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Understanding length dependences of effective thermal conductivity of nanowires

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ABSTRACT

We have studied the length dependence of effective thermal conductivity of silicon nanowires by a thermon gas model and MD simulations. After modifications of the force term by considering the resistance enhancements from thermon gas interactions with the confined surfaces and the ends (inlet and outlet), the theoretical predictions of effective thermal conductivity agree well with the results of MD simulations in the length range of 4 to 550 nm. The result suggests that the resistance enhancement effect by thermon–boundary interactions, instead of the heat inertia, plays the dominating role in the non-Fourier heat conduction in silicon nanowires.

1. Introduction

Nanowires have received significant attention because of their unique properties and various potential applications in electronic, optical and energy conversion devices [1–3]. Particularly a better understanding of thermal transport in nanowires is crucial for new designs and optimizations of thermoelectric power generation [3–7] and thermal managements of microelectronics [8]. Numerous experiments have reported that the thermal transport in nanowires violates the Fourier's law [9–11], which describes a linear relationship between heat flux and temperature gradient [12]. Theoretical analysis based on the phonon Boltzmann equation often ascribes this violation to the contribution of ballistic conductions because of the smaller characteristic length than the phonon mean free path in nanomaterials [13–16]. The atomistic simulations are often used to predict the thermal properties of nanowires [17–19], and an early work has reported violations of Fourier's law even when the phonon mean free path is much shorter than the characteristic length in silicon nanowires [20]. The numerical results demonstrated such violations coming from insufficient phonon–phonon interactions in the low-dimensional lattice model [20–22]. Recently the phonon hydrodynamics [23–25] and the thermomass model [26,27] have been used to predict effective thermal conductivities of nanomaterials respectively, and have both received acceptable agreements with the experimental data. It is notable that both models assume that the heat carrier flows in nanomaterials are similar as rarefied gas flows in porous structures even though they are derived from different bases. Therefore in such models, heat transport is not necessarily divided into diffusive or ballistic regime, while the effective thermal conductivity depends on a Knudsen number defined as the ratio of characteristic lengths of the heat carrier and the nanomaterials [25–27].

Our early work has compared the predictions of length dependence of thermal conductivity for silicon nanowires based on the thermomass concept with the molecular dynamic (MD) simulation results and obtained excellent agreements for nanowires longer than 50 nm [27]. When the length was smaller than 50 nm, the theoretical predictions of thermomass model was higher than the MD results, but the mechanism was not revealed yet. In this Letter, we will revisit the resistance term in the governing equations of thermomass model and modify the equations by borrowing the newest results of resistance of rarefied gas flows in microchannels. Comparisons will be made between thermomass predictions with MD simulations for a wide length range. The mechanism of heat conduction in nanowires will be therefore discussed.

2. Theoretical and numerical methods

Since the famous caloric-dynamic argument in the 19th century, heat has been generally regarded as a process of energy transfer instead of substance transport [28]. Therefore constitutive equations for describing heat transfer behavior have hardly been established as those for substance transport, such as object movement or fluid flow. The Einsteinian mass–energy relationship proposed in

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the early 20th century presents the inherent equivalence between mass and energy [29], based on which a thermomass concept was proposed [30], stating that heat owns a mass–energy duality by exhibiting energy-like features in conversion and mass-like characteristics in transfer [31]. The effective mass of heat in transfer is calculated by the mass–energy equivalence, which is normally extremely small, i.e. 10^{-16} kg for 1 J heat, so that it has seldom measured before. However this mass may show its significance in ultrafast heating or ultrahigh-rate heat transfer process, such as in nanomaterials [26].

Since the heat carriers are different for different media, such as molecules for gas, phonons for pure crystals, electrons and phonons for pure metals, and more complicated for general media, let us name the quasi-particles carrying the transferred thermal energy on these carriers collectively "thermon". Because the moving speed of thermons is much less than the speed of light, the movement of thermons is still able to be described by the Newtonian mechanics. Borrowing from the classical gas dynamics, Guo et al. [32–34] derived the governing equations of thermomass in continuum, including the equation of state, the continuity equation and the momentum equation, and obtained a novel heat conduction equation for heat flux and temperature which is much more general and beyond the Fourier's law [31]. Eq. (1) shows the one-dimensional form of Guo's heat conduction equation in bulk solid materials based on thermomass concept,

$$\frac{\partial \mathbf{q}}{\partial t} - \frac{\mathbf{q}}{T}\frac{\partial T}{\partial t} + \frac{\mathbf{q}}{\rho C T}\frac{\partial \mathbf{q}}{\partial x} - \frac{q^2}{\rho C T^2}\frac{\partial T}{\partial x} + 2\gamma\rho C^2 T\frac{\partial T}{\partial x} + \mathbf{f}_T c^2 = 0, \tag{1}$$

where q is the heat flux, T the local thermodynamic equilibrium temperature, t the time, x the position, ρ the solid density, C the solid specific heat capacity, γ the Grüneisen constant, and $\mathbf{f}_T$ the effective resistance force per unit volume when the thermomass flows through the material lattices. The first two terms of Eq. (1) come from the temporal inertia of heat and the second two terms from the spatial inertia of heat. The fifth term comes from the driving force for thermomass by temperature differences. The resistance $\mathbf{f}_T$ can be determined by compatibility with the Fourier's law when all the inertial effects are negligible. For bulk materials, the effective resistance is

$$\mathbf{f}_T = \frac{2\gamma\rho C^2 T}{c^2 k}\mathbf{q}, \tag{2}$$

where c is the speed of light and k the intrinsic thermal conductivity of material. It is noticed that Eq. (1) has an analogous form to the phonon gas hydrodynamics equation [35] except for the resistance term. We use an effective resistance force instead of the viscous term in the phonon hydrodynamics equation. This simplification avoids troubles from thermon gas viscosity determination for complex materials and from exact description of interaction effects between the thermons and the lattices in materials. Therefore simple models can be incorporated to reflect geometric effects on the resistance of thermon gas flows [27].

Through this thermon gas model, the heat conduction driven by a temperature difference in a nanowire could be regarded analogous to the gas flow driven by a pressure difference in a nanochannel, as shown in Fig. 1. Our previous study on gas flow in microchannels [36] has indicated that the effective resistance is enhanced by interactions between gas and confined surfaces, following an exponential function of the characteristic length ratio [37]. Inspired by this result of gas flow, we proposed an effective resistance enhancement factor for the thermon gas model:

$$\mathbf{f}_T = \frac{2\gamma\rho C^2 T}{c^2 k(1 - e^{-D/l})}\mathbf{q}, \tag{3}$$

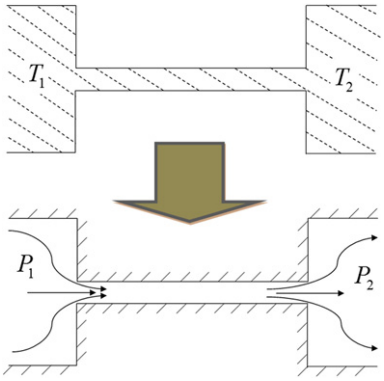

Fig. 1. Analogy of heat conduction in a nanowire to gas flow in a nanochannel in the thermon gas model. The effective resistance of thermon gas flow in nanowire will be modified based on the results of real gas flow in a nanochannel.

where D is the characteristic length of nanostructure vertical to the heat flux direction and l the characteristic length of heat conduction calculated by $qk/2\gamma C(\rho C T)^2$ [26,27]. Such a modification has led to excellent predictions of effective thermal conductivities of nanotubes [26] and nanofilms [27], compared with the available experimental data. However as stated before, the predictions for a silicon nanowire could only agree well with the MD simulations for high length to width ratio ($L/D > 50$ with L representing the length of nanowire). When the nanowire length gets smaller, the thermon gas model overrates the effective thermal conductivity of the nanowire, which means the resistance of the thermon gas is over-evaluated.

To understand the effective resistance of thermon gas in short nanowires, we may have to go back to real gas flows in nanochannels, as shown in Fig. 1. For gas flows in short channels, the ends effect, including inlet and outlet, also influences the overall effective resistance, besides the gas–surface interactions. A careful atomistic modeling study has shown that the ends effect also leads to an effective resistance enhancement by an exponential relationship with a finite ends region [37]. Involving this enhancement as well, the effective resistance for thermon gas flow in nanowires becomes:

$$\mathbf{f}_T = \frac{2\gamma\rho C^2 T}{c^2 k(1 - e^{-D/l})e^{-ND/L}}\mathbf{q}, \tag{4}$$

where L is the length of nanowire and N can be understood as the dimensionless length of ends effect region normalized by the channel width. For a straight channel, an empirical value of N is 5. It is clear that the ends effect will become negligible when the wire is long enough ($L/D > 50$).

Substituting the new effective resistance, Eq. (4), into Eq. (1) and neglecting the time dependent term lead to the governing equation for the steady non-Fourier heat conduction:

$$\left(1 - \frac{q^2}{2\gamma\rho^2 C^3 T^3}\right)k\frac{\partial T}{\partial x} + \frac{\mathbf{q}}{(1 - e^{-D/l})e^{-ND/L}} = 0, \tag{5}$$

where we define a dimensionless factor for inertial effect $F_i = q^2/2\gamma\rho^2 C^3 T^3$ and another dimensionless factor for resistance enhancement effect $F_r = 1/(1 - e^{-D/l})e^{-ND/L}$ to characterize each influence respectively. Note the k in this equation representing the intrinsic thermal conductivity, while the effective thermal conductivity is then calculated by $k_{eff} = qL/\Delta T$ after Eq. (5) numerically solved.

To validate the modified thermon gas model, we compare the predictions with our atomistic modeling results. In this Letter, we focus on silicon nanowires along [1 0 0] direction with cross-section of 3×3 unit cells, corresponding to a square cross-section

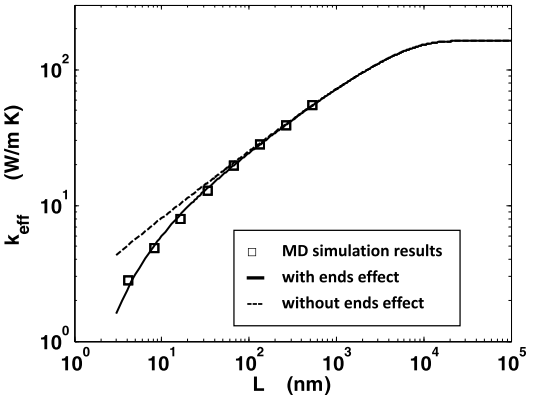

Fig. 2. Length dependence of thermal conductivity for silicon nanowires. The nanowires have a square cross-section of 1.22×1.22 nm^2. The given temperatures at the two ends are 330 K and 270 K. The symbols are data from MD simulations, the solid line is the theoretical prediction based on the thermon gas model with the ends effect considered (on Eq. (4)), and the dashed line is the prediction without ends effect (on Eq. (3)). The material properties used in the present predictions are $\rho = 2330$ kg/m^3, $k = 163$ W/m K, $C = 700$ J/kg K, and $\gamma = 1$ [44].

area at 1.22×1.22 nm^2. Free surface boundary conditions are adopted on the outer surface of the nanowire. The length of nanowire varies from 4 to 550 nm. We use the non-equilibrium molecular dynamics (NEMD) method in simulations, coded by ourselves. The Stillinger–Weber (SW) potential is used for silicon [38], which has been widely used to study the thermal property of silicon nanowires and silicon bulk materials [17,20,39–41]. To establish a temperature gradient along the nanowire, the atoms close to the two ends are put into heat bathes with given a higher or a lower temperature respectively. The Nose–Hoover heat bath is applied in our simulations [42,43]. While we have also checked that the results are independent of heat bath method [20]. Simulations are carried out long enough to reach a stationary state where the local heat flux is a constant along the nanowire and independent of time. After the system reaches a steady state, a time averaging of heat current is performed in 20 ns. The details of simulations and calculation formulae are available in Ref. [20].

3. Results and discussion

Fig. 2 shows the predictions of effective thermal conductivities of silicon nanowires changing with length by the thermon gas model (lines) compared with the MD simulation results (symbols). Two ends of nanowires are set at given temperatures 330 K and 270 K, which are high enough so that the quantum localization effects are negligible. The dashed line represents the predictions without the ends effect considered, which agree well with the MD results when the nanowire is long enough ($L/D > 50$) [27]. However for short nanowires, the predictions without the ends effect (Eq. (3)) overrate the effective thermal conductivity. The deviation increases as the nanowire becomes shorter, which means the resistance of thermon gas flow is underestimated and the ends effect is not negligible for short nanowires. After considering the resistance enhancement by the ends effect, the predictions by the thermon gas model agree with the MD simulations again for all the length range from 4 to 550 nm. The results also indicate that if the nanowire is rather long, longer than 10^4 nm for this case, the effective thermal conductivity will get close to the bulk value.

The effective thermal conductivity of nanowire may be much lower than the bulk value, and decreases with the reduced length nonlinearly. So far based on the thermon gas model, Eq. (5) suggests two factors that dominate the non-Fourier conduction in nanowire: the inertial effect of high-rate heat and the interactions between heat and surface in confined nanosurfaces [27]. Another

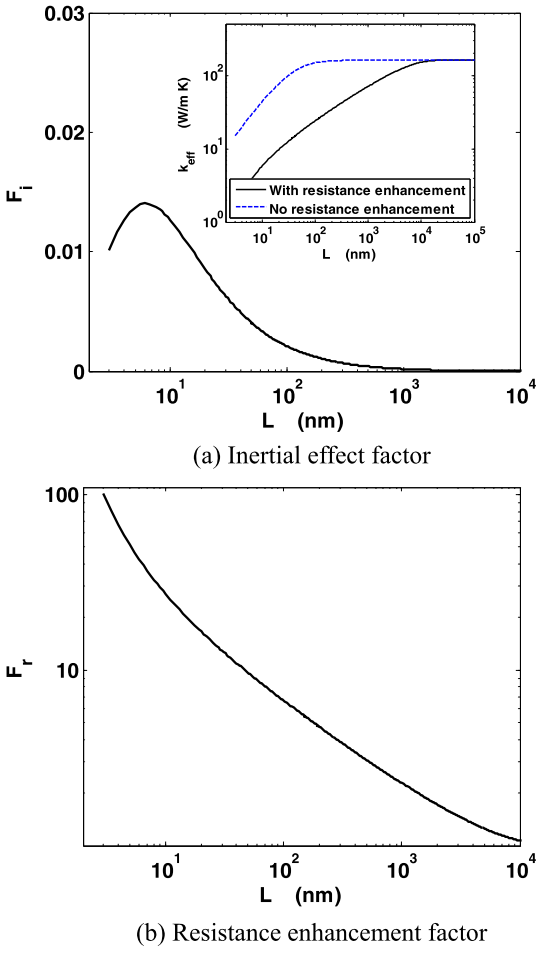

Fig. 3. Factors that characterize influences of the inertial effect of heat and the heat-surface interaction respectively for varying nanowire length. The factor equals unity if the corresponding influence is negligible. The subplot in (a) shows the predictions with or without resistance enhancement considered.

interesting question may rise consequently: which one is more important? Figs. 3(a) and (b) show the two dimensionless factors, F_i and F_r, to compare the relative significance changing with the length. A factor much less than unity means the corresponding effect negligible. The inertial effect factor is always less than 1.5% while the resistance enhancement factor may reach as high as 100 for very short nanowire cases. It indicates that the resistance enhancement effect by thermon–boundary interactions plays a dominating role in the non-Fourier heat conduction in nanowires. This result is warmly similar as that from the phonon hydrodynamics [45,46]. Meanwhile the very small value of F_i does not suggest that the inertial effect of heat is always negligible. The subplot in Fig. 3(a) shows the predicted thermal conductivities with or without resistance enhancement effect considered. It is clear that the inertia of heat can also lead to a significant decrease of conductivity for short nanowires. Since the influence of heat inertia depends on the heat flux value, it is just weakened because of the reduced heat flux impeded by the thermon–boundary interactions, as shown in Fig. 3.

In the previous study, the decrease of effective thermal conductivity of nanowires with length was ascribed to the insufficient phonon–phonon interaction in such a ballistic transport [20]. In this study, the non-Fourier heat conduction in nanowire is dominated by the thermon–boundary interactions, including the thermon–surface interaction and the ends effect, based on the thermon gas model. The understandings are different but not conflicting with each other. The phonon theory was based on a

microscopic model for qualitative analysis in that work, while the thermon gas model is a macroscopic model for quantitative predictions in this Letter. The insufficient phonon–phonon interaction suggests that the thermon gas in nanowires is a rarefied gas, therefore the resistance enhance model for the thermon gas flow is inspired by the real rarefied gas flow, which has been proved correct by comparisons with the MD results. The quantitative analysis based on our thermon gas model has led to more details of the nonlinear heat transfer in nanowires, such as the comparisons in Figs. 2 and 3, and more accurate understanding of mechanisms of length dependence of effective thermal conductivity of nanowires.

4. Conclusions

In summary, we have studied the length dependence of effective thermal conductivity of silicon nanowires by a thermon gas model, and compared the theoretical predictions with MD simulation results. After modifications of the force term by considering the resistance enhancements from thermon gas interactions with the confined surfaces and the ends (inlet and outlet), the theoretical predictions of effective thermal conductivity agree well with the MD simulations in the range of length scale, from 4 to 550 nm. The result suggests that the resistance enhancement effect by thermon–boundary interactions, instead of the heat inertia, plays the dominating role in the non-Fourier heat conduction in silicon nanowires. This research may help to understand non-Fourier thermal transport in low-dimensional nanomaterials and be useful to design and optimize devices in nanosystems as well since nanowires have numerous promising applications.

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References

[1] X.F. Duan, Y. Huang, R. Agarwal, C.M. Lieber, Nature 421 (2003) 241.
[2] M. Law, J. Goldberger, P.D. Yang, Annual Review of Materials Research 34 (2004) 83.
[3] A.I. Hochbaum, R.K. Chen, R.D. Delgado, W.J. Liang, E.C. Garnett, M. Najarian, A. Majumdar, P.D. Yang, Nature 451 (2008) 163.
[4] E.C. Garnett, M.L. Brongersma, Y. Cui, M.D. McGehee, Annual Review of Materials Research 41 (2011) 269.
[5] A.I. Boukai, Y. Bunimovich, J. Tahir-Kheli, J.K. Yu, W.A. Goddard, J.R. Heath, Nature 451 (2008) 168.
[6] S.K. Bux, R.G. Blair, P.K. Gogna, H. Lee, G. Chen, M.S. Dresselhaus, R.B. Kaner, J.P. Fleurial, Advanced Functional Materials 19 (2009) 2445.
[7] N. Yang, G. Zhang, B. Li, Nano Letters 8 (2008) 276.
[8] R.K. Chen, Nanowires for Thermal Energy Conversion and Management, Department of Mechanical Engineering, University of California, Berkeley, 2008.
[9] D.Y. Li, Y.Y. Wu, P. Kim, L. Shi, P.D. Yang, A. Majumdar, Applied Physics Letters 83 (2003) 2934.
[10] R. Chen, A.I. Hochbaum, P. Murphy, J. Moore, P.D. Yang, A. Majumdar, Physical Review Letters 101 (2008) 105501.
[11] A.J. Minnich, J.A. Johnson, A.J. Schmidt, K. Esfarjani, M.S. Dresselhaus, K.A. Nelson, G. Chen, Physical Review Letters 107 (2011) 095901.
[12] W. Dreyer, H. Struchtrup, Continuum Mechanics and Thermodynamics 5 (1993) 3.
[13] G. Chen, Physical Review Letters 86 (2001) 2297.
[14] G. Chen, Nanoscale Energy Transfer and Conversion, Oxford University Press, Oxford, 2005.
[15] R. Prasher, T. Tong, A. Majumdar, Nano Letters 8 (2008) 99.
[16] C. Dames, G. Chen, Journal of Applied Physics 95 (2004) 682.
[17] S.G. Volz, G. Chen, Applied Physics Letters 75 (1999) 2056.
[18] Y.F. Chen, D.Y. Li, J.R. Lukes, A. Majumdar, Journal of Heat Transfer – Transactions of the ASME 127 (2005) 1129.
[19] S.C. Wang, X.G. Liang, X.H. Xu, T. Ohara, Journal of Applied Physics 105 (2009) 014316.
[20] N. Yang, G. Zhang, B.W. Li, Nano Today 5 (2010) 85.
[21] J.S. Wang, B.W. Li, Physical Review Letters 92 (2004) 074302.
[22] B.W. Li, J. Wang, Physical Review Letters 91 (2003) 044301.
[23] F.X. Alvarez, D. Jou, Applied Physics Letters 90 (2007) 083109.
[24] F.X. Alvarez, D. Jou, Journal of Applied Physics 103 (2008) 094321.
[25] F.X. Alvarez, D. Jou, A. Sellitto, Applied Physics Letters 97 (2010) 033103.
[26] M. Wang, Z.Y. Guo, Physics Letters A 374 (2010) 4312.
[27] M.R. Wang, N. Yang, Z.Y. Guo, Journal of Applied Physics 110 (2011) 064310.
[28] E. Mendoza, Physics Today 14 (1961) 32.
[29] A. Einstein, H.A. Lorentz, H. Minkowski, H. Weyl, The Principle of Relativity, Dover Publications, New York, 1952.
[30] Z.Y. Guo, Journal of Engineering Thermophysics 27 (2006) 631.
[31] M. Wang, B. Cao, Z.Y. Guo, Frontiers in Heat and Mass Transfer 1 (2010) 013005.
[32] Z.Y. Guo, H.Y. Zhu, Journal of Engineering Thermophysics 28 (2007) 86.
[33] B.Y. Cao, Z.Y. Guo, Journal of Applied Physics 102 (2007) 2775215.
[34] Z.Y. Guo, B.Y. Cao, Acta Physica Sinica 57 (2008) 4273 (in Chinese).
[35] V.A. Cimmelli, A. Sellitto, D. Jou, Physical Review B 82 (2010) 184302.
[36] M.R. Wang, Z.X. Li, Journal of Engineering Thermophysics 25 (2004) 840.
[37] X.D. Shan, M. Wang, Microfluidics Nanofluidics (2012), in press.
[38] F.H. Stillinger, T.A. Weber, Physical Review B 31 (1985) 5262.
[39] J. Chen, G. Zhang, B.W. Li, Nano Letters 12 (2012) 2826.
[40] Y. Lee, G.S. Hwang, Physical Review B 85 (2012) 125204.
[41] R. Rurali, Reviews of Modern Physics 82 (2010) 427.
[42] S. Nose, Journal of Chemical Physics 81 (1984) 511.
[43] W.G. Hoover, Physical Review A 31 (1985) 1695.
[44] http://en.wikipedia.org/wiki/.
[45] F.X. Alvarez, D. Jou, A. Sellitto, Journal of Applied Physics 105 (2009) 014317.
[46] F.X. Alvarez, D. Jou, A. Sellitto, Journal of Heat Transfer – Transactions of the ASME 133 (2011) 022402.