Supporting Information

## Dynamic tailorable local luminescence patterns on single

## upconversion fluoride microcrystals via in situ oxidation through

## laser irradiation

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**Figure S1** Specific single-particle confocal experimental devices (a), schematic of excitation approach for the spot-excitation approach (b) and the waveguiding-excitation approach (c).



**Figure S2** Simplified energy level diagrams of  $Yb^{3+}$  and  $Ho^{3+}$  ions and proposed upconversion mechanisms for NaYbF<sub>4</sub>: Ho microtube.



**Figure S3** EDX spectra of NaYbF<sub>4</sub>:Ho microtube before and after laser irradiation by a femtosecond laser with a power density of 15 mJ/cm<sup>2</sup> per pulse. Therein, (a) primitive NaYbF<sub>4</sub>:Ho microtube, (b) NaYbF<sub>4</sub>:Ho microtube after laser irradiation.



Fig. S4 XRD pattern of NaYbF<sub>4</sub>:2% Ho<sup>3+</sup> microtube before and after irradiation by laser with a power density of 15 mJ/cm<sup>2</sup> per pulse. The bottom panels are standard data for hexagonal NaYF<sub>4</sub> (JCPDS file number 27-1427) and hexagonal YbOF (JCPDS file number 19-1433).



Fig. S5 The normalized luminescence decay curves for  $NaYbF_4$ :Ho microrods and microtubes with the similar size under a 980 nm laser excitation. The corresponding monitoring wavelength is marked in the map. The longer luminescent lifetime of  $NaYbF_4$ :Ho microrods indicates the higher crystal quality.



Fig. S6 The red-to-green ratio of upconversion luminescence intensities excited by a femtosecond laser of 980 nm wavelength with different pulsed power densities.