

## EXPERIMENTAL SECTION

### Sample preparation and optical measurements

$\text{LaMnAl}_{11}\text{O}_{19}:\text{x}\%\text{Cr}^{3+}$  samples were synthesized by high temperature solid phase method. The dopant Cr was added in the form of  $\text{Cr}_2\text{O}_3$  with different molar concentrations x % (x = 3,4,5,6,7). Using  $\text{La}_2\text{O}_3$  (99.9%) 、  $\text{Al}_2\text{O}_3$ (99.9%) 、  $\text{MnO}$  (99.9%) 、  $\text{Cr}_2\text{O}_3$  (99.9%) as raw materials. 5wt%  $\text{H}_3\text{BO}_3$  was added as a flux as flux, which can promote the uniformity of grain, improve the sintering density and beneficial to obtain pure phase. At first, the required raw materials are calculated by stoichiometric ratio and further weighed, transferred to agate mortar for uniform grinding, and then placed in an alumina crucible. All samples were sintered at 1600 °C for 4 h in a tube furnace at the atmosphere of  $\text{H}_2/\text{N}_2$ . After the mixture was cooled to ambient temperature, and the obtained products were ground again into fine powders for subsequent characterizations.

### Characterization

The X-ray diffraction (XRD) patterns were collected by the Bruker D8 Focus diffractometer with  $\text{Cu K}\alpha$  radiation ( $\lambda=1.5406 \text{ \AA}$ , 40 kV, 40 mA).  $\text{LaMnAl}_{11}\text{O}_{19}$  host was Rietveld refined using the GSAS program. The crystal structure was drawn by VESTA software. The morphology of  $\text{LaMnAl}_{11}\text{O}_{19}:\text{4\%Cr}^{3+}$  phosphor was observed by scanning electron microscopy (SEM, Hitachi Regulus SU8230). Furthermore, the photoluminescence (PL) and photoluminescence excitation (PLE) spectra were measured on the Edinburgh FS1000 spectrometer with a 150 W continuous Xenon lamp as the excitation source; while fluorescence decay curves were obtained by the same equipment with 5 W Xenon flash lamp. Finally, the temperature-dependent photoluminescence spectra in the range of 298–473 K were obtained by using the same FS5 spectrometer with a temperature controller (TAP-02). The EL spectrum of NIR LED device was measured by the OHSP-350M integrating sphere spectroradiometer system.

**Table S1** Relative difference in the ionic radius between the matrix cations and the dopant  $\text{Cr}^{3+}$  ion.

Ions	Radius	CN	$D_r$
$\text{La}^{3+}$	1.032	12	40.4%
$\text{Mn}^{2+}$	1.18	4	47.8%
$\text{Al}^{3+}$	0.39	4	57.6%
	0.48	5	28.1%
	0.535	6	14.9%
$\text{Cr}^{3+}$	0.615	6	

To figure out the specific position within the cation lattice that will be inhabited by the  $\text{Cr}^{3+}$ ion, the percent radius deviation ( $D_r$ ) can be obtained by the following equation:<sup>47</sup>

$$D_r = \frac{[R_m(\text{CN}) - R_d(\text{CN})]}{R_m(\text{CN})}$$

Herein, the coordination number is denoted by  $CN$ ,  $R_m(CN)$  represents the radius of the host cation, and  $R_d(CN)$  denotes the radius of the substituent ion.

**Table S2** Luminescence performance parameters of several  $\text{Cr}^{3+}$  doped phosphors

Hosts	$\lambda_{\text{ex}}$ [nm]	$\lambda_{\text{em}}$ [nm]	IQE/EQE [%]	FWHM [nm]	Refs.
$\text{BaSnSi}_3\text{O}_9$	449	806	16	162	[48]
$\text{BaZnAl}_{10}\text{O}_{17}$	562	691	-	11	[49]
$\text{Sr}_3\text{Sc}_4\text{O}_9$	468	761	87	120	[50]
$\text{Lu}_3\text{Ga}_5\text{O}_{12}$	438	706	20	40	[51]
$\text{La}_2\text{MgGeO}_6$	470	710	-	-	[52]

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LaMnAl <sub>11</sub> O <sub>19</sub>	426	706	55.6	43	This work
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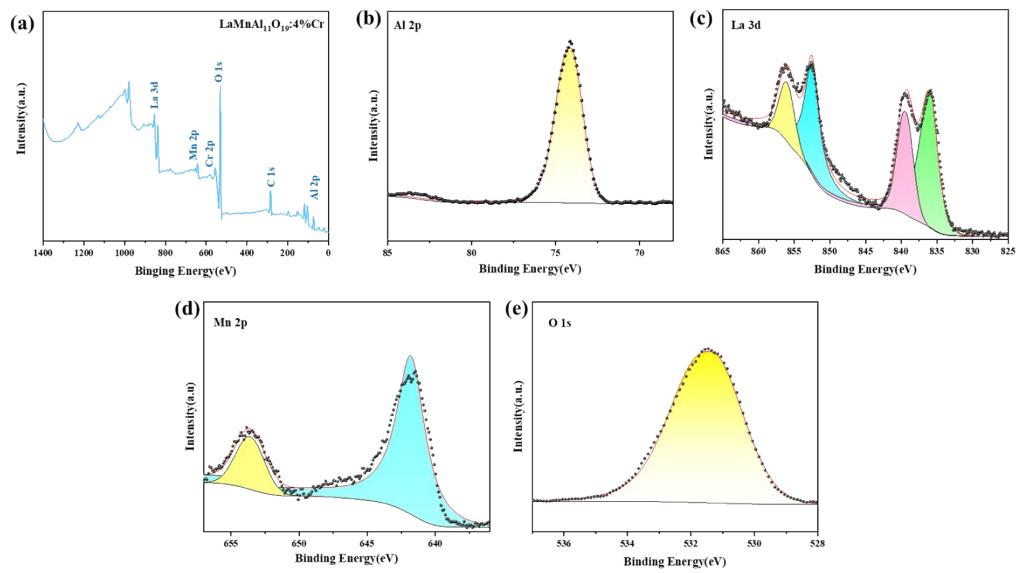


Figure S1. (a) XPS survey scan of LaMnAl<sub>11</sub>O<sub>19</sub>:4%Cr<sup>3+</sup>. (b) Narrow scan of Al 2p. (c) Narrow scan of La 3d. (d) Narrow scan of Mn 2p. (e) Narrow scan of O1s.