Uniqueness of Intraband Plasmon Dispersion of a Single Layer Graphene


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ABSTRACT

We have studied the dispersion of intraband plasmon excitation of a single layer graphene (SLG) formed on 6H-SiC(0001) surface by using high-resolution electron energy loss spectroscopy. The dispersion (q) measured from SLG appears to be quite different from that of other multilayer graphene systems reported so far but in good accord with recent theory. We also discuss how to form and identify the presence of a SLG by monitoring the spectral change of Fuchs-Kliewer (F-K) phonon and interband transition during the initial graphitization process on the 6H-SiC(0001) surface. We thus present the unique behavior of intraband plasmon of SLG not hampered by multilayer effects or coupling with other excitations revealing another intrinsic nature of a SLG in addition to the well-known linear Dirac band.

Keywords: single layer graphene, intraband plasmon, energy-momentum dispersion

1 INTRODUCTION

Graphene, two dimensional form of carbon atoms arranged in hexagonal lattice, has recently attracted a great deal of research interest due to its peculiar phenomena as well as its potential applications in next-generation electronics and photonics [1]. A. K. Geim and K. S. Novoselov first extracted a single layer graphene (SLG) from a piece of graphite in 2004 [2], however, such a method is not suitable for large-scale integration. As one of the alternatives, an epitaxial graphene grown on SiC substrate has been well documented [10]. Graphene, two dimensional form of carbon atoms arranged in hexagonal lattice, has recently attracted a great deal of research interest due to its peculiar phenomena as well as its potential applications in next-generation electronics and photonics [1]. A. K. Geim and K. S. Novoselov first extracted a single layer graphene (SLG) from a piece of graphite in 2004 [2], however, such a method is not suitable for large-scale integration. As one of the alternatives, an epitaxial graphene grown on SiC substrate has been well documented [10].

Our experiments have been performed using spot-profile analysis of low energy electron diffraction (SPA-LEED) and Leybold-Heraeus ELS-22 spectrometer with two 127° cylindrical deflectors for both monochromator and analyzer. The optimum energy resolution and the half-acceptance angle of the detector are 19 meV and 2°, respectively. An epitaxial SLG was produced on Si terminated n-type 6H-SiC(0001) wafer purchased from Cree, Inc. When the sample is first cleaned by heating at 830°C under Si flux for 1 hour, a Si-rich (3 × 3) reconstruction can be obtained. After this process, a (√3 × √3)R30° (√3 for shot), a buffer layer of (6√3 × 6√3)R30° (6√3) and the graphene layers are formed by further annealing at higher temperature, as presented elsewhere [10]. The annealing temperature is monitored by an optical pyrometer assuming an emissivity of 0.90. In order to avoid any multilayer effect in our EELS measurement, we have prepared an early-stage sample of forming SLG onto the insulating 6√3 buffer layer. Details will be discussed in below.

2 EXPERIMENTAL DETAILS

The sequence of surface phases in Si-terminated 6H-SiC(0001) surface, from the Si-rich (3 × 3) to the epitaxial graphene, has been well documented [10]. Figure 1 shows the characteristic loss peaks for each intermediate stages, the √3 and the 6√3 phase, during the graphitization process of SiC surface. The L1 loss peak observed from √3 phase at 2.3 eV has been understood...
as an inter-Hubbard band transition [11]. In addition, for the first time we find a L₂ peak at 1.7 eV from the 6√3 phase. In order to identify the origin of this new loss peak, we consider the surface electronic structure of the 6√3 reconstruction determined by previous photoemission (PES) studies. The buffer layer where the linear Dirac band has not yet formed due to partial hybridizations of p₂ orbitals with the SiC substrate [12] has several flat surface bands, g₁ at 0.5 eV and g₂ at 1.6 eV below Fermi level, and A and B, at 1.0 and 2.4 eV, above Fermi level, respectively [13]. Therefore, we suggest an interband transition from g₁ to A as a possible origin for the L₂. Interestingly, the L₂ rapidly decays with the growth of SLG, while the Drude tail of metallic continuum becomes significantly intense due to the appearance of a single π-band crossing a Fermi level.

In present work, the formation of SLG has been probed by a combination of several tools such as SPA-LEED, APRES and HREELS. Previous LEED study has shown that intensity versus energy (I-V) curve for the first order diffraction spot of graphene sensitively reflects the change in the number of graphene layer [14]. We have observed such a remarkable change in our I-V curve upon the formation of bilayer graphene (not shown). Figure 2(a) shows the evolution of EELS spectra for interband π-plasmon near 6 eV with increasing thickness of graphene layer, which agrees with that reported earlier [15]. The absence of multilayer graphene is also confirmed by observing the unsplit linear π-band of graphene in the angle-resolved PES intensity map, because the split and parabolic π-bands begin to develop by further annealing. Figure 2(b) shows the band image for the SLG sample at an early stage of formation on the insulating buffer layer, where the single π-band coexists with two weak states (g₁ and g₂) from the buffer layer not covered by SLG.

Although the formation of SLG can be identified as described above, we discuss another method to control the growth of graphene layer by monitoring the spectral change of F-K phonon. The F-K phonon peak resolved at loss energy of 117 meV from the buffer layer, progressively shifts toward high loss energy with increasing thickness of graphene layer as shown in figure 3(a), and its energy reaches up to 140 ∼ 150 meV when few layer graphene (FLG) is formed. T. Angot et al. have already reported that the F-K phonon peak observed at 124 meV may indicate the formation of FLG [16]. Here, the SLG of hω_{FK}=120 meV represents the early-stage of SLG with no FLG formed. Figure 3(b) shows the EELS spectra obtained from the SLG and from the FLG. The characteristic L₃ loss peak is identified as a low-energy intraband π-plasmon excitation [5]–[8] since its linewidth is too broad to be a loss peak either from a local atomic vibration or from a phonon. Moreover, we may also rule out the possibility of an interband transition because the peak is observed even at small q region below q_c. In order to understand such novel collective excitation of massless Dirac fermions, we have fitted our experimental data with a theoretical curve calculated for doped graphene [3],[4]. The collective mode within RPA is given by

\[ \varepsilon(q, \omega) = 1 + v(q) \Pi(q, \omega) = 0 \]  

(1)

where \( v(q) = 2\pi e^2/\kappa q \) is the 2D Coulomb interaction in the wave vector space and \( \Pi(q, \omega) \) is the 2D polarizabil-
ity written as a summation of $\Pi^-(q,\omega)$ and $\Pi^+(q,\omega)$ for a doped graphene. In the fitting, we have used a Gaussian line-shape for the loss peak after subtracting the background with a polynomial function [17]. We have fitted with only one fit-parameter, electron density $n$, and adopted the reasonable values for the SLG on SiC surface; a background dielectric constant $\kappa = (1 + \kappa_{SIC})/2 = 5.5$ [3] and the Fermi velocity $v_F = 1.1 \times 10^6 m/s$ [12], [18].

As displayed in figure 4, the plasmon dispersion $\omega(q)$ of SLG is well described by the recent theory for a doped graphene (green curve) with a linear band dispersion. We obtain the best-fit value for $n = 1.2 \times 10^{13}/cm^2$, which agrees with previous estimation from ARPES experiment [12], [18]. Therefore, the Fermi wave vector and the Fermi energy are estimated to be $k_F = \sqrt{\pi n} = 0.061 \AA^{-1}$ and $E_F = \hbar v_F k_F = 0.44 eV$. In contrast to a typical 2D plasmon, the plasmon from SLG survives far beyond $q_c$ because it does not enter the intraband single-particle excitation (SPE) region. However, Stern’s full formula (orange curve) [9] usually applied to normal 2D systems does not reproduce our experimental data. And the dispersion (red square) measured from FLG appear also quite distinct from that of SLG. Such a difference in the plasmon dispersion may come from interaction with other excitations such as in-plane plasmon mode and parabolic $\pi$-bands.

4 SUMMARY

In summary, we have prepared an early-stage SLG sample formed on the insulating $6\sqrt{3}$ buffer layer, and measured the dispersion relation of the low-energy intraband $\pi$-plasmon, using HREELS. The measured energy-momentum dispersion appears to be quite distinct from that of normal 2D electrons as well as of FLG. And the plasmon dispersion from SLG with no multilayer effects is well described by theory predicted within RPA method. We also identify loss peaks characteristic to the intermediate phases during the graphitization process on 6H-SiC(0001) substrate, which may be used as an in-situ probe to sensitively and conveniently identify the formation of SLG.

REFERENCES