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Thermal conductivity model for nanoporous thin films

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ABSTRACT

Nanoporous thin films have attracted great interest because of their extremely low thermal conductivity and potential applications in thin thermal insulators and thermoelectrics. Although there are some numerical and experimental studies about the thermal conductivity of nanoporous thin films, a simplified model is still needed to provide a straightforward prediction. In this paper, by including the phonon scattering lifetimes due to film thickness boundary scattering, nanopore scattering and the frequency-dependent intrinsic phonon-phonon scattering, a fitting-parameter-free model based on the kinetic theory of phonon transport is developed to predict both the in-plane and the cross-plane thermal conductivities of nanoporous thin films. With input parameters such as the lattice constants, thermal conductivity, and the group velocity of acoustic phonons of bulk silicon, our model shows a good agreement with available experimental and numerical results of nanoporous silicon thin films. It illustrates that the size effect of film thickness boundary scattering not only depends on the film thickness but also on the size of nanopores, and a larger nanopore leads to a stronger size effect of the film thickness. Our model also reveals that there are different optimal structures for getting the lowest in-plane and cross-plane thermal conductivities.

1. Introduction

Nanoporous thin films have attracted great interest during the last two decades due to their extremely low thermal conductivity and their potential applications in thermoelectrics [1] and thin thermal insulation [2,3]. Many numerical models such as Monte Carlo (MC) simulations [4–11], Boltzmann transport equation (BTE) methods [12–18], and modified kinetic theory [19–21] have been developed to predict the thermal conductivity of nanoporous materials and thin films. Recently, with nanopore boundary scattering and film thickness considered, the thermal conductivity models including the phonon spectrum for the in-plane and cross-plane directions in a nanoporous thin film were respectively established by Alaie et al. [22] and Hua and Cao [6]. Concerning that previous methods and models usually bring about tedious calculations, in this paper, based on the kinetic theory of phonon transport, we develop a simplified general model to predict the thermal conductivity of two-dimensional nanoporous thin films, with only the lattice constant, thermal conductivity, and the group velocity of acoustic phonons as input. Such kind of simplified analytical models have already been established for the thermal conductivity of nanoporous materials [23] and thin films [24].

The thermal conductivity of a nanoporous thin film depends on the influence factors including porosity, film thickness, size of nanopore, nanopore arrangement and shape [20,21,25–30]. In our model, focusing on the nanoporous thin films with cylindrical nanopores, we applied an effective parameter to include the influence of nanopore size and arrangements. Along the in-plane direction of a nanoporous film, the average phonon-transfer-channel width is adopted as the effective phonon mean free path (MFP) for the nanopore scattering. Considering that the phonon transport along the cross-plane direction of a nanoporous film (which is indeed an inverse structure of nanowires) resembles that in a nanowire [28], the effective hydraulic diameter is applied as the corresponding MFP for the nanopore scattering.

In a thin film, it should be also noted that certain film-boundary roughness can introduce partially diffusive phonon scattering [31–33]. Actually, only when the temperature is below 10 K, the roughness will destroy coherence of short-wavelength phonons [34,35], otherwise, the roughness will not destroy phonons coherence and the surface can be treated as diffusive [36]. In this work, focusing on the classical system (i.e., room temperature), the film boundary is assumed to be diffusive.

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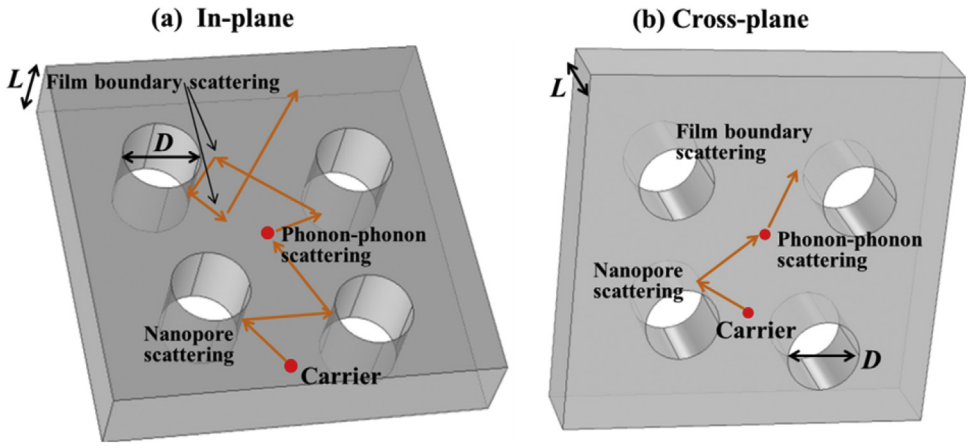

Fig. 1. Schematic showing the scattering processes of an energy carrier in a nanoporous thin film (a) along the in-plane direction and (b) along the cross-plane direction.

By considering simultaneously phonon scatterings by both nanopores and film boundary and frequency-dependent intrinsic phonon-phonon scattering that occurs in the bulk material, an effective thermal conductivity model for nanoporous thin films is established. This general model for nanoporous thin films can be simplified to both nanoporous materials with cylindrical nanopores case and thin film case. The model predictions compare well with available experimental and numerical results of nanoporous silicon thin films. This model can be used to guide the design of nanoporous thin films with reduced thermal conductivity for thermal insulation or thermoelectric use.

2. Analytical model

Fig. 1 shows the geometry along with the scattering processes of an energy carrier in a two-dimensional nanoporous thin film. Here the film thickness is assumed to be L. Cylindrical nanopores with uniform diameter D are dispersed in the film with the pore axis perpendicular to the film thickness direction. Defining n with the unit $1/m^3$ as the volumetric number density of nanopores, the porosity can be then calculated as $\varphi = \frac{\pi}{4}D^2Ln$.

Due to the large structure size considered here, with typical values $L > 10$ nm and $D > 1$ nm, larger than the wavelength of dominant heat carrying phonons (about 1 nm for silicon at room temperature [37,38]), the coherent phonon wave effect is ignored and a phonon particle model is used with bulk phonon properties as input. Based on the kinetic theory (phonon particle model), using Debye approximation where phonon dispersion is assumed to be $\omega = v\kappa$ (κ is wave vector and v is group velocity), the thermal conductivity can be generally written as [24,28,39],

$$\lambda_i(\perp \text{or } \parallel) = \frac{3k_B}{8\pi^3}\int_0^{\omega_D}\int_0^{2\pi}\int_0^{\pi}\frac{v_i^2}{v^3}\tau\omega^2\sin\theta d\theta d\phi d\omega \tag{1}$$

where i denotes the in-plane ($\parallel$) or the cross-plane ($\perp$) direction, k_B is the Boltzmann constant, θ and ϕ are the spherical angles for phonons travel in a 3-dimensional space, v_i is the i component of the group velocity as listed in Table 1, τ is the effective phonon relaxation time (discussed later), ω_D is the Debye frequency described as $\omega_D = v(6\pi^2/\Omega)^{\frac{1}{3}}$ [24], here Ω is the primitive cell volume. For silicon, $\Omega = a^3/4$, where a is the lattice constant of an face-centered cubic lattice.

Table 1
Projected phonon velocities, boundary scattering lifetimes and nanopore scattering lifetimes. Here θ and ϕ are the spherical angles for phonons travel in a 3-dimensional space.

Directions	Projected phonon velocity, v_i [24]	Film boundary scattering lifetime, τ_b [24]	Nanopore scattering lifetime, τ_{pore}
In-plane ($\parallel$)	$v\sin\theta\cos\phi$	$\frac{L}{2v\lvert\cos\theta\rvert}$	$\frac{D}{8\varphi v}$
Cross-plane ($\perp$)	$v\cos\theta$	$\frac{L}{2v\lvert\cos\theta\rvert}$	$\frac{(1-\varphi)D}{2\pi\varphi v}$

For a nanoporous thin film, the scattering processes of an energy carrier include the intrinsic phonon-phonon scattering that regularly happen in bulk materials, the film boundary scattering in the thickness direction, and the scattering with nanopores, as illustrated in Fig. 1. According to the Matthiessen rule [24], the effective phonon relaxation time due to different scattering mechanisms can be expressed as,

$$\frac{1}{\tau} = \frac{1}{\tau_\infty} + \frac{1}{\tau_b} + \frac{1}{\tau_{pore}} \tag{2}$$

where τ_∞, τ_b and τ_{pore} correspond to the intrinsic (bulk) phonon-phonon scattering, film thickness boundary scattering and phonon-nanopore scattering. Here we model the intrinsic phonon-phonon lifetimes using a relationship proposed by Callaway [40], $\tau_\infty = A_0/\omega^2$, where the coefficient A_0 is calculated as $A_0 = 2\pi^2 v\lambda_\infty/k_B\omega_D$ by the bulk limit under Debye approximation, where λ_∞ is the bulk thermal conductivity [24]. The expressions for the film boundary scattering τ_b are obtained from Ref. [24] and listed in Table 1. In the following part, we derive τ_{pore} for both the in-plane direction and the cross-plane direction respectively.

Let us derive first the effective MFP, Λ_{pore}, due to nanopore scattering in the in-plane direction. With the projected area of a cylindrical nanopore on the cross-plane direction defined as $A = DL$, if an energy carrier travels a distance, H, along the in-plane direction, it will be scattered for $A \times H \times n$ times within the swept volume [41]. Thus, the effective MFP can be calculated as,

$$\Lambda_{pore} = \frac{H}{AHn} = \frac{1}{DLn} \tag{3}$$

where Λ_{pore} is thought as the average phonon-transfer-channel width. Substituting the definition of porosity into Eq. (3), the effective MFP due to nanopore scattering is rewritten as,

$$\Lambda_{pore} = \frac{\pi D}{4\varphi} \tag{4}$$

Because the MFP is the average distance traveled ballistically by phonons along the heat-transfer direction [42,43], the distance can therefore be calculated by,

$$\Lambda = \frac{\Lambda_{pore}}{\int_0^{2\pi}\int_0^{\frac{\pi}{2}} cos\theta d\theta d\phi} = \frac{D}{8\varphi} \tag{5}$$

Thus, the corresponding relaxation time due to nanopore scattering