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Supporting Information

<u>*Title:*</u> Sunlight-activated long persistent luminescence in the ultraviolet-B from Bi^{3+} -doped garnet phosphors for covert optical tagging

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Fig. S1. XRD patterns of $Y_3Ga_{5-x}Al_xO_{12}$: 0.3%Bi³⁺ (x = 1, 2) phosphors. Standard data of cubic $Y_3Ga_5O_{12}$ phase (JCPDS No. 43-0512) is also displayed as reference.



Fig. S2. Emission spectra of YGG:x%Bi (x=0.3, 0.5, 1, 2, 3) phosphors. The emission spectraare acquired under 289 nm light excitation. The inset depicts intensities of 313 nm and 475 nmasafunctionofBidopingconcentration.



Fig. S3. (a) The excitation and emission spectra of YGG:Bi phosphor at 77 K.(b) The decaycurveofYGG:Biphosphor.



Fig. S4. The emission spectra of YGG:Bi phosphor at different temperatures ranging from 25 °C to 300 °C.



Fig. S5. Persistent luminescence decay curve (grey dash curve) monitored at 313 nm. The decay curve is normalized at the 20 s decay instant and the red line curve is the thus obtained fitting relationship between the persistent luminescence intensity (I) and the decay time (t). The measured persistent luminescence intensities are also normalized at the 20 s decay instant and displayed as green dots in the Fig. 3a.



Fig. S6. Effect of Bi doping concentration on the persistent luminescence performance of YGG:Bi phosphors. The persistent luminescence decay curves were monitored at 313 nm after irradiation by a 254 nm UV lamp for 10 min.



Fig. S7. Effect of excitation duration on the charging capability and the persistent luminescence performance of YGG:Bi phosphor. The persistent luminescence decay curves were monitored at 313 nm after irradiation by a 254 nm UV lamp for various time from 1 min to 15 min.



Fig. S8. UVB persistent luminescence decay curves of $Y_3Ga_{5-x}Al_xO_{12}:0.3\%Bi^{3+}$ (x = 0, 1, 2) phosphor disks irradiated by a 254 nm UV lamp for 15 min at room temperature.



Fig. S9. (a) UVB persistent luminescence images of YGG:Bi ceramic disc taken at different decay instants (1-60 min) in an indoor-lighting environment after irradiation by a 254 nm lamp for 15 min. (b) Persistent luminescence images of Chinese character "fu" ("fortune" in English) formed by YGG:Bi phosphor powders on whiteboard in bright indoor environment. The pattern was irradiated by 254 nm UV light for 15 min.



Fig. S10. Persistent luminescence decay curves of YGG:Bi phosphor irradiated by 240–405 nmlights for 5 min. The monitoring wavelength is 313 nm. The afterglow intensity at time of 30 safter the stoppage of the irradiation was used to plot the persistent luminescence excitationspectrumshowninFig.4a.



Fig. S11. UVB persistent luminescence decay curves of YGG:Bi phosphor disk irradiated by direct sunlight for 30 min at different moments between sunrise and sunset during the day. a, at 9:00. b, at 11:00. c, at 13:30. d, at 15:30.



Fig. S12. UVB persistent luminescence decay curves of $Y_3Ga_{5-x}Al_xO_{12}:0.3\%Bi^{3+}$ (x = 1, 2) phosphor disks after irradiation by direct sunlight for 30 min on a sunny day.



Fig. S13. UVB persistent luminescence decay curves of YGG:Bi phosphor disk irradiated by direct sunlight for 30 min in different solutions.



Fig. S14. (a) Thermoluminescence spectra of YGG:Bi after illumination by monochromic light over the 240–410 nm spectral range for 5 min. (b) Thermoluminescence spectra obtained by illuminating the YGG:Bi sample using a 340 nm monochromatic light for different exposure times from 30 s to 15 min.



Fig. S15. The reciprocal of the persistent luminescence intensity (I^{-1}) versus *t* after irradiation by 280 nm UV light for 15 min at 77 K.



Fig. S16. Thermoluminescence curve recorded over 80–673 K on a YGG:Bi phosphor disc. The sample was pre-irradiated by 254 nm UV light for 30 s at 77 K.



Fig. S17. Persistent luminescence decay curves after 340 nm excitation for 15 min at 77 K and 300 K.

Sample	YGG:Bi	YGG
Space group	Ia-3d	Ia-3d
a (Å)	12.2805(7)	12.2730(10)
b (Å)	12.2805(7)	12.2730(10)
c (Å)	12.2805(7)	12.2730(10)
V	1852.062(2)	1848.6(3)
α=β=γ (°)	90	90
Density (g/cm ³)	5.7906(8)	5.80
Rp and Rwp	8.803% and 10.805%	

Table S1. Rietveld refinement parameters of YGG:Bi phosphor. The standard data of cubic $Y_3Ga_5O_{12}$ phase is given as reference.

Decay time (s)	Intensity, I_m (pW)		
10	298.7		
20	160.2		
30	108.4		
40	85.6		
50	70.4		
60	60.8		
70	52.9		
80	47.0		
90	42.3		
100	39.1		
110	37.0		
120	32.2		
130	33.3		
140	31.2		
150	30.1		

Table S2. Intensities measured by a Newport UV photodetector (Source data are provided as a

 Source Data file).

Materials	Excitation	Maximum	Duration	Sunlight	Ref.
	sources	emission		excitable	
Lu ₃ Al ₂ Ga ₃ O ₁₂ :Pr ³⁺ , Gd ³⁺	Blue LED	313 nm	~ 1h	No	1
$Sr_3Gd_2Si_6O_{18}$: Pr^{3+}	UV lamp	311 nm	>12h	No	2
$Y_2GdAl_2Ga_3O_{12}:Pr^{3+}$	UV lamp	311 nm	>12h	No	2
$Sr_{3}Y_{2}Si_{6}O_{18}:Pb^{2+}$	UV lamp	299 nm	> 12h	No	2
$Sr_3Gd_2Si_6O_{18}{:}Pb^{2+}$	UV lamp	311 nm	> 12h	No	2
Y ₃ Al ₂ Ga ₃ O ₁₂ :Bi ³⁺	UV lamp	307 nm	~ 6h	No	2
$Y_2GdAl_2Ga_3O_{12}{:}Bi^{3+}$	UV lamp	311 nm	> 12h	No	2
BaLu ₂ Al ₂ Ga ₂ SiO ₁₂ :Pr ³⁺	UV lamp	301 nm	> 3h	No	3
Y ₃ Ga ₅ O ₁₂ :Bi ³⁺	UV lamp	313 nm	>60h	Yes	This work

Table S3. Comparison of the reported UVB persistent phosphors

[1] S. Yan, F. Liu, J. Zhang, X. Wang, Y. Liu, *Phys. Rev. Applied*, 2020, **13**, 044051.

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[3] W. Yuan, T. Tan, H. Wu, R. Pang, S. Zhang, L. Jiang, D. Li, Z. Wu,

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