Uniform Epitaxial Growth of Graphene for High-Frequency Transistors in the Terahertz Band

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The authors have developed a novel method for the fabrication of uniform epitaxial graphene on SiC substrates. Graphene was grown on the C-face 6H-SiC substrates with a sputtered SiC film by annealing at high temperatures. Observation using Raman mapping and low-energy electron micron microscopy revealed that 95% of this graphene consisted of two layers on the fractional area in a 75×75 μm square. This uniformity is quite high compared to that of graphene grown with the conventional method without sputtered SiC films, indicating the new method is more likely to be suitable for the fabrication of high frequency wireless devices.

Keywords: Graphene, SiC, Terahertz, Sputtered film

1. Introduction

The capacity of wireless communication has been increasing drastically, and a transmission rate of 100 Gbit/s will be necessary several years from now. Wireless communication requires high-frequency devices and transistors that work in the terahertz band will be needed to realize these transmission rates.

Graphene attracts attention as one of the candidates for the materials of these wireless communication devices. Graphene has extremely high mobility around 200,000 cm^2/Vs, much higher than that of known semiconductors, due to its anomalous two-dimensional carbon honeycomb structure.(1)-(6) It is therefore expected as an ideal material for high frequency analog devices used in terahertz wireless communications where high mobility of more than 10,000 cm^2/Vs is required.

A number of early fabrication methods for graphene were developed as typified by the well-known mechanical exfoliation method.(1) However, most methods have problems in industrial use due to the number of processes and the difficulty of the transfer process prerequisite to these methods. Epitaxial graphene (EG), which is fabricated by thermal decomposition of SiC substrates, has attracted recent attention as a candidate for electronic devices because it requires no transfer process. Characteristics differ greatly when EG is grown on the SiC (000-1) (the C-face*1) or the SiC (0001) (the Si-face*2). Carrier mobility of EG on the SiC C-face is superior to that of EG on the SiC Si-face because there is no buffer layer to degrade carrier mobility.(7) EG on the SiC C-face, however, has a drawback in the on-wafer uniformity of the distribution of the number of layers due to high decomposition activity. To compensate for this drawback, we have developed a novel method to use a different film of SiC as the raw material for the formation of graphene. This novel method enables the fabrication of uniform epitaxial graphene on the SiC C-face of substrates by using a sputtered SiC film on the substrate. This paper demonstrates that graphene from SiC sputtered film has superior uniformity in the number of graphene layers.

2. The Fabrication and Characterization of Graphene

The graphitization was conducted at temperatures ranging from 1400°C to 1900°C in 1 to 30 minutes under Ar ambient at atmosphere pressure by high-frequency induction heating. To compare the quality, graphene was grown on the C-face of semi-insulating 6H-SiC substrates with and without the sputtered SiC film. The SiC film was deposited by RF magnetron sputtering.

The graphene was characterized by using Raman scattering, atomic force microscopy (AFM), optical microscopy, transmission electron microscopy (TEM), low energy electron microscopy (LEEM), and low energy electron diffraction (LEED). Carrier mobility of the graphene was measured using the Hall effect.

3. Graphene Formation Using Sputtered SiC Film

3-1 The quality of crystal structures of graphene made using sputtered SiC film

First, we confirmed the formation of graphene with and without sputtered SiC film on the SiC C-face using Raman scattering and TEM. Figure 1 shows Raman scattering spectra of samples with and without the sputtering SiC film. Distinct G and G' peaks that indicate a high quality of crystal structure, were confirmed in both samples. TEM images from both samples (Fig. 2) showed formation of 2 or 3 layers of graphene. These results demonstrate the formation of high quality of crystal structure of graphene on sputtered SiC film on the SiC C-face.

3-2 Dependence of the distribution of the number of layers on the thickness of sputtered SiC film

In order to investigate the dependence of the distribution of the number of layers on the thickness of sputtered SiC film, we used SiC films ranging in thickness from 1 nm to 5 nm. Figure 3 shows the optical microscopy images of the samples with sputtered SiC film. An image without the sputtered SiC film is also shown for comparison. The

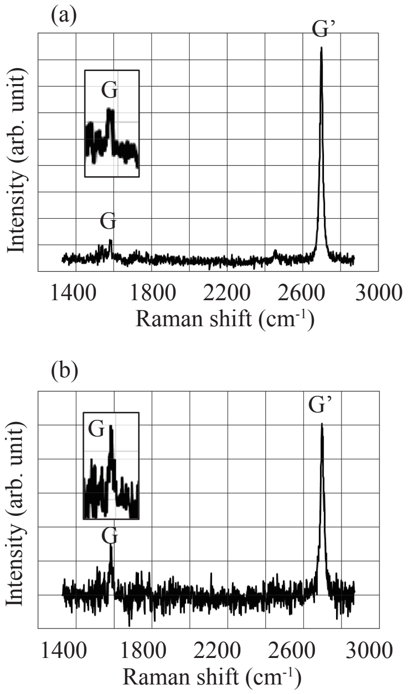

Fig. 1. Raman scattering spectra of graphene fabricated from SiC C-face substrates (a) without and (b) with sputtered SiC films

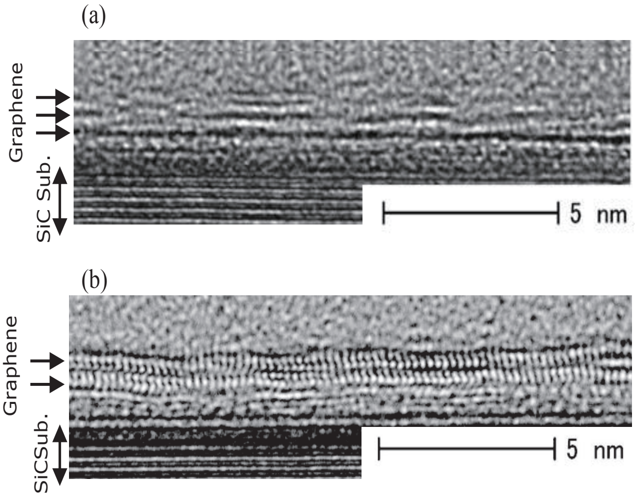

Fig. 2. Cross-sectional TEM images of graphene fabricated from SiC C-face substrates (a) without and (b) with sputtered SiC films

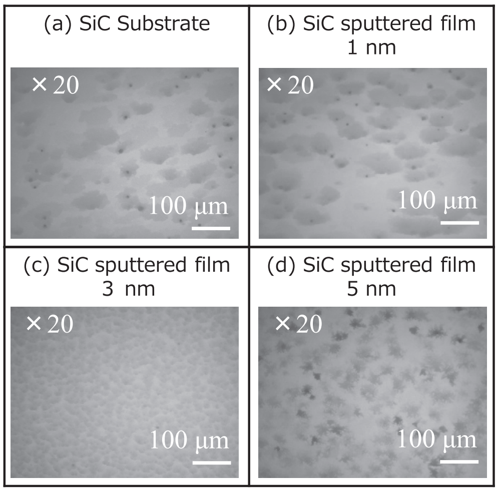

Fig. 3. Optical microscopy images of samples fabricated from SiC C-face substrates (a) without and with sputtered SiC films of (b) 1 nm, (c) 3 nm, and (d) 5 nm

optical microscopy images were taken in optical transmission mode, so that the contrast of the images indicates the difference in the number of layers of graphene. In Fig. 3, large contrast fluctuations are observed in the samples without the sputtered SiC film and those with the sputtered 1 nm and 5 nm thick SiC film. Only the graphene fabricated on a sputtered SiC film with a thickness of 3 nm exhibits remarkably less contrast, indicating its high uniformity in distribution of layer numbers. The graphene formed on a 1 nm sputtered SiC film showed a similar image to that of conventional SiC substrates. We attribute this to insufficient coverage of the SiC film over the SiC substrate. The graphene formed on a 5 nm sputtered SiC film, on the other hand, showed a larger contrast fluctuation, which is attributed to the SiC film being too thick to allow the growth of epitaxial graphene. Consequently, we conclude that the optimal thickness for sputtered SiC film is around 3 nm.

3-3 Uniformity of graphene

The distribution of the number of layers was quantitatively characterized by Raman scattering mapping, LEED, and LEEM. Figure 4 shows the LEED pattern of the graphene formed on sputtered SiC films. The LEED pattern exhibits a single set of hexagonal spots, which indicates that the direction of the crystals of the multiple layer numbers of graphene is identical, i.e. graphene is stacked Bernally.*3 Usually, graphene fabricated on the SiC C-face is stacked with a randomly rotating crystal orientation, i.e. graphene is stacked non-Bernally. The graphene formed on SiC sputtered film exhibits a uniform Bernal stacking. Figure 5 shows the Raman mappings of the distribution of the G' peak intensity in a 75 μm square obtained from the graphene grown on the SiC C-face substrate without sputtered film (a) and with a 3 nm sputtered SiC film (b). Assuming the intensity of the G' peaks indicates the layer number and the quality of crystal structure of graphene, the

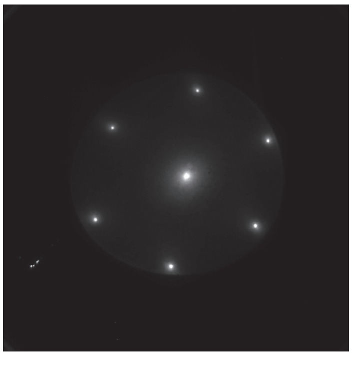

Fig. 4. LEED pattern of graphene from the sputtered 3 nm SiC film on SiC C-face substrate

distribution of the number of layers in the graphene fabricated on the sputtered SiC film is much smaller than that fabricated without SiC film. The area ratio of graphene with the same layer numbers is 95% for the graphene fabricated on the sputtered SiC film calculated from the fractional ratio of distribution of G’ intensity. Figure 6 shows an LEEM image of the graphene formed on the sputtered SiC film. In LEEM, we can obtain the layer number of graphene from the reflection spectrum of each observation point exactly and the distribution of the number of layers as an observable image. As shown in Fig. 6 (a), there are two distinctive areas in the LEEM image; the dark and the bright areas. The spectrum of the LEEM from the small dark area shows one dip (Fig. 6 (b)) and suggests the presence of one-layer graphene.(8) The spectrum of the LEEM from the large bright area shows a pair of dips (Fig. 6 (c)), suggesting the presence of two-layer graphene. Two-layer graphene is mainly observed, occupying an area of 75 μm in diameter while one-layer graphene is scattered in Fig. 6 (a). These results have shown that graphene fabricated from sputtered 3 nm SiC film exhibits extremely uniform distribution of the number of layers.

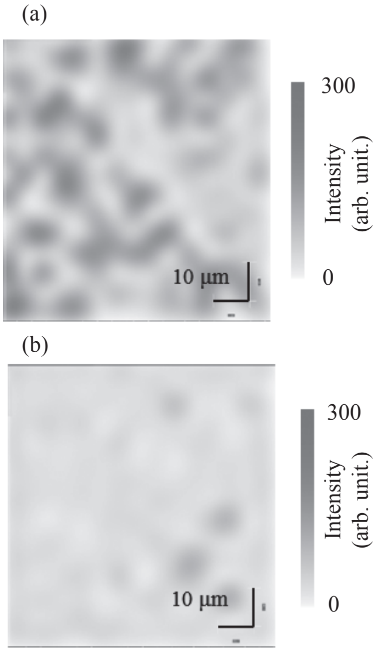

Fig. 5. Raman scattering mapping images of G’ peak intensity of graphene fabricated from the (a) SiC C-face substrate and (b) the sputtered 3 nm SiC film

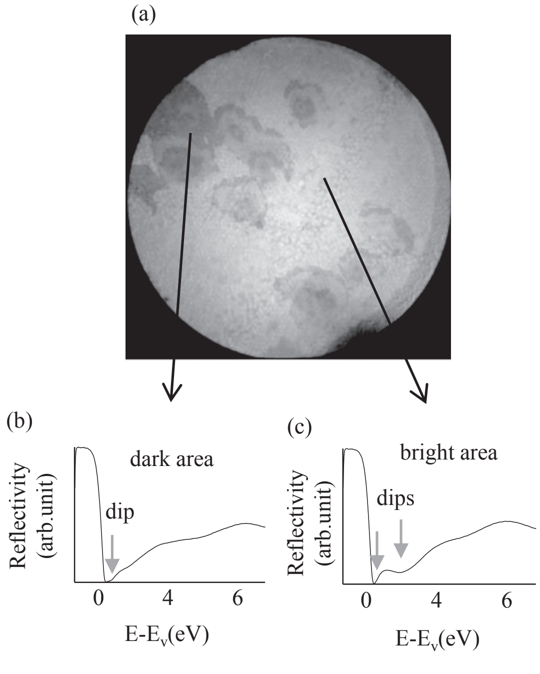

Fig. 6. (a) LEEM image and spectra of (b) bright area and (c) dark area from the 3 nm sputtered SiC film

3-4 The electrical characteristics of graphene

The carrier mobility of graphene fabricated on sputtered SiC film was measured using the Hall effect. Carrier type, carrier concentration, and carrier mobility were determined to be n-type, 1.5×10^{10} cm^{-2}, and 95,000 cm^2/Vs, respectively. The carrier mobility is the highest ever reported for epitaxial graphene.(9) This high mobility is obtained due to remarkably low carrier concentration. Covering the surface with sputtered SiC film presumably inhibited carrier doping from SiC substrates.

4. The Mechanism for the Formation of Graphene

Finally, we briefly discuss the formation mechanism for graphene on sputtered SiC film on the SiC C-face of substrates. Figure 7 compares the AFM images of graphene that formed on conventional SiC C-face substrate (a) to

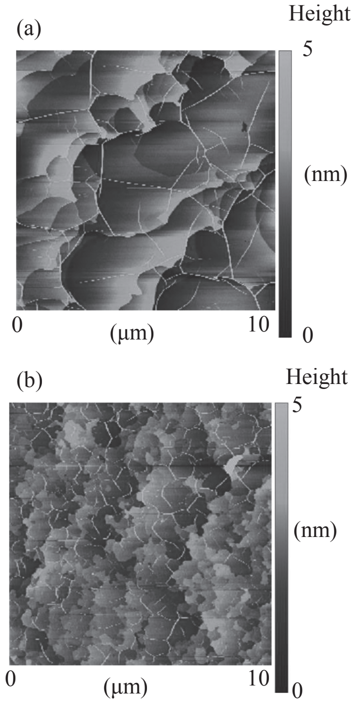

Fig. 7. AFM images of graphene from (a) a conventional SiC C-face substrate and (b) the sputtered 3 nm SiC film on a SiC C-face substrate

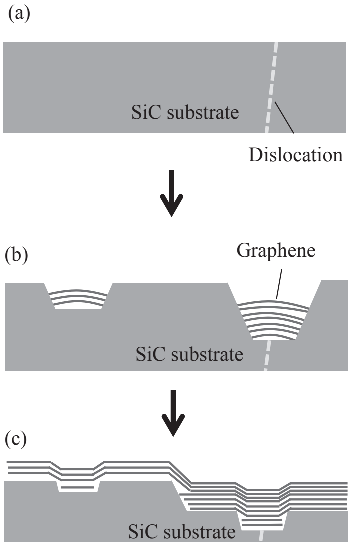

Fig. 8. Formation mechanism models of graphene on SiC C-face substrates

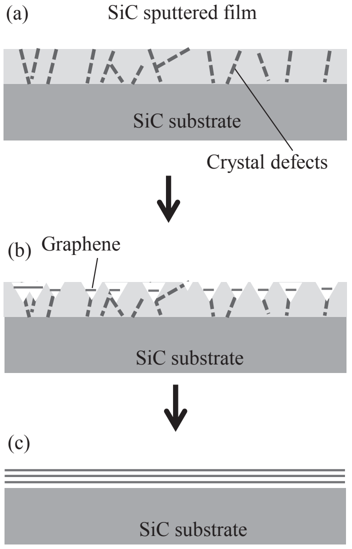

Fig. 9. Formation mechanism models of graphene on sputtered SiC films

that of graphene formed on sputtered 3 nm SiC film on the SiC C-face of the substrate (b). Steps, terraces, and fine-net-like structures are observed in the surface morphology of both samples. Their sizes, however, are different. While, 1-5 µm is the typical size for the structures in (a), it is greatly reduced to 0.5-1 µm in (b). Based on these results, we propose a model for the formation mechanism of graphene in Figs. 8 and 9. On a conventional SiC C-face, the origins of graphene are not well defined and are randomly distributed (Fig. 8 (a)).(7) Differences in the decomposition speed can occur due to a dispersion in SiC crystallinity during graphene formation (Fig. 8 (b)), and give rise to the onset of localized multilayer graphene (Fig. 8 (c)). In contrast, Fig. 9 shows the formation of graphene on SiC sputtered film. In the case of graphitization on sputtered SiC film, small steps and terraces of graphene cover the surface in a short time due to the presence of many defects on the surface of sputtered SiC film (Fig. 9 (a) and (b)). These defects become the origins of graphene (Fig. 9 (b)). This may in consequence lead to less distribution of the layer number over a large area (Fig. 9 (c)) through optimizing the thickness with sputtered SiC film.

5. Conclusion

A novel method to fabricate highly uniform epitaxial graphene on SiC C-face substrates has been demonstrated by using sputtered SiC film. The optimum thickness for the sputtered SiC film was 3 nm, on which the fractional area of two-layer graphene was about 95% in a 75 × 75 µm square. A carrier mobility of 95,000 cm^2/Vs was obtained. This two-layer graphene was considered to be suitable for the creation of transistors as it poses a bandgap structure.(10) We conclude that this novel method is promising for manufacturing analog high frequency devices.

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Technical Terms

∗1 C-face: The crystal structure of SiC substrates that has polarity. The C-face indicates (000-1) structure.

∗2 Si-face: The crystal structure of SiC substrates that has polarity. The Si-face indicates (0001) structure.

∗3 Bernally stacking: The stacking structure of graphene that indicates the {ABAB….} stacking type.

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