## **Supporting Information**

## Core-shell nanoarchitecture: a strategy to significantly enhance white-light upconversion of lanthanide-doped nanoparticles

Bo Zhou,<sup>ab‡</sup> Lili Tao,<sup>a‡</sup> Yuen H. Tsang,<sup>\*a</sup> and Wei Jin<sup>c</sup>

<sup>a</sup> Department of Applied Physics and Materials Research Centre, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong

<sup>b</sup> Institute of Materials Research and Engineering, Agency for Science, Technology

and Research (A\*STAR), 3 Research Link, Singapore 117602

<sup>c</sup> Department of Electrical Engineering, The Hong Kong Polytechnic University, Hung

Hom, Kowloon, Hong Kong

<sup>‡</sup> These authors contributed equally to this work.

\* E-mail address: yuen.tsang@polyu.edu.hk



**Figure S1.** The UC emission spectrum of core CaF<sub>2</sub>:Yb/Tm (20/0.5 mol%) in 300-520 nm wavelength region upon a 980-nm LD excitation. The emission band at ~360 nm originates from the Tm<sup>3+</sup>:  ${}^{1}D_{2} \rightarrow {}^{3}H_{6}$  transition. The energy in Tm<sup>3+</sup>  ${}^{1}D_{2}$  also leads to a blue emission at ~450 nm when it decays to the first excited state Tm<sup>3+</sup>  ${}^{3}F_{4}$ .



**Figure S2.** Power dependence of UC emission intensities in CaF<sub>2</sub>:Yb/Ho (20/0.5 mol%) and CaF<sub>2</sub>:Yb/Tm (20/0.5 mol%) under 980-nm excitation. The slope values are fitted to be 1.63, 1.71 and 2.57 for Ho<sup>3+</sup>:  ${}^{5}F_{5} \rightarrow {}^{5}I_{8}$ , Ho<sup>3+</sup>: ( ${}^{5}F_{4}, {}^{5}S_{2}$ ) $\rightarrow {}^{5}I_{8}$  and Tm<sup>3+</sup>:  ${}^{1}G_{4} \rightarrow {}^{3}H_{6}$  transitions, respectively, confirming the red and green emissions of Ho<sup>3+</sup> are two-step UC processes and blue of Tm<sup>3+</sup> is a three-step UC process.



Figure S3. Comparison of UC emission spectra from core-shell samples of (a)  $CaF_2:Yb/Tm/Ho$  (20/0.5/2.0 mol%)@NaYF4:Yb (20 mol%), (b)  $CaF_2:Yb/Tm/Ho$  (20/0.5/2.0 mol%)@CaF2:Yb (20 mol%), (c)  $NaYF_4:Yb/Tm/Ho$  (20/0.5/2.0 mol%)@CaF2:Yb (20 mol%) and core only samples of (d)  $CaF_2:Yb/Tm/Ho$  (20/0.5/2.0 mol%) and (e)  $NaYF_4:Yb/Tm/Ho$  (20/0.5/2.0 mol%) recorded under 980-nm excitation.



Figure S4. UC emission spectra from (a) core  $CaF_2$ :Yb/Tm/Ho (40/0.5/2.0 mol%) and (b) core-shell  $CaF_2$ :Yb/Tm/Ho (40/0.5/2.0 mol%)@NaYF<sub>4</sub>:Yb (20 mol%) recorded under 980-nm excitation. The UC emission from (c)  $CaF_2$ :Yb/Tm/Ho (20/0.5/2.0 mol%) is also plotted for comparison.