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## **Supporting Information**

## **6**-NaYF₄:Yb,Tm:upconversion properties by controlling the transition probabilities at the same energy level

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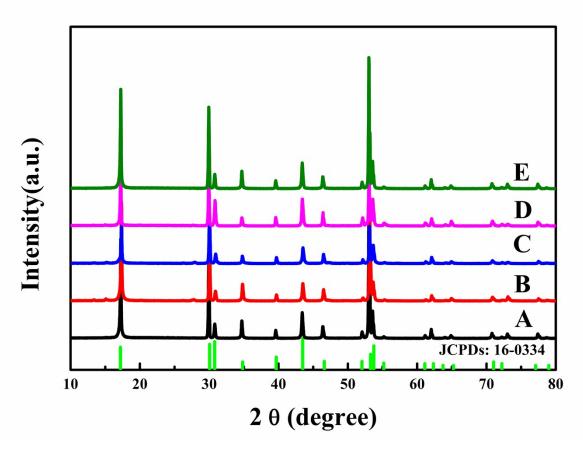
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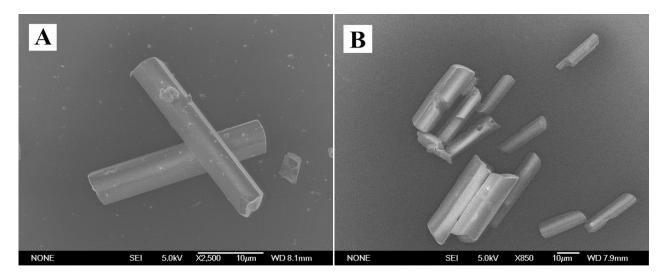
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**Table S5.** The emission intensities table of  $\beta$ -NaYF<sub>4</sub>: 10%Yb, 5%Mn, 0.5%Tm prepared with different Pump power at different wavelengths, The integrated intensities of two photon transition(**T**) and The intensities of the overall integrated emission (four photon transition group + three photon transition group + two photon transition group)(**A**).

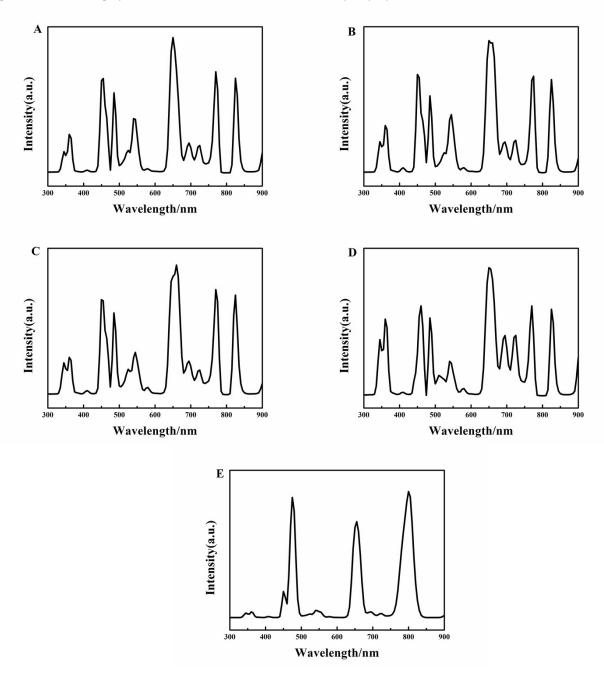


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Figure S2. SEM micrograph of the  $\theta$ -NaYF<sub>4</sub>: 10%Yb<sup>3+</sup>, 0.5%Tm<sup>3+</sup> samples prepared at 180°C with reaction time 24 h



**Figure S3.** Room temperature UC emission spectra of (A) the  $\theta$ -NaYF<sub>4</sub>: 20%Yb<sup>3+</sup>, 0.5%Tm<sup>3+</sup>. (B) the  $\theta$ -NaYF<sub>4</sub>: 30%Yb<sup>3+</sup>, 0.5%Tm<sup>3+</sup>. (C) the  $\theta$ -NaYF<sub>4</sub>: 50%Yb<sup>3+</sup>, 0.5%Tm<sup>3+</sup>. (D) the  $\theta$ -NaYF<sub>4</sub>: 70%Yb<sup>3+</sup>, 0.5%Tm<sup>3+</sup>. (E) the  $\theta$ -NaYF<sub>4</sub>: 99.5%Yb<sup>3+</sup>, 0.5%Tm<sup>3+</sup>. The samples were tested under laser excitation at 980nm with the pump power of 250mW.

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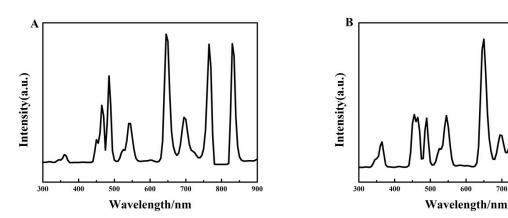


Figure S4. Room temperature UC emission spectra of (A) the  $\theta$ -NaYF<sub>4</sub>: 10%Yb<sup>3+</sup>, 1.0%Mn<sup>2+</sup>, 0.5%Tm<sup>3+</sup>. (B) the  $\theta$ -NaYF<sub>4</sub>: 10%Yb<sup>3+</sup>, 2.0%Mn<sup>2+</sup>, 0.5% Tm<sup>3+</sup>. The samples were tested under laser excitation at 980nm with the pump power of 250mW.

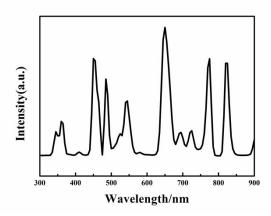
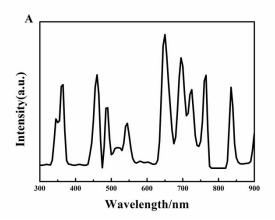
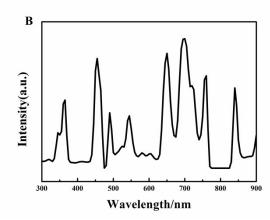


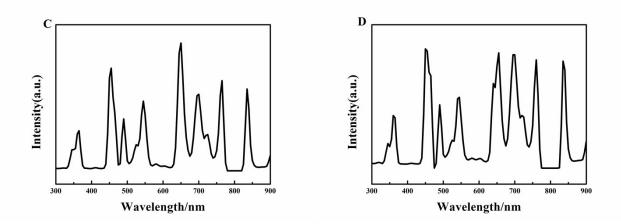
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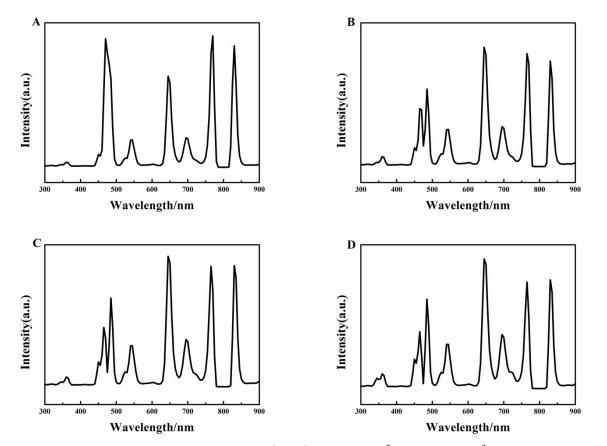


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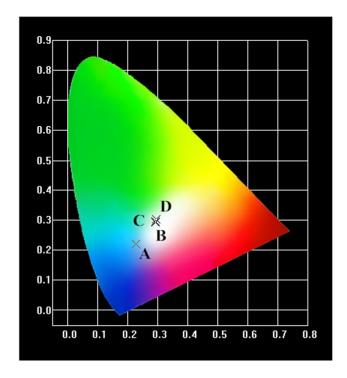


**Figure S6.** Room temperature UC emission spectra of (A) the  $\theta$ -NaYF<sub>4</sub>: 10%Yb<sup>3+</sup>, 1%Tm<sup>3+</sup>. (B) the  $\theta$ -NaYF<sub>4</sub>: 10%Yb<sup>3+</sup>, 2%Tm<sup>3+</sup>. (C) the  $\theta$ -NaYF<sub>4</sub>: 10%Yb<sup>3+</sup>, 4%Tm<sup>3+</sup>. (D) the  $\theta$ -NaYF<sub>4</sub>: 10%Yb<sup>3+</sup>, 8%Tm<sup>3+</sup>. The samples were tested under laser excitation at 980nm with the pump power of 250mW.



**Figure S7.** Room temperature UC emission spectra of the  $\theta$ -NaYF<sub>4</sub>: 10%Yb<sup>3+</sup>,1.0%Mn, 1%Tm<sup>3+</sup> under laser excitation at 980nm with different pump power: (A) 150mW, (B) 200mW, (C) 250mW, (D) 300mW.

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**Figure S8.** CIE chromaticity diagram of the  $\theta$ -NaYF<sub>4</sub>: 10%Yb, 1.0%Mn, 0.5%Tm samples prepared under laser excitation at 980nm with different Pump power: (A) 150mW, (B) 200mW, (C) 250mW, (D) 300mW.

**Table S1.** The emission intensities table of  $\theta$ -NaYF<sub>4</sub>: xYb, 0.5%Tm prepared with different concentration of Yb<sup>3+</sup> at different wavelengths, The integrated intensities of two photon transition(**T**) and The intensities of the overall integrated emission (four photon transition group + three photon transition group + two photon transition group)(**A**). The pump power is 250mW at 980nm.

content	350nm	450nm	475nm	550nm	650nm	Т	Α
5%	1.21091E6	3.10826E6	4.03861E7	2.85854E7	4.66132E7	1.03327E8	2.18174E8
10%	1.28849E7	2.96692E7	1.40751E7	1.93289E7	5.46488E7	7.51999E7	2.05807E8
20%	1.35290E7	3.54517E7	1.73052E7	2.45435E7	6.64177E7	7.53523E7	2.32599E8
30%	1.91423E7	3.52734E7	1.67406E7	2.34426E7	7.81401E7	7.38938E7	2.46633E8
50%	1.41098E7	3.76859E7	1.91857E7	2.76701E7	8.19835E7	8.88300E7	2.69465E8
70%	3.77945E7	3.17367E7	1.82998E7	1.56425E7	7.65863E7	9.76660E7	2.77726E8
99.5%	1.81898E6	5.78565E6	4.58349E7	3.6801E6	5.14672E7	9.31085E7	2.01695E8

**Table S2.** The emission intensities table of  $\beta$ -NaYF<sub>4</sub>: 10%Yb, xMn, 0.5%Tm prepared with different concentration of Mn<sup>2+</sup> at different wavelengths, The integrated intensities of two photon transition(**T**) and The intensities of the overall integrated emission (four photon transition group + three photon transition group + two photon transition group)(**A**). The pump power is 250mW at 980nm.

content	350nm	450nm	475nm	550nm	650nm	T	Α
0.5%	1.40010E6	1.2133E7	1.53714E7	3.31835E7	3.33888E7	5.45448E7	1.62624E8
1.0%	2.03494E6	1.15387E7	1.61338E7	1.46687E7	3.99755E7	6.36254E7	1.47977E8
2.0%	7.03617E6	2.39349E7	1.08266E7	2.01444E7	4.80903E7	6.93742E7	1.79407E8
8.0%	1.05587E7	3.33183E7	1.16215E7	1.8233E7	4.55734E7	7.57524E7	1.95057E8

**Table S3.** The emission intensities table of β-NaYF<sub>4</sub>: 10%Yb, 0.5%Tm prepared with different reaction times at different wavelengths, The integrated intensities of two photon transition(**T**) and The intensities of the overall integrated emission (four photon transition group + three photon transition group + two photon transition group)(**A**). The pump power is 250mW at 980nm.

time	350nm	450nm	475nm	550nm	650nm	T	Α
0.5h	0	10854	713708	95125	192311	1.37776E6	2.38976E6
6.0h	6.33139E6	7.91809E6	4.14534E6	4.92504E6	1.36817E7	2.09786E7	5.79801E7
12h	6.49636E6	7.81754E6	4.12552E6	5.09903E6	1.06504E7	2.19474E7	5.61363E7
24h	1.28849E7	2.96692E7	1.40751E7	1.93289E7	5.46488E7	7.51999E7	2.05807E8

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**Table S4.** The emission intensities table of β-NaYF<sub>4</sub>: 10%Yb, xTm prepared with different concentration of  $Tm^{3+}$  at different wavelengths, The integrated intensities of two photon transition(**T**) and The intensities of the overall integrated emission (four photon transition group + three photon transition group + two photon transition group)(**A**). The pump power is 250mW at 980nm.

Content	350nm	450nm	475nm	550nm	650nm	T	Α
0.5%	1.28849E7	2.96692E7	1.40751E7	1.93289E7	5.46488E7	7.51999E7	2.05807E8
1%	1.63974E7	3.98101E7	1.15991E7	1.9056E7	6.21167E7	1.12183E8	2.61162E8
2%	2.16587E7	4.02696E7	1.03391E7	2.00332E7	4.49127E7	1.44560E8	2.81773E8
4%	1.07098E7	3.7887E7	1.06998E7	2.478E7	4.38053E7	8.41078E7	2.11989E8
8%	1.2157E7	3.99085E7	1.0426E7	2.33243E7	4.61303E7	1.03228E8	2.35174E8

**Table S5.** The emission intensities table of  $\beta$ -NaYF<sub>4</sub>: 10%Yb, 1.0%Mn, 0.5%Tm prepared with different Pump power at different wavelengths, The integrated intensities of two photon transition(**T**) and The intensities of the overall integrated emission (four photon transition group + three photon transition group + two photon transition group)(**A**).

Pump power	350nm	450nm	475nm	550nm	650nm	T	Α
150mW	596902.79885	1.55618E6	3.81773E7	6.67475E6	1.98513E7	5.50617E7	1.21918E8
200mW	1.16481E6	1.08227E7	1.27096E7	1.06087E7	3.19507E7	6.05922E7	1.27849E8
250mW	1.42033E6	1.1243E7	1.30177E7	1.06972E7	3.12463E7	6.02035E7	1.29660E8
300mW	2.74021E6	1.17677E7	1.47312E7	1.27151E7	3.61397E7	6.38436E7	1.41938E8