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## Supporting Information

## An Orange-Yellow-Emitting Lu<sub>2-x</sub>Mg<sub>2</sub>Al<sub>2-y</sub>Ga<sub>y</sub>Si<sub>2</sub>O<sub>12</sub>: xCe<sup>3+</sup> Phosphor-in-Glass Film for Laser-Driven White Light

Shisheng Lin, <sup>1</sup> Hang Lin, <sup>1, 2, 3</sup> \* Pengfei Wang, <sup>1, 4</sup> Ping Sui, <sup>1, 4</sup> Hongyi Yang, <sup>5</sup> Ju Xu, <sup>1</sup> Yao Cheng, <sup>1</sup> Yuansheng Wang<sup>1,\*</sup>

<sup>1</sup> Key Laboratory of Optoelectronic Materials Chemistry and Physics, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, Fujian, 350002 (P. R. China)

E-mail: <u>lingh@fjirsm.ac.cn;</u> E-mail: <u>yswang@fjirsm.ac.cn;</u>

<sup>2</sup> Fujian Science & Technology Innovation Laboratory for Optoelectronic Information of China, Fuzhou, Fujian, 350108 (P. R. China)

<sup>3</sup> State Key Laboratory of Structural Chemistry, Fuzhou, Fujian, 350002 (P. R. China)

<sup>4</sup> College of Chemistry and Materials Science, Fujian Normal University, Fuzhou, Fujian, 350007 (P. R. China)

<sup>5</sup> Xiamen Institute of Rare-earth Materials, Haixi Institutes, Chinese Academy of Sciences, Xiamen, Fujian, 361000 (P. R. China)



Figure S1. Schematical illustration of preparation procedure of the LMAGS: Ce<sup>3+</sup> PiG film-on-SP.



Figure S2. Photograph of the photo-sensitive paper upon blue laser irradiation



**Figure S3.** Rietveld refinement on  $Lu_{2.0-x}Mg_2Al_{1.5}Ga_{0.5}Si_2O_{12}$ :  $xCe^{3+}$  phosphor, showing the observed (black crosses) and calculated (red line) XRD profiles, and the difference between them (blue line).



**Figure S4.** Rietveld refinement on  $Lu_{1.9}Mg_2Al_{2.0-y}Ga_ySi_2O_{12}$ : 0.1Ce<sup>3+</sup> phosphor, showing the observed (black crosses) and calculated (red line) XRD profiles, and the difference between them (blue line).



**Figure S5.** SEM observations on the cross section of LMAGS: Ce<sup>3+</sup> PiG film-on-SP with different film thickness.



Figure S6. PL spectra of  $Lu_{2.0-x}Mg_2Al_{1.5}Ga_{0.5}Si_2O_{12}$ :  $xCe^{3+}$  PiG film-on-SP under 450 nm excitation.



Figure S7. Relationship between log(x) and log(I/x) in the  $Lu_{2.0-x}Mg_2Al_{1.5}Ga_{0.5}Si_2O_{12}$ :  $xCe^{3+}$  phosphor.

Discussions on Figure S7:

In order to study the concentration quenching mechanism, the parameter of  $R_c$  reflecting the average distance of  $Ce^{3+}$  is introduced by using the following expression <sup>[1]</sup>:

$$R_c \approx 2 \left[ \frac{3V}{4\pi x_c N} \right]^{1/3} \tag{1}$$

where V is the volume of unit cell,  $x_c$  the critical concentration of activator and N the number of available sites for the dopant in a unit cell. Taken V=1688.219 Å<sup>3</sup>,  $x_c$ =0.10, and N=8, R<sub>c</sub> is evaluated to be ~16 Å. The electronic exchange interaction should be only effective at R<sub>c</sub><5 Å to achieve energy transfer among Ce<sup>3+</sup> ions. As such, the electric multipolar-multipolar interaction should be the main mechanism. Then, we adopted the Dexter theory to analyze the type of multipolarmultipolar interaction by the following equation<sup>[2]</sup>:

$$I / x = K \left[ 1 + \beta(x)^{\frac{\theta}{3}} \right]^{-1}$$
(2)

where K and  $\beta$  are constants,  $\theta$ =6, 8. 10 means the electric multipole index corresponding to the dipole-dipole (d-d), dipole-quadrupole (d-q) and quadrupolequadrupole (q-q) interaction, respectively. By plotting log(I/x) versus log(x),  $\theta$  is calculated as 5.13, indicating that the main mechanism for concentration quenching in LMAGS: Ce<sup>3+</sup> is the d-d electric multipolar-multipolar interaction.



Figure S8. The measured luminescent curves of  $Lu_{1.9}Mg_2Al_{2.0-y}Ga_ySi_2O_{12}$ : 0.1Ce<sup>3+</sup> PiG film-on-SP to calculate quantum efficiency.



**Figure S9.** Temperature-dependent PL spectra in  $Lu_{1.9}Mg_2Al_{2.0-y}Ga_ySi_2O_{12}$ : 0.1Ce<sup>3+</sup> PiG film-on-SP from 300 to 600 K under 450 nm excitation.



Figure S10. In-line transmittance spectra of sapphire, AR-coated sapphire and BP-coated sapphire.



Figure S11.  $P_{in}$  dependent electroluminescent spectra of LMAGS: Ce<sup>3+</sup> PiG film-on-SP with different mass ratios of LMAGS: Ce<sup>3+</sup> phosphor to glass powders.



Figure S12.  $P_{in}$  dependent electroluminescent spectra of LMAGS: Ce<sup>3+</sup> PiG film-on-SP with different film thickness.



**Figure S13.** Pumping power dependent luminous efficacy of the LMAGS: Ce<sup>3+</sup> PiG film-on-SP with (a) different weight ratios of LMAGS: Ce<sup>3+</sup> phosphor to glass powders and (b) different film thicknesses.



**Figure S14.**  $P_{in}$  dependent EL spectra of LMAGS: Ce<sup>3+</sup> PiG film-on-SP with different Gaconcentration: (a) y=0.00, (b) y=0.50, and (c) y=1.00; insets are the corresponding magnified spectra in the range of 500-700 nm. (d)  $P_{in}$  dependent EL spectra of Lu<sub>1.9</sub>Mg<sub>2</sub>Al<sub>2</sub>Si<sub>2</sub>O<sub>12</sub>: 0.1Ce<sup>3+</sup> PiG filmon-SP "phosphor wheel" measured under rotatory-reflective mode; insets are the digital photo of phosphor wheel (left) and the enlarged spectra in the region of 500-800 nm (right).



**Figure S15.** Pumping power dependent luminous efficacy of the LMAGS: Ce<sup>3+</sup> PiG film-on-SP with different Ga-concentration.



**Figure S16.** Diffuse-reflectance spectrum of the un-doped LMAGS, inset shows the relationship of  $[\alpha hv]^2$  versus photon energy *hv* to determine the optical bandgap of LMAGS.



**Figure S17.** Local temperature of LMAGS: Ce<sup>3+</sup> PiG film-on-SP with different Ga-concentration at the laser spot.



**Figure S18.** the variations in chromaticity coordinates of LMAGS: Ce<sup>3+</sup> PiG film-on-SP with different Ga-concentration.



Figure S19. The derived P<sub>in</sub> dependent luminous efficacy in rotatory-reflective mode.



**Figure S20.** The incident power dependent absorption efficiencies of the LMAGS: Ce<sup>3+</sup> PiG filmon-SP under rotatory-reflective mode.



Figure S21. EL spectra of LMAGS: Ce PiG film-on-SP with different Ce-concentration under different input power density in rotatory-reflective mode.



Figure S22. EL spectra of LMAGS: Ce PiG film-on-SP with different Ga-concentration under different input power density in rotatory-reflective mode.

## References

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