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Numerical Computation of Time-Dependent Properties of Isotopically Disordered One-Dimensional Harmonic Crystal Lattices

Robert J. RUBIN

National Bureau of Standards, Washington 25, D. C., U.S.A.

The principal purpose of this work is to investigate numerically the statistical dynamical properties of isotopically disordered harmonic crystal lattices. Two properties are studied in detail: 1) The decay of initial periodic disturbances $\dot{Q}_k(\tau)$ which are normal modes of the same lattice structure when all particles have the same mass; and 2) the dipole moment correlation function $\langle\dot{M}(0)M(\tau)\rangle$. The numerical calculation of the time evolution of either $\dot{Q}_k(\tau)$ or $\langle\dot{M}(0)M(\tau)\rangle$ involves the solution of a single initial value problem for a given random distribution of the isotope masses on the lattice sites. The time evolution is investigated for values of the isotope mass ratio m_1/m_2 and of k for which the perturbation results of Maradudin, Weiss, and Jepsen are not applicable.

A new formulation of the formal solution of the normal mode decay problem is proposed. This formulation should make it possible to calculate the initial stages of decay in an infinite lattice for the extreme values of m_1/m_2 and k studied in this paper.

1. Introduction

The principal purpose of the work reported in this paper is to investigate numerically the effect of isotope disorder on the statistical dynamical properties of one-dimensional harmonic crystal lattices. The lattice model which is treated is a perfect periodic lattice in which a fraction p of the lattice particles have mass m_1 and the remaining fraction have mass m_2. The forces between particles are linear in their relative displacements. Two dynamical properties are studied in detail: 1) The decay of initial periodic disturbances which are normal modes of the same lattice structure when all particles have the same mass; and 2) the dipole moment correlation function $\langle\dot{M}(0)M(\tau)\rangle$ which has been related to the dielectric susceptibility by Kubo.[1] The first property is determined from an initial value problem; and the second property, the ensemble average $\langle\dot{M}(0)M(\tau)\rangle$, is also determined from a single initial value problem. We have carried out numerical computations for a series of one-dimensional lattices, each consisting of 100 particles with nearest-neighbor interactions and periodic boundary conditions; 50 of the particles have mass m_1 and 50 have mass m_2. For a series of twenty different random distributions of the particles over the lattice sites, and for two values of the relative mass m_1/m_2, we have solved the pertinent initial value problems with the aid of the IBM 7090. A few calculations have also been made for lattices containing 200 particles.

There has been considerable interest recently[2)-5)] in the calculation of the time-dependent properties of isotopically disordered harmonic crystal lattices. Maradudin, Weiss, and Jepsen[3] (MWJ) have investigated the same two problems which we consider in this paper. However they use a perturbation formalism based on techniques developed by van Hove[6] and Prigogine and co-workers[7] for the treatment of dissipative effects in many-particle systems. Consequently some aspects of our exact numerical calculations for a system of 100 particles can be compared with the perturbation results of MWJ for an infinite system. In the remainder of this Section we will summarize those results obtained by MWJ which have motivated our own work.

MWJ obtain an approximate identity between $\langle\dot{M}(0)M(\tau)\rangle$ and an initial value problem involving the optically active normal mode (highest frequency mode) in a lattice of identical particles [relation (a)]. In addition they show that in the perfect lattice normal mode representation, the k^{th} normal mode of the perfect lattice, as an initial condition in the disordered lattice, decays exponentially as

$$\dot{Q}_k(\tau)=\exp(-\Gamma_k\tau)\cos\omega_k^{(0)}\tau \quad \text{[relation (b)]}, \tag{A1}$$

where to the lowest order in the perturbation

$$\Gamma_k=\frac{1}{32}\left(\frac{m_1^2-m_2^2}{m_1m_2}\right)^2\frac{\omega_k^{(0)2}}{[\omega_N^{(0)2}-\omega_k^{(0)2}]^{1/2}} \quad \text{for } 0\leqslant\omega_k^{(0)}<\omega_N^{(0)}; \tag{A2}$$

and $\omega_k^{(0)}$ is the frequency of the k^{th} normal mode of the perfect lattice when all particles have the harmonic mean mass $\mu=2[m_1^{-1}+m_2^{-1}]^{-1}$.

With relation (b) of MWJ in mind, we have studied numerically, in the perfect lattice normal mode representation, the evolution of the k^{th} normal mode of the perfect lattice as the initial condition in a series of isotopically disordered lattices. For mass ratios m_1/m_2 of 5/4 and 2, we determine the decay of mode $k=50$ in a 100 particle lattice, *i.e.*, a mode whose frequency lies in the middle of the band. For the larger mass ratio $m_1/m_2=2$, the MWJ perturbation result cannot be expected to be valid because it [Eq. (A1)] is derived on the assumption that the deviation from the average mass at a lattice site constitutes a weak perturbation.

We have also studied the decay of the highest frequency perfect lattice normal mode for a series of isotically disordered lattices with $m_1/m_2=5/4$. The decay of this mode is of particular interest for two reasons. First, MWJ have established a connection between it and the decay of $\langle\dot{M}(0)M(\tau)\rangle$, relation (a); and second, the expression which they give for the damping constant must be carefully interpreted because the denominator in Eq. (A2) is zero.

2. Formal Solution of the Initial Value Problems

In this Section we obtain formal solutions of the initial value problems associated with the decay of a perfect lattice normal mode and the decay of the dipole moment correlation function. The formal solutions are well-suited for numerical computation.

The kinetic and potential energy quadratic forms for a one-dimensional harmonic crystal lattice can be written most conveniently in matrix notation as

$$\frac{1}{2}\dot{\boldsymbol{x}}^T\mathscr{M}\dot{\boldsymbol{x}} \quad \text{and} \quad \frac{1}{2}\boldsymbol{x}^T\mathscr{V}\boldsymbol{x},$$

respectively, where $\mathscr{M}$ is a diagonal matrix whose i^{th} diagonal element is the mass of the particle at lattice site i. In this paper we assume that the lattice is composed of an equal number of particles of mass m_1 and mass m_2. $\mathscr{V}$ is the potential energy matrix and exhibits the periodicity of the lattice. The quantities $\boldsymbol{x}$ and $\dot{\boldsymbol{x}}$ are column vectors whose i^{th} components are, respectively, the displacement from equilibrium and the velocity of particle i. The superscript ^{T} denotes the transpose of a matrix (or vector). The classical equations of motion in this notation are

$$\mathscr{M}\ddot{\boldsymbol{x}}=-\mathscr{V}\boldsymbol{x}. \tag{B1}$$

To solve Eq. (B 1), introduce the vector $\boldsymbol{\xi}=\mathscr{M}^{1/2}\boldsymbol{x}$ and obtain

$$\ddot{\boldsymbol{\xi}}=-\boldsymbol{V}\boldsymbol{\xi},$$

where $\boldsymbol{V}=\mathscr{M}^{-1/2}\mathscr{V}\mathscr{M}^{-1/2}$. Then define the new vector $\mathscr{Q}=\mathscr{T}^T\boldsymbol{\xi}$, where the j^{th} column of $\mathscr{T}$, $\boldsymbol{t}_j$, is the j^{th} normalized eigenvector of the symmetric matrix $\boldsymbol{V}$ associated with the frequency ω_j. [$\mathscr{T}$ is an orthogonal matrix and $\sum_j\mathscr{T}_{jk}^2=\sum_k\mathscr{T}_{jk}^2=1$.] The equation of motion in the new variables is diagonal in form

$$\ddot{\mathscr{Q}}=-\boldsymbol{\Omega}^2\mathscr{Q}, \tag{B2}$$

where the j^{th} equation of motion is $\ddot{\mathscr{Q}}_j=-\omega_j^2\mathscr{Q}_j$. The general solution of Eq. (B2) is

$$\mathscr{Q}(\tau)=\boldsymbol{\Omega}^{-1}\sin\boldsymbol{\Omega}\tau\dot{\mathscr{Q}}(0)+\cos\boldsymbol{\Omega}\tau\mathscr{Q}(0) \tag{B3}$$

and

$$\dot{\mathscr{Q}}(\tau)=\cos\boldsymbol{\Omega}\tau\dot{\mathscr{Q}}(0)-\boldsymbol{\Omega}\sin\boldsymbol{\Omega}\tau\mathscr{Q}(0), \tag{B4}$$

where the matrix functions $\sin\boldsymbol{\Omega}\tau$ and $\cos\boldsymbol{\Omega}\tau$ are $\sum_{m=0}^{\infty}(-1)^m\frac{\tau^{2m+1}}{(2m+1)!}\boldsymbol{\Omega}^{2m+1}$ and $\sum_{m=0}^{\infty}(-1)^m\times\frac{\tau^{2m}}{(2m)!}\boldsymbol{\Omega}^{2m}$, respectively.

The general solution of Eq. (B1) is

$$\boldsymbol{x}(\tau)=\mathscr{M}^{-1/2}\mathscr{T}\boldsymbol{\Omega}^{-1}\sin\boldsymbol{\Omega}\tau\dot{\mathscr{Q}}(0)+\mathscr{M}^{-1/2}\mathscr{T}\cos\boldsymbol{\Omega}\tau\mathscr{Q}(0) \tag{B5}$$

and

$$\dot{\boldsymbol{x}}(\tau)=\mathscr{M}^{-1/2}\mathscr{T}\cos\boldsymbol{\Omega}\tau\dot{\mathscr{Q}}(0)-\mathscr{M}^{-1/2}\mathscr{T}\boldsymbol{\Omega}\sin\boldsymbol{\Omega}\tau\mathscr{Q}(0); \tag{B6}$$

and in terms of the initial values $\boldsymbol{x}(0)$ and $\dot{\boldsymbol{x}}(0)$ the solution is

$$\boldsymbol{x}(\tau)=\mathscr{M}^{-1/2}\boldsymbol{V}^{-1/2}\sin\boldsymbol{V}^{1/2}\tau\mathscr{M}^{1/2}\dot{\boldsymbol{x}}(0)+\mathscr{M}^{-1/2}\cos\boldsymbol{V}^{1/2}\tau\mathscr{M}^{1/2}\boldsymbol{x}(0) \tag{B7}$$

and

$$\dot{\boldsymbol{x}}(\tau)=\mathscr{M}^{-1/2}\cos\boldsymbol{V}^{1/2}\tau\mathscr{M}^{1/2}\dot{\boldsymbol{x}}(0)$$

$$-\mathscr{M}^{-1/2}\boldsymbol{V}^{1/2}\sin\boldsymbol{V}^{1/2}\tau\mathscr{M}\boldsymbol{x}(0). \tag{B8}$$

Now consider the initial state of the lattice $\boldsymbol{x}(0)=0$ and $\dot{\boldsymbol{x}}(0)=\boldsymbol{t}_k^{(0)}$, where $\boldsymbol{t}_k^{(0)}$ is the k^{th} normal mode of the perfect lattice in which all particles have the harmonic mean mass $\mu=2(m_1^{-1}+m_2^{-1})^{-1}$. The positions and velocities of the lattice particles at time τ are

$$\boldsymbol{x}(\tau)=\mathscr{M}^{-1/2}\boldsymbol{V}^{-1/2}\sin\boldsymbol{V}^{1/2}\tau\mathscr{M}^{1/2}\boldsymbol{t}_k^{(0)} \tag{B9}$$

and

$$\dot{\boldsymbol{x}}(\tau)=\mathscr{M}^{-1/2}\cos\boldsymbol{V}^{1/2}\tau\mathscr{M}^{1/2}\boldsymbol{t}_k^{(0)}. \tag{B10}$$

The components of $\boldsymbol{x}(\tau)$ and $\dot{\boldsymbol{x}}(\tau)$ in the $\boldsymbol{t}_k^{(0)}$ direction are, respectively

$$\begin{aligned}Q_k(\tau)&=\boldsymbol{t}_k^{(0)T}\boldsymbol{x}(\tau)\\&=\boldsymbol{t}_k^{(0)T}\mathscr{M}^{-1/2}\boldsymbol{V}^{-1/2}\sin\boldsymbol{V}^{1/2}\tau\mathscr{M}^{1/2}\boldsymbol{t}_k^{(0)}\end{aligned} \tag{B11}$$

and

$$\begin{aligned}\dot{Q}_k(\tau)&=\boldsymbol{t}_k^{(0)T}\dot{\boldsymbol{x}}(\tau)\\&=\boldsymbol{t}_k^{(0)T}\mathscr{M}^{-1/2}\cos\boldsymbol{V}^{1/2}\tau\mathscr{M}^{1/2}\boldsymbol{t}_k^{(0)}.\end{aligned} \tag{B12}$$

Eq. (B 12) is the formal solution of the equations of motion describing the decay of the perfect lattice normal mode $\boldsymbol{t}_k^{(0)}$. In the case of the perfect lattice, Eqs. (B11) and (B12) reduce to

$$Q_k(\tau)=\omega_k^{(0)-1}\sin\omega_k^{(0)}\tau$$

and

$$\dot{Q}_k(\tau)=\cos\omega_k^{(0)}\tau.$$

The oscillatory decay in the amplitude of $Q_k(\tau)$ and $\dot{Q}_k(\tau)$ obtained by MWJ[3] [see Eq. (A1)] and the oscillatory decay found in the numerical computations described in the next Section is caused by the irregularity in the distribution of the masses m_1 and m_2.

We now obtain an expression for the ensemble average dipole moment correlation function for the same chain of alternately charged ions considered by MWJ. The dipole moment is given by

$$\begin{aligned}M(\tau)&=e\sum_i(-1)^i x_i(\tau)\\&=e\boldsymbol{q}^T\boldsymbol{x}(\tau)\end{aligned} \tag{B13}$$

where $\boldsymbol{q}$ is a column vector whose i^{th} component q_i is $(-1)^i$, and e is the magnitude of the effective ionic charge. Periodic boundary conditions are assumed. [$\boldsymbol{q}$ is proportional to the highest frequency normal mode vector $\boldsymbol{t}_N^{(0)}$ of the perfect lattice; $\boldsymbol{q}=N^{1/2}\boldsymbol{t}_N^{(0)}$ where N is the number of particles in the lattice.] Form the product $\dot{M}(0)M(\tau)$ using Eqs. (B13), (B5) and (B6),

$$\begin{aligned}\dot{M}(0)M(\tau)&=e^2[\boldsymbol{q}^T\dot{\boldsymbol{x}}(0)][\boldsymbol{q}^T\boldsymbol{x}(\tau)]\\&=e^2[\sum_{i,k}q_i\mathscr{M}_i^{-1/2}\mathscr{T}_{ik}\dot{\mathscr{Q}}_k(0)]\\&\quad\times[\sum_{j,l}q_j\mathscr{M}_j^{-1/2}(\mathscr{T}\boldsymbol{\Omega}^{-1}\sin\boldsymbol{\Omega}\tau)_{jl}\dot{\mathscr{Q}}_l(0)\\&\quad+q_j\mathscr{M}_j^{-1/2}(\mathscr{T}\cos\Omega\tau)_{jl}\mathscr{Q}_l(0)]\ .\end{aligned} \tag{B14}$$

In a canonical ensemble the coordinates and velocities have a simple gaussian distribution,

$$\mathscr{N}\exp\left[-\frac{1}{2kT}\dot{\mathscr{Q}}(0)^T\dot{\mathscr{Q}}(0)-\frac{1}{2kT}\mathscr{Q}(0)^T\boldsymbol{\Omega}^2\mathscr{Q}(0)\right],$$

so the ensemble averages of $\langle\dot{\mathscr{Q}}_k(0)\dot{\mathscr{Q}}_l(0)\rangle$ and $\langle\dot{\mathscr{Q}}_k(0)\mathscr{Q}_l(0)\rangle$ are, respectively, $kT\delta_{kl}$ and 0. Using these ensemble averages the ensemble average $\langle\dot{M}(0)M(\tau)\rangle$ can be written compactly as

$$\begin{aligned}&\langle\dot{M}(0)M(\tau)\rangle\\&=Ne^2kT\boldsymbol{t}_N^{(0)T}\mathscr{M}^{-1/2}\boldsymbol{V}^{-1/2}\\&\quad\times\sin\boldsymbol{V}^{1/2}\tau\mathscr{M}^{-1/2}\boldsymbol{t}_N^{(0)}\ .\end{aligned} \tag{B15}$$

Upon comparing Eqs. (B15) and (B12), it is seen that **the ensemble average $\langle\dot{M}(0)M(\tau)\rangle/Ne^2kT$ is**

$$\langle\dot{M}(0)M(\tau)\rangle/Ne^2kT=\boldsymbol{t}_N^{(0)T}\boldsymbol{x}^{(N)}(\tau)\ , \tag{B16}$$

where $\boldsymbol{x}^{(N)}(\tau)$ is derived from the initial condition for the lattice: $\boldsymbol{x}(0)=\boldsymbol{o}$ and $\dot{\boldsymbol{x}}(0)=\mathscr{M}^{-1}\times\boldsymbol{t}_N^{(0)}$. Relation (a) of MWJ is an approximation in which $\mathscr{M}^{-1}$ is replaced by $\mu^{-1}\mathbf{1}$ in $\dot{\boldsymbol{x}}(0)=\mathscr{M}^{-1}\boldsymbol{t}_N^{(0)}$, where $\mu^{-1}=1/2(m_1^{-1}+m_2^{-1})$.

3. Numerical Computations and Results

The method used for computing $\boldsymbol{x}(\tau)$ and $\dot{\boldsymbol{x}}(\tau)$ from initial conditions is best illustrated by considering the series expansion of any one of the three matrix functions of $\boldsymbol{V}^{1/2}$ in Eqs. (B7) and (B8), *e.g.*,

$$\cos\boldsymbol{V}^{1/2}\tau=\mathbf{1}-\frac{\tau_0^2}{2!}\boldsymbol{V}+\frac{\tau_0^4}{4!}\boldsymbol{V}^2-\frac{\tau_0^6}{6!}\boldsymbol{V}^3+\cdots\ . \tag{C1}$$

These series involve only **integer** powers of the known matrix $\boldsymbol{V}$ and therefore can be computed term by term. For a value of τ_0 which is roughly one-eighth of the shortest characteristic period of the perfect lattice composed of particles with the harmonic mean mass, the first eight or nine terms in the expansion of $\cos\boldsymbol{V}^{1/2}\tau$ are sufficient to give values of its elements correct to eight significant figures. In this way elements of the propagator matrices in Eqs. (B7) and (B8) are calculated using an IBM 7090. Once these matrices are known, the values of $\boldsymbol{x}(\tau_0)$ and $\dot{\boldsymbol{x}}(\tau_0)$ can be computed from the initial conditions. This process, when repeated n times, produces values of $\boldsymbol{x}(n\tau_0)$ and $\dot{\boldsymbol{x}}(n\tau_0)$.

We first describe the calculation of $\dot{Q}_k(n\tau_0)$ for a series of isotopically disordered lattices with $m_1/m_2=5/4$ and $m_1/m_2=2$. The lattices consist of 100 particles, 50 of mass $1.00m$ and 50 of mass $1.25m$ [or $2.00m$] with periodic boundary conditions. The mass distributions are formed by the computing machine using a random number generator.[8] The initial perfect lattice normal mode is $\boldsymbol{t}_{50}^{(0)T}=(2^{1/2}/10)(\cdots,1,0,-1,0,1,0,-1,0,\cdots)$. The associated frequency is $\omega_{50}^{(0)}=(1+4/5)^{1/2}$ [or $\omega_{50}^{(0)}=(1+1/2)^{1/2}$] in time units for which the maximum frequency is $2^{1/2}(1+4/5)^{1/2}$ [or $2^{1/2}(1+1/2)^{1/2}$]. The time interval τ_0 which is used in these calculations $\tau_0=\pi/4\omega_{50}^{(0)}$, one-eighth of the period of the corresponding perfect lattice normal mode. From the computed values of $\dot{Q}_{50}(n\tau_0)$ for each random lattice, we have determined the relative maxima of the oscillating decay curve for $\dot{Q}_{50}(\tau)$ by selecting the relative maxima of $\dot{Q}_{50}(n\tau_0)$. In this way a lower limit for the decay curve envelope is determined. In Fig. 1 these estimates of the relative maxima of $\dot{Q}_{50}(\tau)$ are plotted as a function of $\tau=8\tau_0$ for $m_1/m_2=1.25$ for 23 isotopically disordered lattices. The dashed curve in Fig. 1 is the envelope of the decay curve Eq. (A1) obtained by Maradudin, Weiss, and Jepsen.[3] It is seen in Fig. 1 that the average behavior for the set of decay curves is not inconsistent with Eq. (A1). During the time interval plotted, a signal propagating with the speed of sound travels approximately 1/10th of the length of the 100-particle lattice. Particularly for times between $\tau=8$ and 20, the average curve is systematically larger than the simple exponential decay formula. This deviation of the average decay envelope from the simple relation $\exp(-\Gamma_{50}\tau)$, and the distribution of individual decay curves around the average should be interpretable within the framework of the perturbation theory of an isotopically disordered lattice consisting of a finite number of particles. Because we have not calculated the next order correction to Γ_{50} using the MWJ procedure, we can only speculate concerning the deviation of the average decay curve from $\exp(-\Gamma_{50}\tau)$. However, we have experimented with the random mass distributions by combining a mass distribution

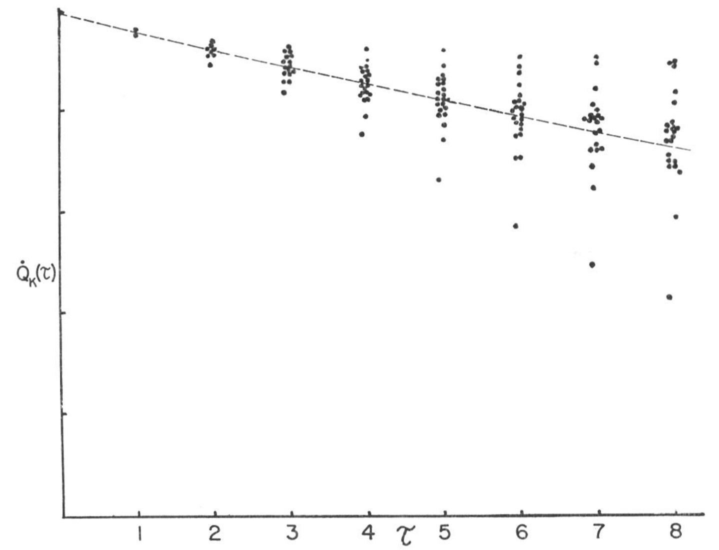

Fig. 1. Lower bounds of the relative maxima of the oscillations of $Q_{50}(\tau)$ *vs* τ for 23 different isotopically disordered 100-particle lattices in which the mass ratio $m_1/m_2=5/4$. The time τ is measured in units of $\tau_{50}^{(0)}$, the period of the monatomic lattice normal mode $k=50$ when the particle mass is equal to the harmonic mean of m_1 and m_2. In the time $\tau=8$, a signal propagating at the speed of sound travels approximately 1/10th of the length of the 100-particle lattice.

giving a high decay curve with one giving a low decay curve. In such a case, when the initial value problem for the combined 200 mass system was solved, the corresponding decay curve was considerably closer to exp $(-\Gamma_{50}\tau)$ and to the average of the two original decay curves.

In Fig. 2 analogous estimates of the relative maxima of the decay curves of $\dot{Q}_{50}(\tau)$ for 10 of the mass distributions included in Fig. 1 are plotted for the case in which $m_1/m_2=2$. The dashed curve, which is the plot of the first order perturbation value of $\exp(-\Gamma_{50}\tau)$ for the mass ratio $m_1/m_2=2$, clearly does not represent the average behavior for the 10 lattices. The average periods of the oscillations in this case as determined form the values of $\dot{Q}_{50}(n\tau_0)$ deviate noticeably from the period $8\tau_0=2\pi/\omega_{50}^{(0)}$. In the calculations in the preceding case, $m_1/m_2=5/4$, the average period remained within one percent of $2\pi/\omega_{50}^{(0)}$ for time intervals two to three times as long as that covered in Fig. 1.

Finally, in Fig. 3, analogous estimates of the decay curves of $\langle \dot{M}(0)M(\tau)\rangle/Ne^2kT$ are plotted as a function of the time τ for the mass ratio 5/4 for 10 disordered lattices. The basic interval τ_0 for these calculations was chosen as $\tau_0=\pi/4[2(1+4/5)]^{1/2}$. In addition to computing

$$\langle \dot{M}(0)M(\tau)\rangle/Ne^2kT=\boldsymbol{t}_{100}^{(0)T}\boldsymbol{x}^{(100)}(\tau)$$

according to Eq. (B16), we have computed simultaneously the quantity $\mu^{-1}\boldsymbol{t}_{100}^{(0)T}\mathscr{M}\boldsymbol{x}^{(100)}(\tau)$ which is equal to $\mu^{-1}Q_{100}(\tau)$. Knowing these two quantities, relation (a) of MWJ can be tested. A comparison of the periods and the phases of $\boldsymbol{t}_{100}^{(0)T}\boldsymbol{x}^{(100)}(\tau)$ and $\mu^{-1}\boldsymbol{t}_{100}^{(0)}\mathscr{M}\boldsymbol{x}^{(100)}(\tau)$ shows that they are approximately the same. If the masses m_1 and m_2 were equal, then $\boldsymbol{t}_{100}^{(0)T}\boldsymbol{x}^{(100)}(\tau)$ would equal $\mu^{-1}\boldsymbol{t}_{100}^{(0)T}\mathscr{M}\boldsymbol{x}^{(100)}(\tau)$. For the 10 isotopically disordered lattices considered here, and for $m_1/m_2=5/4$, the relative maxima of of $\boldsymbol{t}_{100}^{(0)T}\boldsymbol{x}^{(100)}(\tau)$ are greater than or equal to the corresponding relative maxima of $\mu^{-1}\boldsymbol{t}_{100}^{(0)T}\times\mathscr{M}\boldsymbol{x}^{(100)}(\tau)$. The deviation of the ratio of the corresponding maxima from the value one varies from lattice to lattice and varies in the course of oscillation of a particular lattice. The variation in the ratio is necessarily less than $|m_1-\mu|/\mu$ but does cover the entire range between 0 and 0.02.

During the first eight oscillations, the computations for Fig. 2 exhibit a large erratic variation in the amplitudes of successive maxima which is not found in the computations for Figs. 1 and 3. The amplitudes of successive maxima for two extreme examples are identified in Fig. 2 by dots in combination with diagonal lines running either from left to right or right to left.

4. Concluding Remarks

Large values of the mass ratio in the dynamical problems considered here lead to strong coupling between the unperturbed or perfect lattice normal modes. Consequently, a low-order perturbation calculation of the damping constant Γ_k would not be expected to be valid. The expression for Γ_k obtained by MWJ cannot be used to estimate the numerical value of Γ_k for the highest frequency modes, even for values of the mass ratio close to unity. Nevertheless, in an isotopically disordered lattice consisting of a large number of particles, it would be expected from the law of large numbers that the decay of a perfect lattice normal mode $\dot{Q}_k(\tau)$ in either of the extreme cases considered here would be given by the average $\dot{Q}(\tau)$ for all isotope configurations. This expectation is based on the structure of the expression for $\dot{Q}_k(\tau)$, which is basically an average over the entire lattice.

Finally we wish to draw attention to a new representation of the propagator matrices in Eqs. (B5) and (B6) which may facilitate the calculation of the early stages of the decay of quantities such as $\dot{Q}_k(\tau)$ for N large and $k\sim N$. The representation is based on a well-known form of the generating function for Bessel functions[9]

$$\begin{aligned}\cos zw &= J_0(z)+2\sum_{n=1}^{\infty}J_{2n}(z)\cos(2n\sin^{-1}w)\\ &= J_0(z)+2\sum_{n=1}^{\infty}(-1)^nJ_{2n}(z)T_{2n}(w),\end{aligned} \tag{C2}$$

where $J_{2n}(z)$ is the Bessel function of the first kind, and $T_{2n}(w)$ is the Tchebycheff polynomial

$$T_{2n}(w)=\cos(2n\cos^{-1}w).$$

If the time scale in the dynamical lattice problem is adjusted so that time is measured in units of the maximum frequency of a lattice composed entirely of light atoms, then the normal mode frequencies appearing in the

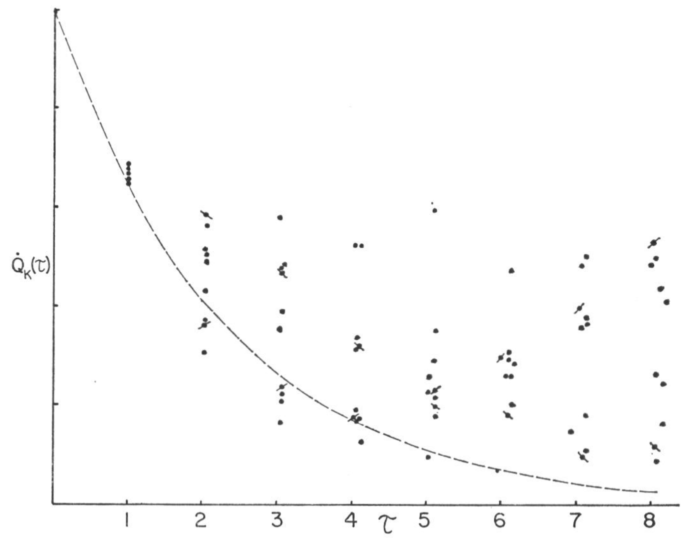

Fig. 2. Lower bounds of the relative maxima of the oscillations of $\dot{Q}_{50}(\tau)$ *vs* τ for 10 different isotopically disordered 100-particle lattices in which the mass ratio $m_1/m_2=2$. The time τ is measured in units of $\tau_{50}{}^{(0)}$, the period of the monatomic lattice normal mode $k=50$ when the particle mass is equal to the harmonic mean of m_1 and m_2. In the time $\tau=8$, a signal propagating at the speed of sound travels approximately 1/10 th of the length of the 100-particle lattice.

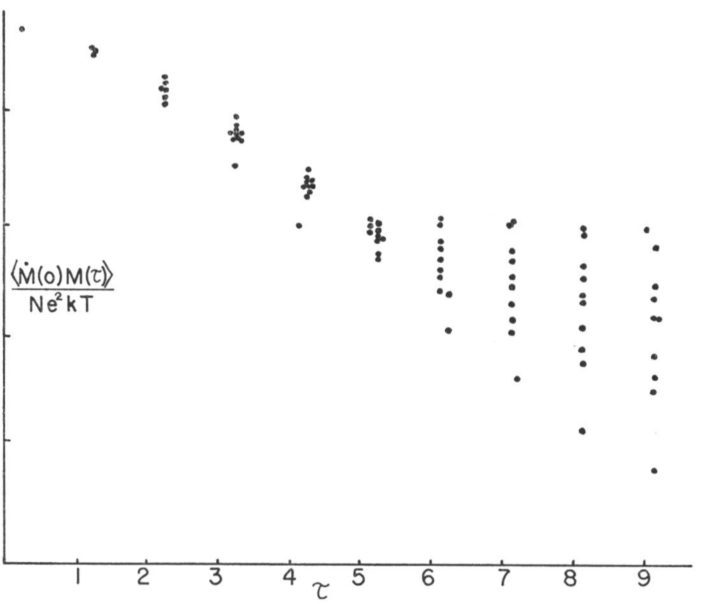

Fig. 3. Lower bounds of the relative maxima of the oscillations of $\langle\dot{M}(0)M(\tau)\rangle/Ne^2kT$ *vs* τ for 10 different isotopically disordered 100-particle lattices in which the mass ratio $m_1/m_2=5/4$. The time τ is measured in units of $\tau_{100}^{(0)}$, the shortest period of the monatomic lattice normal modes when the particle mass is equal to the harmonic mean of m_1 and m_2. In the time $\tau=8$, a signal propagating at the speed of sound travels approximately 1/14th of the length of the 100-particle lattice.

matrix $\cos \tau \boldsymbol{\Omega}$ are all necessarily less than one. It is then possible to replace $\cos \tau \boldsymbol{\Omega}$ by[10]

$$J_0(\tau)\mathbf{1}+2\sum_{n=1}^{\infty}(-1)^n J_{2n}(\tau)T_{2n}(\boldsymbol{\Omega})$$

and $\boldsymbol{\Omega}^{-1}\sin \boldsymbol{\Omega}\tau$ by a similar expression. Eq. (B12) for $Q_k(\tau)$ may then be written as

$$\begin{aligned}Q_k(\tau)&=\boldsymbol{t}_k^{(0)T}\mathscr{M}^{-1/2}\{J_0(\tau)\mathbf{1}\\&\quad+2\sum_{n=1}^{\infty}(-1)^n J_{2n}(\tau)T_{2n}(\boldsymbol{V}^{1/2})\}\mathscr{M}^{1/2}\boldsymbol{t}_k^{(0)}\\&=J_0(\tau)+2\sum_{n=1}^{\infty}(-1)^n J_{2n}(\tau)\\&\quad\times[\boldsymbol{t}_k^{(0)T}\mathscr{M}^{-1/2}T_{2n}(\boldsymbol{V}^{1/2})\mathscr{M}^{1/2}\boldsymbol{t}_k^{(0)}]\,.\end{aligned}$$

For τ not too large in an infinite lattice, $\dot{Q}_k(\tau)$ for $\tau<\tau'$ is expressible as a linear combination of a relatively small number of Bessel functions. The coefficients of the Bessel function of order $2p$ involves the p^{th} and smaller powers of $\mathscr{M}^{-1}\mathscr{V}$.

Acknowledgements

I wish to thank Dr. Hansjorg Oser for not only supervising the computations but for suggesting the particular method of calculation which was used. I also wish to thank David, Judith, and Karl Rubin for generating the first group of random mass distributions.

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10 The suggestion that z be identified with τ and w with frequency was made by Dr. R. Zwanzig in a conversation concerning the usefulness of Eq. (C2).

DISCUSSION

Hori, J.: How will the situation change if the number of particles becomes larger and larger?

Rubin, R. J.: As the number of particles in the lattice increases, fluctuations from the average behavior will decrease. For sufficiently small values of the mass difference and for k not too large, the average decay curve should be the same as that given by MWJ.