

## Supporting Information

### Conversion of Anisotropically Phase-segregated Pd/ $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> Nanoparticles into Exchange-coupled fct-FePd/ $\alpha$ -Fe Nanocomposite Magnets

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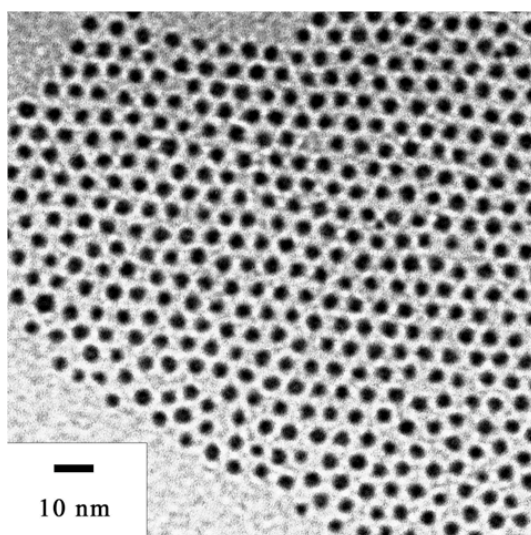
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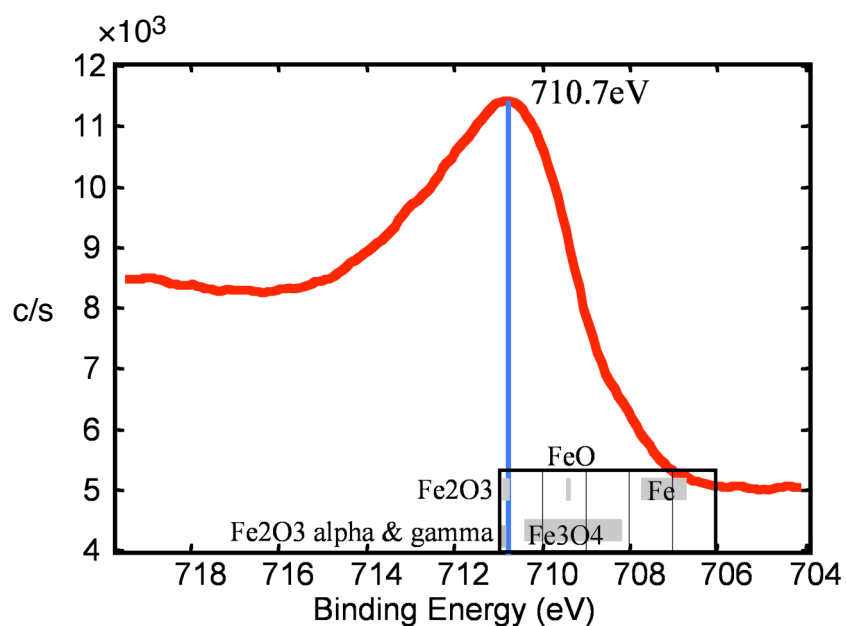
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#### Synthesis of monodisperse 4.9±0.3 nm Pd nanoparticles

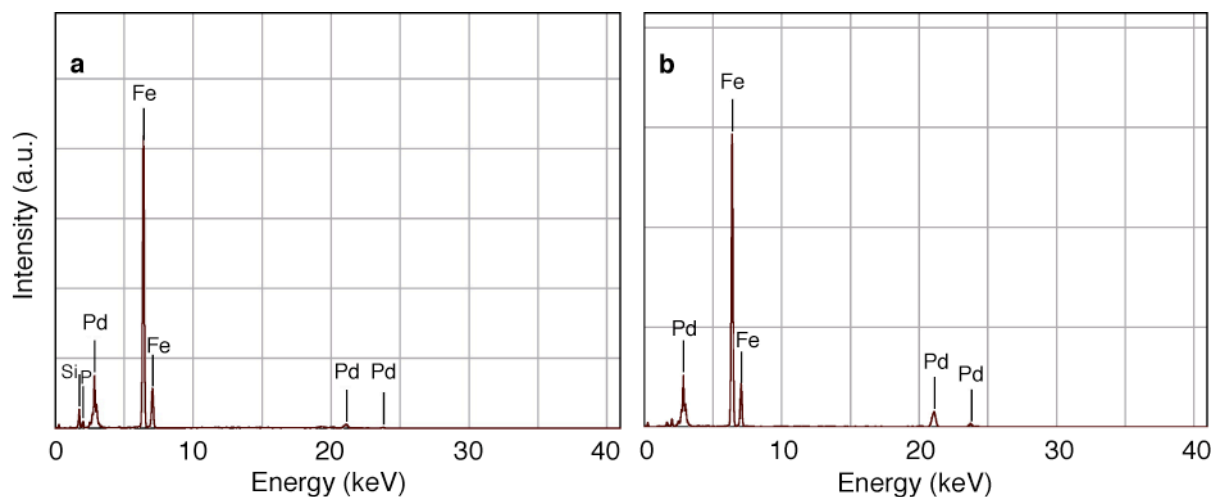
Monodisperse Pd nanoparticles of 4.9±0.3 nm in size were synthesized as follows: 100 mg of Pd(II) acetylacetonate was added into 1 mL of triocrylphosphine (TOP) and the mixture was stirred to completely dissolve the precursor. The solution was added to 10 mL of distilled oleylamine, and the resulting solution was slowly heated to 250 °C and was aged at that temperature for 30 min. The initial orange color of the solution was gradually changed to dark brown. The resulting solution was cooled to room temperature and 50 mL of ethanol was added to yield the pure 4.9 nm TOP-protected Pd (TOP-Pd) nanoparticles as a black precipitate, which was then separated by centrifuging.



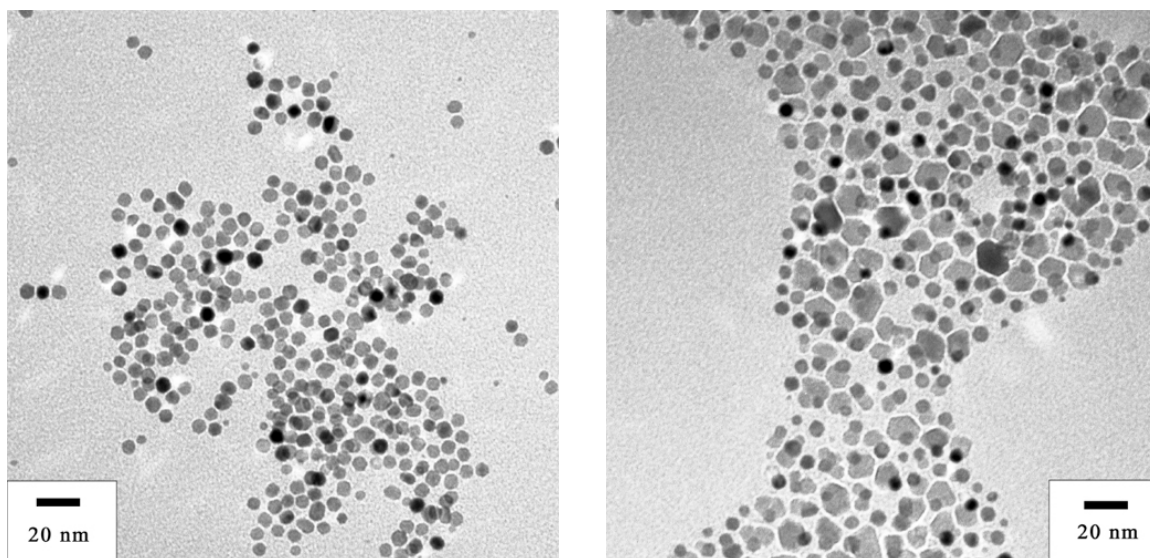
**Figure S1.** TEM image of 4.9±0.3 nm TOP-Pd nanoparticles.



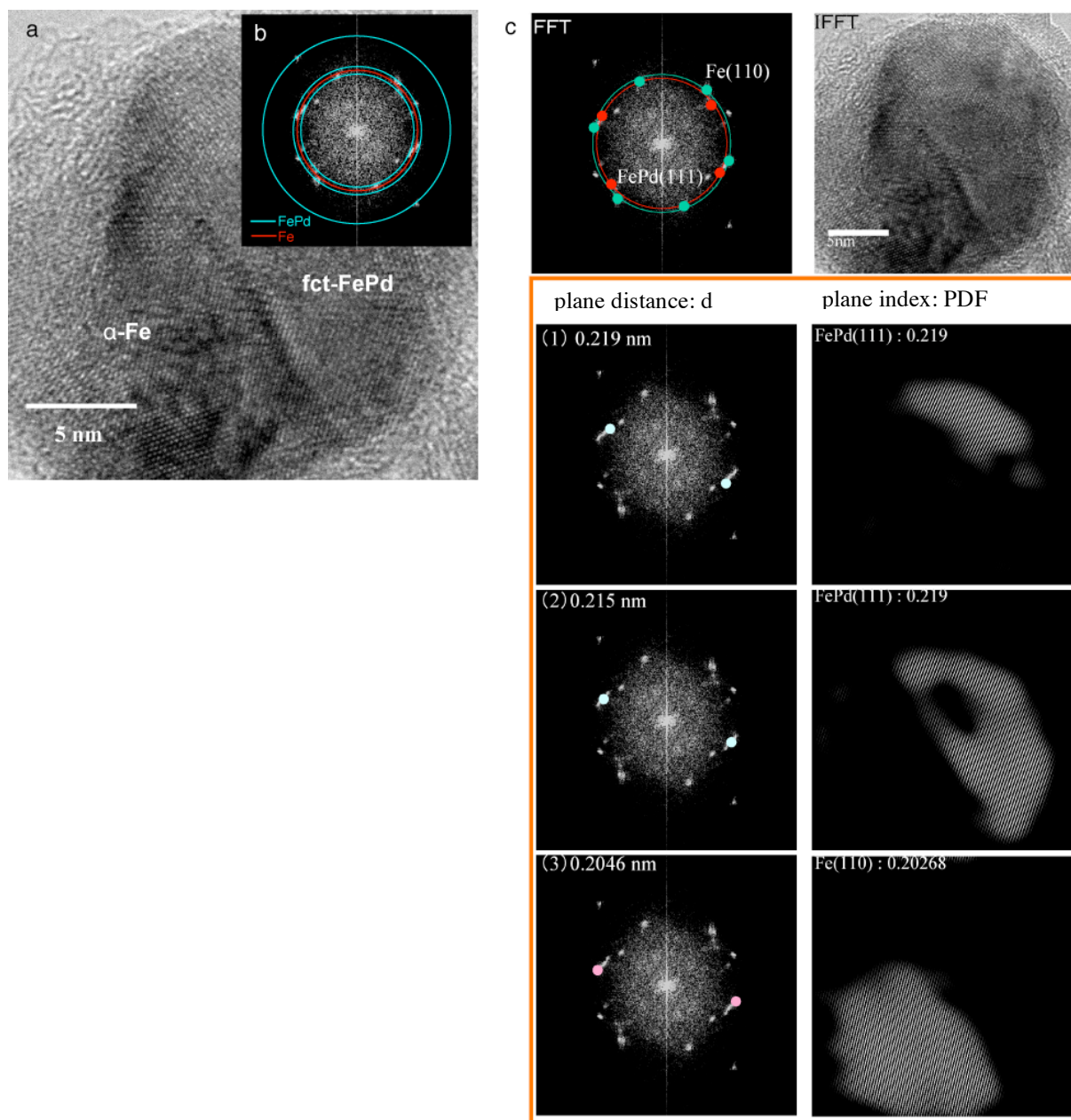
**Figure S2.** Fe $2p_{3/2}$  core-level XPS spectrum for anisotropically phase-segregated Pd/Fe $_2$ O $_3$  nanoparticles. The element ratio of Fe/Pd collected from XPS data is 79/21.



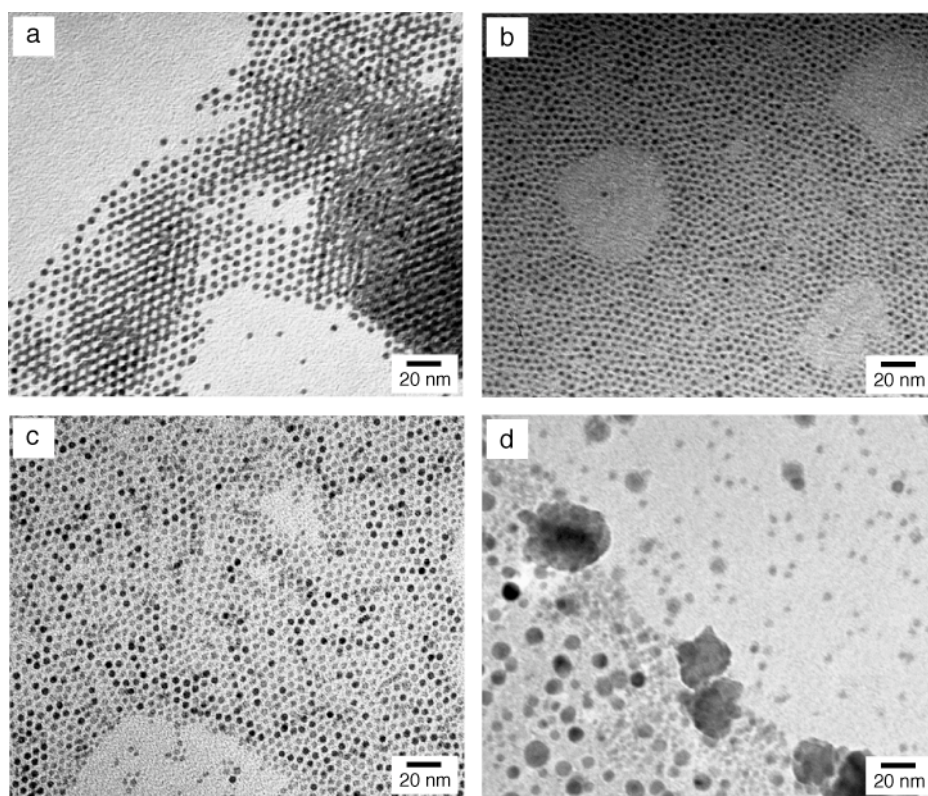
**Figure S3.** X-ray fluorescence spectra for (a) anisotropically phase-segregated Pd/Fe $_2$ O $_3$  nanoparticles and (b) fct-FePd/ $\alpha$ -Fe nanocomposite magnets.



**Figure S4.** TEM images of (a)  $7.1 \pm 0.9$  nm PVP-Pd nanoparticles and (b) anisotropically phase-segregated Pd/Fe<sub>2</sub>O<sub>3</sub> nanoparticles.



**Figure S5.** (a) HRTEM image, (b) FFT image, and (c) inverse-FFT images of fct-FePd/ $\alpha$ -Fe nanocomposite magnets. The inverse-FFT analysis in (c) shows that the (111) lattice plane of fct-FePd hard phase gradually deforms and connects with (110) plane of  $\alpha$ -Fe soft phase, demonstrating an effective exchange coupling.



**Figure S6.** TEM images of (a)  $4.9\pm0.3$  nm TOP-Pd nanoparticles, (b)  $3.6\pm0.4$  nm Fe<sub>3</sub>O<sub>4</sub> nanoparticles, and the mixture of two kinds of nanoparticles (c) before and (d) after annealing under Ar + 4% H<sub>2</sub> at 500°C for 1 h.