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Monitoring anharmonic phonon transport across interfaces in one-dimensional lattice chains

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Modeling thermal transport through interfaces has been one of the most challenging problems in nanoscale heat transfer. Although continuous theoretical efforts have been made, there has been no consensus on how to rigorously incorporate temperature effect and anharmonicity. In this paper, we adopt the self-consistent anharmonic phonon concept for nonlinear lattices to investigate phonon propagation within the materials as well as across interfaces based on equilibrium molecular dynamics simulations. Based on linear response theory, we propose an efficient method to calculate the frequency-dependent transmission coefficient in a nonlinear lattice. The transmission spectrum is extracted directly from velocity correlations, which naturally includes anharmonic effects. Phonon renormalization at finite temperature can also be easily handled using the proposed method. Our results are consistent with the atomistic Green’s function method at the limit of weak anharmonicity. For nonlinear lattices under high temperatures, the anharmonicity is found to increase the cutoff frequency of the transmission coefficient due to phonon renormalization. Further analysis shows that the anharmonicity also promotes interfacial thermal conductance by causing the redistribution of the spectral flux of the excited vibrational waves during their propagation.

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I. INTRODUCTION

The knowledge of phonon transport across interfaces between dissimilar materials is fundamental to addressing heat dissipation in nanodevices [1] and to designing good thermoelectric and thermal insulating materials [2,3]. When phonons encounter interfaces, they experience reflection, transmission, and mode conversion, leading to local temperature jumps at the interfaces and thus additional thermal resistance. While the interfacial thermal resistance can be measured through experiments and used to roughly estimate the effective thermal conductivity of nanostructures in which multiple interfaces exist [4], it is highly desirable to have the knowledge of mode-dependent or frequency-dependent phonon transmission across interfaces as this information could be employed as inputs to precisely determine the thermal conductivity of the nanostructures through mesoscopic phonon modeling tools, such as Boltzmann transport equation based methods [5–7].

In the past few decades, continuous efforts have been devoted to predicting phonon transmission across interfaces and several theoretical methods had been proposed, including mismatch models: the acoustic mismatch model (AMM) [8,9] and diffuse mismatch model (DMM) [9], and atomistic simulations, such as the molecular dynamics based wave packet method [10], linear lattice dynamics [11], and the atomistic Green’s function (AGF) approach [12,13]. The AMM and DMM models assume that the phonon transmission is determined by the acoustic impedances of the materials and that phonon scatterings at interfaces are fully diffusive, respectively. Despite the simple expressions of the mismatch models, which only include the phonon spectra of the bulk materials on both sides, these empirical models cannot take the detailed atomic structure near the interface into account and are found to fail to accurately predict the interfacial thermal conductance (the inverse of interfacial thermal resistance). Unlike the mismatch models, the phonon transmission across realistic interfacial regions could be computed using the atomistic simulations, which are based on the linear approximation of interatomic forces (harmonic approximation) and cannot directly capture anharmonic effects on phonon scatterings. These atomistic approaches have been successfully applied to a wide range of interfaces and used to study the effects of interface roughness [14], species mixing [15], local strain field [16], etc., on phonon transmission and interfacial thermal conductance, but there is yet no consensus on how temperature and lattice anharmonicity affect phonon transport across interfaces.

The experimental evidence of the anharmonic effects on phonon transmission across interfaces was first given by Lyee and Cahill [17], where the conductance of Pb or Bi on hydrogen-terminated diamond measured by the time-domain thermoreflectance (TDTR) approach was found to increase linearly with increasing temperature. The linear dependence was also confirmed by nonequilibrium molecular dynamics (NEMD) by Stevens et al. [18]. The temperature-dependent interfacial thermal conductance could be explained by neither the mismatch models nor the atomistic methods under the harmonic approximation, due to the neglecting of anharmonicity and temperature dependence of phonon properties. Using nonequilibrium molecular dynamics, Wu and Luo [19] and...
Le et al. [20] examined the effect of the anharmonicity both within the bulk materials and in the interfacial region on the interfacial thermal conductance. By modifying the harmonic force constants of the interatomic potential, the authors found that the temperature-dependent interfacial thermal conductance mainly originated from the anharmonicity inside the materials. Compared with interfacial thermal conductance, it is more challenging to determine the anharmonic phonon transmission. To explain the temperature effects, the DMM model was extended by Hopkins and co-workers [21,22]. In their models, the contributions to the phonon transmission due to multiple-phonon inelastic scatterings right at the interfaces add to the original expression of the DMM model. While better agreement in trends between interfacial thermal conductance calculations and experimental data was found with the improvements on the DMM model, there is no direct evidence to validate predicted phonon transmission from the calculations. Sääskilahti et al. [23] derived an expression to calculate the spectral decomposition of the thermal conductance in NEMD simulations based on the correlation function between atom displacement and atom velocity, but in this method the interatomic forces are explicitly decomposed into harmonic and anharmonic components, which requires heavy computation burden. Le et al. [20] also conducted NEMD simulations, where wavelet analysis was performed to calculate the spectral energy density in the interfacial system. Due to the nature of the wavelet transform, the frequency resolution highly depends on the time window and the information of phonon frequency is not exact. The above attempts relied on NEMD simulations, in which the imposed temperature gradients are usually much larger than the realistic value in measurements.

When the temperature is high or the anharmonicity of the lattice is strong, the phonon dispersion of the material deviates from the harmonic prediction. While phonon renormalization is believed to affect phonon transport considerably in quite a few crystalline materials, the role of temperature-induced phonon renormalization on phonon transmission has been rarely studied. Zhang et al. [24] derived an explicit analytical expression of transmission coefficient through the scattering boundary method. However, this method can only deal with the anharmonicity right at the interface, and thus the phonon renormalization within bulk materials could not be correctly treated. Later, He et al. [25] proposed a self-consistent approach to obtain an effective harmonic Hamiltonian for the anharmonic system, which is used to compute the phonon transmission under the framework of the Green’s function approach. Since the phonon scatterings are ignored, this method fails to capture the conversion among different phonon modes if the dimension of the system is larger than the phonon mean free path.

Recently, Liu et al. [26] pointed out that there is a contradiction between the concept of phonons, which are nondecaying, stable propagating waves existing in harmonic lattices, and the existence of phonon decay in real anharmonic lattices. To address the dilemma, they proposed an equilibrium molecular dynamics (EMD) based method to mimic the tuning fork experiments through which the evolution of the excited vibrational waves can be monitored. In this method, a driving force is first imposed on a certain atom to excite the phonons in a homogeneous lattice; then the decay of the excited lattice wave could be monitored by calculating the thermally averaged profile of the magnitude of atomic velocity. Using this method, mean free paths of the self-consistent anharmonic phonons can be easily obtained from the decaying distance.

In this paper, we adopt the anharmonic phonon concept for nonlinear lattices to visualize the phonon propagation within the materials as well as across interfaces based on equilibrium molecular dynamics simulations. Based on the linear response theory, a method to extract the frequency-dependent anharmonic phonon transmittance is proposed by analyzing the evolution of the anharmonic phonon waves excited by external perturbing forces. For simplicity without losing generality, we study the phonon transport across interfaces between one-dimensional Fermi-Pasta-Ulam-β (FPU-β) lattices [27,28]. Unlike AGF, this method can calculate the phonon transmission coefficient of an anharmonic lattice from velocity correlation functions directly instead of using force constants as inputs. The effects of phonon renormalization at high temperature in strongly anharmonic lattice on phonon transmission can be easily handled by the proposed method. Our simulations show that the phonon renormalization at finite temperatures enhances the phonon transmission of high-frequency phonons. In addition, the analysis on the spectral distribution of the excited phonon waves at different locations confirms that during the propagation high-frequency phonons are converted to low-frequency phonons due to the anharmonicity of the lattice. These low-frequency phonons can transmit through the interface more easily than high-frequency phonons, leading to larger interfacial conductance when the location of the heat source becomes farther away from the interface.

II. THEORETICAL METHOD

Figure 1(a) illustrates the concept of anharmonic phonons in a homogeneous anharmonic lattice [26]. We apply this method to two kinds of typical interfacial systems, single-interface systems and heterojunction systems [see Figs. 1(b) and 1(c)], to explore the role of anharmonicity in phonon transmission across the interfacial regions. In Sec. II A, we
briefly review the concept of anharmonic phonons and extend it into interfacial systems. The numerical details of molecular dynamics simulations are given in Sec. II B.

A. Triggering anharmonic phonons

When a lattice system is in thermal equilibrium, the average velocity of each atom is zero. If one applies a weak external periodic force $f_i(t)$ on an atom in the lattice, the thermally averaged velocities of other atoms become no longer zero. For a one-dimensional lattice where an external force acts on atom $i$, as shown in Fig. 1(a), the Hamiltonian function of the lattice system is expressed as

$$H_{\text{total}} = H(r, p) - f_i(t)x_i,$$

where $x_i$ is the displacement of atom $i$ away from its equilibrium position. $H(r, p)$ in Eq. (1) is the Hamiltonian of the unperturbed system, which reads [29]

$$H(r, p) = \sum_{j=1}^{n} \left[ \frac{p_j^2}{2m_j} + V(x_{j+1} - x_j) + U(x_j) \right].$$

where $p_j$ and $m_j$ are the momentum and mass of atom $j$, $V(x_{j+1} - x_j)$ is the interatomic potential, and $U(x_j)$ represents the on-site potential. According to the canonical linear response theory [30,31], the thermal averaged velocity of the $j$th atom excited by the external force acting on atom $i$ can be written as

$$\langle v_j(t) \rangle_e = \int_{-\infty}^{t} \psi_{ji}(t-t')f_k(t')dt'.$$

In Eq. (3), $\psi_{ji}$ is the correlation function between the velocities of the $j$th and $i$th atoms, which is defined by

$$\psi_{ji}(t-t') = \frac{1}{k_B T} \langle v_j(t-t')v_i(0) \rangle,$$

where $k_B$ is the Boltzmann constant, $T$ is the temperature of the system, and $\langle \cdots \rangle$ represents the ensemble average. If we decompose the external force into the components corresponding to different frequencies, that is, $f_i(t) = \sum_n f_n(t)e^{i\omega_n t}$, Eq. (3) is then converted to

$$\langle v_j(t) \rangle_e = \sum_n \left\{ \langle f_n(t) \rangle \left[ \frac{1}{k_B T} \int_{0}^{\infty} \langle v_j(\tau)v_i(0) \rangle e^{i\omega_n \tau} d\tau \right] e^{i\omega_n t} \right\}.$$

Using the so-called dynamic susceptibility,

$$\chi_{ji}(\omega) = \frac{1}{k_B T} \int_{0}^{\infty} \langle v_j(\tau)v_i(0) \rangle e^{i\omega \tau} d\tau = |\chi_{ji}(\omega)|e^{-i\phi_{ji}(\omega)},$$

the thermal averaged velocity of the $j$th atom excited by the external force on atom $i$ can be further simplified as

$$\langle v_j(t) \rangle_e = \sum_{\omega} \left\{ \langle f(\omega) \rangle |\chi_{ji}(\omega)|e^{-i\phi_{ji}(\omega)} \right\} = \sum_{\omega} \langle v_j(\omega) \rangle e^{i\omega t}.$$

In Eq. (7), $\phi_{ji}$ is the phase difference and is caused by the fact that extra time is needed for the excited waves to travel across the distance between the two atoms; $\langle v_j(\omega) \rangle = f(\omega)|\chi_{ji}(\omega)|e^{-i\omega t}$ is the thermal average velocity triggered by the driving force with a frequency of $\omega$. For the homogeneous lattice, due to the translational invariance, $\chi_{ji}$ and $\phi_{ji}$ only depend on the relative position $|j-i|$. Since the phase difference has to obey a linear relation with the distance between the two atoms, one has

$$\phi_{ji}(\omega) = -k(\omega)(j-i) + \phi_0(\omega),$$

where $k$ and $\phi_0$ are two frequency-dependent constants. The coefficient $k$ in Eq. (8) can be interpreted as the wave number. Therefore, the phonon dispersion relation of the anharmonic lattice can be determined directly by analyzing the thermal averaged velocities of atoms in different locations.

The physical meaning of $\chi_{ji}(\omega)$ has been examined through a Green’s function analysis [26]. Under the harmonic approximation, the dynamic susceptibility $\chi_{ji}(\omega)$ is correlated to the Green’s function of the lattice through

$$\chi_{ji}(\omega) = i\omega G_{ji}(\omega),$$

where $G_{ji}(\omega)$ is the component of the Green’s function matrix $G(\omega) = (\Phi - \omega^2 I)^{-1}$ with harmonic matrix $\Phi$ and the identity matrix $I$. For a driving force $f_i(t) = f_i(\omega)e^{i\omega t}$ acting on atom $i$ with a small coefficient $f_i(\omega)$, the displacement of the $j$th atom due to the excitation is written as

$$x_j = G_{ji}(\omega)f_i(\omega)e^{i\omega t} = f_i(\omega)\frac{\chi_{ji}(\omega)e^{i\omega t}}{i\omega}.$$

Thus, $A_{ji}(\omega) = f_i(\omega)|\chi_{ji}(\omega)|/\omega$ means the amplitude of the excited wave at the position of atom $j$.

In anharmonic lattices, $|\chi_{ji}(\omega)|$ decays with the length $|j-i|$. This is because the excited phonon waves lose coherence and the energy carried by the excited phonon waves remains dissipated during the propagation due to phonon scatterings. The phonon mean free path $l(\omega)$ can be therefore extracted through the decay of the susceptibility $|\chi_{ji}(\omega)|$ by

$$|\chi_{j-i}(\omega)| = B_0(\omega)\exp[|j-i|/l(\omega)],$$

where $B_0(\omega)$ is a frequency-dependent coefficient. According to Ref. [32], the energy of the degree of freedom associated with a particular frequency $\omega$ can be calculated by

$$E_j(\omega) = M_j\omega^2|A_{ji}(\omega)|^2,$$

with the atomic mass $M$. Furthermore, the energy flux $I$ at the position of the $n$th atom by triggering the atom $i$ can be written as [11,14]

$$I_{n,i} = \sum_{\omega} c_n(\omega)E_j(\omega) = \sum_{\omega} c_n(\omega)M_n[f_i(\omega)|\chi_{ni}(\omega)|]^2 = \sum_{\omega} I_{n,i}(\omega),$$

where $c_n$ is the phonon group velocity, and $I_{n,i}(\omega) = c_n(\omega)M_n[f_i(\omega)|\chi_{ni}(\omega)|]^2$ is the spectral energy flux.

After obtaining the spectral energy flux, the transmission coefficient between any pair of lattice sites $(m,n)$ can be calculated through the ratio of the spectral energy fluxes:

$$T_{m,n}(\omega) = \frac{I_{n,i}(\omega)}{I_{m,i}(\omega)}.$$
where \( i \) represents the index of the atom that the driving force is acting on. Conventionally, the denominator in the definition of the transmission coefficient is the flux due to the incident wave, but the spectrum energy flux \( I_{m,i}(\omega) \) in Eq. (14) contains the signals of both the incident and reflected waves if the atom \( m \) is before the interface. Therefore, one practical problem when using Eq. (14) to compute the transmission coefficient is to distinguish these two signals if the location of the atom is nearly ahead of the interface. In order to conveniently get the signal of the incident waves, we perform an additional simulation of a homogeneous lattice which is identical to the left part of the interfacial system, and obtain the corresponding susceptibility. The obtained susceptibility is then used to calculate the thermal average velocity, which is treated as the left part of the interfacial system, and obtain the corresponding simulation of a homogeneous lattice which is identical to the right part of the interfacial system, and obtain the corresponding simulation of a homogeneous lattice which is identical to the right part of the interfacial system.

Therefore, the frequency-dependent transmission coefficient of the incident wave in the interfacial system can be calculated through

\[
\Gamma_{m,n}(\omega) = \frac{I_{m,n}(\omega)}{I_{m,m}(\omega)} = \frac{\left| \chi_{m,n}^{m}(\omega) \right|^2}{\left| \chi_{m,m}^{m}(\omega) \right|^2} \frac{M_{m}c_{m}(\omega)}{M_{m}c_{m}(\omega)},
\]

where the superscript “homo” denotes the homogeneous lattice, and “int” represents the interfacial system.

Compared with the existing harmonic based approaches that are used to compute phonon transmission, such as the atomistic Green’s function approach and wave-packet method, the advantages of the current method include the following: (1) All the quantities to compute the transmission coefficient in Eq. (15) can be directly extracted by analyzing the thermal average velocity, and therefore the harmonic force constants are not needed as the inputs; (2) this method can be applied to the strongly anharmonic and high-temperature systems, where phonon renormalization is non-negligible.

Although this derivation is based on a one-dimensional lattice, in principle our method can also be generalized to higher-dimensional cases according to the discussion by Liu et al. [26]. To monitor the wave propagation along the direction \( q = x\mathbf{b}_1 + y\mathbf{b}_2 + z\mathbf{b}_3 \) in a three-dimensional (3D) structure with \( \mathbf{b}_1, \mathbf{b}_2, \) and \( \mathbf{b}_3 \) as its three primitive vectors of the reciprocal lattice, we could choose an orthogonal plane of direction \( q \) and denote it as \( \sigma_1 \) first, and then apply the excited forces to all the atoms in this plane. The longitudinal anharmonic phonons are triggered when the excited forces are out-of-plane forces while the transverse phonons are triggered when the excited forces are in-plane forces. Similar to the one-dimensional (1D) chain model, the thermal averaged velocity of anharmonic phonons reaching atom \( n \) in plane \( \sigma_2 \) triggered by atom \( i \) in plane \( \sigma_1 \) can be calculated by

\[
\langle v_{n,i}(t) \rangle_f = \frac{1}{k_B T} \int_0^\infty \langle v_n(t) v_i(0) \rangle_f e^{-\beta(t - \tau)} d\tau = \sum_\omega \left[ f_{n,i}(\omega) \chi_{n,i}(\omega) e^{-i\omega(t - \tau)} \right].
\]

Then, the spectral energy flux at the position of the plane \( \sigma_2 \) induced by triggering the atoms in plane \( \sigma_1 \) can be written as

\[
I_{\sigma_2,\sigma_1}(\omega) = \sum_{n} \sum_{i \in \sigma_1} c_n(\omega) M_n \frac{f_{n,i}(\omega) \chi_{n,i}(\omega)}{\omega^2},
\]

where \( i \in \sigma_1 \) and \( n \in \sigma_2 \) mean atom \( i \) in plane \( \sigma_1 \) and atom \( n \) in plane \( \sigma_2 \), respectively. Finally, the transmission coefficient between any pair of atomic planes \( (\sigma_2, \sigma_3) \) can be calculated through the ratio of the spectral energy fluxes:

\[
\Gamma_{\sigma_2,\sigma_3}(\omega) = \frac{I_{\sigma_2,\sigma_3}(\omega)}{I_{\sigma_2,\sigma_2}(\omega)}.
\]

### B. Numerical details

The simulation systems studied in this work consist of \( N = 2400 \) atoms, with the lattice constant \( a_0 = 2 \) Å and the reference mass \( M_0 = 28 \) g/mol. The interatomic potential is described by the FPU-\( \beta \) model, where \( V(x) = \frac{k_0}{2}x^2 + \frac{\beta}{4}x^4 \) and \( U(x) = 0 \) with the constants \( k_0 \) and \( \beta \) characterizing the strengths of the harmonic and quartic interactions. Dimensionless variables are introduced to present the results, including lengths in units of \( [a_0] \), frequencies in units of \( 2\sqrt{\frac{k_0}{M_0}} \), anharmonicity strength \( \beta \) in units of \( [\frac{k_0}{a_0^4}] \), and momenta in units of \( [2a_0\sqrt{k_0M_0}] \). We use the superscript “*” to denote the dimensionless quantities in the remainder of the paper.

In the simulations, the periodic boundary condition is applied to the simulation systems and a dimensionless time step \( t^* = 2t/\sqrt{\frac{M_0}{k_0}} \approx 0.18 \) is chosen to integrate the equation of motion. During each simulation, we first generate the initial velocity for each atom randomly according to the Maxwell distribution, and then run \( 5 \times 10^5 \) steps under the canonical (NVT) ensemble to thermalize the system at the target temperature, followed by another \( 5 \times 10^5 \) steps being performed to further equilibrate the system under the microcanonical (NVE) ensemble. According to the ergodicity theory, the ensemble average in Eq. (4) is evaluated by performing a time average during the simulation. The velocity correlation function \( \langle v_n(\tau) v_i(0) \rangle \) is calculated by averaging the data over \( 1.4 \times 10^5 \) steps. When performing the numerical integration in Eq. (6), we typically set the correlation time at 5000 steps, which is longer than the time needed for waves to propagate to the 400th atom.

To reduce the influence of system uncertainty, five sets of parallel simulation calculations with different initial random atomic velocities are performed for all the results of TCOA. The final result of every TCOA is the average result of five parallel calculations. At the same time, the standard deviations are also calculated among every set of five parallel calculations.

### III. RESULTS AND DISCUSSION

#### A. Transmission coefficients of anharmonic phonon

When the anharmonicity is weak, the predicted phonon transmission coefficients through interfaces from the anharmonic phonon approach should be consistent with the methods based on harmonic approximation. To validate our approach, we apply it to two interfacial systems, as shown in Figs. 1(b) and 1(c), constructed by connecting different FPU-\( \beta \) lattices with weak anharmonicity. The temperature and strength of the quartic interaction are chosen as \( T^* = 0.0065 \) (equivalently, \( T = 300 \) K) and \( \beta^* = 0.8 \). To ensure
that with the choice of these parameters the current approach converges to harmonic solution, the phonon properties, including phonon dispersion and phonon mean free paths, of these FPU-\(\beta\) lattices are examined first.

Figure 2 shows the velocity correlation function \(\langle v_i(t)v_i(0)\rangle\) of a homogeneous FPU-\(\beta\) lattice illustrated in Fig. 1(a) with \(T^* = 0.0065\), \(\beta^* = 0.8\), and \(m_1 = M_0\). The oscillation behavior for the velocity-velocity correlation function can be clearly identified. With the increasing of the distance between two atoms, the time needed for phonon waves excited at the position of the first atom propagating to the \(n\)th atom becomes longer. Meanwhile, the amplitude of the oscillation in the velocity correlation function becomes smaller, indicating that the excited phonons are decaying in the anharmonic lattice. Using the velocity correlation function, the dynamic susceptibility is extracted for different frequencies according to Eq. (6). As shown in Fig. 3(a), the amplitude \(|\chi_{n,1}\)| decays exponentially with increasing \(n\). By fitting the relation between \(|\chi_{n}|\) and \(n\) through Eq. (11), the phonon mean free paths of anharmonic phonons \(l(\omega)\) are determined. The wave-number-dependent phonon mean free paths for two chains with different atomic masses \((m_1 = M_0\) and \(10M_0))\) are plotted in Fig. 3(b). The amplitude \(|\chi_{n,1}\)| decays faster with increasing the frequency, so the phonons with lower dimensionless frequencies tend to be of longer mean free paths. In particular, the mean free paths become divergent when the frequency approaches zero. These observations are in accordance with the theoretical analysis on the FPU-\(\beta\) lattice which predicts that \(l \sim k^{-1.70}\) [33–35]. In addition, the results illustrate that the mean free paths with the same dimensionless frequency are larger in the lattice with small atomic mass than those in the heavy counterpart and the mean free paths for both two systems are larger than 75 lattice constants.

Figure 3(c) shows the dependence of the phases \(\psi_{j,i}\) on \(|j-i|\). As expected, a linear relation between the phase and the distance is identified. Using the relation \(k(\omega) = -\psi_{j,i}(\omega)/|j-i|\), the phonon dispersion of anharmonic phonons under different temperatures is calculated, as depicted in Fig. 3(d). The phonon dispersion of the corresponding harmonic lattice is also plotted as the blue dashed line in Fig. 3(d). It is evident that the phonon dispersion of the homogeneous anharmonic lattice determined by the anharmonic phonon approach is very close to the prediction from harmonic lattice dynamics. Both the long phonon mean free paths and the agreement of the phonon dispersion from the anharmonic phonon method with that from harmonic lattice dynamics suggest that the anharmonicity in these two homogeneous lattice systems is so weak that the phonon renormalization can be neglected in these two lattices under \(T^* = 0.0065\).

We then apply the anharmonic phonon method to interfacial systems made by connecting the two lattices with different masses, \(M_0\) on the left-hand side and \(10M_0\) on the right-hand side. Both lattices are kept at low temperature and have weak anharmonicity \((T^* = 0.0065\) and \(\beta^* = 0.8))\), the same as the discussed above. In such a system of one-dimensional interface, the atom triggered by external forces launches an incident plane wave propagating from the left to the right direction and another plane wave moving along the opposite direction. The interface is at the 200th atom. Since the lattice chain is so long that the time needed for the left-going wave to encounter the interface is much longer than that for the right-going wave, only the right-going wave is analyzed in this work. In this case, if the position of the atom is on the left side of the interface \((n < 200)\), both incident and reflected waves can be observed, while only the transmitted wave can be found when the trigerring atom is on the right side of the interface, as shown in Fig. 4.

Figure 5(a) shows the amplitude of susceptibility across the interface region. A sharp decline of the calculated \(|\chi_{n}|\) at the location of the interface \((p = 40)\) can be clearly identified, suggesting that only a portion of energy transmits through the interface. In Sec. III B, the effects of the location of the excited atom away from the interface on phonon transmission will be discussed. Figure 5(b) compares the frequency-dependent phonon transmission coefficients of the anharmonic phonons for different combinations of \(m\) (position chosen for calculating incident energy flux) and \(n\) (position chosen for...
calculating transmitted energy flux). The standard deviations of transmission coefficients are smaller than 10% for most frequencies. Compared with the transmission coefficients from a single simulation, we find that averaging the results of five parallel simulations could significantly reduce the uncertainties; otherwise the obtained transmission coefficients of low-frequency phonons might unphysically exceed unity. We expect more accurate results can be obtained by continuously increasing the number of independent simulations. In theory, one would expect that the phonon transmission coefficients should decrease both with \(|n - p|\) and \(|p - m|\) as the longer distance away from the interface leads to more phonon-phonon scatterings within the bulk region and thus fewer phonons can travel from the position of atom \(m\) to atom \(n\). If we set \(m\) and \(n\) to be the atoms just before and after the interface, phonon scattering should be eliminated across the interface, and the transmission coefficients calculated from the anharmonic phonon approach are indeed very close to the AGF prediction [see Fig. 5(b)], confirming that these two approaches are essentially equivalent in weakly anharmonic interfacial systems. We then further increase the propagating distances before and after the interfaces \((|n - p|, |p - m|)\). The phonon transmission coefficients are found to decrease with the increase of \(|n - p|\), especially in the high-frequency region, while the phonon transmission coefficients are found to be close to those with different \(|p - m|\) when \(|n - p|\) is fixed. However, the decrease of transmission coefficients of anharmonic phonons with increasing \(|p - m|\) is too negligible to be observed due to the long mean free paths of the lattice with lighter atomic mass.

In addition to the system with a single interface, the anharmonic phonon based method is also extended to heterostructures with multiple interfaces. In the heterostructures studied here [see Fig. 1(e)], three atoms with heavier atomic masses are inserted in a homogeneous lattice with atomic masses of \(M_0\) to form a heterojunction. The atomic mass of the three inserted atoms is \(2M_0\) or \(4M_0\). The comparison between the transmission coefficients and the results of AGF for a heterojunction system is shown in Fig. 6. The resultant phonon transmission coefficients are again consistent with the prediction by the AGF approach. At low frequencies, the transmission function is unity, since the long-wavelength phonons cannot see the nanostructure, whose characteristic length is much smaller than the phonon wavelength. With the increase of the frequency (the decrease of phonon wavelength), the transmission is reduced due to the scattering by the heterogeneous interfaces. One can observe two Fabry-Perot peaks, where the transmission function approaches unity, caused by the resonant scattering of phonons. The obtained phonon transmission coefficients from the anharmonic approach are slightly larger than one for some frequencies near the Fabry-Perot peaks. This is probably caused by the aforementioned numerical uncertainty when computing the velocity correlation function.

To explore the phonon transmission on strongly anharmonic lattices, we compute the transmission coefficients through the heterojunction with \(M_1 = 4M_0\) and \(M_2 = 4M_0\) at higher temperatures \((T^* = 0.0129\) and \(0.0324\)), as shown in Fig. 7(a). The phonon transmission curves under different temperatures show a similar trend. However, there are some quantitative differences. With the increasing of the temperature, the high-frequency phonons \((\omega^* > 0.5)\) seem more likely to transmit through the interfaces. In addition, the maximum
The theory predicts phonon-phonon interaction caused by anharmonicity as derived based on the ergodic hypothesis and expresses the which are computed according to Eq. (8). The predictions presents the results of the harmonic lattice.

frequency of phonons that can transmit keeps increasing with the temperature. In fact, this frequency cutoff even exceeds the frequency that the lattice with $M_2 = 4M_0$ can support based on the prediction from the harmonic lattice dynamics. Since the phonon dispersion is affected by the lattice constant of the material, one possible explanation for the high-frequency cutoff is that the lattice constants are altered under high temperatures. From our analysis on the simulation data of atomic positions, the lattice constants of both materials are almost unchanged during the molecular dynamics simulations. Therefore, the temperature-dependent lattice constants should not be related to the change of the frequency cutoff.

To understand the temperature effects on phonon transmission coefficients, the phonon dispersion relations of anharmonic phonons under high temperatures are shown in Fig. 7(b), which are computed according to Eq. (8). The predictions from the renormalized phonon theory (the dashed lines) [36,37] are also plotted. The renormalized phonon theory is derived based on the ergodic hypothesis and expresses the phonon-phonon interaction caused by anharmonicity as an effective harmonic one. The theory predicts

$$\omega(k) = 2\alpha(T) \sin \frac{k}{2}, \quad (19)$$

where the renormalized factor $\alpha(T)$ is temperature dependent and quantifies the strength of anharmonicity. For the FPU-$\beta$ lattice, $\alpha(T)$ can be written as

$$\alpha(T) = \left(1 + \frac{\int x^4 e^{-V(x)/T} dx}{\int x^2 e^{-V(x)/T} dx} \right)^{\frac{1}{2}}. \quad (20)$$

The results from the renormalized phonon theory are consistent with those from the anharmonic phonon method. Both approaches indicate the fact that the span of phonon dispersion is upshifted in an anharmonic system compared with a harmonic lattice and the upshifting becomes more evident with higher temperature. Meanwhile, the cutoff frequency of the transmission coefficient is almost the same as the cutoff frequency of dispersions of anharmonic phonons under the corresponding temperature. Therefore, the temperature-induced change of phonon dispersion should be responsible for the temperature-dependent phonon transmission coefficients.

While in the FPU-$\beta$ lattices the anharmonicity comes from the quartic terms in the potential $V$, for realistic materials the anharmonicity to all orders occurs and affects the phonon spectra under finite temperatures. Although a few approaches have been proposed to treat phonon renormalization in bulk materials [38–40], these attempts cannot be applied to interfacial systems directly due to the breakdown of the translational invariance. It is worth mentioning that the current method does not have such a limitation and can be used in complex interfacial systems, as the renormalization effects are intrinsically considered in the molecular dynamics simulations.

B. Redistribution effect of anharmonicity in interfacial systems

While we have explored the temperature-dependent transmission coefficients in anharmonic interfacial systems, it is worth having a closer look at the role of anharmonicity in the collective behaviors of phonon transport across interfaces. In the previous NEMD simulations, a positive correlation between the obtained thermal conductance and distance of thermal reservoirs away from the interface had been clearly observed [41–43]. This phenomenon is opposite to the expectation for a purely harmonic interfacial system, where the thermal conductance should be independent of the system size, indicating that the anharmonicity in the lattice does alter the energy transfer across the interface. Since our approach monitors the response of phonon waves to the external excitations, one can easily change the location of the interface away from the excited atom and explore the size effects on energy transmission.

Size effects of thermal conductance can be understood through our anharmonic phonon picture as follows. Energy is injected in the heat source, where the population of phonons with different frequencies are disturbed, and deviates away from the equilibrium distribution. During the propagation of these phonons, the spectral distribution of the incident energy would be different from that at the heat source due to phonon-phonon scatterings. Farther away from the heat source, there will be stronger redistribution of spectral flux. Therefore, the population of the excited phonons reaching the interface should be size dependent. In addition to the spectrum of the incident phonons, the thermal conductance is also related to the transmission possibility of the phonons reaching the interface. Whether the phonon transmission coefficient is size dependent should affect the thermal conductance in the anharmonic lattice as well.

To test the above hypothesis, we perform a series of calculations by changing the position of the heterojunction $p$ and then compute the energy flux of the two atoms right before and after the heterojunction ($m$ and $n$) according to Eq. (13). In Eq. (13), $f(\omega)$ is the Fourier transform of the driving force. When analyzing the energy flux induced by the external force, we set $f(\omega)$ to be a constant 1 to let the same amount of energy be injected into the phonons with different frequency.

Figure 8(a) shows the energy flux of the incident anharmonic phonons at the atom before the heterojunction and that of the transmitted anharmonic phonons at the atom just after the heterojunction. The energy flux of incident anharmonic

![FIG. 7. (a) The TCOA for a heterojunction system under different anharmonicity when the mass ratio is 4. (b) The phonon dispersion of anharmonic phonons under different anharmonicities. RT denotes the corresponding result of phonon renormalization theory. Red, black, and blue represent the results of different dimensionless temperatures 0.0065, 0.0129, and 0.0324, respectively. Green represents the results of the harmonic lattice.](image-url)
The temperature is 0.0065 and the dimensionless anharmonic strength is different positions in a homogeneous lattice when the dimensionless and the mass ratio is 4. (c) Composition of anharmonic phonons at junctions (\(\text{transmitted energy flux}\)). The red line represents the energy flux of incident anharmonic phonons reaching the heterojunctions when the heterojunctions position is changed, the ratio \(y(\omega) = \frac{|\chi_{j,i}(\omega)|}{\int |\chi_{j,i}(\omega)| d\omega}\) (22) is calculated to present the composition of the phonon energy of a frequency \(\omega\) occupying the total energy. This definition of \(y\) is similar to that provided by Latour and Chalopin [44], where they proposed to use the cross-spectral density function, \(m_{i,j}(\omega)\), to represent the composition of phonons. When atoms \(i\) and \(j\) are the same atom, the ratio \(y\) becomes proportional to the phonon density of states. On the other hand, if they are different atoms, \(y\) represents the portion of phonons that keep their phase information during the propagation and \((1 - y)\) means the portion of those that are scattered. Figure 8(c) shows the composition of anharmonic phonons at different positions in a homogeneous lattice with atom mass \(M_0\) when \(T^* = 0.0065\) and \(\beta^* = 0.8\). The high-frequency anharmonic phonons are found to occupy the majority when the atom position is located nearby the driving source and the composition of low-frequency anharmonic phonons keeps increasing with the distance between the atom position and the driving source increasing. Hence, due to the anharmonicity in the lattice, the spectrum of phonons that arrive at the interface is distinct to the distribution of the excited phonons at the source, and highly depends on the distance between the source and the interface.

To see whether the probability of an anharmonic phonon transmitting through the interface (transmission coefficient) is relevant to the location of the excited atom, we calculated the frequency-dependent phonon transmission in several systems with the heterojunction placed at different positions. As shown in Fig. 9, the phonon transmission is almost independent of the position of the heterojunction. Since the external force acting on the “heat source” atom is weak, the perturbation in the system is too weak to affect the spectrum of phonons in both sides significantly. Thus, the distance between the heat source and interface does not change the phonon transmission.

Based on these observations, the length-dependent spectrum distribution of phonons reaching the interface should...
be the origin of the increase of the coherent energy flux transmission coefficient with the distance between the excited atom and heterojunction. The length-dependent spectrum distribution of phonons is caused by the anharmonic phonon scatterings. Unlike harmonic lattices, the MFPs of phonons in our anharmonic lattices are no longer infinite because of the anharmonic scatterings. In addition, as we have mentioned, the MFPs of high-frequency anharmonic phonons are much smaller than the low-frequency anharmonic phonons, indicating that high-frequency phonons experience much stronger anharmonic scatterings. To display the redistribution effect more clearly, we also plot the spectral energy flux at different positions in a system with its heterojunctions located just after the 40th atom in Fig. 10. It shows that the energy flux in the high-frequency domain decreases rapidly as the atoms get closer to the heterojunctions, which is consistent with our above analysis. Although the above analysis from the anharmonic phonon approach only captures the transport of the coherent phonons, the results can also be used to explain the size dependence of thermal resistance at interfaces. During the propagation, high-frequency phonons are continuously converted to low-frequency phonons through phonon-phonon scatterings. While the calculated energy flux does not include the contribution of these scattered phonons, these scattered phonons could more easily go through the interface due to the larger phonon transmission coefficient. When the source of the energy that is injected into the system becomes farther away from the interface, the larger amount of low-frequency phonons reaches the interface resulting in the larger total thermal conductance.

In addition to the size effects, we also investigate the dependence of the energy flux transmission coefficient on the temperature and anharmonicity of the lattice. We fix the position of the heterojunction at the 40th atom, and change the temperature ($T^*$) and the anharmonicity strength ($\beta^*$), respectively. The obtained CETCs are shown in Fig. 11. The obtained CETCs continuously increase with the temperature or the strength of the anharmonicity, which could be understood by the fact that the higher temperature and stronger anharmonicity strength both can promote the redistribution of anharmonic phonons, resulting in the enhancement of thermal conductance.

IV. CONCLUSIONS

Based on the concept of triggering anharmonic phonons in a nonlinear lattice, we have visualized phonon transmitting through an interfacial region. The basic idea was to excite an atom and to monitor the propagation of the resultant phonon waves. By analyzing the velocity correlation function between atoms before and after the interface and the excited atom from EMD simulations, an efficient approach to extract frequency-dependent phonon transmission coefficients is developed. We tackle the interfacial systems with different FPU-$\beta$ lattices using the proposed method. While under low temperatures or with weakly anharmonic lattices the obtained phonon transmission coefficients are consistent with the prediction from the atomistic Green's function, under high temperatures the phonon transmission coefficients are found altered considerably due to phonon renormalization. Moreover, we also examine the energy flux before and after the interface, and find that the distance between the excited atom and the interface severely affects the energy transmission coefficient. This phenomenon can be explained by the redistribution process of anharmonic phonons modes. More specifically, the high-frequency phonons, which are less likely to transmit through the interface, are converted to low-frequency phonons because of the anharmonicity in the bulk region.

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