Nonlinear dynamics and anomalous damping of electroacoustic waves in order–disorder ferroelectrics

M. B. Belonenko and M. M. Shakirzyanov

E. K. Zavoĭskiĭ Physicotechnical Institute, Kazan Branch of the Academy of Sciences of the USSR, Kazan
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A theory of the electroacoustic echo in single-domain ferroelectrics of the order–disorder type is constructed in the pseudospin formalism. The Heisenberg equations of motion for the mean values of the pseudospin components and the equation for the acoustic vibrations are employed in the random phase approximation to yield equations describing the dynamics of a ferroelectric subjected to pulses of an external alternating field with allowance for damping in both the pseudospin and acoustic subsystems. The multiple scales method is used to obtain from the equations describing the dynamics of the ferroelectric a system of differential equations for the envelopes of the forward and backward electroacoustic wave packets. The equations describing the dynamics of the envelopes of interacting wave packets have the form of coupled nonlinear Schrödinger equations with a perturbation and can be solved by the Karpman–Maslov method if the changes in the eigenfunctions of the Zakharov–Shabat operator are neglected. The solutions are used to investigate the conditions under which the electroacoustic echo arises and the dependence of the echo signal on the temperature, on the amplitude and durations of the alternating-field pulses, on deuteration of the sample, and on the applied static electric field. The behavior of the effective damping of electroacoustic waves as a function of temperature is obtained, and it is shown that the damping rate decreases sharply as the point of the phase transition is approached.

Experimental investigations of the electroacoustic echo effect in single crystals of the ferroelectrics KDP and Rochelle salt[1,2] have detected anomalous behavior of the damping coefficient for electroacoustic waves near the point of the order–disorder phase transition. Both the damping and amplitude of the echo signal were found to depend strongly not only on the parameters of the pulses of the external alternating field (frequency, duration, intensity) but also on the equilibrium characteristics of the ferroelectric itself. The explanation of the experimental relationships that was offered in Ref. 1 on the basis of the phenomenological theory does not take into account all of the features of the rf dynamics of ferroelectrics with thermal phase transitions of the order–disorder type. In particular, tunneling plays an important role in the dynamics of ferroelectrics[3] and must be taken into account in order to obtain the correct dispersion relations of electroacoustic waves and to account for the attenuation of the echo signal upon deuteration of the samples.

In our view, the most complete and consistent description of all the important features of the dynamics of ferroelectrics excited by alternating electric fields can be obtained in the pseudospin formalism, which is widely employed in the theory of ferroelectrics.[3,4]

1. In this paper we use the pseudospin formalism to construct a theory of the electric echo in ferroelectrics of the KDP type, which are characterized by a symmetric double-well potential for the protons in the hydrogen bond. The Hamiltonian of such a system in the pseudospin representation has the form of an Ising Hamiltonian in crossed fields:[3]

$$\mathcal{H}=-\Omega\sum_j S_j^x-\frac{1}{2}\sum_{ij}J_{ij}S_i^zS_j^z -E_0\sum_j S_j^z-\sum_j E_j(t)S_j^z+\mathcal{H}_{sa}, \tag{1}$$

where S_j^x and S_j^z are the tunneling and electric dipole moment operators of the jth cell, Ω is the tunneling integral, J_{ij} is the exchange integral renormalized for the thermal motion of the atoms,[3,4] and E_0 and $E_j(t)$ are the static and alternating electric fields applied to the sample. The operator $\mathcal{H}_{sa}$ is the interaction Hamiltonian of the pseudospins with acoustic vibrations excited in the sample by the alternating electric field owing to the piezoelectric effect.

The specific form of $\mathcal{H}_{sa}$ will depend on the mode and direction of propagation of the acoustic waves with respect to the crystallographic axes x', y', z'. Let us assume it is a transverse acoustic wave propagating along the z' axis and polarized along the y' axis. Then the displacement vector has only one nonzero component: $\mathbf{U}\to(0,U(z',t),0)$. If it is assumed that the equilibrium electric polarization vector $\mathbf{P}$ of the sample is parallel to the static electric field $\mathbf{E}_0$, which is applied parallel to the $x'y'$ plane, and that the acoustic excitation is due to the linear piezoelectric effect, the Hamiltonian $\mathcal{H}_{sa}$ can be written

$$\mathcal{H}_{sa}=-\sum_j d_{123}\frac{\partial U(z',t)}{\partial z'}S_j^z, \tag{2}$$

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where $d_{123} = d_0$ is the corresponding piezoelectric constant. This choice of interaction Hamiltonian $\mathscr{H}_{sa}$ corresponds to the experiment of Ref. 1 (see Fig. 1). We note that the coordinate system $x'y'z'$ is tied to the crystallographic axes, while the coordinate system xyz is defined in pseudospin space.

The dynamics of the system in external fields is described by the Heisenberg equations of motion for the mean values of the pseudospin operators; in the random phase approximation with allowance for transverse relaxation with a time T_2^*, these equations have the form[3-6]

$$\frac{d\langle S_j^x\rangle}{dt}=M_j\langle S_j^y\rangle-\frac{1}{2}\frac{\langle S_j^z\rangle-\langle S^z\rangle_0}{T_2^*}\times\sin 2\varphi-\frac{\langle S_j^x\rangle-\langle S^x\rangle_0}{T_2^*}\sin^2\varphi,$$

$$\frac{d\langle S_j^y\rangle}{dt}=\Omega\langle S_j^z\rangle-M_j\langle S_j^x\rangle-\frac{\langle S_j^y\rangle}{T_2^*},\qquad \mathrm{tg}\,\varphi=\frac{\langle S^z\rangle_0}{\langle S^x\rangle_0}=\chi, \tag{3}$$

$$\frac{d\langle S_j^z\rangle}{dt}=-\Omega\langle S_j^y\rangle-\frac{\langle S_j^z\rangle-\langle S^z\rangle_0}{T_2^*}\times\cos^2\varphi-\frac{\langle S_j^x\rangle-\langle S^x\rangle_0}{2T_2^*}\sin 2\varphi,$$

$$M_j=\sum_i J_{ij}\langle S_i^z\rangle+E_0+E_j(t)+d_0\frac{\partial U(z',t)}{\partial z'},$$

where $\langle S^\alpha\rangle_0$ are the equilibrium mean values of the α-components of the pseudospin ($\alpha = x,y,z$), and φ is the angle between the x axis and the direction of the equilibrium molecular field in pseudospin space. It is assumed in Eqs. (3) that the pseudospin relaxes to an equilibrium value determined by the equilibrium value of the molecular field rather than the instantaneous value. An account of the relaxation of the pseudospin to the state determined by the instantaneous value of the molecular field would go beyond the accuracy of the approximations made below. The appearance of a dependence of the angle φ in the relaxation terms is due simply to the choice of coordinate system in pseudospin space, wherein the z axis, which is parallel to E_0, lies at an angle $\pi/2 - \varphi$ to the direction of the equilibrium molecular field.[4] The system of equations (3) should clearly be supplemented by the equation of propagation of the acoustic wave, which, with allowance for what was said above, can be written in the form[7]

$$\frac{\partial^2 U}{\partial t^2}=V^2\frac{\partial^2 U}{(\partial z')^2}+\frac{1}{\rho}\frac{\partial\sigma}{\partial z'}-\gamma_a\frac{\partial U}{\partial t},\qquad U\equiv U(z',t), \tag{4}$$

where ρ is the density of the crystal, and V and γ_a are the velocity and damping rate of the acoustic wave in the absence of pseudospin–phonon coupling (which are therefore free of anomalies at the phase transition point). The component of the stress tensor σ arising during the motion of a coupled electroacoustic wave excited in the sample by an alternating field is given as

$$\sigma=\frac{\partial\langle\mathscr{H}_{sa}\rangle}{\partial(\partial U/\partial z')}.$$

2. The system of equations (3) contains $3N$ equations, where N is the number of ferroelectric cells of the sample, and it cannot be solved in the form in which it is given. However, ferroelectrics of the KDP type have a layered structure and can be represented as a set of planes parallel to the plane $x'y'$ containing the spontaneous polarization vector $P_0 \propto \langle S^z\rangle_0 = \langle S_j^z\rangle_0$. The exchange interaction between pseudospins lying in different planes is substantially smaller than the interaction between pseudospins in the same plane.[3,4] This circumstance makes it possible to consider only the interaction of nearest-neighbor planes, with indices $n-1$, n, and $n+1$.

On the other hand, since the wavelength of the electric component of the exciting field is ordinarily much larger than the dimensions of the sample along the $x'y'$ plane, the field in this direction can be considered uniform. Then, for identical initial conditions for the pseudospins of a given plane it is easy to show that $\langle S_j^\alpha(t)\rangle = \langle S_l^\alpha(t)\rangle$ if $\langle S_j^\alpha(0)\rangle = \langle S_l^\alpha(0)\rangle$, $l \neq j$. Then, if the index j specifying the position of the pseudospin in the lattice is represented as a set of two indices n and k, where n is the number of the plane in which the spontaneous polarization vector lies and k describes the position of the pseudospin within the plane, one can write

$$\sum_j J_{ij}\langle S_i^z\rangle=\sum_{n,k}J_{nk;n'k'}\langle S_{nk}^z\rangle\approx\langle S_{n'k'}^z\rangle\sum_k J_{n'k;n'k'}+\langle S_{(n'+1)k'}^z\rangle\sum_k J_{(n'+1)k;n'k'}+\langle S_{(n'-1)k'}^z\rangle\sum_k J_{(n'-1)k;n'k'}. \tag{5}$$

Since the wavelength λ of the alternating field is much greater than the distance a between neighboring planes, it can be assumed that the mean value of the z component of the pseudospin vector changes only slightly from plane to plane, and we can expand $\langle S_{(n\pm1)k}\rangle$ in a Taylor series:

$$\langle S_{(n\pm1)k}^z\rangle\approx\langle S_{nk}^z\rangle\pm a\frac{\partial\langle S_{nk}^z\rangle}{\partial z'}+\frac{a^2}{2}\frac{\partial^2\langle S_{nk}^z\rangle}{(\partial z')^2}+\ldots \tag{6}$$

Then, with allowance for (5) and (6), the system of equations (3) and (4) can be written

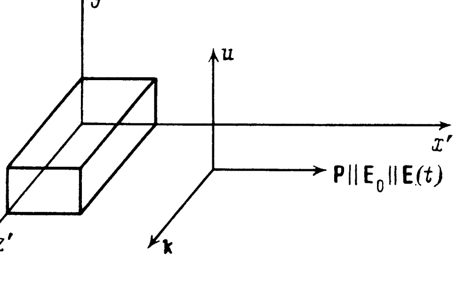

FIG. 1. Relationships among the polarization direction **P**, the direction of wave propagation **k**, the nonzero component of the displacement vector $U(z',t)$, and the crystallographic axes x', y', z'.

$$\frac{d\langle S^x\rangle}{dt}=M\langle S^y\rangle-\frac{\langle S^z\rangle-\langle S^z\rangle_0}{2T_2^*}$$

$$\times\sin 2\varphi-\frac{\langle S^x\rangle-\langle S^x\rangle_0}{T_2^*}\sin^2\varphi,$$

$$\frac{d\langle S^y\rangle}{dt}=\Omega\langle S^z\rangle-M\langle S^x\rangle-\frac{\langle S^y\rangle}{T_2^*},\qquad \langle S^\alpha\rangle\equiv\langle S^\alpha(z',t)\rangle,$$

$$(7)$$

$$\frac{d\langle S^z\rangle}{dt}=-\Omega\langle S^y\rangle-\frac{\langle S^z\rangle-\langle S^z\rangle_0}{T_2^*}$$

$$\times\cos^2\varphi-\frac{\langle S^x\rangle-\langle S^x\rangle_0}{2T_2^*}\sin 2\varphi,$$

$$\frac{d^2U}{dt^2}-V^2\frac{\partial^2U}{(\partial z')^2}+\gamma_a\frac{dU}{dt}+\frac{d_0}{\rho}\frac{\partial\langle S^z\rangle}{\partial z'}=0,\ U\equiv U(z',t),$$

where we have introduced the notation

$$\langle S_{nk}{}^\alpha\rangle\equiv\langle S^\alpha\rangle,\qquad J=\sum_k\,[J_{n'k;n'k'}+2J_{(n'+1)k;n'k'}],$$

$$M=J\langle S^z\rangle+A\frac{\partial^2\langle S^z\rangle}{(\partial z')^2}+E_0+E(t)+d_0\frac{\partial U}{\partial z'},$$

$$A=a^2\sum_k J_{(n'+1)k;n'k'}.$$

To investigate the system (7) we use the multiple scales technique,[8,9] according to which we seek a solution of (7) in the form

$$\langle S^z\rangle=\langle S^z\rangle_0+Z=\langle S^z\rangle_0+\varepsilon Z^{(1)}+\varepsilon^2Z^{(2)}+\dots,$$

$$Z^{(1)}=Z_+\exp\{i(\omega t-kz')\}+Z_-\exp\{-i(\omega t+kz')\}+\text{c.c.},\qquad (8)$$

$$Z_\pm\equiv Z_\pm(\varepsilon t,\ \varepsilon^2t;\ \varepsilon z',\ \varepsilon^2z')=Z_\pm(T_1,\ T_2;\ z_1),$$

$$T_n=\varepsilon^nt,\quad z_n=\varepsilon^nz'.$$

Here ω and $\mathbf{k}$ are the frequency and wave vector of the traveling electroacoustic waves, Z_+ and Z_- are the slowly varying amplitude of the forward and backward (traveling in the opposite direction) waves, T_n and z_n are slow variables, and ε is a small parameter characterizing the deviation of the parameters of the pseudospin system from their equilibrium values. It should be noted that $Z_\pm$ depends only on the variables T_1, T_2, and z_1, since it has been shown[8] that the dependence on the variable z_2 can always be eliminated by transforming to a new coordinate system.

The derivation of the effective equations characterizing the dynamics of the envelopes $Z_\pm$ of the electroacoustic waves (whose solution describes the electroacoustic echo effect) from Eqs. (7) by the multiple scales technique involves a great volume of unwieldy transformations. As the required series of transformations has been reported in detail in the literature,[8,9] here we shall only discuss and justify the approximations made in the course of the calculations and indicate the structure of the transformations. First of all one must determine correctly the order of smallness of the terms describing the damping and the excitation by the external alternating field. It is assumed that the quantities $(T_2^*)^{-1}$ and γ_a are of order ε. This is because the change in the amplitude of the envelope on account of damping processes takes place over times which are slower than the vibrational period of the electroacoustic waves, which determines the fast time. In determining the degree of smallness of the terms involving the alternating field one must keep in mind that efficient excitation of intercoupled pseudospin and acoustic waves by the alternating field occurs at the frequency of the natural oscillations, which are determined from the solution of the linearized system (7) for $E(t)=0$. Then, since the particular solution (dependent on the external alternating field) of system (7) to first order in ε and the solution of the homogeneous linearized system (7) run together (they have the same dependence on the space and time coordinates), the effect of the alternating field cannot be isolated in this case.[10] To solve this problem, i.e., to take into account the effect of an alternating field having the same order of smallness as the deviation of the dynamical system from equilibrium, a method of renormalization of the amplitude of the external field has been proposed.[10] In this method, the effect of the alternating field with the renormalized amplitude is taken into account simultaneously with the nonlinear terms, i.e., to third order in ε in the present case. Finally, we note that the first three equations of system (7) yield an approximate quasi-integral of the motion:

$$\sum_\alpha(\langle S^\alpha\rangle)^2\approx\text{const},\qquad (9)$$

which is exact upon neglect of relaxation processes in the pseudospin system. Thus, with allowance for what we have said, after a series of transformations the system of equations (7) can be reduced to a single equation:

$$\frac{\partial^2Q}{\partial t^2}-V^2\frac{\partial^2Q}{(\partial z')^2}+\gamma_a\frac{\partial Q}{\partial t}+\frac{d_0{}^2\Omega\langle S^x\rangle_0}{\rho}\frac{\partial^2Z}{(\partial z')^2}=0,$$

$$Q=L+S+K+O(Z^4),$$

$$L=\ddot Z+\Omega^2Z-\Omega\langle S^x\rangle_0\left\{JZ+A\frac{\partial^2Z}{(\partial z')^2}+E(z',t)\right\}+\tilde P\Omega\chi Z,$$

$$(10)$$

$$S=\frac{\tilde P\Omega}{2\langle S^x\rangle_0}\left\{Z^2(1+3\chi^2)+\frac{\dot Z^2}{\Omega^2}\right\}+\chi\frac{Z}{\langle S^x\rangle_0}(\ddot Z+\Omega^2Z)$$

$$+\frac{\dot Z}{T_2^*}\{1+\cos^2\varphi(1-\chi^2)\},$$

$$K=\chi^2\frac{Z^2}{(\langle S^x\rangle_0)^2}\{\ddot Z+\Omega^2Z+\tilde P\Omega\chi Z\}$$

$$+\frac{1}{2(\langle S^x\rangle_0)^2}\left\{Z^2(1+\chi^2)+\frac{\dot Z^2}{\Omega^2}\right\}$$

$$\times\{\ddot{Z}+\Omega^2 Z+3\tilde{P}\Omega\chi Z\}-\frac{\ddot{Z}\dot{Z}\cos^2\varphi}{T_2^{\cdot}\Omega^2}\chi$$

$$-\frac{\dot{Z}Z\cos^2\varphi}{T_2^{\cdot}\langle S^x\rangle_0}\chi\left\{1+\frac{1}{\cos^2\varphi}-3\chi^2\right\},$$

$$\tilde{P}=J\langle S^z\rangle_0+E_0,\quad \chi=\frac{\langle S^z\rangle_0}{\langle S^x\rangle_0},\quad \dot{Z}=\frac{\partial Z}{\partial t},\quad \ddot{Z}=\frac{\partial^2 Z}{\partial t^2}.$$

3. The multiple scales technique is based on the successive elimination of the rapidly oscillating secular terms, proportional to $\exp[\pm i(\omega t-kz')]$ in each order of the expansion of Z in the parameter ε. For example, the requirement of eliminating the secular terms in first order in ε yields the dispersion relation for electroacoustic waves. The corresponding equation for the secular terms obtained in second order in ε yields equations describing the dynamics of the envelopes of noninteracting packets of forward and backward electroacoustic waves. Finally, in third order in ε one obtains equations describing the dynamics of the envelopes of interacting forward and backward wave packets.[8,9] The dispersion relation obtained is of the form

$$(\omega^2-\omega_a^2)(\omega^2-\omega_e^2)-\Omega\langle S^x\rangle_0 d_0^2k^2/\rho\equiv l(\omega,k)=0,$$
$$\omega_a^2=k^2V^2,\quad \omega_e^2=\Omega^2-\Omega\langle S^x\rangle_0(J-Ak^2)+\Omega\tilde{P}\chi, \tag{11}$$

where ω_a and ω_e are, respectively, the eigenfrequencies of acoustic vibrations and pseudospin waves with wave vector **k** in the absence of coupling between them. The equations for the envelopes of the noninteracting forward and backward wave packets are

$$\frac{\partial Z_\pm}{\partial T_1}\pm\frac{d\omega}{dk}\frac{\partial Z_\pm}{\partial z_1}+\gamma Z_\pm=0,$$
$$\gamma=\frac{1}{2}\frac{(\omega^2-\omega_a^2)\gamma_s+(\omega^2-\omega_e^2)\gamma_a}{2\omega^2-\omega_e^2-\omega_a^2},\quad \gamma_s=\frac{1}{T_2^{\cdot}}(1+\cos2\varphi); \tag{12}$$

the solution of these equations can be written

$$Z_\pm=\tilde{Z}_\pm\left(\left(z_1\mp\frac{d\omega}{dk}T_1\right);\ T_2\right)\exp(-\gamma T_1), \tag{13}$$

where $\tilde{Z}_\pm$ is some function of the slow variables $z_1\mp(d\omega/dk)T_1$; T_2.

Thus the wave packet of the forward (backward) electroacoustic wave Z_+ (Z_-) moves with group velocity $d\omega/dk$ ($-d\omega/dk$) and is damped at a rate γ which is a function of the damping coefficients of the pseudospin (γ_s) and acoustic (γ_a) waves and also of their frequencies. Let us analyze the temperature dependence of the damping coefficient γ from the experimental studies.

It is known that the damping coefficients of both the pseudospin and acoustic waves exhibit a very weak temperature dependence.[11,13] Therefore, the temperature dependence of the damping coefficient is governed primarily by the strong temperature dependence of the frequency ω_e of the pseudospin wave. At a fixed frequency of the external alternating field ω the frequencies ω_a and ω_e of the acoustic and pseudospin waves depend on the magnitude of the wave vector **k** (see Eq. (11)). At the same time, since the dependence of ω_e on k is due to the very weak interplane exchange interaction A ($A\ll J$), we can neglect this dependence and assume that $\omega_e^2\sim\omega_0^2$, where $\omega_0^2=\Omega^2-\Omega\langle S^x\rangle_0 J+\Omega\tilde{P}\chi$ is the soft mode frequency.[3] In this approximation an expression for k^2 is easily obtained from the dispersion relation (11):

$$k^2\simeq\frac{\omega^2(\omega^2-\omega_0^2)}{B+V^2(\omega^2-\omega_0^2)},\quad B=\frac{\Omega\langle S^x\rangle_0 d_0^2}{\rho}. \tag{14}$$

Then the expression for γ becomes

$$\gamma\approx\frac{1}{2}\gamma_a+\frac{1}{2}\frac{\gamma_s-\gamma_a}{1+\tilde{\Delta}(1+\tilde{\Delta}V^2\omega^2/B)},\quad \tilde{\Delta}=\frac{\omega^2-\omega_0^2}{\omega^2}. \tag{15}$$

Because the piezoelectric constant $d_0=d_{123}$ is zero in the ferrophase ($T<T_c$, where T_c is the phase transition temperature), the electroacoustic waves coupled with it are not excited in the ferrophase. In the paraphase ($T>T_c$) the temperature dependence of d_{123} is of the form $d_{123}=d_1+d'/(T-T_c)$ (Ref. 11). The behavior of the soft mode frequency with temperature has been well studied.[3,4] Specifically, while $\omega_0\sim\Omega$ in the high-temperature region, as T approaches T_c the soft mode frequency decreases sharply, and for $E_0\neq0$ it becomes a small quantity of order $2\mu E_0\ll\omega$ (μ is the dipole moment of a single ferroelectric cell). Thus in the high-temperature region, where $\tilde{\Delta}\sim\Omega^2/\omega^2$, $B\sim\Omega^2d_0^2/\rho k_BT$, we can write approximately

$$\gamma=\gamma_1\approx\frac{1}{2}\gamma_a+\frac{1}{8}\frac{(\gamma_s-\gamma_a)d_0^2\omega^2}{\rho V^2\Omega^2k_BT}. \tag{16}$$

Since $\gamma_s\gg\gamma_a$ (Refs. 3 and 4), it follows from (16) that the damping of the electroacoustic wave increases as the temperature is lowered. Since the soft-mode frequency decreases substantially with decreasing temperature, there is a region of temperatures near T_c ($T>T_c$) in which $\omega_0\sim\omega$ ($\omega\ll\Omega$). In this case, according to Eq. (15), we have

$$\gamma=\gamma_2\approx{}^1/_2\gamma_s;\quad \gamma_2\gg\gamma_1. \tag{17}$$

i.e., the damping is much greater than in the high-temperature region. In the immediate vicinity of the phase transition point T_c ($T\to T_c$), Eq. (15) gives

$$\gamma=\gamma_3\approx\frac{1}{4}(\gamma_s+\gamma_a)+\frac{1}{8}(\gamma_s-\gamma_a)\frac{T-T_c}{T_c}\frac{\Omega^2}{\omega^2},\quad \gamma_3<\gamma_2. \tag{18}$$

Thus it follows from the expressions obtained for the damping coefficients in different temperature regions (Eqs. (16)–(18)) that the damping of electroacoustic waves is maximum in the temperature region where the soft mode frequency is close to the frequency of the external alternating field. Such behavior of the damping coefficient is in qualitative agreement with the experimentally measured temperature dependence of the damping of echo signals in KDP crystals.[1,2]

4. The simplest way of exciting echo signals is to apply two successive pulses of an external alternating field with a time between pulses $\Delta t = \tau_0$, the first pulse have duration τ_1 and frequency ω and the second having duration τ_2 and frequency 2ω. Such a succession of electric field pulses can be written in the form[1,2,12] [$\theta(x)$ is the theta function]:

$$E(z', t) = {}^1/_2 E_1 f(z_1)\{\theta(t) - \theta(t+\tau_1)\}$$
$$\times \exp[i(\omega t - kz')] + {}^1/_2 E_2\{\theta(t+\tau_0+\tau_1)$$
$$-\theta(t+\tau_1+\tau_0+\tau_2)\}\exp(i2\omega t) + \text{c.c.}$$

In accordance with what we have said, equations (10) to third order in ε yield equations describing the excitation and dynamics of the envelopes of interacting forward and backward electroacoustic wave packets:

$$\mp i l_\omega \frac{\partial Z_\pm}{\partial T_2} - \frac{1}{2} l_{\omega\omega} \frac{\partial^2 Z_\pm}{\partial T_1^2} - \frac{1}{2} l_{kk} \frac{\partial^2 Z_\pm}{\partial z_1^2}$$
$$\pm l_{\omega\omega} \frac{\partial^2 Z_\pm}{\partial T_1\, \partial z_1} + \Delta |Z_\pm|^2 Z_\pm$$
$$+\Phi |Z_\mp|^2 Z_\pm \pm R_3 \frac{\partial Z_\pm}{\partial z_1} - R_4 \frac{\partial Z_\pm}{\partial T_1} - R_5 Z_\pm + h_\pm E_1 f(z_1)$$
$$+ (R_1\langle S^z\rangle_0 + R_2 E_0) g_\pm(E_2) Z_\mp = 0,$$
$$l_\xi = \frac{\partial l(\omega, k)}{\partial \xi}, \quad l_{\xi\tilde\eta} = \frac{\partial^2 l(\omega, k)}{\partial \xi \partial \tilde\eta}, \quad h_- = 0$$
$$h_+ = \frac{1}{2}\Omega\langle S^x\rangle_0(\omega^2 - \omega_a^2)[\theta(t) - \theta(t+\tau_1)]; \quad \xi, \tilde\eta = \omega, \mathbf{k}, \tag{19}$$

where

$$\Delta = \frac{\omega_a^2 - \omega^2}{(\langle S^x\rangle_0)^2}\Omega\Big\{H\langle S^x\rangle_0\Big[\tilde P\Big(1+3\chi^2+\frac{\omega^2}{\Omega^2}\Big) + \frac{\chi}{\Omega}(2\Omega^2-\omega^2)\Big]$$
$$+\frac{3\chi}{\Omega}(\Omega^2-\omega^2+\Omega\tilde P\chi) + \frac{3}{2}\Big(1+\chi^2+\frac{\omega^2}{3\Omega^2}\Big)\Big(3\tilde P\chi + \frac{\Omega^2-\omega^2}{\Omega}\Big)\Big\},$$
$$H = \frac{4(\omega_a^2-\omega^2)}{l(2\omega, 2k)(\langle S^x\rangle_0)^2}\Big[\frac{\tilde P}{2}\Big(1+3\chi^2-\frac{\omega^2}{\Omega^2}\Big) + \frac{\chi}{\Omega}(\Omega^2-\omega^2)\Big],$$

$$R_2 = \frac{\omega_a^2 - \omega^2}{\langle S^x\rangle_0}\Omega\Big(1+3\chi^2+2\frac{\omega^2}{\Omega^2}\Big),$$

$$R_1 = JR_2 + \frac{(\omega_a^2-\omega^2)(2\Omega^2-5\omega^2)}{(\langle S^x\rangle_0)^2},$$

$$g_\pm(E_2) = -\frac{E_2\omega^2}{2l(2\omega, 0)}[\theta(t+\tau_0+\tau_1) \pm \theta(t+\tau_1+\tau_0+\tau_2)].$$

Here R_3, R_4, R_5, and Φ are complicated functions of the parameters of the system Hamiltonian, equilibrium mean values $\langle S^\alpha\rangle_0$, and damping constants γ_s and γ_a.

In order to solve Eqs. (19) we must specify the initial and boundary conditions. Assuming that the system is initially in a state of thermodynamic equilibrium, we write the initial conditions in the form

$$Z_\pm(z_1)|_{t=T_1=T_2=0} = 0. \tag{20}$$

Under the condition that the first pulse excites traveling waves in the sample, the boundary conditions can be specified for a semi-infinite sample. Since the corresponding component of the total stress tensor is equal to zero at the boundary of the sample,[11] the boundary conditions are

$$Z_\pm(T_1, T_2)|_{z_1\to\pm\infty} \to 0. \tag{21}$$

We note that boundary conditions (21) for a semi-infinite sample correspond to specifying boundary conditions on the components of the electric displacement vector in the form:

$$E^{(1)}_{tang} = E^{(2)}_{tang}, \quad D^{(1)}_{norm} = D^{(2)}_{norm}.$$

It follows from system of equations (19) and initial conditions (20) that until the start of the second pulse there is only a forward electroacoustic wave in the sample ($Z_- = 0$), propagating in the direction of increasing z'. Making the substitution $Z_+ = q_+ \exp(i\mu_+ T_2 + i\eta z_1)$, where

$$\mu_+ = \Big(-\frac{1}{2} l_{\omega\omega}\gamma^2 - R_5 + R_4\gamma\Big) l_\omega^{-1} - \frac{1}{2}\frac{d^2\omega}{dk^2}\eta^2,$$

$$\eta = i\Big\{l_{\omega\omega}\frac{l_k}{l_\omega}\gamma - l_{k\omega}\gamma + R_3 - R_4\frac{l_k}{l_\omega}\Big\}\Big(\frac{d^2\omega}{dk^2} l_\omega\Big)^{-1} + \frac{d\omega}{dk}\Big(\varepsilon\frac{d^2\omega}{dk^2}\Big)^{-1},$$

we obtain with the aid of relation (13) [here Im (f) is the imaginary part of f]

$$-i\frac{\partial q_+}{\partial T_2} + \frac{1}{2}\frac{d^2\omega}{dk^2}\frac{\partial^2 q_+}{\partial z_1^2} + \frac{\Delta}{l_\omega}|q_+|^2 q_+ + R_+ = 0,$$
$$R_+ = h_+ E_1 f(z_1) l_\omega^{-1}\exp(\gamma T_1 - i\mu_+ T_2 - i\eta z_1) \tag{22}$$
$$+ \Delta l_\omega^{-1}|q_+|^2 q_+\{\exp[-2\gamma T_1 - 2\,\text{Im}(\mu_+ T_2) - 2\,\text{Im}(\eta z_1)] - 1\}.$$

The Cauchy problem for Eq. (22) with boundary conditions (21) on the half line $z_1 \in [0; +\infty]$ is equivalent to the Cauchy problem on the straight line $z_1 \in]-\infty; +\infty[$ with the initial and boundary conditions

$$q_+|_{z_1\to\pm\infty} \Rightarrow 0, \quad q_+|_{T_2=0} = 0, \tag{23}$$

if the definition of the term proportional to h_+ is extended onto the half line $z_1 \in]-\infty; 0]$ as a function which is odd with respect to the argument z_1 and which is equal to zero at $z_1 = 0$ (see, e.g., Refs. 14 and 15). In this case Eq. (22) with conditions (23) can be regarded as a nonlinear Schrödinger equation with zero boundary conditions in the presence of an external perturbation R_+. Equation (22) can be solved by the Karpman–Maslov method[16] if the change in the eigenfunctions of the Zakharov–Shabat operator under the influence of the external perturbation R_+ is neglected. This approximation corresponds to the case when the amplitude of the first alternating-field pulse is small. In fact, the change in the eigenfunctions of the Zakharov–Shabat operator can be neglected under the condition[16]

$$\Big|\frac{\Delta}{l_\omega}\Big| \gg R_+.$$

Hence, using expressions (22) and (19) and neglecting the damping that occurs during the alternating-field pulse, we obtain a condition on the amplitude E_1:

$$E_1 \ll \frac{\Delta}{(\omega^2-\omega_a^2)\Omega\langle S^x\rangle_0}.$$

in the paraphase of the ferroelectric, when $\langle S^z\rangle_0 \sim 0$ (in the ferrophase there are no electroacoustic waves coupled with the given piezoelectric constant; see above), the quantity Δ is of the order of

$$\Delta \sim \frac{3}{2}(\omega^2-\omega_a^2)\frac{\Omega^2}{(\langle S^x\rangle_0)^2},$$

and thus for E_1 we obtain

$$E_1 \ll \frac{3}{2}\frac{\Omega}{(\langle S^x\rangle_0)^3}.$$

This inequality is clearly satisfied for the values of E_1 ordinarily used in experiment,[1,2] since, for the KDP crystal, for example, the tunneling integral $\Omega \sim 10^{13}$ s^{-1}, and $\langle S^x\rangle_0 \sim \Omega/J < 1$ in the vicinity of the phase transition point.

According to the Karpman–Maslov method, the solution of Eq. (22) it determined from the scattering data $c_{11}(w)$, $c_{12}(w)$ as[16]

$$q_+ = \left(\left|\frac{8l_\omega}{\Delta}\right|\right)^{1/2} K(\delta\cdot z_1, \delta\cdot z_1), \qquad \delta = \left(\left|\frac{1}{2}\frac{d^2\omega}{dk^2}\right|\right)^{-1/2}, \tag{24}$$

where the function $K(x,y)$ is a solution of the integral equation

$$K(x,y) - F^*(x+y) + \int_x^\infty\int_x^\infty ds\, ds' K(x,s)F(s+s')F^*(s'+y) = 0,$$

$$F(r) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dw\, e^{iwr}\frac{c_{11}(w)}{c_{12}(w)}. \tag{25}$$

The evolution of the scattering data in time is governed by the equations

$$\frac{\partial c_{12}(w)}{\partial T_2} = 0, \qquad \frac{\partial c_{11}(w)}{\partial T_2} - i4w^2 c_{11}(w)$$
$$= -i\left(\left|\frac{\Delta}{2l_\omega^3}\right|\right)^{1/2}\delta\int_{-\infty}^{\infty} dz_1 R_+^*(z_1;T_2)\exp(-2iw\delta\cdot z_1) \tag{26}$$

with initial conditions according to (23):

$$c_{11}(w)|_{T_2=0} = 0, \quad c_{12}(w)|_{T_2=0} = 1.$$

The solutions of equations (26) at times $T_2 > \tau_1$ are

$$c_{11}(w) = \Omega E_1\langle S^x\rangle_0\left(\left|\frac{\Delta}{2l_\omega^3}\right|\right)^{1/2}\Gamma(w)(\mu_+ + 4w^2)^{-1}(\omega^2-\omega_a^2)$$
$$\times[\exp(-i\mu_+\tau_1+\gamma\tau_1) - \exp(4iw^2\tau_1)]\exp(4iw^2T_2),$$
$$c_{12}(w) = 1, \tag{27}$$

where $\Gamma(w)$ is the Fourier transform of the first pulse:

$$\Gamma(w) = \delta\int_{-\infty}^{\infty} dz_1 f(z_1)\exp(-i\eta z_1)\exp(i2w\delta\cdot z_1).$$

By expressing Z_+ in terms of the scattering data $c_{11}(w)$ and $c_{12}(w)$, it is easy to determine the evolution of the forward electroacoustic wave up until the start of the second pulse.

The interaction of the second pulse, having frequency 2ω, with the forward electroacoustic wave is parametric and so gives rise to a backward electroacoustic wave. This interaction, which is characterized by the term proportional to $g_-(E_2)$ in the equation for the amplitude of the backward wave envelope (the equation for Z_-) in system (19), acts as an external driving force. The backward wave moves in the $-z'$ direction with the same group velocity as the forward wave, and it is detected as an echo response. It follows from the last term of Eq. (19) [the term proportional to $g_-(E_2)$] that excitation of a backward wave through the interaction of the forward wave and the second field pulse is possible only under the condition $R_1\langle S^z\rangle_0 + R_2E_0 \neq 0$. Thus, since $\langle S^z\rangle_0 = 0$ in the paraphase, the formation of a backward wave (and hence an echo signal) in this phase requires the presence of a static electric field ($E_0 \neq 0$). At the same time, in the ferrophase, where $\langle S^z\rangle_0 \neq 0$, electroacoustic waves are not excited because the corresponding piezoelectric constant $d_{123} = 0$.

In the free-evolution period after the end of the second pulse, the system (19) in the approximation of noninteracting waves is a system of independent equations. After the change of variables

$$Z_- = q_-\exp(i\mu_-T_2 + i\eta z_1), \qquad \mu_- = -\mu_+ + \frac{d^2\omega}{dk^2}\eta^2 \tag{28}$$

the equation for the amplitude of the backward wave envelope can be written

$$i\frac{\partial q_-}{\partial T_2} + \frac{1}{2}\frac{d^2\omega}{dk^2}\frac{\partial^2 q_-}{(\partial z_1)^2} + \frac{\Delta}{l_\omega}|q_-|^2q_- + R_- = 0,$$
$$R_- = l_\omega^{-1}(R_1\langle S^z\rangle_0 + R_2E_0)g_-(E_2)q_+\exp[-i(\mu_- - \mu_+)T_2]$$
$$+\Delta l_\omega^{-1}|q_-|^2q_-\{\exp[-2\gamma T_1 - 2\,\mathrm{Im}(\eta z_1) - 2\,\mathrm{Im}(\mu_-T_2)] - 1\}. \tag{29}$$

Because the function q_+ is odd with respect to z_1, Eq. (29) is defined on the straight line $z_1 \in]+\infty; -\infty[$ with zero initial and boundary conditions. Thus Eq. (29), which describes the dynamics of the backward wave, is completely analogous to Eq. (22) which governs the evolution of the forward electroacoustic wave. Consequently, the solution of equation (29) is analogous to the solution (24)–(26) of equation (22). The scattering data $\tilde{c}_{11}(w)$, $\tilde{c}_{12}(w)$, which govern the evolution of the backward wave at times $T_2 > \tau_1 + \tau_0 + \tau_2$, have the form

$$\tilde{c}_{11}(w) = E_1E_2\Omega\langle S^x\rangle_0(R_1\langle S^z\rangle_0 + R_2E_0)\Gamma(w)\left(\left|\frac{\Delta}{2l_\omega^5}\right|\right)^{1/2}$$
$$\times\frac{\omega^2-\omega_a^2}{\mu_+ + 4w^2}(\mu_+ - \mu_- + 8w^2)^{-1}[\exp(-i\mu_+\tau_1 + \gamma\tau_1)$$
$$-\exp(4iw^2\tau_1)]$$
$$\times\exp[-i4w^2(T_2 - 2\tau_0 - \tau_1 - \tau_2)]\{\exp[i(4w^2 + \mu_+ - \mu_-)\tau_2]$$
$$-\exp(-i4w^2\tau_2)\} + O(E_1^2),$$
$$\tilde{c}_{12}(w) = 1. \tag{30}$$

The condition imposed on the amplitude E_2 of the second pulse in order for Eqs. (30) to be a valid solution of equations (29) is analogous to the condition obtained for the amplitude E_1 of the first pulse and is given by the inequality $|\Delta/l_\omega| \gg R_-$. With the same approximations as were used for E_1, we obtain for E_2

$$E_2 \ll \frac{3}{2}\frac{\Omega^2}{E_0(\langle S^x\rangle_0)^2 q_+}.$$

From expressions (24)–(27), if we neglect the dispersion of the envelope of the forward wave during the alternating-field pulse, we obtain for the amplitude of the envelope q_+

$$q_+ \propto \frac{2E_1\langle S^x\rangle_0}{\Omega}.$$

Then, with allowance for the experimental[1,2] conditions $E_0 \sim E_1 \sim E_2$, we can write the following inequality for E_2:

$$E_2 \ll \Omega/\langle S^x\rangle_0,$$

which is practically always satisfied.

We note that the time T_2 appears in expressions (26) and (30) with opposite signs. This corresponds to the well-known "phase conjugation" effect, wherein the backward wave seem to evolve backwards in time. To determine q_- from the scattering data $\tilde{c}_{12}(w)$ and $\tilde{c}_{11}(w)$ one can use relations (24) and (25), after transforming the function $F(r)$ as

$$F(r) \to F_{\rm back}(r) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dw \frac{\tilde{c}_{11}(w)}{\tilde{c}_{12}(w)} e^{iwr}. \quad (31)$$

For alternating-field pulses having low amplitudes and satisfying the inequalities obtained above, we obtain from Eq. (25)

$$K(x, y) \approx F^*(x+y).$$

In this case the amplitude of the backward wave envelope is given approximately by

$$Z_-(T_1, T_2, z_1) \approx E_1E_2(R_1\langle S^z\rangle_0 + R_2E_0)\Omega\langle S^x\rangle_0(\omega^2-\omega_a^2) \times l_\omega^{-1} W e^{-\gamma T_1} \exp i\{\mu_- T_2 + \eta z_1\}, \quad (32)$$

$$W = \frac{1}{\pi}\int_{-\infty}^{\infty} dw \frac{\Gamma(w)}{\mu_+ + 4w^2} \times \frac{[\exp(-i\mu_+\tau_1 + \gamma\tau_1) - \exp(i4w^2\tau_1)]\exp(i2w\delta\cdot z_1)}{\mu_+ - \mu_- + 8w^2}$$

$$\times[\exp i(4w^2 + \mu_+ - \mu_-)\tau_2 - \exp(-i4w^2\tau_2)]$$

$$\times \exp[-i4w^2(T_2 - 2\tau_0 - \tau_1 - \tau_2)]. \quad (33)$$

5. Let us examine how the amplitude of the backward electroacoustic wave envelope (32) depends on time, on the amplitudes and durations of the pulses, and on the parameters of the sample. Using the method of stationary phase, one can easily show that the integral for W in (33) has a maximum at the time $T_2 = \tau_1 + 2\tau_0 + \tau_1$. Consequently, at the time $T_2 + \tau_1 + 2\tau_0 + \tau_2$ the amplitude of the backward wave increases sharply and is observed as an echo response. Ordinarily in experiments $\tau_0 \gg \tau_1, \tau_2$ and $T_2 \approx 2\tau_0$, i.e., the time at which the echo signal appears is determined by the interval between pulses. We note that according to (32), for short pulse durations τ_1 and τ_2 the echo amplitude $A_e = Z_-(T_2 \approx 2\tau_0)$ is proportional to τ_1 and τ_2:

$$A_e \propto E_1E_2\tau_1\tau_2.$$

It also follows from (30) that for small τ_2

$$\tilde{c}_{11}(w,\ T_2 = 2\tau_0 + \tau_1 + \tau_2) = c_{11}(w,\ T_2 = \tau_1)E_2\tau_2(R_1\langle S^z\rangle_0 + R_2E_0)l_\omega^{-1}. \quad (34)$$

According to (25), the scattering data $c_{22}(w, T_2 = \tau_1)$ determine the shape of the envelope of the forward wave $Z_+(T_2 = \tau_1)$ after the end of the second pulse:

$$Z_+(T_2 = \tau_1) \propto \int_{-\infty}^{\infty} dw \frac{c_{11}(w)}{c_{12}(w)} \exp(2iwz_1).$$

According to (34), the shape of the echo signal

$$Z_-^e(T_2 = 2\tau_0 + \tau_1 + \tau_2) \propto \int_{-\infty}^{\infty} dw \frac{\tilde{c}_{11}(w)}{\tilde{c}_{12}(w)} \exp(2iwz_1), \quad (35)$$

is also determined by the scattering data $c_{11}(w, T_2 = \tau_1)$, and, consequently, is analogous to that of the forward wave envelope. As the intensity of the alternating-field pulses is increased, it becomes necessary to take into account the integral term in Eq. (25). Assuming that E_1 and E_2 still satisfy the corresponding inequalities, we can write

$$K(x, y) \approx F^*(x+y) - \int_x^{\infty}\int_x^{\infty} ds\, ds' F^*(x+s)F(s+s')F^*(s'+y), \quad (36)$$

and, consequently, the amplitude of the backward wave will have the form

$$Z_-'(T_1, T_2, z_1) \approx Z_- - (E_1E_2)^3\tilde{N}, \quad (37)$$

where Z_- is solution (32) and $\tilde{N}$ is the function given by the double integral in (36). Using the method of stationary phase, we easily see that at the time $T_2 = 2\tau_0 + \tau_1 + t_2$ the signs of the functions $F^*(x+y)$ and $\tilde{N}$ are the same.

Thus, it follows from expressions (36) and (37) that the echo amplitude saturates as the amplitudes of the alternating-field pulses are increased. It should be noted, however, that solution (37) of the linear integral equation (25) was obtained by successive approximations in the small parameter $E_i/\Omega \ll 1$. Obviously, the conclusion that the echo signal saturates can be considered valid only when the inequalities $E_i/\Omega \ll 1$ hold for the amplitudes of the alternating fields ($\langle S^x\rangle_0 \lesssim 0.1$ in the paraphase). At the same time, we note that in the case of strong fields analysis of the dependence of the echo signal on the parameters of the pulse train requires correct allowance for the changes in the eigenfunctions of the Zakharov–Shabat operator and also for the possible contribution of solitons generated by the alternating fields. Consideration of these questions is beyond the scope of this paper.

As we see from expression (30), the amplitude of the echo signal is proportional to the tunneling integral Ω:

$$A_e = Z_-(T_2 \approx 2\tau_0) \propto \Omega \langle S^x \rangle_0 \approx \Omega^2 / 4 k_B T$$

(for $T > T_c$ one has $\langle S^x \rangle_0 \approx \Omega / 4 k_B T$).[4] Upon deuteration of the sample tunneling integral Ω decreases by a factor of 10–100.[3,4] Consequently, the echo signal should decrease on deuteration by a factor of 10^2–10^4, in good agreement with experiment.

Let us summarize the main results of this paper. We have found the effective equations for the envelopes of the forward and backward electroacoustic wave packets. The equations for the envelopes of noninteracting packets yield an expression for the effective damping rate of the electroacoustic waves, with a temperature dependence that agrees with experiment. We have obtained an expression for the amplitude of the electroacoustic echo arising as a result of the parametric interaction of the forward electroacoustic wave and the uniform electric field of the second pulse. We have explained the linear dependence of the echo amplitude on the amplitudes of the alternating fields and durations of the pulses when a ferroelectric is excited by low-intensity pulses. The amplitude of the echo signal becomes saturated as the intensity of the pulses increases. We have accounted for the lack of an echo in the paraphase of a ferroelectric for $E_0 = 0$ and for the strong attenuation of the echo signal upon deuteration of the sample. Thus, we have constructed a theory of the electroacoustic echo that in the main correctly describes the existing experimental data and permits calculation of the characteristics of the electroacoustic echo on the basis of microscopic concepts of ferroelectricity.

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