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Negative Index of Refraction in Anisotropic Nonmagnetic Materials

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We analyze the possibility of negative refractive index and/or negative refraction of energy flow in a uniaxial nonmagnetic dielectric material. It is shown that an extraordinary p-polarized electromagnetic wave can exhibit a negative refraction provided the parallel (with respect to the optical axis) and perpendicular components of the permittivity have different signs. Such a situation can be encountered near phonon resonant lines, as for example in mercurous halides. The dispersion curves of surface and guided modes in an anisotropic nonmagnetic slab are calculated. The effect of space dispersion is briefly discussed.

Keywords Anisotropic dielectric; negative refractive index; surface modes; guided modes

Introduction

In the past few years, materials showing simultaneously negative permittivity ε and permeability μ in a particular frequency range have been intensively investigated. These, so called negative refractive materials (NRM), exhibit unusual electromagnetic (EM) wave propagation properties predicted by Veselago [1] almost 40 years ago and recently demonstrated experimentally in the microwave range [2, 3]. The essential feature of isotropic NRM is that the phase $\mathbf{v}_{ph}$ and group $\mathbf{v}_g$ velocities of EM waves have opposite directions. Straightforward consequences of this property are the negative refraction of the EM beams described by a negative refractive index n_{ph} introduced into the Snell's law, a reversed Doppler shift, negative radiation pressure and an obtuse angle for Cherenkov radiation [1]. Of particular interest is Veselago's perfect lens [1], a slab of NRM with $\varepsilon = \mu = -1$, which could dramatically improve the resolution in 3d imaging. Physics of isotropic NRM was recently reviewed in detail by Ramakrishna [4].

Anisotropic Dielectrics

A study of EM wave propagation in magnetic anisotropic materials for which not all of the principal components of the permittivity and/or the permeability tensors have the same sign has been reported recently; in certain classes of anisotropy negative refraction and other related properties have been found [5–7]. It is well known [8] that in an anisotropic

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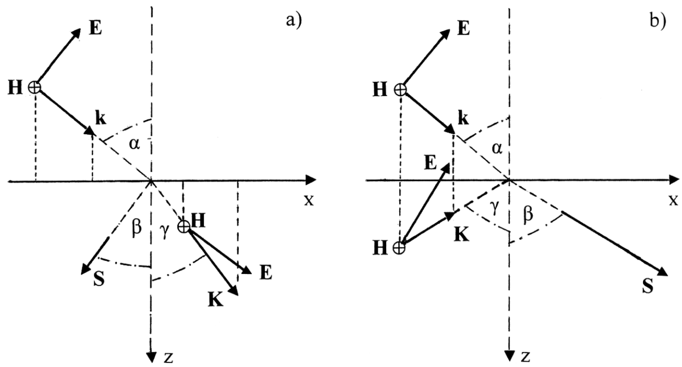

Figure 1. The refraction of a TM wave: a) $\varepsilon_x = 0.5$, $\varepsilon_z = -0.5$; b) $\varepsilon_x = -0.5$, $\varepsilon_z = 0.5$; with this special choice of the values of the permittivity components $\beta = -\gamma$ in both cases. The magnetic field **H** is oriented in the positive direction of the y-axis.

nonmagnetic ($\mu_{\mathrm{ik}} = \delta_{\mathrm{ik}}$) material $\mathbf{v}_{\mathrm{g}}$ makes an acute angle with $\mathbf{v}_{\mathrm{ph}}$, i.e. with the wave vector **k** of the monochromatic plane wave. (If the spatial dispersion of the permittivity is taken into account, the angle between $\mathbf{v}_{\mathrm{g}}$ and **k** may assume all values between 0 and π for different directions of wave propagation [8].) A question arises: which kind of dielectric anisotropy can lead to the negative refraction? Indeed, it has been recently shown that in a planar waveguide with anisotropic dielectric core and metallic waveguide boundaries negative refraction can be achieved [9].

The aim of this paper is to provide a systematic study of EM wave propagation in a uniaxial anisotropic nonmagnetic material. We study both a semi-infinite medium and a plane-parallel slab with the z-axis perpendicular to the sample surface. The EM wave propagation takes place in the (x,z)-plane; the principal axes of the permittivity tensor of the sample are along x, y, and z. Since, obviously, the TE waves (ordinary waves; the electric field **E** perpendicular to the plane of incidence $y = 0$) are not affected by anisotropy of the medium, we shall consider in the following the TM waves (extraordinary waves; the magnetic field **H** perpendicular to the plane of incidence: $\mathrm{H}_y \mathrm{expi}[\mathbf{k} \cdot \mathbf{r} - \omega \mathrm{t}]$) only. The signs of the in-plane (ε_x) and normal (ε_z) components of the permittivity tensor are assumed to be different. Such a situation can be frequently encountered near phonon lines in anisotropic dielectrics, e.g., mercurous halides exhibit this type of behavior in the THz spectral range. One can distinguish two cases: (i) the in-plane component of the permittivity is negative (i.e. $\varepsilon_x < 0$, $\varepsilon_z > 0$) and (ii) the normal component of the permittivity is negative (i.e., $\varepsilon_x > 0$, $\varepsilon_z < 0$) (see Fig. 1).

Refraction

Let us first consider the refraction of a monochromatic EM plane wave with the angular frequency ω and the wave-vector $\mathbf{k}(k_x, 0, k_z)$ incident from vacuum on the surface z = 0 of a semi-infinite uniaxial dielectric with the optical axis along the z-axis of the coordinate system (see Fig. 1). From Fresnel equation we get for the z-component of the wave-vector **K** of the refracted wave (x-component is conserved, $K_x = k_x$)

$$K_z = \pm\sqrt{\varepsilon_x k_0^2 - \frac{\varepsilon_x}{\varepsilon_z} k_x^2} \tag{1}$$

where $k_0 = \omega/c$. In order to obtain a propagative refracted EM wave, we need $\varepsilon_x k_0^2$ - $(\varepsilon_x/\varepsilon_z)k_x^2 > 0$, i.e.,

$$\varepsilon_x - \frac{\varepsilon_x}{\varepsilon_z}\sin^2\alpha > 0. \tag{2}$$

The components of time averaged energy flux (the Poynting vector) **S** read as

$$\begin{aligned} S_x &= \frac{ck_x}{2\pi k_0 \varepsilon_z}\left(\frac{\varepsilon_x k_z}{K_z + \varepsilon_x k_z}\right)^2 |H_y|^2, \\ S_z &= \frac{c\varepsilon_x K_z}{2\pi k_0}\left(\frac{k_z}{K_z + \varepsilon_x k_z}\right)^2 |H_y|^2. \end{aligned} \tag{3}$$

Obviously, the energy of the EM wave is flowing from the interface into the medium and hence S_z is always positive, which means that $\varepsilon_x K_z > 0$, i.e. the actual sign of K_z in Eq. (1) is determined by the sign of ε_x component of the permittivity tensor. From (3) we get the well known relation [10] between the directions of **S** and **K** (see Fig. 1), i.e., of $\mathbf{v}_g$ and $\mathbf{v}_{ph}$,

$$\tan\beta = \frac{\varepsilon_x}{\varepsilon_z}\tan\gamma.$$

Negative refraction of **S** or **K** can be achieved when ε_x and ε_z have different sign. When $\varepsilon_x > 0$, $\varepsilon_z < 0$ (Fig. 1a), S_x is negative and the refracted wave is propagative for an arbitrary angle of incidence α. On the other hand, when $\varepsilon_x < 0$, K_z must be negative (Fig. 1b) and, as it follows from (2) for $\varepsilon_z > 0$, the angle of incidence is limited by the condition $\sin^2\alpha > \varepsilon_z$. It is interesting to note that unlike in an isotropic medium, there exist specific angles of incidence α_0 at which the wave vector **K** or the Poynting vector **S** are not refracted, i.e. $\gamma = \alpha_0$ or $\beta = \alpha_0$. These critical angles are $30^0$and 60^0 when $\varepsilon_x = 0.5$, $\varepsilon_z = –0.5$ (Fig. 1a) and $\varepsilon_x = –0.5$, $\varepsilon_z = 0.5$ (Fig.1b), respectively. It should be emphasized that the vectors **K**, **E**, **H** form a right-handed triad even when **K** refracts negatively, i.e. $K_z < 0$ (Fig. 1b). In a magnetic anisotropic material, when **K** refracts negatively, the triad becomes left-handed [7].

Let us now consider the dependence of the refraction angles β and γ on the incident angle α. It is easy to find that

$$\frac{\sin\alpha}{\sin\gamma} = \frac{K}{k_0} = n = (sign\ \ \varepsilon_x)\sqrt{\varepsilon_x + \left(1 - \frac{\varepsilon_x}{\varepsilon_z}\right)\sin^2\alpha}$$

and

$$\frac{\sin\alpha}{\sin\beta} = (sign\ \ \varepsilon_z)\sqrt{\frac{\varepsilon_z^2}{\varepsilon_x} + \left(1 - \frac{\varepsilon_z}{\varepsilon_x}\right)\sin^2\alpha}$$

where n is the usual refractive index of the extraordinary beam which is a function of the incident angle α. Refractive properties of a slab of a uniaxial material would be of interest for direct applications when β is practically independent of α. This condition is fulfilled when $|\varepsilon_z| \gg \varepsilon_x$,1; in this case $\sin\alpha \gg \sin\beta$. However, Veselago's lens cannot be realized since $\sin\alpha/\sin\beta = –1$ cannot be achieved for an arbitrary α. The refraction angles as a function of the angle of incidence are plotted in Fig. 2.

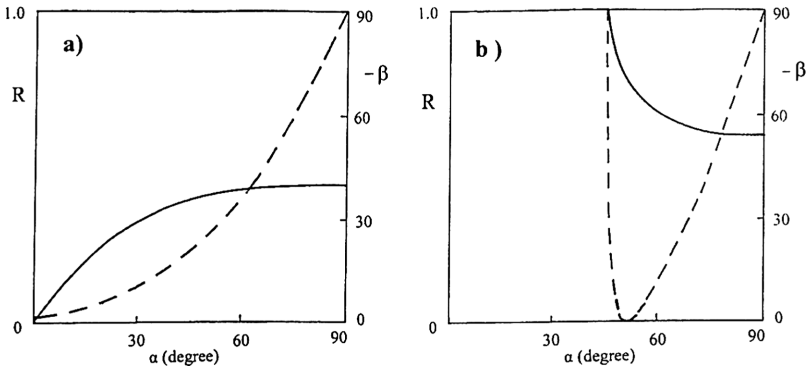

Figure 2. The refraction angle β (full curve; $\gamma = -\beta$ for the chosen values of ε_x and ε_z) and the reflectivity R (dashed curve) as a function of the incident angle α of a material with a) $\varepsilon_x = 0.5$, $\varepsilon_z = -0.5$ and b) $\varepsilon_x = -0.5$, $\varepsilon_z = 0.5$, respectively. In the case a) R is nonzero for any α since $\varepsilon_x < 1$.

Which dielectric materials may display negative refraction? Negative permittivity $\varepsilon(\omega)$ naturally occurs, for example, near optical phonon resonant lines in polar dielectrics which, moreover, should be anisotropic. Mercurous halides Hg_2X_2 (X = Cl, Br, I) are good candidates for observing the negative refraction since these materials exhibit very large dielectric anisotropy in the infrared region: two strong resonance lines appear which are shifted in frequency with respect to each other and correspond to differently polarized polaritons (see Fig. 3). Note that the two cases discussed in this paper (i.e. $\varepsilon_x < 0$, $\varepsilon_z > 0$ and $\varepsilon_x > 0$,

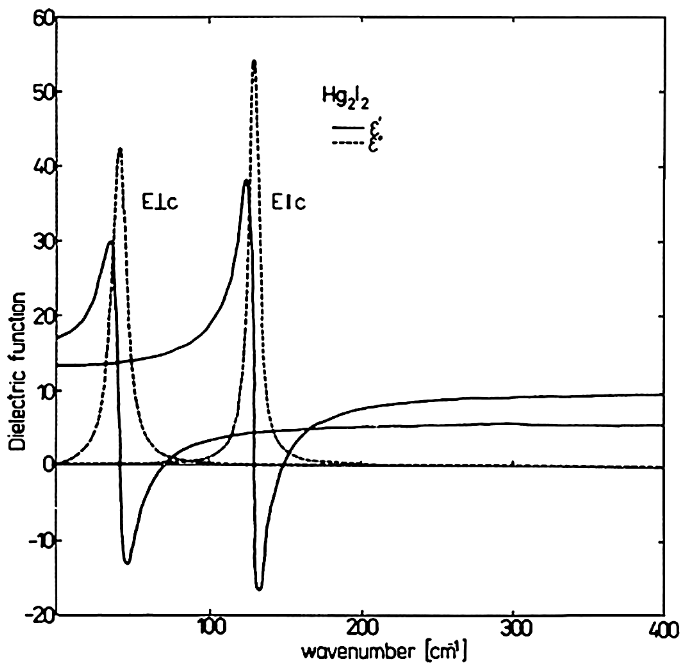

Figure 3. The dielectric function calculated from the single classical-oscillator fit of the reflectivity spectrum of Hg_2I_2 by Petzelt et al. [11].

$\varepsilon_z < 0$) are experimentally achievable using a single slab in two distinct spectral ranges in the far infrared.

Reflection

The reflectivity R is given by the formula

$$R = \frac{|K_z - \varepsilon_x k_z|^2}{|K_z + \varepsilon_x k_z|^2}$$

which can be rewritten for a medium with negligibly small imaginary parts of ε_i (K_z real) in the form

$$R = \left(\frac{n\cos\gamma - \varepsilon_x\cos\alpha}{n\cos\gamma + \varepsilon_x\cos\alpha}\right)^2$$

where $n(\alpha)$ is the refraction index. The course of R for a TM polarized EM wave as a function of α when $\varepsilon_x > 0$, $\varepsilon_z < 0$ is qualitatively similar to that of an isotropic material: first it decreases with increasing α, it reaches zero at the Brewster angle provided $\varepsilon_x > 1$ and then it increases to unity at $\alpha = \pi/2$.

The reflection from a dielectric exhibiting a negative index of refraction n, i.e. with $\varepsilon_x < 0$, $\varepsilon_z > 0$ is more interesting. If $\varepsilon_z \geq 1$ (Re $K_z = 0$), the surface is totally reflecting (R = 1) for any angle α of incidence. For the special case $\varepsilon_x = -1$, $\varepsilon_z = 1$ we have $K_z = \mathrm{i}k_z$ and the phase of the reflected wave is shifted by $\pi/2$. Therefore the surface acts as a mirror and the reflected wave will enhance a wave radiated from a source (an electric dipole antenna) placed $(4\mathrm{N} + 1)\lambda/8$ away from the surface [6]. The reflectivity R as a function of the incident angle α is plotted in the Fig. 2.

We remind that the reflectance R_d of a film of thickness d is given by the formula [10]

$$R_d = \frac{4R\sin^2\phi}{(1-R)^2 + 4R\sin^2\phi}$$

where $\Phi = K_z\mathrm{d}$ and R is the reflectivity of a semi-infinite medium.

It has been shown that the sign of K_z is determined by the sign of ε_x. Therefore combining two layers (I and II) of appropriate thickness having opposite signs of ε_x, the phase advance of the transmitted wave across this bilayer can be made zero. If, moreover, the two layers are impedance matched ($\varepsilon_x^I K_z^{II} = \varepsilon_x^{II} K_z^I$), the bilayer exhibits a unit transfer function T. Such a compensated bilayer has been proposed by Smith and Schurig [6] composed of appropriate magnetic anisotropic materials. The important point is that T = 1 can be achieved for all incident angles and the bilayer could be used for imaging in a similar manner as the perfect lens [6]. Unfortunately nonmagnetic ($\mu = 1$) anisotropic bilayer has this property only for a particular value of the incident angle.

Surface and Guided Modes in an Anisotropic Slab

Surface modes (SM's) in a semi-infinite medium and in a slab of an isotropic magnetic material have been studied in context with the negative index of refraction [12, 13, 14]. SM's in nonmagnetic anisotropic semi-infinite dielectrics were discussed long time ago [8]. We complete those studies considering SM's and guided modes (GM's) in a uniaxial anisotropic nonmagnetic dielectric slab in vacuum.

Using Maxwell equations and continuity of fields at the slab surfaces we get two types of GM's of the form $H_y \exp[i(k_x x - \omega t)]$:

1) Symmetric modes (with respect to z)

$$\begin{aligned} H_y(z) &= H_y(0)\cos(\beta z), && -d/2 \le z \le d/2 \\ H_y(z) &= H_y(d/2)\exp[-\beta_1(|z| - d/2)] && \text{outside the slab,} \end{aligned} \tag{4a}$$

with dispersion relation

$$\frac{\varepsilon_x \beta_1}{\beta}\cot\left(\beta\frac{d}{2}\right) = 1.$$

2) Anti-symmetric modes

$$\begin{aligned} H_y(z) &= H\sin(\beta z), && -d/2 \le z \le d/2 \\ H_y(z) &= H_y(d/2)\exp[-\beta_1(z - d/2)], && z \ge d/2 \\ H_y(z) &= -H_y(d/2)\exp[\beta_1(z + d/2)]\,, && z \le -d/2 \end{aligned} \tag{4b}$$

with dispersion relation

$$\frac{\varepsilon_x \beta_1}{\beta}\tan\left(\beta\frac{d}{2}\right) = -1.$$

Here $\beta_1 = (k_x^2 - k_0^2)^{1/2}$ and $\beta = (\varepsilon_x k_0^2 - [\varepsilon_x/\varepsilon_z]k_x^2)^{1/2}$. The condition $\varepsilon_x k_0^2 - [\varepsilon_x/\varepsilon_z]k_x^2 > 0$ limits the range of allowed k_x. The condition $k_x^2 - k_0^2 > 0$ simply means that the GM's are nonradiative. When β becomes purely imaginary we get SM's localized near the slab surfaces. Their dispersion relations and $H_y(z)$ are obtained by making the substitution $\beta \to -i([\varepsilon_x/\varepsilon_z]k_x^2 - \varepsilon_x k_0^2)^{1/2}$ in the above formulae. We remind that SM's can exist only if $\varepsilon_x < 0$.

We consider the frequency dependence of both ε_x and ε_z in the form of a resonant line

$$\varepsilon_i(\omega) = \frac{\varepsilon_{\infty i}\left(\omega_{Li}^2 - \omega^2\right)}{\omega_{Ti}^2 - \omega^2}$$

which is characteristic for optical phonons in crystals. The course of dispersion curves depends on the relative value of the frequencies ω_{Ti} and ω_{Li} of transverse and longitudinal phonons, respectively. In Fig. 4 the dispersion curves of SM's and GM's are plotted using $\varepsilon_x(\omega)$ and $\varepsilon_z(\omega)$ corresponding to Hg_2I_2 (see Fig. 3). We use the reduced coordinates: $\Omega = \omega/\omega_{Lz}$, $\kappa = ck_x/\omega_{Lz}$.

Note that the anti-symmetric SM continuously transforms (with κ^2) into the anti-symmetric GM. In the frequency region where $\varepsilon_x < 0$, there exist simultaneously both SM's and GM's which cannot be observed in materials with the same sign of the permittivity components. At the point on the lower bulk polariton branch where $\varepsilon_x \to \pm\infty$ all GM's are degenerate. Starting from certain values of k_x the GM's in the region of $\varepsilon_z < 0$ ($\varepsilon_x > 0$) have negative group velocity $v_{gx} = d\omega/dk_x$, opposite to the phase velocity. The same conclusion can be drawn, as it should be, from the formula for the energy flow which reads (for symmetric modes, for example)

$$S_x = \frac{ck_x}{16\pi k_0}\left\{\frac{2\cos^2\left(\beta\frac{d}{2}\right)}{\beta_1} + \frac{d}{\varepsilon_z}\left(1 + \frac{\sin\beta d}{\beta d}\right)\right\} H_y(0)^2\,.$$

The first term describes the energy flow outside the slab (always positive) and the second one corresponds to the energy flow inside the slab (negative for $\varepsilon_z < 0$). The ratio of these two terms determines the direction of the total energy flow. Note that the negative v_g occurs just in the region $\varepsilon_x > 0$, $\varepsilon_z < 0$ of the negative refraction of **S** (see Fig. 1a).

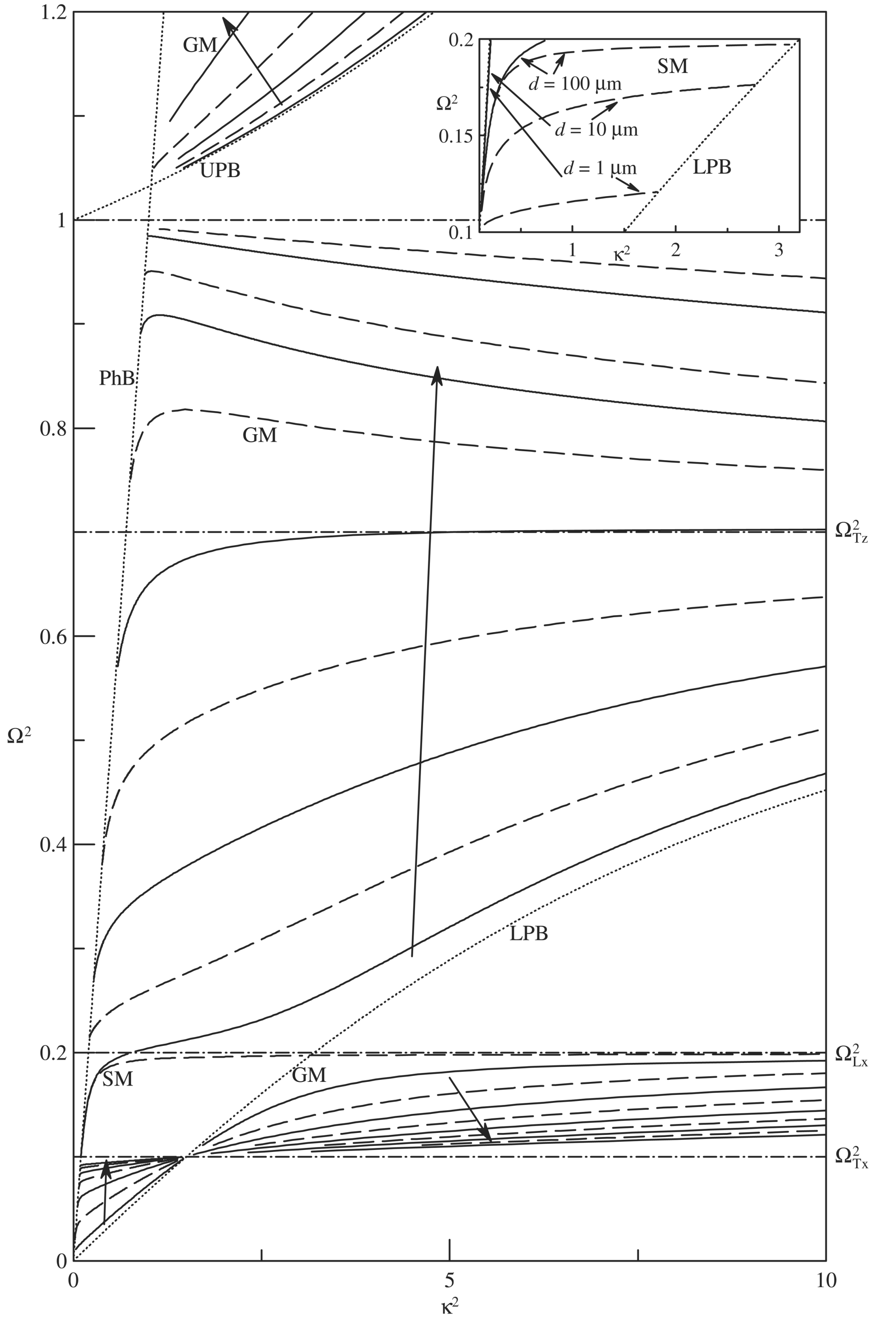

Figure 4. The dispersion curves of SM's and GM's in the slab of thickness $d = 100\,\mu$m. $\Omega^2 \equiv \omega^2/\omega^2_{Lz}$ ($\omega_{Lz} \approx 3\times 10^{13}$ s^{-1}, $\Omega^2_{Tx} \equiv \omega^2_{Tx}/\omega^2_{Lz} = 0.1$, $\Omega^2_{Lx} = 0.2$, $\Omega^2_{Tz} = 0.7$); $\kappa^2 \equiv (c^2/\omega^2_{Lz})k^2_x$. In the frequency ranges (0.1–0.2) and (0.7–1.0), delimited by dashed-dotted horizontal lines in the plot, $\varepsilon_x < 0$ and $\varepsilon_z < 0$, respectively. The straight dotted line is the photon branch in vacuum (PhB), the dotted curves represent upper and lower bulk polariton branches (UPB and LPB) polarized in the z-direction. The full and dashed lines correspond to symmetric and anti-symmetric modes, respectively. SM's are confined to the region above the LPB where $\varepsilon_x < 0$. The series of the dispersion curves of GM's correspond to different values of β, i.e., to different variation of $H_y(z)$ versus z (4a,b); arrows indicate the growing β (there is an infinite number of dispersion curves in each region). Inset: SM's for several thicknesses d.

To observe the SM's and GM's directly, the attenuated total reflection method or Raman scattering could be used.

The Effect of Spatial Dispersion

It is well-known that a mode with negative group velocity may exist when spatial dispersion of $\varepsilon(\omega, \mathbf{k})$ is taken into account [8, 15]. This is the case of so called additional or new waves when the spatial dispersion of ε is of the form [8] (for a non-gyrotropic medium)

$$\varepsilon(\omega, k) = \varepsilon_\infty + \frac{A}{\omega_T^2 + \beta k^2 + \gamma k^4 - \omega^2} \tag{5}$$

with $\beta < 0$, $\gamma > 0$. The group velocity may be negative in this case as it follows directly from the expression for the energy flow which contains an additional term proportional to $-[\partial\varepsilon(\omega, \mathrm{k})/\partial \mathrm{k}]_{k=0} \sim \beta < 0$ [8].

The spatial dispersion of the form (5) with negative β is typical for ferroelectric materials which exhibit an incommensurate phase like in thiourea and sodium nitrite [16]. In these materials there exist soft transverse optic modes with negative slopes of their dispersion curves at $\mathrm{k} \simeq 0$ (Fig. 5).

A question arises whether these additional waves (one of them having the negative group velocity) can be observed in reflection or transmission spectra of EM waves in a slab of incommensurate materials. In principle, measuring the intensity of monochromatic EM wave passing through a slab as a function of its thickness, one could observe the oscillations of the intensity due to interference of the three different waves. In reflection and/or transmission spectra one can expect additional features due to the existence of additional waves. Such effects could be observed obviously only if the damping of the modes is small.

Detailed theoretical analysis of reflection and transmission spectra could be made generalizing the procedure used in [17] for an anisotropic slab.

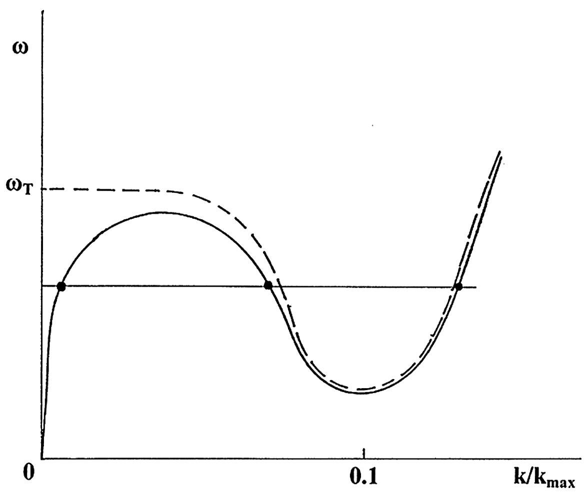

Figure 5. Schematic illustration of the dispersion curve of the soft optic phonon (dashed line) and polariton (solid line) in incommensurate ferroelectrics. At a given frequency three modes with different wavelengths denoted by full circles can propagate.

Conclusion

This paper is a complementary study to a more general case of a material showing both magnetic and dielectric anisotropy [5, 6, 7]. We have discussed under which circumstances negative refraction may occur in a material exhibiting dielectric anisotropy only. We propose that mercurous halides are good candidates of anisotropic materials in which negative refraction of either the wave-vector or the Poynting vector may be observed. The advantage of such materials is that they are natural, in contrast to sophisticated artificial materials [4], and that the frequency range of interest is shifted from the microwave to the infrared region. On the other hand, some important properties (Veselago lens, for example) are lost.

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