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Local piezoelectric effect in disordered dielectrics

L I Deich
Kirensky Institute of Physics, Krasnoyarsk 660036, USSR

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Abstract. We consider for the first time the influence of the local violation of symmetry with respect to spatial inversion on the properties of disordered systems. A specific example of this influence is the local piezoelectric effect in disordered dielectrics. The simplest static and dynamic properties of these systems are investigated.

1. Introduction

The local violation of spatial homogeneity is one of the definitive properties of the various kinds of disordered systems (amorphous materials, glasses etc). The majority of papers in this field deal with the investigation of the influence of this property on different physical phenomena. Violation of spatial homogeneity results in symmetry violation with respect to inversion. However, I do not know of any papers in which this fact was considered. At the same time it is clear that this element of symmetry is very important, as its presence leads to the preclusion of a whole number of physical phenomena. Thus the local violation of this preclusion may influence the properties of various disordered materials.

In the present paper this idea is demonstrated on the example of the local piezoelectric effect in disordered dielectrics. The piezoelectric properties of a continuous medium are known to be characterised by a tensor of the third rank ν_{ijk}, which only differs from zero in the absence of central symmetry [1]. By a local piezoelectric effect we mean the linear relation of a local electric field to a local tensor of deformations. This relation is described by a random tensor field $\nu_{ijk}(\boldsymbol{r})$. Its average value over the ensemble (which is equivalent to the space average when spatial ergodicity is present) will be considered to be zero. In specific calculations we shall use the simplest form of this tensor with a minimal number of parameters:

$$\nu_{ijk} = \nu_1 l_j \delta_{jk} + \nu_2 (l_j \delta_{ik} + l_k \delta_{ij}). \tag{1}$$

Here ν_1 and ν_2 are piezomodules that will be considered to be constant parameters, and $\boldsymbol{l}$ is a unit random vector with the following statistical properties

$$\langle l_i(\boldsymbol{r})\rangle = 0 \qquad \langle l_i(\boldsymbol{r}) l_j(\boldsymbol{r}')\rangle = \tfrac{1}{3}\delta_{ij} K_l(\boldsymbol{r}-\boldsymbol{r}'). \tag{2}$$

Here $K_l(\boldsymbol{r})$ is a normalised correlation function characterised by an arbitrary correlation radius r_c. Expressions (1) and (2) imply the presence of local anisotropy in the system that is in agreement with modern representations concerning the structure of disordered systems and a popular model of local uniaxial anisotropy (see e.g. [2–4]). However, we

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shall neglect this anisotropy in the elastic and dielectric characteristics of the system. Its influence on the results will be considered in the process of investigation.

Thus, the free energy of the system F will be written in the form

$$F = \tfrac{1}{2}\lambda u_{ii}^2 + \mu u_{ij} u_{ji} - (1/8\pi)(\varepsilon E^2 + H^2) - \nu_1 (\boldsymbol{l}\cdot\boldsymbol{E}) u_{ii} - \nu_2 E_i l_j (u_{ij} + u_{ji}) \tag{3}$$

where $u_{ij} = \partial u_i/\partial x_j$ is distorsion tensor, λ and μ are elastic modules, ε is a local dielectric constant of the medium, and $\boldsymbol{E}$ is an electric field.

From (3) there follows an important equation relating a tensor of elastic stresses σ_{ij} and a vector of electric induction $\boldsymbol{D}$ to a distortion tensor u_{ij} and the electric field $\boldsymbol{E}$, respectively

$$\sigma_{ij} = \partial F/\partial u_{ij} = \lambda\delta_{ij} u_{ll} + \mu(u_{ij} + u_{ji}) - \nu_1 \delta_{ij} (\boldsymbol{l}\cdot\boldsymbol{E}) - \nu_2 (E_i l_j + E_j l_i) \tag{4a}$$

$$D_i = -4\pi\, \partial F/\partial E_i = \varepsilon E_i + 4\pi[\nu_1 l_i u_{jj} + \nu_2 l_j (u_{ij} + u_{ji})]. \tag{4b}$$

2. Static properties of materials with a local piezoelectric effect

First of all, let us study several of the simplest static properties of systems with a local piezoelectric effect. In principle, the essence of these properties is the same as in the case of the normal piezoelectric effect—the appearance of deformations under the application of an electric field—and *vice versa*. The difference is the following. In the case under consideration it is necessary to speak of the local deformation and the local electric field, which are random fields and, therefore, require a statistical approach for their description. It is quite evident that it is not sufficient to investigate only average values, and so we shall also consider two-point autocorrelation functions of these fields.

To determine average values, let us consider the problem of a mean field inside an infinite ellipsoid placed in the external electric field $\boldsymbol{E}_{\mathrm{ex}}$. The infinity is required to simplify the elastostatic part of the problem. In this case one can use a well-known expression for the Green tensor of an infinite isotropic medium [5]. In an electrostatic sense, an ellipsoid is considered to be finite, with the usual boundary conditions.

Finding the deformation tensor u_{ij} from the elastostatic equations and substituting the result into (4b), we obtain a closed system of electrostatic equations. Its peculiarity is in the non-local character of the relation between the vectors of the electric induction $\boldsymbol{D}$ and the field $\boldsymbol{E}$. The main problem here is finding an effective dielectric constant that would enable us to replace a precise equation by a generally relevant local one relating the average values $\langle \boldsymbol{D}\rangle$ and $\langle \boldsymbol{E}\rangle$. In the process of averaging we need to decouple correlators of the form $\langle E_i l_j l_k\rangle$ multiplied by a coefficient that is quadratic over the peizomodules. Let us represent the field $\boldsymbol{E}$ by the form $\boldsymbol{E} = \langle \boldsymbol{E}\rangle + \tilde{\boldsymbol{E}}$, where $\tilde{\boldsymbol{E}}$ is a fluctuating part of this field: $\langle \tilde{\boldsymbol{E}}\rangle = 0$. Substituting the expression into the correlator we obtain $\langle E_i l_j l_k\rangle = \langle E_i\rangle\langle l_j l_k\rangle + \langle \tilde{E}_i l_j l_k\rangle$. Since the value $\tilde{E}_i$ is of the first order over the piezomodules, the second summand would appear to be of the third order. Thus, it will be neglected from now on. We have therefore arrived at the desired effective dielectric constant:

$$\varepsilon_{\mathrm{eff}} = \varepsilon_0 + (4\pi/9)[4\nu_2^2/\mu + (3\nu_1^2 + 4\nu_1\nu_2 + 4\nu_2^2)/(\lambda + 2\mu)]. \tag{5}$$

To solve the problem further one should use well-known formulae that can be found in any textbook (see, e.g. [1]) taking into account expression (5).

One should here note that the fluctuating anisotropy in the local dielectric constant (which was neglected) also contributes to $\varepsilon_{\mathrm{eff}}$. Proceeding from the results of such simple calculations it is not possible to separate these contributions. However, one should bear in mind the existence of these two mechanisms forming ε_{eff}: usually only one, namely the presence of a fluctuating part in $\varepsilon(\boldsymbol{r})$, is taken into account.

We have mentioned above that to give a more detailed description of the system under consideration it is necessary to investigate the correlation properties of random fields appearing in the system. We now investigate two-point autocorrelation functions both of the electric field in the sample when subjected to the external stress and of the deformation field in the sample when placed in the external electric field. And we restrict ourselves to the first approximation over the piezomodules. It is easy to show that the end effects have no influence on these correlation functions if the sample sizes are much greater than the correlation radius that appears in (2). It is then convenient to make a Fourier transformation of the corresponding equations and to calculate correlators of the following form: $\langle \tilde{v}_{ij}(\boldsymbol{k})\tilde{v}^*_{kl}(\boldsymbol{k}')\rangle$ and $\langle \tilde{E}_i(\boldsymbol{k})\tilde{E}^*_j(\boldsymbol{k}')\rangle$, where $\tilde{E}_i$ and $\tilde{v}_{ij}$ are Fourier transforms of the corresponding centred values; $\tilde{v}_{ij} = v_{ij} - \langle v_{ij}\rangle$, $v_{ij} = (u_{ij} + u_{ji})/2$. Performing simple calculations we obtain that

$$\langle \tilde{E}_i(\boldsymbol{k})\tilde{E}^*_j(\boldsymbol{k}')\rangle = \varphi_{ij}(k)\delta(\boldsymbol{k}-\boldsymbol{k}') \tag{6}$$

$$\langle \tilde{v}_{ij}(\boldsymbol{k})\tilde{v}^*_{kl}(\boldsymbol{k}')\rangle = T_{ijkl}(k)\delta(\boldsymbol{k}-\boldsymbol{k}') \tag{7}$$

where

$$\varphi_{ij}(k) = (4\pi/\varepsilon k^2)^2 k_i k_j \,|\,\nu_1 U^{(0)}_{nn} k_m + 2\nu_2 k_j U^{(0)}_{jm}\,|^2\, S_l(k) \tag{8}$$

and the expression for the tensor T_{ijkl} is too bulky to give in full. Therefore we shall restrict ourselves to the convolution $T_{iikk} \equiv T_V(k)$, which in the linear approximation over v_{ij} describes local fluctuations of the volume $\Delta V(\boldsymbol{r})$:

$$T_V(k) = [\nu_1/(\lambda+2\mu)]^2[E_0^2 + 3(\nu_2/\nu_1)^2(\boldsymbol{k}\cdot\boldsymbol{E}_0)^2/k^2]S_l(k). \tag{9}$$

$U^{(0)}_{ij}$ and E_0 in (9) represent the deformation tensor and the electric field of the medium, respectively, calculated in the zero-order piezoelectric effect approximation. $S_l(k)$ is a Fourier transform of the correlation function (2). Expressions (6) and (7) are obtained if one assumes that $U^{(0)}_{ij}$ and E_0 are homogeneous inside the sample. They mean that under these conditions the fields $v_{ij}(\boldsymbol{r})$ and $\boldsymbol{E}(\boldsymbol{r})$ are homogeneous random fields. Their correlation properties are determined by the Fourier-transformation of values (8) and (9):

$$K^E_{ij}(r) = \left(\frac{4\pi}{\varepsilon}\right)^2 \int \frac{k_i k_j}{k^2}\left|\frac{\nu_1 U^{(0)}_{nn}k_m + 2\nu_2 k_j U^{(0)}_{jm}}{k}\right|^2 S_l(k)\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\,\mathrm{d}^3k \tag{10}$$

$$K_V(r) = \left(\frac{\nu_1 E_0}{\lambda+2\mu}\right)^2 K_l(r) + 3\left(\frac{\nu_2}{\lambda+2\mu}\right)^2 \int \frac{(\boldsymbol{k}\cdot\boldsymbol{E}_0)^2}{k^2} S_l(k)\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\,\mathrm{d}^3k. \tag{11}$$

($K_l(r)$ is determined from (2).)

Further analysis requires a specific definition of the correlation function (2) and of the corresponding function $S_l(k)$. We shall consider two types of the correlation function:

$$K^{\mathrm{I}}_l(r) = \exp(-r^2/2r_c^2) \qquad S^{\mathrm{I}}_l(r) = [r_c^3/(2\pi)^{3/2}]\exp(-k^2r_c^2/2) \tag{12}$$

$$K^{\mathrm{II}}_l(r) = (1 - r^2/3r_c^2)\exp(-r^2/2r_c^2) \qquad S^{\mathrm{II}}_l(r) = [k^2r_c^5/3(2\pi)^{3/2}]\exp(-k^2r_c^2/2). \tag{13}$$

The main difference between them is in the fact that expression (13) describes a random

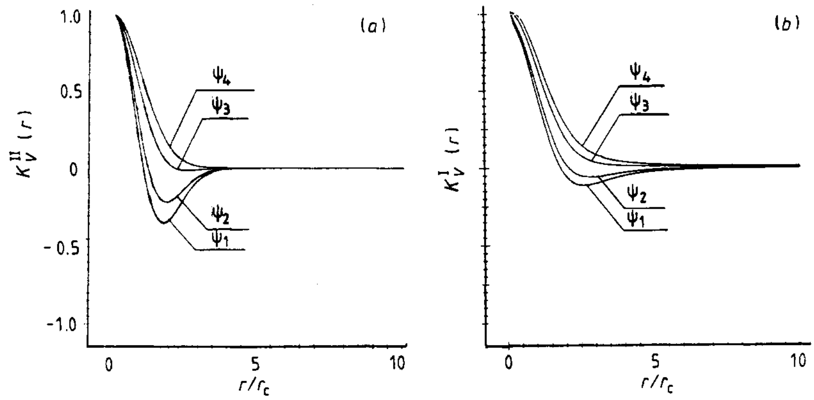

Figure 1. Plots of the dependence of the correlation functions of local volume fluctuations on the distance along different directions with respect to the field. The corresponding angles are: $\psi_1 = 0°$, $\psi_2 = 30°$, $\psi_3 = 60°$, $\psi_4 = 90°$.

field with anticorrelation effects, and $\int K_l^{II}(r)\,d^3r = S_l^{II}(0) = 0$. Functions of this type were first used to model disordered systems in [6]. It was shown there that the properties of the system may in this case change considerably.

For the correlation function of the volume fluctuations (11) the calculations may be worked through to the end. So we obtain

$$K_V^{I}(r) = [(\nu_1^2 + 3\nu_2^2\cos^2\psi)/(\lambda+2\mu)^2]E_0^2\exp(-r^2/2r_c^2) + 3[\nu_2 E_0/(\lambda+2\mu)]^2 \times (2\cos^2\psi - \sin^2\psi)(r_c^2/r^2)[\exp(-r^2/2r_c^2) - (r_c/r)\sqrt{(\pi/2)}\,\mathrm{erf}(r/\sqrt{2}r_c)]\dagger \tag{14}$$

using $S^{I}(k)$ and

$$K_V^{II}(r) = \{[\nu_1 E_0/(\lambda + 2\mu)]^2 + 6[\nu_2 E_0/(\lambda + 2\mu)]^2[1 - (r^2/r_c^2)\cos^2\psi]\}\exp(-r^2/2r_c^2) \tag{15}$$

using $S^{II}(k)$, where ψ is the angle between the direction of observation and the field E_0.

The main difference between (14) and (15) is in their behaviour at infinity (see figure 1). The behaviour of the correlation function (15) is similar to that of the initial function (13). At the same time, in expression (14) instead of the Gaussian law (13) there appears a power tail of the form

$$K_V^{I}(r) \sim [E_0\nu_2/(\lambda + 2\mu)]^2(\sin^2\psi - 2\cos^2\psi)r_c^3/r^3 \tag{16}$$

the sign of which changes at $\psi \simeq 55°$.

When investigating the correlation properties of a random electric field $\boldsymbol{E}(\boldsymbol{r})$ described by the tensor (10) we confined ourselves to a numerical integration of expression (10) using spectral density $S^{I}(k)$ (12). The tensor of the external deformations $U_{ij}^{(0)}$ is chosen in the form $U_{ij}^{(0)} = U^{(0)}n_i n_j$, which describes a uniaxial deformation in the direction of a unit vector, $\boldsymbol{n}$. In the system of coordinates with an axis $0z$ parallel to $\boldsymbol{n}$, the

† $\mathrm{erf}(x) = \frac{2}{\sqrt{\pi}}\int_0^x e^{-t^2/2}\,dt$ is the well-known error function.

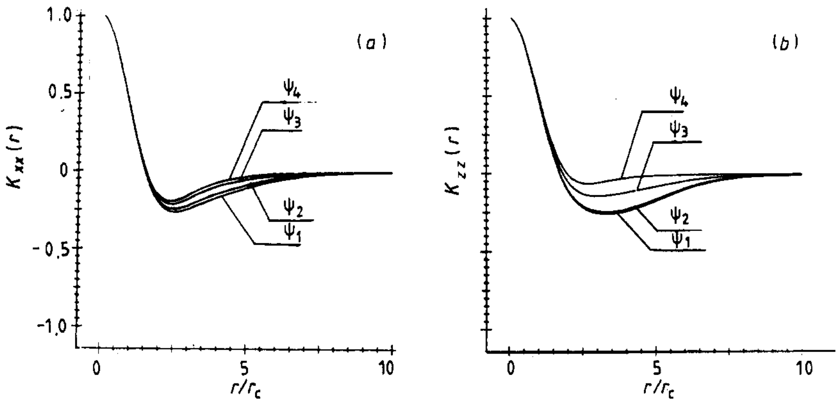

Figure 2. Plots of the dependence of the correlators (*a*) $\langle E_x(\boldsymbol{r})E_x(0)\rangle$ and (*b*) $\langle E_z(\boldsymbol{r})E_z(0)\rangle$ on the distance along different directions with respect to deformation axis. Angle values are the same as in figure 1.

correlation tensor $K_{ij}(\boldsymbol{r})$ is diagonal, and $K_{xx} = K_{yy}$. Figure 2 shows plots of components of this tensor: K_{xx}(figure 2(*a*)) and K_{zz} (figure 2(*b*)). They describe the behaviour of the correlators $\langle \tilde{E}_x(\boldsymbol{r})\tilde{E}_x(0)\rangle$ and $\langle \tilde{E}_z(\boldsymbol{r})\tilde{E}_z(0)\rangle$ respectively. It is seen that there is also a 'tail' here, which is analogous to the tail of the correlation function $K_V^{\text{I}}(\boldsymbol{r})$, however, it is 'shorter' than that in figure 1: the law $1/r^3$ (see (16)) is replaced by the law $1/r^5$. Let us also pay attention to the fact that anticorrelation effects are characteristic of fluctuations of the electric field at any angles ψ between the direction of observation and the deformation axis $\boldsymbol{n}$.†

In conclusion, we may write down the following expressions for the dispersions γ_{ij}^E and γ^V of the electric field and the local volume fluctuations:

$$\begin{aligned} \gamma_{ij}^E &= \langle \tilde{E}_i(0)\tilde{E}_j(0)\rangle^{1/2} = (4\pi U_0/\sqrt{3}\varepsilon)[\nu_1^2 + \tfrac{4}{15}\nu_2(\nu_1+\nu_2)]^{1/2}\delta_{ij} \\ \gamma^V &= \langle \tilde{u}_{ii}^2(0)\rangle^{1/2} = [(\nu_1^2+\nu_2^2)^{1/2}/(\lambda+2\mu)]E_0. \end{aligned} \tag{17}$$

We would now like to emphasise the dependence of γ_{ij}^E and γ^V on the value of the corresponding external action. This enables us, in principle, to influence these parameters and to create models of disordered systems with a regulated degree of disorder.

3. Elastic and electromagnetic waves

In this section we investigate the influence of the local piezoelectric effect on the dispersion laws for elastic and electromagnetic waves. To obtain them we have two different but equivalent methods. One can solve the initial stochastic equation using perturbation theory. The series obtained is then averaged and summed using one approximation or another (see, e.g. [7]). The second method is the following. First we average the equation itself and then we use perturbation theory to obtain an average of the value under consideration. There are several equivalent formulations of this method, e.g. [8, 9]. We applied the approach developed in [9]. And we restricted ourselves to

† Using the correlation function $K_l^{\text{II}}(r)$, replace the law r^{-5} by the law r^{-3} and the others features of these correlators are not changed.

the first non-disappearing correction for the dispersion law that is equivalent to the well known Bourret approximation [10].

Thus, solving Maxwell's equations and those of elasticity theory for the dispersion law of a transverse elastic wave, we obtain the following expression:

$$\omega = s_\perp k\left[1 + \frac{2\pi}{3\varepsilon}\frac{\nu_2^2}{\mu}\left(2 + \int \mathrm{d}^3 q \frac{q^2 + q_x^2}{k^2\beta_\perp^{-2} - q^2} S_l(\boldsymbol{k} - \boldsymbol{q})\right)\right] \tag{18}$$

where $s_\perp$ is the unperturbed velocity of the transverse wave and $\beta_\perp = c/\sqrt{\varepsilon} s_\perp \simeq 10^3$–$10^5$. The analysis of similar expressions was performed for the first time in [11]. The results are as follows. In the region of wavenumbers $k \ll \beta_\perp k_c$ (k_c is a correlation wavenumber of inhomogeneities $l(\boldsymbol{r})$, $k_c \simeq 1/r_c$) the dispersion law is linear with the renormalised velocity

$$\tilde{s}_\perp = s_\perp[1 + (4\pi/9\varepsilon)(\nu_2^2/\mu)]. \tag{19}$$

Deviations from (19) begin to appear at $k \simeq \beta_\perp k_c$. However, if one considers the order of value $\beta_\perp$, it may be seen that this region lies far beyond the limits of the wavenumbers possible for a continuous theory at reasonable values, for disordered materials $r_c \simeq 1$–100 Å. Thus, local piezoelectric effect does not modify the dispersion law of elastic waves. The modification will appear if we need to take into account the random anisotropy in the tensor of the elastic modules. Such a problem without an account of the piezoelectric effect has been solved in [12]. It is clear that a purely piezoelectric contribution will not change the nature of this modification. However, there is also a cross term which takes into account the cross-correlation of the anisotropy of the elastic modules and a piezotensor. We shall not here consider the contribution of this term that is valid under the condition $\nu_1^2/b \ll 1$, where b is an elastic module characterising the anisotropy value.

For a transverse electromagnetic wave, the modified dispersion law has the form

$$\omega = \frac{ck}{\sqrt{\varepsilon}}\left[1 + \frac{2\pi}{3}\left(\frac{(\nu_1 + 2\nu_2)^2}{\lambda + 2\mu}\int \mathrm{d}^3 q \frac{q_x^2 S_l(\boldsymbol{k} - \boldsymbol{q})}{\beta_\parallel^2 k^2 - q^2} + \frac{\nu_1^2}{\lambda + 2\mu}\right.\right.$$
$$\left.\left.\times \int \mathrm{d}^3 q \frac{q_x^2 + q_z^2}{\beta_\parallel^2 k^2 - q^2} S_l(\boldsymbol{k} - \boldsymbol{q}) + \frac{2\nu_2^2}{\mu}\int \mathrm{d}^3 q \frac{q_x^2 + q_z^2}{\beta_\perp^2 k^2 - q^2} S_l(\boldsymbol{k} - \boldsymbol{q})\right)\right] \tag{20}$$

where $\beta_\parallel = c/\sqrt{\varepsilon} s_\parallel$. Here the situation is more complicated, as there are terms corresponding to the scattering of the electromagnetic wave both by longitudinal elastic waves (terms containing $\beta_\parallel$) and by transverse ones (terms containing $\beta_\perp$). However, this complication is not very important, as usually $\beta_\perp \simeq \beta_\parallel$ (or more exactly, $\beta_\perp \geqslant \sqrt{2}\beta_\parallel$) and therefore deviations from the linear law due to both groups of terms will be approximately in one region, $k \simeq k_c/\beta$. Taking into account the value β, it is clear that the dispersion law modification of the electromagnetic wave shifts considerably towards the region of the wavenumbers less than the correlation wavenumber k_c, by some three to five orders of magnitude. In other words, inhomogeneities of the size of r_c appear in the dispersion law in the region of wavelengths $\lambda \simeq 10^3$–$10^5 r_c$. A similar effect was first discovered in [11] when studying electromagnetic waves in disordered metals. It was called the 'microscope' effect.

To get a notion of the dispersion curve behaviour, let us find asymptotes of the dispersion law (20) in two limit cases $k \ll k_c/\beta$ and $k \gg k_c/\beta$.

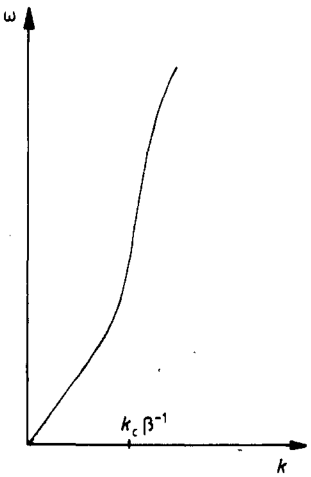

Figure 3. Qualitative form of the dispersion law of the electromagnetic waves in the region of wavenumbers $k \sim k_c/\beta$. In the region $k \sim k_c$, one more modification related to fluctuations of dielectric constant may be observed.

At $k \ll k_c/\beta$

$$\omega \approx (ck/\sqrt{\varepsilon})\{1 - \tfrac{2}{9}\pi[(3\nu_1^2 + 4\nu_1\nu_2 + \nu_2^2)/(\lambda + 2\mu) + 4\nu_2^2/\mu]\}. \tag{21}$$

At $k \gg k_c/\beta$

$$\omega \approx (ck/\sqrt{\varepsilon})[\![1 + \tfrac{2}{3}\pi\{[\nu_1^2/(\lambda + 2\mu)]\beta_\parallel^{-2} + (2\nu_2^2/\mu)\beta_\perp^{-2}\}]\!]. \tag{22}$$

As $\beta \gg 1$, the last terms in (22) become too small to be taken into account. An approximate form of the curve is given in figure 3.

A consideration of the fluctuations of the dielectric constant (e.g. local anisotropy) leads to an equal additional negative contribution to both (21) and (22), since the dispersion law modification related to these fluctuations is in the region $k \simeq k_c$. The contribution of these terms, taking into account the cross-correlation of these fluctuations with the piezotensor ν_{ijk}, may always be neglected.

The local piezoelectric effect leads to the appearance of a longitudinal component of the electromagnetic field. The equation for its average amplitude in our approximation has the form

$$\langle E_z\rangle\left[1 - \frac{4\pi}{3\varepsilon}\left(\frac{(\nu_1 + 2\nu_2)^2}{\lambda + 2\mu}\int d^3q\,\frac{q_z^2 S(\boldsymbol{k} - \boldsymbol{q})}{\omega^2 s_\parallel^{-2} - q^2} + \frac{2\nu_1^2}{\lambda + 2\mu}\right.\right.$$
$$\left.\left.\times\int d^3q\,\frac{q_x^2 S(\boldsymbol{k} - \boldsymbol{q})}{\omega^2 s_\parallel^{-2} - q^2} + \frac{2\nu_2^2}{\mu}\int d^3q\,\frac{q_x^2 + q_z^2}{\omega^2 s_\perp^{-2} - q^2}\,S(\boldsymbol{k} - \boldsymbol{q})\right)\right] = 0. \tag{23}$$

At $\omega < \omega_c$, where ω_c is a critical frequency, the expression in the square brackets has a negative sign. Consequently, the dispersion equation following from (23) has no solutions at these frequencies and $\langle E_z\rangle$ is equal to zero. We can find the longitudinal component by calculating $\langle E_z^2\rangle$, which does not vanish; however, this problem is not in the framework of our paper.

At $\omega > \omega_c$, the integrals change sign and, in principle, $\langle E_z\rangle \neq 0$ may exist. However, the critical value of the frequency implies the existence of the critical value of the piezomodule ν_c. This is easy to see in the limiting case, when in the denominator of the subintegral expression the term q^2 may be neglected. From this it follows that the case $\omega > \omega_c$ $(\nu > \nu_c)$ is beyond the limit of applicability of the approximation considered.

Nevertheless, the result obtained may indicate the possibility of the appearance of a non-zero mean amplitude of the longitudinal electromagnetic wave at a rather strong disorder. More accurate investigation of this problem, as well as the study of the character of such an excitation, requires consideration in a separate paper.

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