

Supporting Information

Considerable improvement of long-persistent luminescence in LiLuSiO₄:Pr³⁺ phosphor by Sm³⁺ co-doping for optical tagging application

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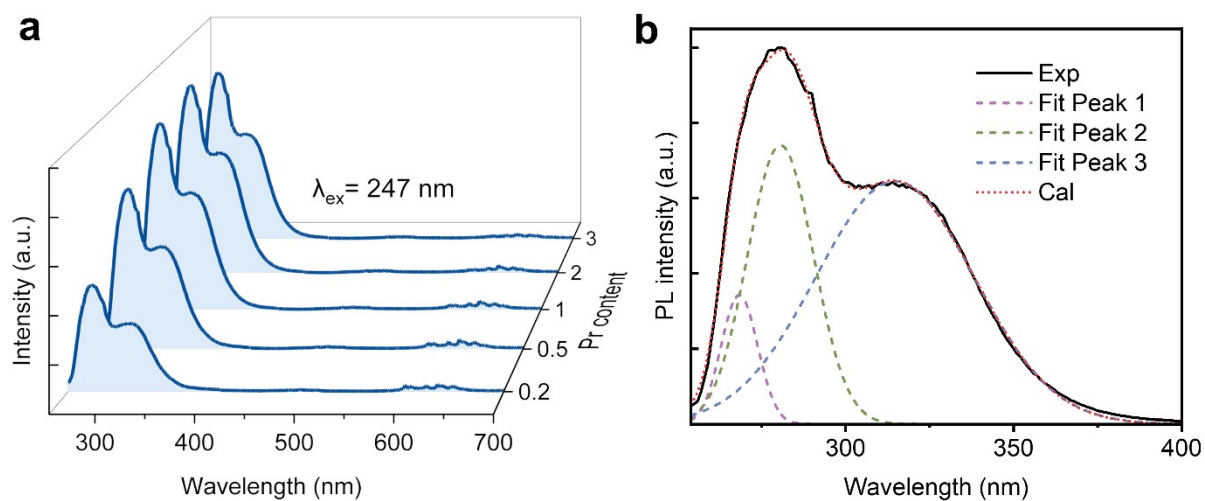


Fig. S1 (a) Photoluminescence emission spectra of $\text{LiLuSiO}_4:x\% \text{Pr}^{3+}$ ($x = 0.2, 0.5, 1, 2, 3$). The spectra were obtained with the excitation of 247 nm. (b) Gaussian decomposition of the emission spectrum of $\text{LiLuSiO}_4:2\% \text{Pr}^{3+}$.

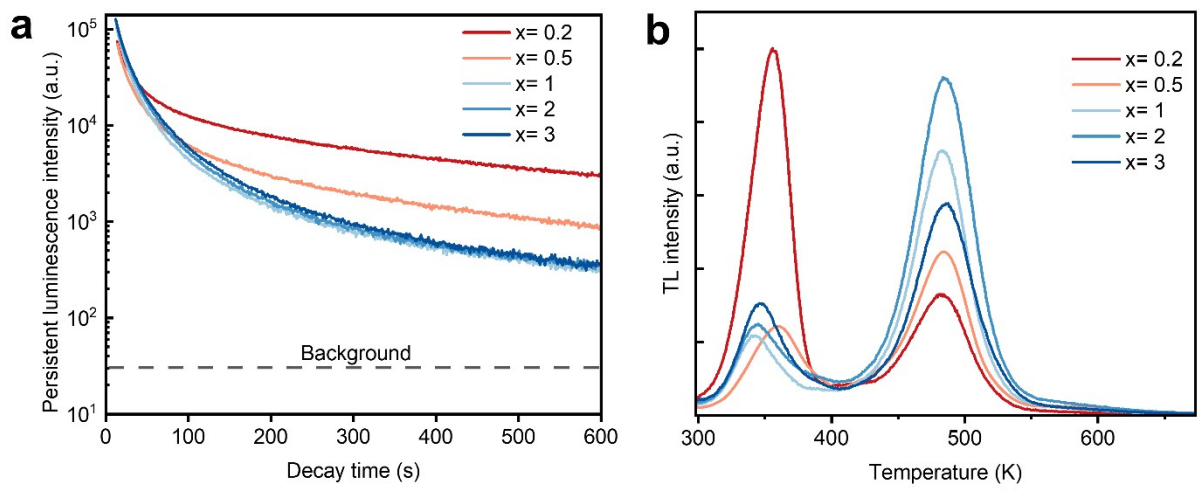


Fig. S2 Persistent luminescence decay curves (a) and thermoluminescence spectra (b) of $\text{LiLuSiO}_4:x\% \text{Pr}^{3+}$ phosphors ($x = 0.2, 0.5, 1, 2, 3$). The samples were pre-irradiated by 254 nm UV lamp for 10 min.

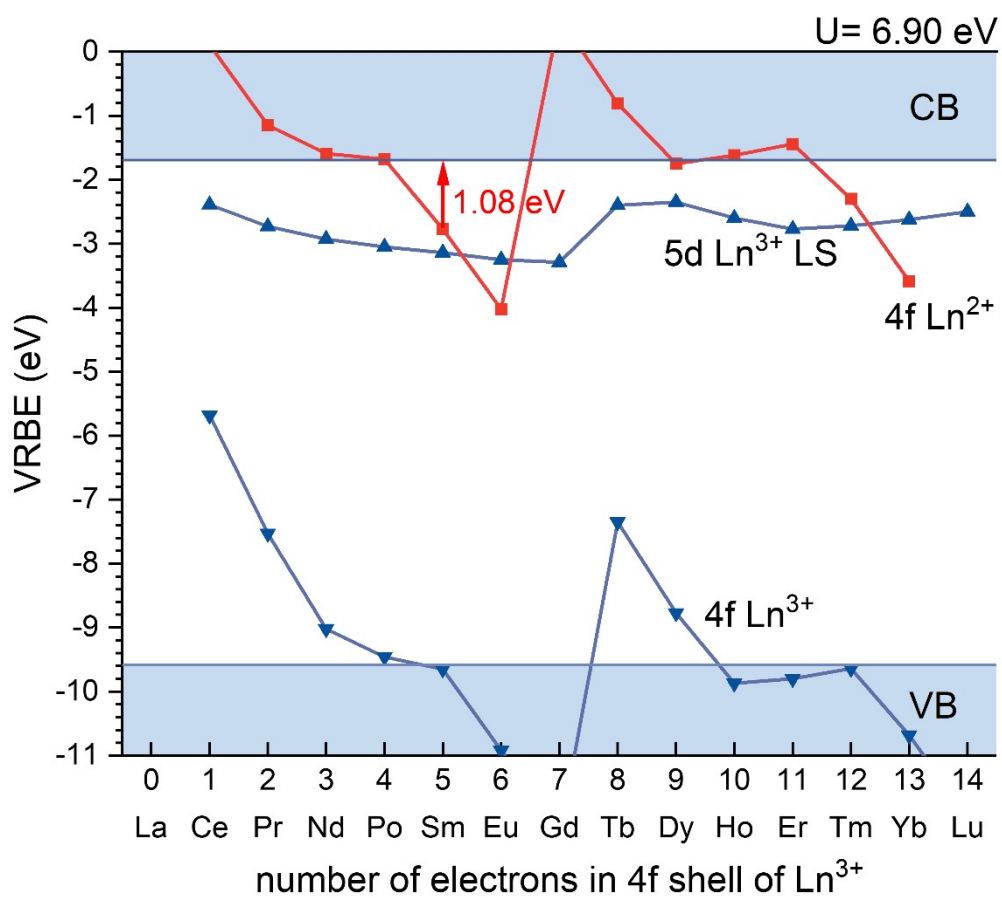


Fig. S3 VRBE diagram of lanthanide ions in LiLuSiO₄.

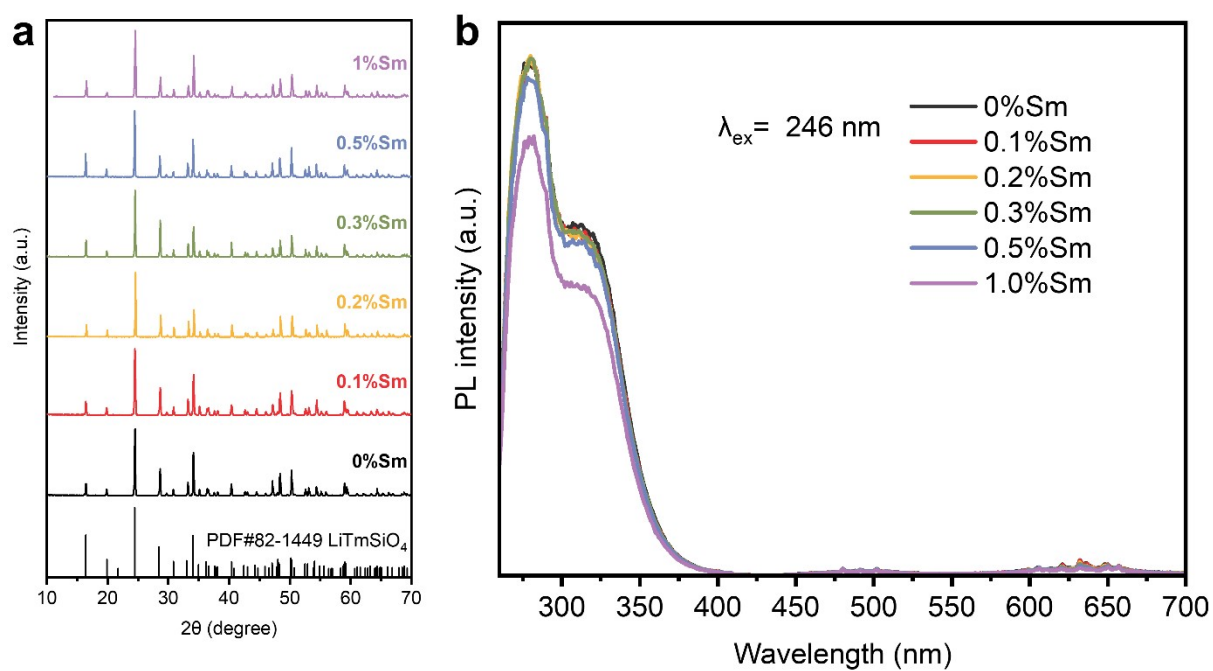


Fig. S4 XRD patterns (a) and photoluminescence emission spectra (b) of LiLuSiO₄:2%Pr³⁺,y%Sm³⁺ (y = 0, 0.1, 0.2, 0.3, 0.5, 1.0) phosphors.

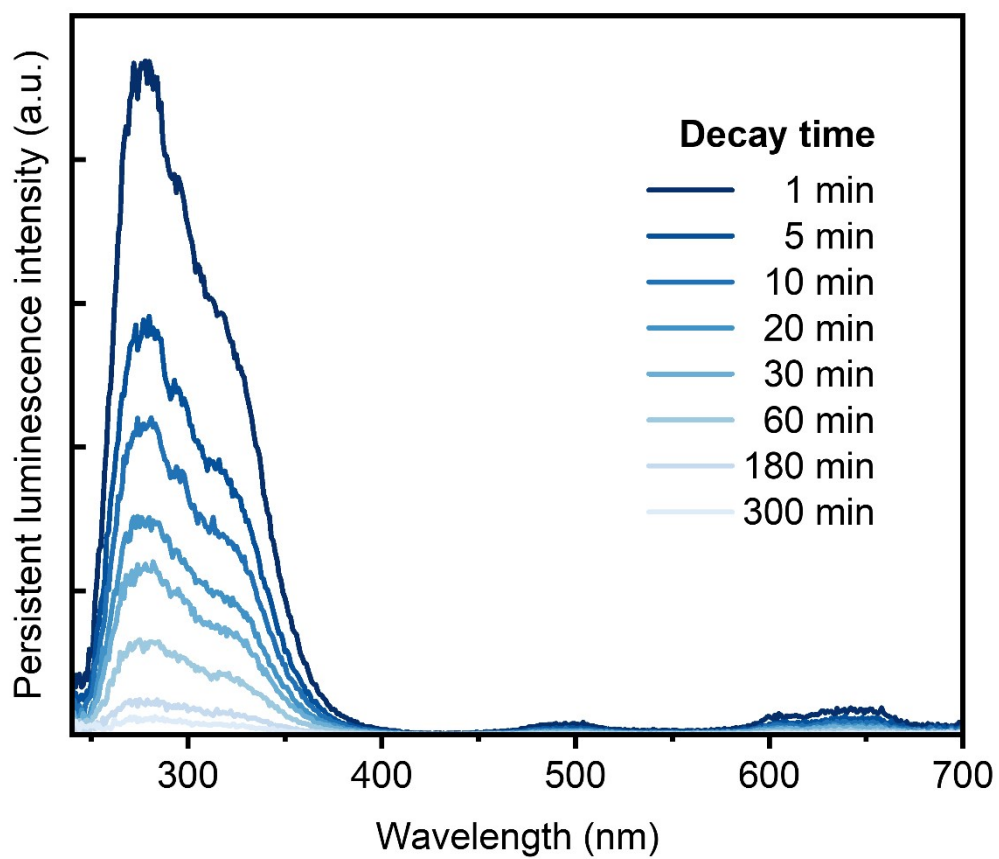


Fig. S5 Persistent luminescence emission spectra of LiLuSiO₄:Pr³⁺, Sm³⁺ phosphor measured at different decay instants after pre-irradiated by a 254 nm UV lamp for 15 min.

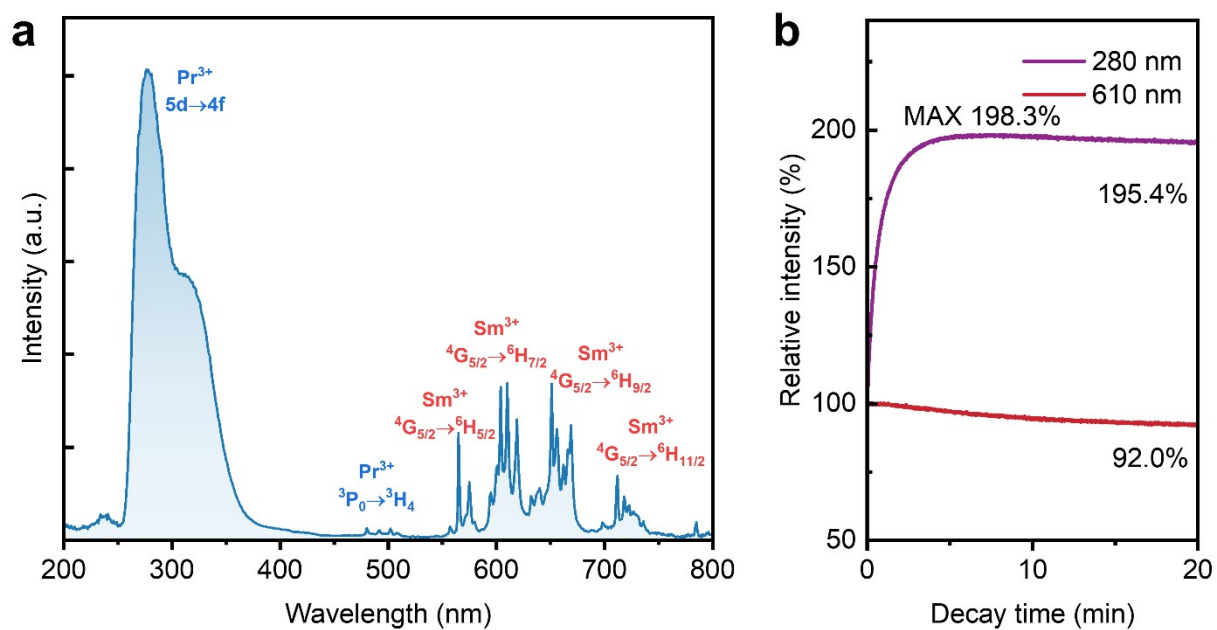


Fig. S6 (a) X-ray excited luminescence spectrum of LLSO:Pr,Sm phosphor. (b) Time-dependent relative emission intensity monitored at 280 nm and 610 nm of LiLuSiO₄:Pr³⁺,Sm³⁺ phosphor

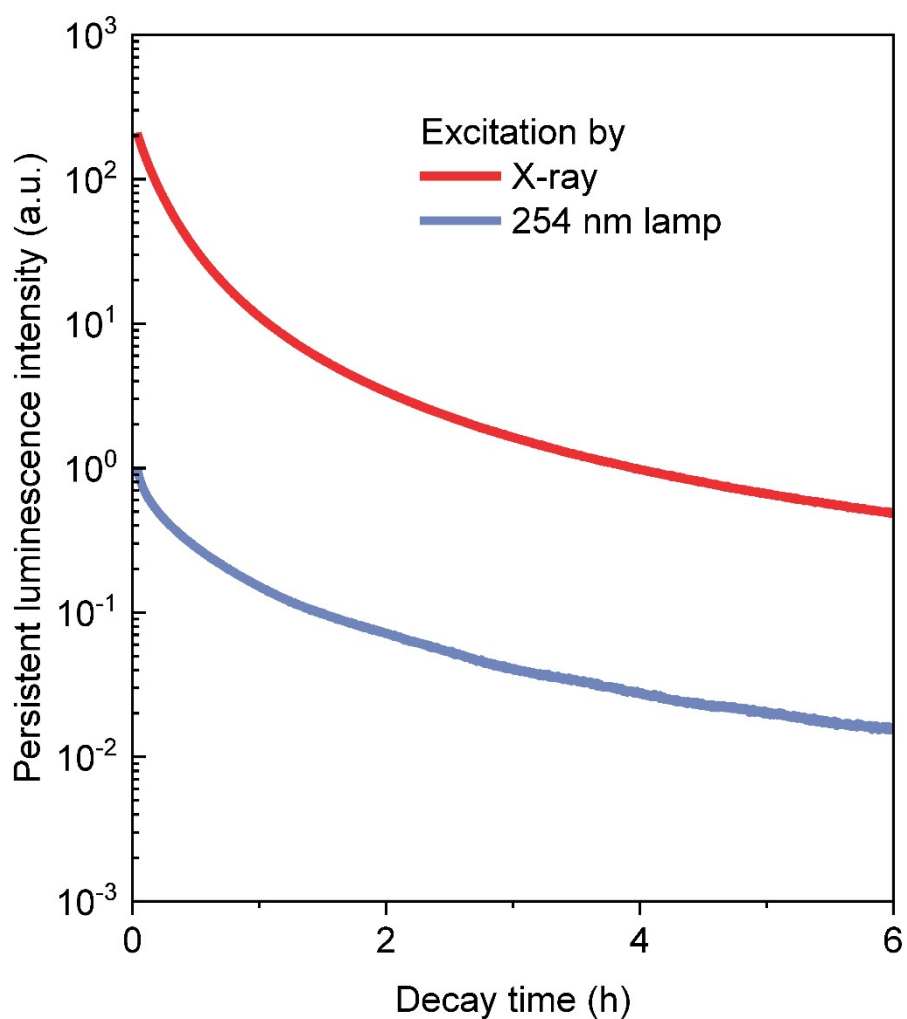


Fig. S7 Persistent luminescence decay curves of the $\text{LiLuSiO}_4:\text{Pr}^{3+},\text{Sm}^{3+}$ phosphor after irradiation by X-ray and 254 nm UV lamp, respectively. The curves were normalized by the initial intensity of the decay curve excited by 254 nm UV lamp.

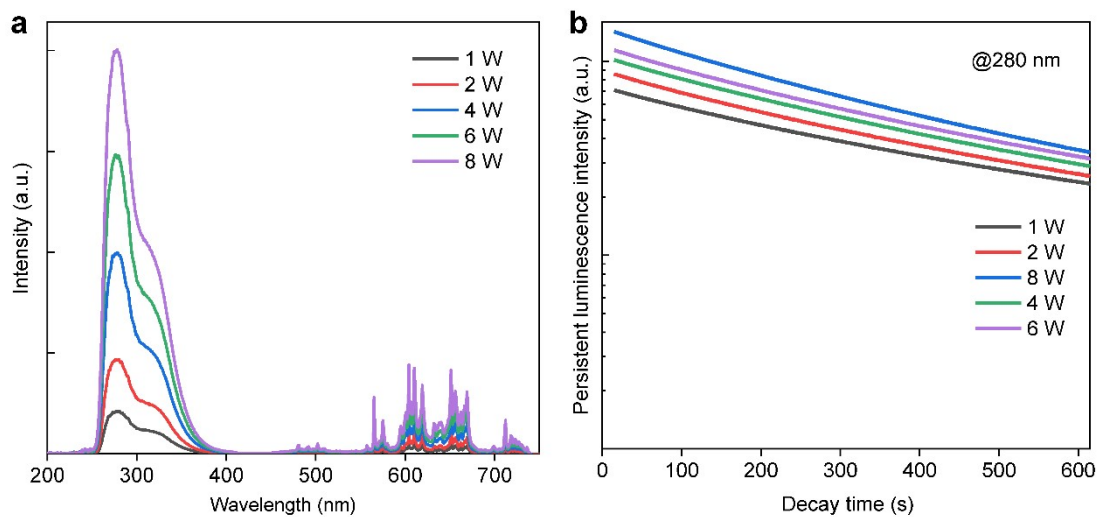


Fig. S8 (a) Radioluminescence spectra of the $\text{LiLuSiO}_4:\text{Pr}^{3+}, \text{Sm}^{3+}$ phosphor upon excitation with a X-ray tube with different output power, respectively. (b) UVC persistent luminescence decay curves after excitation with a X-ray tube with different output power, respectively.

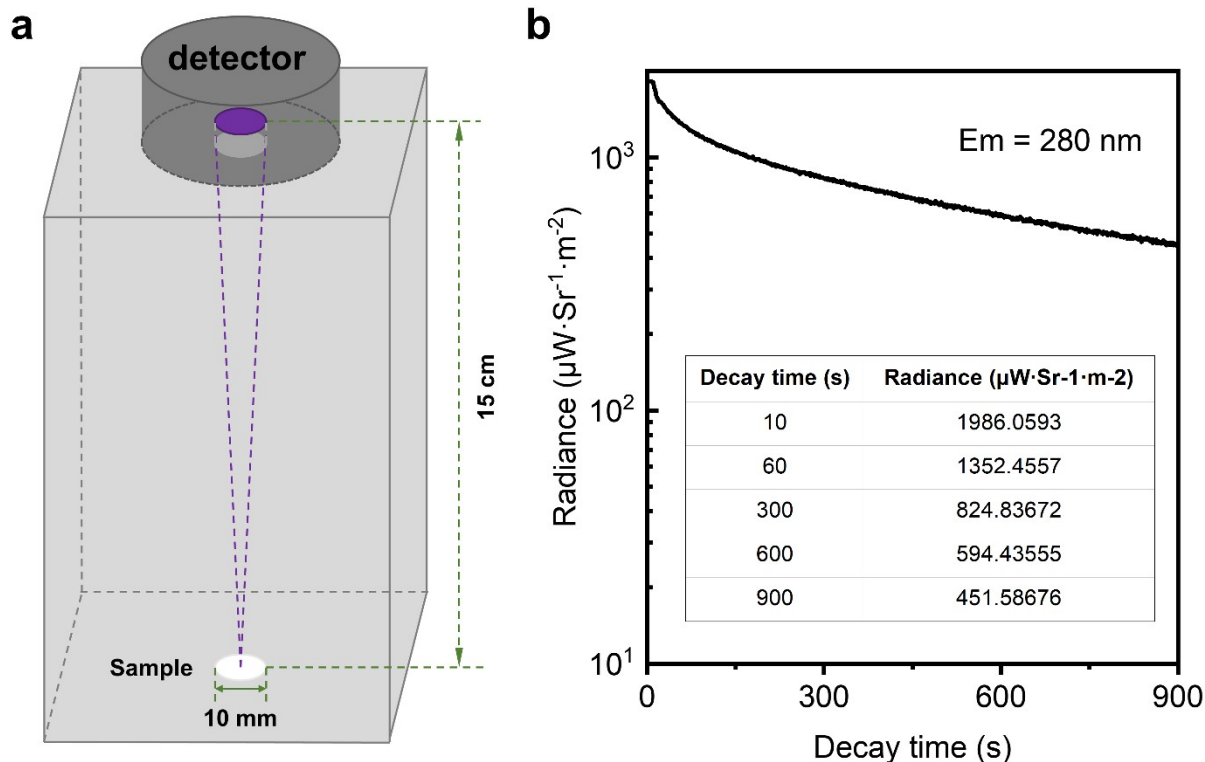


Fig. S9 (a) Measurement device for persistent luminescence radiance intensity. The optical power test is carried out in a self-made black box. The Newport 918D-UV-OD3R UV enhanced silicon photodetector was placed at the top of the black box, and the sample is placed at the bottom. The distance from the detector surface to the sample is controlled to be 15 cm. For a typical procedure, the phosphor disc is placed at the bottom of the black box after X-ray irradiation, and then the measurement is started. The calculated radiance decay curve is shown in Fig. S10b.

The semi-sphere model is used to estimate the afterglow output power of the sample at different decay times, which assuming that the sample disc is a point light source. The persistent luminescence intensity (R) of the phosphor can be calculated using the following formula:

$$R = \frac{P}{2\pi \times (1 - \cos\theta) \times \pi r_s^2}$$

where P is the intensity measured by the power meter (pW), $\theta = \arctan(r_d/h)$, h is the distance between the detector and the sample (15 cm), r_d is the radius of the detector sensor (5 mm), and r_s is the radius of the phosphor disc (5 mm).

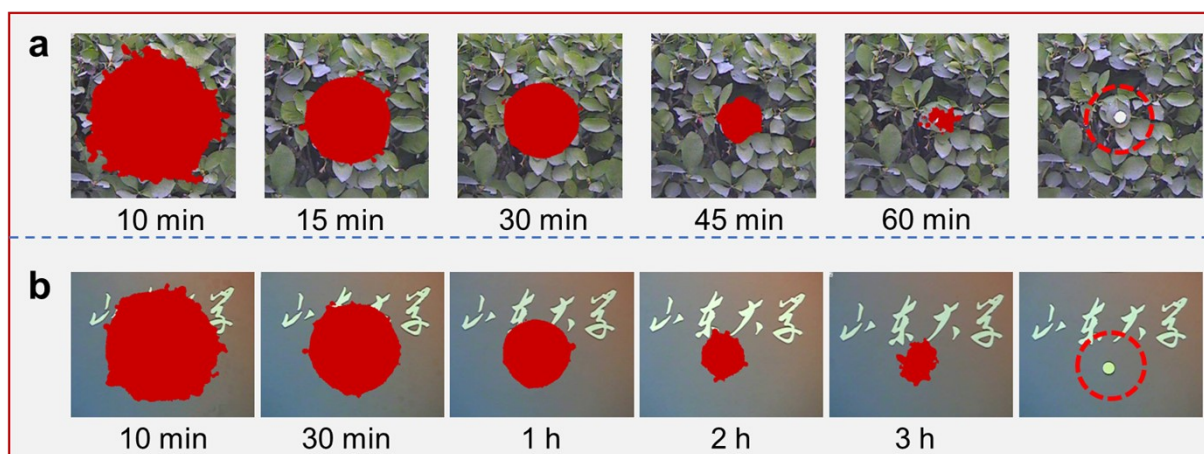


Fig. S10 Solar-blind optical tagging demonstrations for $\text{LiLuSiO}_4:\text{Pr}^{3+},\text{Sm}^{3+}$ phosphors at different environmental conditions. The phosphor discs were placed under outdoor bush (a) and indoor poster (b) to compare the work capability under the photostimulation of sunlight and indoor-lighting. The samples were pre-irradiated by X-ray for 15 min.

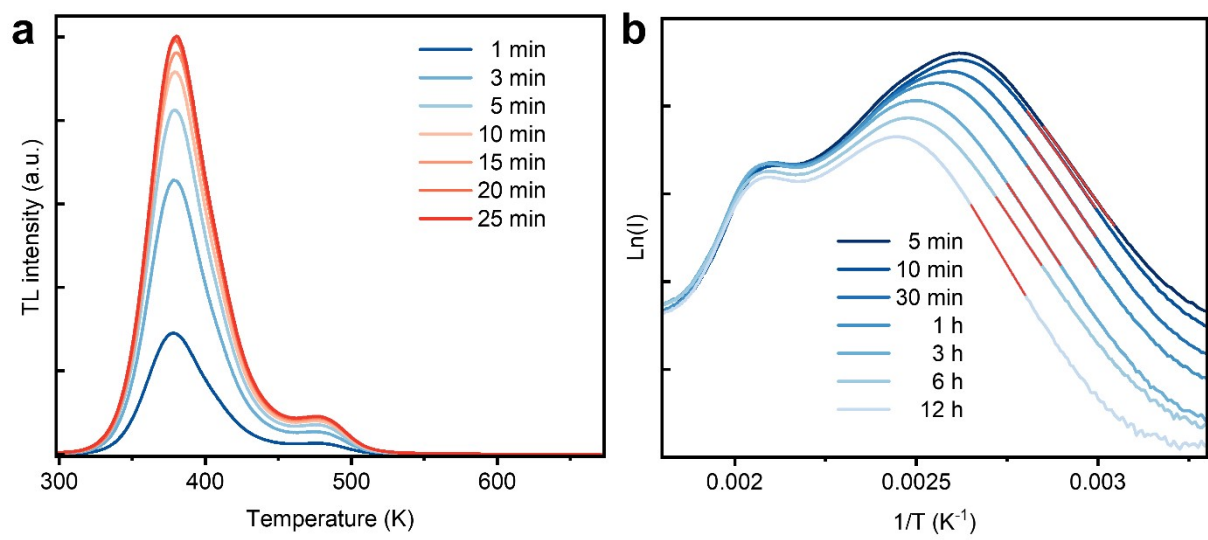


Fig. S11 (a) TL curves of LiLuSiO₄:Pr³⁺, Sm³⁺ with different excitation duration by 254 nm UV lamp. (b) Initial rising analysis for the TL curves of LiLuSiO₄:Pr³⁺, Sm³⁺ at different decay instants after 254 nm UV lamp excitation.

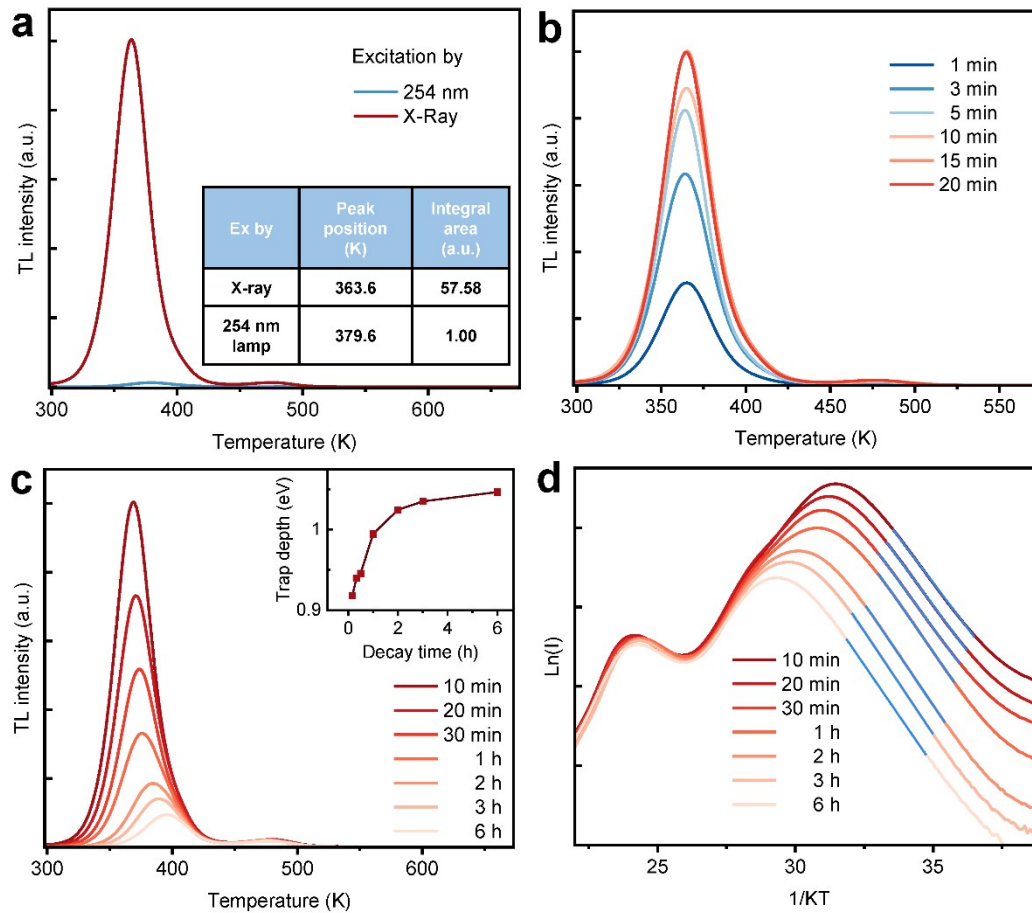


Fig. S12 (a) TL curves of $\text{LiLuSiO}_4:\text{Pr}^{3+},\text{Sm}^{3+}$ phosphor after irradiation by 254 nm UV lamp and X-ray (50 kV, 80 mA), respectively. (b) TL curves of $\text{LiLuSiO}_4:\text{Pr}^{3+},\text{Sm}^{3+}$ with different excitation durations by X-ray (50 kV, 80 mA). (c) TL curves of $\text{LiLuSiO}_4:\text{Pr}^{3+},\text{Sm}^{3+}$ phosphor undergoing different decay time. The sample was pre-irradiated by X-ray for 15 min. (d) Initial rising analysis for the TL curves of $\text{LiLuSiO}_4:\text{Pr}^{3+},\text{Sm}^{3+}$ at different decay instants after X-ray charging.

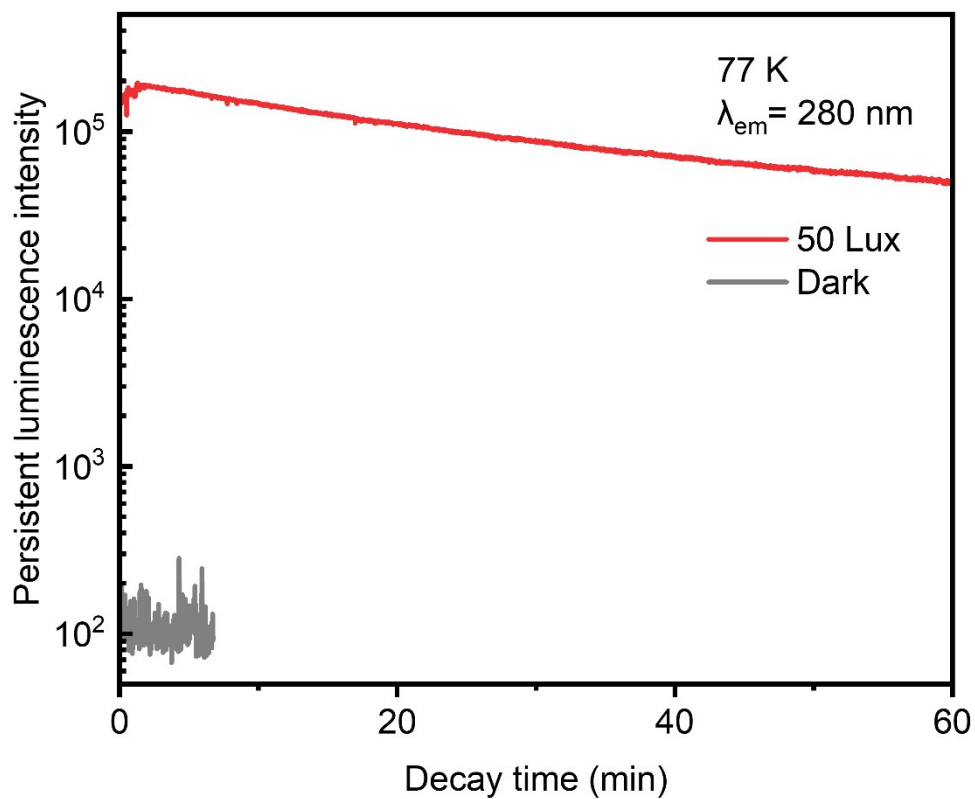


Fig. S13 UVC persistent luminescence decay curves of $\text{LiLuSiO}_4:\text{Pr}^{3+}, \text{Sm}^{3+}$ phosphor at dark and 50 lux white light condition. The sample were pre-irradiated by X-ray for 20 min at room temperature, then measured at 77 K.

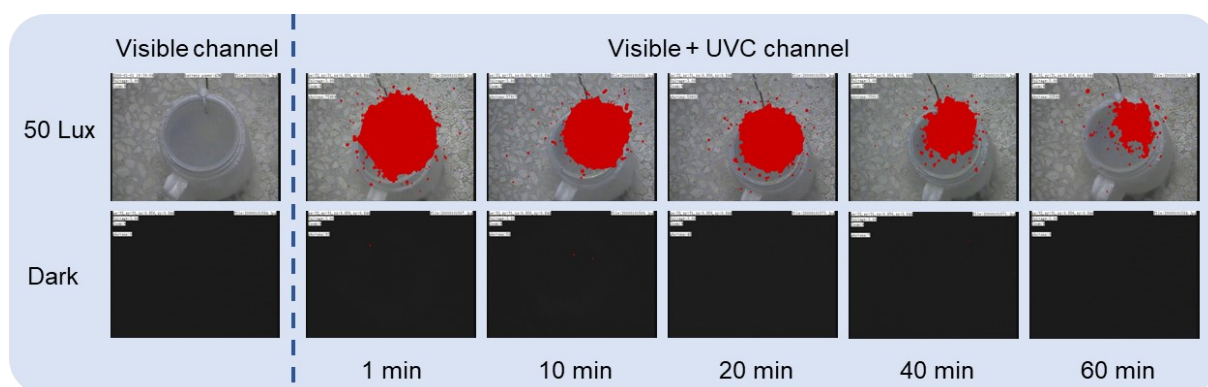


Fig. S14 UVC persistent luminescence images of $\text{LiLuSiO}_4:\text{Pr}^{3+}, \text{Sm}^{3+}$ phosphor at dark and 50 lux white light condition. The sample was pre-irradiated by X-ray for 20 min at room temperature, then immerse into liquid nitrogen.

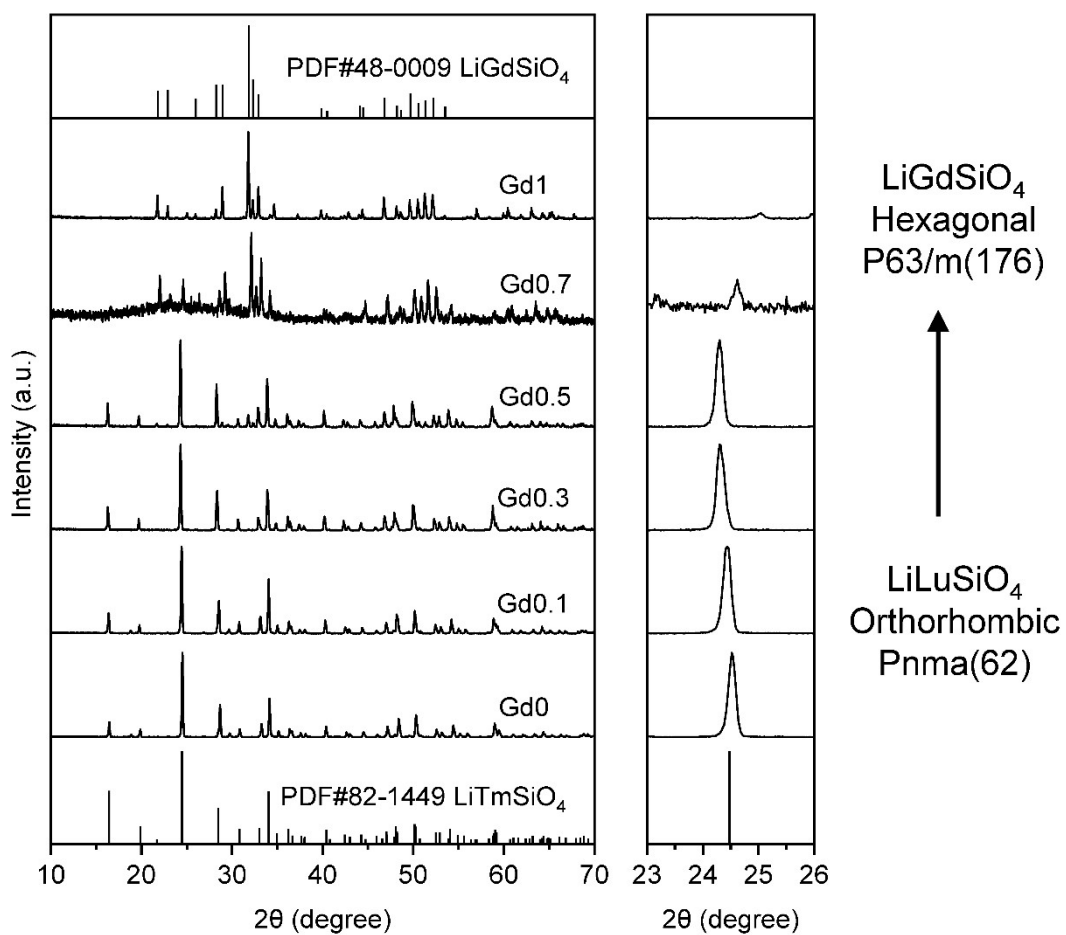


Fig. S15 XRD patterns of $\text{LiLu}_{1-z}\text{Gd}_z\text{SiO}_4:2\%\text{Pr}^{3+},0.1\%\text{Sm}^{3+}$ ($z = 0, 0.1, 0.3, 0.5, 0.7$ and 1) phosphors.

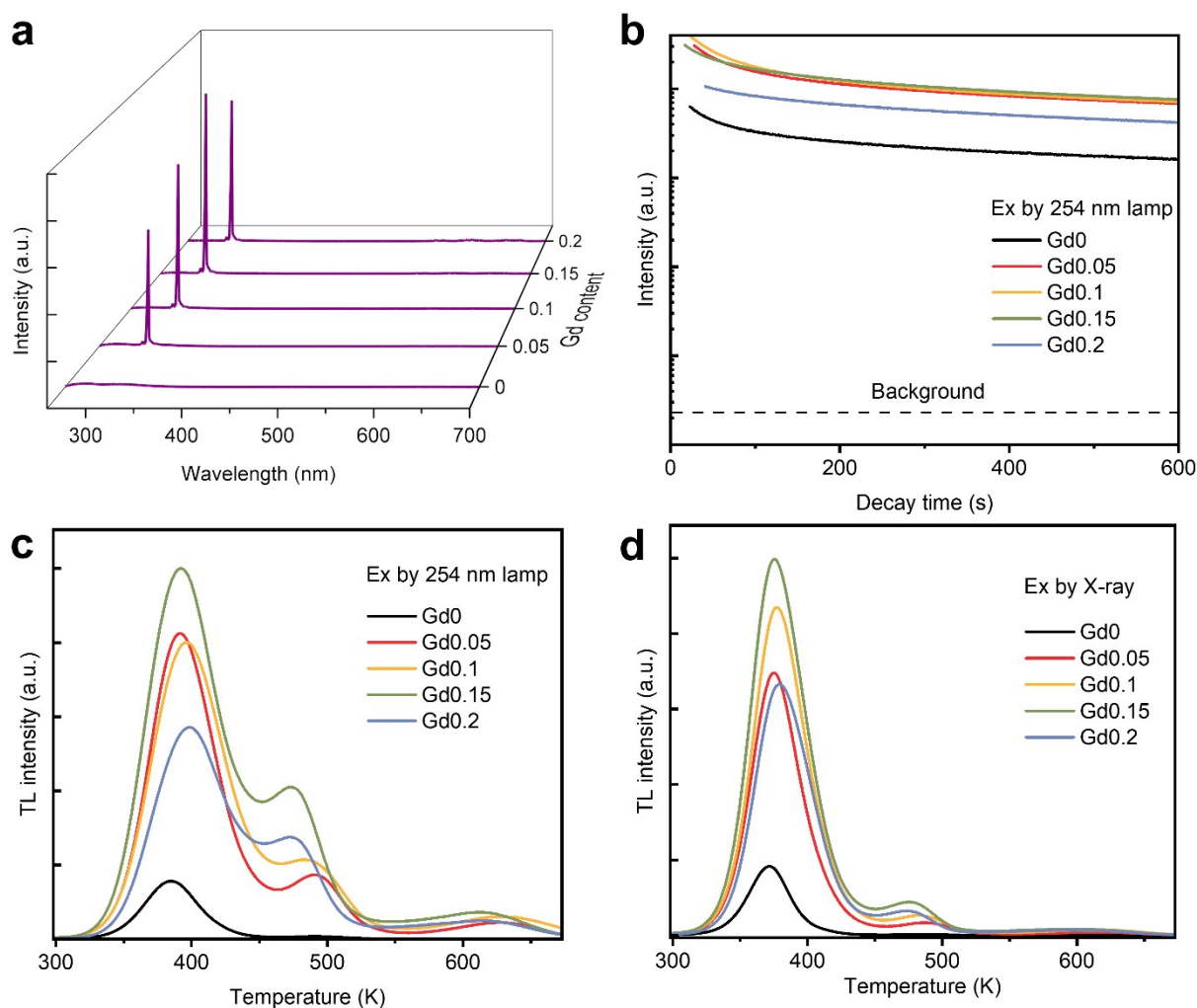


Fig. S16 (a) Photoluminescence emission spectra of $\text{LiLu}_{1-z}\text{Gd}_2\text{SiO}_4:2\%\text{Pr}^{3+},0.1\%\text{Sm}^{3+}$ ($z = 0, 0.05, 0.1, 0.15, 0.2$) upon 248 nm excitation. (b) Persistent luminescence decay curves of $\text{LiLu}_{1-z}\text{Gd}_2\text{SiO}_4:2\%\text{Pr}^{3+},0.1\%\text{Sm}^{3+}$ phosphor after charging by 254 nm UV lamp. (c) TL curves of $\text{LiLu}_{1-x}\text{Gd}_x\text{SiO}_4:2\%\text{Pr}^{3+},0.1\%\text{Sm}^{3+}$ phosphor after UV irradiation for 20 min. (d) TL curves of $\text{LiLu}_{1-z}\text{Gd}_z\text{SiO}_4:2\%\text{Pr}^{3+},0.1\%\text{Sm}^{3+}$ phosphor after X-ray irradiation for 15 min.

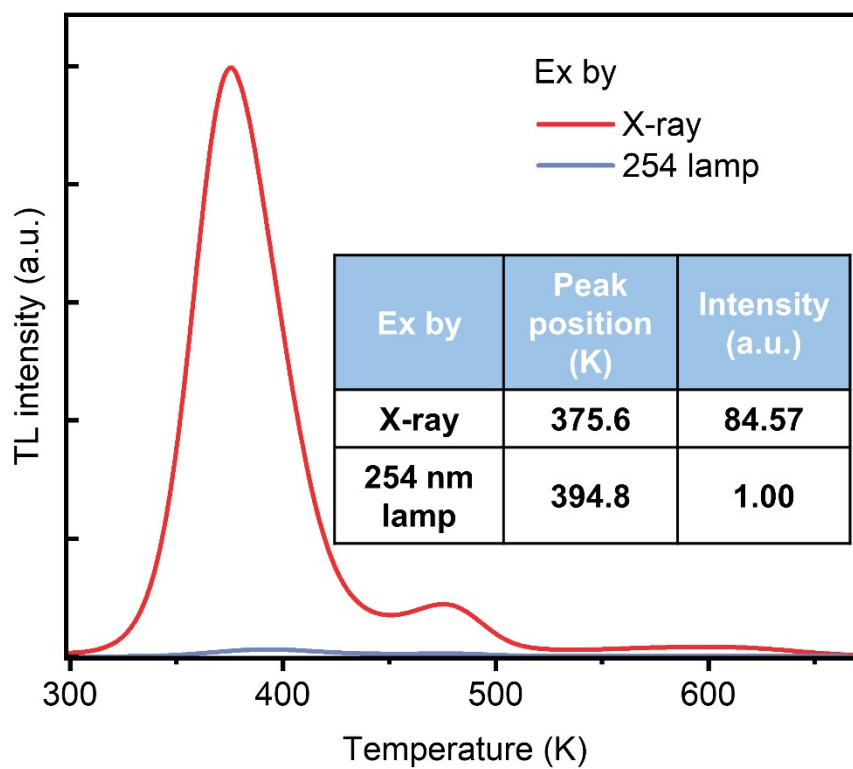


Fig. S17 TL curves of $\text{LiLu}_{0.85}\text{Gd}_{0.15}\text{SiO}_4:\text{Pr}^{3+},\text{Sm}^{3+}$ phosphor after charging with different excitation sources.