

Electronic Supplementary Information

Oxide-assisted growth of scalable single-crystalline graphene with seamlessly stitched millimeter-sized domains on commercial copper foils

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Fig. S1. Growth of aHGDs on different types of commercial Cu foils using oxide-assisted surface monocrystallization. Cu foil product numbers: #13382 and #42972 from Alfa Aesar China Chemical Co., Ltd., #113243 from Nilaco Co., Ltd. All scale bars: 200 μm .



Fig. S2. Growth of aHGDs with prolonged annealing process by H₂. The results show that the alignment of graphene domains does not change by the H₂ annealing, indicating that the surface monocrystallization maintains even with the H₂ annealing process.

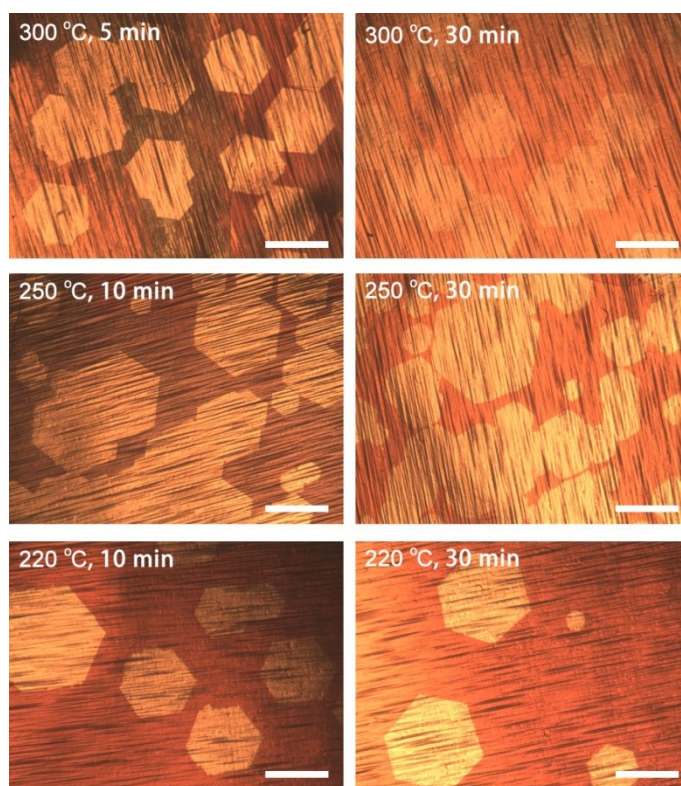


Fig. S3. Growth of aHGDs with different pre-oxidation temperature and time. All the growth periods in the experiments are the same as 30 min. All scale bars: 0.5 mm.

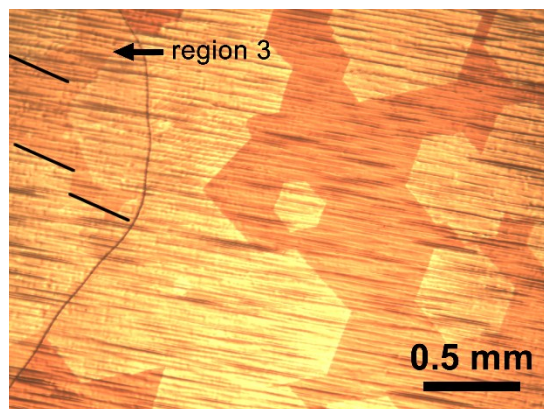


Fig. S4. Enlarged OM image in region 3 for Figure 2b for domain sizes and orientations.

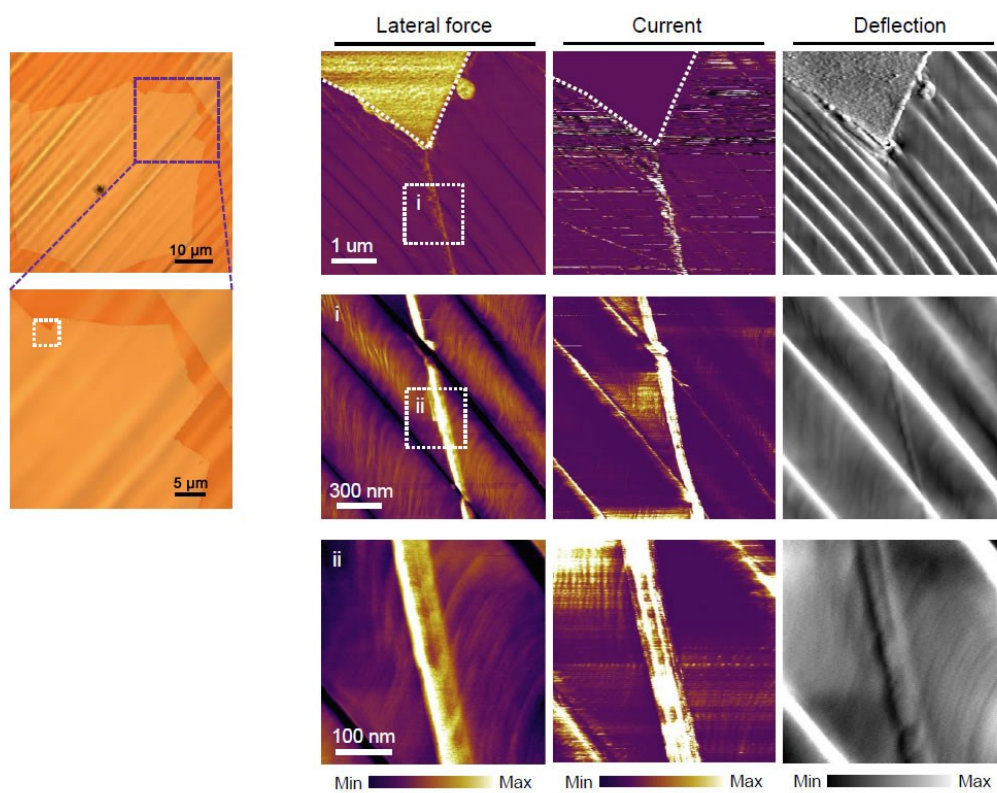


Fig. S5. The AFM lateral force, current and deflection measurements on the coalescence interface of two misoriented domains. Clear enhanced signals were observed in all the images.

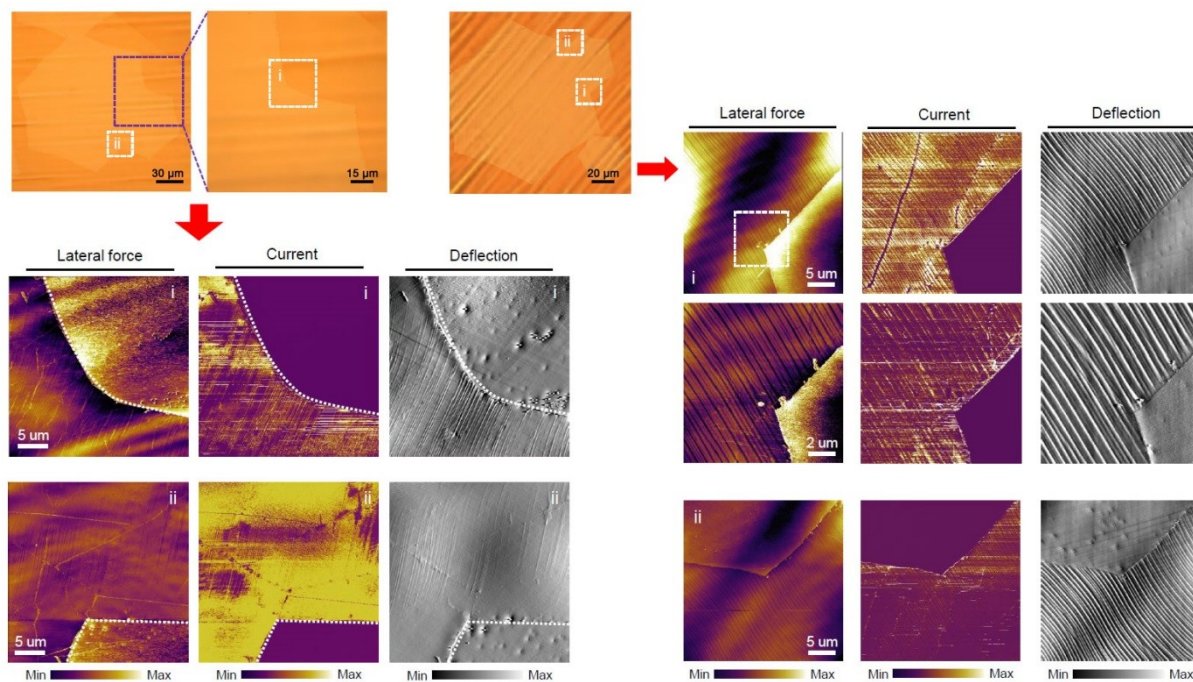


Fig. S6. The AFM lateral force, current and deflection measurements on the coalescence interfaces of different aHGds.

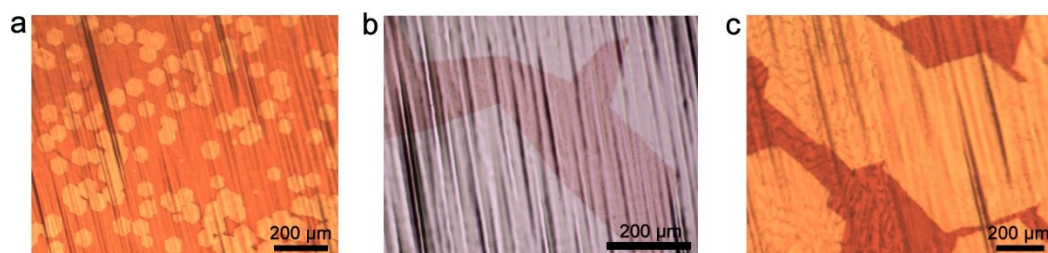


Fig. S7. Control experiments for the investigation of role of surface oxide layer in the growth of aHGds. The experimental condition and results are also summarized in Table S1.

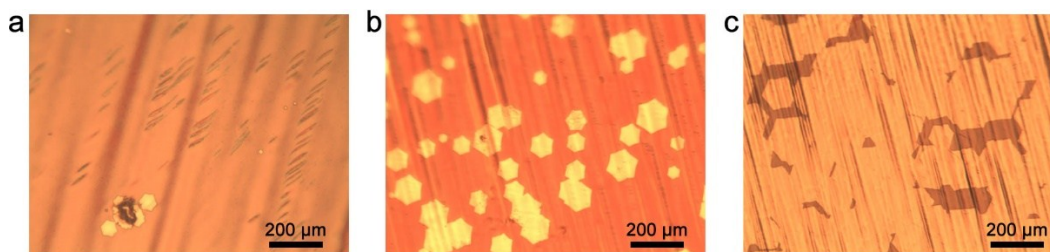


Fig. S8. CVD aHGD growth results with different periods of (a) 5 min, (b) 10 min, and (c) 15 min.

Table S1. Conditions and results for control experiments in Fig. S7

Exp.	Surface oxide	Heat and Anneal	Growth (40 min)	Alignment	Domain size
1	No	Ar/H ₂	Ar/H ₂ /CH ₄	No	Small
2	Yes	Ar/H ₂	Ar/H ₂ /CH ₄	No	Large
3	Yes	Ar then Ar/H ₂	Ar/H ₂ /CH ₄	Yes	Large
4 (general)	Yes	Ar	Ar/H ₂ /CH ₄	Yes	Large