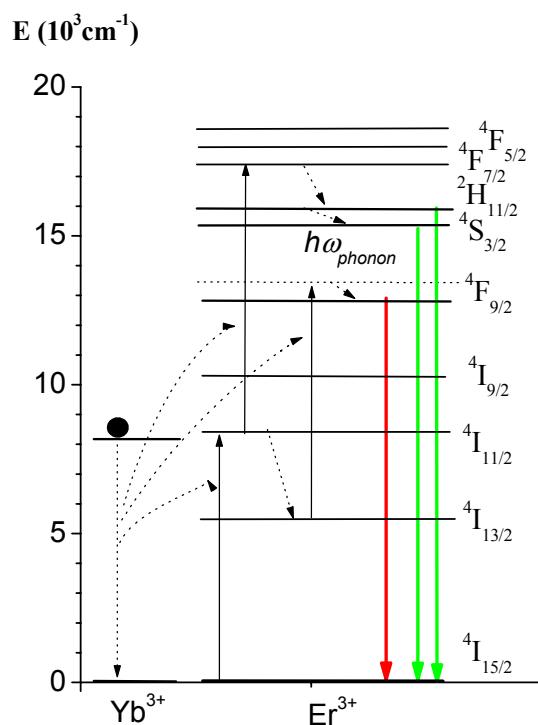


## Supporting information

### S1. Upconversion Mechanism

The schematic of the populating and UCL processes for the green and red emissions was drawn in Figure S1. In  $\text{Yb}^{3+}$  and  $\text{Er}^{3+}$  co-doped systems,  $\text{Yb}^{3+}$  ions act as sensitizers and  $\text{Er}^{3+}$  ions as activators. The green and red UCL both occur via a two-step energy transfer from the  $\text{Yb}^{3+}$  to the  $\text{Er}^{3+}$ . First, the  $\text{Er}^{3+}$  ion is excited from the ground state  $^4\text{I}_{15/2}$  to the excited state  $^4\text{I}_{11/2}$  via energy transfer (ET) of neighboring  $\text{Yb}^{3+}$  and  $\text{Er}^{3+}$ . Subsequent nonradiative relaxations of  $^4\text{I}_{11/2}$ - $^4\text{I}_{13/2}$  populate the  $^4\text{I}_{13/2}$  level. In the second-step excitation, the same laser pumps the excited-state atoms from the  $^4\text{I}_{11/2}$  to the  $^4\text{F}_{7/2}$  levels via ET and excited-state absorption (ESA), or from the  $^4\text{I}_{13/2}$  to  $^4\text{F}_{9/2}$  states via phonon-assisted ET. The populated  $^4\text{F}_{7/2}$  may mostly nonradiatively relax to two lower levels:  $^2\text{H}_{11/2}$  and  $^4\text{S}_{3/2}$ , which produce two green upconversion emissions. The populated  $^4\text{F}_{9/2}$  level of the  $\text{Er}^{3+}$  ion most relaxes radiatively to the ground state  $^4\text{I}_{15/2}$  level, which causes red emissions.



**Figure S1** Upconversion population processes in Yb, Er codoped system under 980-nm laser-diode excitation.

## S2. R<sub>HS</sub> deduced at different conditions

Table S1. The R<sub>HS</sub> values of NaYF<sub>4</sub>:Yb<sup>3+</sup>,Er<sup>3+</sup> and NaYF<sub>4</sub>:Yb<sup>3+</sup>,Er<sup>3+</sup>@Ag NPs in powders

Power Density(W/mm <sup>2</sup> )	R <sub>HS</sub> of NaYF <sub>4</sub> in powder	R <sub>HS</sub> of NaYF <sub>4</sub> @ Silver in powder
0.52	0.301887	0.441767
1	0.350063	0.61453
1.44	0.400794	0.74876
2	0.461505	0.873939
2.48	0.513187	1.022209
2.92	0.550917	1.140845
3.44	0.603927	1.237279
3.96	0.665154	1.361868
4.44	0.715277	1.438503
5	0.766843	1.555951
5.36	0.813674	1.612383
5.8	0.854898	1.669049
6.24	0.911843	1.792619
6.72	0.984386	1.866111
7.16	1.0304	1.93661
7.68		2.037462

Table S2. The R<sub>HS</sub> values of NaYF<sub>4</sub>:Yb/Er and NaYF<sub>4</sub>:Yb/Er@Silver NPs in water

Power Density(W/mm <sup>2</sup> )	R <sub>HS</sub> of NaYF <sub>4</sub> in water	R <sub>HS</sub> of NaYF <sub>4</sub> @ Silver in water
3.96	0.258696	0.273891
5	0.25611	0.275723
5.8	0.264016	0.273973
6.72	0.26183	0.277136
7.68	0.257585	0.268888
8.64	0.257436	0.273714
9.56	0.262309	0.273782
10.56	0.260779	0.27621
11.56	0.260766	0.274112
12.64	0.256829	0.282844
13.6	0.26271	0.289084
14.56	0.263017	0.290598
15.52	0.258301	0.305525