Electronic Supplementary Information

Pillar[5]arene Noncovalent Assembly Boosting Full-color Lanthanide Luminescent Supramolecular Light Switch

Wei-Lei Zhou^{+a,b}, Xian-Yin Dai^{+a,c}, Wenjing Lin^a, Yong Chen^a, and Yu Liu^{*a}

^a College of Chemistry, State Key Laboratory of Elemento-Organic Chemistry, Nankai University, Tianjin 300071,

P. R. China, E-mail: yuliu@nankai.edu.cn

^b College of Chemistry and Material Science, Innovation Team of Optical Functional Molecular Devices, Inner

Mongolia Minzu University, Tongliao 028000, P. R. China.

^c School of Chemistry and Pharmaceutical Engineering, Shandong First Medical University & Shandong Academy

of Medical Sciences, Taian, Shandong 271016, P. R. China

⁺ These authors contributed equally to this work.

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1. Synthetic Protocols



Fig. S1 ^1H NMR (400 MHz) spectrum of 1 in CDCl3 at 25 °C.



Fig. S2 HR-MS (ESI) spectrum of 1.



Fig. S3 ¹H NMR (400 MHz) spectrum of 2 in CDCl₃ at 25 °C.



Fig. S4 HR-MS (ESI) spectrum of 2.



Fig. S5 ¹H NMR (400 MHz) spectrum of H in DMSO-d₆ at 25 °C.



Fig. S6 HR-MS (ESI) spectrum of H.



Fig. S7 ¹H NMR (400 MHz) spectrum of 4 in DMSO-d₆ at 25 °C.



Fig. S8 HR-MS (ESI) spectrum of 4.



Fig. S9 ¹H NMR (400 MHz) spectrum of G_1 in DMSO-d₆ at 25 °C.



Fig. S10 HR-MS (ESI) spectrum of G₁.



Fig. S11 ¹H NMR (400 MHz) spectrum of G_2 in D_2O at 25 °C.



Fig. S12 HR-MS (ESI) spectrum of G₂.

2. Characterization Data



Fig. S13 (a) ¹H NMR spectral change from G_1 to G_1' upon irradiation with UV light (254 nm) and (b) a complete recovery from G_1' to G_1 upon subsequent irradiation with visible light (>450 nm) in DMSO-d₆ at 25 °C.

Fig. S14 Absorption spectra of the closed-ring state G₁' (a) at different time and (b) at different temperature in water([H] =

0.01 mM).

Fig. S15 Absorption spectra of (a) $H+G_2$, (b) $H+H_2O$ and (c) Job' plots for host molecule H and guest G_2 in aqueous solution (absorption changes at 295nm, $[H] + [G_2] = 0.05$ mM).

Fig. S16 (a) Absorption spectra of H (0.03 mM) upon addition of G_2 (from 0 to 5.0 eq) in water. (b) Emission spectra of H (0.05 mM) upon addition of G_2 (from 0 to 1.1 eq). Plots of the emission change at 355 nm versus H concentration in water.

Fig. S17 Time-resolved photoluminescence decay fitting curves of (a) H/Tb^{3+} ; (b) $H/Tb^{3+}/G_1$ measured for 545 nm and (c) H/Eu^{3+} (d) $H/Eu^{3+}/G_1$ for 615 nm in aqueous solution at 298 K (H = 0.02 mM).

Fig. S18 The emission spectrum changes of reference G₂ in H (a) aqueous solution and (b) organic solution, respectively.

Fig. S19 Emission spectra of H (0.01 mM) upon addition of (a) Tb³⁺ and (b) Eu³⁺in CH₂Cl₂ solution.

Fig. S20 Time-resolved photoluminescence decay fitting curves of (a) H/Tb^{3+} ; (b) $H/Tb^{3+}/G_1$ measured for 545 nm and (c) H/Eu^{3+} ; (d) $H/Eu^{3+}/G_1$ measured for 615 nm in CH_2Cl_2 solution (H = 0.02 mM).

Fig. S21 The optical transmittance of H and H/Eu³⁺/G₁ at 500 nm in water ([H] = 1.0×10^{-4} M)

Fig. S22 TEM images of (a) H, (b) H/Eu^{3+} and (c) $H/Eu^{3+}/G_1$.

Fig. S23 The TEM-EDS spectrum of (a) H, (b) H/Eu^{3+} and (c) $H/Eu^{3+}/G_1.$

Fig. S24 The DLS data of (a) H and (b) $H/Eu^{3+}/G_1$ assembly (H = 0.02 mM).

Fig. S25 Zeta potential results of H, H/Eu^{3+} , $H/Eu^{3+}/G_1$ and $H/Eu^{3+}/G_1'$ (pH = 6.8).

Fig. S26 The excitation spectra of $H/Eu^{3+}/G_1$.

Fig. S27 Time-resolved photoluminescence decay curves of H/Ln³⁺/G₁' (a) in aqueous solution and (b) CH₂Cl₂ solution.

Fig. S28 Time-resolved photoluminescence decay fitting curves of $H/Tb^{3+}/G_1$ ' (a) in aqueous solution; (b) CH_2Cl_2 solution, and $H/Eu^{3+}/G_1$ ' (c) in aqueous solution; (d) CH_2Cl_2 solution.

Fig. S29 Luminescent Cyclic images changes under UV light of characters "NKU" written by $H/Eu^{3+}/G_1$ dichloride solution under alternant UV (254 nm) and visible (>450 nm).

Fig. S30 (a) Colors change under nature light and (b) Luminescent images changes under UV light of characters "N" written by Commercially available 2-(3', 3'-Dimethyl-6-nitrospiro[chromene]-2,2'-indolin]-1'-yl)ethanol) dye methanol solution (5.0 $\times 10^{-5}$ M).