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Stepwise quantum decay of self-localized solitons

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The two-phonon decay of self-localized solitons in a one-dimensional monatomic anharmonic lattice caused by cubic anharmonicity is considered. It is shown that the decay takes place with emission of phonon bursts. The average rate of emission of phonons is of the order of the vibrational quantum per vibrational period. The characteristic relaxation time is determined by the quantum anharmonicity parameter; this time may vary from a few (quantum lattices, large anharmonicity) to thousands (ordinary lattices, small anharmonicity) of vibrational periods. [S0163-1829(97)50526-1]

The dynamics of strong nonlinear excitations in polymers and quasi-one-dimensional biomolecular chains is an active research field. Theoretical studies of anharmonic perfect lattices have shown the existence of localized vibrations (self-localized solitons, SLS's) with frequencies above the phonon band or in the gap of the phonon spectrum (see Refs. 1–10 and references therein). SLS's are solitonlike excitations in discrete lattices, and are thus closely related to ordinary solitons. The existence of stationary and moving SLS's was derived in the frame of classical mechanics. Until now there was little discussion about the influence of quantum (and thermal) fluctuations on stability of the SLS. An exception is the paper of Ovchinnikov[1] who argues that the decay of the SLS caused by these fluctuations diminishes with the increase of the mode amplitude. This statement, however, is based on a perturbational consideration which cannot be applied to the description of the evolution of vibrations with a strong amplitude.

Recently we showed[11–13] that the perturbational treatment of the effect of quantum and thermal fluctuations on a local mode associated with a defect atom in a lattice fails for the case when the amplitude of the vibration is large. In fact, the two-phonon damping of the local mode, caused by cubic anharmonicity, behaves dramatically with the change of the amplitude: at definite ''critical'' amplitudes relaxation jumps take place being accompanied by a generation of phonon bursts. This effect indicates that the quantum and thermal fluctuations may dominate in the dynamics of strong vibrations with the energy of the mode being within a specific range. The strong field of the local vibration causes the transformation of phonon operators and the increase of the number of phonons in time.[11,12] This mechanism of phonon generation by a local vibration is analogous to the mechanism of black hole radiation.[14–16]

In this paper we extend the theory[11–13] to the SLS in a monatomic one-dimensional lattice (chain) with cubic and hard quartic anharmonicity. This case is of special interest since the monatomic chain with both anharmonicities is the simplest model for the investigation of the quantum relaxation of SLS in a perfect lattice. Hard quartic anharmonicity is a prerequisite of the existence of the SLS, while the cubic anharmonicity stands for the two-phonon decay of the SLS. We note that SLS's in this model are stable in the classical limit, since all harmonics of the mode are out of resonance with lattice phonons.[3] The odd SLS is examined and an analytical nonperturbative solution of the problem and results of numerical calculations are presented.

The potential energy operator in a monatomic one-dimensional lattice, which includes linear, and the first two nonlinear terms, and takes into account the nearest-neighbor interaction, has the following form:

$$\hat{V}=\sum_{n}\sum_{r=2}^{4}\frac{K_r}{r}(\hat{U}_{n+1}-\hat{U}_n)^r, \tag{1}$$

where $\hat{U}_r$ is the operator of the longitudinal displacement of the nth atom from its equilibrium position, K_r are harmonic ($r=2$) and anharmonic (cubic: $r=3$, quartic: $r=4$) springs. The operators $\hat{U}_n$ satisfy the following equations of motion:

$$\frac{\partial^2\hat{U}_n}{\partial t^2}=\sum_{r=2}^{4}\overline{K_r}[(\hat{U}_{n+1}-\hat{U}_n)^{r-1}-(\hat{U}_n-\hat{U}_{n-1})^{r-1}], \tag{2}$$

$\overline{K_r}=K_r/M$, M is the mass of an atom. We suppose that an SLS with the frequency $\omega_l<2\omega_D$ is excited at the time $t=0$ at the sites $n=0$ and its neighbors (ω_D is the maximum harmonic frequency, $\omega_D=2\sqrt{\overline{K_2}}$). Anharmonic interactions are supposed to be weak, satisfying the condition $a_0^{(0)}\ll A_0$, where $a_0^{(0)}\sim\sqrt{\hbar/(2\omega_D M)}$ is the amplitude of zero-point vibrations, $A_0\sim\sqrt{K_2/K_4}$ is the amplitude of the self-localized vibration (for physical reasons the value of A_0 should not exceed the value of the lattice constant d, i.e., $K_4>K_2/d^2$; it is well fulfilled in realistic models). The condition $a_0^{(0)}\ll A_0$ means that the characteristic energy of the SLS, being of the order of K_2^2/K_4 is much larger than the characteristic vibrational quantum $\hbar\omega_D$:

$$\mathcal{K}=(K_2^2/K_4)/\hbar\omega_D=K_2^{3/2}M^{1/2}/2\hbar K_4\gg 1\,. \tag{3}$$

Note that the reversed dimensionless parameter $\mathcal{K}^{-1}$ characterizes the degree of the quantum anharmonicity of the lattice: it increases as the anharmonic term K_4 increases and as the mass of the atoms M decreases. In quantum crystals He and Ne this parameter is of the order of 1, while in ordinary crystals (characterized by the small amplitude of zero-point vibrations as compared to the lattice constant) it can reach many hundreds. In the problem under investigation $\mathcal{K}$ plays an essential role determining the time scale of the energy relaxation (see below).

In order to take account of the SLS decay we introduce the operators $\hat{U}_n$ in the form

$$\hat{U}_n = u_n(t) + \xi_n + \hat{q}_n , \tag{4}$$

where the ''classical'' displacements u_n are supposed to be nearly periodic functions: $u_n(t) \simeq A_n(t)\cos\omega_l t$, $A_n(t)$ and $\xi_n(t)$ are, slowly changing with time, amplitudes and shifts satisfying the equations

$$-\omega_l^2 A_n = \overline{K_2}(\overline{A_{n+1}} - \overline{A_n}) + 2\overline{K_3}(\overline{A_{n+1}}\,\overline{\xi_{n+1}} - \overline{A_n}\,\overline{\xi_n}) + 3\overline{K_4}\left(\frac{\overline{A^3_{n+1}}}{4} + \overline{A_{n+1}}\xi^2_{n+1} - \frac{\overline{A^3_n}}{4} - \overline{A_n}\,\overline{\xi^2_n}\right), \tag{5}$$

$$0 = \overline{K_2}(\overline{\xi_{n+1}} - \overline{\xi_n}) + \overline{K_3}\left(\frac{\overline{A^2_{n+1}}}{2} + \overline{\xi^2_{n+1}} - \frac{\overline{A^2_n}}{2} - \overline{\xi^2_n}\right) + \overline{K_4}\left(\frac{3\overline{A^2_{n+1}}\,\overline{\xi_{n+1}}}{2} + \overline{\xi^3_{n+1}} - \frac{3\overline{A^2_n}\,\overline{\xi_n}}{2} - \overline{\xi^2_n}\right), \tag{6}$$

where we keep just the $\cos\omega t$ term neglecting $\sim\cos 3\omega_l t$, $\cos 5\omega_l t, \ldots$[3–5] and introduce the new variables $\overline{A_n} = A_n - A_{n-1}$, $\overline{\xi_n} = \xi_n - \xi_{n-1}$. Variations of the A_n and ξ_n in time, due to quantum fluctuations, are described by means of time-dependent operators $\hat{q}_n(t)$, which satisfy the equations

$$d^2\hat{q}_n/dt^2 = W_{n+1}(t)(\hat{q}_{n+1} - \hat{q}_n) - W_n(t)(\hat{q}_n - \hat{q}_{n-1}) . \tag{7}$$

Here $W_n(t) = \overline{K_2} + k_n + 2w_n\cos\omega_l t$, where $k_n = 2\overline{K_3}\,\overline{\xi_n} + 3\overline{K_4}(\overline{A^2_n}/2 + \overline{\xi^2_n})$ and $w_n = \overline{K_3}\,\overline{A_n} + 3\overline{K_4}\,\overline{A_n}\,\overline{\xi_n}$ determine the change of springs caused by the SLS. The terms $\sim K_3(\hat{q}_n - \hat{q}_{n-1})^2$, $K_4\overline{A_n}(\hat{q}_n - \hat{q}_{n-1})^2$, $K_4\overline{\xi_n}(\hat{q}_n - \hat{q}_{n-1})^2$, and $K_4(\hat{q}_n - \hat{q}_{n-1})^3$ are supposed to be small and they were neglected. It is important to take these terms into account when $\omega_l > 2\omega_D$, i.e., when the two-phonon decay under consideration is forbidden by the energy conservation law.

The phonon Hamiltonian is defined as $\hat{H}_{ph}(t) = \hat{T}(t) + \hat{W}(t)$, where $\hat{T} = \frac{1}{2}\Sigma_n \hat{\dot{q}}^2_n$ and $\hat{W} = \hat{W}_0 + \hat{W}_1 + \hat{W}_t$ are the operators of kinetic and potential energy, $\hat{W}_0 = K_2/2\Sigma_n(\hat{q}_n - \hat{q}_{n-1})^2$, $\hat{W}_1 = 1/2\Sigma_n k_n(\hat{q}_n - \hat{q}_{n-1})^2$, $\hat{W}_t = \cos\omega_l t\Sigma_n w_n(\hat{q}_n - \hat{q}_{n-1})^2$; $\hbar = 1$. $\hat{W}_1$ describes the stationary perturbation of the phonon subsystem by the SLS, while $\hat{W}_t$ takes account of the oscillatory time dependence of the springs induced by the SLS. The time dependence of $\hat{H}(t)$ causes the phonon emission by the SLS (phonons are generated by pairs[11,12]).

Introducing properly chosen configurational coordinates $\hat{X}_i = \Sigma_n S_{in}\hat{q}_n$ and $\hat{Q}_m = \Sigma_n s_{mn}\hat{q}_n$, one can diagonalize $\hat{W}_1$ and $\hat{W}_t$: $\hat{W}_1 = \frac{1}{2}\Sigma_i \eta_i \hat{X}^2_i$, $\hat{W}_t = \cos\omega_l t\Sigma_m v_m \hat{Q}^2_m$. Both $\hat{W}_1$ and $\hat{W}_t$ have one zero eigenvalue; all other η_i are positive, while v_m are sign alternating, being symmetric with respect to the sign change. The mode with zero eigenvalue is totally symmetric and therefore does not enter into $\hat{q}_n - \hat{q}_{n-1}$.

The Hamiltonian $\hat{H}_{ph}$ is diagonalized by introducing time-dependent operators as follows. First the time-independent Hamiltonian $\hat{H} = \hat{H}^{(0)}_{ph} + \hat{W}_0$ is diagonalized by applying the standard methods of local dynamics[18,19] ($\hat{H}^{(0)}_{ph}$ is the Hamiltonian of the chain in harmonic approximation). Then the Hamiltonian $\hat{H}_{ph}(t)$ is diagonalized by the method presented in Refs. 11, 12, and 20. The time-dependent phonon Hamiltonian in the diagonal representation allows one to find the energy of phonons $E_{ph}(t)$ and the rate of phonon generation. The latter is equal to[11,12]

$$dE_{ph}(t)/dt = -dE_l(t)/dt = I(t) . \tag{8}$$

Here $E_l \simeq \Sigma_n(\omega_l^2 M\ A^2_n/2 + \Sigma_r K_r\ \xi^r_n/r)$ is the energy of the SLS,

$$I = \frac{\pi\omega_l}{8}\int_{\omega_D - \omega_l}^{\omega_D} d\omega\ \mathrm{Tr}\{P(\omega)P(\omega_l - \omega)\}[1 + 2n(\omega)] \tag{9}$$

denotes the intensity of the emission of phonons, the temperature factor is included by the multiplier $[1 + 2n(\omega)]$

$$P(\omega) = \frac{2}{\pi}\ v\ \mathrm{Im}[G(\omega)]\{I - vG(\omega - \omega_l)vG(\omega)\}^{-1}, \tag{10}$$

$G_{mm'}(\omega) = -i\int_0^\infty dt e^{i\omega t - \epsilon t}\langle[\hat{Q}_m(t), \hat{Q}_{m'}(0)]\rangle$ is the dynamical Green's function, $\hat{Q}_m(t) = e^{i\hat{H}t}Q_m e^{-i\hat{H}t}$. Use of the diagonal representation of the operator $\hat{W}_1$ allows one to easily express the Green's function $G_{mm'}$ via the one-site Green's function of the perfect lattice[21] $G^{(0)}_0(\omega) = i/(\omega\sqrt{\omega_D^2 - \omega^2})$ as follows:

$$G_{mm'} = \sum_{ii'} \sigma_{mi}\sigma_{m'i'}\overline{G_{ii'}}(\omega);$$

$$\overline{G_{ii'}} = ([I - G^{(0)}\eta]^{-1}\overline{G^{(0)}})_{ii'}, \tag{11}$$

where $\overline{G^{(0)}_{ii'}} = G^{(0)}_0\Sigma_{nn'}S_{ni}S_{n'i'}\cos\kappa(n - n')$, $\sigma_{mi} = \Sigma_n S_{mn}S_{in}$, $\eta_{ii'} = \eta_i\delta_{ii'}$, $\cos\kappa = 1 - 2\omega^2/\omega_D^2$, $\omega^2 < \omega_D^2$.

In the limit of small v_m one can neglect the term $\sim v^2$ in the denominator of formula (10). This limit coincides with the corresponding formula of the perturbation theory.[17] For realistic K_3 and the considered values of ω_l between $1.3\omega_D$ and $2\omega_D$ the term mentioned is not small. At some $\omega_l = \omega_k$, $\mathrm{Re}[P(\omega)] = \mathrm{Re}[P(\omega_l - \omega)]$ and $\mathrm{Im}[P(\omega)]$ turn to infinity and the perturbation theory fails. Near such ω_k, $I \sim |\omega_l - \omega_k|^{-1} \sim |t - t_k|^{-1/2}$, i.e., a sharp burst of phonons is generated causing a relaxation jump (t_k corresponds to the time moment when $\omega_l = \omega_k$).

Note that v^2_m is not a linear function of the energy of the SLS. Therefore the relaxation of the SLS is always nonexponential, including the case when perturbation theory is applied. In the latter case $E_l(t) \sim (t_l - t)^\alpha$, where $\alpha \sim 0.75 \ldots 0.83$, and t_l is the lifetime of the SLS which is finite in this approximation.

As an example the quantum decay of an odd SLS is examined below. The properties of the SLS depend essentially on the dimensionless parameter $\delta = \sqrt{K^2_3/(K_2K_4)}$. For realistic one-well pair potentials this parameter has a value of between 1 and $\sqrt{32/9}$. For Ar-Ar and Kr-Kr, potentials δ

$=1.37$; for K-Br, potential $\delta=1.31$. For such values of δ the odd SLS is well localized if $\omega_l \gtrsim 1.3\omega_D$. In this case SLS is localized on three central atoms: $A_1=A_{-1}\simeq -A_0/2$, $|A_n| \lesssim 0.05|A_0|$, $n\geqslant 2$; $\xi_n=-\xi_{-n}\simeq\xi$, $n\geqslant 1$, $\xi_0=0$.[3,4] This allows one to take account of only A_0, $A_1=A_{-1}\simeq A_0/2$ and $\overline{\xi_1}=-\overline{\xi_{-1}}\simeq\xi$. The parameters A_0 and ξ of the classical SLS problem in this approximation are determined by Eqs. (5) $(n=0)$ and (6) $(n=1)$:

$$\omega_l^2=3\overline{K_2}+6\overline{K_3}\xi+\tfrac{81}{16}\overline{K_4}A_0^2+9\overline{K_4}\xi^2, \qquad (12)$$

$$0=\overline{K_2}\xi+\overline{K_3}(A_0^2+\xi^2)+\overline{K_4}\xi\left(\tfrac{27}{8}A_0^2+\xi^2\right). \qquad (13)$$

Introducing dimensionless amplitude $a=\sqrt{\overline{K_4}A_0^2/\overline{K_2}}$ and shift $z=\xi\sqrt{\overline{K_4}/\overline{K_2}}$, one gets

$$a^2=-z(z^2+z\delta+1)/\left(\delta+\tfrac{27}{8}z\right), \qquad (14)$$

where z is determined by the real solutions of the equation

$$\omega_l^2=\frac{3}{4}+\frac{81}{64}\,\frac{z\left(5z^2+\frac{43}{9}z\delta+\frac{32}{27}\delta^2-1\right)}{\delta+\frac{27}{8}z} \qquad (15)$$

(here and below the units $\omega_D=1$ are used). This is a cubic equation for z which for considered ω_l and δ has only one solution with $a^2>0$. This solution determines the parameters of the SLS in the classical limit.

In the approximation considered, the SLS causes changes only in central and next to central springs: $k_0=k_1=(16z\delta+24z^2+27a^2)/32$, $k_2=k_{-1}=3a^2/32$, $w_0=-w_1=a(\delta+3z)/8$, $w_2=-w_{-1}=a\delta/8$. In this case $\hat{W}_1$ and $\hat{W}_t$ can be diagonalized analytically. The eigenvalues η_i and v_m and the components of the eigenvectors are the following: $\eta_1=0$,

$$\eta_{2,3}=(k_2+3k_0/2)\pm[(k_2-3k_0/2)^2+k_2]^{1/2},$$

$$\eta_{4,5}=(k_2+k_0/2)\pm[(k_2-k_0/2)^2-k_2]^{1/2},$$

$$v_3=0,\quad v_{1,2,4,5}=\pm\tfrac{1}{2}[8w_2^2+6w_1^2+4w_1w_2 \pm 2(16w_2^4+8w_1^2w_2^2+16w_2^3w_1 +9w_1^4+12w_1^3w_2)^{1/2}]^{1/2}$$

($++$ corresponds to v_1, $-+$ to v_2, $+-$ to v_4, and $--$ to v_5);

$$S_{kn}=\tilde{S}_{kn}/\sqrt{\textstyle\sum_n \tilde{S}_{kn}^2},\quad \tilde{S}_{1n}=\tilde{S}_{k,1}=1,\quad \tilde{S}_{k2}=1-\eta_k/\beta,$$

$$\tilde{S}_{k3}=-4+2\eta_k/\beta,\ \ \tilde{S}_{k4}=1-\eta_k/\beta,\ \ \tilde{S}_{k5}=1,\ \ k=2,3,$$

$$\tilde{S}_{k3}=0,\ \ \tilde{S}_{k4}=-1+\eta_k/\beta,\ \ \tilde{S}_{k5}=-1,\ \ k=4,5.$$

$$s_{mn}=\tilde{s}_{mn}/\sqrt{\textstyle\sum_n \tilde{s}_{mn}^2},\quad \tilde{s}_{i1}=-\sum_{n=2}^{5}\tilde{s}_{in},$$

$$\tilde{s}_{i2}=1-v_i(a_i-w_1/w_2+1)/w_1,\ \ \tilde{s}_{i3}=1,\ \ \tilde{s}_{i4}=1+a_i,$$

$$\tilde{s}_{i5}=1+v_i/w_2,\ \ a_i=v_i(v_i+w_1+2w_2)/(w_1w_2).$$

Energy of the SLS equals

$$E_l\simeq\hbar\omega_D\mathcal{K}\varepsilon_l;\quad \varepsilon_l=3\omega_l^2a^2+z^2\left(\tfrac{1}{2}+z/3+z^2/4\right); \qquad (16)$$

one sees that parameter $\mathcal{K}$ determines the quantum scale of the SLS energy. Note that the intensity of phonon emission does not depend on $\mathcal{K}$ [the unit of I in Eq. (9) is $\hbar\omega_D^2$]. Consequently, parameter $\mathcal{K}$ determines the time scale of energy relaxation.

We performed calculations of the frequency dependence of the intensity of phonon emission by the SLS and of the time dependence of the intensity of phonon emission and the SLS energy (at the temperature $T=0$). The calculation procedure was the following. First we fixed the initial SLS frequency (usually at $\omega_l=1.98\omega_D$) and calculated z, a^2, E_l, $G(\omega)$, I. The second step was to take a lower value of ω_l and respectively calculate new values of parameters. We repeated this procedure until the frequency of the SLS got a value of 1.3. The time step was calculated using the relation $\Delta t=-\Delta E_l/I$.

In Fig. 1 the dependence of the intensity of phonon emission on the value of ω_l is shown. Indeed, the intensity I has peaks $\sim|\omega_l-\omega_k|^{-1}$ at critical frequencies ω_k (this was confirmed numerically). This means that the SLS is unstable in the vicinity of ω_k. The time dependences of the intensity of phonon emission and of the mode energy E_l are given in Fig. 2.

In all cases the lowest considered frequency $\omega_l=1.3\omega_D$ corresponds to a small energy as compared to the initial energy for $\omega_l\approx 2\omega_D$. Consequently, the main part of the energy of the SLS with initial frequency $\omega_l\sim 2\omega_D$ is lost during the time considered [note that an extrapolation of $E_l(t)$ to larger t also gives the finite lifetime of SLS]. Depending on the value of δ and on the initial energy, this time can be either longer or shorter than that given by the perturbation theory. The relaxation law is essentially nonexponential: sharp bursts of phonons are generated at critical values of time t_k when ω_l approaches ω_k, causing relaxation jumps.

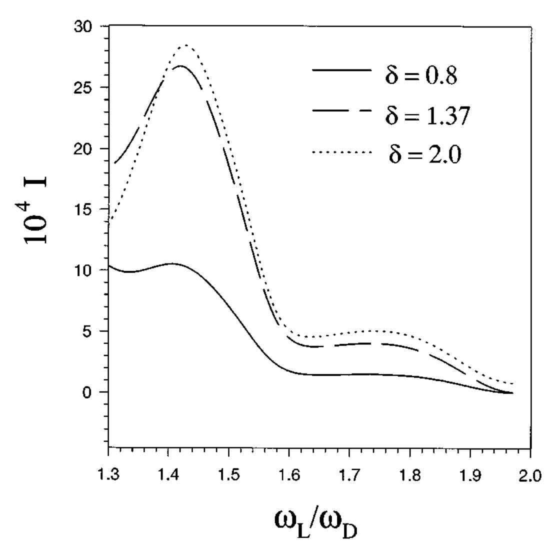

FIG. 1. Frequency dependence of the intensity of emission of phonons by the odd SLS. The peaks correspond to the regions where the SLS is unstable with respect to quantum fluctuations; $\delta=\sqrt{K_3^2/K_2K_4}$.

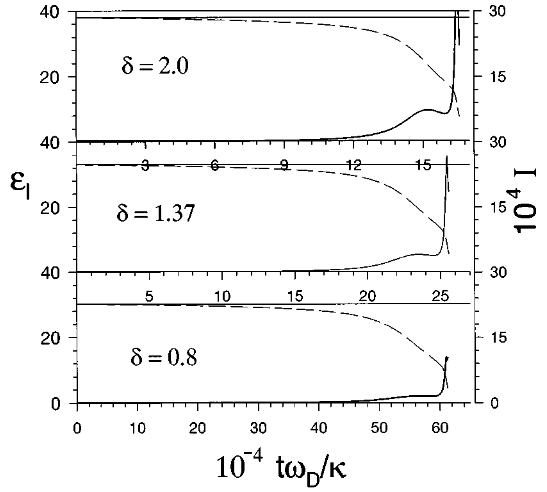

FIG. 2. Time dependence of the SLS energy E_l (thick dashed line) and intensity of phonon emission by the SLS I (thick solid line); decay of the SLS energy calculated with the perturbation theory (thin solid line). Sharp peaks in the $I(t)$ dependence describe emission of phonon bursts by the SLS. $E_l=\varepsilon_l K_2^2/K_4$, $\mathcal{K}=K_2^2/(K_4\hbar\omega_D)$. I is measured in units of $\hbar\omega_D^2$.

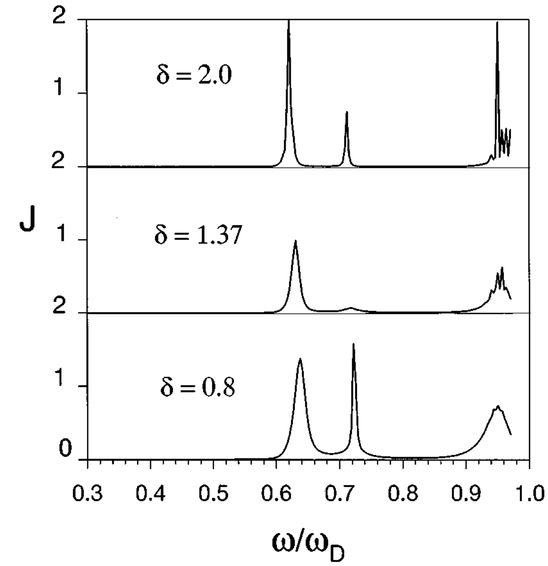

FIG. 3. Spectrum of generated phonons $J(\omega)$ (in a.u.).

The time dependence of the intensity I near t_k is $I\sim|t-t_k|^{-1/2}$. The peaks mentioned are caused by the poles in the integrand function Eq. (9); they correspond to the emission of quasimonochromatic phonons and give sharp lines in the spectrum of generated phonons $J(\omega)$ (the integrand function in (9) summed over time) in Fig. 3.

In the case $\delta^2<3/4$ there may exist[2] large-size SLS with ω_l close to ω_D. The energy of such an SLS $E_l\sim 8\sqrt{2(\omega_l-1)}K_2^2/[K_4(3-4\delta^2)]$ exceeds the unit $(\hbar\omega_D)$ only if δ^2 approaches 3/4 from below. The problem of quantum decay of this large-size SLS is analogous to that of solitons in continuous media and it will be considered elsewhere.

In conclusion, we examined the quantum relaxation of the odd SLS in a one-dimensional anharmonic monatomic lattice, caused by the zero-point fluctuations of the phonons and gave an analytical nonperturbative solution of the problem. We found that the fluctuations can cause generation of very sharp bursts of quasimonochromatic phonons and very fast, stepwise jumps of the SLS energy. The rate of energy loss is on average of the order of the phonon quantum $\hbar\omega_D$ per period of vibrations $2\pi/\omega_D$. The full relaxation time is determined by the quantum anharmonicity parameter $\mathcal{K}$; in chains with potentials corresponding to the quantum crystals, this time is of the order of a period of vibrations, while in lattices with potentials of ordinary crystals it may reach thousands of periods.

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