## **Supplementary Material (ESI)**

## Post annealing induced manipulation of phase and upconversion luminescence of Cr<sup>3+</sup> doped NaYF<sub>4</sub>:Yb,Er crystals<sup>†</sup>

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Fig.S1. Fiber coupled excitation-collection laser (980 nm) based experimental setup.



Fig.S2. FESEM images of the NaYF:Yb<sup>3+</sup>/Er<sup>3+</sup> crystals codoped with different amount of  $Cr^{3+}$  ions annealed at 600 °C : (a) 0 mol%, (b) 5 mol%, (c)10 mol%, (d)15 mol%, (e) 20 mol%.



Fig.S3. Summarized pseudo phase diagram (T vs mol% Cr)

Fig.S3 Show the summarized pseudo phase diagram obtained from the XRD data. Here  $\alpha$  refers to the pure cubic phase,  $\beta$  refers to pure hexagonal phase, ( $\alpha$ +  $\beta$ ) and ( $\beta$ + $\alpha$ ) refers to cubic phase dominant mixed phase and hexagonal phase dominant mixed phase respectively.



Fig.S4. Emission spectra of the NaYF<sub>4</sub>:Yb<sup>3+</sup>/Er<sup>3+</sup> crystals codoped with different amount of Cr<sup>3+</sup> ions annealed at different temperature: (a) 5 mol%, (b) 10 mol%, (c) 15 mol%, (d) 20 mol%.



Fig.S5. Laser power dependence emission spectra of NaYF<sub>4</sub>:Yb<sup>3+</sup>/Er<sup>3+</sup> crystals annealed at different temperature: (a) 200°C, (b) 400°C, (C) 600°C.



Fig.S6. Laser power dependence emission spectra of 5 mol% Cr<sup>3+</sup> doped NaYF<sub>4</sub>:Yb<sup>3+</sup>/Er<sup>3+</sup> microcrystals annealed at different temperature: (a) 200°C, (b) 400°C, (C) 600°C.



Fig.S7. Laser power dependence emission spectra of 10 mol% Cr<sup>3+</sup> doped NaYF<sub>4</sub>:Yb<sup>3+</sup>/Er<sup>3+</sup> crystals annealed at different temperature: (a) 200°C, (b) 400°C, (C) 600°C.



Fig.S8. Laser power dependence emission spectra of 15 mol% Cr<sup>3+</sup> doped NaYF<sub>4</sub>:Yb<sup>3+</sup>/Er<sup>3+</sup> crystals annealed at different temperature: (a) 200°C, (b) 400°C, (C) 600°C



Fig.S9. Laser power dependence emission spectra of 20 mol% Cr<sup>3+</sup> doped NaYF<sub>4</sub>:Yb<sup>3+</sup>/Er<sup>3+</sup> crystals annealed at different temperature: (a) 200°C, (b) 400°C, (C) 600°C.