

Manipulation of effective thermal conductivity of multilayer thin film by varying thickness ratio of layers using Monte Carlo simulation



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ABSTRACT

Using the kinetic-type Monte Carlo method, we have simulated the effective thermal conductivity of multilayer thin film with spectral diffuse mismatch model for interface treatment. The results show a monotonously increasing relationship between the thermal conductivity and the material thickness closer to isothermal boundary with a varied size ratio whereas the fixed material amount in two four-layer thin films. This is explained by relatively more significant size effect of the layer closer to the isothermal boundary than that of the layer apart from the isothermal boundary. The finding in the present work guides a new way for optimization and design of interface structures at micro- and nano-scale.

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1. Introduction

With the rapid development of micro- and nano-technology, heat transport at micro- and nanoscale has attracted more and more attentions [1–4]. However the mechanism at this scale remains a challenge because of the unavailability of classical heat conduction law, the Fourier's law [5–7]. At the micro- and nano-scale, the interface contributes much on thermal resistance. This contribution makes nanostructures with interfaces a potential new direction for highly efficient thermoelectric materials [8]. However, the further optimization of efficiency meets a bottle neck due to lack of available and clearer knowledge of interfacial phonon transport.

Previous studies show that the thermal boundary resistance varies with the size of system [9–11] and is even different for same material pair at different positions in a same system [12,13], which is attributed to different local non-equilibrium effects. A linear relationship between one period thickness and thermal conductivity is found in superlattice of Debye crystal under gray assumption [14]. The same relationship between one-layer thickness and thermal conductivity is also observed in the four-layer thin film with realistic isotropic dispersions when varying same multiple for each layer thickness [13]. However, it is still unclear for the relationship

when varying layer thickness ratio for constant material amount in the same system.

Currently, there are mainly two classical interface models for phonon transmission coefficient quantifying the transmitting probability when phonons encounter the interface, including acoustic mismatch model (AMM) [15] and diffuse mismatch model (DMM) [16]. DMM and its extended models have been shown to well explain the experiments around ordinary temperature for rough and disordered interface [16–18]. Furthermore, recent measurements and microscopic calculations give the frequency-dependent feature of transmission coefficient [19,20]. The spectral diffuse mismatch model (SDMM) is the best theoretical model accounting for this spectral property at interface to the best knowledge of the authors [21,22].

Monte Carlo method is a capable method to solve Boltzmann transport equation with the clear physical figure and easy treatment for complex interface geometries [11,13,23,24]. A kinetic-type Monte Carlo method (KMC) developed recently especially reduces the fluctuation and performs efficiently in phonon transport simulation at small temperature assumption [23,25]. In the present work, we use KMC to model the thermal conductivity for different layer thickness ratios of a four-layer thin film with constant material amount incorporating SDMM for interface treatment. The content of remainder of this article is organized as below: we will begin our discussion with the numerical and physical models used in the present work in Section 2. Section 3 gives the validations of the present KMC algorithm by modeling cross-plane phonon trans-

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port through single-layer and double-layer thin film. The results and discussions for thermal conductivity of four-layer thin film by varying thickness ratio of layers are shown in Section 4 in details. Finally, the conclusion part is discussed in Section 5.

2. Numerical and physical models

An efficient Monte Carlo method for phonon transport, namely kinetic-type Monte Carlo method (KMC) [23,25], and its computational model adopted in the present work will be introduced in this section. KMC is an improvement for traditional ensemble phonon Monte Carlo method [26–28] under the assumption of small temperature difference [23,25]. It actually solves the linearized version of energy-based deviational phonon Boltzmann equation under this assumption [23,25]:

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

where $e^d = \hbar\omega(f - f_{Teq}^{eq})$ denotes the deviational energy distribution with the reduced Planck constant $\hbar$, phonon angular frequency ω , phonon distribution f and the Bose–Einstein distribution $f_{Teq}^{eq} = 1/[\exp(\hbar\omega/k_B T_{eq}) - 1]$ at the referenced equilibrium temperature T_{eq} with Boltzmann constant k_B ; $\mathbf{v}_g$ is the phonon group velocity and $\tau(\omega, \mathbf{p}, T)$ is the phonon relaxation time with angular frequency ω , polarization $\mathbf{p}$ at the thermodynamic temperature T ; $e_{loc}^{eq} = \hbar\omega f_{loc}^{eq}$ and $e_{Teq}^{eq} = \hbar\omega f_{Teq}^{eq}$ denote the pseudo-equilibrium and equilibrium energy distributions at pseudo-equilibrium temperature T_{loc} and referenced equilibrium temperature T_{eq} respectively.

KMC tracks the energy packets one by one emitted from boundary and initial terms until all packets have been ergodic. This is much different from traditional ensemble phonon Monte Carlo method which tracks many packets synchronously and makes this method less memory consumption and more efficient. The tracked packet will be emitted from boundary or initial terms with the initial phonon properties, such as frequency, polarization, group velocity, sign, initial position, and initial time. Then it undergoes advection, Umklapp scattering and interface scattering until it is absorbed by isothermal boundary or simulation time approaches to the end. Finally the macroscopic information (temperature distribution and heat flux) will be calculated by averaging over these tracked packets at a certain time.

In the present work, the interface scattering is treated by SDMM neglecting inelastic scattering and polarization conversion [21]. Its frequency-dependent phonon transmissivity from material 1 to material 2 is expressed as a function of wave number as [21]:

$$\alpha_{12}(\omega, \mathbf{p}) = \frac{[k_2(\omega, \mathbf{p})]^2}{[k_1(\omega, \mathbf{p})]^2 + [k_2(\omega, \mathbf{p})]^2}. \quad (2)$$

The frequency-dependent phonon transmissivity at the reversed direction can be obtained with the application of the principle of detailed balance as [21]:

$$\alpha_{21}(\omega, \mathbf{p}) = \frac{[k_1(\omega, \mathbf{p})]^2}{[k_1(\omega, \mathbf{p})]^2 + [k_2(\omega, \mathbf{p})]^2}. \quad (3)$$

Both Eq. (2) and Eq. (3) are applied to determine the transmission probability of a phonon encountering the interface. Note that for elastic scattering and no polarization conversion, the transmitted or reflected packet will keep its frequency and polarization while other properties may be changed by following the dispersions of material at the new or previous side [11]. Furthermore, the SDMM requires that phonons scatter diffusely thus the directions

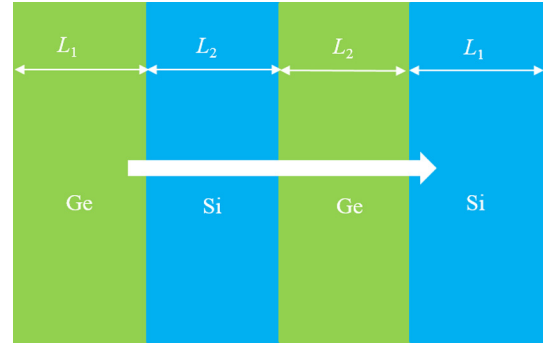


Fig. 1. Schematic of four-layer thin film system formed by material Ge and Si, and L_1 and L_2 denote the thicknesses of the layers. The unidirectional arrow represents the direction of heat flux and the four bidirectional arrows are the ranges for the one layer length.

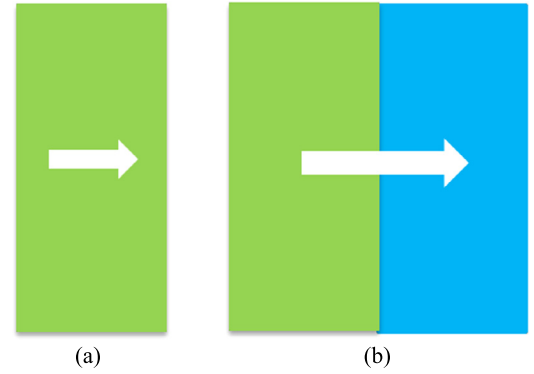


Fig. 2. Schematic of cross-plane phonon transport through the single-layer and double-layer thin film: (a) the single-layer thin film made by Ge and Si separately; (b) the double-layer thin film made by Ge/Si. Both of the unidirectional arrows represent the directions of heat flux.

of group velocity of the transmitted and reflected phonons are reset following the Lambert's cosine law [11,29]. During the tracked phonon travelling within the system, two scattering time will be determined by displacement formula, namely Umklapp scattering time and interface scattering time. If the interface scattering happens first, one random number is generated and compared to the frequency-dependent interfacial phonon transmissivity to determine whether it transmits or reflects [11].

In the present work, phonon transport across a four-layer system formed by material Ge and Si has been modeled to derive the effective thermal conductivity, as is shown in Fig. 1. The middle interface is located at the middle of system and the system is symmetric with respect to it in the present work specially. Therefore, there only exist two thickness values for each four-layer system. L_1 and L_2 are the thicknesses of the layers and will be changed while $(L_1 + L_2)$ is fixed at two values in the present work. The temperatures of left and right side are kept at 301 K and 299 K respectively. Finally we adopt the dispersion relations and relaxation time expressions of Ge and Si from [30] and [31] respectively. And the optical branches are ignored by their negligible contributions to heat transport at steady state.

3. Validations

To validate the present KMC algorithm, we model phonon transport through a single-layer thin film made by Ge and Si separately and double-layer thin film made by Ge/Si with the present KMC algorithm, as are shown in Fig. 2. The benchmark for interfacial phonon transport is not easy mainly by following two reasons: (1) coupling effects of many uncertain factors (such as

interfacial roughness, disorder, dislocations and bonding [32]) in realistic measurements lead to almost impossible direct comparison to measurements; (2) the existed theoretical solution for thermal boundary conductance (also known as Landauer formalism [33]) is obtained from the difference of emitted phonon temperatures at the interface which are difficult to derive from KMC. However, discrete-ordinates method (DOM) is capable of calculating the thermal boundary conductance based on emitted phonon temperatures. Therefore, the DOM scheme with SDMM is developed and validated by calculating Landauer's thermal boundary conductance [11]. Finally, the comparisons between KMC results and DOM results are adopted as the validations for the present KMC algorithm.

The cross-plane phonon transport through the single-layer is modeled firstly by the present KMC algorithm. The system is maintained at 299 K initially and suddenly left and right side are changed to $T_l = 301$ K and $T_r = 299$ K respectively. Subsequently, the temperature of whole system will evolve to a steady state and then both of the temperature and heat flux are calculated by [25]:

$$T = T_{eq} + \sum_i s_i \eta_{eff}^d / C_V V, \quad (4)$$

$$q = \sum_i s_i \eta_{eff}^d v_{i,x} / V, \quad (5)$$

where s_i is the sign of the tracked packet, η_{eff}^d being the effective deviational energy amount of one energy packet, C_V denoting the volumetric heat capacity, V being the volume of unit cell, and $v_{i,x}$ is the group velocity of the tracked packet along the general direction of the heat flux. The effective thermal conductivity is calculated by $\lambda_{eff} = q_{ave} L / (T_l - T_r)$, where L denotes the total thickness of the thin film and q_{ave} is the average value of heat flux along the thin film at steady state. The non-dimensional temperature distributions and effective thermal conductivities obtained by the present KMC are all compared to that calculated by DOM in Fig. 3. To better illustrate the relationship between the effective thermal conductivity and thickness, we introduce an average Knudsen number as [34]:

$$\langle Kn \rangle = \frac{\bar{\Lambda}}{L}, \quad (6)$$

where the average MFP is defined as: $\bar{\Lambda} = \sum_p \int_{\omega} D \frac{de_{eq}}{dT} v_g \tau d\omega / \sum_p \int_{\omega} D \frac{de_{eq}}{dT} d\omega$ with the phonon density of states D . And the comparisons show good agreements between them and validate our KMC algorithm.

The cross-plane phonon transport through double-layer thin film made by Ge/Si is simulated by the present KMC algorithm. The volume ratio of Ge and Si keeps one and the total thickness is varied. Similarly, the temperature and heat flux will be calculated from the present KMC algorithm after the system approaches to the steady state. And the thermal boundary conductance defined by equivalent equilibrium temperature is also obtained by $G = q_{ave} / \Delta T$, where ΔT is the equivalent equilibrium temperature jump at the interface. Both of the non-dimensional temperature distributions and thermal boundary conductance are compared with that by DOM shown in Fig. 4, which show good agreements and thus validate the present algorithm.

4. Results and discussion

After validations, we apply the numerical method to study cross-plane phonon transport through a four-layer thin film formed by Ge/Si shown in Fig. 1. The total thickness of this thin film will

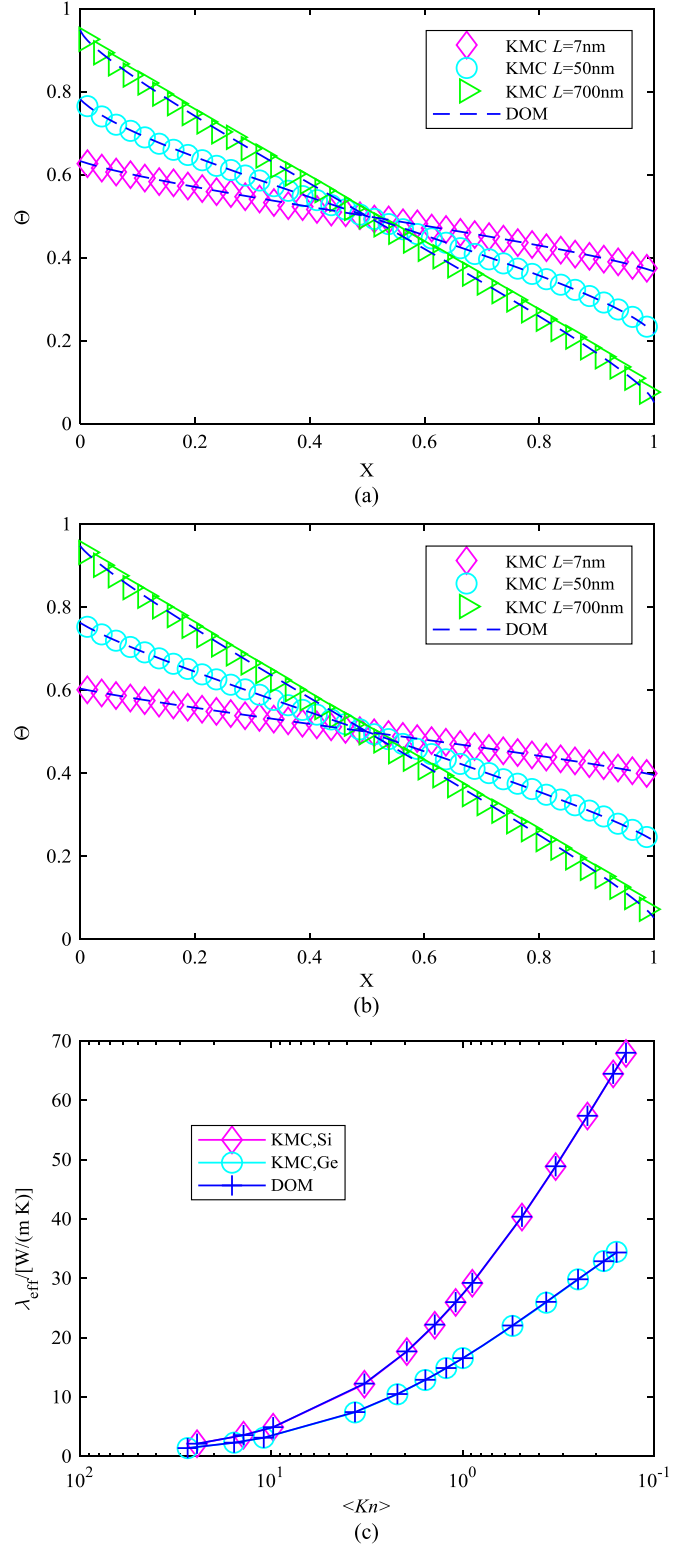


Fig. 3. The non-dimensional temperature distributions and the effective thermal conductivities of cross-plane phonon transport through a single-layer thin film made by Si and Ge: (a) and (b) are the non-dimensional temperature distributions for Si and Ge respectively, including three thicknesses of 7, 50, and 700 nm. Symbols are the results calculated from the present KMC algorithm and dash lines are those calculated from DOM scheme. The non-dimensional temperature Θ in (a) and (b) are defined as the ratios of two temperature differences of temperatures along system and at right boundary relative to that of right boundary. And the definitions for the X in (a) and (b) are all based on system ordinate over the total length of layer. (c) is the effective thermal conductivity for Si and Ge thin films at different average Knudsen numbers, whose horizontal ordinate is defined as the logarithmic length scale.

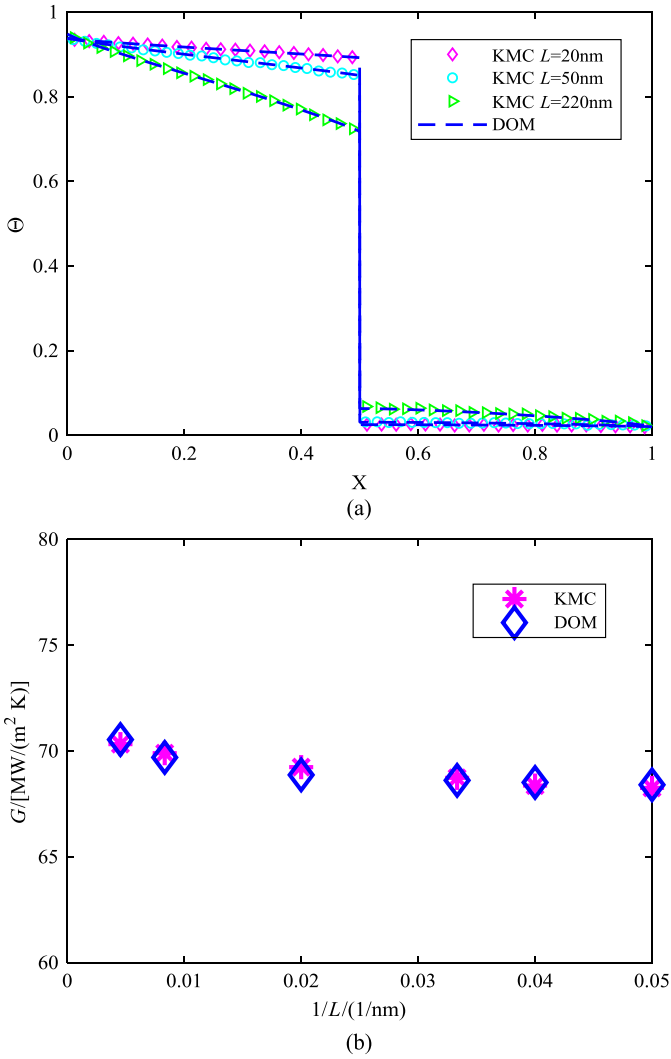


Fig. 4. The non-dimensional temperature distributions and thermal boundary conductance of cross-plane phonon transport through a double-layer thin film made by Ge/Si: (a) the non-dimensional temperature distributions for Ge/Si material pair, including three thicknesses of 20, 50 and 220 nm. Symbols are the results calculated from the present KMC algorithm and dash lines are those calculated from DOM scheme. The non-dimensional temperature Θ is defined as the ratio of two temperature differences of temperatures along system and at right boundary relative to that of right boundary. And the definition for the X is based on system ordinate over the total length of layer. (b) Thermal boundary conductance for Ge/Si material pair at different total thicknesses.

be set as 200 nm and 400 nm. L_1 is varied from 5 nm to 95 nm for the previous case whereas from 10 nm to 190 nm for the latter case then L_2 will be determined. The system will evolve from an initial temperature 299 K with the left and right boundary temperatures kept at $T_l = 301$ K and $T_r = 299$ K respectively and the referenced temperature $T_{eq} = 300$ K. After the system approaches to the steady state, the temperature distribution and heat flux will be calculated. The total evolving time, the time interval and the spatial interval are set as 80 ns, 4 ps and 1 nm for the previous thickness whereas 200 ns, 10 ps and 1 nm for the later thickness. Finally, the total numbers of tracked energy packets are 900 million and 1200 million for the smaller and larger thickness respectively. We average all computational results from the last 100 time intervals to reduce the fluctuations of temperature and heat flux distributions in the KMC simulation.

The effective cross-plane thermal conductivity of this four-layer thin film is derived from the average heat flux at steady state and the exerted temperature difference based on Fourier's law:

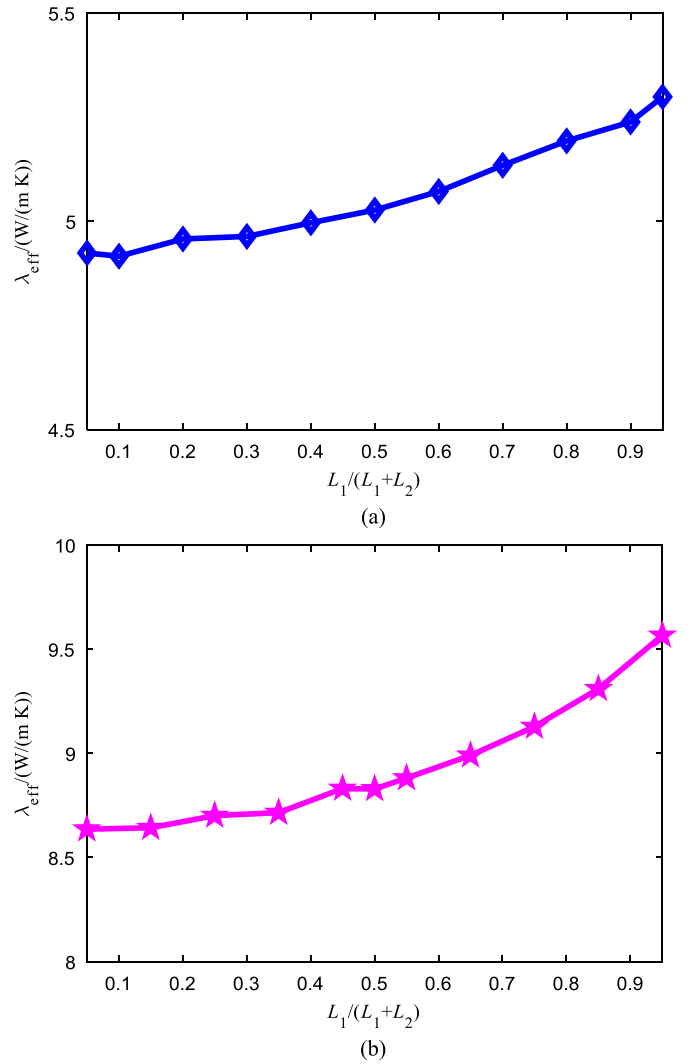


Fig. 5. The effective thermal conductivity at different thickness ratios of layers for the four-layer thin film: (a) total thickness 200 nm; (b) total thickness 400 nm. Since $(L_1 + L_2)$ is fixed at a constant, and L_1 varies from 0 to this constant, which is corresponding to L_2 varies from this constant to 0. Therefore $L_1/(L_1 + L_2)$ varies from 0 to 1 for each case.

$\lambda_{eff} = 2(L_1 + L_2)q_{ave}/(T_l - T_r)$, which is given in Fig. 5. The effective thermal conductivity increases with increasing L_1 or decreasing L_2 whereas constant $(L_1 + L_2)$. The maximum increases are shown about 8% and 11% for the smaller and larger total thickness respectively. This conclusion is not obvious due to the total quantity of material Ge and Si is constant while only the layer ratio in four-layer thin film for each case is changed. It can be explained by the different degrees of size effects of different layers in this four-layer thin film system as below in details.

There are mainly two kinds of layer in the present system whose boundaries are isothermal and interface boundaries or interface and interface boundaries respectively, as shown in Fig. 6. In other words, the layers in (a) and (b) represent the layers contacting with and apart from the isothermal boundary. The interface condition means that phonons encountering with interface will be transmitted or reflected. And the isothermal boundary condition means that phonons encountering with it will be completely absorbed and also phonons will be emitted diffusely from it, which performs just like an interface with one transmissivity and diffuse scattering. Therefore, the interface can cause a bigger resistance than isothermal boundary due to reflection of phonons. The contribution from interface to total resistance is larger than that from

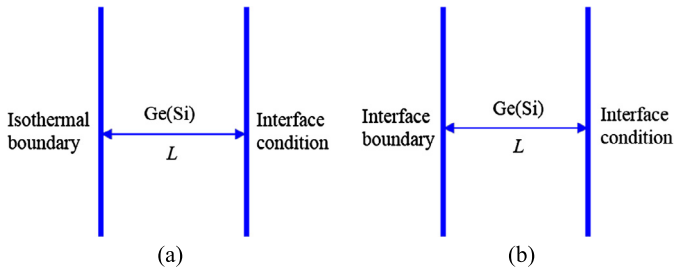


Fig. 6. Schematic of two kinds of layers in this four-layer thin film system: (a) the layer with isothermal and interface boundaries; (b) the layer with interface and interface boundaries.

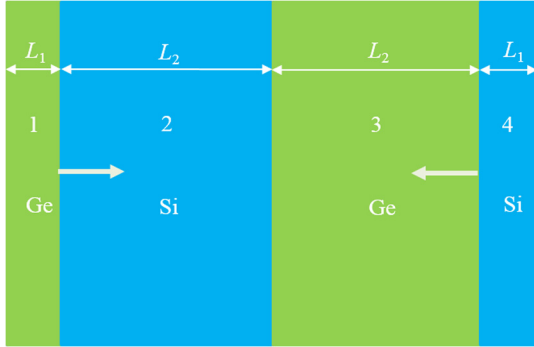


Fig. 7. Schematic of four-layer thin film system formed by material Ge and Si, and L_1 and L_2 denote the thickness of one layer: increasing thickness L_1 and decreasing thickness L_2 whereas keeping total thickness constant. Two unidirectional arrows indicate the moving directions of the interfaces for one total length system. Four bidirectional arrows represent the ranges for one layer length.

isothermal boundary with the same material pair and length L as a result. Then the relative contribution from intrinsic thermal resistance over total resistance of system (a) is larger than that of system (b). The size effect will appear and influence the intrinsic thermal resistance when the thickness of one layer is changed. It is clearer for the system whose contribution from intrinsic thermal resistance is larger. Therefore, the size effect of system (a) is more significant than that of system (b). As is shown in Fig. 7, the thermal conductivities of layer 1 and 4 increase and those of layer 2 and 3 decrease when increasing thickness L_1 whereas maintaining $(L_1 + L_2)$. However, the relative increase of layer 1 and 4 is larger than the relative decrease of layer 2 and 3 due to their more obvious size effects as explained before. Finally, the effective thermal conductivity increases with increasing L_1 or decreasing L_2 even constant $(L_1 + L_2)$ or total material amount. It means that the closer distance to the isothermal boundary, the thicker material amount being suggested to be made. This operation can obtain a multilayer material with as high effective thermal conductivity as possible whereas cost constant material amount. This finding is helpful for design of nanostructures in thermal management.

5. Conclusions

In summary, we have studied the effective thermal conductivity of a Ge/Si four-layer thin film with KMC. The present KMC algo-

rithm is validated firstly in modeling cross-plane phonon transport in single-layer and double-layer thin film. Then we consider a multilayer system of two total thicknesses with varied size ratio of layers whereas fixed material volume. Both numerical results for two total thicknesses give an increasing effective thermal conductivity with increasing thickness of layer next to the isothermal boundary. This increase is attributed to different degrees of size effects of different layers with different boundary conditions. Generally, the size effect of layer with isothermal and interface boundaries is more significant than that of layer with interface and interface boundaries. The present work gives a new way to manage the effective thermal properties of multilayer system and for the optimized design of interface structures.

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