Rapid Growth of Large-Area Single-Crystal Graphene Film by Seamless Stitching Using Resolidified Copper Foil on Molybdenum Substrate

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Fig. S1 The process diagram of the single-crystal graphene film growth.



Fig. S2 SEM image of graphene domains after the cooling process of 1085°C -1070°C.



**g. S3** (a, b) EBSD mappings and (c) XRD spectra of single-crystal Cu foil after annealing at 1085°C and polycrystalline Cu foil after annealing at 1070°C. Insets of Fig. S3a and Fig.S3b show the inverse pole figure orientation coloring scheme of the

EBSD mappings. (d) HRTEM image of single-crystal Cu foil shows its fcc (111) surface orientation. Inset of Fig.S3d shows the fast Fourier transformation pattern of the corresponding HRTEM image.



**Fig. S4** (a) SEM image and (b) the corresponding EBSD mapping of Cu (111) film on Mo substrate after single-crystal graphene domains growth. (c) SEM image and (d) EBSD mapping of Cu (111) film on Mo substrate after single-crystal graphene film growth. Insets of Fig. S4b and Fig. S4d show the inverse pole figure orientation coloring scheme of the EBSD mappings.



**Fig. S5** (a) Digital photo of Cu (111) film on Mo substrate. (b) Inverse pole figure orientation coloring scheme of the EBSD mappings. (c-i) EBSD mappings of Fig. S5a from any seven areas show its fcc (111) surface orientation.



**Fig. S6** (a) SEM image of the Cu and Mo substrate cross section, EDS mappings of (b) Mo element and (c) Cu element.



**Fig. S7** (a-d) Single-crystal graphene film from selected regions of Fig. S7e. (f-q) SAED patterns from the representative regions indicated in Fig. S7a-S7d.



**Fig. S8** AFM images of (a) height-mode and (b) phase-mode of the stitched singlecrystal graphene domains. AFM images of (c) height-mode and (d) phase-mode of single-crystal graphene film.



**Fig. S9** (a) Optical image and (b) enlarged SEM image of the single-crystal graphene film edge. (c) Optical image and (d) enlarged SEM image of the polycrystalline graphene film edge.



Fig. S10 (a, b) Optical images of the single-crystal graphene films transferred to  $SiO_2/Si$  substrates after H<sub>2</sub> etching. (c, d) Optical images of the polycrystalline graphene films transferred to  $SiO_2/Si$  substrates after H<sub>2</sub> etching.