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Spatial dispersion and negative refraction of light †

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Abstract. Negative refraction occurs at interfaces as a natural consequence of the negative group velocity of waves in one of the interfacing media. The historical origin of this understanding of the phenomenon is briefly discussed. We consider several physical systems that may exhibit normal electromagnetic waves (polaritons) with negative group velocity at optical frequencies. These systems are analyzed in a unified way provided by the spatial dispersion framework. The framework utilizes the notion of the generalized dielectric tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ representing the electromagnetic response of the medium to perturbations of frequency ω and wave vector k. Polaritons with negative group velocity can exist in media (whether in natural or in artificial meta-materials) with a sufficiently strong spatial dispersion. Our examples include both gyrotropic and nongyrotropic systems, and bulk and surface polariton waves. We also discuss the relation between the spatial dispersion approach and the more familiar, but more restricted, description involving the dielectric permittivity $\varepsilon(\omega)$ and the magnetic permeability $\mu(\omega)$.

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1. Introduction

In this review, the phenomenon of negative refraction is discussed in terms of the dispersion $\omega(\mathbf{k})$ of *polaritons*, which are normal electromagnetic waves propagating in a medium in the vicinity of excitonic resonances. We focus on a macroscopically uniform and isotropic medium with negligible dissipation: in this case, no additional complications arise, and the basic physics of the phenomena under consideration is especially transparent. In other words, we consider bodies with a characteristic size of the order of or greater than the wavelength λ of waves in the medium. In an isotropic medium, the frequency ω depends only on the absolute value $k = |\mathbf{k}|$ of the wave vector $\mathbf{k}$, and therefore the group velocity of the wave packet

$$\mathbf{v}_{\mathrm{g}} = \frac{\mathrm{d}\omega(\mathbf{k})}{\mathrm{d}\mathbf{k}} = \frac{\mathbf{k}}{k}\,\frac{\mathrm{d}\omega(k)}{\mathrm{d}k} \tag{1}$$

is codirected with either $\mathbf{k}$ or $-\mathbf{k}$, depending on the sign of $\mathrm{d}\omega(k)/\mathrm{d}k$. As was noted by L I Mandel'shtam [1–3], the latter case of 'negative group velocity' $\mathrm{d}\omega(k)/\mathrm{d}k < 0$ is related to the phenomenon of negative refraction. The English physicist Arthur Schuster also mentions this possibility in his book [4]. But he considered the region of anomalous dispersion in the vicinity of a resonance, where the definition of the group velocity in (1) is inapplicable.

As is well known (see, e.g., Refs [3, 5–7]), in a medium with *small dissipation*, the energy propagation velocity

coincides with the group velocity, and therefore the energy flux vector (the Poynting vector for electromagnetic waves) is the product

$$\mathbf{S} = U\mathbf{v}_{\mathrm{g}}\,, \tag{2}$$

where U is the time-averaged energy density. In *thermodynamic equilibrium*, $U > 0$, and hence, for waves with negative group velocity, the energy flux vector $\mathbf{S}$ is directed opposite to the wave vector $\mathbf{k}$. The negative refraction of light and all the unusual properties of materials with negative refraction are natural consequences of such a relation between $\mathbf{S}$ and $\mathbf{k}$. In this review, we consider only the negative refraction of electromagnetic waves. However, it was clearly shown by Mandel'shtam (see Section 2.1) that negative refraction is a general property of waves of any nature with a negative group velocity.

We discuss several physical systems in which polaritons with negative group velocity can appear and, consequently, in which one can try to realize the negative refraction (including the optical frequency range). The existence of polaritons with negative group velocity is possible for a medium with a sufficiently strong *spatial dispersion* of its dielectric properties [7–9]. The presence of the spatial dispersion signifies a nonlocal dielectric response; it is manifested by the dependence of the generalized dielectric tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ on the wave vector $\mathbf{k}$ [6, 7].

We show in what follows that the approach based on accounting for spatial dispersion also includes, as a special case, a more familiar approach, which is typically used in describing the negative refraction of light in a medium with simultaneously negative dielectric permittivity, $\varepsilon(\omega) < 0$, and magnetic permeability, $\mu(\omega) < 0$. In connection with such media, Veselago's work [10] is usually referred to, although this case was in fact discussed much earlier in Sivukhin's paper [11] and afterwards in Pafomov's papers [12, 13]. In particular, these works contain the remark on the negative group velocity of waves in such a medium. The negative group velocity branch is clearly seen in Fig. 1. Figure 1a shows the dispersion law $\omega(k)$ of transverse polaritons defined by the well-known expression

$$\omega^2\varepsilon(\omega)\,\mu(\omega) = \omega^2 n^2(\omega) = c^2k^2\,, \tag{3}$$

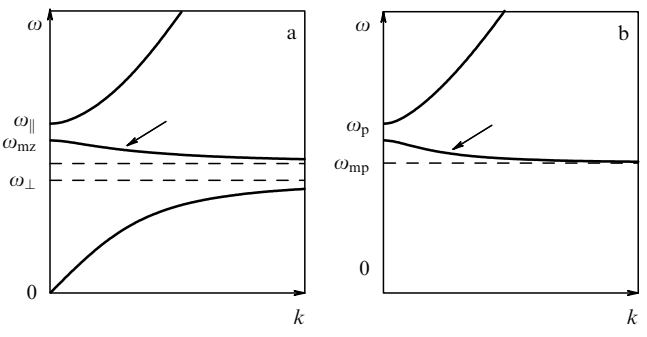

Figure 1. Dispersion $\omega(k)$ of transverse polaritons in materials described by model magnetic permeability (5) and dielectric permittivities given by (a) Eqn (4) and (b) Eqn (6) for a specific arrangement of characteristic frequencies. Polariton branches with negative group velocity are indicated by arrows. We note that neither this figure nor the following figures are plotted to scale: the numerical parameters have been chosen only for the purpose of better displaying qualitative features of the phenomena.

where $n(\omega)$ is the refractive index. Here, the model equation for the dielectric permittivity has a resonant structure,

$$\varepsilon(\omega) = 1 + \frac{F_{\mathrm{e}}}{\omega_{\perp}^2 - \omega^2} = \frac{\omega_{\parallel}^2 - \omega^2}{\omega_{\perp}^2 - \omega^2}\,, \tag{4}$$

and

$$\mu(\omega) = 1 + \frac{F_{\mathrm{m}}}{\omega_{\mathrm{mp}}^2 - \omega^2} = \frac{\omega_{\mathrm{mz}}^2 - \omega^2}{\omega_{\mathrm{mp}}^2 - \omega^2}\,. \tag{5}$$

One of three polariton branches shown in Fig. 1a is evidently characterized by a negative group velocity, because the polariton frequency ω decreases with the increase in the wave vector k (this branch is indicated by an arrow). Of course, the negative group velocity branch exists exactly in that region of frequencies where both $\varepsilon(\omega)$ in (4) and $\mu(\omega)$ in (5) are negative. With the particular choice of the parameters used in Fig. 1, the values of the pole (ω_{mp}) and of the zero (ω_{mz}) of the magnetic permeability are located inside the well-known transverse–longitudinal ($\omega_{\perp}$–$\omega_{\parallel}$) splitting gap, which appears due to the resonance of the dielectric permittivity. Of course, other arrangements of these frequencies are possible.

Figure 1b shows the polariton dispersion with the same expression (5) for $\mu(\omega)$, but with the model dielectric permittivity given by

$$\varepsilon(\omega) = 1 - \frac{\omega_{\mathrm{p}}^2}{\omega^2} \tag{6}$$

instead of Eqn (4). This model expression corresponds to the frequently discussed case of metallic systems, in which the transverse frequency $\omega_{\perp}$ vanishes and $\omega_{\parallel}$ equals the plasma frequency ω_{p}. One of two polariton branches has negative group velocity.

2. The nature of negative refraction: historical remarks

2.1 L I Mandel'shtam and the negative refraction of light

Recent observation of the negative refraction of microwaves [14] and the theoretical prediction of perfect lensing [15] have resulted in a burst of interest in materials with negative refraction. A huge number of articles has been published in scientific and popular journals, and even in newspapers. Very often, the aforementioned work by Veselago [10], done in 1968, is regarded as the origin of the subject. In reality, as already mentioned in the Introduction, the history of negative refraction started much earlier, because a deep understanding of the essence of this phenomenon was achieved by Mandel'shtam at least as early as 1940, and the paper by Veselago just did not contain the references to the previous works.

The founder of a remarkable Moscow physics school (see, e.g., Ref. [16]), Mandel'shtam gave a series of informal lecture courses at Moscow State University that started in the 1930s and continued for many years. The lectures covered many important and subtle topics in optics, the theory of relativity, and quantum mechanics and were famous for their in-depth analysis. They were well attended not only by students but also by many venerable professors. Thanks to lecture notes taken by Mandel'shtam's collaborators S M Rytov and M A Leontovich, the lectures were then published as a part of Mandel'shtam's *Complete Works* and, much later, separately [3].

At one of the lectures on the theory of oscillations in 1944 [1], Mandel'shtam gave a detailed analysis of the negative refraction occurring at a plane interface with a medium supporting waves with negative group velocity. We quote a part of Mandel'shtam's lecture below. After discussing the conditions under which the group velocity gives the velocity of energy propagation, Mandel'shtam continues:

''Let these conditions be satisfied and, hence, the energy propagate with the group velocity. But we know that the group velocity can be negative. This means that the group (and the energy) propagates in the direction opposite to the propagation direction of the phase of the wave. Is this possible in reality?

In 1904, Lamb invented some artificial mechanical models of one-dimensional 'media' in which the group velocity can be negative. He himself probably did not think that his examples may have physical applications. It turns out, however, that there exist real media where the phase and group velocities are directed opposite to each other in some frequency ranges. This happens for the so-called 'optical' branches of the vibrational spectrum of the crystal lattice considered by M Born. The existence of such situations allows one to reconsider such seemingly well-known phenomena as reflection and refraction of a plane wave at the interface between two nonabsorbing media. While traditional discussions of this process do not even mention the notion of the group velocity, the way it occurs significantly depends on the sign of the group velocity.

Indeed, what is the idea underlying the derivation of Fresnel's formulas?

One considers a sinusoidal plane wave incident at an angle φ on the interface plane $y = 0$,

$$E_{\text{inc}} = \exp\left\{\mathrm{i}\left[\omega t - k(x\sin\varphi + y\cos\varphi)\right]\right\}$$

and, in addition, two other waves: the reflected one

$$E_{\text{refl}} = \exp\left\{\mathrm{i}\left[\omega t - k(x\sin\varphi' - y\cos\varphi')\right]\right\}$$

and the refracted one

$$E_{\text{refr}} = \exp\left\{\mathrm{i}\left[\omega t - k_1(x\sin\varphi_1 + y\cos\varphi_1)\right]\right\}.$$

At the plane $y = 0$, these waves should obey the so-called boundary conditions. For elastic bodies, those are the conditions of continuity for the stresses and displacements on both sides of the interface. In the electromagnetic problem, tangential components of the fields and normal components of the inductions must be continuous at the interface. It is easy to show that with the reflected wave alone (or with the refracted wave alone), the boundary conditions cannot be satisfied. But with both reflected and refracted waves, the boundary conditions can always be satisfied. From this consideration, however, it does not follow that there should be only *three* waves involved and not a larger number of waves: in fact, boundary conditions permit one more, the fourth wave propagating in the second medium at the angle $\pi - \varphi_1$. Conventionally, it is tacitly assumed that this wave is not involved and only *one* wave propagates in the second medium.

The boundary conditions immediately imply the laws of reflection

$$\sin\varphi = \sin\varphi', \quad \varphi = \varphi'$$

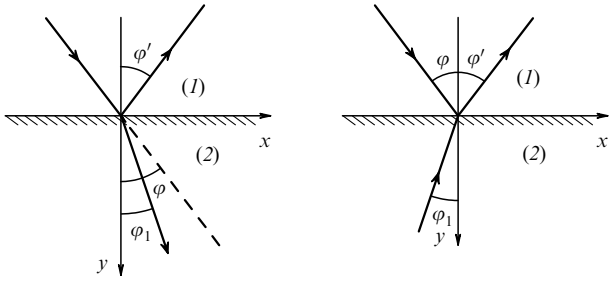

Figure 2. Reflection and refraction of an incident plane wave. (The picture is taken from Mandelstam's lectures [1, 3].)

and of refraction

$$k\sin\varphi = k_1\sin\varphi_1 .$$

The last equation is, however, satisfied not only by the angle φ_1 but also by the angle $\pi - \varphi_1$. The wave corresponding to φ_1 propagates in the second medium away from the interface (left panel in Fig. 2). On the contrary, the wave corresponding to $\pi - \varphi_1$ propagates towards the interface (right panel in Fig. 2). It is considered self-evident that the second wave cannot be involved because the light is incident from the first medium onto the second medium and, hence, the *energy* in the second medium should propagate away from the interface. But what is the relation to the energy? The direction of the wave propagation is determined by its *phase* velocity, while the energy propagates with the *group* velocity. A leap of logic is thus made here, which goes undetected just because we are used to the notion that the directions of the energy and phase propagation coincide. If these directions indeed coincide, i.e., if the group velocity is positive, then everything is correct. But if we have the case of negative group velocity, quite a realistic case as I have already discussed, then everything changes. Still requiring that the energy in the second medium *propagates away* from the interface, we conclude that the phase in this case should propagate towards the interface, and therefore the refracted wave would propagate at the angle $\pi - \varphi_1$ [as shown in the right panel in Fig. 2]. Although such a conclusion is unusual, there is, of course, nothing surprising in it, because the phase velocity does not say anything about the energy propagation direction.''

These remarks by Mandel'shtam made more than 60 years ago actually explain the physical origin and the nature of negative refraction. It is instructive that in his explanation of the nature of negative refraction, Mandel'shtam speaks in terms of the wave vector, group velocity, and causality principle and not in terms of the negative refractive index, which is quite popular presently. It follows from the causality principle that in thermodynamic equilibrium, the intensity of the wave propagating away from the interface should decay. This rule determines the sign of the imaginary part of the refractive index and, thereby, the sign of its real part, because both should result simultaneously from the sign in the expression $n(\omega) = \pm\sqrt{\varepsilon(\omega)\,\mu(\omega)}$ following from Eqn (3).

The relation between negative refraction and negative group velocity established by Mandel'shtam clearly identifies negative refraction as a general wave phenomenon. It also indicates a way of finding materials in which negative refraction can in principle be observed, by examining the dispersion $\omega(k)$ of the waves they can support. For a brief

recent review of the history of the negative group velocity problem, see Ref. [17], which refers to the early discussions such as those by Lamb [18] and von Laue [19].

The importance of the notion of group velocity for crystal optics is extensively discussed in the monograph [7]. The negative refraction occurring at the interface of a gyrotropic medium was explicitly indicated already in its 1966 edition, accompanied by the now very familiar Fig. 2 (see p. 264 in Ref. [7]).

2.2 Cherenkov radiation

In media supporting waves with negative group velocity, Cherenkov radiation has a peculiar character, which was also understood a long time ago. From the theory of Cherenkov radiation (see, e.g., Ref. [6]) and from the sign of the group velocity, the origin of the 'unusual' direction of the propagation of the radiation can be readily seen. We suppose that a charged particle moves in a transparent medium along the x axis with a velocity v. As a result, the medium can emit electromagnetic waves with a frequency ω and a wave vector $\mathbf{k}$ such that $\omega = k_x v$. On the other hand, the wave vector and the frequency are related by $k = n\omega/c$, where $n = \sqrt{\varepsilon}$ is the refractive index. Because $k > k_x$, it follows that we must have $v > v_{\rm ph} = c/n(\omega)$, that is, the radiation of waves with the frequency ω is possible if the velocity of the particle exceeds the phase velocity $v_{\rm ph}$. If θ is the angle between the direction of the particle motion and the wave vector $\mathbf{k}$ of the radiation, we immediately find that

$$\cos\theta = \frac{c}{n(\omega)v}\,. \tag{7}$$

Quoting from Ref. [6], "...the radiation of each frequency is emitted in the forward direction with respect to the particle motion, and is distributed over the surface of a cone with the vertical angle 2θ, where θ is given by Eqn (7)."

It is clear from the logic of the above derivation that the conclusion on the direction of the emission tacitly assumed that the group velocity $\mathbf{v}_{\rm g}$ corresponding to the wave vector $\mathbf{k}$ was positive, that is, directed along $\mathbf{k}$ (this situation is depicted in Fig. 3a). If, instead, the group velocity was negative, i.e., $\mathbf{v}_{\rm g}$ was directed opposite to $\mathbf{k}$, the direction of the emission (the energy flow $\mathbf{S}$) would in fact be opposite. The radiation in the latter case forms an obtuse angle with the direction of the particle motion, as was first discussed by Pafomov [13]. The Cherenkov radiation emitted backwards is shown in Fig. 3b. It is distributed over the surface of the cone with the same vertical angle.

We show in what follows that negative group velocity waves may occur in crystals due to the presence of spatial dispersion. Various manifestations of spatial dispersion in Cherenkov radiation have been discussed in monograph [7] (see pp. 400, 401). Particularly interesting effects have been indicated both in gyrotropic [7, 20] and nongyrotropic [7] media in the vicinity of excitonic resonances: the direction of the Cherenkov cone is modified from forward radiation to backward radiation upon a decrease in the velocity of the moving charge.

It is also interesting how the negative group velocity influences the transition radiation of a charged particle crossing the boundary between two media with different dielectric constants. The important role of the sign of the group velocity for the transition radiation and peculiarities of the 'inverse' Doppler effect were originally clarified in papers by Frank [21], Barsukov [22], and Pafomov [12].

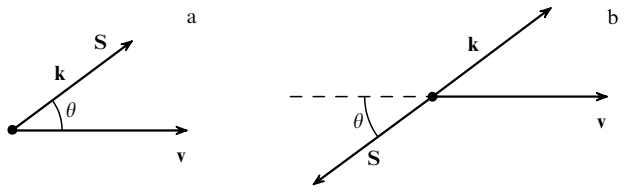

Figure 3. Illustration on the direction of the Cherenkov radiation in a medium with (a) positive and (b) negative group velocity. Here, $\mathbf{v}$ denotes the direction of the particle velocity, $\mathbf{k}$ the direction of the wave vector of the emitted radiation, and $\mathbf{S}$ the direction of the Poynting vector. The vector $\mathbf{S}$ is directed along the group velocity $\mathbf{v}_{\rm g}$ and determines the actual direction of the emission.

3. Maxwell equations and spatial dispersion

3.1 The dielectric permittivity tensor

The macroscopic Maxwell equations form the basis of the electrodynamics of continuous media [6]. They are derived by averaging the 'microscopic' electromagnetic fields, charges, and current densities and have to be supplemented by the so-called matter equations, which determine the relations between the averaged fields. The matter equations are determined by the response of the medium to the fields. Following Landau and Lifshits [6] (see also Refs [7, 23]), we find it more correct and appropriate to use an approach based on taking spatial dispersion into account. In this approach, only three macroscopic fields — $\mathbf{E}$, $\mathbf{D}$, and $\mathbf{B}$ — are considered, while the fourth field, $\mathbf{H}$, is set equal to $\mathbf{B}$. The results of averaging *all* microscopic currents are then included in the definition of the field $\mathbf{D}$. The macroscopic Maxwell equations for monochromatic plane waves then take the form

$$\begin{aligned} &c\,\mathbf{k}\times\mathbf{E} = \omega\mathbf{B}\,,\\ &c\,\mathbf{k}\times\mathbf{B} = -\omega\mathbf{D}\,,\\ &\mathbf{k}\,\mathbf{D} = 0\,,\\ &\mathbf{k}\,\mathbf{B} = 0\,, \end{aligned} \tag{8}$$

while the matter equation relating the components of $\mathbf{D}$ and $\mathbf{E}$ in such waves is given by

$$D_i = \varepsilon_{ij}(\omega,\mathbf{k})\,E_j\,. \tag{9}$$

In Eqn (9), the generalized dielectric tensor $\varepsilon_{ij}(\omega,\mathbf{k})$ depends on the wave vector $\mathbf{k}$. This means that the *spatial dispersion* is taken into account, that is, that the electric induction $\mathbf{D}$ at a given spatial point depends not only on the electric field $\mathbf{E}$ at the same point (the local medium response) but also on the electric field in some neighborhood (the nonlocal response). As such, this tensor $\varepsilon_{ij}(\omega,\mathbf{k})$ describes both dielectric and magnetic responses of the medium (the latter through a natural account of the spatial derivatives of $\mathbf{E}$). The spatial dispersion appears in addition to the more familiar temporal, or frequency, dispersion, which is reflected in the dependence of the tensor on the frequency ω. The spatial dispersion effects are usually much weaker than those arising from the frequency dispersion, but they can lead to qualitatively new phenomena, such as gyrotropy or the appearance of additional electromagnetic waves. The consideration of spatial dispersion is simplified if the relevant parameter $ka \sim a/\lambda$ is small, where a is the characteristic microscopic length or the

mean free path of the charge carriers. In many cases, the smallness of the parameter ka allows using only the first terms (linear and/or quadratic) in the power-series expansion for the tensor $\varepsilon_{ij}(\omega,\mathbf{k})$ in the components of the wave vector $\mathbf{k}$ [6, 7]:

$$\varepsilon_{ij}(\omega,\mathbf{k}) = \varepsilon_{ij}(\omega) + \mathrm{i}\gamma_{ijl}(\omega)k_l + \alpha_{ijlm}(\omega)k_l k_m\,, \tag{10}$$

$$\varepsilon_{ij}^{-1}(\omega,\mathbf{k}) = \varepsilon_{ij}^{-1}(\omega) + \mathrm{i}\delta_{ijl}(\omega)k_l + \beta_{ijlm}(\omega)k_l k_m\,. \tag{11}$$

Various tensors occurring in expansions (10) and (11) reflect the symmetry properties of the system under consideration and satisfy Onsager's principle of symmetry of kinetic coefficients. In particular, in a system with the inversion symmetry, the second terms of the expansions (those proportional to the first power of k_l) vanish.

Because Maxwell equations (8) immediately yield

$$\mathbf{D} = \frac{c^2k^2}{\omega^2}\left(\mathbf{E} - \frac{\mathbf{k}(\mathbf{kE})}{k^2}\right), \tag{12}$$

it follows that Eqns (12) and (9), taken together, *determine the dispersion relations* $\omega(\mathbf{k})$ of the electromagnetic waves propagating in the medium.

The time-averaged energy density and the Poynting vector in the discussed $(\mathbf{E},\mathbf{D},\mathbf{B})$-approach can be found via an analysis of the wave packets [6, 7]. These quantities are given by

$$U = \frac{1}{16\pi}\left[\frac{\partial(\omega\varepsilon_{ij})}{\partial\omega}\,E_i E_j^* + |\mathbf{B}|^2\right], \tag{13}$$

$$\mathbf{S} = \frac{c}{8\pi}\,\mathrm{Re}\,(\mathbf{E}^*\times\mathbf{B}) - \frac{\omega}{16\pi}\,\frac{\partial\varepsilon_{ij}}{\partial\mathbf{k}}\,E_i^* E_j\,. \tag{14}$$

In the absence of dissipation, expressions (13) and (14) satisfy the energy conservation law. Equation (14) contains an additional (second) term [24, 25], entirely due to accounting for spatial dispersion. This term plays a crucial role for the appearance of negative group velocity waves.

In Section 3.2, we provide a comparative discussion of the $(\mathbf{E},\mathbf{D},\mathbf{B})$-approach, which accounts for the spatial dispersion, and of the so-called 'symmetric' approach based on the consideration of all four fields $\mathbf{E},\mathbf{D},\mathbf{B}$, and $\mathbf{H}$. We nevertheless refer the reader to book [26], reviews [27–29], and recent paper [8] for discussions of the different standpoints, other arguments, and details.

3.2 An isotropic medium with the spatial inversion symmetry

When spatial dispersion is taken into account, the dielectric response must be described by a tensor even in isotropic systems, because the vector $\mathbf{k}$ selects a certain direction. Consequently, for an isotropic medium having the inversion symmetry (a nongyrotropic medium), the general form of the dielectric tensor is [6]

$$\varepsilon_{ij}(\omega,\mathbf{k}) = \varepsilon_\perp(\omega,k)\left[\delta_{ij} - \frac{k_i k_j}{k^2}\right] + \varepsilon_\parallel(\omega,k)\,\frac{k_i k_j}{k^2}\,, \tag{15}$$

where the *transverse* $\varepsilon_\perp(\omega,k)$ and *longitudinal* $\varepsilon_\parallel(\omega,k)$ dielectric permittivities depend on the absolute value of the wave vector k only. These two functions provide a complete description of the properties of the medium. In accordance with Eqns (12) and (9), the dispersion law $\omega(k)$ of the transverse $(\mathbf{E}\perp\mathbf{k})$ polaritons can be found from

$$\omega^2\varepsilon_\perp(\omega,k) = c^2k^2\,, \tag{16}$$

while the equation

$$\varepsilon_\parallel(\omega,k) = 0 \tag{17}$$

determines the dispersion of longitudinal $(\mathbf{E}\parallel\mathbf{k},\ \mathbf{D}=0,\ \mathbf{B}=0)$ waves.

The symmetric approach, which involves only the frequency-dependent dielectric permittivity $\varepsilon(\omega)$ and magnetic permeability $\mu(\omega)$, corresponds to the limit $k\to 0$ in the approach based on accounting for the spatial dispersion [6]:

$$\varepsilon(\omega) = \varepsilon_\perp(\omega,0) = \varepsilon_\parallel(\omega,0)\,, \tag{18}$$

$$1 - \frac{1}{\mu(\omega)} = \lim_{k\to 0}\frac{\omega^2\left[\varepsilon_\perp(\omega,k) - \varepsilon_\parallel(\omega,k)\right]}{c^2k^2}\,. \tag{19}$$

It can be easily seen that the particular choice

$$\varepsilon_\perp(\omega,k) = \varepsilon(\omega) + \frac{c^2k^2}{\omega^2}\left(1 - \frac{1}{\mu(\omega)}\right) \tag{20}$$

makes the dispersion equation for transverse polaritons (16) *identical* to Eqn (3) derived within the $\varepsilon(\omega)-\mu(\omega)$ description, where $k^2c^2/\omega^2 = n^2 = \varepsilon(\omega)\,\mu(\omega)$. This already clearly demonstrates a more comprehensive scope of the approach based on accounting for spatial dispersion, because it allows studying various effects of spatial dispersion with an accuracy even higher than that given by accounting for the terms $\propto k^2$ in $\varepsilon_\perp(\omega,k)$. In contrast, the $\varepsilon(\omega)-\mu(\omega)$ approach allows taking only these terms into account (and not even all of them). Moreover, even within the k^2-accuracy, there is a qualitative advantage offered by the spatial dispersion approach. Indeed, in the isotropic system under consideration, the response tensor α_{ijlm} entering Eqn (10) is in general characterized by *two independent* parameters. These parameters (a and b) can be chosen, for instance, such that the equality

$$\alpha_{ijlm} = a\delta_{ij}\delta_{lm} + \frac{b}{2}\left(e_{ril}\,e_{rjm} + e_{rim}\,e_{rjl}\right), \tag{21}$$

is satisfied (e_{ril} denotes the antisymmetric unit tensor of rank three). Equation (21) is manifestly symmetric in both the first (ij) and the second (lm) pairs of indices, and therefore the longitudinal and transverse dielectric permittivities can be written as

$$\begin{aligned}\varepsilon_\perp(\omega,k) &= \varepsilon(\omega) + \left[a(\omega) + b(\omega)\right]k^2\,,\\ \varepsilon_\parallel(\omega,k) &= \varepsilon(\omega) + a(\omega)k^2\,.\end{aligned} \tag{22}$$

The corresponding matter relation (9), as follows from Eqns (10) and (21), is

$$\mathbf{D} = \left(\varepsilon(\omega) + a(\omega)k^2\right)\mathbf{E} + b(\omega)\,\mathbf{k}\times(\mathbf{E}\times\mathbf{k})\,. \tag{23}$$

Equations (22) and (23) make it clear that the parameter $b(\omega)$ determines, roughly speaking, the measure of the spatial dispersion due to the 'magnetic response' of the system: the parameter $b(\omega)$ is related to the magnetic permeability [see Eqn (19)] as

$$\frac{\omega^2 b(\omega)}{c^2} = 1 - \frac{1}{\mu(\omega)}\,.$$

The parameter $a(\omega)$, on the other hand, determines the measure of the spatial dispersion due to the 'electric response.' The parameter $a(\omega)$ and its dependence on ω cannot be taken into account within the $\varepsilon(\omega)-\mu(\omega)$ description. Both types of responses contribute similarly to the dispersion of transverse polaritons [Eqns (22) and (16)], but only the electric response can affect the longitudinal waves [Eqns (22) and (17)]. Particularly noteworthy is that polaritons with negative group velocity and, consequently, negative refraction may occur in systems with $\mu(\omega) = 1$ [that is, with $b(\omega) = 0$] if the response coefficient $a(\omega)$ has an appropriate frequency dependence.

3.3 The connection with a microscopic description

The dielectric tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ describes the response of the medium to electromagnetic perturbations of *arbitrary* frequencies ω and wave vectors $\mathbf{k}$. This tensor has certain well-known analytic properties and can in principle be derived from the microscopic description of elementary excitations of the medium using various methods (see, e.g., Refs [7, 23, 30–32] for discussions of many important aspects of this question). An example of such a microscopic relation for a perturbed ground state of a system of N charged particles of charge e and mass m in a volume V is given by the expression [7]

$$\varepsilon_{ij}(\omega, \mathbf{k}) = \left(1 - \frac{4\pi e^2 N}{m\omega^2 V}\right)\delta_{ij} - \frac{4\pi c^2}{\hbar\omega^2 V}\sum_n \left(\frac{M_i^{n*}(\mathbf{k})\, M_j^n(\mathbf{k})}{\omega - \omega_n} - \frac{M_i^n(-\mathbf{k}) M_j^{n*}(-\mathbf{k})}{\omega + \omega_n}\right). \quad (24)$$

The vectors $\mathbf{M}^n(\mathbf{k})$ entering Eqn (24) are the perturbation matrix elements written in Cartesian coordinates:

$$\mathbf{M}^n(\mathbf{k}) = -\frac{e}{2mc}\sum_\alpha \left\langle n\middle|\mathbf{p}^\alpha \exp\left(\mathrm{i}\mathbf{k}\mathbf{r}^\alpha\right) + \exp\left(\mathrm{i}\mathbf{k}\mathbf{r}^\alpha\right)\mathbf{p}^\alpha\middle|0\right\rangle, \quad (25)$$

where $\mathbf{r}^\alpha$ is the position vector of the αth particle and $\mathbf{p}^\alpha = -\mathrm{i}\hbar\,\partial/\partial\mathbf{r}^\alpha$ is its momentum operator. Here, $|0\rangle$ is the wave function of the ground state of the system and $|n\rangle$ are the unperturbed wave functions of various excited states with energies $\hbar\omega_n$. These zeroth-order states, which we call *excitons* ('mechanical excitons,' following the terminology in Ref. [7]), are to be calculated without taking the macroscopic electromagnetic field into account.

It is instructive to consider the microscopic origin of expressions of type (20) and (22) in an isotropic system with the inversion symmetry. For the simplest model of independent atoms or molecules, in accordance with Eqn (24), the dielectric permittivity $\varepsilon(\omega)$ is determined by the elements $\mathbf{M}^n(\mathbf{k} = 0)$ (more exact models are considered in Refs [33, 34]):

$$M_i^n = -\frac{e}{mc}\sum_\alpha \left\langle n\middle| p_i^\alpha\middle|0\right\rangle; \quad (26)$$

therefore, only electric-dipole-allowed transitions (also called E1-transitions) contribute to $\varepsilon(\omega)$. Then, with the corresponding transition frequencies denoted as ω_{en}, it follows from Eqn (24) that

$$\varepsilon(\omega) = 1 + \sum_n \frac{F_{en}}{\omega_{en}^2 - \omega^2}, \quad (27)$$

where the 'oscillator strengths' F_{en} satisfy the sum rule $\sum_n F_{en} = 4\pi e^2 N/mV$. In the vicinity of a single resonance frequency $\omega_\perp$, Eqn (27) has the structure

$$\varepsilon(\omega) = \epsilon_\mathrm{b} + \frac{F_\mathrm{e}}{\omega_\perp^2 - \omega^2} = \epsilon_\mathrm{b}\,\frac{\omega_\parallel^2 - \omega^2}{\omega_\perp^2 - \omega^2}, \quad (28)$$

with $\omega_\parallel^2 = \omega_\perp^2 + F_\mathrm{e}/\epsilon_\mathrm{b}$, where the background constant ϵ_b accounts for all other allowed transitions [$\epsilon_\mathrm{b} = 1$ in Eqn (4)].

The term $\propto k^2$ in Eqn (20) has a very different origin, because it appears due to electric-dipole-forbidden transitions. In the molecular picture, such forbidden transitions become possible due to the next term in the expansion of $\exp(\mathrm{i}\mathbf{k}\mathbf{r}^\alpha)$ in Eqn (25), and their nonvanishing contribution to $M_i^n(\mathbf{k})$ is

$$M_i^n(\mathbf{k}) = -\frac{e}{2mc}\sum_\alpha \mathrm{i}k_l \left\langle n\middle| p_i^\alpha r_l^\alpha + r_l^\alpha p_i^\alpha\middle|0\right\rangle = \mathrm{i}k_l X_{il}^n. \quad (29)$$

Magnetic dipole transitions (M1-transitions) are due to the antisymmetric combination

$$r_l^\alpha p_i^\alpha - r_i^\alpha p_l^\alpha. \quad (30)$$

This combination should occur between $\langle n|$ and $|0\rangle$ in Eqn (29). But the actual combination in those brackets differ from (30) by

$$p_i^\alpha r_l^\alpha + r_i^\alpha p_l^\alpha. \quad (31)$$

It is well known that the combination in Eqn (31) leads to electric quadrupole transitions (E2-transitions). The difference between the magnetic dipole and electric quadrupole transitions is reflected in the symmetry of the tensor X_{il}^n defined in Eqn (29): it is antisymmetric ($X_{il}^{n(\mathrm{m})} = -X_{li}^{n(\mathrm{m})}$) for the former but symmetric ($X_{il}^{n(\mathrm{q})} = X_{li}^{n(\mathrm{q})}$) for the latter. The contribution of both E2- and M1-transitions to the tensor α_{ijlm} in (21) is given by

$$\frac{4\pi c^2}{\hbar\omega^2 V}\sum_n \frac{\omega_n (X_{il}^{n*} X_{jm}^n + X_{jl}^{n*} X_{im}^n)}{\omega_n^2 - \omega^2}. \quad (32)$$

We note that the magnetic dipole combinations $X_{il}^{n(\mathrm{m})*} X_{jm}^{n(\mathrm{m})}$ entering Eqn (32) indeed contribute only to the magnetic response coefficient $b(\omega)$ in Eqn (21). On the other hand, the electric quadrupole combinations of the type $X_{il}^{n(\mathrm{q})*} X_{jm}^{n(\mathrm{q})}$ can contribute to both response coefficients $a(\omega)$ and $b(\omega)$ defined in Eqn (21). The relevant examples can be found, e.g., in Ref. [7] (for a general discussion of the electric quadrupole polarization in macroscopic electrodynamics, see Ref. [35]).

Equation (32) explicitly shows that magnetic dipole and electric quadrupole transitions can lead to contributions of the same type to the transverse dielectric function $\varepsilon_\perp(\omega, k)$. To take such a contribution from a single isolated resonance with a frequency ω_f into account, we must replace Eqn (28) with the expression

$$\varepsilon_\perp(\omega, k) = \epsilon_\mathrm{b} + \frac{F_\mathrm{e}}{\omega_\perp^2 - \omega^2} + \frac{c^2 k^2}{\omega^2}\,\frac{F_\mathrm{f}}{\omega_\mathrm{f}^2 - \omega^2}, \quad (33)$$

where F_f determines the strength of the transition. The properties of the medium, which follow from Eqn (33), are determined by the interplay of two resonances: one that is electric-dipole allowed, and the other that is electric-dipole forbidden. As a result of this interplay, polaritons with

negative group velocity can appear (see Fig. 1). It is easy to see from Eqns (4), (5), and (20) that the frequency ω_{mz} at which $\mu(\omega) = 0$ corresponds to the frequency ω_f of the forbidden transition ($\omega_{mz} = \omega_f$), and $F_m = F_f$.

It follows from the above derivation that the strength of forbidden electronic transitions in atomic or molecular materials is in general much weaker than the strength of electric-dipole-allowed transitions:

$$\frac{F_f}{F_e} \sim \frac{\omega_\perp \omega_f a^2}{c^2} \sim \frac{v^2}{c^2} \ll 1\,, \tag{34}$$

where a is the characteristic atomic or molecular length and v is the typical electron velocity. In this connection, we recall that in Fig. 1, F_e/ϵ_b determines the magnitude of the $\omega_\parallel - \omega_\perp$ splitting, while $F_f = F_m$ determines the band width of the polariton branch with negative group velocity.

3.4 An isotropic medium without the spatial inversion symmetry

In media with a broken spatial inversion symmetry (gyrotropic media), spatial dispersion already manifests itself in the first-order terms in the wave vector $\mathbf{k}$, because the tensors γ_{ijl} and δ_{ijl} in expansions (10) and (11) do not vanish. The interesting features of the polariton dispersion in such media can readily be outlined even if only these linear terms are retained [7, 36]. In isotropic systems, tensors of the general forms γ_{ijl} and δ_{ijl} reduce to the unit antisymmetric tensor and the expansions become

$$\varepsilon_{ij}(\omega, \mathbf{k}) = \varepsilon(\omega)\,\delta_{ij} + \mathrm{i}\gamma(\omega)\,e_{ijl}\,k_l\,, \tag{35}$$

$$\varepsilon_{ij}^{-1}(\omega, \mathbf{k}) = \frac{1}{\varepsilon(\omega)}\,\delta_{ij} + \mathrm{i}\delta(\omega)\,e_{ijl}\,k_l\,. \tag{36}$$

As we discuss in Sections 4.2 and 4.3, Eqn (35) is applicable in the vicinity of the longitudinal frequency $\omega_\parallel$: $\varepsilon(\omega_\parallel) = 0$, while Eqn (36) is to be used in the vicinity of the resonance frequency $\omega_\perp$: $1/\varepsilon(\omega_\perp) = 0$.

It is instructive to see the microscopic origin of the dielectric tensor written in forms (35) and (36). For a set of independent gyrotropic molecules, for instance, it is well known (see Refs [32–34]) that optical activity results from transitions to the states $|n\rangle$ with nonvanishing matrix elements of both types [(26) and (29)]. Such transitions indeed lead to the appearance of terms linear in k_l in Eqn (35): they give rise to the tensor γ_{ijl} (10) via combinations of the type

$$\frac{4\pi c^2}{\hbar\omega^2 V}\sum_n \frac{\omega_n(M_i^n X_{jl}^{n*} - M_j^n X_{il}^{n*} + \text{c.c.})}{\omega_n^2 - \omega^2}\,. \tag{37}$$

The microscopic meaning of the function $\delta(\omega)$ in Eqn (36) is addressed in Section 4.2.

4. Polaritons with negative group velocity

As was already mentioned in Section 3.1, it is the second term in expression (14) for the Poynting vector $\mathbf{S}$ that explicitly shows how spatial dispersion can 'invert' the direction of the energy propagation with respect to the wave vector $\mathbf{k}$. Indeed, the first term in (14) in an isotropic medium is a vector directed along $\mathbf{k}$. For the group velocity to become negative, the second term has to be codirected with $-\mathbf{k}$ and be larger in magnitude. In particular, this implies that the spatial dispersion $\partial\varepsilon_\perp(\omega, k)/\partial k$ must be strong enough. This is precisely the case in a medium characterized by Eqn (33) with negative $\varepsilon(\omega)$ and $\mu(\omega)$ for frequencies below the forbidden frequency ω_f. In Sections 4.1–4.3, we discuss several other examples where substantial spatial dispersion of the dielectric permittivity leads to the appearance of polaritons with negative group velocity.

4.1 Excitons with negative effective mass in a nongyrotropic medium

Pekar [37] first noted in 1957 that spatial dispersion of the dielectric permittivity near excitonic resonances could lead to the appearance of an additional propagating light (exciton–polariton) wave. This possibility is connected to the fact that excitons in the medium can move (e.g., from one molecule to another) and their energy depends on the wave vector $\mathbf{k}$. We consider expression (28) for the transverse dielectric permittivity, which determines the response corresponding to an isolated electric-dipole-allowed excitonic transition at the frequency $\omega_\perp$. Matrix elements (25) 'select' the excitonic states $|n\rangle$ with the (quasi)momentum $\hbar\mathbf{k}$ only, and hence the energies ω_n must correspond to such a momentum. In the effective mass approximation, the dispersion of the exciton energy is

$$\omega_\perp(k) = \omega_\perp + \frac{\hbar^2 k^2}{2M_{exc}}\,. \tag{38}$$

Correspondingly, the transverse dielectric function is

$$\varepsilon_\perp(\omega, k) = \epsilon_b + \frac{F_e}{\omega_\perp^2(k) - \omega^2}\,, \tag{39}$$

which is to coincide with Eqn (28) for motionless excitons ($M_{exc} = \infty$). Of course, the oscillator strength F_e must also acquire some k-dependence, but we limit our discussion to a stronger effect related to the resonance denominator in Eqn (39). We note that the type of spatial dispersion exhibited, for instance, in Eqn (39) cannot be described within the $\varepsilon(\omega)-\mu(\omega)$ framework. It is easy to find the dispersion of transverse polaritons from Eqns (16) and (39); the illustrative examples are displayed in Fig. 4.

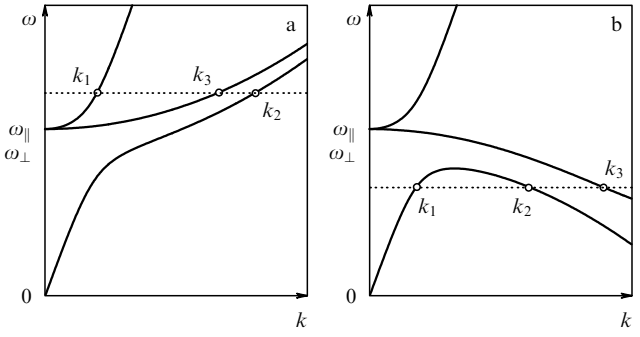

Figure 4. Dispersion of two transverse polariton branches and of the longitudinal wave in a system with exciton dispersion (38): (a) positive exciton effective mass, $M_{exc} > 0$; (b) negative effective mass, $M_{exc} < 0$. The points of intersection of the dashed lines with the dispersion curves indicate the values of the wave vector k for the waves at a given frequency ω: k_1 and k_2 are for transverse polaritons and k_3 is for the longitudinal wave. In case (b), k_2 corresponds to a polariton with negative group velocity.

Figure 4 shows that within a certain range of frequencies ω, there can indeed be two different values of k for each frequency, corresponding to two transverse polariton waves of the same polarization. The one with larger k (denoted as k_2) is the additional wave predicted by Pekar.

The existence of additional exciton – polariton waves has been demonstrated in many crystals; the most convincing experiments were conducted in semiconductors near the Wannier – Mott exciton resonances (see Ref. [7] for the references and discussion). Of principal importance for the sign of the polariton group velocity is the sign of the exciton effective mass. The effective mass of Wannier – Mott excitons is usually positive, $M_{\rm exc} = m_{\rm e} + m_{\rm h} > 0$, where $m_{\rm e}$ and $m_{\rm h}$ are the effective masses of electrons and holes. This is the situation depicted in Fig. 4a. Evidently, the additional exciton – polariton wave then has a positive group velocity.

In organic crystals, however, the radius of Frenkel excitons is typically small. The resonant intermolecular interaction then strongly depends on the orientation of the molecules, and as a result, the exciton effective mass can in general be negative or have different signs in different directions. The situation with a negative exciton mass is depicted in Fig. 4b. It clearly illustrates that for some range of frequencies ω, the additional transverse polariton wave (k_2-wave in the figure) has a negative group velocity. This is the transverse wave that experiences negative refraction.

Also shown in Fig. 4 is the dispersion of the longitudinal waves determined by Eqn (17). For definiteness, we assumed $\varepsilon_{\parallel}(\omega, k) = \varepsilon_{\perp}(\omega, k)$. The wave vector of the longitudinal wave is denoted by k_3. In the case where $M_{\rm exc} < 0$, the longitudinal wave also has a negative group velocity. In general, all three waves (two transverse and one longitudinal) can be excited in a medium by an incident wave of an appropriate frequency. To solve the problem of reflection and refraction of the waves in such circumstances, we must specify so-called additional boundary conditions (ABCs) because the usual Maxwell boundary conditions are insufficient to find the amplitudes of all the excited waves. The form of the ABCs does depend on the microscopic nature of excitons. For molecular crystals, this question is extensively discussed in Ref. [7].

A numerical study of the reflection and refraction of light by a planar slab of a medium supporting excitons with a negative effective mass $M_{\rm exc} < 0$ (Fig. 4b) was recently performed in Ref. [38]. These results convincingly show that due to the negative refraction of waves with negative group velocity, such a slab can indeed focus light. The simulation in [38] also indicates that for an experimental realization of such a system, one has to use crystals with large oscillator strength of the excitonic transition and rather weak dissipation of additional polaritons at the frequencies below the excitonic resonance frequency.

4.2 A gyrotropic medium in the vicinity of excitonic transitions

Gyrotropic systems are well known due to the phenomena of optical activity and circular dichroism. They can be expected to support polaritons with negative group velocity within certain regions of frequencies ω. We start our discussion from the case of a frequency region in the vicinity of the exciton transition frequency $\omega_{\perp}$. The additional waves in this frequency region were considered by Ginzburg [36]. Because the transition frequency corresponds to the pole of the dielectric permittivity $\varepsilon(\omega)$, it is more convenient to use expansion (36) for the inverse dielectric tensor in this region. The inverse dielectric permittivity vanishes at the transition frequency $\varepsilon^{-1}(\omega_{\perp}) = 0$, highlighting the qualitative importance of the next (depending on the spatial dispersion) term in that expansion.

Equation (36) corresponds to the matter relation

$$\mathbf{E} = \frac{1}{\varepsilon(\omega)}\,\mathbf{D} + \mathrm{i}\delta(\omega)\,\mathbf{D}\times\mathbf{k} \tag{40}$$

between $\mathbf{E}$ and $\mathbf{D}$ fields, where the parameter $\delta(\omega)$ determines the 'strength' of gyrotropy. Relation (40) combined with wave equation (12) for *transverse* waves leads to the equation

$$\frac{\omega^2}{c^2k^2}\,\mathbf{D} = \frac{1}{\varepsilon(\omega)}\,\mathbf{D} + \mathrm{i}\delta(\omega)\,\mathbf{D}\times\mathbf{k}\,, \tag{41}$$

whose nontrivial solutions describe transverse polaritons in this system. These solutions are known to correspond to circularly polarized waves: for instance, $D_y/D_x = \pm\mathrm{i}$ for waves propagating in the z direction. The polariton dispersion $\omega(k)$ can be determined from the condition that the determinant of Eqn (41) vanishes:

$$\left(\frac{1}{\varepsilon(\omega)} - \frac{\omega^2}{c^2k^2}\right)^2 = \delta^2(\omega)k^2 \tag{42}$$

or, for waves of different circular polarizations,

$$\frac{1}{\varepsilon(\omega)} - \frac{\omega^2}{c^2k^2} = \pm\bigl|\delta(\omega)\bigr|k\,. \tag{42a}$$

Equation (42a) is a third-order equation in $k(\omega)$; as a result, in some frequency regions, three waves can propagate simultaneously in the medium for a given ω. Figure 5a illustrates the transverse polariton dispersion resulting from Eqn (42a) with the model dielectric permittivity $\varepsilon(\omega)$ in Eqn (28) and a constant $\delta(\omega) = \delta$ [36].

It can be easily seen that similarly to the case of a medium with the spatial inversion symmetry (see Section 4.1), the dispersion of polaritons can be understood as being due to a specific dependence of the exciton energies on the wave vector $\mathbf{k}$ [7, 39]. To verify this, we consider the frequency

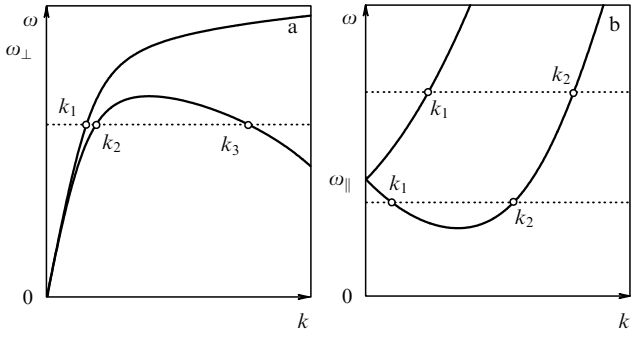

Figure 5. Dispersion of transverse polaritons in a gyrotropic medium. We note distinctly different ranges of frequencies (and wave vectors) in panels (a) and (b). Shown in panel (a) is a range around the frequency $\omega_{\perp}$ and below; the neighborhood of $\omega_{\parallel}$ would be above this plot. Shown in panel (b) is a range around the frequency $\omega_{\parallel}$ and above; the neighborhood of $\omega_{\perp}$ would be below this plot. The points of intersection of dashed lines with the dispersion curves show the allowed values of the wave vector k at a given frequency ω. Both panels show polaritons with negative group velocity.

range in the vicinity of the resonance $\omega_\perp$, where the inverse permittivity can be approximated by a linear function as

$$\varepsilon^{-1}(\omega) \approx A_\perp(\omega_\perp - \omega)\,, \qquad A_\perp = \frac{2\omega_\perp}{F_e}\,. \tag{43}$$

We next take the limit as $c \to \infty$ in Eqn (42a) (i.e. we neglect the retarded interaction between the charges). Then

$$\omega_\perp(k) = \omega_\perp \mp \frac{\delta}{A_\perp}\,k\,. \tag{44}$$

Thus, the 'microscopic' origin of Eqn (42) is due to the presence in the dispersion of the exciton of a term linear in **k**, which has different signs for excitons with different polarizations.

Linear behavior (44) represents the first terms of the k-expansion of the exciton dispersion in a gyrotropic medium with the gyrotropy parameter δ. The linear dependence of the frequency of dipole-active excitations on the wave vector was observed for the first time in spectra of Raman scattering by optical phonons propagating along the optical axis in quartz crystals [40].

As is evident from Fig. 5a, the additional wave with the wave vector k_3, which corresponds to the lower polariton branch, has a negative group velocity. There must also be two other waves excited at the same frequency ω, whose wave vectors are denoted as k_1 and k_2. An experimental realization of negative refraction of k_3-waves requires large oscillator strength of the excitonic transition, as well as large rotatory power and rather small dissipation of additional polaritons at the frequencies below the resonance frequency.

4.3 A gyrotropic medium in the vicinity of the frequency of longitudinal vibrations

A negative refraction of microwaves in an artificial gyrotropic medium was recently considered by Pendry [41] in the vicinity of the frequency $\omega_\parallel$ of longitudinal vibrations using parameters $\varepsilon(\omega)$ and $\mu(\omega)$.[1] In our consideration, we use the approach based on accounting for spatial dispersion, which allows going beyond the low-frequency region. For more details, we refer the reader to paper [46].

Because the longitudinal frequency corresponds to a zero of the dielectric permittivity $\varepsilon(\omega)$, it is appropriate to use expansion (35) of the dielectric tensor. The vanishing $\varepsilon(\omega_\parallel) = 0$ makes it clear that the next term in the expansion, which accounts for spatial dispersion, is qualitatively important. It is seen from definition (28) of the dielectric permittivity that

$$\omega_\parallel = \sqrt{\omega_\perp^2 + \frac{F_e}{\epsilon_b}}\,,$$

and therefore $\varepsilon(\omega)$ is a linear function of ω in the vicinity of $\omega_\parallel$:

$$\varepsilon(\omega) \approx A_\parallel(\omega - \omega_\parallel)\,, \qquad A_\parallel = \frac{2\epsilon_b^2\omega_\parallel}{F_e}\,. \tag{45}$$

[1] We here mention theoretical works [42, 43], in which negative refraction was achieved by formally fitting the parameters ε and μ, as well as works [44, 45], where the possibility of focusing is demonstrated for circularly polarized waves by numerical simulation.

Equation (35) in an isotropic medium takes the form

$$\mathbf{D} = \varepsilon(\omega)\mathbf{E} + \mathrm{i}\gamma(\omega)\,\mathbf{E}\times\mathbf{k}\,, \tag{46}$$

where the parameter $\gamma(\omega)$ determines the strength of gyrotropy. Using Eqn (12), we find that the fields of *transverse* polaritons satisfy the equation

$$\frac{c^2k^2}{\omega^2}\,\mathbf{E} = \varepsilon(\omega)\mathbf{E} + \mathrm{i}\gamma(\omega)\,\mathbf{E}\times\mathbf{k}\,. \tag{47}$$

Nontrivial solutions of Eqn (47) are circularly polarized waves whose dispersion can be determined from

$$\varepsilon(\omega) - \frac{c^2k^2}{\omega^2} = \pm\bigl|\gamma(\omega)\bigr|k\,, \tag{48}$$

where the plus and minus signs correspond to waves with different circular polarizations. Figure 5b illustrates the transverse polariton dispersion in the frequency range around and above $\omega_\parallel$. These curves were obtained from Eqn (48) when using the model dielectric permittivity $\varepsilon(\omega)$ in Eqn (28).

It is easy to qualitatively understand the character of the polariton spectrum shown in Fig. 5b by substituting expression (45) in dispersion equation (48). This substitution immediately yields the polariton dispersion curves in the form of 'displaced parabolas,'

$$\omega_\pm(k) \approx \omega_\parallel + \frac{c^2}{A_\parallel\omega_\parallel^2}\,k^2 \pm \frac{\gamma}{A_\parallel}\,k\,, \tag{49}$$

where $\gamma = \gamma(\omega_\parallel)$. As is evident from Eqn (49) and from Fig. 5b, there are two types of solutions for each frequency ω at which the waves can exist. We let the corresponding wave vectors be denoted by k_1 and k_2, and let $k_1 \leqslant k_2$. For the frequencies $\omega > \omega_\parallel$, k_1- and k_2-waves belong to branches with different polarizations [$\omega_+(k)$ and $\omega_-(k)$], while for the frequencies $\omega < \omega_\parallel$, they belong to the same branch, $\omega_-(k)$. This branch has a minimum $\omega_-(k_{\min}) = \omega_\parallel - \Delta$ (it corresponds to the lowest frequencies allowed for the propagating wave). This minimum is achieved at $k = k_{\min} \simeq (\omega_\parallel^2/2c^2)\gamma$. Its depth

$$\Delta = \omega_\parallel - \omega_-(k_{\min}) \approx \frac{\gamma^2\omega_\parallel^2}{4A_\parallel c^2} \tag{50}$$

strongly depends not only on $\omega_\parallel$ and γ but also on the value of $A_\parallel$. It is clear that the branch $\omega_-(k < k_{\min})$ (k_1-waves) has negative group velocity at $\omega < \omega_\parallel$, because the frequency of this part of the branch decreases as the wave vector k_1 increases. All other parts of the spectrum in (49) have usual (positive) group velocities. At the bottom $\omega_\parallel - \Delta$ of the allowed frequency interval, $k_1 = k_2 = k_{\min}$, and both waves have a vanishing group velocity.

It is interesting to note that similarly to the case illustrated in Fig. 1, the waves with negative group velocity in Fig. 5b occur in the frequency range forbidden for electromagnetic waves in the absence of magnetic resonance. In a gyrotropic medium, only one of two waves with different polarizations has a negative group velocity, and the other one has the ordinary positive group velocity dispersion.

Because left- and right-hand polarized waves propagate with different phase velocities, linearly polarized light experi-

ences rotation of its plane of polarization. It is useful to note that the exact relation

$$k_2 - k_1 = \frac{\gamma\omega^2}{c^2} \tag{51}$$

for the difference between the wave vector magnitudes k_2 and k_1 at the same frequency $\omega > \omega_{\parallel}$ follows from Eqn (48) for an arbitrary dependence of $\varepsilon(\omega)$ and $\gamma(\omega)$ on ω. Its exact counterpart for frequencies $\omega < \omega_{\parallel}$ determines the sum of the wave vector magnitudes at the same frequency:

$$k_2 + k_1 = \frac{\gamma\omega^2}{c^2}\,. \tag{52}$$

Equations (51) and (52) lead to the same result for the rotatory power [46],

$$\rho = \frac{\gamma\omega^2}{2c^2}\,,$$

both below and above the frequency $\omega_{\parallel}$ (see Ref. [46]). Measuring ρ (the rotation of the polarization plane per unit length of the ray passage) thus provides experimental access to the gyrotropy parameter $\gamma(\omega)$.

Dissipation can considerably complicate the practical realization of negative refraction conditions. We give a quantitative illustration in the above case of a gyrotropic medium in the vicinity of the longitudinal frequency. It is obvious that the dispersion of the waves shown in Fig. 5b retains its physical meaning only if in the region of frequencies around $\omega_{\parallel}$, the minimum depth Δ in (50) is large enough in comparison with the dissipative width Γ of *transverse* electromagnetic waves. As discussed in Ref. [46] with several examples, this restriction in fact leads to quite demanding requirements on the 'allowed' magnitudes of gyrotropy and dissipation.

It is shown in Ref. [46] that the ordinary specular reflection can be useful in experimental studies of the question of whether a gyrotropic material can be appropriate for the observation of negative refraction: the interesting frequency range around $\omega_{\parallel}$ would be directly detectable in the features of the reflection spectrum of linearly polarized incident light.

4.4 Surface polaritons

Surface waves can also have negative group velocity. As an example, we consider surface polariton waves near the resonance with the modes of a surface transition layer. It is known that a surface transition layer (e.g., a thin film on a substrate) can drastically alter the dispersion of surface polaritons if they are in resonance with vibrational or electronic excitations of the layer [47]. If chosen properly, the transition layer can lead to surface polariton dispersion curves exhibiting regions of negative group velocity.

We consider a system composed of a thin film of thickness $d \gg a$ (a being the lattice constant) with a dielectric permittivity $\varepsilon(\omega)$, placed between two semi-infinite media with dielectric permittivities $\varepsilon_1(\omega) > 0$ and $\varepsilon_2(\omega) < 0$. Surface polaritons in this system exist in a certain frequency range, and their dispersion curve $\omega(k)$ is determined by the equation [47]

$$\frac{\varkappa_1}{\varepsilon_1} + \frac{\varkappa_2}{\varepsilon_2} + k^2 p + \frac{\varkappa_1}{\varepsilon_1}\,\frac{\varkappa_2}{\varepsilon_2}\,q = 0\,, \tag{53}$$

where k is the magnitude of the *two-dimensional* wave vector of surface polaritons directed along the interface (the medium is supposed to be isotropic in the interface plane). The parameters in Eqn (53) are defined as

$$\varkappa_i = \sqrt{k^2 - \frac{\omega^2}{c^2}\,\varepsilon_i}\,, \qquad i = 1, 2\,,$$
$$q = (\varepsilon - \varepsilon_2)d\,, \qquad p = \left(\frac{1}{\varepsilon} - \frac{1}{\varepsilon_2}\right)d\,,$$

and $kd \ll 1$ is assumed. For $d = 0$, the parameters p and q also vanish, and Eqn (53) reduces to the familiar dispersion equation of surface polaritons at a single interface between two semi-infinite media. The effect we describe arises due to the thin film, that is, due to $d \neq 0$. However, because $kd \ll 1$, it is clear that the terms proportional to d in Eqn (53) become especially important for those frequency regions where either the dielectric permittivity $\varepsilon(\omega) \approx 0$ (longitudinal resonance) or its inverse $\varepsilon^{-1}(\omega) \approx 0$ (transverse resonance). The effect of a thin film on the surface polariton dispersion is often stronger in the first of these two cases.

To illustrate the substantial effect of a thin film overlayer on surface polaritons near a resonance, we consider a thin metallic film covering a metal substrate. In this case, $\varepsilon_1 = 1$ and we can approximate the optical responses of the film and of the substrate by the respective Drude-model expressions

$$\varepsilon(\omega) = 1 - \frac{\omega_{\mathrm{p}}^2}{\omega^2}\,, \qquad \varepsilon_2(\omega) = 1 - \frac{\omega_{2\mathrm{p}}^2}{\omega^2}\,. \tag{54}$$

In the absence of the thin film, the surface plasmon–polaritons of the substrate exist in the frequency interval

$$0 < \omega < \frac{\omega_{2\mathrm{p}}}{\sqrt{2}}\,.$$

We now let $\omega_{\mathrm{p}} \ll \omega_{2\mathrm{p}}$, such that the surface polaritons of the substrate are resonant with the plasmons of the thin metallic film at a frequency $\omega \approx \omega_{\mathrm{p}}$.

Figure 6 shows the dispersion of polaritons that occur in such a system. We use the value of the ratio $(\omega_{2\mathrm{p}}/\omega_{\mathrm{p}})^2 = 15.2$ and the film thickness $d \approx 26$ Å corresponding to the experiments in [48] made for an aluminum substrate coated by a silver film. Due to the resonance, the polariton spectrum in Fig. 6 is split into two branches with a frequency gap appearing between them. Evidently, the lower branch of the polariton spectrum exhibits two modes for a given frequency ω. The mode with larger k (denoted as k_2) is the additional surface polariton wave with a negative group velocity. It is clearly seen in the figure that the polariton wave frequency decreases linearly with the increase in the wave vector; the reason for this can be readily revealed in the following analysis.

For $\omega \approx \omega_{\mathrm{p}} \ll \omega_{2\mathrm{p}}$, the magnitudes of dielectric permittivities (54) must satisfy the conditions

$$-\varepsilon_2(\omega) \gg 1\,, \qquad \left|\varepsilon(\omega)\right| \ll 1\,.$$

The second and the fourth terms in Eqn (53) can then be neglected, because $|\varkappa_2/\varepsilon_2| \ll \varkappa_1$ and $\left|(\varkappa_1\varkappa_2/\varepsilon_1\varepsilon_2)q\right| \ll k^2|p|$. At large enough k, it is true that $\varkappa_1 \approx k$, and Eqn (53) immediately yields the following dispersion equation for the polariton:

$$\omega(k) \approx \omega_{\mathrm{p}}\left(1 - \frac{kd}{2}\right). \tag{55}$$

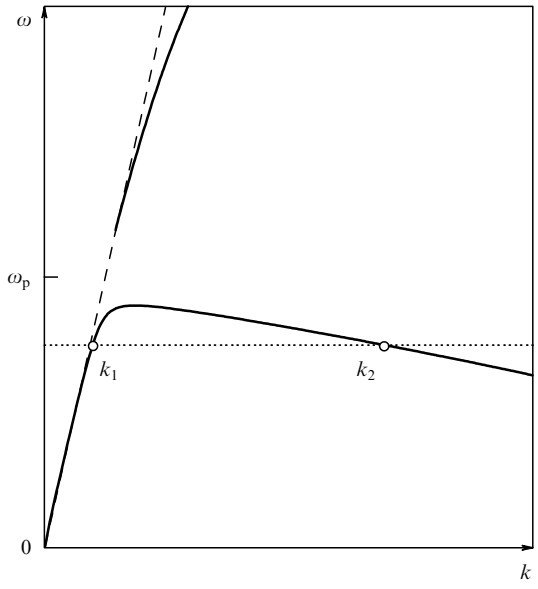

Figure 6. Dispersion of surface polaritons at a resonance with vibrations in a thin overlayer. The resonance is at a frequency ω_p. Both the gap in the polariton spectrum and the branch with negative group velocity (exemplified by wave vector k_2 at a given frequency) are clearly seen.

Equation (55) describes the negative group velocity of the lower polariton branch seen in Fig. 6.

Thermally excited radiation from such surface polaritons with negative group velocity was observed experimentally in Ref. [49] in systems of ZnSe films on Al and Cr substrates. Also, experiments in [50] for thin films of LiF on a sapphire substrate confirmed the square-root dependence of the frequency gap magnitude on the film thickness (the magnitude of the gap being proportional to $\sqrt{d}$) following from Eqn (53). This gap can increase substantially as the resonant plasma frequency increases. In fact, a gap of the magnitude 0.4 eV was reported in Ref. [48] for surface plasmons on an aluminum substrate coated by a silver film of $d = 2.6$ nm, in good agreement with the theoretical estimate. The splitting in the surface polariton dispersion curve has also been observed in systems consisting of an organic monolayer [51] and of a thin organic film [52] on silver substrates.

The theory of surface wave propagation with additional surface waves and the diffraction of waves at the edge of the film taken into account was developed in Ref. [53]. The presence of the diffraction and conversion of surface waves into bulk radiation and, vice versa, of bulk radiation into surface waves, considerably complicates the problem of finding ABCs for surface waves.

5. Magnetic permeability at optical frequencies

In Sections 3.1, 3.2, and 3.4, we have already discussed certain aspects of the correspondence between two approaches used in the electrodynamics of continuous media. One of the approaches is based on taking spatial dispersion into account; in the framework of this approach, three fields $(\mathbf{E}, \mathbf{D}, \mathbf{B})$ supplemented by matter equation (9) with the dielectric tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ are considered. In the other, perhaps more familiar (so-called 'symmetric') approach, all four fields $(\mathbf{E}, \mathbf{D}, \mathbf{B}, \mathbf{H})$ are explicitly considered, and the matter equations

$$\mathbf{D} = \varepsilon(\omega)\mathbf{E}\,, \qquad \mathbf{B} = \mu(\omega)\mathbf{H} \tag{56}$$

are used for monochromatic fields.

Using Eqn (56) with the Maxwell equations leads to the standard dispersion law (3) for plane waves propagating in a *spatially uniform* medium.

In this section, we discuss conditions at which the magnetic permeability $\mu(\omega)$ in Eqn (56) may retain its physical meaning in the description of a continuous medium. An analysis of this question for natural materials has been provided in the textbook by Landau and Lifshits [6] with the conclusion that "unlike $\varepsilon(\omega)$, when the frequency increases, the magnetic permeability $\mu(\omega)$ ceases to have any physical meaning at relatively low frequency." What does this mean? As is well known, a macroscopic description involves spatial averaging and, therefore, necessitates that a microscopic length a (there can be more than one such length) characterizing the medium be much smaller than the length of the spatial variation of the macroscopic electromagnetic fields (that is, for instance, than the wavelength λ of the electromagnetic waves in the medium: $a \ll \lambda$). For natural materials, a is typically of an atomic or molecular size, like a crystal lattice constant, or of the order of the mean free path of charge carriers.

In many recent studies that followed Pendry's work [54], the macroscopic Maxwell equations are used to study wave propagation and negative refraction in artificial periodic or amorphous structures (meta-materials). References to earlier investigations within the same approach in both amorphous and periodic artificial structures can be found in Ref. [55]. These materials are composites comprising objects of various shapes (spheres, rods, pillars, etc.). The geometrical sizes of these constituent objects ('artificial molecules') and the corresponding lattice constants (new length scale a) can be hundreds of times larger than in natural materials. As an example, we mention a structure comprised of pairs of gold nano-pillars of about 80 – 200 nm studied in Ref. [56] within the range of vacuum wavelengths from 400 to 700 nm. Another example is a recent work [57], in which a double-periodic array of pairs of parallel gold nano-rods was used, with the rods measuring $780 \times 220 \times 50$ nm. The wavelength of the illuminating light varied between 500 and 2000 nm. The structures used in Refs [56, 57] were developed with the goal of fabricating meta-materials with a negative refractive index at optical frequencies. However, in both cases, not bulk materials but 'monolayers' were in fact fabricated.

Two different ways to analyze the properties of such a composite can be distinguished. Because the size of the objects is much larger than the atomic sizes, each of the objects can be described within the framework of the usual macroscopic theory, being characterized, e.g., by appropriate $\varepsilon(\omega)$ and $\mu(\omega)$. The wave propagation in the composite can then be studied by applying the Maxwell boundary conditions on the objects' surfaces, for example, within the finite-difference time domain method of computational electrodynamics [58]. Evidently, in this powerful straightforward approach, there is no need to evaluate the effective material constants of the meta-material, while customary values of $\varepsilon(\omega)$ and $\mu(\omega)$ are spatially variable. Any restriction on the meaning of $\mu(\omega)$ in this approach would be the same as for natural materials.

A different way, which is conceptually attractive and allows an analytical solution, is to perform a 'secondary averaging' over composite's structure and to use the macroscopic Maxwell equations in the resulting effectively uniform medium. This method is applicable as long as $\lambda \gg a$, i.e., as long as the medium can be characterized by the corresponding *effective* permittivity and permeability. It is important that wave propagation can be considered similarly to the way typically used in natural homogeneous condensed matter with $a \ll \lambda$ only if coordinate-independent effective parameters ε, μ, or $\varepsilon_{ij}(\omega, \mathbf{k})$ can be introduced. It turns out, however, that the notion of the effective permeability $\mu(\omega)$ has a restricted range of applicability [6].

5.1 The magnetic moment of a macroscopic body

A difficulty with the definition of the physical meaning of $\mu(\omega)$ at higher frequencies, which is important for theory as well as for the interpretation of experiments, is related in [6] to the fact that it may be impossible to 'measure' the permeability by measuring the *total* induced magnetic moment of a macroscopic body. Indeed, the induced macroscopic current density $\mathbf{J}$ in time-dependent fields arises not only from the magnetization

$$\mathbf{M} = \frac{\mathbf{B} - \mathbf{H}}{4\pi} \tag{57}$$

but also from the dielectric polarization $\mathbf{P} = (\mathbf{D} - \mathbf{E})/4\pi$:

$$\mathbf{J} = c\nabla \times \mathbf{M} + \frac{\partial \mathbf{P}}{\partial t}\,. \tag{58}$$

Equation (58) follows, on the one hand, directly from the averaged macroscopic Maxwell equations

$$\begin{aligned} \nabla \times \mathbf{B} &= \frac{4\pi}{c}\,\mathbf{J} + \frac{1}{c}\,\frac{\partial \mathbf{E}}{\partial t}\,, \\ \nabla \times \mathbf{H} &= \frac{1}{c}\,\frac{\partial \mathbf{D}}{\partial t}\,, \end{aligned} \tag{59}$$

and, on the other hand, from the derivation of this current as the average of the microscopic current density $\mathbf{J} = \langle \rho \mathbf{v} \rangle$ using positions and velocities of charged particles in the medium [33, 35].

The induced total magnetic moment

$$\mathbf{M}^{\mathrm{tot}} = \frac{1}{2c}\int (\mathbf{r} \times \mathbf{J})\, \mathrm{d}V$$

of a macroscopic body must therefore also have two corresponding contributions:

$$\mathbf{M}^{\mathrm{tot}} = \mathbf{M}_1^{\mathrm{tot}} + \mathbf{M}_2^{\mathrm{tot}}\,, \tag{60}$$

where

$$\mathbf{M}_1^{\mathrm{tot}} = \int \mathbf{M}\, \mathrm{d}V, \tag{61}$$

$$\mathbf{M}_2^{\mathrm{tot}} = \frac{1}{2c}\int \left(\mathbf{r} \times \frac{\partial \mathbf{P}}{\partial t}\right) \mathrm{d}V. \tag{62}$$

Thus, the magnetization $\mathbf{M}$ has the physical meaning of a magnetic moment of a unit volume of the body when the contribution (62) from the time-dependent dielectric polarization can be omitted in (60). Only if this contribution can be neglected can the permeability $\mu(\omega)$ be treated as the physical quantity that determines the magnetic moment of a unit volume.

We note that the analogous problem does not arise for the electric dipole moment [6], because the total electric dipole moment is defined by an expression similar to (61): $\mathbf{P}^{\mathrm{tot}} = \int \mathbf{P}\, \mathrm{d}V$.

A natural question arises: under which conditions is the contribution $\mathbf{M}_2^{\mathrm{tot}}$ into $\mathbf{M}^{\mathrm{tot}}$ small? Using Maxwell equation (59) and the definitions of $\mathbf{M}$ in (57) and of $\mathbf{P}$, we can immediately evaluate relative contributions to the induced current (58) for a monochromatic electromagnetic wave. For the magnetic current to dominate, i.e., to have

$$|c\nabla \times \mathbf{M}| \gg \left|\frac{\partial \mathbf{P}}{\partial t}\right|,$$

which allows neglecting the term $\mathbf{M}_2^{\mathrm{tot}}$, the following inequality must be satisfied:

$$R(\omega) = \left|\frac{\varepsilon(\omega)\big(\mu(\omega) - 1\big)}{\varepsilon(\omega) - 1}\right| \gg 1\,. \tag{63}$$

Therefore, if $R(\omega) \gg 1$ for given $\varepsilon(\omega)$ and $\mu(\omega)$, then $\mathbf{M}_2^{\mathrm{tot}}$ can be neglected, and the quantity $\mu(\omega)$ entering one of equations (56), with reasonable accuracy, defines the magnetic moment of a unit volume in the field of the plane electromagnetic wave propagating in the medium. If, on the contrary, inequality (63) is not satisfied, then the magnetic moment of a unit volume is dominated by the contribution of the electric polarization current and the physical meaning of the magnetic permeability $\mu(\omega)$ (which, in particular, defines the magnitude of the refraction coefficient of the waves) is unclear. Because $\mu(\omega)$ no longer determines the magnetic moment of a unit volume, the appropriateness of using this quantity and, consequently, the symmetric approach, is doubtful. Nevertheless, the physical meaning of the quantity $\mu(\omega)$ can also be defined in this case if it can be independently measured. It is not the best way to define the magnetic permeability $\mu(\omega)$ using the plane wave, as was done to obtain inequality (63), because the electromagnetic wave does not provide the most favorable conditions for reduction of the value of $\mathbf{M}_2^{\mathrm{tot}}$ due to a relatively strong electric field of the wave. Instead, as discussed in Ref. [6], a small macroscopic body can be placed in a time-dependent (monochromatic) magnetic field produced by some external current density $\mathbf{J}_{\mathrm{ext}}$. The electric field must be relatively weak, such that the contribution of electric polarization into the magnetic moment of a unit volume can be reduced. To solve the problem analytically, we consider a cylindrical sample of length L and radius l placed in a solenoid. Let an external circular current produce a magnetic field inside the solenoid. The smallness of the sample in this geometry means that

$$l \ll \lambda\,. \tag{64}$$

But the sample must still be macroscopic,

$$l \gg a\,, \tag{65}$$

for the very notion of the (effective) macroscopic permeability to be valid.

If Eqn (64) is satisfied, then the magnetic field in the sample is mostly produced by the external current. Let H be the magnitude of this uniform field. This field leads to the

uniform magnetization $M = \big(\mu(\omega) - 1\big)H/4\pi$ of the sample, and its contribution (61) to the total magnetic moment is

$$M_1^{\rm tot} = \left| l^2 L \big(\mu(\omega) - 1\big)\, \frac{H}{4} \right| . \tag{66}$$

But the time-dependent magnetic field also induces the electric field in the sample according to the Maxwell equation

$$\nabla \times \mathbf{E} = -\frac{1}{c}\, \frac{\partial \mathbf{B}}{\partial t} .$$

In our geometry, the magnitude of this field varies with the distance x from the cylinder axis as $E = \big|\omega\mu(\omega)Hx/2c\big|$. The magnitude of the dielectric polarization current density is therefore $|\partial P/\partial t| = \big|\omega^2\mu(\omega)\big(\varepsilon(\omega) - 1\big)Hx/8\pi c\big|$, and hence the second contribution (62) to the total magnetic moment is

$$M_2^{\rm tot} = \left| l^4 L\, \omega^2 \mu(\omega) \big(\varepsilon(\omega) - 1\big)\, \frac{H}{32c^2} \right| . \tag{67}$$

Equations (66) and (67) show that for the magnetization contribution to dominate, i.e., for

$$|\mathbf{M}_1^{\rm tot}| \gg |\mathbf{M}_2^{\rm tot}|$$

to be true, the following inequality must be satisfied:

$$\frac{8c^2}{\omega^2 l^2} \left| \frac{1 - 1/\mu(\omega)}{\varepsilon(\omega) - 1} \right| \gg 1 . \tag{68}$$

With the corresponding wavelength $\lambda(\omega) = 2\pi c/\omega\sqrt{\varepsilon\mu}$ for plane waves in the medium used instead of the frequency ω, criterion (68) can be rewritten as

$$\frac{2}{\pi^2}\, R(\omega) \left(\frac{\lambda(\omega)}{l} \right)^2 \gg 1 . \tag{69}$$

If criterion (68) is satisfied, the quantity $\mu(\omega)$ preserves its physical meaning independently of whether inequality (63) is satisfied. This criterion is 'weaker' than Eqn (63) owing to condition (64). Clearly, the numerical coefficients in Eqns (68) and (69) are affected by the particular shape of the sample considered, and the frequency intervals where condition (64) is not satisfied must be excluded from the above consideration.

We note that inequalities (63) and (68) naturally follow from the comparison of the medium response contributions to the generalized dielectric permittivity $\varepsilon_\perp(\omega, k) - 1$ in Eqn (20): these inequalities require that the contribution of the spatial dispersion term $\propto k^2$ be larger than the term without the spatial dispersion. For a given frequency ω, inequality (63) corresponds in Eqn (20) to the wave vector k of the wave in the medium, while inequality (68) corresponds to the wave vector $k \sim 1/l$, i.e., $1/k$ is of the order of the sample size.

For inequality (68) to be better satisfied, the size of the sample l should be made as small as possible but still large enough for the sample to remain macroscopic [see Eqn (65)]. Evidently, the smaller the microscopic scale a, the smaller l can be, and the easier it is to satisfy inequality (69). The smallest possible magnitudes of a (of atomic or molecular size) are found in natural materials. The presence of the factor ω^2 in the denominator in the left-hand side of (68) clearly shows that this criterion can be easily satisfied at sufficiently low frequencies because the ω-dependence of $\varepsilon(\omega)$ and $\mu(\omega)$ is then weak. It becomes in general harder and harder to satisfy criterion (68) as the frequency increases.

Of course, the possibility of satisfying this criterion also depends on the details of the frequency dependence of the functions $\varepsilon(\omega)$ and $\mu(\omega)$: for instance, using model expressions (4) and (5), we can write the left-hand side of Eqn (68) as

$$\frac{8c^2}{\omega^2 l^2}\, \frac{F_{\rm m}}{F_{\rm e}} \left| \frac{\omega_\perp^2 - \omega^2}{\omega_{\rm mz}^2 - \omega^2} \right| . \tag{70}$$

The magnitude of expression (70) has a 'hump' in a narrow region around the zero of the magnetic permeability $\omega_{\rm mz}$, which is in reality substantially mitigated by dissipation. Apart from that, the magnitude of expression (70) is determined by the factor

$$\frac{c^2}{\omega^2 l^2}\, \frac{F_{\rm m}}{F_{\rm e}} \sim \frac{a^2}{l^2} , \tag{71}$$

where the estimate in the right-hand side is made for natural (consisting of atoms or molecules) materials at optical frequencies $\omega \sim \omega_\perp \sim \omega_{\rm mz}$ [see Eqn (34)]. Evidently, for a given macroscopic size of sample (65), inequality (68) cannot be satisfied at optical frequencies in general. A measurement or a model calculation of the *total* magnetic moment of a macroscopic body in this frequency region is not related to the magnetization $\mathbf{M}$, with the exception, perhaps, of some frequency intervals.

It seems reasonable to assume the same estimate (71) and the same conclusion to be valid for a meta-material built of sufficiently small ($a \ll \lambda$) metallic or other structures if the electric and magnetic resonance frequencies are of the same order as $\omega_{\rm p}$, while the quantity equivalent to $F_{\rm m}/F_{\rm e}$ is of the order of $\omega_{\rm p}^2 a^2/c^2$. Results for various structural shapes suggested in the literature can be used to check whether a condition similar to Eqn (68) is satisfied and to establish the frequency range where the permeability $\mu(\omega)$ retains its physical meaning within a macroscopic description of the sample. Although the characteristic length scale a in meta-materials greatly exceeds the atomic size (it is a few dozen or hundred nanometers), it is obvious that the range of frequencies ω with the reasonably satisfied inequalities

$$a \ll l \ll \lambda(\omega) \tag{72}$$

must in general shift to lower frequencies as the size a increases. In fact, it is possible that in meta-materials with larger a, inequalities (72) cannot be satisfied in a wide range of frequencies, but the wavelength λ is still appreciably larger than a. It is then impossible to measure the permeability (and, consequently, to define its physical meaning) with the help of the aforementioned solenoidal setup. In this situation, only criterion (63) is left for estimates. We are not aware of a better configuration to 'measure' the permeability.

So far, with $\lambda \gg a$, a meta-material can of course be treated as a continuous medium, and the approach based on taking spatial dispersion into account and using the tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ is a powerful alternative to the approach involving $\varepsilon(\omega)$ and $\mu(\omega)$ at the frequencies where $\mu(\omega)$ loses its physical meaning. But it follows from our discussion in Section 3 that as long as the spatial dispersion is restricted to terms $\propto k^2$ [e.g., as in Eqn (22)], the formal description of transverse polaritons within the $\varepsilon(\omega) - \mu(\omega)$ formalism is adequate if

some effective $\mu(\omega)$ is introduced via Eqn (20) with the resulting standard expression (3) for the refractive index. But the discussion in this section implies that $\mu(\omega)$ thus defined cannot be related to the total magnetic moment of a macroscopic body at optical frequencies in general, because only the part of spatial dispersion due to magnetic-dipole-allowed transitions is taken into account. The method based on the account of spatial dispersion, of course, also allows analyzing other types of dispersion and the corresponding qualitatively new effects (such as additional polariton waves), which are completely missed in the description of the material properties of the medium in terms of $\varepsilon(\omega)$ and $\mu(\omega)$.

As soon as any of the structural sizes a in meta-materials becomes comparable to the wavelength λ in the medium, the analysis of the wave propagation in the composite with the help of the electrodynamics of continuous media ceases to be possible: as already mentioned, a composite cannot be treated as an 'effectively continuous' medium, and descriptions involving spatially variable (position-dependent) material response functions must then be used.

An analysis of the applicability of the $\varepsilon(\omega)-\mu(\omega)$ approach to the already published results claiming the observation of negative refraction in the optical domain in meta-materials is of current importance. Unfortunately, the data on the dispersions of $\varepsilon(\omega)$ and $\mu(\omega)$ are not always available in the papers. In some cases, the reported imaginary part of the refractive index is around or even larger than its real part, which does not allow taking the published statements seriously. It is also important that the experimentally studied structures be truly three-dimensional bodies, rather than two-dimensional monolayers: even for artificial materials, monolayers should be taken into account only via a corresponding modification of the boundary conditions. Experiments with 'monolayers' have, in general, absolutely no relation to negative refraction in bulk materials.

6. Related interesting effects

6.1 The generation of harmonics in a medium with negative group velocity

The generation of harmonics in media with negative group velocity can have some peculiarities. Here, following Ref. [8], we briefly consider one of the interesting effects on a qualitative level. We consider a semi-infinite medium that supports waves with a negative group velocity in some frequency interval. Usually, the spectral width $\Delta\omega$ of this interval is relatively narrow: $\Delta\omega \ll \omega$. We suppose that a laser beam of a frequency ω_l is incident on the medium from a vacuum, with ω_l being within the range $\Delta\omega$. Then the frequencies of the second ($2\omega_l$) and higher harmonics belong to those regions of the spectrum where the medium supports waves with positive group velocities. As is known, the sources of harmonics generation are determined by a tensor product of the nonlinear susceptibilities $\chi^{(2)}, \chi^{(3)}, \ldots$ and the amplitudes of the fields in the medium. For example, the source of the generation of the second harmonic is given by $\chi^{(2)}_{ij} E_i(\omega, \mathbf{k}) E_j(\omega, \mathbf{k})$, and similarly for higher harmonics. At low intensities, the field $E(\omega, \mathbf{k})$ can be calculated in the linear approximation neglecting the nonlinear interaction. Because the incoming refracted wave is in the frequency interval of negative refraction, its wave vector $\mathbf{k}$ is directed from the bulk of the medium to its surface, as depicted in Fig. 7a. Then the wave vector of the source, for instance, of the second harmonic, is $2\mathbf{k}$, and is also directed towards the vacuum – medium interface. On the other hand, the wave vector of the wave at the frequency $2\omega_l$, which carries the energy from the surface into the bulk of the nonlinear medium, is directed from the interface into the bulk of the medium. Therefore, the wave vectors of the source of the second harmonic and of this normal transmitted wave are phase-mismatched, their interaction is weak, and this wave is also excited only weakly. This mismatch then causes the dominant part of the energy from the source of the second harmonic to be transferred to the second harmonic propagating in the vacuum away from the interface, as schematically shown in Fig. 7a. Thus, a medium with negative group velocity, as it were, reflects the second harmonic generated by the incident laser beam, and thus works as an effective 'mirror.' Details of the corresponding calculations can be found in Refs [8, 59] (see Ref. [60] for the discussion of the generation of harmonics of acoustic waves in one-dimensional phononic crystals with negative refraction). Experimental studies of nonlinear effects are only beginning; we can mention paper [61], where an enhancement of the intensity of the reflected second harmonic in transmission lines with negative group velocity caused by nonlinear effects

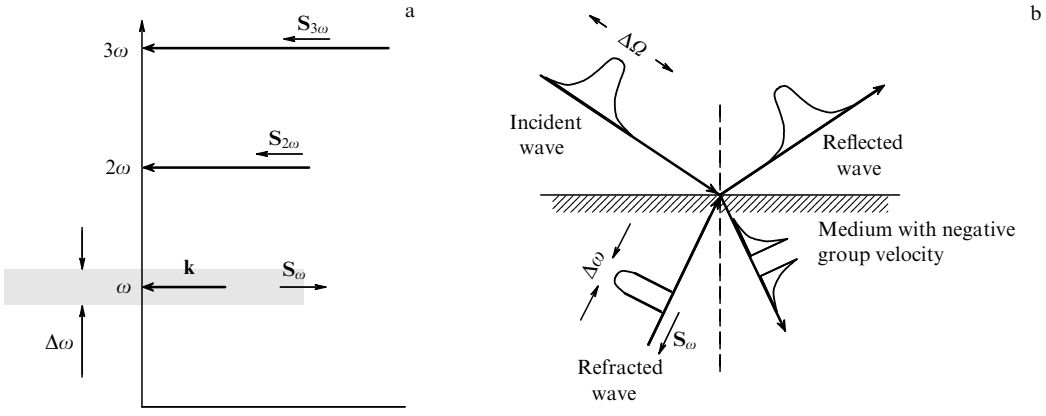

Figure 7. Schematic illustrations for the effects discussed in Section 6. (a) Harmonic generation. The frequency ω of the incident wave belongs to a narrow interval $\Delta\omega$ of frequencies for which the waves in the medium have a negative group velocity. The energy transferred to higher harmonics 2ω and 3ω mostly propagates (the Poynting vector $\mathbf{S}$) in the reflected mode. (b) An ultra-short pulse ($\Delta\Omega > \Delta\omega$) incident on a medium with negative group velocity leads to the appearance of two refracted pulses with different spectral contents.

has been observed. Other nonlinear properties of artificial materials with negative group velocity are discussed in Refs [62 – 64].

6.2 Ultra-short pulse propagation in a medium with negative group velocity

Ultra-short pulses are currently available in a wide frequency range, from the terahertz region to the far ultraviolet. An interesting manifestation of negative refraction may occur when the spectral width $\Delta\Omega$ of a pulse is appreciably larger than the spectral width $\Delta\omega$ of the frequency range where waves with negative group velocity exist in a negative-refraction material [8]. Qualitatively speaking, the ultra-short pulse can be decomposed into a sum of its Fourier components, the propagation of each component can then be followed, and the components composed after their propagation through the medium.

If $\Delta\Omega \gg \Delta\omega$, then the pulse incident on the medium with negative group velocity is expected to split into three outgoing pulses with different spectral content, as schematically shown in Fig. 7b. The reflected pulse has approximately the same spectral content as the incident pulse. The two transmitted pulses have different propagation directions and different spectral contents. The central part of the pulse spectrum (of the width $\Delta\omega$) experiences negative refraction at the interface, but the components from the 'wings' located outside the interval $\Delta\omega$ propagate according to the rules of ordinary 'positive' refraction. Therefore, the frequency interval $\Delta\omega$ can be determined by spectroscopical methods examining spectra of differently refracted transmitted pulses.

Interesting effects can also be expected when ultra-short pulses are used for harmonic generation and wave mixing, because higher harmonics also propagate in an unusual way. Only some part of the spectrum of the input or output signals experiences negative refraction, and therefore the output pulses of transmitted and reflected light can be drastically different: the energy, pulse shape, spectral content, and direction of propagation may differ from those expected in an ordinary nonlinear medium. The details of the description are complicated and depend on the spectral content of the ultra-short pulse and on the material with negative refraction.

7. Conclusion

It was a pleasure for us in this review to once more pay homage to L I Mandel'shtam, who pointed out in the early 1940s that negative refraction of waves at interfaces occurs as a result of a negative group velocity of waves in one of the interfacing media [1 – 3]. The understanding of this fact is a motivation for drawing special attention to various factors that may affect the dispersion law $\omega(\mathbf{k})$ of waves propagating in the medium.

The most general approach to analyze such factors for electromagnetic waves in an effectively homogeneous medium is based on taking spatial dispersion into account. In the framework of this approach, a generalized dielectric tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ is introduced, which represents the response of the medium to perturbations of the frequency ω and wave vector $\mathbf{k}$. Normal waves (polaritons) with negative group velocity can propagate in a medium (whether in natural or in artificial meta-materials) when the spatial dispersion (the dependence of the dielectric tensor on $\mathbf{k}$) is strong enough. A particular case where such a situation arises (which corresponds to the spatial dispersion $\propto k^2$) is better known as the case of a material in which the dielectric permittivity $\varepsilon(\omega)$ and the magnetic permeability $\mu(\omega)$ are simultaneously negative. The approach based on taking spatial dispersion into account also allows working in the optical frequency region, where $\mu(\omega)$ loses its traditional physical meaning, and even in situations where the medium does not exhibit a response of the magnetic dipole type.

The tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ also allows considering more complicated matter equations and the resulting qualitatively new effects, such as additional polariton waves *in a unified fashion*. In this review, we have used this approach to describe several physical systems in which conditions for the propagation of polaritons with negative group velocity at optical frequencies exist. Our examples included gyrotropic and nongyrotropic systems and bulk and surface polariton waves. We hope that these examples can be useful in choosing materials suitable for experiments.

We focused mainly on the physical origin of the appearance of polaritons with negative group velocity. Due to this, we were unable to discuss many important factors that affect the possibility of practically realizing the effects related to the existence of the negative refraction. One of those factors is the presence of dissipation, which is, of course, the problem common to all frequency intervals. This means, for instance, that the crystals with intensive and narrow excitonic resonances deserve special attention. Another problem consists in the relatively low efficiency of the excitation of additional polaritons due to wave vector mismatch. Certain schemes have been proposed to increase the efficiency of their investigation for crystals with a positive group velocity of additional waves. These schemes can possibly also be applied to crystals with negative refraction.

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