

Supplementary Information for

**Revisit the SrAl<sub>2</sub>O<sub>4</sub>:Eu<sup>2+</sup>,Nd<sup>3+</sup> persistent phosphor**

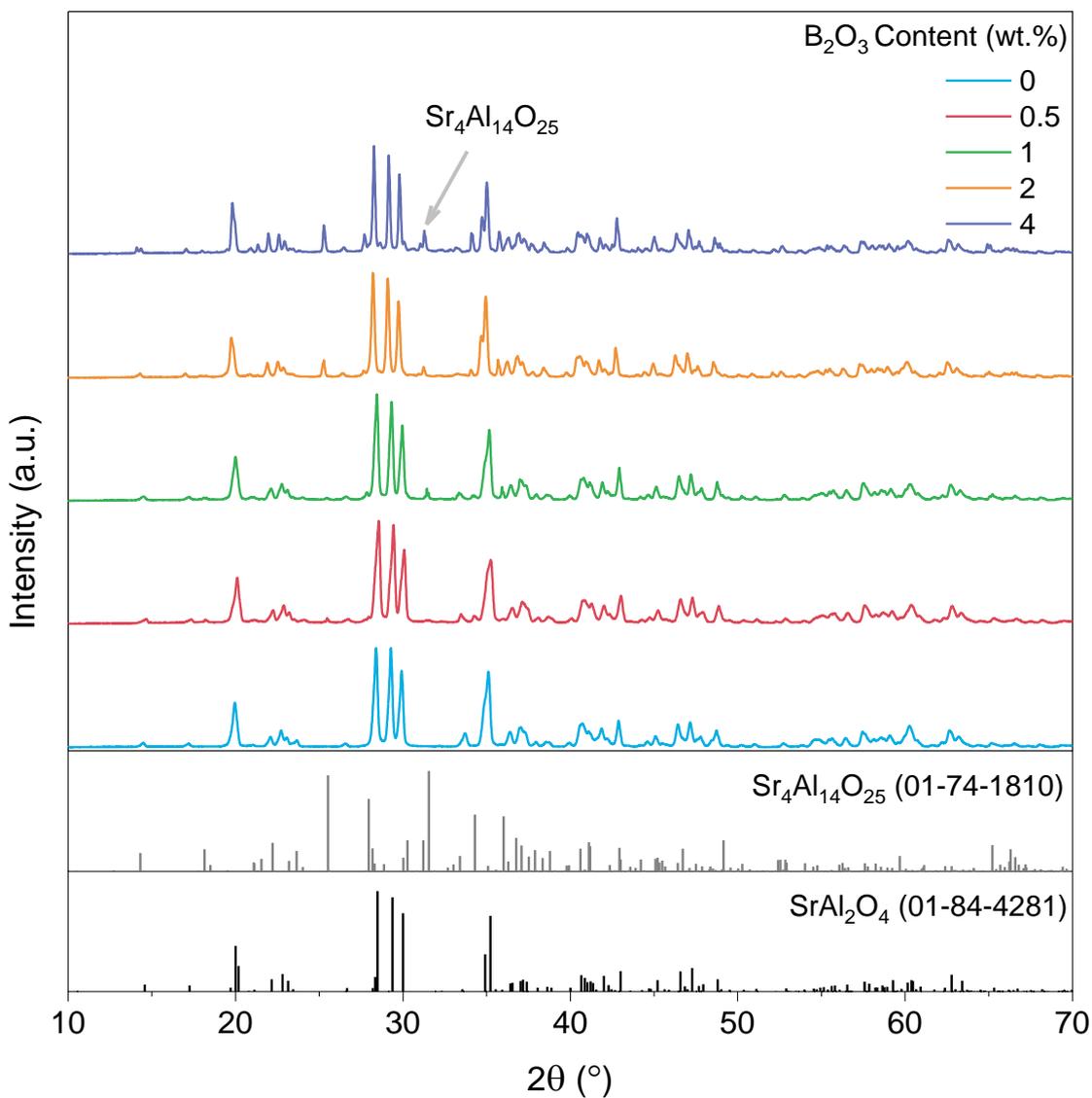
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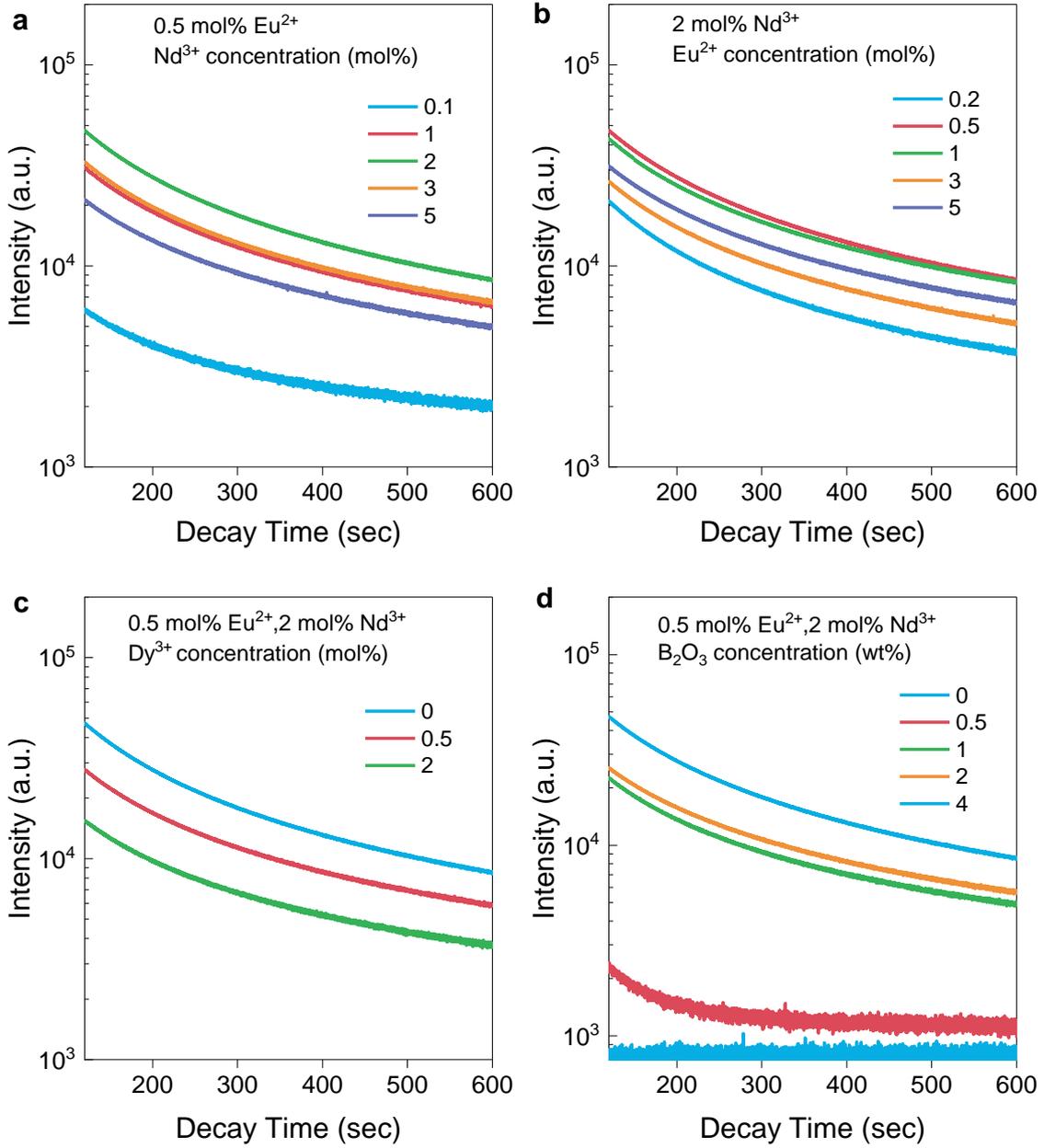
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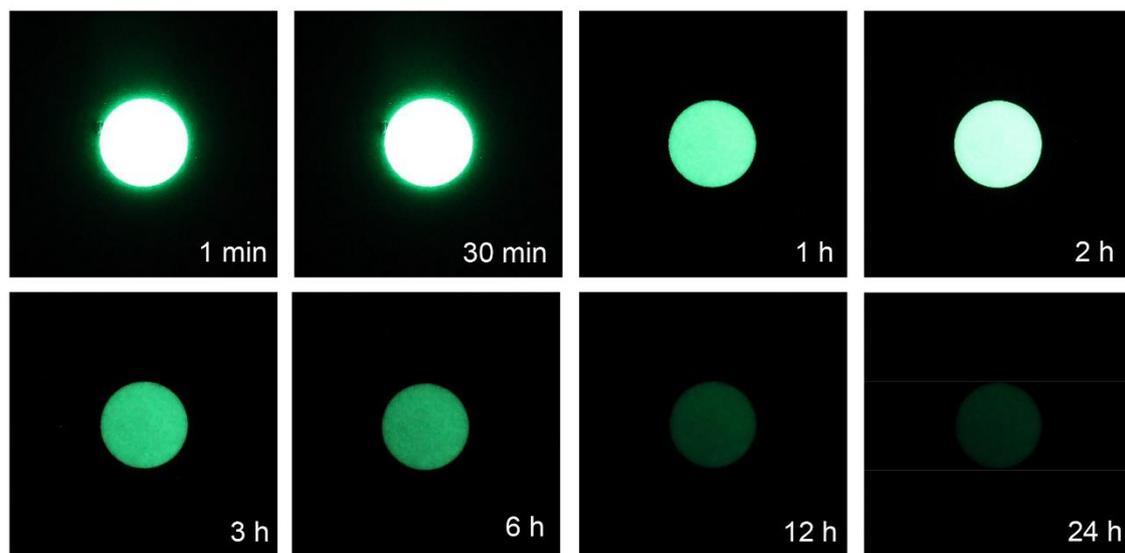
**Brief summary** – This file contains: X-ray diffraction patterns of SAO:Eu,Nd; PersL decay curves of SAO:Eu,Nd; green and NIR PersL imaging; green and NIR PersL intensities of SAO:Eu,Nd; excitation efficacy of 450 nm LED with varying output intensities; PersL excitation spectrum of SrAl<sub>2</sub>O<sub>4</sub>:Eu<sup>2+</sup>,Dy<sup>3+</sup>; TL spectra of SAO:Eu,Nd; PL excitation spectra of SAO:Eu,Nd at RT and LNT; and PL excitation and emission spectra of SrAl<sub>2</sub>O<sub>4</sub> host.



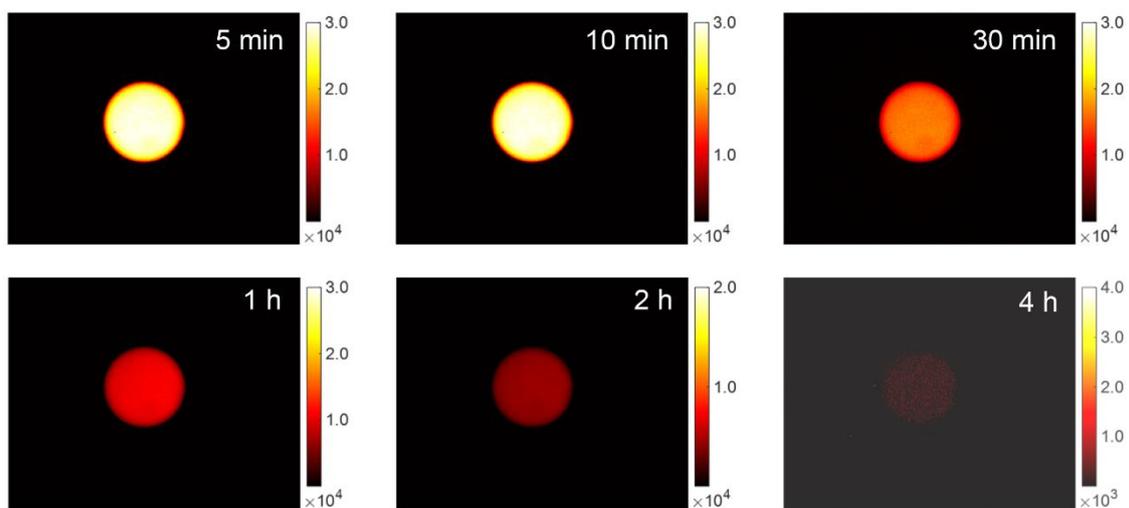
**Figure S1** X-ray diffraction patterns of SrAl<sub>2</sub>O<sub>5</sub>:0.5%Eu<sup>2+</sup>,2%Nd<sup>3+</sup> phosphors with varying B<sub>2</sub>O<sub>3</sub> contents (0–4 wt.%). The indexing of monoclinic SrAl<sub>2</sub>O<sub>4</sub> (ICDD #01-84-4281) and orthorhombic Sr<sub>4</sub>Al<sub>14</sub>O<sub>25</sub> (ICDD #01-74-1810) are also presented. As the B<sub>2</sub>O<sub>3</sub> content increases, Sr<sub>4</sub>Al<sub>14</sub>O<sub>25</sub> phase appears.



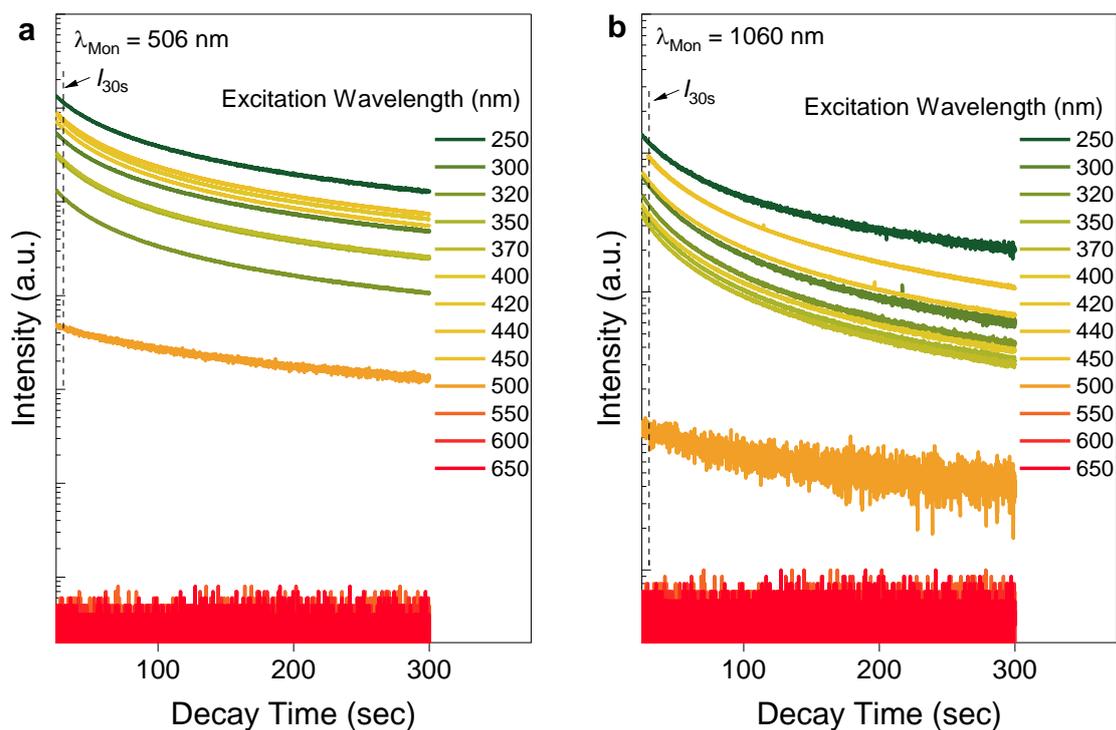
**Figure S2** Determination of optimal  $\text{Eu}^{2+}$ ,  $\text{Nd}^{3+}$ ,  $\text{Dy}^{3+}$ , and  $\text{B}_2\text{O}_3$  contents in  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+},\text{Nd}^{3+}$  persistent phosphors according to the PersL decay of  $\text{Nd}^{3+}$ . (a) Phosphors with 0.5 mol%  $\text{Eu}^{2+}$  and 0.1–5 mol%  $\text{Nd}^{3+}$ . (b) Phosphors with 2 mol%  $\text{Nd}^{3+}$  and 0.2–5 mol%  $\text{Eu}^{2+}$ . (c) Phosphors with 0.5 mol%  $\text{Eu}^{2+}$ , 2 mol%  $\text{Nd}^{3+}$ , and 0–2 mol%  $\text{Dy}^{3+}$ . (d) Phosphors with 0.5 mol%  $\text{Eu}^{2+}$ , 2 mol%  $\text{Nd}^{3+}$ , and 0–4 wt.%  $\text{B}_2\text{O}_3$ . The samples were charged by a white LED flashlight for 1 min. The PersL decay curves were acquired by monitoring the 1060 nm emission of  $\text{Nd}^{3+}$ .



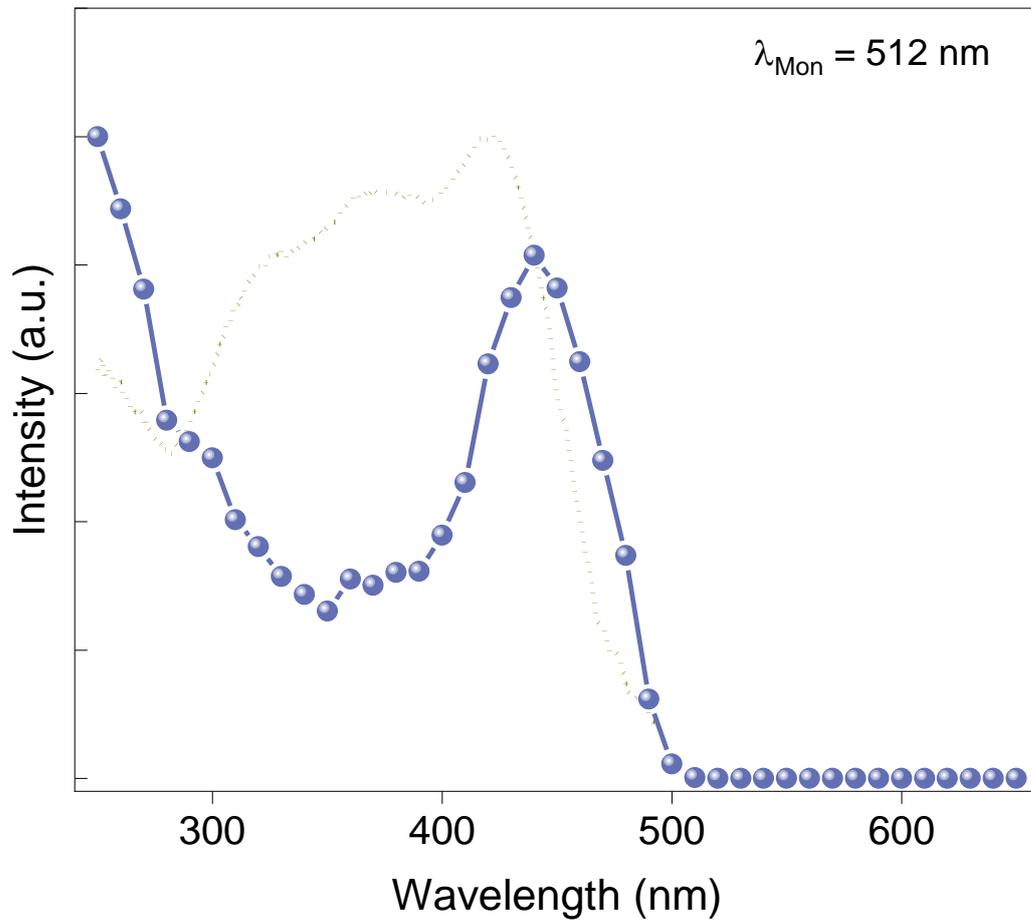
**Figure S3** Green PersL images of a SAO:Eu,Nd persistent phosphor disc taken at different decay points (1 min to 24 h) after illumination with a white LED flashlight for 1 min using a Canon digital camera. The imaging parameter was manual/ISO 400/3 s.



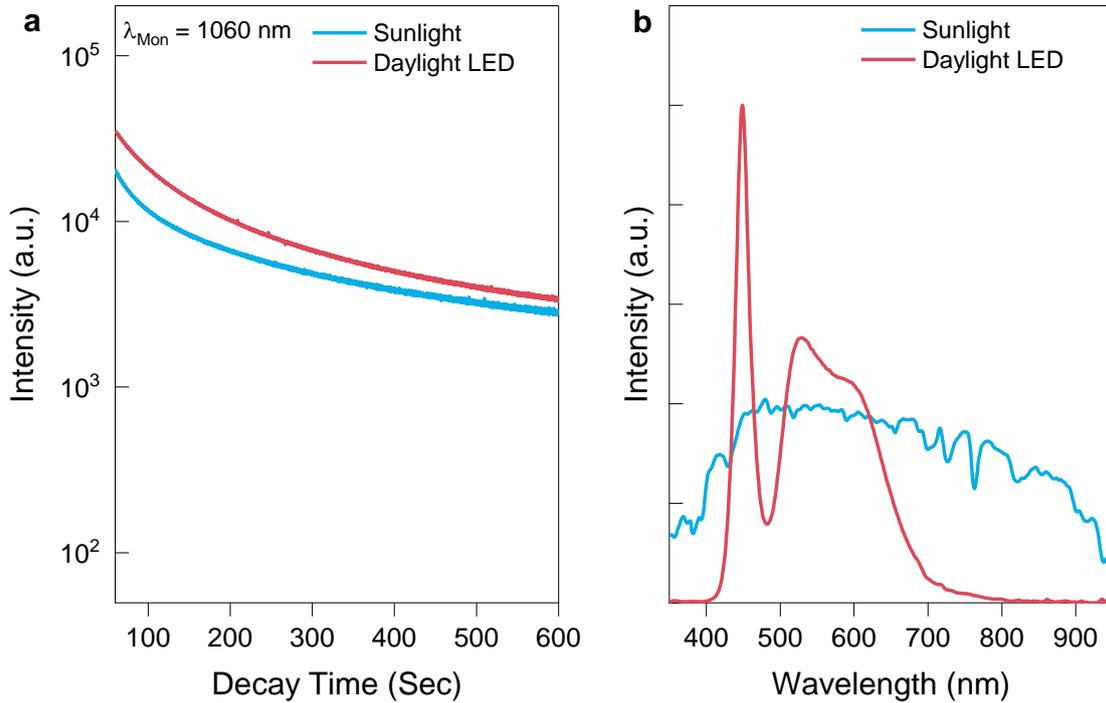
**Figure S4** NIR PersL images of a SAO:Eu,Nd persistent phosphor disc taken at different decay points (5 min to 4 h) after irradiated by a white LED flashlight for 1 min using a Teledyne Princeton Instruments NIRvana 640 InGaAs array camera. The exposure time was 30 s.



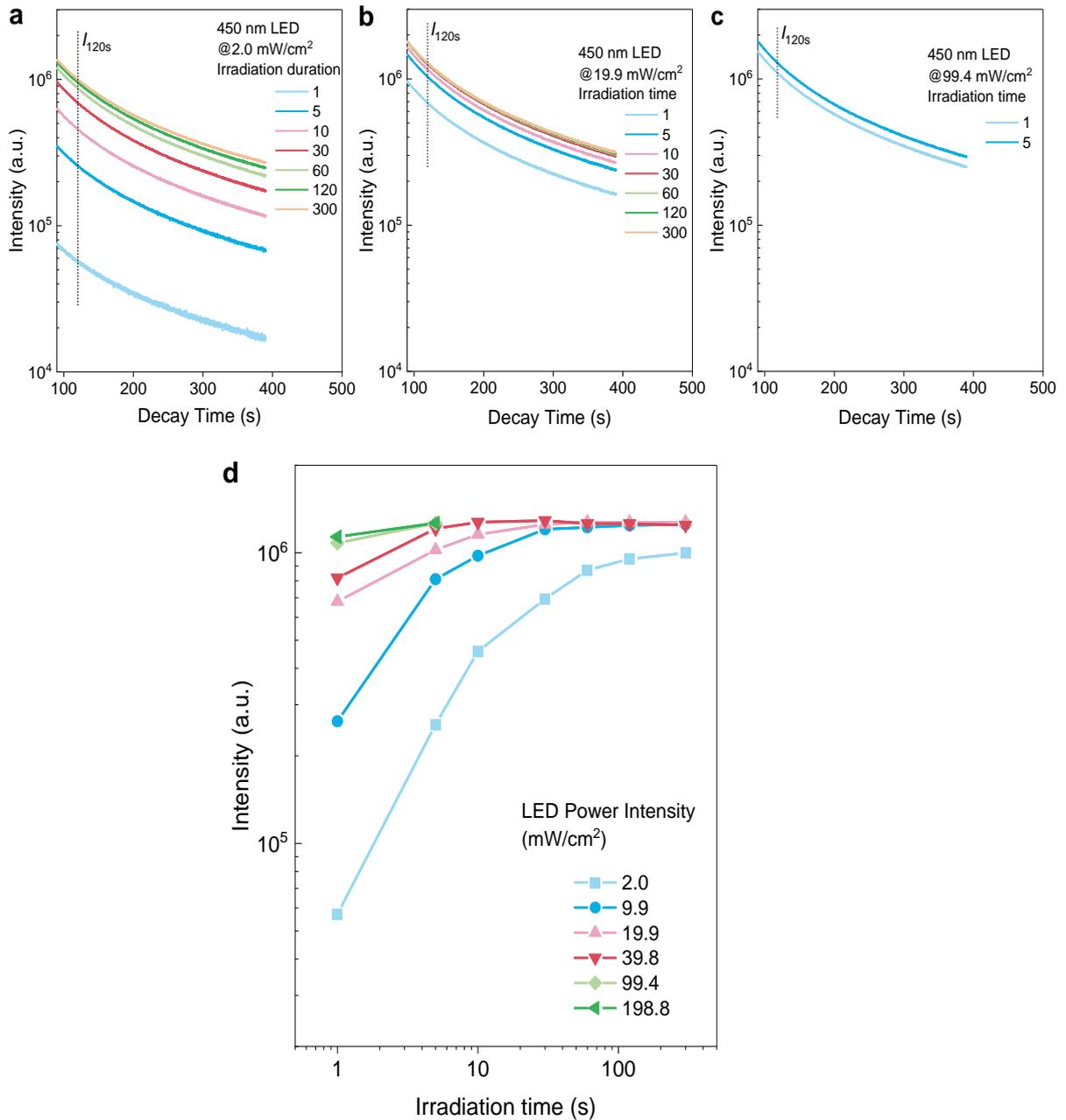
**Figure S5** Acquiring PersL excitation spectra of  $\text{Eu}^{2+}$  and  $\text{Nd}^{3+}$  in SAO:Eu,Nd persistent phosphor. (a) PersL decay curves by monitoring the 506 nm emission of  $\text{Eu}^{2+}$ . (b) PersL decay curves by monitoring the 1060 nm emission of  $\text{Nd}^{3+}$ . The sample was irradiated by monochromatic xenon light between 250–650 nm for 5 min in 10 nm steps. The PersL intensity at time of 30 s after the stoppage of the irradiation ( $I_{30s}$ ) was used to plot the PersL excitation spectra of  $\text{Eu}^{2+}$  and  $\text{Nd}^{3+}$  shown in Fig. 3a and Fig. 3b of the main text, respectively.



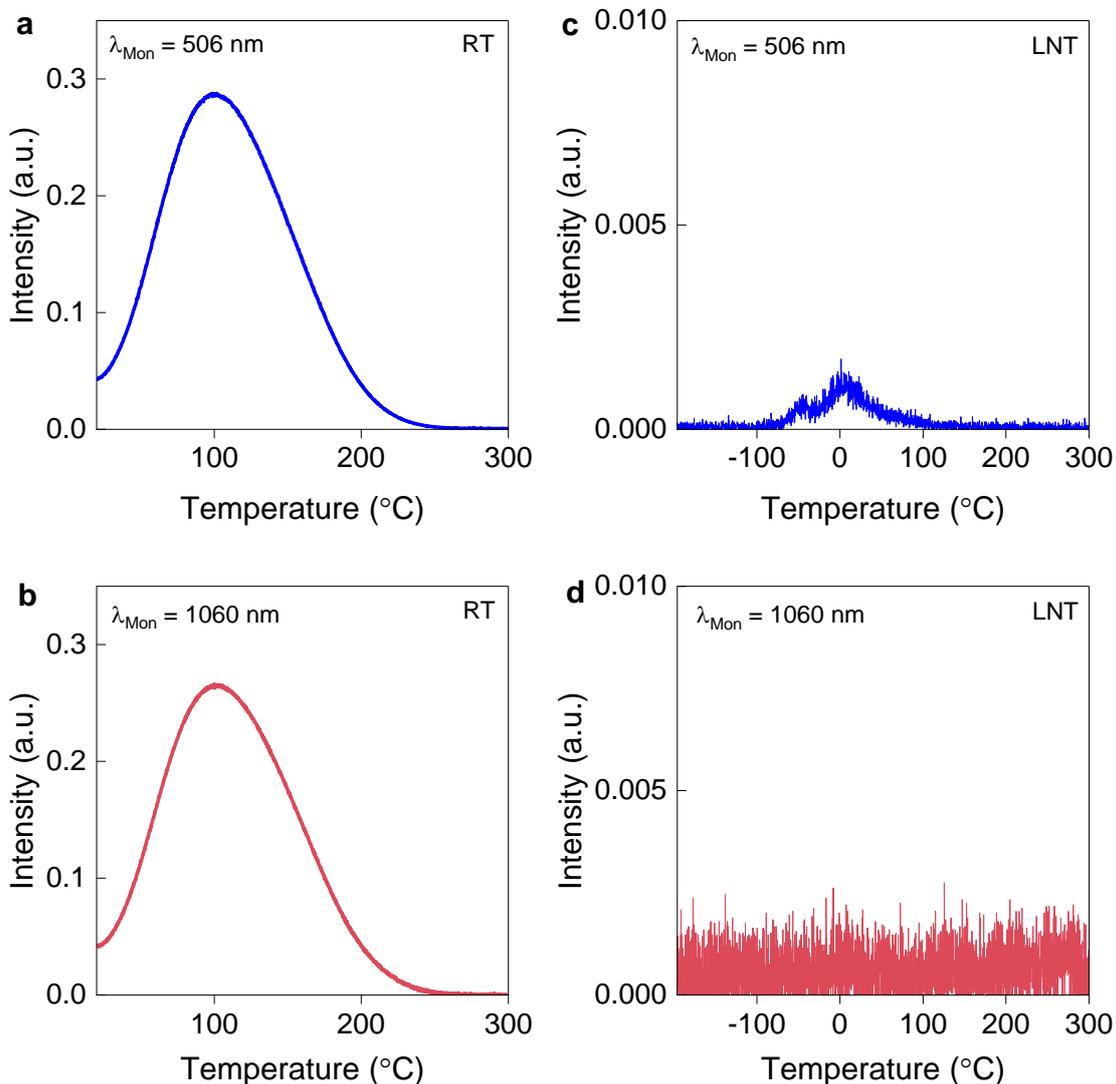
**Figure S6** PersL excitation spectrum of  $\text{SrAl}_2\text{O}_4:0.5\%\text{Eu}^{2+},2\%\text{Dy}^{3+}$  persistent phosphor. The method is the same as that used to obtain the PersL excitation spectra of  $\text{SAO};\text{Eu},\text{Nd}$  phosphor shown in Fig. 3 of the main text. The monitoring wavelength is 512 nm. The grey dot-line curve is the PL excitation spectrum of  $\text{SrAl}_2\text{O}_4:0.5\%\text{Eu}^{2+},2\%\text{Dy}^{3+}$  phosphor by monitoring the 512 nm emission.



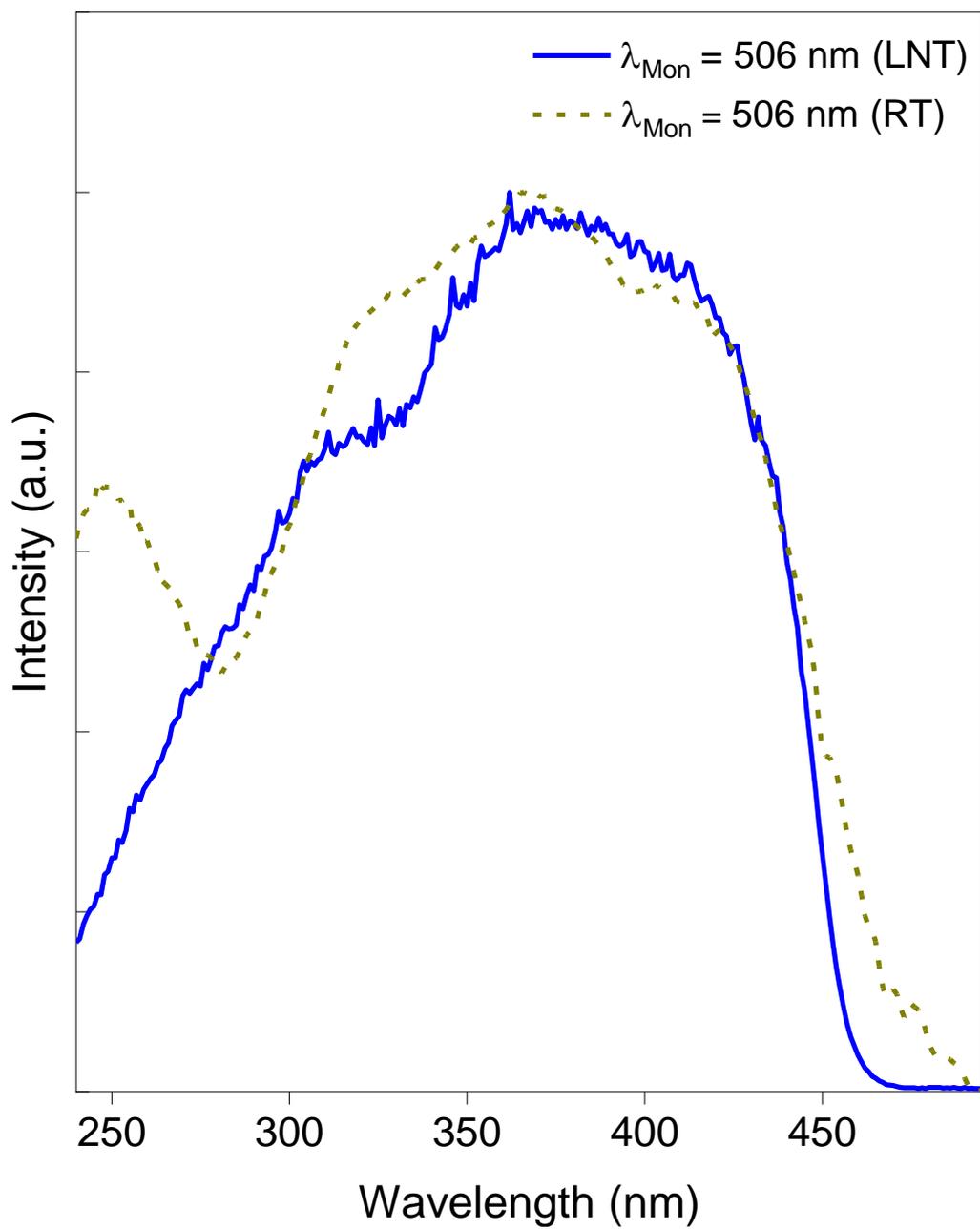
**Figure S7** Excitation of a SAO:Eu,Nd phosphor disc using natural sunlight and a Philips 9.5-W daylight LED bulb (6500 K, 1055 lm). (a) PersL decay curves recorded by monitoring the 1060 nm emission of Nd<sup>3+</sup>. The charging durations were both 5 min. The LED bulb was placed 10 cm above the sample. (b) Normalized emission spectra of natural sunlight and the daylight LED bulb. The spectra were measured using a LISUN LMS-6000S portable CCD spectroradiometer. In measuring the solar spectrum, the detector faced the Sun. In measuring the LED spectrum, the detector was placed towards the bulb at 10 cm distance.



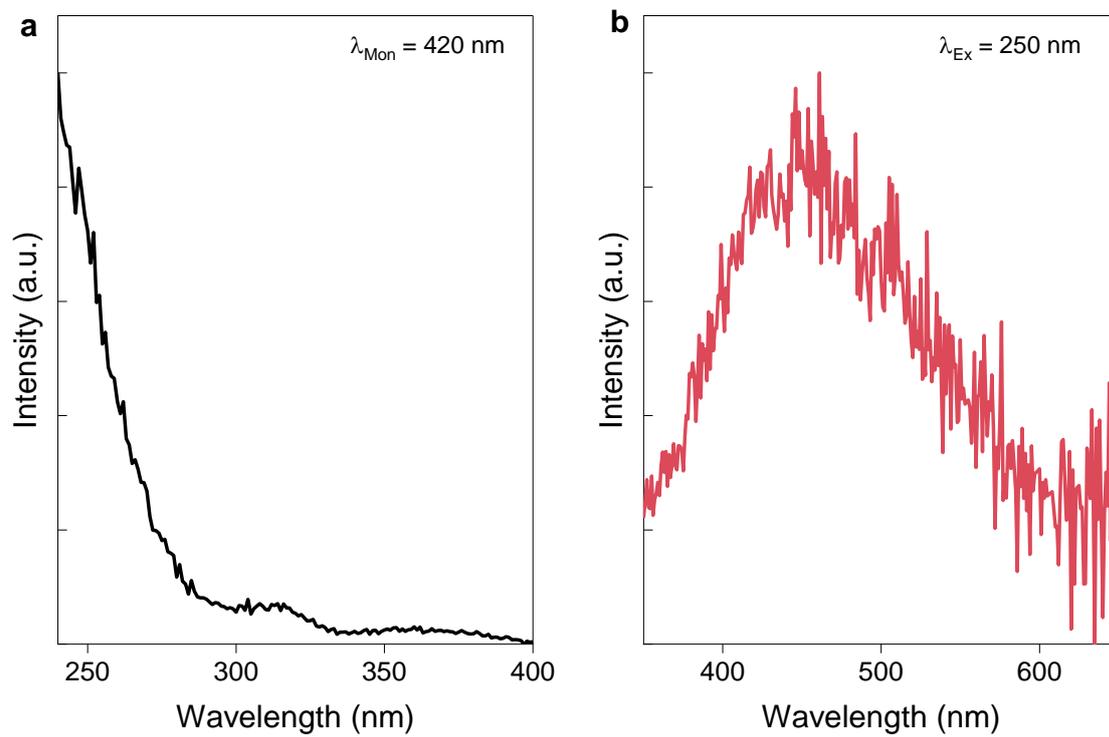
**Figure S8** Charging a SAO:Eu,Nd persistent phosphor disc using a power-tunable 450 nm LED at output intensities of 2–200 mW/cm<sup>2</sup> for 1 s to up to 300 s. Before each charging, the disc was heat-treated at 400 °C for 15 min to empty the traps. After each charging, the PersL decay curve was acquired by monitoring the 506 nm emission of Eu<sup>2+</sup>. (a–c) PersL decay curves acquired after irradiation by a 450 nm LED at light intensities of 2 mW/cm<sup>2</sup>, 19.9 mW/cm<sup>2</sup>, and 99.4 mW/cm<sup>2</sup>, respectively. (d) Relationship of PersL emission intensity ( $I_{120s}$ ), 450 nm light intensity, and irradiation time. The PersL emission intensities are the intensities acquired at 120 s decay points, as indicated by the dash-lines in (a–c).



**Figure S9** TL curves of SAO:Eu,Nd persistent phosphor charged by 440 nm light at RT or LNT. (a,b) TL curves monitored at 506 nm emission of  $\text{Eu}^{2+}$  and 1060 nm emission of  $\text{Nd}^{3+}$  for charging at RT, respectively. The measuring temperature range was 20–300 °C. (c,d) TL curves monitored at 506 nm emission of  $\text{Eu}^{2+}$  and 1060 nm emission of  $\text{Nd}^{3+}$  for charging at LNT, respectively. The measuring temperature range was –196–300 °C. These TL curves have already been shown in Fig. 4 of the main text. They are re-displayed here to highlight the difference in charging effect at RT and LNT.



**Figure S10** Normalized PL excitation spectra of Eu<sup>2+</sup> in SAO:Eu,Nd persistent phosphor at LNT and RT. The monitoring wavelength was 506 nm.



**Figure S11** PL excitation and emission spectra of SrAl<sub>2</sub>O<sub>4</sub> host at RT. The emission spectrum was acquired under 250 nm excitation and the excitation spectrum was obtained by monitoring the 420 nm emission.

**Table S1.** Determination of the green (Eu<sup>2+</sup>) and NIR (Nd<sup>3+</sup>) PersL intensities of SAO:Eu,Nd persistent phosphor at different initial decay instants (10–120 s).

Decay instant (s)	Intensity of green PersL ( $\mu\text{W}/\text{cm}^2$ )	Intensity of NIR PersL ( $\mu\text{W}/\text{cm}^2$ )
10	61.0	0.69
30	15.4	0.23
60	5.9	0.11
120	2.4	0.07

**Measurement note:** The measurements were conducted in a dark chamber setup as depicted in the Supplementary Information in ref. 8. A SAO:Eu,Nd phosphor disc was irradiated by a white LED flashlight for 1 min. The green and NIR PersL emission powers (in  $\mu\text{W}$  or nW) at different initial decay instants (10–120 s) were collected by a Newport 918D-SL-OD3R silicon photodetector and recorded by a Newport 2936-R optical power and energy meter. The measured powers (in  $\mu\text{W}$  or nW) were then calculated into the absolute unit of  $\mu\text{W}/\text{cm}^2$  using a semi-sphere irradiation geometry model by considering the geometry of the dark chamber and the size of the Si detector using equation:  $I = I_m \times 2\pi h^2 / (\pi \times r^2)$ , where  $I_m$  is the measured power,  $h$  is the distance between sensor and disc, and  $r$  is the radius of sensor (see the detail in ref. 8).