

## Supporting Information

### High Density Ordered Arrays of CoPt<sub>3</sub> Nanoparticles with Individually Addressable Out-of-Plane Magnetization

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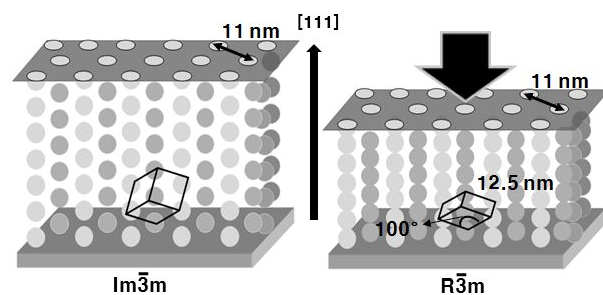
## **1. MTF templates for fabricating 2D hexagonal array structures of replicas**

Mesoporous materials are synthesized by the self-assembly process between an inorganic precursor and a surfactant. Because of the different chemical properties and the attractive interaction between the two components, microscopic phase segregation occurs, through which many different types of ordered structures can be formed depending on many parameters including the nature of the surfactant and the inorganic precursor and their ratio. After the self-assembly is completed, the surfactant is removed to result in a porous inorganic material with regularly structured pores. Although the range of pore size of mesoporous materials is defined to be 2-50 nm by IUPAC, the typical size is in the range of 2-10 nm and pores larger than 10 nm is hard to achieve especially with commercial surfactants.

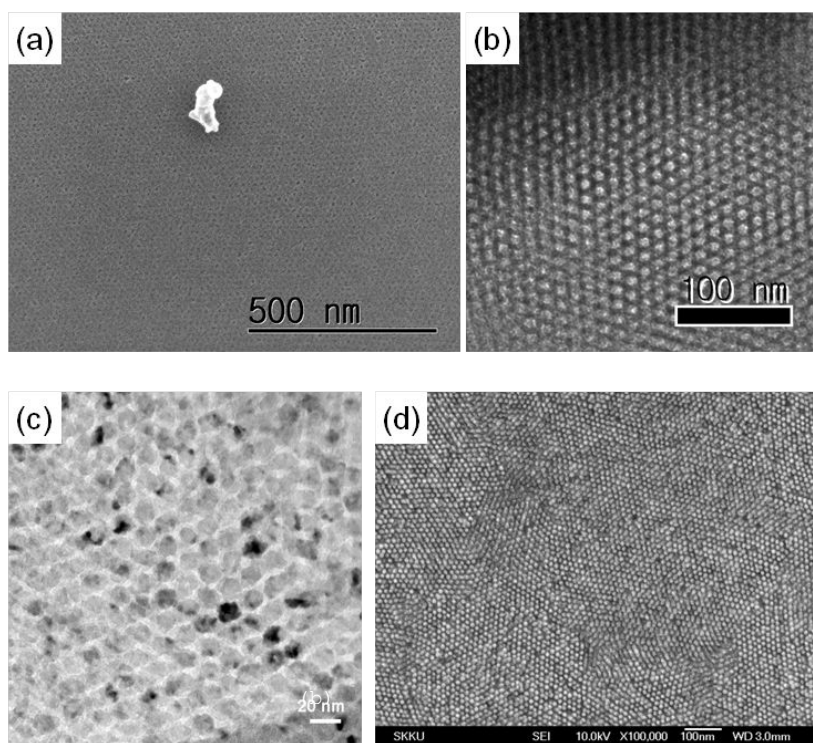
The synthesis of MTF makes use of the same principle of self-assembly between an inorganic precursor and a surfactant, although additional considerations are required. In a typical procedure to synthesize a MTF, a solution of the two components in a volatile solvent is coated on a substrate. During the solvent evaporation, the two components become concentrated to reach the critical micelle concentration, at which point their self-assembly takes place to result in an ordered structure between the two components. Calcination of the ordered structure yields a MTF.

The most common structure of mesoporous material is called the hexagonal structure which is composed of parallel long pore channels. MTFs of such a hexagonal structure have a strong tendency to align the pore channels parallel to the substrate plane because such an orientation is the most stable thermodynamically. Therefore, MTFs with the hexagonal pore structures cannot be utilized in BPM.

On the other hand, it has been shown that MTFs with the cage-like ( $Im\bar{3}m$ ) pore structure can be processed into usable templates. When a cage-like MTF is formed in such a way to share its (111) face with the substrate plane, the adjacent pores become connected to form perpendicular pore channels during the aging of the green material (Fig. S1). Some examples of cage-like MTFs are shown in Fig. S2(a) and S2(b) and replicas in Fig. S2(c) and S2(d).



**Figure S1.** Schematic drawing of the structures of a cage-like pore MTF oriented to share its (111) plane parallel to the substrate. As-prepared film (left) and after calcination (right).<sup>S1</sup>

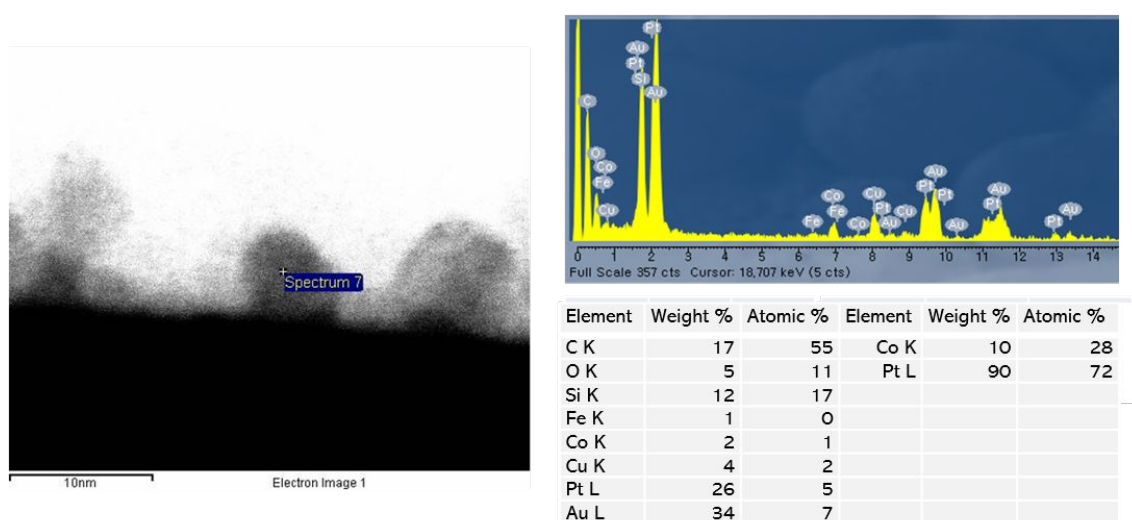


**Figure S2.** Electron microscopic images of MTFs with cage-like pores and their replicas: (a) Top view SEM image of a MTF made of  $\text{TiO}_2$ ,<sup>S1</sup> (b) top view TEM image of a MTF made of  $\text{SiO}_2$ ,<sup>S2</sup> (c) top view TEM image of CdSe replica<sup>S3</sup> and (d) top view SEM image of Pt replica<sup>S4</sup>. The replicas in (c) and (d) were formed by using MSTF templates as shown in (b).

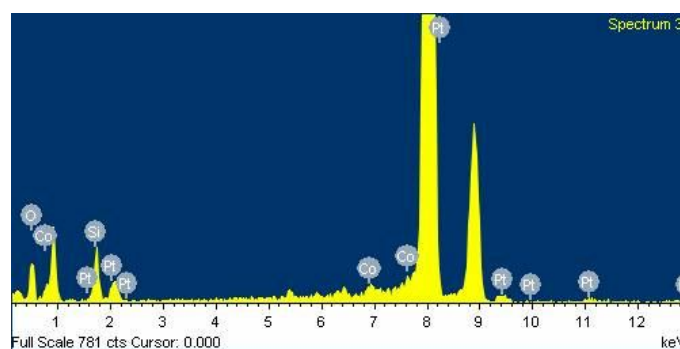
## 2. Analysis data on CoPt<sub>3</sub> NP arrays

**Table S1.** Composition of Co-Pt NPs formed by using MSTFs with different thicknesses

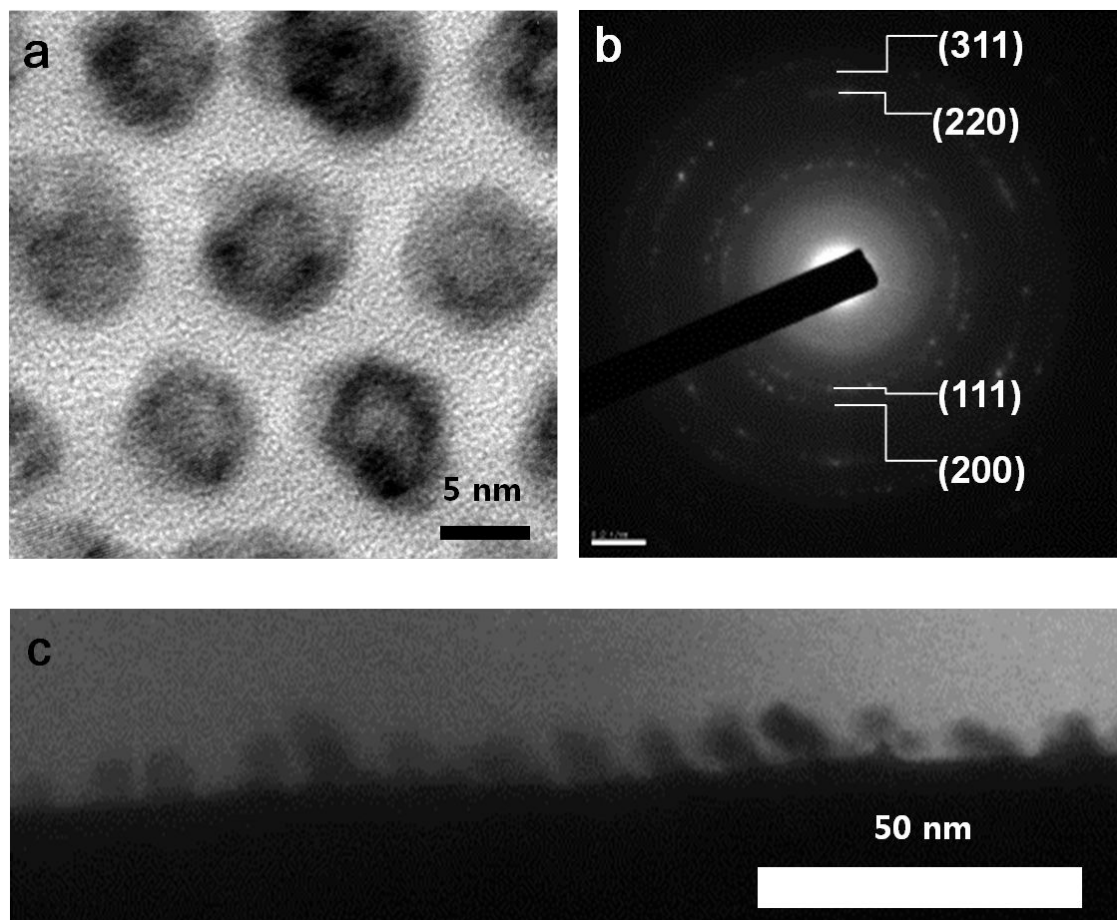
Thickness of MSTF	soaking time (min)	deposition time (s)	Co/Pt atomic ratio	EDS spectrum
250 nm	30	0.1	1/3	Figure S3
90 nm	30	0.1	1/1	Figure S4



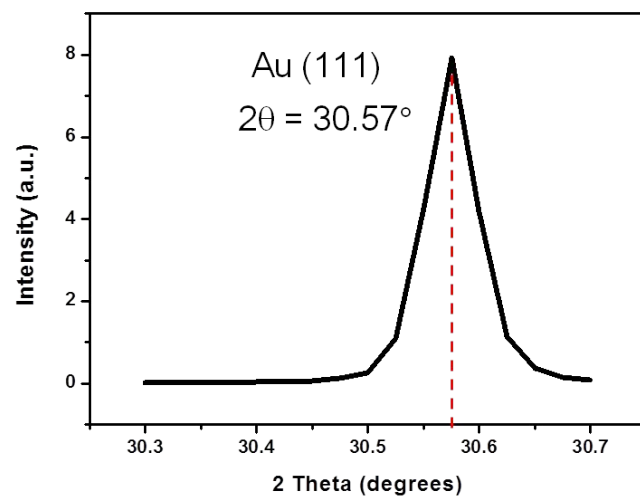
**Figure S3.** TEM image of a Co-Pt NP synthesized by using a 250 nm MSTF template and its EDS spectrum.



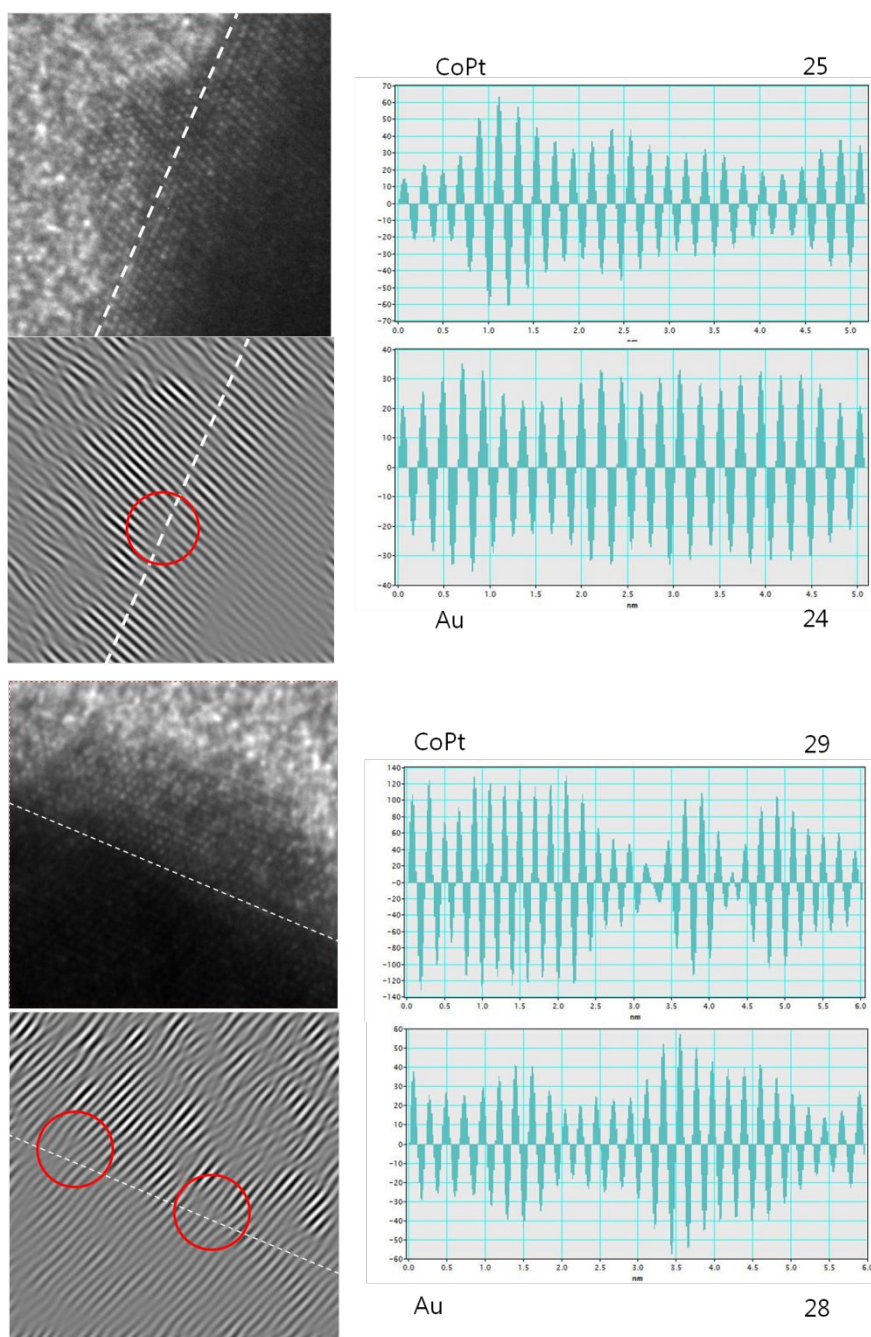
**Figure S4.** EDS spectrum of Co-Pt NPs formed by using a 100 nm thick MSTF template.



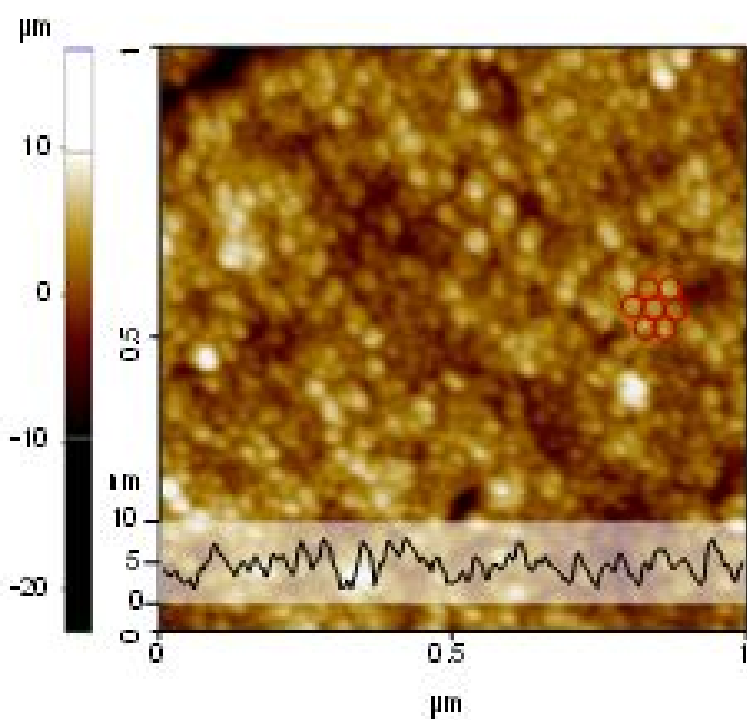
**Figure S5.** Electron microscopy images on CoPt<sub>3</sub> NPs formed on Au (111)/SiO<sub>2</sub> substrate: (a) Top view HVTEM image, (b) selected area electron diffraction pattern on CoPt<sub>3</sub> NPs collected by scraping off the sample, (c) side view TEM image.



**Figure S6.** GIXS spectrum of CoPt<sub>3</sub>/Au (111).  $\lambda = 0.123984$  nm.



**Figure S7.** Cross-section HR-TEM images of CoPt<sub>3</sub> NPs grown on Au (111), corresponding 1D FFT images, and plots of spacings of CoPt<sub>3</sub> NP and Au parts. The frequency of the line dislocation in CoPt<sub>3</sub> NPs is one in every 25-30 Au atomic layers.



**Figure S8.** AFM image of CoPt<sub>3</sub> NPs on Au (111)/SiO<sub>2</sub> substrate and height variation across the image.



## References

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- S2. Lee, U.-H.; Yang, J.-H.; Lee, H.-J.; Park, J.-Y.; Lee, K.-R.; Kwon, Y.-U., “Facile and adaptable synthesis method of mesostructured silica thin films”, *J. Mater. Chem.*, **2008**, 18, 1881–1888.
- S3. Kim, Y.-T.; Han, J. H., Hong, B. H.; Kwon, Y.-U., “Electrochemical Synthesis of CdSe Quantum-Dot Arrays on a Graphene Basal Plane Using Mesoporous Silica Thin-Film Templates”, *Adv. Mater.*, **2010**, 22, 515-518.
- S4. Lee, H.-J.; Lee, U.-H.; Park, J.-Y.; Yoo, S.-H.; Park, S.; Kwon, Y.-U., “Platinum Films with Controlled 3-Dimensional Nanoscopic Morphologies and Their Effects on Surface Enhanced Raman Scattering”, *Chem.-Asian J.* **2009**, 4, 1284-1288.