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SIZE-DEPENDENT MODEL FOR THIN FILM THERMAL CONDUCTIVITY

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ABSTRACT

We present a closed-form classical model for the size dependence of thin film thermal conductivity. The model predictions are compared to Stillinger-Weber silicon thin film thermal conductivities (in-plane and cross-plane directions) calculated using phonon properties obtained from lattice dynamics calculations. By including the frequency dependence of the phonon-phonon relaxation times, the model is able to capture the approach to the bulk thermal conductivity better than models based on a single relaxation time.

INTRODUCTION

Thin films are ubiquitous in electronic and optoelectronic applications. By confining materials to sub-micron dimensions, it is possible to control the transport of electrons and phonons. For example, the quantum wells in the active region of a light-emitting diode (LED) allow for the recombination of electrons and holes to produce photons [1, 2]. The closely-spaced interfaces in a semiconductor superlattice scatter phonons while allowing electrons to pass, qualities necessary for a good thermoelectric material [3–5].

The thermal conductivity of a thin film in both the in-plane and cross-plane directions is less than that of the corresponding bulk material [6–9]. The reduced thermal conductivity makes it difficult to remove excess heat (e.g., generated by non-radiative recombination in an LED). This reduction is due to (i) changes in the phonon density of states (sometimes referred to as phonon confinement), and (ii) phonon-boundary scattering. The density of states effect is only expected to be important in very thin films {e.g., thinner than about 20 nm for a Stillinger Weber (SW) silicon film with free surfaces [8] and thinner than about 2 nm for SW silicon thin films bounded by large extents of SW germanium [10]} and is not discussed here. Our focus is on the boundary scattering effect in films with bulk-like density of states.

Modeling work at different levels of sophistication has attempted to predict the thermal conductivity reduction in thin films. Flik and Tien [11] and Majumdar [12] proposed simple models based on a single phonon group velocity and mean free path (i.e., under the gray approximation). Sellan et al. used lattice dynamics calculations to predict phonon properties for the full Brillouin zone of bulk Stillinger-Weber (SW) silicon [9]. From these, they included boundary scattering using the Matthiessen rule and predicted thin film thermal conductivity in the cross-plane direction using a solution to the Boltzmann trans-

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port equation (BTE). They found that their thermal conductivities approached the bulk value at a slower rate than the Majumdar model. Turney et al. [8] found a similar result for the in-plane direction using a boundary scattering model that does not require the Matthiessen rule. While accurate and allowing for consideration of all the phonon modes, the required lattice dynamics calculations are computationally demanding and do not allow for quick calculations for a range of materials.

In a recent paper, Sellan et al. proposed a closed-from classical model for the cross-plane thermal conductivity of a thin film and used it to assess size effects in molecular dynamics thermal conductivity predictions [13]. Here, we compare the predictions of their model, along with an extension to the in-plane direction, to lattice-dynamics based predictions of thin film thermal conductivity. We find that the Sellan model better captures the trends in the lattice dynamics thermal conductivity predictions compared to the Flik and Majumdar models, particularly for the in-plane direction.

MODEL DERIVATION

We first review the Sellan thermal conductivity model [13]. The film has thickness L and is shown in Fig. 1. The in-plane and cross-plane directions correspond to the x and z directions. The derivation begins with an expression for the thermal conductivity in the i direction that is obtained by solving the BTE under the relaxation time approximation and using the Fourier law [14,15]:

$$k_i(L) = \sum_{\nu} \sum_{\boldsymbol{\kappa}} c_{ph} v_{g,i}^2(\boldsymbol{\kappa},\nu)\tau(\boldsymbol{\kappa},\nu,L). \quad (1)$$

The summation is over all phonon modes with wave vector $\boldsymbol{\kappa}$ and dispersion branch ν, the volumetric specific heat of each mode, c_{ph}, is k_B/V for the classical systems considered here, and $v_{g,i}(\boldsymbol{\kappa},\nu)$ is the i component of the group velocity vector, $\mathbf{v}_g(\boldsymbol{\kappa},\nu)$. The phonon transport is described using a set of mode- and system size-dependent relaxation times, $\tau(\boldsymbol{\kappa},\nu,L)$, defined as the average time between successive scattering events. The mode-dependent phonon mean free path, $\Lambda(\boldsymbol{\kappa},\nu,L)$ is defined as $|\mathbf{v}_g(\boldsymbol{\kappa},\nu)|\tau(\boldsymbol{\kappa},\nu,L)$.

We begin by converting the summation over all phonon modes in Eq. (1) to an integral over the first Brillouin zone, resulting in

$$k_i(L) = \frac{V}{8\pi^3} \sum_{\nu} \int c_{ph} v_{g,z}^2 \tau d\boldsymbol{\kappa}. \quad (2)$$

Considering the cross-plane (z) direction, assuming that optical phonons do not contribute to thermal conductivity, and transforming the integral to spherical coordinates gives

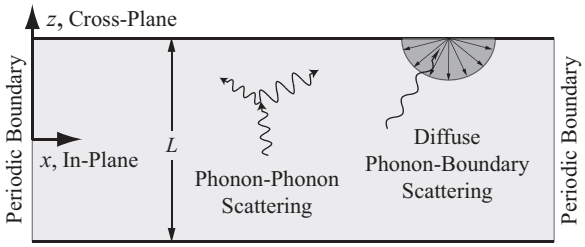

FIGURE 1. THIN FILM MODEL.

$$k_{CP}(L) = \frac{V}{8\pi^3} \sum_{\nu}^{ac} \int_0^{\kappa_{BZ}(\theta,\phi)} \int_0^{2\pi} \int_0^{\pi} (c_{ph}|\mathbf{v}_g \cos\theta|^2 \tau\kappa^2 \sin\theta d\theta d\phi d\kappa), \quad (3)$$

where $\kappa_{BZ}(\theta,\phi)$ is the magnitude of the wave vector at the first BZ boundary along the θ, ϕ direction and the summation is restricted to acoustic modes (ac). Note that multiplying the magnitude of the velocity vector by $\cos\theta$ gives its component in the z direction. These assumptions are reasonable for single-element crystals with face-centered cubic or diamond lattices (e.g., silicon). Optical modes contribute less than 5% to the bulk thermal conductivity of SW silicon [9] and do not exist in crystals with a monoatomic primitive cell.

We now make the Debye approximation for the phonon dispersion, which assumes a single acoustic branch (i.e., no distinction between longitudinal and transverse polarization), such that $\omega = v_{ac}\kappa$. Under this approximation for a classical system, which will allow us to obtain a closed form solution for $k(L)$, Eq. (3) can be simplified to

$$k_{CP}(L) = \frac{3}{8\pi^3} \int_0^{\omega_D} \int_0^{2\pi} \int_0^{\pi} k_B \frac{\cos^2\theta}{v_{ac}} \tau\omega^2 \sin\theta d\theta d\phi d\omega, \quad (4)$$

where the integral over wave-vector has been changed to an integral over frequency and c_{ph} has been replaced with k_B/V. The Debye frequency, ω_D, is defined as [14, 15]

$$\omega_D = v_{ac} \left(\frac{6\pi^2}{\Omega}\right)^{\frac{1}{3}}, \quad (5)$$

where Ω is the volume of the primitive cell and is equal to $a^3/4$ for face centered cubic and diamond crystal lattices, where a is the lattice constant.

The Matthiessen rule is then used to combine the phonon-

phonon and diffuse phonon-boundary scattering, so that

$$\frac{1}{\tau} = \frac{1}{\tau_\infty} + \frac{2v_{\mathrm{ac}}|\cos\theta|}{L}. \tag{6}$$

The phonon-phonon relaxation times are modeled using a relationship derived by Callaway for low frequencies, $\tau_\infty = A/\omega^2$. This form is in agreement with the lattice dynamics predicted phonon-phonon relaxation times for SW silicon at frequencies below 3 THz [13, 16]. The constant A is calculated in the bulk limit and is

$$A = \frac{2\pi^2 v_{\mathrm{ac}} k_\infty}{k_{\mathrm{B}}\,\omega_{\mathrm{D}}}, \tag{7}$$

where k_∞ is the bulk thermal conductivity. Substituting Eqs. (5), (6), and (7) into Eq. (4) and integrating over θ, ϕ, and ω yields

$$\frac{k_{CP}(\mathscr{L})}{k_\infty} = \frac{6}{7} + \frac{3}{14}\mathscr{L} - \frac{3}{7}\mathscr{L}^2 + \frac{3}{7}\mathscr{L}^3 \ln\left(1+\frac{1}{\mathscr{L}}\right) - \frac{6}{7\sqrt{\mathscr{L}}}\arctan\left(\sqrt{\mathscr{L}}\right), \tag{8}$$

where $\mathscr{L}$ is a non-dimensional length defined as

$$\mathscr{L} \equiv \frac{6k_{\mathrm{B}}v_{ac}L}{k_\infty a^3}. \tag{9}$$

By using the kinetic theory expression for the bulk thermal conductivity,

$$k_\infty = \frac{1}{3}Cv_{ac}^2\tau_g = \frac{1}{3}Cv_{ac}\Lambda_g, \tag{10}$$

where C is the volumetric specific heat and τ_g and Λ_g correspond to the relaxation time and mean free path under the gray approximation (i.e., all phonons have the same properties), $\mathscr{L}$ can be expressed as

$$\mathscr{L} = \frac{3}{2}\frac{1}{n_{pc}}\frac{L}{\Lambda_g}. \tag{11}$$

Here, n_{pc} is the number of atoms in the primitive cell (two for silicon) and the ratio L/Λ_g is that used in the Flik and Majumdar models.

For the in-plane direction, Eq. (3) can be recast as

$$k_{IP}(L) = \frac{V}{8\pi^3}\sum_{\nu}^{ac}\int_0^{\kappa_{\mathrm{BZ}}(\theta,\phi)}\int_0^{2\pi}\int_0^{\pi}(c_{ph}|\mathbf{v}_g\sin\theta\cos\phi|^2 \tau\kappa^2\sin\theta d\theta d\phi d\kappa), \tag{12}$$

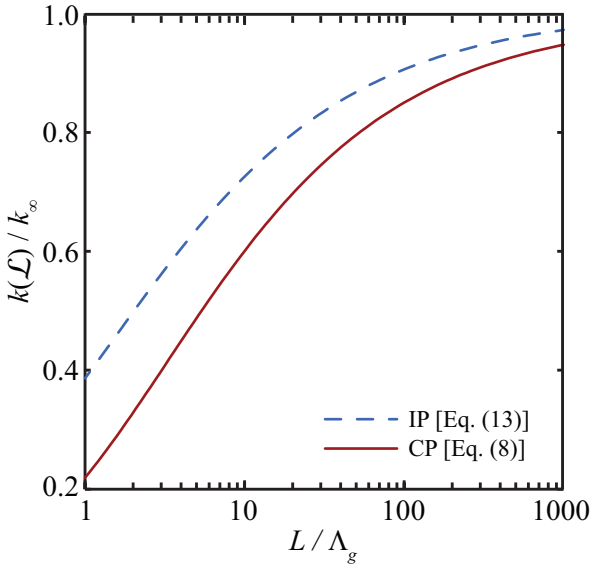

FIGURE 2. IN-PLANE AND CROSS-PLANE THERMAL CONDUCTIVITY MODELS.

and the length-dependent thermal conductivity is

$$\frac{k_{IP}(\mathscr{L})}{k_\infty} = \frac{4}{7} - \frac{3}{28}\mathscr{L} + \frac{3}{14}\mathscr{L}^2 + \frac{1}{2}\mathscr{L}\ln\left(1+\frac{1}{\mathscr{L}}\right) - \frac{3}{14}\mathscr{L}^3\ln\left(1+\frac{1}{\mathscr{L}}\right) - \frac{4}{7\sqrt{\mathscr{L}}}\arctan\left(\sqrt{\mathscr{L}}\right). \tag{13}$$

As shown in Fig. 2, $k(\mathscr{L})$ approaches k_∞ as $\mathscr{L}\to\infty$. The approach to bulk is slower than what would be seen in an experimental sample as the model does not include phonon scattering by defects (e.g., isotopes, vacancies, grain boundaries). The in-plane thermal conductivity is always higher than the cross-plane value. As discussed by Sellan et al. [9], this result can be understood by considering the frequency dependence of the thermal conductivity and how low-frequency phonons, which dominate the bulk thermal conductivity, scatter differently when traveling in the in-plane and cross-plane directions.

MODEL ASSESSMENT

Comparison to Predictions from Lattice Dynamics Calculations

Without making any assumptions, Eq. (1) can be evaluated directly using phonon properties obtained from lattice dynamics calculations [17–20]. For the cross-plane direction, Sellan et al. performed such calculations for SW silicon thin films at a temperature of 300 K [9]. They included diffuse boundary scattering using the Matthiessen rule, Eq. (6). Here, their bulk phonon

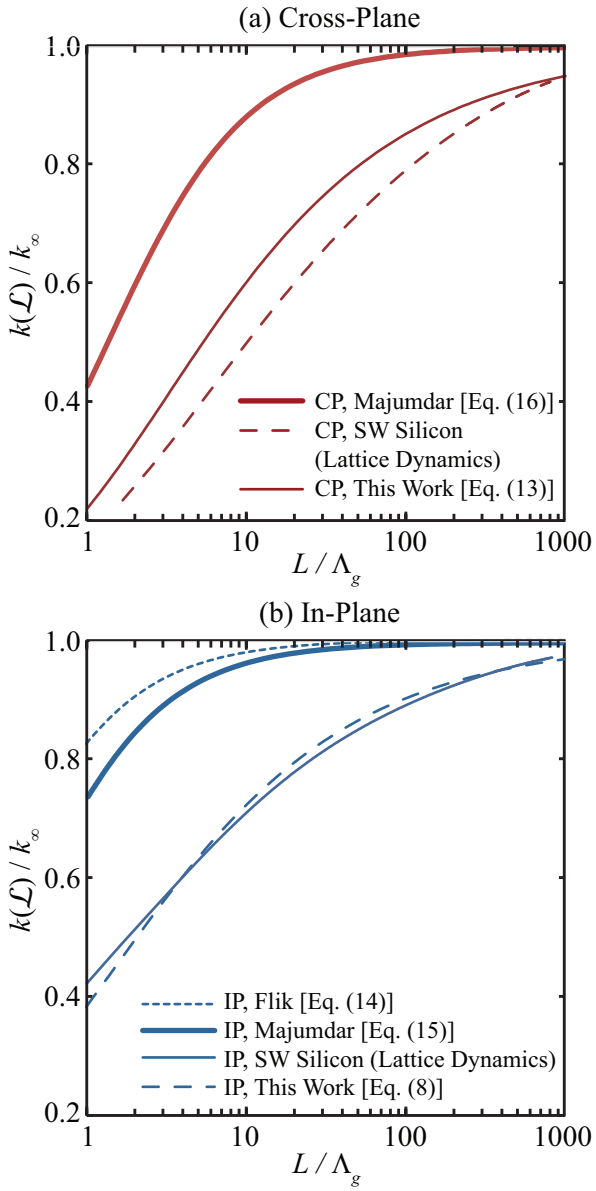

FIGURE 3. COMPARISON OF THERMAL CONDUCTIVITY MODELS AND LATTICE DYNAMICS/BTE PREDICTIONS: (a) CROSS-PLANE, (b) IN-PLANE.

properties data are used to also predict the length dependence of the in-plane thermal conductivity. The lattice dynamics calculations are carried out in the classical limit to be consistent with the derivations of Eqs. (8) and (13).

The lattice dynamics-predicted thin film thermal conductivities are plotted in Figs. 3(a) (cross-plane) and 3(b) (in-plane). The phonon mean free path in the gray approximation, Λ_g, used to scale the thin film thickness, is calculated using Eq. (10). The velocity is taken as an average of the one longitudinal and two transverse acoustic phonon branches in the low-frequency limit in the [001] direction and is 5973 m/s [13]. The specific heat in the classical limit is 2.07×10^6 J/m^3-K. Using the lattice dynamics predicted bulk thermal conductivity of 495 W/m-K gives an average mean free path of 120 nm. The bulk thermal conductivity is higher than that measured experimentally due to (i) the use of the Stillinger-Weber potential, which is known to overpredict thermal conductivity [13, 21] and (ii) the lack of defects in the calculation model.

The lattice dynamics-based in-plane thermal conductivities are in excellent agreement with Eq. (13). The cross-plane predictions are not as good. The difference can be understood by considering the frequency dependence of thermal conductivity. In bulk SW silicon, thermal conductivity is dominated by low frequency phonons [9], which follow the $\tau_\infty = A/\omega^2$ dependence used in the model formulation. Low frequency phonons in the thin film traveling in the in-plane direction are not strongly scattered by the boundaries, and dominate thermal transport, as in bulk. Thus, the model and lattice dynamics predictions are in good agreement. Low frequency phonons traveling in the cross-plane direction, however, are strongly scattered by the boundaries and their mean free path is effectively reduced to the film thickness. Because of this, phonons with larger frequencies, which do not obey the $\tau_\infty = A/\omega^2$ dependence, contribute more to the thermal conductivity. The model predictions are thus not as good for the cross-plane direction, particularly for thinner films.

Comparison to Gray Models

The Flik [11] and Majumdar [12] thermal conductivity models are both derived under the gray approximation for diffuse boundary scattering and $L/\Lambda \gg 1$. The Flik model for the in-plane direction is

$$\frac{k_{IP,F}}{k_\infty} = \frac{1}{1+\frac{2\Lambda_g}{3\pi L}}. \tag{14}$$

The Majumdar model for the in-plane direction is

$$\frac{k_{IP,M}}{k_\infty} = \frac{1}{1+\frac{3\Lambda_g}{8L}} \tag{15}$$

and for the cross-plane direction is

$$\frac{k_{CP,M}}{k_\infty} = \frac{1}{1+\frac{4\Lambda_g}{3L}}. \tag{16}$$

Eqs. (14), (15), and (16) are plotted in Figs. 3(a) and 3(b). For both directions, these gray models predict a faster approach

to the bulk thermal conductivity than the lattice dynamics data and the models derived in this paper. From this result, the importance of considering some spectral dependence of the mean free path (relaxation time) is clearly seen.

SUMMARY

We derived a closed-form classical model for the size dependence of thin film thermal conductivity in the in-plane and cross-plane directions. The model assumes an isotropic, Debye medium, considers a simple-frequency dependence for the phonon relaxation times, uses the Matthiessen rule to include diffuse boundary scattering, and ignores the contribution of optical phonons. We tested the model by comparing against predictions from lattice dynamics calculations on SW silicon thin films where the frequency dependence of all phonon properties (group velocities and relaxation times) are included. The model does an excellent job of predicting the length dependence for the in-plane direction, where low frequency phonons dominate thermal transport. The agreement is not as good for the cross-plane direction, where phonons across a wider range of frequencies make important contributions to the thermal conductivity. When compared to gray models, our model better capture the approach of the thin film thermal conductivity to the bulk value.

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