Numerical study of phonon localization in disordered systems

Michael L. Williams and Humphrey J. Maris
Department of Physics, Brown University, Providence, Rhode Island 02912
(Received 13 July 1984)

A new numerical method has been developed to find the frequencies and vibration patterns of normal modes of harmonic systems. We have used this method to calculate the phonon density of states, and the phonon localization length for a two-dimensional square lattice with mass disorder.

I. INTRODUCTION

A large amount of work has been devoted to the calculation of the phonon density of states in disordered systems.[1] However, very little is known about the actual nature of the phonon states. By analogy with the corresponding problem of electrons in disordered systems, one expects that for strong disorder, phonon states will be localized excitations in the Anderson sense.[2] Phonon localization in one-dimensional solids has been considered by Ishii[3] and Jäckle.[4] Numerical studies have been made by Nagel, Rahman, and Grest[5] for Lennard-Jones glasses. The glasses were prepared by means of molecular-dynamics simulations of rapid cooling from the liquid state. Once a glass was formed, the normal modes of vibration were studied numerically. It was found that above a certain frequency the modes were localized, as expected from the general ideas of Anderson localization.

We have developed a new numerical method to calculate phonon eigenstates for disordered systems. The purpose of this paper is to describe this method, and as an example to use the method to study phonon localization in two-dimensional lattices with mass disorder.

II. NUMERICAL METHOD

Let us consider a set of N atoms which are coupled together by linear springs. For simplicity, we consider the particles to move only in a single direction (e.g., the z direction), but our method can easily be extended to cover general motions. The equation of motion of the masses is

$$M_l\ddot{u}_l(t)=-\sum_{l'}\phi_{ll'}u_{l'}(t)\ . \tag{1}$$

M_l and $u_l(t)$ are the mass and displacement of the lth mass and $\phi_{ll'}$ describes the strength of the spring coupling atoms l and l'. The displacement can be decomposed into a set of normal modes according to

$$u_l(t)=\sum_{\lambda}Q_\lambda(t)\frac{e_l(\lambda)}{\sqrt{M_l}}\ , \tag{2}$$

where Q_λ is the amplitude of the normal mode λ and $e_l(\lambda)$ is the displacement pattern or "polarization vector" of the mode λ. The $\{e_l(\lambda)\}$ and the frequencies $\{\omega_\lambda\}$ satisfy the equations

$$\sum_{l'}\phi_{ll'}(M_lM_{l'})^{-1/2}e_{l'}(\lambda)=\omega_\lambda^2 e_l(\lambda)\ . \tag{3}$$

Thus, to find the frequencies and displacement patterns of the normal modes one has to find the eigenvalues and eigenvectors of an $N\times N$ matrix. Conventional methods require a large amount of computer time as N becomes large. Thus, one has to find another approach.

A. Method for the density of states

One can find the density of states by the following method. We start with each atom at rest and with zero displacement. We then apply a force on each atom given by

$$F_l\cos(\Omega t)\ , \tag{4}$$

where F_l is independent of time. After a time, large compared to the typical period of oscillation of the atoms, the total energy of the system is

$$E=\tfrac{1}{2}\sum_{\lambda}\left[\sum_{l}\frac{F_le_l(\lambda)}{\sqrt{M_l}}\right]^2\frac{\sin^2\{[(\omega_\lambda-\Omega)/2]t\}}{(\omega_\lambda-\Omega)^2}\ . \tag{5}$$

Thus, for large times the periodic force excites only those modes whose frequency is close to Ω. Now let us choose

$$F_l=F_0\sqrt{M_l}\cos(\phi_l)\ , \tag{6}$$

where F_0 is a constant and ϕ_l is a random quantity. If we average over all possible values of ϕ_l we find that the average value of E is

$$\langle E\rangle=\frac{F_0^2}{4}\sum_{\lambda}\frac{\sin^2\{[(\omega_\lambda-\Omega)/2]t\}}{(\omega_\lambda-\Omega)^2}\ . \tag{7}$$

We have used the orthonormality of the mode patterns:

$$\sum_{l}e_l(\lambda)e_l(\lambda')=\delta_{\lambda\lambda'}\ . \tag{8}$$

The modes which contribute to the sum in (7) are those whose frequencies lie within about $\pm(2\pi/t)$ of Ω. Let us choose t such that

$$\Omega t\gg 1\ , \tag{9}$$

$$4\pi N/\omega_m t\gg 1\ , \tag{10}$$

where ω_m is the maximum frequency of the system. Equation (9) means that only modes in a narrow band of frequency on the scale of Ω contribute to the sum. Equation (10) ensures that the number of such modes is much larger than unity.[6] Thus, if these conditions are met we have

$$\langle E\rangle \approx \frac{\pi t F_0^2}{8}\sum_\lambda \delta(\omega_\lambda-\Omega)=\frac{\pi t F_0^2 N g(\Omega)}{8}\ , \tag{11}$$

where $g(\Omega)$ is the phonon density of states. Thus, provided that a way can be found to carry out the time development in the presence of the periodic force, we can find $g(\Omega)$ from

$$g(\Omega)=\frac{8\langle E\rangle}{\pi t F_0^2 N}\ . \tag{12}$$

For a sufficiently large system, one expects that it will not be necessary to carry out explicitly the average over all values of the $\{\phi_l\}$, i.e., it will be sufficient to make one random choice of the $\{\phi_l\}$.

B. Mode patterns

If the system is driven for longer times there will eventually be only a very small number of modes whose excitation level is important. When t is very large, i.e., such that

$$4\pi N/\Omega t<1\ , \tag{13}$$

the resonance factor in Eq. (7) is so sharp that the width $\Delta\omega_R$ of the resonance is less than the spacing $\Delta\omega_\lambda$ between the frequencies of adjacent frequency normal modes. Then most of the energy of the system will be in just a few of the normal modes. If the modes of frequency around Ω are localized, one should then find that the energy distribution in the system will be very inhomogeneous. There will be regions of essentially negligible energy density, and regions of high energy density where the highly excited modes are localized. Since the number of excited modes is small, they will have a very small probability of overlapping. Hence, any region of high density can be assumed to be caused by a single mode.[7] Consequently, the displacements u_l of the atoms in such a region must be

$$u_l=Ce_l(\lambda_0)/\sqrt{M_l}\ , \tag{14}$$

where C is a coefficient independent of l and $e_l(\lambda_0)$ is the polarization vector of the particular mode λ_0. Thus, by looking at these displacements, $u_l, e_l(\lambda_0)$ can be found.

In the case that the modes are extended this method does not work. No matter how large t is one always has significant excitation of a few modes, and when these modes are extended their vibration patterns overlap.

We have devised the following scheme to isolate single modes when the modes are only weakly localized, or even extended. We first drive the system for a time $\tilde{t}$, at which point the displacement of atom l is $u_l^{(1)}$. We can write this displacement as the sum of couplings to the normal modes:

$$u_l^{(1)}=\frac{1}{\sqrt{M_l}}\sum_\lambda F_\lambda h(\Omega,\omega_\lambda,\tilde{t})e_l(\lambda)\ , \tag{15}$$

where

$$F_\lambda=\sum_l \frac{F_l e_l(\lambda)}{\sqrt{M_l}}\ , \tag{16}$$

$$h(\Omega,\omega_\lambda,\tilde{t})=\frac{2\sin[\frac{1}{2}(\Omega-\omega_\lambda)\tilde{t}]\sin[\frac{1}{2}(\Omega+\omega_\lambda)\tilde{t}]}{\Omega^2-\omega_\lambda^2}\ . \tag{17}$$

We then change the amplitude F_l of the force applied to atom l to a new value,

$$F_l^{(1)}=u_l^{(1)}M_l\ , \tag{18}$$

and, again starting with all particles at rest and with zero displacement, we drive the system for a time $\tilde{t}$. The resulting displacements we call $\{u_l^{(2)}\}$. We then reset the forces using

$$F_l^{(2)}=u_l^{(2)}M_l \tag{19}$$

and continue the iteration sequence. After p iterations the displacement of atom l is

$$u_l^{(p)}=\frac{1}{\sqrt{M_l}}\sum_\lambda F_\lambda h^p(\Omega,\omega_\lambda,\tilde{t})e_l(\lambda)\ . \tag{20}$$

After a sufficient numer of iterations the displacement pattern becomes dominated by the mode λ_1 for which $|h(\Omega,\omega_\lambda,\tilde{t})|$ $(\equiv|h_1|)$ is the largest, regardless of the values of $\{F_\lambda\}$. Thus, for sufficiently large p,

$$u_l^{(p)}\approx Ce_l(\lambda_1)/\sqrt{M_l}\ , \tag{21}$$

where C is independent of l. Hence, this provides a way to determine the pattern for the mode λ_1. Consider now the accuracy of this procedure. Let the mode which has the next-largest value of $|h|$ be λ_2, and let the value of h be h_2 for this mode. The accuracy of the result for the mode pattern depends critically on the ratio of $|h_1|$ to $|h_2|$. From Eq. (20) we see that the error in the mode pattern is proportional to

$$(|h_2|/|h_1|)^p\ . \tag{22}$$

Now, if $\tilde{t}$ is short (i.e., only a few times the period of a typical mode), $h(\Omega,\omega_\lambda,\tilde{t})$ will be a slowly varying function of the mode frequency ω_λ. Thus, $|h_2|/|h_1|$ will be only slightly less than 1 and a large value of p will be needed to get good accuracy. It is more efficient to make $\tilde{t}$ sufficiently large so that only a few modes have large values of $|h|$. Thus, one needs $\tilde{t}$ of the order of [see Eq. (13)]

$$\frac{4\pi N}{\Omega}\ . \tag{23}$$

Then $|h_2|$ will be significantly less than $|h_1|$, (e.g., $\frac{1}{2}|h_1|$) and the procedure converges rapidly.

C. Numerical algorithm

The problem thus reduces to the solution of the equations of motion of the system in the presence of a periodic force. These equations can be written as

$$\dot{v}_l(t)=M_l^{-1}\left[-\sum_{l'}\phi_{ll'}u_{l'}(t)+F_l\cos(\Omega t)\right]\ , \tag{24}$$

$$\dot{u}_l(t)=v_l(t)\ . \tag{25}$$

The standard approach to the time development is to replace t by $n\tau$ where τ is a small time step, and n an integer. Then a time-development algorithm is

$$v_l(n+1)=v_l(n)+M_l^{-1}\left[\sum_{l'}\phi_{ll'}u_{l'}(n)+F_l\cos(\Omega n\tau)\right]\tau\ , \tag{26}$$

$$u_l(n+1)=u_l(n)+v_l(n)\tau\ . \tag{27}$$

Normally one would choose τ to be small compared to the characteristic time scale of the system, i.e., the period τ_m of the highest-frequency mode. If we set

$$\tau=z\tau_m\ , \tag{28}$$

with $z\ll 1$, we see that the number n of time steps needed to study localized mode patterns [Eq. (13)], has to satisfy

$$n\gg\frac{4\pi N}{\Omega z\tau_m}\ . \tag{29}$$

If we take $\Omega\tau_m$ roughly equal to 2π then we need n much larger than $2N/z$. In a typical case one might use $z=0.01$.

Since the computing time is proportional to n, we would like to find a way to make z as large as possible. We have discovered the following interesting method. Consider a single harmonic oscillator of frequency ω, and ignore for the moment the driving force. Then

$$\dot{v}=-\omega^2 u\ , \tag{30}$$

$$\dot{u}=v\ , \tag{31}$$

and so we could try as an algorithm

$$v(n+1)=v(n)-\omega^2\tau u(n)\ , \tag{32}$$

$$u(n+1)=u(n)+\tau v(n)\ . \tag{33}$$

Unless $\tau\ll\omega^{-1}$ this algorithm does not work well. Let us start with $u(0)=u_0$ and $v(0)=0$. Even though the oscillator is not driven, we find that after one cycle the amplitude has increased by a factor r, which for small $\omega\tau$ is

$$r\approx 1+\pi\omega\tau\ . \tag{34}$$

One can understand this as follows. Regard the algorithm as a sequence of transforms acting on a vector $\mathbf{X}_n$ in (u,v) space, i.e.,

$$\mathbf{X}_n\equiv[u(n),v(n)]\ . \tag{35}$$

For the algorithm to give solutions which are periodic, we may guess that it is desirable for the Jacobian J of the transform to be 1. For the transform of Eqs. (32) and (33) we have

$$J=1+\omega^2\tau^2\ . \tag{36}$$

To get $J=1$ we make a simple change:

$$v(n+1)=v(n)-\omega^2\tau u(n)\ , \tag{37}$$

$$u(n+1)=u(n)+\tau v(n+1)\ . \tag{38}$$

For initial values $v(0)$ and $u(0)$ we find that

$$u(n)=u(0)\cos(n\alpha)+\left[\frac{\tau}{\sin\alpha}v(0)-\tan\left[\frac{\alpha}{2}\right]u(0)\right]\sin(n\alpha)\ , \tag{39}$$

$$v(n)=v(0)\cos(n\alpha)+\left[v(0)-\frac{2u(0)}{\tau}\right]\tan\left[\frac{\alpha}{2}\right]\sin(n\alpha)\ , \tag{40}$$

where

$$\alpha=2\sin^{-1}\left[\frac{\omega\tau}{2}\right]\ . \tag{41}$$

Thus, the solution is perfectly periodic in the sense that the magnitudes of the oscillations of v or u do not change as the iteration proceeds. The oscillator acts as though it has a frequency,

$$\tilde{\omega}=\frac{\alpha}{\tau}=\frac{2}{\tau}\sin^{-1}\left[\frac{\omega\tau}{2}\right]\ . \tag{42}$$

The relation between the apparent frequency $\tilde{\omega}$ and the real frequency ω is shown in Fig. 1. Equations (39)—(42) remain true provided that $\omega\tau<2$. Above this value, α becomes complex and the solution is no longer periodic. Thus, the simulation can be used provided

$$\tau\le\frac{\tau_0}{\pi} \tag{43}$$

where τ_0 is the period of the oscillator. Thus, we must have $z\le\pi^{-1}$.

Although the solutions (39) and (40) are periodic, the energy calculated as

$$\epsilon=\tfrac{1}{2}mv^2+\tfrac{1}{2}m\omega^2u^2\ , \tag{44}$$

is not constant, but oscillates at frequency $2\tilde{\omega}$ about a mean value $\bar{\epsilon}$. For small z, the amplitude of the oscillations $\ll\bar{\epsilon}$.

Consider now an oscillator driven by a force $F_0\sqrt{m}\cos(\Omega t)$. If we use the same algorithm to perform the time development, we obtain some complicated expressions for $u(n)$ and $v(n)$. Near to resonance the result for $\bar{\epsilon}$ for large n is approximately

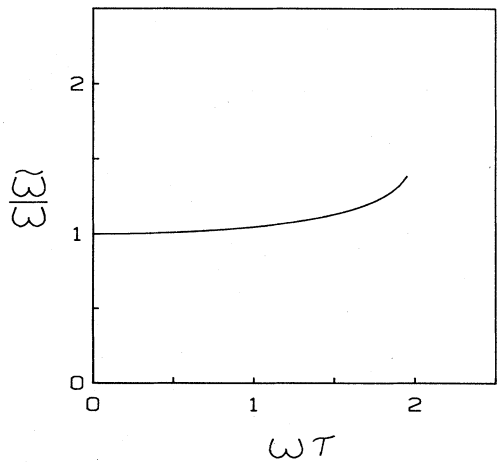

FIG. 1. Shift in frequency of an oscillator produced by the algorithm. $\tilde{\omega}$ is the apparent frequency, ω is the real frequency, and τ is the time step.

$$\bar{\epsilon}=\frac{F_0^2\tau^2}{2\cos^2(\alpha/2)}\frac{\sin^2[(n/2)(\alpha-\alpha_0)]}{(\alpha-\alpha_0)^2}\ , \tag{45}$$

where $\alpha_0=\Omega\tau$.

We can apply this algorithm to the system of N atoms driven by the force (6). This system can be thought of as N oscillators, and so the algorithm effectively time-develops each of these oscillators with a shifted frequency given by (42). The algorithm does *not* change the displacement patterns associated with the modes. Thus, we can determine these patterns by the method already described, i.e., we drive the system for a long time so that only a few modes are excited. The real frequencies ω_λ of the resonant modes are now related to the driving frequency Ω by

$$\omega_\lambda=\frac{2}{\tau}\sin\left[\frac{\Omega\tau}{2}\right]\ . \tag{46}$$

To find the density of states we drive the system for a shorter time, so that many modes are still appreciably excited. Then, we have the result, analogous to (12), that

$$g\left[\frac{2}{\tau}\sin\left[\frac{\Omega\tau}{2}\right]\right]=\frac{8\langle E\rangle\cos(\Omega\tau/2)}{\pi n\tau F_0^2 N}\ . \tag{47}$$

The step size τ in these calculations has to be sufficiently small so that (43) is satisfied for all of the modes of the system. Thus, we need

$$\tau\le 2/\omega_m\ , \tag{48}$$

where ω_m is the maximum frequency. In fact, this condition provides a convenient method to determine ω_m. We set the applied force equal to zero, and give the atoms some random initial displacements and positions. Then the algorithm is used to time develop the system, and after each step the total energy E is calculated. If τ satisfies (48), E will undergo small fluctuations about a mean value. We then increase τ and repeat the procedure. As soon as τ exceeds the limit (48), we see that the energy begins to increase as the iterations proceed. This is because α for the highest frequency mode of the system has become complex, and the energy of this mode grows exponentially. A measurement of the critical value of τ can thus be used to give ω_m.

III. RESULTS AND DISCUSSIONS

We have applied our numerical method to study localization in a simple two-dimensional model. The atoms form a simple-cubic lattice in the x-y plane, and are allowed to move only in the z direction. The atoms interact only with nearest neighbors via springs of strength k. If we label the atoms by coordinates l_x,l_y, the equation of motion is

$$\dot{v}_{l_x l_y}=M^{-1}_{l_x l_y}\sum_{l'_x l'_y}[k(u_{l'_{x+1},l'_y}+u_{l'_x,l'_{y+1}}+u_{l'_{x-1},l'_y}+u_{l'_x,l'_{y-1}}-4u_{l_x,l_y})+F_{l_x l_y}\cos(\Omega t)]\ . \tag{49}$$

We take all the spring constants to be the same, and introduce a disorder through the masses.

A. Density of states

We have calculated $g(\omega)$ for lattices of 1600 atoms in a 40×40 array. The spring constant k was 1, and the masses were randomly distributed in the range

$$1-q<M<1+q\ . \tag{50}$$

Periodic boundary conditions were used. $g(\omega)$ was calculated at ω intervals of 0.1. Results are shown in Fig. 2. We have several checks we can apply to these results. For the ordered lattice (all masses $=1$) we can compare with the exact solution[8] which can be written as

$$g(\omega)=\frac{\omega}{\pi^2}K\left[\frac{\omega^2(8-\omega^2)}{16}\right]\ , \tag{51}$$

where K is the complete elliptic integral of the first kind. This is shown by the dashed line in Fig. 2. The agreement with the numerical results is excellent. As a check of the disordered lattices we can use the moments of the frequency spectrum.[9] It is straightforward to show that

$$g_0\equiv\int g(\omega)d\omega=1\ , \tag{52}$$

$$g_2\equiv\int g(\omega)\omega^2 d\omega=\frac{2k}{q}\ln\left[\frac{1+q}{1-q}\right]\ , \tag{53}$$

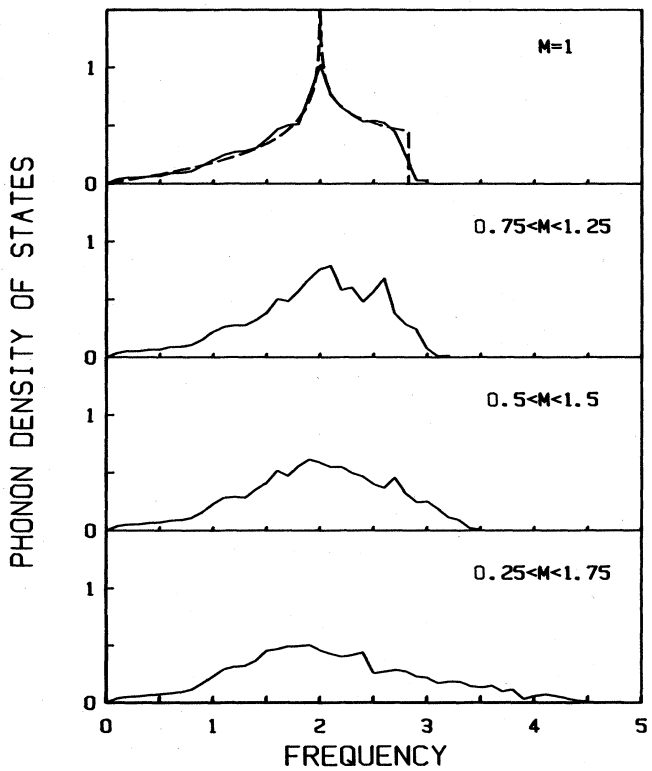

FIG. 2. Phonon density of states for two-dimensional square lattices. The range of masses is as indicated. The dashed line shows the exact result for the case of no mass disorder.

TABLE I. Comparison between the moments of the frequency spectra calculated numerically (N columns) and from the exact expressions Eqs. (52), (53), and (54) (E columns). q measures the amount of disorder as described in the text.

	g_0		g_2		g_4	
q	N	E	N	E	N	E
0	1.005	1	3.962	4	19.65	20
0.25	1.016	1	4.133	4.087	21.52	21.24
0.5	0.988	1	4.232	4.394	24.69	26.16
0.75	0.988	1	5.051	5.189	41.48	43.30

$$g_4 \equiv \int g(\omega)\omega^4 d\omega = k^2 \left[\frac{1}{q^2} \ln^2 \left[\frac{1+q}{1-q} \right] + \frac{16}{1-q^2} \right] . \tag{54}$$

In Table I we compare these exact results with the moments of the computed spectra. The agreement is very good.

B. Localization length

From the calculated displacement pattern $e_{l_x l_y}(\lambda)$ we can calculate how the energy of the mode λ is spatially distributed. Let the amplitude Q_λ of the mode be 1. Then, we define the energy of atom $l_x l_y$ as

$$E_{l_x l_y} \equiv \tfrac{1}{4} e^2_{l_x l_y}(\lambda)\omega^2_\lambda + \tfrac{1}{8}k \times \sum_{\substack{l'_y \\ l'_x}} [e_{l'_x l'_y}(\lambda)/(M_{l'_x l'_y})^{1/2} - e_{l_x l_y}(\lambda)/(M_{l_x} M_{l_y})^{1/2}]^2 , \tag{55}$$

where the sum is over those atoms $l'_x l'_y$ which are nearest neighbors of $l_x l_y$. Thus, $E_{l_x l_y}(\lambda)$ is the sum of the average kinetic energy of atom $l_x l_y$ plus half of the average potential energy of the bonds between this atom and its nearest neighbors. We can then define the center of the mode λ as

$$\mathbf{r}_0(\lambda) \equiv \sum_{l_x l_y} \mathbf{r}_{l_x l_y} E_{l_x l_y}(\lambda) / \sum_{l_x l_y} E_{l_x l_y}(\lambda) , \tag{56}$$

where the position vector of atom $l_x l_y$ is

$$\mathbf{r}_{l_x l_y} = l_x \hat{i} + l_y \hat{j} . \tag{57}$$

As a measure of this size of the mode, we have used the quantity

$$\zeta(\lambda) \equiv \sum_{l_x l_y} | \mathbf{r}_{l_x l_y} - \mathbf{r}_0(\lambda) | E_{l_x l_y}(\lambda) / \sum_{l_x l_y} E_{l_x l_y}(\lambda) . \tag{58}$$

Results for the localization length in units of the interatomic spacing are shown in Fig. 3. These calculations are for $q = 0.25$, 0.5, and 0.75. In each case the upper end of the frequency range is simply determined by the maximum frequency of the system. The lowest frequency is the point below which the mode becomes so large that the effect of the finite size of the system becomes significant. The error bars indicate the standard deviation in ζ, determined typically from 4 to 6 modes of essentially the same frequency. Thus, the uncertainty in the average value of ζ at each frequency is smaller than the error bars by a factor of $\sqrt{4}$ to $\sqrt{6}$.

In two dimensions it is believed[10,11] that all states are localized, and so there is no true mobility edge separating localized and extended states. However, it is expected that there will be ranges of energy (for electrons) or frequency (for phonons) where ζ will be very large, and hence the states are only weakly localized. Our results show that the range of frequency for which ζ is short (strongly localized) extends rapidly as the disorder is increased. The structure of the states with very short ζ (i.e., ~ 0.5) is very simple (Fig. 4). One atom (atom 0) has an amplitude u_0 which is considerably larger than that of any other atom. The four neighboring atoms have displacements in the opposite direction. Thus, the structure of the mode is qualitatively similar to the vibration pat-

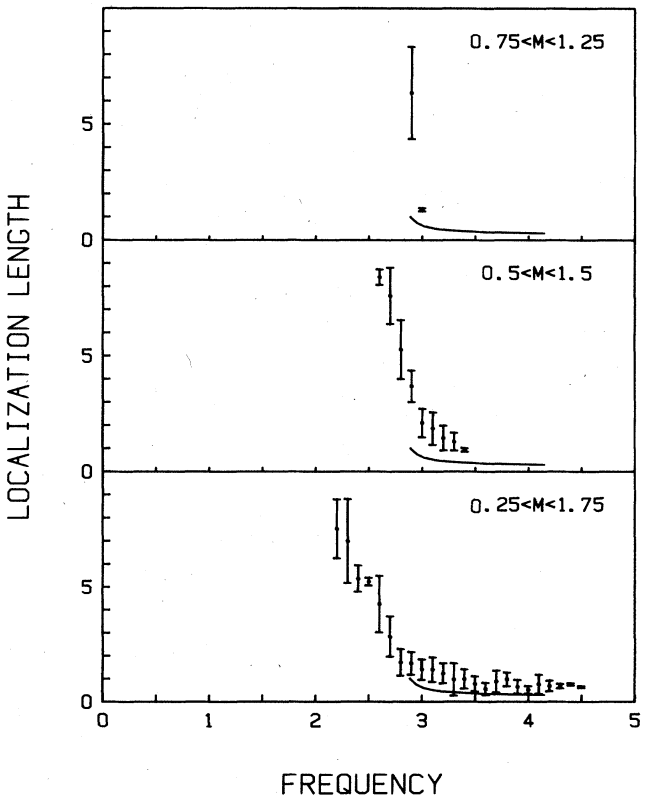

FIG. 3. Localization length ζ in units of the interatomic spacing as a function of phonon frequency for three amounts of disorder.

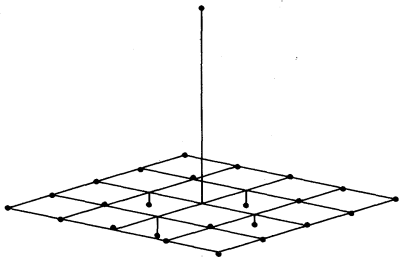

FIG. 4. Displacement pattern of highly localized modes. In this example the frequency is 4.0, the disorder parameter $q=0.75$, and the mass of the center atom is 0.269.

tern of a single light impurity in an otherwise perfect crystal. To develop this idea further we have made the following analysis. For each localized mode we have found the mass M_0 of the atom with the largest average energy. The average of M_0 for all the modes studied with a given ω is shown in Fig. 5. (This figure includes only those modes which have fairly short localization lengths. When ξ is large, there are often several atoms which have energy nearly as large as the energy of the highest-energy atom. Thus, the determination of M_0 becomes rather arbitrary.) For a single impurity of mass M_0 in a perfect lattice of mass M, the impurity-mode frequency is the solution of the equation[9]

$$M=(M-M_0)\omega^2\int_0^\infty \frac{g(\zeta)}{\omega^2-\zeta^2}d\zeta\ . \tag{59}$$

The relation between M_0 and ω is shown as the solid curve in Fig. 5. It is in reasonably good agreement with the numerical results.

We have tried to use the same simple picture to explain the localization length. For a single-impurity atom of mass M_0 in a lattice of atoms of mass M, it is straightforward to calculate the localization length ζ_I of the mode associated with the impurity. The result of this calculation is shown by the solid lines in Fig. 3. ζ_I is significantly smaller (typically by a factor of 2 or 3) than the ζ computed numerically for the disordered system. Examination of the displacement patterns for the modes in the disordered system reveals the following explanation. There will nearly always be at least one of the nearest or next-nearest neighbors of atom 0 which has a mass fairly close to M_0. This atom (or atoms) has a natural resonant frequency close to the mode frequency, and hence tends to have an unusually large amplitude This, in turn, makes a large contribution to ζ. For a single isolated impurity in an ordered lattice, this effect does not occur.

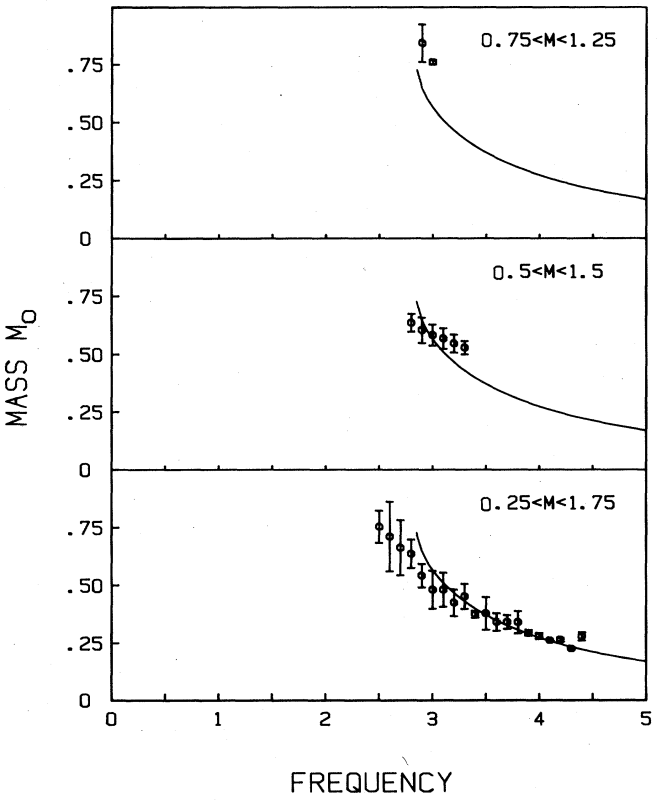

FIG. 5. Mass M_0 of the atom with the largest energy in a localized mode as a function of mode frequency ω. The solid curve is discussed in the text.

C. Mode patterns

We have already described the way the modes look for the highly localized states. For the modes with larger values of ζ, the mode pattern depends not only on ζ, but also on the amount of disorder q. Figure 6 shows two modes which have the same frequency (3.0), almost the same ζ [1.29 for (a), and 1.33 for (b)], but are in lattices with $q=0.25$ for (a) and 0.75 for (b). The atom with the largest energy is at the center of each plot (coordinates 0,0). Mode (a) has a frequency very close to the maximum frequency for disorder 0.25. This mode has a pattern which is locally identical with the pattern for the highest frequency mode in the ordered lattice, i.e., the mode at the corner of the Brillouin zone. The magnitude $|e_{l_x l_y}|$ varies fairly smoothly from atom to atom. For

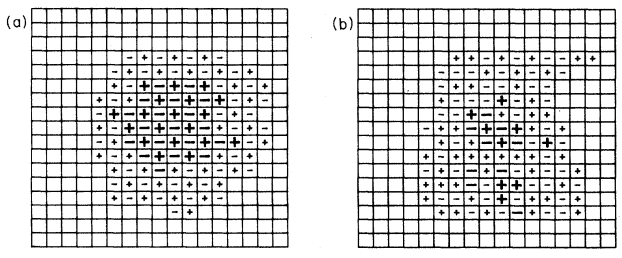

FIG. 6. Mode patterns. The atom with largest energy is at the center of the pattern. Large + and − denote atoms whose amplitudes have magnitudes greater than 0.1 of the amplitude e_0 of the center atom. Small + and − denote atoms whose amplitudes have magnitudes in the range 0.01 to 0.1 for e_0. (a) $\omega=3.0$, $\zeta=1.29$, $q=0.25$. (b) $\omega=3.0$, $\zeta=1.33$, $q=0.75$.

mode (b) the displacements are much more irregular. The atom with largest energy has mass 0.53. There is a second group of atoms with large amplitude around atom $(1,-4)$. This atom has mass 0.41, and thus has a resonant frequency close to the mode frequency.

D. Efficiency of the algorithm

Consider first the number n_1 of computational steps required to calculate a single-mode pattern. To make an order-of-magnitude estimate of n_1, it is sufficient to consider how n_1 depends on the the total number N of atoms in the system. Consider first what happens when the modes are sufficiently localized that the overlap of modes of nearly the same frequency is not significant. Thus, the simple method of Sec. II B can be used. The time t that the system must be driven $\propto N$, and so the number of time steps also $\propto N$. Each time step involves a number of computations $\propto N$, and so the total number of computational steps is

$$n_1 \propto N^2 . \tag{60}$$

If the modes overlap, it is necessary to use the more elaborate scheme in which the applied forces are changed. In this scheme there are now $\sim N^2$ steps between each time at which the force is changed. The accuracy of the method increases exponentially with the number of times p the force is changed, but for a given accuracy this number does not depend on N. Hence, we still have $n_1 \propto N^2$, but n_1 is larger than the n_1 needed when the modes do not overlap by a factor which is independent of N. This factor increases logarithmically as the required accuracy increases. We have typically used $p=10$ to 30, which gives modes patterns which have errors of less than $\pm 10^{-4}$.

Consider now the number of steps n_2 required to calculate the density of states g. In our method the number of modes $\tilde{N}$ which contribute to the energy transfer E [Eq. (5)] is of order

$$\frac{4\pi N}{\omega_m t} .$$

The coupling to each of these modes is random because of the random phases $\{\phi_l\}$. Thus, there will be random fluctuations in the energy transferred which cause E to differ from $\langle E \rangle$ by an amount which is of the order of $\langle E \rangle \tilde{N}^{-1/2}$. Hence, the fractional error ϵ in g is of order

$$\epsilon \sim \left[\frac{\omega_m t}{4\pi N} \right]^{1/2} . \tag{61}$$

It appears from this result that the error is smallest for small t. However, one has to recognize that the range of frequency $\Delta\Omega$ of the excited modes is $\sim 4\pi/t$. Thus, we actually determine the *average* of the density of states over the range $\Delta\Omega$, and this range increases as t decreases. Let us suppose therefore that we want to determine $g(\Omega)$ within a given frequency range of $\Delta\Omega$ and with a fractional uncertainty of ϵ. To do this we must have t and N of the order of

$$t \sim 1/\Delta\Omega , \tag{62}$$

$$N \sim \omega_m/\epsilon^2 \Delta\Omega . \tag{63}$$

Using our algorithm the time step is of order ω_m^{-1}. It follows that the number of computational steps needed is of order

$$n_2 \sim \left[\frac{\omega_m}{\epsilon \Delta\Omega} \right]^2 . \tag{64}$$

This argument assumes, of course, that N is sufficiently large that the density of states can be considered to be independent of N.

We now compare these results with other methods. Reviews have been given by Dean[1] and by Bell.[12] Straightforward diagonalization of a general $N \times N$ matrix requires of the order of N^3 steps, as well as storage of order N^2. However, for any system in which the interatomic forces are of short range the matrix to be diagonalized is sparse. One can then use the negative eigenvalue theorem[1] (NET) to find the number of eigenvalues less than any chosen value. It is then possible to proceed to find the normal mode frequencies, the density of states, and the mode patterns. The number of steps required for one application of the NET is of order Ns^2, where s is the width of the band of nonzero elements along the diagonal of the matrix. s is of order $N^{1-1/d}$ (d=dimensionality) and so the number of steps is of order

$$N^{3-2/d} . \tag{65}$$

The number of steps n_1' needed to find an eigenfunction is of this same order of magnitude. Consider now the number of steps n_2' needed to find the density of states. By two applications of the NET, one can find the number of eigenvalues $\tilde{N}$ in the range $\Delta\Omega$. It is clear that this number must have an error which is at least of order unity, and so the fractional error in g is

$$\epsilon \sim \tilde{N}^{-1} ; \tag{66}$$

but $\tilde{N}$ is of order $N\Delta\Omega/\omega_m$, and so one must choose

$$N \sim \frac{\omega_m}{\epsilon \Delta\Omega} . \tag{67}$$

Hence, the number of steps needed to find g averaged over a range $\Delta\Omega$ with a fractional uncertainty ϵ is

$$n_2' \sim \left[\frac{\omega_m}{\epsilon \Delta\Omega} \right]^{3-2/d} . \tag{68}$$

We can now compare n_1 and n_2 for our method [Eqs. (60) and (64)] with the corresponding n_1' and n_2' for the NET method. We see that in 2 dimensions the number of steps is the same. In this case our method may have some advantage, however, because it is known that for the NET method, there are some stability problems associated with finding the eigenfunctions. These difficulties do not occur in our method. In three dimensions, our method is clearly faster. For example, to find a mode pattern the number of steps needed is smaller by a factor of $N^{1/3}$.

We currently apply this numerical method to study phonon localization in three dimensions, and the effect of different types of disorder, such as topological defects. We will report these results in a later paper.

ACKNOWLEDGMENTS

This work was supported in part by the National Science Foundation through the Materials Research Laboratory at Brown University. We wish to thank P. J. Stiles and R. Zeller for the use of their computer, and J. M. Kosterlitz for helpful discussions.

---

[1]See, for example, P. Dean, Rev. Mod. Phys. **44**, 127 (1972).

[2]P. W. Anderson, Phys. Rev. **109**, 1492 (1958).

[3]K. Ishii, Prog. Theor. Phys. Suppl. **53**, 77 (1973).

[4]J. Jäckle, Solid State Commun. **39**, 1261 (1981).

[5]S. R. Nagel, A. Rahman, and G. S. Grest, Phys. Rev. Lett. **47**, 1665 (1981).

[6]This statement is true provided the density of states is not unusually small at the frequency Ω. The number of modes per frequency interval is of the order of $N/\omega_{\max}$, and so the number which are within $\pm(2\pi/t)$ of Ω is $4\pi N/\omega_{\max}t$.

[7]This assumption can be checked by time developing the lattice further without the driving force and comparing the mode patterns. Overlapping modes will display beating. This procedure was used for every mode discussed in the results.

[8]W. A. Bowers and H. B. Rosenstock, J. Chem. Phys. **18**, 1056 (1950).

[9]For a discussion, see A. A. Maradudin, E. W. Montroll, and G. H. Weiss, in *Solid State Physics,* edited by F. Seitz and D. Turnbull (Academic, New York, 1963), Suppl. 3.

[10]E. Abraham, P. W. Anderson, D. C. Licciardello, and T. V. Ramakrishnan, Phys. Rev. Lett. **42**, 673 (1979).

[11]S. John, H. Sompolinsky, and M. J. Stephen, Phys. Rev. B **27**, 5592 (1983).

[12]R. J. Bell, in *Methods in Computational Physics,* edited by G. Gilat (Academic, New York, 1976), Vol. 15, p. 215.