

## Supporting Information

### One-pot self-assembly of multifunctional mesoporous nanoprobe with magnetic nanoparticles and hydrophobic upconversion nanocrystals

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#### Experimental

**Preparation of monodisperse magnetic Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub> nanoparticles:** The spherical magnetic Fe<sub>3</sub>O<sub>4</sub> particles were prepared by hydrothermal synthesis reported previously.<sup>1</sup> The core-shell structured Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub> nanoparticles with narrow size distribution were prepared by a slightly modified Stöber process. In a typical procedure, as-prepared Fe<sub>3</sub>O<sub>4</sub> (0.05 g) nanoparticles were dispersed in a mixture of ethanol (80 mL), distilled water (20 mL) and further ultrasonic treatment for 10 min. Subsequently, concentrated ammonia aqueous solution (28 wt %, 1.0 mL) and TEOS (0.15 g) were then added to the solution under mechanically stirring and the reaction was allowed to proceed for 12 h. Finally, the products were separated using a magnet and washed with ethanol and distilled water.

**Preparation of Nanocrystals NaYF<sub>4</sub>:Yb<sup>3+</sup>/Tm<sup>3+</sup>:** The synthesis of NaYF<sub>4</sub>:2% Tm<sup>3+</sup>/20% Yb<sup>3+</sup> was done following a reported procedure.<sup>2</sup>

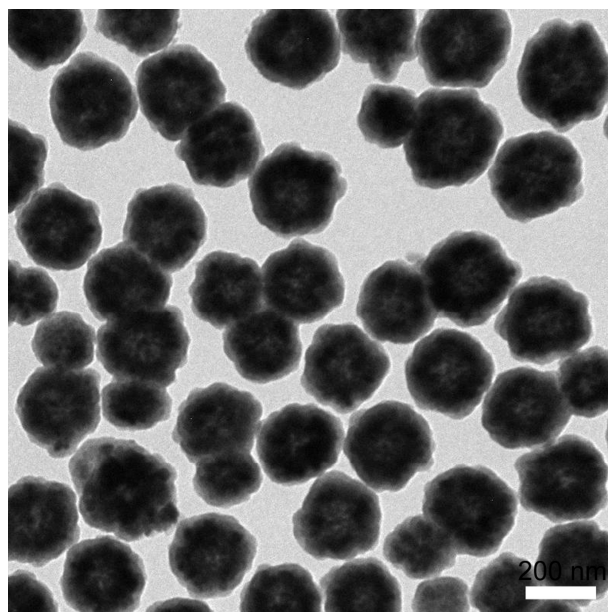
**Preparation of Magnetic, Luminescent Mesoporous Silica Nanospheres (MFNPs):** In a typical synthesis, the oleate-capped UCNPs (5.0 mg) dispersed in 1.0 mL of chloroform was mixed with 0.1 g of cetyltrimethylammonium bromide (CTAB) and 10 mL of water. The mixture was then sonicated and stirred vigorously, a homogeneous oil-in-water microemulsion was obtained. After vigorous stirring of the resulting solution at 60 °C for 15 min, the chloroform solvent was boiled off from the solution. Then a mixture of 60 mg of Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub> and 40 mL of distilled water

prehomogenized by sonication was added. The UCNPs and CTAB would self-assemble around magnetic silica spheres. Then ethanol (30 mL) and concentrated ammonia solution (0.6 mL, 28 wt %) were successively added to the solution under stirring for 30 min at 30 °C. Afterward, 65  $\mu$ L of tetraethylorthosilicate (TEOS) was added dropwise under continuous stirring. After stirring for 6 h at 30 °C, the product was collected with a magnet and washed with ethanol and distilled water. The CTAB surfactants were removed from the mesopores by dispersing the as-synthesized materials in 50 mL of acetone and refluxing at 80 °C for 48h. The extraction was repeated 3 times to ensure a complete removal of CTAB templates. Finally, the materials were then centrifuged and washed with ethanol.

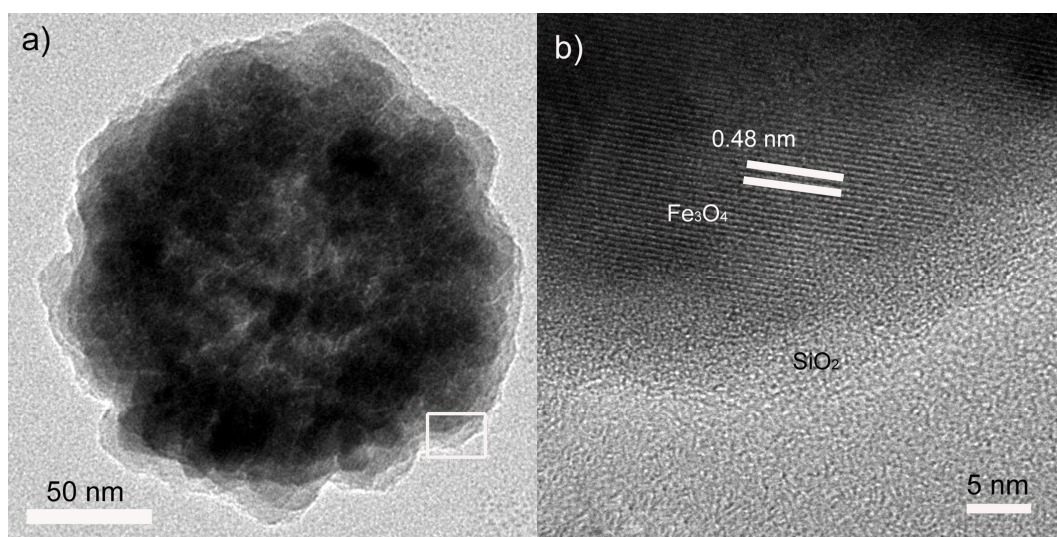
**Characterization:** Low-resolution TEM images were obtained on a JEM 200CX microscope, using an accelerating voltage of 120 kV. High-resolution TEM images were taken on a JEOL JEM-2010F microscope operated at 200 kV. Scanning electron microscopy (SEM) images were obtained on a field emission JEOL JSM-6700F microscope. The zeta potential and particle size distribution were measured using the Malvern Zetasizer 3000HS. X-ray powder diffraction patterns were recorded using a D/MAX-2550 diffractometer, equipped with a rotating anode and with a Cu K $\alpha$  radiation source ( $\lambda$ = 0.154178 nm). The N<sub>2</sub> adsorption–desorption isotherm was measured using a Micromeritics ASAP 2020 M apparatus. The specific surface area was calculated by the Brunauer–Emmett–Teller (BET) method. The pore size distribution was obtained from the Barret–Joner–Halenda (BJH) method. Magnetization measurements were conducted using VSM at room temperature. Upconversion luminescence spectra were measured with an Edinburgh LFS-920 fluorescence spectrometer, equipped with an external 0~800 mW adjustable continuous wave laser (980 nm, Beijing Hi-Tech Optoelectronic Co., China) as the excitation source.

## References

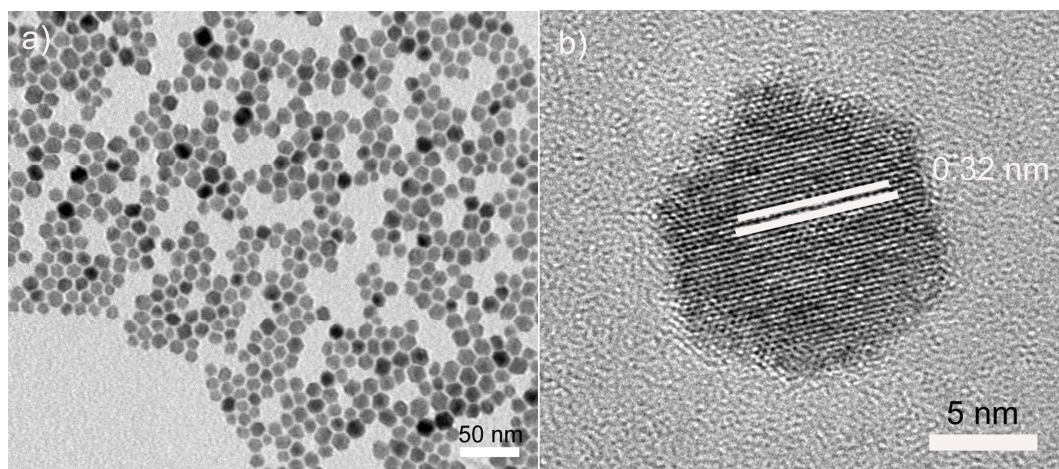
- (1) W. Cheng, K. Tang, Y. Qi, J. Sheng and Z. Liu, *J. Mater. Chem.*, 2010, **20**, 1799-1805.
- (2) G. Chen, T. Y. Ohulchansky, R. Kumar, H. Ågren and P. N. Prasad, *ACS Nano*, 2010, **4**, 3163-3168.



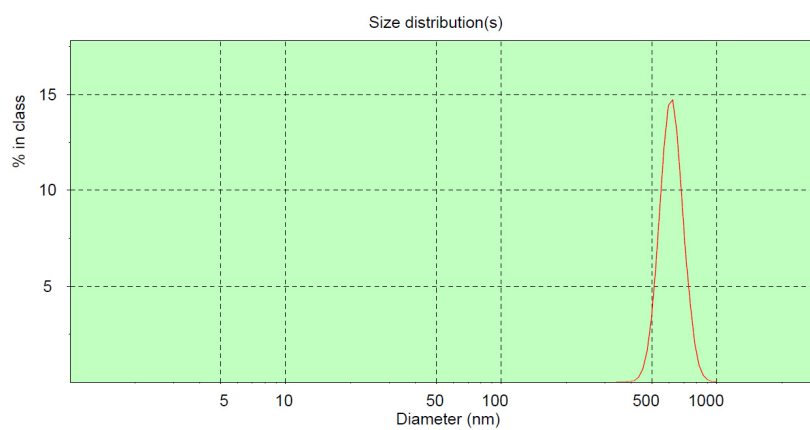
**Figure S1.** TEM image of Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>.



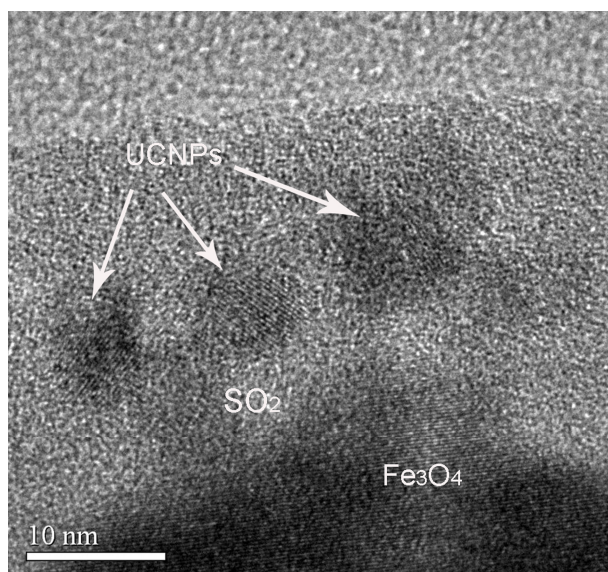
**Figure S2.** (a) HR-TEM image of Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub> and (b) a magnified image of the square-marked area in (a), the fringe spacing 0.48 nm is consistent with the interplanar distance of (220) lattice planes.



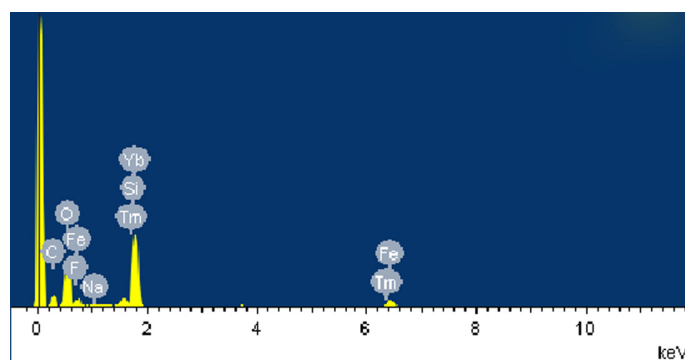
**Figure S3.** (a) TEM image of UCNPs ( $\text{NaYF}_4:2\% \text{Tm}^{3+}/20\% \text{Yb}^{3+}$ ), (b) HR-TEM image of a single upconversion nanoparticle, showing (111) lattice fringes.



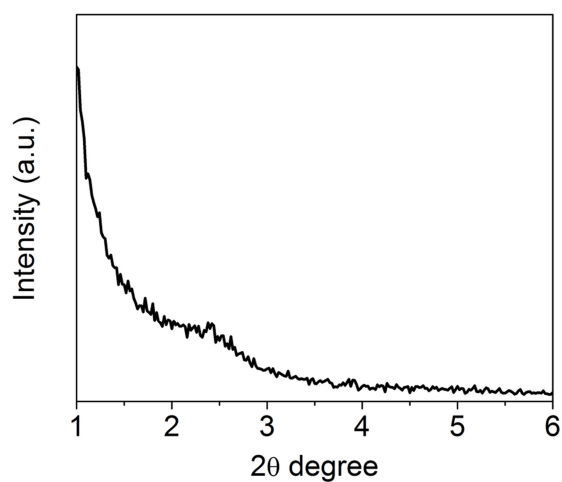
**Figure S4.** Particle size distribution of MFNPs measured by dynamic light scattering (DLS) method in aqueous solution.



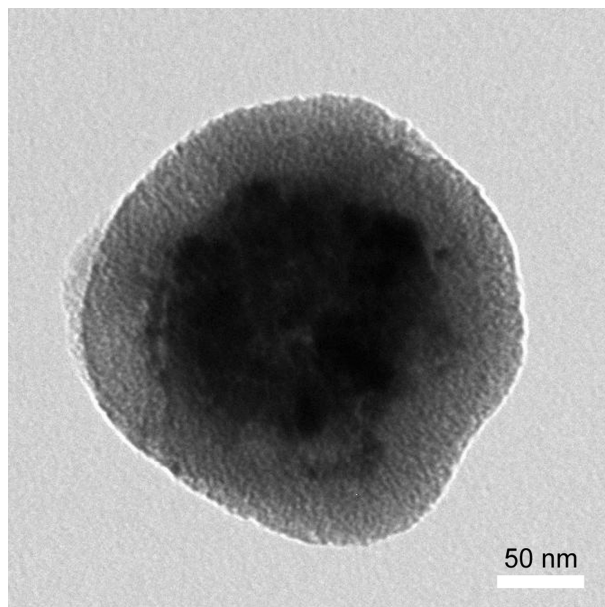
**Figure S5.** HR-TEM image of MFNPs, in which the UCNP were assembled around the  $\text{Fe}_3\text{O}_4@ \text{SiO}_2$ .



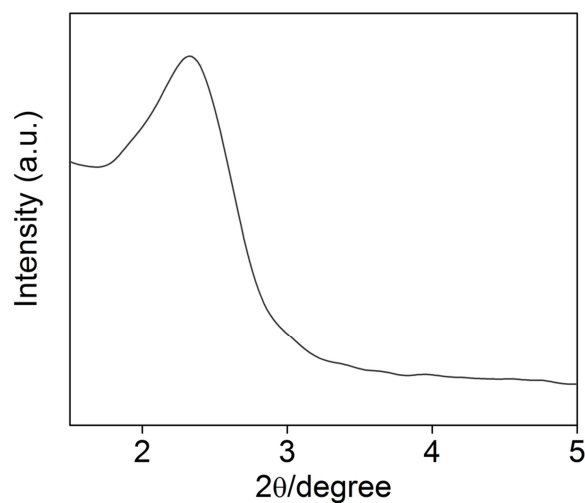
**Figure S6.** EDX analysis of MFNPs.



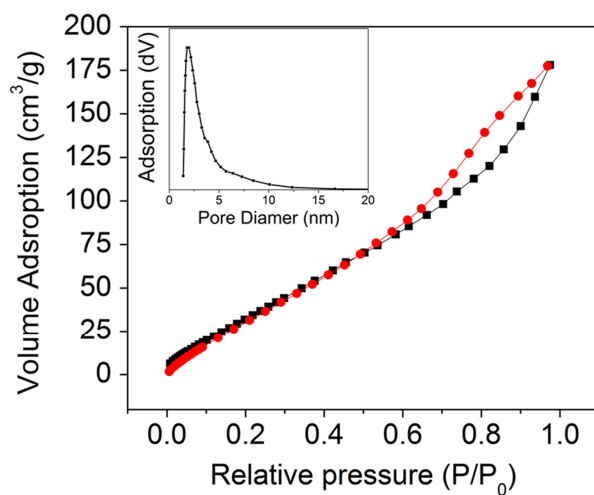
**Figure S7.** Low-angle XRD pattern of the MFNPs with a mesoporous shell.



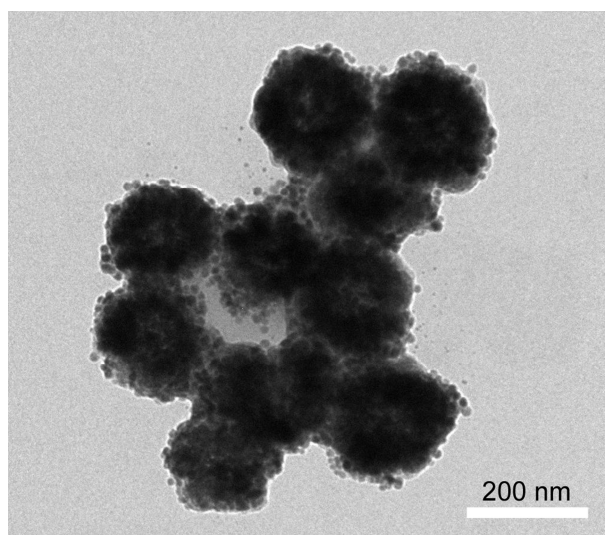
**Figure S8.** HR-TEM image of the obtained mesoporous nanospheres with twice the amount of TEOS used. Although some twisted disordered regions were observed clearly, the mesopores at the edge of the shell were developed generally in a uniform and orderly fashion.



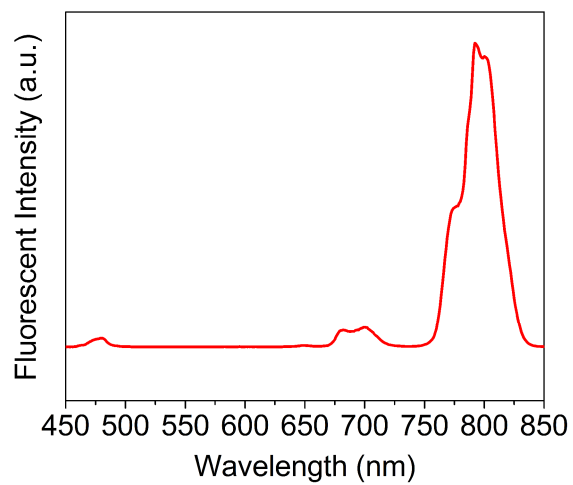
**Figure S9.** Low-angle XRD pattern of the mesoporous nanospheres with twice the amount of TEOS used.



**Figure S10.** N<sub>2</sub> adsorption–desorption isotherms and BJH pore diameter distribution curve at adsorption branch (inset) of the mesoporous nanospheres with the twice the amount of TEOS used.



**Figure S11.** TEM image captured before TEOS was added (CTAB coated UCNPs assembled around Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>).



**Figure S12.** Emission spectrum of colloidal solution of UCNPs (NaYF<sub>4</sub>:2% Tm<sup>3+</sup>/20% Yb<sup>3+</sup>) in cyclohexane excited at 980 nm.