Molecular Beam Epitaxy Growth of Graphene and Ridge-Structure Network of Graphene Using Cracked Ethanol

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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. Hence, establishing alternative ways of forming wafer-scale FLG is of great interest from the viewpoint of large-scale integration, and some new alternative approaches have been proposed. Recently, we have proposed a new method based on gas-source molecular beam epitaxy (MBE), in which a cracked-ethanol source is employed [1]. In our previous study, although we showed the feasibility of this growth method, the quality of the graphene was not sufficient. To optimize this growth method to produce high-quality graphene, we examined the substrate temperature dependence of the growth in this study. In addition, we observed a ridge-structure network, consisting of FLG, on graphene after the growth.

As a substrate, we used graphene, whose layers were around 1.3 monolayers, formed on n-type SiC(0001) by annealing at 1800°C in Ar ambient at about 100 Torr. For the MBE growth, the substrate had been heated at temperatures between 600 and 915°C under the flow of cracked ethanol for four hours. Then, we transferred the samples to the analysis system equipped with a monochromatized Al Kα source (1486.6 eV) and a photoelectron analyzer via an ultra-high vacuum and performed *in situ* measurements. Further details are described in [2].

Figure 1 shows the substrate temperature dependence of growth thickness, estimated using the peaks of graphene and SiC substrate derived from the C 1s spectra by a fitting procedure. We found that the thickness decreased with increasing substrate temperature. This trend indicates that the substrate temperature is in a region where the rate-limiting factor is the mass transport, which is higher than the temperature region where the growth is kinematically limited [3]. The growth limited by the mass transport means that the growth rate dose not simply increase even when the supply of ethanol gas increased, which is consistent with the previous study [2]. We obtained a high-contrast transmission electron microscope (TEM) image of the MBE-grown graphene layer (Fig. 2, closed arrow) at 915°C, the highest growth temperature in this study. This layer is better than those in the previous study, in which there are vacancies and fluctuation in the graphene layers [1]. Thus, high-temperature growth improves the quality of the MBE-grown graphene layers, though the growth rate becomes too slow to be of practical use. Since the previous study [2] suggests that the rate-limiting factor may be etching reaction, we believe its suppression is key to increase the growth rate and to improve the graphene quality by the high-temperature growth.

Further, in the center of the TEM image in Fig. 2, we can see a ridge structure of FLG grown in out of plane direction. This structure would be formed by the collision between incommensurate domains of graphene at their interface. An atomic force microscope (AFM) image of the same sample (Fig. 3) indicates that this ridge structure forms a network on the graphene surface. If our assumption is correct, these structures indicate the domain boundaries of graphene and the domain size is estimated to be less than 100 nm.

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References

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Figures

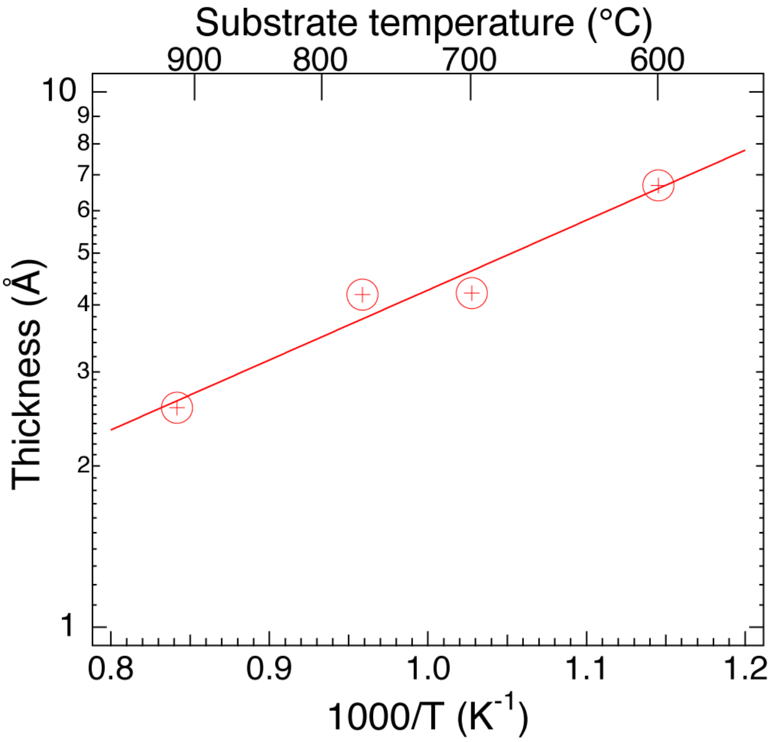

Fig. 1. Substrate temperature dependence of growth thickness.

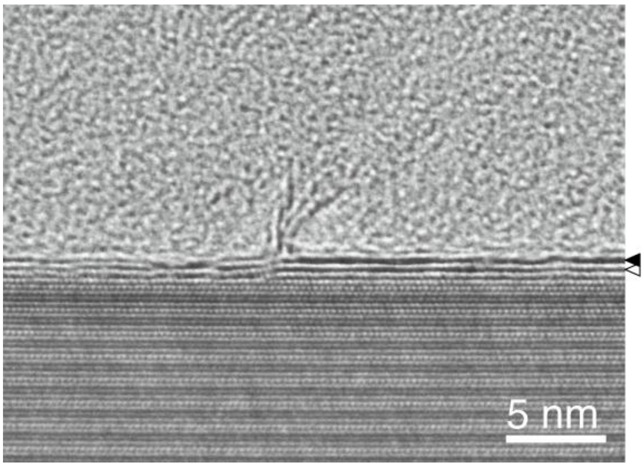

Fig. 2. Cross-section TEM image after MBE growth at 915 °C. The closed arrow points to the MBE-grown graphene layer and the open arrow points to the initial graphene formed by thermal decomposition of SiC.

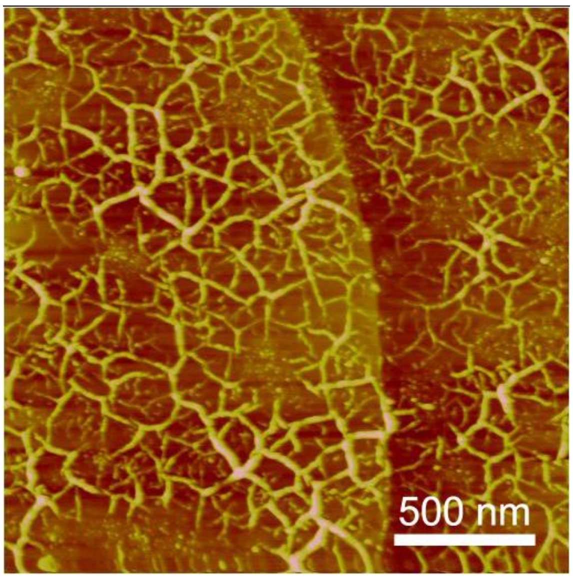

Fig. 3. AFM image after MBE growth at 915 °C