

## Supporting Information

### **Rational Design of Nd<sup>3+</sup>-Sensitized Multifunctional Nanoparticles with Highly Dominant Red Emission**

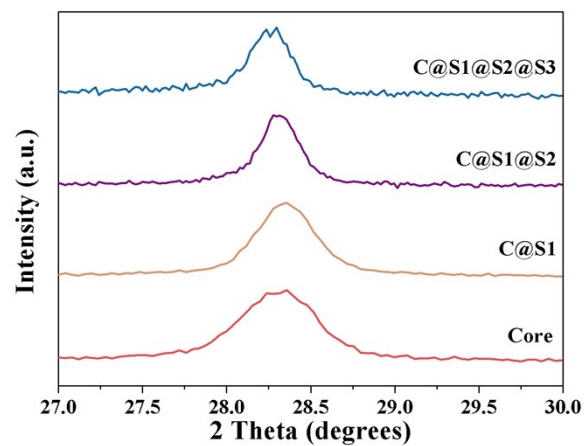
Xia Xu,<sup>a,b</sup> Pengpeng Lei,<sup>a,b</sup> Lile Dong,<sup>a,b</sup> Xiuling Liu,<sup>a</sup> Yue Su,<sup>a,b</sup> Shuyan Song,<sup>a</sup> Jing  
Feng<sup>\*a</sup> and Hongjie Zhang<sup>\*a</sup>

<sup>a</sup> *State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied  
Chemistry, Chinese Academy of Sciences, 5625 Renmin Street, Changchun 130022, China.*

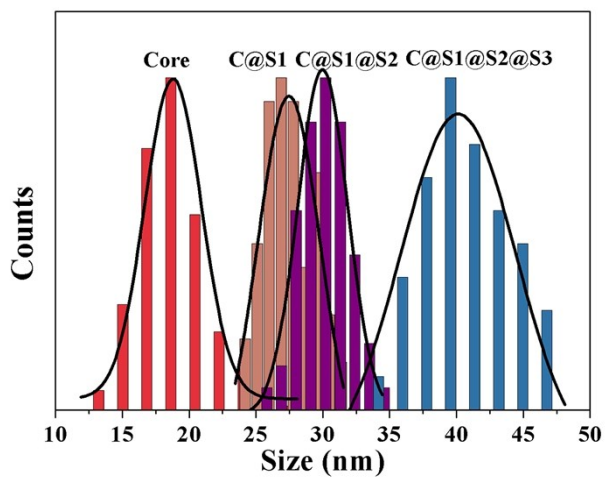
<sup>b</sup> *University of Chinese Academy of Sciences, Beijing 100049, China*

\* Corresponding authors. Tel.: +86 431 85262127; fax: +86 431 85698041.

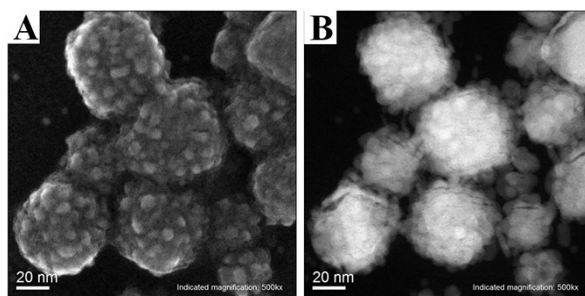
*E-mail addresses:* fengj@ciac.ac.cn (J. Feng), hongjie@ciac.ac.cn (H. Zhang).



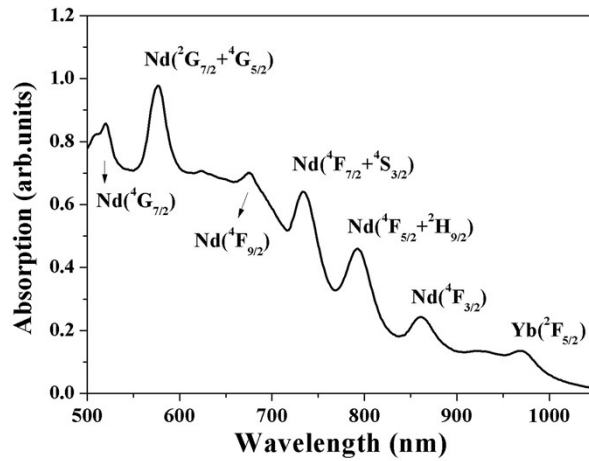
**Fig. S1** The magnified XRD patterns of Core, C@S1, C@S1@S2 and C@S1@S2@S3 nanoparticles.



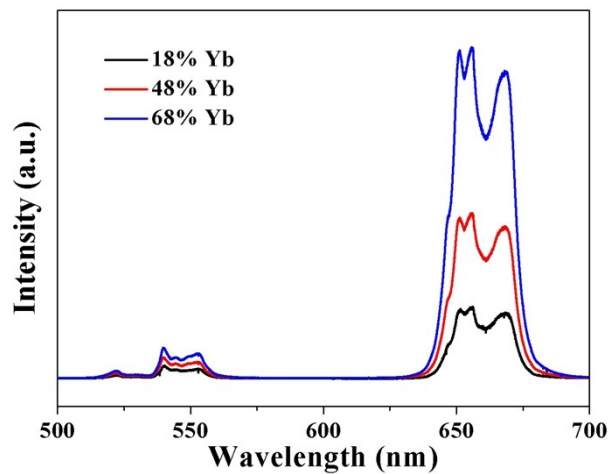
**Fig. S2** Size distribution of Core, C@S1, C@S1@S2 and C@S1@S2@S3 nanoparticles.



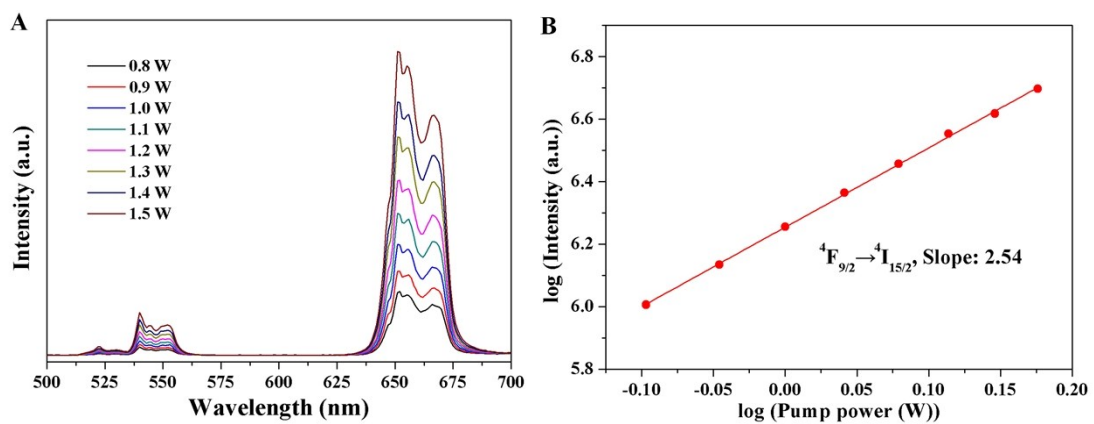
**Fig. S3** A) High-resolution SEM image and B) HAADF-STEM image of C@S1@S2@S3 UCNPs.



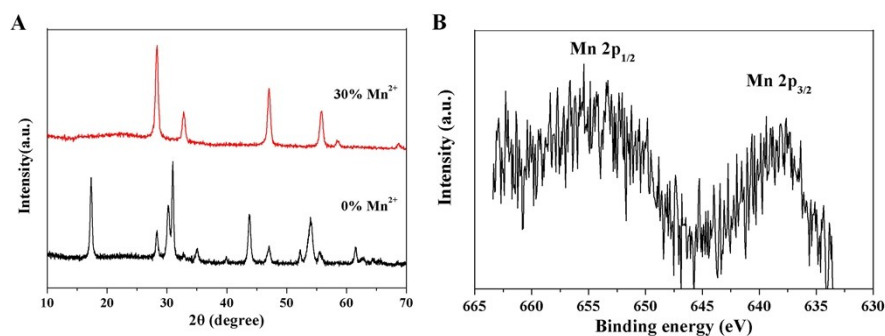
**Fig. S4** The absorption spectrum of C@S1@S2 UCNPs.



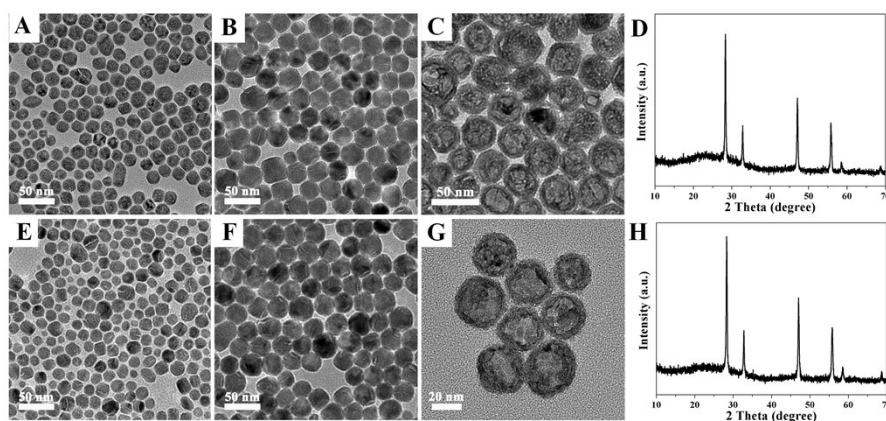
**Fig. S5** The Yb<sup>3+</sup> concentration-dependent UCL spectra of C@S1@S2 UCNPs under 980 nm excitation.



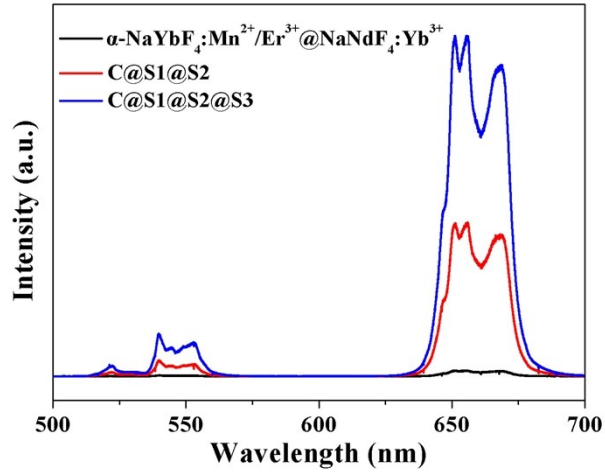
**Fig. S6** A) UCL spectrum of C@S1@S2@S3 UCNPs excited with a series of 808 nm pump power, B) log-log plot of the UCL intensities of C@S1@S2@S3 UCNPs peaked at 653 nm as a function of 808 nm pump power.



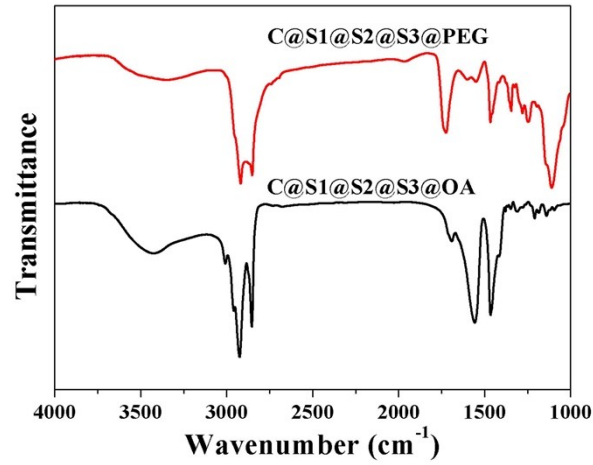
**Fig. S7** A) XRD pattern of NaYbF<sub>4</sub>:18%Yb<sup>3+</sup>, 2%Er<sup>3+</sup> without and with 30% Mn<sup>2+</sup> doped. B) The expanded spectrum of Mn 2p obtained from the C@S1@S2 UCNPs. The Mn 2p<sub>1/2</sub> and 2p<sub>3/2</sub> signals are located at 654.3 and 638.8 eV, respectively.



**Fig. S8** TEM image of A) α-NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>(18%)/Er<sup>3+</sup>, B) α-NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>(18%)/Er<sup>3+</sup>@NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>, C) α-NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>(18%)/Er<sup>3+</sup>@NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>@NaNdF<sub>4</sub>:Yb<sup>3+</sup> and D) XRD pattern of (C). TEM image of E) α-NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>(48%)/Er<sup>3+</sup>, F) α-NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>(48%)/Er<sup>3+</sup>@NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>, G) α-NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>(48%)/Er<sup>3+</sup>@NaLuF<sub>4</sub>:Mn<sup>2+</sup>/Yb<sup>3+</sup>@NaNdF<sub>4</sub>:Yb<sup>3+</sup> and H) the XRD pattern of (G).



**Fig. S9** The UCL spectra of  $\alpha\text{-NaYbF}_4\text{:Mn}^{2+}/\text{Er}^{3+}\text{@NaNdF}_4\text{:Yb}^{3+}$ ,  $\text{C@S1@S2}$  and  $\text{C@S1@S2@S3}$  under 980 nm excitation.



**Fig. S10** FTIR spectra of  $\text{C@S1@S2@S3@OA}$  and  $\text{C@S1@S2@S3@PEG}$ .