Electronic Properties of Large-scale Graphene Films Chemical Vapor Synthesized on Nickel and on Copper

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We report the electronic properties of large scale graphene films grown by chemical vapor synthesis on Ni and Cu, and then transferred to SiO_2/Si substrates. We have observed electric field effect and measured carrier mobilities in the samples to be on the order of $10^3 \text{ cm}^2/\text{Vs}$. We have also measured low temperature magnetoresistance and studied characteristic universal conductance fluctuations on samples grown on Ni.

Introduction

Graphene, single-atom-thick carbon film¹, attracted tremendous interests as a candidate electronic material for "post-silicon" electronics. One of the main challenges of the graphene-based electronics fabrication is the large scale synthesis of high quality graphene. Recently, growth by chemical vapor deposition (CVD) and related methods on various metallic substrates (for example, Nickel^{2,4-8} and Copper^{3,18}) has been demonstrated as a rational synthesis route for producing wafer size ultrathin graphene films. After transferring the synthesized graphene films to other substrates, people have shown these films are promising for various electronic applications, such as flexible and transparent electronics^{19,20}. In this paper, we present the electronic transport properties of graphene films grown on Ni and Cu.

Sample Preparation

The synthesis of graphene film on Ni and Cu substrates was conducted at Univ. of Houston and follows the similar recipes used in our previous research^{2,8,17}. The growth methods are also similar to those used by other groups³⁻⁷ with minor differences. In our case, polycrystalline Ni and Cu foils from Alfa Aesar were cut as 5mm×5 mm pieces. Precursor gases are CH₄ carried by Ar with *ambient* pressure. The carbon decomposition/dissolution time was 5~20 min at 1000 °C. Samples were cooled down by mechanically pushing the sample holder to lower temperature zones in the range of 30~500 °C in Ar atmosphere. Cooling rates were monitored by a thermal couple on the sample holder. Then we transfer the graphene films to SiO₂/Si substrates using acidic solutions for electric property measurements.

A series of Hall bar shaped devices with different lengths (from 200µm to 5µm) have been

fabricated by photolithography (for relatively large devices as shown in Fig. 1b) or e-beam lithography (for relatively small devices as shown in Fig. 3a), with plasma etching and metal deposition (Cr/Au or Ti/Au electrodes).

Electronic Properties of Graphene Films Grown on Ni

Fig. 1a shows electrical resistance of a device ("A") measured at low temperature (T = 6.5 K). We used the highly doped Si as the back gate, and the gate voltage (V_{gate}) is varied between 0 V to 90 V. An ambipolar field effect is evident. Similar field effects are observable up to room temperature, though at low T, a larger range of V_{gate} can be accessed without gate leakage. The field effect is so far only observed in ultrathin 2D graphitic systems, where electronic transport in the field effect is dominated by only few graphene layers⁹⁻¹¹. We have measured similar field effect in multiple samples, with extract carrier mobility ranging from several hundreds to several thousands of cm²/Vs.



Fig. 1. (a) Two-terminal resistance as a function of back gate voltage, measured in sample ("A") at a temperature of 6.5 K., showing the electric field effect. (b) Optical microscope image of a typical field effect transistor (FET) device (top view) fabricated from a large scale transferred film grown on Ni, patterned into the Hall bar shape. (c) Schematic diagram (not to scale) of a FET device.



Fig. 2. (a) Two consecutively measured traces of R_{xx} vs B at 1.5K, showing reproducible fluctuations. (b) Magnetoconductance fluctuations (normalized by e^2/h) (after subtracting a smooth background) at several temperatures. Traces except for the one at

0.3K are vertically shifted for clarity.

At low temperature (down to 300mK), we observed reproducible magnetoconductance fluctuations in a smaller sample "B" (~10 μ m). R_{xx} vs B for two back-to-back sweeps of magnetic field has been plotted in Fig. 2a, which suggests that the fluctuation is intrinsic and can be attributed to the universal conductance fluctuations (UCF). UCF in magnetic field originates from the quantized flux enclosed in the area defined by phase coherent length (L_{ϕ})¹³. UCF has been recently observed in both exfoliated^{12,15,16} and epitaxial (on SiC)¹⁴ graphene.

From the UCF, we can estimate the phase coherence length via $\Delta B \cdot L_{a}^{a} \otimes \phi_{0}$, where

\Delta B is the correlation field, **\beta_0 = \frac{1}{2}** is the flux quantum, *e* is electron charge, *h* is the Planck constant. To quantify the magnetoconductance fluctuation, a smooth background has been subtracted from the original data (Fig. 2b). The values for **\Lambda R** are seen to be roughly 1T at 0.3K and 2.5T at 30K, corresponding to a value of L_{φ} on the order of 50-100nm. L_{φ} is much smaller than the sample size (~10 µm), and on the similar order of magnitude with the estimate from weak localization (WL) measurements in our previous work⁸. By analyzing the magneto-transport behavior such as UCF and WL, we obtain information on the quantum transport of carriers. Such information can be valuable to design and improve graphene-based electronics --- including novel quantum coherent devices¹⁷, where phase coherent electron transport is utilized for new device functionalities or improved performance.

Electronic Properties of Graphene films grown on Cu

Transistor devices (Fig. 3a) have also been fabricated on the graphene films grown on Cu and then transferred to Si/SiO₂ substrates. Fig. 3b shows the electric field effect at 3K on sample "C". The carrier mobility reaches \sim 3000 cm²/Vs. Some hysteresis (depending on the sweep direction of gate voltage) is observable and might be related to charged impurities near graphene (eg. in the substrate) and/or to the metal contacts. More measurements are currently underway to further study the field effect as well as magnetotransport in such graphene grown on Cu.



Fig. 3. (a) Optical microscope image of a device (top view) fabricated by e-beam lithography from a graphene film grown on Cu then transferred to SiO_2/Si . (b) Four-terminal resistance as a function of back gate voltage, showing the electric field effect.

Summery

By measuring the electronic transport (such as field effect and magnetoresistance) in graphene films grown on Ni and Cu, we found such large scale graphene films can have excellent electronic properties and could open many application possibilities for carbon-based electronics, such as high speed/high frequency devices, conductive coatings and transparent or flexible electronics.

Acknowledgments

This research has benefited from support from Miller Family Endowment, Birck Director's Fund and Semiconductor Research Corporation (SRC)'s Nanoelectronics Research Initiative (NRI) via Midwest Institute for Nanoelectronics Discovery (MIND). HC acknowledges support from a Grodzins summer fellowship. Acknowledgment is also made to the donors of the American Chemical Society Petroleum Research Fund for partial support of this research. QY acknowledges support by NSF Grant 0620906 and CAM Special Funding. A portion of this work was carried out at the National High Magnetic Field Laboratory, which is supported by NSF Cooperative Agreement No. DMR-0084173, by the State of Florida and DOE. We thank Jun-Hyun Park and Eric Palm for experimental assistance.

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