

# Controlled Growth of High-Quality Graphene Films: Manipulating the Crystal Orientation via Chemical Vapor Deposition

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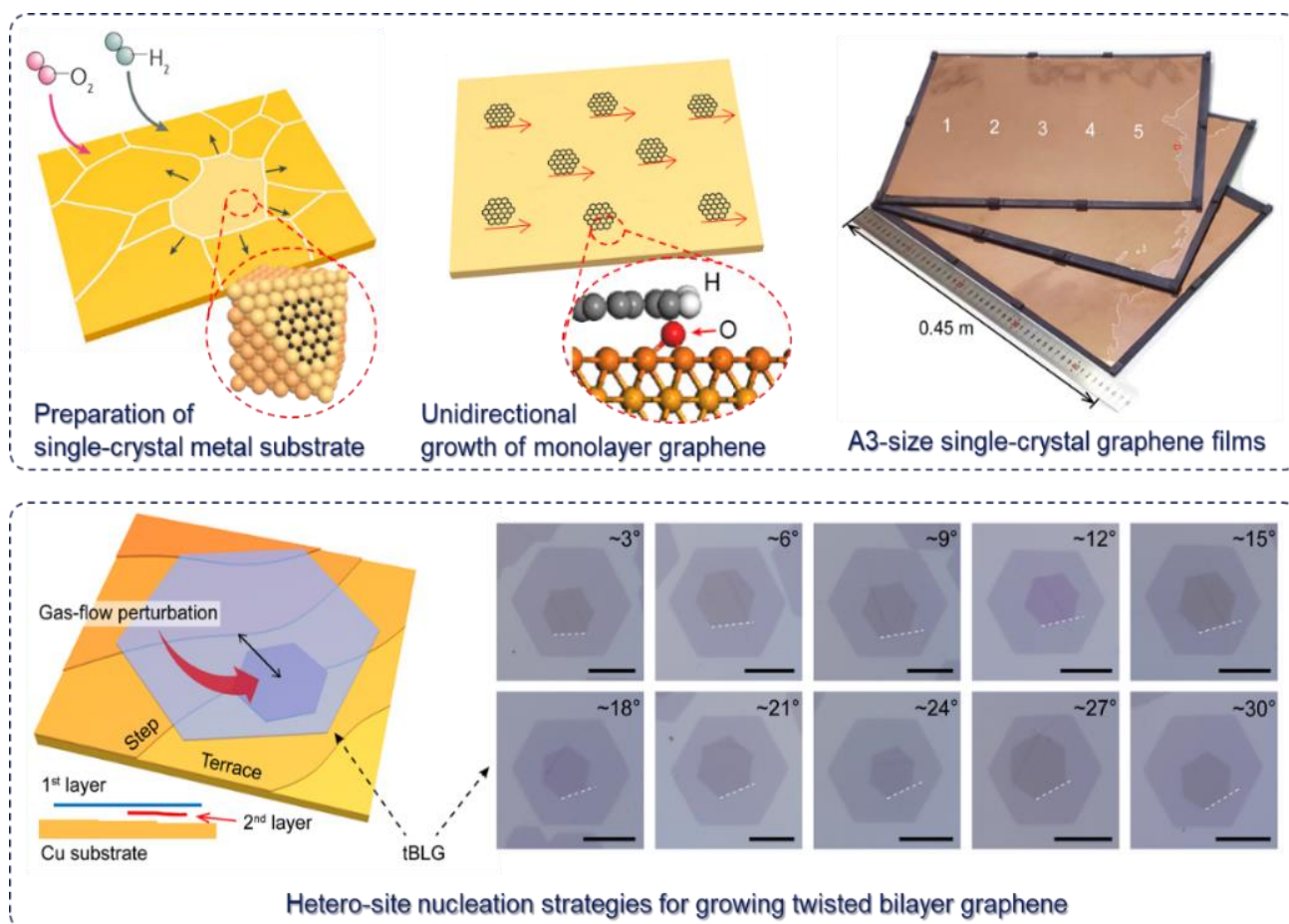
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## Graphical Abstract

Epitaxial growth of well-aligned graphene relies on the achievement of single-crystal metal substrate and strong graphene-substrate interaction, while the obtaining of twisted bilayer graphene is enabled by gas-flow perturbation and hetero-site nucleation strategy.



## Abstract

Graphene has garnered widespread interest and confer remarkable potential for next-generation technological applications, which relies on the controllable preparation of high-quality graphene films. Chemical vapor deposition (CVD) is considered the most promising method, and great progress has been achieved over the last decade. Currently, this field is being pushed to new heights that pursuit structure control (e.g., orientation, layer, stacking order, contamination, doping, etc.) and low-cost production (e.g., increasing the production capacity and growth rate) [1,2]. In this talk, I will introduce our recent works on controlled growth of high-quality graphene films via CVD approach, especially on controlling the crystallographic orientation of graphene. By designing and preparing single-crystal Cu(111) foils, we have opportunities in realizing the epitaxial growth of large-area single-crystal graphene film [3,4]. We designed and constructed a pilot-scale CVD system suitable for producing A3-size graphene films, which works well and output high-quality graphene films with high capacity. In another hand, we also explore the possibility on controlling the layer number and stacking order, which is motivated by the emerging twistrionics. Here I will present our state-of-the-art hetero-site nucleation method for growing twisted bilayer graphene (tBLG) [5]. Gas-flow perturbation and switching of the graphene edge termination play crucial roles in triggering the formation of interlayer twist. The growth mechanism is carefully investigated by using an isotope-labelling technique. The as-obtained tBLGs show high crystalline quality, which is confirmed by the Raman spectra, atomically clear Moiré patterns in TEM image and ultrahigh carrier mobility ( $68,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  at room temperature).

**Keywords:** Chemical vapor deposition; single-crystal graphene, twisted bilayer graphene.

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