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Approach to Phonon Relaxation Time and Mean Free Path in Nonlinear Lattices

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Based on the self-consistent phonon theory, the spectral energy density is calculated by the canonical transformation and the Fourier transformation. Through fitting the spectral energy density by the Lorentzian profile, the phonon frequency as well as the phonon relaxation time is obtained in one-dimensional nonlinear lattices, which is validated in the Fermi–Pasta–Ulam-β (FPU-β) and ϕ^4 lattices at different temperatures. The phonon mean free path is then evaluated in terms of the phonon relaxation time and phonon group velocity. The results show that, in the FPU-β lattice, the phonon mean free path as well as the phonon relaxation time displays divergent power-law behavior. The divergent exponent coincides well with that derived from the Peierls–Boltzmann theory at weak anharmonic nonlinearity. The value of the divergent exponent expects a power-law divergent heat conductivity with system size, which violates Fourier's law. For the ϕ^4 lattice, both the phonon relaxation time and mean free path are finite, which ensures normal heat conduction.

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It is well known that heat conduction through macroscopic materials is ruled by Fourier's law[1]

$$j = -\kappa \nabla T, \tag{1}$$

where j, T and κ are the heat current density, temperature and heat conductivity, respectively. On the contrary, it is quite different for heat conduction in low-dimensional microscopic systems, which has drawn considerable attention in recent years as a fundamental topic for nonequilibrium statistical mechanics.[2,3] For momentum-conserving nonlinear lattices, e.g., the Fermi–Pasta–Ulam-β (FPU-β) lattice, heat conductivity κ diverges with the system size, which indicates the violation of Fourier's law and has been not only observed numerically,[4,5] but also observed experimentally in one-dimensional system[6] and analytically proved by renormalization group theory,[7] the mode coupling theory[2] and the nonlinear fluctuation thermodynamic method.[8] For momentum-nonconserving lattices, e.g., the ϕ^4 lattices, heat conductivity generally converges to a nonzero constant in the thermodynamic limit, in other words, Fourier's law is satisfied.[9–11] However, while much progress has been achieved, there are still some remarkable problems to be answered, which require in-depth understanding of phonons.

The properties of heat conduction for insulating materials are largely dominated by phonons which are collective excitations of atomic vibrations. For harmonic lattices, phonons transport ballistically with infinite mean free path, indicating the breaking of Fourier's law.[12] For anharmonic lattices, phonons interact with one another, which induces a complex shift and broadening of phonon frequencies. With anharmonicity involved, the contradiction between phonon excitation and existence of phonon decay emerges, which has attracted wide interests. A main difficulty of analyses lies in the presence of anharmonicity. For thermal conduction in systems of weak anharmonicity, a nonequilibrium Green's function method has been proposed to deal with phonon-phonon interactions.[13] For systems of strong anharmonicity, the self-consistent phonon theory (SCPT)[14–19] and the effective phonon theory[20–27] have been proposed, through finding an effective harmonic interaction. Recently, Liu *et al.* put forward a tuning-fork method,[28] which allows one to explicitly visualize the picture of phonon and evaluate the phonon dispersion relation and the phonon mean free path through molecular dynamics simulation. Additionally, in terms of the linear response theory, Xu and Wang proposed the resonance phonon approach,[29,30] via which the wave-number-dependent phonon frequency and relaxation time can be obtained by fitting the spatial Fourier transform of the velocity correlation function.

In this Letter, we propose an alternative method to determine the wave-number-dependent phonon frequency and relaxation time in one-dimensional nonlinear lattices via employing SCPT and the Lorentzian fitting.[31,32] Our method is validated in two typical models, namely, the FPU-β lattice and the ϕ^4 lattice. Based on SCPT, we demonstrate that the effective phonon modes remain unchanged even with the shifting of phonon frequencies for one-dimensional anharmonic systems of the homogeneous structure. Then the Fourier transformation of the canonical momen-

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tum, i.e., the spectral energy density, can be fitted by Lorentzian function in order to obtain the vibrating frequency and relaxation time of phonons. The effective phonon frequencies obtained via Lorentzian fitting agree well with those predicted by SCPT. The scaling behavior of the phonon relaxation time and mean free path is consistent with that obtained in previous studies.[28–30]

In literature, SCPT, which can be traced back to Feynman,[14] has been developed to study the properties of thermal conduction in low-dimensional nonlinear lattices[15–19] and the temperature of microcanonical ensemble.[33] Without loss of generality, we consider the Hamiltonian of a one-dimensional homogeneous system in the form of

$$H = \sum_n \frac{p_n^2}{2\,m} + W(\delta q_n) + V(q_n), \tag{2}$$

where $\delta q_n = q_n - q_{n-1}$, $W(\delta q)$ and $V(q)$ are the nearest-neighbor interaction and on-site potential, respectively. The partition function of systems in the canonical ensemble can be written as a standard form:

$$Z = \int \exp\Big[-\frac{H}{k_{\rm B}T}\Big] d\boldsymbol{q} d\boldsymbol{p}. \tag{3}$$

However, an exact evaluation of the integral of the above partition function is generally difficult or even impossible, due to the presence of nonlinear terms in Hamiltonian. The key idea of SCPT is to replace the original Hamiltonian by a trial Hamiltonian that allows an approximate evaluation of the exact partition function given by Eq. (3). A reasonable choice of the trial Hamiltonian is a chain of N coupled harmonic oscillators in the form

$$H_0 = \sum_n \frac{p_n^2}{2\,m} + \frac{f_{\rm c}}{2}(p_n - p_{n-1})^2 + \frac{f}{2}p_n^2, \tag{4}$$

where the trial parameters $f_{\rm c}$ and f are to be obtained by minimizing the right-hand side of the Feynman–Jensen inequality for free energies,

$$\mathcal{F} \leqslant \mathcal{F}_0 + \langle H - H_0\rangle_0. \tag{5}$$

In Eq. (5) the average is taken with respect to the bare harmonic system [Eq. (4)] of free energy $\mathcal{F}_0 = -k_{\rm B}T \ln Z_0$, where the trial partition function is written as

$$Z_0 = \int \exp\Big[-\frac{H_0}{k_{\rm B}T}\Big] d\boldsymbol{q} d\boldsymbol{p}. \tag{6}$$

The average in Eq. (5) can be easily calculated because the integrand is of a quadratic form. Then one can obtain the renormalized phonon frequency

$$\tilde{\omega}_k^2 = \frac{2}{m}\Big\{\frac{\partial V_\rho}{\partial \rho^2} + 4\sin^2\Big(\frac{k}{2}\Big)\frac{\partial W_\gamma}{\partial \gamma^2}\Big\}, \tag{7}$$

where V_ρ and W_γ are the average value of the on-site potential energy and the interaction potential energy, respectively,

$$\begin{aligned} V_\rho &= \int \frac{dy}{\sqrt{2\pi\rho^2}} e^{-\frac{y^2}{2\rho^2}} V(y), \\ W_\gamma &= \int \frac{dy}{\sqrt{2\pi\gamma^2}} e^{-\frac{y^2}{2\gamma^2}} W(y). \end{aligned} \tag{8}$$

In Eq. (8), ρ^2 and γ^2 correspond to the lattice displacement and the two-point correlation function, i.e.,

$$\rho^2 \equiv \langle q_n^2\rangle = \frac{k_{\rm B}T}{Nm}\sum_k \tilde{\omega}_k^{-2}, \tag{9}$$

$$\gamma^2 \equiv \langle (q_n - q_{n-1})^2\rangle = \frac{k_{\rm B}T}{Nm}\sum_p \frac{4\sin^2\left(\frac{k}{2}\right)}{\tilde{\omega}_k^2}. \tag{10}$$

One can obtain the parameters $f_{\rm c}$ and f in Eq. (4) by solving the self-consistent Eqs. (7)–(10).

One can immediately check SCPT for the harmonic system, for which $W(\delta q_n) = \frac{k_0}{2}\delta q_n^2$ and $V(q_n) = \frac{f_0}{2}q_n^2$. Here k_0 is the coupling strength and f_0 the strength of the harmonic on-site potential. According to Eq. (8),

$$W_\gamma = \frac{k_0}{2}\gamma^2, \quad V_\rho = \frac{f_0}{2}\rho^2. \tag{11}$$

It is then straightforward to obtain the phonon frequency of the harmonic system as

$$\omega_k^2 = \frac{f_0}{m} + \frac{4k_0}{m}\sin^2\Big(\frac{k}{2}\Big), \tag{12}$$

which is consistent with the eigen equation

$$\boldsymbol{F}\boldsymbol{e}_k = \omega_k^2 \boldsymbol{e}_k, \tag{13}$$

where $\boldsymbol{e}_k$ represents the eigenvector of phonon with the wave-number k, and $\boldsymbol{F}$ denotes the force matrix. For the anharmonic system, the eigen equation corresponding to the trial Hamiltonian[Eq. (4)] can be written as

$$\boldsymbol{F}_0\tilde{\boldsymbol{e}}_k = \tilde{\omega}_k^2\tilde{\boldsymbol{e}}_k. \tag{14}$$

One can then find the effective dispersion relation

$$\tilde{\omega}_k^2 = \frac{f}{m} + \frac{4f_{\rm c}}{m}\sin^2\Big(\frac{k}{2}\Big), \tag{15}$$

and

$$\tilde{\boldsymbol{e}}_k = \boldsymbol{e}_k, \tag{16}$$

which indicates that, for one-dimensional homogeneous anharmonic systems as given by Eq. (2), the effective dispersion relation shifts but effective phonon modes are the same in form as the corresponding bare phonon modes of the harmonic system in the framework of SCPT.

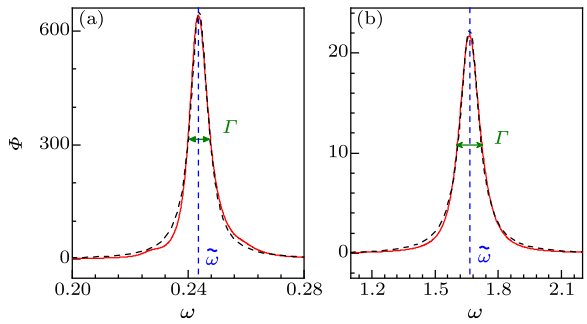

Fig. 1. The spectral energy density (red line) and the corresponding Lorentzian fitting (black dashed line) for (a) $k = \frac{\pi}{32}$ of the FPU-β lattice and (b) $k = \frac{\pi}{2}$ of the ϕ^4 lattice. The blue dashed line denotes the peak position $\tilde{\omega}'$, which agrees well with the renormalized phonon frequency $\tilde{\omega}$ [Eq. (15)]. The green dashed line indicates the width of half maximum Γ. Here $\beta = 1$ for the FPU-β lattice and $\lambda = 1$ for the ϕ^4 lattice. For both the cases, the temperature $T = 0.5$.

In order to obtain the phonon relaxation time, we perform the spectral phonon analysis using the molecular dynamics simulation. In this formulism, the canonical momentum is calculated as

$$P_k(t) = \sum_{n=1}^{N} p_n(t)\tilde{e}_k^n, \tag{17}$$

where $\tilde{e}_k^n$ represents nth component of the effective eigenvector of phonon with the wave-number k. The power spectrum of canonical momentum P_k, namely, the spectral energy density, is then evaluated as

$$\Phi_k(\omega) = \left| \int_{-\infty}^{\infty} P_k(t) e^{-i\omega t} dt \right|^2, \tag{18}$$

which can be fitted with the Lorentzian function (see Refs. [31,32] for details),

$$\Phi_k(\omega) = \frac{A_k}{(\omega - \tilde{\omega}_k')^2 + \Gamma_k^2}, \tag{19}$$

where A_k, $\tilde{\omega}_k'$ and Γ_k are fitting parameters. Here the peak position $\tilde{\omega}_k'$ is the fitting effective phonon frequency for the kth mode, and width at half maximum Γ_k can determine the phonon relaxation time

$$\tau_k = \frac{1}{2\Gamma_k}. \tag{20}$$

The mean free path is defined by

$$l_k = v_k \tau_k, \tag{21}$$

where v_k is the phonon group velocity.

We apply our numerical methods to two typical one-dimensional nonlinear lattices, namely, the FPU-β lattice and the ϕ^4 lattice. The spectral energy density $\Phi_k(\omega)$ defined in Eq. (18) can be evaluated numerically using the fast Fourier transformation (FFT). In our calculations, the time series' length of the instantaneous momentum $P_n(t)$ is 2^{17} and the sample interval $\Delta = 50$ h with the time step $h = 0.01$ for the integration. The spectral energy density $\Phi_k(\omega)$ is averaged over 2000 realizations to reduce numerical fluctuations in the sense of the microcanonical ensemble. Note that the spectral energy density $\Phi_k(\omega)$ could also be evaluated as the Fourier transformation of autocorrelation function of canonical momentum, according to the Wiener-Khinchin theorem. The equation of motion derived from the Hamiltonian at Eq. (2) is integrated by the $SABA_2C$ symplectic algorithm. The system size $N = 512$ and the fixed boundary condition is used. In our calculations, the Boltzmann constant k_B, the mass m and harmonic force constant are set as units, namely, $k_B = 1$, $m = 1$ and $k_0 = 1$. For the sake of clarification, we keep k_B, m and k_0 in all the following formulas.

Note that the spectral energy density Φ_k will be a delta function if the harmonic lattice is concerned, for which the peak position $\tilde{\omega}_k' = \omega_k$ and the width $\Gamma_k = 0$ corresponding to infinite phonon relaxation time. In the presence of anharmonicity, the peak position is shifted and the width is broadened. As seen from Fig. 1, the spectral energy density fits well with the Lorentzian function for both the FPU-β lattice and the ϕ^4 lattice. The peak position $\tilde{\omega}_k'$ as well as the width at half maximum Γ_k can then be determined. One can see that the renormalized phonon frequency $\tilde{\omega}_k$, theoretically derived by SCPT (see details in what follows), agrees well with the peak position of Lorentzian profile $\tilde{\omega}_k'$ for both the models. Therefore, the phonon group velocity at Eq. (21) can be determined by

$$v_k = \frac{\partial \tilde{\omega}_k}{\partial k}. \tag{22}$$

The FPU-β lattice, originally proposed to study the problem of energy equipartition,[34] has the on-site potential $V(x) = 0$ and the interaction potential

$$W(\delta q_n) = \frac{k_0}{2}\delta q_n^2 + \frac{\beta}{4}\delta q_n^4. \tag{23}$$

This model has been given evidence of superdiffusive heat transport.[4,5] In terms of Eq. (8), one can easily obtain the average interaction potential

$$W_\gamma = \frac{k_0}{2}\gamma^2 + \frac{3}{4}\beta\gamma^4. \tag{24}$$

Noting that sinusoidal terms in the summation of Eq. (10) are canceled, one can explicitly get the two-point correlation function

$$\gamma^2 = \frac{-k_0 + \sqrt{k_0^2 + 12\beta k_B T}}{6\beta} \tag{25}$$

and the effective force constant

$$f_c = k_0 + 3\beta\gamma^2 = \frac{k_0 + \sqrt{k_0^2 + 12\beta k_B T}}{2}. \tag{26}$$

The renormalized phonon frequency $\tilde{\omega}_k$ can be obtained by Eq. (15). As shown in Fig. 2(a), the dispersion relation obtained from the Lorentzian fitting agrees well with the prediction by SCPT, even in the large wave-number regime. Interestingly, Fig. 3(a) shows that the phonon relaxation time diverges in a power law $\tau_k \sim k^{-\mu}$ for small k at different temperatures. The exponent $\mu \approx 1.7$, which agrees with the result in the previous studies,[28,29] is close to $\mu = -5/3$ predicted by the Peierls–Boltzmann theory in the weak anharmonicity regime.[35–37] The phonon mean free path displays the divergent behavior with the same exponent [Fig. 4(a)] since v_k here can be regarded as a constant in the low-frequency limit.

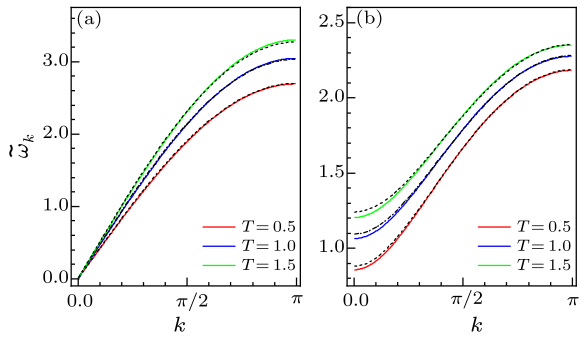

Fig. 2. The dispersion relation for (a) the FPU-β lattice and (b) the ϕ^4 lattice. The red, blue and green curves denote the dispersion relations obtained by the Lorentzian fitting at temperatures $T = 0.5$, 1.0 and 1.5, respectively. The dashed black curves are obtained from SCPT (see text for details). Here $\beta = 1$ for the FPU-β lattice, $\lambda = 1$ for the ϕ^4 lattice.

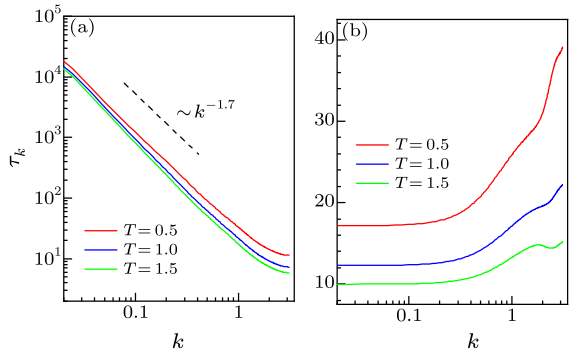

Fig. 3. The phonon relaxation time τ_k as a function of the wave number k for (a) the FPU-β lattice and (b) the ϕ^4 lattice. The red, blue and green curves denote the phonon relaxation time obtained by the Lorentzian fitting at temperatures $T = 0.5$, 1.0 and 1.5, respectively. The dashed black line in (a) is drawn as a line of reference for the power-law behavior $\tau_k \sim k^{-1.7}$. Parameters are given as the same as those for Fig. 2.

Heat conductivity κ can be calculated by the Debye formula

$$\kappa = \sum_k c_k v_k^2 \tau_k = \sum_k c_k v_k l_k, \tag{27}$$

where c_k denotes heat capacity of wave number k, which can also be seen as a constant.[15] In small-k limit, the phonon mean free path is longer than the system size because l_k approaches to divergence as k decreases, leading to anomalous heat conduction with thermal conductivity $\kappa \sim N^\alpha$, shown in one-dimensional momentum-conserving systems.[2,7] The relation between the exponents α and μ is given by $\alpha = 1 - \frac{1}{\mu}$.[3,35] For the FPU-$\beta$ lattice, the exponent $\alpha \approx 0.41$ for $\mu \approx 1.7$, which agrees with the previous studies.[4,5]

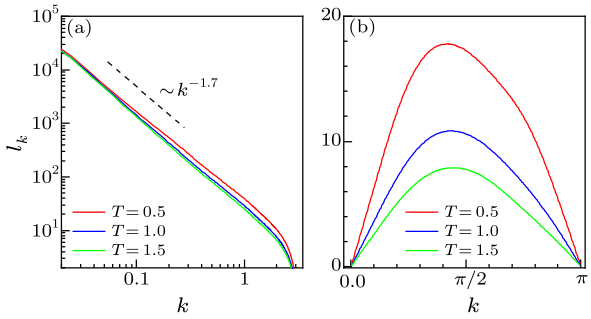

Fig. 4. The phonon mean free path l_k as a function of the wave number k for (a) the FPU-β lattice and (b) the ϕ^4 lattice. The red, blue and green curves denote the phonon mean free path obtained by definition $l_k = v_k \tau_k$ at temperatures $T = 0.5$, 1.0 and 1.5, respectively. The dashed black line in (a) is drawn as a line of reference for the power-law behavior $l_k \sim k^{-1.7}$. Parameters are given as the same as those for Fig. 2.

The ϕ^4 lattice is a typical model of non-conserved total momentum and exhibits normal heat conduction.[9–11] Its Hamiltonian consists of not only a harmonic interaction potential

$$W(\delta q_n) = \frac{k_0}{2} \delta q_n^2, \tag{28}$$

but also a nonlinear on-site potential

$$V(q_n) = \frac{\lambda}{4} q_n^4. \tag{29}$$

In terms of Eq. (8), W_γ and V_ρ are given by

$$W_\gamma = \frac{k_0}{2}\gamma^2, \qquad V_\rho = \frac{3}{4}\lambda\rho^4. \tag{30}$$

The summation of Eq. (9) can be replaced by an integral as the system size is large, which yields

$$\rho^2 = \frac{k_B T}{\sqrt{3\lambda\rho^2(3\lambda\rho^2 + 4k_0)}}. \tag{31}$$

Then the effective force constant f in Eq. (4) is given by the following self-consistent equation

$$f = 3\lambda\rho^2 = \frac{3\lambda k_B T}{\sqrt{f(f + 4k_0)}}, \tag{32}$$

which can be solved numerically. Meanwhile, one can easily find that $f_c = k_0$. The renormalized phonon frequency can also be given by Eq. (15).

As shown in Fig. 2(b), our numerical results of the dispersion relation obtained from the Lorentzian fitting agree well with the prediction of SCPT. Only very slight deviations occur in the low-frequency regime. The phonon relaxation time can be regarded as a constant in the long-wavelength limit [Fig. 3(b)]. As for the mean free path, as shown in Fig. 4(b), long-wavelength phonons exhibit finite values, which confirms the validity of normal heat conduction for the ϕ^4 lattice.[9–11]

In summary, we have studied the wave-number-dependent phonon frequency, relaxation time and mean free path for one-dimensional FPU-β and ϕ^4 lattices. Fitting the spectral energy density, i.e., the magnitude of Fourier transform of canonical momentum, by Lorentzian function, the k-dependent oscillation frequency $\tilde{\omega}'_k$ and phonon relaxation time τ_k can be obtained. The canonical momentum can be calculated by the canonical transformation of momentum of particles with the invariable phonon modes in the framework of SCPT. It is shown that the dispersion relation obtained from the Lorentzian fitting agrees well with the prediction of SCPT for both the models. For the FPU-β lattice, the mean free path exhibits divergent power-law behavior l_k(also τ_k) $\sim k^{-1.7}$ in long-wavelength limit. The exponent closely matches 5/3 predicted by the Peierls–Boltzmann theory. As for the ϕ^4 lattice, neither phonon relaxation time nor mean free path diverges, which indicates normal heat conduction. The method may be extended to study various systems, such as diatomic,[29,30] defect,[38] interfacial[39,40] and disordered systems.

The numerical simulations were performed on TianHe-1 (A) at National Supercomputer Center in Tianjin.

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