



Extended Data Fig. 1. | Comparison of grain boundaries (GBs), twin boundaries (TBs), and tilted twin boundaries (TTBs) observed in three different Cu thin films. a, EBSD map of polycrystalline and **b,** misorientation line map of the GB-dominant Cu thin film. **c,** Schematic illustration showing the crystallographic relationships near the representative regions indicated in the GB case. **d, g,** EBSD maps of single-crystalline Cu thin films exhibiting perfect alignment along the out-of-plane (111) direction. **e, h,** Misorientation line maps of **d** and **g** consisting predominantly of TBs, including **e** regions containing slightly TTBs and **h** regions composed

exclusively of coherent TBs without tilted segments. Schematic illustrations of the local microstructures associated with **f.** TTB and **i.** coherent TB.