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Rotated domains in chemical vapor deposition-grown monolayer graphene on Cu(111) : an angle-resolved photoemission study

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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, we studied the interactions using angle-resolved photoemission spectroscopy. The evolution of both the Shockley surface state of the Cu(111) and the π 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 π 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.

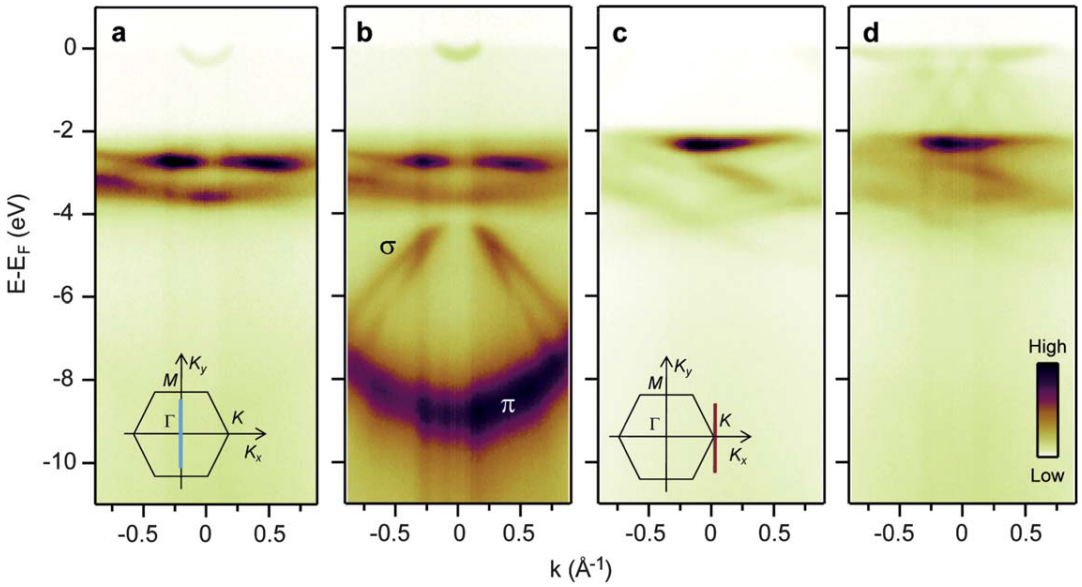

Fig. 1 Band structures of the clean Cu(111) substrate and the graphene–Cu(111) system around the (a and b) Γ and (c and d) *K* points of the first BZ of graphene