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Study on temperature-dependent carrier transport for bilayer graphene

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HIGHLIGHTS

- The bilayer graphenes were synthesized by chemical vapor deposition with C_2H_2.
- Temperature-dependent carrier transport for bilayer graphene was investigated.
- The carrier density of the bilayer graphene followed to thermal activation model.
- The carrier mobility was temperature-independent for the bilayer graphene.

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ABSTRACT

In order to investigate the temperature-dependent carrier transport property of the bilayer graphene, graphene films were synthesized on Cu foils by a home-built chemical vapor deposition (CVD) with C_2H_2. Samples regularity, transmittance (T) and layer number were analyzed by transmission electron microscope (TEM) images, transmittance spectra and Raman spectra. Van Der Pauw method was used for resistivity measurements and Hall measurements at different temperatures. The results indicated that the sheet resistance (R_s), carrier density (n), and mobility (μ) were 1096.20 Ω/sq, 0.75×10^{12} cm^{-2}, and 7579.66 cm^2 V^{-1} s^{-1} at room temperature, respectively. When the temperature increased from 0 °C to 240 °C, carrier density (n) increased from 0.66×10^{12} cm^{-2} to 1.55×10^{12} cm^{-2}, sheet resistance (R_s) decreased from 1215.55 Ω/sq to 560.77 Ω/sq, and mobility (μ) oscillated around a constant value 7773.99 cm^2 V^{-1} s^{-1}. The decrease of the sheet resistance (R_s) indicated that the conductive capability of the bilayer graphene film increased with the temperature. The significant cause of the increase of carrier density (n) was the thermal activation of carriers from defects and unconscious doping states. Because the main influence on the carrier mobility (μ) was the lattice defect scattering and a small amount of impurity scattering, the carrier mobility (μ) was temperature-independent for the bilayer graphene.

1. Introduction

Transparent conducting films (TCFs) have been widely applied in many fields such as flat panel display, thin film solar cells, and gas sensors [1–5]. Recently, tin-doped indium oxide (ITO) had a wide range of commercial applications as transparent conducting oxides because of its unique optical (~85% in visible region) and electrical ($\sim1\times10^{-4}$ Ω cm) properties [6]. However, as a rare metal, indium was expensive, and the preparation cost of ITO was also high. Moreover, ITO was so fragile that it could not satisfy some new applications, such as flexible liquid crystal display and organic solar cells. Graphene as a special two-dimensional material had several attractive properties such as high transparency (~98% in visible region for single-layer graphene), high carrier mobility (~200,000 cm^2 V^{-1} s^{-1} theoretically) [7,8], excellent flexibility and good chemical stability. Therefore, graphene since its discovery in 2004 was regarded as a promising alternative material for ITO as TCFs [9–12]. Graphene film had attracted much attention as TCFs also due to its low cost of raw material and relative ease of synthesis. The two most important performance metrics for TCFs were the transmittance (T) and the sheet resistance (R_s) [13]. Meanwhile, T and R_s were determined by the carrier density (n) and mobility (μ). The measurement of the carrier transport properties of the graphene has become the hot spot.

In Bult's work, the sheet resistance (R_s) of single-layer graphene

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film they measured by four-point probe was 4262 Ω/sq, and the mobility (μ) that they measured by Hall measurement was 2920 $cm^2 V^{-1} s^{-1}$ [13]. In Jo's work, graphene films were grown on Ni by chemical vapor deposition (CVD), and sheet resistance (R_s) they measured was 620 Ω/sq [14]. In Teng's work, they reported the mobility (μ) of the graphene film was 600 $cm^2 V^{-1} s^{-1}$ [15]. Well, Edwin Kim prepared graphene films on Cu foils, and the mobility (μ) was 1200 $cm^2 V^{-1} s^{-1}$[16]. However, the experimental study on the carrier transport properties of bilayer graphene was rarely reported so far.

Bilayer graphene had quadratic energy dispersion similar to the regular 2D electron gas, but its effective Hamiltonian was chiral without band gap similar to the single-layer graphene [17]. Therefore, bilayer graphene was worthy of being studied due to its unusual electronic structure. Furthermore, for thin film solar cells, gas sensors, etc., they may work in different environmental temperatures. Therefore, the influence of temperature on the carrier transport properties of graphene TCFs was also worthy of being investigated. Some previous work studied the carrier transport properties of bilayer graphene at finite temperatures by theoretical calculation (from 0 K to 300 K) [13,18]. However, there was not enough attention on the carrier transport properties of bilayer graphene at higher temperatures (> 300 K).

In this work, high-quality bilayer graphene films were synthesized by atmospheric pressure CVD (APCVD) on Cu foils with C_2H_2. The layer number, transmittance (T) and crystallinity of graphene films were characterized by Raman spectra, transmittance spectra and transmission electron microscope (TEM) images, respectively. We investigated the sheet resistance (R_s), carrier density (n), and mobility (μ) by the Van Der Pauw method at room temperature [19]. The carrier features of bilayer graphene films under the different temperatures (0 °C (273 K)–240 °C (513 K)) were also explored and discussed.

2. Experimental works

2.1. Synthesis and characterization of graphene

Graphene films were grown on 25 μm Cu foils (Alfa Aesar, 25-μm-thick, square area of ~6 cm^2, 99.8%) in a home-built CVD furnace with a quartz tube. Firstly, Cu foils were cleaned by an ultrasonic cleaner for ~15 min in alcohol and acetone. Then, the Cu foils were loaded into the quartz tube of CVD furnace. The synthesis process could be described as follows: the Cu foils were heated to 1000 °C during 60 min and then annealed for 20 min under the constant flowing of 300 standard-state cubic centimeters per minute (sccm) Ar and 300 sccm H_2. 1 sccm C_2H_2 was introduced as carbon source during the growth process and the grown time was set to 10 min. When the growth was finished, the as-grown sample was cooled down to room temperature under the same H_2 and Ar atmosphere as the growth condition. Finally, the Cu foils were dissolved in the etchant ($Fe(NO_3)_3$:HCl:H_2O = 1 g:1 ml:20 ml) [20].

The obtained graphene samples were characterized by TEM (Tecnai G2 F20 s-Twin) and Raman spectrometer (Laboratory Ram HR800, with laser excitation wavelength at 514 nm). The transmittance spectra of the graphene films in the 400–1100 nm wavelength range were also measured by micro-spectrometer (PG 2000-Pro-Ex).

2.2. Device fabrication and transport measurements

Graphene films were transferred onto a quartz substrate for resistivity measurements and Hall measurements. In order to use the Van Der Pauw method, the graphene film must be symmetrical and without isolated holes [21]. Length and width of the sample were both ~15 mm. Four ohmic contacts (as small as possible) were placed on the boundary of the sample. Conductive adhesive was pressed onto the corners of samples to make good electrical contact between the graphene film and wires. Fig. 1(a) displayed the connection method of the wires.

For resistivity measurements, the current flowed along one edge of the sample (I_{12}) and the voltage across the opposite edge (V_{34}) was measured, the resistance ($R_{12,34}$) could be calculated by using Ohm's law. Another resistance ($R_{23,41}$) could also be found by the same method. The sheet resistance (R_s) of the graphene sample could be determined from two of these resistances ($R_{12,34}$ and $R_{23,41}$).

For Hall measurements, a magnetic field (0.10 T) was applied in the direction of the sample, the voltage was applied between the contacts at diagonally opposite corners (V_{24}) and the current (I_{13}) flowing between them was measured. Fig. 1(b) showed the placement of the sample in the magnetic field. Sheet resistance (R_s), carrier density (n) and mobility (μ) were calculated from the corresponding measured data.

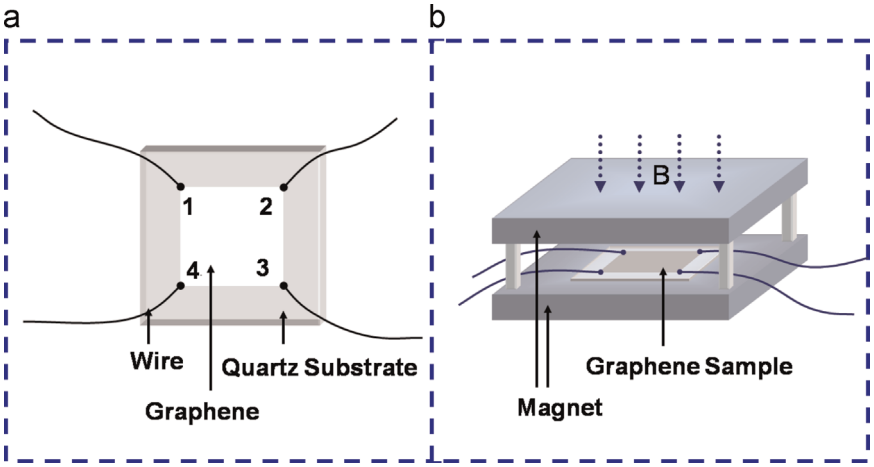

Fig. 1. Schematics of the Hall device. (a) The connection of the wire and the graphene film and (b) placement of the sample in the magnetic field.