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Nianbei Li

Junjie Liu

Changqin Wu

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PAPER

Temperature and frequency dependent mean free paths of renormalized phonons in nonlinear lattices

Nianbei Li[1], **Junjie Liu**[2], **Changqin Wu**[3] and **Baowen Li**[4]

[1] Institute of Systems Science and Department of Physics, College of Information Science and Engineering, Huaqiao University, Xiamen 361021, People's Republic of China
[2] Singapore-MIT Alliance for Research and Technology Center, 1 CREATE Way, Singapore 138602, Singapore
[3] State Key Laboratory of Surface Physics and Department of Physics, Fudan University, Shanghai 200433, People's Republic of China
[4] Department of Mechanical Engineering, University of Colorado Boulder, CO 80309, United States of America

E-mail: nbli@hqu.edu.cn

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Abstract

Unraveling general properties of renormalized phonons are of fundamental relevance to the heat transport in the regime of strong nonlinearity. In this work, we directly study the temperature and frequency dependent mean free path (MFP) of renormalized phonons with the newly developed numerical tuning fork method. The typical 1D nonlinear lattices such as Fermi–Pasta–Ulam β lattice and ϕ^4 lattice are investigated in detail. Interestingly, it is found that the MFPs are inversely proportional to the frequencies of renormalized phonons rather than the square of phonon frequencies predicted by existing phonon scattering theory.

1. Introduction

Phonon is a basic concept in solid state physics describing the collective motions of lattice vibrations. The phonon description is rigorously precise only for Harmonic lattices. For nonlinear lattices especially when nonlinearity cannot be treated as a small perturbation, the concept of renormalized phonons emerges and theoretical efforts have been devoted to describe these novel collective motions. The renormalized phonons are discovered by different groups independently in various research areas ranging from lattice vibrations [1, 2], heat conduction [3], field thermalization [4] and nonlinear waves [5, 6]. The theoretical predictions of the dispersion relations of renormalized phonons from these different approaches are found to be slightly different [7, 8]. This puzzle is solved by a recent variational approach which unifies the renormalized phonon theory by applying suitable approximations in a systematical way [9, 10]. The existence of renormalized phonons in general 1D nonlinear lattices has been verified numerically in computer simulations [1, 2, 5–7, 11, 12]. The role of renormalized phonons as the heat energy carriers has also been proposed and testified by numerical simulations [11–16].

In the regime of strong nonlinearity, the validity of conventional perturbative phonon transport theories is questionable. The phenomenological effective phonon theory [7, 11–13, 16] is developed within the framework of renormalized phonons dedicated to the explanations of temperature dependence of thermal conductivities for nonlinear lattices. This theory can predict the actual exponents of the power-law dependence of thermal conductivities as the function of temperature for typical 1D nonlinear lattices. For example, the effective phonon theory predicts the temperature dependent thermal conductivities $\kappa(T)$ of 1D Fermi–Pasta–Ulam β (FPU-β) lattice are inversely proportional to temperature as $\kappa(T) \propto T^{-1}$ at low temperature region and proportional to the quartic root of temperature as $\kappa(T) \propto T^{1/4}$ at high temperature region [13, 16]. It also predicts the temperature dependence is $\kappa(T) \propto T^{1/2-1/n}$ for H_n lattices [16]. For lattices with on-site potentials, the effective phonon theory predicts for 1D ϕ^4 lattice the temperature dependence is $\kappa(T) \propto T^{-4/3}$ [11]. For general nonlinear Klein–Gordon lattices where ϕ^4 lattice is a special example of $n = 4$, this theory predicts the general temperature dependence as $\kappa(T) \propto T^{4(2-n)/(n+2)}$ [12]. Extensive numerical simulations have verified these predictions quantitatively and consistently for FPU-β lattice [13, 16, 17], H_n lattices with $n = 3, 4, 5$ [16],

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nonlinear Klein–Gordon lattices with $n = 1.25, 1.5, 1.75, 2.5, 3, 3.5, 4$ [11, 12, 18]. It should be emphasized that these theoretical predictions are derived from effective phonon theory without any fitting parameter.

In determining the thermal conductivities, the most important information is the temperature and frequency dependence of mean free paths (MFPs) of energy carriers. In the framework of the effective phonon theory, the dependence of MFPs on the frequency is assumed to follow a phenomenological relation [11, 16]:

$$\hat{l}_k \propto \frac{\hat{v}_k}{\varepsilon \hat{\omega}_k}, \tag{1}$$

where $\hat{v}_k = \partial\hat{\omega}_k/\partial k$ is the group velocity of renormalized phonons. The dimensionless nonlinearity strength ε is defined as the ratio of ensemble averaged nonlinear potential energy E_n and total potential energy $E_t = E_l + E_n$ to be $\varepsilon = E_n/(E_l + E_n)$ where E_l is the ensemble averaged linear potential energy. However, the validity of the conjecture (see equation (1)) in the whole parameter regime remains an open question to us.

In this work, we will directly test the validity of this conjecture by using the newly developed tuning fork method which enables us to calculate the MFPs for every phonon mode in a direct way [19]. The temperature and frequency dependent MFPs of renormalized phonons will be calculated and compared with the conjecture for typical 1D FPU-β, H_4 and ϕ^4 lattices. The good agreement between numerical results and the conjecture indicates that the MFP of renormalized phonons is indeed inversely proportional to the renormalized phonon's frequency and the nonlinearity strength, which is beyond the scope of any conventional perturbative phonon transport theory.

2. The MFPs of renormalized phonons: conjecture and numerical verifications

We consider the 1D nonlinear lattices of N atoms with the general Hamiltonian

$$H = \sum_i \left[\frac{p_i^2}{2} + V(q_{i+1} - q_i) + U(q_i) \right], \tag{2}$$

where p_i and q_i denote the momentum and displacement of the ith atom, respectively. For simplicity, periodic boundary conditions of $q_i = q_{N+i}$ and dimensionless units have been used. The $V(x)$ represents the inter-atom potential energy where only nearest neighbor interaction has been considered and the $U(x)$ is the on-site potential energy. In this work, three typical nonlinear lattices, namely, the 1D FPU-β, H_4 and ϕ^4 lattices are the focus of the investigation. The combination of $V(x) = x^2/2 + x^4/4$ and $U(x) = 0$ describes the FPU-β lattice while the combination of $V(x) = x^4/4$ and $U(x) = 0$ denotes the H_4 lattice. For the ϕ^4 lattice, the potential energy takes the form of $V(x) = x^2/2$ and $U(x) = x^4/4$.

The renormalized phonon frequency $\hat{\omega}_k$ can be expressed in a general form as $\hat{\omega}_k = \sqrt{\alpha\omega_k^2 + \gamma}$ where $\omega_k = 2\sin\frac{k}{2}$, $-\pi < k \leqslant \pi$ is the familiar phonon frequency of the Harmonic lattice [16]. The renormalization coefficient α is determined only by the inter-atom potential energy $V(x)$ while the coefficient γ depends only on the on-site potential energy $U(x)$. For Harmonic lattice, $\gamma = 0$ and $\alpha = 1$ ensure that $\hat{\omega}_k = \omega_k$ is recovered. The nonlinearity strength ε defined as $\varepsilon = E_n/(E_l + E_n)$ is dimensionless. For simplicity, we only consider lattices with symmetrical hard potential with which the ε will not be ill-defined to be negative value [20]. Since in asymmetrical lattices, the situation is more complicated as the energy diffusion is ballistic while the heat conduction is anomalous instead of ballistic [21]. For the Harmonic lattice, the nonlinearity strength ε equals zero which should be expected since Harmonic lattice is a linear system. The existence of renormalized phonons in these lattices have been verified by numerical simulations [1, 2, 5–7, 9–12].

The newly developed tuning fork method is able to calculate the renormalized phonon dispersion relations as well as the MFPs for these anharmonic vibrations directly [19]. For a nonlinear lattice at thermal equilibrium, one can first determine the correlation function between the momenta of mth and nth atoms:

$$\Phi_{mn}(t) = \langle p_m(t) p_n(0) \rangle, \tag{3}$$

where it only depends on the relative positions of $m - n$ here. A Fourier transform of this correlation function gives rise to the susceptibility $\chi(\omega)$:

$$\chi(\omega) = \int_0^\infty \Phi_{mn}(t) e^{-i\omega t} dt = |\chi_{|m-n|}(\omega)| e^{i\phi_{|m-n|}}. \tag{4}$$

If the amplitude $|\chi_{|m-n|}(\omega)|$ decays exponentially as

$$|\chi_{|m-n|}(\omega)| \propto e^{-\frac{|m-n|}{l(\omega)}}, \tag{5}$$

where $l(\omega)$ is a frequency dependent characteristic length. This characteristic length $l(\omega)$ can be explained as the MFP for the nonlinear vibration with frequency ω. Most recently, a resonance phonon approach method has

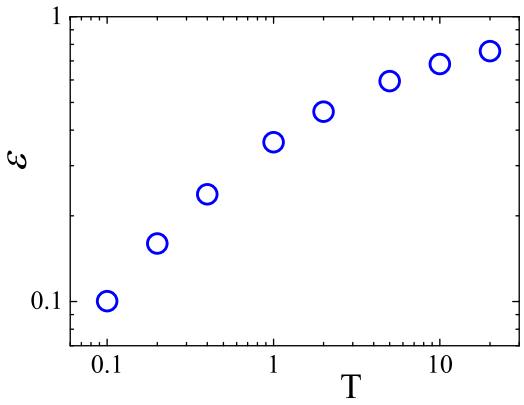

Figure 1. The dimensionless nonlinearity strength ε as the function of temperature for the FPU-β lattice. The ε increases linearly with temperature as $\varepsilon \propto T$ in the low temperature regime and saturates to maximum value of 1 as $\varepsilon \to 1$ in the high temperature limit. These values are evaluated from equation (7) and these temperature points will be used to calculate the MFPs of renormalized phonons as in figure 2.

been proposed which is also able to calculate the renormalized phonon frequencies and relaxation times in nonlinear lattices [20, 22].

2.1. The FPU-β lattice

For lattices without on-site potential, the renormalization coefficient γ is zero. The expression of renormalized phonon frequency and its group velocity can be simplified as

$$\hat{\omega}_k = \sqrt{\alpha}\,\omega_k, \quad \hat{v}_k = \sqrt{\alpha}\,v_k, \tag{6}$$

where $v_k = \cos\frac{k}{2}$ is the phonon group velocity for the Harmonic lattice. Here the renormalization coefficient α can be analytically obtained as $\alpha = 1 + \int_0^\infty x^4 \mathrm{e}^{-(x^2/2+x^4/4)/T}\mathrm{d}x \Big/ \int_0^\infty x^2 \mathrm{e}^{-(x^2/2+x^4/4)/T}\mathrm{d}x$ which is only temperature or equivalently nonlinearity dependent [15].

For the FPU-β lattice, the nonlinearity strength ε can be expressed as $\varepsilon = \langle x^4/4\rangle / (\langle x^2/2\rangle + \langle x^4/4\rangle)$ where $\langle\cdot\rangle$ means ensemble average. It can further be analytically resolved as

$$\varepsilon = \frac{1}{2\dfrac{\int_0^\infty x^2 \mathrm{e}^{-(x^2/2+x^4/4)/T}\mathrm{d}x}{\int_0^\infty x^4 \mathrm{e}^{-(x^2/2+x^4/4)/T}\mathrm{d}x} + 1}. \tag{7}$$

This temperature dependence of ε for the 1D FPU-β lattice is plotted in figure 1. At low temperature limit, ε increases with temperature linearly as $\varepsilon \propto T$, while ε approaches to maximum value 1 at the high temperature limit. The temperatures of these data points will be the temperatures at which the MFPs of renormalized phonons are calculated. It can be seen that the range of ε values considered here is about one order of magnitude.

According to the conjecture of equation (1), the MFPs of renormalized phonons for the FPU-β lattice is $\hat{l}_k \propto v_k / (\varepsilon\omega_k)$ since the coefficient α is canceled for $\hat{v}_k$ and $\hat{\omega}_k$. If we only consider the temperature dependence of MFPs $\hat{l}_k$, we have

$$\hat{l}_k \propto \frac{1}{\varepsilon}, \tag{8}$$

which is a global effect. It says that for the temperature effect, the MFP of every renormalized phonon is inversely proportional to the nonlinearity strength ε.

In figure 2(a), the MFPs of renormalized phonons are plotted for different temperatures from $T = 0.1$ to $T = 20$. The lower the temperature, the longer the MFPs due to its small nonlinearity strength ε as indicated from equation (8). In order to quantitatively verify the dependence of equation (8), we plot the rescaled MFPs of $\hat{l}_k\varepsilon$ as the function of temperature in figure 2(b). It can be seen that all the rescaled MFPs $\hat{l}_k\varepsilon$ at different temperatures collapse into a single curve, which is a clear verification of the equation (8).

It should be mentioned that the numerically calculated MFPs diverges as $\hat{l}_k \propto k^{-1.70}$ at low frequency limit as $k \to 0$ as observed in [19], which finally gives rise to a divergent thermal conductivity as $\kappa \propto N^{0.41}$ known as the anomalous heat conduction [23–56]. Since the raw frequency ω_k is proportional to wave vector k as $\omega_k \propto k$ in the low frequency limit, the calculated MFPs thus follows the behavior of $\hat{l}_k \propto \omega_k^{-1.70}$. However, the MFPs should depend on the renormalized frequency as $\hat{l}_k \propto \hat{\omega}_k^{-1}$ as seen from figure 2(b). As $\hat{\omega}_k \propto \omega_k$, the extra $\omega_k^{-0.70}$ dependence cannot come from the renormalized phonon frequency $\hat{\omega}_k$ which will bring additional temperature

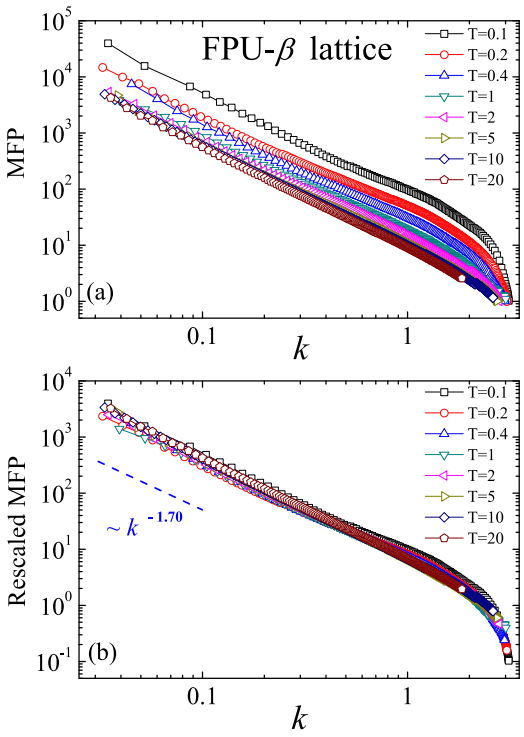

Figure 2. (a) MFP $\hat{l}_k$ of renormalized phonons as the function of k for FPU-β lattice at different temperatures. The larger the temperature, the larger the nonlinearity and the shorter the MFPs as revealed by equation (8). (b) Rescaled MFP ($\hat{l}_k \cdot \varepsilon$) as the function of k for FPU-β lattice at different temperatures. The dashed line of $k^{-1.70}$ is the guide for the eye. The calculations are performed on lattices with size $N = 1024$ for $T < 5$ and $N = 2048$ for $T \geqslant 5$.

dependence to MFPs $\hat{l}_k$. This property is unique for momentum-conserving lattices and the reason for this is still an open issue.

Since the phonon group velocities $v_k = \sqrt{\alpha}\cos k/2$ approach to zero as $k \to \pi$, their MFP will approach to zero as well. However, the phonon MFP $\hat{l}_k$ is meaningless if it is comparable to the lattice constant which is set as unit here. The higher the temperature, the shorter the phonon MFP. In numerical simulations, we only calculate the MFP for $\hat{l}_k > 1$ which occurs for $k < \pi$ as can be seen from figure 2(a).

2.2. The H_4 lattice

As a momentum-conserving lattice, the H_4 lattice has the same expression for renormalized phonon frequency and its group velocity as in equation (6). But the renormalization coefficient α has a different expression as $\alpha = 2\Gamma(5/4)/\Gamma(3/4)T^{1/2}$ [15].

The H_4 lattice is the high temperature limit of the FPU-β lattice and is a pure nonlinear lattice untreatable for any perturbative phonon transport theory. There is only pure nonlinear term in the inter-atom potential energy of $V(x) = x^4/4$. By definition, the nonlinearity strength ε achieves the maximum value as $\varepsilon = 1$ for all temperatures. Same as the case for the FPU-β lattice, the MFPs $\hat{l}_k$ of H_4 lattice shares the same temperature dependence as $\hat{l}_k \propto \varepsilon^{-1}$. However, since $\varepsilon = 1$ at any temperature for the special H_4 lattice, the MFPs

$$\hat{l}_k \propto \text{constant} \tag{9}$$

which does not depend on temperature at all. This behavior can also be obtained from pure mathematical scaling analysis, while the physical consistence between H_4 and FPU-β lattice cannot be recovered.

In figure 3, the MFPs of renormalized phonons in the H_4 lattice are plotted for temperatures from $T = 0.1$ to $T = 20$. All the MFPs collapse into a single curve showing no sign of temperature dependence. The remarkable feature of equation (9) is clearly verified.

2.3. The ϕ^4 lattice

The ϕ^4 lattice has an on-site potential and exhibits normal heat conduction behavior [18, 57, 58]. According to definition, the renormalization coefficient α can be obtained as $\alpha = 1$. The expressions of renormalized phonon frequency and its group velocity are

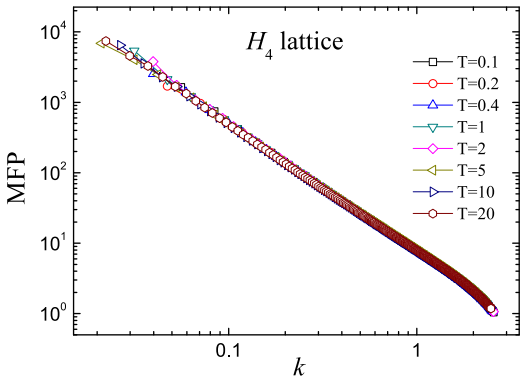

Figure 3. MFPs $\hat{l}_k$ of renormalized phonons as the function of k for H_4 lattice at different temperatures. The MFPs are found to be totally independent of temperatures which is exactly what the conjecture predicts. The calculations are performed on lattices with size $N = 1024$ for $T < 5$ and $N = 2048$ for $T \geqslant 5$.

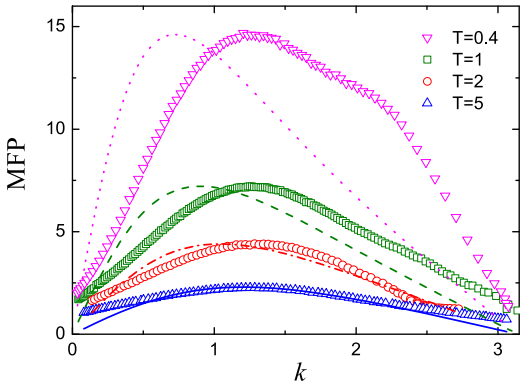

Figure 4. MFPs $\hat{l}_k$ of renormalized phonons as the function of k for ϕ^4 lattice at different temperatures at $T = 0.4,\ 1,\ 2$ and 5. The hollow symbols are the numerical data of MFPs. The dotted, dashed, dashed–dotted and solid lines are the curves of $\hat{l}_k = A(T)\frac{\sin k}{\omega_k^2+\gamma}$ where $A(T)$ is the fitting parameter for $T = 0.4,\ 1,\ 2$ and 5, respectively. Here the fitting $A(T)$ is $A(T = 0.4) = 25.8$, $A(T = 1) = 18.3$, $A(T = 2) = 15.0$ and $A(T = 5) = 12.0$. The calculations are performed on lattices with size $N = 1024$.

$$\hat{\omega}_k = \sqrt{\omega_k^2 + \gamma},\ \ \hat{\nu}_k = \frac{\sin k}{\sqrt{\omega_k^2 + \gamma}}, \tag{10}$$

where the renormalization coefficient $\gamma = \langle x^4\rangle / \langle x^2\rangle$ can be derived from a classical field approach as $\gamma \approx 1.230\ T^{2/3}$ [4].

According to equation (1), the MFPs $\hat{l}_k$ of renormalized phonons of the ϕ^4 lattice should follow a dependence on nonlinearity strength ε and wave vector k as

$$\hat{l}_k \propto \frac{1}{\varepsilon}\frac{\sin k}{\omega_k^2 + \gamma}, \tag{11}$$

where $\gamma \approx 1.230 \cdot T^{2/3}$ is valid for temperatures not high enough [11]. Since the heat conduction is normal for the ϕ^4 lattice, we are able to test both the frequency and temperature dependence of MFPs here.

We first test the k or frequency dependence of MFPs $\hat{l}_k$ as in equation (11). By introducing a temperature dependent but k independent fitting parameter $A(T) \propto \varepsilon^{-1}$, it is easy to have $\hat{l}_k = A(T)\frac{\sin k}{\omega_k^2+\gamma}$. In figure 4, the numerical calculated MFPs are plotted as hollow symbols at four different temperatures $T = 0.4,\ 1,\ 2$ and 5. These data are compared with $\hat{l}_k = A(T)\frac{\sin k}{\omega_k^2+\gamma}$ with $A(T = 0.4) = 25.8$, $A(T = 1) = 18.3$, $A(T = 2) = 15.0$ and $A(T = 5) = 12.0$ plotted as the dotted, dashed, dashed–dotted and solid lines in figure 4, respectively. It can be seen that the k dependence of numerically calculated MFPs can be qualitatively described by the conjectured k dependence of $\hat{l}_k$ of equation (11). For $T = 0.4,\ 1$ and 2, the peak positions of conjectured curves are left-shifted compared to the numerical data. At $T = 5$, the peak positions are consistent between numerical data and the conjecture. The MFPs around $k \to 0$ are underestimated by the conjecture and this effect might be understood by noticing that the numerical calculated renormalized phonon frequencies $\hat{\omega}_k$ are smaller than what are predicted by the renormalized phonon theory [19].

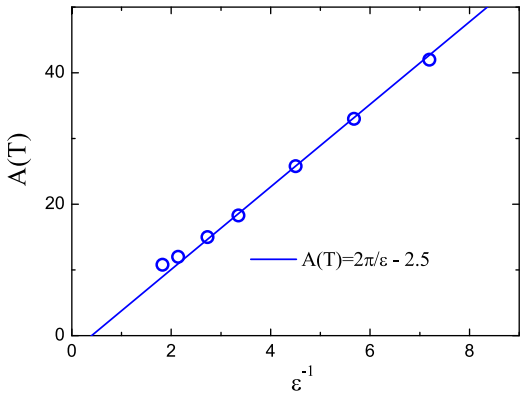

Figure 5. The fitting parameter $A(T)$ as the function of inverse nonlinearity strength ε^{-1} for ϕ^4 lattice. The $A(T)$ is fitted from the numerical calculated MFPs with the fitting formula of $\hat{l}_k = A(T)\frac{\sin k}{\omega_k^2+\gamma}$. The ε is calculated in thermal equilibrium for a lattice with $N = 100$. The solid line of $A(T) = 2\pi/\varepsilon - 2.5$ is the guide for the eye. The good agreement indicates the phonon MFPs in ϕ^4 lattice is inversely proportional to the nonlinearity strength as in equation (11).

The MFP $\hat{l}_k$ is also conjectured to be inversely proportional to the nonlinearity strength ε for the ϕ^4 lattice in equation (11). The nonlinearity strength ε can be expressed as $\varepsilon = \langle q_i^4/4\rangle/(\langle(q_{i+1} - q_i)^2/2\rangle + \langle q_i^4/4\rangle)$ by noticing the ensemble average is independent of atom index i. Unfortunately the ε cannot be analytically calculated here. We can only numerically calculate the nonlinearity strength ε at different temperatures at thermal equilibrium. In figure 5, the fitting prefactors $A(T)$ are plotted as the function of ε^{-1} for temperatures ranging from $T = 0.1$ to $T = 10$. For higher temperatures, the numerical simulation is hard to achieve thermal equilibrium as the ϕ^4 lattice approaches to the anti-continuous limit with only N separate nonlinear oscillators located at each site. The linear dependence between $A(T)$ and ε^{-1} verifies the dependence of nonlinearity strength for the MFP $\hat{l}_k$ conjectured as in equation (11) for the ϕ^4 lattice. Most interestingly, the slope of 2π of this dependence suggests the renormalized phonon's MFP $\hat{l}_k$ of the ϕ^4 lattice can be written as

$$\hat{l}_k = \hat{v}_k \frac{1}{\epsilon}\frac{2\pi}{\hat{\omega}_k}, \tag{12}$$

where $2\pi/\hat{\omega}_k$ is nothing but one period of renormalized phonon with mode k.

3. Conclusions

In summary, we have quantitatively investigated the temperature and frequency dependence of MFPs of renormalized phonons in 1D nonlinear lattice with the newly developed tuning fork method. The conjecture made in the effective phonon theory is verified suggesting the MFPs should be inversely linear proportional to the renormalized frequency $\hat{\omega}_k$ and nonlinearity strength ε. This is different from the analysis of Umklapp phonon scattering theory where a MFP $\propto\omega^{-2}$ dependence is predicted [59]. But our work is consistent with the recent results numerically obtained for carbon nanotubes where the dependence of MFP $\propto\omega^{-1}$ is claimed [60]. The current work reinforces the role of renormalized phonons during the heat transport process in strong nonlinear materials where the nonlinear effect cannot be ignored.

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ORCID iDs

Nianbei Li https://orcid.org/0000-0002-9151-7675

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