

Sunlight stimulated solar-blind ultraviolet phosphor

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As a matter of experience, people always take it for granted that ambient light should be eliminated for the research and application of storage phosphors. Such an empirical viewpoint has hindered the development of storage phosphors in a lighting environment. Herein, we introduce a glow-in-the-daylight phenomenon, which is essentially photostimulated luminescence of ultraviolet phosphor upon stimulation by polychromatic daylight. As a proof of concept, imaging and spectral experiments are carried out using an x-ray-charged $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor to demonstrate the glow performance. The charged phosphor exhibits solar-blind ultraviolet (200–280 nm) emission under sunlight, while its persistent luminescence is nearly absent in a dark environment, revealing the role of sunlight in releasing the stored energy of the phosphor. Moreover, inspired by the case of $\text{LuPO}_4:\text{Pr}^{3+}$, we expect to revisit other ultraviolet phosphors that do not exhibit persistent luminescence in the dark, enabling more glow-in-the-daylight materials to be developed.

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When the glow-in-the-dark phenomenon is mentioned, people think of storage phosphor, which can store external excitation energy and exhibit delayed light emission under a certain stimulation [1,2]. Thermal energy or light illumination may play the role of stimulus to release the stored energy of the storage phosphor, giving rise to persistent luminescence or photostimulated luminescence, respectively [1–4]. To date, although the persistent luminescence and photostimulated luminescence have been well studied, they suffer from some limitations in practical applications. As a matter of experience, people always consider that a dark environment is necessary for detecting persistent luminescence [5–11], and take it for granted that monochromatic light illumination is the prerequisite for photostimulated luminescence [12–18]. It is not clear what impact polychromatic ambient illumination (e.g., sunlight) has on the stimulated luminescence process, hindering further development of storage phosphors in ambient lighting conditions.

Persistent luminescence and photostimulated luminescence of storage phosphors with visible emission wavelength can hardly be detected in a bright environment, since the bright ambient light will submerge the weak emission signals from the phosphors. Therefore, to make the stimulated luminescence detectable in an ambient lighting condition (e.g., in daylight), emission wavelengths of storage phosphors should be outside the spectral range of ambient light [19–21]. For

instance, taking into account the absence of deep ultraviolet background in sunlight on the Earth's surface, phosphors emitting in the solar-blind ultraviolet region (200–280 nm) may be candidates to testify to the effect of outdoor ambient light on the stimulated luminescence.

For ultraviolet luminescence, trivalent praseodymium ion (Pr^{3+}) has received considerable attention due to its efficient $4f5d \rightarrow 4f^2$ allowed transitions in phosphors [22,23]. To date, the available ultraviolet phosphors for Pr^{3+} photoluminescence include fluorides (e.g., LiYF_4), silicates (e.g., Y_2SiO_5), sulfates (e.g., CaSO_4), aluminates (e.g., $\text{LaMgAl}_{11}\text{O}_{19}$), borates (e.g., LuBO_3), and phosphates (e.g., LaPO_4) [24–27]. Besides the conventional photoluminescence, persistent or photostimulated luminescence of Pr^{3+} -doped phosphors in the ultraviolet region is predictable, since Pr^{3+} may be oxidized in some phosphors and thus the electron transfer between the praseodymium ion and energy traps will take place. Recently, persistent phosphors activated by Pr^{3+} with emissions in the solar-blind ultraviolet have been reported [20,28,29], whose luminescence performance has been characterized by using the spectral method in a dark environment. So far, however, there is little scientific evidence on the role of ambient light illumination in the luminescence process of Pr^{3+} -doped storage phosphors. Alternative conceptual approaches are therefore needed to investigate the solar-blind ultraviolet glow luminescence in an ambient lighting environment.

In this work, we report a glow-in-the-daylight phenomenon of storage phosphors, whose ultraviolet emission is outside the spectral range of outdoor sunlight illumination. Taking $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor as an example, solar-blind ultraviolet emission has been achieved when the charged phosphor is exposed to sunlight, while such emission nearly disappears in the dark. Ultraviolet imaging demonstration and spectral

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measurements indicate that the stored energy in the phosphor is gradually released by the ambient illumination energy, which continuously empties the traps by polychromatic ambient light stimulation to yield the long-lasting emission. The effects of illuminance and wavelength of ambient light on the solar-blind ultraviolet glow have been quantitatively evaluated by the spectral approach. The present study suggests that the glow-in-the-daylight phenomenon may be observed in many other ultraviolet phosphors.

To testify the glow-in-the-daylight phenomenon, we prepare $\text{LuPO}_4:0.1\% \text{Pr}^{3+}$ phosphor through a usual solid-state reaction method. Raw materials of Lu_2O_3 , $(\text{NH}_4)_2\text{HPO}_4$ (5% excess) and Pr_6O_{11} powders are mixed and ground. The mixture is sintered 1 h at 800°C . After an additional grinding, the mixture is pressed into a disk, which is sintered again for 2 h at 1300°C to obtain the sample for spectral measurement.

Luminescence performance of the $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor is characterized by imaging and spectral methods. Prior to each spectral measurement, the energy traps are emptied thermally by heating the phosphor at 500°C . Subsequently, the cleaned phosphor is charged by an x-ray tube (Moxtek), whose dose rate is 0.1 Gy per min at the position of the phosphor. Ultraviolet images of the phosphors are taken at 1 min after the x-ray irradiation (for 1000 s) by using an Ofil UVollé-C camera, which is sensitive in the range of 240–280 nm. Each picture is presented by superimposing an ultraviolet image onto a true color image. The ultraviolet emission is highlighted by a red false color overlay in the picture. The outdoor imaging experiment was conducted on June 23, 2021 in natural sunlight. The experimental location is Changchun, China, whose geographic coordinates are $43^\circ51'38''\text{N}$ and $105^\circ19'26''\text{E}$. Spectral performance is characterized using a spectrofluorometer (PTI QuantaMaster 8075-11). The glow emission spectrum of the phosphors is measured at 2 min after the end of the x-ray irradiation. Ultraviolet emission intensities of the phosphors are evaluated using an optical power meter (Newport 2936-R) equipped with a silicon photodetector (Newport 918D). Thermoluminescence curves are recorded by using a TL Reader (Rongfan, SL08-L). The heating rate is 4°C s^{-1} for the thermoluminescence measurements. The photostimulation spectrum of the phosphor is measured by tuning the stimulating wavelengths from 350 to 850 nm (from a filtered xenon lamp; power densities are in the range of $0.25 - 0.7 \text{ mW cm}^{-2}$) and recording the maximum intensity of each ultraviolet emission decay curve. Diffuse reflectance spectra are measured on a Shimadzu UV-3600 spectrophotometer. Filtered sunlight from a solar simulator (Sun 2000, Abet Technology) served as the primary ambient lighting source in the spectral measurements. A visible bandpass filter, which passes wavelengths between 400 and 700 nm [290 nm full width at half maximum (FWHM)], was employed to block the ultraviolet and infrared components of the sunlight. The ultraviolet emission of the phosphor is collected after transmission through a filter combination, which includes an XUS0385-SP filter and an XUV0400-SP filter (Asahi Spectra USA Inc.). Such a filter combination is employed to pass ultraviolet light in the range of 245–385 nm. An illuminance meter (T-10A, Konica Minolta) is applied to record the sunlight illuminance, which is adjusted by using a series of neutral density filters. All spec-



FIG. 1. Imaging demonstration in night and sunlight. A glow-in-the-daylight phosphor disk $\text{LuPO}_4:\text{Pr}^{3+}$ and a commercial glow-in-the-dark phosphor disk $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}, \text{Dy}^{3+}$ are attached to a tree trunk, as shown in the middle picture. The left picture is taken on a dark night, while the right picture is taken in the daytime. The charged $\text{LuPO}_4:\text{Pr}^{3+}$ nearly does not emit in the dark environment (the location of the phosphor is marked by a dashed circle in the left picture) but exhibits solar-blind ultraviolet emission in the outdoor sunlight environment (the ultraviolet signal is highlighted by a red false color overlay in the right picture).

tra presented have been corrected for the system responses (including the xenon lamp, monochromator, and detector) as well as the transmissions of the filters.

To visually demonstrate the glow-in-the-daylight feature of $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor, we perform an imaging experiment by comparing the present phosphor with a commercial glow-in-the-dark phosphor ($\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}, \text{Dy}^{3+}$) at night and in the daytime. The middle picture of Fig. 1 shows that the two phosphor disks taken by a solar-blind ultraviolet camera in outdoor environments, in which the two disks have been thermally emptied and are attached to a tree trunk. At night, as shown in the left picture, the x-ray charged $\text{LuPO}_4:\text{Pr}^{3+}$ disk nearly does not emit, while the $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}, \text{Dy}^{3+}$ material emits bright green persistent luminescence. That is, unlike the glow-in-the-dark phosphor, there is no detectable persistent luminescence from the $\text{LuPO}_4:\text{Pr}^{3+}$ in the dark. In contrast, in the daytime under natural sunlight (illuminance at the location of the disks is around 15 klx), as shown in the right picture, the charged $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor gives solar-blind ultraviolet emission, which is indicated by a red pattern. In this picture, the emission from the charged $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}, \text{Dy}^{3+}$ has been submerged by sunlight and cannot be observed. Compared with the well-studied glow-in-the-dark phosphors [1], there is little scientific investigation on the glow-in-the-daylight phenomenon, so not much is known about such phosphor.

We next demonstrate the glow-in-the-daylight performance of the $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor using spectral methods. Under x-ray excitation, the phosphor exhibits ultraviolet emissions peaking at 233, 245, 262, and 272 nm, which have been assigned to $\text{Pr}^{3+} 4f5d \rightarrow 4f^2$ ($^3\text{H}_4$, $^3\text{H}_5$, $^3\text{H}_6 + ^3\text{F}_2$, and $^3\text{F}_3 + ^3\text{F}_4$) transitions (see Fig. S1 in the Supplemental Material [30]). When the x-ray excitation ceases, the charged phosphor gives solar-blind ultraviolet emission in a sunlight illumination environment. The corresponding glow emission spectrum is recorded upon illumination with the sunlight (100 klx). As shown in Fig. 2(a), the spectral shape of the glow emission is identical to that of steady-state emission, confirming that such glow emission originates from Pr^{3+} (the two spectra have been normalized at 262 nm). Note that the glow-in-the-daylight spectrum is only recorded over

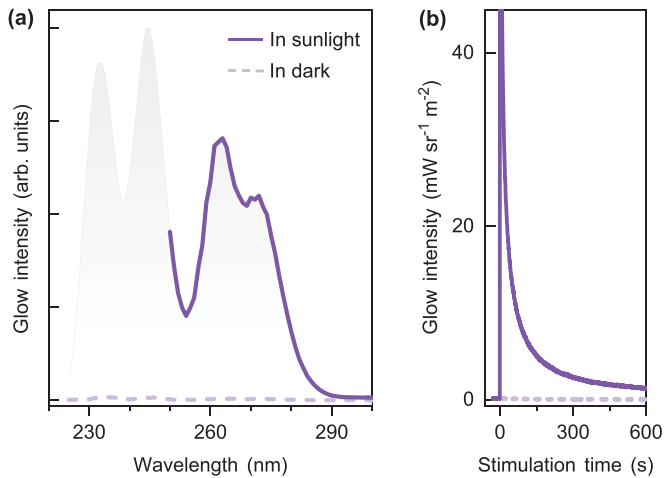


FIG. 2. (a) Glow spectra of $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor recorded at room temperature under sunlight illumination (solid line) and in a dark environment (dashed line). A steady-state luminescence emission spectrum excited by the x-ray is also presented as the curve in shadow. (b) Time evolutions of emission intensities by monitoring at 262 nm upon sunlight illumination (100 klx) and in a dark environment. The time interval between the end of x-ray irradiation and the beginning of sunlight illumination is 1 min.

250–290 nm due to the block of the employed filters. For comparison, we also record the emission spectrum of the phosphor in a dark environment after the x-ray irradiation, as shown by the dashed line in Fig. 2(a). Unlike the emission performance in sunlight, the $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor exhibits an extremely weak afterglow emission in the dark. This spectral result can be expected, which is consistent with the imaging demonstration presented in Fig. 1. We further measure decay curves of the x-ray charged $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor under the sunlight illumination (100 klx), as shown in Fig. 2(b). For the glow-in-the-daylight decay curve (solid line), the emission intensity reaches maximum at the initial moment of the sunlight stimulation, and then decays with time. In contrast to the glow-in-the-daylight emission, the glow emission intensity of the phosphor is nearly undetectable in ambient dark (dashed line), implying that the ambient temperature cannot contribute effectively to the releasing of stored energy in the phosphor. That is, the excitation energy can be stored in the phosphor for a long time in a dark environment (see Fig. S2 in the Supplemental Material [30]).

Subsequently, to gain insight into the glow-in-the-daylight phenomenon of $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor, we carry out thermoluminescence measurements, which can provide information on the energy traps involved in the glow emission. The dashed line in Fig. 3(a) represents the thermoluminescence curve recorded on a phosphor that has been kept in the dark for 10 min after the x-ray irradiation. The thermoluminescence curve consists of three distinct peaks at 115 °C, 215 °C, and 370 °C, respectively. These thermoluminescence peaks correspond to electrons trapped in various oxygen vacancies, involving free oxygen vacancies and thermally more stable vacancies in $[\text{PO}_4]$ units [29]. Note that there is no significant change in intensity of the thermoluminescence in the 10 min

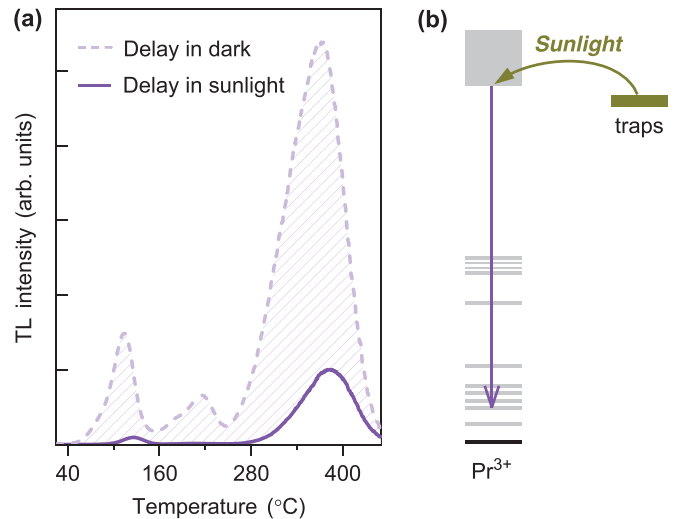


FIG. 3. (a) Thermoluminescence (TL) curves for 262 nm emission of $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor recorded after ceasing the x-ray radiation. Before the measurements, the charged phosphor is kept in the dark for 10 min (dashed line) or under sunlight illumination for 10 min (solid line). (b) Schematic illustration of the stimulation in the glow-in-the-daylight emission process.

(see Fig. S3 in the Supplemental Material [30]). To evaluate the contribution of ambient illumination on the distribution of traps, we also record thermoluminescence curves on the charged phosphor that has been kept being exposed to sunlight (100 klx) for 10 min, as shown by the solid-line curve in Fig. 3(a). In comparison with the dashed-line curve, the solid-line curve shows a remarkable decrease in all the thermoluminescence peaks, indicating that all the traps have been emptied by the sunlight illumination. The gradual depletion of traps upon sunlight illumination is further confirmed by an extended thermoluminescence experiment, in which the charged phosphor experiences different illumination durations (Fig. S4 in the Supplemental Material [30]). Based on the above results, one can deduce that the glow-in-the-daylight emission presented in Figs. 1 and 2 originates from the liberation of trapped electrons caused by the sunlight stimulation (note: the phosphor cannot be charged by the present sunlight). Such a glow emission is essentially photostimulated luminescence upon stimulation by polychromatic sunlight. That is, during x-ray irradiation, the excitation energy is stored in the phosphor as trapped electrons. After ceasing the irradiation, the polychromatic sunlight illumination energy may continuously liberate the trapped electrons, giving rise to the glow emission from Pr^{3+} , as schematically illustrated in Fig. 3(b).

Since sunlight can stimulate the $\text{LuPO}_4:\text{Pr}^{3+}$ phosphor, the effects of sunlight illuminance and stimulating wavelength on the glow emission are important to understanding the glow-in-the-daylight phenomenon. Accordingly, we measure the emission decay curves of the phosphor upon sunlight illumination with different illuminance (Fig. S5 in the Supplemental Material [30]). Our measurement shows that the maximum intensity of each decay curve is proportional to the sunlight illuminance [Fig. 4(a)]. Such a result is quite

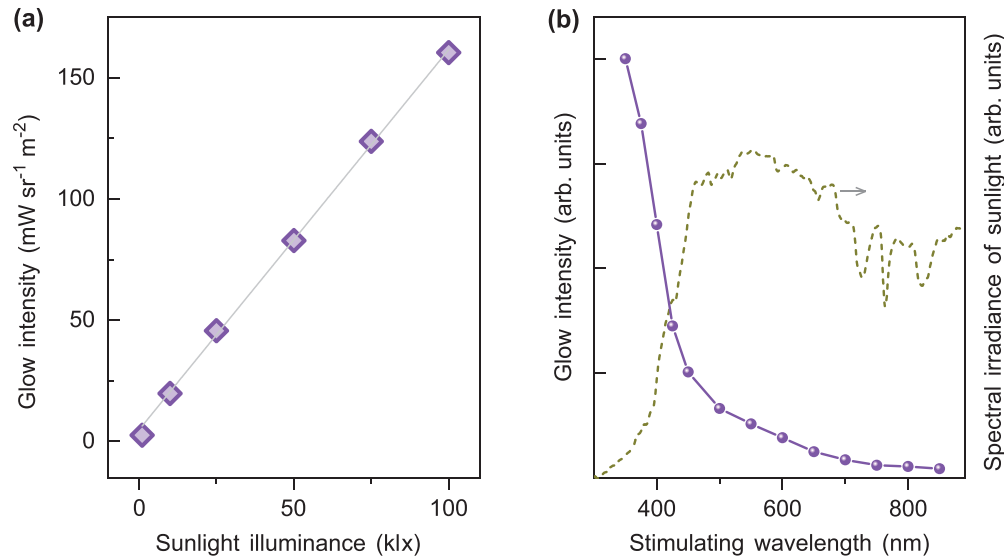


FIG. 4. (a) Glow-in-the-daylight emission intensities of $\text{LuPO}_4\text{:Pr}^{3+}$ phosphor versus the sunlight illuminance. The straight line is a linear fit of the plot. (b) Photostimulation spectrum derived by plotting the maximum intensities of photostimulated luminescence decay curves as a function of the stimulating wavelengths. The solar spectrum recorded at the experimental location is also presented as the dashed-line curve.

understandable since the present glow-in-the-daylight phenomenon is essentially a photostimulated luminescence, whose emission intensity is proportional to the incident photon flux [2]. Subsequently, to learn the effectivity of different stimulating wavelengths in the polychromatic sunlight to the stimulation of the phosphor, we further record the stimulated decay curves upon monochromatic light illumination with different wavelengths (Fig. S6 in the Supplemental Material [30]). We therefore obtain a photostimulation spectrum by recording the maximum intensities versus the stimulating wavelengths. Figure 4(b) shows that the charged $\text{LuPO}_4\text{:Pr}^{3+}$ can be stimulated effectively by wavelengths over 350–600 nm. Such a wavelength range coincides with the absorption spectrum of the photochromic center in the phosphor (Fig. S7 in the Supplemental Material [30]), further confirming the contribution of electron traps to the photostimulation process [17]. Moreover, the photostimulation spectrum in Fig. 4(b) also indicates that the stimulating illumination with a shorter wavelength in the visible region will yield a more intense emission. For example, comparing typical indoor lighting, in which the ultraviolet component is usually absent, sunlight illumination with a larger useful fraction is preferred

to stimulate the phosphor (Fig. S8 in the Supplemental Material [30]).

In summary, we have introduced the glow-in-the-daylight phenomenon by using $\text{LuPO}_4\text{:Pr}^{3+}$ phosphor as an example. The charged phosphor continuously gives a solar-blind ultraviolet emission in sunlight, while such emission is nearly absent in a dark environment. Since not much is known about the glow-in-the-daylight phenomenon from the point of view of spectroscopic investigations, there remain plenty of opportunities to discover and develop it. For instance, we have observed the glow-in-the-daylight phenomenon in various solar-blind ultraviolet phosphors (Fig. S9 in the Supplemental Material [30]). The approach presented in this work appears to be generally applicable to these phosphors. Subsequently, we expect that spectroscopic investigations, involving the thermoluminescence and photostimulation measurements, may reveal more intrinsic properties of the glow-in-the-daylight phosphors.

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- [1] J. Xu and S. Tanabe, Persistent luminescence instead of phosphorescence: History, mechanism, and perspective, *J. Lumin.* **205**, 581 (2019).
- [2] S. Schweizer, Physics and current understanding of x-ray storage phosphors, *Phys. Status Solidi A* **187**, 335 (2001).
- [3] Z. W. Pan, Y. Y. Lu, and F. Liu, Sunlight-activated long-persistent luminescence in the near-infrared from Cr^{3+} -doped zinc gallogermanates, *Nat. Mater.* **11**, 58 (2012).
- [4] J. J. Joos, K. Korthout, L. Amidani, P. Glatzel, D. Poelman, and P. F. Smet, Identification of $\text{Dy}^{3+}/\text{Dy}^{2+}$ as Electron Trap in Persistent Phosphors, *Phys. Rev. Lett.* **125**, 033001 (2020).
- [5] T. Matsuzawa, Y. Aoki, N. Takeuchi, and Y. Murayama, A new long phosphorescent phosphor with high brightness, $\text{SrAl}_2\text{O}_4\text{:Eu}^{2+}$, Dy^{3+} , *J. Electrochem. Soc.* **143**, 2670 (1996).
- [6] F. Liu, Y. J. Liang, and Z. W. Pan, Detection of Up-Converted Persistent Luminescence in the Near Infrared Emitted by the $\text{Zn}_3\text{Ga}_2\text{GeO}_8\text{:Cr}^{3+}$, Yb^{3+} , Er^{3+} Phosphor, *Phys. Rev. Lett.* **113**, 177401 (2014).
- [7] T. Maldiney, A. Bessière, J. Seguin, E. Teston, S. K. Sharma, B. Viana, A. J. J. Bos, P. Dorenbos, M. Bessodes, D. Gourier, D. Scherman, and C. Richard, The *in vivo* activation of persistent

- nanophosphors for optical imaging of vascularization, tumours and grafted cells, *Nat. Mater.* **13**, 418 (2014).
- [8] Y. J. Liang, F. Liu, Y. F. Chen, X. J. Wang, K. N. Sun, and Z. W. Pan, New function of the Yb^{3+} ion as an efficient emitter of persistent luminescence in the short-wave infrared, *Light Sci. Appl.* **5**, e16124 (2016).
- [9] X. Y. Ou, X. Qin, B. L. Huang, J. Zan, Q. X. Wu, Z. Z. Hong, L. L. Xie, H. Y. Bian, Z. G. Yi, X. F. Chen, Y. M. Wu, X. R. Song, J. Li, Q. S. Chen, H. H. Yang, and X. G. Liu, High-resolution x-ray luminescence extension imaging, *Nature (London)* **590**, 410 (2021).
- [10] P. Pei, Y. Chen, C. X. Sun, Y. Fan, Y. M. Yang, X. Liu, L. F. Lu, M. Y. Zhao, H. X. Zhang, D. Y. Zhao, X. G. Liu, and F. Zhang, X-ray-activated persistent luminescence nanomaterials for NIR-II imaging, *Nat. Nanotechnol.* **16**, 1011 (2021).
- [11] Y. X. Zhuang, D. R. Chen, W. J. Chen, W. X. Zhang, X. Su, R. R. Deng, Z. F. An, H. M. Chen, and R. J. Xie, X-ray-charged bright persistent luminescence in $\text{NaYF}_4\text{:Ln}^{3+}@\text{NaYF}_4$ nanoparticles for multidimensional optical information storage, *Light Sci. Appl.* **10**, 132 (2021).
- [12] E. G. Yukihara and S. W. S. McKeever, Optically stimulated luminescence (OSL) dosimetry in medicine, *Phys. Med. Biol.* **53**, R351 (2008).
- [13] P. Leblans, D. Vandenbroucke, and P. Willems, Storage phosphors for medical imaging, *Materials* **4**, 1034 (2011).
- [14] A. Meijerink and G. Blasse, Photostimulated luminescence and thermally stimulated luminescence of some new x-ray storage phosphors, *J. Phys. D: Appl. Phys.* **24**, 626 (1991).
- [15] J. R. Qiu, Y. Shimizugawa, Y. Iwabuchi, and K. Hirao, Photostimulated luminescence in Eu^{2+} -doped fluoroaluminate glasses, *Appl. Phys. Lett.* **71**, 759 (1997).
- [16] W. Chen, R. Sammynaiken, and Y. Huang, Photoluminescence and photostimulated luminescence of Tb^{3+} and Eu^{3+} in zeolite-Y, *J. Appl. Phys.* **88**, 1424 (2000).
- [17] F. Liu, W. Z. Yan, Y. J. Chuang, Z. P. Zhen, J. Xie, and Z. W. Pan, Photostimulated near-infrared persistent luminescence as a new optical read-out from Cr^{3+} -doped LiGa_5O_8 , *Sci. Rep.* **3**, 1554 (2013).
- [18] S. S. Lin, H. Lin, C. G. Ma, Y. Cheng, S. Z. Ye, F. L. Lin, R. F. Li, J. Xu, and Y. S. Wang, High-security-level multidimensional optical storage medium: Nanostructured glass embedded with $\text{LiGa}_5\text{O}_8\text{:Mn}^{2+}$ with photostimulated luminescence, *Light Sci. Appl.* **9**, 22 (2020).
- [19] S. Y. Yan, F. Liu, J. H. Zhang, X. J. Wang, and Y. C. Liu, Persistent Emission of Narrowband Ultraviolet-B Light Upon Blue-Light Illumination, *Phys. Rev. Appl.* **13**, 044051 (2020).
- [20] X. L. Wang, Y. F. Chen, F. Liu, and Z. W. Pan, Solar-blind ultraviolet-C persistent luminescence phosphors, *Nat. Commun.* **11**, 2040 (2020).
- [21] X. Y. Zhao, F. Liu, X. J. Wang, and Y. C. Liu, Emission from Storage Phosphors that Glow Even in Bright Ambient Light, *Phys. Rev. Appl.* **15**, 064039 (2021).
- [22] A. M. Srivastava, Aspects of Pr^{3+} luminescence in solids, *J. Lumin.* **169**, 445 (2016).
- [23] F. Liu and X. J. Wang, Effects of non- $4f$ states on Pr^{3+} luminescence in phosphors, *Chin. J. Lumin.* **38**, 1 (2017).
- [24] A. Zych, M. de Lange, C. de Mello Donega, and A. Meijerink, Analysis of the radiative lifetime of Pr^{3+} $d-f$ emission, *J. Appl. Phys.* **112**, 013536 (2012).
- [25] E. Van der Kolk, P. Dorenbos, A. P. Vink, R. C. Perego, and C. W. E. van Eijk, Vacuum ultraviolet excitation and emission properties of Pr^{3+} and Ce^{3+} in MSO_4 ($M = \text{Ba}$, Sr , and Ca) and predicting quantum splitting by Pr^{3+} in oxides and fluorides, *Phys. Rev. B* **64**, 195129 (2001).
- [26] M. L. H. Ter Heerdt, E. Van der Kolk, W. M. Yen, and A. M. Srivastava, Vacuum ultraviolet spectroscopy of Pr^{3+} in CaAl_4O_7 , $\text{LaMgAl}_{11}\text{O}_{19}$ and SrLaAlO_4 , *J. Lumin.* **100**, 107 (2002).
- [27] A. M. Srivastava, M. Jennings, and J. Collins, The interconfigurational ($4f^15d^1 \rightarrow 4f^2$) luminescence of Pr^{3+} in LuPO_4 , $\text{K}_3\text{Lu}(\text{PO}_4)_2$ and LiLuSiO_4 , *Opt. Mater.* **34**, 1347 (2012).
- [28] Y. M. Yang, Z. Y. Li, J. Y. Zhang, Y. Lu, S. Q. Guo, Q. Zhao, X. Wang, Z. J. Yong, H. Li, J. P. Ma, Y. Kuroiwa, C. Moriyoshi, L. L. Hu, L. Y. Zhang, L. R. Zheng, and H. T. Sun, X-ray-activated long persistent phosphors featuring strong UVC afterglow emissions, *Light Sci. Appl.* **7**, 88 (2018).
- [29] H. Li, Q. Liu, J. P. Ma, Z. Y. Feng, J. D. Liu, Q. Zhao, Y. Kuroiwa, C. Moriyoshi, B. J. Ye, J. Y. Zhang, C. K. Duan, and H. T. Sun, Theory-guided defect tuning through topochemical reactions for accelerated discovery of UVC persistent phosphors, *Adv. Opt. Mater.* **8**, 1901727 (2020).
- [30] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevResearch.4.L012028> for Figs. S1–S9.