Chapter 2
Macroscopic Quantum Electrodynamics

The theory of macroscopic quantum electrodynamics is developed which forms the basis for the analysis of dispersion forces presented in this book. Basic concepts of classical electrodynamics and quantum electrodynamics in free space are recalled. Using these as a guiding principle, we then construct a consistent theory of the quantised electromagnetic field in the presence of magnetoelectric media based on the classical Green's tensor for the electromagnetic field. Atom–field interactions are implemented via the minimal and multipolar coupling schemes.

2.1 Elements of Classical Electrodynamics

Let us first review the basic concepts of classical electrodynamics. Starting with the simplest case of electrodynamics in free space, we then discuss the more general cases of charged particles and magnetoelectric media being present.

2.1.1 Electrodynamics in free Space

Classical electrodynamics is governed by the classical Maxwell equations for the electromagnetic field. In the absence of charges or currents, they take their simplest form

$$\nabla \cdot \boldsymbol{E} = 0\,, \tag{2.1}$$

$$\nabla \cdot \boldsymbol{B} = 0\,, \tag{2.2}$$

$$\nabla \times \boldsymbol{E} + \dot{\boldsymbol{B}} = \boldsymbol{0}\,, \tag{2.3}$$

$$\nabla \times \boldsymbol{B} - \frac{1}{c^2}\,\dot{\boldsymbol{E}} = \boldsymbol{0}\,, \tag{2.4}$$

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where both the electric field $\boldsymbol{E} = \boldsymbol{E}(\boldsymbol{r}, t)$ and the magnetic field[1] $\boldsymbol{B} = \boldsymbol{B}(\boldsymbol{r}, t)$ are functions of position and time. The first of these equations is commonly known as the Gauss law, the second one states the non-existence of magnetic monopoles, the third one is the Faraday law of induction and the last one is the Ampère law.

To simplify this system of equations, we introduce the scalar potential ϕ and the vector potential $\boldsymbol{A}$ for the electromagnetic field according to

$$\boldsymbol{E} = -\boldsymbol{\nabla}\phi - \dot{\boldsymbol{A}}\,, \tag{2.5}$$

$$\boldsymbol{B} = \boldsymbol{\nabla} \times \boldsymbol{A}\,. \tag{2.6}$$

Equations (2.2) and (2.3) are then automatically fulfilled and the remaining equations (2.1) and (2.4) are equivalent to

$$-\Delta\phi - \boldsymbol{\nabla} \cdot \dot{\boldsymbol{A}} = 0\,, \tag{2.7}$$

$$\frac{1}{c^2}\,\boldsymbol{\nabla}\dot{\phi} + \frac{1}{c^2}\,\ddot{\boldsymbol{A}} - \Delta\boldsymbol{A} + \boldsymbol{\nabla}(\boldsymbol{\nabla} \cdot \boldsymbol{A}) = \boldsymbol{0}\,. \tag{2.8}$$

Note that ϕ and $\boldsymbol{A}$ are not uniquely defined. In particular, the electric and magnetic fields as given by (2.5) and (2.6) are invariant under the gauge transformation

$$\phi \to \phi - \dot{\Lambda}\,, \tag{2.9}$$

$$\boldsymbol{A} \to \boldsymbol{A} + \boldsymbol{\nabla}\Lambda \tag{2.10}$$

with some arbitrary scalar field Λ. One can exploit this freedom in order to simplify (2.7) and (2.8). Throughout this book, we employ the Coulomb gauge imposed by the condition

$$\boldsymbol{\nabla} \cdot \boldsymbol{A} = 0\,. \tag{2.11}$$

We recall that any vector field $\boldsymbol{f}$ can be uniquely decomposed into its longitudinal ($\|$) and transverse ($\perp$) parts defined by

$$\boldsymbol{f} = \boldsymbol{f}^{\|} + \boldsymbol{f}^{\perp} \tag{2.12}$$

with

$$\boldsymbol{\nabla} \times \boldsymbol{f}^{\|} = \boldsymbol{0}\,, \qquad \boldsymbol{\nabla} \cdot \boldsymbol{f}^{\perp} = 0\,. \tag{2.13}$$

They can explicitly be given by

$$\boldsymbol{f}^{\|(\perp)}(\boldsymbol{r}) = \int \mathrm{d}^3r'\, \boldsymbol{\delta}^{\|(\perp)}(\boldsymbol{r} - \boldsymbol{r}') \cdot \boldsymbol{f}(\boldsymbol{r}') \tag{2.14}$$

[1] Note that $\boldsymbol{B}$ is alternatively referred to as the induction field.

with

$$\boldsymbol{\delta}^{\|}(\boldsymbol{r}) = -\boldsymbol{\nabla}\,\boldsymbol{\nabla}\,\frac{1}{4\pi r}, \qquad \boldsymbol{\delta}^{\perp}(\boldsymbol{r}) = \boldsymbol{\nabla} \times (\boldsymbol{\nabla} \times \boldsymbol{I})\,\frac{1}{4\pi r} \tag{2.15}$$

being the longitudinal and transverse delta functions, recall that

$$\boldsymbol{\delta}(\boldsymbol{r}) = \delta(\boldsymbol{r})\boldsymbol{I} = -\Delta\,\frac{1}{4\pi r}\,\boldsymbol{I} \tag{2.16}$$

($\boldsymbol{I}$: unit tensor). In Coulomb gauge, (2.5) uniquely relates ϕ and $\boldsymbol{A}$ to the longitudinal and transverse parts of the electric field, respectively,

$$\boldsymbol{E}^{\|} = -\,\boldsymbol{\nabla}\phi\,, \tag{2.17}$$

$$\boldsymbol{E}^{\perp} = -\,\dot{\boldsymbol{A}}\,. \tag{2.18}$$

As a consequence of this gauge, the equations for ϕ and $\boldsymbol{A}$ decouple. The scalar potential is seen to be identical with the electrostatic Coulomb potential (hence the name Coulomb gauge) satisfying a Laplace equation

$$\Delta\phi = 0\,, \tag{2.19}$$

while the vector potential obeys a Helmholtz equation

$$\frac{1}{c^2}\,\ddot{\boldsymbol{A}} - \Delta\boldsymbol{A} = \boldsymbol{0}\,. \tag{2.20}$$

The Laplace equation is trivially solved by $\phi = 0$. In view of the quantisation of the electromagnetic field, it is favourable to reformulate the remaining dynamical equation for $\boldsymbol{A}$ within a Langrangian/Hamiltonian based formalism. To that end, we introduce the Lagrangian of the transverse electromagnetic field

$$L = \int \mathrm{d}^3 r\,\mathcal{L}\,, \tag{2.21}$$

with the Lagrangian density being given by

$$\mathcal{L} = \frac{1}{2}\left[\varepsilon_0\dot{\boldsymbol{A}}^2 - \frac{1}{\mu_0}\,(\boldsymbol{\nabla}\times\boldsymbol{A})^2\right]. \tag{2.22}$$

One can show that

$$\int \mathrm{d}^3r\, (\nabla \times \boldsymbol{A})^2 = \int \mathrm{d}^3r \left\{\mathrm{tr}\Big(\boldsymbol{A}\overleftarrow{\nabla} \cdot \nabla\boldsymbol{A}\Big) - \mathrm{tr}[(\nabla\boldsymbol{A}) \cdot (\nabla\boldsymbol{A})]\right\}$$
$$= \int \mathrm{d}^3r \left\{\mathrm{tr}\Big(\boldsymbol{A}\overleftarrow{\nabla} \cdot \nabla\boldsymbol{A}\Big) - \nabla \cdot (\boldsymbol{A} \cdot \nabla\boldsymbol{A}) + \boldsymbol{A} \cdot \nabla\nabla \cdot \boldsymbol{A}\right\}$$
$$= \int \mathrm{d}^3r\, \mathrm{tr}\Big(\boldsymbol{A}\overleftarrow{\nabla} \cdot \nabla\boldsymbol{A}\Big) \tag{2.23}$$

($\overleftarrow{\nabla}$: differentiation to the left), where the second term on the second line leads to a surface integral at infinity which vanishes while the third term vanishes according to the Coulomb gauge used. The alternative Lagrangian density

$$\mathcal{L} = \frac{1}{2}\left[\varepsilon_0 \dot{\boldsymbol{A}}^2 - \frac{1}{\mu_0}\,\mathrm{tr}\Big(\boldsymbol{A}\overleftarrow{\nabla} \cdot \nabla\boldsymbol{A}\Big)\right] \tag{2.24}$$

is therefore equivalent to the one above. To ensure that the Lagrangian is correct, we need to verify that the associated Lagrange equation

$$\frac{\delta L}{\delta \boldsymbol{A}} = \frac{\mathrm{d}}{\mathrm{d}t}\frac{\delta L}{\delta \dot{\boldsymbol{A}}} \tag{2.25}$$

is equivalent to the dynamical equation (2.20). Calculating

$$\frac{\delta L}{\delta \boldsymbol{A}} = \frac{\partial \mathcal{L}}{\partial \boldsymbol{A}} - \nabla \cdot \frac{\partial \mathcal{L}}{\partial \nabla\boldsymbol{A}} = \frac{1}{\mu_0}\Delta\boldsymbol{A}$$
$$= \frac{\mathrm{d}}{\mathrm{d}t}\frac{\delta L}{\delta \dot{\boldsymbol{A}}} = \frac{\mathrm{d}}{\mathrm{d}t}\frac{\partial \mathcal{L}}{\partial \dot{\boldsymbol{A}}} = \varepsilon_0 \ddot{\boldsymbol{A}}\,, \tag{2.26}$$

we see that this is indeed the case.

Next, we introduce the canonically conjugate momentum associated with $\boldsymbol{A}$ via

$$\boldsymbol{\Pi} = \frac{\delta L}{\delta \dot{\boldsymbol{A}}} = \frac{\partial \mathcal{L}}{\partial \dot{\boldsymbol{A}}} = \varepsilon_0 \dot{\boldsymbol{A}} = -\varepsilon_0 \boldsymbol{E}^{\perp}\,. \tag{2.27}$$

Applying a Legendre transformation

$$H = \int \mathrm{d}^3r\, \boldsymbol{\Pi} \cdot \dot{\boldsymbol{A}} - L \tag{2.28}$$

and expressing the result in terms of canonically conjugate fields $\boldsymbol{A}$ and $\boldsymbol{\Pi}$ rather than $\boldsymbol{A}$ and $\dot{\boldsymbol{A}}$, we find the Hamiltonian of the transverse electromagnetic field

$$H = \int \mathrm{d}^3r\, \mathcal{H}\,, \tag{2.29}$$

with the Hamilton density being given by

$$\mathcal{H} = \frac{1}{2}\left[\frac{1}{\varepsilon_0}\boldsymbol{\Pi}^2 + \frac{1}{\mu_0}(\nabla\times\boldsymbol{A})^2\right]. \tag{2.30}$$

Recalling (2.23), we may equivalently use

$$\mathcal{H} = \frac{1}{2}\left[\frac{1}{\varepsilon_0}\boldsymbol{\Pi}^2 + \frac{1}{\mu_0}\,\mathrm{tr}\Big(\boldsymbol{A}\overleftarrow{\nabla}\cdot\nabla\boldsymbol{A}\Big)\right]. \tag{2.31}$$

Note that H has the physical meaning of being the energy of the (purely transverse) electromagnetic field,

$$H = \frac{1}{2}\int \mathrm{d}^3r\,\left[\varepsilon_0\boldsymbol{E}^2 + \frac{1}{\mu_0}\boldsymbol{B}^2\right]. \tag{2.32}$$

The associated Hamilton equations

$$\dot{\boldsymbol{A}} = \frac{\delta H}{\delta\boldsymbol{\Pi}}, \qquad \dot{\boldsymbol{\Pi}} = -\frac{\delta H}{\delta\boldsymbol{A}} \tag{2.33}$$

are equivalent to both the original dynamical equation (2.20) and the Lagrange equation (2.25), as can be easily seen by calculating

$$\dot{\boldsymbol{A}} = \frac{\delta H}{\delta\boldsymbol{\Pi}} = \frac{\partial\mathcal{H}}{\partial\boldsymbol{\Pi}} = \frac{1}{\varepsilon_0}\boldsymbol{\Pi}, \tag{2.34}$$

$$\dot{\boldsymbol{\Pi}} = -\frac{\delta H}{\delta\boldsymbol{A}} = -\frac{\partial\mathcal{H}}{\partial\boldsymbol{A}} + \nabla\cdot\frac{\partial\mathcal{H}}{\partial\nabla\boldsymbol{A}} = \frac{1}{\mu_0}\Delta\boldsymbol{A}. \tag{2.35}$$

We cast the Hamilton equations may be cast into an alternative form which can later be transferred to the quantum case by means of the correspondence principle. To that end, we introduce the Poisson bracket of two fields f and g according to

$$\{f, g\} = \int \mathrm{d}^3r\,\left(\frac{\delta f}{\delta\boldsymbol{A}}\cdot\frac{\delta g}{\delta\boldsymbol{\Pi}} - \frac{\delta f}{\delta\boldsymbol{\Pi}}\cdot\frac{\delta g}{\delta\boldsymbol{A}}\right). \tag{2.36}$$

When calculating the Poisson brackets of the canonically conjugate fields $\boldsymbol{A}$ and $\boldsymbol{\Pi}$, the fact that these quantities are transverse has to be taken into account. By writing

$$\boldsymbol{A}(\boldsymbol{r}) = \int \mathrm{d}^3r'\,\boldsymbol{\delta}^{\perp}(\boldsymbol{r}-\boldsymbol{r}')\cdot\boldsymbol{A}(\boldsymbol{r}'), \tag{2.37}$$

$$\boldsymbol{\Pi}(\boldsymbol{r}) = \int \mathrm{d}^3r'\,\boldsymbol{\delta}^{\perp}(\boldsymbol{r}-\boldsymbol{r}')\cdot\boldsymbol{\Pi}(\boldsymbol{r}'), \tag{2.38}$$

one finds

$$\frac{\delta A_i(\boldsymbol{r})}{\delta A_j(\boldsymbol{r}')} = \frac{\delta\Pi_i(\boldsymbol{r})}{\delta\Pi_j(\boldsymbol{r}')} = \delta^{\perp}_{ij}(\boldsymbol{r}-\boldsymbol{r}'), \qquad \frac{\delta A_i(\boldsymbol{r})}{\delta\Pi_j(\boldsymbol{r}')} = \frac{\delta\Pi_i(\boldsymbol{r})}{\delta A_j(\boldsymbol{r}')} = 0, \tag{2.39}$$

so the canonical Poisson brackets are given by

$$\{\boldsymbol{A}(\boldsymbol{r}), \boldsymbol{A}(\boldsymbol{r}')\} = \{\boldsymbol{\Pi}(\boldsymbol{r}), \boldsymbol{\Pi}(\boldsymbol{r}')\} = \mathbf{0}\,, \tag{2.40}$$

$$\{\boldsymbol{A}(\boldsymbol{r}), \boldsymbol{\Pi}(\boldsymbol{r}')\} = \boldsymbol{\delta}^{\perp}(\boldsymbol{r}-\boldsymbol{r}')\,. \tag{2.41}$$

Using (2.39), the definition of Poisson brackets leads to a more compact form for the Hamilton equations,

$$\dot{\boldsymbol{A}} = \{\boldsymbol{A}, H\}, \qquad \dot{\boldsymbol{\Pi}} = \{\boldsymbol{\Pi}, H\}\,. \tag{2.42}$$

More generally, the time derivative of any physical quantity f, which is a functional of $\boldsymbol{A}$ and $\boldsymbol{\Pi}$, is given by

$$\dot{f} = \{f, H\}\,, \tag{2.43}$$

as can be easily verified by using the chain rule

$$\dot{f} = \int \mathrm{d}^3 r \left(\frac{\delta f}{\delta \boldsymbol{A}} \cdot \dot{\boldsymbol{A}} + \frac{\delta f}{\delta \boldsymbol{\Pi}} \cdot \dot{\boldsymbol{\Pi}} \right) \tag{2.44}$$

together with the Hamilton equation (2.33). As we will see in Sect. 2.2, the fundamental Poisson brackets (2.40) and (2.41), the Hamilton equation (2.42) and the dynamical equation (2.43) remain valid in the quantum case with slight modifications.

2.1.2 Electrodynamics in the Presence of Charged Particles

Next, we introduce point particles α with charges q_α and masses m_α at positions $\boldsymbol{r}_\alpha$. They give rise to a charge density

$$\rho(\boldsymbol{r}) = \sum_\alpha q_\alpha \delta(\boldsymbol{r}-\boldsymbol{r}_\alpha) \tag{2.45}$$

and a current density

$$\boldsymbol{j}(\boldsymbol{r}) = \sum_\alpha q_\alpha \dot{\boldsymbol{r}}_\alpha \delta(\boldsymbol{r}-\boldsymbol{r}_\alpha) \tag{2.46}$$

which obey the continuity equation

$$\dot{\rho} + \nabla \cdot \boldsymbol{j} = 0\,. \tag{2.47}$$

Charge and current density act as sources for the electromagnetic field, so that two of the Maxwell equations, namely the Gauss law and the Ampère law, become inhomogeneous:

$$\varepsilon_0 \nabla \cdot \boldsymbol{E} = \rho \,, \tag{2.48}$$

$$\frac{1}{\mu_0} \nabla \times \boldsymbol{B} - \varepsilon_0 \dot{\boldsymbol{E}} = \boldsymbol{j} \,. \tag{2.49}$$

Introducing scalar and vector potentials (2.5) and (2.6) for the electromagnetic field, the homogeneous Maxwell equations (2.2) and (2.3) are again automatically fulfilled, while the inhomogeneous ones lead to

$$-\varepsilon_0 \Delta\phi - \varepsilon_0 \nabla \cdot \dot{\boldsymbol{A}} = \rho \,, \tag{2.50}$$

$$\varepsilon_0 \nabla\dot{\phi} + \varepsilon_0 \ddot{\boldsymbol{A}} - \frac{1}{\mu_0} \Delta\boldsymbol{A} + \frac{1}{\mu_0} \nabla(\nabla \cdot \boldsymbol{A}) = \boldsymbol{j} \,. \tag{2.51}$$

As before, we employ the Coulomb gauge $\nabla \cdot \boldsymbol{A} = 0$ which results in an effective decoupling of the two equations. The first one simplifies to the Poisson equation

$$-\varepsilon_0 \Delta\phi = \rho \tag{2.52}$$

which can easily be integrated by making use of the delta-function representation (2.16):

$$\phi(\boldsymbol{r}) = \int \mathrm{d}^3 r' \, \frac{\rho(\boldsymbol{r}')}{4\pi\varepsilon_0 |\boldsymbol{r} - \boldsymbol{r}'|} = \sum_\alpha \frac{q_\alpha}{4\pi\varepsilon_0 |\boldsymbol{r} - \boldsymbol{r}_\alpha|} \,. \tag{2.53}$$

In Coulomb gauge, the scalar potential ϕ is thus just the electrostatic Coulomb potential associated with the particles. Combining the Poisson equation with the continuity equation, we find

$$\nabla \cdot (\varepsilon_0 \nabla\dot{\phi} - \boldsymbol{j}) = 0 \,, \tag{2.54}$$

so that

$$\varepsilon_0 \nabla\dot{\phi} = \boldsymbol{j}^{\parallel} \,; \tag{2.55}$$

hence the vector potential is subject to the inhomogeneous Helmholtz equation

$$\varepsilon_0 \ddot{\boldsymbol{A}} - \frac{1}{\mu_0} \Delta\boldsymbol{A} = \boldsymbol{j}^{\perp} \,. \tag{2.56}$$

In Coulomb gauge, the vector potential thus only depends on the transverse current density, hence the alternative name transverse gauge.

So far, we have only considered the influence of the particles on the electromagnetic field. Conversely, the electromagnetic field acts on charged particles via Lorentz forces

$$\boldsymbol{F}_\alpha = q_\alpha [\boldsymbol{E}(\boldsymbol{r}_\alpha) + \dot{\boldsymbol{r}}_\alpha \times \boldsymbol{B}(\boldsymbol{r}_\alpha)] \,, \tag{2.57}$$

where by use of definitions (2.45) and (2.46), the total Lorentz force acting on the particles can be written as

$$\boldsymbol{F}=\sum_{\alpha}\boldsymbol{F}_{\alpha}=\int \mathrm{d}^3r\,(\rho\boldsymbol{E}+\boldsymbol{j}\times\boldsymbol{B})\,. \tag{2.58}$$

The Newton equations describing the non-relativistic motion of the individual particles under the influence of the Lorentz force are given by

$$m_{\alpha}\ddot{\boldsymbol{r}}_{\alpha}=\boldsymbol{F}_{\alpha}=q_{\alpha}[\boldsymbol{E}(\boldsymbol{r}_{\alpha})+\dot{\boldsymbol{r}}_{\alpha}\times\boldsymbol{B}(\boldsymbol{r}_{\alpha})]\,. \tag{2.59}$$

Upon introducing the potentials (2.5) and (2.6) for the electromagnetic field and using (2.53) for the Coulomb potential, one obtains

$$m_{\alpha}\ddot{\boldsymbol{r}}_{\alpha}=q_{\alpha}\left\{\sum_{\beta\neq\alpha}\frac{q_{\beta}(\boldsymbol{r}_{\alpha}-\boldsymbol{r}_{\beta})}{4\pi\varepsilon_0|\boldsymbol{r}_{\alpha}-\boldsymbol{r}_{\beta}|^3}-\dot{\boldsymbol{A}}(\boldsymbol{r}_{\alpha})+\dot{\boldsymbol{r}}_{\alpha}\times[\boldsymbol{\nabla}\times\boldsymbol{A}(\boldsymbol{r}_{\alpha})]\right\}. \tag{2.60}$$

Note that the electric field associated with the Coulomb potential (2.53) contains contributions from all particles, so the Lorentz force (2.57) or (2.58) on a particle α contains an unphysical divergent self-force associated with the action of the particle's own electrostatic field. By writing out the Coulomb forces explicitly in (2.60), we have been able to remove the self-force by discarding the term $\beta=\alpha$.

In contrast to the free-space case, we now have a coupled system of equations of motion for the dynamical degrees of freedom of the field, $\boldsymbol{A}$, and the particles, $\boldsymbol{r}_{\alpha}$. Note that the scalar potential ϕ has been completely eliminated, it does not play the role of a dynamical degree of freedom. As in the free-space case, we reformulate the dynamical equations within the context of a Lagrangian/Hamiltonian framework in order to facilitate quantisation of the system. According to the minimal coupling scheme, the interaction of the particles with the transverse electromagnetic field may be implemented by means of a term $\int \mathrm{d}^3r\;\boldsymbol{j}\cdot\boldsymbol{A}$, so that the non-relativistic Lagrangian of the system is given by

$$L=\frac{1}{2}\sum_{\alpha}m_{\alpha}\dot{\boldsymbol{r}}_{\alpha}^2-\sum_{\alpha\neq\beta}\frac{q_{\alpha}q_{\beta}}{8\pi\varepsilon_0|\boldsymbol{r}_{\alpha}-\boldsymbol{r}_{\beta}|}+\frac{1}{2}\int\mathrm{d}^3r\left[\varepsilon_0\dot{\boldsymbol{A}}^2-\frac{1}{\mu_0}(\boldsymbol{\nabla}\times\boldsymbol{A})^2\right]$$
$$+\int\mathrm{d}^3r\;\boldsymbol{j}\cdot\boldsymbol{A}\,. \tag{2.61}$$

The correctness of this Lagrangian can be easily verified by deriving the associated Lagrange equations. We calculate

$$\frac{\delta L}{\delta \boldsymbol{A}} = \frac{1}{\mu_0} \Delta \boldsymbol{A} + \boldsymbol{j}^{\perp}$$
$$= \frac{\mathrm{d}}{\mathrm{d}t} \frac{\delta L}{\delta \dot{\boldsymbol{A}}} = \varepsilon_0 \ddot{\boldsymbol{A}}\,, \tag{2.62}$$

showing that the Lagrange equation (2.25) for the vector potential $\boldsymbol{A}$ is equivalent to the Helmholtz equation (2.56). The Lagrange equations

$$\frac{\partial L}{\partial \boldsymbol{r}_\alpha} = \frac{\mathrm{d}}{\mathrm{d}t} \frac{\partial L}{\partial \dot{\boldsymbol{r}}_\alpha} \tag{2.63}$$

for the particle positions $\boldsymbol{r}_\alpha$ are found to be equivalent to the Newton equation (2.60) by calculating

$$\frac{\partial L}{\partial \boldsymbol{r}_\alpha} = q_\alpha \nabla \boldsymbol{A}(\boldsymbol{r}_\alpha) \cdot \dot{\boldsymbol{r}}_\alpha + \sum_{\beta \neq \alpha} \frac{q_\alpha q_\beta (\boldsymbol{r}_\alpha - \boldsymbol{r}_\beta)}{4\pi\varepsilon_0 |\boldsymbol{r}_\alpha - \boldsymbol{r}_\beta|^3}$$
$$= \frac{\mathrm{d}}{\mathrm{d}t} \frac{\partial L}{\partial \dot{\boldsymbol{r}}_\alpha} = m_\alpha \ddot{\boldsymbol{r}}_\alpha + q_\alpha \dot{\boldsymbol{A}}(\boldsymbol{r}_\alpha) + q_\alpha \dot{\boldsymbol{r}}_\alpha \cdot \nabla \boldsymbol{A}(\boldsymbol{r}_\alpha)\,, \tag{2.64}$$

where definition (2.46) of the current density has been recalled.

According to the Lagrangian (2.61), the canonically conjugate momentum associated with the vector potential $\boldsymbol{A}$ is given by

$$\boldsymbol{\Pi} = \frac{\delta L}{\delta \dot{\boldsymbol{A}}} = \varepsilon_0 \dot{\boldsymbol{A}} = -\varepsilon_0 \boldsymbol{E}^{\perp}\,, \tag{2.65}$$

whereas the canonically conjugate momenta of the particles are given by

$$\boldsymbol{p}_\alpha = \frac{\delta L}{\delta \dot{\boldsymbol{r}}_\alpha} = m_\alpha \dot{\boldsymbol{r}}_\alpha + q_\alpha \boldsymbol{A}(\boldsymbol{r}_\alpha)\,. \tag{2.66}$$

The non-relativistic Hamiltonian of the system can thus be constructed by applying a Legendre transformation

$$H = \sum_\alpha \boldsymbol{p}_\alpha \cdot \dot{\boldsymbol{r}}_\alpha + \int \mathrm{d}^3 r\, \boldsymbol{\Pi} \cdot \dot{\boldsymbol{A}} - L \tag{2.67}$$

and eliminating $\dot{\boldsymbol{r}}_\alpha$ and $\dot{\boldsymbol{A}}$ in favour of $\boldsymbol{p}_\alpha$ and $\boldsymbol{\Pi}$. This results in

$$H = \sum_\alpha \frac{[\boldsymbol{p}_\alpha - q_\alpha \boldsymbol{A}(\boldsymbol{r}_\alpha)]^2}{2m_\alpha} + \sum_{\alpha \neq \beta} \frac{q_\alpha q_\beta}{8\pi\varepsilon_0 |\boldsymbol{r}_\alpha - \boldsymbol{r}_\beta|}$$
$$+ \frac{1}{2} \int \mathrm{d}^3 r \left[\frac{1}{\varepsilon_0} \boldsymbol{\Pi}^2 + \frac{1}{\mu_0} (\nabla \times \boldsymbol{A})^2 \right]. \tag{2.68}$$

Comparing this with the Hamiltonian of the particles and the electromagnetic field without interactions (obtained by letting $q_\alpha \to 0$),

$$H = \sum_\alpha \frac{\boldsymbol{p}_\alpha^2}{2m_\alpha} + \frac{1}{2}\int \mathrm{d}^3r \left[\frac{1}{\varepsilon_0}\boldsymbol{\Pi}^2 + \frac{1}{\mu_0}(\boldsymbol{\nabla}\times\boldsymbol{A})^2\right], \tag{2.69}$$

we note that the minimal coupling scheme is equivalent to making the replacement $\boldsymbol{p}_\alpha \mapsto \boldsymbol{p}_\alpha - q_\alpha \boldsymbol{A}(\boldsymbol{r}_\alpha)$ and adding a Coulomb interaction term. The full Hamiltonian again has the physical meaning of being the total energy of the system,

$$H = \frac{1}{2}\sum_\alpha m_\alpha \dot{\boldsymbol{r}}_\alpha^2 + \sum_{\alpha\neq\beta}\frac{q_\alpha q_\beta}{8\pi\varepsilon_0|\boldsymbol{r}_\alpha - \boldsymbol{r}_\beta|} + \frac{1}{2}\int \mathrm{d}^3r \left[\varepsilon_0\big(\boldsymbol{E}^\perp\big)^2 + \frac{1}{\mu_0}\boldsymbol{B}^2\right], \tag{2.70}$$

where the first term is the kinetic energy of the particles, the second one their Coulomb energy and the last term is the energy of the transverse part of the electromagnetic field. By combining (2.45), (2.52) and (2.53) and integrating by parts, one can write

$$\sum_{\alpha\neq\beta}\frac{q_\alpha q_\beta}{8\pi\varepsilon_0|\boldsymbol{r}_\alpha - \boldsymbol{r}_\beta|} = \frac{1}{2}\int \mathrm{d}^3r\,\rho\phi = -\frac{1}{2}\int \mathrm{d}^3r\,\varepsilon_0\phi\Delta\phi = \frac{1}{2}\int \mathrm{d}^3r\,\varepsilon_0\big(\boldsymbol{E}^\parallel\big)^2, \tag{2.71}$$

so the Coulomb energy may alternatively be regarded as the energy associated with the longitudinal part of the electromagnetic field, $\boldsymbol{E}^\parallel = -\boldsymbol{\nabla}\phi$. In this picture, the total energy of the system is given by

$$H = \frac{1}{2}\sum_\alpha m_\alpha \dot{\boldsymbol{r}}_\alpha^2 + \frac{1}{2}\int \mathrm{d}^3r \left[\varepsilon_0\boldsymbol{E}^2 + \frac{1}{\mu_0}\boldsymbol{B}^2\right] \tag{2.72}$$

(note that integrals over mixed longitudinal/transverse vector fields vanish), where the second term is the energy of the whole electromagnetic field. It is worth mentioning that by rewriting the electrostatic interaction in terms of the Coulomb potential (or the longitudinal part of the electric field, respectively), we have introduced divergent self-energies associated with the interaction of each particle with its own electrostatic field. Such contributions are not present when writing out the Coulomb interaction explicitly as in (2.70) and excluding the terms $\alpha = \beta$, in close analogy to the exclusion of self-forces from the Lorentz force in (2.60) discussed above.

The Hamiltonian gives rise to Hamilton equations for both the electromagnetic field and the particles. The Hamilton equation (2.33) for the transverse part of the electromagnetic field are equivalent to the respective Lagrange equation (2.25) as well as the original Helmholtz equation (2.56):

$$\dot{\boldsymbol{A}} = \frac{\delta H}{\delta \boldsymbol{\Pi}} = \frac{1}{\varepsilon_0}\boldsymbol{\Pi}, \tag{2.73}$$

$$\dot{\boldsymbol{\Pi}} = -\frac{\delta H}{\delta \boldsymbol{A}} = \frac{1}{\mu_0} \Delta \boldsymbol{A} + \sum_\alpha \frac{q_\alpha}{m_\alpha} [\boldsymbol{p}_\alpha - q_\alpha \boldsymbol{A}(\boldsymbol{r}_\alpha)] \cdot \boldsymbol{\delta}^\perp(\boldsymbol{r} - \boldsymbol{r}_\alpha)\,. \tag{2.74}$$

The Hamilton equations for the particles

$$\dot{\boldsymbol{r}}_\alpha = \frac{\partial H}{\partial \boldsymbol{p}_\alpha}\,, \qquad \dot{\boldsymbol{p}}_\alpha = -\frac{\partial H}{\partial \boldsymbol{r}_\alpha} \tag{2.75}$$

are in turn equivalent to the respective Lagrange equation (2.63) and the Newton equation (2.59), as is seen from

$$\dot{\boldsymbol{r}}_\alpha = \frac{\partial H}{\partial \boldsymbol{p}_\alpha} = \frac{\boldsymbol{p}_\alpha - q_\alpha \boldsymbol{A}(\boldsymbol{r}_\alpha)}{m_\alpha}\,, \tag{2.76}$$

$$\begin{aligned} \dot{\boldsymbol{p}}_\alpha &= -\frac{\partial H}{\partial \boldsymbol{r}_\alpha} \\ &= \sum_{\beta \neq \alpha} \frac{q_\alpha q_\beta (\boldsymbol{r}_\alpha - \boldsymbol{r}_\beta)}{4\pi\varepsilon_0 |\boldsymbol{r}_\alpha - \boldsymbol{r}_\beta|^3} + \frac{q_\alpha}{m_\alpha} [\boldsymbol{p}_\alpha - q_\alpha \boldsymbol{A}(\boldsymbol{r}_\alpha)] \cdot [\boldsymbol{\nabla} \boldsymbol{A}(\boldsymbol{r}_\alpha)]\,. \end{aligned} \tag{2.77}$$

In order to reformulate the Hamilton equations in terms of the Poisson bracket, its definition must be generalised to account for all dynamical degrees of freedom of the system:

$$\begin{aligned} \{f, g\} = &\sum_\alpha \left(\frac{\partial f}{\partial \boldsymbol{r}_\alpha} \cdot \frac{\partial g}{\partial \boldsymbol{p}_\alpha} - \frac{\partial f}{\partial \boldsymbol{p}_\alpha} \cdot \frac{\partial g}{\partial \boldsymbol{r}_\alpha} \right) \\ &+ \int \mathrm{d}^3 r \left(\frac{\delta f}{\delta \boldsymbol{A}} \cdot \frac{\delta g}{\delta \boldsymbol{\Pi}} - \frac{\delta f}{\delta \boldsymbol{\Pi}} \cdot \frac{\delta g}{\delta \boldsymbol{A}} \right). \end{aligned} \tag{2.78}$$

The fundamental Poisson brackets then read

$$\{\boldsymbol{r}_\alpha, \boldsymbol{r}_\beta\} = \{\boldsymbol{p}_\alpha, \boldsymbol{p}_\beta\} = \boldsymbol{0}\,, \tag{2.79}$$

$$\{\boldsymbol{r}_\alpha, \boldsymbol{p}_\beta\} = \delta_{\alpha\beta} \boldsymbol{I} \tag{2.80}$$

for the particle variables, while the Poisson brackets (2.40) and (2.41) for the field variables remain valid. Obviously, mixed Poisson brackets of particle and field variables vanish. With the aid of the Poisson bracket, the Hamilton equations can be presented in the compact form

$$\dot{\boldsymbol{r}}_\alpha = \{\boldsymbol{p}_\alpha, H\}\,, \qquad \dot{\boldsymbol{p}}_\alpha = \{\boldsymbol{r}_\alpha, H\}\,, \tag{2.81}$$

$$\dot{\boldsymbol{A}} = \{\boldsymbol{A}, H\}\,, \qquad \dot{\boldsymbol{\Pi}} = \{\boldsymbol{\Pi}, H\}\,. \tag{2.82}$$

The dynamics of an arbitrary physical quantity f depending on the fundamental variables can be expressed in terms of the generalised Poisson bracket according to (2.42), as can be seen by applying the chain rule

$$\dot{f} = \sum_{\alpha} \left(\frac{\partial f}{\partial \boldsymbol{r}_{\alpha}} \cdot \dot{\boldsymbol{r}}_{\alpha} + \frac{\partial f}{\partial \boldsymbol{p}_{\alpha}} \cdot \dot{\boldsymbol{p}}_{\alpha} \right) + \int \mathrm{d}^3 r \left(\frac{\delta f}{\delta \boldsymbol{A}} \cdot \dot{\boldsymbol{A}} + \frac{\delta f}{\delta \boldsymbol{\Pi}} \cdot \dot{\boldsymbol{\Pi}} \right) \tag{2.83}$$

together with the Hamilton equations (2.33) and (2.75).

We have thus formulated the dynamics of the coupled system of electromagnetic field and charged particles in a suitable form in terms of Poisson brackets, which can conveniently be carried over to the quantum case by means of the correspondence principle. Before discussing this in Sect. 2.2, we need to generalise our theory to allow for the presence of macroscopic media.

2.1.3 Electrodynamics in Media

Magnetoelectric media, usually present in the form of one or more macroscopic bodies, may be thought of as an aggregate of a very large number of mutually bound charged particles. In principle, the microscopic description of the particles interacting with the electromagnetic field described in the previous section remains valid. In most cases of interest however, the number of particles and hence also the number of coupled equations is enormously large[2] and the system becomes practically unsolvable. Fortunately, it is in most cases sufficient to describe the gross influence of these particles on the electromagnetic field in an effective, macroscopic theory, as will be outlined in the following.

Let us first concentrate on the case of no free charges, so that only the bound charges contained in the magnetoelectric media are present. Denoting the internal charge and current densities associated with these charges by ρ_{in} and $\boldsymbol{j}_{\mathrm{in}}$, the inhomogeneous Maxwell equations take the form

$$\varepsilon_0 \boldsymbol{\nabla} \cdot \boldsymbol{E} = \rho_{\mathrm{in}} \,, \tag{2.84}$$

$$\frac{1}{\mu_0} \boldsymbol{\nabla} \times \boldsymbol{B} - \varepsilon_0 \dot{\boldsymbol{E}} = \boldsymbol{j}_{\mathrm{in}} \,. \tag{2.85}$$

Assuming the internal charge and current densities to obey the continuity equation

$$\dot{\rho}_{\mathrm{in}} + \boldsymbol{\nabla} \cdot \boldsymbol{j}_{\mathrm{in}} = 0 \,, \tag{2.86}$$

one can cast these equations into a homogeneous form by introducing a polarisation $\boldsymbol{P}$ and a magnetisation $\boldsymbol{M}$. Defining the polarisation according to

$$\rho_{\mathrm{in}} = -\boldsymbol{\nabla} \cdot \boldsymbol{P} \,, \tag{2.87}$$

the Gauss law (2.84) can be rewritten as

[2] For instance, 12 g of carbon contain $(6+1) \times N_{\mathrm{A}} \approx 4 \times 10^{24}$ particles.

$$\nabla \cdot (\varepsilon_0 \boldsymbol{E} + \boldsymbol{P}) = 0\,. \tag{2.88}$$

The continuity equation implies that

$$\nabla \cdot (\boldsymbol{j}_{\text{in}} - \dot{\boldsymbol{P}}) = 0\,, \tag{2.89}$$

so the quantity in brackets can be written as the curl of a vector field—the aforementioned magnetisation—hence[3]

$$\boldsymbol{j}_{\text{in}} = \dot{\boldsymbol{P}} + \nabla \times \boldsymbol{M}\,. \tag{2.90}$$

Upon substitution of this relation, the Ampère law (2.85) reads

$$\nabla \times \left(\frac{1}{\mu_0}\boldsymbol{B} - \boldsymbol{M}\right) - \frac{\partial}{\partial t}(\varepsilon_0 \boldsymbol{E} + \boldsymbol{P}) = \boldsymbol{0}\,. \tag{2.91}$$

By introducing electric excitation $\boldsymbol{D}$ and magnetic excitation $\boldsymbol{H}$ according to[4]

$$\boldsymbol{D} = \varepsilon_0 \boldsymbol{E} + \boldsymbol{P}\,, \tag{2.92}$$

$$\boldsymbol{H} = \frac{1}{\mu_0}\boldsymbol{B} - \boldsymbol{M}\,, \tag{2.93}$$

we can write the Gauss and the Ampère laws in their well-known forms

$$\nabla \cdot \boldsymbol{D} = 0\,, \tag{2.94}$$

$$\nabla \times \boldsymbol{H} - \dot{\boldsymbol{D}} = \boldsymbol{0}\,. \tag{2.95}$$

So far, we have only formally simplified our set of equations, because the behaviour of the bound charges (now represented by $\boldsymbol{P}$ and $\boldsymbol{M}$) under the influence of the electromagnetic field is still given by an enormously large number of equations of the type (2.60). We now introduce the simplifying assumption that these charges may be described by some equilibrium arrangement which is only weakly perturbed by the presence of an electromagnetic field. Assuming the response of the charges to this perturbance to be linear and causal, the constitutive relations for the media is given by the Langevin equations[5]

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[3] Note that (2.87) and (2.90) do not uniquely define $\boldsymbol{P}$ and $\boldsymbol{M}$. For instance, the relations remain valid when making the replacements $\boldsymbol{P} \to \boldsymbol{P} + \nabla \times \boldsymbol{f}$, $\boldsymbol{M} \to \boldsymbol{M} - \dot{\boldsymbol{f}} + \nabla\Lambda$ for any vector field $\boldsymbol{f}$ and scalar field Λ.

[4] Alternatively, $\boldsymbol{D}$ is often called displacement field and $\boldsymbol{H}$ is referred to as magnetic field.

[5] A recently proposed new derivation of the macroscopic Maxwell equations results in an alternative form governed by a single constitutive relation between $\boldsymbol{j}_{\text{in}}$ and $\boldsymbol{A}$ [1].

$$\boldsymbol{P}(\boldsymbol{r},t)=\varepsilon_0\int_{-\infty}^{\infty}\mathrm{d}\tau\int\mathrm{d}^3r'\,\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}',\tau)\cdot\boldsymbol{E}(\boldsymbol{r}',t-\tau)+\boldsymbol{P}_\mathrm{N}(\boldsymbol{r},t)\,, \tag{2.96}$$

$$\boldsymbol{M}(\boldsymbol{r},t)=\frac{1}{\mu_0}\int_{-\infty}^{\infty}\mathrm{d}\tau\int\mathrm{d}^3r'\,\boldsymbol{\zeta}(\boldsymbol{r},\boldsymbol{r}',\tau)\cdot\boldsymbol{B}(\boldsymbol{r}',t-\tau)+\boldsymbol{M}_\mathrm{N}(\boldsymbol{r},t)\,. \tag{2.97}$$

As expected for an effective, statistical description of a large number of particles, polarisation and magnetisation have reactive and random parts. The reactive part is given by a linear response to the applied electromagnetic field, with the respective response functions being the electric susceptibility $\boldsymbol{\chi}$ and the magnetic susceptibilities $\boldsymbol{\zeta}$ of the medium. Causality requires that

$$\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}',\tau)=\mathbf{0}\,,\quad \boldsymbol{\zeta}(\boldsymbol{r},\boldsymbol{r}',\tau)=\mathbf{0}\qquad\text{for } |\boldsymbol{r}-\boldsymbol{r}'|>c\tau\,. \tag{2.98}$$

In other words, $\boldsymbol{\chi}$ and $\boldsymbol{\zeta}$ are retarded response functions; the reactive part of polarisation and magnetisation at a given instant only depends on previous, not on future influences. The random parts are given by noise polarisation $\boldsymbol{P}_\mathrm{N}$ and noise magnetisation $\boldsymbol{M}_\mathrm{N}$ which account for the fluctuations occurring in the medium. Note that the noise terms vanish on the classical average,

$$\langle\boldsymbol{P}_\mathrm{N}\rangle_\mathrm{cl}=\mathbf{0}\,,\qquad \langle\boldsymbol{M}_\mathrm{N}\rangle_\mathrm{cl}=\mathbf{0}\,. \tag{2.99}$$

In complete analogy to (2.87) and (2.90) above, noise polarisation and magnetisation are related to the noise charge density

$$\rho_\mathrm{N}=-\boldsymbol{\nabla}\cdot\boldsymbol{P}_\mathrm{N} \tag{2.100}$$

and noise current density

$$\boldsymbol{j}_\mathrm{N}=\dot{\boldsymbol{P}}_\mathrm{N}+\boldsymbol{\nabla}\times\boldsymbol{M}_\mathrm{N}\,, \tag{2.101}$$

with the respective continuity equation

$$\dot{\rho}_\mathrm{N}+\boldsymbol{\nabla}\cdot\boldsymbol{j}_\mathrm{N}=0 \tag{2.102}$$

being automatically fulfilled.

For simplicity, we will assume in the following that the medium response is local

$$\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}',\tau)=\boldsymbol{\chi}(\boldsymbol{r},t-t')\delta(\boldsymbol{r}-\boldsymbol{r}')\,, \tag{2.103}$$

$$\boldsymbol{\zeta}(\boldsymbol{r},\boldsymbol{r}',\tau)=\boldsymbol{\zeta}(\boldsymbol{r},t-t')\delta(\boldsymbol{r}-\boldsymbol{r}') \tag{2.104}$$

and isotropic

$$\boldsymbol{\chi}(\boldsymbol{r}, t - t') = \chi(\boldsymbol{r}, t - t')\boldsymbol{I}\,, \tag{2.105}$$

$$\boldsymbol{\zeta}(\boldsymbol{r}, t - t') = \zeta(\boldsymbol{r}, t - t')\boldsymbol{I}\,. \tag{2.106}$$

The constitutive relations are then given by

$$\boldsymbol{P}(\boldsymbol{r}, t) = \varepsilon_0 \int_{-\infty}^{\infty} \mathrm{d}\tau\, \chi(\boldsymbol{r}, \tau)\boldsymbol{E}(\boldsymbol{r}, t - \tau) + \boldsymbol{P}_\mathrm{N}(\boldsymbol{r}, t)\,, \tag{2.107}$$

$$\boldsymbol{M}(\boldsymbol{r}, t) = \frac{1}{\mu_0} \int_{-\infty}^{\infty} \mathrm{d}\tau\, \zeta(\boldsymbol{r}, \tau)\boldsymbol{B}(\boldsymbol{r}, t - \tau) + \boldsymbol{M}_\mathrm{N}(\boldsymbol{r}, t)\,, \tag{2.108}$$

with the χ and ζ fulfilling the causality requirement

$$\chi(\boldsymbol{r}, \tau) = 0\,, \quad \zeta(\boldsymbol{r}, \tau) = 0 \quad \text{for } \tau < 0\,. \tag{2.109}$$

Polarisation and magnetisation depend on the history of the electromagnetic field. This integral time-dependence can be disentangled by working in the Fourier space. We introduce the Fourier transform of an arbitrary function f according to

$$\underline{f}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}t\, f(t)\mathrm{e}^{\mathrm{i}\omega t}\,, \tag{2.110}$$

with the inverse relations being given by

$$f(t) = \int_{-\infty}^{\infty} \mathrm{d}\omega\, \underline{f}(\omega)\mathrm{e}^{-\mathrm{i}\omega t} = \int_{0}^{\infty} \mathrm{d}\omega\, \underline{f}(\omega)\mathrm{e}^{-\mathrm{i}\omega t} + \mathrm{C.c.} \tag{2.111}$$

[note that $\underline{f}^*(\omega) = \underline{f}(-\omega^*)$ for real $f(t)$]. As a consequence of the convolution theorem, the constitutive relations in Fourier space take the much simpler form

$$\underline{\boldsymbol{P}}(\boldsymbol{r}, \omega) = \varepsilon_0 \chi(\boldsymbol{r}, \omega)\underline{\boldsymbol{E}}(\boldsymbol{r}, \omega) + \underline{\boldsymbol{P}}_\mathrm{N}(\boldsymbol{r}, \omega)\,, \tag{2.112}$$

$$\underline{\boldsymbol{M}}(\boldsymbol{r}, \omega) = \frac{\zeta(\boldsymbol{r}, \omega)}{\mu_0}\underline{\boldsymbol{B}}(\boldsymbol{r}, \omega) + \underline{\boldsymbol{M}}_\mathrm{N}(\boldsymbol{r}, \omega) \tag{2.113}$$

where

$$\chi(\boldsymbol{r},\omega)=2\pi\underline{\chi}(\boldsymbol{r},\omega)=\int\limits_{-\infty}^{\infty}\mathrm{d}\tau\,\chi(\boldsymbol{r},\tau)\mathrm{e}^{\mathrm{i}\omega\tau}\,, \tag{2.114}$$

$$\zeta(\boldsymbol{r},\omega)=2\pi\underline{\zeta}(\boldsymbol{r},\omega)=\int\limits_{-\infty}^{\infty}\mathrm{d}\tau\,\zeta(\boldsymbol{r},\tau)\mathrm{e}^{\mathrm{i}\omega\tau}\,. \tag{2.115}$$

The response functions giving the reactive parts of polarisation and magnetisation are thus $\varepsilon_0\chi$ and ζ/μ_0, respectively. According to fluctuation–dissipation theorem of classical statistical physics [2, 3], the fluctuation spectrum of a physical quantity f can be related to the imaginary part of the respective response function. Denoting classical fluctuations by

$$\Delta f = f - \langle f\rangle_{\mathrm{cl}}\,, \tag{2.116}$$

this theorem reads in our case

$$\langle\Delta\boldsymbol{P}_{\mathrm{N}}(\boldsymbol{r},\omega)\Delta\boldsymbol{P}^{*}_{\mathrm{N}}(\boldsymbol{r}',\omega')\rangle_{\mathrm{cl}}=\frac{k_{\mathrm{B}}T}{\pi\omega}\,\varepsilon_0\mathrm{Im}\,\chi(\boldsymbol{r},\omega)\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\delta(\omega-\omega')\,, \tag{2.117}$$

$$\langle\Delta\boldsymbol{M}_{\mathrm{N}}(\boldsymbol{r},\omega)\Delta\boldsymbol{M}^{*}_{\mathrm{N}}(\boldsymbol{r}',\omega')\rangle_{\mathrm{cl}}=\frac{k_{\mathrm{B}}T}{\pi\omega}\,\frac{\mathrm{Im}\,\zeta(\boldsymbol{r},\omega)}{\mu_0}\,\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\delta(\omega-\omega') \tag{2.118}$$

(k_{B}: Boltzmann constant, T: temperature). The imaginary parts of the response functions determine whether the medium is absorbing or amplifying. We will only consider absorbing media for which χ and ζ have a positive imaginary part. The above equations thus reveal the intrinsic connection between fluctuations and absorption: Fluctuations are necessarily present in any absorbing system at non-zero temperature. Note that the classical fluctuations vanish in the zero-temperature limit.

Before introducing polarisation and magnetisation, we had described the internal charges by their charge and current densities. It is therefore instructive to note that these quantities can also separated into reactive part and random parts. Combining the respective results for the polarisation (2.112) and magnetisation (2.113) into the defining relations (2.87), (2.90) and using (2.100) and (2.101), one easily finds

$$\underline{\rho}_{\mathrm{in}}(\boldsymbol{r},\omega)=-\varepsilon_0\nabla\cdot\left[\chi(\boldsymbol{r},\omega)\underline{\boldsymbol{E}}(\boldsymbol{r},\omega)\right]+\underline{\rho}_{\mathrm{N}}(\boldsymbol{r},\omega)\,, \tag{2.119}$$

$$\underline{\boldsymbol{j}}_{\mathrm{in}}(\boldsymbol{r},\omega)=-\mathrm{i}\omega\varepsilon_0\chi(\boldsymbol{r},\omega)\underline{\boldsymbol{E}}(\boldsymbol{r},\omega)+\frac{1}{\mu_0}\nabla\times\left[\zeta(\boldsymbol{r},\omega)\underline{\boldsymbol{B}}(\boldsymbol{r},\omega)\right]+\underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r},\omega)\,. \tag{2.120}$$

The response of the internal charge and current densities to the electromagnetic field is slightly more involved than that of the polarisation and magnetisation given above, while the random terms are simply the noise charge and current densities introduced earlier.

The constitutive relations complete our system of equations for the electromagnetic field. In Fourier space, this system is given by the Maxwell equations

$$\nabla \cdot \underline{\boldsymbol{D}} = 0\,, \tag{2.121}$$

$$\nabla \cdot \underline{\boldsymbol{B}} = 0\,, \tag{2.122}$$

$$\nabla \times \underline{\boldsymbol{E}} - \mathrm{i}\omega \underline{\boldsymbol{B}} = \boldsymbol{0}\,, \tag{2.123}$$

$$\nabla \times \underline{\boldsymbol{H}} + \mathrm{i}\omega \underline{\boldsymbol{D}} = \boldsymbol{0} \tag{2.124}$$

together with the constitutive relations, which upon combining (2.92), (2.93), (2.112) and (2.113) can be written in the form

$$\underline{\boldsymbol{D}} = \varepsilon_0 \varepsilon \underline{\boldsymbol{E}} + \underline{\boldsymbol{P}}_{\mathrm{N}}\,, \tag{2.125}$$

$$\underline{\boldsymbol{H}} = \frac{1}{\mu_0 \mu} \underline{\boldsymbol{B}} - \underline{\boldsymbol{M}}_{\mathrm{N}}\,. \tag{2.126}$$

Here, we have introduced the electric permittivity

$$\varepsilon(\boldsymbol{r}, \omega) = 1 + \chi(\boldsymbol{r}, \omega) \tag{2.127}$$

and the magnetic permeability

$$\mu(\boldsymbol{r}, \omega) = \frac{1}{1 - \zeta(\boldsymbol{r}, \omega)} \tag{2.128}$$

of the medium.

Before solving the equations for the electromagnetic field, it is worthwhile discussing the introduced permittivity and permeability in little more detail. We first provide some examples. According to the Drude–Lorentz model, the response of a homogeneous dielectric $\varepsilon(\boldsymbol{r}, \omega) = \varepsilon(\omega)$ may be derived by considering a uniform density η of electrons bound in harmonic potentials. The displacement $\boldsymbol{x}$ of one such electron from its equilibrium position under the influence of an electric field is governed by the equation

$$m_e\big(\ddot{\boldsymbol{x}} + \gamma \dot{\boldsymbol{x}} + \omega_{\mathrm{T}}^2 \boldsymbol{x}\big) = -e\boldsymbol{E} \tag{2.129}$$

where m_e and e denote the electron mass and charge, respectively, $\gamma > 0$ is the damping constant accounting for absorption and ω_{T} is the transverse resonance frequency of the electron. This equation can be easily solved in Fourier space, leading to

$$\underline{\boldsymbol{x}} = -\frac{e\underline{\boldsymbol{E}}}{m_e(\omega_{\mathrm{T}}^2 - \omega^2 - \mathrm{i}\gamma\omega)}\,. \tag{2.130}$$

The polarisation associated with all bound electrons is hence given by

$$\underline{\boldsymbol{P}} = -e\underline{\boldsymbol{x}}\eta = \frac{e^2\eta}{m_e} \frac{1}{(\omega_\mathrm{T}^2 - \omega^2 - \mathrm{i}\gamma\omega)} \underline{\boldsymbol{E}} . \tag{2.131}$$

Comparison with (2.112) shows that the permittivity (2.127) of the dielectric can be described by the single-resonance form

$$\varepsilon(\omega) = 1 + \frac{e^2\eta}{\varepsilon_0 m_e} \frac{1}{(\omega_\mathrm{T}^2 - \omega^2 - \mathrm{i}\gamma\omega)} = 1 + \frac{\omega_\mathrm{P}^2}{(\omega_\mathrm{T}^2 - \omega^2 - \mathrm{i}\gamma\omega)} . \tag{2.132}$$

Here, we have introduced the plasma frequency $\omega_\mathrm{P} = e\sqrt{\eta/(\varepsilon_0 m_e)}$ which characterises the behaviour of a plasma of unbound and undamped electrons. Note that the positive damping constant γ present for an absorbing medium ensures that the imaginary part of $\varepsilon(\omega)$ is positive for all real ω. An analogous single-resonance form is commonly used for the permeability μ.

The electric response of a metal may be addressed by using the Drude model, where the focus is on the conduction electrons. With the motion of these unbound electrons being described by

$$m_e\big(\ddot{\boldsymbol{x}} + \gamma\dot{\boldsymbol{x}}\big) = -e\boldsymbol{E} , \tag{2.133}$$

the permittivity can be given in the form

$$\varepsilon(\omega) = 1 - \frac{\omega_\mathrm{P}^2}{\omega(\omega + \mathrm{i}\gamma)} = 1 + \mathrm{i}\,\frac{\sigma(\omega)}{\omega} \tag{2.134}$$

where we have introduced the conductivity

$$\sigma(\omega) = \frac{\omega_\mathrm{P}^2}{\gamma - \mathrm{i}\omega} . \tag{2.135}$$

These simple models can be generalised by combining the response of bound and conduction electrons and by considering more than one resonance. Alternatively, ε and μ for real materials can be obtained from measurements rather than theoretical models.

Independently of the specific form obtained from either a model or measured data, permittivity and permeability exhibit a few general properties. They are both complex-valued functions of position and frequency. The frequency-dependence is usually referred to as dispersion, because it leads to the well-known dispersion of light (i.e., the spreading of a wave packet as it passes through a dispersive medium). As noted in the example, absorption is associated with a positive imaginary part of ε and μ. We require all present media to be absorbing, so that

$$\operatorname{Im}\varepsilon(\boldsymbol{r},\omega) > 0\,, \qquad \operatorname{Im}\mu(\boldsymbol{r},\omega) > 0 \tag{2.136}$$

for real frequencies. From the fact that $\chi(\boldsymbol{r},\tau)$ and $\zeta(\boldsymbol{r},\tau)$ are real, it follows by virtue of (2.114) and (2.115) that the Schwarz reflection principle holds for the susceptibilities, $\chi^*(\boldsymbol{r},\omega) = \chi(\boldsymbol{r},-\omega^*)$, $\zeta^*(\boldsymbol{r},\omega) = \zeta(\boldsymbol{r},-\omega^*)$, and hence also for the permittivity and permeability

$$\varepsilon^*(\boldsymbol{r},\omega) = \varepsilon(\boldsymbol{r},-\omega^*), \qquad \mu^*(\boldsymbol{r},\omega) = \mu(\boldsymbol{r},-\omega^*)\,. \tag{2.137}$$

Further general properties of ε and μ follow from the causality conditions (2.109). Together with definitions (2.114) and (2.115), they imply that $\chi(\boldsymbol{r},\omega)$ and $\zeta(\boldsymbol{r},\omega)$ and hence also $\varepsilon(\boldsymbol{r},\omega)$ and $\mu(\boldsymbol{r},\omega)$ are analytic functions of ω in the upper half of the complex frequency plane including the real axis, provided that $\chi(\boldsymbol{r},\tau)$ and $\zeta(\boldsymbol{r},\tau)$ are finite and vanish in the limit $\tau\to\infty$. This is true for all (magneto)dielectrics, but not for metals. In the latter case $\chi(\boldsymbol{r},\tau)$ has a non-vanishing long-time asymptote, leading to a simple pole of $\varepsilon(\boldsymbol{r},\omega)$ at $\omega = 0$ (as already seen for the Drude model). For metals, we can thus write

$$\varepsilon(\boldsymbol{r},\omega) = \varepsilon_{\text{bound}}(\boldsymbol{r},\omega) + \frac{\mathrm{i}\sigma(\boldsymbol{r},\omega)}{\omega} \tag{2.138}$$

where both the component of the permittivity associated with the bound charges, $\varepsilon_{\text{bound}}(\boldsymbol{r},\omega)$, and the conductivity associated with the free charges, $\sigma(\boldsymbol{r},\omega)$, are again analytic in the whole upper half of the complex frequency plane. Another consequence of causality is the asymptotic high-frequency behaviour [4]

$$\varepsilon(\boldsymbol{r},\omega) - 1 = \mathrm{O}\big(1/\omega^2\big)\,, \qquad \mu(\boldsymbol{r},\omega) - 1 = \mathrm{O}\big(1/\omega^2\big)\,. \tag{2.139}$$

Together with the analyticity, these asymptotes imply the Kramers–Kronig relations which relate the real and imaginary parts of $\varepsilon(\boldsymbol{r},\omega)$ or $\mu(\boldsymbol{r},\omega)$ on the real frequency axis. They read [4]

$$\operatorname{Re}\varepsilon(\boldsymbol{r},\omega) = 1 + \frac{2}{\pi}\mathcal{P}\int_0^\infty \mathrm{d}\omega'\,\frac{\omega'\operatorname{Im}\varepsilon(\boldsymbol{r},\omega')}{\omega'^2-\omega^2}\,, \tag{2.140}$$

$$\operatorname{Im}\varepsilon(\boldsymbol{r},\omega) = -\frac{2\omega}{\pi}\mathcal{P}\int_0^\infty \mathrm{d}\omega'\,\frac{\operatorname{Re}\varepsilon(\boldsymbol{r},\omega')-1}{\omega'^2-\omega^2} \tag{2.141}$$

($\mathcal{P}$: principal value) and

$$\operatorname{Re}\mu(\boldsymbol{r},\omega) = 1 + \frac{2}{\pi}\mathcal{P}\int_0^\infty \mathrm{d}\omega'\,\frac{\omega'\operatorname{Im}\mu(\boldsymbol{r},\omega')}{\omega'^2-\omega^2}\,, \tag{2.142}$$

$$\mathrm{Im}\,\mu(\boldsymbol{r},\omega) = -\frac{2\omega}{\pi}\,\mathcal{P}\int_0^\infty \mathrm{d}\omega'\,\frac{\mathrm{Re}\,\mu(\boldsymbol{r},\omega')-1}{\omega'^2-\omega^2} \tag{2.143}$$

for (magneto)dielectrics, whereas for a metals, (2.140) and (2.141) need to be replaced with

$$\mathrm{Re}\,\varepsilon(\boldsymbol{r},\omega) = 1+\frac{1}{\pi}\,\mathcal{P}\int_{-\infty}^\infty \mathrm{d}\omega'\,\frac{\mathrm{Im}\,\varepsilon(\boldsymbol{r},\omega')}{\omega'-\omega}\,, \tag{2.144}$$

$$\mathrm{Im}\,\varepsilon(\boldsymbol{r},\omega) = \frac{\sigma(\boldsymbol{r},0)}{\omega}-\frac{1}{\pi}\,\mathcal{P}\int_{-\infty}^\infty \mathrm{d}\omega'\,\frac{\mathrm{Re}\,\varepsilon(\boldsymbol{r},\omega')-1}{\omega'-\omega}\,. \tag{2.145}$$

After this little digression, let us construct a solution to the Maxwell equations. In contrast to the free-space case where all quantities were expressed in terms of the vector potential $\boldsymbol{A}$, it is here more convenient to formulate a dynamical equation for $\boldsymbol{E}$. Substituting the Faraday law (2.123) together with the constitutive relations (2.125) and (2.126) into the Ampère law (2.124), we find that the electric field is subject to an inhomogeneous Helmholtz equation

$$\left[\nabla\times\frac{1}{\mu}\nabla\times\;-\frac{\omega^2}{c^2}\,\varepsilon\right]\underline{\boldsymbol{E}} = \mathrm{i}\mu_0\omega\,\underline{\boldsymbol{j}}_{\mathrm{N}}\,, \tag{2.146}$$

with the source term being given by the noise current density

$$\underline{\boldsymbol{j}}_{\mathrm{N}} = -\mathrm{i}\omega\underline{\boldsymbol{P}}_{\mathrm{N}}+\nabla\times\underline{\boldsymbol{M}}_{\mathrm{N}}\,. \tag{2.147}$$

A formal solution to this equation can be given by

$$\underline{\boldsymbol{E}}(\boldsymbol{r},\omega) = \mathrm{i}\mu_0\omega\int \mathrm{d}^3r'\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega) \tag{2.148}$$

where the Green's tensor $\boldsymbol{G}$ is a solution to

$$\left[\nabla\times\frac{1}{\mu(\boldsymbol{r},\omega)}\nabla\times\;-\frac{\omega^2}{c^2}\,\varepsilon(\boldsymbol{r},\omega)\right]\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega) = \boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\,. \tag{2.149}$$

The Green's tensor is uniquely defined by this partial differential equation together with the boundary condition

$$\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\to\boldsymbol{0}\quad\text{for } |\boldsymbol{r}-\boldsymbol{r}'|\to\infty \tag{2.150}$$

for any given arrangement of absorbing bodies characterised by $\varepsilon(\boldsymbol{r},\omega)$ and $\mu(\boldsymbol{r},\omega)$, provided that the strict inequalities $\mathrm{Im}\,\varepsilon(\boldsymbol{r},\omega) > 0$ and $\mathrm{Im}\,\mu(\boldsymbol{r},\omega) > 0$ hold. The general properties of the permittivity and permeability stated above imply some useful general properties of the Green's tensor. It is an analytic function of frequency in the whole upper half of the complex plane. Furthermore one can show that the Schwarz reflection principle

$$\boldsymbol{G}^*(\boldsymbol{r},\boldsymbol{r}',\omega) = \boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',-\omega^*)\,, \tag{2.151}$$

Onsager reciprocity

$$\boldsymbol{G}^{\mathrm{T}}(\boldsymbol{r},\boldsymbol{r}',\omega) = \boldsymbol{G}(\boldsymbol{r}',\boldsymbol{r},\omega) \tag{2.152}$$

and the integral relation

$$\begin{aligned}\int \mathrm{d}^3 s\, \biggl\{&-\frac{\mathrm{Im}\,\mu(\boldsymbol{s},\omega)}{|\mu(\boldsymbol{s},\omega)|^2}\Bigl[\boldsymbol{G}(\boldsymbol{r},\boldsymbol{s},\omega)\times\overleftarrow{\nabla}_{\boldsymbol{s}}\Bigr]\cdot\bigl[\nabla_{\boldsymbol{s}}\times\boldsymbol{G}^*(\boldsymbol{s},\boldsymbol{r}',\omega)\bigr]\\ &+\frac{\omega^2}{c^2}\,\mathrm{Im}\,\varepsilon(\boldsymbol{s},\omega)\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{s},\omega)\cdot\boldsymbol{G}^*(\boldsymbol{s},\boldsymbol{r}',\omega)\biggr\} = \mathrm{Im}\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\end{aligned} \tag{2.153}$$

are valid [5, 6] (see App. B.1).

The solution (2.148) for the electric field can be used to construct explicit expressions for the other relevant fields. From (2.123), we conclude that the magnetic field must be given by

$$\underline{\boldsymbol{B}}(\boldsymbol{r},\omega) = \frac{1}{\mathrm{i}\omega}\nabla\times\underline{\boldsymbol{E}}(\boldsymbol{r},\omega) = \mu_0\int \mathrm{d}^3 r'\,\nabla\times\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega)\,, \tag{2.154}$$

while (2.125) and (2.126) imply that the electric and magnetic excitations read

$$\underline{\boldsymbol{D}}(\boldsymbol{r},\omega) = \mathrm{i}\,\frac{\omega}{c^2}\,\varepsilon(\boldsymbol{r},\omega)\int \mathrm{d}^3 r'\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega) + \underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r},\omega)\,, \tag{2.155}$$

$$\underline{\boldsymbol{H}}(\boldsymbol{r},\omega) = \frac{1}{\mu(\boldsymbol{r},\omega)}\int \mathrm{d}^3 r'\,\nabla\times\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega) - \underline{\boldsymbol{M}}_{\mathrm{N}}(\boldsymbol{r},\omega)\,. \tag{2.156}$$

The Maxwell equations (2.123) and (2.124) as well as the constitutive relations (2.125) and (2.126) are thus fulfilled by construction. Our solution further automatically obeys the remaining two Maxwell equations: The validity of (2.122) follows directly from (2.154). The Gauss law (2.121) can be verified by calculating

$$\begin{aligned}\nabla\cdot\underline{\boldsymbol{D}}(\boldsymbol{r},\omega) &= \mathrm{i}\,\frac{\omega}{c^2}\,\nabla\cdot\varepsilon(\boldsymbol{r},\omega)\int \mathrm{d}^3 r'\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega) + \nabla\cdot\underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r},\omega)\\ &= \frac{1}{\mathrm{i}\omega}\nabla\cdot\bigl[\underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r},\omega) + \mathrm{i}\omega\underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r},\omega)\bigr] = 0\,,\end{aligned} \tag{2.157}$$

where we have used $\underline{\boldsymbol{j}}_{\mathrm{N}} = -\mathrm{i}\omega\underline{\boldsymbol{P}}_{\mathrm{N}} + \nabla \times \underline{\boldsymbol{M}}_{\mathrm{N}}$ as well as the relation

$$\frac{\omega^2}{c^2}\,\nabla\cdot\varepsilon(\boldsymbol{r},\omega)\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega) = -\nabla\cdot\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\,. \tag{2.158}$$

The latter can be found by taking the divergence of (2.149).

Comparing the electromagnetic field in free space with that in the presence of media, we note two important differences. Firstly, the electromagnetic field in free space obeys a homogeneous Helmholtz equation, while the field in the presence of media is subject to an inhomogeneous Helmholtz equation with the noise current density acting as a source. Secondly, in contrast to the free-space case, the electromagnetic field in the presence of media is a fluctuating quantity. These field fluctuations are an immediate consequence of the source fluctuations. For example, the fluctuation spectrum of the electric field can be inferred from the fluctuation spectra (2.117) and (2.118) of the noise polarisation and magnetisation by using the representation (2.148) of the electric field:

$$\begin{aligned}
&\langle\Delta\boldsymbol{E}(\boldsymbol{r},\omega)\Delta\boldsymbol{E}^*(\boldsymbol{r}',\omega')\rangle_{\mathrm{cl}}\\
&= \mu_0^2\omega\omega'\int\mathrm{d}^3s\int\mathrm{d}^3s'\,\Big\{\omega\omega'\boldsymbol{G}(\boldsymbol{r},\boldsymbol{s},\omega)\cdot\langle\Delta\boldsymbol{P}_{\mathrm{N}}(\boldsymbol{s},\omega)\Delta\boldsymbol{P}_{\mathrm{N}}^*(\boldsymbol{s}',\omega')\rangle_{\mathrm{cl}}\cdot\boldsymbol{G}^*(\boldsymbol{s}',\boldsymbol{r}',\omega)\\
&\quad -\Big[\boldsymbol{G}(\boldsymbol{r},\boldsymbol{s},\omega)\times\overleftarrow{\nabla}_{\boldsymbol{s}}\Big]\cdot\langle\Delta\boldsymbol{M}_{\mathrm{N}}(\boldsymbol{s},\omega)\Delta\boldsymbol{M}_{\mathrm{N}}^*(\boldsymbol{s}',\omega')\rangle_{\mathrm{cl}}\cdot\big[\nabla_{\boldsymbol{s}'}\times\boldsymbol{G}^*(\boldsymbol{s}',\boldsymbol{r}',\omega)\big]\Big\}\\
&= \frac{\mu_0 k_{\mathrm{B}}T}{\pi}\,\omega\,\mathrm{Im}\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\delta(\omega-\omega')\,,
\end{aligned} \tag{2.159}$$

where $\underline{\boldsymbol{j}}_{\mathrm{N}} = -\mathrm{i}\omega\underline{\boldsymbol{P}}_{\mathrm{N}} + \nabla \times \underline{\boldsymbol{M}}_{\mathrm{N}}$ and the integral relation (2.153) for the Green's tensor have been used.

Next, consider the case where in addition to the internal charges comprised in the medium, free charges are also present, which give rise to additional charge and current densities ρ and $\boldsymbol{j}$ as given by (2.45) and (2.46). The inhomogeneous Maxwell equations are then given by

$$\varepsilon_0\nabla\cdot\boldsymbol{E} = \rho_{\mathrm{in}} + \rho\,, \tag{2.160}$$

$$\frac{1}{\mu_0}\nabla\times\boldsymbol{B} - \varepsilon_0\dot{\boldsymbol{E}} = \boldsymbol{j}_{\mathrm{in}} + \boldsymbol{j}\,. \tag{2.161}$$

In complete analogy to the case of no free charges, one can relate the charge and current densities ρ_{in} and $\boldsymbol{j}_{\mathrm{in}}$ of the internal charges to polarisation $\boldsymbol{P}$ and magnetisation $\boldsymbol{M}$ via (2.87) and (2.90) and then introduce electric and magnetic excitations $\boldsymbol{D}$ and $\boldsymbol{H}$ according to (2.92) and (2.93). Due to the presence of the free charges, the Gauss and the Ampère laws are now inhomogeneous:

$$\nabla \cdot \boldsymbol{D} = \rho \,, \tag{2.162}$$

$$\nabla \times \boldsymbol{H} - \dot{\boldsymbol{D}} = \boldsymbol{j} \,. \tag{2.163}$$

As before, we describe the behaviour of the internal charges under the influence of the electromagnetic field in an effective way by assuming the medium response to be linear, local and isotropic, so that the constitutive relations (2.107) and (2.108) hold. On the contrary, the motion of the free charges is described in an exact way by the Newton equations (2.59).

A solution for the electromagnetic field can again be constructed by working in the Fourier domain. The homogeneous Maxwell equations (2.122) and (2.123) as well as the constitutive relations (2.125) and (2.126) remain their form whereas the inhomogeneous Maxwell equations now read

$$\nabla \cdot \underline{\boldsymbol{D}} = \underline{\rho} \,, \tag{2.164}$$

$$\nabla \times \underline{\boldsymbol{H}} + \mathrm{i}\omega \underline{\boldsymbol{D}} = \underline{\boldsymbol{j}} \,. \tag{2.165}$$

Combining these equations, one obtains a Helmholtz equation for the electric field

$$\left[\nabla \times \frac{1}{\mu} \nabla \times \; - \frac{\omega^2}{c^2}\,\varepsilon\right] \underline{\boldsymbol{E}} = \mathrm{i}\mu_0\omega\big(\underline{\boldsymbol{j}}_{\mathrm{N}} + \underline{\boldsymbol{j}}\big) \,, \tag{2.166}$$

with the sources being given by both the noise current density and the current density associated with the free charges. The relevant electromagnetic fields can again be expressed in terms of the Green's tensor, where in slight generalisation of the case without free charges, one has

$$\underline{\boldsymbol{E}}(\boldsymbol{r},\omega) = \mathrm{i}\mu_0\omega \int \mathrm{d}^3r'\, \boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot[\underline{\boldsymbol{j}}(\boldsymbol{r}',\omega) + \underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega)] \,, \tag{2.167}$$

$$\underline{\boldsymbol{B}}(\boldsymbol{r},\omega) = \mu_0 \int \mathrm{d}^3r'\, \nabla \times \boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot[\underline{\boldsymbol{j}}(\boldsymbol{r}',\omega) + \underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega)] \,, \tag{2.168}$$

$$\underline{\boldsymbol{D}}(\boldsymbol{r},\omega) = \mathrm{i}\,\frac{\omega}{c^2}\,\varepsilon(\boldsymbol{r},\omega) \int \mathrm{d}^3r'\, \boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot[\underline{\boldsymbol{j}}(\boldsymbol{r}',\omega) + \underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega)] + \underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r},\omega) \,, \tag{2.169}$$

$$\underline{\boldsymbol{H}}(\boldsymbol{r},\omega) = \frac{1}{\mu(\boldsymbol{r},\omega)} \int \mathrm{d}^3r'\, \nabla \times \boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot[\underline{\boldsymbol{j}}(\boldsymbol{r}',\omega) + \underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}',\omega)] - \underline{\boldsymbol{M}}_{\mathrm{N}}(\boldsymbol{r},\omega) \,. \tag{2.170}$$

This solution satisfies Maxwell equations (2.123) and (2.165) as well as the constitutive Equations (2.125) and (2.125) by construction. The remaining two Maxwell equations also hold: Equation (2.122) follows immediately from (2.168) and the validity of (2.164) is seen from

$$
\begin{aligned}
&\nabla \cdot \underline{\boldsymbol{D}}(\boldsymbol{r}, \omega) \\
&= \mathrm{i}\, \frac{\omega}{c^2}\, \nabla \cdot \varepsilon(\boldsymbol{r}, \omega) \int \mathrm{d}^3 r'\, \boldsymbol{G}(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \big[\underline{\boldsymbol{j}}(\boldsymbol{r}', \omega) + \underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}', \omega)\big] + \nabla \cdot \underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r}, \omega) \\
&= \frac{1}{\mathrm{i}\omega} \nabla \cdot \big[\underline{\boldsymbol{j}}(\boldsymbol{r}', \omega) + \underline{\boldsymbol{j}}_{\mathrm{N}}(\boldsymbol{r}, \omega) + \mathrm{i}\omega \underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r}, \omega)\big] = \underline{\rho}(\boldsymbol{r}, \omega)
\end{aligned}
\tag{2.171}
$$

where we have used $\underline{\boldsymbol{j}}_{\mathrm{N}} = -\mathrm{i}\omega \underline{\boldsymbol{P}}_{\mathrm{N}} + \nabla \times \underline{\boldsymbol{M}}_{\mathrm{N}}$ and (2.158) as well as the continuity equation

$$
-\mathrm{i}\omega \underline{\rho} + \nabla \cdot \underline{\boldsymbol{j}} = 0 \tag{2.172}
$$

for the free charges.

Note that (2.167) can be used to infer the fluctuation spectrum of the electric field without referring to the fluctuations of polarisation and magnetisation. To see this, we write this equation in the form

$$
\underline{\boldsymbol{E}}(\boldsymbol{r}, \omega) = \mathrm{i}\mu_0 \omega \int \mathrm{d}^3 r'\, \boldsymbol{G}(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \underline{\boldsymbol{j}}(\boldsymbol{r}', \omega) + \underline{\boldsymbol{E}}_{\mathrm{N}}(\boldsymbol{r}, \omega)\,, \tag{2.173}
$$

where the fist term is the reactive part of the field and the second term is the random part. This equation is analogous to (2.96) where the roles of electric field and the charges are now reversed: While (2.96) describes the response of the internal charges to an applied electric field, the equation here characterises the response of the electric field to the free charges. To get a complete analogy, one has to introduce the polarisation associated with the free charges instead of their current density via a relation similar to (2.90). The response function hence being given by $\mu_0 \omega^2 \boldsymbol{G}$, the fluctuation–dissipation theorem [2, 3] implies

$$
\langle \Delta \boldsymbol{E}(\boldsymbol{r}, \omega) \Delta \boldsymbol{E}^*(\boldsymbol{r}', \omega') \rangle_{\mathrm{cl}} = \frac{k_{\mathrm{B}} T}{\pi \omega}\, \mu_0 \omega^2\, \mathrm{Im}\, \boldsymbol{G}(\boldsymbol{r}, \boldsymbol{r}', \omega) \delta(\omega - \omega')\,, \tag{2.174}
$$

in agreement with the previously derived (2.159).

2.1.4 Duality

In the absence of free charges, the electric and magnetic field quantities are governed by very similar laws. This can be exploited to determine magnetic field configurations without a calculation simply by comparison with the respective analogous electric field configuration and vice versa. This symmetry is known as the duality of electric and magnetic fields and the interchange of these fields constitutes a duality transformation. In the following, we will establish explicit prescriptions for duality transformations of the various fields and establish under which conditions duality is a valid symmetry. Duality will be very useful in the later chapters when studying dispersion forces.

The Maxwell equations in the absence of free charges or currents are homogeneous and may be grouped into two pairs of analogous equations

$$\nabla \cdot \boldsymbol{D} = 0\,, \tag{2.175}$$
$$\nabla \cdot \boldsymbol{B} = 0\,, \tag{2.176}$$

and

$$\nabla \times \boldsymbol{E} + \dot{\boldsymbol{B}} = \boldsymbol{0}\,, \tag{2.177}$$
$$\nabla \times \boldsymbol{H} - \dot{\boldsymbol{D}} = \boldsymbol{0} \tag{2.178}$$

where the relations between the fields and the excitations forms a third such pair,

$$\boldsymbol{D} = \varepsilon_0 \boldsymbol{E} + \boldsymbol{P}\,, \tag{2.179}$$
$$\boldsymbol{H} = \frac{1}{\mu_0}\boldsymbol{B} - \boldsymbol{M}\,. \tag{2.180}$$

Accordingly, we group the electromagnetic field quantities into six-component dual-pair vectors $(\boldsymbol{E}, Z_0\boldsymbol{H})^{\mathrm{T}}$ $(Z_0\boldsymbol{D}, \boldsymbol{B})^{\mathrm{T}}$ and $(Z_0\boldsymbol{P}, \mu_0\boldsymbol{M})^{\mathrm{T}}$ where the vacuum impedance $Z_0 = \sqrt{\mu_0/\varepsilon_0}$ has been introduced for dimensional reasons. In this dual-pair notation, the Maxwell equations assume the compact form

$$\nabla \cdot \begin{pmatrix} Z_0\boldsymbol{D} \\ \boldsymbol{B} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}\,, \tag{2.181}$$
$$\nabla \times \begin{pmatrix} \boldsymbol{E} \\ Z_0\boldsymbol{H} \end{pmatrix} + \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix} \begin{pmatrix} Z_0\dot{\boldsymbol{D}} \\ \dot{\boldsymbol{B}} \end{pmatrix} = \begin{pmatrix} \boldsymbol{0} \\ \boldsymbol{0} \end{pmatrix}; \tag{2.182}$$

and the relation between the fields and excitations reads

$$\begin{pmatrix} Z_0\boldsymbol{D} \\ \boldsymbol{B} \end{pmatrix} = \frac{1}{c}\begin{pmatrix} \boldsymbol{E} \\ Z_0\boldsymbol{H} \end{pmatrix} + \begin{pmatrix} Z_0\boldsymbol{P} \\ \mu_0\boldsymbol{M} \end{pmatrix}\,. \tag{2.183}$$

It is now immediately obvious that these equations are invariant with respect to a duality transformation

$$\boxed{\begin{pmatrix} \boldsymbol{x} \\ \boldsymbol{y} \end{pmatrix}^{\circledast} = \mathcal{D}(\theta)\begin{pmatrix} \boldsymbol{x} \\ \boldsymbol{y} \end{pmatrix}\,, \quad \text{with } \mathcal{D}(\theta) = \begin{pmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{pmatrix} \in \mathrm{SO}(2)} \tag{2.184}$$

being the most general real matrix that commutes with the matrix in (2.182), up to a global constant rescaling factor. Such a matrix $\mathcal{D}(\theta)$ is commonly know as a symplectic matrix. The transformation may be viewed as a rotation in the space of dual pairs ($0 \leq \theta < 2\pi$). The name of the duality transformation can be understood from the fact that in relativistic free-space electrodynamics, it mixes the electromagnetic-field tensor (with electric and magnetic fields as components) with its dual tensor (having magnetic and electric excitations as components) [7, 8]. Alternatively, the real-valued electromagnetic fields are often combined into complex Riemann–Silberstein vectors $\boldsymbol{x} + \mathrm{i}\boldsymbol{y}$ [9], in which case duality invariance manifests itself as a U(1) symmetry. In a more general, metric-free formulation of electrodynamics especially suited for curved spacetimes, the notion electric/magnetic reciprocity is used in place of duality [10].

In order for duality to be a valid symmetry transformation in the presence of magnetoelectric media, it must also be compatible with the constitutive relations

$$\underline{\boldsymbol{D}} = \varepsilon_0\varepsilon\underline{\boldsymbol{E}} + \underline{\boldsymbol{P}}_{\mathrm{N}}\,, \tag{2.185}$$

$$\underline{\boldsymbol{H}} = \frac{1}{\mu_0\mu}\underline{\boldsymbol{B}} - \underline{\boldsymbol{M}}_{\mathrm{N}}\,. \tag{2.186}$$

Recall that they are most conveniently formulated for the Fourier components of the fields which are complex-valued. This is why we use 6-vectors rather than the complex Riemann–Silberstein vectors; it would be difficult extract the electric and magnetic field components $\boldsymbol{x}$ and $\boldsymbol{y}$ from the latter. In dual-pair notation, the constitutive relations read

$$\begin{pmatrix} Z_0\underline{\boldsymbol{D}} \\ \underline{\boldsymbol{B}} \end{pmatrix} = \frac{1}{c}\begin{pmatrix} \varepsilon & 0 \\ 0 & \mu \end{pmatrix}\begin{pmatrix} \underline{\boldsymbol{E}} \\ Z_0\underline{\boldsymbol{H}} \end{pmatrix} + \begin{pmatrix} 1 & 0 \\ 0 & \mu \end{pmatrix}\begin{pmatrix} Z_0\underline{\boldsymbol{P}}_{\mathrm{N}} \\ \mu_0\underline{\boldsymbol{M}}_{\mathrm{N}} \end{pmatrix} \tag{2.187}$$

and hence their invariance under duality transformations requires that

$$\begin{aligned}\begin{pmatrix} \varepsilon^{\circledast} & 0 \\ 0 & \mu^{\circledast} \end{pmatrix} &= \mathcal{D}(\theta)\begin{pmatrix} \varepsilon & 0 \\ 0 & \mu \end{pmatrix}\mathcal{D}^{-1}(\theta) \\ &= \begin{pmatrix} \varepsilon\cos^2\theta + \mu\sin^2\theta & (\mu-\varepsilon)\sin\theta\cos\theta \\ (\mu-\varepsilon)\sin\theta\cos\theta & \varepsilon\sin^2\theta + \mu\cos^2\theta \end{pmatrix}\end{aligned} \tag{2.188}$$

and

$$\begin{pmatrix} \underline{\boldsymbol{P}}_{\mathrm{N}} \\ \underline{\boldsymbol{M}}_{\mathrm{N}}/c \end{pmatrix}^{\circledast} = \begin{pmatrix} \cos\theta & \mu\sin\theta \\ -(1/\mu^{\circledast})\sin\theta & (\mu/\mu^{\circledast})\cos\theta \end{pmatrix}\begin{pmatrix} \underline{\boldsymbol{P}}_{\mathrm{N}} \\ \underline{\boldsymbol{M}}_{\mathrm{N}}/c \end{pmatrix}. \tag{2.189}$$

The first of these conditions can only be fulfilled in two ways: It holds if the relative impedance of the media is equal to unity, $Z = \sqrt{\mu/\varepsilon} = 1$. In this case, which includes both free space and a perfect lens medium ($\varepsilon = \mu = -1$, cf. Sect. 4.1), duality is manifest as a continuous SO(2) symmetry of the electromagnetic field and one has $\varepsilon^{\circledast} = \mu^{\circledast} = \varepsilon$ as well as

$$\begin{pmatrix} \underline{\boldsymbol{P}}_{\mathrm{N}} \\ \underline{\boldsymbol{M}}_{\mathrm{N}}/c \end{pmatrix}^{\circledast} = \begin{pmatrix} \cos\theta & \varepsilon\sin\theta \\ -(1/\varepsilon)\sin\theta & \cos\theta \end{pmatrix}\begin{pmatrix} \underline{\boldsymbol{P}}_{\mathrm{N}} \\ \underline{\boldsymbol{M}}_{\mathrm{N}}/c \end{pmatrix}. \tag{2.190}$$

For media with a nontrivial impedance, (2.188) holds for $\theta = n\pi/2$ with $n \in \mathbb{Z}$ only. The presence of such media hence reduces the duality invariance from the full SO(2) group to a discrete $\mathbb{Z}_4$ symmetry with the four distinct members

$$\mathcal{D}_0 = \mathcal{I}, \quad \mathcal{D}_1 = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}, \quad \mathcal{D}_2 = -\mathcal{I}, \quad \mathcal{D}_3 = -\mathcal{D}_1, \tag{2.191}$$

where (2.188) and (2.189) imply the transformations

$$\begin{pmatrix} \varepsilon \\ \mu \end{pmatrix}^{\circledast} = \begin{pmatrix} \cos^2\theta & \sin^2\theta \\ \sin^2\theta & \cos^2\theta \end{pmatrix} \begin{pmatrix} \varepsilon \\ \mu \end{pmatrix}, \tag{2.192}$$

$$\begin{pmatrix} \underline{\boldsymbol{P}}_{\mathrm{N}} \\ \underline{\boldsymbol{M}}_{\mathrm{N}}/c \end{pmatrix}^{\circledast} = \begin{pmatrix} \cos\theta & \mu\sin\theta \\ -(1/\varepsilon)\sin\theta & \cos\theta \end{pmatrix} \begin{pmatrix} \underline{\boldsymbol{P}}_{\mathrm{N}} \\ \hat{\underline{\boldsymbol{M}}}_{\mathrm{N}}/c \end{pmatrix}. \tag{2.193}$$

The Maxwell equations in the absence of free charges and currents are thus invariant with respect to a duality transformation in one of its two forms. The same must hold for their unique explicit solutions (2.148), (2.154)–(2.156). Upon expressing the noise current density in terms of noise polarisation and magnetisation via (2.147), the latter can be written in the alternative forms

$$\begin{aligned} \underline{\boldsymbol{E}}(\boldsymbol{r}, \omega) = &-\frac{1}{\varepsilon_0} \int \mathrm{d}^{3r'}\, \boldsymbol{G}_{ee}(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r}', \omega) \\ &- Z_0 \int \mathrm{d}^3 r'\, \boldsymbol{G}_{em}(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \underline{\boldsymbol{M}}_{\mathrm{N}}(\boldsymbol{r}', \omega), \end{aligned} \tag{2.194}$$

$$\begin{aligned} \underline{\boldsymbol{B}}(\boldsymbol{r}, \omega) = &-Z_0 \int \mathrm{d}^3 r'\, \boldsymbol{G}_{me}(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r}', \omega) \\ &- \mu_0 \int \mathrm{d}^3 r'\, \boldsymbol{G}_{mm}(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \underline{\boldsymbol{M}}_{\mathrm{N}}(\boldsymbol{r}', \omega), \end{aligned} \tag{2.195}$$

$$\begin{aligned} \underline{\boldsymbol{D}}(\boldsymbol{r}, \omega) = &-\int \mathrm{d}^3 r' \left[\varepsilon(\boldsymbol{r}, \omega)\boldsymbol{G}_{ee}(\boldsymbol{r}, \boldsymbol{r}', \omega) - \boldsymbol{\delta}(\boldsymbol{r} - \boldsymbol{r}')\right] \cdot \underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r}', \omega) \\ &- \frac{\varepsilon(\boldsymbol{r}, \omega)}{c} \int \mathrm{d}^3 r'\, \boldsymbol{G}_{em}(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \underline{\boldsymbol{M}}_{\mathrm{N}}(\boldsymbol{r}', \omega), \end{aligned} \tag{2.196}$$

$$\begin{aligned} \underline{\boldsymbol{H}}(\boldsymbol{r}, \omega) = &-\frac{c}{\mu(\boldsymbol{r}, \omega)} \int \mathrm{d}^3 r'\, \boldsymbol{G}_{me}(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \underline{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r}', \omega) \\ &- \int \mathrm{d}^3 r' \left[\frac{\boldsymbol{G}_{mm}(\boldsymbol{r}, \boldsymbol{r}', \omega)}{\mu(\boldsymbol{r}, \omega)} + \boldsymbol{\delta}(\boldsymbol{r} - \boldsymbol{r}')\right] \cdot \underline{\boldsymbol{M}}_{\mathrm{N}}(\boldsymbol{r}', \omega), \end{aligned} \tag{2.197}$$

making the dual structure more apparent. Here, we have introduced the tensors

$$\boldsymbol{G}_{ee}(\boldsymbol{r},\boldsymbol{r}',\omega) = \frac{\mathrm{i}\omega}{c}\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\,\frac{\mathrm{i}\omega}{c}\,, \tag{2.198}$$

$$\boldsymbol{G}_{mm}(\boldsymbol{r},\boldsymbol{r}',\omega) = \nabla\times\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\times\overleftarrow{\nabla}'\,, \tag{2.199}$$

$$\boldsymbol{G}_{em}(\boldsymbol{r},\boldsymbol{r}',\omega) = \frac{\mathrm{i}\omega}{c}\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\times\overleftarrow{\nabla}'\,, \tag{2.200}$$

$$\boldsymbol{G}_{me}(\boldsymbol{r},\boldsymbol{r}',\omega) = \nabla\times\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\,\frac{\mathrm{i}\omega}{c}\,, \tag{2.201}$$

where $\boldsymbol{G}_{\lambda\lambda'}$ relates the induced electric and magnetic fields (for $\lambda = e, m$) to their two possible sources polarisation and magnetisation (for $\lambda' = e, m$). The duality invariance of these solutions can be used to determine the transformation behaviour of the Green's tensor. We concentrate on the duality transformation $\mathcal{D}_1$ which is a generator of the entire discrete $\mathbb{Z}_4$ group, i.e., the three other transformations can be generated by repeated application of $\mathcal{D}_1$. According to (2.192), we have ($\theta = \pi/2$) $\varepsilon^{\circledast} = \mu$ and $\mu^{\circledast} = \varepsilon$, so the dual Green's tensor is the solution to the differential equation

$$\left[\nabla\times\frac{1}{\varepsilon(\boldsymbol{r},\omega)}\nabla\times-\frac{\omega^2}{c^2}\,\mu(\boldsymbol{r},\omega)\right]\boldsymbol{G}^{\circledast}(\boldsymbol{r},\boldsymbol{r}',\omega) = \boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\,. \tag{2.202}$$

In order to relate it to the original Green's tensor, we apply the duality transformation to both sides of (2.194) and (2.195), where the dual Green's tensor appears on the right hand side of the transformed equations together with $\underline{\boldsymbol{P}}_{\mathrm{N}}^{\circledast}$, $\underline{\boldsymbol{M}}_{\mathrm{N}}^{\circledast}$, $\varepsilon^{\circledast}$ and $\mu^{\circledast}$. We express these fields and response functions as well as the fields on the left hand side in terms of the untransformed ones via (2.184), (2.192) and (2.193) ($\theta = \pi/2$). Finally, we expand the fields on the left hand side in terms of the $\underline{\boldsymbol{P}}_{\mathrm{N}}$, $\underline{\boldsymbol{M}}_{\mathrm{N}}$ by using (2.194)–(2.197). Comparing the coefficients on the two sides of the resulting equations, we find the transformation laws

$$\boldsymbol{G}_{ee}^{\circledast}(\boldsymbol{r},\boldsymbol{r}',\omega) = \frac{1}{\mu(\boldsymbol{r},\omega)}\,\boldsymbol{G}_{mm}(\boldsymbol{r},\boldsymbol{r}',\omega)\,\frac{1}{\mu(\boldsymbol{r}',\omega)} + \frac{1}{\mu(\boldsymbol{r},\omega)}\,\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\,, \tag{2.203}$$

$$\boldsymbol{G}_{mm}^{\circledast}(\boldsymbol{r},\boldsymbol{r}',\omega) = \varepsilon(\boldsymbol{r},\omega)\boldsymbol{G}_{ee}(\boldsymbol{r},\boldsymbol{r}',\omega)\varepsilon(\boldsymbol{r}',\omega) - \varepsilon(\boldsymbol{r},\omega)\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\,, \tag{2.204}$$

$$\boldsymbol{G}_{em}^{\circledast}(\boldsymbol{r},\boldsymbol{r}',\omega) = -\frac{1}{\mu(\boldsymbol{r},\omega)}\,\boldsymbol{G}_{me}(\boldsymbol{r},\boldsymbol{r}',\omega)\varepsilon(\boldsymbol{r}',\omega)\,, \tag{2.205}$$

$$\boldsymbol{G}_{me}^{\circledast}(\boldsymbol{r},\boldsymbol{r}',\omega) = -\varepsilon(\boldsymbol{r},\omega)\boldsymbol{G}_{em}(\boldsymbol{r},\boldsymbol{r}',\omega)\,\frac{1}{\mu(\boldsymbol{r}',\omega)}\,. \tag{2.206}$$

It can easily be seen that these laws are self-inverse; by applying them twice, one returns to the original Green's tensor (note that the roles of ε and μ have to be

Table 2.1 Dual-partner fields and response functions and their behaviour under the duality transformation $\mathcal{D}_1 = \mathcal{D}(\theta = \pi/2)$

Dual partners	Duality transformation	
$\boldsymbol{E}$, $\boldsymbol{H}$:	$\boldsymbol{E}^{\circledast} = Z_0\boldsymbol{H}$,	$\boldsymbol{H}^{\circledast} = -\boldsymbol{E}/Z_0$
$\boldsymbol{D}$, $\boldsymbol{B}$:	$\boldsymbol{D}^{\circledast} = \boldsymbol{B}/Z_0$,	$\boldsymbol{B}^{\circledast} = -Z_0\boldsymbol{D}$
$\boldsymbol{P}$, $\boldsymbol{M}$:	$\boldsymbol{P}^{\circledast} = \boldsymbol{M}/c$,	$\boldsymbol{M}^{\circledast} = -c\boldsymbol{P}$
$\boldsymbol{P}_{\mathrm{N}}$, $\boldsymbol{M}_{\mathrm{N}}$:	$\boldsymbol{P}_{\mathrm{N}}^{\circledast} = \mu\boldsymbol{M}_{\mathrm{N}}/c$,	$\boldsymbol{M}_{\mathrm{N}}^{\circledast} = -c\boldsymbol{P}_{\mathrm{N}}/\varepsilon$
ε, μ:	$\varepsilon^{\circledast} = \mu$,	$\mu^{\circledast} = \varepsilon$
$\boldsymbol{G}_{ee}$, $\boldsymbol{G}_{mm}$:	$\boldsymbol{G}_{ee}^{\circledast} = (1/\mu)\boldsymbol{G}_{mm}(1/\mu) + (1/\mu)\boldsymbol{\delta}$,	$\boldsymbol{G}_{mm}^{\circledast} = \varepsilon\boldsymbol{G}_{ee}\varepsilon - \varepsilon\boldsymbol{\delta}$
$\boldsymbol{G}_{em}$, $\boldsymbol{G}_{me}$:	$\boldsymbol{G}_{em}^{\circledast} = -(1/\mu)\boldsymbol{G}_{me}\varepsilon$,	$\boldsymbol{G}_{me}^{\circledast} = -\varepsilon\boldsymbol{G}_{em}(1/\mu)$

exchanged for the second transformation). By starting from (2.196) and (2.197) instead of (2.227) and (2.195), one would arrive at equivalent transformation laws for the Green's tensor.

The action of the duality transformation associated with $\mathcal{D}_1 = \mathcal{D}(\theta = \pi/2)$ on the various fields and response functions is summarised in Table 2.1. The transformation exchanges the two fields of each dual pair while in addition, one of them acquires a minus sign; the transformation therefore needs to be applied four times in order to return to the original state. On the contrary, the duality transformation is self-inverse when applied to the response functions.

As we have seen, duality in its discrete form with the transformation laws as given in Table 2.1 is an exact symmetry of the Maxwell equations in magnetoelectric media in the absence of free charges or currents. It will break down in the presence of charged systems like single electrons or ions where the Maxwell equations (2.175) and (2.178) become inhomogeneous, thus breaking the symmetry with the homogeneous Maxwell equations (2.176) and (2.177). Duality invariance can be recovered under specific circumstances if only electrically neutral systems are present which may be polarisable or magnetisable. In particular, duality is a useful symmetry of the dispersion forces between atoms and/or bodies in free space, as will be shown in Sects. 3.2, 4.4, and 5.3.

2.2 Field Quantisation in free Space

In a quantum theory, physical observables are represented by operators acting on an appropriate Hilbert space. For a system prepared in a state $|\psi\rangle$ of this Hilbert space, the quantum average of an observable $\hat{f}$ is given by

$$\langle \hat{f} \rangle = \langle \psi | \hat{f} | \psi \rangle \tag{2.207}$$

and its quantum fluctuations can be calculated according to

$$\langle(\Delta\hat{f})^2\rangle = \langle\hat{f}^2\rangle - \langle\hat{f}\rangle^2 \tag{2.208}$$

with

$$\Delta\hat{f} = \hat{f} - \langle\hat{f}\rangle . \tag{2.209}$$

Two operators $\hat{f}$ and $\hat{g}$ representing different observables do not necessarily commute, this is accounted for by introducing the commutator

$$[\hat{f},\hat{g}] = \hat{f}\hat{g} - \hat{g}\hat{f} . \tag{2.210}$$

According to the Heisenberg uncertainty principle, the fluctuations of two observables are related to their commutator by

$$\langle(\Delta\hat{f})^2\rangle\langle(\Delta\hat{g})^2\rangle \geq \frac{1}{4}\left|\langle[\hat{f},\hat{g}]\rangle\right|^2 , \tag{2.211}$$

showing that fluctuations necessarily occur when considering two non-commuting observables.

The quantum electromagnetic field is thus given by field operators $\hat{\boldsymbol{E}}$ and $\hat{\boldsymbol{B}}$ which represent probabilistic, fluctuating quantities; in contrast to the classical c-number fields $\boldsymbol{E}$ and $\boldsymbol{B}$. In spite of this fundamental difference, QED must resemble classical electrodynamics as closely as possible. Thus, we require that operators describing the quantum electromagnetic field in free space are also subject to the Maxwell equations

$$\boldsymbol{\nabla}\cdot\hat{\boldsymbol{E}} = 0 , \tag{2.212}$$

$$\boldsymbol{\nabla}\cdot\hat{\boldsymbol{B}} = 0 , \tag{2.213}$$

$$\boldsymbol{\nabla}\times\hat{\boldsymbol{E}} + \dot{\hat{\boldsymbol{B}}} = \boldsymbol{0} , \tag{2.214}$$

$$\boldsymbol{\nabla}\times\hat{\boldsymbol{B}} - \frac{1}{c^2}\dot{\hat{\boldsymbol{E}}} = \boldsymbol{0} , \tag{2.215}$$

so that the averages of the fluctuating quantum electromagnetic field behave in the same way as the non-fluctuating classical electromagnetic field.

As we will see, this requirement is automatically fulfilled when employing canonical quantisation, a procedure which at the same time renders the yet unknown commutators of the quantum electromagnetic field. Canonical quantisation starts from a Hamiltonian formulation of quantum electrodynamics, as was given at the end of Sect. 2.1.1. According to the correspondence principle, classical observables in this formulation may then simply be replaced by the corresponding operators, whereby classical Poisson brackets must be replaced by commutators

$$\{f,g\} \mapsto \frac{1}{\mathrm{i}\hbar}[\hat{f},\hat{g}] . \tag{2.216}$$

The vector potential of the electromagnetic field $\boldsymbol{A}$ and its canonically conjugate momentum $\boldsymbol{\Pi}$, which represent the dynamical degrees of freedom, thus become operator valued and the classical Poisson brackets (2.40) and (2.41) imply that the canonical equal-time commutation relations are given by

$$\left[\hat{\boldsymbol{A}}(\boldsymbol{r}), \hat{\boldsymbol{A}}(\boldsymbol{r}')\right] = \left[\hat{\boldsymbol{\Pi}}(\boldsymbol{r}), \hat{\boldsymbol{\Pi}}(\boldsymbol{r}')\right] = \mathbf{0}\,, \tag{2.217}$$

$$\left[\hat{\boldsymbol{A}}(\boldsymbol{r}), \hat{\boldsymbol{\Pi}}(\boldsymbol{r}')\right] = \mathrm{i}\hbar\boldsymbol{\delta}^{\perp}(\boldsymbol{r} - \boldsymbol{r}')\,. \tag{2.218}$$

The relation between these fundamental fields and the electromagnetic field is assumed to be completely analogous to the classical one. In Coulomb gauge, we have

$$\hat{\boldsymbol{E}} = \hat{\boldsymbol{E}}^{\perp} = -\frac{1}{\varepsilon_0}\hat{\boldsymbol{\Pi}}\,, \tag{2.219}$$

$$\hat{\boldsymbol{B}} = \nabla \times \hat{\boldsymbol{A}}\,, \tag{2.220}$$

recall (2.6), (2.18) and (2.27). Hence, the equal-time commutation relations of the electromagnetic field are given by

$$\left[\hat{\boldsymbol{E}}(\boldsymbol{r}), \hat{\boldsymbol{E}}(\boldsymbol{r}')\right] = \left[\hat{\boldsymbol{B}}(\boldsymbol{r}), \hat{\boldsymbol{B}}(\boldsymbol{r}')\right] = \mathbf{0}\,, \tag{2.221}$$

$$\left[\hat{\boldsymbol{E}}(\boldsymbol{r}), \hat{\boldsymbol{B}}(\boldsymbol{r}')\right] = \frac{\mathrm{i}\hbar}{\varepsilon_0}\nabla \times \boldsymbol{\delta}(\boldsymbol{r} - \boldsymbol{r}') \tag{2.222}$$

where we have recalled the property $\nabla \times \boldsymbol{\delta}^{\parallel}(\boldsymbol{r}) = \mathbf{0}$.

The classical Hamiltonian of the free electromagnetic field is replaced by the Hamilton operator

$$\hat{H} = \frac{1}{2}\int \mathrm{d}^3r \left[\frac{1}{\varepsilon_0}\hat{\boldsymbol{\Pi}}^2 + \frac{1}{\mu_0}(\nabla \times \hat{\boldsymbol{A}})^2\right], \tag{2.223}$$

which is the operator representing the energy of the (transverse) electromagnetic field

$$\hat{H} = \frac{1}{2}\int \mathrm{d}^3r \left[\varepsilon_0\hat{\boldsymbol{E}}^2 + \frac{1}{\mu_0}\hat{\boldsymbol{B}}^2\right]. \tag{2.224}$$

More importantly, the Hamiltonian governs the equations of motion in a similar way as in the classical theory. As implied by the correspondence principle, the dynamics of a quantum observable $\hat{f}$ is given not by the classical equation (2.43), but by the Heisenberg equation of motion

$$\dot{\hat{f}} = \frac{1}{\mathrm{i}\hbar}\left[\hat{f}, \hat{H}\right] \tag{2.225}$$

instead. When writing down the Heisenberg equations of motion for the canonically conjugate fields, we have to calculate the commutators of the Hamiltonian with these fields. As a direct consequence of the canonical commutation relations, the commutator of the Hamiltonian with respect to one of these fields is directly related to its derivative with respect the other one; this can be verified by repeated use of the rule

$$\left[\hat{a}, \hat{b}\hat{c}\right] = \hat{b}\left[\hat{a}, \hat{c}\right] + \left[\hat{a}, \hat{b}\right]\hat{c} \,. \tag{2.226}$$

The Heisenberg equations of motion for the canonically conjugate fields

$$\dot{\hat{\boldsymbol{A}}} = \frac{1}{\mathrm{i}\hbar}\left[\hat{\boldsymbol{A}}, \hat{H}\right] = \frac{\delta \hat{H}}{\delta \hat{\boldsymbol{\Pi}}} = \frac{1}{\varepsilon_0}\,\hat{\boldsymbol{\Pi}} \,, \tag{2.227}$$

$$\dot{\hat{\boldsymbol{\Pi}}} = \frac{1}{\mathrm{i}\hbar}\left[\hat{\boldsymbol{\Pi}}, \hat{H}\right] = -\frac{\delta \hat{H}}{\delta \hat{\boldsymbol{A}}} = \frac{1}{\mu_0}\,\Delta\hat{\boldsymbol{A}} \,. \tag{2.228}$$

are thus of exactly the same form as the classical Hamilton equations. As in the classical case, these equations are equivalent to the Helmholtz equation

$$\frac{1}{c^2}\ddot{\hat{\boldsymbol{A}}} - \Delta\hat{\boldsymbol{A}} = \boldsymbol{0} \,. \tag{2.229}$$

The Helmholtz equation, together with the definitions (2.219) and (2.220) implies that the Maxwell equations (2.212)–(2.215) for the quantum electromagnetic field in free space hold, as required.

We have thus successfully obtained a quantum theory for the electromagnetic field from classical electrodynamics by means of canonical quantisation. The use of a Hamiltonian formulation together with the correspondence principle has ensured that the quantum theory is very analogous to the classical theory.

2.3 Field Quantisation in Media

Canonical quantisation cannot be applied to the electromagnetic field in the presence of media, since a Hamiltonian formulation of the respective classical theory is not readily available. Instead, we employ three guiding principles in order to construct a quantum theory for the macroscopic electromagnetic field in the presence of media (macroscopic QED): (i) The equations of motion for the quantum electromagnetic field must be the same as the classical ones, i.e. the Maxwell equations together with the constitutive relations must hold. (ii) The quantum fluctuations of the electromagnetic field must obey the fluctuation–dissipation theorem, just as the classical fluctuations do. (iii) In a medium, one has to distinguish between the electric and magnetic fields and excitations. The latter contain contributions from the medium degrees of freedom via polarisation and magnetisation. The electric and magnetic fields, however, represent purely field degrees of freedom, so their commutation relations must be identical to those in free space. To summarise, we require the behaviour of the quantum macroscopic electromagnetic field to resemble that of the classical field as closely as possible while being consistent with the properties of the quantum electromagnetic field in free space derived in the previous section.

Let us begin by quantising the macroscopic electromagnetic field in the absence of free charges or currents. As in the free-space case, all classical fields must be replaced by operator-valued quantum observables, where according to requirement (i), the Maxwell equations together with the constitutive relations must be valid for the quantum fields. Again, it is convenient to introduce frequency components,

$$\hat{f} = \int_0^\infty \mathrm{d}\omega\, \underline{\hat{f}}(\omega) + \mathrm{H.c.}\,, \tag{2.230}$$

where we have accounted for the operator nature of the fields by using the hermitian conjugate. Further, one should note that our definition of the frequency components holds both in the Schrödinger picture where operators are time-independent and in the Heisenberg picture where they explicitly depend on time. We require the frequency components of the electromagnetic field to obey Maxwells equations

$$\nabla \cdot \underline{\hat{\boldsymbol{D}}} = 0\,, \tag{2.231}$$

$$\nabla \cdot \underline{\hat{\boldsymbol{B}}} = 0\,, \tag{2.232}$$

$$\nabla \times \underline{\hat{\boldsymbol{E}}} - \mathrm{i}\omega \underline{\hat{\boldsymbol{B}}} = \boldsymbol{0}\,, \tag{2.233}$$

$$\nabla \times \underline{\hat{\boldsymbol{H}}} + \mathrm{i}\omega \underline{\hat{\boldsymbol{D}}} = \boldsymbol{0} \tag{2.234}$$

and the constitutive relations

$$\underline{\hat{\boldsymbol{D}}} = \varepsilon_0 \varepsilon \underline{\hat{\boldsymbol{E}}} + \underline{\hat{\boldsymbol{P}}}_{\mathrm{N}}\,, \tag{2.235}$$

$$\underline{\hat{\boldsymbol{H}}} = \frac{1}{\mu_0 \mu} \underline{\hat{\boldsymbol{B}}} - \underline{\hat{\boldsymbol{M}}}_{\mathrm{N}}\,. \tag{2.236}$$

Quantum noise polarisation and magnetisation are related to the corresponding noise charge and current densities according to

$$\underline{\hat{\boldsymbol{j}}}_{\mathrm{N}} = -\mathrm{i}\omega \underline{\hat{\boldsymbol{P}}}_{\mathrm{N}} + \nabla \times \underline{\hat{\boldsymbol{M}}}_{\mathrm{N}}\,, \tag{2.237}$$

$$\underline{\hat{\rho}}_{\mathrm{N}} = -\nabla \cdot \underline{\hat{\boldsymbol{P}}}_{\mathrm{N}} \tag{2.238}$$

and the continuity equation holds,

$$-\mathrm{i}\omega \underline{\hat{\rho}}_{\mathrm{N}} + \nabla \cdot \underline{\hat{\boldsymbol{j}}}_{\mathrm{N}} = 0\,. \tag{2.239}$$

As the noise fields are now quantum operators, we must specify their commutation relations. Our choice must be such that noise polarisation and magnetisation vanish on their quantum average and that their fluctuation spectrum obeys the fluctuation–dissipation theorem. As we will show in the following, these conditions can be fulfilled by relating polarisation and magnetisation to fundamental creation and annihilation operators $\hat{\boldsymbol{f}}^{\dagger}_{\lambda}(\boldsymbol{r},\omega)$ and $\hat{\boldsymbol{f}}_{\lambda}(\boldsymbol{r},\omega)$ [5, 6, 11–13],

$$\underline{\hat{\boldsymbol{P}}}_{\mathrm{N}}(\boldsymbol{r},\omega) = \mathrm{i}\sqrt{\frac{\hbar\varepsilon_0}{\pi}\,\mathrm{Im}\,\varepsilon(\boldsymbol{r},\omega)}\,\hat{\boldsymbol{f}}_{e}(\boldsymbol{r},\omega)\,, \tag{2.240}$$

$$\underline{\hat{\boldsymbol{M}}}_{\mathrm{N}}(\boldsymbol{r},\omega) = \sqrt{\frac{\hbar}{\pi\mu_0}\,\frac{\mathrm{Im}\,\mu(\boldsymbol{r},\omega)}{|\mu(\boldsymbol{r},\omega)|^2}}\,\hat{\boldsymbol{f}}_{m}(\boldsymbol{r},\omega) \tag{2.241}$$

which obey bosonic commutation relations

$$\big[\hat{\boldsymbol{f}}_{\lambda}(\boldsymbol{r},\omega),\hat{\boldsymbol{f}}_{\lambda'}(\boldsymbol{r}',\omega')\big] = \big[\hat{\boldsymbol{f}}^{\dagger}_{\lambda}(\boldsymbol{r},\omega),\hat{\boldsymbol{f}}^{\dagger}_{\lambda'}(\boldsymbol{r}',\omega')\big] = \boldsymbol{0}\,, \tag{2.242}$$

$$\big[\hat{\boldsymbol{f}}_{\lambda}(\boldsymbol{r},\omega),\hat{\boldsymbol{f}}^{\dagger}_{\lambda'}(\boldsymbol{r}',\omega')\big] = \delta_{\lambda\lambda'}\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\delta(\omega-\omega')\,. \tag{2.243}$$

They represent the collective, polariton-like [14] bosonic excitations of the body–field system. The system's ground state $|\{0\}\rangle$ is the ground state of the bosonic operators, defined by

$$\hat{\boldsymbol{f}}_{\lambda}(\boldsymbol{r},\omega)|\{0\}\rangle = \boldsymbol{0} \quad \forall\,\lambda,\boldsymbol{r},\omega\,. \tag{2.244}$$

In particular, it implies that the electromagnetic field is in its ground state, which is the quantum vacuum mentioned in the introduction. The complete Hilbert space of the body–field system can be spanned by Fock states obtained in the usual way by repeated application of the creation operators $\hat{\boldsymbol{f}}^{\dagger}_{\lambda}$ to the ground state. For instance, single- and two-quantum Fock states are given by

$$|\boldsymbol{1}_{\lambda}(\boldsymbol{r},\omega)\rangle = \hat{\boldsymbol{f}}^{\dagger}_{\lambda}(\boldsymbol{r},\omega)|\{0\}\rangle\,, \tag{2.245}$$

$$|\boldsymbol{1}_{\lambda}(\boldsymbol{r},\omega)\boldsymbol{1}_{\lambda'}(\boldsymbol{r}',\omega')\rangle = \frac{1}{\sqrt{2}}\,\hat{\boldsymbol{f}}^{\dagger}_{\lambda'}(\boldsymbol{r}',\omega')\hat{\boldsymbol{f}}^{\dagger}_{\lambda}(\boldsymbol{r},\omega)|\{0\}\rangle\,, \tag{2.246}$$

respectively; they are special cases of the general n-quantum Fock state

$$|\boldsymbol{1}_{\lambda_1}(\boldsymbol{r}_1,\omega_1)\ldots\boldsymbol{1}_{\lambda_n}(\boldsymbol{r}_n,\omega_n)\rangle = \frac{1}{\sqrt{n!}}\,\hat{\boldsymbol{f}}^{\dagger}_{\lambda_n}(\boldsymbol{r}_n,\omega_n)\cdots\hat{\boldsymbol{f}}^{\dagger}_{\lambda_1}(\boldsymbol{r}_1,\omega_1)|\{0\}\rangle\,. \tag{2.247}$$

The normalisation factor $1/\sqrt{n!}$ accounts for the fact that the product of two n-quantum states is the sum of $n!$ products of delta functions. From the above definition of the ground state it immediately follows that the creation and annihilation operators have a vanishing ground-state average,

$$\langle \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega)\rangle = \langle\{0\}|\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega)|\{0\}\rangle = \mathbf{0}\,, \tag{2.248}$$

$$\langle \hat{\boldsymbol{f}}^\dagger_\lambda(\boldsymbol{r},\omega)\rangle = \langle\{0\}|\hat{\boldsymbol{f}}^\dagger_\lambda(\boldsymbol{r},\omega)|\{0\}\rangle = \mathbf{0}\,. \tag{2.249}$$

Invoking the bosonic commutation relations, one can further find the following results for ground-state averages of quadratic combinations of $\hat{\boldsymbol{f}}_\lambda$ and $\hat{\boldsymbol{f}}^\dagger_\lambda$:

$$\langle \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega)\hat{\boldsymbol{f}}_{\lambda'}(\boldsymbol{r}',\omega')\rangle = \mathbf{0}\,, \tag{2.250}$$

$$\langle \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega)\hat{\boldsymbol{f}}^\dagger_{\lambda'}(\boldsymbol{r}',\omega')\rangle = \delta_{\lambda\lambda'}\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\delta(\omega-\omega')\,, \tag{2.251}$$

$$\langle \hat{\boldsymbol{f}}^\dagger_\lambda(\boldsymbol{r},\omega)\hat{\boldsymbol{f}}_{\lambda'}(\boldsymbol{r}',\omega')\rangle = \mathbf{0}\,, \tag{2.252}$$

$$\langle \hat{\boldsymbol{f}}^\dagger_\lambda(\boldsymbol{r},\omega)\hat{\boldsymbol{f}}^\dagger_{\lambda'}(\boldsymbol{r}',\omega')\rangle = \mathbf{0}\,. \tag{2.253}$$

The above relations imply that noise polarisation and magnetisation vanish on their ground-state average,

$$\langle \hat{\boldsymbol{P}}_\mathrm{N}\rangle = \mathbf{0}\,,\qquad \langle \hat{\boldsymbol{M}}_\mathrm{N}\rangle = \mathbf{0}\,, \tag{2.254}$$

and their fluctuations agree with the fluctuation–dissipation theorem [2, 3],

$$\langle \mathcal{S}[\Delta\hat{\boldsymbol{P}}_\mathrm{N}(\boldsymbol{r},\omega)\Delta\hat{\boldsymbol{P}}^\dagger_\mathrm{N}(\boldsymbol{r}',\omega')]\rangle = \frac{\hbar}{2\pi}\,\varepsilon_0\,\mathrm{Im}\,\chi(\boldsymbol{r},\omega)\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\delta(\omega-\omega')\,, \tag{2.255}$$

$$\langle \mathcal{S}[\Delta\hat{\boldsymbol{M}}_\mathrm{N}(\boldsymbol{r},\omega)\Delta\hat{\boldsymbol{M}}^\dagger_\mathrm{N}(\boldsymbol{r}',\omega')]\rangle = \frac{\hbar}{2\pi}\,\frac{\mathrm{Im}\,\zeta(\boldsymbol{r},\omega)}{\mu_0}\,\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\delta(\omega-\omega')\,, \tag{2.256}$$

in accordance with our requirement (ii). Here, $\mathcal{S}(\hat{a}\hat{b}) = \frac{1}{2}(\hat{a}\hat{b}+\hat{b}\hat{a})$ denotes a symmetrised operator product. Note that the average thermal energy $k_\mathrm{B}T$ appearing in the classical fluctuation–dissipation theorem (2.117) and (2.118) has been replaced with the quantum ground-state energy $\frac{1}{2}\hbar\omega$ of a bosonic system. The use of a noise polarisation with a spectrum governed by (2.255) is known as Rytov theory [15], it lies at the heart of Lifshitz' famous calculation of the Casimir force [16].

Having introduced the fundamental variables of the system and their commutation relations, explicit quantisation of the electromagnetic field can be performed by expressing all field operators in terms of these variables; this can be achieved by solving the Maxwell equations. In complete analogy to the classical case, we can combine (2.233)–(2.236) into a Helmholtz equation for the electric field,

$$\left[\boldsymbol{\nabla}\times\frac{1}{\mu}\boldsymbol{\nabla}\times\;-\frac{\omega^2}{c^2}\,\varepsilon\right]\underline{\hat{\boldsymbol{E}}} = \mathrm{i}\mu_0\omega\underline{\hat{\boldsymbol{j}}}_\mathrm{N}\,, \tag{2.257}$$

which can be formally solved by means of the classical Green's tensor:

$$\underline{\hat{\boldsymbol{E}}}(\boldsymbol{r},\omega) = \mathrm{i}\mu_0\omega\int \mathrm{d}^3r'\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\underline{\hat{\boldsymbol{j}}}_\mathrm{N}(\boldsymbol{r}',\omega)\,. \tag{2.258}$$

Expressing the noise current density in terms of the fundamental fields $\hat{\boldsymbol{f}}$ and $\hat{\boldsymbol{f}}^{\dagger}_{\lambda}$ by means of $\underline{\hat{\boldsymbol{j}}}_{\mathrm{N}} = -\mathrm{i}\omega\underline{\hat{\boldsymbol{P}}}_{\mathrm{N}} + \nabla \times \underline{\hat{\boldsymbol{M}}}_{\mathrm{N}}$ together with (2.240) and (2.241), we obtain the desired expansion of the electric field,

$$\begin{aligned}\hat{\boldsymbol{E}}(\boldsymbol{r}) &= \int_0^\infty \mathrm{d}\omega\, \underline{\hat{\boldsymbol{E}}}(\boldsymbol{r},\omega) + \mathrm{H.c.} \\ &= \int_0^\infty \mathrm{d}\omega \sum_{\lambda=e,m} \int \mathrm{d}^3r'\, \boldsymbol{G}_\lambda(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}',\omega) + \mathrm{H.c.}\end{aligned} \tag{2.259}$$

The coefficients

$$\boldsymbol{G}_e(\boldsymbol{r},\boldsymbol{r}',\omega) = \mathrm{i}\,\frac{\omega^2}{c^2}\sqrt{\frac{\hbar}{\pi\varepsilon_0}\,\mathrm{Im}\,\varepsilon(\boldsymbol{r}',\omega)}\;\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\,, \tag{2.260}$$

$$\boldsymbol{G}_m(\boldsymbol{r},\boldsymbol{r}',\omega) = \mathrm{i}\,\frac{\omega}{c}\sqrt{\frac{\hbar}{\pi\varepsilon_0}\,\frac{\mathrm{Im}\,\mu(\boldsymbol{r}',\omega)}{|\mu(\boldsymbol{r}',\omega)|^2}}\left[\nabla'\times\boldsymbol{G}(\boldsymbol{r}',\boldsymbol{r},\omega)\right]^{\mathrm{T}}. \tag{2.261}$$

obey the integral relation

$$\sum_{\lambda=e,m}\int \mathrm{d}^3s\, \boldsymbol{G}_\lambda(\boldsymbol{r},\boldsymbol{s},\omega)\cdot \boldsymbol{G}^{*\mathrm{T}}_\lambda(\boldsymbol{r}',\boldsymbol{s},\omega) = \frac{\hbar\mu_0}{\pi}\,\omega^2\,\mathrm{Im}\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega) \tag{2.262}$$

which follows directly from the integral relation (2.153). Expansions of all other relevant fields in terms of the dynamical variables follow from this result by virtue of the Maxwell equations and constitutive relations in frequency space. Thus, (2.233) leads to

$$\begin{aligned}\hat{\boldsymbol{B}}(\boldsymbol{r}) &= \int_0^\infty \mathrm{d}\omega\, \underline{\hat{\boldsymbol{B}}}(\boldsymbol{r},\omega) + \mathrm{H.c.} \\ &= \int_0^\infty \frac{\mathrm{d}\omega}{\mathrm{i}\omega} \sum_{\lambda=e,m} \int \mathrm{d}^3r'\, \nabla\times\boldsymbol{G}_\lambda(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}',\omega) + \mathrm{H.c.}\end{aligned} \tag{2.263}$$

while (2.235), (2.236), (2.240) and (2.241) show that

$$\hat{\boldsymbol{D}}(\boldsymbol{r}) = \int_0^\infty \mathrm{d}\omega\, \underline{\hat{\boldsymbol{D}}}(\boldsymbol{r},\omega) + \text{H.c.}$$
$$= \int_0^\infty \mathrm{d}\omega \Bigg[\varepsilon_0\varepsilon(\boldsymbol{r},\omega) \sum_{\lambda=e,m} \int \mathrm{d}^3r'\, \boldsymbol{G}_\lambda(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}',\omega)$$
$$+\,\mathrm{i}\sqrt{\frac{\hbar\varepsilon_0}{\pi}\,\mathrm{Im}\,\varepsilon(\boldsymbol{r},\omega)}\;\hat{\boldsymbol{f}}_e(\boldsymbol{r},\omega)\Bigg] + \text{H.c.} \tag{2.264}$$

and

$$\hat{\boldsymbol{H}}(\boldsymbol{r}) = \int_0^\infty \mathrm{d}\omega\, \underline{\hat{\boldsymbol{H}}}(\boldsymbol{r},\omega) + \text{H.c.}$$
$$= \int_0^\infty \mathrm{d}\omega \Bigg[\frac{1}{\mathrm{i}\omega\mu_0\mu(\boldsymbol{r},\omega)} \sum_{\lambda=e,m} \int \mathrm{d}^3r'\, \nabla\times\boldsymbol{G}_\lambda(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}',\omega)$$
$$-\sqrt{\frac{\hbar\kappa_0}{\pi}\,\frac{\mathrm{Im}\,\mu(\boldsymbol{r},\omega)}{\mu(\boldsymbol{r},\omega)}}\;\hat{\boldsymbol{f}}_m(\boldsymbol{r},\omega)\Bigg] + \text{H.c.} \tag{2.265}$$

As in the classical case, the Maxwell equations and constitutive relations in Fourier space are fulfilled by construction.

Commutation relations for the fields can be deduced from the bosonic commutation relations of the fundamental fields in a straightforward way, as demonstrated in App. A. In particular, it may be shown that electric and magnetic field obey the equal-time commutation relations [5, 6, 11–13]

$$\big[\hat{\boldsymbol{E}}(\boldsymbol{r}),\hat{\boldsymbol{E}}(\boldsymbol{r}')\big] = \big[\hat{\boldsymbol{B}}(\boldsymbol{r}),\hat{\boldsymbol{B}}(\boldsymbol{r}')\big] = \boldsymbol{0}\,, \tag{2.266}$$
$$\big[\hat{\boldsymbol{E}}(\boldsymbol{r}),\hat{\boldsymbol{B}}(\boldsymbol{r}')\big] = \frac{\mathrm{i}\hbar}{\varepsilon_0}\nabla\times\boldsymbol{\delta}(\boldsymbol{r}-\boldsymbol{r}')\,. \tag{2.267}$$

They agree with those in free space, in accordance with our requirement (iii). Similarly, the fundamental commutation relations can be used to calculate the ground-state fluctuation spectrum of the electric field (2.259). Upon using the integral relation (2.262), one finds [5, 11]

$$\big\langle \mathcal{S}\big[\Delta\hat{\boldsymbol{E}}(\boldsymbol{r},\omega)\Delta\hat{\boldsymbol{E}}^\dagger(\boldsymbol{r}',\omega')\big]\big\rangle = \frac{\hbar}{2\pi}\,\mu_0\omega^2\,\mathrm{Im}\,\boldsymbol{G}(\boldsymbol{r},\boldsymbol{r}',\omega)\delta(\omega-\omega')\,, \tag{2.268}$$

in agreement with the fluctuation–dissipation theorem. In comparison with the classical fluctuation spectrum (2.174), the average thermal energy $k_\mathrm{B}T$ has again been replaced with the quantum ground-state energy $\frac{1}{2}\hbar\omega$.

Next, we must specify the Hamiltonian of the system, which governs its dynamics. We require this Hamiltonian to generate the correct time-dependence of the field operators in the Heisenberg picture such that the Maxwell equations together with the constitutive equations hold. As these equations are fulfilled in the frequency domain by construction, we simply have to ensure that the time-dependent frequency components of the field operators are ordinary Fourier components,

$$\underline{\hat{f}}(\omega, t) = \underline{\hat{f}}(\omega)\mathrm{e}^{-\mathrm{i}\omega t}\,, \tag{2.269}$$

so that the Fourier relation

$$\hat{f}(t) = \int_0^\infty \mathrm{d}\omega\, \underline{\hat{f}}(\omega)\mathrm{e}^{-\mathrm{i}\omega t} + \mathrm{H.c.} \tag{2.270}$$

holds. This is achieved by the Hamiltonian [5, 6, 11–13]

$$\hat{H}_\mathrm{F} = \sum_{\lambda=e,m} \int \mathrm{d}^3 r \int_0^\infty \mathrm{d}\omega\, \hbar\omega\, \hat{\boldsymbol{f}}^\dagger_\lambda(\boldsymbol{r}, \omega) \cdot \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}, \omega)\,, \tag{2.271}$$

as can be seen as follows: $\hat{H}_\mathrm{F}$ generates the Heisenberg equations of motion

$$\dot{\hat{\boldsymbol{f}}}_\lambda(\boldsymbol{r}, \omega) = \frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}, \omega), \hat{H}_\mathrm{F}\big] = -\mathrm{i}\omega\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}, \omega) \tag{2.272}$$

which are solved by

$$\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}, \omega, t) = \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}, \omega)\mathrm{e}^{-\mathrm{i}\omega t}\,. \tag{2.273}$$

The electromagnetic field operators introduced in the section being linear combinations of the fundamental fields, their time-dependent frequency components are thus ordinary Fourier components, as required. Upon using the Fourier relation (2.270) it follows that the Maxwell equations and constitutive relations in frequency space (2.231)–(2.236) also hold in the time domain [5, 6, 11, 12],

$$\boldsymbol{\nabla} \cdot \hat{\boldsymbol{D}} = 0\,, \tag{2.274}$$

$$\boldsymbol{\nabla} \cdot \hat{\boldsymbol{B}} = 0\,, \tag{2.275}$$

$$\boldsymbol{\nabla} \times \hat{\boldsymbol{E}} + \dot{\hat{\boldsymbol{B}}} = \boldsymbol{0}\,, \tag{2.276}$$

$$\boldsymbol{\nabla} \times \hat{\boldsymbol{H}} - \dot{\hat{\boldsymbol{D}}} = \boldsymbol{0}\,, \tag{2.277}$$

$$\hat{\boldsymbol{D}}(\boldsymbol{r},t) = \varepsilon_0 \hat{\boldsymbol{E}}(\boldsymbol{r},t) + \varepsilon_0 \int_{-\infty}^{\infty} \mathrm{d}\tau\, \chi(\boldsymbol{r},\tau) \hat{\boldsymbol{E}}(\boldsymbol{r},t-\tau) + \hat{\boldsymbol{P}}_{\mathrm{N}}(\boldsymbol{r},t)\,, \tag{2.278}$$

$$\hat{\boldsymbol{H}}(\boldsymbol{r},t) = \frac{1}{\mu_0} \hat{\boldsymbol{B}}(\boldsymbol{r},t) - \frac{1}{\mu_0} \int_{-\infty}^{\infty} \mathrm{d}\tau\, \zeta(\boldsymbol{r},\tau) \hat{\boldsymbol{B}}(\boldsymbol{r},t-\tau) - \hat{\boldsymbol{M}}_{\mathrm{N}}(\boldsymbol{r},t)\,. \tag{2.279}$$

Having established the system's Hamiltonian (2.271), let us find its eigenstates. The ground-state $|\{0\}\rangle$ is obviously an eigenstate,

$$\hat{H}_{\mathrm{F}} |\{0\}\rangle = 0\,, \tag{2.280}$$

as follows from its definition (2.244). By invoking the bosonic commutation relations (2.242) and (2.243), one can further show that the single- and two-quantum Fock states (2.245) and (2.246) are also energy-eigenstates,

$$\hat{H}_{\mathrm{F}} |\mathbf{1}_{\lambda}(\boldsymbol{r},\omega)\rangle = \hbar\omega |\mathbf{1}_{\lambda}(\boldsymbol{r},\omega)\rangle\,, \tag{2.281}$$

$$\hat{H}_{\mathrm{F}} |\mathbf{1}_{\lambda}(\boldsymbol{r},\omega)\mathbf{1}_{\lambda'}(\boldsymbol{r}',\omega')\rangle = \hbar(\omega+\omega') |\mathbf{1}_{\lambda}(\boldsymbol{r},\omega)\mathbf{1}_{\lambda'}(\boldsymbol{r}',\omega')\rangle\,. \tag{2.282}$$

Even more generally, every n-quantum Fock state is an energy eigenstate whose energy is just the sum of the energies associated with the excitations involved,

$$\begin{aligned} &\hat{H}_{\mathrm{F}} |\mathbf{1}_{\lambda_1}(\boldsymbol{r}_1,\omega_1)\ldots\mathbf{1}_{\lambda_n}(\boldsymbol{r}_n,\omega_n)\rangle \\ &\quad = \hbar(\omega_1+\ldots+\omega_n) |\mathbf{1}_{\lambda_1}(\boldsymbol{r}_1,\omega_1)\ldots\mathbf{1}_{\lambda_n}(\boldsymbol{r}_n,\omega_n)\rangle\,. \end{aligned} \tag{2.283}$$

The ground state $|\{0\}\rangle$ is obviously the eigenstate with the lowest energy, as required.

Let us summarise the constructed quantisation scheme: By using the Maxwell equations in the frequency domain as a guide and expressing all relevant fields in terms of appropriately chosen bosonic variables, we have thus succeeded in constructing a macroscopic QED that fulfils our three initial requirements: It is compatible with (i) classical macroscopic electrodynamics (because the Maxwell equations and constitutive relations hold), (ii) statistical physics (the quantum fluctuations of both the noise fields and the electric field being in accordance with the fluctuation–dissipation theorem) and (iii) free-space QED (with the electromagnetic field obeying the correct commutation relations).

Since the Maxwell equations for the quantised have exactly the same form as the classical Maxwell equations, all our results regarding the duality of electric and magnetic fields as given in Sect. 2.1.4 remain valid in the quantum case. In particular, the duality transformation laws for the quantum fields are completely analogous to the classical fields as listed in Table 2.1; and duality transformations leave the equations of motion (2.274)–(2.279) invariant. Since the Heisenberg equations of motion are generated by the Hamiltonian (2.271), we can conclude that this Hamiltonian itself must be duality-invariant. To see this more directly, let us determine the transformation behaviour of the fundamental fields $\hat{\boldsymbol{f}}_\lambda$ and $\hat{\boldsymbol{f}}^\dagger_\lambda$. Writing the relations (2.240) and (2.241) between those fields and the noise polarisation and magnetisation in dual-pair notation as

$$\begin{pmatrix} Z_0\underline{\hat{\boldsymbol{P}}}_\mathrm{N} \\ \mu_0\underline{\hat{\boldsymbol{M}}}_\mathrm{N} \end{pmatrix} = \sqrt{\frac{\hbar\mu_0}{\pi}} \begin{pmatrix} \mathrm{i}\sqrt{\mathrm{Im}\,\varepsilon} & 0 \\ 0 & \sqrt{\mathrm{Im}\,\mu}/|\mu| \end{pmatrix} \begin{pmatrix} \hat{\boldsymbol{f}}_e \\ \hat{\boldsymbol{f}}_m \end{pmatrix} \tag{2.284}$$

and recalling Eqs. (2.190) and (2.193), they are seen to transform as

$$\begin{pmatrix} \hat{\boldsymbol{f}}_e \\ \hat{\boldsymbol{f}}_m \end{pmatrix}^{\circledast} = \begin{pmatrix} \cos\theta & -\mathrm{i}(\mu/|\mu|)\sin\theta \\ -\mathrm{i}(|\varepsilon|/\varepsilon)\sin\theta & \cos\theta \end{pmatrix} \begin{pmatrix} \hat{\boldsymbol{f}}_e \\ \hat{\boldsymbol{f}}_m \end{pmatrix} \tag{2.285}$$

in both the continuous ($\varepsilon = \mu$) and discrete ($\theta = n\pi/2$ with $n \in \mathbb{Z}$) cases. For the nontrivial transformations, the fundamental variables are thus exchanged and multiplied with phase factors. The phase factors do not affect the Hamiltonian which is quadratic and symmetric in $\hat{\boldsymbol{f}}_e$ and $\hat{\boldsymbol{f}}_m$. It follows that a duality transformation leaves the Hamiltonian invariant, $\hat{H}_F^{\circledast} = \hat{H}_F$, as expected.

As a preparation for the following section, where atom–field interactions will be discussed, it is useful to introduce scalar and vector potentials for the electric and magnetic fields in the usual way,

$$\hat{\boldsymbol{E}} = -\nabla\hat{\phi} - \dot{\hat{\boldsymbol{A}}}\,, \tag{2.286}$$

$$\hat{\boldsymbol{B}} = \nabla \times \hat{\boldsymbol{A}}\,. \tag{2.287}$$

In Coulomb gauge $\nabla \cdot \hat{\boldsymbol{A}} = 0$, we have

$$\hat{\boldsymbol{E}}^\parallel = -\nabla\hat{\phi}\,, \qquad \hat{\boldsymbol{E}}^\perp = -\dot{\hat{\boldsymbol{A}}}\,, \tag{2.288}$$

so that $\hat{\phi}$ and $\hat{\boldsymbol{A}}$ can be expressed in terms of the fundamental variables by using the representation of the electric field (2.259):

$$\begin{aligned} \nabla\hat{\phi}(\boldsymbol{r}) &= \int_0^\infty \mathrm{d}\omega\, \nabla\underline{\hat{\phi}}(\boldsymbol{r},\omega) + \mathrm{H.c.} \\ &= -\sum_{\lambda=e,m} \int \mathrm{d}^3r' \int_0^\infty \mathrm{d}\omega\, {}^\parallel\boldsymbol{G}_\lambda(\boldsymbol{r},\boldsymbol{r}',\omega) \cdot \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}',\omega) + \mathrm{H.c.}\,, \end{aligned} \tag{2.289}$$

$$\hat{\boldsymbol{A}}(\boldsymbol{r}) = \int_0^\infty \mathrm{d}\omega\, \underline{\hat{\boldsymbol{A}}}(\boldsymbol{r},\omega) + \mathrm{H.c.}$$

$$= \sum_{\lambda=e,m} \int \mathrm{d}^3r' \int_0^\infty \frac{\mathrm{d}\omega}{\mathrm{i}\omega} \, {}^{\perp}\boldsymbol{G}_\lambda(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}', \omega) + \text{H.c.} \tag{2.290}$$

where we denote left/right longitudinal or transverse components of a tensor field $\boldsymbol{T}(\boldsymbol{r}, \boldsymbol{r}')$ by

$${}^{\parallel/\perp}\boldsymbol{T}^{\parallel/\perp}(\boldsymbol{r}, \boldsymbol{r}') = \int \mathrm{d}^3s \int \mathrm{d}^3s' \, \boldsymbol{\delta}^{\parallel/\perp}(\boldsymbol{r}-\boldsymbol{s}) \cdot \boldsymbol{T}(\boldsymbol{s}, \boldsymbol{s}') \cdot \boldsymbol{\delta}^{\parallel/\perp}(\boldsymbol{s}'-\boldsymbol{r}'), \tag{2.291}$$

recall (2.15). Note that for a tensor fulfilling Onsager reciprocity (2.152), one has

$${}^{\parallel/\perp}\boldsymbol{G}(\boldsymbol{r}, \boldsymbol{r}') = \boldsymbol{G}^{\mathrm{T}\parallel/\perp}(\boldsymbol{r}', \boldsymbol{r}). \tag{2.292}$$

As in the free-space case, the canonically conjugate momentum associated with the vector potential is given by

$$\hat{\boldsymbol{\Pi}} = -\varepsilon_0 \hat{\boldsymbol{E}}^{\perp}, \tag{2.293}$$

as can be easily demonstrated by verifying the canonical equal-time commutation relations (Appendix A)

$$\left[\hat{\boldsymbol{A}}(\boldsymbol{r}), \hat{\boldsymbol{A}}(\boldsymbol{r}')\right] = \left[\hat{\boldsymbol{\Pi}}(\boldsymbol{r}), \hat{\boldsymbol{\Pi}}(\boldsymbol{r}')\right] = \boldsymbol{0}, \tag{2.294}$$

$$\left[\hat{\boldsymbol{A}}(\boldsymbol{r}), \hat{\boldsymbol{\Pi}}(\boldsymbol{r}')\right] = \mathrm{i}\hbar\boldsymbol{\delta}^{\perp}(\boldsymbol{r}-\boldsymbol{r}'). \tag{2.295}$$

We conclude the section with a few remarks concerning the validity and physical interpretation of the macroscopic QED outlined above as well as its relation to other theories. The validity of our quantisation scheme depends crucially on the assumption that all space is filled with absorbing media, so that $\mathrm{Im}\,\varepsilon(\boldsymbol{r}, \omega) > 0$ and $\mathrm{Im}\,\mu(\boldsymbol{r}, \omega) > 0$ hold everywhere. This condition acts as a regularisation which guarantees the convergence of spatial integrals of the type (2.259). At the same time, it is obviously vital for preserving the correct equal-time commutation relations for the electromagnetic field: In the extreme example that $\mathrm{Im}\,\varepsilon(\boldsymbol{r}, \omega) = 0$ and $\mathrm{Im}\,\mu(\boldsymbol{r}, \omega) = 0$ everywhere, the electric and magnetic field operators would vanish and thus commute trivially. As a consequence, even in free-space regions or regions where absorption is very small and can be neglected in practice, the imaginary parts of permittivity and permeability must not be set equal to zero in the integrands of expressions of the type (2.259). To allow for free-space regions, the limits $\mathrm{Im}\,\varepsilon(\boldsymbol{r}, \omega) \to 0$ and $\mathrm{Im}\,\mu(\boldsymbol{r}, \omega) \to 0$ may be performed after taking expectation values and having carried out all spatial integrals. In this sense the theory provides the quantised electromagnetic field in the presence of an arbitrary arrangement of linear, causal magnetoelectric bodies characterised by their permittivities and permeabilities, where $\mathrm{Im}\,\varepsilon(\boldsymbol{r}, \omega) \geq 0$ and $\mathrm{Im}\,\mu(\boldsymbol{r}, \omega) \geq 0$; it may thus be regarded as a generalisation of the free-space QED presented in the previous section.

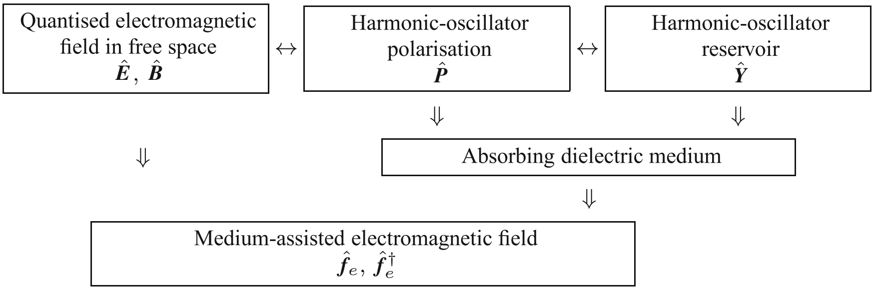

Fig. 2.1 The Huttner–Barnett model

Instead of introducing bosonic operators as the fundamental fields in terms of which the electromagnetic field is expressed, one can also construct a quantisation scheme by using noise polarisation and magnetisation as the fundamental quantities whose commutations relations are chosen such that the fluctuation–dissipation theorem holds in the form of (2.255) and (2.256). Such a variant of the macroscopic QED presented here was initially developed for dielectrics [17, 18] and was later extended to magnetodielectrics [19]. Alternatively, macroscopic QED can be constructed by expressing the electromagnetic field in terms of auxiliary fields whose dynamics is such that the Maxwell equations are fulfilled [20]. It has been shown that this approach is equivalent to the one presented here [21].

All of the macroscopic theories presented above are effective theories where the true behaviour of the charged particles constituting the medium is characterised approximately by the introduced permittivity and permeability; such an approximation obviously breaks down on length scales comparable to the interatomic distances within the medium. When applying the macroscopic QED presented here to a specific scenario, specific permittivities and permeabilities describing the media in question are used as input to the theory, they can be obtained either from microscopic model calculations or from experimental measurements. One of the strengths of the approach is its generality: In principle, arbitrary linear, causal response functions can be used for ε and μ in order to account for the specific properties of the respective media. The theory can be further generalised to include media with a non-local and/or anisotropic response, recall (2.96) and (2.97) [12, 22].

For some special cases of simple model media, more explicit treatments of the field–medium interaction have been carried out. They provide for an additional justification of our theory while shedding some light on the physical meaning of the fundamental fields. This approach was first pursued by Huttner and Barnett [23] who studied the case of a bulk absorbing single-resonance dielectric, their work was later extended to inhomogeneous dielectric bodies [24–26], including bodies exhibiting non-local properties [27]. The Huttner–Barnett model consists of the quantised free-space QED field interacting with a harmonic-oscillator polarisation field which in turn interacts with an additional harmonic-oscillator field acting as a reservoir to model absorption (see Fig. 2.1). Employing a Fano diagonalisation [28], Huttner and

Barnett found the collective variables of this interacting system in two steps. First, they diagonalised the Hamiltonian describing the coupled interaction of polarisation field plus reservoir, obtaining the collective variables of the absorbing medium. They then treated the interaction of this medium with the electromagnetic field in a similar way, leading to collective, bosonic variables which may be identified with our fundamental variables $\hat{\boldsymbol{f}}_e$ and $\hat{\boldsymbol{f}}_e^\dagger$. Their calculation hence shows that the fundamental fields $\hat{\boldsymbol{f}}_\lambda$ and $\hat{\boldsymbol{f}}_\lambda^\dagger$ describe collective excitations of the absorbing medium and the electromagnetic field. The expansions (2.259) and (2.263) may be regarded as projections of the collective variables onto the field subspace of the medium–field system.

2.4 Atom–Field Interactions

Let us next describe how the medium-assisted electromagnetic field characterised in the previous section interacts with one or several atoms. To ensure that our macroscopic description of the medium remains valid, we will typically assume that the atoms are situated in free space and that the medium is present in the form of one or more magnetoelectric bodies [described by regions where $\varepsilon(\boldsymbol{r}, \omega) \neq 1$ or $\mu(\boldsymbol{r}, \omega) \neq 1$], with the atoms being well separated from the bodies. In particular, the atom–body separations must be large compared to the interatomic distances of the atoms forming the medium.

A neutral atom or molecule A (briefly referred to as atom in the following) may be envisioned as a system of particles $\alpha \in A$ with charges q_α ($\sum_{\alpha\in A} q_\alpha = 0$), masses m_α, positions $\hat{\boldsymbol{r}}_\alpha$ and canonically conjugate momenta $\hat{\boldsymbol{p}}_\alpha$. The Poisson brackets (2.79) and (2.80) for classical particles imply by virtue of the correspondence principle (2.216) that the canonical commutation relations are given by

$$\left[\hat{\boldsymbol{r}}_\alpha, \hat{\boldsymbol{r}}_\beta\right] = \left[\hat{\boldsymbol{p}}_\alpha, \hat{\boldsymbol{p}}_\beta\right] = 0, \qquad \left[\hat{\boldsymbol{r}}_\alpha, \hat{\boldsymbol{p}}_\beta\right] = \mathrm{i}\hbar\delta_{\alpha\beta}\boldsymbol{I}\,. \tag{2.296}$$

From the classical Hamiltonian (2.70), we conclude that the Hamiltonian for a nonrelativistic atom is given by

$$\hat{H}_A = \sum_{\alpha\in A} \frac{\hat{\boldsymbol{p}}_\alpha^2}{2m_\alpha} + \sum_{\substack{\alpha,\beta\in A\\ \alpha\neq\beta}} \frac{q_\alpha q_\beta}{8\pi\varepsilon_0|\hat{\boldsymbol{r}}_\alpha - \hat{\boldsymbol{r}}_\beta|}\,. \tag{2.297}$$

In close analogy to the classical case, one may introduce charge and current densities associated with the atom,

$$\hat{\rho}_A(\boldsymbol{r}) = \sum_{\alpha\in A} q_\alpha \delta(\boldsymbol{r} - \hat{\boldsymbol{r}}_\alpha)\,, \tag{2.298}$$

$$\hat{\boldsymbol{j}}_A(\boldsymbol{r}) = \sum_{\alpha\in A} q_\alpha \mathcal{S}\Big[\dot{\hat{\boldsymbol{r}}}_\alpha \delta(\boldsymbol{r} - \hat{\boldsymbol{r}}_\alpha)\Big]\,, \tag{2.299}$$

which obey the continuity equation

$$\dot{\hat{\rho}}_A(\boldsymbol{r}) + \nabla\cdot\hat{\boldsymbol{j}}_A(\boldsymbol{r}) = 0\,. \tag{2.300}$$

The symmetrisation operator $\mathcal{S}$ arises, because $\hat{\boldsymbol{r}}_\alpha$ and $\dot{\hat{\boldsymbol{r}}}_\alpha$ do not commute. It is defined as the weighted sum over all possible orderings of an operator product, e.g.,

$$\mathcal{S}(\hat{a}\hat{b}) = \frac{1}{2}\big(\hat{a}\hat{b} + \hat{b}\hat{a}\big)\,, \qquad \mathcal{S}(\hat{a}\hat{b}^2) = \frac{1}{3}\big(\hat{a}\hat{b}^2 + \hat{b}\hat{a}\hat{b} + \hat{b}^2\hat{a}\big)\,. \tag{2.301}$$

The definition can be extended to analytic functions $f(a,b) = \sum_{i,j=0}^{\infty} f_{ij} a^i b^j$ in a straightforward way,

$$\mathcal{S} f(\hat{a},\hat{b}) = \sum_{i,j=0}^{\infty} f_{ij} \mathcal{S}\big(\hat{a}^i \hat{b}^j\big)\,. \tag{2.302}$$

For expressions containing the delta function, symmetrisation is implied after integrating, e.g.,

$$\int \mathrm{d}^3 r\, f(\boldsymbol{r}, \hat{\boldsymbol{p}}_\alpha)\mathcal{S}\delta(\boldsymbol{r} - \hat{\boldsymbol{r}}_\alpha) = \mathcal{S} f(\boldsymbol{r}_\alpha, \hat{\boldsymbol{p}}_\alpha)\,. \tag{2.303}$$

By repeated use of the operator identity (2.226), one can show that time derivatives automatically lead to symmetrised expressions, e.g.,

$$\frac{\partial}{\partial t}\delta(\boldsymbol{r} - \hat{\boldsymbol{r}}_\alpha) = \frac{1}{\mathrm{i}\hbar}\big[\delta(\boldsymbol{r} - \hat{\boldsymbol{r}}_\alpha), \hat{H}\big] = \mathcal{S}\Big[\dot{\hat{\boldsymbol{r}}}_\alpha \cdot \nabla\delta(\boldsymbol{r} - \hat{\boldsymbol{r}}_\alpha)\Big]\,. \tag{2.304}$$

Introducing the atomic Coulomb potential

$$\hat{\phi}_A(\boldsymbol{r}) = \int \mathrm{d}^3 r' \frac{\hat{\rho}_A(\boldsymbol{r}')}{4\pi\varepsilon_0|\boldsymbol{r} - \boldsymbol{r}'|} = \sum_{\alpha\in A} \frac{q_\alpha}{4\pi\varepsilon_0|\boldsymbol{r} - \hat{\boldsymbol{r}}_\alpha|}\,, \tag{2.305}$$

which is the solution to the Poisson equation

$$-\varepsilon_0 \Delta\hat{\phi}_A = \hat{\rho}_A\,, \tag{2.306}$$

the Coulomb term in the atomic Hamiltonian can be rewritten as

$$\sum_{\substack{\alpha,\beta\in A\\ \alpha\neq\beta}} \frac{q_\alpha q_\beta}{8\pi\varepsilon_0|\hat{\boldsymbol{r}}_\alpha - \hat{\boldsymbol{r}}_\beta|} = \frac{1}{2}\int \mathrm{d}^3 r\, \hat{\rho}_A \hat{\phi}_A = \frac{1}{2}\int \mathrm{d}^3 r\, \varepsilon_0 \big(\nabla\hat{\phi}_A\big)^2\,. \tag{2.307}$$

Note that replacing the explicit form of the Coulomb interaction in (2.295) by an expression in terms of the Coulomb potential $\hat{\phi}_A$ has the effect of introducing divergent self-energy contributions, recall the discussion below (2.72) in Sect. 2.1.2.

Since an atom is a bound system of charges, it is useful to introduce centre-of-mass and relative coordinates

$$\hat{\boldsymbol{r}}_A = \sum_{\alpha\in A} \frac{m_\alpha}{m_A}\hat{\boldsymbol{r}}_\alpha\,, \qquad \hat{\bar{\boldsymbol{r}}}_\alpha = \hat{\boldsymbol{r}}_\alpha - \hat{\boldsymbol{r}}_A \tag{2.308}$$

($m_A = \sum_{\alpha\in A} m_\alpha$). The appropriate associated momenta are given by [29]

$$\hat{\boldsymbol{p}}_A = \sum_{\alpha\in A} \hat{\boldsymbol{p}}_\alpha\,, \qquad \hat{\bar{\boldsymbol{p}}}_\alpha = \hat{\boldsymbol{p}}_\alpha - \frac{m_\alpha}{m_A}\hat{\boldsymbol{p}}_A\,, \tag{2.309}$$

such that the commutation relations

$$\left[\hat{\boldsymbol{r}}_A, \hat{\boldsymbol{r}}_A\right] = \left[\hat{\boldsymbol{p}}_A, \hat{\boldsymbol{p}}_A\right] = \boldsymbol{0}\,, \qquad \left[\hat{\boldsymbol{r}}_A, \hat{\boldsymbol{p}}_A\right] = \mathrm{i}\hbar\boldsymbol{I}\,, \tag{2.310}$$

$$\left[\hat{\bar{\boldsymbol{r}}}_\alpha, \hat{\bar{\boldsymbol{r}}}_\beta\right] = \left[\hat{\bar{\boldsymbol{p}}}_\alpha, \hat{\bar{\boldsymbol{p}}}_\beta\right] = \boldsymbol{0}\,, \qquad \left[\hat{\bar{\boldsymbol{r}}}_\alpha, \hat{\bar{\boldsymbol{p}}}_\beta\right] = \mathrm{i}\hbar\left(\delta_{\alpha\beta} - \frac{m_\beta}{m_A}\right)\boldsymbol{I} \simeq \mathrm{i}\hbar\delta_{\alpha\beta}\boldsymbol{I}\,, \tag{2.311}$$

$$\left[\hat{\bar{\boldsymbol{r}}}_\alpha, \hat{\boldsymbol{r}}_A\right] = \left[\hat{\bar{\boldsymbol{p}}}_\alpha, \hat{\boldsymbol{p}}_A\right] = \left[\hat{\bar{\boldsymbol{r}}}_\alpha, \hat{\boldsymbol{p}}_A\right] = \left[\hat{\boldsymbol{r}}_A, \hat{\bar{\boldsymbol{p}}}_\alpha\right] = \boldsymbol{0} \tag{2.312}$$

follow from the canonical commutation relations (2.296), where the approximation is valid for electrons. Using these definitions, the atomic Hamiltonian may be written in the form

$$\hat{H}_A = \frac{\hat{\boldsymbol{p}}_A^2}{2m_A} + \sum_{\alpha\in A}\frac{\hat{\bar{\boldsymbol{p}}}_\alpha^2}{2m_\alpha} + \sum_{\substack{\alpha,\beta\in A\\ \alpha\neq\beta}} \frac{q_\alpha q_\beta}{8\pi\varepsilon_0|\hat{\bar{\boldsymbol{r}}}_\alpha - \hat{\bar{\boldsymbol{r}}}_\beta|} = \frac{\hat{\boldsymbol{p}}_A^2}{2m_A} + \sum_n E_n|n\rangle\langle n| \tag{2.313}$$

with E_n and $|n\rangle$ denoting the eigenenergies and eigenstates of the internal Hamiltonian.

As a consequence of the continuity equation, the atomic charge and current densities can be related to the atomic polarisation and magnetisation, recall the discussion at the beginning of Sect. 2.1.3. Defining these quantities as

$$\hat{\boldsymbol{P}}_A(\boldsymbol{r}) = \sum_{\alpha\in A} q_\alpha\hat{\bar{\boldsymbol{r}}}_\alpha \int_0^1 \mathrm{d}\sigma\,\delta\big(\boldsymbol{r} - \hat{\boldsymbol{r}}_A - \sigma\hat{\bar{\boldsymbol{r}}}_\alpha\big)\,, \tag{2.314}$$

$$\hat{\boldsymbol{M}}_A(\boldsymbol{r}) = \sum_{\alpha\in A} q_\alpha \int_0^1 \mathrm{d}\sigma\,\sigma\mathcal{S}\Big[\hat{\bar{\boldsymbol{r}}}_\alpha \times \dot{\hat{\bar{\boldsymbol{r}}}}_\alpha\delta\big(\boldsymbol{r} - \hat{\boldsymbol{r}}_A - \sigma\hat{\bar{\boldsymbol{r}}}_\alpha\big)\Big]\,, \tag{2.315}$$

one has

$$\hat{\rho}_A = -\nabla \cdot \hat{\boldsymbol{P}}_A \,, \tag{2.316}$$

$$\hat{\boldsymbol{j}}_A = \dot{\hat{\boldsymbol{P}}}_A + \nabla \times \hat{\boldsymbol{M}}_A + \hat{\boldsymbol{j}}_{\mathrm{R}} \,. \tag{2.317}$$

Note that the Röntgen current density [29, 30]

$$\hat{\boldsymbol{j}}_{\mathrm{R}}(\boldsymbol{r}) = \nabla \times \mathcal{S}[\hat{\boldsymbol{P}}_A(\boldsymbol{r}) \times \dot{\hat{\boldsymbol{r}}}_A] \,, \tag{2.318}$$

which is due to the centre-of-mass motion of the atom, may alternatively be included in the contribution from the magnetisation in order to make the analogy with the classical relation (2.90) more complete. When combined with the Poisson equation, the relation (2.316) implies that

$$\nabla \hat{\phi}_A = \frac{1}{\varepsilon_0} \hat{\boldsymbol{P}}_A^{\parallel} \,. \tag{2.319}$$

Within leading order of the relative particle coordinates, polarisation and magnetisation are well described by the electric and magnetic dipole moments of the atom,

$$\hat{\boldsymbol{d}} = \sum_{\alpha \in A} q_\alpha \hat{\overline{\boldsymbol{r}}}_\alpha = \sum_{\alpha \in A} q_\alpha \hat{\boldsymbol{r}}_\alpha \,, \tag{2.320}$$

$$\hat{\boldsymbol{m}} = \sum_{\alpha \in A} \frac{q_\alpha}{2} \hat{\overline{\boldsymbol{r}}}_\alpha \times \dot{\hat{\overline{\boldsymbol{r}}}}_\alpha \,, \tag{2.321}$$

where we have made use of the fact that the atom is neutral.

A duality transformation (2.184) applies to the atomic polarisation and magnetisation in the same way as it does to a macroscopic polarisation and magnetisation. In particular, application of the discrete transformation $\mathcal{D}_1$ results in

$$\hat{\boldsymbol{P}}_A^{\circledast} = \hat{\boldsymbol{M}}_A / c \,, \qquad \hat{\boldsymbol{M}}_A^{\circledast} = -c \hat{\boldsymbol{P}}_A \,. \tag{2.322}$$

The transformation behaviour of the dipole moments follows immediately:

$$\hat{\boldsymbol{d}}^{\circledast} = \hat{\boldsymbol{m}} / c \,, \qquad \hat{\boldsymbol{m}}^{\circledast} = -c \hat{\boldsymbol{d}} \,. \tag{2.323}$$

Finally, note that using the atomic Hamiltonian (2.313) together with the commutation relations (2.311) and the definition (2.309), one can easily verify the useful relation

$$\sum_{\alpha \in A} \frac{q_\alpha}{m_\alpha} \langle m | \hat{\overline{\boldsymbol{p}}}_\alpha | n \rangle = \mathrm{i} \omega_{mn} \boldsymbol{d}_{mn} \tag{2.324}$$

with $\omega_{mn} = (E_m - E_n)/\hbar$ and $\boldsymbol{d}_{mn} = \langle m | \hat{\boldsymbol{d}} | n \rangle$. It implies the Thomas–Reiche–Kuhn sum rule [31–33]

$$\frac{1}{2\hbar}\sum_k \omega_{kn}(\boldsymbol{d}_{nk}\,\boldsymbol{d}_{kn} + \boldsymbol{d}_{kn}\,\boldsymbol{d}_{nk}) = \sum_{\alpha\in A}\frac{q_\alpha^2}{2m_\alpha}\,\boldsymbol{I}\,. \tag{2.325}$$

2.4.1 Minimal Coupling

As in the classical case, there is a mutual interaction between the body-assisted field and the charged particles constituting the atom: The atomic charges act as sources in the Maxwell equations for the electromagnetic field which in turn influences their motion via Lorentz forces. In the classical theory, we have studied the coupled dynamics by working in Fourier space. In quantum theory, however, the time-dependent frequency components of the electromagnetic field, as prescribed by an appropriate Hamiltonian, do not necessarily coincide with ordinary Fourier components, so a Fourier analysis is not particularly well adapted to the quantum dynamics. Instead, we construct the interacting theory from our separate descriptions of the body-assisted electromagnetic field and the atom by means of the minimal coupling scheme: In close analogy to the free-space case, the Hamiltonian of the interacting atom–field system is obtained by summing the separate Hamiltonians $\hat{H}_A$ and $\hat{H}_\mathrm{F}$, making the replacement $\hat{\boldsymbol{p}}_\alpha \mapsto \hat{\boldsymbol{p}}_\alpha - q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)$ and adding the Coulomb interaction of the atom with the body-assisted field. One obtains [5, 6, 11–13, 34]

$$\begin{aligned}\hat{H} &= \sum_{\alpha\in A}\frac{\left[\hat{\boldsymbol{p}}_\alpha - q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\right]^2}{2m_\alpha} + \sum_{\substack{\alpha,\beta\in A\\ \alpha\neq\beta}}\frac{q_\alpha q_\beta}{8\pi\varepsilon_0|\hat{\boldsymbol{r}}_\alpha - \hat{\boldsymbol{r}}_\beta|}\\ &\quad + \sum_{\lambda=e,m}\int \mathrm{d}^3r\int_0^\infty \mathrm{d}\omega\,\hbar\omega\,\hat{\boldsymbol{f}}_\lambda^\dagger(\boldsymbol{r},\omega)\cdot\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega) + \sum_{\alpha\in A}q_\alpha\hat{\phi}(\hat{\boldsymbol{r}}_\alpha)\\ &= \hat{H}_A + \hat{H}_\mathrm{F} + \hat{H}_\mathrm{AF}\end{aligned} \tag{2.326}$$

where $\hat{H}_A$ and $\hat{H}_\mathrm{F}$ are given by (2.313) and (2.271), respectively, and the atom–field interaction reads

$$\hat{H}_\mathrm{AF} = \sum_{\alpha\in A}q_\alpha\hat{\phi}(\hat{\boldsymbol{r}}_\alpha) - \sum_{\alpha\in A}\frac{q_\alpha}{m_\alpha}\,\hat{\boldsymbol{p}}_\alpha\cdot\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha) + \sum_{\alpha\in A}\frac{q_\alpha^2}{2m_\alpha}\,\hat{\boldsymbol{A}}^2(\hat{\boldsymbol{r}}_\alpha)\,, \tag{2.327}$$

note that the scalar product of $\hat{\boldsymbol{p}}_\alpha$ and $\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)$ commutes in the Coulomb gauge. The expansions (2.259), (2.263), (2.264) and (2.265) for the fields $\hat{\boldsymbol{E}}$, $\hat{\boldsymbol{B}}$, $\hat{\boldsymbol{D}}$ and $\hat{\boldsymbol{H}}$ remain valid. Due to the presence of the atom, not all of them coincide with the physical electric/magnetic fields and excitations, which for distinctness are denoted by curly letters and are given by

$$\hat{\boldsymbol{\mathcal{E}}} = \hat{\boldsymbol{E}} - \nabla\hat{\phi}_A\,, \qquad \hat{\boldsymbol{\mathcal{B}}} = \hat{\boldsymbol{B}}\,, \tag{2.328}$$

$$\hat{\boldsymbol{\mathcal{D}}} = \hat{\boldsymbol{D}} - \varepsilon_0\nabla\hat{\phi}_A\,, \quad \hat{\boldsymbol{\mathcal{H}}} = \hat{\boldsymbol{H}}\,. \tag{2.329}$$

To verify the correctness of the above Hamiltonian, we have to show that the physical fields obey the Maxwell equations

$$\nabla\cdot\hat{\boldsymbol{\mathcal{B}}} = 0\,, \tag{2.330}$$

$$\nabla\cdot\,\hat{\boldsymbol{\mathcal{D}}} = \hat{\rho}_A\,, \tag{2.331}$$

$$\nabla\times\,\hat{\boldsymbol{\mathcal{E}}} + \dot{\hat{\boldsymbol{\mathcal{B}}}} = \boldsymbol{0}\,, \tag{2.332}$$

$$\nabla\times\,\hat{\boldsymbol{\mathcal{H}}} - \dot{\hat{\boldsymbol{\mathcal{D}}}} = \hat{\boldsymbol{j}}_A \tag{2.333}$$

and that the motion of the particles constituting the atom is given by the Newton equations

$$m_\alpha\ddot{\hat{\boldsymbol{r}}}_\alpha = q_\alpha\hat{\boldsymbol{\mathcal{E}}}(\hat{\boldsymbol{r}}_\alpha) + q_\alpha\mathcal{S}\Big[\dot{\hat{\boldsymbol{r}}}_\alpha\times\hat{\boldsymbol{\mathcal{B}}}(\hat{\boldsymbol{r}}_\alpha)\Big]. \tag{2.334}$$

The canonical commutation relations for $\hat{\boldsymbol{r}}_\alpha$ and $\hat{\boldsymbol{p}}_\alpha$ imply that the particle velocities are given by

$$\dot{\hat{\boldsymbol{r}}}_\alpha = \frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{r}}_\alpha, \hat{H}\big] = \frac{1}{m_\alpha}\left[\hat{\boldsymbol{p}}_\alpha - q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\right], \tag{2.335}$$

in close analogy with the classical relation (2.66). Let us begin with the Maxwell equations. The first Maxwell equation follows directly from the expansion (2.263),

$$\begin{aligned}\nabla\cdot\hat{\boldsymbol{\mathcal{B}}}(\boldsymbol{r}) &= \sum_{\lambda=e,m}\int_0^\infty\frac{\mathrm{d}\omega}{\mathrm{i}\omega}\int\mathrm{d}^3r'\,\nabla\cdot\nabla\times\boldsymbol{G}_\lambda(\boldsymbol{r},\boldsymbol{r}',\omega)\cdot\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}',\omega) + \text{H.c.}\\ &= 0\,.\end{aligned} \tag{2.336}$$

It should be stressed again that due to the influence of the atom–field coupling, the time-dependent frequency components $\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega,t) \equiv \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega)$ in the Heisenberg picture are not ordinary Fourier components (2.273) in the presence of the atom. Similarly, the expansion (2.264) leads to the Gauss law

$$\nabla \cdot \hat{\boldsymbol{\mathcal{D}}}(\boldsymbol{r}) = \int_0^\infty \mathrm{d}\omega \left[\varepsilon_0 \nabla \cdot \varepsilon(\boldsymbol{r}, \omega) \sum_{\lambda=e,m} \int \mathrm{d}^3 r' \, \boldsymbol{G}_\lambda(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}', \omega) \right.$$
$$\left. + \mathrm{i}\sqrt{\frac{\hbar\varepsilon_0}{\pi} \operatorname{Im} \varepsilon(\boldsymbol{r}, \omega)}\, \hat{\boldsymbol{f}}_e(\boldsymbol{r}, \omega, t) \right] + \mathrm{H.c.} - \varepsilon_0 \Delta \hat{\phi}_A(\boldsymbol{r}) = \hat{\rho}_A(\boldsymbol{r}) \tag{2.337}$$

where we have recalled definitions (2.260) and (2.261) together with the identity (2.158) to realize that the contribution from the terms in the square brackets is vanishing; and we have invoked the Poisson equation $-\varepsilon_0 \Delta \hat{\phi}_A = \hat{\rho}_A$. The Faraday law can be verified by calculating the time derivative according to the Heisenberg equation of motion. Since $\hat{\boldsymbol{B}}$ commutes with $\hat{\boldsymbol{A}}$ [recall (2.294) and (2.287)] and $\hat{\phi}$ (cf. App. A) and hence also with $\hat{H}_{\mathrm{AF}}$, the expansion (2.263) leads to

$$\begin{aligned}
\dot{\hat{\boldsymbol{\mathcal{B}}}}(\boldsymbol{r}) &= \frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{B}}(\boldsymbol{r}), \hat{H}_{\mathrm{F}}\big] \\
&= \sum_{\lambda=e,m} \int_0^\infty \frac{\mathrm{d}\omega}{\mathrm{i}\omega} \int \mathrm{d}^3 r' \, \nabla \times \boldsymbol{G}_\lambda(\boldsymbol{r}, \boldsymbol{r}', \omega) \cdot \big[-\mathrm{i}\omega \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r}', \omega)\big] + \mathrm{H.c.} \\
&= -\nabla \times \hat{\boldsymbol{\mathcal{E}}}(\boldsymbol{r})
\end{aligned} \tag{2.338}$$

where we have used the commutator (2.272). The Ampère law is a bit more involved since $\dot{\hat{\boldsymbol{\mathcal{D}}}}$ has several contributions:

$$\begin{aligned}
\dot{\hat{\boldsymbol{\mathcal{D}}}}(\boldsymbol{r}) &= \frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{D}}(\boldsymbol{r}), \hat{H}_{\mathrm{F}}\big] + \frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{D}}(\boldsymbol{r}), \hat{H}_{\mathrm{AF}}\big] \\
&\quad - \frac{1}{\mathrm{i}\hbar}\big[\varepsilon_0 \nabla \hat{\phi}_A(\boldsymbol{r}), \hat{H}_A\big] - \frac{1}{\mathrm{i}\hbar}\big[\varepsilon_0 \nabla \hat{\phi}_A(\boldsymbol{r}), \hat{H}_{\mathrm{AF}}\big].
\end{aligned} \tag{2.339}$$

Using the commutator (2.272) and the relation (2.234), one finds

$$\frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{D}}(\boldsymbol{r}), \hat{H}_{\mathrm{F}}\big] = -\int_0^\infty \mathrm{d}\omega \, \mathrm{i}\omega \underline{\hat{\boldsymbol{D}}}(\boldsymbol{r}, \omega) + \mathrm{H.c.} = \nabla \times \hat{\boldsymbol{\mathcal{H}}}(\boldsymbol{r}) \tag{2.340}$$

The commutator of $\hat{\boldsymbol{D}}$ with $\hat{H}_{\mathrm{AF}}$ can be found by noting that $\hat{\boldsymbol{D}}$ commutes with $\hat{\phi}$ while

$$\big[\hat{\boldsymbol{D}}(\boldsymbol{r}), \hat{\boldsymbol{A}}(\boldsymbol{r}')\big] = \mathrm{i}\hbar \boldsymbol{\delta}^\perp(\boldsymbol{r} - \boldsymbol{r}') \tag{2.341}$$

(cf. App. A), so that

$$\frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{D}}(\boldsymbol{r}), \hat{H}_{\mathrm{AF}}\big] = -\sum_{\alpha \in A} \frac{q_\alpha}{m_\alpha}\big[\hat{\boldsymbol{p}}_\alpha - q_\alpha \hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\big] \cdot \boldsymbol{\delta}^\perp(\boldsymbol{r} - \hat{\boldsymbol{r}}_\alpha) = -\hat{\boldsymbol{j}}_A^\perp(\boldsymbol{r}). \tag{2.342}$$

The contributions due to $\hat{\phi}_A$ as given by (2.305) can be calculated by means of the canonical commutation relations for $\hat{\boldsymbol{r}}_\alpha$ and $\hat{\boldsymbol{p}}_\alpha$. From the Hamiltonians (2.297) and (2.327), we find

$$-\frac{1}{\mathrm{i}\hbar}\big[\varepsilon_0\nabla\hat{\phi}_A(\boldsymbol{r}),\hat{H}_A\big] = -\sum_{\alpha\in A}\left[\frac{\hat{\boldsymbol{p}}_\alpha}{2m_\alpha}\cdot\boldsymbol{\delta}^{\parallel}(\boldsymbol{r}-\hat{\boldsymbol{r}}_\alpha)+\boldsymbol{\delta}^{\parallel}(\boldsymbol{r}-\hat{\boldsymbol{r}}_\alpha)\cdot\frac{\hat{\boldsymbol{p}}_\alpha}{2m_\alpha}\right], \tag{2.343}$$

$$-\frac{1}{\mathrm{i}\hbar}\big[\varepsilon_0\nabla\hat{\phi}_A(\boldsymbol{r}),\hat{H}_{\mathrm{AF}}\big] = \sum_{\alpha\in A}\frac{q_\alpha}{m_\alpha}\,\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\cdot\boldsymbol{\delta}^{\parallel}(\boldsymbol{r}-\hat{\boldsymbol{r}}_\alpha) \tag{2.344}$$

[recall definition (2.15) of the longitudinal delta function], so that

$$-\frac{1}{\mathrm{i}\hbar}\big[\varepsilon_0\nabla\hat{\phi}_A(\boldsymbol{r}),\hat{H}_A\big]-\frac{1}{\mathrm{i}\hbar}\big[\varepsilon_0\nabla\hat{\phi}_A(\boldsymbol{r}),\hat{H}_{\mathrm{AF}}\big] = -\hat{\boldsymbol{j}}_A^{\parallel}(\boldsymbol{r})\,, \tag{2.345}$$

cf. (2.335) above. Combining these results, we finally obtain the Ampère law.

Next, we address the Newton equation (2.334) by considering the Heisenberg equation of motion for $\dot{\hat{\boldsymbol{r}}}_\alpha$. The relation $m_\alpha\dot{\hat{\boldsymbol{r}}}_\alpha = \hat{\boldsymbol{p}}_\alpha - q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)$ together with the Hamiltonian (2.335) shows that we have to calculate five contributions to the Lorentz force:

$$\begin{aligned} m_\alpha\ddot{\hat{\boldsymbol{r}}}_\alpha = \frac{1}{\mathrm{i}\hbar}\big[m_\alpha\dot{\hat{\boldsymbol{r}}}_\alpha,\hat{H}\big] &= \frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{p}}_\alpha,\hat{H}_A\big]+\frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{p}}_\alpha,\hat{H}_{\mathrm{AF}}\big]-\frac{1}{\mathrm{i}\hbar}\big[q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha),\hat{H}_A\big] \\ &\quad -\frac{1}{\mathrm{i}\hbar}\big[q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha),\hat{H}_{\mathrm{F}}\big]-\frac{1}{\mathrm{i}\hbar}\big[q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha),\hat{H}_{\mathrm{AF}}\big]. \end{aligned} \tag{2.346}$$

Four of these follow directly from the Hamiltonians (2.297) and (2.327) together with the canonical commutations relations for $\hat{\boldsymbol{r}}_\alpha$ and $\hat{\boldsymbol{p}}_\alpha$,

$$\frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{p}}_\alpha,\hat{H}_A\big] = -q_\alpha\nabla\hat{\phi}_A(\hat{\boldsymbol{r}}_\alpha), \tag{2.347}$$

$$\frac{1}{\mathrm{i}\hbar}\big[\hat{\boldsymbol{p}}_\alpha,\hat{H}_{\mathrm{AF}}\big] = q_\alpha\hat{\boldsymbol{E}}^{\parallel}(\hat{\boldsymbol{r}}_\alpha)+q_\alpha\mathcal{S}\big\{\big[\nabla\,\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\big]\cdot\dot{\hat{\boldsymbol{r}}}_\alpha\big\}, \tag{2.348}$$

$$-\frac{1}{\mathrm{i}\hbar}\big[q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha),\hat{H}_A\big] = -\frac{q_\alpha}{m_\alpha}\,\mathcal{S}\big\{\hat{\boldsymbol{p}}_\alpha\cdot\big[\nabla\,\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\big]\big\}, \tag{2.349}$$

$$-\frac{1}{\mathrm{i}\hbar}\big[q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha),\hat{H}_{\mathrm{AF}}\big] = \frac{q_\alpha^2}{m_\alpha}\,\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\cdot\big[\nabla\,\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\big], \tag{2.350}$$

where we have recalled $\hat{\boldsymbol{E}}^{\parallel} = -\nabla\hat{\phi}$ and $m_\alpha\dot{\hat{\boldsymbol{r}}}_\alpha = \hat{\boldsymbol{p}}_\alpha - q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)$ and exploited the fact that $\hat{\boldsymbol{A}}$ and $\hat{\phi}$ commute (App. A). The remaining term in (2.346) follows trivially from the commutator (2.272) upon comparison of definitions (2.259) and (2.290)

$$-\frac{1}{\mathrm{i}\hbar}\big[q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha),\hat{H}_\mathrm{F}\big]=q_\alpha\hat{\boldsymbol{E}}^\perp(\hat{\boldsymbol{r}}_\alpha)\,. \tag{2.351}$$

Combining these results by using the rule $\boldsymbol{a}\times(\boldsymbol{b}\times\boldsymbol{c})=\boldsymbol{b}(\boldsymbol{a}\cdot\boldsymbol{c})-\boldsymbol{c}(\boldsymbol{a}\cdot\boldsymbol{b})$ and recalling the relations $\hat{\boldsymbol{\mathcal{E}}}=\hat{\boldsymbol{E}}-\nabla\hat{\phi}_A$ and $\hat{\boldsymbol{\mathcal{B}}}=\hat{\boldsymbol{B}}=\nabla\times\hat{\boldsymbol{A}}$, we find that the Newton equation (2.334) is valid.

Macroscopic QED for a single atom interacting with the body-assisted electromagnetic field has thus been successfully established by obtaining the total Hamiltonian of system via the minimal coupling scheme and relating the electromagnetic field to the expansions in terms of fundamental fields as obtained for the non-interacting case. Some properties of the interacting electromagnetic field are directly inherited from the case of no atom being present; in particular, the fundamental equal-time commutation relations (2.266) and (2.267) as well as the fluctuation–dissipation theorem (2.268) remain valid. Furthermore, the dynamics of the theory is in accordance with the Maxwell equations for the electromagnetic field in the presence of charged particles and the Newton equations for the motion of the particles under the influence of the field.

In most cases of practical interest one may assume that the atom is small compared to the wavelength of the relevant electromagnetic field. It is then useful to apply the leading-order long-wavelength approximation by performing a leading-order expansion of the interaction Hamiltonian (2.327) in terms of the relative particle coordinates (2.308). For a neutral atom, this results in

$$\hat{H}_\mathrm{AF}=-\hat{\boldsymbol{d}}\cdot\hat{\boldsymbol{E}}^\parallel(\hat{\boldsymbol{r}}_A)-\sum_{\alpha\in A}\frac{q_\alpha}{m_\alpha}\hat{\overline{\boldsymbol{p}}}_\alpha\cdot\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_A)+\sum_{\alpha\in A}\frac{q_\alpha^2}{2m_\alpha}\hat{\boldsymbol{A}}^2(\hat{\boldsymbol{r}}_A)\,. \tag{2.352}$$

In the minimal coupling scheme, the leading-order long-wavelength approximation may alternatively be referred to as an electric-dipole approximation because results obtained from $\hat{H}_\mathrm{AF}$ as given above generally only depend on the electric dipole moment of the atom. Note that the last term of the interaction Hamiltonian has become independent of the relative particle coordinates, hence it does not affect the internal state of the atom. When considering processes caused by resonant transitions between different internal states of the atom, it may therefore be neglected.

2.4.2 Multipolar Coupling

An equivalent description of the atom–field dynamics that is widely used, is the multipolar coupling scheme. The multipolar-coupling Hamiltonian can be obtained from the minimal coupling form by means of a Power–Zienau–Woolley transformation [35–37]

$$\hat{f}'=\hat{U}\hat{f}\hat{U}^\dagger\quad\text{with }\hat{U}=\exp\left[\frac{\mathrm{i}}{\hbar}\int\mathrm{d}^3r\,\hat{\boldsymbol{P}}_A\cdot\hat{\boldsymbol{A}}\right]. \tag{2.353}$$

The unitarity of the transformation (together with the fact that all relevant equal-time commutators are c-numbers) implies that the transformed variables obey the same equal-time commutation relations as the unprimed ones. In particular, (2.266), (2.267), (2.294) and (2.295) for the electromagnetic field, the bosonic commutation relations (2.242) and (2.243) for the fundamental fields as well as (2.296) for the particles also hold for the transformed variables. Unitarity further implies that the expansions of the electromagnetic fields in terms of the fundamental fields remain valid after the transformation, i.e., (2.259), (2.263)–(2.265), (2.289) and (2.290) apply with primed operators instead of unprimed ones. Similarly, the definitions of derived atomic quantities, (2.298), (2.299), (2.305), (2.308), (2.309), (2.314),(2.315), (2.318), (2.320) and (2.321) hold for the primed operators.

Explicit expressions for the transformed variables in terms of the untransformed ones can be calculated with the aid of the operator identity

$$\mathrm{e}^{\hat{a}}\hat{b}\,\mathrm{e}^{-\hat{a}} = \hat{b} + \big[\hat{a},\hat{b}\big] + \frac{1}{2!}\big[\hat{a},\big[\hat{a},\hat{b}\big]\big] + \cdots \tag{2.354}$$

The following quantities remain unchanged, because they commute with both $\hat{\boldsymbol{r}}_\alpha$ [and hence with $\hat{\boldsymbol{P}}_A$, recall (2.314)] and $\hat{\boldsymbol{A}}$ [recall (2.294) and note (A.21), (A.23)]:

$$\hat{\boldsymbol{B}}' = \hat{\boldsymbol{B}}\,, \qquad \hat{\boldsymbol{A}}' = \hat{\boldsymbol{A}}\,, \qquad \hat{\phi}' = \hat{\phi}\,, \tag{2.355}$$

$$\hat{\boldsymbol{r}}'_\alpha = \hat{\boldsymbol{r}}_\alpha\,, \qquad \hat{\boldsymbol{r}}'_A = \hat{\boldsymbol{r}}_A\,, \qquad \hat{\bar{\boldsymbol{r}}}'_\alpha = \hat{\bar{\boldsymbol{r}}}_\alpha\,, \tag{2.356}$$

$$\hat{\rho}'_A = \hat{\rho}_A\,, \qquad \hat{\phi}'_A = \hat{\phi}_A\,, \qquad \hat{\boldsymbol{P}}'_A = \hat{\boldsymbol{P}}_A\,. \tag{2.357}$$

The Heisenberg dynamics (2.225) still being given by the same Hamiltonian (2.326), it follows that time derivatives of the above quantities are also unaffected by the transformation, hence

$$\dot{\hat{\boldsymbol{r}}}'_\alpha = \dot{\hat{\boldsymbol{r}}}_\alpha\,, \qquad \dot{\hat{\boldsymbol{r}}}'_A = \dot{\hat{\boldsymbol{r}}}_A\,, \qquad \dot{\hat{\bar{\boldsymbol{r}}}}'_\alpha = \dot{\hat{\bar{\boldsymbol{r}}}}_\alpha\,, \tag{2.358}$$

$$\hat{\boldsymbol{m}}'_A = \hat{\boldsymbol{M}}_A\,, \qquad \hat{\boldsymbol{j}}'_A = \hat{\boldsymbol{j}}_A\,, \qquad \hat{\boldsymbol{j}}'_\mathrm{R} = \hat{\boldsymbol{j}}_\mathrm{R}\,. \tag{2.359}$$

Next, we turn to those operators which do not commute with both $\hat{\boldsymbol{P}}_A$ and $\hat{\boldsymbol{A}}$ and are hence affected by the Power–Zienau–Woolley transformation in a nontrivial way. Upon invoking the commutation relation (A.12) for $\hat{\boldsymbol{A}}$ and $\hat{\boldsymbol{E}}$, the rule (2.354) implies that

$$\hat{\boldsymbol{E}}' = \hat{\boldsymbol{E}} + \frac{1}{\varepsilon_0}\,\hat{\boldsymbol{P}}_A^\perp\,. \tag{2.360}$$

The canonically conjugate field momentum $\hat{\boldsymbol{\Pi}} = -\varepsilon_0\hat{\boldsymbol{E}}^\perp$ thus transforms as

$$\hat{\boldsymbol{\Pi}}' = \hat{\boldsymbol{\Pi}} - \hat{\boldsymbol{P}}_A^\perp\,. \tag{2.361}$$

Similarly, the transformed fundamental fields follow from their bosonic commutation relations (2.242) and (2.243); upon using the definition (2.290) of $\hat{\boldsymbol{A}}$, one finds

$$\hat{\boldsymbol{f}}'_\lambda(\boldsymbol{r},\omega) = \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega) + \frac{1}{\hbar\omega}\int \mathrm{d}^3r'\, \hat{\boldsymbol{P}}_A^\perp(\boldsymbol{r}') \cdot \boldsymbol{G}^*_\lambda(\boldsymbol{r}',\boldsymbol{r},\omega)\,. \tag{2.362}$$

The transformation of the canonically conjugate particle momenta is slightly more involved. Upon recalling definition (2.314) of $\hat{\boldsymbol{P}}_A$, application of the commutation relations (2.296) and use of the rule $\boldsymbol{a}\times(\boldsymbol{b}\times\boldsymbol{c}) = \boldsymbol{b}(\boldsymbol{a}\cdot\boldsymbol{c}) - \boldsymbol{c}(\boldsymbol{a}\cdot\boldsymbol{b})$ results in

$$\begin{aligned}
\hat{\boldsymbol{p}}'_\alpha = \hat{\boldsymbol{p}}_\alpha &- \sum_{\beta\in A} q_\beta \left(\delta_{\alpha\beta} - \frac{m_\alpha}{m_A}\right)\int_0^1 \mathrm{d}\sigma\, \hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_A + \sigma\hat{\bar{\boldsymbol{r}}}_\beta) \\
&+ \int \mathrm{d}^3r \sum_{\beta\in A} q_\beta \int_0^1 \mathrm{d}\sigma \left[\sigma\delta_{\alpha\beta} + (1-\sigma)\frac{m_\alpha}{m_A}\right] \hat{\boldsymbol{A}}(\boldsymbol{r})\big(\hat{\bar{\boldsymbol{r}}}_\beta\cdot\nabla\big)\delta\big(\boldsymbol{r}-\hat{\boldsymbol{r}}_A-\sigma\hat{\bar{\boldsymbol{r}}}_\beta\big) \\
&- \int \mathrm{d}^3r \sum_{\beta\in A} q_\beta \int_0^1 \mathrm{d}\sigma \left[\sigma\delta_{\alpha\beta} + (1-\sigma)\frac{m_\alpha}{m_A}\right] \delta\big(\boldsymbol{r}-\hat{\boldsymbol{r}}_A-\sigma\hat{\bar{\boldsymbol{r}}}_\beta\big)\,\hat{\bar{\boldsymbol{r}}}_\beta\times\hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}) \\
= \hat{\boldsymbol{p}}_\alpha &- q_\alpha\hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha) - \int \mathrm{d}^3r\, \hat{\boldsymbol{\varXi}}_\alpha \times \hat{\boldsymbol{B}}
\end{aligned} \tag{2.363}$$

with

$$\begin{aligned}
\hat{\boldsymbol{\varXi}}_\alpha(\boldsymbol{r}) = \hat{\boldsymbol{\varXi}}'_\alpha(\boldsymbol{r}) = q_\alpha\hat{\bar{\boldsymbol{r}}}_\alpha &\int_0^1 \mathrm{d}\sigma\,\sigma\delta\big(\boldsymbol{r}-\hat{\boldsymbol{r}}_A-\sigma\hat{\bar{\boldsymbol{r}}}_\alpha\big) \\
&- \frac{m_\alpha}{m_A}\sum_{\beta\in A} q_\beta\hat{\bar{\boldsymbol{r}}}_\beta \int_0^1 \mathrm{d}\sigma\,\sigma\delta\big(\boldsymbol{r}-\hat{\boldsymbol{r}}_A-\sigma\hat{\bar{\boldsymbol{r}}}_\beta\big) + \frac{m_\alpha}{m_A}\hat{\boldsymbol{P}}_A(\boldsymbol{r})\,.
\end{aligned} \tag{2.364}$$

The second equality in (2.363) has been obtained by partially integrating with respect to σ and exploiting the fact that the atom is neutral. The derived transformation rules can be summarised by noting that the Power–Zienau–Woolley transformation affects the canonically conjugate momenta $\hat{\boldsymbol{\varPi}}$ and $\hat{\boldsymbol{p}}_\alpha$ while leaving $\hat{\boldsymbol{A}}$ and $\hat{\boldsymbol{r}}_\alpha$ unchanged.

In the multipolar coupling scheme, the transformed variables are used as basic variables instead of the original ones. In particular, the multipolar Hamiltonian is obtained by expressing the Hamiltonian (2.326) of the system in terms of these new variables. We start by applying the above transformation rule for the fundamental fields to calculate

$$\sum_{\lambda=e,m}\int \mathrm{d}^3r\int_0^\infty \mathrm{d}\omega\,\hbar\omega\,\hat{\boldsymbol{f}}^\dagger_\lambda(\boldsymbol{r},\omega)\cdot\hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega)$$
$$=\sum_{\lambda=e,m}\int \mathrm{d}^3r\int_0^\infty \mathrm{d}\omega\,\hbar\omega\,\hat{\boldsymbol{f}}'^\dagger_\lambda(\boldsymbol{r},\omega)\cdot\hat{\boldsymbol{f}}'_\lambda(\boldsymbol{r},\omega)$$
$$-\int \mathrm{d}^3r\,\hat{\boldsymbol{P}}'^\perp_A\cdot\hat{\boldsymbol{E}}'+\frac{1}{2\varepsilon_0}\int \mathrm{d}^3r\big(\hat{\boldsymbol{P}}'^\perp_A\big)^2\,; \tag{2.365}$$

where the expansion (2.259) of $\hat{\boldsymbol{E}}'$ has been recalled and the identities (2.262) and (A.11) have been employed. As this result depends on $\hat{\boldsymbol{P}}'^\perp_A$ and $\hat{\boldsymbol{E}}'$, the re-expressed Hamiltonian assumes its most compact form by writing the other terms of the Hamiltonian in terms of these fields as well. Combining the identities (2.307) and $\varepsilon_0\nabla\hat{\phi}_A = \hat{\boldsymbol{P}}^\parallel_A$ and using the fact that $\hat{\boldsymbol{P}}_A$ is not affected by the Power–Zienau–Woolley transformation, the atomic Coulomb interaction can be written as

$$\sum_{\substack{\alpha,\beta\in A\\ \alpha\neq\beta}}\frac{q_\alpha q_\beta}{8\pi\varepsilon_0|\hat{\boldsymbol{r}}_\alpha-\hat{\boldsymbol{r}}_\beta|}=\frac{1}{2\varepsilon_0}\int \mathrm{d}^3r\big(\hat{\boldsymbol{P}}^\parallel_A\big)^2=\frac{1}{2\varepsilon_0}\int \mathrm{d}^3r\big(\hat{\boldsymbol{P}}'^\parallel_A\big)^2\,. \tag{2.366}$$

Similarly, upon noting that $\nabla\hat{\phi}=-\hat{\boldsymbol{E}}^\parallel$, the Coulomb interaction of the atom with the body-assisted field may be rewritten as

$$\sum_{\alpha\in A}q_\alpha\hat{\phi}(\hat{\boldsymbol{r}}_\alpha)=\int \mathrm{d}^3r\,\hat{\rho}_A\hat{\phi}=-\int \mathrm{d}^3r\,\hat{\boldsymbol{P}}^\parallel_A\cdot\hat{\boldsymbol{E}}^\parallel=-\int \mathrm{d}^3r\,\hat{\boldsymbol{P}}'^\parallel_A\cdot\hat{\boldsymbol{E}}'^\parallel\,, \tag{2.367}$$

as both $\hat{\boldsymbol{P}}_A$ and $\hat{\boldsymbol{E}}^\parallel$ are invariant under the transformation. Combining the results (2.363) and (2.365)–(2.367), the Hamiltonian (2.326) can be expressed in terms of the new variables. Noting that integrals over mixed scalar products of longitudinal/transverse vector field vanish, the resulting multipolar-coupling form of the same Hamiltonian reads [5, 6, 11–13, 34]

$$\begin{aligned}\hat{H} &= \sum_{\lambda=e,m} \int \mathrm{d}^3r \int_0^\infty \mathrm{d}\omega\, \hbar\omega\, \hat{\boldsymbol{f}}_\lambda'^\dagger(\boldsymbol{r},\omega) \cdot \hat{\boldsymbol{f}}_\lambda'(\boldsymbol{r},\omega) \\ &\quad + \sum_{\alpha\in A} \frac{1}{2m_\alpha} \mathcal{S}\left[\hat{\boldsymbol{p}}_\alpha' + \int \mathrm{d}^3r\, \hat{\boldsymbol{\Xi}}_\alpha' \times \hat{\boldsymbol{B}}'\right]^2 \\ &\quad + \frac{1}{2\varepsilon_0} \int \mathrm{d}^3r\, \hat{\boldsymbol{P}}_A'^2 - \int \mathrm{d}^3r\, \hat{\boldsymbol{P}}_A' \cdot \hat{\boldsymbol{E}}' \\ &= \hat{H}_A' + \hat{H}_\mathrm{F}' + \hat{H}_\mathrm{AF}' \end{aligned} \tag{2.368}$$

In the multipolar coupling scheme, the atomic, field and interaction parts of the Hamiltonian are given by

$$\begin{aligned}\hat{H}_A' &= \sum_{\alpha\in A} \frac{\hat{\boldsymbol{p}}_\alpha'^2}{2m_\alpha} + \frac{1}{2\varepsilon_0} \int \mathrm{d}^3r\, \hat{\boldsymbol{P}}_A'^2 \\ &= \frac{\hat{\boldsymbol{p}}_A'^2}{2m_A} + \sum_{\alpha\in A} \frac{\hat{\overline{\boldsymbol{p}}}_\alpha'^2}{2m_\alpha} + \frac{1}{2\varepsilon_0} \int \mathrm{d}^3r\, \hat{\boldsymbol{P}}_A'^2 \\ &= \frac{\hat{\boldsymbol{p}}_A'^2}{2m_A} + \sum_n E_n' |n'\rangle\langle n'|\,, \end{aligned} \tag{2.369}$$

$$\hat{H}_\mathrm{F}' = \sum_{\lambda=e,m} \int \mathrm{d}^3r \int_0^\infty \mathrm{d}\omega\, \hbar\omega\, \hat{\boldsymbol{f}}_\lambda'^\dagger(\boldsymbol{r},\omega) \cdot \hat{\boldsymbol{f}}_\lambda'(\boldsymbol{r},\omega)\,, \tag{2.370}$$

$$\begin{aligned}\hat{H}_\mathrm{AF}' &= -\int \mathrm{d}^3r\, \hat{\boldsymbol{P}}_A' \cdot \hat{\boldsymbol{E}}' - \int \mathrm{d}^3r\, \hat{\boldsymbol{M}}_A' \cdot \hat{\boldsymbol{B}}' + \sum_{\alpha\in A} \frac{1}{2m_\alpha} \left[\int \mathrm{d}^3r\, \hat{\boldsymbol{\Xi}}_\alpha' \times \hat{\boldsymbol{B}}'\right]^2 \\ &\quad - \frac{1}{m_A} \int \mathrm{d}^3r\, \hat{\boldsymbol{P}}_A' \times \hat{\boldsymbol{p}}_A' \cdot \hat{\boldsymbol{B}}'\,. \end{aligned} \tag{2.371}$$

Note that in contrast to the physical magnetisation (2.315), the canonical magnetisation

$$\hat{\boldsymbol{M}}_A'(\boldsymbol{r}) = \sum_{\alpha\in A} \frac{q_\alpha}{2m_\alpha} \int_0^1 \mathrm{d}\sigma\, \sigma \mathcal{S}\big[\hat{\overline{\boldsymbol{r}}}_\alpha' \times \hat{\overline{\boldsymbol{p}}}_\alpha' \delta\big(\boldsymbol{r} - \hat{\boldsymbol{r}}_A' - \sigma\hat{\overline{\boldsymbol{r}}}_\alpha'\big)\big] \tag{2.372}$$

is here defined in terms of the canonically conjugate momenta rather than the velocities, as is required in a Hamiltonian formalism. The Hamiltonian (2.368) is the generalisation of the multipolar Hamiltonian obtained earlier for moving atoms in free space [29, 38, 39] to the case where dispersing and absorbing magnetoelectric bodies are present.

One major advantage of the multipolar coupling scheme is the fact that it allows for a systematic expansion in terms of the electric and magnetic multipole moments of the atom. Thus in the long-wavelength approximation, after retaining only the leading-order termsin the relative coordinates, the interaction Hamiltonian in the multipolar coupling scheme simplifies to

$$\hat{H}'_{\mathrm{AF}} = -\hat{\boldsymbol{d}}' \cdot \hat{\boldsymbol{E}}'(\hat{\boldsymbol{r}}'_A) - \hat{\boldsymbol{m}}' \cdot \hat{\boldsymbol{B}}'(\hat{\boldsymbol{r}}'_A) + \sum_{\alpha \in A} \frac{q_\alpha^2}{8m_\alpha} \big[\hat{\bar{\boldsymbol{r}}}'_\alpha \times \hat{\boldsymbol{B}}'(\hat{\boldsymbol{r}}'_A)\big]^2 + \frac{3}{8m_A} \big[\hat{\boldsymbol{d}}' \times \hat{\boldsymbol{B}}'(\hat{\boldsymbol{r}}'_A)\big]^2 - \frac{1}{m_A} \hat{\boldsymbol{d}}' \times \hat{\boldsymbol{p}}'_A \cdot \hat{\boldsymbol{B}}'(\hat{\boldsymbol{r}}'_A) \tag{2.373}$$

where the canonical magnetic dipole moment

$$\hat{\boldsymbol{m}}' = \sum_{\alpha \in A} \frac{q_\alpha}{2m_\alpha} \hat{\bar{\boldsymbol{r}}}'_\alpha \times \hat{\bar{\boldsymbol{p}}}'_\alpha \tag{2.374}$$

is again different from the physical magnetic dipole moment (2.321). The first two terms $\hat{H}'_{\mathrm{AF}}$ represent electric and magnetic dipole interactions, respectively; the next two terms describe the diamagnetic interaction; and the term on the second line is the Röntgen interaction due to the centre-of-mass motion. In the multipolar coupling scheme, the long-wavelength approximation is obviously not an electric-dipole approximation, because magnetic interactions are still explicitly present. The long-wavelength approximation thus has different meanings in the two schemes, because the Taylor expansion in terms of relative particle coordinates does not commute with the Power–Zienau–Woolley transformation. For non-magnetic atoms, the interaction Hamiltonian reduces to its electric-dipole form

$$\hat{H}'_{\mathrm{AF}} = -\hat{\boldsymbol{d}}' \cdot \hat{\boldsymbol{E}}'(\hat{\boldsymbol{r}}'_A) - \frac{1}{m_A} \hat{\boldsymbol{d}}' \times \hat{\boldsymbol{p}}'_A \cdot \hat{\boldsymbol{B}}'(\hat{\boldsymbol{r}}'_A)\,. \tag{2.375}$$

At the end of this section, let us compare the minimal and multipolar coupling schemes. As the total Hamiltonian is the same in both formalisms, the eigenenergies of the total system and the equations of motion for the physical variables are the same in both schemes. However, the separation of the total Hamiltonian into a field part, an atomic part and an interaction part is different in the two schemes, as can be seen by comparing (2.326) and (2.368). Hence, the ground state $|\{0'\}\rangle$ of $\hat{H}'_{\mathrm{F}}$,

$$\hat{\boldsymbol{f}}'_\lambda(\boldsymbol{r},\omega)|\{0'\}\rangle = \boldsymbol{0} \quad \forall\, \lambda, \boldsymbol{r}, \omega\,, \tag{2.376}$$

is different from that of $\hat{H}_{\mathrm{F}}$; and similarly the eigenstates $|n'\rangle$ of $\hat{H}'_A$ are different from the eigenstates $|n\rangle$ of $\hat{H}_A$ (and these uncoupled eigenstates are not simply related to each other via the Power–Zienau–Woolley transformation). When accounting for the atom–field interaction only in a perturbative way, two different approximations to the same exact eigenenergy of the coupled system may thus occur in general. Depending on the specific perturbative calculation, one of the two schemes may yield a better approximation. For instance, it was found that the multipolar scheme leads to a more realistic result for the shape of resonance lines of the H atom [40].

The second main difference between the two formalisms is the different relation of the canonically conjugate momenta to the physical variables. In the minimal coupling scheme, physical and canonical particle momenta differ by the term $q_\alpha \hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)$ [recall (2.335)], whereas in the multipolar formalism we have

$$m_\alpha \dot{\hat{\boldsymbol{r}}}_\alpha = m_\alpha \dot{\hat{\boldsymbol{r}}}'_\alpha = \frac{1}{\mathrm{i}\hbar}\big[m_\alpha \hat{\boldsymbol{r}}'_\alpha, \hat{H}\big] = \hat{\boldsymbol{p}}'_\alpha + \int \mathrm{d}^3 r\, \hat{\boldsymbol{\Xi}}_\alpha \times \hat{\boldsymbol{B}}\,, \tag{2.377}$$

as follows from the Hamiltonian (2.368) together with the canonical commutation relations for $\hat{\boldsymbol{r}}'_\alpha$ and $\hat{\boldsymbol{p}}'_\alpha$. In a certain sense, the canonical particle momenta in the multipolar coupling scheme are thus more intuitive, since they coincide with the physical momenta in the long-wavelength approximation,

$$\hat{\boldsymbol{p}}'_\alpha = m_\alpha \dot{\hat{\boldsymbol{r}}}_\alpha\,. \tag{2.378}$$

The situation is reversed for the field momenta: The canonical momentum in the minimal coupling scheme is directly proportional to the transverse part of the physical electric field,

$$\hat{\boldsymbol{\Pi}} = -\varepsilon_0 \hat{\boldsymbol{\mathcal{E}}}^\perp \tag{2.379}$$

while in the multipolar coupling scheme, canonical field momentum differs from the transverse part of the physical electric field according to

$$\hat{\boldsymbol{\Pi}}' = -\varepsilon_0 \hat{\boldsymbol{\mathcal{E}}}^\perp - \hat{\boldsymbol{P}}_A^\perp\,. \tag{2.380}$$

2.4.3 Multiple Atoms

Our treatment of the interaction of the electromagnetic field with a single atom as described in the previous sections can easily be extended to more than one atom. In the case of multiple atoms, the charged particles α may be grouped into several atoms A, so in straightforward generalisation of (2.326), the total Hamiltonian of the system in the minimal coupling scheme reads [12, 41]

$$\begin{aligned}\hat{H} &= \sum_A \sum_{\alpha\in A} \frac{\big[\hat{\boldsymbol{p}}_\alpha - q_\alpha \hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha)\big]^2}{2m_\alpha} + \sum_{A,B} \sum_{\substack{\alpha\in A,\beta\in B\\ \alpha\neq\beta}} \frac{q_\alpha q_\beta}{8\pi\varepsilon_0 |\hat{\boldsymbol{r}}_\alpha - \hat{\boldsymbol{r}}_\beta|} \\ &\quad + \sum_{\lambda=e,m} \int \mathrm{d}^3 r \int_0^\infty \mathrm{d}\omega\, \hbar\omega\, \hat{\boldsymbol{f}}^\dagger_\lambda(\boldsymbol{r},\omega)\cdot \hat{\boldsymbol{f}}_\lambda(\boldsymbol{r},\omega) + \sum_A \sum_{\alpha\in A} q_\alpha \hat{\phi}(\hat{\boldsymbol{r}}_\alpha) \\ &= \sum_A \hat{H}_A + \sum_{A<B} \hat{H}_{AB} + \hat{H}_\mathrm{F} + \sum_A \hat{H}_{A\mathrm{F}}\,. \end{aligned} \tag{2.381}$$

Here, the individual atomic Hamiltonians $\hat{H}_A$ are given by (2.313), where one has to bare in mind that their eigenenergies $E_n \to E_n^A$ and eigenstates $|n\rangle \to |n_A\rangle$ may be different for different atomic species. The atom–field coupling Hamiltonians $\hat{H}_{AF}$ are given by (2.327). In long-wavelength approximation, they reduce to (2.352) where the atomic dipole moment $\hat{\boldsymbol{d}} \to \hat{\boldsymbol{d}}_A$ is also species-dependent. In addition to these trivial generalisations, an atom–atom interaction term has appeared which is due to the Coulomb interaction of particles belonging to different atoms:

$$\hat{H}_{AB} = \sum_{\alpha\in A,\beta\in B} \frac{q_\alpha q_\beta}{4\pi\varepsilon_0|\hat{\boldsymbol{r}}_\alpha - \hat{\boldsymbol{r}}_\beta|}\,. \tag{2.382}$$

In long-wavelength approximation, it reduces to the dipole–dipole interaction

$$\hat{H}_{AB} = \frac{\hat{\boldsymbol{d}}_A \cdot \hat{\boldsymbol{d}}_B - 3(\hat{\boldsymbol{d}}_A \cdot \hat{\boldsymbol{e}}_{AB})(\hat{\boldsymbol{e}}_{AB} \cdot \hat{\boldsymbol{d}}_B)}{4\pi\varepsilon_0 \hat{r}_{AB}^3} \tag{2.383}$$

$[\hat{r}_{AB} = |\hat{\boldsymbol{r}}_A - \hat{\boldsymbol{r}}_B|, \hat{\boldsymbol{e}}_{AB} = (\hat{\boldsymbol{r}}_A - \hat{\boldsymbol{r}}_B)/\hat{r}_{AB}]$.

The multi-atom Hamiltonian is completely analogous to the single-atom one with the charged particles just being grouped in a different way. It is therefore evident that the Newton equation (2.334) for the particle motion and the homogeneous Maxwell equations (2.330) and (2.331) remain valid, while the source terms of the inhomogeneous Maxwell equations are now given by the total charge and current densities of all atoms

$$\nabla \cdot \hat{\boldsymbol{\mathcal{D}}} = \sum_A \hat{\rho}_A\,, \tag{2.384}$$

$$\nabla \times \hat{\boldsymbol{\mathcal{H}}} - \dot{\hat{\boldsymbol{\mathcal{D}}}} = \sum_A \hat{\boldsymbol{j}}_A\,. \tag{2.385}$$

The multipolar coupling scheme can be obtained by applying the generalised Power–Zienau–Woolley transformation

$$\hat{f}' = \hat{U}\hat{f}\hat{U}^\dagger \quad \text{with } \hat{U} = \exp\left[\frac{\mathrm{i}}{\hbar}\int \mathrm{d}^3r \sum_A \hat{\boldsymbol{P}}_A \cdot \hat{\boldsymbol{A}}\right]. \tag{2.386}$$

Again, the transition from the single to the multi-atom case can be made by using our result (2.368) for the transformed Hamiltonian of the field interacting with one atom, while regrouping the particles into several atoms. Assuming that the atoms are well separated from one another, so that their electronic wave functions do not overlap, we have

$$\int \mathrm{d}^3r\, \hat{\boldsymbol{P}}_A' \cdot \hat{\boldsymbol{P}}_B' = 0 \quad \text{for } A \neq B\,. \tag{2.387}$$

The total Hamiltonian in the multipolar coupling scheme then reads [12, 41]

$$
\begin{aligned}
\hat{H} = & \sum_{\lambda=e,m} \int \mathrm{d}^3 r \int_0^\infty \mathrm{d}\omega\, \hbar\omega\, \hat{\boldsymbol{f}}_\lambda'^\dagger(\boldsymbol{r},\omega) \cdot \hat{\boldsymbol{f}}_\lambda'(\boldsymbol{r},\omega) \\
& + \sum_A \sum_{\alpha \in A} \frac{1}{2m_\alpha} \mathcal{S} \left[\hat{\boldsymbol{p}}_\alpha' + \int \mathrm{d}^3 r\, \hat{\boldsymbol{\Xi}}_\alpha' \times \hat{\boldsymbol{B}}' \right]^2 \\
& + \frac{1}{2\varepsilon_0} \int \mathrm{d}^3 r \sum_A \hat{\boldsymbol{P}}_A'^2 - \int \mathrm{d}^3 r \sum_A \hat{\boldsymbol{P}}_A' \cdot \hat{\boldsymbol{E}}' \\
= & \sum_A \hat{H}_A' + \hat{H}_\mathrm{F}' + \sum_A \hat{H}_{A\mathrm{F}}'
\end{aligned}
\tag{2.388}
$$

where in contrast to the minimal coupling scheme, no atom–atom interaction term is present. Again, the individual atom and atom–field Hamiltonians (2.369) and (2.371) remain valid, and so do the long-wavelength and electric dipole forms (2.373) and (2.375) of the latter.

2.4.4 Magnetic Atoms

In order to obtain an accurate description of magnetic atoms, we have to slightly generalise our theory by taking into account the particle spins. In analogy to the orbital angular momentum $\hat{\bar{\boldsymbol{r}}}_\alpha \times \hat{\bar{\boldsymbol{p}}}_\alpha$ of a particle α, its spin $\hat{\boldsymbol{s}}_\alpha$ satisfies the angular momentum commutation relations

$$
\left[\hat{\boldsymbol{s}}_\alpha, \hat{\boldsymbol{s}}_\beta\right] = -\mathrm{i}\hbar\delta_{\alpha\beta}\boldsymbol{I} \times \hat{\boldsymbol{s}}_\alpha\,. \tag{2.389}
$$

The particle spin induces a magnetic dipole moment

$$
\hat{\boldsymbol{m}}_\alpha = \gamma_\alpha \hat{\boldsymbol{s}}_\alpha\,, \tag{2.390}
$$

where the proportionality constant γ_α is called the gyromagnetic ratio. It can be given as a multiple of an elementary unit, the magneton μ_α, according to $\gamma_\alpha = \mathrm{sign}(q_\alpha) g_\alpha \mu_\alpha/\hbar$, with the dimensionless constant g_α being the (spin) g-factor. For electrons, the relevant magneton is the Bohr magneton $\mu_\mathrm{B} = e\hbar/(2m_e)$ and the g-factor is approximately given by $g_e \approx 2$, leading to a gyromagnetic ratio $\gamma_\alpha \approx -e/m_e$. The electron spin and the induced magnetic dipole moment are hence antiparallel to each other. The gyromagnetic ratio of nuclei is commonly given in terms of the nuclear magneton $\mu_p = e\hbar/(2m_p)$ (m_p: proton mass), so that $\gamma_\alpha = g_\alpha e/(2m_p)$; the spin and its induced magnetic dipole moment for a nucleus are parallel. Note that the g-factors and gyromagnetic ratios can strongly differ for different nuclei.

The magnetic dipole moments associated with the particles' spins present an additional contribution to the atomic current density and magnetisation which modify to

$$\hat{\boldsymbol{j}}_A(\boldsymbol{r}) = \sum_{\alpha\in A} q_\alpha \mathcal{S}\left[\dot{\hat{\boldsymbol{r}}}_\alpha \delta(\boldsymbol{r}-\hat{\boldsymbol{r}}_\alpha)\right] - \sum_{\alpha\in A} \hat{\boldsymbol{m}}_\alpha \times \nabla\delta(\boldsymbol{r}-\hat{\boldsymbol{r}}_\alpha)\,, \tag{2.391}$$

$$\hat{\boldsymbol{M}}_A(\boldsymbol{r}) = \sum_{\alpha\in A} \frac{q_\alpha}{2} \int_0^1 \mathrm{d}\sigma\, \sigma \mathcal{S}\left[\hat{\bar{\boldsymbol{r}}}_\alpha \times \dot{\hat{\bar{\boldsymbol{r}}}}_\alpha \delta\big(\boldsymbol{r}-\hat{\boldsymbol{r}}_A-\sigma\hat{\bar{\boldsymbol{r}}}_\alpha\big)\right] + \sum_{\alpha\in A} \hat{\boldsymbol{m}}_\alpha \delta(\boldsymbol{r}-\hat{\boldsymbol{r}}_\alpha)\,. \tag{2.392}$$

Note that the relation (2.317) between atomic current density, magnetisation and polarisation remains valid. The magnetic dipole moment of the atom also acquires a contribution from the particle spin, so that it now reads

$$\hat{\boldsymbol{m}}_A = \sum_{\alpha\in A} \left(\frac{q_\alpha}{2} \hat{\bar{\boldsymbol{r}}}_\alpha \times \dot{\hat{\bar{\boldsymbol{r}}}}_\alpha + \gamma_\alpha \hat{\boldsymbol{s}}_\alpha\right)\,. \tag{2.393}$$

One can also assign a gyromagnetic ratio to the orbital angular momentum contribution to the magnetic moment, the first term in the above equation; it obviously reads $-e/(2m_e)$ for electrons, with the associated g-factor being equal to unity. Finally, one may of course define a gyromagnetic ratio for the total angular momentum of each particle, with the associated g-factor being known as the Landé g-factor.

The coupling of the particle spin with the electromagnetic field can be implemented by means of a Pauli interaction term, which tends to align the spins with the magnetic field. The atom–field interaction Hamiltonian for magnetic atoms in the minimal coupling scheme thus reads [12, 42]

$$\begin{aligned}\hat{H}_{\mathrm{AF}} = {} & \sum_{\alpha\in A} q_\alpha \hat{\phi}(\hat{\boldsymbol{r}}_\alpha) - \sum_{\alpha\in A} \frac{q_\alpha}{m_\alpha} \hat{\boldsymbol{p}}_\alpha \cdot \hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_\alpha) + \sum_{\alpha\in A} \frac{q_\alpha^2}{2m_\alpha} \hat{\boldsymbol{A}}^2(\hat{\boldsymbol{r}}_\alpha) \\ & - \sum_{\alpha\in A} \gamma_\alpha \hat{\boldsymbol{s}}_\alpha \cdot \hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}_\alpha)\,,\end{aligned} \tag{2.394}$$

it reduces to

$$\begin{aligned}\hat{H}_{\mathrm{AF}} = {} & -\hat{\boldsymbol{d}}_A \cdot \hat{\boldsymbol{E}}^{\parallel}(\hat{\boldsymbol{r}}_A) - \sum_{\alpha\in A} \frac{q_\alpha}{m_\alpha} \hat{\bar{\boldsymbol{p}}}_\alpha \cdot \hat{\boldsymbol{A}}(\hat{\boldsymbol{r}}_A) + \sum_{\alpha\in A} \frac{q_\alpha^2}{2m_\alpha} \hat{\boldsymbol{A}}^2(\hat{\boldsymbol{r}}_A) \\ & - \sum_{\alpha\in A} \gamma_\alpha \hat{\boldsymbol{s}}_\alpha \cdot \hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}_A)\end{aligned} \tag{2.395}$$

in the long-wavelength approximation.

One can easily see that the Maxwell equations remain valid in their usual form: Equations (2.330) and (2.331) or (2.384) obviously still hold, since the occurring quantities are not affected by the particle spin. Noting that the Pauli interaction term commutes with $\hat{\boldsymbol{B}} = \hat{\boldsymbol{\mathcal{B}}}$, the same is true for (2.332). The commutator (A.20) of $\hat{\boldsymbol{D}}$ with $\hat{\boldsymbol{A}}$ being non-trivial, the Pauli interaction does give rise to an additional contribution to $\dot{\hat{\boldsymbol{\mathcal{D}}}}$:

$$\frac{1}{\mathrm{i}\hbar}\left[\hat{\boldsymbol{\mathcal{D}}}(\boldsymbol{r}), -\sum_{\alpha\in A}\gamma_\alpha\hat{\boldsymbol{s}}_\alpha\cdot\hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}_\alpha)\right] = \frac{1}{\mathrm{i}\hbar}\left[\hat{\boldsymbol{D}}(\boldsymbol{r}), -\sum_{\alpha\in A}\gamma_\alpha\hat{\boldsymbol{s}}_\alpha\cdot\hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}_\alpha)\right]$$
$$= \sum_{\alpha\in A}\gamma_\alpha\hat{\boldsymbol{s}}_\alpha\times\nabla\delta(\boldsymbol{r}-\hat{\boldsymbol{r}}_\alpha)\,. \tag{2.396}$$

As this additional term is exactly cancelled by the spin-dependent contribution to the current density, the Ampère law (2.333) or (2.385) also remains valid, with $\hat{\boldsymbol{j}}_A$ being given by (2.391) for a magnetic atom. The Newton equations for the particles are modified due to the action of the magnetic field on the particle spins. Using the canonical commutation relations for $\hat{\boldsymbol{r}}_\alpha$ and $\hat{\boldsymbol{p}}_\alpha$, one finds that the Pauli interaction gives rise to an additional term

$$\frac{1}{\mathrm{i}\hbar}\left[m_\alpha\dot{\hat{\boldsymbol{r}}}_\alpha, -\sum_{\alpha\in A}\gamma_\alpha\hat{\boldsymbol{s}}_\alpha\cdot\hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}_\alpha)\right] = \frac{1}{\mathrm{i}\hbar}\left[\hat{\boldsymbol{p}}_\alpha, -\sum_{\alpha\in A}\gamma_\alpha\hat{\boldsymbol{s}}_\alpha\cdot\hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}_\alpha)\right]$$
$$= \gamma_\alpha\nabla_\alpha\big[\hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}_\alpha)\cdot\hat{\boldsymbol{s}}_\alpha\big]\,, \tag{2.397}$$

so that the Newton equations are modified to

$$m_\alpha\ddot{\hat{\boldsymbol{r}}}_\alpha = q_\alpha\hat{\boldsymbol{\mathcal{E}}}(\hat{\boldsymbol{r}}_\alpha) + q_\alpha\mathcal{S}\Big[\dot{\hat{\boldsymbol{r}}}_\alpha\times\hat{\boldsymbol{\mathcal{B}}}(\hat{\boldsymbol{r}}_\alpha)\Big] + \nabla_\alpha\big[\hat{\boldsymbol{m}}_\alpha\cdot\hat{\boldsymbol{\mathcal{B}}}(\hat{\boldsymbol{r}}_\alpha)\big]\,, \tag{2.398}$$

with the last term being the Zeeman force.

When applying the Power–Zienau–Woolley transformation to the minimal coupling Hamiltonian of a magnetic atom, one notes that $\hat{\boldsymbol{B}}' = \hat{\boldsymbol{B}}$ and

$$\hat{\boldsymbol{s}}'_\alpha = \hat{\boldsymbol{s}}_\alpha\,, \tag{2.399}$$

so the Pauli interaction term is invariant:

$$-\sum_{\alpha\in A}\gamma_\alpha\hat{\boldsymbol{s}}_\alpha\cdot\hat{\boldsymbol{B}}(\hat{\boldsymbol{r}}_\alpha) = -\sum_{\alpha\in A}\gamma_\alpha\hat{\boldsymbol{s}}'_\alpha\cdot\hat{\boldsymbol{B}}'(\hat{\boldsymbol{r}}'_\alpha)\,. \tag{2.400}$$

Its contribution to $\hat{H}'_{\mathrm{AF}}$ can be included by redefining the canonical magnetisation (2.372) according to [12, 42]

$$\hat{\boldsymbol{M}}'_A(\boldsymbol{r}) = \sum_{\alpha\in A} \frac{q_\alpha}{2m_\alpha} \int_0^1 \mathrm{d}\sigma\, \sigma \mathcal{S}\big[\hat{\bar{\boldsymbol{r}}}'_\alpha \times \hat{\bar{\boldsymbol{p}}}'_\alpha \delta\big(\boldsymbol{r} - \hat{\boldsymbol{r}}'_A - \sigma\hat{\bar{\boldsymbol{r}}}'_\alpha\big)\big] + \sum_{\alpha\in A} \gamma_\alpha \hat{\boldsymbol{s}}'_\alpha \delta(\boldsymbol{r} - \hat{\boldsymbol{r}}'_\alpha)\,, \tag{2.401}$$

so that the multipolar interaction Hamiltonian (2.371) remains valid. Accordingly, upon redefining the canonical magnetic dipole moment as

$$\hat{\boldsymbol{m}}' = \sum_{\alpha\in A} \left(\frac{q_\alpha}{2m_\alpha} \hat{\bar{\boldsymbol{r}}}'_\alpha \times \hat{\bar{\boldsymbol{p}}}'_\alpha + \gamma_\alpha \hat{\boldsymbol{s}}'_\alpha \right), \tag{2.402}$$

the multipolar interaction in long-wavelength approximation is still given by (2.375).

References

1. K. Cho, *Reconstruction of Macroscopic Maxwell Equations: A Single Susceptibility Theory*, Springer Tracts in Modern Physics (Springer, Berlin, 2010)
2. H.B. Callen, T.A. Welton, Phys. Rev. **83**(1), 34 (1951)
3. R. Kubo, Rep. Prog. Phys. **29**(I), 255 (1966)
4. J.D. Jackson, *Classical Electrodynamics*, 3rd edn. (Wiley, New York, 1998)
5. L. Knöll, S. Scheel, D.G. Welsch, in *Coherence and Statistics of Photons and Atoms*, ed. by J. Peřina (Wiley, New York, 2001), p. 1
6. D.T. Ho, S.Y. Buhmann, L. Knöll, D.G. Welsch, S. Scheel, J. Kästel, Phys. Rev. A **68**(4), 043816 (2003)
7. C.W. Misner, J.A. Wheeler, Ann. Phys. **2**(6), 525 (1957)
8. M.K. Gaillard, B. Zumino, Nucl. Phys. B **193**(1), 221 (1981)
9. L. Silberstein, Ann. Phys. (Leipzig) **327**(3), 579 (1907)
10. F.H. Hehl, Y.N. Obukhov, *Foundations of Classical Electrodynamics* (Birkhäuser, Boston, 2003)
11. S.Y. Buhmann, D.G. Welsch, Prog. Quantum Electron. **31**(2), 51 (2007)
12. S. Scheel, S.Y. Buhmann, Acta Phys. Slovaca **58**(5), 675 (2008)
13. D.T. Ho, S.Y. Buhmann, L. Knöll, S. Scheel, D.G. Welsch, in *Proceedings of the 8th International Conference on Squeezed States and Uncertainty Relations*, ed. by H. Moya-Cessa, R. Jáuregui, S. Hacyan, O. Castaños (Rinton Press, Princeton, New Jersey, 2003), pp. 110–117
14. C. Klingshirn, in Advances, in *Energy Transfer Processes: Proceedings of the 16th Course of the International School of Atomic and Molecular Spectroscopy*, ed. by B. Di Bartolo, X. Chen (World Scientific Publishing Co. Pte. Ltd., Singapore, 2001), pp. 165–214
15. S.M. Rytov, V.I. Kravtsov, Y.A. Tatarskii, *Principles of Statistical Radiophysics III: Elements of Random Fields* (Springer, Berlin, 1989)
16. E.M. Lifshitz, Sov. Phys. JETP **2**(1), 73 (1956)
17. R. Matloob, R. Loudon, S.M. Barnett, J. Jeffers, Phys. Rev. A **52**(6), 4823 (1995)
18. R. Matloob, Phys. Rev. A **69**(5), 052110 (2004)
19. R. Matloob, Phys. Rev. A **70**(2), 022108 (2004)
20. A. Tip, Phys. Rev. A **57**(6), 4818 (1998)
21. A. Tip, L. Knöll, S. Scheel, D.G. Welsch, Phys. Rev. A **63**(4), 043806 (2001)
22. C. Raabe, S. Scheel, D.G. Welsch, Phys. Rev. A **75**(5), 053813 (2007)
23. B. Huttner, S.M. Barnett, Phys. Rev. A **46**(7), 4306 (1992)
24. M. Wubs, L.G. Suttorp, Phys. Rev. A **63**(4), 043809 (2001)
25. L.G. Suttorp, M. Wubs, Phys. Rev. A **70**(1), 013816 (2004)
26. L.G. Suttorp, A.J. van Wonderen, Europhys. Lett. **67**(5), 766 (2004)

27. O. Di Stefano, S. Savasta, R. Girlanda, J. Opt. B: Quantum Semiclass. Opt. **3**(4), 288 (2001)
28. U. Fano, Phys. Rev. **103**(5), 1202 (1956)
29. C. Baxter, M. Babiker, R. Loudon, Phys. Rev. A **47**(2), 1278 (1993)
30. D.P. Craig, T. Thirunamachandran, *Molecular Quantum Electrodynamics* (Dover, New York, 1998)
31. W. Thomas, Naturwissenschaften **13**(28), 627 (1925)
32. W. Kuhn, Z. Phys. **33**(1), 408 (1925)
33. F. Reiche, W. Thomas, Z. Phys. **34**(1), 408 (1925)
34. S.Y. Buhmann, L. Knöll, D.G. Welsch, D.T. Ho, Phys. Rev. A **70**(5), 052117 (2004)
35. E.A. Power, S. Zienau, Philos. Trans. R. Soc. Lond. Ser. A **251**(999), 427 (1959)
36. R.G. Woolley, Proc. R. Soc. Lond. Ser. A **321**(1547), 557 (1971)
37. C. Cohen-Tannoudji, J. Dupont-Roc, G. Grynberg, *Photons and Atoms* (Wiley, New York, 1989)
38. W.P. Healy, J. Phys. A: Math. Gen. **10**(2), 279 (1977)
39. V.E. Lembessis, M. Babiker, C. Baxter, R. Loudon, Phys. Rev. A **48**(2), 1594 (1993)
40. W.E. Lamb, Phys. Rev. **85**(2), 259 (1952)
41. H. Safari, S.Y. Buhmann, D.G. Welsch, D.T. Ho, Phys. Rev. A **74**(4), 042101 (2006)
42. H. Safari, D.G. Welsch, S.Y. Buhmann, S. Scheel, Phys. Rev. A **78**(6), 062901 (2008)

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