Supporting Information

Broadband Near-infrared (NIR) emission realized by crystal-field engineering of $Y_{3-x}Ca_xAl_{5-x}Si_xO_{12}:Cr^{3+}$ (x = 0 - 2.0) garnet phosphors

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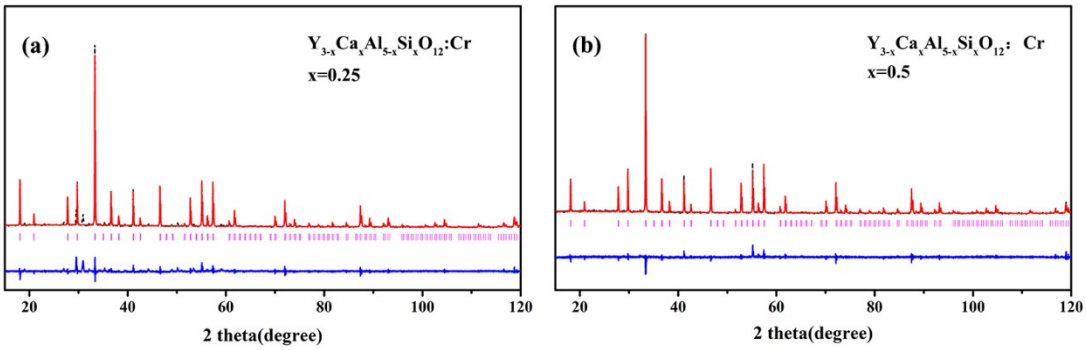

Figure S1. Rietveld refinement X-ray diffraction of $Y_{3-x}Ca_xAl_{5-x}Si_xO_{12}$:0.4%$Cr^{3+}$ (a) x = 0.25; (b) x = 0.5

Table S1. Crystallographic data of $Y_{3-x}Ca_xAl_{5-x}Si_xO_{12}$:0.4%$Cr^{3+}$ with different *x* values

Formula	x = 0.25	x = 0.5	x = 1
Crystal system		Cubic	
Space group		Iad	
Lattice parameters			
a = b = c (Å)	12.0122	11.9971	11.9913
α = β = γ (°)	90	90	90
Cell volume (Å³)	1733.263	1726.727	1724.234
R_p	5.12	5.11	5.48
X^2	4.24	1.88	1.93

Table S2. Atomic parameters of $Y_{3-x}Ca_xAl_{5-x}Si_xO_{12}$:0.4%$Cr^{3+}$ with different *x* values

Atom	**Y**			**Ca**			**Si**		
Wyck.	24c			24c			24d		
x Value	0.25	0.5	1	0.25	0.5	1	0.25	0.5	1
x	0.12500	0.12500	0.12500	0.12500	0.12500	0.12500	0.37500	0.37500	0.37500
y	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
z	0.25000	0.25000	0.25000	0.25000	0.25000	0.25000	0.25000	0.25000	0.25000
Occupancy	0.229	0.208	0.167	0.021	0.042	0.083	0.021	0.042	0.083
Atom	**Al1**			**Al2**			**O**		
Wyck.	16a			24d			96h		
x Value	0.25	0.5	1	0.25	0.5	1	0.25	0.5	1
x	0.00000	0.00000	0.00000	0.37500	0.37500	0.37500	-0.03462	-0.03898	-0.03649
y	0.00000	0.00000	0.00000	0.00000	0.00000	0.0000	0.04752	0.04498	0.04988
z	0.00000	0.00000	0.00000	0.25000	0.25000	0.25000	0.14942	0.14903	0.14875
Occupancy	0.167	0.167	0.167	0.229	0.208	0.167	1.000	1.000	1.000

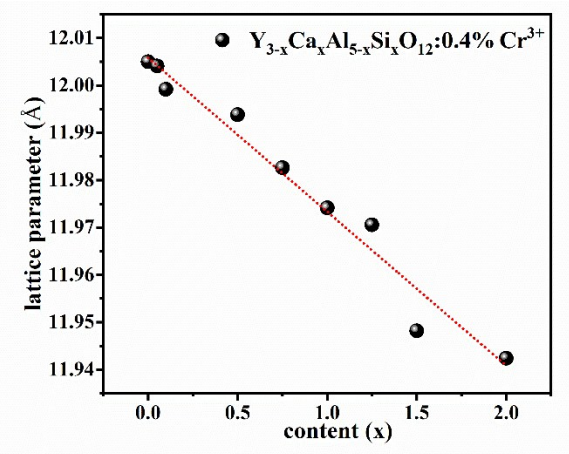

Figure S2. Lattice constant of $Y_{3-x}Ca_xAl_{5-x}Si_xO_{12}$:0.4%$Cr^{3+}$ (x = 0 - 2) after co-substitution, obtanined by structure refinement

Table S3. Distortion of ligands with different Ca^{2+}-Si^{4+} contents

ligand	Ca^{2+}-Si^{4+}content (x)	Distortion index	Effective coordination number
dodecahedron	0.25	0.03066	7.7051
	0.5	0.03020	7.7144
	1	0.01935	7.8889
octahedron	0.25	0	6
	0.5	0	6
	1	0	6
tetrahedron	0.25	0	4
	0.5	0	4
	1	0	4

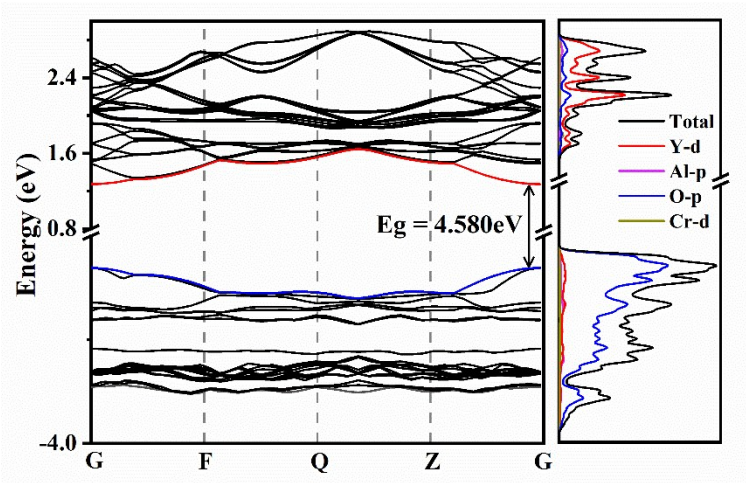

Figure S3. PDOS of $Y_3Al_5O_{12}$:Cr^{3+} (x = 0)

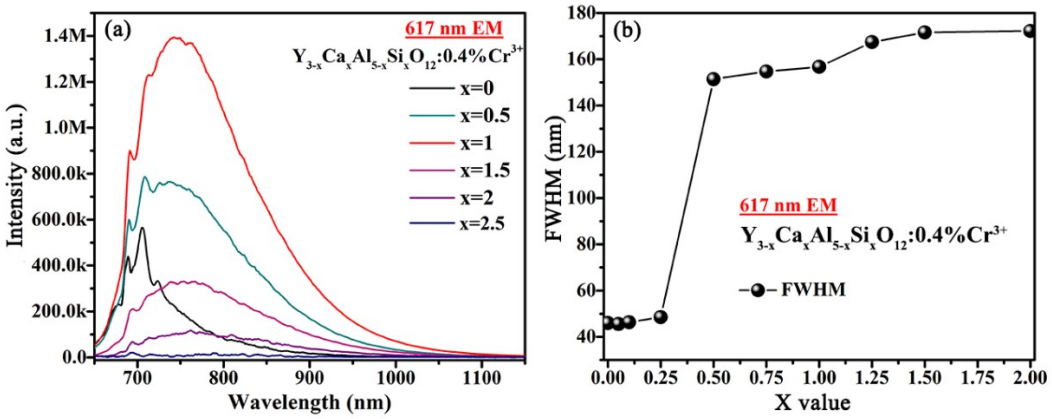

Figure S4. (a) Emission spectra and (b) FWHM of $Y_{3-x}Ca_xAl_{5-x}Si_xO_{12}$:0.4%$Cr^{3+}$ at room temperature under 617 nm excitation

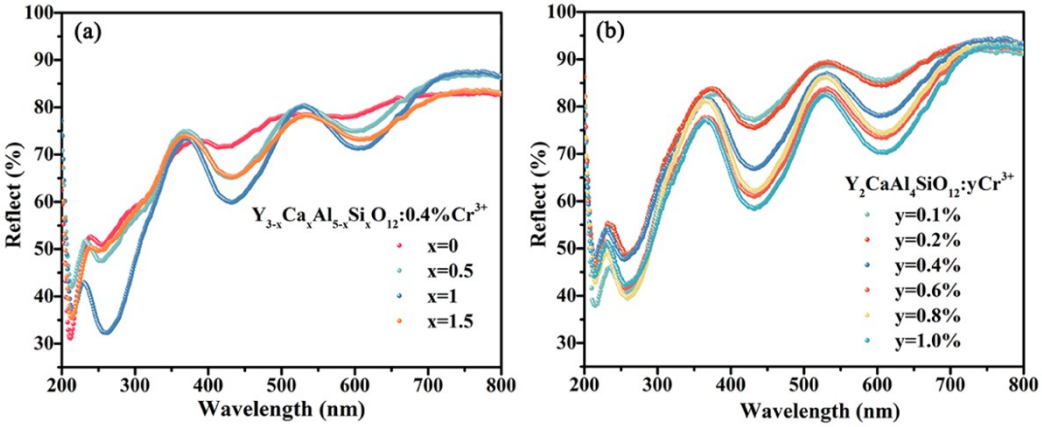

Figure S5. Diffuse reflection spectra of (a) $Y_{3-x}Ca_xAl_{5-x}Si_xO_{12}$:0.4%$Cr^{3+}$ (x = 1 - 1.5) and (b) $Y_2CaAl_4SiO_{12}$:yCr^{3+} (y = 0.1 - 1.0%) at room temperature

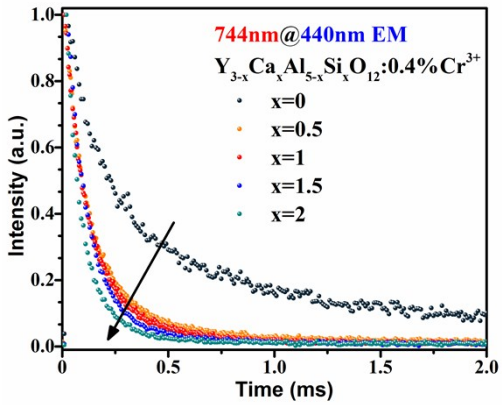

Figure S6. Decay time curves of $Y_{3-x}Ca_xAl_{5-x}Si_xO_{12}$:0.4%$Cr^{3+}$ at room temperature

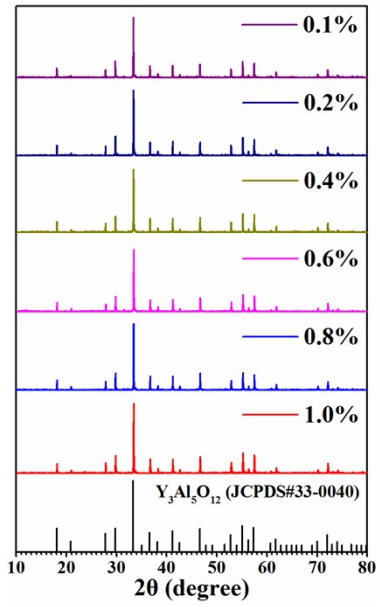

Figure S7. XRD pattern of $Y_2CaAl_4SiO_{12}$:yCr^{3+} and JCPDS reference data of $Y_3Al_5O_{12}$

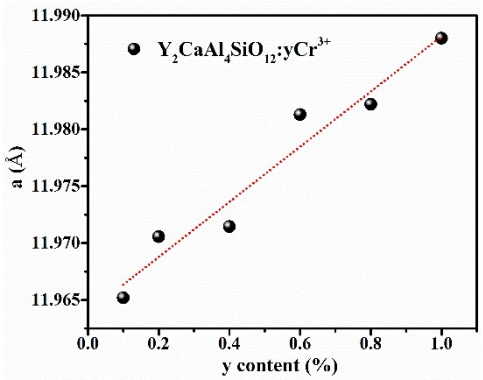

Figure S8. Variation of the lattice constant of $Y_2CaAl_4SiO_{12}$:yCr^{3+} phosphor powders under 440nm excitation

Table S3. The chemical formula for the material in Figure 4

Abbreviation	Chemical formula
LaSGG	$La_3Sc_2Ga_3O_{12}$
CAGG	$Ca_3Al_2Ge_3O_{12}$
GSGG	$Gd_3Sc_2Ga_3O_{12}$
CYMGG	$CaY_2Mg_2Ge_3O_{12}$
YSGG	$Y_3Sc_2Ga_3O_{12}$
YGG	$Y_3Ga_5O_{12}$
GGG	$Gd_3Ga_5O_{12}$
LuSGG	$Lu_3Sc_2Ga_3O_{12}$
YAG	$Y_3Al_5O_{12}$
LuGG	$Lu_3Al_5O_{12}$
LuAG	$Lu_3Al_5O_{12}$

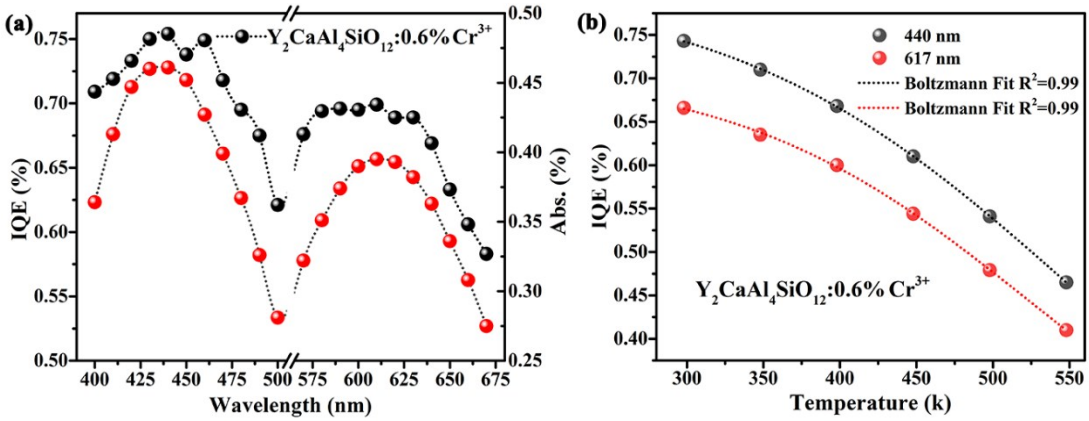

Figure S9. IQE as a function of (a) excitation wavelength and (b) temperature for the $Y_2CaAl_4SiO_{12}$:0.6%Cr^{3+} phosphor

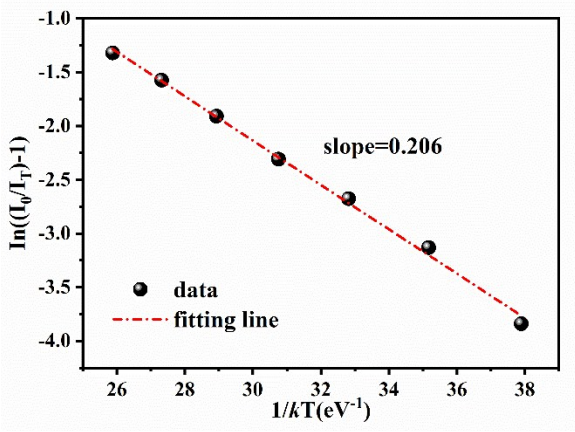

Figure S10. Activation energy using the modified Arrhenius equation for the $Y_2CaAl_4SiO_{12}$:Cr phosphor