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Inelastic phonon transport across atomically sharp metal/semiconductor interfaces

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Understanding thermal transport across metal/semiconductor interfaces is crucial for heat dissipation of electronics. The dominant heat carriers in non-metals, phonons, transport elastically across most interfaces, except for a few extreme cases where the two materials that formed the interface are highly dissimilar with a large difference in Debye temperature. In this work we show that even for two materials with similar Debye temperatures (Al/Si, Al/GaN), a substantial portion of phonons will transport inelastically across their interfaces at high temperatures, significantly enhancing interface thermal conductance. Moreover, we find that interface roughness strongly affects phonon transport process. For atomically sharp interfaces, phonons are allowed to transport inelastically and interface thermal conductance linearly increases at high temperatures. With increasing interface roughness, inelastic phonon transport rapidly diminishes. Our results provide new insights on phonon transport across interfaces and open up opportunities to engineering interface thermal conductance specifically for materials of relevance to microelectronics.

In modern electronics, thermal resistance of interfaces (reciprocal of thermal conductance) is the main limiting factor for heat dissipation, especially for power electronics and high energy density applications¹⁻⁵. The scattering of heat carriers, predominately phonons, leads to interface thermal resistance. Over the past several decades, there were extensive studies on thermal conductance across metal/semiconductor interfaces, both experimentally and theoretically⁶⁻¹². However, the rare agreement between experiments and calculations as well as the scattered experimental results even for the same interface warrant study for much better understanding of thermal transport across interfaces^{6,13-15}.

Theories have been developed to explain interface thermal conductance since 1950s, such as the widely used acoustic mismatch model (AMM) and diffuse mismatch model (DMM)^{16,17}. AMM is based on the assumption that phonon is reflected or transmitted specularly, while DMM assumes that phonon scattering is completely diffusive at the interface. Both DMM and AMM assume phonon transport across interface is elastic, which means the transmitted/reflected phonon has the same frequency as the incident phonon. The elastic transport assumption predicts that interface thermal conductance will reach a plateau at temperatures higher than the lower Debye temperature (TD) of the two materials that formed the interface, when all phonons in this side have been excited. In recent years, advanced calculation methods such as molecular dynamics (MD) and atomistic Green function (AGF) have been used to study phonon transport process across interfaces^{6-8,10,11,18}. Inelastic phonon transport process has been predicted to exist in interfaces between very dissimilar materials, where the transmitted phonons do not have the same frequency as the incident phonons^{7,11,19}, and anharmonicity was found to be of fundamental importance

for the inelastic phonon transport across interfaces. Despite these advances, there are still controversies related to under what conditions inelastic process will occur in the first place. For example, Landry and McGaughey predicted that inelastic phonon transport will become dominant when temperature is higher than ~ 500 K, whereas Feng and Ruan computed that inelastic phonon transport contributes more than 50% to the total thermal conductance for Si/Ge interfaces even at room temperature^{7,11}.

Most experimental results show that phonon transport across metal/semiconductor interface is an elastic process, as interface thermal conductance saturates at high temperatures for most interfaces under study^{6,15,20}. There are only a few exceptions to this trend^{12,13,21}, and all of these suggest a large Debye temperature difference across interface leads to the observation of non-saturated thermal conductance at high temperatures. The most notable one is highly dissimilar Bi/diamond interface (the TD ratio of diamond and Bi is ~ 19), where Lyeo and Cahill observed a linear increase of thermal conductance with temperature even at high temperatures¹². This is attributed to the temperature-dependent inelastic phonon transport, which add an additional thermal transport channel across interfaces^{12,16}. However, it is still an open question how large a Debye temperature difference is required for inelastic phonon transport to occur, which needs to be examined in details.

The lack of high-quality interface limited experimental study of phonon transport across interfaces. Extrinsic phonon scattering centers, such as atomic intermixing, interface roughness and contamination, would easily scatter phonons and bury the phonon's intrinsic elastic and inelastic transport processes across interfaces²². Epitaxial metal/semiconductor interfaces are usually used to study the intrinsic interface phonon transport due to their importance and high quality^{6,22-24}. However, previous studies on epitaxial interfaces used typically lacked atomic-level structural details^{12,21} with interface roughness and atomic intermixing often ignored, thus only lead to qualitative analysis and limit our understanding of intrinsic phonon transport across interfaces.

Here, we report the observation of inelastic phonon transport across metal/semiconductor interfaces, with a clear crossover from elastic-dominated to inelastic-dominated phonon transport following the rise of temperature and reduction of interface roughness. Our results show that, even in an interface formed with highly similar materials with Debye temperature ratio less than 1.5, inelastic phonon transport still exists and significantly enhances thermal conductance at high temperatures, suggesting that inelastic phonon transport is universal across interfaces even in acoustically similar materials. We also observed that inelastic phonon transport could only dominate the process when the interface is atomically sharp. Our MD simulations also confirmed that the interface roughness is crucial for inelastic phonon transport.

We build high-quality metal/semiconductor interfaces by epitaxial growth of Al(111) on Si(111) and GaN (0001) using molecular beam epitaxy (MBE) (See Methods and Supplementary Information Note

I). Before the growth of Al/Si interface, a Si wafer was cleaned by hydrofluoric acid to remove native oxide and then was heated in vacuum at 300 °C for 30 minutes to degas. The growth was in ultrahigh vacuum to ensure that the interface is free of oxide layer and adsorbates. To fabricate an Al/Si interface with controlled interface quality, Al growth was proceeded at different temperatures, 100 °C and 300 °C (denoted as Sample 1 and Sample 2, respectively), knowing that 100 °C is the optimum temperature for Al deposition in our prior work²⁵ (see Supplementary Information Note I). For Al/GaN interface, GaN thin film was grown on sapphire substrate in MBE chamber first, and then the temperature was ramped down to 150 °C to grow Al layer. For comparison, an Al/SiO₂/Si sample was also prepared by e-beam evaporation of Al on Si substrate in presence of native oxide.

We measure the thermal conductance of Al/Si and Al/GaN interfaces over a wide range of temperatures (80-700 K) by time-domain thermoreflectance (TDTR)^{26,27}. The details of our TDTR setup, data analysis and uncertainty estimation can be found in Methods and Supplementary Information Note IV. Note that only the Al/Si Sample 1 can reach 700 K for TDTR measurement. For Al/Si Sample 2 and Al/GaN, the Al films start to melt and large pores form when temperature is higher than 600 K, making TDTR measurement impossible.

The measured thermal conductance, G , of Al/Si and Al/GaN interfaces as a function of temperature is plotted in Fig. 1a and Fig. 1b. At room temperature, Al/Si Sample 1 and Al/GaN show a record high thermal conductance of 370 MW m⁻² K⁻¹ and 410 MW m⁻² K⁻¹, respectively. For both Al/Si Sample 1 and Al/GaN, our results can be clearly divided into two regimes. At temperatures lower than the Debye temperature of Al ($T_D = 428$ K), the thermal conductance of Al/Si Sample 1 and Al/GaN gradually saturate with the increase of temperature, which has the same trend as previous measured thermal conductance of Al/Si and Al/GaN interfaces. However, when temperature approaches 400 K (close to the Debye temperature of Al) and beyond, both Al/Si Sample 1 and Al/GaN show linear increase in thermal conductance with temperature instead of reaching a plateau. For Al/Si Sample 2, it has a thermal conductance of 330 MW m⁻² K⁻¹ at room temperature. Throughout the low temperatures ($T < T_D$), the thermal conductance of Al/Si Sample 2 is ~10% lower than that of Al/Si Sample 1. However, unlike Al/Si Sample 1 and Al/GaN, Al/Si Sample 2 shows a saturated thermal conductance when $T > T_D$.

For metal/semiconductor interface, there are four thermal transport pathways, which are the phonon-phonon transport across interface including both elastic and inelastic phonon transport, as well as the electron-phonon coupling in the metal and across interfaces. The effect of electron-phonon coupling across interfaces is negligible¹³. The electron-phonon coupling in Al adds an additional thermal resistance in series with the phonon-phonon interactions²⁸, which is driven by the thermal non-equilibrium between electrons and phonons near the interface. To calculate the phonon-phonon transport

induced interface thermal conductance alone, we follow Majumdar and Reddy's treatment of electron-phonon coupling to use $G = \frac{G_{ep}G_{pp}}{G_{ep}+G_{pp}}$, where G is the total thermal conductance, G_{ep} and G_{pp} are electron-phonon coupling and phonon-phonon transport induced thermal conductance, respectively²⁸. The electron-phonon coupling induces the conductance $G_{ep} = \sqrt{g\Lambda_p}$, where g is the electron cooling rate and Λ_p is the lattice thermal conductivity of Al. We use experimental determined g ²⁹ and first-principles calculated Λ_p ³⁰, which have been reported previously and are well accepted, to determine G_{ep} . The calculated G_{pp} for Al/Si Sample 1, Sample 2 and Al/GaN are shown in Fig. S3. It shows that G_{pp} of Al/Si Sample 1 and Al/GaN interfaces has a stronger temperature dependence than G , while G_{pp} of Al/Si Sample 2 changes slightly with temperature at high temperatures.

To study the relationship between interface quality and thermal conductance and understand the difference of the two Al/Si interfaces, high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) was used to study the cross-sectional interface structure, as shown in Fig. 2. The interface structure of Al/Si Sample 1 is shown in Fig. 2a, showing crystalline Al film epitaxially grown on Si substrate. The interplanar spacing between Al is 2.3 Å, in agreement with the lattice constant of Al(111)²⁵. The enlarged area in yellow box demonstrates that the interface between Al and Si is atomically sharp, with only 1 - 2 distorted layers of interface atoms observed. The cross-sectional HAADF-STEM image of Al/Si Sample 2 is shown in Fig. 2b and Fig. S2a. Unlike the Al/Si Sample 1, it shows a higher interface roughness of around 1 - 2 nm, which roughly equals the thickness of 3-6 atomic layers of Si. Further structure analysis in Supplementary Fig. S2 shows that the rough interface of Sample 2 results from the intermixing of Al and Si atoms, which is due to the high growth temperature that facilitates atomic diffusion across interfaces.

Fig. 1 | Thermal conductance of Al/Si and Al/GaN interfaces. (a) Thermal conductance of Al/Si Sample 1 (red spheres) and Sample 2 (blue spheres). Black and green dashed lines are interface thermal conductance calculated by phonon radiation limit and DMM, respectively. For comparison, we show previously measured Al/Si thermal conductance in open squares, triangles, and diamond by Minnich³¹, Wilson³², and Jiang³³, respectively. Yellow solid triangles are measured thermal conductance of Al/Si with a native oxide layer, comparing with the results by Hopkins, shown in open circles¹⁴. (b) Thermal conductance of Al/GaN interface (red spheres). For comparison, calculated thermal conductance of phonon radiation limit (black dashed line) and DMM (green dashed line) is plotted. Previous measurement results by Donovan³⁴ are shown in open circles, the Al film of which was deposited by e-beam evaporation. Phonon dispersion relations of Al/Si (c) and Al/GaN (d) are calculated from first-principles.

Fig. 2 | Interface structure of Al/Si Sample 1 and Sample 2. Cross-sectional TEM image of Al(111)/Si(111) Sample 1 (a) and Sample 2 (b). Scale bars are 2.5 nm.

To understand the phonon transport mechanism, we use non-equilibrium molecular dynamics (NEMD) to compute interfacial thermal conductance of Al/Si interfaces with different roughness (see Methods and Supplementary Information Note VI). The Al/Si structure along the (111) orientation is established with sharp and rough interface, as shown in Fig. S5a and Fig. S5b. The sharp interface is composed of Si(111) 3×3 unit cell and Al(111) 4×4 unit cell. Before calculating the thermal interface conductance, the structure was fully relaxed to reduce the stress at the interface. A rough interface was attained by locally melting the interface at 3000 K and quenched at 300 K in the MD simulation, and the thickness of the rough interface is about 1.1 nm. The simulation results with a heat bath temperature difference of 60 K at the sharp interface of 500 K is shown in Supplementary Information Note VI, and the thermal conductance was calculated as $738 \text{ MW m}^{-2} \text{ K}^{-1}$. The thermal conductance predicted by MD at high temperature is shown in Fig. 3a. The interface thermal conductance at the rough interface is lower than the value of sharp interface, which is consistent with the experiment results. As temperature rises, the increasing slope of thermal conductance as a function of temperature at the sharp interface is much higher than that of rough interface, demonstrating that the surface roughness is crucial for the observation of inelastic phonon scattering. Considering that MD simulation does not make assumptions on the

phonon scattering mechanisms, the temperature dependence of thermal conductance at high temperatures is possibly universal, indicating that inelastic phonon scattering is always expected at high-quality interfaces.

Fig. 3 | Phonon transport behavior across Al/Si interface computed by molecular dynamics. (a) Calculated thermal conductance of sharp (red dashed line) and rough (violet dashed line) Al/Si interfaces. (b) Phonon transmission coefficient for sharp (red) and rough (violet) interfaces (c) Schematic of temperature distributions near the sharp and rough interfaces. Here $T_{p,h}$ and $T_{p,l}$ represent temperatures of high- and low-energy phonons. ΔT_s and ΔT_r are temperatures drop across sharp and rough interfaces, respectively.

To further explain the observed distinct temperature dependence of the thermal conductance, we calculate the spectral phonon transmissivity using atomistic Green's function, as shown in Fig. 3b. For the sharp interface, the transmissivity of low-energy phonons (here we define phonons within the energy overlap between Al and Si, *i.e.* lower than 10 THz, as low-energy phonons) is higher than the high-energy phonons. This will result in a relatively large temperature difference between these phonons, as sketched in Fig. 3c. Such a thermal non-equilibrium between high-energy phonons and low-energy phonons leads to mode conversion and energy communication between them through phonon scatterings. Thus, the high-energy phonons with low transmission probability will convert to high transmissivity low-energy phonons before they undergo the transport process across interface, leading to inelastic phonon transport.

For rough interfaces, the difference of transmissivity between the low- and high-energy phonons becomes smaller comparing with that of sharp interfaces, as the transmissivity for all phonons reduces. As a result, the temperature difference of different phonons is expected to be smaller, thus the phonon non-equilibrium is smaller for rough interfaces. The reduced phonon non-equilibrium leads to less energy communication between high and low energy phonons, and the inelastic phonon transport will diminish.

Our results point out that large Debye temperature difference is not required for inelastic phonon

transport process to occur, as the Debye temperature ratio of both Al/Si and Al/GaN is less than 1.5. This finding is in stark contrast to what previous experiments suggested, where inelastic transport can be only observed across interfaces formed with large Debye temperature difference^{12,13,21}. This is also the first direct observation of the crossover from elastic-dominated to inelastic-dominated interface phonon transport processes over the wide temperature range far below to far above T_D . Our work also suggest that optical phonons contribute significantly in the inelastic transport. As shown in Fig. 1d, the high-energy phonons are predominately optical phonons in GaN. For Al/Si, the thermal conductance does not saturate at 700 K, which is higher than the Debye temperature of Si, suggesting that high frequency optical phonons excited at temperatures higher than 700 K contribute to the thermal conductance.

A recent work by Cheng *et al.* claimed that no inelastic phonon transport is observed in atomically sharp Al/Al₂O₃ interface grown by MBE⁶. We would like to point out that at least 5 monolayers of Al₂O₃ near the interface are distorted, as shown in their TEM image, which is similar to our Al/Si Sample 2 and not as sharp as our Al/Si Sample 1. Their work actually echoes with our finding, that significant inelastic thermal transport can only be observed at atomically sharp interfaces.

In summary, we report the observation of inelastic phonon transport across high-quality Al/Si and Al/GaN interfaces grown by MBE. We observed a continuously increasing thermal conductance at high temperatures, which is attributed to inelastic phonon transport process across the interface. The inelastic phonon transport is expected to occur at atomically sharp interfaces where the strong phonon non-equilibrium exists, in contrast to rough interfaces. This work sheds light on increasing thermal conductance across interface at high temperatures and improving heat dissipation of electronic devices.

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Methods

Samples. The epitaxial growth of Al was carried out in a molecular beam epitaxy (MBE) system with a base pressure of 2.7×10^{-7} Torr. A Si wafer was cleaned by hydrofluoric acid to remove the native oxide layer before loading into MBE chamber. The Si wafer was degassed at 900 °C for 90 minutes, and then cooled down for aluminum growth. To form an Al/Si interface with controlled quality, Al growth was proceeded at different temperature: 100 °C for Sample 1 and 300 °C for Sample 2. The deposition rate of Al is 15 nm/min. On GaN substrate, Al is grown at 150 °C.

TDTR measurement. We measure interface thermal conductance of Al/Si at 80-700 K by time-domain thermoreflectance (TDTR) in a Janis VPF-800 cryostat²⁶. The measurement was performed with 5x objective lens with $1/e^2$ radius of 10.6 μm and a modulation frequency of 10.1 MHz for pump beam. The laser power is in the range of 100-200 mW for low temperature measurements, with steady-state temperature rise less than 2 K.

DMM and radiation limit calculation. Assuming a Debye approximation of phonon dispersion relation, the phonon transmission probability $\alpha_A(\omega)$ can be written as¹⁶,

$$\alpha_A(\omega) = \frac{\sum_j v_B^{-2}}{\sum_j v_A^{-2} + \sum_j v_B^{-2}} \quad (1.)$$

where ω is phonon frequency, v is phonon group velocity, superscript A and B refers to materials at one and the other side of interface, respectively, j is the branch of phonons. In this way, interface thermal conductance G is given by¹⁶,

$$G = \frac{1}{4} \sum_j \int_0^{\omega_{A,j}^{\text{Debye}}} D_{A,j}(\omega) \cdot \frac{\partial}{\partial T} f(\omega, T) \cdot \hbar \omega \cdot v_{A,j} \cdot \alpha_A(\omega) d\omega \quad (2.)$$

where D is the phonon density of states, f is Bose-Einstein distribution, v is phonon group velocity, $\omega_{A,j}^{\text{Debye}}$ is the Debye frequency of phonon mode j in material A.

The maximum interface thermal conductance based on elastic phonon scattering is considered as phonon radiation limit (G_{RL}). In this case, all the phonons in material B below the Debye frequency of material A are assumed to transmit across the interface with unit transmission probability ($\alpha_B = 1$). With this assumption, the thermal conductance can be written as,

$$G_{\text{RL}} = \frac{1}{4} \sum_j \int_0^{\omega_{A,j}^{\text{Debye}}} D_{B,j}(\omega) \cdot \frac{\partial}{\partial T} f(\omega, T) \cdot \hbar \omega \cdot v_{B,j} d\omega \quad (3.)$$

MD simulation. The cross-section area is $19.89 \times 22.97 \times 10^{-20} \text{ m}^2$. The simulation is based on non-equilibrium molecular dynamics (NEMD) method, which applies heat bath at both ends of the structure and calculates the interface thermal conductance using Fourier's Law:

$$G = \frac{J}{A\Delta T}$$

where A is heat flux across interface at unit time (W), J is cross-section area (m^2), and ΔT is temperature drop across interface (K). The simulation is carried using LAMMPS by MEAN potential function with time step of 1fs. The system is under canonical ensemble (NVT) with a relaxation time of 500 ps. The temperature and heat flux are recorded under micro-canonical ensemble (NVE) by applying heat bath using Langevin method. The structure length L is set as 44 nm and 49 nm for comparison, and the structure along the other two dimensions are set as infinite.

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Data availability

Source data are available for this paper. All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

Code availability

The analysis codes that support the findings of the study are available from the corresponding authors upon reasonable request.

Author contributions

B.S. and X.W. conceived the project and supervised the research with J.W. and F.K. H.S., Q.L. and L.Z. carried out the TDTR measurement and the DMM calculations. S.Y., F.L., and X.W. grew and characterized the samples. T.W., H.J. and J.W. performed the TEM analysis. S.H. and X.G. conducted the MD simulations. B.S. and X.G. figured out the heat transport mechanism with help from Y.K.K. All authors contributed to the discussions and manuscript preparation.

Competing interests

The authors declare no competing interests.

Figures

Figure 1

Thermal conductance of Al/Si and Al/GaN interfaces. (a) Thermal conductance of Al/Si Sample 1 (red spheres) and Sample 2 (blue spheres). Black and green dashed lines are interface thermal conductance calculated by phonon radiation limit and DMM, respectively. For comparison, we show previously measured Al/Si thermal conductance in open squares, triangles, and diamond by Minnich³¹, Wilson³²,

and Jiang³³, respectively. Yellow solid triangles are measured thermal conductance of Al/Si with a native oxide layer, comparing with the results by Hopkins, shown in open circles¹⁴. (b) Thermal conductance of Al/GaN interface (red spheres). For comparison, calculated thermal conductance of phonon radiation limit (black dashed line) and DMM (green dashed line) is plotted. Previous measurement results by Donovan³⁴ are shown in open circles, the Al film of which was deposited by e-beam evaporation. Phonon dispersion relations of Al/ Si (c) and Al/GaN (d) are calculated from first-principles.

Figure 2

Interface structure of Al/Si Sample 1 and Sample 2. Cross-sectional TEM image of Al(111)/Si(111) Sample 1 (a) and Sample 2 (b). Scale bars are 2.5 nm.

Figure 3

Phonon transport behavior across Al/Si interface computed by molecular dynamics. (a) Calculated thermal conductance of sharp (red dashed line) and rough (violet dashed line) Al/Si interfaces. (b) Phonon transmission coefficient for sharp (red) and rough (violet) interfaces (c) Schematic of temperature distributions near the sharp and rough interfaces. Here $T_{p,h}$ and $T_{p,l}$ represent temperatures of high- and low-energy phonons. ΔT_s and ΔT_r are temperatures drop across sharp and rough interfaces, respectively.

Supplementary Files

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