

1 Article-Supporting Information

2 **LaAlO<sub>3</sub>:Mn<sup>4+</sup> as near-infrared emitting persistent**  
 3 **luminescence phosphor for medical imaging: A**  
 4 **charge compensation study**

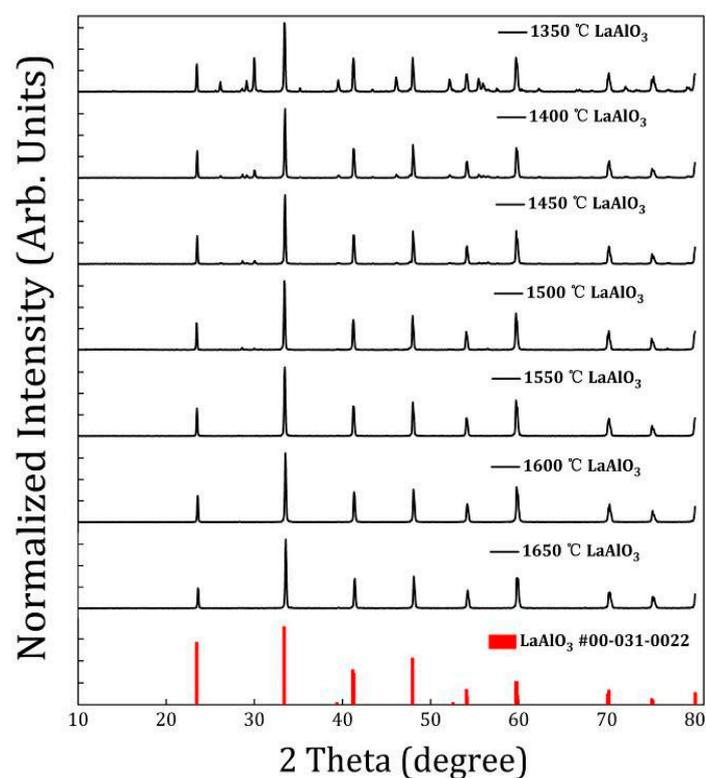
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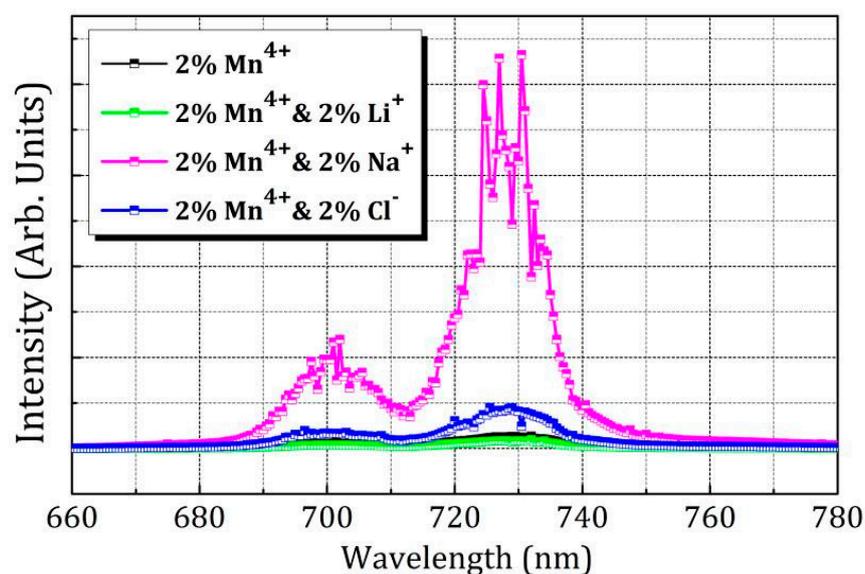
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10 Figure S1-S16; Table S1,S2



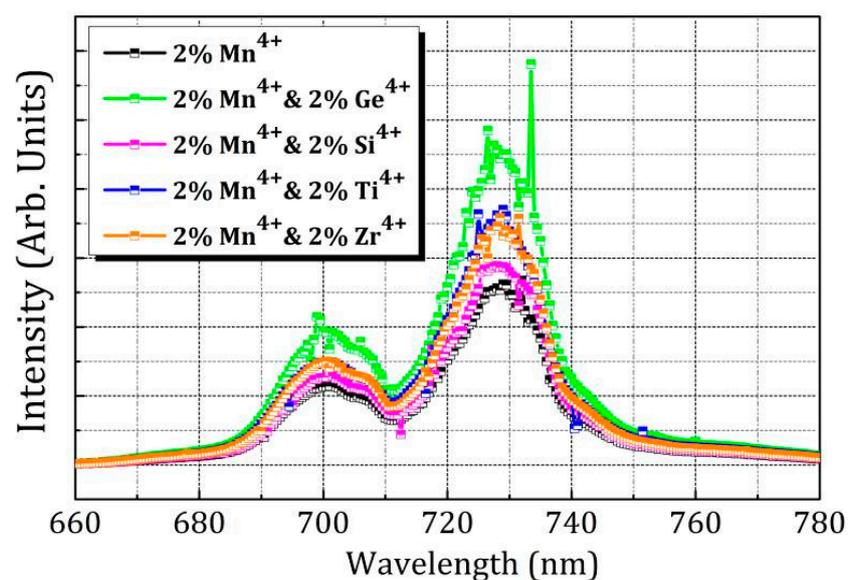
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12 **Figure S1.** XRD pattern of LaAlO<sub>3</sub> synthesized through a solid-state reaction method. The synthesis  
 13 temperature is indicated next to the diffractograms. The intensities of the XRD patterns are normalized to  
 14 arbitrary units [0, 1].



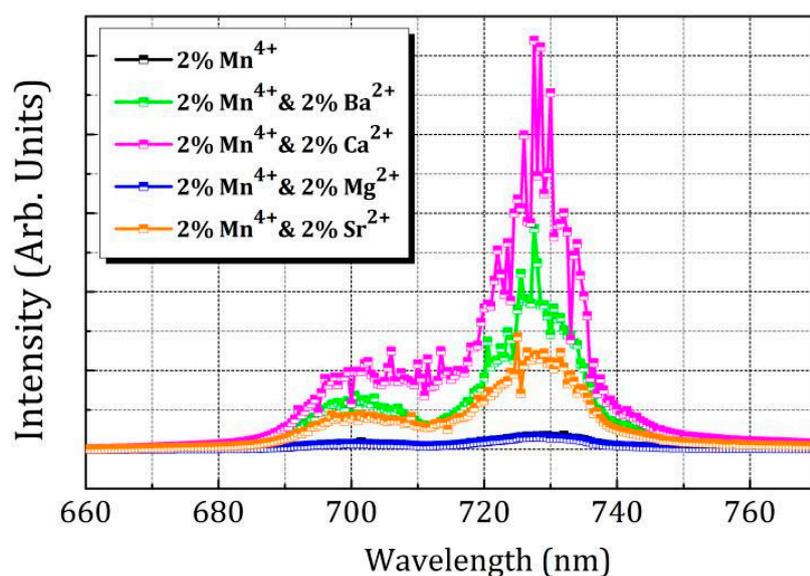
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16 **Figure S2.** Photoluminescence (PL) spectra of LaAlO<sub>3</sub>:2%Mn<sup>4+</sup>,2%Li<sup>+</sup>, LaAlO<sub>3</sub>:2%Mn<sup>4+</sup>,2%Na<sup>+</sup>, and  
 17 LaAlO<sub>3</sub>:2%Mn<sup>4+</sup>,2%Cl<sup>-</sup> phosphors. All the PL spectra ( $\lambda_{\text{ex}}=335$  nm) are in the range 660-780 nm and PL spectrum  
 18 of LaAlO<sub>3</sub>:2%Mn<sup>4+</sup> phosphor is shown in black for comparison.



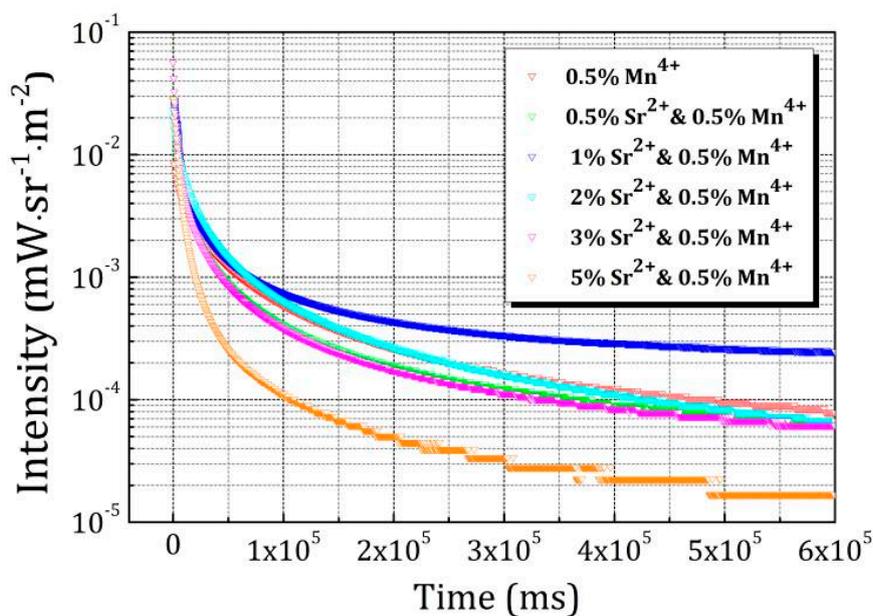
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20 **Figure S3.** Photoluminescence (PL) spectra of LaAlO<sub>3</sub>:2%Mn<sup>4+</sup>,2%Ge<sup>4+</sup>, LaAlO<sub>3</sub>:2%Mn<sup>4+</sup>,2%Si<sup>4+</sup>,  
 21 LaAlO<sub>3</sub>:2%Mn<sup>4+</sup>,2%Ti<sup>4+</sup>, and LaAlO<sub>3</sub>:2%Mn<sup>4+</sup>,2%Zr<sup>4+</sup> phosphors. All the PL spectra ( $\lambda_{\text{ex}}=335$  nm) are in the range  
 22 660-780 nm and PL spectrum of LaAlO<sub>3</sub>: 2%Mn<sup>4+</sup> phosphor is shown in black for comparison.



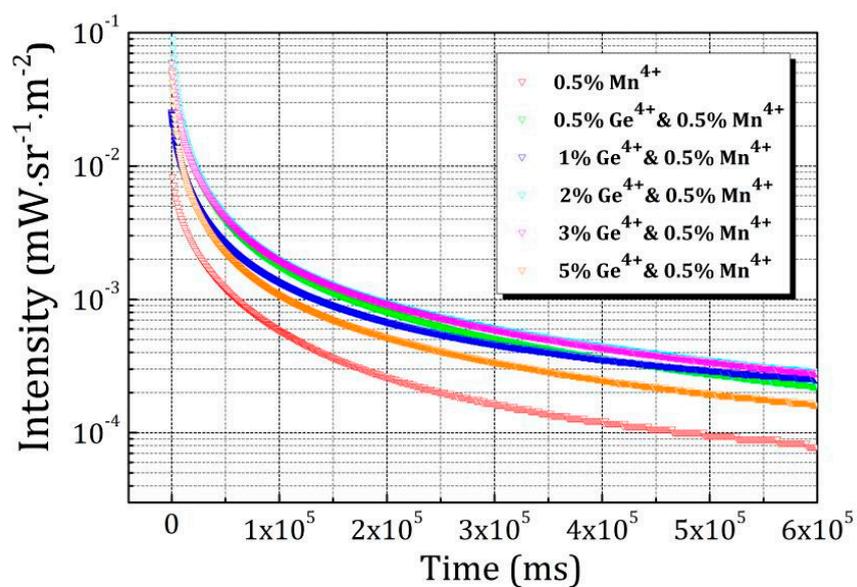
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24 **Figure S4.** Photoluminescence (PL) spectra of  $\text{LaAlO}_3:2\%\text{Mn}^{4+}, 2\%\text{Ba}^{2+}$ ,  $\text{LaAlO}_3:2\%\text{Mn}^{4+}, 2\%\text{Ca}^{2+}$ ,  
 25  $\text{LaAlO}_3:2\%\text{Mn}^{4+}, 2\%\text{Mg}^{2+}$ , and  $\text{LaAlO}_3:2\%\text{Mn}^{4+}, 2\%\text{Sr}^{2+}$  phosphors. All the PL spectra ( $\lambda_{\text{ex}} = 335 \text{ nm}$ ) are in the  
 26 range 660–780 nm and PL spectrum of  $\text{LaAlO}_3:2\%\text{Mn}^{4+}$  phosphor is shown in black for comparison.

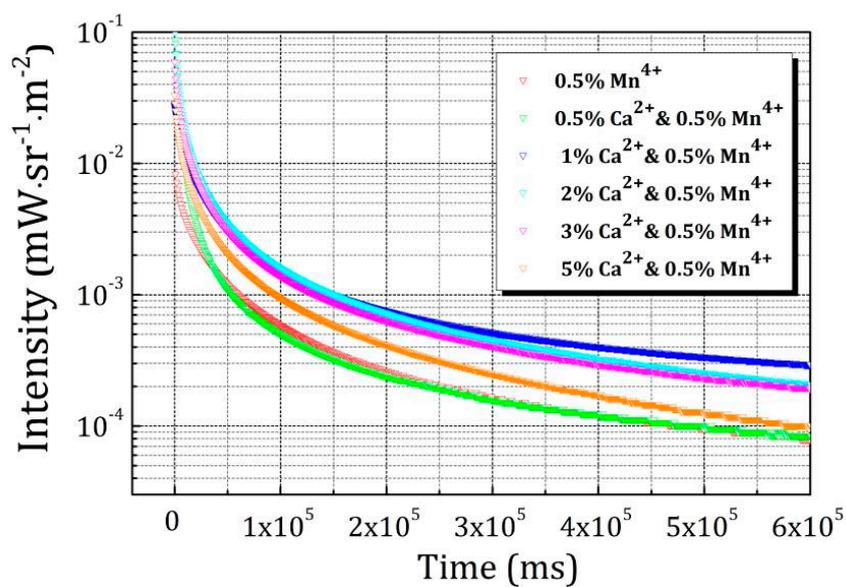


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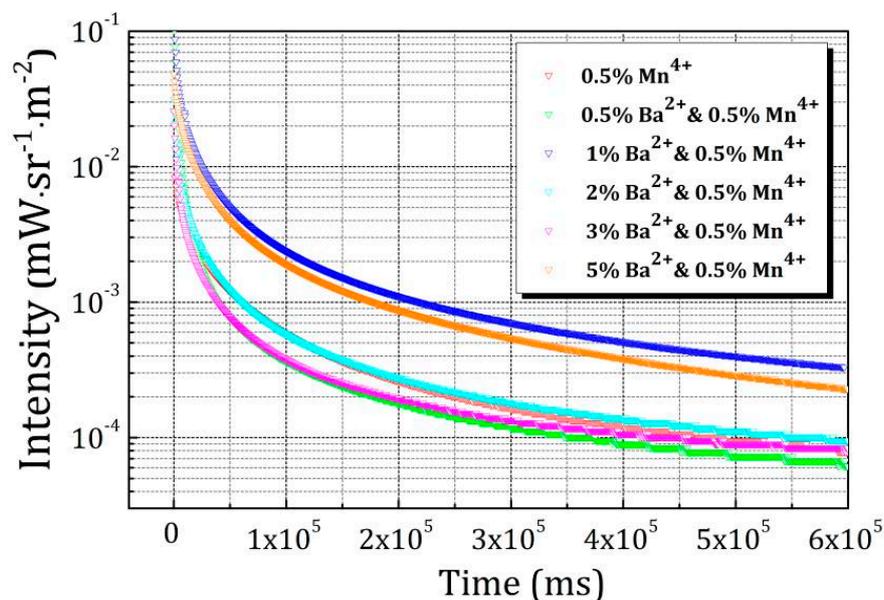
28 **Figure S5.** Persistent luminescence decay curves of  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+}, y\text{Sr}^{2+}$  ( $y = 0.5\%, 1\%, 2\%, 3\%$ , and  $5\%$ )  
 29 phosphors after 5 min of irradiation with a Xenon arc lamp. The red curve corresponds to  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+}$  as  
 30 an intensity benchmark of persistent luminescence.



31  
 32 **Figure S6.** Persistent luminescence decay curves of  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},y\text{Ge}^{4+}$  ( $y = 0.5\%, 1\%, 2\%, 3\%, \text{ and } 5\%$ )  
 33 phosphors after 5 min of irradiation with a Xenon arc lamp. The red curve corresponds to  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+}$  as  
 34 an intensity benchmark of persistent luminescence.

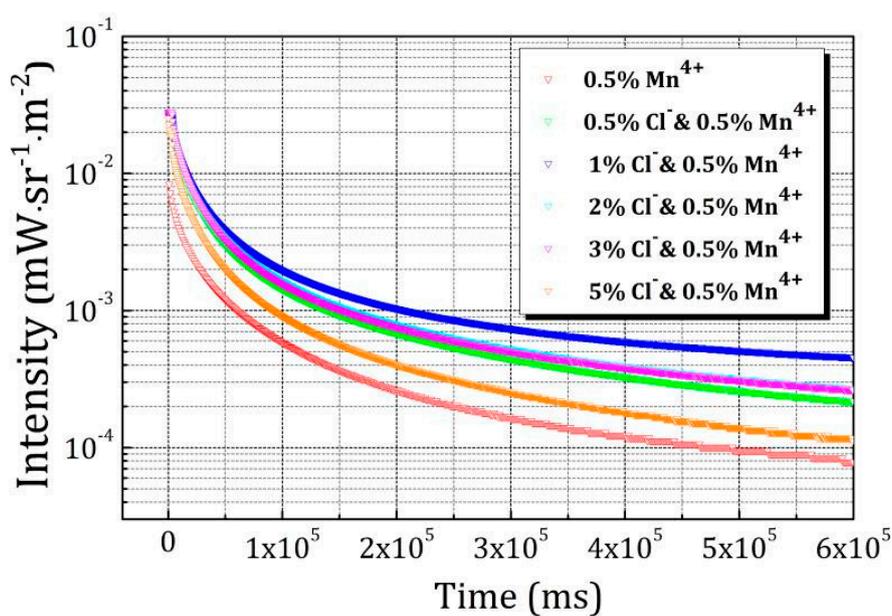


35  
 36 **Figure S7.** Persistent luminescence decay curves of  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},y\text{Ca}^{2+}$  ( $y = 0.5\%, 1\%, 2\%, 3\%, \text{ and } 5\%$ )  
 37 phosphors after 5 min of irradiation with a Xenon arc lamp. The red curve corresponds to  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+}$  as  
 38 an intensity benchmark of persistent luminescence.



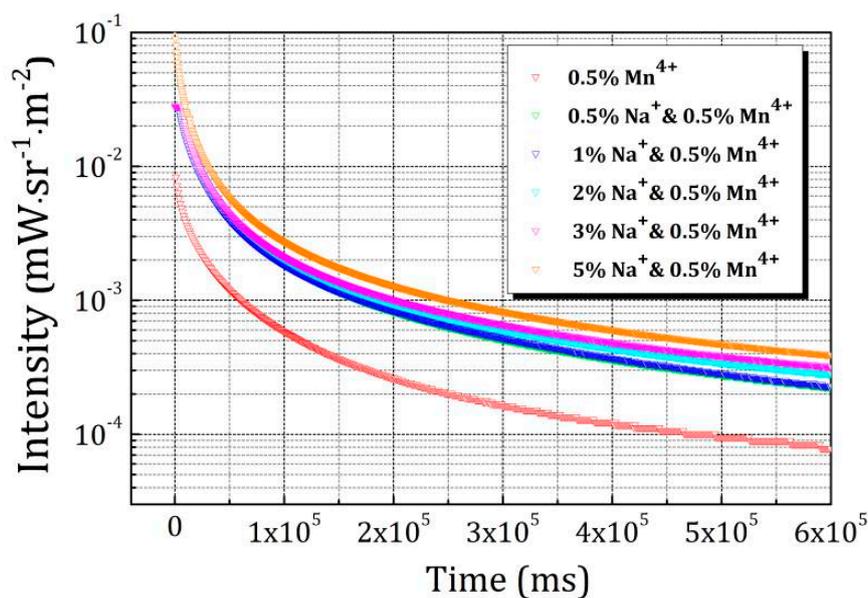
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40 **Figure S8.** Persistent luminescence decay curves of  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},y\text{Ba}^{2+}$  ( $y = 0.5\%, 1\%, 2\%, 3\%, \text{ and } 5\%$ )  
41 phosphors after 5 min of irradiation with a Xenon arc lamp. The red curve corresponds to  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+}$  as  
42 an intensity benchmark of persistent luminescence.



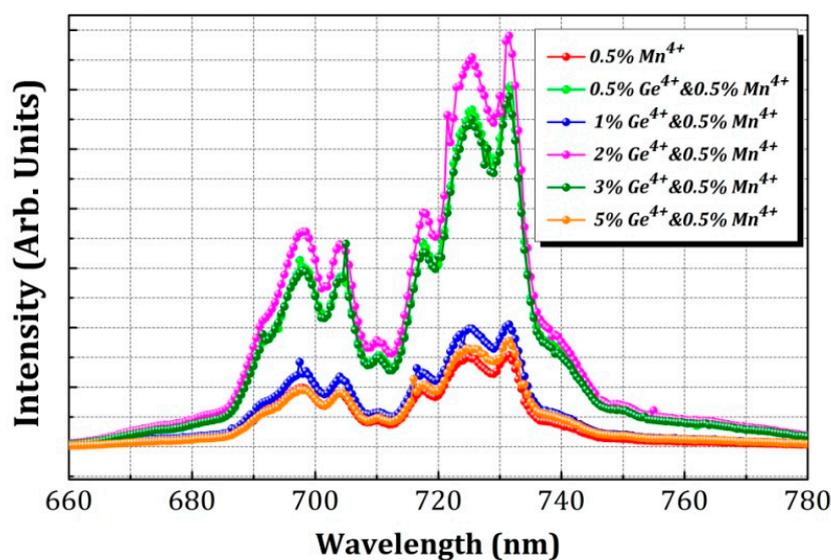
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44 **Figure S9.** Persistent luminescence decay curves of  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},y\text{Cl}^-$  ( $y = 0.5\%, 1\%, 2\%, 3\%, \text{ and } 5\%$ )  
45 phosphors after 5 min of irradiation with a Xenon arc lamp. The red curve corresponds to  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+}$  as  
46 an intensity benchmark of persistent luminescence.



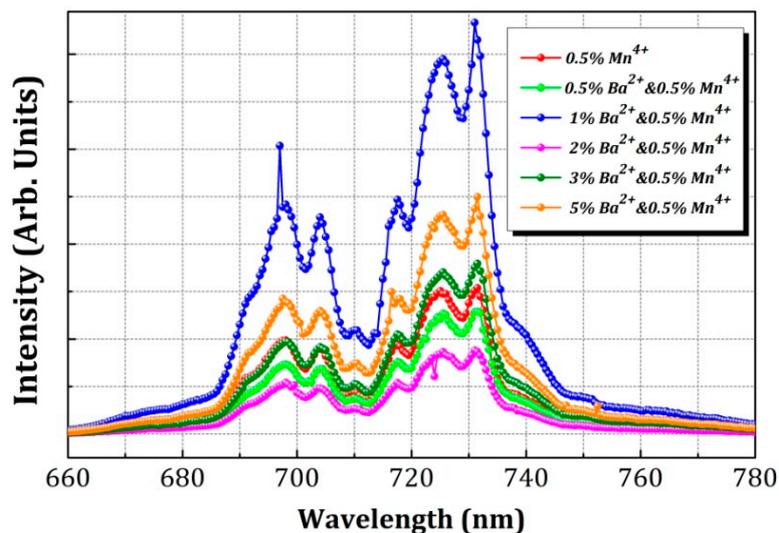
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48 **Figure S10.** Persistent luminescence decay curves of  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},y\text{Na}^+$  ( $y = 0.5\%, 1\%, 2\%, 3\%, \text{ and } 5\%$ )  
 49 phosphors after 5 min of irradiation with a Xenon arc lamp. The red curve corresponds to  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+}$  as  
 50 an intensity benchmark of persistent luminescence.



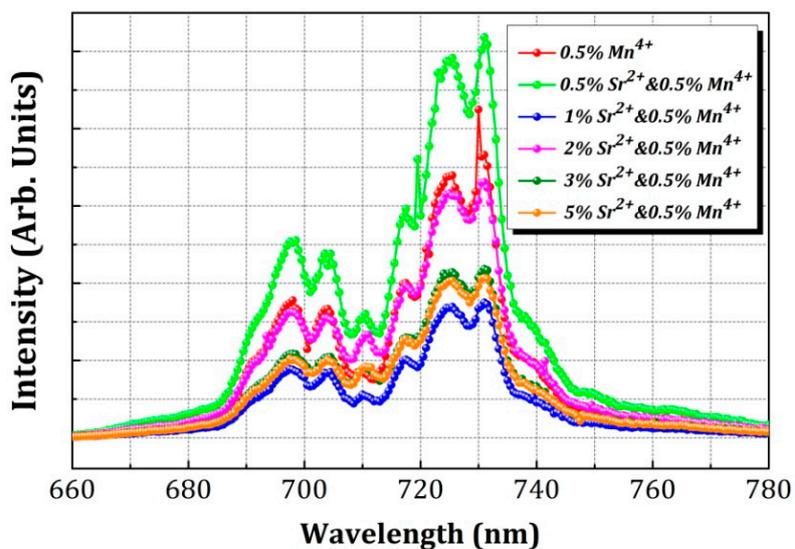
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52 **Figure S11.** Photoluminescence (PL) spectra of  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},0.5\%\text{Ge}^{4+}$ ,  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},1\%\text{Ge}^{4+}$ ,  
 53  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},2\%\text{Ge}^{4+}$ ,  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},3\%\text{Ge}^{4+}$  and  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+},5\%\text{Ge}^{4+}$  phosphors. All the PL  
 54 spectra ( $\lambda_{\text{ex}} = 335 \text{ nm}$ ) are in the range 660-780 nm and PL spectrum of  $\text{LaAlO}_3:0.5\%\text{Mn}^{4+}$  phosphor is shown in  
 55 red for comparison.



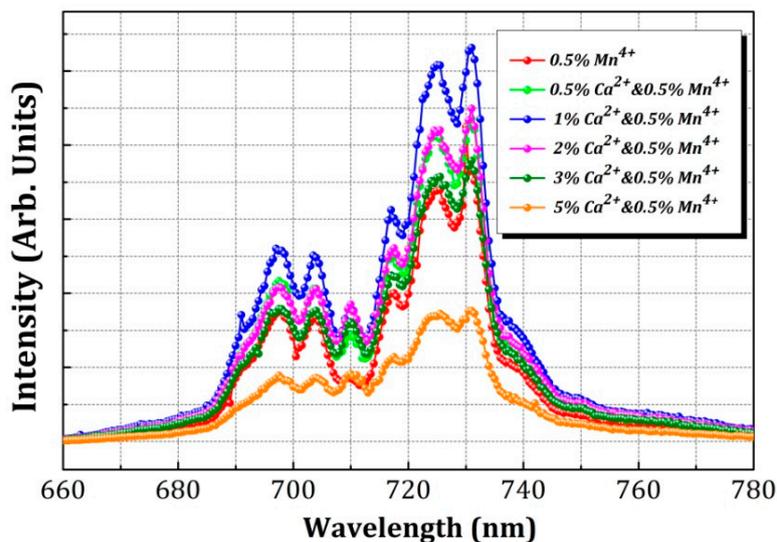
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57 **Figure S12.** Photoluminescence (PL) spectra of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,0.5%Ba<sup>2+</sup>, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,1%Ba<sup>2+</sup>,  
 58 LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,2%Ba<sup>2+</sup>, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,3%Ba<sup>2+</sup> and LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,5%Ba<sup>2+</sup> phosphors. All the PL spectra  
 59 ( $\lambda_{\text{ex}}= 335 \text{ nm}$ ) are in the range 660-780 nm and PL spectrum of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup> phosphor is in red for  
 60 comparison.



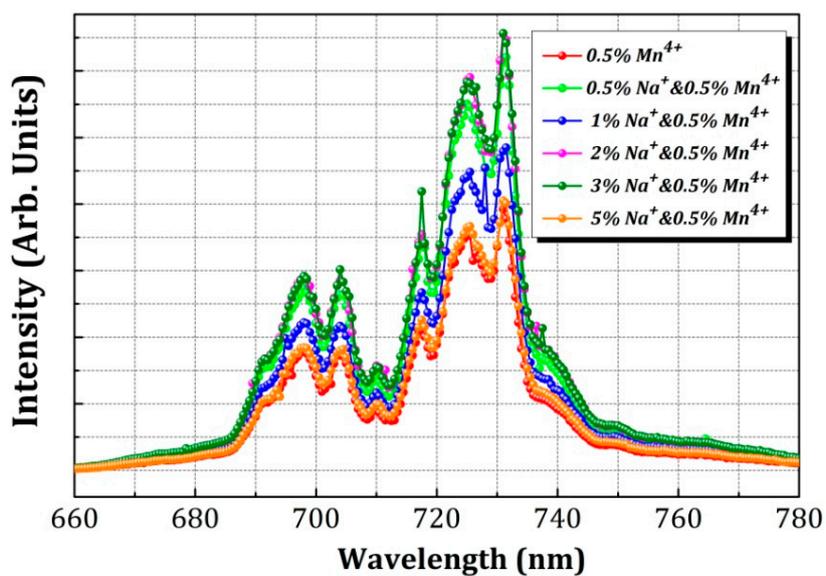
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62 **Figure S13.** Photoluminescence (PL) spectra of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,0.5%Sr<sup>2+</sup>, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,1%Sr<sup>2+</sup>,  
 63 LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,2%Sr<sup>2+</sup>, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,3%Sr<sup>2+</sup> and LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,5%Sr<sup>2+</sup> phosphors. All the PL spectra  
 64 ( $\lambda_{\text{ex}}= 335 \text{ nm}$ ) are in the range 660-780 nm and PL spectrum of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup> phosphor is in red for  
 65 comparison.



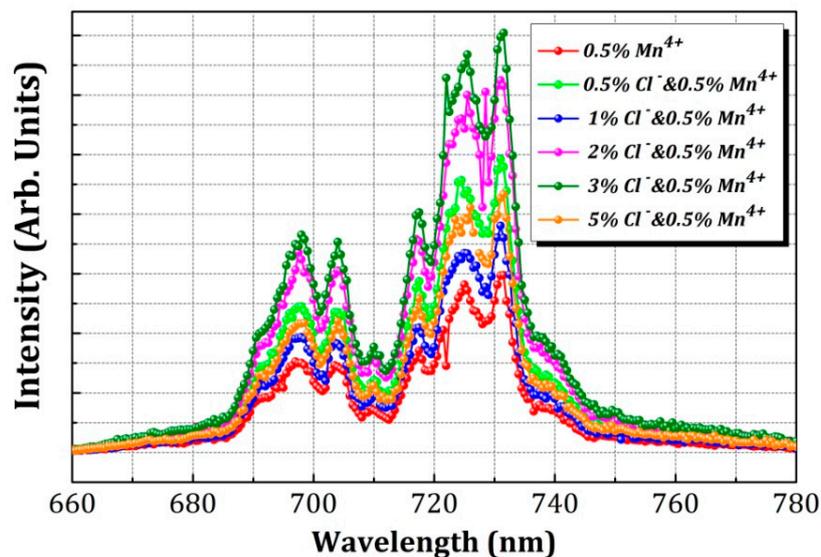
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67 **Figure S14.** Photoluminescence (PL) spectra of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,0.5%Ca<sup>2+</sup>, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,1%Ca<sup>2+</sup>,  
68 LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,2%Ca<sup>2+</sup>, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,3%Ca<sup>2+</sup> and LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,5%Ca<sup>2+</sup> phosphors. All the PL  
69 spectra ( $\lambda_{\text{ex}}=335$  nm) are in the range 660-780 nm and PL spectrum of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup> phosphor is in red for  
70 comparison.



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72 **Figure S15.** Photoluminescence (PL) spectra of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,0.5%Na<sup>+</sup>, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,1%Na<sup>+</sup>,  
73 LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,2%Na<sup>+</sup>, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,3%Na<sup>+</sup> and LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,5%Na<sup>+</sup> phosphors. All the PL spectra  
74 ( $\lambda_{\text{ex}}=335$  nm) are in the range 660-780 nm and PL spectrum of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup> phosphor is in red for  
75 comparison.



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77 **Figure S16.** Photoluminescence (PL) spectra of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,0.5%Cl, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,1%Cl,  
 78 LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,2%Cl, LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,3%Cl and LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup>,5%Cl phosphors. All the PL spectra  
 79 ( $\lambda_{\text{ex}} = 335 \text{ nm}$ ) are in the range 660-780 nm and PL spectrum of LaAlO<sub>3</sub>:0.5%Mn<sup>4+</sup> phosphor is in red for  
 80 comparison.

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**Table S1.** Ionic radius of some common dopant cations for the substitution on Al<sup>3+</sup> site

Dopant Cations	Coordination Number (Substitution on Octahedral Al <sup>3+</sup> Site)	Ionic Radius (pm)	$ R_{\text{DC}^{2+}} - R_{\text{Al}^{3+}}  / R_{\text{Al}^{3+}}$ (%)
Li <sup>+</sup>	VI	76	42.05
Na <sup>+</sup>	VI	102	90.65
K <sup>+</sup>	VI	138	157.94
Rb <sup>+</sup>	VI	152	184.11
Cs <sup>+</sup>	VI	167	212.14
Ag <sup>+</sup>	VI	115	114.95
Au <sup>+</sup>	VI	137	156.07
Be <sup>2+</sup>	VI	45	15.88
Mg <sup>2+</sup>	VI	72	34.57
Ca <sup>2+</sup>	VI	100	86.91
Sr <sup>2+</sup>	VI	118	120.56
Ba <sup>2+</sup>	VI	135	152.33
Cu <sup>2+</sup>	VI	73	36.44
Zn <sup>2+</sup>	VI	74	38.31
Cd <sup>2+</sup>	VI	95	77.57
Hg <sup>2+</sup>	VI	102	90.65
Sc <sup>3+</sup>	VI	74.5	39.25
Y <sup>3+</sup>	VI	90	68.22
B <sup>3+</sup>	VI	27	49.53

Ga <sup>3+</sup>	VI	62	15.88
In <sup>3+</sup>	VI	80	49.53
Ti <sup>4+</sup>	VI	60.5	13.08
Zr <sup>4+</sup>	VI	72	34.57
Mn <sup>4+</sup>	VI	53	0.93
Si <sup>4+</sup>	VI	40	25.23
Ge <sup>4+</sup>	VI	53	0.93
Sn <sup>4+</sup>	VI	69	28.97

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**Table S2.** Ionic radius of some common dopant cations for the substitution on La<sup>3+</sup> site

Dopant Cations	Coordination Number (Substitution on La <sup>3+</sup> Site)	Ionic Radius (pm)	$ R_{DC^{2+}} - R_{La^{3+}}  / R_{La^{3+}}$ (%)
Na <sup>+</sup>	XII	139	2.20
K <sup>+</sup>	XII	164	20.58
Rb <sup>+</sup>	XII	172	26.47
Cs <sup>+</sup>	XII	188	38.23
Ca <sup>2+</sup>	XII	134	1.47
Sr <sup>2+</sup>	XII	144	5.88
Ba <sup>2+</sup>	XII	161	18.38
Cd <sup>2+</sup>	XII	131	3.67

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