

# On the form of constitutive equations in electrodynamics

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**Abstract.** A comparative analysis of various forms of the Maxwell equations for condensed matter is presented. It is shown that the so-called Casimir form contains enough information to solve any electromagnetic problem. The Landau – Lifshitz form intended for describing media with spatial dispersion requires an additional constitutive equation for the surface current, which does not set an additional boundary condition but acts as a replacement of usual Maxwell's continuity conditions for tangential field components.

## 1. Introduction

The creation of chiral, percolation, and other types of new artificial materials challenged the researchers to develop an adequate description of phenomena observed in these materials. The important factors a new theory is to account for are the multipole interaction and retardation effects on the inhomogeneity scale. Previous theories of this kind described phenomena occurring in crystalline or diluted systems [1–4]. Unfortunately, these theories only treated small effects in terms of perturbation theory — an approach allowing one not to bother much about the rigorous definition of the quantities and concepts used. Today, we cannot indulge in this liberty anymore. This is particularly true for the description of chiral media. There are currently two scientific schools employing two different forms of the Maxwell equations: one proposed by Landau and Lifshitz, and the other by Born and Fedorov. When the general aspects of spatial dispersion are discussed, the former is often referred to as a Casimir form and even though it is harder to justify, we will show below that it provides a more simple and consistent description of many phenomena.

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## 2. Various forms of the Maxwell equations in material media

At high frequencies, electromagnetic field disturbances due to the presence of the medium microinhomogeneities<sup>1</sup> may be of a solenoidal nature [5]. As a consequence, the induced currents — whether displacement [6] or conduction ones [7, 8] — lead to the artificial magnetization of the heterogeneous medium, even though the medium is a locally nonpermeable. On the other hand, the external field of a plane wave is of solenoidal character in itself. On going over to a macroscopic picture of electromagnetic phenomena in microinhomogeneous media, it is essential to distinguish between the solenoidal nature of the external field and that associated with the inhomogeneity of the medium [5]. Besides, introducing the permittivity and permeability concepts in a consistent way requires the decomposition of the induced current into two parts, each accounting either for the magnetization or the electric polarization only. What makes this decomposition problematic is that there are generally different scales on which the material properties of the medium fluctuate. The current at a given point may at one and the same time contribute to the electric polarization on one scale, and to the magnetization on another [9]. Until recently, both the procedures appeared to be not only cumbersome but ambiguous as well [5, 10]. To avoid all these problems, Landau and Lifshitz [5] (see also books [10, 11]) suggested incorporating all the induced currents into the generalized polarization as follows:

$$\mathbf{j} = \frac{\partial \mathbf{P}^{\text{LL}}}{\partial t} = \frac{1}{4\pi} \frac{\partial (\mathbf{D}^{\text{LL}} - \mathbf{E})}{\partial t}, \quad (1)$$

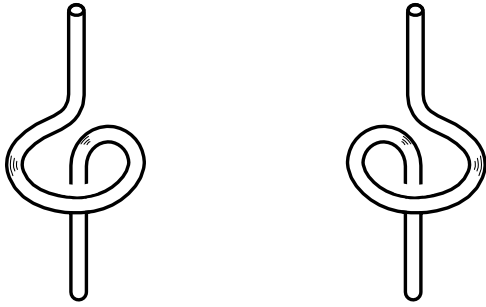
where  $\mathbf{D}^{\text{LL}} = \mathbf{E} + 4\pi \mathbf{P}^{\text{LL}}$ ,  $\mathbf{P}^{\text{LL}}$ , and  $\mathbf{E}$  are the macroscopic values of the generalized induction, generalized polarization, and electric field intensity, respectively. To close the Maxwell equations, a constitutive equation relating  $\mathbf{P}^{\text{LL}}$  or  $\mathbf{D}^{\text{LL}}$  to  $\mathbf{E}$

<sup>1</sup> By microinhomogeneity we mean such changes in material parameters for which the spatial scale is small compared with the wavelength in vacuum but large enough for the usual Maxwell constitutive equations to be employed.

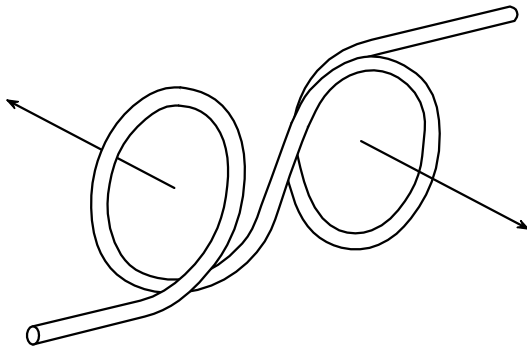
must be added. In the general case these relations will be nonlocal [5, 11]. For time-harmonic fields<sup>2</sup>, the most general form of a linear constitutive equation is given by

$$D_i^{\text{LL}}(\omega, \mathbf{r}) = \int \varepsilon_{ij}^{\text{LL}}(\mathbf{r}, \mathbf{r}', \omega) E_j(\mathbf{r}', \omega) dV'. \quad (2)$$

Generally speaking (see, for example, Ref. [39]),  $\mathbf{P}^{\text{LL}}$  and  $\mathbf{D}^{\text{LL}}$  may depend not only on the electric field but also with the magnetic field (Fig. 1) and even depend on the spatial derivatives (Fig. 2) of the magnetic field. Reducing the constitutive equation to the form (2) implies that both the magnetic field and all its spatial derivatives can be expressed in terms of the electric field using the Maxwell equations. These latter, as is well known, have a scale of length all their own — the wavelength — which may appear as a parameter in the expression for the kernel in Eqn (2). This makes  $\varepsilon_{ij}^{\text{LL}}$  in Eqn (2) a fairly complex operator reflecting not only the properties of the medium but those of the Maxwell equations as well.



**Figure 1.** Placing chiral particles in a magnetic field parallel to their axis induces an electric dipole moment in them.



**Figure 2.** Inclusion with a dipole moment proportional to the spatial derivative of an alternating magnetic field. The arrows indicate the instantaneous values of the field.

For a translationally invariant (and, of necessity, infinite) medium one can introduce the permittivity tensor dependent on the frequency and the wave vector as follows

$$\varepsilon_{ij}^{\text{LL}}(\mathbf{k}, \omega) = \int_0^\infty d\tau \int d(\mathbf{r} - \mathbf{r}') \times \{ \varepsilon_{ij}^{\text{LL}}(\mathbf{r} - \mathbf{r}', \tau) \exp[i\omega\tau - i\mathbf{k}(\mathbf{r} - \mathbf{r}')] \}. \quad (3)$$

<sup>2</sup> Fields are assumed to vary with time as  $\exp(-i\omega t)$ .

It is the latter dependence which gave the name ‘spatial dispersion’ to the effects related to the nonlocality of constitutive equations.

Thus, the Landau–Lifshitz (LL) form of the Maxwell equations links only three fields, namely,  $\mathbf{D}^{\text{LL}}$ ,  $\mathbf{E}$ , and  $\mathbf{B}$ :

$$e_{ij} k_j E_i = k_0 B_i, \quad (4)$$

$$e_{ij} k_j B_i + k_0 D_i^{\text{LL}} = -i \frac{4\pi}{c} (j_{\text{ext}})_i, \quad (5)$$

$$k_i B_i = 0, \quad ik_i D_i^{\text{LL}} = 4\pi \rho_{\text{ext}}, \quad (6)$$

where  $k_0 = \omega/c$ .

If it is possible in the process of homogenization to decompose the induced current into the average, eddy, and saddle-shaped parts (Fig. 3), then the Casimir form of the constitutive equations can be employed. This approach uses the following representation of the macroscopic current [9, 12]:

$$\mathbf{j} = \frac{\partial \mathbf{P}}{\partial t} - \left[ \mathbf{V} \cdot \frac{\partial \hat{\mathcal{Q}}}{\partial t} \right] + c[\mathbf{V} \times \mathbf{M}], \quad (7)$$

where  $\mathbf{j}$ ,  $\mathbf{P}$ , and  $\hat{\mathcal{Q}}$  are the macroscopic densities of current and of the electric dipole and electric quadrupole moments, and  $\mathbf{M}$  is the macroscopic density of the magnetic dipole moment. The polarization current  $\partial \mathbf{P} / \partial t$  is an average of the induced current (Fig. 3a). The magnetization current  $c[\mathbf{V} \times \mathbf{M}]$  includes all the microscopic currents that close up in the volume of averaging (Fig. 3b). The quadrupole part of the current,  $-\left[ \mathbf{V} \cdot (\partial \hat{\mathcal{Q}} / \partial t) \right]$ , contains the remaining part of the induced currents, namely, the microscopic currents which originate and terminate at the walls of the volume of averaging (Fig. 3c) and that part of the currents closed within the volume whose magnetic moments are mutually compensated for. This qualitative picture illustrates the rigorous mathematical fact that any vector field  $J_i$  can be represented in the form

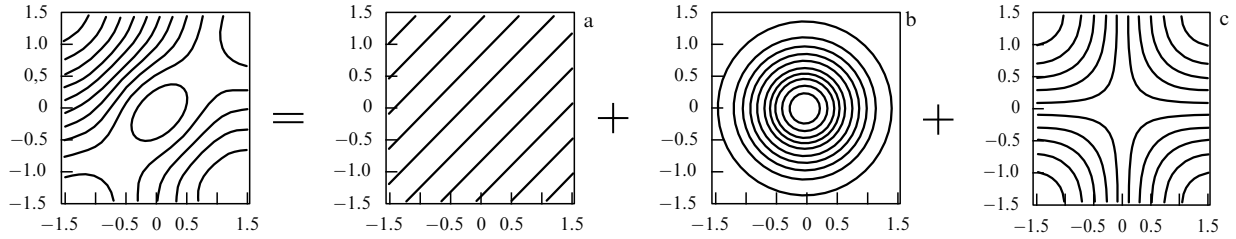
$$J_i \equiv ce_{ijk} \frac{\partial}{\partial x_j} \left( \frac{1}{2c} e_{klm} x_l J_m \right) + \frac{\partial}{\partial x_k} \left( \frac{1}{2} (x_i J_k + x_k J_i) \right) - x_i \frac{\partial}{\partial x_k} J_k.$$

Of course, the current distribution may also possess higher moments but, as discussed in Ref. [9], these can be neglected if the homogenization procedure is considered as averaging over a physically infinitesimal volume. Notice that Eqn (7) is not a truncated perturbation series but a rigorous result (cf. Ref. [9]) of the unique decomposition of the macroscopic current. Moreover, on each scale of averaging (see Ref. [9] for more details) the quantities  $\mathbf{P}$ ,  $\hat{\mathcal{Q}}$ , and  $\mathbf{M}$  are calculated from the standard formulas for the electric dipole, electric quadrupole, and magnetic dipole moments taken about the centre of the volume [12]:

$$\frac{\partial}{\partial t} \mathbf{P} = - \int (\mathbf{x} \operatorname{div} \mathbf{J}) d^3x,$$

$$\frac{\partial}{\partial t} Q_{ij} = \frac{1}{2} \int (x_i J_j + x_j J_i) d^3x,$$

$$\mathbf{M} = \frac{1}{2c} \int [\mathbf{x} \times \mathbf{J}] d^3x.$$



**Figure 3.** Schematic of the induced current decomposition into the average (a), eddy (b), and saddle-shaped (c) currents.

The next step is, first, to introduce the magnetic field  $\mathbf{H}$  and the permeability  $\mu$ :

$$\mathbf{H} = \mathbf{B} - 4\pi\mathbf{M}, \quad \mathbf{B} = \mu\mathbf{H}, \quad (8)$$

and, second, to redefine the electric displacement [9, 12–15]:

$$\mathbf{D}^C = \mathbf{E} + 4\pi\mathbf{P} - 4\pi[\nabla \cdot \hat{Q}]. \quad (9)$$

Finally, the resulting Maxwell equations take the Casimir (C) form traditionally used for the case of negligible spatial dispersion:

$$e_{ijl}k_j H_l + k_0 D_i^C = -i \frac{4\pi}{c} (j_{\text{ext}})_i, \quad e_{ijl}k_j E_l - k_0 B_i = 0, \quad (10)$$

$$k_j B_j = 0, \quad ik_j D_j^C = 4\pi\rho_{\text{ext}}. \quad (11)$$

In the framework of the C form, the relationship between the macroscopic moment densities and macroscopic fields may also be nonlocal, viz.

$$D_i^C(t, \mathbf{r}) = \int_0^\infty \int \varepsilon_{ij}^C(\mathbf{r}, \mathbf{r}', \tau) E_j(\mathbf{r}', t - \tau) dV' d\tau, \quad (12)$$

$$B_i(t, \mathbf{r}) = \int_0^\infty \int \mu_{ij}^C(\mathbf{r}, \mathbf{r}', \tau) H_j(\mathbf{r}', t - \tau) dV' d\tau. \quad (13)$$

Since no simplifying assumptions have been made thus far in deriving the LL and C forms, these forms should yield an equivalent description of the phenomena. In other words, a relation should exist between the tensor in Eqn (3) and the Fourier transforms of the tensors in Eqns (12) and (13).

Eliminating  $D_i^C$ ,  $D_i^{\text{LL}}$ , and  $H_i$  from the Maxwell equations and assuming that the fields  $\mathbf{E}$  and  $\mathbf{B}$  are identical in both forms — which is a not quite evident assumption and will be discussed below — we obtain the following relationship between the LL and C tensor forms (for the sake of convenience, Fourier representation of variables is used, see Ref. [16]):

$$\varepsilon_{ij}^{\text{LL}} = \varepsilon_{ij}^C + k_0^{-2} e_{ilm} e_{jnp} k_l k_n [\delta_{mp} - (\mu_{mp}^C)^{-1}]. \quad (14)$$

Depending on the authors' personal preferences (see the discussion in Ref. [17]), relation (14) is treated differently in the scientific literature. First, it is often viewed as expressing the equivalence of the two forms of the constitutive equations [5, 10, 11]. It is then assumed that in the isotropic case  $\varepsilon_{ij}^C$  and  $\mu_{ij}^C$  are in fact scalars, and  $\varepsilon_{ij}^{\text{LL}}$  is considered to be a tensor whose structure is determined by the wave vector

$$\varepsilon_{ij}^{\text{LL}}(\omega, \mathbf{k}) = \varepsilon^{\text{LLtr}}(\omega, k) \left( \delta_{ij} - \frac{k_i k_j}{k^2} \right) + \varepsilon^{\text{LLl}}(\omega, k) \frac{k_i k_j}{k^2}, \quad (15)$$

with  $\varepsilon^{\text{LLtr}}(\omega, k)$  and  $\varepsilon^{\text{LLl}}(\omega, k)$  being scalars. In this case we have two LL parameters and two C parameters and can establish a one-to-one correspondence between these parameters [10, 18]:

$$\varepsilon^{\text{LLl}}(\omega, k) = \varepsilon^C(\omega, k),$$

$$1 - \frac{1}{\mu^C(\omega, k)} = \frac{k_0^2}{k^2} (\varepsilon^{\text{LLtr}}(\omega, k) - \varepsilon^{\text{LLl}}(\omega, k)). \quad (16)$$

In spite of the apparent equivalence of these forms, using the LL representation presents some difficulties. In particular, to solve the dispersion equation in the C form, viz.

$$k^2 = k_0^2 \varepsilon^C(\omega, k) \mu^C(\omega, k),$$

we must define the square root as a regular single-valued function. To do this it suffices to take a cut along the negative real axis and calculate the square root for the permittivity and permeability of the medium separately [19]:

$$k = k_0 \sqrt{\varepsilon^C(\omega, k)} \sqrt{\mu^C(\omega, k)}.$$

Defining the square root in this way ensures physically correct solutions both for active and passive media (including Veselago media [38] with  $\varepsilon'$ ,  $\mu'$ , and  $k'$  being negative).

When dealing with the dispersion equation<sup>3</sup>

$$k^2 = k_0^2 \varepsilon^{\text{LLtr}}(\omega, k)$$

obtained within the framework of an LL form, it is necessary each time to redefine the square root

$$k = k_0 \sqrt{\varepsilon^{\text{LLtr}}(\omega, k)}$$

for finding a physically correct solution. To describe active and passive media in which the permittivity and permeability have their real parts positive, we must take a cut along the negative real axis. When the permittivity and permeability of the medium have negative real parts (see Ref. [38]), a cut should be taken along the positive real axis. But this is very inconvenient.

The situation becomes even more troublesome if, following Refs [16, 20], we assume that the C-form permittivity is also a tensor:

$$\varepsilon_{ij}^C(\omega, \mathbf{k}) = \varepsilon^{\text{Ctr}}(\omega, k) \left( \delta_{ij} - \frac{k_i k_j}{k^2} \right) + \varepsilon^{\text{Cl}}(\omega, k) \frac{k_i k_j}{k^2},$$

<sup>3</sup> Relation (16) simplifies to  $\varepsilon^{\text{LLtr}}(\omega, k) = \varepsilon^C(\omega, k) \mu^C(\omega, k)$  for waves propagating in a space free of the external sources.

where  $\varepsilon^{\text{C tr}}(\omega, k)$  and  $\varepsilon^{\text{C l}}(\omega, k)$  are the scalars which generally differ from both  $\varepsilon^{\text{LL tr}}(\omega, k)$  and  $\varepsilon^{\text{LL l}}(\omega, k)$ . It has been hypothesized [16] that the permeability of the medium may also be a tensor with a similar structure, viz.

$$\mu_{ij}^{\text{C}}(\omega, \mathbf{k}) = \mu^{\text{C tr}}(\omega, k) \left( \delta_{ij} - \frac{k_i k_j}{k^2} \right) + \mu^{\text{C l}}(\omega, k) \frac{k_i k_j}{k^2}. \quad (17)$$

Relation (14) under the assumptions (15)–(17) becomes (see also Ref. [37])

$$\varepsilon^{\text{LL l}}(\omega, k) = \varepsilon^{\text{C l}}(\omega, k), \quad (18)$$

$$\varepsilon^{\text{LL tr}}(\omega, k) = \varepsilon^{\text{C tr}}(\omega, k) + \left( \frac{k}{k_0} \right)^2 (1 - \mu^{\text{C tr}}(\omega, k)^{-1}). \quad (19)$$

We see that the assumption of the C-form permittivity (permeability) with a tensor structure implies that Eqn (14) relates two LL parameters and three C parameters, a situation which makes this equation irreversible. Whereas knowing the C-form permittivity (permeability) allows one to reconstruct the LL-form permittivity, the inverse problem of reconstructing the C properties from their LL counterparts cannot be solved. The researchers who take this view believe that the C and LL forms are not equivalent. Some authors [16, 20, 37] show a preference for the LL form and consider the irreversibility of Eqn (14) as a consequence of the lack of uniqueness in expansion (7). In other words, they consider the LL form to be more general.

It is noteworthy that the quantity  $\mu^{\text{C l}}(\omega, k)$  did not enter into relations (18), (19). This is most likely due to the scalar nature of the permeability [20, 37]. Indeed, since

$$\begin{aligned} k_l B_l &= \mu^{\text{C tr}} \left( k_l \delta_{lm} - k_l \frac{k_l k_m}{k^2} \right) H_m + \mu^{\text{C l}} k_l \frac{k_l k_m}{k^2} H_m \\ &= \mu^{\text{C l}} k_m H_m = 0, \end{aligned}$$

it follows that either

$$k_m H_m = 0 \quad (20)$$

or

$$\mu^{\text{C l}} = 0. \quad (21)$$

If equality (20) holds true, then we need only one scalar  $\mu^{\text{C tr}}$  to describe the fields, because

$$B_l = \mu^{\text{C tr}} \left( \delta_{lm} - \frac{k_l k_m}{k^2} \right) H_m + \mu^{\text{C l}} \frac{k_l k_m}{k^2} H_m = \mu^{\text{C tr}} H_l.$$

Equality (21) is identical to the magnetostatic wave excitation condition [21]. It is well known that these waves do not carry an electric field and are described by the reduced system of equations

$$\mathbf{V} \cdot \mathbf{B} = 0, \quad [\mathbf{V} \times \mathbf{H}] = 0.$$

The absence of the electric field implies that  $[\mathbf{V} \times \mathbf{E}] = 0$  and hence that the transverse component of the magnetic field also vanishes. For  $H_{\text{tr}} \neq 0$  this is possible only when  $\mu^{\text{C tr}} = 0$ . In other words, the condition for the excitation of magnetostatic waves entails the equality  $\mu^{\text{C l}} = \mu^{\text{C tr}}$ . It is unlikely that these two quantities are equal only at a fixed frequency and differ at others. The fact that the longitudinal and transverse

components of the permeabilities are equal implies that the C-form permeability is a scalar.

The last remark does not solve the problem concerned, because we still have two LL and three C parameters on our hands.

Note here that the reference to an ambiguous character of the representation (7) [16, 20, 37] is unconvincing because of the conditions for the unique decomposition (7)<sup>4</sup> being presented in Ref. [9]. If the conditions of Ref. [9] are not met, the fields in a homogeneous system turn out not to be invariant with respect to averaging over the volume and hence the use of Maxwell's macroscopic constitutive equations becomes problematic. Thus, when the conditions set in Ref. [9] are fulfilled, the arguments of the authors of Refs [16, 20] fail to explain the irreversible nature of Eqn (14). To elucidate the cause of the irreversibility, it is appropriate now to look at the opinions of a third group of researchers.

This group [22–24] considers the C form to be the most general. Their arguments are as follows. It is well known that the C form of the Maxwell equations is invariant under a Serdyukov–Fedorov transformation [20, 22–24], i.e. one can introduce a pair of fields  $\mathbf{Q}$  and  $\mathbf{F}$  and redefine the fields entering into the Maxwell equations in such a way that these new fields satisfy the same equations:

$$\mathbf{D}' = \mathbf{D} + \text{rot } \mathbf{Q}, \quad \mathbf{H}' = \mathbf{H} + \frac{1}{c} \frac{\partial}{\partial t} \mathbf{Q}, \quad (22)$$

$$\mathbf{B}' = \mathbf{B} + \text{rot } \mathbf{F}, \quad \mathbf{E}' = \mathbf{E} + \frac{1}{c} \frac{\partial}{\partial t} \mathbf{F}. \quad (23)$$

Clearly, the new primed fields are related by novel constitutive equations and satisfy different boundary conditions at the interface [22, 23]. In terms of the Serdyukov–Fedorov transformation, the transition from the C to the LL form is equivalent to setting  $\mathbf{Q} = \mathbf{M}$  and  $\mathbf{F} = 0$ . The vector  $\mathbf{Q}$  is chosen in such a way that  $\mathbf{H}' = \mathbf{B}$  (see Refs [20, 25]). Thus, the LL form is a special case of the C form. As a consequence, only when the C-form boundary conditions are known, the correct LL-form boundary equations can be obtained [22, 23]. It is assumed that the C-form boundary conditions are identical to the ordinary Maxwell boundary conditions (the continuity of the tangential components of the  $\mathbf{E}$  and  $\mathbf{H}$  fields).

It should be emphasized that despite the attempts to prove the equivalence of the two forms by means of redefinitions [22, 23], the assumption made in Refs [20, 22–24] implies that Eqn (14) is irreversible, because the LL form, unlike the C form, is not invariant under the Serdyukov–Fedorov transformation. To see this, note that the Serdyukov–Fedorov transformation in this case is given by

$$\mathbf{D}' = \mathbf{D} + \text{rot } \mathbf{Q}, \quad \mathbf{B}' = \mathbf{B} + \frac{1}{c} \frac{\partial}{\partial t} \mathbf{Q}, \quad (24)$$

$$\mathbf{B}' = \mathbf{B} + \text{rot } \mathbf{F}, \quad \mathbf{E}' = \mathbf{E} + \frac{1}{c} \frac{\partial}{\partial t} \mathbf{F}. \quad (25)$$

This means that we must introduce a relationship between the vectors  $\mathbf{Q}$  and  $\mathbf{F}$ :

$$c \text{rot } \mathbf{F} = \frac{\partial \mathbf{Q}}{\partial t},$$

<sup>4</sup> The conditions imposed in Ref. [9] imply the existence of a ‘dividing’ scale  $l_s$  such that  $\xi \ll l_s \ll \lambda$ , where  $\xi$  is the maximum scale of inhomogeneity.

and not only  $\mathbf{D}$  but also  $\mathbf{E}$  is changed. Whereas, as noted above, the field  $\mathbf{E}$  is assumed to be the same when employing the C and LL forms.

In our view, the fact that the LL-form boundary conditions can be obtained from the C-form boundary conditions and at the same time from a macroscopic theory leading to the LL form<sup>5</sup> makes it possible to obtain the missing relationship between the C and LL parameters — one which makes Eqn (14) reversible.

To illustrate this point, let us consider a homogeneous magnetic material, say, a ferrite. We neglect below the gyrotropy of the material for the sake of clarity.

To obtain the LL form of the constitutive equations, we must incorporate all the currents into the LL polarization:

$$\frac{\partial(\mathbf{D}^{\text{LL}} - \mathbf{E})}{\partial t} = 4\pi\mathbf{j} = 4\pi \frac{\partial\mathbf{P}^{\text{C}}}{\partial t} + 4\pi c \text{rot } \mathbf{M}, \quad (26)$$

or

$$\mathbf{D}^{\text{LL}} = \varepsilon^{\text{C}}(\omega)\mathbf{E} + i \frac{4\pi}{k_0} \text{rot } \mathbf{M} \quad (27)$$

$$= \varepsilon^{\text{C}}(\omega)\mathbf{E} + i\xi(\omega) \text{rot } \mathbf{B} \quad (28)$$

$$= \varepsilon^{\text{C}}(\omega)\mathbf{E} + \frac{\xi(\omega)}{k_0} \text{rot rot } \mathbf{E}, \quad (29)$$

where  $\xi(\omega) = (1 - \mu^{-1})/k_0$ . By applying the Fourier transformation to Eqn (29) we obtain the following constitutive LL parameters:

$$\varepsilon^{\text{LLtr}}(\omega, k) = \varepsilon^{\text{C}}(\omega) + \frac{k^2}{k_0} \xi(\omega), \quad (30)$$

$$\varepsilon^{\text{LLl}}(\omega, k) = \varepsilon^{\text{C}}(\omega). \quad (31)$$

When using the LL form, we are dealing with a formally nonmagnetic material and must therefore apply the Maxwell boundary condition usual for nonmagnetic materials<sup>6</sup>. We will demonstrate this point by taking the constitutive equation in the form (28). Let us rewrite the Ampere–Maxwell equation as follows

$$\text{rot } \mathbf{B} = -ik_0\mathbf{D}^{\text{LL}} = -ik_0[\varepsilon(\omega)\mathbf{E} + i\xi(\omega) \text{rot } \mathbf{B}]. \quad (32)$$

Transposing the term  $k_0\xi(\omega) \text{rot } \mathbf{B}$  to the right-hand side we obtain

$$\text{rot } \mathbf{B} = -\frac{ik_0\varepsilon(\omega)}{1 - k_0\xi(\omega)} \mathbf{E}. \quad (33)$$

Observe that the right-hand side of Eqn (33) does not have singularities at the surface, so that the standard procedure for this situation yields the following boundary condition at the

interface:

$$B_{1t} - B_{2t} = 0. \quad (34)$$

The treatment of the present problem using the C form predicts another form of the boundary condition at the interface. To obtain this condition we follow the standard procedure of taking the surface integral of both sides of Eqn (5) [5]. The surface of integration is bounded by a rectangular contour having its long sides on either side of the boundary. The contracting surface is assumed to be perpendicular to the interface. The surface integral of the curl of any function transforms into a curvilinear integral. After contraction of the contour along its short side, the surface integral containing the electric field vanishes because the electric field has no singularities at the boundary. Finally, we obtain the boundary condition in the form [5, 27]

$$B_{1t} - B_{2t} = -4\pi M_{2t}. \quad (35)$$

This relation is identical to the conventional Maxwell relation  $H_{1t} - H_{2t} = 0$ . The cause of the difference between Eqns (34) and (35) will be discussed below.

The boundary condition (35) is equivalent to the ordinary Maxwell boundary condition in the presence of a surface current — as indeed it must, because, as is known, even the uniform magnetization of a medium produces a surface current (see Fig. 4 and Refs [26, 27]).

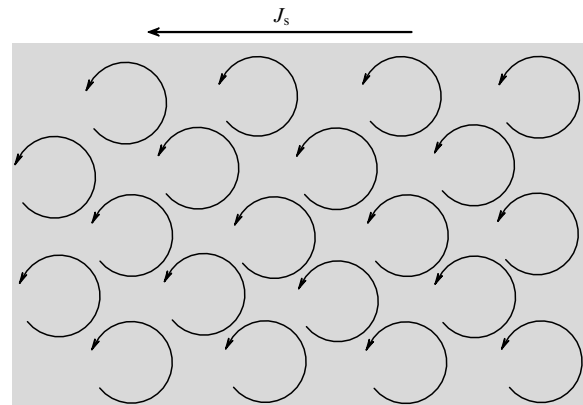


Figure 4. Magnetization of a medium gives rise to a surface current.

Using Eqns (31), (32), the missing condition for the reversibility of Eqn (14) for ferrite is readily shown to be

$$\varepsilon^{\text{LLtr}}(\omega, k) - \varepsilon^{\text{LLl}}(\omega, k) = \frac{k^2}{k_0} \xi(\omega) = \frac{k^2}{k_0^2} [1 - (\mu^{\text{Ctr}})^{-1}]. \quad (36)$$

### 3. Weak spatial dispersion

In the previous section the equivalence of the LL and C forms has been proved for the integral form of the constitutive equations. Actually, the integral form is rarely adapted. Preference is given for the differential form. Using this latter form one should be especially careful to avoid trouble.

The presence of an integral in Eqn (2) suggests that its kernel should decrease with increasing distance  $\mathbf{p} = \mathbf{r}' - \mathbf{r}$ . If the size  $a$  of an inclusion and the average distance  $d$  between

<sup>5</sup> The true boundary conditions are derived by considering the behavior of the fields within a layer near the interface. Clearly, the kernel in Eqn (2) depends not only on the difference between the arguments, but also on each of them separately [11].

<sup>6</sup> When dealing with nonmagnetic media we need to rely only on one field when describing the magnetic part of electromagnetic fields. Unfortunately, this field — the magnetic induction, physically — is usually denoted by the symbol  $H$  and called the magnetic field. This causes no trouble because  $\mu = 1$  and hence  $B = H$ . However, to emphasize the physical meaning of the quantity, we follow Landau and Lifshitz [5] and call it the magnetic induction (thus denoting it by the letter  $B$ ).

inclusions are much less than the wavelength, then so is the kernel radius, and the field under the integral can be expanded in a Taylor series. As a result we arrive at the constitutive equations in the differential form:

$$D_i(\omega, r) = \varepsilon_{ij}^{\text{LL}(0)}(\omega, r)E_j + \varepsilon_{ijl}^{\text{LL}(1)} \frac{\partial E_j}{\partial x_l} + \varepsilon_{ijklm}^{\text{LL}(2)} \frac{\partial^2 E_j}{\partial x_l \partial x_m} + \dots, \quad (37)$$

where the following notation was used:  $\varepsilon_{ijl_1 \dots l_n}^{\text{LL}(n)}(\omega, r) = \int \varepsilon_{ij}(r, \rho - r) \rho_{l_1} \dots \rho_{l_n} d^3 \rho$ .

The incorrect interpretation of this series may be a source of much trouble. When dealing with the spatial dispersion effects one should remember that, as mentioned above, there are at least two geometrical scales involved in the problem: the size of the inclusion (radius of the kernel)  $a$  and the wavelength. For example, the kernel may be of the form  $\varepsilon_{ij}(r, \rho - r) \sim \exp(-\rho/a) f(\rho/\lambda)$ , where the function  $f$  can be expanded in a Laurent series<sup>7</sup>.

Thus, the coefficients of the derivatives entering into Eqn (37) depend on both the scales, but only dependence on  $a$  correspond to the terms appeared in perturbation theory in  $ka$ . The terms with  $\lambda$ -dependent coefficients should be transformed using the Maxwell equation. This point is often ignored and all the coefficients in Eqn (37) are assumed to be powers of  $a$ . As a consequence, the Fourier transform of the series (37) is considered as a series in powers of  $ka$ . Actually, the coefficient  $\varepsilon_{ijl_1 \dots l_n}^{\text{LL}(n)}$  is a heterogeneous polynomial of two variables:

$$\sum_{m=0}^{m=n} c_m^n a^{n-m} \lambda^m,$$

where  $c_m^n$  are the dimensionless coefficients. To obtain a perturbation series, the terms with  $m \neq 0$  should be transformed, namely, the spatial derivatives of the fields must be replaced by temporal ones using the Maxwell equations.

As an illustration, let us return to the derivation of the boundary conditions (34). Expression (29) can be obtained formally from Eqn (37). In this case Eqn (29) takes the form

$$\varepsilon_{ijk}^{\text{LL}(1)} = 0, \quad (38)$$

$$\varepsilon_{ijkl}^{\text{LL}(2)} = \frac{\zeta(\omega)}{k_0} e_{ikm} e_{mlj} \quad (39)$$

(see, for instance, Ref. [28]).

At first sight, the distinction between Eqns (27) and (29) is that we simply rename Eqns (38) and (39). The analysis of a restricted system shows, however, that the problem goes deeper. In particular, this distinction leads to different

<sup>7</sup> Composite materials, which are heterogeneous substances, present multiscale problems, i.e. not only the geometrical sizes involved but also the wavelengths of waves in the constituents are important. The reason is that the response of a composite depends not only on the constituent properties but also on the inclusion shapes. Also, the response of a particle in an alternating field depends strongly on the incident radiation wavelength. An example is the case in which the skin effect on an individual inclusion should be taken into account. Thus, the magnetic polarizability of a conducting ball depends on the ratio between the ball radius and the skin depth:  $a/\delta \sim a\lambda^{3/2}/\sqrt{\sigma/c}$ . The latter quantity is the ‘wavelength’ in the conductor. For instance, at low frequencies  $\alpha' \sim -a^4 \lambda^{-4} (\sigma/\omega)^2 \sim -a^4 \lambda^{-2} (\sigma/c)^2$ . In both cases the powers of  $a$  and  $\lambda$  are different, which is just what the present work declares.

boundary equations. Indeed, if instead of Eqn (27) we use expression (28), which is a consequence of Eqn (29), it can be shown that the jump in the tangential component of magnetic induction at the interface is always zero. The reason is that whereas in the LL form we employ only a part of the series (37), in the C form an exact expression is used.

The most dramatic manifestation of this problem is encountered in chiral media.

#### 4. Chiral (optically active) media

As is well known, chirality represents a first-order effect (in  $ka$ ) [29]. However, retaining only terms with first derivatives in series (37) and in the series obtained from Eqns (12) and (13), we can arrive at different conclusions.

The constitutive equations in the LL form are as follows

$$\mathbf{D}^{\text{LL}} = \varepsilon \mathbf{E} + \gamma \text{rot } \mathbf{E}, \quad (40)$$

where  $\gamma$  is a pseudoscalar [5]. This constitutive equation predicts that the polarization plane rotates as a plane wave propagates. When combined with Maxwell’s boundary conditions, the equation predicts that in the case of a normally incident, linearly polarized plane wave, the reflected wave will be elliptically polarized. Also, the major axis of the polarization ellipse undergoes an azimuthal rotation with respect to the original polarization (the effect of optical activity on reflection). This behavior is a property of nonreciprocal media. However, a chiral system made up of reciprocal elements must be reciprocal, too. It has been suggested [30] that this behavior may relate to the existence of a transition layer near the interphase surface.

Indeed, the boundary acts to violate the translational invariance, and the kernel in Eqn (2) depends not only on the difference of spatial variables but also on where the point of observation is located. The authors of Ref. [30] accounted for this circumstance by introducing an additional term into the constitutive equation:

$$\mathbf{D}^{\text{LL}} = \varepsilon \mathbf{E} + \gamma \text{rot } \mathbf{E} + [\text{grad } \gamma \times \mathbf{E}]. \quad (41)$$

Unfortunately, the mathematics of Ref. [30] is not rigorous enough, and this theory can only be considered as phenomenological. The effect of the additional term leads to the fact that the conditions at the boundary differ from the Maxwell conditions [30]:

$$B_{1t} - B_{2t} = \frac{\gamma}{c} \frac{\partial E_t}{\partial t}. \quad (42)$$

Such a boundary condition changes the sign of the angle through which the axis of the polarization ellipse in the reflected wave rotates. However, this result, although consistent with the experiment presented in Ref. [30]<sup>8</sup>, by no means settles the problem of nonreciprocity of the effect.

To save the theory, the following generalized form of the constitutive equation (42) was proposed [11, 32]:

$$\mathbf{D}^{\text{LL}} = \varepsilon \mathbf{E} + \gamma_1 \text{rot } \mathbf{E} + [\text{grad } \gamma_2 \times \mathbf{E}], \quad (43)$$

<sup>8</sup> Notice that the effects we discuss are extremely small for naturally active materials. In all likelihood, the experiment is being carried out on the limits of accuracy and so cannot be used for testing the hypotheses. While some researchers predict reflection-related optical activity [30], others argue that it is absent [31].

where it is assumed that  $\gamma_1 = 2\gamma_2$ . This relation between  $\gamma_1$  and  $\gamma_2$  is obtained from the principle of symmetry of the coefficients ( $\int E'_i D''_i dv = \int E''_i D'_i dv$ ) [33, 34]. The analysis of the standard procedure of deriving the energy equation [22] suggests that this equation takes the form of a continuity equation only if (1) the expressions for the energy density and the Poynting vector are redefined, and (2) the additional condition  $\gamma_1 = 2\gamma_2$  is imposed. Both Eqn (43) and the C form yield zero reflection-related optical activity when used with Maxwell's boundary conditions.

Although the Landau–Lifshitz and Born–Fedorov (Casimir) approaches have been successfully reconciled, a feeling remains that there is something artificial about the arguments leading to the relation  $\gamma_1 = 2\gamma_2$ . First, this relates to the possibility of extending the above arguments to the C form, thus obtaining the ‘constitutive equations for inhomogeneous media’:

$$\begin{aligned} \mathbf{D}^{\text{BF}} &= \epsilon \mathbf{E} + (\epsilon\beta) \text{rot } \mathbf{E} + [\text{grad } (0.5\epsilon\beta) \times \mathbf{E}], \\ \mathbf{B}^{\text{BF}} &= \mu \mathbf{H} + (\mu\beta) \text{rot } \mathbf{H} + [\text{grad } (0.5\mu\beta) \times \mathbf{H}]. \end{aligned}$$

This provokes novel algebraic exercises but gives no insight into the physical aspects of the problem. What casts further doubt on these arguments is, in the author's view, the following. It is known that the rotation of the plane of polarization on radiation reflection is the property of nonreciprocal media [21]. The reason is the presence of a characteristic vector (the magnetization vector  $\mathbf{M}$  in ferromagnets, and the vector  $\mathbf{L}$  in antiferromagnets) in the problem. The presence of a transition layer also introduces an additional vector into the problem, namely, the gradient of material parameters. In nonactive media such a gradient is a polar vector — in marked contrast to magnetically ordered systems, where the vectors  $\mathbf{M}$  and  $\mathbf{L}$  are axial. As for the gradient of a pseudoscalar, it is an axial vector. On the one hand, the presence of a transition layer could lead to nonreciprocal effects, but then symmetry arguments require changing the sign of this gradient for double-primed fields, i.e. we need to consider a totally different system and hence obtain little information from the relations obtained. On the other hand, a composite made up of reciprocal constituents only must possess the property of reciprocity.

The above discussion shows that the arguments in favor of introducing an additional term  $[\text{grad } \gamma_2 \times \mathbf{E}]$  have the nature of a justification rather than a proof. Because the additional term  $[\text{grad } \gamma_2 \times \mathbf{E}]$  is nonzero only near the boundary, it follows that its introduction is equivalent to introducing an additional surface current and changing the boundary conditions. The introduction of the nonreciprocal boundary condition (42) actually compensates for the nonreciprocal nature of the constitutive equation (40)<sup>9</sup>.

<sup>9</sup> If the constitutive equations are written in the form  $\mathbf{D} = \epsilon \mathbf{E} + (\epsilon\beta_c) \text{rot } \mathbf{E}$ ,  $\mathbf{B} = \mu \mathbf{H} + (\mu\beta_m) \text{rot } \mathbf{H}$  (which includes both the LL and C forms), then the Lorentz lemma reduces to the expression

$$\begin{aligned} \int \left( \frac{4\pi}{c} \mathbf{E}' \mathbf{j}'' - \frac{4\pi}{c} \mathbf{E}'' \mathbf{j}' \right) dv &= ik_0 \mu \beta_m \int (\text{div} [\mathbf{H}'' \times \mathbf{H}']) dv \\ &+ ik_0 \epsilon \beta_c \int (\text{div} [\mathbf{E}' \times \mathbf{E}'']) dv. \end{aligned}$$

The right-hand side of this equation vanishes only if  $\beta_m = \beta_c$ . In the LL form this relation does not hold because  $\beta_m = 0$ , while  $\beta_c = \gamma$  in this formulation. I thank B Z Katsenelenbaum for drawing my attention to this fact (see also Ref. [36]).

In the author's opinion, the trouble with the application of the LL form to bounded media is that some first-order terms (in  $ka$ ) are omitted in writing the constitutive equations. Comparing Eqn (40) with the Born–Fedorov constitutive equations it is seen that one should retain terms with up to at least third derivatives in the expansion in the LL form. The second-order term makes a contribution to the permeability of a medium:  $c_2 \text{rot rot } \mathbf{E} \sim c_2 k_0 \text{rot } \mathbf{B} \sim c_2 (1 - 1/\mu) \text{rot } \mathbf{M}$ . The term with the third derivative is responsible for chirality:  $c_3 \text{rot rot rot } \mathbf{E} \sim c_3 k_0 \text{rot rot } \mathbf{B}$ . According to the arguments put forward at the end of Section 2, the coefficients of these additional terms are proportional to the corresponding powers of  $\lambda$ .

Using the Born–Fedorov equations together with Maxwell's boundary equations does not require any major modifications of the theory (such as the redefinitions of the Poynting vector and energy density, the reformulation of the Lorentz lemma, etc.) and yields a satisfactory agreement with experiment.

## 5. Conclusions

In conclusion, let us examine the physical meaning of the fields appearing in the Maxwell equations. Present-day authors rely almost exclusively on the Rosenfeld postulate. Rosenfeld's proposal [35] was to define the fields  $\mathbf{E}$  and  $\mathbf{B}$  without solving the Maxwell equations and only considering the motion of a charged probe particle. Rosenfeld also assumed that this particle moves under the action of the Lorentz force  $\mathbf{F} = e\mathbf{E} + e[\mathbf{u} \times \mathbf{B}]$ , where  $\mathbf{E}$  and  $\mathbf{B}$  are the fields occurring in the Maxwell equations [10]. This is undoubtedly correct for a particle moving in vacuum. Thus, at least in vacuum we have an independent definition of the fields. Unfortunately, this definition cannot be applied to condensed media. Indeed, any charged particle travelling through a medium experiences the influence of the latter. To describe this influence does not mean just substituting macroscopic fields into the Lorentz force. The particle polarizes the surrounding medium and loses its energy. This gives rise to an additional force, and we need the force's explicit form for applying the Rosenfeld postulate. Unfortunately, this force depends on the form of the constitutive equations. But these latter, because of the ambiguity produced by the Serdyukov–Fedorov transformation, depend on how the fields are defined. Thus we have a vicious circle here: the definition of fields in terms of the Lorentz force is only possible after the fields themselves have been defined.

There is an alternative way to define the fields, which involves the definition of the boundary conditions. Let us forget for a moment that we are dealing with heterogeneous media and recall that the field inside a cavity is  $\mathbf{E}(\mathbf{H})$  if the cavity is elongated along the lines of force. If the cavity is flattened, the field is  $\mathbf{D}(\mathbf{B})$ . This fact is a consequence of Maxwell's boundary conditions. Thus, if we assume that Maxwell's boundary conditions are valid, we have a method by which all the fields can be measured. Application of the Serdyukov–Fedorov transformation leads to a change in the boundary conditions, and, as a consequence, the fields in the cavity are no longer equal to those in the medium.

The cavity method provides a consistent definition of macroscopic fields. Indeed, to solve any electromagnetic problem for bounded bodies requires matching the fields outside and inside a body. Specifying boundary conditions is a necessary and sufficient condition for the solution of this

problem. That the cavity-based definition of fields relies on the same boundary conditions makes these fields measurable, and the solution of the electromagnetic problem is assigned a physical meaning.

It is necessary to note that Maxwell's boundary conditions are not a consequence of the Maxwell equations being written in an integral form. There is no rigorous proof that the Maxwell boundary conditions are *a priori* valid for fields in the Casimir or Landau–Lifshitz representations. The adequacy of Maxwell's boundary conditions can be proved only experimentally or by a microscopic theory describing the transition layer structure. Indeed, using boundary conditions of some kind implies that we have by this very fact defined a method for measuring fields inside the medium and that at the same time have determined the physical meaning of the fields  $\mathbf{E}$  and  $\mathbf{H}$ . The inverse procedure — of first postulating the physical meaning of the fields involved in the equations and then deriving the boundary conditions — is a vague enough and may lead to erroneous conclusions. For example, the assumption that the fields  $\mathbf{E}$  and  $\mathbf{B}$  in the LL form are identical to those in the C form appears unjustified unless the equivalence of the corresponding boundary conditions is proved. When deriving boundary conditions for the C form, we identify the physical meaning of  $\mathbf{H}$  in vacuum and that of  $\mathbf{H}$  in the medium, whereas for the LL form we must recognize that in vacuum  $\mathbf{H} = \mathbf{B}$  and identify the physical meaning of  $\mathbf{H}$  in vacuum with that of  $\mathbf{B} = \mathbf{H}' \neq \mathbf{H}$  in the medium.

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