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Metal-free synthesis of few-layer graphene films on insulating SiO_2 and SiC substrates by chemical vapor deposition

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Abstract

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Recently, metal-catalyzed chemical vapor deposition (CVD) has attracted intensive interest as a process for preparing single crystal graphene. However, graphene prepared with metal-catalyzed CVD must be transferred onto an insulating substrate in order to be developed into useful electronic devices, which inevitably degrades its quality and introduces contamination. In this regard, we successfully developed an approach of growing few-layer graphene directly on SiO₂ and SiC without metal catalysts, which uniformly covered the entire substrate surface. Under the optimal growth conditions for each case, a faster growth rate was seen for SiC comapring with SiO₂. The surface morphology and crystalline quality of the graphene have been characterized by scanning electron microscopy (SEM), atomic force microscopy (AFM) and Raman spectroscopy, which reveals different growth mechanisms on SiO₂ (SK mode) and SiC (VM mode). Hall bar devices have been fabricated with graphene grown on both SiO₂ and SiC substrates, revealed high Hall mobilities: 608 cm² V⁻¹ s⁻¹ and 1265 cm² V⁻¹ s⁻¹, respectively. These results provide important knowledge on the future application of graphene in the field of microelectronic devices.

1. Introduction

Graphene, a single layer of graphite consisting of carbon atoms arranged in a honeycomb lattice, features a number of exceptional properties, such as high carrier mobility [1], low resistivity [2], excellent thermal conductivity [3], and high transparency for visible light [4]. Such properties make graphene a material with a great potential in the field of nano-electronic devices [5, 6], composite materials [7] and energy storage devices [8]. Meanwhile, several mainstream graphene preparation methods have been developed including mechanical and liquid-phase exfoliations [6, 9], chemical vapor deposition (CVD) [10–12], SiC epitaxial growth [13, 14] and oxidation reduction [15]. Among these methods, metal-catalyzed CVD, particularly with Cu is known for ability of the large scale single crystal graphene production [16], but such graphene eventually needs to be transferred onto an insulating substrate in order to make electronic devices, which inevitably impairs the graphene's crystal quality and physical properties.

Aside from developing transfer techniques [17-21], much effort has been devoted to graphene direct growth on insulating substrates instead in the past several years, which includes SiO₂ [22–24], SiC [25], Al₂O₃ [26, 27], etc. However, due to the absence of metal catalysts, the graphene growth has encountered multiple challenges, such as high nucleation density, slow growth rate, many lattice defects, which unavoidably will lead to the poor electronic performance [22, 23, 28].

Herein, we report the growth method of few-layer graphene without metal catalysts on both SiO₂ and SiC employing the CVD method. By varying gas flow rate, growth temperature and time. The significant differences in graphene film's morphology have been observed. Hall bar devices have been fabricated to investigate the



carrier mobility of the grown graphene films. In short, few-layer graphene films with high substrate coverage on SiO₂ and SiC were achieved at a temperature of 1150 °C, with a H_2/CH_4 flow ratio of 30/1.5 sccm and growth time of 4 h. The growth manner was volmer-weber (3D) mode on SiC comparing to Stranski–Krastanov (2D to 3D) mode on SiO₂ [29]. This growth method on insulating SiC and SiO₂ eliminates the complexity of graphene transfer for further developing electronic devices, which are highly compatible with current large-scale semiconductor fabrication technology.

2. Experimental

2.1. Growth and characterization of graphene on SiO₂ and SiC

Methane (CH₄) was used as the carbon source for direct graphene growth on both SiO₂ and SiC substrates under an atmospheric CVD process. Un-doped 6H-SiC substrate was purchased from TankeBlue Co. (Beijing, China), and then diced into pieces of 3.5×4.5 mm. The growth was performed on the Si-face (0001) of SiC substrate. The highly n-doped (ρ : 0.01–0.05 Ω cm) Si (100) substrate with a 285 nm SiO₂ layer was purchased from Hefei Kejing Materials Technology Co., Ltd (Heifei, China) and then diced into pieces of 10×10 mm. Before the growth of graphene, the substrates were ultrasonically cleaned by a three-step process: (1) 15 min in acetone, (2) 15 min in ethanol, and (3) 15 min in deionized water. After they were dried in a dry N₂ flow then placed on a corundum boat within a quartz tube of 1200 mm $\times \Phi$ 60 mm in the center zone of the CVD furnace as described in figure S1(a) is available online at stacks.jop.org/MRX/6/105604/mmedia.

After loading the substrate, the tube was then pumped down and keep at the pressure below 10 Pa for 20 min by a mechanical pump, and then filled up with a mixture gas of H₂ at 30 sccm flow rate and Ar at 120 sccm flow rate to atmosphere. Then, the substrate was heated up to the growth temperature at a rising rate of 17 °C min⁻¹ with the gas flow continuation. Keeping this temperature to anneal the substrate for 10 min. Then followed by film growth procedure, the CH₄ was sequently injected into the furnace at the rate of 1.5 sccm controlled by a MKS flow meter. After the growth prossess, the sample was cooled down to 400 °C with the rate of 17 °C min⁻¹ and followed by a natural cooling process until to the room temperature. The entire temperature control diagram for the growth is shown in figure 1. In addition, multiple graphene growth following the previously mentioned process have been performed by varying parameters including: growth temperature, CH₄ flow rate, H₂ flow rate and growth time as illustrated in table S1.

The surface morphology of graphene films were characterized with a Park Systems atomic force microscope (AFM) in non-contact mode and a Hitachi SU3500 scanning electron microscope (SEM) under 5 kV beam energy with a secondary electron detector. The thickness and quality of graphene films were characterized by a Renishaw Raman spectrometer using a 532 nm laser beam and 1 μ m spot size under 5 mW laser power. All the Raman spectra of graphene collected on SiC substrates were results of a blank SiC background subtraction.

2.2. Fabrication of Hall bar device and carrier mobility measurements

Hall bar devices were fabricated to evaluate the grown graphene films' electronic properties as shown in figure 2. Firstly, the UV-lithography was used to generate the pattern for the subsequent metal deposition. Secondly, Cr/Au (with a thickness of 30/15 nm) were deposited onto the patterned surface by a e-beam evaporator with a deposition rate of 0.5 Å s⁻¹ in 5.0×10^{-4} Pa vacuum to form the source and drain electrodes. Next, the sample was patterned again using UV-lithography to form the pattern for graphene channel. At last, reactive ion etching (RIE) assisted by oxygen plasma was performed to remove the unwanted graphene other than the channel. The



etching was performed in a Tailong electronics (Beijing, China) RIE-150 system using a continuous 30 sccm O_2 flow rate with 25 W RF power for 25 s. The electronic properties of graphene then were evaluated using a four probe Keithley 2450 system. The schematic of the measurement configuration for field effect mobility and Hall mobility as shown in figures S1(b) and (c).

3. Results and discussion

Four main parameters of graphene growth including the growth temperature, CH_4 flow rate, H_2 flow rate and growth time are studied systematically. The film growth has been performed with temperatures ranging from 1050 °C to 1150 °C under a constant growth time of 5 h, CH_4 flow rate of 1.5 sccm and H_2 flow rate of 30 sccm, the corresponding AFM and SEM images have been shown in figure S2 for SiC and S3 for SiO₂. When the temperature is lower than 1150 °C, only small seeds forming on SiC or SiO₂, which is consistent with the previous results reported by Pang *et al* [22].

Figures S4 and S5 show the AFM and SEM images of graphene growing on SiC and SiO₂ respectively, which are under the growth condition of 1150 °C growth temperature, 5 h growth time, 30 sccm H₂ flow rate, but the CH₄ flow rate is varied from 1.0 sccm to 2.0 sccm. As we can see, when the flow rate is 2.0 sccm, the grown graphene films have very rough surface both for SiC and SiO₂ substrates. However, if the flow rate is lowed to 1.0 sccm, grown graphene patches are still not dense enough to cover the whole substrate, but when we tune the gas flow to 1.5 sccm, for both substrates, they are finally covered by graphene.

In addition, the effect of H_2 flow rate during graphene growth on SiC and SiO₂ has also been studied by AFM and SEM (figures S7 and S8). The growth condition of 1150 °C, 4 h growth time, 1.5 sccm CH₄ flow rate, but the H_2 flow rate is varied from 20 sccm to 40 sccm. By comparison for both SiC and SiO₂ substrates, it can be found that when the flow rate is 20 sccm, the formed graphene films is thicker and its surface is rough., when the flow rate is 30 sccm, graphene films become more smoother on the substrates. However, when the flow rate is 40 sccm, the grown graphene films are not continuous;

By setting the growth temperature constantly at 1150 °C, CH₄ flow rate at 1.5 sccm and H₂ flow rate at 30 sccm, the AFM and SEM images of graphene grown under 2 h, 4 h and 5 h have been shown in figure 3 for SiC and figure 4 for SiO₂, respectively. When the growth time is 2 h, the randomly distributed graphene flakes can be observed in figures 3(a), (d) and (g). In addition, small sharp protuberances are always observed at the centre of each flake. The average flake size is about 500 \pm 30 nm. In those figures, each of them has two pairs of arrows been used for indicating the height differences between the edge of the flake and the substrate (red) and the edge of the flake to the centre (blue). Accordingly, the phase diagram has also been marked for a better demonstration as shown in figure 3(d). The measured thickness of the flake is about 0.707 to 0.771 nm, which approximately equals to the thickness of monolayer graphene as reported [28, 30-32]. However, the small protuberances in the centre of flake is around $11.429 \sim 15.283$ nm, which is a clear indication of Volmer-Weber or 3D island growth. Such difference in growth mode has been observed by Ta et al on copper surface by varying the feedstock ratio between CH_4 and H_2 [29]. When the growth time is extended to 4 h, the morphology of graphene films has been shown in figures 3(b), (e) and (h). To be noticed, both of the darker and lighter regions in the SEM image (figure 3(b)) have been confirmed to be covered by multi-layer and single layer graphene from the subsequent Raman spectroscopy measurements as shown in figure 5(a). When the growth time is increased to 5 h, the surface morphology remains similar to the 4 h, but with more multi-layer films coverage as shown in figures 3(c), (f) and (i).

In comparison, the AFM and SEM images of graphene grown on SiO_2 under 2 h, 4 h and 5 h have been shown in figure 4. When the growth time is 2 h, similar as SiC, graphene flakes also randomly disperse across the substrate but with more irregular geometries. When the growth time is extended to 4 h, the graphene flakes have expended and started to merge, while the substrate still has not been fully covered yet. More noticeably, the height distribution of the merged flakes is more evenly distributed without clear 3D islands structures





comparing to the ones grown on SiC (figure 3(d)). The height difference is measured at upper and lower boundaries of the gaps across the film showing the film thickness is about 0.552 to 0.645 nm, which also falls into the single layer graphene range [31]. After increasing the growth time to 5 h, a full coverage of the substrate is eventually achieved with also multi-layer flakes forming at the top as shown in figures 4(c), (f) and (i). It's also more obvious from the AFM phase diagram like that on SiC. Therefore, the growth mechanism on SiO₂ has been determined to be the regime of 2D to 3D or Stranski–Krastanov growth [29]. However, in our work, since the growth conditions have been set exactly the same on each substrate, the difference in graphene growth mechanisms are very likely controlled by the different intrinsic surface properties of the two substrates [28].

To evaluate the quality of graphene films and the film-substrate interactions, Raman spectroscopy measurements were performed. The Raman spectra of graphene mainly consist three peaks, D, G and 2D, they are at the position of 1350 cm⁻¹, 1580 cm⁻¹ and 2700 cm⁻¹, respectively [33]. In addition, the values of I_G/I_D (the intensity ratio between G peak and D peak) and I_{2D}/I_G (the intensity ratio between 2D peak and G peak) were calculated to evaluate the defects level of graphene films and the number of graphene layers [34–36].

In the similar order as the previous AFM and SEM characterizations, Raman spectra of graphene on SiC and SiO₂ substrates under various growth temperatures are shown in figures S6(a) and (b). When the growth temperature is 1050 °C, none of three peaks are observed on either SiC or SiO₂ substrates. When the growth temperature is raised up to 1100 °C, D, G and 2D peaks stand out but rather weak for SiC. However, they are much more observable on SiO₂. When the temperature is increased to 1150 °C, strong D, G and 2D peaks are on the spectrum of graphene from both substrates. Meanwhile, the G peaks of graphene grown on both the SiC and SiO₂ substrates are blue-shifted (~8 cm⁻¹ for SiC and ~11 cm⁻¹ for SiO₂) with respect to graphene which are transferred onto SiO₂/Si substrates but grown on copper foil substrate. This indicates a strong compressive strain of the graphene due to the strong interaction between the graphene and underlying substrates [35, 37, 38]. However, for both substrates, the graphene is well ordered in terms of structure [35].





The Raman spectra of graphene on SiC and SiO₂ substrates which grown under various CH₄ flow rates have been shown in figures S6(c) and (d). In figure S6(f) we can see, when the CH₄ flow rate is 1.5 sccm, the Raman spectra of graphene grown on SiC have a sharp symmetric 2D peak with the full width half maximum (FWHM) of 59 cm⁻¹, which indicates a perfect bilayer graphene [35]. The D peak is always rather high for both the high or low flow rate. For a better demonstration, the trend of I_{2D}/I_G and I_G/I_D rations have been shown in figure S6(e). It shows that the I_{2D}/I_G ratio is obviously lower for the graphene on SiC than that of SiO₂ during the initial film growth, which indicates a better wetting surface of graphene on SiO₂ comparing to SiC. Besides, the largest value of I_G/I_D is observed in graphene films on SiC and SiO₂ both with a CH₄ flow rate of 1.5 sccm. Moreover, the FWHM of the grown graphene films under various CH₄ flow rates have been shown in figure S6(f). For SiO₂, all of the FWHM values are over 60 cm⁻¹ and increase linearly with the increase of the flow rate, it indicates that the films are majorly multilayer under such conditions. For SiC, a lowest FWHM is observed when the CH₄ flow rate is 1.5 sccm, it is the suitable condition for few-layer graphene growth.

The Raman spectra of graphene on SiC and SiO₂ substrates which grown under various H₂ flow rates have been shown in figures S9(a) and (b). The trend of I_{2D}/I_G , I_G/I_D ratios and the FWHM of 2D peak have been shown in figures S9(c) and (d) respectively. It can be seen that the largest value of I_{2D}/I_G is observed in graphene films on SiC and SiO₂ both with the H₂ flow rate of 30 sccm. Moreover, a lowest FWHM is observed when the H₂ flow rate is 30 sccm for SiO₂, while for SiC is observed at 40 sccm H₂ flow rate but the films have not covered the SiC substrate yet. When the H₂ flow rate is 30 sccm, a continuous few-layer graphene films are formed on SiC substrate. Thus, the suitable H₂ flow rate is 30 sccm.

Raman spectra of graphene on SiC and SiO₂ substrates under various growth times are shown in figures 5(a) and (b). D, G and 2D peaks all appear after 2 h of growth on SiO₂, and all the peaks are stronger than those growth on SiC substrate. When the growth time is extended over 4 h, the stronger graphene characteristic D, G, and 2D peaks appeared both on SiC and SiO₂. A sharp symmetric 2D peak with a small FWHM of 41 cm⁻¹ as shown in figure 5(d) and the largest value of I_{2D}/I_G is observed from the graphene grown on SiO₂ substrate as shown in figure 5(c). This indicates a single layer graphene growth under such condition [34, 35]. Additionally,



Figure 5. (a), (b) The Raman spectra of graphene on SiC and SiO₂ substrates growth at 1150 °C with 1.5 sccm CH₄ flow rate under various growth times: 2 h, 4 h and 5 h, respectively. (c) The Raman peak intensity ratio I_{2D}/I_G , I_G/I_D as a function of growth time. (d) The FWHM of graphene 2D peaks as a function of growth time on SiC and SiO₂ substrates.

the FWHM of graphene grown on SiC is always higher than that of SiO₂ that reveals a smoother growth mode upon SiO₂ as inferred from the previous AFM and SEM data. Moreover, the G peaks of all graphene grown on the SiC and SiO₂ substrates are blue-shifted, it is about 20 cm⁻¹ for SiO₂ and 10 cm⁻¹ for SiC, which indicates the stronger interaction between graphene with SiO₂ than that with SiC. Such phenomena are presumably resulted from the higher electronegativity of O than C, which leads to a stronger interaction with C atoms on SiO₂ than SiC under the same graphene. In this way, we eventually discovered an optimal condition to grow 2D graphene films with few-layer directly on SiO₂ and SiC substrates.

In order to further evaluate the electronic performance of the directly grown graphene films, the Hall bar devices were fabricated. As the figure 6(a) shown, a typical I–V characteristic for the Hall bar device on SiO₂ with a channel length (L) of ~100 μ m and a channel width (W) of ~10 μ m was measured. The linear gating behaviour indicates the typical unipolarity property [6]. The transfer curves of positive and negative scan of the device are shown in figure 6(b), here the voltage across the source and drain was set to 0.5 V, the measured drain-source current (I_{DS}) value decreases first and then increases with the graphene charge neutrality point (CNP) at about 75 V, which indicates a highly p-type doped behaviour [6]. In an ideal case, the CNP of graphene locates at zero back gate voltage, but the obvious p-doping and hysteresis effects are observed in our case as shown in figure 6(b), which is not a surprise for graphene under ambient conditions [39]. Usually, the external doping to graphene is considered to be responsible for the hysteresis behaviour, which can be explained by the existence of charge traps and movable charges coupling with the graphene carriers [40]. It is mainly attributed to effect of O₂, H₂O and the residue of photoresist on the graphene during the device fabrication [41–43]. From the transfer curve as shown in figure 6(b), based on field effect mobility formula:

$$\mu_{FE} = \frac{\partial \sigma}{\partial ne} = \frac{\partial I_{DS}}{\partial V_{GS}} \frac{L}{WC_{ox} V_{DS}}$$
(1)

where C_{OX} is the gate oxide capacitance, the hole and electron field effect mobilities are extracted from the maximum value of the transconductance of the Hall bar as about 209 cm² V⁻¹ s⁻¹ and 104 cm² V⁻¹ s⁻¹ respectively in air. These values fall in the middle range of previously reported results [26–28, 32, 44, 45]. Du *et al* reported the growth of graphene on non-catalytic substrates with a flake size of around 21 nm and showing a hole field effect mobility of 147 cm² V⁻¹ s⁻¹ [44], our graphene mobility is larger because of the much larger single crystal flake, the size reaches about 500 nm. It significantly reduces the carrier scattering. However, Chen *et al* reported a hole and electron field effect mobility of 1518 cm² V⁻¹ s⁻¹ and 478 cm² V⁻¹ s⁻¹ respectively for a more continuous and smoother graphene films in air [28]. Subsequently, they also reported a larger graphene single crystal with the growth time of 72 h and the growth temperature of 1180 °C. Since there is no grain



Figure 6. (a) Output curves of the Hall bar device fabricated with 10 μ m channel width and 100 μ m channel length, respectively. The inset OM image shows this Hall bar device. (b) Transfer curves of positive and negative positive and negative scan of the device at $V_{DS} = 0.5 \text{ V}$. (c) V_{H} -B curve for the Hall bar device, the magnetic field intensity B was varied from 50 to 150 G. (d) Comparison of transfer curves of before and after aluminium deposition. All the measurement in air at room temperature.



Figure 7. (a) I–V curve for the Hall bar device with a channel length of 160 μ m and channel width of 5 μ m, the inset SEM image shows this Hall bar device. (b) V_H-B curve for a Hall bar device, the magnetic field intensity B was varied from 0 to 100 G. All the measurement in air at room temperature also.

boundary scattering in graphene films, the field effect mobility is over 5000 cm² V⁻¹ s⁻¹ [24]. Figure 6(c) shows the V_H-B curve for the Hall bar device, where the magnetic field intensity (B) varied from 50 to 150 G, for which a linear relationship between Hall voltage (V_H) and B has been observed. According to the following equations:

$$n_{s} = \left[|e| \frac{d\rho_{xy}}{dB} \right]^{-1} = \frac{I/|e|}{dV_{H}/dB} \quad \mu_{H} = \frac{1}{|e|} \frac{1}{n_{s}\rho_{xx}} = \frac{I/|e|}{n_{s}V_{x}W/L}$$
(2)

the carrier concentration n_s and Hall mobility μ_H were calculated as: $n_s = 2.86 \times 10^{12}$ /cm² and μ_H = 608cm²V⁻¹ s⁻¹, which is larger than their field effect mobility reported previously [45]. In order to test the tunability of the CNP in graphene films, figure 6(d) shows the comparison of transfer curves before and after the deposition of 5 nm aluminium films. We observed that the CNP was shifted from 75 V to 10 V, this has been previously predicted by Khomyakov *et al* [46]. They found that although the surface bonds between graphene and Al, Ag, Cu, Au and Pt are very week, but the electronic structure of graphene will be strongly disturbed by the physical adsorption. A slightly different device was fabricated on the graphene/SiC sample for its diminished substrate size. Figure 7(a) shows the I–V curves for the Hall bar device with a channel length (L) of ~160 μ m and a channel width (W) of ~5 μ m, the two wire and four wire resistances were measured. The four-wire resistance value is smaller as a reason of avoiding contact resistance between the probe and electrode. Figure 7(b) shows the V_H-B curve for the Hall bar device where the magnetic field intensity B was varied from 0 to 100 G, for which a linear relationship between V_H and B has been observed. Thus, according to the Hall mobility formula (2), the carrier concentration n_s and Hall mobility μ_H were calculated as: $n_s = 5.87 \times 10^{12}/\text{cm}^2$ and $\mu_H = 1265 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Compared with the Hall mobility on SiO₂ substrate, it is much larger, possibly because of the smaller channel size [47] and the continuity of graphene films on SiC than SiO₂.

4. Conclusion

To sum up, we have developed an effective metal-free CVD method recipe for growing few-layer graphene on dielectric substrates. CH_4 was chosen as the carbon source, the graphene both on SiO₂ and SiC substrates were successfully grown by an atmospheric CVD process without any metal catalyst. With SEM, AFM and Raman spectroscopy, the structure and surface morphology of the graphene films were characterized, and two different growth modes on SiO₂ and SiC under the same growth temperature of 1150 °C, H₂/CH₄ flow ratio of 30/1.5 sccm and growth time of 4 h were revealed, the growth was determined to be 2D to 3D growth on SiO₂ comparing to 3D growth on SiC and a faster growth rate was seen for SiC comapring to SiO₂. The Hall mobility was measured about 608 cm² V⁻¹ s⁻¹ for the Hall bar device on SiO₂ and about 1265 cm² V⁻¹ s⁻¹ on SiC. These results have proved that few-layer graphene films can directly grow on insulating SiC and SiO₂ substrates without any metal catalysts. Although the graphene samples here grown are only on the size of the substrate but it is can be easily scaled up by simply use larger sized substrate and proper sized CVD furnace accordingly as which are commonly practiced in modern industry. Therefore, our recipe indicates a great potential application for future scalable graphene-based electronic devices fabrication.

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