

# Dye Sensitization for Ultraviolet Upconversion Enhancement

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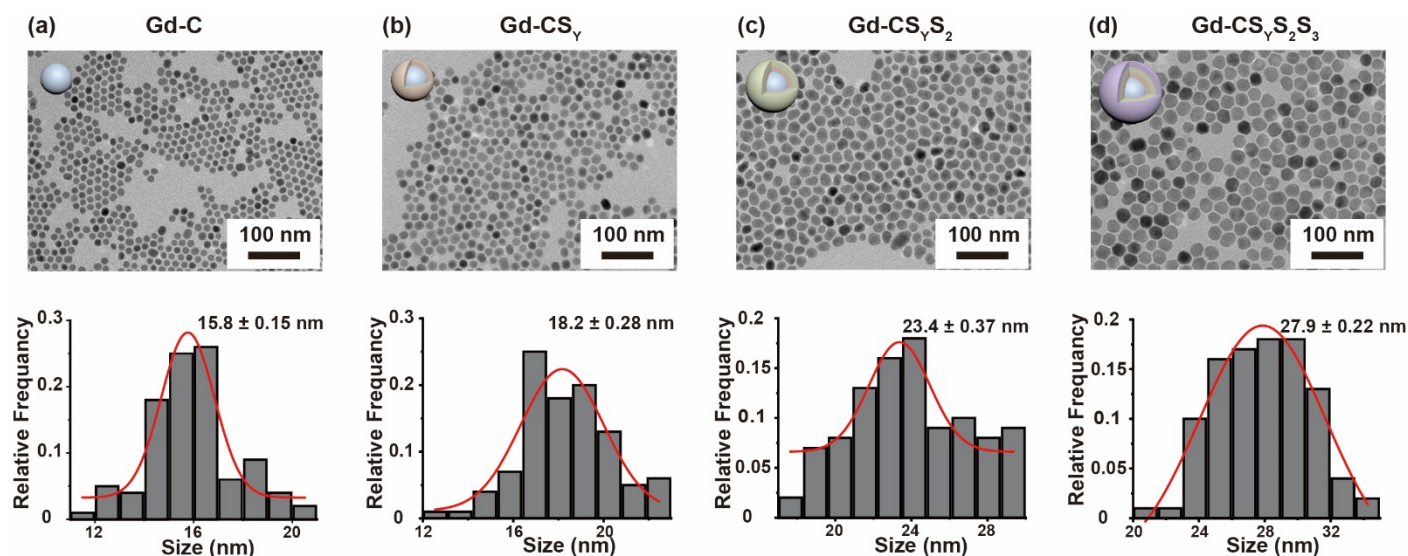
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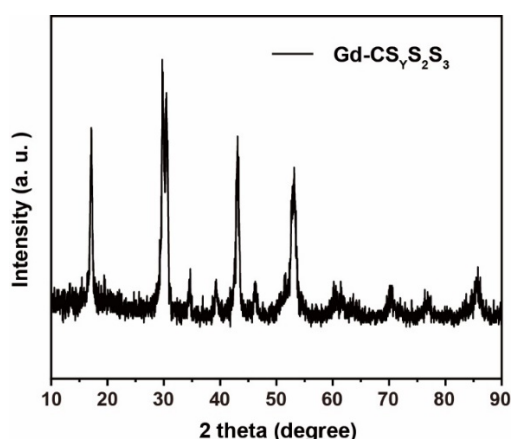
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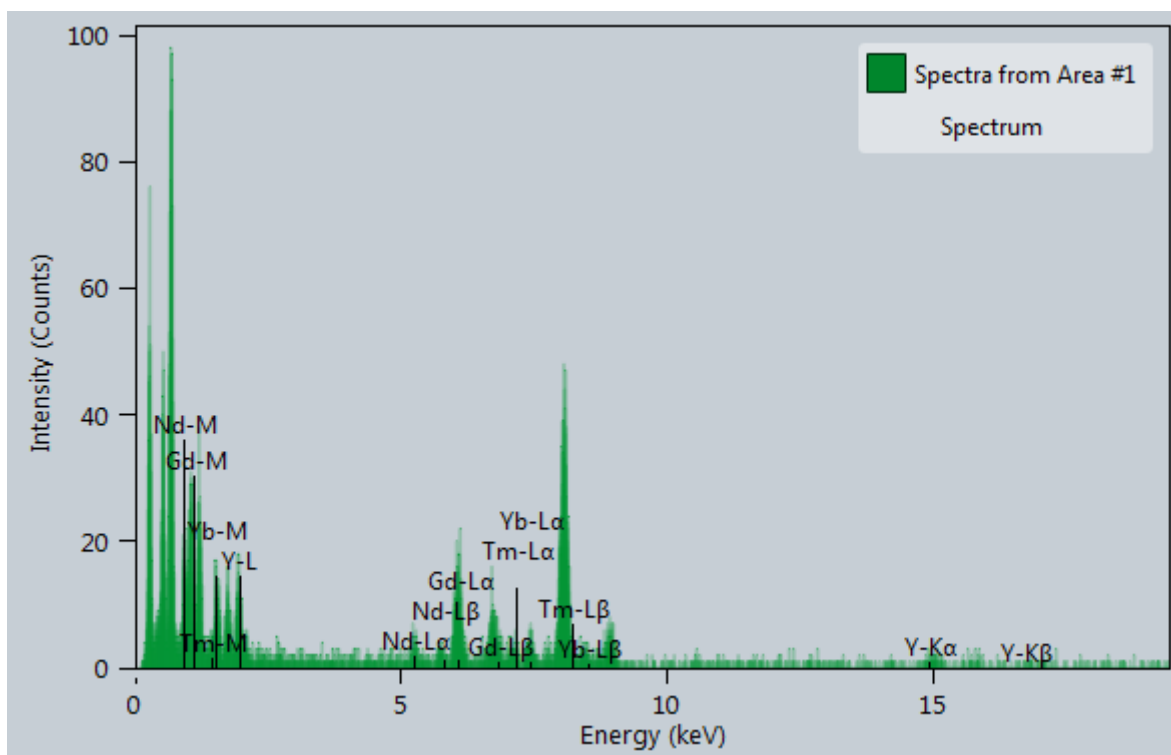
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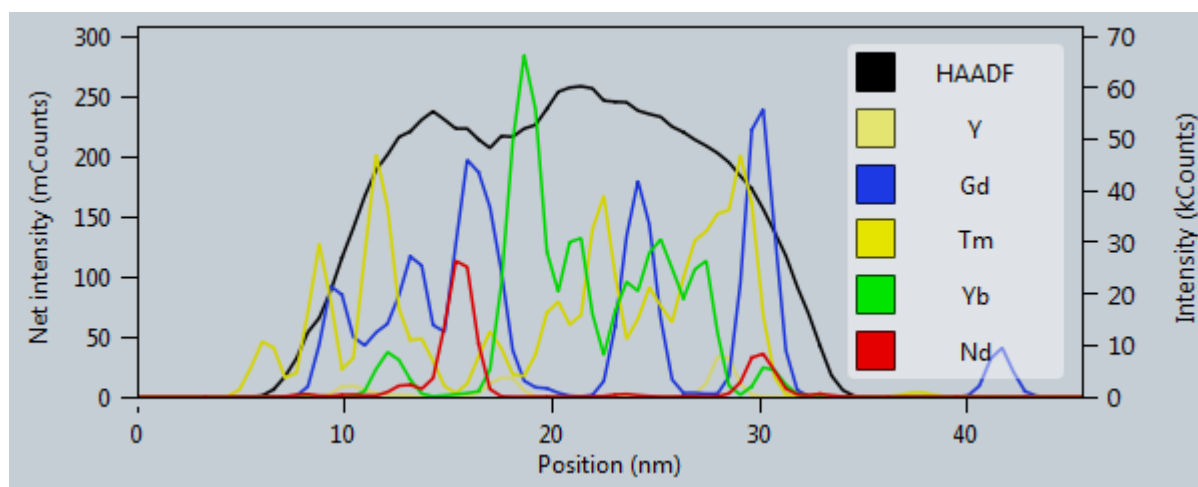
**Figure S1.** Transmission electron microscopy (TEM) and nanoparticles size distribution of as-prepared NaGdF<sub>4</sub>:49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd@NaGdF<sub>4</sub> (Gd-CS<sub>Y</sub>S<sub>2</sub>S<sub>3</sub>). (a) core, (b) core-shell, (c) core-shell-shell, (d) core-multishell. The size distribution of nanoparticles is matched by a Gaussian curve.



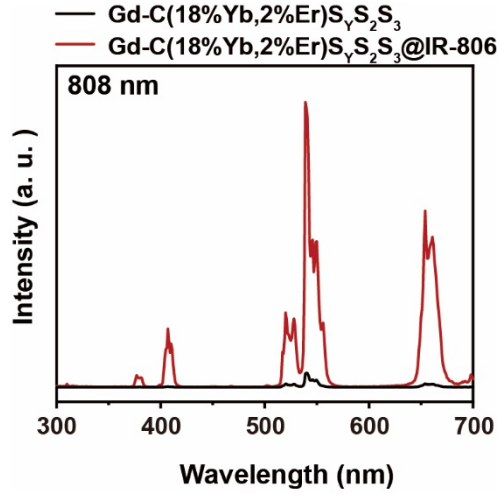
**Figure S2.** X-ray powder diffraction (XRD) curve of the as-synthesized heterogeneous core-multishell nanoparticles.



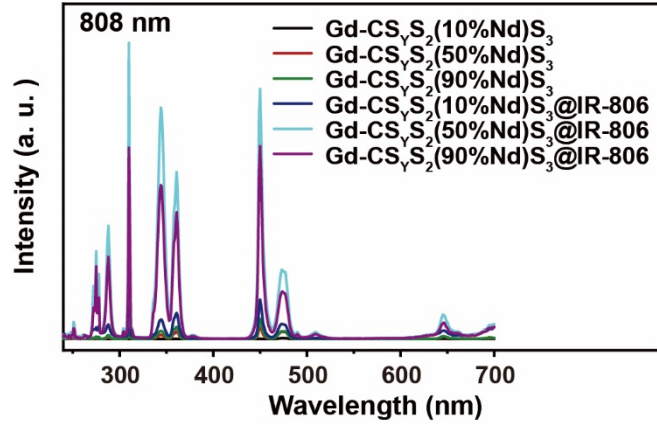
**Figure S3.** Energy dispersive X-ray (EDX) spectrum of Gd-CS<sub>y</sub>S<sub>2</sub>S<sub>3</sub> nanoparticles.



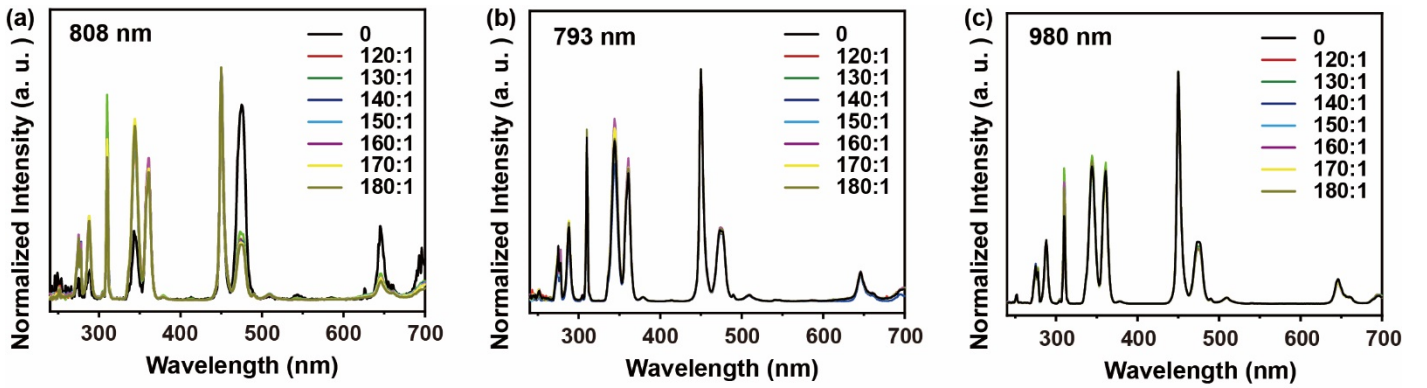
**Figure S4.** Energy-dispersive X-ray lining analysis of the as-synthesized heterogeneous core-multishell nanoparticles.



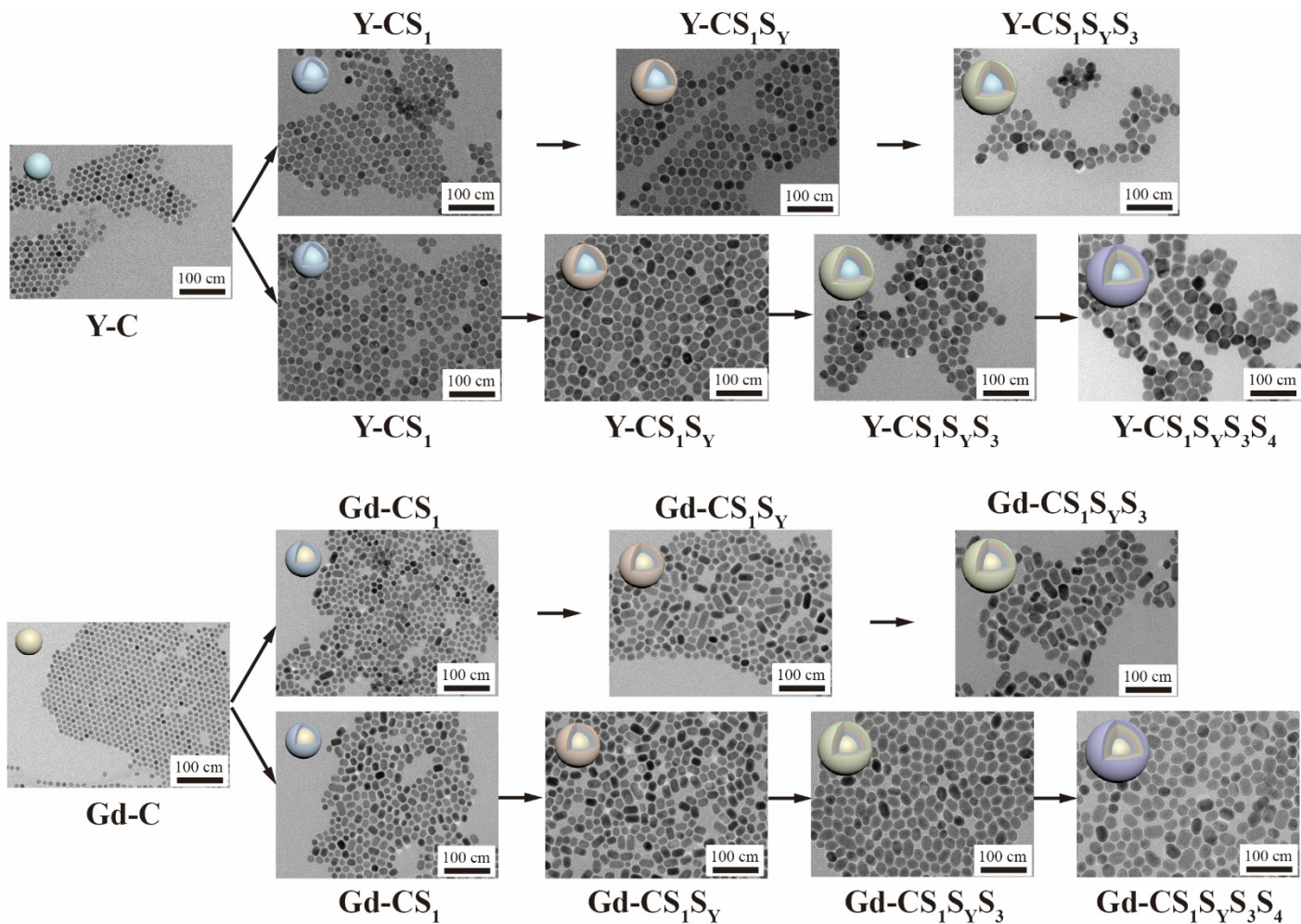
**Figure S5.** The luminescence emission spectrum of NaGdF<sub>4</sub>:18%Yb,2%Er@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd@NaGdF<sub>4</sub>(Gd-C(18%Yb,2%Er)S<sub>2</sub>S<sub>3</sub>) with and without IR-806 under 808 nm laser excitation.



**Figure S6.** The luminescence emission spectrum of NaGdF<sub>4</sub>:49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:x%Nd (x = 10%, 50%, 90%), 10%Yb@NaGdF<sub>4</sub> with and without IR-806 for 808 nm laser exciting.

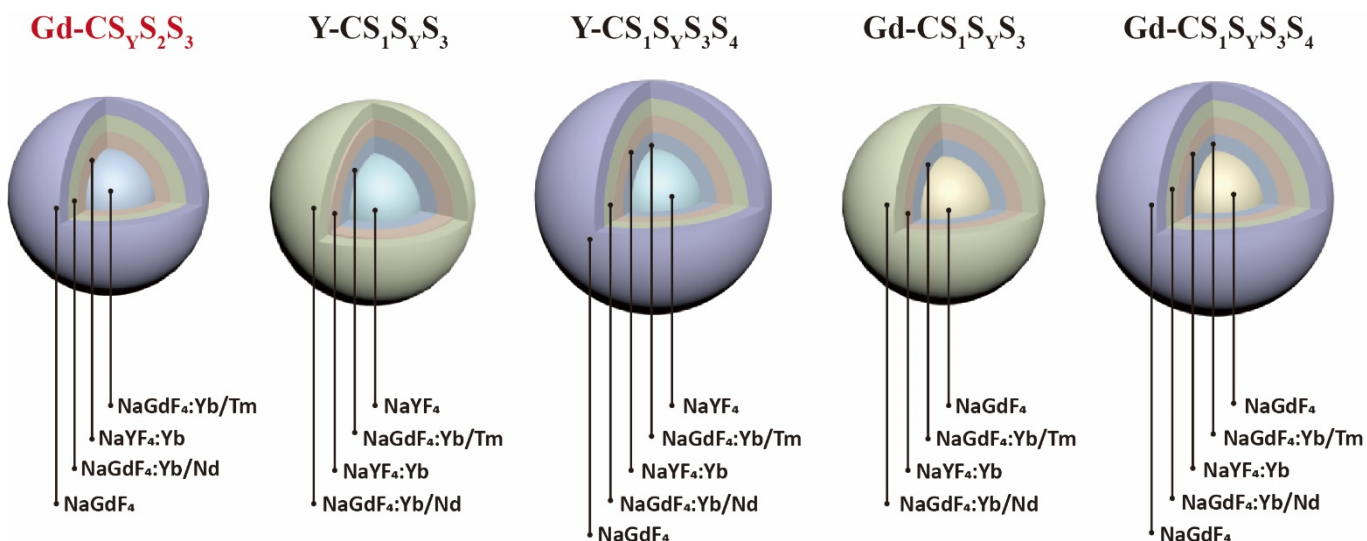


**Figure S7.** Normalized intensity of luminescence spectra of Gd-CS<sub>2</sub>S<sub>3</sub> with various content of IR-806 (m<sub>NP</sub> : m<sub>IR-806</sub> = 0, 120:1, 130:1, 140:1, 150:1, 160:1, 170:1, 180:1) under 808 nm, 793 nm and 980 nm.

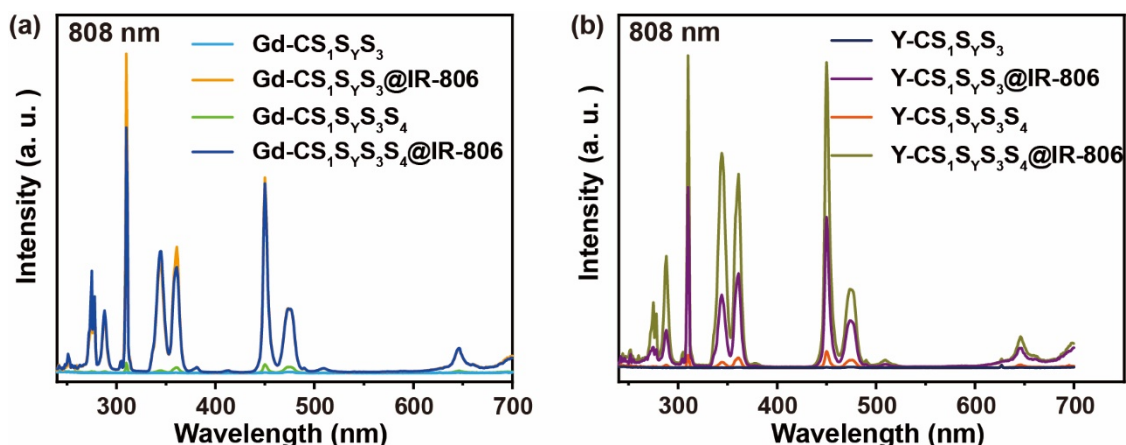


**Figure S8.** Transmission electron microscopy (TEM) and particles size distribution of as-prepared NaYF<sub>4</sub>@NaGdF<sub>4</sub>:49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd (Y-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>), NaYF<sub>4</sub>@NaGdF<sub>4</sub>:49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd@NaGdF<sub>4</sub> (Y-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>S<sub>4</sub>), NaGdF<sub>4</sub>@NaGdF<sub>4</sub>:49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd (Gd-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>), NaGdF<sub>4</sub>@NaGdF<sub>4</sub>:49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd@NaGdF<sub>4</sub> (Gd-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>S<sub>4</sub>).

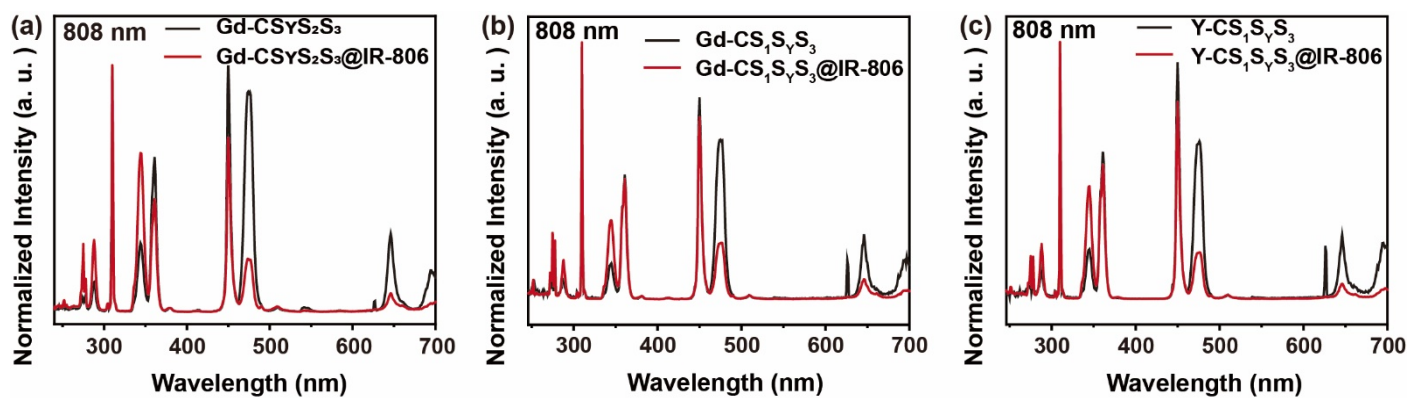




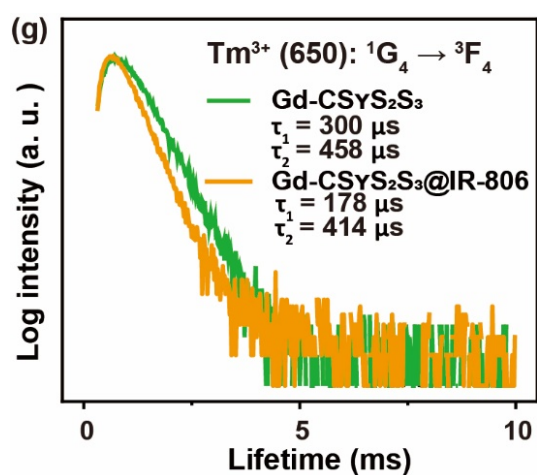
**Figure S9.** Schematic illustration of five types core-multishell structures including NaGdF<sub>4</sub>: 49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd@NaGdF<sub>4</sub> (Gd-CS<sub>Y</sub>S<sub>2</sub>S<sub>3</sub>), NaYF<sub>4</sub>@NaGdF<sub>4</sub>:49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd (Y-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>), NaYF<sub>4</sub>@NaGdF<sub>4</sub>: 49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd@NaGdF<sub>4</sub> (Y-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>S<sub>4</sub>), NaGdF<sub>4</sub>@NaGdF<sub>4</sub>: 49%Yb,1%Tm@NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd@NaGdF<sub>4</sub> (Gd-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>), NaGdF<sub>4</sub>@NaGdF<sub>4</sub>:49%Yb,1%Tm @NaYF<sub>4</sub>:20%Yb@NaGdF<sub>4</sub>:10%Yb,50%Nd@NaGdF<sub>4</sub> (Gd-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>S<sub>4</sub>).



**Figure S10.** Luminescence emission spectra of Gd-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>, Gd-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>@IR-806, Gd-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>S<sub>4</sub>, Gd-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>S<sub>4</sub>@IR-806 and Y-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>, Y-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>@IR-806, Y-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>S<sub>4</sub>, Y-CS<sub>1</sub>S<sub>Y</sub>S<sub>3</sub>S<sub>4</sub>@IR-806 under 808 nm CW diode laser, with excitation power is 10 W/cm<sup>2</sup>.



**Figure S11.** Normalized intensity of luminescence spectra of Gd-CSyS<sub>2</sub>S<sub>3</sub> and Gd-CSyS<sub>2</sub>S<sub>3</sub>@IR-806, Gd-CS<sub>1</sub>S<sub>1</sub>S<sub>3</sub> and Gd-CS<sub>1</sub>S<sub>1</sub>S<sub>3</sub>@IR-806, Y-CS<sub>1</sub>S<sub>1</sub>S<sub>3</sub> and Y-CS<sub>1</sub>S<sub>1</sub>S<sub>3</sub>@IR-806 by 808 nm laser excitation.



**Figure S12.** The decay curves of Tm<sup>3+</sup> at 650 nm in Gd-CSyS<sub>2</sub>S<sub>3</sub> and Gd-CSyS<sub>2</sub>S<sub>3</sub>@IR-806 nanoparticles under 808 nm excitation.