

# Chemical vapour deposition (CVD) growth of graphene on copper grains of different crystallographic orientations

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Large-scale production of high quality graphene thin films is necessary to integrate it into optoelectronic devices. Chemical vapor deposition (CVD) of graphene on copper is a scalable route for high quality deposition because copper is easy to etch, facilitating transfer onto insulating substrates [1] and it is inexpensive. In order to produce graphene films of high quality and variable thickness it is necessary to understand the mechanism of growth, which is dictated by the nature of graphene nucleation sites and their kinetics on the Cu surface. However the interrelated mechanisms of activation of graphene nucleation [2] and density of such sites as a function of the copper morphology, size and crystallographic orientation of grains, are not yet fully understood.

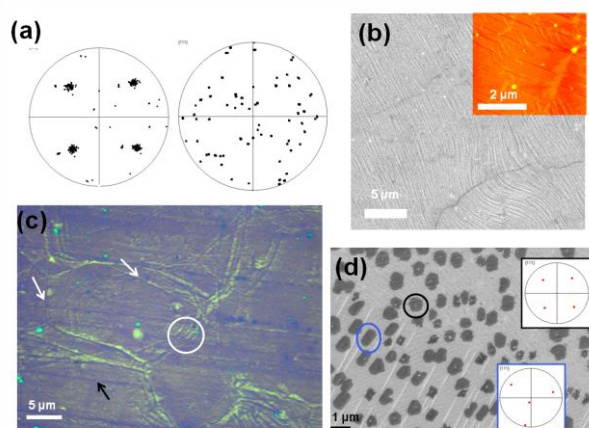
Here we demonstrate the thickness dependence of graphene on CH<sub>4</sub> (carbon precursor source) flow rate, growth pressure, different polycrystalline textures, morphologies and purity of Cu foils (Figure 1). We used different gas mixtures, namely CH<sub>4</sub>/H<sub>2</sub> and CH<sub>4</sub>/H<sub>2</sub>/Ar to grow graphene by low pressure (LP) CVD (0.03-0.5 Torr) and copper foils which underwent different manufacturing processes (Figure 1a).

Uniform graphene coverage (Figure 1b) was obtained at low CH<sub>4</sub> flow rate (0.5 sccm) on copper foils with uniform crystallographic orientations of the grains (Figure 1a). Higher growth pressure increased the graphene thickness only in specific areas that were higher in roughness. While applying the same growth conditions on Cu foils with random crystallographic orientation of the grains, the resulting film was composed of monolayers and 2-3 layers where the distribution of thicknesses reflects the underlying copper morphologies (Figure 1c). Finally, perceptible difference in the graphene nuclei growth directions and rate on different Cu orientations [(111) versus (100)] are reported (Figure 1d). Our results reveal that the graphene nucleation sites can be activated differently from the evolution dynamics of the Cu surface at high temperature, as well as from the CH<sub>4</sub> partial pressure. Hence, altering the copper texture and the feedstock could lead to control of the graphene thickness.

## References

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**Figure 1.** Electron Backscatter Diffraction (EBSD) pole figures display a strong texture of the Cu films in the first case (left) while a random crystallographic orientations of the Cu grains in the second case (right); (b) scanning electron microscope (SEM) image of single layer graphene grown on Cu film with preferential crystallographic orientations of the grains. Atomic force microscope (AFM) image of the copper morphology is reported in the inset; (c) optical image of transferred graphene on 300 nm SiO<sub>2</sub>. The film thickness varies depending upon of the pre-existing copper morphologies. White arrows and circle indicate the profile of grooves while black arrow indicates areas of higher roughness; (d) SEM image of graphene nuclei grown on Cu grains of different crystallographic orientations. Nuclei with elongated shape are preferentially found on Cu (111) (blue circle), while nuclei with isotropic shape are preferentially found on Cu (100). EBSD pole figures display (111) and (100) orientation of the two adjacent Cu grains.