

## Supplementary information

### Size- and Composition-Controlled Pt-Sn Bimetallic Nanoparticles Prepared by Atomic Layer Deposition

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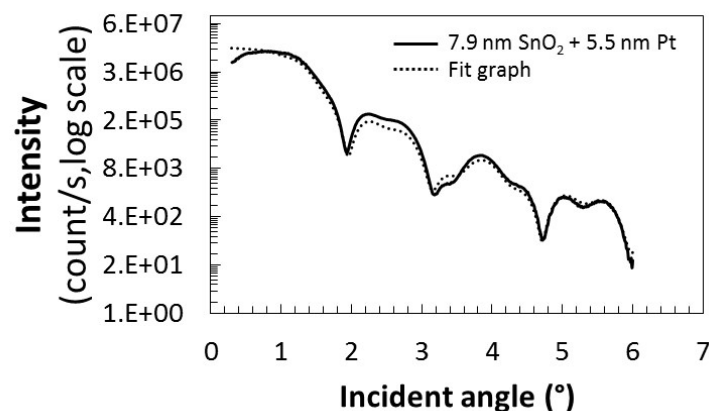
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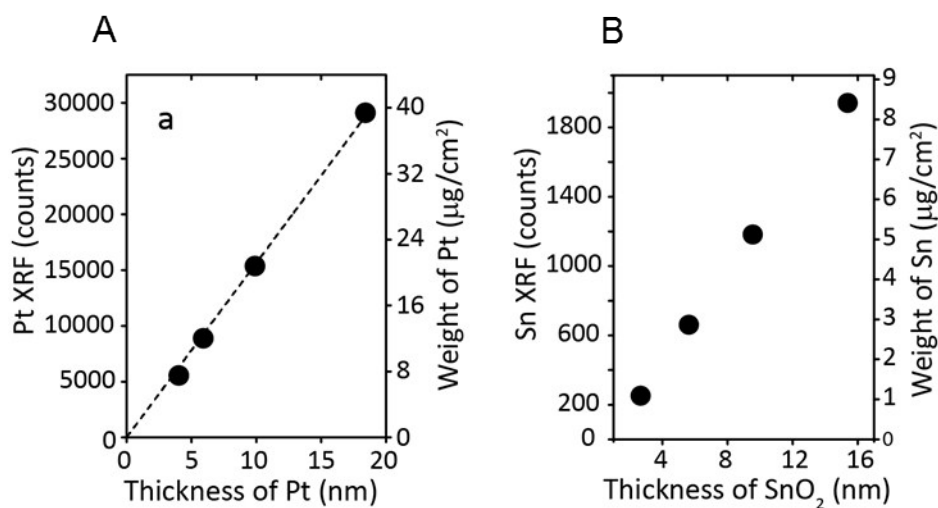
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#### Composition determination.

The thickness of the bilayer samples was determined using X-ray reflectivity (XRR) measurements, as shown in **Figure S1** for a bilayer consisting of 7.9 nm SnO<sub>2</sub> and 5.5 nm Pt. From the thickness, the specific weight of Pt and Sn was calculated by assuming the densities of the films equal to that of bulk Pt and SnO<sub>2</sub>, respectively. Alternatively, and especially for thinner samples for which XRR analysis becomes difficult, the thickness was extracted from calibrated X-ray fluorescence data. For this, a series of pure films of Pt and SnO<sub>2</sub> with different thicknesses were deposited on planar SiO<sub>2</sub> substrates and their thickness was determined by XRR. XRF measurements were performed on each film and the respective fluorescence signal (Pt L $\alpha$  and Sn L $\alpha$ ) was integrated over a period of 100 s. The XRF intensity counts and specific weight of Pt and Sn were plotted against the thickness of the films (**Figure S2**), yielding calibration curves that could be used for determining the composition of the bilayer materials.



**Figure S1.** X-ray reflectivity measurement for a bilayer sample with 7.9 nm SnO<sub>2</sub> and 5.5 nm Pt.

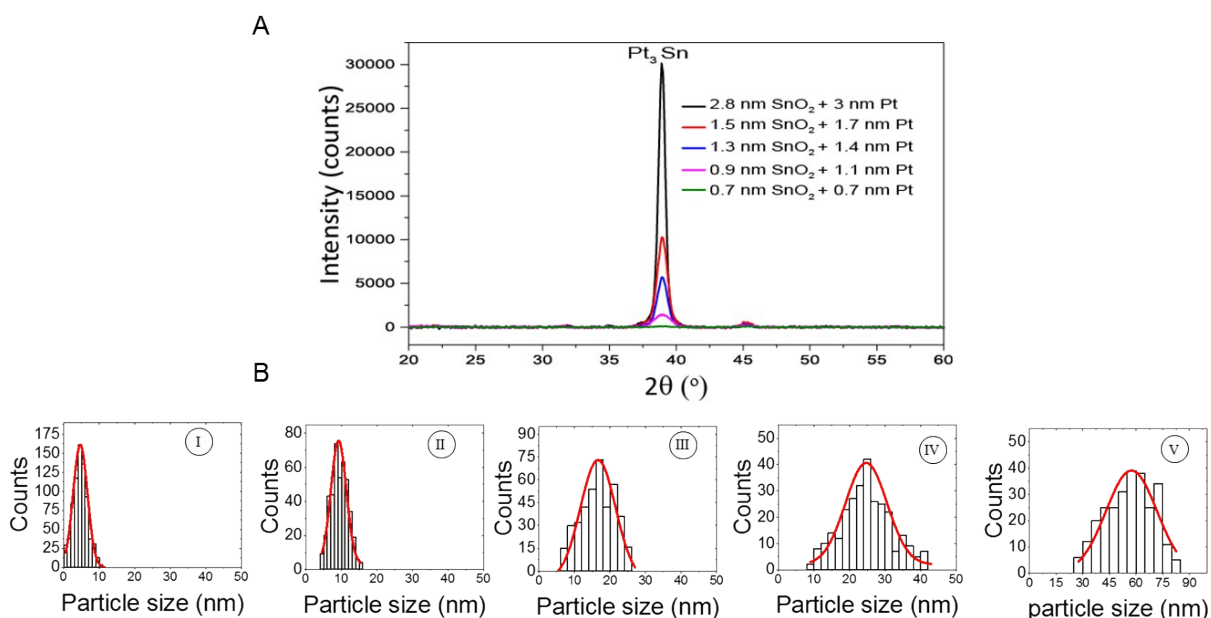


**Figure S2.** Variation of Pt (A) and Sn (B) L $\alpha$  XRF intensity (left y-axis) and weight (right axis) against the Pt and SnO<sub>2</sub> film thickness.

As a consequence of the nucleation-controlled growth mode of Pt ALD on oxide surfaces, a 3D island morphology is observed for low amounts of Pt ( $< 5 \mu\text{g}/\text{cm}^2$ ). In this case, the Pt thickness obtained from the relation should therefore be considered as an equivalent thickness. The equivalent thickness equals the thickness of a 2D Pt thin film containing the same number of Pt atoms as the real deposited 3D Pt morphology.

### Pt<sub>x</sub>Sn<sub>y</sub> size tuning experiment.

A series of bilayer films with a constant Pt/(Pt+Sn) atomic ratio of *ca.* 75%, but different total thickness, ranging from 1.4 to 5.8 nm, was subjected to a TPR up to 700°C in 10% H<sub>2</sub>/N<sub>2</sub>. The formation of Pt<sub>3</sub>Sn was confirmed by XRD measurements (**Figure S3 A**). The morphology of the films was studied by scanning electron microscopy (SEM), where the particle size distribution was obtained using the ImageJ software. A Gaussian fit to the particle size distribution was made for determining the average particle size and the half of the full width at half maximum (FWHM) was taken as the standard deviation of the particles size distribution. **Figure S3 B** shows the particle size distributions and the corresponding Gaussian fits.



**Figure S3.** (A) XRD patterns and (B) particle size distribution calculated from the SEM images measured after TPR in 10 % H<sub>2</sub>/N<sub>2</sub> up to 700 °C of Pt/SnO<sub>2</sub> bilayers with Pt/(Pt+Sn) atomic ratios of *ca.* 75 % and total thickness of 1.4, 2.0, 2.7, 3.2 and 5.8 nm. The red curves show the Gaussian fit to the particle size distribution.