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Width dependent intrinsic thermal conductivity of suspended monolayer graphene

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ABSTRACT

Size dependence is one of the most important unique features of thermal conductivity in two-dimensional materials. Suspended single-layer graphene (SLG) provides a perfect platform for studying the size dependent phonon transport. Here we report measurement and theoretical analysis of heat conduction in suspended SLG as a function of width and temperature. The thermal conductivity of graphene was larger for wider SLG. This width effect was smaller at higher temperatures. In suspended SLG, the long wave-length phonons tend to be more scattered at the lateral boundaries of narrow SLG ribbon, in which the mean free path of phonons is close to the sample width. This behavior can be understood as a mode selectivity of phonon-boundary scattering for suspended SLG. The result revealed the unique width dependence of thermal conductivity in suspended SLG and provided useful guidelines for the future SLG-based thermal applications.

1. Introduction

Graphene is a promising two-dimensional (2D) material for distributing waste heat generated in micro-electronic devices due to its ultra-high thermal conductivity [1,2]. Understanding its unique 2D heat transport mechanisms lays the foundation for developing novel graphene-based heat spreaders [1,3]. The size/geometry dependent thermal conductivity is one important feature of 2D materials. Regarding the length effect (heat flows in the length direction), some numerical simulations for 2D lattice systems demonstrate logarithmic divergence of thermal conductivity [4–6]. Recently, this $\lambda - \log L$ relationship has been verified by the experiment of suspended single-layer graphene (SLG), where λ and L are the thermal conductivity and the length of SLG ribbon respectively [7]. The authors suggested that this unique divergent λ came from the 2D nature of graphene, resulting through the combination of reduced dimensionality and displacement of phonon populations at stationary non-equilibrium conditions.

Besides the length effect, the thermal conductivity of graphene ribbon also depends on its width, which is perpendicular to the heat flow direction. For the width effect of suspended graphene, the phonon scattering at lateral boundaries and edge roughness play a dominant role, especially when the mean free path (MFP) of in-plane phonons is comparable to the width of graphene sample. To date, some numerical simulation results have shown that the thermal conductivity of graphene increases with increasing width [8–11]. However, compelling experimental evidence for this width dependence is still missing. A great challenge comes from the difficulty in fabricating suspended SLG micro-electronic devices that make it possible to measure the width-dependent thermal conductivity.

We have successfully fabricated suspended SLG microelectronic devices for measuring the thermal conductivity in our previous work, in which the effect of defect on heat transport in SLG was quantitatively evaluated [12]. In the present work, we report a clear width-dependent thermal conductivity of SLG. The underlying physical mechanism is explained by using a lattice dynamics model. The result is useful for understanding the 2D nature of heat transport in graphene and related thermal applications in the future.

2. Suspended monolayer graphene device and measurement principle

The fabrication method and measurement principle have been introduced in our previous work [12,13]. Different from the normal suspending process of graphene, an effective gas etching technique was employed to create a deep trench (>8 μm) beneath the

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graphene ribbon. The large etching depth ensured that a large area graphene ribbon could be suspended from the substrate. The graphene membrane was sandwiched between a polymer resist layer and SiO_2 layer during the gas etching process. Without contacting with the ambient reactive gas, the pristine graphene lattice structure could be well maintained. In the final step, both polymer and SiO_2 layers were removed by wet etching, leaving a free-standing SLG ribbon above the substrate.

Fig. 1 shows the scanning-electron-microscope (SEM) images of four fabricated SLG ribbon samples. The SLG bridges between a micro-beam sensor and heat sink pad, where the distance between them determines the length of the SLG ribbon. The samples have been fabricated on the same silicon chip following the same electron beam (EB) lithography pattern. Thus, the length was approximately 1.6 μm for all samples. The widths of SLG ribbons were measured using the SEM images and shown in the caption of Fig. 1. The width of a SLG ribbon was smaller at the middle due to the edge deformation. The largest width at the base was therefore considered as the width of SLG. The suspended micro-beam sensor consisted of a 100-nm thick Au film and an adhesion layer of 10-nm thick Cr. The typical dimensions of the sensor were about 10 μm in length and 1 μm in width.

The T-type sensor method is an effective way of measuring the thermal conductivity due to its simplicity in principle and good repeatability. It was used for measuring the thermal conductivity of SLG ribbon [12]. The sensor and SLG ribbon form a "T" shape structure. The SLG ribbon works as an additional channel for heat transport from the sensor to the heat sink. Under the same Joule heating power condition, the temperature of sensor decreases after connecting with the SLG at the middle. By measuring the average temperature difference of sensor with and without the SLG ribbon, the thermal conductivity of SLG can be calculated. The experiment was conducted through the following *in situ* measurement procedure to ensure a high accuracy. Firstly, the micro-beam sensor connected with a SLG ribbon was placed on a Peltier heating/cooling stage in an FEI Versa 3D™ dual-beam system. The average temperature of the sensor was measured under different ambient temperature and heating power conditions. Then a focused ion beam was used to cut the SLG ribbon. The average temperature of the bare sensor was measured again under the same conditions. Based on a 2D thermal analysis model, the thermal conductivity of SLG was calculated.

Fig. 2 shows the temperature distribution of sensor calculated by using a commercial finite-element software, COMSOL Multiphysics™. In the thermal analysis model, the sample dimensions, thermal conductivity of sensor and Joule heating power were given as known parameters. The thermal conductivity of SLG was the only unknown parameter, which was determined by comparing the simulated temperature difference between Fig. 2(a) and (b) with the experimental data. Considering the uncertainties from the geometric dimensions, resistance measurement, temperature fluctuation of Peltier stage and thermal analysis model, the final measurement uncertainty was estimated to be about 10%.

3. Theoretical model

Lattice dynamics theory was used to predict the thermal conductivity of SLG [14]:

$$\lambda = \frac{1}{4\pi k_B T^2 \delta} \sum_{s=\substack{\mathrm{TA,LA,ZA,}\\ \mathrm{TO,LO,ZO}}} \int_{q_{\min}}^{q_{\max}} \left\{ [\hbar\omega_s(q) v_s(q)]^2 \times \tau_s(q) \frac{\exp[\hbar\omega_s(q)/k_B T]}{(\exp[\hbar\omega_s(q)/k_B T] - 1)^2} q \right\} dq, \quad (1)$$

where λ, k_B, $\hbar$, ω_s, τ_s, q and T are the thermal conductivity, Boltzmann constant, reduced Planck constant, phonon frequency, relaxation time, wave vector and temperature, respectively. $\delta = 0.35$ nm

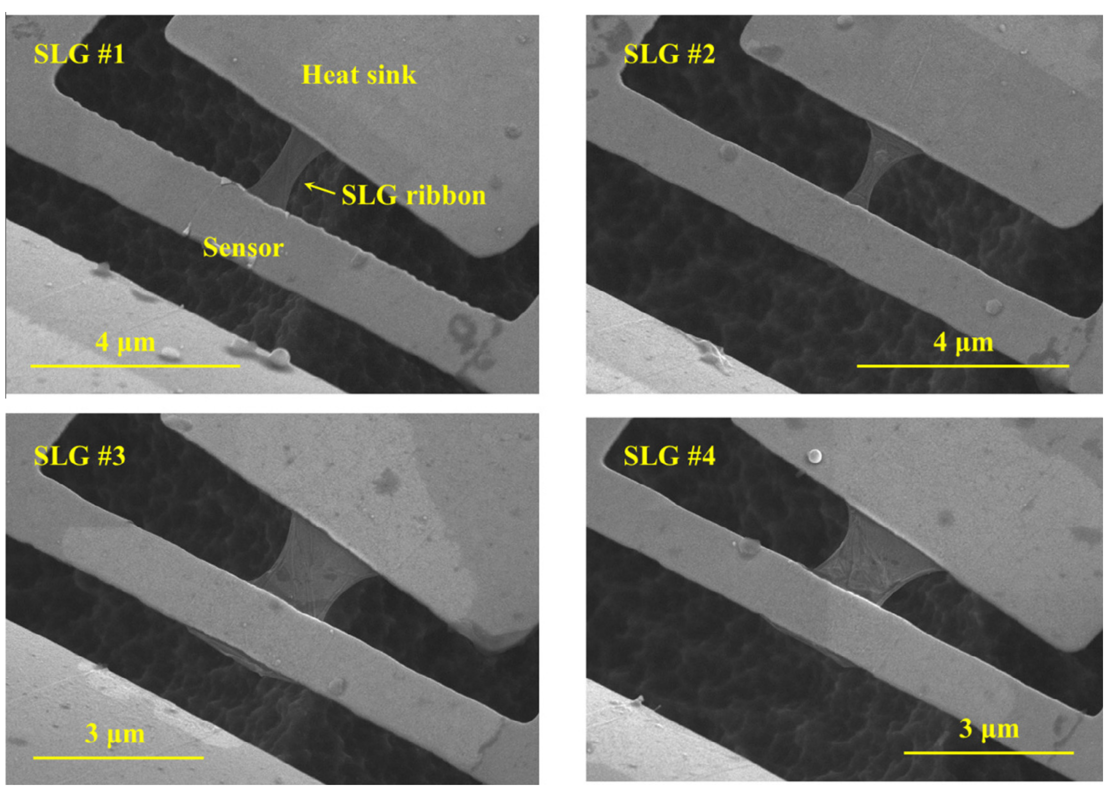

Fig. 1. SEM images of four SLG samples. Samples #1 and #2 are narrow ribbons, whose widths are 0.85 μm and 1.22 μm, respectively. Samples #3 and #4 are wide ribbons, whose widths are 1.92 μm and 2.08 μm, respectively. The SLG sample is suspended between a micro-beam sensor and heat sink pad. The sensor serves as both Joule heater and precise resistance thermometer.