Attenuation of ultrasound in quasi-one-dimensional ferroelectrics

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It is shown that the anomalies in propagation of ultrasound, observed in recent experiments, with a phase transition in quasi-one-dimensional ferroelectric CsH_2PO_4 are explained by the strong anisotropy of the spectrum of critical fluctuations of the order parameter.

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A considerable interest has recently arisen in studying the phase transition in uniaxial ferroelectric (Fe) CsH_2PO_4 (CDP) and its deuterated analog CsD_2PO_4 (DCDP), which have a number of peculiarities due to the quasi-one-dimensional nature of the ordering of protons (deuterons) in hydrogen bonds.[1] In particular, critical anomalies of the velocity and attenuation of ultrasound in CDP, which differ considerably from the predictions of the theory for uniaxial ferroelectrics with three-dimensional ordering,[3] have been observed by Yakushkin *et al.*[2]

To determine the renormalization of the velocity of sound and its attenuation, it is necessary to calculate the mass operator of acoustical phonons, which interacts with fluctuations of the order parameter. Using the approximation of interacting modes, which gives a very accurate description in the case of uniaxial FE, we obtain for the mass operator

$$\Sigma_\mu(\mathbf{q},\omega) \approx |V_\mu(\mathbf{q})|^2 \Big\{ 4P_0^2\, \chi(\mathbf{q},\omega) + \frac{2}{\pi^2}\iint_{-\infty}^{\infty} \frac{d\omega_1 d\omega_2}{\omega_1-\omega_2-\omega}\,\frac{1}{2}\Big(\mathrm{cth}\frac{\omega_1}{2T}-\mathrm{cth}\frac{\omega_2}{2T}\Big)\sum_{\mathbf{k}} \mathrm{Im}\,\chi(\mathbf{k}-\mathbf{q},\omega_1)\,\mathrm{Im}\,\chi(\mathbf{k},\omega_2)\Big\}\ , \quad (1)$$

where the matrix element of the interaction $V_\mu(\mathbf{q})$ of phonons on the branch μ with frequency $\omega=\omega_\mu(\mathbf{q})$ and wave vector $\mathbf{q}$ is proportional to the electrostriction tensor $g_{\alpha\beta,yy}=\{g_{\alpha\alpha,yy}, g_{xz,yy}\}$, $\alpha = x, y, z$, and y is the FE axis. Fluctuations of the order parameter are described by the relaxation function in the Landau-Khalatnikov approximation:

$$\chi(k,\omega) = \chi(k)\,[\,1 + i\omega\tau(\mathbf{k})\,]^{-1} \quad (2)$$

with relaxation time $\tau(\mathbf{k}) = \tau_0\chi(\mathbf{k})$. Here the critical retardation is described by the static susceptibility $\chi(k)$, which, according to Ref. 1, can be written as follows in the long-wavelength approximation ($k\to 0$): $\chi(\mathbf{k}) = (a(t) + \lambda^2 k_\parallel^2/k^2 + s_\parallel^2 k_\parallel^2 + s_\perp^2 k_\perp^2)^{-1}$. The ratio of the constants determining the variance of the fluctuations across and along the FE axis is: $\eta = s_\perp^2/s_\parallel^2 \approx 10^{-2}$ (CDP)–10^{-3}. (DCDP), $a(t)\sim t^\gamma$ for $t=(T/T_c-1)\to 0$, λ^2 is the dipole interaction constant, and $k^2 = k_\perp^2 + k_\parallel^2$.

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The first term in (1), which is proportional to the square of the equilibrium value of the order parameter P_0, describes the Landau-Khalatnikov relaxation contribution to the ferroelectric phase and has the usual behavior for uniaxial FE (see Ref. 3). The second fluctuation term, which does not depend on the direction of $\mathbf{q}$ in the long-wavelength limit, differs from zero both below and above T_c. A standard estimation of integrals over the frequency in (1), using (2), gives the following expressions for the renormalization of the velocity of sound and its attenuation coefficient:

$$\frac{\Delta c_\mu}{c_\mu} \approx -T \frac{|V_\mu(\mathbf{q})|^2}{\omega_\mu^2(\mathbf{q})} \sum_{\mathbf{k}} \chi^2(\mathbf{k}), \tag{3}$$

$$\alpha_\mu(\mathbf{q}) \approx \frac{T}{c_\mu} |V_\mu(\mathbf{q})|^2 \sum_{\mathbf{k}} \chi^2(\mathbf{k})\,\tau(\mathbf{k}). \tag{4}$$

In view of the strong anisotropy of the spectrum of fluctuations in (2), in estimating the temperature dependence of (3) and (4), the region of quasi-one-dimensional ($1D$) fluctuations (where $\eta\omega_D^2 \ll a^2(T_{1D}) \ll \omega_D^2$, $\omega_D^2 = s_\|^2 k_0^2$, k_0 is the cutoff momentum) must be distinguished from the region of three-dimensional (3D) ordering ($a^2(T_{3D}) \ll \eta\omega_D^2$). In the last (very narrow) region, we obtain the following, usual for uniaxial FE, estimates[3]:

$$\Delta c_\mu / c_\mu \sim (\ln \lambda / a(T)) / \lambda s_\perp^3, \quad \alpha_\mu \sim 1/s_\perp^3 a^2(T), \tag{5}$$

where in the mean-field approximation $a^2(T) \sim t$. In the region of 1D fluctuations, which can be rather broad for small η, the asymptotic estimate of integrals in (3) and (4) is different:

$$\Delta c_\mu / c_\mu \sim [s_\|^2 s_\perp a^2(t)]^{-1} \quad \alpha_\mu \sim [s_\|^2 s_\perp a^4(t)]^{-1}, \tag{6}$$

This situation leads to a stronger anomaly with respect to t than in (6). On the whole, however, the fluctuation contribution for quasi-one-dimensional FE turns out to be

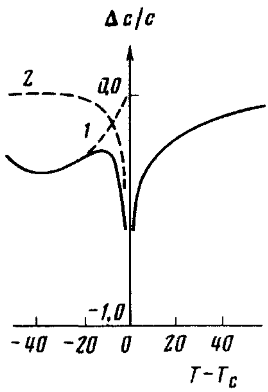

FIG. 1. Temperature dependence of the renormalization of the velocity of longitudinal sound in CDP, propagating along the ferroelectric axis.

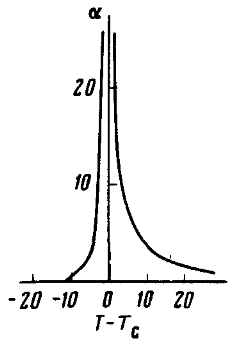

FIG. 2. Temperature dependence of the attenuation factor of longitudinal sound in CDP, propagating along the ferroelectric axis.

large with respect to the anisotropy parameter:

$$\frac{(\Delta c_\mu)_{1D}}{(\Delta c_\mu)_{3D}} \sim \frac{(\alpha_\mu)_{1D}}{(\alpha_\mu)_{3D}} \sim \frac{s_{\parallel}^3}{s_{\perp}^3} \sim 10^3 \tag{7}$$

and in the limit $t \to 0$ (in the region $a^2(t) \ll \omega_D^2$), its asymptotic behavior with respect to t changes. Such a considerable amplification of acoustical anomalies is observed in the experiment described in Ref. 2, in which the values of the critical indices agree qualitatively with asymptotic expression (6).

To make a quantitative comparison of Eq. (1)–(4) with the experiment in Ref. 2, we have also performed numerical calculations on a computer using a more accurate expression for the static susceptibility $\chi(\mathbf{k})$, obtained with a self-consistent solution of the system of equations for a quasi-one-dimensional Ising model in the paraelectric and ferroelectric phases (see, for example, Ref. 4). The results of this calculation for CDP are shown in Figs. 1 and 2. In the ferroelectric phase, the dashed curves show separately the relaxation (1) and fluctuation (2) contributions to the renormalization of the velocity of sound. In selecting the parameters of the model, we use the results of dielectric[4] and neutron[1] measurements: $\eta = 2.9 \times 10^{-2}$, $\lambda^2/J_{\parallel} = 4.93$. The absolute values of $(\Delta c/c)$ and $\alpha(T)$ for longitudinal sound are presented in units of $g^2_{yy,yy}\,\mu^4/c_y^2 M J_{\parallel}$ and $g^2_{yy,yy}\,\omega_y^2 \tau_0 \mu^2/c_y^3 M J_{\parallel}$, respectively, where μ is the dipole moment, and M is the mass per unit cell. Analogous curves were obtained for DCDP (where $\eta = 2.4 \times 10^{-3}$ $\lambda^2/J_{\parallel} = 0.42$), but the absolute values of the quantities (in the same units) turn out to be a factor of 10–20 higher due to the even larger anisotropy. The weak transverse coupling between chains also gives rise to stronger suppression of critical anomalies in an external electric field (along the FE axis): it should be observed in fields that are approximately a factor of η weaker than in isotropic FE.

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