

Molecular-dynamics simulation of the decay kinetics of uniform excitation of an anharmonic 1D chain

V. M. Burlakov and S. A. Kiselev

Institute of Spectroscopy, Academy of Sciences of the USSR

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The decay of the top-band phonon mode has been studied by molecular-dynamics simulation in monatomic and diatomic anharmonic 1D lattices (chains). Distinct stages are found in the decay. An excitation of small-radius localized intrinsic vibrations is also found. These vibrations draw energy from the surrounding spatial regions. Some physical realizations of the observed effects are discussed.

INTRODUCTION

The problem of seeking stable excitations of nonlinear systems arises both in nonlinear optics and quantum electronics, on the one hand, and in several problems in solid state physics, on the other. Among the latter problems are those involving vibrational excitations of polymer chains¹ and the appearance of superstructures in the course of ferroelectric phase transitions (see the review by Strukov²). The form of the stable solutions depends on the nature and magnitude of the nonlinearity of the potential, which in a real physical system may in turn depend on several external parameters (the pressure and the temperature) as well as the excitation amplitude. The purpose of the present study was accordingly not to search for the stable solutions themselves but to study the process by which an excitation which is uniform in terms of amplitude and is stable in a harmonic system decays in a 1D chain with a nearest-particle interaction potential which includes terms of the third and fourth orders in the anharmonicity. That a uniform classical excitation of a nonlinear system is unstable is a fairly well-known fact.³ Distinctive features of the system which we have selected are its discrete nature and its incomplete integrability. These features are linked with some effects found below: a pronounced concentration of energy in an intermediate stage of the decay and the establishment of a thermal equilibrium (a uniform distribution of energy among the various degrees of freedom) in the chain.

The procedure for the numerical simulation is as follows. We begin with a chain of particles with a nearest-neighbor interaction potential

$$U(r) = \frac{K_2}{2} r^2 + \frac{K_3}{3} r^3 + \frac{K_4}{4} r^4, \quad (1)$$

where K_2 , K_3 , and K_4 are constants of the potential. At the initial time we specify a configuration of particle displacements u_n from their equilibrium positions, [$u_n = (-1)^n A_0$, where A_0 is the mode amplitude corresponding to the phonon mode (which we will call Q_m) with the wave vector $k = \pi/h$, where h is the lattice constant]. The displacements of the particles at later times are found from the equations of motion and cyclic boundary conditions.⁴⁻⁶ A small perturbation (with a size less than 0.1% in terms of amplitude) is specified at the initial time in order to initiate the decay of the mode Q_m . This perturbation does no more than tie-in the beginning of the decay with the time $T = 0$, since a decay would occur even in the absence of this perturbation, because of a buildup of the error of the numeri-

cal integration. This error, however, depends on the value of A_0 in a complex way, so the initial time turns out to be shifted with respect to $T = 0$, and it becomes difficult to analyze the dynamics of the process.

RESULTS OF THE NUMERICAL SIMULATION AND DISCUSSION

Monatomic chain

1. Figure 1 shows a typical decay of an original perturbation. Specifically, this figure shows the time evolution of the maximum energy in a chain of 100 particles. We can distinguish three stages on this curve.

1) First comes the actual decay of the original perturbation into a series of localized excitations of the envelope-soliton type—"intrinsic (natural) localized modes"⁶⁻⁹—occurs in interval OA, which amounts to about $100T_m$, where T_m is the vibration period of the excited phonon mode in the harmonic approximation. The energy distribution in the chain shown in Fig. 2a corresponds to this stage of the decay. We see that an energy several times the original energy is concentrated at several of the particles.

2) Next comes a redistribution of energy among the intrinsic localized modes (this is interval AB, which covers the time interval from $100T_m$ to $700T_m$). As a result of this

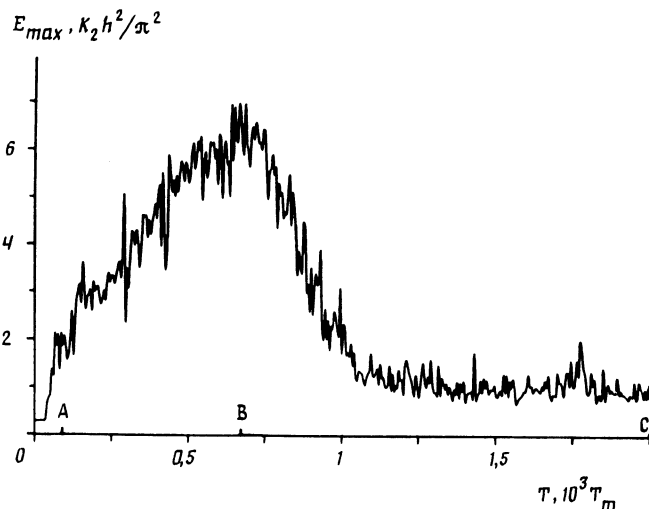


FIG. 1. Time evolution of the maximum energy E_{max} localized at a particle in the course of the decay of mode Q_m in a chain of 100 particles of mass $m = 1$. The initial amplitude of the mode is $A_0 = 0.1h$; the anharmonicity parameter is $\lambda A_0^2 = 0.10$, where $\lambda = K_4/K_2$.

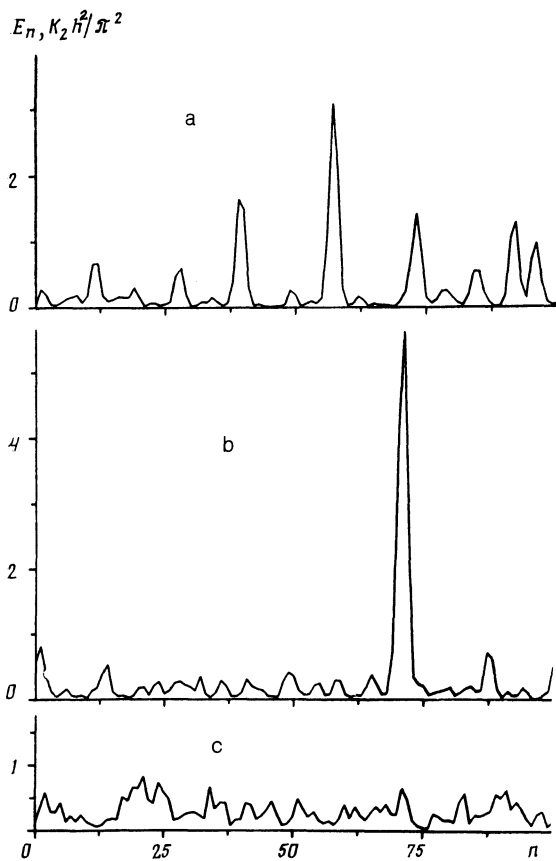


FIG. 2. Distribution of the energy E_n in a chain of 100 particles of mass $m = 1$ during the decay of mode Q_m , at various times. a— $T = 200T_m$; b— $T = 700T_m$; c— $T = 1500T_m$. The anharmonicity parameter is $\lambda A_0^2 = 1.21$.

redistribution, the concentration of energy becomes even more pronounced (Fig. 2b). More than 50% of the entire energy of the chain is concentrated in three or four intrinsic localized modes which form, and in the most intense of these modes the maximum energy per particle is in fact tens of times the average energy.

3) Next comes the decay of the intrinsic localized modes (interval BC, covering the time interval from $700T_m$ to $2000T_m$). As a result of this decay, the energy turns out to be distributed more or less uniformly along the chain again.

The subsequent evolution of the system does not cause any changes in either the spatial or spectral distribution of the energy.

In the spectral representation, the process by which mode Q_m having the maximum wave vector decays has two characteristic stages. The first is characterized by a redistribution of energy between the top-band modes of the chain and the intrinsic localized modes. Figure 3a shows the mode spectrum of a chain of 40 particles in this stage of the decay of Q_m . The highest-frequency peak in the spectrum in Fig. 3a corresponds to intrinsic localized modes whose vibration frequencies lie above the band of phonon modes. The peak at frequencies near ω_m ($\omega_m = 2\pi/T_m$) corresponds to the upper edge of the phonon band, which is the position of the spectrum of the Q_m vibrations. The low-frequency states are filled only slightly in this stage. Next comes a redistribution of energy from the top-band part of the spectrum to the bot-

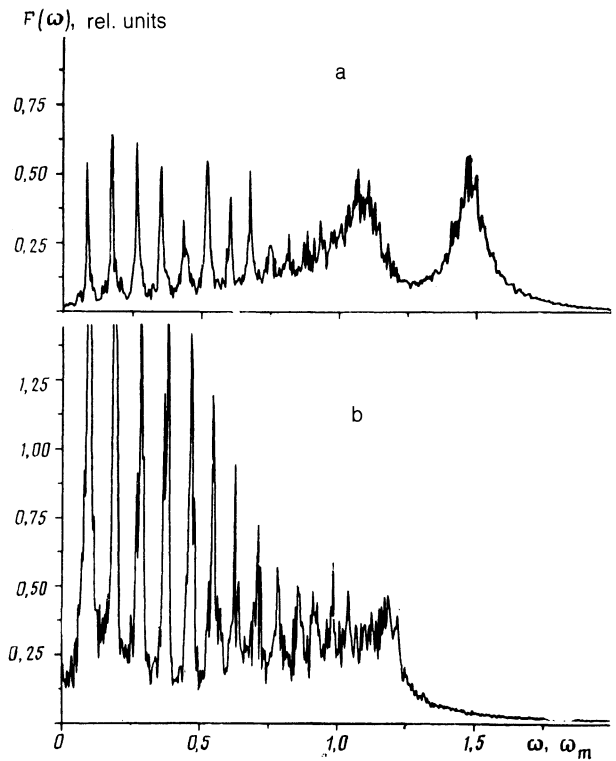


FIG. 3. Fourier spectrum $F(\omega)$ of the vibrations of a chain of 40 particles of mass $m = 1$ during the decay of mode Q_m . a—During the existence of the intrinsic localized modes, at $T \approx 700T_m$; b—after the decay of these modes, at $T \approx 4000T_m$. The anharmonicity parameter is $\lambda A_0^2 = 0.10$.

tom-band part. In a thermalized chain the amplitude of the bottom-band modes is more than an order of magnitude greater than that of the top-band modes (Fig. 3b). The distribution of the amplitude of the Fourier spectrum, $F(\omega)$, in the final phase is indeed characteristic of a thermal equilibrium, since the law $F(\omega)^2 \omega = \text{const}$ holds.

2. Let us take a more detailed look at each of these stages in the decay of mode Q_m .

1) The actual decay of Q_m occurs exponentially; more precisely, the increase in the amplitude of the intrinsic localized modes occurs exponentially (Fig. 4). The time constant of the decay depends on the relative nonlinearity of the potential (see the caption of Fig. 4).

The stability properties of mode Q_m in 1D chains with a particle interaction potential as in (1) (a Fermi-Pasta-Ulam lattice) have been studied theoretically by Budinsky and Bountis.¹⁰ They were interested in the threshold value of the energy per particle, i.e., the value at which an instability first arises, and also the asymptotic value of this threshold energy as the number of particles increased. We are interested primarily in the rate of decay of mode Q_m as a function of the amplitude and the characteristic wave vector of the excitation generated during the decay of Q_m . We will study the decay process in the continuum approximation. The corresponding equations of motion are

$$\rho u_{tt} - 2(K_2 + \frac{3}{2}K_4 u_z u_z^*) u_{zz} - \frac{3}{2}K_4 u_z^2 u_{zz}^* = 0, \quad (2)$$

where ρ is the mass density of the chain; $u(t, z)$ is the displacement of a unit length element from its equilibrium posi-

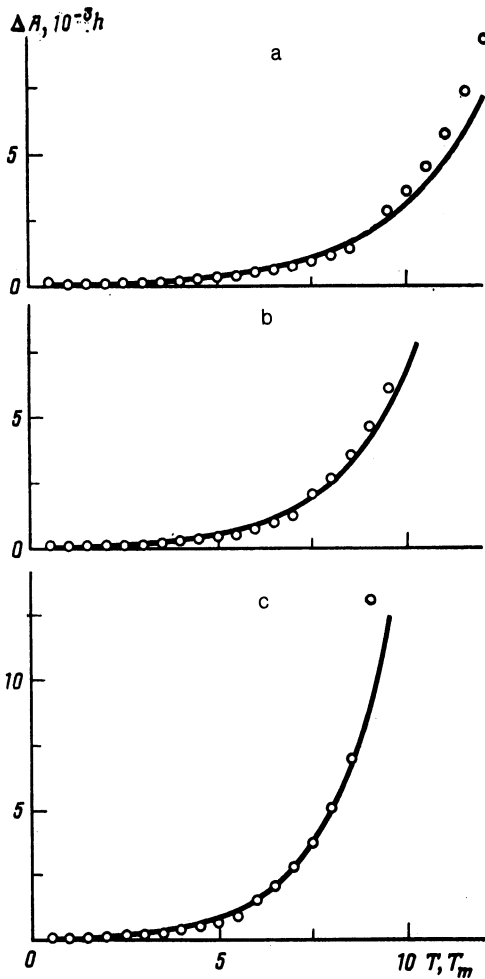


FIG. 4. Exponential growth of the amplitude ΔA in the initial stage of the decay of mode Q_m (region OA in Fig. 1) for various initial amplitudes: a— $A_0 = 0.09h$; b— $A_0 = 0.10h$; c— $A_0 = 0.11h$. The anharmonicity parameter is $\lambda h^2 = 10$. Points—Values found by the molecular-dynamics method; solid lines—best fit of the exponential function $\exp(T/s)$, with (a) $s = 2.36$, (b) 1.98, and (c) 1.70.

tion; u_{tt} , u_z , and u_{zz} are respectively the time derivative and the spatial derivatives of the displacement; and K_2 and K_4 are parameters of potential (1). From (2) we easily find the vibration frequency ω_0 of mode Q_m :

$$\omega_0^2 = \frac{2k^2}{\rho} \left(K_2 + \frac{3}{4} K_4 k^2 A_0^2 \right), \quad (3)$$

where k and A_0 are the wave vector and amplitude of Q_m $\{Q_m = A_0 \exp[-i(\omega t - kz)]\}$, respectively. The stability of Q_m is analyzed by the standard method here, through the addition of a small perturbation $\xi(t, z)$ to the original excitation:

$$\begin{aligned} \bar{Q}_m &= A_0 \exp[-i(\omega t - kz)] + \xi(t, z) \\ &= A_0 \exp[-i(\omega t - kz)] + \varphi(t) \psi(z). \end{aligned} \quad (4)$$

The functions $\varphi(t)$ and $\psi(z)$ depend on the time and on the coordinate, respectively. Substituting (4) into (2), and using (3), we find

$$\frac{\varphi''}{\varphi} - \frac{\omega_0^2}{k^2} \frac{\psi''}{\psi} = \frac{3iK_4 k^2 A_0^2}{\rho} \frac{\psi'}{\psi''}. \quad (5)$$

We seek a solution in the form

$$\psi = \psi_0 e^{-ikz}, \quad \varphi = \varphi_0 e^{Tt}. \quad (6)$$

Substituting (6) into (5), we find

$$\gamma^2 = \frac{3K_4 k^2 A_0^2}{\rho} \kappa - \frac{\omega_0^2}{k^2} \kappa^2. \quad (7)$$

It can be seen from (7) that the decay of Q_m involves a threshold if a perturbation wave vector κ is given, since a decay in (7) corresponds to the requirement $\gamma^2 > 0$. If the perturbation has a more or less continuous spectrum of spatial harmonics, one can determine that value of κ at which γ has its maximum value. We denote this particular value by $\hat{\kappa}$. Differentiating (7) with respect to κ , we find

$$\hat{\kappa} = k \Delta \omega^2 / \omega_0^2, \quad (8)$$

where $\Delta \omega^2$ is the anharmonic shift of the square of the frequency.

The value found for κ with the help of (8) agrees fairly well with that observed in the numerical simulation.

In precisely the same way we can use (7) to estimate the decay rate (more precisely, the rate at which the amplitude of the intrinsic localized modes increases). We find the following expression for $\hat{\gamma}$ (per $\hat{\kappa}$):

$$\hat{\gamma} = \Delta \omega^2 / \omega_0. \quad (9)$$

A substitution of the numerical values into (9) again reveals a satisfactory agreement with the experimental value of γ .

2) The concentration of energy which occurs during time interval AB results from an inelastic interaction of the intrinsic localized modes with each other. As was shown in Ref. 5, two of these modes may merge to form a single localized mode in the course of an interaction, or these intrinsic localized modes may pass through each other without undergoing any significant loss of energy. On the whole, it is thus in this stage that the bottom-band phonons form. These phonons play the most important role in the decay of the small-radius intrinsic localized modes. Specifically, in the first stage of the decay of mode Q_m the chain contains top-band phonons, envelope solitons (the peak in the spectrum at $\omega \approx \omega_m$ in Fig. 3a), and intrinsic localized modes (the highest-frequency peak in the spectrum in Fig. 3a).

According to Flytzanis *et al.*¹¹ (see also the bibliography in Ref. 11), the evolution of the excitations of the first two types is described by a nonlinear Schrödinger equation, and these excitations do not interact with each other. Consequently, generation of low-frequency modes can be expected in processes in which intrinsic localized modes interact with envelope solitons and with top-band phonons. Figure 5, a and b, shows a collision of a soliton with immobile intrinsic localized modes. The amplitude of the latter modes in the second case is higher by a factor of $5^{1/2}$ than the amplitude of the intrinsic localized modes in Fig. 5a. The soliton has the same amplitude and the same velocity in the two collisions.

In a collision of an envelope soliton with a local mode whose anharmonicity is relatively slight (Fig. 5a), only a small part of the soliton is reflected from the intrinsic localized mode, while in a collision of a soliton with a highly anisotropic intrinsic localized mode nearly half the soliton is reflected back. The local mode, in contrast, undergoes almost no change as a result of the collision in the second case, and in the first case it recovers its shape after a rather long time. In each case, after one collision of a soliton with an intrinsic localized mode this mode nearly recovers the initial value of its amplitude.

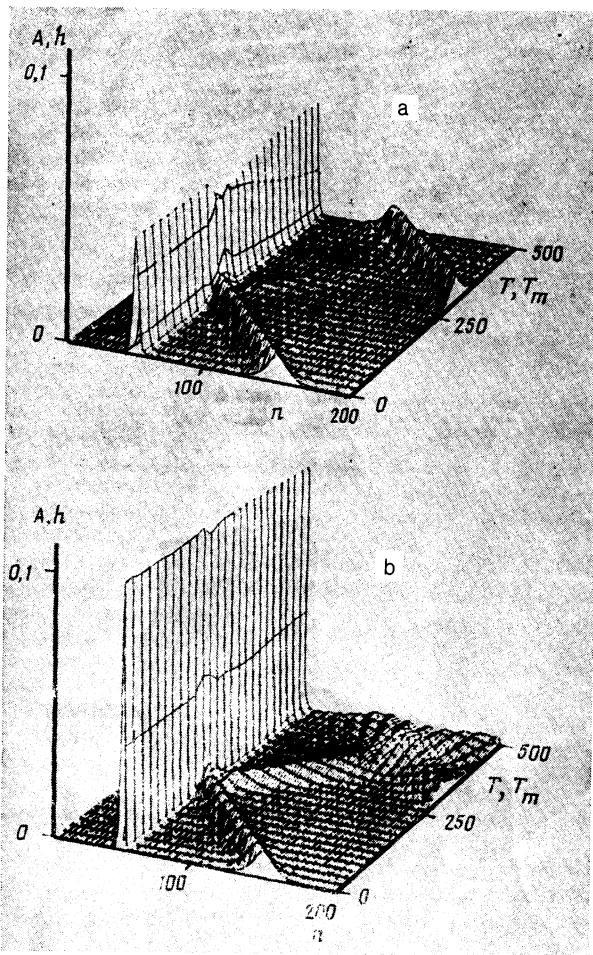


FIG. 5. Collision of an envelope soliton of amplitude $A_s = 0.01h$ with a local mode of amplitude (a) $A_1 = 0.1h \cdot (0.2)^{1/2}$ and (b) $A_2 = 0.1h$ in a chain of 200 particles of mass $m = 1$, with cyclic boundary conditions. The vibration amplitude A of the particles is plotted along the vertical axis. The anharmonicity parameter is $\lambda h^2 = 10$.

Figure 6, a and b, shows vibration spectra of particles of the chain far from an intrinsic localized mode after about four collisions of a soliton with a local mode. In each spectrum, the highest-frequency peak corresponds to the carrier frequency of the soliton. To the left of this peak we see two smaller peaks in each case, which correspond to bottom-band excitations generated in the course of the collision of the soliton with an intrinsic localized mode. The frequency difference between these excitations corresponds precisely to the frequency difference between the local mode and the soliton (the particle vibration spectra shown in Fig. 6, a and b, are for particles far from the immobile intrinsic localized mode, so the frequency of the latter mode is not seen).

In the case of an intrinsic localized mode which is only slightly anharmonic, with the local-mode frequency ω_1 ($\omega_1 \approx 1.08\omega_m$) lying close to the soliton frequency ω_s ($\omega_s \approx 0.99\omega_m$), excitations are generated in the collision which are more intense and which have higher frequencies than in the case of a collision of a soliton with a very anisotropic intrinsic local mode ($\omega_2 \approx 1.36\omega_m$). This circumstance is reflected in the intensity of the low-frequency peaks in Fig. 6, a and b. One might suggest that a collision of a bottom-band phonon with a local mode involves a corre-

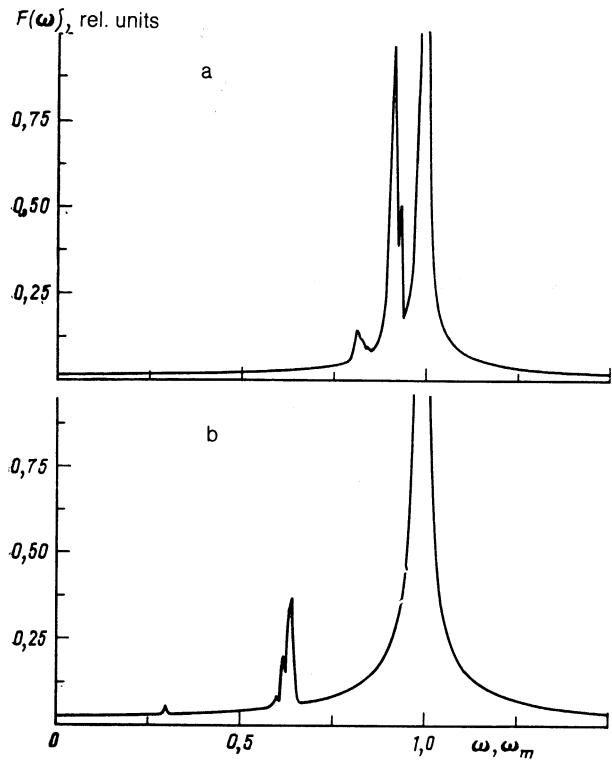


FIG. 6. Fourier spectrum $F(\omega)$ of the vibrations of a particle of a chain of mass $m = 1$ far from an intrinsic localized mode, after four collisions of an envelope soliton with a local mode. The amplitude of the envelope soliton is $A_s = 0.01h$. The amplitude of the intrinsic localized mode is (a) $A_1 = 0.1h \cdot (0.2)^{1/2}$ or (b) $A_2 = 0.1h$, in correspondence with Fig. 5. The high peak at the right corresponds to the soliton frequency.

sponding interaction process, in which a bottom-band excitation is created, since the motion of the phonon obeys a nonlinear Schrödinger equation, as the motion of a soliton does.

3) In the final stage of the thermalization of the chain, the localized modes decay against the background of generation of bottom-band modes. To identify the mechanism for the decay of the intrinsic localized modes, we consider the conditions for the existence of a local mode in a chain in which a low-frequency mode is excited.

The equation of motion for the particles of a chain with a quartic anharmonicity is

$$m\ddot{u}_n = K_2(u_{n+1} + u_{n-1} - 2u_n) + K_4[(u_{n+1} - u_n)^3 + (u_{n-1} - u_n)^3], \quad (10)$$

where m is the mass of the particles, and u_n are the displacements of the particles of the chain. The particle displacements corresponding to a low-frequency mode are

$$u_n(t) = A_q \cos(\omega t - qhn), \quad (11)$$

where A_q is the amplitude of the mode which has a wave vector q and a frequency ω , and h is the chain constant. Assuming that the size of the intrinsic localized mode is small in comparison with $1/q$, and assuming that the frequency ω_l of the localized mode is significantly higher than the frequency of the low-frequency mode, ω , we consider the equation of motion of an intrinsic localized mode against the background of a constant gradient of displacements of the chain particles. Singling out the slowly varying gradient, we seek the displacements in the form

$$u_n(t) = -A_q q h n + v_n(t), \quad (12)$$

where $v_n(t)$ are displacements which describe the local mode. Substituting (12) into (10), we easily find the following equations for $v_n(t)$:

$$\begin{aligned} m\ddot{v}_n = & (1 + 3\lambda A_q^2 q^2 h^2) K_2 (v_{n+1} + v_{n-1} - 2v_n) \\ & - 3A_q q h K_4 [(v_{n+1} - v_n)^2 - (v_{n-1} - v_n)^2] \\ & + K_4 [(v_{n+1} - v_n)^3 + (v_{n-1} - v_n)^3], \end{aligned} \quad (13)$$

where $\lambda = K_4/K_2$ is again the anharmonicity parameter. Equation (13) describes vibrations in a chain with anharmonicities of third and fourth orders, in a nearest-neighbor potential. In other words, a low-frequency mode has created for the intrinsic localized modes an effective potential

$$V_q(r) = \frac{Q_2}{2} r^2 + \frac{Q_3}{3} r^3 + \frac{Q_4}{4} r^4, \quad (14)$$

where

$$\begin{aligned} Q_2 &= (1 + 3\lambda A_q^2 q^2 h^2) K_2, \\ Q_3 &= -3A_q q h K_4, \\ Q_4 &= K_4. \end{aligned} \quad (15)$$

The question of the decay of an intrinsic localized mode against the background of low-frequency modes which have appeared now reduces to the question of the existence of local modes in a chain with anharmonicities of third and fourth order. We have accordingly studied the stability of intrinsic localized modes as a function of the relation between the anharmonicity constants Q_3 and Q_4 in potential (14). A local mode was excited through a displacement of two neighboring particles toward each other, while the other particles remained at rest in their equilibrium positions. The amplitude of the initial displacements of the two particles which were moved was chosen to lie above the threshold for the excitation of intrinsic localized modes, in accordance with Refs. 4 and 5. The degree of stability of intrinsic localized modes in a potential of this sort was estimated from the fraction of the energy which formed a local mode at the particles which were initially moved. Studies of potentials with various relations between the anharmonicities of third and fourth orders showed that the stability of intrinsic local modes falls off as the shape of the particle interaction potential approaches that of a Toda potential¹²

$$V_T(r) = \frac{a}{b} \exp(-br) + ar - \frac{a}{b}, \quad (16)$$

where a and b are constants ($ab > 0$). For a chain with potential (16), a localized mode does not form, and an initial perturbation leaves the region of the initial excitation in the form of solitons. Correspondingly, for a chain of particles with a nearest-neighbor potential (14) the localized modes quickly decay if the relations among the constants Q_2 , Q_3 , and Q_4 correspond to the coefficients of the first terms in a Taylor-series expansion of Toda potential (16) in powers of r near the equilibrium position. The equation which we are seeking is

$$\frac{Q_4}{Q_3} = - \left(\frac{2}{3} \frac{Q_4}{Q_2} \right)^{1/2}. \quad (17)$$

The absence of localized solutions from a chain with a Toda potential also implies the absence of envelope solitons.⁸ Substituting expressions (15) into (17), we find a condition on

the amplitude (A_q) of a low-frequency mode with a wave vector q whose appearance would result in a complete destruction of the local modes:

$$A_q q = \left(\frac{3^{1/2} - 1}{6\lambda h^2} \right)^{1/2}. \quad (18)$$

Let us apply condition (18) to the case of the decay of intrinsic localized modes during the thermalization of a chain of 100 particles, as discussed above. The typical size of the localized modes can be taken to be about $4h$ (Fig. 2b), so a phonon with a wave vector $q \approx \pi/4h$ will create the gradient which we need in order to destroy the intrinsic localized modes. Substituting the value $\lambda h^2 = 10$ for the anharmonicity parameter for the particular potential used into (18), we find the amplitude of a phonon which will destroy the intrinsic localized modes:

$$A_q \approx 0,15h. \quad (19)$$

Results found on the decay of mode Q_m , as shown in Figs. 1 and 2, demonstrate that the destruction of the intrinsic localized modes occurs before the amplitude of the low-frequency modes reaches the value $\approx 0,3h$, in agreement with (19).

3. Let us examine the thermalization of a chain with an initial excitation of mode Q_m as the amplitude of this excitation is varied. This variation of the excitation amplitude causes a variation of the extent to which potential (1) is nonlinear. Figure 7 shows trajectories of the time evolution

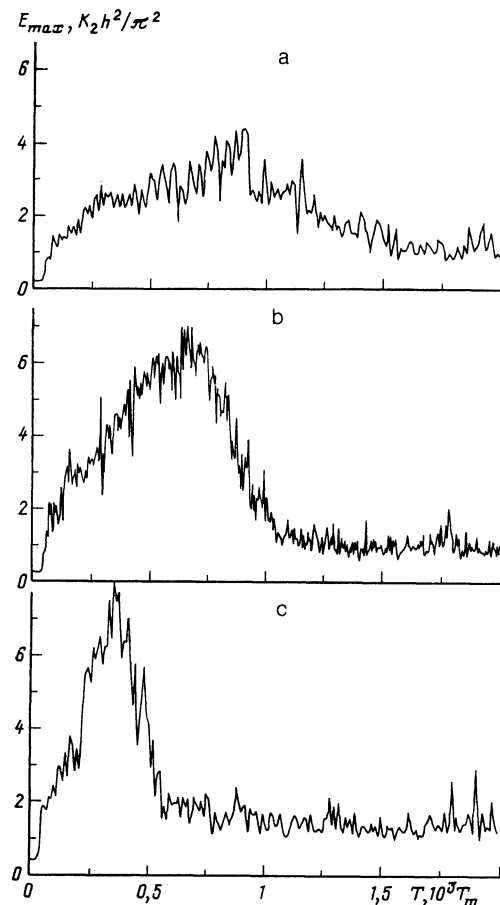


FIG. 7. Time evolution of the maximum energy E_{\max} which is localized at a particle for various initial values of the amplitude and anharmonicity of mode Q_m . a— $A_0 = 0.10h$, $\lambda A_0^2 = 0.10$; b— $A_0 = 0.11h$, $\lambda A_0^2 = 0.121$; c— $A_0 = 0.13h$, $\lambda A_0^2 = 0.169$. The chain contains 100 particles of mass $m = 1$.

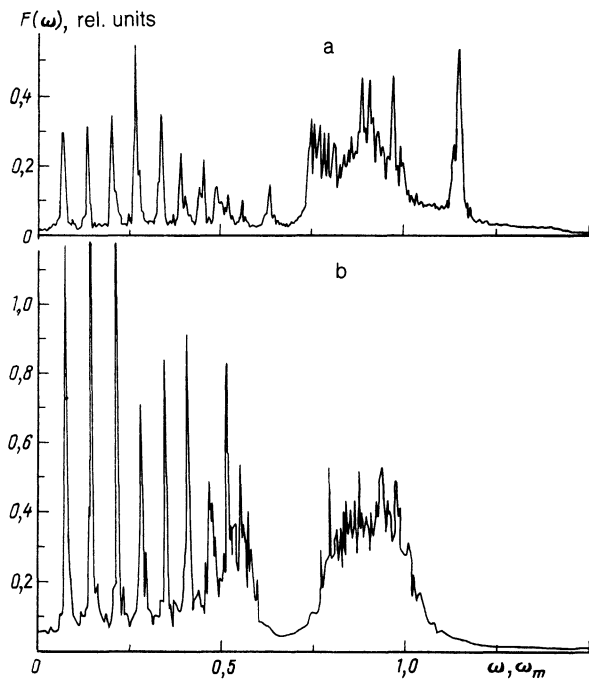


FIG. 8. Fourier spectrum $F(\omega)$ of the vibrations of a diatomic chain ($m_1 = 1$, $m_2 = 2$) of 40 particles during the decay of mode Q_m at (a) $T \approx 2800T_m$ and (b) $T \approx 10700T_m$. The anharmonicity parameter is $\lambda A_0^2 = 0.10$.

of the maximum energy E_{\max} in the chain for various amplitudes Q_m . We observe a pronounced contraction of the time intervals of all the stages of the decay which we discussed above as the anharmonicity increases. A 30% increase in the amplitude (compare Fig. 7c with Fig. 7a), for example, shortens the complete thermalization time from $T \approx 1600T_m$ to $T \approx 600T_m$, i.e., by a factor of nearly 3. The relative increase in E_{\max} which occurs in the intermediate stage is roughly the same in all three cases.

Diatomic chain

The process by which Q_m decays basically involves the same stages as in a diatomic chain, but the presence of a gap in the vibration spectrum introduces a qualitatively new result: Complete thermalization does not occur. The reason is not difficult to see, when we go back to the mechanism for the filling of the phonon states. A generation of low-frequency phonons occurs as the result of an interaction of a small-radius intrinsic localized mode with envelope solitons and with high-frequency phonons. The higher the energy of the intrinsic localized mode (the smaller the localization radius), the lower the frequency of the phonon which is generated. The gap in the spectrum of a diatomic chain has the consequence that a threshold is involved in the generation of low-frequency excitations. Consequently, after the intrinsic localized mode has lost a fraction of its energy above the threshold value, the general thermalization comes to a halt, and a thermalization occurs only within each band (Fig. 8).

CONCLUSION

From this study of the decay kinetics of a uniform classical excitation of a nonlinear chain we can draw several conclusions which concern the dynamics of nonlinear sys-

tems as such and which are applicable to certain problems in the solid state physics. The most important points in our opinion are the following:

- the comparatively rapid decay of an initial excitation into randomly positioned intrinsic localized modes, with a decay rate which increases with increasing K_4 ;
- the high concentration of energy in small-radius intrinsic localized modes;
- the governing role played by the small-radius intrinsic localized modes in the relaxation to a thermal equilibrium.

The first two of these consequences are exceedingly important for concrete physical realizations. The rapid and irreversible decay of mode Q_m as the nonlinearity of the vibrations increases may reflect the behavior of the damping of a soft mode near a structural phase transition. Near the temperature phase-transition T_c , the nonlinearity of the soft-mode potential (K_4) increases, at the expense of K_2 , which decreases (Ref. 13), the quasiharmonic frequency $\omega_m(k_{cr})$ becomes comparable to the anharmonic increments, and the soft mode itself becomes a classical excitation. According to (9), γ increases to the extent that the mode may become a relaxation mode. Since γ is very sensitive to k , the spectrum of spatial fluctuations of the order parameter may contain intense harmonics with $k \ll k_{cr}$. In other words, we might expect that modulated and random phases would arise in such systems. The key factor for this conclusion is the quasi-one-dimensional nature of the soft-phonon band, rather than the one-dimensional nature of the overall system.

The pronounced concentration of energy which occurs during the decay of a uniform classical excitation of this system may describe a specific mechanism for the generation of point defects in a condensed medium.

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