## Pressure-Controlled Chemical Vapor Deposition of Single-Layer Graphene with Millimeter-Size Domains on Thin Cu Film

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Using a thin copper film deposited on a flat, smooth and rigid substrate as catalyst for the chemical vapor deposition (CVD) of graphene offers exciting prospects in terms of graphene quality and processability. However, producing graphene on thin Cu films rather than on widely used Cu foils raises an additional challenge as their quasi-2D nature makes them more prone to degradation upon the harsh environment required to grow high-quality graphene.

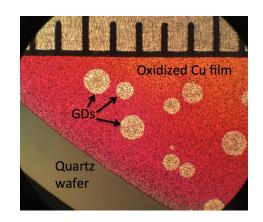
In this work, we demonstrate the synthesis of single-layer araphene with compact millimeter-size domains on thin Cu films (Figures 1 & 2). This has been achieved by carefully controlling the conditions inside the CVD furnace as the graphene synthesis procedure proceeds. A wide range of CVD parameters (global pressure, methane partial pressure, methane to hydrogen ratio, etc) has been systematically investigated to better understand the involved fundamental graphene growth mechanisms and to determine optimal conditions for the different process steps including Cu substrate annealing, graphene nucleation and growth.

Our results show that using a high global pressure during the graphene growth is essential to grow defect-free compact domains. The low nucleation site density required for the synthesis of large graphene domains (GDs) has been achieved by combining a high H<sub>2</sub>-to-CH<sub>4</sub> ratio during the graphene growth step and an in-situ Cu film

oxidation induced by a high pressure level of a non reducing atmosphere (argon) during the Cu annealing step.

Finally, it is found that a brief evacuation of the CVD furnace from its argon atmosphere prior to the graphene growth step is a key process step to maintain the physical integrity of the Cu film during the CVD process. Our method provides a scalable and reproducible way to produce high quality graphene on 3-inch thin Cu film substrate which is a convenient platform for the realization of graphene-based devices.

## Figures:



**Figure 1:** Mm-size graphene domains (GDs) produced on a thin Cu film deposited on a fused quartz wafer. The Cu film surface has been slightly oxidized after the CVD process in order to make graphene visible.



**Figure 2:** Mm-size graphene domains transferred on a Si/SiO<sub>2</sub> substrate.