Phonon backscattering and thermal conductivity suppression in sawtooth nanowires

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The effect of surface roughness on phonon transport in a nanowire has often been described by treating the surface as flat with a specularity parameter (p) in the range between 0 and 1. A lower thermal conductivity limit is approached at $p=0$ for diffuse surface. It is demonstrated here by Monte Carlo simulation that sawtooth roughness on a nanowire can cause phonon backscattering and suppress the thermal conductivity below the diffuse surface limit. The backscattering effect can be accounted for only by a negative p if the detail of the surface roughness is ignored. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2970044]

In the current literature phonon-boundary scattering is treated as either purely diffuse, purely specular or a combination of both.[1] The specularity parameter (p) is generally used as a fitting parameter to simulate the probability of specular scattering.[2–4] In the case of a nanowire (NW) of diameter d, the effective mean free path (l_B) due to boundary scattering is given by[1,4]

$$l_B = \frac{1+p}{1-p} d. \tag{1}$$

For randomly distributed surface roughness that follows a Gaussian distribution, the specularity parameter p is often determined using the formula of Ziman as[1]

$$p = \exp(-16\pi^3 \delta^2/\lambda^2), \tag{2}$$

where δ is the root mean square surface roughness and λ is the phonon wavelength. For a perfectly smooth surface with δ several orders of magnitude smaller than the dominant phonon wavelength (λ_0), p from Eq. (2) approaches the upper bound of 1 corresponding to purely specular surface scattering. This type of scattering poses no resistance to phonon transport and thus no reduction in thermal conductivity (k) compared to bulk. When the surface roughness is just 0.3 nm, the formula of Ziman yields a value of $p \approx 0$ for $\lambda_0 \approx 1$ nm in silicon at room temperature. Corresponding to purely diffuse surface scattering, $p=0$ yields the lowest k of a NW of randomly distributed surface roughness when the bulk phonon dispersion is still applicable. This limit is referred as the diffuse surface limit (k_{diffuse}). Further increase in the surface roughness does not lead to appreciable reduction in either p from Eq. (2) or k calculated using the p value. Some existing k measurement results of NWs were found to be close to k_{diffuse}.[5,6] For a 22 nm diameter NW grown using a vapor liquid solid (VLS) method and 50–70 nm diameter rough silicon NWs obtained by electrochemical etching, the measured k was much lower than k_{diffuse}.[5,7] Note that the diameters of these silicon NWs are much larger than λ_0 for most of the temperature range of the measurement, so that the bulk phonon dispersion should still be applicable and the main phonon confinement effect is increased surface scattering and consequently suppressed k.

Here we investigate whether the surprisingly low k found in these two silicon NW systems is caused by phonon backscattering at the rough surface. Backscattering of gas molecules in a rough channel has been reported.[8] If such backscattering takes place for phonons in a NW with rough surfaces, k could potentially be reduced well below k_{diffuse}. Besides the rough NWs reported recently,[7] we note that sawtooth NWs have also been synthesized.[9,10] Using phonon transport analysis and Monte Carlo (MC) simulation, we show that phonon backscattering can indeed occur on such sawtooth surfaces, reducing k below k_{diffuse}.

To investigate the possibility of phonon backscattering we first employ a radiation analogy to analyze phonon scattering at a single sawtooth surface of a semi-infinite, isotropic, and isothermal solid. It is assumed that randomly distributed surface roughness is superimposed on both the left and right surfaces of the sawtooth so that both surfaces scatter phonons diffusely. Because of the diffuse surface scattering and the short coherence length of phonons, phonon interference is ignored. The phonon irradiation incident on the sawtooth is considered to be diffuse so that the phonon irradiation intensity I_i is independent of the incident angle (θ, ϕ) in the spherical coordinate. We calculate the rates of phonon energy incident from the left side, or with a positive velocity component along the x direction shown in Fig. 1(a), on the left and right sawtooth surfaces as

$$q_{i,+x,\text{left}} = w\frac{\delta}{\sin\theta_1} I_i \left(\int_{\theta=0}^{\theta_1} \int_{\varphi=0}^{2\pi} d\varphi \sin\theta\cos\theta d\theta + \int_{\theta=\theta_1}^{\pi/2} \int_{\varphi=0}^{\varphi_1} d\varphi \sin\theta\cos\theta d\theta \right) \tag{3a}$$

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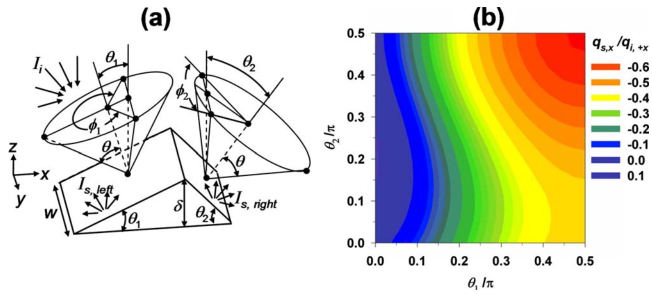

FIG. 1. (Color online) (a) Schematic of one sawtooth. (b) The calculated $q_{s,x}/q_i$ ratio as a function of sawtooth angles θ_1 and θ_2.

$$q_{i,+x,\mathrm{right}} = w\frac{\delta}{\sin\theta_2} I_i \int_{\theta=\theta_2}^{\pi/2}\int_{\varphi=0}^{\varphi_2} d\varphi \sin\theta\cos\theta d\theta, \qquad (3b)$$

respectively. In Eq. (3), $\varphi_1 = 2\pi - 2\cos^{-1}(\tan\theta_1/\tan\theta)$, $\varphi_2 = 2\cos^{-1}(\tan\theta_2/\tan\theta)$, and all the lengths and angles are shown in Fig. 1(a). The phonons incident from the left produce a diffuse scattered phonon intensity $I_{s,\mathrm{left}} = q_{i,+x,\mathrm{left}}\sin\theta_1/w\delta\pi$ and $I_{s,\mathrm{right}} = q_{i,+x,\mathrm{right}}\sin\theta_2/w\delta\pi$ respectively, on the left and right surfaces of the sawtooth. The net scattered phonon energy flux vector by each surface of the sawtooth is along the normal direction of the surface. By projection of the net scattered phonon flux vector from each surface along the x direction, we obtain the x component of the net rate of the scattered phonon energy by the sawtooth as

$$q_{s,x} = w\delta\left(\int_0^{2\pi} d\phi \int_0^{\pi/2} I_{s,\mathrm{right}}\sin\theta\cos^2\theta d\theta - \int_0^{2\pi} d\phi\int_0^{\pi/2} I_{s,\mathrm{left}}\sin\theta\cos^2\theta d\theta\right). \qquad (4)$$

As shown in Fig. 1(b), the $q_{s,x}/q_{i,+x}$ ratio varies from about 0.11 to −0.67, where $q_{i,+x} = q_{i,+x,,\mathrm{left}} + q_{i,+x,\mathrm{right}}$. At $\theta_1 = \theta_2 = 0$, this ratio is zero, which corresponds to completely diffuse scattering from a flat surface of random surface roughness that produces no net phonon flux along the x direction because the possibilities of incident phonons being forward scattered or backscattered are equal. At $\theta_1 < \pi/20$ and a wide θ_2 range, this ratio takes a small positive value of up to 0.11, which corresponds to partially forward and partially diffuse scattering by the sawtooth. For $\theta_1 > \pi/20$, the ratio is negative, suggesting incident phonon flux with a positive velocity component along the x direction are scattered by the sawtooth to produce a net scattered phonon flux along the $-x$ direction. Such scattering is equivalent to partially diffuse and partially backward scattering by the sawtooth.

The above analysis shows that backscattering of phonons is possible by an individual sawtooth. To understand the impact of periodic sawtooth roughness on k in a NW, we have employed MC simulations to calculate k of a Si NW with symmetric sawtooth facets and a square cross section, as shown in Fig. 2(a). The NW length was made to be much longer than the Umklapp phonon-phonon scattering mean free path at each temperature to avoid the temperature jumps noted in previous MC phonon transport simulations.[11] Specifically, our NW lengths were 20, 15, and 10 μm for temperatures of 100, 200, and 300 K, respectively. The minimum NW cross section was chosen to be 22×22 nm^2, or $d = 22$ nm in Fig. 2(a).

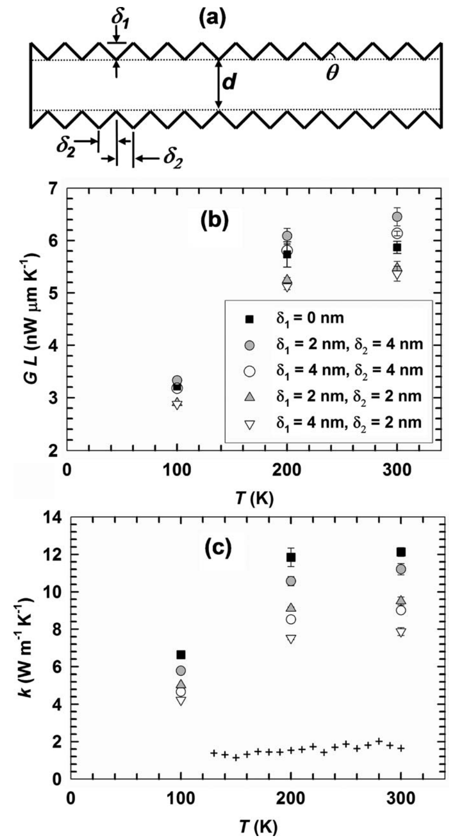

FIG. 2. (a) Schematic diagram, (b) thermal conductance-length product, and (c) thermal conductivity of a silicon NW with a square cross section and four side surfaces of symmetric sawtooth roughness. The legend in (b) is also applied to (c), except that the cross hair in (c) is the measured thermal conductivity of a 52 nm rough Si NW from Ref. 7.

The MC method follows the trajectories of a representative fraction of the entire phonon population that are subject to Umklapp and Normal phonon-phonon scattering, phonon-boundary scattering, and phonon-impurity scattering.[12] Different phonon polarizations and phonon dispersions are taken into account by considering the dependence of phonon lifetime on frequency, polarization, and temperature. Chen *et al.* have used the MC simulation method to calculate the thermal conductivity of silicon NWs of various diameters for a wide temperature range.[11] In the two reported MC simulations of Si thin films and NWs,[11,12] the rough surfaces were treated as flat. Without characterizing the actual phonon scattering event by the surface roughness in detail, these works incorporated the effect of surface roughness by introducing partially diffuse and partially specular phonon-surface scattering via the use of p between 0 and 1. For diffuse surface scattering, the velocity components were redistributed at the surface via random numbers while maintaining the phonon group velocity, effectively randomizing the direction of the reflection. Thus, phonon-boundary scattering was treated statistically according to the defined specularity parameter p and the generated random number rather than the specific nature of the surface.

In this work, we set all four side walls of the NW to possess sawtooth roughness elements. Specifically, periodic sawtooth elements are added to the four flat surfaces of the minimum 22×22 nm^2 square cross section, as shown in Fig. 2(a). The sawtooth height (δ_1) and length (δ_2) are varied from 2 to 4 nm. Each sawtooth face is assumed to scatter phonons following the diffuse surface conditions commonly used in MC simulation of thermal radiation transport.[13] Except for this modified boundary condition and the use of a simpler, more efficient algorithm of Chen *et al.* for impurity and Umklapp scattering, we have followed MC method of Chen *et al.*[11] Each case was run three times, with the starting value of the random number generator changed for each run. The data presented is the average of the three runs, with errors bars representing the random uncertainty at 90% confidence.

The thermal conductivity obtained by the MC simulation for the case of flat walls ($\delta_1=0$) and specular surfaces is 1061, 445, and 133 W/m K at temperature 100, 200, and 300 K, respectively, in agreement with literature values for pure bulk silicon[14] and the previous MC work.[11] For $\delta_1=0$ and diffuse surfaces, k is 6.64, 11.85, and 12.13 W/m K, respectively, for temperatures of 100, 200, and 300 K. These values for diffuse flat surfaces are essentially k_{diffuse}, and are considerably higher than the measurement results of a 22 nm VLS Si NW and rough Si NWs of larger diameters.[5,7]

In Fig. 2(b), we have plotted the thermal conductance (G)-length (L) product for the symmetric sawtooth surfaces of different δ_1 and δ_2 values. For the two cases of $\delta_1=2$ nm, $\delta_2=4$ nm and $\delta_1=4$ nm, $\delta_2=4$ nm, G is higher than the thermal conductance (G_{diffuse}) for the flat and diffuse case of $\delta_1=0$ nm, as generally expected because of the increase in the effective cross section area (A). However, when δ_1 is kept the same as these two cases and δ_2 is decreased from 4 to 2 nm to increase the sawtooth angle (θ), G becomes lower than G_{diffuse}. For these two latter cases, adding surface roughness elements on top of a flat NW suppresses G despite the increase in A. This effect is in agreement with backscattering of phonons by the added sawtooth surface roughness elements, as suggested by the more negative $q_{s,x}/q_{i,+x}$ ratio for larger θ_1 and θ_2 values of a single sawtooth in Fig. 1(b). Nevertheless, the backscattering effect for periodic sawtooth roughness is much more complicated than the case of an individual sawtooth element because of the presence of multiple phonon-surface scattering events between opposing sawtooth surfaces.

The cross section area A of a rough NW is not well defined. If we define A as the ratio between the volume and the length of the NW and use it to calculate k, the obtained k values in Fig. 2(c) of the sawtooth NWs would always be lower than k_{diffuse} of a 22×22 nm^2 flat and diffuse NW, which is expected to be lower than k_{diffuse} of a flat and diffuse NW with the same effective A as the sawtooth NW. For the two cases with a larger δ_1 of 4 nm, the calculated k is rather close to the measurement result of the 22 nm VLS NW,[5] but still considerably higher than those for rough Si NWs of 50–75 nm diameter.[7]

In conclusion we have shown that surface roughness can be engineered to introduce phonon backscattering and reduce k below k_{diffuse} in silicon NWs. If the details of surface roughness are ignored and the surface is represented with p, only a negative p in the range between 0 and −1 can yield $k<k_{\text{diffuse}}$. The limited p range between 0 and 1 from the formula of Ziman is practically useful only for randomly distributed roughness.

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[1]J. M. Ziman, *Electrons and Phonons: The Theory of Transport Phenomena in Solids* (Clarendon, Oxford, 1962).
[2]N. Mingo, L. Yang, D. Li, and A. Majumdar, Nano Lett. **3**, 1713 (2003).
[3]N. Mingo, Phys. Rev. B **68**, 113308 (2003).
[4]C. Dames and G. Chen, J. Appl. Phys. **95**, 682 (2004).
[5]D. Y. Li, Y. Y. Wu, P. Kim, L. Shi, P. D. Yang, and A. Majumdar, Appl. Phys. Lett. **83**, 2934 (2003).
[6]L. Shi, Q. Hao, C. H. Yu, N. Mingo, X. Y. Kong, and Z. L. Wang, Appl. Phys. Lett. **84**, 2638 (2004).
[7]A. I. Hochbaum, R. K. Chen, R. D. Delgado, W. J. Liang, E. C. Garnett, M. Najarian, A. Majumdar, and P. D. Yang, Nature (London) **451**, 163 (2008).
[8]A. S. Berman and W. J. Maegley, Phys. Fluids **15**, 772 (1972).
[9]H. L. Peng, S. Meister, C. K. Chan, X. F. Zhang, and Y. Cui, Nano Lett. **7**, 199 (2007).
[10]F. M. Ross, J. Tersoff, and M. C. Reuter, Phys. Rev. Lett. **95**, 146104 (2005).
[11]Y. F. Chen, D. Y. Li, J. R. Lukes, and A. Majumdar, J. Heat Transfer **127**, 1129 (2005).
[12]S. Mazumder and A. Majumdar, J. Heat Transfer **123**, 749 (2001).
[13]R. Siegel and J. Howell, *Thermal Radiation Heat Transfer* (Taylor & Francis, New York, 2002).
[14]C. J. Glassbrenner and G. A. Slack, Phys. Rev. **134**, A1058 (1964).