

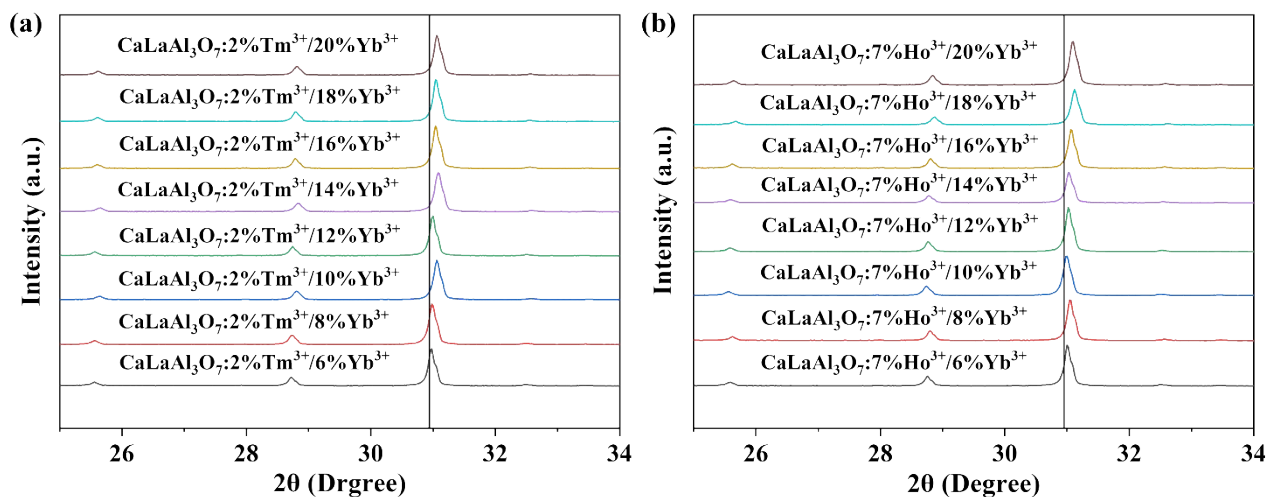
## Supporting Information

### Upconversion phosphors of $\text{CaLaAl}_3\text{O}_7\text{:RE}^{3+}/\text{Yb}^{3+}$ (RE=Tm, Ho) and their multifunctional applications for multi-color anti-counterfeiting and laser displays

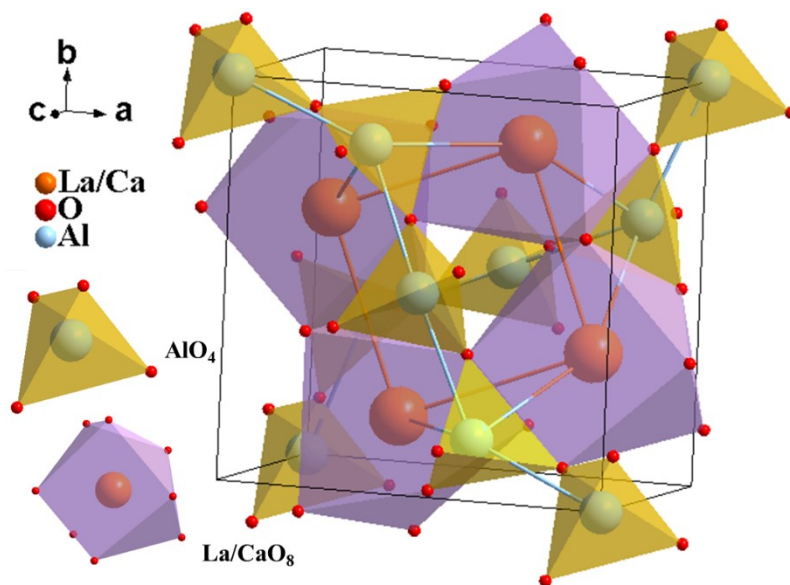
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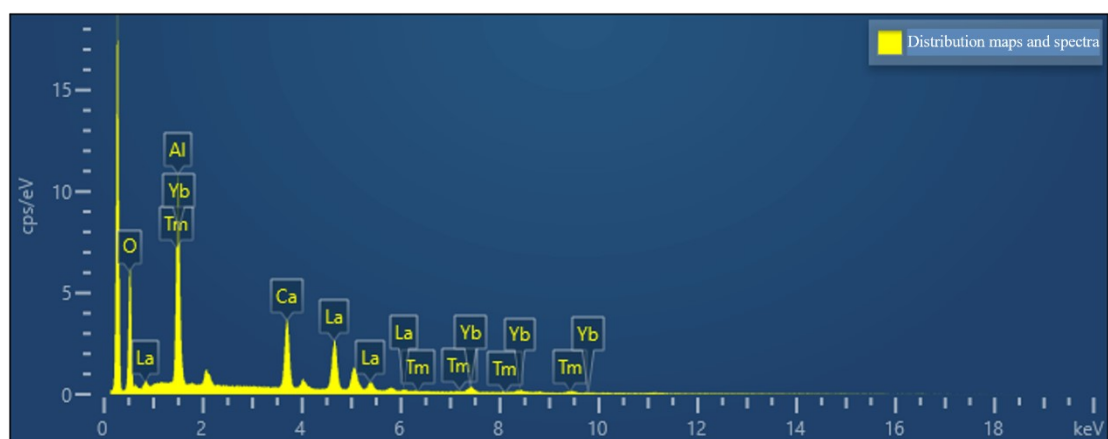
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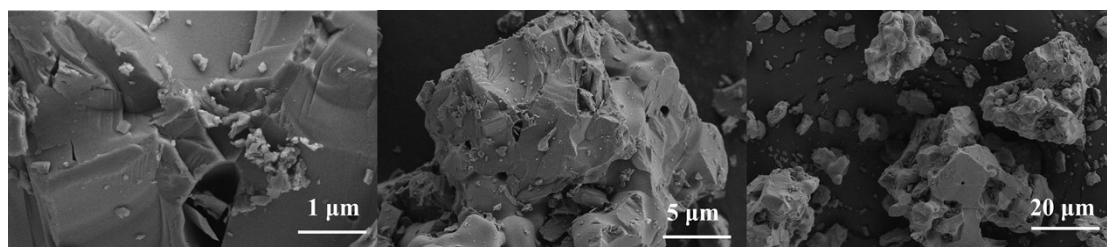
**Fig. S1** XRD patterns peak shifts: **(a)**  $\text{CaLaAl}_3\text{O}_7\text{:}2\%\text{Tm}^{3+}/z_1\%\text{Yb}^{3+}$  ( $z_1 = 6, 8, 10, 12, 14, 16, 18, 20$ ) powder samples **(b)**  $\text{CaLaAl}_3\text{O}_7\text{:}7\%\text{Ho}^{3+}/z_2\%\text{Yb}^{3+}$  ( $z_2 = 6, 8, 10, 12, 14, 16, 18, 20$ ) powder samples.



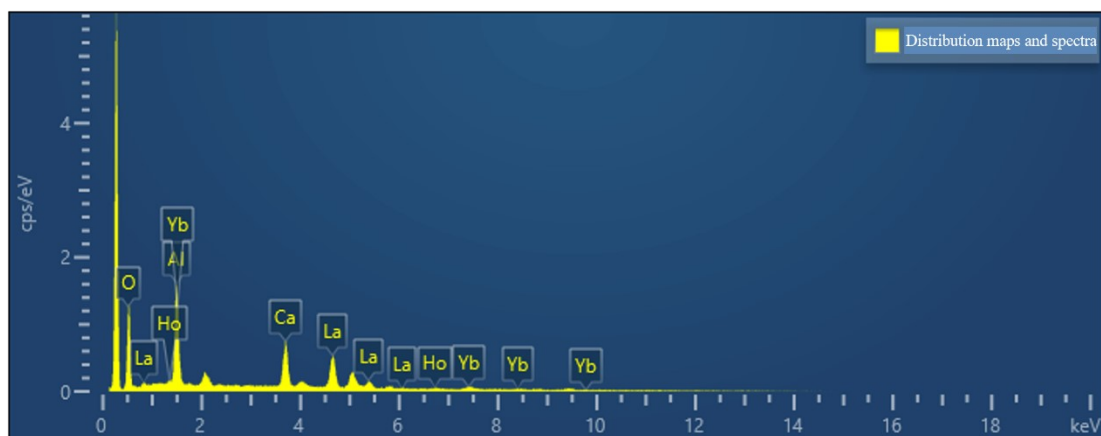
**Fig. S2** Crystal structure information of the  $\text{CaLaAl}_3\text{O}_7$  matrix.



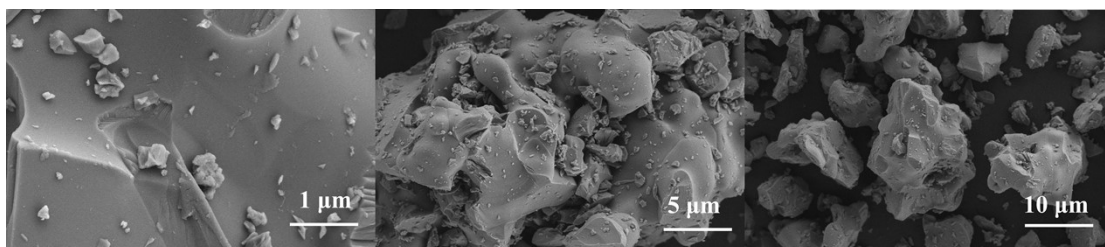
**Fig. S3** EDS element distribution of the  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/18\%\text{Yb}^{3+}$  phosphor.



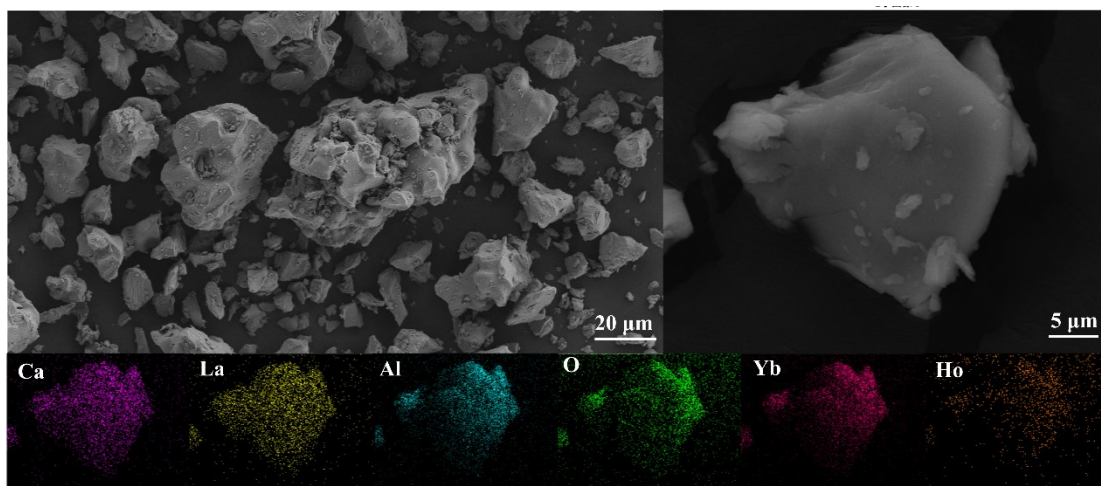
**Fig. S4** SEM images of the  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/18\%\text{Yb}^{3+}$  representative at different magnifications.



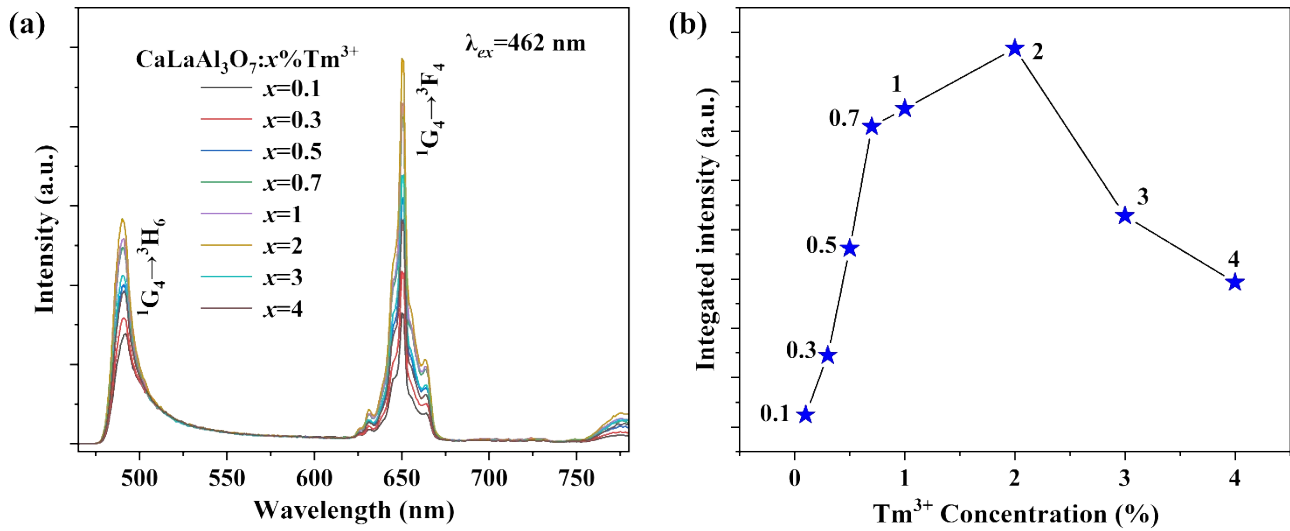
**Fig. S5** EDS element distribution of the  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/14\%\text{Yb}^{3+}$  phosphor.



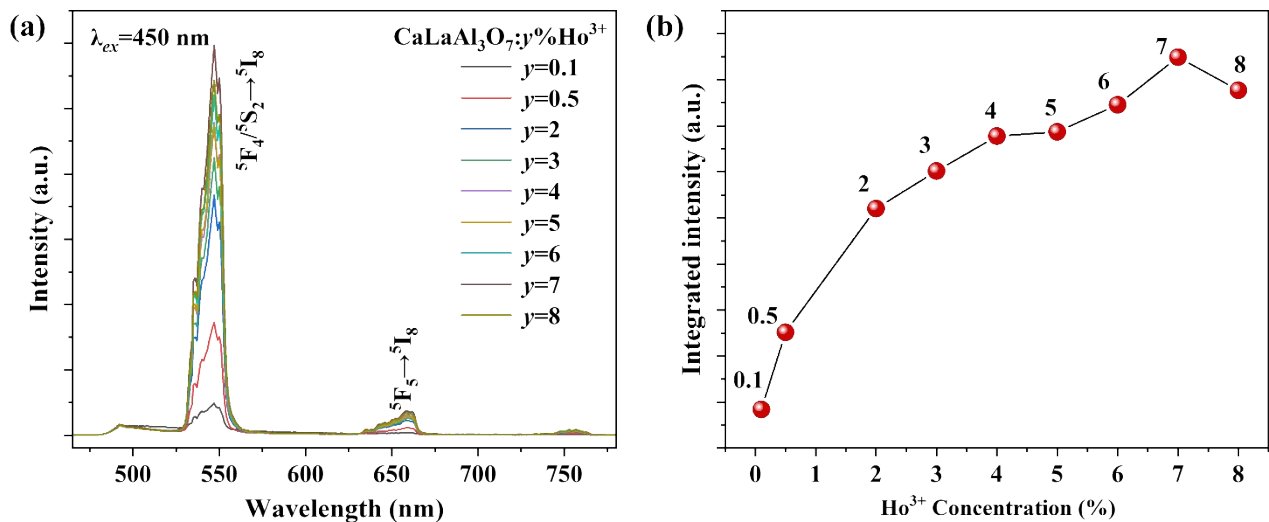
**Fig. S6** SEM images of the  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/14\%\text{Yb}^{3+}$  representative at different magnifications.



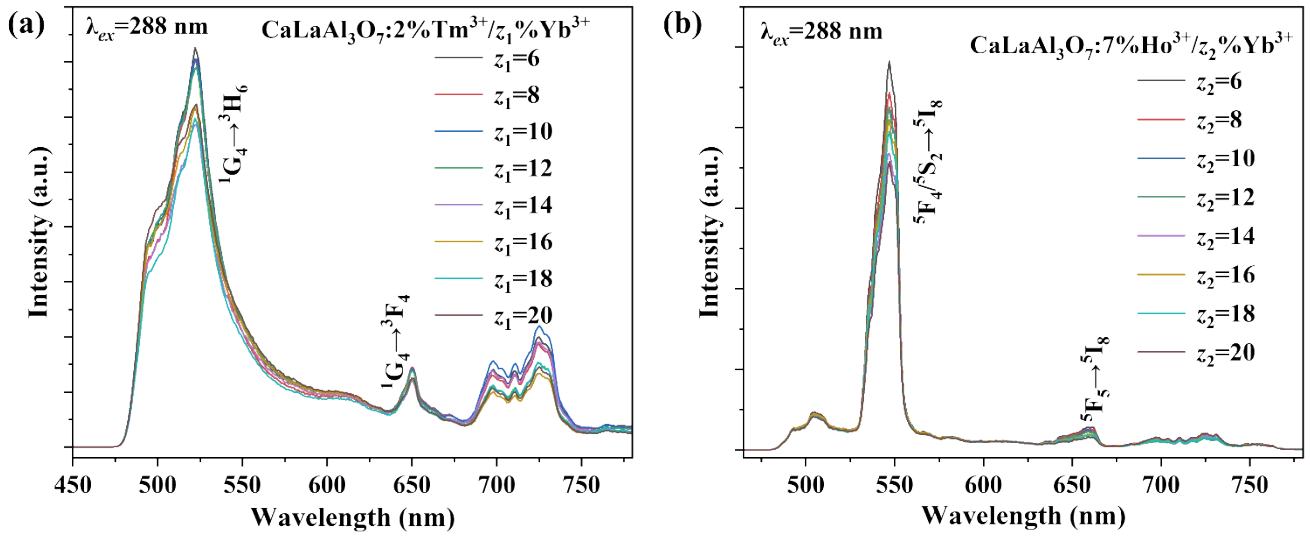
**Fig. S7** SEM photograph and EDS elemental mapping image of the  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/14\%\text{Yb}^{3+}$  phosphor.



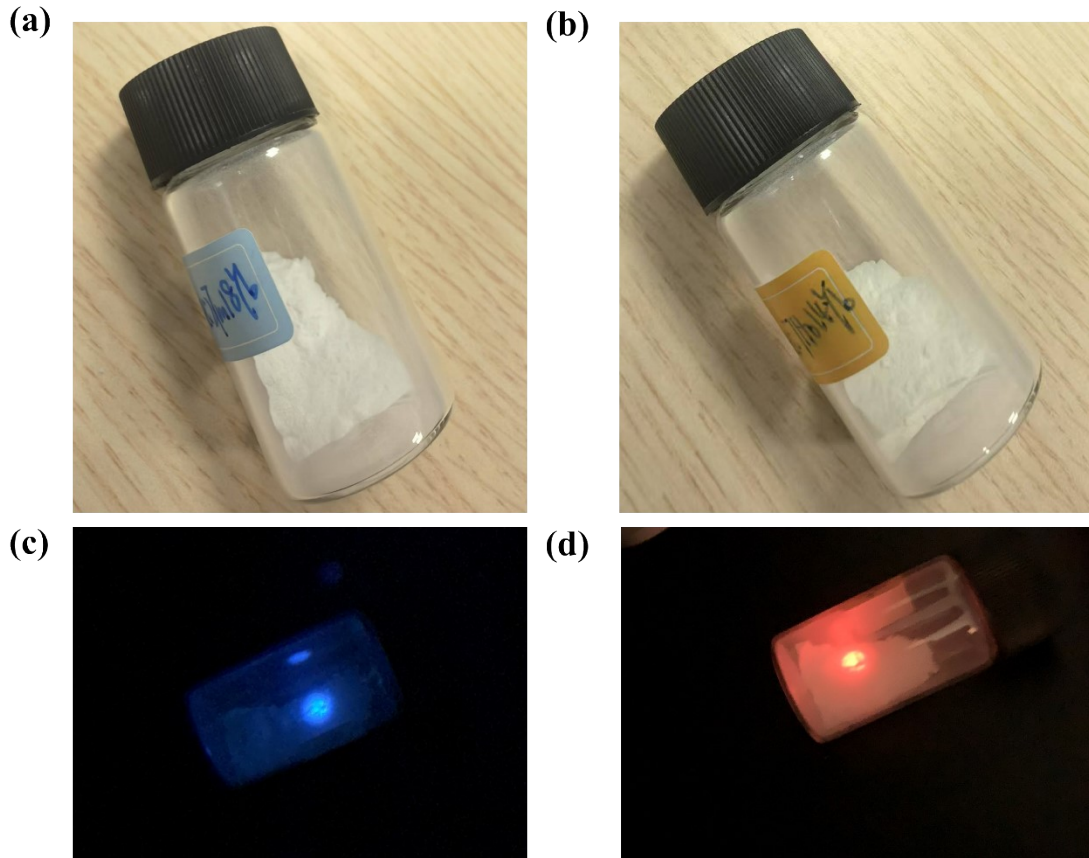
**Fig. S8** (a) PL of  $\text{CaLaAl}_3\text{O}_7:x\%\text{Tm}^{3+}$  ( $x = 0.1, 0.3, 0.5, 0.7, 1, 2, 3, 4$ ) phosphors; (b) Upconversion luminescence integrated intensity in the range of 625–675 nm varying with  $\text{Tm}^{3+}$  concentration



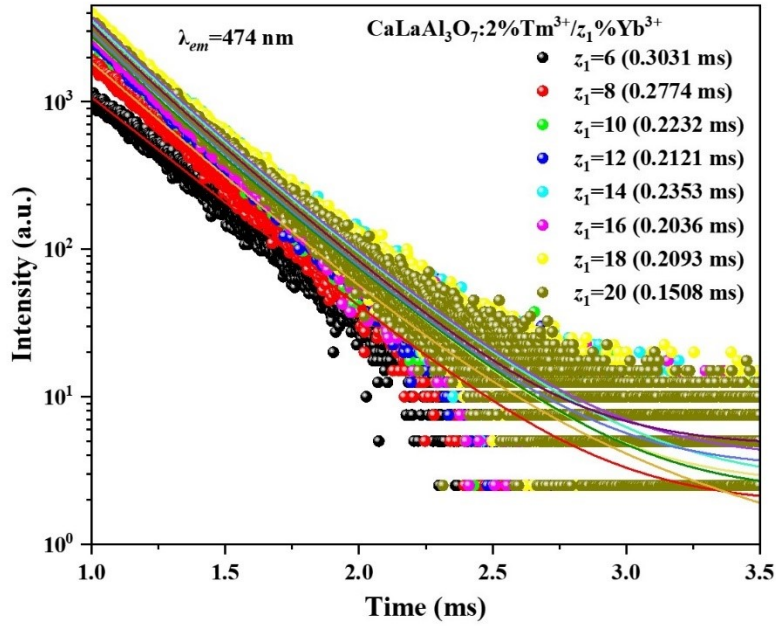
**Fig. S9** (a) PL of  $\text{CaLaAl}_3\text{O}_7:y\%\text{Ho}^{3+}$  ( $y = 0.1, 0.5, 2, 3, 4, 5, 6, 7, 8$ ) phosphors; (b) Upconversion luminescence integrated intensity in the range of 528–560 nm varying with  $\text{Ho}^{3+}$  concentration



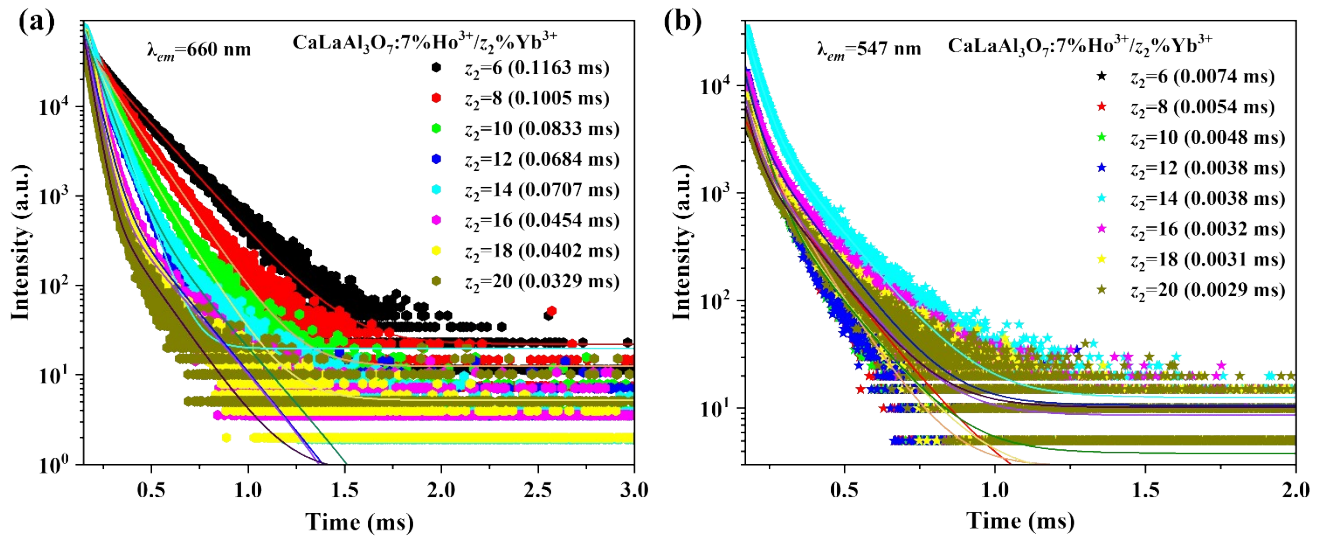
**Fig. S10** (a) PL of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/z_1\%\text{Yb}^{3+}$  ( $z_1 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors under 288 nm excitation; (b) PL of  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/z_2\%\text{Yb}^{3+}$  ( $z_2 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors under 288 nm excitation



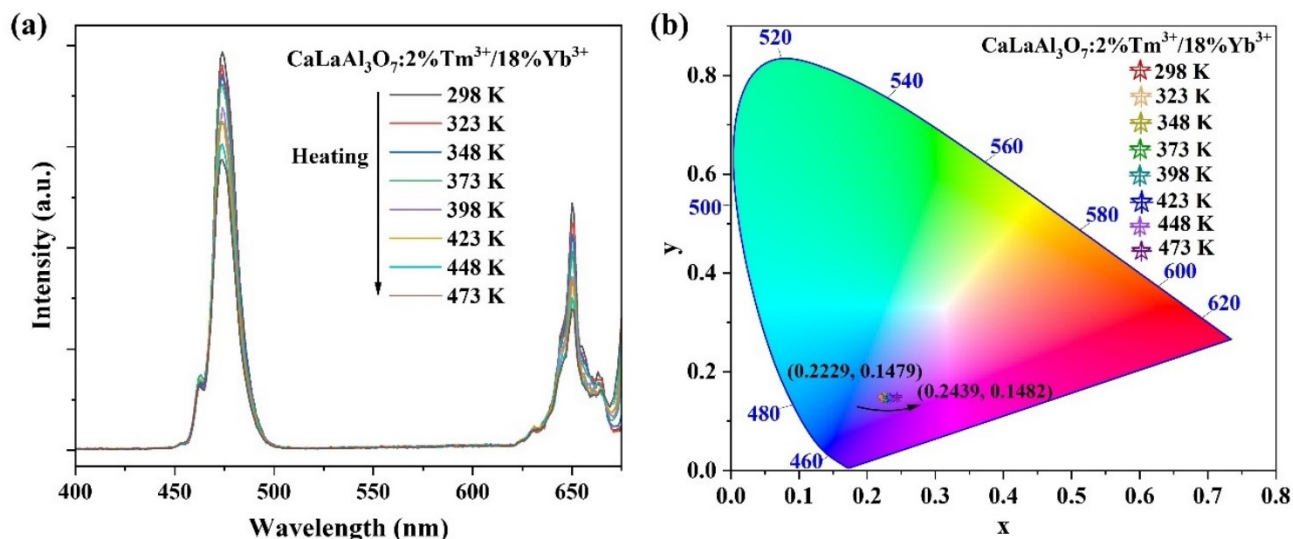
**Fig. S11** (a)  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/18\text{Yb}^{3+}$  sample under natural light; (b)  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/14\text{Yb}^{3+}$  phosphor under natural light; (c)  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/18\text{Yb}^{3+}$  sample under 980 nm laser excitation; (d)  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/14\text{Yb}^{3+}$  phosphor under 980 nm laser excitation



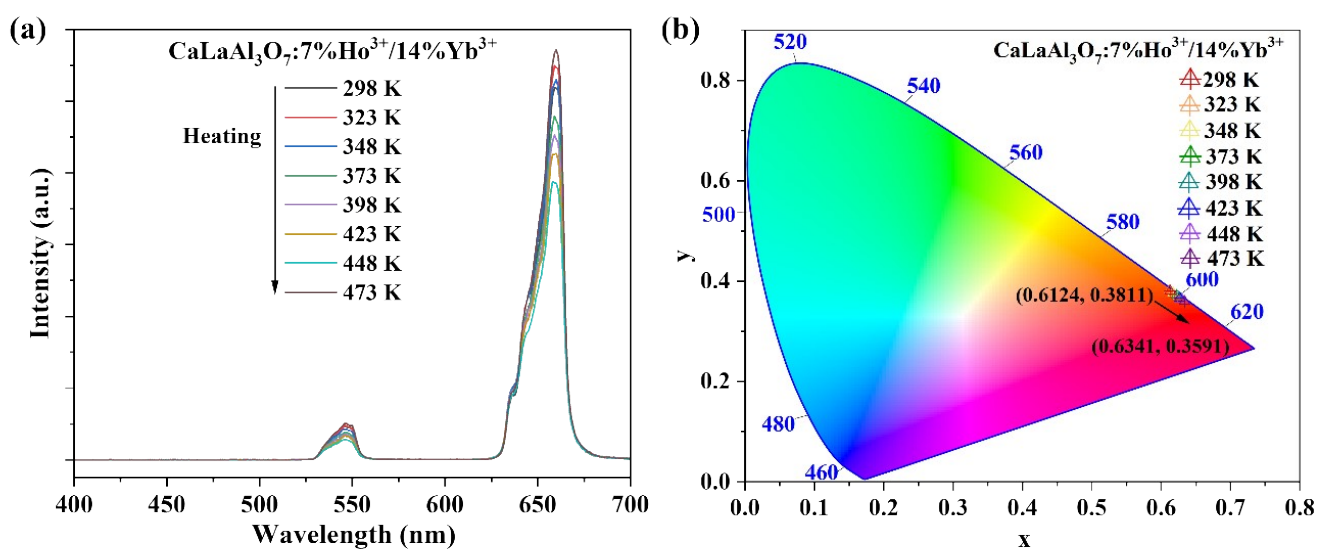
**Fig. S12** Upconversion emission decay curves of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/z_1\%\text{Yb}^{3+}$  ( $z_1 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors at a monitoring emission wavelength of 474 nm upon 980 nm pulsed excitation.



**Fig. S13** Upconversion emission decay curves of  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/z_2\%\text{Yb}^{3+}$  ( $z_2 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors at a monitoring emission wavelength of (a) 660 nm, (b) 547 nm upon 980 nm pulsed excitation.



**Fig. S14 (a)** Thermal stability of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/18\%\text{Yb}^{3+}$  phosphor between 298-473K, with a testing interval of 25 K **(b)** Changes in chromaticity coordinates of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/18\%\text{Yb}^{3+}$  phosphor at different temperatures



**Fig. S15 (a)** Thermal stability of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/18\%\text{Yb}^{3+}$  phosphor between 298-473K, with a testing interval of 25 K **(b)** Changes in chromaticity coordinates of  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/14\%\text{Yb}^{3+}$  phosphor at different temperatures

**Table S1** Results of EDS mapping of the  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/18\%\text{Yb}^{3+}$  representative.

| Element | wt%    | Wt% Sigma | At%    |
|---------|--------|-----------|--------|
| O       | 38.30  | 0.43      | 66.87  |
| Al      | 20.02  | 0.38      | 20.73  |
| Ca      | 8.72   | 0.14      | 6.07   |
| La      | 25.22  | 0.34      | 5.07   |
| Tm      | 0.93   | 0.32      | 0.15   |
| Yb      | 6.82   | 0.39      | 1.10   |
| Total   | 100.00 |           | 100.00 |

**Table S2** Results of EDS mapping of the  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/14\%\text{Yb}^{3+}$  representative.

| Element | wt%    | Wt% Sigma | At%    |
|---------|--------|-----------|--------|
| O       | 28.98  | 0.65      | 61.65  |
| Al      | 14.61  | 0.39      | 18.42  |
| Ca      | 10.74  | 0.30      | 9.12   |
| La      | 37.22  | 0.81      | 9.12   |
| Ho      | 2.87   | 1.02      | 0.59   |
| Yb      | 5.57   | 0.85      | 1.10   |
| Total   | 100.00 |           | 100.00 |

**Table S3** CIE parameters of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/z_1\%\text{Yb}^{3+}$  ( $z_1 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors.

| Yb <sup>3+</sup> Concentration<br>$z_1$ | Excitation<br>wavelength<br>(nm) | CIE coordinate |        |
|-----------------------------------------|----------------------------------|----------------|--------|
|                                         |                                  | $x$            | $y$    |
| 6                                       | 288                              | 0.2803         | 0.6215 |
| 8                                       | 288                              | 0.2782         | 0.6231 |
| 10                                      | 288                              | 0.2810         | 0.6206 |
| 12                                      | 288                              | 0.2818         | 0.6192 |
| 14                                      | 288                              | 0.2893         | 0.6135 |
| 16                                      | 288                              | 0.2870         | 0.6143 |
| 18                                      | 288                              | 0.2845         | 0.6177 |
| 20                                      | 288                              | 0.2855         | 0.6135 |

**Table S4** CIE parameters of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/z_1\%\text{Yb}^{3+}$  ( $z_1 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors.

| Yb <sup>3+</sup> Concentration<br>$z_1$ | Excitation<br>wavelength<br>(nm) | CIE coordinate |        |
|-----------------------------------------|----------------------------------|----------------|--------|
|                                         |                                  | $x$            | $y$    |
| 6                                       | 355                              | 0.2891         | 0.5838 |
| 8                                       | 355                              | 0.2897         | 0.5829 |
| 10                                      | 355                              | 0.2909         | 0.5832 |
| 12                                      | 355                              | 0.2875         | 0.5850 |
| 14                                      | 355                              | 0.2957         | 0.5827 |
| 16                                      | 355                              | 0.2908         | 0.5847 |
| 18                                      | 355                              | 0.2894         | 0.5856 |
| 20                                      | 355                              | 0.2882         | 0.5858 |

**Table S5** CIE parameters of  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/z_2\%\text{Yb}^{3+}$  ( $z_2 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors.

| Yb <sup>3+</sup> Concentration<br>$z_2$ | Excitation<br>wavelength<br>(nm) | CIE coordinate |        |
|-----------------------------------------|----------------------------------|----------------|--------|
|                                         |                                  | $x$            | $y$    |
| 6                                       | 288                              | 0.2926         | 0.6773 |
| 8                                       | 288                              | 0.2935         | 0.6760 |
| 10                                      | 288                              | 0.2937         | 0.6751 |
| 12                                      | 288                              | 0.2933         | 0.6754 |
| 14                                      | 288                              | 0.2952         | 0.6706 |
| 16                                      | 288                              | 0.2935         | 0.6738 |
| 18                                      | 288                              | 0.2934         | 0.6749 |
| 20                                      | 288                              | 0.2950         | 0.6717 |

**Table S6** CIE parameters of  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/z_2\%\text{Yb}^{3+}$  ( $z_2 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors.

| Yb <sup>3+</sup> Concentration<br>$z_2$ | Excitation<br>wavelength<br>(nm) | CIE coordinate |        |
|-----------------------------------------|----------------------------------|----------------|--------|
|                                         |                                  | $x$            | $y$    |
| 6                                       | 361                              | 0.2827         | 0.6992 |
| 8                                       | 361                              | 0.2824         | 0.6996 |
| 10                                      | 361                              | 0.2821         | 0.6997 |
| 12                                      | 361                              | 0.2822         | 0.6996 |
| 14                                      | 361                              | 0.2819         | 0.6990 |
| 16                                      | 361                              | 0.2814         | 0.7005 |
| 18                                      | 361                              | 0.2816         | 0.6997 |
| 20                                      | 361                              | 0.2817         | 0.6995 |

**Table S7** CIE parameters of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/z_1\%\text{Yb}^{3+}$  ( $z_1 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors.

| Yb <sup>3+</sup> Concentration<br>$z_1$ | Excitation<br>wavelength<br>(nm) | CIE coordinate |        |
|-----------------------------------------|----------------------------------|----------------|--------|
|                                         |                                  | $x$            | $y$    |
| 6                                       | 462                              | 0.3404         | 0.4105 |
| 8                                       | 462                              | 0.3404         | 0.4091 |
| 10                                      | 462                              | 0.3370         | 0.4145 |
| 12                                      | 462                              | 0.3394         | 0.4112 |
| 14                                      | 462                              | 0.3318         | 0.4273 |
| 16                                      | 462                              | 0.3313         | 0.4269 |
| 18                                      | 462                              | 0.3354         | 0.4225 |
| 20                                      | 462                              | 0.3335         | 0.4225 |

**Table S8** CIE parameters of  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/z_2\%\text{Yb}^{3+}$  ( $z_2 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors.

| Yb <sup>3+</sup> Concentration<br>$z_2$ | Excitation<br>wavelength<br>(nm) | CIE coordinate |        |
|-----------------------------------------|----------------------------------|----------------|--------|
|                                         |                                  | $x$            | $y$    |
| 6                                       | 450                              | 0.2756         | 0.7041 |
| 8                                       | 450                              | 0.2749         | 0.7044 |
| 10                                      | 450                              | 0.2748         | 0.7041 |
| 12                                      | 450                              | 0.2745         | 0.7043 |
| 14                                      | 450                              | 0.2738         | 0.7043 |
| 16                                      | 450                              | 0.2736         | 0.7054 |
| 18                                      | 450                              | 0.2730         | 0.7054 |
| 20                                      | 450                              | 0.2732         | 0.7048 |

**Table S9** CIE parameters of  $\text{CaLaAl}_3\text{O}_7:2\%\text{Tm}^{3+}/z_1\%\text{Yb}^{3+}$  ( $z_1 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors.

| Yb <sup>3+</sup> Concentration<br>$z_1$ | Excitation<br>wavelength<br>(nm) | CIE coordinate |        |
|-----------------------------------------|----------------------------------|----------------|--------|
|                                         |                                  | $x$            | $y$    |
| 6                                       | 980                              | 0.2272         | 0.1740 |
| 8                                       | 980                              | 0.2140         | 0.1609 |
| 10                                      | 980                              | 0.2052         | 0.1528 |
| 12                                      | 980                              | 0.2072         | 0.1497 |
| 14                                      | 980                              | 0.2023         | 0.1498 |
| 16                                      | 980                              | 0.2017         | 0.1493 |
| 18                                      | 980                              | 0.1993         | 0.1467 |
| 20                                      | 980                              | 0.2011         | 0.1489 |

**Table S10** CIE parameters of  $\text{CaLaAl}_3\text{O}_7:7\%\text{Ho}^{3+}/z_2\%\text{Yb}^{3+}$  ( $z_2 = 6, 8, 10, 12, 14, 16, 18, 20$ ) phosphors.

| Yb <sup>3+</sup> Concentration<br>$z_2$ | Excitation<br>wavelength<br>(nm) | CIE coordinate |        |
|-----------------------------------------|----------------------------------|----------------|--------|
|                                         |                                  | $x$            | $y$    |
| 6                                       | 980                              | 0.6131         | 0.3803 |
| 8                                       | 980                              | 0.6094         | 0.3845 |
| 10                                      | 980                              | 0.6010         | 0.3931 |
| 12                                      | 980                              | 0.6017         | 0.3927 |
| 14                                      | 980                              | 0.5964         | 0.3986 |
| 16                                      | 980                              | 0.5863         | 0.4083 |
| 18                                      | 980                              | 0.5779         | 0.4163 |
| 20                                      | 980                              | 0.5761         | 0.4183 |