Pressure-Dependent Nucleation and Growth of Graphene Islands on Cu by Chemical Vapor Deposition

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We observed the initial stages of graphene growth on Cu catalyst films by thermal chemical vapor deposition (CVD) under various growth conditions. It was revealed that the graphene growth mode, represented by the shape and spatial density of graphene islands, sensitively depends on the partial pressure of the source gas (P_{source}). Our results also suggest that the morphology of the Cu surface, such as grain boundaries and steps, can play a crucial role in the nucleation and growth of graphene islands under a relatively-high P_{source} condition.

Since the discovery of extraordinary electronic properties of single-layer graphene, a number of efforts have been made toward the preparation of graphene and its device applications. Recently, high-quality graphene has been synthesized on metal catalysts, such as Ni, Co, Ru, by means of the thermal CVD method. Especially, Li and co-workers found that single-layer graphene with good uniformity can be formed on the surface of Cu foil [1]. Since it is difficult to synthesize single-layer graphene uniformly with other catalysts, graphene growth using Cu is a promising way to prepare graphene for various applications. Initial stages of graphene growth on Cu surface have also been investigated by several groups [2, 3]; however, many questions still remain to be solved, which include the nucleation and growth mechanisms, and their dependence on the growth condition and Cu-surface morphology. Understanding these issues is essential to control the domain size and the number of graphene layers. We investigated graphene islands on Cu films formed by the thermal CVD method, and found that the graphene growth mode sensitively depends on experimental conditions such as P_{source}, growth time and the nature of the Cu catalyst surface.

In our experiments, graphene was synthesized on a sputtered Cu film on Si/SiO_2 wafer placed in a cold-wall type high-vacuum chamber using C_2H_4 or CH_4 diluted by Ar and H_2 as the carbon source. After H_2 / Ar annealing, graphene growth was performed at temperatures ranged from 700 to 1000 ^{o}C. P_{source} was 0.1, 0.7, or 4.5 Pa, while the total pressure was kept at 1 kPa. The growth time was varied from several seconds to 60 min.

Graphene islands grown at P_{source} of 0.1 Pa yielded mottled patterns (dark regions) on the Cu surface where some islands stepped over the Cu grain boundaries as seen in scanning electron microscopy (SEM) images in Fig. 1. As P_{source} increases, while the total amount of source gas supply [P_{source} $\times$ growth time] is kept constant, the island size becomes smaller and the islands' density becomes higher as shown in Fig. 2 (P_{source} = 0.7 Pa). In addition, the islands form into a scale-like shape and they tend to be aligned along steps on the Cu surface. This result indicates that the surface morphology such as grain boundaries and surface steps strongly influences the graphene growth mode under this condition. The graphene islands grown at P_{source} of 4.5 Pa (Fig. 3) can be seen to be elongated along the steps. This kind of island shape is different from those reported in previous papers [2, 3]. It was also reported that the shape of islands depended on the total gas pressure [4], but our results clearly indicate that the shape also depends on P_{source} .

We then made back-gate field-effect transistors using graphene grown at P_{source} of 0.7 Pa. For transistor fabrication, graphene was transferred to an SiO_2/Si substrate and patterned by the conventional photolithography technique. The fabricated transistors were successfully modulated by the gate voltage, and exhibited ambipolar behavior. The transistors were found to have fileld effect mobilities exceeding 1000 cm^2 / Vs.

References

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Figures

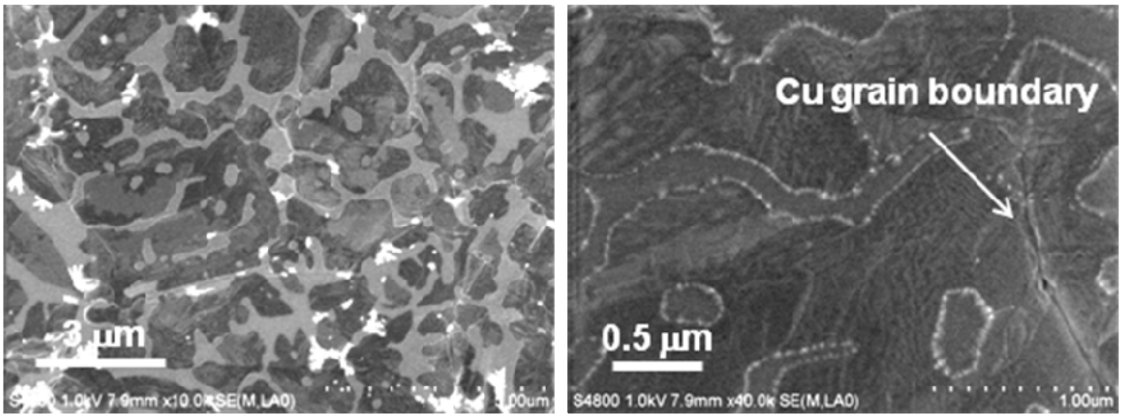

Figure 1

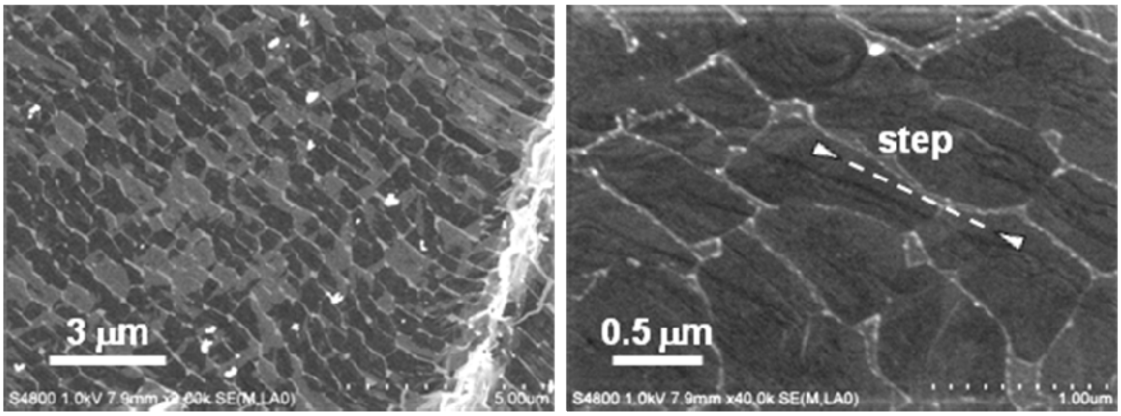

Figure 2

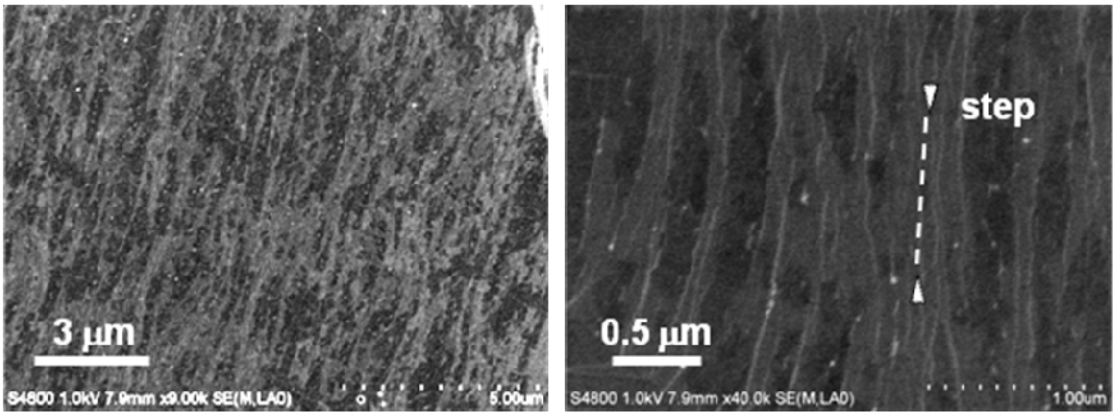

Figure 3

Figure caption
SEM images of graphene islands grown at lower (Fig.1), at medium (Fig.2) and at higher (Fig.3) P_{source} *conditions.*