

Supporting Information

Luminescence Tuning of Imidazole-Based Lanthanide (III)

Complexes [Ln = Sm, Eu, Gd, Tb, Dy]

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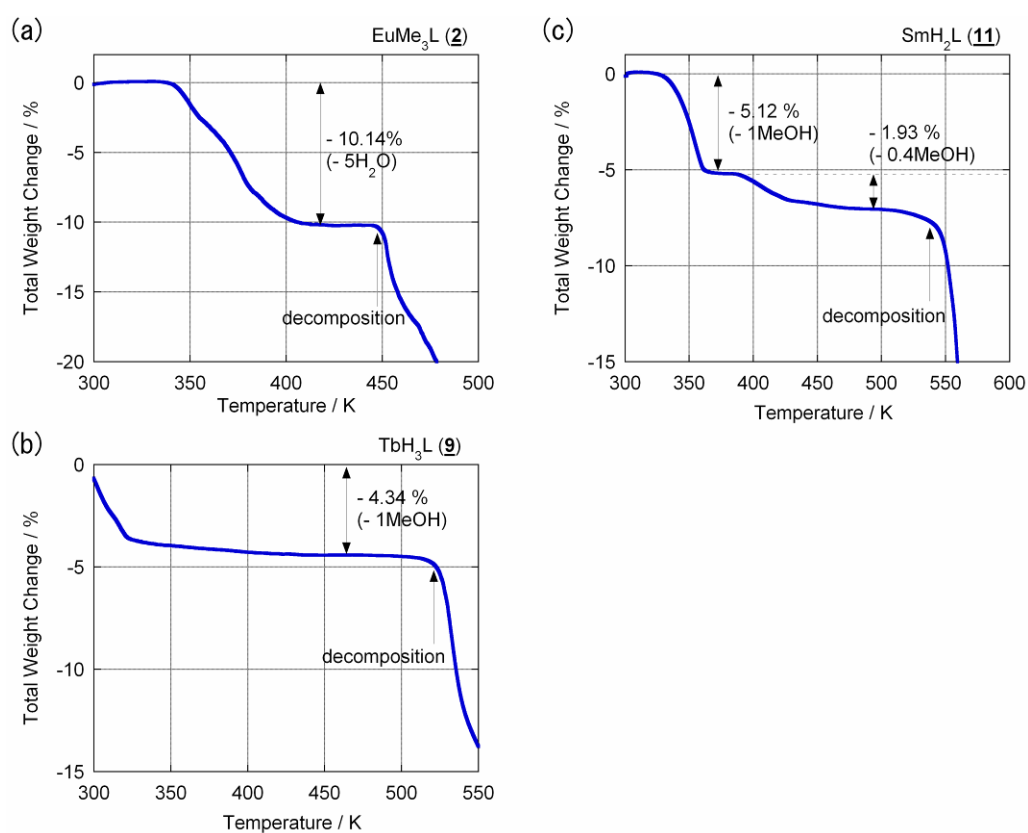


Fig. S1 Thermogravimetric (TG) curves of EuMe_3L **2** (a), TbH_3L **9** (b), and SmH_2L **11** (c), as a representative example. The heating rate was 5 °C/min in a dried nitrogen stream. One of the methanol molecules of H_2L complexes could not be removed before the decomposition of complex.

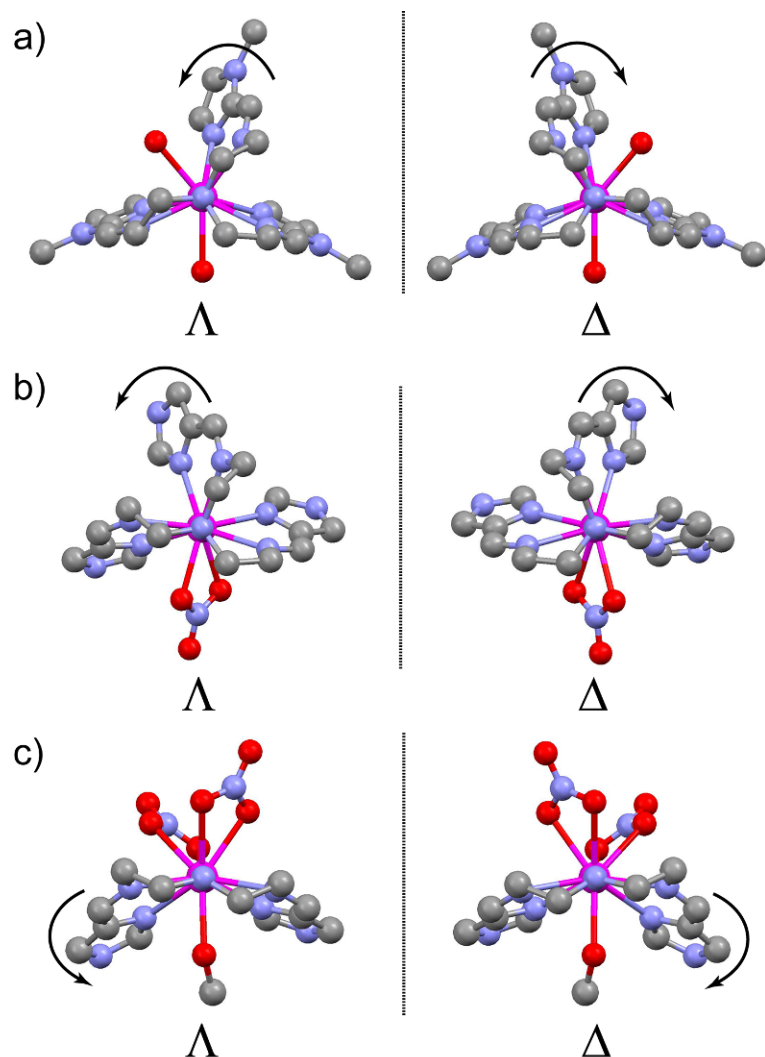


Fig. S2 Clockwise (Δ) and anti-clockwise (Λ) enantiomorphs of (a) Me₃L complex, (b) H₃L complex, and (c) H₂L complex.

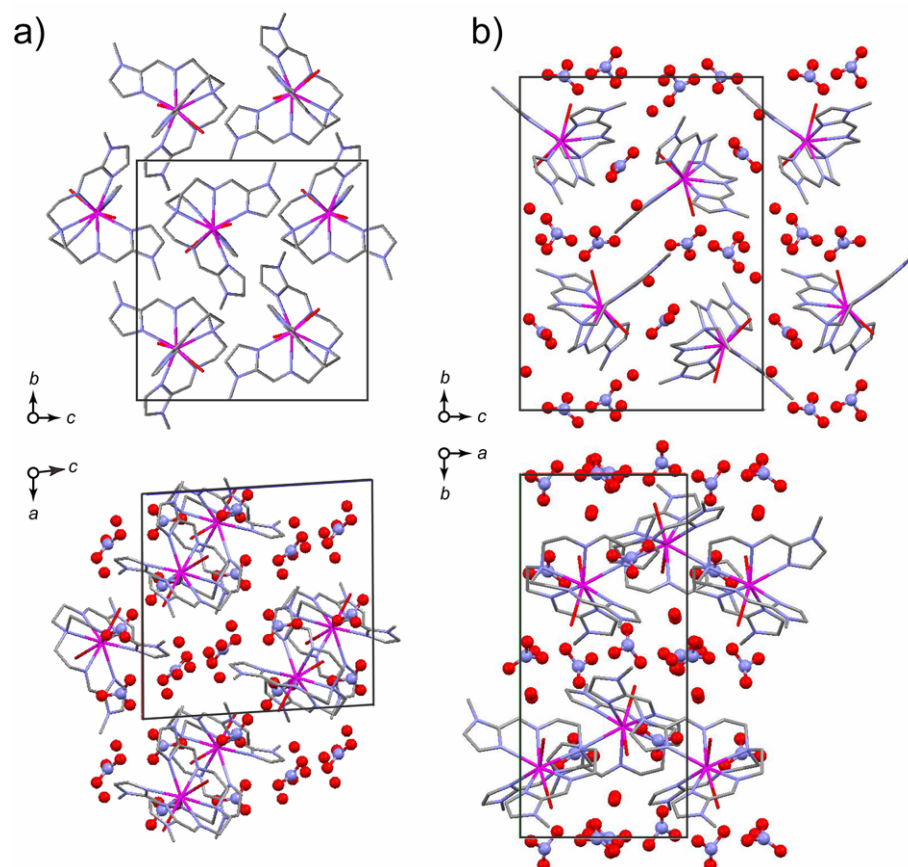


Fig. S3. Crystal packing diagram of Sm·1 (a) projected along the *a*-axis (top) and *b*-axis (bottom) and Gd·3; (b) projected along the *a*-axis (top) and *c*-axis (bottom). Crystalline solvents and counter anions are omitted for clarity in the *a*-axis view of Sm·1.

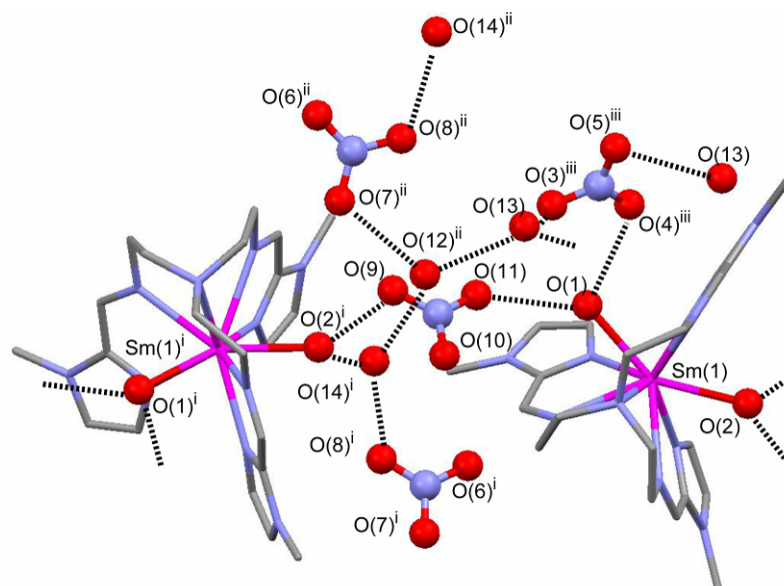


Fig. S4. Crystal structure of Sm·1, showing network structure connected by hydrogen bonds through coordinated H₂O molecule, crystalline water, and counter anion, NO₃[−]. Symmetry operations: i (0.5+x, 0.5−y, −0.5+z), ii (1−x, 1−y, 1−z), iii (1.5−x, 0.5+y, 1.5−z).

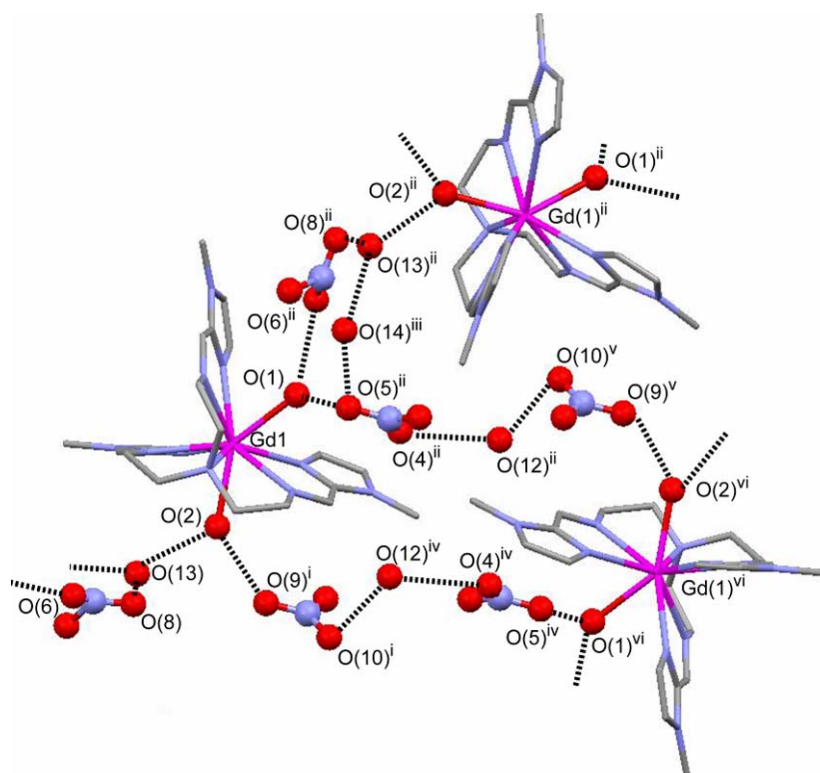


Fig. S5. Crystal structure of Gd·3, showing network structure connected by hydrogen bonds through coordinated H₂O molecules, crystalline water, and counter anion, NO₃[−]. Symmetry operations: i (x, y, −1+z), ii (0.5−x, 0.5+y, 0.5−z), iii (1.5−x, 0.5+y, 0.5−z), iv (−0.5+x, 1.5−y, 0.5+z), v (1−x, 2−y, 1−z), vi (1−x, 2−y, −z).

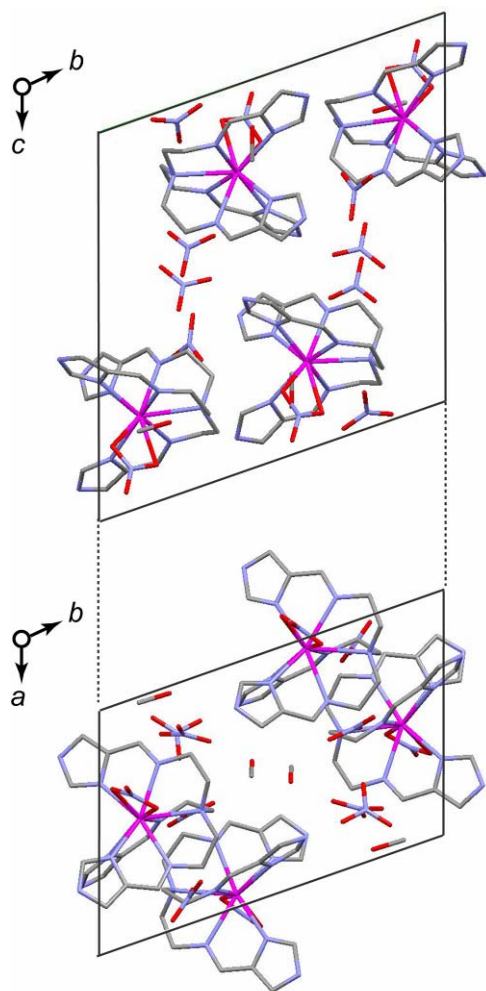


Fig. S6. Crystal packing diagram of Sm·6 (a) projected along the *a*-axis (top) and *c*-axis (bottom) (hydrogen atoms omitted for clarity).

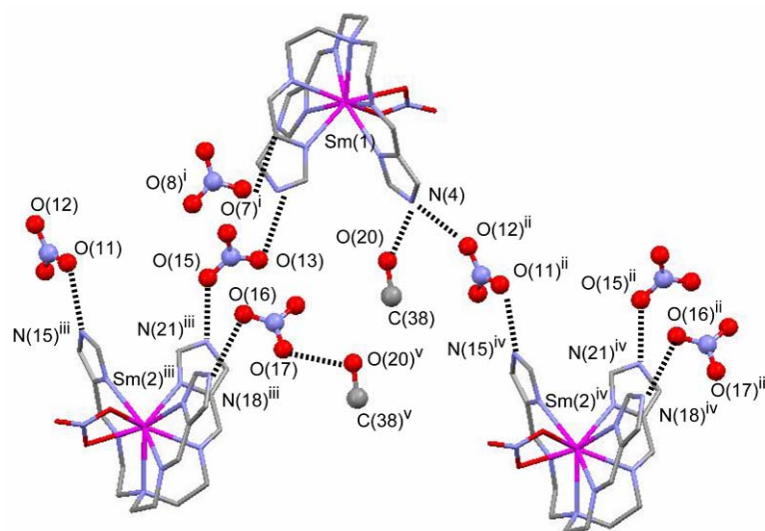


Fig. S7. Crystal structure of Sm·6, showing a network structure connected by hydrogen bonds through crystalline MeOH and counter anion, NO_3^- . Symmetry operations: i ($1+x, y, z$), ii ($x, y, -1+z$), iii ($x, 1+y, z$), iv ($x, 1+y, -1+z$), v ($-1+x, y, z$).

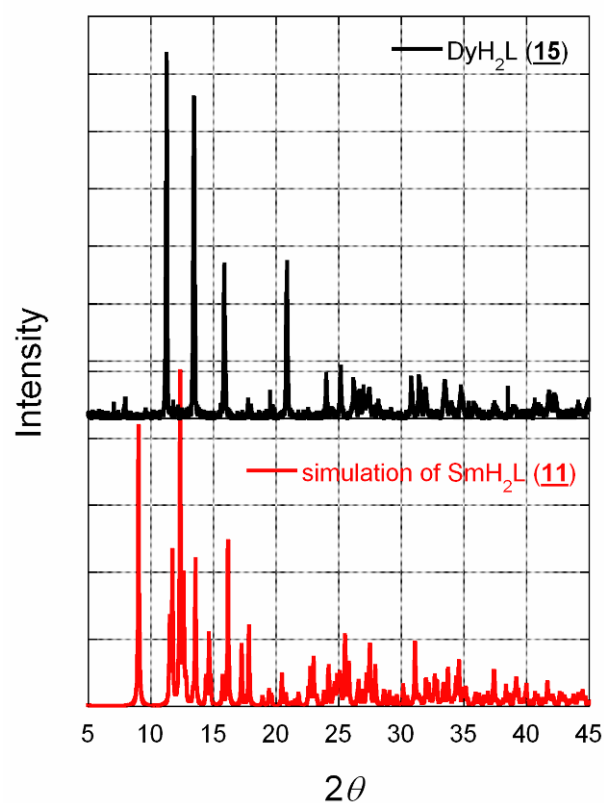


Fig. S8. X-ray powder diffraction pattern for DyH_2L **15**. Red line is the simulated X-ray powder diffraction patterns obtained from the single-crystal structural data of SmH_2L **11**.

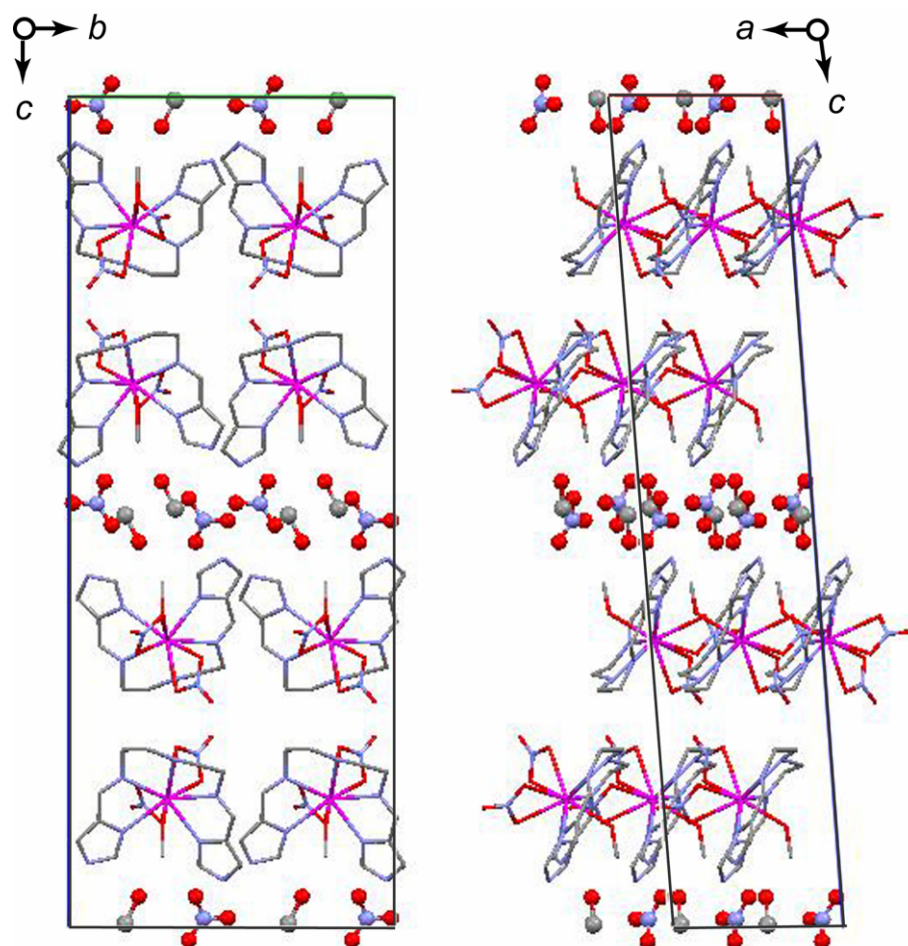


Fig. S9. Crystal packing diagram of Sm·11 (a) projected along the *a*-axis (left) and *b*-axis (right) (hydrogen atoms omitted for clarity).

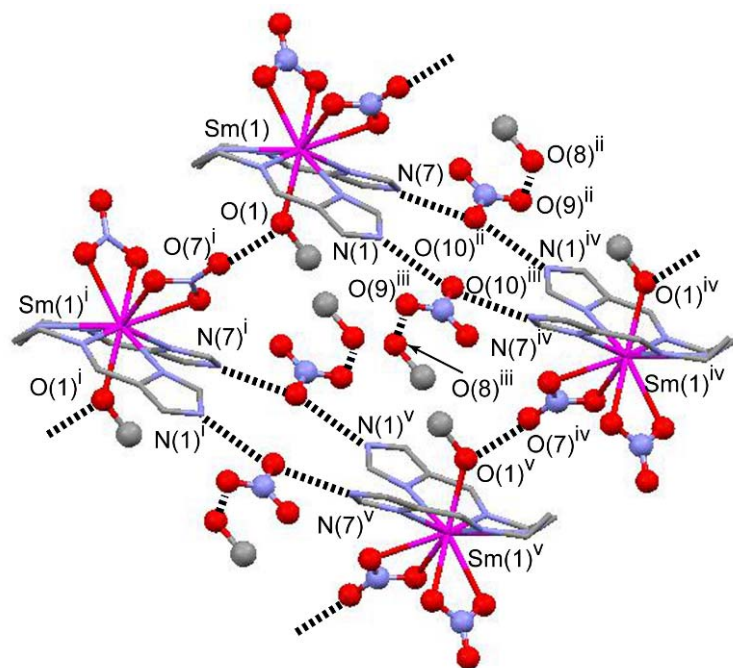


Fig. S10. Crystal structure of Sm·**11**, showing a network structure connected by hydrogen bonds through crystalline MeOH and counter anion, NO_3^- . Symmetry operations: i ($1+x, y, z$), ii ($-x, -y, -z$), iii ($-0.5+x, 0.5+y, z$), iv ($-0.5-x, 0.5-y, -z$), v ($0.5-x, 0.5-y, -z$).

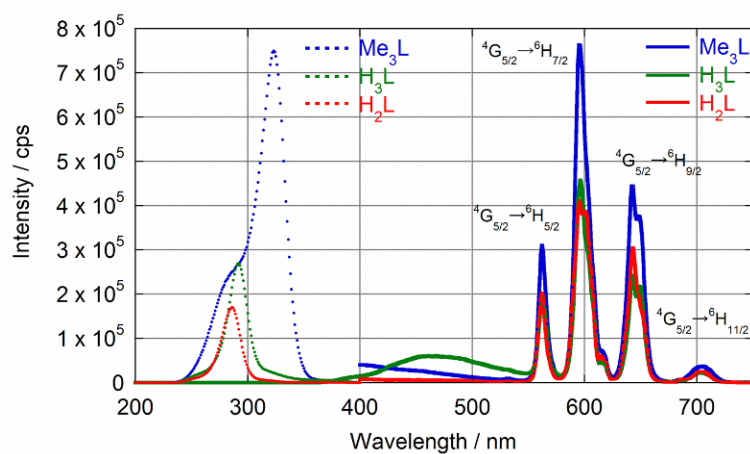


Fig. S11. Excitation (200–400 nm, dotted line) and emission spectra (400–750 nm, solid line) of Sm^{3+} complexes, **1**, **6**, and **11** in the methanol solution (1.0×10^{-4} M) at 298 K. Excitation spectra were obtained by monitoring emission at 597 nm ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$). Emission spectra were measured with excitation at 350, 286, and 286 nm for **1**, **6**, and **11**, respectively.

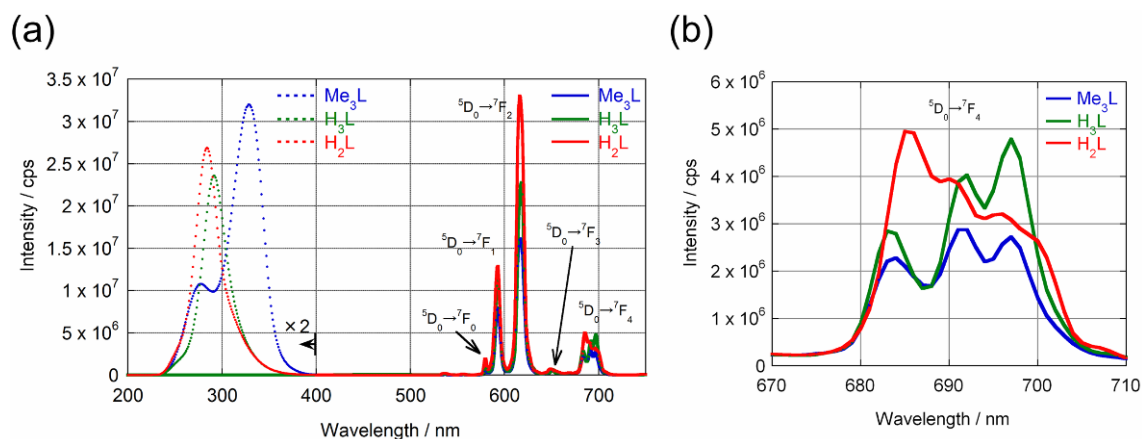


Fig. S12. (a) Excitation (200–400 nm, dotted line) and emission spectra (400–750 nm, solid line) of Eu³⁺ complexes of **2**, **7**, and **12** in the methanol solution (1.0×10^{-4} M) at 298 K. Excitation spectra were obtained by monitoring emission at 617 nm (⁵D₀ → ⁷F₂). The emission spectra were measured with excitation at 329, 291, and 284 nm for **2**, **7**, and **12**, respectively, (b) the ⁵D₀ → ⁷F₄ transition of Eu³⁺ ion.

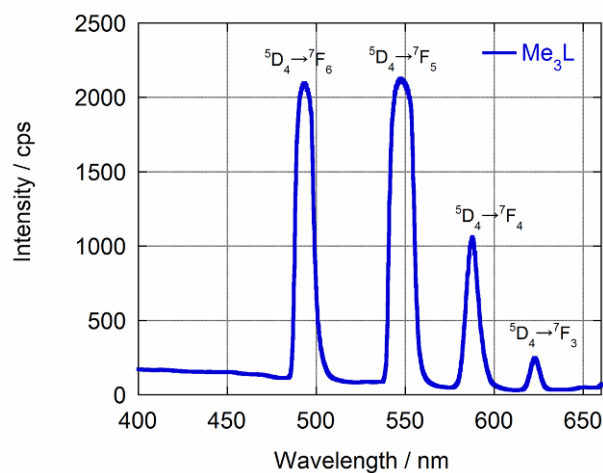


Fig. S13. Emission spectra of Tb³⁺ complex **4** in the solid state at 77 K. The samples were excited at 350 nm. Ligand phosphorescence was cut off by a mechanical light chopper.

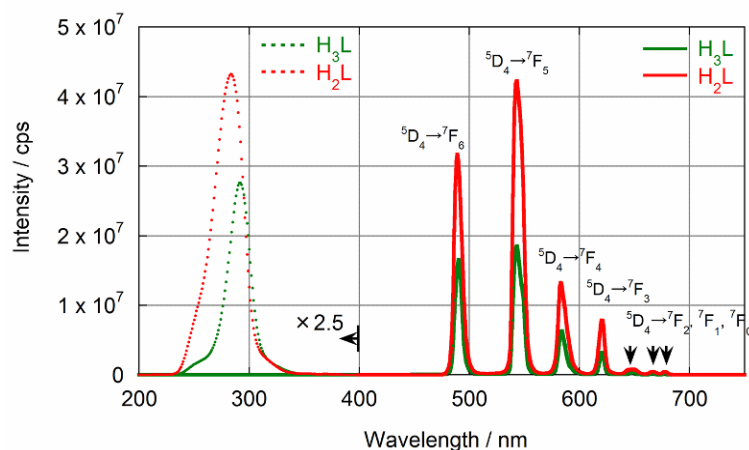


Fig. S14. Excitation (200–400 nm, dotted line) and emission spectra (400–750 nm, solid line) of Tb^{3+} complexes **9** and **14** in methanol solution (1.0×10^{-4} M) at 298 K. Excitation spectra were obtained by monitoring the emission wavelength at 543 nm ($^5\text{D}_4 \rightarrow ^7\text{F}_5$). The emission spectra were measured with sample excitation at 292 and 284 nm for **9** and **14**, respectively.

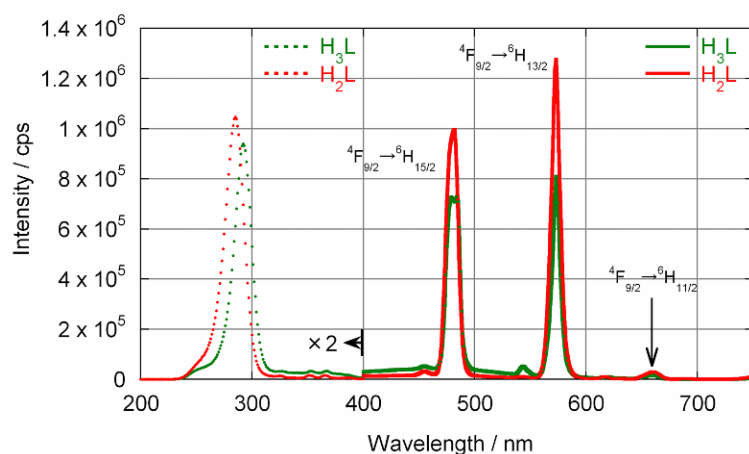


Fig. S15. Excitation (200–400 nm, dotted line) and emission spectra (400–750 nm, solid line) of Dy^{3+} complexes **10** and **15** in methanol solution (1.0×10^{-4} M) at 298 K. Excitation spectra were obtained by monitoring the emission wavelength at 574 nm ($^4\text{F}_{9/2} \rightarrow ^6\text{H}_{13/2}$). The emission spectra were measured while the samples were excited at 292 and 285 nm for **10** and **15**, respectively.

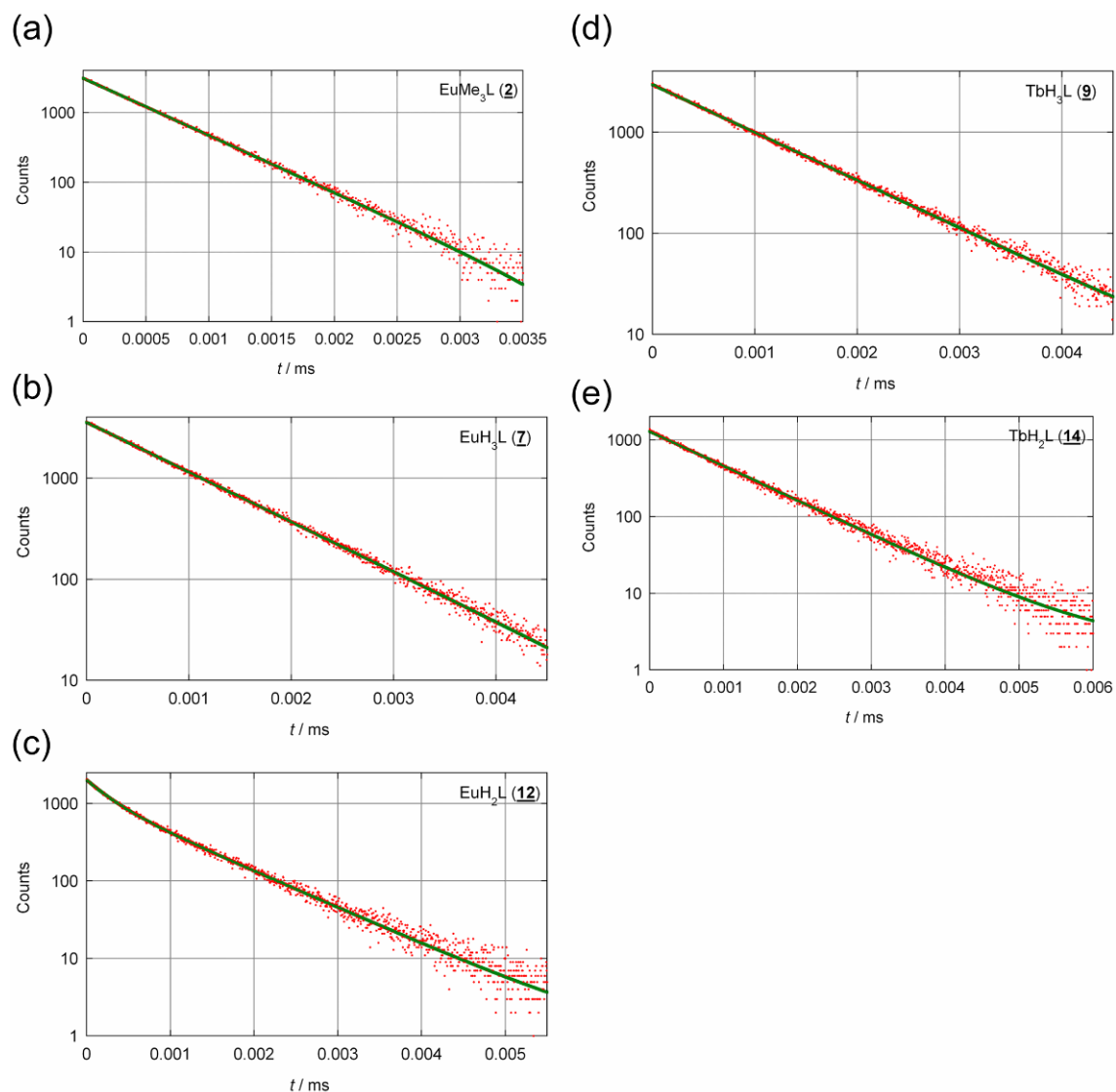


Fig. S16. Luminescence decay curves for Eu^{3+} complexes of (a) **2**, (b) **7**, and (c) **12**, and Tb^{3+} complexes of (d) **9** and (e) **14** in the solid state at 298 K. All data, except for **12**, were fitted to an exponential decay with one component. Two-component analysis was adopted in complex **12** to yield times of 272 μs (minor component $\sim 17\%$) and 916 μs (major component $\sim 83\%$).

Table S1. Crystal data and structure refinements for **1–5**.

Compound	Sm· 1	Eu· 2	Gd· 3	Tb· 4	Dy· 5
Empirical formula	C ₂₁ H ₄₀ N ₁₃ O ₁₄ Sm	C ₂₁ H ₄₀ EuN ₁₃ O ₁₄	C ₂₁ H ₄₀ GdN ₁₃ O ₁₄	C ₂₁ H ₄₀ N ₁₃ O ₁₄ Tb	C ₂₁ H ₄₀ DyN ₁₃ O ₁₄
<i>M</i> / g mol ⁻¹	849.01	850.62	855.91	857.58	861.16
<i>T</i> / K	100	100	93	90	90
Crystal system	Monoclinic	Monoclinic	Monoclinic	Monoclinic	Monoclinic
Space group	<i>P</i> 2 ₁ /n	<i>P</i> 2 ₁ /n	<i>P</i> 2 ₁ /n	<i>P</i> 2 ₁ /n	<i>P</i> 2 ₁ /n
<i>a</i> / Å	14.5310(9)	14.540(6)	10.1450(10)	14.4831(7)	14.4882(7)
<i>b</i> / Å	15.3647(10)	15.354(6)	21.166(2)	15.3207(7)	15.3415(7)
<i>c</i> / Å	15.0069(10)	14.989(6)	16.3695(17)	14.9738(7)	14.9839(7)
<i>α</i> / °	90	90	90	90	90
<i>β</i> / °	93.6430(10)	93.250(9)	106.850(2)	93.3460(10)	93.2490(10)
<i>γ</i> / °	90	90	90	90	90
<i>V</i> / Å ³	3343.7(4)	3341(2)	3364.1(6)	3316.9(3)	3325.1(3)
<i>Z</i>	4	4	4	4	4
<i>D</i> _{calcd} / g cm ⁻³	1.687	1.691	1.690	1.717	1.72
Reflections collected	23460	16441	24121	24004	24461
Independent reflections (<i>R</i> _{int})	8306 (0.0225)	8119 (0.0531)	9473 (0.0385)	8130 (0.0274)	8276 (0.0269)
Goodness of fit	1.067	1.174	1.043	1.343	1.210
^a <i>R</i> 1 (<i>I</i> > 2σ (all data))	0.0243 (0.0293)	0.0718 (0.0913)	0.0378 (0.0487)	0.0513 (0.1019)	0.0409 (0.0436)
^b <i>wR</i> 2 (<i>I</i> > 2σ (all data))	0.0581 (0.0603)	0.1755 (0.1889)	0.1028 (0.1074)	0.0533 (0.1027)	0.0859 (0.0871)
Least diff. peak (hole) / e Å ⁻³	1.470 (−0.386)	3.198 (−2.485)	2.452 (−1.394)	1.551 (−1.70)	1.823 (−1.063)

$$^a R1 = \sum ||F_o| - |F_c|| / \sum |F_o|, \quad ^b wR2 = \{ \sum [w(F_o^2 - F_c^2)^2] / \sum [w(F_o^2)^2] \}^{1/2}$$

Table S2. Crystal data and structure refinements for **6–10**.

Compound	Sm· 6	Eu· 7	Gd· 8	Tb· 9	Dy· 10
Empirical formula	C ₁₉ H ₂₈ N ₁₃ O ₁₀ Sm	C ₁₉ H ₂₈ EuN ₁₃ O ₁₀	C ₁₉ H ₂₈ GdN ₁₃ O ₂₀	C ₁₉ H ₂₈ N ₁₃ O ₁₀ Tb	C ₁₉ H ₂₈ Dy ₂₁₃ O ₁₀
<i>M</i> / g mol ⁻¹	1497.79	1501.01	1511.59	1514.93	1522.09
<i>T</i> / K	90	90	90	90	90
Crystal system	Triclinic	Triclinic	Triclinic	Triclinic	Triclinic
Space group	<i>P</i> -1	<i>P</i> -1	<i>P</i> -1	<i>P</i> -1	<i>P</i> -1
<i>a</i> / Å	11.0955(13)	11.126(4)	11.145(4)	11.172(3)	11.205(3)
<i>b</i> / Å	16.3259(19)	16.104(6)	16.089(7)	16.142(5)	16.115(5)
<i>c</i> / Å	17.628(2)	17.585(7)	17.548(7)	17.528(5)	17.472(5)
α / °	103.252(2)	102.599(6)	102.336(6)	101.836(5)	100.834(5)
β / °	107.393(2)	107.322(6)	107.675(6)	108.106(4)	108.384(4)
γ / °	103.955(2)	104.235(6)	104.145(6)	104.166(5)	104.729(5)
<i>V</i> / Å ³	2796.4(6)	2767.4(19)	2762.4(19)	2774.1(14)	2769.1(13)
<i>Z</i>	4	4	4	4	4
<i>D</i> _{calcd} / g cm ⁻³	1.779	1.801	1.817	1.814	1.825
Reflections collected	18351	10528	12358	14266	10769
Independent reflections (<i>R</i> _{int})	13501 (0.0402)	6529 (0.0448)	8075 (0.0577)	9779 (0.0424)	6577 (0.0542)
Goodness of fit	1.015	1.059	1.117	1.090	1.057
^a <i>R</i> 1 (<i>I</i> > 2σ (all data))	0.0901 (0.1131)	0.0769 (0.1034)	0.0929 (0.1298)	0.0760 (0.1073)	0.0677 (0.0949)
^b w <i>R</i> 2 (<i>I</i> > 2σ (all data))	0.2449 (0.2640)	0.1876 (0.2068)	0.1798 (0.1954)	0.1464 (0.1579)	0.1598 (0.1789)
Least diff. peak (hole) / e Å ⁻³	8.691 (-9.665)	7.737 (-1.449)	3.016 (-2.264)	4.694 (-2.021)	3.846 (-1.857)

^a *R*1 = $\Sigma ||F_o| - |F_c|| / \Sigma |F_o|$. ^b w*R*2 = $\{ \Sigma [w(F_o^2 - F_c^2)^2] / \Sigma [w(F_o^2)^2] \}^{1/2}$

Table S3. Crystal data and structure refinements for **11–14**.

Compound	Sm· 11	Eu· 12	Gd· 13	Tb· 14
Empirical formula	C ₁₄ H ₂₅ N ₁₀ O ₁₁ Sm	C ₁₄ H ₂₅ EuN ₁₀ O ₁₁	C ₁₄ H ₂₅ GdN ₁₀ O ₁₁	C ₁₄ H ₂₅ N ₁₀ O ₁₁ Tb
<i>M</i> / g mol ^{−1}	659.79	661.40	666.69	668.36
<i>T</i> / K	90	90	90	90
Crystal system	Monoclinic	Monoclinic	Monoclinic	Monoclinic
Space group	<i>C</i> 2/ <i>c</i>	<i>C</i> 2/ <i>c</i>	<i>C</i> 2/ <i>c</i>	<i>C</i> 2/ <i>c</i>
<i>a</i> / Å	8.1731(15)	8.1695(14)	8.1523(10)	8.1574(19)
<i>b</i> / Å	15.354(3)	15.333(3)	15.2716(17)	15.206(4)
<i>c</i> / Å	39.204(7)	39.165(7)	39.174(4)	39.097(9)
α / °	90	90	90	90
β / °	94.183(3)	94.318(3)	94.396(2)	94.717(3)
γ / °	90	90	90	90
<i>V</i> / Å ³	4906.5(15)	4892.0(14)	4862.8(10)	4833(2)
<i>Z</i>	8	8	8	8
<i>D</i> _{calcd} / g cm ^{−3}	1.786	1.796	1.821	1.837
Reflections collected	16361	16325	15564	15928
Independent reflections (<i>R</i> _{int})	6096 (0.0398)	6074 (0.0445)	5673 (0.0554)	5963 (0.0383)
Goodness of fit	1.045	1.032	1.037	1.109
^a <i>R</i> 1 (<i>I</i> > 2σ (all data))	0.0335 (0.0440)	0.0341 (0.0444)	0.0361 (0.0547)	0.0386 (0.0464)
^b w <i>R</i> 2 (<i>I</i> > 2σ (all data))	0.0605 (0.0638)	0.0696 (0.0737)	0.0602 (0.0765)	0.0750 (0.0776)
Least diff. peak (hole) / e Å ^{−3}	1.109 (−1.147)	1.264 (−1.213)	0.726 (−0.765)	1.540 (−2.191)

$$^a R1 = \sum ||F_o| - |F_c|| / \sum |F_o|, \quad ^b wR2 = \{ \sum [w(F_o^2 - F_c^2)^2] / \sum [w(F_o^2)^2] \}^{1/2}$$

Table S4. Selected bond distances (Å) and angles (deg) with estimated standard deviations in parentheses for **1–5**.

Sm·1		Eu·2		Gd·3		Tb·4		Dy·5	
Sm(1)–N(1)	2.6831(18)	Eu(1)–N(1)	2.684(7)	Gd(1)–N(1)	2.661(3)	Tb(1)–N(1)	2.655(4)	Dy(1)–N(1)	2.651(3)
Sm(1)–N(2)	2.5021(18)	Eu(1)–N(2)	2.499(6)	Gd(1)–N(2)	2.509(3)	Tb(1)–N(2)	2.470(4)	Dy(1)–N(2)	2.461(3)
Sm(1)–N(3)	2.6290(17)	Eu(1)–N(3)	2.623(6)	Gd(1)–N(3)	2.581(3)	Tb(1)–N(3)	2.608(4)	Dy(1)–N(3)	2.609(3)
Sm(1)–N(5)	2.6143(17)	Eu(1)–N(5)	2.611(6)	Gd(1)–N(5)	2.593(3)	Tb(1)–N(5)	2.573(4)	Dy(1)–N(5)	2.569(3)
Sm(1)–N(6)	2.5489(17)	Eu(1)–N(6)	2.539(7)	Gd(1)–N(6)	2.535(3)	Tb(1)–N(6)	2.504(4)	Dy(1)–N(6)	2.497(3)
Sm(1)–N(8)	2.5420(17)	Eu(1)–N(8)	2.530(6)	Gd(1)–N(8)	2.513(3)	Tb(1)–N(8)	2.509(4)	Dy(1)–N(8)	2.501(3)
Sm(1)–N(9)	2.5630(17)	Eu(1)–N(9)	2.552(6)	Gd(1)–N(9)	2.545(3)	Tb(1)–N(9)	2.522(4)	Dy(1)–N(9)	2.517(3)
Sm(1)–O(1)	2.4596(16)	Eu(1)–O(1)	2.448(6)	Gd(1)–O(1)	2.445(3)	Tb(1)–O(1)	2.425(3)	Dy(1)–O(1)	2.403(3)
Sm(1)–O(2)	2.4539(16)	Eu(1)–O(2)	2.448(6)	Gd(1)–O(2)	2.439(3)	Tb(1)–O(2)	2.430(3)	Dy(1)–O(2)	2.409(3)
Sm(1)···Sm(1) ^a	8.6593(5)	Eu(1)···Eu(1) ^a	8.643(3)	Gd(1)···Gd(1) ^a	8.6195(7)	Tb(1)···Tb(1) ^a	8.6153(4)	Dy(1)···Dy(1) ^a	8.6216(4)
O(2)–Sm(1)–O(1)	139.87(6)	O(1)–Eu(1)–O(2)	139.8(2)	O(2)–Gd(1)–O(1)	137.36(10)	O(1)–Tb(1)–O(2)	139.96(11)	O(2)–Dy(1)–O(1)	139.94(11)
O(1)–Sm(1)–N(1)	75.82(5)	O(1)–Eu(1)–N(1)	76.08(19)	O(1)–Gd(1)–N(1)	94.64(10)	O(1)–Tb(1)–N(1)	75.91(12)	O(1)–Dy(1)–N(1)	75.98(10)
O(2)–Sm(1)–N(1)	96.90(6)	O(2)–Eu(1)–N(1)	96.8(2)	O(2)–Gd(1)–N(1)	79.78(9)	O(2)–Tb(1)–N(1)	96.80(12)	O(2)–Dy(1)–N(1)	96.85(10)
N(2)–Sm(1)–N(1)	67.07(6)	N(2)–Eu(1)–N(1)	67.4(2)	N(2)–Gd(1)–N(1)	66.99(10)	N(2)–Tb(1)–N(1)	67.78(12)	N(2)–Dy(1)–N(1)	67.85(10)
N(3)–Sm(1)–N(1)	128.39(5)	N(3)–Eu(1)–N(1)	128.80(19)	N(3)–Gd(1)–N(1)	128.93(10)	N(3)–Tb(1)–N(1)	129.12(12)	N(3)–Dy(1)–N(1)	129.31(9)
N(5)–Sm(1)–N(1)	63.71(5)	N(5)–Eu(1)–N(1)	63.6(2)	N(5)–Gd(1)–N(1)	64.09(9)	N(5)–Tb(1)–N(1)	64.01(12)	N(5)–Dy(1)–N(1)	64.01(10)
N(6)–Sm(1)–N(1)	127.47(5)	N(6)–Eu(1)–N(1)	127.72(19)	N(6)–Gd(1)–N(1)	128.56(9)	N(6)–Tb(1)–N(1)	128.29(13)	N(6)–Dy(1)–N(1)	128.61(10)
N(8)–Sm(1)–N(1)	66.12(5)	N(8)–Eu(1)–N(1)	66.24(19)	N(8)–Gd(1)–N(1)	66.22(9)	N(8)–Tb(1)–N(1)	66.28(13)	N(8)–Dy(1)–N(1)	66.49(10)
N(9)–Sm(1)–N(1)	130.70(5)	N(9)–Eu(1)–N(1)	131.4(2)	N(9)–Gd(1)–N(1)	132.70(9)	N(9)–Tb(1)–N(1)	131.80(12)	N(9)–Dy(1)–N(1)	132.17(10)

^a Symmetry operations: 1.5–x, 0.5+y, 1.5–z. for **1**, **2**, **4**, and **5**, 0.5+x, 1.5–y, 0.5+z, for **3**.

Table S5. Hydrogen-bond distances (Å) less than 3.0 Å with estimated standard deviations in parentheses for **1–5**.

	Sm· 1	Eu· 2	Tb· 4	Dy· 5		Gd· 3
O(1)···O(4)	2.780(2) ^{#1}	2.786(8) ^{#1}	2.770(4) ^{#1}	2.784(4) ^{#1}	O(1)···O(5) ^{#6}	2.766(5)
O(1)···O(11)	2.819(2)	2.812(8)	2.814(5)	2.820(4)	O(1)···O(6) ^{#6}	2.866(7)
O(2)···O(9)	2.851(2) ^{#2}	2.849(8) ^{#2}	2.849(5) ^{#2}	2.862(4) ^{#2}	O(2)···O(9) ^{#7}	2.774(4)
O(2)···O(14)	2.623(2)	2.631(9)	2.627(5)	2.623(4)	O(2)···O(13)	2.764(5)
O(3)···O(13)	2.786(2) ^{#3}	2.78(1) ^{#3}	2.782(6) ^{#3}	2.787(4) ^{#3}	O(4)···O(12)	2.888(6)
O(5)···O(13)	2.831(2) ^{#4}	2.83(1) ^{#4}	2.830(6) ^{#4}	2.836(5) ^{#4}	O(8)···O(13)	2.897(6)
O(7)···O(12)	2.837(2)	2.84(1)	2.840(6)	2.843(5)	O(10)···O(12) ^{#8}	2.852(5)
O(8)···O(14)	2.786(2)	2.785(9)	2.753(6)	2.767(5)	O(13)···O(14) ^{#9}	2.75(1)
O(12)···O(13)	2.765(3)	2.74(1)	2.758(6)	2.761(5)		
O(12)···O(14)	2.687(2) ^{#5}	2.68(1) ^{#5}	2.679(6) ^{#5}	2.684(5) ^{#5}		

Symmetry operations: #1 (1.5−x, 0.5+y, 1.5−z), #2 (−0.5+x, 0.5−y, 0.5+z), #3 (0.5+x, 0.5−y, 0.5+z), #4 (1.5−x, −0.5+y, 1.5−z), #5 (0.5−x, 0.5+y, 1.5−z), #6 (0.5−x, 0.5+y, 0.5−z), #7 (x, y, −1+z), #8 (0.5+x, 1.5−y, 0.5+z), #9 (−1+x, y, z).

Table S6. Selected bond distances (Å) and angles (deg) with estimated standard deviations in parentheses for **6** and **7**.

Sm· 6				Eu· 7			
Sm(1)–N(1)	2.739(7)	Sm(2)–N(12)	2.693(7)	Eu(1)–N(1)	2.712(12)	Eu(2)–N(12)	2.659(12)
Sm(1)–N(2)	2.549(8)	Sm(2)–N(13)	2.489(7)	Eu(1)–N(2)	2.507(15)	Eu(2)–N(13)	2.486(13)
Sm(1)–N(3)	2.565(8)	Sm(2)–N(14)	2.543(8)	Eu(1)–N(3)	2.556(13)	Eu(2)–N(14)	2.503(13)
Sm(1)–N(5)	2.573(8)	Sm(2)–N(16)	2.543(7)	Eu(1)–N(5)	2.510(9)	Eu(2)–N(16)	2.517(12)
Sm(1)–N(6)	2.567(8)	Sm(2)–N(17)	2.536(7)	Eu(1)–N(6)	2.575(12)	Eu(2)–N(17)	2.515(12)
Sm(1)–N(8)	2.525(7)	Sm(2)–N(19)	2.544(7)	Eu(1)–N(8)	2.496(13)	Eu(2)–N(19)	2.498(12)
Sm(1)–N(9)	2.556(8)	Sm(2)–N(20)	2.586(9)	Eu(1)–N(9)	2.533(12)	Eu(2)–N(20)	2.533(13)
Sm(1)–O(1)	2.481(6)	Sm(2)–O(4)	2.496(6)	Eu(1)–O(1)	2.480(10)	Eu(2)–O(4)	2.470(10)
Sm(1)–O(2)	2.481(6)	Sm(2)–O(5)	2.496(7)	Eu(1)–O(2)	2.457(10)	Eu(2)–O(5)	2.495(10)
Sm(1)···Sm (2) ^a	7.7006(9)			Eu(1)···Eu(2) ^a	7.635(2)		
O(1)–Sm(1)–N(1)	120.3(2)	O(4)–Sm(2)–N(12)	119.2(2)	O(1)–Eu(1)–N(1)	119.8(4)	O(4)–Eu(2)–N(12)	118.5(3)
O(2)–Sm(1)–N(1)	78.8(2)	O(5)–Sm(2)–N(12)	78.4(2)	O(2)–Eu(1)–N(1)	78.2(4)	O(5)–Eu(2)–N(12)	77.8(4)
N(2)–Sm(1)–N(1)	66.1(3)	N(13)–Sm(2)–N(12)	67.0(2)	N(2)–Eu(1)–N(1)	66.0(5)	N(13)–Eu(2)–N(12)	67.9(4)
N(3)–Sm(1)–N(1)	127.3(2)	N(14)–Sm(2)–N(12)	129.2(2)	N(3)–Eu(1)–N(1)	126.7(4)	N(14)–Eu(2)–N(12)	129.4(4)
N(5)–Sm(1)–N(1)	62.8(2)	N(16)–Sm(2)–N(12)	63.9(2)	N(5)–Eu(1)–N(1)	64.3(4)	N(16)–Eu(2)–N(12)	63.5(4)
N(6)–Sm(1)–N(1)	121.8(2)	N(17)–Sm(2)–N(12)	123.6(2)	N(6)–Eu(1)–N(1)	122.4(4)	N(17)–Eu(2)–N(12)	123.5(4)
N(8)–Sm(1)–N(1)	65.3(2)	N(19)–Sm(2)–N(12)	65.3(2)	N(8)–Eu(1)–N(1)	65.6(4)	N(19)–Eu(2)–N(12)	65.8(4)
N(9)–Sm(1)–N(1)	119.8(2)	N(20)–Sm(2)–N(12)	120.5(2)	N(9)–Eu(1)–N(1)	120.2(4)	N(20)–Eu(2)–N(12)	121.5(4)

^a Symmetry operations: $x, y, -1+z$.

Table S7. Selected bond distances (Å) and angles (deg) with estimated standard deviations in parentheses for **8** and **9**.

Gd· 8				Tb· 9			
Gd(1)–N(1)	2.709(15)	Gd(2)–N(12)	2.649(14)	Tb(1)–N(1)	2.703(10)	Tb(2)–N(12)	2.659(10)
Gd(1)–N(2)	2.530(13)	Gd(2)–N(13)	2.490(15)	Tb(1)–N(2)	2.500(9)	Tb(2)–N(13)	2.452(10)
Gd(1)–N(3)	2.551(15)	Gd(2)–N(14)	2.496(15)	Tb(1)–N(3)	2.545(11)	Tb(2)–N(14)	2.494(10)
Gd(1)–N(5)	2.471(19)	Gd(2)–N(16)	2.504(12)	Tb(1)–N(5)	2.508(12)	Tb(2)–N(16)	2.5519(9)
Gd(1)–N(6)	2.535(14)	Gd(2)–N(17)	2.516(12)	Tb(1)–N(6)	2.514(10)	Tb(2)–N(17)	2.496(10)
Gd(1)–N(8)	2.472(14)	Gd(2)–N(19)	2.511(13)	Tb(1)–N(8)	2.475(10)	Tb(2)–N(19)	2.486(9)
Gd(1)–N(9)	2.517(13)	Gd(2)–N(20)	2.544(13)	Tb(1)–N(9)	2.517(9)	Tb(2)–N(20)	2.529(9)
Gd(1)–O(1)	2.443(10)	Gd(2)–O(4)	2.474(11)	Tb(1)–O(1)	2.448(8)	Tb(2)–O(4)	2.471(8)
Gd(1)–O(2)	2.441(11)	Gd(2)–O(5)	2.461(12)	Tb(1)–O(2)	2.433(8)	Tb(2)–O(5)	2.460(8)
Gd(1)···Gd(2) ^a	7.614(2)			Tb(1)···Tb(2) ^a	7.598(1)		
O(1)–Gd(1)–N(1)	120.5(4)	O(4)–Gd(2)–N(12)	119.0(4)	O(1)–Tb(1)–N(1)	120.0(3)	O(4)–Tb(2)–N(12)	119.2(3)
O(2)–Gd(1)–N(1)	77.9(4)	O(5)–Gd(2)–N(12)	78.3(4)	O(2)–Tb(1)–N(1)	78.0(3)	O(5)–Tb(2)–N(12)	78.1(3)
N(2)–Gd(1)–N(1)	65.8(4)	N(13)–Gd(2)–N(12)	67.6(5)	N(2)–Tb(1)–N(1)	65.9(3)	N(13)–Tb(2)–N(12)	67.8(3)
N(3)–Gd(1)–N(1)	127.1(4)	N(14)–Gd(2)–N(12)	129.4(5)	N(3)–Tb(1)–N(1)	127.7(3)	N(14)–Tb(2)–N(12)	130.0(3)
N(5)–Gd(1)–N(1)	64.1(5)	N(16)–Gd(2)–N(12)	64.1(4)	N(5)–Tb(1)–N(1)	64.4(4)	N(16)–Tb(2)–N(12)	64.4(3)
N(6)–Gd(1)–N(1)	123.2(4)	N(17)–Gd(2)–N(12)	123.6(5)	N(6)–Tb(1)–N(1)	123.6(3)	N(17)–Tb(2)–N(12)	124.6(3)
N(8)–Gd(1)–N(1)	65.5(4)	N(19)–Gd(2)–N(12)	65.4(4)	N(8)–Tb(1)–N(1)	65.8(3)	N(19)–Tb(2)–N(12)	65.5(3)
N(9)–Gd(1)–N(1)	120.1(4)	N(20)–Gd(2)–N(12)	121.5(4)	N(9)–Tb(1)–N(1)	120.8(3)	N(20)–Tb(2)–N(12)	121.6(3)

^a Symmetry operations: $x, y, -1+z$.

Table S8. Selected bond distances (Å) and angles (deg) with estimated standard deviations in parentheses for **10**.

Dy· 10			
Dy(1)–N(1)	2.688(13)	Dy(2)–N(12)	2.641(13)
Dy(1)–N(2)	2.473(12)	Dy(2)–N(13)	2.453(12)
Dy(1)–N(3)	2.527(12)	Dy(2)–N(14)	2.465(13)
Dy(1)–N(5)	2.509(12)	Dy(2)–N(16)	2.507(12)
Dy(1)–N(6)	2.486(12)	Dy(2)–N(17)	2.470(12)
Dy(1)–N(8)	2.481(12)	Dy(2)–N(19)	2.463(12)
Dy(1)–N(9)	2.517(11)	Dy(2)–N(20)	2.507(13)
Dy(1)–O(1)	2.454(10)	Dy(2)–O(4)	2.443(10)
Dy(1)–O(2)	2.419(9)	Dy(2)–O(5)	2.444(9)
Dy(1)···Dy(2) ^a	7.571(1)		
O(1)–Dy(1)–N(1)	119.5(4)	O(4)–Dy(2)–N(12)	119.5(5)
O(2)–Dy(1)–N(1)	77.8(4)	O(5)–Dy(2)–N(12)	77.7(4)
N(2)–Dy(1)–N(1)	66.4(4)	N(13)–Dy(2)–N(12)	68.0(4)
N(3)–Dy(1)–N(1)	128.2(4)	N(14)–Dy(2)–N(12)	129.8(4)
N(5)–Dy(1)–N(1)	63.8(4)	N(16)–Dy(2)–N(12)	63.5(4)
N(6)–Dy(1)–N(1)	122.3(4)	N(17)–Dy(2)–N(12)	125.1(4)
N(8)–Dy(1)–N(1)	65.5(4)	N(19)–Dy(2)–N(12)	65.8(4)
N(9)–Dy(1)–N(1)	121.7(4)	N(20)–Dy(2)–N(12)	122.2(4)

^a Symmetry operations: $x, y, -1+z$.

Table S9. Hydrogen–bond distances (Å) less than 3.0 Å with estimated standard deviations in parentheses for **6–10**.

	Sm· 6	Eu· 7	Gd· 8	Tb· 9	Dy· 10
N(4)···O(12)	2.92(1) ^{#1}	2.88(3) ^{#1}	2.90(2) ^{#1}	2.93(1) ^{#1}	2.91(2) ^{#1}
N(7)···O(7)	2.91(1) ^{#2}	2.94(1) ^{#2}	2.89(2) ^{#2}	2.90(1) ^{#2}	2.79(1) ^{#2}
N(10)···O(13)	2.85(1)	2.87(1)	2.87(2)	2.85(1)	2.83(1)
N(15)···O(11)	2.84(1) ^{#3}	2.96(3) ^{#3}	2.95(2) ^{#3}	2.88(2) ^{#3}	2.84(3) ^{#3}
N(18)···O(16)	2.76(1) ^{#3}	2.75(2) ^{#3}	2.73(2) ^{#3}	2.74(1) ^{#3}	2.71(1) ^{#3}
N(21)···O(15)	2.73(1) ^{#3}	2.69(2) ^{#3}	2.71(2) ^{#3}	2.73(1) ^{#3}	2.76(2) ^{#3}

Symmetry operations: #1 (x, y, -1+z), #2 (1+x, y, z), #3 (x, -1+y, z).

Table S10. Selected bond distances (Å) and angles (deg) with estimated standard deviations in parentheses for **11–14**.

Sm· 11		Eu· 12		Gd· 13		Tb· 14	
Sm(1)–N(2)	2.593(3)	Eu(1)–N(2)	2.584(3)	Gd(1)–N(2)	2.575(4)	Tb(1)–N(2)	2.552(3)
Sm(1)–N(3)	2.533(3)	Eu(1)–N(3)	2.528(3)	Gd(1)–N(3)	2.507(4)	Tb(1)–N(3)	2.490(4)
Sm(1)–N(4)	2.698(3)	Eu(1)–N(4)	2.695(3)	Gd(1)–N(4)	2.677(4)	Tb(1)–N(4)	2.653(3)
Sm(1)–N(5)	2.594(3)	Eu(1)–N(5)	2.579(3)	Gd(1)–N(5)	2.569(4)	Tb(1)–N(5)	2.549(4)
Sm(1)–N(6)	2.548(3)	Eu(1)–N(6)	2.533(3)	Gd(1)–N(6)	2.522(4)	Tb(1)–N(6)	2.498(3)
Sm(1)–O(1)	2.466(2)	Eu(1)–O(1)	2.450(3)	Gd(1)–O(1)	2.434(4)	Tb(1)–O(1)	2.430(3)
Sm(1)–O(2)	2.508(2)	Eu(1)–O(2)	2.496(3)	Gd(1)–O(2)	2.481(4)	Tb(1)–O(2)	2.454(3)
Sm(1)–O(3)	2.622(2)	Eu(1)–O(3)	2.601(3)	Gd(1)–O(3)	2.591(3)	Tb(1)–O(3)	2.580(3)
Sm(1)–O(5)	2.554(2)	Eu(1)–O(5)	2.549(2)	Gd(1)–O(5)	2.532(3)	Tb(1)–O(5)	2.534(3)
Sm(1)–O(6)	2.657(2)	Eu(1)–O(6)	2.666(2)	Gd(1)–O(6)	2.672(4)	Tb(1)–O(6)	2.663(3)
Sm(1)···Sm (1) ^a	7.495(1)	Eu(1)···Eu(1) ^a	7.476(1)	Gd(1)···Gd(1) ^a	7.4696(8)	Tb(1)···Tb(1) ^a	7.4265(17)
O(1)–Sm(1)–N(4)	75.82(9)	O(1)–Eu(1)–N(4)	75.69(10)	O(1)–Gd(1)–N(4)	75.71(13)	O(1)–Tb(1)–N(4)	75.83(12)
O(2)–Sm(1)–N(4)	100.15(8)	O(2)–Eu(1)–N(4)	100.10(9)	O(2)–Gd(1)–N(4)	100.20(13)	O(2)–Tb(1)–N(4)	98.56(14)
O(3)–Sm(1)–N(4)	63.82(8)	O(3)–Eu(1)–N(4)	63.89(9)	O(3)–Gd(1)–N(4)	63.91(12)	O(3)–Tb(1)–N(4)	64.37(13)
O(5)–Sm(1)–N(4)	116.29(8)	O(5)–Eu(1)–N(4)	116.17(9)	O(5)–Gd(1)–N(4)	116.08(12)	O(5)–Tb(1)–N(4)	115.12(11)
O(6)–Sm(1)–N(4)	160.57(8)	O(6)–Eu(1)–N(4)	160.15(9)	O(6)–Gd(1)–N(4)	159.93(12)	O(6)–Tb(1)–N(4)	157.60(12)
N(2)–Sm(1)–N(4)	122.82(8)	N(2)–Eu(1)–N(4)	123.24(9)	N(2)–Gd(1)–N(4)	123.52(13)	N(2)–Tb(1)–N(4)	124.43(12)
N(3)–Sm(1)–N(4)	63.60(9)	N(3)–Eu(1)–N(4)	63.34(9)	N(3)–Gd(1)–N(4)	63.51(13)	N(3)–Tb(1)–N(4)	63.39(12)
N(5)–Sm(1)–N(4)	62.63(8)	N(5)–Eu(1)–N(4)	62.62(9)	N(5)–Gd(1)–N(4)	63.09(13)	N(5)–Tb(1)–N(4)	63.39(11)
N(6)–Sm(1)–N(4)	125.10(8)	N(6)–Eu(1)–N(4)	125.48(9)	N(6)–Gd(1)–N(4)	125.80(12)	N(6)–Tb(1)–N(4)	126.94(11)

^a Symmetry operations: $-x, y, 0.5-z$.

Table S11. Hydrogen–bond distances (Å) less than 3.0 Å with estimated standard deviations in parentheses for **11–14**.

	Sm· 11	Eu· 12	Gd· 13	Tb· 14
N(1)···O(10)	2.881(3) ^{#1}	2.877(3) ^{#1}	2.874(5) ^{#1}	2.865(4) ^{#1}
N(7)···O(10)	2.841(3) ^{#2}	2.834(3) ^{#2}	2.832(4) ^{#2}	2.824(4) ^{#2}
O(1)···O(7)	2.732(3) ^{#3}	2.733(3) ^{#3}	2.730(5) ^{#3}	2.710(4) ^{#3}
O(2)···O(6)	2.803(3)	2.793(3)	2.780(4)	2.765(4)
O(3)···O(5)	2.858(3)	2.834(3)	2.810(4)	2.766(5)
O(8)···O(9)	2.796(3)	2.799(3)	2.796(4) ^{#3}	2.778(4)

Symmetry operations: #1 (−0.5+x, 0.5+y, z), #2 (−x, −y, −z), #3 (1+x, y, z).