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Nonlinear electron-phonon dynamics: The appearance of solitary excitations and localized modes

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The existence of solitary excitations is shown through a quantum-mechanical model consisting of electronic two-level centers coupled to the lattice vibrations of the surrounding crystal. Simultaneously localized phonon modes appear in the region around these stable excitations. Within a time-dependent projection-operator formalism coupled equations of motion for the solitary waves and the phonons are derived.

I. INTRODUCTION

The transport properties of nonequilibrium acoustic phonons in insulating crystals have been investigated extensively in the last few years. New experimental techniques were developed which allow the time-resolved detection of nonequilibrium phonon pulses.[1,2] Vibronic sideband spectroscopy[3] as well as monochromatic phonon creation and detection[4] have been used successfully to study the spectral and spatial dynamics of the phonons in insulating crystals.

The most important interaction processes for nonequilibrium acoustic phonons are the elastic impurity scattering processes[5] leading to diffusive phonon propagation. In addition phonon decay processes which are caused by lattice anharmonicities[6] give rise to a rapid decay of high-frequency phonons into low-frequency modes. Both interaction processes together are combined by Bron, Levinson, and O'Connor into the model of quasidiffusive phonon transport.[1]

The electron-phonon coupling, however, was not taken into account in this model. On the other hand the importance of the electron-phonon interaction for the phonon dynamics is demonstrated in an experiment of Engelhardt, Happek, and Renk.[4] By x-ray irradiation in a ruby crystal they convert Cr^{3+} to Cr^{2+} ions[4] which leads to a strong decrease of the phonon lifetime. Therefore this experiment is a direct indication of the influence of the electron-phonon interaction on the transport properties of nonequilibrium phonons. In their model of the "spectral spatial" diffusion[7] they assumed that Raman scattering processes are the relevant interaction processes which change the frequency distribution of a nonequilibrium phonon pulse.

Alternatively the authors[8,9] introduced a model which only takes into account the interaction of phonons with electronic scattering centers, represented by a distribution of two-level centers.

Using a unitary transformation it was shown, that "anharmonic" three phonon decay processes can be induced by the electron-phonon coupling. Since in this model the anharmonic processes are coupled with electronic transitions this special interaction gives rise to a "quasiresonant" phonon transport behavior and therefore differs qualitatively from the models of "quasidiffusion" and "spectral spatial" diffusion.[9]

In the calculations,[8,9] however, it was assumed that the phonons are not exactly in resonance with the electronic transitions. In this contribution we mainly will concentrate on these resonant phonons and we will show that they induce some important effects. In particular in the following three topics will be discussed.

(i) The interaction of acoustic phonons with the electronic two level centers can lead to a coupled coherent pulse propagation which is similar to the phenomenon of self-induced transparency in nonlinear optics. This will be shown in Secs. II and III. First the electron-phonon Hamiltonian is rewritten by applying a unitary transformation which allows to separate the resonant and nonresonant part of the interaction leading to coherent and noncoherent motions. The Heisenberg equations for the coherent system can be integrated and it turns out that the electron phonon systems bear solitary wave solutions.

(ii) In Sec. IV the influence of the solitary excitation on the residual phonon modes is calculated and it is shown that the renormalization of the phonon spectrum leads to localized phonon modes.

(iii) Finally, in Sec. V from the microscopic model, coupled equations of motion for the phonon-soliton system are derived.

These equations represent a generalization of the phonon Boltzmann equation derived recently.[8]

II. MODEL HAMILTONIAN

The Hamiltonian of the coupled system of electronic two-level centers and optical phonon modes has the following form ($\hbar=1$):

$$H=\Delta\sum_m \sigma_m^z+\sum_k \omega_k b_k^\dagger b_k +\kappa\sum_m\sum_k[\sigma_m^+ b_k\exp(ikm)+\sigma_m^- b_k^\dagger\exp(-ikm)] . \quad (2.1)$$

The two-level centers at the lattice sites m are described by the quasispin operators σ_m^z and $\sigma_m^\pm$, which obey the usual commutation relations of an SU(2) algebra

$$[\sigma_m^+,\sigma_{m'}^-]=2\sigma_m^z\delta_{m,m'} ,\quad [\sigma_m^\pm,\sigma_{m'}^z]=\mp\sigma_m^\pm\delta_{m,m'} . \tag{2.2}$$

The Bose operators b_k ($b_k^\dagger$) destroy (create) phonons with wave vectors k and frequency ω_k where

$$[b_k,b_{k'}^\dagger]=\delta_{k,k'} . \tag{2.3}$$

For simplicity the branch index is neglected. Δ is the energy splitting of the electronic levels and κ the electron phonon coupling constant.

In the following we will divide the phonon spectrum into two parts, namely into phonons which are energetically in resonance with the electronic two-level systems and into phonons which are off resonance. In order to define the region of resonance we assume the energy splitting Δ to have a small distribution within a range 2η:

$$\Delta_0-\eta\leqq\Delta\leqq\Delta_0+\eta ,$$

where

$$2\eta\ll\Delta_0 .$$

Thus resonant phonons are phonons whose energies ω_k lie in the energy region of Δ. We now can define a special kind of course-graining operator C^m

$$C^m b_l=\sum_{l=k_0}^{k_1} b_l e^{ilm}\equiv B_L(m) , \tag{2.4}$$

where C^m selects from the whole phonon spectrum just the small part of resonant phonons with wave vectors

$$k_0\leqq k\leqq k_1 .$$

According to (2.4) this group of phonons is represented by a single collective mode $B_L(m)$, which still contains m as a parameter.

In order to make the separation of the resonant and nonresonant phonon modes more explicit we perform a unitary transformation to the variables $\sigma_m^\pm$, $B_L^\mp$ which is defined as

$$\begin{aligned}\sigma_m^+&=\tilde\sigma_m^+\cos\alpha+\tilde B_L^+(m)\sin\alpha ,\\ B_L^+(m)&=-\tilde\sigma_m^+\sin\alpha+\tilde B_L^+(m)\cos\alpha ,\\ \sigma_m^-&=\tilde\sigma_m^-\cos\alpha+\tilde B_L(m)\sin\alpha ,\\ B_L(m)&=-\tilde\sigma_m^-\sin\alpha+\tilde B_L(m)\cos\alpha .\end{aligned} \tag{2.5}$$

Choosing

$$\alpha=\frac{1}{2}\arctan\frac{2\kappa}{\Omega_L} , \tag{2.5a}$$

the interaction terms between the B_L and σ_m can be eliminated. Ω_L is the mean frequency of the resonance phonons, $\Omega_L=\bar\Delta=\Delta_0$. The transformed Hamiltonian reaches the form

$$\begin{aligned}H=&\sum_m(\tilde\Delta\sigma_m^z+\tilde\Omega_L B_L^+B_L)+\delta+\sum_k\omega_k b_k^\dagger b_k\\ &+\sum_m\sum_k\kappa\{\cos\alpha[\sigma_m^+b_k\exp(ikm)+\sigma_m^-b_k^\dagger\exp(-ikm)]\\ &\quad+\sin\alpha[B_L^+b_k\exp(ikm)\\ &\quad+B_Lb_k^\dagger\exp(-ikm)]\} ,\end{aligned} \tag{2.6}$$

where

$$\tilde\Delta=\Delta\cos^2\alpha,\quad \delta=\tfrac{1}{2}\Omega_L\cos^2\alpha ,$$

$$\tilde\Omega_L=-\tfrac{1}{2}\Delta+(\tfrac{1}{2}\Delta+\Omega_L)\cos^2\alpha+\kappa\sin(2\alpha) .$$

The symbol $\sim$ has been dropped in the operator expressions $B_L^\pm$, $\sigma_m^\pm$. The resonance phonons and the electronic two-level systems are now decoupled except for an indirect interaction between these subsystems via the nonresonant phonons [last term in (2.6)].

For the discussion of the dynamic behavior of the total system it is convenient to perform a second unitary transformation of the form

$$\tilde H=e^{-S}He^S , \tag{2.7}$$

where in close analogy to the model of quasiresonant phonon transport S is chosen in such a form that it eliminates the first-order terms in the interaction part of Eq. (2.6), e.g., we claim

$$[H_0,S]+H_I=0 . \tag{2.8}$$

This relation is satisfied for[8,9]

$$S=-i\lim_{\epsilon\to0^+}\int_{-\infty}^{0}dt\exp(-\epsilon|t|+iH_0t)\times H_I(t=0)\exp(-iH_0t) \tag{2.9}$$

where H_0 and H_I are given by

$$\begin{aligned}H_0=&\tilde\Delta\sum_m(\sigma_m^z+\tilde\Omega_LB_L^+B_L)+\delta+\sum_k\omega_kb_k^\dagger b_k ,\\ H_I=&\sum_m\sum_k\kappa\{\cos\alpha[\sigma_m^+b_k\exp(ikm)+\text{H.c.}]\\ &+\sin\alpha[B_L^+b_k\exp(ikm)+\text{H.c.}]\} .\end{aligned} \tag{2.10}$$

As a result we get

$$S=\sum_k\sum_m\kappa\left\{\frac{\cos\alpha}{\omega_k-\tilde\Delta}[\sigma_m^+b_k\exp(ikm)-\text{H.c.}]+\frac{\sin\alpha}{\omega_k-\tilde\Delta}[B_L^+b_k\exp(ikm)-\text{H.c.}]\right\} . \tag{2.11}$$

Since $\omega_k\neq\tilde\Delta,\tilde\Omega_L$ and assuming the electron-phonon coupling to be weak we may expand (2.7) in a power series and obtain up to third order in κ using Eq. (2.11):

$$\tilde H=H_0+\tfrac{1}{2}[H_I,S]+\tfrac{1}{3}[[H_I,S],S]+\cdots\equiv H_1+H_2+H_3 ,$$

where

$$H_1=\sum_m(\widetilde{\Delta}\sigma_m^z+\widetilde{\Omega}_L B_L^+B_L)+\delta+\sum_k\sum_m\sum_{m_1}\frac{\kappa^2}{\omega_k-\widetilde{\Omega}_L}\exp[ik(m-m_1)](\sigma_m^+\cos\alpha+B_L^+\sin\alpha)(\sigma_{m_1}^-\cos\alpha+B_L\sin\alpha)\,,$$

$$H_2=\tfrac{1}{2}\sum_k\sum_m\sum_{k_1}\sum_{m_1}\frac{\kappa^2}{\omega_{k_1}-\widetilde{\Delta}}(\delta_{m,m_1}\sigma_m^z\cos^2\alpha-\sin^2\alpha)\{b_{k_1}^\dagger b_k\exp[i(km-k_1m_1)]+\text{H.c.}\}+\sum_k\omega_k b_k^\dagger b_k\,, \tag{2.12}$$

$$H_3=\tfrac{1}{3}\sum_k\sum_{k_1}\sum_m\sum_{m_1}\sum_{m_2}\frac{\kappa^3}{(\omega_{k_1}-\Delta)^2}\{\sigma_m^+b_k\exp(ikm)(\cos^3\alpha)\delta_{m,m_1}\delta_{m,m_2}$$
$$+3(\sin^2\alpha-\sigma_m^z\cos^2\alpha\delta_{m,m_1})(\sigma_{m_2}^+\cos\alpha+B_L^+\sin\alpha)b_k\exp[ik(m_2-m_1)+ikm]\}$$
$$+\tfrac{1}{3}\sum_k\sum_{k_1}\sum_m\sum_{m_1}\sum_{m_2}\frac{\kappa^3}{(\omega_k-\widetilde{\Delta})(\omega_{k_1}-\widetilde{\Delta})}\{\sigma_m^+b_{k_1}\exp(ik_1m)(\cos^3\alpha)\delta_{m,m_1}\delta_{m,m_2}$$
$$+3(\sin^2\alpha-\sigma_m^z\cos^2\alpha\delta_{m,m_1})(\sigma_{m_2}^+\cos\alpha+B_L^+\sin\alpha)b_k\exp[ik_1m+ik(m_2-m)]\}$$
$$+\tfrac{1}{3}\sum_m\sum_k\sum_{k_1}\sum_{k_2}(\cos^3\alpha)\left[\frac{\kappa^3}{(\omega_{k_1}-\widetilde{\Delta})(\omega_{k_2}-\widetilde{\Delta})}\sigma_m^+b_k^+b_{k_1}b_{k_2}\exp[im(k_1+k_2-k)]\right.$$
$$\left.+\frac{\kappa^3}{(\omega_k-\widetilde{\Delta})(\omega_{k_2}-\widetilde{\Delta})}\sigma_m^+b_{k_1}^+b_{k_2}b_k\exp[im(k+k_2-k_1)]\right]+\text{H.c.}$$

In (2.12) we divided $\widetilde{H}$ into three parts, H_1, H_2, H_3 which will be investigated in the following sections from different viewpoints. In the next section we show that when using H_1 to derive equations of motion for the σ_m, B_L we obtain solitonlike excitations. The influence of the soliton solutions on the phonon part H_2 is studied in Sec. IV. Finally, in Sec. V the coupled electron-phonon dynamics is treated by means of a time-dependent projection operator technique.

III. COHERENT PHONON MOTION: SOLITARY EXCITATIONS

In this section we derive coupled equations of motion for the electronic two-level systems and the resonant phonon mode B_L starting from

$$H_1=\delta+\sum_m[\widetilde{\Delta}+L(m)]\sigma_m^z+L+\sum_m(\widetilde{\Omega}_L+L)B_L^+B_L$$
$$+\sum_m\sum_{m_1}[T(m-m_1)\sigma_m^+\sigma_{m_1}^-+\text{H.c.}]$$
$$+\sum_m[A(m)\sigma_m^+B_L+\text{H.c.}]\,, \tag{3.1}$$

where

$$T(m-m_1)=\sum_k\frac{\kappa^2}{\omega_k-\widetilde{\Delta}}\cos^2\alpha\exp[ik(m-m_1)]\,,$$
$$A(m)=\sum_{m_1}T(m-m_1)\tan\alpha\,,$$
$$L=\sum_k\frac{\kappa^2}{\omega_k-\widetilde{\Delta}}\cos^2\alpha\,.$$

The Heisenberg equations of motion read

$$i\frac{\partial}{\partial t}O=[O,H]\,, \tag{3.2}$$

where O are the phonon and electron operators ($\hbar=1$).

In the explicit derivation of the equations of motion we assume m to be a continuous variable and therefore we may expand all nonlocal operator products in analogy to a procedure used earlier by Haken and Schenzle:[10]

$$\sum_{m_1}T(m-m_1)\sigma_{m_1}^+=\sum_{\nu=0}^{\infty}\frac{(i)^\nu}{\nu!}\frac{\partial^\nu T(k')}{\partial k'^\nu}\bigg|_{k=k'}\frac{\partial^\nu}{\partial m^\nu}\sigma_m^+\,, \tag{3.3}$$

where

$$T(m-m_1)=\frac{1}{N}\sum_{k'}T(k')\exp[-ik'(m-m_1)]\,. \tag{3.4}$$

N is the number of atoms in the chain. Keeping only terms up to first order in (3.3) we obtain the following set of coupled equations

$$-i\dot{B}_L^+=\widetilde{\Omega}_L B_L^+ + iv_{\text{ph}}\frac{\partial B_L^+}{\partial m}+A(m)\sigma_m^+\,, \tag{3.5a}$$

$$-i\dot{B}_L=-\widetilde{\Omega}_L B_L - iv_{\text{ph}}\frac{\partial B_L}{\partial m}-A(m)\sigma_m^-\,, \tag{3.5b}$$

$$-i\frac{\partial}{\partial t}(B_L^+B_L)=A(m)(\sigma_m^+B_L-\text{H.c.})\,, \tag{3.5c}$$

$$-\dot{\sigma}_m^z=-2v_k\frac{\partial}{\partial m}(\sigma_m^+\sigma_m^-)+iA(m)(\sigma_m^-B_L^+-\text{H.c.})\,, \tag{3.5d}$$

$$-i\dot{\sigma}_m^+=\widetilde{\Delta}\sigma_m^++i\left[\frac{\partial\sigma^+}{\partial m}\right]\sigma_m^z\frac{\partial T_{(k)}}{\partial k}$$
$$-E\sigma_m^+-2\sigma_m^zB_L^+A(m)\,, \tag{3.5e}$$

$$i\dot{\sigma}_m^- = \tilde{\Delta}\sigma_m^- - i\left[\frac{\partial\sigma}{\partial m}\right]\sigma_m^z \frac{\partial T(k)}{\partial k} + E\sigma_m^- + 2\sigma_m^z B_L A(m) , \tag{3.5f}$$

where we used the abbreviations

$$E = 2T(0) + \tilde{\Delta} + L ,$$

and

$$v_k = \frac{\partial T(k)}{\partial k} = v_{el} ,$$

$$v_{ph} = \frac{\partial \Omega_L}{\partial L} .$$

v_{el} and v_{ph} are the electron and phonon velocities, respectively.

Since we are interested in pulselike solutions we assume that the spatial and time variables appear in the phonon and electron variables only in the form

$$v\tau = m - vt .$$

v is the velocity common for the phonon and electron pulse. This leads us to an ansatz for $B_L^{\pm}$, $\sigma_m^{\pm}$ of the form

$$\begin{aligned} \sigma_m^{\pm}(t) &= \exp(\pm i\tilde{\Delta}t)\sigma^{\pm}(m - vt) , \\ B_L^{\pm}(m,t) &= \exp(\pm i\tilde{\Omega}t)B^{\pm}(m - vt) . \end{aligned} \tag{3.6}$$

With Eq. (3.6) one obtains from Eqs. (3.5) equations of motion for the slowly varying "envelope functions," $\sigma(m - vt)$, $B(m - vt)$. Neglecting in (3.5) the spatial derivatives and multiplying (3.5e) by σ_m^- and (3.5f) by σ_m^+ and adding both equations together we end up with

$$\frac{\partial}{\partial \tau}(\sigma^+\sigma^-) = 2iA(\tilde{\sigma}^z - \tfrac{1}{2})(B_L^+\sigma^- - \text{H.c.}) . \tag{3.7}$$

In (3.7) $\tilde{\sigma}^z$ is defined as

$$\tilde{\sigma}^z = \sigma^z + \tfrac{1}{2} .$$

Starting from (3.5d) and integrating over τ we get

$$\sigma^+\sigma^- = -(\tilde{\sigma}^z)^2 + \tilde{\sigma}^z + C_1 , \tag{3.8}$$

where the operators σ_m, B_L are treated as C numbers.[10] C_1 is a constant of integration. From (3.5a) and (3.5b) we obtain with (3.6)

$$\lambda^2 \frac{\partial B_L}{\partial \tau} = -iA\sigma^- , \tag{3.9a}$$

$$\lambda^2 \frac{\partial B_L^+}{\partial \tau} = iA\sigma^+ , \tag{3.9b}$$

and

$$\lambda^2 = -\frac{v}{v_{ph}} + 1 .$$

We insert (3.9) into (3.8). Without loss of generality the integration constant C_1 is chosen to be zero.

$$\tilde{\sigma}^z = \lambda^2 B_L^+ B_L + C_2 , \tag{3.10}$$

where C_2 is a second constant of integration. Again we put

$$C_2 = 0 .$$

Finally, combining (3.9), (3.10), and (3.8) we derive a nonlinear equation of motion for B_L in the form

$$\frac{\lambda^2}{A^2(m)}\left[\frac{\partial B_L^+}{\partial \tau}\right]\left[\frac{\partial B_L}{\partial \tau}\right] = -\lambda^2(B_L^+B_L)^2 + B_L^+B_L . \tag{3.11}$$

This equation for the phonon part of the pulse is formally equivalent to the photon pulse equation in the case of self-induced transparency obtained by Haken and Schenzle.[10] Following these authors with the ansatz

$$B_L^{\pm} = B_L e^{\pm i\phi} ,$$

one obtains for the field amplitude the solution

$$B_L^2 = \frac{a_3}{a_1\cosh[2\gamma(\tau - \tau_0)] + a_2} , \tag{3.12}$$

with

$$\gamma^2 = \left[\frac{A}{\lambda}\right]^2 - \tfrac{1}{4}(\tilde{\Omega} - E - \tilde{\Delta})^2 ,$$

$$a_3 = 2\gamma^2, \quad a_2 = 2a_4 ,$$

$$a_1 = (a_4^2 + 4\gamma^2)^{1/2}, \quad a_4 = [A^2 + \tfrac{1}{2}\lambda^2 T(k)(\tilde{\Omega} - E - \tilde{\Delta})] .$$

This result describes the coherent motion of a solitary excitation built up by an electron and a phonon pulse which interact resonantly.

From this we conclude that a coherent phonon field can lead to solitary pulse propagation when interacting resonantly with an electronic medium. This phenomenon may be observed in phonon scattering or time-of-flight experiments in the form of very slowly decaying phonon modes.[4] For an application we may think of insulating crystals such as Al_2O_3 (Cr^{2+}), where defect ions interact resonantly with phonons.[8,9]

IV. LOCALIZED PHONON MODES

As we discussed in the previous sections, when separating the phonon system into "nonresonant" and "resonant" modes, the interaction of the resonant phonons with the electronic two-level centers led to a solitary wave propagation. In this section we will see in which way the nonresonant phonons are influenced by the solitary wave solution. Similar to the case of one-dimensional chains, where it can be shown that around the soliton or polaron solutions localized phonon modes are created, we will demonstrate in the following that in our model the existence of the solitary solution (4.2) also will lead to a localization of the nonresonant phonons (4.1) as soon as the soliton is built up.

We start from the (nonresonant) phonon part of the Hamiltonian (2.12):

$$H_2=\sum_k \omega_k b_k^\dagger b_k + \tfrac{1}{2}\sum_{k_1}\sum_k\sum_m\sum_{m_1}\left[\frac{\kappa^2}{\omega_{k_1}-\tilde{\Delta}}\sigma_m^z b_k^\dagger b_{k_1} \times\exp(ikm-ik_1m_1)+\text{H.c.}\right] . \quad (4.1)$$

For simplicity in (4.1) we have put $\alpha=0$, because the first unitary transformation (2.5) only acts on the resonant phonons and the electronic system.

We can introduce the soliton solution via σ_m^z. Since for finite soliton velocities $a_1 \gg a_2$, we can neglect a_2 in (3.12) and we get together with (3.10):

$$\tilde{\sigma}^z(\tau)=\tilde{\sigma}_0^z\text{sech}[2\gamma(\tau-\tau_0)] . \quad (4.2)$$

The identities

$$\sum_m \exp[im(k-k_1)]=\delta_{k,k_1} ,$$

$$\sum_k \exp[ik(m-m_1)]=\delta_{m,m_1} ,$$

lead from (4.1) using (4.2) to a new Hamiltonian

$$H_2=H_2^{\text{d}}+H_2^{\text{nd}} , \quad (4.3)$$

where the diagonal H_2^{d} and the nondiagonal H_2^{nd} part read

$$H_2^{\text{d}}=\sum_k \omega_k b_k^\dagger b_k+\sum_k b_k^\dagger b_k \frac{\kappa^2}{\tilde{\Delta}-\omega} , \quad (4.3\text{a})$$

$$H_2^{\text{nd}}=\sum_k\sum_{k_1}\left[b_k^\dagger b_{k_1}\frac{\kappa^2}{\tilde{\Delta}-\omega}\frac{\pi}{4\gamma}\tilde{\sigma}_0^z\text{sech}\left[\frac{\pi}{4\gamma}(k-k_1)\right]+\text{H.c.}\right] . \quad (4.3\text{b})$$

For simplicity in (4.3) we have neglected the dispersion of the nonresonant phonon modes

$$\omega_k \to \omega .$$

Physically this means, that our calculations do not take account of the whole phonon spectrum; we only calculate the effect of phonon localization for one single mode. This, however, changes neither the general validity of the argumentation nor the basic effects of the localization behavior.

With this simplification H_2^{nd} and H_2^{d} commute

$$[H_2^{\text{d}},H_2^{\text{nd}}]=0 .$$

The eigenvalues of H_{ph}^{d} are known and we are left with the problem of solving the eigenvalue equation

$$\sum_k\sum_{k_1}\left[b_k^\dagger b_{k_1}\frac{\kappa^2}{\tilde{\Delta}-\omega}\frac{\pi}{4\gamma}\tilde{\sigma}_0^z\text{sech}\left[\frac{\pi}{4\gamma}(k-k_1)\right]+\text{H.c.}\right]=\sum_\mu \epsilon_\mu b_\mu^\dagger b_\mu . \quad (4.4)$$

This can be done by applying a linear canonical transformation:

$$b_\mu=\sum_k f_{\mu k}b_k , \quad (4.5)$$

where the unitary matrix $f_{\mu k}$ transforms the operators b_k to new operators b_μ for which H_2 is diagonal. Inserting the transformation into (4.4) one obtains an equation for the unknown coefficients $f_{\mu k}$

$$\sum_{k_1}\left[\frac{\kappa^2}{\tilde{\Delta}-\omega}\frac{\pi}{4\gamma}\tilde{\sigma}_0^z\text{sech}\left[\frac{\pi}{4\gamma}(k-k_1)\right]\right]f_{\mu k_1}=\varepsilon_\mu f_{\mu k} . \quad (4.6)$$

As an ansatz for f_{μ_k}, we choose

$$f_{\mu k}=\frac{k^{\mu-1}}{\sinh\left[\frac{\pi}{4\gamma}k\right]}+\frac{k^\mu}{\cosh\left[\frac{\pi}{4\gamma}k\right]} , \quad (4.7)$$

where μ is an integer.

For even (odd) values of μ the function $f_{\mu k}$ has even (odd) parity:

$$f_{\mu,-k}=(-1)^\mu f_{\mu,k} . \quad (4.8)$$

Converting the sum into an integral, the diagonalization of (4.6) is reduced to the problem of calculating the eigenvalues E_μ and eigenfunctions $f_\mu(k)$ of the integral equation[12]

$$\varepsilon_\mu f_\mu(k)=\int dk_1 K(k,k_1)f_\mu(k_1) , \quad (4.9)$$

where the integral kernel $K(k,k')$ is defined as

$$K(k,k_1)=\text{sech}\left[\frac{\pi}{4\gamma}(k-k_1)\right]\tilde{\sigma}_0^z\frac{\pi}{4\gamma}\frac{\kappa^2}{\tilde{\Delta}-\omega} . \quad (4.10)$$

By inspection we find that the ansatz solves the integral equation for both even and odd parities of $f_{\mu k}$. The eigenvalues are given by

$$\varepsilon_\mu=-\frac{\kappa^2}{\tilde{\Delta}-\omega}\tilde{\sigma}_0^z\frac{\pi}{4\gamma}\frac{1}{\mu},\quad \mu=1,2,\ldots,\infty \quad (4.11)$$

and the corresponding eigenfunctions are

$$f_{\mu k}=\frac{1}{(N_1)^{1/2}}\left[\frac{\left[\frac{\pi}{4\gamma}\right]^{\mu-1}k^{\mu-1}}{\sinh\left[\frac{\pi}{4\gamma}k\right]}+\frac{\left[\frac{\pi}{4\gamma}\right]^{\mu}k^{\mu}}{\cosh\left[\frac{\pi}{4\gamma}k\right]}\right] . \quad (4.12)$$

N_1 is a normalization factor.

With this result the phonon Hamiltonian H^{ph} of Eq. (4.3) can be written in the diagonalized form

$$H_2=\sum_\mu \omega_\mu b_\mu^\dagger b_\mu , \quad (4.13)$$

where the frequencies of the phonon modes b_μ are defined as

$$\omega_\mu=\omega-\frac{\kappa^2}{\tilde{\Delta}-\omega}\frac{\pi}{4\gamma}\tilde{\sigma}_0^z\frac{1}{\mu} . \quad (4.14)$$

From (4.12) one sees that in the presence of a solitary excitation of the form (4.2) the energy spectrum of the nonresonant phonons is changed. The energy of phonons, which are lower in energy than the resonant phonons, is decreased, whereas the energy of phonons, which are higher in energy than the resonant phonons, is increased. In this way an energy gap around the resonance energy is built up, which leads to an enhanced stability of the soliton.

When one considers the eigenfunctions of these shifted phonon modes in real space one sees that they become localized. For the first four modes ($\mu=1,\ldots,4$), which can be written in the form

$$f_\mu(x)=\sum_k e^{ikx} f_{\mu k} , \tag{4.15}$$

where $x=na$ (a is the lattice constant, $n=1,2,\ldots,L$), one obtains

$$f_1(x)=\frac{1}{(2\pi N_1)^{1/2}}\tanh\left[\frac{x}{a}\right]\left[1+\frac{\pi}{4\gamma a}\frac{1}{\cosh(\gamma x/a)}\right] , \tag{4.15a}$$

$$f_2(x)=\frac{2\gamma}{(2\pi N_1)^{1/2}}\left[-\frac{\pi}{4\gamma a}\frac{1}{\cosh^2(x/a)}+\left[\frac{\pi}{4\gamma a}\right]^2\left[\frac{2}{\cosh^3(\gamma x/a)}-\frac{1}{\cosh(\gamma x/a)}\right]\right] , \tag{4.15b}$$

$$f_3(x)=\frac{4\gamma}{(2\pi N_1)^{1/2}}\left[\left[\frac{\pi}{4\gamma a}\right]^2\frac{\tanh(\gamma x/a)}{\cosh^2(\gamma x/a)}(-2)+\frac{\pi}{4\gamma a}\left[1-\frac{6}{\cosh^2(\gamma x/a)}\right]\right] , \tag{4.15c}$$

$$\begin{aligned}f_4(x)=\frac{4\gamma}{(2\pi N_1)^{1/2}}&\left[\left[\frac{\pi}{4\gamma a}\right]^3\left[\frac{1}{\cosh^4(\gamma x/a)}-\frac{2\tanh^2(\gamma x/a)}{\cosh^4(\gamma x/a)}\right]\right.\\&\left.+\left[\frac{\pi}{4\gamma a}\right]^4\left[\frac{1}{\cosh(\gamma x/a)}-\frac{2}{\cosh^3(\gamma x/a)}-\frac{6}{\cosh^5(\gamma x/a)}+\frac{18\tanh^2(\gamma x/a)}{\cosh^3(\gamma x/a)}\right]\right] .\end{aligned} \tag{4.15d}$$

Because the arguments of the trigonometric functions are the same as those in our soliton solution (3.12), the phonon modes are localized around the center of the soliton.

V. MASTER EQUATIONS

In the previous two sections we showed the existence of solitary-wave solutions in the model Hamiltonian (2.12). Due to the localized soliton solution the spectrum of the nonresonant phonons was renormalized.

The scattering terms between the solitary excitation and the nonresonant localized phonon modes given in H_3 [Eq. (2.12)], however, have not yet been considered. This interaction influences the dynamics of the unperturbed soliton as well as the spectral and spatial distribution of the phonon modes.

In the following we shall discuss the most general situation where the phonon and electron lifetimes vary on the same time scales. In this case one cannot eliminate one of the subsystems adiabatically. Instead, equations of motion of both coupled systems have to be derived.

This can be done using time-dependent projection operators.[12,13] Therefore we separate the Hamiltonian (2.12) into two parts

$$H=H_0+H_I , \tag{5.1}$$

where H_0 describes the unperturbed soliton and phonon systems

$$H_0=H_0^{\rm sol}+H_0^{\rm ph} . \tag{5.2}$$

Consequently the scattering processes in (2.12), where only one nonresonant phonon is involved, e.g.,

$$\sigma_m^+ b_{k1},\quad B_L^+ b_k ,$$

can be neglected, because of energy conservation.

Then we end up with the Hamiltonian

$$H_I=\sum_m\sum_{k_1}\sum_{k_2}\sum_{k_3}[\kappa(m,k_1,k_2,k_3)\sigma_m^+ b_{k_1}^\dagger b_{k_2} b_{k_3}+\text{H.c.}] , \tag{5.3}$$

where

$$\kappa(m,k_1,k_2,k_3)=\frac{2}{3}\frac{(\kappa\cos\alpha)^3}{(\omega_{k_2}-\tilde{\Delta})(\omega_{k_3}-\tilde{\Delta})}\exp[im(k_2+k_3-k_1)] .$$

Here only the electronic part of the soliton system couples to the nonresonant phonons. From this it follows that we can describe the phonon soliton dynamics in terms of a statistical operator ρ which only depends on the electronic and nonresonant phonon degrees of freedom. The statistical operator ρ can be split into a relevant and irrelevant part, ρ_r, ρ_i

$$\rho=\rho_r+\rho_i . \tag{5.4}$$

Assuming that the relevant part ρ_r factorizes into a pure phonon ρ_{ph} and pure electron ρ_e operator

$$\rho_r = \rho_{ph}\rho_e \ , \tag{5.5}$$

ρ_r can be obtained by applying a time-dependent projection operator $P(t)$ to the statistical operator ρ:[13]

$$\rho_r(t) = P(t)\rho(t) \ , \tag{5.6}$$

$$P(t) = \rho_e(t)D_{ph} + \rho_{ph}(t)D_e - \rho_e(t)\rho_{ph}(t)D_e D_{ph} \ . \tag{5.7}$$

D_e and D_{ph} are projection operators for the electronic and phonon subsystem as, e.g., introduced by Grabert and Weidlich.[13]

With Eqs. (5.4)–(5.7), two coupled master equations for ρ_p and ρ_e can be derived. Using the Born-Markov approximation, one obtains in the interaction picture

$$\dot{\rho}_{ph} = -\int_0^\infty ds \exp(-\Gamma s) \times \mathrm{Tr}_e[\mathcal{L}_I(t)\mathcal{L}_I(t-s)\rho_e(t)\rho_{ph}(t)] \ , \tag{5.8}$$

$$\dot{\rho}_e = -\int_0^\infty ds \exp(-\Gamma s) \times \mathrm{Tr}_{ph}[\mathcal{L}_I(t)\mathcal{L}_I(t-s)\rho_e(t)\rho_{ph}(t)] \ . \tag{5.9}$$

In Eqs. (5.8) and (5.9), $\mathcal{L}_I$ is the Liouville operator $\mathcal{L}_I = [H_I, \ldots]$ and

$$\langle \cdots \rangle_e = \mathrm{Tr}_e(\rho_e \cdots) \ .$$

The influence of thermal reservoirs is treated phenomenologically by introducing the damping constant Γ.

The master equations (5.8) and (5.9) are two symmetrical equations for ρ_{ph} and ρ_e. This can be seen by changing the index $e \leftrightarrow$ph. Therefore they are a direct generalization of the master equation in the Argyres-Kelley projection operator formalism which describes the dynamics of one dynamical system coupled to a heat bath.[14] This close analogy turns out to be quite useful when deriving from (5.8) and (5.9) equations of motion for electronic and phonon expectation values, because the calculations are similar to the one done earlier in our model of "quasiresonant phonon propagation"[8,9] to which we refer. There a phonon Boltzmann equation for space- and k-dependent one-particle Wigner functions was derived using the Argyres-Kelley formalism. The electronic two-level centers were treated a heat bath.

In the model presented in this paper the electronic system has its own dynamic behavior and therefore can no longer play the role of a reservoir. It represents the dynamics of the solitary excitations. In this sense Eq. (5.9) is a generalization of the phonon transport equations presented earlier.[8,9]

The excitations in the phonon system can be described by one-particle Wigner functions ($\hbar = 1$):

$$f(n,k,t) = \frac{1}{8\pi^3}\int dq \exp(inq) \times \mathrm{Tr}_{ph}[\rho_{ph}(t) b^\dagger_{k-q/2} b_{k+q/2}] \ , \tag{5.10}$$

where n is the lattice site and k the wave vector of the phonon. As an ansatz for the N-particle Wigner functions, we neglect phonon-phonon correlations and write[8]

$$F(n_1,\ldots,n_N,k_1,\ldots,k_N) = \prod_{i=i}^{N} f(n_i,k_i,t) \ . \tag{5.11}$$

The nonresonant phonons are described by a set of occupation number states

$$\langle b^\dagger_{k-q/2} b_{k+q/2} \rangle \neq 0, \quad \langle b_k \rangle = \langle b_k^\dagger \rangle = 0 \ . \tag{5.12}$$

The electron system, however has a coherent motion, i.e.,

$$\langle \sigma_m^- \rangle, \langle \sigma_m^+ \rangle \neq 0 \tag{5.13}$$

which is described by the soliton solutions derived in Sec. III. Therefore the coupled master equations represent the dynamics of a soliton interacting with the nonresonant phonons b_k.

From Eqs. (5.8) and (5.9), equations of motion for the relevant electron and phonon operators can be derived. Using Eqs. (5.10)–(5.12), the continuum approximation of Sec. III and the properties of the coherent soliton states,

$$\langle \sigma_m^+ \sigma_n^- \rangle = \langle \sigma_m^+ \rangle \langle \sigma_m^- \rangle \ ,$$

we obtain from Eqs. (5.8) and (5.9)

$$\frac{\partial}{\partial t}\langle \tilde{\sigma}_m^z \rangle = -\sum_{k_1}\sum_{k_2}\sum_{k_3} W(k_1;k_2,k_3) f(m,k_1,t) f(m,k_2,t) f(m,k_3,t) - \sum_{k_2}\sum_{k_3} W(k_2,k_3) f(m,k_2,t) f(m,k_3,t) \ , \tag{5.14}$$

$$\begin{aligned}\left[\frac{\partial}{\partial t} + v_k \frac{\partial}{\partial m}\right] f(m,k,t) = & -\sum_{k_1}\sum_{k_2} W(k;k_1,k_2)(\langle \tilde{\sigma}_m^z \rangle - \langle \tilde{\sigma}_m^z \rangle^2) \\ & \times \{ f(m,k,t)[1+f(m,k_1,t)][1+f(m,k_2,t)] - [1+f(m,k,t)] f(m,k_1,t) f(m,k_2,t) \} \\ & - \sum_{k_1}\sum_{k_2} 2W(k_1;k_2,k)(\langle \tilde{\sigma}_m^z \rangle - \langle \tilde{\sigma}_m^z \rangle^2) \\ & \times \{ f(m,k,t) f(m,k_2,t)[1+f(m,k_1,t)] - [1+f(m,k,t)][1+f(m,k_2,t)] f(m,k_1,t) \} \ . \end{aligned} \tag{5.15}$$

v_k is the phonon group velocity.

The transition probabilities read

$$W(k;k_1,k_2)=W(k;k_2,k_1)$$
$$=\frac{16}{9}\frac{\cos^6\alpha}{\Gamma}\kappa^6\left[\frac{1}{(\omega_{k_1}-\tilde{\Delta})^2(\omega_{k_2}-\tilde{\Delta})(\omega_k-\tilde{\Delta})}+\frac{1}{(\omega_{k_2}-\tilde{\Delta})^2(\omega_{k_1}-\tilde{\Delta})(\omega_k-\tilde{\Delta})}+\frac{1}{(\omega_{k_1}-\tilde{\Delta})^2(\omega_{k_2}-\tilde{\Delta})^2}\right] \tag{5.16}$$

and

$$W(k_1,k_2)=W(k_2,k_1)=\frac{16}{9}\frac{(\kappa\cos\alpha)^6}{\Gamma}\frac{1}{(\omega_{k_1}-\tilde{\Delta})^2(\omega_{k_2}-\tilde{\Delta})^2} . \tag{5.17}$$

Since we have assumed that the electronic two-level centers are identical the coupling parameters are consequently identical and therefore independent of m. By calculating the transition probabilities we used the simplification

$$\frac{1}{\Gamma\pm i(\Delta+\omega_{k_1}-\omega_{k_2}-\omega_{k_3})}\rightarrow\frac{1}{\Gamma} .$$

In the derivation of Eqs. (5.16) and (5.17) we used Eq. (3.8).

As we have mentioned, the phonon transport equation (5.15) is a generalization of the Boltzmann equation derived earlier,[8] where the time dependence of the electronic system is described by σ_m^z. We want to note that through the interaction (5.3) in the collision term of the transport equation (5.15) phonon-phonon correlations in the Wigner functions are initiated leading to an anharmonic phonon decay.

The electronic master equation can be rewritten as

$$\frac{\partial}{\partial t}\langle\tilde{\sigma}_m^z\rangle=\frac{1}{\tau(m,t)}\langle\tilde{\sigma}_m^z\rangle , \tag{5.18}$$

where τ^{-1} is a space- and time-dependent relaxation time:

$$\frac{1}{\tau(m,t)}=\sum_{k_1}\sum_{k_2}\sum_{k_3}W(k_1;k_2,k_3)f(m,k_1,t)f(m,k_2,t)f(m,k_3,t)+\sum_{k_2}\sum_{k_3}W(k_2,k_3)f(m,k_2,t)f(m,k_3,t) . \tag{5.19}$$

$$\langle\sigma_m^z\rangle_t=\langle\sigma_m^z\rangle_0\exp[-t/\tau(m,t)] . \tag{5.20}$$

The shape of the soliton pulse decays due to the "anharmonic interaction" (5.3) with the nonresonant phonons.

Since the nonequilibrium phonon distributions are only weakly occupied for low temperatures ($f\ll 1$), we see from Eqs. (5.16) and (5.19) that

$$\frac{1}{\tau(m,t)}\ll\Gamma . \tag{5.21}$$

This means that the soliton pulse $\langle\sigma_m^z\rangle$ decays on a much slower time scale than the phonon-phonon scattering processes in Eq. (5.15) take place.

Therefore in Eq. (5.15), $\langle\sigma_m^z\rangle$ is nearly constant. Consequently, in this case the results obtained in the model of "quasiresonant phonon propagation"[9] are still valid also in the presence of solitary excitations. The resonant modes decay much slower than the nonresonant modes.

However, as seen in the present calculations the electronic decay leads to a decrease in the scattering rates, which can be seen by inserting (5.20) in (5.15). The transition probabilities in (5.15) becomes renormalized by the factor $\sim\langle\sigma_m^z\rangle_0 e^{-t/\tau}$. In this way the time scales at which the electronic and phonon systems move are shifted toward each other. Therefore the electronic system can no longer be treated as a heat bath as in the model of "quasiresonant phonon transport" where the electronic system was time and space independent, which means $\tau\rightarrow\infty$ in (5.20).

In the derivation of Eqs. (5.14) and (5.15) we implicitly assumed the soliton velocity to be constant. To go beyond this approximation one has to treat the dynamics of the resonant phonons more explicitly, which leads to additional master equations for the resonant-phonon system.

VI. CONCLUSIONS

In this article we presented a microscopic model which describes the interaction of electronic two-level systems and phonons. The phonon spectrum exhibits both, phonons which are in resonance with the two-level systems and nonresonant ones. This is a generalization of the model of "quasiresonant phonon transport" introduced earlier.[8,9] The coupled motion of resonant phonons and electronic two-level centers is treated in the frame of Heisenberg equations of motion and the possibility of solitary wave solutions is shown. It turns out that the existence of solitons in the resonant subsystems leads to localization behavior of the nonresonant modes of the phonon spectrum.

In addition the phonon transport equations derived recently[8,9] are generalized to include a dynamic behavior of the electronic system. The phonon transport equation turns out to be a generalized Boltzmann equation, whereas the electronic transport equation describes the motion of a solitary excitation interacting with the nonresonant phonons.

In conclusion we want to note that we treated the dynamical behavior of the resonant and nonresonant phonons separately. The formation of a solitary wave is caused by the interaction between the electronic two-level centers and the resonant phonon mode. Therefore this phonon mode builds up a propagating state and in this sense is "dynamically unbounded."[15]

In the molecular crystal model this argument of Holstein and co-workers leads them to transform to a "soliton centroid coordinate." Around the soliton solution they assume small fluctuations, which they identify as localized phonon modes. In this article we outlined a totally different procedure which starting from a microscopic Hamiltonian leads to a closed and natural description of the solitary excitations and their interaction with the phonons.

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