

Thermal conductivity of suspended single-layer graphene measured by Raman spectroscopy

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Since Balandin et al. first reported an extremely large value for the thermal conductivity of $\sim 5,000 \text{ Wm}^{-1}\text{K}^{-1}$ for single layer graphene [1], the superior thermal properties of graphene has attracted much interest. Several groups since have measured thermal conductivity of mechanically exfoliated [1-3] or chemical vapor deposition (CVD)-grown [4,5] graphene using different methods. For suspended, exfoliated single-layer graphene, the reported values range between 660 and $5,300 \text{ Wm}^{-1}\text{K}^{-1}$ [1,2]. Given the importance of this key parameter for device applications, an accurate measurement is crucial. In this work, we present the measurement of the thermal conductivity for suspended single-layer graphene at temperatures between 300 K and 500 K using Raman spectroscopy on a clean sample prepared directly on a patterned substrate without involving a transfer process.

The substrates with round holes with various diameters were prepared by photolithography and dry etching of Si substrates covered with a 300 nm-thick SiO_2 layer. The depth of the holes is $\sim 1.7 \mu\text{m}$, deep enough to prevent interference from laser light reflected and scattered from the bottom of the holes [6]. The diameters were 2.6, 3.6, 4.6, and $6.6 \mu\text{m}$. The samples were prepared directly on the cleaned substrate by mechanical exfoliation from natural graphite flakes. No chemical treatment of the sample was involved in the preparation process. This ensures that the sample surface is free from chemical contaminants that may affect the measured thermal conductivity values. The sample used was a single-layer graphene flake of $35 \times 60 \mu\text{m}^2$ dimensions identified by the Raman spectrum [7,8]. The 514.5-nm (2.41 eV) beam of an Ar ion laser was focused onto the graphene sample by a $50\times$ microscope objective lens (N.A. 0.8), and the scattered light was collected and collimated by the same objective. The laser spot size was measured using the modified knife-edge method [5], and the beam spot size was approximately $0.29 \mu\text{m}$ in radius. The scattered signal was dispersed with a Jobin-Yvon Triax 550 spectrometer (1800 grooves/mm) and detected with a liquid-nitrogen-cooled charge-coupled-device detector. The spectral resolution was about 0.7 cm^{-1} .

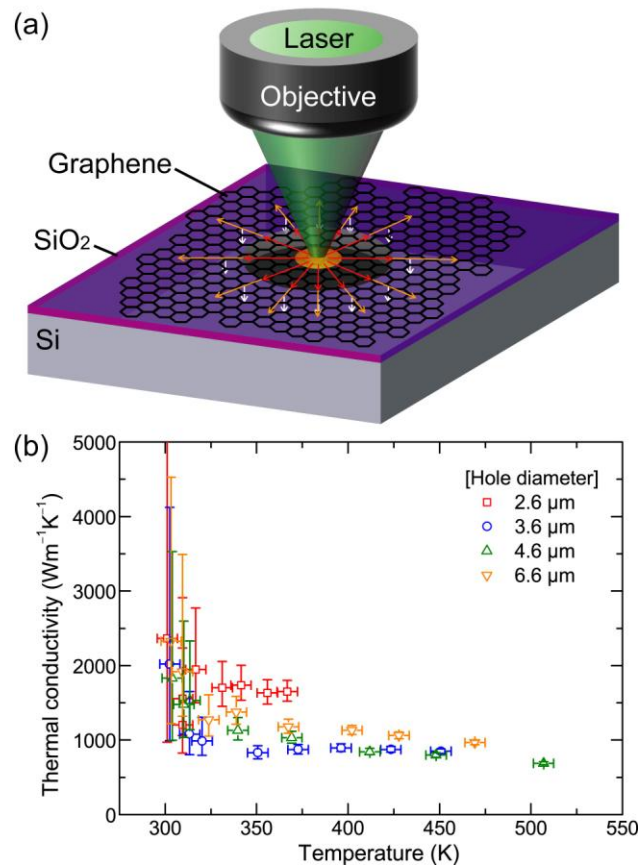
The light absorption induces local heating that raises the temperature in the vicinity of the laser spot. In a steady state, there exists a temperature gradient that depends on the total power supplied by the laser beam, thermal conductivity, and the boundary conditions at the edge of the hole. The local temperature at the laser spot can be estimated from the Raman spectrum. The temperature coefficients of the *G* and *2D* bands for suspended graphene were measured recently by Chen *et al.* [4]. Since the *2D* band is more sensitive to temperature than the *G* band, we used the *2D* band for the estimate of the temperature. As the laser power increases, the *2D* frequency redshifts due to increased heating.

In order to estimate the thermal conductivity, we used the heat diffusion equation ignoring the heat conduction to the ambient air. We considered heat conduction through suspended graphene and graphene on the substrate as well as between graphene and the substrate. The substrate is assumed to be a heat sink at the ambient temperature. From analyses with the heat diffusion equation, we estimated the thermal conductivity of suspended graphene. It decreases as the temperature increases: from $\sim 2000 \text{ Wm}^{-1}\text{K}^{-1}$ near 300 K to $\sim 690 \text{ Wm}^{-1}\text{K}^{-1}$ at 500 K.

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Figures



(a) Schematic diagram of experiment which measured the thermal conductivity of suspended pristine graphene. (b) Estimated the thermal conductivity of graphene as a function of temperature.