# **Electronic Supplementary Material**

# Towards growth of pure AB-stacked bilayer graphene single crystals

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**Figure S1** EDS Mapping of a silica particle on Cu foil. (a) SEM image of a silica particle. (b)–(d) EDS mapping of Cu (b), Si (c), and O (d) corresponding to (a).

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Figure S2 Photograph of the graphite box.

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**Figure S3** (a) optical image of the foil annealed without the box. Particles can be seen clearly on the surface. (b)–(h) optical images of the foil annealed in the box at different areas. No particles can be found. The image sizes of (a)–(h) are the same.



**Figure S4** Optical images of a graphene island (a) and its zoom-in image (b) grown on Cu/Ni (1 1 1) foils by conventional CVD method. Multilayer and twisted islands can be found due to random particles.



**Figure S5** Statistical optical image of misalignment regions in bilayer graphene grown without (a) and with heat-resisting box (b). The twisted and multilayer regions are marked as black circle and line, respectively.



**Figure S6** Raman spectra of the bilayer graphene samples. (a) Raman mapping of the  $I_{2D}/I_G$  ratio of bilayer graphene. (b) Raman spectra of the as-grown bilayer and monolayer graphene. (c) Lorentz curve fitting of the 2D band of the bilayer graphene sample, the red curves are four Lorentz curves with the FWHM of 31 cm<sup>-1</sup>.



Figure S7 (a) SAED pattern of the AB-stacked bilayer graphene. (b) SAED intensity profile along the direction marked in (a).



Figure S8 LEED patterns of the AB-stacked bilayer graphene at different areas. The nearly identical orientations indicate perfect alignment of the graphene lattice.



**Figure S9** Photograph of the bilayer graphene transferred onto a SiO<sub>2</sub>/Si substrate.

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**Figure S10** Optical images of graphene samples grown on Cu/Ni (1 1 1) alloy films with different Ni concentrations: (a) 12.5 at.%, (b) 14.8 at.%, (c) 17.1 at.%, (d) 19.2 at.%. The growth conditions were the same for all samples.



**Figure S11** Optical images of graphene grown on Cu/Ni (1 1 1) foils and transferred onto SiO<sub>2</sub>/Si substrates with different temperature: (a) 1000 °C, (b) 1050 °C, (c) 1100 °C, and different  $H_2/CH_4$  ratio: (d) 30:2, (e) 15:2, (f) 5:2. The growth conditions were the same for all samples.