Functional Approach to Electrodynamics in Media

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Abstract

We put forward an approach to classical electrodynamics in media which identifies induced electromagnetic fields as the microscopic counterparts of polarization and magnetization and which systematically employs the mutual functional dependences of induced, external, and total field quantities. This allows for a unified, relativistic description of the electromagnetic response independent of any assumption about the material’s possible composition of electric or magnetic dipoles. Using this approach we derive universal (material-independent) relations between electromagnetic response functions such as the generalized dielectric tensor, generalized magnetic susceptibility and microscopic conductivity tensor. These reduce to well-known identities in special cases, but include more generally the effects of inhomogeneity, non-isotropy and relativistic retardation. We further provide general expressions for the constitutive dyadics of bianistropic media in terms of the nine causal response functions as represented by the conductivity tensor.

Keywords: Electrodynamics, Polarization, Response function, Bianisotropy

1. Introduction

Electrodynamics in media aims at describing the response of a material probe to external electromagnetic perturbations \cite{1, 2, 3, 4}. For this pur-
pose, Maxwell equations alone are not enough, because these relate only the electromagnetic fields to their sources and do not describe the reaction of currents and charges in the material to the electromagnetic fields. The classical approach to electrodynamics in media is therefore to introduce additional field quantities, which are—besides the electric field and magnetic induction \((E, B)\)—the electric polarization and magnetization \((P, M)\) as well as the displacement field and magnetic field \((D, H)\) defined by

\[
D = \varepsilon_0 E + P, \quad H = \frac{1}{\mu_0} B - M, \quad (1)
\]

where \(\varepsilon_0\) and \(\mu_0\) denote the vacuum permittivity and permeability, respectively. The fields \((P, M)\) are interpreted as electric and magnetic dipole densities inside the material. Macroscopic Maxwell equations are then written in the following form:

\[
\begin{align*}
\nabla \cdot D &= \rho_{ext}, \\
\nabla \times H - \frac{\partial D}{\partial t} &= j_{ext}, \\
\nabla \cdot B &= 0, \\
\nabla \times E + \frac{\partial B}{\partial t} &= 0,
\end{align*} \quad (2)
\]

where \((\rho_{ext}, j_{ext})\) represent the external charge and current sources \([1]\). (Often these are also identified with the free charges as opposed to those bound in the medium \([3, 4]\).) These general equations are complemented by material-specific constitutive relations, which in the simplest case read

\[
P = \varepsilon_0 \chi_e E, \quad M = \chi_m H, \quad (4)
\]

with electric and magnetic susceptibilities \(\chi_e\) and \(\chi_m\). Together these equations determine (up to the freedom to choose appropriate boundary conditions) electric and magnetic fields inside the material medium in terms of the external sources. They are usually supposed to hold on a macroscopic scale, i.e., \((E, B)\) represent suitable averages of the microscopic fields, and \((P, M)\) are regarded as average dipole moments of macroscopic volume elements \([1, 2, 3, 4, 5]\). We will henceforth refer to this approach as the classical approach to electrodynamics in media. Traditional textbooks (such as \([3, 4]\)) set up this approach and derive Eqs. \((2) - (3)\) based on the assumption of microscopic dipoles induced by the electromagnetic fields, as e.g. in the Clausius-Mossotti model \([6]\). Such simplified models have been ubiquitous in the description of the electromagnetic response of solids for more than a century since Maxwell’s original work in 1865 \([7, 8]\).
However, two modern developments in solid-state physics have put severe limitations on the classical approach: the quest for an \textit{ab initio} description of materials properties \cite{9, 10, 11, 12} on the one hand, and the discovery of new materials with exotic electromagnetic responses on the other hand. As for the first point, the Modern Theory of Polarization \cite{13, 14, 15} has raised conceptual questions on the interpretation of $P$ and $M$ in terms of electric and magnetic dipole densities. It has been shown that the localized polarizable units of the Clausius-Mossotti model are in stark contrast to the actual delocalized electronic charge distributions in real materials, and hence this model fails in most cases to describe polarization effects adequately \cite{14}. More fundamentally it was argued that the polarization of a crystalline solid cannot, even in principle, be defined as a bulk quantity in terms of periodic electronic charge distributions, and instead the polarization \textit{change} in a typical measurement setup was defined in terms of a macroscopic charge flow in the interior of the sample \cite{13, 14}. Regarding the second point, the advent of metamaterials \cite{16, 17, 18}, in particular bianisotropic media with magnetoelectric coupling \cite{19, 20, 21}, demonstrated the need to generalize the constitutive relations \cite{4} and to reformulate the theory of electromagnetic responses in a more systematic way \cite{22, 23}. Since often these materials properties are determined by nanoscale resonant inclusions ("meta-atoms") \cite{24}, macroscopic averaging procedures have also come under intense investigation \cite{25, 26, 27}. Further systems showing large magnetoelectric effects are multiferroics, which combine multiple long-range orderings in a single phase \cite{28, 29}, as well as composites based on magnetostrictive and piezoelectric materials \cite{30, 31, 32}. \cite{1}

In this paper, we will develop a \textit{functional approach} to electrodynamics in media, which identifies induced electromagnetic fields as the microscopic counterparts of macroscopic polarizations, and these are interpreted as functionals of the external perturbations. Thus we start from the following iden-

\begin{footnote}
\textsuperscript{1}The research on magnetoelectric coupling has indeed a much longer history going back to W.C. Röntgen and P. Curie in the 19th century \cite{32}. In traditional single-phase materials such as Cr$_2$O$_3$, however, the effect is generally too weak for practical applications \cite{33}.
\end{footnote}
ifications on a microscopic scale:

\[
P(x, t) = -\varepsilon_0 E_{\text{ind}}(x, t), \quad M(x, t) = B_{\text{ind}}(x, t)/\mu_0, \quad (5)
\]

\[
D(x, t) = \varepsilon_0 E_{\text{ext}}(x, t), \quad H(x, t) = B_{\text{ext}}(x, t)/\mu_0, \quad (6)
\]

\[
E(x, t) = E_{\text{tot}}(x, t), \quad B(x, t) = B_{\text{tot}}(x, t). \quad (7)
\]

The quantities on the right hand side refer to induced, external and total microscopic fields, respectively, which will be characterized precisely below (see also \[34, 1\]). By microscopic fields we mean that these are derived from microscopic charge and current distributions, which in turn are derived from continuous quantum many-body wave functions. The macroscopic electric/magnetic dipole moments of a distribution of charges and currents inside volume \(V\) are consequently defined as

\[
P(t) = -\frac{\varepsilon_0}{V} \int d^3 x E_{\text{ind}}(x, t), \quad M(t) = \frac{1}{\mu_0 V} \int d^3 x B_{\text{ind}}(x, t), \quad (8)
\]

where the integration range is the whole three-dimensional space. We will show below that both the classical approach to electrodynamics in media and the fundamental equation of the Modern Theory of Polarization are naturally recovered from these definitions.

Microscopic approaches to electrodynamics in media have already been described by Hirst \[35\] and in the metric-free formulation by Truesdell and Toupin \[36\] as well as Hehl and Obukhov \[37, 38\] (cf. also the textbook of Kovetz \[39\]). Especially, the metric-free approach takes the Maxwell equations (2)–(3) as microscopic equations and has the advantage of being independent of the spacetime geometry, fact which is also relevant to the experiment \[40\]. By comparison, our approach does not require any assumption about the localization of charges and currents inside the material medium, it provides a unified description of electromagnetic fields and polarizations and does not depend on the constitutive relations of the medium. Therefore it can be used to derive universal (material-independent) relations between electromagnetic response functions, which are the main result of this article (see Section 3). We further note that in the textbook of Fließbach \[1\], Eqs. (5)–(7) are deduced for the macroscopically averaged fields in the special case of homogeneous, isotropic media, while they represent the most general definition of microscopic polarizations in our approach (see the discussion in Section 2.3).
Our main focus is on the microscopic electromagnetic response functions which are represented by partial functional derivatives

$$\frac{\delta E^i_{\text{ind}}(x, t)}{\delta E^j_{\text{ext}}(x', t')}, \frac{\delta E^i_{\text{ind}}(x, t)}{\delta B^j_{\text{ext}}(x', t')}, \frac{\delta B^i_{\text{ind}}(x, t)}{\delta E^j_{\text{ext}}(x', t')}, \frac{\delta B^i_{\text{ind}}(x, t)}{\delta B^j_{\text{ext}}(x', t')}.$$ (9)

These are not independent of each other, and hence it is possible to formulate electrodynamics in media as a single susceptibility theory \[23\] where all linear responses are derived from a single susceptibility tensor. In magneto-optics, e.g., it has been suggested to introduce an effective permittivity tensor relating the $D$ and $E$ fields while setting $H = B$ identically \[41, 42, 43\]. By contrast, we keep here the usual definition of electromagnetic response functions \(9\) and relate each of them to the microscopic conductivity tensor, which also describes the most general linear electromagnetic response \[34, 44, 45, 46\]. Starting from the model-independent definitions \(5\)–\(7\), we will thus obtain universal relations, which can be used to study the electromagnetic response not only of solid crystals, but also liquids, single atoms or molecules, or even the vacuum of quantum electrodynamics (see Section 2.2).

The paper is organized as follows: In Section 2 we review the general electromagnetic response formalism based on the functional dependence of the induced four-current on the external four-potential. We propose equivalent response functionals and thereby establish the connection to the Schwinger-Dyson equations in quantum electrodynamics and Hedin’s equations in electronic structure theory. Further we relate the functional approach to the classical approach and the Modern Theory of Polarization. In Section 3 we derive universal relations between the generalized dielectric tensor, generalized magnetic susceptibility and the microscopic conductivity tensor. These reduce to well-known identities in special cases, but apply more generally to inhomogeneous, nonisotropic materials and take into account relativistic retardation effects. We further provide explicit expressions for the cross-coupling coefficients between electric and magnetic fields, and summarize our results in Section 3.5 by a set of four universal relations between electromagnetic response functions. Finally we relate the constitutive dyadics used in the context of bianisotropic materials to the nine causal response functions as represented by the conductivity tensor.
2. Functional setup of electrodynamics in media

2.1. Fundamental response functions

In a general experimental setup, the electromagnetic response of a material probe is determined under an externally applied electromagnetic perturbation. For the whole system comprising the probe and the external perturbation, this means that all electromagnetic quantities (such as fields, charges and currents) are split into internal and external contributions. Thinking of the external perturbation as inducing a redistribution of charges and currents in the sample, a natural starting point for the theoretical description is the functional (see e.g. \[47\])

$$j_{\text{int}}^\mu = j_{\text{int}}^\mu[A_{\text{ext}}^\nu],$$

(10)

where $j_{\text{int}}^\mu$ is the four-current of the internal charges and currents, and $A_{\text{ext}}^\nu$ is the four-potential of the external electromagnetic fields. In general, this functional dependence may be complicated, depend on past history (hysteresis), be nonlinear, etc. [3]. Here we only assume that it is analytic, and hence up to first order the Taylor series expansion reads

$$j_{\text{int}}^\mu(x) = j_{\text{int},0}^\mu(x) + \int d^4x' \frac{\delta j_{\text{int}}^\mu(x)}{\delta A_{\text{ext}}^\nu(x')} (A_{\text{ext}}^\nu(x') - A_{\text{ext},0}^\nu(x')),$$

(11)

where $x = (x, t)$ and $d^4x = d^3x dt$, and the functional derivative is to be evaluated at the initial potential $A_{\text{ext},0}^\nu$. The difference between internal currents in the presence and in the absence of external sources is called \textit{induced four-current}, $j_{\text{ind}}^\mu = j_{\text{int}}^\mu - j_{\text{int},0}^\mu$. The above expansion is typically performed around $A_{\text{ext},0}^\nu \equiv 0$, but it is also possible to consider small perturbations around a finite initial potential (in this way one can describe, e.g., the current response to an applied voltage in the presence of a constant external magnetic field, i.e. the Hall conductivity). From the functional point of view, linear response theory restricts attention to the first order terms, i.e. calculation of the functional derivatives

$$\chi_{\mu\nu}(x, x') = \frac{\delta j_{\text{ind}}^\mu(x)}{\delta A_{\text{ext}}^\nu(x')},$$

(12)

We will refer to this tensor as the \textit{fundamental response tensor}, and to its components as \textit{fundamental response functions}. We do not presuppose that the system under consideration actually behaves linearly, but instead we
will make generally valid statements on the first order derivatives of induced quantities with respect to external perturbations.

The following constraints on \( \chi_{\mu \nu} \) are required by the continuity equation and respectively the gauge invariance of the induced current [47, 48]:

\[
\partial \mu \chi_{\mu \nu}(x, x') = 0, \quad \text{and} \quad \partial^\nu \chi_{\mu \nu}(x, x') = 0. \tag{13}
\]

Therefore, at most 9 of the 16 fundamental response functions are independent of each other (cf. [22, 23, 49]). Apart from reducing the number of independent response functions, focussing on the fundamental response functions in the first place has several advantages: They constitute a second-rank Lorentz tensor and thus yield a relativistic description of the electromagnetic response, which is especially relevant for the study of moving media [50, 51, 52]. Their transformation behavior reads explicitly

\[
\chi_{\mu \nu}(x, x') = \Lambda_{\alpha \mu} \Lambda_{\beta \nu} \chi_{\alpha \beta}(\Lambda^{-1} x, \Lambda^{-1} x') \tag{14}
\]

with Lorentz transformations \( \Lambda \in O(3,1) \). Moreover, the coupling of \( A^\mu \) to a Hamiltonian can easily be written down on the most fundamental level: it is given by the minimal coupling prescription \( p^\mu \rightarrow p^\mu - eA^\mu \). A quantum field theoretical expression for the fundamental response functions is therefore given by the Kubo formula in terms of the current-current correlation function [47]. However, the crucial question is whether one can derive closed expressions for the usual electromagnetic response functions (such as conductivity, dielectric tensor and magnetic susceptibility) in terms of the fundamental response tensor. This question will be addressed in Section 3.

2.2. Equivalent formulations and connection to Hedin’s equations

As described in the previous section, the functional approach to electrodynamics in media is based on the splitting of all electromagnetic quantities into external and internal (or induced) quantities. Each of these subsystems can be described by its charges and currents \( j^\mu \), by the gauge potential \( A^\mu \) or by the electromagnetic fields \( (E, B) \). These descriptions are equivalent in the following sense: (i) Given the gauge potential, the electromagnetic fields are determined by \( E = -\nabla \Phi - \partial_t A \) and \( B = \nabla \times A \). (ii) Given the electromagnetic fields, the currents are determined by the inhomogeneous Maxwell equations. (iii) Given the currents, the gauge potential is determined by \( A^\mu = (D_0)^\mu_{\nu} j^\nu + \partial^\mu f \) with a free, retarded propagator \( D_0 \) and an arbitrary pure gauge \( \partial^\mu f \). As we deal with electromagnetic perturbations of a sample
Figure 1: Definition of external, internal, induced and total fields (cf. [34, 1]). (a) \((E_{\text{int,0}}, B_{\text{int,0}})\) are the fields present in the sample without external sources. (b) The external fields are produced by the external sources in the absence of the medium. (c) In the presence of external sources, the fields inside the medium are a superposition of external and internal fields. (d) Subtracting from the latter the fields \((E_{\text{int,0}}, B_{\text{int,0}})\) yields the induced fields, whereas the total fields are defined as external plus induced fields.

which are produced in the laboratory, for both the external perturbation and the reaction of the sample it is sensible to assume that the electromagnetic fields vanish before some initial switching-on. Mathematically this requirement leads to the choice of a retarded propagator \(D_0\).

In particular, the induced electromagnetic fields are related by inhomogeneous and homogeneous Maxwell equations to the induced charges and currents, which were defined in the previous section. Schematically, the external, internal, induced and total fields are shown in Fig. 1 (they agree with the definitions in [34, 1]). Now given the functional dependence of the induced current on the external four potential, it follows that all other electromagnetic response functions (specifying e.g. the induced current in terms of the external electric field or the external charges and currents) can be calculated from it. Especially, we will consider in Section 3 the functionals \(E_{\text{ind}}[E_{\text{ext}}, B_{\text{ext}}]\) and \(B_{\text{ind}}[E_{\text{ext}}, B_{\text{ext}}]\), and relate their partial functional derivatives to the fundamental response tensor. Moreover, it follows that Eq. (12) defines only one of a number of equivalent response tensors. Instead
of describing the response of the system in terms of the induced quantities, one may as well characterize it by the functional dependence of the total on the external, or even the induced on the total field quantities. Each of these possibilities gives rise to an equivalent set of response functions, all of which are—within the linear response regime—mutually related by functional chain rules.

As an example, we consider the converse point of view to Eq. (12), namely the dependence of the total four-potential on the external current density. Defining the full electromagnetic propagator as

$$D_{\mu \nu}(x, x') = \frac{\delta A_{\mu}^{\text{tot}}(x)}{\delta j_{\nu}^{\text{ext}}(x')},$$

(15)

a straightforward application of the functional chain rule in the form

$$\frac{\delta A_{\text{tot}}}{\delta j_{\text{ext}}} = \frac{\delta A_{\text{ext}}}{\delta j_{\text{ext}}} + \frac{\delta A_{\text{ind}}}{\delta j_{\text{ind}}} \frac{\delta A_{\text{ext}}}{\delta j_{\text{ext}}}$$

(16)

leads to the equation

$$D = D_0 + D_0 \chi D_0,$$

(17)

which relates $D$ to the fundamental response tensor $\chi$ via the free electromagnetic propagator $D_0$. Notice that the formal products refer to the natural multiplication in the linear space of $4 \times 4$ tensorial integral kernels.

Further, we introduce irreducible response functions $\tilde{\chi}$ specifying the response of the induced currents to the total (instead of external) four-potentials,

$$\tilde{\chi}_{\mu \nu}(x, x') = \frac{\delta j_{\mu}^{\text{ind}}(x)}{\delta A_{\nu}^{\text{tot}}(x')},$$

(18)

Again, the functional chain rule

$$\frac{\delta j_{\text{ind}}}{\delta A_{\text{ext}}} = \frac{\delta j_{\text{ind}}}{\delta A_{\text{tot}}} \frac{\delta A_{\text{tot}}}{\delta A_{\text{ext}}} = \frac{\delta j_{\text{ind}}}{\delta A_{\text{tot}}} + \frac{\delta j_{\text{ind}}}{\delta A_{\text{tot}}} \frac{\delta A_{\text{ind}}}{\delta j_{\text{ind}}} \frac{j_{\text{ind}}}{\delta A_{\text{ext}}}$$

(19)

shows that these quantities are related to the fundamental response tensor through

$$\chi = \tilde{\chi} + \tilde{\chi} D_0 \chi.$$  

(20)

In terms of these irreducible response functions, the tensor $D$ satisfies

$$D = D_0 + D_0 \tilde{\chi} D_0,$$

(21)
as follows again by a functional chain rule. This last equation has a Dyson-like structure and formally coincides with the Schwinger-Dyson equation for the full electromagnetic propagator in the sense of quantum electrodynamics under the identification of $\tilde{\chi}$ with the irreducible photon self-energy. In fact, the photon self-energy is given by the current-current correlation function evaluated in the vacuum, which corresponds to the Kubo formula for the fundamental response function $\chi$. This shows the literal analogy between quantum electrodynamics and solid state physics, where in the former case the vacuum plays the role of the polarizable medium. Moreover, our analysis provides via Eq. (15) a classical interpretation of the full electromagnetic propagator: just as $D_0$ propagates the electromagnetic four-potential in terms of its own sources, $D$ propagates the total four-potential in terms of the external four-current, i.e., to linear order $A^\mu_{\text{tot}} = D^\mu_{\nu} j^\nu_{\text{ext}}$. We note, however, that in quantum electrodynamics one usually works with time-ordered instead of retarded response functions.

A non-relativistic version of the Schwinger-Dyson equations is known in electronic structure theory as Hedin’s equations. These are among the most important first-principles techniques used nowadays for describing single-electron excitations in real materials (see or for a recent discussion in the context of Green function theory). They can be obtained from the relativistic Schwinger-Dyson equations by replacing the electronic Dirac propagator by the propagator of the Schrödinger or Pauli equation and approximating the full electromagnetic propagator in the Coulomb gauge by its 00-component. Indeed, keeping in Eq. (21) only the Coulomb potential $V = (D_0)_{00} e^2 c^2$ and the irreducible density-density response function $\tilde{P} = \tilde{\chi}_{00}/e^2 c^2$, we obtain the approximate relation for $W = (D)_{00} e^2 c^2$:

$$ W = V + V \tilde{P} W. $$

This is precisely Hedin’s equation for the screened potential $W$, where $\tilde{P}$ is known in electronic structure theory as the polarizability.

2.3. Average electromagnetic fields versus polarizations

From the functional point of view Eqs. (2)–(3) represent the inhomogeneous Maxwell equations relating the external fields to the external sources and the homogeneous Maxwell equations for the total fields, respectively. They hold on a microscopic scale and do not need to be derived as in the classical approach (based on the induction of electric and magnetic dipoles,
cf. [3, 4]). In this respect our approach agrees with the work of Hirst [35] and the metric-free approach as mentioned above [36, 37, 38, 40]. By comparison, we focus on the derivation of universal relations between electromagnetic response functions, and in this respect the advantages of our approach are: (i) By distinguishing between induced and external (instead of bound and free) charges and currents, it is independent of any assumption about the medium. External and induced sources are not necessarily located only “outside” or “inside” the material probe, respectively; instead this distinction implies that the external sources can be controlled experimentally [34]. Thus we use the term “external fields” in precisely the same sense as in classical or quantum mechanics, where one considers particles moving in external fields as opposed to particles in the fields created by themselves. (ii) It provides by Eqs. (5)–(7) a unique definition of microscopic polarization and magnetization without referring to the constitutive relations of the material. Explicitly, the induced fields are related to the induced charges and currents by the microscopic Maxwell equations

\[ -\nabla \cdot P(x, t) = \rho_{\text{ind}}(x, t), \quad \nabla \times M(x, t) + \frac{\partial}{\partial t} P(x, t) = j_{\text{ind}}(x, t), \quad (23) \]

\[ \nabla \cdot M(x, t) = 0, \quad -\nabla \times P(x, t) + \frac{1}{c^2} \frac{\partial}{\partial t} M(x, t) = 0. \quad (24) \]

\[2\]While the first two equations are standard also in the classical approach to electrodynamics in media, the analogy between \( P(x) \), \( M(x) \) and electromagnetic fields as generated by internal sources has been called “a deceptive parallel” in [4, Sec. 4.3.2, 6.3.2]. In macroscopic magnetostatics, e.g., the bar magnet with uniform magnetization parallel to its axis is considered an example where the condition \( \nabla \cdot M(x) = 0 \) does not hold. There the magnetization is assumed constant inside the cylinder and zero outside, hence a nonvanishing divergence appears at the top and bottom surfaces. However, the so defined field \( M(x) \) is not observable itself and indeed only serves to determine by \( \nabla \times M(x) = j(x) \) the current which is localized at the lateral surface of the cylinder. Any curl-free vector field can be added to \( M(x) \) without changing this information, fact which has been referred to as “gauge freedom” of the magnetization in [34, 55, 62]. A careful examination of the procedure described in [4] shows that the real, observable magnetic field is given in terms of the current by \( \nabla \times B(x) = \mu_0 j(x) \) and \( \nabla \cdot B(x) = 0 \), or in terms of the uniform magnetization by taking the transverse part \( B(x) = \mu_0 M_T(x) \). Similarly, in electrostatics the observable electric field is given in terms of the uniform polarization of a macroscopic body by the longitudinal part \( E(x) = -\varepsilon_0 P_L(x) \). Such problems actually do not concern electrodynamics in media but the determination of electromagnetic fields as generated by macroscopic surface currents or charges.
Thus our approach is suitable for deriving universal relations between electromagnetic response functions as shown in Section (iii) Macroscopically averaged fields defined by

\[ \langle E \rangle(x, t) = \int d^3x' f(x - x') E(x', t) \] (25)

(with \( f(x) \) being smooth, localized at \( x = 0 \) and ranging over distances large compared to atomic dimensions in the material) satisfy the same Maxwell equations as the microscopic fields, because the averaging procedure commutes with the partial derivatives \[5\]. Moreover, the relations \[1\] hold as exact identities while they are only first order approximations in other approaches such as \[63\].

The classical interpretation of \( P \) and \( M \) can be recovered from the microscopic identities \[3\]–\[7\], because the volume integral of the electric field is proportional to the total dipole moment of the corresponding charge density \[2, 3\]:

\[ \int d^3x E_i(x, t) = \sum_k \int d^3x (\partial_k x_i) E_k(x, t) = -\sum_k \int d^3x x_i \partial_k E_k(x, t), \] (26)

hence with \( \nabla \cdot E = \rho/\varepsilon_0 \) we obtain for the macroscopic polarization

\[ P(t) = -\varepsilon_0 \int \frac{1}{V} d^3x E_{\text{ind}}(x, t) = \frac{1}{V} \int d^3x x \rho_{\text{ind}}(x, t), \] (27)

where the integration range is the whole three-dimensional space. In this sense, one can regard \( P \) as macroscopic dipole moment even without assuming the presence of polarizable molecules as in the Clausius-Mossotti picture. Similarly, one finds in the case of static magnetic fields \[2, 3\]

\[ M = \frac{1}{\mu_0 V} \int d^3x B_{\text{ind}}(x) = \frac{1}{2V} \int d^3x x \times j_{\text{ind}}(x). \] (28)

Hence by this averaging procedure, the functional approach to electrodynamics in media recovers the classical approach.

Eqs. \[27\]–\[28\] are valid as long as the fields decay sufficiently fast at infinity, such that the boundary terms in the partial integration, e.g. in Eq. \[26\], can be neglected. However, the resulting formulas for \( P \) and \( M \) in terms of \( \rho_{\text{ind}} \) and respectively \( j_{\text{ind}} \) require an integration over the whole
sample volume, and hence are not useful for calculating the polarization of a crystalline solid from the charge and current distributions of periodic Bloch wavefunctions [14]. The Modern Theory of Polarization has therefore replaced Eq. (27) by a fundamental equation for the change in macroscopic polarization in terms of a transient current through the sample [13, 14] (see also [12, Eq. (22.4)] and [64, 65, 66]):

$$P(\Delta t) - P(0) = \int_0^{\Delta t} dt' j_{\text{ind}}(t'), \quad \text{where} \quad j_{\text{ind}}(t') = \frac{1}{V} \int d^3x' j_{\text{ind}}(x', t').$$

(29)

We now show that the identification of induced electromagnetic fields with electric and magnetic polarizations is consistent with this new approach, as Eq. (29) can be derived from the Maxwell equations for the microscopic induced fields (23)–(24): Integrating Ampère’s law over the whole space yields

$$\int d^3x \nabla \times M(x, t) = j_{\text{ind}}(t) - \frac{dP(t)}{dt}.$$  

(30)

If we can show that the left hand side vanishes under realistic assumptions, then a time integration leads immediately to (29). The equation of motion

$$\Box M(x, t) = \nabla \times j_{\text{ind}}(x, t),$$

which follows from (23)–(24), can be solved for the magnetization using the retarded Green function,

$$M(x, t) = \frac{1}{4\pi} \int d^3x' \nabla' \times j_{\text{ind}}(x', t')|_{t' = t_{\text{ret}}},$$

(31)

where $t_{\text{ret}} = t - |x - x'|/c$. Considering typical measurement setups for induced polarizations [14], we may assume that the current $j_{\text{ind}}(x, t)$ sets in at a finite time $t_0$ and stays in a finite volume $V$ for all times. This implies that for any time $t$, there exists $r > 0$ such that for all $x$ with $|x| > r$ and all $x' \in V$, we have $t_{\text{ret}} < t_0$ and consequently $M(x, t) = 0$. Consider now the integral

$$\int d^3x (\nabla \times M)_i(x, t) = \sum_k \int d^3x (\partial_k x_i)(\nabla \times M)_k(x, t).$$

(32)

By partial integration, this equals

$$- \sum_k \int d^3x x_i \partial_k (\nabla \times M)_k(x, t) + \sum_k \int d^3x \partial_k [x_i (\nabla \times M)_k(x, t)].$$

(33)
Both terms vanish, because the first one contains the divergence of a rotational vector field, and the second one can be converted to a surface integral, which is zero as
\[
\int_{|x|=R} dS x \cdot (\nabla \times M)(x, t) = 0 \quad \text{for any } R > r.
\] (34)

We have thus shown that the left hand side of Eq. (30) vanishes, and the fundamental equation of the Modern Theory of Polarization follows from Maxwell’s equations for the induced electromagnetic fields by macroscopic averaging. The advantage of Eq. (29) as compared to (27) is that it can be reexpressed—assuming a periodic current distribution in a crystalline solid—through the current of a single unit cell, while Eq. (24) cannot be expressed through the dipole moment of one unit cell [14].

3. Universal relations between electromagnetic response functions

3.1. Electromagnetic properties from fundamental response functions

The electromagnetic properties of any material are (within the linear regime and in the absence of magnetoelectric coupling) characterized by the electric susceptibility \( \leftrightarrow \chi_e \) giving the polarization in terms of the total electric field, \( P = \varepsilon_0 \leftrightarrow \chi_e E_{\text{tot}} \), the magnetic susceptibility \( \leftrightarrow \chi_m \) giving the magnetization in terms of the external magnetic field, \( \mu_0 \mu M = \leftrightarrow \chi_m B_{\text{ext}} \), and the conductivity specifying the response of the induced current to the external electric field, \( j_{\text{ind}} = \leftrightarrow \sigma E_{\text{ext}} \). Other common response functions are the relative permittivity or dielectric tensor defined by \( E_{\text{ext}} = \leftrightarrow \varepsilon E_{\text{tot}} \), and the relative magnetic permeability defined by \( B_{\text{tot}} = \leftrightarrow \mu B_{\text{ext}} \). These are related to the corresponding susceptibilities by \( \leftrightarrow \varepsilon = 1 + \leftrightarrow \chi_e \) and \( \leftrightarrow \mu = 1 + \leftrightarrow \chi_m \), respectively. All these response functions represent \( 3 \times 3 \) tensorial integral kernels with respect to the space and time variables. Our main goal in this subsection is to derive also general relations between \( \leftrightarrow \varepsilon \) and \( \leftrightarrow \sigma \), as well as \( \leftrightarrow \chi_m \) and \( \leftrightarrow \sigma \), showing that the microscopic conductivity tensor already contains the complete information on all linear electromagnetic responses.

In deriving electromagnetic materials properties from the fundamental response tensor we are facing two questions: (i) How is the response of \( P \) and \( M \) related to the induced four-current \( j_{\text{ind}}^\mu \), and (ii) how do we express the response to \( E_{\text{ext}} \) and \( B_{\text{ext}} \) in terms of the response to the external four-potential \( A_{\text{ext}}^\mu \). The first problem is solved under the identifications (5)–(7):
The Maxwell equations for the induced fields (23)–(24) imply the equations of motion

\[ \Box P(x, t) = \nabla \rho_{\text{ind}}(x, t) + \frac{1}{c^2} \frac{\partial}{\partial t} j_{\text{ind}}(x, t), \]  
(35)

\[ \Box M(x, t) = \nabla \times j_{\text{ind}}(x, t). \]  
(36)

By using the retarded Green function for the d’Alembert operator, \( \Box^{-1} = \frac{D_0}{\mu_0} \) (this coincides with the diagonal entries of the free electromagnetic propagator in the Lorentz gauge), we can thus obtain the induced fields in terms of the induced currents. As for the second problem, it is natural to consider the functional chain rule,

\[ \frac{\delta j_{\text{ind}}^\mu(x)}{\delta E_{\text{ext}}^\ell(x')} = \int d^4y \chi_\alpha^\mu(x, y) \frac{\delta A_{\text{ext}}^\alpha(y)}{\delta E_{\text{ext}}^\ell(x')}, \]  
(37)

and the analogous equation for the external magnetic field. However, the functional dependence of the four-potential on the electromagnetic fields depends on the gauge, and therefore it remains to show that the above equation is gauge independent. Since two potentials \( A^\mu \) and \( A'^\mu \) determining the same fields differ by a pure gauge, \( A^\mu = A'^\mu - \partial^\mu f \), it is enough to show that

\[ \int d^4y \chi_\alpha^\mu(x, y) \frac{\delta \partial^\alpha f(y)}{\delta E_{\text{ext}}^\ell(x')} = 0 \]  
(38)

for an arbitrary function \( f \). Using that the functional derivative commutes with the partial derivative and by a partial integration, this condition follows from the constraint (13) on the fundamental response function.

### 3.2. Conductivity as universal electromagnetic response

In a first step, we relate the microscopic conductivity tensor to the fundamental response tensor:

\[ \sigma_{kl}(x, x'; t - t') = \frac{\delta j_{\text{ind}}^k(x, t)}{\delta E_{\text{ext}}^l(x', t')} = \sum_i \int d^3y \int ds \frac{\delta j_{\text{ind}}^k(x, t)}{\delta A_{\text{ext}}^i(y, s)} \frac{\delta A_{\text{ext}}^i(y, s)}{\delta E_{\text{ext}}^l(x', t')} \]  
(39)

As shown in the previous section, the result does not depend on the gauge, hence we may choose \( \varphi_{\text{ext}} = 0 \) and \( E_{\text{ext}} = -\partial_t A_{\text{ext}} \). Thus we obtain

\[ A_{\text{ext}}(x, t) = -\int_{-\infty}^t dt' E_{\text{ext}}(x, t'), \]  
(40)
and the formal functional derivative
\[
\frac{\delta A^k_{\text{ext}}(\mathbf{x}, t)}{\delta E^\ell_{\text{ext}}(\mathbf{x}', t')} = -\delta_{k\ell} \theta(t - t') \delta(\mathbf{x} - \mathbf{x}').
\] (41)

This leads to the well-known result [49]
\[
\sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; t - t') = -\int_{-\infty}^{t-t'} ds \chi^k_{\ell}(\mathbf{x}, \mathbf{x}'; s),
\] (42)
or by Fourier transformation with respect to the time variable\[3\]
\[
\sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; \omega) = \frac{1}{i\omega} \chi^k_{\ell}(\mathbf{x}, \mathbf{x}'; \omega).
\] (44)

It follows that the microscopic conductivity tensor contains the complete information on all linear electromagnetic responses [44], because $\chi^\mu_{\nu}$ can be reconstructed from $\sigma_{k\ell}$: Using the constraints (13), we can express also $\chi^0_{0}$, $\chi^0_{\ell}$ and $\chi^\ell_{0}$ in terms of $\sigma_{k\ell}$ as follows:
\[
\chi^0_{\ell}(\mathbf{x}, \mathbf{x}'; \omega) = c \sum_k \frac{\partial}{\partial x_k} \sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; \omega),
\] (45)
\[
\chi^0_{\ell}(\mathbf{x}, \mathbf{x}'; \omega) = c \sum_\ell \frac{\partial}{\partial x'^\ell} \sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; \omega),
\] (46)
\[
\chi^0_{0}(\mathbf{x}, \mathbf{x}'; \omega) = \frac{c^2}{i\omega} \sum_{k, \ell} \frac{\partial}{\partial x^k} \frac{\partial}{\partial x'^\ell} \sigma_{k\ell}(\mathbf{x}, \mathbf{x}'; \omega).
\] (47)

### 3.3. Generalized dielectric tensor

As a first example we derive the most general relation between the frequency and wave-vector dependent dielectric tensor and the microscopic conductivity tensor. The former quantity determines the (magneto)optical properties of media [67] and contains the entire information about elementary

---

\[3\] The Fourier transformation of a general response function $F(\mathbf{x}, \mathbf{x}'; t - t')$ is defined by
\[
F(\mathbf{x}, \mathbf{x}'; t - t') = \int d^3k \int d\omega \int d^3k' e^{i(k \cdot \mathbf{x} - k' \cdot \mathbf{x}') - i\omega(t - t')} F(k, k'; \omega).
\] (43)

In our convention for indices, any genuine $3 \times 3$ tensor has only lower indices, e.g. $\sigma_{k\ell} = \delta j^k_{\text{ind}} / \delta E^\ell_{\text{ext}}$, whereas $\chi^k_{\ell}$ denotes the spatial components of the $4 \times 4$ Lorentz tensor $\chi^\mu_{\nu}$.
excitations and collective modes in solids [68]. With $E_{\text{tot}} = E_{\text{ext}} + E_{\text{ind}}$ and $P = -\varepsilon_0 E_{\text{ind}}$, we have
\[
[(\varepsilon^{-1})_{ij}(x, x')] = \frac{\delta E_{\text{tot}}^i(x)}{\delta E_{\text{ext}}^j(x')} = \delta_{ij}\delta(x - x') + (\varepsilon_{\text{eE}})_{ij}(x, x'), \tag{48}
\]
where the second term determines the response of the induced to the external electric field and is given by the fundamental relation
\[
(\chi_{\text{eE}})_{ij}(x, x') = -\frac{1}{\varepsilon_0} \delta_{ij} \frac{\delta P_i(x)}{\delta E_{\text{ext}}^j(x')} = -\frac{1}{\varepsilon_0} \int d^4y \int d^4y' \frac{\delta P_i(x)}{\delta j^i_{\text{ind}}(y)} \chi_{\nu \mu}(y, y') \frac{\delta A_{\nu}^\nu(y')}{\delta E_{\text{ext}}^j(x')}.	ag{49}
\]
The first term in the integrand is evaluated as described in Section 3.1: With the retarded Green function of the d'Alembert operator (see e.g. [3])
\[
D_0(x - y; t - s) = \frac{\mu_0}{4\pi} \frac{\delta(t - t' - |x - x'|/c)}{|x - x'|} \tag{50}
\]
we can express the induced field as
\[
P_i(x, t) = \int d^3y \int ds \frac{1}{\mu_0} D_0(x - y; t - s) \left( \frac{\partial}{\partial y^i} \rho_{\text{ind}}(y, s) + \frac{1}{c^2} \frac{\partial}{\partial s} j^i_{\text{ind}}(y, s) \right). \tag{51}
\]
Hence after a partial integration, we obtain the functional derivatives
\[
\frac{1}{c} \frac{\delta P^i(x, t)}{\delta \rho_{\text{ind}}(y, s)} = -\frac{1}{c\mu_0} \frac{\partial}{\partial y^i} D_0(x - y; t - s), \tag{52}
\]
\[
\frac{\delta P^i(x, t)}{\delta j^k_{\text{ind}}(y, s)} = -\delta_{ik} \frac{1}{c^2 \mu_0} \frac{\partial}{\partial s} D_0(x - y; t - s). \tag{53}
\]
The last term in the integrand of Eq. (49) is again evaluated in the gauge where $\varphi_{\text{ext}} = 0$, such that
\[
\frac{\delta A_{\nu}^\nu(y', s')}{\delta E_{\text{ext}}^j(x', t')} = -\delta_{ij} \theta(s' - t') \delta(y' - x'). \tag{54}
\]
Putting (52)–(54) into (48)–(49), performing a partial integration and reexpressing the fundamental response tensor in terms of the conductivity tensor by means of (44)–(47), we arrive at
\[
[(\varepsilon^{-1})_{ij}(x, x'; t - t')] = \delta_{ij}\delta(t - t')\delta(x - x') - \int d^3y \int ds \int ds' D_0(x - y; t - s) \left( -c^2 \frac{\partial}{\partial y^i} \frac{\partial}{\partial y^m} + \delta_{im} \frac{\partial^2}{\partial s^2} \right) \sigma_{mj}(y, x'; s - s') \theta(s' - t'). \tag{55}
\]
Here and in the following we sum over all doubly appearing indices. Eq. (55) represents the most general relation between the generalized dielectric tensor and the microscopic conductivity tensor (assuming only homogeneity in time). By Fourier transformation with respect to the space and time variables it is equivalent to

$$
[(\hat{\varepsilon})^{-1}]_{ij}(k, k'; \omega) = \delta_{ij} - \frac{i}{\omega} D_0(k; \omega) \left( c^2 k_i k_m - \omega^2 \delta_{im} \right) \sigma_{mj}(k, k'; \omega). \quad (56)
$$

In matrix notation this identity can be written more compactly as

$$
(\hat{\varepsilon})^{-1}(k, k'; \omega) = \hat{1} - \frac{i}{\omega} D_0(k; \omega) \left( c^2 |k|^2 \hat{P}_L(k) - \omega^2 \hat{1} \right) \hat{\sigma}(k, k'; \omega), \quad (57)
$$

where \((P_L)_{ij}(k) = k_i k_j / |k|^2\) denotes the projection operator on the longitudinal part. Let us now see how this general relation simplifies by restricting to special cases. First, in the non-relativistic limit where \(\omega \ll c|k|\) and hence retardation effects are neglected, the propagator is given by \(D_0(k; \omega) = \mu_0 / |k|^2\) and therefore,

$$
(\hat{\varepsilon})^{-1}(k, k'; \omega) = \hat{1} - \frac{i}{\omega \varepsilon_0} \hat{P}_L(k) \hat{\sigma}(k, k'; \omega). \quad (58)
$$

Second, in a homogeneous medium the conductivity is proportional to \(\delta(k - k')\), hence it depends only on one momentum \(k\). Then the same is true for the dielectric tensor and we can write

$$
(\hat{\varepsilon})^{-1}(k, \omega) = \hat{1} - \frac{i}{\omega \varepsilon_0} \hat{P}_L(k) \hat{\sigma}(k, \omega). \quad (59)
$$

Third, specializing to an isotropic medium where

$$
\hat{\sigma}(k, \omega) = \sigma_L(k, \omega) \hat{P}_L(k) + \sigma_T(k, \omega) \hat{P}_T(k) \quad (60)
$$

with longitudinal and transverse conductivities \(\sigma_L\) and \(\sigma_T\), respectively, we obtain the simpler relation

$$
(\hat{\varepsilon})^{-1}(k, \omega) = \hat{1} - \frac{i}{\omega \varepsilon_0} \sigma_L(k, \omega) \hat{P}_L(k). \quad (61)
$$

In particular, the longitudinal dielectric constant \(\varepsilon_L\) is related to the longitudinal conductivity by

$$
\varepsilon_L^{-1}(k, \omega) = 1 - \frac{i}{\omega \varepsilon_0} \sigma_L(k, \omega), \quad (62)
$$

which is a well-known identity [69 Eq. (6.51)]. We have shown that it follows from the general relation (57) in a non-relativistic approximation and by restricting to homogeneous, isotropic materials.
3.4. Generalized magnetic susceptibility

As a second example consider the generalized magnetic susceptibility, which contains the description of magnetic orderings and excitation spectra and is directly related to the neutron scattering cross section \[70\]. Again we start from the fundamental relation

\[ (\chi_m)_{ij}(x, x') = \frac{\delta M^i(x)}{\delta B^j_{\text{ext}}(x')} = \int d^4y \int d^4y' \frac{\delta M^i(x)}{\delta j^\mu_{\text{ind}}(y)} \chi^{\mu\nu}(y, y') \frac{\delta A^\nu_{\text{ext}}(y')}{\delta B^j_{\text{ext}}(x')}. \]  

(63)

Similar as above, we obtain for the first term

\[ \frac{\delta M^i(x, t)}{\delta j^k_{\text{ind}}(y, s)} = -\frac{1}{\mu_0} \varepsilon_{imk} \frac{\partial}{\partial y^m} D_0(x - y, t - s), \]  

(64)

where \( \varepsilon_{imk} \) is the Levi-Civita symbol. For the external vector potential we choose the Coulomb gauge where \( \delta A^0/\delta B^i = 0 \) and \( \nabla \cdot A = 0 \). Then we can express the vector potential explicitly in terms of the magnetic field as

\[ A_{\text{ext}}(x, t) = \frac{1}{4\pi} \nabla \times \int d^3x' \frac{B_{\text{ext}}(x', t)}{|x - x'|}. \]  

(65)

This yields for the third term in the above integral,

\[ \frac{\delta A^l_{\text{ext}}(y')}{\delta B^j_{\text{ext}}(x')} = -\frac{1}{4\pi} \varepsilon_{jln} \frac{\partial}{\partial y^n} \frac{\delta(s' - t')}{|y' - x'|}. \]  

(66)

Putting (64) and (66) into (63) and by partial integration with respect to the \( y^m \) and \( y^m \) variables, we obtain the general identity

\[ (\chi_m)_{ij}(x, x'; t - t') = \int d^3y \int ds \int d^3y' \int ds' \frac{1}{\mu_0} D_0(x - y; t - s) \left( \varepsilon_{imk} \varepsilon_{jnl} \frac{\partial}{\partial y^n} \chi^k_l(y, y'; s - s') \right) \frac{1}{4\pi} \frac{\delta(s' - t')}{|y' - x'|}. \]  

(67)

\[ ^4 \text{In this context, if } \chi^{\mu\nu}_m \text{ is associated with the response of a microscopic charge current to the electromagnetic potential, the corresponding response function } \chi_m \text{ refers only to the orbital contribution to the magnetic susceptibility } [71]. \text{ Spin contributions can be included in our picture by a divergence free contribution to the current of the Pauli equation, as it is motivated by the non-relativistic limit of the Dirac equation (cf. [72] chap. XX, §29).} \]
By Fourier transformation with respect to the space and time variables,

\[
(\chi_m)_{ij}(k, k'; \omega) = \frac{1}{\mu_0} D_0(k; \omega) \left( \varepsilon_{imk} \varepsilon_{jn\ell} k_m k'_n \chi_{\ell}(k, k'; \omega) \right) \frac{1}{|k'|^2}.
\]  

(68)

Reexpressing the fundamental response function \(\chi^k_{\ell}\) in terms of \(\sigma_{k\ell}\) by (44) yields a closed formula for the generalized magnetic susceptibility in terms of the microscopic conductivity tensor. Let us simplify again Eq. (68) by restricting to special cases. First, neglecting retardation effects \((\omega \ll c|k|)\), we have

\[
(\chi_m)_{ij}(k, k'; \omega) = \frac{1}{|k|^2} \left( \varepsilon_{imk} \varepsilon_{jn\ell} k_m k'_n \chi_{\ell}(k, k'; \omega) \right) \frac{1}{|k'|^2}.
\]  

(69)

In homogeneous materials, this formula reduces to

\[
(\chi_m)_{ij}(k, \omega) = \frac{1}{|k|^4} \varepsilon_{imk} \varepsilon_{jn\ell} k_m k'_n \chi_{\ell}(k, \omega),
\]  

(70)

which agrees for \(\omega = 0\) with [73, Eq. (A1.6)]. In isotropic media, where

\[
\chi^k_{\ell}(k, \omega) = \chi_L(k, \omega) \frac{k_k k_\ell}{|k|^2} + \chi_T(k, \omega) \left( \delta_k\ell - \frac{k_k k_\ell}{|k|^2} \right)
\]  

(71)

with longitudinal and transverse response functions \(\chi_L\) and \(\chi_T\), respectively, we further obtain

\[
(\chi_m)_{ij}(k, \omega) = \frac{\chi_T(k, \omega)}{|k|^2} \left( \delta_{ij} - \frac{k_i k_j}{|k|^2} \right).
\]  

(72)

Since \(\chi_m\) acts on magnetic fields, which are purely transverse, the transverse projection operator in brackets can be omitted, and we arrive at

\[
\chi_m(k, \omega) = \frac{\chi_T(k, \omega)}{|k|^2}.
\]  

(73)

In particular, we recover the well-known formula for the (orbital) magnetic susceptibility [49, Eq. (3.183)],

\[
\chi_m = \lim_{|k| \to 0} \frac{\chi_T(k, \omega = 0)}{|k|^2},
\]  

(74)

where the limit \(|k| \to 0\) corresponds to integrating out spatial dependence. The identity [68] generalizes this result to inhomogeneous, anisotropic media and includes relativistic retardation effects.
3.5. Electromagnetic cross-coupling responses

In the most general case, there are besides electric and magnetic susceptibilities also cross-coupling (magnetoelastic) coefficients, which specify the response of an induced magnetic field to an external electric field and vice versa \[52\]. We can write the most general linear constitutive relations as

\[
\begin{align*}
E_{\text{ind}} &= i\chi_{eE} E_{\text{ext}} + c\chi_{eB} B_{\text{ext}}, \\
B_{\text{ind}} &= \frac{1}{c}i\chi_{bE} E_{\text{ext}} + \chi_{bB} B_{\text{ext}}.
\end{align*}
\] (75) (76)

The factor \( c = 1/\sqrt{\varepsilon_0 \mu_0} \) was introduced such that the response functions \( \chi_{eE}, \chi_{eB}, \chi_{bE}, \chi_{bB} \) are dimensionless. Within the functional approach, the cross-coupling responses can be calculated as partial functional derivatives analogous as in the two examples above, and we summarize all results by the following set of equations:

\[
\begin{align*}
(\chi_{eE})_{ij}(\mathbf{k}, \mathbf{k}'; \omega) &= D_0(\mathbf{k}; \omega) \left( \delta_{im} - \frac{e^2 k_i k_m}{\omega^2} \right) i\omega \sigma_{mj}(\mathbf{k}, \mathbf{k}'; \omega) \quad (77) \\
(\chi_{eB})_{ij}(\mathbf{k}, \mathbf{k}'; \omega) &= D_0(\mathbf{k}; \omega) \left( \delta_{im} - \frac{e^2 k_i k_m}{\omega^2} \right) \left( \varepsilon_{jnl} \frac{\omega k_n'}{|\mathbf{k}'|^2} \right) i\omega \sigma_{ml}(\mathbf{k}, \mathbf{k}'; \omega) \quad (78) \\
(\chi_{bE})_{ij}(\mathbf{k}, \mathbf{k}'; \omega) &= D_0(\mathbf{k}; \omega) \left( \varepsilon_{imk} \frac{ck_m}{\omega} \right) i\omega \sigma_{jk}(\mathbf{k}, \mathbf{k}'; \omega) \quad (79) \\
(\chi_{bB})_{ij}(\mathbf{k}, \mathbf{k}'; \omega) &= D_0(\mathbf{k}; \omega) \left( \varepsilon_{imk} \frac{ck_m}{\omega} \right) \left( \varepsilon_{jnl} \frac{\omega k_n'}{|\mathbf{k}'|^2} \right) i\omega \sigma_{kl}(\mathbf{k}, \mathbf{k}'; \omega) \quad (80)
\end{align*}
\]

Again we sum over all doubly appearing indices and

\[D_0(\mathbf{k}; \omega) = \frac{\mu_0}{|\mathbf{k}|^2 - (\omega/c + i\eta)^2}\] (81)

with \( \eta \to 0^+ \) denotes the retarded Green function of the d’Alembert operator. These relations between electromagnetic response functions are universal, i.e. valid in any material, owing to our model-independent definition of electric and magnetic polarizations \[5\]. We have derived them for the microscopic response functions, but they hold as well between the corresponding response functions of macroscopically averaged fields, as e.g.

\[
gf{k}{t} = \frac{\delta(j_{\text{ind}})(\mathbf{x}, t)}{\delta(E_{\text{ext}})(\mathbf{x}', t')} \quad (82)
\]
with the averages on the right hand side defined by Eq. (25). (When the variation of the external fields over atomic scales in the material is negligible, we may have \( \langle E_{\text{ext}} \rangle(x, t) \simeq E_{\text{ext}}(x, t) \).) The reason for this is that the average fields satisfy the same Maxwell equations as the microscopic fields (cf. Section 2.3), which we have used in the derivation. Furthermore, the universal relations hold in any inertial frame, as they are derived from the Lorentz tensor of fundamental response functions. Given a particular microscopic model of the medium, Eqs. (77)–(80) can—at least in principle—be evaluated in the Kubo formalism, where the conductivity tensor is expressed by a current-current correlation function [47].

Finally, let us relate the causal response functions defined above to the constitutive dyadics used in the context of bianisotropic materials [16, 52]. These are defined by

\[
D = \varepsilon_0 \hat{\varepsilon} E + \sqrt{\varepsilon_0 \mu_0} \hat{\xi} H, \\
B = \mu_0 \hat{\mu} H + \sqrt{\varepsilon_0 \mu_0} \hat{\zeta} E,
\]

where factors \( \mu_0, \varepsilon_0 \) were again introduced such that \( \hat{\varepsilon}, \hat{\xi}, \hat{\zeta}, \hat{\mu} \) are dimensionless. With the identifications (5)–(7), these relations are equivalent to

\[
E_{\text{ext}} - \frac{c}{\varepsilon} \hat{\xi} B_{\text{ext}} = \hat{\varepsilon} E_{\text{tot}}, \\
B_{\text{tot}} - \frac{1}{c} \hat{\zeta} E_{\text{tot}} = \hat{\mu} B_{\text{ext}}.
\]

The so defined constitutive dyadics do not constitute causal response functions, i.e. they are not retarded, as only the induced fields are causally connected to the external fields (but not, e.g., the external to the total fields). A straightforward calculation yields the relationship between the constitutive

\[\text{In carrying over the universal relations to a macroscopic scale, however, one must keep in mind the following limitation: Each macroscopic material property is usually dominated by a certain class of degrees of freedom, e.g. the conductivity by the conduction electrons’ orbital motion, whereas the magnetic susceptibility is often dominated by localized spin degrees of freedom. Thus by describing each macroscopic response only in terms of its most relevant degrees of freedom, the universal relations between them get lost. By contrast, the microscopic response function in principle refers to all degrees of freedom contributing to the electromagnetic four-current, and by the universal relations each degree of freedom contributes to any electromagnetic response.}\]
dyadics and the causal response functions,

\[ \varepsilon = (1 + \chi_{eE})^{-1} \]  \hspace{1cm} (87)
\[ \xi = - (1 + \chi_{eE})^{-1} \chi_{eB} \]  \hspace{1cm} (88)
\[ \zeta = \chi_{bE} (1 + \chi_{eE})^{-1} \]  \hspace{1cm} (89)
\[ \mu = 1 + \chi_{bB} - \chi_{bE} (1 + \chi_{eE})^{-1} \chi_{eB} \]  \hspace{1cm} (90)

Combining these equations with Eqs. (77)–(80), we have thus shown that even in the case of a bianisotropic medium there are only nine independent response functions, and the four constitutive dyadics (each with nine components) can be expressed completely in terms of the conductivity tensor.

4. Conclusion

We have developed a functional approach which disentangles electrodynamics in media from macroscopic electrodynamics: While the latter corresponds to spatial and/or temporal averaging of fields which can be performed with or without media, electrodynamics in media is set up by regarding induced quantities as functionals of external perturbations on the microscopic or on the macroscopic scale. In terms of first order functional derivatives, this approach describes a unified, relativistic account of electromagnetic materials responses. We have shown that the equivalence of several response tensors translates into the Schwinger-Dyson equations of quantum electrodynamics or Hedin’s equations of electronic structure theory. While being independent of model assumptions on the material’s composition of electric and magnetic dipoles, the functional approach recovers both the classical approach to electrodynamics in media and the fundamental equation of the Modern Theory of Polarization on the level of macroscopically averaged fields. Within the microscopic domain, we have established universal (material-independent) relations between the generalized dielectric tensor, generalized magnetic susceptibility and the conductivity tensor. These reduce to well-known identities in special cases, but apply more generally to nonisotropic, inhomogeneous media; they take into account relativistic retardation effects and hold in any inertial frame. Moreover, we have derived general expressions for the cross-coupling coefficients between electric and magnetic fields, which may provide a theoretical basis for the quantitative description of magnetoelectric effects especially in bianisotropic materials.
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