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Thermal transport behaviors of suspended graphene sheets with different sizes

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We experimentally investigated the thermal transport behaviors of suspended single-layer graphene (SLG) and graphene sheets with different sizes. The length of graphene sheet suspended on a groove of SiO_2/Si substrate is equal to the groove width (508 μm). Laser cutting technology is used to tailor SLG to obtain graphene sheets with different widths. A one-dimensional steady-state method is applied to measure the thermal conductivity, k, of graphene sheet and SLG at different conditions. k of the graphene sheets with different widths is much higher than that of SLG and increases with the decrease of graphene sheet width, and the increase in k appears significant when the width decreases greatly. We proposed that the Umklapp scattering may become dominant in phonon transport with the increasing of graphene sheet width, resulting in the thermal conductivity decrease. Another anomalous behavior is that k increases firstly and then decreases with an increase in temperature, and has a peak value at around 50 °C. The competition effects of boundary scattering, point defect scattering, grain-boundary phonon scattering, and Umklapp scattering in phonon transport may contribute to this phenomenon. A maximum k value of 2450.55 W m^{-1} K^{-1} is obtained under the condition of 50 °C when the SLG is cut for the fourth times. The electrical conductivity of graphene sheets with different widths at room temperature is in the range of $0.7584–2.7002 \times 10^6$ Ω^{-1} m^{-1}. The graphene sheet with small width exhibits better thermal and electronic properties that make them attractive for the fabrication of nanoscale electronics and promising for the application in thermal management.

1. Introduction

Materials with high thermal conductivity are needed for thermal management due to the developments in miniaturizing electronic devices. Among these materials, graphene, a two-dimensional crystal consisting of a single atomic layer of carbon, has been demonstrated by Mingo et al. [1] to have higher thermal conductivity than carbon nanotube. In their works the lattice dynamics method was applied. The same method was applied by Shi et al. [2] and the analytical results are that the thermal conductivity of graphene with different widths and defect densities show different temperature dependence. Nika et al. [3] make use of valence-force field method to calculate thermal conductivity of graphene and the resulted value is between 2000 and 5000 W/(m K). They also found that the thermal conductivity value of graphene is dependent on the width, concentration of defects, and edge rough degree of graphene. Zhang et al. [4] have employed the classical non-equilibrium molecular dynamics (NEMD) method to investigate the thermal conductivity of sawtooth-like graphene nanoribbons (GNRs). It is found that the thermal conductivity of these GNRs is nearly independent on the length, while it is very sensitive to the width. Hu et al. [5] used the similar technique to investigate the effect of edge-passivation by hydrogen and isotope mixture on the thermal conductivity of rectangular GNRs. The results revealed that the thermal conductivity was considerably reduced by the edge H-passivation and the isotope mixing could reduce the thermal conductivities. This NEMD was also employed to explore the thermal conductivity of both zigzag and armchair GNRs possessing different densities of Stone–Thrower–Wales defects, the thermal conductivity of hydrogenated or fluorinated graphene, and the effect of boron or nitrogen atom substitution on the thermal conductivity of graphene [6–11]. Verma et al. has addressed a physics-based closed-form analytical model of flexural phonon-dependent diffusive thermal conductivity of suspended rectangular single layer GNR [12]. Theoretical studies suggested that graphene have unusually high thermal conductivity [13–15],

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and many theoretical methods were developed to study the thermal conductivity of different kinds of graphene. Several experimental measurements were also reviewed and discussed in our previous report [16]. Due to more or less deficiency existing in the reported experimental measurements, it is necessary to apply a new method to study the graphene thermal conductivity. Recently, we have reported an effective and reliable method to measure the temperature dependent in-plane thermal conductivity of a suspended and long GNR [16]. Compared with other reported studies, in which the confocal micro-Raman spectroscopy was applied [17,18], or graphene was deposited on SiO_2 substrate [19], or graphene was encased between SiO_2 layers [20], our reported method avoid the great errors resulted from different assumptions on the efficiency value of the graphene's optical absorbance and the influence caused by a substrate.

In theoretical studies, size dependence of thermal conductivity of GNRs has been observed [21–25]. Other size dependent thermal conductivity of low dimensional materials such as carbon nanotubes [26,27] and silicon nanowires [28] was also studied by using molecular dynamics simulations. GNR, the narrow layers of graphene, is similar to the cases of nanotubes and nanowires, and its one dimensional nano-structures are very promising platforms to verify fundamental thermal transport theories [22]. A very recent study on the simulation and experiment of the size and temperature dependent thermal conductivity of suspended GNRs has been reported by Xu et al. [29] They firstly obtained from experimental measurement the length dependent thermal conductivity of suspended GNRs in the range from 300 nm to 9 μm and found that the experiments were in excellent agreement with the simulations. To verify the theoretical results on size dependence of thermal conductivity of GNRs, more experimental observations are needed. In this study, we applied our previous reported method to measure the thermal conductivity of suspended graphene sheets with different sizes, and the effect of temperatures on the thermal conductivity is also investigated.

2. Experimental

2.1. Preparation of measurements

The samples were prepared by suspending single-layer graphene (SLG) film on an insulated groove at the middle position of a SiO_2/Si substrate with gold coating film. The details about the preparation have been described in our previous report [16], in which the transfer for graphene was according to the Ref. [30]. In brief, firstly, Cu foil supporting graphene film with polymethylmethacrylate (PMMA) protection layer was etched in an aqueous solution of iron trichloride and hydrochloric. Secondly, the obtained graphene film with PMMA protection layer was transferred into water. Thirdly, an insulated groove (see Fig. 1(b)) was fabricated at the middle position of SiO_2/Si substrate with gold coating film. The SiO_2/Si substrate was brought to contact with the graphene film to 'pull' it from the water. The graphene film with PMMA protection layer was suspended on the groove. The PMMA aiding to the transfer of graphene was further removed with acetone. The edge of the suspended graphene film was rough. Laser cutting technology was applied to tailor the suspended graphene film, and after being further cut for several times the suspended graphene sheets with different size were obtained. Fig. 1(a) illustrates the schematic diagram of a fabricated sample structure. Four copper wires were fixed on the gold film at four different orientations. A schematic figure illustrating the Si wafer with Au electrodes can be seen in our previous report [16]. The fabricated sample with measurement circuit was installed on an isothermal plate as can be seen in Fig. 1(b). The isothermal plate with fabricated sample was placed in a sample cavity. The experimental temperatures can be controlled in a temperature range of 77–500 K, and the sample cavity temperature can be adjusted by controlling the liquid nitrogen flow and electrical heating power. A high vacuum level of ~3×10^{-6} Torr was applied through all the measurements process to suppress residual gas conduction and convective thermal losses. The measuring instruments include constant current source, standard resistance, and two high-accuracy voltmeters. The temperature difference between the experimental graphene sheet and ambience was controlled within 10 K, so the radiation heat losses could be ignored.

2.2. Graphene sheets with different sizes

Fig. 2 shows the SEM images of SLG film and graphene sheet suspended on the groove. The width of groove is 508 um, which can be taken as the length of the suspended SLG film and graphene sheet. It can be seen from Fig. 2(a) that the edge of the suspended SLG film is rough, being highlighted by the white curves, and is not perpendicular to the boundary line of groove. It is apparent that the width of the suspended SLG film is bigger than its length. After measurements the suspended SLG film was cut for the first time by using laser radiation and a suspended SLG film with straight edge was obtained. The edge is almost perpendicular to the boundary line of groove. After being cut, the width of the suspended SLG film is 385um, which is smaller than its length. We can consider that the suspended SLG film is tailored into graphene sheet which is shown in Fig. 2(b). The suspended SLG film was cut for the second time and the image of the resulted film is shown in Fig. 2(c). The width of the obtained SLG film is 163–176um. It is obvious that the graphene sheet is not standard rectangle because no coordinate can be used in cutting the graphene sheet. The difference from eye-observation made it difficult to keep graphene sheet to be standard rectangle. The widths are 118–128 μm and 43–50 μm after the graphene

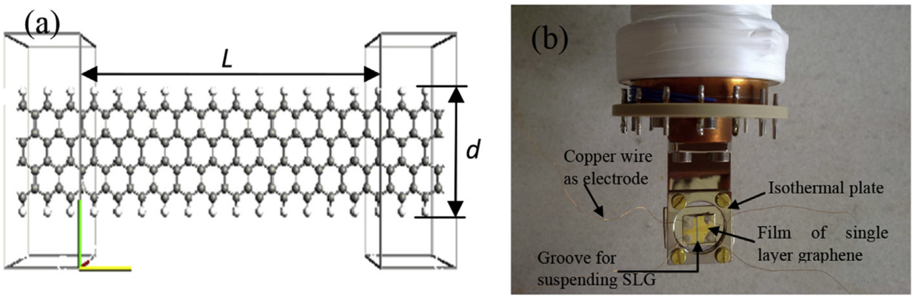

Fig. 1. Schematic diagram of a graphene sheet (a) and digital photo of a chip with the measurement circuit (b).