

Phonon Wave-Packet Simulations of Ar/Kr Interfaces

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Abstract—The frequency- and direction-dependence of transmission coefficients at interfaces was investigated theoretically. The interfaces are formed by having two Lennard-Jones (L-J) materials differing in mass and interatomic potential equally divided at the center of an FCC lattice system. A single frequency wave-packet is generated at one end of the system and allowed to propagate through the system until all interactions with the interface are complete. The transmission coefficient is then calculated by comparing the energy of the packet that is transmitted with the original wave-packet. Results show a difference in transmission when the wave-packet originates from opposite sides.

NOMENCLATURE

a	lattice constant (m)
A	amplitude (m)
k	wave vector (m^{-1})
l	device length (m)
T	temperature (K)
V_g	phonon group velocity (ms^{-1})
w	device width (m)
z	location along device (m)
z_n	location of nth atom along device (m)
z_o	location of center of wave-packet (m)
ϵ	polarization vector
ω	phonon frequency (Hz)
τ	phonon transmission coefficient
ξ	width of wave-packet (m)

I. INTRODUCTION

Thermal rectification is a phenomenon where the magnitude of transport in a specific direction through a material is dependent on the sign of the temperature gradient, and though observations of the effect in solids have been rare, rectifying behavior could have widespread applications. Materials with different thermal transport properties in opposite directions along the same axis would revolutionize how thermal management problems from the nano and microscale to the macroscale are approached. Thermal management has been an issue and is becoming even more important in microelectronics where device sizes are becoming smaller and new technologies such as 3D circuit designs make heat removal extremely difficult. Materials that exhibit thermal rectification would drastically improve our ability to control the flow of heat in microelectronic devices similar to the way we are able to control the

flow of charge carriers. Thermally rectifying materials would allow engineers to design chips that preferred transport along a specific direction (eg. to a location to dissipate heat) while providing resistance to the flow of heat towards temperature sensitive portions of the chip (logic circuits). Clearly this would allow for more compact and higher performance circuits. Records of thermal rectification date back as early as 1935 when Starr [1] found that copper-cuprous oxide systems showed thermal as well as electrical rectification. In 2006 Walker [2] reviewed several candidate theoretical models based on geometric asymmetries with unequal thermal expansion and offered several alternate explanations for rectification behavior based on non-equilibrium effects. Examples of these models include lattice vibrations which reflect differently depending on the geometry and characteristics of the structure or transmission across an interface. These theories suggest that rectification could be exploited with our improved ability to manipulate materials at the nanoscale. There have been several reports of thermal rectification at interfaces, but many of these are due to a difference in the temperature dependence of the thermal conductivity or a results of a change in thermal contact resistance [3, 4]

In 2002 Terraneo et al. [5] demonstrated theoretical rectification behavior using a nonlinear one-dimensional chain of atoms between two thermostats at different temperatures. In their system, Terraneo was able to change the chain from a normal conductor in one direction to a nearly perfect insulator in the other direction due to the non-linearity of the potential. In a similar study, Li et al. [6] simulated a nonlinear lattice and calculated a difference in conduction between the two directions to be 100 times that of ref. [5]. Also, Hu and Yang [7] found they could adjust the thermal conductivity by 10,000 times by adjusting the external potential. Hu and Yang also presented a design of a thermal diode and two designs of a thermal rectifier by coupling two or three Frenkel-Kontronova chains with different periodicity. In 2005 Casati [8] used a 1D nonlinear chain to show that, by acting on the parameter that controls the on site potential strength within the chain, the heat flow in the system can be controlled resulting in conducting or insulating behavior. Segal and Nitzan [9] argued that an asymmetry, in addition to nonlinear interactions, is necessary in a system for thermally rectifying behavior to exist. In 2006

Echmann and Mejia-Monasterio [10] showed the possibility to observe thermal rectification in asymmetric billiard-like mechanical systems. Chang et al. [11] suggested solitons could be responsible for rectification and showed experimentally that greater conduction resulted in the direction of decreasing mass density in an engineered material having a non-uniform mass distribution along nanotubes. The non-uniform mass distribution was obtained by depositing $C_9H_{16}Pt$ with a much higher concentration at one end of the nanotube than the other. Following this experiment, several theoretical studies followed to try to verify Chang’s hypothesis. In 2007 Wu and Li [12] used molecular dynamics to investigate thermal rectification in intramolecular junctions in carbon nanotubes. Wu and Li argued that optical phonons are the major contributors to thermally rectifying behavior at interfaces. Alaghemandi et al. [13] performed molecular dynamics simulations of carbon nanotubes with a mass gradient (similar to ref. [11]), but found the results to be the opposite of Chang’s measurements. Alaghemandi et al. showed a preferential transport direction of low-mass to high-mass. Similarly, Roberts and Walker [14] showed, via Monte Carlo simulations, that devices with asymmetric and frequency-dependent boundaries could exhibit thermally self-biasing behavior based on the difference in boundary reflections between phonons traveling in opposite directions that encounter a boundary.

Interfaces have also been proposed as a mechanism for thermal rectification. Hu et al. [15] showed rectifying behavior in a silicon-amorphous polyethylene interface from a vibrational analysis. Roberts and Walker [16] also presented theoretical evidence from molecular dynamics simulations of systems with a single interface between two dissimilar materials. Roberts and Walker used the Diffuse Mismatch Model (DMM) to estimate the maximum rectification in a system of two dissimilar materials divided by a single interface. Their molecular dynamics results agreed well with the maximum theoretical rectification from the DMM assuming no inelastic scattering at the interface. Other mechanism for thermal rectification have been proposed by Miller et al. [17] which include asymmetric defects, inclusions and voids. Miller et al. argues that these can result in asymmetric phonon transmission through a device resulting in thermal rectification.

Theoretical methods have been used to investigate thermal transport properties of low dimensional materials with some success (eg. Monte Carlo and molecular dynamics). There have been several groups that have used molecular dynamics to study thermal boundary resistance by generating single phonon wave-packets and allowing them to interact with the interface [18–21].

In this work Roberts and Walker’s hypothesis of thermal rectification at interfaces between two dissimilar materials due to a difference in phonon spectra will be tested using a Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [22]. Phonon wave-packet or zero-degree molecular dynamics simulations are performed in which a wave-packet is created by displacing a set of atoms and allowing the packet to propagate through the system while the rest

of the system is held at zero degrees. Phonon wave-packet simulations have been performed for interfaces [18–21], but these studies have only looked at transmission in one direction and never compared the transmission in opposite directions. The current work looks at the transmission coefficients from argon to krypton and krypton to argon as a function of phonon frequency.

II. SIMULATION METHOD

LAMMPS, a general purpose molecular dynamics package, is used to perform phonon wave-packet simulations in an argon/krypton system. The argon and krypton in the system uses a Lennard-Jones (L-J) potential parameters from the literature [23]. The transmission coefficients are determined for a range of frequencies from argon to krypton and the reverse. The transmission coefficients are obtained by first generating a wave-packet given by [18],

$$u_n = A\epsilon k \exp ik(z_n - z_o)^2 \exp -(z_n - z_o)^2/\xi^2, \quad (1)$$

where A is the amplitude of the displacement, ϵ is the polarization vector, k is the wave vector, z_n is the location of the atom along the direction of transport, z_o is the center of the wave-packet and ξ is the width of the wave-packet. In all the simulations we used $A = 0.002a$, $\epsilon = 1$, $z_o = 250a$, k ranges from $0.1(2\pi/a)$ to $0.9(2\pi/a)$ and $\xi = 100a$. The system we simulate is $1000a \times 2a \times 2a$ so that it is long enough to create a large enough wave packet ($100 - 200a$) far enough away from the interface so that it could propagate, interact and move away from the interface in order to collect information on the energy transmitted and reflected during the interaction. The system must also satisfy $\omega_{min} < \xi < L$ where ω_{min} is the minimum phonon frequency and L is the system length [21].

The simulations are then run at zero-degrees so that only this phonon wave-packet exists in the lattice and it is allowed to propagate and interact with the interface. The transmission coefficient is calculated by taking the ratio of the amplitudes of the transmitted and incident wave-packets given by [21]

$$\left(\frac{E_{tr}}{E}\right)^2. \quad (2)$$

Validation of the simulation was performed by initializing a wave-packet in a device of the same size, but composed of a single material (argon in this case). The full range of wave-vectors were simulated and the result was perfect transmission of the wave-packet along the device.

III. SIMULATION RESULTS

In this work the wave-packet was first generated in the argon side of the interface and was monitored as it propagated and interacted with the interface. This interaction with the interfaces resulted in some transmission into the krypton and some reflection back into the argon. In the next step the wave-packet was generated in the krypton and allowed to propagate and interact with the interface. Again in this second case some of the wave-packet transmitted into the argon while some was

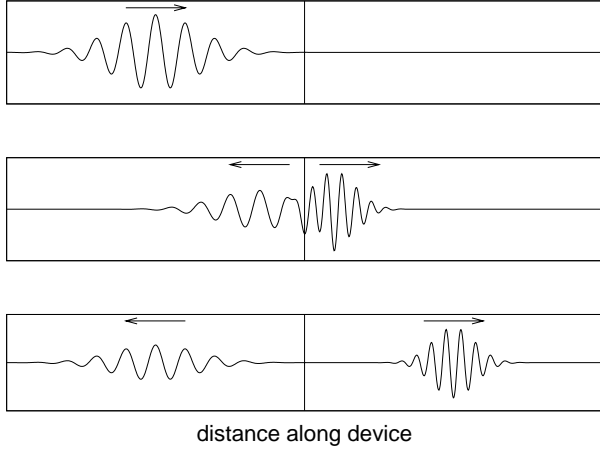


Fig. 1. Wave-packet propagation originating in argon with a normalized wave vector of 0.2

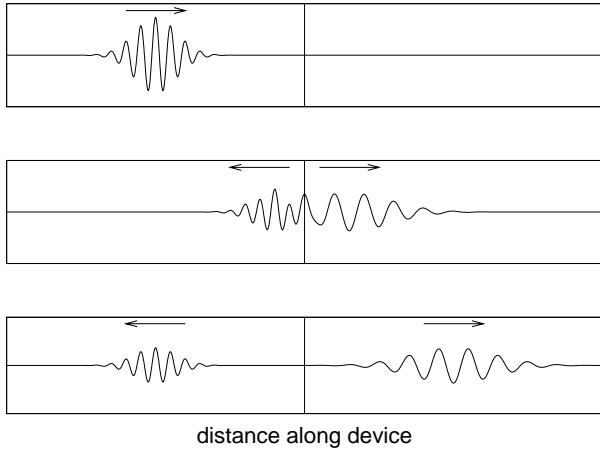


Fig. 2. Wave-packet propagation originating in krypton-like material with a normalized wave vector of 0.2

reflected back into the krypton. Figure 1 shows the propagation of a wave-packet originating in argon with a wave vector of $k = 0.2(2\pi/a)$. Figure 2 shows the propagation of a wave-packet originating in the krypton with the same wave vector as in Figure 1. By comparing these two figures we see that the transmission at these wave vectors are similar, but the frequency of the wave-packets are different because of the difference in phonon dispersion. The difference in dispersion can be observed in Figure 3 where an analytic dispersion is plotted for argon and krypton. From the dispersion of the two materials we expect that a wave-packet originating in the argon with a wave vector greater than about $0.5(2\pi/a)$ would be completely reflected because there are no transverse acoustic phonons at frequencies that large in krypton.

The transmission coefficients calculated for both the wave-packet originating in the argon and the wave-packet originating in the krypton are shown in Figure 4. The Acoustic Mismatch

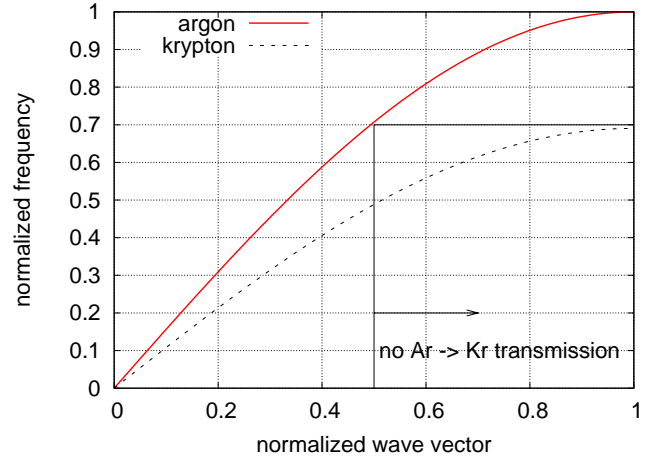


Fig. 3. Comparison of a single branch of an analytic dispersions for argon and krypton-like materials

model (AMM), given by

$$\tau_{AMM} = \frac{4Z_{Ar}Z_{Kr}}{(Z_{Ar} + Z_{Kr})^2}, \quad (3)$$

is a model used for the calculation of transmission probabilities. The AMM assumes that phonons are governed by continuum acoustics and that phonons can only reflect, reflect and mode convert, refract or refract and mode convert [24]. This model does not include any directional dependence and is based on the impedance of the two materials creating the interface. The Diffuse Mismatch model (DMM), is given by

$$\tau_{DMM,i}(\omega) = \frac{V_{g,i+1}(\omega)N_{i+1}(\omega, T)}{V_{g,i}(\omega)N_i(\omega, T) + V_{g,i+1}(\omega)N_{i+1}(\omega, T)}, \quad (4)$$

where V_g is the phonon group velocity and N is the phonon occupation number. The DMM assumes that when a phonon interacts with an interface it loses all memory of where it originated and the phonon selects which material it transmits into based on the probability calculated. A formulation is needed here to account for the probability that a phonon originated from a specific side (either the argon or krypton) in order to calculate the transmission from one material into the other. Both the AMM and the DMM predictions are included in Figure 4. From the phonon wave-packet simulations at low wave vectors the transmission from the argon to the krypton is greater and is close to the prediction from the AMM as expected [24]. At higher wave vectors the transmission is greater from the krypton to the argon and diverges from the AMM, but does not match the DMM. The transmission coefficient from the argon to the krypton becomes zero at a wave vector of 0.6. No transmission is expected in this case because there are no frequency modes in the krypton at this particular frequency (see Figure 3). Our expectation that no transmission should occur is based on the difference in phonon dispersion and the assumption that there is no inelastic scattering at the interface [20]. Since the DMM calculation only considers the frequency dependence of the group velocity

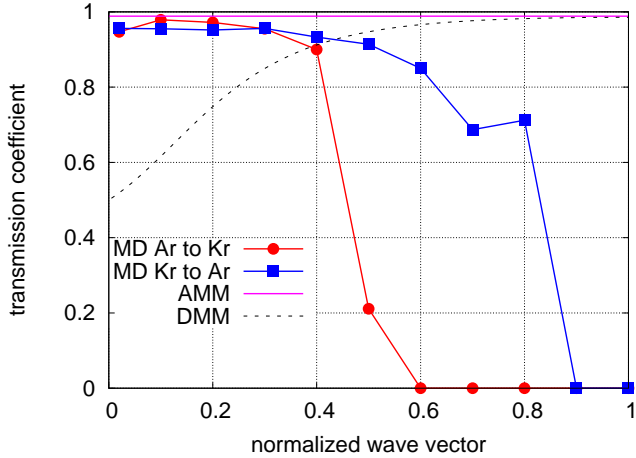


Fig. 4. Transmission coefficients of wave-packets propagating from argon and krypton into the opposite as a function of wave vector

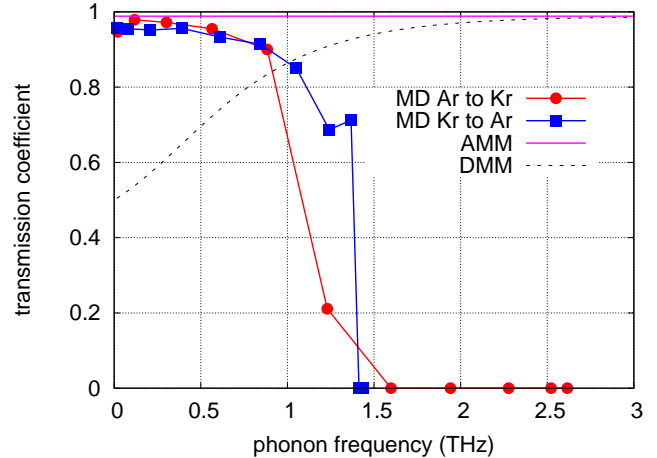


Fig. 5. Transmission coefficients of wave-packets propagating from argon and krypton into the opposite as a function of frequency

and the ratio of density of states and not the dispersion there is a calculated probability of transmission even when those phonon frequencies are not possible when no inelastic scattering occurs. The DMM also does not consider the origin of the phonon which makes calculating transmission rates from the DMM impossible for comparison. A system at zero degrees and having a perfect planar interface should exhibit no inelastic scattering and therefore perfect reflection. These results show that there is indeed a wave vector dependence on the phonon transmission at the interface between dissimilar materials. A dependence on wave vector is not necessarily of importance here since we are interested in energy transport across an interface. Figure 5 shows the phonon transmission coefficient as a function of phonon frequency. We obtained the frequency of the wave-packet for the specific wave vector from the molecular dynamics simulations which agreed well with the analytic dispersion and the literature [25]. The transmission as a function of frequency appears to be slightly different around the maximum frequency in the krypton when no inelastic scattering occurs. This different in transmission at this small frequency range could possibly be exploited to produce thermal rectification, but in order to determine this a thermal conductance will need to be calculated.

IV. CONCLUSION

Thermal rectification studies have been performed, but this is the first to look at frequency dependence of phonon transmission at an interface. The AMM does not contain any directional dependence on the transmission coefficient therefore we are not able to compare the value we calculated to the theoretical prediction of the AMM unless we assume an average transmission coefficient. Clearly from the wave-packet results here there is a difference in phonon transmission from the argon to the krypton than from the krypton to the argon. With a strong frequency dependence in phonon transmission at perfect interfaces thermally rectifying materials are possible to obtain when no inelastic scattering occurs..

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