Electronic Supplementary Material (ESI) for Nanoscale.


# Supplementary Information

## Synthesis of sub-millimeter single-crystal grains of aligned hexagonal boron nitride on epitaxial Ni film

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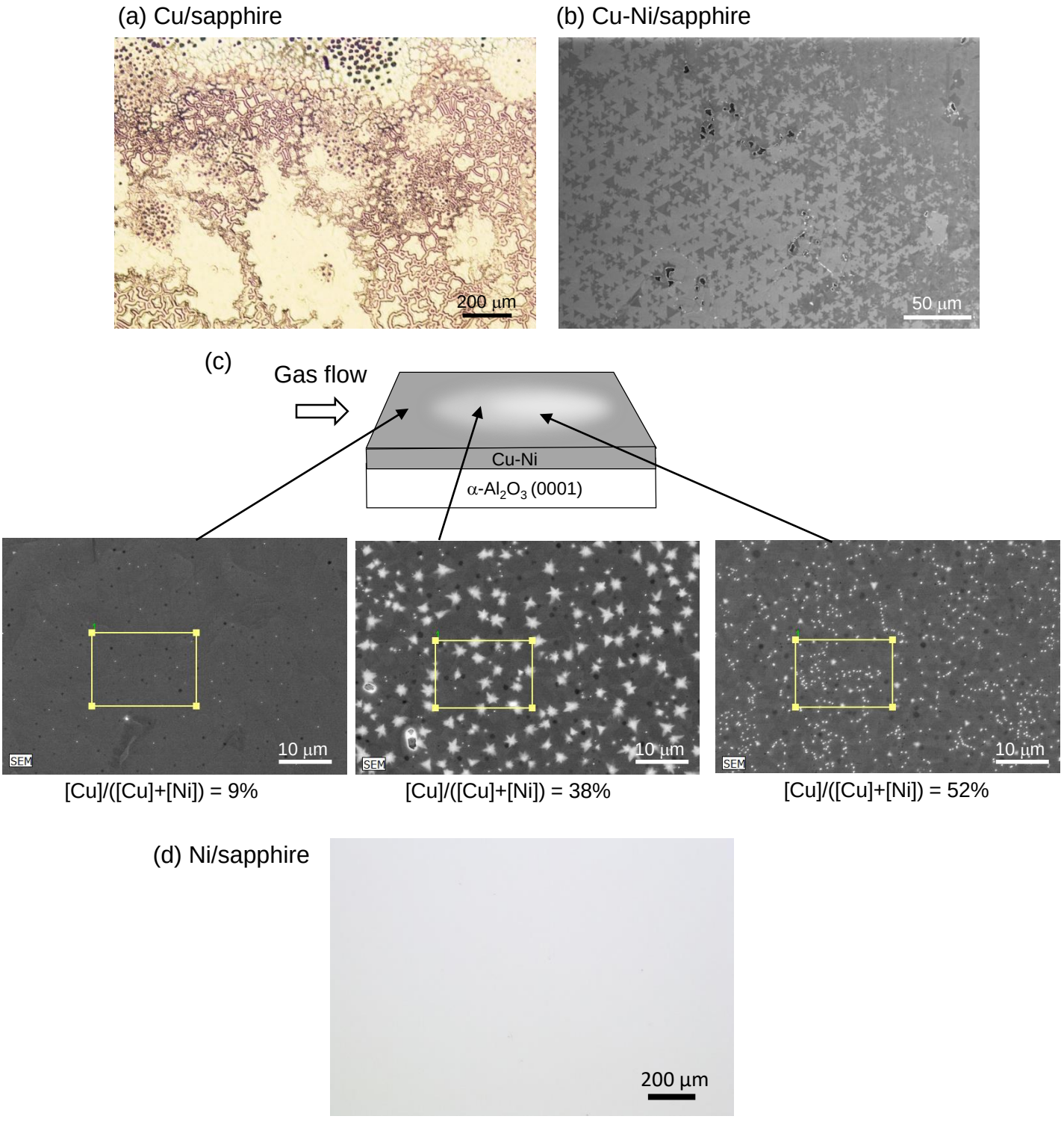



**Figure S1.** (a) Optical micrograph of the Cu(111) surface after the CVD at 1050 ºC. Severe evaporation of Cu makes rough surface with many pits reaching to the underlying sapphire substrate. (b) SEM image of the Cu-Ni(111) surface after the CVD at 1050 ºC. A number of very small h-BN grains were observed. (c) Schematic of the Cu-Ni surface after the CVD and the SEM images taken at the marked positions. The initial Cu concentration, *i.e.* [Cu]/([Cu]+[Ni]), of the alloy film is ~80%. The concentration of Cu was strongly dependent on the substrate position, indicating the significant evaporation of Cu from the alloy film during the CVD process. The spatial distribution of Cu concentration suggests that the Cu evaporation is enhanced by the borazine vapor. (d) Optical micrograph of the Ni(111) surface after the CVD at 1100 ºC, which shows clean and flat surface.

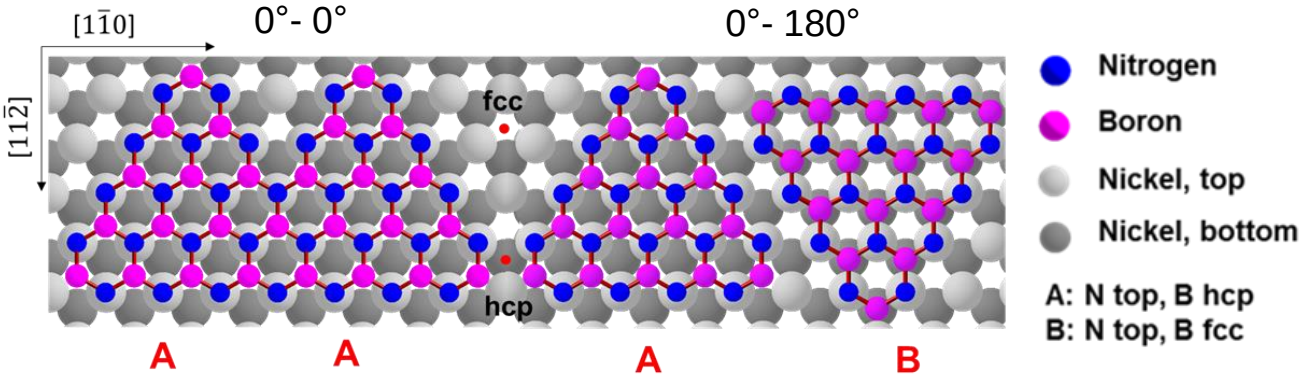
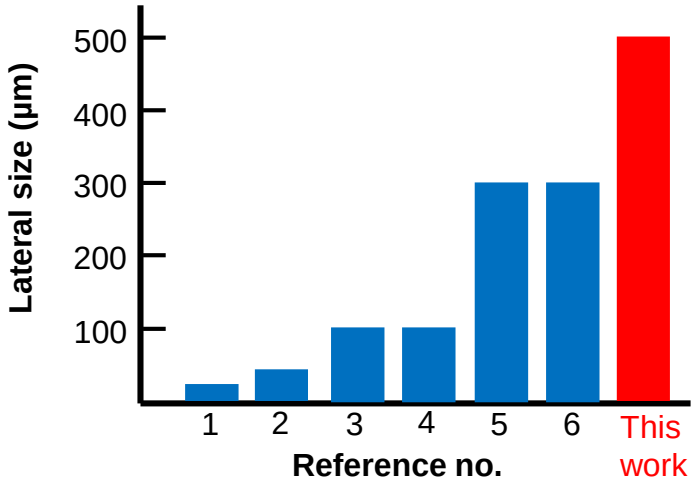



**Figure S2.** Atomic model of aligned h-BN grains on the Ni(111) surface.




[1] S. Caneva *et al., Nano Lett*. **16**, 1250 (2016)
[2] J. Yin *et al., Small* **11**, 4497 (2015)
[3] G. Lu *et al., Nat. Commun*. **6**, 6160 (2015)
[4] Q. Wu *et al*., *Sci. Rep*. **5**,16159 (2015)
[5] S. Caneva *et al., Nano Lett.* **15**, 1867 (2015)
[6] Y. Ji *et al., ACS Nano* **11**, 12057 (2017)

**Figure S3.** Comparison of the grain size of monolayer h-BN grown by thermal CVD method.

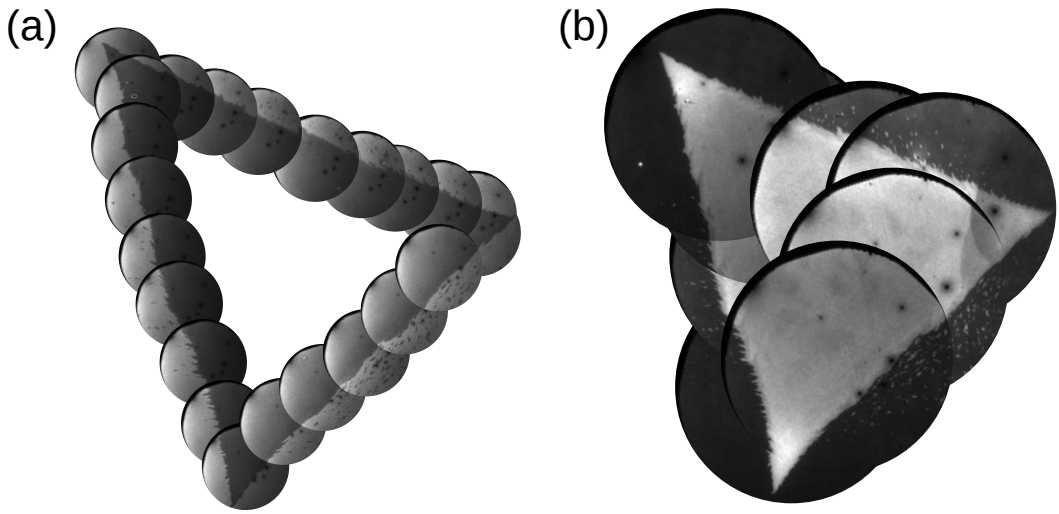



**Figure S4.** (a) BF-LEEM and (b) PEEM images of the same h-BN grain shown in Figure 3a. Due to large grain size, both images are composed of multiple view images. The LEEM images at the central area of (a) are omitted. Fields-of-view are 40 μm and 100 μm for BF-LEEM and PEEM images, respectively.

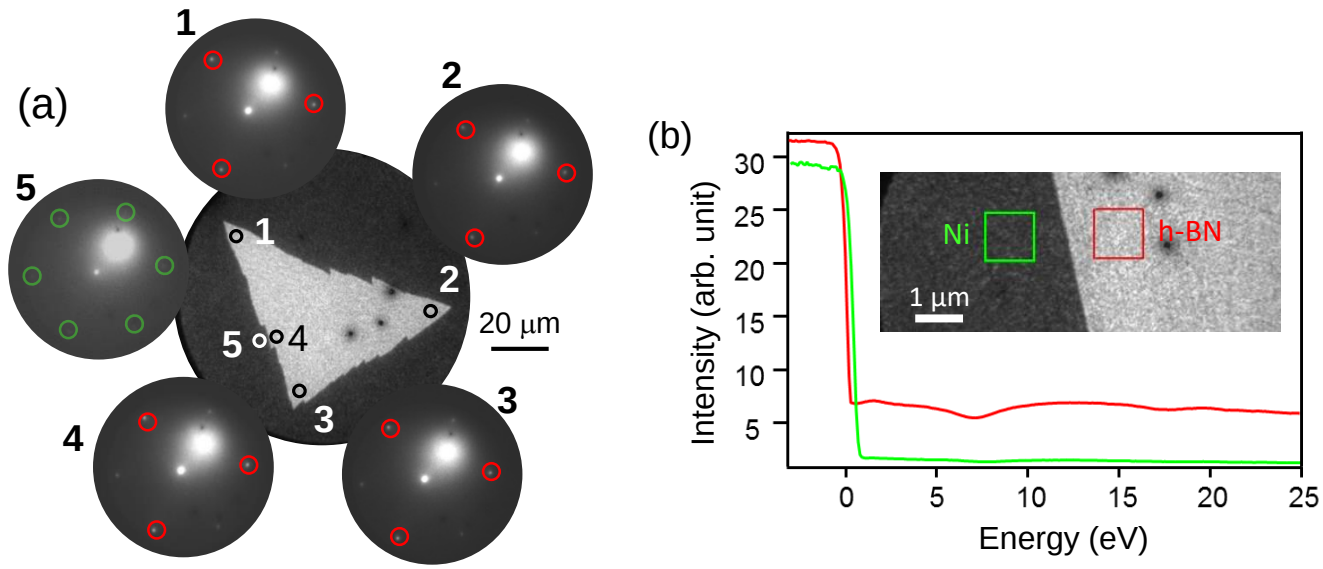



**Figure S5.** (a) BF-LEEM image and corresponding diffraction patterns. The h-BN grain is different from that shown in Figure 3a. (b) Electron reflectivity curves taken at h-BN and Ni surfaces of the grain shown in (a).

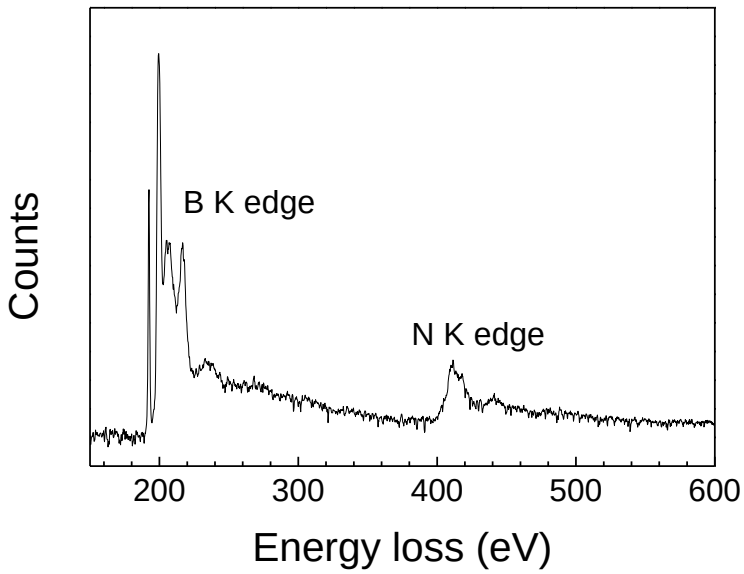



**Figure S6.** EELS spectrum collected from the sample shown in Figure 4d.

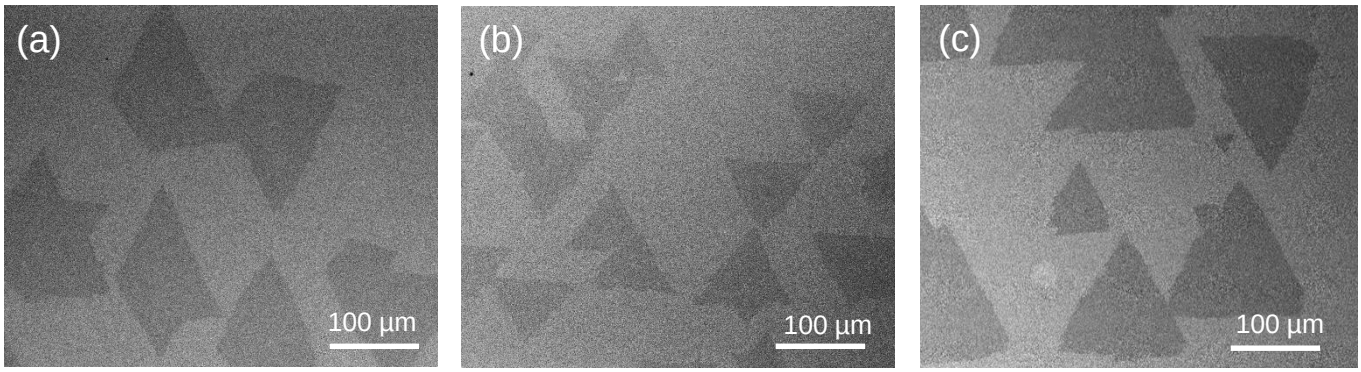



**Figure S7.** Influence of $N_2$ gas added during the h-BN growth. The concentration of $N_2$ gas: (a) 0 vol%, (b) 10 vol%, and (c) 25 vol%.

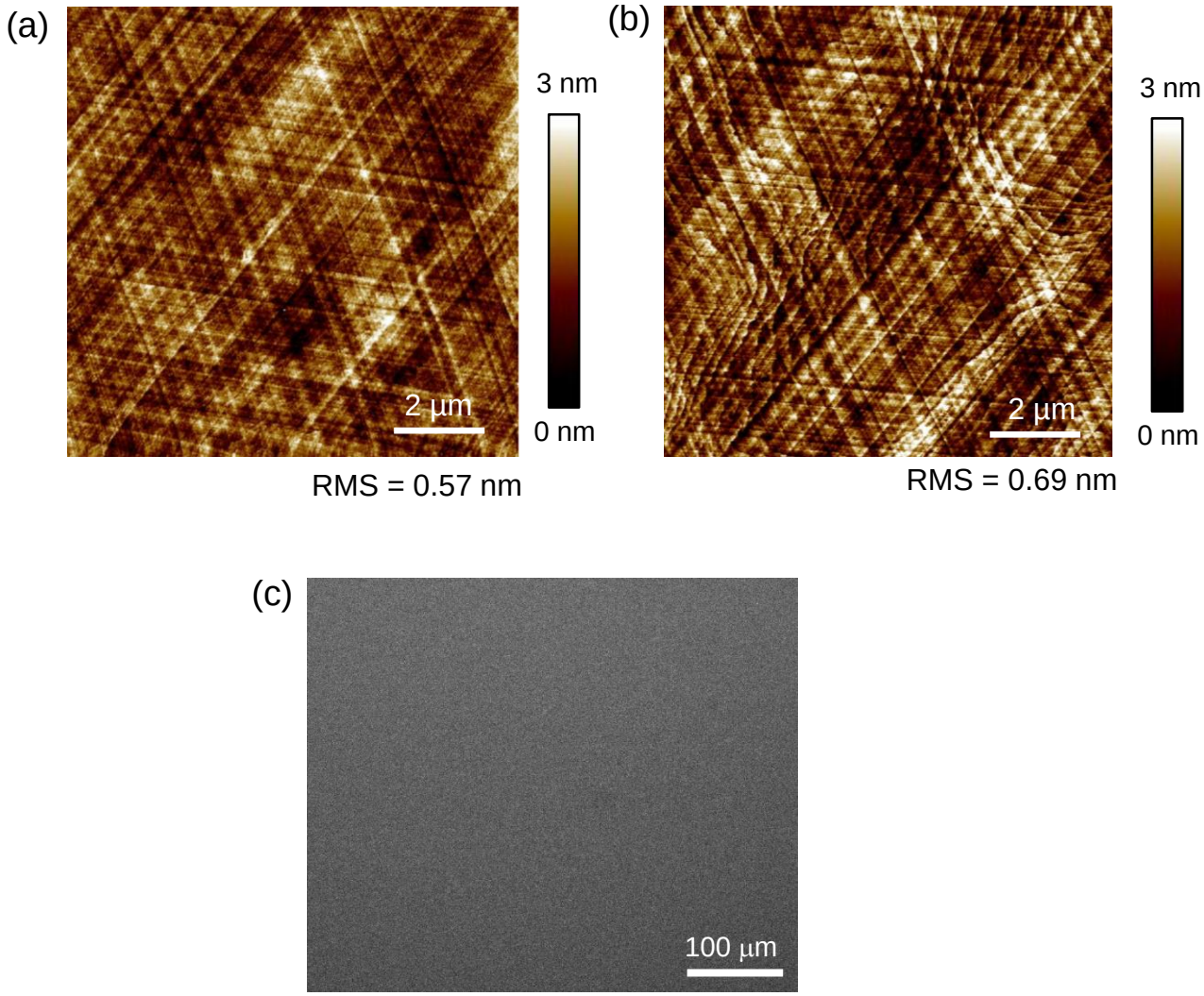



**Figure S8.** AFM images of Ni(111) surface after annealing $H_2$ gas at 1000 Pa (a) and 30 Pa (b). The Ni annealed at 30 Pa shows rougher surface than that annealed at 1000 Pa, strongly suggesting Ni evaporation at the low pressure (30 Pa). The RMS value was measured for the whole scanned area. (c) SEM image of the Ni(111) surface after the CVD performed at 30 Pa. As the rough Ni surface increased the nucleation density, the Ni surface was fully covered with h-BN after the CVD under the standard condition (10 min at 1100 ºC).