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Quantum Hall Effect and Band Dispersion in Artificially Stacked CVD Graphene

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Abstract

Graphene continues to attract intense interest as a two-dimensional electron system in which charge carriers are governed by Dirac equation in the limit of zero mass. Interestingly, recent studies of Raman spectroscopy imply that the feature of Dirac-cone dispersion is maintained even when CVD monolayer graphene is artificially stacked. In addition, the dramatic improvement of carrier mobility is reported in such artificially stacked CVD graphene. Here we investigate the quantum Hall effects (QHEs) of artificially stacked CVD graphene and verify the feature of band dispersion through Landau level (LL) splitting. The unusual broken-symmetry QHEs are observed in two- and three-layer graphene, indicating the band shift and level crossing due to the presence of interlayer coupling. We thus conclude that a complex low-lying dispersion is formed in the vicinity of the *K* point although no explicit band splitting is observed in Raman spectroscopy.

1. Introduction

Monolayer graphene exhibits linear band dispersion (Dirac-cone dispersion) in which charge carriers are governed by Dirac equation in the limit of zero mass [1-3]. Thanks to its linear band dispersion, high carrier mobility is realized and a variety of sensor applications are expected [4-7]. On the other hand, well-ordered multilayer graphene (i.e., AB or ABA-stacked graphene) exhibits higher conductivity than monolayer graphene because of parallel conductance. However, such multilayer graphene exhibits non-linear band dispersion similar to bulk graphite, yielding the suppression of carrier mobility.

The chemical vapor deposition (CVD) method has been one of the most popular techniques for synthesizing graphene. It is recently reported that carrier mobility is significantly improved in artificially stacked graphene by the repeated transfer of CVD monolayer graphene [8]. Furthermore, the G' peak in Raman spectroscopy remained narrow and almost symmetric line shape even after multiple transfer steps, implying linear band dispersion due to weak interlayer coupling [8].

Here we investigate the quantum Hall effects (QHEs) in monolayer, two- and three-layer stacked CVD graphene and verify the feature of band dispersion through Landau level (LL) splitting. The simplest multilayer model of well-ordered stacked graphene, in which only the nearest intra- and interlayer couplings are considered, results in symmetric electron and hole bands [9]. It follows that symmetric QHE states are expected with the $4 \times l$-fold degeneracy of zero-energy LL (zLL) (l: the number of layers). When the next-nearest layer coupling is considered, electron-hole symmetry is broken associated with band offset [9]. Accordingly, the broken-symmetry QHE states are observed and the degeneracy of zLL is lifted at a given magnetic field [9,10]. It is also shown that the band dispersion and material properties are extremely sensitive to the stacking angle [11-13]. In order to verify the low-lying energy spectrum, we here focus on the electron-hole symmetry and the degeneracy of zLLs in the QHE states of artificially stacked CVD graphene.

2. Experiments

Two-layer (2L) and three-layer (3L) artificially stacked graphene were prepared by the repeated transfer of CVD monolayer (1L) graphene onto a SiO_2/Si substrate. The detail synthesis processes of CVD graphene are described in Ref. [8]. Multiple electrodes are fabricated on the samples using electron-beam lithography followed by Au/Cr (50/5 nm) evaporation and a lift-off process (Fig. 1).

Transport properties were measured at 300 mK in a ^{3}He cryostat with a superconducting magnet. Raman spectra of the graphene devices were acquired at room temperature using laser excitation of 532 nm (incident laser energy E_L = 2.33 eV) with a 100× objective lens (typical spot diameter: 1 μm).

3. Results and discussion

The carrier mobility was measured through the conductivity as a function of the applied back-gate voltage V_g. The mobilities in 1L, 2L and 3L graphene are estimated to be 5000 $cm^2V^{-1}s^{-1}$, 11000 $cm^2V^{-1}s^{-1}$, and 10000 $cm^2V^{-1}s^{-1}$ at 300 mK,

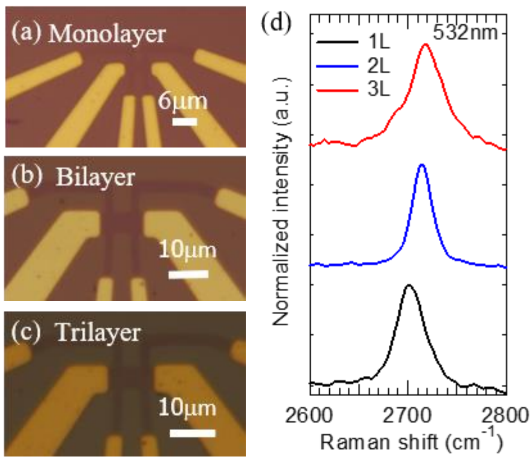

Fig. 1 Optical micrograph of (a) monolayer (1L), (b) two-layer (2L) and (c) three-layer (3L) graphene. (d) Raman spectra of these devices.

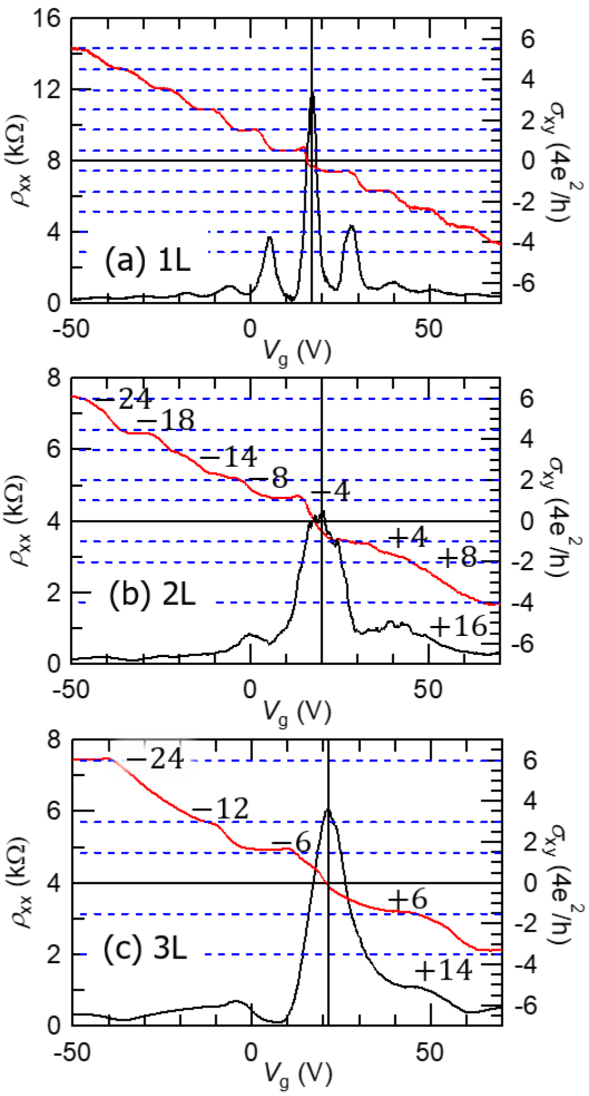

Fig. 2 Longitudinal resistivity and Hall conductivity as a function of V_g at $B = 9$ T in (a) 1L, (b)2L and (c) 3L graphene.

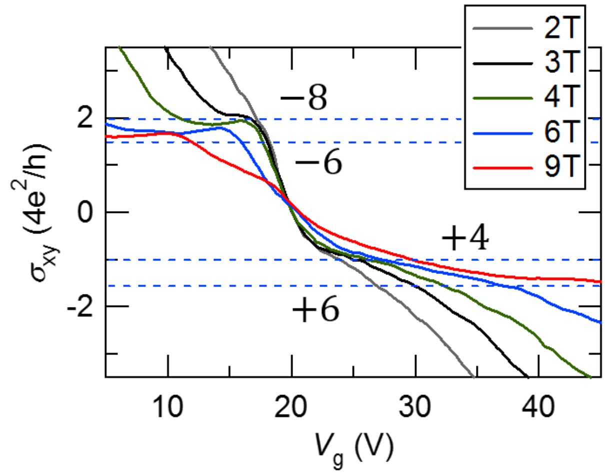

Fig. 3 Magnetic-field dependence of QHE plateaus in 3L graphene.

respectively. The mobility is significantly improved in artificially stacked CVD graphene as reported in Ref. [8].

Figure 1(d) shows Raman spectra of our graphene devices near 2700 cm^{-1} (G' peak). It is known that an asymmetric broad line shape is observed due to the splitting of electronic band dispersion in well-ordered stacked graphene [14]. However, the G' peak exhibits symmetric line shape in our devices as reported by previous studies [8]. This suggests that the splitting of band dispersion is negligibly small in the optically excited energy level ($E \sim E_L/2$).

Figure 2 shows the longitudinal resistivity and Hall conductivity as a function of back-gate voltage (V_g) at a magnetic field of $B = 9$ T for 1L, 2L and 3L CVD graphene. Well-defined symmetric QHE states are observed in 1L graphene, indicating the high quality of our synthesized CVD graphene. On the other hand, unusual broken-symmetry behaviors are observed in the QHE states of artificially stacked 2L and 3L grap hene. In addition, it is found that the zLL ($\nu = \pm 6$) is sensitive to magnetic fields in 3L graphene. The zLL is shifted to $\nu = -8$ and $+4$ around 4 T (Fig. 3) while the twelve-fold degeneracy is maintained.

In well-ordered stacked graphene, the Hall plateaus should follow the symmetric QHE states with LL filling factors, $\nu = \pm 4, \pm 8, \pm 12, \cdots$, for AB stacked 2L graphene [15] and $\nu = \pm 6, \pm 10, \pm 14, \cdots$, for ABC stacked 3L graphene [16]. It is also reported that the electron-hole symmetry is broken and the degeneracy of the zLL is lifted at a given magnetic-filed in ABA stacked 3L graphene because of the band offset and level crossing [10]. In addition, twisted bilayer graphene also exhibits an asymmetric QHE state that sensitively depends on magnetic fields through its specific interlayer coupling [13]. The unusual QHE behaviors observed in our experiments thus provide signs of the band offset and level crossing due to the presence of interlayer coupling.

4. Conclusions

We have studied artificially stacked CVD graphene through magnetotransport experiments and Raman spectroscopy. Our results indicate that a complex band dispersion is formed in the low-lying energy levels although no explicit band splitting is observed in the optically excited higher energy levels.

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