Supplementary information

Visualization of trap distribution in $SrAl_2O_4:Eu^{2+},Dy^{3+}$ revealed by simulations of luminescence change in charging and emission of persistent luminescence and thermoluminescence based on a random walk model

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S-1. Experimental

Preparation of PersL SAOED$_x$ (x = 2, 4, 5, 6, 8, 10)

$Sr_{0.9}Al_2O_4:Eu^{2+}_{0.01x},Dy^{3+}_{(0.1-0.01x)}$ (x = 2, 4, 5, 6, 8, 10), SAOED$_x$, were synthesized according to the procedure described in the literature.[s1] To a mixture of $SrCO_3$ and Al_2O_3 powders in the molar ratio 0.9 : 1.0, Eu_2O_3 and Dy_2O_3 powders were added so as to be the molar ratios of 0.02 : 0.08, 0.04 : 0.06, 0.05 : 0.05, 0.06 : 0.04, 0.08 : 0.02, or 0.10 : 0.00. After grinding for 1 h with a mortar and a pestle, the resulting powders were pressed under 2 t for 15 min to become pellet-like solid. The pellets of SAOED$_x$ were placed in a SILICONIT electric furnace TEXSH-530 under argon atmosphere containing 1% hydrogen gas and heated up from room temperature to 1500 °C for over 5 h. The temperature of the electric furnace was maintained at 1500 °C for 2 h and then decreased to room temperature over one night.

Reference

s1. R. M. Calderón-Olvera, E. A. Albanés-Ojeda, M. García-Hipólito, J. M. Hernández-Alcántara, M. A. Álvarez-Perez, C. Falcony, O. Álvarez-Fregoso, *Ceram. Int.*, 2018, **44**, 7917.

Measurement

PL and PeasL of SAOED$_x$ (x = 2, 4, 5, 6, 8, 10) were recorded on a JASCO spectrofluorometer FP-6500. PXRD measurements were performed using a Bruker AXS NEW D8 ADVANCE with the Cu Kα radiation (1.5406 Å) at 40 kV and 40 mA for SAOED$_x$ powders obtained by griding with mortar and pestle. We employed an Oxford microstat N equipped with a thermo-controller Oxford ITC502 for the temperature control for TL spectra. The temperature of SAOED$_x$ powders increased at a rate of ca. 1 K/s from 200 K to 500 K. TL spectra were depicted against temperature corresponding to time from the beginning of the observation. For the TL measurement, after the samples were heated at 500 K for de-trapping under the dark and cooled to 200 K by liquid nitrogen, they were excited with 370 nm (10 W) LED light for 10 min and left to stand in the dark.

S-2. PL emission and excitation spectra of SAOED$_x$ (x = 2, 4, 5, 6, 8, 10)

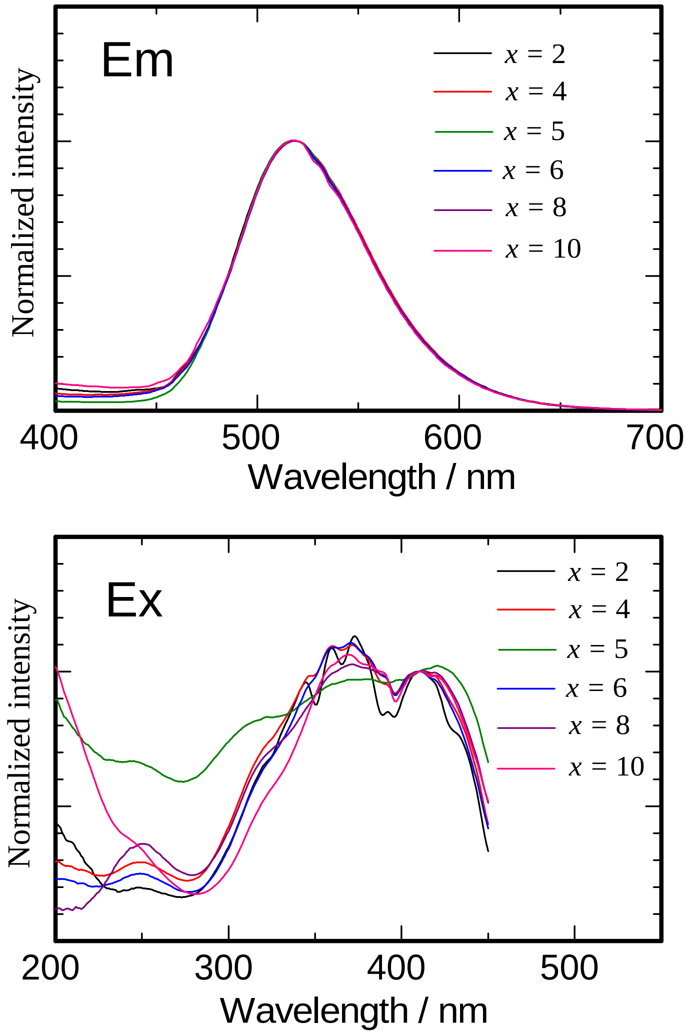

Figure S1. PL emission spectra (top) and excitation spectra (bottom) of SAOED$_x$ (x = 2 (red), 4 (orange), 5 (green), 6 (dark green), 8 (cyan), 10 (blue).

S-3. PXRD of SAOED$_x$ (x = 2, 4, 5, 6, 8, 10)

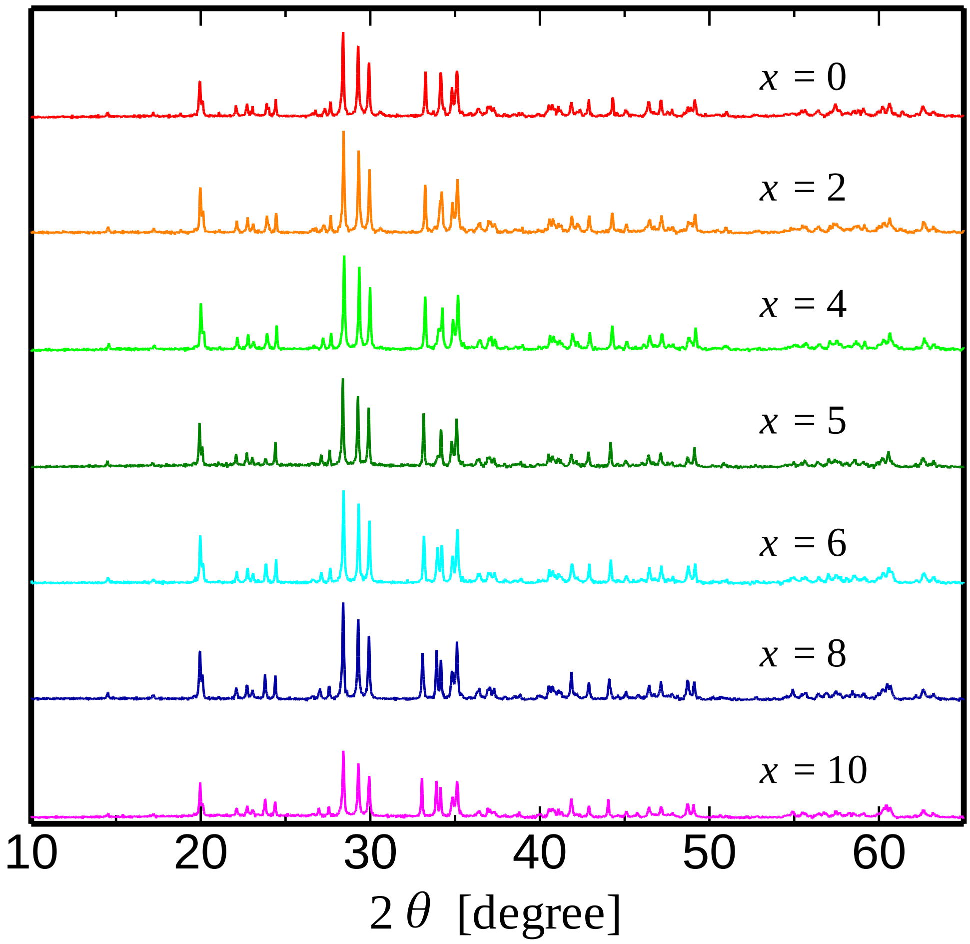

Figure S2. PXRD patterns of SAOED$_x$, where x = 0 (red), 2 (orange), 4 (green), 5 (dark green), 6 (cyan), 8 (blue), 10 (purple).

S-4. Lattice constants (*a*, *b*, *c*, β) for SAOED$_x$ (*x* = 2, 4, 5, 6, 8, 10) crystals

Table S1. Lattice constants (*a*, *b*, *c*, β) for SAOED$_x$ in the monoclinic crystal system evaluated from the PXRD patterns using the Bragg's equation.

x	*a* / Å	*b* /Å	*c* /Å	*β* /°
0	8.384	8.887	5.198	93.403
2	8.381	8.879	5.193	93.400
4	8.367	8.870	5.188	93.396
5	8.380	8.904	5.208	93.406
6	8.374	8.887	5.198	93.401
8	8.382	8.887	5.191	93.257
10	8.383	8.887	5.186	93.298

S-5. Electron populations in reservoirs N_m (m = 0-6) for SAOED$_x$ (x = 2, 4, 5, 6, 8, 10)

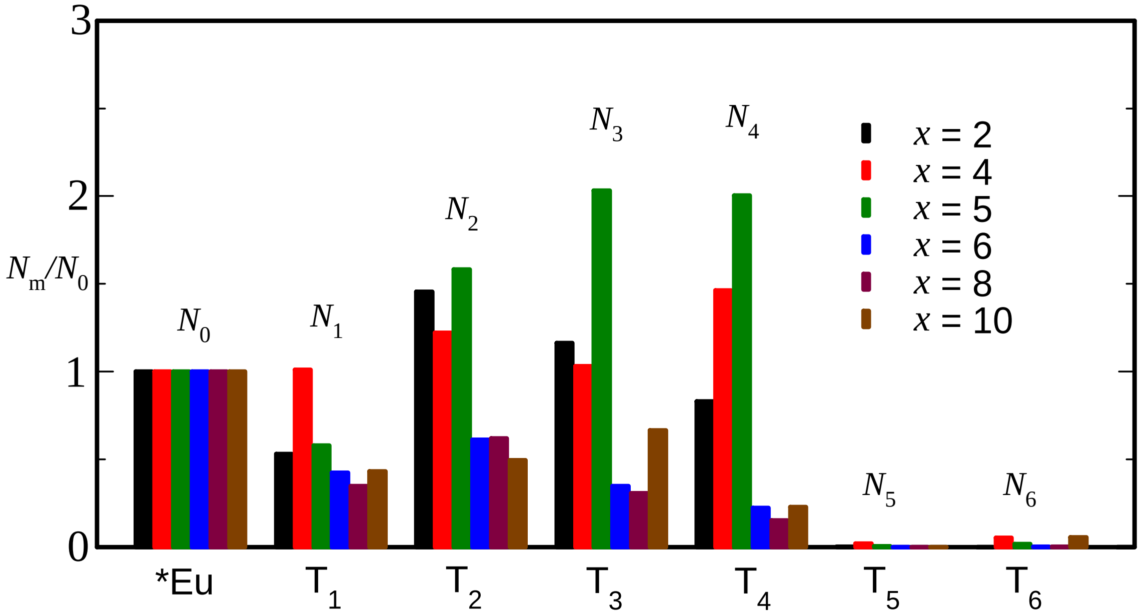

Figure S3. Electron populations in reservoirs N_m (m = 0–6) for SAOED$_x$ (x = 2, 4, 5, 6, 8, 10) predicted by the simulation of charging and PersL processes based on RWM.

S-6. Luminescence changes in charging and PersL processes for SAOED$_x$ (x = 2, 4, 5, 6, 8, 10)

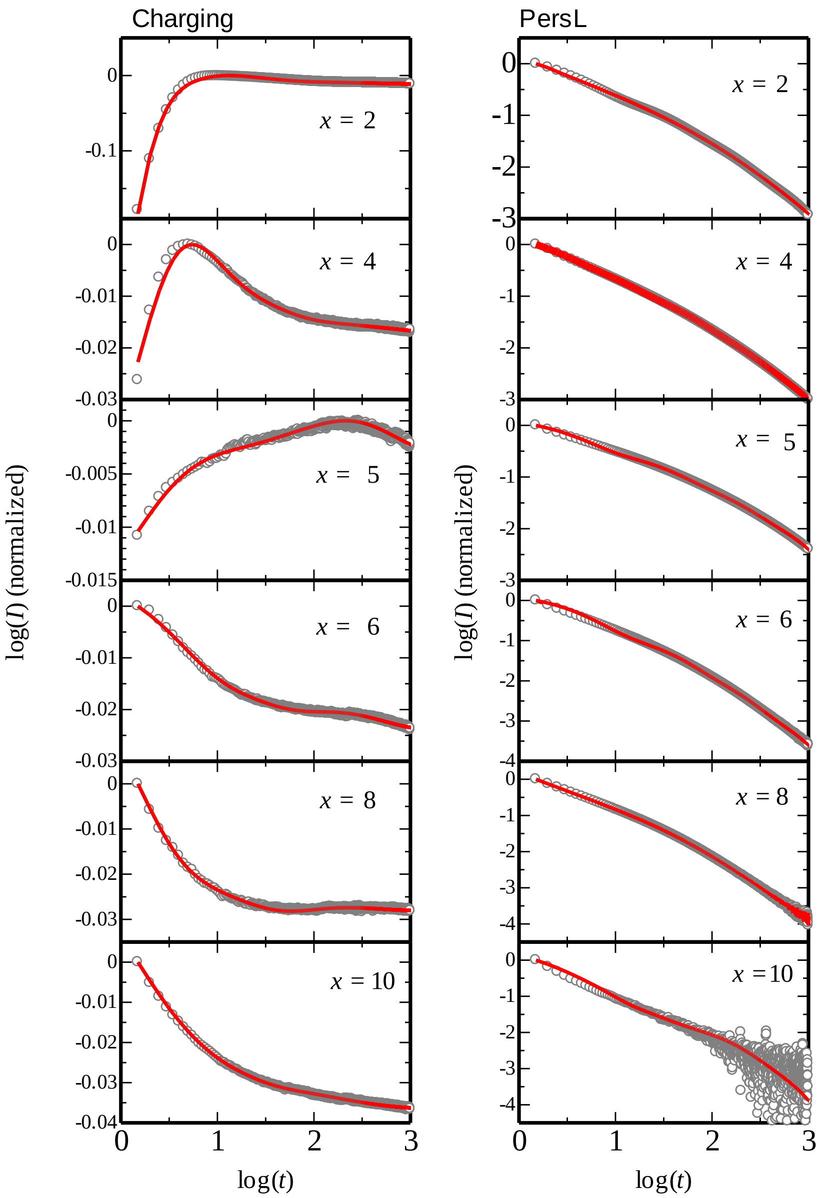

Figure S4. Luminescence changes in charging and PersL processes for SAOED$_x$ (x = 2, 4, 5, 6, 8, 10). The red lines are the best fits for the curve fitting using equations $I_c = I_{c1}\exp(-t/\tau_{c1}) + I_{c2}\exp(-t/\tau_{c2}) - I_{c3}\exp(-t/\tau_{c3}) + I_{c0}$ for the charging (left) and $I_p = I_{p1}\exp(-t/\tau_{p1}) + I_{p2}\exp(-t/\tau_{p2}) + I_{p3}\exp(-t/\tau_{p3})$ for PersL (right).

S-7. Parameters determined through the simulation of the luminescence changes in charging and PersL for SAOED$_x$ (x = 2, 4, 5, 6, 8, 10)

Table S2. Parameters determined through the simulation of the luminescence changes in charging and PersL for SAOED$_x$ (x = 2, 4, 5, 6, 8, 10) using equations $I_c = I_{c1}\exp(-t/\tau_{c1}) + I_{c2}\exp(-t/\tau_{c2}) + I_{c3}\exp(-t/\tau_{c3}) + I_{c0}$ for the charging and $I_p = I_{p1}\exp(-t/\tau_{p1}) + I_{p2}\exp(-t/\tau_{p2}) + I_{p3}\exp(-t/\tau_{p3})$.

x	Charging		PersL	
2	I_{c1} = 0.025 I_{c2} = 0.26 I_{c3} = -1.1 I_{c0} = 0.72	τ_{c1} = 44 s τ_{c2} = 51000 s τ_{c3} = 1.3 s	I_{p1} = 89% I_{p2} = 10% I_{p3} = 1%	τ_{p1} = 5.0 s τ_{p2} = 65 s τ_{p3} = 430 s
4	I_{c1} = 0.040 I_{c2} = 0.26 I_{c3} = -0.46 I_{c0} = 0.70	τ_{c1} = 23 s τ_{c2} = 61000 s τ_{c3} = 0.74 s	I_{p1} = 89% I_{p2} = 9% I_{p3} = 1%	τ_{p1} = 7.3 s τ_{p2} = 71 s τ_{p3} = 450 s
5	I_{c1} = 0.019 I_{c2} = -0.025 I_{c3} = -0.0083 I_{c0} = 0.98	τ_{c1} = 2600 s τ_{c2} = 3.0 s τ_{c3} = 70 s	I_{p1} = 92% I_{p2} = 7% I_{p3} = 1%	τ_{p1} = 3.7 s τ_{p2} = 49 s τ_{p3} = 450 s
6	I_{c1} = 490 I_{c2} = 0.017 I_{c3} = 6.3×10^{-6} I_{c0} = 0.94	τ_{c1} = 8.0 s τ_{c2} = 1400 s τ_{c3} = 10000 s	I_{p1} = 90% I_{p2} = 9% I_{p3} = 1%	τ_{p1} = 2.6 s τ_{p2} = 27 s τ_{p3} = 170 s
8	I_{c1} = 0.098 I_{c2} = 0.029 I_{c3} = 1.9×10^{-5} I_{c0} = 0.94	τ_{c1} = 1.5 s τ_{c2} = 8.5 s τ_{c3} = 590 s	I_{p1} = 91% I_{p2} = 8% I_{p3} = 1%	τ_{p1} = 4.9 s τ_{p2} = 57 s τ_{p3} = 380 s
10	I_{c1} = 0.079 I_{c2} = 0.035 I_{c3} = 0.013 I_{c0} = 0.92	τ_{c1} = 1.9 s τ_{c2} = 11 s τ_{c3} = 190 s	I_{p1} = 93% I_{p2} = 6% I_{p3} = 1%	τ_{p1} = 3.2 s τ_{p2} = 49 s τ_{p3} = 270 s