Phase-transition-induced nonlinear acoustic phenomena in ferroelectrics

V. I. Kozub and A. K. Tagantsev

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The propagation of an intense acoustic wave at a phase transition in a ferroelectric is investigated theoretically. Attention is directed mainly to the case when the deformation amplitude in the wave is sufficient to induce the phase transition. The shape of the wave contour is obtained for the case of weak attenuation. The acoustic nonlinearity in a ferroelectric without the piezoeffect in the paraphase due to polarization viscosity is also analyzed. It is shown that for a ferroelectric with the piezoeffect in the paraphase the development of a shock wave should be possible, and the time needed for it to form is estimated. Order of magnitude and numerical estimates of criteria for these phenomena to be observed are presented.

As is well known, an investigation of the propagation of sound in ferroelectrics provides a convenient tool to use in the study of their various physical properties. This is because the phase transition temperature, where the most interesting behavior is observed, is itself (generally speaking) a function of stress. Thus, the question arises: in the case of sufficiently high-intensity acoustic (i.e., stress) waves, to what extent can this stress dependence be neglected? In point of fact, one might expect that if the stress amplitude is high enough, the phase transition could occur in those portions of the wave profile where the stress exceeds a certain critical value. Provided that the elastic moduli of the different phases are noticeably different (Ref. 1), the equations of elasticity for such a system are significantly nonlinear. The basic goal of this paper will be to analyze the effects of this type of nonlinearity, i.e., one with a marked threshold character, on the propagation of sound.

In this paper, we will assume everywhere that the acoustic frequency is so low that the time it takes the phase transition to occur can be neglected in comparison with the wave period, i.e., that the transition occurs instantaneously. This in turn implies that the elastic modulus is a function of the local value of the deformation. Now, a deformation dependence of this kind can give rise to shock wave formation (see Ref. 2), since the possibility of formation of such waves is determined by the relationship between nonlinearity and dispersion and dissipation. As was pointed out above, in the case under investigation here an increase in stress above a certain critical value produces a significant change in the elastic moduli, so that we have a system with very strong nonlinearity. We further observe that if the amplitude of a sound wave entering the ferroelectric crystal exceeds a certain value, it cannot propagate in the crystal in the form of a sinusoidal wave. In practice, shock fronts form immediately at the surface of a crystal, which guarantees the subsequent evolution of nonsinusoidal waves. It is interesting that this evolution results not in complete attenuation (see e.g., Ref. 2) but in the emergence of a certain nonsinusoidal wave profile whose subsequent propagation is determined by conventional mechanisms.

We remark that for the particular systems we have investigated, the wave propagation problem is equivalent to that of propagation of finite-amplitude sound waves in a gas-liquid system near a critical point. That problem was investigated by Pokrovskii and Kamenskii (Ref. 3). We have found that our results differ both qualitatively and quantatively from those obtained in Ref. 3; in our opinion, the differences are connected with a certain inconsistency in the way the original equations in Ref. 3 were formulated.

Provided that the nonlinearity described above is due only to the change in elastic moduli at the phase transition, it can make itself felt even in the systems in which the equations of elasticity theory both above and below the transition are themselves linear. It is well known that precisely this sort of behavior is characteristic of ferroelectrics which are nonpiezoelectric in the paraelectric phase. For this type of ferroelectric, if no account is taken of fluctuations (Ref. 4) and polarization viscosity, there are no nonlinear terms in the equations of elasticity theory. If, however, the ferroelectric properties give rise to such nonlinearities even in the absence of the phase transition (i.e., taking place in the wave field), then the latter guarantee a change in the form of the wave (growth of harmonics and subsequent formation of shock fronts) even for sonic intensities below the critical value. Such behavior is naturally expected for ferroelectrics which are piezoelectric in the paraelectric phase. However, we also want to point out that even in the absence of piezoelectricity in the paraelectric phase, taking into account the polarization viscosity can still lead to the appearance of nonlinear terms in the equations of elasticity when this viscosity is treated within the framework of Landau's theory (that is, without taking fluctuations into account). It is evident that this type of nonlinearity can manifest itself only below the transition point. We will investigate the growth of harmonics in this situation; for the case of ferroelectricity accompanied by piezoelectricity in the paraelectric phase, we will only give estimates of the time it takes shock fronts to form.

1. THRESHOLD NONLINEARITY FOR FERROELECTRICS WHICH ARE NONPIEZOELECTRIC IN THE PARAELECTRIC PHASE

For simplicity we will investigate the case of acoustic propagation in a single-phase ferroelectric; we will be assuming that the direction of propagation is selected in such a way

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that the sonic wave is not accompanied by an electric field (in the presence of an electric field, as will be shown further on, the nonlinear effects are suppressed). Sound propagation in such a situation is described by the following system of equations:

$$\alpha P+\beta_T P^3+2Q_T P\frac{\partial u}{\partial x}=-\mu\dot{P}, \tag{1a}$$

$$\frac{\partial}{\partial x}\left(\Lambda_T\frac{\partial u}{\partial x}+Q_T P^2-\Lambda_T aT\right)=\rho\ddot{u}, \tag{1b}$$

$$\dot{S}=-\frac{\partial\alpha}{\partial T}P\dot{P}+\Lambda_T a\frac{\partial \dot{u}}{\partial x}+\frac{c}{T}\dot{T}=0. \tag{1c}$$

Here, u is the elastic displacement associated with the sound wave, α and β_T are coefficients for the second-order and fourth-order terms in an expansion of the free energy in powers of the polarization P; Q_T, Λ_T, a are the corresponding components of the electrostrictive and elastic-modulus tensors, along with the tensor coefficients of thermal expansion (we remark that the parameters β_T, Q_T and Λ_T are defined with respect to isothermal conditions; in addition, Λ_T is for the paraelectric phases); μ is the polarization vioscosity and c the heat capacity at constant polarization.

Equation (1a), which is the equation of motion for the polarization, is obtained by neglecting spatial dispersion. This corresponds to fulfillment of the condition

$$\alpha\gg\varkappa\omega^2/w^2, \tag{2}$$

where ω and w are the frequency and sound velocity and $\varkappa$ is the coefficient of the gradient term in the free energy expansion. Equation (1c)—the equation of thermal balance—is derived by neglecting the non-adiabatic character of the sound wave. Later on we will neglect the polarization viscosity in Eq. (1a), assuming that the frequency of sound is bounded from above by the following inequality:

$$\omega\ll\alpha/\mu. \tag{3}$$

The physics of this restriction implies in particular that the time it takes to establish the polarization at the phase transition is short compared to the wave period.

To begin with, we eliminate the last of Eqs. (1a)–(1c). This gives rise to a renormalization of the parameters β, Q and Λ corresponding to a transition from isothermal to adiabatic values:

$$\beta=\beta_T\left[1+\frac{1}{2}\left(\frac{\partial\alpha}{\partial T}\right)^2\frac{T}{c\beta_T}\right],$$
$$Q=Q_T\left[1-\frac{1}{2}\frac{\partial\alpha}{\partial T}\frac{T}{c}\frac{\Lambda_T a}{Q_T}\right], \tag{4}$$
$$\Lambda=\Lambda_T\left[1+\frac{aT}{c}\Lambda_T a\right].$$

As a result, we are led to the equations

$$P\left(\alpha+\beta P^2+2Q\frac{\partial u}{\partial x}\right)=0, \tag{5a}$$

$$\frac{\partial}{\partial x}\left(\Lambda\frac{\partial u}{\partial x}+QP^2\right)=\rho\ddot{u}. \tag{5b}$$

Clearly, the first of equations (5) can have different steady-state solutions with different functional dependences on the magnitude of the deformation $U=\partial u/\partial x$, namely[1)]

$$P^2=-(\alpha+2QU)/\beta,\quad 2QU<-\alpha,$$
$$P=0,\qquad 2QU>-\alpha.$$

Hence, taking into account (5), it is not difficult to see that if $|U|_{\max}<|U_c|$ (where $U_c=-\alpha/2Q$), then the system is described by the usual equations of elasticity. If, however, $|U|_{\max}>|U_c|$, then the nonlinear regime results; in this case, the nonlinearity is due entirely to the action of the phase transition induced by the wave. Therefore, the second of Eqs. (5) can be cast in the form

$$\frac{\partial}{\partial x}[\Lambda(U)U]=\rho\ddot{u}, \tag{6}$$

where

$$\Lambda(U)=\begin{cases}\Lambda, & U<U_c\\ \Lambda-2Q^2/\beta, & U>U_c\end{cases}$$

(for definiteness we assume that $Q<0$, $\alpha>0$). As is well known from the theory of nonlinear waves, equations of this kind admit solutions in the form of simple waves (see, e.g., Ref. 5):

$$U=\Phi[x-w(U)t], \tag{7a}$$

$$w^2(U)=[\Lambda(U)+\Lambda'(U)U]/\rho, \tag{7b}$$

where Φ is an arbitrary function determined by the initial conditions. We will investigate the evolution of a sinusoidal wave

$$U=U_0\cos(\omega t-kx),$$

introduced into a crystal. From (7) it is clear that if $U_0<|U_c|$ we have the case of a pure sinusoidal wave which does not change its form during propagation. We note that within the framework of our approximation we neglect linear damping of the wave caused by polarization viscosity. This neglect is permissible if we are analyzing the behavior of waves whose distance from the crystal surface is smaller than the absorption depth

$$l=|\alpha|/\Delta wk^2\mu,\quad \Delta w=Q^2/\beta\rho w_0,$$
$$w_0^2=\Lambda/\rho.$$

If, however, the wave amplitude is large enough, i.e., $U_0>|U_c|$, the velocities of selected portions of the wave contour begin to depend on the local deformation (Fig. 1). The

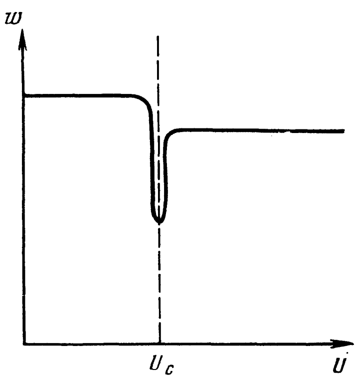

FIG. 1. Velocity dependence of a portion of the wave contour $w(U)$ versus local deformation U for $Q<0$, $\alpha>0$.

spike in Fig. 1 arises from differentiating the jump in (6). The presence of a delta-function singularity in the velocity profile implies that a portion of the wave contour with infinite derivative, i.e., a shock front, forms practically at once. We note that from a physical point of view the jump in elastic modulus leads to the appearance of a jump in elastic stress at the same point, i.e., the force density at this point also diverges. This, in its turn, causes the local velocity of surrounding points to diverge, and in the final analysis leads to the generation of the shock front. In the real situation, the jump in elastic modulus in (6) is washed out. The physical cause of this washing out can be traced to the fact that the real phase transition takes place through creation of domains of the new phase whose size scale is on the order of the correlation length. The smallness of this distance compared to a wavelength ensures that inequality (2) is satisified by a large margin. As a result, within the critical region a "medium" is generated with intermediate values of the parameters. As regards the possible influence of viscosity in the critical region, we notice that the corresponding time scale, by virtue of inequality (3), is much smaller than the wave period, so that its inclusion cannot significantly change the qualitative picture we describe here. As a result, the singularity under discussion is washed out, and thus the shock front acquires a finite width. Inequality (3) ensures the narrowness of the front compared to a wavelength.[2)]

As was pointed out in the introduction, Kamenskii and Pokrovskii[3] have investigated a problem which is analogous in its mathematical formulation to this one. They studied the propagation of sound in a liquid-gas system near the critical point. The authors of Ref. 3 did not undertake a systematic derivation of the nonlinear equations of elaticity [of type (6)], but at once took advantage of a solution in the form of a simple wave (described by a Riemann invariant), whereupon they identified the velocity of the simple wave with the velocity of sound in one of the two media. Such an identification (without taking into account the nonlinear properties of the medium) is justified only for domains which correspond to a specific phase; it cannot be justified for the interphase boundary. In our case, this would correspond to taking into account only the first term in (7b). It is easy to see, however, that the presence of the second term is of major importance, since it is linked with the emergence of a secondary shock front which was not considered in Ref. 3. Later on it will be clear that this secondary front plays an extremely important role in the later evolution of the wave.

It is evident that the solution (7) is not useful in the shock-front region since it leads to a multiple-valued dependence on the coordinates (see Fig. 2b). In reality the wave evolution is determined by the motion of the shock front. We can obtain the following relation (see Ref. 2) for the front velocity v:

$$v^2=\{\sigma\}/\rho\{U\}, \tag{8}$$

where $\{\sigma\}$ and $\{U\}$ are the jumps in elastic stress $\sigma = \Lambda(U)U$ and deformation at the discontinuity. In order to determine $\{\sigma\}$ and $\{U\}$ we also need an equation describing the motion of points on the profile in the neighborhood of the discontinuity, where the solution (7) can be used. Differentiating (7), we obtain

$$\dot{U}=[v-w(U)]\left[\frac{1}{\Phi'}+\frac{\partial w}{\partial U}t\right]^{-1}. \tag{9}$$

The system (8) and (9) allows us to describe the wave evolution. It is apparent that in the situation we are investigating this evolution will continue as long as some portion of the wave profile is above the linearity boundary $U = U_c$ (Fig. 2). At the instant that this portion disappears (corresponding to the "collision" of the two shock fronts) the steady-state form of the wave profile becomes fixed and (neglecting linear dissipation) will evolve no further. This instant satisfies $\Delta t \sim (w_0/\Delta w)(\pi/\omega)$. It is easy to see that if inequality (3) is fulfilled, then the distance over which this evolution occurs is shorter than the linear attenuation length.

This profile is shown in Fig. 2(e); the phases where the "kinks" shown in that figure appear are determined from the following equation

$$\sin\varphi_1=\sin\varphi_0-\cos\varphi_0(2\pi+\varphi_0-\varphi_1) \tag{10}$$

[the derivation of (10) and a detailed analysis of the evolution of the wave contour are given in the Appendix). From (10) it is easy to see that the steady-state contour does not include any zero-order harmonic, i.e., "DC level" (we note that the solution obtained in Ref. 3 will in general contain some DC level].

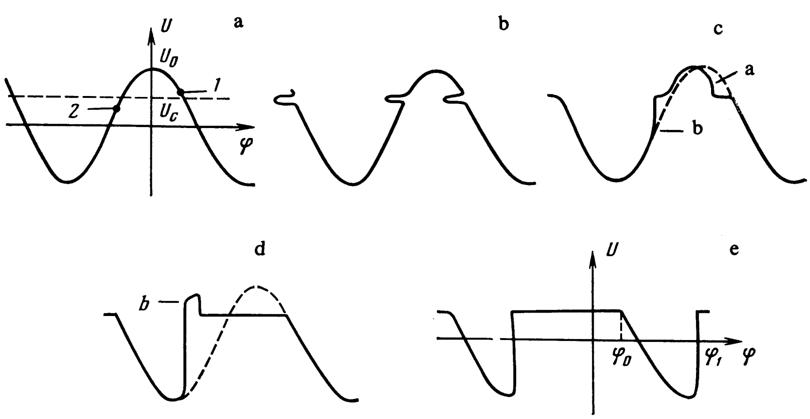

FIG. 2. (a), (c), (d) and (e): steps in the evolution of the wave profile for the case of a threshold nonlinearity. For $\varphi_0 < \tan^{-1}(\pi/2)$ the profile evolves by omitting phase (d). (b) shows the multiple-valued solution (7) for very short times.

2. HIGHER HARMONICS OF SOUND WAVES FOR SONIC INTENSITIES LOWER THAN CRITICAL IN FERROELECTRICS WHICH ARE NON-PIEZO-ELECTRIC IN THE PARAELECTRIC PHASE

In Sec. 1, we investigated the appearance of strong acoustic nonlinearity, caused by the presence of a phase transition, in a wave with sufficiently high sonic intensity. We now want to turn our attention to the case of a medium which exhibits nonlinearity even for intensities below critical, in particular a medium whose nonlinearity is connected with the presence of polarization viscosity. Provided that the paraelectric phase for $U_0 < |U_c|$ is characterized by $P = 0$, this nonlinearity occurs only in the ferroelectric ($P \neq 0$) phase. If we take viscosity into account, in place of (5a) we have

$$\alpha+\beta P^2+2QU=-\frac{\mu}{2}\frac{\partial}{\partial t}(\ln P^2). \tag{11}$$

Iterating (11) in the magnitude of the polarization viscosity, we obtain a correction to the term P^2 (which appears in the elasticity equation):

$$\delta(P^2)=-\frac{\mu}{2\beta}\frac{2Q\dot{U}}{\alpha+2QU}=\frac{\mu}{2\beta}\frac{\dot{U}}{U_c-U}. \tag{12}$$

Substituting (12) into (5b), we obtain

$$\rho\ddot{u}=\frac{\partial}{\partial x}\left[\left(\Lambda-\frac{2Q^2}{\beta}\right)U+\frac{Q\mu}{2\beta}\frac{\dot{U}}{U_c-U}\right]. \tag{13}$$

The range of applicability of (13) is bounded in the low-polarization-viscosity case only by the condition $|U| < |U_c|$. To simplify further analysis we assume, however, that $|U| \ll |U_c|$. In this situation we can iterate (13) in powers of U/U_c and investigate the propagation of the corresponding acoustic harmonics. We will confine ourselves to an analysis of the behavior of the second harmonic. Following standard procedures, we assume $U_2(x, t) = U_{02}(x) \times \exp[2i(\omega t - kx)]$; substituting this expression (along with the deformation which corresponds to the fundamental harmonic) in (13), we obtain for the slowly-varying amplitude U_{02} the following equation:

$$\frac{\partial U_{02}}{\partial x}+2\frac{\Delta w}{w}4k(\omega\tau)U_{02}=-2\frac{\Delta w}{w}k(\omega\tau)\frac{U_{01}^2}{U_c}, \tag{14}$$

$$w^2=(\Lambda-2Q^2/\beta)/\rho,\quad \tau=-\mu/2\alpha.$$

The boundary condition on (14) is the requirement $U_{02}(x=0)=0$. We remark that, strictly speaking, the amplitude of the fundamental harmonic U_{01} itself depends on x because of attenuation which occurs over a distance $l \sim w/2\Delta wk\omega\tau$. We will assume, however, that the sample length satisfies $L \lesssim l$ and neglect this dependence. As a result, we obtain for U_{02} at $x = L$:

$$U_{02}\simeq\frac{1}{4}\frac{(U_{01})^2}{U_c}\left[1-\exp\left(-\frac{4L}{l}\right)\right]. \tag{15}$$

As regards higher-frequency harmonics, the correct iteration procedure for (13) is quite tedious. However, it is not hard to see that

$$U_{0n}\sim\left(\frac{U_{01}}{U}\right)^{n-1}U_{01}\left[1-\exp\left(-\frac{n^2L}{l}\right)\right]. \tag{16}$$

It is clear[3] that U_{0n} diverges as $T \to T_c$.

We will now trace the evolution of the higher-harmonic signal at temperatures in the vicinity of T_c, taking into account both the "nonadiabatic" nonlinearity we have investigated and the formation of the shock wave. We remark that the characteristic behavior of the wave profile can be qualitatively different for different values of x. For the present analysis there exist three length scales which characterize the wave evolution: the temperature-dependent attenuation length of the fundamental harmonic $l(T)$, the "critical" value of this length $l(T_t)$ where the temperature T_t satisfies the condition $2QU_{01} = -\alpha(T_t)$, and finally the distance the wave propagates before it establishes a steady-state profile as a result of the process of shock wave evolution $\mathscr{L} = w/\Delta wk$. If $\mathscr{L} > l(T_t)$, then the shock wave will form, and only the viscosity mechanism can give rise to nonlinear effects. The temperature dependence of the higher harmonic amplitudes is different for temperature regions where the relations $x < l(T)$ and $x \ll l(T)$ are satisfied: for $x > l(T)$,

$$U_{0n} \propto (T_c-T)^{1-n}\exp(-nx/l);$$

while for $x \ll l(T)$,

$$U_{0n} \propto (T_c-T)^{-n}.$$

In the intermediate region, it is clear that a maximum will be reached whose amplitude is determined by the estimates (15) and (16), and depends on temperature as $(T_c - T)^{1-n}$.

If, however, $\mathscr{L} < l(T_t)$, then for $T > T_t$ a shock wave forms in the system. However, at a distance $x > l(T_t)$, the overall character of the temperature dependence is in practice no different from that investigated above, since on the one hand for points here $T > T_t$ the harmonic amplitudes experience a rapid fall because of the strong attenuation of the fundamental wave, while on the other hand it is easy to convince oneself that for such conditions the order of magnitude of the harmonics which arise in the course of evolution of the shock wave is the same as that connected with the "viscosity" mechanism for $U \sim U_c$. The most interesting behavior is observed in the region $x < l(T_t)$. In this region, for $T < T_t$ a growth in harmonic amplitude is observed, which varies at $(T_c - T)^{-n}$, caused by the "viscosity" nonlinearity. For $T > T_t$ a shock wave is generated in the system, and from that point on (for $T_c - T \sim T_c - T_t$) the harmonics are associated basically with the steady-state profile. In contrast to the mechanism of "viscous" nonlinearity (for which $U_{02} \sim U_{01}\omega\tau\Delta w/w$, the amplitude U_{02} in the steady-state profile is $\gtrsim U_{01}$, so that in a fairly small temperature interval around T_c (where $T_c - T \sim T_c - T_t$), the harmonic amplitude grows by a factor of $w/\Delta w\omega\tau$. Subsequently, however, for a further approach to T_c there is a substantial reduction in the amplitude of the steady-state contour (which varies according to the relation $U_c(T) \propto T_c - T$) and so the amplitude of the corresponding harmonics falls as $T_c - T$. Finally, in the intermediate vicinity of T_c for $l(T) \sim x$ this law of decrease changes over to exponential.

3. FERROELECTRICS WITH PIEZOELECTRICITY IN THE PARAELECTRIC PHASE

As is well know, in crystals of this sort there is a linear relationship between deformation and order parameter both

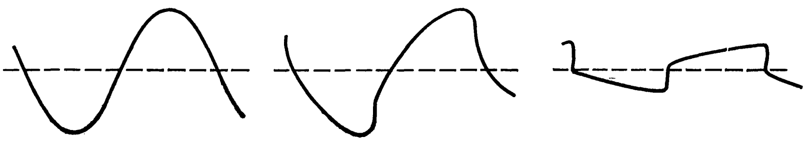

FIG. 3. Stages in the evolution of the wave profile for linear coupling between the wave deformation and the order parameter.

above and below the transition point; in addition, the dynamic behavior of the order parameter is distinguished by substantial nonlinearity. By virtue of the circumstances mentioned above, there arises a nonlinear contribution to the effective elastic modulus of the medium, the equation for which can be written in the form[4)] (for definiteness we pick the paraelectric phases):

$$\Lambda(U)=\Lambda_0-d^2/\alpha+\beta(d/\alpha)^4U^2. \tag{17}$$

In order to write (17) we include a term in the free energy expansion bilinear in the deformation and polarization; d is the coefficient of this term. Expression (17) is valid for moderately large values of the deformation U such that the following inequality is satisfied:

$$U^2(\beta\Lambda_0^3/d^4)\ll 1. \tag{18}$$

We remark that the nonlinear term in (17) is entirely due to the dielectric nonlinearity; the term which would be present if both piezoelectricity and electrostriction were taken into account is absent by virtue of symmetry considerations. The point is that piezoelectricity and electrostriction are connected with different components of the deformation tensor.

In order to analyze the wave evolution, it is necessary to substitute (17) into the elasticity equation (6). The corresponding nonlinear equation was analyzed in Ref. 2 for the specific case of a quadratic nonlinearity. As shown in Ref. 2, for any value of the deformation potential a nonlinearity of this form leads to the formation of a shock front at a time

$$\Delta t=\frac{\pi}{\omega}\frac{\Lambda_0-d^2/\alpha}{\beta(d/\alpha)^4U_0^2}. \tag{19}$$

The subsequent wave evolution sketched in Fig. (3) in fact results in damping which is nonexponential in character (satisfying a $1/t$ law). In order to observe the behavior under consideration here, it is necessary that the time it takes the shock front to form, i.e., (19), be smaller than the linear attenuation time

$$\Delta t_a=\frac{\pi}{\omega}\frac{\Lambda_0-d^2/\alpha}{d^2\omega\mu/\alpha^2}.$$

This condition leads to the inequality

$$U_0^2(\beta\Lambda_0^3/d^4)\gg\omega\mu/\alpha. \tag{20}$$

Compatibility of inequalities (20) and (18) is assured if the limit on acoustic frequency (3) is fulfilled.

4. CONCLUSIONS

Let us go through an analysis of the possibility of observing the phenomena discussed here experimentally. To begin with, we consider some estimates relating to the threshold nonlinearity. First of all, a limit on sonic frequency can be derived from inequality (3). Applied to the ferroelectric TGS, which has a phase transition of the second kind, this inequality corresponds to the restriction

$$\omega\ll 1/3\times10^4\,(T_c-T)\ ^\circ\mathrm{K\,sec^{-1}}$$

(here and further on the material parameters are taken from Refs. 1, 8 and 9). On the other hand, as we have seen, the ferroelectric phase transition can occur under the influence of the sound wave if

$$|T-T_c|\lesssim|T_t-T_c|=2Q(\partial\alpha/\partial T)^{-1}(2J/w\Lambda)^{1/2}$$

(where J is the wave intensity). For TGS this implies that $|T-T_c|\lesssim 10^{-2}\,J^{1/2}$ (V/cm²). Taking into account the above-mentioned relations, wave evolution in TGS can be observed if $J\sim 10$ V/cm², $T-T_c\sim .03$ °K and $\omega/2\pi\lesssim 50$ MHz.

As regards harmonic formation deriving from the nonadiabatic (i.e., viscous) nonlinearity, it is natural to compare the corresponding contribution with that of the fluctuation nonlinearity. It is not difficult to show that for uniaxial ferroelectrics (Ref. 4) the order of magnitude of this contribution is given by the expression:

$$[(T/\varkappa^{3/2})\beta(\alpha/\mu\omega)]^{-1}.$$

Hence we find that the nonadiabatic nonlinearity is more important if $\omega\tau>T/Mw^2$, where M is the ionic mass. The inequality is easily fulfilled in conjunction with the condition $\omega\tau<1$.

APPENDIX

We will assume that the jump in sound velocity at the phase transition is relatively small ($\Delta w\ll w_0$). In that case, to lowest order in $\Delta w/w_0$, from (8) and (9) we obtain

$$v=w_0[1+\{\tilde\sigma\}/2\rho\{U\}], \tag{A1}$$

where $\tilde\sigma$ is the nonlinear part of the elastic stress:

$$\tilde\sigma=-2\Lambda\frac{\Delta w}{w}U\theta(U-U_c).$$

In turn, (7b) gives

$$w=w_0\left[1+\frac{1}{2\rho}\frac{\partial\tilde\sigma}{\partial U}\right]. \tag{A2}$$

In order to investigate the evolution of the shock wave, we will assume that the θ-function in (A1) is "smeared out" on a scale $\Gamma\ll U$. As follows from (9), the points where the front appears (and converts the derivative $\partial u/\partial x$ to infinity)—i.e., points 1 and 2 in Fig. 2a—correspond to maximal values of $|\partial w/\partial U|$; the inequality $\partial w/\partial U>0$ corresponds to the leading edge of the shock, while $\partial w/\partial U<0$ is the trailing edge. It is not difficult to convince oneself that, starting from Eqs. (9), (A1) and (A2), the evolution of the fronts in the initial instants will be determined by the motion of their edges next to the line $U=U_c$; this stage is concluded when the velocities of these edges, described by (9), fall to the values corresponding to Eq. (A1). This takes place during a time much smaller than the characteristic time for evolution

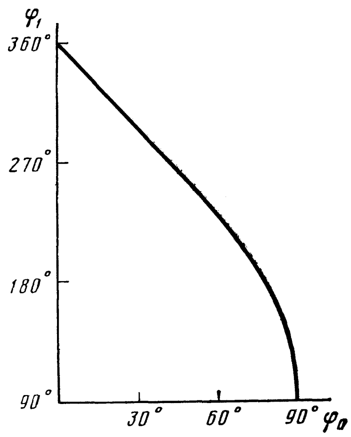

FIG. 4. Solution to Eq. (10) for the "kink" phase angles shown in Fig. 2(e).

of the wave; the smallness of this time is ensured by the smallness of the scale Γ of "smearing-out" of the θ-function compared to the characteristic scale of the change in U. At the instant in question one can assume that within a single-phase domain the velocity w is constant, while the jumps corresponding to the fronts equal respectively $-2U_+\Delta w/w_0$ for the leading-edge front (where U_+ is the value of the deformation at point a in Fig. 2c) and $-2U_c\Delta w/w_0$ for the trailing edge. Thus, for the leading edge velocity we have the expression

$$v_1=w_0\left[1-\frac{\Delta w}{w_0}\frac{U_+}{U_+-U_c}\right], \tag{A3}$$

and for the trailing edge velocity the expression

$$v_2=w_0\left[1-\frac{\Delta w}{w}\frac{U_c}{U_c-U_-}\right] \tag{A4}$$

(where U_- is the value of the deformation at point b in Fig. 2c).

Taking into account what was said above, we can obtain the following equations

$$\dot{U}_+=-\Delta w\frac{U_c}{U_+-U_c}\Phi', \tag{A5}$$

$$\dot{U}_-=-\Delta w\frac{U_c}{U_c-U_-}\Phi'. \tag{A6}$$

Transforming to polar coordinates, i.e., rewriting $U_+=U_0\times\cos\varphi_+$, $U_-=U_0\cos\varphi_-$, $U_c=U_0\cos\varphi_0$, and taking into account that $\Phi'=k\partial U/\partial\varphi$ along with the initial conditions, $\varphi_-|_{t=0}=-\varphi_0$, $\varphi_+|_{t=0}=\varphi_0$, we substitute (A3) and (A4) into (A5) and (A6), and find an equation for the motion of the front

$$\Omega t=\varphi_+-\varphi_0+\frac{\sin\varphi_0-\sin\varphi_+}{\cos\varphi_0}, \tag{A7}$$

$$\Omega t=-\varphi_--\varphi_0+\frac{\sin\varphi_0+\sin\varphi_-}{\cos\varphi_0}, \tag{A8}$$

where $\Omega=\Delta w\cdot k$. If $\varphi_0<\mathrm{arctg}(\pi/2)$, then the evolution is completed by "truncation" of the peaks by the leading edge; the time for this truncation is determined by the condition $\varphi_+=-\varphi_0$. The position of the trailing edge in the final instant of evolution, taking into account (A8), is given by the equation

$$\sin\varphi_-=\sin\varphi_0-\cos\varphi_0(\varphi_0-\varphi_-). \tag{A9}$$

Equation (A9), by means of the replacement $\varphi_-=\varphi_1-2\pi$ leads to (10), which we were required to show.

If, however, $\varphi_0>\mathrm{arctg}(\pi/2)$, then the evolution is completed by "collapse" of the fronts. In this case, in place of (A6) we have a system of equations for the values of the deformation at the front boundaries.

$$\dot{U}_-=-\Delta w\frac{U_{+-}}{U_{+-}-U_-}\Phi',\qquad \dot{U}_{+-}=\dot{U}_-+\Delta w\Phi', \tag{A10}$$

where U_{+-} is the deformation at point b in Fig. 2d). The initial conditions for (A10) are $\varphi_{+-}|_{t=t_0}=-\varphi_0$, $\varphi_-|_{t=t_0}=-\pi+\varphi_0$, where $t_0=(\pi-2\varphi_0)/\Omega$. An analysis of (A10) once again leads to Eq. (A9) and correspondingly to (10). The form of $\varphi_1(\varphi_0)$, satisfying Eq. (10), is shown in Fig. 4. For different components of the deformation tensor.

[1)] We remark that if the sound wave is accompanied by an electric field, correction terms appear in the equation of motion for the polarization, the inclusion of which (neglecting the background dielectric susceptibility) leads to the change (Ref. 6) $\alpha\to\alpha+4\pi k_z^2k^2$. Provided that this correction does not reduce to zero at the phase transition point, it will lead to an abrupt increase in U_c.

[2)] We note that in order to estimate the width of the shock front, it is in general necessary to take into account the effect of spatial dispersion along with viscosity, if in the corresponding region there is an abrupt change in the medium's characteristics. However, the exact nature of the mechanism which causes the front to steepen is for us irrelevant, provided that we do not underake to analyze the detailed shape there.

[3)] We note that the problem of nonlinear sound propagation in ferroelectrics which are nonpiezoelectric in the paraelectric phase, and which are far away from the tricritical point so that fluctuations can be neglected, was investigated in Ref. 7. In this work it is inferred that this nonlinearity does not have any anomalies in the neighborhood of the phase transition. In our opinion, this inference is connected with certain inconsistencies in the use of the iterative method.

[4)] We remark that although the nonlinear term in (17) is proportional to d^4, for a fixed value of the linear elastic modulus an increase in d leads to a decrease in the nonlinearity. Formally, this is connected with the fact that near the transition $\alpha\sim d^2/\Lambda_0$. For atomic values of the piezomodulus $d^2\sim\Lambda_0$, corresponding to a nonlinear contribution comparable to that of the elastic nonlinearity (which in the situation under investigation, by virtue of symmetry considerations, is appreciable only for an increase of U by three orders of magnitude).

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