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**Supporting information** 



Fig. S1 PL and PLE spectra of Gd<sub>3-y</sub>Ca<sub>y-0.009</sub>GaO<sub>6</sub>:0.009Eu<sup>2+</sup> (y=0.1).



Fig. S2 PL spectra of  $Gd_{2.9}Ca_{0.1}GaO_6$ :xEu<sup>2+</sup> (x = 0.009-0.025).



Fig. S3 Crystal structure of Gd<sub>3</sub>GaO<sub>6</sub>.



Fig. S4 TL spectra of  $Gd_{3-y}Ca_{y-0.02}GaO_6:0.02Eu^{2+}$  (y = 0.1-0.5).



Fig. S5 Temperature-dependent PL spectra of  $Gd_{3-y}Ca_{y-0.02}GaO_6:0.02Eu^{2+}$  (y=0.2).



Fig. S6 Temperature-dependent PL spectra of Gd<sub>3-y</sub>Ca<sub>y-0.02</sub>GaO<sub>6</sub>:0.02Eu<sup>2+</sup>, 0.2K<sup>+</sup>

(y=0.2).



Fig. S7 Temperature-dependent PL spectra of  $Gd_{3-y}Ca_{y-0.02}GaO_6:0.02Eu^{2+}$  (y=0.2).



Fig. S8 Temperature-dependent PL spectra of Gd<sub>3-y</sub>Ca<sub>y-0.02</sub>GaO<sub>6</sub>:0.02Eu<sup>2+</sup>, 0.2K<sup>+</sup>

(y=0.2).



Fig. S9 Variation of color coordinates with temperature of Gd<sub>2.8</sub>Ca<sub>0.2</sub>GaO<sub>6</sub>:0.02Eu<sup>2+</sup>.



Fig. S10 Quantum efficiency.

$$I(t) = I_0 + A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right)$$
(S1)

where I and  $I_0$  are the emission intensities at the time of  $\tau$  and 0, respectively,  $A_1$  and  $A_2$  represent constants; t is the time,  $t_1$  and  $t_2$  are the first and the second values represent the life of the exponential fitting, respectively.

$$\tau = (A_1\tau_1^2 + A_2\tau_2^2) / (A_1\tau_1 + A_2\tau_2)$$
(S2)

where t is time and I(t) is the intensity at time t.

Formula	y=0	y=0.05	y=0.1	y= 0.15	y= 0.2	y= 0.25
Crystal system	Orthorhombic					
Space group		Cmc21				
a(Å)	8.9802	8.9910	8.9946	8.9963	8.9981	9.0073
b(Å)	11.2651	11.2755	11.2808	11.2812	11.2875	11.2881
c(Å)	5.4741	5.4780	5.4799	5.4820	5.4830	5.4868
2theta interval	10-80	10-80	10-80	10-80	10-80	10-80
$\chi^2$	2.959	2.796	6.763	2.792	2.799	3.599
$R_{wp}$	6.27%	7.17%	8.29%	7.17%	7.41%	7.47%
R <sub>p</sub>	4.6%	5.41%	5.28%	5.14%	5.36%	5.30%

Table S1 The crystal structure data and refinement parameters for samples Gd<sub>3-y</sub>Ca<sub>y-0.02</sub>GaO<sub>6</sub>:0.02Eu<sup>2+</sup>.

у	Gd1O <sub>7</sub>	Gd2O <sub>7</sub>	$GaO_4$
0	19.424	19.817	5.861
0.05	19.456	19.851	3.006
0.10	19.481	19.876	3.010
0.15	19.490	19.886	3.011
0.20	19.509	19.904	3.014
0.25	19.523	19.919	3.016

Table S2 Variation of crystal cell with doping concentration of  $Ca^{2+}$  (Å<sup>3</sup>).

	y=0	y=0.05	y=0.1	y=0.15	y=0.2	y=0.25
Gd <sub>1</sub> -O1	2.0843	2.0863	2.087	2.0946	2.0883	2.0896
Gd <sub>1</sub> -O2	2.2076	2.2093	2.2103	2.1943	2.2116	2.2123
Gd <sub>1</sub> -O3	2.3563	2.3590	2.3600	2.3370	2.3613	2.3626
Gd <sub>1</sub> -O4	2.3900	2.3923	2.3933	2.3990	2.3946	2.3960
Gd <sub>1</sub> -O5	2.4016	2.4040	2.4050	2.4490	2.4060	2.4080
Gd <sub>1</sub> -O6	2.4730	2.4753	2.4763	2.5513	2.4776	2.4790
Gd <sub>1</sub> -O7	2.7420	2.7443	2.7456	2.7413	2.7473	0.7486
Gd <sub>2</sub> -O1	2.2073	2.2096	2.2106	2.2056	2.2116	2.2130
Gd <sub>2</sub> -O2	2.2073	2.2096	2.2106	2.2056	2.2116	2.2130
Gd <sub>2</sub> -O3	2.4576	2.4600	2.4610	2.4253	2.4623	2.4636
Gd <sub>2</sub> -O4	2.4576	2.4600	2.4610	2.4680	2.4623	0.4636
Gd <sub>2</sub> -O5	2.4830	2.4853	2.4863	2.4850	2.4876	2.4893
Gd <sub>2</sub> -O6	2.4830	2.4853	2.4863	2.5613	2.4876	2.4893
Gd <sub>2</sub> -O7	2.6006	2.6030	2.6040	2.6393	2.6053	2.6060

Table S3 Variation of bond length with doping concentration of  $Ca^{2+}$  (Å).

Temperature (°C)	CIE	
25	(0.6773,0.3218)	
50	(0.6733,0.3257)	
75	(0.6689,0.3301)	
100	(0.6646,0.3344)	
125	(0.6600,0.3390)	
150	(0.6553,0.3436)	
175	(0.6506,0.3483)	
200	(0.6458,0.3531)	

Table S4 CIE coordinates at different temperatures.