Negative permeability and negative refraction in composite systems with finite-size inclusions

A. Smolyakov

Department of Physics and Engineering Physics, University of Saskatchewan, Saskatoon, Saskatchewan, Canada S7N 5A2

E. Fourkal

Department of Radiation Physics, Fox Chase Cancer Center, Philadelphia, Pennsylvania 19111, USA

It is shown that the spatial dispersion in composite systems formed by inclusion of finite size spheres may lead to negative group velocity and, subsequently, to negative refraction. Longitudinal and transverse electromagnetic modes exhibiting negative dispersion are found in the system of finite size charged clouds. It is also shown that similar mechanism due to the electric quadrupole coupling leads to excitation of longitudinal and transverse eigenmodes with negative dispersion in a random set of nonmagnetic metal spheres embedded in a dielectric host. It is shown that the negative refraction related to the spatial dispersion effects may alternatively be viewed as a result of simultaneously negative permittivity and permeability.

I. INTRODUCTION

Metamaterials posses unusual (and fascinating) optical and electromagnetic (EM) properties and their studies have recently attracted a great deal of attention and effort. There are several related but not equivalent phenomena which define many unique properties of metamaterials. The existing literature on metamaterials refers to different approaches in the description and characterization of these media, also resulting in different names being used for such materials: negative-refraction media, negative-index materials, left-handed media, etc. Several recent reviews emphasize various aspects of metamaterials, of which two key phenomena are negative refraction and amplification of evanescent waves. A physical picture of negative refraction can be most easily formulated in terms of the negative group velocity (or negative dispersion) $\partial \omega / \partial \mathbf{k} \sim -\mathbf{k}$, when it is opposite to the direction of the wave vector (phase velocity) [1]. In media with negative group velocity, the energy flow is opposite to the direction of the wave front propagation. As a result, at the interface between a regular (positive-index) material and a metamaterial (negative-index material), the refracted wave ray is deflected into the same side as the incident ray, hence the name of negative refraction. In this approach, the negative-refraction material is characterized only by its dispersion properties. For high-frequency electromagnetic waves, it is possible to fully characterize such dispersive materials with a single dielectric tensor $\widetilde{\boldsymbol{\varepsilon}} = \widetilde{\boldsymbol{\varepsilon}}(\omega, \mathbf{k})$ [2–5] while assuming that $\mu = 1$. An alternative description of negative refraction is based on the notion of the negative refractive index $n = -\sqrt{\varepsilon \mu} < 0$ [6], which requires the conditions $\varepsilon < 0$ and $\mu < 0$ [5,7], thus leading to the names of negative-index and double-negative materials.

In this paper, we investigate dispersive properties of simple composite physical systems consisting of finite-size objects. We show that negative refraction occurs as a result of the dispersion caused by finite-size inclusions in the host medium. One example of such a system is a gas of charged particles of a finite size. These systems are often used to simulate the behavior of plasmas. Since the number of particles in real systems is computationally prohibitive even for modern computers, the real charged particles are approximated by charged clouds of a finite size (particle-in-cell calculations) [8]. In fact, this procedure is identical to the finite-size averaging employed in derivations of the macroscopic electrodynamics equation [9] for a dielectric medium. We show here that in such systems negative refraction occurs as a result of the finite dimension of the interacting clouds (or the finite averaging size of the sampling volume [9]). This finding suggests that other simple systems with electrically active objects of finite size may also exhibit negative refraction. Indeed, by considering a simple system of metal spheres immersed in a host medium, we show that the second-order effects in a/λ (corresponding to the quadrupole moments of the spheres) result in the appearance of additional propagating modes with negative dispersion. To our knowledge this is the simplest example of a negative-refraction medium. Our analysis is primarily based on the characterization of the medium by the dielectric tensor $\widetilde{\boldsymbol{\varepsilon}} = \widetilde{\boldsymbol{\varepsilon}}(\omega, \mathbf{k})$. We also show that physical results based on the dispersive properties of the dielectric tensor with $\mu = 1$ can be equivalently described within the ε-μ approach with simultaneously negative ε and μ.

II. MAGNETIC RESPONSE OF DISPERSIVE MEDIA

Although media with negative μ do not exist in nature, a magnetic response occurs for many nonmagnetic materials. In particular, it has been emphasized that optical magnetism in plasmonic metamaterials is a result of plasmonic (electrostatic) resonance [10,11]. In fact, a magnetic response is a common property of strongly dispersive materials, e.g., plasmas. This fact may have been obscured in part due to a common approach in plasma physics which does not rely on the notion of magnetic permeability. As noted in [2], the notion of magnetic permeability for high-frequency phenomena is not directly related to the density of the magnetic moment of the material. For such cases, it is possible to write Maxwell equations with $\mathbf{B} = \mathbf{H}$ (i.e., $\mu = 1$) so that all currents (including bound currents, normally associated with magnetization) are assigned to the generalized polarization vector $\mathbf{P}$. In this so-called three-field ($\mathbf{E}$, $\mathbf{D}$, $\mathbf{B}$) model, the dielectric response becomes

a tensor dependent on the wave vector $\widetilde{\varepsilon} = \widetilde{\varepsilon}(\omega,\mathbf{k})$. For an isotropic medium, the most general form of this permittivity tensor is

$$\widetilde{\boldsymbol{\varepsilon}}(\omega,\mathbf{k}) = \varepsilon_T(\omega,k)\left(\mathbf{I} - \frac{\mathbf{k} \otimes \mathbf{k}}{k^2}\right) + \varepsilon_L(\omega,k)\frac{\mathbf{k} \otimes \mathbf{k}}{k^2}, \quad (1)$$

and the electric current is determined by $\mathbf{j} = -i\omega[\widetilde{\boldsymbol{\varepsilon}}(\omega,\mathbf{k}) - \mathbf{I}] \cdot \mathbf{E}/4\pi$. As an example, in the absence of an external magnetic field, the components of the dielectric tensor for warm plasma are [3]

$$\varepsilon_T(\omega,k) = 1 - \frac{\omega_p^2}{\omega^2}[1 - W(\omega/kv_T)], \quad (2a)$$

$$\varepsilon_L(\omega,k) = 1 + \frac{k_D^2}{k^2}W(\omega/kv_T), \quad (2b)$$

where W is the plasma dispersion function. Alternatively, the dispersive medium characterized by the tensor (1) can be described by two tensor functions $\boldsymbol{\varepsilon}(\omega,\mathbf{k})$ and $\boldsymbol{\mu}(\omega,\mathbf{k})$ [12]. In this description, the electric current is

$$\mathbf{j} = \frac{\partial \mathbf{P}}{\partial t} + c\nabla \times \mathbf{M}, \quad (3)$$

where the effective polarization vector $\mathbf{P}$ and magnetization vector $\mathbf{M}$ are defined by the expressions

$$\begin{aligned} 4\pi\mathbf{P} &= [\boldsymbol{\varepsilon}(\omega, \mathbf{k}) - \mathbf{I}] \cdot \mathbf{E}, \\ 4\pi\mathbf{M} &= [\mathbf{I} - \boldsymbol{\mu}^{-1}(\omega, \mathbf{k})] \cdot \mathbf{B}. \end{aligned} \quad (4)$$

Thus dispersion effects can be described either by the dielectric tensor (1) or with two tensor functions $\boldsymbol{\varepsilon}(\omega,\mathbf{k})$ and $\boldsymbol{\mu}(\omega,\mathbf{k})$. Both approaches are equivalent and lead to the same physical conclusions. The transition from $\boldsymbol{\varepsilon}(\omega,\mathbf{k})$ and $\boldsymbol{\mu}(\omega,\mathbf{k})$ to $\widetilde{\boldsymbol{\varepsilon}}(\omega,\mathbf{k})$ is unique, while the inverse transformation is not [13,14]. However, in the limit of small wave numbers k, the inverse transformation $\widetilde{\boldsymbol{\varepsilon}}(\omega,\mathbf{k})$ to $\varepsilon(\omega)$ and $\mu(\omega)$ is well defined and can be given as follows [2]:

$$\begin{aligned} \varepsilon(\omega) &= \lim_{k \to 0} \varepsilon_L(\omega,k), \\ \frac{1}{\mu(\omega)} &= 1 + \lim_{k \to 0} \frac{\omega^2}{k^2 c^2}[\varepsilon(\omega) - \varepsilon_T(\omega,k)]. \end{aligned} \quad (5)$$

These relations show that the magnetic response can be viewed merely as a manifestation of the spatial dispersion in the system, which in itself arises not only from the magnetic dipole (and higher-order magnetic-type) moments, but also from those associated with the electric polarization currents. For example, warm plasma in the example (2a) and (2b) can be characterized (in the lowest dispersive order) by the dielectric function $\varepsilon(\omega) = 1 - \omega_p^2/\omega^2$ and magnetic permeability

$$\mu(\omega) = \frac{1}{1 + \omega_p^2 v_T^2/(\omega^2 c^2)}. \quad (6)$$

As one can see, in the $\varepsilon(\omega)$-$\mu(\omega)$ description, the plasma becomes magnetically active and its response is described by $\mu < 1$, which is a known result for the diamagnetic nature of the plasma state. The dispersion relation for the transverse electromagnetic waves in the $\varepsilon(\omega)$-$\mu(\omega)$ approach is $k^2 c^2/\omega^2 = \varepsilon(\omega)\mu(\omega)$. An identical result can be obtained from the approach with $\widetilde{\boldsymbol{\varepsilon}}(\omega,\mathbf{k})$ and $\mu = 1$ in which the

dispersion relation for transverse electromagnetic waves is $k^2 c^2/\omega^2 = \varepsilon_T(\omega,k)$.

III. NEGATIVE PERMEABILITY AND NEGATIVE REFRACTION IN SYSTEMS OF FINITE-SIZED CHARGED CLOUDS

Computer simulation of real plasmas with a large number of electrons and ions requires very high spatial resolution in order to describe the dynamics of particle collisions. This would make such simulations prohibitively slow. However, if one is interested in the collective behavior, effects of the short-range forces are not important and can be smeared out by applying a spatial field-averaging procedure. This procedure is analogous to the averaging procedure employed in the derivation of the macroscopic Maxwell equations [9] and equivalent to the introduction of finite-size particles. It is known as a particle-in-cell (PIC) method in plasma kinetic simulation codes. In this averaging procedure, the charge distribution of a point particle system,

$$\rho(\mathbf{x}) = \sum_i \mathbf{q}_i \delta(\mathbf{x} - \mathbf{x}_i), \quad (7)$$

is averaged with respect to the weight function $S(\mathbf{x})$ [9],

$$\bar{\rho}(\mathbf{x}) = \int d^3x' S(\mathbf{x}')\rho(\mathbf{x} - \mathbf{x}'). \quad (8)$$

In Fourier representation one can write

$$\bar{\rho}(\mathbf{k}) = S(\mathbf{k})\rho(\mathbf{k}). \quad (9)$$

The averaging function $S(\mathbf{k})$ is a smooth localized function in space, which decays to zero at distances larger than the characteristic length scale a. The latter would be a characteristic size of the finite-size object. A typical choice is the Gaussian test function $S(k) \sim e^{-k^2 a^2/2}$, which suppresses spatial fluctuations of the total charge density at high $k > 1/a$, yet retaining its long-wavelength features. In real space, the spatial average of the charge density becomes

$$\bar{\rho}(\mathbf{x}) = \sum_i q_i S(\mathbf{x} - \mathbf{x}_i). \quad (10)$$

The above expression can be recast in the form of the Taylor series expansion corresponding to the multipole expansion [9]. It is easy to see that introduction of particle clouds in the PIC technique is equivalent to the spatial averaging procedure used in the macroscopic electrodynamics of continuous media [9]. Electromagnetic properties of particle-in-cell systems can be studied by using the dielectric response functions calculated by standard methods [8]. The longitudinal and transverse dielectric response functions for the Maxwellian distribution of finite-size particles are [8]

$$\begin{aligned} \varepsilon_l(k,\omega) &= 1 + S(k)^2 \frac{k_D^2}{k^2} W\left(\frac{\omega}{kv_T}\right), \\ \varepsilon_t(k,\omega) &= 1 - S(k)^2 \frac{\omega_p^2}{\omega^2}\left[1 - W\left(\frac{\omega}{kv_T}\right)\right]. \end{aligned} \quad (11)$$

For the Gaussian averaging function $S(k) \sim e^{-k^2 a^2/2}$, the dispersion relation for the longitudinal electrostatic waves in
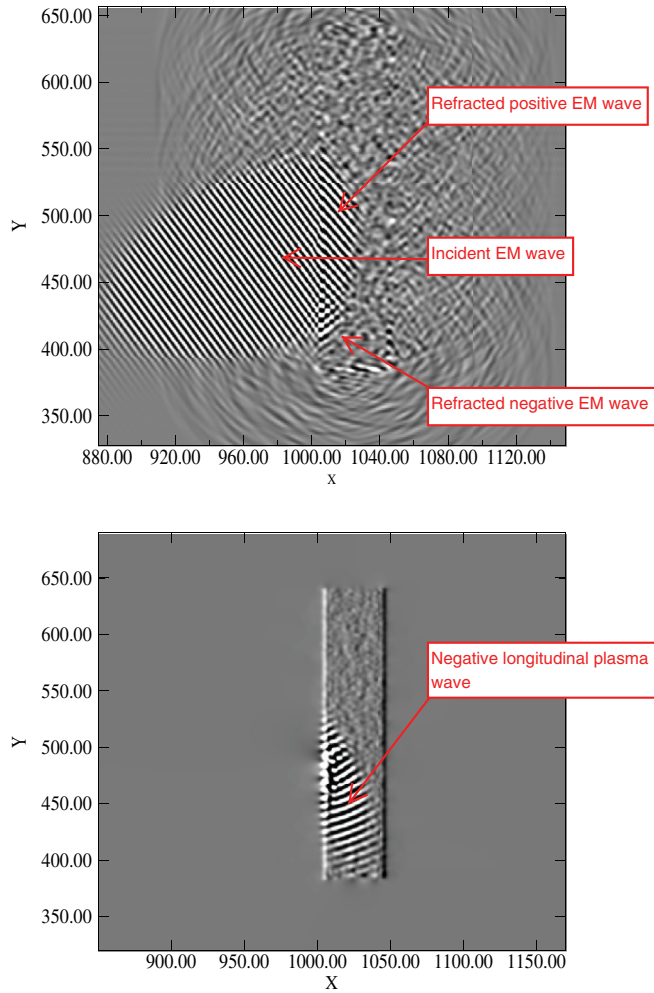
FIG. 1. (Color online) The two-dimensional spatial distribution of the transverse magnetic field $B_z(x,y)$ (the top picture) and longitudinal electric field $E_x(x,y)$ (the bottom picture) obtained from PIC simulations. The incident laser pulse carryer frequency is $\omega_0 = 7.85 \times 10^{14}$ s^{-1}, and the slab's plasma frequency $\omega_p = 8.05 \times 10^{14}$ s^{-1}. The Debye wavelength is $\lambda_D \approx 0.47 \times 10^{-7}$ m and the particle radius $a \approx 4.486 \times 10^{-7}$ m. The spatial coordinates X and Y are normalized to the grid spacing $\Delta \approx 6.3 \times 10^{-7}$ m.

the limit $k v_T/\omega \ll 1$ and $ka \ll 1$ becomes

$$\omega^2 = \omega_p^2 + 3k^2 v_T^2 \left(1 - \frac{1}{3} a^2 k_d^2\right),$$

where $k_D = 1/\lambda_D =$ is the Debye wave number. The group velocity of the plasma wave becomes negative when $a > 3\lambda_D$. A negative plasma wave has been observed in PIC simulations of a laser pulse interaction with a cold plasma slab, shown in Fig. 1 (the bottom picture). In this case, the incident laser pulse with the carrying frequency $\omega_0 < \omega_p$ directly excites a negative-refractive plasma wave.

Analogously, the dispersion relation for the transverse wave is obtained as

$$\omega^2 = \omega_p^2 e^{-k^2 a^2} + k^2 c^2.$$

Taking the limit $ka \ll 1$, one arrives at the condition $a > c/\omega_p$ for the existence of the transverse electromagnetic mode with negative group velocity. Figure 1 (the top picture) shows the snapshot of the spatial distribution of the transverse magnetic field for the case when the laser pulse is incident on the plasma slab. One can clearly see two propagating branches inside the plasma, one of which has a positive dispersion and the other negative.

Substituting expressions (11) into Eq. (5) one readily arrives at the electric permittivity $\varepsilon(\omega) = 1 - \omega_p^2/\omega^2$ and magnetic permeability $\mu(\omega) = [1 - \omega_p^2 a^2/c^2 + \omega_p^2 v_T^2/(\omega^2 c^2)]^{-1}$ of the system of finite-sized charges in the ε-μ description. As one can see, both quantities are negative when $a\omega_p/c > 1$ and $\omega < \omega_p$ ($v_T \ll c$).

IV. NEGATIVE PERMEABILITY AND NEGATIVE REFRACTION IN A SYSTEM OF FINITE-SIZE CONDUCTING SPHERES

In the previous example, negative refraction occurs as a result of the dispersion due to the finite size of the charged clouds. Now we investigate similar effects for a composite material consisting of a random set of finite-size metal spheres embedded in a dielectric host.

Consider a nonmagnetic dielectric sphere of radius a and dielectric constant ε_1 embedded in a medium with dielectric
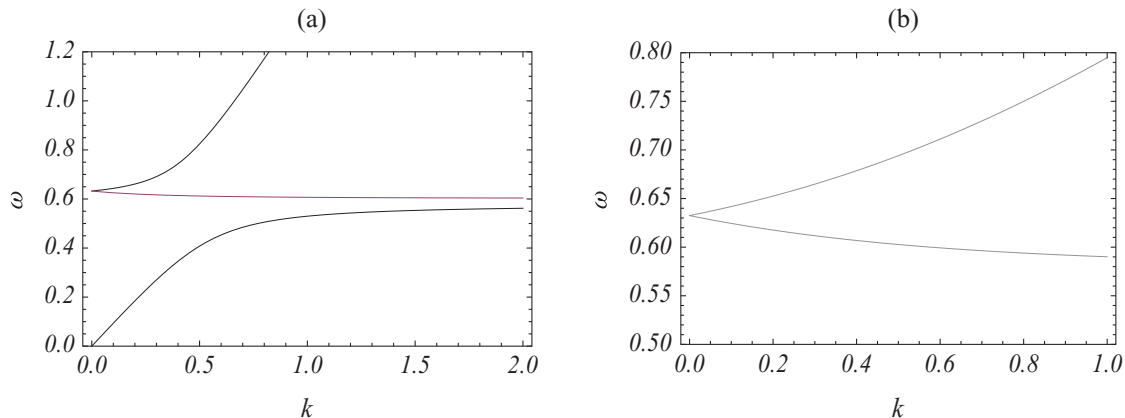

FIG. 2. (Color online) (a) Transverse modes with electric quadrupole and magnetic dipole corrections. The negative-dispersion branch is seen in the gap region; $n_s a^3 = 0.2$, $a = 3c/\omega_p$, where ω_p is the plasma frequency of the metal sphere, ω is normalized to the plasma frequency, and k is in units of c/ω_p. (b) Longitudinal modes with electric quadrupole corrections.

constant ε_2 and magnetic permeability $\mu = 1$. An external inhomogeneous electric field induces a set of multipole moments on the sphere. In the lowest order in $a/\lambda < 1$, the scattered electric field is approximated by its magnitude as well as the first-order spatial gradients, which correspond to the induced magnetic dipole and electric quadrupole moments. The explicit expressions for these moments are [15]

$$\mathbf{p} = \frac{1}{4\pi} \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} a^3 \mathbf{E}, \tag{12}$$

$$q_{ij} = \frac{1}{4\pi} \frac{\varepsilon_1 - \varepsilon_2}{2\varepsilon_1 + 3\varepsilon_2} a^5 \left[\frac{1}{2}(\nabla_j E_i + \nabla_i E_j) - \frac{1}{3}\mathbf{\nabla} \cdot \mathbf{E}\delta_{ij} \right], \tag{13}$$

$$\mathbf{m} = \frac{\omega^2 a^5}{30 c^2}(\varepsilon_1 - \varepsilon_2)\mathbf{B}. \tag{14}$$

Using Eq. (3) together with Eqs. (12), (13), and (14) one obtains the total dielectric tensor in the form of (1) with

$$\varepsilon_L = 1 + \frac{(\varepsilon_1 - \varepsilon_2)n_s a^3}{\varepsilon_1 + 2\varepsilon_2} + \frac{2}{3}\frac{(\varepsilon_1 - \varepsilon_2)n_s a^3}{2\varepsilon_1 + 3\varepsilon_2}k^2 a^2, \tag{15}$$

$$\varepsilon_T = 1 + \frac{(\varepsilon_1 - \varepsilon_2)n_s a^3}{\varepsilon_1 + 2\varepsilon_2} + \frac{1}{2}\frac{(\varepsilon_1 - \varepsilon_2)n_s a^3}{2\varepsilon_1 + 3\varepsilon_2}k^2 a^2$$
$$+ \frac{4\pi (\varepsilon_1 - \varepsilon_2)n_s a^3}{30}k^2 a^2, \tag{16}$$

where n_s is the concentration of the spheres. The third and the fourth terms in the expression for the transverse dielectric function come from the electric quadrupole and the magnetic dipole contributions to the generalized dielectric response tensor, respectively. As expected, both moments are of the same order ($k^2 a^2$ terms) and are responsible for the spatial dispersion. The dispersion relations $\varepsilon_T = k^2 c^2/\omega^2$ and $\varepsilon_L(\omega,k) = 0$ define propagating transverse and longitudinal electromagnetic modes, respectively shown in Figs. 2(a) and 2(b). In each case, there is one mode with negative dispersion. It should be noted here that the existence of negative waves for the system of conducting spheres of the same size is solely due to the contribution of the induced electric quadrupole moment. The induced magnetic dipole moment of the sphere does not contribute to negative dispersion in this particular case, unlike in the example of the composite medium consisting of two sub-lattices of dielectric spherical particles with different radii embedded in a host material [16], where the induced magnetic dipole moment of the bi-sphere structure can take on negative values in a certain frequency range.

Alternatively, the negative-refraction mode can be viewed as a result of negative ε and negative μ defined by
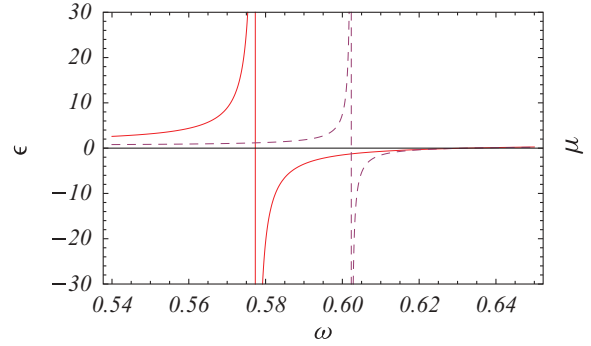

FIG. 3. (Color online) Effective dielectric function $\varepsilon(\omega)$ (solid line) and magnetic permeability $\mu(\omega)$ (dashed line) for a system of spheres in vacuum versus normalized frequency; $n_s a^3 = 0.2$, $a = 3c/\omega_p$. The dielectric permittivity and magnetic permeability are negative for $0.6023 < \omega < 0.6325$, where ω is normalized to the plasma frequency ω_p.

Eq. (5):

$$\varepsilon(\omega) = 1 + \frac{(\varepsilon_1 - \varepsilon_2)n_s a^3}{\varepsilon_1 + 2\varepsilon_2},$$

$$\mu(\omega) = \left(1 - \frac{1}{2}\frac{(\varepsilon_1 - \varepsilon_2)n_s a^3}{2\varepsilon_1 + 3\varepsilon_2}\frac{\omega^2 a^2}{c^2} \right.$$
$$\left. - \frac{4\pi (\varepsilon_1 - \varepsilon_2)n_s a^3 \omega^2 a^2}{30 c^2}\right)^{-1}.$$

Figure 3 shows the electric permittivity $\varepsilon(\omega)$ and the magnetic permeability $\mu(\omega)$ for a system of conducting spheres of radius a in a vacuum. Both functions are negative in the frequency range that exactly corresponds to the region where the negative mode obtained from the dispersion relation $\varepsilon_T - k^2 c^2/\omega^2 = 0$ exists (see Fig. 2).

V. SUMMARY

We have considered several simple systems that exhibit negative-refraction phenomena. We have applied the formalism of the total dielectric tensor to characterize a dispersive medium with opposite directions of the group and phase velocities. The latter is a crucial signature of a negative-refraction medium. Alternatively, such dispersive media can be described by simultaneously negative ε and μ. A system of metal spheres seems to be the simplest configuration exhibiting negative dispersion (negative refraction). The role of the quadrupole resonance in negative dispersion was also noted in Ref. [17]. These negative-refraction modes propagate with low group velocity ($v_{gr} \approx 0.1$), which could be of interest for slow-light devices.

[1] L. I. Mandel'stam, *Lectures on Optics, Relativity and Quantum Mechanics* (Nauka, Moscow, 1972) (in Russian).

[2] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media*, 2nd ed. (Pergamon Press, Oxford, 1984).

[3] S. Ichimaru, *Statistical Plasma Physics. Volume I: Basic Principles* (Addison-Wesley, Reading, MA, 1991).

[4] V. M. Agranovich and Y. N. Gartstein, Phys. Usp. **49**, 1029 (2006).

[5] A. P. Vinogradov, A. V. Dorofeenko, and S. Zouhdi, Phys. Usp. **51**, 485 (2008).

[6] V. G. Veselago, Phys. Usp. **10**, 509 (1968).

[7] A. L. Pokrovsky and A. L. Efros, Solid State Commun. **124**, 283 (2002).

[8] C. K. Birdsall and A. B. Langdon, *Plasma Physics via Computer Simulation* (Taylor and Francis, London, 2005).

[9] J. D. Jackson, *Classical Electrodynamics*, 3rd ed. (John Wiley and Sons, New York, 1999).

[10] Y. A. Urzhumov and G. Shvets, Solid State Commun. **146**, 208 (2008).

[11] A. Alu and N. Engheta, Opt. Express **17**, 5723 (2009).

[12] V. M. Agranovich, Y. R. Shen, R. H. Baughman, and A. A. Zakhidov, Phys. Rev. B **69**, 165112 (2004).

[13] D. A. Kirzhnits, Phys. Usp. **30**, 575 (1987).

[14] A. P. Vinogradov, Phys. Usp. **45**, 331 (2002).

[15] J. A. Stratton, *Electromagnetic Theory*, 1st ed. (McGraw-Hill, New York, 1941).

[16] I. Vendick, O. Vendick, I. Kolmakov, and M. Odit, Opto-Electronics Review **14**, 179 (2006).

[17] D. Han, Y. Kai, K. H. Fung, Z.-Q. Zhang, and C. T. Chan, Phys. Rev. B **79**, 195444 (2009).