Breather in a dispersive medium with a resonant two-phonon transition

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We propose a mechanism generating a nonlinear wave in a dispersive medium with a resonant two-phonon transition. We consider an acoustic surface wave propagating in a film-substrate medium. We show that for an acoustic wave with a relatively small energy a breather state can be formed. We indicate the conditions for the realization of this effect.

1. INTRODUCTION

When acoustic waves with sufficiently large amplitude propagate in a nonlinear medium nonlinear acoustic waves may be formed. Nonlinear waves of a stable shape—solitons—are of particular interest among those nonlinear waves. The determination of mechanisms causing the formation of solitons is one of the main problems of the physics of nonlinear waves. Acoustic solitons may be formed in paramagnetic dielectrics through the McCall–Hahn mechanism, i.e., when there is a coherent interaction between acoustic waves and the paramagnetic impurities contained in the medium and the conditions for self-induced transparency (SIT) are satisfied.[1,2] Under well defined conditions there may also appear apart from solitons, pulsating acoustic wave solitons (breathers).[3] Besides this effect acoustic breathers can be formed in dispersive paramagnetic dielectrics when anharmonic crystal lattice vibrations and the coherent interaction of acoustic waves with resonant paramagnetic impurities are simultaneously effective.[4] The physical picture changes if the wave causes a two-phonon excitation of the paramagnetic impurities. Under SIT conditions a soliton is then formed in the form of a 2π pulse with a Lorentz shape.[5,6] The question of whether the excitation of an acoustic breather is possible in that case is an open one.

The main statement of the present paper is that breather states may be formed for an acoustic pulse with a relatively small energy which causes two-phonon excitations of the paramagnetic impurities, if the medium in which the wave propagates is dispersive.

2. DERIVATION OF THE EQUATIONS

We study a mechanism which produces a nonlinear acoustic wave in a dispersive medium with a resonant two-phonon transition using the example of an acoustic surface wave (ASW) propagating in a solid halfspace-solid layer (substrate-film) system. We assume that the substrate—a nonmetallic diamagnetic solid with electron (J) and nuclear (I) spins—occupies the $x<0$ halfspace (for the sake of simplicity we shall assume $J=I=\frac{1}{2}$). Different kinds of ASW can propagate in this system; they differ from one another by the boundary conditions characterizing the state of the wave process at the boundaries of the media.[7] We consider an ASW pulse with vertical polarization of length $T \ll T_{1,2}$, frequency $\omega_{\mathbf{k}}$, and wavevector $\mathbf{k}$ propagating in the direction of the positive z axis (the $T_{1,2}$ are the longitudinal and transverse relaxation times). An external constant magnetic field H_0 is applied in the same direction. If the condition $2\omega_{\mathbf{k}}=\omega_J+\omega_I$ is satisfied the ε_{zz} component of the deformation tensor of the ASW will cause two-phonon transitions in the electron-nuclear spin system of the impurities (ω_J and ω_I are the Zeeman frequencies of the electron and nuclear spins).[8,9] The spin-phonon interaction in this case does not change the nature of the boundary conditions and, hence, the transverse structure of the field and the dispersion law of the ASW are determined in the linear limit.[7] Using an expansion in coherent states of the acoustic field we can write the component of the deformation tensor of the ASW in the following form:[7,10]

$$\varepsilon_{zz}={}^1/_2(\varepsilon^+ +\varepsilon^-),\qquad \varepsilon^\pm=\mp 2i\sum_{\mathbf{k}} k a_{\mathbf{k}}^{\pm} e^{\mp ikz}\varkappa_{\mathbf{k}}\gamma_{\mathbf{k}}(x),$$

where

$$\varkappa_{\mathbf{k}}^2=\hbar/2\omega_{\mathbf{k}}\rho N_0 V,$$

$a_{\mathbf{k}}^+$ and $a_{\mathbf{k}}^-$ are Bose creation and annihilation operators for acoustic surface waves, ρ is the density of the medium, N_0 is the number of sites in the lattice, V is the volume of the medium, and $\gamma_{\mathbf{k}}(x)$ is a function which determines the vertical structure of the field and which depends on the actual shape of the ASW.[7] We shall assume in what follows that we have $\hbar=N_0=V=1$.

The dispersion relation is given by the equation[11]

$$\omega_{\mathbf{k}}^2=c^2k^2[1-(hk)^2], \tag{1}$$

where c is the speed of the linear ASW and the quantity h is determined by the thickness of the film.

The Hamiltonian of this model has the form

$$\mathscr{H}=\mathscr{H}_z+\mathscr{H}_A+\mathscr{H}_{\rm ph}+\mathscr{H}_{\rm sph}, \tag{2}$$

where

$$\mathscr{H}_Z=\omega_J J^z-\omega_I I^z$$

is the Zeeman Hamiltonian,

$$\mathscr{H}_A=A\sum_i \mathbf{J}_i\mathbf{I}_i$$

is the hyperfine interaction Hamiltonian,

$$\mathscr{H}_{\rm ph}=\sum_{\mathbf{k}}\omega_{\mathbf{k}}(a_{\mathbf{k}}^+a_{\mathbf{k}}^- + {}^1/_2)$$

is the phonon system Hamiltonian,

$$\mathscr{H}_{\rm sph}=\beta H_0 F_{zzzzzz}J^z\varepsilon_{zz}^2$$

is the spin-phonon interaction Hamiltonian under conditions when two-phonon transitions can be excited, A is the

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hyperfine interaction constant, β is the Bohr magneton, and F_{zzzzzz} is a component of the spin-phonon interaction tensor.

Performing a canonical transformation using the unitary operator[9]

$$\mathcal{U}=\exp\left(i\int_0^t \mathcal{H}_{sf}(t')dt'\right),$$

we determine the representation in which in the rotating wave approximation the evolution of the system is given by the Hamiltonian

$$\tilde{\mathcal{H}}=\omega_J J^z-\omega_I I^z+A\sum_i J_i^z I_i^z+\sum_{\mathbf{k}}\omega_{\mathbf{k}}(a_{\mathbf{k}}^+a_{\mathbf{k}}^-+{}^1/_2)$$
$$+{}^1/_2 L[J^+I^-(\varepsilon^-)^2+J^-I^+(\varepsilon^+)^2], \tag{3}$$

where we have

$$L=A\beta H_0 F_{zzzzzz}/2\omega_{\mathbf{k}}.$$

Using the standard procedure[3,4] we obtain from the Hamiltonian (3) a set of equations for the averages of the operators of the acoustic field and of the spin variables:

$$\begin{aligned}\ddot{\alpha}_{-\mathbf{k}}^*&=-\omega_{\mathbf{k}}^2\alpha_{-\mathbf{k}}^*+4iLB^+k\varkappa_{\mathbf{k}}\gamma_{-\mathbf{k}}(x)\omega_{\mathbf{k}}e^{-ikz}\varepsilon_{zz},\\ \ddot{\alpha}_{\mathbf{k}}&=-\omega_{\mathbf{k}}^2\alpha_{\mathbf{k}}+4iLB^-k\varkappa_{\mathbf{k}}\gamma_{-\mathbf{k}}(x)\omega_{\mathbf{k}}e^{-ikz}\varepsilon_{zz},\end{aligned} \tag{4}$$

$$\dot{B}^+=i\omega_0B^+-iL(u^+)^2N,\quad \dot{N}={}^1/_2 iL[(u^+)^2B^--(u^-)^2B^+], \tag{5}$$

where

$$B^\pm=\langle J^\pm I^\mp\rangle,\ N=\tfrac{1}{2}\langle(J^z-I^z)\rangle,\quad \alpha_{\mathbf{k}}=\langle\{\alpha_{\mathbf{k}}\}|a_{\mathbf{k}}^-|\{\alpha_{\mathbf{k}}\}\rangle,$$

$$|\{\alpha_{\mathbf{q}}\}\rangle=\prod_{\mathbf{q}}|\alpha_{\mathbf{q}}\rangle,\quad a_{\mathbf{k}}^-|\alpha_{\mathbf{k}}\rangle=\alpha_{\mathbf{k}}|\alpha_{\mathbf{k}}\rangle,$$

$$u^\pm=\langle\varepsilon^\pm\rangle,\quad \omega_0=\omega_J+\omega_I$$

$|\alpha_{\mathbf{k}}\rangle$ is the coherent state vector of the kth mode of ASW, and the symbol $\{\alpha_{\mathbf{k}}\}$ indicates the set of all amplitudes $\alpha_{\mathbf{k}}$.

We multiply Eq. (4) by the quantity $ik\varkappa_{\mathbf{k}}\gamma_{\mathbf{k}}(0)\times\exp(ikz)$ and sum over **k**; as a result we obtain on the boundary of the media for $x=0$ a nonlinear equation for the ASW:

$$\ddot{U}+\frac{1}{2\pi}\int W(x)U(z-x)dx+I_0[U]=0, \tag{6}$$

where we have

$$U=\langle\varepsilon_{zz}\rangle,\quad W(x)=\sum_{\mathbf{k}}\omega_{\mathbf{k}}^2e^{ikx},$$

$$I_0[U]=R^2U(B^++B^-),\quad R^2=\sum_{\mathbf{k}}4Lk^2\varkappa_{\mathbf{k}}^2\omega_{\mathbf{k}}\gamma_{\mathbf{k}}(0)\gamma_{-\mathbf{k}}(0).$$

Apart from the notation this equation is valid also in the case when the ASW causes two-phonon transitions of only the electron spins of the paramagnetic impurities with $J=1$. The interaction between the ASW and the paramagnetic impurities will then change the boundary conditions at the boundary between the film and the substrate for $x=0$. This is caused by the fact that two-phonon transitions in impurities here are caused by the ε_{xz} component of the deformation tensor of the ASW. The change in the boundary conditions can reflect differently on the nonlinear wave process. In particular, in the effects studied in Ref. 12 the change in the boundary conditions caused by the spin-phonon interaction are of a very significant nature and lead to a qualitative change in the physical picture of the wave process, but in some situations[13] such a change is not fundamental. When the ASW causes two-phonon excitations of the paramagnetic impurities with $J=1$ spins we shall follow Ref. 13 and also approximately use the linear boundary conditions and, hence, the results obtained in the present paper remain valid also for that case.

3. ASW BREATHER STATE

Using the slowly changing profile method we can significantly simplify the set of Eqs. (5) and (6). To do this we write U in the form

$$U=\frac{1}{2}\sum_l Z_l\mathscr{E}_l, \tag{7}$$

where we have $Z_l=\exp[il(kz-\omega_{\mathbf{k}}t)]$, and $\mathscr{E}_l$ is the slowly changing amplitude of the acoustic wave. As the quantity U is real it follows that $\mathscr{E}_l=\mathscr{E}^*_{-l}$.

Bearing in mind that the ASW causes two-phonon excitations of the paramagnetic impurities and that in the simplest case we consider there are only two energy levels, we can write the average magnetization of the paramagnetic impurities in the form

$$B^++B^-=\sum_l P_{-l}Z_l(\delta_{l,2}+\delta_{l,-2}). \tag{8}$$

When the ASW interact with paramagnetic impurities the most characteristic features of the nonlinear phenomena usually show up when the condition for exact resonance, $2\omega_{\mathbf{k}}=\omega_0$, is satisfied. We therefore consider just that case. We assume here that all paramagnetic impurities before the ASW pulse enters the medium are in the ground state, i.e., that we have $N_{\text{init}}=-\frac{1}{2}$ for $t\to-\infty$.

We note that when there are no phase changes in the wave, i.e., when the conditions $\mathscr{E}_l=\mathscr{E}^*_{-l}=\mathscr{E}_{-l}$ are satisfied, substitution of Eqs. (7) and (8) into the set of Eqs. (5) and (6) in a nondispersive medium, when $h\to 0$, leads to the well known soliton solution in the form of a 2π pulse with a Lorentz shape.[5,6]

We consider in the present paper the solution of the set of Eqs. (5) and (6) in the case $|\Theta_l|\ll 1$ where the quantity

$$\Theta_l(z,t)=\frac{L}{2}\int_{-\infty}^t \mathscr{E}_l^2(z,t')dt'$$

is proportional to the pulse energy. We use the reductive perturbation method, developed in Ref. 14, according to which we can write the quantity $\mathscr{E}_l$ as follows

$$\mathscr{E}_l(z,t)=\sum_{\alpha=1}^\infty\sum_{n=-\infty}^\infty \varepsilon^\alpha Y_n\varphi_{l,n}^{(\alpha)}(\zeta,\tau)=\sum_{\alpha=1}^\infty\varepsilon^\alpha\mathscr{E}_l^{(\alpha)}, \tag{9}$$

where we have

$$Y_n = e^{in(Qz-\Omega t)}, \quad \zeta = \varepsilon Q(z - v_g t), \quad \tau = \varepsilon^2 t, \quad v_g = \frac{\partial \Omega}{\partial Q},$$

and ε is a small parameter determining the degree of nonlinearity. Such a representation enables us to split off from $\mathscr{E}_l$ the even more slowly changing quantity $\varphi_{l,n}^{(\alpha)}(\zeta,\tau)$. Hence we assume that the quantities $\varphi_{l,n}^{(\alpha)}$, Ω, and Q satisfy the inequalities

$$\omega_k \gg \Omega, \quad k \gg Q, \quad |\dot{\varphi}_{l,n}^{(\alpha)}| \ll \Omega |\varphi_{l,n}^{(\alpha)}|, \quad \left|\frac{\partial \varphi_{l,n}^{(\alpha)}}{\partial z}\right| \ll Q |\varphi_{l,n}^{(\alpha)}|. \tag{10}$$

We substitute the expansions (7) to (9) into Eq. (6) and use the fact that from the set of Eqs. (5) we find that

$$P_{\pm 2} = \pm \varepsilon^2 i \frac{L n_0}{2} \int_{-\infty}^{t} (\mathscr{E}_{\mp 1}^{(1)})^2 \, dt' + O(\varepsilon^4).$$

Using the inequalities (10) we can transform Eq. (6) to the following form:

$$\sum_{n=-\infty}^{+\infty} Y_n \Bigg\{ \sum_{\alpha=1}^{\infty} \varepsilon^\alpha [W_{\pm 1,n} + \varepsilon J_{\pm 1,n} \partial_\zeta + \varepsilon^2 H_{\pm 1,n} \partial_{\zeta\zeta}{}^2 + \varepsilon^2 h_{\pm 1,n} \partial_\tau] \varphi_{\pm 1,n}^{(\alpha)} + \varepsilon^3 \frac{R^2 L n_0}{2\Omega} \sum_{n',n''} \frac{1}{n-n'} \varphi_{\pm 1,n-n'-n''}^{(1)} \varphi_{\pm 1,n'}^{(1)} \varphi_{\pm 1,n''}^{(1)} + O(\varepsilon^4) \Bigg\} = 0,$$

$$l = \pm 1, \tag{11}$$

where we have

$$W_{l,n} = W_l - 2ln\omega_k\Omega + nQA_l - n^2\Omega^2 + n^2Q^2C_l,$$
$$J_{l,n} = i(2l\omega_k Q v_g - A_l Q + 2n\Omega Q v_g - 2nQ^2 C_l),$$
$$H_{l,n} = Q^2(v_g^2 - C_l), \quad h_{l,n} = -2i(l\omega_k + n\Omega),$$
$$W_l = \omega_{kl}{}^2 - l^2\omega_k{}^2, \quad A_l = \frac{1}{k}\frac{\partial \omega_{kl}{}^2}{\partial l}, \quad C_l = \frac{1}{2k^2}\frac{\partial^2 \omega_{kl}{}^2}{\partial l^2}. \tag{12}$$

To determine the quantities $\varphi_{l,n}^{(\alpha)}$ in Eq. (11) we separately set equal to zero the terms with the same powers of ε. As a result we obtain

$$W_{\pm 1,n} \varphi_{\pm 1,n}^{(1)} = 0, \tag{13}$$

$$W_{\pm 1,n} \varphi_{\pm 1,n}^{(3)} + J_{\pm 1,n} \partial_\zeta \varphi_{\pm 1,n}^{(2)} + H_{\pm 1,n} \partial_{\zeta\zeta}{}^2 \varphi_{\pm 1,n}^{(1)} + h_{\pm 1,n} \partial_\tau \varphi_{\pm 1,n}^{(1)} + \frac{R^2 n_0 L}{2\Omega} \sum_{n',n''} \frac{1}{n-n'} \varphi_{\pm 1,n-n'-n''}^{(1)} \varphi_{\pm 1,n'}^{(1)} \varphi_{\pm 1,n''}^{(1)} = 0. \tag{14}$$

It follows from the dispersion relation (1) that in dispersive media we have $W_0 = W_{\pm 1} = 0$. Hence it follows from (13) that from all quantities $\varphi_{\pm 1,n}^{(1)}$ only the terms $\varphi_{\pm 1,\pm 1}^{(1)}$ are nonvanishing ($\varphi_{\pm 1,+1}^{(1)} = \varphi_{-1,-1}^{(1)*}$) and the relation between the quantities Ω and Q is then determined from the equations

$$2\omega_k\Omega \mp QA_{\pm 1} + \Omega^2 - Q^2C_{\pm 1} = 0, \quad l, n = \pm 1. \tag{15}$$

Comparing Eqs. (12) to (15) we can prove that the following relations hold:

$$J_{\pm 1,\pm 1} = 0, \quad H_{\pm 1,\pm 1} = 3c^4k^4Q^2h^2/(\omega_k + \Omega)^2,$$
$$h_{\pm 1,\pm 1} = \mp 2i(\omega_k + \Omega), \quad v_g = (2QC_{\pm 1} \pm A_{\pm 1})/2(\omega_k + \Omega). \tag{16}$$

Note that Eqs. (13) to (16) are obtained from Eq. (11) by expanding in powers of ε up to third order. One checks easily that an expansion to higher powers of ε does not lead to relations which are independent of (13)–(16).

Substituting (16) into (14) and using (15) we get in the variables $y = z - v_g t$, t an equation for the quantities $\chi_\pm = \varepsilon \varphi_{\pm 1,\pm 1}^{(1)}$:

$$\mp i\partial_t \chi_\pm + P\partial_{yy}{}^2 \chi_\pm + q\chi_\pm |\chi_\pm|^2 = 0, \tag{17}$$

where we have

$$P = \frac{3c^4k^4h^2}{2(\omega_k + \Omega)^3}, \quad q = \frac{R^2 n_0 L}{8\Omega(\omega_k + \Omega)}.$$

This is the well known nonlinear Schrödinger equation (NLS) which for $Pq > 0$ has a soliton solution.[15] It is clear from the expressions for P and q that this inequality is satisfied. We consider a single-soliton solution of the NLS

$$\chi_\pm = \mp 2i\eta \exp[\pm i\Phi] \cosh^{-1} 2\eta f, \tag{18}$$

where we have

$$\Phi = 2\xi\mu z - 2[\xi\mu v_g + 2(\eta^2 - \xi^2)q]t - \varphi_0,$$
$$f = \mu z + (4\xi q - \mu v_g)t - Y_0,$$
$$\mu = \left(\frac{q}{P}\right)^{1/2}, \quad \varphi_0 = \arg b(0), \quad Y_0 = \frac{1}{2\eta} \ln |b(0)|,$$

ξ, η, and b are the scattering data for the soliton, η determines the soliton amplitude and ξ its velocity $v = -4\xi$.

Substituting the solution (18) into (9) and using (10) we obtain for the quantities $\mathscr{E}_{\pm 1}$

$$\mathscr{E}_{\pm 1} = \mp 2i\eta \exp[\pm i(\Phi + Qz - \Omega t)] \cosh^{-1} 2\eta f. \tag{19}$$

The occurrence of the factor $\exp[\pm i(Qz - \Omega t)]$ in this expression indicates the appearance of periodic "beats" in position and time, which are slow compared to $\exp[\pm i(kz - \omega_k t)]$, with characteristic parameters Q and Ω; as a result the soliton solution (18) is transformed into the solution (19) which has the shape of a breather.

4. DISCUSSION OF THE RESULTS

When a pulse propagates in a dispersive medium its shape changes—the width of the pulse increases while it propagates if $\partial^2\omega_k/\partial k^2 \neq 0$. This is connected with the fact that in a dispersive medium waves with different wavelengths propagate with different velocities. In the NLS equation the dispersive effects are taken into account by the term $P\partial_{yy}^2\chi_\pm$.

On the other hand, nonlinear effects caused by the coherent nonlinear two-phonon interaction of the wave with the paramagnetic impurities lead to a progressive deformation of the initial pulse profile. In the NLS equation the nonlinear effects are taken into account by the term $q\chi_\pm |\chi_\pm^2|^2$. As a result of the competition between the nonlinear effects which increase the torsion of the pulse profile and the dispersion effect, due to which the profile gets smeared out, the shape of the nonlinear wave stabilizes—an ASW breather state is formed.

The condition for a balance between the dispersive and nonlinear effects in the particular case when for $t=0$ the pulse $|\chi_-(z,0)|$ has a rectangular shape of amplitude H and length L can be written as

$$H\Lambda\mu\sim 2^{1/2},\quad 4HT(Pq/2)^{1/2}\sim\Lambda. \tag{20}$$

The solution (19) can occur in a dispersive medium in which h is sufficiently large so that condition (20) is satisfied. Such a situation is realized when the dispersion is "external" and caused by the presence of a film on the surface of the substrate, i.e., for ASW. One can determine from (20) the (order of magnitude of the) film width necessary for the realization of the ASW breather (19). In the continuous-medium approximation,[16] i.e., when we neglect the discrete structure of the medium, we have $h\to 0$ for acoustic bulk waves and, hence, the proposed mechanism for the formation of a nonlinear wave is not realized.

We have thus shown that in dispersive media (e.g., in a film-substrate system) containing paramagnetic impurities a nonlinear wave in the shape of an ASW breather can be formed in the case of the propagation of an ASW of sufficiently small energy, $|\Theta_l|\ll 1$, which is able to cause two-phonon excitations of the impurities. The explicit shape of this wave is for $x=0$ given by Eq. (19) and the transverse structure of the field is determined by the function $\gamma_{\mathbf{k}}(x)$. The dispersion law and the connection between the quantities Ω and Q are given, respectively, by Eqs. (1) and (15). Phase modulation results.

We note that the results given here are valid for pulses with a sufficiently smooth envelope, provided that the pulse is large compared to the wavelength, i.e., $\Lambda k\gg 1$. Moreover, the length of the breather must be significantly larger than the characteristic length over which the periodic "beats" change: $\Lambda Q\gg 1$. These conditions are satisfied for the solution (18) but are not satisfied for other solutions. It is, for instance, well known[17] that Eq. (17) contains, apart from (18), also N-soliton solutions, the behavior of which is more complicated. In particular, for the many-soliton solutions of the NLS characteristic oscillations of the envelope and strong compression of the peaks of the pulse occur in the initial stage of wave propagation. Under these conditions we cannot apply the slowly varying envelope approximation (7) and even less (9)—the splitting off from $\mathscr{E}_l$ of the even more slowly varying quantity $\varphi_{l,n}^{(\alpha)}$. Hence, the scheme for above such solutions proposed is invalid and for their study we need a completely different approach (see, e.g., Ref. 17).

We note that the results given above are valid only in the case when the spin-phonon interaction constant L is not too small. In the opposite case one must use stronger pulses, which leads to the necessity of taking into account anharmonic crystal lattice vibrations.[4]

In the present paper we have considered the case when we have exact resonance, $2\omega_{\mathbf{k}}=\omega_0$, and uniform broadening of the spectral line. It is not difficult to generalize to the $2\omega_{\mathbf{k}}\neq\omega_0$ case and nonuniform broadening of the spectral line. It is clear that in that case one should not expect results which are qualitatively new compared to ones given here.

One can find characteristic values of the parameters of the acoustic field and of the medium necessary for an experimental observation of the effects discussed above in Refs. 2 and 4. As substance it is advisable to use a crystal of the CaF_2 fluorite with U^{4+} impurities in which two-phonon resonance transitions have been found[18] and also the effect of acoustic SIT under conditions of single-phonon excitations of the impurities.[2]

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