

## Electronic Supplementary Information

### A method for covering a substrate with highly-oriented single crystalline hexagonal zinc structures under ambient pressure and room temperature

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#### A. Materials and experimental procedures

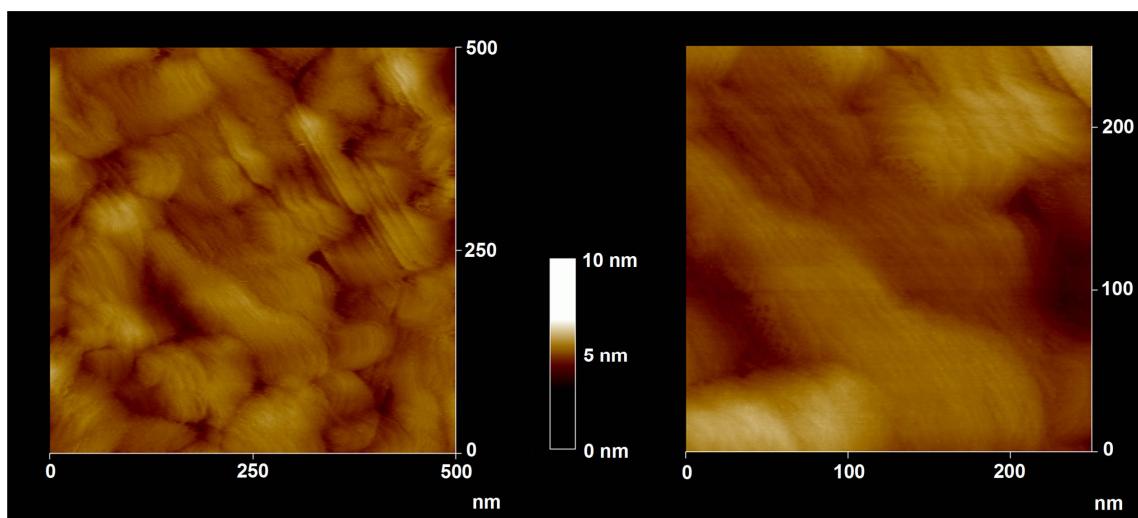
All chemicals used in this study were of analytical grade and were used without further purification. A 100 mL transparent aqueous solution containing 0.01 M zinc acetate dihydrate ( $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ , 99%, Samchun) and ammonia water (28.0~30.0 wt%, Samchun, 5 mL) was produced at room temperature (pH 11.37). To prepare the substrate, a 100 nm thick Al film was deposited as the reductant on a Si wafer by evaporation in a vacuum chamber. The substrate was transferred into the solution and maintained at 25 °C for 6 h. After the reaction, the substrate was removed from the solution and washed with deionized water, after which it was dried in an oven.

#### B. Characterizations

The morphology, crystallinity, crystalline nature, and chemical composition of the samples were determined by using a field-emission scanning electron microscope (FESEM, JEOL, JMS-7400F, operating at 10 keV), X-ray diffraction (XRD, Mac Science, M18XHF), a high-resolution scanning transmission electron microscope (Cs corrected HR-STEM, JEOL, JEM-2200FS with an energy-dispersive X-ray spectrometer, operating at 200 kV), X-ray photoelectron spectroscopy (XPS, VG Scientific, EscaLab 200iXL), Scanning tunneling microscope (STM, Digital Instruments, Nanoscope IIIa) and inductively coupled plasma atomic emission spectroscopy (ICP-AES, Spectro, SPECTRO FRAME MODULA E).

### C. Al film characterizations

We characterized the Al deposited Si substrate before a reaction by using field-emission scanning electron microscope (FESEM), Scanning tunneling microscope (STM), X-ray photoelectron spectroscopy (XPS) and inductively coupled plasma atomic emission spectroscopy (ICP-AES).



**Fig. S1.** STM image of the Al deposited Si substrate before a reaction.

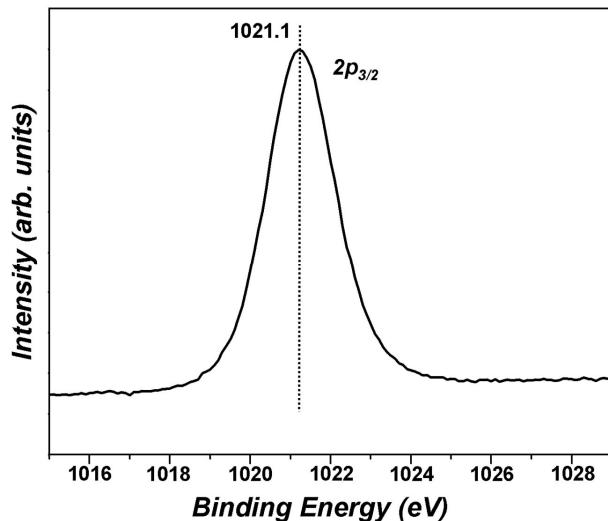
Surface topology was characterized with STM (Fig. S1). The thickness of Al film deposited on the Si wafer is  $100\text{ nm} \pm 5\text{ nm}$  (from SEM observations of substrate cross-section view). From XPS, the thickness of native oxide ( $\sim 1.9\text{ nm}$ ) was calculated using the Strohmeier Equation. Quantitative chemical analysis with (ICP-AES) revealed that the composition of Al film before a reaction has values of 98.88at.%: 1.12at%. (Al: O). Native oxides of the metal films consumed in replacement reactions, in which the more reactive component is consumed without any external electron source, have not been significantly considered.<sup>C1,C2</sup>

### Reference

- C1. L. A. Poter, Jr., H. C. Choi, A. E. Ribbe and J. M. Buriak, *Nano Lett.*, 2002, **2**, 1067.  
C2. Y. Sun and Y. Xia, *J. Am. Chem. Soc.*, 2004, **126**, 3892.

#### D. XPS analysis of hexagonal zinc structures

We characterized the products after reaction with an aqueous ammonia solution containing Zn ions at 25 °C for 6 h by using X-ray photoelectron spectroscopy (XPS).



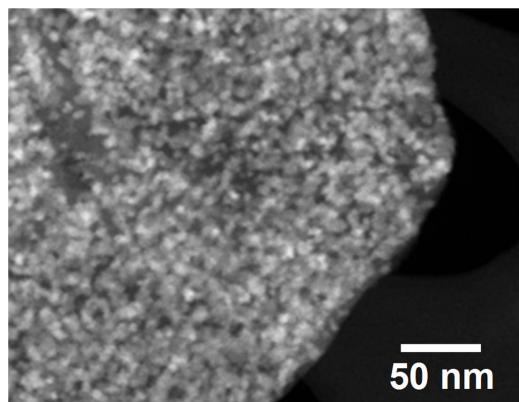
**Fig. S2.** X-ray photoelectron spectrum in the Zn 2p region of the products after reaction with an aqueous ammonia solution containing Zn ions at 25 °C for 6 h.

Fig. S2 shows an X-ray photoelectron spectrum in the Zn 2p region. The location of 2p<sub>3/2</sub> peak (binding energy) indicates that Zn species in the products dominantly exist as Zn(0), not Zn(II).<sup>D1,D2</sup>

#### References

- D1. J. Cheon, L. H. Dubois and G. S. Girolami, *Chem. Mater.*, 1994, **6**, 2279.  
D2. Y. Zhu, H. Wang and P. P. Ong, *J. Phys.: Condens. Matter*, 2001, **13**, 787.

**E. High-angle annular dark-field scanning transmission electron microscope (HAADF-STEM) image**



**Fig. S3.** A HAADF image of a Zn structure obtained after reaction with an aqueous ammonia solution containing Zn ions at 25 °C for 20 min.

The HAADF image (Fig. S3) shows that the structure, which was synthesized after reaction with an aqueous ammonia solution containing Zn ions at 25 °C for 20 min, is composed of nanoparticles.