Controlling the size, number of layers and planarity of CVD graphene single-crystals

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Chemical vapor deposition (CVD) of graphene on copper catalyst is presently considered as the most promising manufacturing approach as it yields large-area graphene with a relatively good control over the structural quality and the number of layers. In order to benefit from graphene's excellent physical properties and fully exploit its potential in advanced technological applications, it is essential to precisely control the CVD process.

Our work focuses on studying the correlation between the conditions employed during the CVD protocol, the type of Cu substrate and the graphene growth results. The last findings which have been made recently in order to reduce the presence of structural defects, limit the thickness to one single layer and prevent the formation of wrinkles and cracks will be presented.

CVD protocols typically consist of four successive steps: the CVD furnace warming, the high temperature Cu annealing, the graphene growth, and the furnace cooling. First, we systematically studied how graphene growth conditions (CH_4 partial pressure, H_2-to-CH_4 ratio, pressure) influence the density, the shape, the structural quality and thickness of graphene domains [1]. We then investigated how the conditions (gas composition, pressure) employed during the furnace warming and Cu annealing steps impact graphene nuclei density and number of layers [2]. Among other results, we found that two different kinds of graphene multilayer growth mode can take place: compact ad-layers formed via the diffusion of carbon species through the Cu foil bulk and branch-like multilayer regions formed through the diffusion of C species on the Cu surface (see Fig. 1) [2]. Finally, we found that employing a thin Cu film pre-deposited on a flat and rigid substrate (fused quartz or crystalline sapphire) as catalyst instead of commonly used Cu foils is essential to improve graphene's planarity and physical integrity (see Fig. 2).

Our findings provide clear guidelines to produce ultra-flat defect-free single crystals of graphene with a lateral size exceeding a few millimetres and a controllable number of layers.

Figures

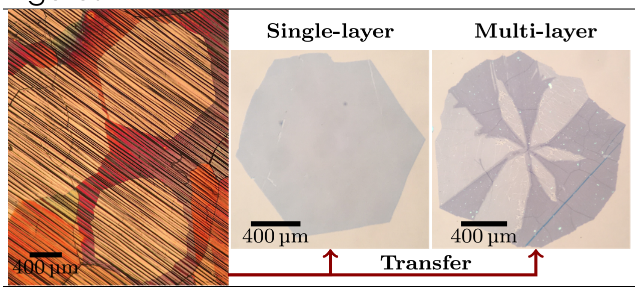

Figure 1: Millimeter-size graphene domains grown on Cu foils. Left panel: On Cu after a slight oxidation to make graphene visible. Right panel: transferred single-layer and branch-like multilayer domains (from [2]).

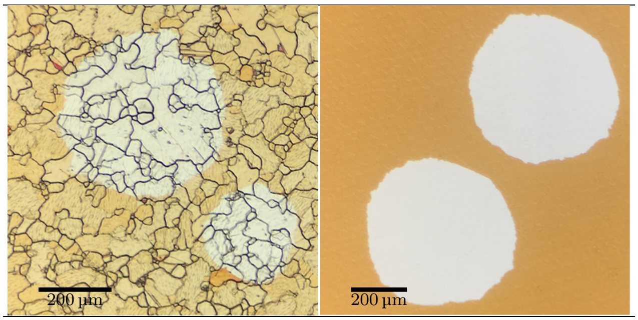

Figure 2: Graphene domains grown on (left panel) polycrystalline and (right panel) single-crystalline Cu films after a slight Cu oxidation to make graphene visible.

References

[1] B. Huet & J.-P. Raskin, *Chemistry of Materials*, 29 (8), 2017, 3431-3440
[2] B. Huet & J.-P. Raskin, *Carbon*, 129, 2018, 270-280