Supporting Material

Stripe distributions of graphene-coated Cu foils and their effects on the reduction of graphene wrinkles

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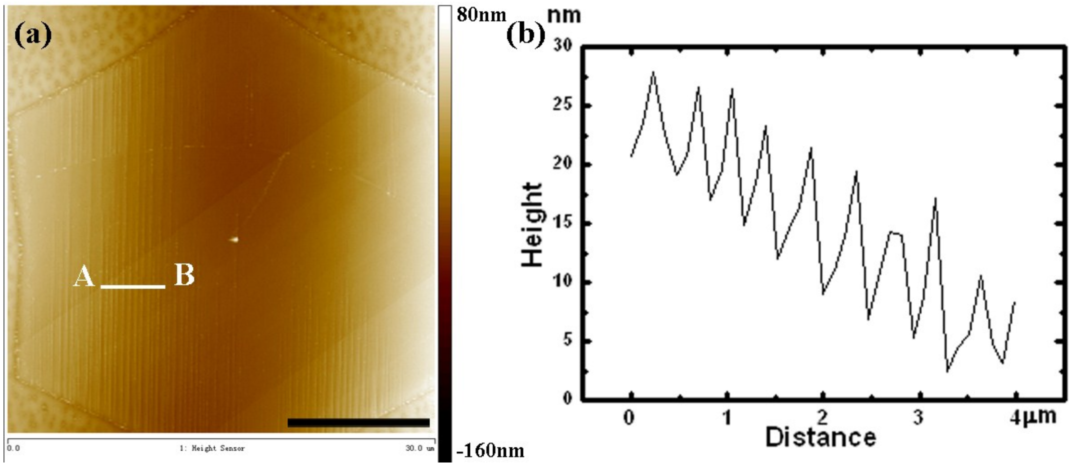

Figure S1. (a) AFM topography image (scale bar, 10 μm) and (b) the corresponding line profile taken along the A–B line.

Parallel stripes were observed on some hexagonal graphene domains. These stripes ran through the whole grain with height and width of approximately 8 nm and 0.5 μm, respectively.

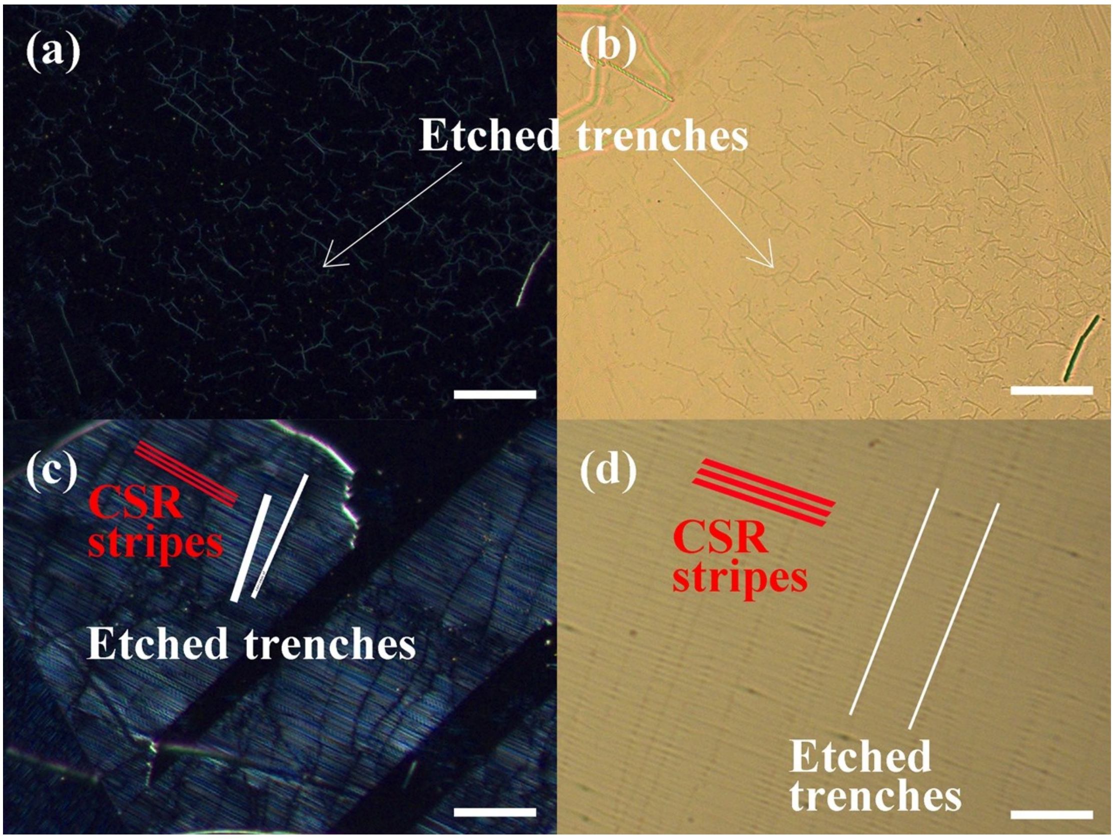

Figure S2. (a) Dark- and (b) bright-field optical images of the etched trenches in non-CSR and those in (c and d) CSR area (not the same area). Scale bar: 100 μm.

Graphene films were etched by hydrogen gas. The results were similar to those of hexagonal single-crystal domains. The reticular etched trenches (figures S2a and S2b) indicated the anisotropy wrinkles. However, the trenches were always perpendicular to the CSR stripes on graphene films (figures S2c and S2d). Hence, the formation of wrinkles along the stripes' direction was suppressed.

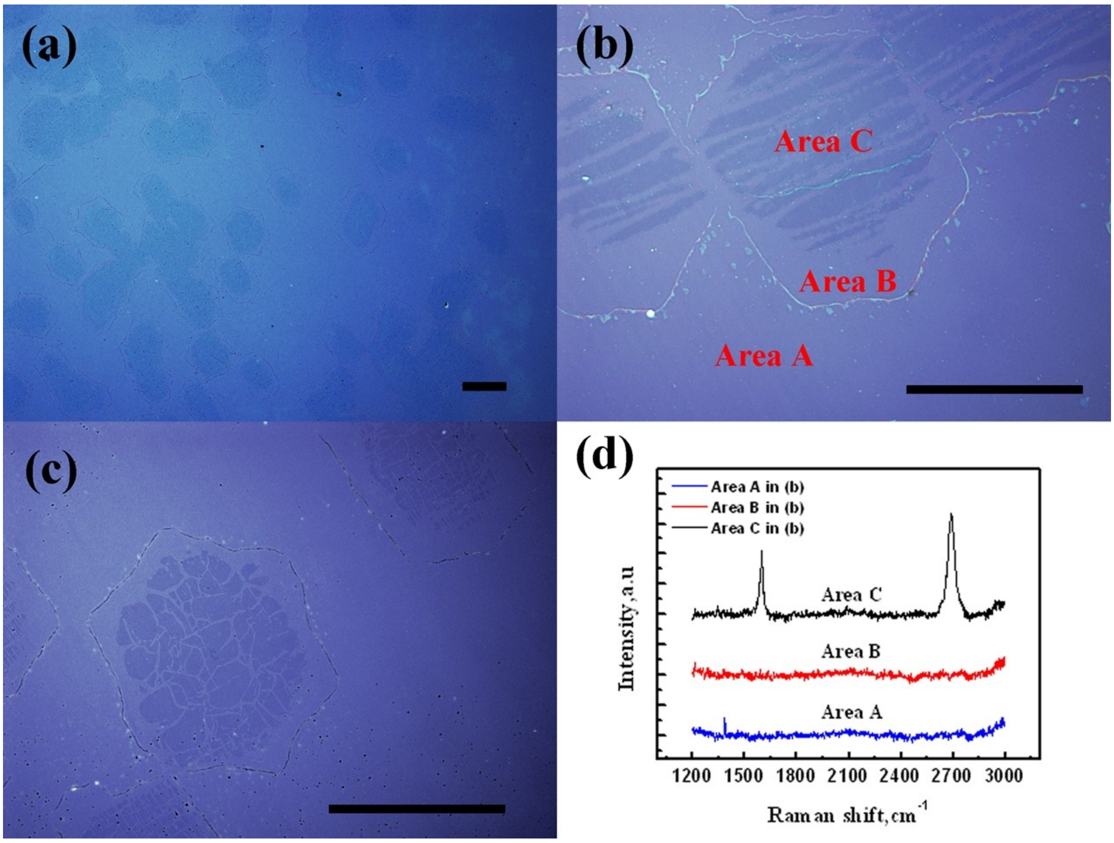

Figure S3. Graphene domains were etched under 1000 sccm Ar and 200 sccm H_2 flow at 1050 °C for 10 min. The samples were then transferred on SiO_2/Si substrate. (a) Optical images of the transferred graphene domains with (b) parallel and (c) reticulate etched trenches. Scale bar: 50 µm. (d) Raman results for the different area in (b).

Graphene underwent thermal hydrogen etching using 1000 sccm Ar and 200 sccm H_2 flow at atmospheric pressure. Subsequently, etching was performed at 900 °C. Partially etched and monolayer graphene domains (figure 2) were transferred to the SiO_2/Si substrate. The original hexagonal edge was observed because of the PMMA residue. The stripes on these domains disappeared (figure S3b). By contrast, CSR stripes were smoothly unfolded when the Cu substrate was dissolved by $FeCl_3$ during transferring.

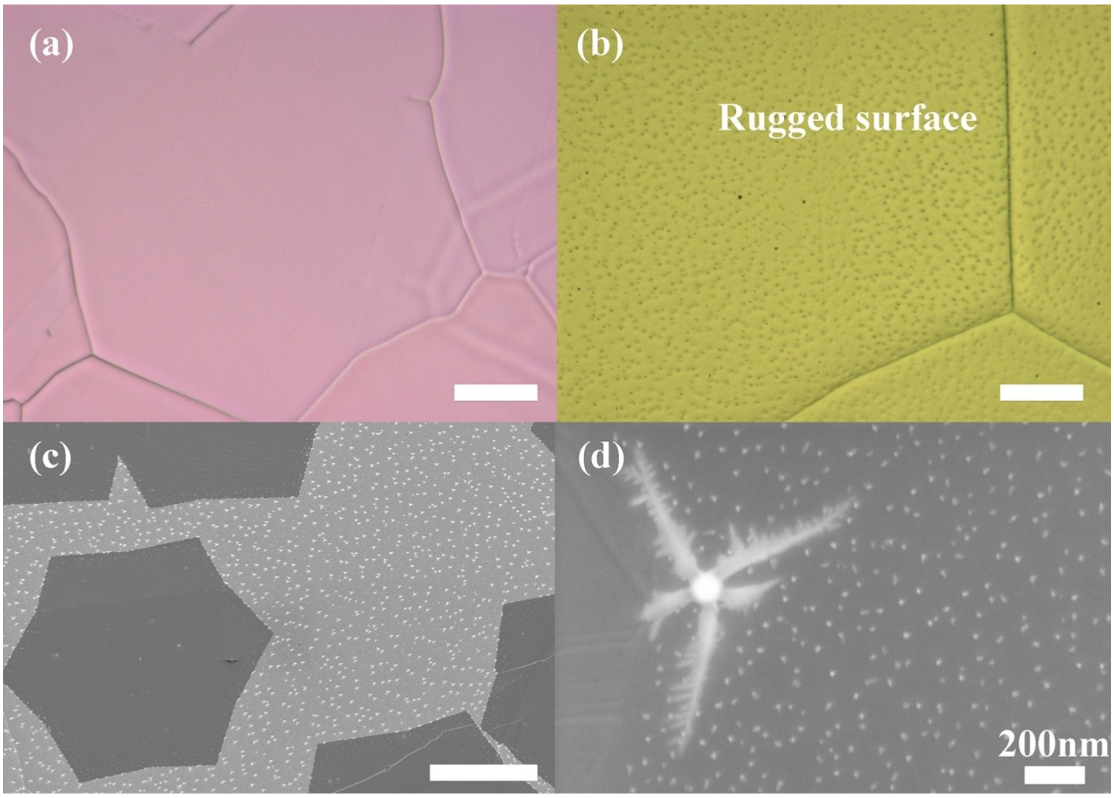

Figure S4. Optical images of Cu surface experiencing (a) fast cooling (200 °C/min); and (b) slow cooling 50 °C/min. (c) SEM images of graphene domains samples experiencing slow cooling. Scale bar: 25 μm. (d) Magnified image of white dendrite-like structures in (c). Scale bar: 200 nm.

Samples experienced fast cooling result in flat surface while the ones experienced slow cooling exhibits rugged surface. The rugged surface is covered with dendrite-like structures shown in (d). Paronyan et.al. believe high temperature result in a thin melted surface layer on the Cu foil with unstable solid-melt interface and these dendrite-like structures are cause by small perturbation in the course of solidification due to the thermal quenching.(In ref.30)

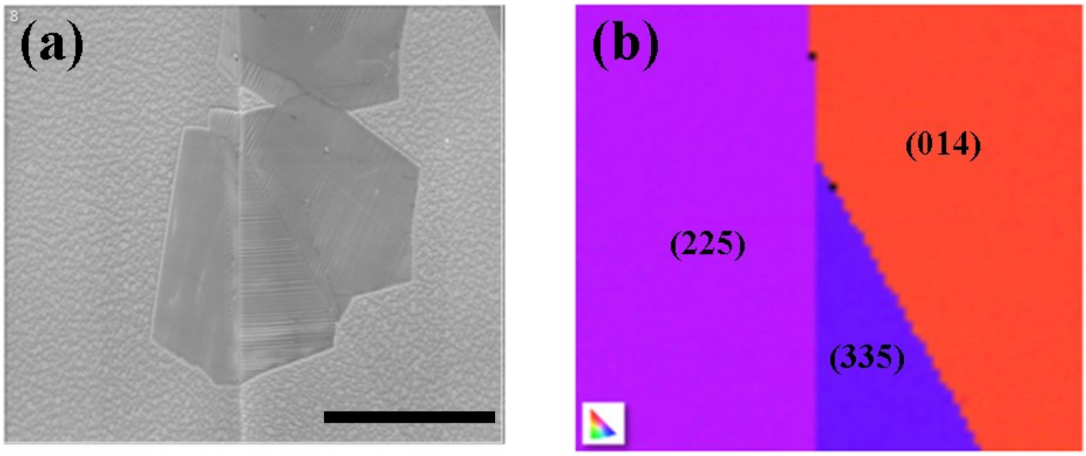

Figure S5. (a) SEM images of the distribution of stripes with Cu grains. (b) EBSD results showing the Cu grain facet index in the same region as (a). Scale bar: 25 μm.

Cu (014) and Cu (335) resulted in the different directions of CSR stripes. These stripes were cut off by the Cu boundary. CSR stripes were not observed on Cu (225), which suggested that these stripes were determined by the distribution of Cu grains.

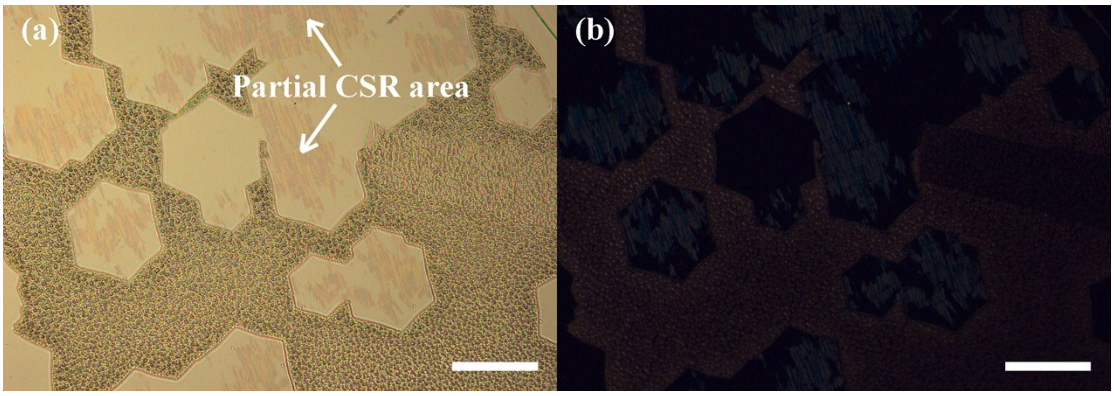

Figure S6. (a) Bright- and (b) dark-field images of the partial CSR stripes on graphene domains. Scale bar: 100 μm.

During slow cooling, numerous CSR stripes failed to run through the whole grain. The partial stripes were formed in Cu grains where CSR could not easily occur because the slow cooling rate provided enough time for the surface to shrink.

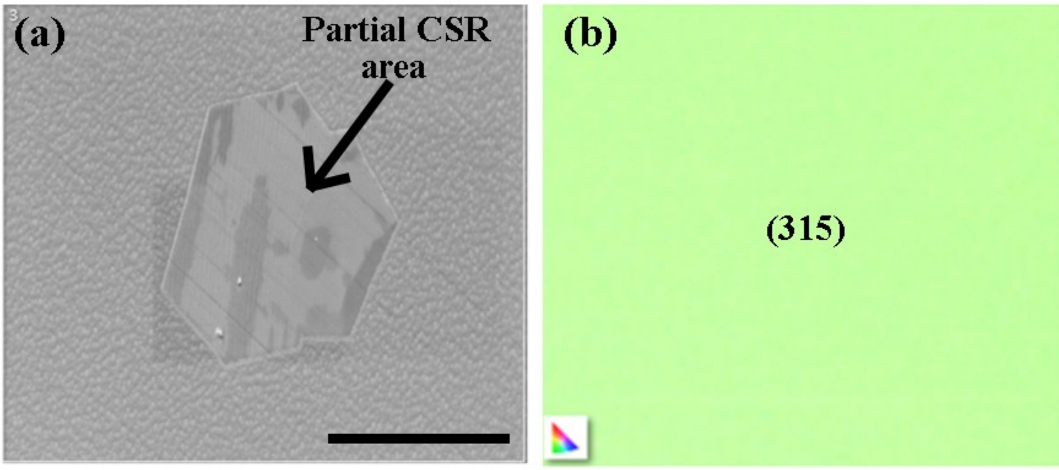

Figure S7. (a) SEM images of the distribution of stripes with Cu grains. (b) EBSD results showing the Cu grain facet index in the same region as (a). Scale bar: 25 μm.

Partial CSR areas were found in one Cu grain by slowing down the cooling process. The CSR stripes were associated to the Cu grains. However, EBSD results implied that these Cu grains contributed to CSR and non-CSR areas. Hence, the cooling rate promoted the CSR phenomenon.