# Introduction to Photonics

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Draft

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# Preface

***Photonics*** deals with controlled production, evolution, and detection of (mainly coherent) light (photons) within the 'optical' region of the electromagnetic spectrum (vacuum wavelength ($\lambda_0$) from 50 nm to 500 $\mu$m or photon energies ($\hbar\omega$) from 25 to 0.002 eV). With the advent of the laser non-linear properties of optical media have become essential.

$\vdots$

This draft should be considered as just the germ of a more complete introduction to photonics.

**Recommended Literature:** (Loudon, 2000; Mandel and Wolf, 1995; Yariv, 1997; Vogel et al., 2001; Shen, 1984; Bachor, 1998; Saleh and Teich, 1991)

# Contents

# Chapter 1

# Fundamentals

## 1.1 Classical Electrodynamics

### 1.1.1 Maxwell Equations

The classical electromagnetic field $\mathbf{E}(\mathbf{x},t)\,,\ \mathbf{B}(\mathbf{x},t)$ is defined (implicitly) by the force density

$$\mathbf{f}(\mathbf{x},t) = \rho_{\text{test}}(\mathbf{x},t)\,\mathbf{E}(\mathbf{x},t) + \boldsymbol{\jmath}_{\text{test}}(\mathbf{x},t)\times\mathbf{B}(\mathbf{x},t) \tag{1.1}$$

that it exerts on the carriers of test 4-current densities $\Big(c\,\rho_{\text{test}}(\mathbf{x},t), \boldsymbol{\jmath}_{\text{test}}(\mathbf{x},t)\Big)$ ; see, e.g., Sections 3.3.3 and 4.1.3 of (Lücke, rel). The field generated by a 4-current density $\Big(c\,\rho(\mathbf{x},t), \boldsymbol{\jmath}(\mathbf{x},t)\Big)$ fulfilling the continuity equation

$$\dot\rho(\mathbf{x},t) + \operatorname{div}\boldsymbol{\jmath}(\mathbf{x},t) = 0 \tag{1.2}$$

has to obey Maxwell's equations[1]

$$\begin{aligned}
\operatorname{curl}\mathbf{B}(\mathbf{x},t) &= \mu_0\Big(\epsilon_0\,\frac{\partial}{\partial t}\mathbf{E}(\mathbf{x},t) + \boldsymbol{\jmath}(\mathbf{x},t)\Big)\,, \\
\operatorname{curl}\mathbf{E}(\mathbf{x},t) &= -\frac{\partial}{\partial t}\mathbf{B}(\mathbf{x},t)\,, \\
\operatorname{div}\mathbf{E}(\mathbf{x},t) &= \frac{1}{\epsilon_0}\,\rho(\mathbf{x},t)\,, \\
\operatorname{div}\mathbf{B}(\mathbf{x},t) &= 0
\end{aligned} \tag{1.3}$$

(we use SI units; see Appendix A.3.3 of (Lücke, edyn)). More precisely, in the classical case, it is the physical solution

$$\mathbf{E}(\mathbf{x},t) = -\operatorname{grad}\Phi_{\text{ret}}(\mathbf{x},t) - \frac{\partial}{\partial t}\mathbf{A}_{\text{ret}}(\mathbf{x},t)\;,\quad \mathbf{B}(\mathbf{x},t) = \operatorname{curl}\mathbf{A}_{\text{ret}}(\mathbf{x},t)$$

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[1]Note that $\mu_0\,\epsilon_0 = c^{-2}$ , where

$$\epsilon_0 \approx 8,85\cdot 10^{-12}\,\frac{\text{As}}{\text{Nm}}$$

and $c$ denotes the velocity of light in vacuum ($\approx 3\cdot 10^8$ m/s).

given by the retarded electromagnetic Potentials

$$
\begin{aligned}
\Phi_{\mathrm{ret}}(\mathbf{x},t) &= \frac{1}{\epsilon_0}\int \frac{\rho\left(\mathbf{x}',t-\frac{|\mathbf{x}-\mathbf{x}'|}{c}\right)}{4\pi\,|\mathbf{x}-\mathbf{x}'|}\,dV_{\mathbf{x}'}\,,\\
\mathbf{A}_{\mathrm{ret}}(\mathbf{x},t) &= \mu_0\int \frac{\boldsymbol{\jmath}\left(\mathbf{x}',t-\frac{|\mathbf{x}-\mathbf{x}'|}{c}\right)}{4\pi\,|\mathbf{x}-\mathbf{x}'|}\,dV_{\mathbf{x}'}\,.
\end{aligned}
\tag{1.4}
$$

In typical applications the 4-current density is approximated as

$$
\begin{aligned}
\rho(\mathbf{x},t) &= \rho_{\mathrm{ex}}(\mathbf{x},t) - \operatorname{div}\boldsymbol{\mathcal{P}}(\mathbf{x},t)\,,\\
\boldsymbol{\jmath}(\mathbf{x},t) &= \boldsymbol{\jmath}_{\mathrm{ex}}(\mathbf{x},t) + \frac{\partial}{\partial t}\boldsymbol{\mathcal{P}}(\mathbf{x},t)\,,
\end{aligned}
\tag{1.5}
$$

with some known 4-current density $\left(c\,\rho_{\mathrm{ex}}(\mathbf{x},t),\boldsymbol{\jmath}_{\mathrm{ex}}(\mathbf{x},t)\right)$ of *excess charges* and some ***generalized polarization*** $\boldsymbol{\mathcal{P}}(\mathbf{x},t)$.

**Remarks:**

1. For every choice of $\rho_{\mathrm{ex}}(\mathbf{x},t),\boldsymbol{\jmath}_{\mathrm{ex}}(\mathbf{x},t)$ obeying the continuity equation there is[2] a time-dependent vector field $\boldsymbol{\mathcal{P}}(\mathbf{x},t)$ fulfilling (1.5).
2. Under suitable conditions we have

$$
\frac{\partial}{\partial t}\boldsymbol{\mathcal{P}}(\mathbf{x},t) = \frac{\partial}{\partial t}\boldsymbol{\mathcal{P}}_{\mathrm{el}}(\mathbf{x},t) + \frac{1}{\mu_0}\operatorname{rot}\boldsymbol{\mathcal{M}}(\mathbf{x},t)\,,
\tag{1.6}
$$

(see, e.g., Chapter 4 of (Lücke, edyn)), where $\boldsymbol{\mathcal{P}}_{\mathrm{el}}(\mathbf{x},t) = \mathbf{D}(\mathbf{x},t) - \epsilon_0\,\mathbf{E}(\mathbf{x},t)$ denotes the electric dipole density and $\boldsymbol{\mathcal{M}}(\mathbf{x},t) = \mathbf{B}(\mathbf{x},t) - \mu_0\,\mathbf{H}(\mathbf{x},t)$ the magnetic dipole density.[3]

Then MAXWELL's equation become equivalent to

$$
\operatorname{curl}\mathbf{B}(\mathbf{x},t) = \mu_0\frac{\partial}{\partial t}\left(\epsilon_0\,\mathbf{E}(\mathbf{x},t)+\boldsymbol{\mathcal{P}}(\mathbf{x},t)\right) + \mu_0\,\boldsymbol{\jmath}_{\mathrm{ex}}(\mathbf{x},t)\,, \tag{1.7}
$$

$$
\operatorname{curl}\mathbf{E}(\mathbf{x},t) = -\frac{\partial}{\partial t}\mathbf{B}(\mathbf{x},t)\,, \tag{1.8}
$$

$$
\operatorname{div}\left(\epsilon_0\,\mathbf{E}(\mathbf{x},t)+\boldsymbol{\mathcal{P}}(\mathbf{x},t)\right) = +\rho_{\mathrm{ex}}(\mathbf{x},t)\,, \tag{1.9}
$$

$$
\operatorname{div}\mathbf{B}(\mathbf{x},t) = 0\,. \tag{1.10}
$$



[2]See (Lücke, 1995), in this connection.

[3]Contrary to a wrong argument given in (Landau und Lifschitz, 1967, § 60) the interpretation of $\boldsymbol{\mathcal{M}}(\mathbf{x},t)$ does **not** imply $\frac{\partial}{\partial t}\boldsymbol{\mathcal{P}}_{\mathrm{el}}(\mathbf{x},t)\approx 0$ for $\boldsymbol{\jmath}_{\mathrm{ex}}(\mathbf{x},t) = 0$. For higher dipole moments see (Bloembergen, 1996b, p. 63).

### 1.1.2 Energy and Energy Flux of the Field

For arbitrary (sufficiently well-behaved) regions $\mathcal{G}$ we have

$$\begin{aligned}
&\int_{\mathcal{G}} \mu_0\, \boldsymbol{\jmath}(\mathbf{x},t)\cdot \mathbf{E}(\mathbf{x},t)\,\mathrm{d}V_{\mathbf{x}} \\
&\underset{(1.3)}{=} \int_{\mathcal{G}} \Big( \underbrace{\mathbf{E}(\mathbf{x},t)\cdot\left(\boldsymbol{\nabla}_{\mathbf{x}}\times\mathbf{B}(\mathbf{x},t)\right)}_{=\mathbf{B}(\mathbf{x},t)\cdot\left(\underbrace{\boldsymbol{\nabla}_{\mathbf{x}}\times\mathbf{E}(\mathbf{x},t)}_{\underset{(1.8)}{=}-\frac{\partial}{\partial t}\mathbf{B}(\mathbf{r},t)}\right)-\boldsymbol{\nabla}_{\mathbf{x}}\cdot\left(\mathbf{E}(\mathbf{x},t)\times\mathbf{B}(\mathbf{x},t)\right)} - \epsilon_0\,\mu_0\,\mathbf{E}(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\mathbf{E}(\mathbf{x},t)\Big)\,\mathrm{d}V_{\mathbf{x}} \\
&= -\int_{\mathcal{G}} \left(\epsilon_0\,\mu_0\,\mathbf{E}(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\mathbf{E}(\mathbf{x},t) + \mathbf{B}(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\mathbf{B}(\mathbf{x},t) + \boldsymbol{\nabla}_{\mathbf{x}}\cdot\left(\mathbf{E}(\mathbf{x},t)\times\mathbf{B}(\mathbf{x},t)\right)\right)\mathrm{d}V_{\mathbf{x}}\,.
\end{aligned}$$

This implies

$$\begin{aligned}
&\frac{1}{2}\frac{\partial}{\partial t}\left(\epsilon_0\,|\mathbf{E}(\mathbf{x},t)|^2 + \frac{1}{\mu_0}\,|\mathbf{B}(\mathbf{x},t)|^2\right) \\
&= -\boldsymbol{\jmath}(\mathbf{x},t)\cdot\mathbf{E}(\mathbf{x},t) - \boldsymbol{\nabla}_{\mathbf{x}}\cdot\frac{\mathbf{E}(\mathbf{x},t)\times\mathbf{B}(\mathbf{x},t)}{\mu_0}
\end{aligned} \tag{1.11}$$

and therefore, by GAUSS' theorem, the POYNTING ***theorem*** [4]

$$\begin{aligned}
&\frac{\mathrm{d}}{\mathrm{d}t}\int_{\mathcal{G}}\frac{1}{2}\left(\epsilon_0\,|\mathbf{E}(\mathbf{x},t)|^2 + \frac{1}{\mu_0}\,|\mathbf{B}(\mathbf{x},t)|^2\right)\mathrm{d}V_{\mathbf{x}} \\
&= -\int_{\mathcal{G}}\boldsymbol{\jmath}(\mathbf{x},t)\cdot\mathbf{E}(\mathbf{x},t)\,\mathrm{d}V_{\mathbf{x}} - \int_{\partial G}\frac{\mathbf{E}(\mathbf{x},t)\times\mathbf{B}(\mathbf{x},t)}{\mu_0}\cdot\mathrm{d}\mathbf{S}_{\mathbf{x}}\,.
\end{aligned} \tag{1.12}$$

**Remark:** If (1.6) holds then (1.12) and GAUSS's theorem, because of

$$\begin{aligned}
\operatorname{div}\left(\mathbf{E}(\mathbf{x},t)\times\boldsymbol{\mathcal{M}}(\mathbf{x},t)\right) &= \boldsymbol{\mathcal{M}}(\mathbf{x},t)\cdot\operatorname{curl}\mathbf{E}(\mathbf{x},t) - \mathbf{E}(\mathbf{x},t)\cdot\operatorname{curl}\boldsymbol{\mathcal{M}}(\mathbf{x},t) \\
&\underset{(1.8)}{=} -\boldsymbol{\mathcal{M}}(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\mathbf{B}(\mathbf{x},t) - \mathbf{E}(\mathbf{x},t)\cdot\operatorname{curl}\boldsymbol{\mathcal{M}}(\mathbf{x},t)\,,
\end{aligned}$$

also imply

$$\begin{aligned}
&\int_{\mathcal{G}}\left(\mathbf{E}(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\mathbf{D}(\mathbf{x},t) + \mathbf{H}(\mathbf{x},t)\,\frac{\partial}{\partial t}\mathbf{B}(\mathbf{x},t)\right)\mathrm{d}V_{\mathbf{x}} \\
&= -\int_{\mathcal{G}}\boldsymbol{\jmath}_{\mathrm{ex}}(\mathbf{x},t)\cdot\mathbf{E}(\mathbf{x},t)\,\mathrm{d}V_{\mathbf{x}} - \int_{\partial G}\left(\mathbf{E}(\mathbf{x},t)\times\mathbf{H}(\mathbf{x},t)\right)\cdot\mathrm{d}\mathbf{S}_{\mathbf{x}}\,,
\end{aligned} \tag{1.13}$$

where

$$\mathbf{D}(\mathbf{x},t) \stackrel{\text{def}}{=} \epsilon_0\,\mathbf{E}(\mathbf{x},t) + \boldsymbol{\mathcal{P}}(\mathbf{x},t)\,, \quad \mathbf{H}(\mathbf{x},t) \stackrel{\text{def}}{=} \frac{\mathbf{B}(\mathbf{x},t) - \boldsymbol{\mathcal{M}}(\mathbf{x},t)}{\mu_0}\,.$$



[4] See also (Kinsler et al., 2009).

Since, by (1.1), $\int_{\mathcal{G}} \boldsymbol{\jmath}(\mathbf{x},t)\cdot\mathbf{E}(\mathbf{x},t)\,\mathrm{d}V_{\mathbf{x}}$ is to be interpreted as the rate of the work done by the electromagnetic field on the charges, (1.12) suggests the interpretation

$$\boxed{\boldsymbol{\mathcal{S}}(\mathbf{x},t) \stackrel{\text{def}}{=} \frac{1}{\mu_0}\mathbf{E}(\mathbf{x},t)\times\mathbf{B}(\mathbf{x},t) = \left\{\begin{array}{l}\text{engergy current density of the}\\ \text{plain electromagnetic field}\end{array}\right.} \tag{1.14}$$

of the (free) POYNTING ***vector*** $\boldsymbol{\mathcal{S}}(\mathbf{x},t)$ and

$$\boxed{\begin{aligned}\mathcal{H}_0(\mathbf{x},t) &\stackrel{\text{def}}{=} \frac{1}{2}\left(\epsilon_0\,|\mathbf{E}(\mathbf{x},t)|^2 + \frac{1}{\mu_0}\,|\mathbf{B}(\mathbf{x},t)|^2\right)\\ &= \left\{\begin{array}{l}\text{energy density of the}\\ \text{plain electromagnetic field}\,.\end{array}\right.\end{aligned}} \tag{1.15}$$

Assuming (1.5) and defining[5]

$$\mathcal{U}(\mathbf{x},t) \stackrel{\text{def}}{=} \left\{\begin{array}{l}\text{work done by the field}\\ \text{on the non-excess charges}\,.\end{array}\right.$$

we get

$$\frac{\partial}{\partial t}\mathcal{U}(\mathbf{x},t) \underset{(1.1)}{=} \mathbf{E}(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\boldsymbol{\mathcal{P}}(\mathbf{x},t)\,. \tag{1.16}$$

**Remark:** Recall that the power performed by a force $\mathbf{F}(t)$ on a masspoint with trajectory $\mathbf{x}(t)$ is $\mathbf{F}(t)\cdot\dot{\mathbf{x}}(t)$. Correspondingly, the power performed by the angular momentum $\mathbf{M}(t)$ on a rigid body rotating with circular angular velocity $\boldsymbol{\omega}(t)$ is $\mathbf{M}\cdot\boldsymbol{\omega}(t)$. Likewise, the power performed by an electrical field $\mathbf{E}(t)$ on an electrical dipole $\mathbf{P}(t)$ is $\mathbf{E}(t)\cdot\dot{\mathbf{P}}(t)$ — **not** $\frac{\mathrm{d}}{\mathrm{d}t}\left(\mathbf{E}(t)\cdot\mathbf{P}(t)\right)$.

Therefore, (1.11) and (1.5) imply

$$\frac{\partial}{\partial t}\Big(\mathcal{H}_0(\mathbf{x},t)+\mathcal{U}(\mathbf{x},t)\Big) = -\boldsymbol{\jmath}_{\text{ex}}(\mathbf{x},t)\cdot\mathbf{E}(\mathbf{x},t) - \boldsymbol{\nabla}_{\mathbf{x}}\cdot\frac{\mathbf{E}(\mathbf{x},t)\times\mathbf{B}(\mathbf{x},t)}{\mu_0}\,.$$

Consequently:

$$\boldsymbol{\jmath}_{\text{ex}} = 0 \quad\Longrightarrow\quad \frac{\mathrm{d}}{\mathrm{d}t}\int\Big(\mathcal{H}_0(\mathbf{x},t)+\mathcal{U}(\mathbf{x},t)\Big)\mathrm{d}V_{\mathbf{x}} = 0\,. \tag{1.17}$$

### 1.1.3 Frequency Analysis

Using the FOURIER transform

$$\widetilde{f}(\mathbf{x},\omega) \stackrel{\text{def}}{=} \frac{1}{\sqrt{2\pi}}\int f(\mathbf{x},t)\,e^{i\omega t}\,\mathrm{d}t$$



[5]Concerning the classical field energy see (Landau und Lifschitz, 1967, Chapt. IX, § 61).

resp.

$$\widetilde{\mathbf{f}}(\mathbf{x},\omega) \stackrel{\text{def}}{=} \frac{1}{\sqrt{2\pi}} \int \mathbf{f}(\mathbf{x},t)\, e^{i\omega t}\, \mathrm{d}t$$

we may write (1.7)–(1.10) in the equivalent form

$$\begin{aligned}
\frac{1}{\mu_0}\operatorname{curl}\widetilde{\mathbf{B}}(\mathbf{x},\omega) &= -i\,\omega\left(\epsilon_0\,\widetilde{\mathbf{E}}(\mathbf{x},\omega)+\widetilde{\mathcal{P}}(\mathbf{x},\omega)\right)+\widetilde{\boldsymbol{\jmath}}_{\text{ex}}(\mathbf{x},\omega)\,, && (1.18)\\
\operatorname{curl}\widetilde{\mathbf{E}}(\mathbf{x},\omega) &= +i\,\omega\,\widetilde{\mathbf{B}}(\mathbf{x},\omega)\,, && (1.19)\\
\operatorname{div}\left(\epsilon_0\,\widetilde{\mathbf{E}}(\mathbf{x},\omega)+\widetilde{\mathcal{P}}(\mathbf{x},\omega)\right) &= +\widetilde{\rho}_{\text{ex}}(\mathbf{x},\omega)\,, && (1.20)\\
\operatorname{div}\widetilde{\mathbf{B}}(\mathbf{x},\omega) &= 0\,. && (1.21)
\end{aligned}$$

> **Remark:** If the FOURIER-integrals do not exist in the ordinary sense — as, e.g., for monochromatic waves — they have to be interpreted in the distributional sense; see Exercise 33 of (Lücke, qft) or Section 3.1.1 of (Lücke, ftm).

The continuity equation for $\rho_{\text{ex}}, \boldsymbol{\jmath}_{\text{ex}}$ implies

$$\widetilde{\rho}_{\text{ex}}(\mathbf{x},\omega) = \frac{1}{i\omega}\operatorname{div}\widetilde{\boldsymbol{\jmath}}_{\text{ex}}(\mathbf{x},\omega) \quad \forall\,\omega\neq 0 \tag{1.22}$$

and, consequently, equivalence of the equations (1.18)–(1.21) to

$$\widetilde{\mathbf{B}}(\mathbf{x},\omega) = \frac{1}{i\,\omega}\operatorname{curl}\widetilde{\mathbf{E}}(\mathbf{x},\omega) \quad \forall\,\omega\neq 0$$

and

$$\boxed{\operatorname{curl}\left(\operatorname{curl}\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right) = \left(\frac{\omega}{c}\right)^2\left(\widetilde{\mathbf{E}}(\mathbf{x},\omega)+\frac{1}{\epsilon_0}\,\widetilde{\mathcal{P}}(\mathbf{x},\omega)\right)+i\,\omega\,\mu_0\,\widetilde{\boldsymbol{\jmath}}_{\text{ex}}(\mathbf{x},\omega)} \tag{1.23}$$

for $\omega\neq 0$.

Of course, the physical fields have to be real. For the electric field, e.g., this means

$$\mathbf{E}(\mathbf{x},t) = \left(\mathbf{E}(\mathbf{x},t)\right)^*,$$

i.e.

$$\widetilde{\mathbf{E}}(\mathbf{x},-\omega) = \left(\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right)^*. \tag{1.24}$$

Therefore, it is sufficient to determine $\widetilde{\mathbf{E}}(\mathbf{x},\omega)$ for $\omega\geq 0$.

## 1.2 Quantized Electromagnetic Radiation

Of course, the classical electromagnetic theory should be quantized.[6] A rigorous relativistic quantum theory describing the interaction of radiation with matter (non-perturbative QED) does not yet exist. Fortunately, as we will see, for many important applications the classical macroscopic MAXWELL theory supplies sufficient information concerning the influence of linear optical devices on photons.[7]

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[6] For a detailed discussion why a semiclassical theory (treating only 'particles' quantum mechanically) plus vacuum fluctuations is insufficient see, e.g., (Scully and Zubairy, 1999).

[7] The notion 'photon' was introduced in (Lewis, 1926). See (Lamb, Jr., 1995) for a historical review.

### 1.2.1 Quantum Aspects of Light

Nowadays (almost) monochromatic electromagnetic radiation of angular frequency $\omega$ is considered to be composed of quanta (photons) having (total) energy $\hbar\omega$ each (and zero *rest mass*). This leads to additional information on optical processes.

To give an example, consider second harmonic generation which has to be considered as formation of quanta having energy $2\hbar\omega$ out of pairs of quanta having energy $\hbar\omega$ each.[8] Reversibility of quantum dynamics predicts also the reverse process, called *spontaneous down conversion*: instantaneous formation of pairs of photons out of single photons. Contrary to the quantized theory[9] classical optics does not give any hint at (essentially) simultaneous creation of both partners of each pair, as confirmed by experiment.[10]

More generally, quantum optics describes multi-photon processes mediated by matter. These processes are called ***nonlinear*** if what happens to a photon depends in an essential way on the presence and properties of other optical photons.[11] This kind of nonlinearity is essential for optical implementations of modern quantum information processing.

### 1.2.2 Field Operators

In the so-called ***radiation gauge*** a classical electromagnetic vacuum field $\mathbf{E}(\mathbf{x},t)$, $\mathbf{B}(\mathbf{x},t)$ (of sufficiently rapid decrease at spatial infinity) may be represented in the form

$$\boxed{\mathbf{B}(\mathbf{x},t) = \operatorname{curl}\mathbf{A}(\mathbf{x},t)\,, \quad \mathbf{E}(\mathbf{x},t) = -\frac{\partial}{\partial t}\mathbf{A}(\mathbf{x},t)} \tag{1.25}$$

with the vector potential

$$\mathbf{A}(\mathbf{x},t) = \frac{1}{4\pi}\int \frac{\operatorname{curl}\mathbf{B}(\mathbf{x}',t)}{|\mathbf{x}-\mathbf{x}'|}\,\mathrm{d}V_{\mathbf{x}'} \tag{1.26}$$

fulfilling the COULOMB ***condition***

$$\operatorname{div}\mathbf{A}(\mathbf{x},t) = 0\,, \tag{1.27}$$

**Remark:** Equations (1.25) follow from (1.26) by $\operatorname{curl}\operatorname{curl} = \operatorname{grad}\operatorname{div} - \Delta$ and the POISSON equation $\Delta_{\mathbf{x}}\frac{1}{|\mathbf{x}-\mathbf{x}'|} = -4\pi\,\delta(\mathbf{x}-\mathbf{x}')$; see, e.g., equations 4.87 and 4.63 of (Lücke, ein). The COULOMB condition follows for (1.26) by $\operatorname{div}\operatorname{curl} = 0$; see, e.g., Equation 4.89 of (Lücke, ein).

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[8] Momentum conservation holds for the momenta **inside** the medium, corresponding to *phase (-velocity) matching* from the classical point of view.

[9] See (Mandel and Wolf, 1995, Sect. 22.4.2).

[10] See (Mandel and Wolf, 1995, Sect. 22.4.7).

[11] This type of *nonlinearity* does not contradict the fundamental linearity of quantum mechanical time evolution!

In this gauge the free MAXWELL equations are equivalent to the vectorial wave equation

$$\square\, \mathbf{A}(\mathbf{x},t) = 0\,, \qquad \square \stackrel{\text{def}}{=} \Delta - \left(\frac{1}{c}\frac{\partial}{\partial t}\right)^2 . \tag{1.28}$$

If no boundary conditions are imposed, every (sufficiently well-behaved, real) solution of (1.28) fulfilling the COULOMB condition is of the form

$$\mathbf{A}(\mathbf{x},t) \stackrel{\text{def}}{=} \mathbf{A}^{(+)}(\mathbf{x},t) + \left(\mathbf{A}^{(+)}(\mathbf{x},t)\right)^* \tag{1.29}$$

with a ***complex vector potential***[12]

$$\mathbf{A}^{(+)}(\mathbf{x},t) = (2\pi)^{-3/2}\sqrt{\mu_0 \hbar c}\int \left(\sum_{j=1}^{2} \boldsymbol{\epsilon}_j(\mathbf{k})\, a_j(\mathbf{k})\right) e^{-i(c|\mathbf{k}|t - \mathbf{k}\cdot\mathbf{x})}\, \frac{dV_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\,, \tag{1.30}$$

where the $a_j(\mathbf{k})$ are (sufficiently well-behaved) complex-valued functions and the $\boldsymbol{\epsilon}_j(\mathbf{k})$ are vector-valued functions forming a

$$\text{right handed orthonormal basis } \left\{\boldsymbol{\epsilon}_1(\mathbf{k}), \boldsymbol{\epsilon}_2(\mathbf{k}), \frac{\mathbf{k}}{|\mathbf{k}|}\right\} \text{ of } \mathbb{R}^3$$

for every $\mathbf{k} \neq 0$.

In the HEISENBERG picture the observables $\hat{\mathbf{E}}(\mathbf{x},t)$ and $\hat{\mathbf{B}}(\mathbf{x},t)$ should obey the same linear relations as their classical counter parts $\mathbf{E}(\mathbf{x},t)$ and $\mathbf{B}(\mathbf{x},t)$. This can be achieved replacing the complex-valued functions $a_j(\mathbf{k})$ by suitable operator-valued functions $\hat{a}_j(\mathbf{k})$ :

$$\boxed{\hat{\mathbf{A}}^{(+)}(\mathbf{x},t) \stackrel{\text{def}}{=} (2\pi)^{-3/2}\sqrt{\mu_0 \hbar c}\int \left(\sum_{j=1}^{2} \boldsymbol{\epsilon}_j(\mathbf{k})\, \hat{a}_j(\mathbf{k})\right) e^{-i(c|\mathbf{k}|t - \mathbf{k}\cdot\mathbf{x})}\, \frac{dV_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\,,} \tag{1.31}$$

$$\begin{aligned}
\hat{\mathbf{B}}^{(+)}(\mathbf{x},t) &\stackrel{\text{def}}{=} \operatorname{curl} \hat{\mathbf{A}}^{(+)}(\mathbf{x},t) \qquad (1.32)\\
&= (2\pi)^{-3/2}\sqrt{\mu_0 \hbar c}\sum_{j=1}^{2}\int i\mathbf{k}\times\boldsymbol{\epsilon}_j(\mathbf{k})\,\hat{a}_j(\mathbf{k})\, e^{-i(c|\mathbf{k}|t - \mathbf{k}\cdot\mathbf{x})}\,\frac{dV_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\,,\\
\hat{\mathbf{E}}^{(+)}(\mathbf{x},t) &\stackrel{\text{def}}{=} -\frac{\partial}{\partial t}\hat{\mathbf{A}}^{(+)}(\mathbf{x},t) \qquad (1.33)\\
&= (2\pi)^{-3/2}\sqrt{\mu_0 \hbar c}\sum_{j=1}^{2}\int ic\,|\mathbf{k}|\;\boldsymbol{\epsilon}_j(\mathbf{k})\,\hat{a}_j(\mathbf{k})\, e^{-i(c|\mathbf{k}|t - \mathbf{k}\cdot\mathbf{x})}\,\frac{dV_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\,,
\end{aligned}$$

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[12]The factor $1/\sqrt{2\,|\mathbf{k}|}$ under the integral will be necessary for LORENTZ covariance of the quantized field tensor. The factor $\sqrt{\mu_0\hbar c}$ is chosen in view of (1.57).

$$\hat{\mathbf{A}}(\mathbf{x},t) \stackrel{\text{def}}{=} \hat{\mathbf{A}}^{(+)}(\mathbf{x},t) + \left(\hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\right)^\dagger , \tag{1.34}$$

$$\hat{\mathbf{B}}(\mathbf{x},t) \stackrel{\text{def}}{=} \hat{\mathbf{B}}^{(+)}(\mathbf{x},t) + \left(\hat{\mathbf{B}}^{(+)}(\mathbf{x},t)\right)^\dagger , \tag{1.35}$$

$$\hat{\mathbf{E}}(\mathbf{x},t) \stackrel{\text{def}}{=} \hat{\mathbf{E}}^{(+)}(\mathbf{x},t) + \left(\hat{\mathbf{E}}^{(+)}(\mathbf{x},t)\right)^\dagger . \tag{1.36}$$

If the operators $\hat{a}_j(\mathbf{k})$ , $(\hat{a}_j(\mathbf{k}))^\dagger$ fulfill the ***canonical commutation relations***[13]

$$\boxed{[\hat{a}_j(\mathbf{k}), \hat{a}_{j'}(\mathbf{k}')]_- = 0\,, \quad \left[\hat{a}_j(\mathbf{k}), \left(\hat{a}_{j'}(\mathbf{k}')\right)^\dagger\right]_- = \delta_{jj'}\,\delta(\mathbf{k}-\mathbf{k}')} \tag{1.37}$$

on a suitable dense subspace $D_0$ , of the Hilbert space $\mathcal{H}_{\text{field}}$ , containing a cyclic normalized ***vacuum*** state vector $\Omega$ characterized by[14]

$$\hat{a}_j(\mathbf{k})\,\Omega = 0\,, \tag{1.38}$$

then the observables $\hat{\mathbf{B}}^{(+)}(\mathbf{x},t)$ , $\hat{\mathbf{E}}^{(+)}(\mathbf{x},t)$ fulfill all reasonable physical postulates.

**Remarks:**

1. These properties fix the quantized theory up to unitary equivalence (see Sect. 4.1.1 of (Lücke, qft) and (Fredenhagen, 2001, Sect. III.4)). This also implies, by the way, that all irreducible realizations of (1.37)/(1.38) are unitarily equivalent.
2. Actually, the $(\hat{a}_j(\mathbf{k}))^\dagger$ have to be considered as operator-valued distributions (generalized functions):

$$\int \hat{a}_j^\dagger(\mathbf{k})\,\varphi^*(\mathbf{k})\,d\mathbf{k} \subset \left(\int \hat{a}_j(\mathbf{k})\,\varphi(\mathbf{k})\,d\mathbf{k}\right)^\dagger \quad \forall\, \varphi \in \mathcal{S}(\mathbb{R}^3)\,.$$

   See, e.g., Section 2.1.3 of (Lücke, qft) for details.

Especially, the field observables fulfill the quantized free Maxwell equations:

$$\operatorname{curl}\hat{\mathbf{B}}(\mathbf{x},t) = \mu_0\,\epsilon_0\,\frac{\partial}{\partial t}\,\hat{\mathbf{E}}(\mathbf{x},t)\,, \tag{1.39}$$

$$\operatorname{curl}\hat{\mathbf{E}}(\mathbf{x},t) = -\frac{\partial}{\partial t}\,\hat{\mathbf{B}}(\mathbf{x},t)\,, \tag{1.40}$$

$$\operatorname{div}\hat{\mathbf{E}}(\mathbf{x},t) = 0\,, \tag{1.41}$$

$$\operatorname{div}\hat{\mathbf{B}}(\mathbf{x},t) = 0\,. \tag{1.42}$$

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[13] We simply write $z$ for $z\,\hat{1}$ , $z \in \mathbb{C}$ , as long as this does not cause any confusion. For a possible realization of (1.37)/(1.38) see A.3.

[14] Because of (1.38) the $\hat{a}_j(\mathbf{k})$ are called ***annihilation operators*** (see (Mizrahi and Dodonov, 2002) in this connection). Here, 'cyclic' means that the linear span of all vectors of the form

$$\underbrace{\int f(\mathbf{k}_1,\ldots,\mathbf{k}_n)\,(\hat{a}_{j_1}(\mathbf{k}_1))^\dagger \cdots (\hat{a}_{j_n}(\mathbf{k}_1))^\dagger\,dV_{\mathbf{k}_1}\ldots dV_{\mathbf{k}_n}}_{\stackrel{\text{def}}{=} 1 \text{ for } n=0}\,\Omega\,, \quad n \in \mathbb{Z}_+\,,\, j_1,\ldots,j_n \in \{1,2\}\,,$$

with square integrable $f$'s is dense in $\mathcal{H}_{\text{field}}$ .

However, the canonical commutation relations also imply

$$\begin{aligned}&\left[(\hat{A}^{j_1})^{(+)}(\mathbf{x}_1,t_1)\,,\,\left((\hat{A}^{j_2})^{(+)}(\mathbf{x}_2,t_2)\right)^\dagger\right]_-\\&= i\,\mu_0\,\hbar\,c\left(\delta_{j_1 j_2}-\frac{\frac{\partial}{\partial x_1^{j_1}}\frac{\partial}{\partial x_1^{j_2}}}{\Delta_{\mathbf{x}_1}}\right)\Delta_0^{(+)}(\mathbf{x}_1-\mathbf{x}_2,t_1-t_2)\end{aligned}\tag{1.43}$$

(in the distributional sense), where

$$\begin{aligned}\Delta_0^{(+)}(\mathbf{x},t) &\stackrel{\text{def}}{=} -i\,(2\pi)^{-3}\int e^{i\,\mathbf{k}\cdot\mathbf{x}}\,e^{-i\,c|\mathbf{k}|t}\,\frac{\mathrm{d}V_{\mathbf{k}}}{2\,|\mathbf{k}|}\\&= -i\,(2\pi)^{-3}\int_{k^0>0}\delta\left(k^0\,k^0-\mathbf{k}\cdot\mathbf{k}\right)e^{-i\left(k^0ct-\mathbf{k}\cdot\mathbf{x}\right)}\,\mathrm{d}V_{\mathbf{k}}\,\mathrm{d}k^0\,.\end{aligned}\tag{1.44}$$

**Outline of proof for (1.43):** Since

$$\begin{aligned}&\left[\sum_{j=1}^{2}\boldsymbol{\epsilon}_j^{j_1}(\mathbf{k})\,\hat{a}_j(\mathbf{k})\,,\,\sum_{j'=1}^{2}\boldsymbol{\epsilon}_{j'}^{j_2}(\mathbf{k}')\left(\hat{a}_{j'}(\mathbf{k}')\right)^\dagger\right]_-\\&= \sum_{j,j'=1}^{2}\epsilon_j^{j_1}(\mathbf{k})\,\epsilon_{j'}^{j_2}(\mathbf{k}')\left[\hat{a}_j(\mathbf{k})\,,\,\left(\hat{a}_{j'}(\mathbf{k}')\right)^\dagger\right]_-\\&= \sum_{j=1}^{2}\epsilon_j^{j_1}(\mathbf{k})\,\epsilon_j^{j_2}(\mathbf{k}')\,\delta\left(\mathbf{k}-\mathbf{k}'\right)\,,\end{aligned}$$

(1.31) implies

$$\begin{aligned}&\left[(\hat{A}^{j_1})^{(+)}(\mathbf{x}_1,t_1)\,,\,\left((\hat{A}^{j_2})^{(+)}(\mathbf{x}_2,t_2)\right)^\dagger\right]_-\\&= (2\pi)^{-3}\,\mu_o\,\hbar\,c\int\underbrace{\sum_{j=1}^{2}\epsilon_j^{j_1}(\mathbf{k})\,\epsilon_j^{j_2}(\mathbf{k}')}_{=\delta_{j_1j_2}-\frac{k^{j_1}k^{j_2}}{|\mathbf{k}|^2}}e^{-i(c|\mathbf{k}|(t_1-t_2)-\mathbf{k}\cdot(\mathbf{x}_1-\mathbf{x}_2))}\,\frac{\mathrm{d}V_{\mathbf{k}}}{2\,|\mathbf{k}|}\,.\qquad\blacksquare\end{aligned}$$

(1.43) implies

$$\boxed{\begin{aligned}&\left[\hat{A}^{j_1}(\mathbf{x}_1,t_1)\,,\,\hat{A}^{j_2}(\mathbf{x}_2,t_2)\right]_-\\&= i\,\mu_0\,\hbar\,c\left(\delta_{j_1 j_2}-\frac{\frac{\partial}{\partial x_1^{j_1}}\frac{\partial}{\partial x_1^{j_2}}}{\Delta_{\mathbf{x}_1}}\right)\Delta_0(\mathbf{x}_1-\mathbf{x}_2,t_1-t_2)\,,\end{aligned}}\tag{1.45}$$

where[15]

$$\Delta_0(\mathbf{x},t)\stackrel{\text{def}}{=}\Delta_0^{(+)}(\mathbf{x},t)-\Delta_0^{(+)}(-\mathbf{x},-t)\,.\tag{1.46}$$

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[15] Note that $\Delta_0(\mathbf{x},t)$, being an anti-symmetric Lorentz invariant distribution, vanishes for $c\,|t|\geq|\mathbf{x}|$. However, the operator on the r.h.s of (1.45) is non-local.

Therefore, we have

$$\left\langle \Omega \,\middle|\, \hat{\mathbf{E}}(\mathbf{x},t)\cdot\hat{\mathbf{E}}(\mathbf{x}',t')\,\Omega \right\rangle \neq 0 \tag{1.47}$$

in spite of[16]

$$\left\langle \Omega \,\middle|\, \hat{\mathbf{E}}(\mathbf{x},t)\,\Omega \right\rangle = 0\,.$$

Since $\hat{\mathbf{E}}(\mathbf{x},t)$ is the observable of the electric field,[17] field, (1.47) shows the presence of ***vacuum fluctuations***[18] (***quantum noise***). (1.45) also implies[19]

$$\begin{aligned}
\left[\hat{A}^{j_1}(\mathbf{x}_1,t)\,,\,\hat{A}^{j_2}(\mathbf{x}_2,t)\right]_- &= 0\,,\\
\left[\hat{A}^{j_1}(\mathbf{x}_1,t)\,,\,\hat{B}^{j_2}(\mathbf{x}_2,t)\right]_- &= 0\\
\left[\hat{B}^{j_1}(\mathbf{x}_1,t)\,,\,\hat{B}^{j_2}(\mathbf{x}_2,t)\right]_- &= 0\,,\\
\left[\hat{E}^{j_1}(\mathbf{x}_1,t)\,,\,\hat{E}^{j_2}(\mathbf{x}_2,t)\right]_- &= 0
\end{aligned} \tag{1.48}$$

and

$$\begin{aligned}
\left[\hat{A}^{j_1}(\mathbf{x}_1,t)\,,\,\hat{E}^{j_2}(\mathbf{x}_2,t)\right]_- &= -i\,\frac{\hbar}{\epsilon_0}\,\delta_\perp^{j_1 j_2}(\mathbf{x}_1-\mathbf{x}_2)\,, \\
\left[\hat{\mathbf{B}}(\mathbf{x}_1,t)\,,\,\hat{E}^{j_2}(\mathbf{x}_2,t)\right]_- &= i\,\frac{\hbar}{\epsilon_0}\,\mathbf{e}_{j_2}\times\boldsymbol{\nabla}_{\mathbf{x}_1}\delta(\mathbf{x}_1-\mathbf{x}_2)\,,
\end{aligned} \tag{1.49}$$

where

$$\delta_\perp^{j_1 j_2}(\mathbf{x}) \stackrel{\text{def}}{=} (2\pi)^{-3}\int\left(\delta_{j_1 j_2}-\frac{k^{j_1}k^{j_2}}{|\mathbf{k}|^2}\right)e^{i\,\mathbf{k}\cdot\mathbf{x}}\,\mathrm{d}V_{\mathbf{k}} \tag{1.50}$$

fulfills

$$\begin{aligned}
\sum_{j,j'=1}^{3}\int\delta_\perp^{jj'}(\mathbf{x}-\mathbf{x}')\,F^j(\mathbf{x}')\,\mathrm{d}V_{\mathbf{x}'}\,\mathbf{e}_{j'} &= \mathbf{F}_\perp(\mathbf{x})\\
&\stackrel{\text{def}}{=} \mathbf{F}(\mathbf{x})+\operatorname{grad}\int\frac{\operatorname{div}\mathbf{F}(\mathbf{x}')}{4\pi\,|\mathbf{x}-\mathbf{x}'|}\,\mathrm{d}V_{\mathbf{x}'}
\end{aligned} \tag{1.51}$$

for sufficiently well-behaved $\mathbf{F}(\mathbf{x})$.

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[16]Also for JOHNSON *noise*, the electric current in a simple circuit resulting from fluctuating electromagnetic forces due to thermal motion of the charges, one has zero mean but nonzero mean square. $\Omega$ being an eigenvector of the (non-hermitean!) operator $\hat{\mathbf{E}}^{(+)}(\mathbf{x},t)$ does **not** mean that the positive-frequency part of the electric field has a definite value in the vacuum state.

[17]I.e. $\left\langle \Phi \,\middle|\, \hat{\mathbf{E}}(\mathbf{x},t)\,\Phi \right\rangle$ is the expectation value for the electric field in the quantum state $\Phi$, for all $\Phi \in D_0$.

[18]These provide, e.g., a simple explanation for the **spontaneous** transitions of exited atoms into lower energy states; see (Milonni, 1994, Chapter 3) for some further effects. Also spontaneous down-conversion can be explained as a result of vacuum fluctuations.

[19]The first three equations hold because of $\Delta_0(\mathbf{x},0) = 0$. The last one follows from $\left(\frac{\partial}{\partial t}\right)^2\Delta_0(\mathbf{x},t)_{|t=0} = 0$.

**Remark:** Note that $\mathbf{F}_\perp(\mathbf{x})$ is just the transversal part of

$$\mathbf{F}(\mathbf{x}) = \operatorname{curl} \int \frac{\operatorname{curl} \mathbf{F}(\mathbf{x}')}{4\pi\,|\mathbf{x}-\mathbf{x}'|}\, dV_{\mathbf{x}} - \operatorname{grad} \int \frac{\operatorname{div} \mathbf{F}(\mathbf{x}')}{4\pi\,|\mathbf{x}-\mathbf{x}'|}\, dV_{\mathbf{x}}$$

(see, e.g., Section 4.5.3 of (Lücke, ein)).

Let us finally note that one may prove relativistic covariance of the theory in the sense that there is a unitary representation[20] $\hat{U}(a,\Lambda)$ of the restricted POINCARÉ group $\mathcal{P}_+^\uparrow$ fulfilling

$$\hat{U}(a,\Lambda)\Omega = 0$$

and

$$\hat{U}^{-1}(a,\Lambda)\hat{F}^{\mu\nu}(\Lambda x + a)\,\hat{U}(a,\Lambda) = \sum_{\alpha,\beta=0}^{3} \Lambda^\mu{}_\alpha \Lambda^\nu{}_\beta \hat{F}^{\alpha\beta}(x)$$

(see, e.g., Chapter 4 of (Lücke, qft) for details), where[21]

$$\left(\hat{F}^{\mu\nu}(x)\right) \stackrel{\text{def}}{=} \begin{pmatrix} 0 & -\frac{1}{c}\hat{E}^1(\mathbf{x},t) & -\frac{1}{c}\hat{E}^2(\mathbf{x},t) & -\frac{1}{c}\hat{E}^3(\mathbf{x},t) \\ +\frac{1}{c}\hat{E}^1(\mathbf{x},t) & 0 & -\hat{B}^3(\mathbf{x},t) & +\hat{B}^2(\mathbf{x},t) \\ +\frac{1}{c}\hat{E}^2(\mathbf{x},t) & +\hat{B}^3(\mathbf{x},t) & 0 & -\hat{B}^1(\mathbf{x},t) \\ +\frac{1}{c}\hat{E}^3(\mathbf{x},t) & -\hat{B}^2(\mathbf{x},t) & +\hat{B}^1(\mathbf{x},t) & 0 \end{pmatrix}$$

and

$$x \stackrel{\text{def}}{=} (x^0,\dots,x^3)\,, \quad x^0 \stackrel{\text{def}}{=} c\,t\,.$$

This together with the above commutation relations also shows that the field measurements at space-like separated space-time points are compatible:

$$\left[\hat{F}^{\mu\nu}(x), F^{\mu'\nu'}(x')\right]_- = 0 \quad \text{for } c\,|t-t'| > |\mathbf{x}-\mathbf{x}'| \qquad (\text{EINSTEIN } \boldsymbol{causality})\,.$$

### 1.2.3 Field Modes

The free Hamiltonian $\hat{H}_{\text{field}}$, characterized as the self-adjoint operator fulfilling the commutation relation

$$\frac{i}{\hbar}\left[\hat{H}_{\text{field}}, \hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\right]_- = \frac{\partial}{\partial t}\hat{\mathbf{A}}^{(+)}(\mathbf{x},t) \tag{1.52}$$

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[20] This means that the $\hat{U}(a,\Lambda)$ are unitary operators fulfilling

$$\hat{U}(a_1,\Lambda_1)\hat{U}(a_2,\Lambda_2) = \hat{U}(a_1+\Lambda_1 a_2, \Lambda_1\Lambda_2) \quad \forall\,(a_1,\Lambda_1),(a_2\Lambda_2) \in \mathcal{P}_+^\uparrow\,.$$

[21] Note that $\hat{F}^{\mu\nu}$ is the quantized version of $\frac{1}{\mu_0}H^{\mu\nu}$, as introduced in 1.1.1.

on $D_0 \subset D_{\hat{H}_{\text{field}}}$ and

$$\hat{H}_{\text{field}}\,\Omega = 0\,,$$

is given by[22]

$$\hat{H}_{\text{field}} = \int \hbar\, c\, |\mathbf{k}|\ \hat{n}(\mathbf{k})\, \mathrm{d}V_{\mathbf{k}}\ , \quad \hat{n}(\mathbf{k}) \stackrel{\text{def}}{=} \sum_{j=1}^{2} \left(\hat{a}_j(\mathbf{k})\right)^{\dagger} \hat{a}_j(\mathbf{k})\,, \tag{1.53}$$

where $\hat{n}(\mathbf{k})$ is interpreted as the observable (in the Heisenberg picture) for the $\mathbf{k}$-space density of the ***number of photons***:

$$\int_{\mathcal{O}} \langle \Phi \mid \hat{n}(\mathbf{k})\, \Phi\rangle\, \mathrm{d}V_{\mathbf{k}} = \begin{cases} \text{expectation value for the} \\ \text{number of photons in the state } \Phi \\ \text{with momentum } \mathbf{p} = \hbar\,\mathbf{k} \in \hbar\,\mathcal{O}\,. \end{cases} \tag{1.54}$$

This is consistent in the sense that, for every open subset $\mathcal{O}$ of the $\mathbf{k}$-space, the $t$-independent observable $\int_{\mathcal{O}} \hat{n}(\mathbf{k})\, \mathrm{d}V_{\mathbf{k}}$ commutes[23] with $\hat{H}_{\text{field}}$ , has only nonnegative integer eigenvalues,[24] and

$$\hat{\mathbf{P}}_0 \stackrel{\text{def}}{=} \int \hbar\,\mathbf{k}\,\hat{n}(\mathbf{k})\, \mathrm{d}V_{\mathbf{k}} \tag{1.55}$$

is the momentum observable[25] as uniquely characterized (on $D_0$) by

$$\frac{i}{\hbar}\left[\hat{\mathbf{P}}_0, \hat{A}^j(\mathbf{x}, t)\right]_- = -\boldsymbol{\nabla}_{\mathbf{x}} \hat{A}^j(\mathbf{x}, t) \tag{1.56}$$

and

$$\hat{\mathbf{P}}_0 \Omega = 0$$

According to (1.54),

$$\hat{N} \stackrel{\text{def}}{=} \int \hat{n}(\mathbf{k})\, \mathrm{d}V_{\mathbf{k}}$$

has to be interpreted as the observable for the total number of photons and the subspace $\mathcal{H}_{\text{field}}^{(n)}$ of $n$-photon state vectors coincides with the closed linear span of the



[22]Note that (1.37) implies $\left[\hat{n}(\mathbf{k}'), \hat{a}_j(\mathbf{k})\right]_- = -\hat{a}_j(\mathbf{k})\,\delta(\mathbf{k}-\mathbf{k}')$. This, actually, is the heuristic motivation for (1.37). See (Green, 1953), in this connection.

[23]Note that (1.37) implies $\left[\left(\hat{a}_j(\mathbf{k})\right)^{\dagger}\hat{a}_j(\mathbf{k})\,,\ \left(\hat{a}_{j'}(\mathbf{k}')\right)^{\dagger}\hat{a}_{j'}(\mathbf{k}')\right]_- = 0\,.$

[24]Since (1.37) implies

$$\begin{aligned} &\left(\left(\hat{a}_j(\mathbf{k})\right)^{\dagger}\hat{a}_j(\mathbf{k})\right)\left(\hat{a}_{j_1}(\mathbf{k}_1)\right)^{\dagger}\cdots\left(\hat{a}_{j_n}(\mathbf{k}_n)\right)^{\dagger} \\ &= \left(\hat{a}_{j_1}(\mathbf{k}_1)\right)^{\dagger}\cdots\left(\hat{a}_{j_n}(\mathbf{k}_n)\right)^{\dagger}\left(\left(\left(\hat{a}_j(\mathbf{k})\right)^{\dagger}\hat{a}_j(\mathbf{k})\right) + \sum_{\nu=1}^{n}\delta_{jj_\nu}\delta(\mathbf{k}-\mathbf{k}_\nu)\right), \end{aligned}$$

(recall the derivation of (A.26)).

[25]This observable is $t$-independent in the Heisenberg picture, since it commutes with $\hat{H}_{\text{field}}$ .

set of all vectors of the form[26]

$$\int f(\mathbf{k}_1, \ldots, \mathbf{k}_n) \left(\hat{a}_{j_1}(\mathbf{k}_1)\right)^\dagger \cdots \left(\hat{a}_{j_n}(\mathbf{k}_1)\right)^\dagger \mathrm{d}V_{\mathbf{k}_1} \ldots \mathrm{d}V_{\mathbf{k}_n}\, \Omega$$

with square integrable $f$'s.

As already indicated in Footnote 12, the normalization factor on the r.h.s. of (1.31) has been chosen to yield[27]

$$\boxed{\begin{aligned} \hat{H}_{\text{field}} &= \frac{1}{2}\int \left(\epsilon_0 :\hat{\mathbf{E}}(\mathbf{x},t)\cdot\hat{\mathbf{E}}(\mathbf{x},t): + \frac{1}{\mu_0} :\hat{\mathbf{B}}(\mathbf{x},t)\cdot\hat{\mathbf{B}}(\mathbf{x},t):\right) \mathrm{d}V_{\mathbf{x}}\,, \\ c^2\,\hat{\mathbf{P}}_0 &= \frac{1}{\mu_0}\int :\hat{\mathbf{E}}(\mathbf{x},t)\times\hat{\mathbf{B}}(\mathbf{x},t): \mathrm{d}V_{\mathbf{x}}\,, \end{aligned}} \tag{1.57}$$

where : : means ***normal ordering***, i.e. in all products the ***creation operators*** $\hat{a}_j^\dagger(\mathbf{k}) \stackrel{\text{def}}{=} \left(\hat{a}_j(\mathbf{k})\right)^\dagger$ have to be placed — irrespective of the initial ordering — to the left of the annihilation operators $\hat{a}_j(\mathbf{k})$.

A dense subset of $\mathcal{H}^{(n)}_{\text{field}}$ is given by the linear span of the set of all (i.g. not yet normalized) vectors of the form

$$\hat{a}_1^\dagger \cdots \hat{a}_n^\dagger\, \Omega$$

with ***modes*** $\hat{a}_1, \ldots \hat{a}_n$, i.e. operators of the form[28]

$$\hat{a}_\nu = \sum_{j=1}^{2} \int \hat{a}_j(\mathbf{k}) \left(f_\nu^j(\mathbf{k})\right)^* \mathrm{d}V_{\mathbf{k}}\,, \tag{1.58}$$

$$\left[\hat{a}_\nu, \hat{a}_\nu^\dagger\right]_- = 1\,. \tag{1.59}$$

Here

$$\left\langle \Omega \mid \mathbf{A}^{(+)}(\mathbf{x},t)\,\hat{a}_\nu^\dagger\,\Omega\right\rangle = (2\pi)^{-\frac{3}{2}} \sqrt{\mu_0\,\hbar\,c} \int \sum_{j=1}^{2} \boldsymbol{\epsilon}_j(\mathbf{k})\, f_\nu^j(\mathbf{k})\, e^{-i(c|\mathbf{k}|t - \mathbf{k}\cdot\mathbf{x})} \frac{\mathrm{d}V_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}} \tag{1.60}$$

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[26] Recall Footnote 14.

[27] Recall Footnote 1. Using the commutation relations for the components of $\hat{\mathbf{A}}(\mathbf{x},t)$, $\hat{\mathbf{E}}(\mathbf{x},t)$ and $\hat{\mathbf{B}}(\mathbf{x},t)$ one can verify (1.52) and (1.56) also directly for (1.57). For the connection between the energy current density and the momentum density of the electromagnetic field see (Landau und Lifschitz, 1967, Footnote 1 on Page 284).

[28] Equation (1.59) is equivalent to

$$\sum_{j=1}^{2} \int \left|f_\nu^j(\mathbf{k})\right|^2 \mathrm{d}V_{\mathbf{k}} = 1\,.$$

can be interpreted as *wave function* of the single photon[29] in the (pure) state $\widehat{=}\, \hat{a}_\nu^\dagger\,\Omega$. Since

$$\begin{aligned}\left\langle \Omega \,\middle|\, \hat{a}_j(\mathbf{k}) \sum_{j=1}^{2} \hat{a}_j^\dagger(\mathbf{k})\, f^j(\mathbf{k})\, \mathrm{d}V_{\mathbf{k}}\,\Omega \right\rangle &= \left\langle \Omega \,\middle|\, \sum_{j=1}^{2} [\hat{a}_{j'}(\mathbf{k}')\,,\,\hat{a}_j^\dagger(\mathbf{k})]_-\, f^j(\mathbf{k})\,\mathrm{d}V_{\mathbf{k}}\,\Omega \right\rangle \\ &= f^{j'}(\mathbf{k}')\,,\end{aligned}$$

for every $\phi^{(1)} \in \mathcal{H}^{(1)}_{\text{field}}$ there is exactly one mode $\hat{a}$ with

$$\phi^{(1)} = \hat{a}^\dagger\,\Omega\,.$$

Motivated by

$$\begin{aligned}\left\langle \hat{a}^\dagger\,\Omega \,\middle|\, \hat{c}^\dagger\,\Omega \right\rangle &= \left\langle \Omega \,\middle|\, \hat{a}\,\hat{c}^\dagger\,\Omega \right\rangle \\ &= \left\langle \Omega \,\middle|\, [\hat{a}\,,\hat{c}^\dagger]_-\,\Omega \right\rangle \\ &= [\hat{a}\,,\hat{c}^\dagger]_-\,,\end{aligned}$$

modes $\hat{a}, \hat{c}$ are said to be ***orthogonal*** if

$$[\hat{a}, \hat{c}^\dagger]_- = 0\,.$$

A family $\{\hat{a}_\nu\}_{\nu\in\mathbb{N}}$ of pairwise orthogonal modes is said to be ***maximal***, if $\left\{\hat{a}_\nu^\dagger\,\Omega : \ \nu \in \mathbb{N}\right\}$ is a maximal orthonormal system (MONS) of $\mathcal{H}^{(1)}_{\text{field}}$. For every maximal family $\{\hat{a}_\nu\}_{\nu\in\mathbb{N}}$ of pairwise orthogonal modes we have

$$\hat{a}_j(\mathbf{k}) = \sum_{\nu=1}^{\infty} \left\langle \Omega \,\middle|\, \hat{a}_j(\mathbf{k})\,\hat{a}_\nu^\dagger\,\Omega \right\rangle \hat{a}_\nu\,;$$

since

$$\begin{aligned}\hat{a}_j^\dagger(\mathbf{k})\,\Omega &= \sum_{\nu=1}^{\infty} \left|\hat{a}_\nu^\dagger\,\Omega\right\rangle\!\left\langle \hat{a}_\nu^\dagger\,\Omega\right|\, \hat{a}_j^\dagger(\mathbf{k})\,\Omega \\ &= \sum_{\nu=1}^{\infty} \left(\langle \Omega \mid \hat{a}_j(\mathbf{k})\,\hat{a}_\nu^\dagger\,\Omega\rangle\right)^*\,\hat{a}_\nu^\dagger\,\Omega \qquad (1.61)\end{aligned}$$

and modes $\hat{a}$ are uniquely characterized by the corresponding 1-photon state vectors $\hat{a}^\dagger\,\Omega$. Therefore, the field operators $\hat{a}_j(\mathbf{k})$ may be replaced by the countable set of ordinary operators $\hat{a}_\nu$ and every normalized 1-photon state vector $\Psi^{(1)}$ may be written in the form

$$\Psi^{(1)} = \sum_{\nu=1}^{\infty} \lambda_\nu\,\hat{a}_\nu^\dagger\,\Omega\,, \qquad \sum_{\nu=1}^{\infty} |\lambda_\nu|^2 = 1\,,$$

where the complex coefficients $\lambda$ are the ***probability amplitudes*** for the 'modes' $\hat{a}_\nu$, i.e.:



[29] If $\sum_{j=1}^{2} \boldsymbol{\epsilon}_j(\mathbf{k})\, f_\nu^j(\mathbf{k}) \propto \boldsymbol{\epsilon}_1(\mathbf{k}) + i\,\boldsymbol{\epsilon}_2(\mathbf{k})$ resp. $\sum_{j=1}^{2} \boldsymbol{\epsilon}_j(\mathbf{k})\, f_\nu^j(\mathbf{k}) \propto \boldsymbol{\epsilon}_1(\mathbf{k}) - i\,\boldsymbol{\epsilon}_2(\mathbf{k})$ the photon is said to have positive resp. negative ***helicity***.

For every $\nu_0 \in \mathbb{N}$
$$|\lambda_{\nu_0}| = \left|\left\langle \hat{a}_\nu^\dagger\,\Omega \,\middle|\, \Psi^{(1)}\right\rangle\right|^2$$
is the probability for finding a photon, randomly chosen from an ensemble (characterized by) $\Psi^{(1)}$, in the mode $\hat{a}_\nu^\dagger$ **if** an ideal test for being in just one one the modes $\hat{a}_1, \hat{a}_2, \ldots$ is performed.[30]

Obviously, for $\nu \in \mathbb{N}$,
$$\hat{n}_\nu \stackrel{\text{def}}{=} \hat{a}_\nu^\dagger\,\hat{a}_\nu$$
is the observable for the number of photons in mode $\hat{a}_\nu$ and for the observable $\hat{N}$ for the total number of photons we get
$$\hat{N} = \sum_{\nu=1}^{n} \hat{n}_\nu\,.$$

**Warning:** According to the simple theory of photodetection considered in 7.2.2 the states corresponding to elements of $\mathcal{H}_{\text{field}}^{(n)}$ are, in fact, $n$-fold localized: The maximal number of photodetectors which may fire simultaneously when testing such a state is $n$.

Typical for ***nonlinear quantum optics***:
$$\hat{b}_\nu^\dagger\,\omega \longmapsto \hat{c}_\nu^\dagger\,\Omega \quad \forall\,\nu \qquad \not\Longrightarrow \qquad \hat{b}_1^\dagger\cdots\hat{b}_n^\dagger\,\Omega \longmapsto \hat{c}_1^\dagger\cdots\hat{c}_n^\dagger\,\Omega \quad \forall\,n\,.$$
Such 'nonlinear' behavior is needed, e.g., for optical implementations of the CNOT gate acting according to
$$\begin{array}{rcl} \mathbf{H}\otimes\mathbf{H} & \longmapsto & \mathbf{H}\otimes\mathbf{H}\,,\\ \mathbf{H}\otimes\mathbf{V} & \longmapsto & \mathbf{H}\otimes\mathbf{V}\,,\\ \mathbf{V}\otimes\mathbf{V} & \longmapsto & \mathbf{V}\otimes\mathbf{H}\,,\\ \mathbf{V}\otimes\mathbf{H} & \longmapsto & \mathbf{V}\otimes\mathbf{V}\,. \end{array}$$

Finally, let us derive some useful relations for modes $\hat{a}$:

A simple consequence of the commutation relations (1.37) and (1.59) is
$$\left(\hat{a}^\dagger\hat{a}\right)\hat{a} = \hat{a}\left(\hat{a}^\dagger\hat{a} - 1\right)\,, \quad \left(\hat{a}^\dagger\hat{a}\right)\hat{a}^\dagger = \hat{a}^\dagger\left(\hat{a}^\dagger\hat{a} + 1\right) \tag{1.62}$$
which, by iteration, implies
$$\left(\hat{a}^\dagger\hat{a}\right)\left(\hat{a}\right)^n = \left(\hat{a}\right)^n\left(\hat{a}^\dagger\hat{a} - n\right)\,, \quad \left(\hat{a}^\dagger\hat{a}\right)\left(\hat{a}^\dagger\right)^n = \left(\hat{a}^\dagger\right)^n\left(\hat{a}^\dagger\hat{a} + n\right)$$



[30] Typical for quantum mechanics is, that such tests are considered even if
$$\Psi^{(1)} = \hat{a}^\dagger\,\Omega\,, \qquad \hat{a} \neq \hat{a}_\nu \quad \forall\,\nu \in \mathbb{N}\,.$$

for all $n \in \mathbb{N}$. This, in turn, implies[31]

$$\left(\hat{a}^\dagger\right)^n \hat{a}^n = \left(\hat{a}^\dagger \hat{a}\right)\left(\hat{a}^\dagger \hat{a} - 1\right)\cdots\left(\hat{a}^\dagger \hat{a} - (n-1)\right) \quad \forall\, n \in \mathbb{N} \tag{1.63}$$

and

$$\left(\hat{a}^\dagger \hat{a}\right)\left(\hat{a}^\dagger\right)^n \Omega = n\left(\hat{a}^\dagger\right)^n \Omega \quad \forall\, n \in \mathbb{N}\,. \tag{1.64}$$

### 1.2.4 'Classical' States of Light

> *They are important,*[32] *not only because they are one of the quantum mechanical states whose properties most closely resemble those of a classical electromagnetic wave, but also because a single-mode laser operated well above threshold generates a coherent state excitation...*
>
> (Loudon, 2000, p. 190)

#### Radiation off Classical Currents

The expectation values for the (no longer free) electromagnetic field associated with a **classical** 4-current $\left(c\,\rho(\mathbf{x},t), \boldsymbol{\jmath}(\mathbf{x},t)\right)$ in the vacuum should fulfill the corresponding MAXWELL equations:

$$\begin{aligned}
\operatorname{curl}\langle \mathbf{B}(\mathbf{x},t)\rangle &= \mu_0\left(\epsilon_0 \frac{\partial}{\partial t}\langle \mathbf{E}(\mathbf{x},t)\rangle + \boldsymbol{\jmath}(\mathbf{x},t)\right), &(1.65)\\
\operatorname{curl}\langle \mathbf{E}(\mathbf{x},t)\rangle &= -\frac{\partial}{\partial t}\langle \mathbf{B}(\mathbf{x},t)\rangle\,, &(1.66)\\
\operatorname{div}\langle \mathbf{E}(\mathbf{x},t)\rangle &= \frac{1}{\epsilon_0}\,\rho(\mathbf{x},t)\,, &(1.67)\\
\operatorname{div}\langle \mathbf{B}(\mathbf{x},t)\rangle &= 0\,. &(1.68)
\end{aligned}$$

In the **interaction picture** the expectation values (for *pure* states) are given by[33]

$$\begin{aligned}
\langle \mathbf{B}(\mathbf{x},t)\rangle &= \left\langle \Psi_t^{\mathrm{I}} \,\middle|\, \hat{\mathbf{B}}(\mathbf{x},t)\,\Psi_t^{\mathrm{I}}\right\rangle\,,\\
\langle \mathbf{E}(\mathbf{x},t)\rangle &= \left\langle \Psi_t^{\mathrm{I}} \,\middle|\, \left(\hat{\mathbf{E}}(\mathbf{x},t) - \operatorname{grad}\int \frac{\rho(\mathbf{x}',t)}{4\pi\,\epsilon_0\,|\mathbf{x}-\mathbf{x}'|}\,dV_{\mathbf{x}'}\right)\Psi_t^{\mathrm{I}}\right\rangle
\end{aligned} \tag{1.69}$$

and, therefore, the equations (1.65)–(1.68) are guaranteed by the $t$-dependence

$$i\hbar \frac{d}{dt}\,\Psi_t^{\mathrm{I}} = \hat{H}_{\mathrm{int}}(t)\,\Psi_t^{\mathrm{I}}\,, \tag{1.70}$$

of the **normalized** state vector $\Psi_t^{\mathrm{I}}$, where

$$\hat{H}_{\mathrm{int}}(t) \stackrel{\mathrm{def}}{=} -\int \boldsymbol{\jmath}(\mathbf{x},t)\cdot\hat{\mathbf{A}}(\mathbf{x},t)\,dV_{\mathbf{x}}\,. \tag{1.71}$$

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[31] Compare with (A.31).

[32] See also (Klauder, 2010).

[33] Note that, in the presence of charges, $\hat{\mathbf{E}}(\mathbf{x},t)$ is only the **transversal** part of the observable for the field strength in the interaction picture (see also Footnote 2 of Chapter 7).

**Outline of proof for (1.65)–(1.68):** While (1.68) resp. (1.67) follows directly from (1.69) and (1.42) resp. (1.41), (1.66) follows according to

$$\begin{aligned}
\operatorname{curl}\langle \mathbf{E}(\mathbf{x},t)\rangle &\underset{(1.69)}{=} \operatorname{curl}\left\langle \Psi_t^{\mathrm{I}} \middle| \hat{\mathbf{E}}(\mathbf{x},t)\,\Psi_t^{\mathrm{I}}\right\rangle \\
&\underset{(1.40)}{=} \left\langle \Psi_t^{\mathrm{I}} \middle| \frac{\partial}{\partial t}\hat{\mathbf{B}}(\mathbf{x},t)\,\Psi_t^{\mathrm{I}}\right\rangle \\
&\underset{(1.70)}{=} \tfrac{\partial}{\partial t}\left\langle \Psi_t^{\mathrm{I}} \middle| \hat{\mathbf{B}}(\mathbf{x},t)\,\Psi_t^{\mathrm{I}}\right\rangle - \left\langle \Psi_t^{\mathrm{I}} \middle| \underbrace{\frac{i}{\hbar}\left[\hat{H}_{\mathrm{int}}(t),\hat{\mathbf{B}}(\mathbf{x},t)\right]_-}_{\underset{(1.71),(1.48)}{=}0}\Psi_t^{\mathrm{I}}\right\rangle .
\end{aligned}$$

(1.65), finally, follows from

$$\begin{aligned}
\frac{1}{\mu_0\,\epsilon_0}\operatorname{curl}\langle \mathbf{B}(\mathbf{x},t)\rangle &\underset{(1.39)}{=} \left\langle \Psi_t^{\mathrm{I}} \middle| \frac{\partial}{\partial t}\hat{\mathbf{E}}(\mathbf{x},t)\,\Psi_t^{\mathrm{I}}\right\rangle \\
&\underset{(1.70)}{=} \frac{\partial}{\partial t}\left\langle \Psi_t^{\mathrm{I}} \middle| \hat{\mathbf{E}}(\mathbf{x},t)\,\Psi_t^{\mathrm{I}}\right\rangle - \left\langle \Psi_t^{\mathrm{I}} \middle| \frac{i}{\hbar}\left[\hat{H}_{\mathrm{int}}(t),\hat{\mathbf{E}}(\mathbf{x},t)\right]_-\Psi_t^{\mathrm{I}}\right\rangle
\end{aligned}$$

and

$$\begin{aligned}
-\tfrac{i}{\hbar}\left[\hat{H}_{\mathrm{int}}(t),\hat{\mathbf{E}}(\mathbf{x},t)\right]_- &\underset{(1.71)}{=} -\frac{i}{\hbar}\int \boldsymbol{\jmath}(\mathbf{x}',t)\cdot\left[\hat{\mathbf{A}}_0(\mathbf{x}',t),\hat{\mathbf{E}}(\mathbf{x},t)\right]_-\,dV_{\mathbf{x}'} \\
&\underset{(1.49),(1.51)}{=} \frac{1}{\epsilon}\,\boldsymbol{\jmath}(\mathbf{x}) + \operatorname{grad}\int\frac{\operatorname{div}\boldsymbol{\jmath}(\mathbf{x}')}{4\pi\,\epsilon_0\,|\mathbf{x}-\mathbf{x}'|}\,dV_{\mathbf{x}} \\
&\underset{\text{cont. eq.}}{=} \frac{1}{\epsilon}\,\boldsymbol{\jmath}(\mathbf{x},t) - \frac{\partial}{\partial t}\operatorname{grad}\int\frac{\rho(\mathbf{x}',t)}{4\,\pi\,\epsilon_0\,|\mathbf{x}-\mathbf{x}'|}\,dV_{\mathbf{x}'}\,. \qquad \blacksquare
\end{aligned}$$

If

$$\left.\begin{array}{rcl}\rho(\mathbf{x},t) &=& 0\\ \boldsymbol{\jmath}(\mathbf{x},t) &=& 0\end{array}\right\}\quad \forall\, t<0 \tag{1.72}$$

then the ***retarded*** solution of (1.70), characterized by

$$\Psi_t^{\mathrm{I}} = \Omega \quad \forall\, t<0\,,$$

is[34]

$$\Psi_t^{\mathrm{I}} = e^{\lambda(t)}\,e^{\hat{A}_t}\,,\quad \hat{A}_t \stackrel{\text{def}}{=} -\frac{i}{\hbar}\int_{-\infty}^{t}\hat{H}_{\mathrm{int}}(t')\,dt' \tag{1.73}$$

(for sufficiently well-behaved $\rho, \boldsymbol{\jmath}$), where

$$\lambda(t) \stackrel{\text{def}}{=} \frac{1}{2}\int_{-\infty}^{t}\frac{d}{dt'}\left[\hat{A}_{t'},\hat{A}_t\right]_-\,dt' \in \mathbb{C}\,.$$

**Remark:** We assume, without proof, that this solution is unique. For the expectation values this is a simple consequence of POYNTING's theorem; see, e.g., Section 5.1.2 of (Lücke, edyn).



[34]For more complicated interactions the DYSON series (see A.4) has to be evaluated.

That this is a solution can be shown by application of the BAKER-HAUSDORFF ***formula***[35]

$$\left[\hat{A},[\hat{A},\hat{B}]_-\right]_- = \left[\hat{B},[\hat{A},\hat{B}]_-\right]_- = 0 \implies e^{\hat{A}+\hat{B}} = e^{-\frac{1}{2}[\hat{A},\hat{B}]_-}\, e^{\hat{A}}\, e^{\hat{B}}\,. \tag{1.74}$$

**Outline of proof for ‘ (1.73) $\Longrightarrow$ (1.70)’:**

$$\begin{aligned}
&\frac{\mathrm{d}}{\mathrm{d}t}\, e^{\hat{A}_t}\,\Omega \\
&= \lim_{\Delta t\to+0} \frac{e^{\hat{A}_{t+\Delta t}}e^{-\hat{A}_t}-1}{\Delta t}\, e^{\hat{A}_t}\,\Omega \\
&\underset{(1.74)}{=} \lim_{\Delta t\to+0} \frac{e^{\hat{A}_{t+\Delta t}-\hat{A}_t}e^{-\frac{1}{2}\left[\hat{A}_{t+\Delta t},\hat{A}_t\right]_-}-1}{\Delta t}\, e^{\hat{A}_t}\,\Omega \\
&= \lim_{\Delta t\to+0} \left(\frac{e^{\hat{A}_{t+\Delta t}-\hat{A}_t}-1}{\Delta t}\, e^{-\frac{1}{2}\left[\hat{A}_{t+\Delta t},\hat{A}_t\right]_-} + \frac{e^{-\frac{1}{2}\left[\hat{A}_{t+\Delta t},\hat{A}_t\right]_-}-1}{\Delta t}\right) e^{\hat{A}_t}\,\Omega \\
&\underset{(1.73)}{=} \left(-\frac{i}{\hbar}\,\hat{H}_{\mathrm{int}} - \lambda'(t)\right) e^{\hat{A}_t}\,\Omega\,. \qquad \blacksquare
\end{aligned}$$

Since

$$\frac{i}{\hbar}\int_{-\infty}^{t} \hat{H}_{\mathrm{int}}(t')\,\mathrm{d}t' = \hat{a}_{\boldsymbol{\jmath}}(t) - \left(a_{\boldsymbol{\jmath}}(t)\right)^{\dagger}, \tag{1.75}$$

where

$$a_{\boldsymbol{\jmath}}(t) \stackrel{\text{def}}{=} \sum_{j=1,2}\int \left(g^j(\mathbf{k},t)\right)^* \hat{a}_j(\mathbf{k})\,\mathrm{d}V_{\mathbf{k}}\,,$$

and

$$g^j(\mathbf{k},t) \stackrel{\text{def}}{=} \frac{i}{\hbar}\sqrt{\frac{\mu_0\hbar c}{2\,|\mathbf{k}|}}\,\boldsymbol{\epsilon}_j(\mathbf{k})\cdot\int_{-\infty}^{t}\left((2\pi)^{-\frac{3}{2}}\int \boldsymbol{\jmath}\,(\mathbf{x},t')\,e^{-i\,\mathbf{k}\cdot\mathbf{x}}\,\mathrm{d}V_{\mathbf{x}}\right) e^{+i\,c|\mathbf{k}|t'}\,\mathrm{d}t'\,,$$

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[35]For operators in **finite** dimensional vector spaces (1.74) may be proved as follows: Since $e^{\lambda\hat{A}}\hat{B}\,e^{-\lambda\hat{A}}$ and $e^{\mathrm{ad}_{\lambda\hat{A}}}\hat{B}$ ($\mathrm{ad}_{\hat{C}}\,\hat{D} \stackrel{\text{def}}{=} [\hat{C},\hat{D}]_-$) fulfill the same first order differential equation and initial condition (for $\lambda=0$), the CAMPBELL-HAUSDORFF ***formula***

$$e^{\hat{A}}\,\hat{B}\,e^{-\hat{A}} = e^{\mathrm{ad}_{\hat{A}}}\,\hat{B}$$

holds for arbitrary $\hat{A}$, $\hat{B}$. Therefore, also

$$f_1(\lambda) \stackrel{\text{def}}{=} e^{\lambda\left(\hat{A}+\hat{B}\right)+\frac{\lambda^2}{2}\,[\hat{A},\hat{B}]_-}$$

and

$$f_2(\lambda) \stackrel{\text{def}}{=} e^{\lambda\,\hat{A}}\,e^{\lambda\,\hat{B}}$$

fulfill the same first order differential equation and initial condition (for $\lambda=0$) and hence $f_1=f_2$ **if** the l.h.s. of (1.74) holds. For details concerning the case of unbounded operators $\hat{A},\hat{B}$ see (Fröhlich, 1977).

(1.73) can be written in the form

$$\Psi_t^{\mathrm{I}} = e^{\lambda(t)}\, e^{\left(a_{\jmath}(t)\right)^\dagger - \hat{a}_{\jmath}(t)}\, \Omega\,. \tag{1.76}$$

**Proof of (1.75):** Using the general definition

$$\mathbf{G}(\mathbf{k},t) \stackrel{\text{def}}{=} (2\pi)^{-\frac{3}{2}} \int \mathbf{G}(\mathbf{x},t) e^{-i\,\mathbf{k}\cdot\mathbf{x}}\, dV_{\mathbf{x}}$$

we get

$$\hat{H}_{\text{int}} \underset{(1.71)}{=} -\int \jmath_{\text{cr}}(-\mathbf{k},t)\cdot \widehat{\mathbf{A}}(\mathbf{k},t)\, dV_{\mathbf{k}}$$

and

$$\widehat{\mathbf{A}}(\mathbf{k},t) \underset{(1.31),(1.34)}{=} \sqrt{\frac{\mu_0 \hbar c}{2\,|\mathbf{k}|}} \sum_{j=1,2} \left(\boldsymbol{\epsilon}_j(\mathbf{k})\, \hat{a}_j(\mathbf{k})\, e^{-i\,c|\mathbf{k}|t} + \boldsymbol{\epsilon}_j(-\mathbf{k}) \left(\hat{a}_j(-\mathbf{k})\right)^\dagger e^{+i\,c|\mathbf{k}|t}\right).$$

Since

$$\jmath(\mathbf{x},t) = \left(\jmath(\mathbf{x},t)\right)^* \quad \Longrightarrow \quad \jmath(-\mathbf{k},t) = \left(\jmath(+\mathbf{k},t)\right)^*,$$

this implies (1.75). ∎

**Remarks:**

1. The state of the electromagnetic field depends only on the time-dependent vector field $\jmath(\mathbf{x},t)$. This is no surprise since the latter — together with the continuity equation and (1.72) — fixes the time-dependent scalar field $\rho(\mathbf{x},t)$.
2. Note that for time intervals without interaction[36] the interaction picture coincides with the free Heisenberg picture.
3. This is why $\hat{a}_{\jmath}(t)$ does not change during time intervals over which $\jmath(\mathbf{x},t)$ vanishes.

### Single Modes

Obviously, the state (1.76) generated from the vacuum by an exterior current is a ***coherent state***,[37] i.e. a pure state corresponding to a state vector $\chi$ of the form

$$\chi \propto \hat{D}_{\hat{a}}(\alpha)\,\Omega\,, \quad \alpha \in \mathbb{C}\,, \tag{1.77}$$

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[36] For the present case this means that $\jmath(\mathbf{x},t) = 0$ over the corresponding time intervals.

[37] These states are called *coherent* since their analogs for the harmonic oscillator correspond to wave packets which do not spread but — apart form shifts of the expectation values — have the same probability distributions for position and momentum as the ground state, for all times; see A.2.3.

where $\hat{a}$ is a mode and

$$\hat{D}_{\hat{a}}(\alpha) \stackrel{\text{def}}{=} e^{\alpha\,\hat{a}^\dagger - \alpha^*\,\hat{a}} \quad \forall\,\alpha \in \mathbb{C}\,. \tag{1.78}$$

Obviously, the operators $\hat{D}_{\hat{a}}(\alpha)$ are unitary. Using the BAKER-HAUSDORFF formula (1.74) we easily see that

$$\hat{D}_{\hat{a}}(\alpha) = e^{+\frac{1}{2}|\alpha|^2}\, e^{-\alpha^*\,\hat{a}}\, e^{\alpha\,\hat{a}^\dagger} = e^{-\frac{1}{2}|\alpha|^2}\, e^{\alpha\,\hat{a}^\dagger}\,\Omega \quad \forall\,\alpha \in \mathbb{C}\,. \tag{1.79}$$

The latter especially implies

$$\begin{aligned} \chi_{\hat{a}}(\alpha) &\stackrel{\text{def}}{=} \underbrace{e^{-\frac{1}{2}|\alpha|^2}}_{\text{normalizing factor}} e^{\alpha\,\hat{a}^\dagger}\,\Omega \\ &= \hat{D}_{\hat{a}}(\alpha)\,\Omega \quad \forall\,\alpha \in \mathbb{C}\,. \end{aligned} \tag{1.80}$$

Now, the relation

$$\left[\hat{a}, e^{\alpha\,\hat{a}^\dagger}\right]_- = \alpha\, e^{\alpha\,\hat{a}^\dagger}\,, \tag{1.81}$$

corresponding to the formal rule

$$[\hat{A},\hat{B}]_- \propto 1 \implies [\hat{A}, f(\hat{B})]_- = [\hat{A},\hat{B}]_-\, f'(\hat{B})\,, \tag{1.82}$$

(compare Exercise E29a) of (Lücke, eine)) shows that

$$\hat{a}\,\chi_{\hat{a}}(\alpha) = \alpha\,\chi_{\hat{a}}(\alpha) \quad \forall\,\alpha \in \mathbb{C} \tag{1.83}$$

More generally we have

$$\begin{aligned} \hat{a}_j(\mathbf{k})\,\hat{D}_{\hat{a}}(\alpha)\,\Omega &= \left[\hat{a}_j(\mathbf{k}), \hat{D}_{\hat{a}}(\alpha)\right]_-\,\Omega \\ &\underset{(1.82)}{=} \alpha\left[\hat{a}_j(\mathbf{k}), \hat{a}^\dagger\right]_-\,\hat{D}_{\hat{a}}(\alpha)\,\Omega \end{aligned}$$

and

$$\begin{aligned} \left[\hat{a}_j(\mathbf{k}), \hat{a}^\dagger\right]_- &= \left\langle \Omega \,\middle|\, \left[\hat{a}_j(\mathbf{k}), \hat{a}^\dagger\right]_-\,\Omega \right\rangle \\ &= \left\langle \Omega \,\middle|\, \hat{a}_j(\mathbf{k})\,\hat{a}^\dagger\,\Omega \right\rangle\,, \end{aligned}$$

hence

$$\hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\,\chi_{\hat{a}}(\alpha) = \underbrace{\left\langle \Omega \,\middle|\, \hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\,\alpha\,\hat{a}^\dagger\,\Omega \right\rangle}_{\substack{= \text{ complex vector potential of the expectation value} \\ \text{of the electromagnetic field in the state } \chi_{\hat{a}}(\alpha) \\ = \ \alpha\times \textit{wave function} \text{ of a photon in the state } \hat{a}^\dagger\,\Omega}} \chi_{\hat{a}}(\alpha)\,. \tag{1.84}$$

Thus:

> In a coherent state $\chi$ the (vector-valued) 'wave function' of every photon coincides (up to normalization) with the complex vector potential $\left\langle \chi \,\middle|\, \hat{\mathbf{A}}_0^{(+)}(\mathbf{x},t)\,\chi \right\rangle$.

**Remarks:**

1. For (Fock-)states of the form
$$\Psi^{(n)} = \frac{1}{\sqrt{n!}}\,\hat{a}^\dagger\,\Omega$$
the phase of the single-photon mode $\hat{a}$ is irrelevant. But it is crucial for coherent superpositions of such states with different values of $n$.

2. The unitary $\hat{D}_{\hat{a}}(\alpha)$ a displacement operator in the sence that[38]
$$[\hat{a}, \hat{D}_{\hat{a}}(\alpha)]_- \underset{(1.81)}{=} \alpha\,\hat{D}_{\hat{a}}(\alpha)\,, \quad [\hat{a}^\dagger, \hat{D}_{\hat{a}}(\alpha)] \underset{(1.81)}{=} \overline{\alpha}\,\hat{D}_{\hat{a}}(\alpha)\,. \quad (1.85)$$

In coherent states $\chi_{\hat{c}}(\alpha)$ the so-called ***quadrature components***
$$\hat{x}_{\hat{a}} \stackrel{\text{def}}{=} \frac{\hat{a}+\hat{a}^\dagger}{\sqrt{2}}\,, \quad \hat{p}_{\hat{a}} \stackrel{\text{def}}{=} \frac{\hat{a}-\hat{a}^\dagger}{i\sqrt{2}}$$
of an arbitrary mode $\hat{a}$ have the same uncertainties as in the vacuum state and their product takes the minimum value allowed by the uncertainty relations[39]
$$\Delta x_{\hat{a}}\,\Delta p_{\hat{a}} \geq \frac{1}{2}\left|[\hat{x}_{\hat{a}}, \hat{p}_{\hat{a}}]_-\right| = \frac{1}{2}\,.$$

**Outline of proof:** With
$$\lambda \stackrel{\text{def}}{=} [\hat{a}, \alpha\,\hat{c}^\dagger]_-$$
we have
$$\begin{aligned}\langle\hat{x}_{\hat{a}}\hat{a}\rangle &= \left\langle\chi_{\hat{c}}(\alpha)\,\middle|\,\hat{x}_{\hat{a}}\,\chi_{\hat{c}}(\alpha)\right\rangle\\ &= \frac{1}{\sqrt{2}}\left(\left\langle\chi_{\hat{c}}(\alpha)\,\middle|\,\hat{a}\,\chi_{\hat{c}}(\alpha)\right\rangle + \left\langle\hat{a}\,\chi_{\hat{c}}(\alpha)\,\middle|\,\chi_{\hat{c}}(\alpha)\right\rangle\right)\\ &= \frac{1}{\sqrt{2}}\,(\lambda+\lambda^*)\end{aligned}$$
and
$$\begin{aligned}2\left\langle(\hat{x}_{\hat{a}})^2\right\rangle &= \left\langle\chi_{\hat{c}}(\alpha)\,\middle|\,\left((\hat{a})^2 + ((\hat{a}^\dagger)^2 + 2\,\hat{a}^\dagger\,\hat{a} + \hat{1}\right)\chi_{\hat{c}}(\alpha)\right\rangle\\ &= (\lambda^*)^2 + \lambda^2 + 2\,\lambda^*\,\lambda + 1\\ &= (\lambda+\lambda^*)^2 + 1\,,\end{aligned}$$



[38]Note that (1.85) implies
$$\hat{a}\,\Psi = \alpha\,\Psi \implies \hat{a}\left(\hat{D}_{\hat{a}}(\beta)\,\Psi\right) = (\alpha+\beta)\left(\hat{D}_{\hat{a}}(\beta)\,\Psi\right).$$
Actually:
$$\hat{D}_{\hat{a}}(\alpha)\,\hat{D}_{\hat{a}}(\beta) \underset{(1.74)}{=} e^{\frac{1}{2}\left(\alpha\,\overline{\beta}-\overline{\alpha}\,\beta\right)}\,\hat{D}_{\hat{a}}(\alpha+\beta) \quad \forall\,\alpha,\beta\in\mathbb{C}\,.$$

[39]Siehe Footnote 14 of Appendix A.

hence

$$\begin{aligned}(\Delta x_{\hat{a}})^2 &= \left\langle (\hat{x}_{\hat{a}})^2 \right\rangle - (\langle \hat{x}_{\hat{a}} \rangle)^2 \\ &= 1/2\,.\end{aligned}$$

Similarly we get

$$(\Delta p_{\hat{a}})^2 = 1/2\,. \qquad \blacksquare$$

In this sense coherent states are 'as classical as possible'.[40]

**Remarks:**

1. For every $t \in \mathbb{R}$ and arbitrary real-valued $f^1(\mathbf{x}), f^1(\mathbf{x}), f^1(\mathbf{x}) \in \mathcal{S}(\mathbb{R}^3)$ there is a mode $\hat{a}$ with

$$\int \hat{\mathbf{E}}(\mathbf{x},t) \cdot \mathbf{f}(\mathbf{x})\, \mathrm{d}V_{\mathbf{x}} \propto \hat{x}_{\hat{a}}\,.$$

2. If $\hat{a}$ is a mode then so is $e^{i\varphi}\,\hat{a}$ for every $\varphi \in \mathbb{R}$.
3. For every mode $\hat{a}$ we have $\hat{p}_{\hat{a}} = \hat{x}_{-i\,\hat{a}}$.

Let $\hat{a}$ be a mode and denote by $\mathcal{H}_{\hat{a}}$ the smallest closed subspace of $\mathcal{H}_{\text{field}}$ that contains $\Omega$ and is invariant under $\hat{a}^\dagger$. Then[41]

$$\sum_{\nu=0}^{\infty} \frac{1}{\nu!} \left| \left(\hat{a}^\dagger\right)^\nu \Omega \right\rangle\!\left\langle \left(\hat{a}^\dagger\right)^\nu \Omega \right| = \hat{P}_{\mathcal{H}_{\hat{a}}}\,, \tag{1.86}$$

holds as a consequence of

$$\left\langle \left(\hat{a}^\dagger\right)^\nu \Omega \mid \left(\hat{a}^\dagger\right)^\mu \Omega \right\rangle = \nu!\,\delta_{\nu\mu}\,. \tag{1.87}$$

**Proof of (1.87):** The statement follows by successive application of

$$\begin{aligned}\left\langle \hat{a}^\dagger \Psi \mid \left(\hat{a}^\dagger\right)^\mu \Omega \right\rangle &= \left\langle \Psi \mid \left[\hat{a}\,, \left(\hat{a}^\dagger\right)^\mu\right]_- \Omega \right\rangle \\ &\underset{(1.82),(1.59)}{=} \mu \left\langle \Psi \mid \left(\hat{a}^\dagger\right)^{\mu-1} \Omega \right\rangle \quad \forall\, \mu \in \mathbb{N}\,,\ \Psi \in \mathcal{H}_{\text{field}}\,. \qquad \blacksquare\end{aligned}$$

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[40] States with $\Delta x_{\hat{a}} < 1/\sqrt{2}$ or $\Delta p_{\hat{a}} < 1/\sqrt{2}$ are called ***squeezed states*** (see (Scully and Zubairy, 1999, Sections 2.5–2.8) for a discussion of these states).

[41] As usual, $\hat{P}_{\mathcal{H}_{\hat{a}}}$ denotes the orthogonal projection onto $\mathcal{H}_{\hat{a}}$.

(1.86) implies[42]

$$\frac{1}{\pi}\int \left|\hat D_{\hat a}(x+iy)\,\Omega\right\rangle\left\langle \hat D_{\hat a}(x+iy)\,\Omega\right|\,dx\,dy = \hat P_{\mathcal{H}_{\hat a}}\,, \tag{1.88}$$

i.e.[43]

$$\chi = \frac{1}{\pi}\int \left\langle \hat D_{\hat a}(x+iy)\,\Omega \mid \chi\right\rangle D_{\hat a}(x+iy)\,\Omega\,dx\,dy \quad \forall\,\chi\in\mathcal{H}_{\hat a}\,.$$

**Proof of (1.88):** According to (1.86) and (1.87) we have to show

$$\left\langle \left(\hat a^\dagger\right)^\nu\Omega\middle|\left(\int\left|\hat D_{\hat a}(x+iy)\,\Omega\right\rangle\left\langle\hat D_{\hat a}(x+iy)\,\Omega\right|\,dx\,dy\right)\left(\hat a^\dagger\right)^\mu\Omega\right\rangle = \pi\,\nu!\,\delta_{\nu\mu}\,.$$

This, however, may be shown as follows:

$$\begin{aligned}
&\left\langle \left(\hat a^\dagger\right)^\nu\Omega\middle|\left(\int\left|\hat D_{\hat a}(x+iy)\,\Omega\right\rangle\left\langle\hat D_{\hat a}(x+iy)\,\Omega\right|\,dx\,dy\right)\left(\hat a^\dagger\right)^\mu\Omega\right\rangle\\
&= \int \left\langle \left(\hat a^\dagger\right)^\nu\Omega \mid \hat D_{\hat a}(x+iy)\,\Omega\right\rangle\left\langle \hat D_{\hat a}(x+iy)\,\Omega\mid\left(\hat a^\dagger\right)^\mu\Omega\right\rangle dx\,dy\\
&\underset{(1.79)}{=} \int e^{-|x+iy|^2}\left\langle\left(\hat a^\dagger\right)^\nu\Omega\mid e^{(x+iy)\hat a^\dagger}\,\Omega\right\rangle\left\langle e^{(x+iy)\hat a^\dagger}\,\Omega\mid\left(\hat a^\dagger\right)^\mu\Omega\right\rangle dx\,dy\\
&= \int e^{-|x+iy|^2}\sum_{\nu',\mu'=0}^{\infty}\frac{(x+iy)^{\nu'}\,(x-iy)^{\mu'}}{\nu'!\,\mu'!}\left\langle\left(\hat a^\dagger\right)^\nu\Omega\mid\left(\hat a^\dagger\right)^{\nu'}\,\Omega\right\rangle\cdot\\
&\qquad\qquad\cdot\left\langle\left(\hat a^\dagger\right)^{\mu'}\,\Omega\mid\left(\hat a^\dagger\right)^\mu\Omega\right\rangle dx\,dy\\
&\underset{(1.87)}{=} \int (x+iy)^\nu\,(x-iy)^\mu\;e^{-|x+iy|^2}\,dx\,dy\\
&= \int_0^\infty r^{\nu+\mu+1}\,e^{-r^2}\,dr\int_0^{2\pi}e^{i(\nu-\mu)\varphi}\,d\varphi\\
&= \frac{1}{2}\int_0^\infty \xi^{(\nu+\mu)/2}\,e^{-\xi}\,d\xi\int_0^{2\pi}e^{i(\nu-\mu)\varphi}\,d\varphi\\
&= \pi\,\nu!\,\delta_{\nu\mu}\,. \quad\blacksquare
\end{aligned}$$



[42]The integral is to be understood in the weak sense:

$$\begin{aligned}
&\left\langle\chi_1\middle|\left(\int\left|\hat D_{\hat a}(x+iy)\,\Omega\right\rangle\left\langle\hat D_{\hat a}(x+iy)\,\Omega\right|\,dx\,dy\right)\chi_2\right\rangle\\
&\stackrel{\text{def}}{=}\int\left\langle\chi_1\middle|\hat D_{\hat a}(x+iy)\,\Omega\right\rangle\left\langle\hat D_{\hat a}(x+iy)\,\Omega\middle|\,\chi_2\right\rangle dx\,dy \quad \forall\,\chi_1,\chi_2\in\mathcal{H}_{\text{field}}\,.
\end{aligned}$$

[43]Note that, for every $\epsilon>0$, already $\left\{\hat D_{\hat a}(\alpha)\,\Omega:\ \alpha\in\mathbb{C}\,,\ |\alpha|<\epsilon\right\}$ is a complete set of states since, e.g.,

$$\left|\left(\hat a^\dagger\right)^n\,\Omega\right\rangle = \left(\left(\frac{d}{d\alpha}\right)^n e^{\alpha\,\hat a^\dagger}\,\Omega\right)_{|\alpha=0}\quad\forall\,n\in\mathbb{Z}_+$$

and

$$\left|\left(\hat a^\dagger\right)^n\,\Omega\right\rangle = \frac{n!}{2\pi}\,e^{r^2/2}\,r^{-n}\int e^{-n\varphi}\left|\hat D_{\hat a}\left(r\,e^{i\varphi}\right)\Omega\right\rangle d\varphi\quad\forall\,n\in\mathbb{Z}_+\,,\ r>0\,.$$

By (1.88), every vector in $\mathcal{H}_{\hat{a}}$ way be written as a continuous superposition of coherent states $\left|\hat{D}_{\hat{a}}(x+iy)\,\Omega\right\rangle$, $z \in \mathbb{C}$. But, of course, this representation is not unique:[44]

$$\left\{\left|\hat{D}_{\hat{a}}(x+iy)\,\Omega\right\rangle : z \in \mathbb{C}\right\} \text{ is an } \textit{over-complete} \text{ subset of } \mathcal{H}_{\hat{a}}\,.$$

The states $\hat{D}_{\hat{a}}(\alpha)\,\Omega$ and $\hat{D}_{\hat{a}}(\alpha)\,\Omega$ are only approximately orthogonal for large $|\alpha-\beta|$ :

$$\begin{aligned}\left\langle \hat{D}_{\hat{a}}(\alpha)\,\Omega \mid \hat{D}_{\hat{a}}(\beta)\,\Omega\right\rangle &\underset{(1.79)}{=} e^{-\frac{1}{2}\left(|\alpha|^2+|\beta|^2\right)}\langle\Omega \mid \underbrace{e^{\alpha^*\hat{a}}\,e^{\beta\hat{a}^\dagger}\,\Omega}_{\underset{(1.83)}{=}\,e^{\alpha^*\beta}\,e^{\beta\hat{a}^\dagger}\,\Omega}\rangle\\ &= e^{-\frac{1}{2}\left(|\alpha|^2+|\beta|^2\right)+\alpha^*\beta}\left\langle e^{\beta^*\hat{a}}\,\Omega \mid \Omega\right\rangle\\ &= e^{-\frac{1}{2}\left(|\alpha|^2+|\beta|^2\right)+\alpha^*\beta}\end{aligned} \tag{1.89}$$

### Multiple Modes

More generally, let $\{\hat{a}_\nu\}_{\nu\in\mathbb{N}}$ be a maximal family of pairwise orthogonal modes and, for $n \in \mathbb{N}$, denote by $\mathcal{H}_{\hat{a}_1,\ldots,\hat{a}_n}$ the smallest closed subspace of $\mathcal{H}_{\text{field}}$ that contains $\Omega$ and is invariant under $\hat{a}_1^\dagger,\ldots,\hat{a}_n^\dagger$. Then

$$\text{s-}\lim_{n\to\infty}\hat{P}_{\mathcal{H}_{\hat{a}_1,\ldots,\hat{a}_n}} = \hat{1}_{\mathcal{H}_{\text{field}}}\,. \tag{1.90}$$

Now also the vectors

$$\Phi_{\alpha_1,\ldots,\alpha_n} \stackrel{\text{def}}{=} e^{-\frac{1}{2}\left(|\alpha_1|^2+\ldots|\alpha_n|^2\right)}\,e^{\alpha_1\hat{a}_1^\dagger+\ldots\alpha_n\hat{a}_n^\dagger}\,\Omega \quad \forall\, n\in\mathbb{N}\,,\ \alpha_1,\ldots,\alpha_n\in\mathbb{C} \tag{1.91}$$

describe coherent states and have the inner products

$$\langle\Phi_{\alpha_1,\ldots,\alpha_n} \mid \Phi_{\beta_1,\ldots,\beta_m}\rangle = \prod_{\nu=1}^{\min\{n,m\}} e^{-\frac{1}{2}\left(|\alpha_\nu|^2+|\beta_\nu|^2\right)+\alpha_\nu^*\beta_\nu}\,. \tag{1.92}$$

**Outline of proof for $n = m$ :**

$$\begin{aligned}&e^{+\left(|\alpha_1|^2+\ldots+|\beta_n|^2\right)}\,\langle\Phi_{\alpha_1,\ldots,\alpha_n} \mid \Phi_{\beta_1,\ldots,\beta_n}\rangle\\ =\ &\langle\Omega \mid e^{\alpha_1^*\hat{a}_1}\,e^{\beta_1\hat{a}_1^\dagger}\ldots\underbrace{e^{\alpha_n^*\hat{a}_n}\,e^{\beta_n\hat{a}_n^\dagger}\,\Omega}_{=e^{\alpha_n^*\beta_n}\,e^{\alpha_n\hat{a}_n^\dagger}\,\Omega}\rangle\\ =\ &e^{\alpha_n^*\beta_n}\langle\underbrace{e^{\alpha_n^*\hat{a}_n}\,\Omega}_{=\Omega} \mid e^{\alpha_1^*\hat{a}_1}\,e^{\beta_1\hat{a}_1^\dagger}\ldots e^{\alpha_{n-1}^*\hat{a}_{n-1}}\,e^{\beta_{n-1}\hat{a}_{n-1}^\dagger}\,\Omega\rangle\\ &\vdots\\ =\ &\prod_{\nu=1}^{n} e^{\alpha_\nu^*\beta_\nu}\,. \qquad \blacksquare\end{aligned}$$

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[44] A simple calculation (see (Mandel and Wolf, 1995, Section 11.6.1)) even shows that

$$\int f(x^2+y^2)\,(x+iy)^n\left|\hat{D}_{\hat{a}}(x+iy)\,\Omega\right\rangle\mathrm{d}x\,\mathrm{d}y = 0$$

holds for every function $f$ and every $n \in \mathbb{N}$ for which the integral exists.

(1.92) implies

$$\boxed{\left|\left\langle\Phi_{\alpha_1,\ldots,\alpha_{n'}} \mid \Phi_{\beta_1,\ldots,\beta_n}\right\rangle\right|^2 = \prod_{\nu=1}^{n} e^{-|\alpha_\nu-\beta_\nu|^2} \quad \text{for } n' \ge n\,.} \tag{1.93}$$

The corresponding generalization of (1.88) is

$$\pi^{-n}\int |\Phi_{\alpha_1,\ldots,\alpha_n}\rangle\langle\Phi_{\alpha_1,\ldots,\alpha_n}|\,\mathrm{d}^2\alpha_1\cdots\mathrm{d}^2\alpha_n = \hat{P}_{\mathcal{H}_{\hat{a}_1,\ldots,\hat{a}_n}}\,, \tag{1.94}$$

where

$$\mathrm{d}^2(x+iy) \stackrel{\text{def}}{=} \mathrm{d}x\,\mathrm{d}y$$

and this gives a nice mode expansion:

$$\boxed{(1.90) \quad\Longrightarrow\quad \hat{1}_{\mathcal{H}_{\text{field}}} = \text{s-}\lim_{n\to\infty}\int |\Phi_{\alpha_1,\ldots,\alpha_n}\rangle\langle\Phi_{\alpha_1,\ldots,\alpha_n}|\,\frac{\mathrm{d}^2\alpha_1}{\pi}\cdots\frac{\mathrm{d}^2\alpha_n}{\pi}\,.} \tag{1.95}$$

This expansion is extremely useful since[45]

$$\boxed{\hat{a}_j(\mathbf{k})\,\Phi_{\alpha_1,\ldots,\alpha_n} = \left[\hat{a}_j(\mathbf{k})\,,\,\alpha_1\hat{a}_1^\dagger + \ldots \alpha_n\hat{a}_n^\dagger\right]_-\,\Phi_{\alpha_1,\ldots,\alpha_n}\,.} \tag{1.96}$$

Now let $\hat{\rho}$ be some density operator on $\mathcal{H}_{\hat{a}_1,\ldots,\hat{a}_n}$. Then, thanks to (1.95), $\hat{\rho}$ is uniquely determined by the matrix elements

$$f(\alpha_1,\ldots,\beta_n) \stackrel{\text{def}}{=} \langle\Phi_{\alpha_1,\ldots,\alpha_n} \mid \hat{\rho}\,\Phi_{\beta_1,\ldots,\beta_n}\rangle\;, \quad n\in\mathbb{N}\,,\; \alpha_1,\ldots,\beta_n\in\mathbb{C}\,.$$

Since, by (1.91) ,

$$P(\alpha_1,\ldots,\alpha_n,\beta_1,\ldots,\beta_n) \stackrel{\text{def}}{=} e^{\frac{1}{2}\sum_{\nu=1}^n\left(|\alpha_\nu|^2+|\beta_\nu|^2\right)} f(\overline{\alpha_1},\ldots,\overline{\alpha_n},\beta_1,\ldots,\beta_n)$$

is a convergent power series of the variables $\alpha_1,\ldots,\alpha_n,\beta_1,\ldots,\beta_n$ the following lemma shows that $\hat{\rho}$ is already determined by the expectation values[46]

$$\langle\Phi_{\alpha_1,\ldots,\alpha_n} \mid \hat{\rho}\,\Phi_{\alpha_1,\ldots,\alpha_n}\rangle\;, \quad n\in\mathbb{N}\,,\; \alpha_1,\ldots,\alpha_n\in\mathbb{C}\,.$$

**Lemma 1.2.1** *Let $n\in\mathbb{N}$ and let $P(z_1,\ldots,z_{2n})$ be a power series of the complex variables $z_1,\ldots,z_{2n}$ that converges for all $(z_1,\ldots,z_{2n})\in\mathbb{C}^{2n}$ and vanishes if*

$$z_\nu = \overline{z_{n+\nu}} \quad \forall\,\nu\in\{1,\ldots,n\}\;.$$

*Then $P$ is identically zero.*

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[45] Recall (1.82) and (1.37).

[46] Note that $\Phi_{\alpha_1,\ldots,\alpha_n} = \Phi_{\alpha_1,\ldots,\alpha_n,0,\ldots,0}$.

**Outline of proof:** Obviously,

$$\check{P}(z_1, \ldots, z_{2n}) \stackrel{\text{def}}{=} P(z_1 - i\, z_{n+1}, \ldots, z_n - i\, z_{n+n}, z_1 + i\, z_{n+1}, \ldots, z_n + i\, z_{n+n})$$

is a power series of the complex variables $z_1, \ldots, z_{2n}$ that vanishes on $\mathbb{R}^{2n}$. Therefore, it vanishes identically. ∎

This suggests that $\hat{\rho}$ may be represented in the form

$$\hat{\rho} = \text{s-}\lim_{n\to\infty} \int \rho_n(\alpha_1, \ldots, \alpha_n)\, |\Phi_{\alpha_1,\ldots,\alpha_n}\rangle\langle\Phi_{\alpha_1,\ldots,\alpha_n}|\, \frac{\mathrm{d}^2\alpha_1}{\pi} \cdots \frac{\mathrm{d}^2\alpha_n}{\pi}$$

with suitably chosen functions $\rho_n(\alpha_1, \ldots, \alpha_n)$. Indeed, careful elaboration on this idea leads to the following theorem:

**Theorem 1.2.2** *For every density operator $\hat{\rho}$ on $\mathcal{H}_{\text{field}}$ there is a sequence of functions $\varphi_N \in \mathcal{S}\left(\mathbb{R}^{2N}\right)$ — see, e.g. Definition 9.1.1 of (Lücke, eine) for the Definition of the* SCHWARTZ *space $\mathcal{S}$ — such that*

$$\operatorname{Tr}\left(\hat{\rho}\, \hat{B}\right) = \lim_{N\to\infty} \int \varphi_N\Big(\Re(\alpha_1), \Im(\alpha_1), \ldots, \Re(\alpha_N), \Im(\alpha_N)\Big) \left\langle \Phi_{\alpha_1,\ldots,\alpha_N} \mid \hat{B}\, \Phi_{\alpha_1,\ldots,\alpha_N} \right\rangle \frac{\mathrm{d}\alpha_1}{\pi} \cdots \frac{\mathrm{d}\alpha_N}{\pi} \tag{1.97}$$

*is uniformly convergent for all $\hat{B} \in \mathcal{L}\left(\mathcal{H}_{\text{field}}\right)$ with $\left\|\hat{B}\right\| \leq 1$.*

**Proof:** See (Klauder and Sudarshan, 1968, Section 8-4 B). ∎

Strictly speaking (1.97) is established only for bounded operators $\hat{B}$. Typically, however, this formula will be applied to experimentally realizable $\rho$ with normally ordered polynomials (eventually of infinite degree) of radiation field operators substituted for $\hat{B}$. The assertion that such applications are justified is called the ***optical equivalence theorem*** since for **coherent** states — thanks to (1.96) — the expectation values of normal ordered products of (positive- and negative-frequency) radiation field operators coincide with the corresponding products of expectation values of these operators.[47]



[47] The latter implies the optical equivalence theorem for normally ordered bounded analytic functions of the $\hat{a}_\nu$. Limits of such functions are used in typical applications.

# Chapter 2

# Linear Optical Media

## 2.1 General Considerations

In situations of practical interest a given medium never extends over all of $\mathbb{R}$. Consequently, nontrivial boundary conditions have to be taken into account and this spoils the use of spatial FOURIER techniques. Then, instead of a CAUCHY problem, one usually considers a scattering problem: For $t \to \infty$ the electromagnetic field tends to a prescribed vacuum solution. This together with (1.7)–(1.10) and suitable constitutive equations determines the field for all times.

Usually, since the evolution of electromagnetic waves inside materials is very delicate, only very special types of (idealized) media and/or waveforms are discussed in the literature. This makes it very difficult to get an overview.

In their most general form the constitutive equations for ***linear media***[1] just specify $\boldsymbol{\mathcal{P}}(\mathbf{x}, t)$ as a **linear** functional of $\mathbf{E}(\mathbf{x}, t)$ (recall Section 3.1.1) and $\mathbf{B}(\mathbf{x}, t)$. Of course, we are not able to establish linear optics in this general form. Rather we will consider simple models for special situations. However, in order to give at least some feeling for complications arising in more general situations, we derive FRESNEL's formulas ßindexFresnel@FRESNEL's formulas for (isotropic) dispersive media with absorption (on both sides of the boundary plane) and present the solutions of exponential type for MAXWELL's equations in nonisotropic media with absorption and dispersion.

### 2.1.1 A Simple Model

Let us consider (non-moving) ***linear*** optical media inside which the generalized polarization $\boldsymbol{\mathcal{P}}(\mathbf{x}, t)$, and the induced macroscopic current density $\boldsymbol{\jmath}_{\text{ind}}(\mathbf{x}, t)$ are given via $t$-dependent tensor fields[2]

$$\overset{\leftrightarrow}{\check{\chi}}(\mathbf{x}, t) = \left(\check{\chi}^j{}_k(\mathbf{x}, t)\right) \quad \text{and} \quad \overset{\leftrightarrow}{\check{\sigma}}(\mathbf{x}, t) = \left(\check{\sigma}^j{}_k(\mathbf{x}, t)\right)$$

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[1]Linearity should be a good approximation for 'normal' electromagnetic fields.

[2]We use matrix multiplication: $\overset{\leftrightarrow}{m}\,\mathbf{A} \overset{\text{def}}{=} \sum_{j,k=1}^{3} m^j{}_k\, A^k\, \mathbf{e}_j$.

by[3]

$$\boxed{\boldsymbol{\mathcal{P}}(\mathbf{x},t) = \frac{\epsilon_0}{\sqrt{2\pi}} \int \overset{\leftrightarrow}{\check{\chi}}(\mathbf{x},t-t')\,\mathbf{E}(\mathbf{x},t')\,dt'\,,} \tag{2.1}$$

$$\boxed{\boldsymbol{\jmath}_{\mathrm{ind}}(\mathbf{x},t) = \frac{1}{\sqrt{2\pi}} \int \overset{\leftrightarrow}{\check{\sigma}}(\mathbf{x},t-t')\,\mathbf{E}(\mathbf{x},t')\,dt'} \tag{2.2}$$

(see Appendix A of (Lücke, ein) for an outline of tensor calculus). Thinking of $\boldsymbol{\mathcal{P}}$ and $\boldsymbol{\jmath}_{\mathrm{ind}}$ as (initially[4]) induced by $\mathbf{E}$ leads to the following ***causality condition***:[5]

$$\boxed{\overset{\leftrightarrow}{\check{\chi}}(\mathbf{x},t) = \overset{\leftrightarrow}{\check{\sigma}}(\mathbf{x},t) = 0 \quad \forall\, t < 0\,.} \tag{2.3}$$

Moreover, the inverse Fourier transforms $\chi^j{}_k(\mathbf{x},\omega)$ , and $\sigma^j{}_k(\mathbf{x},\omega)$ — usually called "material constants" — should be ordinary functions, converging rapidly to 0 for $\omega \to \pm\infty$. Then also $\check{\chi}^j{}_k(\mathbf{x},t)$ and $\check{\sigma}^j{}_k(\mathbf{x},t)$ are ordinary functions which should decrease exponentially for $t \to +\infty$ (damping).

**Exercise 1** Show that the oscillation $x(t)$, induced by $f(t)$ according to

$$\ddot{x}(t) + 2\rho\,\dot{x}(t) + \omega_0^2\,x(t) = f(t)\,,$$
$$x(t) = f(t) = 0 \quad \text{für } t < t_0\,,$$

is of the form

$$x(t) = \frac{1}{\sqrt{2\pi}} \int \check{r}(t-t')\,f(t')\,dt'$$

where

$$r(\omega) = \frac{1}{\omega_0^2 - \rho^2 - (\omega + i\rho)^2}$$

can be analytically continued into an open neighborhood of the closed upper half plane. Moreover, show that

$$\begin{aligned}\check{r}(t) &= \frac{1}{\sqrt{2\pi}} \int r(-\omega)\,e^{i\omega t}\,d\omega \\ &= \frac{1}{\sqrt{\omega_0^2-\rho^2}}\,e^{-\rho t}\,\sin\left(t\,\sqrt{\omega_0^2-\rho^2}\right)\,\theta(t)\end{aligned}$$

holds for weak damping, i.e. for $0 < \rho \ll \omega_0$.



[3]This means — among other things — that we exclude ferroelectrics, ferromagnets, and nonlocal effects such as optical rotation in quartz (*optical activity*, see (Saleh and Teich, 1991, Eq. (6.4-2))) or anomalous skin effect, here. Media with (spatially) nonlocal response are discussed in (Agranovich and Ginzburg, 1984).

[4]Concerning the backreaction of $\boldsymbol{\mathcal{P}}$ onto $\mathbf{E}$ see, e.g., (Mandel and Wolf, 1995, Sect. 16.3).

[5]Exact vanishing for $t < 0$ is not essential but quite convenient and reasonable for macroscopic considerations.

### 2.1.2 Exploiting Homogeneity

Let us assume that the medium is ***homogeneous***, i.e. that

$$\boxed{\overset{\leftrightarrow}{\check{\chi}}(\mathbf{x},t) = \overset{\leftrightarrow}{\check{\chi}}(t)\,, \quad \overset{\leftrightarrow}{\check{\sigma}}(\mathbf{x},t) = \overset{\leftrightarrow}{\check{\sigma}}(t)\,.}$$

Then the causality condition (2.3) implies that

$$\begin{aligned}
\chi^k{}_l(z) &\stackrel{\text{def}}{=} \frac{1}{\sqrt{2\pi}} \int \check{\chi}^k{}_l(t)\, e^{izt}\, dt \quad (\text{generalized } \textit{\textbf{susceptibility}}) \\
\sigma^k{}_l(z) &\stackrel{\text{def}}{=} \frac{1}{\sqrt{2\pi}} \int \check{\sigma}^k{}_l(t)\, e^{izt}\, dt \quad (\textit{\textbf{specific conductivity}})
\end{aligned}$$

are bounded holomorphic functions on a sufficiently small neighborhood of the closed upper half plane.

> **Remark:** This together with (2.4), (2.5), and the causality condition allows for the derivation of dispersion relations (see Sect. 4.1 of (Lücke, ftm) providing valuable insight into the frequency dependence of the permittivity (see (Mills, 1998, Sect. 2.1)).

The same, consequently, holds for the (relative) ***permittivity***[6]

$$\overset{\leftrightarrow}{\epsilon}(z) \stackrel{\text{def}}{=} \overset{\leftrightarrow}{1} + \overset{\leftrightarrow}{\chi}(z)\,.$$

Since $\check{\chi}^k{}_l$ and $\check{\sigma}^k{}_l$ have to be real-valued, the so-called ***crossing relations***

$$\boxed{\left.\begin{array}{rcl} \overset{\leftrightarrow}{\epsilon}(z) &=& \overset{\leftrightarrow}{\epsilon}{}^*(-z^*) \\ \overset{\leftrightarrow}{\sigma}(z) &=& \overset{\leftrightarrow}{\sigma}{}^*(-z^*) \end{array}\right\} \quad \text{for } \Im(z) \geq 0} \tag{2.4}$$

have to be fulfilled[7] according to Rule 7 for the FOURIER transform[8] and the principle of analytic continuation. The asymptotic behaviour along the real axis is:[9]

$$\boxed{\left.\begin{array}{rcl} \overset{\leftrightarrow}{\epsilon}(\omega) &\to& \overset{\leftrightarrow}{1} \\ \overset{\leftrightarrow}{\sigma}(\omega) &\to& \overset{\leftrightarrow}{0} \end{array}\right\} \quad \text{for } \omega \to \pm\infty\,.} \tag{2.5}$$

If (2.1) holds, we may use

$$\epsilon_0\, \widetilde{\mathbf{E}}(\mathbf{x},\omega) + \widetilde{\mathcal{P}}(\mathbf{x},\omega) = \epsilon_0\, \overset{\leftrightarrow}{\sigma}(\omega)\, \widetilde{\mathbf{E}}(\mathbf{x},\omega) \tag{2.6}$$

in (1.18) resp. (1.20) and get

$$\boxed{\operatorname{curl} \widetilde{\mathbf{B}}(\mathbf{x},\omega) = \frac{-i\,\omega}{c^2}\, \overset{\leftrightarrow}{\epsilon}(\omega)\, \widetilde{\mathbf{E}}(\mathbf{x},\omega) + \mu_0\, \widetilde{\jmath}_{\text{ex}}(\mathbf{x},\omega)} \tag{2.7}$$

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[6] The inverse $\overset{\leftrightarrow}{\epsilon}{}^{-1}(z)$ is called ***impermeability***.

[7] By definition, the matrix $\overset{\leftrightarrow}{m}{}^*$ results from $\overset{\leftrightarrow}{m}$ by complex conjugation of the entries.

[8] See Appendix.

[9] Compare Exercise 1 and (Römer, 1994, Sect. 2.8).

resp.

$$\boxed{\operatorname{div}\left(\epsilon_0 \overset{\leftrightarrow}{\sigma}(\omega)\, \widetilde{\mathbf{E}}(\mathbf{x},\omega)\right) = +\widetilde{\rho}_{\mathrm{ex}}(\mathbf{x},\omega)\,,} \tag{2.8}$$

**If** also $\jmath_{\mathrm{ex}} = \jmath_{\mathrm{ind}}$ holds with $\jmath_{\mathrm{ind}}$ given by (2.2), then (2.7) becomes[10]

$$\operatorname{curl} \widetilde{\mathbf{B}}(\mathbf{x},\omega) = \frac{-i\,\omega}{c^2}\left(\overset{\leftrightarrow}{\epsilon}(\omega) + i\,\frac{\overset{\leftrightarrow}{\sigma}(\omega)}{\omega\,\epsilon_0}\right)\widetilde{\mathbf{E}}(\mathbf{x},\omega)\,. \tag{2.9}$$

**Remark:** The material tensors $\overset{\leftrightarrow}{\epsilon}(\omega)$, $\overset{\leftrightarrow}{\sigma}(\omega)$ enter MAXWELL's equations only via the combination

$$\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega) \stackrel{\mathrm{def}}{=} \overset{\leftrightarrow}{\epsilon}(\omega) + i\,\frac{\overset{\leftrightarrow}{\sigma}(\omega)}{\omega\,\epsilon_0}\,,$$

usually called ***complex dielectric 'constant'*** — even though, from the physical point of view,[11] already $\epsilon(\omega)$ should be complex-valued.[12] From the purely mathematical point of view, if only $\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)$ is given, ${\epsilon^k}_l(\omega)$ and ${\sigma^k}_l(\omega)$ may be considered as real-valued (but not separately analytic). Similar remarks apply to the ***complex conductivity***

$$\overset{\leftrightarrow}{\sigma}_{\mathrm{c}}(\omega) \stackrel{\mathrm{def}}{=} \overset{\leftrightarrow}{\sigma}(\omega) - i\,\omega\,\epsilon_0\,\overset{\leftrightarrow}{\epsilon}(\omega) \quad \left(= -i\,\omega\,\epsilon_0\,\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\right)\,.$$

**Exercise 2** Given $\omega \in \mathbb{R}\setminus\{0\}$ and assuming (1.7)–(2.2) show that

$$\left.\begin{array}{l} 0 \neq \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega) \sim \overset{\leftrightarrow}{1} \\ \jmath_{\mathrm{ex}}(\mathbf{x},t) = \jmath_{\mathrm{ind}}(\mathbf{x},t) \end{array}\right\} \Longrightarrow \quad \widetilde{\rho}_{\mathrm{ex}}(\mathbf{x},\omega) = 0\,.$$

Thanks to (1.19), the radiative Part of $\mathbf{B}(\mathbf{x},t)$ as as a function of time is already fixed by $\mathbf{E}(\mathbf{x},t)$ and its first order spatial derivatives **at the same position** in space. Therefore:

> For radiation fields inside homogeneous linear media it is sufficient to determine $\mathbf{E}(\mathbf{x},t)$.

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[10]Since the 4-current $\jmath^\nu_{\mathrm{cr}} \stackrel{\mathrm{def}}{=} \jmath^\nu_{\mathrm{ex}} - \jmath^\nu_{\mathrm{ind}}$ creating the electromagnetic wave (see Section 2.1.3) is not included, (2.9) is — strictly speaking — only relevant for times $t$ with $\jmath^\nu_{\mathrm{cr}}(\mathbf{x},t') = 0 \quad \forall\, t' \geq t$.

[11]Compare Exercise 1.

[12]Thus allowing for a phase difference between $\widetilde{\mathbf{E}}(\mathbf{x},\omega)$ and $\widetilde{\mathbf{D}}(\mathbf{x},\omega)$.

In the following we consider only media for which (2.1) holds. Then we have

$$\begin{aligned}
\operatorname{curl}\left(\operatorname{curl}\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right) &\underset{(1.19)}{=} i\,\omega\operatorname{curl}\widetilde{\mathbf{B}}(\mathbf{x},\omega)\\
&\underset{(2.7)}{=} \frac{\omega^2}{c^2}\overset{\leftrightarrow}{\epsilon}(\omega)\,\widetilde{\mathbf{E}}(\mathbf{x},\omega) + i\,\omega\,\mu_0\,\widetilde{\jmath}_{\mathrm{ex}}(\mathbf{x},\omega)\\
&\underset{(2.2)}{=} \frac{\omega^2}{c^2}\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\,\widetilde{\mathbf{E}}(\mathbf{x},\omega) + i\,\omega\mu_0\,\widetilde{\jmath}_{\mathrm{cr}}(\mathbf{x},\omega)\,,
\end{aligned} \tag{2.10}$$

where

$$\widetilde{\jmath}_{\mathrm{cr}}(\mathbf{x},\omega) \stackrel{\mathrm{def}}{=} \widetilde{\jmath}_{\mathrm{ex}}(\mathbf{x},\omega) - \widetilde{\jmath}_{\mathrm{ind}}(\mathbf{x},\omega)\,. \tag{2.11}$$

Thanks to

$$\boldsymbol{\nabla}_{\mathbf{x}}\times\left(\boldsymbol{\nabla}_{\mathbf{x}}\times\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right) = \boldsymbol{\nabla}_{\mathbf{x}}\left(\boldsymbol{\nabla}_{\mathbf{x}}\cdot\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right) - \Delta_{\mathbf{x}}\,\widetilde{\mathbf{E}}(\mathbf{x},\omega)\,,$$

(2.10) is equivalent to

$$\boxed{\left(\Delta_{\mathbf{x}} + \left(\frac{\omega}{c}\right)^2\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\right)\widetilde{\mathbf{E}}(\mathbf{x},\omega) = \operatorname{grad}\left(\operatorname{div}\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right) - i\,\omega\,\mu_0\,\widetilde{\jmath}_{\mathrm{cr}}(\mathbf{x},\omega)\,.} \tag{2.12}$$

Therefore

$$\widetilde{\mathbf{B}}(\mathbf{x},\omega) \underset{(1.19)}{=} \frac{\operatorname{curl}\widetilde{\mathbf{E}}(\mathbf{x},\omega)}{i\,\omega} \quad \text{for } \omega\neq 0 \tag{2.13}$$

together with (2.12) implies (2.7). (1.21) is a direct consequence of (2.13) and (2.8), finally, serves as a definition for $\widetilde{\rho}_{\mathrm{ex}}$ respecting the continuity equation. The latter implies

$$\begin{aligned}
\widetilde{\rho}_{\mathrm{ex}}(\mathbf{x},\omega) &= \widetilde{\rho}_{\mathrm{cr}}(\mathbf{x},\omega) + \widetilde{\rho}_{\mathrm{ind}}(\mathbf{x},\omega) \quad \text{for } \omega\neq 0\,, &(2.14)\\
\text{where:}\quad \widetilde{\rho}_{\mathrm{cr}}(\mathbf{x},\omega) &\stackrel{\mathrm{def}}{=} \operatorname{div}\left(\widetilde{\jmath}_{\mathrm{cr}}(\mathbf{x},\omega)\right)\Big/(i\omega)\,, &(2.15)\\
\widetilde{\rho}_{\mathrm{ind}}(\mathbf{x},\omega) &\stackrel{\mathrm{def}}{=} \operatorname{div}\left(\overset{\leftrightarrow}{\sigma}(\omega)\,\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right)\Big/(i\omega)\,. &(2.16)
\end{aligned}$$

We conclude:

| |
|---|
| Solving (1.18)–(1.21) for $\omega\neq 0$ is equivalent to solving (2.12) and determining $\widetilde{\mathbf{B}}(\mathbf{x},\omega)$ by (2.13). |

**Lemma 2.1.1** *Let* $\mathbf{E}(\mathbf{x},t)$ *be a solution of (2.12) for* $\widetilde{\jmath}_{\mathrm{cr}}(\mathbf{x},\omega) = 0$ *with*

$$\operatorname{supp}\mathbf{E}(\mathbf{x},t)\subset U_R(V_+)$$

*for some* $R>0$*. Then* $\mathbf{E}(\mathbf{x},t) = 0$*.*

**Proof:** Use the tube theorem, the edge-of-the-wedge theorem, and (2.5). ∎

### 2.1.3 Radiation off a Given Source

In this subsection we consider only ***isotropic***[13] homogeneous media, i.e. we assume

$$\overset{\leftrightarrow}{\check{\chi}}(\mathbf{x},t) = \check{\chi}(t)\,\overset{\leftrightarrow}{1}\,, \quad \overset{\leftrightarrow}{\check{\sigma}}(\mathbf{x},t) = \check{\sigma}(t)\,\overset{\leftrightarrow}{1}\,. \tag{2.17}$$

Then, by (2.8), (2.12) becomes

$$\left(\Delta_{\mathbf{x}} + \left(\frac{\omega}{c}\right)^2 \epsilon_{\mathrm{c}}(\omega)\right)\widetilde{\mathbf{E}}(\mathbf{x},\omega) = \frac{1}{\epsilon_0\,\epsilon(\omega)}\operatorname{grad}\widetilde{\rho}_{\mathrm{ex}}(\mathbf{x},\omega) - i\,\omega\,\mu_0\,\widetilde{\boldsymbol{\jmath}}_{\mathrm{cr}}(\mathbf{x},\omega)\,. \tag{2.18}$$

Defining

$$\begin{aligned}\widetilde{\mathbf{E}}_{\mathrm{ret}}(\mathbf{x},\omega) \overset{\mathrm{def}}{=} & -\operatorname{grad}\int \widetilde{\rho}_{\mathrm{cr}}(\mathbf{x}',\omega)\,\frac{e^{i\frac{\omega}{c}\mathcal{N}(\omega)|\mathbf{x}-\mathbf{x}'|}}{4\pi\,\epsilon_0\,\epsilon_{\mathrm{c}}(\omega)\,|\mathbf{x}-\mathbf{x}'|}\,dV_{\mathbf{x}'} \\ & -\mu_0\int (-i\omega)\,\widetilde{\boldsymbol{\jmath}}_{\mathrm{cr}}(\mathbf{x}',\omega)\,\frac{e^{i\frac{\omega}{c}\mathcal{N}(\omega)|\mathbf{x}-\mathbf{x}'|}}{4\pi\,|\mathbf{x}-\mathbf{x}'|}\,dV_{\mathbf{x}'}\,,\end{aligned} \tag{2.19}$$

where

$$\mathcal{N}(\omega) \overset{\mathrm{def}}{=} \sqrt{\epsilon_{\mathrm{c}}(\omega)}\,, \quad \Re\Big(\mathcal{N}(\omega)\Big) \geq 0\,,$$

and using (2.15) we get

$$\operatorname{div}\widetilde{\mathbf{E}}_{\mathrm{ret}}(\mathbf{x},\omega) = \frac{\widetilde{\rho}_{\mathrm{cr}}(\mathbf{x},\omega)}{\epsilon_0\,\epsilon_{\mathrm{c}}(\omega)} \tag{2.20}$$

and hence, by (2.14) and (2.16),

$$\widetilde{\rho}_{\mathrm{cr}}(\mathbf{x},\omega)/\epsilon_{\mathrm{c}}(\omega) = \widetilde{\rho}_{\mathrm{ex}}(\mathbf{x},\omega)/\epsilon(\omega)\,.$$

Therefore $\widetilde{\mathbf{E}}(\mathbf{x},\omega) = \widetilde{\mathbf{E}}_{\mathrm{ret}}(\mathbf{x},\omega)$ is a solution of (2.18) (for $\omega \neq 0$). By (2.13) this implies

$$\widetilde{\mathbf{B}}_{\mathrm{ret}}(\mathbf{x},\omega) \overset{\mathrm{def}}{=} \operatorname{curl}\int \mu_0\,\widetilde{\boldsymbol{\jmath}}_{\mathrm{cr}}(\mathbf{x}',\omega)\,\frac{e^{i\frac{\omega}{c}\mathcal{N}(\omega)|\mathbf{x}-\mathbf{x}'|}}{4\pi\,|\mathbf{x}-\mathbf{x}'|}\,dV_{\mathbf{x}'}\,. \tag{2.21}$$

Fourier transforming (2.19) and (2.21) we get solutions

$$\mathbf{E}(\mathbf{x},t) = -\operatorname{grad}\Phi(\mathbf{x},t) - \frac{\partial}{\partial t}\mathbf{A}(\mathbf{x},t)\,, \quad \mathbf{B}(\mathbf{x},t) = \operatorname{curl}\mathbf{A}(\mathbf{x},t) \tag{2.22}$$

of Maxwell's equations (1.7)–(1.10) for[14]

$$\begin{aligned}\rho_{\mathrm{ex}}(\mathbf{x},t) &= -\int_{-\infty}^{t}\left(\frac{1}{\sqrt{2\pi}}\int \check{\sigma}(t''-t')\operatorname{div}\mathbf{E}(\mathbf{x},t')\,dt'\right)dt'' + \rho_{\mathrm{cr}}(\mathbf{x},t)\,, \\ \boldsymbol{\jmath}_{\mathrm{ex}}(\mathbf{x},t) &= \frac{1}{\sqrt{2\pi}}\int \check{\sigma}(t-t')\,\mathbf{E}(\mathbf{x},t')\,dt' + \boldsymbol{\jmath}_{\mathrm{cr}}(\mathbf{x},t)\end{aligned} \tag{2.23}$$

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[13]All crystals of the cubic system are isotropic. It would be interesting to generalize the results of this subsection for nonisotropic media.

[14]Note that (2.23) and the continuity equation for $_{\mathrm{ex}}$ imply the continuity equation for $_{\mathrm{cr}}$.

with ***electromagnetic Potentials*** $\Phi$ , $\mathbf{A}$ of the form

$$
\begin{aligned}
\Phi(\mathbf{x},t) &= \Phi_{\rm ret}(\mathbf{x},t) \stackrel{\rm def}{=} \frac{1}{\sqrt{2\pi}} \int \widetilde{\rho}_{\rm cr}(\mathbf{x}',\omega)\, \frac{e^{-i\frac{\omega}{c}(ct-\mathcal{N}(\omega)|\mathbf{x}-\mathbf{x}'|)}}{4\pi\,\epsilon_0\,\epsilon_{\rm c}(\omega)\;|\mathbf{x}-\mathbf{x}'|}\, dV_{\mathbf{x}'}\, d\omega\,, \\
\mathbf{A}(\mathbf{x},t) &= \mathbf{A}_{\rm ret}(\mathbf{x},t) \stackrel{\rm def}{=} \frac{1}{\sqrt{2\pi}} \int \mu_0\, \widetilde{\boldsymbol{\jmath}}_{\rm cr}(\mathbf{x}',\omega)\, \frac{e^{-i\frac{\omega}{c}(ct-\mathcal{N}(\omega)|\mathbf{x}-\mathbf{x}'|)}}{4\pi\;|\mathbf{x}-\mathbf{x}'|}\, dV_{\mathbf{x}'}\, d\omega\,.
\end{aligned}
\tag{2.24}
$$

Using standard techniques one may prove (see (Borchers, 1990) and, for the main techniques, also Sect. 4.1 of (Lücke, ftm)) that

$$
\begin{aligned}
&\rho_{\rm cr}(\mathbf{x},t)\,,\ \boldsymbol{\jmath}_{\rm cr}(\mathbf{x},t) = 0 \quad \text{for} \quad (|\mathbf{x}|\,,t) \notin [0,R]\times[0,\infty] \\
&\Longrightarrow \quad \mathbf{B}_{\rm ret}(\mathbf{x},t) = 0 \quad \text{for } t \notin \left[0, \tfrac{|\mathbf{x}|-R}{c}\right].
\end{aligned}
$$

and, assuming[15]

$$
\int \frac{1}{\epsilon_{\rm c}(\omega)}\, e^{-i\omega t}\, dt = 0 \quad \forall\, t<0\,, \tag{2.25}
$$

(in addition to (2.3)) also

$$
\begin{aligned}
&\rho_{\rm cr}(\mathbf{x},t)\,,\ \boldsymbol{\jmath}_{\rm cr}(\mathbf{x},t) = 0 \quad \text{for} \quad (|\mathbf{x}|\,,t) \notin [0,R]\times[0,\infty] \\
&\Longrightarrow \quad \mathbf{E}_{\rm ret}(\mathbf{x},t) = 0 \quad \text{for } t \notin \left[0, \tfrac{|\mathbf{x}|-R}{c}\right].
\end{aligned}
\tag{2.26}
$$

holds. In other words:

The speed of a lightning inside the medium is strictly bounded by $c$ .

All this becomes obvious for constant[16] real $\epsilon_{\rm c}$ . Then we have

$$
\Phi_{\rm ret}(\mathbf{x},t) = \frac{\rho_{\rm cr}\left(\mathbf{x}', t-\frac{n}{c}\,|\mathbf{x}-\mathbf{x}'|\right)}{4\pi\,\epsilon_0\,\epsilon_{\rm c}\,|\mathbf{x}-\mathbf{x}'|}\ , \qquad \mathbf{A}_{\rm ret}(\mathbf{x},t) = \mu_0\, \frac{\boldsymbol{\jmath}_{\rm cr}\left(\mathbf{x}', t-\frac{n}{c}\,|\mathbf{x}-\mathbf{x}'|\right)}{4\pi\;|\mathbf{x}-\mathbf{x}'|}
$$

with $n = \mathcal{N}(\omega)$ , showing that the speed of light in such a medium is $c_{\rm medium} = c/n$ . Taken together with the former argument we see that

$$
\mathbb{R} \ni \epsilon_{\rm c} \text{ constant} \quad \Longrightarrow \quad n \geq 1\,. \tag{2.27}
$$

**Remarks:**

1. By Lemma 2.1.1, there is at most one retarded solution for given $\rho_{\rm cr}$ , $\boldsymbol{\jmath}_{\rm cr}$ .
2. Therefore we consider the solution (2.22)/(2.24) as the physical one.
3. Then (2.20) implies
$$
\operatorname{div} \widetilde{\mathbf{E}}(\mathbf{x},\omega) = 0 \tag{2.28}
$$
outside the (spatial) support of $\rho_{\rm cr}(\mathbf{x},\omega)$ .

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[15] This assumption may presumably be proved using (2.5).

[16] Strictly speaking, this is only compatible with (2.5) if $\epsilon = \mu = 1$ and $\sigma = 0$ .

## 2.2 Monochromatic Waves of Exponential Type

### 2.2.1 Basic Equations

In the following we consider only the case

$$\boxed{\mathcal{J}_{\mathrm{cr}} = 0\,.}$$

Then (2.12) becomes

$$\left(\Delta_{\mathbf{x}} + \left(\frac{\omega}{c}\right)^2 \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\right) \widetilde{\mathbf{E}}(\mathbf{x},\omega) = \operatorname{grad}\left(\operatorname{div}\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right) \tag{2.29}$$

Let us look for monochromatic solutions of exponential type

$$\boxed{\widetilde{\mathbf{E}}(\mathbf{x},\omega') = \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\, \exp\left(i\,\frac{\omega}{c}\,\mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s}\cdot\mathbf{x}\right)\, \sqrt{2\pi}\,\delta(\omega'-\omega)\,,} \tag{2.30}$$

i.e. for (damped) ***monochromatic plane waves***[17]

$$\mathbf{E}(\mathbf{x},t) = \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\, e^{-i\,\frac{\omega}{c}\left(c\,t - \mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s}\cdot\mathbf{x}\right)}\,,$$

where

$$\mathbf{s}\,,\ \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) \in \mathbb{C}^3$$

and $\mathbf{s}$ is a (complex) ***direction***, i.e. [18]

$$\boxed{\mathbf{s}\cdot\mathbf{s} = 1\,.} \tag{2.31}$$

Then (2.29) holds iff[19]

$$\boxed{\begin{array}{c} \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\,\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) = \left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2 \overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\,, \\ \text{where:}\quad \overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\mathbf{z} \overset{\text{def}}{=} \mathbf{z} - (\mathbf{s}\cdot\mathbf{z})\,\mathbf{s} \quad \forall\,\mathbf{z}\in\mathbb{C}^3\,. \end{array}} \tag{2.32}$$



[17]Note that, by (2.31), $\mathcal{N}(\omega)\,\mathbf{s}$ can only be real if $\left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2$ is real. One may show (see (Mills, 1998, p. 12)) that $\Re\left(\mathcal{N}_{\mathbf{s}}(\omega)\right) > 0 \implies \omega\,\Im\left(\mathcal{N}_{\mathbf{s}}(\omega)\right) \le 0\,.$ Actually, $\mathcal{N}_{\mathbf{s}}(\omega)$ may depend on the polarization (see 2.2.3).

[18]We use standard notation:

$$\mathbf{a}\cdot\mathbf{b} \overset{\text{def}}{=} \sum_{j=1}^{3} a^j\,b^j\,,\quad \overline{a^1\mathbf{e}_1 + a^2\mathbf{e}_2 + a^3\mathbf{e}_3} \overset{\text{def}}{=} \overline{a^1}\mathbf{e}_1 + \overline{a^2}\mathbf{e}_2 + \overline{a^3}\mathbf{e}_3\,,\quad |\mathbf{a}| \overset{\text{def}}{=} \sqrt{\overline{\mathbf{a}}\cdot\mathbf{a}} \ge 0$$

$$(a^1\mathbf{e}_1 + a^2\mathbf{e}_2 + a^3\mathbf{e}_3)\times(b^1\mathbf{e}_1 + b^2\mathbf{e}_2 + b^3\mathbf{e}_3) \overset{\text{def}}{=} (a^2b^3 - a^3b^2)\,\mathbf{e}_1 + (a^3b^1 - a^1b^3)\,\mathbf{e}_2 + (a^1b^2 - a^2b^1)\,\mathbf{e}_3\,.$$

Note that the symmetric bilinear form $\mathbf{a}\cdot\mathbf{b}$ is an **indefinite** inner product on $\mathbb{C}^3$ and that $\Im\left(\mathbf{s}\cdot\mathbf{s}\right) = 0 \implies \Re(\mathbf{s})\cdot\Im(\mathbf{s}) = 0\,.$ Modes with $0 \ne \mathbf{e}\cdot\mathbf{s} \in i\,\mathbb{R}$ for some $\mathbf{e}\in\mathbb{R}^3$ are called ***evanescent***.

[19](2.32) implies (2.8) for $\rho_{\mathrm{ex}} = \rho_{\mathrm{ind}}$ according to (2.16). Note that, thanks to (2.31), $\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}$ is a projection operator: $\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\overset{\leftrightarrow}{P}_{\perp\mathbf{s}} = \overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,.$

According to (2.13), the corresponding magnetic field is given by

$$\boxed{\widetilde{\mathbf{B}}(\mathbf{x},\omega) = \mu_0\,\frac{\mathcal{N}_{\mathbf{s}}(\omega)}{Z_0}\,\mathbf{s}\times\widetilde{\mathbf{E}}(\mathbf{x},\omega)} \tag{2.33}$$

for $\omega \neq 0$, where

$$Z_0 \stackrel{\text{def}}{=} \mu_0\,c \approx 377\,\Omega \quad \textbf{\textit{vacuum impedance}}\,.$$

Even though the object of physical interest is the real field[20]

$$\begin{aligned}\mathbf{E}(\mathbf{x},t)+\overline{\mathbf{E}(\mathbf{x},t)} \;=\; & 2\,e^{-\frac{\omega}{c}\Im\left(\mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s}\right)\cdot\mathbf{x}}\,\Big(\Re\left(\boldsymbol{\mathcal{E}}_{\mathbf{s}}\right)\,\cos\Big(\omega\,t-\frac{\omega}{c}\,\Re(\mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s})\cdot\mathbf{x}\Big)\\ & +\Im\left(\boldsymbol{\mathcal{E}}_{\mathbf{s}}\right)\,\sin\Big(\omega\,t-\frac{\omega}{c}\,\Re(\mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s})\cdot\mathbf{x}\Big)\Big)\end{aligned} \tag{2.34}$$

it is mathematically quite convenient to work with the complex field[21] $\mathbf{E}(\mathbf{x},t)$. For the corresponding POYNTING vector $\mathbf{S}(\mathbf{x},t)$ we have

$$\begin{aligned}\mu_0\,\langle\mathbf{S}(\mathbf{x},t)\rangle \;&=\; 4\,\Big\langle\Re\Big(\mathbf{E}(\mathbf{x},t)\Big)\times\Re\Big(\mathbf{B}(\mathbf{x},t)\Big)\Big\rangle\\ &\underset{(2.33)}{=}\; \mu_0\,\frac{2}{Z_0}\,e^{-2\,\frac{\omega}{c}\,\Im\left(\mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s}\right)\cdot\mathbf{x}}\,\Re\Big(\overline{\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)}\times\left(\mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s}\times\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\right)\Big)\end{aligned} \tag{2.35}$$

where $\langle\;\rangle$ means averaging over $t$. By (2.31) and

$$\mathbf{a}\times(\mathbf{b}\times\mathbf{c}) = (\mathbf{a}\cdot\mathbf{c})\,\mathbf{b}-(\mathbf{a}\cdot\mathbf{b})\,\mathbf{c} \quad \forall\,\mathbf{a},\mathbf{b},\mathbf{c}\in\mathbb{C}^3 \tag{2.36}$$

this gives[22]

$$\boxed{\begin{aligned}\langle\mathbf{S}(\mathbf{x},t)\rangle \;=\; & \frac{2}{Z_0}\,e^{-2\,\frac{\omega}{c}\,\Im\left(\mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s}\right)\cdot\mathbf{x}}\,\Re\Big(\left|\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\right|^2\,\left(\mathcal{N}_{\mathbf{s}}(\omega)\,\mathbf{s}\right)\\ & -\mathcal{N}_{\mathbf{s}}(\omega)\,\left(\overline{\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)}\cdot\mathbf{s}\right)\,\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\Big)\,.\end{aligned}} \tag{2.37}$$

For real $\mathbf{s}$ and $\overset{\leftrightarrow}{\epsilon}(\omega) = \epsilon(\omega)\,\overset{\leftrightarrow}{1}$ the ***polarization*** of the wave is given, according to



[20]Note that with (1.24) the crossing relations (2.4) imply that (2.29) also holds for $\omega$ replaced by $-\omega$. Similar formulas hold for $\mathbf{B}(\mathbf{x},t)$ and $\widetilde{\mathbf{B}}(\mathbf{x},\omega)$ with $\boldsymbol{\mathcal{B}}_{\mathbf{s}} = \mu_0\frac{\mathcal{N}(\omega)}{Z_0}\,\mathbf{s}\times\boldsymbol{\mathcal{E}}_{\mathbf{s}}$.

[21]See (Bloembergen, 1996b, Section 1–2) for the justification of this convention.

[22]Note that for isotropic media and real $\mathbf{s}$ the wave is propagating into the direction of $\Re\Big(\mathcal{N}_{\mathbf{s}}(\omega)\Big)\mathbf{s}$ perpendicular to the planes of constant phase while the planes of constant amplitude are perpendicular to $\Im\big(\mathcal{N}_{\mathbf{s}}(\omega)\big)\mathbf{s}$.

| polarization | JONES vector |
|---|---|
| linear | $e^{i\varphi}\begin{pmatrix}\cos\alpha\\ \sin\alpha\end{pmatrix}$ |
| right circular[24] | $\frac{e^{i\varphi}}{\sqrt{2}}\begin{pmatrix}1\\ i\end{pmatrix}$ |
| left circular | $\frac{e^{i\varphi}}{\sqrt{2}}\begin{pmatrix}1\\ -i\end{pmatrix}$ |
| elliptic | else[25] |

Table 2.1: Polarization of monochromatic plane waves

Table 2.1, by the corresponding JONES ***vector***[23]

$$\mathbf{J_s}(\mathbf{x},\omega) \stackrel{\text{def}}{=} \begin{pmatrix} \mathbf{a_s}\cdot\dfrac{\widetilde{\mathbf{E}}(\mathbf{x},\omega)}{\left|\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right|} \\ \mathbf{b_s}\cdot\dfrac{\widetilde{\mathbf{E}}(\mathbf{x},\omega)}{\left|\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right|}\end{pmatrix},$$

relative to a right-handed orthonormal basis $(\mathbf{a_s},\mathbf{b_s},\mathbf{s})$ of $\mathbb{R}^3$.

### 2.2.2 Simple Reflection and Refraction

For isotropic media, i.e. if (2.17) holds, the solution of (2.32) is obvious:

$$\mathbf{s}\cdot\boldsymbol{\mathcal{E}}_\mathbf{s}(\omega)=0\,,\quad \pm\mathcal{N}_\mathbf{s}(\omega)=\mathcal{N}(\omega)\stackrel{\text{def}}{=}\sqrt{\epsilon_c(\omega)}\,,\quad \Re\Big(\mathcal{N}(\omega)\Big)\geq 0\,.$$

Let us now consider the following situation:[26]

> The region $\mathcal{G}_\pm \stackrel{\text{def}}{=} \{\mathbf{x}\in\mathbb{R}^3 : \pm x^1 \geq 0\}$ is filled by some homogeneous **isotropic** linear optical medium with (generalized) permittivity $\overleftrightarrow{\epsilon}_\pm(\omega) =$



[23]See (Jones, 1941; Azzam and Bashara, 1987; Kliger et al., 1990). For actual measurement of polarization see (Gjurchinovski, 2002). .

[24]Here — contrary to the convention used, e.g., in (Reider, 1997, Sect. 1.5) — *right circular* means that the $\mathbf{b_s}$ component of $\mathbf{E}(\mathbf{x},t)$ lags the $\mathbf{a_s}$ component of $\mathbf{E}(\mathbf{x},t)$ by $90°$. This way — as pointed out in (Jackson, 1975, Sect. 7.2) — right circular light corresponds to *positive helicity*, hence to *right handed* photons.

[25]Note that, for $\omega\neq 0$: $\dfrac{\mathrm{d}}{\mathrm{d}t}\left|\Re\left(\dfrac{e^{-i\omega t}}{\sqrt{2}}\begin{pmatrix}1\\ z\end{pmatrix}\right)\right|^2 = 0\;\forall\, t \iff z\in\{-i,+i\}\,.$

[26]See also (Stratton, 1941, Ch. IX) for the special case that $\mathcal{G}_-$ is a perfect dielectric. For realizable refractive indices see (Skaar and Seip, 2006; Grigorenko, 2006). For generalization to curved boundaries see (Hentschel and Schomerus, 2002; Hentschel and Schomerus, 2006).

$\epsilon_\pm(\omega)\overset{\leftrightarrow}{1}$, and conductivity $\overset{\leftrightarrow}{\sigma}_\pm(\omega) = \sigma_\pm(\omega)\overset{\leftrightarrow}{1}$. Moreover, assume

$$\Big(\mathcal{N}_+(\omega)\Big)^2 \neq \Big(\mathcal{N}_-(\omega)\Big)^2, \tag{2.38}$$

where

$$\mathcal{N}_\pm(\omega) \overset{\text{def}}{=} \sqrt{\epsilon_{\text{c}}^\pm(\omega)}\,, \quad \Re\Big(\mathcal{N}_\pm(\omega)\Big) \geq 0\,.$$

Then scattering theory suggests to look for modes of the following form:[27]

- Inside $\mathcal{G}_-$ the electric field has the form

$$\mathbf{E}_-(\mathbf{x},t) = \boldsymbol{\mathcal{E}}_{\mathbf{s}_-}\, e^{-i\frac{\omega}{c}\left(c\,t - \mathcal{N}_-(\omega)\,\mathbf{s}_-\cdot\mathbf{x}\right)} + \boldsymbol{\mathcal{E}}_{\mathbf{s}'_-}\, e^{-i\frac{\omega}{c}\left(c\,t - \mathcal{N}_-(\omega)\,\mathbf{s}'_-\cdot\mathbf{x}\right)}\,.$$

- The electric field inside $\mathcal{G}_+$ has the form

$$\mathbf{E}_+(\mathbf{x},t) = \boldsymbol{\mathcal{E}}_{\mathbf{s}_+}\, e^{-i\frac{\omega}{c}\left(c\,t - \mathcal{N}_+(\omega)\,\mathbf{s}_+\cdot\mathbf{x}\right)}\,.$$

- $\mathbf{s}_-, \mathbf{s}'_-, \mathbf{s}_+, \boldsymbol{\mathcal{E}}_{\mathbf{s}_-}, \boldsymbol{\mathcal{E}}_{\mathbf{s}'_-}, \boldsymbol{\mathcal{E}}_{\mathbf{s}_+} \in \mathbb{C}^3 \setminus \{0\}$ .
- (2.31) holds for $\mathbf{s} \in \{\mathbf{s}_-, \mathbf{s}'_-, \mathbf{s}_+\}$ .
- $\mathbf{s}_- - \hat{P}_{\mathbf{e}_1}\mathbf{s}_-$ is not isotropic[28] and $\hat{P}_{\mathbf{e}_1}\mathbf{s}_- \overset{\text{def}}{=} (\mathbf{e}_1\cdot\mathbf{s}_-)\,\mathbf{e}_1 \neq 0$.

For this Ansatz the boundary condition for the elctric field requires

$$\mathbf{e}_1 \times \left(\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}\, e^{-i\,\frac{\omega}{c}\mathcal{N}_-(\omega)\,\mathbf{s}_-\cdot\mathbf{x}} + \boldsymbol{\mathcal{E}}_{\mathbf{s}'_-}\, e^{-i\,\frac{\omega}{c}\mathcal{N}_-(\omega)\,\mathbf{s}'_-\cdot\mathbf{x}} - \boldsymbol{\mathcal{E}}_{\mathbf{s}_+}\, e^{-i\frac{\omega}{c}\mathcal{N}_+(\omega)\,\mathbf{s}_+\cdot\mathbf{x}}\right)_{|_{x^1=0}} = 0$$

for all $x^2, x^3 \in \mathbb{R}$. This is equivalent to

$$\mathbf{e}_1 \times \Big(\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}(\omega) + \boldsymbol{\mathcal{E}}_{\mathbf{s}'_-}(\omega) - \boldsymbol{\mathcal{E}}_{\mathbf{s}_+}(\omega)\Big) = 0 \tag{2.39}$$

and SNELL ***'s law*** [29]

$$\boxed{\mathbf{e}_1 \times \mathcal{N}_-(\omega)\,\mathbf{s}'_- = \mathbf{e}_1 \times \mathcal{N}_-(\omega)\,\mathbf{s}_- = \mathbf{e}_1 \times \mathcal{N}_+(\omega)\,\mathbf{s}_+\,.} \tag{2.40}$$

Defining

$$\mathbf{c}_3 \overset{\text{def}}{=} \frac{\mathbf{s}_- - \hat{P}_{\mathbf{e}_1}\mathbf{s}_-}{\sqrt{\Big(\mathbf{s}_- - \hat{P}_{\mathbf{e}_1}\mathbf{s}_-\Big)\cdot\Big(\mathbf{s}_- - \hat{P}_{\mathbf{e}_1}\mathbf{s}_-\Big)}}\,, \quad \mathbf{c}_2 \overset{\text{def}}{=} \mathbf{c}_3 \times \mathbf{e}_1$$



[27] For nonisotropic media the Ansatz has to be refined.

[28] A vector $\mathbf{z} \in \mathbb{C}^3$ is called ***isotropic*** iff $\mathbf{z}\cdot\mathbf{z} = 0$.

[29] Usually SNELL's law is given for the special case $\mathcal{N}_\pm(\omega) = n_\pm(\omega) \in \mathbb{R}$, $\mathbf{s}'_-, \mathbf{s}_\pm \in \mathbb{R}^3$. Then:

$$\boxed{n_-(\omega)\,\sin\theta'_- = n_-(\omega)\,\sin\theta_- = n_+(\omega)\,\sin\theta_+\,,}$$

where

$$\mathbf{s}_\pm = \cos\theta_\pm\,\mathbf{e}_1 + \sin\theta_\pm\,\mathbf{e}_3\,; \quad \mathbf{s}'_- = -\cos\theta'_-\,\mathbf{e}_1 + \sin\theta'_-\,\mathbf{e}_3\,, \quad \mathbf{e}_3 \overset{\text{def}}{=} \mathbf{c}_3 \in \mathbb{R}^3\,.$$

we get a basis $\{\mathbf{e}_1, \mathbf{c}_2, \mathbf{c}_3\}$ of $\mathbb{C}^3$ that is orthonormal in the sense that

$$\mathbf{c}_j \cdot \mathbf{c}_k = \delta_{jk}\,, \quad \mathbf{c}_1 \stackrel{\text{def}}{=} \mathbf{e}_1\,.$$

For $\mathbf{c}_3$ introduced this way SNELL's law implies

$$\mathbf{s}'_-,\ \mathbf{s}_-,\ \mathbf{s}_+ \in \operatorname{Span}\{\mathbf{e}_1, \mathbf{c}_3\} \subset \mathbb{C}^3\,. \tag{2.41}$$

Similarly to (2.39), assuming vanishing permeability, we get

$$\mathbf{e}_1 \times \left(\boldsymbol{\mathcal{B}}_{\mathbf{s}_-}(\omega) + \boldsymbol{\mathcal{B}}_{\mathbf{s}'_-}(\omega) - \boldsymbol{\mathcal{B}}_{\mathbf{s}_+}(\omega)\right) = 0\,. \tag{2.42}$$

By (2.33) the latter is equivalent to

$$0 = \mathbf{e}_1 \times \left(\mathcal{N}_-(\omega)\ \left(\mathbf{s}_- \times \boldsymbol{\mathcal{E}}_{\mathbf{s}_-}(\omega) + \mathbf{s}'_- \times \boldsymbol{\mathcal{E}}_{\mathbf{s}'_-}(\omega)\right) - \mathcal{N}_+(\omega)\,\mathbf{s}_+ \times \boldsymbol{\mathcal{E}}_{\mathbf{s}_+}(\omega)\right)\,. \tag{2.43}$$

Thanks to (2.41) and (2.31) the splitting[30]

$$\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) = \mathcal{E}^{\perp}_{\mathbf{s}}(\omega)\,\mathbf{c}_2 + \mathcal{E}^{\parallel}_{\mathbf{s}}(\omega)\,(\mathbf{c}_2 \times \mathbf{s}) \quad \text{for } \mathbf{s} = \mathbf{s}_\pm, \mathbf{s}'_-\,. \tag{2.44}$$

is possible iff (2.32) holds.

**Proof of (2.44):** Obviously, due to our assumptions concerning the material constants, (2.32) is equivalent to

$$\mathbf{s} \cdot \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) = 0\,. \tag{2.45}$$

According to (2.41) and (2.31) $\{\mathbf{s}\,, \mathbf{c}_2\,, \mathbf{c}_2 \times \mathbf{s}\}$ is a Basis[31] of $\mathbb{C}^3$. Therefore, a splitting of the form

$$\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) = \mathcal{E}^{\perp}_{\mathbf{s}}(\omega)\,\mathbf{c}_2 + \mathcal{E}^{\parallel}_{\mathbf{s}}(\omega)\,(\mathbf{c}_2 \times \mathbf{s}) + \mathcal{E}^{\mathrm{add}}_{\mathbf{s}}(\omega)\,\mathbf{s}$$

is possible. But, since $\mathbf{s} \cdot \mathbf{c}_2 = 0 = \mathbf{s} \cdot (\mathbf{c}_2 \times \mathbf{s})$, $\mathcal{E}^{\mathrm{add}}_{\mathbf{s}}(\omega)$ vanishes iff (2.45) holds. ∎

With this splitting, because (2.36) and (2.41) imply

$$\mathbf{e}_1 \times \Big(\mathbf{s} \times (\mathbf{c}_2 \times \mathbf{s})\Big) = \mathbf{e}_1 \times \mathbf{c}_2\,, \quad \mathbf{e}_1 \times (\mathbf{c}_2 \times \mathbf{s}) = (\mathbf{e}_1 \cdot \mathbf{s})\,\mathbf{c}_2\,,$$

(2.43) becomes equivalent to

$$\begin{gathered}
\mathcal{N}_-(\omega)\,\left(\mathcal{E}^{\parallel}_{\mathbf{s}_-}(\omega) + \mathcal{E}^{\parallel}_{\mathbf{s}'_-}(\omega)\right) = \mathcal{N}_+(\omega)\,\mathcal{E}^{\parallel}_{\mathbf{s}_+}(\omega)\,,\\
\mathcal{N}_-(\omega)\,\left((\mathbf{e}_1 \cdot \mathbf{s}_-)\,\mathcal{E}^{\perp}_{\mathbf{s}_-}(\omega) + (\mathbf{e}_1 \cdot \mathbf{s}'_-)\mathcal{E}^{\perp}_{\mathbf{s}'_-}(\omega)\right) = \mathcal{N}_+(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_+)\,\mathcal{E}^{\perp}_{\mathbf{s}_+}(\omega)\,.
\end{gathered} \tag{2.46}$$

Similarly we see that (2.39) is equivalent to

$$\begin{gathered}
\mathcal{E}^{\perp}_{\mathbf{s}_-}(\omega) + \mathcal{E}^{\perp}_{\mathbf{s}'_-}(\omega) = \mathcal{E}^{\perp}_{\mathbf{s}_+}(\omega)\,,\\
(\mathbf{e}_1 \cdot \mathbf{s}_-)\,\mathcal{E}^{\parallel}_{\mathbf{s}_-}(\omega) + (\mathbf{e}_1 \cdot \mathbf{s}'_-)\mathcal{E}^{\parallel}_{\mathbf{s}'_-}(\omega) = (\mathbf{e}_1 \cdot \mathbf{s}_+)\,\mathcal{E}^{\parallel}_{\mathbf{s}_+}(\omega)\,.
\end{gathered} \tag{2.47}$$

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[30] The notions $^\perp$ and $^\parallel$ refer to the $\mathbf{e}_1$-$\mathbf{c}_3$-plane in $\mathbb{C}^3$, $\mathbf{z}_1 \perp \mathbf{z}_2$ meaning $\mathbf{z}_1 \cdot \mathbf{z}_2 = 0$.

[31] Note that $(\mathbf{c}_2 \times \mathbf{s}) \cdot (\mathbf{c}_2 \times \mathbf{s}) = (\mathbf{c}_2 \cdot \mathbf{c}_2)\,(\mathbf{s} \cdot \mathbf{s}) - (\mathbf{c}_2 \cdot \mathbf{s})^2 = 1$.

**Exercise 3** Using (2.31) and (2.41), show the following:

a) (2.47), (2.46), (2.38), and SNELL's law imply

$$\mathbf{e}_1 \cdot \mathbf{s}'_- = -\,\mathbf{e}_1 \cdot \mathbf{s}_- \,, \tag{2.48}$$

if solutions with $\mathcal{E}^{\|}_{\mathbf{s}_+}(\omega) \neq 0$ as well as those with $\mathcal{E}^{\perp}_{\mathbf{s}_+}(\omega) \neq 0$ exist.

b)

$$\mathbf{s}'_- = (\mathbf{c}_3 \cdot \mathbf{s}_-)\,\mathbf{c}_3 - (\mathbf{e}_1 \cdot \mathbf{s}_-)\,\mathbf{e}_1 \,. \tag{2.49}$$

c)

$$\mathbf{s}_+ = \frac{\mathcal{N}_-(\omega)}{\mathcal{N}_+(\omega)}\,(\mathbf{c}_3 \cdot \mathbf{s}_-)\,\mathbf{c}_3 \pm \sqrt{1 - \left(\frac{\mathcal{N}_-(\omega)}{\mathcal{N}_+(\omega)}\,(\mathbf{c}_3 \cdot \mathbf{s}_-)\right)^2}\;\mathbf{e}_1 \,. \tag{2.50}$$

By (2.48), both (2.46) and (2.47) together are equivalent to FRESNEL***'s formulas***[32]

$$\boxed{\mathcal{E}^{\|}_{\mathbf{s}_+}(\omega) = \frac{2\,\mathcal{N}_-(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_-)}{\mathcal{N}_-(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_+) + \mathcal{N}_+(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_-)}\,\mathcal{E}^{\|}_{\mathbf{s}_-}(\omega)\,,} \tag{2.51}$$

$$\boxed{\mathcal{E}^{\perp}_{\mathbf{s}_+}(\omega) = \frac{2\,\mathcal{N}_-(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_-)}{\mathcal{N}_-(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_-) + \mathcal{N}_+(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_+)}\,\mathcal{E}^{\perp}_{\mathbf{s}_-}(\omega)\,,} \tag{2.52}$$

$$\boxed{\mathcal{E}^{\|}_{\mathbf{s}'_-}(\omega) = \frac{\mathcal{N}_+(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_-) - \mathcal{N}_-(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_+)}{\mathcal{N}_+(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_-) + \mathcal{N}_-(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_+)}\,\mathcal{E}^{\|}_{\mathbf{s}_-}(\omega)\,,} \tag{2.53}$$

$$\boxed{\mathcal{E}^{\perp}_{\mathbf{s}'_-}(\omega) = \frac{\mathcal{N}_-(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_-) - \mathcal{N}_+(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_+)}{\mathcal{N}_-(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_-) + \mathcal{N}_+(\omega)\,(\mathbf{e}_1 \cdot \mathbf{s}_+)}\,\mathcal{E}^{\perp}_{\mathbf{s}_-}(\omega)\,.} \tag{2.54}$$

**Exercise 4** Using SNELL law, show the following:

a) (2.43) implies

$$\mathbf{e}_1 \cdot \left(\epsilon_{\mathrm{c}}^-(\omega)\,\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}(\omega) + \epsilon_{\mathrm{c}}^-(\omega)\,\boldsymbol{\mathcal{E}}_{\mathbf{s}'_-}(\omega) - \epsilon_{\mathrm{c}}^+(\omega)\,\boldsymbol{\mathcal{E}}_{\mathbf{s}_+}(\omega)\right) = 0\,.$$

b) grad $(\mathbf{c}_2 \cdot \mathbf{E}(\mathbf{x},t))$ is continuous.

c) grad $(\mathbf{c}_3 \cdot \mathbf{E}(\mathbf{x},t))$ is **not** continuous, in general.



[32] We assume that the denominators on the r.h.s. of (2.51)–(2.54) do not vanish. For the evaluation of these formulas in the case of nonabsorbing media and $\mathbf{s} \in \mathbb{R}^3$ see, e.g., (Monzón and Sánchez-Soto, 2001).

Let us consider the case[33]

$$\boxed{\Re\left(\mathbf{e}_1 \cdot \mathbf{s}_\pm\right) > 0\,.} \tag{2.55}$$

Then $\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}\, e^{-i\frac{\omega}{c}\left(c\,t-\mathcal{N}_-(\omega)\,\mathbf{s}_-\cdot\mathbf{x}\right)}$ may be interpreted as incoming wave,[34] reflected into $\mathcal{G}_-$ as $\boldsymbol{\mathcal{E}}_{\mathbf{s}'_-}\, e^{-i\frac{\omega}{c}\left(c\,t-\mathcal{N}_-(\omega)\,\mathbf{s}'_-\cdot\mathbf{x}\right)}$ and transmitted into $\mathcal{G}_+$ as $\boldsymbol{\mathcal{E}}_{\mathbf{s}_+}\, e^{-i\frac{\omega}{c}\left(c\,t-\mathcal{N}_+(\omega)\,\mathbf{s}_+\cdot\mathbf{x}\right)}$. As a direct consequence of (2.37) we have

$$\mathbf{e}_1 \cdot \left\langle \mathbf{S}_{\mathbf{s}_-}(\mathbf{x},t)\right\rangle_{|_{x^1=0}} = \mathbf{e}_1 \cdot \left\langle \mathbf{S}^{\perp}_{\mathbf{s}_-}(\mathbf{x},t)\right\rangle_{|_{x^1=0}} + \mathbf{e}_1 \cdot \left\langle \mathbf{S}^{\parallel}_{\mathbf{s}_-}(\mathbf{x},t)\right\rangle_{|_{x^1=0}}\,,$$

where[35]

$$\begin{aligned}
\mathbf{S}_{\mathbf{s}_-}(\mathbf{x},t) &\stackrel{\text{def}}{=} \text{\textsc{Poynting} vector of} \quad \boldsymbol{\mathcal{E}}_{\mathbf{s}_\pm}\, e^{-i\frac{\omega}{c}\left(c\,t-\mathcal{N}_-(\omega)\,\mathbf{s}_-\cdot\mathbf{x}\right)}\,,\\
\mathbf{S}^{\perp}_{\mathbf{s}_-}(\mathbf{x},t) &\stackrel{\text{def}}{=} \text{\textsc{Poyn}<\textsc{ting} vector of} \quad \mathcal{E}^{\perp}_{\mathbf{s}_-}\,\mathbf{c}_2\, e^{-i\frac{\omega}{c}\left(c\,t-\mathcal{N}_-(\omega)\,\mathbf{s}_-\cdot\mathbf{x}\right)}\,,\\
\mathbf{S}^{\parallel}_{\mathbf{s}_-}(\mathbf{x},t) &\stackrel{\text{def}}{=} \text{\textsc{Poynting} vector of} \quad \mathcal{E}^{\parallel}_{\mathbf{s}_-}\left(\mathbf{c}_2\times\mathbf{s}_-\right) e^{-i\frac{\omega}{c}\left(c\,t-\mathcal{N}_-(\omega)\,\mathbf{s}_-\cdot\mathbf{x}\right)}\,.
\end{aligned}$$

Since, similar relations hold for $\mathbf{s}'_-$ and $\mathbf{s}_+$ instead of $\mathbf{s}_-$, we may analyze the energy flow for the $^\perp$- and $^\parallel$-modes separately. Let us define

$$\left.\begin{aligned}
R^{\perp}_{\mathbf{s}_-}(\omega) &\stackrel{\text{def}}{=} -\frac{\mathbf{e}_1\cdot\left\langle \mathbf{S}^{\perp}_{\mathbf{s}'_-}(\mathbf{x},t)\right\rangle_{|_{x^1=0}}}{\mathbf{e}_1\cdot\left\langle \mathbf{S}^{\perp}_{\mathbf{s}_-}(\mathbf{x},t)\right\rangle_{|_{x^1=0}}}\,,\\
T^{\perp}_{\mathbf{s}_-}(\omega) &\stackrel{\text{def}}{=} +\frac{\mathbf{e}_1\cdot\left\langle \mathbf{S}^{\perp}_{\mathbf{s}_+}(\mathbf{x},t)\right\rangle_{|_{x^1=0}}}{\mathbf{e}_1\cdot\left\langle \mathbf{S}^{\perp}_{\mathbf{s}_-}(\mathbf{x},t)\right\rangle_{|_{x^1=0}}}
\end{aligned}\right\}\quad \text{for } \mathbf{e}_1\cdot\left\langle \mathbf{S}^{\perp}_{\mathbf{s}_-}(\mathbf{x},t)\right\rangle_{|_{x^1=0}} \neq 0$$

and similarly for $^\parallel$ instead of $^\perp$. Then by (2.37) and SNELL's law (2.40) we have

$$\begin{aligned}
R^{\perp}_{\mathbf{s}_-}(\omega) &= -\frac{\mathbf{e}_1\cdot\Re(\mathcal{N}_-(\omega)\mathbf{s}'_-)}{\mathbf{e}_1\cdot\Re(\mathcal{N}_-(\omega)\mathbf{s}_-)}\left|\mathcal{E}^{\perp}_{\mathbf{s}'_-}(\omega)\right|^2 \Big/ \left|\mathcal{E}^{\perp}_{\mathbf{s}_-}(\omega)\right|^2\\
&\underset{(2.48),(2.54)}{=} \left|\frac{\mathcal{N}_-(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_-\right)-\mathcal{N}_+(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_+\right)}{\mathcal{N}_-(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_-\right)+\mathcal{N}_+(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_+\right)}\right|^2
\end{aligned} \tag{2.56}$$

and

$$\begin{aligned}
T^{\perp}_{\mathbf{s}_-}(\omega) &= +\frac{\mathbf{e}_1\cdot\Re(\mathcal{N}_+(\omega)\mathbf{s}_+)}{\mathbf{e}_1\cdot\Re(\mathcal{N}_-(\omega)\mathbf{s}_-)}\left|\mathcal{E}^{\perp}_{\mathbf{s}_+}(\omega)\right|^2 \Big/ \left|\mathcal{E}^{\perp}_{\mathbf{s}_-}(\omega)\right|^2\\
&\underset{(2.53)}{=} \frac{\Re\Big(\mathcal{N}_+(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_+\right)\Big)}{\Re\Big(\mathcal{N}_-(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_-\right)\Big)}\left|\frac{2\,\mathcal{N}_-(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_-\right)}{\mathcal{N}_-(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_-\right)+\mathcal{N}_+(\omega)\left(\mathbf{e}_1\cdot\mathbf{s}_+\right)}\right|^2
\end{aligned} \tag{2.57}$$



[33] For $\Re\left(\mathbf{e}_1\cdot\mathbf{s}_+\right) < 0$ we have incoming waves from both sides of the boundary.

[34] Actually we should form wave packets by integrating with suitable weights over $\mathbf{s}_-$ and $\omega$.

[35] Recall (2.44).

Therefore[36]

$$
\begin{aligned}
& R^{\perp}_{\mathbf{s}_-}(\omega) + T^{\perp}_{\mathbf{s}_-}(\omega) = 1 \\
\iff & \frac{\Re\Big(\mathcal{N}_+(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_+)\Big)}{\Re\Big(\mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-)\Big)} = \Re\left(\frac{\mathcal{N}_+(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_+)}{\mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-)}\right) \\
\iff & \Big\{\mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-)\,,\,\mathcal{N}_+(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_+)\,\overline{\mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-)}\Big\} \cap \mathbb{R} \neq \emptyset\,,
\end{aligned}
$$

i.e.:

> In general, the usual interpretation of $R^{\perp}_{\mathbf{s}_-}(\omega)$ as ***reflectance*** and $T^{\perp}_{\mathbf{s}_-}(\omega)$ as ***transmittance*** (of the $^\perp$-mode) is not strictly justified.

For the $^{\|}$-mode the situation is more complicated, since, e.g.,[37]

$$
\Re\left(\mathcal{N}_-(\omega)\left(\overline{\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}(\omega)}\cdot\mathbf{s}_-\right)\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}(\omega)\right) = i\,\mathbf{c}_2\cdot\left(\overline{\mathbf{s}_-}\times\mathbf{s}_-\right)\Im\left(\mathcal{N}_-(\omega)\,\overline{\mathcal{E}^{\|}_{\mathbf{s}_-}(\omega)}\,\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}(\omega)\right)\,.
$$

The latter, however, implies

$$
\mathbf{e}_1\cdot\Re\left(\mathcal{N}_-(\omega)\left(\overline{\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}(\omega)}\cdot\mathbf{s}_-\right)\boldsymbol{\mathcal{E}}_{\mathbf{s}_-}(\omega)\right) = i\,\mathbf{c}_2\cdot\left(\overline{\mathbf{s}_-}\times\mathbf{s}_-\right)\left|\mathcal{E}^{\|}_{\mathbf{s}_-}(\omega)\right|^2\Im\Big(\mathcal{N}_-(\omega)\,(\mathbf{c}_3\cdot\mathbf{s}_-)\Big)\,. \tag{2.58}
$$

and, therefore, similarly to (2.56) and (2.57) we get

$$
\begin{aligned}
& \Im\Big(\mathcal{N}_-(\omega)\,(\mathbf{c}_3\cdot\mathbf{s}_-)\Big)\left(\overline{\mathbf{s}_-}\times\mathbf{s}_-\right) = \Im\Big(\mathcal{N}_+(\omega)\,(\mathbf{c}_3\cdot\mathbf{s}_+)\Big)\left(\overline{\mathbf{s}_+}\times\mathbf{s}_+\right) = 0 \\
\Longrightarrow\ & R^{\|}_{\mathbf{s}_-}(\omega) = \left|\frac{\mathcal{N}_+(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-) - \mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_+)}{\mathcal{N}_+(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-) + \mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_+)}\right|^2
\end{aligned} \tag{2.59}
$$

and

$$
\begin{aligned}
& \Im\Big(\mathcal{N}_-(\omega)\,(\mathbf{c}_3\cdot\mathbf{s}_-)\Big)\left(\overline{\mathbf{s}_-}\times\mathbf{s}_-\right) = \Im\Big(\mathcal{N}_+(\omega)\,(\mathbf{c}_3\cdot\mathbf{s}_+)\Big)\left(\overline{\mathbf{s}_+}\times\mathbf{s}_+\right) = 0 \\
\Longrightarrow\ & T^{\|}_{\mathbf{s}_-}(\omega) = \frac{\Re\Big(\mathcal{N}_+(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_+)\Big)}{\Re\Big(\mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-)\Big)}\left|\frac{2\,\mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-)}{\mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_+) + \mathcal{N}_+(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-)}\right|^2
\end{aligned} \tag{2.60}
$$

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[36] Note that

$$
\frac{\Re(z_+)}{\Re(z_-)} = \Re\left(\frac{z_+}{z_-}\right) \iff \overline{z_+}\,z_-\left(\overline{z_-} - z_-\right) = z_+\,\overline{z_-}\left(\overline{z_-} - z_-\right)\,.
$$

For a discussion of the reflection and transmission in the case of normal incidence see (Lodenquai, 1991).

[37] Note that, e.g.,

$$
i\,\mathbf{c}_2\cdot\left(\overline{\mathbf{s}_-}\times\mathbf{s}_-\right) = 2\,\alpha\,\beta \quad \text{for } \mathbf{s}_- = \alpha\,\mathbf{e}_1 + i\,\beta\,\mathbf{c}_3\ \ \alpha,\beta\in\mathbb{R}\,.
$$

**Warning:** Note that (2.56), (2.57), (2.59), and (2.60) hold only for $\Re\Big(\mathcal{N}_-(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_-)\Big)\neq 0\,.$

A direct consequence of (2.57) is that $\Re\Big(\mathcal{N}_+(\omega)\,(\mathbf{e}_1\cdot\mathbf{s}_+)\Big)=0$ implies $T^{\perp}_{\mathbf{s}_-}(\omega)=0\,,$ i.e. ***total reflection*** of the $^{\perp}$-mode.[38] If, in addition,

$$\Im\Big(\mathcal{N}_-(\omega)\,(\mathbf{c}_3\cdot\mathbf{s}_-)\Big)\,(\overline{\mathbf{s}_-}\times\mathbf{s}_-)=\Im\Big(\mathcal{N}_+(\omega)\,(\mathbf{c}_3\cdot\mathbf{s}_+)\Big)\,(\overline{\mathbf{s}_+}\times\mathbf{s}_+)=0$$

holds then, by (2.60), also $T^{\parallel}_{\mathbf{s}_-}(\omega)=0\,.$

Strictly speaking, however, total reflection appears only since $\mathcal{G}_+$ has infinite diameter. If $\mathcal{G}_+$ is replaced by a layer of finite depth, then ***optical tunneling*** is possible, i.e. partial transmission of radiation under the conditions of otherwise total reflection. Then, however, there is a superposition of two evanescent modes inside $\mathcal{G}_+$ with non-vanishing $\mathbf{e}_1$-component of the total Poynting vector.[39]

**Remarks:**

1. Multiple total reflection may be used for building useful optical devices (e.g. wave guides).
2. In case of total reflection the evanescent wave inside $\mathcal{G}_+$ has higher **spatial** frequency ($\widehat{=}$higher resolution) in the direction of propagation than usual and may have considerable amplitudes **near** the boundary of the media. These modes are exploited by **near**-field scanning optical microscopy[40] (NSOM), converting the evanescent modes into propagating ones.
3. Usually scattered light contains evanescent near-field components (Wolf and Nieto-Vesperinas, 1985).
4. Optical tunneling may be used for coupling wave guides.

**Exercise 5** For real $\mathcal{N}_\pm(\omega)=\mathcal{N}_\pm(\omega)=n_\pm(\omega)$ and $\mathbf{s}_-\in\mathbb{R}^3$ show that $R^{\parallel}_{\mathbf{s}_-}(\omega)$ vanishes iff

$$\frac{(\mathbf{e}_1\cdot\mathbf{s}_-)^2}{1-(\mathbf{e}_1\cdot\mathbf{s}_-)^2}=\left(\frac{n_-(\omega}{n_+(\omega)}\right)^2\,,$$



[38] Usually the case

$$\mathcal{N}_\pm(\omega)=n_\pm(\omega)\,,\quad n_-(\omega)\,\sin\theta_->n_+(\omega)\,,\quad \mathbf{s}_-\in\mathbb{R}^3$$

is considered, corresponding to total reflection since the $\mathbf{e}_1$-component of (2.50) becomes purely imaginary.

[39] And the results for incident monochromatic plane waves should not be applied too naively to almost monochromatic incident rays!

[40] See, e.g., (Ohtsu and Hori, 1999) and (Paesler and Moyer, 1996).

i.e. iff $\angle \mathbf{e}_1, \mathbf{s}_-$ coincides with the so-called Brewster ***angle*** $\arctan \dfrac{n_-(\omega)}{n_+(\omega)}$ . Moreover, show that in this case

$$R^{\|}_{\mathbf{s}_-}(\omega) = 0 \quad \Longleftrightarrow \quad \mathbf{s}'_- \cdot \mathbf{s}_+ = 0\,.$$

### 2.2.3 Birefringent Media (Crystal Optics)

Generally, (2.32) is equivalent to

$$\overset{\leftrightarrow}{M}_{\mathbf{s}}(\omega)\, \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) = 0\,, \tag{2.61}$$

where

$$\overset{\leftrightarrow}{M}_{\mathbf{s}}(\omega) \stackrel{\text{def}}{=} \Big(\mathcal{N}_{\mathbf{s}}(\omega)\Big)^2 \left(\overset{\leftrightarrow}{1} - \begin{pmatrix} s^1 s^1 & s^1 s^2 & s^1 s^3 \\ s^2 s^1 & s^2 s^2 & s^2 s^3 \\ s^3 s^1 & s^3 s^2 & s^3 s^3 \end{pmatrix}\right) - \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\,.$$

(2.61) can be fulfilled iff

$$\det\Big(\overset{\leftrightarrow}{M}_{\mathbf{s}}(\omega)\Big) = 0\,. \tag{2.62}$$

Therefore, we have to choose the ***complex refractive index*** $\mathcal{N}_{\mathbf{s}}(\omega)$ such that (2.62) is fulfilled and determine the (complex) direction of $\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)$ from (2.32) for such $\mathcal{N}_{\mathbf{s}}(\omega)$ .

Reasonable physical assumptions imply that[41]

$$\mathbf{a} \cdot \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \mathbf{b} = \mathbf{b} \cdot \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \mathbf{a} \quad \forall\, \mathbf{a}, \mathbf{b} \in \mathbb{C}^3\,. \tag{2.63}$$

Therefore, if $\mathcal{N}_{\mathbf{s}}(\omega)$ resp. $\mathcal{N}'_{\mathbf{s}}(\omega)$ are solutions of (2.62) and $\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)$ resp. $\boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega)$ corresponding solutions of (2.32), we have

$$\boxed{\Big(\mathcal{N}_{\mathbf{s}}(\omega)\Big)^2 \neq \Big(\mathcal{N}'_{\mathbf{s}}(\omega)\Big)^2 \implies \Big(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\Big) \cdot \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega) = 0\,.} \tag{2.64}$$

**Proof of (2.64):**

$$\begin{aligned}
&\left(\mathcal{N}'_{\mathbf{s}}(\omega)\right)^2 \Big(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\Big) \cdot \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega) \\
\underset{(2.32)}{=}\; & \left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2 \left(\mathcal{N}'_{\mathbf{s}}(\omega)\right)^4 \overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\, \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) \cdot \overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\, \boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega) \\
=\; & \left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2 \left(\mathcal{N}'_{\mathbf{s}}(\omega)\right)^4 \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) \cdot \overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\, \boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega) \\
\underset{(2.32)}{=}\; & \left(\mathcal{N}_{\mathbf{s}}(\omega)\, \mathcal{N}'_{\mathbf{s}}(\omega)\right)^2 \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) \cdot \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega) \\
\underset{(2.63)}{=}\; & \left(\mathcal{N}_{\mathbf{s}}(\omega)\, \mathcal{N}'_{\mathbf{s}}(\omega)\right)^2 \boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega) \cdot \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) \\
\underset{(2.32)}{=}\; & \left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2 \Big(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega)\Big) \cdot \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\, \boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\,.
\end{aligned}$$



[41] See (Römer, 1994, Ends of Sections 2.4 and 2.7).

Because of

$$\mathbf{a}\cdot\mathbf{b}=\mathbf{b}\cdot\mathbf{a}\quad\forall\,\mathbf{a},\mathbf{b}\in\mathbb{C}^3$$

this implies

$$\left(\left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2-\left(\mathcal{N}'_{\mathbf{s}}(\omega)\right)^2\right)\left(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\,\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\right)\cdot\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\,\boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega)=0$$

and hence (2.64). ∎

Note that (2.64), (2.32), and (2.31) imply

$$\left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2\neq\left(\mathcal{N}'_{\mathbf{s}}(\omega)\right)^2\implies\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\cdot\boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega)=\left(\mathbf{s}\cdot\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\right)\left(\mathbf{s}\cdot\boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega)\right).\tag{2.65}$$

$\mathbf{s}$ is said to correspond to an ***optical axis*** for given $\omega$, if there is a unique complex refractive index $\mathcal{N}_{\mathbf{s}}(\omega)=\mathcal{N}^{\mathrm{ord}}_{\mathbf{s}}(\omega)$ for which there are two independent solutions $\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)$ of (2.32).

**Lemma 2.2.1** $\mathbf{s}$ *corresponds to an optical axis for given* $\omega$ *iff*

$$\mathbf{s}\cdot\mathbf{e}=0\quad\implies\quad\mathbf{e}\cdot\left(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\right)^{-1}\mathbf{e}=\left(\mathcal{N}^{\mathrm{ord}}_{\mathbf{s}}(\omega)\right)^{-2}\tag{2.66}$$

*holds for all (complex) directions* $\mathbf{e}$.

**Proof:** Let $\mathbf{s}$ correspond to an optical axis and let $\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega),\boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega)$ be independent solutions of (2.32) for $\mathcal{N}_{\mathbf{s}}(\omega)=\mathcal{N}^{\mathrm{ord}}_{\mathbf{s}}(\omega)$. Then, because of

$$\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)=\left(\mathcal{N}^{\mathrm{ord}}_{\mathbf{s}}(\omega)\right)^2\left(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\right)^{-1}\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega),\tag{2.67}$$

(and the corresponding equation for $\boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega)$) also $\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)$ and $\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\boldsymbol{\mathcal{E}'}_{\mathbf{s}}(\omega)$ have to be independent. Moreover, (2.67) implies

$$\left(\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\right)=\left(\mathcal{N}^{\mathrm{ord}}_{\mathbf{s}}(\omega)\right)^2\overset{\leftrightarrow}{I}_{\mathrm{s}}\left(\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega)\right)$$

for

$$\overset{\leftrightarrow}{I}_{\mathrm{s}}\overset{\mathrm{def}}{=}\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\left(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\right)^{-1}\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}.$$

Therefore,

$$\overset{\leftrightarrow}{I}_{\mathrm{s}}=\frac{1}{\left(\mathcal{N}^{\mathrm{ord}}_{\mathbf{s}}(\omega)\right)^2}\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}.$$

Since

$$\mathbf{s}\cdot\mathbf{e}=0\iff\mathbf{e}=\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\mathbf{e}$$

and

$$\mathbf{a}\cdot\left(\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\mathbf{b}\right)=\left(\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\,\mathbf{a}\right)\cdot\mathbf{b}\quad\forall\,\mathbf{a},\mathbf{b}\in\mathbb{C}^3,$$

we conclude[42] that (2.66) holds. Conversely, if (2.66) holds, then

$$\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) = \left(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\right)^{-1} \mathbf{z}$$

is a solution of (2.32) for every $\mathbf{z} \in \overset{\leftrightarrow}{P}_{\perp\mathbf{s}} \mathbb{C}^3$. The latter shows that $\mathbf{s}$ corresponds to an optical axis. ∎

Let us assume that $\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)$ may be diagonalized:

$$\boxed{\begin{gathered}\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega) = \begin{pmatrix} \epsilon_{\mathrm{c}}^1(\omega) & 0 & 0 \\ 0 & \epsilon_{\mathrm{c}}^2(\omega) & 0 \\ 0 & 0 & \epsilon_{\mathrm{c}}^3(\omega) \end{pmatrix} \quad \text{w.r.t. } \{\mathbf{z}_1(\omega), \mathbf{z}_2(\omega), \mathbf{z}_3(\omega)\} \subset \mathbb{C}^3\,, \\ \mathbf{z}_j(\omega) \cdot \mathbf{z}_k(\omega) = \delta_{jk} \quad \forall\, j,k \in \{1,2,3\}\,.\end{gathered}} \tag{2.68}$$

**Remark:** (2.68) is guaranteed by (2.63), for suitable $\mathbf{z}_j(\omega) \in \mathbb{C}^3$, if $\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)$ does not have any isotropic eigenvector; see Lemma 7.4.13 of (Lücke, eine). Note, however, that there are media (monoclinic and triclinic crystals) for which the eigenvectors of $\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)$ depend on $\omega$.

If (2.68) holds with $\epsilon_{\mathrm{c}}^2(\omega) = \epsilon_{\mathrm{c}}^3(\omega)$, then $\pm\mathbf{z}_1(\omega)$ corresponds to an optical axis and

$$\boxed{\mathcal{N}^{\mathrm{ord}}_{\mathbf{z}_1(\omega)}(\omega) = \sqrt{\epsilon_{\mathrm{c}}^2(\omega)}\,.} \tag{2.69}$$

In this case, whenever $\mathbf{s} \not\sim \mathbf{z}_1(\omega)$,

$$\boldsymbol{\mathcal{E}}_{\mathbf{s}}(\omega) = \boxed{\boldsymbol{\mathcal{E}}^{\mathrm{ord}}_{\mathbf{s}}(\omega) \sim \mathbf{s} \times \mathbf{z}_1(\omega)\,,} \tag{2.70}$$

is a solution of (2.32) for

$$\mathcal{N}_{\mathbf{s}}(\omega) = \mathcal{N}^{\mathrm{ord}}_{\mathbf{z}_1(\omega)}(\omega)\,.$$

Note that, by (2.37),

$$\mathbf{s} \in \mathbb{R} \quad \Longrightarrow \quad \left\langle \mathbf{S}^{\mathrm{ord}}_{\mathbf{s}}(\mathbf{x},t)\right\rangle \sim \mathbf{s}\,. \tag{2.71}$$

To determine the second solution of (2.62) we may assume, without loss of generality, that

$$\mathbf{s} = s^1(\omega)\,\mathbf{z}_1(\omega) + s^3(\omega)\,\mathbf{z}_3(\omega)\,.$$

Then, w.r.t. $\{\mathbf{z}_1(\omega), \mathbf{z}_2(\omega), \mathbf{z}_3(\omega)\}$, by (2.31), we have

$$\overset{\leftrightarrow}{M}(\mathbf{k}_0) = \left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2 \begin{pmatrix} +s^3(\omega)s^3(\omega) & 0 & -s^1(\omega)s^3(\omega) \\ 0 & 1 & 0 \\ -\,s^3(\omega)s^1(\omega) & 0 & +s^1(\omega)s^1(\omega) \end{pmatrix} - \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)$$



[42] Recall that $\overset{\leftrightarrow}{P}_{\perp\mathbf{s}}\overset{\leftrightarrow}{P}_{\perp\mathbf{s}} = \overset{\leftrightarrow}{P}_{\perp\mathbf{s}}$.

and hence (2.62) holds iff either $\mathcal{N}_{\mathbf{s}}(\omega) = \mathcal{N}^{\text{ord}}_{\mathbf{z}_1(\omega)}(\omega)$ or

$$\begin{aligned} 0 &= \left(\left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2 s^3(\omega)s^3(\omega) - \epsilon^1_{\text{c}}(\omega)\right)\left(\left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^2 s^1(\omega)s^1(\omega) - \epsilon^3_{\text{c}}(\omega)\right) \\ &\qquad - \left(\mathcal{N}_{\mathbf{s}}(\omega)\right)^4 s^1(\omega)s^1(\omega)s^3(\omega)s^3(\omega) \\ &= \epsilon^1_{\text{c}}(\omega)\,\epsilon^3_{\text{c}}(\omega)\left(1 - \frac{\left(\mathcal{N}_{\mathbf{s}}(\omega)\,s^1(\omega)\right)^2}{\epsilon^3_{\text{c}}(\omega)} - \frac{\left(\mathcal{N}_{\mathbf{s}}(\omega)\,s^3(\omega)\right)^2}{\epsilon^1_{\text{c}}(\omega)}\right). \end{aligned}$$

Therefore another solution $\boldsymbol{\mathcal{E}}'_{\mathbf{s}}(\omega)$ of (2.32) exists for $\mathcal{N}_{\mathbf{s}}(\omega) = \mathcal{N}^{\text{exo}}_{\mathbf{s}}(\omega)$, the latter being implicitly defined by

$$\boxed{\left(\frac{\mathcal{N}^{\text{exo}}_{\mathbf{s}}(\omega)\,s^1(\omega)}{\mathcal{N}^{\text{ord}}_{\mathbf{s}}(\omega)}\right)^2 + \left(\frac{\mathcal{N}^{\text{exo}}_{\mathbf{s}}(\omega)\,s^2(\omega)}{\sqrt{\epsilon^1_{\text{c}}(\omega)}}\right)^2 + \left(\frac{\mathcal{N}^{\text{exo}}_{\mathbf{s}}(\omega)\,s^3(\omega)}{\sqrt{\epsilon^1_{\text{c}}(\omega)}}\right)^2 = 1\,.} \tag{2.72}$$

In the nonisotropic case, (2.70) and (2.64) imply

$$\boxed{\boldsymbol{\mathcal{E}}^{\text{exo}}_{\mathbf{s}}(\omega) \sim \left(\overset{\leftrightarrow}{\epsilon}_{\text{c}}(\omega)\right)^{-1}\left(\mathbf{s}\times\left(\mathbf{s}\times\mathbf{z}_1(\omega)\right)\right).} \tag{2.73}$$

In agreement with (2.65) we have $\boldsymbol{\mathcal{E}}^{\text{exo}}_{\mathbf{s}}(\omega)\cdot\boldsymbol{\mathcal{E}}^{\text{ord}}_{\mathbf{s}}(\omega) = 0$, in the present case.[43]

**Exercise 6** Show that

$$\boldsymbol{\mathcal{E}}^{\text{ord}}_{\mathbf{s}}(\omega)\cdot\left\langle\mathbf{S}^{\text{ord}}_{\mathbf{s}}(\mathbf{x},t)\right\rangle = \boldsymbol{\mathcal{E}}^{\text{exo}}_{\mathbf{s}}(\omega)\cdot\langle\mathbf{S}^{\text{exo}}_{\mathbf{s}}(\mathbf{x},t)\rangle = 0$$

and

$$\langle\mathbf{S}^{\text{exo}}_{\mathbf{s}}(\mathbf{x},t)\rangle \in \text{Span}\,\{\mathbf{s},\boldsymbol{\mathcal{E}}^{\text{exo}}_{\mathbf{s}}(\omega)\} \subset \text{Span}\left\{\mathbf{s},\mathbf{z}_1(\omega)\right\}$$

**if** $\mathbf{z}_1(\omega) \not\sim \mathbf{s} \in \mathbb{R}^3$ and $\overset{\leftrightarrow}{\epsilon}_{\text{c}} = \left(\overset{\leftrightarrow}{\epsilon}_{\text{c}}\right)^\dagger$.

**Remarks:**

1. For real directions $\mathbf{e}$ and real $\overset{\leftrightarrow}{\epsilon}_{\text{c}}(\omega)$ we have

$$\left(\overset{\leftrightarrow}{\epsilon}_{\text{c}}(\omega)\right)^{-1}\mathbf{e} = \frac{1}{2}\left(\boldsymbol{\nabla}_{\mathbf{x}}\left(\mathbf{x}\cdot\left(\overset{\leftrightarrow}{\epsilon}_{\text{c}}(\omega)\right)^{-1}\mathbf{x}\right)\right)_{|\mathbf{x}=\mathbf{e}}$$

and, therefore, $\left(\overset{\leftrightarrow}{\epsilon}_{\text{c}}(\omega)\right)^{-1}\mathbf{e} \sim \mathbf{n}(\mathbf{e})$, where $\mathbf{n}(\mathbf{e})$ denotes the normal of the ***index ellipsoid***

$$\left\{\mathbf{x}\in\mathbb{R} : \left(\mathbf{x}\cdot\left(\overset{\leftrightarrow}{\epsilon}_{\text{c}}(\omega)\right)^{-1}\mathbf{x}\right) = 1\right\}$$

at $\mathbf{x}\sim\mathbf{e}$.

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[43]Note that, in the present case, $\left(\overset{\leftrightarrow}{\epsilon}_{\text{c}}(\omega)\right)^{-1}$ leaves the linear span of $\mathbf{s}$ and $\mathbf{z}_1(\omega)$ invariant.

2. Since, therefore, $\mathbf{s} \cdot \boldsymbol{\mathcal{E}}^{\mathrm{exo}}_{\mathbf{s}}(\omega) \overset{\text{i.g.}}{\neq} 0$ Exercise 6 and (2.71) show that

$$\langle \mathbf{S}^{\mathrm{exo}}_{\mathbf{s}}(\mathbf{x},t)\rangle \overset{\text{i.g.}}{\not\sim} \mathbf{s} \sim \big\langle \mathbf{S}^{\mathrm{ord}}_{\mathbf{s}}(\mathbf{x},t)\big\rangle\,;$$

i.e. the medium is ***birefringent***.

**Lemma 2.2.2** *If (2.68) holds with $\epsilon^1_{\mathrm{c}}(\omega) \neq \epsilon^2_{\mathrm{c}}(\omega)$ and if $\mathbf{s}$ is a (complex) direction corresponding to an optical axis for given $\omega$ then* $\prod_{j=1}^{3} \mathbf{s} \cdot \mathbf{z}_j(\omega) = 0\,.$

**Outline of proof:** Let $\mathbf{s}$ correspond to an optical axis. Then, by Lemma 2.2.1,

$$\mathbf{s} \cdot \mathbf{x} = 0 \neq \mathbf{x} \cdot \mathbf{x} \implies \sum_{j=1}^{3} (x^j)^2/\epsilon^j_{\mathrm{c}} = C\,\mathbf{x} \cdot \mathbf{x} \qquad \forall \mathbf{x} \in \mathbb{C}^3$$

holds for some $C = C(\omega) \in \mathbb{C}$, where $x^j \overset{\text{def}}{=} \mathbf{x} \cdot \mathbf{z}_j(\omega)$ for $j = 1,2,3$. Now assume $\prod_{j=1}^{3} s^j \neq 0$. Then

$$\mathbf{s} \cdot \mathbf{x} = 0 \iff \left(x^1\right)^2 = \left(\frac{s^2x^2 + s^3x^3}{s^1}\right)^2$$

and, therefore, we have

$$\begin{aligned}
\left(\frac{s^2x^2 + s^3x^3}{s^1}\right)^2 &+ \left(x^2\right)^2 + \left(x^3\right)^2 \neq 0 \\
&\implies \frac{1}{\epsilon^1_{\mathrm{c}}}\left(\frac{s^2x^2 + s^3x^3}{s^1}\right)^2 + \frac{1}{\epsilon^2_{\mathrm{c}}}\left(x^2\right)^2 + \frac{1}{\epsilon^3_{\mathrm{c}}}\left(x^3\right)^2 \\
&\qquad = C\left(\left(\frac{s^2x^2 + s^3x^3}{s^1}\right)^2 + \left(x^2\right)^2 + \left(x^3\right)^2\right)
\end{aligned}$$

for all $x^2, x^3 \in \mathbb{C}$. Comparing the $x^2x^3$-terms on both sides of the latter equation shows that $C = 1/\epsilon^1_{\mathrm{c}}$. Similarly we conclude that $C = 1/\epsilon^2_{\mathrm{c}}$. Therefore, $\prod_{j=1}^{3} s^j \neq 0$ can**not** hold if $\epsilon^1_{\mathrm{c}} \neq \epsilon^2_{\mathrm{c}}$. ∎

**Corollary 2.2.3** *If (2.68) holds with $\epsilon^1_{\mathrm{c}}(\omega) \neq \epsilon^2_{\mathrm{c}}(\omega)$ and if $\mathbf{s}$ is a (complex) direction with $\mathbf{s} \cdot \mathbf{z}_2(\omega) = 0$, for given $\omega$, then $\mathbf{s}$ corresponds to an optical axis if and only if $\epsilon^1_{\mathrm{c}}(\omega) \neq \epsilon^3_{\mathrm{c}}(\omega)$ and*

$$\mathbf{s} = \pm\sqrt{1 - (s^3)^2}\,\mathbf{z}_1(\omega) + s^3\,\mathbf{z}_3(\omega)\,, \quad s^3 = \pm\sqrt{\frac{\epsilon^3_{\mathrm{c}}(\omega) - \epsilon^2_{\mathrm{c}}(\omega)}{\epsilon^3_{\mathrm{c}}(\omega) - \epsilon^1_{\mathrm{c}}(\omega)}\,\frac{\epsilon^1_{\mathrm{c}}(\omega)}{\epsilon^2_{\mathrm{c}}(\omega)}}\,. \tag{2.74}$$

**Outline of proof:** Let the direction $\mathbf{s} \perp \mathbf{z}_2(\omega)$ correspond to an optical axis. Then there are complex numbers $C$ and $s^3$ with

$$\mathbf{s} = \pm\sqrt{1 - (s^1)^2}\, \mathbf{z}_1(\omega) + s^3\, \mathbf{z}_3(\omega)$$

and

$$\mathbf{s} \cdot \mathbf{s}' = 0 \implies \mathbf{s}' \cdot \left(\overleftrightarrow{\epsilon}_{\mathrm{c}}(\omega)\right)^{-1} \mathbf{s}' = C$$

for all directions $\mathbf{s}'$ (by Lemma 2.2.1). Especially for

$$\mathbf{e} \stackrel{\text{def}}{=} \mathbf{z}_2(\omega)\,, \quad \mathbf{e}' \stackrel{\text{def}}{=} s^3\, \mathbf{z}_1(\omega) \mp \sqrt{1 - (s^1)^2}\, \mathbf{z}_3(\omega)\,,$$

therefore, we have

$$\begin{aligned} 1/\epsilon_{\mathrm{c}}^2 &= \mathbf{e} \cdot \left(\overleftrightarrow{\epsilon}_{\mathrm{c}}(\omega)\right)^{-1} \mathbf{e} \\ &= \mathbf{e}' \cdot \left(\overleftrightarrow{\epsilon}_{\mathrm{c}}(\omega)\right)^{-1} \mathbf{e}' \\ &= \frac{(s^3)^2}{\epsilon_{\mathrm{c}}^1} + \frac{1 - (s^3)^2}{\epsilon_{\mathrm{c}}^3}\,. \end{aligned}$$

This implies $\epsilon_{\mathrm{c}}^1 \neq \epsilon_{\mathrm{c}}^3$ (since $\epsilon_{\mathrm{c}}^1 \neq \epsilon_{\mathrm{c}}^2$, by assumption) and (2.74). Conversely, since $\mathbf{e} \cdot \left(\overleftrightarrow{\epsilon}_{\mathrm{c}}(\omega)\right)^{-1} \mathbf{e}' = 0$, it is obvious (by Lemma 2.2.1) that $\mathbf{s}$ corresponds to an optical axis under these conditions. ∎

## Remarks:

1. For the case
$$\epsilon_{\mathrm{c}}^1(\omega) \neq \epsilon_{\mathrm{c}}^2(\omega) = \epsilon_{\mathrm{c}}^3(\omega)\,, \tag{2.75}$$
Lemma 2.2.2 and Corollary 2.2.3 show that only $\pm\mathbf{z}_1(\omega)$ corresponds to an optical axis. Therefore media for which (2.68) and (2.75) hold are called ***uniaxial***.

2. Media with
$$\epsilon_{\mathrm{c}}^1(\omega) \neq \epsilon_{\mathrm{c}}^2(\omega) \neq \epsilon_{\mathrm{c}}^3(\omega) \neq \epsilon_{\mathrm{c}}^1(\omega)$$
are called ***biaxial***. Actually, by Corollary 2.2.3, there are 6 optical axes,[44] but only those corresponding to 'sufficiently real' directions are relevant for physical applications.

3. For biaxial media with
$$\epsilon_{\mathrm{c}}^3(\omega) > \epsilon_{\mathrm{c}}^2(\omega) > \epsilon_{\mathrm{c}}^1(\omega) > 0 \tag{2.76}$$
the real directions $\mathbf{e}$ corresponding to optical axes are given by Corollary 2.2.3 (for the choice of indices corresponding to (2.76)).

4. Crystals of the hexagonal, tetragonal and trigonal system are uniaxial and the optical axis coincides with the crystal axis of six-, four-, or three-fold symmetry. Crystals of the orthorhombic, monoclinic and triclinic system are optically biaxial.



[44] Note that the choice of indices in (2.68) is arbitrary

## 2.3 Monochromatic Light Rays[45]

In this section we consider only isotropic media and — in order to be able to apply standard FOURIER analysis (for tempered distributions) — only the case[46]

$$\boxed{\mathcal{N}(\omega) = n(\omega) > 0\,.}$$

Then, by ***monochromatic light ray*** we mean a solution of MAXWELL's equations fulfilling the following two conditions for suitable $\omega$ and $\mathbf{s} \in \mathbb{R}^3$ with $|\mathbf{s}| = 1$ :

1. $\mathbf{E}(\mathbf{x}, t) = \boldsymbol{\mathcal{E}}(\mathbf{x})\, e^{-i\,\omega\, t} \quad \Big(\text{i.e.}:\ \widetilde{\mathbf{E}}(\mathbf{x}, \omega') = \boldsymbol{\mathcal{E}}(\mathbf{x})\, \delta(\omega' - \omega)\Big).$

2. $\left|\check{\boldsymbol{\mathcal{E}}}(\mathbf{k})\right| \ll 1$ unless $|\mathbf{k}|^2 - (\mathbf{s}\cdot\mathbf{k})^2 \ll |\mathbf{k}|^2$ ,

where

$$\check{\boldsymbol{\mathcal{E}}}(\mathbf{k}) \stackrel{\text{def}}{=} (2\pi)^{-3/2} \int \boldsymbol{\mathcal{E}}(\mathbf{x})\, e^{i\mathbf{x}\cdot\mathbf{k}}\, dV_{\mathbf{x}}\,. \tag{2.77}$$

Let us assume $\mathbf{s} = \mathbf{e}_3$ and define

$$\boldsymbol{\mathcal{F}}(k^1, k^2; x^3) \stackrel{\text{def}}{=} e^{-i\frac{\omega}{c}n(\omega)x^3}\, \frac{1}{2\pi} \int \boldsymbol{\mathcal{E}}(\mathbf{x})\, e^{-i\left(x^1k^1 + x^2k^2\right)}\, dx^1 dx^2\,.$$

Then the HELMHOLTZ equation (2.29) gives[47]

$$\left(-\left(k^1\right)^2 - \left(k^2\right)^2 + \left(\frac{\partial}{\partial x^3}\right)^2 + \left(\frac{\omega}{c}\, n(\omega)\right)^2\right)\left(e^{+i\frac{\omega}{c}n(\omega)x^3}\, \boldsymbol{\mathcal{F}}(k^1, k^2; x^3)\right) = 0\,.$$

Since[48]

$$\begin{aligned}
&\left(\frac{\partial}{\partial x^3}\right)^2 \left(e^{+i\frac{\omega}{c}n(\omega)x^3}\, \boldsymbol{\mathcal{F}}(k^1, k^2; x^3)\right)\\
&= e^{+i\frac{\omega}{c}n(\omega)x^3}\left(-\left(\frac{\omega}{c}n(\omega)\right)^2 + 2\,i\,\frac{\omega}{c}n(\omega)\frac{\partial}{\partial x^3} + \left(\frac{\partial}{\partial x^3}\right)^2\right)\boldsymbol{\mathcal{F}}(k^1, k^2; x^3)\\
&\approx e^{+i\frac{\omega}{c}n(\omega)x^3}\left(-\left(\frac{\omega}{c}n(\omega)\right)^2 + 2\,i\,\frac{\omega}{c}n(\omega)\frac{\partial}{\partial x^3}\right)\boldsymbol{\mathcal{F}}(k^1, k^2; x^3)\,,
\end{aligned}$$

we conclude:

$$\boxed{\left(\left(k^1\right)^2 + \left(k^2\right)^2 - 2\,i\,\frac{\omega}{c}n(\omega)\frac{\partial}{\partial x^3}\right)\boldsymbol{\mathcal{F}}(k^1, k^2; x^3) \approx 0\,.} \tag{2.78}$$

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[45] See also (Gomez-Reino et al., 2002).

[46] Recall (2.30).

[47] Recall that, in the isotropic case, (2.29) implies $\operatorname{div}\widetilde{\mathbf{E}}(\mathbf{x}, \omega) = 0$ .

[48] Thanks to the second defining condition for *light rays*, we may neglect $\left(\frac{\partial}{\partial x^3}\right)^2 \boldsymbol{\mathcal{F}}(k^1, k^2; x^3)$ .

This justifies to approximate $\boldsymbol{\mathcal{F}}(k^1, k^2; x^3)$ by the unique solution[49]

$$\boldsymbol{\mathcal{F}}_{\approx}(k^1, k^2; x^3) = e^{-i \frac{(k^1)^2 + (k^2)^2}{2 \frac{\omega}{c} n(\omega)} x^3} \boldsymbol{\mathcal{F}}(k^1, k^2; 0) \tag{2.79}$$

of the initial-value problem

$$\begin{aligned}
\frac{\partial}{\partial x^3} \boldsymbol{\mathcal{F}}_{\approx}(k^1, k^2; x^3) &= \frac{(k^1)^2 + (k^2)^2}{2\, i\, \frac{\omega}{c} n(\omega)} \boldsymbol{\mathcal{F}}_{\approx}(k^1, k^2; x^3)\,, \\
\boldsymbol{\mathcal{F}}_{\approx}(k^1, k^2; 0) &= \boldsymbol{\mathcal{F}}(k^1, k^2; 0) = \frac{1}{2\pi} \int \boldsymbol{\mathcal{E}}(x^1, x^2, 0)\, e^{-i\left(x^1 k^1 + x^2 k^2\right)}\, \mathrm{d}x^1 \mathrm{d}x^2\,.
\end{aligned}$$

Because of

$$\boldsymbol{\mathcal{E}}(\mathbf{x}) = \frac{1}{2\pi} \int e^{+i \frac{\omega}{c} n(\omega) x^3}\, \boldsymbol{\mathcal{F}}(k^1, k^2; x^3)\, e^{+i\left(x^1 k^1 + x^2 k^2\right)}\, \mathrm{d}k^1\, \mathrm{d}k^2 \tag{2.80}$$

this means:

$$\boxed{\begin{aligned}
\boldsymbol{\mathcal{E}}(\mathbf{x}) \approx (2\pi)^{-2} \int \left( \int \boldsymbol{\mathcal{E}}(x^1, x^2, 0)\, e^{-i\left(x^1 k^1 + x^2 k^2\right)}\, \mathrm{d}x^1 \mathrm{d}x^2 \right) \cdot \\
\cdot\ e^{-i \frac{(k^1)^2 + (k^2)^2}{2 \frac{\omega}{c} n(\omega)} x^3}\, e^{+i\left(x^1 k^1 + x^2 k^2\right)}\, \mathrm{d}k^1 \mathrm{d}k^2\,.
\end{aligned}} \tag{2.81}$$

Hence:

> For monochromatic light rays,[50] $\boldsymbol{\mathcal{E}}(\mathbf{x})$ can be calculated everywhere approximately from its values on any plane that is perpendicular to the propagation direction $\mathbf{s}$.

Especially for (linearly polarized) GAUSS***ian rays***, characterized (up to spatial rotation and translation) by

$$\boldsymbol{\mathcal{E}}(\mathbf{x})_{|_{x^3=0}} = \boldsymbol{\mathcal{E}}_0 \exp\left( - \frac{\left(x^1\right)^2 + \left(x^2\right)^2}{\delta^2} \right) \,, \quad \mathbf{s} = \mathbf{e}_3\,,$$

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[49] We skip the detailed mathematical prove of uniqueness.

[50] We do not elaborate on which profiles $\boldsymbol{\mathcal{E}}(\mathbf{x})_{|_{x^3=0}}$ are actually possible for monochromatic light rays.

(2.81) gives

$$
\begin{aligned}
\boldsymbol{\mathcal{E}}(\mathbf{x}) \;\approx\;& (2\pi)^{-2}\,\boldsymbol{\mathcal{E}}_0 \int \left( \int e^{-\frac{\left(x^1\right)^2+\left(x^2\right)^2}{\delta^2}}\, e^{-i\left(x^1k^1+x^2k^2\right)}\,\mathrm{d}x^1\mathrm{d}x^2 \right)\cdot \\
&\qquad\qquad\qquad\qquad \cdot\, e^{-i\,\frac{(k^1)^2+(k^2)^2}{2\,\frac{\omega}{c}\,n(\omega)}\,x^3}\, e^{+i\left(x^1k^1+x^2k^2\right)}\,\mathrm{d}k^1\mathrm{d}k^2 \\
=\;& \frac{\delta^2}{4\pi}\,\boldsymbol{\mathcal{E}}_0 \int e^{-\left(\left(k^1\right)^2+\left(k^2\right)^2\right)\delta^2/4}\, e^{-i\,\frac{(k^1)^2+(k^2)^2}{2\,\frac{\omega}{c}\,n(\omega)}\,x^3}\, e^{+i\left(x^1k^1+x^2k^2\right)}\,\mathrm{d}k^1\mathrm{d}k^2 \\
=\;& \frac{\delta^2}{4\pi}\,\boldsymbol{\mathcal{E}}_0 \prod_{j=1}^{2} \int \exp\left( -\left(k\,\mathcal{R}(x^3)\right)^2 + i\,x^j\,k \right)\mathrm{d}k \\
=\;& \frac{\delta^2}{4\pi}\,\boldsymbol{\mathcal{E}}_0 \prod_{j=1}^{2} \left( e^{-\left(\frac{x^j}{2\,\mathcal{R}(x^3)}\right)^2} \int \exp\left( -\left( k\,\mathcal{R}(x^3) - i\,\frac{x^j}{2\,\mathcal{R}(x^3)} \right)^2 \right)\mathrm{d}k \right) \\
=\;& \left( \frac{\delta}{2\,\mathcal{R}(x^3)} \right)^2 \exp\left( -\left( \frac{x^1}{2\,\mathcal{R}(x^3)} \right)^2 - \left( \frac{x^2}{2\,\mathcal{R}(x^3)} \right)^2 \right),
\end{aligned}
$$

where

$$
\mathcal{R}(x^3) \stackrel{\text{def}}{=} \sqrt{(\delta/2)^2 + i\,\frac{x^3}{2\,\frac{\omega}{c}n(\omega)}}\,, \quad \Re\left(\mathcal{R}(x^3)\right) > 0\,,
$$

since

$$
\Re\left(\mathcal{R}^2\right) > 0 < \Re(\mathcal{R}) \implies \boxed{\int_{-\infty}^{+\infty} e^{-(\mathcal{R}x+z_0)^2}\,\mathrm{d}x = \frac{\sqrt{\pi}}{\mathcal{R}}} \quad \forall\,\mathcal{R}\,,\ z_0 \in \mathbb{C} \tag{2.82}
$$

(see, e.g., Sect. 2.1.3 of (Lücke, ftm) for a proof of (2.82)) and hence

$$
\frac{1}{\sqrt{2\pi}} \int e^{-x^2}\, e^{-i\,x\,k}\,\mathrm{d}x = \frac{1}{\sqrt{2}}\, e^{-k^2/4} \quad \forall\, k \in \mathbb{R}\,. \tag{2.83}
$$

## 2.4 Group-Velocity Dispersion

See (Reider, 1997, Sect. 3.2.1).

# Chapter 3

# Nonlinear Optical Media

## 3.1 General Considerations

If, in Exercise 1, the damped harmonic oscillator is replaced by an anharmonic one then its evolution depends nonlinearly on the driving force. As a consequence — even if the driving force is monochromatic — higher harmonics show up in the motion of the oscillator; mainly second order harmonics if the medium is not inversion symmetric[1] (compare (Reider, 1997, Sect. 8.1)). Moreover, if the driving force is a superposition of two monochromatic ones, the motion of the oscillator contains harmonic components corresponding to the sum as well as to the difference of both frequencies.[2]

If the damped harmonic oscillator model is applied to the electric dipoles of an optical medium then the accelerated charge of the dipoles create a radiation field acting on the other charges in addition. Therefore, actually, the driving field is a superposition of the incident exterior field with the radiation emitted from the medium's electric dipoles. Now, if the harmonic oscillator is replaced by a more realistic nonlinear one, then the radiation contributed by the medium itself (to the macroscopic electromagnetic field) contains higher harmonics of every monochromatic component of the incident (microscopic) field as well as components with frequencies corresponding to the sum or difference of any two frequencies present in the incident field.

Usually, very strong electromagnetic fields — as provided by laser pulses of high intensity — or long optical path length's in low-loss nonlinear optical media (optical fibers, see (Agrawal, 1999, Sect. 1.3.3)) — have to be applied in order to gain recognizable nonlinear effects. Then, however,

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[1] Since, e.g., silicia glasses — the material of choice for low-loss optical fibers, formed by fusing $SiO_2$ molecules — are inversion symmetric they do not normally exhibit second-order nonlinear effects.

[2] See (Mills, 1998, Sect. 3.1) for a perturbative calculation.

interesting phenomena show up several of which will be studied in this chapter.

### 3.1.1 Functionals of the Electric Field

Usually, especially in optical applications, $\boldsymbol{\mathcal{P}}(\mathbf{x},t)$ is assumed to be a (sufficiently well-behaved) functional of $\mathbf{E}$. Then[3] we have the functional TAYLOR expansion[4]

$$\mathcal{P}_j(\mathbf{x},t) = \sum_{\nu=0}^{\infty} \mathcal{P}_j^{(\nu)}(\mathbf{x},t)\,, \tag{3.1}$$

$$\begin{aligned}\mathcal{P}_j^{(\nu)}(\mathbf{x},t) &= \frac{\epsilon_0}{(2\pi)^{\frac{\nu}{2}}} \int \check{\chi}^{(\nu)}_{jk_1\ldots k_\nu}(\mathbf{x},t;\mathbf{x'}_1,t'_1;\ldots;\mathbf{x'}_\nu,t'_\nu)\, E^{k_1}(\mathbf{x'}_1,t'_1)\ldots \\ &\qquad \ldots E^{k_\nu}(\mathbf{x'}_\nu,t'_\nu)\,\mathrm{d}V_{\mathbf{x'}_1}\,\mathrm{d}t'_1\ldots \mathrm{d}V_{\mathbf{x'}_\nu}\,\mathrm{d}t'_\nu\,,\end{aligned} \tag{3.2}$$

w.r.t. an arbitrary Basis $\{\mathbf{b}_1,\mathbf{b}_2,\mathbf{b}_3\}$ of $\mathbb{R}$, where[5]

$$\mathcal{P}_j(\mathbf{x},t) \stackrel{\text{def}}{=} \mathbf{b}_j\cdot\boldsymbol{\mathcal{P}}(\mathbf{x},t)\,,\quad E^k(\mathbf{x}',t') \stackrel{\text{def}}{=} \mathbf{b}^k\cdot\mathbf{E}(\mathbf{x},t)\,.$$

Note that the coefficients $\check{\chi}^{(\nu)}$ fulfill the symmetry condition[6]

$$\check{\chi}^{(\nu)}_{jk_1\ldots k_\nu}(\mathbf{x},t;\mathbf{x'}_1,t'_1;\ldots;\mathbf{x'}_\nu,t'_\nu) = \check{\chi}^{(\nu)}_{jk_{\check{\pi}(1)}\ldots k_{\check{\pi}(\nu)}}(\mathbf{x},t;\mathbf{x'}_{\check{\pi}(1)},t'_{\check{\pi}(1)};\ldots;\mathbf{x'}_{\check{\pi}(\nu)},t'_{\check{\pi}(\nu)}) \tag{3.3}$$

for every permutation $\check{\pi}$ of $(1,\ldots,\nu)$. If, in addition, we assume homogeneity in space and time these coefficients have to be of the form

$$\check{\chi}^{(\nu)}_{jk_1\ldots k_\nu}(\mathbf{x},t;\mathbf{x'}_1,t'_1;\ldots;\mathbf{x'}_\nu,t'_\nu) = \check{\chi}^{(\nu)}_{jk_1\ldots k_\nu}(\mathbf{x}-\mathbf{x'}_1,t-t'_1;\ldots;\mathbf{x}-\mathbf{x'}_\nu,t-t'_\nu)\,.$$

Assuming, moreover, spatial nonlocality of the electric field contributions to be negligible we have

$$\check{\chi}^{(\nu)}_{jk_1\ldots k_\nu}(\mathbf{x},t;\mathbf{x'}_1,t'_1;\ldots;\mathbf{x'}_\nu,t'_\nu) = \check{\chi}^{(\nu)}_{jk_1\ldots k_\nu}(t-t'_1,\ldots,t-t'_\nu)\,\delta(\mathbf{x}-\mathbf{x'}_1)\ldots\delta(\mathbf{x}-\mathbf{x'}_\nu)\,.$$

Thus,

$$\begin{aligned}&\frac{(2\pi)^{\frac{\nu-1}{2}}}{\epsilon_0}\,\mathcal{P}_j^{(\nu)}(\mathbf{x},t) \\ &= \frac{1}{\sqrt{2\pi}}\int \chi^{(\nu)}_{jk_1\ldots k_\nu}(\omega_1,\ldots,\omega_\nu)\,\widetilde{E}^{k_1}(\mathbf{x},\omega_1)\ldots\widetilde{E}^{k_\nu}(\mathbf{x},\omega_\nu)\,e^{-i(\omega_1+\ldots+\omega_\nu)t}\,\mathrm{d}\omega_1\ldots\mathrm{d}\omega_\nu\end{aligned} \tag{3.4}$$

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[3]See Appendix A.1.

[4]We use EINSTEIN's summation convention meaning, e.g., $\chi_{jk}\,E^k \equiv \sum_{k=1}^{3}\chi_{jk}\,E^k$.

[5]Recall that the ***reciprocal basis*** $\{\mathbf{b}^1,\mathbf{b}^2,\mathbf{b}^3\}$ is characterized by:

$$\mathbf{b}_j\cdot\mathbf{b}^k = \begin{cases}1 & \text{for } j=k\,,\\ 0 & \text{else}\,.\end{cases}$$

[6]Recall (A.1).

holds for the (generalized) ***susceptibilities***

$$\chi^{(\nu)}_{jk_1\ldots k_\nu}(\omega_1,\ldots,\omega_\nu) \stackrel{\text{def}}{=} (2\pi)^{-\frac{n}{2}} \int \check{\chi}^{(\nu)}_{jk_1\ldots k_\nu}(t_1,\ldots,t_\nu)\, e^{i(\omega_1 t_1+\ldots+\omega_\nu t_\nu)}\, \mathrm{d}t_1\ldots\mathrm{d}t_\nu$$

and (3.3) is equivalent to

$$\boxed{\chi^{(\nu)}_{jk_1\ldots k_\nu}(\omega_1,\ldots,\omega_\nu) = \chi^{(\nu)}_{jk_{\check\pi(1)}\ldots k_{\check\pi(\nu)}}(\omega_{\check\pi(1)},\ldots,\omega_{\check\pi(\nu)})\,.} \tag{3.5}$$

The FOURIER transform of (3.4) is[7]

$$\boxed{\begin{aligned}&\frac{(2\pi)^{\frac{\nu-1}{2}}}{\epsilon_0}\,\widetilde{\mathcal{P}}^{(\nu)}_j(\mathbf{x},\omega)\\ &= \int \chi^{(\nu)}_{jk_1\ldots k_\nu}(\omega_1,\ldots,\omega_\nu)\,\widetilde{E}^{k_1}(\mathbf{x},\omega_1)\ldots\widetilde{E}^{k_\nu}(\mathbf{x},\omega_\nu)\,\delta(\omega_1+\ldots+\omega_\nu-\omega)\,\mathrm{d}\omega_1\ldots\mathrm{d}\omega_\nu\,.\end{aligned}} \tag{3.6}$$

An optical medium modeled by (3.6) is called a ***linear optical medium*** if all the ***nonlinear susceptibilities*** $\chi^{(\nu)}_{jk_1\ldots k_\nu}(\omega_1,\ldots,\omega_\nu)\,,\ \nu>1\,,$ vanish.[8]

Usually,

$$\boldsymbol{\mathcal{P}}(\mathbf{x},t) = \boldsymbol{\mathcal{P}}_{\text{el}}(\mathbf{x},t)$$

is assumed for optical media and the susceptibilities should be caculated by (time-dependent) quantum mechnical perturbation theory.[9] For the tensor $\chi^{(1)}_{jk}(\omega)$ of the **linear susceptibility** first order perturbation theory is sufficient.

### 3.1.2 Various Electric Polarization Effects

See (Römer, 1994, Chapter 5).

### 3.1.3 Perturbative Solution of MAXWELL's Equations

If we replace $\boldsymbol{\mathcal{P}}$ by $\boldsymbol{\mathcal{P}}^{(1)}$ in (2.1) and $\overset{\leftrightarrow}{\chi}$ by $\overset{\leftrightarrow}{\chi}^{(1)}$ in the definition of the permittivity $\overset{\leftrightarrow}{\epsilon}$ then the FOURIER transformed MAXWELL equations (1.18)–(1.21) become equivalent to:

$$\boxed{\operatorname{curl}\widetilde{\mathbf{H}}(\mathbf{x},\omega) = -i\,\omega\,\epsilon_0\,\overset{\leftrightarrow}{\epsilon}(\omega)\,\widetilde{\mathbf{E}}(\mathbf{x},\omega) + \widetilde{\boldsymbol{\jmath}}_{\text{ex}}(\mathbf{x},\omega) - i\,\omega\,\widetilde{\boldsymbol{\mathcal{P}}}^{\text{nl}}(\mathbf{x},\omega)\,,} \tag{3.7}$$

$$\operatorname{curl}\widetilde{\mathbf{B}}(\mathbf{x},\omega) = i\,\omega\,\widetilde{\mathbf{H}}(\mathbf{x},\omega)\,, \tag{3.8}$$

$$\boxed{\operatorname{div}\left(\epsilon_0\,\overset{\leftrightarrow}{\epsilon}(\omega)\,\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right) = \widetilde{\rho}_{\text{ex}}(\mathbf{x},\omega) - \operatorname{div}\left(\widetilde{\boldsymbol{\mathcal{P}}}^{\text{nl}}(\mathbf{x},\omega)\right),} \tag{3.9}$$

$$\operatorname{div}\widetilde{\mathbf{B}}(\mathbf{x},\omega) = 0\,, \tag{3.10}$$



[7]For a listing of the dependence of the lowest nonlinear susceptibilities on the symmetry classes for crystals see (Brunner and Junge, 1982, Sect. 3.2.2.1).

[8]Note that nonlinearity does not necessarily mean that $\chi^{(\nu)}_{jk_1\ldots k_\nu}(\omega,\ldots,\omega)\neq 0$ for some $\nu>1$.

[9]See (Armstrong et al., 1962a).

where

$$\widetilde{\boldsymbol{\mathcal{P}}}^{\mathrm{nl}}(\mathbf{x},\omega) \stackrel{\text{def}}{=} \widetilde{\boldsymbol{\mathcal{P}}}(\mathbf{x},\omega) - \widetilde{\mathcal{P}}_j^{(1)}(\mathbf{x},\omega)\,\mathbf{b}^j\,.$$

Now we have to replace (2.12) by the nonlinear equation[10]

$$\boxed{\boxed{\hat{H}\widetilde{\mathbf{E}}(\mathbf{x},\omega) = -i\,\omega\,\mu_0\,\widetilde{\boldsymbol{\jmath}}_{\mathrm{cr}}(\mathbf{x},\omega) - \omega^2\mu_0\,\widetilde{\boldsymbol{\mathcal{P}}}^{\mathrm{nl}}\Big(\mathbf{x},\omega;\widetilde{\boldsymbol{E}}(\mathbf{x},.)\Big)}} \tag{3.11}$$

in which we have written $\widetilde{\boldsymbol{\mathcal{P}}}^{\mathrm{nl}}\Big(\mathbf{x},\omega;\widetilde{\boldsymbol{E}}(\mathbf{x},.)\Big)$ instead of $\widetilde{\boldsymbol{\mathcal{P}}}^{\mathrm{nl}}(\mathbf{x},\omega)$ in order to indicate the functional (nonlinear) dependence on $\widetilde{\boldsymbol{E}}(\mathbf{x},.)$, determined by the nonlinear susceptibilities, and where

$$\boxed{\boxed{\hat{H}\,\widetilde{\mathbf{E}}(\mathbf{x},\omega) \stackrel{\text{def}}{=} \left(\Delta_{\mathbf{x}} + \left(\frac{\omega}{c}\right)^2 \overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\right)\widetilde{\mathbf{E}}(\mathbf{x},\omega) - \operatorname{grad}\left(\operatorname{div}\widetilde{\mathbf{E}}(\mathbf{x},\omega)\right)\,.}} \tag{3.12}$$

Motivated by the results of 2.1.3, let us assume that there is a unique (matrix-valued, restarted) response function $\overset{\leftrightarrow}{\check{r}}(\mathbf{x},t)$ of the corresponding linear medium such that

$$\boldsymbol{\mathcal{R}}(\mathbf{x},t;\boldsymbol{\jmath}) \stackrel{\text{def}}{=} \frac{1}{\sqrt{2\pi}}\int \overset{\leftrightarrow}{\check{r}}(\mathbf{x}-\mathbf{x}',t-t')\,\boldsymbol{\jmath}(\mathbf{x}',t')\,\mathrm{d}V_{\mathbf{x}'}\,\mathrm{d}t' \tag{3.13}$$

fulfills the conditions

$$\begin{aligned} &\boldsymbol{\jmath}(\mathbf{x},t) = 0 \quad \text{for } \ (|\mathbf{x}|\,,t) \notin [0,+R]\times[0,+\infty) \\ &\Longrightarrow \quad \boldsymbol{\mathcal{R}}(\mathbf{x},t;\boldsymbol{\jmath}) = 0 \quad \text{for } t \notin \left[0,\tfrac{|\mathbf{x}|-R}{c}\right] \end{aligned}$$

and

$$\hat{H}\,\widetilde{\boldsymbol{\mathcal{R}}}(\mathbf{x},\omega;\widetilde{\boldsymbol{\jmath}}) = -i\,\omega\,\mu_0\,\widetilde{\boldsymbol{\jmath}}(\mathbf{x},\omega) \tag{3.14}$$

for all sufficiently well-behaved $\boldsymbol{\jmath}$, where

$$\begin{aligned} \widetilde{\boldsymbol{\mathcal{R}}}(\mathbf{x},\omega;\widetilde{\boldsymbol{\jmath}}) &\stackrel{\text{def}}{=} \frac{1}{\sqrt{2\pi}}\int \boldsymbol{\mathcal{R}}(\mathbf{x},t;\boldsymbol{\jmath})\,e^{+i\,\omega\,t}\,\mathrm{d}t \\ &= \int \overset{\leftrightarrow}{\check{r}}(\mathbf{x}-\mathbf{x}',\omega)\,\widetilde{\boldsymbol{\jmath}}(\mathbf{x}',\omega)\,\mathrm{d}V_{\mathbf{x}'}\,. \end{aligned} \tag{3.15}$$

Let us only consider the case

$$\boxed{\boxed{\widetilde{\boldsymbol{\jmath}}_{\mathrm{cr}} = 0\,.}}$$

Then $\widetilde{\mathbf{E}}(\mathbf{x},\omega)$ is a solution of (3.11) iff the linear equation

$$\hat{H}\,\widetilde{\mathbf{E}}_0(\mathbf{x},\omega) = 0\,. \tag{3.16}$$



[10] Again, (3.9) serves as a definition of $\widetilde{\rho}_{\mathrm{ex}}(\mathbf{x},\omega)$ for $\omega \neq 0$.

is fulfilled for

$$\widetilde{\mathbf{E}}_0(\mathbf{x},\omega) = \widetilde{\mathbf{E}}(\mathbf{x},\omega) - \widetilde{\boldsymbol{\mathcal{R}}}\Big(\mathbf{x},\omega;\widetilde{\boldsymbol{\jmath}}_{\widetilde{\boldsymbol{E}}}\Big)\,,$$

where

$$\widetilde{\boldsymbol{\jmath}}_{\widetilde{\boldsymbol{E}}}(\mathbf{x},\omega) \stackrel{\text{def}}{=} -i\omega\,\widetilde{\boldsymbol{\mathcal{P}}}^{\text{nl}}\Big(\mathbf{x},\omega;\widetilde{\boldsymbol{E}}(\mathbf{x},.)\Big)\,. \tag{3.17}$$

This suggests the following perturbative solution of (3.11):

> Take a solution $\widetilde{\mathbf{E}}_0(\mathbf{x},\omega)$ of (3.16) and define recursively
>
> $$\widetilde{\mathbf{E}}_\nu(\mathbf{x},\omega) \stackrel{\text{def}}{=} \widetilde{\mathbf{E}}_0(\mathbf{x},\omega) + \widetilde{\boldsymbol{\mathcal{R}}}\Big(\mathbf{x},\omega;\widetilde{\boldsymbol{\jmath}}_{\widetilde{\boldsymbol{E}}_{\nu-1}}\Big) \quad \text{for } \nu = 1,2,3,\ldots$$
>
> (compare Section 5.1.3 von (Lücke, ein)). Then
>
> $$\widetilde{\mathbf{E}}(\mathbf{x},\omega) = \lim_{\nu\to\infty} \widetilde{\mathbf{E}}_\nu(\mathbf{x},\omega)\,,$$
>
> **if** the limit exists (in a suitable topology), is a solution of (3.11).

Certainly, one will try to get along with the first order approximation

$$\boxed{\widetilde{\mathbf{E}}(\mathbf{x},\omega) \approx \widetilde{\mathbf{E}}_0(\mathbf{x},\omega) - i\omega \int \overset{\leftrightarrow}{r}(\mathbf{x}-\mathbf{x}',\omega)\,\widetilde{\boldsymbol{\mathcal{P}}}^{\text{nl}}\Big(\mathbf{x}',\omega;\widetilde{\boldsymbol{E}}_0(\mathbf{x}',.)\Big)\,dV_{\mathbf{x}'}} \tag{3.18}$$

(for properly chosen $\widetilde{\mathbf{E}}_0(\mathbf{x},\omega)$) as far as possible. Note that, by (3.11)–(3.13), we have

$$\hat{H}\,\widetilde{\mathbf{E}}(\mathbf{x},\omega) = -\omega^2\mu_0\,\widetilde{\boldsymbol{\mathcal{P}}}^{\text{nl}}\Big(\mathbf{x},\omega;\widetilde{\boldsymbol{E}}_0(\mathbf{x},.)\Big) \tag{3.19}$$

in this approximation.

## 3.2 Classical Nonlinear Optical Effects[11]

### 3.2.1 Phase Matching

Let us discuss the approximations (3.18) and

$$\widetilde{\mathcal{P}}_j^{\text{nl}}(\mathbf{x},\omega) \approx \widetilde{\mathcal{P}}_j^{(2)}(\mathbf{x},\omega) \underset{(3.6)}{=} \int \chi^{(2)}_{jkl}(\omega',\omega-\omega')\,\widetilde{E}^{\,k}(\mathbf{x},\omega')\,\widetilde{E}^{\,l}(\mathbf{x},\omega-\omega')\,d\omega'\,. \tag{3.20}$$

By (3.19), then, we have to expect that

$$\left.\begin{array}{l} \omega = \omega_1 + \omega_2\,, \\ \chi^{(2)}_{jkl}(\omega_1,\omega_2)\,\widetilde{E}^k_0(\mathbf{x},\omega_1)\,\widetilde{E}^l_0(\mathbf{x},\omega_1) \neq 0 \end{array}\right\} \text{for suitable } \omega_1,\omega_1 \in \mathbb{R}$$
$$\Longrightarrow\ \widetilde{\mathbf{E}}(\mathbf{x},\omega) \neq 0$$



[11] A freeware program for calculating nonlinear frequency conversion processes is offered at: www.sandia.gov/imrl/XWEB1128/xxtal.htm

and hence

$$\left(\widetilde{\boldsymbol{E}}_0(\mathbf{x},\omega_1) \neq 0 \neq \widetilde{\boldsymbol{E}}_0(\mathbf{x},\omega_2) \Longrightarrow \widetilde{\boldsymbol{E}}(\mathbf{x},\omega_1+\omega_2) \neq 0\right) \; \forall \omega_1,\omega_2 \in \mathbb{R}$$

holds for sufficiently nontrivial[12] $\chi^{(2)}$. Especially for $\omega_1 = \omega_2$ this means ***second harmonic generation***. Of course, the crucial question is how strong these effects are.

Actually, as can be explained by simple models, MILLER***'s rule***[13]

$$\boxed{\Delta_{jkl}(\omega_1,\omega_2) \stackrel{\text{def}}{=} \frac{\chi^{(2)}_{jkl}(\omega_1,\omega_2)}{\chi^{(1)}_{jj}(\omega_1+\omega_2)\,\chi^{(1)}_{kk}(\omega_1)\,\chi^{(1)}_{ll}(\omega_2)} \approx \text{independent of } \omega_1,\omega_2\,,} \tag{3.21}$$

w.r.t. the principal directions in the medium, is valid for a large variety of crystals. Therefore the dispersion relations[14] for the linear susceptibilities $\chi^{(1)}_{jk}$, qualitatively described by Exercise 1, are of great importance also for the second order susceptibilities.

Let us analyze (3.18)/(3.19) for the special case that the solution $\widetilde{\mathbf{E}}_0(\mathbf{x},\omega)$ of (3.16) is of the form[15]

$$\begin{aligned} \widetilde{\mathbf{E}}_0(\mathbf{x},\omega) &= \widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{x},\omega)\left(\delta(\omega_1-\omega)+\delta(\omega_2-\omega)\right) \\ &\quad + \overline{\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{x},-\omega)}\left(\delta(\omega_1+\omega)+\delta(\omega_2+\omega)\right). \end{aligned} \tag{3.22}$$

Then we have

$$\begin{aligned} &\widetilde{E}^k_0(\mathbf{x},\omega')\,\widetilde{E}^l_0(\mathbf{x},\omega-\omega') \\ &= \widetilde{\mathcal{E}}^k_0(\mathbf{x},\omega')\,\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega-\omega')\left(\delta(\omega_1-\omega')+\delta(\omega_2-\omega')\right)\left(\delta(\omega_1-\omega+\omega')+\delta(\omega_2-\omega+\omega')\right) \\ &\quad + \widetilde{\mathcal{E}}^k_0(\mathbf{x},\omega')\,\overline{\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega'-\omega)}\left(\delta(\omega_1-\omega')+\delta(\omega_2-\omega')\right)\left(\delta(\omega_1+\omega-\omega')+\delta(\omega_2+\omega-\omega')\right) \\ &\quad + \overline{\widetilde{\mathcal{E}}^k_0(\mathbf{x},-\omega')}\,\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega-\omega')\left(\delta(\omega_1+\omega')+\delta(\omega_2+\omega')\right)\left(\delta(\omega_1-\omega+\omega')+\delta(\omega_2-\omega+\omega')\right) \\ &\quad + \overline{\widetilde{\mathcal{E}}^k_0(\mathbf{x},-\omega')}\,\overline{\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega'-\omega)}\left(\delta(\omega_1+\omega')+\delta(\omega_2+\omega')\right)\left(\delta(\omega_1+\omega-\omega')+\delta(\omega_2+\omega-\omega')\right). \end{aligned}$$

In the approximation (3.20), this together with

$$\chi^{(2)}_{jkl}(-\omega'_1,-\omega'_2) = \overline{\chi^{(2)}_{jkl}(\omega'_1,\omega'_2)}$$

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[12]For inversion-symmetric media $\chi^{(2)}$ must vanish, because both $\mathcal{P}^{(2)}_j$ and $\widetilde{E}^l$ change sign under total spatial reflection.

[13]See (Miller, 1964). Obviously, this rule is consistent with the generally valid relations

$$\chi^{(2)}_{jlk}(\omega_2,\omega_1) = \chi^{(2)}_{jkl}(\omega_1,\omega_2) = \overline{\chi^{(2)}_{jkl}(-\omega_1,-\omega_2)}\,, \quad \chi^{(1)}_{jl}(-\omega) = \overline{\chi^{(1)}_{jl}(\omega)}\,.$$

[14]Recall the remark on page 37 of Chapter 2.

[15]We shall see, however, that such a choice may be inappropriate for first order approximation.

gives

$$\widetilde{\boldsymbol{\mathcal{P}}}^{\rm nl}\Big(\mathbf{x},\omega;\widetilde{\boldsymbol{E}}_0(\mathbf{x},.)\Big) = \widetilde{\boldsymbol{\mathcal{P}}}_0(\mathbf{x},\omega) + \overline{\widetilde{\boldsymbol{\mathcal{P}}}_0(\mathbf{x},-\omega)}$$

where[16]

$$\begin{aligned}\widetilde{\boldsymbol{\mathcal{P}}}_0(\mathbf{x},\omega) \;\stackrel{\rm def}{=}\; & 2\,\delta(\omega_1+\omega_2-\omega)\,\chi^{(2)}_{jkl}(\omega_1,\omega_2)\,\widetilde{\mathcal{E}}^k_0(\mathbf{x},\omega_1)\,\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega_2)\\ & +\delta(2\,\omega_1-\omega)\,\chi^{(2)}_{jkl}(\omega_1,\omega_1)\,\widetilde{\mathcal{E}}^k_0(\mathbf{x},\omega_1)\,\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega_1)\\ & +\delta(2\,\omega_2-\omega)\,\chi^{(2)}_{jkl}(\omega_2,\omega_2)\,\widetilde{\mathcal{E}}^k_0(\mathbf{x},\omega_2)\,\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega_2)\\ & +2\,\delta(\omega_1-\omega_2-\omega)\,\chi^{(2)}_{jkl}(-\omega_2,\omega_1)\,\overline{\widetilde{\mathcal{E}}^k_0(\mathbf{x},\omega_2)}\,\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega_1)\\ & +\delta(\omega)\Big(\chi^{(2)}_{jkl}(\omega_1,-\omega_1)\,\widetilde{\mathcal{E}}^k_0(\mathbf{x},\omega_1)\,\overline{\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega_1)}\\ & \qquad\qquad +\chi^{(2)}_{jkl}(\omega_2,-\omega_2)\,\widetilde{\mathcal{E}}^k_0(\mathbf{x},\omega_2)\,\overline{\widetilde{\mathcal{E}}^l_0(\mathbf{x},\omega_2)}\Big)\,.\end{aligned}$$

Therefore, in the approximation (3.18) $\widetilde{\mathbf{E}}(\mathbf{x},\omega)$ is of the form

$$\widetilde{\mathbf{E}}(\mathbf{x},\omega) = \widetilde{\mathbf{E}}_0(\mathbf{x},\omega) + \sum_{\omega'\in M_{\omega_1,\omega_2}} \widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega)\,\delta(\omega-\omega')\,,$$

where

$$M_{\omega_1,\omega_2} \stackrel{\rm def}{=} \{\omega_1+\omega_2, 2\,\omega_1, 2\,\omega_2, \omega_1-\omega_2, -\omega_1-\omega_2, -2\,\omega_1, -2\,\omega_2, \omega_2-\omega_1\}\ .$$

The static part of $\widetilde{\boldsymbol{\mathcal{P}}}^{\rm nl}\Big(\mathbf{x},\omega;\widetilde{\boldsymbol{E}}_0(\mathbf{x},.)\Big)$ (***optical rectification***[17]) does not show up in (3.18) because of the $\omega$-factor.[18] Let us assume, for simplicity, that

$$\{\omega_1,\omega_2\}\cap M_{\omega_1,\omega_2} = \emptyset\,.$$

Then special consequences of (3.19) are the equation

$$\boxed{\hat H\,\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},2\,\omega_1) = -\,(2\,\omega_1)^2\,\mu_0\,\chi^{(2)}_{jkl}(\omega_1,\omega_1)\,\widetilde{\mathcal{E}}^k(\mathbf{x},\omega_1)\,\widetilde{\mathcal{E}}^l(\mathbf{x},\omega_1)\,\mathbf{b}^j} \tag{3.23}$$

for ***second harmonic generation***, and the equation

$$\boxed{\hat H\,\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega_1+\omega_2) = -2\,(\omega_1+\omega_2)^2\,\mu_0\,\chi^{(2)}_{jkl}(\omega_1,\omega_2)\,\widetilde{\mathcal{E}}^k(\mathbf{x},\omega_1)\,\widetilde{\mathcal{E}}^l(\mathbf{x},\omega_2)\,\mathbf{b}^j} \tag{3.24}$$

for ***sum-frequency generation***, where we have set

$$\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega_1) = \widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{x},\omega_1) \quad \text{for } j=1,2\,. \tag{3.25}$$



[16]Recall Footnote 13.

[17]See, e.g., (Shen, 1984, Sect. 5.1) for a discussion of this effect.

[18]But it does contribute to $\widetilde{\boldsymbol{E}}_0(\mathbf{x},0)$. Note that $\hat H\,\widetilde{\boldsymbol{E}}(\mathbf{x},0)=0$ does **not** imply $\widetilde{\rho}_{\rm ex}(\mathbf{x},0)=0$.

More precisely, (3.24) is a consequence of

$$\begin{aligned}&\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega_1+\omega_2)\\&=-i\,(\omega_1+\omega_2)\int \overset{\leftrightarrow}{r}(\mathbf{x}-\mathbf{x}',\omega_1+\omega_2)\left(2\,\chi^{(2)}_{jkl}(\omega_1,\omega_2)\,\widetilde{\mathcal{E}}^k(\mathbf{x}',\omega_1)\,\widetilde{\mathcal{E}}^l(\mathbf{x}',\omega_2)\,\mathbf{b}^j\right)\mathrm{d}V_{\mathbf{x}'}\end{aligned}\tag{3.26}$$

in very the same way as (3.19) is a consequence of (3.18).

Let us specialize further to[19]

$$\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{x},\omega)=\mathcal{E}_{\mathbf{e}_1}(\omega)\,\exp\left(i\,\frac{\omega}{c}\,\mathcal{N}_{\mathbf{e}_1}(\omega)\,\mathbf{e}_1\cdot\mathbf{x}\right)\quad\forall\,\omega\in\{\omega_1,\omega_2\}\tag{3.27}$$

and

$$\{\mathbf{b}_1,\mathbf{b}_2,\mathbf{b}_3\}=\{\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3\}\quad\text{right-handed orthonormal basis of }\mathbb{R}^3\,.$$

Then (3.25) and (3.26) imply

$$\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega)=\widetilde{\boldsymbol{\mathcal{E}}}\left(x^1,\omega\right)\quad\forall\,\omega\in\{\omega_1,\omega_2,\omega_1+\omega_2\}\tag{3.28}$$

and therefore

$$\Delta_{\mathbf{x}}\,\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega)-\boldsymbol{\nabla}_{\mathbf{x}}\left(\boldsymbol{\nabla}_{\mathbf{x}}\cdot\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega)\right)=\left(\frac{\partial}{\partial x^1}\right)^2\hat{P}_{\perp\mathbf{e}_1}\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega)\tag{3.29}$$

holds for $\omega\in\{\omega_1,\omega_2,\omega_1+\omega_2\}$. Especially for $\omega=\omega_1+\omega_2$ this means, according to the definition of $\hat{H}$ (Equation (3.12)), that (3.24) is equivalent to

$$\begin{aligned}&\epsilon_0\left(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega_1+\omega_2)\,\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega_1+\omega_2)+\left(\frac{\partial}{\partial x^1}\right)^2\hat{P}_{\perp\mathbf{e}_1}\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega_1+\omega_2)\right)\\&=-2\,\chi^{(2)}_{jkl}(\omega_1,\omega_2)\,\widetilde{\mathcal{E}}^k(\mathbf{x},\omega_1)\,\widetilde{\mathcal{E}}^l(\mathbf{x},\omega_2)\,\mathbf{e}_j\,,\end{aligned}$$

i.e. to the equations

$$\left(\epsilon_0\,\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega_1+\omega_2)\,\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega_1+\omega_2)\right)\cdot\mathbf{e}_1=-2\,\chi^{(2)}_{1kl}(\omega_1,\omega_2)\,\widetilde{\mathcal{E}}^k(\mathbf{x},\omega_1)\,\widetilde{\mathcal{E}}^l(\mathbf{x},\omega_2)$$

and

$$\begin{aligned}&\left(\epsilon_0\,\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega_1+\omega_2)\,\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega_1+\omega_2)\right)\cdot\mathbf{e}_j+\epsilon_0\left(\frac{\partial}{\partial x^1}\right)^2\widetilde{\mathcal{E}}^j(\mathbf{x},\omega_1+\omega_2)\\&=-2\,\chi^{(2)}_{jkl}(\omega_1,\omega_2)\,\widetilde{\mathcal{E}}^k(\mathbf{x},\omega_1)\,\widetilde{\mathcal{E}}^l(\mathbf{x},\omega_2)\quad\text{for }j=2,3\,.\end{aligned}\tag{3.30}$$

For simplicity, let us assume that the medium is uniaxial with axis along $\mathbf{e}_1$. Then

$$\begin{aligned}\mathcal{N}_{\mathbf{e}_1}(\omega)=\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega)&\underset{(2.69)}{=}\sqrt{\epsilon^2_{\mathrm{c}}(\omega)}\,,\\\left(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega)\,\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega)\right)\cdot\mathbf{e}_j&=\epsilon^2_{\mathrm{c}}(\omega)\,\widetilde{\mathcal{E}}^j(\mathbf{x},\omega)\quad\text{for }j=2,3\end{aligned}\tag{3.31}$$



[19]Compare with (2.30).

and, corresponding to (3.27), the ***slowly varying amplitude approximation***[20]

$$\left|\left(\frac{\partial}{\partial x^1}\right)^2 \mathcal{A}(x^1,\omega)\right| \ll \left|\frac{\omega}{c}\,\mathcal{N}_{\mathbf{e}_1}(\omega)\frac{\partial}{\partial x^1}\,\mathcal{A}(x^1,\omega)\right| \tag{3.32}$$

should be applicable to

$$\mathcal{A}(x^1,\omega) \stackrel{\text{def}}{=} \exp\left(-i\,\frac{\omega}{c}\,\mathcal{N}_{\mathbf{e}_1}(\omega)\,\mathbf{e}_1\cdot\mathbf{x}\right)\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega) \tag{3.33}$$

for $\omega = \omega_1 + \omega_2$ . (3.30)–(3.33) imply

$$\frac{\partial}{\partial x^1}\mathcal{A}^j(x^1,\omega_1+\omega_2) = \mathcal{C}^j\,e^{-i\,\Delta\mathcal{K}\,x^1} \quad \text{for } j = 2,3\ , \tag{3.34}$$

where

$$\mathcal{C}^j \stackrel{\text{def}}{=} i\,c\,\frac{\omega_1+\omega_2}{\mathcal{N}_{\mathbf{e}_1}(\omega_1+\omega_2)}\,\mu_0\,\chi^{(2)}_{jkl}(\omega_1+\omega_2)\,\mathcal{E}^k_{\mathbf{e}_1}(\omega_1)\,\mathcal{E}^l_{\mathbf{e}_1}(\omega_2)$$

and

$$\Delta\mathcal{K} \stackrel{\text{def}}{=} \frac{\omega_1+\omega_2}{c}\,\mathcal{N}_{\mathbf{e}_1}(\omega_1+\omega_2) - \frac{\omega_1}{c}\,\mathcal{N}_{\mathbf{e}_1}(\omega_1) - \frac{\omega_2}{c}\,\mathcal{N}_{\mathbf{e}_1}(\omega_2)\,.$$

The general solution of (3.34) is[21]

$$\begin{aligned}\mathcal{A}^j(x^1,\omega_1+\omega_2) &= \frac{i}{\Delta\mathcal{K}}\,\mathcal{C}^j\left(e^{-i\,\Delta\mathcal{K}\,x^1}-1\right)+\hat{\mathcal{C}}^j\\ &= x^1\,\mathcal{C}^j\,e^{-\frac{i}{2}\,\Delta\mathcal{K}\,x^1}\,\frac{\sin\left(\frac{1}{2}\,\Delta\mathcal{K}\,x^1\right)}{\frac{1}{2}\,\Delta\mathcal{K}\,x^1}+\hat{\mathcal{C}}^j\,.\end{aligned} \tag{3.35}$$

with arbitrary $\hat{\mathcal{C}}^j = \hat{\mathcal{C}}^j(\omega_1+\omega_2)$ . Thus we see:

1. Since $\lim\limits_{x\to 0}\dfrac{\sin x}{x} = 1$ :

$$\left|\mathcal{A}^j(x^1,\omega_1+\omega_2)-\hat{\mathcal{C}}^j\right| \approx e^{\Im(\Delta\mathcal{K})\,x^1/2}\,\left|x^1\,\mathcal{C}^j\right| \quad \text{for } \left|\Delta\mathcal{K}\,x^1\right| \ll 1\,;\ j=2,3\,.$$

2. Therefore, for sufficiently good ***phase matching*** $|\Delta\mathcal{K}| \approx 0$ arbitrary increase of $\left|\widetilde{\boldsymbol{\mathcal{E}}}(\mathbf{x},\omega_1+\omega_2)\right| = \left|e^{-\frac{\omega_1+\omega_2}{c}\,\Im\left(\mathcal{N}_{\mathbf{e}_1}(\omega_1+\omega_2)\right)\,x^1}\,\mathcal{A}^j(x^1,\omega_1+\omega_2)\right|$ in the direction of propagation is possible in spite of absorption related to $\Im\left(\mathcal{N}\right)$ .

3. This indicates that low order perturbative approximations may have to be restricted to sufficiently small regions and that $\widetilde{\boldsymbol{\mathcal{E}}}_0(\mathbf{x},\omega)$ has to be adapted to the region under consideration.

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[20]Compare Footnote 48 of Chapter 2.3. Now, however, we allow for $\Im\left(\mathcal{N}_{\mathbf{e}_1}(\omega)\right) \neq 0$ .
[21]Note that (3.35) is consistent with (3.32) if $|\Delta\mathcal{K}| \ll \left|\frac{\omega_1+\omega_2}{c}\,\mathcal{N}_{\mathbf{e}_1}(\omega_1+\omega_2)\right|$ .

Actually, under the conditions considered above, good phase matching is very unlikely for $\omega_1, \omega_2 > 0$. To simplify the argument, let us consider the special case $\omega_1 = \omega_2$ and $\mu = 1$. Then

$$\Delta\mathcal{K} = 0 \iff \mathcal{N}_{\mathbf{e}_1}(2\,\omega_1) = \mathcal{N}_{\mathbf{e}_1}(\omega_1) \underset{(3.31)}{=} \epsilon_{\mathrm{c}}^2(2\,\omega_1) = \epsilon_{\mathrm{c}}^2(\omega_1)$$

and the latter condition is usually violated in sufficiently lossless media as can be understood by inspection of dispersion relations.

Fortunately, there are uniaxial media (with frequency-independent optical axis) for which ***type-I*** phase matching

$$\frac{\omega_1+\omega_2}{c}\,\mathcal{N}_{\mathbf{e}_1}^{\mathrm{exo}}(\omega_1+\omega_2) \approx \frac{\omega_1}{c}\,\mathcal{N}_{\mathbf{e}_1}^{\mathrm{ord}}(\omega_1) + \frac{\omega_2}{c}\,\mathcal{N}_{\mathbf{e}_1}^{\mathrm{ord}}(\omega_2) \tag{3.36}$$

or ***type-II*** phase matching

$$\frac{\omega_1+\omega_2}{c}\,\mathcal{N}_{\mathbf{e}_1}^{\mathrm{exo}}(\omega_1+\omega_2) \approx \frac{\omega_1}{c}\,\mathcal{N}_{\mathbf{e}_1}^{\mathrm{exo}}(\omega_1) + \frac{\omega_2}{c}\,\mathcal{N}_{\mathbf{e}_1}^{\mathrm{ord}}(\omega_2) \tag{3.37}$$

is possible without substantial damping **if** the optical axis is suitably oriented (**not** along $\mathbf{e}_1$).

**Exercise 7**

a) Using (2.72), show that

$$\begin{aligned}&\left\{|\mathcal{N}_{\mathbf{s}}^{\mathrm{exo}}(\omega)|^2 : \mathbf{s}\in\mathbb{R}^3\,,\ \mathbf{s}\cdot\mathbf{s}=1\right\}\\ &= \left[\min\left\{\left|\epsilon_{\mathrm{c}}^{\|}(\omega)\right|,\left|\epsilon_{\mathrm{c}}^{\perp}(\omega)\right|\right\}\,,\ \max\left\{\left|\epsilon_{\mathrm{c}}^{\|}(\omega)\right|,\left|\epsilon_{\mathrm{c}}^{\perp}(\omega)\right|\right\}\right],\end{aligned}$$

where $\epsilon_{\mathrm{c}}^{\|}$ denotes the nondegenerate and $\epsilon_{\mathrm{c}}^{\perp}$ the degenerate eigenvalue of $\overset{\leftrightarrow}{\epsilon}{}^{\mathrm{c}}$.

b) For $\omega_1 = \omega_2$ and the standard low-loss case

$$\left|\epsilon_{\mathrm{c}}^{\perp}(\omega_1)\right| < \left|\epsilon_{\mathrm{c}}^{\perp}(2\,\omega_1)\right|$$

show that

$$(3.36) \implies \left|\epsilon_{\mathrm{c}}^{\|}(2\,\omega_1)\right| \le \left|\epsilon_{\mathrm{c}}^{\perp}(\omega_1)\right|\,.$$

### 3.2.2 Three-Wave Mixing

Let us consider a uniaxial medium with axis along $\mathbf{e}_0 \in \mathbb{R}$ and positive frequencies $\omega_1, \omega_2$ for which the phase matching conditions are such that a solution of (3.11) (for $\widetilde{\jmath}_{\mathrm{cr}}(\mathbf{x},\omega) = 0$) with the following properties exists:

1. For every sufficiently small region $\mathcal{G}$ in $\mathbf{x}$-space there is a good first order perturbative approximation (3.18) with a suitably chosen zero order approximation (solution of (3.16)) consisting only of

   (a) two ordinary plane waves[22] of the form

$$\begin{aligned}&\mathbf{e}_{\mathrm{ord}}\,\mathcal{A}_{\mathcal{G}}(\omega)\,e^{i\,\frac{\omega}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega)\,x^1}\,\delta(\omega_j-\omega)\\&+\,\mathbf{e}_{\mathrm{ord}}\,\overline{\mathcal{A}_{\mathcal{G}}(\omega)\,e^{i\,\frac{\omega}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega)\,x^1}}\,\delta(\omega_j+\omega)\,;\quad j=1,2\end{aligned}$$

   where

$$\mathbf{e}_{\mathrm{ord}}\stackrel{\mathrm{def}}{=}\frac{\mathbf{e}_1\times\mathbf{e}_0}{|\mathbf{e}_1\times\mathbf{e}_0|}\,,\tag{3.38}$$

   and

   (b) an extraordinary plane wave[23] of the form

$$\begin{aligned}&\mathbf{e}_{\mathrm{exo}}\,\mathcal{A}_{\mathcal{G}}(\omega)\,e^{i\,\frac{\omega}{c}\,\mathcal{N}^{\mathrm{exo}}_{\mathbf{e}_1}(\omega)\,x^1}\,\delta(\omega_1+\omega_2-\omega)\\&+\mathbf{e}_{\mathrm{exo}}\,\overline{\mathcal{A}_{\mathcal{G}}(\omega)\,e^{i\,\frac{\omega}{c}\,\mathcal{N}^{\mathrm{exo}}_{\mathbf{e}_1}(\omega)\,x^1}}\,\delta(\omega_1+\omega_2+\omega)\,,\end{aligned}$$

   where

$$\mathbf{e}_{\mathrm{exo}}\stackrel{\mathrm{def}}{=}\frac{\left(\overset{\leftrightarrow c}{\epsilon}(\omega)\right)^{-1}\Big(\mathbf{e}_1\times(\mathbf{e}_1\times\mathbf{e}_0)\Big)}{\left|\left(\overset{\leftrightarrow c}{\epsilon}(\omega)\right)^{-1}\Big(\mathbf{e}_1\times(\mathbf{e}_1\times\mathbf{e}_0)\Big)\right|}\,.\tag{3.39}$$

2. There is a good slowly varying amplitude approximation for $\widetilde{\mathbf{E}}(\mathbf{x},\omega)$. More precisely: For some $\mathcal{A}_j(x^1,\omega)$ fulfilling[24]

$$\left|\left(\frac{\mathrm{d}}{\mathrm{d}x^1}\right)^2\mathcal{A}(x^1,\omega)\right|\ll\left|\frac{\omega}{c}\,\mathcal{N}_{\mathbf{e}_1}(\omega)\frac{\partial}{\partial x^1}\,\mathcal{A}(x^1,\omega)\right|\quad\forall\,x^1\in\mathbb{R}\,,\omega\in\mathbb{R}\tag{3.40}$$

   with

$$\mathcal{N}_{\mathbf{e}_1}(\omega)=\begin{cases}\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega)&\text{for }\omega=\omega\in\{\omega_1,\omega_2\}\,,\\\mathcal{N}^{\mathrm{exo}}_{\mathbf{e}_1}(\omega)&\text{for }\omega=\omega_1+\omega_2\,.\end{cases}$$

   we have

$$\begin{aligned}\widetilde{\mathbf{E}}(\mathbf{x},\omega)\;\approx\;&\mathbf{e}_{\mathrm{ord}}\,\mathcal{A}(x^1,\omega)\,e^{i\,\frac{\omega}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega)\,x^1}\Big(\delta(\omega_1-\omega)+\delta(\omega_2-\omega)\Big)\\&+\mathbf{e}_{\mathrm{exo}}\,\mathcal{A}(x^1,\omega)\,e^{i\,\frac{\omega}{c}\,\mathcal{N}^{\mathrm{exo}}_{\mathbf{e}_1}(\omega)\,x^1}\,\delta(\omega_1+\omega_2-\omega)\end{aligned}$$

   in some neighborhood of $\{\omega_1,\omega_2,\omega_1+\omega_2\}$.



[22]Recall (2.70) and (2.71).
[23]Recall (2.73) and Remark 2 in Section 2.2.3.
[24]Compare (3.32).

Under these conditions — according to (3.19), (3.18), and (3.5) — the following equations are approximately valid:

$$\hat{H}\left(\mathcal{A}(x^1,\omega_1)\,e^{i\,\frac{\omega_1}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1)\,x^1}\,\mathbf{e}_{\mathrm{ord}}\right)$$
$$= -\,(\omega_1)^2\left(\mathcal{A}(x^1,\omega)\,e^{i\,\frac{\omega}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega)\,x^1}\right)_{|\omega=\omega_1+\omega_2}\overline{\mathcal{A}(x^1,\omega_2)\,e^{i\,\frac{\omega_2}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_2)\,x^1}}\,\mathbf{a}\,, \tag{3.41}$$

$$\hat{H}\left(\mathcal{A}(x^1,\omega_2)\,e^{i\,\frac{\omega_2}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1)\,x^1}\,\mathbf{e}_{\mathrm{ord}}\right)$$
$$= -\,(\omega_2)^2\left(\mathcal{A}(x^1,\omega)\,e^{i\,\frac{\omega}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega)\,x^1}\right)_{|\omega=\omega_1+\omega_2}\overline{\mathcal{A}(x^1,\omega_1)\,e^{i\,\frac{\omega_1}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1)\,x^1}}\,\mathbf{b}\,, \tag{3.42}$$

$$\hat{H}\left(\mathcal{A}(x^1,\omega_1+\omega_2)\,e^{i\,\frac{\omega_1+\omega_2}{c}\,\mathcal{N}^{\mathrm{exo}}_{\mathbf{e}_1}(\omega_1+\omega_2)\,x^1}\,\mathbf{e}_{\mathrm{exo}}\right)$$
$$= -\,(\omega_1+\omega_2)^2\,\mathcal{A}(x^1,\omega_1)\,e^{i\,\frac{\omega_1}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1)\,x^1}\,\mathcal{A}(x^1,\omega_2)\,e^{i\,\frac{\omega_2}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_2)\,x^1}\,\mathbf{c}\,, \tag{3.43}$$

where:

$$\begin{aligned} a_j &\stackrel{\mathrm{def}}{=} 2\,\mu_0\,\chi^{(2)}_{jkl}(\omega_1+\omega_2,-\omega_2)\,e^k_{\mathrm{exo}}\,e^l_{\mathrm{ord}}\,,\\ b_j &\stackrel{\mathrm{def}}{=} 2\,\mu_0\,\chi^{(2)}_{jkl}(\omega_1+\omega_2,-\omega_1)\,e^k_{\mathrm{exo}}\,e^l_{\mathrm{ord}}\,,\\ c_j &\stackrel{\mathrm{def}}{=} 2\,\mu_0\,\chi^{(2)}_{jkl}(\omega_1,\omega_2)\,e^k_{\mathrm{ord}}\,e^l_{\mathrm{ord}}\,. \end{aligned} \tag{3.44}$$

The coupling of the components with frequencies $\omega_1$ , $\omega_2$ , and $\omega_1+\omega_2$ via equations (3.41)–(3.43) is called ***three-wave mixing***.

For simplicity, let us consider only the favorable case[25]

$$\mathbf{e}_0 = \mathbf{e}_2\,. \tag{3.45}$$

Then[26]

$$\mathbf{e}_{\mathrm{ord}} = \mathbf{e}_3\,,\quad \mathbf{e}_{\mathrm{exo}} = -\mathbf{e}_2 \tag{3.46}$$

and, therefore, also for the extraordinary sum-frequency component the propagation direction[27] is $\mathbf{e}_1$ . Application of $\hat{P}_{\perp\mathbf{e}_1}$ to (3.41)–(3.43) and use of (3.40) gives the

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[25] Since beams (suitable superpositions of plane wave solutions) are used in real experiments the propagation directions of all components should essentially coincide for most efficient sum-frequency generation. See (Armstrong et al., 1962b, Sect. VI) for adaption to general $\mathbf{e}_0$ .

[26] Note that in this case

$$\left(\overset{\leftrightarrow}{\epsilon}_{\mathrm{c}}(\omega_1+\omega_2)\right)^{-1}\left(\mathbf{e}_1\times(\mathbf{e}_1\times\mathbf{e}_0)\right) = -\left(\overset{\leftrightarrow}{\epsilon}^{\mathrm{c}}(\omega_1+\omega_2)\right)^{-1}\mathbf{e}_0 = -\epsilon^{\mathrm{c}}_{\|}(\omega_1+\omega_2)\,\mathbf{e}_0\,.$$

[27] Recall Exercise 6.

approximate equations[28]

$$\boxed{\begin{aligned}
\frac{\partial}{\partial x^1}\,\mathcal{A}(x^1,\omega_1) &= \frac{i\,\omega_1\,c}{\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1)}\,e^{i\,x^1\Delta_1\mathcal{K}}\,\mathcal{A}(x^1,\omega_1+\omega_2)\,\overline{\mathcal{A}(x^1,\omega_2)}\,a_3\,,\\
\frac{\partial}{\partial x^1}\,\mathcal{A}(x^1,\omega_2) &= \frac{i\,\omega_2\,c}{\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_2)}\,e^{i\,x^1\,\Delta_2\mathcal{K}}\,\mathcal{A}(x^1,\omega_1+\omega_2)\,\overline{\mathcal{A}(x^1,\omega_1)}\,b_3\,,\\
\frac{\partial}{\partial x^1}\,\mathcal{A}(x^1,\omega_1+\omega_2) &= -\frac{i\,(\omega_1+\omega_2)\,c}{\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1+\omega_2)}\,e^{i\,x^1\Delta_3\mathcal{K}}\,\mathcal{A}(x^1,\omega_1)\,\mathcal{A}(x^1,\omega_2)\,c_2\,,
\end{aligned}} \tag{3.47}$$

where:

$$\begin{aligned}
\Delta_1\mathcal{K} &\stackrel{\mathrm{def}}{=} \frac{\omega_1}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1) - \frac{\omega_1+\omega_2}{c}\,\mathcal{N}^{\mathrm{exo}}_{\mathbf{e}_1}(\omega_1+\omega_2) + \frac{\omega_2}{c}\,\overline{\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_2)}\,,\\
\Delta_2\mathcal{K} &\stackrel{\mathrm{def}}{=} \frac{\omega_2}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_2) - \frac{\omega_1+\omega_2}{c}\,\mathcal{N}^{\mathrm{exo}}_{\mathbf{e}_1}(\omega_1+\omega_2) + \frac{\omega_1}{c}\,\overline{\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1)}\,,\\
\Delta_3\mathcal{K} &\stackrel{\mathrm{def}}{=} \frac{\omega_1+\omega_2}{c}\,\mathcal{N}^{\mathrm{exo}}_{\mathbf{e}_1}(\omega_1+\omega_2) - \frac{\omega_1}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_1) - \frac{\omega_2}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_2)\,.
\end{aligned}$$

Note that (3.44) and (3.46) imply

$$\begin{aligned}
a_3 &= -2\,\mu_0\,\chi^{(2)}_{323}(\omega_1+\omega_2,-\omega_2)\,,\\
b_3 &= -2\,\mu_0\,\chi^{(2)}_{323}(\omega_1+\omega_2,-\omega_1)\,,\\
c_2 &= +2\,\mu_0\,\chi^{(2)}_{233}(\omega_1,\omega_2)\,.
\end{aligned}$$

We consider only the case

$$\Delta k \stackrel{\mathrm{def}}{=} \Delta_1\mathcal{K} = \Delta_2\mathcal{K} = -\Delta_3\mathcal{K} \in \mathbb{R}\,. \tag{3.48}$$

(lossless media) and[29]

$$K \stackrel{\mathrm{def}}{=} a_3 = b_3 = -c_2 \in \mathbb{R}\,. \tag{3.49}$$

Then the system of equations (3.47) simplifies to

$$\begin{aligned}
\mathcal{A}_1'(x) &= \frac{i\,(\omega_1)^2\;K}{k_1}\,e^{+i\,x\,\Delta k}\,\mathcal{A}_3(x)\,\overline{\mathcal{A}_2(x)}\,,\\
\mathcal{A}_2'(x) &= \frac{i\,(\omega_2)^2\;K}{k_2}\,e^{+i\,x\,\Delta k}\,\mathcal{A}_3(x)\,\overline{\mathcal{A}_1(x)}\,,\\
\mathcal{A}_3'(x) &= \frac{i\,(\omega_3)^2\;K}{k_3}\,e^{-i\,x\,\Delta k}\,\mathcal{A}_1(x)\,\mathcal{A}_2(x)\,,
\end{aligned} \tag{3.50}$$

where

$$\begin{aligned}
\mathcal{A}_j(x) &\stackrel{\mathrm{def}}{=} \mathcal{A}(x,\omega_j) \quad \text{for } j=1,2,3\,,\\
\omega_3 &\stackrel{\mathrm{def}}{=} \omega_1+\omega_2\,,\\
k_j &\stackrel{\mathrm{def}}{=} \tfrac{\omega_j}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_j) \quad \text{for } j=1,2\,,\\
k_3 &\stackrel{\mathrm{def}}{=} \tfrac{\omega_3}{c}\,\mathcal{N}^{\mathrm{ord}}_{\mathbf{e}_1}(\omega_3)\,.
\end{aligned}$$

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[28]Recall (3.12) and (3.29).

[29]See (Miller, 1964).

If we write the $\mathcal{A}_j$ in polar form

$$\mathcal{A}_j(x) = \rho_j(x)\, e^{i\varphi_j(x)} \quad \forall\, j \in \{1, 1, 3\}$$

and define

$$\theta(x) \stackrel{\text{def}}{=} x\,\Delta k + \varphi_3(x) - \varphi_1(x) - \varphi_2(x)$$

then (3.50) becomes equivalent to

$$\begin{aligned}
\rho_1'(x) + i\,\varphi_1'(x)\,\rho_1(x) &= \frac{i\,(\omega_1)^2\,K}{k_1}\,\rho_2(x)\,\rho_3(x)\,e^{+i\,\theta(x)}\,,\\
\rho_2'(x) + i\,\varphi_2'(x)\,\rho_2(x) &= \frac{i\,(\omega_2)^2\,K}{k_2}\,\rho_3(x)\,\rho_1(x)\,e^{+i\,\theta(x)}\,,\\
\rho_3'(x) + i\,\varphi_3'(x)\,\rho_3(x) &= \frac{i\,(\omega_3)^2\,K}{k_3}\,\rho_1(x)\,\rho_2(x)\,e^{-i\,\theta(x)}\,.
\end{aligned}$$

Separated into real and imaginary parts these equations read:

$$\begin{aligned}
\rho_1'(x) &= -\frac{(\omega_1)^2\,K}{k_1}\,\rho_2(x)\,\rho_3(x)\,\sin\theta(x)\,, & \varphi_1'(x)\,\rho_1(x) &= -\rho_1'(x)\,\cot\theta(x)\,,\\
\rho_2'(x) &= -\frac{(\omega_2)^2\,K}{k_2}\,\rho_3(x)\,\rho_1(x)\,\sin\theta(x)\,, & \varphi_2'(x)\,\rho_2(x) &= -\rho_2'(x)\,\cot\theta(x)\,,\\
\rho_3'(x) &= +\frac{(\omega_3)^2\,K}{k_3}\,\rho_1(x)\,\rho_2(x)\,\sin\theta(x)\,, & \varphi_3'(x)\,\rho_3(x) &= +\rho_3'(x)\,\cot\theta(x)\,.
\end{aligned} \tag{3.51}$$

The equations on the l.h.s of (3.51) imply that[30]

$$W \stackrel{\text{def}}{=} \sum_{j=1}^{3} \frac{k_j}{\omega_j}\,\left|\mathcal{A}_j(x)\right|^2 \quad \text{is independent of } x\,.$$

Therefore, with the definitions

$$u_j \stackrel{\text{def}}{=} \sqrt{\frac{k_j}{(\omega_j)^2\,W}}\,\rho_j \quad \forall\, j \in \{1, 2, 3\}$$

and

$$\zeta \stackrel{\text{def}}{=} K\sqrt{W\,\frac{(\omega_1)^2(\omega_2)^2(\omega_3)^2}{k_1\,k_2\,k_3}}\,x$$

the system of equations (3.51) become equivalent to[31]

$$\begin{aligned}
u_1'(\zeta) &= -u_2(\zeta)\,u_3(\zeta)\,\sin\theta(\zeta)\,, & \varphi_1'(\zeta)\,u_1(\zeta) &= -u_1'(\zeta)\,\cot\theta(\zeta)\,,\\
u_2'(\zeta) &= -u_3(\zeta)\,u_1(\zeta)\,\sin\theta(\zeta)\,, & \varphi_2'(\zeta)\,u_2(\zeta) &= -u_2'(\zeta)\,\cot\theta(\zeta)\,,\\
u_3'(\zeta) &= +u_1(\zeta)\,u_2(\zeta)\,\sin\theta(\zeta)\,, & \varphi_3'(\zeta)\,u_3(\zeta) &= +u_3'(\zeta)\,\cot\theta(\zeta)\,.
\end{aligned} \tag{3.52}$$

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[30] By (2.37), if $\mu = 1$, $\frac{2}{Z_0}W$ is the total power flow of the components with frequencies $\omega_1, \omega_2, \omega_1 + \omega_3$ (mediated over time), since we assumed the medium to be lossless.

[31] Thanks to the different physical dimensions of variables we do not need extra symbols for functions when identifying $f(\xi)$ with $f(z)$ in spite of $z \neq \zeta$.

The equations on the r.h.s. of (3.52) imply

$$\theta'(\zeta) = \Delta S + \cot\theta(\zeta)\,\frac{\mathrm{d}}{\mathrm{d}\zeta}\ln\Big(u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\Big)$$

resp.

$$\begin{aligned}&\cos\theta(\zeta)\,\frac{\mathrm{d}}{\mathrm{d}\zeta}\Big(u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\Big) - u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\,\theta'(\zeta)\,\sin\theta(\zeta)\\ &= -\Delta S\,u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\,\sin\theta(\zeta)\,,\end{aligned}\tag{3.53}$$

where

$$\Delta S \stackrel{\text{def}}{=} \frac{\Delta k}{K}\sqrt{\frac{k_1\,k_2\,k_3}{W\,(\omega_1)^2(\omega_2)^2(\omega_3)^2}}\,.$$

Since the last equation on the l.h.s. of (3.52) implies

$$u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\,\sin\theta(\zeta) = \frac{1}{2}\,\frac{\mathrm{d}}{\mathrm{d}\zeta}\Big(u_3(\zeta)\Big)^2$$

and since

$$\begin{aligned}&\cos\theta(\zeta)\,\frac{\mathrm{d}}{\mathrm{d}\zeta}\Big(u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\Big) - u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\,\theta'(\zeta)\,\sin\theta(\zeta)\\ &= \frac{\mathrm{d}}{\mathrm{d}\zeta}\Big(u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\,\cos\theta(\zeta)\Big)\,,\end{aligned}$$

(3.53) implies

$$\boxed{\cos\theta(\zeta) = \frac{\Gamma - \frac{1}{2}\Big(u_3(\zeta)\Big)^2\Delta S}{u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)}\,,\qquad \Gamma = \text{const.}\,.}\tag{3.54}$$

This may be used to eliminate $\theta$ in, e.g., the last equation on the l.h.s. of (3.52):

$$\frac{\mathrm{d}}{\mathrm{d}\zeta}\Big(u_3(\zeta)\Big)^2 = \pm 2\sqrt{\Big(u_1(\zeta)\,u_2(\zeta)\,u_3(\zeta)\Big)^2 - \left(\Gamma - \frac{1}{2}\Big(u_3(\zeta)\Big)^2\Delta S\right)^2}\,.\tag{3.55}$$

Thanks to the equations on the l.h.s. of (3.52), we have are constant:

$$\boxed{\begin{array}{lcl} m_1 & \stackrel{\text{def}}{=} & \Big(u_2(\zeta)\Big)^2 + \Big(u_3(\zeta)\Big)^2 = \text{const.}\,,\\ m_2 & \stackrel{\text{def}}{=} & \Big(u_3(\zeta)\Big)^2 + \Big(u_1(\zeta)\Big)^2 = \text{const.}\,,\\ m_3 & \stackrel{\text{def}}{=} & \Big(u_1(\zeta)\Big)^2 - \Big(u_2(\zeta)\Big)^2 = \text{const.}\,. \end{array}}\tag{3.56}$$

This together with (3.54) gives

$$\boxed{\zeta = \pm\frac{1}{2}\int_{(u_3(0))^2}^{(u_3(\zeta))^2}\frac{\mathrm{d}\lambda}{\sqrt{\lambda\,(m_2-\lambda)\,(m_1-\lambda) - \left(\Gamma - \frac{1}{2}\,\lambda\,\Delta S\right)^2}}\,.}\tag{3.57}$$

Now the intensities $\left(u_j(\zeta)\right)^2$ may be determined from (3.54)–(3.56) and the (boundary) values of the electromagnetic wave at $\zeta = 0$. For a detailed discussion see (Armstrong et al., 1962b).

For the simple case

$$\begin{array}{rcll} u_1 & = & u_2 & \text{(second harmonic generation[32])}\,, \\ \Delta S & = & 0 & \text{(perfect phase matching)}\,, \\ u_3(0) & = & 0 & \text{(second harmonic vanishing at the boundary)}\,, \\ \Gamma & = & 0 & \text{(no singularity in (3.54))}\,. \end{array}$$

(3.56) and (3.57) imply

$$\begin{aligned} \zeta &= \pm\frac{1}{2}\int_{\left(u_3(0)\right)^2}^{\left(u_3(\zeta)\right)^2} \frac{\mathrm{d}\lambda}{\sqrt{\lambda}\,(m_1-\lambda)} \\ &= \pm\frac{\tanh^{-1}\left(u_3(\zeta)/\sqrt{m_1}\right)}{\sqrt{m_1}} \end{aligned}$$

for $\zeta > 0$, hence

$$u_3(\zeta) = \sqrt[+]{m_1}\,\tanh\left(\sqrt[+]{m_1}\,|\zeta|\right)$$

inside the medium.

### 3.2.3 Four-Wave Mixing

See (Shen, 1984, Chapter 14).



[32]We suppress an additional scaling of amplitudes that is necessary in this case.

## 3.3 Quantum Aspects of Three-Wave Mixing

### 3.3.1 Quantized Radiation Inside Linear Media

Let us consider a uniaxial linear crystal with (essentially) real permittivity

$$\overset{\leftrightarrow}{\epsilon}(\omega) = \hat{1} + \overset{\leftrightarrow}{\chi}^{(1)}(\omega) = \begin{pmatrix} \epsilon_1(\omega) & 0 & 0 \\ 0 & \epsilon_2(\omega) & 0 \\ 0 & 0 & \epsilon_2(\omega) \end{pmatrix} \quad \text{w.r.t. the ONB } (\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3) \ .$$

Then, as shown in 2.2.3, for every (real) direction $\mathbf{s}$ there are monochromatic plane waves of the forms

$$\mathbf{E}^{\mathrm{o}}_{\mathbf{s},\omega}(\mathbf{x},t) \propto (\mathbf{s} \otimes \mathbf{e}_1)\, \Re\left(e^{-i\left(\omega\, t - \frac{\omega}{c}\, n^{\mathrm{o}}(\omega)\, \mathbf{s}\cdot\mathbf{x}\right)}\right) \quad \text{(ordinary wave)}$$

and

$$\mathbf{E}^{\mathrm{e}}_{\mathbf{s},\omega}(\mathbf{x},t) \propto \Big(\mathbf{s} \times (\mathbf{s} \otimes \mathbf{e}_1)\Big)\, \Re\left(e^{-i\left(\omega\, t - \frac{\omega}{c}\, n^{\mathrm{e}}_{\mathbf{s}}(\omega)\, \mathbf{s}\cdot\mathbf{x}\right)}\right) \quad \text{(extraordinary wave)}\ ,$$

where

$$n^{\mathrm{o}}(\omega) \stackrel{\text{def}}{=} \sqrt{\epsilon_2(\omega)} \quad \text{(independent of } \mathbf{s})$$

and[33]

$$n^{\mathrm{e}}_{\mathbf{s}}(\omega) \stackrel{\text{def}}{=} 1 \Big/ \sqrt{\left(\frac{s^1}{n^{\mathrm{o}}(\omega)}\right)^2 + \frac{(s^2)^2 + (s^3)^2}{\epsilon_1(\omega)}}\ .$$

Correspondingly, the quantized electromagnetic field inside the $\overset{\leftrightarrow}{\chi}^{(1)}$-crystal is of the form

$$\hat{\mathbf{E}}_{\overset{\leftrightarrow}{\chi}^{(1)}}(\mathbf{x},t) = \Big(\hat{\mathbf{E}}^{(+)}_{\mathrm{o}}(\mathbf{x},t) + \hat{\mathbf{E}}^{(+)}_{\mathrm{e}}(\mathbf{x},t)\Big) + \text{h.c.}\,,$$

where

$$\hat{\mathbf{E}}^{(+)}_{\mathrm{o}}(\mathbf{x},t) \stackrel{\text{def}}{=} (2\pi)^{-\frac{3}{2}} \int \underbrace{f_{\mathrm{o}}(\mathbf{k})\, \hat{a}_1(\mathbf{k})\, \underbrace{\boldsymbol{\epsilon}_1(\mathbf{k})}_{\stackrel{\text{def}}{=} \frac{\mathbf{k}}{|\mathbf{k}|}\times\mathbf{e}_1}\, e^{-i(\omega_{\mathrm{o}}(\mathbf{k})\, t - \mathbf{k}\cdot\mathbf{x})}}_{\text{ordinary classical wave}} \frac{dV_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}$$

and

$$\hat{\mathbf{E}}^{(+)}_{\mathrm{e}}(\mathbf{x},t) \stackrel{\text{def}}{=} (2\pi)^{-\frac{3}{2}} \int \underbrace{f_{\mathrm{e}}(\mathbf{k})\, \hat{a}_2(\mathbf{k})\, \underbrace{\boldsymbol{\epsilon}_2(\mathbf{k})}_{\stackrel{\text{def}}{=} \frac{\mathbf{k}}{|\mathbf{k}|}\times\left(\frac{\mathbf{k}}{|\mathbf{k}|}\times\mathbf{e}_1\right)}\, e^{-i(\omega_{\mathrm{e}}(\mathbf{k})\, t - \mathbf{k}\cdot\mathbf{x})}}_{\text{extraordinary classical wave}} \frac{dV_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\ .$$

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[33]Recall that the optical axis is along $\mathbf{e}_1$ : $n^{\mathrm{e}}_{\mathbf{e}_1}(\omega) = n^{\mathrm{o}}(\omega)$.

Of course, the Hamiltonian $\hat{H}_{\underset{\chi}{\leftrightarrow}(1)}$ has to fulfill

$$\frac{i}{\hbar}\left[\hat{H}_{\underset{\chi}{\leftrightarrow}(1)}\,,\,\hat{\mathbf{E}}_{\underset{\chi}{\leftrightarrow}(1)}(\mathbf{x},t)\right]_{-} = \frac{\partial}{\partial t}\,\hat{\mathbf{E}}_{\underset{\chi}{\leftrightarrow}(1)}(\mathbf{x},t)\,.$$

Therefore (up to an additive constant) we have

$$\hat{H}_{\underset{\chi}{\leftrightarrow}(1)} = \int \Big(\hbar\,\omega_{\rm o}(\mathbf{k})\,\Big(\hat{a}_1(\mathbf{k})\Big)^{\dagger}\hat{a}_1(\mathbf{k}) + \hbar\,\omega_{\rm e}(\mathbf{k})\,\Big(\hat{a}_2(\mathbf{k})\Big)^{\dagger}\hat{a}_2(\mathbf{k})\Big)\,\mathrm{d}V_{\mathbf{k}}\,.$$

Correspondingly, the generator of translations, often misinterpreted[34] as momentum observable, is

$$\hat{\mathbf{P}}_{\underset{\chi}{\leftrightarrow}(1)} = \int \hbar\,\mathbf{k}\,\Big(\Big(\hat{a}_1(\mathbf{k})\Big)^{\dagger}\hat{a}_1(\mathbf{k}) + \Big(\hat{a}_2(\mathbf{k})\Big)^{\dagger}\hat{a}_2(\mathbf{k})\Big)\mathrm{d}V_{\mathbf{k}}\,.$$

The amplitudes $f_{\rm o}(\mathbf{k})\,,\ f_{\rm e}(\mathbf{k})$ are fixed essentially by identification of the Hamiltonian with the quantum version of $\int \Big(\mathcal{H}_0(\mathbf{x},t) + \mathcal{U}(\mathbf{x},t)\Big)\mathrm{d}V_{\mathbf{x}}$, i.e. by postulating[35] that

$$\int \Big(\hbar\,\omega_{\rm o}(\mathbf{k})\,\Big(\hat{a}_1(\mathbf{k})\Big)^{\dagger}\hat{a}_1(\mathbf{k}) + \hbar\,\omega_{\rm e}(\mathbf{k})\,\Big(\hat{a}_2(\mathbf{k})\Big)^{\dagger}\hat{a}_2(\mathbf{k})\Big)\,\mathrm{d}V_{\mathbf{k}} = \int \hat{\mathcal{H}}(\mathbf{x},t)\,\mathrm{d}V_{\mathbf{x}} \qquad (3.58)$$

holds for some quantum field $\hat{\mathcal{H}}(\mathbf{x},t)$ fulfilling the conditions

$$\lim_{t\to-\infty}\Big\langle \Phi \,\Big|\, \hat{\mathcal{H}}(\mathbf{x},t)\,\Phi\Big\rangle = 0 \quad \text{for sufficiently well-behaved } \Phi \in \mathcal{H}_{\rm field} \qquad (3.59)$$

and

$$\frac{\partial}{\partial t}\,\hat{\mathcal{H}}(\mathbf{x},t) = :\epsilon_0\,\hat{\mathbf{E}}(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\hat{\mathbf{E}}(\mathbf{x},t) + \frac{1}{\mu_0}\,\hat{\mathbf{B}}(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\hat{\mathbf{B}}(\mathbf{x},t) + \hat{\mathbf{E}}(\mathbf{x},t)\cdot\hat{\boldsymbol{\mathcal{P}}}(\mathbf{x},t):\,, \qquad (3.60)$$

where[36]

$$\begin{aligned}
\hat{\boldsymbol{\mathcal{P}}}(\mathbf{x},t) &\stackrel{\rm def}{=} \hat{\boldsymbol{\mathcal{P}}}^{(+)}(\mathbf{x},t) + \Big(\hat{\boldsymbol{\mathcal{P}}}^{(+)}(\mathbf{x},t)\Big)^{\dagger}\,,\\
\hat{\boldsymbol{\mathcal{P}}}^{(+)}(\mathbf{x},t) &\stackrel{\rm def}{=} (2\pi)^{-\frac{3}{2}}\int f_{\rm o}(\mathbf{k})\,\hat{a}_1(\mathbf{k})\,\epsilon_0\,\overset{\leftrightarrow}{\epsilon}\Big(\omega_{\rm o}(\mathbf{k})\Big)\boldsymbol{\epsilon}_1(\mathbf{k})\,e^{-i(\omega_{\rm o}(\mathbf{k})\,t-\mathbf{k}\cdot\mathbf{x})}\,\frac{\mathrm{d}V_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\\
&\quad + (2\pi)^{-\frac{3}{2}}\int f_{\rm e}(\mathbf{k})\,\hat{a}_2(\mathbf{k})\,\epsilon_0\,\overset{\leftrightarrow}{\epsilon}\Big(\omega_{\rm e}(\mathbf{k})\Big)\boldsymbol{\epsilon}_2(\mathbf{k})\,e^{-i(\omega_{\rm o}(\mathbf{k})\,t-\mathbf{k}\cdot\mathbf{x})}\,\frac{\mathrm{d}V_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\,.
\end{aligned}$$

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[34] See (Garrison and Chiao, 2004), in this connection.

[35] The requirements (3.58)–(3.60), adopted to (1.15)–(1.17), guarantee that the energy expection values in coherent states coincide with the corresponding classical energies.

[36] Recall (2.1) and (3.6).

Straightforward calculation yields

$$\begin{aligned}\int \hat{\mathcal{H}}(\mathbf{x},t)\,\mathrm{d}V_{\mathbf{x}} &= \int \frac{\left(n^{\mathrm{o}}(\mathbf{k})\right)^2}{c^2\,|\mathbf{k}|\,\mu_0}\,|f_{\mathrm{o}}(\mathbf{k})|^2\,\left(\hat{a}_1(\mathbf{k})\right)^\dagger \hat{a}_1(\mathbf{k})\,\mathrm{d}V_{\mathbf{k}} \\ &\quad + \int \frac{\left(n^{\mathrm{e}}(\mathbf{k})\right)^2}{c^2\,|\mathbf{k}|\,\mu_0}\,|f_e(\mathbf{k})|^2\,\left(\hat{a}_2(\mathbf{k})\right)^\dagger \hat{a}_2(\mathbf{k})\,\mathrm{d}V_{\mathbf{k}}\end{aligned} \tag{3.61}$$

and thus

$$f_j(\mathbf{k}) = i\,c\,|\mathbf{k}|\,\sqrt{\mu_0\,\hbar\,c}\,\sqrt{\frac{\omega_j(\mathbf{k})}{c\,|\mathbf{k}|\,\left(n^j(\mathbf{k})\right)^2}}\,,\quad \text{for } j\in\{\mathrm{o}\,,\,\mathrm{e}\}\,,$$

by (3.58), up to irrelevant phase factors.[37]

**Proof of (3.61):** ...................

**Final remark:** One should not be surprised that the interaction with the infinitely extended medium cannot be described in the naive interaction picture:

For fixed $t$, unless $\overset{\leftrightarrow}{\chi} = \hat{1}$, the quantum field $\hat{\mathbf{E}}_{\overset{\leftrightarrow}{\chi}^{(1)}}(\mathbf{x},t)$ is **not** unitarily equivalent to the free field $\hat{\mathbf{E}}(\mathbf{x},t)$.

### 3.3.2 Nonlinear Perturbation

Let us now consider a finitely extended $\chi^{(1)}$-$\chi^{(2)}$ crystal embedded[38] in the $\chi^{(1)}$ cystal, i.e. that

$$\overset{\leftrightarrow}{\check{\chi}}{}^{(1)}(\mathbf{x},t;\mathbf{x}_1',t_1') = \delta(\mathbf{x}-\mathbf{x}_1')\,(2\pi)^{-\frac{1}{2}}\int \begin{pmatrix}\epsilon_1(\omega) & 0 & 0\\ 0 & \epsilon_2(\omega) & 0\\ 0 & 0 & \epsilon_2(\omega)\end{pmatrix} e^{-i\,\omega\left(t-t_1'\right)}\,\mathrm{d}\omega_1$$

and

$$\begin{aligned}&\check{\chi}^{(2)}_{jk_1k_2}(\mathbf{x},t;\mathbf{x}_1',t_1';\mathbf{x}_2',t_2')\\ &= \chi_{\mathcal{G}}(\mathbf{x})\,\delta(\mathbf{x}-\mathbf{x}_1')\,\delta(\mathbf{x}-\mathbf{x}_2')\,\frac{1}{2\pi}\int \chi^{(2)}_{jk_1k_2}(\omega_1,\omega_2)\,e^{-i\left(\omega_1\left(t-t_1'\right)+\omega_2\left(t-t_2'\right)\right)}\,\mathrm{d}\omega_1\,\mathrm{d}\omega_2\end{aligned}$$



[37] Obviously, such phase factors could be removed by corrsponding redefinition of the annihilation operators $a_1(\mathbf{k}), a_2(\mathbf{k})$ not changing their (canonical) commutation relations.

[38] This way we (essentially) prohibit diffraction at the boundaries of the $\chi^{(1)}$-$\chi^{(2)}$ crystal.

for some bounded region $\mathcal{G}$ whereas

$$\check{\chi}^{(\nu)} = 0 \quad \forall\, \nu > 2\,.$$

The Hamiltonian of the quantized theory now is[39]

$$\hat{H} = \hat{H}_{\overset{\leftrightarrow}{\chi}^{(1)}} + \int_{\mathcal{G}} \hat{\mathcal{H}}_{\mathrm{int}}(\mathbf{x},0)\, \mathrm{d}V_{\mathbf{x}} \tag{3.62}$$

with $\hat{\mathcal{H}}_{\mathrm{int}}(\mathbf{x},0)$ being the (normally ordered) quantum version of $\mathcal{H}_{\mathrm{int}}(\mathbf{x},t)$, the latter being characterized by

$$\lim_{t\to-\infty} \mathcal{H}_{\mathrm{int}}(\mathbf{x},t) = 0 \quad \text{for sufficiently well-behaved } \mathbf{E}(\mathbf{x},t) \tag{3.63}$$

and

$$\frac{\partial}{\partial t}\,\mathcal{H}_{\mathrm{int}}(\mathbf{x},t) = E^j(\mathbf{x},t)\cdot\frac{\partial}{\partial t}\,\mathcal{P}^{(2)}_j(\mathbf{x},t)\,, \tag{3.64}$$

where

$$\mathcal{P}^{(2)}_j(\mathbf{x},\omega) \underset{(3.6)}{=} \frac{\epsilon_0}{\sqrt{2\pi}} \int \chi^{(2)}_{jk_1k_2}(\omega_1,\omega_2)\, \widetilde{E}^{k_1}(\mathbf{x},\omega_1)\, \widetilde{E}^{k_2}(\mathbf{x},\omega_2)\, \delta(\omega_1+\omega_2-\omega)\, \mathrm{d}\omega_1\, \mathrm{d}\omega_2\,.$$

Inserting the latter into (3.64) we get

$$\begin{aligned}
\frac{\partial}{\partial t}\,\mathcal{H}_{\mathrm{int}}(\mathbf{x},t) &= -i\,\frac{\epsilon_0}{(2\pi)^{\frac{3}{2}}} \int \widetilde{E}^j(\mathbf{x},\omega')\,(\omega-\omega')\,\chi^{(2)}_{jk_1k_2}(\omega_1,\omega_2)\,\widetilde{E}^{k_1}(\mathbf{x},\omega_1)\\
&\qquad\qquad \widetilde{E}^{k_2}(\mathbf{x},\omega_2)\,\delta\Big(\omega_1+\omega_2-(\omega-\omega')\Big)\,\mathrm{d}\omega_1\,\mathrm{d}\omega_2\,\mathrm{d}\omega'\,e^{-i\,\omega\,t}\,\mathrm{d}\omega\\
&= -i\,\frac{\epsilon_0}{(2\pi)^{\frac{3}{2}}} \int (\omega_1+\omega_2)\,e^{-i\,(\omega'+\omega_1+\omega_2)\,t}\,\chi^{(2)}_{jk_1k_2}(\omega_1,\omega_2)\\
&\qquad\qquad \widetilde{E}^j(\mathbf{x},\omega')\,\widetilde{E}^{k_1}(\mathbf{x},\omega_1)\,\widetilde{E}^{k_2}(\mathbf{x},\omega_2)\,\mathrm{d}\omega_1\,\mathrm{d}\omega_2\,\mathrm{d}\omega'
\end{aligned}$$

and thus, thanks to (3.5),

$$\begin{aligned}
\frac{1}{2}\frac{\partial}{\partial t}\,\mathcal{H}_{\mathrm{int}}(\mathbf{x},t) &= -i\,\frac{\epsilon_0}{(2\pi)^{\frac{3}{2}}} \int \omega_1\,e^{-i\,(\omega_1+\omega_2+\omega_3)\,t}\,\chi^{(2)}_{k_3k_1k_2}(\omega_1,\omega_2)\\
&\qquad\qquad \widetilde{E}^{k_1}(\mathbf{x},\omega_1)\,\widetilde{E}^{k_2}(\mathbf{x},\omega_2)\,\widetilde{E}^{k_3}(\mathbf{x},\omega_3)\,\mathrm{d}\omega_1\,\mathrm{d}\omega_2\,\mathrm{d}\omega_3\\
&= -i\,\frac{\epsilon_0}{(2\pi)^{\frac{3}{2}}} \int \omega_2\,e^{-i\,(\omega_1+\omega_2+\omega_3)\,t}\,\chi^{(2)}_{k_3k_1k_2}(\omega_1,\omega_2)\\
&\qquad\qquad \widetilde{E}^{k_1}(\mathbf{x},\omega_1)\,\widetilde{E}^{k_2}(\mathbf{x},\omega_2)\,\widetilde{E}^{k_3}(\mathbf{x},\omega_3)\,\mathrm{d}\omega_1\,\mathrm{d}\omega_2\,\mathrm{d}\omega_3\,.
\end{aligned}$$

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[39] Recall (1.15)–(1.17).

For situations in which also

$$\begin{aligned}
&\int \omega_1\, e^{-i\,(\omega_1+\omega_2+\omega_3)\,t}\, \chi^{(2)}_{k_3 k_1 k_2}(\omega_1,\omega_2)\, \widetilde{E}^{k_1}(\mathbf{x},\omega_1)\, \widetilde{E}^{k_2}(\mathbf{x},\omega_2)\, \widetilde{E}^{k_3}(\mathbf{x},\omega_3)\, \mathrm{d}\omega_1\, \mathrm{d}\omega_2\, \mathrm{d}\omega_3 \\
&= \int \omega_3\, e^{-i\,(\omega_1+\omega_2+\omega_3)\,t}\, \chi^{(2)}_{k_3 k_1 k_2}(\omega_1,\omega_2)\, \widetilde{E}^{k_1}(\mathbf{x},\omega_1)\, \widetilde{E}^{k_2}(\mathbf{x},\omega_2)\, \widetilde{E}^{k_3}(\mathbf{x},\omega_3)\, \mathrm{d}\omega_1\, \mathrm{d}\omega_2\, \mathrm{d}\omega_3
\end{aligned} \tag{3.65}$$

holds and $\chi^{(2)}_{k_3 k_1 k_2}(\omega_1,\omega_2)$ can be replaced by the constant tensor $\chi^{(2)}_{k_3 k_1 k_2}$ this implies

$$\begin{aligned}
&\frac{3}{2}\,\mathcal{H}_{\mathrm{int}}(\mathbf{x},t) \\
&= \epsilon_0\, \chi^{(2)}_{k_3 k_1 k_2} \int e^{-i\,(\omega_1+\omega_2+\omega_3)\,t}\, \widetilde{E}^{k_1}(\mathbf{x},\omega_1)\, \widetilde{E}^{k_2}(\mathbf{x},\omega_2)\, \widetilde{E}^{k_3}(\mathbf{x},\omega_3)\, \mathrm{d}\omega_1\, \mathrm{d}\omega_2\, \mathrm{d}\omega_3 \\
&= \epsilon_0\, \chi^{(2)}_{k_3 k_1 k_2}\, E^{k_1}(\mathbf{x},t)\, E^{k_2}(\mathbf{x},t)\, E^{k_3}(\mathbf{x},t)\,.
\end{aligned}$$

Hence, if $\hat{P}_{\mathrm{II}}$ is the projector onto a corresponding subspace of $\mathcal{H}_{\mathrm{field}}$ , we have

$$\hat{P}_{\mathrm{II}}\, \hat{\mathcal{H}}_{\mathrm{int}}(\mathbf{x},0) \hat{P}_{\mathrm{II}} = \frac{2}{3}\,\epsilon_0\, \hat{P}_{\mathrm{II}} : \hat{E}^{k_1}_{\underset{\chi}{\leftrightarrow}(1)}(\mathbf{x},0)\, \hat{E}^{k_2}_{\underset{\chi}{\leftrightarrow}(1)}(\mathbf{x},0)\, \hat{E}^{k_3}_{\underset{\chi}{\leftrightarrow}(1)}(\mathbf{x},0) : \hat{P}_{\mathrm{II}}$$

⋮

### 3.3.3 Type-II Spontaneous Down Conversion

⋮

# Chapter 4

# Photodetection

## 4.1 Simple Detector Models

### 4.1.1 Single Localized Detectors

The counting rate at time $t$ for a photodetector localized at $\mathbf{x}$ in an **almost monochromatic classical** radiation field with complex vector potential is (the local space-time average of)

$$\sum_{j_1,j_2=1}^{3} A^{(-)^{j_1}}(\mathbf{x},t)\, A^{(+)^{j_2}}(\mathbf{x},t)\, \eta^{j_1 j_2}\,,$$

where the $\eta^{j_1 j_2}$ (depending on the mean frequency of the radiation) characterize the efficiency of the detector.[1]

> **Remark:** For almost monochromatic (not quantized) light with mean angular frequency $\omega_0$
>
> $$\mathbf{E}(\mathbf{x},t) = -\frac{\partial}{\partial t}\,\mathbf{A}(\mathbf{x},t)$$
>
> implies:
>
> $$E^{(-)^{j_1}}(\mathbf{x},t)\, E^{(+)^{j_2}}(\mathbf{x},t) \;\approx\; (\omega_0)^2\; A^{(-)^{j_1}}(\mathbf{x},t)\, A^{(+)^{j_2}}(\mathbf{x},t)\,.$$

For quantized almost monochromatic radiation in the (pure) state $\phi$ one can show[2] that the corresponding detection rate is given in first order perturbation theory the formally similar expression[3]

$$P = \sum_{j_1,j_2=1}^{3} \left\langle \phi \,\middle|\, \hat{A}^{(-)j_1}(\mathbf{x},t)\, \hat{A}^{(+)j_2}(\mathbf{x},t)\, \phi \right\rangle \eta^{j_1 j_2}\,.$$

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[1] For achievable efficiencies see (Jackson and Hockney, 2004).

[2] See Section 7.2.2.

[3] Hence, in this approximation, the counting rate in the vacuum state is zero.

**Special cases:**

1. For $\phi = \chi_{\hat{a}}(\alpha)$ we have
$$\hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\,\phi = \alpha\,\mathbf{A}^{(+)}(\mathbf{x},t)\,\phi$$
with
$$\mathbf{A}^{(+)}(\mathbf{x},t) \stackrel{\text{def}}{=} \left\langle \Omega \,\middle|\, \hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\,\hat{a}^\dagger\,\Omega \right\rangle$$
and hence
$$P = |\alpha|^2 \sum_{j_1,j_2=1}^{3} A^{(-)j_1}(\mathbf{x},t)\,A^{(+)j_2}(\mathbf{x},t)\,\eta^{j_1 j_2}\,.$$

2. For the (normalized) $n$-photon state
$$\phi = \frac{1}{\sqrt{n!}}\left(\hat{a}^\dagger\right)^n \Omega$$
we have
$$\left\langle \phi \,\middle|\, \hat{A}^{(-)j_1}(\mathbf{x},t) \underbrace{\hat{A}^{(+)j_2}(\mathbf{x},t)\,\phi}_{=\frac{n}{\sqrt{n!}}\left[\mathbf{A}_0^{(+)}(\mathbf{x},t)j_2(\mathbf{x},t),\hat{a}^\dagger\right]_-\left(\hat{a}^\dagger\right)^{n-1}\Omega} \right\rangle = n\,A^{(-)j_1}(\mathbf{x},t)\,A^{(+)j_2}(\mathbf{x},t)\,,$$
in spite of
$$\left\langle \phi \,\middle|\, \hat{\mathbf{E}}(\mathbf{x},t)\,\phi \right\rangle = 0\,.$$

**Remarks:**

1. The phases of the 1-photon state vectors do not play any role for photodetection!
2. Therefore[4] the counting rates in the **mixed** state[5]
$$\begin{aligned}\hat{\rho}_{\hat{a}}(\alpha) &\stackrel{\text{def}}{=} \frac{1}{2\pi}\int_0^{2\pi} \left|\chi_{\hat{a}}(|\alpha|\,e^{i\varphi})\right\rangle\!\left\langle\chi_{\hat{a}}(|\alpha|\,e^{i\varphi})\right| \\ &= \sum_{\nu=0}^{\infty}\left|\frac{\alpha^\nu}{\nu!}\right|^2 \left|\left(\hat{a}^\dagger\right)^n\Omega\right\rangle\!\left\langle\left(\hat{a}^\dagger\right)^n\Omega\right|\end{aligned}$$
are the same as in the (pure) coherent state $\chi_{\hat{a}}(\alpha)$.
3. Usually, experimental checks of predictions for $n$-photon states are performed via *post selection*.[6]



[4] See (Sanders et al., 2003) and (Nemoto and Braunstein, 2003) in this connection.

[5] This state corresponds fairly well to the field of a single-mode laser inside the resonant cavity in case of high pump intensity; see (Mandel and Wolf, 1995, Sect. 18.5.2) and (Wiseman, 2004).

[6] Since coherent superposition with the vacuum state, at least, are unavoidable.

### 4.1.2 Independent Detectors

For an **almost monochromatic classical** radiation field with complex vector potential $\mathbf{A}^{(+)}(\mathbf{x},t)$

$$\underbrace{P_{\mathbf{A}^{(+)}}(\mathbf{x}_1,t_1;\mathbf{x}_2,t_2;\ldots;\mathbf{x}_n,t_n)}\qquad\qquad \mathrm{d}t_1\cdots\mathrm{d}t_n$$
$$\stackrel{\text{def}}{=} \prod_{\nu=1}^{n}\left(\sum_{j_\nu,j'_\nu=1}^{3}\hat{A}^{(-)j_\nu}(\mathbf{x}_\nu,t_\nu)\,\hat{A}^{(+)j'_\nu}(\mathbf{x}_\nu,t_\nu)\,\eta_\nu^{j_\nu j'_\nu}\right)$$

is the probability that for all $\nu\in\{1,\ldots,n\}$ the detector localized at $\mathbf{x}_\nu$ fires during the time interval $[t_\nu,t_\nu+\mathrm{d}t_\nu]$. Here $\eta_\nu^{j_\nu j'_\nu}$ characterizes the efficiency of the $\nu$-th detector and we assume that the detectors have no (essential) effect on each other.

Correspondingly, for a **coherent** state $\chi$ this probability can be shown to be

$$P_\chi(\mathbf{x}_1,t_1;\ldots;\mathbf{x}_n,t_n)\,\mathrm{d}t_1\cdots\mathrm{d}t_n$$

in first order perturbation theory,[7] where

$$\begin{aligned}
&P_\chi(\mathbf{x}_1,t_1;\ldots;\mathbf{x}_n,t_n)\\
&\stackrel{\text{def}}{=}\sum_{j_1,\ldots,j'_n=1}^{3}\left\langle\phi\,\middle|\,\hat{A}^{(-)j_1}(\mathbf{x}_1,t_1)\cdots\hat{A}^{(-)j_n}(\mathbf{x}_n,t_n)\,\hat{A}^{(+)j'_n}(\mathbf{x}_n,t_n)\cdots\right.\\
&\qquad\qquad\left.\cdots\hat{A}^{(+)j'_1}(\mathbf{x}_1,t_1)\,\phi\right\rangle\eta_1^{j_1j'_1}\cdots\eta_n^{j_nj'_n}\quad\forall\,\phi\in\mathcal{H}_{\text{field}}\,.
\end{aligned}$$

This together with the general rule (6.11) and the optical equivalence theorem shows:

> For **every** physically relevant state $\widehat{=}\,\hat\rho$ of the radiation field — to first order of quantum mechanical perturbation theory — the probability that for all $\nu\in\{1,\ldots,n\}$ the detector localized at $\mathbf{x}_\nu$ fires during the time interval $[t_\nu,t_\nu+\mathrm{d}t_\nu]$ is
>
> $$P_{\hat\rho}(\mathbf{x}_1,t_1;\mathbf{x}_2,t_2;\ldots;\mathbf{x}_n,t_n)\,\mathrm{d}t_1\cdots\mathrm{d}t_n\,,$$
>
> where:
>
> $$\begin{aligned}
&P_{\hat\rho}(\mathbf{x}_1,t_1;\mathbf{x}_2,t_2;\ldots;\mathbf{x}_n,t_n)\\
&\stackrel{\text{def}}{=}\sum_{j_1,\ldots,j'_n=1}^{3}\mathrm{Tr}\Big((\hat\rho\,\hat{A}^{(-)j_1}(\mathbf{x}_1,t_1)\cdots\hat{A}^{(-)j_n}(\mathbf{x}_n,t_n)\,\hat{A}^{(+)j_n}(\mathbf{x}_n,t_n)\cdots\\
&\qquad\qquad\cdots\hat{A}^{(+)j_1}(\mathbf{x}_1,t_1)\Big)\,\eta_1^{j_1j'_1}\cdots\eta_n^{j_nj'_n}\,.
\end{aligned}\tag{4.1}$$

We may also consider idealized ‘distributed’ detectors[8] which sample the field at various points of space-time and are sensitive to the resulting effective field. A



[7] See Section 7.2.2.

[8] See (Klauder and Sudarshan, 1968, p. 150).

typical example is YOUNG's double-slit experiment, considered in 4.3.1, where the actual detector is sensitive to the field sampled from both slits.[9] The probability density for arrays of such detectors may be given by

$$\begin{aligned}
&P_{\hat\rho}(\mathbf{x}_1, t_1; \ldots ; \mathbf{x}_n, t_n) \\
&\approx \sum_{j_1,\ldots,j_n,j'_1,\ldots,j'_n=1}^{3} \operatorname{Tr}\Big(\hat\rho\, {\hat A^{(-)}_{\mathrm{eff},1}}{}^{j_1}(\mathbf{x}_1,t_1)\cdots {\hat A^{(-)}_{\mathrm{eff},n}}{}^{j_n}(\mathbf{x}_n,t_n) {\hat A^{(+)}_{\mathrm{eff},n}}{}^{j'_n}(\mathbf{x}_n,t_n)\cdots \\
&\qquad\qquad \cdots {\hat A^{(+)}_{\mathrm{eff},1}}{}^{j'_1}(\mathbf{x}_1,t_1)\Big)\, \eta_1^{j_1 j'_1}\cdots \eta_n^{j_n j'_n}\,, \\
&\left({\hat A^{(-)}_{\mathrm{eff},\nu}}{}^{j}(\mathbf{x},t)\right)^* \stackrel{\mathrm{def}}{=} {\hat A^{(+)}_{\mathrm{eff},\nu}}{}^{j}(\mathbf{x},t) \stackrel{\mathrm{def}}{=} \int {\hat A_0^{(+)}}{}^{k}(\mathbf{x}-\mathbf{x}', t-t')\, S_\nu^{jk}(\mathbf{x}',t')\,\mathrm{d}V_{\mathbf{x}'}\,\mathrm{d}t'\,,
\end{aligned}$$

instead of (4.1), with suitable distributions $S_\nu^{jk}(\mathbf{x}',t')$.

All this suggests that, in principle, all the correlation functions

$$\begin{aligned}
&G_{\hat\rho}^{(n,m)}(\mathbf{x}_1,t_1,j_1;\ldots;\mathbf{x}_{n+m},t_{n+m},j_{n+m}) \\
&\stackrel{\mathrm{def}}{=} \operatorname{Tr}\left(\hat\rho\left(\prod_{\nu=1}^{n} \hat A_0^{(-)j_\nu}(\mathbf{x}_\nu,t_\nu)\right)\prod_{\mu=n+1}^{n+m}\hat A_0^{(+)j_\mu}(\mathbf{x}_\mu,t_\mu)\right)
\end{aligned} \tag{4.2}$$

with $\underline{\underline{n=m}} \in \mathbb{N}$ are determined by the rate information from all possible counting arrangements.

**Remark:** If $G_{\hat\rho}^{(n,m)}$ exist for all $n,m \in \mathbb{N}$ — as is usually the case for physically realizable $\hat\rho$ — then the totality of all these correlation functions characterizes $\hat\rho$ uniquely, as may be shown by methods of axiomatic field theory (see, e.g., Corollary 2.2.16 of (Lücke, qft)). Actually, according to (1.31), every $G_{\hat\rho}^{(n,m)}$ is already fixed by its values at $t_1 = \ldots = t_{n+m} = 0$.

## 4.2 Quantum Theory of Coherence

### 4.2.1 Correlation Functions in General

As explained in 4.1.2, the counting rates of corresponding detector arrays for the quantum state $\hat\rho$ of the radiation field are given (to first approximation) by the $\eta_\nu^{j_\nu j'_\nu}$ and the ***correlation functions***

$$G_{\hat\rho}^{(n,m)}(\mathbf{x}_1,t_1,j_1;\ldots;\mathbf{x}_n,t_n,j_n) \stackrel{\mathrm{def}}{=} \operatorname{Tr}\left(\hat\rho\left(\prod_{\nu=1}^{n} \hat A^{(-)j_\nu}(\mathbf{x}_\nu,t_\nu)\right)\prod_{\mu=n+1}^{n+m}\hat A^{(+)j_\mu}(\mathbf{x}_\mu,t_\mu)\right).$$



[9] See, e.g. (Ficek and Swain, 2001) and (Brukner and Zeilinger, 2002).

In the following we make use of the identification

$$x_\nu \equiv (\mathbf{x}_\nu, t_\nu, j_\nu) \in \mathcal{M} \stackrel{\text{def}}{=} \mathbb{R}^3 \times \mathbb{R} \times \{1,2,3\} \,.$$

In this notation we have, e.g.,[10]

$$G^{(n,m)}_{\hat\rho^{\mathrm{I}}}(x_1;\ldots;x_{n+m}) = \left(G^{(n,m)}_{\hat\rho^{\mathrm{I}}}(x_{n+m};\ldots;x_1)\right)^* \quad \forall\, n,m \in \mathbb{N}\,,\; x_1,\ldots,x_{n+m} \in \mathcal{M}\,. \tag{4.3}$$

**Lemma 4.2.1** *The density operator $\hat\rho^{\mathrm{I}}$ on $\mathcal{H}_{\text{field}}$ is a coherent state,[11] i.e. there is a mode $\hat a$ and a complex number $\alpha$ with*

$$\hat\rho^{\mathrm{I}} = \left|\hat D_{\hat a}(\alpha)\,\Omega\right\rangle\left\langle \hat D_{\hat a}(\alpha)\,\Omega\right| \,, \tag{4.4}$$

*iff there is a function $V(x)$ on $\mathcal{M}$ with*

$$G^{(n,m)}_{\hat\rho^{\mathrm{I}}}(x_1;\ldots;x_{n+m}) = \prod_{\nu=1}^{n}\left(V(x_\nu)\right)^* \prod_{\mu=n+1}^{n+m} V(x_\mu) \quad \forall\, n,m \in \mathbb{N}\,,\; x_1,\ldots,x_{n+m} \in \mathcal{M}\,. \tag{4.5}$$

*More precisely, (4.4) implies (4.5) for*[12]

$$\begin{aligned} V(\mathbf{x},t,j) &= \left\langle \hat D_{\hat a}(\alpha)\,\Omega \mid \underbrace{\hat A_0^{(+)^j}(\mathbf{x},t)\,\hat D_{\hat a}(\alpha)\,\Omega}_{\stackrel{\sim}{(1.84)}\;\hat D_{\hat a}(\alpha)\,\Omega}\right\rangle \\ &\underset{(1.84)}{=} \alpha \left\langle \Omega \mid \hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\,\hat a^\dagger\,\Omega\right\rangle . \end{aligned} \tag{4.6}$$

**Outline of proof:** The density operator $\hat\rho^{\mathrm{I}}$ is uniquely fixed by the correlation functions $G^{(n,m)}_{\hat\rho^{\mathrm{I}}}$ (recall the remark at the end of Sectionnlqo-S-indDet). Since, obviously, (4.4) and (4.6) imply (4.5) this proves the statement. ∎

The state $\hat\rho^{\mathrm{I}}$ of the radiation field is said to have $M$***th-order coherence***[13] if there is a function $V(x)$ on $\mathcal{M}$ with

$$G^{(n,n)}_{\hat\rho^{\mathrm{I}}}(x_1;\ldots;x_{2n}) = \prod_{\nu=1}^{n}\left(V(x_\nu)\right)^* \prod_{\mu=n+1}^{2n} V(x_\mu) \tag{4.7}$$

for all $n \in \{1,\ldots,M\}$. It is said to have ***infinite-order coherence*** if it has $M$th-order coherence for all $M \in \mathbb{N}$.



[10] For additional properties see (Walls and Milburn, 1995, Section 3.2).

[11] Recall Sections 1.2.4 and 1.2.4.

[12] Comparison of (4.6) with (4.9) confirms: $\langle \hat n\rangle = |\alpha|^2$.

[13] Here we use the terminology of, e.g., (Klauder and Sudarshan, 1968; Walls and Milburn, 1995; Ficek and Swain, 2001) rather than that of, e.g., (Mandel and Wolf, 1995; Hariharan and Sanders, 1996).

**Remarks:**

1. In a more restricted sense only the
$$G_{\hat{\rho}}^{(n,n)}(\underline{x}_1; \ldots ; \underline{x}_n; \underline{x}_n; \ldots ; \underline{x}_1)$$
are relevant. But for 'distributed' detectors also the $G^{(n,n)}$ with different arguments are relevant.
2. According to (Mandel and Wolf, 1995, Section 12.5) also the $G_{\hat{\rho}^{\mathrm{I}}}^{(n,m)}$ with $n \neq m$ are experimentally accessible, in principle, if nonlinear dielectric media are exploited in the counting arrangement.
3. Both the pure (coherent) states
$$e^{-\frac{1}{2}|\alpha|^2} \sum_{n=0}^{\infty} \frac{1}{n!} \left(\alpha\, e^{i\,\varphi_n}\, \hat{a}^\dagger\right)^n \Omega \qquad (= \chi_{\hat{a}}(\alpha) \text{ für } \varphi_n = 0)$$
and the mixed state
$$\hat{\rho}_{\hat{a}}(|\alpha|) = e^{-|\alpha|^2} \sum_{n=0}^{\infty} \frac{|\alpha|^{2n}}{n!} \left| \frac{1}{\sqrt{n!}} \left(\hat{a}^\dagger\right)^n \Omega \right\rangle \left\langle \frac{1}{\sqrt{n!}} \left(\hat{a}^\dagger\right)^n \Omega \right|$$
have infinite order coherence.
4. For these states the probability $p(n)$ for exactly $n$ photons 'being present' is given by a Poisson distribution:
$$\begin{aligned} p(n) &= \frac{\langle n \rangle}{n!}\, e^{-\langle n \rangle} \\ \langle n \rangle &= |\alpha|^2 \qquad \text{(expectation value for the photon number)}\ . \end{aligned}$$

### 4.2.2 Correlation Functions of Single-Mode States

**Lemma 4.2.2** *For every* ***single-mode state****, i.e. every* $\hat{\rho}^{\mathrm{I}}$ *fulfilling*[14]
$$\hat{\rho}^{\mathrm{I}} = \hat{P}_{\mathcal{H}_{\hat{a}}}\, \hat{\rho}^{\mathrm{I}}\, \hat{P}_{\mathcal{H}_{\hat{a}}}$$
*for some* ***mode*** $\hat{a}$ *(as considered in 1.2.4),*
$$G_{\hat{\rho}^{\mathrm{I}}}^{(n,m)}(x_1; \ldots ; x_{n+m}) = g_{nm} \prod_{\nu=1}^{n} \Big(V(x_\nu)\Big)^* \prod_{\mu=n+1}^{n+m} V(x_\mu) \quad \forall\, n,m \in \mathbb{N}\,,\ x_1, \ldots, x_{n+m} \in \mathcal{M} \tag{4.8}$$



[14] As usual, we denote by $\hat{P}_{\mathcal{H}_{\hat{a}}}$ the orthogonal projection onto the subspace $\mathcal{H}_{\hat{a}}$ . The subspace $\mathcal{H}_{\hat{a}}$ was defined below Equation (1.82).

*holds with*

$$V(\mathbf{x},t,j) = \sqrt{\langle\hat{n}\rangle}\left[\hat{A}_0^{(+)^j}(\mathbf{x},t)\,,\,\hat{a}^\dagger\right]_- \quad , \quad \langle\hat{n}\rangle \stackrel{\text{def}}{=} \operatorname{Tr}\left(\hat{\rho}^{\mathrm{I}}\,\hat{a}^\dagger\hat{a}\right)\,. \tag{4.9}$$

*and*

$$g_{nm} \stackrel{\text{def}}{=} \frac{\operatorname{Tr}\left(\hat{\rho}^{\mathrm{I}}\,\left(\hat{a}^\dagger\right)^n\,\hat{a}^m\right)}{\langle\hat{n}\rangle^{(n+m)/2}} \quad \forall\, n,m\in\mathbb{N}\,. \tag{4.10}$$

**Outline of proof:**

$$\begin{aligned}
&\operatorname{Tr}\left(\hat{\rho}^{\mathrm{I}}\prod_{\nu=1}^{n}\hat{A}_0^{(-)^{j_\nu}}(\mathbf{x}_\nu,t_\nu)\prod_{\mu=n+1}^{n+m}\hat{A}_0^{(+)^{j_\mu}}(\mathbf{x}_\mu,t_\mu)\right)\\
&= \sum_{\gamma,\rho=0}^{\infty}\frac{1}{\gamma!\,\rho!}\left\langle\left(\hat{a}^\dagger\right)^\gamma\Omega\middle|\hat{\rho}^{\mathrm{I}}\left(\hat{a}^\dagger\right)^\rho\Omega\right\rangle\left\langle\left(\hat{a}^\dagger\right)^\rho\Omega\middle|\prod_{\nu=1}^{n}\hat{A}_0^{(-)^{j_\nu}}(\mathbf{x}_\nu,t_\nu)\cdot\right.\\
&\qquad\left.\cdot\prod_{\mu=n+2}^{n+m}\hat{A}_0^{(+)^{j_\mu}}(\mathbf{x}_\mu,t_\mu)\;\underbrace{\hat{A}_0^{(+)^{j_{n+1}}}(\mathbf{x}_{n+1},t_{n+1})\left(\hat{a}^\dagger\right)^\gamma\Omega}_{\stackrel{(1.82),(1.37)}{=}\langle\hat{n}\rangle^{-\frac{1}{2}}V(x_{n+1})\,\gamma\,(\hat{a}^\dagger)^{\gamma-1}\Omega}\right\rangle\\
&= \frac{1}{\langle\hat{n}\rangle^{\frac{n+m}{2}}}\sum_{\rho=n}^{\infty}\sum_{\gamma=m}^{\infty}\frac{1}{(\rho-n)!\,(\gamma-m)!}\prod_{\nu=1}^{n}\Big(V(x_\nu)\Big)^*\prod_{\mu=n+1}^{n+m}V(x_\mu)\left\langle\left(\hat{a}^\dagger\right)^\gamma\Omega\middle|\hat{\rho}^{\mathrm{I}}\left(\hat{a}^\dagger\right)^\rho\Omega\right\rangle\\
&\qquad\underbrace{\left\langle\left(\hat{a}^\dagger\right)^{\rho-n}\Omega\mid\left(\hat{a}^\dagger\right)^{\gamma-m}\Omega\right\rangle}_{=(\rho-n)!\,\delta_{\rho,\gamma+n-m}}\\
&= \frac{1}{\langle\hat{n}\rangle^{\frac{n+m}{2}}}\prod_{\nu=1}^{n}\Big(V(x_\nu)\Big)^*\prod_{\mu=n+1}^{n+m}V(x_\mu)\underbrace{\sum_{\gamma=0}^{\infty}\frac{1}{\gamma!}\left\langle\left(\hat{a}^\dagger\right)^{\gamma+m}\Omega\middle|\hat{\rho}^{\mathrm{I}}\left(\hat{a}^\dagger\right)^{\gamma+n}\Omega\right\rangle}_{=\operatorname{Tr}(\hat{a}^m\,\hat{\rho}^{\mathrm{I}}\,(\hat{a}^\dagger)^n)}\,.\qquad\blacksquare
\end{aligned}$$

**Theorem 4.2.3** *For every state $\hat{\rho}^{\mathrm{I}}$ of the radiation field the following three statements are equivalent:*[15]

1. $\left|G^{(1,1)}_{\hat{\rho}^{\mathrm{I}}}(x_1;x_2)\right|^2 = G^{(1,1)}_{\hat{\rho}^{\mathrm{I}}}(x_1;x_1)\,G^{(1,1)}_{\hat{\rho}^{\mathrm{I}}}(x_2;x_2) \quad \forall\,x_1,x_2\in\mathcal{M}\,.$

2. *$\hat{\rho}^{\mathrm{I}}$ has 1st-order coherence.*

3. *$\hat{\rho}^{\mathrm{I}}$ is a single-mode state.*

**Outline of proof:**[16] Assume the first statement to be valid and $G^{(1,1)}_{\hat{\rho}^{\mathrm{I}}}(x,y)$ sufficiently well-behaved.[17] Then, using the shorthand notation

$$G(x,y) \stackrel{\text{def}}{=} G^{(1,1)}_{\hat{\rho}^{\mathrm{I}}}(x,y)\,,$$

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[15] In this context it is essential that, for $n=1$, (4.7) is an **exact** equality. Unfortunately this can never be checked experimentally.

[16] We essentially follow (Klauder and Sudarshan, 1968, Section 8–2 B).

[17] Otherwise we could consider suitably 'smeared' versions of $G^{(1,1)}_{\hat{\rho}^{\mathrm{I}}}(x,y)$; see (4.17) and (4.18), below.

we have

$$|G(x,y)| = \sqrt{G(x,x)}\,\sqrt{G(y,y)}$$

and hence

$$G(x,y) = \underbrace{r(x)}_{\geq 0}\, r(y)\, e^{i\,\varphi(x,y)} \quad \forall\, x,y \in \mathbb{R}^3 \times \mathbb{R} \times \{1,2,3\} \tag{4.11}$$

for some real-valued function $\varphi(x,y)$ , where

$$r(x) \stackrel{\text{def}}{=} \sqrt{G(x,x)}\,.$$

Since

$$G(x,y) = \Big(G(y,x)\Big)^*\,,$$

$\varphi$ has to fulfill

$$\varphi(x,y) = -\varphi(y,x) \in \mathbb{R} \qquad \forall\, x,y \in \mathbb{R}^3 \times \mathbb{R} \times \{1,2,3\}\ . \tag{4.12}$$

Consider arbitrary $x_1, x_2, x_3 \in \mathbb{R}^3 \times \mathbb{R} \times \{1,2,3\}$ fulfilling

$$r(x_j) \neq 0 \quad \forall\, j \in \{1,2,3\} \tag{4.13}$$

and define

$$\begin{aligned} {M^j}_k &\stackrel{\text{def}}{=} e^{i\,\varphi(x_j,x_k)} \quad \forall\, j,k \in \{1,2,3\}\ , \\ \mathbb{M} &\stackrel{\text{def}}{=} (\,{M^j}_k\,)\ . \end{aligned} \tag{4.14}$$

Then (4.12) implies

$$\mathbb{M} = \mathbb{M}^* \tag{4.15}$$

and hence

$$\mathbb{M} = \begin{pmatrix} 1 & {M^1}_2 & {M^1}_3 \\ \left({M^1}_2\right)^* & 1 & {M^2}_3 \\ \left({M^1}_3\right)^* & \left({M^2}_3\right)^* & 1 \end{pmatrix}.$$

Therefore we have

$$\frac{1}{2}\,\det(\mathbb{M}) = \Re({M^1}_2{M^2}_3{M^3}_1) - 1$$

and, consequently,

$$\det(\mathbb{M}) \geq 0 \implies {M^1}_2{M^2}_3{M^3}_1 = 1\,. \tag{4.16}$$

Another consequence of (4.14) is

$$\begin{aligned} \sum_{j,k=1}^{3} \Big(\Big(r(x_j)\,\zeta_j\Big)\Big)^* \Big(r(x_k)\,\zeta_k\Big){M^j}_k &= \sum_{j,k=1}^{3} (\zeta_j)^*\,\zeta_k\, G(x_j,x_k) \\ &\geq 0 \quad \forall\, \zeta_1,\zeta,\zeta_3 \in \mathbb{C}\,, \end{aligned}$$

which, together with (4.13) and(4.15), implies

$$\det(\mathbb{M}) \geq 0\,.$$

The latter, together with (4.16) implies

$${M^1}_2{M^2}_3{M^3}_1 = 1$$

and hence

$$\varphi(x_1,x_2) + \varphi(x_2,x_3) + \varphi(x_3,x_1) = 0\,,$$

by (4.14). This, together with (4.12) and (4.14), shows that

$$\mathbb{M}^2 = 3\,\mathbb{M}\,.$$

Therefore, by (4.15), $\mathbb{M}/3$ is a projection operator, i.e.

$$M^j{}_k = A^j\,\left(A^k\right)^* \quad \forall\, j,k \in \{1,2,3\}$$

holds for suitable $A^1, A^2, A^3 \in \mathbb{C}$ with $\left|A^1\right|^2 + \left|A^2\right|^2 + \left|A^3\right|^2 = 3\,.$ Since

$$M^j{}_j = 1 \quad \forall\, j \in \{1,2,3\}\ ,$$

we have

$$M^j{}_k = e^{i\,\varphi_j}\,e^{-i\,\varphi_k}$$

for suitable $\varphi_1, \varphi_2, \varphi_3 \in \mathbb{R}\,.$ Together with (4.14) this implies

$$\varphi(x_j, x_k) = \varphi_j - \varphi_k \quad \forall\, j,k \in \{1,2,3\}$$

and hence

$$r(x)\,r(y) \neq 0 \implies \varphi(x,y) = \varphi(x) - \varphi(y) \qquad \forall\, x,y \in \mathbb{R}^3 \times \mathbb{R} \times \{1,2,3\}$$

for some real-valued $\varphi(x)\,.$ This, together with (4.11), finally implies

$$G(x,y) = \Big(V(x)\Big)^*\,V(y) \qquad \forall\, x,y \in \mathbb{R}^3 \times \mathbb{R} \times \{1,2,3\}$$

with

$$V(x) \stackrel{\text{def}}{=} r(x)\,e^{i\,\varphi(x)} \quad \forall\, x \in \mathbb{R}^3 \times \mathbb{R} \times \{1,2,3\}\ .$$

This proves, that the first statement implies the second one.

Now assume that the second statement is valid. Then, since[18]

$$\begin{aligned}
\hat a_{\mathbf g} &\stackrel{\text{def}}{=} \int \Big(\mathbf g(\mathbf x)\Big)^* \cdot \hat{\mathbf A}_0^{(+)}(\mathbf x, 0)\,dV_{\mathbf x} \\
&\underset{(1.30)}{=} \sqrt{\mu_0\,\hbar\,c}\sum_{j=1}^{2}\Big(\mathbf g(\mathbf k)\Big)^* \cdot \boldsymbol\epsilon_j(\mathbf k)\,\hat a_j(\mathbf k)\,\frac{dV_{\mathbf k}}{\sqrt{|\mathbf k|}}
\end{aligned} \tag{4.17}$$

shows that all modes (1.58) are of the form $\hat a_{\mathbf g}$ , there is a sequence of complex numbers $z_1, z_2, \ldots$ with

$$\mathrm{Tr}(\hat\rho^{\mathrm I}\,\hat a_\nu^\dagger\,\hat a_\mu) = (z_\nu)^*\ z_\mu \quad \forall\, \nu,\mu \in \mathbb{Z}\,. \tag{4.18}$$

We need only consider the physically relevant case

$$0 < \sum_{\nu=1}^{\infty} |z_\nu|^2 < \infty\,.$$

Then the definition

$$\hat b \stackrel{\text{def}}{=} \frac{\sum_{\mu=1}^{\infty} (z_\nu)^*\ \hat a_\nu}{\sqrt{\sum_{\nu=1}^{\infty} |z_\nu|^2}} \tag{4.19}$$



[18]We denote by $\mathbf g(\mathbf k)$ the FOURIER transform of $\mathbf g(\mathbf x)$ :

$$\mathbf g(\mathbf k) \stackrel{\text{def}}{=} (2\pi)^{-3/2}\int \mathbf g(\mathbf x)\,e^{-i\,\mathbf x\cdot\mathbf k}\,dV_{\mathbf x} \quad \forall\, \mathbf k \in \mathbb{R}^3\,.$$

is allowed and specifies a mode $\hat{b}$ $\left(\left|\hat{b}^\dagger\,\Omega\right| = 1\right)$. We may choose a complete system of modes $\left\{\hat{b}_\nu : \nu \in \mathbb{N}\right\}$ (of type (1.58)) with

$$\hat{b}_1 = \hat{b}\,. \tag{4.20}$$

Then

$$\hat{b}_\mu = \sum_{\nu=1}^{\infty} u_{\mu\nu}\,\hat{a}_\nu \quad \forall\,\mu \in \mathbb{N} \tag{4.21}$$

holds for suitable complex $u_\mu\nu$ fulfilling

$$\sum_{\lambda=1}^{\infty}\Bigl(u_{\mu\lambda}\Bigr)^* u_{\nu\lambda} = \delta_{\mu\nu} \quad \forall\,\mu,\nu \in \mathbb{N}\,. \tag{4.22}$$

(4.19)–(4.21) imply

$$u_{1\nu} = \frac{(z_\nu)^*}{\sqrt{\sum_{\nu=1}^{\infty}|z_\nu|^2}} \quad \forall\,\nu \in \mathbb{N}\,. \tag{4.23}$$

We conclude that

$$\begin{aligned}
\mathrm{Tr}(\hat{\rho}^{\mathrm{I}}\,\hat{b}_\lambda^\dagger\,\hat{b}_\lambda) &\underset{(4.21),(4.18)}{=} \sum_{\nu,\mu=1}^{\infty}\Bigl(u_{\lambda\mu}\,z_\mu\Bigr)^*(u_{\lambda\nu}\,z_\nu)\\
&\underset{(4.23)}{=} \underbrace{\sum_{\nu,\mu=1}^{\infty}(u_{\lambda\mu})^*\,u_{1\mu}\,u_{\lambda_\nu}\,(u_{1\nu})^*}_{\underset{(4.23)}{=}\,\delta_{1\lambda}}\sum_{\nu=1}^{\infty}|z_\nu|^2
\end{aligned}$$

and hence

$$\mathrm{Tr}(\hat{\rho}^{\mathrm{I}}\,\hat{b}_\lambda^\dagger\,\hat{b}_\lambda) = 0 \quad \forall\,\lambda > 1\,.$$

This shows that $\hat{\rho}^{\mathrm{I}}$ characterizes a single mode state with mode $\hat{b}_1$.

That the third statement implies the first one is a direct consequence of 4.2.2. ∎

A direct consequence of Lemma 4.2.2 and (1.63) is:

$$g_{\nu\nu} = \underbrace{\frac{n\,(n-1)\cdots\Bigl(n-(\nu-1)\Bigr)}{n^\nu}}_{=0 \ \text{for}\ \nu>n} \quad \text{for single-mode } n\text{-photon states with } n \neq 0\,.$$

This especially shows:

> Single-mode $n$-photon states with $n \neq 0$ have coherence of only first order.[19]



[19]Since for these states we have $g_{11} = 1 \neq \dfrac{n-1}{n} = g_{22}$.

Lemma 4.2.2 also shows that[20]

$$g_{nn} = n! \quad \text{for } \hat\rho \propto e^{-\beta\,\hat a^\dagger\,\hat a}\,,\ \beta > 0\,.$$

Thus:

Also the single-mode thermal states have coherence of only first order.

**Corollary 4.2.4** *The state $\hat\rho^{\mathrm{I}}$ of the radiation field has infinite-order coherence iff*

$$\left|G^{(n,n)}_{\hat\rho^{\mathrm{I}}}(x_1;\ldots;x_{2n})\right|^2 = \prod_{\mu=1}^{2n} G^{(1,1)}_{\hat\rho^{\mathrm{I}}}(x_\mu;x_\mu) \quad \forall\, n\in\mathbb{N}\,,\ x_1,\ldots,x_{n+m}\in\mathcal{M}\,.$$

**Corollary 4.2.5** *If $\hat\rho^{\mathrm{I}}$ is a state describing photons of the single mode $\hat a$ then $\hat\rho^{\mathrm{I}}$ has infinite-order coherence iff*[21]

$$p_{\hat\rho^{\mathrm{I}}}(m) = \frac{\langle\hat n\rangle^m}{m!}\,e^{-\langle\hat n\rangle} \quad \forall\, m\in\mathbb{Z}_+\,, \tag{4.24}$$

*where*

$$p_{\hat\rho^{\mathrm{I}}}(m) \stackrel{\mathrm{def}}{=} \mathrm{Tr}\left(\hat\rho^{\mathrm{I}}\,\frac{1}{m!}\left|\left(\hat a^\dagger\right)^m\Omega\right\rangle\!\left\langle\left(\hat a^\dagger\right)^m\Omega\right|\right) \tag{4.25}$$



[20]Since

$$\begin{aligned}
&\mathrm{Tr}\Big(e^{-\beta\,\hat a^\dagger\,\hat a}(\hat a^\dagger)^n\hat a^n\Big)\\
&\underset{(1.63)}{=} \mathrm{Tr}\bigg(\Big(e^{-\beta\,\hat a^\dagger\,\hat a}\,\big(\hat a^\dagger\,\hat a\big)\big(\hat a^\dagger\,\hat a-1\big)\ldots\big(\hat a^\dagger\,\hat a-(n-1)\big)\Big)\bigg)\\
&\underset{(1.64)}{=} \sum_{\mu=0}^\infty \frac{1}{\mu!}\Big\langle(\hat a^\dagger)^\mu\,\Omega\,\Big|\,e^{-\beta\,\hat a^\dagger\,\hat a}\,\big(\hat a^\dagger\,\hat a\big)\big(\hat a^\dagger\,\hat a-1\big)\ldots\big(\hat a^\dagger\,\hat a-(n-1)\,(\hat a^\dagger)^\mu\,\Omega\Big\rangle\\
&= \sum_{\mu=0}^\infty \lambda^\mu\,\mu\,(\mu-1)\ldots\big(\mu-(n-1)\big)\\
&= \lambda^n\left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^n\underbrace{\sum_{\mu=0}^\infty\lambda^\mu}_{=1/(1-\lambda)}\\
&= \lambda^n\,\frac{n!}{(1-\lambda)^{n+1}}\\
&= \frac{n!\,e^{-n\,\beta}}{\left(1-e^{-\beta}\right)^{n+1}}\,,
\end{aligned}$$

and, therefore, especially

$$\mathrm{Tr}\left(e^{-\beta\,\hat a^\dagger\,\hat a}\right) = \frac{1}{1-e^{-\beta}}\,,\quad \mathrm{Tr}\left(e^{-\beta\,\hat a^\dagger\,\hat a}\,\hat N\right) = \frac{e^{-\beta}}{\left(1-e^{-\beta}\right)^2}\,.$$

[21]In other words: $m\mapsto p_{\hat\rho^{\mathrm{I}}}(m)$ is a Poisson ***distribution***.

*denotes the probability for exactly $m$ photons 'being present' in this state and*

$$\langle\hat{n}\rangle \stackrel{\text{def}}{=} \mathrm{Tr}\left(\hat{\rho}^{\mathrm{I}}\,\hat{a}^\dagger\hat{a}\right) .$$

**Outline of proof:** From the general relations

$$\begin{aligned}
\sum_{\nu=0}^{\infty}(1+\lambda)^\nu\, p_{\hat{\rho}^{\mathrm{I}}}(\nu) &= \sum_{\nu=0}^{\infty}(1+\lambda)^\nu\,\frac{1}{\nu!}\left\langle\left(\hat{a}^\dagger\right)^\nu\Omega\mid\hat{\rho}^{\mathrm{I}}\,\left(\hat{a}^\dagger\right)^\nu\Omega\right\rangle \\
&= \sum_{\nu=0}^{\infty}\frac{1}{\nu!}\left\langle\left(\hat{a}^\dagger\right)^\nu\Omega\mid\hat{\rho}^{\mathrm{I}}\,e^{\nu\,\ln(1+\lambda)}\left(\hat{a}^\dagger\right)^\nu\Omega\right\rangle \\
&= \sum_{\nu=0}^{\infty}\frac{1}{\nu!}\left\langle\left(\hat{a}^\dagger\right)^\nu\Omega\mid\hat{\rho}^{\mathrm{I}}\,e^{\ln(1+\lambda)\,\hat{a}^\dagger\hat{a}}\left(\hat{a}^\dagger\right)^\nu\Omega\right\rangle \\
&= \mathrm{Tr}\left(\hat{\rho}^{\mathrm{I}}\,e^{\ln(1+\lambda)\,\hat{a}^\dagger\hat{a}}\right) \\
&\underset{\text{Lemma ??}}{=} \mathrm{Tr}\left(\hat{\rho}^{\mathrm{I}}:e^{\lambda\,\hat{a}^\dagger\hat{a}}:\right) \qquad (4.26)
\end{aligned}$$

and

$$p_{\hat{\rho}^{\mathrm{I}}}(m) = \frac{1}{m!}\left(\left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^m\sum_{\nu=0}^{\infty}(1+\lambda)^\nu\,p_{\hat{\rho}^{\mathrm{I}}}(\nu)\right)_{|\lambda=-1}$$

we conclude that

$$p_{\hat{\rho}^{\mathrm{I}}}(m) = \frac{1}{m!}\left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^m\sum_{\nu=0}^{\infty}\frac{\lambda^\nu}{\nu!}\,\mathrm{Tr}\left(\hat{\rho}^{\mathrm{I}}\,\left(\hat{a}^\dagger\right)^\nu\hat{a}^\nu\right)_{|\lambda=-1} \qquad \forall\, m\in\mathbb{Z}_+\,. \qquad (4.27)$$

If $\hat{\rho}^{\mathrm{I}}$ has infinite-order coherence, then (4.27) and Lemma 4.2.2 imply (4.24). Conversely, if (4.24) holds then

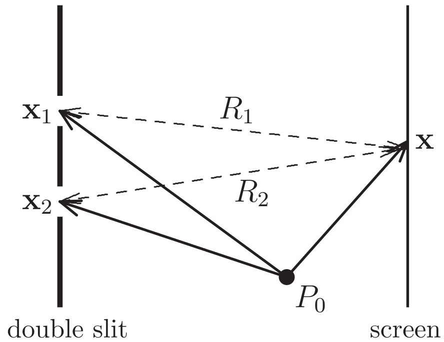
$$\begin{aligned}
\mathrm{Tr}\left(\hat{\rho}^{\mathrm{I}}\,\left(\hat{a}^\dagger\right)^m\hat{a}^m\right) &= \left(\left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^m\mathrm{Tr}\left(\hat{\rho}^{\mathrm{I}}:e^{\lambda\,\hat{a}^\dagger\hat{a}}:\right)\right)_{|\lambda=0} \\
&\underset{(4.26)}{=} \left(\left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^m\sum_{\nu=0}^{\infty}(1+\lambda)^\nu\,p_{\hat{\rho}^{\mathrm{I}}}(\nu)\right)_{|\lambda=0} \\
&\underset{(4.24)}{=} \left(\left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^m\sum_{\nu=0}^{\infty}(1+\lambda)^\nu\,\frac{\langle\hat{n}\rangle^\nu}{\nu!}\right)_{|\lambda=0} e^{-\langle\hat{n}\rangle} \\
&= \left(\left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^m e^{\lambda\langle\hat{n}\rangle}\right)_{|\lambda=0} \\
&= \langle\hat{n}\rangle^m
\end{aligned}$$

together with Lemma 4.2.2 implies that $\hat{\rho}^{\mathrm{I}}$ has infinite-order coherence. ∎

## 4.3 Simple Interference Experiments

### 4.3.1 YOUNG's Double-Slit Experiment

For the 'distributed' detector of YOUNG's double-slit experiment




the first order correlation function $G^{(1,1)}_{\hat{\rho}_{\text{in}}}(\underline{x}_1;\underline{x}_2)$ of the quantized radiation field **before the apertures** determines the counting rates.

One can show[22] that the ***first order degree of coherence***

$$g^{(1)}_{\hat{\rho}_{\text{in}}}(\underline{x}_1;\underline{x}_2) \stackrel{\text{def}}{=} \frac{G^{(1,1)}_{\hat{\rho}_{\text{in}}}(\underline{x}_1;\underline{x}_2)}{\sqrt[+]{G^{(1,1)}_{\hat{\rho}_{\text{in}}}(\underline{x}_1;\underline{x}_1)\, G^{(1,1)}_{\hat{\rho}_{\text{in}}}(\underline{x}_2;\underline{x}_2)}}$$

determines the ***visibility***

$$\mathcal{V} \stackrel{\text{def}}{=} \frac{\langle I\rangle_{\text{max}} - \langle I\rangle_{\text{min}}}{\langle I\rangle_{\text{max}} + \langle I\rangle_{\text{min}}}\,.$$

For instance, we have

$$\mathcal{V}(\mathbf{x},t) \approx \left| g^{(1)}_{\hat{\rho}_{\text{in}}}\left(\mathbf{x}_1, t-\frac{R_1}{c}, 3\,;\, \mathbf{x}_2, t-\frac{R_2}{c}, 3\right)\right| \quad \text{for vertical polarization}\,.$$

### 4.3.2 HANBURY-BROWN-TWISS Effect

For the so-called HANBURY-BROWN-TWISS ***effect*** (interference of photocurrents) the second order correlation function

$$G^{(2,2)}_{\hat{\rho}_{\text{in}}}(\underline{x}_1;\ldots;\underline{x}_4) \quad \text{mit } (\underline{x}_3;\underline{x}_4) = (\underline{x}_2;\underline{x}_1)$$

is the relevant entity. Note that, for independent simple detectors,

$$P_{\hat{\rho}_{\text{in}}}(\mathbf{x}_1,t_1;\mathbf{x}_2,t_2) \propto \left\{ \begin{array}{l} \text{product of the photocurrents} \\ \text{of both detectors} \end{array} \right.$$



[22] See, e.g. (Mandel and Wolf, 1995, Sect. 4.3.1).

**if**

$$\eta_\nu^{j_\nu j'_\nu} \propto \delta_{j_\nu j'_\nu} \quad \forall\, \nu \in \{1,2\}\,,\; j_\nu, j'_\nu \in \{1,2,3\}\,.$$

Especially for coherent states of the form

$$\begin{aligned}\chi &= e^{-\frac{|\alpha|^2+|\beta|^2}{2}}\, e^{\alpha\,\hat a^\dagger + \beta\,\hat b^\dagger}\Omega \\ &= \left(e^{-\frac{|\alpha|^2}{2}}\, e^{\alpha\,\hat a^\dagger}\Omega\right) \otimes \left(e^{-\frac{|\beta|^2}{2}}\, e^{\beta\,\hat b^\dagger}\Omega\right) \\ &\mathrel{\widehat{=}} \text{double light source}\end{aligned}$$

with $\hat a \perp \hat b$ we have

$$\begin{aligned}\hat A^{(+)^j}(\mathbf{x},t)\,\chi &= \left\langle \Omega \,\middle|\, \hat A^{(+)^j}(\mathbf{x},t)\left(\alpha\,\hat a^\dagger + \beta\,\hat b^\dagger\right)\Omega\right\rangle \chi \\ &= \left(A_a^{(+)^j}(\mathbf{x},t) + A_b^{(+)^j}(\mathbf{x},t)\right)\chi\end{aligned}$$

where

$$\begin{aligned}A_a^{(+)^j}(\mathbf{x},t) &\stackrel{\text{def}}{=} \left\langle \Omega \,\middle|\, \hat A^{(+)^j}(\mathbf{x},t)\,\alpha\,\hat a^\dagger\,\Omega\right\rangle, \\ A_b^{(+)^j}(\mathbf{x},t) &\stackrel{\text{def}}{=} \left\langle \Omega \,\middle|\, \hat A^{(+)^j}(\mathbf{x},t)\,\beta\,\hat b^\dagger\,\Omega\right\rangle.\end{aligned}$$

If

$$\begin{aligned}A_a^{(+)^j}(\mathbf{x},t) &\approx \underbrace{A_a^j}_{>0}\, e^{-i(\omega\, t - \mathbf{k}_a\cdot\mathbf{x} + \varphi_a)}\,, \\ A_b^{(+)^j}(\mathbf{x},t) &\approx \underbrace{A_b^j}_{>0}\, e^{-i(\omega\, t - \mathbf{k}_b\cdot\mathbf{x} + \varphi_b)}\end{aligned}$$

in the considered space-time region then this implies

$$\begin{aligned}G^{(2,2)}_{\hat\rho_{\rm in}}(\underline{x}_1;\underline{x}_2;\underline{x}_2;\underline{x}_1) &= \left|A_a^{j_1}\, e^{-i(\omega\, t_1 - \mathbf{k}_a\cdot\mathbf{x}_1 + \varphi_a)} + A_b^{j_1}\, e^{-i(\omega\, t_1 - \mathbf{k}_b\cdot\mathbf{x}_1 + \varphi_b)}\right| \times \\ &\qquad \times \left|A_a^{j_2}\, e^{-i(\omega\, t_2 - \mathbf{k}_a\cdot\mathbf{x}_2 + \varphi_a)} + A_b^{j_2}\, e^{-i(\omega\, t_2 - \mathbf{k}_b\cdot\mathbf{x}_2 + \varphi_b)}\right| \\ &= \left(\left|A_a^{j_1}\right|^2 + \left|A_b^{j_1}\right|^2 + A_a^{j_1}\, A_b^{j_1}\left(e^{-i(\varphi_a-\varphi_b)}\, e^{(\mathbf{k}_a-\mathbf{k}_b)\cdot\mathbf{x}_1} + \text{c.c.}\right)\right) \times \\ &\qquad \times \left(\left|A_a^{j_2}\right|^2 + \left|A_b^{j_2}\right|^2 + A_a^{j_2}\, A_b^{j_2}\left(e^{-i(\varphi_a-\varphi_b)}\, e^{(\mathbf{k}_a-\mathbf{k}_b)\cdot\mathbf{x}_2} + \text{c.c.}\right)\right)\end{aligned}$$

and hence

$$\begin{aligned}\left\langle G^{(2,2)}_{\hat\rho_{\rm in}}(\underline{x}_1;\underline{x}_2;\underline{x}_2;\underline{x}_1)\right\rangle &= \left(\left|A_a^{j_1}\right|^2 + \left|A_b^{j_1}\right|^2\right)\left(\left|A_a^{j_2}\right|^2 + \left|A_b^{j_2}\right|^2\right) \\ &\quad + 2\, A_a^{j_1}\, A_a^{j_2}\, A_b^{j_1}\, A_b^{j_2}\, \underbrace{\cos\Big((\mathbf{k}_a - \mathbf{k}_b)\cdot(\mathbf{x}_1 - \mathbf{x}_2)\Big)}_{\text{interference term!}},\end{aligned}$$

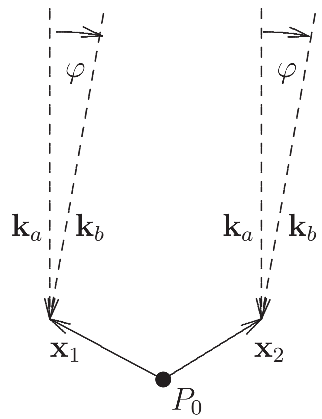
where by $\langle\ \rangle$ we indicate averaging over one (or both) of the angles $\varphi_a, \varphi_b$. Hence for

$$\hat\rho = \frac{e^{-|\alpha|^2-|\beta|^2}}{(2\pi)^2}\int_{\varphi_a=0}^{2\pi}\int_{\varphi_b=0}^{2\pi}\left|e^{\alpha\, e^{i\,\varphi_a}\,\hat a^\dagger + \beta\, e^{i\,\varphi_b}\,\hat b^\dagger}\Omega\right\rangle\left\langle e^{\alpha\, e^{i\,\varphi_a}\,\hat a^\dagger + \beta\, e^{i\,\varphi_b}\,\hat b^\dagger}\Omega\right|\, d\varphi_a\, d\varphi_b$$

we have

$$P_{\hat\rho}(\mathbf{x}_1,t_1;\mathbf{x}_2,t_2) \;\approx\; C_1 + C_2\,\cos\Big((\mathbf{k}_a-\mathbf{k}_b)\cdot(\mathbf{x}_1-\mathbf{x}_2)\Big)$$

with suitable constants $C_1, C_2$ . The interference term may be used to determine the angular distance of distant pairs of corresponding light sources:




$$
\begin{aligned}
&|\mathbf{k}_a| = |\mathbf{k}_b| = \tfrac{\omega}{c}\,,\\
&\angle\,(\mathbf{k}_a-\mathbf{k}_b,\mathbf{x}_1-\mathbf{x}_2) \ll 1\,,\\
&\underline{\underline{(\mathbf{k}_a-\mathbf{k}_b)\cdot(\mathbf{x}_1-\mathbf{x}_2)}} \;\approx\; |\mathbf{k}_a-\mathbf{k}_b|\,|\mathbf{x}_1-\mathbf{x}_2|\\
&\qquad\qquad\qquad\qquad\qquad\;\approx\; \underline{\underline{\varphi\,c\,\omega\,|\mathbf{x}_1-\mathbf{x}_2|}}\,.
\end{aligned}
$$

‘Corresponding light sources’ are, e.g.:

- Almost monochromatic lasers with stochastic phase $\Big(\widehat{=}\ \hat\rho_{\hat a}(|\alpha|),\hat\rho_{\hat b}(|\beta|)\Big)$ .
- Almost monochromatic thermal (single-mode) light sources (tar light).

Note that for ‘corresponding light sources’

$$G^{(1,1)}_{\hat\rho}(\underline{x};\underline{x}) = \text{konstant}$$

holds (within the considered space-time region) and hence the (spatial) interference is determined by the ***second order degree of coherence***[23]

$$g^{(2)}_{\hat\rho}(\underline{x}_1;\underline{x}_2) \;\stackrel{\text{def}}{=}\; \frac{G^{(2,2)}_{\hat\rho}(\underline{x}_1;\underline{x}_2;\underline{x}_2;\underline{x}_1)}{G^{(1,1)}_{\hat\rho}(\underline{x}_1;\underline{x}_1)\,G^{(1,1)}_{\hat\rho}(\underline{x}_2;\underline{x}_2)}\,.$$



[23] For the relation to $g^{(1)}_{\hat\rho}$ for stationary polarized light see (Mandel and Wolf, 1995, p. 708).

# Chapter 5

# Applications of Non-Classical Light

## 5.1 Criteria for Non-Classical Light

According to GLAUBER a state $\hat{\rho}$ of the radiation field is called ***classical*** if in Theorem 1.2.2 the $\rho_N$ can be chosen nonnegative.[1]

The optical equivalence theorem explains why interference experiments with typical interference pattern — as in YOUNG's double-slit experiment or the HANBURY-BROWN-TWISS effect — do not show any striking difference between classical and non-classical light.

Inequalities, however, which are valid for classical light may be violated by non-classical light, since inequalities are not invariant under averaging with non-positive weights.

For states $\hat{\rho}_{\text{Fock}}$ with strictly bounded photon number this is quite obvious, since they fulfill the condition

$$G^{(n,n)}_{\hat{\rho}_{\text{Fock}}} \equiv 0 \quad \text{for sufficiently large } n\,, \tag{5.1}$$

whereas for classical light we always have

$$\sum_{j_1,\ldots,j_n=1}^{3} \int G^{(n,n)}_{\hat{\rho}}(\underline{x}_1;\ldots;\underline{x}_n)\,\mathrm{d}\mathbf{x}_1\cdots\mathrm{d}\mathbf{x}_n > 0\,.$$

Unfortunately, preparation of states with strictly bounded photon number is impossible and, moreover, (5.1) cannot be rigorously checked.

Better suited for experimental checks is the inequality

$$\boxed{\begin{aligned}\left|G^{(2,2)}_{\hat{\rho}}(\underline{x}_1;\underline{x}_2;\underline{x}_2;\underline{x}_1)\right|^2 \quad &\leq \quad G^{(2,2)}_{\hat{\rho}}(\underline{x}_1;\underline{x}_1;\underline{x}_1;\underline{x}_1)\,G^{(2,2)}_{\hat{\rho}}(\underline{x}_2;\underline{x}_2;\underline{x}_2;\underline{x}_2)\\ &\qquad \text{for } \textbf{classical} \text{ states } \hat{\rho}\in\mathcal{B}(\mathcal{H}_{\text{field}})\,.\end{aligned}} \tag{5.2}$$



[1]See (Johansen, 2004) for critical remarks on GLAUBER's classicality criterion. See also (Miranowicz et al., 2010).

**Outline of proof for (5.2):** Let $\{\hat{a}_1, \hat{a}_2, \ldots\}$ be a maximal family of pairwise orthogonal modes and define

$$G^{(n,m)}_{\alpha_1,\ldots,\alpha_N}(\underline{x}_1; \ldots; \underline{x}_{n+m})$$
$$\stackrel{\text{def}}{=} \operatorname{Tr}\left( |\chi_{\alpha_1,\ldots,\alpha_N}\rangle\langle\chi_{\alpha_1,\ldots,\alpha_N}| \left(\prod_{\nu=1}^{n} \hat{A}^{(-)j_\nu}(\mathbf{x}_\nu, t_\nu)\right) \prod_{\mu=n-1}^{n+m} \hat{A}^{(+)j_\mu}(\mathbf{x}_\mu, t_\mu)\right).$$

Then
$$G^{(2,2)}_{\alpha_1,\ldots,\alpha_N}(\underline{x}_1; \underline{x}_2; \underline{x}_2; \underline{x}_1) = G^{(1,1)}_{\alpha_1,\ldots,\alpha_N}(\underline{x}_1; \underline{x}_1)\, \underbrace{G^{(1,1)}_{\alpha_1,\ldots,\alpha_N}(\underline{x}_2; \underline{x}_2)}_{\geq 0}.$$

For **classical** states this implies

$$\begin{aligned}
&\left| G^{(2,2)}_{\hat{\rho}}(\underline{x}_1; \underline{x}_2; \underline{x}_2; \underline{x}_1) \right|^2 \\
&= \lim_{N\to\infty} \left| \int \underbrace{\rho_N(\xi_1, \ldots, \eta_N)}_{\geq 0}\, G^{(2,2)}_{\xi_1+i\,\eta_1,\ldots,\xi_N+i\,\eta_N}(\underline{x}_1; \underline{x}_2; \underline{x}_2; \underline{x}_1) \frac{d\xi_1\, d\eta_1}{\pi} \cdots \frac{d\xi_N\, d\eta_N}{\pi} \right|^2 \\
&= \lim_{N\to\infty} \left| \int \left( \sqrt[+]{\rho_N(\xi_1, \ldots, \eta_N)}\, G^{(1,1)}_{\xi_1+i\,\eta_1,\ldots,\xi_N+i\,\eta_N}(\underline{x}_1; \underline{x}_1) \right)^* \times \right. \\
&\qquad \left. \times \sqrt[+]{\rho_N(\xi_1, \ldots, \eta_N)}\, G^{(1,1)}_{\xi_1+i\,\eta_1,\ldots,\xi_N+i\,\eta_N}(\underline{x}_2; \underline{x}_2) \frac{d\xi_1\, d\eta_1}{\pi} \cdots \frac{d\xi_N\, d\eta_N}{\pi} \right|^2 \\
&\underset{\text{SCHWARZ}}{\leq} \lim_{N\to\infty} \int \rho_N(\xi_1, \ldots, \eta_N) \underbrace{\left| G^{(1,1)}_{\xi_1+i\,\eta_1,\ldots,\xi_N+i\,\eta_N}(\underline{x}_1; \underline{x}_1) \right|^2}_{=G^{(2,2)}_{\xi_1+i\,\eta_1,\ldots,\xi_N+i\,\eta_N}(\underline{x}_1;\underline{x}_1;\underline{x}_1;\underline{x}_1)} \frac{d\xi_1\, d\eta_1}{\pi} \cdots \frac{d\xi_N\, d\eta_N}{\pi} \times \\
&\qquad \times \int \rho_N(\xi_1, \ldots, \eta_N) \underbrace{\left| G^{(1,1)}_{\xi_1+i\,\eta_1,\ldots,\xi_N+i\,\eta_N}(\underline{x}_2; \underline{x}_2) \right|^2}_{=G^{(2,2)}_{\xi_1+i\,\eta_1,\ldots,\xi_N+i\,\eta_N}(\underline{x}_2;\underline{x}_2;\underline{x}_2;\underline{x}_2)} \frac{d\xi_1\, d\eta_1}{\pi} \cdots \frac{d\xi_N\, d\eta_N}{\pi} \\
&= G^{(2,2)}_{\hat{\rho}}(\underline{x}_1; \underline{x}_1; \underline{x}_1; \underline{x}_1)\, G^{(2,2)}_{\hat{\rho}}(\underline{x}_2; \underline{x}_2; \underline{x}_2; \underline{x}_2). \quad \blacksquare
\end{aligned}$$

**Definition 5.1.1** *Let $\mathcal{G} = \mathcal{O} \times (\tau_1, \tau_2)$ be an open space-time region and let $\hat{\rho}^{\mathrm{I}}$ be a density operator on $\mathcal{H}_{\text{field}}$. Then $\hat{\rho}^{\mathrm{I}}$ is called **stationary in** $\mathcal{G}$ iff*[2]

$$G^{(n,m)}_{\hat{\rho}^{\mathrm{I}}}(x_1; \ldots; x_{n+m}) = G^{(n,m)}_{\hat{\rho}^{\mathrm{I}}}(x_1 + \tau; \ldots; x_{n+m} + \tau),$$

*where*
$$x_\nu + \tau \stackrel{\text{def}}{=} (\mathbf{x}_\nu, j_\nu, t_\nu + \tau),$$
*holds for all $n, m \in \mathbb{N}$ and for all $x_1, \ldots, x_{n+m} \in \mathcal{M}$ and $\tau \in \mathbb{R}$ with*

$$x_1, \ldots, x_{n+m}, x_1 + \tau, \ldots, x_{n+m} + \tau \in \mathcal{G}.$$

**Remark:** of course, also for stationary states there are stochastic fluctuations.

*Draft, November 5, 2011*

[2]It is stationary in all of space-time iff $e^{-\frac{i}{\hbar}\hat{H}_{\text{field}}\,\tau}\,\hat{\rho}^{\mathrm{I}}\,e^{+\frac{i}{\hbar}\hat{H}_{\text{field}}\,\tau} = \hat{\rho}^{\mathrm{I}} \quad \forall \tau \in \mathbb{R}$.

**Corollary 5.1.2** *Let $\mathcal{G}$ be as in Definition 5.1.1 and let $\hat{\rho}$ be a* **classical** *state of the radiation field that is* **stationary** *in $\mathcal{G}$. Then*

$$G^{(2,2)}_{\hat{\rho}}(x; x+\tau; x+\tau; x) \leq G^{(2,2)}_{\hat{\rho}}(x; x; x; x) \tag{5.3}$$

*holds for all $x \in \mathcal{M}$ and all $\tau \in \mathbb{R}$ with $x, x+\tau \in \mathcal{G}$.*

**Outline of proof:**

$$\Big(\underbrace{G^{(2,2)}_{\hat{\rho}}(\underline{x};\underline{x};\underline{x};\underline{x})}_{=\Big(\underbrace{G^{(1,1)}_{\hat{\rho}}(\underline{x};\underline{x})}_{\geq 0}\Big)^2}\Big)^2 \underset{\text{stat.}}{=} G^{(2,2)}_{\hat{\rho}}(\underline{x};\underline{x};\underline{x};\underline{x})\, G^{(2,2)}_{\hat{\rho}}(\underline{x}+\tau;\underline{x}+\tau;\underline{x}+\tau;\underline{x}+\tau)$$

$$\underset{(5.2)}{\geq} \Big|\underbrace{G^{(2,2)}_{\hat{\rho}}(\underline{x};\underline{x}+\tau;\underline{x}+\tau;\underline{x})}_{\geq 0}\Big|^2 . \qquad \blacksquare$$

While ***bunching***

$$G^{(2,2)}_{\hat{\rho}}(\underline{x};\underline{x}+\tau;\underline{x}+\tau;\underline{x}) < G^{(2,2)}_{\hat{\rho}}(\underline{x};\underline{x};\underline{x};\underline{x}) \,,$$

is possible for stationary classical light Corollary 5.1.2 shows that ***anti-bunching***

$$G^{(2,2)}_{\hat{\rho}}(\underline{x};\underline{x}+\tau;\underline{x}+\tau;\underline{x}) > G^{(2,2)}_{\hat{\rho}}(\underline{x};\underline{x};\underline{x};\underline{x}) \,,$$

is **not** possible for stationary classical light!

Bunching is typical for thermal light:

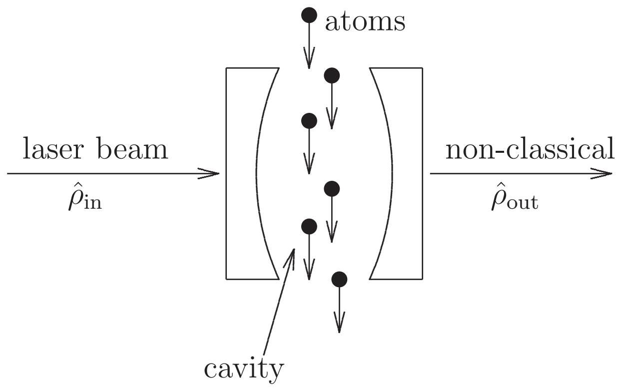
> If a detector fires then with high probability the intensity of the radiation field is considerable which — for coherent states — implies high probability for detection of additional photons at that instant. Therefore, strong stochastic fluctuations of the momentary coherent state of classical radiation support bunching, i.e.:
>
> > The probability for detecting a second photon almost immediately after a first one decreases with increasing time delay.

Anti-bunching is to be expected if one can arrange that single photons arrive in (essentially) fixed time intervals. This could be observed for resonance fluorescence.[3]



[3] See (Mandel and Wolf, 1995, Sect. 15.6.5) and (Kurtsiefer et al., 2000).

A typical experiment of this kind is described, e.g., in (Carmichael, 2001):




> Tuning of laser light and cavity to a certain atomic transition makes the states of the single atoms evolve in periodic cycles causing periodic stochastic fluctuations of the (stationary) radiation leaking out of the cavity. Naively formulated.
>
> > An atom that has just emitted a photon has to be reexcited before it can emit another photon.

In this case, by the way, also the inequality $g^{(2)}(0) \geq 1$ is violated which has to be fulfilled for classical light because of

$$\left\langle \left( A - \langle A \rangle \right)^2 \right\rangle = \left\langle A^2 \right\rangle - \langle A \rangle^2 \,.$$

## 5.2 The Action of Beam Splitters on Photons

### 5.2.1 General Action of Linear Optical Devices[4]

From experience we know that the following is a fairly accurate description of the influence of **linear** optical media on the quantized electromagnetic field:

> The expectation values of the quantized field are solutions of the classical macroscopic MAXWELL equations for the linear medium.

In this approximation the action of passive linear optical devices like beam splitters, phase shifters etc. is as follows:[5]

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[4] See also (Leonhardt, 2003).

[5] Note that the time evolution of the quantized electromagnetic field in the HEISENBERG picture fixes the Hamiltonian of the field up to an additive constant.

If $\{\hat{a}_\nu\}_{\nu\in\mathbb{N}}$ is a maximal family of pairwise orthogonal modes and if

$$\left\langle \Omega \,\middle|\, \hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\,\hat{a}_\nu^\dagger\,\Omega \right\rangle \longmapsto \left\langle \Omega \,\middle|\, \hat{\mathbf{A}}^{(+)}(\mathbf{x},t)\,\hat{b}_\nu^\dagger\,\Omega \right\rangle$$

agrees with the transformation of the complex vector potential in the corresponding classical optics (for every $\nu \in \mathbb{N}$) then the state transformation for the quantized electromagnetic field in the interaction picture is

$$f(\hat{a}_1^\dagger,\ldots,\hat{a}_2^\dagger,\ldots)\,\Omega \longmapsto f(\hat{b}^\dagger,\ldots,\hat{b}_2^\dagger,\ldots)\,\Omega$$

for sufficiently well-behaved $f$.

For coherent states this is a direct consequence of (1.84). Therefore, according to the optical equivalence theorem, this holds for all states of the quantized radiation field.

### 5.2.2 Classical Description of Lossless Beam Splitters

Consider the action of a *beam splitter*, as sketched in Figure 5.1, on an incoming monochromatic classical light beam corresponding to the complex vector potential

$$\mathbf{A}_{\mathrm{in}}^{(+)}(\mathbf{x},t) = (2\pi)^{-3/2}\sqrt{\mu_0\hbar c}\int \mathbf{a}_{\mathrm{in}}(\mathbf{k})e^{-i(\omega t-\mathbf{k}\cdot\mathbf{x})}\,\frac{dV_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\,,$$

where[6]

$$\begin{aligned}\mathbf{a}_{\mathrm{in}}(\mathbf{k}) &= z_{\mathrm{in}}^1\,\mathbf{a}_1^{\mathrm{V}}(\mathbf{k}) + z_{\mathrm{in}}^2\,\mathbf{a}_2^{\mathrm{V}}(\mathbf{k})\,,\\ \mathbf{a}_j^{\mathrm{V}}(\mathbf{k}) &\stackrel{\mathrm{def}}{=} \left(\frac{\omega}{c}\right)^{\frac{3}{2}}\delta(\mathbf{k}-\mathbf{k}_j)\,\mathbf{e}_2 \quad \forall\, j\in\{1,2\}\end{aligned} \tag{5.4}$$

and[7]

$$\mathbf{k}_1 \stackrel{\mathrm{def}}{=} \frac{\omega}{c}\,\frac{\mathbf{e}_1-\mathbf{e}_3}{\sqrt{2}}\,,\quad \mathbf{k}_2 \stackrel{\mathrm{def}}{=} \frac{\omega}{c}\,\frac{\mathbf{e}_1+\mathbf{e}_3}{\sqrt{2}}\,.$$

Linearity of the (plane) beam splitter implies that there are $r_1, r_2, t_1, t_2 \in \mathbb{C}$ such that the outcoming light beam corresponds to a complex vector potential

$$\mathbf{A}_{\mathrm{out}}^{(+)}(\mathbf{x},t) = (2\pi)^{-3/2}\sqrt{\mu_0\hbar c}\int \mathbf{a}_{\mathrm{out}}(\mathbf{k})e^{-i(\omega t-\mathbf{k}\cdot\mathbf{x})}\,\frac{dV_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}$$

with

$$\mathbf{a}_{\mathrm{out}}(\mathbf{k}) = z_{\mathrm{out}}^1\,\mathbf{a}_1^{\mathrm{V}}(\mathbf{k}) + z_{\mathrm{out}}^2\,\mathbf{a}_2^{\mathrm{V}}(\mathbf{k})$$



[6]The $z^j$ are just complex numbers without any physical dimension.

[7]Actually, it would be sufficient that $\mathbf{k}_1\cdot\mathbf{e}_3 = -\mathbf{k}_2\cdot\mathbf{e}_3$. Then, however the action of the beam splitter would depend on $\mathbf{k}_1\cdot\mathbf{e}_3$, in general.

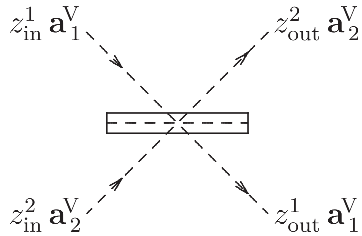



Figure 5.1: Classical Action of a Beam Splitter

and[8]

$$\begin{pmatrix} z_{\rm out}^1 \\ z_{\rm out}^2 \end{pmatrix} = \mathbb{S} \begin{pmatrix} z_{\rm in}^1 \\ z_{\rm in}^2 \end{pmatrix} , \quad \mathbb{S} \stackrel{\rm def}{=} \begin{pmatrix} t_1 & r_2 \\ r_1 & t_2 \end{pmatrix} .$$

For the fields

$$\begin{aligned} \mathbf{E}_{1,\rm in}(\mathbf{x},t) &\stackrel{\rm def}{=} (2\pi)^{-3/2}\sqrt{\frac{\mu_0\,\hbar\,\omega^2}{2c}}\left(i\,\omega\,z_{\rm in}^1\,e^{-i(\omega\,t-\mathbf{k}_1\cdot\mathbf{x})} - i\,\omega\,\overline{z_{\rm in}^1}\,e^{+i(\omega\,t-\mathbf{k}_1\cdot\mathbf{x})}\right)\mathbf{e}_2 \\ \mathbf{B}_{1,\rm in}(\mathbf{x},t) &\stackrel{\rm def}{=} (2\pi)^{-3/2}\sqrt{\frac{\mu_0\,\hbar\,\omega^2}{2c}}\left(i\,z_{\rm in}^1\,e^{-i(\omega\,t-\mathbf{k}_1\cdot\mathbf{x})} - i\,\overline{z_{\rm in}^1}\,e^{+i(\omega\,t-\mathbf{k}_1\cdot\mathbf{x})}\right)\mathbf{k}_1\times\mathbf{e}_2 \end{aligned}$$

corresponding to the complex vector

$$\mathbf{A}_{1,\rm in}^{(+)}(\mathbf{x},t) \stackrel{\rm def}{=} (2\pi)^{-3/2}\sqrt{\mu_0\hbar c}\int z_{\rm in}^1\mathbf{a}_1^{\rm V}(\mathbf{k})e^{-i(\omega\,t-\mathbf{k}\cdot\mathbf{x})}\,\frac{\mathrm{d}V_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}$$

with $\mathbf{a}_1^{\rm V}(\mathbf{k})$ given by (5.4) we have

$$\begin{aligned} \mathbf{E}_{1,\rm in}(\mathbf{x},t)\times\frac{1}{\mu_0}\mathbf{B}_{1,\rm in}(\mathbf{x},t) &= \frac{1}{\mu_0\,c}\,\mathbf{E}_{1,\rm in}(\mathbf{x},t)(\mathbf{x},t)\times\left(\frac{\mathbf{k}_1}{|\mathbf{k}_1|}\times\mathbf{E}_{1,\rm in}(\mathbf{x},t)\right) \\ &= \frac{1}{\mu_0\,c}\,|\mathbf{E}_{1,\rm in}(\mathbf{x},t)|^2\,\frac{\mathbf{k}_1}{|\mathbf{k}_1|} - \underbrace{\mathbf{E}_{1,\rm in}(\mathbf{x},t)\cdot\frac{\mathbf{k}_1}{|\mathbf{k}_1|}}_{=0}\,\mathbf{E}_{1,\rm in}(\mathbf{x},t)\,. \end{aligned}$$

Since[9]

$$\int_0^{\frac{2\pi}{\omega}}\left(i\,z_{\rm in}^1\,e^{-i(\omega\,t-\mathbf{k}_1\cdot\mathbf{x})} - i\,\overline{z_{\rm in}^1}\,e^{+i(\omega\,t-\mathbf{k}_1\cdot\mathbf{x})}\right)^2\mathrm{d}t = \frac{2\pi}{\omega}\left(\left|z_{\rm in}^1\right|^2 + \left|z_{\rm in}^1\right|^2\right)$$



[8] Of course, $\mathbb{S}$ depend on the frequency and also on the polarization of the incoming radiation.

[9] Note that

$$\int_0^{\frac{2\pi}{\omega}} e^{\pm i\,2\omega\,t}\,\mathrm{d}t = 0\,.$$

this gives for the ***intensity***, i.e. the modulus of the time-averaged energy current density, of this stationary field

$$\left| \frac{\omega}{2\pi} \int_0^{\frac{2\pi}{\omega}} \mathbf{E}_0(\mathbf{x}, t) \times \frac{1}{\mu_0} \mathbf{B}_0(\mathbf{x}, t)\, \mathrm{d}t \right| = (2\pi)^{-3} \frac{\hbar\, \omega^4}{2\, c^2} \left( \left| z^1_{\mathrm{in}} \right|^2 + \left| z^1_{\mathrm{in}} \right|^2 \right) .$$

Since similar results hold for the fields in the other arms of the interferometer, for a lossless beam splitter the condition

$$\left| z^1_{\mathrm{in}} \right|^2 + \left| z^2_{\mathrm{in}} \right| = \left| z^1_{\mathrm{out}} \right|^2 + \left| z^2_{\mathrm{out}} \right|$$

has to be fulfilled. This means that $\mathbb{S}$ has to be unitary, hence of the form

$$\mathbb{S} = \begin{pmatrix} \zeta^1 & -\overline{\zeta^2}\, e^{i\psi} \\ \zeta^2 & \overline{\zeta^1}\, e^{i\psi} \end{pmatrix} , \quad \psi \in \mathbb{R} ,\, \zeta^1, \zeta^2 \in \mathbb{C} ,\, \left|\zeta^1\right|^2 + \left|\zeta^2\right|^2 = 1 .$$

The latter may also be written in the form

$$\mathbb{S} = e^{i\,\varphi} \begin{pmatrix} a & -\bar{b} \\ b & \bar{a} \end{pmatrix} \quad \phi \in \mathbb{R} ,\, a, b \in \mathbb{C} ,\, |a|^2 + |b|^2 = 1 \tag{5.5}$$

(with $\varphi = \psi/2$ , $a = \zeta^1\, e^{-i\psi/2}$ , and $b = \zeta^2\, e^{-i\psi/2}$) or

$$\mathbb{S} = e^{i\,\varphi} \begin{pmatrix} a & \bar{b} \\ b & -\bar{a} \end{pmatrix} \quad \phi \in \mathbb{R} ,\, a, b \in \mathbb{C} ,\, |a|^2 + |b|^2 = 1$$

(with $\varphi = (\psi + \pi)/2 + \pi$ , $a = \zeta^1\, e^{-i(\psi+\pi)/2}$ , and $b = \zeta^2\, e^{-i(\psi+\pi)/2}$). Anyway, the ***transmittivity*** $T$ resp. the ***reflectivity*** $R$ is the same for both incoming rays:

$$R = |r_1|^2 = |r_2|^2 \;, \quad T = |t_1|^2 = |t_2|^2 \;, \quad R + T = 1 .$$

Beam splitters with $R = T$ are called ***50/50-beam splitter***s. Beam splitters with $r_1 = r_2$ and $t_1 = t_2$ are called ***symmetric***. Thus:

$$\mathbb{S} = \frac{e^{i\varphi}}{\sqrt{2}} \begin{pmatrix} 1 & \mp i \\ \mp i & 1 \end{pmatrix} \quad \text{for symmetric 50/50 beam splitters} \tag{5.6}$$

(see Exercise E34c) of (Lücke, eine)).

### 5.2.3 Transformation of Photon Modes

Let us describe the effect of beam splitters on photons as a scattering process in the interaction picture.[10]



[10] See also (Yurke et al., 1986).

The photon mode corresponding to the classical mode $\mathbf{a}_1^{\mathrm{V}}(\mathbf{k})$ resp. $\mathbf{a}_2^{\mathrm{V}}(\mathbf{k})$ is $\left(\frac{\omega}{c}\right)^{\frac{3}{2}}\hat{a}^{\mathrm{V}}(\mathbf{k}_1)$ resp. $\left(\frac{\omega}{c}\right)^{\frac{3}{2}}\hat{a}^{\mathrm{V}}(\mathbf{k}_2)$, where

$$\hat{a}^{\mathrm{V}}(\mathbf{k}) = \hat{a}_2(\mathbf{k}) \quad \forall\, \mathbf{k} \perp \mathbf{e}_2\,,$$

for the choice

$$\boldsymbol{\epsilon}_2(\mathbf{k}) \stackrel{\text{def}}{=} \mathbf{e}_2 \quad \forall\, \mathbf{k} \perp \mathbf{e}_2\,,$$

and the beam splitter acts according to[11]

$$\begin{aligned} \overline{z_{\text{in}}^1}\,\hat{a}^{\mathrm{V}}(\mathbf{k}_1) + \overline{z_{\text{in}}^2}\,\hat{a}^{\mathrm{V}}(\mathbf{k}_2) \quad &\longmapsto \quad \overline{z_{\text{out}}^1}\,\hat{a}^{\mathrm{V}}(\mathbf{k}_1) + \overline{z_{\text{out}}^2}\,\hat{a}^{\mathrm{V}}(\mathbf{k}_2) \\ &= \quad \left(t_1\,\overline{z_{\text{in}}^1} + r_2\,\overline{z_{\text{in}}^2}\right)\hat{a}^{\mathrm{V}}(\mathbf{k}_1) + \left(r_1\,\overline{z_{\text{in}}^1} + t_2\,\overline{z_{\text{in}}^2}\right)\hat{a}^{\mathrm{V}}(\mathbf{k}_2) \end{aligned}$$

for arbitrary $z_{\text{in}}^1, z_{\text{in}}^2 \in \mathbb{C}$. This is equivalent to linearity plus

$$\boxed{\begin{pmatrix} \hat{a}_-^{\mathrm{V}}(\mathbf{k}_1) \\ \hat{a}_-^{\mathrm{V}}(\mathbf{k}_2) \end{pmatrix} \stackrel{\text{def}}{=} \begin{pmatrix} \hat{a}^{\mathrm{V}}(\mathbf{k}_1) \\ \hat{a}^{\mathrm{V}}(\mathbf{k}_2) \end{pmatrix} \longmapsto \begin{pmatrix} \hat{a}_+^{\mathrm{V}}(\mathbf{k}_1) \\ \hat{a}_+^{\mathrm{V}}(\mathbf{k}_2) \end{pmatrix} \stackrel{\text{def}}{=} \underbrace{\begin{pmatrix} \overline{t_1} & \overline{r_1} \\ \overline{r_2} & \overline{t_2} \end{pmatrix}}_{=\mathbb{S}^* = \mathbb{S}^{-1}} \begin{pmatrix} \hat{a}^{\mathrm{V}}(\mathbf{k}_1) \\ \hat{a}^{\mathrm{V}}(\mathbf{k}_2) \end{pmatrix}.} \tag{5.7}$$

**Remark:** The scattering matrix $\hat{S}_0$ in the interaction picture fulfills

$$\hat{S}_0\,\Omega = \Omega$$

and

$$\hat{a}_+^{\mathrm{V}}(\mathbf{k}_j) = \hat{S}_0\,\hat{a}_-^{\mathrm{V}}(\mathbf{k}_j)\,\hat{S}_0^{-1} \quad \forall\, j \in \{1,2\}\,.$$

The scattering matrix $\hat{S}$ in the HEISENBERG picture — used, e.g., in (Mandel and Wolf, 1995, Section 12.12) and (Bowmeester et al., 2002, Chapter 4) — fulfills

$$\Omega_{\text{in}} = \hat{S}\,\Omega_{\text{out}}$$

and

$$\hat{a}_{\text{in}}^{\mathrm{V}}(\mathbf{k}_j) = \hat{S}\,\hat{a}_{\text{out}}^{\mathrm{V}}(\mathbf{k}_j)\,\hat{S}^{-1} \quad \forall\, j \in \{1,2\}\,,$$

where[12]

$$\begin{pmatrix} \hat{a}_{\text{in}}^{\mathrm{V}}(\mathbf{k}_1) \\ \hat{a}_{\text{in}}^{\mathrm{V}}(\mathbf{k}_2) \end{pmatrix} = \mathbb{S}^{-1} \begin{pmatrix} \hat{a}_{\text{out}}^{\mathrm{V}}(\mathbf{k}_1) \\ \hat{a}_{\text{out}}^{\mathrm{V}}(\mathbf{k}_2) \end{pmatrix}.$$

See Section 2.3.1 of (Lücke, qft) for a detailed discussion of the two pictures of scattering theory.



[11] Recall (1.58) and (1.60).

[12] Some authors mystify the fact that, e.g.,

$$\hat{a}_{\text{out}}^{\mathrm{V}}(\mathbf{k}_1) = t_1\,\hat{a}_{\text{in}}^{\mathrm{V}}(\mathbf{k}_1) + r_2\,\hat{a}_{\text{in}}^{\mathrm{V}}(\mathbf{k}_2)$$

holds even if the input situation at the lower arm of the beam splitter corresponds to the vacuum.

For (improper) incoming $n$-photon states (5.7) means that the beam splitter acts as

$$
\begin{aligned}
&\sum_{j_1,\ldots,j_n\in\{1,2\}} c_{j_1,\ldots,j_n}\left(\hat a^{\mathrm V}(\mathbf k_{j_1})\cdots\hat a^{\mathrm V}(\mathbf k_{j_n})\right)^\dagger\,\Omega\\
&\longmapsto \sum_{j_1,\ldots,j_n\in\{1,2\}} c_{j_1,\ldots,j_n}\left(\sum_{j_1'=1}^{2} S^{j_1'}{}_{j_1}\,\hat a^{\mathrm V}(\mathbf k_{j_1'})\right)^\dagger\cdots\left(\sum_{j_n'=1}^{2} S^{j_n'}{}_{j_n}\,\hat a^{\mathrm V}(\mathbf k_{j_n'})\right)^\dagger\Omega\,,
\end{aligned}
\tag{5.8}
$$

where

$$
\begin{pmatrix} S^1{}_1 & S^1{}_2\\ S^2{}_1 & S^2{}_2\end{pmatrix} \stackrel{\text{def}}{=} \begin{pmatrix} t_1 & r_2\\ r_1 & t_2\end{pmatrix}.
$$

Obviously, the total number of photons is not changed by the beam splitter.

Let us consider, for example, the special action[13]

$$
\begin{aligned}
\hat a^{\mathrm V}(\mathbf k_1)^\dagger\,\hat a^{\mathrm V}(\mathbf k_2)^\dagger\,\Omega \;&\longmapsto\; \frac12\left(\hat a^{\mathrm V}(\mathbf k_1)+i\,\hat a^{\mathrm V}(\mathbf k_2)\right)^\dagger\left(i\,\hat a^{\mathrm V}(\mathbf k_1)+\hat a^{\mathrm V}(\mathbf k_2)\right)^\dagger\\
&=\; \frac{i}{2}\left(\hat a^{\mathrm V}(\mathbf k_1)^\dagger\,\hat a^{\mathrm V}(\mathbf k_1)^\dagger+\hat a^{\mathrm V}(\mathbf k_2)^\dagger\,\hat a^{\mathrm V}(\mathbf k_2)^\dagger\right)\Omega
\end{aligned}
$$

of a (symmetric, 50/50-) beam splitter with

$$
\mathbb S = \frac{1}{\sqrt2}\begin{pmatrix} 1 & -i\\ -i & 1\end{pmatrix}. \tag{5.9}
$$

Here the incoming state is a 2-photon state with one photon definitely entering the beam splitter from above and the other photon definitely entering the beam splitter from below. As if agreed upon before, the photons leave the beam splitter either both on the upper side (with probability $\frac12$) or both on the lower side. This so-called Hong-Ou-Mandel ***effect*** effect is often coined *2-photon interference*,[14] even though the relative phase of the two photon modes is irrelevant, in this case. Needless to say, there is no classical analogue for this coalescence effect.

If the action of a beam splitter with $S$-matrix (5.9) does not depend on the polarization then it transforms the (improper) Bell ***states***

$$
\begin{aligned}
\Phi^{\pm}_{=}(\mathbf k_1,\mathbf k_2) &\stackrel{\text{def}}{=} \frac{1}{\sqrt2}\left(\hat a^{\mathrm V}(\mathbf k_1)\,\hat a^{\mathrm V}(\mathbf k_2)\pm\hat a^{\mathrm H}(\mathbf k_1)\,\hat a^{\mathrm H}(\mathbf k_2)\right)^\dagger\Omega\\
\Phi^{\pm}_{\neq}(\mathbf k_1,\mathbf k_2) &\stackrel{\text{def}}{=} \frac{1}{\sqrt2}\left(\hat a^{\mathrm V}(\mathbf k_1)\,\hat a^{\mathrm H}(\mathbf k_2)\pm\hat a^{\mathrm H}(\mathbf k_1)\,\hat a^{\mathrm V}(\mathbf k_2)\right)^\dagger\Omega\,,
\end{aligned}
\tag{5.10}
$$



[13]Recall Footnote 7. For more general input $\mathbf k$-configurations see (Scully and Zubairy, 1999, Sect. 4.4.3), where the Heisenberg picture is used.

[14]See (Di Giuseppe et al., 2003) for experimental verification and (McDonald and Wang, 2003; Lim and Beige, 2005) for generalization. This effect may be exploited for measurement of the spectral density matrix of 1-photon states (Wasilewski et al., 2007). See also (Kim and Grice, 2003) and (Zavatta et al., 2004) for additional aspects of two-photon interference. For an extensive treatment of two-photon interference in beam splitters see (Wang, 2003) and references given there.

where

$$\hat{a}^{\mathrm{H}}(\mathbf{k}) = \hat{a}_1(\mathbf{k})\,, \qquad \boldsymbol{\epsilon}_2(\mathbf{k}) = \mathbf{e}_2 \times \frac{\mathbf{k}}{|\mathbf{k}|} \quad \forall \underbrace{\mathbf{k}}_{\neq 0} \perp \mathbf{e}_2\,,$$

according to

$$\begin{aligned}
\Phi^{\pm}_{=}(\mathbf{k}_1,\mathbf{k}_2) &\longmapsto \frac{1}{2\sqrt{2}} \sum_{j=1}^{2} \left(\hat{a}^{\mathrm{V}}(\mathbf{k}_j)\,\hat{a}^{\mathrm{V}}(\mathbf{k}_j) + \hat{a}^{\mathrm{H}}(\mathbf{k}_j)\,\hat{a}^{\mathrm{H}}(\mathbf{k}_j)\right)^{\dagger}\Omega\,,\\
\Phi^{+}_{\neq}(\mathbf{k}_1,\mathbf{k}_2) &\longmapsto \frac{i}{\sqrt{2}} \sum_{j=1}^{2} \left(\hat{a}^{\mathrm{V}}(\mathbf{k}_j)\,\hat{a}^{\mathrm{H}}(\mathbf{k}_j)\right)^{\dagger}\Omega\,,\\
\Phi^{-}_{\neq}(\mathbf{k}_1,\mathbf{k}_2) &\longmapsto \frac{1}{\sqrt{2}} \left(\hat{a}^{\mathrm{V}}(\mathbf{k}_1)\,\hat{a}^{\mathrm{H}}(\mathbf{k}_2) - \hat{a}^{\mathrm{H}}(\mathbf{k}_1)\,\hat{a}^{\mathrm{V}}(\mathbf{k}_2)\right)^{\dagger}\Omega\,.
\end{aligned}$$

Note that $\Phi^{-}_{\neq}$ is the only BELL state that is transformed into a state with exactly one photon leaving the beam splitter on each side.[15] This effect was exploited in the first experiment on (partial) *quantum teleportation* (Bowmeester et al., 1997); see also (Gisin et al., 2003).

> **Comment:** By straightforward calculation (see 5.3.3) one can show that
>
> $$\begin{aligned}
> &\chi(\mathbf{k}_1) \otimes \Phi^{-}_{\neq}(\mathbf{k}_2,\mathbf{k}_3)\\
> &\quad = \frac{1}{2}\Big(\Phi^{\pm}_{=}(\mathbf{k}_1,\mathbf{k}_2) \otimes \hat{U}_1\,\chi(\mathbf{k}_3) - \Phi^{+}_{\neq}(\mathbf{k}_1,\mathbf{k}_2) \otimes \hat{U}_2\,\chi(\mathbf{k}_3)\\
> &\qquad\quad + \Phi^{-}_{=}(\mathbf{k}_1,\mathbf{k}_2) \otimes \hat{U}_3\,\chi(\mathbf{k}_3) - \Phi^{-}_{\neq}(\mathbf{k}_1,\mathbf{k}_2) \otimes \chi(\mathbf{k}_3)\Big)
> \end{aligned}$$
>
> holds for
>
> $$\chi(\mathbf{k}) \stackrel{\mathrm{def}}{=} \Big(\lambda_{\mathrm{V}}\,\hat{a}^{\mathrm{V}}(\mathbf{k}) + \lambda_{\mathrm{H}}\,\hat{a}^{\mathrm{H}}(\mathbf{k})\Big)^{\dagger}\Omega$$
>
> and
>
> $$\hat{U}_j\,\chi(\mathbf{k}) \stackrel{\mathrm{def}}{=} \begin{cases} \Big(-\lambda_{\mathrm{H}}\,\hat{a}^{\mathrm{V}}(\mathbf{k}) + \lambda_{\mathrm{V}}\,\hat{a}^{\mathrm{H}}(\mathbf{k})\Big)^{\dagger}\Omega & \text{for } j=1\,,\\ \Big(\lambda_{\mathrm{V}}\,\hat{a}^{\mathrm{V}}(\mathbf{k}) - \lambda_{\mathrm{H}}\,\hat{a}^{\mathrm{H}}(\mathbf{k})\Big)^{\dagger}\Omega & \text{for } j=2\,,\\ \Big(\lambda_{\mathrm{H}}\,\hat{a}^{\mathrm{V}}(\mathbf{k}) + \lambda_{\mathrm{V}}\,\hat{a}^{\mathrm{H}}(\mathbf{k})\Big)^{\dagger}\Omega & \text{for } j=3\,. \end{cases}$$
>
> This allows for quantum teleportation:
>
> Alice checks in which of the four BELL states photon 1 and 2 (corresponding to $\mathbf{k}_1$ and $\mathbf{k}_2$) are and communicates (by classical means) the result to Bob. If, e.g., Alice found $\Phi^{\pm}_{=}(\mathbf{k}_1,\mathbf{k}_2)$ then Bob just has to apply $\hat{U}_1^{-1}$ to photon 3 (corresponding to $\mathbf{k}_3$) in order to get its polarization state equal to the **unknown**[16] original polarization state of photon 1.

Of great practical use are also ***polarizing beam splitter***s, transmitting the horizontal modes and reflecting the vertical modes. Appropriately oriented, these

Draft, November 5, 2011

[15]See also (Pittman and Franson, 2003) in this connection.

[16]See (Caves et al., 2002) concerning the notion of *unknown state* in quantum mechanics.

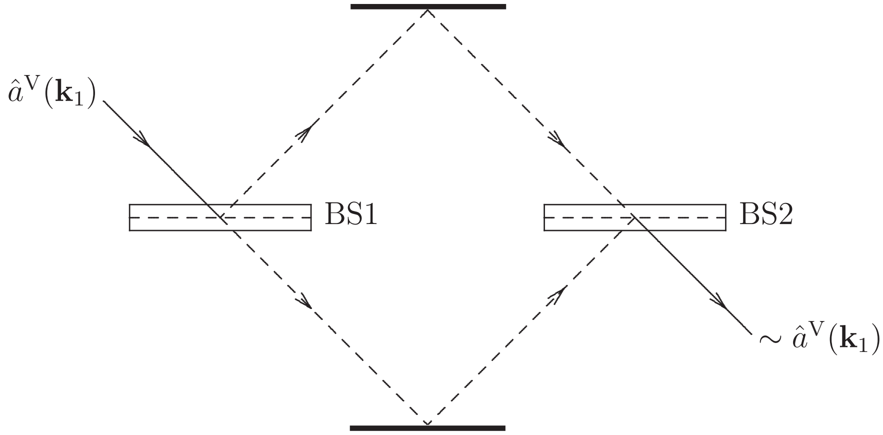



Figure 5.2: Balanced Mach-Zehnder interferometer

devices act according to

$$\boxed{\begin{pmatrix} \hat{a}^{\mathrm{H}}(\mathbf{k}_1) \\ \hat{a}^{\mathrm{H}}(\mathbf{k}_2) \\ \hat{a}^{\mathrm{V}}(\mathbf{k}_1) \\ \hat{a}^{\mathrm{V}}(\mathbf{k}_2) \end{pmatrix} \longmapsto \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix} \begin{pmatrix} \hat{a}^{\mathrm{H}}(\mathbf{k}_1) \\ \hat{a}^{\mathrm{H}}(\mathbf{k}_2) \\ \hat{a}^{\mathrm{V}}(\mathbf{k}_1) \\ \hat{a}^{\mathrm{V}}(\mathbf{k}_2) \end{pmatrix}} \tag{5.11}$$

### 5.2.4 Mach-Zehnder Interferometer

The quantum effect of beam splitters on one-photon states is already very puzzling. Consider, for example a Mach-Zehnder ***interferometer*** as sketched in Fig. 5.2.
Its action on an incoming single photon in the (improper) mode $\hat{a}^{\mathrm{V}}(\mathbf{k}_1)$ corresponds (up to irrelevant phase factors) to the following sequence of transformations:

$$\begin{aligned} \hat{a}^{\mathrm{V}}(\mathbf{k}_1)^\dagger\,\Omega \quad & \underset{\mathrm{BS1}}{\longmapsto} \quad \frac{1}{\sqrt{2}}\left(\hat{a}^{\mathrm{V}}(\mathbf{k}_1) + i\,\hat{a}^{\mathrm{V}}(\mathbf{k}_2)\right)^\dagger\Omega \\ & \underset{\mathrm{mirrors}}{\longmapsto} \quad \frac{1}{\sqrt{2}}\left(\hat{a}^{\mathrm{V}}(\mathbf{k}_2) + i\,\hat{a}^{\mathrm{V}}(\mathbf{k}_1)\right)^\dagger\Omega \\ & \underset{\mathrm{BS2}}{\longmapsto} \quad \frac{1}{2}\Big(\left(i\,\hat{a}^{\mathrm{V}}(\mathbf{k}_1) + \hat{a}^{\mathrm{V}}(\mathbf{k}_2)\right) + i\left(\hat{a}^{\mathrm{V}}(\mathbf{k}_1) + i\,\hat{a}^{\mathrm{V}}(\mathbf{k}_2)\right)\Big)^\dagger\Omega \\ & \quad = \quad i\,\hat{a}^{\mathrm{V}}(\mathbf{k}_1)^\dagger\,\Omega\,. \end{aligned}$$

For classical light there is no surprise:

> The interferometer is *balanced* in the sense that there is complete destructive interference for the waves transmitted through BS2 from below and reflected at BS2 from

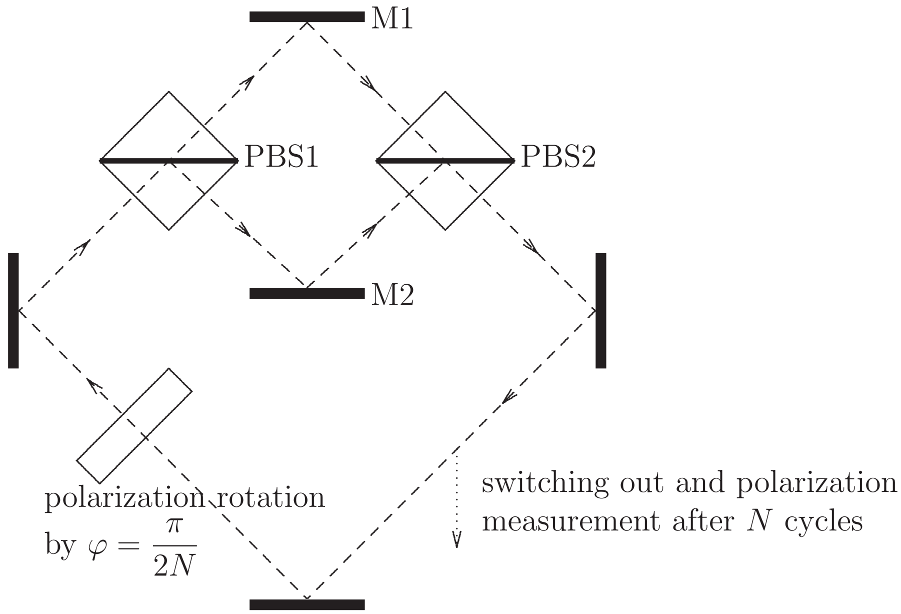



Figure 5.3: Principle of 'interaction free' measurements.

> above. If part of the waves is screened within the interferometer then this interference is disturbed and light is emitted also from the upper side of BS2.

For the quantized electromagnetic field in a one-photon state, however, there is an obvious problem:

> If, e.g., the lower path within the interferometer is blocked then, with equal probability, the photon may leave BS2 either on the upper or the lower side. Now the photon, considered as a particle, can only leave BS2 if it takes the upper path within the interferometer. Then, however, the photon should behave as if the lower path were not blocked at all, and there is no explanation for the photon's ability to leave BS2 on the upper side.

If the photon leaves the interferometer on the upper side of BS2 it 'knows' that the lower path is blocked although it never entered that region. Obviously, the photon is more than just a 'particle'. The described effect inspired ELITZUR and VAIDMAN to suggest what they coined *interaction free* measurements (Elitzur and Vaidman, 1993). The principle of a refined version, devised by KWIAT et al. (Kwiat et al., 1995; Kwiat et al., 1999), is sketched in Fig. 5.3:

> The interferometer is built with polarizing beam splitters rather than with polarization independent ones. A horizontally polarized photon will be inserted such that it enters the polarization rotator first. If there is no object inside the interferometer — built by PBS1, M1, PBS2, and M2 — then a photon entering the interferometer will pass it without any change of polarization. If, however, the path via M2 is blocked, then the photon will either be absorbed — with probability $\sin^2 \frac{\pi}{2N}$ — or pass with horizontal polarization. Thus, when the photon is switched out after $N$ cycles (if not

already absorbed) and checked for horizontal or vertical polarization, there are three possible outcomes:

1. The photon will not be detected at all. This may be due to failure of the detector or absorption by some object inside the interferometer. If an object is inserted as described, the probability for absorption of the photon is $N \sin^2 \frac{\pi}{2N}$ — which may be made arbitrarily small.
2. The photon will be detected with horizontal polarization. This can only happen if the absorber is inserted into the interferometer.
3. The photon will detected with vertical polarization. This can only happen if the absorber is not inserted into the interferometer.

This way, with probability arbitrarily close to 1, an object may be detected without interacting with it in any way.

## 5.3 Applications to Quantum Information Processing

### 5.3.1 Single-Photon States and Quantum Cryptography

In general, non-classical effect require sufficient — preferably individual — control of photons.

A standard technique for preparing single-photon states is sketched in Figure 5.4.

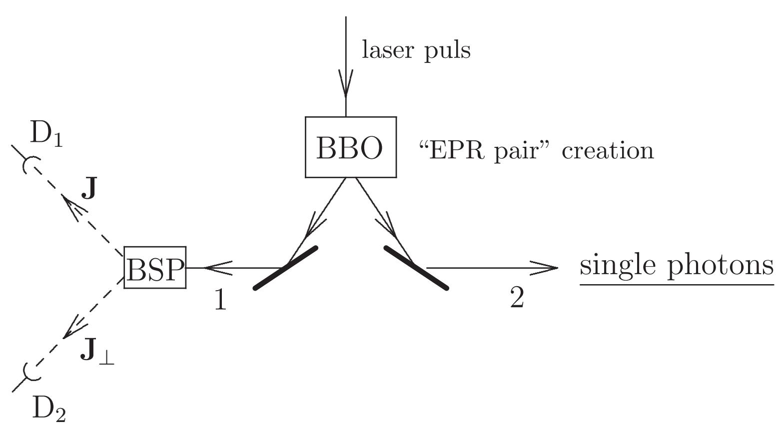



Figure 5.4: Possible preparation of single-photon states.

A laser pulse incident on a BBO (barium borate) crystal produces a pair of photon with polarization state

$$\Psi_- \stackrel{\text{def}}{=} \frac{1}{\sqrt{2}}\Big(\underbrace{\mathbf{V}}_{=\begin{pmatrix}1\\0\end{pmatrix}} \otimes \underbrace{\mathbf{H}}_{=\begin{pmatrix}0\\1\end{pmatrix}} - \mathbf{H}\otimes\mathbf{V}\Big)$$

via *spontaneous down conversion*[17] and this internal state fulfills the condition

$$\left.\begin{array}{l}\mathbf{J}\in\mathbb{C}^3\\ \|\mathbf{J}\|=1\end{array}\right\} \Longrightarrow \Psi_- = \frac{1}{\sqrt{2}}\left(\mathbf{J}\otimes\mathbf{J}_\perp - \mathbf{J}_\perp\otimes J\right) ,$$

where:

$$\begin{pmatrix}a\\b\end{pmatrix}_\perp \stackrel{\text{def}}{=} \begin{pmatrix}+b^*\\-a^*\end{pmatrix} \quad \forall\, a,b\in\mathbb{C}\,.$$

From this we conclude:

- The **internal** quantum state of the **ensemble** of those photons running to the right and having a partner making detector $D_2$ fire after passing a $\mathbf{J}$-$\mathbf{J}_\perp$ beam splitter has to be described (according to quantum mechanics) by the density operator $\hat\rho = |\mathbf{J}\rangle\langle\mathbf{J}|$.
- The **internal** quantum state of the **ensemble** of **all** photons running to the right — irrespective of the behavior of their partners — corresponds to

$$\hat\rho = \frac{1}{2}\,\hat 1 = \frac{1}{2}\,|\mathbf{J}\rangle\langle\mathbf{J}| + \frac{1}{2}\,|\mathbf{J}_\perp\rangle\langle\mathbf{J}_\perp|\ .$$

The result of a $\mathbf{J}$-$\mathbf{J}_\perp$ test ("which detector fires?") on photon 1 (left running) predetermines the result of an eventually following $\mathbf{J}$-$\mathbf{J}_\perp$ test on photon 2 (running to the right):

$$\begin{array}{lll}\mathbf{J}_\perp \text{ for photon 1} & \Longrightarrow & \mathbf{J} \text{ for photon 1}\\ \mathbf{J} \text{ for photon 1} & \Longrightarrow & \mathbf{J}_\perp \text{ for photon 1}\end{array}$$

(strict correlations).

If it were possible to produce exact copies of the unknown state of photon 2, owned by *Bob* (far away from *Alice* ), then Bob could produce an **ensemble** of photons with internal state $\mathbf{J}$ resp. $\mathbf{J}_\perp$ if photon 1, owned by *Alice*, just passed a $\mathbf{J}$-$\mathbf{J}_\perp$ beam splitter and made detector 2 resp. 1 fire. Since the states of (sufficiently large) ensembles can be determined (*quantum state tomography*) *Bob* could immediately check *Alice*'s choice of $\mathbf{J}$-$\mathbf{J}_\perp$ alternative. This could be exploited by *Alice* for superluminal communication of information to *Bob* encoded in different choices of $\mathbf{J}$-$\mathbf{J}_\perp$ alternatives.[18]

**Conclusion:**



[17] See, e.g., (Gatti et al., 2003) for details.
[18] See(Herbert, 1982) in this connection.

> The principles of special relativity exclude the possibility of producing exact copies of unknown quantum states (*no cloning theorem*).

The reason:

> Every attempt to gain information **not available a priori** about a quantum system usually results in uncontrollable changes of the system's state.

The no cloning theorem opens up the possibility of single-photon quantum cryptography:

If *Alice* and *Bob* both are provided with some random $N$-bit sequence

$$\mathbf{k} = (k_1, \ldots, k_N)$$

known only to them they can use this for encryption[19]

$$\mathbf{b} = (b_1, \ldots, b_N) \longmapsto \mathbf{b} \oplus \mathbf{k} \stackrel{\text{def}}{=} (b_1 \oplus k_1, \ldots, b_N \oplus k_N)$$

of any $N$-bit information $\mathbf{b}$. *Alice* can send the $N$-bit sequence $\mathbf{b}+\mathbf{k}$ through some public channel to *Bob* without being afraid that any eavesdropper could make use of this. *Bob* however can decrypt the message according to

$$\mathbf{b} \oplus \mathbf{k} \longmapsto (\mathbf{b} \oplus \mathbf{k}) \oplus \mathbf{k} = \mathbf{b}$$

(thanks to $\mathbf{k} \oplus \mathbf{k} = (0, \ldots, 0)$). Thus:

> The essential point for secure secret information exchange is the ability of *Alice* and *Bob* to create a common random ***key*** $\mathbf{k}$ known only to them.

A well established[20] technology for secret key distribution is based on the so-called ***BB84 protocol*** suggested by (Bennet and Brassard, 1984):

*Alice* and *Bob* agree upon two alternatives $\mathbf{J}$-$\mathbf{J}_\perp$ and $\mathbf{J}'$-$\mathbf{J}'_\perp$ with

$$\left|\langle \mathbf{J} \mid \mathbf{J}' \rangle\right|^2 = \frac{1}{2}\,.$$

Moreover they agree on certain instants of time at which *Alice* will send a photon to *Bob* with internal state being randomly chosen from the set $\{\mathbf{J}, \mathbf{J}_\perp, \mathbf{J}', \mathbf{J}'_\perp\}$. *Bob* randomly chooses a test of either the $\mathbf{J}$-$\mathbf{J}_\perp$ or the $\mathbf{J}'$-$\mathbf{J}'_\perp$ alternative for the received



[19] As usual, $\oplus$ means modulo-2 addition:

$$b \oplus k \stackrel{\text{def}}{=} (b+k) \bmod 2 \quad \forall\, b, k \in \{0, 1\}\,.$$

[20] See http://www.idquantique.com. For more recent implementations of quantum key distribution see (Sasaki et al., 2011) and references given there.

photon. After all photons are sent and tested *Alice* and *Bub* determine, via public communication, those instances for which by chance the had chosen the same alternative. For these instances their results are strictly correlated (in the ideal case) — unless the sending war perturbed (by an eavesdropper, for example). If these correlations are confirmed for certain randomly chosen instance then the other $N$ results, kept secret, provide the common key.

> The crucial point, however, is to make sure that (essentially) only **single** photons are sent[21] in order to prohibit — thanks to the no cloning theorem — eavesdropping.

### 5.3.2 Entangled Pairs of Photons

Figure 5.4 also serves as a good illustration of the following well-known consideration by Einstein, Rosen, and Podolsky (EPR):

1. According to EPR the result of any test of a $\mathbf{J}, \mathbf{J}_\perp$ alternative for photon 2 should be predetermined (*determinism*) since: Without influencing photon 2 in any way (*locality*) one <u>could</u> test the $\mathbf{J}, \mathbf{J}_\perp$ alternative on photon 1 and use the result to predict — thanks to the strict correlation of the photons — the outcome of a corresponding subsequent test on photon 2 with certainty.

2. From this point of view quantum mechanics appears to be incomplete (existence of *hidden variables*) since there is no quantum mechanical state predicting for all $\mathbf{J}$-$\mathbf{J}_\perp$ alternatives the outcome of a corresponding test with certainty.

This should be confronted with another well-know consideration by Bell which — in a slightly modified form — is as follows:

Let $\Phi$ be a (large but finite) ensemble of photon pairs in the state $\Psi_-$. Then, according to EPR the definition

$$\Phi_{\mathbf{J},\mathbf{J}'} \stackrel{\text{def}}{=} \left\{ \text{pairs from } \Phi \text{ with: } \left\{ \begin{array}{l} \mathbf{J} \text{ predeterminded for photon 1 and} \\ \mathbf{J}' \text{ predetermined for photon 2} \end{array} \right. \right\}$$

should be allowed (<u>determinism</u>) for every ensemble of EPR pairs (photons 1 and 2) and

$$\Phi_{\mathbf{J}} \stackrel{\text{def}}{=} \Phi_{\mathbf{J},\mathbf{J}'} \cup \Phi_{\mathbf{J},\mathbf{J}'_\perp}$$

should be independent of $\mathbf{J}'$ (<u>locality</u>). The <u>correlations</u> of the EPR pairs then imply

$$\boxed{\Phi_{\mathbf{J},\mathbf{J}'} = \Phi_{\mathbf{J}} \cap \Phi_{\mathbf{J}'} \quad \forall\, \mathbf{J}, \mathbf{J}'} \tag{5.12}$$



[21] See (Alleaume et al., 2003) and references given there for realizations of single-photon sources.

and, consequently, for all JONES vectors $\mathbf{J}_2, \mathbf{J}_2, \mathbf{J}_3$ the inequality[22]

$$\left|\Phi_{\mathbf{J}_1,\mathbf{J}_2}\right| \;\le\; \left|\Phi_{\mathbf{J}_1,\mathbf{J}_3}\right| + \left|\Phi_{\mathbf{J}_2,\mathbf{J}_{3\perp}}\right| \tag{5.13}$$

has to be fulfilled.

**Outline of proof:** The inequality follows from

$$\chi_1\,\chi_2 \le \chi_1\,\chi_3 + \chi_2\,(1-\chi_3) \quad \forall\,\chi_1,\chi_2,\chi_3 \in \{0,1\}\ ,$$

since[23]

$$|\Phi_{\mathbf{J}_\nu}| = \sum_{x\in\Phi_{\mathbf{J}_\nu}} \chi_{\Phi_{\mathbf{J}_\nu}}(x)\ , \quad |\Phi_{\mathbf{J}_{\nu\perp}}| = \sum_{x\in\Phi_{\mathbf{J}_\nu}} \left(1-\chi_{\Phi_{\mathbf{J}_\nu}}(x)\right)$$

and

$$\chi_{\Phi_{\mathbf{J}_\nu}\cap\Phi_{\mathbf{J}_\mu}}(x) = \chi_{\Phi_{\mathbf{J}_\nu}}(x)\,\chi_{\Phi_{\mathbf{J}_\mu}}(x)\,. \quad \blacksquare$$

Therefore (5.12) **contradicts** the quantum mechanical predictions

$$\boxed{\left.\begin{array}{l} |\Phi_{\mathbf{J}}|\,/\,|\Phi| = \dfrac{1}{2}\,, \\ |\Phi_{\mathbf{J}',\mathbf{J}_\perp}|\,/\,|\Phi| = \dfrac{1}{2}\,|\langle\mathbf{J}\mid\mathbf{J}'\rangle|^2 \end{array}\right\}\quad \forall\,\mathbf{J},\mathbf{J}'\,.} \tag{5.14}$$

This is because for, e.g.,

$$\mathbf{J}_\nu \stackrel{\text{def}}{=} \begin{pmatrix} \cos\left(\frac{\nu\,\pi}{8}\right) \\ \sin\left(\frac{\nu\,\pi}{6}\right) \end{pmatrix} \qquad \forall\,\nu\in\{1,2,3\}$$

(5.14) implies

$$\begin{aligned} \frac{|\Phi_{\mathbf{J}_1,\mathbf{J}_2}|}{|\Phi|} &= \frac{1}{2}\,\cos^2(\pi/6) \\ &= 3/8 \\ &\ge 2/8 \\ &= \frac{1}{2}\,\cos^2(\pi/3) + \frac{1}{2}\left(1-\cos^2(\pi/6)\right) \\ &= \frac{|\Phi_{\mathbf{J}_1,\mathbf{J}_3}| + |\Phi_{\mathbf{J}_2,\mathbf{J}_{3\perp}}|}{|\Phi|} \end{aligned}$$

rather than (5.13). Naive proposal:



[22]By $|\Phi_\nu|$ we denote the number of elements of the set $\Phi_\nu$ .

[23]As usual we denote by $\chi_{\Phi'}$ the *characteristic function* of the set $\Phi'$ :

$$\chi_{\Phi'}(x) \stackrel{\text{def}}{=} \begin{cases} 1 & \text{for } x\in\Phi'\,, \\ 0 & \text{sonst}\,. \end{cases}$$

Check (5.14) experimentally.

The vast majority of physicists considers (5.14) to be experimentally confirmed[24] and conclude:

> *Local determinism as assumed by EPR is inconsistent with physical reality.*

Concerning the actually remaining **loopholes** see, e.g., (Gisin and Gisin, 1999), (Semenov and Vogel, 2010), (Morgan, 2008), (Stobińska et al., 2010), (Gerhardt et al., 2011), and (Wittmann et al., 2011).

> *Generally speaking, we could take the attitude that every "no-go theorem" comes with some small-print, that is, certain conditions and assumptions that are considered totally natural and reasonable by the authors, but which may be violated in the real world.*
>
> ('t Hooft, 2002, p. 309)

The seeming conflict of quantum mechanics with local realism appears even more dramatic if state vectors of the form

$$\frac{1}{\sqrt{2}}(\mathbf{H}\otimes\mathbf{H}\otimes\mathbf{H}+\mathbf{V}\otimes\mathbf{V}\otimes\mathbf{V})$$

(***Greenberger-Horne-Zeilinger*** states) are considered (Pan et al., 2000).

### 5.3.3 Quantum Teleportation

As discussed in 5.3.2, the internal quantum state $\widehat{=}\ \Psi_-$ of the EPR pairs exhibits correlations which are incompatible with local realism.

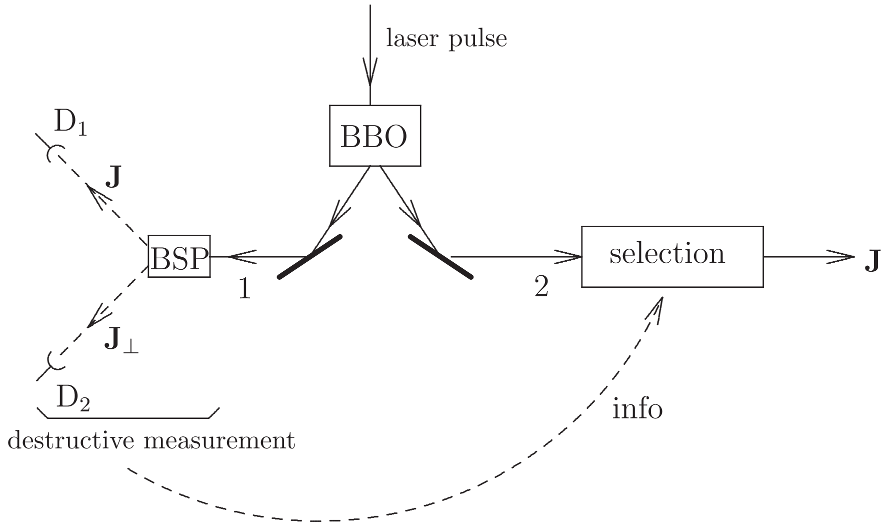
Even without deeper insight into the origin of these correlations they may be exploited for new technologies:

Their use for preparation of single-photon states with prescribed polarization **J**



[24] See (Aspect, 2004),(Kaltenbaek et al., 2003), (Simon and Irvine, 2003), and (Rosenberg et al., 2005) for experimental checks. For classical light, of course, the corresponding Bell inequality is fulfilled.

was already mentioned:[25]




An especially valuable application is ***quantum teleportation***, basing on the identity

$$2\,\mathbf{J}\otimes\Psi_- = \Phi_+\otimes\hat{U}_1\,\mathbf{J} + \Psi_+\otimes\hat{U}_2\,\mathbf{J} + \Phi_-\otimes\hat{U}_3\,\mathbf{J} + \Psi_-\otimes\hat{U}_4\,\mathbf{J} \tag{5.15}$$

where:

$$\begin{aligned}
\Psi_\pm &\stackrel{\text{def}}{=} \frac{1}{\sqrt{2}}\left(\begin{pmatrix}1\\0\end{pmatrix}\otimes\begin{pmatrix}0\\1\end{pmatrix}\pm\begin{pmatrix}0\\1\end{pmatrix}\otimes\begin{pmatrix}1\\0\end{pmatrix}\right),\\
\Phi_\pm &\stackrel{\text{def}}{=} \frac{1}{\sqrt{2}}\left(\begin{pmatrix}1\\0\end{pmatrix}\otimes\begin{pmatrix}1\\0\end{pmatrix}\pm\begin{pmatrix}0\\1\end{pmatrix}\otimes\begin{pmatrix}0\\1\end{pmatrix}\right),\\
\hat{U}_j\begin{pmatrix}\alpha\\\beta\end{pmatrix} &\stackrel{\text{def}}{=} \begin{cases}\begin{pmatrix}-\beta\\\alpha\end{pmatrix} & \text{for } j=1\,,\\ \begin{pmatrix}-\alpha\\\beta\end{pmatrix} & \text{for } j=2\,,\\ \begin{pmatrix}\beta\\\alpha\end{pmatrix} & \text{for } j=3\,,\\ -\begin{pmatrix}\alpha\\\beta\end{pmatrix} & \text{for } j=4\,.\end{cases}
\end{aligned}$$

**Outline of proof:** With the identification

$$(\Phi_1,\Phi_2,\Phi_3,\Phi_4) = (\Phi_+,\Psi_+,\Phi_-,\Psi_-)$$

we have

$$\Phi_j = \mathbf{J}\otimes f_j(\mathbf{J}) + \mathbf{J}_\perp\otimes f_j(\mathbf{J}_\perp) \quad \forall\, J\in\mathbb{C}^2\,,\ j\in\{1,\dots,4\}\,,$$

for the anti-linear mappings

$$f_j(\mathbf{J}) \stackrel{\text{def}}{=} \sum_{\nu=1}^{2}\langle\mathbf{J}\otimes\mathbf{e}_\nu \mid \Phi_j\rangle\,\mathbf{e}_\nu \quad \forall\, J\in\mathbb{C}^2\,,$$



[25]See (Migdall et al., 2002) for possibility to increase the efficiency of such sources.

not depending on the choice of orthonormal basis $\{\mathbf{e}_1, \mathbf{e}_2\}$ of $\mathbb{C}^2$. This implies

$$\begin{aligned}
&\left(\hat{P}_{\Phi_j} \otimes 1\right)(\mathbf{J} \otimes \Phi_4) \\
&= \left(\hat{P}_{\Phi_j} \otimes 1\right)\left(\mathbf{J} \otimes f_j(\mathbf{J}) \otimes f_4\Big(f_j(\mathbf{J})\Big) + \mathbf{J} \otimes f_j(\mathbf{J})_\perp \otimes f_4\Big(f_j(\mathbf{J})_\perp\Big)\right) \\
&= \left(\hat{P}_{\Phi_j} \otimes 1\right)\Big(\mathbf{J} \otimes f_j(\mathbf{J}) \otimes (f_4 \circ f_j)(\mathbf{J})\Big) \\
&= \langle \Phi_j \mid \mathbf{J} \otimes f_j(\mathbf{J})\rangle\, \Phi_j \otimes (f_4 \circ f_j)(\mathbf{J}) \quad \forall\, J \in \mathbb{C}^2,\ j \in \{1, \ldots, 4\}.
\end{aligned}$$

Since $\{\Phi_1, \ldots, \Phi_4\}$ is an orthonormal basis of $\mathbb{C}^2 \otimes \mathbb{C}^2$ this together with

$$2\,(f_4 \circ f_j)(\mathbf{J}) = \hat{U}_j\,\mathbf{J} \quad \forall\, j \in \{1, \ldots, 4\}$$

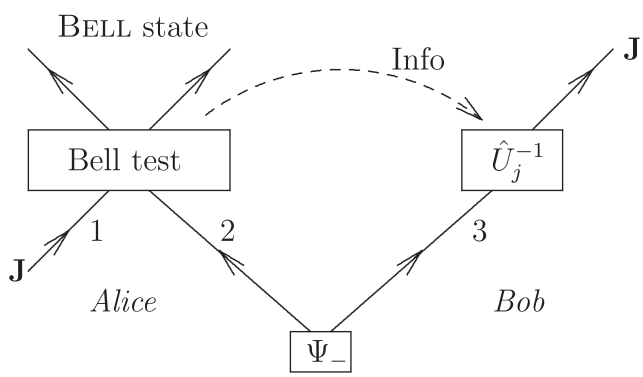
proves the statement. ∎

**Remark:** Quantum teleportation correspond to the following **virtual** flow of information:

> Since the initial (internal) state of photon 1 was (the unknown) $\mathbf{J}$, photon 2 must have 'been' in the initial state $f_j(\mathbf{J})$ if the result of the BELL measurement on photons 1 and 2 was $\Phi_j$. Then, because of the correlation of photons 2 and 3 the resulting state of photon 3 must be $2\,f_4 \circ f_j(\mathbf{J})$. Therefore application of $\hat{U}_j^{-1}$ turns the state of photon 3 into $\mathbf{J}$.

This kind of reasoning has been elaborated into a remarkable theory.[26]

Since the four BELL ***states*** $\Psi_\pm, \Phi_\pm$ form an orthonormal basis of $\mathbb{C}^2 \otimes \mathbb{C}^2$ we just have to test in which of these 2-photon states states photons 1 and 2 'are' (BELL test) in order to know the transformation $U_j$ turning the state of photon 3 into the unknown $\mathbf{J}$:




Once *Alice* and *Bob* share an EPR pair (photons 2 and 3) they do not need a quantum channel any more in order to teleport the unknown initial state $\mathbf{J}$ of photon 1 onto photon 3. However, the necessary classical communication (from *Alice* to *Bob*) of the result of the BELL test can only be performed with subluminal speed. Note also that the BELL test destroys the original state of photon 1.



[26]See (Coecke, 2004, Theorem 3.3), (Abramsky and Coecke, 2004), and references given there.

**Conclusion:**

> If *Alice* and *Bob* have the possibility to order, store, and release partners of shared EPR pairs then they can exchange unknown quantum information (in principle) reliably without any chance for potential eavesdroppers.

One possibility to implement a complete BELL test results form the following action of (nonlinear) BBO crystals[27]

$$\Phi_\pm \underset{\text{Typ-I}}{\longmapsto} \frac{1}{\sqrt{2}}\left(\hat{H}' \pm \mathbf{V}'\right) ,$$
$$\Psi_\pm \underset{\text{Typ-II}}{\longmapsto} \frac{1}{\sqrt{2}}\left(\hat{H}' \pm \mathbf{V}'\right) .$$

According to (Kim et al., 2001) this may be exploited as sketched in Figure 5.5. If detector the $D_j$ fires then*Alice* instructs *Bob* to apply $\hat{U}_j$.

A severe problem of long distance quantum teleportation is the exponential increase of photoabsorption (especially in optical fibers). In principle this problem can be mastered exploiting ***teleportation of entanglement***,[28] sketched in figure 5.6.

Teleporting the correlations of photons 0 and 1, described by $\Psi_-$, onto the pair of photons 0 and 3 relies on the identity

$$2\sqrt{2}\,\Psi_- \otimes \Psi_- = \sum_{j=1}^{4} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \otimes \Phi_+ \otimes \hat{U}_j \begin{pmatrix} 0 \\ 1 \end{pmatrix} - \begin{pmatrix} 0 \\ 1 \end{pmatrix} \otimes \Phi_+ \otimes \hat{U}_j \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

following from (5.15), which we may also write as

$$|\Psi_-\rangle_{0,1} \otimes |\Psi_-\rangle_{2,3} = \sum_{j=1}^{4} |\Phi_j\rangle_{1,2} \otimes \left|\left(\hat{1} \otimes \hat{U}_j\right) \Psi_-\right\rangle_{0,3} .$$

However, useful application of such methods requires the possibility to check success of the necessary operations. This could be provided by implementations of quantum nondemolition (QND) measurements of the photon number which are of great interest by themselves.

One suggestion to implement such measurements for small photon numbers $n$ is

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[27] We are writing $\mathbf{H}', \mathbf{V}'$ instead of $\mathbf{H}, \mathbf{V}$ in order to indicate doubling of frequency via conversion of the pairs into single photons.

[28] See (Duan et al., 2001). Another theoretical but barely practicable possibility to fight absorption is described in (Gingrich et al., 2003).

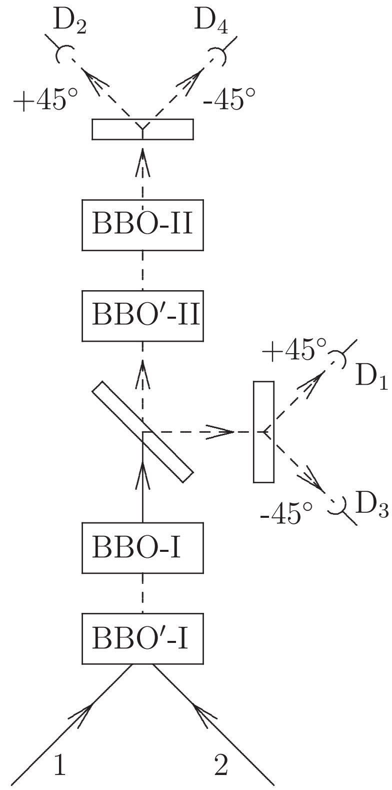



Figure 5.5: Possibility of a complete Bell test.

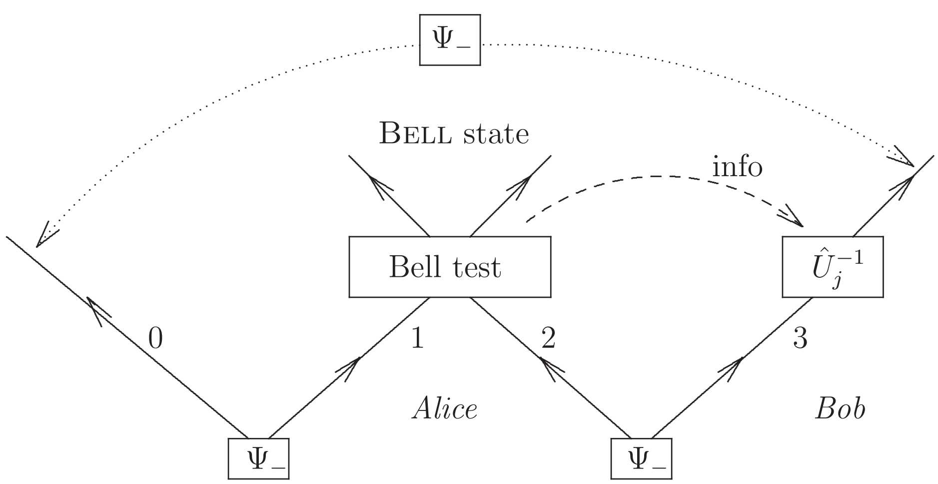
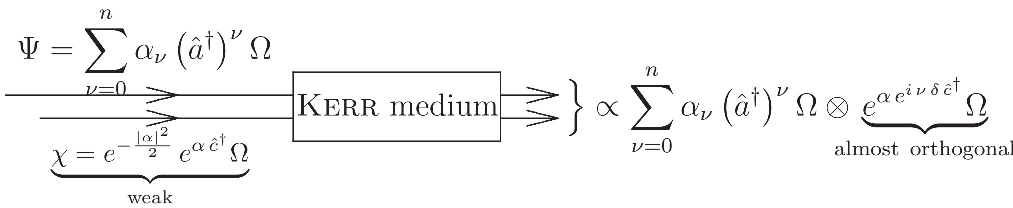


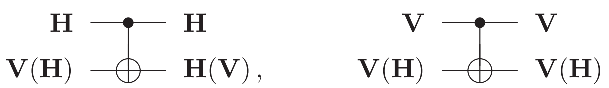

Figure 5.6: Teleportation of Entanglement

described in (Munro et al., 2005):




Such implementations require extremely strong cross-KERR nonlinearities which can be provided[29] by electromagnetically induced transparency (EIT), treated in 8.3.2.

Such nonlinearities can also be used to implement CNOT gates, i.e. 2-qubit gates acting according to



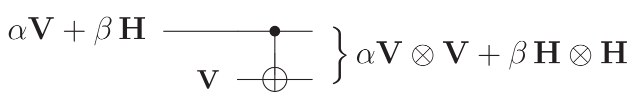

One just has to construct a ***nonlinear phase gate*** (NS) acting according

$$\left(\alpha\,\hat 1 + \beta\,\hat a^\dagger_{\mathrm H} + \gamma\left(\hat a^\dagger_{\mathrm H}\right)^2\right)\Omega \quad \underset{\mathrm{NS}}{\longmapsto} \quad \left(\alpha\,\hat 1 + \beta\,\hat a^\dagger_{\mathrm H} - \gamma\left(\hat a^\dagger_{\mathrm H}\right)^2\right)\Omega\,.$$

Then the arrangement sketched in Figure 5.7 represents a CNOT gate. Here 2-photon interference is exploited at the HADAMARD beam splitters.

**Remark:** For the detailed action of the (non-symmetric) HADAMARD beam splitters see Figure 3.2 of (Lücke, qip).

The CNOT gate is also called ***measurement gate*** since it may be used for QND measurement of the polarization of the target photon:[30]




Destructive 'measurement' of the (lower) ancillary photon (with initial state $\mathbf V$) yields an ideal test of the $\mathbf V$-$\mathbf H$ alternative[31] for the (upper) target photon.



[29] See (Ottaviani et al., 2006).
[30] Note that is acts like a copying machine if only the states $\mathbf V$ and $\mathbf H$ are allowed for the target photon.
[31] Tests for general $\mathbf J$-$\mathbf J_\perp$ alternatives may be easily implemented by additional unitary single-photon transformations.

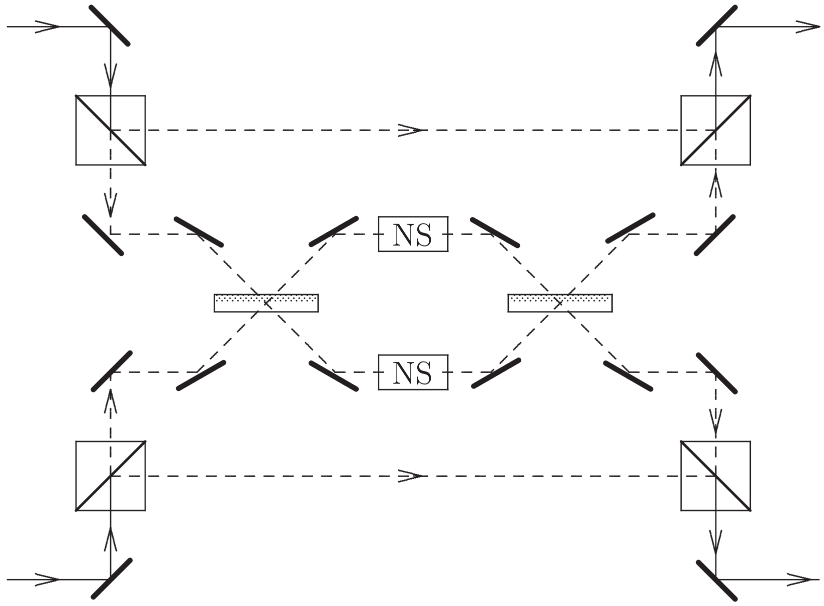
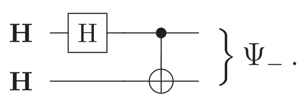



Figure 5.7: Implementation of a CNOT gate.

Note that CNOT gates may be used to produce EPR pairs:




Already for this reason also indeterministic CNOT gates are of considerable interest — if appropriate quantum memory is available.[32] here ***indeterministic*** means:

> The gate acts as desired only with a certain probability. However, proper action is highlighted by some ancillary system (similarly to QND measurements).

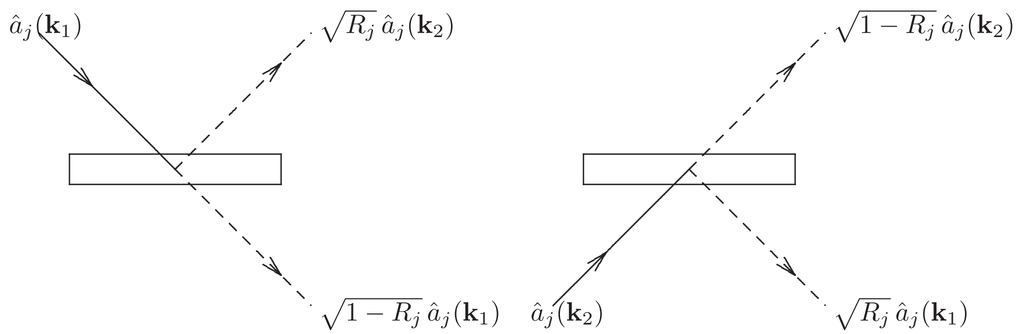
One possibility for implementing indeterministic gates exploits the following effect:[33]



[32] See (Pittman and Franson, 2002) and (Liu et al., 2001; Juzeliunas et al., 2003; Fleischhauer and Lukin, 2002; van der Wal et al., 2003) for possibilities of storing quantum information.

[33] See (Sanaka et al., 2003).

A beam splitter acting according to




also acts according to

$$
\begin{aligned}
&\left(\hat{a}_1^\dagger(\mathbf{k}_1)\right)^m \left(\hat{a}_2^\dagger(\mathbf{k}_1)\right)^n \hat{a}_2^\dagger(\mathbf{k}_2)\,\Omega \\
&\longmapsto \sum_{\mu=0}^{m}\sum_{\nu=0}^{n+1} c_{\mu\nu} \left(\hat{a}_1^\dagger(\mathbf{k}_2)\right)^\mu \left(\hat{a}_2^\dagger(\mathbf{k}_2)\right)^\nu \left(\hat{a}_1^\dagger(\mathbf{k}_1)\right)^{m-\mu} \left(\hat{a}_2^\dagger(\mathbf{k}_1)\right)^{n+1-\mu} \Omega\,,
\end{aligned}
\tag{5.16}
$$

where we especially have

$$
c_{mn} = \left(\sqrt[+]{R_1}\right)^m \left(\sqrt[+]{R_2}\right)^{n-1} \left(R_2 - n\,(1-R_2)\right) \tag{5.17}
$$

corresponding to the two possibilities, sketched in Figure 5.8, for exactly one photon leaving the lower right arm of the beam splitter and having polarization $\widehat{=}\ \mathbf{H}$. For $m = 0$, $n = 1$ and $R_2 = 1/2$ equation (5.17) reflects the familiar 2-photon interference:

$$
c_{01} = 0 \quad \text{for } R_2 = \frac{1}{2}\,.
$$

As indicated in Figure 5.9 the upper arms of the beam splitter can be considered as input and output ports of an indeterministic $NS'$ gate in the sense that

$$
\text{success} \ \widehat{=} \left\{ \begin{array}{l} \hat{a}_2(\mathbf{k}_1) \ \widehat{=} \text{ state of the} \\ \text{lower output of the beam splitter}\,, \end{array} \right.
$$

if an ancillary photon in the state $\hat{a}_2(\mathbf{k}_2)$ is used as lower input for the beam splitter.

For suitable choice of $R_1, R_2$ two such NS$'$ gates can be combined with two Hadamard beam splitters and two Hadamard gates $\boxed{\mathrm{H}}$ to yield an indeterministic CNOT gate as sketched in Figure 5.10.

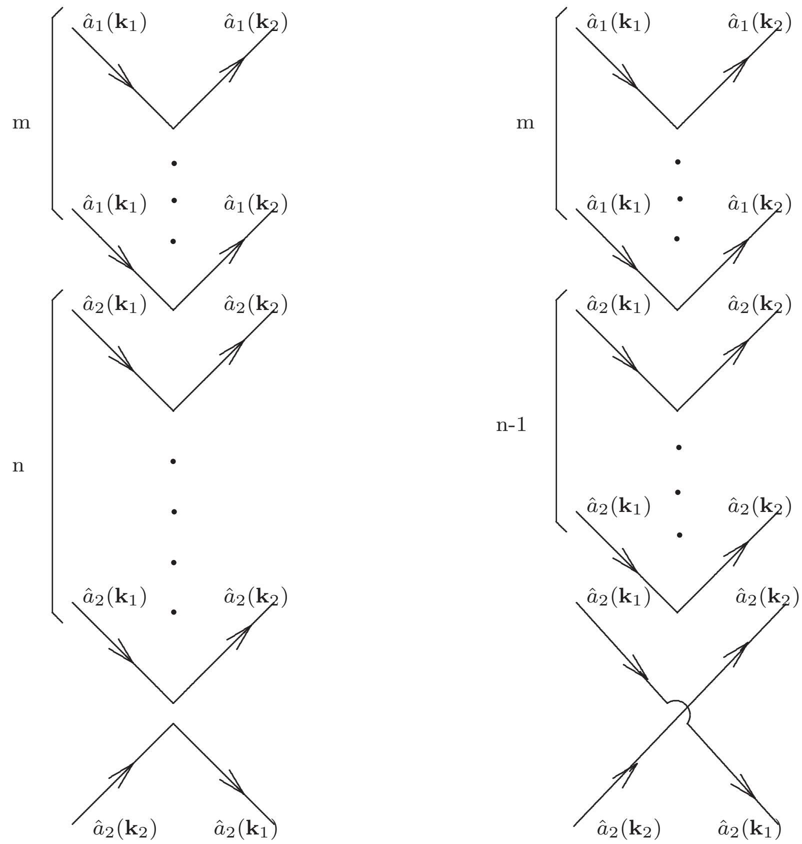



Figure 5.8: Selected Transitions at the Beam Splitter.

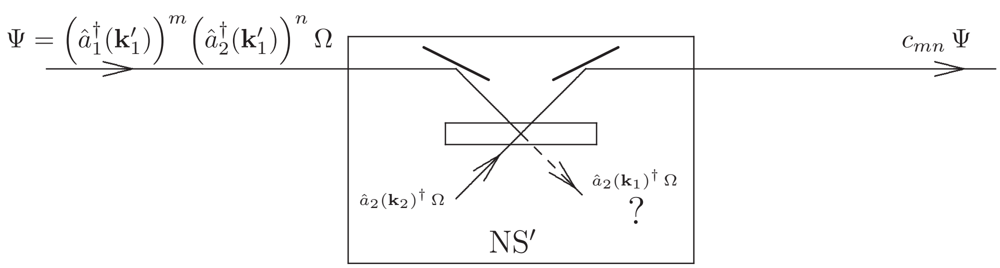



Figure 5.9: Indeterministic NS Gate.

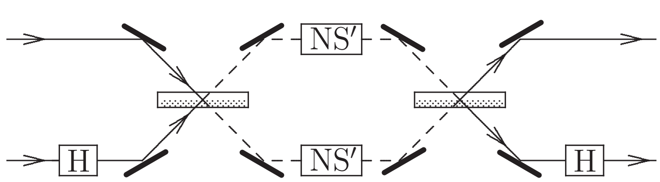



Figure 5.10: Indeterministic CNOT Gate.

The HADAMARD beam splitters act according to

$$\begin{aligned}\Psi_- &\stackrel{\mathrm{def}}{=} \frac{1}{\sqrt{2}}\Big(\hat a_1^\dagger(\mathbf{k}_1)\,\hat a_2^\dagger(\mathbf{k}_2) - \hat a_2^\dagger(\mathbf{k}_1)\,\hat a_1^\dagger(\mathbf{k}_2)\Big)\,\Omega\\ &\longmapsto \tfrac{1}{\sqrt{2}}\Big(\Big(\hat a_1^\dagger(\mathbf{k}_1)+\hat a_1^\dagger(\mathbf{k}_2)\Big)\Big(\hat a_2^\dagger(\mathbf{k}_2)-\hat a_2^\dagger(\mathbf{k}_1)\Big)\\ &\qquad\qquad - \Big(\hat a_2^\dagger(\mathbf{k}_1)+\hat a_2^\dagger(\mathbf{k}_2)\Big)\Big(\hat a_1^\dagger(\mathbf{k}_2)-\hat a_1^\dagger(\mathbf{k}_1)\Big)\Big)\,\Omega\\ &= \Psi_-\,,\end{aligned}$$

$$\begin{aligned}\Psi_+ &\stackrel{\mathrm{def}}{=} \frac{1}{\sqrt{2}}\Big(\hat a_1^\dagger(\mathbf{k}_1)\,\hat a_2^\dagger(\mathbf{k}_2) + \hat a_2^\dagger(\mathbf{k}_1)\,\hat a_1^\dagger(\mathbf{k}_2)\Big)\,\Omega\\ &\longmapsto \Psi_0 \stackrel{\mathrm{def}}{=} \frac{1}{\sqrt{2}}\Big(\hat a_1^\dagger(\mathbf{k}_2)\,\hat a_2^\dagger(\mathbf{k}_2) - \hat a_1^\dagger(\mathbf{k}_1)\,\hat a_2^\dagger(\mathbf{k}_1)\Big)\,\Omega\,,\end{aligned}$$

$$\begin{aligned}&\hat a_j^\dagger(\mathbf{k}_1)\,\hat a_j^\dagger(\mathbf{k}_2)\,\Omega\\ &\longmapsto \frac{1}{2}\Big(\Big(\hat a_j^\dagger(\mathbf{k}_1)+\hat a_j^\dagger(\mathbf{k}_2)\Big)\Big(\hat a_j^\dagger(\mathbf{k}_2)-\hat a_j^\dagger(\mathbf{k}_1)\Big)\,\Omega\\ &= \Psi_j \stackrel{\mathrm{def}}{=} \frac{1}{2}\Big(\Big(\hat a_j^\dagger(\mathbf{k}_2)\Big)^2 - \Big(\hat a_j^\dagger(\mathbf{k}_1)\Big)^2\Big)\,\Omega\,.\end{aligned}$$

According to Figure 5.9 and equation (5.17) joint succesfull operation of both NS′ gates has the following effect on the relevant intermediate states:[34]

$$\begin{aligned}\Psi_- &\longmapsto \underbrace{c_{10}\,c_{01}}_{=\sqrt[+]{R_1R_2}(2\,R_2-1)}\,\Psi_-\,,\\ \Psi_0 &\longmapsto \underbrace{c_{11}\,c_{00}}_{=c_{10}\,c_{01}}\,\Psi_0\,,\\ \Psi_1 &\longmapsto \underbrace{R_1R_2}_{=c_{20}\,c_{00}}\,\Psi_1\,,\\ \Psi_2 &\longmapsto \underbrace{R_2\,(3\,R_2-2)}_{=c_{02}\,c_{00}}\,\Psi_2\,.\end{aligned}$$

Since the two HADAMARD gates (without the NS′ gates but combined with the necessary mirrors) form a balanced MACH-ZEHNDER interferometer (see Figure 3.4 of (Lücke, qip)). The whole arrangement outlined in Figure 5.10, but without the two HADAMARD gates, acts in case of success according to

$$\Psi \longmapsto \begin{cases} -R_1R_2\,\Psi & \text{for } \Psi \propto \mathbf{H}\otimes\mathbf{H}\,,\\ +R_1R_2\,\Psi & \text{for } \Psi \perp \mathbf{H}\otimes\mathbf{H}\,,\end{cases}$$

**if**

$$-\,R_2\,(3\,R_2-2) = R_1R_2 = \sqrt[+]{R_1R_2}\,(2\,R_2-1)\;. \tag{5.18}$$

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[34]Equation (5.17) implies

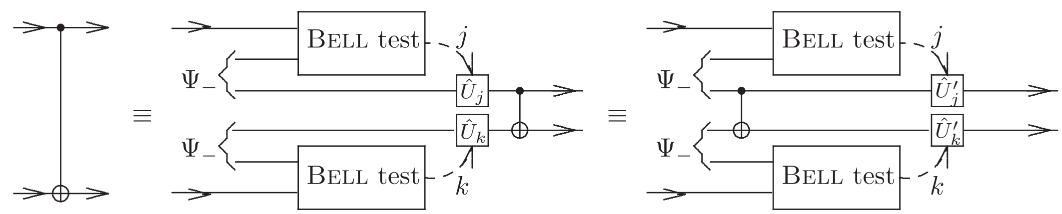
$$\begin{aligned}c_{10} &= \sqrt[+]{R_1R_2}\,, & c_{01} &= (2\,R_2-1)\,, & c_{11} &= \sqrt[+]{R_1}\,(2\,R_2-1)\,,\\ c_{00} &= \sqrt[+]{R_2}\,, & c_{20} &= R_1\,\sqrt[+]{R_2}\,, & c_{02} &= \sqrt[+]{R_2}\,(3\,R_2-2)\,.\end{aligned}$$

Addition of the two HADAMARD gates, finally, converts it into an indeterministic CNOT gate. The conditions (5.18) are fulfilled for

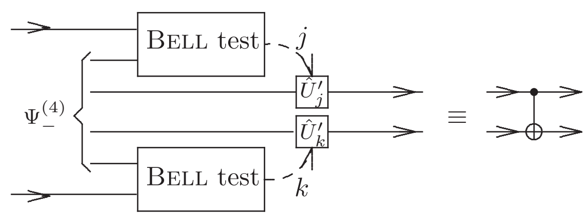
$$R_2 = \frac{3+\sqrt{2}}{7} \approx 0.63\,, \quad R_1 = \frac{5-3\sqrt{2}}{7} \approx 0.1\,.$$

The corresponding probability for successful operation[35] is $|R_1 R_2|^2 \approx 0.0047$.

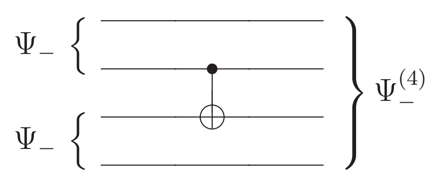
A crucial discovery by (Gottesman and Chuang, 1999) is the following (see also Sect. 3.1.3 of (Lücke, qip)):




This suggests the following implementation of **deterministic** CNOT gate:




Here an ancillary 4-photon state $\Psi_-^{(4)}$ is used that may be prepared by means of an indeterministic CNOT gate according to[36]




This, in principle, allows scalable quantum computing using only linear optical devices, photon number resolving detectors, and quantum memory.



[35] For a more efficient, slightly more complicated, implementation see (Sanaka and Resch, 2003).

[36] See (Knill et al., 2000; Knill et al., 2001) and (Ralph et al., 2002; Dowling et al., 2004).

# Chapter 6

# Coupling of Quantum Systems

*Now it is customary to define the method of measuring physical quantities without defining these quantities themselves. In fact we have no satisfactory reason for ascribing objective existence to physical quantities as distinguished from the numbers obtained when we make the measurements which we correlate with them ... It would evidently be philosophically more exact if we spoke of "making measurements" of this, that, or the other type instead of saying that we measure this, that, or the other "physical quantity" ... however, we can continue to use the old language, reinterpreting the terms "physical quantity" and "dynamical variable," and allowing them to stand for the corresponding operator which fixes the nature of the measurement under consideration.*

(Kemble, 1937, Section 36b)

## 6.1 Closed Systems

### 6.1.1 States and Observables in the Heisenberg Picture

In the Heisenberg ***picture*** the physical state of a closed[1] quantum system is characterized by a density operator $\hat{\rho}^{\mathrm{H}}$ on its ***state space*** $\mathcal{H}$ — being a Hilbert space — and a time-dependent injection

$$Q \longmapsto \hat{P}^{\mathrm{H}}_{Q}(t)$$

of the (naively assumed) testable 'properties' $Q$ into the (orthogonal) projectors $\hat{P}^{\mathrm{H}}_{Q}(t)$ on $\mathcal{H}$ such that:[2]

$$\boxed{p_{Q,t}\big(\hat{\rho}^{\mathrm{H}}\big) \stackrel{\mathrm{def}}{=} \mathrm{Tr}\Big(\hat{\rho}^{\mathrm{H}}\, \hat{P}^{\mathrm{H}}_{Q}(t)\Big) = \begin{cases}\text{probability for: "}Q\text{ at time }t\text{" to be}\\ \text{confirmed by optimal tests}\,.\end{cases}} \tag{6.1}$$

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[1]For a discussion of the applicability of the concept of a "closed system" (isolated system) see (Leggett, 2001, Lecture 3).

[2]If every step of a test for '$Q$ at time $t$' is delayed by $\Delta t$ then this amount to a test for '$Q$ at time $t + \Delta t$'. We do not yet require that $Q$ refers to an instant of time.

**Remarks:**

1. A ***density operator*** (***statistical operator***) is a positive trace class operator $\hat{\rho}$ with
$$\mathrm{Tr}(\hat{\rho}) = 1$$
(see, e.g., Definition 8.3.19 of (Lücke, eine)).

2. The simplest density operators are those of the form
$$\hat{\rho} = |\Psi\rangle\langle\Psi| \ , \Psi \in \mathcal{H} \, ,$$
Those states are called ***vector states*** and fulfill [3]
$$\mathrm{Tr}\left(\hat{\rho}\,\hat{P}\right) = \left\langle \Psi \mid \hat{P}\,\Psi \right\rangle = \left\|\hat{P}\,\Psi\right\|^2 \tag{6.2}$$
for all projectors $\hat{P}$ on $\mathcal{H}_{\text{field}}$ .

3. Every density operator $\hat{\rho}$ on $\mathcal{H}_{\text{field}}$ may be written in the form
$$\hat{\rho} = \sum_{\nu=1}^{\infty} \underbrace{\lambda_\nu}_{\geq 0} |\phi_\nu\rangle\langle\phi_\nu| \ , \quad \sum_{\nu=1}^{\infty} \lambda_\nu = 1 = \|\Phi_\mu\| \quad \forall\, \mu \in \mathbb{N} \, ,$$
suggesting the following **naive** interpretation:[4]

   The actual state is one of the vector states $\phi_1, \phi_2, \ldots$ and $\lambda_\nu$ is the probability for this being $\phi_\nu$ .

4. Note that (6.1) is assumed only over a time interval during which the system can be considered as closed. Thus, especially, the preparation of the state has to be done before this time interval; and nontrivial tests ('measurements') — since inevitably changing the state — are allowed only after this time interval.

5. More generally ***effects***, i.e. bounded Operators $\hat{A}$ on $\mathcal{H}$ with $0 \leq \hat{A} \leq \hat{1}$ , could be used[5] instead of the projectors $\hat{P}^{\mathrm{H}}_Q(t)$ .

The (naive) idea behind (6.1) is the following:

- $\hat{P}^{\mathrm{H}}_Q(t)$ projects $\mathcal{H}$ onto the closed subspace corresponding to those vector states for which "$Q$ at time $t$" will be confirmed by **every** optimal test.



[3] Of course, $\mathrm{Tr}(\hat{\rho}) = 1$ implies $\|\Psi\| = 1$ .

[4] In general, however, there is no unique choice for the states $\phi_\nu$ . See (Timpson and Brown, 2005) in this connection.

[5] See (Davies, 1976, Section 2.2), (Kraus, 1983, § 1), or (Busch et al., 1995) for the corresponding physical interpretation.

- $\neg\hat{P}^{\mathrm{H}}_{Q}(t) \stackrel{\text{def}}{=} \hat{1} - \hat{P}^{\mathrm{H}}_{Q}(t)$ projects $\mathcal{H}$ onto the closed subspace corresponding to those vector states for which "$Q$ at time $t$" will **never** be confirmed by any optimal test.

- Since $\left\|\hat{P}^{\mathrm{H}}_{Q}(t)\,\Psi\right\|^2 + \left\|\neg\hat{P}^{\mathrm{H}}_{Q}(t)\,\Psi\right\|^2 = 1$ it seems natural to interpret
$$\left\|\hat{P}^{\mathrm{H}}_{Q}(t)\,\Psi\right\|^2 = \mathrm{Tr}\Big(|\Psi\rangle\langle\Psi|\,\hat{P}^{\mathrm{H}}_{Q}(t)\Big)$$
as the probability for "$Q$ at time $t$" in a state described by $\hat{\rho}^{\mathrm{H}} = |\Psi\rangle\langle\Psi|$.

- This motivates (6.1) for vector states.

- Due to linearity of the trace this implies (6.1) for states represented by density operators of the form
$$\hat{\rho}^{\mathrm{H}} = \sum_{\nu=0}^{\infty} \underbrace{p_\nu}_{\geq 0} |\Psi\rangle_\nu\langle\Psi|_\nu \;, \quad \sum_{\nu=0}^{\infty} p_\nu = 1\,, \tag{6.3}$$
if,[6] for every $\nu$, $p_\nu$ is (naively) identified with the probability for the system to be in the vector state corresponding to $\Psi_\nu$.

- Since every density operator can be written in the form (6.3), thanks to the Hilbert-Schmidt theorem (see, e.g., Corollary 8.3.18 of (Lücke, eine)), this implies (6.1) for all states.

There is a one-one correspondence between self-adjoint operators $\hat{A}$ (see, e.g., Definition 8.3.7 (Lücke, eine)) and families $\left\{\hat{E}^{\hat{A}}_{\lambda}\right\}_{\lambda\in\mathbb{R}}$ of projectors fulfilling the following requirements (see Theorem 8.3.24 of (Lücke, eine)):

1. $$\left(\lambda_1 \leq \lambda_2 \implies \left\langle \Psi \mid \hat{E}^{\hat{A}}_{\lambda_1}\Psi\right\rangle \leq \left\langle \Psi \mid \hat{E}_{\lambda_2}\Psi\right\rangle\right) \quad \forall\,\Psi\in\mathcal{H}\,,\ \lambda_1,\lambda_2\in\mathbb{R}\,.$$

2. $$\lim_{\epsilon\to-\infty}\left\langle \Psi \mid \hat{E}^{\hat{A}}_{\lambda}\Psi\right\rangle = 0 \quad \forall\,\Psi\in\mathcal{H}\,,\ \lambda\in\mathbb{R}\,.$$

3. $$\lim_{\epsilon\to+\infty}\left\langle \Psi \mid \hat{E}^{\hat{A}}_{\lambda}\Psi\right\rangle = \langle\Psi\mid\Psi\rangle \quad \forall\,\Psi\in\mathcal{H}\,,\ \lambda\in\mathbb{R}\,.$$

4. $$\lim_{\epsilon\to+0}\left\langle \Psi \mid \hat{E}^{\hat{A}}_{\lambda+\epsilon}\Psi\right\rangle = \left\langle \Psi \mid \hat{E}^{\hat{A}}_{\lambda}\Psi\right\rangle \quad \forall\,\Psi\in\mathcal{H}\,,\ \lambda\in\mathbb{R}\,.$$



[6] See also Section 6.2.2 and (Zeh, 2000) for mixed states arising for subsystems from vector states of a composed system.

5.
$$D_{\hat{A}} = \left\{ \Psi \in \mathcal{H} : \int |\lambda|^2 \, \mathrm{d}\left\langle \Psi \mid \hat{E}^{\hat{A}}_\lambda \, \Psi \right\rangle < \infty \right\} .$$

6.
$$\left\langle \Psi \mid \hat{A} \, \Psi \right\rangle = \int \lambda \, \mathrm{d}\left\langle \Psi \mid \hat{E}^{\hat{A}}_\lambda \, \Psi \right\rangle \quad \forall \, \Psi \in D_{\hat{A}} .$$

Therefore self-adjoint operators are interpreted as ***observables*** of physical quantities — like 'energy at time $t$' — in the following sense:

$$\mathrm{Tr}\left(\hat{\rho}^{\mathrm{H}} \, \hat{E}^{\hat{A}}_\lambda\right) = \begin{cases} \text{probability for: “}A \in (-\infty, \lambda]\text{” to be} \\ \text{confirmed by optimal tests} \, . \end{cases}$$

Consequently:

$$\boxed{\mathrm{Tr}\left(\hat{\rho}^{\mathrm{H}} \, \hat{A}\right) = \text{expectation value of } A \text{ in the state} \mathrel{\widehat{=}} \hat{\rho}^{\mathrm{H}} \, .} \tag{6.4}$$

Projectors are just observables of physical quantities for which only 0 and 1 are 'possible values'.

### 6.1.2 Time Evolution and Projective Measurements

For a closed quantum system the time evolution is assumed to be given by a self-adjoint operator $\hat{H}$ called the ***Hamiltonian*** of the system, in the sense that

$$\boxed{\hat{P}^{\mathrm{H}}_Q(t) = e^{+\frac{i}{\hbar} \hat{H} \, t} \, \hat{P}^{\mathrm{H}}_Q(0) \, e^{-\frac{i}{\hbar} \hat{H} \, t} \quad \forall \, t \in \mathbb{R}} \tag{6.5}$$

holds for all 'testable properties' $Q$ .

**Remark:** See, e.g., (Lücke, 1996, Section2.5) for a justification of this assumption and Definition 8.3.30 of (Lücke, eine) for the definition of functions of self-adjoint operators.

This allows switching to the SCHRÖDINGER ***picture***

$$\mathrm{Tr}\left(\hat{\rho}^{\mathrm{S}}_t \, \hat{P}^{\mathrm{S}}_Q\right) = \begin{cases} \text{probability for “}Q \text{ at time } t\text{” to be} \\ \text{confirmed by optimal tests} \, , \end{cases} \tag{6.6}$$

where

$$\hat{\rho}^{\mathrm{S}}_t \stackrel{\mathrm{def}}{=} e^{-\frac{i}{\hbar} \hat{H} \, t} \, \hat{\rho}^{\mathrm{H}} \, e^{+\frac{i}{\hbar} \hat{H} \, t} \, , \tag{6.7}$$

$$\begin{aligned} \hat{P}^{\mathrm{S}}_Q &\stackrel{\mathrm{def}}{=} \hat{P}^{\mathrm{H}}(0) \\ &= e^{-\frac{i}{\hbar} \hat{H} \, t} \, \hat{P}^{\mathrm{H}}_Q(t) \, e^{+\frac{i}{\hbar} \hat{H} \, t} \, . \end{aligned} \tag{6.8}$$

Note that, formally, (6.7) is equivalent to the LIOUVILLE ***equation***

$$i\hbar \frac{\partial}{\partial t} \hat{\rho}_t^{\mathrm{S}} = \left[\hat{H} \,,\, \hat{\rho}_t^{\mathrm{S}}\right]_- \tag{6.9}$$

— to be suitably interpreted.

If the Hamiltonian is of the form

$$\boxed{\hat{H} = \hat{H}_0 + \hat{V}} \tag{6.10}$$

with $e^{-\frac{i}{\hbar}\hat{H}_0\, t}$ known and $\hat{V}$ 'small' in some sense then it may be convenient to switch to the ***interaction picture***

$$\mathrm{Tr}\Big(\hat{\rho}_t^{\mathrm{I}}\, \hat{P}_Q^{\mathrm{I}}(t)\Big) = \begin{cases}\text{probability for "$Q$ at time $t$" to be} \\ \text{confirmed by optimal tests}\,,\end{cases} \tag{6.11}$$

where

$$\hat{\rho}_t^{\mathrm{I}} \stackrel{\text{def}}{=} e^{+\frac{i}{\hbar}\hat{H}_0\, t}\, e^{-\frac{i}{\hbar}\hat{H}\, t}\, \hat{\rho}^{\mathrm{H}}\, e^{+\frac{i}{\hbar}\hat{H}\, t}\, e^{-\frac{i}{\hbar}\hat{H}_0\, t}\,, \tag{6.12}$$

$$\hat{P}_Q^{\mathrm{I}}(t) = e^{+\frac{i}{\hbar}\hat{H}_0\, t}\, \hat{P}_Q^{\mathrm{H}}(0)\, e^{-\frac{i}{\hbar}\hat{H}_0\, t}\,. \tag{6.13}$$

The reason will become evident in Section 6.1.3.

For simplicity we require that for every 'testable property' $Q$ there is a ***projective measurement***, i.e. a test changing the HEISENBERG state according to[7]

$$\hat{\rho}^{\mathrm{H}} \longmapsto p_{Q,t}\big(\hat{\rho}^{\mathrm{H}}\big)\, \hat{\rho}_{Q,t}^{\mathrm{H}} + \Big(1 - p_{Q,t}\big(\hat{\rho}^{\mathrm{H}}\big)\Big)\hat{\rho}_{\neg Q,t}^{\mathrm{H}} \tag{6.14}$$

with

$$\hat{\rho}_{Q,t}^{\mathrm{H}} \stackrel{\text{def}}{=} \frac{\hat{P}_Q^{\mathrm{H}}(t)\hat{\rho}^{\mathrm{H}}\hat{P}_Q^{\mathrm{H}}(t)}{\mathrm{Tr}\Big(\hat{P}_Q^{\mathrm{H}}(t)\hat{\rho}^{\mathrm{H}}\hat{P}_Q^{\mathrm{H}}(t)\Big)}$$

describing the subensemble of all those individual systems for which '$Q$ at time $t$' has been confirmed and

$$\hat{\rho}_{\neg Q,t}^{\mathrm{H}} \stackrel{\text{def}}{=} \frac{\hat{P}_{\neg Q}^{\mathrm{H}}(t)\hat{\rho}^{\mathrm{H}}\hat{P}_{\neg Q}^{\mathrm{H}}(t)}{\mathrm{Tr}\Big(\hat{P}_{\neg Q}^{\mathrm{H}}(t)\hat{\rho}^{\mathrm{H}}\hat{P}_{\neg Q}^{\mathrm{H}}(t)\Big)}\,, \qquad \hat{P}_{\neg Q}^{\mathrm{H}}(t) = \hat{1} - \hat{P}_Q^{\mathrm{H}}(t)\,,$$

describing the subensemble of all those individual systems for which '$Q$ at time $t$' is denied by the test. This assumption is consistent in the sense that

$$\mathrm{Tr}\big(\hat{\rho}^{\mathrm{H}}\big) = \mathrm{Tr}\Big(p_{Q,t}\big(\hat{\rho}^{\mathrm{H}}\big)\, \hat{\rho}_{Q,t}^{\mathrm{H}} + \Big(1 - p_{Q,t}\big(\hat{\rho}^{\mathrm{H}}\big)\Big)\hat{\rho}_{\neg Q,t}^{\mathrm{H}}\Big)\,.$$

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[7] Recall (6.1).

Note that

$$\begin{aligned} &p_{Q',t}\left(\hat{\rho}^{\mathrm{H}}\right) = p_{Q',t}\left(p_{Q,t}\left(\hat{\rho}^{\mathrm{H}}\right)\hat{\rho}^{\mathrm{H}}_{Q,t} + \left(1 - p_{Q,t}\left(\hat{\rho}^{\mathrm{H}}\right)\right)\hat{\rho}^{\mathrm{H}}_{\neg Q,t}\right) \quad \forall\, \hat{\rho}^{\mathrm{H}} \\ \iff\ & \left[\hat{P}^{\mathrm{H}}_{Q}(t)\,,\, \hat{P}^{\mathrm{H}}_{Q'}(t)\right]_{-} = 0\,, \end{aligned}$$

since

$$\operatorname{Tr}\left(\hat{\rho}\,\hat{A}\right) = \operatorname{Tr}\left(\hat{A}\,\hat{\rho}\right) \tag{6.15}$$

holds for all trace class operators $\hat{\rho}$ and all bounded operators $\hat{A}$. Therefore 'testable properties' $Q$, $Q'$ with commuting $\hat{P}^{\mathrm{H}}_{Q}(t)$, $\hat{P}^{\mathrm{H}}_{Q'}(t)$ are called ***compatible***.[8]

### 6.1.3 Time Dependent Perturbation Theory[9]

In the interaction picture the time-dependent density operator (6.12) fulfills the differential equation[10]

$$\boxed{i\hbar\frac{\mathrm{d}}{\mathrm{d}t}\hat{\rho}^{\mathrm{I}}_{t} = \left[\hat{V}^{\mathrm{I}}(t), \hat{\rho}^{\mathrm{I}}_{t}\right]_{-}\,,} \tag{6.16}$$

where

$$\hat{V}^{\mathrm{I}}(t) \stackrel{\mathrm{def}}{=} e^{+\frac{i}{\hbar}\hat{H}_0\,t}\,\hat{V}\,e^{-\frac{i}{\hbar}\hat{H}_0\,t}\,. \tag{6.17}$$

(6.16) is (formally) equivalent to the integral equation

$$\hat{\rho}^{\mathrm{I}}_{t} = \hat{\rho}^{\mathrm{I}}_{0} - \frac{i}{\hbar}\int_0^t \left[\hat{V}^{\mathrm{I}}(t'), \hat{\rho}^{\mathrm{I}}_{t'}\right]_{-}\,\mathrm{d}t'$$

which may be solved for given $\hat{\rho}^{\mathrm{I}}_{0}$ by iteration

$$\begin{aligned} \hat{\rho}^{(0)}_{t} &\stackrel{\mathrm{def}}{=} \hat{\rho}^{\mathrm{I}}_{0}\,, \\ \hat{\rho}^{(\nu)}_{t} &\stackrel{\mathrm{def}}{=} \hat{\rho}^{\mathrm{I}}_{0} - \frac{i}{\hbar}\int_0^t \left[\hat{V}^{\mathrm{I}}(t'), \hat{\rho}^{(\nu-1)}_{t'}\right]_{-}\,\mathrm{d}t' \quad \text{for } \nu = 1, 2, \ldots \end{aligned} \tag{6.18}$$

and taking the limit[11]

$$\hat{\rho}^{\mathrm{I}}_{t} = \lim_{\nu\to\infty}\hat{\rho}^{(\nu)}_{t}$$



[8]Note that

$$\left[\hat{P}^{\mathrm{H}}_{Q}(0)\,,\, \hat{P}^{\mathrm{H}}_{Q'}(0)\right]_{-} = 0 \underset{(6.5)}{\iff} \left(\left[\hat{P}^{\mathrm{H}}_{Q}(t)\,,\, \hat{P}^{\mathrm{H}}_{Q'}(t)\right]_{-} = 0\;\forall\, t \in \mathbb{R}\right)\,.$$

[9]In this subsection we do not care about mathematical details since even for a divergent perturbative expansion the lowest order may provide useful results.

[10]Compare Equation (6.9).

[11]For simplicity, we do not specify the type of convergence.

**if** $\hat{V}^{\mathrm{I}}(t)$ is 'sufficiently small'. For typical applications the approximation

$$\begin{aligned}\hat{\rho}^{\mathrm{I}}_t \approx \hat{\rho}^{(2)}_t \quad &= \quad \hat{\rho}^{\mathrm{I}}_0 - \frac{i}{\hbar}\int_0^t \left[\hat{V}^{\mathrm{I}}(t'), \hat{\rho}^{\mathrm{I}}_0\right]_- \mathrm{d}t' \\ &\quad -\hbar^{-2}\int_0^t \left(\int_0^{t'} \left[\hat{V}^{\mathrm{I}}(t'), \left[\hat{V}^{\mathrm{I}}(t''), \hat{\rho}^{\mathrm{I}}_0\right]_-\right]_- \mathrm{d}t''\right) \mathrm{d}t'\end{aligned}$$

is good enough. Note that, thanks to (6.15),

$$\begin{aligned}&\hat{P}\hat{\rho}^{\mathrm{I}}_0 = \hat{\rho}^{\mathrm{I}}_0\,\hat{P} = 0 \\ &\Longrightarrow\ \mathrm{Tr}\left(\hat{\rho}^{(2)}_t\,\hat{P}\right) = \hbar^{-2}\int_{[0,t]\times[0,t]} \mathrm{Tr}\left(\hat{V}^{\mathrm{I}}(t')\,\hat{\rho}^{\mathrm{I}}_0\,\hat{V}^{\mathrm{I}}(t'')\,\hat{P}\right) \mathrm{d}t''\,\mathrm{d}t'\,.\end{aligned} \tag{6.19}$$

This is in agreement with DYSON's perturbation theory for state vectors.[12]

# 6.2 Bipartite Systems

## 6.2.1 Composition of Distinguishable Systems

Let $\mathcal{S}_1$ and $\mathcal{S}_2$ be **distinguishable** quantum systems not interacting with each other. In order to describe $\mathcal{S}_1$ and $\mathcal{S}_2$ together as a single quantum system $\mathcal{S}$ in the HEISENBERG picture one takes the state space $\mathcal{H} = \mathcal{H}_1 \otimes \mathcal{H}_2$, where $\mathcal{H}_1$ resp. $\mathcal{H}_2$ is the state space of $\mathcal{S}_1$ resp. $\mathcal{S}_2$.

> **Remark:** See, e.g., Section 8.4.2 of (Lücke, eine) for the definition of the tensor products $\otimes$.

The Hamiltonian of $\mathcal{S}$ is

$$\hat{H} = \hat{H}_1 \otimes \hat{1} + \hat{1} \otimes \hat{H}_2\,, \tag{6.20}$$

where $\hat{H}_1$ resp. $\hat{H}_2$ is the Hamiltonian of $\mathcal{S}_1$ resp. $\mathcal{S}_2$, and therefore

$$e^{+\frac{i}{\hbar}\hat{H}\,t}\left(\hat{P}^{\mathrm{H}}_{Q_1}(0) \otimes \hat{P}^{\mathrm{H}}_{Q_2}(0)\right) e^{-\frac{i}{\hbar}\hat{H}\,t} = \hat{P}^{\mathrm{H}}_{Q_1}(t) \otimes \hat{P}^{\mathrm{H}}_{Q_2}(t) \quad \forall\, t \in \mathbb{R} \tag{6.21}$$

holds for all 'testable properties' $Q_1$ and $Q_2$ of $\mathcal{S}_1$ resp. $\mathcal{S}_2$.

If the HEISENBERG state of $\mathcal{S}$ is given by

$$\hat{\rho}^{\mathrm{H}} = \hat{\rho}^{\mathrm{H}}_1 \otimes \hat{\rho}^{\mathrm{H}}_2 \tag{6.22}$$

then

$$\mathrm{Tr}\left(\hat{\rho}^{\mathrm{H}}\left(\hat{P}^{\mathrm{H}}_{Q_1}(t) \otimes \hat{P}^{\mathrm{H}}_{Q_2}(t)\right)\right) = \mathrm{Tr}\left(\hat{\rho}^{\mathrm{H}}_1\,\hat{P}^{\mathrm{H}}_{Q_1}(t)\right)\mathrm{Tr}\left(\hat{\rho}^{\mathrm{H}}_2\,\hat{P}^{\mathrm{H}}_{Q_2}(t)\right).$$

This suggests the interpretation

$$\mathrm{Tr}\left(\hat{\rho}^{\mathrm{H}}\left(\hat{P}^{\mathrm{H}}_{Q_1}(t) \otimes \hat{P}^{\mathrm{H}}_{Q_2}(t)\right)\right) = \begin{cases}\text{probability for: "}Q_1\ \textbf{and}\ Q_2 \text{ at time } t\text{"} \\ \text{to be confirmed by optimal tests}\,.\end{cases} \tag{6.23}$$



[12] See Appendix A.4.

As long as (6.20) holds (6.23) is equivalent to

$$\boxed{\operatorname{Tr}\left(\hat{\rho}_t^{\mathrm{S}}\left(\hat{P}_{Q_1}^{\mathrm{S}} \otimes \hat{P}_{Q_2}^{\mathrm{S}}\right)\right) = \begin{cases}\text{probability for: "}Q_1 \textbf{ and } Q_2 \text{ at time } t\text{"}\\ \text{to be confirmed by optimal tests}\,,\end{cases}} \tag{6.24}$$

thanks to (6.21). However, if an interaction between $\mathcal{S}_1$ and $\mathcal{S}_2$ is introduced, corresponding to the addition of a ***perturbation***

$$\hat{V} \neq \hat{V}_1 \otimes \hat{1} + \hat{1} \otimes \hat{V}_2 \quad \forall\, \hat{V}_1\,,\, \hat{V}_2 \tag{6.25}$$

on the r.h.s. of (6.20), then (6.21) is no longer consistent for all $Q_1, Q_2$. Then we try to keep (6.24) for as many $Q_1, Q_2$ as possible.[13]

Even for the restricted class of $Q_1, Q_2$ do ***correlations*** arise, i.e.

$$\operatorname{Tr}\left(\hat{\rho}_t^{\mathrm{S}}\left(\hat{P}_{Q_1}^{\mathrm{S}} \otimes \hat{P}_{Q_2}^{\mathrm{S}}\right)\right) \overset{\text{i.g.}}{\neq} \operatorname{Tr}\left(\hat{\rho}_t^{\mathrm{S}}\left(\hat{P}_{Q_1}^{\mathrm{S}} \otimes \hat{1}\right)\right)\operatorname{Tr}\left(\hat{\rho}_t^{\mathrm{S}}\left(\hat{1} \otimes \hat{P}_{Q_2}^{\mathrm{S}}\right)\right),$$

since:

> Even if the Schrödinger state at time 0 is ***factorized***, i.e. if
>
> $$\hat{\rho}_0^{\mathrm{S}} = \hat{\rho}_1 \otimes \hat{\rho}_2\,,$$
>
> $\hat{\rho}_t^{\mathrm{S}}$ is typically not even separable[14] for $t > 0$.

Correlations of subsystems also arise for classical systems. But:

> The subsystems $\mathcal{S}_1$, $\mathcal{S}_2$ of $\mathcal{S}$ should be considered as ***classically correlated*** at time $t$ only if $\hat{\rho}_t^{\mathrm{S}}$ is separable (Werner, 1989).

### 6.2.2 Partial States

Now let us consider a bipartite system $\mathcal{S}$ as described in 6.2.1 and ask only for properties $Q_1$ of the subsystem $\mathcal{S}$, i.e. for the ***partial state*** of $\mathcal{S}_1$ given by the mapping

$$(Q_1, t) \longmapsto \operatorname{Tr}\left(\hat{\rho}_t^{\mathrm{S}}\left(\hat{P}_{Q_1}^{\mathrm{S}} \otimes \hat{1}\right)\right) = \begin{cases}\text{probability for: "}Q_1 \text{ at time } t\text{"}\\ \text{to be confirmed by optimal tests}\,.\end{cases}$$

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[13] Problems arise whenever the relation between two observables depends on the dynamics; a typical example being the observables of position and velocity.

[14] A (momentary Schrödinger) state with density operator $\hat{\rho}$ is called ***separable*** if for every $\epsilon > 0$ there exist $N \in \mathbb{N}$, $p_1, \ldots, p_N \geq 0$, and density operators $\hat{\rho}_{1,\nu}, \ldots, \hat{\rho}_{2,N}$ with

$$\operatorname{Tr}\left|\hat{\rho} - \sum_{\nu=1}^{N} p_\nu\, \hat{\rho}_{1,\nu} \otimes \hat{\rho}_{2,\nu}\right| < \epsilon\,.$$

Otherwise the state is called ***entangled***.

One may show that (for every $t$) there is a density operator $\mathrm{Tr}_2\left(\hat{\rho}_t^{\mathrm{S}}\right)$ on $\mathcal{H}_1$, called the ***partial trace*** of $\hat{\rho}_t^{\mathrm{S}}$ w.r.t. $\mathcal{H}_2$, for which

$$\mathrm{Tr}\left(\hat{\rho}_t^{\mathrm{S}}\left(\hat{P}_{Q_1}^{\mathrm{S}} \otimes \hat{1}\right)\right) = \mathrm{Tr}\left(\mathrm{Tr}_2\left(\hat{\rho}_t^{\mathrm{S}}\right) \hat{P}_{Q_1}^{\mathrm{S}}\right) \quad \forall\, Q_1 . \tag{6.26}$$

Obviously, $\mathrm{Tr}_2\left(\hat{\rho}_t^{\mathrm{S}}\right)$ represents the partial SCHRÖDINGER state of $\mathcal{S}_1$ at time $t$. But:

> Even if $\hat{\rho}_t^{\mathrm{S}}$ represents a vector state of $\mathcal{S}$ its partial trace w.r.t. $\mathcal{H}_2$ usually does not represent a vector state[15] of $\mathcal{S}_1$.

Moreover, the interaction between $\mathcal{S}_1$ and $\mathcal{S}_2$ has to be very special[16] for all the $\mathrm{Tr}_2\left(\hat{\rho}_t^{\mathrm{S}}\right)$ with $t > 0$ to be determined by $\mathrm{Tr}_2\left(\hat{\rho}_0^{\mathrm{S}}\right)$, at all. Usually, therefore, the HEISENBERG picture does not work for subsystems.

### 6.2.3 Composition of Indistinguishable Systems

Two quantum systems $\mathcal{S}_1$, $\mathcal{S}_2$ are called ***indistinguishable*** if they may be described in the same way, especially with

$$\mathcal{H}_1 = \mathcal{H}_2 , \quad \hat{H}_1 = \hat{H}_2 , \quad \{Q_1\} = \{Q_2\} .$$

Then there is a natural decomposition

$$\mathcal{H}_1 \otimes \mathcal{H}_2 = \mathcal{H}^+ \oplus \mathcal{H}^- ,$$

where $\mathcal{H}^\sigma$ denotes the HILBERT subspace of $\mathcal{H}_1 \otimes \mathcal{H}_2$ generated by all vectors $\Psi$ of the form

$$\Psi = \psi_1 \otimes \psi_2 + \sigma \psi_2 \otimes \psi_1 , \quad \psi_1, \psi_2 \in \mathcal{H}_1 ,$$

and the composition described in 6.2.1 has to be modified in the following way:[17]

1. The state space of the composed system is $H^\sigma$ with $\sigma = +$resp. $\sigma = -$ if $\mathcal{S}_1$ is a ***Boson*** resp. a ***Fermion***.

2. Only those properties of the composed system can be considered as 'testable properties' which do not depend on the enumeration of the subsystems. Thus, for instance, if $Q_1 \neq Q_2$ are compatible properties of $\mathcal{S}_1$ then

   $$(\text{`}Q_1 \text{ for } \mathcal{S}_1\text{'} \wedge \text{`}Q_2 \text{ for } \mathcal{S}_2\text{'}) \vee (\text{`}Q_2 \text{ for } \mathcal{S}_1\text{'} \wedge \text{`}Q_1 \text{ for } \mathcal{S}_2\text{'})$$

   is a 'testable property' of the composed system represented by the projector

   $$\left(\hat{P}_{Q_1}^{\mathrm{S}} \otimes \hat{P}_{Q_2}^{\mathrm{S}} + \hat{P}_{Q_2}^{\mathrm{S}} \otimes \hat{P}_{Q_1}^{\mathrm{S}} - \left(\hat{P}_{Q_1}^{\mathrm{S}}\, \hat{P}_{Q_2}^{\mathrm{S}}\right) \otimes \left(\hat{P}_{Q_1}^{\mathrm{S}}\, \hat{P}_{Q_2}^{\mathrm{S}}\right)\right) \restriction \mathcal{H}^\sigma$$

   in the SCHRÖDINGER picture. But

   $$\text{`}Q_1 \text{ for } \mathcal{S}_1\text{'} \wedge \text{`}Q_2 \text{ for } \mathcal{S}_2\text{'}$$

   cannot be considered as a testable.



[15] The evolution of an initial partial vector state into a mixed state is called ***decoherence***.

[16] See Section 6.3

[17] Here we assume $\hat{V} = 0$, for simplicity.

3. The Hamiltonian has to be replaced by

$$\left(\hat{H}_1 \otimes \hat{1} + \hat{1} \otimes \hat{H}_2 + \hat{V}\right) \upharpoonright \mathcal{H}^\sigma$$

with $\hat{V}$ leaving $\mathcal{H}^\sigma$ invariant.

Recall that particles with integer spin like photons are Bosons while particles with half-integer spin like electrons are Fermions.

For multipartite systems and the possibility of parastatistics we refer to (Ohnuki and Kamefuchi, 1982

## 6.3 Open Systems

See (Davies, 1976; Kraus, 1983; Carmichael, 1993; Alicki, 2003).

# Chapter 7

# Perturbation Theory of Radiative Transitions

*Being able to predict things or describe them, however accurately, is not at all the same thing as understanding them.*

(Deutsch, 1997, p. 2)

## 7.1 Single Atoms in General

### 7.1.1 Naive Interaction Picture

Let $\mathcal{S}_1$ be a single 1-electron atom with approximate Hamiltonian[1]

$$\hat{H}_{\text{atom}} \stackrel{\text{def}}{=} \frac{\hat{\mathbf{p}}_{\text{can}}^2}{2m} + qcA^0(\hat{\mathbf{x}})$$

acting on $\mathcal{H}_{\text{atom}} = L^2(\mathbb{R}^3)$ and let $\mathcal{S}_2$ be the electromagnetic radiation system with state space $\mathcal{H}_{\text{field}}$ and Hamiltonian

$$\hat{H}_{\text{field}} = \frac{1}{2} \int \left( \epsilon_0 : \hat{\mathbf{E}}_\perp(\mathbf{x},0) \cdot \hat{\mathbf{E}}_\perp(\mathbf{x},0) : + \frac{1}{\mu_0} : \hat{\mathbf{B}}(\mathbf{x},0) \cdot \hat{\mathbf{B}}(\mathbf{x},0) : \right) \mathrm{d}V_{\mathbf{x}} \,,$$

where $\hat{\mathbf{E}}_\perp(\mathbf{x},0)$ resp. $\hat{\mathbf{B}}(\mathbf{x},0)$ denotes the observable of the electric[2] resp. magnetic field at time $t = 0$.

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[1]We assume that the atom's center of mass fixed at $\mathbf{x} = 0$ and neglect spin-orbit interaction and the like. $q < 0$ is the electron's charge, $m > 0$ its (reduced) mass , $c \approx 3 \cdot 10^{-10}$cm the velocity of light in vacuum, and $cA^0(\hat{\mathbf{x}})$ the electric (binding) potential of the nucleus. $\hat{\mathbf{p}}_{\text{can}}$ denotes the electron's canonical momentum operator and $\hat{\mathbf{x}}$ its position operator:

$$\left(\hat{p}^j_{\text{can}}\, \psi\right)(\mathbf{x}) = \frac{\hbar}{i} \frac{\partial}{\partial x^j} \psi(\mathbf{x}) \,, \quad \left(\hat{x}^j\, \psi\right)(\mathbf{x}) = x^j\, \psi(\mathbf{x}) \,.$$

[2]Note that only in the absence of charges the total electric field $\hat{\mathbf{E}}(\mathbf{x},0)$ coincides with its *transversal* part $\hat{\mathbf{E}}_\perp(\mathbf{x},0)$ (recall Footnote 33 of Chapter 1).

**Agreement:** From now on we identify operators $\hat{B}$ on $\mathcal{H}_{\text{atom}}$ with the corresponding operators $\hat{B}\otimes\hat{1}_{\text{field}}$ on $\mathcal{H}_{\text{atom}}\otimes\mathcal{H}_{\text{field}}$. Similarly we identify operators $\hat{C}$ on $\mathcal{H}_{\text{field}}$ with the corresponding operators $\hat{1}_{\text{atom}}\otimes\hat{C}$ on $\mathcal{H}_{\text{atom}}\otimes\mathcal{H}_{\text{field}}$.

Since the interaction between $\mathcal{S}_1$ and $\mathcal{S}_2$ cannot be switched off let us consider these systems as components of a closed bipartite system, as described in 6.2.1, with ***minimal coupling***[3]

$$\begin{aligned}\hat{V} &= -\frac{q}{2m}\left(\hat{\mathbf{p}}_{\text{can}}\cdot\hat{\mathbf{A}}\left(\hat{\mathbf{x}},0\right)+\hat{\mathbf{A}}\left(\hat{\mathbf{x}},0\right)\cdot\hat{\mathbf{p}}_{\text{can}}\right)\\ &\quad+\frac{q^2}{2m}{:}\hat{\mathbf{A}}^{\,2}(\hat{\mathbf{x}},0){:}\,,\end{aligned}\tag{7.1}$$

where we assume that the naive interaction picture works, i.e. that the state space and the time zero fields $\hat{\mathbf{E}}_\perp(\mathbf{x},0)$, $\hat{\mathbf{B}}(\mathbf{x},0)$, $\hat{\mathbf{A}}(\mathbf{x},0)$ can be chosen identical with those of the free theory described in 1.2.2.

**Warning:** Actually, as stressed in (Hoffmann, 1994, Sect. 3.5), $\hat{V}$ is **not** well-defined. This is a simple consequence of the equations

$$\begin{aligned}\hat{A}_0^{j_1}(\mathbf{x}_1,t_1)\,\hat{A}_0^{j_2}(\mathbf{x}_2,t_2) &= {:}\hat{A}_0^{j_1}(\mathbf{x}_1,t_1)\,\hat{A}_0^{j_2}(\mathbf{x}_2,t_2){:}\\ &\quad+\left\langle\Omega\mid\hat{A}_0^{j_1}(\mathbf{x}_1,t_1)\,\hat{A}_0^{j_2}(\mathbf{x}_2,t_2)\,\Omega\right\rangle\end{aligned}\tag{7.2}$$

and

$$\begin{aligned}&\left\langle\Omega\mid\hat{A}_0^{j_1}(\mathbf{x}_1,t_1)\,\hat{A}_0^{j_2}(\mathbf{x}_2,t_2)\,\Omega\right\rangle\\ &= \left\langle\Omega\mid\left[(\hat{A}_0^{j_1})^{(+)}(\mathbf{x}_1,t_1)\,,\,\left((\hat{A}_0^{j_2})^{(+)}(\mathbf{x}_2,t_2)\right)^\dagger\right]_-\Omega\right\rangle\\ &\underset{(1.45)}{=} (2\pi)^{-3}\,\mu_0\,\hbar\,c\int\left(\delta_{j_1j_2}-\frac{k^{j_1}k^{j_2}}{|\mathbf{k}|^2}\right)e^{i\mathbf{k}(\mathbf{x}_1-\mathbf{x}_2)}\,e^{-i\,c|\mathbf{k}|(t_1-t_2)}\,\frac{\mathrm{d}V_{\mathbf{k}}}{2\,|\mathbf{k}|}\end{aligned}\tag{7.3}$$

for the **free** field operators $\hat{A}_0(\mathbf{x},t)$. Therefore we should replace $\hat{\mathbf{A}}(\mathbf{x},0)$ by $\int\hat{\mathbf{A}}_0(\mathbf{x}-\mathbf{y},0)\,\varphi(\mathbf{y})\,\mathrm{d}\mathbf{y}$ with $\varphi\in\mathcal{S}(\mathbb{R}^3)$ (introduce a *ultraviolet cutoff* this way) and determine the limit dynamics for $\varphi\to\delta$. However, we are not going to elaborate on such mathematical details.[4]

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[3]Our notation should be understood in the sense that, e.g.,

$$\left\langle\psi'\otimes\chi'\mid\hat{\mathbf{A}}\left(\hat{\mathbf{x}},0\right)(\psi\otimes\chi)\right\rangle\stackrel{\text{def}}{=}\int\left(\psi'(\mathbf{x})\right)^*\psi(\mathbf{x})\left\langle\chi'\mid\hat{\mathbf{A}}\left(\mathbf{x},0\right)\,\chi\right\rangle_{\text{field}}\mathrm{d}V_{\mathbf{x}}$$

holds for sufficiently well-behaved $\psi\otimes\chi,\psi'\otimes\chi'\in\mathcal{H}$. See (Simon, 1971) in this context.

[4]See (Fröhlich et al., 2003) for such techniques.

**Remark:** (7.2) is a special case of WICK's theorem; see, e.g., Sect. 3.2.1 of (Lücke, qft).

Now the time evolution of the composed system is generated by the total Hamiltonian

$$\hat{H} = \hat{H}_{\rm atom} + \hat{H}_{\rm field} + \hat{V} \tag{7.4}$$

$$= \frac{1}{2m} : \Big( \underbrace{\hat{\mathbf{p}}_{\rm can} - q\,\hat{\mathbf{A}}(\hat{\mathbf{x}},0)}_{=\hat{\mathbf{p}}_{\rm kin} \stackrel{\rm def}{=} m\,\frac{i}{\hbar}\left[\hat{H},\hat{\mathbf{x}}\right]_-} \Big)^2 : + qcA^0(\hat{\mathbf{x}}) + \hat{H}_{\rm field}\,. \tag{7.5}$$

For (sufficiently well-behaved) vector states the dynamical law (6.7) suggests validity of the SCHRÖDINGER equation[5]

$$i\hbar \frac{\rm d}{{\rm d}t}\Psi_t = \hat{H}\,\Psi_t \quad \forall\, t \in \mathbb{R}\,, \tag{7.6}$$

where $\hat{\rho}^{\rm S}_t = |\Psi_t\rangle\langle\Psi_t|$ .

**Remark:** See Section 9.1.1 of (Lücke, eine) for arbitrary vector states.

Assuming[6]

$$\left.\begin{array}{l} \Psi_t \approx \psi_t \otimes \chi_t\,, \\ \|\chi_t\| = 1 \end{array}\right\} \quad \forall\, t \in \mathbb{R} \tag{7.7}$$

and taking the inner product of (7.6) with $\psi' \otimes \chi_t$ gives

$$\begin{aligned}
&\int \big(\psi'(\mathbf{x})\big)^* i\hbar \frac{\partial}{\partial t}\psi_t(\mathbf{x})\, {\rm d}V_{\mathbf{x}} \\
&= \left\langle \psi' \otimes \chi_t \,\middle|\, i\hbar \frac{\rm d}{{\rm d}t}(\psi_t \otimes \chi_t) \right\rangle - \langle \psi' \mid \psi_t \rangle \left\langle \chi_t \middle| i\hbar \frac{\rm d}{{\rm d}t}\chi_t \right\rangle \\
&\underset{(7.7),(7.6)}{\approx} \left\langle \psi' \otimes \chi_t \mid \hat{H}\,(\psi_t \otimes \chi_t) \right\rangle - \langle \psi' \mid \psi_t \rangle \left\langle \chi_t \middle| i\hbar \frac{\rm d}{{\rm d}t}\chi_t \right\rangle \\
&\underset{(7.4),(7.1)}{=} \left\langle \psi' \mid \hat{H}_{\rm atom}\,\psi_t \right\rangle + \langle \psi' \mid \psi_t \rangle \left\langle \chi_t \middle| \left(\hat{H}_{\rm field} - i\hbar\frac{\rm d}{{\rm d}t}\right)\chi_t \right\rangle \\
&\quad -\frac{q}{2m}\int \big(\psi'(\mathbf{x})\big)^* \left(\hat{\mathbf{p}}_{\rm can} \cdot \mathbf{A}_{\rm ext}(\mathbf{x},t) + \mathbf{A}_{\rm ext}(\mathbf{x},t) \cdot \hat{\mathbf{p}}_{\rm can}\right)\psi_t(\mathbf{x})\, {\rm d}V_{\mathbf{x}} \\
&\quad +\frac{q^2}{2m}\int \big(\psi'(\mathbf{x})\big)^* \left\langle \chi_t \middle| :\hat{\mathbf{A}}^{\,2}(\mathbf{x},0): \chi_t \right\rangle \psi_t(\mathbf{x})\, {\rm d}V_{\mathbf{x}}\,,
\end{aligned}$$

where

$$\mathbf{A}_{\rm ext}(\mathbf{x},t) \stackrel{\rm def}{=} \left\langle \chi_t \mid \hat{\mathbf{A}}(\mathbf{x},0)\,\chi_t \right\rangle .$$



[5]Note, however, that the time evolution of $\hat{\rho}^{\rm S}_t$ — contrary to that of $\Psi_t$ — is not changed by addition of a constant to $\hat{H}$ .

[6]This possibility seems reasonable for coherent states $\chi_0$ ; see end of 7.2.1.

Since $\psi'$ is arbitrary, this establishes the ***exterior field formalism***

$$i\hbar \frac{\partial}{\partial t}\psi_t(\mathbf{x}) \approx \left(\frac{1}{2m}\left(\frac{\hbar}{i}\boldsymbol{\nabla} - q\mathbf{A}_{\mathrm{ext}}(\mathbf{x},t)\right)^2 + qcA^0_{\mathrm{ext}}(\mathbf{x},t) + qcA^0(\mathbf{x})\right)\psi_t(\mathbf{x})\,, \tag{7.8}$$

**if** (7.7) holds,

with suitably defined $A^0_{\mathrm{ext}}(\mathbf{x},t)$. We postpone the discussion under which conditions (7.7) is reasonable.[7]

### 7.1.2 Electric Field Representation[8]

Let us choose some $\varphi \in \mathcal{S}(\mathbb{R}^3)$ with

$$\varphi(\mathbf{x}) = \left(\varphi(\mathbf{x})\right)^* \approx \delta(\mathbf{x}) \tag{7.9}$$

and exploit the WIGNER symmetry corresponding to the unitary operator

$$\hat{U} = \exp\left(-\frac{i}{\hbar}\,q\,\hat{\mathbf{x}} \cdot \int \hat{\mathbf{A}}(\mathbf{y},0)\,\varphi(\mathbf{y})\,d\mathbf{y}\right)\,. \tag{7.10}$$

**Remark:** See,e.g., Section 8.3.4 of (Lücke, eine) for the notion of WIGNER ***symmetry***.

This means that all operators $\hat{B}$ have to be replaced by

$$\mathrm{Ad}_{\hat{U}}\hat{B} \stackrel{\mathrm{def}}{=} \hat{U}\,\hat{B}\,\hat{U}^{-1} \tag{7.11}$$

without changing the physical interpretation. The new observable for the transversal part of the electric field, for example, in the SCHRÖDINGER picture is now

$$\mathrm{Ad}_{\hat{U}}\hat{\mathbf{E}}_\perp(\mathbf{x},0) = \hat{\mathbf{E}}_\perp(\mathbf{x},0) - \frac{1}{\epsilon_0}\hat{\mathbf{P}}^{(0)}_\perp(\mathbf{x})\,, \tag{7.12}$$

where

$$\hat{\mathbf{P}}^{(0)}_\perp(\mathbf{x}) \stackrel{\mathrm{def}}{=} \epsilon_0 \sum_{j=1}^{3} q\,\hat{x}^j\,\frac{i}{\hbar}\int \left[\hat{A}^j(\mathbf{y},0), \hat{\mathbf{E}}_\perp(\mathbf{x},0)\right]_-\varphi(\mathbf{y})\,d\mathbf{y}\,. \tag{7.13}$$

**Proof of (7.12):** As usual, we (formally) define

$$\mathrm{ad}_{\hat{A}}\,\hat{B} \stackrel{\mathrm{def}}{=} \left[\hat{A},\hat{B}\right]_-\,.$$

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[7]See (Gemmer and Mahler, 2001) for the case of bipartite systems with finite dimensional state spaces.

[8]Here we follow (Cohen-Tannoudji et al., 1992, Appendix 5), to some extent, rather than (Mandel and Wolf, 1995, Section 14.1.3). See also (Scully and Zubairy, 1999, App. 5.A) for the corresponding exterior field formalism.

Then (1.37) implies that $\mathrm{ad}_{\ln(\hat{U})}\hat{\mathbf{E}}_\perp(\mathbf{x},0)$ is a function of $\hat{\mathbf{x}}$ and, consequently,

$$\left(\mathrm{ad}_{\ln(\hat{U})}\right)^\nu \hat{\mathbf{E}}_\perp(\mathbf{x},0) = 0 \quad \forall\, \nu > 1\,.$$

Therefore (7.12) follows from the CAMPBELL-HAUSDORFF ***formula***[9]

$$\mathrm{Ad}_{\exp(\hat{A})}\,\hat{B} = \exp(\mathrm{ad}_{\hat{A}})\,\hat{B} \tag{7.14}$$

and

$$\mathrm{ad}_{\ln(\hat{U})}\hat{\mathbf{E}}_\perp(\mathbf{x},0) = -\frac{i}{\hbar}\,q\sum_{j=1}^{3}\hat{x}^j\int\left[\hat{A}^j(\mathbf{y},0),\hat{\mathbf{E}}_\perp(\mathbf{x},0)\right]_-\,\varphi(\mathbf{y})\,\mathrm{d}\mathbf{y}\,. \qquad \blacksquare$$

Similarly we get:

$$\begin{aligned}
\mathrm{Ad}_{\hat{U}}\,\hat{\mathbf{x}} &= \hat{\mathbf{x}}\,,\\
\mathrm{Ad}_{\hat{U}}\,\hat{\mathbf{p}}_{\mathrm{kin}} &= \hat{\mathbf{p}}_{\mathrm{kin}} + q\,\frac{i}{\hbar}\sum_{j=1}^{3}\left[\hat{\mathbf{p}}_{\mathrm{can}},\hat{x}^j\right]_-\int\hat{A}^j(\mathbf{y},0)\,\varphi(\mathbf{y})\,\mathrm{d}\mathbf{y}\\
&= \hat{\mathbf{p}}_{\mathrm{can}} - q\,\hat{\mathbf{A}}\,(\hat{\mathbf{x}},0) + q\int\hat{\mathbf{A}}(\mathbf{y},0)\,\varphi(\mathbf{y})\,\mathrm{d}\mathbf{y} \qquad (7.15)\\
\mathrm{Ad}_{\hat{U}}\hat{\mathbf{B}}(\mathbf{x},0) &\underset{(1.48)}{=} \hat{\mathbf{B}}(\mathbf{x},0)\,,\\
\mathrm{Ad}_{\hat{U}}\hat{H} &= \frac{1}{2m}{:}(\mathrm{Ad}_{\hat{U}}\hat{\mathbf{p}}_{\mathrm{kin}})^2{:} + qcA^0(\hat{\mathbf{x}}) + \hat{H}_{\mathrm{field}}\\
&\quad -\int\hat{\mathbf{P}}^{(0)}_\perp(\mathbf{x})\cdot\hat{\mathbf{E}}_\perp(\mathbf{x},0)\,\mathrm{d}V_{\mathbf{x}} + \frac{1}{2\epsilon_0}\int\left(\hat{\mathbf{P}}^{(0)}_\perp(\mathbf{x})\right)^2\mathrm{d}V_{\mathbf{x}}\,(7.16)
\end{aligned}$$

**Proof of (7.16):** According to (7.5) it is sufficient to show

$$\mathrm{Ad}_{\hat{U}}\hat{H}_{\mathrm{field}} = \hat{H}_{\mathrm{field}} - \int\hat{\mathbf{P}}^{(0)}_\perp(\mathbf{x})\cdot\hat{\mathbf{E}}_\perp(\mathbf{x},0)\,\mathrm{d}V_{\mathbf{x}} + \frac{1}{2\epsilon_0}\int\left(\hat{\mathbf{P}}^{(0)}_\perp(\mathbf{x})\right)^2\mathrm{d}V_{\mathbf{x}}\,.$$

This follows from

$$\mathrm{Ad}_{\hat{U}}\hat{H}_{\mathrm{field}} = \frac{1}{2}\int\left(\epsilon_0 : \left(\mathrm{Ad}_{\hat{U}}\hat{\mathbf{E}}_\perp(\mathbf{x},0)\right)^2 : + \frac{1}{\mu_0} : \hat{\mathbf{B}}^2(\mathbf{x},0) :\right)\mathrm{d}V_{\mathbf{x}}$$

and (7.12). $\blacksquare$



[9]Formally (7.14) is a consequence of the fact that $\mathrm{Ad}_{\exp(\lambda\hat{A})}\,\hat{B}$ and $\exp\!\left(\mathrm{ad}_{\lambda\hat{A}}\right)\hat{B}$, considered as operator-valued functions of $\lambda$, fulfill the same first order differential equation and the same initial condition for $\lambda = 0$.

Note that (1.49) and (7.13) imply[10]

$$\hat{\mathbf{P}}_\perp^{(0)}(\mathbf{x}) = q \sum_{j,j'=1}^{3} \hat{x}^j \, \mathbf{e}_{j'} \int \delta_\perp^{jj'}(\mathbf{y}-\mathbf{x}) \, \varphi(\mathbf{y}) \, \mathrm{d}\mathbf{y} \,. \tag{7.17}$$

Since

$$\int \delta_\perp^{jj'}(\mathbf{y}-\mathbf{x}) \, \hat{E}_\perp^{j'}(\mathbf{x}) \, \mathrm{d}V_\mathbf{x} = \hat{E}_\perp^{j}(\mathbf{y}) \,,$$

(7.17) implies

$$\int \hat{\mathbf{P}}_\perp^{(0)}(\mathbf{x}) \cdot \hat{\mathbf{E}}_\perp(\mathbf{x},0) \, \mathrm{d}V_\mathbf{x} = q \, \hat{\mathbf{x}} \cdot \int \hat{\mathbf{E}}_\perp(\mathbf{y},0) \, \varphi(\mathbf{y}) \, \mathrm{d}\mathbf{y} \,.$$

Substituting the latter into (7.16) we get

$$\boxed{\begin{aligned} \mathrm{Ad}_{\hat{U}} \hat{H} \;=\; & \left( \frac{1}{2m} \!:\! \left(\mathrm{Ad}_{\hat{U}} \, \hat{\mathbf{p}}_{\mathrm{kin}}\right)^2 \!:\! + qcA^0(\hat{\mathbf{x}}) + \frac{1}{2\epsilon_0} \int \left(\hat{\mathbf{P}}_\perp^{(0)}(\mathbf{x})\right)^2 \mathrm{d}V_\mathbf{x} \right) + \hat{H}_{\mathrm{field}} \\ & - q \, \hat{\mathbf{x}} \cdot \int \hat{\mathbf{E}}_\perp(\mathbf{y},0) \, \varphi(\mathbf{y}) \, \mathrm{d}\mathbf{y} \,. \end{aligned}} \tag{7.18}$$

Therefore we call the new representation, resulting from the Wigner symmetry transformation $\mathrm{Ad}_{\hat{U}}$, the ***electric field representation***.

> **Warning:** The new observable of the (transversal part of the) electric field is $\mathrm{Ad}_{\hat{U}} \hat{\mathbf{E}}_\perp(\mathbf{x},0)$ rather than $\hat{\mathbf{E}}_\perp(\mathbf{x},0)$. According to (7.12)/(7.13), both coincide only 'outside' the atom.

### 7.1.3 First Order Perturbation Theory

Let us go back to the representation described in 7.1.1 which we shall call the Coulomb ***representation***. Then we have

$$\hat{H}_0 \stackrel{\mathrm{def}}{=} \hat{H}_{\mathrm{atom}} + \hat{H}_{\mathrm{field}}$$



[10]The observable of the transversal polarization density is the operator-valued distribution

$$\begin{aligned} \hat{\mathbf{P}}_\perp(\mathbf{x}) & \stackrel{\mathrm{def}}{=} q \sum_{j,j'=1}^{3} \hat{x}^j \, \mathbf{e}_{j'} \delta_\perp^{jj'}(\hat{\mathbf{x}}-\mathbf{x}) \\ & = \epsilon_0 \sum_{j=1}^{3} q \, \hat{x}^j \, \frac{i}{\hbar} \left[ \hat{A}^j(\hat{\mathbf{x}},0), \hat{\mathbf{E}}_\perp(\mathbf{x},0) \right]_- \,. \end{aligned}$$

as *unperturbed* Hamiltonian and

$$\hat{V}^{\mathrm{I}}(t) \underset{(6.17)}{=} e^{+\frac{i}{\hbar}\hat{H}_0\,t}\,\hat{V}\,e^{-\frac{i}{\hbar}\hat{H}_0\,t}$$

with $\hat{V}$ given by (7.1).

Let us consider the special case

$$\left.\begin{array}{rcl} \hat{\rho}_0^{\mathrm{I}} &=& |\psi\otimes\chi\rangle\langle\psi\otimes\chi|\;, \\ \hat{P} &=& |\check{\psi}\otimes\check{\chi}\rangle\langle\check{\psi}\otimes\check{\chi}|\;, \\ \hat{P}\,\hat{\rho}_0^{\mathrm{I}} &=& \hat{\rho}_0^{\mathrm{I}}\,\hat{P} = 0\,. \end{array}\right\} \tag{7.19}$$

Then[11]

$$\begin{aligned}
&p_t\left(\psi\otimes\chi\to\check{\psi}\otimes\check{\chi}\right)\\
&\quad\overset{\mathrm{def}}{=}\quad \mathrm{Tr}\left(\hat{\rho}_t^{\mathrm{I}}\,\hat{P}\right)\\
&\quad\underset{(6.19)}{\approx}\quad \hbar^{-2}\left|\int_0^t\left\langle\check{\psi}\otimes\check{\chi}\mid\hat{V}^{\mathrm{I}}(t')\,(\psi\otimes\chi)\right\rangle\mathrm{d}t'\right|^2\\
&\quad\underset{(6.17)}{=}\quad \hbar^{-2}\left|\int_0^t\left\langle\check{\psi}_{t'}^{(0)}\otimes\check{\chi}_{t'}^{(0)}\mid\hat{V}\left(\psi_{t'}^{(0)}\otimes\chi_{t'}^{(0)}\right)\right\rangle\mathrm{d}t'\right|^2\\
&\quad\underset{(7.1)}{=}\quad \hbar^{-2}\left|\int_0^t\left(-\frac{q}{m}\int\sum_{j=1}^3\left\langle\check{\chi}_{t'}^{(0)}\mid\hat{A}^j(\mathbf{x},0)\,\chi_{t'}^{(0)}\right\rangle\left(\check{\psi}_{t'}^{(0)}(\mathbf{x})\right)^*\frac{\hbar}{i}\frac{\partial}{\partial x^j}\,\psi_{t'}^{(0)}(\mathbf{x})\,\mathrm{d}V_{\mathbf{x}}\right.\right.\\
&\qquad\qquad\left.\left.+\frac{q^2}{2m}\int\left\langle\check{\chi}_{t'}^{(0)}\mid:\hat{\mathbf{A}}^2(\mathbf{x},0):\chi_{t'}^{(0)}\right\rangle\left(\check{\psi}_{t'}^{(0)}(\mathbf{x})\right)^*\psi_{t'}^{(0)}(\mathbf{x})\,\mathrm{d}V_{\mathbf{x}}\right)\mathrm{d}t'\right|^2,\\
&\quad=\quad \hbar^{-2}\left|\int_0^t\left(-\frac{q}{m}\int\sum_{j=1}^3\left\langle\check{\chi}\mid\hat{A}^j(\mathbf{x},t')\,\chi\right\rangle\left(\check{\psi}_{t'}^{(0)}(\mathbf{x})\right)^*\frac{\hbar}{i}\frac{\partial}{\partial x^j}\,\psi_{t'}^{(0)}(\mathbf{x})\,\mathrm{d}V_{\mathbf{x}}\right.\right.\\
&\qquad\qquad\left.\left.+\frac{q^2}{2m}\int\left\langle\check{\chi}\mid:\left(\hat{\mathbf{A}}\right)^2(\mathbf{x},t'):\chi\right\rangle\left(\check{\psi}_{t'}^{(0)}(\mathbf{x})\right)^*\psi_{t'}^{(0)}(\mathbf{x})\,\mathrm{d}V_{\mathbf{x}}\right)\mathrm{d}t'\right|^2,
\end{aligned}$$

where we used

$$\hat{\mathbf{A}}(\mathbf{x},t) = e^{+\frac{i}{\hbar}\hat{H}_{\mathrm{field}}\,t}\,\hat{\mathbf{A}}(\mathbf{x},0)\,e^{-\frac{i}{\hbar}\hat{H}_{\mathrm{field}}\,t} \tag{7.20}$$

and

$$\psi_t^{(0)} \overset{\mathrm{def}}{=} e^{-\frac{i}{\hbar}\hat{H}_{\mathrm{atom}}\,t}\,\psi\;,\quad \check{\psi}_t^{(0)} \overset{\mathrm{def}}{=} e^{-\frac{i}{\hbar}\hat{H}_{\mathrm{atom}}\,t}\,\check{\psi}\,,$$

$$\chi_t^{(0)} \overset{\mathrm{def}}{=} e^{-\frac{i}{\hbar}\hat{H}_{\mathrm{field}}\,t}\,\chi\;,\quad \check{\chi}_t^{(0)} \overset{\mathrm{def}}{=} e^{-\frac{i}{\hbar}\hat{H}_{\mathrm{field}}\,t}\,\check{\chi}\,.$$

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[11] Note that $\boldsymbol{\nabla}\cdot\hat{\mathbf{A}}(\mathbf{x},0) = 0\,.$

Usually $\left\langle \check{\chi} \mid : \left(\hat{\mathbf{A}}\right)^2 (\mathbf{x}, t') : \chi \right\rangle$ will be neglected:[12]

$$\begin{aligned} & p_t\big(\psi \otimes \chi \to \check{\psi} \otimes \check{\chi}\big) \\ & \approx \frac{q^2}{m^2 \hbar^2} \left| \int_0^t \Big( \int \sum_{j=1}^3 \left\langle \check{\chi} \mid \hat{A}^j(\mathbf{x}, t')\, \chi \right\rangle \left( \check{\psi}_{t'}^{(0)}(\mathbf{x}) \right)^* \frac{\hbar}{i} \frac{\partial}{\partial x^j}\, \psi_{t'}^{(0)}(\mathbf{x})\, \mathrm{d}V_{\mathbf{x}} \Big) \mathrm{d}t' \right|^2 \\ & \textbf{if } (7.19) \text{ holds}. \end{aligned} \tag{7.21}$$

For obvious reasons[13]

$$p_t\big(\psi \otimes \chi \to \check{\psi} \otimes \check{\chi}\big) \underset{(6.7),(6.12)}{=} \operatorname{Tr}\Big( \hat{\rho}_t^{\mathrm{S}}\, e^{-\frac{i}{\hbar} \hat{H}_0 t}\, \hat{P}\check{}\, e^{+\frac{i}{\hbar} \hat{H}_0 t} \Big)$$

is called the ***transition probability*** for the transition of the SCHRÖDINGER state $\widehat{=}\, e^{-\frac{i}{\hbar} \hat{H}_0 t} (\psi \otimes \chi)$ into the SCHRÖDINGER state $\widehat{=}\, e^{-\frac{i}{\hbar} \hat{H}_0 t} \big( \check{\psi} \otimes \check{\chi} \big)$ at time $t$.

In typical quantum optical applications also the approximation

$$\begin{aligned} & \int \sum_{j=1}^3 \left\langle \check{\chi} \mid \hat{A}^j(\mathbf{x}, t')\, \chi \right\rangle \left( \check{\psi}_{t'}^{(0)}(\mathbf{x}) \right)^* \frac{\hbar}{i} \frac{\partial}{\partial x^j}\, \psi_{t'}^{(0)}(\mathbf{x})\, \mathrm{d}V_{\mathbf{x}} \\ \approx\ & \int \sum_{j=1}^3 \left\langle \check{\chi} \mid \hat{A}^j(0, t')\, \chi \right\rangle \left( \check{\psi}_{t'}^{(0)}(\mathbf{x}) \right)^* \frac{\hbar}{i} \frac{\partial}{\partial x^j}\, \psi_{t'}^{(0)}(\mathbf{x})\, \mathrm{d}V_{\mathbf{x}} \\ =\ & \left\langle \check{\chi} \mid \hat{\mathbf{A}}(0, t')\, \chi \right\rangle \cdot \left\langle \check{\psi}_{t'}^{(0)} \mid \hat{\mathbf{p}}_{\mathrm{can}}\, \psi_{t'}^{(0)} \right\rangle \\ =\ & \left\langle \check{\chi} \mid \hat{\mathbf{A}}(0, t')\, \chi \right\rangle \cdot \left\langle \check{\psi}_{t'}^{(0)} \mid m\, \frac{i}{\hbar} \left[ \hat{H}_{\mathrm{atom}}, \hat{\mathbf{x}} \right]_- \psi_{t'}^{(0)} \right\rangle \\ =\ & \frac{m}{q} \left\langle \check{\chi} \mid \hat{\mathbf{A}}(0, t')\, \chi \right\rangle \cdot \frac{\mathrm{d}}{\mathrm{d}t'} \left\langle \check{\psi}_{t'}^{(0)} \mid q\, \hat{\mathbf{x}}\, \psi_{t'}^{(0)} \right\rangle \end{aligned}$$

is justified. Substituting this into (7.21) we get the so-called ***electric dipole approximation***

$$\boxed{\begin{aligned} & p_t\big(\psi \otimes \chi \to \check{\psi} \otimes \check{\chi}\big) \\ & \approx \int_{[0,t]^2} \sum_{j_1, j_2 = 1}^3 \left\langle \chi \mid \hat{A}^{j_1}(0, t_1)\, \check{\chi} \right\rangle \left\langle \check{\chi} \mid \hat{A}^{j_2}(0, t_2)\, \chi \right\rangle f_{\psi, \check{\psi}}^{j_1 j_2}(t_1, t_2)\, \mathrm{d}t_1\, \mathrm{d}t_2\,, \\ & \text{in the COULOMB representation, where} \\ & f_{\psi, \check{\psi}}^{j_1 j_2}(t_1, t_2) \stackrel{\mathrm{def}}{=} \hbar^{-2} \left( \frac{\mathrm{d}}{\mathrm{d}t_1} \left\langle \psi_{t_1}^{(0)} \mid q\, \hat{x}^{j_1}\, \check{\psi}_{t_1}^{(0)} \right\rangle \right) \left( \frac{\mathrm{d}}{\mathrm{d}t_2} \left\langle \check{\psi}_{t_2}^{(0)} \mid q\, \hat{x}^{j_2}\, \psi_{t_2}^{(0)} \right\rangle \right). \end{aligned}} \tag{7.22}$$

Obviously, the $f$-factor gives rise to ***selection rules***:[14]

$$\left\langle \check{\psi}_{t'}^{(0)} \mid q\, \hat{\mathbf{x}}\, \psi_{t'}^{(0)} \right\rangle = 0\ \forall\, t' \in [0, t] \quad \Longrightarrow \quad p_t\big(\psi \otimes \chi \to \check{\psi} \otimes \check{\chi}\big) \approx 0\,. \tag{7.23}$$



[12] See (Mandel and Wolf, 1995, End of Section 14.1.2) for justification.

[13] Recall (6.6).

[14] The l.h.s. of (7.23) is often implied by symmetries.

**Remark:** Note that
$$\left\langle \check{\chi} \mid \hat{\mathbf{A}}(\mathbf{x},t)\,\Omega \right\rangle \overset{\text{i.g.}}{\neq} 0 \quad \left( = \left\langle \Omega \mid \hat{\mathbf{A}}(\mathbf{x},t)\,\Omega \right\rangle \right).$$
Therefore (7.22) may be non-negligible even for $\chi = \Omega$. This explains, e.g., the occurrence of *spontaneous decay*[15] of excited atomic states in the vacuum.

# 7.2 Photoelectric Detection of Light

## 7.2.1 Ionisation of One-Electron Atoms

Let us assume that the projection $\hat{P}_{\text{ion}}$ onto the subspace of $\mathcal{H}_{\text{atom}}$ corresponding to ionisation is of the form
$$\hat{P}_{\text{ion}} = \int_{\check{E}\geq 0} \underbrace{\sigma(\check{E},\check{\Omega})}_{\geq 0} \left|\check{\psi}(\check{E},\check{\Omega})\right\rangle\left\langle\check{\psi}(\check{E},\check{\Omega})\right| \, \mathrm{d}\check{E}\,\mathrm{d}\check{\Omega}\,, \tag{7.24}$$
where the $\check{\psi}(\check{E},\check{\Omega})$ are (improper) eigenstates of $\hat{H}_{\text{atom}}$ :
$$\hat{H}_{\text{atom}}\,\check{\psi}(\check{E},\check{\Omega}) = \check{E}\,\check{\psi}(\check{E},\check{\Omega})\,.$$
Moreover, let $\{\check{\chi}_\nu\}_{\nu\in\mathbb{N}}$ be a maximal orthonormal system (MONS) of $\mathcal{H}_{\text{field}}$. Then, if
$$\hat{H}_{\text{atom}}\,\psi = E\,\psi\,, \quad E < 0\,,$$
the probability $p_t^{\text{ion}}(\psi,\chi)$ for ionisation at time $t$ is
$$\begin{aligned}
&p_t^{\text{ion}}(\psi,\chi)\\
&\quad = \operatorname{Tr}\!\left(\hat{\rho}_t^{\mathrm{I}}\left(\hat{P}_{\text{ion}}\otimes\hat{1}\right)\right)\\
&\quad = \sum_{\nu=1}^{\infty}\int_{\check{E}\geq 0}\sigma(\check{E},\check{\Omega})\,p_t\!\left(\psi\otimes\chi\to\check{\psi}(\check{E},\check{\Omega})\otimes\check{\chi}_\nu\right)\mathrm{d}\check{E}\,\mathrm{d}\check{\Omega}\\
&\underset{(7.22)}{\approx} \int_{[0,t]^2}\sum_{j_1,j_2=1}^{3}\left\langle\chi\mid\hat{A}^{j_1}(0,t_1)\,\hat{A}^{j_2}(0,t_2)\,\chi\right\rangle I_\psi^{j_1 j_2}(t_1-t_2)\,\mathrm{d}t_1\,\mathrm{d}t_2\,,
\end{aligned}$$
where
$$\begin{aligned}
&I_\psi^{j_1 j_2}(t_1-t_2)\\
&\overset{\text{def}}{=} \int_{\check{E}\geq 0}\sigma(\check{E},\check{\Omega})\,\left(E-\check{E}\right)^2\left\langle\psi\mid q\,\hat{x}^{j_1}\,\check{\psi}(\check{E},\check{\Omega})\right\rangle\left\langle\check{\psi}(\check{E},\check{\Omega})\mid q\,\hat{x}^{j_2}\,\psi\right\rangle\cdot\\
&\qquad\qquad\cdot e^{+\frac{i}{\hbar}\left(E-\check{E}\right)(t_1-t_2)}\,\mathrm{d}\check{E}\,\mathrm{d}\check{\Omega}\\
&= \int_{\check{E}\geq 0}\sigma(\check{E},\check{\Omega})\,f^{j_1 j_2}_{\psi,\check{\psi}(\check{E},\check{\Omega})}(t_1,t_2)\,\mathrm{d}\check{E}\,\mathrm{d}\check{\Omega}\,.
\end{aligned}$$



[15]See, e.g., (Baym, 1969, Chapter 13).

This together with (7.2) gives

$$\begin{aligned} &p_t^{\mathrm{ion}}(\psi,\chi) \\ &\approx \int_{[0,t]^2} \sum_{j_1,j_2=1}^{3} \left\langle \chi \mid :\hat{A}^{j_1}(0,t_1)\,\hat{A}^{j_2}(0,t_2):\chi \right\rangle I_\psi^{j_1 j_2}(t_1-t_2)\,\mathrm{d}t_1\,\mathrm{d}t_2 + \mathcal{V}_t(\psi)\,, \end{aligned} \tag{7.25}$$

where

$$\begin{aligned} \mathcal{V}_t(\psi) \quad &\stackrel{\mathrm{def}}{=} \quad \int_{[0,t]^2} \sum_{j_1,j_2=1}^{3} \left\langle \Omega \mid \hat{A}^{j_1}(0,t_1)\,\hat{A}^{j_2}(0,t_2)\,\Omega \right\rangle I_\psi^{j_1 j_2}(t_1-t_2)\,\mathrm{d}t_1\,\mathrm{d}t_2 \\ &\underset{(7.3)}{=} \quad (2\pi)^{-3}\,\mu_0\,\hbar\,c \sum_{j_1,j_2=1}^{3} \int \left( \delta_{j_1 j_2} - \frac{k^{j_1}k^{j_2}}{|\mathbf{k}|^2} \right) \cdot \\ &\qquad \cdot \left( \int_{[0,t]^2} I_\psi^{j_1 j_2}(t_1-t_2)\,e^{-i\,c|\mathbf{k}|(t_1-t_2)}\,\mathrm{d}t_1\,\mathrm{d}t_2 \right) \frac{\mathrm{d}V_{\mathbf{k}}}{2\,|\mathbf{k}|}\,. \end{aligned} \tag{7.26}$$

Since

$$\left\langle \Omega \mid :\hat{A}^{j_1}(0,t_1)\,\hat{A}^{j_2}(0,t_2):\Omega \right\rangle = 0\,,$$

we have

$$\mathcal{V}_t(\psi) \underset{(7.25)}{\approx} p_t^{\mathrm{ion}}(\psi,\Omega)$$

and, therefore, expect $\mathcal{V}_t(\psi)$ to be negligible.

**Additional consideration:** By change of variables[16]

$$(t_1,t_2) \longrightarrow (\tau,t') \stackrel{\mathrm{def}}{=} \left( t_1 - t_2, \frac{t_1+t_2}{2} \right)$$

we see — in the typical case,[17] that $I_\psi^{j_1 j_2}(\tau)$ is sufficiently concentrated around $\tau = 0$ —- that

$$\begin{aligned} \int_{[0,t]^2} I_\psi^{j_1 j_2}(t_1-t_2)\,e^{-i\,c|\mathbf{k}|(t_1-t_2)}\,\mathrm{d}t_1\,\mathrm{d}t_2 \quad &= \quad \int_{-t}^{+t} \left( I_\psi^{j_1 j_2}(\tau)\,e^{-i\,c|\mathbf{k}|\tau} \int_{\left|\frac{\tau}{2}\right|}^{t-\left|\frac{\tau}{2}\right|} \mathrm{d}t' \right) \mathrm{d}\tau \\ &= \quad \int_{-t}^{+t} (t-|\tau|)\, I_\psi^{j_1 j_2}(\tau)\,e^{-i\,c|\mathbf{k}|\tau}\,\mathrm{d}\tau \\ &\approx \quad t \int_{-t}^{+t} I_\psi^{j_1 j_2}(\tau)\,e^{-i\,c|\mathbf{k}|\tau}\,\mathrm{d}\tau \\ &\approx \quad t \int_{-\infty}^{+\infty} I_\psi^{j_1 j_2}(\tau)\,e^{-i\,c|\mathbf{k}|\tau}\,\mathrm{d}\tau \end{aligned}$$



[16]Note that

$$(t_1,t_2) \in [0,t]^2 \quad \Longleftrightarrow \quad 0 \le t' \pm \frac{\tau}{2} \le t \quad \Longleftrightarrow \quad \left|\frac{\tau}{2}\right| \le t' \le t - \left|\frac{\tau}{2}\right|\,.$$

[17]See comments to (Mandel and Wolf, 1995, (14.2-13)).

for relevant $t$. Together with (7.26) and

$$\begin{aligned}
&\int_{-\infty}^{+\infty} I_\psi^{j_1 j_2}(\tau)\, e^{-i\,c|\mathbf{k}|\tau}\, \mathrm{d}\tau \\
&= \int_{\check{E}\geq 0} \sigma(\check{E},\check{\Omega})\, \left(E-\check{E}\right)^2 \langle \psi \mid q\,\hat{x}^{j_1}\, \check{\psi}(\check{E},\check{\Omega})\rangle \langle \check{\psi}(\check{E},\check{\Omega}) \mid q\,\hat{x}^{j_2}\,\psi\rangle \cdot \\
&\qquad \cdot \underbrace{\left(\int_{-\infty}^{+\infty} e^{-\frac{i}{\hbar}\left(\check{E}-(E-\hbar c|\mathbf{k}|)\right)\tau}\, \mathrm{d}\tau\right)}_{=2\pi\,\delta\left(\check{E}-\underbrace{(E-\hbar c\,|\mathbf{k}|)}_{<0}\right)} \mathrm{d}\check{E}\,\mathrm{d}\check{\Omega} \\
&= 0
\end{aligned}$$

this implies $\mathcal{V}_t(\psi,\chi) \approx 0$.

Therefore, by (7.25),

$$p_t^{\mathrm{ion}}(\psi,\chi) \approx \int_{[0,t]^2} \sum_{j_1,j_2=1}^{3} \left\langle \chi \mid : \hat{A}^{j_1}(0,t_1)\, \hat{A}^{j_2}(0,t_2) : \chi \right\rangle I_\psi^{j_1 j_2}(t_1-t_2)\, \mathrm{d}t_1\, \mathrm{d}t_2 \tag{7.27}$$

for relevant $t$. Obviously,

for normalized **coherent** states $\chi$:

$$\begin{aligned}
\left\langle \chi \mid : \hat{A}^{j_1}(\mathbf{x}_1,t_1)\, \hat{A}^{j_2}(\mathbf{x}_2,t_2) : \chi \right\rangle &= A_\chi^{j_1}(\mathbf{x}_1,t_1)\, A_\chi^{j_2}(\mathbf{x}_2,t_2)\,, \\
\text{where[18]} \quad A_\chi^{j}(\mathbf{x},t) &\stackrel{\mathrm{def}}{=} \left\langle \chi \mid \hat{A}^{j}(\mathbf{x},t)\, \chi \right\rangle .
\end{aligned} \tag{7.28}$$

Since, on the other hand,

$$\begin{aligned}
&\mathrm{Tr}\Big(\hat{\rho}_t^{\mathrm{I}}\left(\hat{P}_{\mathrm{ion}} \otimes |\chi\rangle\langle\chi|\right)\Big) \\
&= \sum_{\nu=1}^{\infty} \int_{\check{E}\geq 0} \sigma(\check{E},\check{\Omega})\, p_t\big(\psi\otimes\chi \to \check{\psi}(\check{E},\check{\Omega}) \otimes \check{\chi}_\nu\big)\, \mathrm{d}\check{E}\, \mathrm{d}\check{\Omega} \\
&\underset{(7.22)}{\approx} \int_{[0,t]^2} \sum_{j_1,j_2=1}^{3} \left\langle \chi \mid \hat{A}^{j_1}(0,t_1)\, \chi \right\rangle \left\langle \chi \mid \hat{A}^{j_2}(0,t_2)\, \chi \right\rangle I_\psi^{j_1 j_2}(t_1-t_2)\, \mathrm{d}t_1\, \mathrm{d}t_2\,,
\end{aligned}$$

we see that

$$p_t^{\mathrm{ion}}(\psi,\chi) \approx \mathrm{Tr}\Big(\hat{\rho}_t^{\mathrm{I}}\left(\hat{P}_{\mathrm{ion}} \otimes |\chi\rangle\langle\chi|\right)\Big)\,.$$

This indicates that the following holds within the limits of our approximations:

> If the (partial) state of the radiation field is a coherent one at time $t=0$, then the interaction influences only the time evolution of the atom's (partial) state.[19]



[18] Usually, the positive-frequency part $A_\chi^{(+)^j}(\mathbf{x},t)$ of $A_\chi^j(\mathbf{x},t)$ is called the ***analytic signal*** associated with $A_\chi^j(\mathbf{x},t)$.

[19] Nevertheless the total energy is conserved thanks to the interaction term (7.1).

Recalling the definitions of $I_\psi^{j_1 j_2}(t_1-t_2)$ and $\hat{\mathbf{A}}(\mathbf{x},t)$ and interchanging integration over $t_1, t_2$ with integration over $\check{E}, \check{\Omega}$ we easily see that (7.27) implies

$$\boxed{p_t^{\mathrm{ion}}(\psi,\chi) \approx \int_{[0,t]^2} \sum_{j_1,j_2=1}^{3} \left\langle \chi \mid \hat{A}^{(-)^{j_1}}(0,t_1)\, \hat{A}^{(+)^{j_2}}(0,t_2)\,\chi \right\rangle I_\psi^{j_1 j_2}(t_1-t_2)\,\mathrm{d}t_1\,\mathrm{d}t_2} \tag{7.29}$$

for sufficiently well-behaved states $\chi$.

### 7.2.2 Simple Photodetectors

A typical photodetector consists of an assembly of 'atoms' to be ionized by the incident radiation. The electrons thus emitted from the 'atoms' are counted.[20] In the following by ' detector' we always mean a photodetector of this type.

First of all, let us consider the special case that the radiation field is initially in a **coherent** state $\chi$ (in the interaction picture). Then, as we have seen, this state remains essentially unchanged by the ionisation processes. Therefore the above considerations of the previous section make it plausible[21] that the counting rate at time $t_0$ of such a detector, localized at $\mathbf{x}_0$, is

$$P_{|\chi\rangle\langle\chi|}(\mathbf{x}_0,t_0) \approx \left( \frac{\partial}{\partial t} \int_{[t_0,t_0+t]^2} \sum_{j_1,j_2=1}^{3} \left\langle \chi \middle| \hat{A}^{(-)^{j_1}}(\mathbf{x}_0,t_1)\, \hat{A}^{(+)^{j_2}}(\mathbf{x}_0,t_2)\,\chi \right\rangle K^{j_1 j_2}(t_1-t_2)\,\mathrm{d}t_1\,\mathrm{d}t_2 \right)_{|t=0}, \tag{7.30}$$

where $K_\psi^{j_1 j_2}(t_1-t_2)$ is a characteristic function of the detector.

**Remarks:**

1. Here, by 'counting rate at time $t_0$' we mean the production rate for photoelectrons at time $t_0$. Of course, actual registration needs some extra time.[22]
2. Strictly speaking, in (7.30), we should not use the derivative w.r.t. $t$ at $t=0$ but rather choose some 'relevant' $t$ and divide by this $t$. This is because $p_t^{\mathrm{ion}}(\psi,\chi)$ becomes proportional to $t^2$ for **too** small $t$.
3. Generalization of the latter is also the reason for the so-called ***quantum*** Zeno ***effect***:[23]



[20] Actually, what we mean by 'atom' here is an electron bound in some photosensitive material. Ionisation of ordinary atoms would require frequencies above the optical domain.

[21] See (Mandel and Wolf, 1995, Sections 414.2–14.4) for further details.

[22] See, e.g., (Paul, 1995, Chapter 5).

[23] See (Lynds, 2003) for Zeno's paradoxes and, concerning quantum mechanics, (Misra and Sudarshan, 1977).

> A quantum mechanical system does not change its state as long as it is permanently checked, by ideal tests, whether it is in this state.

4. Justification of (7.30) as a consequence of (7.29) is not as straightforward, as usually tacitly assumed:

   (7.29) is the probability for finding the atom ionized **if** measured at time $t$. So in order to relate (7.29) to actual ionisation we have to assume that such measurements automatically occur at random times. However, these 'measurements' must not appear too often in order to avoid the ***quantum*** ZENO ***effect***!

In typical cases we may substitute $\eta^{j_1 j_2}\,\delta(t_1 - t_2)$ for $K^{j_1 j_2}(t_1 - t_2)$, where $\eta^{j_1 j_2}$ describes the efficiency of the counter (slowly varying with the frequency range of the radiation field). Then (7.30) becomes[24]

$$\boxed{P_{|\chi\rangle\langle\chi|}(\mathbf{x}_0, t_0) \approx \sum_{j_1, j_2 = 1}^{3} \left\langle \chi \right| \hat{A}^{(-)^{j_1}}(\mathbf{x}_0, t_0)\, \hat{A}^{(+)^{j_2}}(\mathbf{x}_0, t_0)\, \chi \Big\rangle\, \eta^{j_1 j_2}\,.} \tag{7.31}$$

Now let us consider $n$ detectors located at the positions $\mathbf{x}_1, \ldots, \mathbf{x}_n$ and define the corresponding ***probability density***[25]

$$P_{|\chi\rangle\langle\chi|}(\mathbf{x}_1, t_1; \ldots; \mathbf{x}_n, t_n) \stackrel{\text{def}}{=} \left( \frac{\partial}{\partial t'_1} \cdots \frac{\partial}{\partial t'_n}\, p_\chi(\mathbf{x}_1, t_1, t'_1; \ldots; \mathbf{x}_n, t_n, t'_n) \right)_{|t'_1 = \ldots = t'_n = 0}\,,$$

for the radiation field being initially in the **coherent** state $\chi$, where $p_\chi(\mathbf{x}_1, t_1, t'_1; \ldots; \mathbf{x}_n, t_n, t'_n)$ is the probability for the following effect:[26]

> For all $\nu \in \{1, \ldots, n\}$ the detector localized at $\mathbf{x}_\nu$ 'clicks' during the time interval $(t_\nu, t'_\nu)$.

Then, assuming that the coherent state suffers no relevant changes by the detectors, we get by (7.31)

$$\begin{aligned}
&P_{|\chi\rangle\langle\chi|}(\mathbf{x}_1, t_1; \ldots; \mathbf{x}_n, t_n) \\
&\approx \prod_{\nu=1}^{n} \sum_{j_\nu, j'_\nu = 1}^{3} \left\langle \chi \right| \hat{A}^{(-)^{j_\nu}}(\mathbf{x}_\nu, t_\nu)\, \hat{A}^{(+)^{j'_\nu}}(\mathbf{x}_\nu, t_\nu)\, \chi \Big\rangle\, \eta_\nu^{j_\nu j'_\nu} \\
&= \sum_{j_1, \ldots, j_n, j'_1, \ldots, j'_n = 1}^{3} \left\langle \chi \right| \hat{A}^{(-)^{j_1}}(\mathbf{x}_1, t_1) \cdots \hat{A}^{(-)^{j_n}}(\mathbf{x}_n, t_n) \hat{A}^{(+)^{j'_n}}(\mathbf{x}_n, t_n) \cdots \\
&\qquad\qquad \cdots \hat{A}^{(+)^{j'_1}}(\mathbf{x}_1, t_1)\, \chi \Big\rangle\, \eta_1^{j_1 j'_1} \cdots \eta_n^{j_n j'_n}\,,
\end{aligned} \tag{7.32}$$



[24] Recall Footnote 16.

[25] Here Remark 1 to (7.30) applies accordingly.

[26] For instance, $P_{\hat{\rho}^{\mathrm{I}}}(\mathbf{x}_1, t_1; \mathbf{x}_2, t_2)$ is measured in the HANBURY BROWN-TWISS experiment; see, e.g., (Mandel and Wolf, 1995, Sections 9.9, 9.10, and 14.6.1).

where, for $\nu \in \{1, \dots, n\}$, the $\eta_\nu^{j_1 j_2}$ characterize the efficiency of the detector localized at $\mathbf{x}_\nu$.

Since $P_{\hat\rho^{\mathrm{I}}}(\mathbf{x}_1, t_1; \dots; \mathbf{x}_n, t_n)$ — to be defined for general $\hat\rho^{\mathrm{I}}$ accordingly[27] — is linear (and suitably continuous) in $\hat\rho^{\mathrm{I}}$ Theorem 1.2.2 tells us that

$$\begin{aligned} &P_{\hat\rho^{\mathrm{I}}}(\mathbf{x}_1, t_1; \dots; \mathbf{x}_n, t_n) \\ &\quad = \lim_{N\to\infty} \int \rho_N(\alpha_1, \dots, \alpha_N) P_{\Phi_{\alpha_1,\dots,\alpha_N}}(\mathbf{x}_1, t_1; \dots; \mathbf{x}_n, t_n) \frac{d\alpha_1}{\pi} \cdots \frac{d\alpha_N}{\pi} \end{aligned}$$

holds with suitable functions $\rho_N$ with[28]

$$\hat\rho^{\mathrm{I}} = \lim_{N\to\infty} \int \rho_N(\alpha_1, \dots, \alpha_N) \, |\Phi_{\alpha_1,\dots,\alpha_N}\rangle\langle\Phi_{\alpha_1,\dots,\alpha_N}| \, \frac{d\alpha_1}{\pi} \cdots \frac{d\alpha_N}{\pi} \,. \tag{7.33}$$

This together with (7.32) and the optical equivalence theorem gives[29]

$$\begin{aligned} &P_{\hat\rho^{\mathrm{I}}}(\mathbf{x}_1, t_1; \dots; \mathbf{x}_n, t_n) \\ &\approx \sum_{j_1,\dots,j_n,j'_1,\dots,j'_n=1}^{3} \mathrm{Tr}\left(\hat\rho^{\mathrm{I}} \, \hat A^{(-)^{j_1}}(\mathbf{x}_1, t_1) \cdots \hat A^{(-)^{j_n}}(\mathbf{x}_n, t_n) \hat A^{(+)^{j'_n}}(\mathbf{x}_n, t_n) \cdots \right. \\ &\qquad\qquad \left. \cdots \hat A^{(+)^{j'_1}}(\mathbf{x}_1, t_1)\right) \eta_1^{j_1 j'_1} \cdots \eta_n^{j_n j'_n} \,. \end{aligned} \tag{7.34}$$



[27] From now on we denote by $\hat\rho^{\mathrm{I}}$ the density matrix of the (partial) state of the electromagnetic field — rather than of the total system — in the interaction picture.

[28] In spite of (7.33), $\hat\rho^{\mathrm{I}}$ need not be an ordinary mixture of coherent states, since the $\rho_N$ need not be nonnegative functions.

[29] Admittedly, this argument involves an interchange of limits that ought to be justified.

# Chapter 8

# Semiclassical Treatment of Few Level Atomic Systems

## 8.1 The Exterior Electric Field Representation

Let us recall the exterior field formalism for a single atom as introduced in 7.1.1:

The atom's state space is

$$\mathcal{H}_{\text{atom}} = L^2(\mathbb{R}^3, \mathrm{d}^3\mathbf{x})$$

and its free Hamiltonian is

$$\hat{H}_{\text{atom}} = \frac{1}{2m}\left(\frac{\hbar}{i}\boldsymbol{\nabla}_{\mathbf{x}}\right)^2 + \underbrace{q\,c\,A^0_{\text{atom}}(\mathbf{x})}_{\text{Bindungspotential}} \ .$$

Interaction with the (optical) radiation field is introduced via ***minimal coupling***:[1]

$$\hat{H}_{\text{atom}} \longmapsto \hat{H}^{\mathrm{S}}(t) \quad \stackrel{\text{def}}{=} \quad \frac{1}{2m}\left(\frac{\hbar}{i}\boldsymbol{\nabla}_{\mathbf{x}} - q\,\mathbf{A}_{\text{ext}}(\mathbf{x},t)\right)^2 + q\,c\,A^0_{\text{atom}}(\mathbf{x})\,.$$

As usual, we evaluate the Schrödinger equation

$$i\,\frac{\mathrm{d}}{\mathrm{d}t}\,\hbar\,\psi_t = \hat{H}^{\mathrm{S}}(t)\,\psi_t$$

only in the so-called ***dipole approximation***

$$\hat{H}^{\mathrm{S}}(t)\psi_t(\mathbf{x}) = \left(\frac{1}{2m}\left(\frac{\hbar}{i}\boldsymbol{\nabla}_{\mathbf{x}} - q\,\mathbf{A}(t)\right)^2 + q\,c\,A^0_{\text{atom}}(\mathbf{x})\right)\psi_t(\mathbf{x})\,,$$

where

$$\mathbf{A}(t) \stackrel{\text{def}}{=} \mathbf{A}_{\text{ext}}(0,t) \quad \forall\, t \in \mathbb{R}\,.$$

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[1] Here we use the Schrödinger picture.

By change of representation

$$\psi_t \;\mapsto\; \phi_t \stackrel{\text{def}}{=} \hat{U}(t)\,\psi_t\,,$$

where

$$\hat{U}(t) \stackrel{\text{def}}{=} e^{-\frac{i}{\hbar}\,q\,\mathbf{x}\cdot\mathbf{A}(t)}\,,$$

the SCHRÖDINGER equation (in dipole approximation) becomes

$$\boxed{\begin{aligned} i\,\hbar\,\frac{\partial}{\partial t}\,\phi_t(\mathbf{x}) &\approx \hat{H}_{\text{cl}}(t)\,\phi_t(\mathbf{x})\,,\\ \text{where:}^{2}\quad \hat{H}_{\text{cl}}(t) &\stackrel{\text{def}}{=} -\frac{\hbar^2}{2m}\Delta_{\mathbf{x}} - q\,\mathbf{x}\cdot\mathbf{E}(t) + q\,c\,A^0_{\text{atom}}(\mathbf{x})\,,\\ \mathbf{E}(t) &\stackrel{\text{def}}{=} -\frac{\partial}{\partial t}\mathbf{A}_{\text{ext}}(0,t)\,. \end{aligned}} \tag{8.1}$$

**Warning:** Recall that — strictly speaking — the usual interpretation of

$$-\frac{\hbar^2}{2m}\Delta_{\mathbf{x}} + q\,c\,A^0_{\text{atom}}(\mathbf{x})$$

as observable of the energy of the atom is not correct.[3]

## 8.2 2-Level Systems

Let us assume that (within the considered time interval essentially) only a *ground* state $|\mathrm{g}\rangle$ and an *excited* state $|\mathrm{e}\rangle$ are occupied[4] corresponding to the energy eigenvalues $E_{\mathrm{g}}, E_{\mathrm{e}}$ :

$$\hat{H}_{\text{atom}}\,|\mathrm{g}\rangle = E_{\mathrm{g}}\,|\mathrm{g}\rangle \quad,\quad \hat{H}_{\text{atom}}\,|\mathrm{e}\rangle = E_{\mathrm{e}}\,|\mathrm{e}\rangle \ .$$

Then, because of

$$\hat{\rho}^{\mathrm{S}}_{\text{atom}}(t) \approx \hat{P}_{\mathcal{H}^{(2)}_{\text{atom}}(t)}\hat{\rho}^{\mathrm{S}}_{\text{atom}}(t)\hat{P}_{\mathcal{H}^{(2)}_{\text{atom}}(t)}\,,$$

restriction to the 2-dimensional subspace

$$\mathcal{H}^{(2)}_{\text{atom}}(t) \stackrel{\text{def}}{=} \mathcal{L}\Big(\{|\mathrm{g}\rangle\,,|\mathrm{e}\rangle\}\Big)$$



[2]Note that:

$$i\,\hbar\,\frac{\partial}{\partial t}\,\hat{U}(t)\,\psi_t(\mathbf{x}) = \left(q\,\mathbf{x}\cdot\frac{\partial}{\partial t}\mathbf{A}(t)\right)\hat{U}(t)\,\psi_t(\mathbf{x})\,,\quad \hat{U}(t)\,\frac{\hbar}{i}\,\boldsymbol{\nabla}_{\mathbf{x}}\,\hat{U}(t)^{-1} = \frac{\hbar}{i}\,\boldsymbol{\nabla}_{\mathbf{x}} + q\,\mathbf{A}(t)\,.$$

[3]See also (Scully and Zubairy, 1999, 5.1.3) and (Shore, 1990a, End of § 3.11).

[4]For simplicity we assume that interaction with the electromagnetic field does not change the level scheme.

of $\mathcal{H}_{\mathrm{atom}}$ is adequate:

$$\boxed{\begin{aligned} i\hbar \frac{\mathrm{d}}{\mathrm{d}t}\,\hat{\rho}^{(2)}_{\mathrm{atom}}(t) &\approx \left[\hat{H}^{(2)}_{\mathrm{cl}}(t), \hat{\rho}^{(2)}_{\mathrm{atom}}(t)\right]_- \\ \text{where:}\quad \hat{\rho}^{(2)}_{\mathrm{atom}}(t) &\stackrel{\mathrm{def}}{=} \frac{\hat{P}_{\mathcal{H}^{(2)}_{\mathrm{atom}}(t)}\,\hat{\rho}^{\mathrm{S}}_{\mathrm{atom}}(t) \restriction \mathcal{H}^{(2)}_{\mathrm{atom}}(t)}{\mathrm{Spur}\left(\hat{P}_{\mathcal{H}^{(2)}_{\mathrm{atom}}(t)}\,\hat{\rho}^{\mathrm{S}}_{\mathrm{atom}}(t)\,\hat{P}_{\mathcal{H}^{(2)}_{\mathrm{atom}}(t)}\right)}\,, \\ \hat{H}^{(2)}_{\mathrm{cl}}(t) &\stackrel{\mathrm{def}}{=} \hat{P}_{\mathcal{H}^{(2)}_{\mathrm{atom}}(t)}\hat{H}_{\mathrm{cl}}(t) \restriction \mathcal{H}^{(2)}_{\mathrm{atom}}(t)\,. \end{aligned}} \tag{8.2}$$

W.r.t. the basis

$$\left(|\mathrm{e}\rangle = \begin{pmatrix}1\\0\end{pmatrix}, |\mathrm{g}\rangle = \begin{pmatrix}0\\1\end{pmatrix}\right)$$

we have

$$\begin{aligned} |\mathrm{g}\rangle\langle\mathrm{e}| &= \hat{b} \stackrel{\mathrm{def}}{=} \begin{pmatrix}0&0\\1&0\end{pmatrix}, \\ |\mathrm{e}\rangle\langle\mathrm{g}| &= \hat{b}^\dagger = \begin{pmatrix}0&1\\0&0\end{pmatrix}, \\ |\mathrm{e}\rangle\langle\mathrm{e}| &= \hat{b}^\dagger\,\hat{b} = \begin{pmatrix}1&0\\0&0\end{pmatrix}, \\ |\mathrm{g}\rangle\langle\mathrm{g}| &= \hat{b}\,\hat{b}^\dagger = \begin{pmatrix}0&0\\0&1\end{pmatrix} = \hat{\tau}^0 - \hat{b}^\dagger\,\hat{b} \end{aligned}$$

and hence

$$\begin{aligned} \hat{H}^{(2)}_{\mathrm{cl}}(t) &= E_{\mathrm{e}}\,|\mathrm{e}\rangle\langle\mathrm{e}| + E_{\mathrm{g}}\,|\mathrm{g}\rangle\langle\mathrm{g}| - q\hat{P}_{\mathcal{H}^{(2)}_{\mathrm{atom}}(t)}\mathbf{x}\cdot\mathbf{E}_{\mathrm{ext}}(\mathbf{x},t)\restriction\mathcal{H}^{(2)}_{\mathrm{atom}}(t) \\ &= \hbar\,\omega_0\hat{b}^\dagger\,\hat{b} + \underbrace{E_{\mathrm{g}}\,\hat{\tau}^0}_{\text{irrelevant}} - q\hat{P}_{\mathcal{H}^{(2)}_{\mathrm{atom}}(t)}\mathbf{x}\cdot\mathbf{E}_{\mathrm{ext}}(\mathbf{x},t)\restriction\mathcal{H}^{(2)}_{\mathrm{atom}}(t)\,, \end{aligned} \tag{8.3}$$

where:

$$\boxed{\omega_0 \stackrel{\mathrm{def}}{=} \frac{E_{\mathrm{e}} - E_{\mathrm{g}}}{\hbar}\,.}$$

For

$$\mathbf{E}_{\mathrm{ext}}(\mathbf{x},t) = 0\,,$$

because of

$$\hat{b}^\dagger\,\hat{b} = \frac{\hat{\tau}^0 + \hat{\tau}^3}{2}\,,$$

the Hamiltonian may be written as

$$\hat{H}^{(2)}_{\mathrm{cl}}(t) = \frac{\hbar\,\omega_0}{2}\,\hat{\tau}^3 + \underbrace{\frac{E_{\mathrm{g}} + E_{\mathrm{e}}}{2}\,\hat{\tau}^0}_{\text{irrelevant}} \tag{8.4}$$

and the Liouville equation (8.2) simplifies to

$$i\hbar\frac{\mathrm{d}}{\mathrm{d}t}\hat{\rho}^{(2)}_{\mathrm{atom}}(t) = \left[\frac{\hbar\,\omega_0}{2}\,\hat{\tau}^3, \hat{\rho}^{(2)}_{\mathrm{atom}}(t)\right]_-\,. \tag{8.5}$$

(8.5) can easily solved by means of the BLOCH ***representation***[5]

$$\hat{\rho}^{(2)}_{\text{atom}}(t) = \frac{1}{2}\left(\hat{\tau}^0 + \underbrace{\mathbf{x}(t)}_{\in\mathbb{R}^3}\cdot\hat{\boldsymbol{\tau}}\right). \tag{8.6}$$

Here

$$\hat{\tau}^0 \stackrel{\text{def}}{=} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad \hat{\tau}^1 \stackrel{\text{def}}{=} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \hat{\tau}^2 \stackrel{\text{def}}{=} \begin{pmatrix} 0 & -i \\ +i & 0 \end{pmatrix}, \quad \hat{\tau}^3 \stackrel{\text{def}}{=} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \tag{8.7}$$

are the well-known PAULI-***matrices***[6] — forming an orthonormal basis of $\mathcal{B}(\mathcal{H}_m)$ w.r.t. the HILBERT-SCHMIDT norm and obeying the relations

$$\begin{aligned} \hat{\tau}^\nu\hat{\tau}^\nu &= \hat{1} && \forall\,\nu \in \{0,1,2,3\}\,, \\ \hat{\tau}^j\hat{\tau}^k &= -\,\hat{\tau}^k\hat{\tau}^j && \forall\,j,k \in \{1,2,3\}\,, j \neq k\,, \\ \hat{\tau}^1\hat{\tau}^2 &= i\,\hat{\tau}^3\,, \\ \hat{\tau}^2\hat{\tau}^3 &= i\,\hat{\tau}^1\,, \\ \hat{\tau}^3\hat{\tau}^1 &= i\,\hat{\tau}^2 \end{aligned} \tag{8.8}$$

(see, e.g., Section 7.2.2 of (Lücke, eine)). In this connection let us note that

$$\hat{\tau}^3 = \hat{b}^\dagger\hat{b} - \hat{b}\,\hat{b}^\dagger = 2\,\hat{b}^\dagger\hat{b} - \hat{\tau}^0\,, \tag{8.9}$$

where the FERMI annihilation operator

$$\hat{b} \stackrel{\text{def}}{=} \frac{\hat{\tau}^1 - i\,\hat{\tau}^2}{2} = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \tag{8.10}$$

acts as

$$\hat{b}\,|g\rangle = 0\,, \quad \hat{b}\,|e\rangle = |g\rangle\,, \quad \hat{b}^\dagger\,|g\rangle = |e\rangle\,, \quad \hat{b}^\dagger\,|e\rangle = 0\,. \tag{8.11}$$

and fulfills the canonical anti-commutation relations

$$\left[\hat{b},\hat{b}^\dagger\right]_+ = \hat{\tau}^0\,, \quad \left[\hat{b},\hat{b}\right]_+ = 0\,. \tag{8.12}$$

In the BLOCH representation (8.5) is equivalent to

$$\begin{aligned} i\,\tfrac{\hbar}{2}\dot{\mathbf{x}}(t)\cdot\hat{\boldsymbol{\tau}} &= \frac{\hbar\,\omega_0}{4}\,\mathbf{x}(t)\cdot[\hat{\tau}^3,\hat{\boldsymbol{\tau}}]_- \\ &= i\,\frac{\hbar}{2}\,\omega_0\left(x^1(t)\,\hat{\tau}^2 - x^2(t)\,\hat{\tau}^1\right) \\ &= i\,\frac{\hbar}{2}\,\omega_0\,\mathbf{e}_3\cdot\left(\mathbf{x}(t)\times\hat{\boldsymbol{\tau}}\right), \end{aligned}$$

hence also to

$$\boxed{\dot{\mathbf{x}}(t) = \omega_0\,\mathbf{e}_3 \times \mathbf{x}(t)\,;}$$

i.e.:

The BLOCH ***vector*** $\mathbf{x}(t)$ rotates[7] with constant angular frequency $\omega_0\,\mathbf{e}_3$.

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[5]See (Leggett, 2001, p. 8) for generalization.
[6]A widespread notation is $\sigma_\nu$ instead of $\hat{\tau}^\nu$.
[7]Since spontaneous decay was not taken into account.

### 8.2.1 Interpretation of the Bloch Vector

Obviously, positivity of $\hat{\rho}^{(2)}_{\text{atom}}(t)$ implies

$$\det\left(\hat{\rho}^{(2)}_{\text{atom}}(t)\right) \geq 0$$

and hence

$$|\mathbf{x}(t)| \leq 1\,,$$

since

$$\begin{aligned}\det\left(\hat{\rho}^{(2)}_{\text{atom}}(t)\right) &\underset{(8.6)}{=} \det\left(\frac{1}{2}\left(\hat{\tau}^0 + \underbrace{\mathbf{x}(t)}_{\in\mathbb{R}^3}\cdot\hat{\boldsymbol{\tau}}\right)\right)\\ &= \frac{1-|\mathbf{x}(t)|^2}{4}\,.\end{aligned}$$

Since, moreover,

$$\begin{aligned}\hat{\rho}^{(2)}_{\text{atom}}(t)\text{ projector} &\iff \left(\hat{\rho}^{(2)}_{\text{atom}}(t)\right)^2 = \hat{\rho}^{(2)}_{\text{atom}}(t)\\ &\iff \frac{1}{4}\left(\hat{\tau}^0 + 2\,\mathbf{x}(t)\cdot\hat{\boldsymbol{\tau}} + |\mathbf{x}(t)|^2\,\hat{\tau}^0\right) = \frac{1}{2}\left(\hat{\tau}^0 + \mathbf{x}(t)\cdot\hat{\boldsymbol{\tau}}\right)\\ &\iff |\mathbf{x}(t)|^2 = 1\end{aligned}$$

we conclude:

$$\hat{\rho}^{(2)}_{\text{atom}}(t)\text{ describes a }\begin{cases}\text{pure state} & \text{if } |\mathbf{x}(t)| = 1\,,\\ \text{mixed state} & \text{if } |\mathbf{x}(t)| < 1\,.\end{cases}$$

Especially we have

$$\begin{aligned}\frac{1}{2}\left(\hat{\tau}^0 + \begin{pmatrix}\sin\vartheta\,\cos\varphi\\ \sin\vartheta\,\sin\varphi\\ \cos\vartheta\end{pmatrix}\cdot\hat{\boldsymbol{\tau}}\right) &= \begin{pmatrix}\cos^2\frac{\vartheta}{2} & \frac{1}{2}\,e^{-i\,\varphi}\,\sin\vartheta\\ \frac{1}{2}\,e^{+i\,\varphi}\,\sin\vartheta & \sin^2\frac{\vartheta}{2}\end{pmatrix}\\ &= \left|\begin{pmatrix}e^{-i\,\frac{\varphi}{2}}\,\cos\frac{\vartheta}{2}\\ e^{+i\,\frac{\varphi}{2}}\,\sin\frac{\vartheta}{2}\end{pmatrix}\right\rangle\left\langle\begin{pmatrix}e^{-i\,\frac{\varphi}{2}}\,\cos\frac{\vartheta}{2}\\ e^{+i\,\frac{\varphi}{2}}\,\sin\frac{\vartheta}{2}\end{pmatrix}\right|\end{aligned}$$

and[8]

$$\begin{aligned}\hat{\rho}_{\mathbf{x}} &\overset{\text{def}}{=} \frac{1}{2}\left(\hat{\tau}^0 + \mathbf{x}\cdot\hat{\boldsymbol{\tau}}\right)\\ &= \frac{1+|\mathbf{x}|}{2}\,\hat{\rho}_{\frac{\mathbf{x}}{|\mathbf{x}|}} + \frac{1-|\mathbf{x}|}{2}\,\hat{\rho}_{-\frac{\mathbf{x}}{|\mathbf{x}|}} \quad \forall\,\mathbf{x}\in\mathbb{R}^3\setminus\{0\}\,. \end{aligned} \tag{8.13}$$

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[8]The vector $\mathbf{x}$ characterizing $\hat{\rho}_{\mathbf{x}}$ is called Bloch ***vector*** resp. Stokes ***vector*** if $\hat{\rho}_{\mathbf{x}}$ describes electron spin resp. photon polarization. The set of all normalized Bloch vectors resp. Stokes vectors is called Bloch ***sphere*** resp. Poincare ***sphere***. In both cases $|\mathbf{x}|$ is called the ***degree of polarization***.

Furthermore, we have

$$\boxed{\hat{\rho}^{(2)}_{\text{atom}}(t) = \hat{\rho}_{\mathbf{x}(t)} \implies \langle\hat{\boldsymbol{\mu}}\rangle(t) = \Re\Big(q\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\Big)\,x^1(t) + \Im\Big(q\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\Big)\,x^2(t)\,,} \tag{8.14}$$

where

$$\hat{\boldsymbol{\mu}} \stackrel{\text{def}}{=} q\,\hat{P}_{\mathcal{H}^{(2)}_{\text{atom}}(t)}\hat{\mathbf{x}}\,\hat{P}_{\mathcal{H}^{(2)}_{\text{atom}}(t)}$$

denotes the observable of the atom's electric dipole moment (truncateded to $\mathcal{H}^{(2)}_{\text{atom}}(t)$).

**Outline of proof for (8.14):** The statement follows from

$$\begin{aligned}
\langle\hat{\boldsymbol{\mu}}\rangle(t) &\stackrel{\text{def}}{=} \mathrm{Tr}\left(\hat{\rho}^{(2)}_{\text{atom}}(t)\,\hat{\boldsymbol{\mu}}\right)\\
&= \mathrm{Tr}\left(\frac{1}{2}\left(\hat{\tau}^0 + \mathbf{x}(t)\cdot\hat{\boldsymbol{\tau}}\right)\Big(q\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\,\hat{b} + \text{h.c.}\Big)\right)\\
&= \frac{1}{2}\,\mathrm{Tr}\Big(\mathbf{x}(t)\cdot\hat{\boldsymbol{\tau}}\,\Big(q\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\,\hat{b} + \text{h.c.}\Big)\Big)
\end{aligned}$$

and

$$\begin{aligned}
\mathrm{Tr}\left(\hat{\boldsymbol{\tau}}\,\hat{b}\right) &= \frac{1}{2}\,\mathrm{Tr}\left(\hat{\boldsymbol{\tau}}\hat{\tau}^1\right) - \frac{i}{2}\,\mathrm{Tr}\left(\hat{\boldsymbol{\tau}}\hat{\tau}^2\right)\\
&= \mathbf{e}_1 - i\,\mathbf{e}_2\,,\\
\mathrm{Tr}\left(\hat{\boldsymbol{\tau}}\,\hat{b}^\dagger\right) &= \mathbf{e}_1 + i\,\mathbf{e}_2\,. \qquad \blacksquare
\end{aligned}$$

### 8.2.2 Rotating Wave Approximation

For $\mathbf{E}(t) \neq 0$ we have

$$q\,\hat{P}_{\mathcal{H}^{(2)}_{\text{atom}}(t)}\hat{\mathbf{x}}\cdot\mathbf{E}(t)\,\hat{P}_{\mathcal{H}^{(2)}_{\text{atom}}(t)} = \hat{\boldsymbol{\mu}}\cdot\mathbf{E}(t)\,.$$

This, together with (8.2) and (8.3) implies

$$i\hbar\,\frac{\mathrm{d}}{\mathrm{d}t}\,\hat{\rho}^{(2)}_{\text{atom}}(t) \approx \left[\hbar\,\omega_0\,\hat{b}^\dagger\hat{b} - \hat{\boldsymbol{\mu}}\cdot\mathbf{E}_{\text{ext}}(0,t), \hat{\rho}^{(2)}_{\text{atom}}(t)\right]_-\,. \tag{8.15}$$

In case[9]

$$\mathbf{E}_{\text{ext}}(0,t) \approx \boldsymbol{\mathcal{E}}\,e^{-i\,\omega\,t} + \text{c.c.} \tag{8.16}$$

it is easier to determine

$$\hat{\rho}_\omega(t) \stackrel{\text{def}}{=} e^{+i\,\omega\,\hat{b}^\dagger\hat{b}\,t}\,\hat{\rho}^{(2)}_{\text{atom}}(t)\,e^{-i\,\omega\,\hat{b}^\dagger\hat{b}\,t}$$

for which (8.15) implies

$$i\hbar\,\frac{\mathrm{d}}{\mathrm{d}t}\,\hat{\rho}_\omega(t) \approx \left[\hbar\,(\omega_0 - \omega)\,\hat{b}^\dagger\hat{b} - \hat{\boldsymbol{\mu}}_\omega(t)\cdot\mathbf{E}_{\text{ext}}(0,t), \hat{\rho}_\omega(t)\right]_-\,,$$



[9] In (Mandel and Wolf, 1995, Eq. (15.3-9)) $\mathbf{E}(t)$ is allowed to vary slowly with time.

where

$$\hat{\boldsymbol{\mu}}_\omega(t) \stackrel{\text{def}}{=} e^{+i\omega\,\hat{b}^\dagger\hat{b}\,t}\,\hat{\boldsymbol{\mu}}\,e^{-i\omega\,\hat{b}^\dagger\hat{b}\,t}\,.$$

Usually the conditions

$$\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{g}\rangle = \langle \mathrm{e}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle = 0$$

are fulfilled.[10] Then

$$e^{+i\omega\,\hat{b}^\dagger\hat{b}\,t}\,\hat{b}\,e^{-i\omega\,\hat{b}^\dagger\hat{b}\,t} \underset{(8.12)}{=} \hat{b}\,e^{-i\omega\,t}$$

implies

$$\begin{aligned}\hat{\boldsymbol{\mu}}_\omega(t)\cdot\mathbf{E}_{\text{ext}}(0,t) &= \left(q\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\,\hat{b}\,e^{-i\omega\,t} + h.c.\right)\cdot\left(\boldsymbol{\mathcal{E}}\,e^{-i\omega\,t} + c.c.\right)\\ &= \left(q\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\cdot\boldsymbol{\mathcal{E}}^*\,\hat{b} + h.c.\right)\\ &\quad + \left(q\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\cdot\boldsymbol{\mathcal{E}}\,\hat{b}\,e^{-i\,2\,\omega\,t} + h.c.\right)\,,\end{aligned}$$

and this is usually replaced by the so-called ***rotating wave approximation*** [11]

$$\hat{\boldsymbol{\mu}}_\omega(t)\cdot\mathbf{E}_{\text{ext}}(0,t) \approx q\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\cdot\boldsymbol{\mathcal{E}}^*\,\hat{b} + h.c.\,.$$

All approximations together result in the equation

$$i\hbar\,\frac{\mathrm{d}}{\mathrm{d}t}\,\hat{\rho}_\omega(t) \approx \left[\hbar\,(\omega_0-\omega)\,\hat{b}^\dagger\hat{b} - \frac{\hbar}{2}\left(\Omega\,\hat{b} + h.c.\right),\hat{\rho}_\omega(t)\right]_-\,,$$

where[12]

$$\Omega \stackrel{\text{def}}{=} \frac{2\,q}{\hbar}\,\langle \mathrm{g}|\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\cdot\boldsymbol{\mathcal{E}}^*\,.$$

For suitable choice of the origin of the time scale we have

$$\boxed{\Omega = |\Omega| \qquad \text{RABI } \textit{frequency}}$$

and, in the BLOCH representation

$$\hat{\rho}_\omega(t) = \frac{1}{2}\left(\hat{\tau}^0 + \mathbf{x}_\omega(t)\cdot\hat{\boldsymbol{\tau}}\right)\,,$$

this implies

$$\begin{aligned} i\hbar\,\dot{\mathbf{x}}_\omega(t)\cdot\hat{\boldsymbol{\tau}} &= \mathbf{x}_\omega(t)\cdot\left[\hbar\,(\omega_0-\omega)\,\hat{b}^\dagger\hat{b} - \frac{\hbar}{2}\,\Omega\left(\hat{b}+\hat{b}^\dagger\right),\hat{\boldsymbol{\tau}}\right]_-\\ &= i\hbar\Big((\omega_0-\omega)\,\mathbf{e}_3\times\mathbf{x}_\omega(t)\Big)\cdot\hat{\boldsymbol{\tau}} - \frac{\hbar}{2}\,\Omega\,\mathbf{x}_\omega(t)\cdot[\hat{\tau}^1,\hat{\boldsymbol{\tau}}]_-\,.\end{aligned}$$

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[10] Especially if $|\mathrm{g}\rangle\,,|\mathrm{e}\rangle$ are parity eigenstates.

[11] For exact solutions of related problems not using the rotating wave approximation approximation see (Angelo and Wreszinski, 2005) and (Chen et al., 2010).

[12] Note the used convention: $\mathbf{E}_{\text{ext}}(0,t) = \Re\big(2\,\boldsymbol{\mathcal{E}}\,e^{-i\,\omega\,t}\big)\,.$

Using

$$\frac{i}{2}[\hat{\tau}^j, \hat{\boldsymbol{\tau}}]_- = \mathbf{e}_j \times \hat{\boldsymbol{\tau}} \quad \forall\, j \in \{1,2,3\} \tag{8.17}$$

we conclude that[13]

$$\dot{\mathbf{x}}_\omega(t) = \boldsymbol{\Omega}_\Delta \times \mathbf{x}_\omega(t)\,,$$

where:

$$\boxed{\begin{array}{lcll} \boldsymbol{\Omega}_\Delta & \stackrel{\text{def}}{=} & \Delta\,\mathbf{e}_3 - \Omega\,\mathbf{e}_1\,, & \\ \Delta & \stackrel{\text{def}}{=} & \omega_0 - \omega & \textbf{\textit{detuning}}\,. \end{array}}$$

Correspondingly we have

$$\boxed{\mathbf{x}_\omega(t) = \hat{D}_{-\vartheta\,\mathbf{e}_2}\,\hat{D}_{|\boldsymbol{\Omega}_\Delta|\,t\,\mathbf{e}_3}\,\hat{D}_{+\vartheta\,\mathbf{e}_2}\,\mathbf{x}_\omega(0)}$$

with

$$\boxed{\hat{D}_{\pm\vartheta\,\mathbf{e}_2} = \begin{pmatrix} \frac{\Delta}{|\boldsymbol{\Omega}_\Delta|} & 0 & \pm\frac{\Omega}{|\boldsymbol{\Omega}_\Delta|} \\ 0 & 1 & 0 \\ \mp\frac{\Omega}{|\boldsymbol{\Omega}_\Delta|} & 0 & \frac{\Delta}{|\boldsymbol{\Omega}_\Delta|} \end{pmatrix}}$$

and

$$\hat{D}_{\varphi\,\mathbf{e}_3} = \begin{pmatrix} \cos\varphi & -\sin\varphi & 0 \\ \sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad \forall\,\varphi \in \mathbb{R}\,.$$

Especially for

$$\mathbf{x}_\omega(0) = -\mathbf{e}_3\,,$$

therefore,

$$\mathbf{x}_\omega(t) = \begin{pmatrix} \frac{\Omega\,\Delta}{|\boldsymbol{\Omega}_\Delta|^2}\Big(1 - \cos\left(|\boldsymbol{\Omega}_\Delta|\,t\right)\Big) \\ -\frac{\Omega}{|\boldsymbol{\Omega}_\Delta|}\,\sin\left(|\boldsymbol{\Omega}_\Delta|\,t\right) \\ -\frac{\Delta^2 + \Omega^2\,\cos\left(|\boldsymbol{\Omega}_\Delta|\,t\right)}{|\boldsymbol{\Omega}_\Delta|^2} \end{pmatrix}.$$

**Remarks:**

1. We see that the population of $|\mathrm{g}\rangle$ and $|\mathrm{e}\rangle$ oscillates with angular frequency $|\boldsymbol{\Omega}_\Delta| = \sqrt{(\omega_0 - \omega)^2 + \Omega^2}$.
2. For $|\Delta| \gg \Omega$ these Rabi ***oscillations*** are negligible.

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[13]Thus:

$$\hat{\rho}_\omega(t) \text{ stationary} \quad \Longleftrightarrow \quad \mathbf{x}_\omega(t) \propto \boldsymbol{\Omega}_\Delta\,.$$

3. If $\mathbf{\Omega}_\Delta$ is 'slowly' changed with time and the initial angle between $\hat{\rho}_\omega(t)$ and $\mathbf{\Omega}_\Delta(t)$ is 'small' then this angle remains essentially constant.

4. This ***adiabatic*** following of $\mathbf{x}_\omega(t)$ may be used for specific state preparation:

   For instance, starting with $\mathbf{x}_\omega(t) = -\mathbf{e}_3$ and $\Delta \gg \Omega$ decrease $\omega$ until $-\Delta \gg \Omega$. Then $\mathbf{x}_\omega$ changes essentially into $+\mathbf{e}_3$, i.e. the atom's state is changed from $|\mathrm{g}\rangle$ essentially to $|\mathrm{e}\rangle$.

   The exact value of $\omega_0$ does not matter in this connection!

5. Of course:
$$\boxed{\mathbf{x}(t) = \hat{D}_{\omega\, t\, \mathbf{e}_3}\, \mathbf{x}_\omega(t)\,.}$$

6. Spontaneously emitted radiation — recognizable as fluorescence but not considered so far — could be taken into account to some extent, as far as $\hat{\rho}_\omega(t)$ is concerned by inserting phenomenological damping terms into the evolution equation.[14]

7. The Rabi oscillation are correlated with a modulation of the amplitude of the emitted light. Therefore side-bands show up at the angular frequencies[15] $\omega \pm \sqrt{(\omega_0 - \omega)^2 + \Omega^2}$.

## 8.3 3-Level Atoms

### 8.3.1 Population Trapping

Now assume that the atom is coupled to the classical field

$$\mathbf{E}_{\mathrm{ext}}(0,t) \approx \left(\boldsymbol{\mathcal{E}}_{\mathrm{p}}\, e^{-i\,\omega_{\mathrm{p}} t} + \boldsymbol{\mathcal{E}}_{\mathrm{c}}\, e^{-i\,\omega_{\mathrm{c}} t}\right) + c.c.$$

and that maximally only the three levels $|\mathrm{g}\rangle\,, |\mathrm{g}'\rangle\,, |\mathrm{e}\rangle$ are (essentially) occupied. Then, if

$$\begin{aligned} \langle \mathrm{g}|\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle \cdot \boldsymbol{\mathcal{E}}_{\mathrm{c}}^* = \langle \mathrm{g}'|\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle \cdot \boldsymbol{\mathcal{E}}_{\mathrm{p}}^* = 0\,, \\ \langle \mathrm{e}|\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle = \langle \mathrm{g}|\, \hat{\mathbf{x}}\, |\mathrm{g}\rangle = \langle \mathrm{g}'|\, \hat{\mathbf{x}}\, |\mathrm{g}'\rangle = \langle \mathrm{g}'|\, \hat{\mathbf{x}}\, |\mathrm{g}\rangle = 0\,, \end{aligned} \tag{8.18}$$

in dipole and rotation wave approximation the Liouville equation restricted to

$$\mathcal{H}^{(3)}_{\mathrm{atom}}(t) \stackrel{\mathrm{def}}{=} \mathcal{L}\Big(\{|\mathrm{g}\rangle\,, |\mathrm{e}\rangle\,, |\mathrm{g}'\rangle\}\Big)$$

is

$$i\hbar\, \frac{\mathrm{d}}{\mathrm{d}t}\, \hat{\rho}^{(3)}_{\mathrm{atom}}(t) = [\hat{H}^{(3)}_{\mathrm{cl}}(t), \hat{\rho}^{(3)}_{\mathrm{atom}}(t)]_-\,, \tag{8.19}$$



[14] See (Scully and Zubairy, 1999, 5.3) and 8.3.2.

[15] See (Mandel and Wolf, 1995, Eq. (15.3-22)) for the special case $\mathbf{x}(0) = -\mathbf{e}_3$.

where:[16]

$$\begin{aligned}
\hat{H}^{(3)}_{\rm cl}(t) &\stackrel{\rm def}{=} \hat{H}^{(3)}_{\rm atom} - \left(\frac{\hbar}{2}\,\Omega_{\rm p}\,e^{+i(\omega_{\rm p}\,t+\phi_{\rm p})}\,|{\rm g}\rangle\langle{\rm e}| + h.c.\right) \\
&\qquad - \left(\frac{\hbar}{2}\,\Omega_{\rm c}\,e^{+i(\omega_{\rm c}\,t+\phi_{\rm c})}\,|{\rm g}'\rangle\langle{\rm e}| + h.c.\right), \\
\Omega_{\rm p} &\stackrel{\rm def}{=} \frac{2\,q}{\hbar}\,\langle{\rm g}|\,\hat{\mathbf{x}}\,|{\rm e}\rangle\cdot\boldsymbol{\mathcal{E}}^*_{\rm p}\,e^{-i\phi_{\rm p}} \geq 0\,, \\
\Omega_{\rm c} &\stackrel{\rm def}{=} \frac{2\,q}{\hbar}\,\langle{\rm g}'|\,\hat{\mathbf{x}}\,|{\rm e}\rangle\cdot\boldsymbol{\mathcal{E}}^*_{\rm c}\,e^{-i\phi_{\rm c}} \geq 0\,.
\end{aligned} \tag{8.20}$$

With the definitions

$$\begin{aligned}
\hat{\rho}_{\rm rot}(t) &\stackrel{\rm def}{=} \hat{U}_{\rm rot}(t)\,\hat{\rho}^{(3)}_{\rm atom}(t)\,\hat{U}_{\rm rot}(-t)\,, \\
\hat{U}_{\rm rot}(t) &\stackrel{\rm def}{=} e^{-i\,(\omega_{\rm p}t+\phi_{\rm p})|{\rm g}\rangle\langle{\rm g}|}\,e^{-i\,(\omega_{\rm c}t+\phi_{\rm c})|{\rm g}'\rangle\langle{\rm g}'|}\,, \\
\hat{H}_{\rm rot}(t) &\stackrel{\rm def}{=} \hat{U}_{\rm rot}(t)\,\hat{H}^{(3)}_{\rm atom}\,\hat{U}_{\rm rot}(-t) + \hbar\,\omega_{\rm c}\,|{\rm g}'\rangle\langle{\rm g}'| + \hbar\,\omega_{\rm p}\,|{\rm g}\rangle\langle{\rm g}| - E_{\rm e}
\end{aligned}$$

this gives

$$i\hbar\,\frac{\rm d}{{\rm d}t}\,\hat{\rho}_{\rm rot}(t) = [\hat{H}_{\rm rot}(t), \hat{\rho}_{\rm rot}(t)]_-\,. \tag{8.21}$$

Using the Campbell-Hausdorff formula[17]

$$e^{\hat{A}}\,\hat{B}\,e^{-\hat{A}} = e^{{\rm ad}_{\hat{A}}}\,\hat{B}\,, \quad {\rm ad}_{\hat{A}}\left(\hat{B}\right) \stackrel{\rm def}{=} [\hat{A},\hat{B}]_- \quad \forall\,\hat{A},\hat{B}\,,$$

and

$$\Big[|{\rm g}\rangle\langle{\rm g}|\,,|{\rm g}\rangle\langle{\rm e}|\Big]_- = |{\rm g}\rangle\langle{\rm e}|\ , \quad \Big[|{\rm g}'\rangle\langle{\rm g}'|\,,|{\rm g}'\rangle\langle{\rm e}|\Big]_- = |{\rm g}'\rangle\langle{\rm e}|$$

we get

$$\begin{aligned}
\hat{H}_{\rm rot}(t) &= -\hbar\,\Delta_{\rm p}\,|{\rm g}\rangle\langle{\rm g}| - \hbar\,\Delta_{\rm c}\,|{\rm g}'\rangle\langle{\rm g}'| - \frac{\hbar}{2}\,\Omega_{\rm p}\Big(|{\rm g}\rangle\langle{\rm e}| + |{\rm e}\rangle\langle{\rm g}|\Big) \\
&\qquad - \frac{\hbar}{2}\,\Omega_{\rm c}\Big(|{\rm g}'\rangle\langle{\rm e}| + |{\rm e}\rangle\langle{\rm g}'|\Big),
\end{aligned} \tag{8.22}$$

where

$$\Delta_{\rm p} \stackrel{\rm def}{=} \frac{E_{\rm e}-E_{\rm g}}{\hbar} - \omega_{\rm p}\ , \quad \Delta_{\rm c} \stackrel{\rm def}{=} \frac{E_{\rm e}-E_{{\rm g}'}}{\hbar} - \omega_{\rm c}\,.$$

For the special case

$$\Delta_{\rm p} = \Delta_{\rm c} = \Delta$$

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[16] Note that

$$\hat{H}^{(3)}_{\rm atom} = E_{\rm e}\,|{\rm e}\rangle\langle{\rm e}| + E_{\rm g}\,|{\rm g}\rangle\langle{\rm g}| + E_{{\rm g}'}\,|{\rm g}'\rangle\langle{\rm g}'|\ .$$

[17] See Footnote 35 of Chapter 1.

this gives

$$\hat{H}_\Delta \stackrel{\text{def}}{=} \hat{H}_{\text{rot}}(t) + \Delta = -\frac{\hbar}{2}\begin{pmatrix} 0 & \Omega_\text{p} & 0 \\ \Omega_\text{p} & -2\Delta & \Omega_\text{c} \\ 0 & \Omega_\text{c} & 0 \end{pmatrix}$$

w.r.t. the basis $\left(|\text{g}\rangle\,, |\text{e}\rangle\,, |\text{g}'\rangle\right)$ and hence

$$\hat{H}_\Delta \begin{pmatrix} \Omega_\text{c} \\ 0 \\ -\Omega_\text{p} \end{pmatrix} = 0\,.$$

This implies that the ***dark state***

$$\hat{\rho}^{(3)}_{\text{atom}}(t) = |\psi_{\text{dark}}(t)\rangle\langle\psi_{\text{dark}}(t)|\;,$$

(not depending on $\Delta$), where

$$\boxed{\psi_{\text{dark}}(t) \stackrel{\text{def}}{=} \frac{1}{\sqrt{\Omega_\text{c}^2 + \Omega_\text{p}^2}}\left(\Omega_\text{c}\, e^{+i(\omega_\text{p}\,t+\phi_\text{p})}\,|\text{g}\rangle - \Omega_\text{p} e^{+i(\omega_\text{c}\,t+\phi_\text{c})}\,|\text{g}'\rangle\right)\;,}$$

is a solution of the LIOUVILLE equation (8.19) — which is also easy to check directly. Thanks to (8.18) the expectation value of the atom's electric dipole moment vanishes in this state. Hence the atom has no optical influence.

**Remarks:**

1. By suitable adiabatic variation of $\Omega_\text{p}, \Omega_\text{c}$ the population may be transferred completely from $|\text{g}\rangle$ to $|\text{g}'\rangle$.[18]
2. Extremely weak pump radiation (with angular frequency $\omega_\text{p}$) has to be treated quantum mechanically since in this case the spontaneously emitted portion of the total radiation with angular frequency $\omega_\text{p}$ can no longer be neglected.[19]

### 8.3.2 Electromagnetically Induced Transparency

Of course, (8.21) can be solved explicitly for $\hat{H}_{\text{rot}}(t)$ given by (8.22). For simplicity, however, let us assume strict resonance

$$\boxed{\Delta_\text{p} = \Delta_\text{c} = 0\,.} \tag{8.23}$$

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[18] See (Kuklinski et al., 1989). For the possible implementation of a universal set of quantum gates based on adiabatic population transfer see (Moller et al., 2007).

[19] See (Fleischhauer and Lukin, 2000) for this case. In (Kuang et al., 2003) both the pump and the coupling radiation are treated quantum mechanically.

Then we have

$$\begin{aligned}\hat{H}_{\mathrm{rot}}(t)\,|\mathrm{g}\rangle &= -\frac{\hbar}{2}\,\Omega_{\mathrm{p}}\,|\mathrm{e}\rangle\ ,\\ \hat{H}_{\mathrm{rot}}(t)\,|\mathrm{g}'\rangle &= -\frac{\hbar}{2}\,\Omega_{\mathrm{c}}\,|\mathrm{e}\rangle\ ,\\ \hat{H}_{\mathrm{rot}}(t)\,|\mathrm{e}\rangle &= -\frac{\hbar}{2}\Big(\Omega_{\mathrm{p}}\,|\mathrm{g}\rangle + \Omega_{\mathrm{c}}\,|\mathrm{g}'\rangle\Big)\end{aligned}$$

and hence

$$\begin{aligned}\hat{H}_{\mathrm{rot}}(t)\Big(\Omega_{\mathrm{c}}\,|\mathrm{g}\rangle - \Omega_{\mathrm{p}}\,|\mathrm{g}'\rangle\Big) &= 0\,, &(8.24)\\ \hat{H}_{\mathrm{rot}}(t)\left(|\mathrm{e}\rangle \pm \frac{\Omega_{\mathrm{p}}}{\Omega}\,|\mathrm{g}\rangle \pm \frac{\Omega_{\mathrm{c}}}{\Omega}\,|\mathrm{g}'\rangle\right) &= -\frac{\hbar}{2}\Big(\Omega_{\mathrm{p}}\,|\mathrm{g}\rangle + \Omega_{\mathrm{c}}\,|\mathrm{g}'\rangle\Big) \mp \frac{\hbar}{2}\left(\frac{\Omega_{\mathrm{p}}^2}{\Omega} + \frac{\Omega_{\mathrm{c}}^2}{\Omega}\right)|\mathrm{e}\rangle\\ &= \mp\frac{\hbar}{2}\,\Omega\left(|\mathrm{e}\rangle \pm \frac{\Omega_{\mathrm{p}}}{\Omega}\,|\mathrm{g}\rangle \pm \frac{\Omega_{\mathrm{c}}}{\Omega}\,|\mathrm{g}'\rangle\right)\ , &(8.25)\end{aligned}$$

where

$$\Omega \stackrel{\mathrm{def}}{=} \sqrt{\Omega_{\mathrm{p}}^2 + \Omega_{\mathrm{c}}^2}\,.$$

(8.24) corresponds to population trapping for the special case (8.23). (8.24) and (8.25) show that the normalized vectors

$$\begin{aligned}\psi_0 &\stackrel{\mathrm{def}}{=} \frac{1}{\Omega}\Big(\Omega_{\mathrm{c}}\,|\mathrm{g}\rangle - \Omega_{\mathrm{p}}\,|\mathrm{g}'\rangle\Big)\\ \psi_\pm &\stackrel{\mathrm{def}}{=} \frac{1}{\sqrt{2}}\left(|\mathrm{e}\rangle \pm \left(\frac{\Omega_{\mathrm{p}}}{\Omega}\,|\mathrm{g}\rangle + \frac{\Omega_{\mathrm{c}}}{\Omega}\,|\mathrm{g}'\rangle\right)\right)\end{aligned}$$

are eigenvectors of $\hat{H}_{\mathrm{rot}}(t)$ :

$$\begin{aligned}\hat{H}_{\mathrm{rot}}(t)\,\Psi_0 &= 0\,,\\ \hat{H}_{\mathrm{rot}}(t)\,\Psi_\pm &= \mp\frac{\hbar}{2}\,\Omega\,\psi_\pm\,.\end{aligned} \qquad (8.26)$$

Note that

$$\psi_+ - \psi_- = \frac{\sqrt{2}}{\Omega}\Big(\Omega_{\mathrm{p}}\,|\mathrm{g}\rangle + \Omega_{\mathrm{c}}\,|\mathrm{g}'\rangle\Big) \qquad (8.27)$$

and, therefore,

$$\begin{aligned}\psi_+ - \psi_- + \sqrt{2}\,\frac{\Omega_{\mathrm{c}}}{\Omega_{\mathrm{p}}}\,\psi_0 &= \left(\sqrt{2}\,\frac{\Omega_{\mathrm{p}}}{\Omega} + \sqrt{2}\,\frac{\Omega_{\mathrm{c}}^2}{\Omega\,\Omega_{\mathrm{p}}}\right)|\mathrm{g}\rangle\\ &= \sqrt{2}\,\frac{\Omega}{\Omega_{\mathrm{p}}}\,|\mathrm{g}\rangle\ .\end{aligned}$$

The latter implies[20]

$$
\begin{aligned}
& e^{-\frac{i}{\hbar}\hat H_{\rm rot}(t)\,t}\,|{\rm g}\rangle \\
&\underset{(8.26)}{=} \quad \frac{\Omega_{\rm c}}{\Omega}\,\psi_0 + \frac{1}{\sqrt{2}}\,\frac{\Omega_{\rm p}}{\Omega}\left(e^{+\frac{i}{2}\Omega t}\,\psi_+ - e^{-\frac{i}{2}\Omega t}\,\psi_-\right) \\
&= \quad \frac{\Omega_{\rm c}}{\Omega}\,\psi_0 + i\,\frac{\Omega_{\rm p}}{\Omega}\,\sin\left(\frac{\Omega}{2}\,t\right)|{\rm e}\rangle + \left(\frac{\Omega_{\rm p}}{\Omega}\right)^2\sin\left(\frac{\Omega}{2}\,t\right)|{\rm g}\rangle + \frac{\Omega_{\rm p}\,\Omega_{\rm c}}{\Omega^2}\cos\left(\frac{\Omega}{2}\,t\right)|{\rm g}'\rangle\,,
\end{aligned}
$$

i.e. the probability for finding the atom in the excited state at time $T$ is

$$
\left|\langle{\rm e}|\,e^{-\frac{i}{\hbar}\hat H_{\rm rot}(t)\,t}\,|{\rm g}\rangle\right|^2 = \left|\frac{\Omega_{\rm p}}{\Omega}\,\sin\left(\frac{\Omega}{2}\,t\right)\right|^2 \leq \left(\frac{\Omega_{\rm p}}{\Omega}\right)^2 \ll 1 \quad \text{for } \Omega_{\rm p} \ll \Omega
$$

if the state at time $t = 0$ is given by $|{\rm g}\rangle$ .

This explains the effect of ***electromagnetically induced transparency*** (EIT):

The coupling radiation $\widehat{=}\ \Omega_{\rm c}$ applied to the atom initially in the ground state $|{\rm g}\rangle$ essentially prohibits transitions into the excited state $|{\rm e}\rangle$ , if $\Omega_{\rm p} \ll \Omega$ , thus making the atom transparent for probe radiation $\widehat{=}\ \Omega_{\rm p}$ .

### 8.3.3 Change of Linear Susceptibility[21]

In order to calculate the susceptibility for the pump frequency, fading out of the transient perturbation — see Exercise 38a) of (Lücke, ein) in this context — has to be incorporated. A convenient heuristic way to achieve this is modifying the evolution equation into[22]

$$
i\hbar\,\frac{\rm d}{{\rm d}t}\,\hat\rho^{(3)}_{\rm atom}(t) = [\hat H^{(3)}_{\rm cl}(t), \hat\rho^{(3)}_{\rm atom}(t)]_- - i\,\frac{\hbar}{2}[\hat\Gamma, \hat\rho^{(3)}_{\rm atom}(t)]_+
$$

by means of a phenomenological ***relaxation matrix***

$$
\hat\Gamma = \underbrace{\gamma_{\rm g}}_{=0}\,|{\rm g}\rangle\langle{\rm g}| + \underbrace{\gamma_{\rm e}}_{>0}\,|{\rm e}\rangle\langle{\rm e}| + \underbrace{\gamma_{{\rm g}'}}_{\geq 0}\,|{\rm g}'\rangle\langle{\rm g}'|\ .
$$

For the interaction picture[23]

$$
\begin{aligned}
\hat\rho^{\rm I}(t) &\stackrel{\rm def}{=} e^{+\frac{i}{\hbar}\hat H^{(3)}_{\rm atom}\,t}\,\hat\rho^{(3)}_{\rm atom}(t)\,e^{-\frac{i}{\hbar}\hat H^{(3)}_{\rm atom}\,t}\,, \\
\hat V^{\rm I}(t) &\stackrel{\rm def}{=} e^{+\frac{i}{\hbar}\hat H^{(3)}_{\rm atom}\,t}\left(\hat H^{(3)}_{\rm cl}(t) - \hat H^{(3)}_{\rm atom}\right)e^{-\frac{i}{\hbar}\hat H^{(3)}_{\rm atom}\,t}
\end{aligned}
$$

Draft, November 5, 2011

[20] If a damping term is added to $\hat H_{\rm rot}(t)$ then $|{\rm g}\rangle$ will be driven into the corresponding dark state; see (Scully, 1994).

[21] We essentially follow the representation in (Scully and Zubairy, 1999, Sect. 7.3).

[22] Actually, one should also add $i\,\hbar\sqrt{\hat\Gamma}\,\hat\rho^{(3)}_{\rm atom}(t)\,\sqrt{\hat\Gamma}$ on the r.h.s. in order to get a Lindblad equation preserving the trace and positivity of $\hat\rho^{(3)}_{\rm atom}(t)$; more generally, see (Ottaviani et al., 2006). However, this would not effect the result for the linear susceptibility.

[23] See 6.1.2.

this implies

$$i\hbar \frac{\mathrm{d}}{\mathrm{d}t}\, \hat{\rho}^{\mathrm{I}}(t) = [\hat{V}^{\mathrm{I}}(t),\, \hat{\rho}^{\mathrm{I}}(t)]_{-} - i\,\frac{\hbar}{2}[\hat{\Gamma},\, \hat{\rho}^{\mathrm{I}}(t)]_{+} \tag{8.28}$$

with

$$\begin{aligned}\hat{V}^{\mathrm{I}}(t) \quad &= \quad -\left(\frac{\hbar}{2}\,\Omega_{\mathrm{p}}\, e^{-i(\Delta_{\mathrm{p}}t-\phi_{\mathrm{p}})}\, |\mathrm{g}\rangle\langle\mathrm{e}| + h.c.\right)\\ &\qquad -\left(\frac{\hbar}{2}\,\Omega_{\mathrm{c}}\, e^{-i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\, |\mathrm{g}'\rangle\langle\mathrm{e}| + h.c.\right)\,.\end{aligned}$$

In order to determine — for given coupling radiation — the linear atomic susceptibility

$$\begin{aligned}\overset{\leftrightarrow(1)}{\chi}_{\mathrm{atom}}(\omega_{\mathrm{p}})\,\boldsymbol{\mathcal{E}}_{\mathrm{p}} \quad &= \quad \lim_{T\to+\infty}\int_0^T \Omega_{\mathrm{p}}\,\frac{\partial}{\partial\Omega_{\mathrm{p}}}\,\langle\hat{\boldsymbol{\mu}}\rangle\,(t)\, e^{+i\,\omega_{\mathrm{p}}\,t}\,\mathrm{d}t\\ \hat{\boldsymbol{\mu}} \quad &\overset{\text{def}}{=} \quad q\,\hat{P}_{\mathcal{H}^{(3)}_{\mathrm{atom}}(t)}\hat{\mathbf{x}} \bigwedge \mathcal{H}^{(3)}_{\mathrm{atom}}(t)\end{aligned}$$

for the pump beam we develop the functions

$$\rho_{jk}(t) \overset{\text{def}}{=} \langle j|\;\hat{\rho}^{\mathrm{I}}(t)\,|k\rangle \quad \forall\, j,k\in\{\mathrm{g},\mathrm{e},\mathrm{g}'\}$$

into power series of $\Omega_{\mathrm{p}}$ :

$$\rho_{jk}(t,\Omega_{\mathrm{p}}) = \sum_{n=0}^{\infty}\rho^{[n]}_{jk}(t)\,(\Omega_{\mathrm{p}})^n\ .$$

Then we have

$$\begin{aligned}&\epsilon_0\,\overset{\leftrightarrow(1)}{\chi}_{\mathrm{atom}}(\omega_{\mathrm{p}})\,\boldsymbol{\mathcal{E}}_{\mathrm{p}}\\ &\quad= \lim_{T\to+\infty}\int_0^T \Omega_{\mathrm{p}}\,\mathrm{Tr}\Bigg(\underbrace{e^{-\frac{i}{\hbar}\,\hat{H}^{(3)}_{\mathrm{atom}}\,t}\sum_{j,k\in\{\mathrm{g},\mathrm{e},\mathrm{g}'\}}\rho^{[1]}_{jk}(t)\,|j\rangle\langle k|\;e^{+\frac{i}{\hbar}\,\hat{H}^{(3)}_{\mathrm{atom}}\,t}}_{=\left(\frac{\partial}{\partial\Omega_{\mathrm{p}}}\,\hat{\rho}^{\mathrm{I}}(t)\right)_{|\Omega_{\mathrm{p}}=0}}\,\hat{\boldsymbol{\mu}}\Bigg)\,e^{+i\,\omega_{\mathrm{p}}\,t}\,\mathrm{d}t\\ &\quad= \lim_{T\to+\infty}\int_0^T\sum_{j,k\in\{\mathrm{g},\mathrm{e},\mathrm{g}'\}}\rho^{[1]}_{jk}(t)\,e^{-\frac{i}{\hbar}(E_j-E_k)t}\,\langle k|\,q\,\hat{\mathbf{x}}\,|j\rangle\,\Omega_{\mathrm{p}}\,e^{+i\,\omega_{\mathrm{p}}\,t}\,\mathrm{d}t\\ &\underset{(8.18)}{=} \lim_{T\to+\infty}\int_0^T\left(\rho^{[1]}_{\mathrm{eg}}(t)\,e^{-\frac{i}{\hbar}(E_{\mathrm{e}}-E_{\mathrm{g}})t}\,\langle\mathrm{g}|\,q\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\,\Omega_{\mathrm{p}} + c.c.\right)e^{+i\,\omega_{\mathrm{p}}\,t}\,\mathrm{d}t\\ &\qquad + \lim_{T\to+\infty}\int_0^T\left(\rho^{[1]}_{\mathrm{eg}'}(t)\,e^{-\frac{i}{\hbar}\left(E_{\mathrm{e}}-E_{\mathrm{g}'}\right)t}\,\langle\mathrm{g}'|\,q\,\hat{\mathbf{x}}\,|\mathrm{e}\rangle\,\Omega_{\mathrm{p}} + c.c.\right)e^{+i\,\omega_{\mathrm{p}}\,t}\,\mathrm{d}t\,.\end{aligned} \tag{8.29}$$

With

$$
\begin{aligned}
\left[\hat{V}^{\mathrm{I}}(t), |\mathrm{g}\rangle\langle\mathrm{g}|\right]_- &= \frac{\hbar}{2}\,\Omega_{\mathrm{p}}\, e^{-i(\Delta_{\mathrm{p}}t-\phi_{\mathrm{p}})}\;|\mathrm{g}\rangle\langle\mathrm{e}| - h.c.\,,\\
\left[\hat{V}^{\mathrm{I}}(t), |\mathrm{g}'\rangle\langle\mathrm{g}'|\right]_- &= \frac{\hbar}{2}\,\Omega_{\mathrm{c}}\, e^{-i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\;|\mathrm{g}'\rangle\langle\mathrm{e}| - h.c.\,,\\
\left[\hat{V}^{\mathrm{I}}(t), |\mathrm{e}\rangle\langle\mathrm{e}|\right]_- &= -\left(\frac{\hbar}{2}\,\Omega_{\mathrm{p}}\, e^{-i(\Delta_{\mathrm{p}}t-\phi_{\mathrm{p}})}\;|\mathrm{g}\rangle\langle\mathrm{e}| - h.c.\right)\\
&\qquad\qquad -\left(\frac{\hbar}{2}\,\Omega_{\mathrm{c}}\, e^{-i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\;|\mathrm{g}'\rangle\langle\mathrm{e}| - h.c.\right)\,,\\
\left[\hat{V}^{\mathrm{I}}(t), |\mathrm{e}\rangle\langle\mathrm{g}|\right]_- &= \frac{\hbar}{2}\,\Omega_{\mathrm{p}}\, e^{-i(\Delta_{\mathrm{p}}t-\phi_{\mathrm{p}})}\Big(|\mathrm{e}\rangle\langle\mathrm{e}| - |\mathrm{g}\rangle\langle\mathrm{g}|\Big)\\
&\qquad\qquad - \frac{\hbar}{2}\,\Omega_{\mathrm{c}}\, e^{-i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\;|\mathrm{g}'\rangle\langle\mathrm{g}|\,,\\
\left[\hat{V}^{\mathrm{I}}(t), |\mathrm{e}\rangle\langle\mathrm{g}'|\right]_- &= \frac{\hbar}{2}\,\Omega_{\mathrm{c}}\, e^{-i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\Big(|\mathrm{e}\rangle\langle\mathrm{e}| - |\mathrm{g}'\rangle\langle\mathrm{g}'|\Big)\\
&\qquad\qquad - \frac{\hbar}{2}\,\Omega_{\mathrm{p}}\, e^{-i(\Delta_{\mathrm{p}}t-\phi_{\mathrm{p}})}\;|\mathrm{g}\rangle\langle\mathrm{g}'|\,,\\
\left[\hat{V}^{\mathrm{I}}(t), |\mathrm{g}\rangle\langle\mathrm{g}'|\right]_- &= -\frac{\hbar}{2}\,\Omega_{\mathrm{p}}\, e^{+i(\Delta_{\mathrm{p}}t-\phi_{\mathrm{p}})}\;|\mathrm{e}\rangle\langle\mathrm{g}'| + \frac{\hbar}{2}\,\Omega_{\mathrm{c}}\, e^{-i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\;|\mathrm{g}\rangle\langle\mathrm{e}|\,.
\end{aligned}
$$

and

$$\left[\hat{\Gamma}, |j\rangle\langle k|\right]_+ = (\gamma_j + \gamma_k)\,|j\rangle\langle k| \quad \forall\, j,k \in \{\mathrm{g},\mathrm{e},\mathrm{g}'\}$$

together with (8.28) we also conclude that

$$
\begin{aligned}
\dot{\rho}^{[0]}_{\mathrm{gg}}(t) &= 0\,,\\
\dot{\rho}^{[0]}_{\mathrm{ee}}(t) &= -\gamma_{\mathrm{e}}\,\rho^{[0]}_{\mathrm{ee}}(t)\,,\\
\dot{\rho}^{[0]}_{\mathrm{g}'\mathrm{g}'}(t) &= -\gamma_{\mathrm{g}'}\,\rho^{[0]}_{\mathrm{g}'\mathrm{g}'}(t)\,,\\
\dot{\rho}^{[0]}_{\mathrm{g}'\mathrm{e}}(t) &= -\frac{\gamma_{\mathrm{g}'}+\gamma_{\mathrm{e}}}{2}\,\rho^{[0]}_{\mathrm{g}'\mathrm{e}}(t)
\end{aligned}
\tag{8.30}
$$

and

$$
\begin{aligned}
\dot{\rho}^{[1]}_{\mathrm{eg}}(t) &= \frac{i}{2}\,e^{+i(\Delta_{\mathrm{p}}t-\phi_{\mathrm{p}})}\left(\rho^{[0]}_{\mathrm{gg}}(t) - \rho^{[0]}_{\mathrm{ee}}(t)\right)\\
&\quad + \frac{i}{2}\,\Omega_{\mathrm{c}}\,e^{+i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\rho^{[1]}_{\mathrm{g}'\mathrm{g}}(t) - \frac{\gamma_{\mathrm{e}}}{2}\rho^{[1]}_{\mathrm{eg}}(t)\,,\\
\dot{\rho}^{[1]}_{\mathrm{g}'\mathrm{g}}(t) &= \frac{i}{2}\,\Omega_{\mathrm{c}}\,e^{-i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\rho^{[1]}_{\mathrm{eg}}(t)\\
&\quad - \frac{i}{2}\,e^{+i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\,\rho^{[0]}_{\mathrm{g}'\mathrm{e}}(t) - \frac{\gamma_{\mathrm{g}'}}{2}\rho^{[1]}_{\mathrm{g}'\mathrm{g}}(t)\,,\\
\dot{\rho}^{[1]}_{\mathrm{eg}'}(t) &= \frac{i}{2}\,\Omega_{\mathrm{c}}\,e^{+i(\Delta_{\mathrm{c}}t-\phi_{\mathrm{c}})}\left(\rho^{[1]}_{\mathrm{g}'\mathrm{g}'}(t) - \rho^{[1]}_{\mathrm{ee}}(t)\right)\\
&\quad + \frac{i}{2}\,e^{+i(\Delta_{\mathrm{p}}t-\phi_{\mathrm{p}})}\,\rho^{[0]}_{\mathrm{gg}'}(t) - \frac{\gamma_{\mathrm{e}}+\gamma_{\mathrm{g}'}}{2}\,\rho^{[1]}_{\mathrm{eg}'}(t)\,.
\end{aligned}
\tag{8.31}
$$

For the special case

$$\boxed{\hat{\rho}^{\mathrm{I}}(t_0) = |\mathrm{g}\rangle\langle\mathrm{g}| \quad \forall\,\Omega_{\mathrm{p}}} \tag{8.32}$$

(8.30) implies

$$\rho^{[0]}_{\mathrm{gg}}(t) = 1\,, \quad \rho^{[0]}_{\mathrm{ee}}(t) = \rho^{[0]}_{\mathrm{g'g'}}(t) = \rho^{[0]}_{\mathrm{g'e}}(t) = 0$$

and hence, by (8.31),

$$\begin{aligned}
\dot{\rho}^{[1]}_{\mathrm{eg}}(t) &= \frac{i}{2}\, e^{+i(\Delta_{\mathrm{p}} t - \phi_{\mathrm{p}})} + \frac{i}{2}\,\Omega_{\mathrm{c}}\, e^{+i(\Delta_{\mathrm{c}} t - \phi_{\mathrm{c}})}\, \rho^{[1]}_{\mathrm{g'g}}(t) - \frac{\gamma_{\mathrm{e}}}{2} \rho^{[1]}_{\mathrm{eg}}(t)\,,\\
\dot{\rho}^{[1]}_{\mathrm{g'g}}(t) &= \frac{i}{2}\,\Omega_{\mathrm{c}}\, e^{-i(\Delta_{\mathrm{c}} t - \phi_{\mathrm{c}})} \rho^{[1]}_{\mathrm{eg}}(t) - \frac{\gamma_{\mathrm{g'}}}{2} \rho^{[1]}_{\mathrm{g'g}}(t)\,.
\end{aligned}$$

If, moreover, we assume

$$\boxed{\omega_{\mathrm{c}} = \omega_{\mathrm{eg'}}} \tag{8.33}$$

then me may conclude

$$\begin{aligned}
\dot{\mathbf{R}}(t) &= -\hat{M}\,\mathbf{R}(t) + \mathbf{A}\,,\\
\mathbf{R}(t_0) &= 0\,,
\end{aligned} \tag{8.34}$$

where:

$$\begin{aligned}
\mathbf{R}(t) &\stackrel{\text{def}}{=} \begin{pmatrix} e^{-i(\Delta_{\mathrm{p}} t - \phi_{\mathrm{p}})}\, \rho^{[1]}_{\mathrm{eg}}(t) \\ e^{-i(\Delta_{\mathrm{p}} t - \phi_{\mathrm{p}})}\, \rho^{[1]}_{\mathrm{g'g}}(t) \end{pmatrix},\\
\hat{M} &\stackrel{\text{def}}{=} \begin{pmatrix} \frac{\gamma_{\mathrm{e}}}{2} + i\Delta_{\mathrm{p}} & -\frac{i}{2}\,\Omega_{\mathrm{c}}\, e^{-i\,\phi_{\mathrm{c}}} \\ -\frac{i}{2}\,\Omega_{\mathrm{c}}\, e^{+i\,\phi_{\mathrm{c}}} & \frac{\gamma_{\mathrm{g'}}}{2} + i\Delta_{\mathrm{p}} \end{pmatrix},\\
\mathbf{A} &\stackrel{\text{def}}{=} \begin{pmatrix} \frac{i}{2} \\ 0 \end{pmatrix}.
\end{aligned}$$

The solution of the initial value problem (8.34) is

$$\begin{aligned}
\mathbf{R}(t) &= \hat{M}^{-1}\mathbf{A} - e^{-\hat{M}(t-t_0)}\, \hat{M}^{-1}\,\mathbf{A}\\
&\xrightarrow[t_0\to-\infty]{} \hat{M}^{-1}\mathbf{A}\,.
\end{aligned} \tag{8.35}$$

**Sketch of proof for (8.35):** Since

$$\det\Bigl(\hat{M} - i\Delta_{\mathrm{p}} - E\Bigr) = \frac{1}{4}\,(\gamma_{\mathrm{e}} - E)(\gamma_{\mathrm{g'}} - E) + (\Omega_{\mathrm{c}}/2)^2\,,$$

$\hat{M} - i\Delta_{\mathrm{p}}$ has only eigenvalues $E$ with $\Re(E) = \frac{1}{2}\,(\gamma_{\mathrm{e}} + \gamma_{\mathrm{g'}}) > 0$. ∎

Therefore,[24] assuming (8.32) and (8.33), asymptotically (for large $t$) we have

$$\rho_{\mathrm{eg}}^{[1]}(t) = \frac{i}{2}\, \frac{e^{+i(\Delta_{\mathrm{p}} t - \phi_{\mathrm{p}})}\left(\frac{\gamma_{\mathrm{g}'}}{2} + i\Delta_{\mathrm{p}}\right)}{\left(\frac{\gamma_{\mathrm{e}}}{2} + i\Delta_{\mathrm{p}}\right)\left(\frac{\gamma_{\mathrm{g}'}}{2} + i\Delta_{\mathrm{p}}\right) + (\Omega_{\mathrm{c}}/2)^2}\,.$$

For

$$\frac{E_{\mathrm{e}} - E_{\mathrm{g}}}{\hbar} > 0 < \frac{E_{\mathrm{e}} - E_{\mathrm{g}'}}{\hbar} \neq \omega_{\mathrm{p}} > 0$$

this, together with

$$\Omega_{\mathrm{p}} \underset{(8.20)}{=} \frac{2\,q}{\hbar}\, \langle \mathrm{e}|\, \hat{\mathbf{x}}\, |\mathrm{g}\rangle \cdot \boldsymbol{\mathcal{E}}_{\mathrm{p}}\, e^{+i\phi_{\mathrm{p}}}$$

and (8.29), implies[25]

$$\begin{aligned}
\epsilon_0\, \overset{\leftrightarrow}{\chi}{}^{(1)}_{\mathrm{atom}}(\omega_{\mathrm{p}})\, \boldsymbol{\mathcal{E}}_{\mathrm{p}} &= \rho_{\mathrm{eg}}^{[1]}(t)\, e^{i(\omega_{\mathrm{p}} - \omega_{\mathrm{eg}})t}\, \langle \mathrm{g}|\, q\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle\, \Omega_{\mathrm{p}} \\
&= \frac{i}{\hbar}\, \frac{\langle \mathrm{g}|\, q\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle \left(\langle \mathrm{e}|\, q\, \hat{\mathbf{x}}\, |\mathrm{g}\rangle \cdot \boldsymbol{\mathcal{E}}_{\mathrm{p}}\right)\left(\frac{\gamma_{\mathrm{g}'}}{2} + i\Delta_{\mathrm{p}}\right)}{\left(\frac{\gamma_{\mathrm{e}}}{2} + i\Delta_{\mathrm{p}}\right)\left(\frac{\gamma_{\mathrm{g}'}}{2} + i\Delta_{\mathrm{p}}\right) + (\Omega_{\mathrm{c}}/2)^2}\,.
\end{aligned}$$

Thus, especially for

$$\boldsymbol{\mathcal{E}}_{\mathrm{p}} \propto \langle \mathrm{e}|\, \hat{\mathbf{x}}\, |\mathrm{g}\rangle$$

we have

$$\overset{\leftrightarrow}{\chi}{}^{(1)}_{\mathrm{atom}}(\omega_{\mathrm{p}})\, \boldsymbol{\mathcal{E}}_{\mathrm{p}} = \chi_{\mathrm{atom}}(\omega_{\mathrm{p}})\, \boldsymbol{\mathcal{E}}_{\mathrm{p}}\,,$$

where

$$\chi_{\mathrm{atom}}(\omega_{\mathrm{p}}) \overset{\mathrm{def}}{=} \frac{i}{\epsilon_0\, \hbar}\, \frac{\left|\langle \mathrm{g}|\, q\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle\right|^2 \left(\frac{\gamma_{\mathrm{g}'}}{2} + i\Delta_{\mathrm{p}}\right)}{\left(\frac{\gamma_{\mathrm{e}}}{2} + i\Delta_{\mathrm{p}}\right)\left(\frac{\gamma_{\mathrm{g}'}}{2} + i\Delta_{\mathrm{p}}\right) + (\Omega_{\mathrm{c}}/2)^2}\,.$$

Together with

$$\begin{aligned}
&\frac{1}{\left(\frac{\gamma_{\mathrm{e}}}{2} + i\Delta_{\mathrm{p}}\right)\left(\frac{\gamma_{\mathrm{g}'}}{2} + i\Delta_{\mathrm{p}}\right) + (\Omega_{\mathrm{c}}/2)^2} \\
&= \frac{\frac{\gamma_{\mathrm{e}}}{2}\frac{\gamma_{\mathrm{g}'}}{2} - (\Delta_{\mathrm{p}})^2 + (\Omega_{\mathrm{c}}/2)^2 - i\left(\frac{\gamma_{\mathrm{e}}}{2} + \frac{\gamma_{\mathrm{g}'}}{2}\right)\Delta_{\mathrm{p}}}{\left|\frac{\gamma_{\mathrm{e}}}{2}\frac{\gamma_{\mathrm{g}'}}{2} - (\Delta_{\mathrm{p}})^2 + (\Omega_{\mathrm{c}}/2)^2 + i\left(\frac{\gamma_{\mathrm{e}}}{2} + \frac{\gamma_{\mathrm{g}'}}{2}\right)\Delta_{\mathrm{p}}\right|^2} \\
&= \frac{\frac{\gamma_{\mathrm{e}}}{2}\frac{\gamma_{\mathrm{g}'}}{2} - (\Delta_{\mathrm{p}})^2 + (\Omega_{\mathrm{c}}/2)^2 - i\left(\frac{\gamma_{\mathrm{e}}}{2} + \frac{\gamma_{\mathrm{g}'}}{2}\right)\Delta_{\mathrm{p}}}{\left(\frac{\gamma_{\mathrm{e}}}{2}\frac{\gamma_{\mathrm{g}'}}{2} - (\Delta_{\mathrm{p}})^2 + (\Omega_{\mathrm{c}}/2)^2\right)^2 + \left(\frac{\gamma_{\mathrm{e}}}{2} + \frac{\gamma_{\mathrm{g}'}}{2}\right)^2 (\Delta_{\mathrm{p}})^2}
\end{aligned}$$

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[24]Note that

$$a\,d - b\,c \neq 0 \quad\Longrightarrow\quad \begin{pmatrix} a & b \\ c & d \end{pmatrix}^{-1} = \begin{pmatrix} +d & -c \\ -b & +a \end{pmatrix} \Big/ (a\,d - b\,c) \qquad \forall\, a, b, c, d \in \mathbb{C}\,.$$

[25]We do not show explicitly that the second contribution on the r.h.s. of (8.29) vanishes.

this, finally, gives:

$$\boxed{\begin{aligned}\Re\Big(\chi_{\mathrm{atom}}(\omega_{\mathrm{p}})\Big) &= \frac{|\langle \mathrm{g}|\, q\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle|^2}{\epsilon_0\, \hbar}\, \frac{\Delta_{\mathrm{p}}\left(\left(\frac{\gamma_{\mathrm{g}'}}{2}\right)^2 + (\Delta_{\mathrm{p}})^2 - (\Omega_{\mathrm{c}}/2)^2\right)}{\left(\frac{\gamma_{\mathrm{e}}}{2}\frac{\gamma_{\mathrm{g}'}}{2} - (\Delta_{\mathrm{p}})^2 + (\Omega_{\mathrm{c}}/2)^2\right)^2 + \left(\frac{\gamma_{\mathrm{e}}}{2} + \frac{\gamma_{\mathrm{g}'}}{2}\right)^2 (\Delta_{\mathrm{p}})^2}\,,\\ \Im\Big(\chi_{\mathrm{atom}}(\omega_{\mathrm{p}})\Big) &= \frac{|\langle \mathrm{g}|\, q\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle|^2}{\epsilon_0\, \hbar}\, \frac{(\Delta_{\mathrm{p}})^2\, \frac{\gamma_{\mathrm{e}}}{2} + \frac{\gamma_{\mathrm{g}'}}{2}\left(\frac{\gamma_{\mathrm{e}}}{2}\frac{\gamma_{\mathrm{g}'}}{2} + (\Omega_{\mathrm{c}}/2)^2\right)}{\left(\frac{\gamma_{\mathrm{e}}}{2}\frac{\gamma_{\mathrm{g}'}}{2} - (\Delta_{\mathrm{p}})^2 + (\Omega_{\mathrm{c}}/2)^2\right)^2 + \left(\frac{\gamma_{\mathrm{e}}}{2} + \frac{\gamma_{\mathrm{g}'}}{2}\right)^2 (\Delta_{\mathrm{p}})^2}\,.\end{aligned}}$$

Defining

$$\begin{aligned}\xi(\lambda) &\overset{\mathrm{def}}{=} \frac{\lambda\,(\lambda^2-1)}{(\lambda^2-1)^2 + \left(\frac{2\,\gamma_{\mathrm{e}}}{\Omega_{\mathrm{c}}}\right)^2 \lambda^2}\,,\\ \eta(\lambda) &\overset{\mathrm{def}}{=} \frac{\lambda^2}{(\lambda^2-1)^2 + \left(\frac{2\,\gamma_{\mathrm{e}}}{\Omega_{\mathrm{c}}}\right)^2 \lambda^2}\\ &= \frac{1}{(\lambda - 1/\lambda)^2 + \left(\frac{2\,\gamma_{\mathrm{e}}}{\Omega_{\mathrm{c}}}\right)^2} \quad \text{für } \lambda \neq 0\end{aligned}$$

we get

$$\gamma_{g'} = 0 < \Omega_{\mathrm{c}} \quad\Longrightarrow\quad \begin{cases}\Re\Big(\chi_{\mathrm{atom}}(\omega_{\mathrm{p}})\Big) = \dfrac{2\,|\langle \mathrm{g}|\, q\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle|^2}{\epsilon_0\, \hbar\, \Omega_{\mathrm{c}}}\, \xi\!\left(\dfrac{2\,\Delta_{\mathrm{p}}}{\Omega_{\mathrm{c}}}\right)\,,\\ \Im\Big(\chi_{\mathrm{atom}}(\omega_{\mathrm{p}})\Big) = \dfrac{2\,|\langle \mathrm{g}|\, q\, \hat{\mathbf{x}}\, |\mathrm{e}\rangle|^2}{\epsilon_0\, \hbar\, \Omega_{\mathrm{c}}}\, \dfrac{\gamma_{\mathrm{e}}}{\Omega_{\mathrm{c}}}\, \eta\!\left(\dfrac{2\,\Delta_{\mathrm{p}}}{\Omega_{\mathrm{c}}}\right)\,,\end{cases}$$

$$\max_{0\neq\lambda\in\mathbb{R}} \eta(\lambda) = \lambda(1)\,.$$

For the special case $\Omega_{\mathrm{c}} = 2\,\gamma_{\mathrm{e}}$ the graphs of the functions $\xi(\lambda)$ and $\eta(\lambda)$ are sketched in Figure 8.1.

Quite generally the complex refractive index of an isotropic medium with linear susceptibility $\chi(\omega_{\mathrm{p}})$ is characterized by

$$\mathcal{N}(\omega_{\mathrm{p}}) \overset{\mathrm{def}}{=} \sqrt{1+\chi(\omega_{\mathrm{p}})} \in \mathbb{R}_+ + i\,\mathbb{R}\,.$$

Because of

$$\left(\sqrt[+]{\frac{|z| + \Re(z)}{2}} + i\,\sqrt[+]{\frac{|z| - \Re(z)}{2}}\,\operatorname{sign}\Big(\Im(z)\Big)\right)^2 = z \quad \forall\, z \in \mathbb{C}\setminus\mathbb{R}\,.$$

the latter is equivalent to

$$\begin{aligned}\Re\Big(\mathcal{N}(\omega_{\mathrm{p}})\Big) &= \sqrt[+]{\frac{|1+\chi(\omega_{\mathrm{p}})| + \Re\Big(1+\chi(\omega_{\mathrm{p}})\Big)}{2}}\,,\\ \Im\Big(\mathcal{N}(\omega_{\mathrm{p}})\Big) &= \sqrt[+]{\frac{|1+\chi(\omega_{\mathrm{p}})| - \Re\Big(1+\chi(\omega_{\mathrm{p}})\Big)}{2}}\,\operatorname{sign}\Big(\Im\Big(\chi(\omega_{\mathrm{p}})\Big)\Big)\,.\end{aligned}$$

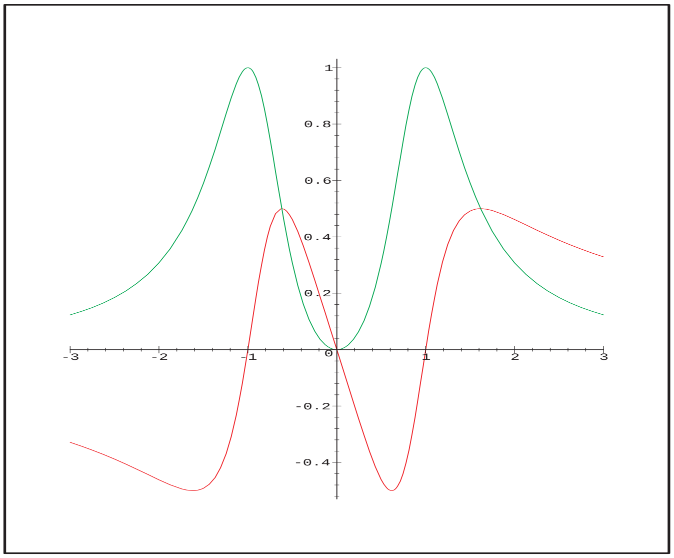


Figure 8.1: Graphs of the functions $\xi(\lambda)$ (red,below) and $\eta(\lambda)$ (green,above).

# Chapter 9

# Cavity QED

The general idea for modeling the coupled system of an atom and radiation in a high $Q$ cavity is the following:

- There are only a few resonant modes dominating the radiation in the cavity. Therefore, in a reasonable approximate description, all other modes of the quantized radiation field may be disregarded.
- There are only a few levels of the atom involved in the interaction with the cavity field. Therefore, in a reasonable approximate description, the internal state space of the atom subsystem may be restricted to describe just these levels.

## 9.1 The Jaynes-Cummings Model

In the simplest model of cavity QED the radiation field is restricted to a single, essentially monochromatic mode $\hat{a}$ and the atom is described as a two-level quantum system at some fixed position in the cavity.

### 9.1.1 Quantized Radiation Field for Selected Modes

Let $\mathbf{a}_1, \mathbf{a}_2, \ldots$ be a maximal family of pairwise orthogonal modes. Then, by (1.37),

$$f^j_\nu(\mathbf{k}) = [\hat{a}_j(\mathbf{k}), \hat{a}^\dagger_\nu]_- \quad \forall\, j \in \{1,2\}\ ,\ \nu \in \mathbb{R}\,,\ \mathbf{k} \in \mathbb{R}^3\,. \tag{9.1}$$

(1.61) together with (1.33) implies

$$\begin{aligned} \hat{\mathbf{E}}^{(+)}_{\hat{a}_1,\ldots,\hat{a}_n}(\mathbf{x}) &\stackrel{\text{def}}{=} \hat{P}_{\mathcal{H}_{\hat{a}_1,\ldots,\hat{a}_n}}\, \hat{\mathbf{E}}^{(+)}_0(\mathbf{x},0)\, \hat{P}_{\mathcal{H}_{\hat{a}_1,\ldots,\hat{a}_n}} \\ &= \sum_{\nu=1}^{n} \boldsymbol{\mathcal{E}}_{\hat{a}_\nu}(\mathbf{x})\, \hat{a}_\nu \quad \forall\, n \in \mathbb{N}\,, \end{aligned} \tag{9.2}$$

where

$$\boldsymbol{\mathcal{E}}_{\hat{a}_\nu}(\mathbf{x}) \stackrel{\text{def}}{=} -i\,(2\pi)^{-\frac{3}{2}}\,\sqrt{\mu_0\hbar c}\sum_{j=1}^{2}\int c\,|\mathbf{k}|\,\boldsymbol{\epsilon}_j(\mathbf{k})\,[\hat{a}_j(\mathbf{k}),\hat{a}_\nu^\dagger]_-\,e^{i\,\mathbf{k}\cdot\mathbf{x}}\,\frac{\mathrm{d}V_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}\,. \tag{9.3}$$

(1.61) together with (1.53) implies

$$\begin{aligned}\hat{H}_{\hat{a}_1,\ldots,\hat{a}_n} &\stackrel{\text{def}}{=} \hat{P}_{\mathcal{H}_{\hat{a}_1,\ldots,\hat{a}_n}}\,\hat{H}_{\text{field}}\,\hat{P}_{\mathcal{H}_{\hat{a}_1,\ldots,\hat{a}_n}}\\ &= \sum_{\nu,\mu=1}^{n} h_{\nu\mu}\,\hat{a}_\nu^\dagger\,\hat{a}_\mu \quad \forall\, n\in\mathbb{N}\,,\end{aligned} \tag{9.4}$$

where

$$\begin{aligned}h_{\nu\mu} &\stackrel{\text{def}}{=} \sum_{j=1}^{2}\int \hbar\,c\,|\mathbf{k}|\,\left(f_\nu^j(\mathbf{k})\right)^*\,f_\mu^j(\mathbf{k})\,\mathrm{d}V_{\mathbf{k}}\\ &= \left(h_{\nu'\nu}\right)^* \quad \forall\,\nu,\mu\in\mathbb{N}\,.\end{aligned} \tag{9.5}$$

(9.4) simplifies to

$$\hat{H}_{\hat{a}_1,\ldots,\hat{a}_n} = \sum_{\nu=1}^{n} h_{\nu\nu}\,\hat{a}_\nu^\dagger\,\hat{a}_\nu$$

if the modes $\hat{a}_\nu$ are ***non-overlapping***, i.e. if

$$\nu\neq\mu \implies f_\nu^j(\mathbf{k})\,f_\mu^j(\mathbf{k}) = 0 \quad \forall\, j\in\{1,2\}\,.$$

> **Remark:** Note that restriction to the non-overlapping modes $\hat{a}_1,\ldots,\hat{a}_n$ can only be consistent if, e.g., the $\hat{a}_1^\dagger\,\Omega,\ldots,\hat{a}_n^\dagger\,\Omega$ have (sufficiently) sharp energy. Actually for cavity modes this is only possible due to a change of boundary conditions resulting in a change of the dynamics. Thus restricting the Hamiltonian of the radiation field to some non-overlapping modes is much more drastic than restricting the Hamiltonian of an atom to a few levels.

### 9.1.2 Interaction of the Two-Level System with the Cavity Field

Let the atom be localized at $\mathbf{x}=0$. Then the electric field representation discussed in 7.1.2 suggests using[1]

$$\begin{aligned}\hat{H}_{\text{cav}} &\stackrel{\text{def}}{=} \left(\hat{P}_{\mathcal{H}_m}\otimes\hat{P}_{\mathcal{H}_{\hat{a}}}\right)\left(\hat{H}_{\text{atom}}+\hat{H}_{\text{field}}-q\,\hat{\mathbf{x}}\cdot\hat{\mathbf{E}}_0(0,0)\right)\left(\hat{P}_{\mathcal{H}_m}\otimes\hat{P}_{\mathcal{H}_{\hat{a}}}\right)\\ &= \hat{H}_m+\hat{H}_{\hat{a}}-q\,\hat{\mathbf{x}}_m\cdot\hat{\mathbf{E}}_{\hat{a}}\,,\end{aligned} \tag{9.6}$$



[1] Recall the agreement made in 7.1.1.

where

$$\hat{\mathbf{E}}_{\hat{a}} \stackrel{\text{def}}{=} \hat{\mathbf{E}}^{(+)}_{\hat{a}}(0) + \left(\hat{\mathbf{E}}^{(+)}_{\hat{a}}(0)\right)^{\dagger} \;, \quad \hat{\mathbf{x}}_{\mathrm{m}} \stackrel{\text{def}}{=} \hat{P}_{\mathcal{H}_m}\, \hat{\mathbf{x}}\, \hat{P}_{\mathcal{H}_m} \,, \tag{9.7}$$

as an approximate Hamiltonian for interacting system of the two-level atom and the quantized single-mode radiation field.

> **Remark:** Note that in the electric field representation $\hat{\mathbf{p}}_{\text{can}}$ is to be interpreted as the (approximate) observable of the mechanical — rather than canonical — momentum of the atom's electron! Correspondingly, $\mathcal{H}_{\text{atom}}$ is to be interpreted as the (approximate) observable of the internal energy of the one-electron atom.

Assuming[2]

$$\boxed{\langle \Psi_\nu \mid \hat{\mathbf{x}}\, \Psi_\nu \rangle = 0 \quad \text{for } \nu = 1, 2} \tag{9.8}$$

and defining

$$\boldsymbol{\mu}_{01} \stackrel{\text{def}}{=} q\, \langle \Psi_0 \mid \hat{\mathbf{x}}\, \Psi_1 \rangle \tag{9.9}$$

we get

$$q\, \hat{\mathbf{x}}_m = \boldsymbol{\mu}_{01}\, \hat{b} + \boldsymbol{\mu}^*_{01}\, \hat{b}^\dagger \tag{9.10}$$

for the (restricted) atomic dipole operator in the matrix representation. This is a simple consequence of (9.7), (9.2), and (8.11). Moreover, defining

$$\boldsymbol{\mathcal{E}} \stackrel{\text{def}}{=} -i\,(2\pi)^{-\frac{3}{2}} \sqrt{\mu_0 \hbar c} \sum_{j=1}^{2} \int c\, |\mathbf{k}|\, \boldsymbol{\epsilon}_j(\mathbf{k})\, [\hat{a}_j(\mathbf{k}), \hat{a}^\dagger_\nu]_- \frac{\mathrm{d}V_{\mathbf{k}}}{\sqrt{2\,|\mathbf{k}|}}$$

we get

$$\hat{\mathbf{E}}_{\hat{a}} = \boldsymbol{\mathcal{E}}\, \hat{a} + \boldsymbol{\mathcal{E}}^* \hat{a}^\dagger \tag{9.11}$$

for the restricted electric field operator as a direct consequence of (9.7), (9.2), and (9.3).

Let us consider the **special** case[3]

$$\boldsymbol{\mu}_{01} = \mu_{\mathrm{m}}\, \frac{\mathbf{e}_1 + i\, \mathbf{e}_2}{\sqrt{2}} \;, \quad \boldsymbol{\mathcal{E}} = \mathcal{E}_0\, \frac{\mathbf{e}_1 + i\, \mathbf{e}_2}{\sqrt{2}} \,. \tag{9.12}$$

Then, up to an irrelevant additive constant, $\hat{H}_{\text{cav}}$ coincides with the JAYNES-CUMMINGS ***Hamiltonian***

$$\hat{H}_{\text{JC}} \stackrel{\text{def}}{=} \hbar\, \omega_0\, \hat{b}^\dagger \hat{b} + \hbar\, \omega\, \hat{a}^\dagger \hat{a} + \lambda\, \hat{a}^\dagger \hat{b} + \lambda^*\, \hat{a}\, \hat{b}^\dagger \,, \tag{9.13}$$

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[2]This assumption is usually fulfilled for the stationary states $\Psi_0, \Psi_1$ of the atom.

[3]The first condition corresponds to $\Psi_1 \to \Psi_0$ being a $\Delta m = \pm 1$-transition. If the conditions (9.12) are not fulfilled (9.13) is called ***rotating-wave approximation***; see, e.g., (Schleich, 2001, Section 14.8.2).

where

$$\omega \stackrel{\text{def}}{=} \hbar^{-1} \sum_{j,j'=1}^{2} \int c\,|\mathbf{k}|\,[\hat{a},\hat{a}_j^\dagger(\mathbf{k})]_-\,[\hat{a}_{j'}(\mathbf{k}),\hat{a}^\dagger]_-\,\mathrm{d}V_{\mathbf{k}}\,, \quad \lambda \stackrel{\text{def}}{=} \mu_{\mathrm{m}}\,\mathcal{E}_0\,.$$

**Proof:** Thanks to

$$(\mathbf{e}_1 + i\,\mathbf{e}_2)^2 = 0\,, \quad (\mathbf{e}_1 + i\,\mathbf{e}_2)\,(\mathbf{e}_1 - i\,\mathbf{e}_2) = 2$$

we have

$$\begin{aligned} -\,q\,\hat{\mathbf{x}}_m \cdot \hat{\mathbf{E}}_{\hat{a}} \underset{(9.10)-(9.12)}{=} & \left(\mu_{\mathrm{m}}\,\frac{\mathbf{e}_1 + i\,\mathbf{e}_2}{\sqrt{2}}\,\hat{b} + \mu_{\mathrm{m}}^*\,\frac{\mathbf{e}_1 - i\,\mathbf{e}_2}{\sqrt{2}}\,\hat{b}^\dagger\right) \\ & \cdot \left(\mathcal{E}_0\,\frac{\mathbf{e}_1 + i\,\mathbf{e}_2}{\sqrt{2}}\,\hat{a} + \mathcal{E}_0^*\,\frac{\mathbf{e}_1 - i\,\mathbf{e}_2}{\sqrt{2}}\,\hat{a}^\dagger\right) \\ = & \ \lambda\,\hat{a}^\dagger\hat{b} + \lambda^*\,\hat{a}\,\hat{b}^\dagger\,. \end{aligned} \tag{9.14}$$

(9.2), (9.3), and (9.1) imply

$$\hat{H}_{\hat{a}} = \hbar\,\omega\,\hat{a}^\dagger\hat{a}\,. \tag{9.15}$$

(8.4) and (8.9), finally, imply

$$\hat{H}_m - \hbar\,\omega_0\,\hat{b}^\dagger\hat{b} \propto \hat{1}\,, \tag{9.16}$$

Now, the statement is a direct consequence of (9.14)–(9.16). ∎

Without loss of generality, we assume that[4]

$$\boxed{\lambda = \lambda^*\,.}$$

With

$$\mathcal{H}^{(0)} \stackrel{\text{def}}{=} \{\alpha\,|0,0\rangle\,:\,\alpha \in \mathbb{C}\}$$

and

$$\mathcal{H}^{(n+1)} \stackrel{\text{def}}{=} \left\{ \begin{pmatrix} \alpha_1 \\ \alpha_0 \end{pmatrix}_n \stackrel{\text{def}}{=} \alpha_0\,|0,n+1\rangle + \alpha_1\,|1,n\rangle\,:\,\alpha_0,\alpha_1 \in \mathbb{C} \right\} \quad \forall\, n \in \mathbb{Z}_+\,,$$

where

$$|j,n\rangle \stackrel{\text{def}}{=} \Psi_j \otimes \frac{1}{\sqrt{n!}}\left(\hat{a}^\dagger\right)^n \Omega \quad \forall\, j \in \{0,1\}\,,\, n \in \mathbb{Z}_+\,.$$

we have

$$\mathcal{H}_m \otimes \mathcal{H}_{\hat{a}} = \bigoplus_{n=0}^{\infty} \mathcal{H}^{(n)}$$

and, obviously, $\hat{H}_{\mathrm{JC}}$ leaves all the $\mathcal{H}^{(n)}$ invariant. For every $n \in \mathbb{Z}_+$, since

$$\hat{a}^\dagger\,|j,n\rangle = \sqrt{n+1}\;|j,n+1\rangle\,, \quad \hat{a}\,|j,n+1\rangle = \sqrt{n+1}\;|j,n\rangle\,,$$



[4] Otherwise we could change $\hat{a}$ by a phase factor to arrange this.

we have

$$\hat{H}_{\mathrm{JC}} \begin{pmatrix} \alpha_1 \\ \alpha_0 \end{pmatrix}_n = \hat{H}_{\mathrm{JC}}^{(n+1)} \begin{pmatrix} \alpha_1 \\ \alpha_0 \end{pmatrix}_n \quad \forall\, \alpha_0, \alpha_1 \in \mathbb{C}$$

with

$$\begin{aligned} \hat{H}_{\mathrm{JC}}^{(n+1)} &\stackrel{\text{def}}{=} \begin{pmatrix} \hbar\,\omega_0 + n\,\hbar\,\omega & \lambda\,\sqrt{n+1} \\ \lambda\sqrt{n+1} & (n+1)\,\hbar\,\omega \end{pmatrix} \\ &= \check{E}_n\,\hat{\tau}^0 + \lambda\sqrt{n+1}\,\hat{\tau}^1 + \hbar\,\frac{\delta}{2}\,\hat{\tau}^3 \end{aligned} \tag{9.17}$$

and

$$\check{E}_n \stackrel{\text{def}}{=} \hbar\left(\omega_0 + (n+1/2)\,\omega\right), \quad \delta \stackrel{\text{def}}{=} \omega_0 - \omega \quad (\textbf{\textit{detuning}})\,. \tag{9.18}$$

In order to exploit (9.17), note that[5]

$$e^{i\,\boldsymbol{\varphi}\cdot\hat{\boldsymbol{\tau}}}\,\hat{\boldsymbol{\tau}}\,e^{-i\,\boldsymbol{\varphi}\cdot\hat{\boldsymbol{\tau}}} = \hat{D}_{2\boldsymbol{\varphi}}\,\hat{\boldsymbol{\tau}} \quad \forall\,\boldsymbol{\varphi} \in \mathbb{R}^3\,. \tag{9.19}$$

**Sketch of proof:** A straightforward calculation using (8.8) shows that

$$\hat{\mathbf{f}}(\varphi) = \hat{\mathbf{f}}_1(\varphi) \stackrel{\text{def}}{=} e^{i\,\varphi\,\mathbf{e}\cdot\hat{\boldsymbol{\tau}}}\,\hat{\boldsymbol{\tau}}\,e^{-i\,\varphi\,\mathbf{e}\cdot\hat{\boldsymbol{\tau}}}$$

is a solution of the initial value problem

$$\frac{\mathrm{d}}{\mathrm{d}\varphi}\,\hat{\mathbf{f}}(\varphi) = 2\,\mathbf{e}\times\hat{\mathbf{f}}(\varphi)\,, \quad \hat{\mathbf{f}}(0) = \hat{\boldsymbol{\tau}}\,.$$

Since also

$$\hat{\mathbf{f}}(\varphi) = \hat{\mathbf{f}}_2(\varphi) \stackrel{\text{def}}{=} \hat{D}_{2\,\varphi\,\mathbf{e}}\,\hat{\boldsymbol{\tau}}$$

is such a solution, both $\hat{\mathbf{f}}_1(\varphi)$ and $\hat{\mathbf{f}}_2(\varphi)$ must coincide. ∎

Thus

$$e^{+i\,\frac{i}{\hbar}\,\hat{H}_{\mathrm{JC}}^{(n+1)}\,t}\,\hat{\boldsymbol{\tau}}\,e^{-i\,\frac{i}{\hbar}\,\hat{H}_{\mathrm{JC}}^{(n+1)}\,t} = \hat{D}_{2\Omega_n t\,\mathbf{e}'_n}\,\hat{\boldsymbol{\tau}}\,, \tag{9.20}$$

where

$$\mathbf{e}'_n \stackrel{\text{def}}{=} \begin{pmatrix} \sqrt{n+1}\,\lambda/\hbar \\ 0 \\ \delta/2 \end{pmatrix} \Big/ \Omega_n$$

and

$$\Omega_n \stackrel{\text{def}}{=} \sqrt{\left(\frac{\lambda}{\hbar}\right)^2 (n+1) + \left(\frac{\delta}{2}\right)^2} \quad (\text{R}{\small ABI}\ \textbf{\textit{frequency}})\,. \tag{9.21}$$

Note that, according to (8.13), the nutation of the Bloch vector (relative to $\mathbf{e}_3$) corresponds to transitions between the two levels of the atom with a circular frequency $\Omega_n$ .

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[5] By $\hat{D}_{\boldsymbol{\varphi}}$ we denote rotation around the axis along $\boldsymbol{\varphi}$ by the angle $|\boldsymbol{\varphi}|$ .

Assume that

$$\hat{\Psi}^{(n+1)} = \begin{pmatrix} \alpha_1 \\ \alpha_0 \end{pmatrix}_n \in \mathcal{H}^{(n+1)}\,, \quad \alpha_1 \geq 0\,,$$

is a normalized eigenstate of $\hat{H}_{\mathrm{JC}}^{(n+1)}$ , $n \in \mathbb{Z}_+$ , Then, by (9.20),

$$\begin{pmatrix} \alpha_1\,(\alpha_0 + \alpha_0^*) \\ -i\,\alpha_1\,(\alpha_0 - \alpha_0^*) \\ |\alpha_1|^2 - |\alpha_0|^2 \end{pmatrix} = \left\langle \hat{\Psi}^{(n+1)} \mid \hat{\boldsymbol{\tau}}\,\hat{\Psi}^{(n+1)} \right\rangle = \sigma \mathbf{e}'_n$$

holds for either $\sigma = +1$ or $\sigma = -1$ . This implies $\sigma\,\alpha_0 \geq 0$ and, together with normalization:

$$(\alpha_1)^2 + (\alpha_0)^2 = 1\,, \quad (\alpha_1)^2 - (\alpha_0)^2 = \frac{\sigma\,\delta}{2\,\Omega_n}\,.$$

Thus,

$$\alpha_1 = \sqrt[+]{\frac{1}{2} + \frac{\sigma\,\delta}{4\,\Omega_n}}\,, \quad \alpha_0 = \sigma \sqrt[+]{\frac{1}{2} - \frac{\sigma\,\delta}{4\,\Omega_n}}$$

holds for either $\sigma = +1$ or $\sigma = -1$ . We conclude that

$$\left\{ \hat{\Psi}_\sigma^{(n+1)} \stackrel{\text{def}}{=} \sqrt[+]{\frac{1}{2} + \frac{\sigma\,\delta}{4\,\Omega_n}}\,|1, n\rangle + \sigma \sqrt[+]{\frac{1}{2} - \frac{\sigma\,\delta}{4\,\Omega_n}}\,|0, n+1\rangle : \sigma \in \{-, +\} \right\}$$

is a maximal orthonormal set of eigenvectors of $\hat{H}_{\mathrm{JC}}^{(n+1)}$ :

$$\hat{H}_{\mathrm{JC}}^{(n+1)}\,\hat{\Psi}_\pm^{(n+1)} = \left(\check{E}_n \pm \hbar\,\Omega_n\right)\hat{\Psi}_\pm^{(n+1)}\,. \tag{9.22}$$

**Proof of (9.22):** The statement is a direct consequence of

$$\begin{aligned} \left(\hat{H}_{\mathrm{JC}}^{(n+1)} - \check{E}_n\,\hat{\tau}^0\right)\begin{pmatrix} \alpha_1 \\ \alpha_0 \end{pmatrix}_n &\underset{(9.17)}{=} \begin{pmatrix} \lambda\sqrt{n+1}\,\alpha_0 + \hbar\,\frac{\delta}{2}\,\alpha_1 \\ \lambda\sqrt{n+1}\,\alpha_1 - \hbar\,\frac{\delta}{2}\,\alpha_0 \end{pmatrix}_n \\ &\underset{(9.17)}{=} \begin{pmatrix} \left(\lambda\sqrt{n+1}\,\frac{\alpha_0}{\alpha_1} + \hbar\,\frac{\delta}{2}\right)\alpha_1 \\ \left(\lambda\sqrt{n+1}\,\frac{\alpha_1}{\alpha_0} - \hbar\,\frac{\delta}{2}\right)\alpha_0 \end{pmatrix}_n \end{aligned}$$

and

$$\begin{aligned} \lambda\sqrt{n+1}\,\sigma\sqrt{\frac{2\,\Omega_n \mp \sigma\,\delta}{2\,\Omega_n \pm \sigma\,\delta}} \pm \hbar\,\frac{\delta}{2} &= \frac{\sigma\,\lambda\sqrt{n+1}\,(2\,\Omega_n \mp \sigma\,\delta)}{\sqrt{4\,(\Omega_n)^2 - \delta^2}} \pm \hbar\,\frac{\delta}{2} \\ &\underset{(9.21)}{=} \frac{\sigma\,\lambda\sqrt{n+1}\,(2\,\Omega_n \mp \sigma\,\delta)}{2\,\frac{\lambda}{\hbar}\,\sqrt{n+1}} \pm \hbar\,\frac{\delta}{2} \\ &= \sigma\,\hbar\,\Omega_n\,. \quad \blacksquare \end{aligned}$$

Since

$$e^{i\,\boldsymbol{\varphi}\cdot\hat{\boldsymbol{\tau}}} = \cos|\boldsymbol{\varphi}|\;\hat{\tau}^0 + i\,\sin|\boldsymbol{\varphi}|\;\frac{\boldsymbol{\varphi}}{|\boldsymbol{\varphi}|}\cdot\hat{\boldsymbol{\tau}} \quad \forall\,\boldsymbol{\varphi}\in\mathbb{R}^3$$

(see Exercise E25b) of (Lücke, eine)), (9.17) also implies

$$e^{-\frac{i}{\hbar}\,\hat{H}_{\mathrm{JC}}^{(n+1)}\,t} = e^{-\frac{i}{\hbar}\,\hat{E}_n\,t}\left(\cos\left(\Omega_n\,t\right)\hat{\tau}^0 - i\,\sin\left(\Omega_n\,t\right)\left(\frac{\lambda\sqrt{n+1}}{\hbar\,\Omega_n}\,\hat{\tau}^1 + \frac{\delta}{2\Omega_n}\,\hat{\tau}^3\right)\right)\,.$$

# Appendix A

## A.1 Functional TAYLOR Expansion

Of course, the polarization of a nonlinear medium should be a **non**linear functional of electromagnetic field. The first order functional derivative characterizes this functional only approximately (fortunately with very high precision for usual fields). This is why a functional TAYLOR expansion is needed.

Let $\mathcal{T}$ be some complex vector space of functions on $\mathbb{R}^n$ with norm[1] $\|.\|$ and let $F$ be a continuous (complex) functional on $\mathcal{T}$, i.e. a continuous mapping from $\mathcal{T}$ into $\mathbb{C}$. Then the ***functional derivative***[2] of $F$ at $g \in \mathcal{T}$, if it exists, is the unique bounded **linear** Functional $L_g$ on $\mathcal{T}$ fulfilling

$$\lim_{\epsilon \to +0} \sup_{\substack{\varphi \in \mathcal{T} \\ \|\varphi\| < \epsilon}} \frac{1}{\epsilon} \left| F(g+\varphi) - F(g) - L_g(\varphi) \right| = 0$$

and is identified with the *generalized function* $\dfrac{\delta}{\delta g(\mathbf{x})} F(g)$ :

$$\int_{\mathbb{R}^n} \left( \frac{\delta}{\delta g(\mathbf{x})} F(g) \right) \varphi(\mathbf{x}) \, \mathrm{d}V_{\mathbf{x}} \stackrel{\text{def}}{=} L_g(\varphi) \quad \forall \, \varphi \in \mathcal{T} \, .$$

Note that

$$\int_{\mathbb{R}^n} \left( \frac{\delta}{\delta g(\mathbf{x})} F(g) \right) \varphi(\mathbf{x}) \, \mathrm{d}V_{\mathbf{x}} = \lim_{\epsilon \to +0} \frac{F(g+\epsilon\,\varphi) - F(g)}{\epsilon} \quad \forall \, \varphi \in \mathcal{T}$$

**if** $L_g$ exists. More generally, the $n$-fold functional derivative

$$\frac{\delta}{\delta g(\mathbf{x}_1)} \cdots \frac{\delta}{\delta g(\mathbf{x}_\nu)} F(g)$$

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[1]The linear space $\mathcal{T}$ need not be complete w.r.t. $\|.\|$ .

[2]This is a special case of the FRÉCHET ***derivative*** (see, e.g., (Choquet-Bruhat et al., 1978, p. 71)).

of $F$ at $g$ is iteratively defined by

$$\begin{aligned}
&\int_{\mathbb{R}^{(\nu+1)n}} \left(\frac{\delta}{\delta g(\mathbf{x}_1)} \dots \frac{\delta}{\delta g(\mathbf{x}_{\nu+1})} F(g)\right) \varphi_1(\mathbf{x}_1) \dots \varphi_{\nu+1}(\mathbf{x}_{\nu+1}) \, \mathrm{d}V_{\mathbf{x}_1} \dots \mathrm{d}V_{\mathbf{x}_{\nu+1}} \\
&\stackrel{\text{def}}{=} \int_{\mathbb{R}^n} \left(\frac{\delta}{\delta g(\mathbf{x})} F_{\varphi_2,\dots,\varphi_{\nu+1}}(g)\right) \varphi_1(\mathbf{x}) \, \mathrm{d}V_{\mathbf{x}} \qquad \forall\, \varphi_1,\dots,\varphi_{\nu+1} \in \mathcal{T}\,,
\end{aligned}$$

where

$$\begin{aligned}
&F_{\varphi_2,\dots,\varphi_{\nu+1}}(g) \\
&\stackrel{\text{def}}{=} \int_{\mathbb{R}^{\nu n}} \left(\frac{\delta}{\delta g(\mathbf{x}_2)} \dots \frac{\delta}{\delta g(\mathbf{x}_{\nu+1})} F(g)\right) \varphi_2(\mathbf{x}_2) \dots \varphi_{\nu+1}(\mathbf{x}_{\nu+1}) \, \mathrm{d}V_{\mathbf{x}_2} \dots \mathrm{d}V_{\mathbf{x}_{\nu+1}} \\
&\qquad\qquad \forall\, \varphi_2,\dots,\varphi_{\nu+1} \in \mathcal{T}\,.
\end{aligned}$$

This amounts to

$$\begin{aligned}
&\int_{\mathbb{R}^{\nu n}} \left(\frac{\delta}{\delta g(\mathbf{x}_1)} \dots \frac{\delta}{\delta g(\mathbf{x}_{\nu})} F(g)\right) \varphi_1(\mathbf{x}_1) \dots \varphi_{\nu}(\mathbf{x}_{\nu}) \, \mathrm{d}V_{\mathbf{x}_1} \dots \mathrm{d}V_{\mathbf{x}_{\nu}} \\
&= \left(\frac{\partial}{\partial \epsilon_1} \dots \frac{\partial}{\partial \epsilon_\nu} F\left(g + \epsilon_1\, \varphi_1 + \dots \epsilon_\nu\, \varphi_\nu\right)\right)_{|\epsilon_1 = \dots = \epsilon_\nu = 0} \qquad \forall\, \varphi_1,\dots,\varphi_\nu \in \mathcal{T}
\end{aligned} \tag{A.1}$$

whenever $\frac{\delta}{\delta g(\mathbf{x}_1)} \dots \frac{\delta}{\delta g(\mathbf{x}_{\nu})} F(g)$ exists. (A.1) especially implies

$$\begin{aligned}
&\left(\left(\frac{\partial}{\partial \epsilon}\right)^{\nu} F\left(g + \epsilon\, \varphi\right)\right)_{|\epsilon=0} \\
&= \int_{\mathbb{R}^{\nu n}} \left(\frac{\delta}{\delta g(\mathbf{x}_1)} \dots \frac{\delta}{\delta g(\mathbf{x}_{\nu})} F(g)\right) \varphi(\mathbf{x}_1) \dots \varphi(\mathbf{x}_{\nu}) \, \mathrm{d}V_{\mathbf{x}_1} \dots \mathrm{d}V_{\mathbf{x}_{\nu}} \quad \forall\, \varphi \in \mathcal{T}\,.
\end{aligned}$$

Therefore, for sufficiently well-behaved $F$, we have the ***functional*** TAYLOR ***expansion***

$$\begin{aligned}
F(\varphi) &= \left(e^{\int \mathrm{d}V_{\mathbf{x}}\, \varphi(\mathbf{x})\, \frac{\delta}{\delta g(\mathbf{x})}} F(g)\right)_{|g=0} \\
&\stackrel{\text{def}}{=} F(0) + \sum_{\nu=1}^{\infty} \frac{1}{\nu!} \int F_\nu(\mathbf{x}_1,\dots,\mathbf{x}_\nu)\, \varphi(\mathbf{x}_1) \dots \varphi(\mathbf{x}_\nu) \, \mathrm{d}V_{\mathbf{x}_1} \dots \mathrm{d}V_{\mathbf{x}_{\nu}}\,,
\end{aligned}$$

where

$$F_\nu(\mathbf{x}_1,\dots,\mathbf{x}_\nu) \stackrel{\text{def}}{=} \left(\frac{\delta}{\delta g(\mathbf{x}_1)} \dots \frac{\delta}{\delta g(\mathbf{x}_{\nu})} F(g)\right)_{|g=0} \qquad \forall\, \nu \in \mathbb{N}\,.$$

More precisely, we have

$$\begin{aligned}
F(\varphi) &= F(0) + \sum_{\nu=1}^{n} \frac{1}{\nu!} \int F_\nu(\mathbf{x}_1,\dots,\mathbf{x}_\nu)\, \varphi(\mathbf{x}_1) \dots \varphi(\mathbf{x}_\nu) \, \mathrm{d}V_{\mathbf{x}_1} \dots \mathrm{d}V_{\mathbf{x}_{\nu}} \\
&\quad + \frac{1}{n!} \int_0^1 (1-\lambda)^n \left(\frac{\mathrm{d}}{\mathrm{d}\lambda}\right)^{n+1} F(\lambda\varphi)\, \mathrm{d}\lambda\,,
\end{aligned}$$

This formalism has a straightforward generalization to vector-valued functions $\mathbf{g}$ on $\mathbb{R}^n$.

# A.2 Elementary Quantum Mechanics[3]

## A.2.1 Scalar Particles in $\mathbb{R}^1$

(In the Schrödinger picture) the (pure) state of a single, free, 1-dimensional, scalar, quantum mechanical 'particle' of mass $m$ is always given by a ***wave function*** $\Psi(x,t)$ fulfilling the ***free*** Schrödinger ***equation***

$$i\,\frac{\partial}{\partial t}\,\Psi(x,t) = \overbrace{\frac{1}{2m}\,\hat{p}^2}^{\hat{H}_{\mathrm{kin}} \stackrel{\mathrm{def}}{=}}\,\Psi(x,t)\,, \tag{A.2}$$

where

$$\boxed{\hat{p} \stackrel{\mathrm{def}}{=} \frac{\hbar}{i}\frac{\partial}{\partial x}\,.} \tag{A.3}$$

Its basic interpretation is

$$\boxed{\int_{\mathcal{G}} |\Psi(x,t)|^2\,\mathrm{d}x = \text{probability for 'position at time } t \text{ inside } \mathcal{G}\text{'}} \tag{A.4}$$

for all (sufficiently well-behaved) region $\mathcal{G} \subset \mathbb{R}$ and requires[4]

$$\int |\Psi(x,t)|^2\,\mathrm{d}x = 1 \tag{A.5}$$

for all $t \in \mathbb{R}$. Simple asymptotic considerations[5] show that the spatial Fourier transform

$$\boxed{\widetilde{\Psi}(p,t) \stackrel{\mathrm{def}}{=} \frac{1}{\sqrt{2\pi\hbar}} \int \Psi(x,t)\,e^{\frac{i}{\hbar}\,p\,x}\mathrm{d}x} \tag{A.6}$$

(adapted to quantum mechanics) has a similar interpretation w.r.t. linear momentum:[6]

$$\boxed{\int_{\widetilde{\mathcal{G}}} \left|\widetilde{\Psi}(p,t)\right|^2\,\mathrm{d}p = \text{probability for 'momentum at time } t \text{ inside } \widetilde{\mathcal{G}}\text{'.}} \tag{A.7}$$

The latter is consistent in the sense that

$$\begin{aligned} \int \left|\widetilde{\Psi}(p,t)\right|^2\,\mathrm{d}p &= \int |\Psi(x,t)|^2\,\mathrm{d}x \\ &= 1\,. \end{aligned} \tag{A.8}$$



[3] For mathematical details see, e.g., (Lücke, fuan).

[4] As usual, we write $\int$ for $\int_{-\infty}^{\infty}$ resp. $\int_{\mathbb{R}}$. One can easily show that (A.2) guarantees that (A.5) holds for all $t$ if fulfilled for $t = 0$.

[5] See, e.g., (Lücke, 1995, Section 5).

[6] Actually, (A.2) implies the l.h.s. of (A.7) — contrary to the l.h.s. of (A.4) — to be independent of $t$.

Defining

$$
\begin{aligned}
\Psi_t(x) &\stackrel{\text{def}}{=} \Psi(x,t)\,,\\
\langle \Psi \mid \Phi \rangle &\stackrel{\text{def}}{=} \int \overline{\Psi(x)}\Phi(x)\,\mathrm{d}x\,,
\end{aligned}
$$

and the ***position observable*** $\hat{x}$ by

$$
\boxed{\left(\hat{x}\,\Psi\right)(x) \stackrel{\text{def}}{=} x\,\Psi(x)} \tag{A.9}
$$

one gets

$$
\langle \Psi_t \mid \hat{x}\,\Psi_t \rangle = \int x\,|\Psi_t(x)|^2\,\mathrm{d}x
$$

and hence[7]

$$
\boxed{\langle \Psi_t \mid \hat{x}\,\Psi_t \rangle = \langle x(t) \rangle \stackrel{\text{def}}{=} \left\{ \begin{array}{l} \text{expectation value for the position variable} \\ x \text{ at time } t \text{ in the state given by } \Psi_t\,. \end{array} \right.} \tag{A.10}
$$

Similarly one has

$$
\boxed{\langle \Psi_t \mid \hat{p}\,\Psi_t \rangle = \langle p(t) \rangle \stackrel{\text{def}}{=} \left\{ \begin{array}{l} \text{expectation value for the momentum variable} \\ p \text{ at time } t \text{ in the state given by } \Psi_t\,, \end{array} \right.} \tag{A.11}
$$

since

$$
\Phi(x,t) = \hat{p}\,\Psi(x,t) \implies \widetilde{\Phi}(p,t) = p\,\widetilde{\Psi}(x,t)
$$

and, therefore, the generalization

$$
\int \overline{\Phi(x)}\,\Psi(x)\,\mathrm{d}x = \int \overline{\widetilde{\Phi}(p)}\,\widetilde{\Psi}(p)\,\mathrm{d}p \tag{A.12}
$$

of (A.8) implies

$$
\langle \Psi_t \mid \hat{p}\,\Psi_t \rangle = \int p\left|\widetilde{\Psi}(p,t)\right|^2\mathrm{d}p\,. \tag{A.13}
$$

In view of (A.11), $\hat{p}$ is called ***momentum observable***. (A.10) and (A.11) are consistent in the sense that

$$
\text{(A.2)} \implies \frac{\mathrm{d}}{\mathrm{d}t}\,\langle x(t) \rangle = \frac{1}{m}\,\langle p(t) \rangle = \text{constant.} \tag{A.14}
$$

Similarly to (A.14) one gets

$$
\langle \Psi_t \mid \underbrace{\hat{H}_{\text{kin}}}_{=\frac{\hat{p}^2}{2m}=-\frac{\hbar^2}{2m}\left(\frac{\partial}{\partial x}\right)^2} \Psi_t \rangle = \int \frac{p^2}{2m}\left|\widetilde{\Psi}(p,t)\right|^2\mathrm{d}p
$$



[7] Note, however, that $\langle x(t) \rangle$ may be infinite.

and, therefore,

$$\left\langle \Psi_t \,\middle|\, \frac{\hat{p}^2}{2m}\,\Psi_t \right\rangle = \langle E_{\mathrm{kin}}(t)\rangle \stackrel{\mathrm{def}}{=} \left\{ \begin{array}{l} \text{expectation value for the kinetic energy} \\ \text{at time } t \text{ in the state given by } \Psi_t\,. \end{array} \right. \tag{A.15}$$

If the particle is not free but interacting with an external force field with potential[8] $V(x)$ then, of course, (A.2) has to be modified. The basic interpretations (A.4) and (A.7), however, are maintained. Therefore, also (A.10) resp. (A.11) has to be maintained for (A.9) resp. (A.3). But now, in addition, we have

$$\langle \Psi_t \mid V(\hat{x})\,\Psi_t\rangle = \langle E_{\mathrm{kin}}(t)\rangle \stackrel{\mathrm{def}}{=} \left\{ \begin{array}{l} \text{expectation value for the potential energy} \\ \text{at time } t \text{ in the state given by } \Psi_t\,. \end{array} \right. \tag{A.16}$$

Assuming that the expectation value of the total energy is the sum of the expectation values for kinetic and potential energy, one concludes from (A.15) and (A.16) that

$$\boxed{\left\langle \Psi_t \mid \hat{H}\,\Psi_t \right\rangle = \langle E(t)\rangle \stackrel{\mathrm{def}}{=} \left\{ \begin{array}{l} \text{expectation value for the total energy} \\ \text{at time } t \text{ in the state given by } \Psi_t\,, \end{array} \right.} \tag{A.17}$$

where the operator

$$\boxed{\hat{H} \stackrel{\mathrm{def}}{=} \frac{\hat{p}^2}{2m} + V(\hat{x}) \quad (\boldsymbol{\mathit{Hamiltonian}})} \tag{A.18}$$

is the observable for the total energy. The latter suggests replacing (A.2) by the ***time dependent*** SCHRÖDINGER ***equation***

$$\boxed{i\,\frac{\partial}{\partial t}\,\Psi(x,t) = \hat{H}\,\Psi(x,t)\,,} \tag{A.19}$$

coinciding with (A.2) for $V = 0$. This is consistent in the sense that — generalizing (A.14) — (A.19) implies the EHRENFEST ***equations***

$$\frac{\mathrm{d}}{\mathrm{d}t}\,\langle x(t)\rangle = \frac{1}{m}\,\langle p(t)\rangle \ , \quad \frac{\mathrm{d}}{\mathrm{d}t}\,\langle p(t)\rangle = \int \left(-\frac{\mathrm{d}}{\mathrm{d}x}\,V(x)\right) |\Psi(x,t)|^2\,\mathrm{d}x$$

and $\langle E(t)\rangle$ =constant.

### A.2.2 Energy Eigenstates of the Harmonic Oscillator

One of the main objectives of quantum mechanics is to determine the (possibly improper) energy eigenfunctions, i.e. those $\Psi(x)$ for which there is an energy value $E$ with

$$\hat{H}\,\Phi(x) = E\,\Psi(x)\,. \tag{A.20}$$



[8]For particles interacting with a general electromagnetic field see 7.1.1.

These eigenfunctions give rise to the special solutions

$$\Psi(x,t) = e^{-\frac{i}{\hbar}E\,t}\,\Psi(x) \tag{A.21}$$

of (A.19) called ***stationary***, since

$$(\text{A.21}) \implies \frac{\partial}{\partial t}\left|\Psi(x,t)\right|^2 = 0\,.$$

Let us solve (A.20) for the ***(quantum mechanical) harmonic oscillator***, characterized by

$$V(x) = \frac{m}{2}\,\omega^2\,x^2\,. \tag{A.22}$$

This problem is easily solvable by means of the operators

$$\hat{a} \stackrel{\text{def}}{=} \frac{\hat{p} - i\,m\,\omega\,\hat{x}}{\sqrt{2\,m\,\hbar\,\omega}}\,,\quad \hat{a}^\dagger \stackrel{\text{def}}{=} \frac{\hat{p} + i\,m\,\omega\,\hat{x}}{\sqrt{2\,m\,\hbar\,\omega}} \tag{A.23}$$

fulfilling the commutation relations[9]

$$\left[\hat{a},\hat{a}^\dagger\right]_- = 1 \tag{A.24}$$

and hence the equations

$$\begin{aligned}\left(\hat{a}^\dagger\hat{a}\right)\hat{a} &= \hat{a}\,\left(\hat{a}^\dagger\hat{a} - 1\right),\\ \left(\hat{a}^\dagger\hat{a}\right)\hat{a}^\dagger &= \hat{a}^\dagger\left(\hat{a}^\dagger\hat{a} + 1\right).\end{aligned}$$

Iterating these equations gives[10]

$$\left(\hat{a}^\dagger\hat{a}\right)\hat{a}^\nu = \hat{a}^\nu\left(\hat{a}^\dagger\hat{a} - \nu\right) \quad \forall\,\nu\in\mathbb{N}\,, \tag{A.25}$$

$$\left(\hat{a}^\dagger\hat{a}\right)\left(\hat{a}^\dagger\right)^\nu = \left(\hat{a}^\dagger\right)^\nu\left(\hat{a}^\dagger\hat{a} + \nu\right) \quad \forall\,\nu\in\mathbb{N}\,. \tag{A.26}$$

Since

$$\begin{aligned}\hat{H} &= \frac{\hbar}{2}\,\omega\left(\hat{a}^\dagger\,\hat{a} + \hat{a}\,\hat{a}^\dagger\right)\\ &= \hbar\,\omega\left(\hat{a}^\dagger\,\hat{a} + \frac{1}{2}\right),\end{aligned} \tag{A.27}$$

we have

$$\hat{H}\,\Psi = E\,\Psi \implies \begin{cases} \hat{H}\,\left(\hat{a}^\nu\,\Psi\right) \underset{(\text{A.25})}{=} (E-\nu)\,\hat{a}^\nu\,\Psi \\ \hat{H}\,\left(\left(\hat{a}^\dagger\right)^\nu\Psi\right) \underset{(\text{A.26})}{=} (E+\nu)\,(\hat{a}^\dagger)^\nu\,\Psi \end{cases}$$

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[9] We simply write $z$ for $z\hat{1}$ (with $z\in\mathbb{C}$) in the following. Note that, in this sense, $[\hat{p}\,,\,\hat{x}] = \frac{\hbar}{i}$.

[10] Note that, e.g., (A.26) also follows from

$$\hat{a}\left(\hat{a}^\dagger\right)^\nu \underset{(1.82)}{=} \left(\hat{a}^\dagger\right)^\nu\hat{a} + \nu\left(\hat{a}^\dagger\right)^{\nu-1}.$$

for all (sufficiently well-behaved) $\Psi$. Thus, if $\Psi$ is an eigenfunction of $\hat{H}$ with eigenvalue $E$ then

$$\left\langle \hat{a}^\nu\,\Psi \mid \hat{H}\,\hat{a}^\nu\,\Psi \right\rangle = (E-\nu)\,\langle \hat{a}^\nu\,\Psi \mid \hat{a}^\nu\,\Psi\rangle$$

could become negative if $\hat{a}^\nu\,\Psi$ were nontrivial for all $\nu \in \mathbb{N}$. The latter, however is impossible because of

$$\begin{aligned} \left\langle \Phi \mid \hat{H}\,\Phi \right\rangle &\underset{(A.27)}{=} \hbar\,\omega\,\langle \hat{a}\,\Phi \mid \hat{a}\,\Phi\rangle + \frac{1}{2}\hbar\,\omega\,\langle \Phi \mid \Phi\rangle \\ &> 0 \quad \forall\,\Phi \neq 0\,. \end{aligned}$$

Therefore:

For every eigenfunction $\Psi$ of $\hat{H}$ there is some $\mu \in \mathbb{Z}_+$ with

$$\hat{a}\,(\hat{a}^\mu\,\Psi) = 0\,. \tag{A.28}$$

Now, $\hat{a}^\mu\,\Psi$ is fixed — up to some factor — by (A.28):

$$\begin{aligned} \hat{a}\,(\hat{a}^\mu\,\Psi) = 0 \quad &\underset{(A.23),(A.3)}{\Longrightarrow} \quad \frac{\mathrm{d}}{\mathrm{d}x}\,(\hat{a}^\mu\,\Psi)\,(x) = -\frac{m\,\omega}{\hbar}\,x\,(\hat{a}^\mu\,\Psi)\,(x) \\ &\Longrightarrow \quad \frac{\mathrm{d}}{\mathrm{d}x}\,\ln\,(\hat{a}^\mu\,\Psi)\,(x) = -\frac{m\,\omega}{\hbar}\,x \\ &\Longrightarrow \quad (\hat{a}^\mu\,\Psi)\,(x) \sim e^{-\frac{m\,\omega}{2\,\hbar}\,x^2}\,. \end{aligned}$$

We conclude that

$$\hat{H}\,\Psi = E\,\Psi \qquad \Longrightarrow \qquad 0 \neq \hat{a}^\mu\,\Psi \sim \Omega \quad \text{for some } \mu \in \mathbb{Z}_+\,, \tag{A.29}$$

where[11]

$$\Omega(x) \stackrel{\text{def}}{=} \left(\frac{m\,\omega}{\pi\,\hbar}\right)^{\frac{1}{4}} e^{-\frac{m\,\omega}{2\,\hbar}\,x^2}\,, \quad \hat{a}\,\Omega = 0\,. \tag{A.30}$$

Iterating

$$\begin{aligned} \left(\hat{a}^\dagger\right)^\nu\,\hat{a}^\nu &= \left(\hat{a}^\dagger\right)^{\nu-1}\left(\hat{a}^\dagger\hat{a}\right)\hat{a}^{\nu-1} \\ &\underset{(A.25)}{=} \left(\hat{a}^\dagger\right)^{\nu-1}\hat{a}^{\nu-1}\Big(\hat{a}^\dagger\hat{a} - (\nu-1)\Big) \end{aligned}$$

we get

$$\left(\hat{a}^\dagger\right)^\nu\,\hat{a}^\nu = \left(\hat{a}^\dagger\hat{a}\right)\left(\hat{a}^\dagger\hat{a}-1\right)\cdots\Big(\hat{a}^\dagger\hat{a}-(\nu-1)\Big) \quad \forall\,\nu \in \mathbb{N} \tag{A.31}$$

and conclude:[12]

$$\hat{H}\,\left(\hat{a}^\dagger\right)^\mu\,\Omega \underset{(A.26),(A.30)}{=} \left(\mu + \frac{1}{2}\right)\hbar\,\omega\,\Omega \quad \forall\,\mu \in \mathbb{Z}_+\,,$$



[11]Note that $\displaystyle\int e^{-x^2}\,\mathrm{d}x = \sqrt{\pi}$ (more generally, see (2.83)), hence $\langle \Omega \mid \Omega\rangle = 1\,.$

[12]Note that

$$\hat{a}^\mu\,\Psi \neq 0 \implies \left(\hat{a}^\dagger\right)^\mu\,\hat{a}^\mu\,\Psi \neq 0\,,$$

$$\hat{H}\,\Psi = E\,\Psi \quad \underset{(A.29),(A.31),(A.27)}{\Longrightarrow} \quad \Psi \sim \left(\hat{a}^\dagger\right)^\mu \Omega \quad \text{for some } \mu \in \mathbb{Z}_+\,,$$

### A.2.3 Coherent States

Recall[13] that the operators

$$\hat{a}_{\text{osc}} \stackrel{\text{def}}{=} \frac{\frac{\hbar}{i}\frac{\mathrm{d}}{\mathrm{d}x} - i\,m\,\omega\,x}{\sqrt{2\,m\,\hbar\,\omega}}\,, \quad \hat{a}^\dagger_{\text{osc}} = \frac{\frac{\hbar}{i}\frac{\mathrm{d}}{\mathrm{d}x} + i\,m\,\omega\,x}{\sqrt{2\,m\,\hbar\,\omega}}$$

of the 1-dimensional harmonic oscillator with Hamiltonian

$$\hat{H}_{\text{osc}} \stackrel{\text{def}}{=} -\frac{\hbar^2}{2\,m}\left(\frac{\mathrm{d}}{\mathrm{d}x}\right)^2 + \frac{1}{2}\,m\,\omega^2\,x^2$$

also obey the commutation relation

$$\left[\hat{a}_{\text{osc}}, \hat{a}^\dagger_{\text{osc}}\right]_- = 1$$

and that

$$\hat{H}_{\text{osc}} = \frac{\hbar}{2}\,\omega\left(\hat{a}^\dagger_{\text{osc}}\,\hat{a}_{\text{osc}} + \hat{a}_{\text{osc}}\,\hat{a}^\dagger_{\text{osc}}\right)\,,$$

hence

$$\hat{H}_{\text{osc}} = \hbar\,\omega\left(\hat{a}^\dagger_{\text{osc}}\,\hat{a}_{\text{osc}} + \frac{1}{2}\right)\,.$$

Here the ground state

$$\Omega_{\text{osc}}(x) \stackrel{\text{def}}{=} \left(\frac{m\,\omega}{\pi\,\hbar}\right)^{\frac{1}{4}} e^{-\frac{m\,\omega}{2\,\hbar}\,x^2}$$

corresponds to the vacuum:

$$\hat{a}_{\text{osc}}\,\Omega_{\text{osc}} = 0\,.$$

Therefore, since

$$\alpha\,\hat{a}^\dagger_{\text{osc}} - \overline{\alpha}\,\hat{a}_{\text{osc}} = \Im(\alpha)\sqrt{\frac{2\,\hbar}{m\,\omega}}\,\frac{\mathrm{d}}{\mathrm{d}x} + i\,\Re(\alpha)\sqrt{\frac{2\,m\,\omega}{\hbar}}\,x\,,$$

the corresponding coherent states, according to (1.79) and (1.74), are

$$\Psi_{\hat{a}_{\text{osc}},\alpha}(x) = \left(\frac{m\,\omega}{\pi\,\hbar}\right)^{\frac{1}{4}} e^{i\,\Re(\alpha)\Im(\alpha)}\,e^{i\,\Re(\alpha)\sqrt{\frac{2\,m\,\omega}{\hbar}}\,x}\,e^{-\frac{m\,\omega}{2\,\hbar}\left(x+\Im(\alpha)\sqrt{\frac{2\,\hbar}{m\,\omega}}\right)^2}\,.$$

This shows that the coherent states of the harmonic oscillator are just displacements in position and momentum of the ground state, hence have the same minimal



[13] See A.2.2.

uncertainty product[14]

$$\Delta x\,\Delta p = \frac{\hbar}{2} = \frac{\hbar}{2}\,\Delta(a + a^\dagger)\,\Delta(i\,a - i\,a^\dagger)\,.$$

In this sense, the coherent states of the quantized electromagnetic field correspond to coherent classical fields as closely as possible.[15]

## A.3 Construction of Field Operators

Denote by $\mathfrak{F}$ the complex vector space of all truncated sequences

$$\mathfrak{f} = \{f_0, f_1, f_2, \ldots\}$$

of (sufficiently well-behaved) functions[16] $f_\nu(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_\nu, j_\nu)$ over $(\mathbb{R}^3 \times \{1,2,3\})^\nu$ fulfilling[17]

$$f_\nu(\mathbf{k}_{\pi 1}, j_{\pi 1}; \ldots; \mathbf{k}_{\pi\nu}, j_{\pi\nu}) = f_\nu(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_\nu, j_\nu) \quad \forall\,\pi \in S_\nu$$

and

$$\sum_{j_1,\ldots,j_\nu=1}^{3} \int |f_\nu(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_\nu, j_\nu)|^2\, \mathrm{d}V_{\mathbf{k}_1} \ldots \mathrm{d}V_{\mathbf{k}_\nu} < \infty\,.$$

Of course, the linear structure is meant to be the natural one:

$$\begin{aligned} z\,\mathfrak{f} &= \{z\,f_0, z\,f_1, z\,f_2, \ldots\} \quad \forall\, z \in \mathbb{C}\,,\ \mathfrak{f} \in \mathfrak{F}\,,\\ \mathfrak{f} + \mathfrak{f}' &\stackrel{\text{def}}{=} \{f_0 + f_0', f_1 + f_1', f_2 + f_2', \ldots\} \quad \forall\, \mathfrak{f}, \mathfrak{f}' \in \mathfrak{F}\,. \end{aligned}$$

With the inner product

$$\langle \mathfrak{f} \mid \mathfrak{f}' \rangle \stackrel{\text{def}}{=} \overline{f_0}\, f_0' + \sum_{j_1,\ldots,j_\nu=1}^{3} \int \overline{f_\nu(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_\nu, j_\nu)}\, f_\nu'(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_\nu, j_\nu) \mathrm{d}V_{\mathbf{k}_1} \ldots \mathrm{d}V_{\mathbf{k}_\nu}$$



[14] Quite generally we have

$$\begin{aligned} \left\|(\hat{A} - a)\Psi\right\|^2 \left\|(\hat{B} - b)\Psi\right\|^2 &\geq \left|\left\langle \left(\hat{A} - a\right)\Psi \,\middle|\, \left(\hat{B} - b\right)\Psi \right\rangle\right|^2 \\ &= \tfrac{1}{4}\left\|\underbrace{\left\langle \Psi \,\middle|\, [\hat{A}, \hat{B}]_-\, \Psi \right\rangle}_{\text{real}}\right\|^2 + \tfrac{1}{4}\left\|\underbrace{\left\langle \Psi \,\middle|\, [\hat{A} - a, \hat{B} - b]_+\, \Psi \right\rangle}_{\text{imaginary}}\right\|^2 . \end{aligned}$$

for all selfadjoint operators $\hat{A}, \hat{B}$, all real numbers $a, b$, and every vector $\Psi \in D_{\hat{B}\hat{A}} \cap D_{\hat{A}\hat{B}}$.

[15] States with $\Delta(a + a^\dagger) < 1$ or $\Delta(i\,a - i\,a^\dagger) < 1$ are called ***squeezed states*** (see (Scully and Zubairy, 1999, Sections 2.5–2.8) for a discussion of these states).

[16] The physical dimension of the values of these functions should be such that the integrals $\int |f_\nu(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_\nu, j_\nu)|^2\, \mathrm{d}V_{\mathbf{k}_1} \ldots \mathrm{d}V_{\mathbf{k}_\nu}$ are just real numbers without any physical dimension.

[17] As usual, we denote by $S_\nu$ the group of all permutations of $(1, \ldots, \nu)$.

F becomes a euklidean vector space (pre-HILBERT space). Now let us introduce operator-valued functions $\hat{b}_j(\mathbf{k})$ by[18]

$$
\begin{aligned}
&\mathfrak{f}^- = \hat{b}_j(\mathbf{k})\,\mathfrak{f} \\
&\overset{\text{def}}{\Longleftrightarrow}\; f^-_\nu(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_\nu, j_\nu) = \ell^{-\frac{3}{2}}\sqrt{\nu}\, f_{\nu+1}(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_\nu, j_\nu; \mathbf{k}, j) \quad \forall\, \nu \in \mathbb{Z}_+\,,
\end{aligned}
$$

where $\ell$ is some fixed length necessary for adjusting the physical dimensions. Then for the adjoint operators $\left(\hat{b}_j(\mathbf{k})\right)^\dagger$, characterized by

$$
\left\langle \mathfrak{f} \,\middle|\, \hat{b}_j(\mathbf{k})\,\mathfrak{f}' \right\rangle = \left\langle \left(\hat{b}_j(\mathbf{k})\right)^\dagger \mathfrak{f} \,\middle|\, \mathfrak{f}' \right\rangle \quad \forall\, \mathfrak{f}, \mathfrak{f}' \in \mathfrak{F}\,,
$$

we have $\mathfrak{f}^+ = \left(\hat{b}_j(\mathbf{k})\right)^\dagger \mathfrak{f}$ iff $f^+_0 = 0$ and:

$$
\begin{aligned}
&\ell^{\frac{3}{2}}\, f^+_{\nu+1}(\mathbf{k}_1, j_1; \ldots; \mathbf{k}_{\nu+1}, j_{\nu+1}) \\
&= \frac{1}{\sqrt{\nu+1}} \sum_{\mu=1}^{\nu} \delta_{j_n u j}\, \delta\left(\mathbf{k}_\mu - \mathbf{k}\right) f_{\nu+1}(\mathbf{k}_1, j_1; \ldots; \cancel{\mathbf{k}_\mu, j_\mu}; \ldots; \mathbf{k}_{\nu+1}, j_{\nu+1}) \quad \forall\, \nu \in \mathbb{Z}_+\,.
\end{aligned}
$$

Consequently, we have the commutation relations

$$
[\hat{b}_j(\mathbf{k}), \hat{b}_{j'}(\mathbf{k}')]_- = 0\,, \qquad \left[\hat{b}_j(\mathbf{k}), \left(\hat{b}_{j'}(\mathbf{k}')\right)^\dagger\right]_- = \ell^{-3}\,\delta_{jj'}\,\delta(\mathbf{k}-\mathbf{k}')
$$

and the definition

$$
\hat{a}_j(\mathbf{k}) \;\overset{\text{def}}{=}\; \ell^{\frac{3}{2}}\,\boldsymbol{\epsilon}_j(\mathbf{k}) \cdot \hat{\mathbf{b}}(\mathbf{k}) \quad \forall\, j \in \{1,2\}\,,\ \mathbf{k} \in \mathbb{R}^3\,,
$$

where

$$
\hat{\mathbf{b}}(\mathbf{k}) \overset{\text{def}}{=} \sum_{j=1}^{3} \hat{b}_j(\mathbf{k})\,\mathbf{e}_j\,,
$$

yields a realization of the canonical commutation relations (1.37) fulfilling (1.38) for

$$
\Omega \overset{\text{def}}{=} (1, 0, 0, \ldots)\,.
$$

Note that

$$
\sum_{j=1}^{2} \boldsymbol{\epsilon}_j(\mathbf{k})\,\hat{a}_j(\mathbf{k}) = \hat{\mathbf{b}}(\mathbf{k}) - \frac{\mathbf{k}}{|\mathbf{k}|} \cdot \hat{\mathbf{b}}(\mathbf{k}) \quad \forall\, \mathbf{k} \neq 0\,.
$$

This shows that $\hat{\mathbf{A}}^{(+)}(\mathbf{x}, t)$ does not depend on the choice of polarization vectors $\boldsymbol{\epsilon}_j(\mathbf{k})$ in (1.31).



[18]Of course '$\ldots; \mathbf{k}_\nu, j_\nu$' should be dropped for $\nu \leq 1$ and also '$\mathbf{k}_1, j_1$' if $\nu = 0$.

## A.4 Dyson Series

In the Schrödinger picture the state vectors (wave functions) $\Psi_t = \Psi_t^{\mathrm{S}}$ describe the momentary situation of the quantum system at time $t$. Thus, if $\hat{A} = \hat{A}^{\mathrm{S}}$ is the observable of a physical quantity $A$ then $\left\langle \Psi_t^{\mathrm{S}} \mid \hat{A}^{\mathrm{S}}\, \Psi_t^{\mathrm{S}} \right\rangle$ is the corresponding expectation value for $A$ at time $t$. The time evolution is described by the Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} \Psi_t^{\mathrm{S}} = \hat{H}^{\mathrm{S}}(t)\, \Psi_t^{\mathrm{S}} . \tag{A.32}$$

Now assume that the Hamiltonian in the Schrödinger picture is of the form

$$\hat{H}^{\mathrm{S}}(t) = \hat{H}_0^{\mathrm{S}} + \hat{V}^{\mathrm{S}}(t) , \tag{A.33}$$

Where $\hat{H}_0^{\mathrm{S}}(t)$ corresponds to some known 'free evolution' and $\hat{V}^{\mathrm{S}}(t)$ is some ***perturbation*** of the 'free' Hamiltonian $\hat{H}_0^{\mathrm{S}}(t)$. Then we may get a perturbative solution of (A.32) by switching to the interaction picture:[19]

$$\Psi_t^{\mathrm{S}} \quad \longmapsto \quad \Psi_t^{\mathrm{I}} \stackrel{\mathrm{def}}{=} e^{\frac{i}{\hbar} \hat{H}_0^{\mathrm{S}}\, t}\, \Psi_t^{\mathrm{S}} , \tag{A.34}$$

$$\hat{A}^{\mathrm{S}}(t) \quad \longmapsto \quad \hat{A}^{\mathrm{I}}(t) \stackrel{\mathrm{def}}{=} e^{+\frac{i}{\hbar} \hat{H}_0^{\mathrm{S}}\, t}\, \hat{A}^{\mathrm{S}}(t)\, e^{-\frac{i}{\hbar} \hat{H}_0^{\mathrm{S}}\, t} . \tag{A.35}$$

This way (A.32) becomes equivalent to

$$i\hbar \frac{\partial}{\partial t} \Psi_t^{\mathrm{I}} = \hat{V}^{\mathrm{I}}(t)\, \Psi_t^{\mathrm{I}} . \tag{A.36}$$

Sure, (A.36) has the same formal structure as (A.32). Now, however, for the sufficiently small $T$ the mapping[20]

$$\mathcal{I} :\ \Phi(t) \longmapsto -\frac{i}{\hbar} \int_0^t \hat{V}^{\mathrm{I}}(t)\, \Phi(t')\, \mathrm{d}t' \tag{A.37}$$

may be ***contracting*** with respect to the norm

$$\|\Phi\|_T \stackrel{\mathrm{def}}{=} \int_0^T \|\Phi(t)\|\ \mathrm{d}t ;$$

i.e. the may be some $\lambda < 1$ with

$$\|\Phi\|_T \leq \lambda\, \|\Phi\|_T \quad \forall\, \Phi .$$

If this is the case we have

$$\Psi_t^{\mathrm{I}} = \sum_{\nu=0}^{\infty} \left( \mathcal{I}^\nu \Psi_0^{\mathrm{I}} \right)(t) \quad \forall\, t \in [0, T] \tag{A.38}$$

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[19] See (Lücke, qft, Sect. 3.1.3) for a qualitative discussion.

[20] Here we assume that $\Phi(t)$ is a (sufficiently well behaved) function of $t$ with values in the state space $\mathcal{H}$ and denote by $\|.\|$ the norm corresponding to the inner product of $\mathcal{H}$.

(compare Section 5.1.3 of (Lücke, ein)), since (A.36) is equivalent to the integral equation

$$\Psi^{\mathrm{I}}_t = \Psi^{\mathrm{I}}_0 - \frac{i}{\hbar}\int_0^t \hat{V}^{\mathrm{I}}(t')\,\Psi^{\mathrm{I}}_{t'}\,\mathrm{d}t'\,.$$

Using ***time ordering***[21]

$$\mathcal{T}\left(\hat{V}^{\mathrm{I}}(t_{\pi 1})\cdots\hat{V}^{\mathrm{I}}(t_{\pi\nu})\right) \stackrel{\text{def}}{=} \hat{V}^{\mathrm{I}}(t_1)\cdots\hat{V}^{\mathrm{I}}(t_\nu) \quad \forall\, t_1\leq\ldots\leq t_\nu\,,\ \pi\in S_\nu$$

we may rewrite (A.38) as DYSON ***series***:[22]

$$\boxed{\begin{aligned}\Psi^{\mathrm{I}}_t &= \mathcal{T}\left(e^{-\frac{i}{\hbar}\int_0^t \hat{V}^{\mathrm{I}}(t')\,\mathrm{d}t'}\right)\Psi^{\mathrm{I}}_0\\ &\stackrel{\text{def}}{=} \Psi^{\mathrm{I}}_0(0) + \sum_{\nu=1}^{\infty}\frac{1}{\nu!}\left(-\frac{i}{\hbar}\right)^{\nu}\int_{[0,t]^\nu}\mathcal{T}\left(\hat{V}^{\mathrm{I}}(t_1)\cdots\hat{V}^{\mathrm{I}}(t_\nu)\right)\Psi^{\mathrm{I}}_0\,\mathrm{d}t_1\cdots\mathrm{d}t_\nu\,.\end{aligned}} \tag{A.39}$$

Usually only the ***first order*** approximation

$$\Psi^{\mathrm{I}}_t \approx \Psi^{\mathrm{I}}_0 - \frac{i}{\hbar}\int_0^t \hat{V}^{\mathrm{I}}(t')\,\Psi^{\mathrm{I}}_0\,\mathrm{d}t' \quad \forall\, t\in[0,T] \tag{A.40}$$

is used — as justified for sufficiently small $T$. This implies for the ***transition probability***

$$\left|\left\langle\Phi^0_0 \mid \Psi^{\mathrm{I}}_t\right\rangle\right| \approx \hbar^{-2}\left|\int_0^t\left\langle\Phi^0_0 \mid \hat{V}^{\mathrm{I}}(t')\,\Psi^{\mathrm{I}}_0\right\rangle\mathrm{d}t'\right|^2 \quad \textbf{if } \left\langle\Phi^0_0 \mid \Psi^{\mathrm{I}}_t\right\rangle = 0\,. \tag{A.41}$$

**Remark:** Since

$$\left|\int_0^t\left\langle\Phi^0_0 \mid \hat{V}^{\mathrm{I}}(t')\,\Psi^{\mathrm{I}}_0\right\rangle\mathrm{d}t'\right|^2 = \int_{[0,t]\times[0,t]}\left\langle\Phi^0_0 \mid \hat{V}^{\mathrm{I}}(t_1)\,\Psi^{\mathrm{I}}_0\right\rangle\left\langle\Psi^{\mathrm{I}}_0 \mid \hat{V}^{\mathrm{I}}(t_2)\,\Phi^0_0\right\rangle\mathrm{d}t_1\,\mathrm{d}t_2\,,$$

the corresponding formula for mixed states with density matrix $\hat{\rho}^{\mathrm{I}}_t$ is

$$\mathrm{trace}\left(\hat{\rho}^{\mathrm{I}}_t\,|\Phi^0_0\rangle\langle\Phi^0_0|\right) \approx \hbar^{-2}\int_{[0,t]\times[0,t]}\left\langle\Phi^0_0 \mid \hat{V}^{\mathrm{I}}(t_1)\,\hat{\rho}^{\mathrm{I}}_0\,\hat{V}^{\mathrm{I}}(t_2)\,\Phi^0_0\right\rangle\mathrm{d}t_1\,\mathrm{d}t_2\,,$$

in agreement with (6.19).

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[21] As usual, we denote by $S_\nu$ the set of all permutations of $1,\ldots,\nu$.

[22] Of course, we tacitly assume also $t\mapsto\hat{V}^{\mathrm{I}}(t)$ to be sufficiently well behaved.

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