Phonon wave-packet interference and phonon tunneling based energy transport across nanostructured thin films

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The molecular dynamics based phonon wave-packet technique is used to study phonon transport across mass-mismatched fcc thin films. Transport behavior of normally incident longitudinal acoustic phonon wave packets with wave vectors ranging in magnitude from 2% to 50% of the ⟨100⟩ first Brillouin zone boundary is examined as a function of thin film thickness when the phonon mean free path exceeds film thickness. The results indicate that for thin film to bulk solid mass ratios up to a factor of 6, the transmission of energy through the thin film can be well described by treating the thin film as a bulk solid. © *2010 American Institute of Physics*. [doi:10.1063/1.3458831]

Recent advances in nanomaterials and processing technologies have enabled the creation of a large number of heterogeneous nanostructures for use in information processing, energy generation, sensors, and biological applications.[1,2] Epitaxially grown thin films have been used to fabricate thermoelectrics,[1] quantum cascade lasers,[3] and thermal interface materials[4] with significantly improved functionality. In these nanostructured devices, phonons can both be beneficial and detrimental. Tuning of device structures is often desired to achieve optimal phonon transport behaviors. However, due to the combination of phonon wave effects at small length scales and diffusive heat transfer at macroscopic scales, the nature of phonon transport in nanostructured thin films is only partially understood.

Phonon wave packets are linear combinations of vibration eigenstates of a perfect crystal. The fundamental premise of the phonon wave-packet technique is to construct a phonon wave packet from a single branch of a solid's phonon dispersion curve with a narrow frequency range and well-defined polarization.[5] A molecular dynamics (MD) simulator then propagates the wave packet towards the interface or materials of interest. Schelling *et al.*[5] have shown that the technique is applicable to studies of phonon scattering at ideal interfaces,[5,6] grain boundaries,[7] rough surfaces,[8] material inclusions,[9] and defects[10] for Lennard–Jones (LJ) solids, semiconductors, carbon nanotubes, and polymers. It was shown from simulations of scattering of phonon wave packets in superlattices that, phonon wave-packet interference plays an important role in determining the energy transmission through superlattices.[6] In this paper, we utilize the MD-based phonon wave-packet method to directly observe phonon transport across single-crystal thin films of different thicknesses. Our focus is on the deviations observed in the transport of acoustic phonons of different frequencies from those predicted by the acoustic mismatch model (AMM).[11]

A single-crystal, mass-mismatched LJ thin film sandwiched inside an Ar solid is used in the simulations where all interfaces are assumed to be perfect. An fcc crystal of Ar, $1000a \times 2a \times 2a$ in dimension, with a thin film starting at $500a$ in x is initialized at 0 K with its [100] direction along the x axis where a is the lattice constant of Ar at 0 K. As discussed in Schelling *et al.*,[6] the length scales involved in the phonon wave-packet study (i.e., the dominant phonon wavelength λ, phonon mean free path l, and spatial width of the wave packet ξ) must satisfy $l > \xi > \lambda$. This results in a wave packet size of $100-200a$ and a domain size of $1000a$. The wave packet is formed by linear combinations of the vibration normal modes, determined from the dynamical matrix of the LJ perfect crystal. A wave packet centered at k_0 in k space and x_0 in real space is generated by setting the displacement as

$$u_n = A\varepsilon(k_0)\exp[ik_0(x_n - x_0)]\exp[-(x_n - x_0)^2/\xi^2], \quad (1)$$

where u_n is the displacement of the nth atom, A is amplitude of the wave, and ε is the polarization vector. The phonon dispersion curve index is set to be that of the longitudinal acoustic (LA) phonons with the polarization vector directed in the x direction. Inverse Fourier transformation is performed to resolve the wave packet in k space, and hence determines the corresponding phonon normal modes. Adding time dependence to the atom displacements based on linear combinations of the normal modes of the crystalline lattice and differentiating with time, the initial velocities of atoms are obtained. When the initial wave packet of amplitude A and frequency ω encounters an interface, elastic scattering at this interface results in a transmitted wave and a reflected wave both at frequency ω. By computing the ratio between the amplitudes of transmitted (A_{tr}) and initial (A) phonon wave packets across a thin film, the energy transmission coefficient α can be determined by[6]

$$\alpha = \left(\frac{A_{tr}}{A}\right)^2. \quad (2)$$

The perfect fcc thin film considered here has mass four times that of the bulk material. Only acoustic phonons are excited due to the presence of a single atom basis. First, small k_0 values are considered. The two sets of parameters used are: (i) $A=0.002$, $\varepsilon=1$, $x_0=250a$, $k_0=0.02(2\pi/a)$, and

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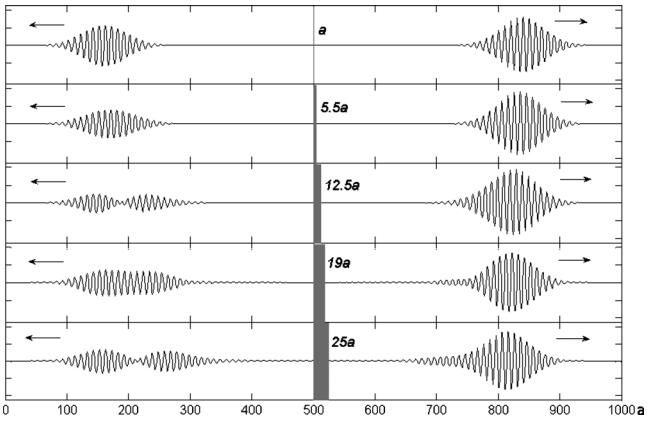

FIG. 1. Snapshots of displacements for a phonon wave packet of $k_0=0.1(2\pi/a)$ at 200 ps. From top to bottom, the thin film thickness, d, equals to a, $5.5a$, $12.5a$, $19a$, and $25a$, respectively.

$\xi=50a$ and (ii) $A=0.0004$, $\varepsilon=1$, $x_0=250a$, $k_0=0.1(2\pi/a)$, and $\xi=50a$. If we assume the thin film has a phonon dispersion relationship given by the bulk fcc solid but with mass four times that of the bulk, we can analytically determine the expected transmission coefficient and compare that to the MD simulations. At the bulk crystal/thin film interface, some phonons are transmitted while others are reflected. The transmitted wave packets soon reach the second thin film/bulk crystal interface, producing transmitted and reflected waves at this second surface. The transmitted and reflected wave packets generated by these two thin film surfaces interfere. Figure 1 shows snapshots of displacements after transmitted and reflected wave packets have separated for a thin film thickness, d, of a, $5.5a$, $12.5a$, $19a$, and $25a$. The results show that the amplitude of the transmitted wave does not vary monotonically with the thin film thickness. The phonon group velocities of transmitted and reflected waves decrease with the increase in thin film thickness due to a smaller group velocity in the denser film region. In addition, two out-of-phase reflected wave packets for the $d=12.5a$ and $25a$ cases are clear evidence of phonon wave-packet interference.

Since the amplitude of reflected wave packets is reduced geometrically after each interaction with the interface, only the first two wave packets going through the thin film are considered. If the two wave packets are in-phase, constructive interference takes place and the transmitted energy is large. However, if the two wave packets are out-of-phase, destructive interference leads to a reduced transmission coefficient. It is important to note that, the wave-packet interference is different from that of plane wave interference due to the Gaussian distribution of the wave packet that limits interference effects to a limited spatial region where the two wave packets overlap. As shown in Fig. 2, the transmission coefficient determined from MD simulation oscillates as a function of the thin film thickness. For a given wave number, the oscillation can be calculated analytically based on superposition of two initial wave packets at frequency ω and assuming a transmission coefficient given by AMM. With these assumptions, the energy transmission coefficient, α, between materials 1 (bulk) and 2 (thin film) is given by

$$\alpha=\frac{4Z_1Z_2}{(Z_1+Z_2)^2}, \tag{3}$$

where $Z_i=\rho_i c_i$, ρ_i is the mass density and c_i is the speed of sound in material i. For $m_2=4m_1$, $Z_2=2Z_1$, and $\alpha=8/9$. By

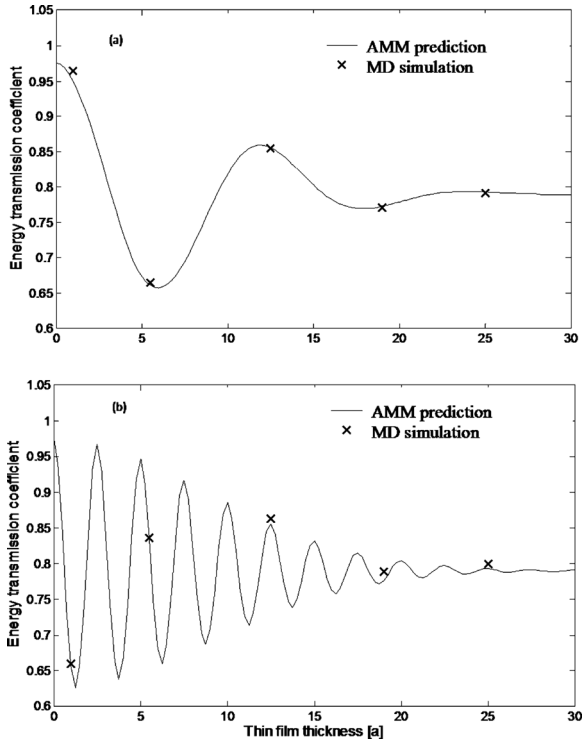

FIG. 2. Comparison of simulated phonon transmission coefficients with the AMM predictions for a wave packet of (a) $k_0=0.02(2\pi/a)$ and (b) $k_0=0.1(2\pi/a)$ across films of various thicknesses.

superposition of the first two wave packets emerging from the thin film with amplitudes of $(\sqrt{\alpha}\cdot\sqrt{\alpha})A=(8/9)A$ and $(\sqrt{\alpha}\cdot\sqrt{1-\alpha}\cdot\sqrt{1-\alpha}\cdot\sqrt{\alpha})A=(8/81)A$, respectively, and a $2d/v$ time delay between them (here v is the bulk phonon group velocity in the thin film, and, with our assumptions, is equal to one half of the phonon group velocity in the Ar bulk material), the phonon transmission coefficient versus thin film thickness can be determined. Wave packets due to additional reflections at the interfaces are neglected due to the geometric decay of amplitudes with common ratio 1/9. Figure 2 shows the comparison between the analytically calculated α values based on AMM [i.e., Eq. (3) and the superposition of the elastically scattered wave packets] and the α values extracted from MD simulations for $k_0=0.02(2\pi/a)$ and $k_0=0.1(2\pi/a)$. Good agreement is achieved for both cases for LA phonon wave packets considered here. Thus, it appears that treating the thin film as if it has the phonon dispersion relationship of a macroscopic crystal accurately describes the transmission of energy under these conditions.

When a large k_0 is considered, the role of thin film changes due to the lack of LA energy states in the film with real wave vectors (again assuming a macroscopic crystal phonon dispersion relationship for the film) at the appropriate frequency. However, states with complex wave vectors do exist in the film at the appropriate frequency. The complex form of the wave vector results in an exponentially decaying wave-packet amplitude as the wave packet traverses the thin film. Figure 3 shows the MD results for the energy transmission coefficient and phonon displacement for thin films of thickness $0.5a$, a, and $1.5a$. The parameters here are $A=0.00001$, $\varepsilon=1$, $x_0=250a$, $k_0=0.5(2\pi/a)$, and $\xi=50a$. The results indicate that the thin film effectively blocks high frequency phonon energy, with only the evanescent wave passing through thin films of thickness comparable to a. This

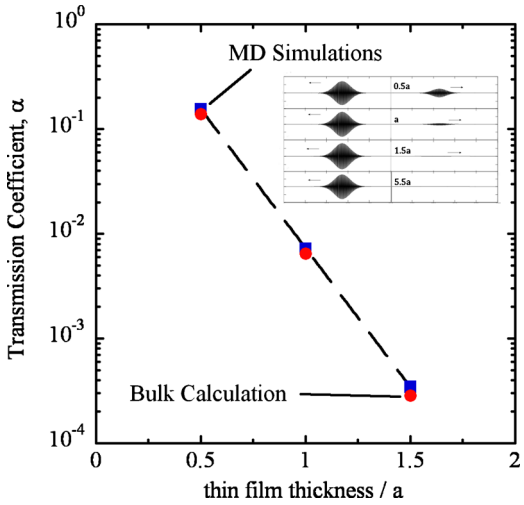

FIG. 3. (Color online) Phonon tunneling across thin films of various thicknesses for a wave packet of $k_0=0.5(2\pi/a)$. Corresponding snapshots of phonon displacements for film thickness of $0.5a$, a, $1.5a$, and $5.5a$ are shown in the upper right inset.

evanescent wave based transfer of energy across the thin film is often termed phonon tunneling.[12–14] If the thin film is treated as a bulk solid, its complex dispersion relationship can be calculated from the dynamical matrix. In Fig. 3, we show the calculated transmission coefficient for a wave packet determined by matching plane wave boundary conditions at the thin film/bulk solid interfaces with a complex wave vector calculated assuming the thin film as a bulk solid. In this case, with a thin film to bulk solid mass ratio of four, the calculated results are in excellent agreement with MD generated results. For $d>2a$, almost no phonon is transmitted across the thin film. Thus, we directly simulate the effect of phonon wave-packet tunneling within the MD framework.

Figure 4 shows MD simulated results for phonon tunneling across pure and composite thin films of various mass values for a wave packet of $k_0=0.5(2\pi/a)$ and film thickness of $0.5a$. We also show the expected transmission coefficient calculated assuming the thin film has the complex dispersion relationship of a bulk solid. Composite films containing a mixture of $m=3$ and 5 atoms are created through random mixtures of the atoms with different masses. These are simulated for the technologically relevant reason that interfaces between solids are not typically ideal but often provide some mixing at the atomic scale.

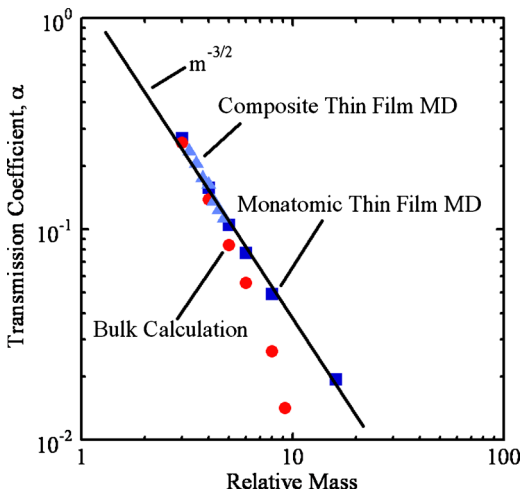

FIG. 4. (Color online) Phonon tunneling across pure and composite thin films of various mass values for a wave packet of $k_0=0.5(2\pi/a)$ and film thickness of $d=0.5a$. Composite films containing a mixture of $m=3$ and 5 atoms (where $m=1$ is for the bulk material) are created by randomly changing $m=3$ with 5 atoms.

The MD determined high frequency transmission coefficient as a function of mass is best described as a power law for mass ratios between three and sixteen, with the transmission coefficient being proportional to $m^{-3/2}$. The observed transmission coefficients for the composite films fall on the same curve. The calculated transmission coefficients based on the bulk complex dispersion curve show good agreement with the MD simulated results up to mass ratios of approximately six. However, the agreement becomes quite poor at mass ratios of eight where the calculated transmission coefficient differs from the simulated result by a factor of two. We note here that for the LJ parameters used in this work, and the phonon wave-packet frequencies considered, the bulk complex dispersion curves obtained for mass ratios beyond approximately 9.25 result in imaginary eigenfrequencies. In the bulk solid, we believe this is due to the large imaginary wave vectors required to achieve the desired wave-packet frequency. At these imaginary wave vectors, the wave-packet decays in a distance less than the solid's interplanar spacing and the concept of a travelling wave becomes nonphysical. Thus, the approximation of treating the thin film as a bulk material clearly breaks down in this mass ratio regime.

In this paper, we have demonstrated the use of the MD-based phonon wave-packet technique to study phonon wave-packet interference and tunneling across nanostructured thin films. The results show good agreement with the AMM theory in the low-frequency limit. At high frequencies, energy transport through the thin films occurs predominantly through evanescent waves. In both frequency regimes, treating the thin film as a bulk solid produces calculated results that are in excellent agreement with simulated results for mass ratios less than six.

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