A new look at thermal properties of nanotubes

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The thermal conductance of nanotubes is examined over a wide temperature range and as a function of mechanical manipulation. It is found that carbon nanotubes maintain impressive thermal conductance up to their decomposition temperature of ~3200 K. For both carbon and boron nitride multiwalled nanotubes the thermal conductance is relatively independent of tube bending angle, but it can be substantially tuned by "telescoping" the tubes.

1   Introduction

Experimental and theoretical studies have demonstrated that both carbon and boron nitride nanotubes have exceptionally high thermal conductance (K) near room temperature. K decreases with decreasing temperature, eventually becoming linear in temperature at low temperature as expected for a 1-D conductor with quantized thermal conductance [1]. Isotope effects on K are large, particularly for boron nitride nanotubes [2]. Relatively unexplored are the high temperature behavior of K and the dependence of K on mechanical nanotube deformation. We here describe K under thermal and geometrical deformation extremes.

2   High temperature behavior

Established methods for determining the thermal conductance of nanotubes fail at high temperature. We have used a new method to examine the thermal properties of multiwall carbon nanotubes from room temperature to well over 3000 K. The method is described in detail elsewhere in this volume, but briefly, it consists of an electrically-contacted (2-probe) nanotube resting on a thin, TEM-electron-transparent membrane. The nanotube and membrane are sprinkled with gold nanocrystal thermometers. The nanotube is self-Joule heated within the TEM and the nanocrystal thermometers establish a thermal profile over the membrane, which can be analyzed to extract K parameters for the nanotube. Above room temperature, K is well described by

$$K(T) = 1/(aT + bT^2) \,, \tag{1}$$

where the first term in the denominator on the right represents two-phonon Umklapp scattering and the second term represents 3-phonon processes. For a typical nanotube representative of several measured, at

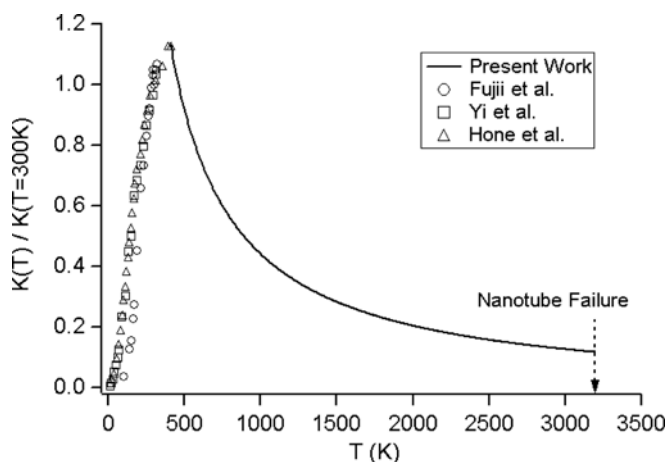* Corresponding author: e-mail: azettl@physics.berkeley.edu

Fig. 1 Temperature dependence of the thermal conductance K of carbon nanotubes. Data below 300 K are from the literature.

1275 K, $K = 150$ W/m with $a = 4.8 \times 10^{-6}$ m/W and $b = 4.3 \times 10^{-10}$ m/WK, as obtained from a fit of Eq. (1) to the experimental data. The contribution of 3-phonon scattering processes becomes non-negligible at temperatures exceeding 1100 K. At 1000 K the nanotube retains 50% of its peak thermal conductance and at $T = 3000$ K it retains approximately 10% of its peak thermal conductance.

Figure 1 shows the full temperature dependence of K for carbon nanotubes. Data for temperatures below room temperature to 410 K have been extracted from the Refs. [4–6], while the high temperature curve is a fit to Eq. (1) to membrane experiments using nanocrystal thermometers. Low temperature data are normalized at 300 K and our fit, valid at high temperature, scaled to extend the previously reported results.

3   Dependence of K on mechanical deformation

It is expected that nanotube transport coefficients, including thermal conductance, be sensitive to mechanical strain. Electronic-band-structure-dependent coefficients, such as the electrical conductivity σ and thermoelectric power S (reflecting the energy derivative of the electrical conductivity), should be particularly vulnerable. This is because the local density of electronic states is highly strain dependent for nanotubes. On the other hand, if the major contributor to thermal conductance is phonons, a more robust transport channel may exist. To test this, we have mounted individual carbon and boron nitride nanotubes on a fixture that allows the nanotube to be mechanically manipulated during measurement. For multiwall carbon nanotubes, the electrical resistance $R(= 1/\sigma)$, S, and K have been measured simultaneously at room temperature as an axial load was applied to the nanotube. With sufficient load, the nanotube first kinks and then begins to fold over onto itself. Figure 2 shows a series of SEM images showing the nanotube cycled through various stages of deformation. Also shown in Fig. 2 are the simultaneously measured normalized coefficients R, S, and K. While R and S faithfully track the deformation (it peaks in frames 7 and 16), suggesting the use of nanotubes as strain gauges or motion sensors, the thermal conductance K is immune to mechanical deformation. Similar robustness for K is observed for boron nitride nanotubes. Indeed, the thermal transport in nanotubes is so robust that they might be termed "phonon waveguides".

4   Tuning the nanotube thermal conductance

Is there a way to manipulate or "tune" the thermal conductance of nanotubes? Certainly, if a nanotube is physically severed, its thermal conductance drops from a high value to zero. We have seen above that less drastic mechanical manipulations, such as bending and kinking, have little effect on K. Is there a happy middle ground?
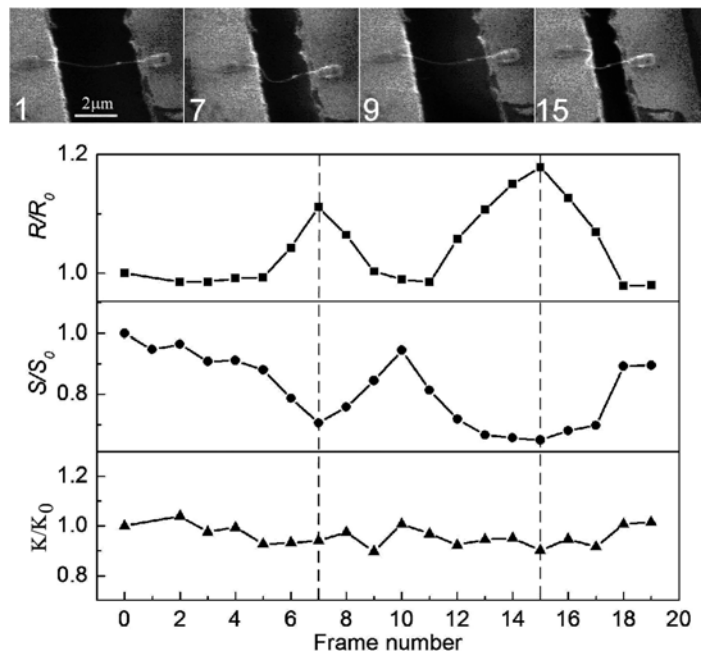
Fig. 2 SEM images of a carbon nanotube undergoing severe mechanical deformation (top series). The simultaneously determined normalized electrical resistance R, thermoelectric power S, and thermal conductance K are also shown. In sharp contrast to R and S, K is relatively immune to mechanical deformations.

Previous experiments [3] on the electrical conductance of "telescoped" nanotubes have shown a remarkable exponential dependence of electrical resistance R on telescopic extension. We have explored analogous thermal conductance experiments. Multiwall carbon nanotubes were telescoped out while measuring K. We find that K is sensitive to telescoping distance x. Indeed, the resulting functional form of the thermal resistance, R_{therm}, follows precisely that found for the electrical resistance,

$$K^{-1}(x) = R_{therm}(x) = R_{0,therm} \exp[2x/D_p] ,$$ (2)

where $R_{0,therm}$ is a constant and D_p is a characteristic length. For carbon nanotubes we find phonon scattering parameter $D_p = 74$ nm (interestingly, the associated characteristic length for the electrical resistance, tied to electron scattering through the same interface, is $1000 - 1500$ nm).

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