Supporting Information

An Orange-Yellow-Emitting Lu_{2-x}Mg_{2}Al_{2-y}Ga_{y}Si_{2}O_{12}: xCe^{3+} Phosphor-in-Glass Film for Laser-Driven White Light

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

1 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: lingh@fjirsm.ac.cn; E-mail: yswang@fjirsm.ac.cn;

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

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

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

5 Xiamen Institute of Rare-earth Materials, Haixi Institutes, Chinese Academy of Sciences, Xiamen, Fujian, 361000 (P. R. China)
Figure S1. Schematic illustration of preparation procedure of the LMAGS: Ce$^{3+}$ PiG film-on-SP.
**Figure S2.** Photograph of the photo-sensitive paper upon blue laser irradiation
Figure S3. Rietveld refinement on Lu$_{2.0x}$Mg$_{2}$Al$_{1.5}$Ga$_{0.5}$Si$_2$O$_{12}$: 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$_2$Al$_2$O$_{12}$: 0.1Ce$^{3+}$ 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$^{3+}$ PiG film-on-SP with different film thickness.
Figure S6. PL spectra of Lu$_{2.0-x}$Mg$_x$Al$_{1.5}$Ga$_{0.5}$Si$_2$O$_{12}$: $x$Ce$^{3+}$ PiG film-on-SP under 450 nm excitation.
Figure S7. Relationship between log($x$) and log($1/x$) in the Lu$_{2.0-x}$Mg$_{1.5}$Ga$_{0.5}$Si$_2$O$_{12}$: $xCe^{3+}$ 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 [1]:

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

(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$^3$, $x_c=0.10$, and $N=8$, $R_c$ is evaluated to be ~16 Å. The electronic exchange interaction should be only effective at $R_c<5$ Å to achieve energy transfer among $Ce^{3+}$ ions. As such, the electric multipolar-multipolar interaction should be the main mechanism. Then, we adopted the Dexter theory to analyze the type of multipolar-
multipolar interaction by the following equation [2]:

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

(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 quadrupole-quadrupole (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\(^{3+}\) is the d-d electric multipolar-multipolar interaction.
Figure S8. The measured luminescent curves of Lu$_{1.9}$Mg$_{2.0}$Al$_{2.0-y}$Ga$_{y}$Si$_2$O$_{12}$: 0.1Ce$^{3+}$ PIG film-on-SP to calculate quantum efficiency.
Figure S9. Temperature-dependent PL spectra in Lu$_{1.9}$Mg$_2$Al$_{2.0-y}$Ga$_y$Si$_2$O$_{12}$: 0.1Ce$^{3+}$ 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$^{3+}$ PiG film-on-SP with different mass ratios of LMAGS: Ce$^{3+}$ phosphor to glass powders.
**Figure S12.** $P_{in}$ dependent electroluminescent spectra of LMAGS: Ce$^{3+}$ PiG film-on-SP with different film thickness.
Figure S13. Pumping power dependent luminous efficacy of the LMAGS: Ce³⁺ PiG film-on-SP with (a) different weight ratios of LMAGS: Ce³⁺ phosphor to glass powders and (b) different film thicknesses.
Figure S14. $P_m$ dependent EL spectra of LMAGS: Ce$^{3+}$ PiG film-on-SP with different Ga-concentration: (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) $P_m$ dependent EL spectra of Lu$_{1.9}$Mg$_2$Al$_2$Si$_2$O$_{12}$: 0.1Ce$^{3+}$ PiG film-on-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$^{3+}$:PiG film-on-SP with different Ga-concentration.
Figure S16. Diffuse-reflectance spectrum of the un-doped LMAGS, inset shows the relationship of $[\alpha h\nu]^2$ versus photon energy $h\nu$ to determine the optical bandgap of LMAGS.
Figure S17. Local temperature of LMAGS: Ce$^{3+}$ PiG film-on-SP with different Ga-concentration at the laser spot.
Figure S18. The variations in chromaticity coordinates of L-MAGS: Ce³⁺ PiG film-on-SP with different Ga-concentration.
Figure S19. The derived P_in dependent luminous efficacy in rotatory-reflective mode.
Figure S20. The incident power dependent absorption efficiencies of the LMAGS: Ce³⁺ PiG film-on-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 LMGAS: Ce PiG film-on-SP with different Ga-concentration under different input power density in rotatory-reflective mode.
References
