Tailoring multi-dimensional traps towards rewritable multi-level optical data storage

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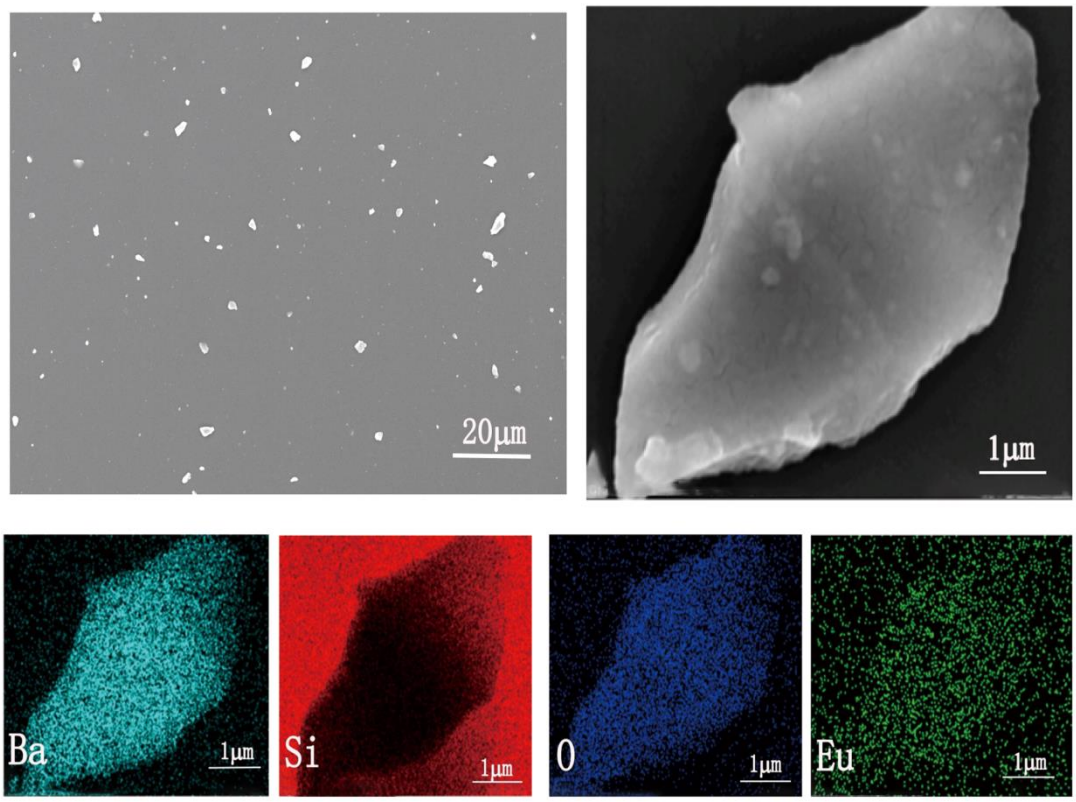

Figure S1. The SEM and element mapping images of BS:0.005Eu^{2+}.

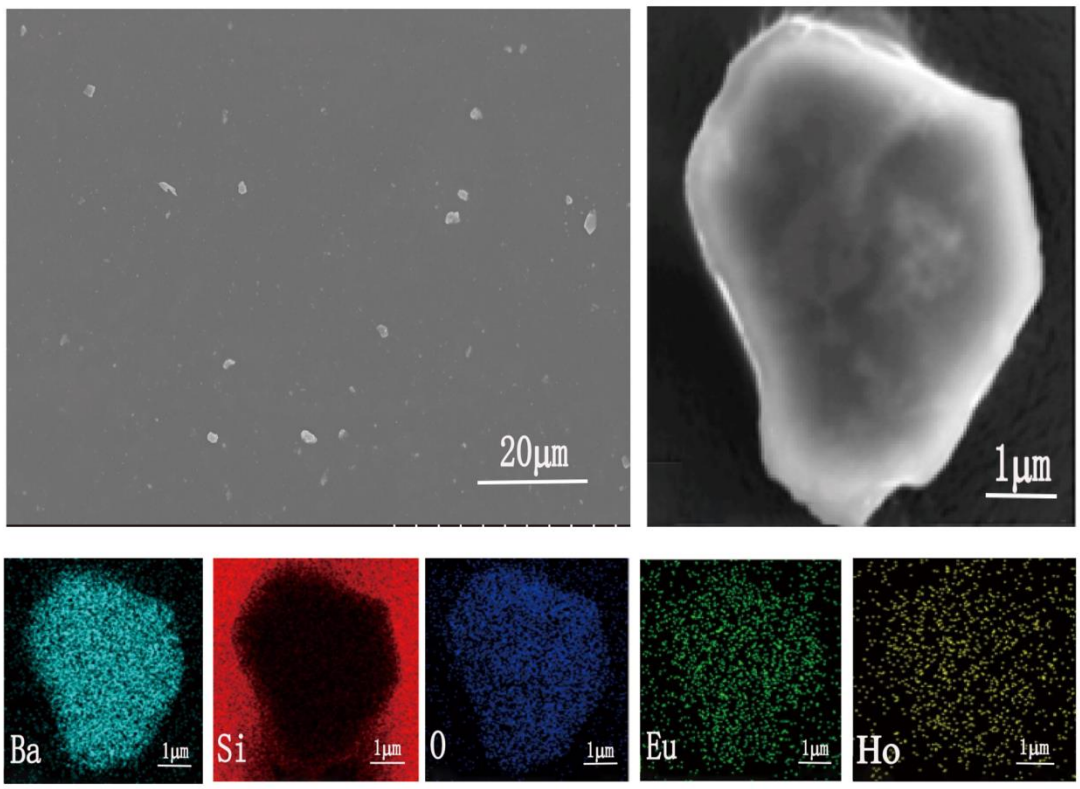

Figure S2. The SEM and element mapping images of BS:0.005Eu^{2+}, 0.01Ho^{3+}.

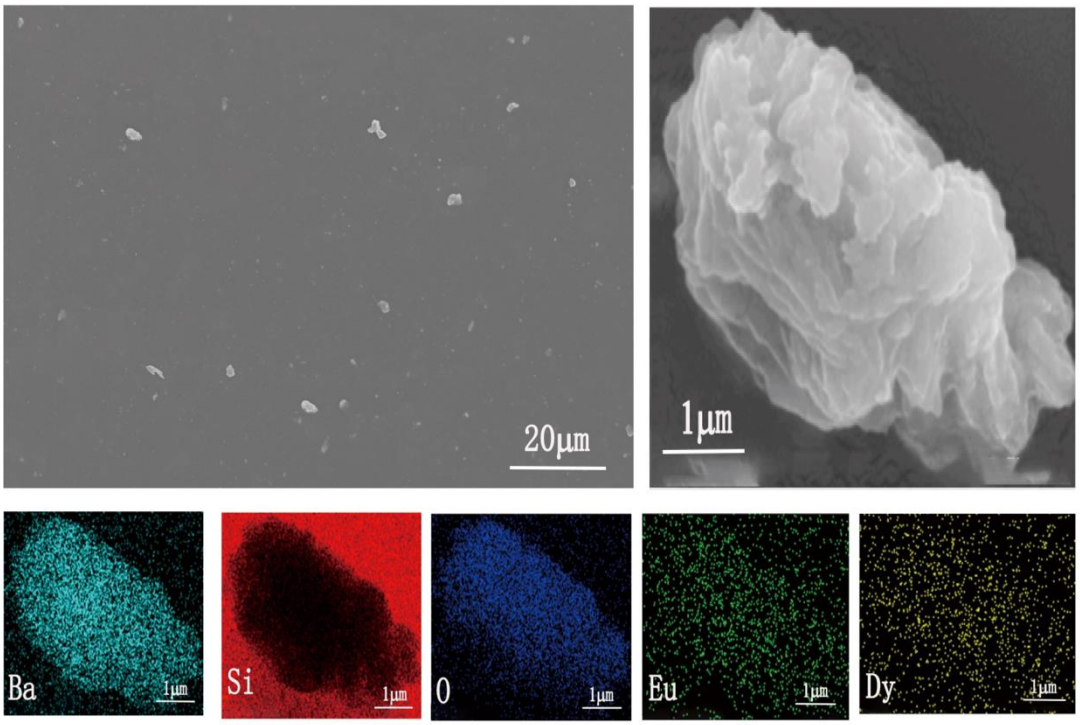

Figure S3. The SEM and element mapping images of BS:0.005Eu^{2+}, 0.02Dy^{3+}

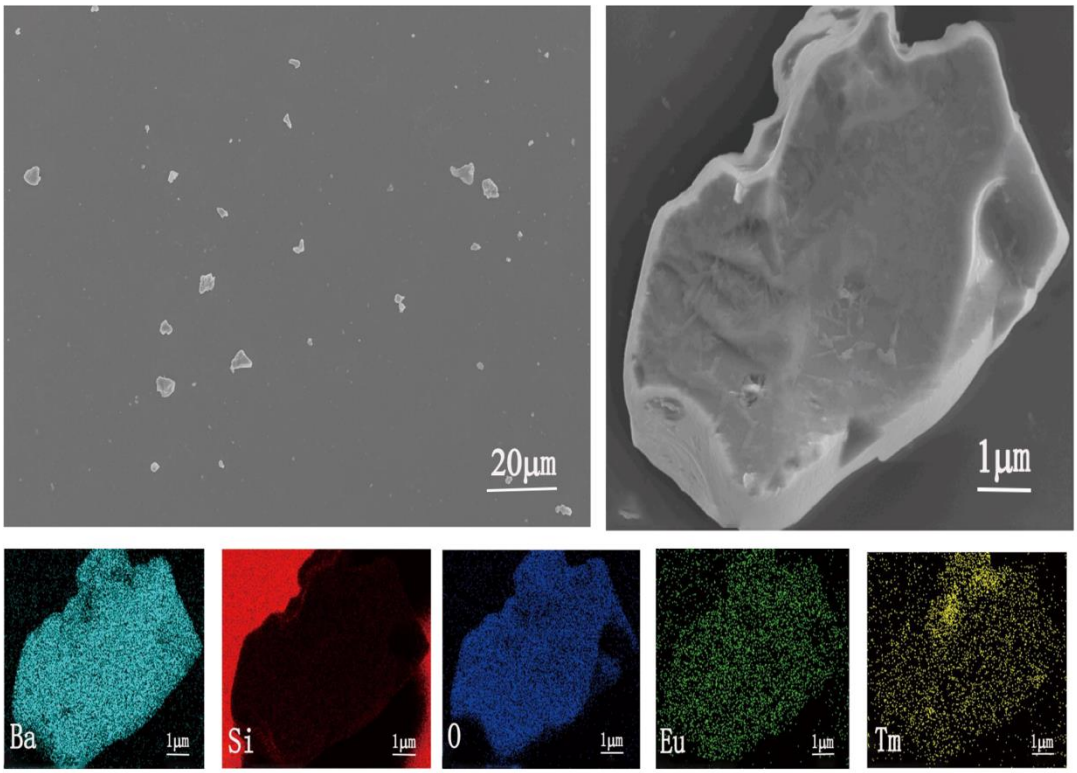

Figure S4. The SEM and element mapping images of BS:0.005Eu^{2+}, 0.01Tm^{3+}

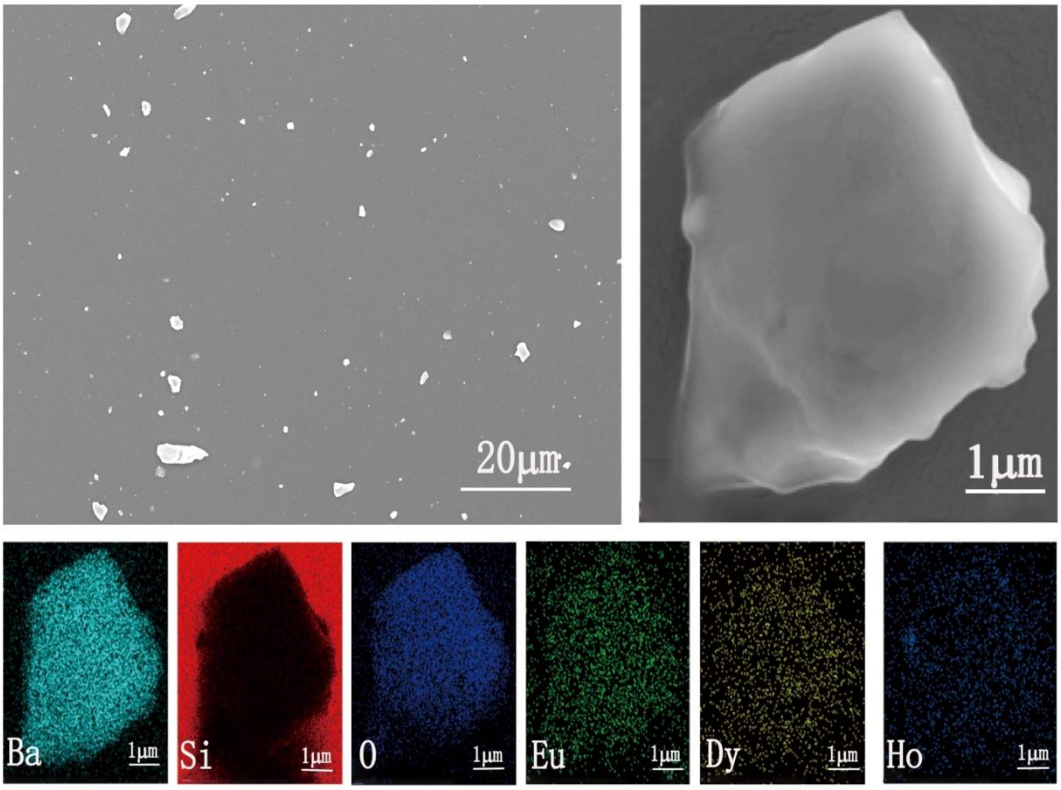

Figure S5. The SEM and element mapping images of BS:0.005Eu^{2+}, 0.002Dy^{3+}, 0.01Ho^{3+}

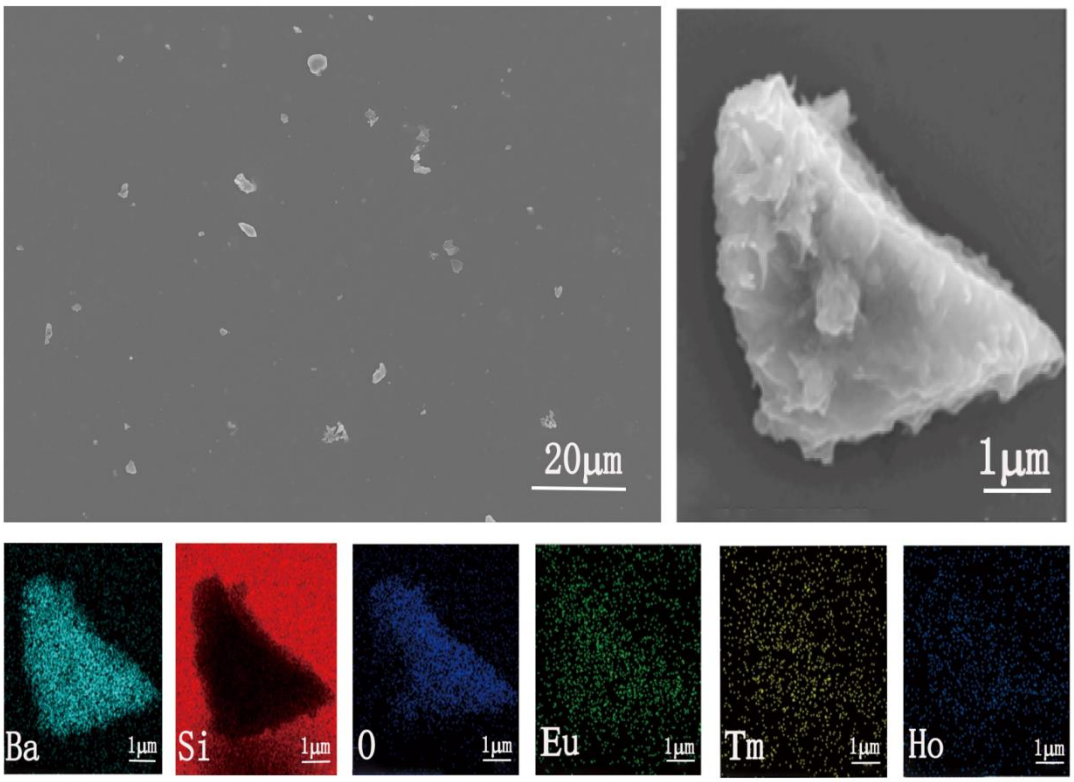

Figure S6. The SEM and element mapping images of BS:0.005Eu^{2+}, 0.001Tm^{3+}, 0.001Ho^{3+}

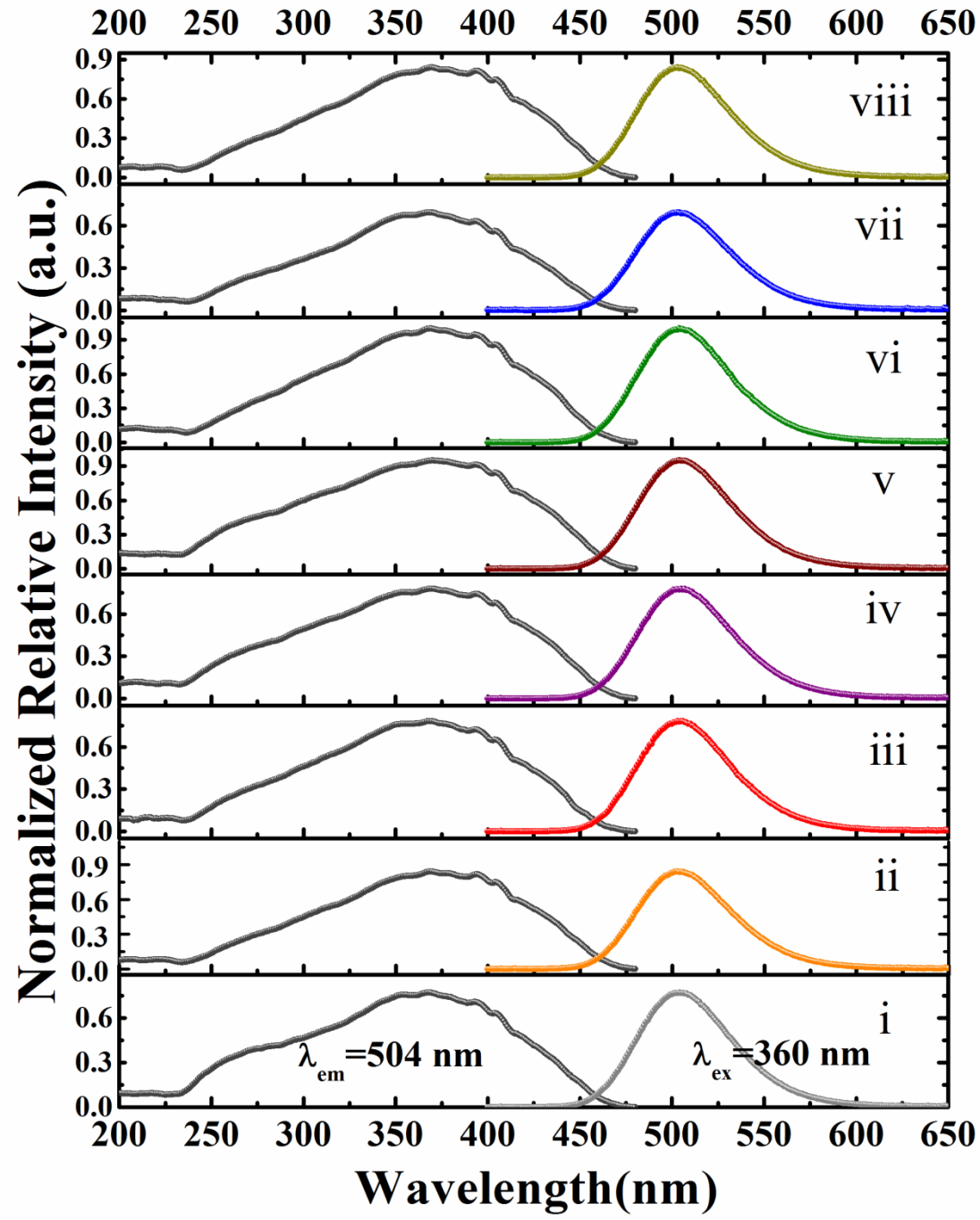

Figure S7. Excitation (λ_{em}=504 nm) and emission (λ_{ex}=360 nm) spectra of BS:0.005Eu^{2+} (i) and BS:0.005Eu^{2+}, Ln^{3+}: Ln=0.01Er (ii), 0.01Ho (iii), 0.002Dy (iv), 0.001Tm (v), 0.002Dy+0.01Ho (vi), 0.001Tm+0.001Ho (vii), and 0.001Tm+0.001Ho+0.0005Dy (viii).

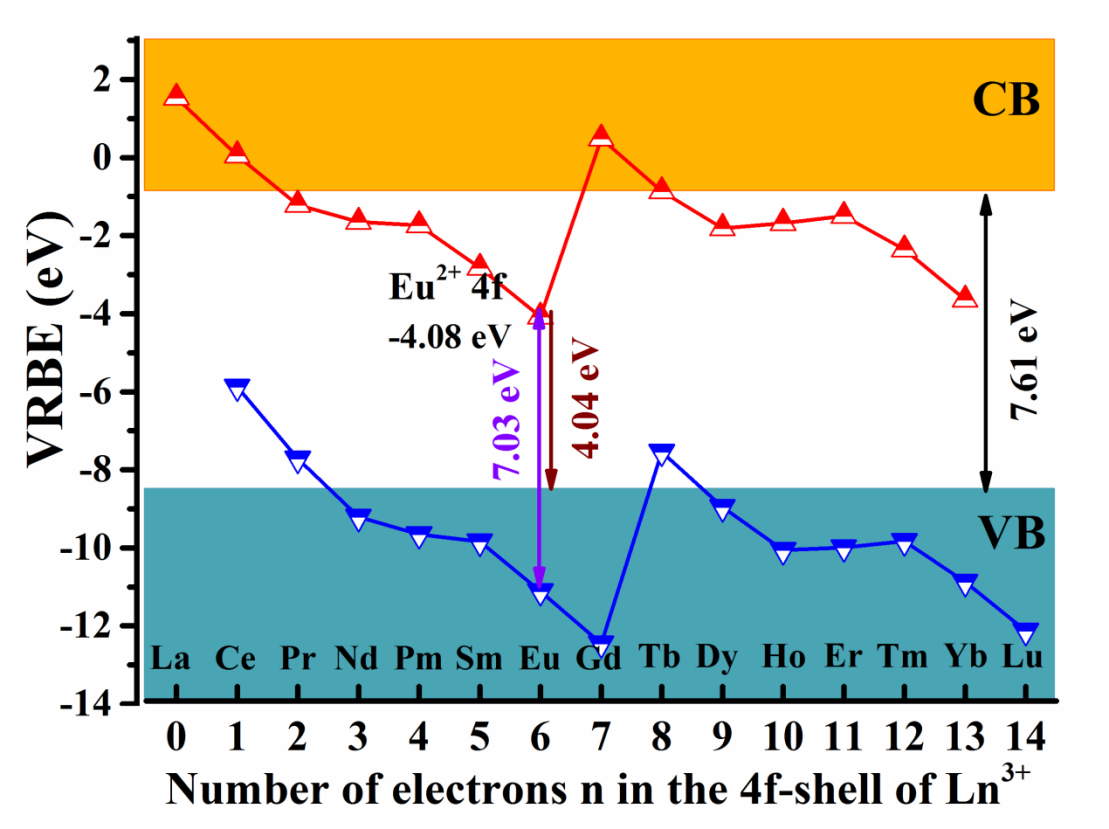

Figure S8. The VRBE schemes of Ln^{2+}/Ln^{3+}-doped Ba_2SiO_4. The 4f ground states of Ln^{2+} and Ln^{3+} are labeled by red regular triangle and blue irregular triangle.

In order to construct a 4f-VRBE scheme, Eqs. (1), (2), and (3) and the required parameters *U(6,A)*, $E^{CT}(6, 3+, A)$ and $E^{ex}(A)$ are extracted from ref. [15].

$$E_{4f}(7,2+,A) = -24.92 + \frac{18.05 - U(6,A)}{0.777 - 0.0353U(6,A)} \quad (1)$$

Where $E_{4f}(7, 2+, A)$ is the 4f-shell electron binding energies of Eu^{2+}. −24.92 eV is the experimentally known 4f-shell electron binding energy for gaseous Eu^{2+} (A=vacuum), and the second term on the right-hand side is the chemical shift.

$$E_V(A) = E_{4f}(7,2+,A) - E^{CT}(6,3+,A) = -\Phi(A) \quad (2)$$

where $E_V(A)$ is equivalent to the work function $-\Phi(A)$, $E^{CT}(6, 3+, A)$ is the energy needed to pump an electron from the top of the valence band into the 4f shell of Eu^{3+} thus generating Eu^{2+}.

$$E_C(A) = E_V(A) + 1.08 \times E^{ex}(A) = -\chi(A) \quad (3)$$

where $E_C(A)$ is the energy at the bottom of the conduction band. $E^{ex}(A)$ represents the host compound excitonic absorption energy.

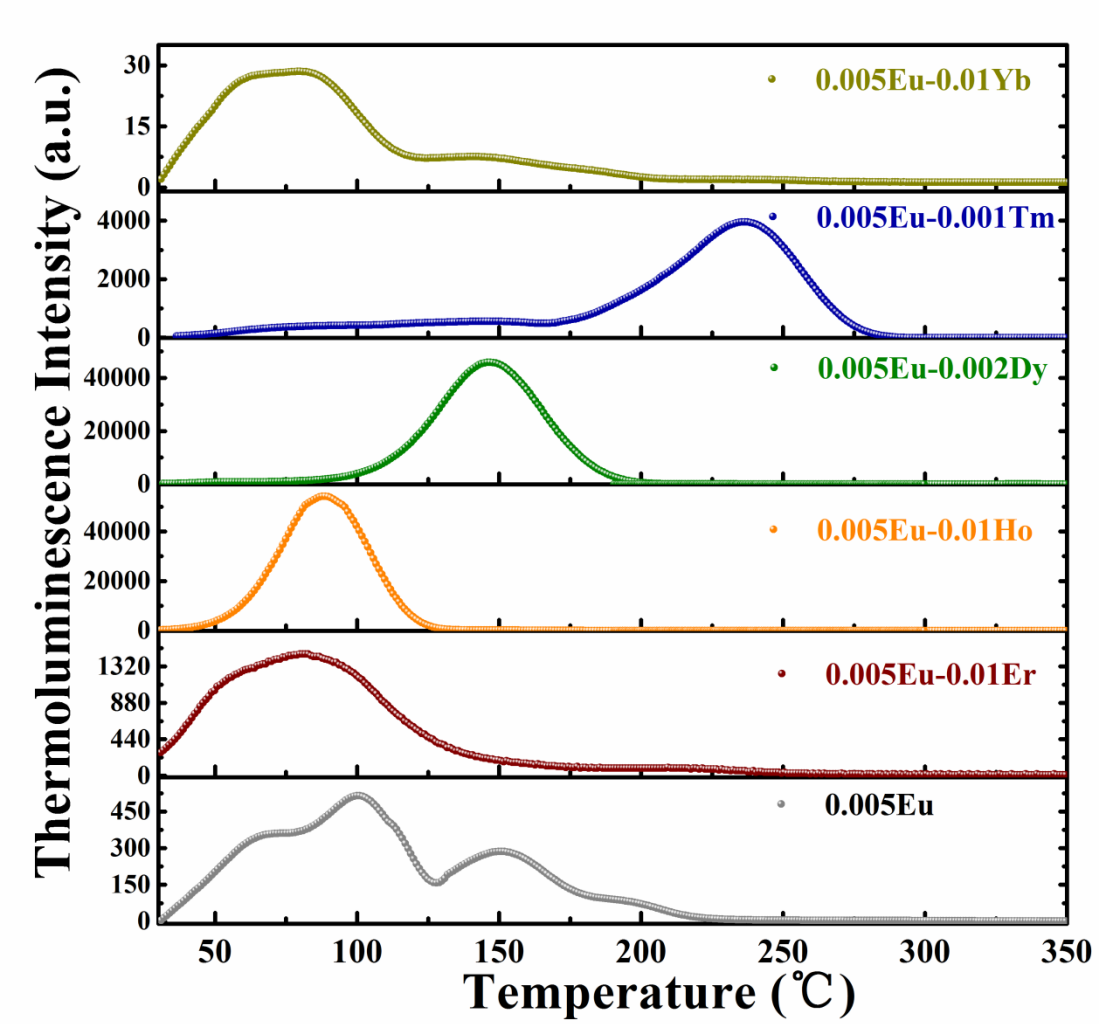

Figure S9. The TL curves of samples BS:0.005Eu^{2+} and BS:0.005Eu^{2+}, Ln^{3+} after excitation by 254 nm light for 5 min. Ln is referred to single element of Er, Ho, Dy, Tm and Yb. The heating rate is set as 2 K/s for all of the measurements.

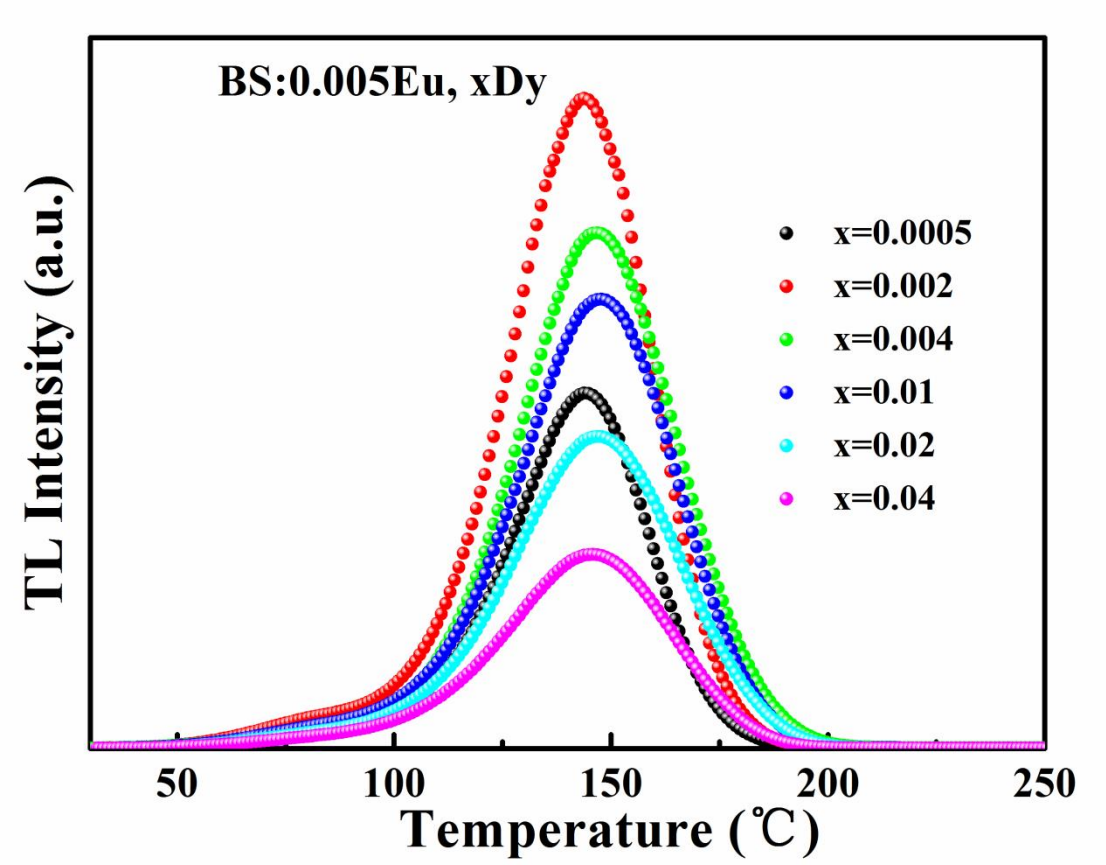

Figure S10. The TL glow curves of BS:0.005Eu^{2+}, xDy^{3+} ($0.0005 \leq x \leq 0.04$) phosphors.

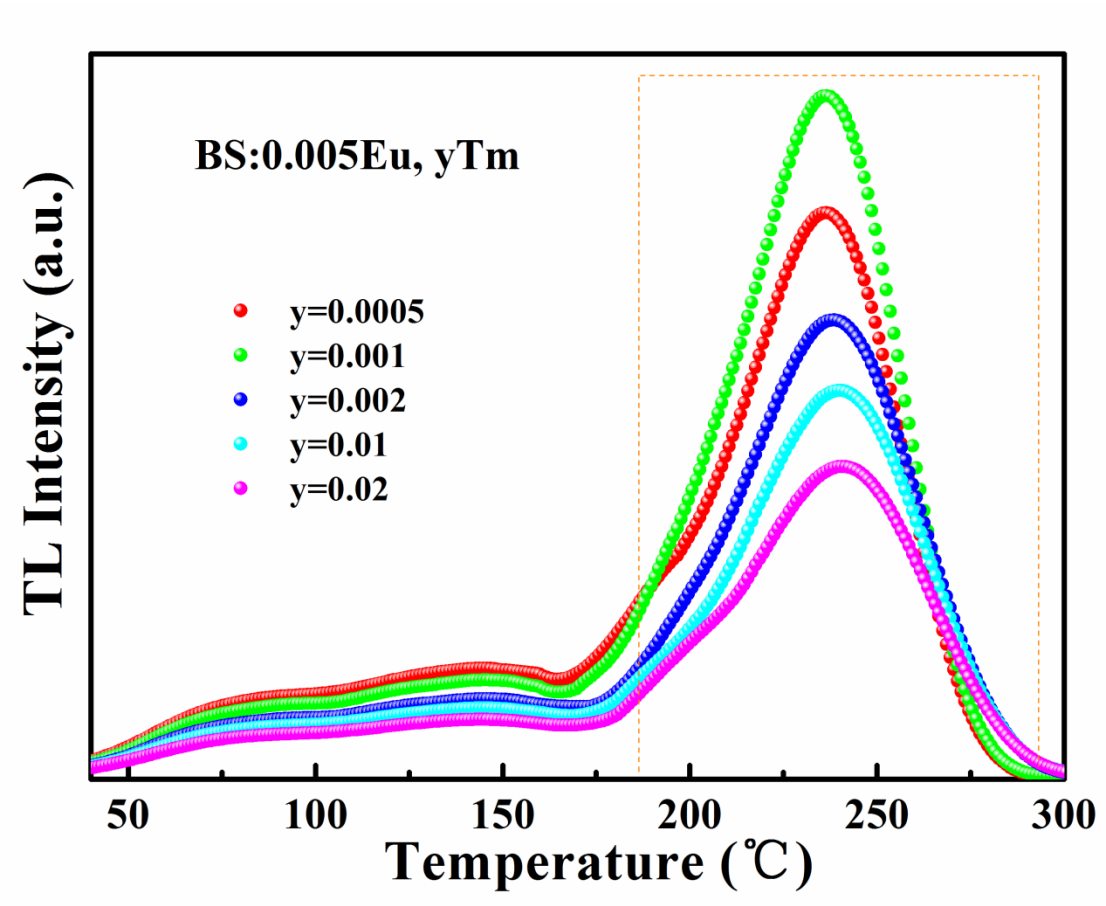

Figure S11. The TL glow curves of BS:0.005Eu^{2+}, yTm^{3+} ($0.0005 \leq y \leq 0.04$) phosphors.

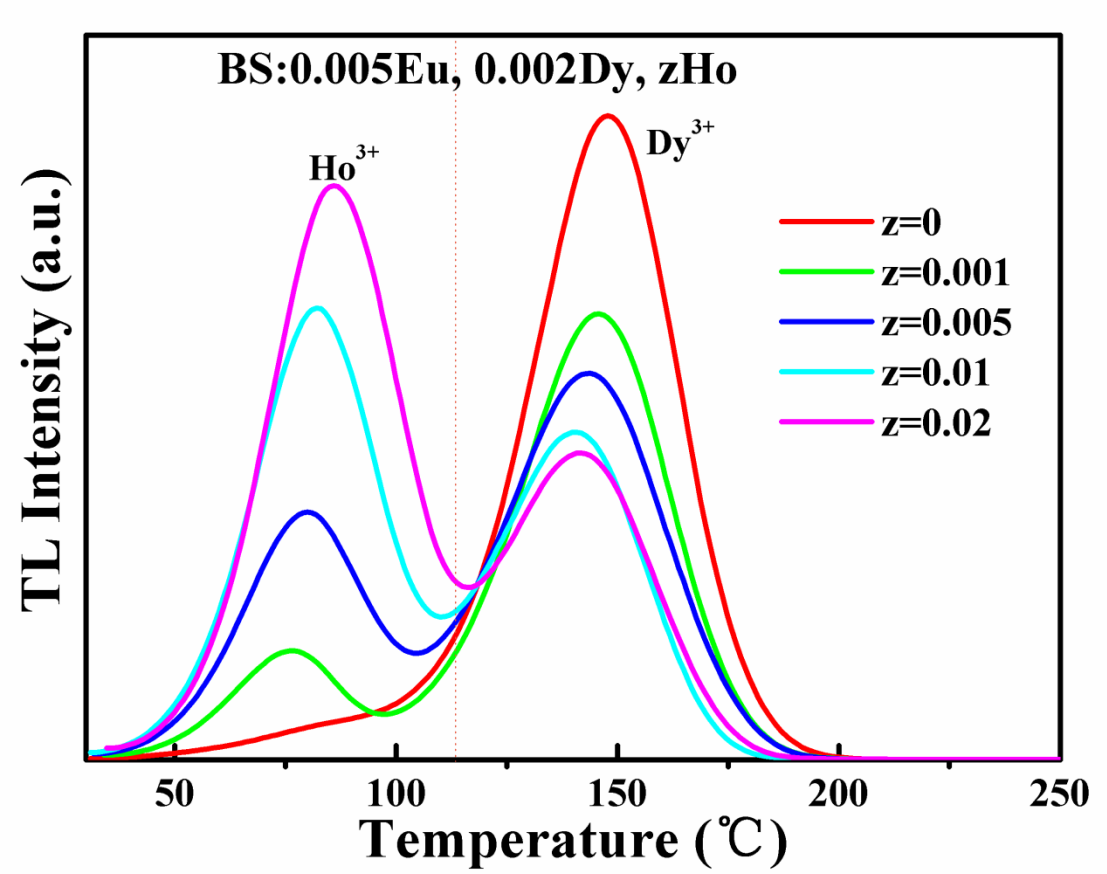

Figure S12. The TL glow curves of BS:0.005Eu^{2+}, 0.002Dy^{3+}, zHo^{3+} ($0 \leq z \leq 0.02$) phosphors.

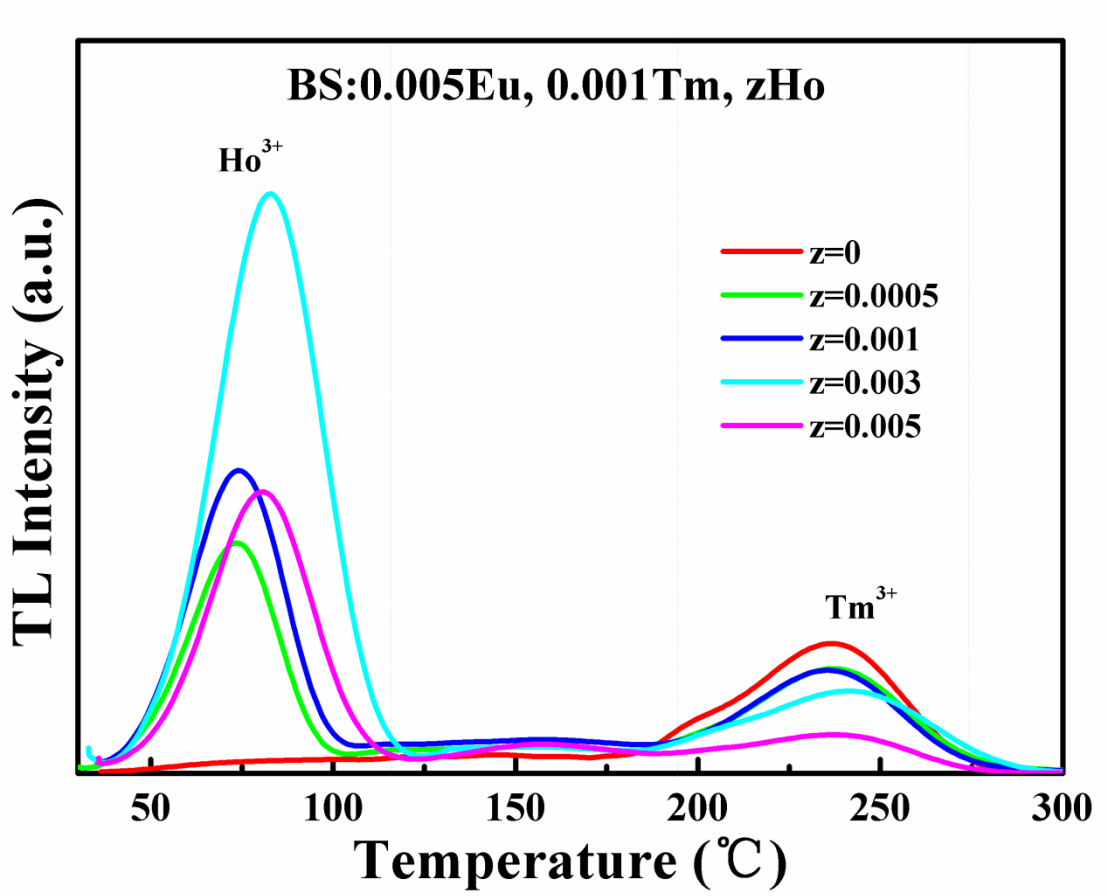

Figure S13. The TL glow curves of BS:0.005Eu^{2+}, 0.001Tm^{3+}, zHo^{3+} ($0 \leq z \leq 0.005$) phosphors.

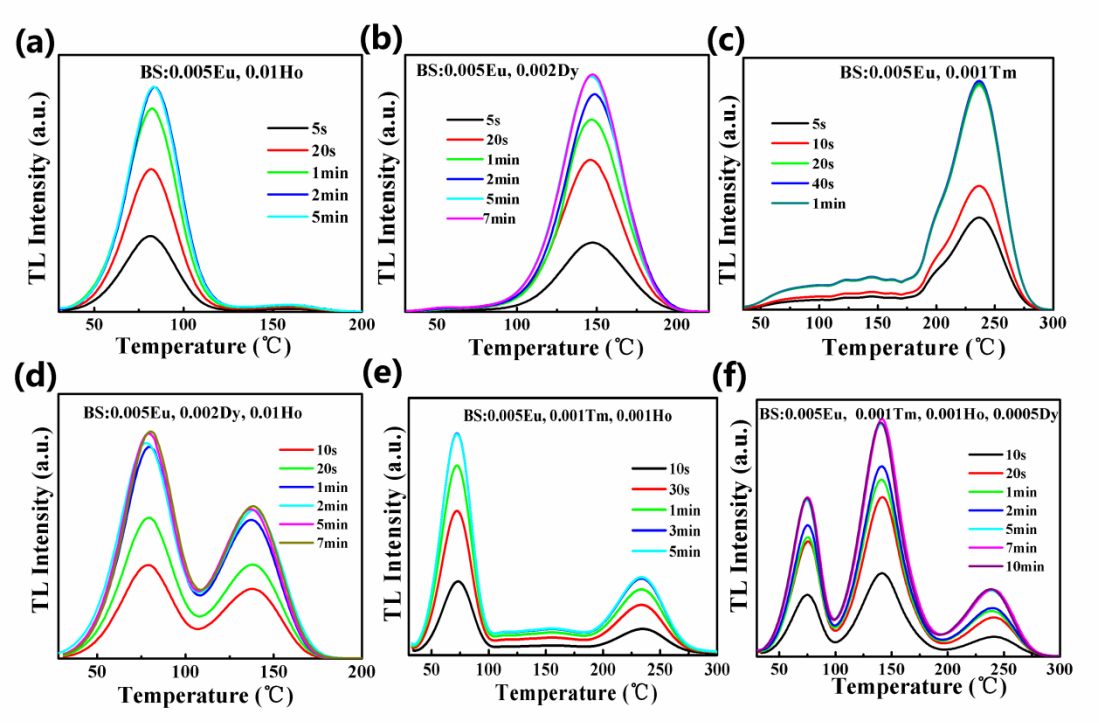

Figure S14. The TL glow curves of BS:0.005Eu^{2+}, Ln^{3+} samples measured after excitation by 254 nm light for various duration: Ln=0.01Ho (a), 0.002Dy (b), 0.001Tm (c), 0.002Dy+0.01Ho (d), 0.001Tm+0.001Ho (e), and 0.001Tm+0.001Ho+0.0005Dy (f).

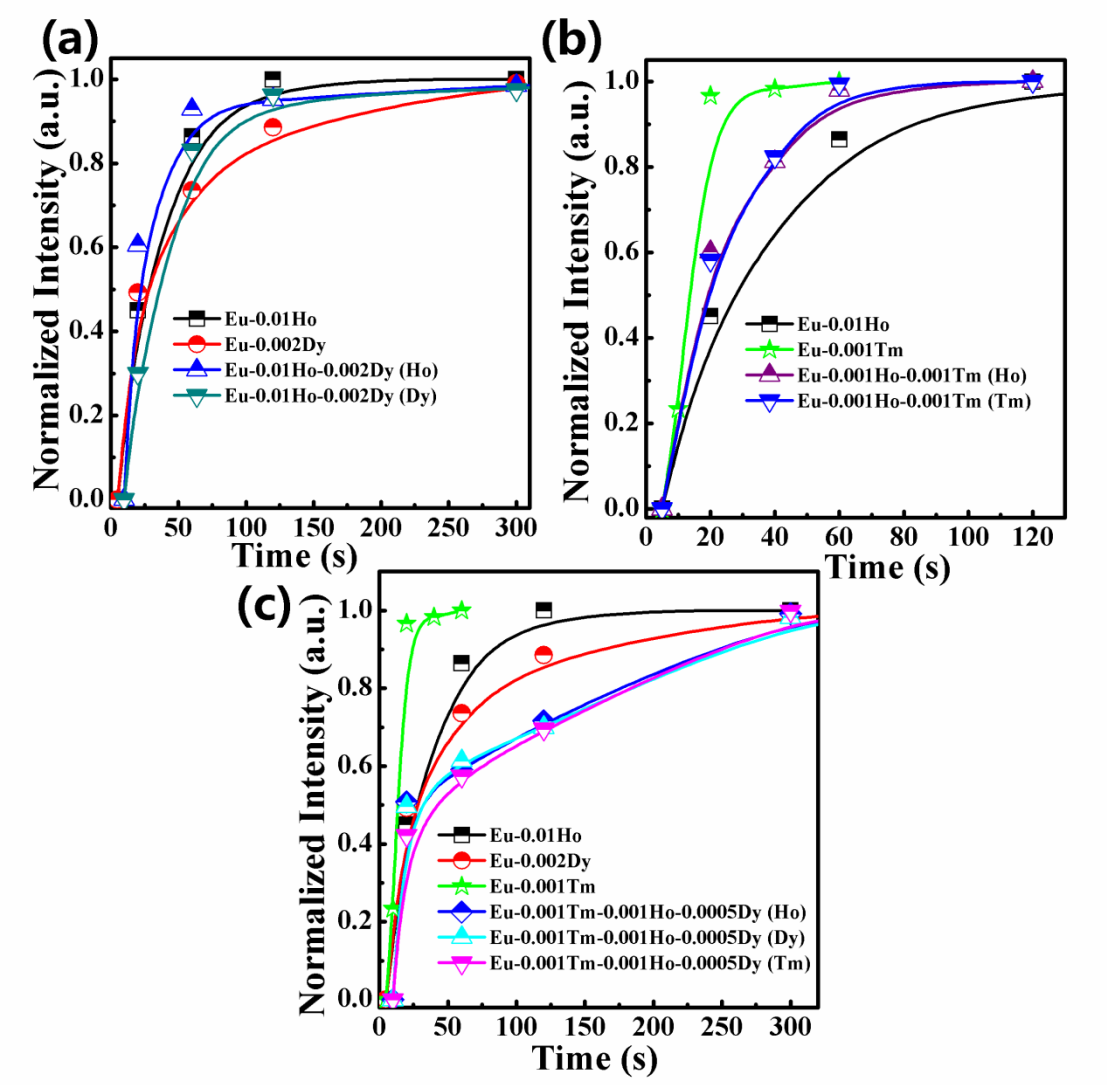

Figure S15. The dependence of trap filling on 254 nm light irradiation duration of samples BS:0.005Eu^{2+}, Ln^{3+}, which is derived from Figure S14. (a): The sample of BS:0.005Eu^{2+}, 0.01Ho^{3+}, BS:0.005Eu^{2+}, 0.002Dy^{3+}, and BS:0.005Eu^{2+}, 0.01Ho^{3+}, 0.002Dy^{3+}. (b) The sample of BS:0.005Eu^{2+}, 0.01Ho^{3+}, BS:0.005Eu^{2+}, 0.001Tm^{3+} and BS:0.005Eu^{2+}, 0.01Ho^{3+}, 0.001Tm^{3+}. (c) The sample of BS:0.005Eu^{2+}, 0.01Ho^{3+}, BS:0.005Eu^{2+}, 0.002Dy^{3+}, BS:0.005Eu^{2+}, 0.001Tm^{3+} and BS:0.005Eu^{2+}, 0.001Tm^{3+}, 0.01Ho^{3+}, 0.002Dy^{3+}

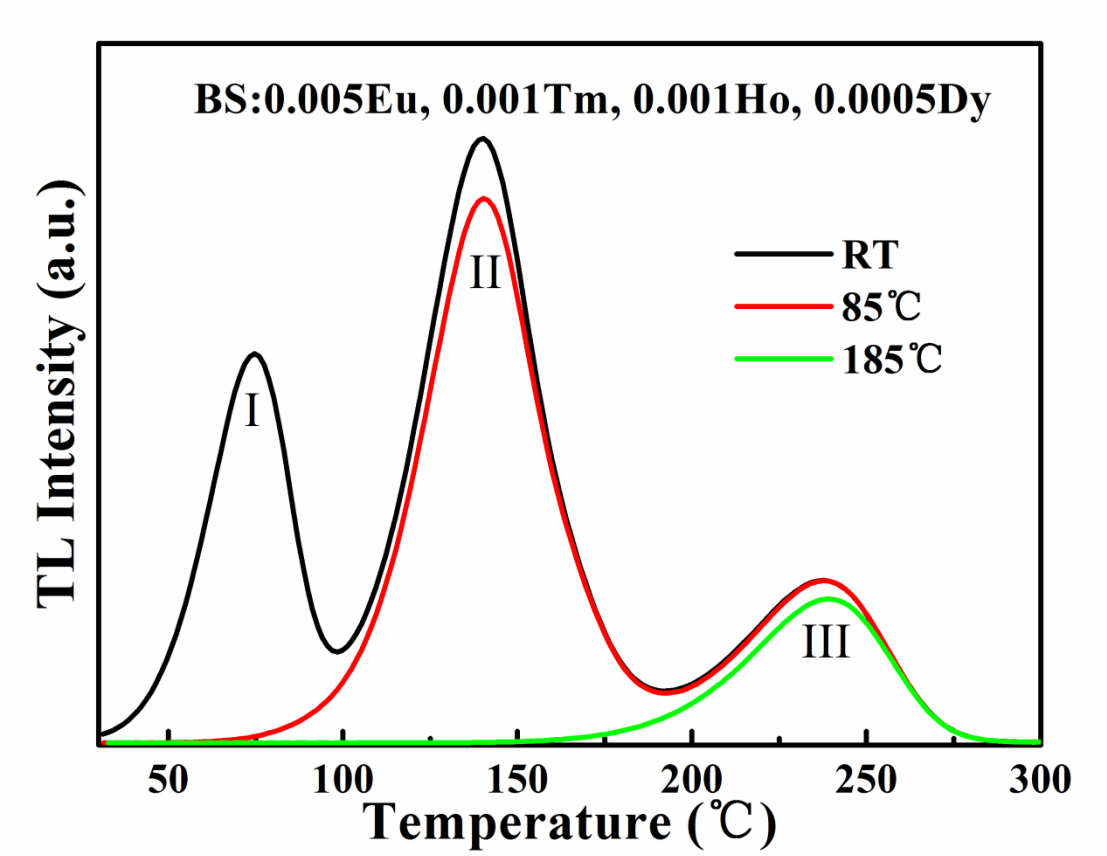

Figure S16. The TL glow curves of sample BS: 0.005Eu, 0.001Tm, 0.001Ho, 0.0005Dy (BS:Eu^{2+}, Tm^{3+}, Ho^{3+}, Dy^{3+}): black, red and green are measured after 254 nm light irradiation for 5 min without further treatment, heated up to 85 ℃ for 20 s, and 185 ℃ for 20 s, respectively.

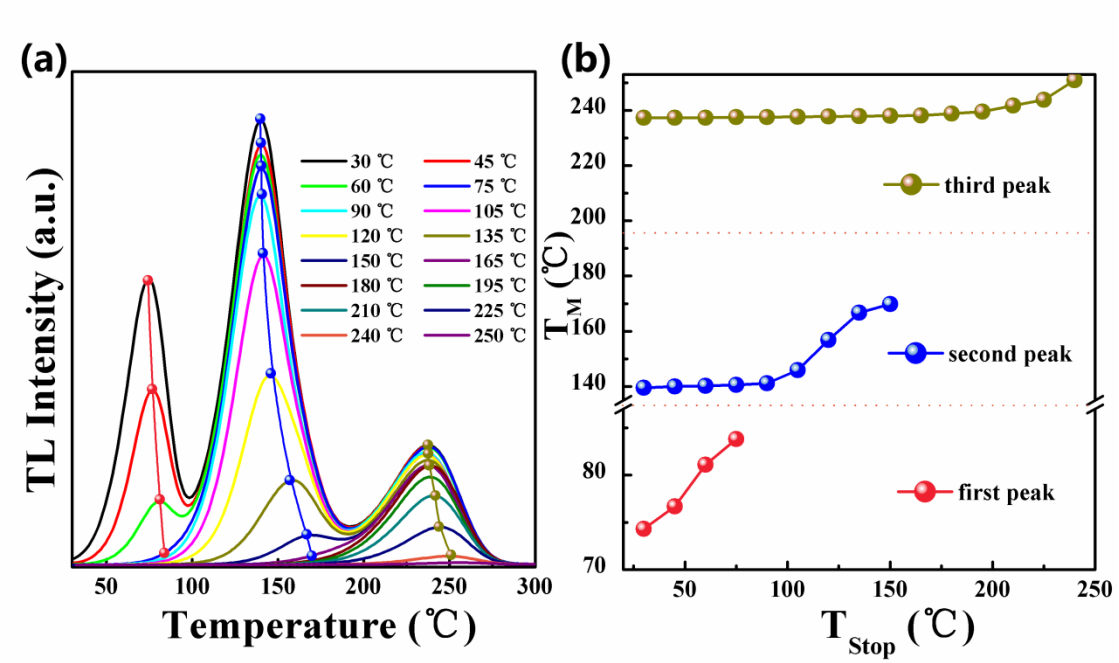

Figure S17 (a) The TL glow curves of the pre-irradiated sample BS:Eu^{2+}, Tm^{3+}, Ho^{3+}, Dy^{3+} using T_M-T_{stop} method. Before each measurement, the sample was irradiated by 254 nm for 5 min with subsequent heating to different temperatures (T_{stop}) for 20 s, and then the TL curves were recorded after cooling down to room temperature. (b) The plots of T_M-T_{stop}. T_M is the temperature at maximum of each individual TL peak.

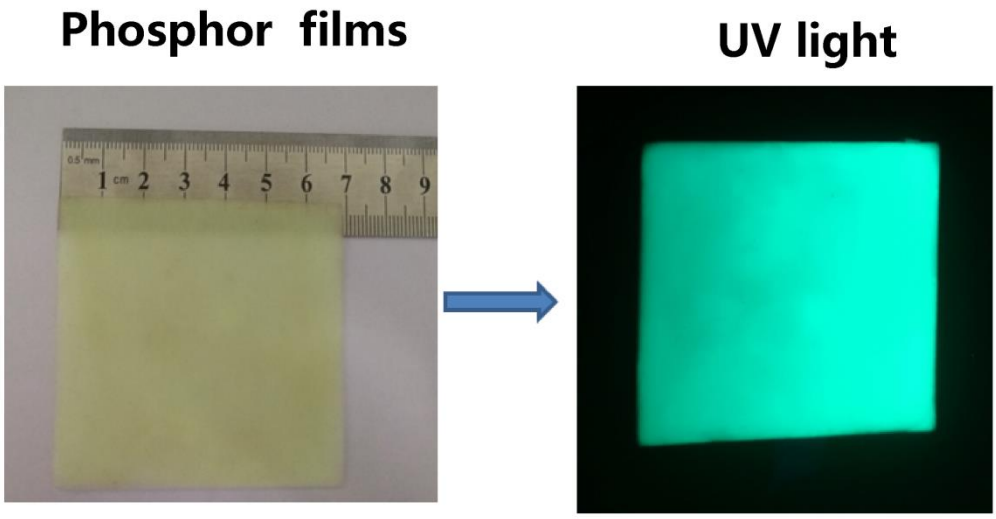

Figure S18. The flexible phosphor films under sunlight and ultraviolent light.

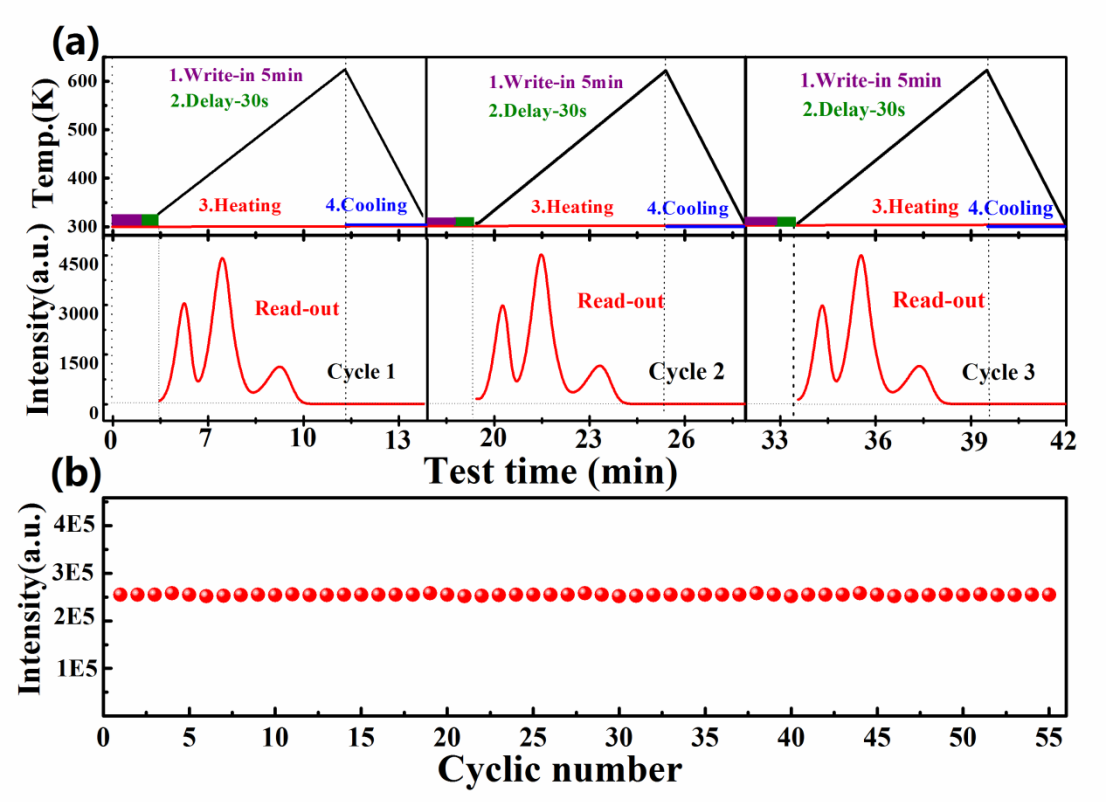

Figure S19. (a) The top panel is the schematic diagram of the whole domain information readout process characterized by TL method. The red line in the bottom panel represents the actual TL curves. (b) The TL signal intensity of the whole traps as a function of cyclic number of 55 cycles.

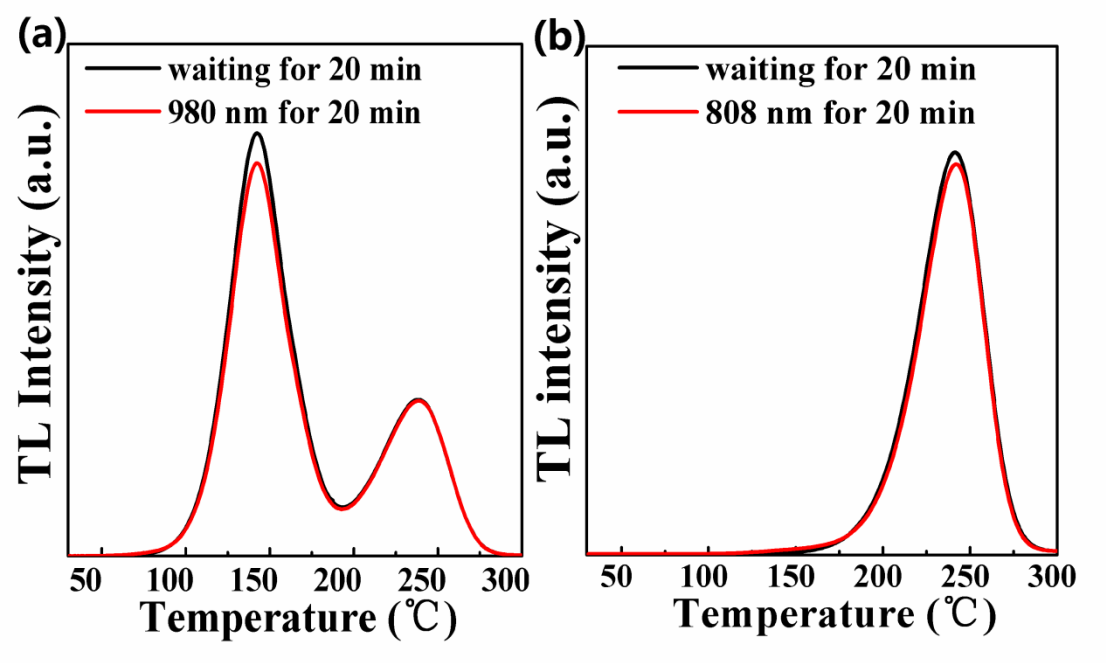

Figure S20. (a) The TL curves of sample BS:Eu^{2+}, Tm^{3+}, Ho^{3+}, Dy^{3+}. Black curve is measured 20 min after the thermal cleaning of trap I, and red curve is after the thermal cleaning of trap I and then illuminated by 980 nm for 20 min. (b) The TL curves of sample BS: Eu^{2+}, Tm^{3+}, Ho^{3+}, Dy^{3+}. Black curve is measured 20 min after the thermal cleaning of trap I, and red curve is after the thermal cleaning of trap I and then illuminated by 808 nm for 20 min.

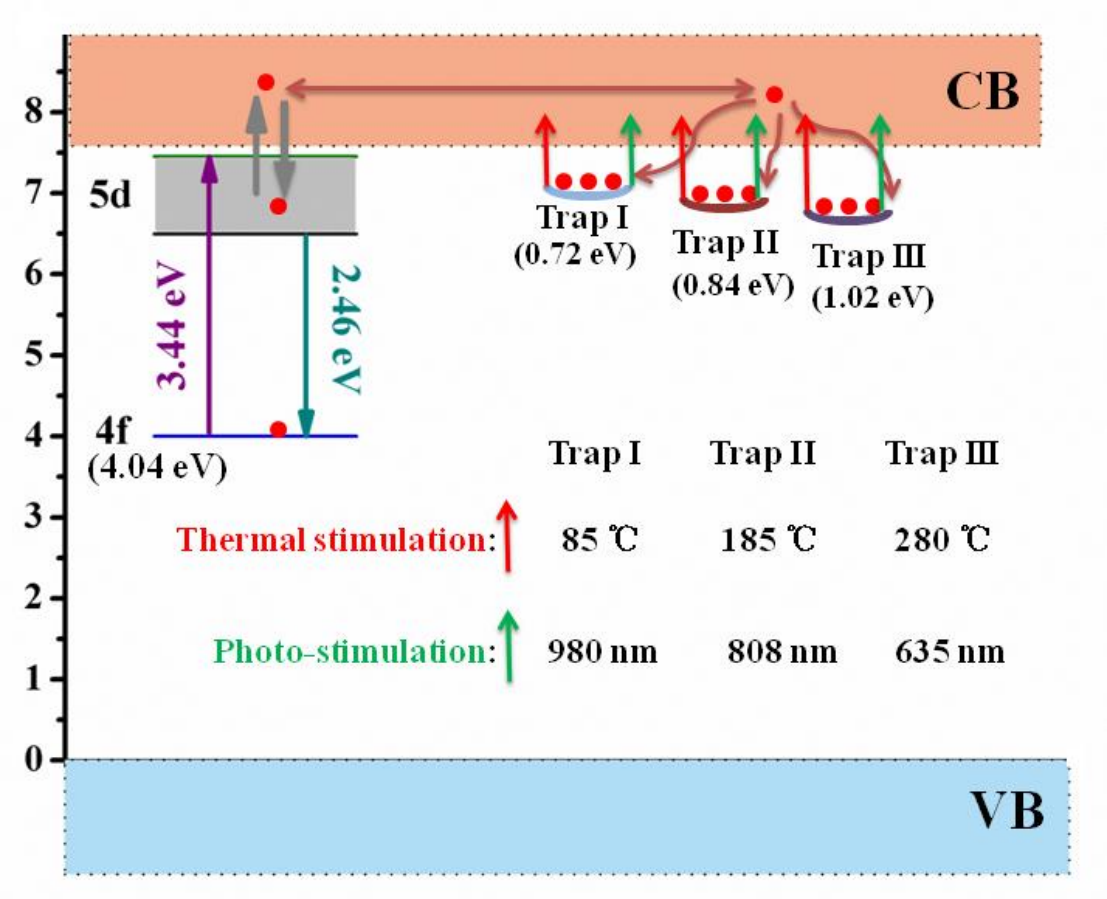

Figure S21. Energy level schematic diagram of the multi-level ODS mechanism of BS: Eu^{2+}, Tm^{3+}, Ho^{3+}, Dy^{3+} phosphor.

Table S1. The trap depths of samples BS:0.005Eu^{2+}+Ln^{3+} (Ln=Ho, Dy, Tm and Er); T_M is the temperature of each TL peak; T_1 and T_2 are the lower and higher temperatures at half maximum, respectively. *E (eV)* and *C (eV)* represent the experimental and theoretical trap depths. The trap distribution (E_T) is estimated by the following equation $E_T = T_2/500 - T_1/500$.

Eu^{2+}+Ln^{3+}	T_1 *(K)*	T_2*(K)*	T_m *(K)*	*E (eV)*	*C (eV)*	E_T *(eV)*
Ln=Ho	343.0	379.6	361.6	0.723	0.82	0.073
Ln=Dy	397.2	441.4	420.5	0.841	0.95	0.088
Ln=Tm	478.5	532.0	509.3	1.02	1.5	0.107
Ln=Er	-	-	-	-	0.62	-
Ln=Yb	-	-	-	-	2.78	-