Transport Properties and Defects at the Intersection of CVD Graphene Domains

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Abstract

We study intersection between two adjacent graphene domains grown by chemical vapor deposition (CVD) method over the hetero-epitaxial Cu catalyst, which is expected to give high quality and single crystal graphene with large area. The defects and carrier transport properties are investigated by Raman mapping and electrical measurement of graphene field effect transistor (g-FET). Our results suggest that some defects remain at the intersection even for domains with identical orientation.

1. Introduction

Graphene is greatly expected as a material for new electronics applications due to its unique and excellent physical properties, such as high carrier mobility and flexibility [1]. For preparation of graphene, CVD growth on Cu catalyst has attracted an interest for large area and low cost synthesis. The CVD graphene has multi domain structure with a number of domain boundaries [2] which limit the physical properties [3,4]. Therefore, it is important to investigate and control the domain structure of CVD graphene [5,6]. We previously reported the orientation-controlled graphene produced by epitaxial CVD growth using single-crystalline Cu and Co films [6-8], but it was unclear how the developing graphene domains merge together at the intersection during the growth.

In this study, we have investigated the intersection between adjacent graphene domains by using Raman mapping and electrical measurement of g-FET.

2. Experimental method

Hexagonal graphene domains were synthesized over hetero-epitaxial Cu(100)/MgO substrate by CVD methods using the mixed gases, CH₄ 10 ppm and H₂ 2.25% in Ar, at 1075 °C under the atmospheric pressure. Then, graphene domains were transferred onto a SiO₂/Si substrate by using polymethyl methacrylate (PMMA), thermal tape, and etching solution of Cu, which consists of FeCl₃ and HCl [7]. Transferred graphene domains were analyzed by optical microscope, atomic force microscope (AFM), and Raman spectroscopy. For g-FET, multiple Au/Ti electrodes were deposited onto hexagonal graphene domains by electron beam lithography and thermal evaporation. Electrical measurements were performed under four terminal system in vacuum ($\sim 2 \times 10^{-5}$ Pa) under the temperature range of 80-280 K.

3. Results and discussion

Large hexagonal-shaped graphene domains with three main orientations, 0, 30, and -10 degrees were obtained on Cu(100) film. The size of these domains is several tens of micrometers. The transferred graphene has uniform optical contrast, and the Raman 2D band is stronger than G-band $(I_{2D}/I_G \sim 1.5)$ and has narrow linewidth (30-40 cm⁻¹). These results indicate that graphene domains are high-quality and single-layer. We selected two merged graphene domains to characterize the structure and properties of the intersection of neighboring domains. It was found that two neighboring graphene domains merged with a different angle always give Raman D-band along the intersection between two graphene domains. On the other hand, two merged domains with the identical angle sometimes gave no D-band at the intersection.

For further understanding of the intersection of two adjacent domains, we measured carrier transport for these hexagonal domains. The carrier transport was measured within a hexagonal domain (intra-domain) and over two merged domains (inter-domain), as shown Figure 1. Here, we studied two types of merged domains, merged with the identical angle and different angles. From Figure 2, the inter-domain region showed lower conductivity than the intra-domain region irrespective of merged angle of domains. The temperature dependence of carrier mobility was observed clearly at intra-domain compared with that at the inter-domain, as shown Figure 3. These phenomena were observed not only for the domains with different angles but also for those with the identical angle. These results suggest that some defects remain at the domain boundary even for domains with identical orientations because they cause electron scattering and reduce carrier mobility.



Figure 1 Optical microscope image of g-FET. Four terminal measurements were carried out at two different positions in two merged hexagonal graphene domains; inter-domain and in-tra-domain areas.



Figure 2 Gate voltage dependence of conductivity of intra-(gray) and inter- domain (black) at 80 K.



Figure 3 Temperature dependence of mobility of inter-(gray) and intra-(black) domain.

3. Conclusions

We present the distributions of Raman D-band and the electrical transport properties across adjacent graphene domains to understand how the developing domains merge together. The results indicated that defects remain at the intersection between two graphene domains, reducing carrier transport at the intersection.

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