

**Achieving Broadband NIR Emission through Multiple Centers of Isolated Cr^{3+}
and Cr^{3+} - Cr^{3+} Ion Pairs Resulting from Phase Segregation in $\text{SrLa}_2\text{Ga}_2\text{O}_7$**

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Experimental section

The X-ray diffraction (XRD) analysis of the phosphor-related materials was performed using a Rigaku SmartLab SE X-ray diffractometer (Japan). The morphology of the samples was examined by scanning electron microscopy (SEM) utilizing a Zeiss Gemini 500. Elemental distribution map was generated via energy dispersive spectroscopy (EDS). The X-ray photoelectron spectroscopy (XPS) analysis of the phosphor was conducted using an ESCALAB XI+ X-ray photoelectron spectrometer. Electron paramagnetic resonance (EPR) measurements were carried out at room temperature using a Bruker EMX PLUS EPR spectrometer. The Raman spectrum of the phosphor was acquired employing a micro-laser confocal Raman

spectrometer, specifically the LabRAM HR Evolution. Additionally, the absorption spectrum of the phosphor was recorded with an Agilent Cary 7000 UMS universal spectrophotometer. The optical properties of the phosphor were evaluated using a UV-Vis-NIR fluorescence spectrometer (FLS 1000), including analyses such as excitation spectra (PLE), emission spectra (PL), temperature-dependent photoluminescence spectra, and fluorescence lifetime decay curves.

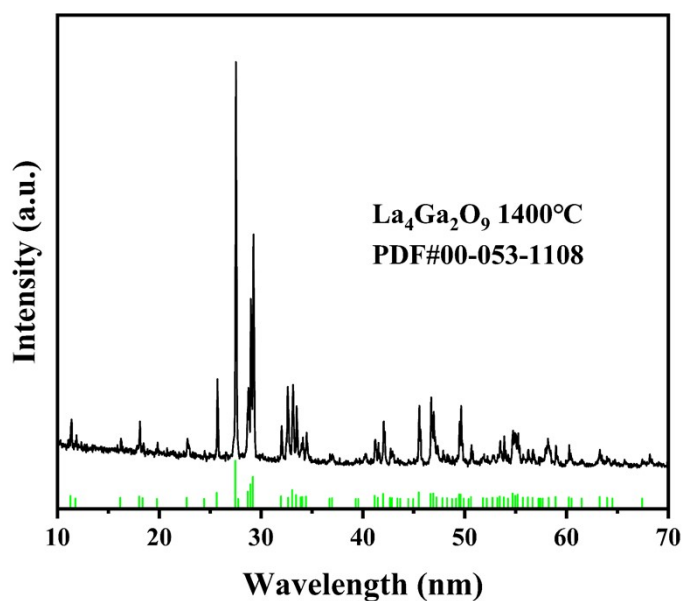


Figure S1. XRD patterns of LaGaO_4 powder samples.

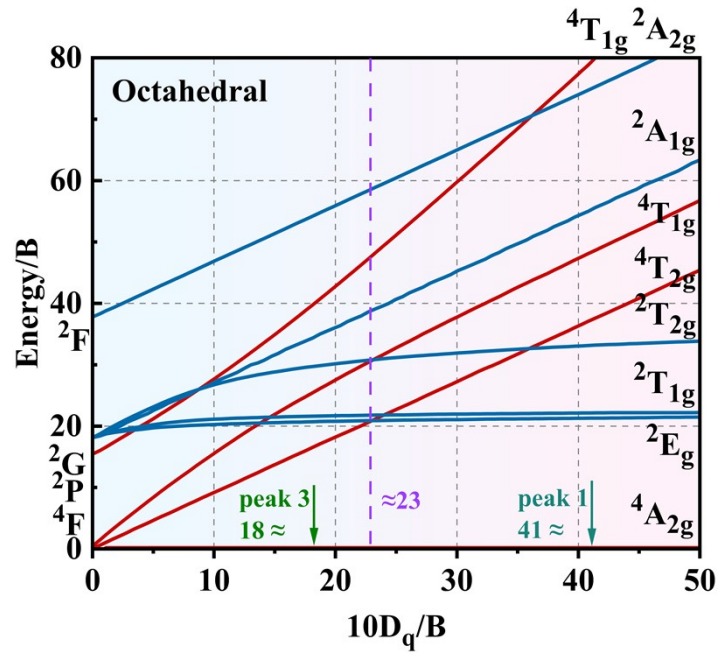


Figure S2. Tanabe-Sugano energy level diagram for Cr^{3+} ions ($3d_3$) lying in the octahedral ligand.

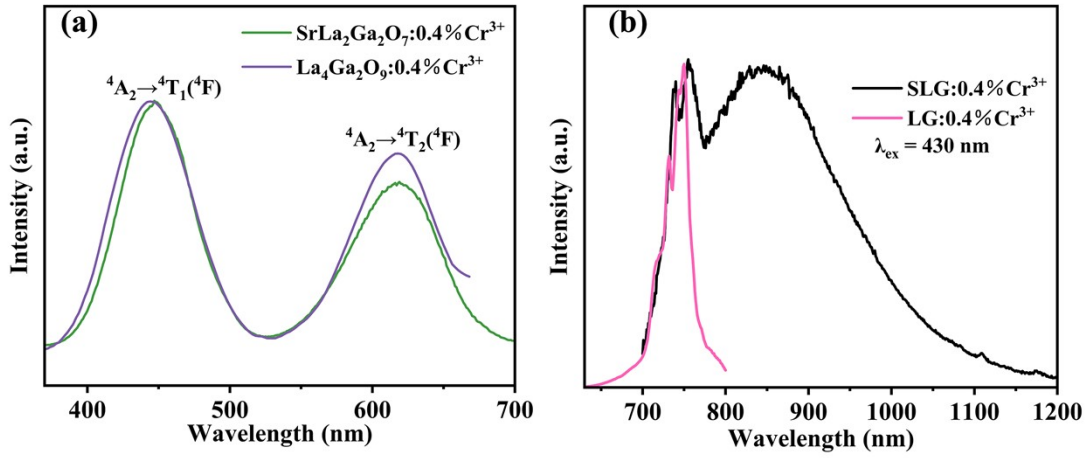


Figure S3. (a) PLE spectra recorded at RT, (b) PL spectra of SLG:0.4% Cr^{3+} and LG:0.4% Cr^{3+} phosphors.

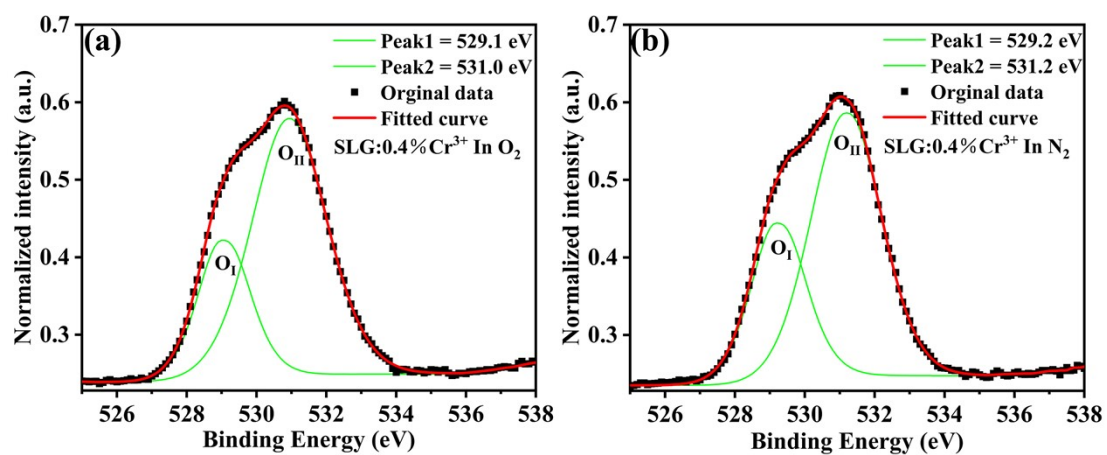


Figure S4. The XPS analysis of O 1s spectra for SLG:0.4%Cr³⁺ phosphors prepared

(a) in O₂ atmosphere and (b) in N₂ atmosphere, with fitted peaks (O_I and O_{II}).

Table S1. Luminescence performance parameters of several Cr³⁺-doped near-infrared fluorescent powders.

Hosts	$\lambda_{\text{ex}}(\text{nm})$	$\lambda_{\text{em}}(\text{nm})$	FWHM (nm)	IQE/EQE(%)	$I_{423\text{K}}/I_{298\text{K}}(\%)$	Refs.
K ₃ AlF ₆	430	750	100	\	\	1
Gd ₃ Sc ₂ Ga ₃ O ₁₂	460	754	90	60	100	2
Ba ₂ LaGa ₁₁ O ₂₀	468	738	133	\	34	3
SrGa ₂ O ₄	420	780	170	\	56	4
LiScSi ₂ O ₆	460	845	156	64.4/33.4	75	5
SrLaMgTaO ₆	470	810	180	37.8/14.18	80.4	6
Na ₃ Ga ₂ Li ₃ F ₁₂	430	764	117	48.7/7.79	71	7
Na ₃ ScF ₆	436	774	108	91.5/40.82	\	8
Na ₃ AlF ₆	420	720	95	75	60	9
K ₃ ScF ₆	432	770	150	71.7	87.3	10
K ₂ NaScF ₆	435	765	100	74	89.6	11
Sr ₂ MgWO ₆	371	763	125	\	\	12
Ca ₂ MgWO ₆	371	800	150	\	\	13
La ₂ MgZrO ₆	460	825	210	58	50	14
GdAl ₃ (BO ₃) ₄	420	733	80	91	88	15
La ₃ Sc ₂ Ga ₃ O ₁₂	490	800	145	35	60	16

LiSrAlF ₆	436	820	155	52.06	55.47	17
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