

## Supporting Information

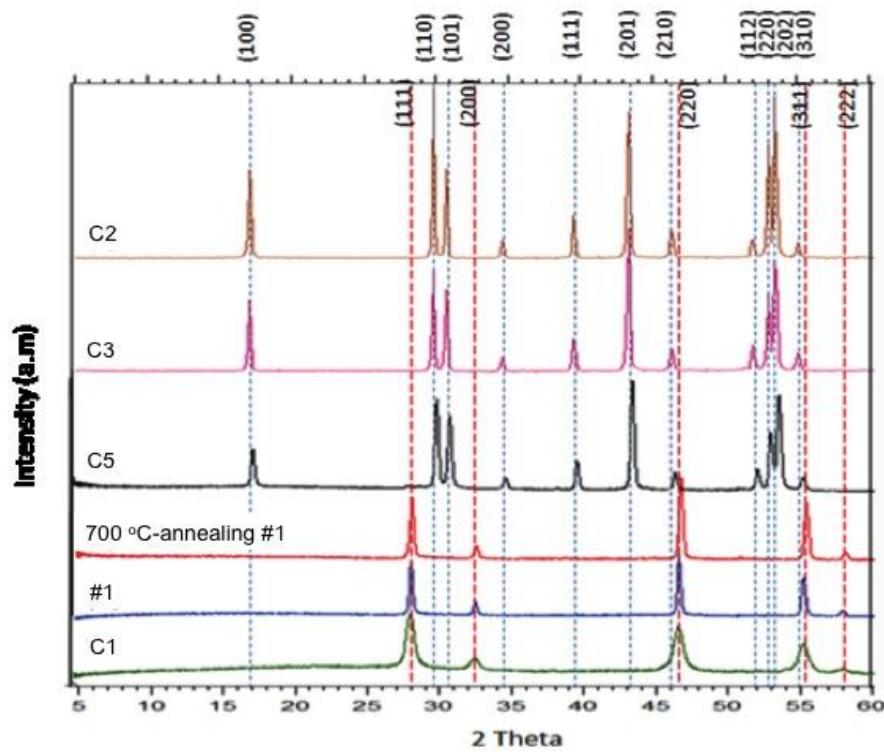
### Annealing-induced ultra-efficient NIR-to-VIS upconversion of nano-/micro-scale $\alpha$ and $\beta$ NaYF<sub>4</sub>:Er<sup>3+</sup>,Yb<sup>3+</sup> crystals

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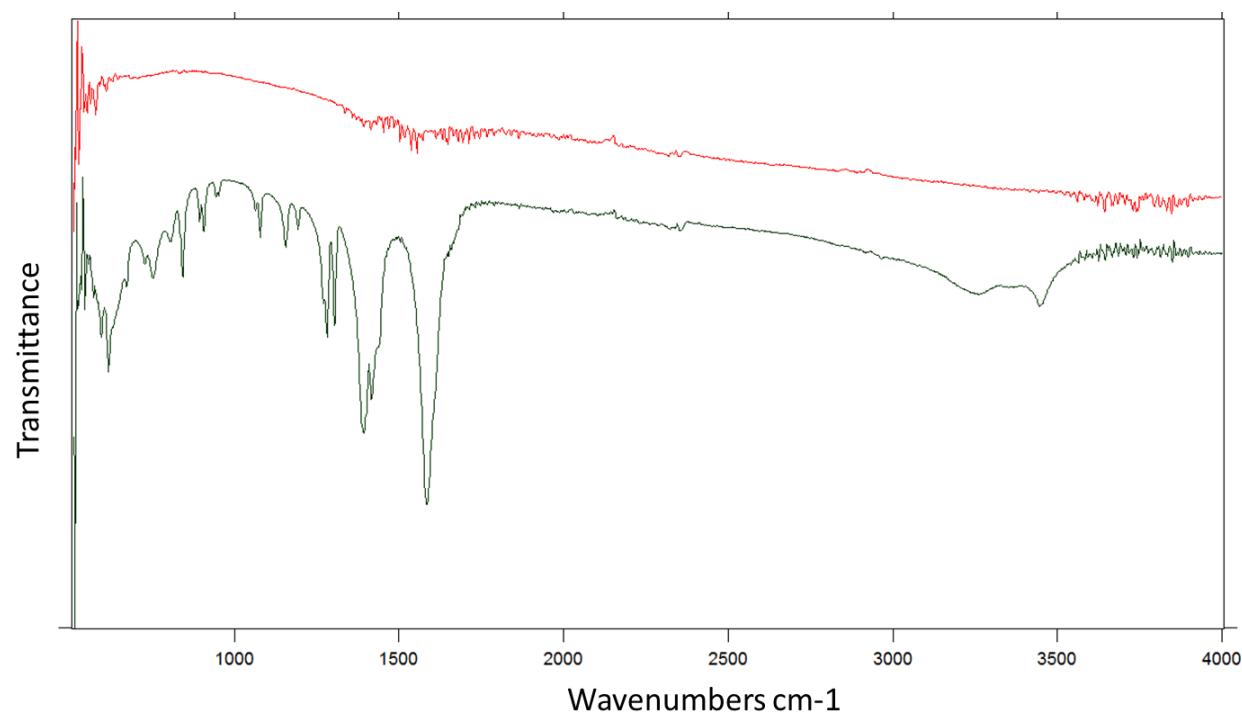
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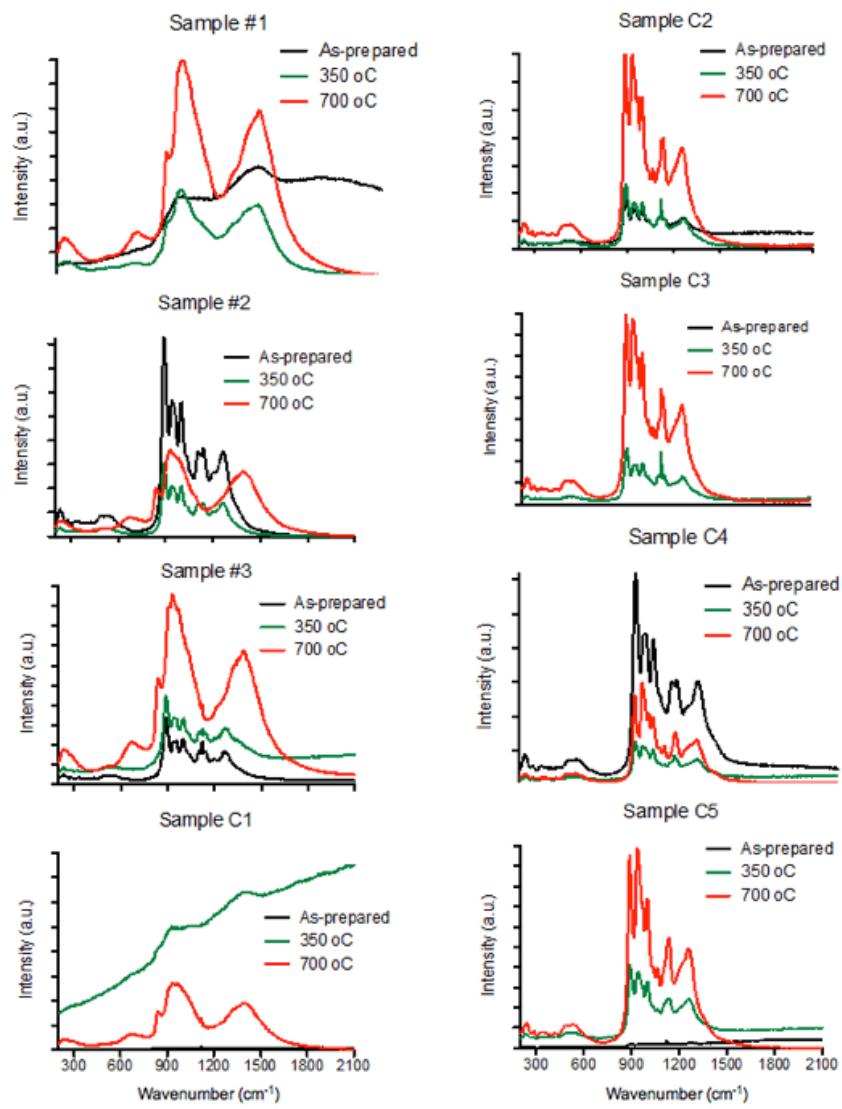
<b>Figure S1.</b> XRD patterns of $\alpha$ -NaYF <sub>4</sub> :Er <sup>3+</sup> ,Yb <sup>3+</sup> (C1, green; #1, blue) and $\beta$ -NaYF <sub>4</sub> :Er <sup>3+</sup> ,Yb <sup>3+</sup> (C2, yellow; C3, pink; C5, black), and 700 °C-annealed $\alpha$ -NaYF <sub>4</sub> :Er <sup>3+</sup> ,Yb <sup>3+</sup> (#1, red) .....
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**Fig.S1** XRD patterns of  $\alpha$ -NaYF<sub>4</sub>:Er<sup>3+</sup>,Yb<sup>3+</sup> (C1, green; #1, blue) and  $\beta$ -NaYF<sub>4</sub>:Er<sup>3+</sup>,Yb<sup>3+</sup> (C2, yellow; C3, pink; C5, black), and 700 °C-annealed  $\alpha$ -NaYF<sub>4</sub>:Er<sup>3+</sup>,Yb<sup>3+</sup> (#1, red)



**Fig.S2.** FTIR spectra of citrate-modified  $\text{NaYF}_4:\text{Er}^{3+},\text{Yb}^{3+}$  crystals (sample C5) and its annealed product at 700°C.

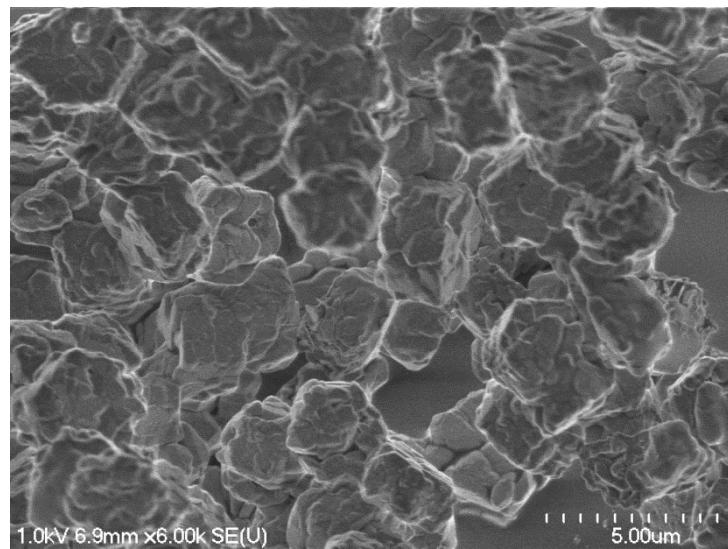


**Fig.S3** Raman spectra of various  $\text{NaYF}_4:\text{Er}^{3+},\text{Yb}^{3+}$  crystals before (as-prepared, black) and after annealing at 350 °C (green) and 700 °C (red)

**Table S1.** Raman spectra data ( $\text{cm}^{-1}$ ) of doped  $\alpha$ - and  $\beta$ - $\text{NaYF}_4:\text{Er}^{3+},\text{Yb}^{3+}$  crystals

#1			#2			#3			C1			C2			C3			C4			C5						
Amb.	350°	700°	Amb.	350°	700°	Amb	350°	700°	Amb	350°	700°	Amb	350°	700°	Amb	350°	700°	Amb	350°	700°	Amb	350°	700°				
$\alpha$	$\alpha$	$\alpha$	$\beta$	$\beta$	$\alpha$	$\beta$	$\beta$	$\alpha$	$\alpha$	$\alpha$	$\alpha$	$\beta$															
1416	1384	1390		1402		1392	1400	1431	1411			1265	1270	1265	1276	1262	1259	1273	1267	1267	1285	1267	1267				
			1274	1271		1273	1273								1240	1137		1228sh	1229					1240			
			1230	1232		1229						1148	1138	1138	1145	1124		1143	1141	1135		1138	1137				
			1144	1144		1144	1143					1122.6			1122			1138	1045		1122	1114	1038				
			1135	1132		1137							1041		1069	1119	1068		1065				1092				
														1030sh									1038	1038			
1003			1001	1003		1003	1004		979	979		1001.5	1003	1000	1000	1000	1000	1001	1001	1000	1001	1000	1000				
932	937		945	948	932	956	960	939				958.6	955	935	961	960	997	955	940	972	985	951	971				
		871	893	893	849	895	891	875				844	893	888	890	895	939	895	891	935	896	899	938				
687	701		552	575	699	555	580	703				714	550	572	555	565	565	888	548br	567	888	587w	560	888			
				516											512	516		533	548	506	359	506	559	521	558		
					362vw		357						350			367			315	314	436			363	311		
					313vw		314								291	291	288	288						292			
279	265		230vw	244	261	242	240	260				266	242	242	245	244	240	244	233	244	253	249	247	249			
179	170				163			170				175			126			126					128		150	147	

B: broad, Amb.: ambient, w: weak intensity, vw: very weak, sh: shoulder, br: broad band



**Fig. S4.** Typical SEM image of 700°C annealed  $\beta$ -NaYF<sub>4</sub>:Er<sup>3+</sup>,Yb<sup>3+</sup> crystals (sample C3). The annealed crystals were ground before their fluorescence was measured (Figure 5).