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Thin Graphitic Structure Formation on Various Substrates by Gas-Source Molecular Beam Epitaxy Using Cracked-Ethanol

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1. Introduction

While the excellent electrical performance of few-layer graphene (FLG) has been demonstrated for exfoliated FLG, the exfoliation process cannot form the basis of a large-scale manufacturing process. Thus, alternative processes are required to obtain wafer-scale FLG. Here, we propose a new approach based on gas-source molecular beam epitaxy, in which a cracked-ethanol source is employed. This growth method allows us to independently control the substrate temperature and the supply of growth material. This could lead to growth mode control in a layer-by-layer manner at high temperature, which would result in high-quality and large-domain growth. To show the feasibility of this growth method, we tried to grow FLG and analyzed the graphitic structure formed on various substrates.

2. Experimental

For the growth, an ultrahigh-vacuum growth system was equipped with ethanol-gas supply system with a cracking unit consisting of a W filament. Using this system, cracked-ethanol was supplied on various substrates. The substrates used in this experiment were Si(111) with native oxide, α-Al_2O_3(0001) (c-surface of sapphire), and graphene layers (thickness: 3~4 layers [1]) formed on 6H-SiC(0001). For the growth, substrate temperature was set at about 600°C and the W-filament temperature was 2000°C.

3. Results and Discussion

After the growth, Raman spectra were captured from these three substrates as shown in Fig. 1. Peaks attributed to G- and D- bands were observed for all of the substrates while the 2D-band peak was observed only for SiC, indicating that graphitic structures were formed on all substrates while, only on the SiC substrate, there is possibility that graphene were formed.

However, on the SiC substrate, because graphene had already been formed by thermal decomposition before the growth, it is difficult to confirm the growth of graphene layers by Raman spectrum alone. Therefore, we precisely analyzed the graphitic structures on the SiC substrate. Figure 2 shows a transmission electron microscope (TEM) image after the growth. A thin layer structure of six to seven layers can be seen in this TEM image, indicating an increase of three to four layers by the growth process. We characterized this layer structure by x-ray photoelectron spectroscopy. In Fig. 3, C 1*s* spectra before and after the growth are shown. Before the growth, a peak and a shoulder structure, which are respectively attributed to graphite and SiC, were observed. After the growth, the peak intensity increased and the typical spectrum for bulk graphite was obtained. No other peaks, such as peaks related to oxides, were observed. These results indicate that only graphitic material grew and covered the surface. The thickness of the grown layers, estimated from attenuation of peak intensities in Si 2*p* of SiC

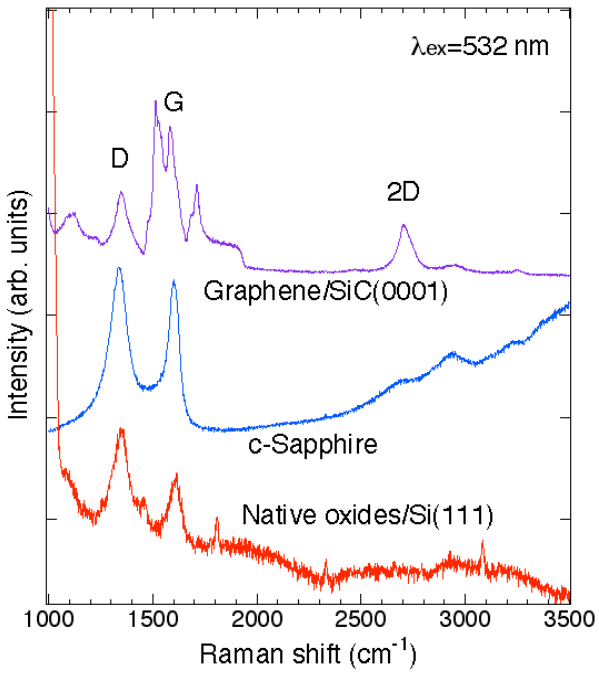

Fig. 1. Raman spectra of various substrates.

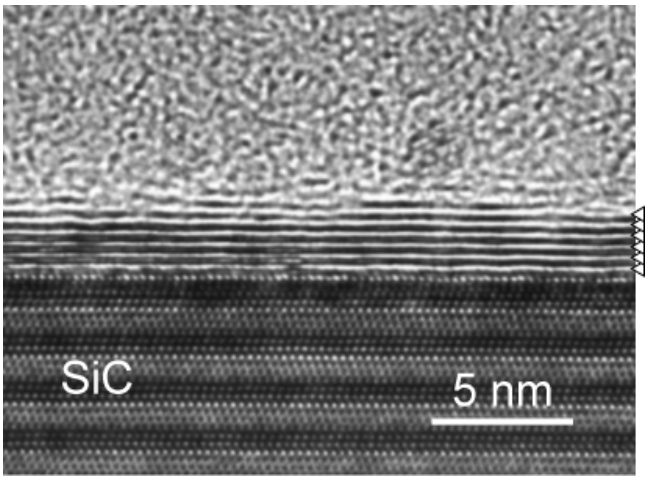

Fig. 2. Cross-sectional TEM image near surface of graphene/SiC(0001) after growth.

substrate was 1.4 nm. This thickness roughly corresponds to four monolayers and is consistent with the TEM observation. These results indicate that FLG was grown on the graphene/SiC(0001) substrate.

In the Raman spectra, the D-band peak intensity was large, indicating that the grown FLG is not high quality. We believe that the optimization of growth condition will improve the quality and that our approach is feasible for the formation of wafer-scale FLG.

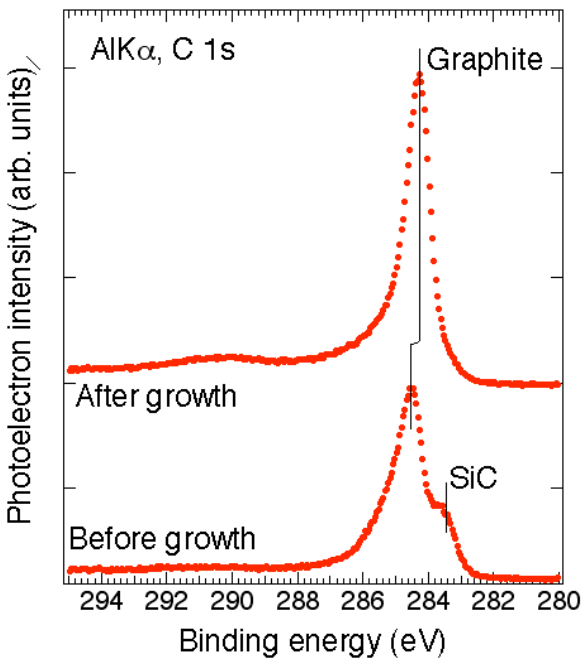

Fig. 3. C 1*s* spectra before and after growth.

Reference

[1] H. Hibino, H. Kageshima, F. Maeda, M. Nagase, Y. Kobayashi, and H. Yamaguchi, Phys. Rev. B **77** (2008) 075413.