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Computer simulation on phonon transport in crystals with defects

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Abstract

Molecular dynamics computer simulation has been carried out to investigate phonon scattering in defect crystals at finite temperatures. One-dimensional mass-spring model crystals were used for the simulation. First, thermalization of input pulsed phonons in an isolated crystal was studied. An equipartition of phonon energies in the crystal was realized after a prolonged time, and thus a definite crystal temperature could be set up. Second, transport of pulsed phonons in a mass-defect crystal was simulated to evaluate the thermal diffusivity of the crystal. Dependences of the diffusivity on the crystal temperature, on the input phonon frequency, on the defect mass and defect concentration were investigated.

1. Introduction

The authors are studying a molecular dynamics computer simulation of phonon transport in one-dimensional mass-spring model crystals. The characteristics of our method are as follows: a transient phenomenon of propagation of pulsed phonons in the crystal is observed, the thermal diffusivity is determined, and the phonon scattering is investigated. In a previous paper [1], a preliminary study was described. The essential features, such as phonon dispersion, reflection of phonons, ballistic propagation of phonons, and diffusive transport of phonons, could be observed. The method was then applied to layer-stacked crystals, and a numerical evaluation was carried out for the case of thermal diffusivity of Langmuir–Blodgett films [2, 3]. The evaluation was compared successfully with our previous pulse-heating experiments on LB films of arachidic acid [4].

It is now intended to develop the simulation method to study phonon transport in crystals with defects. A trial is made for mass-defect crystals at various temperatures. First, the temperature must be set up, and then the diffusivity in the crystal is determined. A short report of our study has been presented [5], and further details of the temperature set-up method and a summary of the results obtained for simulated thermal diffusivity will be given here.

2. Methods

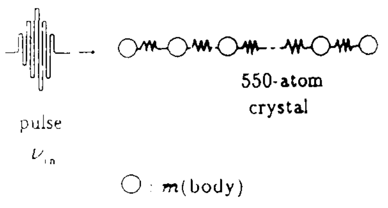
2.1. Models

Two kinds of one-dimensional mass-spring chain, as shown in Fig. 1, are taken as the model crystals.

2.1.1. Homogeneous crystal. The same masses (m), or atoms, are connected with the same springs. The number of atoms is 550, and the spacings between all atoms are the same (L). The springs are non-linear, and an anharmonic potential up to the fourth order is taken into account. The force acting on the ith atom from the neighbouring atoms is

$$F_i = -C^{(1)}(D_i - D_{i-1}) - C^{(2)}(D_i - D_{i-1})^2 - C^{(3)}(D_i - D_{i-1})^3 + C^{(1)}(D_{i+1} - D_i) + C^{(2)}(D_{i+1} - D_i)^2 + C^{(3)}(D_{i+1} - D_i)^3 \quad (1)$$

(a) Homogeneous crystal



(b) Mass-defect crystal

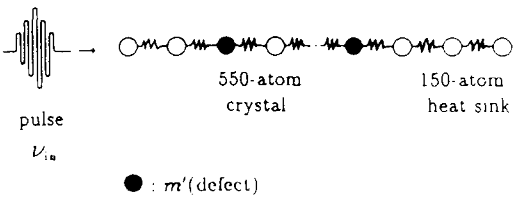


Fig. 1. Model crystals used for simulation of phonon thermalization and phonon transport.

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where D_i is the displacement of the ith atom, and $C^{(j)}$ is the jth-order force constant. We choose the ratio $C^{(1)}:C^{(2)}:C^{(3)}=1:-10/L:100/L^2$, being consistent with a conclusion of the higher-order elasticity theory [6] and also able to produce a reasonable interatomic potential curve.

2.1.2. Mass-defect crystal. Among the body atoms (m), a certain amount of defect atoms (m') are randomly distributed. The total number of atoms is 550 and the interatomic spacings and force constants are the same as in 2.1.1. To the right end of the crystal, another 150-atom homogeneous crystal is connected, which acts as a heat sink for propagating phonons.

2.2. Computation

The method of molecular dynamics (MD) is adopted in the present study. The equation of motion for the ith atom with mass m is

$$m\left(\frac{\mathrm{d}^2 D_i}{\mathrm{d}T^2}\right)=F_i \tag{2}$$

where T is the time. The equations for all of the atoms are numerically integrated by the Runge–Kutta–Gill method [7]. Displacement, velocity and kinetic energy of each atom are determined by the step-by-step method. A system of computational units (MD units), $L=1000$, $m=1$, $C^{(1)}=1$, is used, and a discrete time interval t (MD step) is taken. The maximum frequency of atomic vibration in a homogeneous harmonic crystal is [8] $\nu_{\mathrm{m}}=1/\pi$ for $m=C^{(1)}=1$, and the minimum period is $T_{\min}=3.14$. The MD step is here chosen as $t=0.1$, and the minimum period is $T_{\min}=31.4$ MD steps.

3. Simulation

3.1. Phonon state

An input phonon pulse is applied to one end of a homogeneous isolated crystal. The pulse is composed of step functions with a triangular envelope (Fig. 1(a)), and the frequency of the step-like vibrations is ν_{in}. The number of vibrations contained in a pulse is 16–250 for the lowest and highest input frequencies. The time variation of the atomic vibrational amplitude, $D(T)$, is recorded for each of the atoms in the crystal. The computation is prolonged to the time $T_0=1.1\times10^6$ MD steps. Two stages of the computation are chosen: the early stage $T=0$–$(1/140)T_0$ and the later stage $T=(139/140)T_0$–T_0. Samplings of the amplitude values are made in these two stages, and the sampling interval is as short as $(1/7.85)T_{\min}$. The conventional discrete or 'fast' Fourier transform procedure is carried out using these sampled points to obtain a frequency spectrum of vibrational amplitude. Spectra for eight different atoms in the crystal are chosen, and then these are averaged. The amplitude $D(\nu)$ thus obtained is squared and multiplied by ν^2 to obtain the frequency spectrum of the kinetic energy of atomic vibration. Obtained results are shown in Fig. 2 for various cases, where the left and the right figures represent those for the early and the later stages.

Computation was first performed for the case of a harmonic crystal ($C^{(2)}=C^{(3)}=0$ in eqn. (1)), and the results were as shown in Figs. 2(a) and (a′). In the early stage (a), peaks appear corresponding to the input frequency ν_{in} and its overtones in the frequency spectrum. It can be seen that the peaks continue in the later stage (a′).

In the case of an anharmonic crystal, the peaks in the early stage (b) disappear in the later stage (b′). The frequency spectrum becomes almost uniform, which means that a thermalization of input phonons and an equipartition of phonon energies in the crystal are realized. The temperature of the crystal can be defined by the averaged energy. Apparently, the inclusion of lattice anharmonicity producing phonon–phonon interaction is needed for the phonon thermalization.

In the above two cases, the frequency of input phonons is rather low: $\nu_{\mathrm{in}}/\nu_{\mathrm{m}}=0.061$. The results for input phonons with higher frequency, $\nu_{\mathrm{in}}/\nu_{\mathrm{m}}=0.83$, are shown in Figs. 2(c) and (c′). The observed behaviours are not well understood in this case. Phonon thermalization cannot be obtained even in the anharmonic crystal.

In conclusion, thermalization of input phonons and setting of crystal temperature are successfully realized, when the lattice anharmonicity is included and input phonons not so high in frequency are used.

3.2. Phonon transport

Transport of pulsed phonons was simulated for the mass-defect model crystals shown in Fig. 1(b). The crystal has been thermalized beforehand, and the same input phonon pulse as before is applied. The phonons propagate along the crystal and reach the heat sink at the right end of the crystal, and the heat sink eliminates the coming phonons. A transient propagation of phonons can thus be observed in the model crystal.

The state of the propagating phonons is represented as follows. The crystal is divided into layers of 10 atoms, and the vibrational energies of each atom are averaged over the layer. The time (T) variation of the energy K is observed at the Xth layer, and the energy-versus-time data are as shown in Fig. 3. By analysing the data we can determine the thermal diffusivity of the crystal, as shown in our previous paper [2].

The simulation conditions for the cases shown in Fig. 3 are:

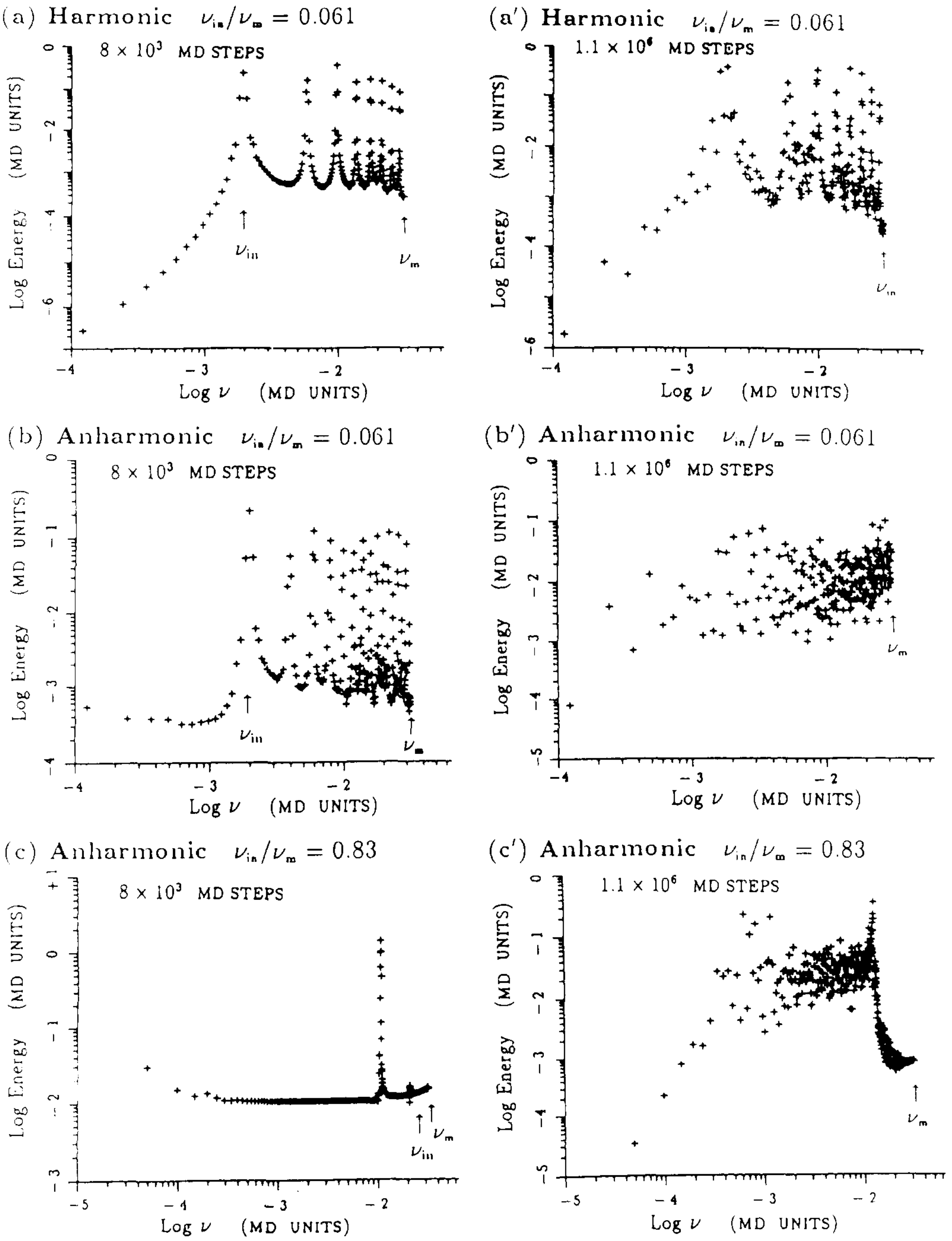

Fig. 2. Frequency spectra of phonons in various cases. The input phonon frequency is ν_{in}, and the maximum atomic vibrational frequency is ν_m: (a, a') harmonic, $\nu_{in}/\nu_m = 0.061$; (b, b') anharmonic; $\nu_{in}/\nu_m = 0.061$; (c, c') anharmonic, $\nu_{in}/\nu_m = 0.83$.

(a) The mass ratio is m'(defect)/m(body) = 3. The defect concentration is 10%. The input phonon frequency is $\nu_{in}/\nu_m = 0.061$. The crystal temperature is Θ_0 (arbitrary units). The observing position is $X = 20$.

(b) The crystal temperature is $5.3\Theta_0$. Other conditions are the same. Note that fluctuation in K *vs.* T is more marked in the high-temperature crystal. However, the determined diffusivity values are nearly the same for the above two cases, $D = 4.0$ (MD units): the thermal diffusivity is not sensitive to temperature in the case of the present mass-defect crystal. The phonon scattering that determines the diffusivity is mainly controlled by the crystal defects.

Simulations to determine the diffusivity are now being carried out for various cases of the mass-defect crystal. In the present stage of the study, the following relation seems to hold for the diffusivity:

$$D \propto \left[1-\left(\frac{\nu}{\nu_m}\right)\right]^{1/2} \times \left(\frac{\Delta m}{m_{av}}\right)^{-1} \times \left(\frac{N'}{N}\right)^{-1} \qquad (3)$$

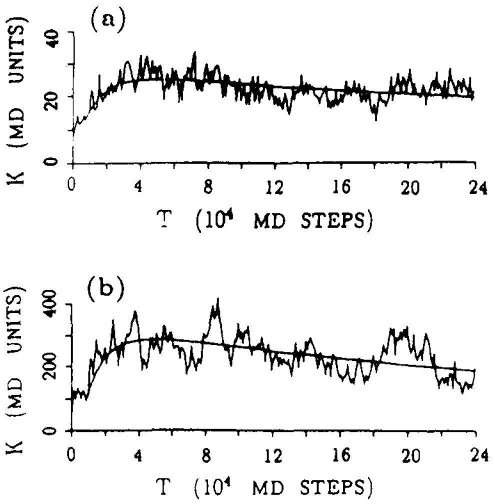

Fig. 3. Transport of phonons in crystal at (a) low and (b) high temperatures, showing kinetic energy K versus time T. The smooth solid curves are the parameter-fitted ones to determine the thermal diffusivity.

where ν is the frequency of input phonons; ν_m is the maximum phonon frequency of the crystal; N and N' are the total number of atoms in the crystal and the number of defect atoms; $m_{av}=m[(N-N')/N]+m'[N'/N]$; $\Delta m=m'-m_{av}$; and here m and m' are the masses of body and defect atoms.

The above result can qualitatively be explained by the relation

$$D\alpha vl \qquad (4)$$

which comes from the gas kinetics equation $\kappa=Cvl$ and the definition $D=\kappa/\rho C$, where κ is the thermal conductivity; C is the heat capacity per unit mass; ρ is the material density; v and l are the velocity and mean free path of phonons. It is considered that the first factor in the right-hand side of eqn. (3) represents the group velocity of phonons in a one-dimensional crystal; the second and third factors correspond to the mean free path controlled by the mass difference and the concentration of defect atoms [8]. The procedures of simulation to obtain the above result are rather lengthy, and details will not be presented here.

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