

完全 AB 積層した二層グラフェンの CVD 成長

Synthesis and stacking evolution of highly uniform AB-stacked bilayer graphene

Pablo Solís-Fernández¹, Yuri Terao¹, Kenji Kawahara¹, Kosuke Nagashio², Yung-Chang Lin³,

Keisuke Yamamoto¹, Hiroshi Nakashima¹, Hiroki Hibino⁴, Kazu Suenaga³, Hiroki Ago¹

Kyushu Univ.¹, Univ. Tokyo², AIST³, Kwansei Gakuin Univ.⁴

E-mail: ps-fernandez@gic.kyushu-u.ac.jp

Control of the stacking order in bilayer graphene (BLG) allows realizing unique physical properties, owing to the intimate correlation between the stacking order and the electronic properties. The possibility of tuning the band gap in AB-stacked BLG (AB-BLG) has a great technological importance for electronic and optoelectronics applications. Most of current methods to produce AB-BLG suffer from inhomogeneous layer thickness and/or coexistence with twisted BLG.¹ Here, we demonstrate a method to synthesize highly pure large-area AB-BLG by chemical vapor deposition (CVD) using Cu-Ni films.² We found that increasing the reaction time resulted in a gradual increase of the AB stacking, with the BLG eventually free from twist regions for the longer times (Fig. 1a), due to catalyst-assisted continuous BLG reconstruction driven by carbon dissolution-segregation processes. This method allows to obtain large areas of BLG, which currently are only limited by the size of the CVD furnace employed (inset of Fig. 1a). The stacking order was confirmed by several techniques, including Raman spectroscopy (Fig. 1b) and STM (Fig. 1c). The opening of a band gap was successfully observed by electrical measurements on dual gate field-effect transistors (FETs) (Fig. 1d). The concept of the continuous reconstruction to achieve highly pure AB-BLG offers a new strategy to control the stacking order of catalytically grown two-dimensional materials.

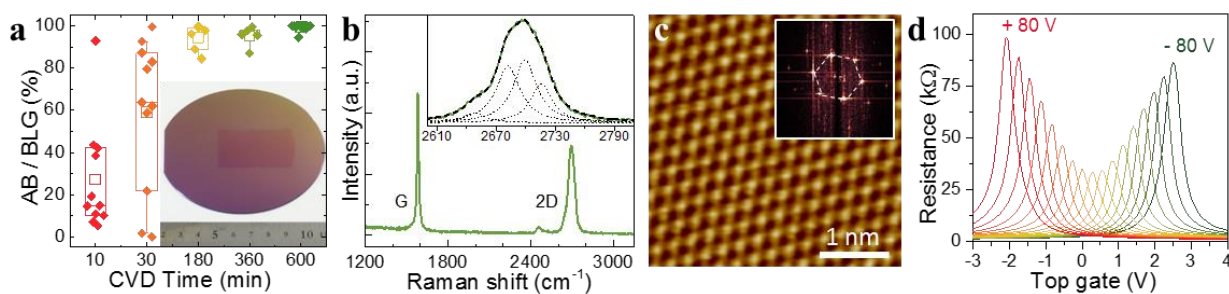


Figure 1. (a) Evolution of the AB stacking ratio with CVD time. Inset shows a large area BLG on a SiO₂ substrate. (b) Raman spectrum of the AB-BLG. Inset shows an enlargement of the 2D band, fitted by 4 Lorentzian peaks. (c) STM image of the AB-BLG. Inset shows the FFT of the image. (d) Resistance of an AB-BLG FET as a function of the top gate voltage, for several applied back gate voltages. The numbers indicate the value of the back gate voltage for the corresponding curve.

References:

1. Takesaki, Y. *et al.* Highly uniform bilayer graphene on epitaxial Cu-Ni(111) alloy. *Chem. Mater.* **28**, 4583–4592 (2016).
2. Solís-Fernández, P. *et al.* Isothermal growth and stacking evolution in highly uniform AB-stacked bilayer graphene. *Submitted for publication* (2019); doi:10.26434/chemrxiv.11459316.v1.