Comparison of the formation process and properties of epitaxial graphenes on Si- and C-face 6H–SiC substrates

Wang Dang-Chao(王党朝)a), Zhang Yu-Ming(张玉明)a), Zhang Yi-Men(张义门)a),
Lei Tian-Min(雷天民)a), Guo Hui(郭辉)a), Wang Yue-Hu(王悦湖)a),
Tang Xiao-Yan(汤晓燕)a), and Wang Hang(王航)a)

a) School of Microelectronics, Xidian University, Key Laboratory of Wide Band-Gap Semiconductor Materials and Devices,
Xidian University, Xi'an 710071, China
b) School of Physics and Electronic Engineering, Xianyang Normal College, Xianyang 712000, China

(Received 15 July 2011; revised manuscript received 30 August 2011)

In this paper, the epitaxial graphene layers grown on Si- and C-face 6H–SiC substrates are investigated under a low pressure of 400 Pa at 1600 °C. By using atomic force microscopy and Raman spectroscopy, we find that there are distinct differences in the formation and the properties between the epitaxial graphene layers grown on the Si-face and the C-face substrates, including the hydrogen etching process, the stacking type, and the number of layers. Hopefully, our results will be useful for improving the quality of the epitaxial graphene on SiC substrate.

Keywords: SiC substrate, epitaxial graphene, Raman spectroscopy

PACS: 81.05.ue, 78.30.–j, 61.48.Gh

DOI: 10.1088/1674-1056/21/3/038102

1. Introduction

Epitaxial graphene grown on single-crystalline silicon carbide (SiC) has attracted great interest due to its compatibility with the current CMOS technology and applications in making graphene-based devices.[1,2] SiC has two polar faces perpendicular to the c axis, called the (0001) Si face and the (0001) C face. It is believed that the epitaxial graphene layers formed on the different faces need different growing conditions and have different electronic properties.[3,4] Previous experiments dealt mainly with the fabrication of the epitaxial graphene under ultra high vacuum (UHV)[3–7] and argon atmospheric conditions.[8] The epitaxial graphene grown in the UHV condition is only 100 nm in size at most and is of poor quality.[9] In the argon atmospheric environments, the epitaxial graphene samples are of better quality with much larger domain sizes, but the corresponding formation temperature needs to increase considerably.[8] The formation mechanism of the epitaxial graphene is still not clearly understood. In this paper, we fabricate epitaxial graphene layers on Si-face and C-face SiC substrates under a lower argon gas pressure (about 400 Pa) at 1600 °C, and make a comparison of their formation and characteristics.

2. Experiment

In our work, 6H–SiC (0001) and (0001) substrates, each with an n-type nitrogen concentration of about 10^{15} cm^{−3}, were used for the epitaxial growth of graphene. They were pre-cleaned to remove surface contaminants by using chemical treatments before entering an Aixtron/Epigress VP 508 hot-wall chemical vapour deposition (CVD) reactor chamber. Two different processes for hydrogen-etched Si- and C-face substrates were adopted: (i) at 1600 °C with a flux of 90 standard liters per minute under a pressure of 1.01×10^{4} Pa, and (ii) at 1600 °C with a flux of 60 standard liters per minute under a pressure of 1.01×10^{5} Pa. Following the etching process, the substrates were deposited at low pressure (400 Pa) with an argon flux at 1600 °C for growing graphene. Using an alternating current (AC) mode atomic force microscopy (AFM: Agilent 5500) in air at ambient pressure, the AFM maps were obtained as shown in Figs. 1 and 2.
3. Measurements and results

Figures 1 and 2 show the AFM images in a 10 μm × 10 μm zone of Si- and C-face of the 6H–SiC substrates processed by hydrogen-etching processes (i) and (ii), respectively. Figure 1(a) shows a more regularly stepped surface with a smaller root mean square roughness (0.496 nm) than that (24.09 nm) in Fig. 1(b), although they are almost the same in height (3 nm). However, figures 2(a) and 2(b) show significantly different structures. Figure 2(b) shows a more regularly stepped surface, while figure 2(a) exhibits an irregularly stepped one.

Raman spectroscopy has been considered as a quick and useful tool for the characterization of the epitaxial graphene on a SiC substrate. The Raman spectrum of the epitaxial graphene shows two main features, the G band and 2D band. The G band at around 1580 cm\(^{-1}\) is the characteristic of the sp\(^2\) hybridization and involves the in-plane optical phonon \(E_{2g}\) near the first Brillouin zone center of the phonon band structure.\(^{[10]}\) The 2D band at around 2700 cm\(^{-1}\) is a second-order band in the highest optical branch near the \(K\) point.\(^{[10]}\) The disorder-induced D band at around 1350 cm\(^{-1}\) often clearly appears in the Raman spectrum, which is sensitive to the domain size and the disorder and the edge defects in the lattice structure.\(^{[3]}\)

Figures 3 and 4 show the Raman spectra of the epitaxial graphene layers grown at 1600 °C under a low argon pressure about 400 Pa using the Si and the C faces of 6H–SiC respectively. It is obvious that the epitaxial graphene layers have indeed grown on the Si and the C faces of the SiC substrates because the G and the 2D bands occur simultaneously. These two types of epitaxial graphene layers have different properties.

For the Si face of 6H–SiC using hydrogen-etching method (i), figure 3 (curve c) shows a narrow, sharp and symmetric G band and 2D band, and a weak D band suggesting that the epitaxial graphene apparently takes the shape with a high quality. It is very
similar to the C face of 6H–SiC, which was grown by using the hydrogen-etching method (ii) and is shown in Fig. 4 (curve c).

Figure 3. Raman spectra of (curve a) 6H-SiC (0001) substrate, (curve b) and (curve c), epitaxial graphene grown on 6H–SiC (0001) substrate at 1600 °C under low pressure of 400 Pa using the hydrogen-etching methods (ii) and (i), respectively.

Figure 4. Raman spectra of (curve a) 6H-SiC (0001) substrate, (curve b) and (curve c), epitaxial graphene grown on 6H–SiC (0001) substrate at 1600 °C under low pressure of 400 Pa using the hydrogen-etching methods (i) and (ii), respectively.

However, as shown in Fig. 3 (curve b), by using the hydrogen-etching method (ii) the G band is invisible due to the effect of the substrate, and the intensity of the 2D band is weak and that of the D band is strong, which suggests that the epitaxial graphene is weakly generated on the substrates. As shown in Fig. 4 (curve b), using the hydrogen-etching method (i), the intensity of the 2D band is feeble, and the intensities of the G and the D bands are strong, suggesting that the epitaxial graphene is also weakly generated on the substrate but with poor quality.

As can be seen from Figs. 3 and 4, under the same growing condition of low argon pressure about 400 Pa at 1600 °, hydrogen etching method (i) for the Si face and hydrogen etching method (ii) for the C face are suited for growing high quality graphene.

Figure 5 shows a comparison between the properties of the epitaxial graphene layers grown on Si- and C-face SiC substrates. The inset in Fig. 5 (curve a) shows that the 2D band can be fitted with a four-Lorentzian function, we can judge that the epitaxial graphene has a property of the bi-layer epitaxial graphene with an A–B stacking.[13–14] While in the inset of Fig. 5 (curve b), the 2D band can be fitted with a single-Lorentzian function with a full width at half maximum (FWHM) of 64 cm⁻¹, and FWHM (D) 35 cm⁻¹, FWHM (G) 26 cm⁻¹, we can deduce that the epitaxial graphene has a property of the turbostratic multilayer graphite, not the A–B stacking structure.[13,14]

Figure 5. (colour online) Raman spectra of high quality epitaxial graphene layers grown on (curve a) Si-face and (curve b) C-face of 6H–SiC substrates at 1600 °C under low pressure. The upper inset shows that the 2D band can be fitted with four-Lorentzian function, and the lower inset shows the turbostratic graphite.

As is known, there are a lot of scratches and several disordered bi-layers on the surface of the as-received SiC substrate, which is harmful to the formation of the epitaxial graphene, so hydrogen-etching is carried out to remove the damaged layers leaving a uniform and well-ordered stepped morphology. The typical terrace width is about a few microns, and the step height is a few nanometers.[3] This is very conducive to the pre-graphitization of carbon atoms in the case of the Si face with a periodic $6\sqrt{3} \times 6\sqrt{3}R30^\circ$ and the C face of the $\sqrt{3} \times \sqrt{3}R30^\circ$ structure.[3] Therefore, different hydrogen-etching processes can form different morphological lattice structures and different properties of the epitaxial graphene.
4. Conclusion

In this work we investigate the formation and the characters of epitaxial graphene layers grown on Si- and C-face 6H–SiC substrates under low pressure of 400 Pa at 1600 °C. It is shown that the epitaxial graphene layers grown on the Si and the C faces are significantly different in several aspects. Although we have not yet understood the difference between these two types of epitaxial graphene layers in detail, we find a way to gain a higher quality epitaxial graphene layer on the SiC substrate.

References