

## Supporting Information

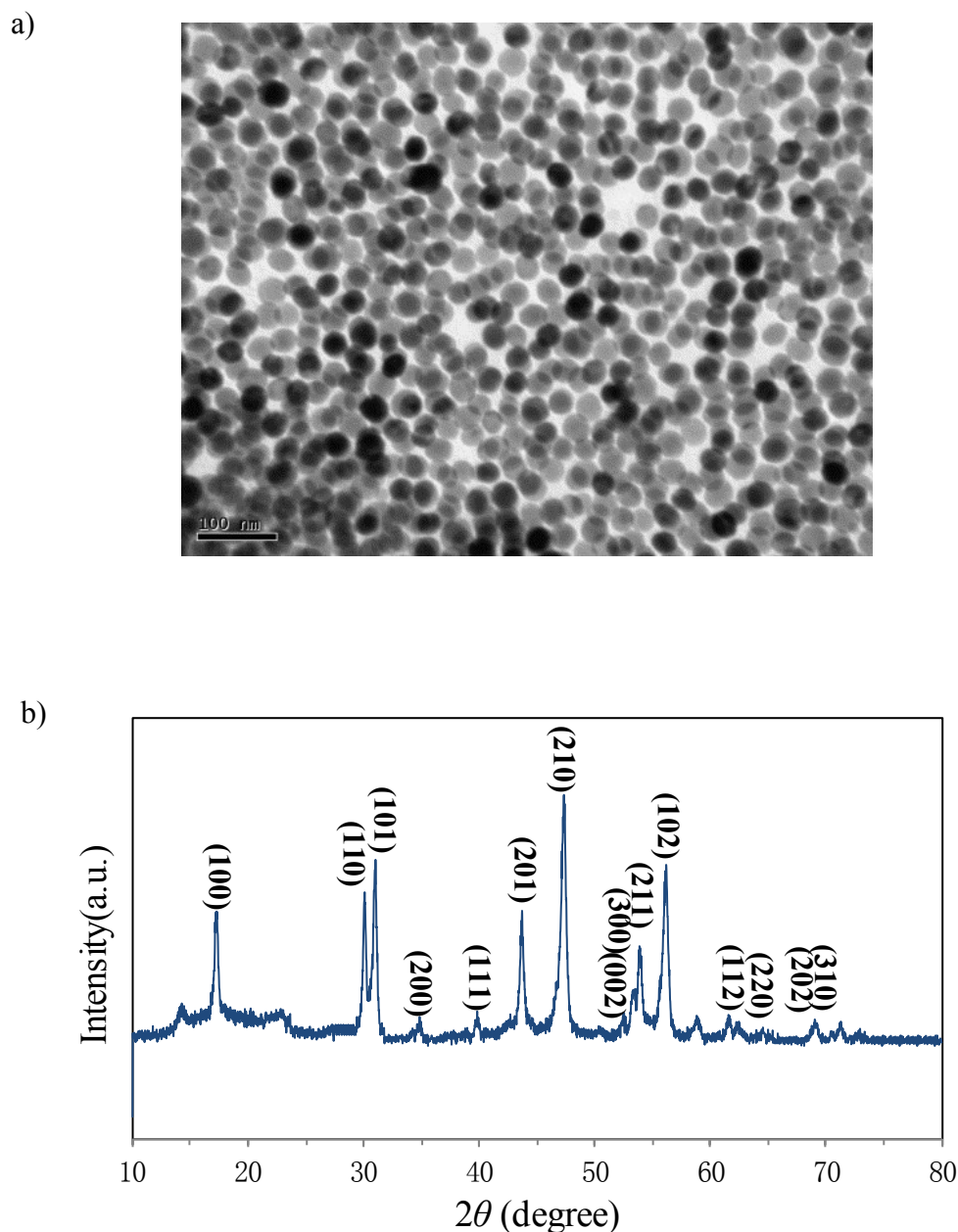
### **Large enhancement of upconversion luminescence by dye/Nd<sup>3+</sup> sensitization of quenching-shield sandwich structural upconversion nanocrystal under 808nm excitation**

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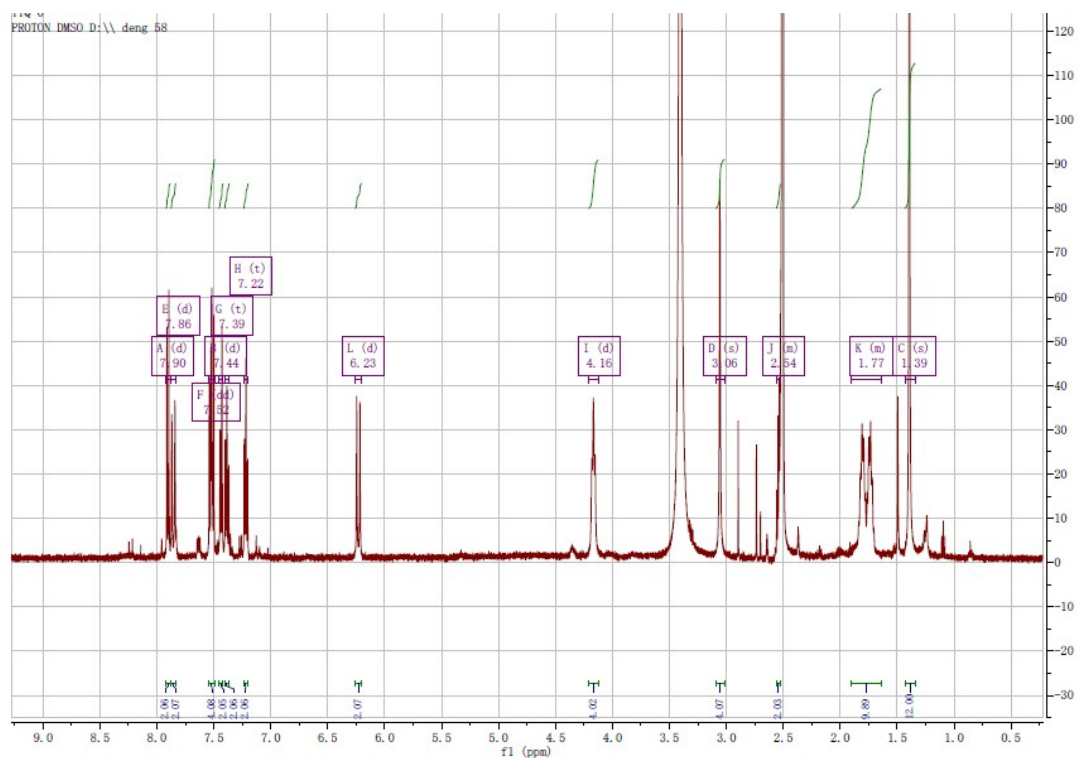


**Figure S1.** a) TEM images of the as-prepared  $\text{NaLuF}_4\text{:Gd,Yb,Tm}(24/20/0.5\%)@ \text{NaLuF}_4\text{:Gd,Yb}(24/10\%)@ \text{NaLuF}_4\text{:Yb}(10\%)$  nanocrystals; b) Experimental powder XRD patterns of the  $\text{NaLuF}_4\text{:Gd,Yb,Tm}(24/20/0.5\%)@ \text{NaLuF}_4\text{:Gd,Yb}(24/10\%)@ \text{NaLuF}_4\text{:Yb}(10\%)$  nanocrystals.

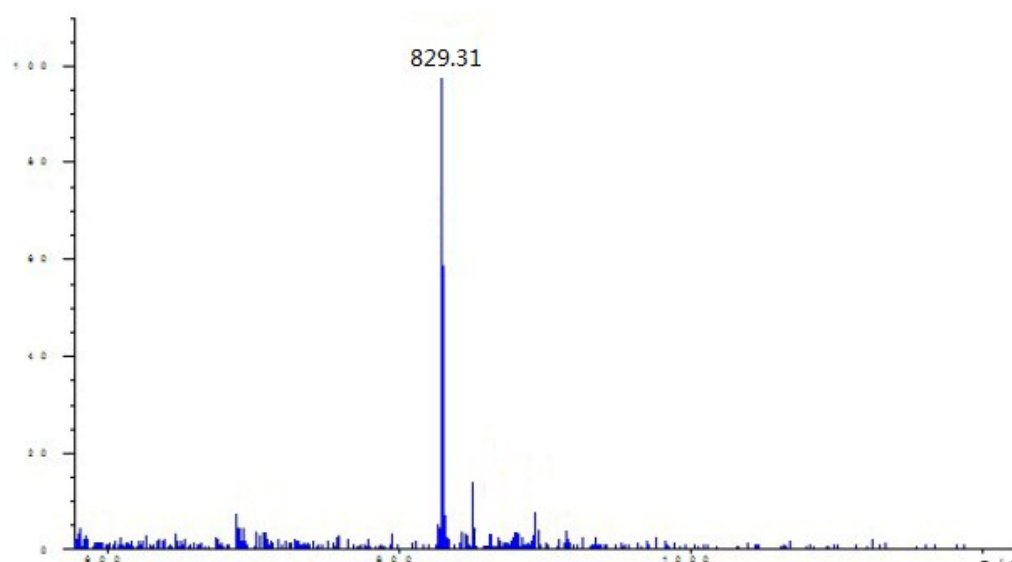
#### Synthesis of IR-820 dye

$^1\text{H-NMR}$  (500 MHz,  $d_6\text{-DMSO}$ ):  $\delta$  = 7.90 (d,  $J$  = 8.6 Hz, 2H), 7.86 (d,  $J$  = 14.0 Hz, 2H), 7.52 (dd,  $J$  = 11.9, 7.9 Hz, 4H), 7.44 (d,  $J$  = 8.0 Hz, 2H), 7.39 (t,  $J$  = 7.7 Hz, 2H), 7.22 (t,  $J$  = 7.7 Hz, 2H), 6.23 (d,  $J$  = 14.0 Hz, 2H), 4.16 (d,  $J$  = 7.5 Hz, 4H), 3.06 (s, 4H), 2.56-2.53 (m, 2H), 1.90-1.64 (m, 10H), 1.39 (s, 12H).

MS: calculated for  $\text{C}_{44}\text{H}_{49}\text{N}_2\text{O}_8\text{S}_3$ : 829.26; found: 829.31.



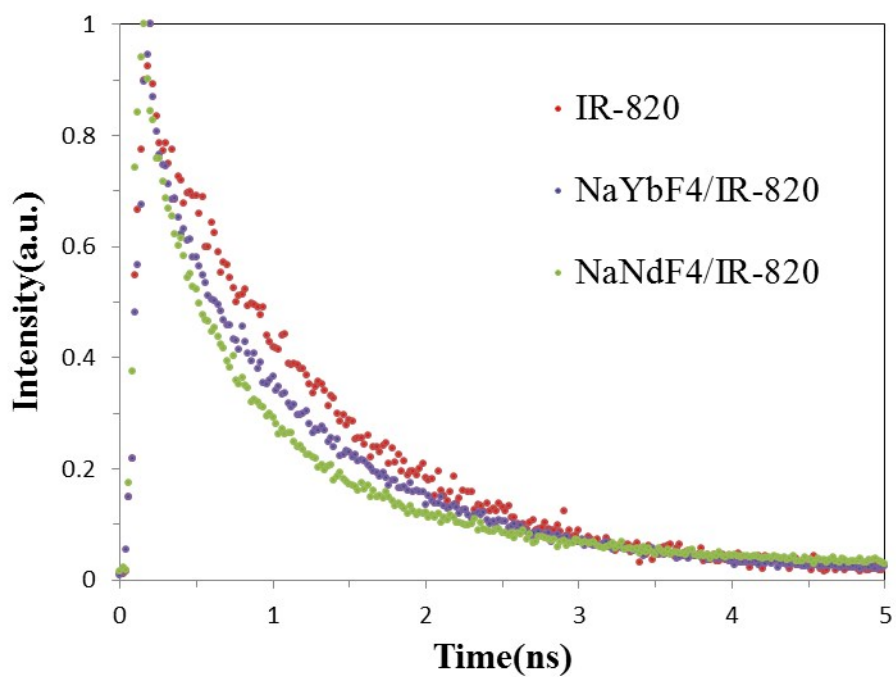
**Figure S2.**  $^1\text{H}$ -NMR (500 MHz,  $d_6$ -DMSO) spectrum of IR-820.



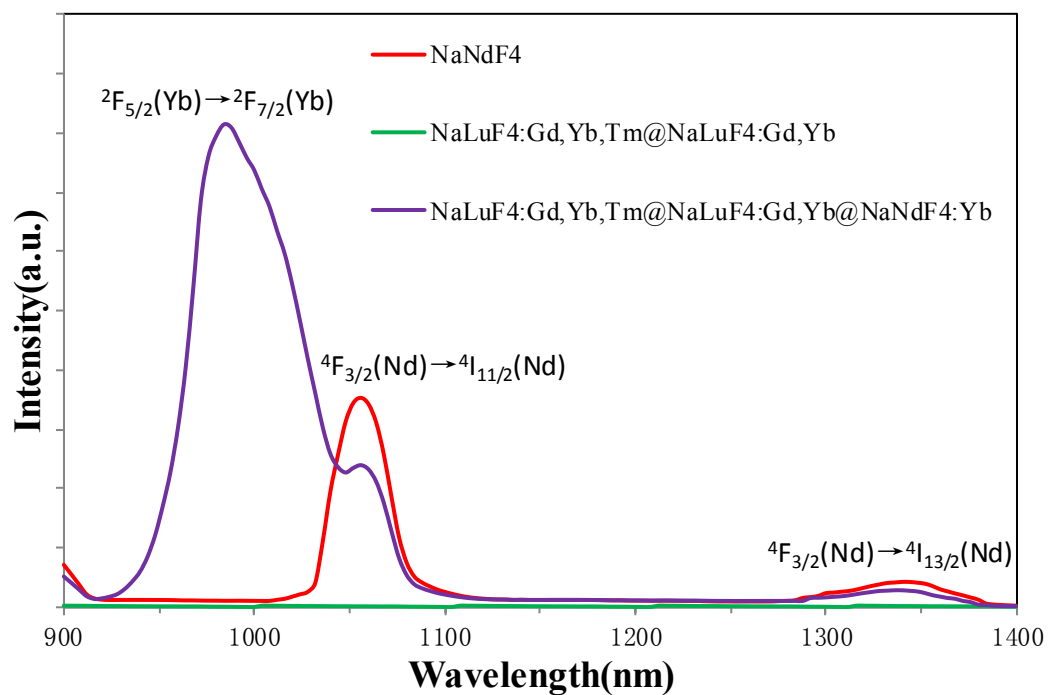
**Figure S3.** ESI Mass spectrum of IR-820



**Figure S4.** Photographic images: a) IR-820 dye; b) OA-coated nanoparticles; c) IR-820 functionalized nanoparticles.



**Figure S5.** Fluorescence decays of the IR-820 dye from the pure dye, the IR-820 dye-sensitized  $\text{NaYbF}_4$  nanoparticles as well as the IR-820 dye-sensitized  $\text{NaNdF}_4$  nanoparticles.



**Figure S6.** A comparison of downconversion luminescence (DCL) from NaNdF<sub>4</sub>, NaLuF<sub>4</sub>:Gd,Yb,Tm@NaLuF<sub>4</sub>:Gd,Yb, and NaLuF<sub>4</sub>:Gd,Yb,Tm@NaLuF<sub>4</sub>:Gd,Yb@NaNdF<sub>4</sub>:Yb nanoparticles (hexane dispersion, excited at 808 nm).