Electronic Supplementary Material (ESI) for Nanoscale
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Simultaneous enhancement of red upconversion luminescence and CT contrast of $NaGdF_4$:Yb,Er nanoparticles *via* Lu^{3+} doping

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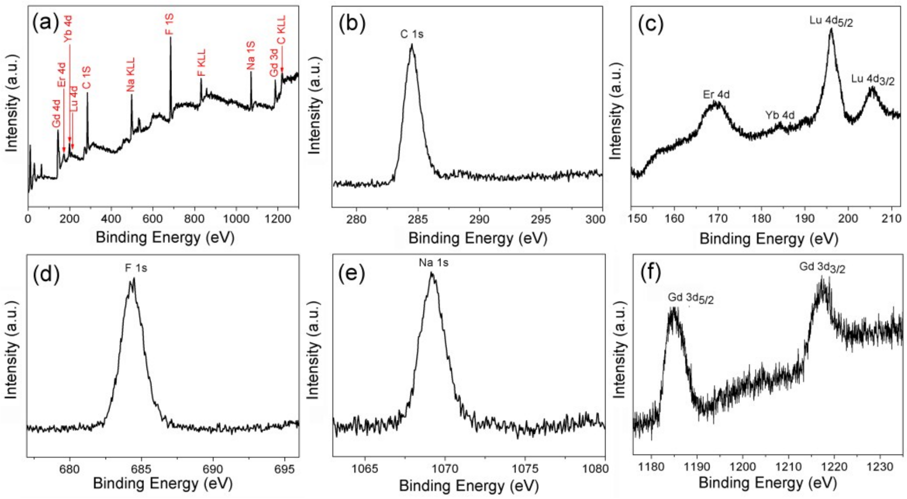

Fig. S1 (a) XPS survey spectrum and (b) C 1s, (c) Yb 4d, Er 4d, Lu $4d_{5/2}$, and Lu $4d_{3/2}$, respectively, (d) F 1s, (e) Na 1s, (f) Gd $3d_{5/2}$ and Gd $3d_{3/2}$ spectra of $NaGdF_4$:18%Yb,2%Er,2.5%Lu NPs.

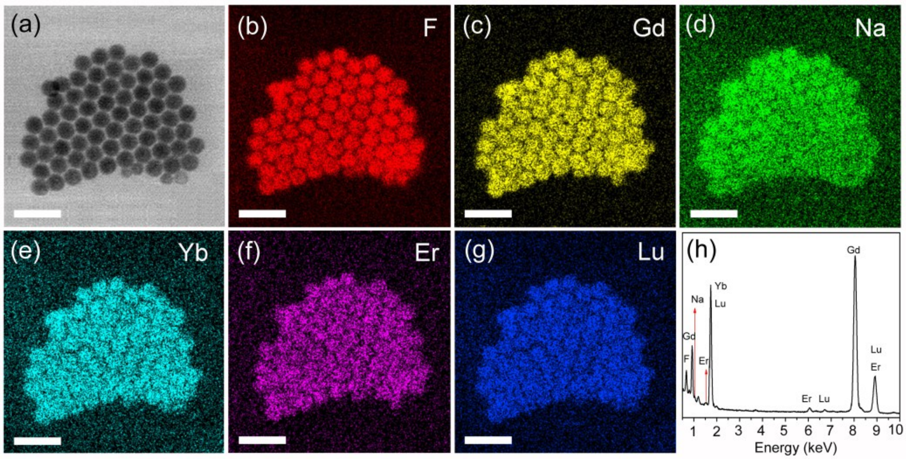

Fig. S2 (a) STEM image (b-g) EDX elemental mapping of $NaGdF_4$:18%Yb,2%Er,2.5%Lu NPs and line-profile analysis of $NaGdF_4$:18%Yb,2%Er,2.5%Lu NPs with different elements (F, Gd, Na, Yb, Er and Lu), (h) EDX spectrum of $NaGdF_4$:18%Yb,2%Er,2.5%Lu NPs. The scale bar is 25 nm.

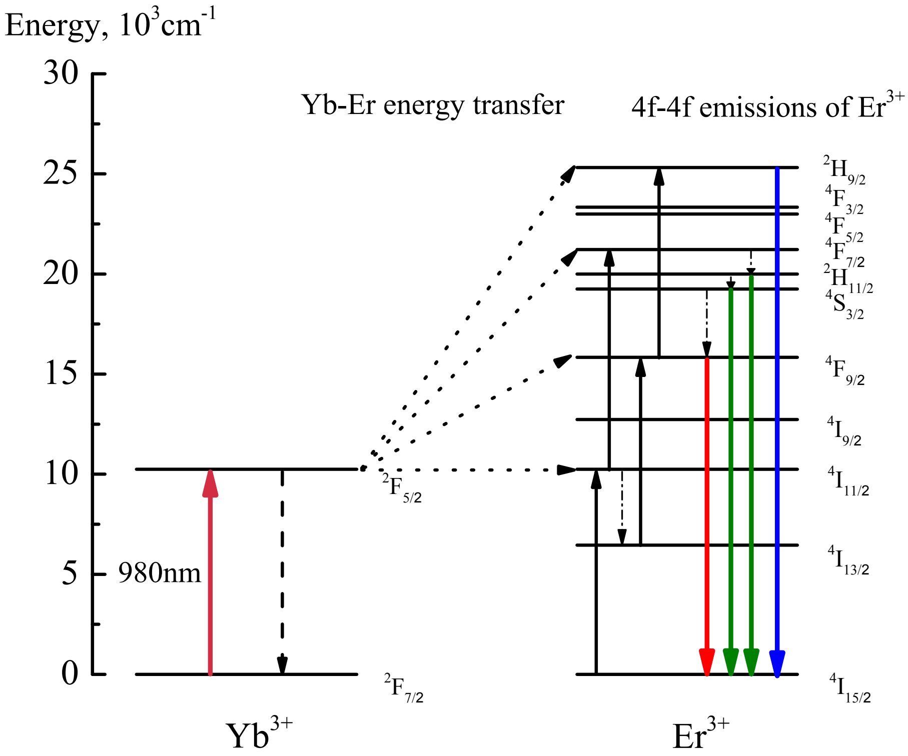

Fig. S3 The energy level diagram for the *4f* electronic configurations of Yb^{3+} and Er^{3+} ions and the upconversion luminescence mechanism of Yb^{3+}/Er^{3+}-codoped materials with the excitation of 980 nm.

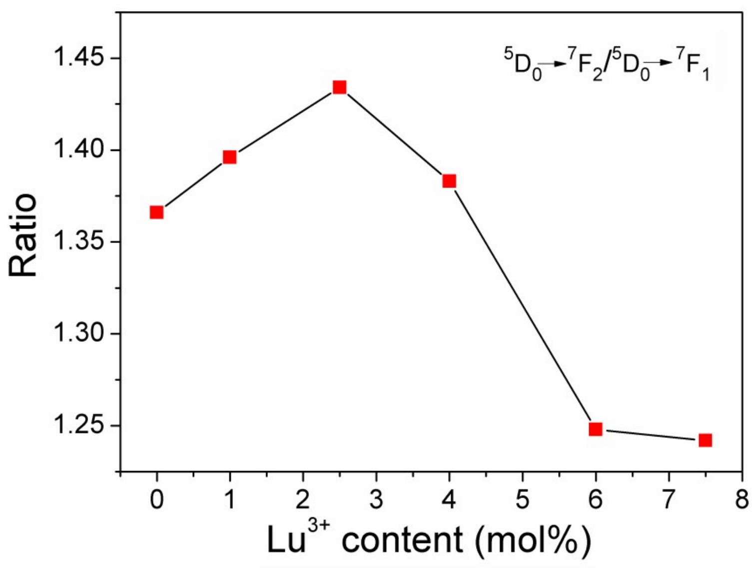

Fig. S4 The dependence of the ratio of intensity ($^5D_0 \rightarrow {}^7F_2/{}^5D_0 \rightarrow {}^7F_1$) on Lu^{3+} doping content in β-$NaGdF_4$:1%Eu NPs doped with different concentrations of Lu^{3+} ions.

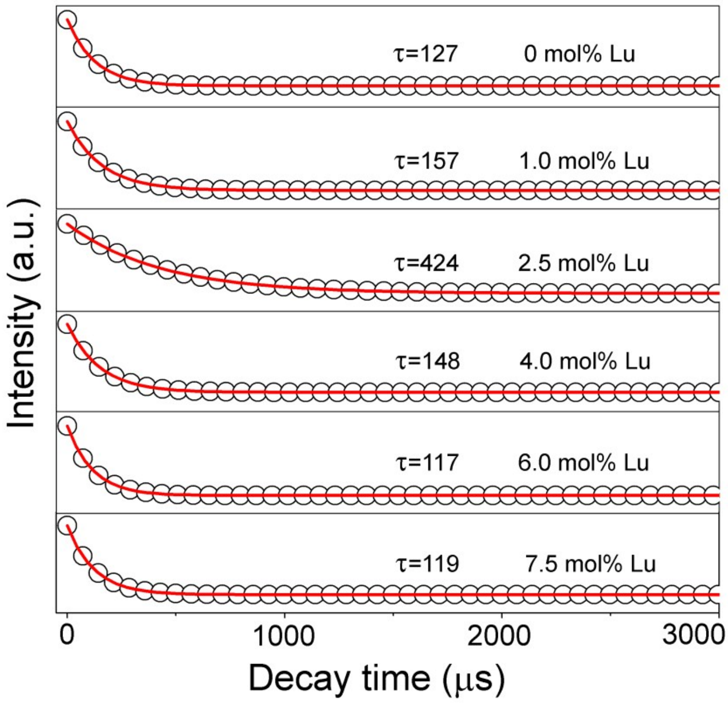

Fig. S5 Temporal evolution of UCL from the $^4F_{9/2}$ level of Er^{3+} in the β-$NaGdF_4$:Yb,Er,*X*%Lu NPs (X = 0, 1, 2.5, 4, 6 and 7.5) under the excitation of a 980 nm pulsed Raman shift laser: experimental data (black circles) and fitting by single-exponential function ($I = I_0\exp(-t/\tau)$) (red solid line).

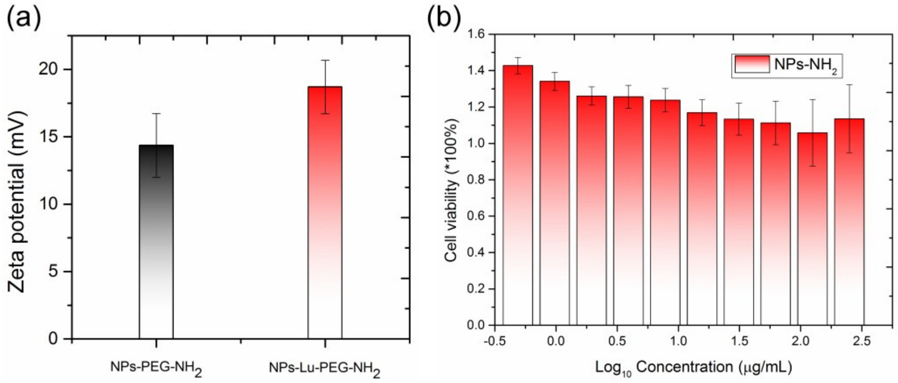

Fig. S6 (a) The zeta potential of NH_2-PEGylated-$NaGdF_4$:Yb,Er (NPs-PEG-NH_2) and $NaGdF_4$:Yb,Er,2.5%Lu NPs (NPs-Lu-PEG-NH_2). (b) *In vitro* cell viabilities of HepG-2 cells with NPs-NH_2 of different concentrations for 24 h.

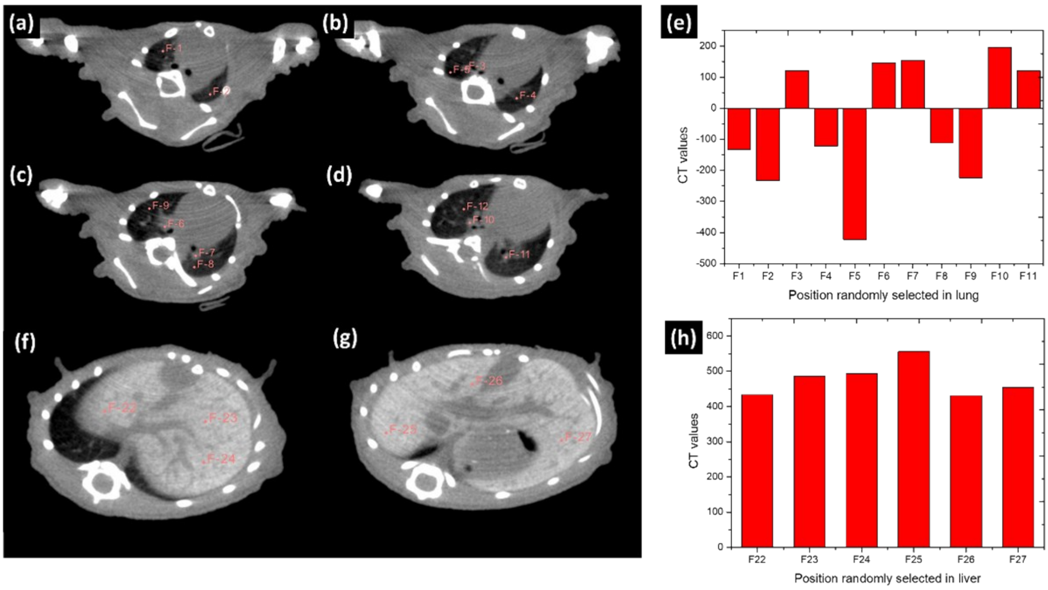

Fig. S7 The computed tomography images as well as CT values of liver and lung. (a-d) The computed tomography images of lung. (e) CT values of lung. (f-g) The computed tomography images of liver. (h) CT values of liver.

Description of the upconversion luminescence (UCL) mechanism

The UCL mechanism can be described by the energy level diagram for the *4f* electronic configurations of Yb^{3+} and Er^{3+} ions,[1] as shown in Fig. S3. The laser excitation of 980 nm widely used in the UCL experiments can pump Yb^{3+} ion in the ground state $^2F_{7/2}$ to its excited state $^2F_{5/2}$. And then such excited Yb^{3+} ions undergo the radiative and non-radiative deexcitation processes, as shown by the emission transition $^2F_{5/2} \rightarrow {}^2F_{7/2}$ and the energy transfer (ET) between Yb^{3+} and Er^{3+} ions. The later ET can effectively induce one and more resonant absorption transitions of Er^{3+} ion, such as $^4I_{15/2} \rightarrow {}^4I_{11/2}$, $^4I_{11/2} \rightarrow {}^4F_{7/2}$, $^4I_{13/2} \rightarrow {}^4F_{9/2}$ and $^4F_{9/2} \rightarrow {}^2H_{9/2}$, to form the two- and three-photon UC excitations with the help of the multiphoton relaxation processes between those $4f^{11}$ energy levels close to each other. And thus the four luminescent energy levels $^4F_{9/2}$, $^4S_{3/2}$, $^2H_{11/2}$ and $^2H_{9/2}$ of Er^{3+} ion can be sufficiently populated so as to generate the red, green and blue emissions to the ground state $^4I_{15/2}$, as observed in most UCL experiments.

Reference

[1] W. T. Carnall, H. Crosswhite, H. M. Crosswhite, *Energy Level Structure and Transition Probabilities in the Spectra of the Trivalent Lanthanides in LaF_3*, Argonne National Laboratory Report ANL 78-XX-95: Lemont, IL, 1978.