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DEPENDENCE OF SOFT PHONON SPECTRA ON FLEXOELECTRIC COUPLING IN FERROELECTRICS[1]

Analytical expressions describing the frequency dispersion of the soft transverse acoustic (TA) and optic (TO) phonon modes in uniaxial ferroelectrics, as well as their dependence on the flexoelectric coupling constant f, have been analyzed in the framework of the Landau–Ginzburg–Devonshire theory. A critical behavior of the TA mode with respect to the f magnitude is revealed.

K e y w o r d s: Landau–Ginzburg–Devonshire theory, flexoelectric coupling, soft phonon modes.

1. Introduction

Unique physical properties of nanosized ferroics attract a permanent attention of researchers [1]. In this work, we would like to focus attention on the most striking and interesting phenomena induced by the flexoelectric effect in both fundamental and applied physics. This effect was first predicted theoretically by Mashkevich and Tolpygo in 1957 [2]. It is inherent in any material, which makes it universal [3–6]. Kvasov and Tagantsev [7] predicted the existence of a cross-term in the kinetic energy and coined its dynamic flexoelectric effect (see reviews [4, 5] and references therein). The influence of flexoelectricity is very important in nanoscaled objects, in which strong strain gradients are inevitably present near the surfaces, in thin films [8–10], at domain walls, and at ferroelectric interfaces [8, 11–13].

Dynamic characteristics of phase transitions in ferroics have attracted a keen attention of scientists for many years, being a source of the valuable information for fundamental physical researches and advanced applications [14]. Any phase transition leads to the instability of a certain soft phonon vibration mode, and static displacements of atoms taking place at the phase transition correspond to frozen displacements of this mode [15]. In particular, for ferroelectrics, the frequency $\omega_{\rm TO}$ of the transverse optic soft mode depends on the temperature T, with $\omega_{\rm TO}(T_{\rm C}) = 0$ at the transition temperature $T = T_{\rm C}$. The main experimental methods that allow one to obtain information about the soft modes and the spatial modulation of the order parameter in ferroics include dielectric measurements [16], inelastic neutron scattering [15, 17–21], X-ray [22–24], Raman [25], and Brillouin [21, 22, 26–29] scattering, as well as the ultrasonic pulse-echo method [25, 27] providing hypersound spectroscopic information.

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The influence of the static and dynamic flexoelectric couplings on soft phonon spectra in ferroics has not been studied till now [30, 31]. In the cited research, we used the Landau–Ginzburg–Devonshire (LGD) approach to consider the influence of the flexocoupling on the appearance of spatially modulated phases (SMPs) and on the properties of optic and acoustic phonons in various ferroelectric and paraelectric phases. Flexocoupling-induced soft acoustic modes and a SMP in ferroelectrics were revealed in work [32].

In this work, in the framework of the LGD theory, we will analyze expressions for the frequency dispersion $\omega(k)$ of soft transverse acoustic (TA) and optic (TO) phonon modes, as well as their dependence on the flexoelectric coupling constant f in ferroelectrics. Using the one-component approximation, which is valid for a number of uniaxial ferroelectrics, we revealed a critical behavior of the TA mode with respect to the magnitude of this parameter, which agrees with the results of work [32].

2. Analytical Description

In the simplest one-dimensional (1D) and one-component case, which is considered below, the Lagrange function L consists of the free energy F and the kinetic energy K [4, 5]:

$$L = \int\limits_t dt\,(F - K). \tag{1}$$

The bulk part of the free energy F depends on the polarization component P, strain component u, and their gradients. It looks like [32]

$$F_V = \int dx \times \times \left(\begin{array}{l} \frac{\alpha(T)}{2}P^2 + \frac{\beta}{4}P^4 + \frac{\gamma}{4}P^6 + \frac{g}{2}\left(\frac{\partial P}{\partial x}\right)^2 - PE - \\ - quP^2 + \frac{c}{2}u^2 + \frac{v}{2}\left(\frac{\partial u}{\partial x}\right)^2 - \frac{f}{2}\left(P\frac{\partial u}{\partial x} - u\frac{\partial P}{\partial x}\right) \end{array} \right). \tag{2}$$

According to the Landau theory [33, 34], the coefficient α for proper ferroelectrics linearly depends on the temperature T: $\alpha(T) = \alpha_T(T - T_C)$, where T_C is the Curie temperature. All other coefficients in Eq. (1) are supposed to be temperature-independent. The coefficient $\beta > 0$ for ferroics with the second-order phase transition, and $\beta < 0$ for ferroics with the first-order one. The nonlinear stiffness γ should be non-negative, $\gamma \geq 0$, for the stability of functional (1) at all P. The gradient coefficients $g > 0$ and $v < 0$ determine the gradient energy. The polarization P interacts with the external electric field E. We also assume that the depolarization field is absent. The considered case corresponds to the transverse variation of polarization components and strains. The electrostriction coefficient q can be either positive or negative. The elastic stiffness c and the strain gradient coefficient ν have to be always positive to provide the functional stability. The coefficient f is the component of the static flexocoupling tensor, and its sign is not fixed.

The kinetic energy

$$K = \int dx \left(\frac{\mu}{2}\left(\frac{\partial P}{\partial t}\right)^2 + M\frac{\partial P}{\partial t}\frac{\partial U}{\partial t} + \frac{\rho}{2}\left(\frac{\partial U}{\partial t}\right)^2 \right), \tag{3}$$

includes the dynamic flexocoupling [7] with magnitude M, ρ is the material density, and μ a kinetic coefficient. The elastic displacement component U is related to the strain u as $u = \partial U/\partial x$.

The dependence of the frequency ω of a soft phonon on its wave vector k, i.e. the dispersion law $\omega(k)$, can be calculated from the time-dependent dynamic equations of state for the polarization, P, and elastic displacement, U, components [30–32]. The dynamic equations of state are obtained by varying the Lagrange function (2) with respect to P and U. The solution of those equations was found in the linear approxinmation, in vicinities of the spontaneous values

$$P_S^2 = \frac{1}{2\gamma}\left(\sqrt{\beta^{*2} - 4\alpha\gamma} - \beta^*\right)$$

and $u_S = \frac{q}{c}P_S^2$, where the coefficient $\beta^* = \beta - 2\frac{q^2}{c}$. The solution looks like [31, 32]

$$\omega_{\mathrm{O,A}}^2(k) = \frac{C(k) \pm \sqrt{D(k)}}{2(\mu\rho - M^2)}, \tag{4}$$

where the function

$$C(k) = \alpha_S\rho + (c\mu - 2fM + g\rho)\,k^2 + \mu v k^4$$

and the determinant

$$D(k) = C^2(k) - 4\left(\mu\rho - M^2\right)k^2 \times \times \left(\alpha_S c - 4q^2P_S^2 + \left(cg + \alpha_S v - f^2\right)k^2 + gvk^4\right)$$

were introduced. The temperature-dependent coefficient $\alpha_S = \alpha_S(T)$ equals

$$\alpha_S(T) = \alpha(T) + \left(3\beta - 2\frac{q^2}{c}\right) P_S^2(T) + 5\gamma P_S^4(T). \quad (5)$$

The dispersion relation (4) describes one optical (O) and one acoustic (A) phonon modes, for which the sign "+" or "−", respectively, before the radical should be taken.

3. Dispersion Dependence on Parameters

In order to make a general analysis of dependence (4) in a material-independent form, let us introduce the following dimensionless parameters [31]: the frequency $\omega^* = \frac{\sqrt{4v\rho}}{c}\omega$ and the wave vector $k^* = \frac{ak}{\pi}$, as well as

$$a^* = \frac{a}{\pi}\sqrt{\frac{c}{2v}}, \quad F^* = \frac{f^2}{cg},$$
$$\alpha_v^* = \frac{v}{cg}\left(\alpha + \left(3\beta - 2\frac{q^2}{c}\right) P_S^2 + 5\gamma P_S^4\right), \quad (6)$$
$$Q^* = 4\frac{q^2 P_S^2}{c\alpha_S}, \quad M^* = \frac{cM}{2\rho f}, \quad \mu^* = \frac{c\mu}{2g\rho},$$

where a is the lattice constant. Then the dispersion law (4) in terms of the dimensionless variables (6) reads [31, 32]

$$\omega_{\mathrm{O,A}}^{*2}(k^*) = \frac{\left(C^* \pm \sqrt{D^*}\right)}{2\left(\mu^* - 2F^*M^{*2}\right)}, \quad (7)$$

where

$$C^* = 2\alpha_v^* + \left(\frac{k^*}{a^*}\right)^2 (2\mu^* + 1 - 4F^*M^*) + \left(\frac{k^*}{a^*}\right)^4 \mu^*,$$
$$D^* = C^{*2} + 4\left(\frac{k^*}{a^*}\right)^2 \left(2F^*M^{*2} - \mu^*\right) \times$$
$$\times \left(4\alpha_v^*(1 - Q^*) + 2\left(\frac{k^*}{a^*}\right)^2 (1 - F^* + \alpha_v^*) + \left(\frac{k^*}{a^*}\right)^4\right).$$

The dimensionless critical value of flexocoupling constant, at which the frequency of the acoustic mode is zeroed, equals

$$F_{\mathrm{cr}}^*(\alpha_v^*) = 1 + \alpha_v^* + 2\sqrt{\alpha_v^*(1 - Q^*)}.$$

The corresponding critical value for the dimensionless wave vector is

$$k_{\mathrm{cr}}^* = a^*\sqrt{(F^* - 1 - \alpha_v^*)}.$$

At $F^* > F_{\mathrm{cr}}^*$, there are two wave-vector values corresponding to the zero frequency of the A-mode,

$$k_{1,2}^{*\mathrm{cr}} =$$
$$= a^*\sqrt{\left(F^* - 1 - \alpha_v^* \pm \sqrt{(F^* - 1 - \alpha_v^*)^2 - 4\alpha_v^*(1 - Q^*)}\right)}.$$

The mode frequency differs from zero in the intervals $0 < k < k_1^{*\mathrm{cr}}$ and $k > k_2^{*\mathrm{cr}}$. We may expect that a spatially modulated phase can appear in the "gap" $k_1^{*\mathrm{cr}} \le k \le k_2^{*\mathrm{cr}}$ [32].

Figure 1, *a* illustrates the influence of the flexocoupling constant F^* on the behavior of the O- and A-modes. The O-mode frequency decreases very slightly, as F^* increases from 0 to 10 (see dotted and solid O-curves). The A-mode frequency decreases noticeably, and there appears a bend in the $\omega^*(k^*)$ dependence, as F^* increases from 0 to 3. The critical value $F_{\mathrm{cr}}^*(\alpha_v^*) = 3.414$ for the chosen parameters, and the corresponding curve for the A-mode touches the k-axis at the point $k_{\mathrm{cr}}^* = 0.12$. For $F^* = 5$, the "softening" of the A-mode takes place at the points $k_1^{*\mathrm{cr}} = 0.06$ and $k_2^{*\mathrm{cr}} \approx 0.2$. A spatially modulated phase exists in the gap $k_1^{*\mathrm{cr}} \le k \le k_2^{*\mathrm{cr}}$. For the O-mode, solid curves depict dispersion relations for various values of flexoconstant F^* (from 1 to 10 with a step of 1), and the dotted curve illustrates the dispersion relation at $F^* = 0$. For the A-mode, the flexoconstant F^* acquires values from the set {1.25, 1.5, 1.75, 2.0, 2.25, 2.5, 2.75, 3.0, 3.414, 5.0} (solid curves) and 0 (dotted curve). In the case of O-mode, the growth of the flexoconstant from 0 to 10 leads to a significant tightening of spectral lines in a vicinity of the flexoconstant $F^* = 10$. In the case of A-mode, the situation is inverse: it is the decrease of the flexoconstant that leads to the tightening of spectral lines. At a certain critical flexoconstant value, the spectral line has a break point at k_{cr}, and a further growth of the flexoconstant generates a gap in the dispersion relation.

The dependences of the dimensionless frequency ω^* on the dimensionless flexoconstant F^* for various wave vectors k^*'s are shown in Fig. 1, *b* for the O-mode and in Fig. 1, *c* for the A-mode. At small k^*'s, the frequency of the O-mode depends very weakly on F^*, but the dependence becomes a bit stronger for larger values of the wave vector. On the contrary, in the case of A-mode, the phonon frequency strongly

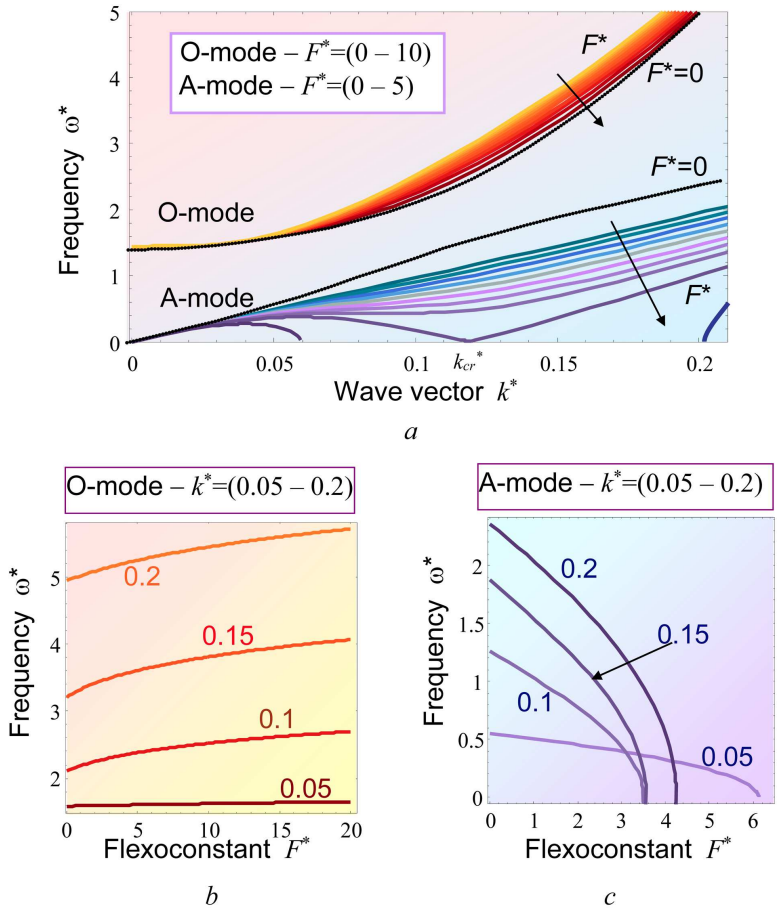

Fig. 1. Dimensionless dependences of the phonon frequency ω^* on the wave vector $k^* = \frac{ak}{\pi}$ for various flexoconstant values $F^* = 1, 2, 3, 4, 5, 6, 7, 8, 9, 10$ (solid curves), and $F^* = 0$ (dotted curve) for the O-mode; and $F^* = 1.25, 1.5, 1.75, 2.0, 2.25, 2.5, 2.75, 3.0, 3.414, 5.0$ (solid curves), and $F^* = 0$ (dotted curve) for the A-mode (a). Dependences of the frequency ω^* on the flexoconstant F^* in the O- (b) and A-modes (c) for various $k^* = 0.05, 0.1, 0.15$, and 0.2 (indicated near the plots). The calculation parameters are $\alpha_v^* = 1$, $Q^* = 0.5$, $M^* = 0.05$, $a^* = 0.1$, and $\mu^* = 1$ (b, c)

and nontrivially depends on the wave vector k^*, including all aforementioned critical points. The strong growth of the frequency together with the wave vector is responsible for a specific behavior of spectral lines in the O-mode. From this figure, it is evident that, for the O-mode, the frequency weakly depends on the flexoconstant, especially in the region of small wave vectors, where the frequency has an almost constant value. In the A-mode, the frequency ω^* also weakly depends on the flexoconstant F^* in the region of small wave vectors, but, as the wave vector values grow, this dependence becomes stronger. In all cases, there is an upper limit for the flexoconstant F^* that determines the condition for a spatially modulated phase to appear. For the parameter values indicated in the figure caption, this critical constant ranges within the limits from about 3 to about 6.

The contour plots in Fig. 2 illustrate the frequency dependences of the wave vector k^* and the flexoconstant F^* in the A- and O-modes. One can observe a weak monotonic dependence on F^* for the O-mode (Fig. 2, b) and a nontrivial dependence with a shifted maximum for the A-mode (Fig. 2, a). In general, the frequency increases with the wave vector in both panels. In panel b, the contours are almost perpendicular to the F^*-axis in vicinities of small wave vectors, which means a strong dependence of the angular frequency on the flexoconstant F^* and,

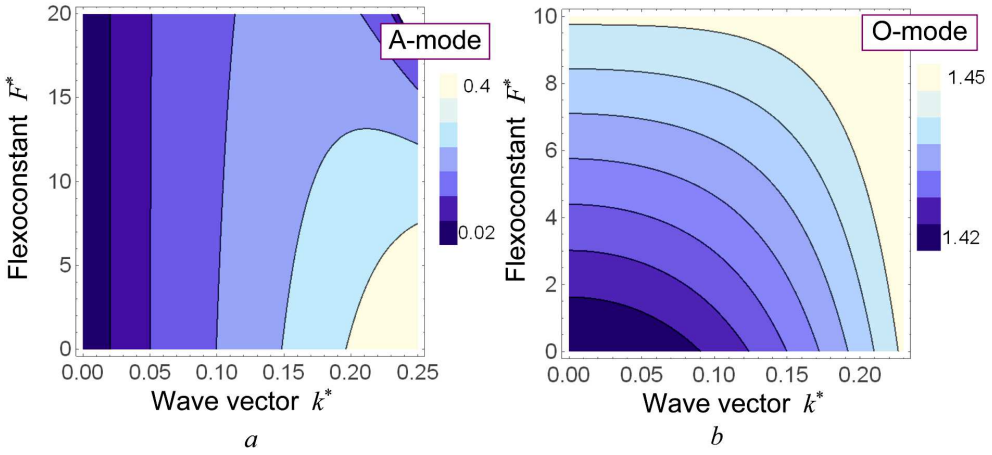

Fig. 2. Contour plots of the dimensionless phonon frequency ω^* on the dimensionless wave vector k^* and flexoconstant F^* for the A- (a) and O-modes (b). The calculation parameters are the same as in Fig. 1

simultaneously, a weak dependence on the wave vector k^*. This behavior correlates well with the optic dispersion relation presented in Fig. 1, a, where the frequency is almost independent of the wave vector at small k^*-values. The $\omega^*(F^*)$ dependence of a certain kind can be observed for the O-mode at relatively large values of wave vector (Fig. 2, b): it significantly softens at large wave vectors. For the A-mode, a strong dependence of the frequency on the wave vector takes place, but the frequency weakly depends on the flexoconstant in the region of small wave vectors.

4. Discussion and Conclusions

Using the one-component approximation, which is valid for some ferroelectrics, the analytical expressions for the frequency dispersion law $\omega(k)$ and their dependence on the flexoelectric coupling constant f have been analyzed for the soft TA and TO phonon modes in uniaxial ferroelectrics. For the O-mode, the growth of the flexoconstant leads to a significant tightening of spectral lines in a vicinity of the flexoconstant. The frequency of the O-mode weakly depends on the flexoconstant in the region of small wave vectors. A critical dependence of the TA mode on the flexocoupling constant has been revealed. Unlike the O-mode, it is a reduction of the flexoconstant that leads to the tightening of spectral lines in the A-mode. At a certain critical flexoconstant value, the spectral line has a break point at $k_{\rm cr}$, and the further growth of the flexoconstant leads to the appearance of a gap in the dispersion relation.

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ЗАЛЕЖНІСТЬ СПЕКТРІВ М'ЯКИХ ФОНОНІВ ВІД ФЛЕКСОЕЛЕКТРИЧНОГО ЗВ'ЯЗКУ В СЕГНЕТОЕЛЕКТРИКАХ

Р е з ю м е

В межах теорії Ландау–Гінзбург–Девоншира (LGD) ми аналізуємо аналітичні вирази для частотної дисперсії м'яких поперечних акустичних (TA) та оптичних (TO) фононних мод залежно від величини константи флексоелектричного зв'язку у одновісних сегнетоелектриках та виявили критичну поведінку TA моди в залежності від константи флексоелектричного зв'язку.