## **Supporting Information**

## Highly Spectral Dependent Enhancement of Upconversion Emission with Sputtered Gold Island Films

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## Synthesis of NaYF4:Yb/Er nanocrystals

NaYF<sub>4</sub>:20%Yb,2%Er NCs was synthesized by thermal decomposition of rare-earth/sodium trifluoroacetate precursors in oleic acid (OA) and octadecene (ODE) as reported previously.<sup>9</sup> All chemicals were purchased from Sigma-Aldrich and used without further purification. In a typical procedure, 0.975 mmol of yttrium(III) oxide ( $Y_2O_3$ , 99.99%), 0.25 mmol of ytterbium(III) oxide ( $Yb_2O_3$ , 99.9%) and 0.025 mmol of erbium(III) oxide ( $Er_2O_3$ , 99.9%) were dissolved in 5 ml trifluoroacetic acid (TFA, 99%) in a 100 ml three-necked flask. The slurry was then heated to 80 °C with vigorous magnetic stirring under vacuum for 30 minutes to remove water and excessive TFA. Next, 2.5 mmol sodium trifluoroacetate (NaCOOCF<sub>3</sub>, 98%) was added, along with 7.5 mL of oleic acid (OA, 90%) and 7.5 ml of 1-octadecene (ODE, 90%) at 100 °C. Afterwards, the solution was heated to 330 °C at a rate of 30 °C min<sup>-1</sup> and maintained at 330 °C for 60 minutes to obtain the NCs. The NCs were thoroughly washed and can be readily dispersed in non-polar organic solvents such as chloroform and toluene.



*Figure S1.* (a) TEM image of upconversion NCs. (Scale Bar: 200 nm) (b) EDX spectrum of upconversion NCs shows only sodium, fluorine and rare-earth elements. The carbon signal comes from the carbon membrane on TEM grid. The atomic ratio of Y:Yb:Er is 72.3:25.0:2.7



*Figure S2.* (a) AFM image of a glass slide sputtered with gold island film; (b) SEM image of a single upconversion NC with sputtered gold island films. (Scale Bar: 100 nm)



*Figure S3.* SEM image of a large area of upconversion NC thin film prepared by Langmuir–Blodgett (LB) assembly. (Scale Bar:  $10 \mu m$ )



*Figure S4.* Emission spectra obtained from 10 different locations in L-B assembled upconversion NC film before (a) and after (b) sputtering GIFs.

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*Figure S5.* Schematic illustration of the two-photon (a) and three-photon (b) upconversion process in NaYF<sub>4</sub>:Yb,Er NCs.



Figure S6. UV-Vis spectrum of the gold island film sputtered on a glass slide.