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Epitaxial chemical vapor deposition growth of boron nitride atomic sheet over Cu(111)/sapphire substrate

Yuki Uchida[1], Tasuku Iwaizako[1], Seigi Mizuno[1], Masaharu Tsuji[2] and Hiroki Ago[1,3]

[1] *Interdisciplinary Graduate School of Engineering, Kyushu University*

[2] *Research and Education Center of Carbon Resources, Kyushu University*

[3] *Global Innovation Center (GIC), Kyushu University*

6-1 Kasugakouen, Kasuga, Fukuoka 816-8580, Japan

Abstract

Hexagonal boron nitride (h-BN), an insulating two-dimensional (2D) material, is an ideal substrate to bring out properties of other 2D materials, such as graphene, transition metal dichalcogenides and black phosphorus. Here, we demonstrate chemical vapor deposition (CVD) growth of high-quality, large-area monolayer h-BN on Cu(111) thin film deposited on c-plane sapphire using BH_3NH_3 as a feedstock. Highly oriented triangular h-BN grains grow on the Cu(111). Low-energy diffraction (LEED) mesurement indicate the hexagonal lattice of the monolayer h-BN is well oriented along the underlaying Cu(111) lattice, implying epitaxial growth of the h-BN.

1. Introduction

Two dimensional (2D) materials such as graphene, transition metal dichalcogenides and black phosphorus have been gaining interest due to the excellent physical properties.[1-3] However, they are sensitive to surroundings owing to their extremely thin 2D structure. Hexagonal boron nitride (h-BN), a layered material with a hexagonal network of boron and nitrogen atoms, is an ideal insulating substrate for bringing out the intrinsic properties of 2D materials.[4] Chemical vapor deposition (CVD) using catalytic substrate has been widely reported as a promising method to grow large-area h-BN films in recent year.[5-7] Most researchers have used polycrystalline catalytic substrate, mainly Cu foil,[5] Ni foil[6] and Pt foil,[7] which have rough surface with different orientations of grains. Consequently, h-BN film grown on polycrystalline metal substrate consists of a number of h-BN grains with different orientations and even different thickness, resulting in polycrystalline h-BN films composed of poorly connected small h-BN grains. Therefore, the control of crystallinity of the metal substrate is essential for the growth of high-quality h-BN films. Here, we demonstrate epitaxial chemical vapor deposition growth of h-BN atomic sheet using thin Cu(111) film deposited on c-plane sapphire substrate by sputtering. Prepared Cu film consists of (111) plane, which produce highly oriented h-BN sheet. We observed that triangular h-BN grains are oriented in two define directions with respect to the Cu(111) thin film. The controlled orientation of the hexagonal lattice in large-area was confirmed by low-energy electron diffraction (LEED) measurement.

2. Experimental

Cu(111) prepared by sputtering was used for catalytic substrate. CVD growth of h-BN was performed using ammonia borane as a feedstock. To investigate morphologies of grown h-BN and surface conditions surface, SEM and AFM measurements were performed. LEED was measured for demonstrating epitaxial growth of h-BN film.

3 Results and discussion

3.1 SEM observation of grown h-BN

Figure 1 show a SEM images of h-BN grains grown on Cu(111) by CVD. After a short CVD process with 5 min reaction, triangular h-BN grains were observed on the Cu(111) surface. It is considered that the edge of triangular h-BN grains have zigzag edges

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and a previous theoretical study suggested that edge structures of h-BN with terminating nitrogen atoms are more stable in triangular grains.[8] Therefore, it is likely that obtained h-BN grains have zigzag edges with terminating nitrogen atoms (Inset of Fig. 1a). As can be seen in Fig. 1a, most of the h-BN grains are oriented in two specific directions. The existence of these two preferred orientations indicates an epitaxial growth of the h-BN on the Cu(111) surface. By extending the CVD growing time, the lateral size of h-BN grains increase, after long enough growing periods grains completely coalesce, and a uniform h-BN sheet covering the entire Cu surface can be obtained. This result suggests that h-BN grows preferentially on a Cu catalyst compared with the surface of h-BN.

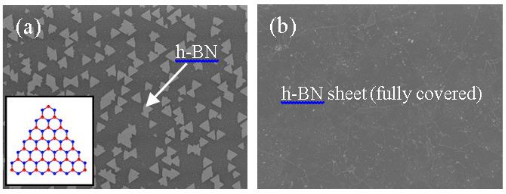

Figure 1. SEM images of h-BN grown on Cu(111)/sapphire substrate at different reaction time of 5 min (a) and 15 min (b). Inset illustrates atomic model of triangular grain of h-BN Scale bar is 20 μm.

3.2 LEED measurement for investigating orientation of h-BN

An orientation of as-grown h-BN on a Cu(111) film was measured by LEED. Because a beam size of LEED we used is about 1mm, it is possible to investigate a macroscopic orientation of h-BN on Cu(111). Fig. 2a and b show LEED pattern measured with 100 eV and the six clear spots which are derived from both h-BN and Cu(111) lattices were measured. Therefore, the diffraction pattern obtained by LEED measurement support a epitaxial growth of h-BN on the Cu(111) suggested by SEM observation. Fig. 2c illustrates the atomic model of triangular h-BN grains oriented in two directions with respect to the Cu(111) lattice determined by our SEM and LEED observations.

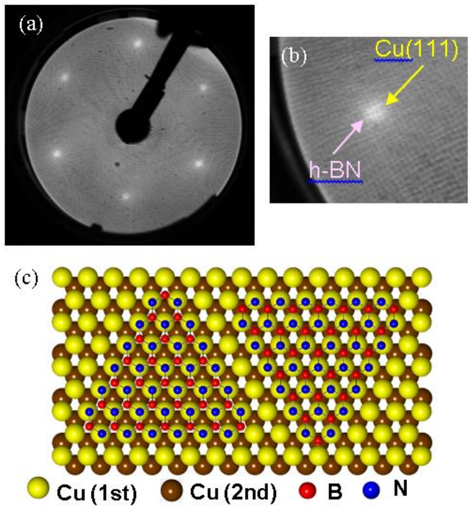

Fig. 2. (a) LEED pattern measured for the fully covered h-BN sheet. (b) Magnified image of one of the diffraction spots. (c) Atomic model of triangular grains of h-BN on Cu(111).

4 Conclusion

Monolayer h-BN was epitaxially grown by CVD on Cu(111) thin film deposited on c-plane sapphire substrates. The use of sapphire substrate realizes a large-scale synthesis of transferrable h-BN. We observed that the orientations of triangular h-BN grains are highly controlled by the Cu(111) lattcice, pointing to two main directions. Further, we obtained a well aligned monolayer h-BN sheet by extending the growth time, which also presents the epitaxy of the monolayer h-BN on Cu(111). Our work is expected to develop an insulating layer for future flexible and transparent 2D devices.

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