Supporting Information

Direct Synthesis of Van der Waals Solids

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The following pages include supporting figures and experimental data.

Figure S1. Raman spectral mapping of the graphene 2D/G intensity ratio provides evidence of graphene layer thickness variation in epitaxial graphene on SiC. Lower ratios correlate with multilayer graphene. Ratios > 1 are typically associated with monolayer QFEG, while ratios of 1 or less are multilayer.¹ (Scale: 3 μm)

Figure S2. X-ray Photoemission spectroscopy (XPS) demonstrates (a) that hydrogenation of epitaxial graphene on SiC (0001) eliminates the buffer layer (identified as S1 and S2) at the interface of graphene and SiC(0001). High resolution XPS of the Mo 3d (b) and S 2p (c) peaks MoS_2 , indicating no Mo-C or C-S bonding following MoS_2 synthesis on EG and QFEG templates.

Figure S3. AFM micrographs illustrating monolayer and pyramidal MoS₂ growth on (a) as-grown epitaxial graphene (EG) and monolayer MoS₂ on (b) quasi-free standing epitaxial graphene (QFEG) with secondary nucleation occurring at wrinkles and step edges. The scale bars are 500 nm in length.

Figure S4. (a) A 5 μm by 5 μm AFM micrographs shows the grain sizes of initial nucleation of MoS₂ layers in the growth for 10 min on EG. (b) The corresponding deformation image mapping the MoS₂ crystal distribution on the EG template.²

60 mV reduction in surface
potential at step edge center

Figure S5: Surface potential force microscopy (SPFM) micrograph of a 5 μm by 5 μm region of as-grown epitaxial graphene on a SiC ($1\bar{1}0\text{n}$) step edge and (0001) terrace (a). The image contrast is directly related to a surface potential difference of 60 mV (light green to violet) across a 7 nm-high step edge between two terraces (b). The reduction in potential is well correlated with a transition from monolayer to multilayer EG, as well as a shift in strain of the EG, confirming the impact of thickness and strain on surface potential and reactivity.

Figure S6. The output characteristics of the MoS₂/QFEG photosensor illuminated with a power at 2.5 μ W under 514 nm excitation wavelength. The measured drain current increases from 250 nA to 3400 nA at $V_{ds} = 3$ V when the 15x20 μ m MoS₂/graphene channel is illuminated by a 500nm diameter laser.

Figure S7. (a) The output characteristic of the bare QFEG photosensor shows the massive current transport. (b) The transient measurement under 514 nm excitation wavelength switched on and off at each 300m sec at 77K in vacuum shows the photoresponse of the bare QFEG photosensor without the affect from heat and ambient.

Figure S8. The XPS spectra of hBN/QFEG/SiC heterostructures. In this survey the measured B/N ratio is 0.95.

REFERENCE:

- [1] Robinson, J. A.; Trumbull, K. A.; LaBella, M.; Cavalero, R.; Hollander, M. J.; Zhu, M.; Wetherington M. T.; Mark Fanton, M.; Snyder, D. W. Effects of Substrate Orientation on The Structural and Electronic Properties of Epitaxial Graphene on SiC (0001). *Appl. Phys Lett.* **2011**, 98, 222109-222109.
- [2] Pittenger, B.; Erina, N.; Su, C Quantitative mechanical property mapping at the nanoscale with PeakForce QNM. *Application Note Veeco Instruments Inc.* **2010**.