Magnetoresistance measurements of tetralayer graphene device with single gate electrode

T Hirahara¹, R Ebisuoka¹, K Watanabe², T Taniguchi² and R Yagi¹

¹Graduate School of Advanced Sciences of Matter, Hiroshima University, Higashi-Hiroshima, Japan
²National Institute for Material Science, Tsukuba, Japan

Abstract. We have studied low temperature magnetoresistance of tetralayer graphene. By analysing quantum magnetoresistance oscillation in detail, carrier density dependence of the fermi surfaces of orbits indicated that tetralayer graphene has two bilayer like bands whose energy of bottom of the bands were different. We estimated ratio of their band mass using carrier density dependence of magnetoresistance.

1. Introduction

Graphene has been studied intensively since the successful discovery of a method to create graphene flake with adhesive tape [1]. Electronic band structure of monolayer, bilayer and trilayer have been uncovered by experiments [1-6], showing unique properties for massless and massive Dirac fermions. Tetralayer, on the other hand, has not been fully understood. It was expected to have two bilayer like bands [7,8]. There are a few experimental works: insulating behavior near charge neutrality points studied by double-gate suspended graphene [9], and landau level structure studied by capacitance measurements [10]. In this paper, we measured Shubnikov-de Haas oscillation (SdHO) by a resistance measurement and studied electronic band structure of tetralayer graphene with a Bernal stacking (i.e., ABAB stacking) focusing on gate voltage dependence of carrier density of fermi surfaces.

2. Methods

We fabricated graphene by mechanical exfoliating high-quality Kish graphite with adhesive tape. Layer number was determined by color intensity (RGB digits) of optical images of graphene. A large number of graphene samples were measured to obtain distribution of RGB digits of graphene color. The distribution exhibited quantization due to discrete change of graphene’s color, which allowed us to determine layer number precisely. To improve quality, graphene flakes were transferred onto high-quality h-BN flakes using a washer method using PVA (poly vinyl alcohol) acrylic resin [6] as shown in Figure 1. Electric leads to measure resistivity were made by a standard electron beam lithography technique. An optical micrograph of a typical sample is displayed in Fig. 2. The graphene is indicated with lines for clarity. The Graphene flake consisted of tetralayer graphene. Fewer layer graphene was also present at the left regime. However we could measure resistance of tetralayer graphene with a four terminal measurement using Electrodes 3 and 1 for current injection and drain, and Electrodes 5 and 6 for voltage measurement, which allowed us to exclude picking up voltage drop due to fewer graphene layers.

yagi@hiroshima-u.ac.jp
Carrier density was tuned by a back gate voltage. A specific back gate capacitance was $C_g = 108 \text{ aF/µm}^2$. Magnetic fields were applied perpendicular to graphene devices using a superconducting solenoid. Measurements were done at a liquid helium temperature, $T = 4.2 \text{ K}$. Electric resistance was measured using a standard lock-in technique with excitation current of 1 µA.

3. Sample characterization

$V_g$-dependence of resistance at $B = 0 \text{ T}$ is shown in Fig. 3. Resistance took a peak (about 800 Ω) at about $V_g = -15 \text{ V}$, which apparently indicated a semiconducting or a semi-metallic behavior rather than a strong insulating one. Sample quality was estimated by a mobility, $\mu = 1/ne\rho$, as shown in Fig. 4. At high carrier density regime, mobility was about 10,000 cm$^2$V$^{-1}$s$^{-1}$, which was moderately higher than those that were expected for graphene samples fabricated on SiO$_2$ substrates.

4. Magnetotransport measurements

Left panel of Figure 5 shows traces of magnetoresistance for different carrier densities. Oscillations are due to Shubnikov-de Hass (SdH) effect arising from Landau quantization. Oscillations are periodic in $1/B$ as shown in a central panel of Fig. 5, showing a replot against reciprocal magnetic fields $1/B$. Oscillations looked complicated: they composed of more than one frequency. Fast Fourier Transform (FFT) was employed to determine frequency of the oscillations. Right panel shows Fourier spectra. Complicated peak structures in low frequency regime were possibly due to back ground of magnetoresistance. At higher frequencies, we could discern two salient peaks that varied with gate voltages. At electron regime, ($i.e. V_g > -13.3 \text{ V}$) two peaks were observed. Apparently these were not higher order harmonics, but would originate from fermi surfaces of different subbands. Figure 6 shows carrier density estimated from the frequency of SdHO. Here we used formulas, $n_{osc} = g f_s \Delta(B)$ and $f_s = 1/\Delta(B')$, where $\Delta(B')$ is a period, $g$ is degeneracy of landau level which we assumed to be 4 considering valley and spin. First, we focus our attention in electron regime where two frequency components were observed. These two would be light mass and heavy mass bilayer bands in tetralayer graphene. From the $V_g$-dependence of carrier density, red and blue points were identified to be light...
and heavy mass bilayer bands, respectively. Rate of variation of $n_{\text{osc}}$ for each orbit as a function of total carrier density $n_{\text{tot}}$ was dependent on the mass of the subbands since $\delta n_{\text{osc}} \propto m \delta E_i$ where $m$ is a band mass and $E_i$ is Fermi energy. Here we assumed that dispersion relation of bilayer bands can be approximated by $E - E_0 \approx \hbar^2 k^2 / 2m$ where $E_0$ is the energy of the band bottom and $m$ is a band mass. From the slope of $V_{xx}$-dependence of $n_{\text{osc}}$, ratio of the masses were estimated to be $m_{\text{light}} / m_{\text{heavy}} \sim 0.4$.

The sum of carrier density for light mass and heavy mass bilayer bands yields total carrier density. Black points in Fig. 6 is a sum of these carrier density determined from oscillation. The sum is approximately the same as $n_{\text{tot}}$ (large about factor of 1.025). This indicated that there are only two bands in this system.

Moreover, we could extract information about the dispersion relation from $n_{\text{tot}}$-dependence of $n_{\text{osc}}$. As mentioned earlier $n_{\text{osc}}$ for two bands both varied linearly with $n_{\text{tot}}$. This was a characteristic feature of subbands with dispersion, $E \propto k^2$. For other dispersion relation, such as $E - E_0 \propto k$, or $E - E_0 \propto k^3$, $n_{\text{osc}}$ varies as $(E_i - E_0)^2$ or $(E_i - E_0)^{2/3}$, resulting in non-linear $n_{\text{tot}}$ dependence of $n_{\text{osc}}$.

Energy of band bottom for light mass bilayer band was larger than that for heavy mass bilayer bands. This can be seen by the $n_{\text{osc}}$ value at $n_{\text{tot}}$, which was determined by extrapolating a relation between $n_{\text{osc}}$ and $n_{\text{tot}}$. A least square fitting for a liner line for light mass bilayer bands (red) intercepts the ordinate at a large value of $n_{\text{osc}}$ than that for heavy mass bilayer bands (blue).

Detailed landau level structure was revealed by the mapping magnetoresistance as a function of magnetic field and carrier density as shown in Fig. 7. We could discern characteristic landau level structure of trilayer graphene. Stripes corresponding to landau levels with different indices arose from SdH. The shape of this fan diagram was different from monolayer, bilayer and trilayer graphene. The fan diagram consisted of two sets of fans which are superposed. This is a direct consequence of above mentioned linearity in $n_{\text{osc}}$ to $n_{\text{tot}}$.

Offset of conduction band bottoms were observed as an offset of zero-mode landau levels. We could easily identify a zero-mode landau level for a heavy mass bilayer band at $n_{\text{tot}} \approx 0$ cm$^{-2}$. White broken lines in the figure indicates filling factors $\nu = 4i$, where $i$ denotes integers. Salient stripes at high magnetic fields are mostly due to heavy mass bilayer bands. Near $n_{\text{tot}} = 1 \times 10^{12}$ cm$^{-2}$, $N = 0$ landau level of the light mass bilayer band gave rise to jumps in the filling factors of the landau levels for the heavy mass. Apparently the filling factors varied by 8, showing the $N = 0$ landau level had eight-fold degeneracy as expected for bilayer bands.

![Figure 5](image-url). (Left) Magnetoresistance as a function of magnetic fields. Back ground was subtracted to obtain oscillatory components. (Center) Replot against $B^{-1}$. (Right) FFT results. Data were offset. From bottom to top $V$, was varied from -70 V (blue) to 70 V.
From the above results, there are two sets of bilayer band. There are three different stacking structures, ABAB, ABCA, and ABCB in tetra-layer graphene. These are expected to have different electronic structures, number of bands and their dispersions are different to one another. The ABAB stacking is the only stacking that has two-sets of bilayer bands.

5. Conclusion
We have succeeded in observing landau level structure of Bernal stacked tetra-layer graphene by magnetotransport measurements. Tetra-layer graphene has verified to have to sets of bilayer bands. The light mass bilayer band had larger energy for conduction band bottom than that for the heavy mass bilayer band. A ratio of band masses was estimated.

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