

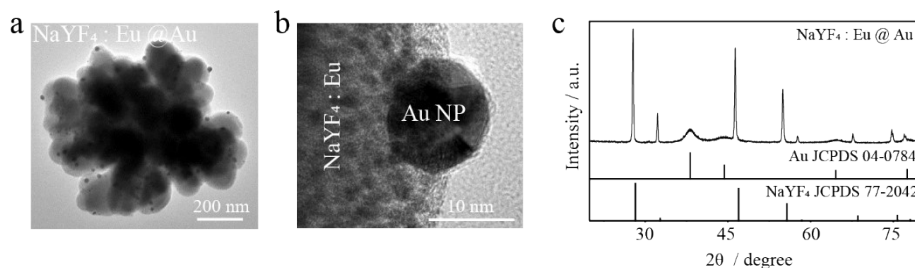
## Supplementary information

### Thermo-plasmonic assisted structure optimization of micro/nano crystal based on single-particle spectroscopy

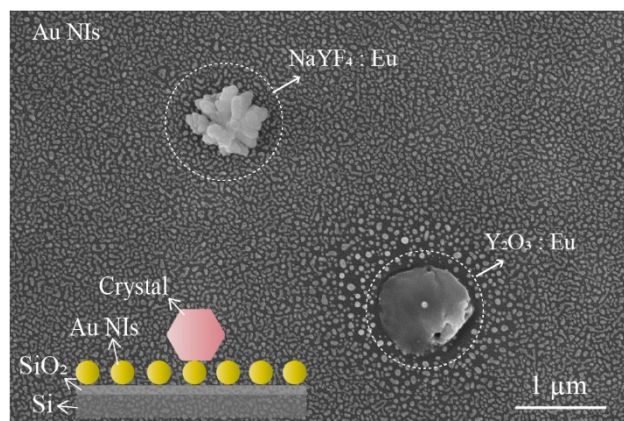
Chengyun Zhang,<sup>a,b</sup> Xilin Zhou,<sup>b</sup> Ting Kong,<sup>a</sup> Lei Xi,<sup>b</sup> Ruobin Zhang,<sup>a</sup> Baobao Zhang,<sup>b</sup> Huan Chen,<sup>b</sup>  
Zhengkun Fu,<sup>b</sup> Zhenglong Zhang<sup>\*,b</sup>

<sup>a</sup> Address here School of Electronic Engineering, Xi'an University of Posts & Telecommunications, Xi'an 710121,  
China

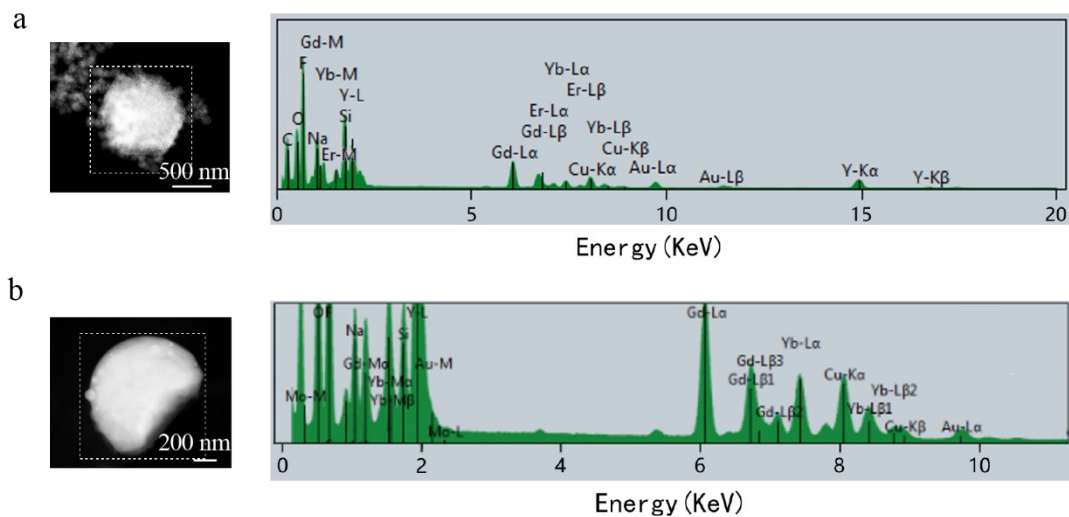
<sup>b</sup> School of Physics and Information Technology, Shaanxi Normal University, Xi'an 710062, China,  
[zizhang@snnu.edu.cn](mailto:zizhang@snnu.edu.cn)



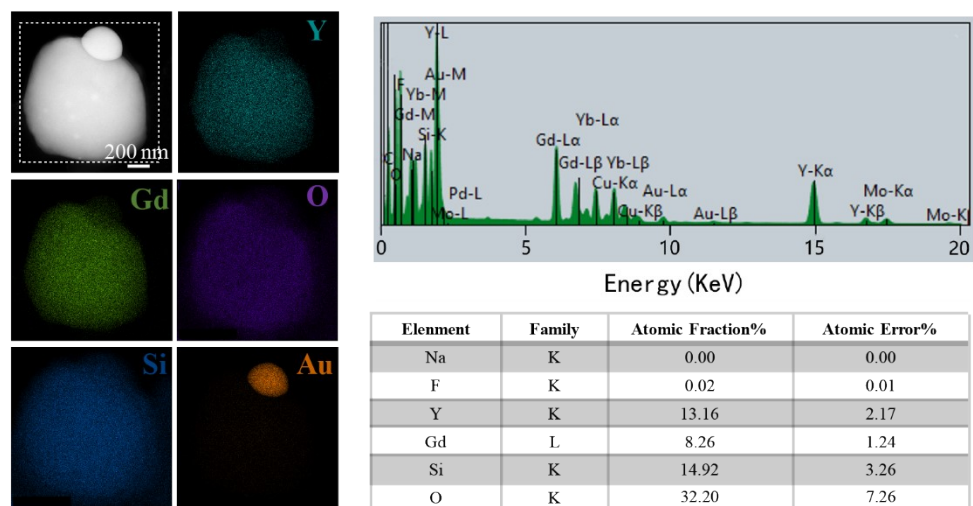
**Figure S1. Characterization of the NaYF<sub>4</sub>:Eu<sup>3+</sup> @Au composite particles.** (a) TEM image of a single composite particle. (b) Enlarged TEM image of the composite particle, showing an Au NP partially embedded in the crystal. (c) X-ray diffraction spectra of the composite structure and the standard patterns of cubic phase Au (JCPDS No.04-0784) and cubic phase NaYF<sub>4</sub> (JCPDS No.77-2042).



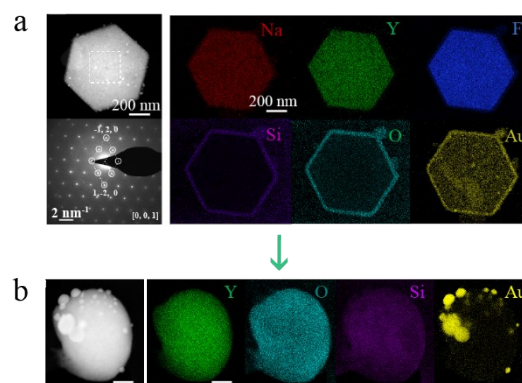
**Figure S2. SEM image of the Au nanoislands with NaYF<sub>4</sub>:Eu<sup>3+</sup> and transformed and sketch of the composite structure**



**Figure S3.** Elemental characterization of the as-synthesized NaYF<sub>4</sub> (Hexagonal phase) @ SiO<sub>2</sub> (Amorphous) @ NaGdF<sub>4</sub> (Amorphous) @ Au (cubic phase) composite structure (a) before and (b) after light irradiation.

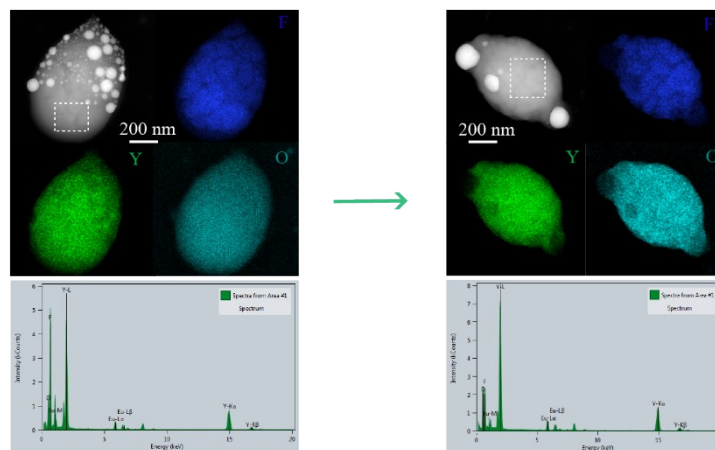


**Figure S4.** Structural and elemental characterization of the particle shown in Figure 5 after further irradiation (5 mW, 0.5 s), which results in the disappear of fluorine and sodium elements in the particle.

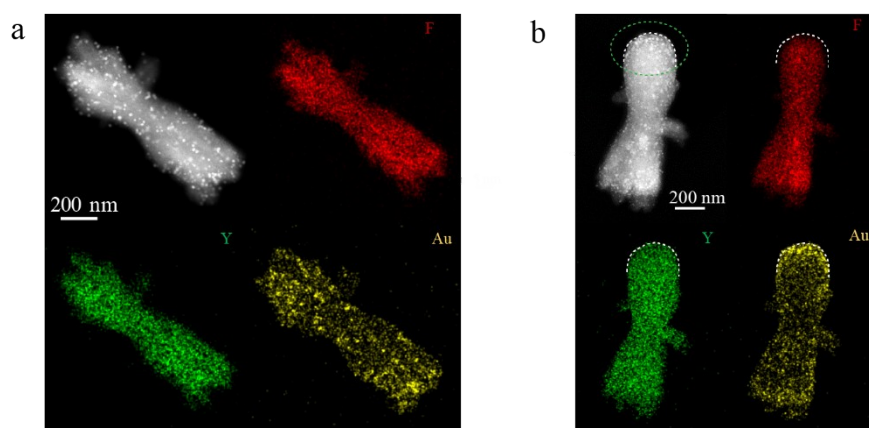


**Figure S5.** Thermo-plasmonic assisted modulation of elemental distribution. Structural and elemental characterization of the as-synthesized NaYF<sub>4</sub> (Hexagonal phase) @ SiO<sub>2</sub> (Amorphous) @ Au (cubic phase)

composite structure (a) before and (b) after light irradiation. SAED pattern taken along [0 0 1] zone axis from the circled region of the NaYF<sub>4</sub> particle with hexagonal structure. After light irradiation (5 mW, 1 s), the Si elements in the shell layer are uniformly dispersed into the crystal particles, while the fluorine and sodium elements in the particles disappear.



**Figure S6. Thermo-plasmonic assisted modulation of elements.** Slight irradiation of light brings about a decrease of fluorine element in fluorine oxide and an increase of oxygen element.



**Figure S7. Thermo-plasmonic assisted regulation of element distribution in microregions.** (a) STEM image and EDX maps of an as-synthesized YF<sub>3</sub>@Au composite particle. It can be seen that both ends of the particles are in the shape of a broom. (b) STEM image and EDX maps of a YF<sub>3</sub>@Au composite particle with one tip of the particle slightly irradiated by light (1 mW, 0.1 s). The area irradiated by light is marked with a green dotted circle. The white dashed line represents the outline of this tip after being irradiated by light. And it can be seen that the irradiated endpoint shows obvious deformation from irregular broom shape to rounded shape, and the fluorine element is obviously