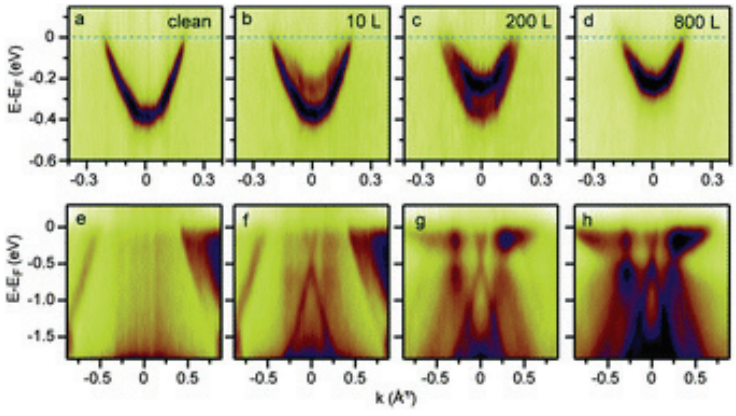
# Rotated Domains in Chemical Vapor Deposition-Grown Monolayer Graphene on Cu(111): An Angle-Resolved Photoemission Study

Graphene, an emergent two-dimensional carbon sheet, has extraordinary electrical properties due to the linear energy band dispersion at the Dirac point. Charge carriers in graphene behave like relativistic particles with zero rest mass, resulting in ballistic transport on the micrometer scale at room temperature. This characteristic makes graphene a promising material for future electronic devices, and enormous progress has been made since the existence of graphene was clearly demonstrated in 2004.

The synthesis of a realistic monolayer graphene *via* a self-limiting process was achieved on Cu foil as a result of the low carbon solubility at growth temperatures. Thus, copper is considered to be the most promising substrate for the growth of high-quality and large area graphene by chemical vapor deposition (CVD), in particular, on the (111) facet. Because the interactions between graphene and Cu substrates influence the orientation, quality, and properties of the synthesized graphene, the interactions using angle-resolved photoemission spectroscopy (ARPES) was investigated in this study [1].

ARPES measurements were conducted at the 4A2 and 10D beamlines of the Pohang Accelerator Laboratory in Korea. All ARPES spectra in this work were taken at the 4A2 beamline using a R4000 analyzer (VG-Scienta) with 30 eV photons at room temperature.




The evolution of both the Shockley surface state of the Cu(111) and the $\pi$ band of the graphene was measured from the initial stage of CVD growth to the formation of a monolayer. Graphene growth was initiated along the Cu(111) lattice, where the Dirac band crossed the Fermi energy ($E_F$) at the $K$ point without hybridization with the d-band of Cu. Then two rotated domains were additionally grown as the area covered with graphene became wider. The Dirac energy was about -0.4 eV and the energy of the Shockley surface state of Cu(111) shifted toward the $E_F$ by ~0.15 eV upon graphene formation. These results indicate weak interactions between graphene and Cu, and that the electron transfer is limited to that between the Shockley surface state of Cu(111) and the $\pi$ band of graphene. This weak interaction and slight lattice mismatch between graphene and Cu resulted in the growth of rotated graphene domains (9.6° and 8.4°), which showed no significant differences in the Dirac band with respect to different orientations. These rotated graphene domains resulted in grain boundaries which would hinder a large-sized single monolayer growth on Cu substrates. Since the graphene was grown via CVD and the (111) facet is the best for uniform graphene growth, this study directly explains the phenomena of the growth of graphene on Cu foils using CVD. We believe that understanding the clear mechanism of formation of rotated domains is the first step toward the growth of large-sized uniform graphene with minimized domain boundaries on Cu.

### *References*

[1] Nanoscale, 2013,5, 8210-8214