Cu(111) epitaxial films on mica(001) substrate used for high quality graphene growth by chemical vapor deposition

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Graphene attracts much attention as a wonder material to overcome the limit of traditional materials in the field of electronics, energy storage, thermal management, MEMS and so on. In particular, application of transparent conductive film (TCF) is expected to adapt for solar cells, flat panel displays, and touch panels, because it allows us the high flexibility, cost efficiency and rare-metal free compared with Indium tin oxide. Recently, large-area with high quality graphene grown on transition metal substrates by chemical vapor deposition and transferring to arbitrary substrates have been reported in some research groups to prepare the graphene TCF[1-3]. However, the sheet resistance of graphene TCF deposited by CVD was not high enough to use for solar cells application. For further improvement of the quality of graphene, the crystallinity of Cu substrates such as symmetry and single crystallization must be needed. Reddy et. al. explored a single crystal Cu(111) thin film deposited on basal-plane sapphire by evaporation as a substrates, and obtained low defect graphene on them[4]. To find a feasible way, in this study, we report that epitaxially grown Cu(111) films on synthetic mica ($KMg_3(AlSi_3O10)F_2$) substrates are excellent to avoid defect creation in thin film Cu. Because has a low surface energy and the pit density could be reduced in the thin film Cu deposited on it. In addition, large-area mica substrates are easily obtained and are reusable by cleaving easily the Cu deposited surface layer at a cleavage plane of mica. These are advantages for industrialization of large-area TCF's.

For the growth of graphene, we used 10 cm x 10 cm synthesized mica(001) with a thickness of 0.5mm as a substrate. Cu film with 530 nm in thickness was deposited on the cleaved surface of the mica(001) by EB evaporation at 600 °C. The Cu deposited mica(001) substrate was loaded into a CVD reactor, and it was annealed for 30 min at 800 °C under the hydrogen atmosphere of 0.22 Torr for 30 min. After that, the substrate temperature was increased to 1000 °C, and turned off the hydrogen gas, then CH_4 gas was introduced at a flow rate of 20 sccm. Growth of graphene was carried out for 30 min at a gas pressure 30 Torr.

In order to characterize the crystalline structure of Cu(111)/mica(001), we performed X-ray diffraction (XRD) and electron backscatter diffraction (EBSD) measurements. XRD results of the Cu thin film after CVD show the Cu(111) and Cu(222) reflections with higher-order spectra from the mica(001) substrate. The lattice constant of Cu (111) was 0.2085 nm, which is in good agreement with that of bulk single crystalline. The in-plane distribution of the crystal orientation of deposited Cu examined by EBSD shows the orientation of the deposited Cu was (111) over the surface, but we observed the boundaries attributed to twin crystalline structure consisting of ABC and ABA stacking structures in fcc., The average domain size surrounded by the boundaries are 600 μm or larger, however, it is large enough to grow graphene. Moreover, AFM study revealed the Cu(111) film on Mica(001) after depositing at 1000°C maintained an atomically flat terrace with step bunching structure. Thus, it is expected to be an excellent template for growth of graphene.

The Raman spectra of the graphene on the Cu(111) film at three different measurement points are shown in Fig. 1. Raman spectra of the graphene have no D peak (1358cm^{-1}), and I_G/I_{2D}, is smaller than unity. These results indicate that the graphene is almost mono-layer with no defects. Moreover, Raman mapping show that the full width at half maximum of I_G at the twin boundary is larger than that on the other area. This result suggests that the six-membered rings are distorted at the twin boundary of Cu(111) to solve the discontinuity of each domain of graphene without introducing defects such as dangling bonds and/or irregular bonds.

From these results, Cu(111)/mica(001) is promising substrate for a high quality and large-area graphene growth in the Cu-based CVD method.

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References

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Figures

Fig. 1

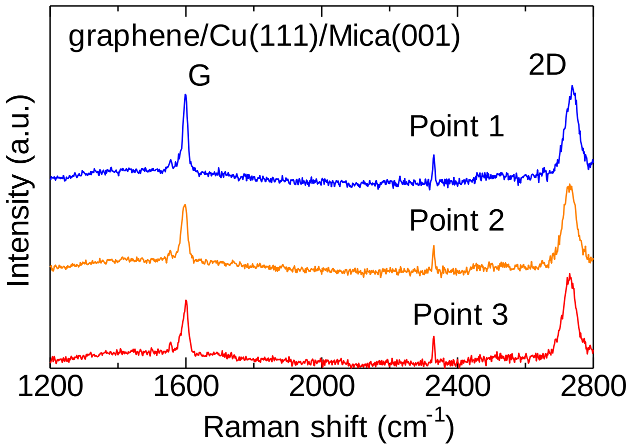

Figure caption

Fig.1 Raman spectra of the grahene/Cu(111)/Mica(001) measured at three different points by using 488 nm excitation wavelength.