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Supporting Information for:

Dy^{3+} and Tb^{3+} codoped mixed garnet crystals with high-disorder structure for promising efficient InGaN laser-diode pumped yellow lasers

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Content

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1. Rietveld refinement result

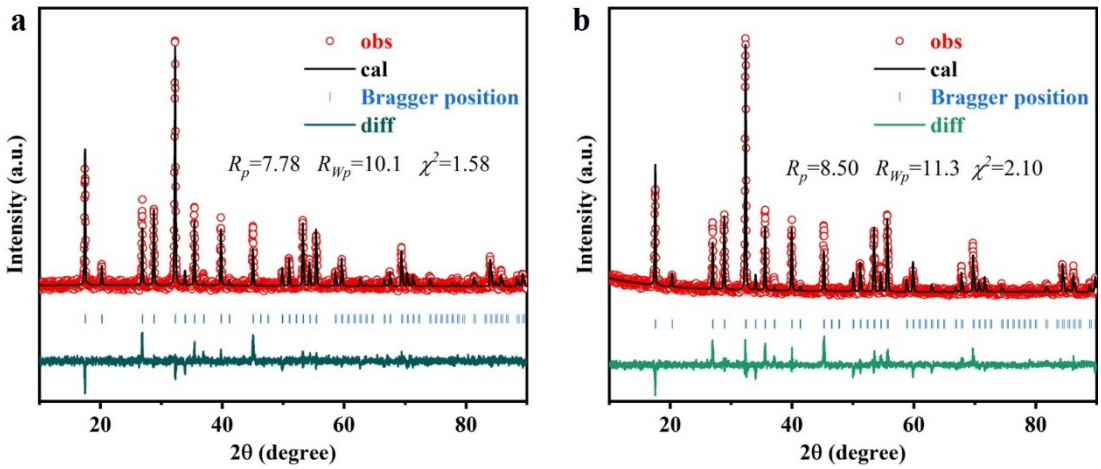

Fig.S1 Rietveld refinement XRD patterns of Dy,Tb:GYSAG(1) and Dy,Tb:GYSAG(2).

Table S1 The structural parameters obtained from the refinement of the Dy,Tb:GYSAG(1) crystal

Atoms	x	y	z	Occupancy	Wyckoff site
Gd	0.375	0.000	0.250	0.247	24d
Al (1)	0.000	0.000	0.000	0.024	16a
Al (2)	0.125	0.000	0.250	0.243	24c
Sc (1)	0.375	0.000	0.250	0.008	24d
Sc (2)	0.000	0.000	0.000	0.145	16a
O	0.0296	-0.043	0.154	1.000	96h
a=b=c=12.143 Å, α=β=γ=90°					

Table S2 The structural parameters obtained from the refinement of the Dy,Tb:GYSAG(2) crystal

Atoms	x	y	z	Occupancy	Wyckoff site
Gd	0.375	0.000	0.250	0.247	24d
Al (1)	0.000	0.000	0.000	0.024	16a
Al (2)	0.125	0.000	0.250	0.243	24c
Sc (1)	0.375	0.000	0.250	0.008	24d
Sc (2)	0.000	0.000	0.000	0.145	16a
O	0.0200	-0.047	0.159	1.000	96h
a=b=c=12.015 Å, α=β=γ=90°					

2. Density of the crystals

The density of the mixed garnet crystals was measured by Archimedes method. The schematic diagram of the measurement is shown Fig.S2. The ρ_0 in the formula is the density of the water at room temperature. The m_0, m_1 and m_2 represents the mass of dried sample in air, the mass of the water and the mass of the sample completely immersed in the water, respectively.

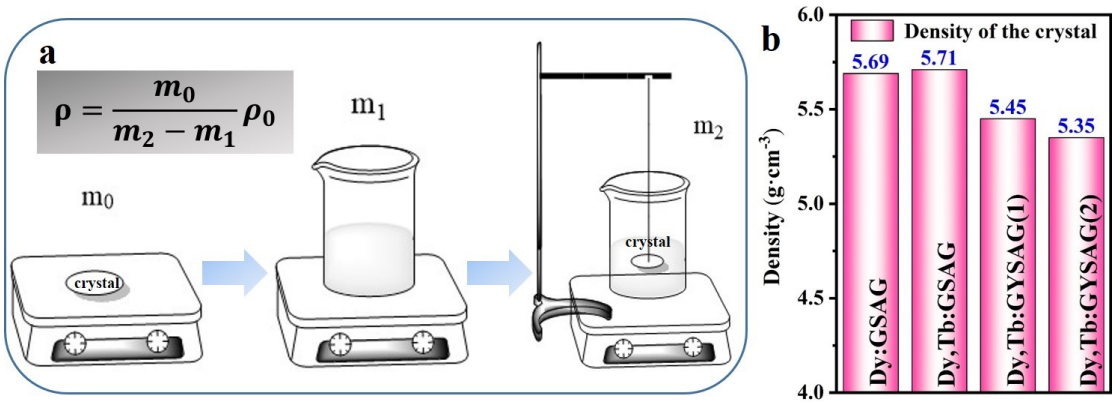

Fig.S2 (a) The schematic diagram of the crystal density measurement. (b) Measured crystal densities of the mixed garnet crystals.

3. Elements mapping and effective segregation coefficient of Dy^{3+}

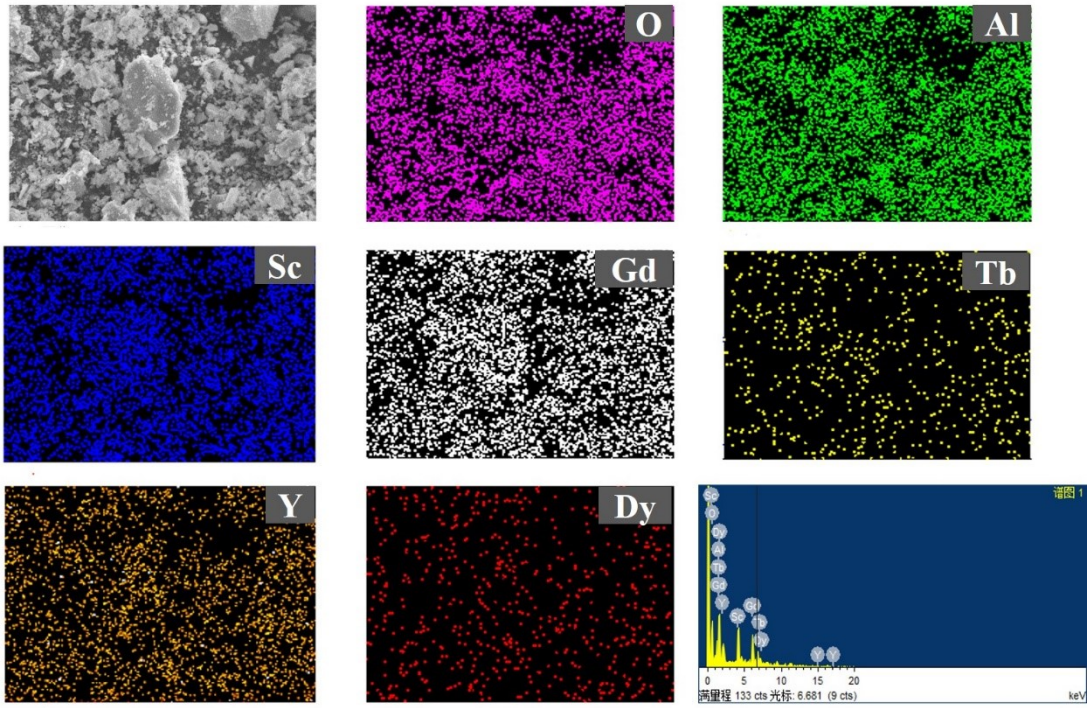

Fig.S3 Elements mapping and quantitative analysis of elements in the crystal by SEM-EDS measurement.

4. Judd-Ofelt (J-O) calculation

In this work, the J-O calculation for Dy,Tb:GYSAG crystal was performed according to the absorption spectrum (exclude the Tb^{3+} absorption). The detailed J-O calculation procedure is similar to that reported in the Ref.[1,2]. The square of the reduced matrix elements $U^{(t)}$ used for the J-O calculation has been described by Carnall et al [3]. The J-O parameters, including experimental and calculated oscillator strengths, were calculated and listed in Table S1. The relative square deviation R were fitted as 7.98%, indicating the high reliability of the calculated results. The three intensity parameters Ω_t (t=2, 4, 6) were fitted as 2.47×10^{-20} cm^2, 0.98×10^{-20} cm^2 and 2.92×10^{-20} cm^2, respectively. Using the fitted J-O intensity parameters, the radiative properties of the $^4F_{9/2}$ excited state of Dy^{3+}, including electric-dipole transition probability (A_{ed}), magnetic-dipole transition probability (A_{md}) and fluorescence branching ratio (β), are calculated as listed in Table S2.

Table S3 Experimental and calculated oscillator strengths as well as J-O intensity parameters of Dy^{3+} in Dy,Tb:GYSAG crystal

$^6H_{15/2}\rightarrow{}^{2S+1}L_J$ transitions	λ (nm)	f_{ed}^{exp} ($\times10^{-6}$)	f_{ed}^{cal} ($\times10^{-6}$)	$f_{ed}^{exp-cal}$ ($\times10^{-6}$)
$^4G_{11/2}$	428	0.1359	0.1330	0.0029
$^4I_{15/2}$	453	0.7521	0.6705	0.0816
$^4F_{9/2}$	475	0.1113	0.2866	-0.1753
$^6F_{3/2}$	751	0.2844	0.3018	-0.0174
$^6F_{5/2}$	805	1.5322	1.5991	-0.0669
$^6F_{7/2}$	901	2.9344	3.4251	-0.4907
$^6F_{9/2}+{}^6H_{7/2}$	1095	3.3194	4.2605	-0.9411
$^6F_{11/2}+{}^6H_{9/2}$	1280	4.3832	5.6381	-1.2549
$^6H_{11/2}$	1690	1.8878	1.4930	0.3948
Relative square deviation: R=7.98%; Ω_2=2.47, Ω_4=0.98, Ω_6=2.92				

Table S4 Transition probability, fluorescence branching ratio, and radiative lifetime of Dy^{3+} in Dy,Tb:GYSAG crystal

$^4F_{9/2}\rightarrow{}^{2S+1}L_J$ transitions	λ (nm)	S_{ed} (10^{-20} cm^2)	S_{md} (10^{-20} cm^2)	A_{ed} (s^{-1})	β (%)	τ_{rad} (ms)
$^6F_{1/2}$	1490	0.00045	0	0.0689	0.0044	0.644
$^6F_{3/2}$	1375	0.00078	0	0.1512	0.0097	
$^6F_{5/2}$	1239	0.02033	0	5.3972	0.3475	
$^6F_{7/2}$	1052	0.01510	0.0200	6.5404	0.9928	

$^6H_{5/2}$	941	0.00815	0	4.9348	0.3177
$^6F_{9/2}$	873	0.00986	0.0120	7.4863	1.0828
$^6H_{7/2}$	856	0.03442	0.00692	27.7509	2.1544
$^6F_{11/2}$	799	0.01868	0.07808	18.5212	6.2949
$^6H_{9/2}$	762	0.01995	0.00449	22.7827	1.8052
$^6H_{11/2}$	667	0.04000	0.01503	68.0019	6.0624
$^6H_{13/2}$	582	0.32492	0	830.123	53.445
$^6H_{15/2}$	490	0.09958	0	426.880	27.483

5. The emission cross section of Dy,Tb:GYSAG(1) crystal

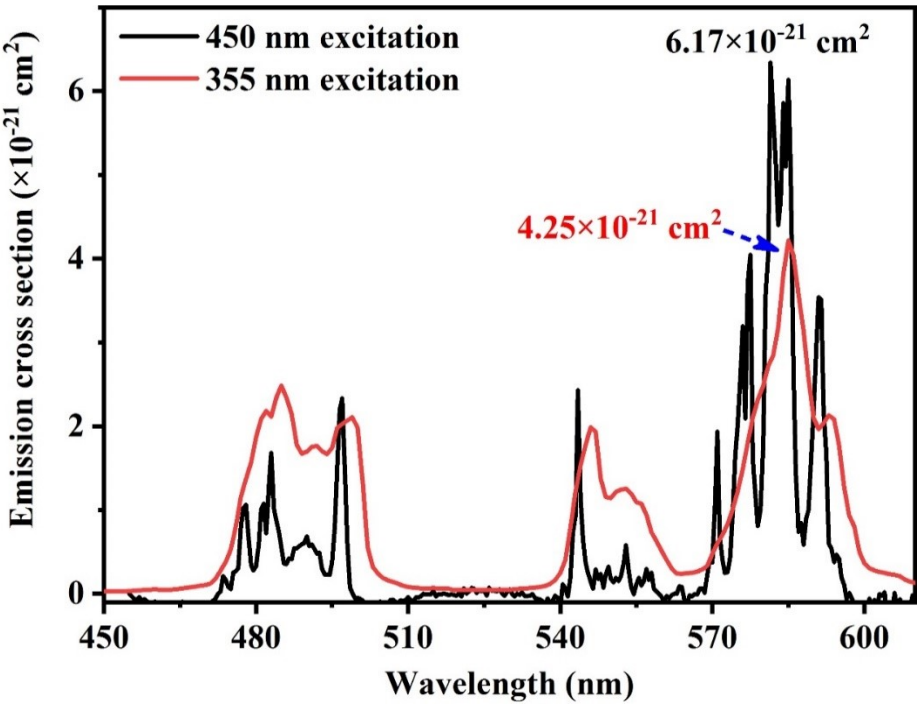

Fig.S4 The calculated emission cross sections upon 355 nm and 450 nm excitation.

6. The CIE 1931 chromaticity coordinate values

Table S5 The CIE 1931 chromaticity coordinate values of Dy^{3+} and Dy^{3+}/Tb^{3+} copdoped mixed garnet crystals upon 450 nm, 355 nm and 266 nm excitation.

Excitation wavelength	Crystals	(CIEx, CIEy)	Peak wavelengths
450 nm	Dy:GSAG	(0.4009, 0.4135)	585 nm
	Dy,Tb:GSAG	(0.3889, 0.4184)	585 nm
	Dy,Tb:GYSAG(1)	(0.3861, 0.4178)	585 nm

	Dy,Tb:GSYAG(2)	(0.3838, 0.4173)	585 nm
355 nm	Dy:GSAG	(0.3162, 0.3522)	485 nm
	Dy,Tb:GSAG	(0.3230, 0.4115)	485 nm
	Dy,Tb:GYSAG(1)	(0.3095, 0.4006)	485 nm
	Dy,Tb:GSYAG(2)	(0.3261, 0.4175)	485 nm
266 nm	Dy:GSAG	(0.3873, 0.3879)	585 nm
	Dy,Tb:GSAG	(0.3193, 0.5326)	545 nm
	Dy,Tb:GYSAG(1)	(0.3229, 0.5337)	546 nm
	Dy,Tb:GSYAG(2)	(0.3182, 0.5342)	546 nm

7. The emission spectra of Dy,Tb:GYSAG(1) crystal upon 355 and 266 nm excitation

It can be seen from the Fig.S5(a) that the emission intensity of $^5D_4 \rightarrow {}^7F_J$ transition (Green color as depicted in Fig.S5(b)) of Tb^{3+} is far stronger than that of $^5D_3 \rightarrow {}^7F_J$ transition (Blue color as depicted in Fig.S5(b)).

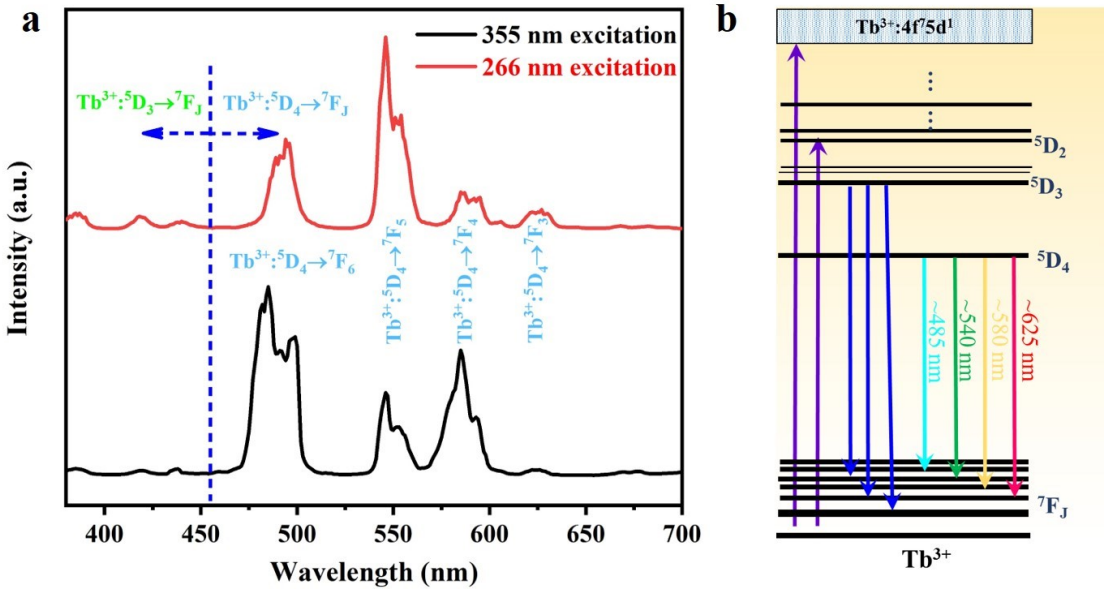

Fig.S5 (a) The 355 nm and 266 nm excited emission spectra of Dy,Tb:GYSAG(1) crystal. (b) The Schematic energy levels of Tb^{3+}.

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