Comment on “Divergent and Ultrahigh Thermal Conductivity in Millimeter-Long Nanotubes”

Lee *et al.* [1] report a continuously divergent length dependence of thermal conductivity in 2-μm-to-1-mm-long single-walled carbon nanotubes (SWCNTs) even for defected samples. The thermal conductivity of an ~1-mm-long SWCNT was measured to be as high as ~13 000 W/mK at room temperature. In the following, we prove that their surprising results are not reliable, because their heat conduction model is far from rigorous. Indeed, it is radiation heat loss that causes significant overestimation of thermal conductivity in ultralong samples.

Figure 1(a) illustrates Lee and co-workers’ experimental scheme and heat conduction model, where an individual SWCNT is connected to four suspended resistive thermometers (RT1–RT4) and an electrical current is applied in RT1 to generate Joule heating. Lee and co-workers regarded a parabolic temperature profile in RT1 that is independent of the properties of the CNT, which actually indicates no heat input from the heater to the CNT according to Fourier’s law. Lee *et al.* further argued that radiation heat loss led to underestimation of the heat flux measured by RT2–RT4, and thus the true thermal conductivity of the CNT segment between J1 and J2 should be even higher than the measured value.

Indeed, the heater RT1 should have a *sectionally* parabolic temperature profile [2–6] that is determined by the thermal resistance of the SWCNT and radiation heat loss [Fig. 1(b)]. Radiation heat loss from the CNT surface lowers ΔT_{J1}, which accounts for overestimation of the thermal conductivity. In the end, what can be measured is the apparent thermal resistance between J1 and J2, which is definitely smaller than the thermal conduction resistance of the CNT owing to radiation. Especially for the millimeter-long and 2-nm-diameter SWCNT sample, its conduction resistance is extremely large, while the radiation heat loss is enhanced with the increased surface area; thus, radiation plays a much

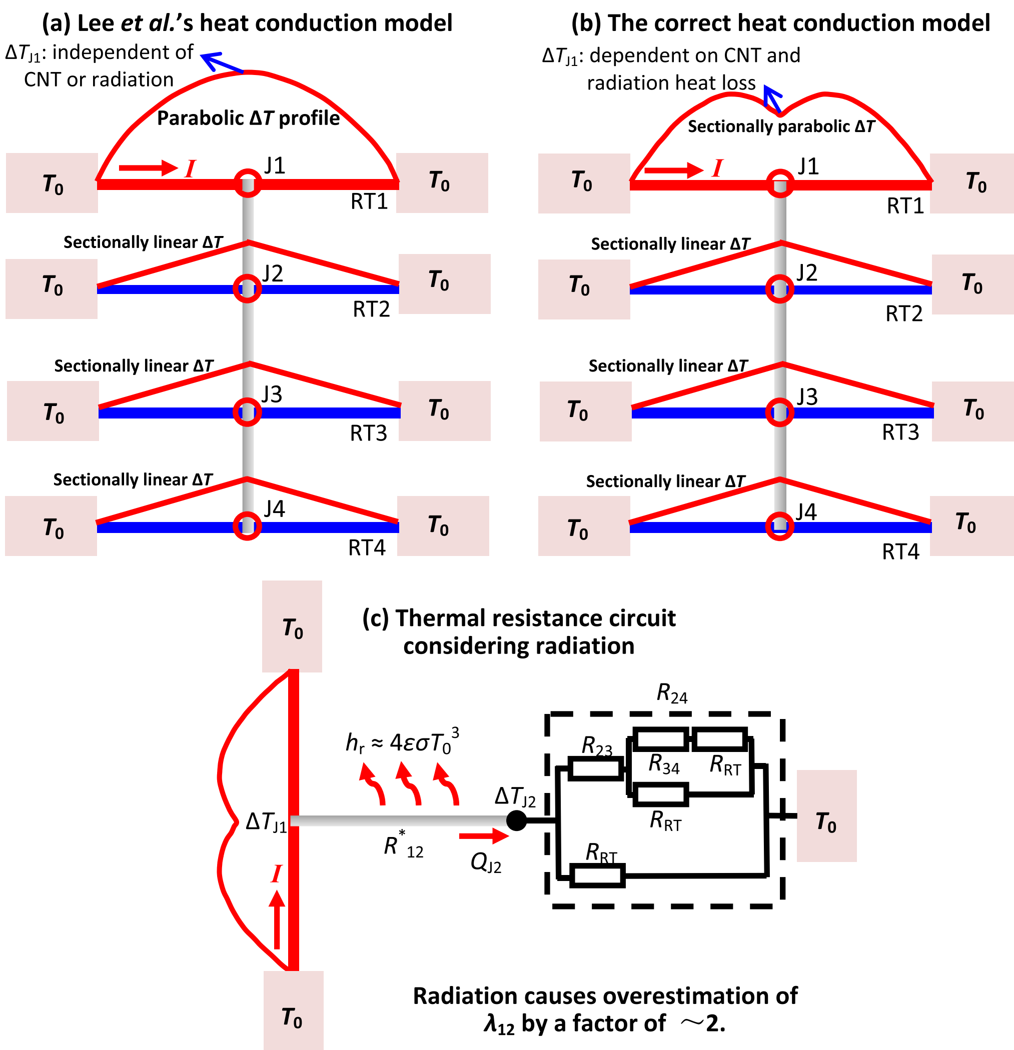

FIG. 1. Heat conduction models.

more pronounced role in overestimating the CNT's thermal conductivity. The apparent thermal resistance considering radiation R_{app} can be approximated by the classical fin model, giving $R_{app} \approx R_{12} \tanh(l_{12}m)/(l_{12}m)$, where $R_{12} = l_{12}/(\lambda_{12}A_{12})$ is the conduction resistance of the CNT and $m = [h_r C_{12}/(\lambda_{12}A_{12})]^{1/2}$, C_{12} being the circumference of the CNT's cross section. Taking λ_{12} as 4000 W/mK, the apparent thermal resistance is calculated to be 46.2% of the conduction resistance for a 1-mm-long SWCNT at 300 K. In other words, the measured thermal conductivity is 2.16 times the true value owing to radiation. We can also rigorously solve differential equations to evaluate the radiation effect [Fig. 1(c)], and the ratio of the measured thermal conductivity to the true value, $\lambda_{12,\mathrm{exp}}/\lambda_{12,\mathrm{true}}$, is deduced to be

$$\frac{\lambda_{12,\mathrm{exp}}}{\lambda_{12,\mathrm{true}}} = \frac{Q_{\mathrm{J2}} l_{12}}{\lambda_{12,\mathrm{true}} A_{12} (\Delta T^*_{\mathrm{J1}} - \Delta T_{\mathrm{J2}})} = \frac{R_{24}}{l_{12}/(\lambda_{12,\mathrm{true}} A_{12})} \left\{ \left(1 + \frac{R_{\mathrm{b1}}}{4R_{\mathrm{tot}}}\right) \times \left[\frac{\sinh(m l_{12})}{R_{24}\sqrt{h_{\mathrm{r}} C_{12} \lambda_{12,\mathrm{true}} A_{12}}} + \cosh(m l_{12}) \right] - 1 \right\}, \quad (1)$$

where ΔT^*_{J1} is the heater's midpoint temperature rise adopted by Lee *et al.* If $\lambda_{12,\mathrm{true}} = 4000$ W/mK, the measured value based on Lee *et al.*'s model should be 7674 W/mK, 91.9% higher than the true value in the 1-mm-long SWCNT.

In summary, the divergent and ultrahigh thermal conductivity data in Ref. [1] has to be taken with care. It turns out that radiation heat loss lowers the apparent thermal resistance and accounts for overestimation of the thermal conductivity by ~2 times in millimeter-long samples.

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