

Electronic Supporting Information (ESI)
**Control of Green and Red Upconversion in NaYF₄:Yb³⁺,Er³⁺ Nanoparticles by
Excitation Modulation**

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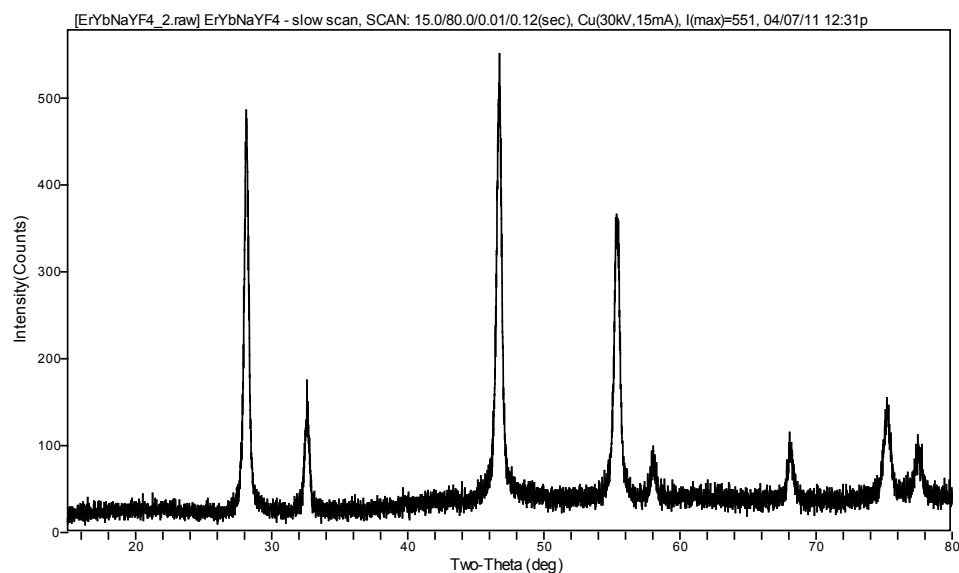


Fig. S1. X-ray powder diffraction (XRD) pattern of NaYF₄:Er³⁺,Yb³⁺ nanoparticles.

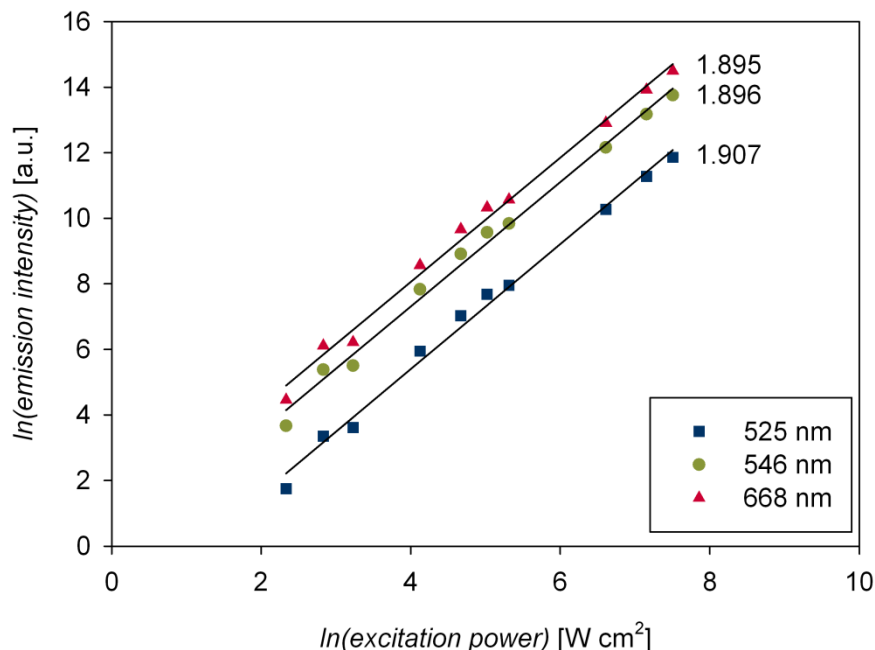


Fig. S2. Power density dependence of three emission peaks in NaYF₄: Yb³⁺, Er³⁺ nanoparticles. Data for visible peaks are well matched to linear fits with slopes of approximately 2, indicating 2-photon processes. Excitation was by 980 nm laser diode pulsed at 500 Hz with a 100 μs pulse width.

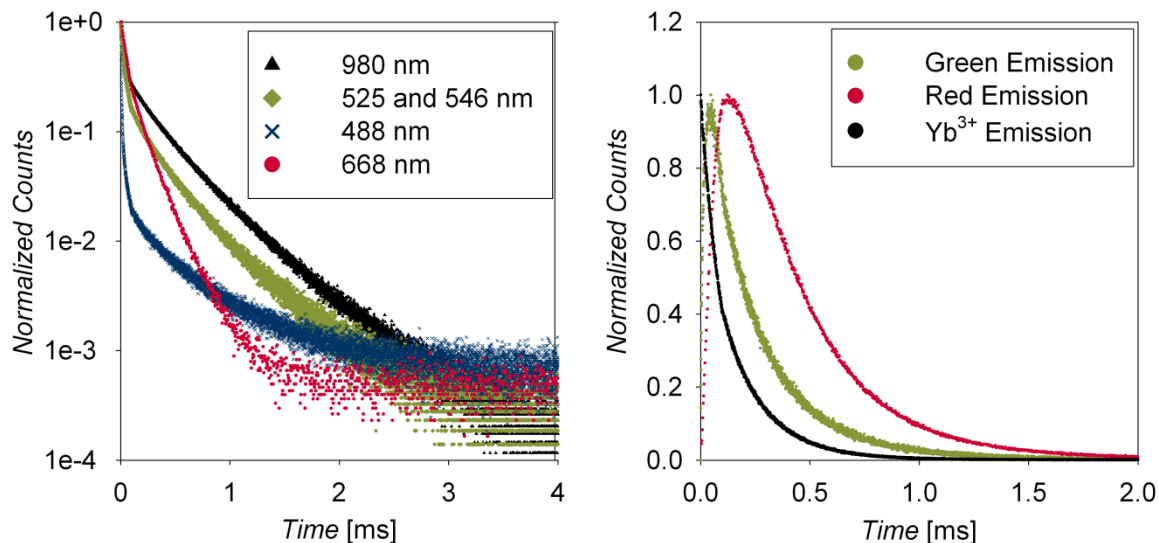


Fig. S3. Left: Luminescence decay of NaYF₄:Er³⁺, Yb³⁺ nanoparticles at several wavelengths following direct excitation of the associated transitions. Excitation and measurement matched the wavelengths listed in the legend. Right: Luminescence decay of transitions resulting in green and red emission in upconverting nanoparticles as well as the luminescence decay of NaYF₄:Yb³⁺ nanoparticles at 980 nm. Excitation for all three was 980 nm. Excitation was by an OPO tuned NdYAG laser pulsed at 20 Hz with 3 ns pulse width for all measurements.

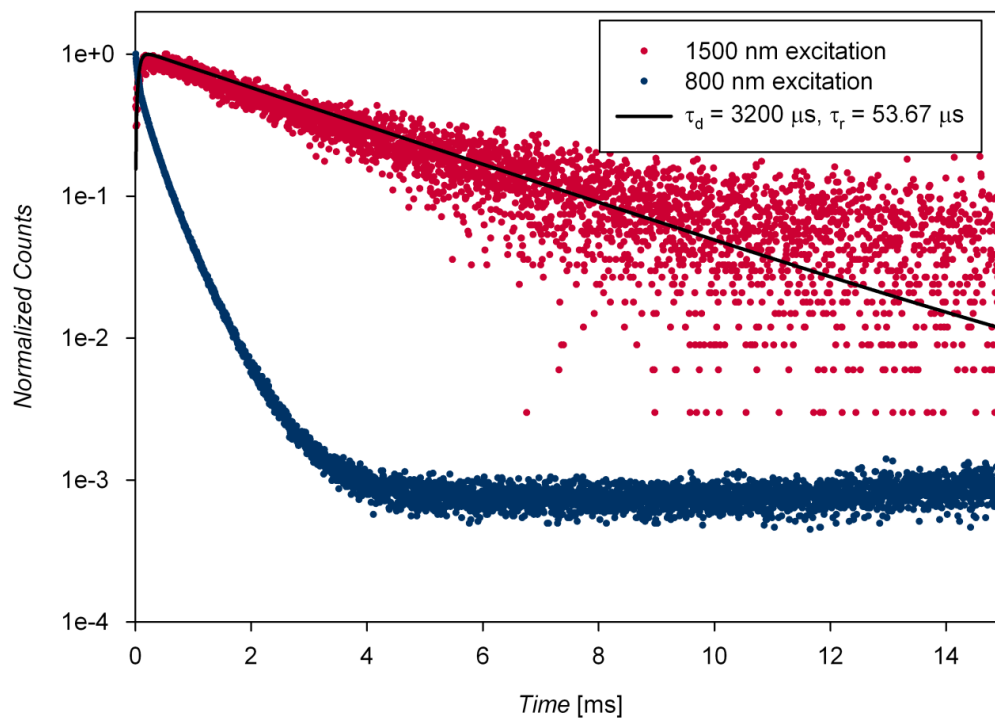


Fig. S4. Luminescence decay measured at 980 nm following excitation by 1500 nm (red) and 800 nm (blue).

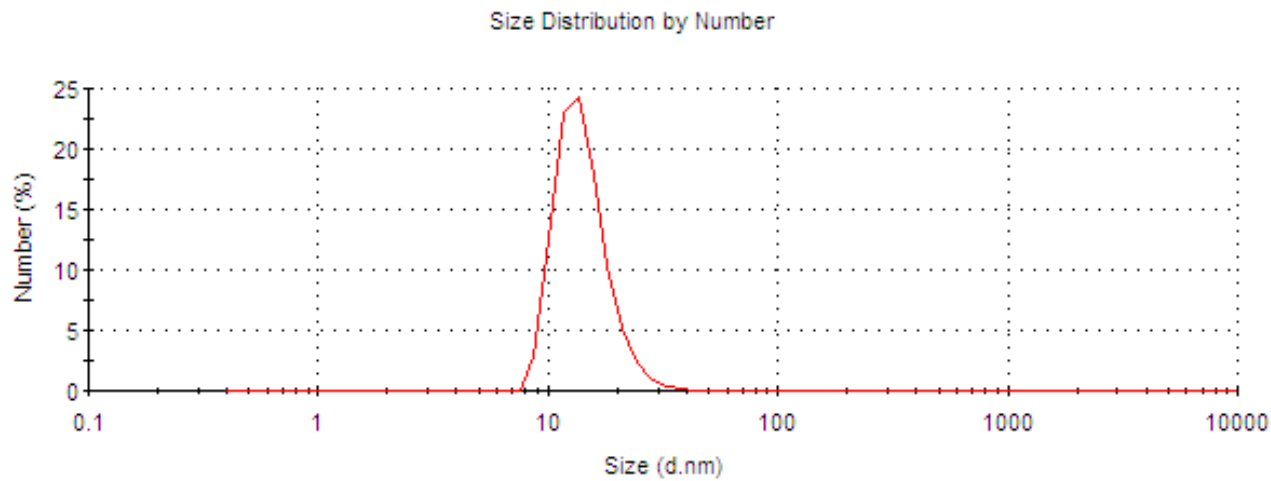


Fig. S5. Size distribution of $\text{NaYF}_4:\text{Er}^{3+}, \text{Yb}^{3+}$ nanoparticles. The peak is at 14.3 nm.

Coefficients for rate equations presented in the main text:

$$C_{22} = \frac{k_{02}N_0N_{S^*}^0}{W_2 - W_S} \text{ and } C_{21} = N_2^0 - \frac{k_{02}N_0N_{S^*}^0}{W_2 - W_S}$$

$$C_{42} = \frac{k_{24}N_{S^*}^0N_2^0}{W_4 - (W_S + W_2)} - \frac{k_{02}k_{24}N_0N_{S^*}^0{}^2}{W_4(W_2 - W_S) - (W_S^2 + W_2^2)}$$

$$C_{43} = \frac{k_{02}k_{24}N_0N_{S^*}^0{}^2}{W_4(W_2 - W_S) - 2(W_2W_S - W_S^2)}, \text{ and } C_{41} = -(C_{42} + C_{43})$$

$$C_{31} = -(C_{32} + C_{33} + C_{34} + C_{35}),$$

$$C_{32} = \frac{k_{13}N_{S^*}^0C_{11}}{W_3 - W_S - W_{10}}, C_{33} = \frac{\left(W_{43}C_{42} - \frac{k_{13}N_{S^*}^0C_{21}}{W_2}\right)}{W_3 - W_S - W_2}$$

$$C_{34} = \frac{\left(W_{43}C_{43} - \frac{k_{13}N_{S^*}^0C_{22}}{W_S}\right)}{W_3 - 2W_S}, \text{ and } C_{35} = \frac{W_{43}C_{41}}{W_3 - W_4}$$