Size-dependent model for thin film and nanowire thermal conductivity

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We present an analytical model for the size-dependence of thin film and nanowire thermal conductivity and compare the predictions to experimental measurements on silicon nanostructures. The model contains no fitting parameters and only requires the bulk lattice constant, bulk thermal conductivity, and an acoustic phonon speed as inputs. By including the mode-dependence of the phonon lifetimes resulting from phonon-phonon and phonon-boundary scattering, the model captures the approach to the bulk thermal conductivity of the experimental data better than gray models based on a single lifetime. © *2011 American Institute of Physics*. [doi:10.1063/1.3644163]

The thermal conductivities of films and wires with micron and nanometer dimensions have been measured and predicted to be less than that of the corresponding bulk material.[1–20] The reduced thermal conductivity makes it difficult to remove excess heat (e.g., generated in electronics) but can benefit thermoelectric energy conversion. This reduction is due to changes in the phonon density of states[4,6,8,13,17] and phonon-boundary scattering. The density of states effect is only important in very small nanostructures [e.g., suspended Stillinger-Weber (SW) silicon films thinner than 20 nm (Ref. 17)] and is not discussed here. Our focus is on the boundary scattering effect in nanostructures with bulk-like density of states.

Modeling work at different levels of sophistication has attempted to predict the thermal conductivity reduction in films and wires. Flik and Tien[1] and Majumdar[2] proposed film models based on a single phonon group velocity and lifetime (i.e., the gray approximation). More detailed calculations that include quantum statistics and the mode dependence of the phonon properties have proven successful at predicting experimental data for silicon nanostructures with minimal fitting parameters.[8,10,16] Lattice dynamics calculations[17,18] and Monte Carlo-based methods[11] have also been used to elucidate the underlying phonon physics.

We recently proposed a closed-from high-temperature model for the cross-plane thermal conductivity of a film and used it to assess size effects in molecular dynamics thermal conductivity predictions.[21] Here, we extend this model, which contains no fitting parameters, to the in-plane direction and to wires. The predictions are then compared to experimental data for silicon nanostructures. Our model does not seek to supplant more detailed calculations but to provide a straightforward method for predicting thermal conductivity reduction in nanostructures.

The film and wire geometries are shown in Figs. 1(a) and 1(b). The film has thickness L, and the wire has diameter D. The derivation begins with an expression for the thermal conductivity in the i direction that is obtained by solving the Boltzmann transport equation under the relaxation time approximation and using the Fourier law[22]

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

The summation is over all phonon modes with wave vector $\boldsymbol{\kappa}$ and dispersion branch ν, c_{ph} is the mode volumetric specific heat, and $v_{g,i}(\boldsymbol{\kappa}, \nu)$ is the i component of the group velocity vector. The phonon transport is described using a set of mode- and system-size-dependent lifetimes, $\tau(\boldsymbol{\kappa}, \nu, L \text{ or } D)$, defined as the average time between successive scattering events.

Converting the summation over wave vector in Eq. (1) to an integral in spherical coordinates over the first Brillouin zone (BZ) and assuming that optical phonons do not contribute to thermal conductivity gives

$$k_i(L \text{ or } D) = \frac{V}{8\pi^3} \sum_{\nu}^{ac} \int_0^{\kappa_{BZ}(\theta,\phi)} \int_0^{2\pi} \int_0^{\pi} c_{ph} v_{g,i}^2 \tau \kappa^2 \sin\theta \, d\theta d\phi d\kappa, \quad (2)$$

where V is the sample volume, $\kappa_{BZ}(\phi, \phi)$ 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). While optical phonons can contribute to thermal conductivity in nanostructures,[18,20] we ignore them here to preserve the simplicity of the model.

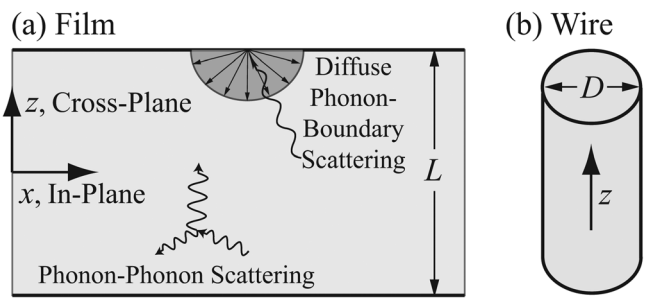

FIG. 1. (a) Film and (b) wire models. The film is infinite in the in-plane directions, and the wire is infinite along its axis.

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TABLE I. Group velocities and diffuse boundary scattering lifetimes. For the film, the average distance traveled ballistically by a phonon before hitting the top or bottom surface is $1/(L|\cos\theta|)\int_0^L z dz = L/(2|\cos\theta|)$. For the wire (with $D=2R$), the average distance traveled ballistically by a phonon before hitting the side is $1/(\pi R^2|\sin\theta|)\int_0^{2\pi}\int_0^R (\sqrt{R^2-r^2\cos^2\phi}-r\sin\phi) r dr d\phi = 4D/(3\pi|\sin\theta|)$. θ and ϕ are the spherical angles.

Geometry	Group velocity, $v_{g,i}$	Boundary scattering lifetime, τ_b
Film, in-plane	$v_{ac}\sin\theta\cos\phi$	$\frac{L}{2v_{ac}\lvert\cos\theta\rvert}$
Film, cross-plane	$v_{ac}\cos\theta$	$\frac{L}{2v_{ac}\lvert\cos\theta\rvert}$
Wire	$v_{ac}\cos\theta$	$\frac{4D}{3\pi v_{ac}\lvert\sin\theta\rvert}$

We now make the Debye approximation for the phonon dispersion, which assumes a single acoustic branch such that $\omega = v_{ac}\kappa$. Under this approximation, for a classical (i.e., high temperature) system, Eq. (2) simplifies to

$$k_i(L \text{ or } D) = \frac{3}{8\pi^3}\int_0^{\omega_D}\int_0^{2\pi}\int_0^{\pi} k_B \frac{v_{g,i}^2}{v_{ac}^3}\tau\omega^2\sin\theta d\theta d\phi d\omega, \quad (3)$$

where the integral over wave vector has been changed to an integral over frequency and c_{ph} has been replaced with k_B/V, where k_B is the Boltzmann constant. The Debye frequency, ω_D, is $v_{ac}(6\pi^2/\Omega)^{\frac{1}{3}}$,[22] where Ω is the primitive cell volume. For silicon, which has a diamond structure, the primitive cell volume is $a^3/4$, where a is the lattice constant. The expressions for $v_{g,i}$ are provided in Table I.

The Matthiessen rule is used to combine the effects of phonon-phonon (τ_∞) and phonon-boundary (τ_b) scattering so that[22]

$$\frac{1}{\tau} = \frac{1}{\tau_\infty} + \frac{1}{\tau_b}. \quad (4)$$

The phonon-phonon lifetimes are modeled using a relationship proposed by Callaway[23] for low frequencies, $\tau_\infty = A/\omega^2$. This form agrees with lattice dynamics predicted phonon-phonon lifetimes times for SW silicon at a temperature of 300 K for frequencies below 3 THz.[21] The constant A is calculated in the bulk limit and is $2\pi^2 v_{ac} k_\infty / k_B \omega_D$, where k_∞ is the bulk thermal conductivity. Phonon-boundary scattering at high temperatures is expected to be diffuse.[24] The expressions for τ_b are provided in Table I.

Following substitution of Eq. (4) into Eq. (3) and using the expressions from Table I, the integrals can be performed analytically for the film, leading to

$$\frac{k_{In-Plane}(\mathcal{L})}{k_\infty} = \frac{4}{7} - \frac{3}{28}\mathcal{L} + \frac{3}{14}\mathcal{L}^2 + \frac{1}{2}\mathcal{L}\ln\left(1+\frac{1}{\mathcal{L}}\right) - \frac{3}{14}\mathcal{L}^3\ln\left(1+\frac{1}{\mathcal{L}}\right) - \frac{4}{7\sqrt{\mathcal{L}}}\arctan(\sqrt{\mathcal{L}}), \quad (5)$$

$$\frac{k_{Cross-Plane}(\mathcal{L})}{k_\infty} = \frac{6}{7} + \frac{3}{14}\mathcal{L} - \frac{3}{7}\mathcal{L}^2 + \frac{3}{7}\mathcal{L}^3\ln\left(1+\frac{1}{\mathcal{L}}\right) - \frac{6}{7\sqrt{\mathcal{L}}}\arctan(\sqrt{\mathcal{L}}). \quad (6)$$

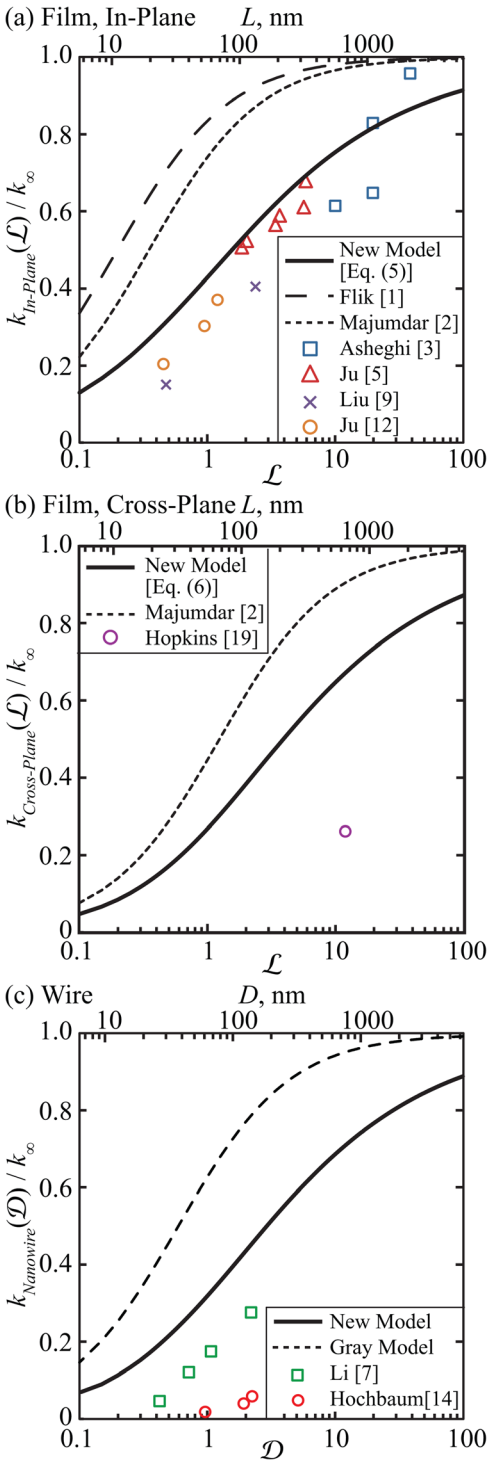

FIG. 2. (Color online) Comparison of thermal conductivity models and experimental data for silicon nanostructures at a temperature of 300 K: (a) film in-plane, (b) film cross-plane, (c) wire.

Here, $\mathcal{L}$ is a non-dimensional length defined as $\frac{3}{2}\frac{k_B v_{ac} L}{k_\infty \Omega}$. For the wire, the angular integrals can be carried out analytically, but the frequency integral must then be performed numerically. While there is no closed-form solution for the thermal conductivity, the ratio $k_{Nanowire}(\mathcal{D})/k_\infty$ can be plotted as a function of $\mathcal{D} = \frac{4}{\pi}\frac{k_B v_{ac} D}{k_\infty \Omega}$.

As shown by the solid lines in Figs. 2(a)–2(c), the thermal conductivity models approach k_∞ as the limiting dimension (L or D) increases. The in-plane thermal conductivity of

the film is always higher than the cross-plane value. This result can be understood for a cubically isotropic material by noting that phonons scatter more frequently with boundaries when traveling in the cross-plane direction.[17,18]

Available experimental thermal conductivity data for silicon films[3,5,9,12,19] and wires[7,14] at a temperature of 300 K are plotted in Figs. 2(a)–2(c). Because the thermal conductivity of silicon is dominated by low-frequency phonons,[17,18] our high-temperature models should give reasonable predictions even though the Debye temperature of silicon is 645 K.[25] In converting the experimental data to dimensionless units, the lattice constant is 5.43 Å, the velocity is the average of the one longitudinal and two transverse acoustic phonon branches in the low-frequency limit in the [001] direction and is 6733 m/s, and the bulk thermal conductivity is 149 W/m-K.[25]

The in-plane thermal conductivity model captures the trend measured by four separate studies. The model predictions are generally higher than the experimental data. The result may be due to the flatness of the transverse acoustic phonon branch in silicon at mid- to high-frequencies,[25] which a single phonon velocity cannot capture. Making the boundary scattering in the model somewhat specular increases the predicted thermal conductivity away from the experimental data. While the agreement of the cross-plane thermal conductivity model with the one available experimental measurement is poor, more data are needed to assess its performance.

For the wire, the model overpredicts the experimental data of Li *et al.*[7] by $\sim 0.1 k/k_\infty$. The similar slopes of the experimental data and the model suggest that the boundary scattering is properly handled. The wire model may suffer due to the single group velocity[8] and/or the possibility that the phonon-phonon scattering is not bulk-like due to confinement in two dimensions. In comparison, the model does not predict either the magnitude or diameter dependence of the rough silicon nanowires of Hochbaum *et al.*,[14] pointing to the different nature of the phonon-boundary scattering in those wires.[15]

The Flik[1] and Majumdar[2] film thermal conductivity models are derived under the gray approximation (i.e., a single, mode-independent velocity, v_{ac}, and lifetime, τ_G) for $L/(\tau_G v_{ac}) \gg 1$ and use the Matthiessen rule for diffuse boundary scattering. They take the form

$$\frac{k}{k_\infty} = \frac{1}{1 + \dfrac{\tau_G v_{ac}}{\alpha L}}. \tag{7}$$

The values of α are $3\pi/2$ (Flik, In-Plane), 8/3 (Majumdar, In-Plane), and 3/4 (Majumdar, Cross-Plane). For the wire, with $D = L$ in Eq. (7), we set $\alpha = 4/3$.[22]

By using the kinetic theory expression for the bulk thermal conductivity, $k_\infty = \frac{1}{3} C v_{ac}^2 \tau_G$, where C is the volumetric specific heat (1.66×10^6 J/m^3-K for silicon at a temperature of 300 K), $\mathcal{L}$ and $\mathcal{D}$ can be expressed as $\frac{3}{2}\frac{1}{n_{pc}}\frac{C_\infty}{C}\frac{L}{\tau_G v_{ac}}$ and $\frac{4}{\pi}\frac{1}{n_{pc}}\frac{C_\infty}{C}\frac{D}{\tau_G v_{ac}}$, where n_{pc} is the number of atoms in the primitive cell (two for silicon) and C_∞ is the high-temperature harmonic specific heat ($3 n_{pc} k_B/\Omega$). The gray models are plotted as dashed lines in Figs. 2(a)–2(c) for all values of $\mathcal{L}$ or $\mathcal{D}$. In all cases, the gray models predict a much faster approach to the bulk thermal conductivity than the experimental data and our models. In full Brillouin-zone lattice dynamics calculations on SW silicon thin films, Sellan *et al.*[18] and Turney *et al.*[17] also found that gray models predict a too-fast approach to the bulk thermal conductivity. This faster approach to bulk occurs because gray models tend to underpredict the lifetime of the majority of the phonons, which can span many orders of magnitude.[18]

In summary, we presented a high-temperature analytical model for the size-dependence of film and wire thermal conductivity that requires no fitting parameters. For the film, the model provides convenient algebraic expressions. We tested the model by comparing its predictions against experimental data for silicon nanostructures. Good agreement was found for the in-plane direction for the film. When compared to gray models, our model better captures the approach to the bulk thermal conductivity for both films and wires. Extending this approach to lower temperatures, where phonon-defect scattering and quantum statistics must be considered, can be done in this framework but will require fitting parameters and numerical integration.

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