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Epitaxial synthesis of graphene on 4H-SiC by microwave plasma chemical vapor deposition

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Abstract

Epitaxial graphene (EG) on semi-insulating SiC prepared by a thermal decomposition method is the most promising strategy for graphene application in large-scale integrated circuits due to compatibility with current semiconductor processes. In this study, high-quality few-layer graphene (FLG) was epitaxially grown on the semi-insulating on-axis 4H-SiC by microwave plasma chemical vapor deposition (MPCVD). Both sides of the SiC substrate were etched with H2 plasma at ∼1000 °C to promote the nucleation and the growth of graphite before epitaxial growth. The surface morphology and properties of EG on SiC(0001) were measured by energy-dispersive x-ray (EDX), x-ray photoelectron spectrogram (XPS), and atomic force microscope (AFM). The qualities of EG grown on the two surfaces of SiC at various temperatures were checked by Raman spectroscopy. Furthermore, the EG growth was controlled by the effect of Ar plasma in the MPCVD and the few-layer (1–3 layers) high-quality EG films were formed on SiC(0001). The room temperature Hall mobility of EGs up to 2790 cm² V⁻¹ s⁻¹ on SiC(0001) was realized.

1. Introduction

Graphene is a 2D layered material that is composed of sp² hybridized carbon arranged in a regular hexagonal pattern [1]. Since graphene was effectively isolated in 2004, many remarkable properties have been reported, such as optical transparency up to 97%, electron mobility up to 27000 cm² V⁻¹ s⁻¹, a thermal conductivity greater than 5000 W mK⁻¹, and the room temperature anomalous quantum Hall effect [2–5]. Due to its unique characteristics, graphene has received great interest from fundamental research and device applications such as electronic devices, catalysts, energy storage, and gas sensors [6–9]. However, scalable synthesis up to the industrial level is still very challenging. Various strategies have been proposed to overcome this issue, such as chemical exfoliation, thermal chemical vapor deposition (TCVD), and thermal decomposition of silicon carbide (SiC) [10–12]. TCVD synthesis has been known as a method with great promise for large-area and uniform graphene preparation [13]. However, it heavily relies on transition metal as a catalyst [14, 15]. For large-scale graphene synthesis, the method of growing epitaxially graphene (EG) on SiC by thermal decomposition is particularly attractive [16, 17]. Si atoms are evaporated and separated selectively from the surface when the SiC substrate is annealed at a high temperature above 1200 °C, and the rest of the Cr atoms form graphene by naturally restructuring [18]. Due to the semi-insulating SiC substrate, the grown graphene can be fabricated into diverse electronic devices directly without substrate corrosion, film migration, and other complicated processes [19, 20]. It is one of the most effective ways to realize the application of graphene in microelectronics [21]. Generally, a high-temperature hot-wall CVD furnace reactor is usually employed in the epitaxial growth of graphene on the SiC [22]. However, it is expensive, high energy consumption, and inefficient for temperature increase. In recent years, microwave plasma chemical vapor deposition (MPCVD) has been shown successfully
in the synthesis of graphene. This deposition technique is being optimized for the synthesis of high-quality graphene at low temperature \[23, 24\]. Nevertheless, the investigations on the epitaxial growth of graphene on the SiC using MPCVD are still lacking so far. In this study, we have successfully synthesized high-quality few-layer graphene (FLG) on the 4H-SiC by MPCVD.

2. Experimental

2.1. Fabrication of epitaxial graphene film

Graphene film on the SiC was synthesized by the MPCVD system (DMT Technology Co., Ltd China). The microwave power was provided by a 2.45 GHz generator (MUEGGE Inc. Germany) with a maximum output power of 6 kW. A schematic diagram of the MPCVD chamber is shown in figure 1(a). The chamber consists mainly of the resonant cavity, stub tuner, quartz window, and cooling stage in addition to the generator. The working principle of MPCVD was described in detail in a previous report \[25\]. The semi-insulating 4H-SiC wafer (purchased from CREE Inc.) was used as the substrates were cut into square pieces with a dimension of 10 × 10 mm, and was chemically cleaned in acetone and methanol to remove the surface organic impurities. The cleaned substrate was placed into the MPCVD chamber with a Mo stage that directly contacted with the cooling stage. The chamber was evacuated to a base pressure of about 0.1 Torr before the process gases entered. Figure 1(b) illustrates the process of graphene MPCVD-synthesis on the SiC. First, the sample was heated up to 1000 °C by a plasma ball generated by the microwave generator. Then the annealing step was performed in 30 Torr of H\(_2\) for 30 min at 1000 °C to etch the SiC surface and broaden the width of the steep terraces. After this step, the temperature was immediately increased by stages to 1200 °C with the mixture gas of H\(_2\) and Ar. The SiC surface was treated to build a condition feasible for EG synthesis \[26\]. After that, the growth step was performed at 1300 °C–1500 °C under the pressure of 50 Torr with H\(_2\)/Ar mixture gas for 30–50 min. Finally, the sample was cooled down to room temperature.

2.2. Characterization and measurements

The surface morphology of H-etched SiC was observed by an AFM (Veeco Inc.), and the results were analysed by the Nanoscope analysis software. The images of the graphene on the SiC and the EDX spectra were obtained by field emission scanning electron microscopy (FE-SEM, FEI Inc.). High-resolution XPS spectrum measurement was performed on a Theta Probe spectrometer (Thermo Fisher Scientific). Raman spectroscopy and mapping image were carried out by a confocal Raman microscope (HORRIBA-JY) using a 532 nm excitation laser.

3. Results and discussion

3.1. Hydrogen etching treatment of SiC substrate

Although the SiC surface has been polished, there are still many mechanical scratches and disordered defects on the SiC substrate. Therefore, the substrate must be etched with hydrogen to facilitate the nucleation and growth of graphite before epitaxial growth \[27, 28\]. Figure 2(a) shows the AFM image of SiC (0001) etched with
Firstly, graphene was epitaxially grown on SiC by MPCVD. The etching conditions included the temperature of 1000 °C, the H2 flow rate of 300 sccm, and the etching time of 30 min. Under this etching condition, the terrace morphology with a width close to the micron level and height about the atom level was obtained. The H2-etched SiC surface has a similar scale but curved terrace morphology, as shown in figure 2(b). These step structures were beneficial to stable nucleation and large area growth of graphene. As shown in reaction equation (1), the specific mechanism of hydrogen etching is that SiC reacts with hydrogen plasma at a high temperature to generate gaseous C2H2 and SiH4. Owing to the existence of H2 plasma in MPCVD, Si atoms reacted with H2 plasma at a lower temperature to form SiH4 gas which was carried away from the SiC surface by H2 flow. Thus, the etching process at a low temperature can be successfully implemented.

\[
\text{SiC} (\text{solid}) + \text{H}_2 (\text{plasma}) \rightarrow \text{C}_2\text{H}_2 (\text{gas}) + \text{SiH}_4 (\text{gas})
\]  

(1)

3.2. Basic epitaxial growth of graphene on SiC (0001)

Firstly, graphene was epitaxially grown on SiC (0001) at a temperature of 1300 °C. Figure 3(a) shows the carbon content which was measured by the energy spectra of EDX of the surface before and after the epitaxial growth. The electron beam energy was 10 kV. The atomic ratio of carbon content on the SiC surface before and after the epitaxial growth was 27.4% and 46.65%, respectively. Therefore, on the surface after growth, the carbon content was significantly higher than that of the original substrate. With the increase of beam energy, the atomic ratio of carbon content in the original SiC had no significant change, but it decreased gradually after the epitaxial growth, which indicates that there was a layer of carbon covering the surface of the SiC substrate. This also proved indirectly the existence of graphene. Figure 3(b) shows the Cls XPS spectrum fitted well by three peaks after epitaxial growth on SiC (0001). The peak of binding energy located at 283.5 eV was from the SiC substrate. The G peak (284.5 eV) at higher binding energy was close to the C-sp² bond position of graphite. This indicated that graphitization had occurred on the silicon face of SiC, which may form graphene by this experimental method. The binding energy position of the B peak (~284.9 eV) could be attributed to the buffer layer [29]. Figure 3(c) shows the AFM image of EG in the 1 μm × 1 μm region on SiC (0001). It can be seen that the grain boundary was obvious and the grain size of graphene was about 100 nm. The RMS roughness in the entire region was about 0.511 nm. The top of figure 3(d) shows the height along the white dotted line marked in figure 3(c). The height between region A and region B was ~0.25 nm, which was equivalent to the thickness of a Si–C diatomic layer. While the height between region B and region C was ~0.34 nm, which was exactly consistent with the thickness of the graphene monolayer. This can be explained by the schematic diagram shown at the bottom of figure 3(d). The epitaxial growth of graphene monolayer was realized in regions A and B on the buffer layer. Region B consumed one more Si–C bilayer than region A. On the contrary, due to the temperature or synthesis time was not sufficient, the buffer layer in region C had not enough energy to form the graphene layer. Figures 3(e) and (f) are the height and phase from AFM image of EG in the 1 μm × 1 μm region on SiC (0001) at 1300 °C, respectively, which show that the graphitic surface exhibits a meshwork of wrinkles. These wrinkles were frequently observed in FLG on SiC (0001) [30]. The origin of the wrinkle-like network with a ridge height of ~2.4 nm was attributed to the compressive stresses that develop upon cooling from the growth temperature [31].
3.3. Change temperature and Ar flow rate

For the graphene epitaxial growth on the SiC substrate, Raman spectra can not only be used to confirm the formation of the graphene but also to estimate the number of layers, doping, defects, stress and other properties of the graphene [32]. Figure 4(a) is the Raman spectra of graphene grown on SiC (0001) with various temperatures by MPCVD epitaxy, and the data have been normalized. The growth temperatures were set at 1200 °C, 1300 °C, 1400 °C, and 1500 °C, respectively. It can be seen clearly that there was no obvious G peak and 2D peak in the Raman spectrum of growth at 1200 °C (black line in figure 4(a)), indicating that there was no graphene formed on the substrate. When the temperature was set at 1300 °C, the G peak (1587 cm$^{-1}$) and 2D peak (2730 cm$^{-1}$) appeared in the Raman spectrum indicating the graphene formation on the substrate [33]. The full width at half maximum (FWHM) value of 2D peak at 1300 °C was ~62.8 cm$^{-1}$ which was close to monolayer graphene (59 cm$^{-1}$) [32]. The intensity of the G peak was lower than that of SiC substrate peaks,
which indicated that the graphene grain size was small and a continuous graphene film was not formed. According to equation (2) [34], the grain size of EG can be estimated:

\[
L_a = 2.4 \times 10^{-10} \lambda_{laser}^4 \left( \frac{I_D}{I_G} \right)^{-1}
\]

where \(L_a\) is the average grain size of graphene, \(\lambda_{laser}\) is the wavelength of the laser beam in Raman spectra, \(I_G\) and \(I_D\) are the intensities of \(G\) peak and \(D\) peak in Raman spectrum respectively. The average grain size of EG at 1300 °C was \(\sim 114\) nm, which was consistent with the observation of AFM.

The obvious \(D\) peak (\(\sim 1360\) cm\(^{-1}\)) in the Raman spectrum of growth at 1400 °C indicated that there were impurities or defects in the lattice structure of graphene. As the growth temperature increased to 1500 °C, the value of \(I_G/I_{SiC}\) became larger, indicating that the graphene fully covered the SiC substrate. The gradual increase of \(I_G/I_D\) indicated that high-quality graphene has been obtained at the growth temperature of 1500 °C. As shown in figure 4(a), the 2D peak in the blue curve is located at \(2713\) cm\(^{-1}\). A red-shift of \(17\) cm\(^{-1}\) was observed compared with that of growth at 1300 °C. The red-shift of the 2D peak in the Raman spectra of EG could be ascribed to the internal stress of the EG layer. When the temperature increases, the lattice mismatch between the substrate and the graphene was induced by the stress generated by thermal expansion. At the same time, the intensity of the 2D peak at 1500 °C was stronger which indicated a thicker graphene layer with an increase of temperature. Figure 4(b) shows the Raman spectra of graphene films grown at various temperatures on SiC (0001). Different from the silicon surface, the intensities of graphene characteristic peaks were highly strong which were almost unaffected by the SiC substrate peaks. As the growth temperature increases from 1300 °C to 1500 °C, the \(G\) and 2D peaks of the Raman spectra of EG on SiC (0001) were located at about \(1587\) cm\(^{-1}\) and \(2706\) cm\(^{-1}\), respectively. The intensities of \(D\) peaks were very small, indicating that the density of defects was low and the crystallization quality was high. As the growth temperature increased, the \(I_G/I_D\) and FWHM values of 2D peaks increased, indicating that the number of graphene layers increased gradually.

The sublimation rate of silicon atom plays an important role in the reconstruction of graphene. High argon pressure can effectively reduce the sublimation rate of silicon atom [35]. Figure 4(c) shows the Raman spectra of the graphene growth on SiC (0001) with Ar flow rates of 100 sccm and 200 sccm, respectively. When Ar flow was at a low level, \(D\) peak was obvious, and the \(I_G/I_{SiC}\) was \(\sim 1.23\). When Ar flux increased to 200 sccm, \(D\) peak was almost invisible, while \(I_G/I_{SiC}\) was \(\sim 0.68\). As the Ar flow rate increased, the intensities of the substrate peaks

![Figure 4. The Raman spectra of graphene grown on SiC: (a) On SiC (0001) at various temperatures by MPCVD epitaxy, (b) On SiC (0001) at various temperatures by MPCVD epitaxy, (c) On SiC (0001) at Ar flow rates of 100 sccm and 200 sccm, respectively, (d) On SiC (0001) at Ar flow rates of 100 sccm and 200 sccm, respectively.](image-url)
increased gradually, and the number of graphene layers and grain boundary size decreased. SiC (0001) graphene Raman spectra are shown in figure 4 (d). Similarly, the $I_G/I_D$ was 12.8 and the $I_{2D}/I_G$ was 0.63 as the Ar flow rate was 100 sccm. While the Ar flow rate increased to 200 sccm, $I_G/I_D$ and $I_{2D}/I_G$ values increased to 24.4 and 0.76, respectively, which suggested that the sublimation of Si atoms can be suppressed and the reconstruction rate of graphene can be better controlled by the Ar flow rate. The controllable growth rate can reduce the density of the dislocations and defects, thus the quality of graphene could be improved.

3.4. Controllable growth of graphene on the SiC (0001)

With the further improvement of the Ar flow rate in MPCVD, EG within 3 layers were obtained on the SiC (0001), as shown in figure 5(a). The $I_{2D}/I_G$ was 5.5 and $I_G/I_D$ was 17, which could be identified to be monolayer graphene. According to equation (2), The average grain size of EG was $\sim$326.4 nm for monolayer. For $I_{2D}/I_G = 1.07$ and $I_G/I_D = 86$, it was a bilayer graphene. The calculated average grain size was $\sim$1651.2 nm, which is larger than that of the monolayer. For $I_{2D}/I_G = 0.44$, the number of graphene layers was three or more. The calculated average grain size had little change because of $I_G/I_D = 82$. Figure 5(b) zooms in on the Raman 2D bands in figure 5(a). It can be seen that when the film was monolayer graphene, the 2D band had good symmetry and can be well fitted with a Lorentz single peak with an FWHM of 25 cm$^{-1}$. According to empirical equation (3) [36]:

$$FWHM(2D) = \left[ -45 \left( \frac{1}{N} \right) + 88 \right] [cm^{-1}]$$

where $N$ is the thickness of graphene and can be calculated to be about 1. This further verifies that EG was a monolayer. While the layer number of EG was greater than or equal to two, FWHM did not increase much. The calculated $N$ was close to 1, and the 2D band can also be well fitted with a Lorentz single peak. This could be explained as that the interlayer of the FLG on the SiC (0001) was rotated by an angle. The interlayer coupling was
weak, and its Raman properties were similar to the monolayer. Because of buffer layer free, the sublimation rate and the formation rate of EG were much faster on the SiC (001) than those on SiC (0001). Therefore, it was quite difficult to control graphene layers by the general high-temperature hot-wall CVD system. In the H\textsubscript{2}/Ar plasma of MPCVD, radiative species such as Ar, H\textsubscript{2}, and H are mainly generated by electron collision, as shown below [37]:

$$\begin{align*}
(a) \quad e + Ar & \rightarrow e + Ar^+ , \\
(b) \quad e + H_2 & \rightarrow e + H_2^* , \\
(c) \quad e + H_2 & \rightarrow e + H + H^* , \\
(d) \quad e + H & \rightarrow e + H^* .
\end{align*}$$

The radiative species of atomic H can penetrate the substrate to form a layer with Si, C and H, which contributes to break the Si–C bond at a relatively low temperature [38]. This can generate SiH\textsubscript{x} gas with Si atoms [39] to form a C-rich layer to synthesize graphene. Ar radiative species can suppress the sublimation growth of SiC, and has a certain etching effect on the formed graphene, making the graphene layers more controllable [35, 40]. Our results show that the rate of SiC decomposition and graphene thickness can be controlled by Ar flow without changing the total pressure and substrate temperature, which is consistent with the reported results by D. Momeni Pakdehi et al [41].

Figure 5(c) shows the Raman 2D peak mapping of graphene grown on the SiC (001) under the above experimental conditions, with a range of 15 μm × 15 μm and a step of 0.5 μm × 0.5 μm. The map was reconstructed exactly at the frequency of 1, 2 and more than 3 layers graphene in figure 5(b). It can be seen that the color of the large area in mapping was consistent and identifies a bilayer graphene area.

The transport properties of EG were measured by the Van der Pauw structure device. The device structure is shown in the inset of figure 5(d). The area of graphene was 10 μm × 10 μm. The EG on SiC (0001) and SiC (0001) were measured at 8 points respectively. The maximum room-temperature Hall mobility of EG was 592 cm\textsuperscript{2} V\textsuperscript{−1} s\textsuperscript{−1} on SiC (0001) and 2790 cm\textsuperscript{2} V\textsuperscript{−1} s\textsuperscript{−1} on SiC (0001), which further confirmed that the decoupling of FLG layers on the SiC (0001) with weak interlayer coupling and high mobility have been obtained.

Based on our present results, the differences in the growth mechanisms of EG on SiC (0001) and SiC (0001) were further discussed. On the SiC (0001), graphene nucleated at the edge of the steps grows laterally on the surface layer by layer [12]. Conversely, several layers of graphene nucleated on the platform of SiC (0001) grow in all directions to keep the number of layers stable [35]. The temperature dependence of the surface coverage of graphene on the two faces is also different. In particular, at the temperature corresponding to the initial growth stage, the coverage on the C-face is significantly higher than the coverage on the Si-face. These differences are considered to stem from the reactivity of the faces. C-face is more reactive than Si face [42]. Therefore, on the highly reactive C-face, local decomposition occurs in various parts of the surface, resulting in faster nucleation of graphene.

### 4. Conclusions

In summary, we have developed a pretreatment process on the SiC substrate surfaces by H\textsubscript{2} plasma etching at comparatively low temperature (1000 °C) to promote the nucleation before epitaxial growth on two surfaces of the SiC. Moreover, we found that the Ar plasma in MPCVD can suppress the sublimation of Si atoms resulting in the control of the number of EG layers on SiC (0001). Excitingly, we successfully prepared layer-controllable, high-quality 1–3 layers EG films on SiC (0001) by Ar plasma-assisted MPCVD. The transport properties of EG films on SiC were measured by the Van der Pauw structure device. The EG films on SiC (0001) exhibit high room-temperature mobility up to 2790 cm\textsuperscript{2} V\textsuperscript{−1} s\textsuperscript{−1} which is much higher than that of growth on SiC (0001), which further confirmed the high-quality of EG films on SiC (0001). Furthermore, we also discussed the differences in the growth mechanisms of EG on SiC (0001) and SiC (0001). On the highly reactive C-face, local decomposition occurs in various parts of the surface, resulting in faster nucleation of graphene.

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