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Hexagonal Boron Nitride Heteroepitaxial Layers on Graphitized 6H-SiC Substrate Grown by Metalorganic Vapor Phase Epitaxy

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1. Introduction

Hexagonal boron nitride (h-BN) has potential for optical device applications in the ultraviolet spectral region and as an exciton-based quantum information processing material because of its wide bandgap and large exciton binding energies of 5.97 eV and 149 meV, respectively [1]. During the past few decades, the lack of bulk h-BN substrate has made it difficult to obtain single-phase h-BN epitaxial layers. Graphite is one of the attractive substrates for h-BN heteroepitaxial growth because it has almost the same crystal structure and relatively small lattice mismatch (1.6%). However, only expensive highly oriented pyrolytic graphite is available. An alternative to graphite is single crystal 6H-SiC substrates, which are commercially available and have advantages of a high melting point, a large diameter, cleavability, and a reasonable price. However, the large lattice-mismatch (19%) between the 6H-SiC and h-BN films has hindered a high-quality heteroepitaxial h-BN growth on 6H-SiC substrate [2]. Here, we propose introducing graphitized 6H-SiC substrate for h-BN heteroepitaxial growth, in which the surface is covered with a layer of graphite having several monolayers thickness, to overcome the obstacle of the large lattice mismatch. Our approach enables us to grow single-phase h-BN heteroepitaxial layers on graphitized 6H-SiC substrate by metalorganic vapor phase epitaxy (MOVPE) for the first time.

2. Experiment

First, (0001) 6H-SiC substrate was introduced into an ultrahigh vacuum (UHV) chamber equipped with a low energy electron diffraction (LEED) apparatus. The working pressure was ~ 10^{-10} Torr. Annealing the substrate in the UHV causes Si to evaporate from the surface and provides the graphitized 6H-SiC, whose surface is covered with a layer of graphite. The graphitization of the basal faces of the 6H-SiC was confirmed by LEED observation after the annealing. The annealing temperatures were measured with a thermocouple mounted inside the sample holder.

Next, the graphitized 6H-SiC substrates were transferred from the UHV chamber to the MOVPE reactor. The BN layers were grown on graphitized and non-graphitized (0001) 6H-SiC substrates by low-pressure (300 Torr) MOVPE. The non-graphitized substrate was directly introduced into the MOVPE chamber without annealing in UHV chamber. The BN growth temperature was 1100 °C. Triethylboron (TEB) and ammonia (NH_3) were the precursors. The molar flux of TEB for BN growth was 10 μmol/min, and the NH_3 flow rate was 50 sccm. Strong parasitic reaction between TEB and NH_3 requires that we make the NH_3 flow rate as low as possible to obtain a practical growth rate [2]. The thickness of the BN layers was 0.8 μm. A high-resolution X-ray diffractometer (Philips X'Pert System) was used to evaluate the structural quality and determine c-lattice parameters.

3. Results and Discussion

3.1 Graphitization of the surface of 6H-SiC substrate

After the substrate was deoxidized by vacuum annealing under a silicon flux, sharp and bright spots of a Si-rich (3 x 3) reconstruction were observed by LEED. After further annealing of the substrate at 1030 °C in the UHV chamber the surface shows the (0001) SiC ($\sqrt{3}$ x $\sqrt{3}$) reconstruction (Fig. 1) made of Si adatoms in the hollow sites of a compact bilayer termination.

Fig. 1 ($\sqrt{3}$ x $\sqrt{3}$) LEED pattern of 6H-SiC.

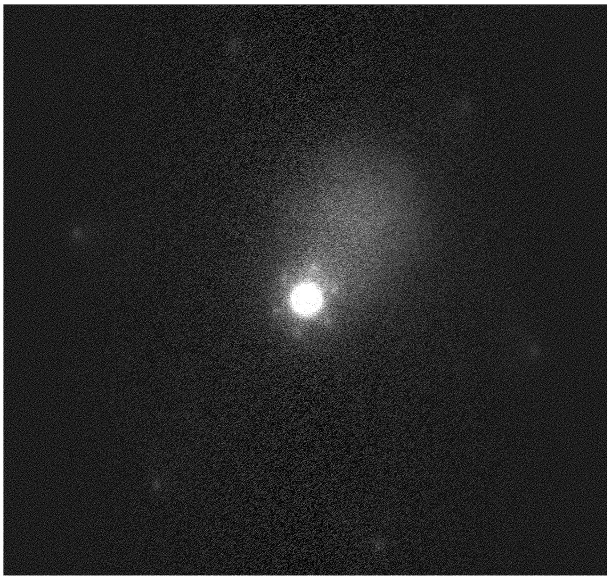

Fig. 2 (1 x 1) LEED pattern of 6H-SiC.

Then further annealing at higher temperature of 1400 °C converts the surface into a layer of graphite having several monolayers thickness. After further annealing at that temperature, the LEED pattern reveals a graphitic termination with only the (1x1) reconstruction spots of crystalline graphite, which is shown in Fig. 2. The graphite layer is monocrystalline on the Si face. The graphite layer has the thickness of 1 – 4 monolayers and on the basis of the LEED pattern shows the same lattice constant as bulk graphite, which is ideal for h-BN heteroepitaxial growth.

3.2 h-BN heteroepitaxial growth on graphitized 6H-SiC

To confirm the effectiveness of the graphitized 6H-SiC substrate, BN layers were grown on graphitized and non-graphitized (0001) 6H-SiC substrates by MOVPE and characterized by XRD. Figure 3 shows 2θ/ω X-ray diffraction patterns of the BN films. No diffraction peak related to BN was observed for the BN sample grown on non-graphitized 6H-SiC substrate, indicating that the structure of the film is amorphous.

In contrast, apart from the (0006) 6H-SiC substrate peak, a strong peak at the diffraction angle of 2θ about 26.5° was observed for BN samples grown on graphitized 6H-SiC substrate. The c lattice constant is 6.72 Å, which is close to the bulk value of 6.66 Å. Therefore, this peak arises from nearly (0001) planes of h-BN. The graphitized 6H-SiC substrate enables us to grow h-BN heteroepitaxial layers successfully for the first time. It should be noted that only turbostratic or amorphous BN layers have been grown on non-graphitized (0001) 6H-SiC substrates [2]. From the viewpoint of crystal quality, BN growth with a larger NH_3 flow rate is preferable. Therefore, further optimization of the growth conditions could lead to h-BN that has the same lattice constant of bulk h-BN.

The exact reasons for this successful formation of an h-BN heteroepitaxial layer on graphitized 6H-SiC are

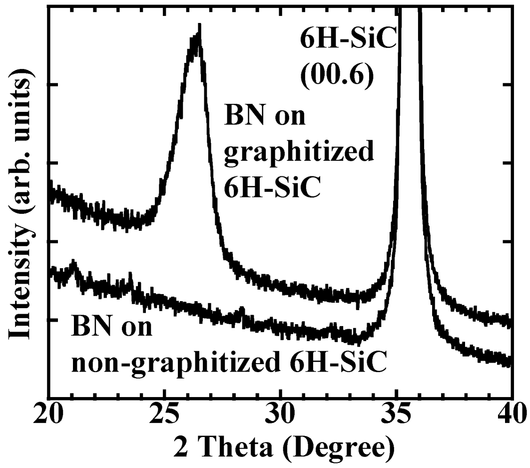

Fig. 3. 2θ/ω X-ray diffraction patterns of BN films on graphitized and non-graphitized 6H-SiC substrates.

still unclear. We speculate that the graphitized 6H-SiC substrate offers two advantages for successful h-BN heteroepitaxial growth. The graphite layer is monocrystalline, having almost the same lattice constant as bulk graphite, which reduces the magnitude of the lattice-mismatch from 19% to 1.6% compared with non-graphitized substrate. Additionally, the graphitization converts the surface structure of 6H-SiC from that with wurtzite sp^3 bonds to one with graphitic sp^2 bonds, circumventing the deterioration of BN crystal quality caused by the mismatch of crystal structure between 6H-SiC and h-BN.

The thickness of the graphite layer is central to achieving h-BN heteroepitaxial layer growth. Thicker graphite layers of over 7 monolayers would lead to exfoliation of the BN films after the growth. A graphite layer with submonolayer thickness could result in turbostratic BN. Therefore, the layer with 1 - 4 monolayer thickness is thought to be optimum.

3. Conclusions

We demonstrated that the h-BN heteroepitaxial layers can be successfully grown on graphitized 6H-SiC substrate by MOVPE for the first time. Graphitized substrates were formed by annealing at 1400 °C in UHV. The thickness of the graphite layer is central to achieving h-BN heteroepitaxial layer growth and thickness of 1 - 4 monolayers is thought to be optimum.

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