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Growth of Graphene/3C-SiC(111)/4H-SiC(0001) by a Sublimation Technique with a Vertical Infrared-Lamp Annealer

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Abstract

We demonstrate simple sublimation growth of 3C-SiC(111) on a 4H-SiC(0001) substrate with a vertical infrared-lamp annealer. Furthermore, using the same annealer, high quality graphene has been grown on the 3C-SiC/4H-SiC by the conventional Si-sublimation technique. The grown 3C-SiC has been confirmed by transmission electron microscopy and Raman scattering spectroscopy. A clear 2D peak of graphene/3C-SiC/4H-SiC has been also observed by Raman scattering spectroscopy. This simple method paves the way to study unique properties of graphene/3C-SiC, such as electrical doping and homogeneity of the number of graphene layers, which depend on the polytype of the SiC substrate.

1. Introduction

The properties of graphene grown on a SiC substrate by the Si-sublimation technique [1] are affected by the SiC substrate, that is to say properties such as electrical doping, surface morphology, and layer number [2-4]. Among many polytypes of SiC with crystal structures such as hexagonal, cubic, and rhombohedral structures, 3C-SiC has a unique property of negligible spontaneous electrical polarization due to its cubic crystal structure [3]. The polarization of a substrate such as 4H- and 6H-SiC with the hexagonal crystal structure induces doping of graphene, which shifts a Fermi level far from the Dirac point [2,3]. Furthermore, 3C-SiC has only one Si-sublimation energy because it consists of only one type of SiC stacking layer. On the other hand, 4H- and 6H-SiC have two and three energies due to those SiC stacking sequences, respectively [5]. To obtain a homogeneous layer number of graphene, one Si-sublimation energy may be favorable because uniform Si sublimation from each SiC stacking layer is expected. However, a high quality 3C-SiC substrate is not commercially available [6]. This hinders from investigating the properties of graphene grown on 3C-SiC, while intensive studies of graphene on 4H- and 6H-SiC have been reported. Here, we demonstrate simple sublimation growth of 3C-SiC(111) on 4H-SiC(0001), and further growth of graphene on 3C-SiC/4H-SiC using an infrared-lamp annealer.

2. Growth method

A vertical infrared-lamp annealer shown in Fig. 1(a) was used to grow 3C-SiC or graphene. A sample is heated by an infrared lamp. The sample is in a quartz tube, where the gas

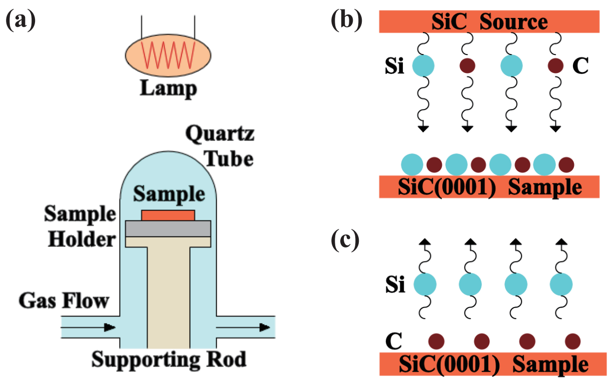

Fig. 1 (a) Schematics of infrared-lamp annealer. (b) Schematic growth mechanism of 3C-SiC, and (c) that of graphene. In 3C-SiC growth, both Si and C sublimate from the source, while only Si sublimates in graphene growth.

pressure is controlled. The sample temperature was monitored with a radiation thermometer.

In the first growth process of 3C-SiC, source and sample SiC were used. A possible growth mechanism is illustrated in Fig. 1(b). Sublimated Si and C from the source attach to the sample SiC. The growth of 3C-SiC was carried out at 1660ºC for 2 - 10 min in an Ar atmosphere of 100 Torr. It is reported that 3C-SiC grows at lower temperatures compared with 4H-, or 6H-SiC [7]. We confirmed the thickness of 3C-SiC increases with increasing the annealing time.

In the second growth process of graphene on 3C-SiC/4H-SiC, the conventional Si-sublimation technique [1] shown in Fig. 1(c) was applied. Only Si sublimates and C remains on the surface. The second growth was carried out at 1620ºC for 5 min in an Ar atmosphere of 100 Torr.

3. Experimental results and discussions

We confirmed that the grown layer on 4H-SiC in the first process is 3C-SiC by atomic force microscopy (AFM), scanning electron microscopy (SEM), transmission electron microscopy (TEM), and Raman scattering spectroscopy. In Fig 2(a), a smooth surface of a 5-µm-square AFM image of the grown layer on 4H-SiC(0001) is shown. The root-mean-square roughness is 0.13 nm. The step height is 0.25 nm, which corresponds to one-layer SiC stacking. In a cross-sectional SEM image of Fig. 2(b), a clear boundary between the grown layer and the 4H-SiC substrate is observed in a 5-min-annealed sample. The SEM contrast is probably caused by the difference of work functions. No double-posi-

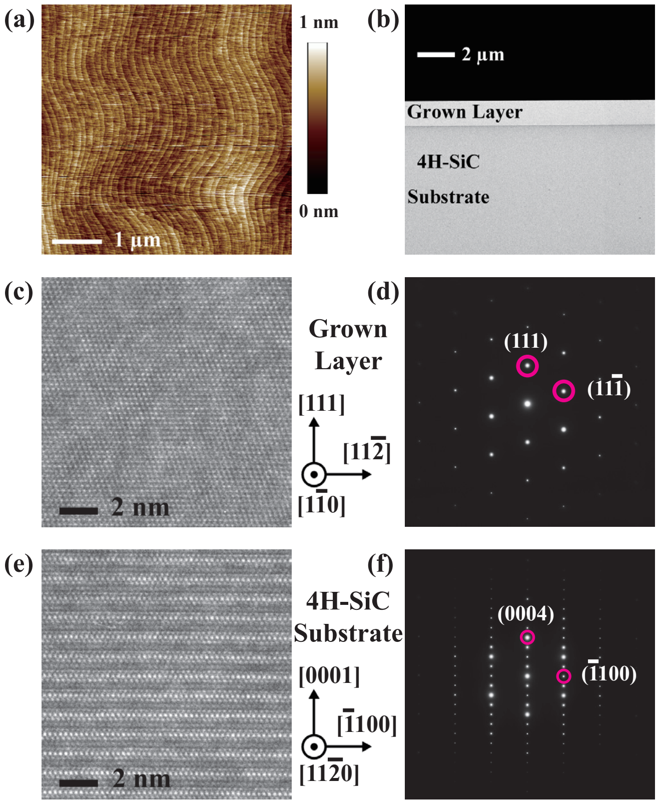

Fig. 2 (a) AFM image of grown layer in the first process. (b) Cross-sectional SEM image of grown layer on the 4H-SiC substrate. The thickness of grown layer is 1.3 μm. TEM lattice and diffraction images of (c) and (d) grown layer, and (e) and (f) 4H-SiC, respectively. Three-layer of SiC stacking and cubic structures are observed in grown layer.

tioning boundary, which is caused by inequivalent structures of 3C-SiC rotated by 60° on the hexagonal structure [8], is observed over 10 μm. Cross-sectional TEM lattice and diffraction images of the grown layer are displayed in Fig. 2(c) and (d), and those of the 4H-SiC substrate are shown in Fig. 2(e) and (f), respectively. In Fig. 2(c) and (d), three-layer SiC stacking and cubic structures are observed, so we verified that the grown layer is 3C-SiC(111). Assigned crystal axes and plane indices are also displayed. As references, four-layer stacking and hexagonal structures of 4H-SiC are shown in Fig. 2(e) and (f), with assigned crystal axes and plane indices. We concluded that 10-μm-large high-quality 3C-SiC/4H-SiC can be grown by our method, which is verified from the small surface roughness, the clear lattice image, and the distinct diffraction spot.

By Raman spectroscopy, different polytypes of SiC can be discriminated from peak positions of transverse optical (TO) and longitudinal optical (LO) phonon modes [9]. Figure 3(a) shows a Raman spectrum of the grown sample in the first process. Peaks of 797 and 976 cm^{-1} are assigned to TO and LO phonon modes of 3C-SiC, respectively. Peaks of 777 and 965 cm^{-1} are assigned to TO and LO phonon modes of the substrate 4H-SiC. We also confirmed homogeneity of the grown sample from a Raman mapping of the ratio between 3C-SiC and 4C-SiC peaks.

Graphene grown on 3C-SiC/4H-SiC is also verified by

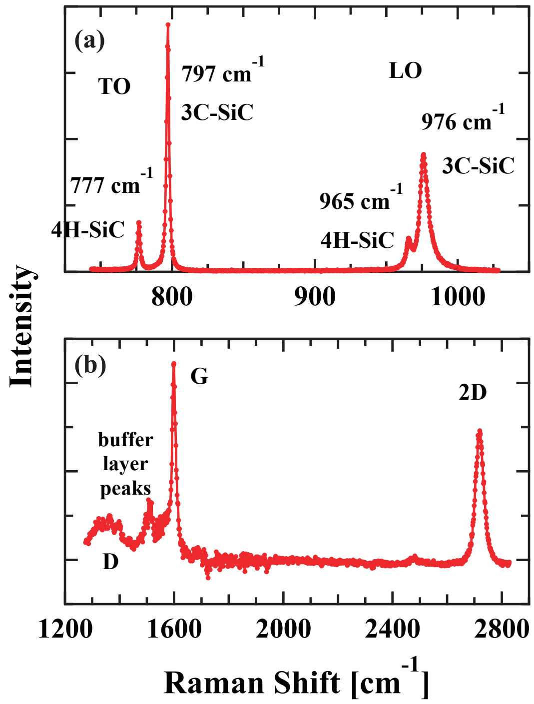

Fig. 3 (a) Raman spectrum of grown sample in the first process. (b) Spectrum of grown sample in the second process. Clear 2D peak is observed. Background of SiC signal is subtracted.

Raman spectroscopy. A clear 2D peak with a full width of half maximum of 35 cm^{-1}, which is a typical value of monolayer graphene, is observed in Fig. 3(b). Peaks of buffer layer, which is a non-conductive carbon layer located between graphene and the SiC substrate, appear in the range of graphene D and G peaks [10]. From Lorentzian curve fitting, it was found that the observed peaks around D are mostly contributed to the buffer layer peaks. So the contribution of graphene D peak is not so large, which indicates our graphene sample quality is good.

4. Summary

By the sublimation technique using the source and the sample SiC, 3C-SiC(111) on the 4H-SiC(0001) substrate were grown and verified by AFM, SEM, TEM, and Raman spectroscopy. After the growth of 3C-SiC, high quality graphene on the 3C-SiC/4H-SiC was grown by the conventional Si-sublimation technique. Our simple growth method is useful for investigating the unique graphene/3C-SiC properties.

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