Supporting Information

Blue-light activated green emitting $BaY_2Al_2Ga_2SiO_{12}$:Ce^{3+}, Bi^{3+}: Efficient persistent luminescence phosphor for AC-LEDs

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Tables and Table Captions

Table S1. The formation energies of 12 equivalent structures by Bi^{3+} doping.

System	Formation energy (eV)
Ba-Al-Al	2.671
Ba-Al-Ga	2.586
Ba-Ga-Al	3.124
Ba-Ga-Ga	2.743
Ba-Si-Al	3.265
Ba-Si-Ga	2.592
Y-Al-Al	1.458
Y-Al-Ga	2.183
Y-Ga-Al	2.074
Y-Ga-Ga	1.649
Y-Si-Al	1.762
Y-Si-Ga	1.836

Table S2. The formation energies of Bi-doped Ba sites and Y sites.

System	Total energy (eV)	Formation energy (eV)
Pure matrix	-1038.873792	/
Doping Ba site	-1034.624087	2.843
Doping Y site	-1030.659895	1.961

Table S3. Refined structural data of BYAGSO, BYAGSO:0.04Ce^{3+}, and BYAGSO:0.04Ce^{3+}, 0.01Bi^{3+}.

Formula	BYAGSO	BYAGSO:0.04Ce^{3+}	BYAGSO:0.04Ce^{3+}, 0.01Bi^{3+}
crystal system	cubic	cubic	cubic
space group	*Ia3d*(230)	*Ia3d*(230)	*Ia3d*(230)
a (Å)	12.1468	12.1473	12.1483
b (Å)	12.1468	12.1473	12.1483
c (Å)	12.1468	12.1473	12.1483
$\alpha = \beta = \gamma$ (deg)	90	90	90
Z	8	8	8
V ($Å^3$)	1792.185	1792.419	1792.868
R_p	0.0521	0.0524	0.0530
R_{wp}	0.0719	0.0742	0.0740
χ^2	2.518	2.847	2.698

Table S4. The atom positions, fraction factors, and thermal vibration parameters of BYAGSO, BYAGSO:0.04Ce^{3+}, and BYAGSO:0.04Ce^{3+}, 0.01Bi^{3+}.

atom	site	x	y	z	Occupancy	U_{iso}
Ba1	24c	0.125	0	0.25	0.333	0.02680
Y1	24c	0.125	0	0.25	0.666	0.02680
Al1	16a	0	0	0	0.591	0.00610
Ga1	16a	0	0	0	0.409	0.00610
Al2	24d	0.375	0	0.25	0.273	0.09059
Ga2	24d	0.375	0	0.25	0.394	-0.04059
Si1	24d	0.375	0	0.25	0.333	-0.04059
O1	96h	-0.04379	0.02761	0.15388	1.000	-0.01400

atom	site	x	y	z	Occupancy	U_{iso}
Ba1	24c	0.125	0	0.25	0.333	-0.00055
Y1	24c	0.125	0	0.25	0.653	0.05055
Ce1	24c	0.125	0	0.25	0.013	-0.01145
Al1	16a	0	0	0	0.591	0.00213
Ga1	16a	0	0	0	0.409	0.00213
Al2	24d	0.375	0	0.25	0.273	0.06694
Ga2	24d	0.375	0	0.25	0.394	-0.01694
Si1	24d	0.375	0	0.25	0.333	-0.01694
O1	96h	-0.04447	0.0307	0.1514	1.000	0.01760

atom	site	x	y	z	Occupancy	U_{iso}
Ba1	24c	0.125	0	0.25	0.333	-0.02416
Y1	24c	0.125	0	0.25	0.650	0.07416
Bi1	24c	0.125	0	0.25	0.003	0.80000
Ce1	24c	0.125	0	0.25	0.013	-0.01044
Al1	16a	0	0	0	0.591	0.01102
Ga1	16a	0	0	0	0.409	0.01102
Al2	24d	0.375	0	0.25	0.273	0.07532
Ga2	24d	0.375	0	0.25	0.394	-0.02532
Si1	24d	0.375	0	0.25	0.333	-0.02532
O1	96h	-0.04435	0.02961	0.15361	1.000	-0.01893

Table S5. PersL phosphors for AC-LED applications

Phosphor	color	Percent flicker
$BaLu_2Al_2Ga_2SiO_{12}$:Ce^{3+}, Bi^{3+}	blue	55.04%
$Gd_3Al_2Ga_3O_{12}$:Ce^{3+}	green	69%
$Mg_3Y_2Ge_{2.7}Si_{0.3}O_{12}$:$Ce^{3+}$	yellow	71.70%
$Lu_{1.98}CaMg_2Si_1Ge_2O_{12}$:$Ce^{3+}$	yellow	64.10%

Figures and Figure Captions

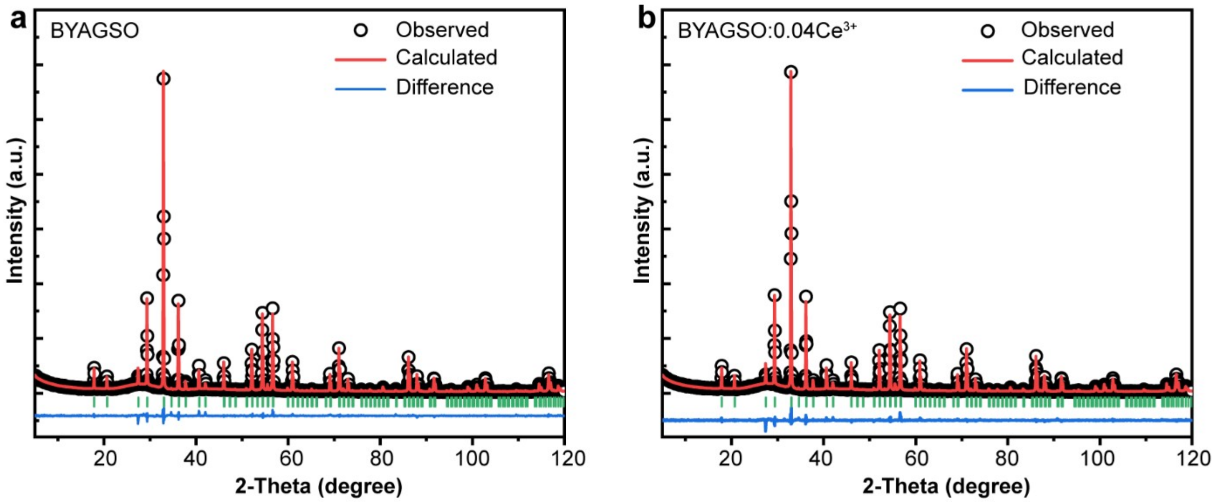

Figure S1. Rietveld refinements of BYAGSO (a) and BYAGSO:0.04Ce^{3+} (b).

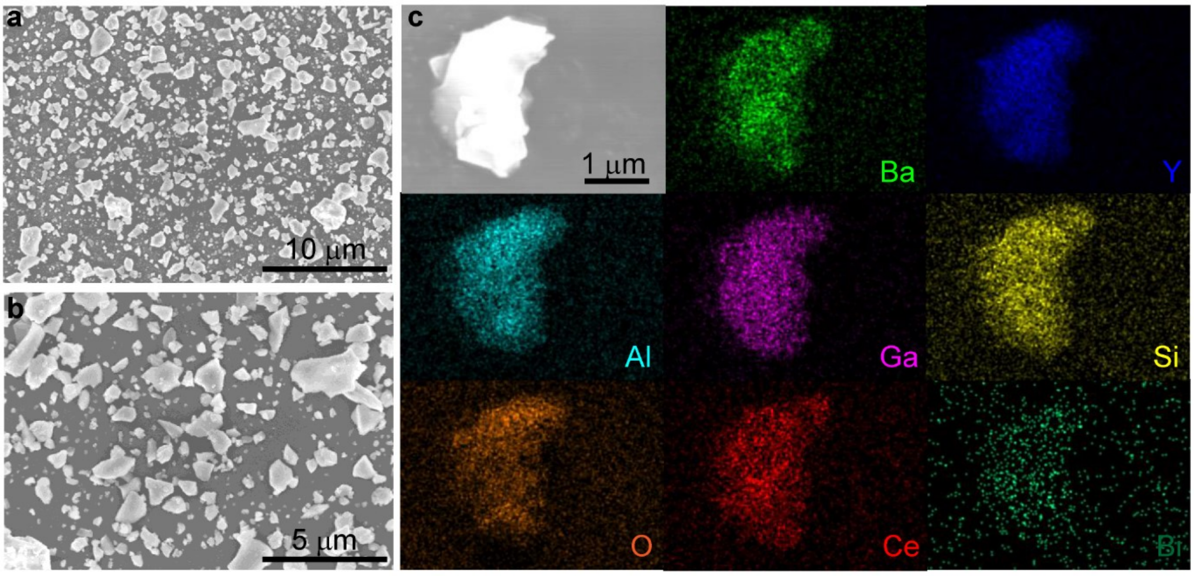

Figure S2. (a) - (b) SEM images of BYAGSO:0.04Ce^{3+}, 0.01Bi^{3+}. (c) Elemental mapping images of BYAGSO:0.04Ce^{3+}, 0.01Bi^{3+}.

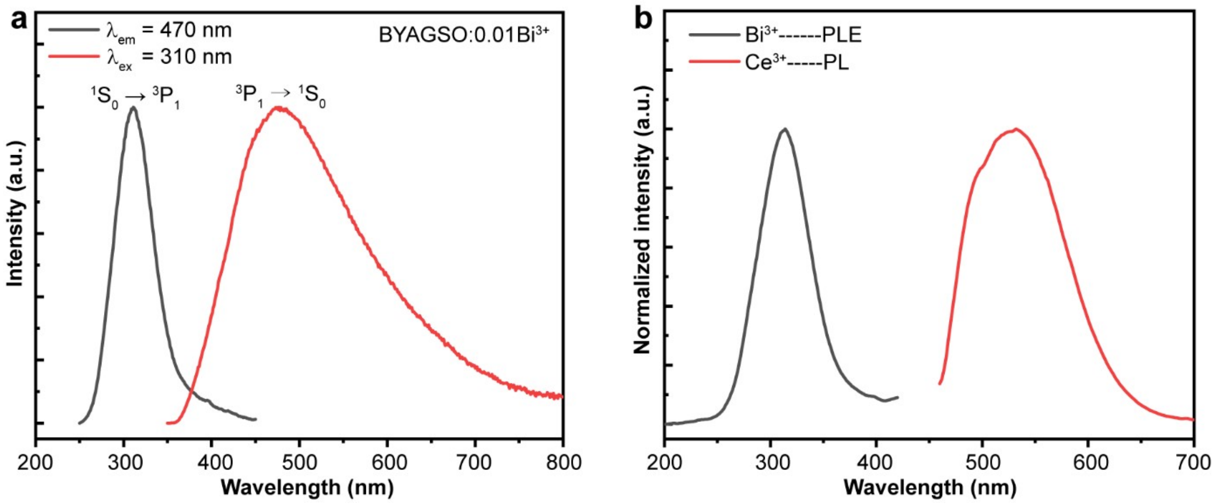

Figure S3. (a) PLE and PL spectra of BYAGSO:0.01Bi^{3+}. (b) PLE spectrum of BYAGSO:0.01Bi^{3+} and PL spectrum of BYAGSO:0.04Ce^{3+}.

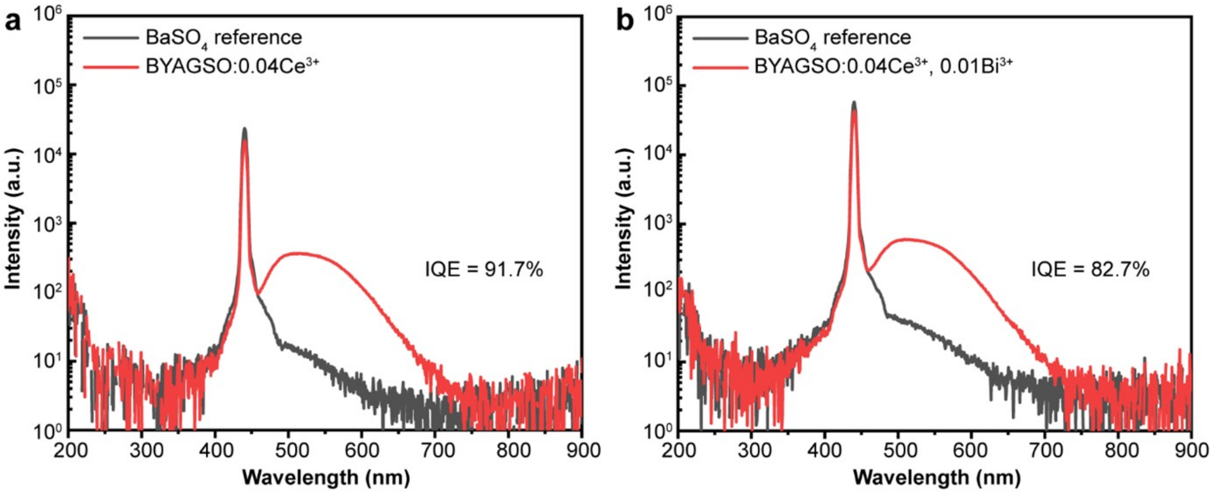

Figure S4. IQEs of BYAGSO:0.04Ce^{3+} (a) and BYAGSO:0.04Ce^{3+}, 0.01Bi^{3+} (b).

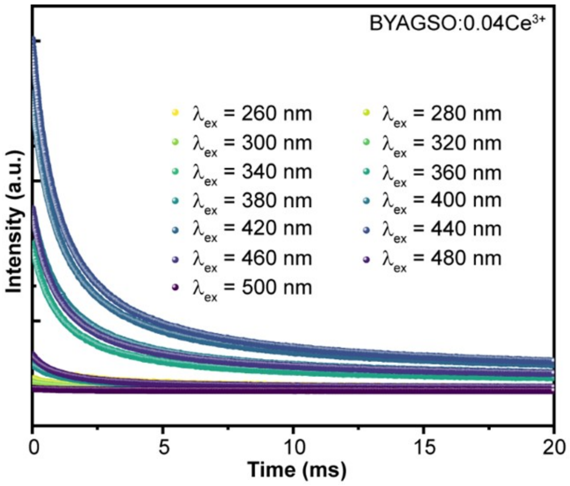

Figure S5. The representative PersL decay versus the excitation wavelength.

Calculation method

First-principles calculations are performed by vienna ab initio simulation package (VASP).[1,2] The generalized gradient approximation(GGA) of Perdew-Burke-Ernzerhof (PBE) is used to describe the exchange-correlation functional. The cut-off energy for the plane wave basis is set to 400 eV and a 2×2×2 Monkhorst-pack mesh is employed. All the structures were fully relaxed (atomic position) up to 10^{-5} eV /Å force minimization and max force of 0.01 eV/ Å.

We calculated the formation energies (E_f) of all these defects in the doped system according to the formula:[3]

$$E_f = E_{df} - E_{ini} + \sum_i n_i \mu_i$$

where E_{ini} and E_{df} are the total energies of the supercell before and after doping, respectively. *ni* is the number of elements added or removed during the formation of the defects, *ui* is the chemical potential of the element. For the metal cations, the chemical potentials were obtained from the calculations of the respective bulk materials. For the oxygen atom, the chemical potentials were obtained from the calculations of oxygen gas.

References

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2 G. Kresse, D. Joubert, From ultrasoft pseudopotentials to the projector augmented-wave method, *Phys. Rev. B*, 1999, **59**, 1758-1775.

3 W. Wu, W.Y. Cong, C. Guan, H. Sun, R. Yin, G. Yu, Y.B. Lu, Investigation of the Mn dopant-enhanced photoluminescence performance of lead-free $Cs_2AgInCl_6$ double perovskite crystals, *Phys. Chem. Chem. Phys.*, 2020, **22**, 1815-1819.