Long-wavelength order parameter fluctuations near the transition to polydomain state in free ferroelectrics and ferromagnets

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In the absence of external fields and currents, ferroelectrics and ferromagnets are known to undergo phase transitions to inhomogeneous (polydomain) structures induced by the dipole-dipole interaction. The long-wavelength order parameter fluctuations near such a transition are studied for $T>T_c$ in the mean-field and scaling regions by considering the quadratic part of the free energy functional for a thin plate of a uniaxial crystal with the polar axis perpendicular to the plate surface. It is shown that the specific anomalies in the long-period responses of a crystal appear due to condensation of the inhomogeneous (periodic) soft mode with period proportional to $\sqrt{L}$ (L is the plate thickness) in accordance with the phenomenological result of Landau and Lifshitz for the c-domain width and the microscopic result of Tarasenko, Chensky, and Dickshtein for $T\rightarrow T_c-0$. The behavior of x-ray and neutron scattering and light propagation in ferroelectrics near such a transition is described. © *1996 American Institute of Physics.* [S1063-7761(96)01109-2]

It is well known that the main cause of the transition to a polydomain structure in ferroelectrics and ferromagnets is the dipole-dipole interaction of the order parameter fluctuations.[1–4] It was shown by Landau and Lifshitz[1] that the reduction of magnetostatic (or electrostatic) energy in a polydomain can be greater than the (positive) domain wall surface energy contribution just below T_c, making the polydomain structure energetically more favorable than the homogeneous one in some interval $T_0<T<T_c$ in free (without external fields, currents, and conductors) ferromagnets and ferroelectrics-insulators. This result actually means that we have transitions in some inhomogeneous (incommensurate) phases in such crystals which must be described by some order parameter functions depending on the form of the crystal. According to Landau and Lifshitz,[1] the c-domain structure period d in a plate of thickness L is proportional to $\sqrt{L}$. Thus in the infinite-volume limit one can regard spatial variations of the order parameter as a surface effect having negligible influence on the critical properties of the crystal. Indeed, this long-period variations could change the spectrum of the soft order parameter fluctuations only at small wave vectors $k \leqslant 1/d \propto 1/\sqrt{L}$. Then the thermodynamical quantities which depend on the whole spectrum of these fluctuations, such as the heat capacity, would not be influenced by these changes. Still it is apparently necessary to determine the actual order parameter distribution and the spectrum of its long-range fluctuations, since some physical properties of free ferroelectrics and ferromagnets can be described only with due account of the specific long-period polydomain structure induced by the dipole-dipole interaction. We can only mention the features in the light and sound propagation and diffuse x-ray and neutron scattering which show the polydomain phase transition to occur. In general, the reaction of a crystal to the external perturbations (fields) with wavelength comparable to the structure period would be significantly modified in comparison to the cases when such structure does not occur, e.g., in a toroidal Heisenberg ferromagnet or short-circuited ferroelectric plate.

The first microscopic treatment of the polydomain phase transition in terms of the free energy functional with magnetostatic contribution was undertaken by Tarasenko, Chensky, and Dickshtein,[5,6] who found in the limit $T\rightarrow T_c-0$ the approximate equilibrium order parameter distribution in the plate of a uniaxial ferromagnet with period $d \propto \sqrt{L}$ and studied the dynamics of the long-wavelength magnetic excitations below T_c. At the same time, the peculiarities of the long-wavelength responses should be exhibited above T_c and their full description could be obtained more simply at least in the mean-field region, since we need to consider only the part of the Landau–Ginzburg free energy functional with the dipole-dipole interaction term quadratic in the order parameter. Moreover, it is easy to show that the mean-field results are also valid in the fluctuation region in the immediate vicinity of T_c after substitution of the renormalized parameters. So in the present work we obtain the exact order parameter correlation function describing the anomalies of long-wavelength responses near the polydomain phase transition for all $T>T_c$ and describe the behavior of x-ray and neutron scattering and light propagation in ferroelectrics near such a transition in a plate of the uniaxial crystal.

To study the phase transition to the polydomain phase in a regular way instead of construction of domain-wall models[1,4,7–9] one should treat the Landau–Ginzburg free energy functional with the dipole-dipole interaction term. In the mean-field region above T_c it is sufficient to consider the part quadratic in the order parameter, which has the form

$$F=\frac{1}{2}\int_V d\mathbf{x}\int_V d\mathbf{x}'\,\mathbf{P}(\mathbf{x})[\hat{\chi}^{-1}(\mathbf{x}-\mathbf{x}')+D_{\alpha\beta}(\mathbf{x}-\mathbf{x}')]\mathbf{P}(\mathbf{x}'), \tag{1}$$

$$D_{\alpha\beta}(\mathbf{x}-\mathbf{x}')\equiv-\nabla_\alpha\nabla_\beta\frac{1}{|\mathbf{x}-\mathbf{x}'|}. \tag{2}$$

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Here $\hat{\chi}(\mathbf{x})$ is the susceptibility tensor with the short-range (Ornstein–Zernike) space dependence and some (or all) divergent $\mathbf{k}=0$ Fourier components

$$\hat{\chi}\equiv\frac{1}{V}\int_V d\mathbf{x}\hat{\chi}(\mathbf{x})=\frac{\hat{C}}{T-T_c}.$$

The order parameter field $\mathbf{P}(\mathbf{x})$ is the polarization in ferroelectrics and the magnetization in ferromagnets. The form of the dipole-dipole interaction term implies the absence of conductors in the ferroelectric case and currents in the case of ferromagnet.

It is easy to see that generally there is no instability with respect to the homogeneous order parameter fluctuations at T_c. Setting $\mathbf{P}(\mathbf{x})=\mathbf{P}$ in (1), we have

$$F=\frac{V}{2}\mathbf{P}(\hat{\chi}^{-1}+4\pi\hat{n})\mathbf{P}, \tag{3}$$

$$n_{\alpha\beta}=-\frac{1}{4\pi V}\int_V d\mathbf{x}\int_V d\mathbf{x}'\nabla_\alpha\nabla_\beta\frac{1}{|\mathbf{x}-\mathbf{x}'|}=\frac{1}{4\pi V}\int_S\int_S\frac{dS_\alpha dS'_\beta}{|\mathbf{x}-\mathbf{x}'|}. \tag{4}$$

The tensor $\hat{n}$ could be called the effective tensor of the depolarizing coefficients, because the exact expression for the (homogeneous) electric field in the ellipsoids with $\mathbf{P}(\mathbf{x})=P$ is[4] $\mathbf{E}=-4\pi\hat{n}\mathbf{P}$. In general case $\hat{n}$ always has nonzero (positive) components due to the relation Tr $\hat{n}=1$, which follows from the equation

$$\Delta\frac{1}{|\mathbf{x}|}=-4\pi\delta(\mathbf{x}).$$

Thus instability with respect to homogeneous fluctuations can occur at T_c only in samples of special form having $n_{\alpha\alpha}\approx n_{\alpha\beta}\approx 0$ if $(\hat{\chi}^{-1})_{\alpha\alpha}\to 0$. The homogeneous phase transition is also possible in the case of a ferroelectric having on its surface conductors with fixed potentials. Then $\hat{D}(\mathbf{x}-\mathbf{x}')$ in Eq. (1) must be replaced by

$$(D_c)_{\alpha\beta}(\mathbf{x}-\mathbf{x}')\equiv-\nabla_\alpha\nabla_\beta G(\mathbf{x},\mathbf{x}'),$$

where $G(\mathbf{x},\mathbf{x}')$ is the Green's function of the Laplace equation satisfying the boundary conditions on the conductor surfaces[10]

$$G(\mathbf{x},\mathbf{x}')|_{\mathbf{x}\in S}=G(\mathbf{x},\mathbf{x}')|_{\mathbf{x}'\in S}=0.$$

The corresponding substitution in Eq. (4) yields $\hat{n}=0$ when the conductor surfaces cover those of ferroelectric sample, and we have a transition to the homogeneous state at T_c in this case.

When the depolarizing field forbids the homogeneous state below T_c, we can find the actual order parameter function $\mathbf{P}_s(\mathbf{x})$ having the most divergent fluctuations just by diagonalizing F in Eq. (1). That is, we should expand

$$\mathbf{P}(\mathbf{x})=\sum_{n=1}^{\infty}c_n\mathbf{P}_n(\mathbf{x})$$

via the eigenfunctions of the linear operator defining F, i.e.,

$$\int_V d\mathbf{x}'[\hat{\chi}^{-1}(\mathbf{x}-\mathbf{x}')+\hat{D}(\mathbf{x}-\mathbf{x}')]\mathbf{P}_n(\mathbf{x}')=\lambda_n\mathbf{P}_n(\mathbf{x}). \tag{5}$$

Then

$$F=\frac{1}{2}\sum_{n=1}^{\infty}\lambda_n c_n^2$$

and the most divergent fluctuations would be associated with the mode with the lowest eigenvalue, say λ_1. Hence the transition occurs as a condensation of $\mathbf{P}_s(\mathbf{x})=\mathbf{P}_1(\mathbf{x})$ when λ_1 becomes zero.

Here we consider the simplest case of an (infinite) plate with finite thickness L, as in.[5,6] Let its surfaces be at $z=\pm L/2$ and introduce

$$\mathbf{P}_{k_z}(\mathbf{k}_\perp)=\frac{1}{\sqrt{L}}\int_{-L/2}^{L/2}dze^{ik_z z}\int d\mathbf{x}_\perp e^{i\mathbf{k}_\perp\mathbf{x}_\perp}\mathbf{P}(\mathbf{x}). \tag{6}$$

Here $k_z=2\pi n/L$, $n=0,\pm 1,\pm 2\ldots$. In this representation the dipole–dipole interaction term can be expressed as

$$D_{\alpha\beta}(\mathbf{k},\mathbf{k}')=2L\int d\mathbf{p}\frac{\mathbf{p}_\alpha\mathbf{p}_\beta}{p^2}S(p_z-k_z)S(p_z-k'_z)\delta(\mathbf{p}_\perp-\mathbf{k}_\perp)\delta(\mathbf{p}_\perp-\mathbf{k}'_\perp),$$
$$S(x)=\frac{2}{xL}\sin\frac{xL}{2}. \tag{7}$$

The integration in Eq. (7) can be easily performed, and we have the following expression for F in terms of $\mathbf{P}_{k_z}(\mathbf{k}_\perp)$:

$$F=\frac{1}{2}\sum_{k_z,k'_z}\int\frac{d\mathbf{k}_\perp}{(2\pi)^2}\mathbf{P}^*_{k_z}(\mathbf{k}_\perp)\hat{G}^{-1}_{k_z,k'_z}(\mathbf{k}_\perp)\mathbf{P}_{k'_z}(\mathbf{k}_\perp), \tag{8}$$

$$(\hat{G}^{-1}_{k_z,k'_z})_{\alpha\beta}(\mathbf{k}_\perp)=\left[(\chi^{-1})_{\alpha\beta}(\mathbf{k})+4\pi\frac{k_\alpha k_\beta}{k^2}\right]\delta_{k_z,k'_z}+\frac{4\pi}{k_\perp L}(1-e^{-k_\perp L})[(\mathbf{c}^\perp_{k_z})_\alpha(\mathbf{c}^\perp_{k'_z})_\beta-\mathbf{c}^\parallel_{k_z}:\mathbf{c}^\parallel_{k'_z}],$$
$$\mathbf{c}^\perp_{k_z}(\mathbf{k}_\perp)=\exp\left(\frac{ik_zL}{2}\right)\frac{k_z\mathbf{k}_\perp-k_\perp^2\mathbf{e}_z}{k^2},$$
$$\mathbf{c}^\parallel_{k_z}(\mathbf{k}_\perp)=\exp\left(\frac{ik_zL}{2}\right)\frac{k_\perp\mathbf{k}}{k^2}. \tag{9}$$

Equation (5) becomes

$$\sum_{k'_z}\hat{G}^{-1}_{k_z,k'_z}(\mathbf{k}_\perp)\mathbf{P}_n(\mathbf{k}_\perp,k'_z)=\lambda_n(\mathbf{k}_\perp)\mathbf{P}_n(\mathbf{k}). \tag{10}$$

The notation $\hat{G}^{-1}_{k_z,k'_z}(\mathbf{k}_\perp)$ is convenient because in the present quadratic (mean-field) approximation

$$\langle P^*_\alpha(\mathbf{k})P_\beta(\mathbf{k}')\rangle=T(G_{k_z,k'_z})_{\alpha\beta}(\mathbf{k}_\perp)\delta(\mathbf{k}_\perp-\mathbf{k}'_\perp).$$

Note that the Fourier transform (6) diagonalizes $\hat{\chi}(\mathbf{x}-\mathbf{x}')$ but not $\hat{D}(\mathbf{x}-\mathbf{x}')$. This difference results from the long-range character of the $\hat{D}(\mathbf{x})$ space dependence. Actually the exact diagonalization requires along with the dependence on the difference $\mathbf{x}-\mathbf{x}'$ that the transforming function has periodicity of L along the z-axis. Since $\hat{\chi}(\mathbf{x}-\mathbf{x}')$ is localized at $|\mathbf{x}-\mathbf{x}'|<a$, where a is on the order of the lattice parameter, the last condition fails only in a small part of the period on the order of a/L. Thus the Fourier transform of $\hat{\chi}(\mathbf{x}-\mathbf{x}')$ has a term of the same order nondiagonal in k_z which can be dropped when $L\gg a$. At the same time, the only length scale which enters $\hat{D}(\mathbf{k}_\perp, z-z')$ is $k_\perp^{-1}$, and the nondiagonal term in Eq. (10) is small only for $k_\perp L \gg 1$. For $k_z = k_z' = 0$ and $k_\perp \to 0$ it has the finite limit, i.e.,

$$\lim_{k_\perp\to 0} (G_{0,0}^{-1})_{\alpha\beta}(\mathbf{k}_\perp) = (\chi_0^{-1})_{\alpha\beta} + 4\pi(e_z)_\alpha(e_z)_\beta,$$

in accordance with the above considerations.

Thus, there is no depolarizing field for the modes with $P_n^z(\mathbf{k}=0)=0$ (if they exist) which actually means that we neglect it when we consider the infinite plate. This neglect can be justified if the condition

$$P_n^z(\mathbf{k}) \gg |\mathbf{P}_n^\perp(\mathbf{k})|$$

holds for the modes with small λ_n which we are interested in. There is only one case when such an assumption can hold and the consideration of the infinite plate makes sense in the uniaxial ferroelectric (ferromagnet) with polar axis perpendicular to the plate surface. Then

$$\hat{\chi}_{\alpha\beta}^{-1}(\mathbf{k}) = \delta_{\alpha\beta}[\chi_\perp^{-1}(1-\delta_{\alpha z}) + \chi_\parallel^{-1}(\mathbf{k})\delta_{\alpha z}],$$
$$\chi_\perp^{-1}(\mathbf{k}) = \chi_\parallel^{-1} + l_\perp^2 k_\perp^2 + l_\parallel^2 k_z^2, \quad \chi_\parallel^{-1} = 4\pi(T-T_c)/C. \tag{11}$$

Here we ignore the wave-vector dependence of the noncritical $\chi_\perp$ and write down explicitly the expansion of $\chi_\parallel^{-1}(\mathbf{k})$ at small $\mathbf{k}$. Then we can use the x and y components of Eq. (10) to express $P_n^z(\mathbf{k})$ via $\mathbf{P}_n^\perp(\mathbf{k})$. For $\lambda_n(\mathbf{k}_\perp)\chi_\perp \ll 1$ we have

$$\mathbf{P}_n^\perp(\mathbf{k}) = (\varepsilon_\perp - 1)\mathbf{k}_\perp \Bigg\{ -\frac{k_z P_n^z(\mathbf{k})}{\varepsilon_\perp^2 k_\perp^2 + k_z^2} + \frac{2\sqrt{\varepsilon_\perp} k_\perp}{L k_z} \sum_{k_z'} [\varepsilon_\perp k_\perp^2 a_{+1}(\mathbf{k}) a_{+1}(k_\perp, k_z') + k_z^2 a_{-1}(\mathbf{k}) a_{-1}(\mathbf{k}_\perp, k_z')] P_n^z(\mathbf{k}_\perp, k_z') \Bigg\}, \tag{12}$$

where

$$a_\sigma(\mathbf{k}) \equiv \frac{\exp(ik_z L/2)(k_z/\sqrt{\varepsilon_\perp} k_\perp)^{(1+\sigma)/2}}{(\varepsilon_\perp k_\perp^2 + k_z^2)\sqrt{\varepsilon_\perp^{\sigma/2} + \coth\sqrt{\varepsilon_\perp} k_\perp L/2}},$$
$$\varepsilon_\perp = 1 + 4\pi\chi_\perp.$$

To obtain Eq. (12) the summation over $k_z = 2\pi n/L$, $n=0$ $\pm 1, \pm 2\ldots$ was performed using the relation

$$\sum_{k_z=-\infty}^{\infty} A(k_z) = \frac{L}{2} \oint_C \frac{du}{2\pi i} \frac{\tilde{A}(u)}{\tan(uL/2)},$$

where the contour C encircles the real axis and $\tilde{A}(u)$ is the analytical continuation of $A(k_z)$. Eliminating $\mathbf{P}_n^\perp(\mathbf{k})$ from the z-component in Eq. (10) we get

$$\left[\chi_\parallel^{-1}(\mathbf{k}) + \frac{4\pi k_z^2}{(\varepsilon_\perp k_\perp^2 + k_z^2) - \lambda_n(\mathbf{k}_\perp)}\right] P_n^z(\mathbf{k}) + \frac{8\pi}{L}(\sqrt{\varepsilon_\perp} k_\perp)^3 \sum_{k_z'} [a_{-1}(\mathbf{k}) a_{-1}(\mathbf{k}_\perp, k_z') - a_1(\mathbf{k}) a_1(\mathbf{k}_\perp, k_z')] P_n^z(\mathbf{k}_\perp, k_z') = 0.$$

The solution of this equation has the form

$$P_n^z(\mathbf{k}) = \frac{c_n(\mathbf{k}_\perp) a_{\sigma_n}(\mathbf{k})}{\chi_\parallel^{-1}(\mathbf{k}) + 4\pi k_z^2/(\varepsilon_\perp k_\perp^2 + k_z^2) - \lambda_n(\mathbf{k}_\perp)},$$
$$\sigma_n = (-1)^n. \tag{13}$$

Representing $\lambda_n(\mathbf{k}_\perp)$ as

$$\lambda_n(\mathbf{k}_\perp) = \chi_\parallel^{-1}(\mathbf{k}_\perp) + q_n^2\left(l_\parallel^2 + \frac{4\pi}{q_n^2 + \varepsilon_\perp k_\perp^2}\right),$$
$$q_n(k_\perp) = \frac{\pi n - 2\delta_n(k_\perp)}{L}, \quad |\delta_n(k_\perp)| < \frac{\pi}{2}, \quad n = 1,2,\ldots, \tag{14}$$

we get the equation for $\delta_n(k_\perp)$:

$$l_\parallel^2(q_n^2 + \varepsilon_\perp k_\perp^2)^2 \left[1 + \frac{(\alpha_n/\varepsilon_\perp l_\parallel k_\perp)^{\sigma_n}}{\tanh(\alpha_n L/2l_\parallel)}\right] = 4\pi\varepsilon_\perp k_\perp^2 \left[\left(\frac{q_n}{\varepsilon_\perp k_\perp \tan\delta_n}\right)^{\sigma_n} - 1\right],$$

$$\alpha_n(k_\perp) = \sqrt{\frac{\varepsilon_\perp k_\perp^2}{q_n^2 + \varepsilon_\perp k_\perp^2}}[4\pi + l_\parallel^2(q_n^2 + \varepsilon_\perp k_\perp^2)].$$

For

$$\pi n/L \ll \sqrt{\varepsilon_\perp} k_\perp \ll l_\parallel^{-1}, \tag{15}$$

we have

$$\delta_n(k_\perp) \approx \frac{\pi n}{\varepsilon_\perp k_\perp L}, \quad \lambda_n(k_\perp) \approx \chi_\parallel^{-1} + l_\perp^2 k_\perp^2 + 4\pi\left(\frac{\pi n}{\sqrt{\varepsilon_\perp} k_\perp L}\right)^2.$$

Hence the lowest eigenvalue belongs to the mode with $n=1$ and $k_\perp = k_0$,

$$k_0 \equiv \frac{(4\pi^3/\varepsilon_\perp)^{1/4}}{\sqrt{L l_\perp}}. \tag{16}$$

Note that this value of k_0 satisfies the condition (15) for

$$L \gg \max(l_\perp, l_\parallel^2/l_\perp).$$

Then

$$\lambda_{\min} \equiv \lambda_1(k_0) \approx \chi_\parallel^{-1} + 2l_\perp^2 k_0^2 = \chi_\parallel^{-1} + 4\pi\sqrt{\frac{\pi}{\varepsilon_\perp}\frac{l_\perp}{L}} \tag{17}$$

differs from $\chi_\parallel^{-1}$ by a small quantity of order $l_\perp/L$. Thus the transition to the polydomain structure takes place at $T = T_c'$, which is only slightly below T_c. From Eqs. (11) and (17) we get

$$T_c' = T_c - C\sqrt{\frac{\pi}{\varepsilon_\perp}\frac{l_\perp}{L}}.$$

The period of the resulting (domain) structure is

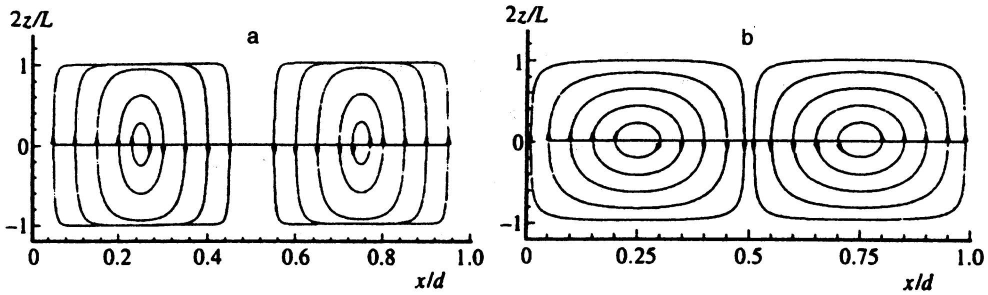

FIG. 1. The directions of $\mathbf{P}_s(\mathbf{x})$ for $\varepsilon_\perp=1.1$ (a) and $\varepsilon_\perp=10$ (b).

$$d=2\pi/k_0=(4\pi\varepsilon_\perp)^{1/4}\sqrt{Ll_\perp}. \tag{18}$$

We obtain the same expression for d as was found in Refs. 5 and 6 for $T<T_c$ confirming the $\sqrt{L}$-dependence of the period as in the Landau and Lifshitz phenomenological result for the c-domain structure.[1] This result also agrees with the experiments in the uniaxial ferroelectric rochelle salt.[3] For $l_\perp\approx a\approx 10^{-9}$ m, $L=10^{-3}$ m, $\varepsilon_\perp\approx 1-10$ we have $d\approx 10^{-6}$ m, which is the typical value for the c-domain structure period in ferroelectrics[11,12] and ferromagnets.[2] Indeed in rochelle salt we have $\sqrt{4\pi\varepsilon_\perp}l_\perp\approx 2\cdot 10^{-8}\,\mathrm{m}^3$ and $\varepsilon_\perp\approx 10$.[12] Hence we have $l_\perp\approx 2\cdot 10^{-9}$ m, which is on the order of lattice parameters in this crystal. The Fourier transform of Eqs. (12) and (13) with $n=1$ and $c_1(\mathbf{k}_\perp)=\delta(k_x-k_0)+\delta(k_x+k_0)$ gives the following order parameter function:

$$\begin{aligned}P_s^z(\mathbf{x})&=\left[\cos[\pi-2\delta_1(k_0)]\,\frac{z}{L}\right.\\&\left.\quad-\frac{\sqrt{\pi}l_\parallel}{2L}\,\frac{\cosh(2\sqrt{\pi}z/l_\parallel)}{\sinh(\sqrt{\pi}L/l_\parallel)}\right]\cos(k_0x),\\P_s^x(\mathbf{x})&=\left[\sin[\pi-2\delta_1(k_0)]\,\frac{z}{L}\right.\\&\left.\quad-\frac{\varepsilon_\perp l_\parallel^2k_0^2}{4\pi}\,\frac{\sinh(2\sqrt{\pi}z/l_\parallel)}{\sinh(\sqrt{\pi}L/l_\parallel)}\right]\cos(k_0x).\end{aligned} \tag{19}$$

This result differs from that found in Refs. 5 and 6 in the limit $T\to T_c-0$ only by the small surface terms with cosh and sinh which were neglected in Refs. 5 and 6 and small higher-order harmonics whose amplitudes vanish at T_c. Curves indicating the directions of $\mathbf{P}_s$ are shown in Fig. 1. Part of the period is occupied by closed loops which do not come out on the surface. Its width can be estimated as

$$w=\frac{d}{\pi}\arccos\left(\frac{d}{2\varepsilon_\perp L}\right)^{1-1/\varepsilon_\perp}.$$

We have

$$w\ll d,\quad \varepsilon_\perp-1\ll 1,\quad w\approx d/2,\quad \varepsilon_\perp\gg 1.$$

Here we note that the $\mathbf{P}_s$ given by Eq. (19) appearing at $T=T_c'$ is substantially modified at lower temperatures. It is rather difficult to find the order parameter distribution below T_c', as this requires the determination of the minimum of the free energy functional with the P^4 term.[5,6] Evidently this term will generate higher-order mode contributions to $\mathbf{P}_s$ given by Eq. (19) as secondary order parameters[4] which could make $\mathbf{P}_s$ more like the experimentally observed domain structures far below T_c'. Still, the period of the c-domain structure generated in this way will be the same value of d in Eq. (18) for all $T<T_c'$. The validity of these general considerations was explicitly demonstrated in Refs. 5 and 6.

At the same time, in the mean-field region above T_c' it is the softening of the $\mathbf{P}_1$ mode that determines the behavior of crystal responses to inhomogeneous external perturbations. Thus, the singular part of the intensity of diffuse neutron and x-ray scattering in ferroelectrics and magnetic neutron scattering in ferromagnets is described by the structure factor $S_\parallel(\mathbf{k})=G^{zz}_{k_z,k_s}(\mathbf{k}_\perp)$. For $k_z=0$ and

$$L^{-1}\ll k_\perp\sqrt{\varepsilon_\perp}\ll l_\parallel^{-1}, \tag{20}$$

inversion of the matrix $\hat{G}^{-1}_{k_z,k_z'}$ in Eq. (9) gives

$$\begin{aligned}S_\parallel(k_\perp)&=\chi_\parallel(k_\perp)\{1-u^{-1}(k_\perp)\tanh[u(k_\perp)]\},\\u(k_\perp)&=\frac{k_\perp L\sqrt{\varepsilon_\perp}}{2\sqrt{4\pi\chi_\parallel(k_\perp)}}.\end{aligned} \tag{21}$$

Using Eqs. (11) and (16), we can also rewrite $u(k)$ as

$$u(k_\perp)=\sqrt{\kappa^2+k_\perp^2}\,\frac{\pi k_\perp}{2k_0^2},\quad \kappa^2=l_\perp^{-2}\chi_\parallel^{-1}.$$

Note that $\chi_\parallel$ and κ^2 can become negative near T_c'. Then $u(k_\perp)$ becomes imaginary, $u(k_\perp)=i|u(k_\perp)|$, for $k_\perp^2<-\kappa^2$ but expression (21) still holds for such $k_\perp$ and can be represented as

$$S_\parallel(k_\perp)=|\chi_\parallel(k_\perp)|[|u^{-1}(k_\perp)|\tan|u(k_\perp)|-1].$$

Specifically, $\kappa^2=-2k_0^2$ holds at $T=T_c'$ (cf. Eq. (17)) and for $k_\perp^2<2k_0^2$ at the transition point we have

$$S_\parallel(k_\perp)=\frac{1}{l_\perp^2(2k_0^2-k_\perp^2)}\times\left[\frac{2k_0^2}{\pi k_\perp\sqrt{2k_0^2-k_\perp^2}}\tan\left(\frac{\pi k_\perp\sqrt{2k_0^2-k_\perp^2}}{2k_0^2}\right)-1\right].$$

Thus, at $T=T_c'$ the intensity of diffuse scattering diverges at $k=k_0$, as one would expect for the transition to incommensurate structure with the period $d=2\pi/k_0$.

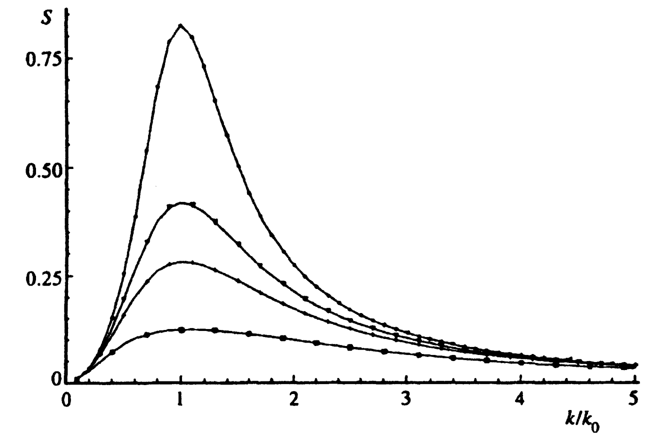

FIG. 2. The intensity of diffuse scattering for $\kappa^2=5k_0^2$ (□), $\kappa^2=k_0^2$ (+), $\kappa^2=0$ (△) and $\kappa^2=-k_0^2$ (*).

The diffuse scattering profiles are shown in Fig. 2 for several $T > T_c'$. They exhibit the peaks near $k=k_0$ which are not difficult to observe experimentally especially in the cases of small $l_\perp$ values and thin samples.

Another spectacular effect appears near T_c' for light propagating in the ferroelectric plate of the geometry in question. It consists of drastic changes in the refractive index dispersion due to the dipole-dipole interaction at long wavelengths $\lambda\approx d$. To describe it one must determine the dynamical dielectric susceptibility

$$\hat{\chi}(\mathbf{k},\omega)=\frac{\partial\mathbf{P}(\mathbf{k},\omega)}{\partial\mathbf{E}(\mathbf{k},\omega)}.$$

When the ferroelectric order parameter fluctuations are soft phonons, the susceptibility $\hat{\chi}(\mathbf{k},\omega)$ can be obtained from equations of motion of the form

$$v^{-2}l_\perp^2(\ddot{\mathbf{P}}+2\Gamma\dot{\mathbf{P}})=-\frac{\partial F}{\partial\mathbf{P}}+\mathbf{E}(\mathbf{k},\omega)\cos(\mathbf{kx}-\omega t). \qquad (22)$$

Here Γ is the phonon damping constant and v is the group velocity of phonons at $T = T_c'$. For $k\gg k_0$ Eq. (22) just describes the propagation of the ordinary longitudinal (LO) and transverse (TO) optical phonons with the frequencies

$$\Omega_{TO1}^2(\mathbf{k})=\frac{v^2}{l_\parallel^2\chi_\parallel(\mathbf{k})},\quad \Omega_{TO2}^2(\mathbf{k})=\frac{v^2}{l_\parallel^2\chi_\perp},$$

$$\Omega_{LO}^2(\mathbf{k})=\frac{v^2}{l_\perp^2}(\chi_\perp^{-1}+4\pi).$$

But it appears that the usual long-wavelength polariton dispersion becomes essentially modified for light with $\lambda\approx d$ propagating in the xy plane and polarized along the polar z-axis when this axis is perpendicular to the plate surface. In this case from (22) for $\chi_{zz}(\mathbf{k}_\perp,k_z=0,\omega)\equiv\chi_\parallel(k_\perp,\omega)$ at $k_\perp$ satisfying the condition (20) we have

$$\chi_\parallel(k_\perp,\omega)=v^2\frac{[1-u^{-1}(k_\perp,\omega)\tanh u(k_\perp,\omega)]}{l_\perp^2[\Omega_{TO1}^2(k_\perp)-2i\Gamma\omega-\omega^2]},$$
$$u(k_\perp,\omega)\equiv\frac{\pi}{2}\frac{vk_\perp}{\Omega_0^2}\sqrt{\Omega_{TO1}^2(k_\perp)-2i\Gamma\omega-\omega^2}, \qquad (23)$$
$$\Omega_0\equiv vk_0.$$

Then we obtain the refractive index $n_{zz}\equiv n_\parallel(\omega)$ as the solution of the equation

$$n_\parallel^2=1+4\pi\chi_\parallel(n_\parallel\omega/c,\omega).$$

Here c is the velocity of light. Assuming that

$$|u(n_\parallel\omega/c,\omega)|<1 \qquad (24)$$

and using the smallness of the v/c ratio, we have

$$n_\parallel^2\simeq2\Omega_L^2\left\{\Omega_L^2-\omega^2+\sqrt{(\Omega_L^2-\omega^2)+\frac{2\pi^2}{5}\frac{v^2\Omega_L^2\omega^4}{c^2\Omega_0^4}[\Omega_{TO1}^2(0)-2i\Gamma\omega-\omega^2]}\right\}^{-1},$$

$$\Omega_L\equiv2\sqrt{\frac{3}{\varepsilon_\perp}\frac{c}{L}}.$$

The conditions for validity of this expression (Eqs. (20) and (24)) can be represented as

$$\omega\leqslant\Omega_L,\quad \sqrt{\varepsilon_\perp}\Omega_L/\omega\ll|n_\parallel|\ll L/l_\parallel.$$

Thus $n_\parallel'\gg n_\parallel''$ holds for $\omega<\Omega_L$ and $n_\parallel'$ grows rapidly in the limit $\omega\to\Omega_L-0$, while for $\omega>\Omega_L$ the light waves become overdamped. We can relate this light absorption to the excitation of the specific collective motion with the frequency Ω_L defined by the plate thickness L. For $L=1$ mm we have $\Omega_L/2\pi c\approx10$ cm^{-1}, and such excitation could be easily observed in the IR experiments.

It is easy to see that the above results remain valid in the fluctuation region above T_c after substitution of the renormalized critical susceptibility[13]

$$\chi_\parallel\to\tilde{\chi}\propto(T-T_c)^{-1}[\ln(T-T_c)^{-1}]^{1/3}$$

and renormalized (finite) $\tilde{l}_\parallel$ and $\tilde{l}_\perp$ instead of the bare values. This is because fluctuations renormalize only the short-range part of the inverse order parameter correlator (i.e., $\tilde{\chi}^{-1}(\mathbf{k})$), while the dipole-dipole interaction term stays the same.[13]

Here we note that the results presented in this work for ideal crystals can be very different in real crystals with impurities and defects. Still, we believe that the approach outlined here to the study of phase transitions to the inhomogeneous state in systems with the dipole–dipole interaction can be also useful for the study of more realistic cases.

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