Local-field effects in magnetodielectric media: Negative refraction and absorption reduction

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We give a microscopic derivation of the Clausius-Mossotti relations for a homogeneous and isotropic magnetodielectric medium consisting of radiatively broadened atomic oscillators. To this end the diagram series of electromagnetic propagators is calculated exactly for an infinite bicubic lattice of dielectric and magnetic dipoles for a small lattice constant compared to the resonance wavelength λ. Modifications of transition frequencies and linewidth of the elementary oscillators are taken into account in a self-consistent way by a proper incorporation of the singular self-interaction terms. We show that in radiatively broadened media sufficiently close to the free-space resonance the real part of the index of refraction approaches the value -2 in the limit of $\rho\lambda^3 \gg 1$, where ρ is the number density of scatterers. Since at the same time the imaginary part vanishes as $1/\rho$, local field effects can have important consequences for realizing low-loss negative index materials.

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I. INTRODUCTION

It is well-known that in dense dielectric materials the induced polarization $\mathbf{P}$ alters the field strength $\mathbf{E}_{loc}$ acting on the constituents (i.e., the local field) compared to the average macroscopic field $\mathbf{E}_m$. Macroscopic considerations show that in systems with high symmetry such as a cubic lattice the two fields are related to each other according to $\mathbf{E}_{loc}=\mathbf{E}_m+\mathbf{P}/(3\varepsilon_0)$ [1,2]. This leads to the well-known Clausius-Mossotti relation for the permittivity $\varepsilon(\omega)$,

$$\varepsilon(\omega) = 1 + \frac{\rho\alpha(\omega)/\varepsilon_0}{1-\rho\alpha(\omega)/(3\varepsilon_0)}, \tag{1}$$

where ρ is the density and $\alpha(\omega)$ the polarizability of the oscillators. Similar arguments hold for a purely magnetic material [3], except that the required densities are usually much higher due to the smallness of magnetic dipole moments and polarizabilities. In linear response $\alpha(\omega)$ is well-described by a damped-oscillator model [1],

$$\alpha(\omega) = \alpha' + i\alpha'' = \frac{\alpha_0}{\omega_0^2-\omega^2-i\gamma\omega}. \tag{2}$$

The corresponding (real-valued) parameters such as the oscillator strength α_0, the resonance frequency and width, ω_0 and γ, are determined by the microscopic model. In general the linewidth γ contains radiative as well as nonradiative contributions. For purely radiative interaction these parameters are strongly affected by the renormalization of energy levels and spontaneous emission processes caused by the interaction with the vacuum electromagnetic field in the medium [4–11]. Since the mode structure of the electromagnetic field inside a dense medium can be substantially modified compared to free space, one would expect that the polarizability entering Eq. (1) is different from that in free space. In a macroscopic approach $\alpha(\omega)$ is, however, an input function and no conclusion can be drawn about possible changes due to the different structure of the vacuum modes inside the medium. To take into account the modification of transition frequencies and radiative linewidth in a dense medium in a self-consistent way requires a microscopic approach.

In the present paper we develop a microscopic approach to local field effects in dense materials with simultaneous dielectric and magnetic response using Green function techniques similar to those used by de Vries and Lagendijk for purely dielectric materials [12]. To this end we consider an infinitely extended bicubic lattice of electric and magnetic point dipoles with isotropic response with a small lattice constant compared to the transition wavelength. We, however, do not make use of the assumptions made in [12] to renormalize the singular self-interaction contributions to the lattice *T*matrix which eliminated radiative contributions to linewidth and transition frequencies altogether. We show that instead the self-interaction contributions can be summed to yield the dressed *t* matrix of an isolated oscillator interacting with the vacuum modes of the electromagnetic field in free space. In this way we derive Clausius-Mossotti relations for general, radiatively broadened, isotropic magnetodielectrica. Apart from nonradiative broadenings, the electric and magnetic polarizabilities entering these equations are shown to be exactly those of free space. We then show that simultaneous local-field corrections to electric *and* magnetic fields in purely radiatively broadened magnetodielectrica have a surprising and potentially important effect: For sufficiently large densities the real part of the refractive index saturates at the level of -2. At the same time, the imaginary part of the complex index approaches zero inversely proportional to the density. Thus the medium becomes transparent and left-handed, i.e., displays a negative index of refraction with low absorption.

II. LOCAL-FIELD EFFECTS AND RENORMALIZATION OF RADIATIVE SELF-INTERACTION IN DIELECTRIC MEDIA

We start by developing a microscopic scattering approach to local-field effects in dielectric media taking into account possible material induced modifications of radiative linewidth and transition frequencies in a self-consistent way. To this end we consider a simple cubic lattice of electric point dipoles with isotropic bare polarizability α_b

$$\alpha_b(\mathbf{r}) = \alpha_b \sum_{\mathbf{R}} \delta(\mathbf{r}-\mathbf{R}), \tag{3}$$

where $\mathbf{R}$ denotes lattice vectors. The dipoles interact with the quantized electromagnetic field $\hat{\mathbf{E}}$ which obeys the vector Helmholtz equation

$$\vec{\nabla}\times\vec{\nabla}\times\hat{\mathbf{E}}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\hat{\mathbf{E}}(\mathbf{r},\omega) = \mu_0\omega^2\hat{\mathbf{P}}. \tag{4}$$

In the weak excitation, i.e., linear response limit, the operator of the microscopic electric polarization $\hat{\mathbf{P}}$ has the form $\hat{\mathbf{P}}(\mathbf{r})=\alpha_b(\mathbf{r})\hat{\mathbf{E}}(\mathbf{r},\omega)$. Solving Eq. (4) we can determine the (isotropic) dispersion relation $k=k(\omega)$ from which the permittivity $\varepsilon(\omega)$ can be extracted. In the linear response limit the solution of the quantum mechanical interaction problem can most easily be obtained by means of Green function techniques. In particular it is sufficient to calculate the scattering T matrix of the oscillator lattice. The dispersion relation can then be obtained via [13–15]

$$\det T^{-1} = 0. \tag{5}$$

The scattering T matrix obeys a linear Dyson equation,

$$T = V + V\mathcal{G}^{(0)}V + \cdots = V + V\mathcal{G}^{(0)}T, \tag{6}$$

where $\mathcal{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega)$ is the free-space retarded propagator of the electric field which is a solution to the classical vector Helmholtz equation

$$\vec{\nabla}\times\vec{\nabla}\times\mathcal{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega) - \frac{\omega^2}{c^2}\mathcal{G}^{(0)}(\mathbf{r},\mathbf{r}',\omega) = \mathbb{1}\delta(\mathbf{r}-\mathbf{r}'), \tag{7}$$

and

$$V(\mathbf{r},\omega) = -\frac{\omega^2\alpha_b(\mathbf{r})}{\varepsilon_0 c^2} \tag{8}$$

is a linear, isotropic point vertex. Note that integration over spatial variables was suppressed in Eq. (6) for notational simplicity.

For a cubic lattice of isotropic scatterers, the series can be summed up to yield [16]

$$T(\mathbf{k},\mathbf{k}') = -\sum_{\mathbf{R}'} e^{i(\mathbf{k}-\mathbf{k}')\mathbf{R}'}\left\{\frac{1}{t(\omega)} + \sum_{\mathbf{R}\neq 0} e^{i\mathbf{k}'\mathbf{R}}\mathcal{G}^{(0)}(\mathbf{R})\right\}^{-1}, \tag{9}$$

where $\mathcal{G}^{(0)}(\mathbf{R})$ stands for $\mathcal{G}^{(0)}(\mathbf{r},\mathbf{r}+\mathbf{R},\omega_0)$ which, due to the discrete translation invariance, is independent of $\mathbf{r}$. The single-particle scattering t matrix $t(\omega)$ is determined by the bare polarizability [12]

$$t(\omega)^{-1} = \left(\frac{\omega^2}{c^2}\frac{\alpha_b}{\varepsilon_0}\right)^{-1} + \mathcal{G}^{(0)}(0). \tag{10}$$

Note that $\mathcal{G}^{(0)}(0)$ is diagonal and isotropic. In Eq. (9) we have separated the contribution of the lattice ($\Sigma_{\mathbf{R}\neq 0}$) from the multiple scattering events at the same oscillator [$\mathcal{G}^{(0)}(0)$]. This separation is crucial since $\mathcal{G}^{(0)}(0)$ is singular. Rather than eliminating this singularity by a regularization procedure as done in [12], we note that expression (10) gives the single-particle scattering t matrix $t(\omega)$ dressed by the interaction with the vacuum field in free space. This quantity is experimentally observable and is related to the single-particle polarizability $\alpha(\omega)$ in free space:

$$\alpha(\omega) = t(\omega)\frac{c^2}{\omega^2}\varepsilon_0; \tag{11}$$

α_b on the other hand is not observable and thus only a theoretical notion. At this point other broadening mechanisms can be incorporated by adding appropriate nonradiative decay rates γ^{nonrad} to the polarizability $\alpha(\omega)$ [Eq. (11)] [cf. Eq. (2) and discussion thereafter].

Obviously, for the radiative part separating the sum $\Sigma_{\mathbf{R}} e^{i\mathbf{k}'\mathbf{R}}\mathcal{G}^{(0)}(\mathbf{R})$ into $\mathcal{G}^{(0)}(0)+\Sigma_{\mathbf{R}\neq 0} e^{i\mathbf{k}'\mathbf{R}}\mathcal{G}^{(0)}(\mathbf{R})$ does the trick of writing the full lattice T matrix in terms of the *known* free space t matrix. As a drawback we are left with the sum over the lattice vectors $\mathbf{R}\neq 0$. Unfortunately this sum cannot be evaluated exactly and has to be treated approximately.

According to Poisson's summation formula

$$\sum_{n=-\infty}^{\infty} f(n) = \sum_{k=-\infty}^{\infty}\int_{-\infty}^{\infty} dx f(x) e^{-2\pi i k x}, \tag{12}$$

the sum over $\mathbf{R}\neq 0$ can be expressed in terms of a real space integral and a sum over inverse lattice vectors $\mathbf{K}$ of the Fourier transform of the free space Green function $\widetilde{\mathcal{G}^{(0)}}(\mathbf{p})$,

$$\sum_{\mathbf{R}\neq 0} e^{i\mathbf{k}\mathbf{R}}\mathcal{G}^{(0)}(\mathbf{R}) = \sum_{\mathbf{K}}\int d\mathbf{r}d\mathbf{p}\frac{\Xi(|\mathbf{r}|)}{(2\pi a)^3} e^{i(\mathbf{p}+\mathbf{k}-\mathbf{K})\mathbf{r}}\widetilde{\mathcal{G}^{(0)}}(p). \tag{13}$$

Here $\Xi(|\mathbf{r}|)$ with $\Xi(0)=0$ and $\Xi(|\mathbf{r}|>0)\to 1$ some smooth function introduced to prevent the integral from touching the excluded singular point $\mathbf{r}=0$.

In the following we restrict the discussion to lattices with a lattice constant much smaller than the resonant wavelength, i.e., $ka \ll 1$. In this limit the lattice of oscillators behaves essentially as a homogeneous medium. Contributions from large $\mathbf{K}$ vectors to the sum, which reflect the discreteness of the lattice, can be neglected as long as the singular contribution from the origin has been excluded. Therefore we only keep the term $\mathbf{K}=0$ and assume a Gaussian cutting function $\Xi(|\mathbf{r}|)=1-e^{-r^2/\delta^2}$, with $\delta \ll a$. This yields

$$\sum_{\mathbf{R}\neq 0} e^{i\mathbf{kR}}\mathcal{G}^{(0)}(\mathbf{R}) \approx \frac{1}{a^3}\widetilde{\mathcal{G}^{(0)}}(k) - \frac{1}{a^3}\frac{\pi^{3/2}\delta^3}{(2\pi)^3} \times \int d\mathbf{p} p^2 e^{-\delta^2(k^2+p^2)/4} e^{-\delta^2\mathbf{k}\cdot\hat{\mathbf{p}}/2}\widetilde{\mathcal{G}^{(0)}}(p), \tag{14}$$

where $\hat{\mathbf{p}}=\mathbf{p}/|\mathbf{p}|$. Apart from the Gaussian p integral which provides a smooth cutoff in reciprocal space, δ can be treated as a small parameter. That allows one to carry out the integration analytically which in leading order of δ yields

$$\sum_{\mathbf{R}\neq 0} e^{i\mathbf{kR}}\mathcal{G}^{(0)}(\mathbf{R}) \approx \frac{1}{a^3}\widetilde{\mathcal{G}^{(0)}}(k) - \frac{1}{a^3}\frac{1}{3\omega^2/c^2}\mathbb{1}. \tag{15}$$

The free-space Green tensor $\widetilde{\mathcal{G}^{(0)}}(k)$ is given by [12]

$$\widetilde{\mathcal{G}^{(0)}}(k) = \left(\frac{\omega^2}{c^2}\mathbb{1} - |\mathbf{k}|^2\Delta_{\mathbf{k}}\right)^{-1}, \tag{16}$$

with $\Delta_{\mathbf{k}}=\mathbb{1}-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}}$ being a projector to directions orthogonal to $\mathbf{k}$.

With this we are ready to evaluate Eq. (5) which reads in the limit $ka \ll 1$

$$\det\left(\frac{1}{\frac{\omega^2}{c^2}\rho\alpha(\omega)/\varepsilon_0}\mathbb{1} + \frac{1}{\frac{\omega^2}{c^2}\mathbb{1} - |\mathbf{k}|^2\Delta_{\mathbf{k}}} - \frac{\mathbb{1}}{3\frac{\omega^2}{c^2}}\right) = 0. \tag{17}$$

Solving Eq. (17) for the (isotropic) dispersion $k=k(\omega)$ with $k(\omega)=\varepsilon(\omega)\omega^2/c^2$ finally yields

$$\varepsilon(\omega) = 1 + \frac{\rho\alpha(\omega)/\varepsilon_0}{1-\rho\alpha(\omega)/3\varepsilon_0}. \tag{18}$$

This is the well-known Clausius-Mossotti relation where for purely radiatively broadened systems $\alpha(\omega)$ is the dressed polarizability of an isolated oscillator interacting with the free-space electromagnetic vacuum field.

III. LOCAL-FIELD EFFECTS FOR MAGNETODIELECTRICS

We now extend the above discussion to the case of a bicubic lattice of electric and magnetic dipole oscillators. The microscopic, space-dependent bare electric polarizability $\alpha_{be}(\mathbf{r})$ is then given by

$$\alpha_{be}(\mathbf{r}) = \alpha_{be}\sum_{\mathbf{R}}\delta(\mathbf{r}-\mathbf{R}) = \frac{\alpha_{be}}{a^3}\sum_{\mathbf{K}} e^{i\mathbf{Kr}} \tag{19}$$

and, similarly, the bare magnetic polarizability by

$$\alpha_{bm}(\mathbf{r}) = \alpha_{bm}\sum_{\mathbf{R}}\delta(\mathbf{r}-\mathbf{R}-\Delta\mathbf{r}) = \frac{\alpha_{bm}}{a^3}\sum_{\mathbf{K}} e^{i\mathbf{K}(\mathbf{r}-\Delta\mathbf{r})}. \tag{20}$$

Here $\mathbf{R}$ denotes again the lattice vectors and $\Delta\mathbf{r}$ the spacing between the electric and magnetic sublattices. The bare atomic polarizabilities α_{be} and α_{bm} are assumed to be scalar for simplicity corresponding to an isotropic medium. The last expressions in Eqs. (19) and (20) give the bare polarizabilities in reciprocal space, with $\mathbf{K}$ being the reciprocal lattice vectors.

Due to the simultaneous presence of electric and magnetic dipole lattices we now have to solve the coupled set of vector Helmholtz equations for the operators of the electric and magnetic fields

$$\nabla\times\nabla\times\hat{\mathbf{E}} - \frac{\omega^2}{c^2}\hat{\mathbf{E}} = i\omega\mu_0\nabla\times\hat{\mathbf{M}} + \mu_0\omega^2\hat{\mathbf{P}} \tag{21}$$

and

$$\nabla\times\nabla\times\hat{\mathbf{H}} - \frac{\omega^2}{c^2}\hat{\mathbf{H}} = \frac{\omega^2}{c^2}\hat{\mathbf{M}} - i\omega\nabla\times\hat{\mathbf{P}}. \tag{22}$$

In linear response the operator of the polarization $\hat{\mathbf{P}}$ and the magnetization $\hat{\mathbf{M}}$ are proportional to the electric and magnetic fields, respectively, $\hat{\mathbf{P}}(\mathbf{r})=\alpha_{be}(\mathbf{r})\hat{\mathbf{E}}(\mathbf{r})$ and $\hat{\mathbf{M}}(\mathbf{r})=\mu_0\alpha_{bm}(\mathbf{r})\hat{\mathbf{H}}(\mathbf{r})$.

In the following we will pursue a slightly different approach to solve the coupled set of equations than used in the previous section. Taking into account the lattice symmetry we first write the field variables in the form

$$\hat{\mathbf{E}}(\mathbf{r}) = \int_{1.\mathrm{BZ}} d\mathbf{k}\sum_{\mathbf{K}}\tilde{\mathbf{E}}(\mathbf{k}-\mathbf{K})e^{i(\mathbf{k}-\mathbf{K})\mathbf{r}}, \tag{23}$$

where the dependence on frequency ω was suppressed for notational simplicity. The subscript denotes integration over the first Brillouin zone. Substituting this and the corresponding expression for $\hat{\mathbf{H}}$ into Eqs. (21) and (22) gives the Helmholtz equations in reciprocal space. After some elementary manipulations the following closed set of equations is derived:

$$\left[\frac{\mathbb{1}}{\frac{\omega^2}{c^2}\rho\alpha_{be}/\varepsilon_0} + \sum_{\mathbf{K}}\frac{1}{\frac{\omega^2}{c^2}\mathbb{1} - |\mathbf{k}-\mathbf{K}|^2\Delta_{\mathbf{k}-\mathbf{K}}}\right]\sum_{\mathbf{K}'}\tilde{\mathbf{E}}(\mathbf{k}-\mathbf{K}') = \frac{\mu_0\alpha_{bm}}{\omega\alpha_{be}}\sum_{\mathbf{K}}\frac{e^{i\mathbf{K}\Delta\mathbf{r}}}{\frac{\omega^2}{c^2}\mathbb{1} - |\mathbf{k}-\mathbf{K}|^2\Delta_{\mathbf{k}-\mathbf{K}}} (\mathbf{k}-\mathbf{K})\times\sum_{\mathbf{K}'}\tilde{\mathbf{H}}(\mathbf{k}-\mathbf{K}')e^{-i\mathbf{K}'\Delta\mathbf{r}}, \tag{24}$$

$$\left[\frac{\mathbb{1}}{\frac{\omega^2}{c^2}\rho\mu_0\alpha_{bm}} + \sum_{\mathbf{K}}\frac{1}{\frac{\omega^2}{c^2}\mathbb{1} - |\mathbf{k}-\mathbf{K}|^2\Delta_{\mathbf{k}-\mathbf{K}}}\right]\sum_{\mathbf{K}'}\tilde{\mathbf{H}}(\mathbf{k}-\mathbf{K}')e^{-i\mathbf{K}'\Delta\mathbf{r}} = -\frac{c^2\alpha_{be}}{\omega\mu_0\alpha_{bm}}\sum_{\mathbf{K}}\frac{e^{-i\mathbf{K}\Delta\mathbf{r}}}{\frac{\omega^2}{c^2}\mathbb{1} - |\mathbf{k}-\mathbf{K}|^2\Delta_{\mathbf{k}-\mathbf{K}}} (\mathbf{k}-\mathbf{K})\times\sum_{\mathbf{K}'}\tilde{\mathbf{E}}(\mathbf{k}-\mathbf{K}'), \tag{25}$$

where $\rho=1/a^3$ is the particle density. The sum in the brackets on the left-hand sides of Eqs. (24) and (25) can be rewritten as

$$\rho\sum_{\mathbf{K}}\frac{1}{\frac{\omega^2}{c^2}\mathbb{1}-|\mathbf{k}-\mathbf{K}|^2\Delta_{\mathbf{k}-\mathbf{K}}} = \sum_{\mathbf{R}} e^{i\mathbf{k}\mathbf{R}}\mathcal{G}^{(0)}(\mathbf{R}) = \mathcal{G}^{(0)}(0) + \sum_{\mathbf{R}\neq 0} e^{i\mathbf{k}\mathbf{R}}\mathcal{G}^{(0)}(\mathbf{R}),$$

where in the second line we have separated the singular contribution $\mathcal{G}^{(0)}(0)$. One recognizes that this term can be added to the expressions containing the bare polarizabilities in Eqs. (24) and (25) yielding the dressed scattering t matrices for isolated electric and magnetic dipoles interacting with the free-space vacuum field:

$$t_e(\omega)^{-1} = \left(\frac{\omega^2}{c^2}\frac{\alpha_{be}}{\varepsilon_0}\right)^{-1} + \mathcal{G}^{(0)}(0), \tag{26}$$

$$t_m(\omega)^{-1} = \left(\frac{\omega^2\mu_0}{c^2}\alpha_{bm}\right)^{-1} + \mathcal{G}^{(0)}(0). \tag{27}$$

The sum over the Green function excluding $\mathbf{R}=0$ can be evaluated in a similar way as in the previous section. If we again assume a lattice constant a much smaller than the resonant wavelength, reciprocal $\mathbf{K}$ vectors different from zero can be disregarded. This leads to

$$\left[\frac{1}{\rho t_e(\omega)} + \widetilde{\mathcal{G}^{(0)}}(\mathbf{k}) - \frac{1}{3\omega^2/c^2}\right]\hat{\mathbf{E}}(\mathbf{k}) = \frac{\mu_0\alpha_{bm}}{\omega\alpha_{be}}\frac{1}{\frac{\omega^2}{c^2}\mathbb{1}-k^2\Delta_{\mathbf{k}}}\mathbf{k}\times\hat{\mathbf{H}}(\mathbf{k}), \tag{28}$$

$$\left[\frac{1}{\rho t_m(\omega)} + \widetilde{\mathcal{G}^{(0)}}(\mathbf{k}) - \frac{1}{3\omega^2/c^2}\right]\hat{\mathbf{H}}(\mathbf{k}) = \frac{c^2\alpha_{be}}{\omega\mu_0\alpha_{bm}}\frac{1}{\frac{\omega^2}{c^2}\mathbb{1}-k^2\Delta_{\mathbf{k}}}\mathbf{k}\times\hat{\mathbf{E}}(\mathbf{k}). \tag{29}$$

Since we are furthermore only interested in propagating, i.e., transversal modes, we can further simplify the calculation by projecting onto transversal modes using $\Delta_{\mathbf{k}}$,

$$\left[\frac{1}{\frac{\omega^2}{c^2}\rho\alpha_e(\omega)/\varepsilon_0} + \frac{1}{\frac{\omega^2}{c^2}-k^2} - \frac{1}{3\frac{\omega^2}{c^2}}\right]\Delta_{\mathbf{k}}\hat{\mathbf{E}}(\mathbf{k}) = \frac{\mu_0\alpha_{bm}}{\omega\alpha_{be}}\frac{1}{\frac{\omega^2}{c^2}-k^2}\mathbf{k}\times\Delta_{\mathbf{k}}\hat{\mathbf{H}}(\mathbf{k}), \tag{30}$$

$$\left[\frac{1}{\frac{\omega^2}{c^2}\rho\mu_0\alpha_m(\omega)} + \frac{1}{\frac{\omega^2}{c^2}-k^2} - \frac{1}{3\frac{\omega^2}{c^2}}\right]\mathbf{k}\times\Delta_{\mathbf{k}}\hat{\mathbf{H}}(\mathbf{k}) = \frac{c^2\alpha_{be}}{\omega\mu_0\alpha_{bm}}\frac{k^2}{\frac{\omega^2}{c^2}-k^2}\Delta_{\mathbf{k}}\hat{\mathbf{E}}(\mathbf{k}). \tag{31}$$

Here we have substituted the dressed single particle t matrices by the free-space dressed polarizabilities $\alpha_{e(m)}(\omega) = t_{e(m)}(\omega)c^2\omega^{-2}\varepsilon_0(\mu_0^{-1})$.

In order to find the dispersion $k(\omega)=n^2\omega^2/c^2$ we have to determine the solution of the secular equation of the linear set of Eqs. (30) and (31), which results in the condition

$$\left[\frac{1}{\frac{\omega^2}{c^2}\rho\alpha_e(\omega)/\varepsilon_0} + \frac{1}{\frac{\omega^2}{c^2}-k^2} - \frac{1}{3\frac{\omega^2}{c^2}}\right]\times\left[\frac{1}{\frac{\omega^2}{c^2}\rho\mu_0\alpha_m(\omega)} + \frac{1}{\frac{\omega^2}{c^2}-k^2} - \frac{1}{3\frac{\omega^2}{c^2}}\right] = 0. \tag{32}$$

Solving for the refractive index of the transversal modes then gives $n^2=\varepsilon\mu$, where

$$\varepsilon = 1 + \frac{\rho\alpha_e(\omega)/\varepsilon_0}{1-\rho\alpha_e(\omega)/3\varepsilon_0}, \tag{33}$$

$$\mu = 1 + \frac{\rho\mu_0\alpha_m(\omega)}{1-\rho\mu_0\alpha_m(\omega)/3} \tag{34}$$

are the relative dielectric permittivity and magnetic permeability, respectively, both satisfying the Clausius-Mossotti relations.

Note that for longitudinal modes Eqs. (28) and (29) decouple. This can be seen by applying the corresponding projector to longitudinal waves $\hat{\mathbf{k}}\otimes\hat{\mathbf{k}}$ which leads to a disappearance of the cross-coupling terms. The dispersion obtained in this way gives either $\varepsilon=0$ corresponding to electric excitons [17,18] or $\mu=0$ for magnetic excitons.

IV. NEGATIVE REFRACTION AND ABSORPTION REDUCTION DUE TO LOCAL FIELD EFFECTS IN MAGNETODIELECTRIC MEDIA

It is interesting to consider the implications of the Clausius-Mossotti relations for radiatively broadened media in the large density limit. Let us first consider a purely dielectric medium and let us assume that the polarizability $\alpha_e(\omega)=\alpha_e'(\omega)+i\alpha_e''(\omega)$ does not depend on the density, i.e., the medium is radiatively broadened. In this case one finds

$$\varepsilon(\omega) \overset{\rho\to\infty}{\to} -2 + i\frac{1}{\rho}\frac{9\varepsilon_0\alpha_e''}{|\alpha_e|^2}. \tag{35}$$

In the high-density limit and sufficiently close to resonance the response saturates at a value of -2 with an imaginary part that vanishes as $1/\rho$. At this point the medium becomes to-

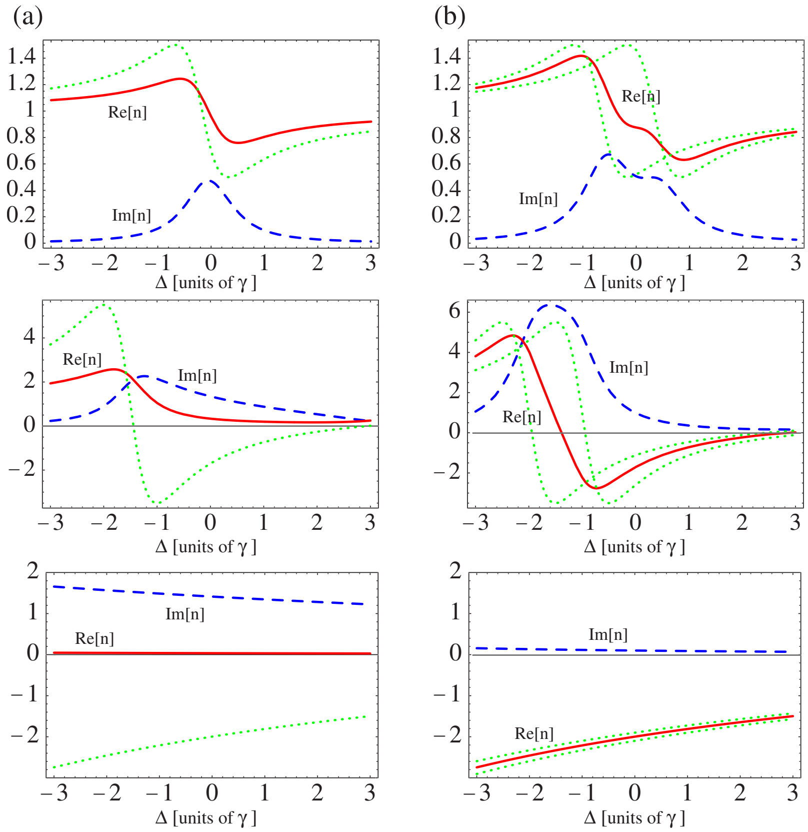

FIG. 1. (Color online) Spectrum of the real (solid) and imaginary (dashed) part of the refractive index as well as the real (dotted) part of the response function(s) ε and/or μ as a function of the detuning Δ for a (a) pure dielectric or magnetic medium for $\rho|\alpha_0|/3$ at $\Delta=0$ equal to $=1/3$ (top), 3 (middle), and 30 (bottom) and (b) magnetodielectric medium for $\rho|\alpha_0|/3$ at $\Delta=0$ equal to $=1/3$ (top), 3 (middle), and 30 (bottom).

tally opaque since the index of refraction attains an imaginary value $n=i\sqrt{2}$ indicating the emergence of a stopping band. This is illustrated in the left column of Fig. 1 for a medium composed of either electric or magnetic dipole oscillators. For small densities ($\rho|\alpha_0|/3=1/3$) the resonance is centered at ω_0 whereas for larger densities ($\rho|\alpha_0|/3=3$) the response shifts to smaller frequencies and is amplified. Eventually ($\rho|\alpha_0|/3=30$) the refractive index becomes almost purely imaginary in which case light cannot propagate any longer.

This behavior changes dramatically if we consider media with overlapping electric and magnetic resonances described by both an electric polarizability $\alpha_e(\omega)$ and a magnetic polarizability $\alpha_m(\omega)$. Independent application of Clausius-Mossotti local-field corrections to the permittivity and the permeability leads in the high density limit to

$$n=-2+i\frac{1}{\rho}\left(\frac{9\varepsilon_0\alpha_e''}{|\alpha_e|^2}+\frac{9\alpha_m''}{\mu_0|\alpha_m|^2}\right). \tag{36}$$

Thus in the spectral overlap region the real part of the index of refraction approaches the value −2, i.e., attains a constant negative value. Furthermore, the imaginary part, responsible for absorption losses, approaches zero in that spectral region as $1/\rho$. This rather peculiar behavior is illustrated in the right column of Fig. 1. One clearly recognizes the emergence of a spectral region around the bare resonance frequency where the real part of the refractive index approaches −2 while the imaginary part is strongly suppressed.

Negative refraction of light is currently one of the most active research areas in photonics [19–21] due to fascinating potential applications such as superlensing [22] or electromagnetic cloaking [23–25]. In recent years substantial progress has been made in realizing negative refraction in so-called metamaterials [26–29]. These are artificial periodic structures of electric and magnetic dipoles with a resonance wavelength much larger than the lattice constant which thus form a quasihomogeneous magnetodielectric medium. In order to achieve a large electromagnetic response, operation close to resonance is needed which is associated with rather substantial losses. The elimination of these losses represents one of the main challenges in the field [30]. We have shown here that in a radiatively broadened medium, i.e., a medium in which a density-dependent broadening mechanism can still be disregarded for sufficiently large densities, local field effects can provide a negative index of refraction and at the same time efficiently suppress absorption losses.

V. SUMMARY

In the present paper we have given a rigorous microscopic derivation of Clausius-Mossotti relations for both the electric and magnetic response in an isotropic, radiatively broadened magnetodielectric medium formed by a simple bicubic lattice of electric and magnetic dipoles. As opposed to previous microscopic approaches we have taken into account possible modifications of the single-particle polarizabilities by the altered electromagnetic vacuum inside the medium in a self-consistent way. For a simple bicubic lattice it has been shown that the polarizabilities entering the Clausius-Mossotti relations are those of single oscillators interacting with the free-space vacuum field. We showed that as a consequence of the local field corrections a radiatively broadened medium with overlapping electric and magnetic resonances becomes lossless with a real part of the refractive index approaching the value −2 in the high-density limit. The latter could provide an interesting avenue to construct artificial materials with negative refraction and low losses.

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