Local field in finite-size metamaterials
application to composites of dielectrics and metal nanoparticles

Bordo, Vladimir

Published in:
Physical Review B

DOI:
10.1103/PhysRevB.97.115410

Publication date:
2018

Document version
Publisher's PDF, also known as Version of record

Citation for published version (APA):

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal

Take down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.
Local field in finite-size metamaterials: Application to composites of dielectrics and metal nanoparticles

V. G. Bordo*

NanoSyd, Mads Clausen Institute, Syddansk Universitet, Alson 2, DK-6400 Sønderborg, Denmark

(Received 27 December 2017; published 9 March 2018)

The theory of the optical response of a metamaterial slab which is represented by metal nanoparticles embedded in a dielectric matrix is developed. It is demonstrated that the account of the reflections from the slab boundaries essentially modifies the local field in the slab and leads to the anisotropy and spatial dispersion of its dielectric function as well as to the emergence of modes which do not exist in an infinite metamaterial. It is shown that these features introduce the existence of self-excited normal waves (polaritons) and mechanical excitons (polarization waves). These findings reveal that the metamaterial slab can be regarded as an active device (“plasmonic oscillator”) which generates sustained polaritons in the presence of dissipation. A relation of this effect with the phenomenon of a plasmonic blackbody or perfect absorber, observed in such structures, is discussed and a possible mechanism of this phenomenon is proposed.

DOI: 10.1103/PhysRevB.97.115410

I. INTRODUCTION

Optical metamaterials—artificial materials which contain nanoscale inclusions—have received tremendous attention due to their striking potential applications in diverse fields of science and engineering [1]. The characteristic scale of their structure is much smaller than the wavelength of interest that allows one to describe the optical response of metamaterials in terms of the effective dielectric function of the macroscopically uniform medium (effective medium).

One of the realizations of the metamaterials is a composite of dielectrics and metal nanoparticles (NPs) which, on the one hand, is important from the viewpoint of applications [2–5] and, on the other hand, has a simple structure suitable for modeling. The effective dielectric function of such a material, \( \epsilon \), is described by the Maxwell Garnett approximation [6,7] which relates it with the dielectric functions of the metal particles, \( \epsilon_p \), and the host dielectric, \( \epsilon_h \).

Maxwell Garnett assumed that the radius of the metal spheres, \( R \), is small compared with the wavelength of the incident light, \( \lambda \). This allowed him to replace the spheres by point dipoles with the dipole moments given by \( p = \alpha \mathbf{E} \) with \( \alpha \) being the sphere polarizability and \( \mathbf{E} \) being the acting periodic electric field. In such a formulation the problem is reduced to the analysis of the local field in a substance, represented by an ensemble of atoms or molecules located in vacuum, given by Lorentz and Lorenz earlier [8].

The Lorentz-Lorenz approach assumes that the medium under consideration has dimensions much larger than the wavelength of light. It can be, however, applied to thin films if the incident field is parallel to the film surfaces [6]. The dielectric function in the orthogonal direction may be different, so the film behaves optically like a uniaxial crystal. The reason for this feature is the lack of the dipole distribution isotropy in a thin film. A similar effect occurs in the boundary layer of extended materials where the Lorentz local field differs from its value in the bulk [9].

Turning to the case of a composite of dielectrics and metal nanoparticles, one should introduce a host medium which is different from vacuum that is usually done by making the substitutions \( \epsilon \rightarrow \epsilon/\epsilon_h \) and \( \epsilon_p \rightarrow \epsilon_p/\epsilon_h \) [7]. In a finite-size metamaterial this introduces also a boundary between the host medium and surrounding vacuum.

The influence of such a boundary is twofold. First, it breaks the isotropy of the dipole environment in the vicinity of the boundary as it was mentioned above. This effect was rigorously treated, for example, in the electromagnetic response of a metamaterial slab modeled by a regular array of point dipoles [10]. Second, the light field scattered by the dipoles is partly reflected from the boundary back into the metamaterial bulk that modifies the local field as well. The latter effect, to the best of our knowledge, has not been so far discussed in the literature on the effective medium theory. It can be neglected in the metamaterial bulk if the reflected field is rapidly attenuated due to the absorption in the host material. However, this is obviously not the case if the host material is transparent in the frequency range where the nanoparticles scatter light (i.e., within their localized surface-plasmon polariton band) or if the metamaterial slab thickness is less than or comparable to the light absorption length.

In the present paper, we investigate the influence of the reflections from the interface of a host material and surrounding medium on the electromagnetic response of a metal-nanoparticles–dielectrics composite slab. We demonstrate that this effect introduces both anisotropy and spatial dispersion in the optical response of an isotropic and dispersionless material [we neglect here the effect of the spatial dispersion in the nanoparticle optical response which has an order of \( (R/\lambda)^2 \); see Refs. [11,12]]. We determine the corresponding dielectric function and investigate the dispersion of both the normal waves and mechanical excitons in such a slab (we follow the terminology adopted in Ref. [11]).

*brordo@mci.sdu.dk
The paper is organized as follows. In Sec. II, the integral equation which determines the local field in the slab is derived and the nonlocal dielectric function is deduced. In Secs. III and IV, the dispersion relations are obtained for both the transverse and longitudinal normal waves, respectively. In Sec. V, the mechanical excitons and the polarization waves are introduced and analyzed. In Sec. VI, the developed theory is illustrated by some numerical calculations for the metamaterial slabs enclosed either by vacuum or by a metal. In Sec. VII the obtained results as well as their relation with the phenomenon of a perfect absorber are discussed. Section VIII summarizes the main results of the paper.

II. LOCAL-FIELD EQUATIONS

Let us consider an infinite slab of a nonmagnetic metamaterial of thickness $d$ which is represented by NPs embedded into a dielectric matrix (host material) with the dielectric function $\epsilon_m$ (see Fig. 1). We choose the $z$ axis of the coordinate system along the normal to the slab surfaces with the origin located at the midpoint between them. We assume that all NPs are spherical particles of radius $R$ with the dielectric function $\epsilon_p(\omega)$ and they are distributed randomly throughout the host material with the number density $N$. Then the polarization of NPs at the frequency $\omega$, $P(r, \omega)$, is found as

$$P(r, \omega) = N\alpha(\omega)E(r, \omega)$$

with

$$\alpha(\omega) = \frac{\epsilon_p(\omega) - \epsilon_h}{\epsilon_p(\omega) + 2\epsilon_h}$$

being the polarizability of a spherical nanoparticle embedded into the host material (in Gaussian units) [13] and $E(r, \omega)$ being the local field.

In what follows we assume that the NPs are metallic and their dielectric function is described by the Drude model

$$\epsilon_p(\omega) = \epsilon_{\infty} - \frac{\omega_p^2}{\omega(\omega + i\Gamma)},$$

where $\omega_p$ is the plasma frequency, $\Gamma$ is the relaxation constant, and $\epsilon_{\infty}$ is the offset which takes into account the interband transitions [1].

The local field acting on a given nanoparticle is different from the mean field which results from the averaging over a volume containing a large number of NPs and which enters the macroscopic Maxwell equations. It is obtained as a sum of the mean field, $E(r, \omega)$, and the field of all other NPs distributed throughout the slab volume $V$ and can be written in the form

$$E'(r, \omega) = E(r, \omega) + \int_{V'} \tilde{F}(r', \omega)P(r', \omega)dr'.$$

where we have formally assumed a continuous distribution of the NP polarization and $V'$ denotes the slab volume after deduction of a small volume around the given nanoparticle that excludes the self-action of the nanoparticle. The field susceptibility tensor, $\bar{F}(r, r'; \omega)$, relates the electric field at the point $r$ generated by a classical dipole, oscillating at frequency $\omega$, with the dipole moment itself, located at $r'$ [14]. This quantity is obtained from the Green’s function of the vector wave equation and can be decomposed into direct and reflected contributions as follows:

$$\tilde{F}(r, r'; \omega) = \tilde{F}^0(r, r'; \omega) + \tilde{F}^R(r, r'; \omega).$$

Here the first term results from the dipole field of NPs and its contribution to Eq. (4) is found from the Lorentz relation as (in Gaussian units) [1,8,15]

$$\int_{V'} \tilde{F}^0(r, r'; \omega)P(r', \omega)dr' = \frac{4\pi}{3\epsilon_h}P(r, \omega).$$

The second term in Eq. (5) originates from the dipole fields of NPs reflected from the metamaterial slab boundaries. For a dipole located between two parallel surfaces it is derived in Ref. [16].

Now, substituting Eq. (4) into Eq. (1) and taking into account Eq. (6), one finds a relation between the polarization of NPs and the mean field in the form of an integral equation:

$$P(r, \omega) - \chi(\omega) \int_V \tilde{F}^R(r, r'; \omega)P(r', \omega)dr' = \chi(\omega)E(r, \omega),$$

where

$$\chi(\omega) = \frac{N\alpha(\omega)}{1 - (4\pi/3\epsilon_h)N\alpha(\omega)}$$

is the linear susceptibility of NPs [8]. The latter quantity is proportional to the volume fraction of NPs:

$$f = \frac{4\pi}{3}R^3N.$$

For the further analysis it is convenient to introduce the spatial Fourier components of both the field and polarization:

$$E(r, \omega; z) = \frac{1}{(2\pi)^2} \int k||E(k||, \omega; z)ei\mathbf{k}_||r||dk||,$$

$$P(r, \omega; z) = \frac{1}{(2\pi)^2} \int k||P(k||, \omega; z)ei\mathbf{k}_||r||dk||,$$

where $r$ is the projection of the radius vector $r$ onto the $xy$ plane and $k||$ is the wave vector in the $xy$ plane. Due to the translational invariance of the structure in the $xy$ plane the field susceptibility tensor depends on the difference $r|| = r||'$. 

FIG. 1. The structure under consideration. The metamaterial slab is represented by metal nanoparticles (blue balls) embedded into a transparent dielectric which is enclosed by the material with the dielectric function $\epsilon_m$. 

115410-2
and is represented as follows [16]:

$$\bar{F}^R(\mathbf{r}, \mathbf{r}'; \omega) = \frac{1}{(2\pi)^2} \int \bar{F}^R(\mathbf{k}_\parallel, \omega; z, z')e^{i\mathbf{k}_\parallel \cdot (\mathbf{r} - \mathbf{r}')}d\mathbf{k}_\parallel, \quad (12)$$

where the Fourier transform can be split into the partial downward- and upward-propagating waves as

$$\bar{F}^R(k_x, \omega; z, z') = \bar{F}^+(k_x, \omega; z)e^{-ik_zz'} + \bar{F}^-(k_x, \omega; z)e^{ik_zz'} \quad (13)$$

with $k_z$ being the $z$ component of the wave vector in the metamaterial slab. In view of the isotropy in the $xy$ plane the Fourier transform here does not depend on the direction of the vector $\mathbf{k}_1$.

The substitution of Eqs. (10)–(12) into Eq. (7) leads to the integral equation for the Fourier transforms:

$$\mathbf{P}(k_x, \omega; z) - \chi(\omega) \int_{-d/2}^{d/2} \bar{F}^R(k_x, \omega; z, z')d\mathbf{P}(k_x, \omega; z')dz' = \chi(\omega)\mathbf{E}(k_x, \omega; z). \quad (14)$$

As far as the structure under consideration is invariant with respect to the inversion of the $z$ coordinate, both the field and polarization components can be chosen to have a certain parity and the components with different parities do not mix with each other. Therefore in what follows we consider either even or odd components relative to the inversion $z \to -z$ and denote them by the superscripts $e$ and $o$, respectively.

Let us represent the kernel in the integral equation (14) in the form

$$\bar{F}^R(k_x, \omega; z, z') = \bar{F}^+(k_x, \omega; z') \cos k_zz + \bar{F}^o(k_x, \omega; z') \sin k_zz, \quad (15)$$

where

$$\bar{F}^+(k_x, \omega; z') = \bar{F}^+(k_x, \omega; z') + \bar{F}^-(k_x, \omega; z'), \quad (16)$$

$$\bar{F}^o(k_x, \omega; z') = i[\bar{F}^+(k_x, \omega; z') - \bar{F}^-(k_x, \omega; z')]. \quad (17)$$

Then the equations for even and odd modes are split as follows:

$$\mathbf{P}^e(k_x, \omega; z) - \chi(\omega) \cos k_zz \int_{-d/2}^{d/2} \bar{F}^+(k_x, \omega; z')\mathbf{P}^e(k_x, \omega; z')dz' = \chi(\omega)\mathbf{E}^e(k_x, \omega; z), \quad (18)$$

$$\mathbf{P}^o(k_x, \omega; z) - \chi(\omega) \sin k_zz \int_{-d/2}^{d/2} \bar{F}^o(k_x, \omega; z')\mathbf{P}^o(k_x, \omega; z')dz' = \chi(\omega)\mathbf{E}^o(k_x, \omega; z). \quad (19)$$

The analysis of the parity of the elements of matrices $\bar{F}^e(\omega) (\alpha = e, o)$ reveals that the diagonal elements of $\bar{F}^e(\omega)$ and the nondiagonal elements of $\bar{F}^o(\omega)$ are even functions of $\omega$, whereas the nondiagonal elements of $\bar{F}^e(\omega)$ and the diagonal elements of $\bar{F}^o(\omega)$ are odd functions of $\omega$. As a consequence, the integral terms in Eqs. (18) and (19) are nonzero only for the diagonal elements of $\bar{F}^e(\omega)$. This leads to the scalar form of those equations ($j = x, y, z$):

$$P^e_j(k_x, \omega; z) - \chi(\omega) \cos k_zz \int_{-d/2}^{d/2} F^e_j(k_x, \omega; z')P^e_j(k_x, \omega; z')dz' = \chi(\omega)E^e_j(k_x, \omega; z), \quad (20)$$

$$P^o_j(k_x, \omega; z) - \chi(\omega) \sin k_zz \int_{-d/2}^{d/2} F^o_j(k_x, \omega; z')P^o_j(k_x, \omega; z')dz' = \chi(\omega)E^o_j(k_x, \omega; z). \quad (21)$$

The even and odd modes of the mean field have the forms

$$E^e_j(k_x, \omega; z) = E^e_j(k_x, \omega) \cos k_zz, \quad (22)$$

$$E^o_j(k_x, \omega; z) = E^o_j(k_x, \omega) \sin k_zz, \quad (23)$$

respectively, which lead to the solutions

$$P^e_j(k_x, \omega; z) = P^e_j(k_x, \omega) \cos k_zz, \quad (24)$$

$$P^o_j(k_x, \omega; z) = P^o_j(k_x, \omega) \sin k_zz \quad (25)$$

with

$$\tilde{P}^e_j(k_x, \omega) = \eta^e_{jj}(k_x, \omega)E^e_j(k_x, \omega), \quad \alpha = e, o. \quad (26)$$

Here the quantities

$$\eta^o_{jj}(k_x, \omega) = \frac{\chi(\omega)}{1 - \chi(\omega)G^o_{jj}(k_x, \omega)} \quad (27)$$

with

$$G^e_{jj}(k_x, \omega) = \int_{-d/2}^{d/2} F^e_{jj}(k_x, \omega; z) \cos k_zzdz \quad (28)$$

and

$$G^o_{jj}(k_x, \omega) = \int_{-d/2}^{d/2} F^o_{jj}(k_x, \omega; z) \sin k_zzdz \quad (29)$$

can be regarded as the tensor components of the linear susceptibility of the metamaterial of finite thickness $d$. Neglecting the reflections from the boundaries ($G^o_{jj} = 0$) they are reduced to the ordinary susceptibility $\chi(\omega)$. However, if the reflections cannot be neglected they introduce both anisotropy and spatial dispersion (dependence on $k_z$) in the optical response of the metamaterial. In addition, the effective susceptibilities $\eta^e_{jj}(k_x, \omega)$ contain resonances where $\chi(\omega) = 1/G^o_{jj}(k_x, \omega)$ which do not exist in an infinite metamaterial.

It should be noted that the quantities $G^o_{jj}(k_x, \omega)$ in the denominator of Eq. (27) depend of the wave-vector component $k_z$ in the metamaterial which is dictated in turn by the effective linear susceptibilities $\eta^e_{jj}(k_x, \omega)$ in all other ways, Eq. (27) should be understood as a self-consistent equation which can be solved, for example, by iterations. In what follows we assume that the volume fraction of NPs is small, $f \ll 1$. Therefore a good enough approximation is provided by the first iteration of Eq. (27) where the quantity $k_z$ in the integrand of $G^o_{jj}(k_x, \omega)$ is replaced by its value in the slab without NPs:

$$k = \sqrt{\frac{\omega^2}{c^2} - \varepsilon_b - k_{\parallel}^2} \quad (30)$$

with $c$ being the speed of light in vacuum. The corresponding quantities $\tilde{G}^o_{jj}(k_x, \omega)$ are given in the Appendix.

One can also introduce the tensors of the dielectric function,
\( \varepsilon_e^{\alpha}(k_\parallel,\omega) \), for both even and odd modes through the relation
\[
\tilde{D}_i^{\alpha}(k_\parallel,\omega) = \varepsilon_e^{\alpha}\tilde{E}_i^{\alpha}(k_\parallel,\omega) + 4\pi \dot{P}_{\parallel i}^{\alpha}(k_\parallel,\omega)
\]
\[
= \sum_j \varepsilon_j^{\alpha}(k_\parallel,\omega)\tilde{E}_j^{\alpha}(k_\parallel,\omega).
\]  
(31)

As it follows from Eq. (26), these tensors are diagonal and turn into even and odd modes with the use of the substitutions
\[
e^{\alpha}_{jj}(k_\parallel,\omega) = \varepsilon_h + 4\pi \eta_j^{\alpha}(k_\parallel,\omega).
\]
(32)

**III. TRANSVERSE NORMAL WAVES**

In this section, we investigate the solutions of homogeneous Maxwell equations (normal waves or polaritons) in the metamaterial (33). Let us choose the z axis along the wave propagation direction specified by the wave vector \( \mathbf{k} \). Then the field components do not depend on the x coordinate because of the translational invariance along this direction. As a result, the solutions of Maxwell’s equations
\[
\nabla \times \mathbf{H}(\mathbf{r},\omega) = -\frac{i\omega}{c}\mathbf{D}(\mathbf{r},\omega),
\]
(33)
\[
\nabla \times \mathbf{E}(\mathbf{r},\omega) = \frac{i\omega}{c}\mathbf{H}(\mathbf{r},\omega)
\]
(34)
are split into TE modes with non-zero-field components \( E_x, H_y \), and \( H_x \) and TM modes with non-zero-field components \( E_x, E_z \), and \( H_y \). The TE and TM modes can be split in their turn into even and odd modes with the use of the substitutions
\[
E_x^{\parallel}(\mathbf{r},\omega) = \tilde{E}_x^{\parallel}(k_\parallel,\omega)e^{ik_zz}\cos k_zz,
\]
(35)
\[
E_x^{\perp}(\mathbf{r},\omega) = \tilde{E}_x^{\perp}(k_\parallel,\omega)e^{ik_zz}\sin k_zz,
\]
(36)
and
\[
H_y^{\parallel}(\mathbf{r},\omega) = \tilde{H}_y^{\parallel}(k_\parallel,\omega)e^{ik_zz}\cos k_zz,
\]
(37)
\[
H_y^{\perp}(\mathbf{r},\omega) = \tilde{H}_y^{\perp}(k_\parallel,\omega)e^{ik_zz}\sin k_zz,
\]
(38)
respectively. Let us note that we define here the parity of the mode as the parity of the transverse field component parallel to the boundaries (\( E_y \) for TE polarization and \( H_y \) for TM polarization).

The condition of existence of nontrivial solutions of Maxwell’s equations (33) and (34) leads to the following dispersion relations:
\[
k_\parallel^2 + k_z^2 = \left(\frac{\omega}{c}\right)^2 \varepsilon_\parallel(TE \text{ even modes}),
\]
(39)
\[
k_\parallel^2 + k_z^2 = \left(\frac{\omega}{c}\right)^2 \varepsilon_\parallel(TE \text{ odd modes}),
\]
(40)
\[
\varepsilon_\parallel k_\parallel^2 + \varepsilon_z k_z^2 = \left(\frac{\omega}{c}\right)^2 \varepsilon_\parallel(TE \text{ even modes}),
\]
(41)
\[
\varepsilon_\parallel k_\parallel^2 + \varepsilon_z k_z^2 = \left(\frac{\omega}{c}\right)^2 \varepsilon_\parallel(TE \text{ odd modes}),
\]
(42)

The continuity of the tangential field components at the slab boundaries \( z = \pm d/2 \) imposes additional restrictions for the allowed values of \( k_z \):
\[
k_z \tan \left(\frac{k_zd}{2}\right) = \alpha \quad (\text{TE even modes}),
\]
(43)
\[
k_z \cot \left(\frac{k_zd}{2}\right) = -\alpha \quad (\text{TE odd modes}),
\]
(44)
\[
k_z \tan \left(\frac{k_zd}{2}\right) = \frac{\alpha}{\varepsilon_m} \quad (\text{TM even modes}),
\]
(45)
\[
k_z \cot \left(\frac{k_zd}{2}\right) = -\alpha \quad (\text{TM odd modes}),
\]
(46)
with
\[
\alpha = \sqrt{k_\parallel^2 - \left(\frac{\omega}{c}\right)^2 \varepsilon_m},
\]
(47)
which formally coincide with the ordinary dispersion relations for the waveguide modes [17].

**IV. LONGITUDINAL NORMAL WAVES**

Among the TM modes considered in the previous section, there is a specific solution for which the electric-field vector \( \mathbf{E} \) is parallel to \( \mathbf{k}_\parallel \) (\( E_z = 0 \)). This solution is only possible if \( E_z \) does not depend on the \( z \) coordinate (\( k_z = 0 \)) and \( H_z \equiv 0 \). The consistency with Eq. (33) requires that
\[
\varepsilon_{xx}(k_\parallel,\omega) = 0,
\]
(48)
which determines the dispersion relation of such a longitudinal mode. [Recently the materials for which the condition (48) holds received the name epsilon-near-zero materials; see Ref. [18] for details.]

As far as the quantity \( \dot{G}_x(k_\parallel,\omega) = 0 \) when \( \kappa = 0 \) (see the Appendix), the reflections from the boundaries do not influence the longitudinal mode dispersion in the adopted approximation and it is reduced to
\[
\varepsilon_h + 4\pi \chi(\omega) = 0.
\]
(49)

Assuming in the Drude model, Eq. (3), \( \Gamma \ll \omega \) and a small volume fraction of NPs, \( f \), one finds the solution of Eq. (49) as follows:
\[
\omega_1 \approx \omega_{\text{SPP}} \left(1 + \frac{\varepsilon_h}{\varepsilon_\infty + 2\varepsilon_h}f\right) - i\frac{\Gamma}{2},
\]
(50)
where
\[
\omega_{\text{SPP}} = \frac{\omega_p}{\sqrt{\varepsilon_\infty + 2\varepsilon_h}}
\]
(51)
is the frequency of the localized surface-plasmon polariton supported by a single nanoparticle. The real part in Eq. (50) gives the frequency of the longitudinal mode which originates from the collective electron density oscillations of the ensemble of NPs (plasmons), whereas the imaginary part provides its relaxation rate.

Let us note that the consideration of the longitudinal mode propagating along the \( z \) axis (\( E_z = 0, k_\parallel = 0 \)) leads to the same results, Eqs. (49) and (50).
V. MECHANICAL EXCITONS

The set of equations
\[ \sum_j \epsilon_{ij}^{-1}(k_j, \omega) D_j(k_j, \omega) = E_i(k_j, \omega) = 0 \] (52)

allows a nontrivial solution \((D \neq 0)\) in the absence of the macroscopic field \((E = 0)\) if
\[ \det \left\{ \epsilon_{ij}^{-1}(k_1, \omega) \right\} = [\epsilon_{xx}(k_1, \omega)\epsilon_{yy}(k_1, \omega)\epsilon_{zz}(k_1, \omega)]^{-1} = 0. \] (53)

Such solutions are known as mechanical excitons \([11]\) or polarization eigenmodes \([10]\). These excitations correspond to the poles of the dielectric function tensor and support a nonzero polarization of the medium. \(P\), without a macroscopic field. If, in addition, \(P\) is perpendicular to \(k_1\), the mechanical excitons are called the polarization waves.

In the metamaterial slab the even and odd mechanical excitons are determined by the poles of the effective susceptibilities \(\eta_j^e(k_1, \omega)\) and \(\eta_j^o(k_1, \omega)\), respectively \([\text{see Eq. (27)}]\). Among them, the poles of \(\eta_j^o(k_1, \omega)\) and \(\eta_j^e(k_1, \omega)\) correspond to the polarization waves.

One of the poles which is common for all polarizations is the pole of the linear susceptibility \(\omega_0\), \(\omega_1\), corresponding to the transverse oscillations of the NP polarization. Assuming \(\Gamma \ll \omega\) and \(f \ll 1\) as before one obtains
\[ \omega_1 \approx \omega_{SPP} \left(1 - \frac{3}{2} \frac{\epsilon_h}{\epsilon_\infty + 2\epsilon_h} f \right) - i \frac{\Gamma}{2}. \] (54)

To find the other poles which emerge due to the reflections from the slab boundaries we consider the wave vector \(k_1\) to be a real parameter, whereas we consider the exciton frequency to be complex, \(\omega_j^\alpha = \omega_j^\alpha + i\omega_j^{\alpha*}\) \((\alpha = e, o; j = x, y, z)\). Such a description gives the so-called virtual modes which can manifest themselves in light scattering with \(\omega_j^{\alpha*}\) being the central frequency of the mode and \(2 | \omega_j^\alpha | \) being its width or the inverse of the lifetime \([19]\). In the same approximation as before the virtual modes satisfy the equations
\[ \omega_j^\alpha (\omega_j^\alpha + i \Gamma) \approx \omega_{SPP}^2 \left[ 1 - \frac{3\epsilon_h f}{\epsilon_\infty + 2\epsilon_h} \left(1 + \frac{3\epsilon_h}{4\pi} \tilde{G}_j(k_1, \omega_j^\alpha) \right) \right]. \] (55)

where the quantities \(\tilde{G}_j(k_1, \omega)\) are given in the Appendix. The latter equation can be solved by iterations and because of the condition \(f \ll 1\) already the first iteration provides a good approximation. In the vicinity of the frequency \(\omega_{SPP}\) one obtains
\[ \omega_j^\alpha(k_1) \approx \omega_{SPP} \left(1 - \frac{3}{2} \frac{\epsilon_h f}{\epsilon_\infty + 2\epsilon_h} \right) \left[ 1 + \frac{3\epsilon_h}{4\pi} \text{Re} \tilde{G}_j(k_1, \omega_{SPP}) \right], \] (56)

\[ \omega_j^{\alpha*}(k_1) \approx -\frac{\Gamma}{2} - \frac{9}{8\pi} \frac{\omega_{SPP}}{\epsilon_\infty + 2\epsilon_h} \text{Im} \tilde{G}_j(k_1, \omega_{SPP}). \] (57)

A remarkable conclusion follows from the analysis of Eq. (57) which determines the linewidth of the virtual modes. In the range of the wave vectors where \(\text{Im} \tilde{G}_j(k_1, \omega_{SPP})\) is negative and, in addition,
\[ \frac{9}{8\pi} \frac{\omega_{SPP}}{\epsilon_\infty + 2\epsilon_h} \text{Im} \tilde{G}_j(k_1, \omega_{SPP}) > \frac{\Gamma}{2}, \] (58)

the imaginary part of the mode frequency becomes positive, which implies an exponential increase of the mode amplitude with time. Such a process is only possible if the energy for the polarization of nanoparticles is supplied from an external source. It corresponds to a self-excitation of the mode and can be understood in terms of a positive feedback loop provided by the reflective boundaries \([20,21]\). In this case Eq. (58) formulates the threshold condition for the mode self-excitation (generation).

VI. NUMERICAL RESULTS

We illustrate the above theory with some numerical calculations carried out for a composite material represented by gold NPs embedded into a dielectric with \(\epsilon_h = 1.53^2\). The parameters for \(\epsilon_p\) in the Drude model, Eq. (3), are as

- \(\omega_h = 532, \epsilon_h = 150 \text{ nm}, \Gamma = 0.203 \times 10^{15} \text{ s}^{-1}\), and \(f = 0.01\). The superscript in the mode notation indicates the parity of the mode, while the subscript indicates either the lower \((l)\) or upper \((u)\) branch.
follows: $\epsilon_\infty = 9$, $\omega_p = 13.8 \times 10^{15} \text{ s}^{-1}$, and $\Gamma \approx \gamma + v_F/R$ with $\gamma = 0.11 \times 10^{15} \text{ s}^{-1}$ and $v_F = 1.4 \times 10^8 \text{ cm/s}$ and the NP radius $R$ is taken to be 15 nm [1]. To elucidate the role of the bordering material we investigate two cases: (i) when the metamaterial slab is suspended in vacuum and (ii) when it is enclosed between two semi-infinite metals.

**A. Metamaterial slab in vacuum**

First, to have a reference model system, we consider the dispersion curves of the normal waves in the slab neglecting the reflections from the boundaries. In this approach the dielectric function of the slab is isotropic and local and is reduced to $\epsilon(\omega) = \epsilon_h + 4\pi\chi(\omega)$ with the linear susceptibility $\chi(\omega)$ given by Eq. (8). These curves plotted for the slab thickness $d = 150 \text{ nm}$ are shown in Fig. 2. Their behavior is well known for an isotropic medium with a dielectric function which has a single resonance [11]. Namely, the dispersion curves for both polarizations can be viewed as a result of avoided crossing between the waveguide modes and the frequency band corresponding to the localized surface-plasmon polariton resonance in NPs [11]. For the given thickness only two waveguide modes (even TE and TM modes) can exist while the other modes are cut off. The vacuum light line $\omega = c k_\parallel$ separates the region of the radiative modes (to the left from the line) and the region of the nonradiative modes (to the right from the line). When $k_\parallel \to 0$ the upper branches tend to the frequency of the longitudinal mode, $\omega_\parallel$, and the lower branches tend to zero frequency. When $k_\parallel \to \infty$ the lower branches tend to the frequency of the transverse oscillations, $\omega_\perp$. Thus, there is a frequency gap $\Delta = \omega_\parallel - \omega_\perp$, where no polaritons can propagate. The gap width can be estimated from Eqs. (50) and (54) as follows:

$$\Delta \approx 5 \frac{\epsilon_h f}{2 \epsilon_\infty + 2\epsilon_h} \omega_{\text{SPP}},$$

i.e., it is proportional to the volume fraction of NPs. This implies that a composite of dielectrics and metal nanoparticles behaves like a photonic band-gap crystal—a conclusion which is valid for a regular point dipole array as well [10].

The account of reflections from the slab boundaries changes this picture dramatically (see Figs. 3 and 4). Additional modes emerge in both TE ($\text{TE}_{11}^e$ and $\text{TE}_{12}^e$) and TM ($\text{TM}_{11}^e$, $\text{TM}_{12}^e$, and $\text{TM}_{22}^e$) polarizations and some of them have odd parity. The dispersions of the modes which can be thought of as originating from the modes shown in Fig. 2 ($\text{TE}_{11}^e$, $\text{TM}_{11}^e$, $\text{TE}_{12}^e$, $\text{TM}_{12}^e$, and $\text{TM}_{22}^e$) are terminated in the regions where the numerical routine fails to resolve close roots of the dispersion equation.
FIG. 5. Dispersion curves (a, c, e) and the relaxation rates (b, d, f) of the mechanical excitons for a metamaterial slab suspended in vacuum with account of reflections from the boundaries. The parameters are the same as in Fig. 2.

The TM\textsuperscript{e}l\textsubscript{1}, and TM\textsuperscript{e}u\textsubscript{1}) are remarkably modified in the frequency range between ω\textsubscript{⊥} and ω\textsubscript{∥}. The most striking result is that two of the modes (TE\textsuperscript{e}l\textsubscript{1} and TM\textsuperscript{e}l\textsubscript{2}) possess a positive imaginary part in this frequency interval that indicates their self-excited character.

To shed light on the origin of this unusual behavior we investigate further the dispersion of the mechanical excitons in the metamaterial slab. We shall call a mechanical exciton \(j\)-polarized (\(j = x, y, z\)) and denote it as ME\textsuperscript{e}\(\alpha\)\(\beta\) [\(\alpha = e, o\); \(\beta\) specifies the lower (\(l\)) or upper (\(u\)) branch] if it is determined by the pole of the susceptibility \(\eta^{e}_{jj}(k, \omega)\). Figure 5 shows both the dispersion curves and the dependence of the relaxation rates on the wave vector for \(x\)-, \(y\)-, and \(z\)-polarized mechanical excitons. One can notice that the \(x\)-polarized (longitudinal) excitons do not display self-excitation behavior, while some of the \(y\)- and \(z\)-polarized excitons (polarization waves) do. These peculiarities occur when the lower branch of the exciton dispersion curve crosses the frequency band between the frequencies ω\textsubscript{⊥} and ω\textsubscript{∥} (excitons ME\textsuperscript{e}yl\textsubscript{1} and ME\textsuperscript{e}zl\textsubscript{1}). The mechanical excitons being self-excited induce in their turn the self-excited normal waves.
FIG. 6. Dispersion curves (a) and the relaxation rates (b) of the self-excited normal waves and mechanical excitons for a metamaterial slab enclosed between two thick gold films. The parameters are the same as in Fig. 2.

[see Eqs. (18) and (19)]. A comparison of Figs. 3 and 4 with Fig. 5 reveals that the self-excited exciton ME_{yl1} induces the self-excited normal wave TE_{el1}, whereas the self-excited exciton ME_{zl1} induces the self-excited normal wave TM_{el2}. This discussion demonstrates that it is sufficient to investigate the behavior of the mechanical excitons in order to predict the features of the normal waves.

B. Metamaterial slab between two gold films

To investigate the influence of the material bordering the metamaterial slab we have calculated the dispersion of both the self-excited mechanical excitons and normal waves for a slab between two thick gold films (see Fig. 6). Surprisingly, the maximum values of \( \omega'' \) for both excitons and normal waves, which determine their maximum self-excitation rate, are the same as for a slab suspended in vacuum, although gold is a much better reflector than vacuum.

The analysis of the expressions for the quantities \( G^\alpha_{jj}(k_\parallel, \omega) \) (see the Appendix) reveals that the maxima in the dependencies \( \omega''(k_\parallel) \) originate from the singularity in the density of optical states which behaves as \( 1/k_z \) (or \( 1/\kappa \) in our approximation) [14,22]. This singularity depends on neither the field polarization nor the reflection coefficient at the boundary and therefore takes place for any surrounding medium. In what follows, we restrict ourselves to the discussion of the self-excited excitons in a metamaterial slab enclosed between two metals keeping in mind a possible application for an electrically driven generator [21].

Figure 7 illustrates how the picture is modified when the slab thickness is increased up to 1 \( \mu \)m. It shows only the \( z \)-polarized self-excited mechanical excitons. Their number is increased up to 3 and the range of the wave vectors where they are self-excited is significantly wider than before. The line \( \alpha = 0 \) in Fig. 7(a) with the parameter \( \alpha \) given by Eq. (47) separates the waveguide modes (below the line) and the modes propagating in the bordering metals (above the line). The shown excitons are therefore localized within the slab and cannot propagate outward.

Figure 8 demonstrates the dependence of the mechanical exciton dispersions on the volume fraction of NPs. One can see that, while the dispersion of the real part of \( \omega \) does not change significantly, the dispersion of its imaginary part modifies dramatically. In particular, the maximum positive value of \( \omega'' \) which determines the rate of self-excitation is very small for small values of \( f \), indicating a threshold value of the volume
oscillators, by feeding some output signal back into the input
as in Fig. 2.

different volume fractions of NPs. The other parameters are the same
for a metamaterial slab enclosed between two thick gold films and for
self-excited mechanical exciton denoted in Fig. 6 as ME

vectors. As it was discussed in Sec. VI A, such excitons excite
excitons have self-excited character in a certain range of wave
fraction for a given slab thickness below which self-excitation
is possible. The increase of \( f \) leads to much larger values
of the self-excitation rate with a much wider range of the wave
vectors where self-excitation is possible.

VII. DISCUSSION

We have demonstrated above that some of the mechanical
excitons have self-excited character in a certain range of wave
vectors. As it was discussed in Sec. VI A, such excitons excite
self-excited polaritons under the same conditions. Such a kind
of normal waves can emerge if the power for them is supplied
from an external source, for example upon illumination of the
metamaterial slab by light. The self-excitation process, which
is also known as self-oscillation or sustained oscillation [23],
amplifies any small field Fourier components \( E(k,\omega; z) \) with
\( k \) being in the self-excitation region thus providing power gain
for them. In this sense, the metamaterial slab can be regarded
as an active device which maintains sustained polaritons in the
presence of dissipation [24, 25]. In our case the role of the output signal is played by the
radiation scattered by nanoparticles, while the input signal is the
field acting on them. In this system the field scattered by
nanoparticles and reflected back to them from the boundaries
forms a feedback loop. In the range of \( k \parallel \), where the quantity
\( \text{Im} G_{j}(k_{\parallel},\omega_{\text{res}}) \) is negative, the phase of the reflected field is
such that it provides a positive feedback. If the rate of this
feedback exceeds the relaxation rate the polariton undergoes self-excitation [see Eq. (58)].

In this context, it is interesting to note that a maser—a
generator of microwave radiation and the precursor of a laser—
was initially called a molecular oscillator, where the term
“oscillator” was adopted from electronics [26, 27]. By analogy,
a dielectric slab containing plasmonic nanoparticles, which
generates sustained polaritons, can be called a “plasmonic
oscillator.”

As far as the self-excited polaritons require a power sup-
ply from the incident light beam, they can exist within the
illuminated spot and cannot propagate too far from it. Their
propagation length outside the spot can slightly depend on the
polarization of the incident light due to the difference in the
group velocities of TE and TM polaritons and can be roughly
estimated as \( L \lesssim (c/\sqrt{\epsilon_{r}})\Gamma^{-1} \). For the parameters used in this
paper above one obtains \( L \lesssim 5 \mu m \).

The range of the wave vectors \( k \parallel \) of self-excited mechanical
excitons falls into the nonradiative region [see Fig. 2(a)] and
therefore they cannot be excited by light incident from vacuum
or air. This obstacle which occurs for smooth boundaries is well
known for surface polaritons and can be overcome with the use
of special configurations exploiting attenuated total reflection
(ATR) [28].

On the other hand, any real surface is rough and even
surfaces of small roughness can provide an increase in \( k \parallel \)
up to 0.03 nm\(^{-1}\) due to light scattering ("Umklapp process")
[28]. Such a scattering process creates an initial self-excited
Fourier component of the NP polarization which, even when
being tiny, experiences a rapid exponential growth on the
time scale of \( \Gamma^{-1} \). This evolution stops when the total power
of the generated waves is balanced by the power of the
incident light beam after deduction of the power absorbed
in the NPs. In other words, in the steady-state regime the
extinction of the incident beam power in the slab is complete.
Being a self-oscillation process, this effect does not depend
on the parameters of the incident radiation which triggers it
[23].

Taking into account that a realistic metamaterial slab has
limited dimensions in the \( xy \) plane, one obtains a quasicontinu-
ous spectrum of Fabry-Perot modes instead of the continuous
parameter \( k \parallel \). These modes have different self-excitation rates
while being fed from a common source. In such a situation,
which is well known for the mode competition in a multimode
laser [29], only the mode which has the largest excitation rate
grows, while the other modes are discriminated. This means
that the mode which survives in the steady-state regime is
determined by the value of \( k_{\parallel}^* \) at which the dependence \( \omega(k) \)
has its maximum. The frequency of this mode can be found
from the dispersion curve as \( \omega^*(k_{\parallel}^*) \). To observe the
generated waves, one can use, for example, a hemispherical
prism attached to the slab. Then the generated radiation will
propagate along a conical surface with the opening angle
given by \( \theta = 2 \sin^{-1}(ck_0^m/\omega\rho n_p) \) with \( n_p > \sqrt{\epsilon_a} \) being the refractive index of the prism.

Let us note that in the same ATR configuration one can observe the other virtual modes which are decaying (i.e., with \( \omega' < 0 \)). In such a case, the ATR spectrum excited by a tunable laser will have the spectral width determined by \( 2|\omega'| \) when the exciting radiation is scanning across the frequency \( \omega'(k_0^d) \), where \( k_0^d \) is fixed by the angle of incidence. On the other hand, one can fix the frequency of the incident laser beam at \( \omega'(k_0^d) \) and vary the angle of incidence \( \theta \) along with \( k_0^d \) in the vicinity of \( k_0^d \). Then the angular spread of the ATR signal will be determined by [19]

\[
\sin^{-1}\left(\frac{k_0^d c}{\omega' - \omega'}\right) \leq \theta \leq \sin^{-1}\left(\frac{k_0^d c}{\omega' + \omega'}\right). \tag{60}
\]

The composites of dielectrics and metal nanoparticles considered in this paper have been a subject of extensive experimental investigations [4,30,31]. Such materials have received the name “plasmonic blackbody” or “perfect absorber” due to extremely high absorption (about 100%) claimed by the authors. It is more correct, however, to talk about high extinction which is deduced from those measurements. The extinction is a sum of the absorbed and scattered power; the latter had been omitted when interpreting the experimental data in the mentioned works.

In the light of the theory developed above the phenomenon of a perfect absorber can be understood as a result of the mechanical excitons’ self-excitation. The excitons induce in their turn self-excited normal waves which are localized within the metamaterial slab and do not radiate outward. This process withdraws the power from the incident light beam very effectively and traps it in the slab. As far as its efficiency is determined primarily by the Umklapp process at the surface roughness, which provides a very broad spatial Fourier spectrum of scattered waves, it does not depend significantly on the incidence angle that leads to an omnidirectional character of the process. As one can see from Fig. 8 that, for relatively large values of the NP volume fraction, the self-excitation region spans a large range of \( k_0^d \) [Fig. 8(b)] which corresponds to a wide spectral interval of the dispersion curve [Fig. 8(a)]. This implies that the self-excitation effect takes place for a broad frequency band of the incident radiation. It only slightly depends on the light polarization as far as the y- and z-polarized excitons can be excited by s- and p-polarized incident light, respectively. As it follows from Fig. 8, the maximum of the self-excitation rate shifts to lower frequencies with the increase of the NP volume fraction—a behavior which is consistent with the observations of Ref. [31].

VIII. CONCLUSION

In this paper, we have developed the theory of the optical response of a metamaterial slab represented by a composite of a dielectric and metal nanoparticles which takes into account the reflections from the boundaries. We have found that the dielectric function of such a slab is anisotropic and nonlocal and contains resonances which do not exist in an infinite metamaterial. We have derived the dispersion relations for both the normal waves and mechanical excitons in the slab.

The numerical calculations carried out for a slab either suspended in vacuum or enclosed between two metals have revealed that there exist normal waves and excitons which are self-excited within a certain range of the wave vectors. The amplitude of such excitations grows exponentially with time if they are fed by an external field. This implies that the metamaterial slab behaves like an active device which maintains sustained polaritons in the presence of dissipation although it does not contain any conventional gain material. These findings confirm the results obtained before [20,21] and can be interpreted as a positive feedback provided by the reflective slab boundaries.

We have demonstrated that this effect is very likely manifested as the phenomenon of a plasmonic blackbody or perfect absorber and suggested a mechanism which explains the experimental observations.

ACKNOWLEDGMENT

The author is grateful to N. Engheta, A. S. Lagutchev, and M. Elbahri for stimulating discussions.

APPENDIX: QUANTITIES \( \tilde{G}^s_{j,\omega}(k_0^d, \omega) \)

The matrix elements \( \tilde{G}^s_{j,\omega}(k_0^d, \omega) \) which are obtained from \( G^s_{j,\omega}(k_0^d, \omega) \), Eqs. (28) and (29), by means of the substitution \( k_z \rightarrow \kappa \) [see Eq. (30)] have the following forms:

\[
\tilde{G}^s_{xs}(k_0^d, \omega) = \frac{2 \pi i}{\sqrt{\varepsilon_h}} k d \left(1 + \frac{\sin \kappa d}{\kappa d}\right) \frac{r_p e^{i \kappa d}}{1 + r_p e^{i \kappa d}}. \tag{A1}
\]

\[
\tilde{G}^s_{ys}(k_0^d, \omega) = \frac{2 \pi i}{\sqrt{\varepsilon_h}} k d \left(1 - \frac{\sin \kappa d}{\kappa d}\right) \frac{r_p e^{i \kappa d}}{1 - r_p e^{i \kappa d}}. \tag{A2}
\]

\[
\tilde{G}^s_{y\gamma}(k_0^d, \omega) = 2 \pi i k d \left(\frac{\omega}{\varepsilon_h}\right)^2 \left(1 + \frac{\sin \kappa d}{\kappa d}\right) \frac{r_p e^{i \kappa d}}{1 - r_p e^{i \kappa d}}. \tag{A3}
\]

\[
\tilde{G}^s_{\gamma\gamma}(k_0^d, \omega) = -2 \pi i k d \left(\frac{\omega}{\varepsilon_h}\right)^2 \left(1 - \frac{\sin \kappa d}{\kappa d}\right) \frac{r_p e^{i \kappa d}}{1 + r_p e^{i \kappa d}}. \tag{A4}
\]

\[
\tilde{G}^s_{zz}(k_0^d, \omega) = \frac{2 \pi i}{\sqrt{\varepsilon_h}} k d \left(\frac{k_0^d}{\kappa}\right)^2 \left(1 + \frac{\sin \kappa d}{\kappa d}\right) \frac{r_p e^{i \kappa d}}{1 - r_p e^{i \kappa d}}. \tag{A5}
\]

\[
\tilde{G}^s_{zz}(k_0^d, \omega) = -\frac{2 \pi i}{\sqrt{\varepsilon_h}} k d \left(\frac{k_0^d}{\kappa}\right)^2 \left(1 - \frac{\sin \kappa d}{\kappa d}\right) \frac{r_p e^{i \kappa d}}{1 + r_p e^{i \kappa d}}. \tag{A6}
\]

Here

\[
r_p = \frac{\epsilon_m \kappa - i \varepsilon_h \alpha}{\epsilon_m \kappa + i \varepsilon_h \alpha} \tag{A7}
\]

and

\[
r_s = \frac{\kappa - i \alpha}{\kappa + i \alpha} \tag{A8}
\]

are the Fresnel reflection coefficients for s- and p-polarized light, respectively, at the interface of the metamaterial slab and bordering material, and the quantity \( \alpha \) is defined by Eq. (47).


[14] Strictly speaking, the relation (6) holds only approximately near the boundaries. See Ref. [9] for details.


[16] See, e.g., C. A. Balanis, *Advanced Engineering Electromagnetics* (Wiley, New York, 1989); Let us note that in this book the definition of the mode parity is different from that adopted in this paper.

[17] I. Liberal and N. Engheta, Nat. Photon. 11, 149 (2017).


[23] See Ref. [24], Sec. 3.4, “Passive versus active devices.”


