On the Problem of Constitutive Parameter of Composite Materials

A. P. Vinogradov[1] and I. I. Skidanov[2]

[1] Institute for Theoretical and Applied Electromagnetism, Russian Academy of Sciences Izhorskaya 13/19, 127412, Moscow, Russia Tel: 7 095 484 26 44, Fax: 7 095 484 26 33, E-mail: vinogr@vinogr.msk.ru

[2] Department for Problems in Physics and Energetics, Moscow Physical and Technological Institute, Dolgoprudnyi, Russia, email: ins@aport.ru

Abstract

A comparison analysis of the Landau-Lifshitz and Casimir forms of the Maxwell equations in condensed media is made. It is shown that the Casimir form comprises sufficient information of the system to solve any electromagnetic problem whereas the Landau-Lifshitz form demands an additional constitutive equation for surface current. It is shown that the main difference in these forms is that the Casimir form being free from seeming spatial dispersion gives more adequate description of effects of spatial dispersion.

1. Introduction

The manufacturing of advanced artificial materials (chiral, percolation and etc.) challenges the researcher to develop an adequate description of the phenomena observed in these materials. The new theory must account for multipole interaction and effects of retardation. The theories of this a kind were well developed for serving phenomena in crystal or dilute systems [1], [2]. Unfortunately, they confine themselves to consideration of weak effects and deal in frame of perturbation theory. In frame of these theories one does not bother his head with strong definitions of involved quantities and concepts. Today we cannot permit ourselves to deal in such a manner. Here we review the existing forms of constitutive equations in the light of their predictions.

2. Constitutive Equations in Forms Suggested by Landau–Lifshitz and by Casimir

In the case of spatial dispersion Landau and Lifshitz [3], [4], [5] suggested including all the induced currents into definition of polarization

$$\frac{\partial \vec{P}^{LL}}{\partial t} = \vec{j} \tag{1}$$

avoiding introduction of magnetization. The most general form of linear constitutive equation looks as:

$$D_i^{LL} = \int_0^\infty d\tau \int d^3 r' \varepsilon_{ij}^{LL}\left(\vec{r}, \vec{r}', \tau\right) E_j\left(\vec{r}', t-\tau\right) \tag{2}$$

where $D_i^{LL} = E_i + 4\pi P_i^{LL}$, E_i, and P_i^{LL} are macroscopic values of electrical induction, electrical field and electrical polarization. The frequency domain Maxwell equations together with this constitution equation could be written as:

$$i\omega\overrightarrow{B} = c\left[\nabla\times\overrightarrow{E}\right], \qquad -i\omega\overrightarrow{D}^{LL} = c\left[\nabla\times\overrightarrow{B}\right] \tag{3}$$

$$\left(\nabla\cdot\overrightarrow{B}\right) = 0, \qquad \left(\nabla\cdot\overrightarrow{D}^{LL}\right) = 0, \tag{4}$$

$$D_i^{LL} = \int d^3r'\varepsilon_{ij}^{LL}\left(\overrightarrow{r},\overrightarrow{r}',\omega\right)E_j\left(\overrightarrow{r}',\omega\right), \tag{5}$$

$$\varepsilon_{ij}^{LL}\left(\overrightarrow{r},\overrightarrow{r}',\omega\right) = \int_0^{\infty} d\tau\exp\left(i\omega\tau\right)\varepsilon_{ij}^{LL}\left(\overrightarrow{r},\overrightarrow{r}',\tau\right) \tag{6}$$

The kernel in (5) decreases with increase of the distance $\overrightarrow{\rho} = \overrightarrow{r} - \overrightarrow{r}'$. If both the dimension a of an inclusion and the mean distance d between the inclusions are small in terms of the wavelength λ then the kernel radius in (5) is also small (the case of weak spatial dispersion). In this case we can expand the field under integral, in the Taylor series. The relation (5) can be rewritten as:

$$\begin{aligned} D_i^{LL}\left(\omega,\overrightarrow{r}\right) &= \widehat{\varepsilon}_{ij}^{LL}\left(\omega,\overrightarrow{r}\right)E_j\left(\omega,\overrightarrow{r}\right) = \\ &\left(\begin{array}{c}\varepsilon_{ij}^{LL(0)}\left(\omega,\overrightarrow{r}\right)+\\ \varepsilon_{ijk}^{LL(1)}\left(\omega,\overrightarrow{r}\right)\nabla_k+\varepsilon_{ijkl}^{LL(2)}\left(\omega,\overrightarrow{r}\right)\nabla_k\nabla_l+\ldots\end{array}\right)E_j\left(\omega,\overrightarrow{r}\right)\end{aligned} \tag{7}$$

where $\varepsilon^{LL(n)}$ are frequency dependent tensors. If during homogenization we separate mean current, eddy current, and saddle current we arrive at the Casimir form of constitutive equations based on the following representation of the macroscopic current ([6])

$$\overrightarrow{j} = \frac{\partial\overrightarrow{P^C}}{\partial t} - c\left(\nabla\cdot\frac{\partial\widehat{Q}}{\partial t}\right) + c\left[\nabla\times\overrightarrow{M}\right] \tag{8}$$

here $\overrightarrow{P}$, and $\widehat{Q}$, are the densities of electric dipole and electric quadrupole moments, $\overrightarrow{M}$ is the density of magnetic dipole moment. The next step is introduction of the magnetic field H and magnetic permeability μ^C:

$$\overrightarrow{H} = \overrightarrow{B} - 4\pi\overrightarrow{M}, \qquad B_i = \mu_{ij}^C H_j \tag{9}$$

And redefine electrical displacement [7], [6], [8]:

$$D_i^C = E_i + 4\pi P_i^C - 4\pi\nabla\cdot\widehat{Q} \tag{10}$$

The current representation (9) implies nonlocal relation of the moment densities to macroscopic field which results in nonlocal constitutive equations

$$D_i^C = \int d^3r'\varepsilon_{ij}^C\left(\overrightarrow{r},\overrightarrow{r}',\omega\right)E_j\left(\overrightarrow{r}',\omega\right), \qquad B_i = \int d^3r'\mu_{ij}^C\left(\overrightarrow{r},\overrightarrow{r}',\omega\right)H_j\left(\overrightarrow{r}',\omega\right) \tag{11}$$

Likewise in (7) we can write (11)

$$\begin{aligned} D_i^C\left(\omega,\overrightarrow{r}\right) &= \widehat{\varepsilon}_{ij}^C\left(\omega,\overrightarrow{r}\right)E_j\left(\omega,\overrightarrow{r}\right) = \\ &\left(\begin{array}{c}\varepsilon_{ij}^{C(0)}\left(\omega,\overrightarrow{r}\right)+\\ \varepsilon_{ijk}^{C(1)}\left(\omega,\overrightarrow{r}\right)\nabla_k+\varepsilon_{ijkl}^{C(2)}\left(\omega,\overrightarrow{r}\right)\nabla_k\nabla_l+\ldots\end{array}\right)E_j\left(\omega,\overrightarrow{r}\right)\end{aligned} \tag{12}$$

$$\begin{aligned} B_i\left(\omega,\overrightarrow{r}\right) &= \widehat{\mu}_{ij}^C\left(\omega,\overrightarrow{r}\right)E_j\left(\omega,\overrightarrow{r}\right) = \\ &\left(\begin{array}{c}\mu_{ij}^{C(0)}\left(\omega,\overrightarrow{r}\right)+\\ \mu_{ijk}^{C(1)}\left(\omega,\overrightarrow{r}\right)\nabla_k+\mu_{ijkl}^{C(2)}\left(\omega,\overrightarrow{r}\right)\nabla_k\nabla_l+\ldots\end{array}\right)E_j\left(\omega,\overrightarrow{r}\right)\end{aligned} \tag{13}$$

The Landau-Lifshitz and Casimir constitutive equations must explain and forecast the same phenomena. In other word there should be an equivalence relation between the tensor in (7) and the tensors in (12) and (13). It is suitable to write the relation for spatial Fourier transforms:

$$\varepsilon_{ij}^{LL}\left(\omega,\overrightarrow{k}\right)=\varepsilon_{ij}^{C}\left(\omega,\overrightarrow{k}\right)+\left(\frac{c}{\omega}\right)^{2}\left\{e_{ikl}e_{jnm}k_{k}k_{n}\left[\left(\mu_{lm}^{C}\right)^{-1}-\delta_{lm}\right]\right\} \tag{14}$$

The last relation is often treated as a condition of equivalence of two forms of constitutive equations. Really, one can reconstruct $\varepsilon_{ij}^{LL}\left(\omega,\overrightarrow{k}\right)$ from ε_{ij}^{C} and μ_{lm}^{C}, but it is impossible to solve the inverse problem of reconstruction ε_{ij}^{C} and μ_{lm}^{C} from ε_{ij}^{LL}. To make the latter problem solvable it often suggested that ε_{ij}^{C} and, μ_{lm}^{C} are scalars whereas ε_{ij}^{C} is a tensor. The last assumption seems to be unjustified. Moreover we loose a series of phenomena. For example, the existence of magnetic longitudinal waves can not be described. If ε_{ij}^{C} and μ_{lm}^{C} are tensors they have the form:

$$\begin{aligned}
\varepsilon_{lm}^{C} &= \varepsilon^{Ctr}(\omega,k)\left(\delta_{lm}-\frac{k_l k_m}{k^2}\right)+\varepsilon^{Cl}(\omega,k)\frac{k_l k_m}{k^2}, && (15)\\
\mu_{lm}^{C} &= \mu^{Ctr}(\omega,k)\left(\delta_{lm}-\frac{k_l k_m}{k^2}\right)+\mu^{Cl}(\omega,k)\frac{k_l k_m}{k^2} && (16)
\end{aligned}$$

It is obvious that along with electrical longitudinal waves there may be magnetic longitudinal waves with μ^{Cl}, $\left(\overrightarrow{k}\cdot\overrightarrow{H}\right)\neq 0$. The examples are stratified medium and composite loaded with be-helix structures. The waves existing in both media are of evanescent. They cannot propagate through the infinite system but the may be generated on the boundaries. The irreversibility of (14) and the existence of the magnetic longitudinal waves means that the Casimir constitutive equations comprise more information of the system.

3. The Physical Sense of the Fields Governed by the Material Maxwell Equations and the Boundary Conditions

In order to understand the reason of incompleteness of the Landau-Lifshitz form we have to consider the problem of the boundary conditions. Dealing with bounded body demands a procedure of sewing together the solution of the Maxwell equation outside and inside the body because just on the boundary the Maxwell equations are not valid. If the dispersion equation

$$k^2=\left(\frac{\omega}{c}\right)^2\varepsilon^{Ctr}(\omega,k)\,\mu^{Ctr}(\omega,k)\,,\qquad\left(\overrightarrow{k}\cdot\overrightarrow{H}\right)=0,\qquad\left(\overrightarrow{k}\cdot\overrightarrow{E}\right)=0 \tag{17}$$

has only one solution for k^2 the common Maxwell boundary conditions are enough. These conditions come about from the assumption that we deal with the same fields inside and outside the medium. In other words the physical sense of the fields $\overrightarrow{E}$ and $\overrightarrow{H}$ should be the same in vacuum and inside the medium. Saying about the physical sense of the fields implies that there must be determined a method of measuring these fields. By now almost all authors proceed from the Rosenfeld Ansatz. Rosenfeld [9] suggested determining the fields investigating the motion a small probe particle with charge e. Rosenfeld assumed that this particle moves under the Lorentz force

$$\overrightarrow{F}=e\overrightarrow{E}+e\left[v\times\overrightarrow{B}\right] \tag{18}$$

where $\overrightarrow{E}$ and $\overrightarrow{B}$ are just the fields staying in the Maxwell equations. "The fields are taken to be the primitive fields" [10]. The fields $\overrightarrow{D}$ and $\overrightarrow{H}$ are often considered being of secondary kind or induction fields [10]. Indeed, one can introduce a new field $\overrightarrow{Q}$ and redefine the fields $\overrightarrow{D}$ and $\overrightarrow{H}$ [11].

$$\overrightarrow{D'} = \overrightarrow{D} + \mathrm{curl}\overrightarrow{Q}, \qquad \overrightarrow{H'} = \overrightarrow{H} + \frac{1}{\mathrm{c}}\frac{\partial}{\partial \mathrm{t}}\overrightarrow{Q} \tag{19}$$

The fields $\overrightarrow{D'}$ and also $\overrightarrow{H'}$ satisfy the Maxwell equations. In particular choosing $\overrightarrow{H'} = \overrightarrow{B}$ we can pass from the Casimir to Landau-Lifshitz form [11]. Unfortunately, this scheme ceases being so attracting if we remember that even in absence of the fields a charged particle moving through the matter looses energy polarizing the surrounding medium. There is another way to determine the fields. This way is tightly connected with boundary conditions. Temporarily forgetting that we deal with heterogeneous media let us recall that the field inside the anisotropically shaped cavity is equal to $\overrightarrow{E}$ ($\overrightarrow{H}$) if the cavity is elongated along the force lines and is equal to $\overrightarrow{D}$ ($\overrightarrow{B}$) if the cavity is flattened along the corresponding direction. This property is a consequence of the Maxwell boundary conditions. Thus if we assume that the Maxwell boundary conditions are valid we have the method of measuring all the fields involved in our problem. There is neither freedom nor uncertainty in definition of any field. Introduction of any auxiliary vector field $\overrightarrow{Q}$ results in changing the boundary conditions.

There are some indications that it is the Casimir form that is accompanied with the Maxwell boundary conditions. First of all along the surface separating two media may flow a surface current that is due to difference in eddy currents induced in different media. Another constitutive equation relating this current to fields should be added to. Thus the Landau-Lifshitz form is incomplete while describing edge effects including the evanescent waves. The suggestion that the Casimir form should be supplemented with the Maxwell boundary conditions means that ε_{ij}^{C} and μ_{lm}^{C} comprise information not only of the wavenumber but also of the impedance. This fact accounts for the irreversibility of (14).

4. Chiral (Optically Active) Media

It is worth emphasizing that really the problem exists if the effects of spatial dispersion are important. If it is not the case the situation becomes trivial. Indeed, the Landau-Lifshitz form produces seeming spatial dispersion. The relation (7) reduces in this case to

$$\overrightarrow{D} = \varepsilon(\omega)\overrightarrow{E} + \xi(\omega)\left[\overrightarrow{k} \times \left[\overrightarrow{k} \times \overrightarrow{E}\right]\right] \tag{20}$$

Employing $e_{ijk}k_jE_k = -(\omega/c)B_i$ we arrive at the usual Casimir form with scalar permeability $\mu = 1/(1 + i\omega\xi(\omega)/c)$. This seeming spatial dispersion may be a source of some troubles. Let us consider the phenomenon of chirality. It is well known that the chirality is the effect of first order in (ka), where is a characteristic dimension of the inclusion and k is the wavenumber. Nevertheless truncating the series (7) and (12), (13) at the same order in (ka) may lead to different consequences. As we shall see the reason of the disagreement is neglect of the seeming spatial dispersion. In the Landau-Lifshitz form the constitutive equations looks as

$$\overrightarrow{D^{LL}} = \varepsilon\overrightarrow{E} + \gamma\mathrm{curl}\overrightarrow{E} \tag{21}$$

where γ is a pseudo-scalar. The constitutive equation predicts rotation of the plane of polarization during propagation. Beside this, the theory employed together with the Maxwell boundary conditions predicts that in the case of normally incident, linearly polarized wave the reflected wave is elliptically polarized. Moreover, the main axis of the polarization ellipse is azimuth rotated (effect of optical activity on reflection). Such a behavior is an attribute of non-reciprocal medium. On the other hand the chiral system made of reciprocal elements should be reciprocal. The authors of [12] suppose that this behavior is connected with existence of transition layer near the boundary surface. Indeed the boundary breaks the translation invariance. The kernel in (5) depends not only on difference of spatial variables but also on position of the point of observation. As a consequence there appear an additional term in the constitutive equation:

$$\overrightarrow{D^{LL}} = \varepsilon \overrightarrow{E} + \gamma \mathrm{curl} \overrightarrow{E} + \left[\mathrm{grad}\gamma \times \overrightarrow{E}\right] \tag{22}$$

The boundary conditions obtained by the usual way change too [12]

$$E_{t1} - E_{t2} = 0, \qquad B_{2t} - B_{1t} = \frac{\gamma}{c}\frac{\partial E_t}{\partial t} \tag{23}$$

The angle of main axis rotation changes his sign but the main result remains: the reflected wave is elliptically polarized. In the problem there appear a vector gradγ. The situation seems to remain the situation in ferromagnetic and antiferromagnetic where the rotation of polarization on reflection is a well known effect. Indeed, in ferromagnetic there is the vector of magnetization $\overrightarrow{M}$ and in antiferromagnetic there is the vector $\overrightarrow{L}$ which is equal to the difference of magnetizations of sublattices. The key moment is that in the last cases the vectors are axial whereas gradγ is a polar vector. Thus we cannot anticipate the appearance of nonreciprocity. To rest the theory in [13] the constitutive equation was generalized:

$$\overrightarrow{D^{LL}} = \varepsilon \overrightarrow{E} + \gamma_1 \mathrm{curl} \overrightarrow{E} + \left[\mathrm{grad}\gamma_2 \times \overrightarrow{E}\right] \tag{24}$$

Where it is assumed that $\gamma_1 = 2\gamma_2$. This relation between the quantities were determined from continuity of the Pointing vector and strange assumption that $\left[\overrightarrow{E} \times \partial \overrightarrow{E}/\partial t\right] \neq 0$. This corrects the boundary conditions so that the effect of optical activity on reflection disappears. All these troubles can be avoided if we deal with Casimir (in our case Born-Fedorov) constitutive equations. There is no effect of optical activity on reflection neither in uniform medium nor while taking into account a transition layer [11]. The background of complexity in boundary conditions appears due to incorrect treatment of derivatives in (7). Dealing with effects of spatial dispersion we must remember that there are two scales in the problem. The first one is the inclusion dimension a . The second scale is the wavelength. Thus coefficients staying in (7) in front of derivatives depend on both scales but only those depending on a contribute in the corresponding term of perturbation theory. The part depending on λ should be rewritten employing the Maxwell equation, relating the first derivatives of $\overrightarrow{E}$ to $\overrightarrow{B}$. In so doing we should take into consideration the term with third order derivative. This term produces the term $\gamma \mathrm{curl} \overrightarrow{B}$ in the Born-Fedorov constitutive equations.

5. Conclusion

Thus the Casimir form of the constitutive equations is more complete in comparison with the Landau-Lifshitz form. The Casimir form produces not only wavevector but also the impedance of the material, whereas the Landau-Lifshitz form demands introduction of additional constitutive equation for the surface current [15]. This additional constitutive equation usually appears as modification of the boundary conditions.

Acknowledgement
This work was supported in part by the Russian Foundation for Basic Research, grants No. 00-15-97570 and No. 99-02-16564.

References

[1] L.D. Barron, *Molecular Light Scattering and Pptical Activity*, Cambridge:Cambridge University Press, 1982.
[2] E. B. Graham, J. Pierrus, and R. E. Raab, *J.Phys. B*, vol. 25, pp. 4673-4684, 1992; R.E. Raab and J. H. Cloete, *JEWA*, vol. 8,pp. 1073-1089, 1994; E. B. Graham and R. E. Raab, *J. Opt. Soc. Am.*, vol. 6, pp. 1239-1248, 1996.
[3] L. D. Landau, E. M. Lifshitz and L. P. Pitaevskii, *Electrodynamics of Continuous Media*, 2nd ed., Oxford: Butterworth Heinemann, 1995.
[4] V.M.Agranovich and V.L. Ginzburg, *Spatial Dispersion in Crystal Optics and the Theory of Excitons*, Monographs and Texts in Physics and Astronomy, Vol. XIII. New York: Interscience, 1966.
[5] V.P. Silin and A.A. Rukhadze *Electromagnetic Properties of Plasma and Plasma-Like Media.* Moscow: GosAtomIzdat, 1961 (in Russian).
[6] A. P. Vinogradov and A. V. Aivazian, *Phys. Rev E*, vol. 60, pp. 987-993, 1999.
[7] J. E. Sipe and J. Van Kranendonk, *Phys. Rev. A* , vol. 9, p. 1806, 1974; *Can. J. Phys.*, vol. 53, p. 2095, 1975; G. Russakoff, *Am. J. Phys.*, vol. 38, p. 1188, 1970.
[8] J. D. Jackson *Classical Electrodynamics*, 2nd ed. Singapore: John Wiley, 1990.
[9] L. Rosenfeld, *Theory of Electrons.* Amsterdam: North-Holland Pub. Company, 1951
[10] W. S. Weiglhofer, *J. Phys. A*, vol. 27, pp. L871-874, 1994.
[11] F. I. Fedorov, *Theory of Girotropy.* Minsk: Nauka i Tekhnika, 1976 (in Russian).
[12] A. R. Bungay, Yu. P. Svirko, and N.I. Zheludev, *Phys. Rev.B*, vol. 47, No. 18, pp. 11730-11735, 1993.
[13] Valkov A. Yu., V. P. Romanov, and A.N. Shalaginov, *Opt. i Spectr.*, vol. 69, pp. 635-639, 1990 (in Russian).
[14] B. V. Bokut and A. N. Serdyukov, *Zh. Prikl. Spectr.*, vol. 20, pp. 677-681, 1974; B. V. Bokut, A. N. Serdyukov, and F. I. Fedorov, *Opt. i Spectr.*, vol. 37, pp. 288-293, 1974 (in Russian).
[15] G. A. Smolenskii, R. V. Pisarev, and I. G. Sinii, *Uspekhi Fiz. Nauk*, vol.116, pp. 231-242, 1975 (im Russian).