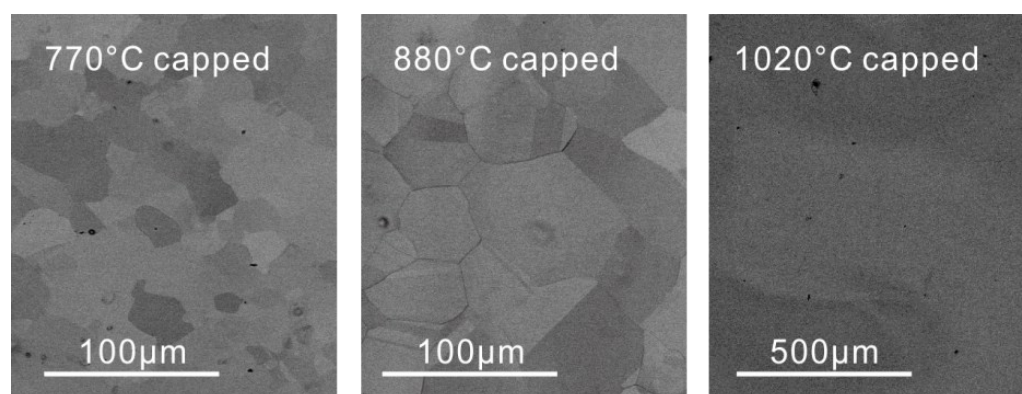


## Recrystallization of copper at a solid interface for improved CVD graphene growth

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### Copper morphology at different annealing temperatures

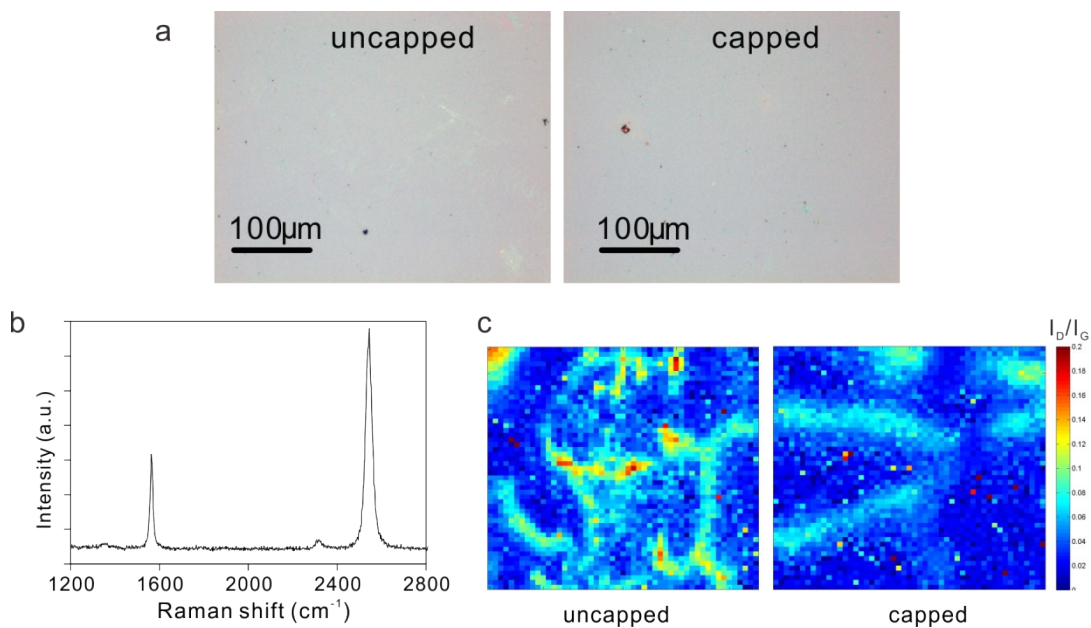
Backscattered electron imaging (BEI) was employed to identify the size of copper grains after 3h annealing.



Suppl. Fig. S1. Representative BEI images of cap annealed Cu foil at different temperatures

### Extended graphene characterization

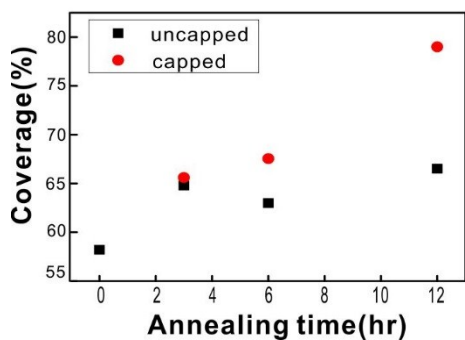
The quality of graphene grown on cap annealed copper foils was compared to traditionally synthesized graphene. For this purpose, graphene was transferred from the growth substrate to 90nm SiO<sub>2</sub> covered Si wafers. Optical micrographs show uniform graphene films with few cracks and bilayer regions which indicates the high quality of graphene (Figure S2(a)). Furthermore, Raman spectra show a small D-band around 1350cm<sup>-1</sup> and sharp G-band and 2D-band features around 1600cm<sup>-1</sup> and 2700cm<sup>-1</sup>, respectively (Figure S2(b)). Finally, high resolution Raman maps (50x50µm) were obtained for capped and uncapped samples. The spatial defect distribution was extracted by fitting the D-Band and G-Band to Lorentzian peaks and their intensity ratio is plotted in Figure S2(c). A larger defect concentration can be observed for graphene grown on uncapped Cu foil.



Suppl. Fig. S2 Characterization of graphene grown on cap-annealed or uncapped Cu foil: (a) Optical micrographs of transferred graphene, (b) representative Raman spectrum, (c) 50x50µm maps of the Raman  $I_D/I_G$  ratio

### Confirmation of graphene continuity

Copper foil coverage was analyzed using film-induced frustrated etching method(FIFE) following previous reports.<sup>1</sup> In brief, copper foil is etched with APS for 20s and rinsed with DI water and dry with  $N_2$ . The resulting etch pits reveal the location and dimension of defects in the graphene.



Suppl. Fig 3. Apparent graphene coverage after 20s etching for different annealing times and conditions

1. Hofmann, M.; Shin, Y. C.; Hsieh, Y.-P.; Dresselhaus, M. S.; Kong, J. *Nano Research* **2012**, 5, (7), 504-511.