Influence of crystallographic orientation and anisotropy on Kapitza conductance via classical molecular dynamics simulations


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We investigate the influence of crystallographic orientation and anisotropy on local phonon density of states, phonon transmissivity, and Kapitza conductance at interfaces between Lennard-Jones solids via classical molecular dynamics simulations. In agreement with prior works, we find that the Kapitza conductance at an interface between two face-centered cubic materials is independent of crystallographic orientation. On the other hand, at an interface between a face-centered cubic material and a tetragonal material, the Kapitza conductance is strongly dependent on the relative orientation of the tetragonal material, albeit this dependence is subject to the overlap in vibrational spectra of the cubic and tetragonal materials. Furthermore, we show that interactions between acoustic phonons in the cubic material and optical phonons in the tetragonal material can lead to the interface exhibiting greater “thermal anisotropy” as compared to that of the constituent materials. Finally, it is noted that the relative match or mismatch between the Debye temperatures of two materials comprising an interface does not serve an accurate gauge of the efficiency of interfacial thermal transport when those materials have different crystal structures.

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I. INTRODUCTION

The thermal transport properties of modern devices are now dictated more so by the interfaces between materials than they are by the materials themselves.1 In response, a significant effort has been put forth to both investigate and explain the behavior of thermal transport across solid-solid interfaces, the efficiency of which can be described by the interface Kapitza conductance, 2 or $h_k$. For example, prior works have demonstrated how the mismatch of phonon spectra,3–7 interfacial structure,8–14 and interfacial chemistry15–21 can each impact conductance. However, despite the many advances within this rapidly growing body of work, relatively few studies have focused on the influence of crystallographic orientation and anisotropy on Kapitza conductance at solid-solid interfaces.

In nanostructured material systems, thermal transport properties can exhibit high degrees of anisotropy due to both geometric and material factors. With regard to the former, reduced characteristic lengths can lead to increased anisotropy through boundary scattering rates that vary based on direction22–26 and through changes in the phonon spectra.24,27 As for the latter, material factors (i.e., thermal anisotropy inherent to the materials comprising the interface) have been less frequently studied. Theoretical28,29 and experimental30 investigations of Kapitza conductance at metal:carbon-nanotube and metal:graphite interfaces have indicated that conductance can vary by an order of magnitude depending on the orientation of the nanotube or graphite; this behavior has been attributed to phonon group velocities that vary with crystallographic direction. On the other hand, Costescu et al.8 measured the conductance of epitaxially grown TiN:MgO(001) and TiN:MgO(111) interfaces via time-domain thermoreflectance (TDTR) and reported no dependence on crystallographic orientation. Similarly, Hopkins et al.31 measured Kapitza conductance at Al:Si(001) and Al:Si(111) interfaces via TDTR and likewise reported no orientational dependence. However, in that same study, conductance at Al:Al2O3(0001) and Al:Al2O3(1120) interfaces differed by up to 40%. As with the earlier metal:nanotube and metal:graphite studies, this effect was attributed to directionally dependent phonon group velocities.

In this paper, we report on the influence of crystallographic orientation and anisotropy on Kapitza conductance at interfaces between Lennard-Jones (LJ) solids via classical molecular dynamics (MD) simulations. In agreement with the aforementioned studies, we find that the Kapitza conductance at an interface between two cubic materials (face-centered cubic or fcc type) is independent of crystallographic orientation, while at an interface between a fcc material and a tetragonal material (L10 type) the Kapitza conductance is strongly dependent on the relative orientation of the L10 material. In contrast with earlier works, this observed effect is
not attributed to directionally dependent group velocities, as the magnitudes of the group velocities in the L10 material do not vary significantly with crystallographic direction. To gain further insight into this observed behavior, we spatially resolve the phonon density of states (DOS) near the interface and determine frequency-dependent phonon transmissivities via wave-packet simulations. Briefly, it is found that the interactions between acoustic phonons in a fcc material and the optical phonons in a L10 material can dramatically enhance thermal anisotropy at the interface beyond that inherent to the constituent materials.

The remainder of this paper will be organized as follows: Sec. II will detail the Lennard-Jones solids considered, including their bulk vibrational properties as calculated via harmonic lattice dynamics (LD); Sec. III will report on the thermal conductivities of the constituent materials and the Kapitza conductances at interfaces between them as determined via non-equilibrium molecular dynamics (NEMD) simulations; Sec. IV will discuss the results in the context of local phonon density of states calculations and wave-packet simulations at interfaces between fcc and L10 solids; finally, Sec. V will provide a summary of major results.

II. PROPERTIES OF CONSTITUENT MATERIALS

We considered three different materials which we will refer to as material A, material B, and the AB alloy. All interatomic interactions were described by the 6–12 LJ potential, \( U(r) = 4\varepsilon[(\sigma/r)^{12} - (\sigma/r)^{6}] \), where \( U \) is potential energy, \( r \) is the interatomic separation, and \( \sigma \) and \( \varepsilon \) are the LJ length and energy parameters, respectively. To reinforce the generality of this work (our objective is not to extract quantitative data for specific interfaces but rather to discover the dependence of Kapitza conductance on interfacial configuration), we present all relevant quantities in non-dimensional LJ units (see Table I for the nondimensionalization scheme).\(^5\)\(^,\)\(^18\) The LJ potential was parameterized identically for each material,\(^32\) and the cut-off distance was set to \( r_c = 2.5\sigma \). Consequently, all materials had the same zero-temperature cubic lattice constant, \( a_{\text{lat}} = 1.55 \), and thus differed only by atomic mass and arrangement. Materials A and B were fcc solids comprised of a single type of atom with a unique mass; \( m_A = 40 \text{ amu} \) and \( m_B = 120 \text{ amu} \). The AB alloy contained both types of atoms in the layered-tetragonal L10 configuration (see Fig. 1). In this structure, an A atom occupies the atomic site at \( (0, 0, 0) \) and a B atom occupies the site at \( (0.5, 0.5, 0.5) \) relative to the primitive vectors of the tetragonal unit cell. This atomic arrangement forms a \( 1 \times 1 \) superlattice in the [001] crystallographic direction, or along the \( c \)-axis.

The bulk phonon DOS and phonon dispersion relations of materials A, B, and the AB alloy were determined via harmonic lattice dynamics calculations. These calculations were carried out with the General Utility Lattice Program (GULP),\(^33\) the results of which are shown in Fig. 2. In the top panel, the phonon DOS is plotted as a function of angular frequency, \( \omega \). The DOS of materials A and B exhibits the characteristic shape associated with fcc materials, and their cutoff frequencies differ by a factor of \( \sqrt{m_B/m_A} = 2.3 \). The shape of the DOS of the AB alloy differs greatly from that of materials A and B, with peaks centered at \( \omega \approx 8.5 \) and 20.

The phonon dispersion relations in selected directions of high symmetry of materials A, B, and the AB alloy at zero temperature are plotted in the lower three panels of Fig. 2.

### TABLE I. Non-dimensional (LJ-reduced) scheme. All quantities were normalized such that \( m_B = m_A = 40 \text{ amu} \). Throughout the text, the asterisks are dropped and non-dimensional quantities are implied.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Non-dimensional</th>
</tr>
</thead>
<tbody>
<tr>
<td>Length, ( r )</td>
<td>( r^* = r \times \sigma^{-1} )</td>
</tr>
<tr>
<td>Temperature, ( T )</td>
<td>( T^* = T \times k_B \varepsilon^{-1} )</td>
</tr>
<tr>
<td>Angular frequency, ( \omega )</td>
<td>( \omega^* = \omega \times \sigma \times \sigma^6 \varepsilon^{-0.5} )</td>
</tr>
<tr>
<td>Thermal flux, ( Q )</td>
<td>( Q^* = Q \times \sigma^3 \varepsilon^{-1.5} )</td>
</tr>
<tr>
<td>Thermal conductivity, ( \kappa )</td>
<td>( \kappa^* = \kappa \times \sigma^2 \varepsilon^{-0.5} \times k_B^{-1} \varepsilon^{-0.5} )</td>
</tr>
<tr>
<td>Kapitza conductance, ( h_K )</td>
<td>( h_{K}^* = h_K \times \sigma^3 \varepsilon^{-0.5} \times k_B^{-1} \varepsilon^{-0.5} )</td>
</tr>
</tbody>
</table>

FIG. 1. Schematics of the (a) tetragonal primitive unit cell and (b) cubic conventional unit cell of the layered-tetragonal L10 AB alloy.

FIG. 2. Bulk DOS (top) and phonon dispersion relations (bottom) in selected directions of high symmetry of materials A, B, and the AB alloy as calculated via harmonic lattice dynamics. The wavevector has been normalized to \( 2\pi/a_{\text{lat}} \), where \( a_{\text{lat}} \) is the lattice constant of the cubic conventional cell. Note that the slopes of the dispersion curves, i.e., phonon group velocities, of the AB alloy in the in-plane and cross-plane directions do not differ significantly.
all three panels, the wavevector has been normalized by $2\pi/a_{\text{AB}}$. Note that the maximum wavevector of the AB alloy in the cross-plane direction ($cp$) is half of that along the in-plane direction ($ip$, or [0.5, 0.5, 0] relative to the primitive vectors of the tetragonal unit cell). Both the DOS and dispersion relations demonstrate that while the vibrational spectra of material A and the AB alloy better match one another in terms of their cutoff frequencies (and hence, Debye temperatures), material B and the AB alloy exhibit better spectral overlap at frequencies below $\omega = 10$. In addition, it is important to note that the phonon group velocities, $v_g = |\partial\omega/\partial k|$, do not differ significantly from the in-plane to cross-plane direction in the AB alloy.

### III. NON-EQUILIBRIUM MOLECULAR DYNAMICS

Classical molecular dynamics simulations are well suited to study thermal transport across interfaces in semiconductor systems as the time-evolution of the positions and energies of atoms around an interface are exactly known. For example, a standard steady-state NEMD simulation can be set up to investigate either the thermal conductivity of a material, $\kappa$, or the Kapitza conductance at the interface between two different materials, $h_{\kappa}$. In either case, a thermal flux, $Q$, is applied across a computational domain in order to establish a steady-state spatial temperature gradient, $\nabla T$. If the thermal conductivity of a single material is sought, the observed spatial temperature gradient can be related to the thermal conductivity by invoking the Fourier law, $Q = \kappa \nabla T$. As for Kapitza conductance, differences in vibrational properties will lead to a temperature discontinuity at the interface between two materials when a flux is applied. This discontinuity, $\Delta T$, is related to conductance through the relationship $Q = h_{\kappa} \Delta T$. In the present study, the thermal conductivities of materials A, B, and the AB alloy, as well as the Kapitza conductances at various interfaces between them, were calculated via NEMD simulations at temperatures from 0.09 to 0.42 (the melt temperature of LJ solids is $T_m \approx 0.5$ (Ref. 5)). The computational domains implemented when calculating conductivities and conductances were rectangular cuboids elongated in the $z$-direction with periodic boundary conditions applied in the $x$- and $y$-directions, thus taking on the general characteristics associated with many previous NEMD simulations. The four outermost monolayers in the positive and negative $z$-directions formed rigid walls. The eight monolayers immediately inside these walls were bath atoms, to which energy would be added or removed to establish a temperature gradient. Further specifics regarding the domains will be discussed in subsequent sections.

During the simulation, the equations of motion for the system were integrated using the Nordsieck fifth-order predictor-corrector algorithm with a time step of 0.002. The systems were first equilibrated at a predefined temperature via a velocity scaling routine and zero pressure, maintained by the Berendsen barostat algorithm. In this routine, pressure was calculated for all dynamic atoms in the system and the volume scaling involved displacement of the rigid walls. Subsequently, the NEMD procedure was implemented. The addition of energy to/removed from the baths was performed through a constant-flux approach. This routine slightly changes the forces acting on a particular atom depending on the amount of energy to be added or removed. The total force acting on atom $i$ is given by

$$F_{i,\text{total}} = F_i + \zeta m_i v_i^T,$$

where $m_i$ is the mass of the atom, $v_i^T$ is the thermal velocity of the atom, and $\zeta$ is a scaling factor. This scaling factor is expressed as

$$\zeta = Q \frac{1}{2\kappa^2} = \frac{\Delta E}{2\Delta T},$$

where $Q$ is the flux across the domain, $\Delta T$ is the time step used in the MD integration of the equations of motion, $\Delta E$ is the amount of energy to be added to or removed from the bath per time step, and $K^T$ is the total thermal kinetic energy of the bath. For thermal conductivity and Kapitza conductance calculations, the applied flux varied between $7.95 \times 10^{-5}$ and $15.9 \times 10^{-5}$ depending on the overall thermal resistivity of the particular domain of interest (in order to make the change in temperature small but significant relative to the noise).

During NEMD, the systems were divided into equally sized bins such that the temperature of each monolayer could be calculated, thus allowing for the construction of spatial temperature profiles along the $z$-axis. Once in steady-state (as determined by monitoring the temporal evolution of the bath temperatures), time-averaged profiles were constructed from 3500 system snapshots taken over $1.75 \times 10^6$ time steps. Linear least-squares fits of these temperature profiles were then performed (separately for each half of the domain in the Kapitza conductance simulations). In certain cases, some non-linearity in the temperature profiles was observed near the baths and/or the interface; these data were not included in the fits. The slopes of these fits were used to calculate the thermal conductivity; the discontinuities between fits at the interfaces were used to calculate Kapitza conductance. When calculating Kapitza conductance, it was ensured that the thermal conductivities of each half of the domain were in agreement with the thermal conductivities of the respective materials as determined from separate thermal conductivity simulations of larger, homogenous domains in which bulk properties could be reproduced. Finally, to ensure that our results were independent of the direction of applied heat flux, additional simulations were conducted with this direction reversed; no systematic dependence was observed.

### A. Thermal conductivity

The thermal conductivities of materials A and B were calculated along the [001] direction and the thermal conductivity of the AB alloy was calculated in both the in-plane and cross-plane directions (the thermal conductivity tensors of materials A and B are isotropic due to their cubic symmetry). Two pre-equilibrated domain sizes were implemented to ensure size effects did not distort results;
12.4 × 12.4 × 93 and 12.4 × 12.4 × 124. These domain lengths, \( L_z \), correspond to 60 and 80 conventional unit cells, respectively. The thermal conductivity of material A as calculated in domains of each size is plotted in Fig. 3. As is evident from the plot, this change in domain length did not produce any systematic difference in the calculated values of thermal conductivity, suggesting finite size effects did not distort our results. Regardless, the \( L_z = 124 \) length was used in all subsequent simulations in order to generate more data points over which to fit when calculating the spatial temperature gradients.

Results of our thermal conductivity simulations are plotted as a function of temperature in Fig. 3. Each data point is the mean result of three independent simulations; standard deviations were less than 6% of the mean value in all cases. The thermal conductivities of the materials exhibit the inverse temperature dependence attributed to phonon-phonon scattering.\(^{27,40}\) This temperature dependence is illustrated by the inverse power fits of the NEMD data represented by the dashed lines in Fig. 3. Across the entire temperature range, the thermal conductivity of the AB alloy in the in-plane direction is approximately a factor of two higher than in the cross-plane direction. This difference is due to the fact that the Brillouin zone of the AB alloy is narrower by a factor of two in the cross-plane direction as compared to the in-plane direction. As a result, the maximum wavevector of phonons traveling along that axis is half that of phonons traveling in the in-plane direction, and thus, the likelihood of Umklapp scattering is higher in the cross-plane direction as compared to that in the in-plane direction.\(^{41,42}\) While the formation of a superlattice typically leads to band flattening and lower phonon group velocities, the dispersion curves in Fig. 2 indicate that directionally dependent group velocities alone cannot explain the conductivity trends of the AB alloy.

### B. Kapitza conductance

Kapitza conductances were calculated at six different interfaces via NEMD simulations; the specifics of these interfaces and the computational domains are listed in Table II. Note that the (111) orientation of sample II prevented a square cross section and thus prescribed different overall dimensions; still, the number of atoms contained in that domain was within 4% of that in the others. The calculated conductances are plotted as a function of temperature in Fig. 4. Each data point represents the mean result of five independent simulations (with the exception of the hollow symbols plotted in Fig. 4(b), which each represent the result of a single simulation) and error bars represent the standard deviation of these five simulations, i.e., repeatability. In some cases, the standard deviation is small enough that the error bars are masked by the data point itself. In Figs. 4(b) and 4(c), the dashed lines are linear fits of the NEMD data.

Samples I and II contained interfaces between materials A and B and differed in terms of their crystallographic orientation; sample I was oriented along the [001] crystallographic direction, whereas sample II was oriented along the [111] direction. While their sizes do differ slightly, this discrepancy does not affect our results, as the difference in size between these domains is small relative to that between those which were checked for finite size effects (see Sec. III A). The Kapitza conductances at the interfaces contained within samples I and II are plotted as a function of temperature in Fig. 4(a). Our results support the conclusion of previous experimental work that the Kapitza conductance at interfaces between two cubic crystals does not vary with orientation.\(^{8,31}\) Unique to this data set is the fact that materials A and B are lattice matched, and thus, the interfaces were atomically perfect. In reality, dislocation densities can vary with orientation in systems that are not lattice matched. Still, prior experimental data at TiN/MgO (Ref. 8) and Al/Si (Ref. 31) interfaces exhibit the same orientational independence observed here. Though dislocation densities at those interfaces did likely vary with orientation, they did not vary substantially enough to affect Kapitza conductance (Ref. 11 suggests it can take several order-of-magnitude changes in dislocation density to vary Kapitza conductance by a factor of two).

Samples III through VI contained interfaces between a cubic material oriented along the [001] direction (either A or B) and the AB alloy oriented in the in-plane or

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**TABLE II.** The pre-equilibrated dimensions, constituent materials, and crystallographic orientations of the computational domains employed in the Kapitza conductance simulations. In all cases, the cold bath was at the end of material 1 and the hot bath was at the end of material 2, with the exception of the hollow symbols plotted in Fig. 4(b), for which the heat flux direction was reversed.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Dimensions</th>
<th>Material 1</th>
<th>Material 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>12.4 × 12.4 × 124</td>
<td>fcc A (001)</td>
<td>fcc B (001)</td>
</tr>
<tr>
<td>II</td>
<td>12.0 × 13.3 × 123</td>
<td>fcc A (111)</td>
<td>fcc B (111)</td>
</tr>
<tr>
<td>III</td>
<td>12.4 × 12.4 × 124</td>
<td>fcc A (001)</td>
<td>L1_0 AB (ip)</td>
</tr>
<tr>
<td>IV</td>
<td>12.4 × 12.4 × 124</td>
<td>fcc A (001)</td>
<td>L1_0 AB (cp)</td>
</tr>
<tr>
<td>V</td>
<td>12.4 × 12.4 × 124</td>
<td>fcc B (001)</td>
<td>L1_0 AB (ip)</td>
</tr>
<tr>
<td>VI</td>
<td>12.4 × 12.4 × 124</td>
<td>fcc B (001)</td>
<td>L1_0 AB (cp)</td>
</tr>
</tbody>
</table>
cross-plane direction; again, all interfaces were atomically perfect. Schematics representing these domains are illustrated in Fig. 5. From the data plotted in Figs. 4(b) and 4(c), it is clear that Kapitza conductance is strongly dependent on the relative orientation of the AB alloy. At interfaces between material A and the AB alloy, Kapitza conductance varies by upwards of a factor of five depending on AB alloy orientation, whereas at interfaces between B and the AB alloy, it varies by a factor of two. To further elucidate this point, we define a thermal anisotropy factor,

\[ \gamma = \frac{K_{ip}}{K_{cp}} \text{ or } \frac{h_{K,ip}}{h_{K,cp}} \]

where the subscripts \( ip \) and \( cp \) refer to the orientation of the AB alloy. In Fig. 6, both the conductivity of the AB alloy and conductance of the B:AB interfaces exhibit nearly identical anisotropy factors; \( \gamma \approx 2 \) across the entire temperature range. On the other hand, the anisotropy factor is not only much higher for the A:AB interfaces, but it also exhibits greater temperature dependence.

IV. DISCUSSION

As detailed in the Introduction, prior works examining the influence of crystallographic orientation and anisotropy on Kapitza conductance have ascribed any observed dependence on orientation to directionally dependent group velocities. While this argument is viable in the case of materials exhibiting large degrees of vibrational and thermal anisotropies, e.g., graphite or carbon nanotubes, the lattice dynamics calculations presented in Sec. II indicate that the phonon group velocities of the L10 AB alloy do not differ substantially with crystallographic direction. In addition, directionally dependent group velocities cannot explain the significant difference in anisotropy between the A:AB and B:AB interfaces. In order to investigate this phenomenon further, we calculate the local phonon density of states and frequency-dependent phonon transmissivity at interfaces corresponding to those contained within samples III through VI.

A. Local phonon density of states

We calculated the local phonon DOS of the bilayer of atoms within the AB alloy immediately adjacent to the interface in samples III through VI. The DOS is proportional to the Fourier transform (\( \mathcal{F} \)) of the velocity correlation function (VACF) but in practice is calculated using standard

\[ \text{FIG. 6. The anisotropy ratio defined by Eq. (3) plotted as a function of temperature. While the anisotropy factor of the B:AB interfaces closely follows that of the conductivity of the AB alloy, the anisotropy factor of the A:AB interfaces is significantly higher while also exhibiting greater temperature dependence.} \]
estimation procedures for power spectral density.\textsuperscript{37} Within each bilayer of interest, the velocity of 32 atoms is obtained at each integration time step to give a velocity fluctuation time series of 73 728 points. The Welch method of power spectral density estimation is then applied by creating eight 50\% overlapping segments of 16 384 points to give an angular frequency resolution of 0.19 based on our time step of 0.002. Each segment is then multiplied by a Hamming window and the fast Fourier transform is computed. The power spectral density, proportional to DOS and equivalent to $S(\omega)$, is then obtained by ensemble averaging the Fourier transform magnitudes of each segment.

From Fig. 2, the optical-type phonons in the AB alloy are “elastically inaccessible” to the acoustic phonons of material B; i.e., no acoustic phonons within material B are at frequencies as high as those of the optical phonons in the AB alloy. On the other hand, the optical phonons in the AB alloy are “elastically accessible” to the acoustic phonons of material A. In Fig. 7, the calculated local phonon DOS of the bilayer of atoms within the AB alloy immediately adjacent to the interface in samples III through VI at $T = 0.09$ are plotted (this low simulated temperature ensures distinct spectral features are observable). Overall, none of the curves exhibit an overwhelming deviation from that of the bulk (and harmonic) phonon DOS plotted in the top panel of Fig. 2, despite the fact that the sampled atoms used to generate these curves were immediately adjacent to an interface. However, in sample IV, the frequency range $18 < \omega < 21$ does exhibit a degree of mode depletion. While this could be expected due to the overlap in vibrational spectra of materials A and the AB alloy, it is interesting that these modes are only depleted for one orientation of the interface. This unique characteristic of the interface within sample IV suggests that interactions between the acoustic phonons of material A and the optical phonons of the AB alloy lead to increases in the thermal anisotropy of A:AB interfaces beyond that of B:AB interfaces, or the AB alloy itself.

### B. Wave-packet simulations

To investigate the frequency dependence of phonon transmissivity, we use the multiple wave-packet method.\textsuperscript{43} For a single wave-packet,\textsuperscript{44} linear combinations of normal modes with different wavevectors $\mathbf{k}$ are used as initial conditions for a molecular dynamics simulation. The normal mode amplitudes for wave-packet $i$ are chosen to be a Gaussian centered on a given central wavevector $\mathbf{k}_0^i$, yielding a wave-packet localized in real space. Moreover, the phases of the modes are chosen such that the packet is initially located at a specified position $\mathbf{R}_0^i$ in real space.

In this work, the propagation direction is along the $z$-axis and localization is accomplished by mixing modes with different $k_z$. Specifically, multiple wave-packets are superposed with packet $i$ containing wavevectors $\mathbf{k} = \mathbf{k}_0^i + (00\delta k_z)$ with normal mode amplitudes $\xi_{\mathbf{k}^i}$ given by

$$\xi_{\mathbf{k}^i} = A^i e^{-\frac{\delta k_z^2}{2}} e^{-i(k_0^i + \delta k_z)z} R_0^i.$$  

(4)

Every wave-packet from phonon branch $\lambda$ has the same energy so that $|A^i|^2$ varies inversely with $\omega_x(k_0^i)^2$ and $\mathbf{R}_0^i$ is chosen so that it begins in the middle of the fcc half of the bicrystal. Allowed wavevectors are reciprocal to the simulation box for the system and the normal modes are from the longitudinal acoustic (LA) branch. Wave-packets propagate along the positive $z$-direction, reaching the interface to either be transmitted to the AB alloy or reflected into the fcc material. By calculating time-dependent normal mode amplitudes at particular simulation times, the phonon transmissivity is obtained. We define this transmissivity to be the ratio of the energy transmitted through the interface in a given range of frequency to the energy incident on the interface in the same frequency range.

These phonon transmissivities at the four interfaces of interest are plotted as a function of angular frequency in Fig. 8. As is evident from the figure, phonon transmissivity is largely insensitive to the relative orientation of the AB alloy at frequencies below $\omega = 12$. On the other hand, phonon transmissivity at A:AB interfaces is strongly dependent on the relative orientation of the AB alloy at frequencies in the range $17 < \omega < 25$, i.e., nearly the same frequency range noted during the discussion of local phonon density of states in Sec. IV A. Again, this is the frequency range in which acoustic phonons within material A are at the same frequencies of optical phonons within the AB alloy. Finally, we note that transmissivity at B:AB interfaces is marginally higher than at A:AB interfaces at frequencies below $\omega = 12$, consistent with the earlier observation that material B and the AB alloy exhibit better spectral overlap across that same frequency range.

These results, in conjunction with the Kapitza conductance data presented in Sec. III B, demonstrate that the observed thermal anisotropy at B:AB interfaces is due almost entirely to that inherent to the AB alloy, whereas the anisotropy at A:AB interfaces is enhanced beyond this baseline (i.e.,

- [Fig. 7](#) The local phonon density of states at $T = 0.09$ within the bilayer of atoms in AB alloy immediately adjacent to the interface in samples (a) III and IV and (b) V and VI; III and V (both ip) are represented by shaded regions, whereas IV and VI (both ip) are represented by solid lines. The frequency range $18 < \omega < 21$ in sample IV exhibits significant mode depletion (as indicated by the bold arrow) as compared to all other samples.
\[
\gamma > 2 \]

due to the dependence of interfacial phonon-phonon interactions on the relative orientation of the AB alloy. In other words, the anisotropy factor defined by Eq. (3) is nearly identical for B:AB Kapitza conductance and AB alloy conductivity (see Fig. 6), despite the fact that phonon transmissivity at B:AB interfaces is independent of the relative orientation of the AB alloy; in turn, the thermal anisotropy of the B:AB interfaces can be ascribed to the thermal anisotropy inherent to the AB alloy. In contrast, the thermal anisotropy of the A:AB interfaces is enhanced beyond that inherent to the AB alloy due to the dependence of phonon transmissivity at A:AB interfaces on the orientation of the AB alloy.

\section*{V. CONCLUSION}

We have studied the influence of crystallographic orientation and anisotropy on Kapitza conductance, local phonon density of states, and phonon transmissivity via classical molecular dynamics simulations. Non-equilibrium molecular dynamics simulations have demonstrated that Kapitza conductance at interfaces between fcc and L10 materials depends on both the relative orientation of the L10 material and the degree of vibrational overlap between the two materials (but not on phonon cutoff frequencies). Local phonon density of states and phonon transmissivity calculations indicate that interactions between acoustic phonons in the fcc material and optical phonons in the L10 material can greatly enhance thermal anisotropy. Furthermore, interfaces may exhibit higher degrees of thermal anisotropy (i.e., a larger dependence on crystallographic orientation) than their constituent materials. Despite the classical nature of these simulations, the results presented provide qualitative insight into the mechanisms responsible for prior experimental observations that Kapitza conductance is critically dependent on crystallographic orientation. Finally, we would like to note that the relative match or mismatch between the cutoff frequencies or Debye temperatures of two materials comprising an interface does not serve an accurate gauge of the efficiency of interfacial thermal transport when those materials have different crystal structures.

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