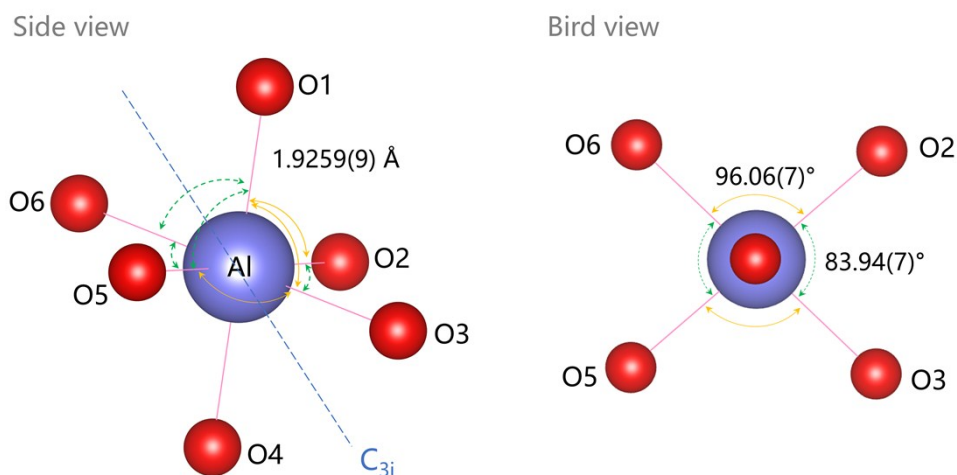


## Supporting Information

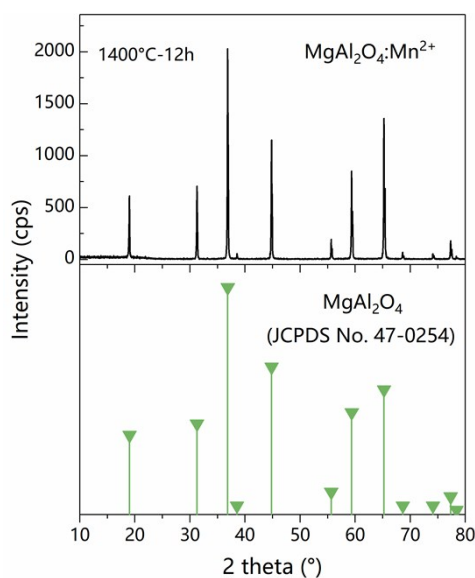
### Ultrabroadband Red Luminescence of Mn<sup>4+</sup> in MgAl<sub>2</sub>O<sub>4</sub> Peaking at 651 nm

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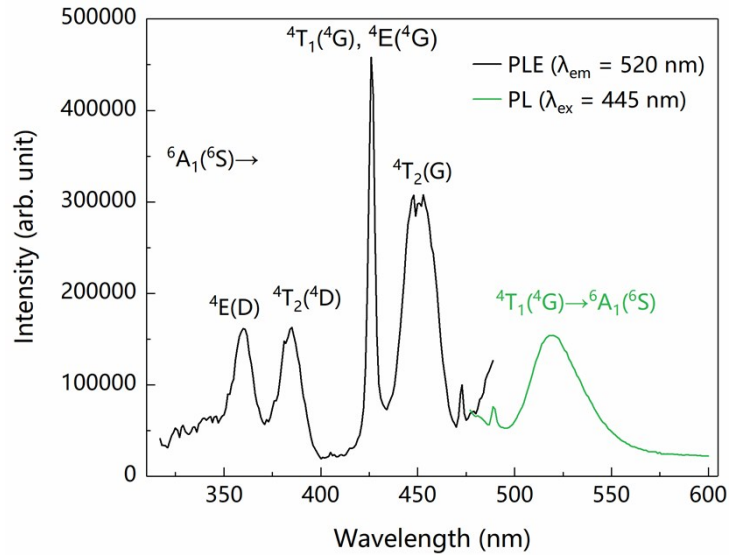
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**Figure S1**  $\text{Al}^{3+}$  octahedral coordination in  $\text{MgAl}_2\text{O}_4$  (ICSD 31373) showing the interatomic distances and angles between the chemical bonds.  $d_{\text{Al-O1}} = d_{\text{Al-O2}} = d_{\text{Al-O3}} = d_{\text{Al-O4}} = d_{\text{Al-O5}} = d_{\text{Al-O6}} = 1.9259(9) \text{ \AA}$ ;  $\angle \text{O1-Al-O2} = \angle \text{O1-Al-O3} = \angle \text{O2-Al-O6} = \angle \text{O3-Al-O5} = \angle \text{O2-Al-O4} = \angle \text{O3-Al-O4} = \angle \text{O4-Al-O5} = \angle \text{O4-Al-O6} = 96.06(7)^\circ$ ,  $\angle \text{O1-Al-O5} = \angle \text{O1-Al-O6} = \angle \text{O2-Al-O3} = \angle \text{O5-Al-O6} = 83.94(7)^\circ$ .

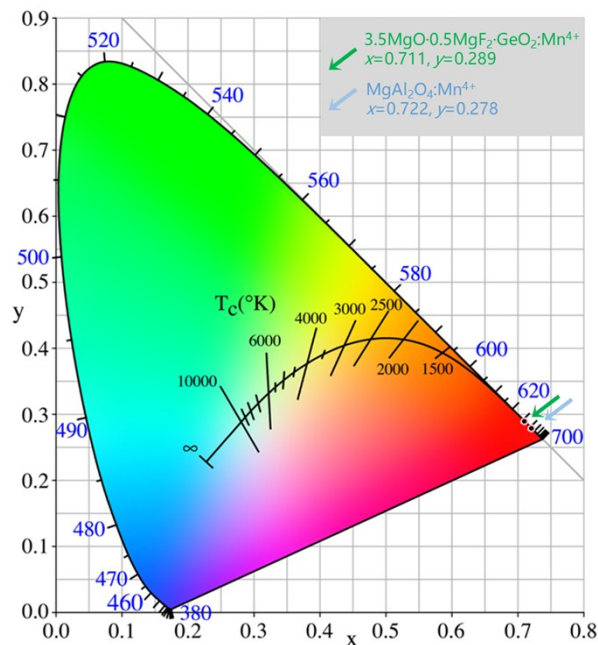


**Figure S2** XRD pattern of the spinel phosphor calcined at 1400 °C for 12 h. The standard pattern for  $\text{MgAl}_2\text{O}_4$  (JCPDS card no. 47-0254) was provided as reference.



**Figure S3** Photoluminescence spectra of the  $\text{MgAl}_2\text{O}_4:\text{Mn}^{2+}$  phosphor calcined at  $1400\text{ }^\circ\text{C}$  for 12 h. The corresponding energy transitions were assigned based on Ref. [1].

- [1] E. H. Song, Y. Y. Zhou, Y. Wei, X. X. Han, Z. R. Tao, R. L. Qiu, Z. G. Xia and Q. Y. Zhang. A thermally stable narrow-band green-emitting phosphor  $\text{MgAl}_2\text{O}_4:\text{Mn}^{2+}$  for wide color gamut backlight display application. *J. Mater. Chem. C*, 2019, 7, 8192-8198.



**Figure S4** Chromaticity coordinate ( $\lambda_{\text{ex}} = 440\text{ nm}$ ) of  $\text{MgAl}_2\text{O}_4:\text{Mn}^{4+}$  in CIE 1931 diagram. The chromaticity of the commercial  $3.5\text{MgO}\cdot 0.5\text{MgF}_2\cdot \text{GeO}_2:\text{Mn}^{4+}$  was also given.

### **Preparation of $\text{MgAl}_2\text{O}_4:\text{Mn}^{4+}$ by the molten salt method:**

MgO (A.R., Sinopharm, China) and  $\text{Al}_2\text{O}_3$  (99.99%,  $\gamma$ -phase,  $\leq 20$  nm, Aladdin, China) were used as the raw chemicals.  $\text{MnCO}_3$  (A.R., Sinopharm, China) and  $\text{LiCl}\cdot\text{H}_2\text{O}$  (A.R., Sinopharm, China) were used as the dopant source and the salt. MgO,  $\text{Al}_2\text{O}_3$ , and  $\text{MnCO}_3$  were weighed according to the formula of  $\text{MgAl}_{1.998}\text{Mn}_{0.002}\text{O}_4$  and then mixed with  $\text{LiCl}\cdot\text{H}_2\text{O}$  in a weight ratio of 1:5. The mixture was then transferred to corundum crucibles covered with a lid and heated at 950 °C for 3 h. After cooling to room temperature, the product was washed with hot deionized water for 5 times to remove the salt, concentrated by centrifugation, and then dried in an oven at 100 °C for 12 h.