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Local field effects in anisotropic metamaterials

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Abstract. We consider dielectric and magnetic properties of metamaterials consisting of single particles. Local field effects are taken into account. The so called method of multiscale consideration is realized: the macroscopic quantities of permittivity and permeability are expressed through the microscopic polarizability and magnetization of single particles. We obtain the generalized Clausius-Mossotti relations for the case of magnetic medium and with anisotropic particles in the long wave approximation. In cases of isotropic magnetic and nonmagnetic particles our results coincide with published results of other authors.

1. Introduction
In the frames of macroscopic electrodynamics there are two ways to determine the constitutive equations. The first way is that we consider only three macroscopic fields: \( E, \ D, \ B \). In this case, permittivity \( \varepsilon(\omega) \) describes both electric and magnetic response of medium. This approach suggests the attractive opportunity to take into account the effects of spatial dispersion, see the article of V. M. Agranovich [1]. The second or “symmetric” way is based on the use of four macroscopic quantities: \( E, \ D, \ B, \ H \). We investigate permittivity \( \varepsilon(\omega) \) and permeability \( \mu(\omega) \) by second way, meaning to develop the theory valid for description of metamaterials with the so called local field effects that are due to interaction of single particles the medium consist of. These particles can be atoms, molecules, nanoparticles or macroscopic particles.

2. Permittivity and permeability with local field effects
Let us consider homogenous magnetic medium which consists of \( N \) anisotropic particles. The microscopic field \( E^{\text{mic}} \) in a medium is different from the incident field \( E^0 \) generated by an external source in vacuum. Relations between the external field \( E^0 \), the macroscopic field \( E \), and the local field \( E^{\text{mic}} \) that lead to the Clausius-Mossotti formula for three-dimensional systems are obtained by using a pair distribution function for molecules [2].

Equation for the density of micro-currents in homogenous magnetic medium can be written as:

\[
j^{\text{mic}}_i (\mathbf{r}, \omega) = -i\omega P_i (\mathbf{r}, \omega) + c \left( \text{rot} \mathbf{M} (\mathbf{r}, \omega) \right),
\]

where \( P(\mathbf{r}, \omega) \) and \( \mathbf{M}(\mathbf{r}, \omega) \) are the average density of the electric and magnetic moments, respectively.

The relationship between the average density of the electric moment and microscopic electric field, the average density of the magnetic moment and the microscopic magnetic field given by the expressions:

\[
P_i (\mathbf{r}, \omega) = \sum_n \alpha^e_n (\omega) E^{\text{mic}}_j (\mathbf{R}_n, \omega) \delta (\mathbf{r} - \mathbf{R}_n),
\]

\[
M_j (\mathbf{r}, \omega) = \sum_n \alpha^m_n (\omega) H^{\text{mic}}_j (\mathbf{R}_n, \omega) \delta (\mathbf{r} - \mathbf{R}_n),
\]
where \( \alpha^\varepsilon_\delta(\omega) \), \( \alpha^\mu_\delta(\omega) \) are polarizability tensor and magnetization tensor, respectively. These tensors can be written in the form

\[
\alpha^\varepsilon_\delta(\omega) = \alpha^\varepsilon_{\emptyset}(\omega)(\delta_\varepsilon - e_\varepsilon e_\varepsilon) + \alpha^\varepsilon_{\emptyset}(\omega)e_\varepsilon e_\varepsilon ,
\]

\[
\alpha^\mu_\delta(\omega) = \alpha^\mu_{\emptyset}(\omega)(\delta_\mu - e_\mu e_\mu) + \alpha^\mu_{\emptyset}(\omega)e_\mu e_\mu .
\]

The microscopic field in medium is the sum of the primary field \( E^0(\mathbf{r},\omega) \), generated by external charges, and secondary fields, produced by all particles of the medium. Maxwell’s equations for Fourier transform of the microscopic fields acting on a particle of medium placed at a point \( \mathbf{r} \) give [2]:

\[
E^\text{mic}(\mathbf{r},\omega) = E^0(\mathbf{r},\omega) + \frac{4\pi i}{c} \int d^3 q e^{iq\mathbf{r}} \left[ k^2 E^\text{mic}(\mathbf{q},\omega) - q(\mathbf{q},E^\text{mic}(\mathbf{q},\omega)) \right],
\]

\[
H^\text{mic}(\mathbf{r},\omega) = H^0(\mathbf{r},\omega) + \frac{4\pi i}{c} \int d^3 q e^{iq\mathbf{r}} \left[ q(\mathbf{q},H^\text{mic}(\mathbf{q},\omega)) \right],
\]

where \( k^2 = \frac{\omega^2}{c^2} \).

The effective field acting on a molecule is formed by adding the fields of many molecules lying in some volume of medium with linear dimensions \( L \) that are large by the comparison with intermolecular distance \( n^{-\frac{1}{3}} \), but small by the comparison with wavelength (long-wave limit). When the inequality

\[
n^{-\frac{1}{3}} = L = \frac{c}{\omega}
\]

is valid, the values of the microscopic electric and magnetic fields acting on a molecule are close to its values averaged over the positions of the over molecules, so-called the local fields. The local field is independent of the positions of other molecules, whereas its dependence on the structure of the medium is preserved by the function of distribution \( f(\mathbf{R}_{\emptyset}) \).

Averaging Eqs. (6) and (7) with two-particle function of distribution

\[
dw(\mathbf{R}_{\emptyset}) \equiv \frac{1}{V} \left[ 1 - f(\mathbf{R}_{\emptyset}) \right] d^3 \mathbf{R}_{\emptyset}
\]

(here, \( V \) is the volume of the medium) i.e. over the positions of all particles in respect to the single particle (see [2] and [3]), one can get equations for the local electric and magnetic fields acting on a molecule located at a point with radius-vector \( \mathbf{R}_{\emptyset} \):

\[
E^\text{loc}_{\emptyset}(\mathbf{R}_{\emptyset},\omega) = E^0_{\emptyset}(\mathbf{R}_{\emptyset},\omega) + \frac{n}{2\omega\pi^2} \int d^3 q \frac{k^2 q - q_\varepsilon q_j}{q^2 - k^2} \int d^3 R_{\emptyset} e^{iq\mathbf{R}_{\emptyset}} \times \left( 1 - f(\mathbf{R}_{\emptyset}) \right) \left[ \omega \alpha^\varepsilon_{\emptyset}(\omega) E^\text{loc}_{\emptyset}(\mathbf{R}_{\emptyset} - \mathbf{R}_{\emptyset},\omega) - c \alpha^\mu_{\emptyset}(\omega) \left[ \mathbf{p}, B^\text{loc}_{\emptyset}(\mathbf{R}_{\emptyset} - \mathbf{R}_{\emptyset},\omega) \right] \right] ,
\]

\[
B^\text{loc}_{\emptyset}(\mathbf{R}_{\emptyset},\omega) = B^0(\mathbf{R}_{\emptyset},\omega) + \frac{n}{2\omega\pi^2} \int d^3 q \frac{1}{q^2 - k^2} \int d^3 R_{\emptyset} e^{iq\mathbf{R}_{\emptyset}} \left( 1 - f(\mathbf{R}_{\emptyset}) \right) \times \left[ \omega \alpha^\varepsilon_{\emptyset}(\omega) \left[ \mathbf{q}, E^\text{loc}_{\emptyset}(\mathbf{R}_{\emptyset} - \mathbf{R}_{\emptyset},\omega) \right] - c \alpha^\mu_{\emptyset}(\omega) \left[ \mathbf{q}, B^\text{loc}_{\emptyset}(\mathbf{R}_{\emptyset} - \mathbf{R}_{\emptyset},\omega) \right] \right] ,
\]

where \( R_{\emptyset} = \mathbf{R}_{\emptyset} - \mathbf{R}_{\emptyset} (k = 1,N-1) \) is the distance between molecules \( \emptyset \) and \( n \).

In these equations \( f(\mathbf{R}_{\emptyset}) \) can be found from equation for the radial distributional function, which in turn can be found from independent experiment, e.g., using x-ray diffraction.
On the other hand, averaging equations Eqs. (6) and (7) over the positions of all particles one get equations for usual average macroscopic fields:

\[
E_i(r,\omega) = E_i^0(r,\omega) + \frac{n}{2\omega\pi^2} \int d^2q e^{i\mathbf{q}\cdot\mathbf{r}} \frac{k^2 \delta_{ij} - q_iq_j}{q^2 - k^2} 
\times (\omega \alpha^{\mu}_{\omega}(\omega) E_k^{\text{loc}}(\mathbf{q},\omega) - c a^{\mu}_{\omega}(\omega)[\mathbf{q}, \mathbf{B}^{\text{loc}}(\mathbf{q},\omega)]_z),
\]

(12)

\[
B_i(r,\omega) = B_i^0(r,\omega) + \frac{n}{2\omega\pi^2} \int d^2q e^{i\mathbf{q}\cdot\mathbf{r}} \frac{1}{q^2 - k^2} 
\times (\omega \alpha^{\mu}_{\omega}(\omega) [\mathbf{q}, \mathbf{E}^{\text{loc}}(\mathbf{q},\omega)]_j - c a^{\mu}_{\omega}(\omega) [\mathbf{q}, [\mathbf{q}, \mathbf{B}^{\text{loc}}(\mathbf{q},\omega)]]_j).
\]

(13)

Substituting vector \( \mathbf{R}_i \) of molecule instead vector \( \mathbf{r} \) in Eqs. (12),(13) gives expressions for macroscopic fields at a point \( \mathbf{R}_i \). Combining Eqs. (10) - (13), in the long-wave limit one can obtain relations between the local field, i.e. locally acting fields, and the average macroscopic field in medium:

\[
E_i(r,\omega) - E_i^{\text{loc}}(r,\omega) = -(4\pi n/3) \alpha^{\mu}_{\omega} E_k^{\text{loc}}(r,\omega),
\]

(14)

\[
B_i(r,\omega) - B_i^{\text{loc}}(r,\omega) = (8\pi n/3) a^{\mu}_{\omega} B_k^{\text{loc}}(r,\omega).
\]

(15)

To obtain Eqs. (14), (15) we used properties of the function \( f(R_{nm}) \). On the one hand, the probability of unlimited approachmment between the two molecules is negligibly small, and therefore \( f(R_{nm}) = 1 \) at \( R_{nm} = n^{-1/3} \). On the other hand, there is no correlation between distant molecules, therefore \( f(R_{nm}) = 0 \) at \( R_{nm} \approx n^{-1/3} \) [2].

Polarization of material can be expressed through the macroscopic field and on the other hand is expressed through the local field:

\[
E_i(r,\omega)(\omega) = 4\pi n a^{\mu}_{\omega} E_k^{\text{loc}}(r,\omega),
\]

(16)

\[
\mu^{\mu}_{\omega}(\omega) B_j(r,\omega) = 4\pi n a^{\mu}_{\omega} B_k^{\text{loc}}(r,\omega).
\]

(17)

Eqs. (14), (15) and (16), (17) let us obtain the dependence of the permittivity tensor \( \varepsilon^{\mu}_{\omega}(\omega) \) from the polarizability tensor \( \alpha^{\mu}_{\omega}(\omega) \) and dependence of the permeability tensor \( \mu^{\mu}_{\omega}(\omega) \) from the magnetization tensor \( \alpha^{\mu}_{\omega}(\omega) \):

\[
\varepsilon^{\mu}_{\omega}(\omega) = \varepsilon^+(\omega) (\delta_{ij} - e_i e_j) + \varepsilon^-(\omega) e_i e_j,
\]

(18)

\[
\mu^{\mu}_{\omega}(\omega) = \mu^+(\omega) (\delta_{ij} - e_i e_j) + \mu^-(\omega) e_i e_j,
\]

(19)

where

\[
\varepsilon^+ = \frac{1 + (8\pi/3)n\alpha^{\mu}_{\omega}(\omega)}{1 - (4\pi/3)n\alpha^{\mu}_{\omega}(\omega)},
\]

(20)

\[
\varepsilon^- = \frac{1 + (8\pi/3)n\alpha^{\mu}_{\omega}(\omega)}{1 - (4\pi/3)n\alpha^{\mu}_{\omega}(\omega)},
\]

\[
\mu^+ = \frac{1 + (8\pi/3)n\alpha^{\mu}_{\omega}(\omega)}{1 - (4\pi/3)n\alpha^{\mu}_{\omega}(\omega)},
\]

(21)

\[
\mu^- = \frac{1 + (8\pi/3)n\alpha^{\mu}_{\omega}(\omega)}{1 - (4\pi/3)n\alpha^{\mu}_{\omega}(\omega)}.
\]
In Eqs. (18) - (20) denominators are not vanish because polarizability and magnetization are complex quantities. In particular case of isotropic medium Eqs. (18), (19) give

\[ \varepsilon(\omega) = \frac{1 + (8\pi/3)n\alpha_e(\omega)}{1 - (4\pi/3)n\alpha_e(\omega)}, \]

(22)

\[ \mu(\omega) = \frac{1 + (8\pi/3)n\alpha_m(\omega)}{1 - (4\pi/3)n\alpha_m(\omega)}. \]

(23)

Eqs. (22),(23) coincides with ones obtained in article [4] with the help of another method. For nonmagnetic material our results go to the well-known Clausius-Mossotti relations. The main approximation we use is that the wavelength of the field must exceed considerably the size of the particles. So, we have obtained the generalized Clausius-Mossotti relations for magnetic media in general and metamaterials in particular.

The conditions when denominators in Eqs. (20) - (23) are small implies the effects of difference between local fields and the macroscopic fields. The strong enhancement of local fields means sharp enhancement of all electromagnetic processes and can be of great interest for numerous applications in physics of solid state, spectroscopy, optics, including nonlinear optics [5], and for artificial periodic structures, in particularly, in metamaterials [6].

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