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Electrical Characterization of Bilayer Graphene Formed by Hydrogen Intercalation of Monolayer Graphene on SiC(0001)

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1. Introduction

Monolayer graphene (MLG) and bilayer graphene (BLG) are attractive materials for electronics applications because of their unique electronic properties, which differ depending on the number of graphene layers [1]. For digital-electronics applications, BLG is particularly of interest because a band gap opens in BLG when an electric field is applied perpendicular to the BLG plane [2, 3, 4]. By controlling the strength of the electric field, the band gap can even be tuned. This property may lead to new electronics devices, such as wavelength-tunable laser diodes. However, one important step required for application of BLG is to establish a method for its mass production. This is because exfoliating BLG from graphite, which is a commonly used method, is not suitable for obtaining BLG with a large area.

Epitaxial bilayer graphene (EBLG), which is grown on SiC(0001) by thermal decomposition of the substrate, is one of the possible ways to grow large-area BLG. Here, the EBLG is situated on top of a buffer layer, a graphene-like structure that exists in between the SiC substrate and the EBLG, and is electronically inactive. Using angle-resolved photoemission microscopy (ARPES), Ohta *et al*. showed that EBLG on SiC(0001) possesses the unique band structure of BLG and that the band gap opens intrinsically due to interaction with the SiC substrate [5]. We have investigated the electronic transport properties of EBLG grown in a UHV and detected band-gap opening [6]. Furthermore, we evaluated the mobility of the EBLG and found out that it is inferior to that of BLG exfoliated from graphite. Improvement of the quality of the EBLG is needed for its wide applications

Recently, it has been demonstrated that hydrogen intercalation of epitaxial monolayer graphene (EMLG) on SiC(0001) decouples the buffer layer from the substrate [7]. The buffer layer then becomes quasi-free-standing monolayer graphene (QFMLG), and as a result, the whole system (EMLG + buffer layer) turns into quasi-free-standing bilayer graphene (QFBLG). The electrical properties of QFBLG have not been reported. Here, we characterize the electronic properties of QFBLG in top-gated devices and compare the quality with that of EBLG on SiC(0001).

2. Experimental

The QFBLG was prepared by first making EMLG on SiC(0001). The EMLG was grown by annealing n-doped 4H-SiC(0001) at around 1800℃ in Ar pressure of less than 100 Torr. Next, EMLG-Hall bars were patterned by photolithography and reactive-ion etching using CF_4. Then, the substrate was annealed at around 1250℃ in H_2 pressure of about 25 Torr. To check whether the hydrogen intercalation had been accomplished, we used low-energy electron microscopy (LEEM) to evaluate the number of graphene layers [8]. After verifying the formation of the QFBLG, we fabricated top-gated devices using a method similar to that described in Ref. 6. The channel width and length were 2.5 and 7.5 μm, respectively. We performed four-terminal measurements at 2 K with a constant direct current of 1 μA.

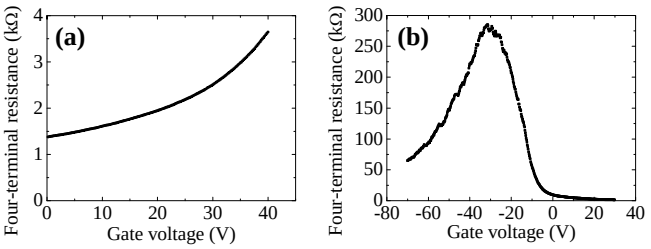

Fig. 1 Gate-voltage dependence of four-terminal resistance of (a) a top-gated QFBLG device and (b) a top-gated EBLG device measured at 2 K.

3. Results and discussion

Figure 1(a) shows the gate-voltage, V_g, dependence of four-terminal resistances, R, of the QFBLG. If the QFBLG is undoped, the maximum R should appear at V_g = 0 V, where the Fermi level reaches the charge neutrality point. However, R shows no peak at V_g = 0 V, and it increases with increasing V_g. This indicates that the QFBLG is p-doped. We also performed Hall measurements and confirmed that the carriers are indeed holes. This is consistent with the ARPES measurement [7]. The observed p-doping is different from that of EBLG on SiC(0001), as shown in Fig. 1(b) [6], where the EBLG is intrinsically n-doped. We also evaluated mobility, μ, as a function of carrier density, n, which is shown in Fig. 2. For comparison, n dependence of μ of EBLG on SiC(0001) in Ref. 6 is also plotted. The μ of the QFBLG increases with increasing n, which is the same as for the EBLG, and reflects the electronic structure of BLG [9]. Note that the carriers are holes for the QFBLG and they are electrons for the EBLG.

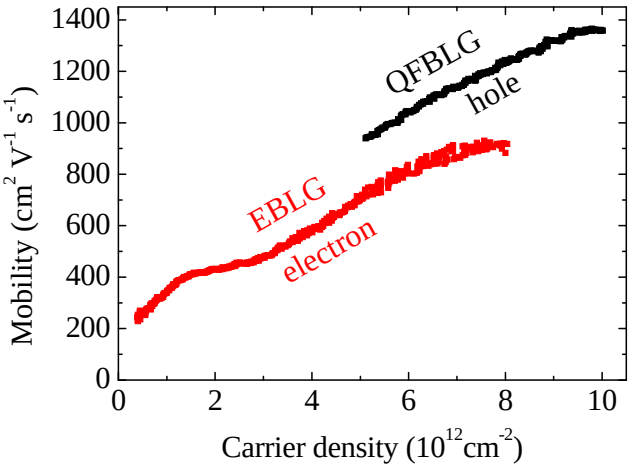

Fig. 2 Carrier density dependence of mobility of QFBLG on SiC(0001) and EBLG on SiC(0001).

In principle, electron mobility and hole mobility are the same in the BLG when n is the same due to its symmetric band structure [1]. Considering these facts, if we assume that the quality of the EBLG and QFBLG are the same, the hole mobility of the QFBLG is expected to be the same or lower than the electron mobility of the EBLG when they are compared at the same n. However, as shown in Fig. 2, μ of the QFBLG is higher than that of the EBLG. Because the device-fabrication process is almost identical between the QFBLG devices and the EBLG devices, notable degradation of sample quality of the EBLG during the fabrication process is unlikely.

Instead, we consider differences in their structural properties as the possible origins of the difference in μ. First, domain size is different between the two. In EBLG on SiC(0001), it is known that two types of domain co-exist, which are different in the stacking order of the two graphene layers [11]. Figure 3 clearly shows that the domain size of the QFBLG is larger than that of EBLG on SiC(0001). The larger the domain size is, the less carrier scattering there would be at the domain boundaries, which would lead to higher μ. Second, the number of atomic defects per unit area may be different between the two. Atomic defects are the sources of short-range scattering, which has a detrimental effect on μ. Because the EMLG and the buffer layer, which are the precursor of the QFBLG, are grown at higher temperature than the EBLG, surface diffusion of carbon and silicon atoms is enhanced [12]. Therefore, there would be fewer atomic defects per unit area compared to the EBLG and, consequently, less short-range scattering. Third, the surface structure underneath the EBLG and QFBLG are different: the EBLG and QFBLG reside on the buffer layer and H-terminated SiC(0001) surface, respectively. Thus, the strength of Coulomb scattering at the interfaces of the underlying layers and the graphene layers may be different. Although further studies are needed to correlate the structural properties of the grown graphene layers with μ, our results shows higher μ in the QFBLG than in EBLG on SiC(0001).

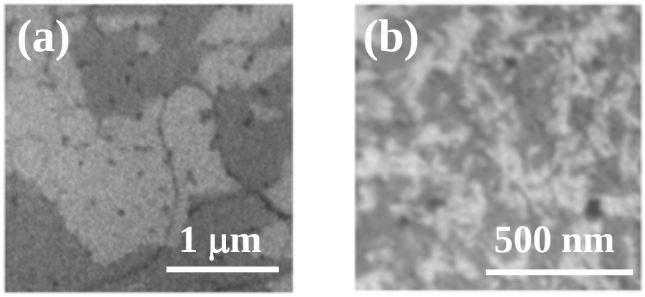

Fig. 3 Typical dark-field LEEM images of (a) QFBLG on SiC(0001) and (b) EBLG on SiC(0001). Two types of stacking domain are distinguished by differences in their contrasts.

4. Conclusions

We conducted electronic transport measurements of quasi-free-standing bilayer graphene on SiC(0001) in top-gated devices. The quasi-free-standing bilayer graphene was prepared by hydrogen intercalation of epitaxial monolayer graphene on SiC(0001). The gate-voltage dependence of resistance and the polarity of the Hall resistance at zero-gate voltage show that the carriers are holes. We also evaluated the mobility at various carrier densities and found out that the quasi-free-standing bilayer graphene shows higher mobility than epitaxial bilayer graphene on SiC(0001) when compared at the same carrier density. This suggest that hydrogen intercalation of epitaxial monolayer graphene on SiC(0001) may be one possible way to attain high-quality bilayer graphene on SiC(0001).

Acknowledgements

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