Supplementary Information for

Dye-Sensitized Lanthanide-Doped Upconversion Nanoparticles

Xindong Wang,[a] Rashid R. Valiev,[b,d] Tymish Y. Ohulchanskyy,[c] Hans Ågren,[b,a] Chunhui Yang[a] and Guanying Chen*[a]

[a]MIIT Key Laboratory of Critical Materials Technology for New Energy Conversion and Storage, School of Chemistry and Chemical Engineering & Key Laboratory of Micro-systems and Micro-structures, Ministry of Education, Harbin Institute of Technology, 150001 Harbin, People's Republic of China.

[b]Department of Theoretical Chemistry and Biology, Royal Institute of Technology, S-10691 Stockholm, Sweden

[c]College of Optoelectronic Engineering, Key Laboratory of Optoelectronic Devices and Systems of Ministry of Education and Guangdong Province, Shenzhen University, 518060 Shenzhen, Guangdong, People's Republic of China

[d]Tomsk State University, 36 Lenin Avenue, 634050, Tomsk, Russia

*Correspondence: chenguanying@hit.edu.cn

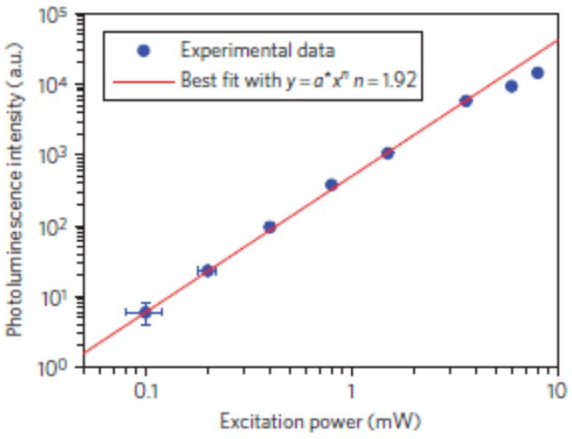

Figure S1. Upconversion photoluminescence intensity as a function of excitation power. Reprinted with permission from Ref. 7 (Copyright 2012, Nature Publishing Group). The upconversion intensity of IR806-sensitized $NaYF_4$:Yb,Er nanoparticles (integrated in the range 500–685 nm) as a function of excitation power from an 800 nm laser. The number of photons which are required to populate the upper emitting state under unsaturated condition can be obtained by the relation,

$$I_f \propto P^n \tag{S1}$$

Where I_f is the luminescence intensity, P is the pump laser power, and n is the number of laser photons required. A best fit with a power function yields the exponent factor n=1.92, indicating a purely two-photon process.

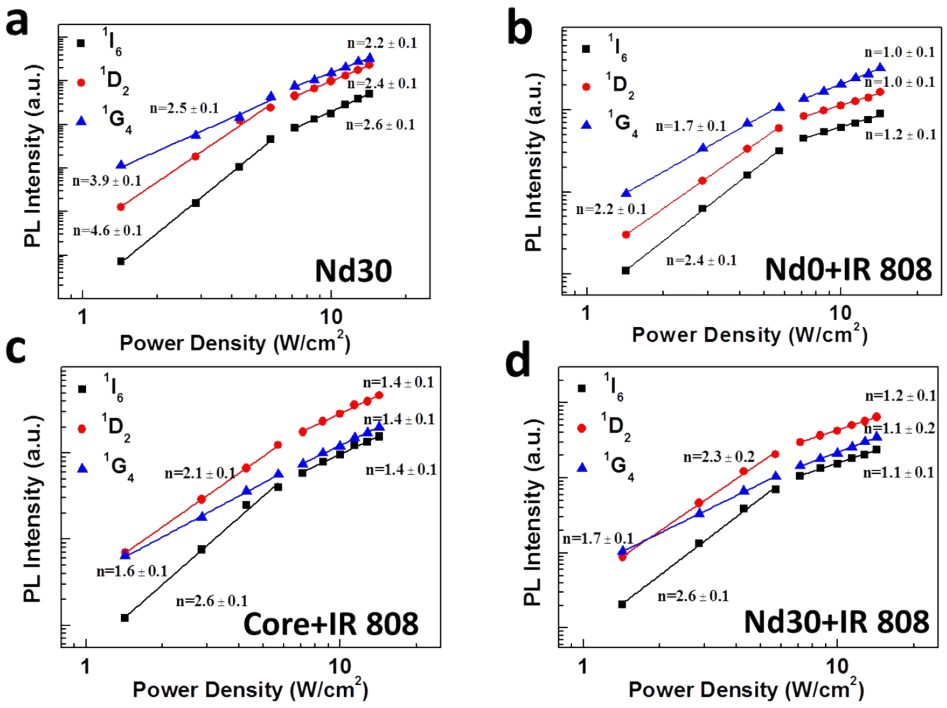

Figure S2. The dependence of the intensities of upconverted luminescence from the $^1I_6$, $^1D_2$, $^1G_4$ states of Tm^{3+} ions from ($NaYbF_4$: Tm^{3+} 0.5%)@$NaYF_4$: Nd^{3+} 30% (Nd30, a), IR-808 sensitized ($NaYbF_4$: Tm^{3+} 0.5%)@$NaYF_4$ (Nd0+IR-808, b), IR-808 sensitized $NaYbF_4$: Tm^{3+} 0.5% (core+IR-808, c), and IR-808 sensitized ($NaYbF_4$: Tm^{3+} 0.5%)@$NaYF_4$: Nd^{3+} 30% (Nd30+IR-808, d) nanoparticles on the excitation power density. Reprinted with permission from Ref. 8. Copyright 2015, American Chemical Society.

Slope values of 2.5, 3.9, and 4.6 for an unsaturated situation below 6 W/cm^2 in ($NaYbF_4$: Tm^{3+} 0.5%)@$NaYF_4$: Nd^{3+} 30% nanoparticles indicate that three-, four-, and five photon processes are involved to populate the $^1G_4$, $^1D_2$, and $^1I_6$ state, respectively (Figure S2 a). However, saturation effects take place for all IR-808 sensitized samples and for all laser power excitation range (Figure S2 b-d). In particular, slope values close to n=1 are observed for all the emitting states of all IR-808 sensitized samples with excitation density above 6 W/cm^2. These results illustrate that the upconverting rate at each intermediate state is significantly higher than its decay rate, in line with the reported high upconversion efficiency.