

## Comment on “Length-dependant thermal conductivity of an individual single-wall carbon nanotube” [Appl. Phys. Lett. 91, 123119 (2007)]

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In a recent letter,<sup>1</sup> Wang *et al.* reported the measurement of length-dependant thermal conductivity ( $\lambda$ ) of individual single-wall carbon nanotubes (SWCNTs) using a  $3\omega$  method. Their results appear to agree with a theoretical calculation.<sup>2</sup>

However, a number of questions need to be clarified before the results of Wang *et al.* can be validated. Based on Eq. (3) of the letter, the obtained  $3\omega$  signal ( $U_{3\omega}$ ) depends on both  $\lambda$  and the line contact thermal resistance ( $R_c$ ) between the SWCNT and the substrate. It is unclear how  $R_c$  was estimated before  $\lambda$  was obtained from Eq. (3). Measuring or calculating  $R_c$  between a SWCNT or nanowire and the substrate has been a challenging topic.<sup>3–7</sup> The lack of knowledge of the contact area and the difficulty in modeling phonon transport through the complex nanometer scale contact make it inaccurate to calculate  $R_c$  based on Fourier’s law of heat conduction. In addition, Eqs. (1) and (3) in the letter of Wang *et al.* appear to contain a mistake, i.e., the  $L/\pi^2 R_c$  term in both equations should be corrected to be  $\pi^2 L/R_c$ . These two equations were obtained by retaining only the first term of a series expansion. Truncating the higher order terms leads to a relative error close to 1/3 for a SWCNT on a substrate when heat dissipation across the line contact between the SWCNT and the substrate becomes dominant; whereas the error is only 1/81 for suspended samples.<sup>8</sup> Moreover, the data analysis of Wang *et al.* in Eqs. (1)–(3) assumes diffusive phonon transport and becomes inaccurate if transport of the long-wavelength phonons inside the SWCNT is ballistic, which is the origin of the length-dependant  $\lambda$ .<sup>2</sup> The assumption of diffusive phonon transport in Wang *et al.*’s data analysis appears to contradict their finding of length-dependant  $\lambda$  or essentially ballistic transport of long wavelength phonons.

Moreover, there is a fundamental question regarding the suggestion of no contact thermal resistance at the two ends of the SWCNTs of Wang *et al.* For either a nanometer scale point or line contact or a nanoparticle such as a SWCNT segment embedded in a medium, the constriction thermal resistance can be rather high because of the ballistic resistance component.<sup>3,9–11</sup> In addition, phonon transmission coefficient at the interface between the SWCNT and the SiO<sub>2</sub> substrate or the metal contact should be well below 1 because of the large acoustic impedance mismatch, further increasing the contact thermal resistance. In fact, scanning thermal microscopy measurements have shown elevated temperature rises in nanotubes near the two metal contacts,<sup>12,13</sup> suggesting appreciable contact thermal resistance or contact heating. As the intrinsic thermal resistance becomes smaller in the shorter SWCNT, the influence of the

contact thermal resistance becomes larger. This effect can result in the finding of a  $\sim 22\%$  lower thermal conductivity in the shortest SWCNT by Wang *et al.*

Wang *et al.* contacted the SWCNTs using focused ion beam (FIB) assisted metal deposition, which could spread a conducting layer between the two very closely placed contact electrodes to their shortest SWCNT sample.<sup>14</sup> In addition, extrapolating the electrical resistance versus length relationship for the two longer SWCNTs to zero length, one can obtain a contact resistance of about 15 k $\Omega$  which is comparable to the resistance of the shortest SWCNT sample. The ballistic electrical resistance of a SWCNT with two one-dimensional electron subbands is about 6.5 k $\Omega$ ,<sup>15</sup> which is the minimum achievable contact electrical resistance to the SWCNT. The contact electrical resistance to a SWCNT cannot be eliminated using a four-terminal setup unless the two middle voltage probes are weakly coupled to the SWCNT and are noninvasive,<sup>16,17</sup> which is unlikely the case for the metal contact deposited using a highly energetic FIB on the SWCNTs of Wang *et al.* Hence, the contact electrical resistance for the shortest SWCNT might be much larger than the 10% value suggested in the letter of Wang *et al.* Either an appreciable contact electrical resistance or the aforementioned FIB spreading could result in their finding of 22% lower thermal conductivity in the shortest SWCNT than in the two longer samples.

There is also a subtle question regarding whether a SWCNT can be used as an accurate resistance thermometer to measure its lattice temperature rise during Joule heating. Electron transport in SWCNTs has been known to be highly nonlinear and characterized by ballistic transport in short SWCNTs at low electric field and optical phonon emission at high electric field.<sup>15,18,19</sup> If the nonlinear current-voltage ( $I$ - $V$ ) characteristic of a SWCNT is fitted using a polynomial, the obtained  $V$  expression can contain a  $I^3$  term caused not only simply by an increased lattice temperature, but also by other nonlinear processes including optical phonon emission. Consequently, the  $3\omega$  voltage  $U_{3\omega}$  is not entirely caused simply by lattice temperature rise, especially at high field or in short SWCNTs where nonequilibrium temperatures of electrons, acoustic phonons, and optical phonons need to be taken into account.<sup>19</sup>

In their letter, Wang *et al.* used a small current in order to validate the relationship  $U_{3\omega} \propto I^3$  expected for self heating of the lattice, and in effect, to avoid other nonlinear processes such as optical phonon emission that occur at high field. Based on their Figs. 3 and 4, the applied peak voltage was slightly smaller than the 160 mV value needed for emission of the dominant 1300 cm<sup>-1</sup> optical phonon mode in

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SWCNTs.<sup>18</sup> However, optical or zone boundary phonon modes with lower energy exist in SWCNTs. Moreover, non-equilibrium could also be caused by ballistic electron transport in short SWCNTs at low field. In fact, the reported exponent of the current was 2.723, 2.89, and 2.908 when the length was 0.509, 4.919, and 6.941  $\mu\text{m}$  long, showing an increased deviation from the expected value of 3 with decreasing SWCNT length. This trend raises a question whether some nonequilibrium processes have occurred in the shortest SWCNT, so that  $U_{3\omega}$  was not simply caused by lattice temperature rise.

There are two additional minor issues in the letter by Wang *et al.* First, the spurious signals at 50 and 100 Hz in Fig. 2 of the letter were likely caused by the first and second harmonics of the power line frequency used in their measurement, instead of heat loss to the substrate suggested in the letter. Moreover, the  $\lambda d$  product in their Fig. 5 is proportional to the thermal conductance-length product when the SWCNT cross section is calculated according to  $S = \pi d \delta$ , where  $d$  is the SWCNT diameter and  $\delta$  is the interlayer spacing of graphite. On the other hand, the  $\lambda d$  product does not seem to represent any meaningful physical property when  $S$  is obtained as  $S = \pi d^2 / 4$ .

These questions on the measurement method of Wang *et al.* raise doubts about their claim of  $\pm 10.5\%$  measurement uncertainty. Given the many issues raised here on the experiment, their finding of  $\sim 22\%$  lower thermal conductivity in the shortest SWCNT sample than in the two other longer samples could be either well within the actual uncertainty of their measurement or caused by one of the possible errors discussed above.

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- <sup>1</sup>Z. L. Wang, D. W. Tang, X. B. Li, X. H. Zheng, W. G. Zhang, L. X. Zheng, Y. T. T. Zhu, A. Z. Jin, H. F. Yang, and C. Z. Gu, *Appl. Phys. Lett.* **91**, 123119 (2007).
- <sup>2</sup>N. Mingo and D. A. Broido, *Nano Lett.* **5**, 1221 (2005).
- <sup>3</sup>R. Prasher, *Nano Lett.* **5**, 2155 (2005).
- <sup>4</sup>H. Maune, H. Y. Chiu, and M. Bockrath, *Appl. Phys. Lett.* **89**, 013109 (2006).
- <sup>5</sup>F. Zhou, J. Szczech, M. T. Pettes, A. L. Moore, S. Jin, and L. Shi, *Nano Lett.* **7**, 1649 (2007).
- <sup>6</sup>C. H. Yu, S. Saha, J. H. Zhou, L. Shi, A. M. Cassell, B. A. Cruden, Q. Ngo, and J. Li, *ASME J. Heat Transfer* **128**, 234 (2006).
- <sup>7</sup>I. K. Hsu, R. Kumar, A. Bushmaker, S. B. Cronin, M. T. Pettes, L. Shi, T. Brintlinger, M. S. Fuhrer, and J. Cumings, *Appl. Phys. Lett.* **92**, 063119 (2008).
- <sup>8</sup>L. Lu, W. Yi, and D. L. Zhang, *Rev. Sci. Instrum.* **72**, 2996 (2001).
- <sup>9</sup>C. H. Yu, L. Shi, Z. Yao, D. Y. Li, and A. Majumdar, *Nano Lett.* **5**, 1842 (2005).
- <sup>10</sup>G. Chen, *ASME J. Heat Transfer* **118**, 539 (1996).
- <sup>11</sup>S. K. Saha and L. Shi, *J. Appl. Phys.* **101**, 074304 (2007).
- <sup>12</sup>L. Shi, S. Plyasunov, A. Bachtold, P. L. McEuen, and A. Majumdar, *Appl. Phys. Lett.* **77**, 4295 (2000).
- <sup>13</sup>J. P. Small, L. Shi, and P. Kim, *Solid State Commun.* **127**, 181 (2003).
- <sup>14</sup>S. B. Cronin, Y. M. Lin, O. Rabin, M. R. Black, J. Y. Ying, M. S. Dresselhaus, P. L. Gai, J. P. Minet, and J. P. Issi, *Nanotechnology* **13**, 653 (2002).
- <sup>15</sup>A. Javey, J. Guo, Q. Wang, M. Lundstrom, and H. J. Dai, *Nature (London)* **424**, 654 (2003).
- <sup>16</sup>S. Datta, *Electronic Transport in Mesoscopic Systems* (Cambridge University Press, New York, 1995).
- <sup>17</sup>M. S. Purewal, B. H. Hong, A. Ravi, B. Chandra, J. Hone, and P. Kim, *Phys. Rev. Lett.* **98**, 186808 (2007).
- <sup>18</sup>Z. Yao, C. L. Kane, and C. Dekker, *Phys. Rev. Lett.* **84**, 2941 (2000).
- <sup>19</sup>E. Pop, D. Mann, J. Cao, Q. Wang, K. Goodson, and H. J. Dai, *Phys. Rev. Lett.* **95**, 155505 (2005).