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Phonon boundary scattering effect on thermal conductivity of thin films

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In the study, we introduced the local mean free path of phonons with boundary effects. The local thermal conductivity distribution from boundary to film bulk region was obtained, and the boundary scattering effect was examined by introducing a phonon Knudsen layer thickness. We calculated the ratio of effective thermal conductivity to the bulk one and the results are in agreement with available data. © *2011 American Institute of Physics*. [doi:10.1063/1.3622317]

Thermal properties of thin films play an important role in performance and reliability design of nanometer-sized devices, such as heat energy dissipation, thermal electric conversion, etc.[1–7] The thermal energy in a solid is due to the thermal vibration energy in the lattice. The lattice vibration energy is quantized, and the energy quantum is called a phonon. The state of lattice vibrations may be characterized by the phonon gas consisting of large numbers of phonons moving randomly.[8] Based on the phenomenological phonon diffusion model, the thermal conductivity of bulk silicon, $k = \rho c_v \nu_a \Lambda_b/3$, in which ρ is the density, c_v is the specific heat, ν_a is the average speed of phonons, and Λ_b is the mean free path of phonons. In thin films, when the characteristic length (film thickness, wire diameter) is comparable to the phonon mean free path, the boundary or interface scattering becomes important. Phonon transport is dominated by scattering with the interface of the thin film. Subsequently, the thermal conductivity becomes size dependent.[9,10]

Zhang[11] presented several approaches to consider the classical size effect on thermal conductivity. When the material is infinitely extended, Λ_b is called the bulk mean free path. When the characteristic scale $L \ll \Lambda_b$, the conductivity ratio can be obtained as $k_f/k_b = \Lambda_f/\Lambda_b = 1/Kn$, where Λ_f is the mean free path, and the Knudsen number is defined as $Kn = \Lambda_b/L$. In the intermediate region, one applies Matthiessen's rule to obtain the effective mean free path Λ_{eff} and effective thermal conductivity k_{eff} is

$$\frac{k_{eff}}{k_b} = \frac{\Lambda_{eff}}{\Lambda_b} = \frac{1}{1+Kn}. \tag{1}$$

When $L \ll \Lambda_b$, assuming that all energy carriers originate from the boundary and the free paths should be averaged over the hemisphere, then applying Matthiessen's rule again, one has

$$\frac{k_{eff}}{k_b} = \frac{\Lambda_{eff}}{\Lambda_b} = \left(1+\frac{Kn}{\ln(Kn)+1}\right)^{-1}, \quad Kn > 5. \tag{2}$$

When $Kn < 1$, we may use

$$\frac{k_{eff}}{k_b} = \left(1+\frac{Kn}{m}\right)^{-1}, \tag{3}$$

where $m \approx 3$ for thin films.[9,10] Interpolation has been taken in the intermediate region when Kn is between 1 and 5.

A weighted average of the free path components in the parallel and normal directions of thin films was employed.[10,12] For the z direction (perpendicular to the thin film), using Matthiessen's rule, one can obtain

$$\frac{k_{eff,z}}{k_b} = \left(1+\frac{Kn}{2-Kn^{-1}}\right)^{-1}, \quad Kn > 5. \tag{4}$$

For $Kn < 1$, Eq. (3) should be used with $m = 3$. For circular wires, applying Matthiessen's rule yields

$$\frac{k_{eff,w}}{k_b} = \left(1+\frac{Kn}{1-(4Kn)^{-1}}\right)^{-1}, \tag{5}$$

which can be applied for $Kn > 5$. For $Kn < 1$ Eq. (3) is a good approximation with $m = 4/3$.[11,13]

Based on the Boltzmann transport equation (BTE) for bulk materials, the following expression for thermal conductivities of film was derived:[11,14]

$$\frac{k_{eff}}{k_b} = 1 - \frac{3Kn}{2}\int_1^\infty \left(\frac{1}{t^3}-\frac{1}{t^5}\right)(1-\exp(-t/Kn))dt \tag{6}$$

The formulas mentioned above can effectively be used to study the boundary scattering on thermal conductivity, nevertheless, there are still something to be examined. First, the Matthiessen's rule is applied in the formulas. Though the Matthiessen rule is commonly used to combine phonon scattering mechanisms, its accuracy has never been directly tested.[15] Second, the interpolation in functions between different Knudsen number regions or empirical coefficient used in some existing formulas will cause numerical inaccuracies and inconvenience. Most important, the detailed information of the phonon boundary scattering effect on the bulk material is not yet clear. In this paper, a method is presented to calculate the thermal conductivity reduction in the thin film based on the non-equilibrium gas kinetic theory with boundary scattering effects. The main objective is to illustrate how boundary scattering reduces the thermal conductivity. This theory was first applied to calculate the mean free path of gas molecules by Stops.[16] As phonons collision in the thin film is similar to gas molecules collision in a bounded

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geometry, by this theory we can directly obtain the overall ratio k_{eff}/k_b at a certain Knudsen number without using Matthiessen's rule and interpolation.

For an unbounded phonon, the probability of a phonon that can travel between two consecutive collisions with other phonons at location $\mathbf{x}$ and $\mathbf{x}+d\mathbf{x}$ is of the form:

$$p(\mathbf{x}) = \exp(-\mathbf{x}/\Lambda_b)d(\mathbf{x}/\Lambda_b). \qquad (7)$$

In the film region, when the phonon is bounded, a number of phonons will be terminated by the boundary, which causes the effective mean free path less than Λ_b. The boundary effect on local phonon mean free path for any given geometry can be derived from the above general Eq. (7).

For a thin film, if we consider phonons bounded by two parallel boundaries at $z=0$ and $z=L$, the local mean free path of the phonons at a distance z $(0<z<L)$ from the lower boundary can be calculated by:[16]

$$\frac{\Lambda(z)}{\Lambda_b} = 1 + (\gamma-1)\exp(-\gamma) - \gamma^2\int_\gamma^\infty \frac{\exp(-t)}{t}dt, \qquad (8)$$

where $\gamma = z/\Lambda_b$ for those phonons moving toward $z=0$ and $\gamma=(L-z)/\Lambda_b$ for those moving toward $z=L$. Since a phonon can move toward these two boundaries with the same probability, the local phonon mean free path for all the phonons in the transmit domain can be determined by averaging these two parts.

Fig. 1 presents the local thermal conductivity ratio profiles from one boundary to the other in the thin film for $Kn=0.01$-10. The local thermal conductivity at the boundary is only half of the bulk value when the Knudsen number is small. However, the increasing Knudsen number leads to that the boundary scattering effects originating from the two surfaces start to overlap with each other, and thus the local thermal conductivity at the boundary decreases further. For example, when $Kn=0.3$, the thermal conductivity ratio at the wall is about 0.49 instead of 0.5. In addition, as the Knudsen number increases, the boundary effect extends deeper into the center region of the thin film such that the thermal conductivity in the center zone goes smaller than the bulk value. For example, at $Kn=0.3$, the local thermal conductivity at the center is about 0.91 k_b, rather than k_b.

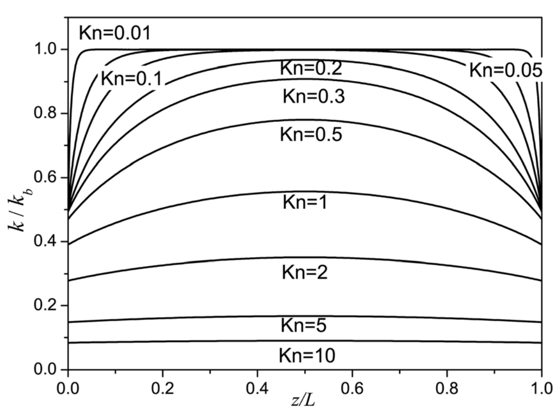

FIG. 1. Local thermal conductivity distribution in a thin film.

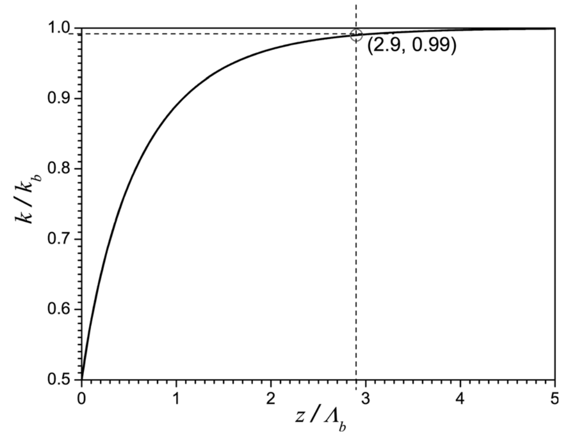

FIG. 2. Local thermal conductivity distribution in a semi-infinite film.

If assuming a semi-infinite film between $z=0$ and $z=\infty$, the local mean free path can be determined as

$$\frac{2\Lambda(z)}{\Lambda_b} = 2 + (\gamma-1)\exp(-\gamma) - \gamma^2\int_\gamma^\infty \frac{\exp(-t)}{t}dt, \qquad (9)$$

where $\gamma = z/\Lambda_b$. Figure 2 shows the local thermal conductivity distribution for the semi-infinite film. The local thermal conductivity is half of the bulk value at the boundary and then increases from the boundary to the bulk as the boundary scattering fades away gradually. Particularly, we can see that the thermal conductivity reaches to 99% of the bulk value at which the distance from the boundary is 2.9 Λ_b. We can define this length as the phonon Knudsen layer thickness, a measure of the distance over which the boundary scattering can exert an effect on the thermal conductivity.

Consider a thin film of thickness dz at a distance z from the lower surface, then, we may assume that the collision rate per unit volume is independent of z, according to Eq. (8) the effective phonon mean free path of these phonons is the average of this free path over the volume. The number of free paths will be ξdz if ξ is the collision rate per unit

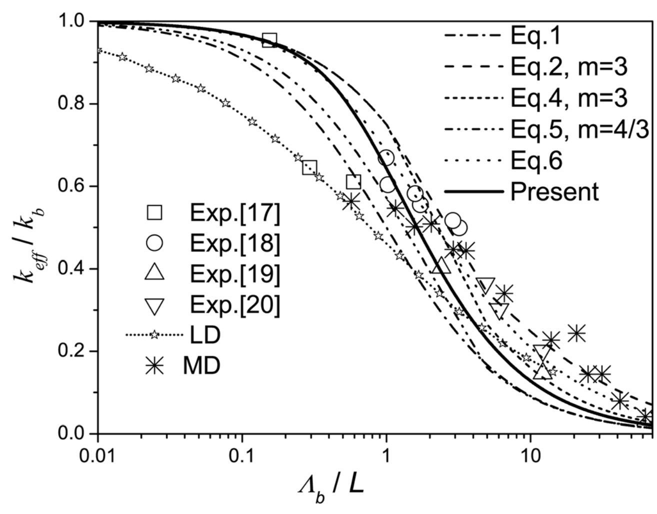

FIG. 3. Comparisons of reduction in effective thermal conductivity between the present model and results from existing formulas, experiments (Refs. 17–20), the lattice dynamics simulation (LD) (Ref. 15), and the molecular dynamics simulation (MD) (Ref. 21).

volume, and the average of this free path over the whole phonon gas volume from $z = 0$ to $z = L$ is

$$\Lambda_1 = \frac{\int_0^L \Lambda_z \xi dz}{\int_0^L \xi dz} = \frac{\Lambda_b^2}{L}\int_0^{1/Kn}\left[1 + (\gamma - 1)\exp(-\gamma) - \gamma^2\int_\gamma^\infty t^{-1}\exp(-t)dt\right]d\gamma. \quad (10)$$

In addition, when phonons collide with the boundary, the reflected phonons will make collisions in the bulk region or even with the other boundary. Therefore, to calculate the overall effective thermal conductivity, one must consider the contribution from those phonons that come from the boundaries. For those phonons that leave the lower boundary at $z = 0$, they can be divided into three groups, those with free paths $r \leq L$ never influenced by the boundary at $z = L$, those with $L < r \leq L/\cos\theta$ unlimited by the boundary, and those with $r > L/\cos\theta$ truncated to $L/\cos\theta$, where θ is the phonon path angle with z-direction. Thus, the mean free path contributions from the phonons leaving from the boundary is

$$\Lambda_2 = \int_0^L p(\mathbf{x})r + \int_L^\infty p(\mathbf{x})\left(\int_{\cos^{-1}L/r}^{\pi/2} r\cos\theta\sin\theta d\theta + \int_0^{\cos^{-1}L/r}(L/\cos\theta)\cos\theta\sin\theta d\theta\right)\Big/\int_0^{\pi/2}\cos\theta\sin\theta d\theta$$
$$= \Lambda_b\left(1 - \exp(-L/\Lambda_b) + \frac{L}{\Lambda_b}\exp(-L/\Lambda_b) - \left(\frac{L}{\Lambda_b}\right)^2\int_{L/\Lambda_b}^\infty t^{-1}\exp(-t)dt\right). \quad (11)$$

By averaging these two contributions of the mean free paths from the boundaries and from the interior, the effective thermal conductivity can be calculated as

$$\frac{k_{eff}}{k_b} = \frac{\Lambda_{eff}}{\Lambda_b} = \frac{\Lambda_1 + \Lambda_2}{2\Lambda_b}. \quad (12)$$

The results calculated from Eq. (12) as a function of the Knudsen number are plotted in Fig. 3, compared with the classical treatments summarized in the Introduction Section. It appears that the trend from the present model is consistent with those formulas and quantitatively intermediates among them. When $Kn > 0.1$ the thermal conductivity reduction increases significantly, and the boundary scattering dominates the thermal conductivity. The present solutions are also compared with available data from experiments,[17–20] the lattice dynamics simulation (LD),[15] and the molecular dynamics simulation (MD).[21] The employed bulk mean free path for silicon at 300 K (Refs. 15 and 17–20) and 400 K (Ref. 21) is 243 nm and 125 nm, respectively, according to Ref. 15. As a whole, the present predictions are in rough agreement with the data from experiments and the MD. When $Kn < 4$ the present solution is higher than the result from LD while it is lower than the LD result when $Kn > 4$.

In summary, by introducing the local phonon mean free path, local thermal conductivity distribution and the overall effective thermal conductivity in the thin film have been obtained analytically. It is found that the boundary scattering extends to the bulk over a distance of 2.9 Λ_b. In addition, the overlap effect due to the multi-boundary scattering will even result in the reduction in local thermal conductivity in the thin film center region. The reduction in effective thermal conductivity from the present model is in reasonable agreement with available data. Though the conclusions of boundary scattering on thermal conductivity are not new, the study provides a simple theoretical method to evaluate the phonon boundary scattering effect particularly provides the quantitative measure of the phonon Knudsen layer thickness.

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