Electronic Supporting Information

Structural Modulation Induced Intensity Enhancement of Full Color Spectra: A Case of $Ba_3ZnTa_{2-x}Nb_xO_9:Eu^{3+}$ Phosphors

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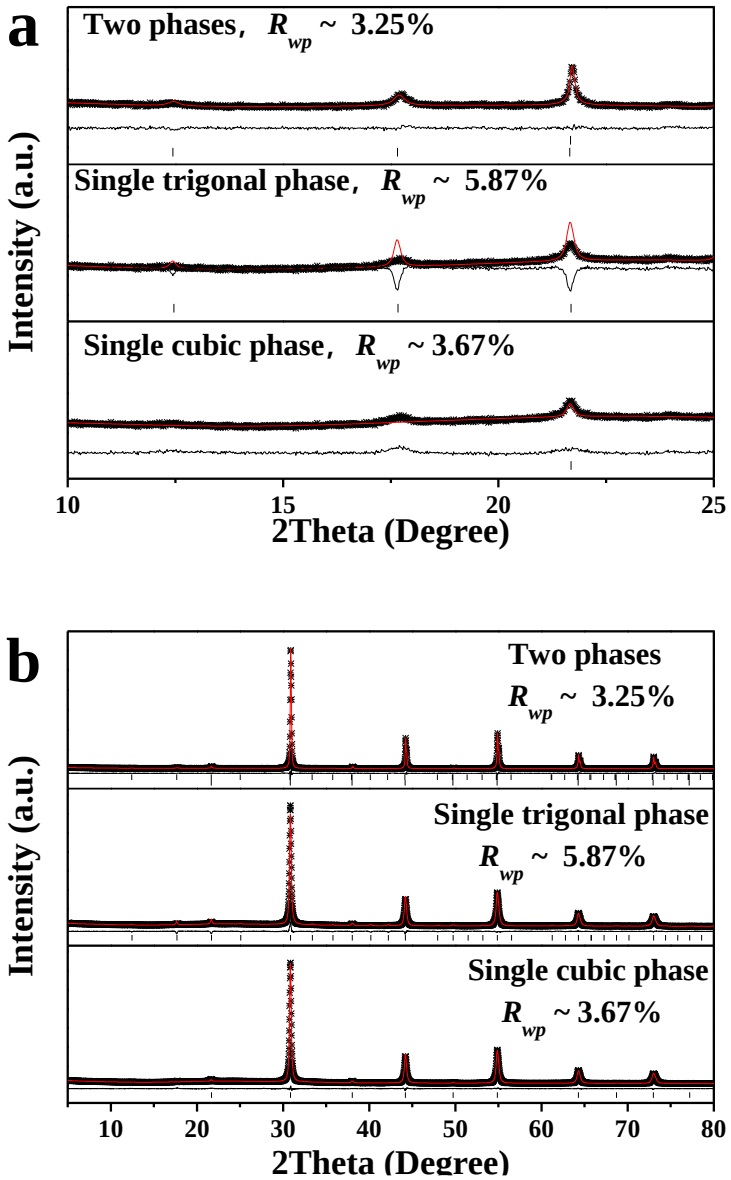


Figure S1. (a) Comparison of Rietveld refinements using single cubic/trigonal phase and both phases for XRD data of BZT obtained at 1100 ℃. (b) And its corresponding enlargements in 10-25 °2θ range.

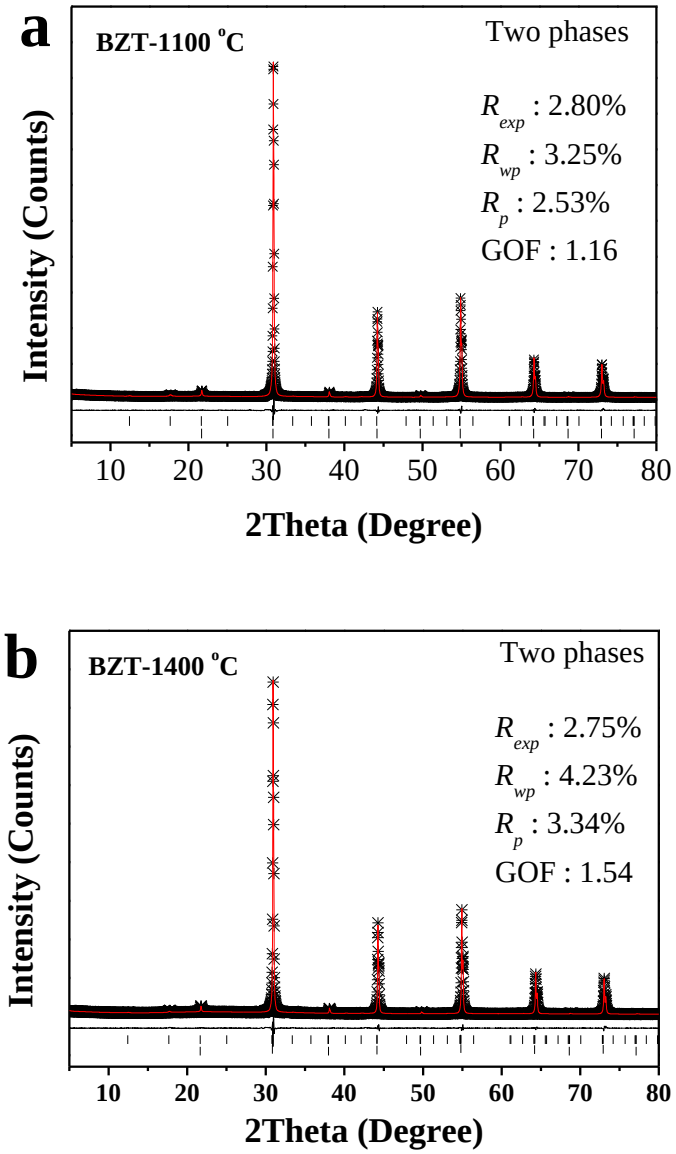


Figure S2. Rietveld refinements using two phases for XRD data of BZT obtained at 1100 ℃ (a) and 1400 ℃ (b), respectively. The refined mass percentages of the cubic phase for BZT obtained at 1100 ℃ and 1400 ℃ are 29.9% and 46.3%, respectively.

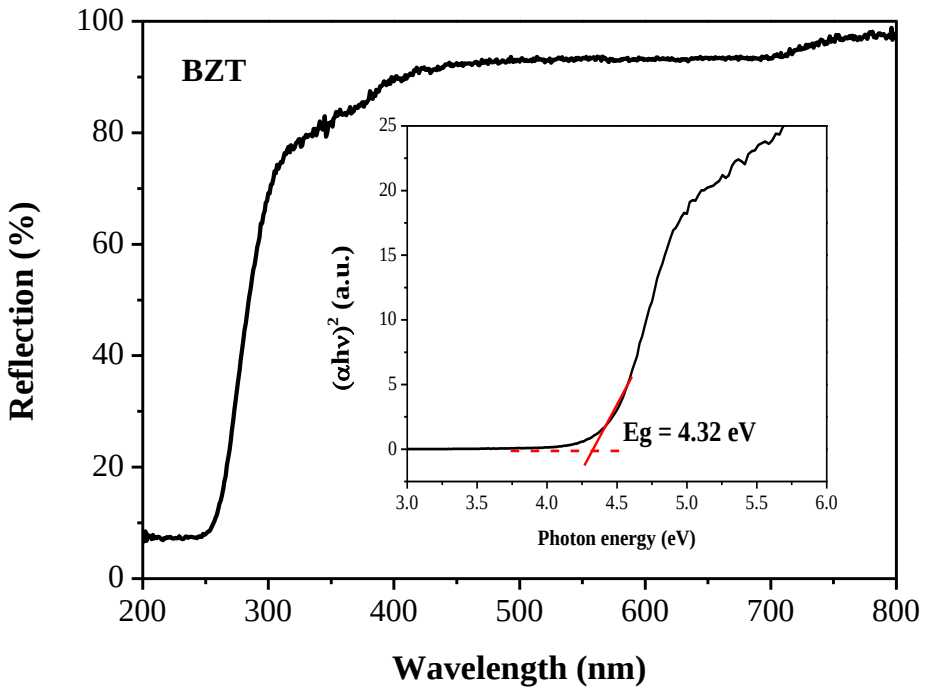
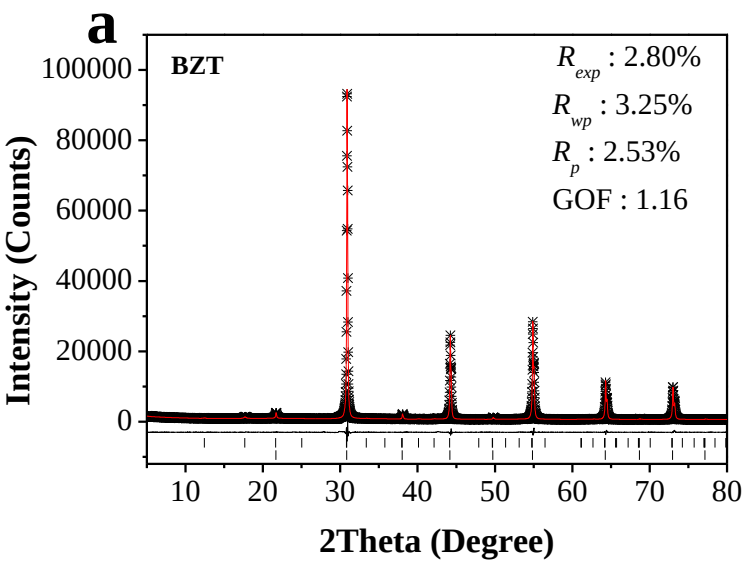
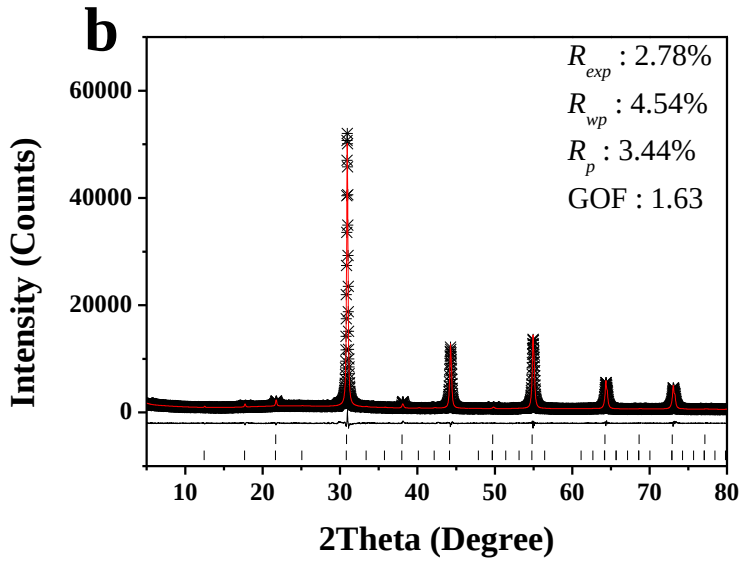
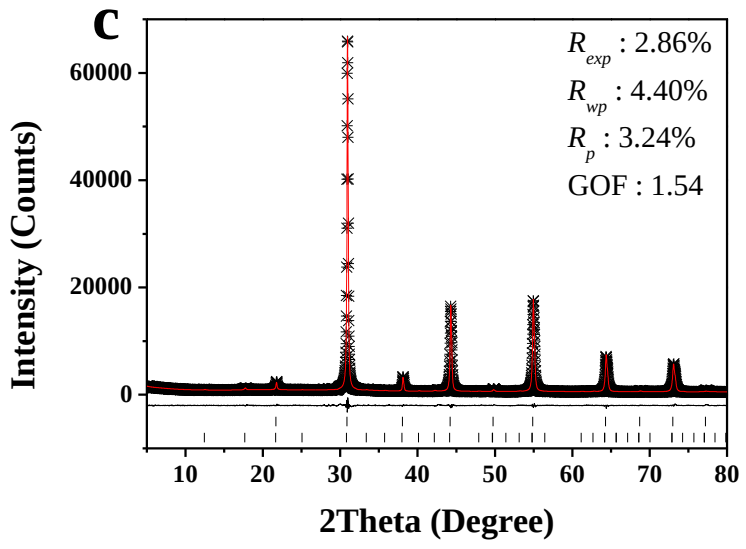


Figure S3. UV-Vis diffuse reflection spectrum of BZT sample. The inset is the $(\alpha h\nu)^2$ ~ $h\nu$ plot.


a
BZT
R_{exp} : 2.80%
R_{wp} : 3.25%
R_p : 2.53%
GOF : 1.16
Intensity (Counts)
2Theta (Degree)


b
R_{exp} : 2.78%
R_{wp} : 4.54%
R_p : 3.44%
GOF : 1.63
Intensity (Counts)
2Theta (Degree)


c
R_{exp} : 2.86%
R_{wp} : 4.40%
R_p : 3.24%
GOF : 1.54
Intensity (Counts)
2Theta (Degree)

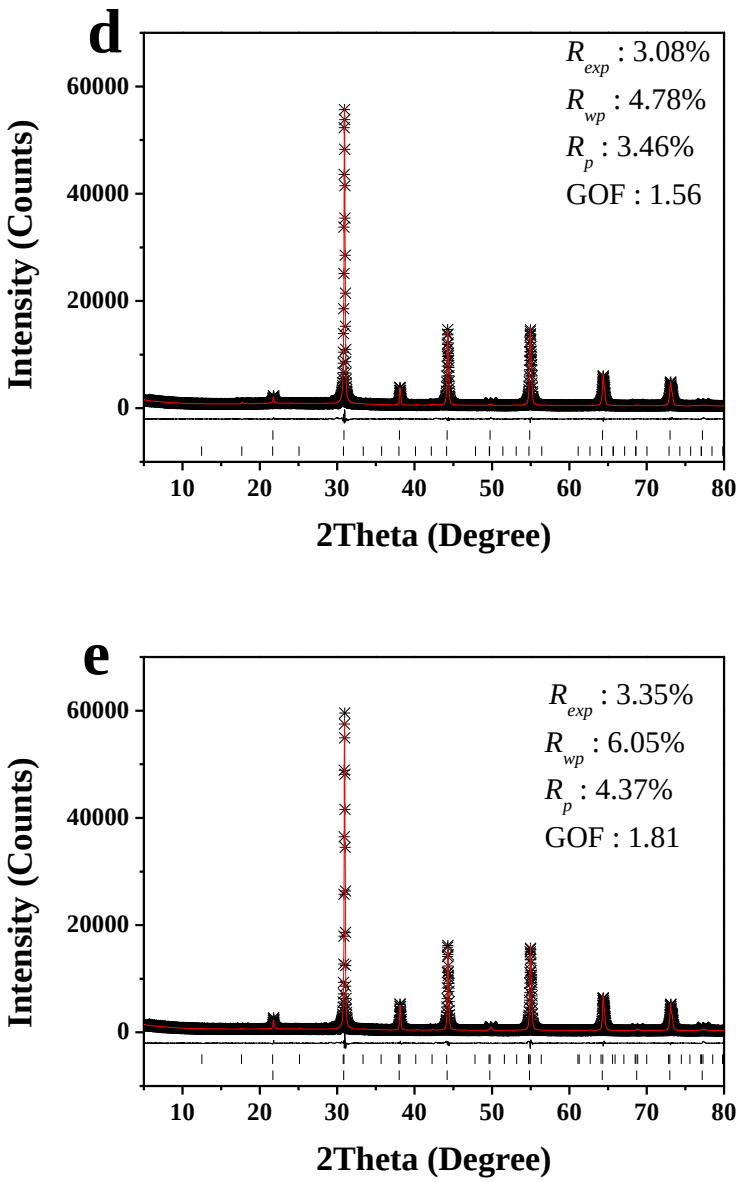


Figure S4. Quantitative Rietveld refinement plots of $Ba_3ZnTa_{2-x}Nb_xO_9$ samples. (a) x = 0.0, (b) x = 0.5, (c) x = 1.0, (d) x = 1.5 and (e) x = 2.0.

Table S1. The refined structural parameters of B-site disordered cubic BZN*.

Atom	Site	x	y	z	Occ	Biso(Å^2)	BVS
Ba	1b	0.5	0.5	0.5	1	1	2.29
Zn	1a	0	0	0	0.333	1	2.37
Nb	1a	0	0	0	0.667	1	4.14
O	3d	0.5	0	0	1	1	1.18

*Space group: *Pm*-3*m*, *a* = *b* = *c* = 4.0959(1) Å. *Z* = 1

Table S2. The refined structural parameters of B-site ordered trigonal BZN*

Atom	Site	x	y	z	Occ	Biso(Å^2)	BVS
Ba1	1a	0	0	0	1	1	2.18
Ba2	2d	1/3	2/3	0.633(3)	1	1	2.42
Zn1	1b	0	0	1/2	1	1	2.19
Nb1	2d	1/3	2/3	0.166(3)	1	1	4.20
O1	3e	0.5	0	0	1	1	1.95
O2	6i	0.169	0.169	0.334	1	1	1.96

*Space group: *P*-3*m*1, *a* = *b* = 5.815(1) Å, *c* = 7.105(3) Å, *Z* = 1.

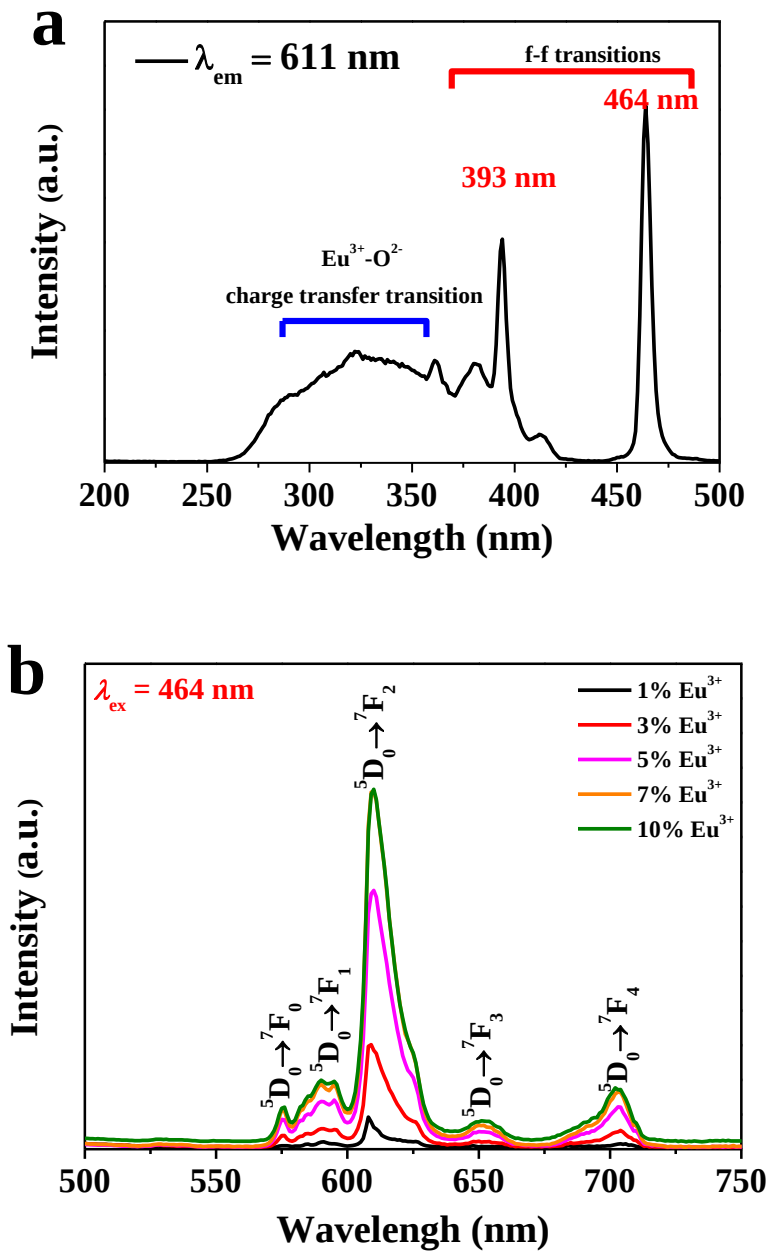


Figure S5. (a) PLE spectrum of $Ba_{3-y}Eu_yZnNb_2O_9$ (y = 10%) by monitoring 611 nm-emission**. (b)** Eu^{3+}-Emission spectra (λ_{ex} = 464 nm) of $Ba_{3-y}Eu_yZnNb_2O_9$ (y = 1.0%, 3.0%, 5.0%, 7.0%, 10%) samples.

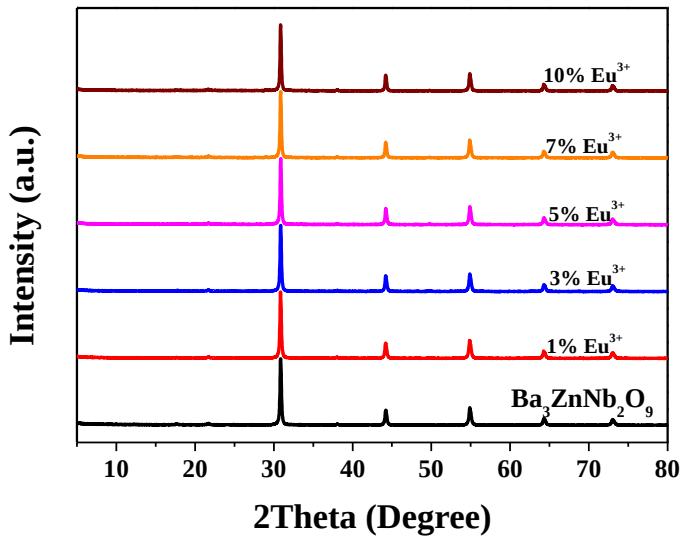


Figure S6. XRD patterns of $Ba_{3-y}Eu_yZnNb_2O_9$ (y =0, 1.0%, 3.0%, 5.0%, 7.0%, 10%) samples.

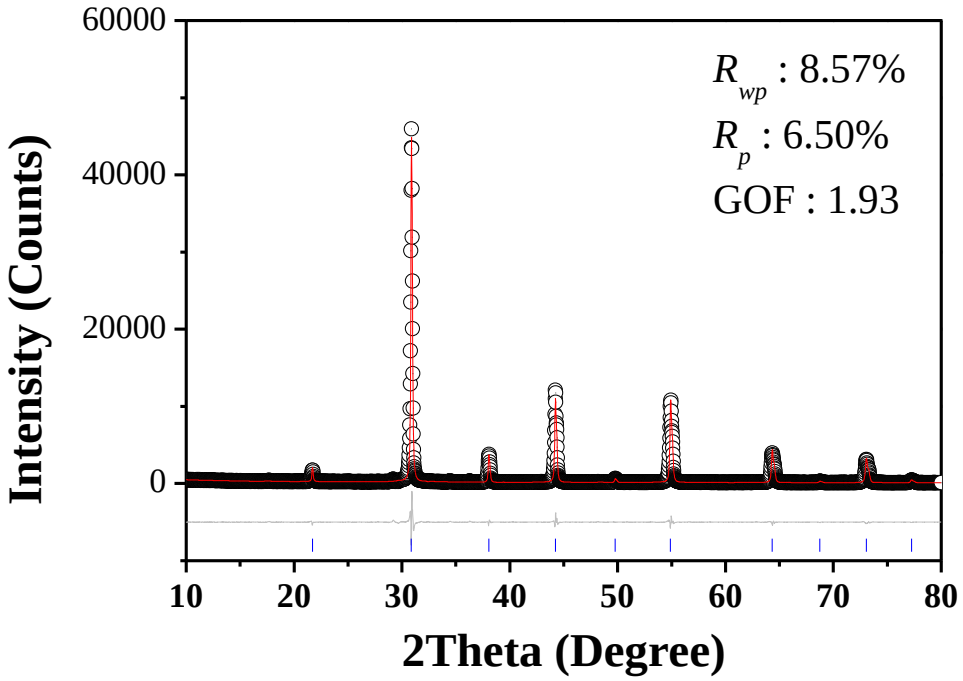


Figure S7. Rietveld refinement plots of $Ba_{3-y}Eu_yZnNb_2O_9$ (y = 0.1) samples.

Table S3. The refined structural parameters of $Ba_{3-y}Eu_yZnNb_2O_9$ (y = 0.1)

Atom	Site	x	y	z	Occ	Biso(Å^2)
Eu	1b	0.5	0.5	0.5	0.03 (1)	1
Ba	1b	0.5	0.5	0.5	0.97 (1)	1
Zn	1a	0	0	0	0.333	1
Nb	1a	0	0	0	0.667	1
O	3d	0.5	0	0	1	1

*Space group: *Pm*-3*m*, *a* = *b* = *c* = 4.093 (1) Å. *Z* = 1

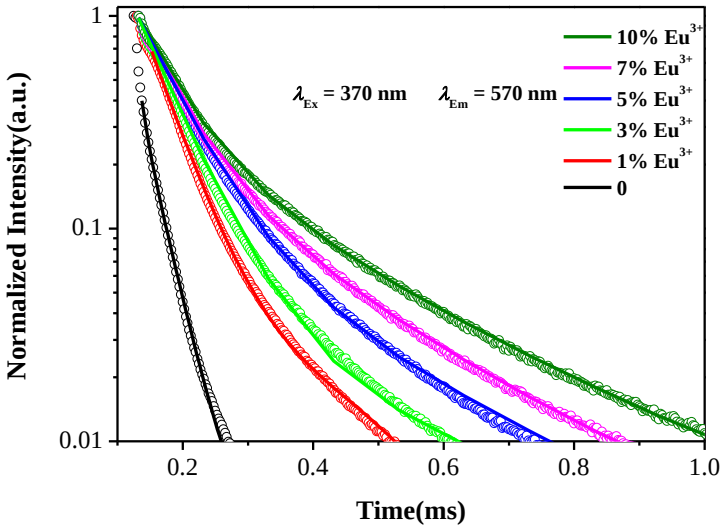


Figure S8. Luminescence decay curves (λ_{ex} = 370 nm, λ_{em} =570 nm) of $Ba_{3-y}Eu_yZnNb_2O_9$ (y =0.0, 1.0%, 3.0%, 5.0%, 7.0%, 10%) samples at room temperature.

The decay curves of $Ba_{3-y}Eu_yZnNb_2O_9$ (y =0.0, 1.0%, 3.0%, 5.0%, 7.0%, 10%) samples can be well fitted with the second-order exponential decay mode by using equation:

$$I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) \quad (1)$$

Where I is the luminescence intensity, A_1 and A_2 are constants, t is the time, τ_1 and τ_2 are rapid and slow lifetime values of exponential components, respectively. According to the equation (1), the A_1, A_2, τ_1 and τ_2 values can be obtained by fitting the decay curves. Thus, the average lifetimes (τ^*) of $Ba_{3-y}Eu_yZnNb_2O_9$ can be calculated as equation (2):

$$\tau^* = (A_1\tau_1^2 + A_2\,\tau_2^2) / (A_1\tau_1 + A_2\,\tau_2) \quad (2)$$

The average lifetimes of $Ba_{3-y}Eu_yZnNb_2O_9$ (y =0.0, 1.0%, 3.0%, 5.0%, 7.0%, 10%) samples were calculated as 8.2 μs, 57.5 μs, 71.3 μs, 85.8 μs, 91.2 μs and 115.3 μs, respectively.

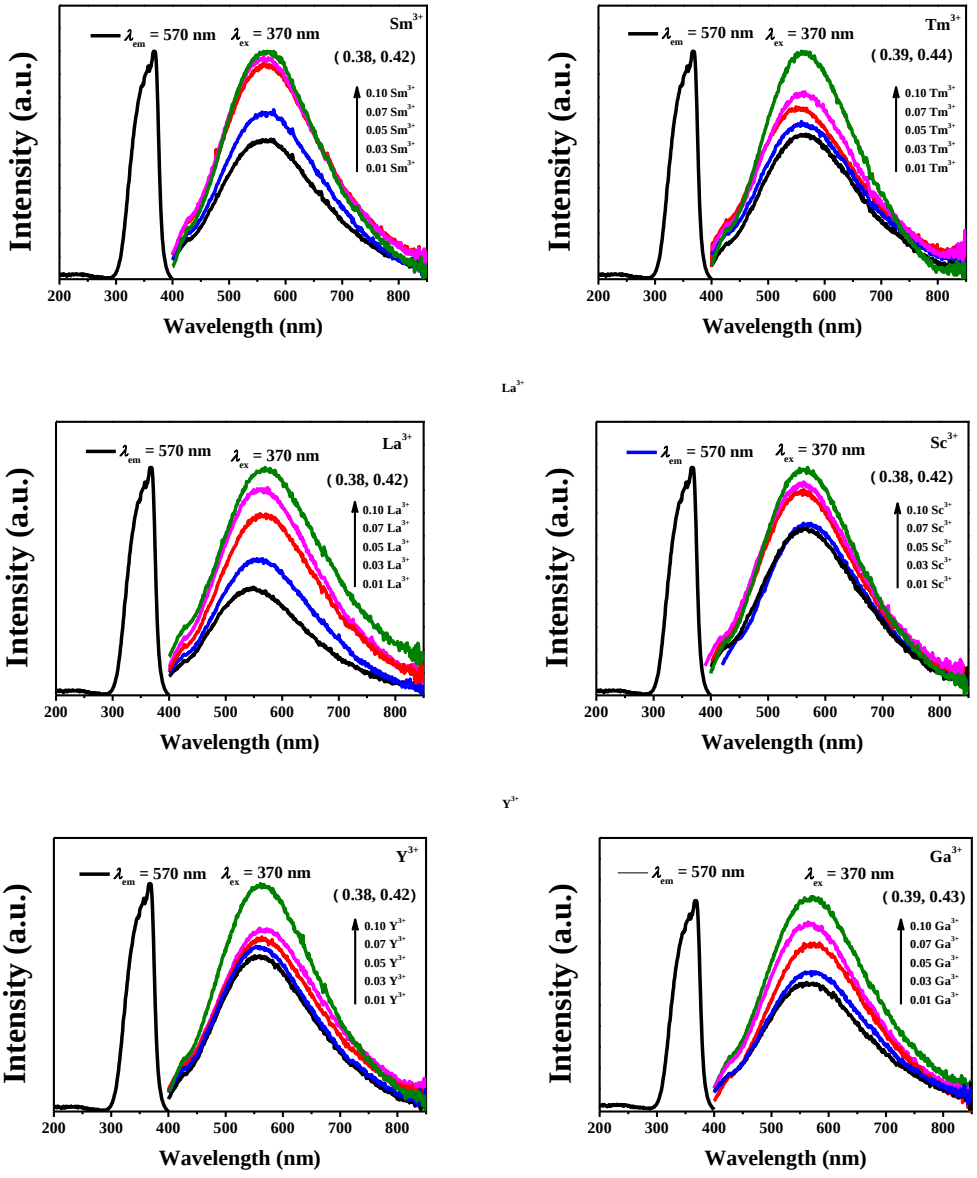


Figure S9. PL and PLE spectra of $Ba_{3-y}Ln_yZnNb_2O_9$ (Ln = Sm^{3+}, Tm^{3+}, La^{3+}, Sc^{3+}, Y^{3+}, Ga^{3+}, y = 0.01-0.10) samples.

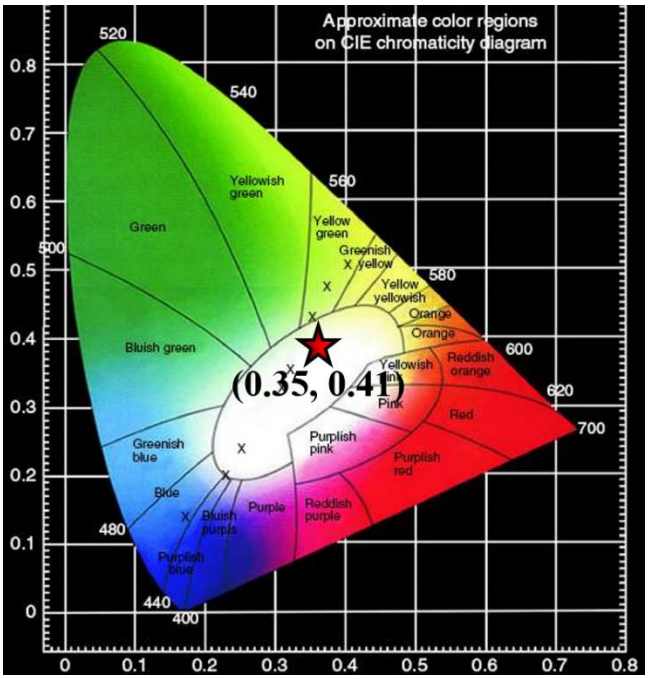


Figure S10. CIE chromaticity diagram of $Ba_{3-y}Eu_yZnNb_2O_9$ (y = 0.1) phosphors.

For the fabrication of LED device, the single $Ba_{3-x}Eu_xZnNbO_9$ (x = 0.1) phosphor sufficiently blended with silicone gel, and then a layer of this mixture was coated on a 370 n-UV LED chip to prepare WLED devices. The electroluminescence (EL) spectra of WLED device were measured on a PMS-80 fluorescence spectrometer.