Nonlinear waves in molecular crystals with quantum-tunneling states

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The dynamics of a crystal whose every unit cell contains a two-level center is investigated theoretically. The interaction between the tunneling states and the vibrations of the crystal lattice is described within the simplest model. The ground state of such a crystal is studied and it is shown that depending upon the magnitude of the interaction constant g, two distinct types of ground states are possible. The interaction constant g is a bifurcation parameter. If g reaches a certain critical value g_c a change of the ground state and a rearrangement in the small-vibration spectrum will occur. Nonlinear properties of the quantum crystal are studied and one-parameter soliton excitations and their energy spectrum are described. It is remarked that algebraic solitons can also exist besides dynamical solitons. The Hamiltonian equations of motion are given for the solitons and their dynamics is studied.

1. INTRODUCTION

Phenomena caused by the quantum nature of atomic motions must play an important role in molecular crystals at very low temperatures. Occurrence of tunneling states in each unit cell of a molecular crystal could serve as a clear example of such phenomena. The ammonia molecule NH_3 is an often exploited (in both meanings of the word) example of a molecule with tunneling states. This molecule has two classically equivalent states which differ in that the N atom is placed on symmetrically opposite sides of the plane defined by the three H atoms. The two states are separated by a potential barrier. Tunneling through this barrier leads to a splitting of the ground-state energy level. The two tunneling states arise in free NH_3 molecules and their energy difference is $\hbar\omega_0 \approx 10^{-4}$ eV ≈ 1 K. Analogous tunneling states occur in N_2, O_2, CH_4, and other molecules in which case the energy difference between the tunneling states is either comparable to or significantly smaller than the $\hbar\omega_0$ given for an NH_3 molecule. This means that the interesting effects caused by the tunneling states should occur at low or super-low temperatures.

We shall study the long-wavelength, macroscopic excitations of a molecular crystal whose every unit cell contains tunneling states with two nondegenerate levels separated by the energy gap $\hbar\omega_0$. In the theory of the interaction of a two-level system with an external field[1] the states of a molecule corresponding to the energies $E = 0$ and $E = \hbar\omega_0$ are often considered as the eigenstates of the Pauli matrix operator σ_3. Then, the characteristic Hamiltonian of a single molecule can be written as

$$H_0 = {}^1/_2\hbar\omega_0(\sigma_3 + 1). \tag{1}$$

As a rule, the direct quantum interaction between the tunneling states of molecules in the neighboring unit cells is small and it is not taken into account. An interaction is also mediated by electromagnetic and elastic fields. The coupling between the tunneling states and the long-wavelength elastic vibrations of the crystal lattice will be analyzed in the present work.

A consideration of physical quantities averaged over volumes containing large numbers of molecules appears necessary in the long-wavelength approximation. We shall consider the operator σ_3 as the corresponding component of the vector operator $\boldsymbol{\sigma}$ (σ_1, σ_2, σ_3) and we shall denote by $\mathbf{S}$ the average value per unit volume of the operator $\boldsymbol{\sigma}$. This vector is usually called the energy spin and it is defined so that its square equals unity: $\mathbf{S}^2 = 1$. Thus, the self-energy density w_0 of the tunneling states which corresponds to the single-molecule Hamiltonian (1) is

$$w_0 = {}^1/_2\hbar\omega_0 n(S_3 + 1), \tag{2}$$

where n is the number of molecules per unit volume ($n = 1/a^3$, a^3 being the unit cell volume). It is clear that the sum $(S_3 + 1)/2$ determines the relative population of the upper level.

Within the simplest model, the energy spin interacts only with the isotropic crystal compression (or expansion) and the interaction Hamiltonian of a molecule and the elastic field is

$$H_i = {}^1/_2 g\sigma_1 \operatorname{div} \mathbf{u}, \tag{3}$$

where $\mathbf{u}$ is the elastic displacement vector, g is the interaction constant (generally speaking, $g < \hbar\omega_0$). This Hamiltonian describes the transitions between the two quantum states. The energy density

$$w_i = {}^1/_2 gnS_1 \operatorname{div} \mathbf{u} \tag{4}$$

corresponds to the Hamiltonian (3). A deformation of the crystal lattice also inevitably influences the energy difference $\hbar\omega_0$ between the two quantum states, as considered, for example, in Ref. 2. Although this effect can be easily taken into account, it does not lead to qualitative changes in our conclusions that the interaction between the elastic waves and the tunneling states in nonlinear. Therefore, when describing nonlinear vibrations of a crystal with tunneling states we shall start from the expressions (2) and (4).

The spectrum of small (harmonic) crystal vibrations will be discussed in Sec. 2, and it will be shown that in the

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presence of the tunneling states a gap in the frequency (energy) spectrum appears. The size and the place of the gap depend on the parameter g. If this parameter is sufficiently small the gap will deform the acoustic (phonon) spectrum of the crystal only slightly. However, for certain values of g the deformation of the spectrum is so significant that a change in the crystal ground state is necessary: a spontaneous deformation of the crystal occurs and with it a finite population of the upper level of the tunneling states. It appears that the parameter g takes the role of a bifurcation parameter whose different values correspond to different ground states of the crystal.

Nonlinear soliton excitations which affect both the elastic deformation of the crystal and the population of the upper level of the tunneling states are analyzed in Sec. 3. These are the one-parameter solitons whose dynamics is characterized by only one quantity, their speed, which determines the magnitude and the degree of localization of the excitation.

The dynamics of one-parameter solitons is studied in Sec. 4, where a dependence of the soliton energy on its momentum is found and it is shown that the solitons can be treated as collective excitations of the crystal. The soliton energy spectrum occupies the energy interval forbidden for the harmonic vibrations of the quantum crystal.

2. GROUND-STATE BIFURCATION AND THE SPECTRUM OF WEAK (LINEAR) EXCITATIONS

The equations of motion for the components of the energy spin vector **S** can be obtained by averaging corresponding operator equations for the Pauli matrices,

$$-i\hbar\frac{\partial\boldsymbol{\sigma}}{\partial t}=[H_0+H_i,\boldsymbol{\sigma}].$$

Elementary calculations lead to the equations

$$\begin{aligned}&\frac{\partial S_1}{\partial t}=-\omega_0 S_2,\quad \frac{\partial S_2}{\partial t}=\omega_0 S_1-\frac{g}{\hbar}S_3\operatorname{div}\mathbf{u},\\&\frac{\partial S_3}{\partial t}=\frac{g}{\hbar}S_2\operatorname{div}\mathbf{u}.\end{aligned}\tag{5}$$

The equations (5) are analogous to the Bloch equations for the magnetic resonance spin precession.[3,4] We shall supplement them with the equations for the crystal lattice vibrations in the long-wavelength limit.

$$\frac{\partial^2\mathbf{u}}{\partial t^2}=c_t^2\frac{\partial^2\mathbf{u}}{\partial x_k^2}+(c_l^2-c_t^2)\operatorname{grad}\operatorname{div}\mathbf{u}+\frac{g}{2m}\operatorname{grad}S_1,\tag{6}$$

where x_k, $k = 1, 2, 3.$ are the spatial coordinates, c_l and c_t are the speeds of the longitudinal and transverse acoustic waves, respectively, and m is the mass of the unit cell. It is clear that the system of equations (6) separates into two systems, one of which describes the longitudinal elastic vibrations,

$$\frac{\partial^2(\operatorname{div}\mathbf{u})}{\partial t^2}=c_l^2\frac{\partial^2(\operatorname{div}\mathbf{u})}{\partial x_k^2}+\frac{g}{2m}\frac{\partial^2 S_1}{\partial x_k^2},\tag{7}$$

while the other describes the transverse elastic vibrations,

$$\frac{\partial^2(\operatorname{rot}\mathbf{u})}{\partial t^2}=c_t^2\frac{\partial^2(\operatorname{rot}\mathbf{u})}{\partial x_k^2}.\tag{8}$$

Since in the given model the transverse elastic vibrations do not interact with the tunneling states, we shall not consider them in the following. In that case, the elastic energy density can be simplified,

$$w_{\rm el}={}^1/{}_2\rho(\partial\mathbf{u}/\partial t)^2+{}^1/{}_2\rho c^2(\operatorname{div}\mathbf{u})^2,\tag{9}$$

where $\rho = mn \equiv m/a^3$ and $c = c_l$ is the speed of the longitudinal acoustic vibrations. Hence, we shall represent the total energy of a vibrating crystal with the tunneling states as

$$\begin{aligned}E=&\frac{1}{2}n\int\{\hbar\omega_0(S_3+1)+gS_1\operatorname{div}\mathbf{u}\}d^3x\\&+\frac{1}{2}\rho\int\left\{\left(\frac{\partial\mathbf{u}}{\partial t}\right)^2+c^2(\operatorname{div}\mathbf{u})^2\right\}d^3x.\end{aligned}\tag{10}$$

Clearly, the energy (10) is invariant with respect to a simultaneous sign change in S_1 and div **u**.

Equations (5) and (7) contain the trivial solutions

$$\operatorname{div}\mathbf{u}=0,\quad S_1=S_2=0,\quad S_3=-1,\tag{11}$$

corresponding to the absence of a lattice deformation and to all two-level centers being in the ground state. The energy of such a state is $E_0 = 0$.

There is another static (equilibrium) solution of the equations (5) and (7):

$$S_1=\frac{g}{\hbar\omega_0}S_3\operatorname{div}\mathbf{u},\quad S_2=0,\quad S_3=\text{const},\quad \operatorname{div}\mathbf{u}=\text{const}.\tag{12}$$

The solution (12) is consistent with (7) if

$$\operatorname{div}\mathbf{u}=\pm\varepsilon_0,\quad \varepsilon_0=\frac{\hbar\omega_0 g}{g_c^2}\left[1-\left(\frac{g_c}{g}\right)^4\right]^{1/2},\quad S_3=-\left(\frac{g_c}{g}\right)^2,\tag{13}$$

where $g_c^2 = 2mc^2\hbar\omega_0$. Essentially, there are two solutions of the type (12), one of which corresponds to a static expansion of the crystal (div $\mathbf{u} > 0$), while the second corresponds to a compression (div $\mathbf{u} < 0$). The states (12) are possible only for sufficiently large values of g,

$$|g|>g_c\equiv c(2m\hbar\omega_0)^{1/2}.\tag{14}$$

The energy of the crystal in the states (12) equals

$$E_s=-{}^1/{}_4N\hbar\omega_0[(g_c/g)-(g/g_c)]^2,\tag{15}$$

where N is the number of sites of the crystal lattice. The sign of the parameter g does not influence the existence of this state—it only determines the relative sign between div **u** and S_1.

Comparing the energies of the two equilibrium states of the crystal, (11) and (12), we find

$$E_s\leqslant E_0\quad\text{for}\quad |g|\geqslant g_c,\tag{16}$$

in which case the equality of the energies is possible only in the case where $|g| = g_c$. The inequality (16) is consistent with the condition (14) for the existence of the second equilibrium state.

Therefore, for small values of the interaction parameter, $|g| < g_c$, the ground state is the undeformed lattice (div

$\mathbf{u} = 0$) with $S_3 = -1$. As the parameter $|g|$ exceeds the critical value g_c a second (competing), doubly degenerate equilibrium state with a uniform deformation of the lattice (div $\mathbf{u} = \pm \varepsilon_0$) and $S_3 = -(g_c/g)^2$ occurs. In this fashion we have clarified that the interaction parameter g is a bifurcation parameter and that its value $|g| = g_c$ is the ground-state bifurcation point. After the bifurcation point $|g| = g_c$ the ground state is the state with a spontaneous deformation of the crystal lattice.

By linearizing (5) near the ground states (11) or (12) we obtain equations for the small vibrations which realize the states of the weak excitations of the crystal. The relationship between the frequency ω of an elementary excitation and its wave vector $\mathbf{k}$ is given by the dispersion law

$$(\omega^2-c^2k^2)(\omega^2-\omega_0{}^2)=(\omega_0ck)^2(g/g_c)^2, \quad |g|\leqslant g_c, \tag{17}$$

$$(\omega^2-c^2k^2)\left(\omega^2-\omega_0{}^2-\frac{g^2}{\hbar^2}\varepsilon_0{}^2\right)=(\omega_0ck)^2, \quad |g|\geqslant g_c, \tag{18}$$

where ε_0 is given by the expression (13).

Let us analyze the dispersion law (17). For $g = 0$ we would have independent longitudinal vibrations of the crystal with the dispersion law $\omega = ck$ and an excitation of the two-level systems with the frequency $\omega = \omega_0$. The inclusion of the interaction between the phonons and the eigenmodes of the two-level molecules leads to the dispersion law (17). For $\omega \ll \omega_0$ we have $\omega = c_* k$ where $c_* = c[1-(g/g_c)^2]^{1/2}$, while for $\omega \gg \omega_0$ we have $\omega = ck$. For $\omega \sim \omega_0$, as a result of the coupling between the phonons and the two-level states, a significant rearrangement of the spectrum occurs. A new characteristic frequency $\omega_* = \omega_0[1-(g/g_c)^2]^{1/2}$ appears in the system. The formation of a gap in the frequency spectrum for the harmonic crystal vibrations occurs as a consequence of the interaction between the elastic vibrations and the two-level molecules. For small values of the coupling parameter, $|g| \leqslant g_c$, the gap $\Delta\omega = \omega_0 - \omega_*$ increases with g from the value $\Delta\omega = 0$ for $g = 0$ to its maximal value $\Delta\omega = \omega_0$ for $|g| = g_c$. A plot of the dispersion law (17) is given in the left-hand portion of Fig. 1. As follows from the reality of the frequency ω_*, the dispersion law (17) can be realized only for $|g| \leqslant g_c$. In this interval ω_* is always smaller than the eigenfrequency of the two-level states.

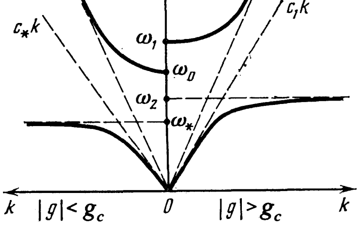

FIG. 1. The dispersion law for small vibrations of a quantum molecular crystal: $c_1 = c[1-(g_c/g)^4]^{1/2}$.

If the bifurcation parameter g takes the value $|g| = g_c$, then the dispersion laws (17) and (18) degenerate into

$$\omega=0, \quad \omega^2=\omega_0{}^2+c^2k^2. \tag{19}$$

Consequently, the low-frequency branch of the longitudinal acoustic vibrations disappears. This means that in the linear approximation with respect to the deformations the crystal offers no resistance to isotropic compression (expansion), which is being compensated by the appearance of a uniform deviation of the vector $\mathbf{S}$ from its equilibrium direction. Indeed, for $|g| = g_c$ a static change of the full energy density for $S_2 = 0$ and $S_1 \ll 1$ can be written as

$$w={}^1/_2\{\hbar\omega_0(S_3+1)+gS_1\operatorname{div}\mathbf{u}$$
$$+mc^2(\operatorname{div}\mathbf{u})^2\}={}^1/_4\{(\hbar\omega_0)^{1/2}S_1 \pm (2mc^2)^{1/2}\operatorname{div}\mathbf{u}\}^2. \tag{20}$$

It follows from (20) that for arbitrarily small div $\mathbf{u}$ a definitive value of S_1 can be determined such that the crystal energy is unchanged ($w = 0$). Of course, the corresponding possibility of populating the upper quantum energy level follows also from the consideration (in the linear approximation) of the equations (5) and (7).

Let us now consider the dispersion law (18) which is realized for $|g| \geqslant g_c$ (its plot is shown in the right-hand portion of Fig. 1). Characteristic frequencies ω_1 and ω_2 are determined by the relations $\omega_1 = \omega_0(g/g_c)^2$, $\omega_2^2 = \omega_1^2 - \omega_0^2$. The frequency ω_1 is greater than the eigenfrequency ω_0 of the two-level states for all values of $|g| > g_c$. The width of the gap $\Delta\omega = \omega_1 - \omega_2$ decreases with an increase in g and for $|g| \gg g_c$ we have $\Delta\omega \approx (\omega_0/2)(g_c/g)^2$.

3. NONLINEAR LOCALIZED EXCITATIONS. ONE-PARAMETER SOLITONS

In addition to the quasiparticles with the dispersion laws (17) and (18), excitations of another type are possible within the studied system. These excitations are the solitons which are spatially localized, traveling, nonuniform distributions of the vectors $\mathbf{S}$ and $\mathbf{u}$.

We shall seek localized solutions to the equations (5) and (7) in the form of functions which depend only on one coordinate (x) and the time (t) through the difference $\xi = x - Vt$, where V is the propagation velocity of the excitation. Note that at infinity the energy spin vector and div $\mathbf{u}$ take on their equilibrium values. Since different equilibrium states (11) and (12) can be realized in the studied system depending on the magnitude of the bifurcation parameter g, the following analysis must be conducted separately for the small ($|g| \leqslant g_c$) and the large ($|g| \geqslant g_c$) value of g.

We begin with the case $|g| \geqslant g_c$. Then, $S_1 = S_2 = 0$, $S_3 = -1$, and div $\mathbf{u} = 0$ at infinity. By taking this into account, integration of the equation (7) gives a simple relation between S_1 and $\varepsilon = \partial u_x/\partial x$:

$$S_1=-\frac{2m}{g}(c^2-V^2)\varepsilon. \tag{21}$$

Equations (5) and (21) imply the nonlinear equation

$$\partial^2\varepsilon/\partial\xi^2-\varkappa^2\varepsilon+{}^1/_2(g/\hbar V)^2\varepsilon^3=0, \tag{22}$$

where

$$\varkappa^2=\frac{\omega_0{}^2}{V^2}\left(\frac{V^2-c_*{}^2}{c^2-V^2}\right).$$

Localized solutions of the equation (22) are the functions

$$\varepsilon = \pm 2\hbar |V| \varkappa / g \operatorname{ch} \varkappa\xi, \tag{23}$$

while the corresponding soliton solutions to the equation (5) are

$$S_1 = \mp \frac{2(V^2 - c_*^2)\omega_0}{\varkappa |V| c_0^2 \operatorname{ch} \varkappa\xi}, \quad S_2 = \pm \frac{2(V^2 - c_*^2)}{c_0^2} \frac{\operatorname{sh} \varkappa\xi}{\operatorname{ch}^2 \varkappa\xi},$$
$$S_3 = \frac{2(V^2 - c_*^2)}{c_0^2} \frac{1}{\operatorname{ch}^2 \varkappa\xi} - 1, \quad c_0^2 = c^2 \left(\frac{g}{g_c}\right)^2 \leqslant c^2. \tag{24}$$

Since the localized solutions are possible only for a real $\varkappa$ the speed of the solitons must lie in the interval

$$c_*^2 \equiv c^2 (1 - g^2/g_c^2) \leqslant V^2 \leqslant c^2. \tag{25}$$

In this way the soliton speed is necessarily greater than the maximal phase velocity of the "slow" linear elastic waves, but it is smaller than the speed of the longitudinal sound in the crystal.

We observe that the phase of a harmonic plane wave can always be represented as

$$kx - \omega t = k(x - \omega k^{-1} t) = k(x - V_p t),$$

where the phase velocity $V_p = \omega/k$ determines the slopes of the rays passing from the origin of Fig. 1 to the dispersion curve for the linear vibrations. These rays pass either below the straight line $\omega = c_* k$ or above the line $\omega = ck$, avoiding the "forbidden" region between them. Possible soliton speeds fill this region. As a result, the set of collective excitations of a crystal is enlarged and the elementary motions excluded from the linear theories are taken into consideration.

The structure of the nonlinear equation (22) is such that the solutions which vanish at infinity can have either sign. Either a compression wave ($\varepsilon < 0$) or an expansion wave ($\varepsilon > 0$) is possible. The choice of the sign is determined by the boundary condition at infinity for the component u_x of the displacement. Indeed, it follows from (23) that

$$u_x = \pm (2\hbar |V|/g) \operatorname{arctg} \operatorname{sh} (\varkappa\xi/2) + {}^1/_2 u_0,$$

where $u_0 = \text{const}$. We shall consider an uncompressed crystal at $x = +\infty$ and a soliton traveling from left to right ($V > 0$). Then,

$$u_x = \pm \frac{2\hbar V}{g} \left(\operatorname{arctg} \operatorname{sh} \frac{\varkappa\xi}{2} - \frac{\pi}{2} \right). \tag{26}$$

The function (26) describes a kink which travels with the speed V and which sharpens rapidly as the limiting value c is approached. Such a nonlinear wave gives rise to a quite specific displacement at $x = -\infty$, namely

$$u_0 = u_x(-\infty) = \mp (2\pi\hbar/g) V. \tag{27}$$

The sign of this displacement chooses one of the possible solitons.

The long-wavelength approximation which we employed requires the inequality $\varkappa a \ll 1$, which is equivalent to the condition

$$[(V^2 - c_*^2)/(c^2 - V^2)]^{1/2} \ll V/a\omega_0. \tag{28}$$

However, precisely because of this long-wavelength approximation it is necessary that $a(\omega_0/V) \ll 1$, so that the condition (28) does not bring additional essential limitations on the soliton speed. The only condition is that the speed V should not be too close to c.

The dependence of S_3 (population of the upper state) upon the soliton speed is interesting. For the minimal speed $V = c_*$ the soliton is completely delocalized ($S_3 = -1$, $\varkappa = 0$). For intermediate speeds (25) $S_3(0) + 1 = 2(V^2 - c_*^2)/c_0^2$ at the center of the soliton. In principle, the maximal value $S_3(0) = 1$ is reached for $V = c$. However, we have seen that a soliton resulting from the equations in the long-wavelength approximation cannot have such a speed.

The described behavior of S_3 is reminiscent of the order parameter behavior of the "dark soliton" which arises in a nonideal Bose gas.[5]

Let us turn to the case $|g| \geqslant g_c$ where the boundary conditions at infinity correspond to the equilibrium states (12) and (13). To be specific, we shall choose the upper sign and we shall introduce the new variables which go to zero at infinity:

$$\mathbf{s} = \mathbf{S} - \mathbf{S}^0, \quad \zeta = \varepsilon - \varepsilon_0.$$

The relationship

$$s_1 = -(2m/g)(c^2 - V^2)\zeta \tag{29}$$

is preserved for the variables determined in this fashion. Nevertheless, a nonlinear equation obtained from (5) and (29) is different from (22). Namely,

$$\frac{\partial^2 \zeta}{\partial \xi^2} - \varkappa^2 \zeta + \frac{1}{2}\left(\frac{g}{\hbar V}\right)^2 \zeta^3 + \frac{3}{2}\left(\frac{g}{\hbar V}\right)^2 \varepsilon_0 \zeta^2 = 0, \tag{30}$$

where

$$\varkappa^2 = \left(\frac{g}{g_c}\right)^2 \frac{\omega_0^2}{V^2} \left(\frac{V^2 - c_1^2}{c^2 - V^2}\right), \quad c_1 = c[1 - (g_c/g)^4]^{1/2}.$$

Equation (30) has two types of localized solutions (solitons):

$$\zeta = \frac{2\hbar |V| \varkappa}{g(\eta \pm (1 + \eta^2)^{1/2} \operatorname{ch} \varkappa\xi)}, \quad \eta = \frac{g\varepsilon_0}{\hbar |V| \varkappa}. \tag{31}$$

Henceforth, the upper and lower signs will be associated and the first and second soliton types, respectively.

Solitons are usually characterized by an exponential decay of the corresponding dynamical field far from the center of the soliton. Nonetheless, the appearance of algebraic solitons is possible for $|g| > g_c$. Indeed, if the limit $\varkappa \to 0$ is taken in (31), then the solution of the first type ($+$) disappears while the solution of the second type ($-$) changes into

$$\zeta = -\frac{4}{\varepsilon_0}\left(\frac{\hbar V}{g}\right)^2 \left\{ \xi^2 + \left(\frac{\hbar V}{g\varepsilon_0}\right)^2 \right\}^{-1}. \tag{32}$$

This is a power-law soliton. We see that the power-law solitons disappear in the limit $|g| \to g_c$.

Solutions of the equation (5) which correspond to the solitons (31) are

$$s_1 = -\frac{4m\hbar (c^2 - V^2) |V| \varkappa}{g^2 (\eta \pm (1 + \eta^2)^{1/2} \operatorname{ch} \varkappa\xi)},$$

$$s_2=\pm\frac{4m\hbar(c^2-V^2)V^2\varkappa^2(1+\eta^2)^{1/2}\,\mathrm{sh}\,\varkappa\xi}{\omega_0 g^2(\eta\pm(1+\eta^2)^{1/2}\,\mathrm{ch}\,\varkappa\xi)^2},$$

$$s_3=\frac{4m\hbar(c^2-V^2)V^2\varkappa^2}{\omega_0 g}\left\{\frac{\eta}{\eta\pm(1+\eta^2)^{1/2}\,\mathrm{ch}\,\varkappa\xi}+\frac{1}{(\eta\pm(1+\eta^2)^{1/2}\,\mathrm{ch}\,\varkappa\xi)^2}\right\}. \quad (33)$$

It is easily seen that the equation (30) and the solutions (31) and (33) go over into the equation (22) and the solutions (23) and (24), respectively, as the bifurcation parameter reaches the value $|g| = g_c$. However, in contrast to the small g case, the soliton solutions are now not symmetric with respect to the equilibrium state (Fig. 2). The difference in the amplitudes of the two types of solitons given by the solutions (31) is $4\varepsilon_0$.

The interval of the allowed soliton speed for $|g|\geqslant g_c$ is found from the reality condition for $\varkappa$:

$$c_1^2\leqslant V^2\leqslant c^2.$$

As in the previously considered case, the rays $\omega = Vk$ fall into the "forbidden" region for the rays which connect the origin with the dispersion curve for the linear vibrations (Fig. 1).

Behavior of the upper level occupancy $S_3 = s_3 + S_3^0$ differs from the case $|g| < g_c$ only in that the magnitudes of the extremal values of $S_3(0)$ do not exceed $(g_c/g)^2$.

4. SOLITON DYNAMICS

Fundamental characteristics of a soliton, viewed as a collective excitation of a crystal, are its energy and momentum. The soliton energy can be calculated with the help of the expression(10) and the solutions (23) and (24) or (31) and (32). Such calculations are very similar in all considered cases so that we shall give only the results for the case $|g|\leqslant g_c$.

A calculation gives

$$E=\frac{8\rho\hbar^2\omega_0}{g^2}V^3\left[\frac{V^2-c_*^2}{c^2-V^2}\right]^{1/2} \quad (34)$$

for the energy per unit area perpendicular to the x axis.

A soliton motion is connected with an actual mass transport so that it is accompanied by a momentum. The momentum of the molecules in an elastic wave, calculated per unit area perpendicular to the direction of the soliton motion, is

$$P_{\mathrm{mol}}=\int_{-\infty}^{\infty}\rho\frac{\partial u_x}{\partial t}dx=-\rho V\int_{-\infty}^{\infty}\frac{\partial u_x}{\partial x}dx=-\rho V[u_x(\infty)-u_x(-\infty)]. \quad (35)$$

Substituting (27) into (35), we obtain

$$P_{\mathrm{mol}}=\mp(2\pi\hbar/g)\rho V^2. \quad (36)$$

However, it is convenient to characterize soliton dynamics not by the momentum of the molecules (36) but by a quantity which stems from crystal homogeneity with respect to a displacement of the long-wavelength excitations, i.e., by the field momentum of a soliton. We emphasize that a soliton is an excitation of the vector fields **u** and **S**. When determining a field momentum it is convenient to start from the Lagrangian density for corresponding fields. To this end, it is convenient to introduce angular variables θ and φ:

$$S_1=\sin\theta\cos\varphi,\quad S_2=\sin\theta\sin\varphi,\quad S_3=\cos\theta.$$

Then, the equations of motion (5) are reduced to the two equations

$$\frac{\partial\theta}{\partial t}=-\frac{g}{\hbar}\sin\varphi\,\mathrm{div}\,\mathbf{u},\qquad \frac{\partial\varphi}{\partial t}=\omega_0-\frac{g}{\hbar}\,\mathrm{ctg}\,\theta\cos\varphi\,\mathrm{div}\,\mathbf{u}. \quad (37)$$

The Lagrangian density which gives rise to the equations (37) and to the equation of motion of the elastic medium (7) can be written as

$$L=\frac{1}{2}n\hbar(1+\cos\theta)\frac{\partial\varphi}{\partial t}+\frac{1}{2}\rho\left(\frac{\partial\mathbf{u}}{\partial t}\right)^2-\frac{1}{2}\rho c^2(\mathrm{div}\,\mathbf{u})^2$$
$$-\frac{1}{2}ng\sin\theta\cos\varphi\,\mathrm{div}\,\mathbf{u}-\frac{1}{2}n\hbar\omega_0(1+\cos\theta). \quad (38)$$

The field momentum (per unit area) is, by definition,

$$P=-\int_{-\infty}^{\infty}\frac{\partial L}{\partial\dot{\mathbf{u}}}\frac{\partial\mathbf{u}}{\partial x}dx-\int_{-\infty}^{\infty}\frac{\partial L}{\partial\dot{\varphi}}\frac{\partial\varphi}{\partial x}dx, \quad (39)$$

where the dot indicates differentiation with respect to t. Therefore, in our case

$$P=\rho V\int_{-\infty}^{\infty}\varepsilon^2\,dx-\frac{1}{2}n\hbar\int_{-\infty}^{\infty}(1+\cos\theta)\frac{\partial\varphi}{\partial x}dx. \quad (40)$$

Let us write the field excitation energy, which depends on a single spatial coordinate, in terms of the angular variables:

$$E=\frac{1}{2}\rho c^2\int_{-\infty}^{\infty}\varepsilon^2\,dx+\frac{1}{2}ng\int_{-\infty}^{\infty}\sin\theta\cos\varphi\varepsilon\,dx$$
$$+\frac{1}{2}\rho\int_{-\infty}^{\infty}\left(\frac{\partial\mathbf{u}}{\partial t}\right)^2dx+\frac{1}{2}n\hbar\omega_0\int_{-\infty}^{\infty}(1+\cos\theta)\,dx. \quad (41)$$

We shall consider $\varepsilon = \mathrm{div}\,\mathbf{u}$, θ, and φ to be functions of a single variable $\xi = x - Vt$ and we shall calculate changes in the soliton energy (41) and momentum (40) for small variations in the functions **u**, θ, and φ. Tedious but in principle simple computations lead us to the following result:

$$\delta E=V\delta P. \quad (42)$$

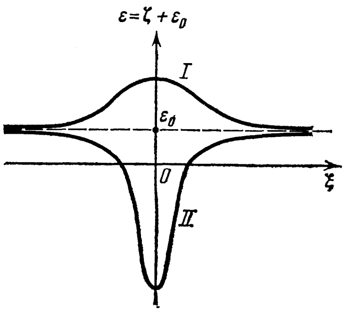

FIG. 2. Profiles of the soliton solutions of the two types for $|g| > g_c$.

Equation (42) indicates that the Hamilton equation

$$V=\partial E(P)/\partial P \tag{43}$$

is valid for the studied solitons in crystals.

Let us analyze the dependence $E=E(P)$ when $V>0$. Substituting the solutions (23) and (24) into (40), we find

$$P=(4\rho\hbar^2\omega_0/g^2)\left\{(3V^2-c^2)\left[\frac{V^2-c_*^2}{c^2-V^2}\right]^{1/2}\right.$$
$$\left.+c_0^2\operatorname{arctg}\left[\frac{V^2-c_*^2}{c^2-V^2}\right]^{1/2}\right\}. \tag{44}$$

The dependence of the soliton energies on the momenta is determined by a comparison between the relations (34) and (44). Near the boundary of the allowed values of the speed we have

$$E=c_*P,\quad V\to c_*; \tag{45}$$

$$E=c(P-P_0),\quad V\to c, \tag{46}$$

where $P_0=\pi n\hbar$.

The fact that the soliton momentum can be comparable to $n\hbar$ is consistent with the long-wavelength approximation. A similar situation occurs in the theory of magnetic solitons[6] and reduces to the following. The soliton can be considered as a bound state of a large number of elementary excitation of the upper tunneling level. Therefore, it is necessary to require that the average momentum per excitation of the ground state be small in comparison to $n\hbar$. The total number N_e of molecular excitations per unit area can be determined as

$$N_e=n\int_{-\infty}^{\infty}(S_3+1)\,d\xi. \tag{47}$$

Substituting the solution (24) into (47), we obtain

$$N_e=4n\frac{V}{\omega_0}\frac{[(V^2-c_*^2)(c^2-V^2)]^{1/2}}{c_0^2}. \tag{48}$$

The maximal value of N_e corresponds to $V^2=(c^2+c_*^2)/2$ and equals

$$N_e^{max}=(n/\omega_0)[2(c^2+c_*^2)]^{1/2}.$$

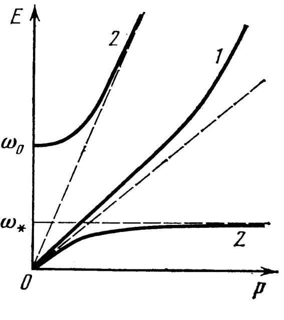

FIG. 3. Soliton dispersion law for $|g|<g_c$ (curve 1). The curves number 2 correspond to the dispersion laws for the harmonic vibrations of the crystal.

Since by assumption $c/\omega_0\gg a$, we have $N_e^{\max}\gg 1$. In this way the region of validity of the long-wavelength approximation determined by the requirement $aP\ll\hbar N_e$ can extend completely over the region of the momenta $P\sim P_0$.

Should we consider a one-dimensional elastic chain, then the soliton energy would be a^2E and its energy a^2P, where a is the molecular separation. Then it is reasonable to compare quantities a^2E and a^2P with the energy and the momentum of the phonons and to show in a single plot the dispersion laws for the solitons and the elementary harmonic excitations. Such a plot is schematically given in Fig. 3. We see that the soliton dispersion curve covers the energy interval $\Delta E=\hbar(\omega_0-\omega_*)$ in which the small vibrations cannot exist. Consequently, it is once more confirmed that the nonlinear solutions of the dynamic equations enlarge the set of collective excitations of a one-dimensional crystal.

In conclusion, we shall discuss the possibility of exciting the described solitons mechanically. At the creation of a soliton at the crystal's left-hand surface ($x=-\infty$), the surface layer of the molecules is displaced by an amount u_0 expressed in terms of the soliton speed by the relation $u_0=2\pi\hbar V/g$. This displacement has to occur during the time which the soliton needs to traverse its own length:

$$\Delta t=1/\varkappa V=\frac{1}{\omega_0}\left[\frac{c^2-V^2}{V^2-c_*^2}\right]^{1/2}. \tag{49}$$

In order that the soliton be formed, a momentum (36) must be transmitted to the molecules of the crystal. This can be provided by an initial disturbance which would create a pressure

$$p=\frac{P_{\text{mol}}}{\Delta t}=\frac{2\pi\hbar\rho\omega_0}{g}V^2\left[\frac{V^2-c_*^2}{c^2-V^2}\right]^{1/2}. \tag{50}$$

Solitons which are described in the long-wavelength approximation must have a speed close to c_* (which is, in turn, of the same order of magnitude as c: $V^2-c_*^2\ll c^2$). In order to excite the solitons it is sufficient to create on a crystal surface a pressure $p\ll(\omega_0\hbar/g)$ for a time duration $\Delta t\gg 1/\omega_0$, where $G=\rho c^2$ is the compression modulus of the crystal. For certain values of the bifurcation parameter g these crystal excitations can be fully realized.

Note added in proof (May 11, 1984): By limiting the elastic crystal energy to the form in (9) and (10) we have lost a contribution from the transverse acoustic vibrations. Therefore, the speed c_t of the transverse sound is absent from all subsequent expressions (formally $c_t=0$). Taking into account the energy of the transverse acoustic vibrations does not change the results pertinent to the soliton excitations but it changes substantially the conclusions contained in the formula (19). At the bifurcation point, the limiting speed c_* of the waves associated with the lower branch turns out not to be equal to zero but to take the minimal possible value for the speed of the longitudinal sound, $2c_t/\sqrt{3}$.

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