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Supporting Information

## Multi-wavelength excitable mid-infrared luminescence and energy transfer in core-shell nanoparticles for nanophotonics

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## **Supporting Figures 1-17**



**Figure S1.** TEM images of the as-synthesized NaErF<sub>4</sub>:Ho(10 mol%) core (left) and NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub> core-shell (right) nanoparticles.



**Figure S2.** XRD patterns of NaErF<sub>4</sub>:Ho(10 mol%) core and NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub> core-shell nanoparticles. The card JCPDS 27-0689 is from hexagonal phase NaErF<sub>4</sub>.



**Figure S3.** Mid-infrared emission spectra of NaYF<sub>4</sub>:Er/Ho(20-99/1 mol%)@NaYF<sub>4</sub> core-shell nanoparticles under 808 nm (left) and 980 nm (right) excitation, respectively.



**Figure S4.** Near-infrared (left) and upconverted (right) emission spectra of NaYF<sub>4</sub>:Er/Ho(20-99/1 mol%)@NaYF<sub>4</sub> core-shell nanoparticles under 980 nm excitation, respectively.



**Figure S5.** Mid-infrared emission spectra of  $NaErF_4$ :Ho(1-20 mol%)@NaYF<sub>4</sub> coreshell nanoparticles under 808 nm (left) and 980 nm (right) excitations, respectively.



Figure S6. TEM images of the as-synthesized  $NaYF_4$ : Yb/Ho(20/2 mol%) core (left) and  $NaYF_4$ : Yb/Ho(20/2 mol%) core-shell (right) nanoparticles.



**Figure S7.** Mid-infrared emission spectra of NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub> coreshell nanoparticles with various inert shell thicknesses under 808 nm (a) and 980 nm (b) excitations, respectively.



**Figure S8.** Upconversion emission spectra of  $NaErF_4$ :Ho(10 mol%)@NaYF<sub>4</sub> coreshell nanoparticles with various inert shell thicknesses under 808 nm (a), 980 nm (b) and 1530 nm excitations, respectively.



**Figure S9.** Mid-infrared emission spectra of NaErF<sub>4</sub>@NaYF<sub>4</sub>:Ho(1-30 mol%)@NaYF<sub>4</sub> samples under 808 nm (a) and 980 nm (b) excitations, respectively.



**Figure S10.** Comparative mid-infrared emission spectra of NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub> and NaErF<sub>4</sub>@NaYF<sub>4</sub>:Ho(15 mol%)@NaYF<sub>4</sub> samples under 808 nm (a) and 980 nm (b) excitations, respectively.



Figure S11. Comparative near-infrared emission spectra of  $NaErF_4@NaYF_4$ ,  $NaErF_4@NaYF_4$ :Ho(15 mol%)@NaYF\_4 and  $NaErF_4$ :Ho(10 mol%)@NaYF\_4 samples under 980 nm excitation.



**Figure S12.** Comparative upconversion emission spectra of NaErF<sub>4</sub>@NaYF<sub>4</sub>:Ho(15 mol%)@NaYF<sub>4</sub> and NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub> samples under 808 nm (a), 980 nm (b) and 1530 nm (c) excitations, respectively.



**Figure S13.** Upconversion emission spectra of NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub>:Yb (20-100 mol%)@NaYF<sub>4</sub> core-shell-shell nanoparticles under 980 nm excitation.



**Figure S14.** Mid-infrared emission spectra of NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub>:Yb(20-100mol%)@NaYF4 core-shell-shell nanoparticles under 980 nm (a), 808 nm (b) and 1550 nm (c) excitations, respectively.



**Figure S15.** TEM images of the NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub>:Yb(60 mol%) coreshell nanoparticles with different Yb shell thicknesses. NaYF<sub>4</sub> inert shell was subsequently coated. The scale bar is 100 nm.



**Figure S16.** Mid-infrared emission spectra of NaErF<sub>4</sub>:Ho(10 mol%)@NaYF<sub>4</sub>:Yb(60 mol%)@NaYF<sub>4</sub> core-shell-shell nanoparticles with various interlayer shell thicknesses under 980 nm excitation.



**Figure S17.** Mid-infrared emission spectra of NaErF<sub>4</sub>:Ho/Yb(10/10 mol%)@NaYF<sub>4</sub>:Yb(60 mol%)@NaYF<sub>4</sub>:Nd/Yb(30/20 mol%) core-shell-shell nanocrystals after modifying with different concentrations of ICG dyes. The excitation wavelength is 808 nm.