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Thermal conductivity of individual silicon nanowires

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The thermal conductivities of individual single crystalline intrinsic Si nanowires with diameters of 22, 37, 56, and 115 nm were measured using a microfabricated suspended device over a temperature range of 20-320 K. Although the nanowires had well-defined crystalline order, the thermal conductivity observed was more than two orders of magnitude lower than the bulk value. The strong diameter dependence of thermal conductivity in nanowires was ascribed to the increased phonon-boundary scattering and possible phonon spectrum modification. © 2003 American Institute of Physics. [DOI: 10.1063/1.1616981]

One-dimensional (1D) materials such as various kinds of nanowires and nanotubes have attracted considerable attention due to their potential application in electronic and energy conversion devices.¹⁻⁴ In contrast with the extensive studies on electron transport, investigation of phonon transport in 1D nanostructures^{5–9} has been initiated only recently. When crystalline solids are confined to the nanometer range, phonon transport within them can be significantly altered due to various effects, namely (i) increased boundary scattering; (ii) changes in phonon dispersion relation; and (iii) quantization of phonon transport. For example, theoretical studies^{7,8} have suggested that, as the diameter of a Si nanowire becomes smaller than 20 nm, the phonon dispersion relation could be modified due to phonon confinement, such that the phonon group velocities would be significantly less than the bulk value. Molecular dynamics simulations⁹ have shown that, for wires of nanometer diameter, the thermal conductivities could be two orders of magnitude smaller than that of bulk silicon. However, to the best of our knowledge, no systematic experimental results have been reported on the thermal conductivities of Si nanowires. It is, therefore, important to experimentally validate these theoretical predictions to understand the underlying physics. In addition, a deeper understanding of thermal transport in nanostructures also has practical implications¹⁰ in the design and performance of modern microelectronic devices that have sub-100 nm features and recently proposed nanowire-based thermoelectric devices. For these reasons, we report in this letter a systematic experimental study of the size effect on Si nanowire thermal conductivity. Results show that Si nanowire thermal conductivity is much lower than the corresponding bulk value, which can be explained by increased phonon boundary scattering. Furthermore, the experimental results for a 22 nm diam silicon nanowire shows that the low-temperature behavior of its thermal conductivity significantly deviates from Debye T^3 law, which suggests possible changes in the phonon dispersion relation due to confinement.

Figure 1 shows a typical microdevice used in our experiments. An individual Si nanowire thermally connects two suspended microfabricated microstructures. The suspended microstructure consists of two silicon nitride (SiN_x) mem-



FIG. 1. SEM image of the suspended heater. The lower inset shows a 100 nm Si nanowire bridging the two heater pads, with wire-pad junctions wrapped with amorphous carbon deposits (shown by arrows). The scale bar in the inset represents 2 μ m.

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FIG. 2. High-resolution TEM image of a 22 nm single crystal Si nanowire. The inset is a selected area electron diffraction pattern of the nanowire.

branes each suspended by five SiN_x beams that are 420 μ m long and 0.5 μ m thick. A thin Pt resistance coil and a separate Pt electrode are patterned onto each membrane. Each resistor is electrically connected to four contact pads by the metal lines on the suspended legs, thus enabling four-point measurement of the voltage drop and resistance of the resistor. Each Pt resistor can serve as a heater to increase the temperature of the suspended island, as well as a resistance thermometer to measure the temperature of each island.

The Si nanowires were synthesized by the vapor– liquid–solid method,¹¹ in which Au clusters were used as a solvent at high temperature. The Si and Au formed a liquid alloy and when the alloy became supersaturated with Si, Si nanowires grew by precipitation at the liquid–solid interface. A high-resolution transmission electron microscopy (TEM) investigation (see Fig. 2) showed that the Si nanowires were single crystalline and grew along the $\langle 111 \rangle$ direction. The wire diameters fell in the range of 10–200 nm and the lengths were several microns.

Once synthesized, the nanowires were first dispersed in isopropanol by sonication, and then drop cast onto suspended heater devices. After drying the solvent, we found that an individual nanowire often bridged the two islands. In order to improve thermal contact between the wires and the suspended devices, amorphous carbon films were locally deposited at the nanowire–heater pad junctions with a scanning electron microscope (SEM) as shown in the inset of Fig. 1.

All the measurements were carried out at a high vacuum level of $\sim 2 \times 10^{-6}$ Torr and temperature ranging up to 320 K to suppress residual gas conduction and radiation loss. A detailed description of the measurement technique and uncertainty analysis is summarized elsewhere.^{12,13} Briefly, bias voltage applied to one of the resistors, R_h , creates Joule heating and increases the temperature, T_0 . Under steady state, part of the heat will flow through the nanowire to the other resistor, R_s , and raise its temperature, T_s . By solving the heat transfer equations of the system,¹³ denoting the thermal conductance of the wire G_w and the suspending legs G_l , we have

$$T_h = T_0 + \frac{G_l + G_w}{G_l (G_l + 2G_w)} P$$

and



FIG. 3. (a) Measured thermal conductivity of different diameter Si nanowires. The number beside each curve denotes the corresponding wire diameter. (b) Low temperature experimental data on a logarithmic scale. Also shown are T^3 , T^2 , and T^1 curves for comparison.

$$T_s = T_0 + \frac{G_w}{G_l(G_l + 2G_w)}P,$$

where $P = I^2(R_h + R_l/2)$. Here R_l is the total electrical lead resistance of Pt lines that connects the heater coil. From the slopes of T_h and T_s vs P, the thermal conductivity of the bridging nanowire can be estimated after considering the diameter and length of the wires. In the experiments, the I-Vcurve is measured as the dc current I is slowly ramped up to a value in the range of 6–12 μ A depending on T_0 . The maximum power dissipation on the heating membrane is below 1 μ W and the maximum rise in temperature on the heating side is below 5 K.

The measured thermal conductance includes the thermal conductance of the junction between the nanowire and the suspended islands in addition to the intrinsic thermal conductance of the nanowire itself. We have estimated the thermal conductance of the junctions with the carbon deposition, and found that the junction contribution is less than 15% of the total thermal transport barrier.¹³

Shown in Fig. 3(a) are the measured thermal conductivities for intrinsic single-crystalline Si nanowires of different diameters (22, 37, 56, and 115 nm). Compared to the thermal conductivity of bulk Si,¹⁴ there are two important features that are common to all the nanowires we measured: (i) The measured thermal conductivities are about two orders of magnitude lower than that of the bulk and, as the wire diameter is decreased, the corresponding thermal conductivity is reduced. This clearly indicates that enhanced boundary scattering has a strong effect on phonon transport in Si nanowires. (ii) For the 37, 56, and 115 nm diam wires, thermal conductivities reach their peak values around 210, 160, and 130 K, respectively. This is in sharp contrast to the peak of bulk Si that occurs at about 25 K. The shift of the peak suggests that, as the wire diameter is reduced, the phonon boundary scattering dominates over phonon–phonon umklapp scattering, which decreases the thermal conductivity with an increase in temperature. In addition, the thermal conductivity of the 22 nm diam wire does not exhibit a peak within the experimental temperature range.

The temperature dependence of the thermal conductivity between 20 and 60 K is plotted on a log-log scale in Fig. 3(b). It can be seen that the data for the 115 and 56 nm diam wires fit Debye T^3 law quite well in this temperature range. This suggests that boundary scattering, which is frequency and temperature independent, is the dominant phonon scattering mechanism, such that the thermal conductivity follows the temperature dependence of specific heat. As the nanowire diameter is reduced, one would expect boundary scattering to dominate even more. However, for the smaller diameter wires, 37 and 22 nm, the power exponent gets smaller as the diameter decreases and for the 22 nm wire the deviation from Debye T^3 law can be clearly seen. This means that besides phonon boundary scattering, some other effects could play important roles. Although more experiments must be performed before solid conclusions can be drawn, several previous investigations^{7,8,15–18} have suggested that, as the characteristic size scales down to the order of 10 nm, the phonon dispersion relation could be modified due to confinement, which would lead to reduced phonon group velocities. It was further claimed⁸ that for a 20 nm silicon nanowire, the overall phonon group velocity would be reduced by a factor of 1/2 compared with the bulk value, which could explain the observed deviation. In addition, a recent experimental study¹⁹ on GaAs nanobeams suggests that using real phonon group velocity, instead of the Debye values, might be important for analyzing the phonon transport in nanostractures. However, changes in phonon dispersion could also modify the selection rules and scattering rate of phonon-phonon scattering, but they have not been studied so far. Hence, it is premature to attribute the deviation of T^3 behavior to any particular effect, since it requires further investigation.

In summary, we have presented thermal conductivity measurements of individual 22, 37, 56, and 115 nm diam single crystalline Si nanowires. The thermal conductivity observed is much lower than the bulk value, suggesting that phonon–boundary scattering controls thermal transport in Si nanowires. At low temperature, the thermal conductivity of the 22 nm diam wire significantly deviates from Debye T^3 law, suggesting that, on this scale, effects other than phonon–boundary scattering, possibly changes in phonon dispersion due to confinement, may have appreciable effects.

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