Computer simulation of intrinsic localized modes in one-dimensional and two-dimensional anharmonic lattices

V. M. Burlakov, S. A. Kiselev, and V. N. Pyrkov
Institute of Spectroscopy, Academy of Sciences of the Union of Soviet Socialist Republics, Troitzk, 142 092 Moscow Region, U.S.S.R.
(Received 12 February 1990)

The properties of stable and movable intrinsic localized vibrations (ILV's) have been investigated by the particle method. It is shown primarily with the help of computer experiments that an ILV in an anharmonic lattice is identical to a localized vibration near a force-constant defect in a harmonic lattice. The temperature dependence of the localization threshold is presented. The differences between ILV's and solitons are discussed. A brief study is also made on the existence of ILV in a two-dimensional square lattice.

INTRODUCTION

It is well understood that the appearance of localized vibrational excitations in a harmonic lattice is caused by mass or force-constant defects.[1,2] It was theoretically predicted recently that the localization may also occur in ideal anharmonic lattices.[3,4] For example, if the vibrational amplitude a for any atom satisfies the condition $a^2 \gg 0.05K_2/K_4$, where K_2 is the harmonic and K_4 the quartic force constants, then according to Ref. 3, there will be a localized vibration with $\omega_l \gg \omega_m$, where ω_m is the top band frequency. It is the relatively large amplitude of the vibration which is a peculiar feature of the localized vibrations (LV) and especially of intrinsic LV 's (ILV's). The ILV is thought to be an interesting object in connection with the problem of local phase transitions (unstable local modes and nucleation sources),[5] and for the thermodynamics of quantum crystals.[4] Here we present the results of investigations of ILV properties in linear one- and two-atomic chains and in a square lattice. Computer experiments have been performed in the range of parameter space where analytical investigation is not possible. A preliminary report of some of these findings has already been made.[6]

SIMULATION TECHNIQUE

The lattice under consideration consists of a linear chain of N particles with nearest-neighbor interactions,

$$V(r)=(K_2/2)(r-r_0)^2+(K_3/3)(r-r_0)^3 + (K_4/4)(r-r_0)^4 , \quad (1)$$

where K_2, K_3, and K_4 are harmonic and two anharmonic force constants. Under an external pressure or for finite temperatures, the lattice constant h is not equal to the minimum r_0 of the potential. Since V is a function of $r-r_0$, the energy does not depend on h and r_0, but only on their difference. We set r_0 equal to h, because one can eliminate the difference $h-r_0$ by a redefinition of force constants K_2, K_3, and K_4. For the Toda lattice this question has been discussed in Ref. 7. In the two-dimensional (2D) square lattice there are four nearest neighbors for any particle. For this reason the equation of motion of the particles should depend not only on the difference between h and r_0, but also on h and r_0 separately.

If it is not specifically mentioned, we set $\hbar=1$, $r_0=1$, and $m=1$, where m is a particle mass for a monatomic chain. We also set $K_2=(2\pi)^2$—this has the advantage that in the limit $K_3, K_4 \to 0$, where V reduces to the harmonic potential, the period of oscillations in this potential equals unity. After this scaling the Hamiltonian reads

$$H=\sum_n [\dot{u}_n^2/2+V(u_{n+1}-u_n)] , \quad (2)$$

where u_n are displacement coordinates.

To characterize the relative contribution of anharmonicity to the vibrational frequency we determine the convenient parameter[3,4]

$$\lambda=3K_4a^2/K_2 . \quad (3)$$

The ILV was excited by providing the initial configuration of particle displacement of various types: (i) only one particle with number i has a nonzero displacement $u_i=u^0$; (ii) two adjacent particles have equal displacements of opposite sign, $u_i=+u^0$ and $u_{i+1}=-u^0$, with remaining elements being at rest; (iii) three adjacent particles have initial displacements $u_{i-1}=-u^0/2$, $u_i=+u^0$, and $u_{i+1}=-u^0/2$, respectively. All three types of excitation will be analyzed below.

After initial excitation, we solve computationally the equations of motion according to the usual procedure.[8] The time step δt was chosen so that the total energy was conserved to within 1% accuracy. This value was $T_m/32$, where $T_m=2\pi/\omega_m$ ($\omega_m=2\sqrt{K_2}=4\pi$) is the period of the top-band vibration in a harmonic case. For the calculations periodic boundary conditions were used.

To characterize the ILV properties, the Fourier component of the particle displacement, $A(\omega,n)$, and the total energy per particle, E, were used. Before the determination of these characteristic parameters, we use up to 1800 steps of randomization. During this time a sharp initial excitation spreads to some adjacent particles and acquires a stationary shape. The function $A(\omega,n)$ was

calculated through the time interval $T_\omega = 512T_m$. Note that the amplitudes of Fourier components contain no information about vibrational and thermodynamic properties of the particle system under consideration because of the absence of ergodicity in these systems.[9]

EXPERIMENTAL RESULTS AND DISCUSSION

Origin of ILV's in a linear chain

It was theoretically predicted that an ILV in an anharmonic lattice is identical to a LV near the force-constant defect in a harmonic lattice.[3] As shown in Figs. 1(b)–1(c), the Fourier spectrum of an ILV's is sufficiently narrow, indicating the quasiharmonic character of the vibration. One can also see distinct spatial localization and splitting of the ILV frequency ω_L from the phonon band depending on the amplitude of type-(ii) excitation [see Figs. 1(c) and 1(d)]. Excitations of type (i) and (ii) differ from type (iii) only in the quantity of the ILV final energy E^1 with respect to the initial one, E^0. For types (i) and (ii) the ratio E^1/E^0 is close to 0.5, whereas for type (iii) it is approximately 0.7. All other properties of the ILV are independent of the type of excitation. Distinct spatial localization and frequency shift are inherent in a harmonic LV near a force constant or mass defect. But, in our case, the defect strength depends on the amplitude of vibration.

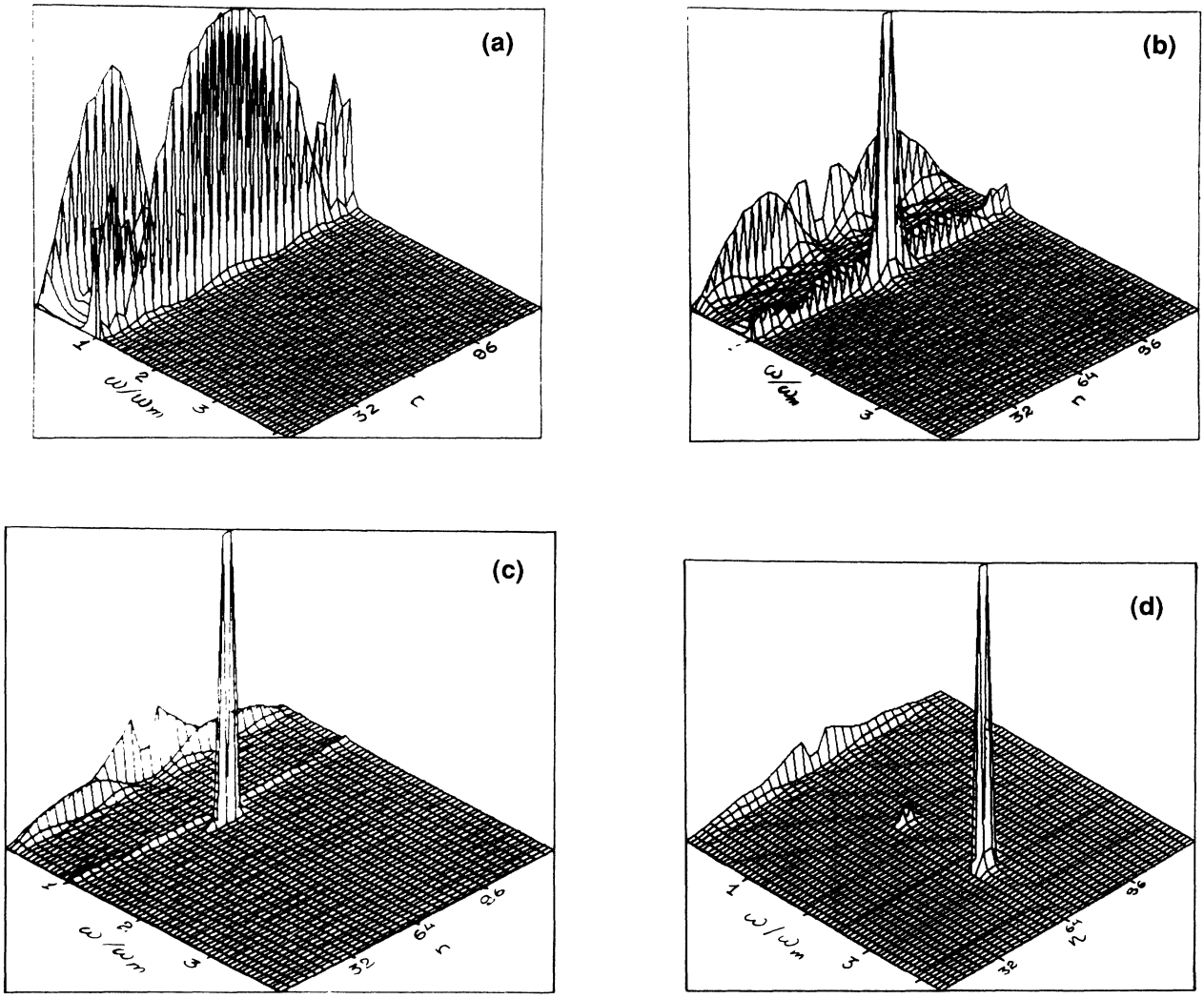

FIG. 1. Fourier-transform spectrum $A(\omega,n)$ of particle displacements in a one-dimensional anharmonic linear chain. The calculation of $A(\omega,n)$ was initiated at $T=256T_m$ and continued during the interval $T_\omega=64T_m$. The excitation of a set of low-frequency modes (including $\omega=0$) is observed. This is caused by the cubic anharmonicity term in the potential. The anharmonic potential parameters are $K_3=-60$, $K_4=800$. The initial displacements of particles of type-(ii) excitation are as follows: (a) $u^0=0.07$, no localized mode; (b) $u^0=0.1$, first observation of a localized mode; (c) $u^0=0.15$, a distinct shift of localized-mode frequency from cutoff frequency; (d) $u^0=0.4$—note the second localized mode in the cutoff region which first appears at $u^0=0.22$.

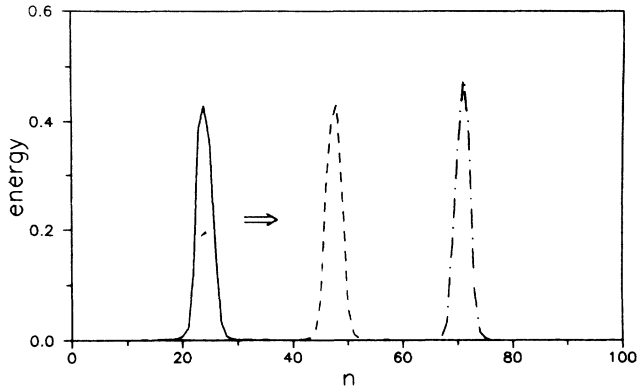

FIG. 2. Moving ILV excited in a linear chain close to the fixed end particle. The type-(ii) excitation with initial amplitudes $u_2=0.1$, $u_3=-0.1$, $u_1\equiv 0$ (fixed particle). Different peaks correspond to ILV at different times: $T=20T_m$ (solid curve), $T=40T_m$ (dashed curve), and $T=60T_m$ (dotted-dashed curve).

There is also another approach to the ILV problem from its solitonlike nature.[10] One may readily excite a moving ILV near the fixed end of a chain. Such an ILV looks like a soliton (Fig. 2). The dispersion law for the ILV (dependence of peak frequency ω_l on velocity) is presented in Fig. 3.

To investigate the origin of ILV's or the extent of the similarity of ILV's to solitons, computer experiments were carried out in three ways: (a) the Fourier analysis was performed on solitons excited using the analytical solution for the Toda lattice;[7] (b) the Fourier analysis was performed on ILV's in the linear chain containing 10 particles; (c) the Fourier analysis was performed on particle vibrations in a linear chain with the potential (1) and with excitation of a so-called envelope soliton (we call an envelope soliton an excitation of the top-band vibration with a solitonlike envelope function; this type of soliton is usually observed in nonlinear optics[11]).

(a) To perform this type of experiment, we have fitted the potential (1) to the potential of the Toda lattice by adjusting the constants. For this purpose the Toda potential was expanded into the Taylor series and the K_2, K_3, and K_4 constants in (1) were chosen to be equal to appropriate coefficients of expansion. For the excitation of a soliton in the lattice with the potential thus obtained, we use the relationship between the particle number n and its displacement u_n in the form[7]

$$\exp[(2K_3/K_2)(u_{n+1}-u_n)]-1=(\beta^2/K_2)\mathrm{sech}(\alpha n+\beta t)\ , \tag{4}$$

where $\beta=\sqrt{K_2}\sinh\alpha$. The soliton width is h/α and the quartic anharmonic force constant $K_4=\frac{2}{3}(K_3^2/K_2)$. After this, the equations of motion for 128 particles, with the particles at the end fixed, were solved for a time interval $T_\omega=512T_m$. During the time of motion there were many reflections of the soliton from the fixed end particles. All this time the shape and the energy of the soliton were observed to be conserved very well (within 1% of the initial energy). The result of the Fourier analysis of the particle displacements is shown in Fig. 4. Note that low-frequency modes are effectively involved in the soliton. Another important result is the relatively low anharmonicity of this excitation: the relative contributions to the particle energy of harmonic, third, and quartic anharmonic terms are equal to 1, 0.1, and 0.01, respectively.

(b) This experiment was performed to answer the question about the type of spectrum (one particle or many particle) forming the ILV. For the detailed investigation of this problem, we use a linear chain containing 10 parti-

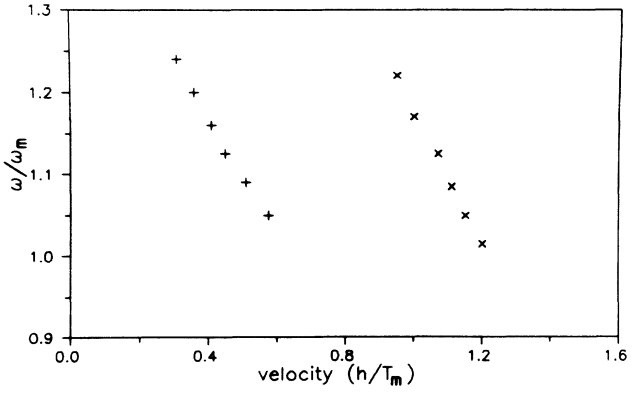

FIG. 3. Dispersion (frequency velocity) curves for excitation of a moving ILV. (×) corresponds to initial excitation $u_2=u^0$, $u_3=-u^0$ ($u_1\equiv 0$, fixed); (+) corresponds to excitation $u_3=u^0$, $u_4=-u^0$ ($u_1\equiv 0$, fixed).

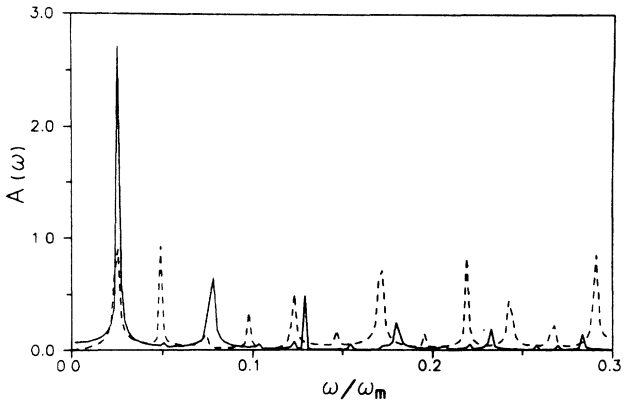

FIG. 4. Frequency spectrum $A(\omega)$ of particles in a linear chain with an excited soliton (solid curve). The potential parameters are $K_3=-218$, $K_4=800$. The characteristic displacements of particles in the soliton are of the order of 0.03. The ratio of corresponding harmonic and anharmonic parts of energy related to K_2, K_3, and K_4, terms is 1:0.1:0.01. Dashed curve: the frequency spectrum of the harmonic chain ($K_3=K_4=0$).

cles and exclude the term with K_3 from the potential (1) to remove the ILV damping. Figure 5(a) demonstrates that for a sufficiently high initial amplitude of type-(ii) excitation ($u^0=0.2$), the $A(\omega)$ spectrum of the particles involved contains phonons (delocalized vibrations) and an ILV. The ILV is represented by the shifted top-band vibration in this figure. The $A(\omega)$ spectrum of the uninvolved particles contain only undisturbed phonons [Fig. 5(b)]. Note that the relative contribution of the anharmonicity, determined, according to (3), by λ, to the frequency of vibration of involved particles is close to unity in this case. Figure 5(c) presents the situation when there is no ILV splitting from the phonon band ($u^0=0.1$).

(c) The results obtained in this experiment present a crossover regime from weakly anharmonic solitons to strongly anharmonic ILV's. Figure 6(a) presents the frequency spectrum of 30 particles involved (solid line) and uninvolved (dashed line) in the envelope soliton. It is clearly seen from Fig. 6(a) that soliton formation in this case requires anharmonic coupling of several modes: there is a wide peak in the frequency spectrum of the involved particles in place of the several narrow peaks corresponding to decoupled modes of uninvolved particles. The relative anharmonic contribution to the total energy in the case shown in Fig. 6(a) is about 3%. The higher anharmonicity (15%) of vibration [Fig. 6(b)] leads to splitting of a single mode from the vibrational band, indicating ILV formation. Note that a genuine single-mode splitting is shown in Fig. 6(b), in which the total number of modes is equal to 15, as in the harmonic case.

The observed results indicate the similarity of an ILV and a harmonic LV near the force-constant defect. This means that the frequency of an ILV may be regarded as a solution of the Lifshitz equation,[12]

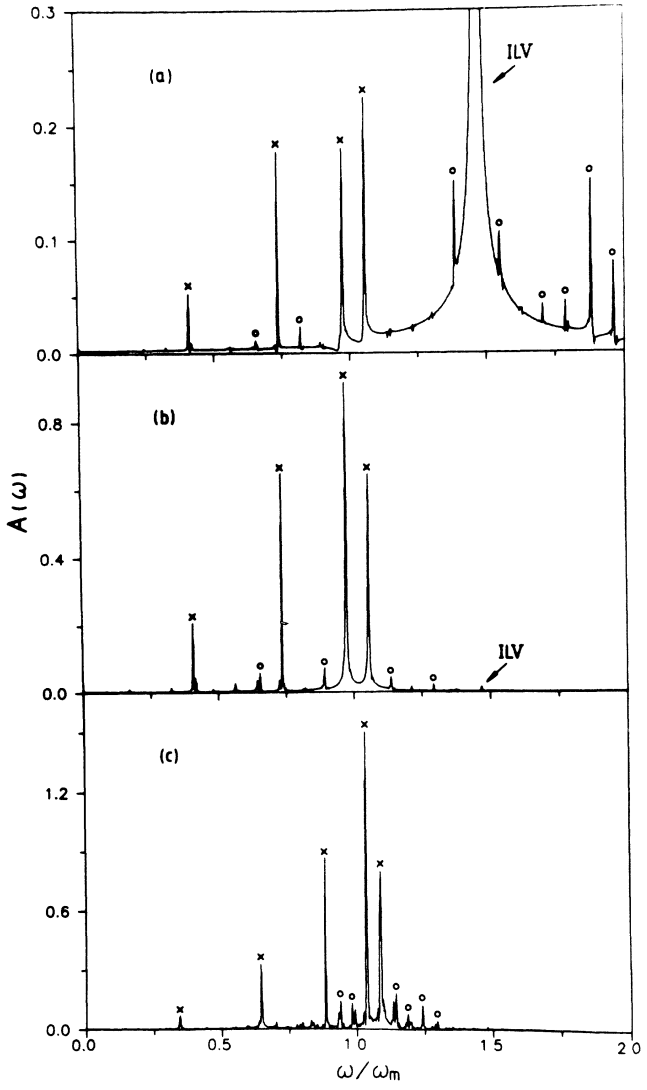

FIG. 5. Fourier transform $A(\omega)$ of particle displacements (a) involved and (b) not involved in the ILV of type-(ii) excitation with initial displacement $u^0=0.2$; (c) corresponds to initial displacement $u^0=0.1$, where the ILV is not excited. The potential parameters are $K_3=0$, $K_4=800$. Phonon peaks are marked by ×, overtones by ○.

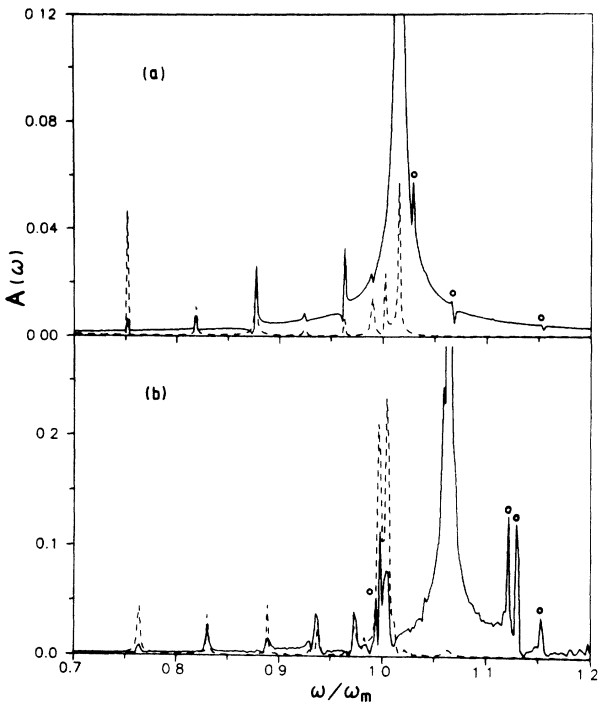

FIG. 6. Frequency spectrum of particles involved (solid curve) and not involved (dashed curve) in (a) the envelope soliton and (b) the ILV excited in a monatomic chain containing 30 particles. Potential parameters are $K_3=0$, $K_4=800$. The form of excitation was represented by the top-band mode of the amplitude (a) ±0.05 and (b) ±0.03 with modulation envelope function sech$[(n-N/2)/3]$. In order to reveal the vibrational-band spectrum, there was an additional random contribution of a relatively smaller quantity to the initial particle displacements. Overtones are marked by ○.

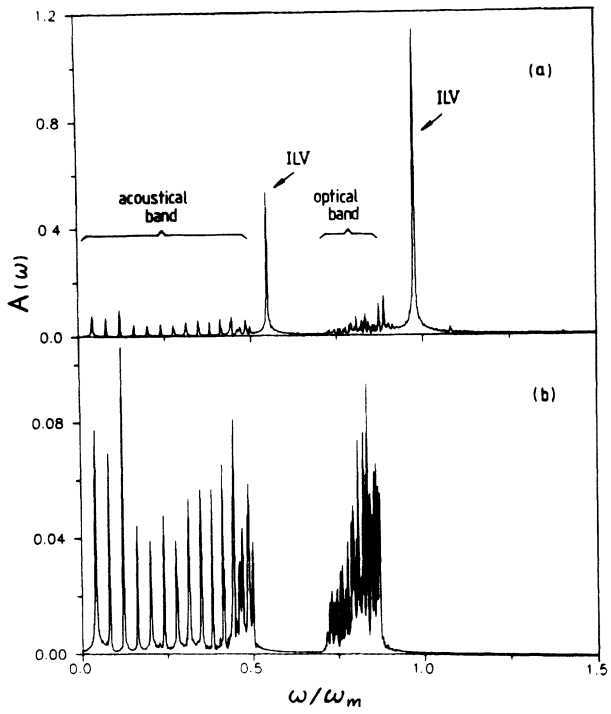

FIG. 7. Type-(ii) ILV excited with $u^0=0.1$ in a diatomic ($m_1=1, m_2=2$) chain containing 64 particles with potential coefficients $K_3=0$, $K_4=800$. Frequency spectrum $A(\omega)$ (a) of particles involved in the ILV; (b) of uninvolved particles.

$$1+\frac{\gamma}{N}\sum_k \frac{1}{\omega^2(k)-\omega^2}=0\ , \tag{5}$$

with the effective defect strength γ and the phonon spectrum $\omega(k)$.

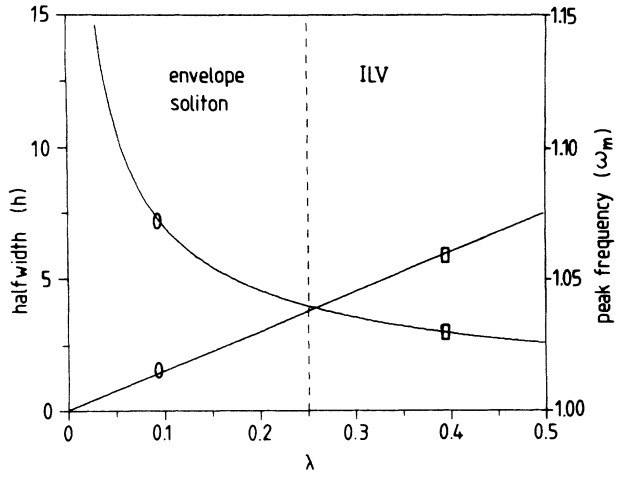

FIG. 8. λ dependence of half-width and peak frequency of localized vibrations in the 1D anharmonic lattice. Parts of the curves to the left of the vertical dashed line corresponds to solitons (multimode complexes) and those to the right to intrinsic localized vibrations (single-mode excitations). ○, envelope soliton; □, ILV with parameters the same as Fig. 6.

Equation (5) is written within the effective point-defect approximation. The role of a point defect is played by the mean-square amplitude of the ILV, which leads to phonon scattering and localization of the top-band vibrational mode. Further increase of the amplitude also leads to localization of the second top-band vibration [see Fig. 1(d)].

The analogy of an ILV to the defect LV is also confirmed by experiments with a diatomic linear chain. If one makes a type-(ii) excitation of an ILV in such a chain, then there appear two ILV's arising near the tops of optical and acoustical bands [see Fig. 7(a)] and they are formed by localization of respective top-band vibrations. Figure 7(b) presents the band states of the diatomic chain without an ILV. The optical ILV can be excited in a diatomic chain by setting the initial displacements of two adjacent particles $u_i=u^0$ and $u_{i+1}=-m_2u^0/m_1$. The main difference in the ILV excitation in a diatomic chain is a decrease in the localization threshold for an ILV excitation near an optical band, because the optical band is narrower. If this threshold, $E_{\rm th}$, is the minimum energy required to form an ILV, the localization threshold for a monatomic chain with $m=1$ is $E_{\rm th}=1.25$, while for a diatomic chain ($m_1=1$, $m_2=5$) with the same potential parameters, it is $E_{\rm th}=0.25$. This result is analogous to the localization threshold observed in the harmonic lattice, and is determined by the strength of the defect parameters and bandwidth of the delocalized excitation spectrum.[2] The results obtained above allow one to recognize the region of existence of different types of localized excitations in the space of the anharmonicity parameter λ. The last parameter was determined for a central particle involved in any type of local vibrations. Figure 8 presents the qualitative λ dependence of the half-width of local vibration in real space and on the peak frequency.

Thus, we may conclude that (a) ILV's are nearly identical to harmonic LV's near a force-constant defect, and (b) The soliton and ILV are different types of localized vibrations. The first corresponds to anharmonic coupling of several vibrational modes and requires relatively low anharmonicity. The second has an essentially high anharmonic contribution and consists of a single top-band vibrational mode split from the vibrational band.

Temperature dependence of the localization threshold

We define the localization-threshold energy as the minimum energy required for the ILV excitation. To describe the temperature dependence of the localization conditions, one may try to use (5).

At first, we will deduce the expression for γ in (5). Let us suppose that an ILV is fairly confined on only one particle with $n=0$ [type-(i) excitation]. One may introduce the effective phonon-phonon scattering using the assumption that the inhomogeneity of the chain is caused by the high vibrational amplitude of this particle:

$$H_{\rm int}=(K_4/4)(a^2-\langle u_n^2\rangle)[(u_1-u_0)^2+(u_{-1}-u_0)^2]\ , \tag{6a}$$

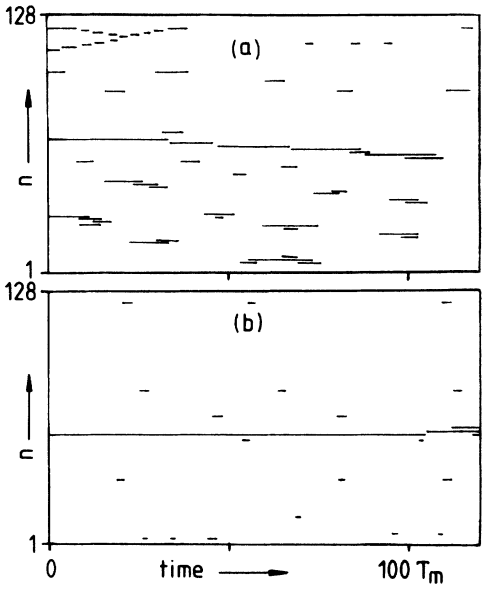

FIG. 9. Time dependence of ILV's for the monatomic linear chain of 128 particles at elevated temperature. The type-(ii) ILV was excited on 64 and 65 particles by setting initial displacements (a) $u^0=0.12$ and (b) $u^0=0.13$. The initial displacement of the other particles was generated by a random number generator with mean-square amplitude $\langle u_n^2\rangle=0.003$. The black lines on the map correspond to a cell with total energy exceeding the 0.5 level of the maximum value.

$$u_n=(1/\sqrt{N})\sum_k [Q_k\exp(ikn)+Q_k^*\exp(-ikn)] , \qquad (6b)$$

where Q_k is the phonon with wave number k (Q_k's are orthogonal, but not normalized to unity because of the nonzero temperature), and $\langle u_n^2\rangle$ is the mean-square amplitude of the host particle. With the assumed units, $\langle u_2^n\rangle$ characterizes the temperature of the chain according to the relation $k_BT=K_2\langle u_n^2\rangle$ (k_B is Boltzmann's constant).

Substituting (6b) into (6a), one obtains

$$H_{\rm int}=4K_4(a^2-\langle u_n^2\rangle)\sum_{k,k}{}'Q_kQ_k^*\sin k\,\sin k' ,$$

$$\pi/2\geq k,k'\geq 0 , \qquad (6c)$$

or, in a simplified form,

$$H_{\rm int}=4K_4\overline{\sin k\,\sin k'}(a^2-\langle u_n^2\rangle)\sum_{k,k'}Q_kQ_{k'}^*=\gamma\sum_{k,k'}Q_kQ_{k'}^* . \qquad (6d)$$

Thus, from (6) it is clear that to maintain the defect strength $a^2-\langle u_n^2\rangle=a^2-k_BT/K_2$ as temperature independent, one must take the temperature dependence of the initial excitation amplitude in the form

$$[u^0(T)]^2=[u^0(0)]^2+k_BT/K_2 . \qquad (7)$$

For excitation of type-(i) vibrations, the initial energy E^0 is related to u^0 by

$$E^0=K_2(u^0)^2+(K_4/2)(u^0)^4 . \qquad (8)$$

Thus, from (7) and (8) one can evaluate the temperature dependence of the localization threshold,

$$E_{\rm th}(T)=E_{\rm th}(0)+[1+(2K_4/K_2^2)E_{\rm th}(0)]^{1/2}k_BT+(K_4/2K_2^2)(k_BT)^2 . \qquad (9)$$

The temperature dependence of $E_{\rm th}$ was determined experimentally using a random number generator for all particles, except that used for ILV excitation. The point

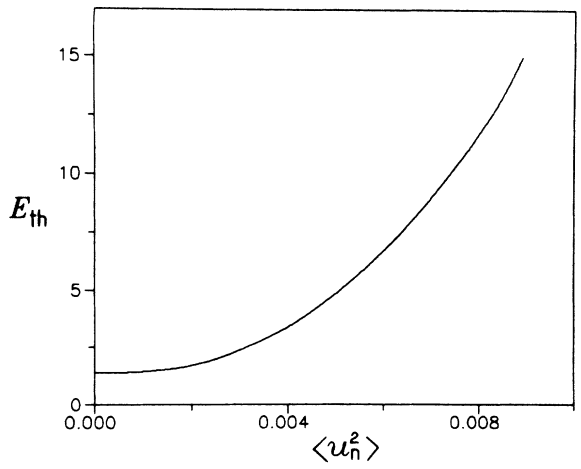

FIG. 10. Temperature dependence of the localization threshold.

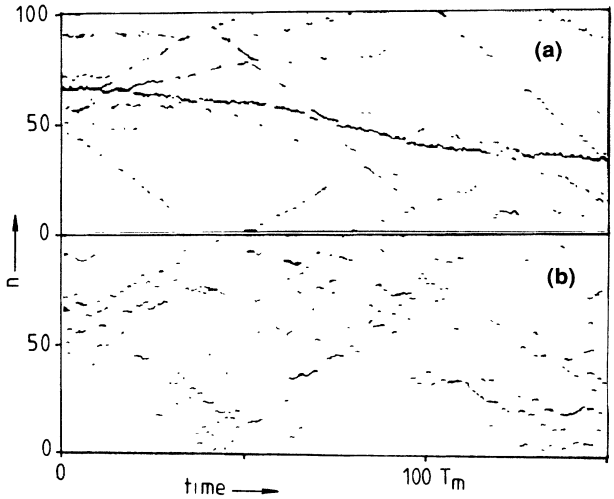

FIG. 11. Thermal ILV excitation by means of the generation of random initial displacements of all particles ($\langle u_n^2\rangle=0.09$) in the chain. (a) With the same potential parameters as described in Fig. 1. (b) The potential without the quartic term ($K_4=0$).

was marked on the map in Fig. 9, if the sum of the potential and kinetic energies of a particle exceeds one-half of the maximum value per atom during the calculation. Two examples of the ILV excitation in a linear chain with the temperature T corresponding to $\langle u_n^2 \rangle = 0.003$ for the excitation energy (a) under and (b) over the threshold energy are shown in Fig. 9. The tracks of the high-energy regions corresponding to an ILV with the initial amplitude $[u^0(T)]^2 = 1.4[u^0(0)]^2$ are observed in Fig. 9(b). Figure 9(a) shows the destruction of the ILV when the initial amplitude is $[u^0(T)]^2 = 1.3[u^0(0)]^2$. The ILV destruction criterion was roughly determined as the disappearance of the track during the time $T = 100T_m$. This quantity is typical for the ratio of the lifetime to the period of vibration of real phonons. The T dependence of the E_{th} thus determined is presented in Fig. 10. One can see a stronger temperature dependence of the experimental E_{th} compared to that given by (9).

It is interesting in connection with the E_{th} temperature dependence to consider the possibility of random excitations of ILV's. Figure 11 shows thermally activated ILV's. The procedure is to set up a random initial displacements for all the particles in the chain in the way described above. Figure 11(a) shows bright tracks corresponding to ILV's. When the quartic term in the potential is eliminated, the ILV disappear [see Fig. 11(b)].

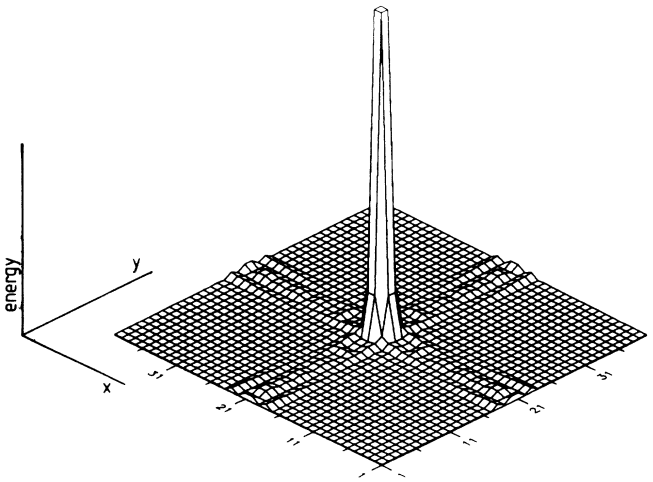

FIG. 12. ILV in a square anharmonic lattice ($h = 0.99$, $r_0 = 1$) with the potential parameters $K_2 = (2\pi)^2$, $K_3 = 0$, and $K_4 = 1400$. The square lattice has 40 particles per side. The initial displacements ($|u_x^0| = |u_y^0| = 0.1$) of the four neighboring particles in the middle of the lattice are directed to the center of the square unit cell. Time after excitations is $40T_m$. The relative anharmonic contribution λ, determined according to (3), is close to unity.

ILV's in a square lattice

It was shown theoretically that solitonlike excitations in 2D discrete space are unstable.[11] With regard to the ILV's in a 2D lattice, a condition very similar to excitations of ILV's in a linear chain exists. The ILV in a square lattice with the potential (1) is presented in Fig. 12. As in a 1D lattice, there is a very strong dependence of the ILV stability on the vibration amplitude in a 2D lattice.

As to the mobility of an ILV in a square lattice, we have failed in exciting a moving ILV by a manner similar to the 1D case. All variants of excitation lead to ILV destruction.

It is obvious that in two dimensions there may be a great variety of ILV types (longitudinal and transverse of different shape). A detailed investigation of such excitations in different 2D lattices (square, triangular, etc.) will be considered in the future.

ACKNOWLEDGMENTS

We thank Professor A. J. Sievers for many discussions and helpful remarks and S. R. Bickham for a critical reading of the manuscript.

[1] A. A. Maradudin, E. W. Montroll, and G. H. Weiss, *Theory of Lattice Dynamics in the Harmonic Approximation*, Suppl. XX of *Solid State Physics* (Academic, New York, 1971).

[2] A. S. Barker, Jr. and A. J. Sievers, Rev. Mod. Phys. **47**, 2 (1975).

[3] A. J. Sievers and S. Takeno, Phys. Rev. Lett. **61**, 970 (1988).

[4] S. Takeno and A. J. Sievers, Solid State Commun. **67**, 1023 (1988).

[5] A. G. Badalyan, P. G. Baranov, V. S. Vihnin, M. M. Petrosyan, and V. A. Hramstov, Zh. Eksp. Teor. Fiz. **61**, 808 (1985).

[6] V. M. Burlakov, S. A. Kiselev, and V. N. Pyrokov, Solid State Commun. **74**, 327 (1990).

[7] M. Toda, *Theory of Nonlinear Lattices*, Vol. 20 of *Springer Series in Solid-State Sciences* (Springer, Berlin, 1989).

[8] R. W. Hockney and J. W. Eastwood, *Computer Simulation Using Particles* (McGraw-Hill, New York, 1981).

[9] E. Fermi, J. Pasta, and S. Ulam, *Collected Papers of Enrico Fermi*, edited by E. Segrè (University of Chicago Press, Chicago, 1965), Vol. 2, p. 978.

[10] S. Takeno, K. Kisoda, and A. J. Sievers, Prog. Theor. Phys. **94**, 242 (1988).

[11] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).

[12] I. M. Lifshitz and A. M. Kosevich, Rep. Prog. Phys. **29**, 1 (1966); **29**, 217 (1966).