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Generalized Lorentz-Lorenz formulas for microstructured materials

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In this work the homogenization approach recently proposed in [M. Silveirinha, Phys. Rev. B **75**, 115104 (2007)] is applied to derive the analog of the classical Lorentz-Lorenz formula in a generic dielectric crystal, considering the effects of frequency and spatial dispersion. In particular, we obtain a rigorous formula for the interaction constants that relate the local fields with the macroscopic fields and the induced dipole moments, when the response of the particles is described by only the dipole-type terms. To illustrate the application of the results, we homogenize a material formed by plasmonic spheres, demonstrating that the spatial dispersion effects are of crucial importance near the plasmonic resonance.

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I. INTRODUCTION

The use of homogenization methods to characterize the interaction of electromagnetic fields with matter has a long history. An interesting review of the pioneering works of Lorentz, Planck, Ewald, and Oseen, can be read in Ref. 1. Lorentz was the first to recognize that to properly describe molecular optics it was necessary to incorporate atomic concepts into Maxwell's equations, and take into account the electric vibrations of the particles. These ideas became known as "the Lorentz program." Lorentz obtained a relation between the dielectric constant and the density of the material at optical frequencies, and established the foundations of macroscopic electromagnetism. More recently, the theory was further developed by studies that clarified averaging procedures,[2] and took into account the resonant interaction of electromagnetic radiation with dielectric crystals coupled via retarded dipole fields.[3] Classical molecular optics was also extended to optically active media, and to spatially dispersive media.[4,5]

In recent years there has been a renewed interest in homogenization methods due to their widespread use in the field of microstructured materials (metamaterials). These materials are formed by properly shaped dielectric or metallic inclusions, designed to obtain a desired effective response of the material. It has been demonstrated that the metamaterial concept may enable the synthesis of materials with unconventional properties, such as negative index of refraction,[6] materials with permittivity near zero,[7] or materials with extreme anisotropy.[8,9] An important feature of such novel materials is that typically the size of the lattice constant "a" is only moderately smaller than the wavelength of light, typically 5–10 times. This contrasts markedly with propagation of radiation in matter where the ratio λ/a is several orders of magnitude larger than that value, even at optical frequencies. Due to this reason, new phenomena may emerge in microstructured materials.[10] In particular, the role of spatial dispersion in microstructured materials has been underlined by recent works.[11–17] The description of these effects using homogenization theory requires the use of sophisticated methods.

In a recent work,[18] we have introduced a completely general method to compute the dielectric function of an arbitrary nonmagnetic periodic material. The formalism assumes that the microstructured material can be described by a nonlocal dielectric function $\bar{\bar{\varepsilon}}_{\text{eff}}(\omega,\mathbf{k})$. The dielectric function is obtained by exciting the periodic material with a spatially uniform density of current $\mathbf{J}_e$. By solving the microscopic Maxwell equations, one can compute the microscopic fields, and afterwards the induced generalized polarization vector and the dielectric function. The analysis presented in Ref. 18 is mainly focused on the calculation of the dielectric function of a completely generic material using numerical methods.

The objective of this work is to apply the homogenization formalism proposed in Ref. 18 to derive a generalization of the Lorentz-Lorenz formula.[19] This classical formula relates the local electromagnetic fields with the macroscopic fields and the polarization vector. Here, it will be proven that it is possible to generalize such a formula to a completely generic dielectric crystal, taking into account the effects of frequency and spatial dispersion. We will analyze with special attention the case where the interaction between the inclusions can be described by only the dipolar terms of the radiated field. In particular, it will be demonstrated that the spatial dispersion effects may be of crucial importance near a resonance of the inclusions, and may originate anomalous phenomena such as negative refraction, consistently with the results of Ref. 20.

This paper is organized as follows. In Sec. II the generalized Lorentz-Lorenz formulas are derived. In Sec. III the new Lorentz-Lorenz formulas are used to calculate the dielectric function of a composite material. In Sec. IV we apply the derived results to homogenize a simple cubic lattice of plasmonic spheres. Finally, in Sec. V the conclusions are drawn. In this work, it is assumed that the fields are monochromatic with time dependence of the form $e^{j\omega t}$.

II. GENERALIZED LORENTZ-LORENZ FORMULAS

Here, we use the methodology proposed in Ref. 18 to generalize the classical Lorentz-Lorenz formula to a generic microstructured material. First, we will derive a result that describes the general relation between the induced microscopic current and the local fields, and then we will particularize the formulas for a case in which only the effect of the dipolar-type terms is considered.

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It is assumed that the medium is a three-dimensional dielectric crystal. The medium is invariant to translations along the primitive vectors $\mathbf{a}_1$, $\mathbf{a}_2$, and $\mathbf{a}_3$ of the crystal. For convenience, the reciprocal lattice vectors $\mathbf{b}_1$, $\mathbf{b}_2$, and $\mathbf{b}_3$, defined by the relations $\mathbf{a}_n\mathbf{b}_m=2\pi\delta_{n,m}$, are also introduced. As in our previous work,[18] the inclusions are either dielectric or metallic, with the magnetic permeability equal to μ_0 and relative permittivity $\varepsilon_r(\mathbf{r})$. Since the medium is periodic, the permittivity verifies $\varepsilon_r(\mathbf{r}+\mathbf{r}_\mathbf{I})=\varepsilon_r(\mathbf{r})$, where $\mathbf{r}_\mathbf{I}=i_1\mathbf{a}_1+i_2\mathbf{a}_2+i_3\mathbf{a}_3$ is a lattice point and $\mathbf{I}=(i_1,i_2,i_3)$ is a multi-index of integers.

A. Overview of the homogenization method

The method described in Ref. 18 allows one to homogenize a generic dielectric crystal by means of a dielectric function of the form $\bar{\bar{\varepsilon}}_{\text{eff}}(\omega,\mathbf{k})$, where ω is the angular frequency, and $\mathbf{k}=(k_x,k_y,k_z)$ is the wave vector. As explained in Ref. 18, the dependence of the dielectric function on the wave vector is a consequence of spatial dispersion effects. For a given $(\omega,\mathbf{k})$, the dielectric function can be obtained by solving the microscopic Maxwell equations under the excitation of an external distribution of electric current as follows:

$$\nabla\times\mathbf{E}=-j\omega\mathbf{B},$$

$$\nabla\times\frac{\mathbf{B}}{\mu_0}=j\omega\mathbf{P}_{e,\text{av}}e^{-j\mathbf{k}\cdot\mathbf{r}}+\mathbf{J}_d+j\omega\varepsilon_0\mathbf{E}. \tag{1}$$

In the above, $\mathbf{E}$ and $\mathbf{B}$ are the microscopic electric and induction fields, respectively, $\mathbf{J}_d$ is the microscopic current induced in the material, and the term $\mathbf{P}_{e,\text{av}}e^{-j\mathbf{k}\cdot\mathbf{r}}$ corresponds to the applied external current, where $\mathbf{P}_{e,\text{av}}$ is a constant vector, independent of $\mathbf{r}$, designated by the "applied polarization vector." In a dielectric crystal, $\mathbf{J}_d$ is given by $\mathbf{J}_d=j\omega\varepsilon_0(\varepsilon_r-1)\mathbf{E}$, where ε_r is the periodic permittivity of the material.

The macroscopic electric field $\mathbf{E}_{\text{av}}$ is defined by

$$\mathbf{E}_{\text{av}}=\frac{1}{V_{\text{cell}}}\int_\Omega\mathbf{E}(\mathbf{r})e^{+j\mathbf{k}\cdot\mathbf{r}}d^3\mathbf{r}, \tag{2}$$

where Ω is the unit cell of the material and $V_{\text{cell}}=|\mathbf{a}_1\cdot(\mathbf{a}_2\times\mathbf{a}_3)|$ is the volume of Ω. The macroscopic induction field $\mathbf{B}_{\text{av}}$ is defined similarly. The generalized polarization vector $\mathbf{P}_{g,\text{av}}$ is given by

$$\mathbf{P}_{g,\text{av}}=\frac{1}{V_{\text{cell}}j\omega}\int_\Omega\mathbf{J}_d(\mathbf{r})e^{+j\mathbf{k}\cdot\mathbf{r}}d^3\mathbf{r}. \tag{3}$$

The vector $\mathbf{P}_{g,\text{av}}$ includes the contribution of both the electric and magnetic dipolar response, and in addition of all other higher order multipoles.

The macroscopic fields satisfy

$$-\mathbf{k}\times\mathbf{E}_{\text{av}}+\omega\mathbf{B}_{\text{av}}=0,$$

$$\omega\mathbf{D}_{g,\text{av}}+\mathbf{k}\times\frac{\mathbf{B}_{\text{av}}}{\mu_0}=-\omega\mathbf{P}_{e,\text{av}}, \tag{4}$$

where the electric displacement is related to the average electric field through the constitutive relation,

$$\mathbf{D}_{g,\text{av}}\equiv\varepsilon_0\mathbf{E}_{\text{av}}+\mathbf{P}_{g,\text{av}}=\bar{\bar{\varepsilon}}_{\text{eff}}(\omega,\mathbf{k})\cdot\mathbf{E}_{\text{av}}. \tag{5}$$

The procedure to obtain the dielectric function for a given $(\omega,\mathbf{k})$, consists of using equations (2) and (3) to calculate the macroscopic electric field $\mathbf{E}_{\text{av}}$ and the generalized polarization vector $\mathbf{P}_{g,\text{av}}$ from the microscopic fields. The dielectric function is then defined so that it is consistent with the constitutive relation (5).

An important result obtained in Ref. 18 is that, for the considered external source, the microscopic fields are completely determined by the value of the macroscopic field $\mathbf{E}_{\text{av}}$. In particular, it was demonstrated that the microscopic electric field has the integral representation,

$$\mathbf{E}(\mathbf{r})=\mathbf{E}_{\text{av}}e^{-j\mathbf{k}\cdot\mathbf{r}}+\int_D\bar{\bar{\mathbf{G}}}_{p0}(\mathbf{r}|\mathbf{r}')\cdot(-j\omega\mu_0)\mathbf{J}_d d^3\mathbf{r}', \tag{6}$$

where D represents the dielectric region in the unit cell, and $\bar{\bar{\mathbf{G}}}_{p0}$ is a suitable Green function defined below. As shown in Ref. 18 this representation can be used to formulate an integral equation, whose solution yields the induced microscopic current $\mathbf{J}_d$ associated with a given $(\omega,\mathbf{k})$ and $\mathbf{E}_{\text{av}}$. In this work, we will use the integral representation (6) to calculate the local fields that act on an individual dielectric inclusion.

The Green function $\bar{\bar{\mathbf{G}}}_{p0}$ is given by $\bar{\bar{\mathbf{G}}}_{p0}(\mathbf{r}|\mathbf{r}')=\left(\bar{\bar{\mathbf{I}}}+\frac{c^2}{\omega^2}\nabla\nabla\right)\Phi_{p0}(\mathbf{r}-\mathbf{r}')$, where the scalar potential Φ_{p0} is defined by

$$\Phi_{p0}(\mathbf{r},\omega,\mathbf{k})=\Phi_p(\mathbf{r},\omega,\mathbf{k})-\Phi_{\text{av}}(\omega,\mathbf{k})e^{-j\mathbf{k}\cdot\mathbf{r}}, \tag{7}$$

with

$$\Phi_{\text{av}}(\omega,\mathbf{k})=\frac{1}{V_{\text{cell}}}\frac{1}{k^2-(\omega/c)^2}, \tag{8}$$

where $c=1/\sqrt{\varepsilon_0\mu_0}$ is the speed of light in vacuum (the host medium). We also introduced the lattice Green function Φ_p, which is given by the solution of the equation,[18,21,22]

$$\nabla^2\Phi_p+\left(\frac{\omega}{c}\right)^2\Phi_p=-\sum_{\mathbf{I}}\delta(\mathbf{r}-\mathbf{r}_\mathbf{I})e^{-j\mathbf{k}\cdot\mathbf{r}}, \tag{9}$$

where δ represents Dirac's distribution.

B. Local fields in a generic dielectric crystal

The local electromagnetic field that acts on the inclusion in the unit cell is the superimposition of the fields radiated by the other particles and of the external field. At the end of the 19th century Lorentz and Lorenz proved independently that for a simple cubic lattice the local electric field is related to the macroscopic field through the formula $\mathbf{E}_{\text{loc}}=\mathbf{E}_{\text{av}}+\mathbf{p}_e/(3\varepsilon_0V_{\text{cell}})$, where $\mathbf{p}_e$ is the electric dipole moment of the particles. Such a formula is only valid in the static limit and for a lattice of electric dipoles. In this section, we generalize the classical Lorentz-Lorenz formula to a completely arbitrary dielectric crystal, taking into account both frequency and spatial dispersion effects.

Such a result can be obtained in a straightforward manner using the integral representation (6). To this end, we introduce the free-space Green dyadic $\bar{\bar{\mathbf{G}}}_f(\mathbf{r}|\mathbf{r}')=\left(\bar{\bar{\mathbf{I}}}+\frac{c^2}{\omega^2}\nabla\nabla\right)\Phi_f(\mathbf{r}$

$-\mathbf{r}'$), where $\Phi_f(\mathbf{r},\omega)=e^{-jr\omega/c}/4\pi r$ is the scalar potential due to a single point source with Sommerfeld radiation conditions at infinity. It is simple to verify that Eq. (6) can be rewritten as

$$\mathbf{E}(\mathbf{r})=\mathbf{E}_{\text{loc}}(\mathbf{r})+\int_D \overline{\overline{\mathbf{G}}}_f(\mathbf{r}-\mathbf{r}')\,.\,(-j\omega\mu_0\mathbf{J}_d)d^3\mathbf{r}', \quad (10)$$

where $\mathbf{E}_{\text{loc}}$ is given by

$$\mathbf{E}_{\text{loc}}(\mathbf{r})=\mathbf{E}_{\text{av}}e^{-j\mathbf{k}\cdot\mathbf{r}}+\int_D \overline{\overline{\mathbf{C}}}_{\text{int}}(\mathbf{r}-\mathbf{r}';\omega,\mathbf{k})\cdot\frac{\mathbf{J}_d}{j\omega\varepsilon_0}d^3\mathbf{r}', \quad (11)$$

and the *interaction* dyadic is defined by

$$\overline{\overline{\mathbf{C}}}_{\text{int}}(\mathbf{r}-\mathbf{r}';\omega,\mathbf{k})=\left(\frac{\omega}{c}\right)^2[\overline{\overline{\mathbf{G}}}_{p0}(\mathbf{r}|\mathbf{r}')-\overline{\overline{\mathbf{G}}}_f(\mathbf{r}|\mathbf{r}')]. \quad (12)$$

It is evident that the second term in the right-hand side of formula (10) is the electric field radiated by the inclusion in the unit cell. Hence, $\mathbf{E}_{\text{loc}}$ can be regarded as the "local electric field," as it is suggested by the notation.

Similarly, using the fact that $\mathbf{B}=-\nabla\times\mathbf{E}/j\omega$ and Eq. (4), it is found that the local induction field is given by

$$\mathbf{B}_{\text{loc}}(\mathbf{r})=\mathbf{B}_{\text{av}}e^{-j\mathbf{k}\cdot\mathbf{r}}-\frac{1}{j\omega}\int_D \nabla\times\overline{\overline{\mathbf{C}}}_{\text{int}}(\mathbf{r}-\mathbf{r}';\omega,\mathbf{k})\cdot\frac{\mathbf{J}_d}{j\omega\varepsilon_0}d^3\mathbf{r}'. \quad (13)$$

Equations (11) and (13) relate the local microscopic fields with the macroscopic fields, and with the induced microscopic currents. These formulas can be regarded as a generalization of the classical Lorentz-Lorenz formula to a completely arbitrary nonmagnetic dielectric crystal. To give further insights into these formulas in the next section, we will consider the case in which the fields scattered by the inclusions have essentially a dipole-type character.

For future reference, we note that the interaction dyadic $\overline{\overline{\mathbf{C}}}_{\text{int}}$ can be rewritten as

$$\overline{\overline{\mathbf{C}}}_{\text{int}}(\mathbf{r}-\mathbf{r}';\omega,\mathbf{k})=\left[\left(\frac{\omega}{c}\right)^2\overline{\overline{\mathbf{I}}}+\nabla\nabla\right]\Phi_{\text{reg}}, \quad (14)$$

where Φ_{reg} is the scalar potential defined by

$$\Phi_{\text{reg}}(\mathbf{r},\omega,\mathbf{k})=\Phi_p(\mathbf{r},\omega,\mathbf{k})-\Phi_f(\mathbf{r},\omega)-\Phi_{\text{av}}(\omega,\mathbf{k})e^{-j\mathbf{k}\cdot\mathbf{r}}. \quad (15)$$

From the definition, it is clear that Φ_{reg} has no singularities at the origin $\mathbf{r}=\mathbf{0}$ of the space domain. On the other hand, from the spectral representation of the lattice Green function,[18,21] it can also be verified that Φ_{reg} has no singularities at the origin $\mathbf{k}=\mathbf{0}$ of the spectral domain. Thus, Φ_{reg} is basically a lattice Green function with singularities extracted in both the space and spectral domains. From Eq. (9) it is immediate that for $\mathbf{r}$ in the unit cell,

$$\nabla^2\Phi_{\text{reg}}+\left(\frac{\omega}{c}\right)^2\Phi_{\text{reg}}=\frac{1}{V_{\text{cell}}}e^{-j\mathbf{k}\cdot\mathbf{r}}, \quad \mathbf{r}\in\Omega. \quad (16)$$

Using this formula and Eq. (14) it can be verified that for $\mathbf{r}\in\Omega$ the interaction dyadic satisfies the equation

$$\nabla\times\nabla\times\overline{\overline{\mathbf{C}}}_{\text{int}}-\left(\frac{\omega}{c}\right)^2\overline{\overline{\mathbf{C}}}_{\text{int}}=-\left(\frac{\omega}{c}\right)^2\overline{\overline{\mathbf{I}}}\frac{1}{V_{\text{cell}}}e^{-j\mathbf{k}\cdot\mathbf{r}}. \quad (17)$$

From Eq. (14) it can also be confirmed that $\overline{\overline{\mathbf{C}}}_{\text{int}}$ has the symmetry properties,

$$\overline{\overline{\mathbf{C}}}_{\text{int}}=\overline{\overline{\mathbf{C}}}_{\text{int}}^{t},$$

$$\nabla\times\overline{\overline{\mathbf{C}}}_{\text{int}}=-(\nabla\times\overline{\overline{\mathbf{C}}}_{\text{int}})^t, \quad (18)$$

where the superscript "t" represents the transpose dyadic.

C. Lattice of dipoles

To study the implications of the generalized Lorentz-Lorenz formulas, next it is assumed that the interaction between the particles is accurately described by only the dipolar terms, and that the rest of the multipole moments are negligible. In this way, considering that the dielectric inclusion in the unit cell is centered at the origin, it follows that,

$$\int_D x_i^n\mathbf{J}_d(\mathbf{r})d^3\mathbf{r}\approx 0 \quad (19)$$

for $n\geq 2$ and $i=1,2,3$ (x_i is the projection of $\mathbf{r}$ into a generic coordinate axis), and that

$$\int_D(\mathbf{r}\mathbf{J}_d+\mathbf{J}_d\mathbf{r})d^3\mathbf{r}\approx 0. \quad (20)$$

The latter condition is equivalent to impose that the quadrupole moment vanishes.[19] The fields radiated by a given inclusion can thus be characterized uniquely by the electric dipole moment $\mathbf{p}_e$, and by the magnetic dipole moment $\mathbf{p}_m$, which are defined by

$$\mathbf{p}_e=\frac{1}{j\omega}\int_D\mathbf{J}_d d^3\mathbf{r}', \quad (21)$$

$$\frac{\mathbf{p}_m}{\mu_0}=\frac{1}{2}\int_D\mathbf{r}'\times\mathbf{J}_d d^3\mathbf{r}'. \quad (22)$$

As is well known, Eq. (20) implies that

$$\int_D\mathbf{r}'\mathbf{J}_d d^3\mathbf{r}'=-\overline{\overline{\mathbf{I}}}\times\frac{\mathbf{p}_m}{\mu_0}. \quad (23)$$

Since the set of all polynomials is complete, it can be verified that the unique distribution of current that satisfies Eqs. (19), (21), and (23) simultaneously is such that

$$\mathbf{J}_d(\mathbf{r})\approx j\omega\mathbf{p}_e\delta(\mathbf{r})+\nabla\times\left\{\frac{\mathbf{p}_m}{\mu_0}\delta(\mathbf{r})\right\}, \quad \mathbf{r}\in\Omega. \quad (24)$$

Thus, within the considered hypotheses, the microscopic current induced at the particle in the unit cell can be modeled as a superimposition of δ-function-type distributions. Next, we evaluate the local fields at the origin for this current distribution.

Substituting the microscopic current $\mathbf{J}_d$ into Eqs. (11) and (13), it is simple to verify that for $\mathbf{r}=0$,

$$\mathbf{E}_{\text{loc}} = \mathbf{E}_{\text{av}} + \bar{\bar{\mathbf{C}}}_{\text{int}} \cdot \frac{\mathbf{p}_e}{\varepsilon_0} - \frac{c^2}{j\omega}\mathbf{p}_m \cdot \nabla \times \bar{\bar{\mathbf{C}}}_{\text{int}},$$

$$\mathbf{B}_{\text{loc}} = \mathbf{B}_{\text{av}} - \frac{1}{j\omega}\nabla \times \bar{\bar{\mathbf{C}}}_{\text{int}} \cdot \frac{\mathbf{p}_e}{\varepsilon_0} + \frac{c^2}{\omega^2}\mathbf{p}_m \cdot \nabla \times \nabla \times \bar{\bar{\mathbf{C}}}_{\text{int}}, \tag{25}$$

where all the dyadics are evaluated at the origin. To obtain the above relations we used the symmetry properties (18) and the auxiliary relation

$$\int \bar{\bar{\mathbf{A}}}(\mathbf{r}-\mathbf{r}') \cdot \nabla' \times (\mathbf{p}_m \delta(\mathbf{r}'))d^3\mathbf{r}' = -\mathbf{p}_m \cdot \nabla \times \bar{\bar{\mathbf{A}}}^t(\mathbf{r}), \tag{26}$$

which is valid for a generic dyadic $\bar{\bar{\mathbf{A}}}(\mathbf{r}-\mathbf{r}')$.

To further simplify the local fields, we use Eq. (17) and the symmetry relations (18). It is readily found that

$$\mathbf{E}_{\text{loc}} = \mathbf{E}_{\text{av}} + \bar{\bar{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k}) \cdot \frac{\mathbf{p}_e}{\varepsilon_0} + \bar{\bar{\mathbf{C}}}_{e,m}(\omega,\mathbf{k}) \cdot c\mathbf{p}_m,$$

$$\frac{\mathbf{B}_{\text{loc}}}{\mu_0} = \mathbf{H}_{\text{av}} - \bar{\bar{\mathbf{C}}}_{e,m}(\omega,\mathbf{k}) \cdot c\mathbf{p}_e + \bar{\bar{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k}) \cdot \frac{\mathbf{p}_m}{\mu_0}, \tag{27}$$

where $\bar{\bar{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k})$ is the interaction dyadic evaluated at the origin, and the dyadic $\bar{\bar{\mathbf{C}}}_{e,m}$ is defined by

$$\bar{\bar{\mathbf{C}}}_{e,m}(\omega,\mathbf{k}) = \frac{c}{j\omega}\nabla \times \bar{\bar{\mathbf{C}}}_{\text{int}}\big|_{\mathbf{r}=\mathbf{0}} = -j\frac{\omega}{c}\nabla\Phi_{\text{reg}}\big|_{\mathbf{r}=\mathbf{0}} \times \bar{\bar{\mathbf{I}}}. \tag{28}$$

The dyadic $\bar{\bar{\mathbf{C}}}_{e,m}$ describes the interaction between dipoles of a different type, which may occur due to the structural properties of the lattice.

In Eq. (27), we introduced the macroscopic magnetic field $\mathbf{H}_{\text{av}}$ consistently with the classical definition as follows:

$$\mathbf{H}_{\text{av}} = \frac{\mathbf{B}_{\text{av}}}{\mu_0} - \frac{1}{V_{\text{cell}}}\frac{\mathbf{p}_m}{\mu_0}. \tag{29}$$

Equations (27) demonstrate that within the dipolar approximation, the local fields are written uniquely in terms of the macroscopic fields and of the induced dipole moments. Notice that the local fields depend explicitly on the wave vector $\mathbf{k}$, i.e., on the phase shift of the fields from cell to cell. This effect, which emerges due to the noncontinuous (discrete) nature of the material, is the root of possible spatial dispersion effects, as will be discussed later. In order to establish a rigorous connection between Eq. (27) and the classical Lorentz-Lorenz formula, in the next subsection the interaction dyadics are further characterized.

D. Interaction dyadics

Provided the lattice has enough symmetries, it is straightforward to obtain a closed form expression for $\bar{\bar{\mathbf{C}}}_{\text{int}}$ in the static limit $\omega=0$ and $\mathbf{k}=\mathbf{0}$. In fact, consider that the lattice is simple cubic. It is clear that by symmetry and for $\mathbf{k}=\mathbf{0}$ we have $\frac{\partial^2\Phi_{\text{reg}}}{\partial x^2}=\frac{\partial^2\Phi_{\text{reg}}}{\partial y^2}=\frac{\partial^2\Phi_{\text{reg}}}{\partial z^2}$ at the origin $\mathbf{r}=\mathbf{0}$. Similarly, by symmetry arguments it can be verified that the crossed derivatives, $\frac{\partial^2\Phi_{\text{reg}}}{\partial x \partial y}=0$, etc., vanish at the origin. On the other hand, from Eq. (16), it follows that in the static limit $\nabla^2\Phi_{\text{reg}}=\frac{1}{V_{\text{cell}}}$. This implies that $\frac{\partial^2\Phi_{\text{reg}}}{\partial x^2}=\frac{\partial^2\Phi_{\text{reg}}}{\partial y^2}=\frac{\partial^2\Phi_{\text{reg}}}{\partial z^2}=\frac{1}{3}\frac{1}{V_{\text{cell}}}$ at the origin. Thus, using Eq. (14) it is found that in the static limit the interaction dyadic for a simple cubic lattice is

$$\bar{\bar{\mathbf{C}}}_{\text{int}}(\omega=0,\mathbf{k}=\mathbf{0}) = \frac{1}{3V_{\text{cell}}}\bar{\bar{\mathbf{I}}} \quad \text{(s.c. lattice)}. \tag{30}$$

Substituting this result into Eqs. (27), we obtain the classical Lorentz-Lorenz formula for an isotropic material formed by uniquely electric dipoles.[19] This confirms that Eqs. (27) are indeed generalizations of the classic Lorentz-Lorenz formula. Unlike the classical formula, the new formulas are not limited to the quasistatic regime.

It is relevant to mention that for $\omega=0$ and $\mathbf{k}=\mathbf{0}$ (static case) and for a generic lattice, the interaction dyadic defined in this work is consistent with the result presented in Ref. 23. In the general case, it is necessary to numerically evaluate Eq. (14) to calculate $\bar{\bar{\mathbf{C}}}_{\text{int}}$. This can be done very efficiently using the representations of the lattice Green function proposed in Refs. 21 and 22. For example, from the results of Ref. 21 it can be verified that Φ_{reg} has the following mixed domain representation with Gaussian convergence:

$$\begin{aligned}\Phi_{\text{reg}}(\mathbf{r},\omega,\mathbf{k}) = {} & \frac{1}{4\pi}\frac{j\sin(\beta r)}{r} + \frac{1}{4\pi}\frac{\cos(\beta r)}{r}[\text{erfc}(Er)-1] \\ & + \sum_{\mathbf{I}\neq 0}\frac{1}{4\pi}\frac{\cos(\beta|\mathbf{r}-\mathbf{r}_{\mathbf{I}}|)}{|\mathbf{r}-\mathbf{r}_{\mathbf{I}}|}\text{erfc}(E|\mathbf{r}-\mathbf{r}_{\mathbf{I}}|)e^{-j\mathbf{k}\cdot\mathbf{r}_{\mathbf{I}}} \\ & + \frac{1}{V_{\text{cell}}}\frac{e^{-j\mathbf{k}\cdot\mathbf{r}}}{2k}\sum_{\pm}\frac{e^{-(k\pm\beta)^2/4E^2}-1}{k\pm\beta} \\ & + \frac{1}{V_{\text{cell}}}\sum_{\mathbf{J}\neq\mathbf{0}}\frac{1}{2|\mathbf{k}_{\mathbf{J}}|}\sum_{\pm}\frac{e^{-(|\mathbf{k}_{\mathbf{J}}|\pm\beta)^2/4E^2}}{|\mathbf{k}_{\mathbf{J}}|\pm\beta}e^{-j\mathbf{k}_{\mathbf{J}}\cdot\mathbf{r}},\end{aligned} \tag{31}$$

where "erfc" denotes the complementary error function, $\beta=\omega/c$, $\mathbf{k}_{\mathbf{J}}=\mathbf{k}+j_1\mathbf{b}_1+j_2\mathbf{b}_2+j_3\mathbf{b}_3$, $\mathbf{r}_{\mathbf{I}}=i_1\mathbf{a}_1+i_2\mathbf{a}_2+i_3\mathbf{a}_3$, and $\mathbf{I}=(i_1,i_2,i_3)$ and $\mathbf{J}=(j_1,j_2,j_3)$ are multi-indices of integers. The sums with index "±" represent the sum of two terms, one with the "+" sign and the other with the "−" sign. The parameter E is an arbitrary positive real number. An adequate choice that optimizes the convergence rate of the two triple series is $E=\sqrt{\pi}/V_{\text{cell}}^{1/3}$. The formula assumes that both β and $\mathbf{k}$ are real valued. It can be verified that all the terms in the right-hand side of Eq. (31) are smooth functions in the vicinity of $\mathbf{r}=\mathbf{0}$ and $\mathbf{k}=\mathbf{0}$.

It is interesting to note that the imaginary part of the interaction dyadic can be evaluated in close analytical form. To this end, Eq. (31) can be rewritten as

$$\Phi_{\text{reg}}(\mathbf{r},\omega,\mathbf{k}) = \frac{1}{4\pi}\frac{j\sin(\beta r)}{r} + V(\mathbf{r},\omega,\mathbf{k}), \tag{32}$$

where $V(\mathbf{r},\omega,\mathbf{k})$ is the sum of the last four terms in the right-hand side of Eq. (31). Direct calculations demonstrate that $V(\mathbf{r})=V^*(-\mathbf{r})$, assuming always that β and $\mathbf{k}$ are real valued. In particular, this property implies that $V(0,\omega,\mathbf{k})$, as well as the second order space derivatives $\frac{\partial^2 V}{\partial x_i \partial x_j}(0,\omega,\mathbf{k})$, are real valued functions. Hence, substituting Eq. (32) into Eq. (14), we obtain

$$\mathrm{Im}\{\overline{\overline{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k})\} = \left.\left[\left(\frac{\omega}{c}\right)^2\overline{\overline{\mathbf{I}}} + \nabla\nabla\right]\left(\frac{1}{4\pi}\frac{\sin(r\omega/c)}{r}\right)\right|_{\mathbf{r}=\mathbf{0}}. \tag{33}$$

After straightforward calculations, the formula simplifies to

$$\mathrm{Im}\{\overline{\overline{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k})\} = \frac{1}{6\pi}\left(\frac{\omega}{c}\right)^3\overline{\overline{\mathbf{I}}}. \tag{34}$$

It will be seen in the next section that this formula is consistent with the fact that the dielectric crystal supports electromagnetic modes that propagate coherently with no radiation loss.

For ω and $\mathbf{k}$ real valued, the interaction dyadic verifies

$$\overline{\overline{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k}) = \overline{\overline{\mathbf{C}}}_{\text{int}}(\omega,-\mathbf{k}). \tag{35}$$

This property follows from the symmetry relation $V(\mathbf{r},\omega,\mathbf{k})=V(\mathbf{r},\omega,-\mathbf{k})^*$ and from Eqs. (14) and (32).

Some useful properties of the dyadic $\overline{\overline{\mathbf{C}}}_{e,m}$ are derived next. From Eq. (31), it is evident that $\Phi_{\text{reg}}(\mathbf{r},\omega,\mathbf{k}=0) =\Phi_{\text{reg}}(-\mathbf{r},\omega,\mathbf{k}=0)$. This proves that for arbitrary ω we have that

$$\overline{\overline{\mathbf{C}}}_{e,m}(\omega,0) = 0. \tag{36}$$

This property implies that for $\mathbf{k}=\mathbf{0}$ there is no coupling between the electric and magnetic dipoles. Another interesting result is obtained noting that from Eq. (32) and from $V(\mathbf{r}) =V^*(-\mathbf{r})$, it follows that $\nabla\Phi_{\text{reg}}|_{\mathbf{r}=\mathbf{0}}$ is purely imaginary. Hence, it is found that the dyadic is real valued as follows:

$$\mathrm{Im}\{\overline{\overline{\mathbf{C}}}_{e,m}(\omega,\mathbf{k})\} = 0. \tag{37}$$

III. DIELECTRIC FUNCTION

As in Sec. II C, here it is assumed that the interaction between the dielectric inclusions can be accurately described by only the dipole-type terms. Moreover, for simplicity, we consider that the material is formed by nonbianisotropic particles. In such case, the relation between the local fields and the induced dipole moments is

$$\mathbf{p}_e = \varepsilon_0\overline{\overline{\alpha}}_{ee}\cdot\mathbf{E}_{\text{loc}}, \quad \mathbf{p}_m = \overline{\overline{\alpha}}_{mm}\cdot\mathbf{B}_{\text{loc}}. \tag{38}$$

The polarizability dyadics $\overline{\overline{\alpha}}_{ee}$ and $\overline{\overline{\alpha}}_{mm}$ are symmetric and have unities of volume $[m^3]$.

Within these hypotheses, it is a simple task to obtain the dielectric function of the composite material using the generalized Lorentz-Lorenz formulas. To this end, first we will obtain the induced dipole moments as a function of the macroscopic fields. In the view of property (36), it is legitimate to neglect the terms that involve $\overline{\overline{\mathbf{C}}}_{e,m}$ in the generalized Lorentz-Lorenz relations (27), at least if $\mathbf{k}$ is near the origin of the Brillouin zone. In such circumstances, it is readily found that

$$\frac{\mathbf{p}_e}{\varepsilon_0}\frac{1}{V_{\text{cell}}} = [\overline{\overline{\varepsilon}}_r - \overline{\overline{\mathbf{I}}}]\cdot\mathbf{E}_{\text{av}},$$

$$\frac{\mathbf{p}_m}{\mu_0}\frac{1}{V_{\text{cell}}} = (\overline{\overline{\mu}}_r - \overline{\overline{\mathbf{I}}})\cdot\mathbf{H}_{\text{av}}, \tag{39}$$

where the dyadics $\overline{\overline{\varepsilon}}_r$ and $\overline{\overline{\mu}}_r$ are defined as

$$\overline{\overline{\varepsilon}}_r(\omega,\mathbf{k}) = \overline{\overline{\mathbf{I}}} + \frac{1}{V_{\text{cell}}}[\overline{\overline{\mathbf{I}}} - \overline{\overline{\alpha}}_{ee}\cdot\overline{\overline{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k})]^{-1}\cdot\overline{\overline{\alpha}}_{ee},$$

$$\overline{\overline{\mu}}_r(\omega,\mathbf{k}) = \overline{\overline{\mathbf{I}}} + \frac{1}{V_{\text{cell}}}(\overline{\overline{\mathbf{I}}} - \overline{\overline{\alpha}}_{mm}\cdot\overline{\overline{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k}))^{-1}\cdot\overline{\overline{\alpha}}_{mm}. \tag{40}$$

It is obvious that provided it is possible to neglect the $\mathbf{k}$ dependence of the above expressions, then $\overline{\overline{\varepsilon}}_r$ and $\overline{\overline{\mu}}_r$ can be regarded as the classical "local" effective permittivity and permeability associated with the standard constitutive relations.[18] These expressions are thus a generalization of the classical Clausius-Mossotti formulas.[19] They establish that the effective parameters can be written in terms of the electric polarizability of the particles and of the interaction dyadic $\overline{\overline{\mathbf{C}}}_{\text{int}}$.

However, in the general case in which the spatial dispersion effects are not negligible, it is not meaningful to introduce a magnetic permeability tensor, and the material is preferably characterized by only a dielectric function $\overline{\overline{\varepsilon}}_{\text{eff}}(\omega,\mathbf{k})$.[24]

To compute the dielectric function we use Eqs. (4), (29), and (39) to obtain

$$\frac{\mathbf{p}_m}{V_{\text{cell}}} = (\overline{\overline{\mathbf{I}}} - \overline{\overline{\mu}}_r^{-1})\cdot\left(\frac{\mathbf{k}}{\omega}\times\mathbf{E}_{\text{av}}\right). \tag{41}$$

On the other hand, substituting Eq. (24) into Eq. (3), we find that the generalized polarization vector is equal to

$$\mathbf{P}_{g,\text{av}} = \frac{1}{V_{\text{cell}}}\left(\mathbf{p}_e - \frac{\mathbf{k}}{\omega}\times\frac{\mathbf{p}_m}{\mu_0}\right). \tag{42}$$

Substituting Eqs. (39) and (41) into Eq. (42), and then using the constitutive relation (5), it is found that

$$\overline{\overline{\varepsilon}}_{\text{eff}}(\omega,\mathbf{k}) = \overline{\overline{\varepsilon}}_r(\omega,\mathbf{k}) + c^2\frac{\mathbf{k}}{\omega}\times(\overline{\overline{\mu}}_r^{-1} - \overline{\overline{\mathbf{I}}})\times\frac{\mathbf{k}}{\omega}. \tag{43}$$

The above formula is consistent with the discussion presented in Ref. 18, and in particular, confirms that for local media (for which $\overline{\overline{\varepsilon}}_r$ and $\overline{\overline{\mu}}_r$ can be considered to a good approximation independent of the wave vector) the dielectric function can be written in terms of the local effective parameters.

It is interesting to note that if the particles are lossless, the dielectric function is real valued. In fact, it is known that in order that the balance between the power radiated by the electric dipole and the power absorbed from the local field be zero, it is necessary that the electric polarizability verifies $\mathrm{Im}\{\overline{\overline{\alpha}}_{ee}^{-1}\}=\frac{1}{6\pi}\left(\frac{\omega}{c}\right)^3\overline{\overline{\mathbf{I}}}$ (it is assumed without loss of generality that $\overline{\overline{\alpha}}_{ee}$ has an inverse, and that the coupling between electric and magnetic dipoles is negligible). This property is sometimes known as the Sipe-Kranendonk condition.[3] Using this power balance consistency condition and Eq. (34), it follows that the dyadic $\overline{\overline{\varepsilon}}_r$ can be rewritten as

$$\overline{\overline{\varepsilon}}_r(\omega,\mathbf{k}) = \overline{\overline{\mathbf{I}}} + \frac{1}{V_{\mathrm{cell}}}(\mathrm{Re}\{\overline{\overline{\alpha}}_{ee}^{-1} - \overline{\overline{\mathbf{C}}}_{\mathrm{int}}(\omega,\mathbf{k})\})^{-1}. \tag{44}$$

In particular, it is seen that due to the Sipe-Kranendonk condition, the dyadic $\overline{\overline{\varepsilon}}_r$ is real valued. Since the dyadic $\overline{\overline{\mu}}_r$ has the same property, it follows that the dielectric function of the material (43) is also real valued, consistently with what could be expected for a lossless medium with a center of symmetry. An alternative proof of these results is presented in Ref. 14.

It is important to mention here that apart from the simplifying assumption $\overline{\overline{\mathbf{C}}}_{e,m}\approx 0$, formula (43) represents the exact solution of the homogenization problem formulated in Ref. 18 for a microscopic current of the form (24). In particular, the dispersion characteristic of the electromagnetic modes can be obtained by substituting the dielectric function into Eq. (4), and by calculating the values of $(\omega,\mathbf{k})$ for which the homogeneous system with $\mathbf{P}_{e,\mathrm{av}}=\mathbf{0}$ has nontrivial solutions. This property is valid even away from the long wavelength limit, where $\overline{\overline{\varepsilon}}_{\mathrm{eff}}(\omega,\mathbf{k})$ has arguably no physical meaning, but can still be mathematically defined in a consistent manner using the formalism developed in Ref. 18.

To conclude this section, we briefly discuss what happens if the dyadic $\overline{\overline{\mathbf{C}}}_{e,m}$ is not neglected. In such a situation it is clear that the material can only be characterized using the classical constitutive relations, provided we introduce two tensors $\overline{\overline{\xi}}$ and $\overline{\overline{\zeta}}$ to describe the interaction between dipoles of a different type, in addition to the tensors $\overline{\overline{\varepsilon}}_r$ and $\overline{\overline{\mu}}_r$. This may suggest that the material is, in general, bianisotropic.[15] However, in our opinion such an interpretation is incorrect. In fact, the designation bianisotropic suggests that the medium may be gyrotropic, which is completely impossible since the structure under study obviously has a center of symmetry and the particles are assumed nonbianisotropic. Thus, since the medium is not gyrotropic, the tensors $\overline{\overline{\xi}}$ and $\overline{\overline{\zeta}}$ have no physical meaning. In fact, using Eq. (36) it can be easily verified that $\overline{\overline{\xi}}$ and $\overline{\overline{\zeta}}$ vanish for $\mathbf{k}=\mathbf{0}$. This means that in the problem under study the tensors $\overline{\overline{\xi}}$ and $\overline{\overline{\zeta}}$ are associated with spatial dispersion effects of second order, and not with spatial dispersion of first order as in conventional gyrotropic media. These considerations only underline the fact that when spatial dispersion is strong it is not meaningful to use the classical constitutive relations, and that the medium should instead be characterized by a nonlocal dielectric function.

IV. HOMOGENIZATION OF A LATTICE OF PLASMONIC SPHERES

As an application of the proposed formalism, in this section we homogenize a crystal formed by plasmonic spheres. Recently, it was found that such microstructured material may enable negative refraction at optical frequencies.[20] The study presented in Ref. 20 was based on the dispersion properties of the electromagnetic modes, but no homogenization model was given. Next, we will calculate the dielectric function of the microstructured material giving further physical insights into the phenomena described in Ref. 20.

As in Ref. 20 it is assumed here that the plasmonic spheres can be modeled as electric dipoles with electric polarizability such that [19]

$$\mathrm{Re}\{\alpha_e^{-1}\} = \frac{(\varepsilon_r+2)}{(\varepsilon_r-1)3V_{\mathrm{sph}}}, \tag{45}$$

where $\varepsilon_r(\omega)$ is the relative permittivity of the plasmonic spheres, and $V_{\mathrm{sph}}=4\pi R^3/3$, where R is the radius of the spheres. For simplicity, the absorption of the particles is neglected here. We also restrict our analysis to the case in which the spheres are arranged in a simple cubic lattice, with lattice constant a.

It is assumed that the permittivity of the plasmonic spheres follows a Drude-type dispersion model with $\varepsilon_r(\omega)=1-3\omega_r^2/\omega^2$, where ω_r is the frequency of the plasmonic resonance. The radius of the spheres is taken equal to $R=a/2.1$. Thus, the volume fraction of the inclusions is 45.2%. The frequency ω_r is assumed to be such that $R\omega_r/c=2\pi/100$ (i.e., $R=\lambda/100$ at frequency ω_r). These parameters are similar to ones chosen in Ref. 20.

In the Appendix it is proven that the interaction dyadic of a simple cubic lattice is such that

$$C_{xx}(\omega,\mathbf{k}) \approx \frac{1}{a^3}\left[\frac{1}{3} - 0.15\left(\frac{\omega}{c}a\right)^2 - 0.026[\cos(k_y a)-1] - 0.026[\cos(k_z a)-1] + 0.052[\cos(k_x a)-1]\right], \tag{46}$$

$$C_{xy}(\omega,\mathbf{k}) \approx \frac{0.105}{a}k_x k_y, \tag{47}$$

where $C_{mn}=\hat{\mathbf{u}}_m\cdot\mathrm{Re}\{\overline{\overline{\mathbf{C}}}_{\mathrm{int}}\}\cdot\hat{\mathbf{u}}_n$. The remaining components of the interaction constant can be obtained by permutation of the wave vector coordinates. The above formulas explicitly show the dependence of the interaction constant on the wave vector. It is apparent that the $\mathbf{k}$ dependence is relatively weak, and this may suggest that the nonlocal effects are negligible. It will be shown that this is not the case, and these effects are of crucial importance near a resonance of the inclusions.

To give an idea of the accuracy of formula (46), in Fig. 1 the interaction constant C_{xx} is depicted as a function of the wave vector along several directions of the crystal. The solid lines represent the exact result computed using Eqs. (14) and (31), and the dashed lines represent the results obtained us-

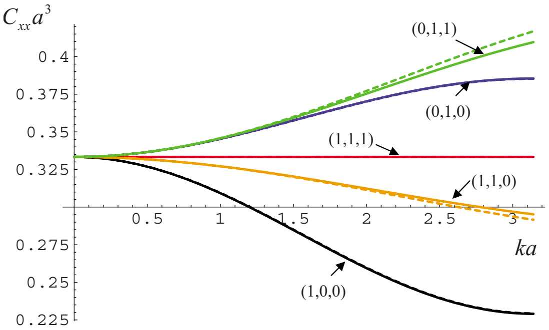

FIG. 1. (Color online) Normalized interaction constant as a function of the wave vector amplitude k, for $\omega=0$ and $\mathbf{k}=k\hat{\mathbf{k}}$. The unit vector $\hat{\mathbf{k}}$ is directed along the direction of the crystal defined by the triplet associated with each curve. The solid lines represent the exact result and the dashed lines the result given by Eq. (46).

ing the approximate formula (46). It is seen that the agreement is excellent, and that for some directions the two curves cannot be distinguished. Similarly, in Fig. 2 the interaction constant is plotted as a function of frequency, and for different wave vectors. The approximate formula describes very accurately the dependence of C_{xx} on frequency for $\omega a/c < 1.0$. This demonstrates that Eq. (46) is an excellent approximation of the exact formula, even for relatively short wavelengths. The rest of the results presented in this section were obtained by using the approximation (46).

From the analysis of Sec. III, it is clear that the dielectric function of the material is such that

$$\bar{\bar{\varepsilon}}_{\text{eff}}(\omega,\mathbf{k}) = \bar{\bar{\mathbf{I}}} + \frac{1}{a^3}(\alpha_e^{-1} - \text{Re}\{\bar{\bar{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k})\})^{-1}. \tag{48}$$

In general, the dielectric function is not diagonal relative to the canonical basis (defined by the primitive vectors of the lattice). In fact, the crossed components of the interaction dyadic are in general different from zero [see Eq. (47)]. However, for propagation along the coordinates axes, the permittivity dyadic becomes diagonal in the canonical basis. It is found that

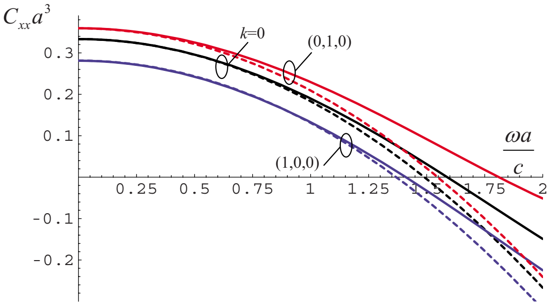

FIG. 2. (Color online) Normalized interaction constant as a function of the frequency ω and for different $\mathbf{k}=k\hat{\mathbf{k}}$ with $ka=\pi/2$. The unit vector $\hat{\mathbf{k}}$ is directed along the direction of the crystal defined by the triplet associated with each curve. The solid lines represent the exact result and the dashed lines the result given by Eq. (46).

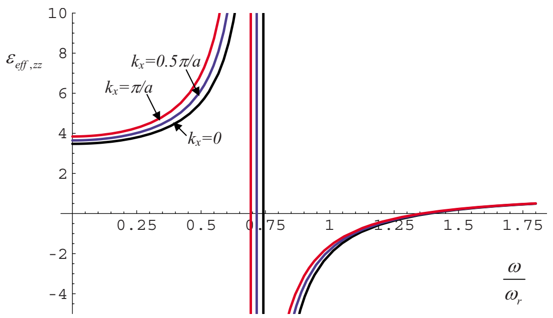

FIG. 3. (Color online) Effective permittivity of the composite material as a function of the normalized frequency, for $k_y=k_z=0$ and different values of k_x.

$$\varepsilon_{\text{eff},xx} = 1 + \frac{1}{a^3}\frac{1}{\text{Re}\{\alpha_e^{-1}\} - C_{xx}(\omega,\mathbf{k})}, \tag{49}$$

where $\varepsilon_{\text{eff},xx}=\hat{\mathbf{u}}_x\cdot\bar{\bar{\varepsilon}}_{\text{eff}}\cdot\hat{\mathbf{u}}_x$. Similar formulas hold for the yy and zz components of the dielectric function.

The ε_{zz} component of the dielectric constant is depicted as a function of frequency in Fig. 3. The composite material has a resonance at $\omega\approx 0.73\omega_r$. The dielectric function is also plotted for different values of k_x, with $k_y=k_z=0$. It is seen that the dependence of the dielectric function on k_x is rather weak, except for the range of frequencies near $\omega\approx 0.7\omega_r$. Notice that the value of the resonance frequency is considerably smaller than the resonance of the individual particles. This property is a consequence of the inclusions being closely packed.[20] This enables a strong mutual coupling that shifts the resonance to the red.

To give further insights, let us consider propagation along the x direction, with $k_y=k_z=0$. It is simple to verify from the dispersion characteristic that the medium supports two transverse electromagnetic (TEM) modes and one longitudinal mode. They verify

$$\left(\frac{\omega}{c}\right)^2\varepsilon_{\text{eff},zz}(\omega,k_x) - k_x^2 = 0, \quad \text{TEM modes},$$

$$\varepsilon_{\text{eff},xx}(\omega,k_x) = 0, \quad \text{longitudinal mode}, \tag{50}$$

where $\varepsilon_{\text{eff},xx}$ and $\varepsilon_{\text{eff},zz}$ are defined consistently with Eq. (49). In Fig. 4, we depict the band structure of the electromagnetic modes. The band associated with the longitudinal mode is nearly dispersionless and has $\omega/\omega_r = 1.38$ at the origin of the Brillouin zone. The remaining bands are doubly degenerated and are associated with TEM modes. It is seen that the dispersion characteristic of the TEM mode, $\omega=\omega(k_x)$, increases with k_x (forward wave) for k_x near the origin of the Brillouin. However, for $k_x a > 0.95$ the dispersion $\omega(k_x)$ decreases with k_x. This behavior is characteristic of a backward wave. The described phenomenon is narrowband. Strategies to improve the bandwidth were discussed in Ref. 20.

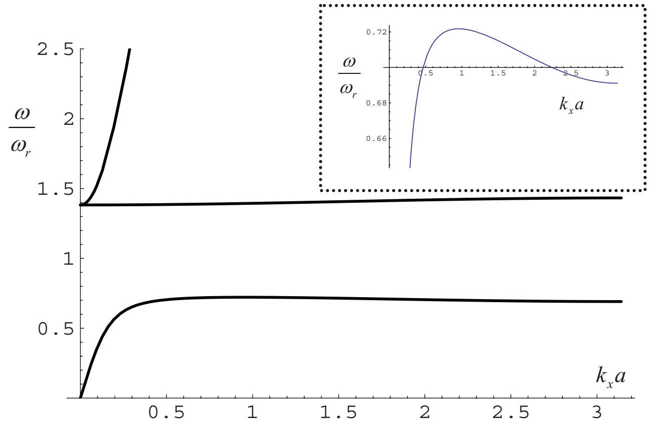

FIG. 4. (Color online) Band structure of the composite material for propagation along the x direction. The inset is a zoom of the first band, showing clearly that backward wave propagation is supported.

The emergence of backward propagation can be further confirmed from the contour plot of the dispersion characteristic. In Fig. 5, the dispersion contours are shown for propagation in the xoy plane. These were obtained by solving numerically the equation

$$\left(\frac{\omega}{c}\right)^2 \varepsilon_{\text{eff},zz}(\omega,k_x,k_y) - k_x^2 - k_y^2 = 0, \quad k_z = 0, \tag{51}$$

where $\varepsilon_{\text{eff},zz}$ is given by an expression analogous to Eq. (49). The above equation corresponds to the dispersion characteristic of the TEM mode with electric field along the z direction. It is seen that consistently with the band structure diagram, for $\omega/\omega_r < 0.64$ the Poynting vector is nearly parallel to the wave vector and there is no backward wave propagation. On the other hand, for $0.64 < \omega/\omega_r < 0.73$ and for each direction in the xoy plane, the medium supports two different modes that have exactly the same polarization. One of the modes is a forward wave and the other is a backward wave (wave vector and Poynting vector are antiparallel). For $\omega/\omega_r \approx 0.72$ the contours of the backward wave mode are nearly circular, and thus the propagation is almost isotropic. These results are completely consistent with the analysis of Ref. 20. The analysis presented here demonstrates that the backward wave phenomenon emerges due to structural spatial dispersion effects. It is clear that these effects are of key importance near the resonance of the plasmonic spheres.

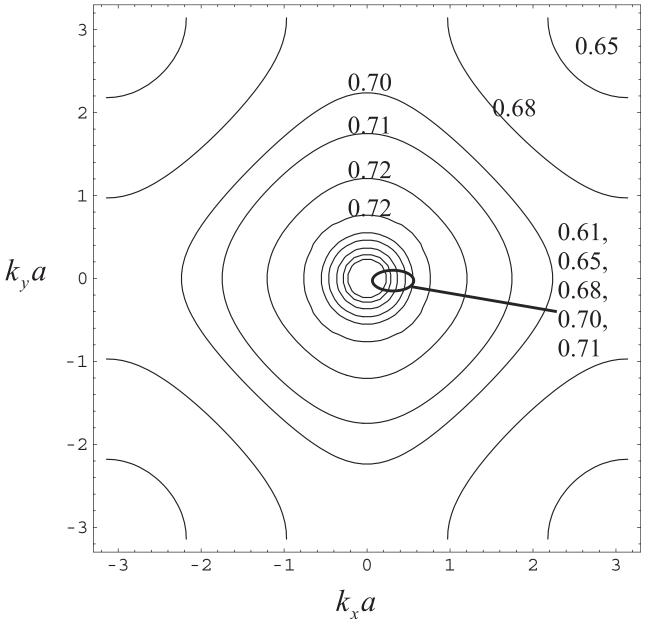

FIG. 5. Contour plot of the dispersion characteristic of the TEM mode for propagation in the xoy plane. The number associated with each curve specifies the value of ω/ω_r.

V. CONCLUSION

In this work, the homogenization concepts introduced in Ref. 18 were applied to obtain a generalized form of the classical Lorentz-Lorenz formulas. We derived a general relation between the local fields, the macroscopic fields, and the induced microscopic current. The main result establishes that the generalized Lorentz-Lorenz formulas are given by Eqs. (27), provided the interaction between the inclusions can be described by only the dipolar terms. These formulas take into account the effects of both frequency dispersion and spatial dispersion, and can be easily generalized to the case in which the unit cell contains more than one particle, or to the case in which higher order multipoles need to be considered.

Using the generalized Lorentz-Lorenz formulas, we obtained the dielectric function of a generic composite medium that is modeled as a lattice of electric and magnetic dipoles. We applied this result to a study of the properties of a material formed by plasmonic spheres. We found out that the spatial dispersion effects are dominant near the resonance of the inclusions, causing the emergence of anomalous phenomena such as backward wave propagation and negative refraction.

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APPENDIX: INTERACTION DYADIC FOR A SIMPLE CUBIC LATTICE

Here, we derive an approximate formula for the interaction dyadic $\overline{\overline{\mathbf{C}}}_{\text{int}}(\omega,\mathbf{k})$ of a simple cubic lattice. Let $C_{m,n} = \hat{\mathbf{u}}_m \cdot \text{Re}\{\overline{\overline{\mathbf{C}}}_{\text{int}}\} \cdot \hat{\mathbf{u}}_n$ be the component of the interaction dyadic along the coordinate axes $\hat{\mathbf{u}}_m$ and $\hat{\mathbf{u}}_n$ $(m,n=x,y,z)$. By symmetry arguments it is sufficient to characterize C_{xx} and C_{xy}, since the remaining elements can be obtained by permutation of the wave vector components.

First, we analyze the dependence of C_{xx} with k_x, supposing that $\omega=0$ and that $k_y=k_z=0$. It can be verified from Eq. (14) that in this case C_{xx} is an even periodic function of k_x. Thus, it has an expansion of the form

$$C_{xx}\Big|_{k_y=k_z=0}^{\omega=0} = A_0 + A_1\cos(k_x a) + A_2\cos(2k_x a) + \cdots , \tag{A1}$$

where A_i are some Fourier coefficients that can be calculated numerically using Fourier theory. Direct calculations show that $A_1 \approx 0.052$. Using Eq. (30), the above formula can be rewritten as

$$C_{xx}\Big|_{k_y=k_z=0}^{\omega=0} = \frac{1}{3a^3} + A_1[\cos(k_x a) - 1] + A_2[\cos(2k_x a) - 1] + \cdots . \tag{A2}$$

Neglecting all the terms except the first two, it is found that to a first approximation,

$$C_{xx}\Big|_{k_y=k_z=0}^{\omega=0} \approx \frac{1}{3a^3} + \frac{0.052}{a^3}[\cos(k_x a) - 1]. \tag{A3}$$

Proceeding along a similar line of thought it can be found that

$$C_{xx}\Big|_{k_x=k_z=0}^{\omega=0} \approx \frac{1}{3a^3} - \frac{0.026}{a^3}[\cos(k_y a) - 1]. \tag{A4}$$

It is obvious that a similar formula holds if the roles of k_y and k_z are interchanged.

Finally, the dependence of C_{xx} on ω can be approximated by a Taylor series of order two. Calculating the derivatives numerically it is found that, $C_{xx}|_{\mathbf{k}=0} \approx 1/3a^3 - (0.15/a^3) \times (\omega a/c)^2$. Using this result and Eqs. (A3) and (A4), and supposing that the variations of the different coordinates are to some extent independent, we arrive at formula (46).

To characterize C_{xy}, we expand the function in a Taylor series of order two at the origin. It can be verified that C_{xy} is an odd function of k_x and k_y, and an even function of k_z. Thus, it is clear that the second order Taylor series only has the term proportional to $k_x k_y$. Explicit calculations yield formula (47).

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