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Toward Controllable Growth of Millimeter-Size CVD Graphene Single Crystals and Its Application in 2D van der Waals Heterostructure Based Photodetector

In conventional chemical vapor deposition process, as-grown large-area graphene films on catalytic metal foils are regarded as coalescence of many small graphene grains with lateral size of less than 50 μm. The dense graphene grain boundaries would introduce severe electron/hole scattering, which degrades physical properties of graphene, such as carrier mobility, therefore retarding graphene-related applications. Through nucleation density control in height-confined graphite reaction cavity, the high-quality millimeter-scale graphene single crystals can be synthesized using commercial chemical vapor deposition equipment.

The graphite reaction cavity is able to mimic the Cu enclosed configuration [1], which can suppress Cu sublimation, therefore reducing roughness of Cu substrates. In addition, the sublimated Cu is trapped in graphite reaction cavity, where the Cu vapor-rich surroundings can appropriately boost the growth rate. Inside the graphite reaction cavity, a height-confined sapphire slit is designed for the insertion of the growth substrate, which can consecutively provide trace amount of oxygen to oxidized the Cu substrate. The formation of copper oxide can block the pathway of graphene growth [2], i.e. the graphene nucleation is restrained. Therefore, through adjusting the height of the sapphire slit as well as ex situ pre-oxidation of Cu substrates, the oxygen concentration near the Cu surface can be controlled, implying the control of graphene nucleation density. Figure 1(c) and (d) shows the size of graphene single crystals is up to millimeter scale before coalescence.

The as-grown millimeter-size graphene single crystals is then transferred by low average molecular weight (AMW) PMMA, which is an optimized supporting layer developed for the low-residue transfer of CVD graphene. Compared with the long-chained PMMA, the graphene sample transferred by low AMW PMMA suffers less p-type doping and contamination. Through X-ray photoelectron spectroscopic analysis in Figure 2, the binding energy intensities of C–C, O–CH3, and O–C=O components confirm the extensive reduction of the residue using low AMW PMMA, which can serve as a promising supporting layer for the transfer of graphene and other two-dimensional (2D) layered materials.

The high-quality large graphene single crystals and well-developed low residue transfer method can be applied in 2D van der Waals heterostructure based photodetectors. In Figure 3, $ReSe_2$ based photodetector is presented. $ReSe_2$ is a promising transition-metal dichalcogenide (TMDC), which has a weakly layer-dependent bandgap (it increases from 1.29 eV to 1.31 eV when passing from bulk to atomically thick layers) but a high photo-responsivity when in few-layer form [4]. Here, the as-fabricated 2D few-layer $ReSe_2$-based photodetector with high photosensitivity (~106 photo-to-dark conductivity ratio, see Figure 3(c)) and fast switching time is reposted. The device has a planar architecture based on high-quality CVD-grown 2D materials that are van der Waals stacked.

References

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Figures

Figure 1: (a) Graphene grown by conventional CVD method. (b) Cu surface pre-treatment in $FeCl_3$/HCl solution. About 600 nuclei/cm2 can be achieved. (c) Large graphene single crystals through a height-confined graphite reaction cavity (d) As-transferred millimeter size graphene single crystal.

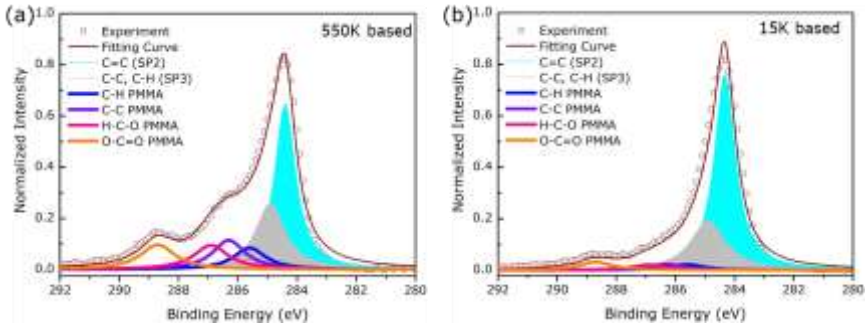

Figure 2: The XPS results of graphene samples transferred by (a) 550K and (b)15K-based PMMA.

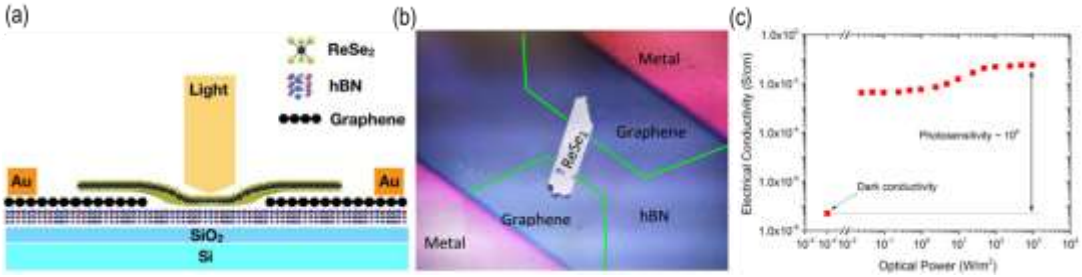

Figure 3: (a) 2D material hetrostructure. (b) Top view of van der Waal stack device. (c) Electrical conductivity as a function of irradiance under white light illumination.