Supporting Information

New role of Yb³⁺—energy reservoir for lanthanide upconversion

luminescence

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Figure S1. Variation of the integral UC intensity of Nd^{3+} single doped $NaYF_4$ NCs with the Nd^{3+} concentration. The integral intensities are normalized by that of 1 mol% Nd^{3+} doped NCs. Insets are the dark-field and bright-field luminescent images of the most efficient 1 mol% Nd^{3+} doping $NaYF_4$ NCs dispersed in cyclohexane with a density of ~0.07 mmol/mL excited at a pumping power of ~1.7 W with the 800 nm laser diode.



Figure S2. Top: TEM photographs of various Nd^{3+}/Yb^{3+} codoped $NaYF_4$ NCs, scale bars are 100 nm. Bottom: XRD pattern of 1 mol% $Nd^{3+}:NaYF_4$, indicating the hexagonal phase of the formed NCs.



Figure S3. Possible transitions of Nd³⁺ overlapping the observed spectrum (bottom of the figure).



Figure S4. Comparison of the integral UC intensities of $10Nd^{3+}/5Yb^{3+}:NaYF_4$, $10Nd^{3+}/5Yb^{3+}:NaYF_4@NaYF_4$, and $10Nd^{3+}:NaYF_4@5Yb^{3+}:NaYF_4$ NCs.



Figure S5. (a) NIR spectra and (b) intensity ratio between the Yb³⁺ emission and the Nd³⁺ emission in low Nd³⁺ doped NaYF₄ NCs; (c) NIR spectra and (d) intensity ratio between the Yb³⁺ emission and the Nd³⁺ emission in high Nd³⁺ doped NaYF₄ NCs. The gradually increasing intensity ratio with Yb³⁺ concentration in both low Nd³⁺ and high Nd³⁺ doping situations clearly manifest the FET process of Nd³⁺ \rightarrow Yb³⁺.



Figure S6. Ln-Ln power dependences of (a) Nd^{3+} doped and (b) Nd^{3+}/Yb^{3+} codoped $NaYF_4 NCs$.



Figure S7. (a) UC spectrum of 0.5Tm³⁺/10Yb³⁺ codoped NaYF₄ NCs upon 800 nm excitation. The green and red emission bands of Er³⁺ evidence the existence of the Er³⁺ impurity. (b) Simplified energy-level diagrams depicting the possible cross-relaxation (CR) quenching mechanisms between Er³⁺ and Tm³⁺ under 800 nm excitation.



Figure S8. TEM images of $NaErF_4@NaYF_4$ and $Na(Er_{0.9}/Yb_{0.1})F_4@NaYF_4$ NCs. Scale bars are 100 nm.



Figure S9. Effects of Yb^{3+} on the UC spectra of Er^{3+} upon 800 nm excitation in low Er^{3+} doped NCs.



Figure S10. Upconversion spectra of $Na(Er/Yb)F_4@NaYF_4$ nanocrystals upon 800, 980, and 1530 nm excitation using identical excitation intensity.



Figure S11. Decay profiles of the Er^{3+} 540 nm emission of NaErF₄@NaYF₄ core-shell NCs and Na($Er_{0.9}$ /Yb_{0.1})F₄@NaYF₄ core-shell NCs upon 1530 nm excitation.



Figure S12. Upconversion spectra of $Er^{3+}\!\!:\!\!Y_2O_3$ and $Er^{3+}\!/Yb^{3+}\!:\!Y_2O_3$ ceramics.

	1Nd	10Nd	20Nd	30Nd	40Nd	60Nd	90Nd	100Nd
0Yb	1.00	0.64	0.21	0.23	0.24			0.22
1Yb	1.83	2.48						NA
2Yb	2.18	4.96						NA
3Yb	2.30	6.25						NA
5Yb	1.93	7.06	4.89	2.87	2.25		1.59	NA
10Yb	1.46	6.13	3.71	3.39	3.58		1.71	NA
20Yb	1.09	5.81	3.21	2.79	2.65		NA	NA
30Yb	0.65	2.58	2.41	1.86	2.21		NA	NA
40Yb	0.41					0.91	NA	NA
60Yb	0.29					NA	NA	NA
70Yb				0.82	NA	NA	NA	NA
80Yb	0.19			NA	NA	NA	NA	NA
99Yb	0.11	NA						

Table S1. Integral intensity of visible light of Nd^{3+}/Yb^{3+} :NaYF₄. All intensities were normalized to the value of 1 mol% Nd^{3+} NCs. Those impossible compositions are labeled as NA. Two typical intensities of $1Nd^{3+}$ and $10Nd^{3+}/5Yb^{3+}$ NCs were given in bold font, respectively, corresponding to the most efficient Nd^{3+} single doped and Nd^{3+}/Yb^{3+} co-doped NCs.