

### *Supplementary Information for*

Tuning upconversion through a sensitizer/activator-isolated NaYF<sub>4</sub> core/shell structure

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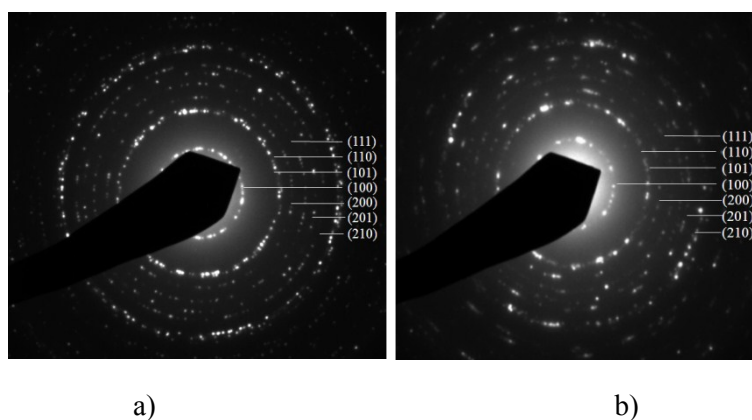
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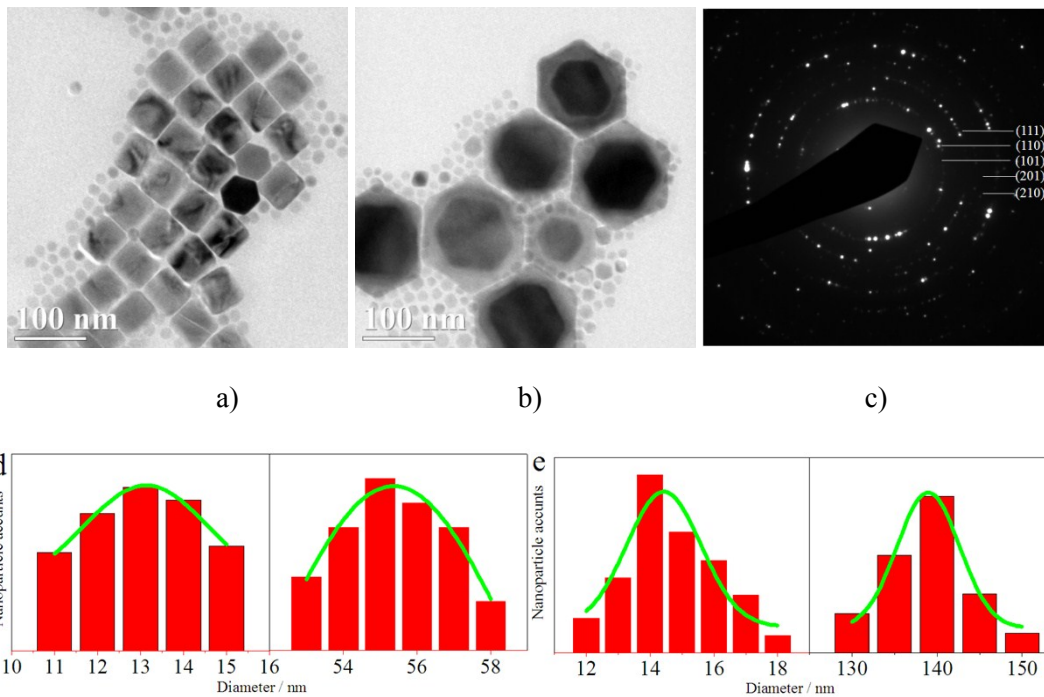
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**Figure S1.** The selected area electron diffraction patterns of (a) the conventional NaYF<sub>4</sub>:Yb<sup>3+</sup>50%, Ho<sup>3+</sup> 1% and (b) the core-shell NaYF<sub>4</sub>:Yb<sup>3+</sup>50%@NaYF<sub>4</sub>:Ho<sup>3+</sup>1% upconversion nanoparticles. The diffraction rings correspond to that of the standard hexagonal NaYF<sub>4</sub> host lattice of JCPDS 28-1192.



**Figure S2.** Transmission electron microscopic images of a) the core-shell  $\text{NaYF}_4:\text{Yb}^{3+}60\%@\text{NaYF}_4:\text{Ho}^{3+}1\%$  UCNPs, b) the core-shell  $\text{NaYF}_4:\text{Yb}^{3+}80\%@\text{NaYF}_4:\text{Ho}^{3+}1\%$  UCNPs. There are clearly two size range nanoparticles appeared in both (a) and (b). The smaller size nanoparticles in (a) might arise from the incompleteness of the Ostwald-ripening process, while the smaller ones in (b) is possibly due to the self-nucleation of the shell host materials. (c) The selected area electron diffraction pattern of the core-shell  $\text{NaYF}_4:\text{Yb}^{3+}60\%@\text{NaYF}_4:\text{Ho}^{3+}1\%$  UCNPs, corresponding to the standard hexagonal  $\text{NaYF}_4$  host lattice of JCPDS 28-1192. The (d) and (e) display the two size distribution of the core-shell  $\text{NaYF}_4:\text{Yb}^{3+}60\%@\text{NaYF}_4:\text{Ho}^{3+}1\%$  UCNPs in (a), and the core-shell  $\text{NaYF}_4:\text{Yb}^{3+}80\%@\text{NaYF}_4:\text{Ho}^{3+}1\%$  UCNPs in (b), respectively.

We calculated the intended element concentrations of each type of rare earth ions in the core @ shell samples of NaYF<sub>4</sub>:Yb50% @ NaYF<sub>4</sub>:Ho2%, NaYF<sub>4</sub>:Yb50% @ NaYF<sub>4</sub>:Er2%, NaYF<sub>4</sub>:Yb50% @ NaYF<sub>4</sub>:Tm 1%, and compared them directly with the measured element concentration using inductively coupled plasma mass spectrometry (ICP-MS). The comparison results are shown in Table S1-S3. To calculate, we used the following equation

$$Con(RE) = \frac{Con(RE)_{core} + Con(RE)_{shell}}{2}$$

Where Con(RE) stands for the concentration of RE (Y, Yb, Ho, Er, Tm) elements, Con(RE)<sub>core</sub> stands for the concentration of RE in the core, Con(RE)<sub>shell</sub> stands for the concentration of RE in the shell. The volume of the core as well as the volume of the shell was both taken into consideration when calculating the intended element concentration.

Table S1. Element concentration of NaYF<sub>4</sub>:50%Yb@NaYF<sub>4</sub>:1%Ho by ICP-MS

| Elements | Theoretical Concentration (%) | Real Concentration (%) |
|----------|-------------------------------|------------------------|
| Y        | 74.5                          | 62.90                  |
| Yb       | 25                            | 36.69                  |
| Ho       | 0.5                           | 0.41                   |

Table S2. Element concentration of NaYF<sub>4</sub>:50%Yb@NaYF<sub>4</sub>:2%Er by ICP-MS

| Elements | Theoretical Concentration (%) | Real Concentration (%) |
|----------|-------------------------------|------------------------|
| Y        | 74                            | 63.25                  |
| Yb       | 25                            | 35.79                  |
| Er       | 1                             | 0.78                   |

Table S3. Element concentration of NaYF<sub>4</sub>:50%Yb@NaYF<sub>4</sub>:1%Tm by ICP-MS

| Elements | Theoretical Concentration (%) | Real Concentration (%) |
|----------|-------------------------------|------------------------|
| Y        | 74.5                          | 63.57                  |
| Yb       | 25                            | 36.04                  |
| Tm       | 0.5                           | 0.39                   |

