Phonon relaxation and heat conduction in one-dimensional Fermi–Pasta–Ulam β lattices by molecular dynamics simulations*

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The phonon relaxation and heat conduction in one-dimensional Fermi–Pasta–Ulam (FPU) β lattices are studied by using molecular dynamics simulations. The phonon relaxation rate, which dominates the length dependence of the FPU β lattice, is first calculated from the energy autocorrelation function for different modes at various temperatures through equilibrium molecular dynamics simulations. We find that the relaxation rate as a function of wave number k is proportional to $k^{1.688}$, which leads to a $N^{0.41}$ divergence of the thermal conductivity in the framework of Green–Kubo relation. This is also in good agreement with the data obtained by non-equilibrium molecular dynamics simulations which estimate the length dependence exponent of the thermal conductivity as 0.415. Our results confirm the $N^{2/5}$ divergence in one-dimensional FPU β lattices. The effects of the heat flux on the thermal conductivity are also studied by imposing different temperature differences on the two ends of the lattices. We find that the thermal conductivity is insensitive to the heat flux under our simulation conditions. It implies that the linear response theory is applicable towards the heat conduction in one-dimensional FPU β lattices.

Keywords: thermal conductivity, phonon relaxation, low-dimensional heat conduction, Fermi–Pasta–Ulam (FPU) β lattice

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1. Introduction

Many one-dimensional (1D) materials, such as nanotubes, nanowires and nanobelts, have been fabricated in the past decades due to the rapid advancement of nanoscience and nanotechnology.[1,2] Based on these nanomaterials, the construction of nanodevices, such as nano-electro-mechanical systems, micro/nanofluidic devices and lab-on-a-chip, can be realized, and new physics at nanoscale have been studied. The basic properties of these nanostructure materials have attracted a great deal of attention. Among various properties, electrical, magnetic, optical and mechanical properties have been studied more comprehensively as compared with thermal properties.[3–5] The thermal conductivity of one-dimensional materials, as an elementary property, however, is significantly related to the energy dissipation and transport, which is one of the most dominant bottlenecks of modern nanodevices. The thermal property of low-dimensional materials, which is usually different from those of its conventional counterparts, is highly desirable to be understood.

Low-dimensional materials have been found to have unique thermal properties compared with their bulk counterparts or other nanostructures. Typically, carbon nanotubes were reported to have a very high thermal conductivity, as high as 2000–6600 W/mK, by experimental measurements,[6–8] theoretical analyses[9–11] and molecular dynamics simulations[12–14] due to the low-dimensional confinement of phonons. The thermal conductivity of carbon nanotubes increases with the increasing tube length by using a power function.[14–16] The low-dimensional effects on thermal transport were also reported to come into play for polymer chains,[17] polyethylene

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nanofibres[18,19] and proteins.[20] In low-dimensional nanostructures, heat often transports in a diffusive-ballistic way, and the thermal conductivity of low-dimensional materials shows unique characteristics. Thus, the heat conduction under low-dimensional conditions remains obscure.

The heat conduction in an ideal one-dimensional Femi–Pasta–Ulam (FPU) β lattice has attracted much attention in recent years since it can reflect the thermal transport characteristics in low-dimensional structures.[21,22] Anomalous heat conduction has been found by several groups.[21−24] The thermal conductivity in a one-dimensional lattice is often proportional to L^{α}, where L is the system length and α is a coefficient. Based on the hydrodynamic equations and renormalization group theory, Narayan *et al.* proved $\alpha = 1/3$ in the thermodynamic limit.[25,26]However, the hydrodynamic equations might break down for a one-dimensional fluid system as pointed out by Hurtado.[27] Lepri put forward the mode-coupling theory and suggested that α should be $2/5$[28], which was also confirmed by the Peierls–Boltzmann theory.[29] In fact, other different coefficients were also reported through theoretical analyses or molecular dynamics simulations.[30,31] Further studies are still necessary towards the mechanism of energy transport through the one-dimensional lattice.

Heat is carried by phonons in the one-dimensional lattice. Although solitions can be excited in the nonlinear lattices, they have been demonstrated to provide no effective contribution to thermal conduction.[32] Therefore, the energy transport process should be dominated by the thermal relaxation of the vibration modes (phonons). However, focused research on this topic is rare. In this paper, we will study the relaxation rate of phonons in the FPU β lattice, and then obtain the relation between the thermal conductivity and the system scale (length L or number of atoms N). Our molecular dynamics simulations give an $N^{2/5}$ dependent thermal conductivity in the FPU β lattice, which validates the viewpoint from the mode-coupling and Peierls–Boltzmann theories.

2. Phonon relaxation and thermal conductivity

The Green-Kubo formula is widely used in discussions of thermal conductivity. The thermal conductivity in a one-dimensional system can be expressed as

$$\lambda = \frac{1}{k_{\mathrm{B}}T^2 L}\lim_{t\to\infty}\int_0^t \langle q\,(t)\,q\,(0)\rangle\,\mathrm{d}t, \tag{1}$$

where λ is the thermal conductivity, T is temperature, L is the system length, k_{B} is the Boltzmann constant, t is time, and $\langle q(t)q(0)\rangle$ is the heat flux autocorrelation function. The brackets denote the ensemble average. In this paper, the contribution of phonons to the heat conduction is considered, and then the heat flux can be expressed as

$$q = \sum_i n_i \hbar\omega_i v_i, \tag{2}$$

where n_i is the number of the phonons of the ith mode, $\hbar$ is the Planck constant, ω_i is the angle frequency, and v_i is the phonon group velocity. When the system is in an equilibrium state, the heat flux is zero; consequently, we can rewrite Eq. (2) as

$$q = \sum_i \delta n_i \hbar\omega_i v_i, \tag{3}$$

where $\delta n_i = n_i - n_{i,0}$, and $n_{i,0}$ is the phonon number of the ith mode in equilibrium. Using Eq. (3), we can obtain the heat flux autocorrelation function as

$$\langle q\,(t)\,q\,(0)\rangle = \sum_{i,j}\langle \delta e_i\,(t)\,\delta e_j\,(0)\rangle\,v_i v_j, \tag{4}$$

where $\delta e_i\,(t) = \delta n_i\,(t)\,\hbar\omega_i$ is the energy fluctuation of the phonon of the ith mode at time t. Assuming that the correlation function of different phonon modes is zero, Eq. (4) can be rewritten as

$$\langle q\,(t)\,q\,(0)\rangle = \sum_i \langle \delta e_i\,(t)\,\delta e_i\,(0)\rangle\,v_i^2. \tag{5}$$

Using the exponential function to describe the autocorrelation function $\langle \delta e_i(t)\delta e_i(0)\rangle$, and taking $\langle(\delta e_i)^2\rangle = (k_{\mathrm{B}}T)^2$, we can obtain

$$\langle q\,(t)\,q\,(0)\rangle = (k_{\mathrm{B}}T)^2\sum_i v_i^2\exp\left(-t/\tau_i\right), \tag{6}$$

where τ_i is the relaxation time of the phonons of the ith mode. Using the integral in place of the sum, and wave number k instead of i, Eq. (6) can be expressed as

$$\begin{aligned}\langle q\,(t)\,q\,(0)\rangle &= (k_{\mathrm{B}}T)^2\frac{L}{2\pi}\int_0^{\pi/a} v_k^2\exp\left(-t/\tau_k\right)\mathrm{d}k\\ &\equiv (k_{\mathrm{B}}T)^2\frac{L}{2\pi}D\,(t)\,,\end{aligned} \tag{7}$$

where a is the lattice constant. Assuming that the relationship between the relaxation rate and the wave vector can be expressed as $1/\tau_k \sim k^{\varsigma}$ (the relationship will be confirmed by molecular dynamics simulations in the next section), and considering only the long wave phonons, we can obtain

$$D\,(t) \sim t^{-1/\varsigma}. \tag{8}$$

Substituting Eqs. (7) and (8) into Eq. (1), we can obtain the relation between the thermal conductivity and the system size as

$$\lambda \sim L^{1-1/\varsigma}. \tag{9}$$

From the above derivation, we can see that the thermal conductivity of the one-dimensional lattice may be a power function of the system length, and the power coefficient relies on the relaxation rate of phonons. Therefore, learning the thermal transport in a one-dimensional lattice is dominated by understanding the relaxation time of phonons.

3. Models and simulation method

We use the molecular dynamics simulation method to study the vibration relaxation process and thermal conductivity of the 1D FPU β lattice. The Hamiltonian of the FPU β lattice reads

$$H = \frac{1}{2}\sum_i \left[u_i^2 + (r_{i+1} - r_i)^2 + \frac{\beta}{2}(r_{i+1} - r_i)^4\right], \tag{10}$$

where u_i and r_i are the velocity and displacement of atom i, respectively. In our study, the coefficient β is 0.1. In our simulations, the whole system includes 50–10000 atoms. The four order Runge–Kutta method is used to integrate the motion equation with a time step $\mathrm{d}t = 0.0005$. Periodic boundary conditions are applied. At the beginning, all atoms are placed at their equilibrium positions, and the velocities are randomly distributed according to the Gaussian distribution. After that the system freely evolves. The system temperature is defined by the average kinetic energy per atom as

$$T = \left\langle u_i^2 \right\rangle. \tag{11}$$

Here, we take the Boltzmann constant and the atom mass as units.

The phonon relaxation process is simulated by using equilibrium molecular dynamics (EMD) simulations. The first 2000000 steps are used to make the system reach a stationary state. In the following 2000000 steps, the velocity and the displacements are recorded to calculate the energy and energy autocorrelation function of various vibration modes. To reduce the error, five independent trajectories are calculated for every case. Finally, the phonon relaxation rate are estimated by fitting the energy autocorrelation function with exponential function $A_0\exp(-t/\tau_k)$, where A_0 is a coefficient.

For comparison, non-equilibrium molecular dynamics (NEMD) simulations are also performed to calculate the thermal conductivity of the FPU β chains with different system scales. In these simulations, fixed boundary conditions are used, and the atoms adjacent to the boundary are attached to Nose–Hoover thermostats at different temperatures. Note that different thermostats may produce different phonon modes in the boundary regions.[21,22] The first 1000000 steps are used to obtain a stationary state. In the following 5000000 steps, the heat flux and the temperature profile are calculated, and the thermal conductivity can be calculated according to Fourier's heat conduction law.

4. Simulation results and discussion

We calculate the phonon relaxation time as a function of wave number k at different temperatures by using EMD simulations first. The results are shown in Fig. 1, in which $k^* = ak/\pi$ is the dimensionless wave number. We can see that the relaxation rate increases with the increasing wave number for long wave phonons, and that the relaxation rate is greatly related to the temperature as well. Using an exponential function

$$\tau_k^{-1} = Ak^{\varsigma}T^C \tag{12}$$

to describe the relaxation rate, we can obtain the coefficients ζ and C by fitting all data. The coefficient ζ is estimated to be 1.688 ± 0.097 in our simulations, which is very close to 5/3 calculated by Pereverzev through the phonon Boltzmann equation.[29] According to Eq. (9), it will result in a length-dependent thermal conductivity as $\lambda \sim N^{0.41\pm0.03}$, which supports Lepri's results based on the mode-coupling theory.[28]

For comparison, we conduct non-equilibrium molecular dynamics (NEMD) simulations to further investigate the thermal transport in the FPU β chains. As the temperature difference of two ends increases, the temperature range also increases. To evaluate the influence of the temperature profile of a wide range, the thermal conductivity of an FPU β chain with 100 atoms is calculated with small temperature differences at various temperatures. As the temperature increases from 0.1 to 10000, the thermal conductivity decreases first, and then increases as shown in Fig. 2. There is a transition zone with the temperature between 1 and 100, in which the thermal conductivity almost does

not change. This phenomenon is in agreement with the results obtained in Refs. [33] and [34]. When investigating the effects of the system scale and heat flux, this temperature zone is preferable to decrease the effects from the temperature.

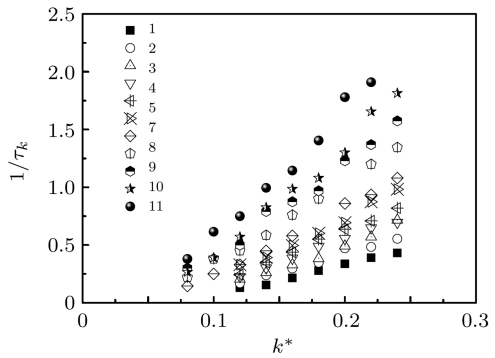


Fig. 1. Phonon relaxation rate as a function of wave number at different temperatures. The legends 1 to 11 represent the different system total energies (or temperatures), i.e., 10 (6.2), 20 (12.6), 30 (19.0), 40 (25.5), 60 (38.4), 100 (63.5), 200 (130.9), 400 (263.2), 600 (395), 1000 (663), 2000 (1327).

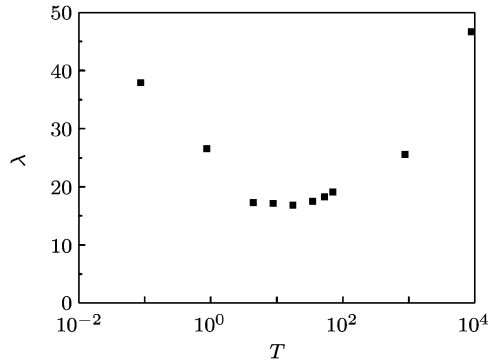


Fig. 2. Thermal conductivity varying with the temperature.

To validate the applicability of Fourier's heat conduction law, we then check the linear response theory in terms of one-dimensional systems. Based on the FPU β model, the effects of the heat flux on the thermal conductivity are studied by using NEMD simulations. Two systems with $N = 25$ and 100 respectively are simulated at $T = 88$. The thermal conductivity as a function of temperature difference at two ends is shown in Fig. 3. The thermal conductivity almost does not change as the temperature difference increases. In fact, the present heat flux is not high enough to cause the inertial and nonlocal effects on heat conduction.[35–37] It implies that the linear response theory has wide applicability in the low-dimensional models.

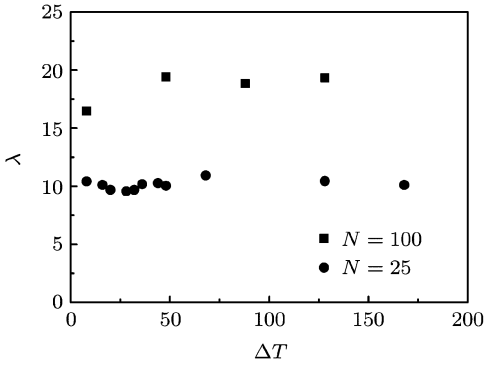


Fig. 3. Effects of the temperature difference ΔT on the thermal conductivity of the FPU β lattice.

Non-equilibrium molecular dynamics simulations are also performed to calculate the thermal conductivity of the FPU β chains with different system scales. The mean system temperature is 88, and the temperatures at the left and right ends are, respectively, maintained at 152 and 24. Since there are temperature jumps at the boundaries, we use the data in the middle part where the temperature profile is an approximate line to calculate the thermal gradient to be used in Fourier's heat conduction law. In our simulations, the thermal conductivity is proportional to $N^{0.415\pm0.013}$ as shown in Fig. 4. This result is consistent with that predicted by EMD calculation. Based on the analyses above, we can conclude that the thermal conductivity divergence results from the large relaxation time of the long wave normal mode in the FPU β lattice. It should be pointed out that those real low-dimensional materials, such as carbon nanotubes, graphene and polymer chains, have different power coefficients due to their different interaction potentials.

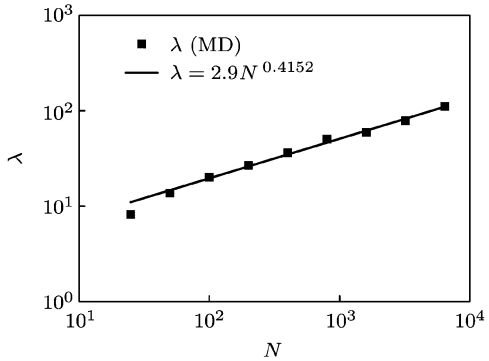


Fig. 4. Dependence of the thermal conductivity of the FPU β lattice on the system scale.

5. Conclusion

We have studied the phonon relaxation and heat conduction in one-dimensional Fermi–Pasta–Ulam β lattices by molecular dynamics simulations. The

phonon relaxation rate as a function of wave number k is proportional to $k^{1.688}$, and it leads to an $N^{0.41}$ divergence of the thermal conductivity. Our EMD and NEMD simulations agree well with each other. This result supports the viewpoint of Lepri based on mode-coupling theory. The heat flux or the temperature difference is found to have no significant effect on the heat conduction, which implies that the linear response theory is applicable to the heat conduction in one-dimensional FPU β lattices. The present results can also be applied to thermal transport in other low-dimensional materials, such as carbon nanotubes, graphene and polymer nanowires.

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