

**Simultaneous Broadening and Enhancement of Cr³⁺ Photoluminescence in
Ba₃ZrTa₄O₁₅ by Chemical Unit Cosubstitution**

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Samples	Atom	x	y	z	Multiplicity & Wyckoff	Occ	Uiso
$\text{Ba}_3\text{ZrTa}_4\text{O}_{15}:\text{Cr}^{3+}$	Ba1	0.00000	0.00000	0.39378	2a	1	0.012
	Ba2	0.17165	0.67165	0.40490	4c	1	0.028
	Zr1	0.00000	0.50000	0.87563	2b	0.2	0.002
	Ta1	0.00000	0.50000	0.87563	2b	0.8	0.002
	Zr2	0.07552	0.21384	0.89881	8d	0.2	0.007
	Ta2	0.07552	0.21384	0.89881	8d	0.8	0.007
	O1	0.00000	0.50000	0.45260	2b	1	0.079
	O2	0.28106	0.78106	0.92480	4c	1	0.022
	O3	0.07270	0.21049	0.36082	8d	1	0.004
	O4	0.34297	0.01190	0.83636	8d	1	0.044
	O5	0.14239	0.06733	2.23864	8d	1	0.035
$\text{Ba}_3\text{Al}_{0.5}\text{Ta}_{4.5}\text{O}_{15}:\text{Cr}^{3+}$	Ba1	0.00000	0.00000	0.40240	2a	1	0.029
	Ba2	0.1722	0.67220	0.37949	4c	1	0.030
	Al1	0.00000	0.50000	0.87605	2b	0.2	0.011
	Ta1	0.00000	0.50000	0.87605	2b	0.8	0.011
	Al2	0.07508	0.21315	0.89932	8d	0.2	0.002
	Ta2	0.07508	0.21315	0.89932	8d	0.8	0.002
	O1	0.00000	0.50000	0.46334	2b	1	0.062
	O2	0.28163	0.78163	0.95045	4c	1	0.025
	O3	0.07258	0.20606	0.34494	8d	1	0.006
	O4	0.35763	0.01105	0.86803	8d	1	0.065
	O5	0.15222	0.06925	0.87630	8d	1	0.010

Table S2. Distances in octahedra of $\text{Ba}_3\text{ZrTa}_4\text{O}_{15}:\text{Cr}^{3+}$ and $\text{Ba}_3\text{Al}_{0.5}\text{Ta}_{4.5}\text{O}_{15}:\text{Cr}^{3+}$.

Octahedr a	$\text{Ba}_3\text{ZrTa}_4\text{O}_{15}:\text{Cr}^{3+}$	$\text{Ba}_3\text{Al}_{0.5}\text{Ta}_{4.5}\text{O}_{15}:\text{Cr}^{3+}$	Octahedra	$\text{Ba}_3\text{ZrTa}_4\text{O}_{15}:\text{Cr}^{3+}$	$\text{Ba}_3\text{Al}_{0.5}\text{Ta}_{4.5}\text{O}_{15}:\text{Cr}^{3+}$
Ba1O ₁₂	2.82665(4)	2.75362(4)	Ba1O ₁₅	2.66585(3)	2.57820(3)
	2.82665(4)	2.75362(4)		2.66585(3)	2.82079(4)
	2.82665(4)	2.75362(4)		2.74315(3)	2.82079(4)
	2.82665(4)	2.75362(4)		2.85641(4)	2.88340(4)
	2.82775(4)	2.81283(4)		3.04814(4)	2.88340(4)
	2.82775(4)	2.81283(4)		3.04814(4)	2.97631(4)
	2.82775(4)	2.81283(4)		3.08500(5)	3.07689(5)
	2.82775(4)	2.81283(4)		3.27080(4)	3.22218(4)
	2.89070(4)	2.95387(4)		3.27080(4)	3.22218(4)
	2.89070(4)	2.95387(4)		3.28468(5)	3.23687(5)
	2.89070(4)	2.95387(4)		3.28468(5)	3.23687(5)
	2.89070(4)	2.95387(4)		3.37916(4)	3.23757(4)
				3.37916(4)	3.23757(4)
				3.44562(5)	3.43675(5)
		3.44562(5)	3.43675(5)		
(Zr/Al/Ta) 1O ₆	1.68868(3)	1.62910(3)	(Zr/Al/Ta) 2O ₆	1.84504(4)	1.76154(4)
	2.00367(3)	1.79402(3)		1.99362(3)	1.97001(3)
	2.00367(3)	1.79402(3)		2.00192(3)	2.00509(3)
	2.00367(3)	1.79402(3)		2.01176(3)	2.05293(3)
	2.00367(3)	1.79402(3)		2.06841(3)	2.11648(4)
	2.30318(4)	2.31822(5)		2.14838(4)	2.19035(4)

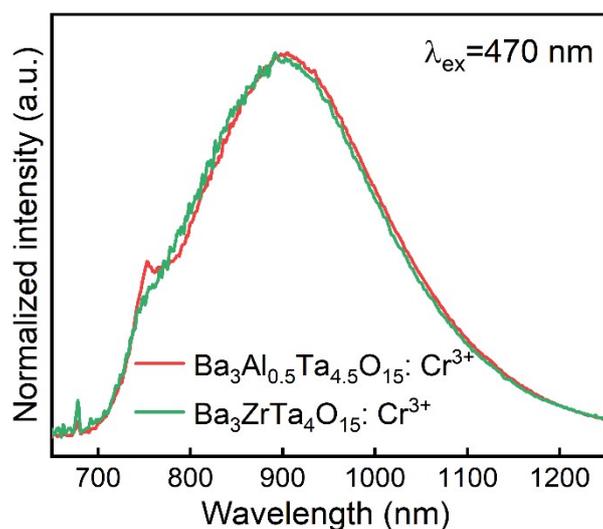


Figure S1. Normalize emission spectra of $\text{Ba}_3\text{ZrTa}_4\text{O}_{15}: 0.04\text{Cr}^{3+}$ and $\text{Ba}_3\text{Al}_{0.5}\text{Ta}_{4.5}\text{O}_{15}: 0.04\text{Cr}^{3+}$. $\text{Ba}_3\text{ZrTa}_4\text{O}_{15}: 0.04\text{Cr}^{3+}$ and $\text{Ba}_3\text{Al}_{0.5}\text{Ta}_{4.5}\text{O}_{15}: 0.04\text{Cr}^{3+}$ phosphors were synthesised via a solid-state reaction method. High-purity starting materials, including BaCO_3 (AR), Al_2O_3 (AR), ZrO_2 (AR), Cr_2O_3 (AR), and Ta_2O_5 (AR), were weighed according to the stoichiometric ratios of the respective chemical formulae. The thoroughly mixed reactants were sintered at 1400°C for 5 h. The resulting products were subsequently cooled to room temperature, ground into fine powders, and ultimately obtained as the final phosphor materials.

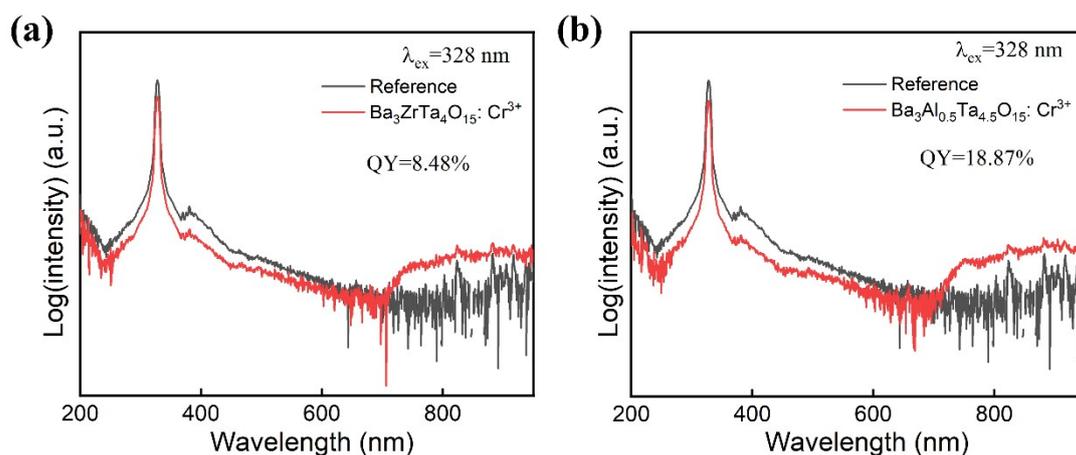


Figure S2. The quantum yield (QY) spectra of $\text{Ba}_3\text{ZrTa}_4\text{O}_{15}: \text{Cr}^{3+}$ and $\text{Ba}_3\text{Al}_{0.5}\text{Ta}_{4.5}\text{O}_{15}: \text{Cr}^{3+}$.

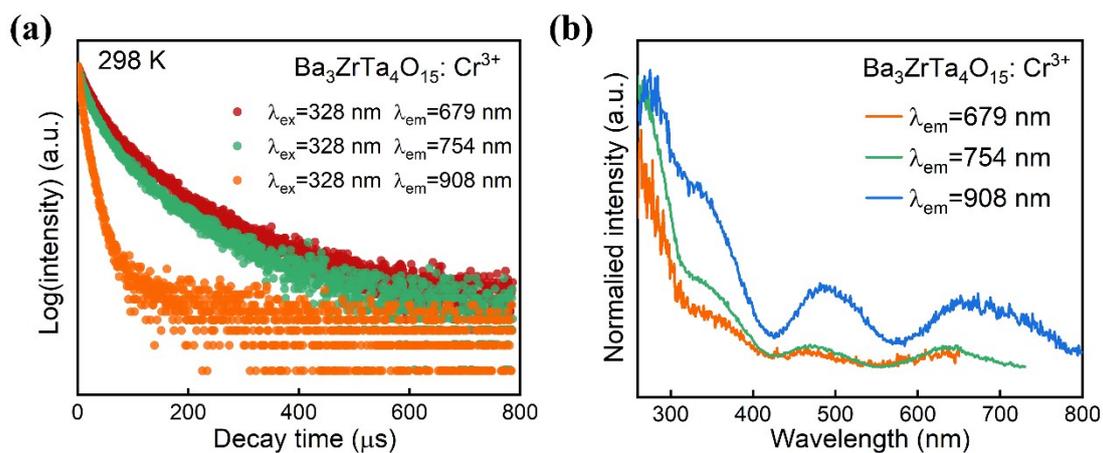


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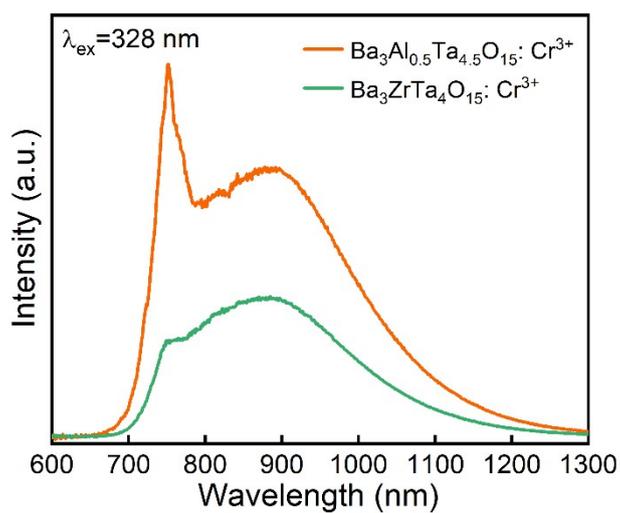


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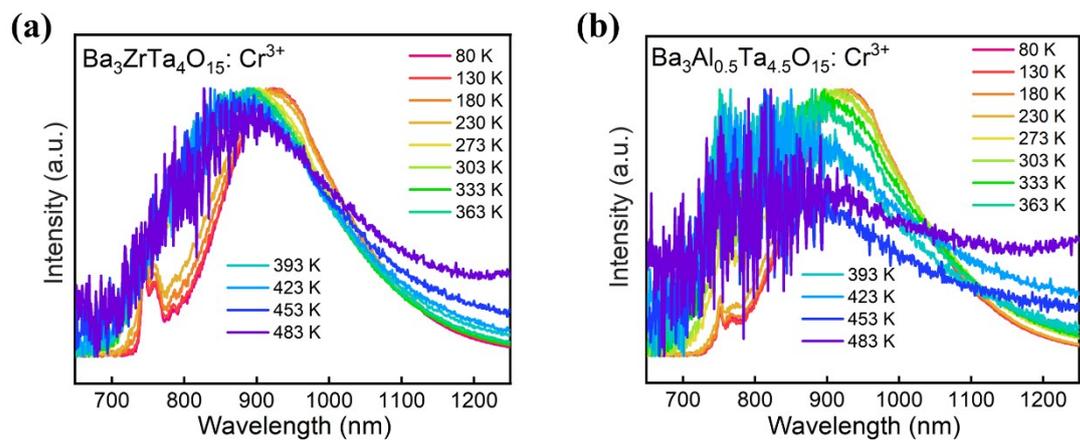


Figure S5. Normalized temperature-dependent emission spectra of (a) $\text{Ba}_3\text{ZrTa}_4\text{O}_{15}:\text{Cr}^{3+}$ and (b) $\text{Ba}_3\text{Al}_{0.5}\text{Ta}_{4.5}\text{O}_{15}:\text{Cr}^{3+}$.