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Length-dependent thermal transport and ballistic thermal conduction

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Probing length-dependent thermal conductivity of a given material has been considered as an important experimental method to determine the length of ballistic thermal conduction, or equivalently, the averaged phonon mean free path (l). However, many previous thermal transport measurements have focused on varying the lateral dimensions of samples, rendering the experimental interpretation indirect. Moreover, deducing l is model-dependent in many optical measurement techniques. In addition, finite contact thermal resistances and variations of sample qualities are very likely to obscure the effect in practice, leading to an overestimation of l. We point out that directly investigating one-dimensional length-dependent (normalized) thermal resistance is a better experimental method to determine l. In this regard, we find that no clear experimental data strongly support ballistic thermal conduction of Si or Ge at room temperature. On the other hand, data of both homogeneously-alloyed SiGe nanowires and heterogeneously-interfaced Si-Ge core-shell nanowires provide undisputed evidence for ballistic thermal conduction over several micrometers at room temperature. [http://dx.doi.org/10.1063/1.4914584]

The empirical Fourier's law suggests that macroscopic heat conduction phenomena are diffusive phenomena, i.e. phonons lose their wave properties when propagating beyond an averaged distance called phonon mean free path (l). However, interesting ballistic thermal conduction would occur when the sample length (L) is shorter than the l. In principle, ballistic thermal conduction should obey Landauer's formulation for quantum thermal conduction (K_Q):[1]

$$K_Q = \frac{k_B^2}{h} \sum_m \int_{x_m}^{\infty} dx \frac{x^2 e^x}{(e^x - 1)^2} T_m(\pi^2 k_B^2/\hbar) \approx \frac{\pi^2 k_B^2 T}{3h}, \qquad (1)$$

where is k_B Boltzmann constant, T_m is the transmission coefficient of the mth phonon mode. At low temperatures, Landauer's formula can be reduced to a simple form expressed by fundamental constants, as shown in Eq. (1). According to Eq. (1), no dissipation will occur inside a ballistic conductor and thermal resistance only happens at the contacts, resulting in a length-independent thermal resistance ($1/K$, where K is thermal conductance) effect for $L < l$. Equivalently, the behavior will result in a linear L-dependence of thermal conductivity (κ) for $L < l$, too. Furthermore, the length scale at which the thermal transport transits from ballistic to diffusive is generally denoted as the experimentally determined l.

Many experiments have been carried out to determine the l of Si. For example, Goodson's group studied thickness-dependent, in-plane κ of silicon-on-insulator wafer by repeated thermal

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oxidation and wet etching.[2,3] The l was estimated to be 300nm instead of 43nm predicted by a dispersionless theory.[3] Liu and Asheghi further studied κ of doped Si films with thinner thickness.[4] Li *et al*. studied diameter-dependent κ of Si nanowires and found that the κ decreases with decreasing diameter.[5] Alvarez and Jou did theoretical analyses on the size dependent thermal transport phenomena of Si.[6] It should be noted that the above measurements were based on samples with different lateral dimensions and thus did not rigorously obey the operational definition of ballistic thermal conduction described above. Because investigating l by varying the lateral dimensions of a sample would need to adapt additional assumptions on the specular/diffusive boundary scatterings or the validity of Casimir limit, the estimation of l is indirect. In fact, the experimental verification of quantum thermal conduction of Eq. (1) provides a good example that the lateral confinement of the SiN_x (200nm wide, 60nm thick) beam does not necessarily affect the ballistic thermal conduction along the beam (~4μm).[7] Rigorously speaking, the experimental data collected from varying lateral dimensions of various Si samples should not be regarded as evidence for ballistic thermal conduction below some characteristic size l.

On the other hand, there have been interests in utilizing optical techniques to probe l's of materials. Here heat is generated after the material absorbs the laser light. Then the temperature variation can be probed by another laser beam based on the temperature-dependence of the refractive index of the material. Recently, various techniques including time-domain thermoreflectance (TDTR),[8,9] frequency-domain thermoreflectance (FDTR)[10] have been employed to study l's. For these optical techniques, heat generation in materials requires absorption of laser power. But the investigated materials may not efficiently absorb light and convert it into heat. Besides, the modulation of refractive index of semiconductors could be mixed with phononic and electronic responses. To solve the problem, Al thin film is commonly employed as a thermal transducer in the experiments. However, it introduces another problem of thermal contact resistances in these optical measurements.[11] The thermal transport at the interface is difficult to model, but it certainly plays an important role in enhancing the anisotropy in an otherwise isotropic material.[11] The interpretation of optical measurement results without considering the role of interface may also lead to inconsistent conclusions. In TDTR or FDTR, changes of κ's were observed by changing spot sizes or the modulation frequency of the pump laser. The variation of spot sizes primarily reveals the in-plane heat conduction where the modulation frequency indicates the cross-plane phonon transport. Wilson and Cahill recently pointed out the discrepancy of previous results originated from anisotropic thermal transport in-plane and cross-plane.[11] Yet deducing the l's is not at all straightforward, as experimental complexities associated with laser spot sizes, laser modulation frequencies, temperature profiles, and interface phonon scatterings are quite involved.[11] Moreover, theoretical interpretations of the optical measurement results remain controversial. While it is commonly assumed that the observed deviation to be due to ballistic phonons, *ab-initio* calculations suggest that agreements on Si are better if simply considering harmonic and anharmonic phonon channels but without directly incorporating ballistic phonons.[12] Thus the interpretation of the experimental data is model-dependent and nontrivial.

One should be reminded that the original definition of ballistic thermal conduction is based on the length-dependent thermal transport. Experimentally, the measured system should be kept as simple as possible to minimize unwanted complexities. So far only a few experiments have followed this guideline to study l's.[13–16] Alvarez-Quintana *et al*. studied thickness-dependent cross-plane κ of germanium-on-insulator wafer and estimated the $l \sim 20$nm.[13] Johnson *et al*. employed laser-induced transient grating method to study in-plane κ of Si membranes and found the κ deviates from diffusive behavior at several micrometers, suggesting $l > 1$μm.[15] In transient grating, changes of κ's were studied by varying the one-dimensional grating periods in silicon membrane and the experiment was designed to avoid problems of interfaces introduced by metal tranducers.[15] The experiment thus reduced the complexities associated with TDTR or FDTR. Recently, we have also recognized the issues mentioned above and directly measured the length-dependent κ of homogeneously-alloyed SiGe nanowires and heterogeneously-interfaced Si-Ge core-shell nanowires.[14,16] Surprisingly, the l of these SiGe nanowires was found to be 5~8μm, which is nearly 1000 times longer than that predicted by some theories.[17] Interestingly, the diameter-dependent of κ of the SiGe nanowires displays a much weaker dependence than those observed in Si or Ge nanowires.[5,14,18] The study on the SiGe nanowires also raises a subtle yet important issue on the validity of probing l by changing the lateral dimensions of a sample, as discussed above.

However, carrying out the desired length-dependent thermal transport measurement to determine l is not a simple task in practice. Because sometimes the data were collected from different samples of various sizes, they may inevitably exhibit large uncertainties due to variations of sample quality. Furthermore, some previous works had focused on the discussions on the length-dependent of κ alone,[13] rendering the observed behavior likely to be confused with effects of contact resistance.

Physically, thermal transport measurements usually require connecting leads to a sample, which would often result in finite contact thermal resistances. Thus the experimentally measured total thermal resistance ($1/K_{total}$) can be expressed as:

$$\frac{1}{K_{total}} = \frac{1}{K_{sample}} + \frac{1}{K_{contact}}, \tag{2}$$

where $1/K_{contact}$ is contact thermal resistance, which can be reasonably modeled as a constant, and $1/K_{sample}$ is sample resistance, which can be described as

$$1/K_{sample} = \begin{cases} 1/K_Q, & \text{for } L \leq l, \\ (1/K_Q)\,\dfrac{L}{l}, & \text{for } L > l, \end{cases} \tag{3}$$

by assuming that ballistic behavior takes place and $1/K_{sample}$ becomes constant when $L < l$, whereas diffusive behavior dominates and is proportional to L when $L > l$. One can refine the simple model of Eq. (3) and smooth out the kink at $L = l$ by introducing a small but finite characteristic length Δ for the ballistic-diffusive transition. By using the integral of the logistic function (viewed as a smoothed step function), a smooth function that provides a phenomenological model of $1/K_{sample}$ can be constructed as

$$\begin{aligned} 1/K_{sample}(L) &= \frac{1}{K_Q} + \frac{1}{K_Q l}\int_0^L \frac{1}{1+e^{-(x-l)/\Delta}}dx \\ &= 7\frac{1}{K_Q} + \frac{1}{K_Q l}\left(\Delta \log\left(e^{l/\Delta} + e^{L/\Delta}\right) - \Delta \log\left(1 + e^{l/\Delta}\right)\right). \end{aligned} \tag{4}$$

If ballistic thermal conductance does not take place at all, we simply have the diffusive behavior $1/K_{sample} = aL$ for all $L > 0$, and consequently the thermal conductivity $\kappa \equiv KL/A$ yields a constant regardless of L. For this reason, the observation that κ decreases with decreasing L is often regarded as a signal of the occurrence of ballistic thermal conductance. This signal, however, could be fallacious, because the decrease of κ with decreasing L may be explained by the presence of a nonzero contact resistance. For finite contact resistance without any ballistic behavior, Eq. (2) reads as

$$\frac{1}{K_{total}} = aL + \frac{1}{K_{contact}}, \tag{5}$$

and consequently we have

$$\kappa \equiv \frac{K_{total}L}{A} = \frac{L/A}{aL + 1/K_{contact}}, \tag{6}$$

which leads to $\kappa \to 0$ as $L \to 0$. To illustrate the fallacy, we plot κ vs. L and $1/K_{total}$ vs. L using Eq. (5) & (6) in Fig. 1(a) & 1(b), with added noises to simulate the experimental data. If one merely focuses on the decrease of κ, the phonon mean free path l could be erroneously identified to be about 7, significantly deviated from the actual value ($l = 0$).

To determine the value of l rigorously, one has to perform regression analysis on the experimental data (of κ or $1/K_{total}$) in response to all range of L, and it turns out Eq. (4) serves as a sound regression function. A reliable result of regression analysis not only gives the best-fitted values of l, $1/K_Q$, and $1/K_{contact}$ but should also yield stringent standard errors for them. If regression analyses yields $l > 0$ with a small standard error, ballistic thermal conduction can be claimed.

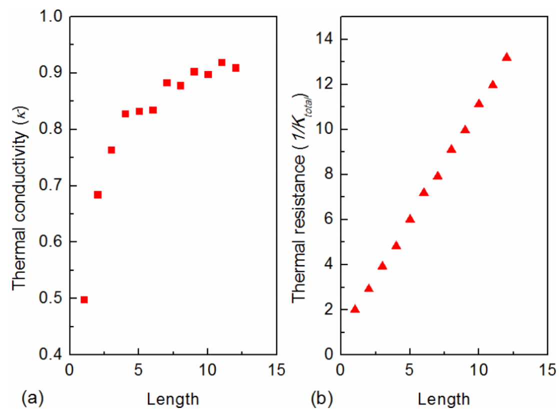

FIG. 1. Simulated (a) κ vs. L and (b) $1/K_{total}$ vs. L relations for $1/K_{contact} = 1$. Here some noises are added to the data to simulate the experimental observation. It can be seen that finite contact thermal resistance will create a pseudo ballistic thermal conduction effect in the κ vs. L relation even if the sample is a diffusive conductor.

To easily visualize the ballistic thermal conduction, a convenient method is to plot the $1/K_{total}$ vs. L relation. The $1/K_{total}$ vs. L relation would allow us to separate the ballistic thermal conduction from the unwanted effect of contact thermal resistance by reading the offset at $L \to 0$, as shown in Fig. 1(b). The ballistic thermal conduction can be seen by observing the deviation from diffusive behavior, extrapolating from the data of large L's (the dashed line of Fig. 1(b)). Although the method may give a conservative estimate on l, it is undoubtedly a more reliable method.

We now reexamine the previous data of Ge and Si,[13,15] whose κ vs. L relation is shown in Fig. 2(a) and 2(c), respectively. Because the effective lengths were collected from different samples of different dimensions, we employ the relation $A/K_{total} = L/\kappa$ and plot normalized thermal resistance (A/K_{total}) vs. L relations in Fig. 2(b) & 2(d) instead. Clearly, the A/K_{total} vs. L relation shown in Fig. 2(b) & 2(d) does not deviate from diffusive transport behavior down to L = 42nm for Ge film and L = 1µm for Si membranes. It is interesting to note that in Ref. 15, heat was actually directly generated in silicon membrane by laser without a metal transducer. Yet a finite "effective contact thermal resistance" is still observed in Fig 2(d). Nevertheless, because the data in Fig. 2(b) & 2(d)

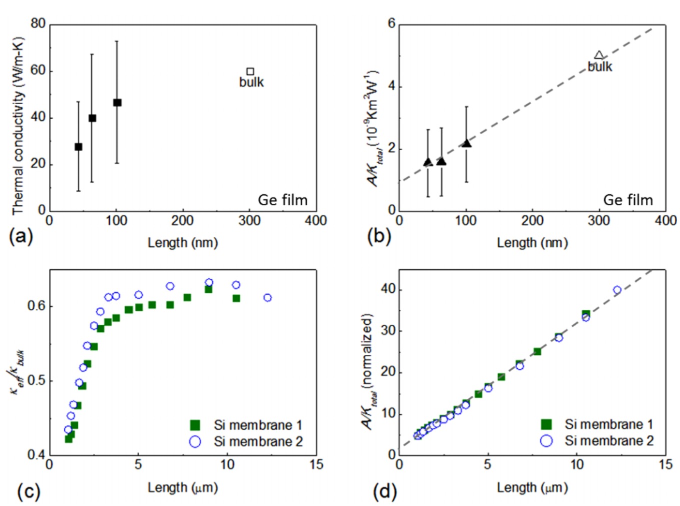

FIG. 2. (a) κ vs. L and (b) A/K_{total} vs. L relations for Ge film. The A/K_{total} vs. L relation follows the diffusive behavior within the error bars for $L > 42$nm, as indicated by the gray dashed line. The data are reproduced from Ref. 13. The open symbols denote the bulk values of Ge. (c) κ vs. L and (d) A/K_{total} vs. L relations for Si membranes. The A/K_{total} vs. L relation follows the diffusive behavior for $L > 1$µm, as indicated by the gray dashed line. The data are reproduced from Ref. 15. Here the lengths are half of the grating period shown in Ref. 15 and the y-axes are normalized by the bulk values.

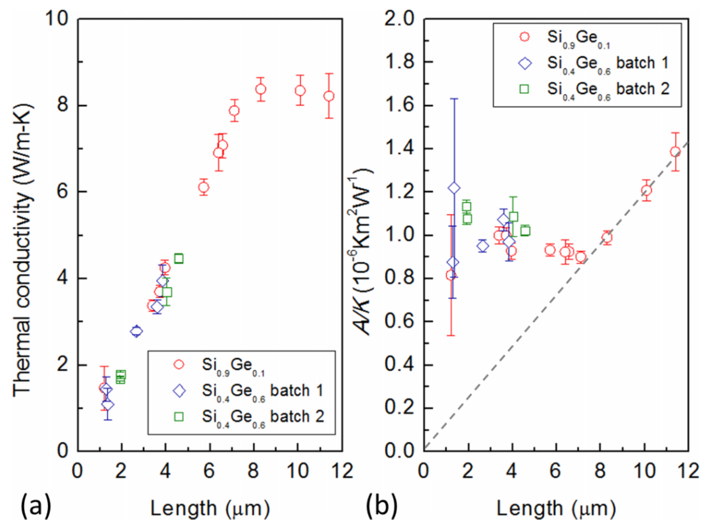

FIG. 3. (a) κ vs. L and (b) A/K_{total} vs. L relations for homogeneously-alloyed SiGe nanowires. Both κ vs. L and A/K_{total} vs. L relations deviate from the diffusive behavior for $L < 8\mu m$, indicating the ballistic thermal conduction. The gray dashed line is a fitted line extrapolating from data of $L > 8\mu m$. The data are reproduced from Ref. 14.

do not deviation from the diffusive behavior, there is no observable ballistic thermal conduction of Si or Ge at room temperature.

The reanalysis shown in Fig. 2(b) & 2(d) certainly raises concerns on the validity of our recent discovery of ballistic thermal conduction over 8.3μm for homogeneously-alloyed SiGe nanowires.[14] Especially, could the data be interpreted as ordinary diffusive thermal conductors with large contact thermal resistance? To answer the question, we plot the κ vs. L and A/K_{total} vs. L on Fig. 3(a) & 3(b). Figure 3(a) shows that the κ increases linearly with L, followed by a slope change at $L = 8.3\mu m$, then the κ saturates at 8.2 W/m-K, agreeing with the bulk value of SiGe. Although Fig. 3(a) alone seems to suggest $l = 8.3\mu m$ for SiGe nanowires, it is necessary to reexamine the data by plotting A/K_{total} vs. L in Fig. 3(b). From Fig. 3(b), A/K_{total} decreases linearly with L for $L > 8\mu m$, indicating the expected diffusive transport behavior. Interestingly, the A/K_{total} vs. L changes the slope at $L = 8\mu m$ and it significantly deviates from the diffusive behavior for $L < 8\mu m$. Applying the regression analysis mentioned above gives $l = 8.05\mu m$ with standard deviation =0.31μm. Moreover, extrapolating the diffusive behavior to $L \to 0$ suggests that the (classical) contact thermal resistance is nearly zero, which indicates that the observed ballistic thermal conduction shown in Fig. 3(a) cannot be attributed to effects of finite contact thermal resistance. We emphasize that the x-axes of Fig. 3(a) & 3(b) are "length" rather than "effective length" and thus the experimental investigations provide the direct probing of l. In addition, experimental demonstrations of quantum contact resistances and non-additive thermal resistance in series have provided independent strong supports to the claimed ballistic thermal conduction.[14] Therefore, reexamining the data reaffirms the ballistic thermal conduction for at least 8μm in homogeneously-alloyed SiGe nanowires. The result highlights the important role of disorders or alloys in filtering out high frequency phonons contributing to the total thermal conductivity.[19–22]

Similar analyses can be applied to data of heterogeneously-interfaced Si-Ge core-shell nanowires, too.[16] However, unlike the homogeneously-alloyed SiGe nanowires discussed above, different heterogeneously-interfaced Si-Ge core-shell nanowires always exhibit distinct thickness of Si-core or Ge-shell. Besides, structural variations, changes of l's, or uncertainties in the contacts in different samples may further complicate the analyses. To minimize the unwanted effects associated with different samples, we thus focus our analyses on a sample investigated by an electron-beam heating technique.[16,23] Figure 4(a) displays κ vs. L for a Si-Ge core-shell nanowire of diameter ~150nm. The slope of the κ vs. L changes at $L = 5\mu m$, which seems to indicate $l = 5\mu m$. Because the measurement was conducted on the same sample, we can plot $1/K_{total}$ vs. L (instead of A/K_{total} vs. L) in Fig. 4(b).

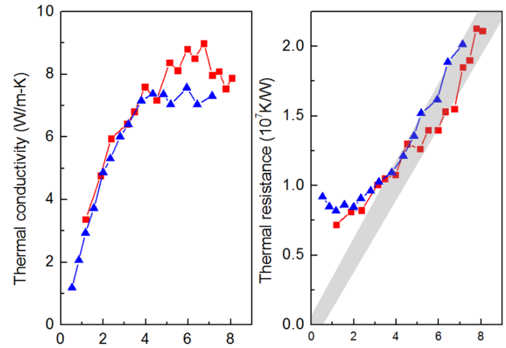

FIG. 4. (a) κ vs. L and (b) $1/K_{total}$ vs. L relations for a heterogeneously-interfaced Si-Ge core-shell nanowire (of diameter ~150nm) investigated by an electron-beam heating method. It can be seen that both κ vs. L and $1/K_{total}$ vs. L relations deviate from the diffusive behavior for $L < 2.5$μm, indicating the ballistic thermal conduction. The gray belt is a fitted line extrapolating from data of $L > 5$μm. The data are reproduced from Ref. 16.

Extrapolating the data of $L > 5$μm to $L \to 0$ indicates that the contact thermal resistance is less than 1.6×10^6 K/W. Interestingly, $1/K_{total}$ deviates from the diffusive behavior at $L \sim 2.5$μm, indicating $l \sim 2.5$μm for the sample (the regression analysis gives $l = 2.61$μm with standard deviation = 0.25μm).

In summary, we point out that directly investigating one-dimensional length-dependent (normalized) thermal resistance is a better experimental method to determine l. It directly follows the definition of ballistic thermal conduction and avoids many model-dependent complexities commonly exist in previous thermal transport or optical measurements. The $1/K_{total}$ vs. L (or A/K_{total} vs. L) relation also allows us to separate unwanted effect of contact thermal resistance. In this regard, we have discussed the problems in previous measurements and found that no clear experimental data strongly support ballistic thermal conduction of Si or Ge at room temperature. On the other hand, applying regression analyses to the data of both homogeneously-alloyed SiGe nanowires and heterogeneously-interfaced Si-Ge core-shell nanowires provide undisputed evidence for ballistic thermal conduction over several micrometers at room temperature.

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