Molecular dynamics studies of material property effects on thermal boundary conductance

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Thermal boundary resistance (inverse of conductance) between different material layers can dominate the overall thermal resistance in nanostructures and therefore impact the performance of the thermal property limiting nano devices. Because relationships between material properties and thermal boundary conductance have not been fully understood, optimum devices cannot be developed through a rational selection of materials. Here we develop generic interatomic potentials to enable material properties to be continuously varied in extremely large molecular dynamics simulations to explore the dependence of thermal boundary conductance on the characteristic properties of materials such as atomic mass, stiffness, and interfacial crystallography. To ensure that our study is not biased to a particular model, we employ different types of interatomic potentials. In particular, both a Stillinger–Weber potential and a hybrid embedded-atom-method + Stillinger–Weber potential are used to study metal-on-semiconductor compound interfaces, and the results are analyzed considering previous work based upon a Lennard-Jones (LJ) potential. These studies, therefore, reliably provide new understanding of interfacial transport phenomena particularly in terms of effects of material properties on thermal boundary conductance. Our most important finding is that thermal boundary conductance increases with the overlap of the vibrational spectra between metal modes and the acoustic modes of the semiconductor compound, and increasing the metal stiffness causes a continuous shift of the metal modes. As a result, the maximum thermal boundary conductance occurs at an intermediate metal stiffness (best matched to the semiconductor stiffness) that maximizes the overlap of the vibrational modes.

I. Introduction

Unlike uniform materials, interfaces between different materials impose an additional thermal boundary resistance.1,2 Such a resistance is termed Kapitza resistance, and its reciprocal is called Kapitza conductance. Kapitza conductance can dominate the overall thermal resistance in nano systems. This phenomenon can be utilized to increase the figure of merit of thermoelectric materials.3,4 It can also cause local temperature accumulation leading to thermal failures in microelectronic devices.5 As a result, Kapitza resistance is often the performance-limiting factor in nanoscale applications.6,7

Kapitza conductance is extremely sensitive to interfacial structures including crystallinity, crystallographic orientation, roughness, interdiffusion, and chemistry.5,8–13 Without understanding the dependence of Kapitza conductance on structural and chemical details, the design of devices has been relying on a trial-and-error approach rather than a science-based optimization.

In molecular dynamics (MD) simulations, a computational material is represented by an assembly of atoms that mutually interact through a force field (i.e., an interatomic potential). Thermal transport phenomena can then be simulated by solving velocities (and hence temperature) of atoms as a function of time from Newton’s equations of motion under any externally applied heat fluxes. The resulting time evolution of temperature enables both calculation of Kapitza conductance and fundamental analysis on such properties as phonon density of states.14 Because MD methods enable atomic details of interfacial structures to be replicated without imposing constraints on the nature of phonon scattering, they have been successfully applied in recent years to study thermal boundary conductance15–23 and thermal conductivity of superlattices with interfaces,24–26 establishing the effects of size,27 interfacial morphology,14 temperature,15,20,21 and interfacial defects.20,21 By changing the relevant parameters of the interatomic potential used in the MD simulations, material properties such as atomic mass, lattice constant, cohesive energy (corresponding to bond strength), and elastic constants (corresponding to bond stiffness) can be systematically varied. Hence, MD methods have also been used to study effects of mass differential of the two layers,5,20–22 lattice mismatch between layers,20,21 and stiffness of
the materials and bond strength at the interface.\textsuperscript{16,28} While these studies represent significant progresses towards interfacial engineering, our current understanding of the thermal boundary conductance is far from the material/structure design requirement. In particular, MD simulations are often performed using a given interatomic potential specific to a particular material system. Because different materials are associated with many different properties that can all affect Kapitza conductance, MD results obtained from specific studies cannot usually guide experiments to optimize interfaces through material selections.

Here we use parallel MD codes LAMMPS\textsuperscript{29} to perform extremely large simulations to explore the dependence of thermal boundary conductance on the characteristic properties of materials such as atomic mass, stiffness, and interfacial crystallography. The estimated total computation of the current work is approximately $7.5 \times 10^6$ CPU hour [2.93 GHz processors], which is, apparently, at least two orders of magnitude increase over what is typically used in literature.\textsuperscript{15–21} Our data is collected from super computer cluster Redsky over a span of more than two years. The expanded computations enable us to effectively reduce the statistical errors of the data, which proves to be critical in driving our research direction towards the discovery of the material property effects.

To ensure that our studies are relevant to realistic metal-on-semiconductor compound interfaces, the interatomic potentials used in our simulations are fitted to the reference Al–GaN system. However, these potentials are used as generic potentials in simulations by varying the corresponding parameter from its reference value to enable individual material property to be continuously varied. To ensure that our study is not biased to a particular model, we construct two different types of generic potentials. In particular, we develop both a Stillinger–Weber potential and a hybrid embedded-atom-method + Stillinger–Weber potential for studying M/AB interfaces, where M and AB refer to a face-centered-cubic (fcc) metal (\textit{i.e.}, modified Al) layer and a binary semiconductor wurtzite (wz) compound (\textit{i.e.}, modified GaN), respectively. These potentials are then used to study the thermal boundary conductance through the M/AB interfaces, and the results are analyzed considering previous work using a Lennard-Jones potential.\textsuperscript{28,30} By using different types of interatomic potentials, by independently changing each of the characteristic properties of materials, and by performing extremely large MD simulations to improve the quality of data, our studies will provide new understanding of effects of materials on thermal boundary conductance.

II. Method
A. Interatomic potential

In this work, we develop two interatomic potential models for the Al–Ga–N ternary systems. The first model uses the Stillinger–Weber (SW) potential format,\textsuperscript{31} with essentially the same parameters as the Al–Ga–N potential developed previously\textsuperscript{14} except that the Al–N parameters are slightly modified so that the new potential reproduces exactly the experimental cohesive energy for AlN. This modification should not affect the results obtained in previous studies of the Al/GaN interfaces\textsuperscript{14,27} where Al was assumed to be in contact with Ga at the interface and no AlN was encountered. This refinement, however, should enable the potential to be used for other studies where AlN may be encountered. For convenience, a complete list of the Al–Ga–N SW potential parameters is shown in Table 2 of Appendix A.

The second model applies a hybrid embedded-atom-method (EAM) + SW potential format. For convenience, this potential is termed ESW potential. Here the elemental Al is described by a well-established Al EAM potential,\textsuperscript{12} GaN is represented by a literature Ga–N SW potential\textsuperscript{13,34} (which is also the same as the Ga–N portion of the Al–Ga–N SW potential), and the cross interactions between Al and Ga and between Al and N are defined by a Morse potential. Eight parameters, $r_0_{AlGa}$, $E_{AlGa}$, $r_0_{AlN}$, $E_{AlN}$, $r_0_{AIN}$, $E_{AIN}$, $r_{AlGa}$, and $r_{AlN}$, are needed for the Morse potential. To simplify the comparison between SW and ESW potentials, the parameters of the Morse potential are fitted to give the same equilibrium bond lengths, bond energies, and the second derivatives of the bond energy of the cross interactions as those by the Al–Ga–N SW potential. These Morse parameters of the ESW potential are listed in Table 3 of Appendix A.

The cohesive energies, lattice constants, and elastic constants predicted by the Al–Ga–N SW and ESW potentials are shown respectively in Tables 4–6 of Appendix A. Note that with the Al interactions defined by EAM, it is impossible for the ESW potential to predict a stable AlN wurtzite. As a result, ESW predictions of the AlN phases are not included in Tables 4–6. The inconsistent properties of the “AlN” phases predicted by the two potentials do not affect the comparisons as these phases are not encountered in our simulations. Table 4 verifies that both the SW and ESW potentials correctly predict the fcc Al, wz GaN, and wz AlN to be the lowest energy phases as compared with other possible structures. The predicted lattice constants and cohesive energies of these lowest energy phases are also in good agreement with the corresponding experimental values. In particular, the SW potential reproduces exactly the experimental cohesive energy of AlN, which improves over the previous model.\textsuperscript{14} Tables 4–6 of Appendix A also indicate that while the SW and ESW potentials predict similar properties of the lowest energy phases of Al, Ga, GaN, they give dramatically different elastic constants (stiffness) for Al. This characteristic provides a convenient path to explore the effects of metal elastic constants.

To use the Al–Ga–N SW potential for simulating a generic M-on-AB system [Al, Ga, and N correspond to M, A, and B, respectively], the stiffness of individual materials are varied by using arbitrary scaling factors $e_{MM}/e_{AlAl}$ or $e_{MM}/e_{GaGa}$ or $e_{MM}/e_{NN}$, and $e_{AB}/e_{MM}$. The stiffness between materials across the interface are varied by using arbitrary scaling factors $e_{AB}/e_{GaAl}$ and $e_{AB}/e_{NAl}$. These scalings do not change equilibrium bond lengths. We can also vary the lattice constants of the materials by scaling the $\sigma$ parameters. However, changing the lattice constants changes the lattice mismatch, which requires the adjustment of system dimensions to minimize lattice mismatch strains. This introduces additional size scaling effect, and is better left for a future study.
To use the Al–Ga–N ESW potential for simulating a generic M-on-AB system, the stiffness of the metal is varied by multiplying the Al embedding energy and the Al–Al pair energy of the Al EAM potential by a scaling factor \( f_{\text{EAM}} \), the stiffness of the AB compound is varied by using arbitrary scaling factors \( \varepsilon_{\text{AM}}/\varepsilon_{\text{AB,M}} \), \( \sigma_{\text{AB}}/\sigma_{\text{M,N}} \), and \( \varepsilon_{\text{AB}}/\varepsilon_{\text{M,AB}} \), and the stiffness between materials across the interface are varied by using arbitrary scaling factors \( E_{\text{B,AM}}/E_{\text{B,GAaN}} \) and \( E_{\text{B,BD}}/E_{\text{B,MAC}} \). These scalings do not change bond lengths. Again the lattice constants can be continuously changed with the ESW potential but are not explored here.

B. Direct method molecular dynamics model

1. M/AB interfaces (SW and ESW potentials). Strictly speaking, thorough studies of material property effects require the development of different interatomic potentials to model various combinations of crystal structures of the two materials bounding the interface. This is clearly a collective effort in literature and is beyond the scope of the present work. Here we focus on interfaces between an fcc metal “M” and a wurtzite semiconductor compound “AB”. As a result, we cannot directly study different crystal structures. However, we explore different crystallographic matches at the interface, corresponding respectively to three fcc metal planes \{100\}, \{110\}, and \{111\} on a given wurtzite semiconductor compound \{0001\} plane. Different combinations of crystallographic planes at the interface may provide insights that can be later used to elucidate the phenomena caused by crystal structures.

The orientations and dimensions of the computational crystals used in the direct method MD simulations are shown in Table 1 for the \{100\}M/{0001}AB, \{110\}M/{0001}AB, and \{111\}M/{0001}AB interfaces. Here the heat flux is assumed to be in the x-direction from the fcc metal at the left side to the wurtzite AB compound at the right side. The wurtzite AB is always oriented with \{0001\} parallel to the x-direction, \{110\} parallel to the y-direction, and \{112\} parallel to the z-direction. For the \{100\}M/{0001}AB interface, the fcc metal is oriented with [100] parallel to the x-direction, [010] parallel to the y-direction, and [001] parallel to the z-direction. For the \{110\}M/{0001}AB interface, the fcc metal is oriented with [110] parallel to the x-direction, [010] parallel to the y-direction, and [001] parallel to the z-direction. For the \{111\}M/{0001}AB interface, the fcc metal is oriented with [111] parallel to the x-direction, [112] parallel to the y-direction, and [110] parallel to the z-direction. The \{0001\} \((x\%)\) direction of an AB wurtzite crystal is stacked with alternating A and B planes of alternating short and large spacings.14 Because the lower energy interface always lies between the larger plane spacing, our model assumes that M is in contact with A at the interface and the next B plane has a short spacing with the contact A.

The length, width, and thickness are represented respectively by \( L_{\text{fcc}} \), \( W_{\text{fcc}} \), and \( t_{\text{fcc}} \) for the metal and \( L_{\text{wz}} \), \( W_{\text{wz}} \), and \( t_{\text{wz}} \) for the AB compound. For convenience, the dimensions are also represented by number of cells in the x-, y-, and z-directions as \( n_x, f_{\text{fcc}} \), \( n_y, f_{\text{fcc}} \), \( n_z, f_{\text{fcc}} \) for the metal and \( n_x, w_{\text{wz}} \), \( n_y, w_{\text{wz}} \), \( n_z, w_{\text{wz}} \) for the compound. Our fcc metal has a lattice constant of \( a_{\text{M}} = 4.05 \text{ Å} \), and our wurtzite AB compound have lattice constants of \( a_{\text{AB}} = 3.19 \text{ Å} \) and \( c_{\text{AB}} = 5.20 \text{ Å} \). The smallest orthogonal cell of the

![Diagram](https://example.com/diagram.png)

**Table 1** Computational geometries of the three M/AB interfaces studied

<table>
<thead>
<tr>
<th>Interface</th>
<th>( L_{\text{fcc}} \times t_{\text{fcc}} \times W_{\text{fcc}} ) (( \text{Å}^3 ))</th>
<th>( L_{\text{wz}} \times t_{\text{wz}} \times W_{\text{wz}} ) (( \text{Å}^3 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>{100}M/{0001}AB</td>
<td>( 97 \times 61 \times 45 )</td>
<td>( 146 \times 61 \times 45 )</td>
</tr>
<tr>
<td></td>
<td>( 24 \times 15 \times 11 )</td>
<td>( 28 \times 11 \times 14 )</td>
</tr>
<tr>
<td>{110}M/{0001}AB</td>
<td>( 103 \times 61 \times 29 )</td>
<td>( 146 \times 61 \times 29 )</td>
</tr>
<tr>
<td></td>
<td>( 36 \times 15 \times 10 )</td>
<td>( 28 \times 11 \times 9 )</td>
</tr>
<tr>
<td>{111}M/{0001}AB</td>
<td>( 98 \times 144 \times 29 )</td>
<td>( 146 \times 144 \times 29 )</td>
</tr>
<tr>
<td></td>
<td>( 14 \times 29 \times 10 )</td>
<td>( 28 \times 26 \times 9 )</td>
</tr>
</tbody>
</table>
metal in the (100), (110), (111), and (112) directions are respectively $a_{(100),M} = a_M$, $a_{(110),M} = 0.5\sqrt{2}a_M$, $a_{(111),M} = \sqrt{3}a_M$ and $a_{(112),M} = 0.5\sqrt{6}a_M$. The smallest orthogonal cell of the AB compound in the (0001), (1100), and (1120) directions are respectively $a_{(0001),AB} = a_{AB}$, $a_{(1100),AB} = 2\cos(\pi/6)a_{AB}$, and $a_{(1120),AB} = a_{AB}$. The absolute value of the length and the number of cells are related by these smallest cells. As discovered from the previous work, the cross-section area does not affect the results. With the constraint of a low lattice mismatch strains between the metal and the compound, namely $W_{\text{fcc}} \approx W_i$, and $t_{\text{fcc}} \approx t_i$, the cross-section dimensions as shown in Table 1 are minimized for reducing the computational cost. In particular, these dimensions give y- and z-mismatch strains of 0.0005 and 0.0025 respectively for the (100)/M/(0001)AB interface, 0.0005 and 0.0025 for the (110) M/(0001)AB interface, and 0.0013 and 0.0025 respectively for the (110)/M/(0001)AB interface. The previous work also indicates that the thermal boundary resistance linearly scales with the inverse of sample length. With this understood, we explore a fixed sample length. As shown in Table 1, all the three interfaces use a metal length of about 100 Å and a compound length of about 146 Å. This choice of sample length does not affect the trends of the results. More importantly, the length scale is relevant to practical applications where each layer of the multi-layered devices can be as thin as a few nanometers. With such dimensions, the (100)/M/(0001)AB, (110)/M/(0001)AB, and (111)/M/(0001)AB interfaces shown in Table 1 contain 50,336, 32,976, and 76,776 atoms respectively.

The computational crystal with the desired dimension is first created based on the 0 K temperature lattice constants of the metal and the AB compound crystals. Appropriate strains in the cross-section directions are applied between the metal and the compound to compensate for the lattice mismatch. To incorporate the thermal expansion effect, a pre-conditioning MD simulation is performed at the simulated temperature (via velocity rescaling) for a total of 20 ps under the periodic boundary conditions in all three coordinate directions using a zero-pressure NPT (constant number of atoms, pressure, and temperature) ensemble. After discarding the first 10 ps simulations, the average dimensions obtained during the last 10 ps are then used to create a new crystal with the thermal expansion effect incorporated. This crystal is used in the main MD thermal transport simulation.

In the main MD simulation, periodic boundary conditions are used in the y- and z-directions such that simulated systems approximate an infinite cross-section area. As in the previous work, free boundary conditions with the fixed ends are used in the x-direction. In particular, one atomic metal layer at the left end and a pair of atomic A and B layers at the right end are held fixed during simulations. MD is then first run for 10 ps under the NVT (constant number of atoms, volume, and temperature) condition using velocity rescaling to set up the desired system temperature $T$. MD is then continued with an NVE (constant number of atoms, volume, and energy) condition while at the same time a constant heat flux is introduced. Typically a heat flux of $j = 0.00012$ eV ps$^{-1}$ Å$^{-2}$ is used. First, about 30 Å long materials immediately beside the two fixed ends, as shown in the dark regions near the left and the right sides in Table 1, are defined as hot (heat source) and cold (heat sink) regions. During simulations, a constant amount of energy is continuously added to the hot region and exactly the same amount of energy is removed from the cold region at each MD time step using velocity rescaling (while preserving linear momentum). This naturally creates a temperature differential when the motion of atoms is solved from Newton’s equations. Our special version of the LAMMPS codes implements finite element calculation capability. To compute the temperature profile, 40 finite element bins are created along the x-direction. A temperature averaged over a designated number of time steps is calculated for each of the bins. To generate extremely precise results, the duration of the NVE simulations is chosen to be at least 110.4 ns (for reference, each Kapitza conductance calculation uses about 7.5 × 10$^4$ CPU hour for the (111)/M/(0001)AB interface). After discarding the first 0.4 ns of simulation to allow the system to reach a dynamic equilibrium, the temperature profile averaged over the remaining 110 ns is used for analysis. While we will also explore effects of temperature up to 500 K, we choose to focus on studies at 300 K. Although 300 K does not ensure to be above Debye temperature for the generic materials explored here, we emphasize that our objective is not to extract quantitative data for specific materials but rather to discover material property effects. The advantages of low systematic/statistic errors due to low temperature simulations are critical for this purpose.

III. Results

The direct method MD model is used to compute thermal boundary conductance. As described above, our objective is to understand the effects of material properties through studies at a given temperature of 300 K. To assess the generality of our results, however, simulations are first performed for all the three M/AB interfaces at five different temperatures 100 K, 200 K, 300 K, 400 K, and 500 K. The basis Al-Ga-N SW potential is used directly without scaling. Examples of temperature profiles across the interface obtained at a heat flux of $j = 0.00012$ eV ps$^{-1}$ Å$^{-2}$ and three different temperatures (100 K, 300 K, 500 K) are shown in Fig. 1 for the three interfaces. As expected from the typical interfacial resistance phenomenon, Fig. 1 shows a large abrupt temperature change at the M/AB interface whereas the temperature changes within both the metal region at the left and the AB region at the right are continuous and relatively very small (e.g., approximately 0.005 K Å$^{-1}$ in the GaN layer). The temperature gradient within the GaN layer is much smaller than the 0.0265 K Å$^{-1}$ value obtained from direct method MD simulation of pure GaN at a much higher heat flux of $j = 0.0015$ eV ps$^{-1}$ Å$^{-2}$. Here the use of a smaller heat flux is necessary or the temperature changes across the interface will be unrealistically higher. Because the temperature gradient appears in the denominator of the thermal conductivity expression, we cannot derive accurate thermal conductivities of the metal and the GaN layers from our simulations.
From the temperature profiles, Kapitza conductance $h$ is calculated as $h = J/D\Delta T$, where $J$ is the heat flux and $\Delta T$ is the abrupt temperature change. For all the three interfaces, the abrupt temperature change decreases as temperature is increased, suggesting that the Kapitza conductance increases with temperature. A similar approach to that described previously was used to estimate the standard deviation of the Kapitza conductance.

The calculated conductance values are shown in Fig. 2 as a function of temperature for all the three interfaces. It can be seen that the statistical errors of our data are extremely small. This verifies that our averaging time of 110 ns, which is more than an order of magnitude increase over those commonly used for MD simulations, has effectively reduced the error to such a point that we can confidently deduce effects of material properties.

Fig. 2 indicates that the Kapitza conductance monotonically increases with temperature, which has been ascribed to the increasing contribution of anharmonic phonon–phonon interactions at elevated temperatures. Interestingly, while the Kapitza conductance is seen to linearly increase with temperature for all the three crystallographic orientations at higher temperature, the relationship becomes non-linear for the $\{100\}M/(0001)\text{AB}$ and $\{111\}M/(0001)\text{AB}$ interfaces at low temperatures. This behavior has also been observed for SiGe. The linear dependence on temperature has also been observed for Lennard-Jones system.

A. Effects of atomic mass

Atomic mass is an obvious characteristic property of materials which affects phonons and their density of states. To explore effects of relative atomic mass on Kapitza conductance, simulations are performed using different atomic masses for the metal layer whereas the atomic masses for the A and B species of the AB compound layer are held constant. Again the basis Al–Ga–N SW potential is used without scaling. The calculated Kapitza conductance values are shown in Fig. 3 as a function of atomic mass of the metal species for the three interfaces.

Fig. 3 indicates that Kapitza conductance depends on atomic masses. In particular, when the atomic masses of A and B are fixed at 69.72 and 14.01 amu respectively (i.e., $A = \text{Ga}$ and $B = \text{N}$), there is a maximum Kapitza conductance at an optimal metal atomic mass between 60 and 80 amu for the $\{111\}M/(0001)\text{AB}$ and the $\{100\}M/(0001)\text{AB}$ interfaces and between 60 and 120 amu for the $\{110\}M/(0001)\text{AB}$ interface. The optimal metal atomic mass seems to closely match the atomic mass of Ga (~69.72 amu). With the notion that metal directly contacts Ga in our systems, our result is consistent with the previous finding from MD simulations of an A/B interface using a Lennard-Jones potential that Kapitza conductance reaches maximum when the atomic masses of A and B are equal.

To understand the mass dependence described above, we calculate the phonon density of states for SW Al, SW “heavy Al”, and GaN with the General Utility Lattice Program (GULP). The results of this calculation are presented in Fig. 4. It can be seen that by increasing the metal atomic mass to about 60–80 amu range, the vibrational spectra is shifted towards lower frequencies (which scales approximately with the square root of the ratio of the elastic stiffness to mass) yielding a better spectral overlap between the Al modes and the acoustic modes of GaN. This results in an increased Kapitza conductance; but unlike in previous studies...
using simpler Lennard-Jones systems the effect is less pronounced. This is most likely due to the disparity in phonon density of states in the metal relative to the semiconductor.

Fig. 3 also indicates that the (110)M/{0001}AB interface has a significantly higher Kapitza conductance than the (111)M/{0001}AB and the (100)M/{0001}AB interfaces. This gives some implication of effects of crystal structures. Because atomically the (110) metal surface is “more mixed” than the (111) and (100) metal surfaces, the result here supports previous findings that the atomically diffused interfaces cause mixed phonon properties (modes) of the two layers, resulting in some average phonon dispersion that promotes thermal transport.

B. Effects of metal stiffness

Elastic constants, which measure bond stiffness, are also characteristic properties of materials. To explore effects of metal bond stiffness on Kapitza conductance, simulations are performed with the SW potential where the parameter $e_{MM}$ is scaled with different scaling factor $e_{MM}/e_{AlAl}$ between 0.8 and 1.6 (this range of parameter has reached the limit of our model as extending the parameter beyond this range leads to unstable interfaces during MD simulations) and all the other parameters are taken as the basis Al–Ga–N parameters. The calculated Kapitza conductance values are shown as a function of the scaling factor in Fig. 5(a) for the three interfaces (dash lines and unfilled symbols). Note that the scaling factor is related to elastic constants of the metal. To more clearly explore the effects of elastic constants, the same Kapitza conductance data is re-plotted as a function of bulk modulus of the metal in Fig. 5(b) (again using the dash lines and unfilled symbols).

Fig. 5 indicates that for the SW potential, the Kapitza conductance increases as the metal bond stiffness decreases. In particular, the Kapitza conductance at the lowest simulated stiffness scaling factor of 0.6 is more than double of the Kapitza conductance at the highest stiffness scaling factor of 1.60 for the (111)M/{0001}AB and (100)M/{0001}AB interfaces. Note that the scaling of the parameter $e$ results in the scaling of both bond energy (corresponds to cohesive energy) and bond stiffness. This, however, is consistent with practical materials. For instance, experimental bulk modulus of metals is found to be proportional to the cohesive energy density $E_c/\Omega$, where $E_c$ is cohesive energy (per atom), and $\Omega$ is atomic volume. On the other hand, the ESW potential gives dramatically different elastic constants but the same cohesive energy for Al as compared with the SW potential.

To unravel the key material properties that determine the Kapitza conductance, the simulations are performed with the ESW potential where the scaling factor $f_{EAM}$ changes from 0.8 to 1.6 (this range is also the limit we can reach for stable interfaces). While this allows the cohesive energy of the metal to change roughly in the same range as the SW potential, the metal elastic constants of the ESW potential are dramatically different from those of the SW potential (e.g., $C_{11}$ predicted by the ESW potential falls in the 0.568–1.136 eV Å$^{-3}$ range as opposed to the 1.374–3.664 eV Å$^{-3}$ range by the SW potential). The results using the ESW potential are shown in Fig. 5 using the solid lines and filled symbols. It can be seen from Fig. 5(a) that except for the (111)M/{0001}AB interface, the ESW potential predicts a decreasing trend of the Kapitza conductance with a decreasing scaling factor for the (110)M/{0001}AB interface.
and {100}M/{0001}AB interfaces, which is, apparently, an opposite trend with respect to cohesive energy for the SW potential. Since SW and ESW potentials predict exactly the same cohesive energy for the metal, the observed opposite effects cannot be attributed to the change of the cohesive energy. On the other hand, if the data are plotted against the bulk modulus \( B \) of the metal as shown in Fig. 5(b), then the results obtained from both the SW and ESW potentials can be consolidated approximately as instances of a trend to peak at some optimal bulk modulus and then decay fairly linearly with increasing \( B \). For instance, the Kapitza conductance for the {110}M/{0001}AB and {100}M/{0001}AB interfaces initially increases as the bulk modulus \( B \) is increased from 0.25 eV Å\(^{-3}\), reaches a maximum at \( B \) between 0.75 and 1.00 eV Å\(^{-3}\), and then monotonically decreases when \( B \) is further increased. The {111}M/{0001}AB interface also appears to have this behavior except that the Kapitza conductance has not reached the maximum at the lowest bulk modulus explored.

To further understand this phenomenon, we once again calculate the phonon density of states of the SW metals of various stiffnesses with GULP and compare the results with those of GaN in Fig. 6. It can be seen that by decreasing the metal stiffness, the vibrational spectra is shifted towards lower frequencies, yielding a better spectral overlap between the metal modes and the acoustic modes of GaN. This leads to an increased Kapitza conductance. Further decreasing the metal stiffness will eventually cause the maximum frequency to go to zero and therefore misalign the vibrational states which again leads to a reduced Kapitza conductance. So despite the complexity of the present systems, they display the same effects again leads to a reduced Kapitza conductance. Therefore, the Kapitza conductance for the {110}M/{0001}AB and {100}M/{0001}AB interfaces remains sharp during the simulations. Contrarily, the {111}M/{0001}AB interface becomes relatively diffused. The atomic details of this diffused interface statistically vary among simulations. This accounts for the scatter data seen in Fig. 7.

C. Effects of metal–semiconductor cross stiffness

The cross bond stiffness between metal and semiconductor is also a characteristic property of the interface between materials. Under the scaling relation between elastic constants and cohesive energy, large cross bond stiffness should give large bond strength, and therefore a large heat of mixing between metal and semiconductor. To explore effects of metal–semiconductor cross bond stiffness on Kapitza conductance, simulations are first performed using the SW potential where parameters \( E_{\text{AM}} \) and \( E_{\text{BM}} \) are scaled with different scaling factor \( \epsilon_{\text{AM}}/\epsilon_{\text{AlAI}} = \epsilon_{\text{BM}}/\epsilon_{\text{GaAI}} \) between 0.2 and 1.6 and all other parameters are kept at their basis Al–Ga–N values. The calculated Kapitza conductance values are shown as a function of the scaling factor in Fig. 8 for the three interfaces (dash lines and unfilled symbols). Fig. 8 reveals an interesting phenomenon that when the cross bond stiffness is very low, almost the same Kapitza conductance is obtained for all the three interfaces. The Kapitza conductance then linearly increases as the cross bond stiffness is increased. When the cross bond stiffness is further increased, the Kapitza conductance for the {110}M/{0001}AB and {100}M/{0001}AB interfaces reaches a maximum and then slightly decreases before it becomes saturated. Over this saturated regime, the relatively “rough” {110}M/{0001}AB exhibits a higher conductance than the less “rough” {100}M/{0001}AB interface as discussed above. However, the Kapitza conductance of the {111}M/{0001}AB interface continuously increases as cross bond stiffness continuously increases. As a result, the Kapitza conductance of the {111}M/{0001}AB interface exceeds the Kapitza conductance of both the {110}M/{0001}AB and {100}M/{0001}AB interfaces at the very high cross bond stiffness where the scaling factor is above 1.2. These results represents a more expansive study of this sensitivity than that found in ref. 28 and have remarkable similarity to the results from one dimensional chains in ref. 50.

To gain a more complete picture of material property effects, similar simulations are also performed with the ESW potential. Here the cross interaction effects are explored by simultaneously scaling parameters \( E_{\text{AM}} \) and \( E_{\text{BM}} \) using the scaling factors \( E_{\text{AM}}/E_{\text{AlAI}} = E_{\text{BM}}/E_{\text{GaAI}} \) between 0.3 and 0.5 (again the limit we can reach for stable interfaces) with the other parameters at their basis Al–Ga–N values. This corresponds to the same stiffness range for the cross interactions used for the SW potential because the basis SW and ESW potentials give the

![Fig. 6](https://example.com/fig6.png)  
**Fig. 6** Phonon density of states for the metal M with various stiffnesses, and GaN. Here the parameter \( \epsilon_{\text{AM}}/\epsilon_{\text{AlAI}} = 0.6, 0.8, 1.0, 1.2, 1.4, 1.6 \) corresponds to Al bulk modulus \( B_{\text{AI}} = 0.92, 1.22, 1.53, 1.84, 2.14, 2.45 \) Å eV\(^{-3}\).
same bond energies and the second derivatives of the bond energies for the cross interactions. Results of the Kapitza conductance calculated from the ESW potential are included in Fig. 8 using filled symbols. The ESW data again shows that the Kapitza conductance increases with the interfacial stiffness. In either the ESW or SW cases, it is likely that increasing the interfacial stiffness yields a greater probability of anharmonic phonon–phonon interactions, thereby offering another channel for which thermal energy can transmit across the interface.28

IV. Conclusions

Large scale molecular dynamics simulations are used to determine the functional dependence of thermal boundary conductance on material properties including masses, stiffness of individual materials, stiffness of interfaces, and crystallographic orientations. Results obtained from different generic interatomic potentials are consolidated, and the following conclusions are obtained:

(a) Thermal boundary conductance is dictated by material stiffness rather than bonding strength as Kapitza conductance calculated from different interatomic potentials indicate a consistent behavior as a function of bulk modulus but not as a function of bonding strength.

(b) When the properties of the semiconductor compound are given, thermal boundary conductance reaches a maximum at an optimum metal atomic mass and an optimum metal stiffness.

(c) Thermal boundary conductance increases with interfacial stiffness.

(d) Thermal boundary conductance is sensitive to crystal orientation. The {110}M/{0001}AB interface usually has a higher conductance compared with the {111}M/{0001}AB and {100}M/{0001}AB interfaces. However, the {111}M/{0001}AB interface can have abnormally high conductance when the metal stiffness is extremely low or the interfacial stiffness is extremely high.

(e) Thermal boundary conductance increases with the overlap of the vibrational spectra between the Al modes and the acoustic modes of GaN. This fundamental mechanism can explain all the material property effects on Kapitza conductance discovered in our simulations.

Appendix A: potential parameters

The $\xi_{\text{NAI}}$, $\sigma_{\text{NAI}}$, $\lambda_{\text{NAI}}$, and $\alpha_{\text{NAI}}$ parameters of the Al–Ga–N SW potential have been modified from the previous work14 to better fit the experimental cohesive energy of the wurtzite AlN. Details of the potential parameterization have been described previously14 and hence will not be repeated. For convenience, a complete list of the Al–Ga–N SW potential parameters are shown in Table 2.
ESW values are shown for the AlN wurtzite which is not stable functional theory is 1.37 eV Å fictitious phase. The bulk modulus of dc N calculated from density potential are fitted to give the same equilibrium bond lengths, the same as given by Table 2, and a Morse potential for the N–N interactions. These Morse parameters are listed in Table 3.

The properties of different Al, Ga, N, GaN, AlN, and GaAl phases, as predicted by the Al–Ga–N SW and ESW potentials, are shown in Tables 4–6 for cohesive energies, lattice constants, and elastic constants respectively, along with the available experimental data.

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### Notes and references
