

## Supporting Information

### The role of Cr<sup>3+</sup> and Cr<sup>4+</sup> in emission brightness enhancement and sensitivity improvements of NIR-emitting Nd<sup>3+</sup>/Er<sup>3+</sup> ratiometric luminescence thermometers

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The integral intensities of the Nd<sup>3+</sup> and Er<sup>3+</sup> bands to calculate LIR values were fitted with Mott-Seitz equation (Eq. S1):

$$I = \frac{I_0}{C \cdot \exp\left(-\frac{W}{k \cdot T}\right) + 1} \quad (\text{Eq. S1})$$

where: I – the intensity in temperature T, I<sub>0</sub> – the intensity in the initial temperature, W - the activation energy, k – Boltzmann constant, C – the dimensionless constant

The average lifetime of the excited states were calculated with the equation Eq. S2:

$$\langle \tau \rangle = \frac{A_1 \tau_1^2 + A_2 \tau_2^2}{A_1 \tau_1 + A_2 \tau_2} \quad (\text{Eq. S2a})$$

where: τ<sub>1</sub>, τ<sub>2</sub> – the average time, which is in accordance with the relation τ = t·ln(2) and A<sub>1</sub>, A<sub>2</sub> – amplitude, which are the parameters of the doubleexponential function:

$$y = y_0 + A_1 \cdot \exp\left(-\frac{x}{t_1}\right) + A_2 \cdot \exp\left(-\frac{x}{t_2}\right) \quad (\text{Eq. S2b})$$

Temperature determination uncertainty was calculated using Eq. S6:

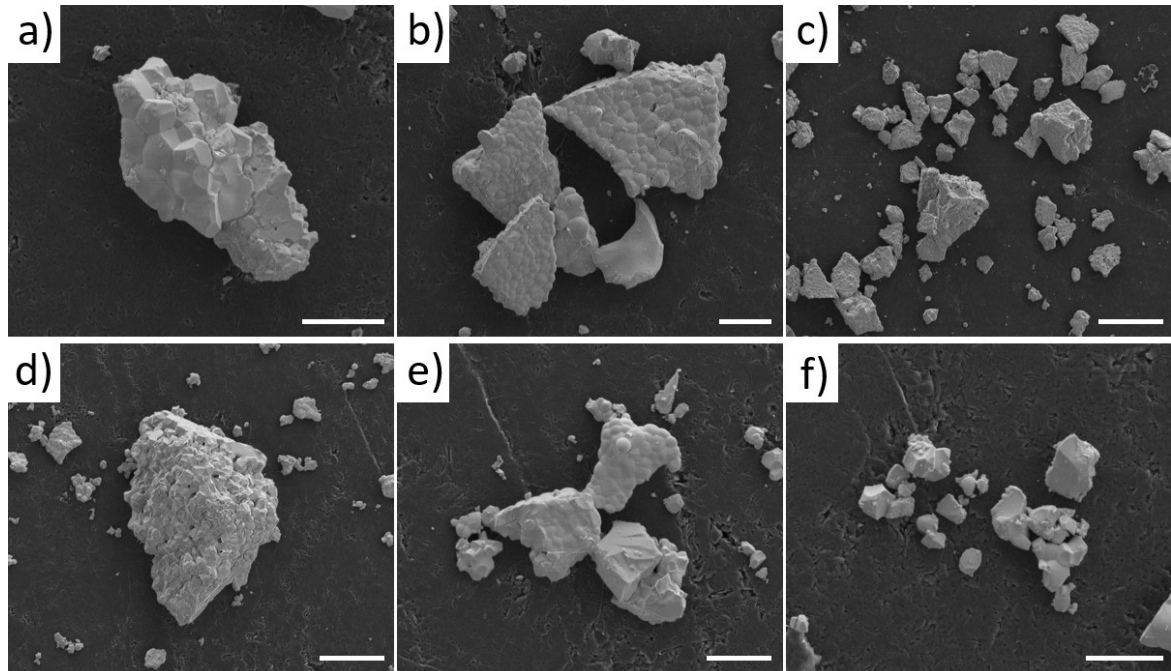
$$\delta T = \frac{1}{S_R} \cdot \frac{\delta LIR}{LIR} \quad (\text{Eq. S3a})$$

$S_R$  is the relative sensitivity and  $\delta LIR/LIR$  determines the uncertainty of the LIR determination, where  $\delta LIR/LIR$  was determined as follows:

$$\frac{\delta LIR}{LIR} = \sqrt{\left(\frac{\delta I_{Tb}}{I_{Tb}}\right)^2 + \left(\frac{\delta I_{Eu}}{I_{Eu}}\right)^2} \quad (\text{Eq. S3b})$$

Table S1. The Rietveld refinement parameters of XRD patterns for YAG:Nd<sup>3+</sup>, Er<sup>3+</sup>, Cr<sup>3+/4+</sup> powders

Sample	Rexp	Rprofile	weight R profile	D statistics	weight S-Statistics
YAG:1%Nd <sup>3+</sup> , 1%Er <sup>3+</sup>	1.52919	1.96616	3.1929	1.33710	1.07269
YAG:1%Nd <sup>3+</sup> , 1%Er <sup>3+</sup> , 1%Cr <sup>3+</sup>	1.55303	3.36102	4.73781	0.40631	0.25277
YAG:1%Nd <sup>3+</sup> , 1%Er <sup>3+</sup> , 2%Cr <sup>3+</sup>	1.53677	2.51937	3.52202	0.70730	0.49693
YAG:1%Nd <sup>3+</sup> , 1%Er <sup>3+</sup> , 5%Cr <sup>3+</sup>	1.52759	2.66131	3.75056	0.57502	0.39426
YAG:1%Nd <sup>3+</sup> , 1%Er <sup>3+</sup> , 10%Cr <sup>3+</sup>	1.55828	1.66312	2.39261	0.81267	0.73250



**Figure S1.** The representative SEM images of YAG:Nd<sup>3+</sup>, Er<sup>3+</sup> – scale bar: 4 μm (a), 5 μm (b), 20 μm (c) and YAG:Nd<sup>3+</sup>, Er<sup>3+</sup>, 10% Cr<sup>3+/4+</sup> – scale bar: 10 μm (d), 5 μm (e), 3 μm (f).

YAG:1%Nd <sup>3+</sup> , 1%Er <sup>3+</sup> , 20%Cr <sup>3+</sup>	1.56894	1.82007	2.74488	0.78064	0.69035
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Table S2. Atomic concentration of the surface of the analysed samples by EDS method.

YAG:1%Nd <sup>3+</sup> , 1%Er <sup>3+</sup> ,						
Element	Cr <sup>3+/4+</sup> -unco-doped			doped with 10% Cr <sup>3+/4+</sup>		
	at% (%)	+/-	<i>r<sub>conc</sub></i> (%)	at% (%)	+/-	<i>r<sub>conc</sub></i> (%)
Y	13.630	0.166		14.097	0.284	
Nd	0.180	0.022	1.321	0.187	0.045	1.324
Er	0.163	0.012	1.198	0.180	0.008	1.277
Al	25.877	0.337		24.773	0.198	
Cr	-	-		1.947	0.123	7.858
O	60.147	0.373		58.817	0.358	

*at%* - atomic percentage

*r<sub>conc</sub>* – the ratio of the dopant concentration in respect to the content of the element which it substitutes (Nd, Er substitutes Y sites, Cr substitutes Al sites)

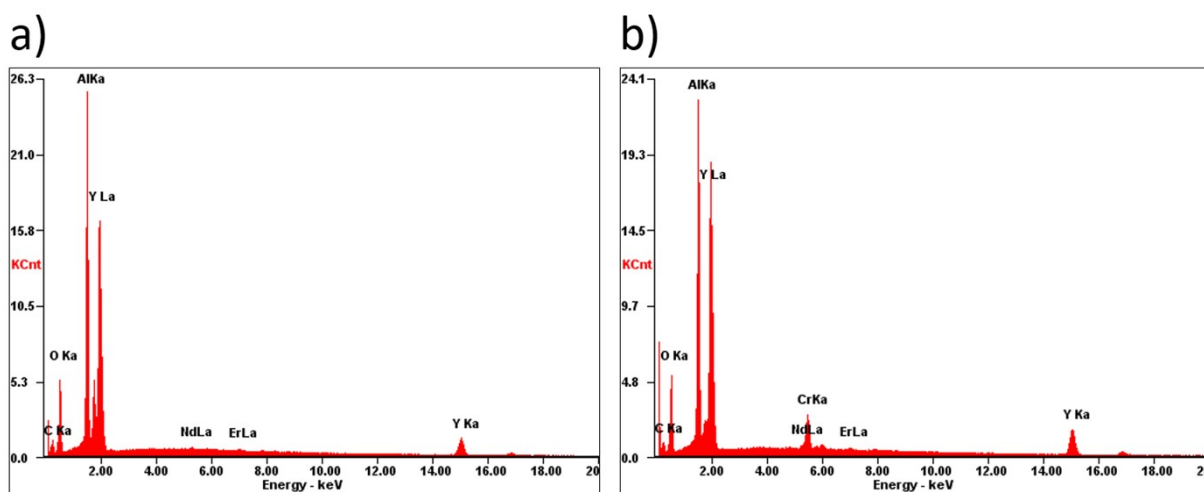


Figure S2. The representative EDS spectra for YAG:Nd<sup>3+</sup>, Er<sup>3+</sup> (a) and YAG:Nd<sup>3+</sup>, Er<sup>3+</sup>, 10% Cr<sup>3+/4+</sup> (b).

Table S3. Atomic concentration from the analyzed samples by XPS method.

The concentration of elements for the YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> samples (%):

Element	x = 1%	x = 5%	x = 10%	x = 20%
Al	11.74	12.97	10.4	7.25
Y	6.73	8.21	6.9	4.6
C	40.25	32.23	43.28	50
O	36.58	42.24	35.07	34.1
N	4.54	4.17	3.96	3.48
Cr	0	0.17	0.39	0.56

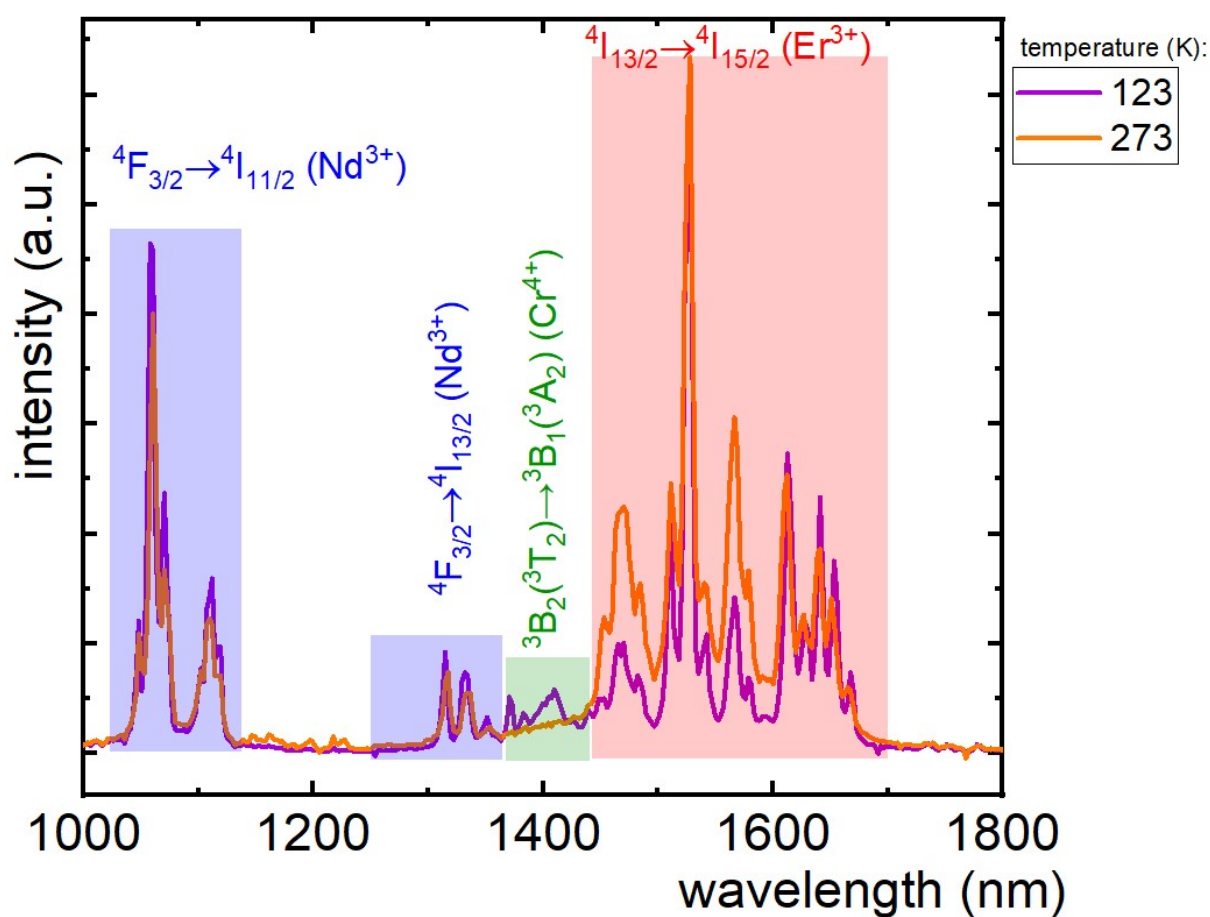
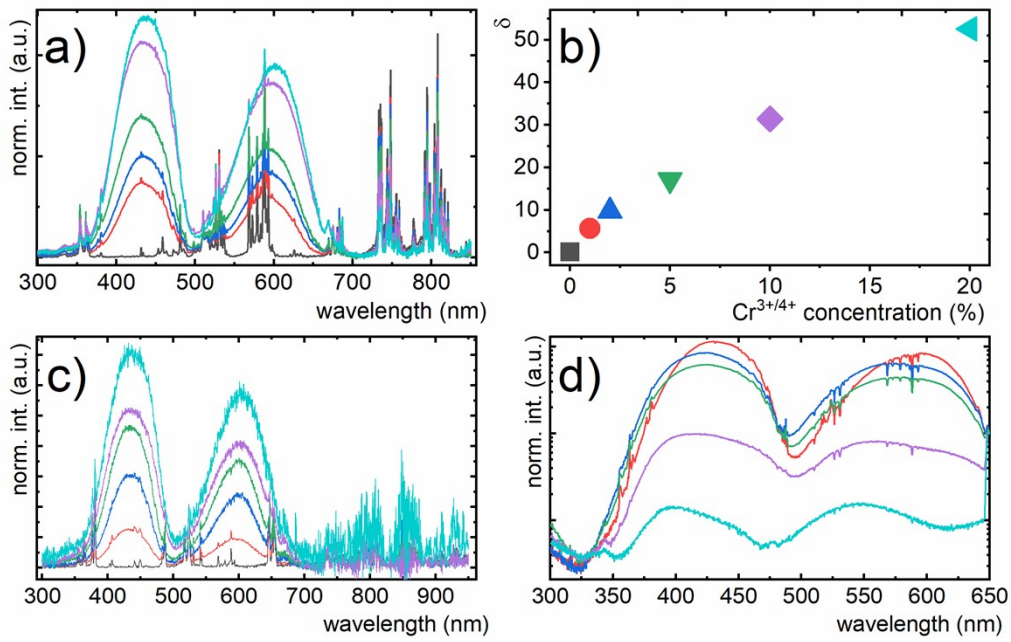
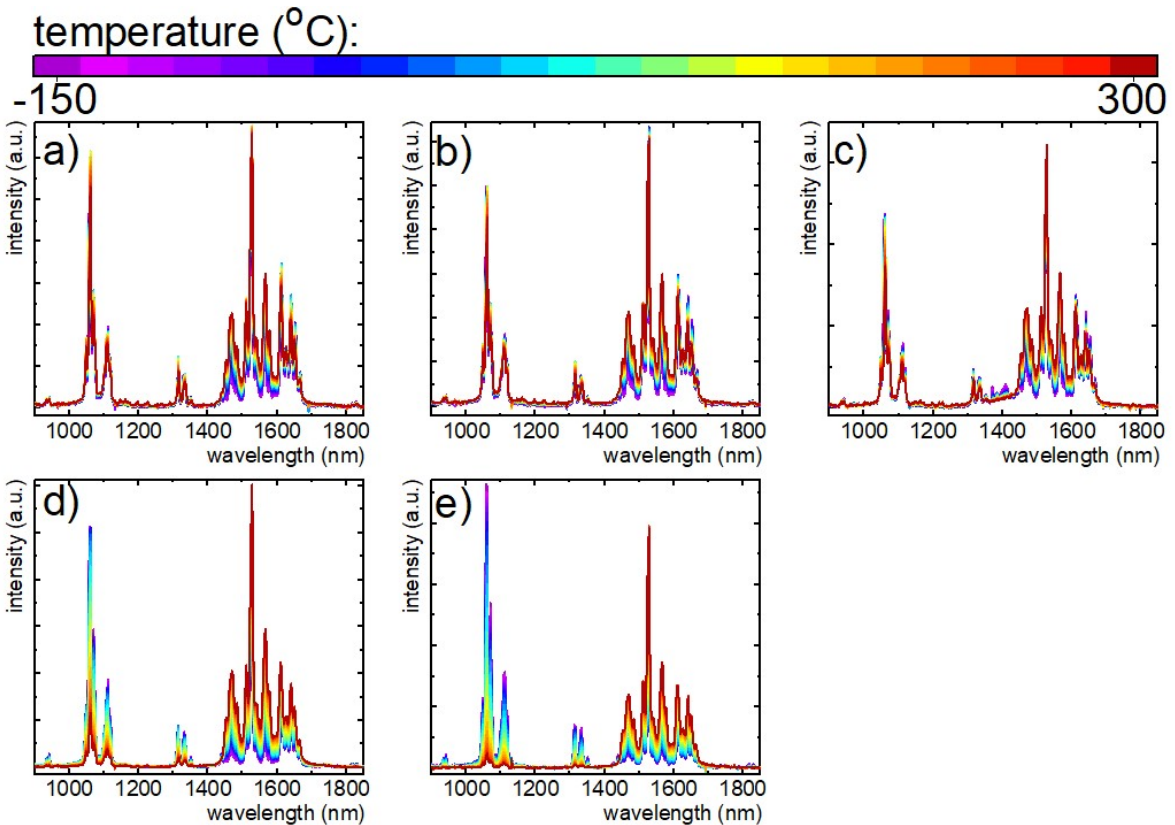


Figure S3. The emission spectra of YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, 5% Cr<sup>3+/4+</sup> at 123 K and at 273K .

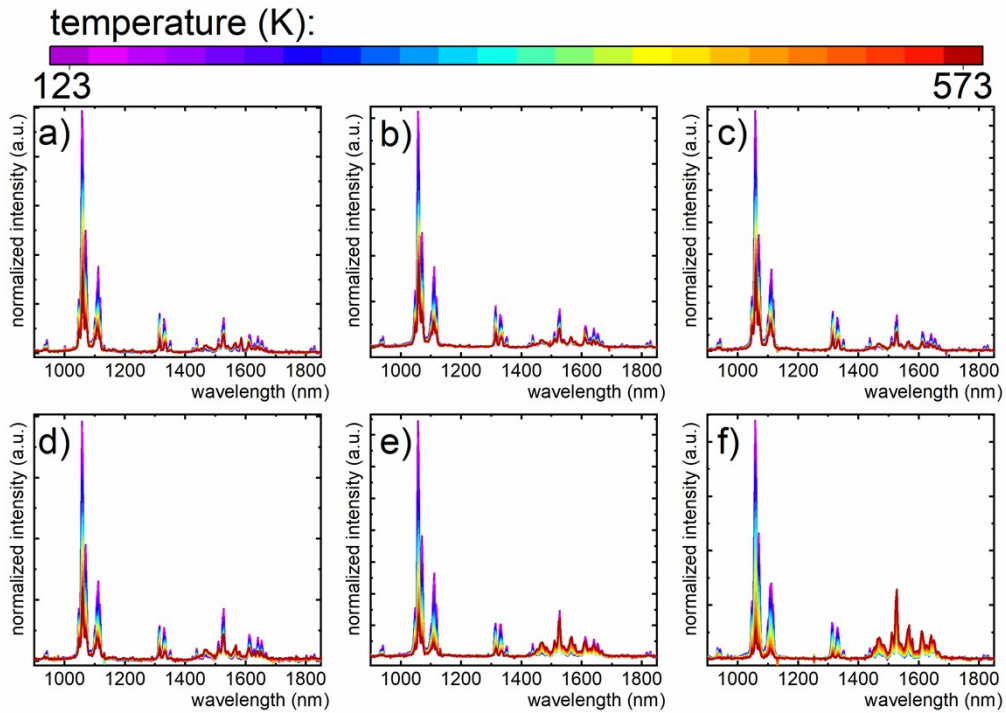
YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup>, x = 0 1 2 5 10 20



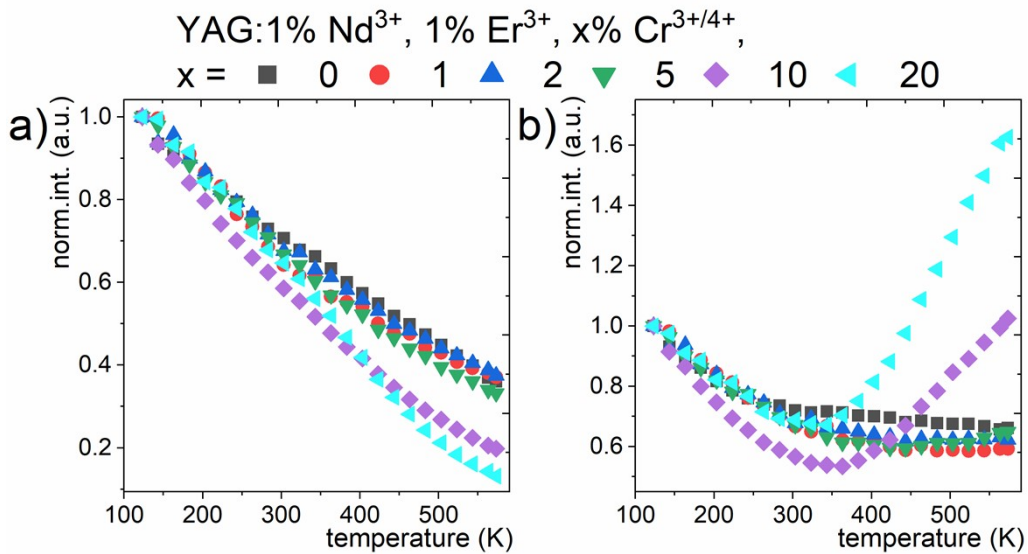
**Figure S4.** The influence of Cr<sup>3+/4+</sup> ions concentration on excitation spectra in YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> for 1064 nm (a) and on the contribution parameter  $\delta$  (b), for 1530 nm (c) and for 445 nm (d).



**Figure S5.** The thermal evolution of the NIR range of emission spectra of YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> powders with  $\lambda_{exc} = 445$  nm, where x = 1 (a), 2 (b), 5 (c), 10 (d), 20 (e).

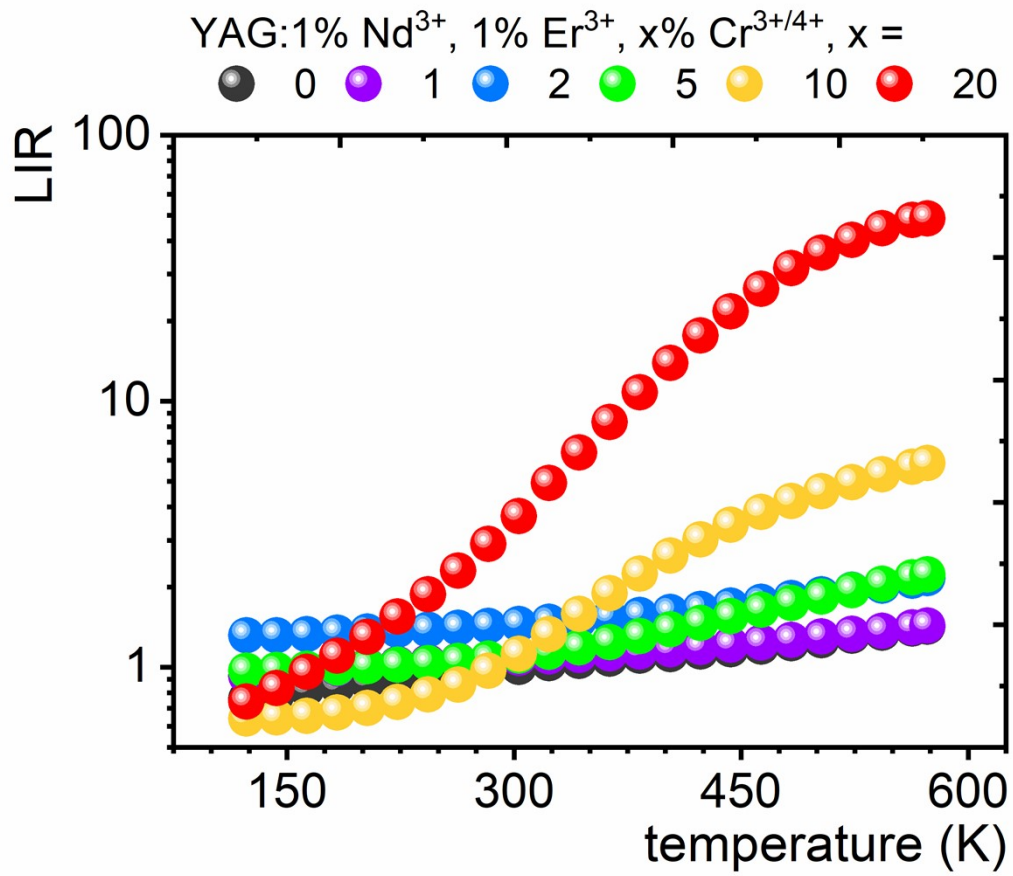


**Figure S6.** The thermal evolution of emission spectra of YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> powders with  $\lambda_{\text{exc}} = 793$  nm, where x = 0 (a), 1 (b), 2 (c), 5 (d), 10 (e), 20 (f).



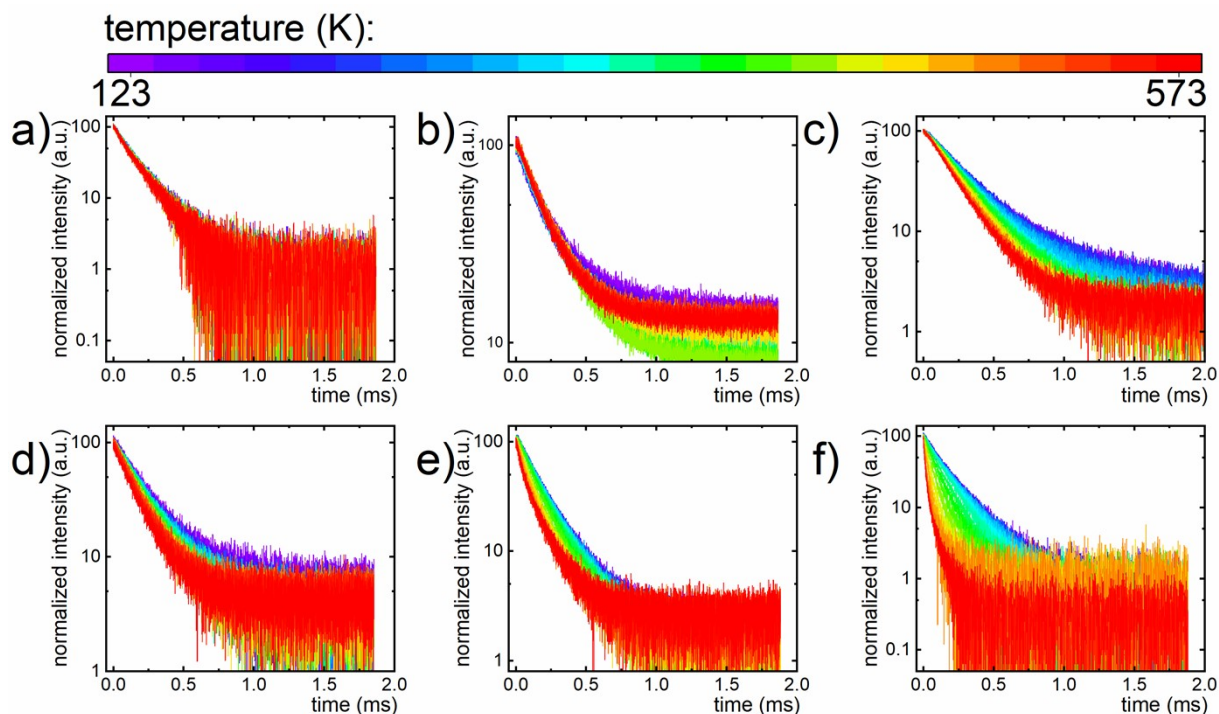
**Figure S7.** The thermal evolution of integral band intensities of  ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$  transition of Nd<sup>3+</sup> ion (a) and  ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$  electronic transition of Er<sup>3+</sup> ion (b) excited by  $\lambda_{\text{exc}} = 793$  nm.



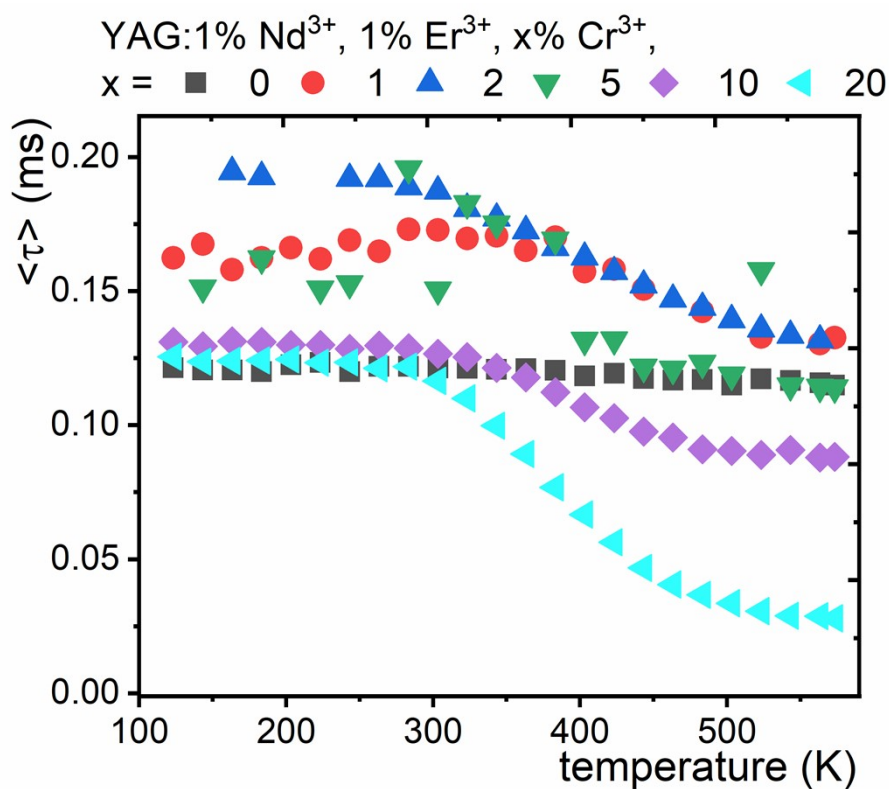


**Figure S8.** Thermal dependence of LIR calculated on the unnormalized data for different Cr<sup>3+</sup>/Cr<sup>4+</sup> dopant concentration

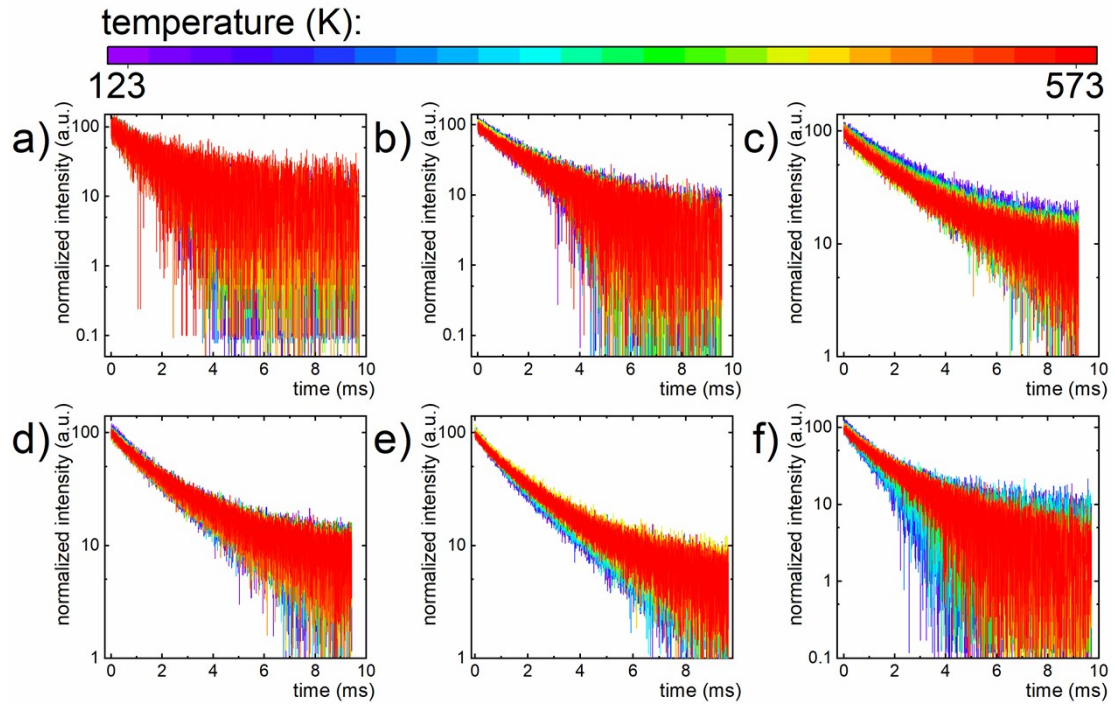




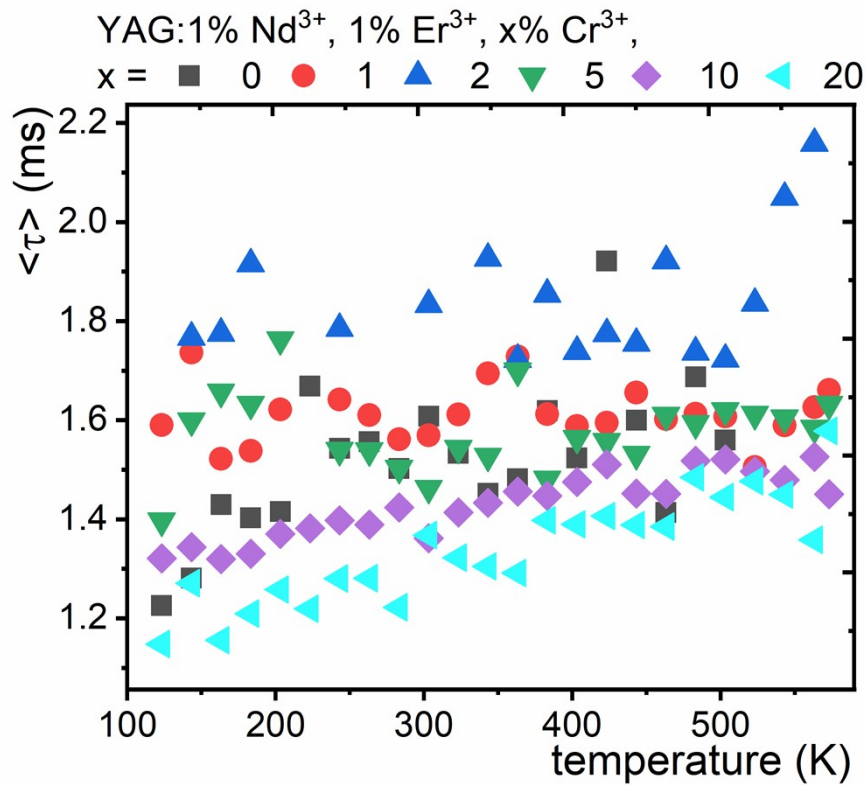
**Figure S9.** The thermal evolution of luminescent decays in YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> for 1064 nm (Nd<sup>3+</sup> excited state), where x = 0 (a), 1 (b), 2 (c), 5 (d), 10 (e), 20 (f).



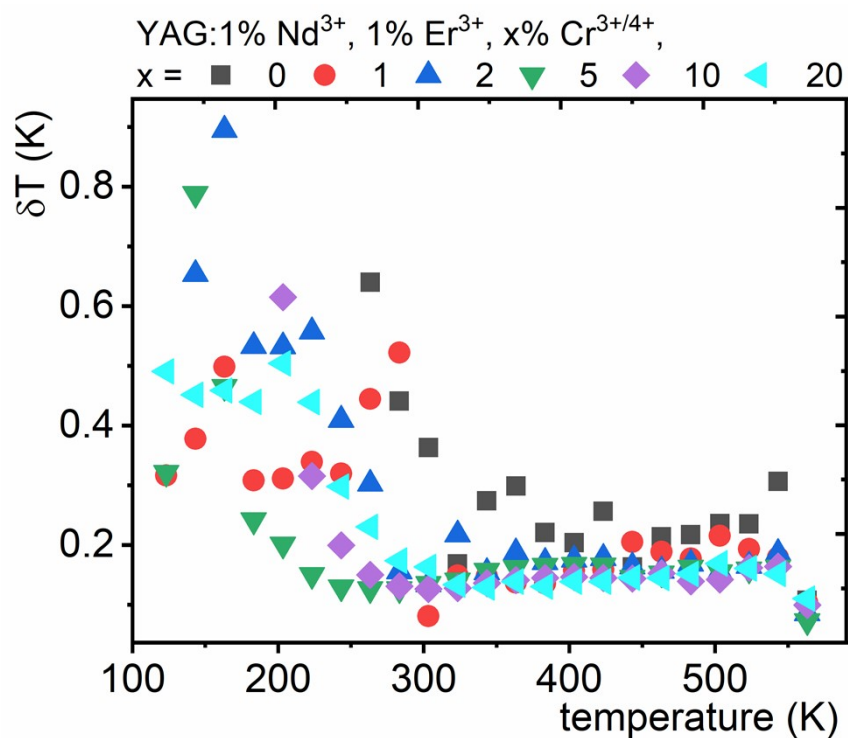
**Figure S10.** The thermal evolution of average lifetime of Nd<sup>3+</sup> excited state for YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> powders.



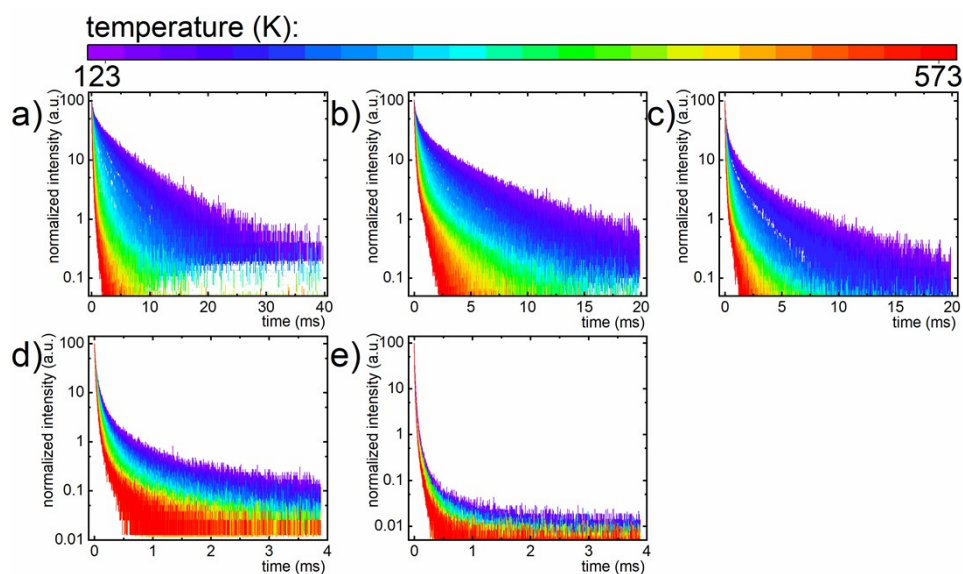
**Figure S11.** The thermal evolution of luminescent decays in YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> for 1530 nm (Er<sup>3+</sup> excited state), where x = 0 (a), 1 (b), 2 (c), 5 (d), 10 (e), 20 (f).



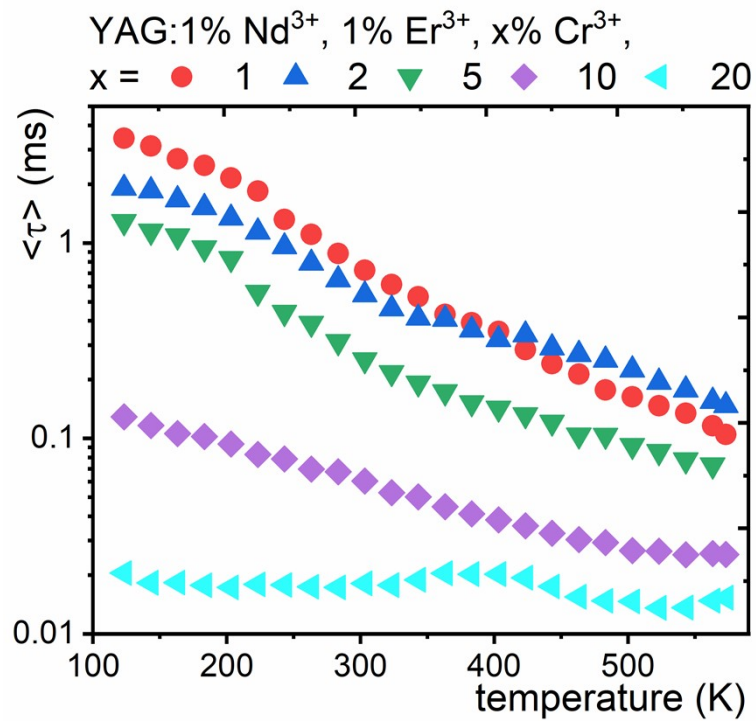
**Figure S12.** The thermal evolution of average lifetime of Er<sup>3+</sup> excited state for YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> powders.



**Figure S13.** The thermal dependence of temperature estimation uncertainty for different Cr<sup>3+/4+</sup> concentration in the YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup>.



**Figure S14.** The thermal evolution of luminescent decays in YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> for 445 nm (Cr<sup>3+</sup> excited state), where x = 1 (a), 2 (b), 5 (c), 10 (d), 20 (e).



**Figure S15.** The thermal evolution of average lifetime of Cr<sup>3+</sup> excited state for YAG:1% Nd<sup>3+</sup>, 1% Er<sup>3+</sup>, x% Cr<sup>3+/4+</sup> powders.