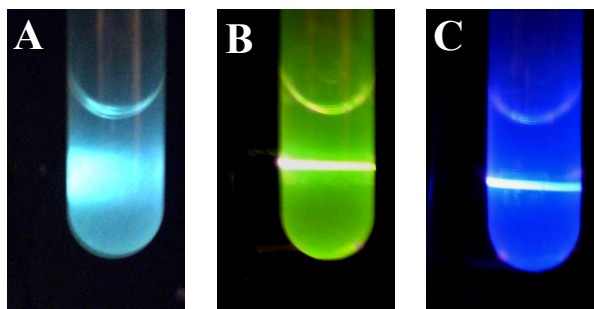
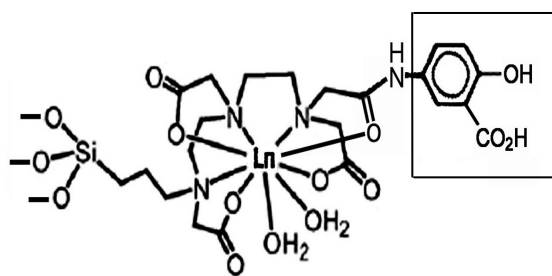


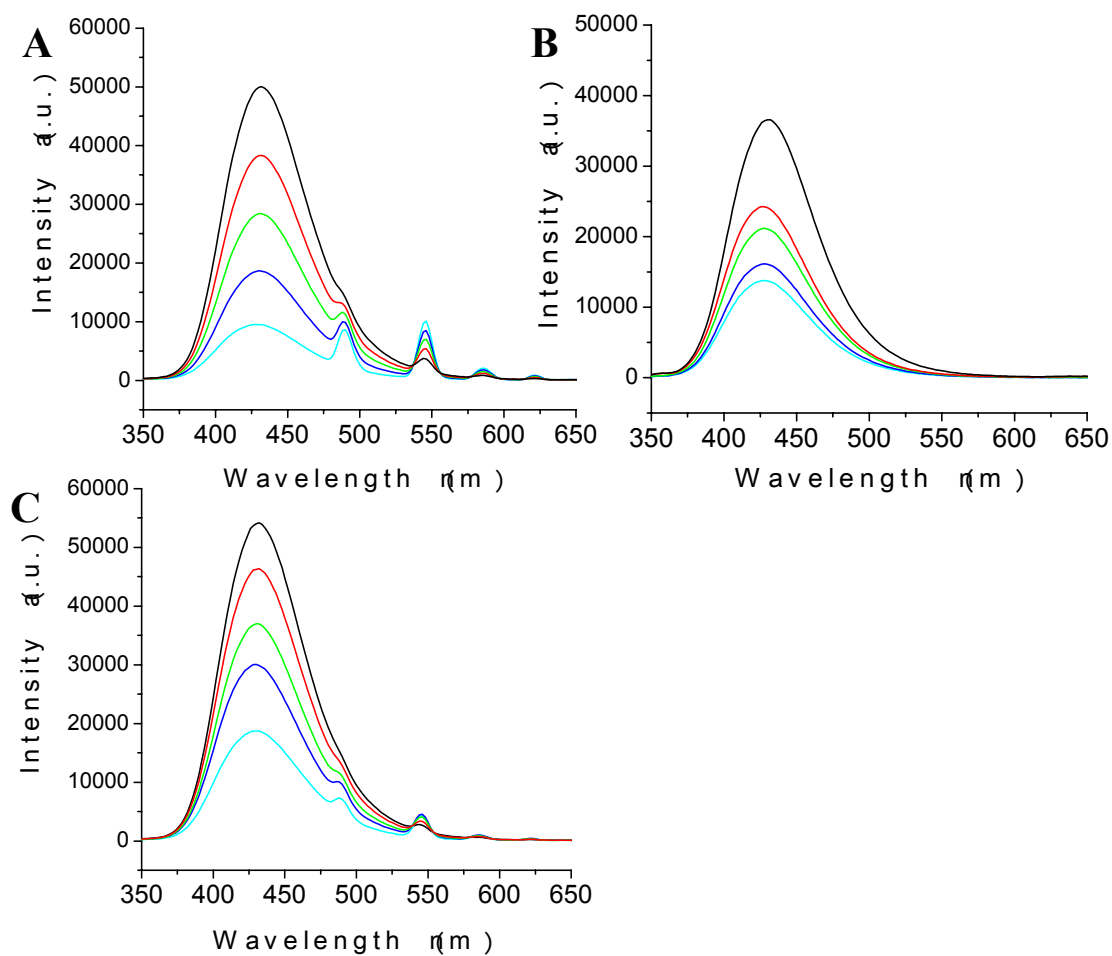
## Supporting information



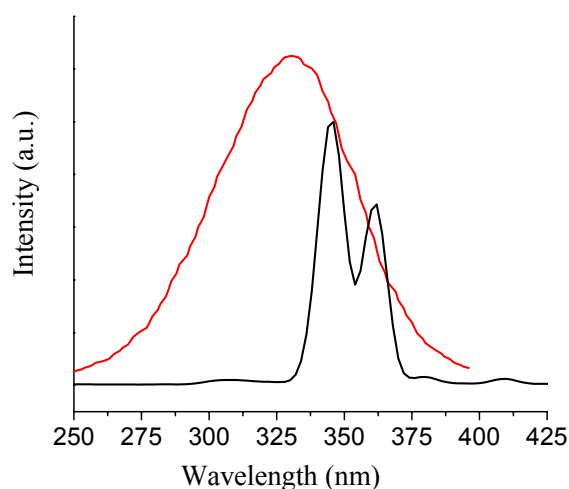
**Figure S1.** Fluorescence images of aqueous nanoparticles solutions. (A) NaYF<sub>4</sub>:Yb,Er@Si-DTPA-4-AS-Tb nanoparticles under excitation of a UV lamp at 325 nm; (B) NaYF<sub>4</sub>:Yb,Er@Si-DTPA-4-AS-Tb nanoparticles under excitation of a 980 nm NIR laser; (C) NaYF<sub>4</sub>:Yb,Tm@Si-DTPA-4-AS nanoparticles under excitation of a 980 nm NIR laser.



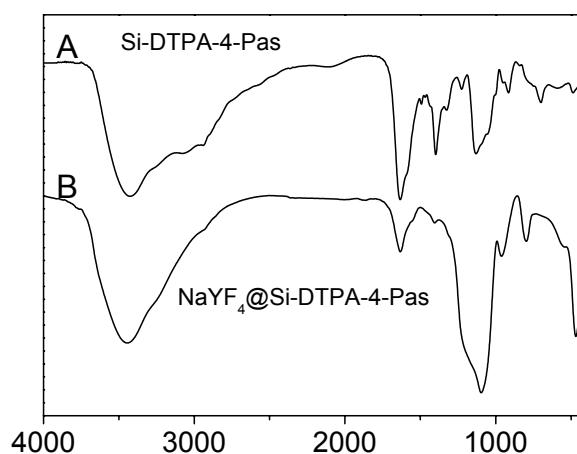
**Figure S2.** Schematic illustration of a single Si-DTPA-5-AS-Ln complex. The steric position of -OH group and -COOH group in Si-DTPA-5-AS is different from that in Si-DTPA-4-AS.



**Figure S3.** Fluorescence emission spectra of 1 mM  $\text{NaYF}_4@Si-5-AS-Gd$  nanoparticles solution metalized with lanthanide ions of different concentrations (0.2 mM, black line; 0.4 mM, red line; 0.6 mM, green line; 0.8 mM blue line; 1 mM, cyan line). (A) Pure Tb; (B) Pure Gd; (C) Equal amount of Tb and Gd.



**Figure S4.** Excitation spectrum of Si-DTPA-4-AS-Tb complex (red line) and up-conversion emission spectrum of NaYF<sub>4</sub>:Yb,Tm nanocrystals under a 980 nm laser. The emission band of the nanocrystals overlaps with the excitation band of the complex, indicating that it is possible to excite the complex by using NIR light through a FRET process.



**Figure S5.** FTIR spectra of (A) pure Si-DTPA-4-AS and (B) NaYF<sub>4</sub>@Si-DTPA-4-AS nanoparticles. After coating a layer of silica and Si-DTPA-4-AS complex on NaYF<sub>4</sub> nanocrystals, the characteristic peaks of the complex could still be clearly observed although the intensity is low due to the low doping concentration (1/10). The strong peak at 1230~1025 cm<sup>-1</sup> owes to SiO<sub>2</sub>, while the peak at 466 cm<sup>-1</sup> corresponds to the lattice vibration of NaYF<sub>4</sub> nanocrystals.