



Interfacial phonon transport with frequency-dependent transmissivity by Monte Carlo simulation

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ABSTRACT

Interfacial phonon transport widely exists in nanosystems, yet the physical mechanism has never been well understood. In this work, a numerical framework is developed for interfacial phonon transport between dissimilar materials with a frequency-dependent transmissivity by introducing the spectral diffuse mismatch model into an efficient kinetic-type Monte Carlo scheme. The numerical method is validated by modeling cross-plane phonon transport through several single-layer and bi-layer thin films, which shows good agreement with the discrete-ordinates solutions. Through mesoscopic modeling, the size effect of Kapitza conductance is found to be weak or vanishing when the equivalent equilibrium temperature or emitted phonon temperature is adopted for defining the interfacial temperature difference respectively. Furthermore, the effective Kapitza conductance decreases when interfacial roughness is introduced, which can be mainly ascribed to the increased interfacial area ratio by roughness. For engineering application, an empirical power law is proposed for the dependence of effective Kapitza conductance on interfacial area ratio. The present work will promote fundamental understanding and modeling capability of interfacial heat transport, as well as engineering design and optimization of interface in nano devices.

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1. Introduction

In the past several decades, with the rapid developments of micro- and nano- manufacturing and nano-technology, there are increasing interests in nanoscale heat transport [1,2]. The thermal management of micro- and nano-electronics is pursuing thermal interfacial materials with high thermal conductivity [3]. The introduction of complex interfaces including dislocations, grain boundaries etc., is one of the main methods to improve the figure of merit of nanostructured thermoelectric material [4–6]. Both of the above two issues require a profound understanding of the physical mechanism in interfacial phonon transport. The classical Fourier's law, which is valid for heat transport in bulk material, becomes no longer available for this situation [1,7], where phonon-interface scattering is dominant over the intrinsic scattering. Therefore, it is essential to develop effective theories and methods for modeling interfacial phonon transport.

The interface between two dissimilar materials will cause a cross-plane thermal transport resistance comparable to the intrinsic thermal resistance of each material layer. There will be a tem-

perature jump across the interface and the ratio of temperature jump to the heat flux across the interface is defined as Kapitza resistance or thermal boundary resistance. The Kapitza conductance or thermal boundary conductance is also often used as the inverse of the Kapitza resistance. There have been mainly two categories of approaches to predicting the thermal boundary conductance: (i) microscopic methods including molecular dynamics simulation [8–10], atomistic Green's function method [11–13], etc.; (ii) mesoscopic modeling based on the phonon Boltzmann equation [14]. The microscopic methods are usually situated for small nanostructures and simple interfaces due to an intensive consumption of computational time and resources. In contrast, the phonon Boltzmann modeling represents a feasible approach for much larger structures and more complex interfaces. The mesoscopic modeling requires an important physical parameter in determining the Kapitza conductance between a material pair: phonon transmission coefficient across the interface.

The classical models for phonon transmission coefficient include the acoustic mismatch model (AMM) [15] and the diffuse mismatch model (DMM) [16]. The AMM treats phonons as a kind of acoustic waves transmitting through the interface. The transmission coefficient is calculated from the acoustic impedance of materials that form the interface [15]. It provides an appreciably good prediction of Kapitza conductance at very low temperature

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where phonons are mainly populated at low frequency with wavelengths much larger than the size of interfacial asperity. In elevated temperature scope, the AMM becomes often overestimating the Kapitza resistance because of the stronger Rayleigh scattering of the phonon population when shifting to higher frequency scope with wavelengths comparable to or even smaller than the size of interface asperity [16,17]. The DMM was thus proposed, treating phonons as a kind of particles transmitting diffusely through the interface without keeping the information of the side they come from. The transmission coefficient is determined by the phonon dispersion relations of both materials. The DMM often works well at higher temperature such as around the room temperature [16]. Some mixed models have also been developed which consider partially specular and partially diffuse transmission through the interface, yet based on simplified gray Debye's approximation [14]. On the other hand, the portion of specular scattering at the interface, known as the interfacial specularity parameter, is difficult to specify in prior and has to be extracted from fitting the experimental results. Therefore, the DMM is currently the most popular one for describing interfacial phonon transport in realistic applications around room temperature inferred from some good agreements between Kapitza conductances measured by experiments and predicted by simulations for different material pairs [18–20].

Taking into account the aforementioned models for phonon transmission coefficient, people have paid much effort to study interfacial phonon transport by solving the phonon Boltzmann equation [14,21–25]. Two categories of numerical schemes are currently available for the solution of Boltzmann equation, including the deterministic method (discrete-ordinates method [14,25], finite volume method [21], lattice Boltzmann method [26] and so on) and the stochastic method (Monte Carlo method [22–24]). Monte Carlo method avoids directly solving the high-dimensional Boltzmann equation by tracking the phonon dynamics through the pseudo-particles. Therefore, the interface boundary treatment in Monte Carlo method is simpler with a clearer physical picture via mimicking the realistic phonon-interface scattering. As a result, the Monte Carlo method is a better choice compared with the deterministic methods for studying interfacial phonon transport with complex geometries. Jeng et al. [22] first used Monte Carlo method to model the thermal conductivity of nanoparticle composites based on the DMM under gray Debye's approximation. Huang et al. [23] presented an improved Monte Carlo scheme to simulate interfacial phonon transport based on the gray mixed interface model proposed in Ref. [14]. Recently, Péraud and Hadji-constantinou [24] modeled the Al/Si interfacial heat transport in transient thermo-reflectance experiments using energy-based variance-reduced Monte Carlo formulations based on a semi-spectral interface model considering gray transmission coefficient for each individual frequency at mono-direction [27]. Besides, there are several Monte Carlo simulations of phonon transport through grain boundaries in polycrystalline nanostructures with empirical expressions for spectral transmission coefficient [28–30]. To sum up, the previous Monte Carlo simulations of interfacial phonon transport merely considered a constant gray transmissivity at one direction or at both directions between dissimilar materials, in spite of an empirical treatment of frequency-dependent transmissivity through grain boundary within a single material. It remains to carefully consider the strongly frequency-dependent interfacial phonon transmissivity between dissimilar materials, which has been demonstrated significant in both microscopic computation [13] and recent experimental measurement [31].

The aim of the present work is to develop a numerical framework for interfacial phonon transport between two material pairs by introducing the spectral diffuse mismatch model (SDMM) into an energy-based variance-reduced Monte Carlo scheme. Although

the SDMM is still a crude approximation to the realistic situation in interfacial phonon transport [18], it is the most appropriate theoretical model available currently. In principle, when supplied with the detailed frequency-dependent phonon transmissivity from recent first-principle calculation [32,33], the phonon Boltzmann modeling can provide a more accurate description of interfacial heat transport. Yet for the convenience of development of numerical framework, we take the classical SDMM into account as a first step. The inclusion of *ab initio* frequency-dependent transmissivity into the present Monte Carlo scheme is straightforward and will be investigated in the future work. The remaining of this article is organized as below: a brief fundamental knowledge of the kinetic-type Monte Carlo method and a detailed introduction of the novel interface boundary treatment, are presented in Section 2. Section 3 gives the validation of our Monte Carlo framework by modeling cross-plane phonon transport through both single-layer and bi-layer thin films. Two pertinent applications are studied in Section 4: including the size effect and roughness effect on Kapitza conductance. Concluding remarks are finally made in Section 5.

2. Numerical method

Phonon Monte Carlo scheme is a kind of pseudo-particle method to solve the phonon Boltzmann equation [34], with its earlier counterpart in rarefied gas flow the direct simulation Monte Carlo (DSMC) [35–38]. It takes statistical samples (phonon energy packets in this work) to simulate phonon dynamics, where the drift process and scattering process take place separately. The required macroscopic information (temperature, heat flux, and so on) is then extracted by averaging over these statistical samples. The kinetic Monte Carlo scheme is adopted for solution of the energy-based deviational phonon Boltzmann equation [24,34,39]:

$$\frac{\partial e^d}{\partial t} + \mathbf{v}_g \cdot \nabla e^d = - \frac{e^d - (e_{loc}^{eq} - e_{Teq}^{eq})}{\tau(\omega, \mathbf{p}, T)}, \quad (1)$$

where $e^d = \hbar\omega(f - f_{Teq}^{eq})$ is the deviational energy distribution with the reduced Planck constant $\hbar$, phonon angular frequency ω , phonon distribution f and the Bose-Einstein distribution $f_{Teq}^{eq} = [\exp(\hbar\omega/k_B T_{eq}) - 1]^{-1}$ at the referenced equilibrium temperature T_{eq} ; $\mathbf{v}_g$ is group velocity, $\tau(\omega, \mathbf{p}, T)$ being the relaxation time for phonons with frequency ω , polarization $\mathbf{p}$ at a thermodynamic temperature T ; $e_{loc}^{eq} = \hbar\omega f_{loc}^{eq}$ and $e_{Teq}^{eq} = \hbar\omega f_{Teq}^{eq}$ being the pseudo-equilibrium and equilibrium energy distributions at pseudo-equilibrium temperature T_{loc} and referenced equilibrium temperature T_{eq} respectively. The linearized version of Eq. (1) is actually solved under tiny temperature difference assumption in kinetic Monte Carlo scheme [34,39], with a more detailed introduction presented in Appendix A.

2.1. Interface treatment

The spectral diffuse mismatch model (SDMM) [40] is introduced into the kinetic Monte Carlo scheme considering only elastic phonon interfacial scattering without any polarization conversion. In other words, we do not consider the conversion of longitudinal acoustic phonon into transverse one or vice versa when the phonon scatters at the interface. The transmission coefficient is derived by applying the principle of detailed balance and the diffuse scattering assumption at interface. The phonon heat flux across the interface from side 1 to side 2 is expressed as [40]:

$$q_{1 \rightarrow 2} = 2\pi \sum_{\mathbf{p}} \int_0^\pi \int_0^{\omega_m} \cos \theta \sin \theta \hbar \omega f(\omega, T_{e1}) \alpha_{12}(\omega, \mathbf{p}) D_1(\omega, \mathbf{p}) v_{1g} d\omega d\theta, \quad (2)$$

where 1 and 2 represent the labels of two dissimilar materials, with T_{e1} the emitted phonon temperature of material 1, $\alpha_{12}(\omega, p)$ being the frequency-dependent transmission coefficient from material 1 to material 2; θ is the polar angle, D being the density of states. The definition of emitted phonon temperature will be discussed later in Section 4.1. When both sides of materials are in an equilibrium state at a temperature T , the phonon heat flux from side 1 to side 2 is equal to that from the opposite direction, i.e., $q_{1 \rightarrow 2} = q_{2 \rightarrow 1}$. Thus one obtains the following relation [40]:

$$\begin{aligned} 2\pi \sum_p \int_0^\pi \int_0^{\omega_m} \cos \theta \sin \theta \hbar \omega f^{\text{eq}}(\omega, T) \alpha_{12}(\omega, p) D_1(\omega, p) v_{1g} d\omega d\theta \\ = 2\pi \sum_p \int_0^\pi \int_0^{\omega_m} \cos \theta \sin \theta \hbar \omega f^{\text{eq}}(\omega, T) \alpha_{21}(\omega, p) D_2(\omega, p) v_{2g} d\omega d\theta. \end{aligned} \quad (3)$$

Based on the diffuse scattering assumption: $\alpha_{21}(\omega, p) = 1 - \alpha_{12}(\omega, p)$ and the principle of detailed balance for individual phonon frequency and polarization, the spectral transmission coefficient is derived as:

$$\alpha_{12}(\omega, p) = \frac{f^{\text{eq}}(\omega, T) D_2(\omega, p) v_{2g}}{f^{\text{eq}}(\omega, T) D_1(\omega, p) v_{1g} + f^{\text{eq}}(\omega, T) D_2(\omega, p) v_{2g}}. \quad (4)$$

With the aid of the expression of density of states, Eq. (4) is rewritten as [40]:

$$\alpha_{12}(\omega, p) = \frac{[k_{p,2}(\omega)]^2}{[k_{p,1}(\omega)]^2 + [k_{p,2}(\omega)]^2}, \quad (5)$$

where k is the magnitude of wave vector.

To implement the interface treatment, we firstly specify which one, among the phonon intrinsic scattering time, boundary scattering time and interface scattering time, is the shortest. The phonon intrinsic scattering time is available from the phonon relaxation time expression; the boundary or interface scattering time is deduced from the knowledge of trajectory, velocity of the present energy packet and the distance between the energy packet and boundary or interface. If the interface scattering time is the shortest, this energy packet with frequency ω and polarization p encounters with interface at first. A random number is then generated and compared to the transmission coefficient $\alpha(\omega, p)$. If the random number is smaller than the transmission coefficient, this energy packet will be diffusely transmitted into the other side; otherwise, it will be diffusely reflected back [23]. The energy packet reflected back or transmitted into the other side keeps its frequency and polarization. For the reflection case, other properties of the packet are computed based on the phonon dispersion of material at old side; whereas for the transmission case, other properties of the packet are computed based on the phonon dispersion of material at the new side. Note the present interface treatment is different from that in previous Monte Carlo simulation of interfacial phonon transport through grain boundary within a single material [28–30], where the properties of energy packet are the same for both the reflection and transmission cases. The other difference is that the SDMM is adopted here whereas an empirical expression of phonon transmission coefficient is used therein [28–30]. The change of the direction of phonon group velocity is based on the Lambert's cosine law. Firstly generate two uniform random numbers $R_1 \in [0, 1]$ and $R_2 \in [0, 1]$ associated with the polar angle θ and azimuthal angle ϕ ; then the velocity components of the energy packet are determined as below:

$$\begin{aligned} v_1 &= v_{g,j} \cos \theta = v_{g,j} R_3 \\ v_2 &= v_{g,j} \sin \theta \cos \phi = v_{g,j} \sqrt{1 - R_3^2} \cos(2\pi R_2) \\ v_3 &= v_{g,j} \sin \theta \sin \phi = v_{g,j} \sqrt{1 - R_3^2} \sin(2\pi R_2) \end{aligned} \quad (6)$$

where v_1 represents velocity component perpendicular to the interface, v_2 and v_3 the velocity component parallel to the interface; $v_{g,j}$ is the phonon group velocity of material at side j as the old side for reflection case or the new side for transmission case; R_3 is the arithmetic square root of R_1 [41]. The flow chart of the interface treatment in kinetic-type Monte Carlo method is summarized in Fig. 1.

3. Numerical validations

In this section, the numerical framework introduced in Section 2 is validated by simulating cross-plane phonon transport through single-layer thin films in Fig. 2(a), made of Si, Al, and Ge separately, and cross-plane interfacial phonon transport through bi-layer thin films in Fig. 2(b), including Al/Si and Ge/Si respectively, at 300 K. The dispersion relations and relaxation time expressions of the three materials are provided in Appendix B. A benchmark for Monte Carlo simulation of interfacial phonon transport between dissimilar materials with frequency-dependent transmission is challenging mainly due to the following two reasons: (i) a comparison to the experimental data is often not direct because of many unknown influencing factors in realistic interface system such as the roughness and disorder; (ii) the available analytical expression of Kapitza conductance (known as Landauer formalism [1]) is derived based on the difference of emitted phonon temperature at the interface, which is difficult to calculate in Monte Carlo method. Therefore, we firstly calibrate a deterministic solver for interfacial phonon transport using discrete-ordinates method (DOM), which is capable of computing the emitted phonon temperature and thus Landauer's Kapitza conductance. Then the DOM numerical results of both temperature distributions across the interface system and the Kapitza conductance based on the usual equivalent equilibrium temperature are adopted as a benchmark to validate the Monte Carlo scheme. The general principle and procedure in DOM solution of phonon Boltzmann equation can be found in our recent work [42]. We extend the DOM scheme to interfacial phonon transport with SDMM model as given in Appendix C.

3.1. Cross-plane phonon transport through thin film

In order to validate our basic kinetic-type Monte Carlo code, we firstly simulate the cross-plane phonon transport through single-layer thin films by comparing the numerical result to that of discrete-ordinates method. Full absorption and diffuse emission

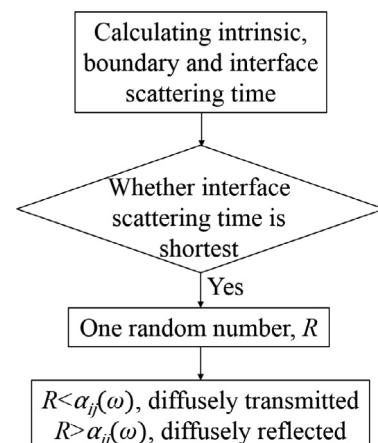


Fig. 1. The flow chart of the interface treatment in kinetic-type Monte Carlo method. After the energy packet scatters with interface, it will be diffusely transmitted or diffusely reflected.

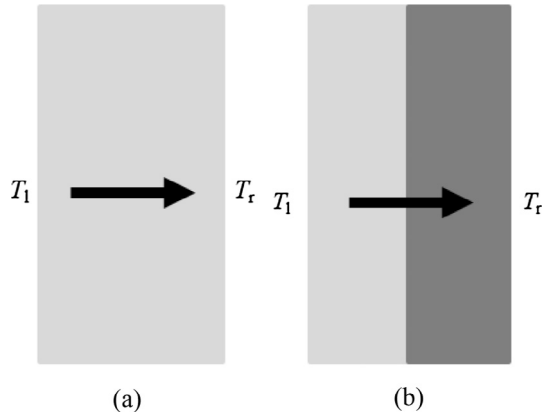


Fig. 2. Schematic of cross-plane phonon transport through a single thin film and cross-plane interfacial phonon transport through bilayer thin films: (a) single-layer thin films, made of Si, Al, and Ge separately; (b) bi-layer thin films, including Al/Si system and Ge/Si system respectively.

are applied to treat the isothermal boundaries at both ends of the thin film. The system is initially maintained at 299 K. Then the temperature of the left-hand boundary increases to 301 K, whereas the right-hand boundary keeps at 299 K. After a sequence of evolution, the system will approach to steady state. The temperature distributions and effective thermal conductivity λ_{eff} , are computed when the system reaches a steady state. The numerical parameters in Monte Carlo simulation for the three cases of Si, Al, and Ge thin film with different thickness are given in Table 1. For all the cases, the referenced equilibrium temperature is chosen as $T_{\text{eq}} = 300\text{K}$.

Fig. 3 shows a comparison between the results by the present kinetic-type Monte Carlo method and those by the discrete-ordinates method. Due to the frequency-dependent phonon mean free path (MFP) dictated by the spectral feature of phonon group velocity and relaxation time, an average Knudsen number is introduced as [43]:

$$\langle Kn \rangle = \frac{\bar{\Lambda}}{L}, \quad (7)$$

with the average MFP defined as: $\bar{\Lambda} = \sum_p \int_{\omega} \hbar \omega D \frac{df_{\text{eq}}}{dT} v_g \tau d\omega$

$\sum_p \int_{\omega} \hbar \omega D \frac{df_{\text{eq}}}{dT} d\omega$ and L the thin film thickness. The temperature distributions within thin film are approximately linear with larger temperature jump at boundary at increasing average Knudsen number. The boundary temperature jump comes from the non-equilibrium interaction between the hot/cold sources and the thin film due to sub-continuum effect. Thus the effective cross-plane thermal conductivity decreases with decreasing thin film thickness or increasing average Knudsen number. The excellent agreement of the present Monte Carlo results with the discrete-ordinates solutions demonstrates the validity of our basic kinetic-type Monte Carlo program.

3.2. Cross-plane interfacial phonon transport

In this sub-section, we aim to validate our interface treatment scheme in Monte Carlo method through modeling the cross-

plane interfacial phonon transport through Al/Si and Ge/Si bi-layer thin films around room temperature. In both two cases of bi-layer thin film, the volume ratio of the two materials forming the interface is 1. When the system reaches steady state after a sequence of evolution, the temperature distributions and Kapitza conductance are computed. The numerical parameters in Monte Carlo simulation for three cases of Al/Si and Ge/Si bilayer thin film systems with different total thickness are given in Table 2.

The comparison between the present results by Monte Carlo method and that by the discrete-ordinates method is shown in Fig. 4 for Al/Si system and in Fig. 5 for Ge/Si system respectively. The Kapitza conductance is defined as: $G = q/\Delta T$, where q is the heat flux across the interface and ΔT denotes the temperature jump across the interface based on the equivalent equilibrium temperature. The equivalent equilibrium temperature is a representation of the total energy of all phonons in a cell thermalized into an equivalent equilibrium state. The present numerical results by kinetic-type Monte Carlo method with the SDMM interface treatment agree well with those by the discrete-ordinates solution. As the total thickness of thin film decreases, the temperature jump across the interface increases due to the strong non-equilibrium effect.

In addition, the semi-spectral interface phonon model developed in Ref. [27] is also adopted to validate the present Monte Carlo method for interface treatment:

$$\alpha_{12} = \frac{\frac{2}{\sum_p \int_{\omega \in (1,2)} \hbar \omega D_1 \frac{df_{\text{eq}}}{dT} v_{1g} d\omega}}{\frac{1}{\sum_p \int_{\omega \in 1} \hbar \omega D_1 \frac{df_{\text{eq}}}{dT} v_{1g} d\omega} + \frac{1}{\sum_p \int_{\omega \in 2} \hbar \omega D_2 \frac{df_{\text{eq}}}{dT} v_{2g} d\omega} + \frac{1}{2G}}. \quad (8)$$

In this model, G is an input Kapitza conductance to extract a constant transmission coefficient from material 1 to material 2, whereas a frequency-dependent transmission coefficient from material 2 to material 1. Based on this interface phonon model and the material data from Ref. [43] for Al/Si interface, the present Monte Carlo scheme produces a Kapitza conductance $G = 110.27 [\text{MW}/\text{m}^2 \cdot \text{K}]$, which shows a good agreement with the targeted input Kapitza conductance $110 [\text{MW}/\text{m}^2 \cdot \text{K}]$ within acceptable numerical error.

4. Results and discussion

In this section, the validated Monte Carlo framework is applied to study two important effects in interfacial phonon transport: size effect and interface roughness effect on Kapitza conductance.

4.1. Size effect on Kapitza conductance

The size effect in nanoscale heat transport refers to the destruction of thermal properties when decreasing the dimension of system. In comparison to extensive and conclusive studies on the size effect of thermal conductivity of nanostructures [44–47], the size effect of Kapitza conductance is relatively rarely considered with still no consistent conclusions in previous work [48–50]. A non-linear relation between the Kapitza resistance and inverse of total system thickness was declared in some work [48], while the linearly increasing or decreasing relations between Kapitza resistance and the inverse of total system thickness have been found

Table 1

Numerical parameters in Monte Carlo simulation for the three cases of Si, Al and Ge thin film.

Material	Si			Al			Ge		
Film thickness (nm)	4	70	700	4	70	700	4	70	700
Sample number (million)	20	50	190	56	80	320	56	200	190

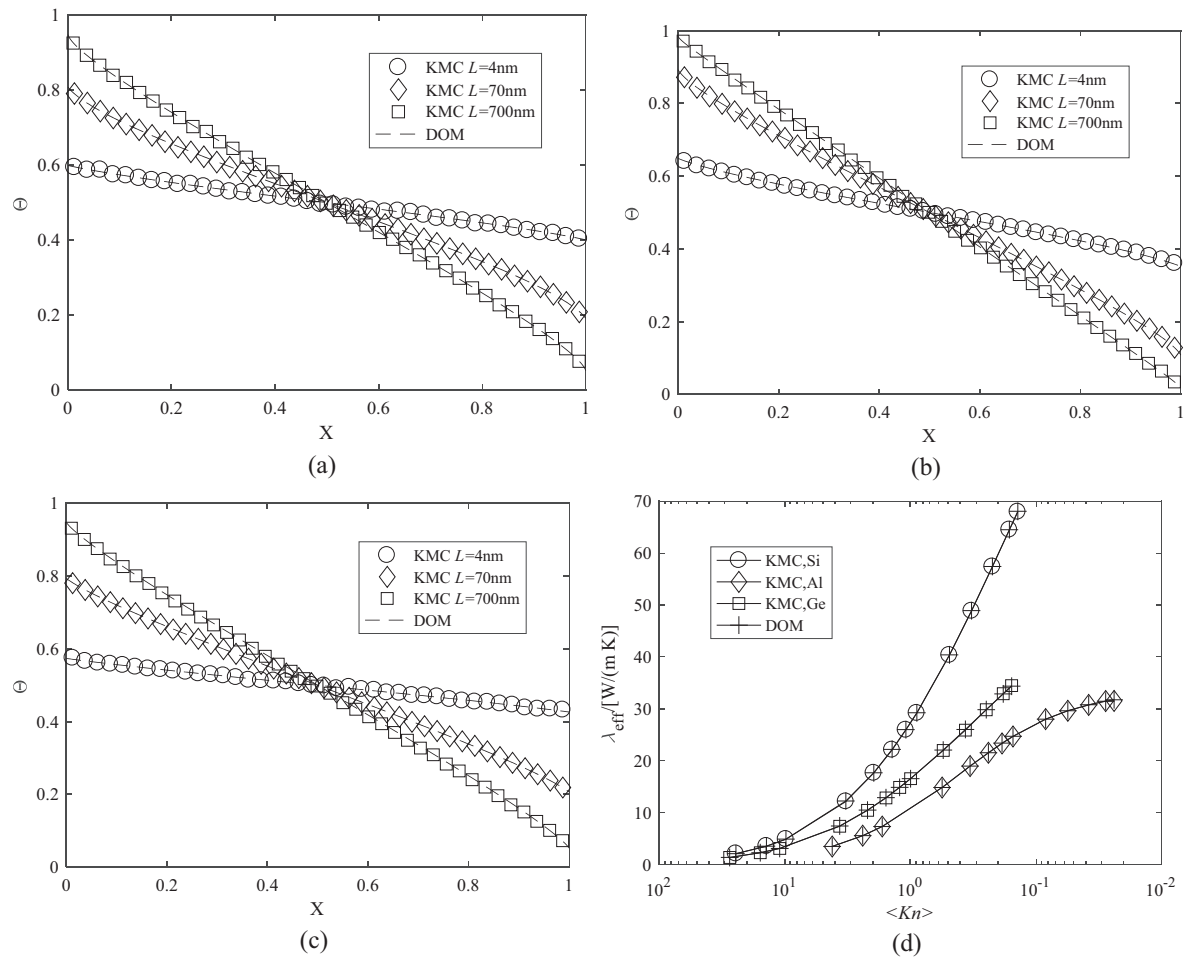


Fig. 3. The non-dimensional temperature distributions and the effective thermal conductivity of cross-plane phonon transport through Si, Al, and Ge single-layer thin films: (a), (b), and (c) are non-dimensional temperature distributions for Si, Al, and Ge, including three cases with a thickness 4 nm, 70 nm, and 700 nm respectively. Symbols are results of the present kinetic-type Monte Carlo method whereas dash lines are that of discrete-ordinates method introduced in Appendix C; (d) is effective thermal conductivity for Si, Al and Ge thin films versus average Knudsen number.

Table 2

Numerical parameters in Monte Carlo simulation for three cases of Al/Si and Ge/Si bi-layer thin film system.

Material	Al/Si			Ge/Si		
Total thickness of film (nm)	20	120	220	20	120	220
Sample number (million)	50	80	100	50	80	100

by Jones et al. [49] and Yang et al. [50] respectively. Therefore, it is essential to provide a clear physical picture and quantitative interpretation of the size effect on Kapitza conductance.

The calculation of Kapitza conductance depends on the temperature jump at the interface. Two different definitions of temperature have been used for evaluating the interfacial temperature jump: (i) the equivalent equilibrium temperature (referred as definition I), and (ii) the emitted phonon temperature (referred as definition II). Definition I has been given in Section 3.2, whereas definition II at the interface is defined as conjectured phonons emitted ballistically from a boundary at equilibrium temperature T_e with infinite MFP (detailed interpretation and calculation can be found in Appendix C). Definition I has been widely used in numerical methods where the equivalent equilibrium temperature is obtained through statistical summation of the energy density within a simulation cell. In comparison, the classical analytical expression of Kapitza conductance, known as Landauer formalism, between two dissimilar materials is derived based on definition II [1]:

$$G_e = \frac{1}{4} \sum_p \int_0^{\omega_m} v_{1g} \alpha_{12} C_{\omega 1} d\omega. \quad (9)$$

As it is difficult to establish an analytical relation between the temperature differences based on definition I and definition II except under gray Debye's approximation [14,51] or other simplified assumption [27], one can hardly derive an analytical Kapitza conductance based on definition I.

The results of Kapitza conductance for Al/Si and Ge/Si interfaces at around room temperature are shown in Fig. 6(a) and (b) respectively. The Kapitza conductance based on definition I is found to be considerably higher than that based on definition II. We thus infer that the temperature difference based on definition I is smaller than that based on definition II, as qualitatively consistent with previous simplified results [14,51]. Furthermore, the Kapitza conductance increases with decreasing the inverse of total system thickness based on definition I and approaches a constant value at a sufficiently large system thickness, while no size effect is

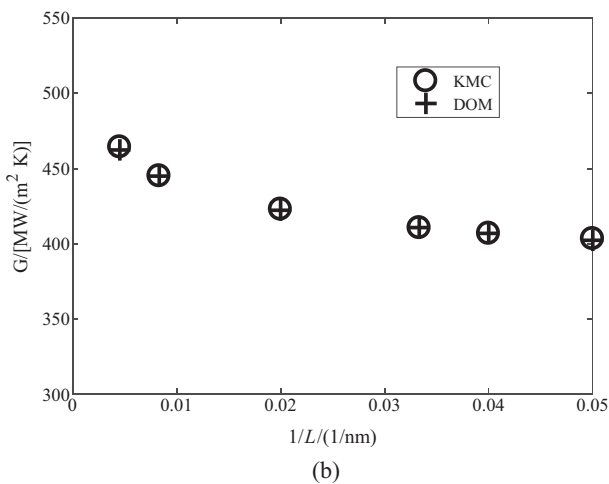
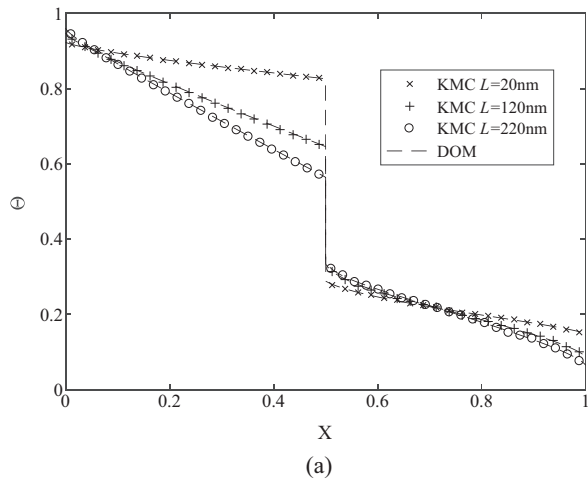


Fig. 4. The non-dimensional temperature distribution and Kapitza conductance in cross-plane interfacial phonon transport through Al/Si bi-layer thin film: (a) non-dimensional temperature distribution for three cases with a total thickness 20 nm, 120 nm and 220 nm respectively; (b) Kapitza conductance versus the inverse of total thickness separately.

obtained when definition II is adopted. The different results based on the two definitions can be understood as below. Definition I represents the energy density of non-equilibrium phonons near the interface, the deviation of which from local equilibrium state is dependent on the total system size. In comparison, definition II is an idealized one for phonons with infinite MFP independent of the system dimension.

The present results have twofold significant indications. Firstly, the controversial relations in the literature between the Kapitza conductance based on definition I and the inverse of total system thickness may be attributed to the simulated thickness range and interface disorder. This relation is nearly linear within the range of small total system thickness, and will become slightly nonlinear at larger system size. The increasing Kapitza conductance with decreasing system size may be caused by the influence from interfacial atomic disorder softening the abrupt vibrational mismatch therein [50]. Secondly, the wide application of Eq. (9) based on emitted phonon temperature in theoretical prediction of interfacial phonon transport in much existing literature remains to be deliberated. As the experimental measurement of Kapitza conductance at around room temperature or even higher temperature is based on the equivalent equilibrium temperature [1], the underestimation of the measured Kapitza conductance by Eq. (9) [19,40,52] is only an artifact due to the inconsistent use of the definition of tem-

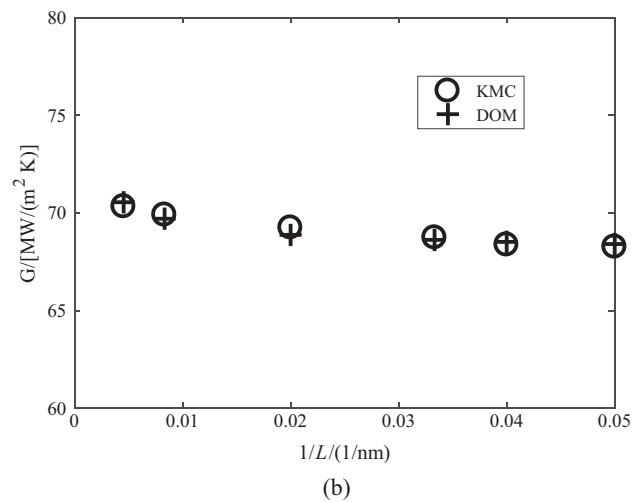
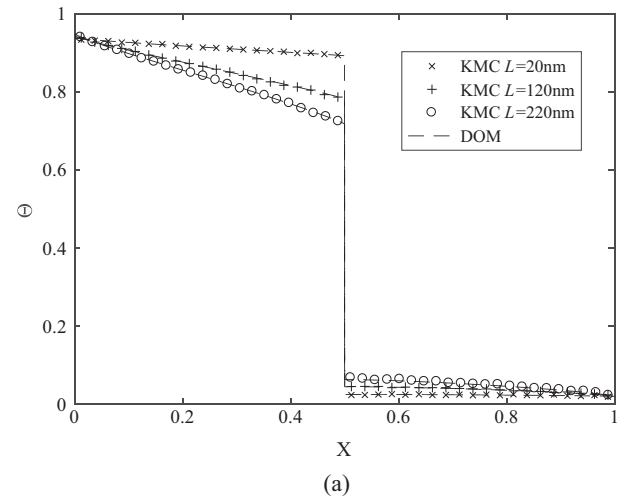


Fig. 5. The non-dimensional temperature distribution and Kapitza conductance in cross-plane interfacial phonon transport through Ge/Si bi-layer thin film: (a) the non-dimensional temperature distribution for three cases with a total thickness 20 nm, 120 nm and 220 nm respectively; (b) Kapitza conductance versus the inverse of total thickness.

perature difference. The predicted Kapitza conductance (~ 400 MW/m² K) by our kinetic-type Monte Carlo method based on definition I is actually very close to the experimental result (345 ± 40 MW/m² K) at 300 K [19] considering the neglected inelastic interface phonon scattering in our present model.

4.2. Interface roughness effect

As asperity is common at material interface from both the manufacturing and application processes, it is crucial to understand the influence of interface roughness on Kapitza conductance. Although some experimental works [53–55] have indicated that the roughness can reduce the Kapitza conductance, the physical mechanism and quantitative description remain elusive. Two aspects of the interface roughness effect are studied in this sub-section: the interfacial area ratio and the roughness height. The interfacial area ratio is defined as the ratio of the true interfacial area to the interfacial area projected along the mean normal vector to the interface (i.e. the overall transport direction).

Heat transport across Al/Si interface is considered with different types of asperity geometries: triangle, square, wavelike, rectangle, cross, T-type and random roughness as shown in Fig. 7. The total

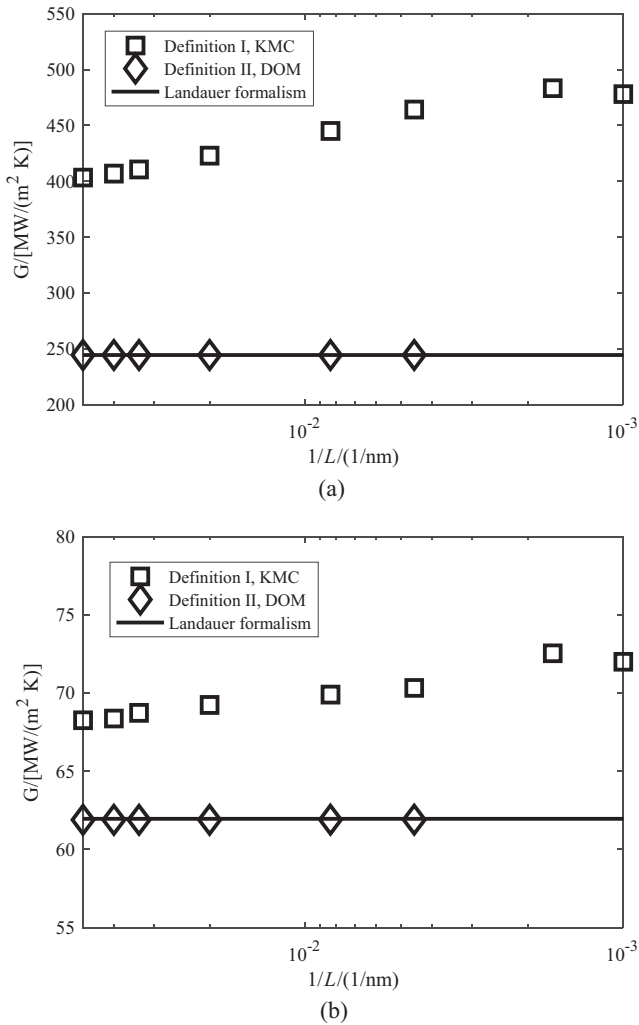


Fig. 6. Kapitza conductance versus system total thickness at room temperature ($T = 300 \text{ K}$): (a) Al/Si interface, and (b) Ge/Si interface. The squares represent the results based on definition I (equivalent equilibrium temperature), whereas the diamonds represent the results based on definition II (emitted phonon temperature).

thickness of the simulated interface system between $T_l = 300.5 \text{ K}$ and $T_r = 299.5 \text{ K}$ is 50 nm with a volume ratio 1. The interface roughness height is fixed at 6 nm in studying the effect of interfacial area ratio. The random roughness is generated following the algorithm in our previous work [56], with two main steps: (i) choose the directional growth probability $D_x = 3 \times 10^{-5}$ and $D_y = 1 \times 10^{-5}$, and randomly locate one seed within every 7 nm length along the lateral direction at 28 nm along the longitudinal direction; (ii) let the right-hand material randomly grow into the left-hand one from these seeds until the asperity height reaches 6 nm and the volume ratio of two materials becomes 1. To eliminate the statistical noise, four random structures with one cell size $1 \text{ nm} \times 1 \text{ nm}$ are generated based on the same growing parameters to yield an average result. The periodic boundary condition is applied along the lateral direction to realize infinite simulation system. Seven units of the periodic element are simulated for all the six regular interfaces to have a consistent comparison to the random interface. The rough interface is piled up by four kinds of interfaces as shown in Fig. 8. We go through all vertical and horizontal interfaces and justify which interface will intersect with the trajectory of the tracking packet. Then this packet will encounter with the nearest interface.

The effective thermal conductivity of the interface system is calculated from the steady-state heat flux distribution and the exerted temperature difference based on the Fourier's law: $\lambda_{\text{eff}} = (\int_0^{L_y} q_x dy \cdot L) / [L_y(T_l - T_r)]$ and given in Fig. 9(a), where L_y is the width of simulated system along y direction. To calculate the effective Kapitza conductance of the rough interface, we present a one-dimensional model ignoring lateral heat transport, with the series relation for thermal resistance [57]:

$$\frac{L}{\lambda_{\text{eff}}} = R_0 + \frac{1}{G_{\text{eff}}}, \quad (10)$$

where R_0 includes both the intrinsic and boundary thermal resistance of the material pair and G_{eff} is defined as an effective Kapitza conductance. The intrinsic and boundary thermal resistance in rough interface is assumed the same as that in the smooth interface with the same total system thickness and volume ratio. In this way, the effective Kapitza conductance for various rough interfaces are computed based on Eq. (10), and shown in Fig. 9(b). The results show that both the effective thermal conductivity and Kapitza conductance will decrease with increasing interfacial area ratio. This is due to larger interfacial area ratio creates stronger interface scattering besides interfacial vibrational mismatch, which results in larger reduction of effective Kapitza conductance.

For the convenience of engineering application, we build up an empirical relation between the Kapitza conductance in bi-layer thin film and the interfacial area ratio. A power law dependence on the interfacial area ratio is assumed for the total thermal resistance of the interface system as below:

$$\frac{L}{\lambda_{\text{eff}}} = L \left[A(S - 1)^B + \frac{1}{\lambda_{\text{smooth}}} \right], \quad (11)$$

where A and B are the fitting parameters, S denoting the interfacial area ratio, and λ_{smooth} being the effective thermal conductivity of bi-layer thin film with smooth interface. The dimension of parameter A is $[\text{m K/W}]$, whereas the parameter B is dimensionless. In the limiting case of $S = 1$, Eq. (11) recovers well the result for smooth interface. The values are obtained as 0.00233 and 3.66946 respectively for parameters A and B through fitting the various numerical results for different types of designed interface roughness in Fig. 9(a). With the help of both Eqs. (10) and (11), the empirical expression for Kapitza conductance is derived as:

$$\frac{1}{G_{\text{eff}}} = LA(S - 1)^B + \frac{1}{G_{\text{smooth}}}, \quad (12)$$

where G_{smooth} denotes the Kapitza conductance for the smooth interface case. With the obtained values for parameters A and B through fitting the total thermal resistance, Eq. (12) gives the Kapitza conductance in good agreement with the numerical result by the present Monte Carlo simulation, as shown in Fig. 9(b).

Finally we study the effect of roughness height on the Kapitza conductance. The Al/Si and Ge/Si interfaces with square and T-type roughness are taken into account with fixed interfacial ratio at 2 and 3 respectively. The following roughness heights are considered respectively: 2 nm , 4 nm , 6 nm , 8 nm , 10 nm for both cases. To guarantee a fixed interfacial ratio, the corresponding structures have been derived by enlarging or narrowing the same structure with a roughness height 6 nm . For instance, the structure with roughness height 4 nm is derived by narrowing each edge of interface $2/3$ times from that with a roughness height 6 nm . For each case, only one periodic element is simulated. The Kapitza conductance drops sharply from the smooth interface and keeps nearly at a constant with increasing roughness height, as shown in Fig. 10. The roughness height is a relatively unimportant factor to influence the Kapitza conductance compared to the interfacial area ratio. Previous experimental works [53–55] suggest lower

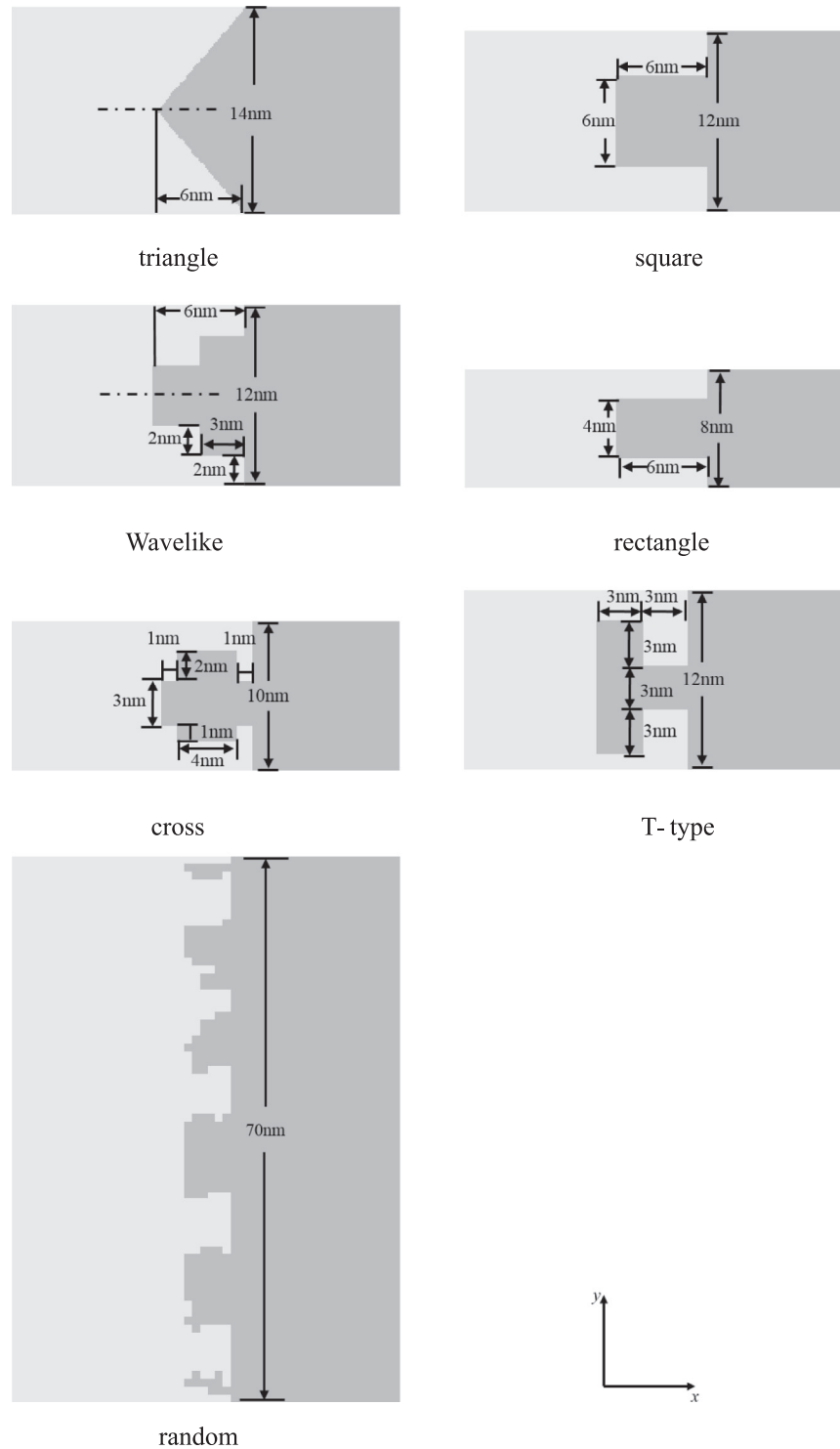


Fig. 7. Illustration of various interface roughness types considered in present work, including triangle, square, wavelike, rectangle, cross, T-type and random interface roughness.

Kapitza conductance of interfacial structures at larger roughness height. This discrimination can be explained by another parameter: correlation length. In the present study, a larger correlation length is obtained when increasing the roughness height to keep a constant interfacial area ratio. In previous experimental studies [53–55], a smaller correlation length or constant one is often corresponding to a larger roughness height. As a benefit, the empirical

relation Eq. (12) between the Kapitza conductance and interfacial area ratio is available for various roughness heights approximately.

5. Conclusions

In summary, we present a Monte Carlo framework to model interfacial phonon transport between dissimilar materials with a

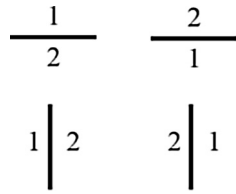


Fig. 8. Schematic of four kinds of interface: vertical interfaces and horizontal interfaces, formed by material 1 and material 2.

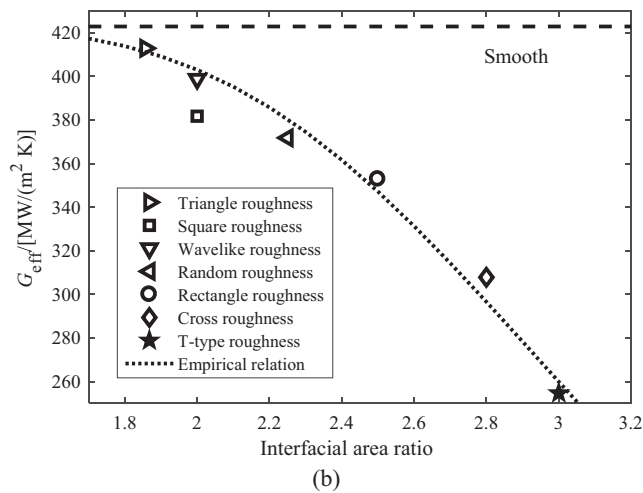
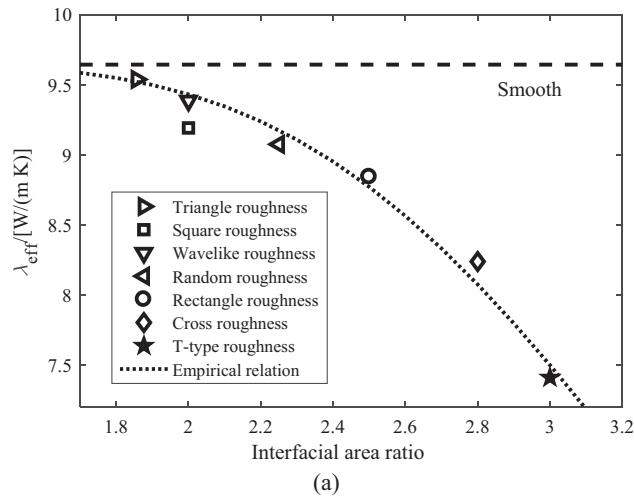


Fig. 9. The effective thermal conductivity and effective Kapitza conductance versus interfacial area ratio: (a) thermal conductivity, (b) Kapitza conductance, right triangle is triangle roughness, square is square roughness, down triangle is wavelike roughness, left triangle is random roughness, circle is rectangle roughness, diamond is cross roughness, star is T-type roughness, dash line is smooth interface and dot line is the empirical relation.

frequency-dependent transmissivity based on the spectral diffuse mismatch model. After careful validations, the present Monte Carlo framework is applied to study the size effect on Kapitza conductance and the interface roughness effect. For the size effect, the results show that: the Kapitza conductance based on the equivalent equilibrium temperature is slightly influenced by system size while this size effect vanishes based on the emitted phonon temperature. Furthermore, the Kapitza conductances based on two temperature definitions are considerably different, which reveals the importance of using a consistent definition in comparing theoretical prediction to experimental results. For the interface

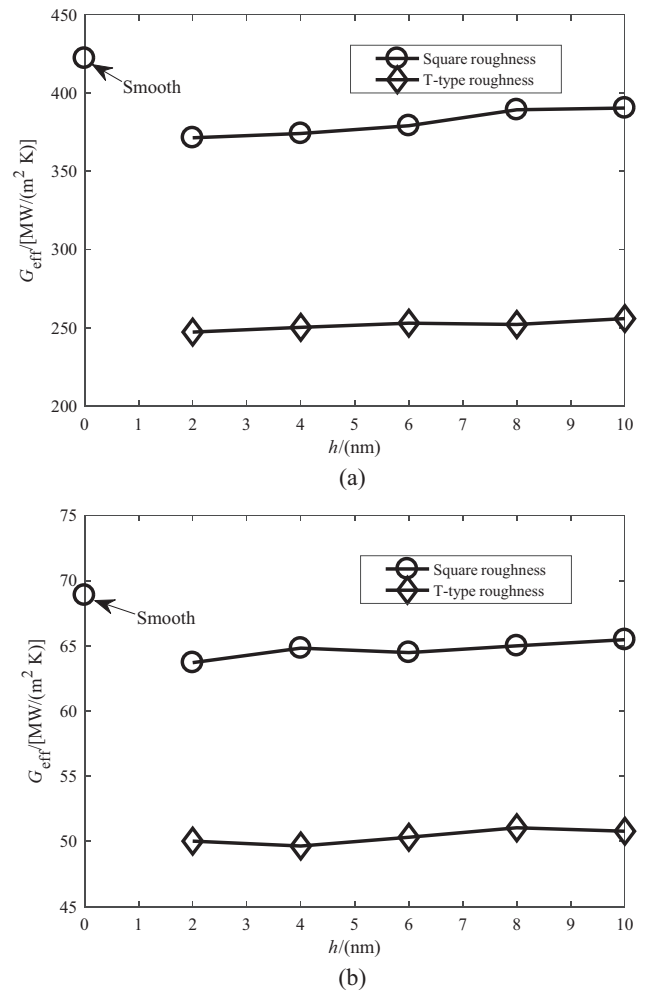


Fig. 10. The Kapitza conductance as a function of roughness height, h , for square roughness and T-type roughness: (a) Al/Si interface and (b) Ge/Si interface.

roughness effect, the results indicate that the Kapitza conductance is mainly influenced by interfacial area ratio defined as the ratio of the true interfacial area to the interfacial area projected along the overall transport direction, while slightly influenced by the interface roughness height. Furthermore, we establish an empirical relation between the Kapitza conductance and interfacial area ratio for convenient engineering application. The conclusions of present work are helpful for the design of interface structure in nanosystems with optimized thermal performance.

Conflict of interest

We declare that there is no conflict of interests for this work.

Acknowledgements

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Appendix A. Kinetic-type Monte Carlo method

Kinetic-type Monte Carlo (KMC) method is a simplified edition of energy-based variance reduction Monte Carlo scheme under the assumption of small temperature difference in the heat transport

process [34]. Under this assumption, the deviational energy distribution and pseudo-equilibrium energy distribution is independent on temperature and pseudo-temperature once normalized [34]. This means the initialization, scattering and boundary emission of phonon can be performed without the knowledge of the thermodynamic temperature and local pseudo-temperature. This leads to a key feature of kinetic-type Monte Carlo method that energy packets with an effective deviational energy $\varepsilon_{\text{eff}}^d$ are tracked one by one, different from traditional phonon Monte Carlo method. The effective deviational energy is specified as the ratio of total deviational energy amount through all simulation time to the total number N of simulated energy packets. The total deviational energy amount includes initial term, boundary term and source term (only former two parts is needed in the present work) [34].

These energy packets are treated independently along their trajectories accumulated by linear segments separated by intrinsic scattering and scattering with boundaries and interfaces [34]. This process tracking one energy packet is described as follows:

- (i) Draw the properties (frequency, polarization, group velocity, sign, initial position and initial time) of one energy packet from the equilibrium distribution of initial term or boundary term;
- (ii) This energy packet then travels in the system until it exits the isothermal boundary or the simulation time domain for the time-dependent case by repeating the following steps [34]: (a) Calculate the time step between the present scattering event and next intrinsic scattering event as $\Delta t = -\tau(\omega, \mathbf{p}, T_{\text{eq}}) \ln(1 - R)$, where R is a uniform random number between 0 and 1; (b) if there is no scattering with boundaries or interfaces during this time step, the next scattering event happens at time $\tilde{t} = t + \Delta t$ and the position of the energy packet at next scattering event is $\tilde{\mathbf{x}} = \mathbf{x} + \mathbf{v}_g \Delta t$, where t and $\mathbf{x}$ denote the time and position of the energy packet at this scattering event; the properties, including frequency, polarization, and group velocity, of this energy packet will be reset from the deviational pseudo-equilibrium energy distribution:

$$\phi = \frac{(T_{\text{loc}} - T_{\text{eq}})D(\omega, \mathbf{p})}{\tau(\omega, \mathbf{p}, T)} \frac{d\varepsilon_{\text{eq}}^d}{dT}, \quad (\text{A1})$$

(c) if a boundary or interface is encountered during this time step, the position $\tilde{\mathbf{x}}$ of this energy packet at next scattering event will be set as the position where this packet encounters with the boundary or interface and the time $\tilde{t}$ of this energy packet at next scattering event is set as the time when this energy packet encounters with the boundary or interface; the resetting of other properties depends on the type of boundaries and interface (for example: diffuse reflection, specular reflection, periodic reinjection, diffuse transmission and so on), and isothermal boundary just terminates the tracking operation of this energy packet;

- (iii) The macroscopic information is calculated at time t_{count} by accumulating the contribution of each energy packet to it at this time; for instance, the contribution of one energy packet to temperature is $s_i \varepsilon_{\text{eff}}^d / C_V V$ and its contribution to heat flux along x direction is $s_i \varepsilon_{\text{eff}}^d v_{i,x} / V$, where s_i is the sign of this energy packet, i , C_V being the heat capacity derived from the phonon dispersions, V the volume of region needing to count the information, and $v_{i,x}$ is velocity component of this energy packet along x direction; thus the temperature and heat flux distributions at counting time t_{count} are obtained by summing up the contributions from all the energy packets based on $T = T_{\text{eq}} + \sum_i s_i \varepsilon_{\text{eff}}^d / C_V V$ and $q = \sum_i s_i \varepsilon_{\text{eff}}^d v_{i,x} / V$.

Appendix B. Numerical data for phonon dispersion relations and scattering rates

In the simulation throughout this study, we use data for the relaxation time and for the isotropic [100] dispersion relations of acoustic mode for three materials, Si, Al and Ge and ignore the contributions of optical mode to heat transport due to its small group velocity compared with acoustic mode.

The dispersion relations of all materials are expressed as a fourth degree polynomial equation. The expression for Si is from Ref. [58]:

$$\omega(k) = A_4 k^4 + A_3 k^3 + A_2 k^2 + A_1 k, \quad (\text{B1})$$

where A_1 , A_2 , A_3 and A_4 are parameters through fitting the experimental measurements. For transverse acoustic (TA) polarization, the four parameters are: $A_1 = 5.511 \times 10^3 \text{ m s}^{-1}$, $A_2 = -1.169 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$, $A_3 = -4.957 \times 10^{-17} \text{ m}^3 \text{ s}^{-1}$, $A_4 = 2.432 \times 10^{-27} \text{ m}^4 \text{ s}^{-1}$; for longitudinal acoustic (LA) polarization: $A_1 = 8.192 \times 10^3 \text{ m s}^{-1}$, $A_2 = -1.140 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$, $A_3 = -2.612 \times 10^{-18} \text{ m}^3 \text{ s}^{-1}$, $A_4 = -5.645 \times 10^{-29} \text{ m}^4 \text{ s}^{-1}$. The experimental data of dispersion relation for Al are taken from Ref. [59] and fitted by:

$$\omega(K) = B_4 K^4 + B_3 K^3 + B_2 K^2 + B_1 K, \quad (\text{B2})$$

where B_1 , B_2 , B_3 and B_4 are fitting parameters and $K = k/k_{\text{max}}$ with $k_{\text{max}} = 2\pi/a$, a being the cubic lattice constant. For transverse acoustic (TA) polarization, the four parameters are $B_1 = 5.147 \times 10^{13} \text{ rad/s}$, $B_2 = 9.578 \times 10^{12} \text{ rad/s}$, $B_3 = -3.204 \times 10^{13} \text{ rad/s}$, $B_4 = 6.750 \times 10^{12} \text{ rad/s}$; for longitudinal acoustic (LA) polarization, four parameters are: $B_1 = 9.825 \times 10^{13} \text{ rad/s}$, $B_2 = 2.226 \times 10^{13} \text{ rad/s}$, $B_3 = -1.216 \times 10^{14} \text{ rad/s}$, $B_4 = 6.208 \times 10^{13} \text{ rad/s}$. The experimental data of dispersion relation for Ge are taken from Ref. [60] and fitted by:

$$\omega(K) = B'_4 K^4 + B'_3 K^3 + B'_2 K^2 + B'_1 K, \quad (\text{B3})$$

For transverse acoustic (TA) polarization, four parameters are: $B'_1 = 2.08583 \times 10^{13} \text{ rad/s}$, $B'_2 = -1.83927 \times 10^{11} \text{ rad/s}$, $B'_3 = -2.39121 \times 10^{12} \text{ rad/s}$, $B'_4 = -3.5444 \times 10^{12} \text{ rad/s}$; for longitudinal acoustic (LA) polarization, four parameters are: $B'_1 = 4.72355 \times 10^{13} \text{ rad/s}$, $B'_2 = -8.06266 \times 10^{12} \text{ rad/s}$, $B'_3 = 4.32529 \times 10^{12} \text{ rad/s}$, $B'_4 = -3.00242 \times 10^{12} \text{ rad/s}$.

For Si, the relaxation time expressions are taken from Ref. [58]: Umklapp scattering rate $\tau_a^{-1} = C_1 T \omega^2 \exp(-C_2/T)$ and impurity scattering rate $\tau_i^{-1} = C_3 \omega^4$. The parameters are the same for both phonon polarizations: $C_1 = 1.4 \times 10^{-19} \text{ s K}^{-1}$, $C_2 = 152 \text{ K}$, $C_3 = 1.32 \times 10^{-45} \text{ s}^3$ respectively. The total relaxation time of Si is derived from the Matthiessen's rule: $\tau^{-1} = \tau_a^{-1} + \tau_i^{-1}$. For Al, the total relaxation time is taken from Ref. [24,27] a $\tau = 10 \text{ ps}$ which yields the desired lattice thermal conductivity. For Ge, we adopt the same relation time expressions as those of Si for both Umklapp scattering rate, $\tau_a^{-1} = D_1 T \omega^2 \exp(-D_2/T)$ and impurity scattering rate $\tau_i^{-1} = D_3 \omega^4$ [60]. The corresponding constants are chosen as: $D_1 = 3.35 \times 10^{-19} \text{ s K}^{-1}$, $D_2 = 57.6 \text{ K}$, $D_3 = 2.4 \times 10^{-44} \text{ s}^3$. The total relaxation time of Ge is also derived from the Matthiessen's rule: $\tau^{-1} = \tau_a^{-1} + \tau_i^{-1}$.

In the numerical simulation, the frequency domain of single material or a material pair is discretized into 1500 uniform units between 0 and maximum frequency (for single material, maximum frequency is the maximum value of its own frequencies while for one material pair, maximum frequency is the maximum value of all of their frequencies). Other properties are thus calculated at each frequency respectively.

Appendix C. Discrete-ordinates method with exact phonon dispersion relations

Multiplying $v_g \hbar \omega D(\omega, p)/4\pi$ in both sides of Eq. (1), a deviational form of EPRT [45] is derived as:

$$v_g \cos \theta \frac{dI_{\omega,p}^d}{dx} = -\frac{I_{\omega,p}^d - (I_{\text{loc},\omega,p}^{\text{eq}} - I_{T_{\text{eq},\omega,p}}^{\text{eq}})}{\tau(\omega, p, T)}, \quad (\text{C1})$$

with the spectral deviational phonon intensity $I_{\omega,p}^d = I_{\omega,p} - I_{T_{\text{eq},\omega,p}}^{\text{eq}}$, and the assumed equilibrium intensity $I_{T_{\text{eq},\omega,p}}^{\text{eq}} = v_g \hbar \omega f_{T_{\text{eq}}}^{\text{eq}} D(\omega, p)/4\pi$. Due to the similarity of discretization of Eq. (C1) inside each material shown in Fig. A1, we just take the left material of interface as an example to explain this process and then give the energy balance conditions at the interface. The discretized space units uniformly divide the system with sampling points in their middle positions. As Fig. A1 shows, the interface is located at the middle of system which is the right side of last unit in material 1, which can avoid sampling at the interface where temperature is discontinuous.

For the inner points in materials 1, when $\cos \theta_j > 0$, the forward difference scheme is applied and the discretized form of Eq. (C1) is:

$$v_g \cos \theta_j \frac{I_{\omega,p,i,j}^d - I_{\omega,p,i-1,j}^d}{\Delta x} = -\frac{I_{\omega,p,i,j}^d - (I_{\text{loc},\omega,p,i,j}^{\text{eq}} - I_{T_{\text{eq},\omega,p,i,j}}^{\text{eq}})}{\tau(\omega, p, T)}, \quad (\text{C2})$$

where $I_{\omega,p,i,j}^d$ and $I_{\text{loc},\omega,p,i,j}^{\text{eq}}$ are the spectral deviational intensity and the pseudo deviational intensity of phonon in unit i and at j direction, θ_j polar angle value at j direction, Δx the size of one unit. Defining $m_1 = (\cos \theta_j v_g \tau)/\Delta x$, Eq. (C2) can be arranged as:

$$I_{\omega,p,i,j}^d = \frac{(I_{\text{loc},\omega,p,i,j}^{\text{eq}} - I_{T_{\text{eq},\omega,p,i,j}}^{\text{eq}}) + m_1 I_{\omega,p,i-1,j}^d}{1 + m_1}. \quad (\text{C3})$$

When $\cos \theta_j < 0$, the final form of Eq. (C1) after discretizing with the backward difference scheme is:

$$I_{\omega,p,i,j}^d = \frac{(I_{\text{loc},\omega,p,i,j}^{\text{eq}} - I_{T_{\text{eq},\omega,p,i,j}}^{\text{eq}}) - m_1 I_{\omega,p,i+1,j}^d}{1 - m_1}. \quad (\text{C4})$$

For the left boundary treatment, i.e. discretizing Eq. (C1) from $1/2$ to 1 , similar to derivation above, the final form using the forward difference scheme is:

$$I_{\omega,p,1,j}^d = \frac{(I_{\text{loc},\omega,p,1,j}^{\text{eq}} - I_{T_{\text{eq},\omega,p,1,j}}^{\text{eq}}) + m_2 I_{\omega,p,1/2,j}^d}{1 + m_2}, \quad (\text{C5})$$

where $m_2 = 2m_1$, and the deviational intensity emitted from left isothermal boundary $I_{\omega,p,1/2,j}^d = I_{T_1,\omega,p}^{\text{eq}} - I_{T_{\text{eq},\omega,p}}^{\text{eq}}$ with the equilibrium intensity $I_{T_1,\omega,p}^{\text{eq}} = v_g \hbar \omega f_{T_1}^{\text{eq}} D(\omega, p)/4\pi$. With the aid of assumption of tiny temperature difference, this term can be linearized as: $I_{\omega,p,1/2,j}^d = (T_1 - T_{\text{eq}}) v_{1g} C_{\omega}/4\pi$ with the volume heat capacity per unit frequency $C_{\omega} = \hbar \omega D(\omega) \frac{df}{dT} \Big|_{T_{\text{eq}}}$. Then treat the interface boundary, i.e.

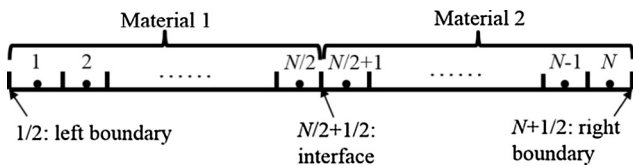


Fig. A1. The schematic of system of cross-plane interfacial phonon transport: the sampling point is at the middle of each unit. The left boundary, the interface and the right boundary are at the left side of the first unit and the right side of the last unit in material 1 and the right side of the last unit in material 2, denoted by $1/2$, $N/2 + 1/2$ and $N + 1/2$ point in the schematic.

discretize Eq. (C1) from $N/2$ to $N/2 + 1/2$ and from $N/2 + 1/2$ to $N/2$. The first case indicates $\cos \theta_j > 0$, and, applying the forward difference scheme, the final discretized form is:

$$I_{\omega,p,N/2+1/2,j}^d = \frac{(I_{\text{loc},\omega,p,N/2,j}^{\text{eq}} - I_{T_{\text{eq},\omega,p}}^{\text{eq}}) + (m_2 - 1) I_{\omega,p,N/2,j}^d}{m_2}. \quad (\text{C6})$$

The second case indicates $\cos \theta_j < 0$, and the final discretized form with the backward difference scheme is:

$$I_{\omega,p,N/2,j}^d = \frac{(I_{\text{loc},\omega,p,N/2,j}^{\text{eq}} - I_{T_{\text{eq},\omega,p}}^{\text{eq}}) - m_2 I_{\omega,p,N/2+1/2,j}^d}{1 - m_2}. \quad (\text{C7})$$

Finally, the energy balance is applied at the interface for individual phonon frequency and polarization:

$$\begin{aligned} \int_{\pi/2}^{\pi} (I_{1,\omega,p}^d)^- \cos \theta \sin \theta d\theta &= \alpha_{21}(\omega, p) \\ \int_{\pi/2}^{\pi} (I_{2,\omega,p}^d)^- \cos \theta \sin \theta d\theta - (1 - \alpha_{12}(\omega, p)) \\ &\int_0^{\pi/2} (I_{1,\omega,p}^d)^+ \cos \theta \sin \theta d\theta, \end{aligned} \quad (\text{C8})$$

$$\begin{aligned} \int_0^{\pi/2} (I_{2,\omega,p}^d)^+ \cos \theta \sin \theta d\theta \\ = -(1 - \alpha_{21}(\omega, p)) \int_{\pi/2}^{\pi} (I_{2,\omega,p}^d)^- \cos \theta \sin \theta d\theta + \alpha_{12}(\omega, p) \\ \times \int_0^{\pi/2} (I_{1,\omega,p}^d)^+ \cos \theta \sin \theta d\theta \end{aligned} \quad (\text{C9})$$

where $(I_{1,\omega,p}^d)^-$, $(I_{1,\omega,p}^d)^+$, $(I_{2,\omega,p}^d)^-$ and $(I_{2,\omega,p}^d)^+$ are the spectral deviational intensities of backward-going and forward-going phonons on left side of interface and backward-going and forward-going phonons on right side of interface respectively.

To get temperature, an equivalent equilibrium deviational intensity of phonon is defined as:

$$\begin{aligned} \sum_p \int_{\Omega} \int_0^{\omega_m} \frac{(f^{\text{eq}} - f_{T_{\text{eq}}}^{\text{eq}})}{4\pi} \hbar \omega D(\omega) d\omega d\Omega \\ = \sum_p \int_{\Omega} \int_0^{\omega_m} \frac{I_{\omega,p}^d}{v_g} d\omega d\Omega, \end{aligned} \quad (\text{C10})$$

where $d\Omega = \sin \theta d\theta d\phi$ is differential of solid angle. Under the assumption of tiny temperature difference, the left side of Eq. (C10) is linearized as an explicit function of temperature:

$$\sum_p \int_{\Omega} \int_0^{\omega_m} \frac{(T - T_{\text{eq}})}{4\pi} C_{\omega} d\omega d\Omega = \sum_p \int_{\Omega} \int_0^{\omega_m} \frac{I_{\omega,p}^d}{v_g} d\omega d\Omega. \quad (\text{C11})$$

Therefore, the temperature can be calculated by the following expression:

$$T = T_{\text{eq}} + \frac{\sum_p \int_0^{2\pi} \int_0^{\pi} \int_0^{\omega_m} \frac{I_{\omega,p}^d}{v_g} \sin \theta d\omega d\theta d\phi}{\sum_p \int_0^{\omega_m} C_{\omega} d\omega}. \quad (\text{C12})$$

Similarly, the calculation of pseudo-temperature is performed as [61]:

$$T_{\text{loc}} = T_{\text{eq}} + \frac{\sum_p \int_0^{2\pi} \int_0^{\pi} \int_0^{\omega_m} \frac{I_{\omega,p}^d}{v_g} \sin \theta d\omega d\theta d\phi}{\sum_p \int_0^{\omega_m} \frac{C_{\omega}}{\tau} d\omega}. \quad (\text{C13})$$

The emitted phonon temperatures mentioned in Section 5.1 are corresponding to the deviational intensities of forward-going phonons on left side of interface and backward-going phonons on right side of interface. The former case is illustrated as:

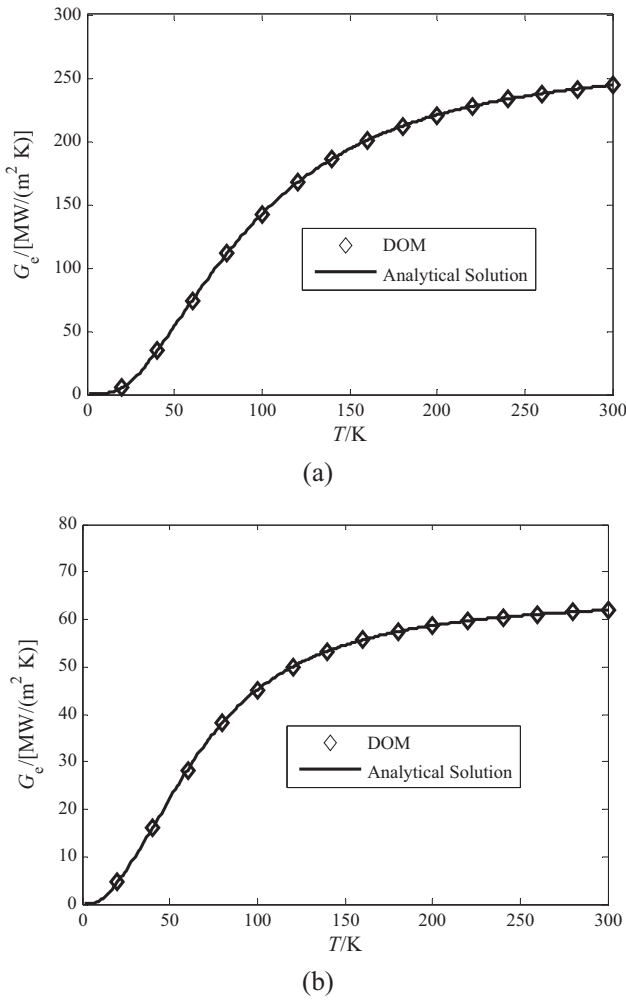


Fig. A2. Comparisons of thermal boundary conductance between discrete-ordinates method and analytical solution at different temperature: (a) Al/Si interface; (b) Ge/Si interface.

$$\begin{aligned} & \sum_p \int_0^{\omega_m} \int_0^{2\pi} \int_0^{\frac{\pi}{2}} \alpha_{12}(I_{1,\omega,p}^d)^+ \cos \theta \sin \theta d\omega d\theta d\varphi \\ &= \sum_p \int_0^{\omega_m} \int_0^{2\pi} \int_0^{\frac{\pi}{2}} \frac{\alpha_{12} \nu_{1g} \hbar \omega f^d(T_{e1}) D_1 \cos \theta \sin \theta}{4\pi} d\omega d\theta d\varphi, \end{aligned} \quad (C14)$$

with $f^d(T_{e1}) = f^{eq}(T_{e1}) - f_{T_{eq}}^{eq}$, T_{e1} being the temperature of the forward-going phonons on left side of interface. Applying the assumption of tiny temperature difference, T_{e1} can be extracted after linearization of right side of Eq. (C14) as:

$$T_{e1} = T_{eq} + \frac{8\pi \sum_p \int_0^{\omega_m} \int_0^{\frac{\pi}{2}} \alpha_{12}(I_{1,\omega,p}^d)^+ \cos \theta \sin \theta d\omega d\theta}{\sum_p \int_0^{\omega_m} \alpha_{12} \nu_{1g} C_{\omega 1} d\omega}. \quad (C15)$$

Similarly, the temperature of the backward-going phonons on right side of interface is:

$$T_{e2} = T_{eq} - \frac{8\pi \sum_p \int_0^{\omega_m} \int_0^{\frac{\pi}{2}} \alpha_{21}(I_{2,\omega,p}^d)^- \cos \theta \sin \theta d\omega d\theta}{\sum_p \int_0^{\omega_m} \alpha_{21} \nu_{2g} C_{\omega 2} d\omega}. \quad (C16)$$

Due to the equilibrium intensity of phonon, $f_{T_{eq,\omega,p}}^{eq}$, have no contribution to heat flux, the heat flux is derived from:

$$q = 2\pi \sum_p \int_0^{\omega_m} \int_0^{2\pi} \cos \theta \sin \theta I_{\omega,p}^d d\omega d\theta. \quad (C17)$$

Besides, all the integrations of polar angle above is performed through Gauss-Legendre quadrature and that of frequency is performed through rectangular integration. At last, the Kapitza conductance defined by emitted phonon temperature in numerical simulation is given by:

$$G_e = \frac{q}{T_{e1} - T_{e2}}. \quad (C18)$$

To validate the present discrete-ordinates method, cross-plane interfacial phonon transport through single Al/Si interface and Ge/Si interface have been simulated at different temperature with the data given in Appendix B, and compared with the classical analytical solution of Kapitza conductance defined by the emitted phonon temperature, i.e. Eq. (9). Fig. A2 gives the comparison results, which validate the present discrete-ordinates method.

References

- [1] G. Chen, *Nanoscale Energy Transport and Conversion: A Parallel Treatment of Electrons, Molecules, Phonons, and Photons*, Oxford University Press, New York, 2005.
- [2] Y. Guo, M. Wang, Phonon hydrodynamics and its applications in nanoscale heat transport, *Phys. Rep.* 595 (2015) 1–44.
- [3] Z. Yan, G. Liu, J.M. Khan, A.A. Balandin, Graphene quilts for thermal management of high-power GaN transistors, *Nat. Commun.* 3 (2012) 827.
- [4] G.J. Snyder, E.S. Toberer, Complex thermoelectric materials, *Nat. Mater.* 7 (2) (2008) 105–114.
- [5] P.E. Hopkins, Thermal transport across solid interfaces with nanoscale imperfections: effects of roughness, disorder, dislocations, and bonding on thermal boundary conductance, *ISRN Mech. Eng.* 2013 (2013) 682586.
- [6] C.-W. Wu, Y.-R. Wu, Optimization of thermoelectric properties for rough nanoridge GaAs/AlAs superlattice structure, *APL Adv.* 6 (11) (2016) 115201.
- [7] M. Wang, N. Yang, Z.-Y. Guo, Non-fourier heat conduction in nanomaterials, *J. Appl. Phys.* 110 (6) (2011) 064310.
- [8] S. Shin, M. Kaviani, T. Desai, R. Bonner, Roles of atomic restructuring in interfacial phonon transport, *Phys. Rev. B* 82 (8) (2010) 081302.
- [9] Y. Chalopin, K. Esfarjani, A. Henry, S. Volz, G. Chen, Thermal interface conductance in Si/Ge superlattices by equilibrium molecular dynamics, *Phys. Rev. B* 85 (19) (2012) 195302.
- [10] Z. Liang, K. Sasikumar, P. Keblinski, Thermal transport across a substrate-thin-film interface: effects of film thickness and surface roughness, *Phys. Rev. Lett.* 113 (6) (2014) 065901.
- [11] W. Zhang, T.S. Fisher, N. Mingo, Simulation of interfacial phonon transport in Si-Ge heterostructures using an atomistic green's function method, *J. Heat Transfer* 129 (4) (2007) 483–491.
- [12] H. Zhao, J.B. Freund, Phonon scattering at a rough interface between two fcc lattices, *J. Appl. Phys.* 105 (1) (2009) 013515.
- [13] X. Li, R. Yang, Effect of lattice mismatch on phonon transmission and interface thermal conductance across dissimilar material interfaces, *Phys. Rev. B* 86 (5) (2012) 054305.
- [14] G. Chen, Thermal conductivity and ballistic-phonon transport in the cross-plane direction of superlattices, *Phys. Rev. B* 57 (23) (1998) 14958–14973.
- [15] W. Little, The transport of heat between dissimilar solids at low temperatures, *Can. J. Phys.* 37 (3) (1959) 334–349.
- [16] E.T. Swartz, R.O. Pohl, Thermal boundary resistance, *Rev. Mod. Phys.* 61 (3) (1989) 605–668.
- [17] A. Majumdar, Effect of interfacial roughness on phonon radiative heat conduction, *ASME Trans. J. Heat Transfer* 113 (1991) 797–805.
- [18] R.R. Kakodkar, J.P. Feser, Probing the validity of the diffuse mismatch model for phonons using atomistic simulations, *Phys. Rev. B* 95 (12) (2017).
- [19] C. Monachon, L. Weber, C. Dames, Thermal boundary conductance: a materials science perspective, *Annu. Rev. Mater. Res.* 46 (1) (2016) 433–463.
- [20] P.M. Norris, P.E. Hopkins, Examining interfacial diffuse phonon scattering through transient thermoreflectance measurements of thermal boundary conductance, *J. Heat Transfer* 131 (4) (2009) 043207.
- [21] J.Y. Murthy, S.R. Mathur, Computation of sub-micron thermal transport using an unstructured finite volume method, *J. Heat Transfer* 124 (6) (2002) 1176–1181.
- [22] M.-S. Jeng, R. Yang, D. Song, G. Chen, Modeling the thermal conductivity and phonon transport in nanoparticle composites using monte carlo simulation, *J. Heat Transfer* 130 (4) (2008) 042410.
- [23] M.-J. Huang, T.-C. Tsai, L.-C. Liu, M.-S. Jeng, C.-C. Yang, A fast Monte-Carlo solver for phonon transport in nanostructured semiconductors, *Comput. Model. Eng. Sci. (CMES)* 42 (2) (2009) 107–129.
- [24] J.-P.M. Péraud, N.G. Hadjiconstantinou, Efficient simulation of multidimensional phonon transport using energy-based variance-reduced Monte Carlo formulations, *Phys. Rev. B* 84 (20) (2011) 205331.

- [25] H. Ali, B.S. Yilbas, Phonon transport in silicon-diamond thin film pairs: consideration of thermal boundary resistance due to cutoff mismatch and diffusive mismatch models, *Num. Heat Transfer Part A: Appl.* 68 (12) (2015) 1307–1330.
- [26] Y. Guo, M. Wang, Lattice Boltzmann modeling of phonon transport, *J. Comput. Phys.* 315 (2016) 1–15.
- [27] A.J. Minnich, G. Chen, S. Mansoor, B.S. Yilbas, Quasiballistic heat transfer studied using the frequency-dependent Boltzmann transport equation, *Phys. Rev. B* 84 (23) (2011) 235207.
- [28] T. Hori, J. Shiomi, C. Dames, Effective phonon mean free path in polycrystalline nanostructures, *Appl. Phys. Lett.* 106 (17) (2015) 171901.
- [29] C. Hua, A.J. Minnich, Importance of frequency-dependent grain boundary scattering in nanocrystalline silicon and silicon-germanium thermoelectrics, *Semicond. Sci. Technol.* 29 (12) (2014) 124004.
- [30] L. Yang, A.J. Minnich, Thermal transport in nanocrystalline Si and SiGe by ab initio based Monte Carlo simulation, *Sci. Rep.* 7 (2017) 44254.
- [31] C. Hua, X. Chen, N.K. Ravichandran, A.J. Minnich, Experimental metrology to obtain thermal phonon transmission coefficients at solid interfaces, *Phys. Rev. B* 95 (20) (2017) 205423.
- [32] A. Alkudri, S. Pailhès, S. Merabia, Critical angle for interfacial phonon scattering: Results from ab initio lattice dynamics calculations, *Appl. Phys. Lett.* 111 (9) (2017) 093101.
- [33] B. Latour, N. Shulumba, A.J. Minnich, Ab initio study of mode-resolved phonon transmission at Si/Ge interfaces using atomistic Green's functions, *Phys. Rev. B* 96 (10) (2017) 104310.
- [34] J.-P.M. Péraud, C.D. Landon, N.G. Hadjiconstantinou, Monte Carlo methods for solving the Boltzmann transport equation, *Ann. Rev. Heat Transfer* 17 (2014) 205–265.
- [35] G. Bird, Monte Carlo simulation of gas flows, *Annu. Rev. Fluid Mech.* 10 (1) (1978) 11–31.
- [36] N.G. Hadjiconstantinou, Analysis of discretization in the direct simulation Monte Carlo, *Phys. Fluids* 12 (10) (2000) 2634–2638.
- [37] M. Wang, Z. Li, Nonideal gas flow and heat transfer in micro- and nanochannels using the direct simulation Monte Carlo method, *Phys. Rev. E* 68 (4 Pt 2) (2003) 046704.
- [38] M. Wang, Z. Li, Simulations for gas flows in microgeometries using the direct simulation Monte Carlo method, *Int. J. Heat Fluid Flow* 25 (6) (2004) 975–985.
- [39] J.-P.M. Péraud, N.G. Hadjiconstantinou, An alternative approach to efficient simulation of micro/nanoscale phonon transport, *Appl. Phys. Lett.* 101 (15) (2012) 153114.
- [40] J.C. Duda, P.E. Hopkins, J.L. Smoyer, M.L. Bauer, T.S. English, C.B. Saltonstall, P. M. Norris, On the assumption of detailed balance in prediction of diffusive transmission probability during interfacial transport, *Nanoscale Microscale Thermophys. Eng.* 14 (1) (2010) 21–33.
- [41] L. Maurer, S. Mei, I. Knezevic, Phonon Monte Carlo: generating random variates for thermal transport simulation, *arXiv preprint:1602.07735*, 2016.
- [42] Y. Guo, M. Wang, Heat transport in two-dimensional materials by directly solving the phonon Boltzmann equation under Callaway's dual relaxation model, *Phys. Rev. B* 96 (2017) 134312.
- [43] J.-P.M. Péraud, N.G. Hadjiconstantinou, Extending the range of validity of Fourier's law into the kinetic transport regime via asymptotic solution of the phonon Boltzmann transport equation, *Phys. Rev. B* 93 (4) (2016) 045424.
- [44] M. Flik, C. Tien, Size effect on the thermal conductivity of high-Tc thin-film superconductors, *J. Heat Transfer* 112 (4) (1990) 872–881.
- [45] A. Majumdar, Microscale heat conduction in dielectric thin films, *J. Heat Transfer* 115 (1993) 7–16.
- [46] M. Simkin, G. Mahan, Minimum thermal conductivity of superlattices, *Phys. Rev. Lett.* 84 (5) (2000) 927–930.
- [47] L. Liang, B. Li, Size-dependent thermal conductivity of nanoscale semiconducting systems, *Phys. Rev. B* 73 (15) (2006) 153303.
- [48] G. Balasubramanian, I.K. Puri, Heat conduction across a solid-solid interface: understanding nanoscale interfacial effects on thermal resistance, *Appl. Phys. Lett.* 99 (1) (2011) 013116.
- [49] R.E. Jones, J.C. Duda, X.W. Zhou, C.J. Kimmer, P.E. Hopkins, Investigation of size and electronic effects on Kapitza conductance with non-equilibrium molecular dynamics, *Appl. Phys. Lett.* 102 (18) (2013) 183119.
- [50] N. Yang, T. Luo, K. Esfarjani, A. Henry, Z. Tian, J. Shiomi, Y. Chalopin, B. Li, G. Chen, Thermal interface conductance between aluminum and silicon by molecular dynamics simulations, *J. Comput. Theor. Nanosci.* 12 (2) (2015) 168–174.
- [51] S. Simons, On the thermal contact resistance between insulators, *J. Phys. C: Solid State Phys.* 7 (22) (1974) 4048–4052.
- [52] J.C. Duda, T.E. Beechem, J.L. Smoyer, P.M. Norris, P.E. Hopkins, Role of dispersion on phononic thermal boundary conductance, *J. Appl. Phys.* 108 (7) (2010) 073515.
- [53] P.E. Hopkins, L.M. Phinney, J.R. Serrano, T.E. Beechem, Effects of surface roughness and oxide layer on the thermal boundary conductance at aluminum/silicon interfaces, *Phys. Rev. B* 82 (8) (2010) 085307.
- [54] P.E. Hopkins, J.C. Duda, C.W. Petz, J.A. Floro, Controlling thermal conductance through quantum dot roughening at interfaces, *Phys. Rev. B* 84 (3) (2011) 035438.
- [55] J.C. Duda, P.E. Hopkins, Systematically controlling Kapitza conductance via chemical etching, *Appl. Phys. Lett.* 100 (11) (2012) 111602.
- [56] M. Wang, Q. Kang, Electrokinetic transport in microchannels with random roughness, *Anal. Chem.* 81 (8) (2009) 2953–2961.
- [57] P.E. Hopkins, J.R. Serrano, L.M. Phinney, S.P. Kearney, T.W. Grasser, C.T. Harris, Criteria for cross-plane dominated thermal transport in multilayer thin film systems during modulated laser heating, *J. Heat Transfer* 132 (8) (2010) 081302.
- [58] P.E. Hopkins, C.M. Reinke, M.F. Su, R.H. Olsson, E.A. Shaner, Z.C. Leseman, J.R. Serrano, L.M. Phinney, I. El-Kady, Reduction in the thermal conductivity of single crystalline silicon by phononic crystal patterning, *Nano Lett.* 11 (1) (2011) 107–112.
- [59] R. Stedman, G. Nilsson, Dispersion relations for phonons in aluminum at 80 and 300 °K, *Phys. Rev.* 145 (2) (1966) 492–500.
- [60] D. Singh, J.Y. Murthy, T.S. Fisher, Effect of phonon dispersion on thermal conduction across Si/Ge interfaces, *J. Heat Transfer* 133 (12) (2011) 122401.
- [61] Q. Hao, G. Chen, M.-S. Jeng, Frequency-dependent Monte Carlo simulations of phonon transport in two-dimensional porous silicon with aligned pores, *J. Appl. Phys.* 106 (11) (2009) 114321.