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Thermal Conductivity of a Nano-Structured Material

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ABSTRACT

In this paper, the phonon Boltzmann equation is solved numerically to study the phonon thermal conductivity of nano-structured thin films opened a nano-hole in a host material. We focused on effects of hole size on the reduction of thermal conductivity. The simulation shows that the temperature profiles in nano-structures are very different from those in conventional bulk materials, due to ballistic phonon transport at nanoscale. The conventional heat conduction equations cannot be applied to solve the heat transfer in solids at nanoscale. The effective thermal conductivity of nano-structures are calculated from temperature gradient. We predict the thermal conductivity dependence on the size of a nano-hole. At constant thin film thickness the larger the hole size, the smaller is the thermal conductivity of two-dimensional nano-structured thin film. The results of this study can be used to the development of thermal management of heat conduction by using artificial physical property.

INTRODUCTION

The thermal conductivity of nano-structures are attracting increasing attention due to their importance in different applications such as the development of efficient thermoelectric devices, thermal management of microelectronic devices and circuits, data storage system, and so on. For example, nano-structured materials such as Bi_2Te_3/Sb_2Te_3 superlattices (Venkatasubramanlan et al., 2001) and PbTe/PbSeTe quantum dot superlattices (Harman et al., 2002) have shown significant increases in efficient compared to their bulk materials due to their suppressed phonon thermal conductivity. In the present study the numerical calculations of heat transfer in dielectric thin films and structures with a nano-hole are conducted. Temperature profiles in the nano-structured materials are calculated using the equations of phonon radiative transfer in two dimensions based on the Boltzmann transport theory

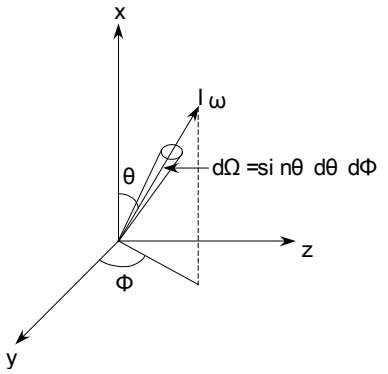


Fig. 1 Schematic diagram of the coordinate system showing the phonon intensity and the various angles

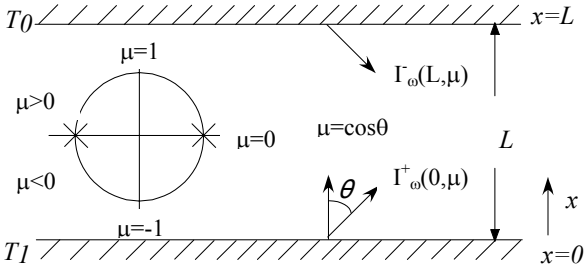


Fig. 2 Schematic diagram of heat transfer in a dielectric film of thickness L at different temperatures

(Yang and Chen, 2004). We also calculate effective thermal conductivities of nano-structures from temperature gradient. We have investigated the effects of the hole size and the phonon Knudsen number, which is defined as the phonon mean free path over the characteristic size the the structure, on the thermal conductivity of the nano-structured materials.

SIMULATION METHODS

The transport of heat in semiconductors and dielectric materials are mainly by the transport of phonons (Majumdar, 1993). The physics of phonons is well documented (Kittel,

1986). In this study phonon transport is analyzed using the Boltzmann transport equation often used to model the transport of particles:

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla f = \left(\frac{\partial f}{\partial t} \right)_s , \tag{1}$$

where f is the distribution function, which depends on time t and particle position $\mathbf{r}$ and velocity $\mathbf{v}$. Phonons that dominate heat transport travel at the speed of sound. For the simplicity the scattering term on the right side of Eq. (1) is approximated by frequency independent relaxation time.

$$\left(\frac{\partial f}{\partial t} \right)_s = \frac{f_0 - f}{\tau} \tag{2}$$

We defined phonon intensity analogous to photon intensity as the energy flow per unit time, per unit area, per solid angle in the direction of phonon propagation, and per unit frequency interval around ω:

$$I(\theta, \phi, x, y, t) = \frac{1}{4\pi} \sum v f \hbar \omega D(\omega) , \tag{3}$$

,where θ is the polar angle, and ϕ the azimuthal angle as shown in Fig.1. By multiplying Eq. (1) by $\mathbf{v}\hbar\omega D(\omega)$ and using the definition of intensity from Eq. (3), the following phonon intensity equation is obtained.

$$\frac{1}{v}\frac{\partial I}{\partial t} + \cos\theta \frac{\partial I}{\partial x} + \sin\theta \cos\phi \frac{\partial I}{\partial y} = \frac{\mathrm{I}_0[T(x,y)] - I}{v\tau} \tag{4}$$

The local equilibrium intensity I^0 corresponds to each temperatures, which in turn depends on the location x, y. We simplify the calculations by invoking a stringent condition of equality at every frequency,

$$I_0[T(x,y)] = \frac{1}{4\pi} \int_0^{2\pi} \int_0^{\pi} I(x, y, \theta, \phi) \sin\theta \, d\theta \, d\phi . \tag{5}$$

Substituting Eq. (5) into Eq.(4) we obtain

$$\frac{1}{v}\frac{\partial I}{\partial t} + \cos\theta \frac{\partial I}{\partial x} + \sin\theta \cos\phi \frac{\partial I}{\partial y} = \frac{\frac{1}{4\pi} \int_0^{2\pi} \int_0^{\pi} I(x, y, \theta, \phi) \sin\theta \, d\theta \, d\phi - I}{v\tau} \tag{6}$$

To numerically solve Eq.(6) it is convenient to separate the intensity I into a forward component I^+ and a backward component I^- depending on the direction of phonon intensity as shown in Figs. 1 and 2. The integral in Eq.(6) is approximated by Gaussian quadrature

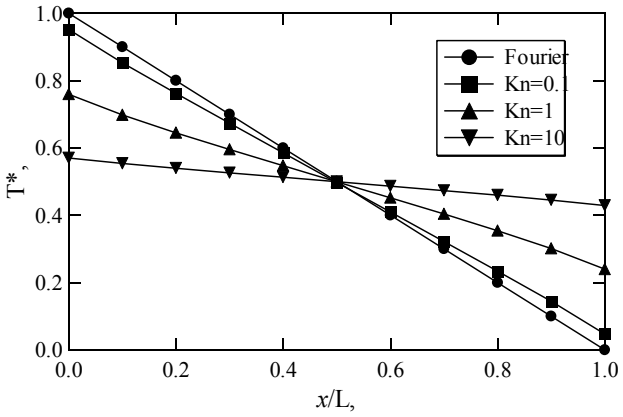
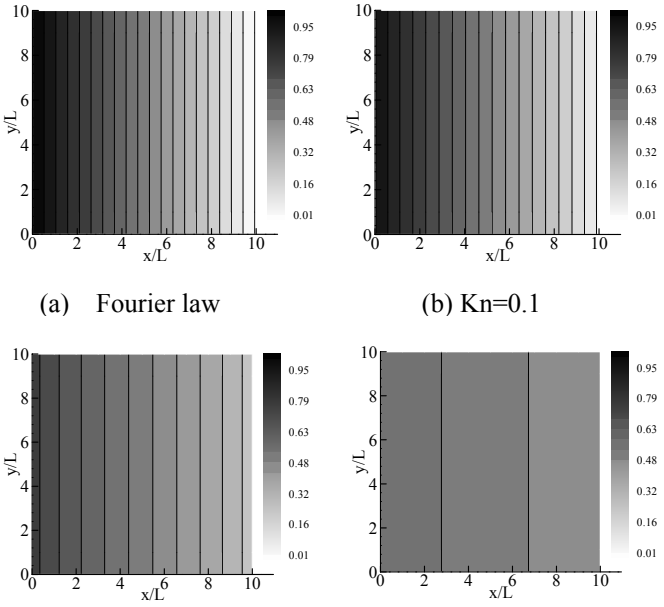


Fig.3 Temperature distribution of thin film in one-dimension



(a) Fourier law (b) Kn=0.1

(c) Kn=1 (d) Kn=10

Fig.4 Non-dimensional temperature distributions calculated by solving two dimensional Bolzmann equations of phonon transport

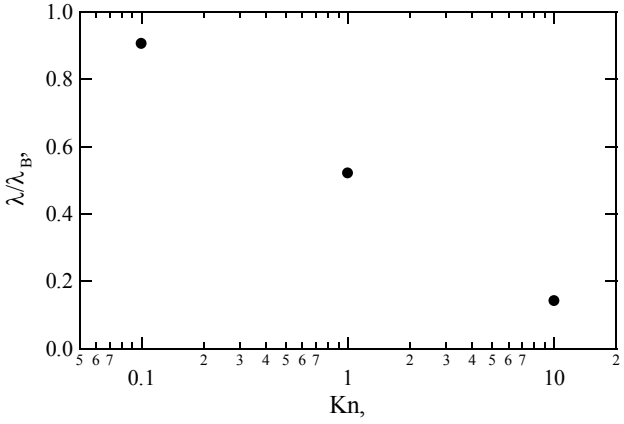


Fig.5 Effective thermal conductivities of thin films

$$\frac{1}{4\pi}\int_0^{2\pi}\int_0^{\pi} I(x,y,\theta,\phi)\sin\theta d\theta d\phi = \frac{2}{4\pi}\sum_m\sum_n I(x,y,\theta,\phi)w_n w_m \quad (7)$$

where w are the weights. Regarding accuracy we chose a quadrature of N=200 for each integral. The boundary conditions in x direction can be written as

$$I^+ = I_0(T_1) \quad \text{at } x/L=1.0 \quad (8)$$

$$I^- = I_0(T_0) \quad \text{at } x/L=0.0 \quad (9)$$

The boundary conditions in y direction are assumed specular scattering of phonons as adiabatic conditions. The following dimensionless parameters have been introduced to the present calculation:

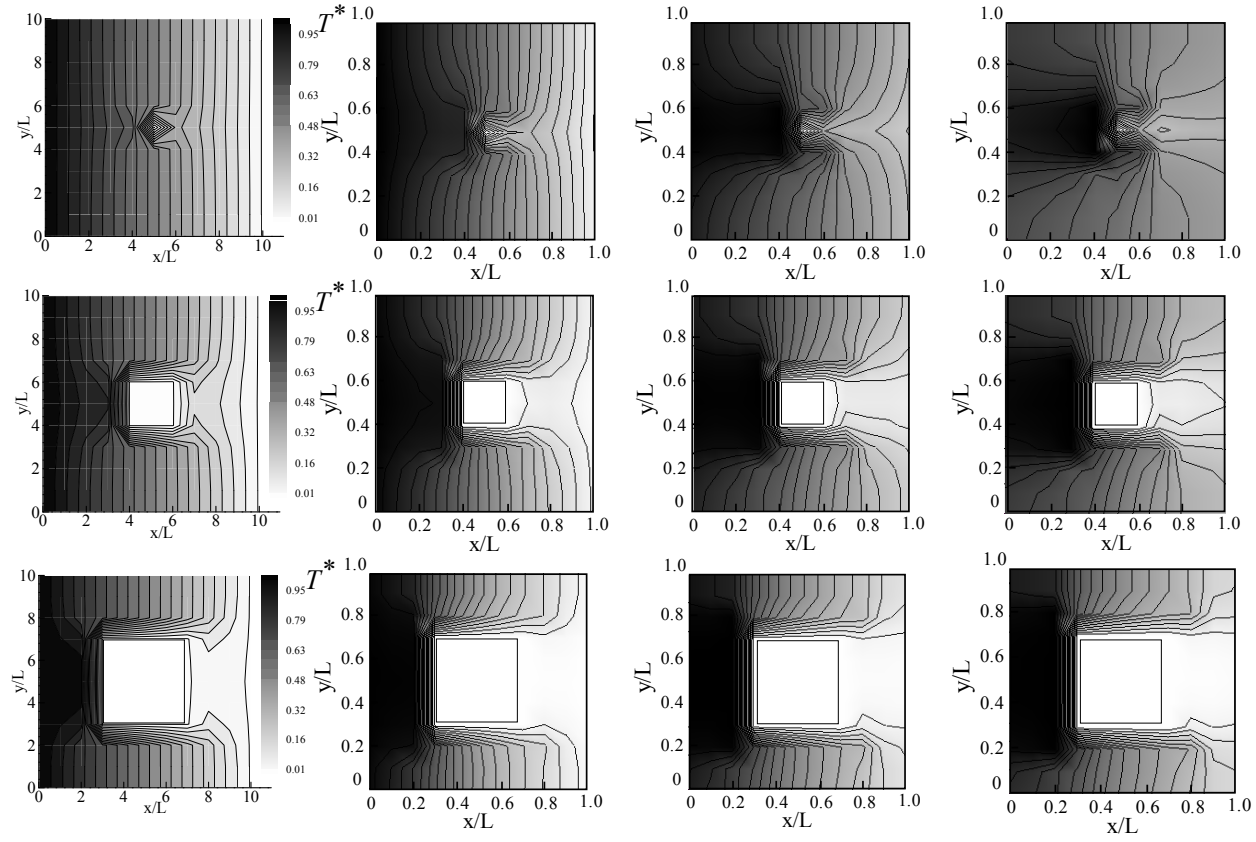
$$x^* = \frac{x}{L}, y^* = \frac{y}{L}, Kn = \frac{\delta}{L}. \quad (10)$$

We defined phonon Knudsen number (Kn) is δ/L where δ is phonon mean free path and L is film thickness. We refer the mean free path of phonon of Si (Dames and Chen, 2004). The value is about 200-300nm at room temperature.

RESULTS AND DISCUSSIONS

Heat Conduction in Solid Dielectric Thin Film

Temperature distributions of thin film without holes are calculated with the specular reflection of phonons at boundaries in y directions in order to check our code for the two-dimensional model. Specular reflection conserves phonon momentum. Therefore constant temperature distributions in y direction are expected in the thin film. Calculated isothermal lines are shown in Fig.4 at each 0.05 non-dimensional temperature. The temperature gradients in Fig. 4 are the same results as those of one-dimensional calculation as shown in Fig.3, which partially validates the solution. When Kn number is large, phonon transport becomes more ballistic, shown by the small slope of the gradient at Kn=10. Therefore temperature jumps at the boundaries becomes more significant for a large Kn number in simulations. We calculated effective thermal conductivities from the gradients of temperature distributions at steady state. The effective thermal conductivity logarithmically decreases with increasing Kn about 10% of the bulk property as shown in Fig.5.



(a) Fourier (b) Kn=0.1 (c) Kn=1 (d) Kn=10

Fig.6 Temperature distribution of thin film with a nano-hole (p=0.3)

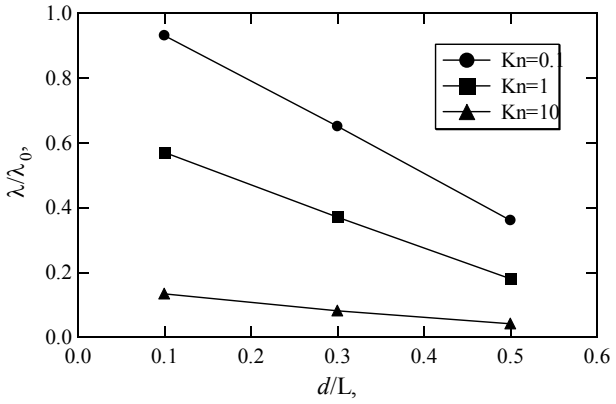


Fig. 7 Effective thermal conductivities of a nano- hole under various Knudsen numbers

Thermal Conductivity of Nano-structured Materials

As we confirmed the validity of our code for a two-dimensional model given by Eq. (6), we studied heat conduction of nano-structured materials. A nano-hole is opened in the calculation domain as a nano-structure. For the boundary condition at the interface of holes, partially diffuse and partially specular scattering interface is used. The interface specularity parameter p is very important for the reduced thermal conductivity value (Chen, 1998) and should be fitted by using experimental results or other analytical results like a molecular dynamics simulations. The interface specularity parameter as an input for BTE simulation can be easily changed to fit available experimental data. In the present study we assumed p=0.3 as an example due to the complexity of the specularity itself. Therefore we focused only on the effects of the size of a hole on the thermal conductivity. The size of a hole is 0.1L, 0.3L, and 0.5L. Temperature distributions of the nano-structure are shown in Fig. 6 by contour plots. When the phonon transport is purely diffusive considered as Fourier law, heat flux calculated from temperature gradient is obtained even behind the hole as shown in Figs. 6 (a). However when the phonon transport becomes ballistic as shown in Figs. 6 (b)(c)(d), heat flux behind the holes is decreased. When the Knudsen number is increased, ballistic phonons are reflected at a nano-hole. As the result thermal transport is much reduced at the high Knudsen number. It means that effective thermal conductivity can be reduced by nano-structures such as a nano-hole. Effective thermal conductivities of nano-structured materials are calculated from temperature gradient at x=0 averaged along y direction (Fig. 7). When the hole size becomes 1/20 of mean path of phonons (Kn=10, d/L=0.5), the effective thermal conductivity is much decreased to less than 5% of the bulk value.

CONCLUSIONS

We solved two-dimensional Boltzmann equations of phonon to calculate heat transfer in dielectric thin films and 2-D structures with a nano-hole. First we calculated temperature distributions in thin films without a hole to confirm the validity of our code for a two-dimensional model. The calculated results agree well with one-dimensional simulation results. Secondly we calculated the thermal conduction of nano-structured materials. The temperature distributions are much different from those of conventional thermal conduction equations due to the ballistic phonon effect at nano-scale. When the size of holes and the Kn number are increased, ballistic phonon effects become dominant. In such case ballistic phonons are reflected at a nano-hole, and heat flux is significantly decreased. When the hole size becomes 1/20 of mean free path of phonon, the effective thermal conductivity is decreased less than 5% of bulk value. In that case hole size corresponds to several nano-meter. The modeling results show that it is possible to suppress the thermal conductivity by using a nano-hole.

NOMENCLATURE

a	acceleration, ms^{-2}
d	hole size, m
C	specific heat per unit volume, $Jm^{-3}K^{-1}$
D(w)	density of states, $m^{-3}s$
F	statistical distribution function
$\hbar$	Planck's constant divided by 2π, Js
I	directional-spectral phonon intensity, $Wm^{-2}sr^{-1}s$
k_B	Boltzmann constant, JK^{-1}
Kn	Knudsen number
L	film thickness, m
p	interface specularity parameter
q	heat flux, Wm^{-2}
t	time, s
T	temperature, K
υ	speed of sound, ms^{-1}
x,y,z	coordinate direction and length, m

Greek symbols

δ	mean free path of phonon, m
ϕ	azimuthal angle, rad
λ	thermal conductivity, $Wm^{-1}K^{-1}$
μ	directional cosine
θ	polar angle, rad
θ_D	Debye temperature, K
τ	relaxation time, s
ω	angular frequency of phonons, s^{-1}
Ω	solid angle, sr

Subscripts

B	bulk
D	Debye cutoff
x	*x*-direction
y	*y*-direction
ω	spectral quantity
0	equilibrium

Superscripts

+	forward direction
–	backward direction
*	non-dimensional

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