

## Supporting information

# Novel persistent phosphors of lanthanide-chromium co-doped yttrium aluminum gallium garnet: design concept with vacuum referred binding energy diagram

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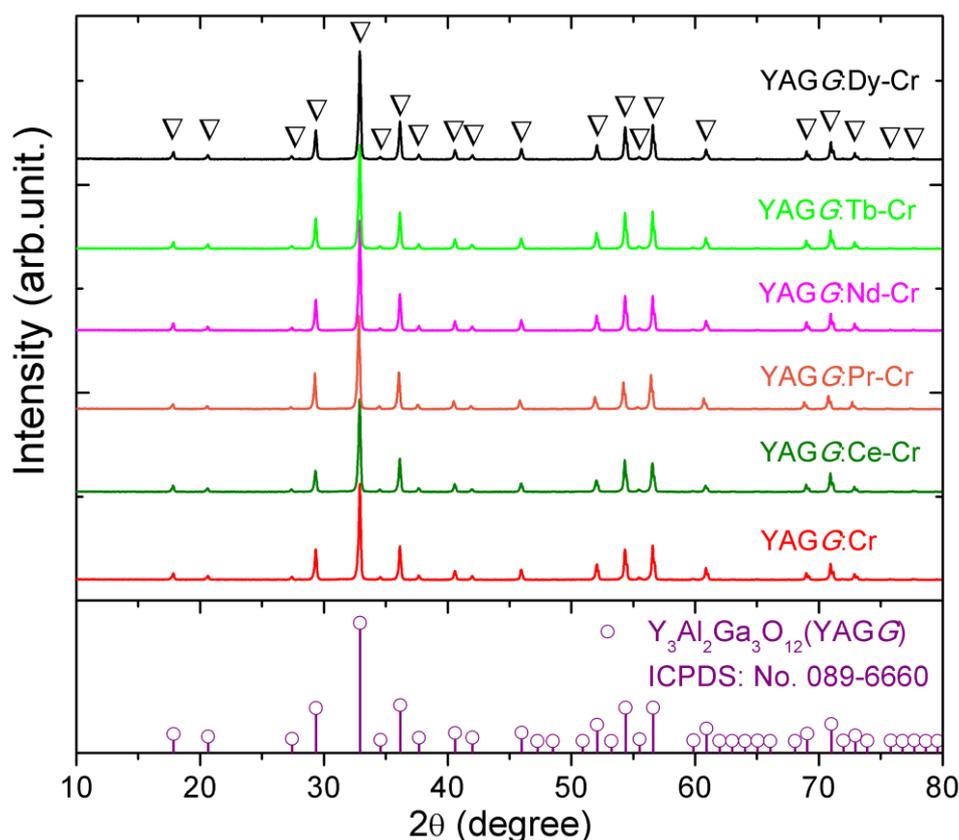


Fig. S1. X-ray diffraction (XRD) patterns of the YAGG:Cr and YAGG:Ln-Cr ( $Ln$ =Ce, Pr, Nd, Tb, Dy) persistent phosphors

The phase compositions of YAGG:Cr and YAGG:Ln-Cr ( $Ln$ =Ce, Pr, Nd, Tb, Dy) transparent ceramic persistent phosphors were measured by X-ray diffraction (XRD) measurements (Ultima IV, Rigaku), utilizing nickel filtered  $\text{CuK}_\alpha$  radiation ( $1.5406 \text{ \AA}$ ). All of the as-prepared samples are confirmed to be single phase matching well with the standard card (ICPDS: No. 089-6660) of  $\text{Y}_3\text{Al}_2\text{Ga}_3\text{O}_{12}$  (YAGG).

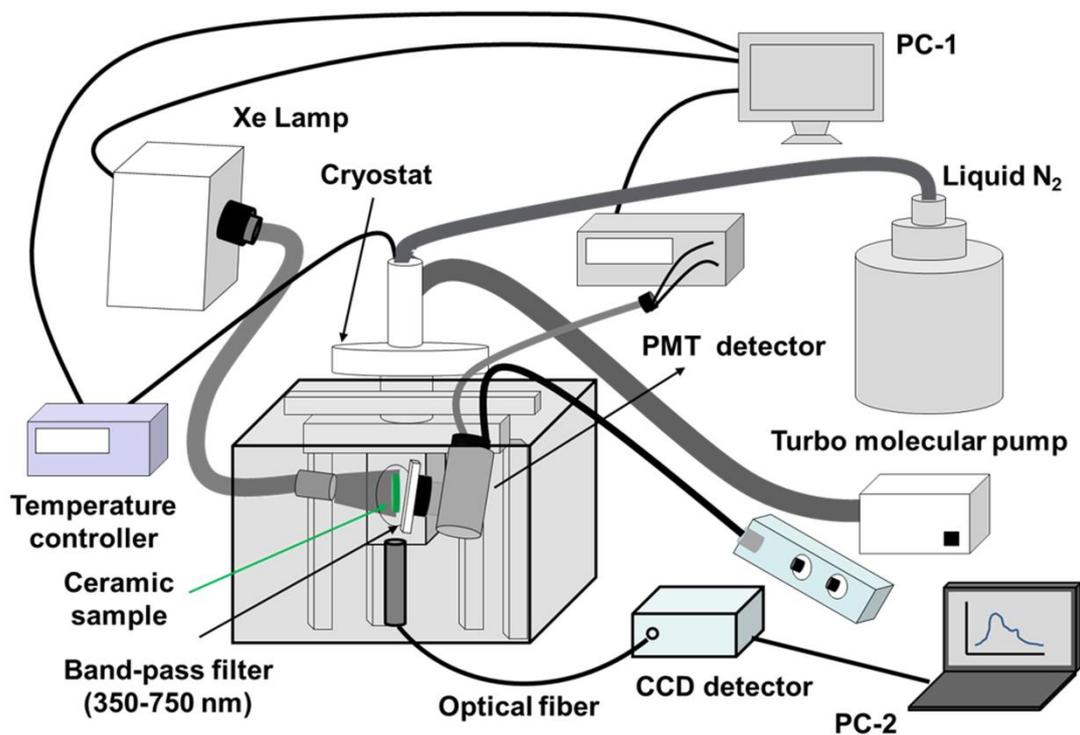


Fig. S2. The measurement setup of thermoluminescence (TL) two-dimensional (2D) plot measurements

A 300 W Xe-lamp (MAX-302, Asahi Spectra) with a UV mirror module (250-400 nm) was used as the excitation source. The ceramic sample was set in a cryostat (Helitran LT3, Advanced Research Systems) to control temperatures and firstly illuminated by UV light at 150 K for 10 min, then heated up to 550 K at a rate of 10 K/min at 10 min after ceasing the illumination, and the thermoluminescence (TL) signals were recorded by a PMT detector (R11041, Hamamatsu Photonics & Co. Ltd.) covered with 350 nm short-cut and 750 nm long-cut filters. The Si CCD spectrometer (QE65-Pro, Ocean Optics) was operated simultaneously with the TL measurement to always monitor the emission spectra at different temperatures.

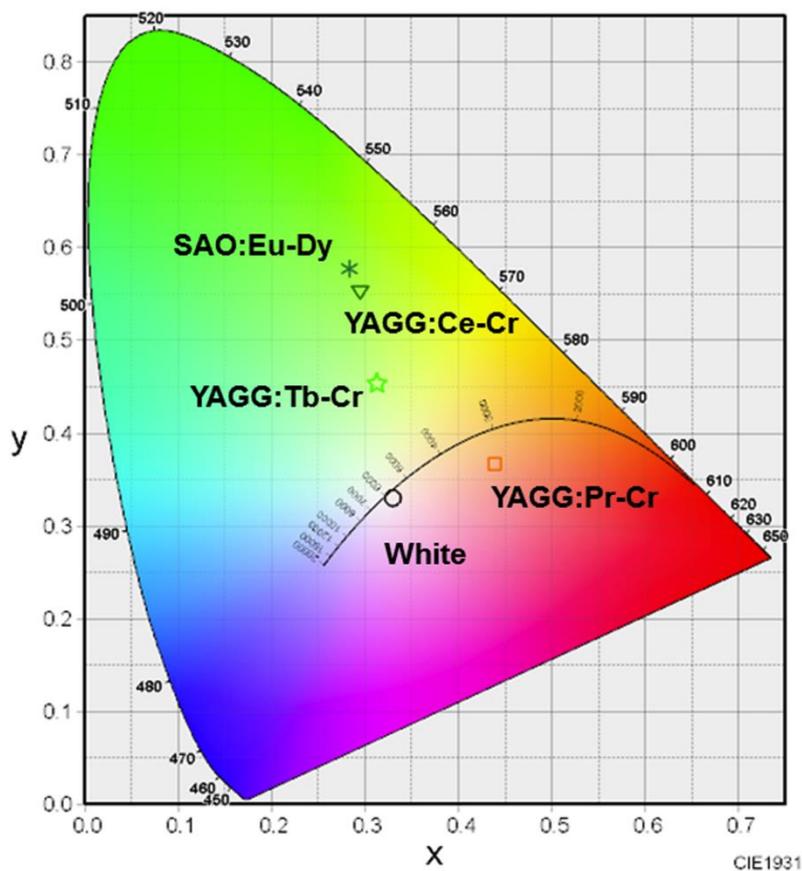


Fig. S3. Persistent luminescent color coordinates of YAGG:Ce-Cr, YAGG:Pr-Cr and YAGG:Tb-Cr transparent ceramic persistent phosphors in CIE 1931 chromaticity diagram compared with that of the commercial  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}\text{-Dy}^{3+}$  green persistent phosphor

The color coordinates of persistent luminescence in CIE 1931 chromaticity diagram are (0.295, 0.553) for YAGG:Ce-Cr, (0.439, 0.367) for YAGG:Pr-Cr, and (0.313, 0.454) for YAGG:Tb-Cr ceramic phosphors compared with (0.283, 0.577) for the commercial  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}\text{-Dy}^{3+}$  green persistent phosphor (LumiNova-GLL300FFS, Nemoto & Co. Ltd.).

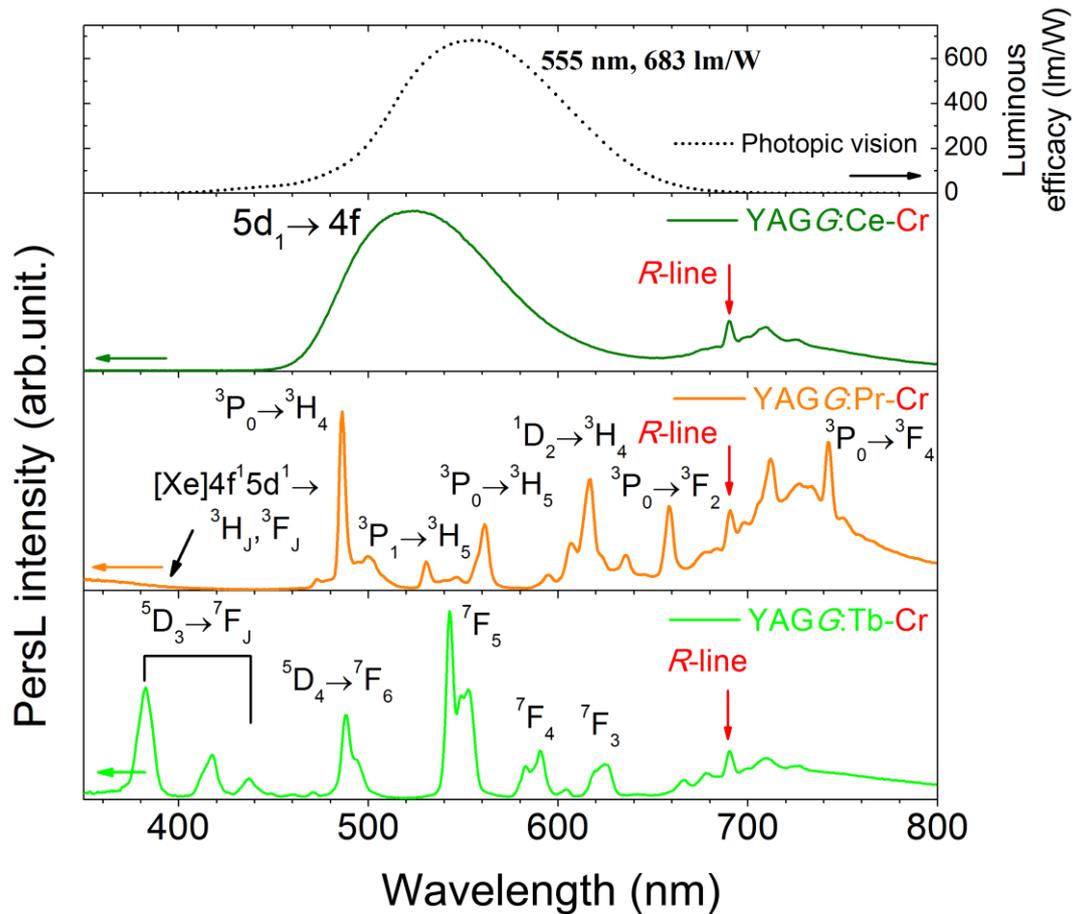


Fig. S4. (a) Luminous efficacy of human eyes in photopic vision and persistent luminescent spectra of (b) YAGG:Ce-Cr (c) YAGG:Pr-Cr (d) YAGG:Tb-Cr persistent phosphors at 5 min after ceasing UV (250-400 nm) illumination

Photopic vision which is mediated by cone cells in human eyes allows color perception and shows a broad band sensitivity curve (from 380 to 780 nm) peaked at 555 nm with 683 lm/W luminous efficacy. The typical band emission due to  $\text{Ce}^{3+}:5d_1 \rightarrow 4f$ , line emissions due to  $\text{Pr}^{3+}:^3P_{J(J=0, 1, 2)} \rightarrow ^3H_{4,5}$ ,  $^1D_2 \rightarrow ^3H_4$ , and  $\text{Tb}^{3+}:^5D_4 \rightarrow ^7F_J$  ( $J=6, 5, 4, 3$ ) in YAGG:Ce-Cr, YAGG:Pr-Cr, YAGG:Tb-Cr ceramic samples match well with the sensitivity curve of photopic vision.

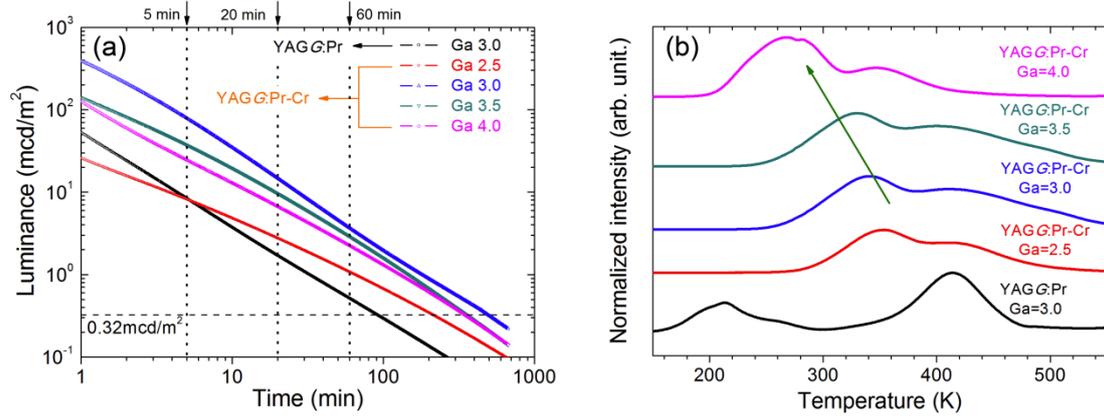


Fig. S5. (a) Persistent luminescent decay curves recorded at 25°C after ceasing UV (250-400 nm) illumination for 5 min of YAGG:Pr (Ga=3.0) and YAGG:Pr-Cr (Ga=2.5, 3.0, 3.5, 4.0) transparent ceramic persistent phosphors (b) normalized thermoluminescence (TL) glow curves of corresponding samples

Table S1. Luminance and duration of YAGG:Pr (Ga=3.0) and YAGG:Pr-Cr (Ga=2.5, 3.0, 3.5, 4.0) transparent ceramic persistent phosphors after ceasing UV (250-400 nm) illumination for 5 min

Sample composition	5 min (mcd/m <sup>2</sup> )	20 min (mcd/m <sup>2</sup> )	60 min (mcd/m <sup>2</sup> )	Duration upon 0.32 mcd/m <sup>2</sup> (min)
YAGG:Pr <sup>3+</sup> (Ga=3.0)	8.4	1.7	0.51	93±1
YAGG:Pr <sup>3+</sup> -Cr <sup>3+</sup> (Ga=2.5)	8.2	2.8	1.07	216±1
YAGG:Pr <sup>3+</sup> -Cr <sup>3+</sup> (Ga=3.0)	79.1	14.8	3.66	499±1
YAGG:Pr <sup>3+</sup> -Cr <sup>3+</sup> (Ga=3.5)	37.4	9.6	2.87	359±1
YAGG:Pr <sup>3+</sup> -Cr <sup>3+</sup> (Ga=4.0)	24.6	6.7	2.24	349±1

Compared with the Pr<sup>3+</sup> singly doped YAGG (YAGG:Pr) persistent phosphor with the same Ga<sup>3+</sup> content (Ga=3.0), the persistent luminescent intensity of the YAGG:Pr-Cr sample was enhanced by co-doping Cr<sup>3+</sup> which acts as an additional electron trap with ideal trap depth for Pr<sup>3+</sup> persistent luminescence working at room temperature.

The peak temperature of the normalized TL glow curves for YAGG:Pr-Cr samples shifts to lower temperature with increasing Ga<sup>3+</sup> content (from 2.5 to 4.0). Since the TL peak temperature is correlated to the trap depth, the downshift of glow peak indicates that the trap depth becomes shallower. Because the bottom of conduction band of Ga<sup>3+</sup>-free garnet is composed by *d* orbitals of lanthanide ions while when Ga<sup>3+</sup> is added, 4*s* orbitals of Ga<sup>3+</sup> start to dominate the conduction band edge [1]. Therefore, the trap depth becomes shallower with increasing Ga<sup>3+</sup> content so that the persistent luminescent behaviors such as initial intensity and duration can be easily modified by changing Ga<sup>3+</sup> content in YAGG:Pr-Cr orange persistent phosphors.

[1] M. Fasoli, A. Vedda, M. Nikl, C. Jiang, B. P. Uberuaga, D. A. Andersson, K. J. McClellan, and C. R. Stanek, *Phys. Rev. B.* **84**, 081102 (2011).

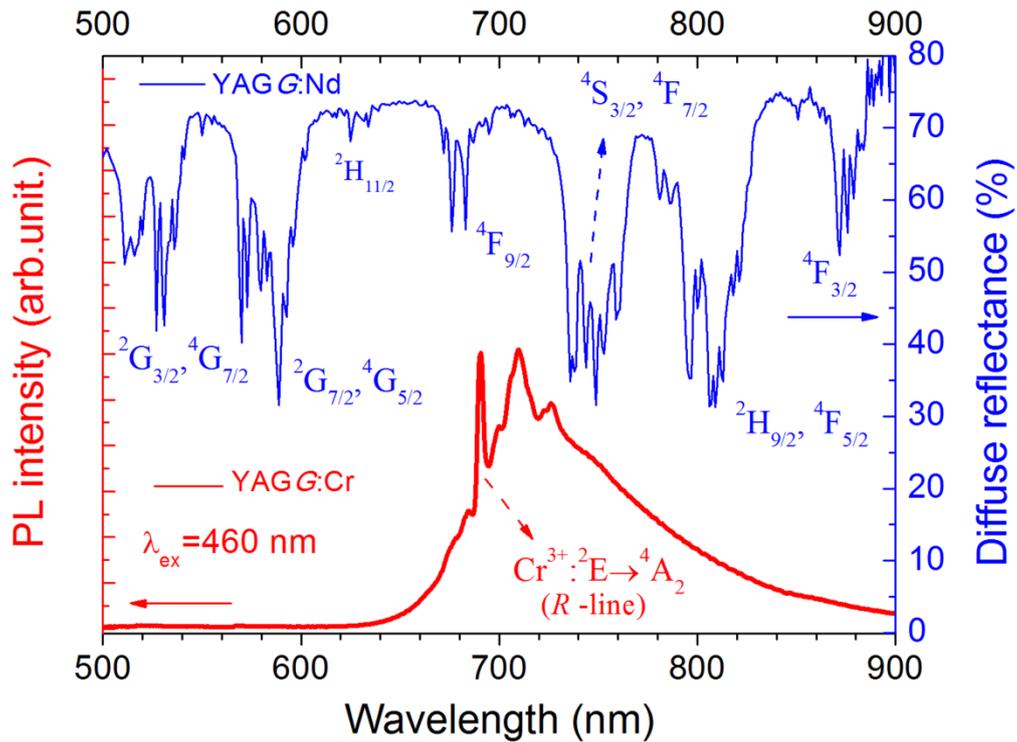


Fig. S6. Photoluminescence (PL) spectrum ( $\lambda_{\text{ex}}=460$  nm) of the YAGG:Cr and diffuse reflectance of the YAGG:Nd ceramic samples

Förster resonant energy transfer takes place when the oscillations of optical-induced electronic coherence on the donor are in resonance with the electronic energy gap of the acceptor. Briefly, this non-radiative energy transfer process requires sufficient spectral overlap between donor emission and acceptor absorption bands. Since the broad band emission of donor ( $\text{Cr}^{3+}$ ) (including *R*-line and its phonon sidebands) matches well with the absorption band of acceptor ( $\text{Nd}^{3+}$ ) in the same  $\text{Y}_3\text{Al}_2\text{Ga}_3\text{O}_{12}$  (YAGG) host, resonant energy transfer is possible from  $\text{Cr}^{3+}$  to  $\text{Nd}^{3+}$ .