

Supporting Information

A family of doped lanthanide metal-organic frameworks for wide-range temperature sensing and tunable white light emission

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Table S1. The original ratios of different lanthanide metal salts and the corresponding ICP results.

Table S2. The parameters containing composition, temperature ranges (K), maximum relative sensitivity values (S_m , % K⁻¹), the temperature at which S_m is maximum (T_m , K) of several selected ratiometric luminescent MOF thermometers.

Table S3. Crystal data and refinement results for **EuL**, **GdL** and **TbL**.

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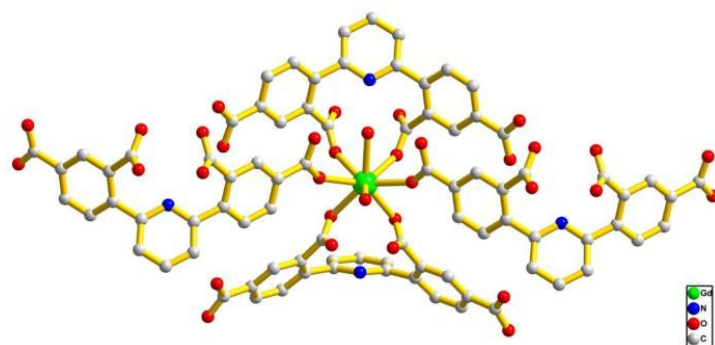


Fig. S1 The coordinated environment of Gd^{3+} ion in compound **GdL**.

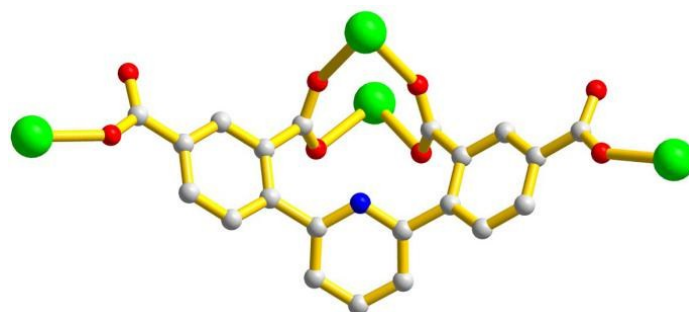


Fig. S2 The coordination mode of $[\text{L}]^{4-}$ in **GdL**: μ_4 -bridging mode.

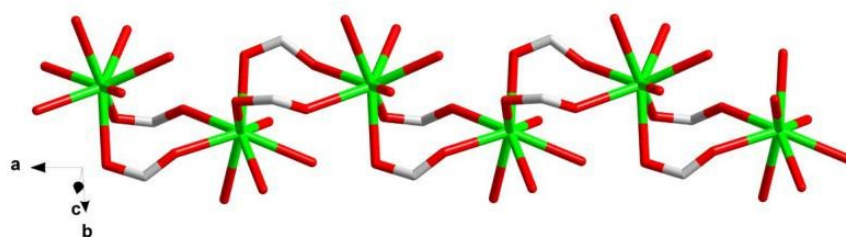


Fig. S3 1D metal-chain $[\text{Gd}_2(\mu_2\text{-COO})_2]_n$ in **GdL**.

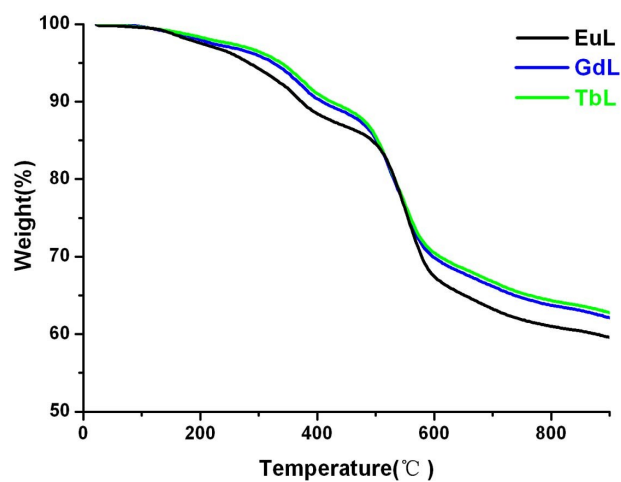


Fig. S4 TGA curve of **EuL**, **GdL** and **TbL**.

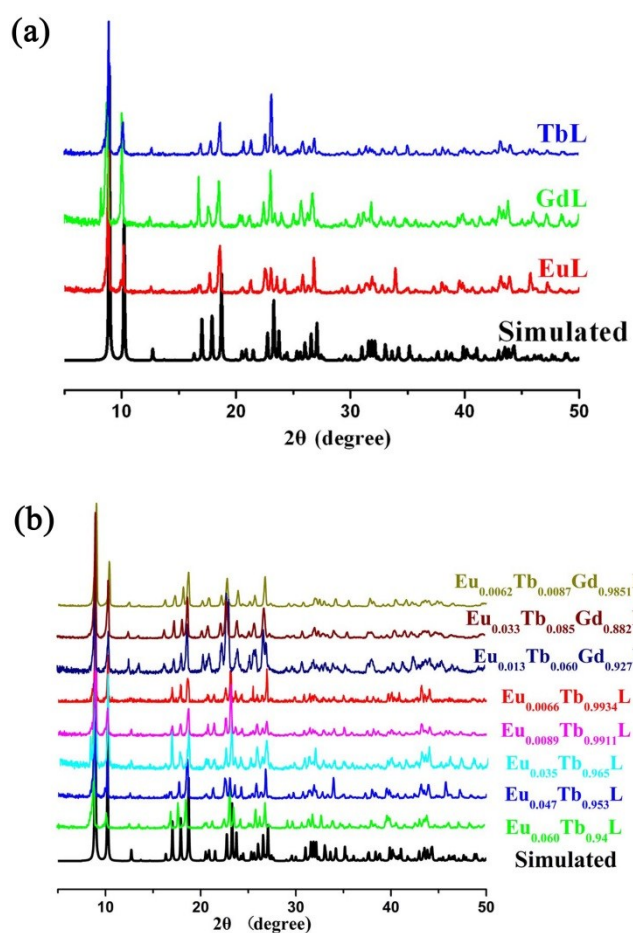


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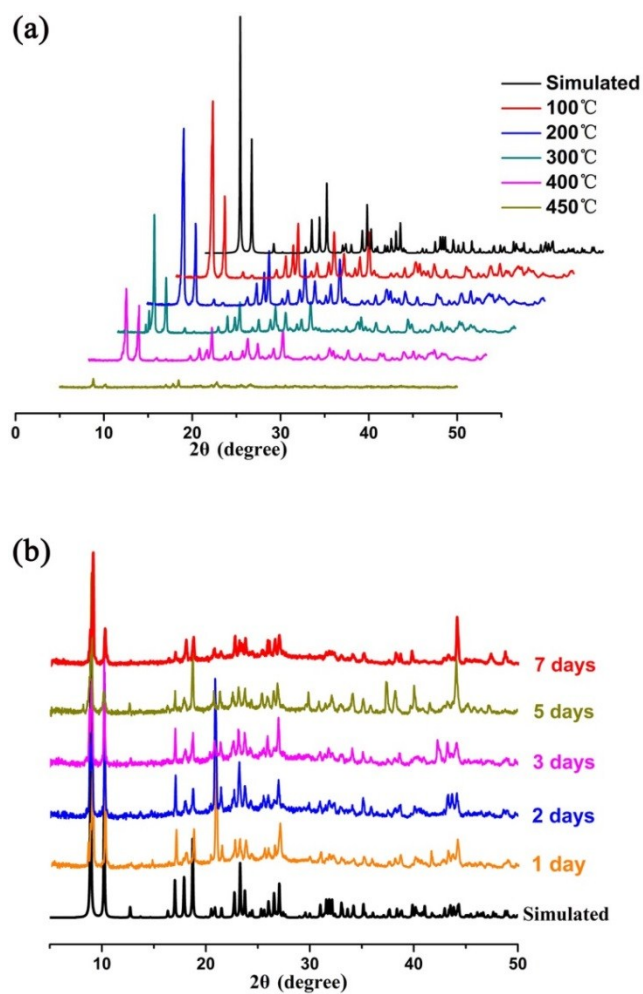


Fig. S6 (a) The X-ray thermodiffractogram of **GdL**. (b) PXRD patterns of **GdL** after exposed to open air containing H₂O vapor for 1 day, 2 days, 3 days, 5 days and 7 days.

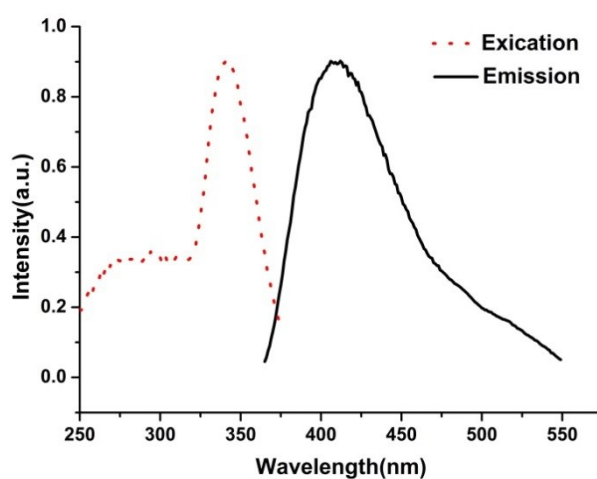


Fig. S7 Solid-state luminescent spectra of the ligand H₄L (excitation: dot, λ_{em} = 412 nm; emission: solid, λ_{ex} = 341 nm) at room temperature.

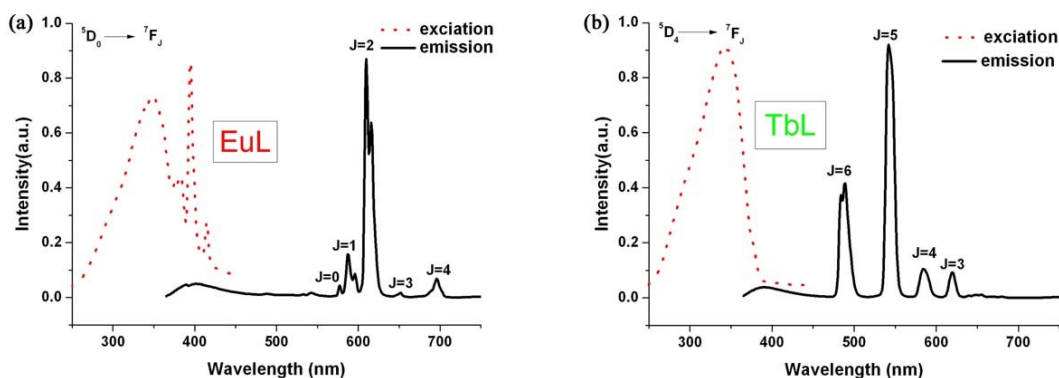


Fig. S8 Solid-state luminescent spectra of (a) **EuL** (excitation: dot, λ_{em} = 612 nm; emission: solid, λ_{ex} = 335 nm) and (b) **TbL** (excitation: dot, λ_{em} = 542 nm; emission: solid, λ_{ex} = 335 nm) at room temperature.

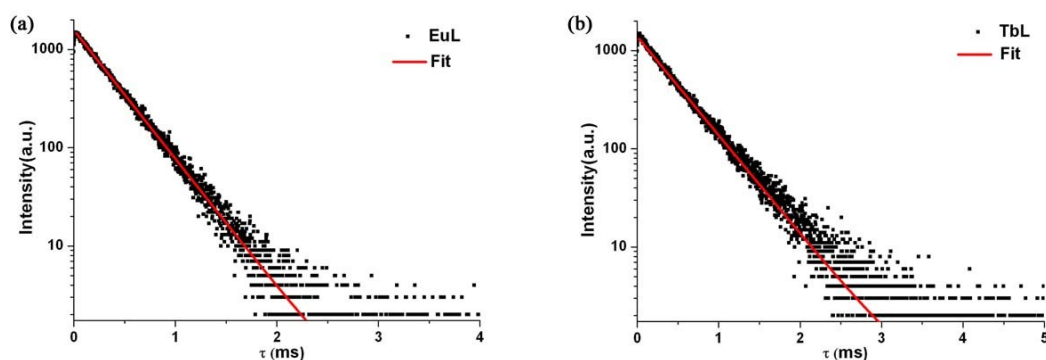


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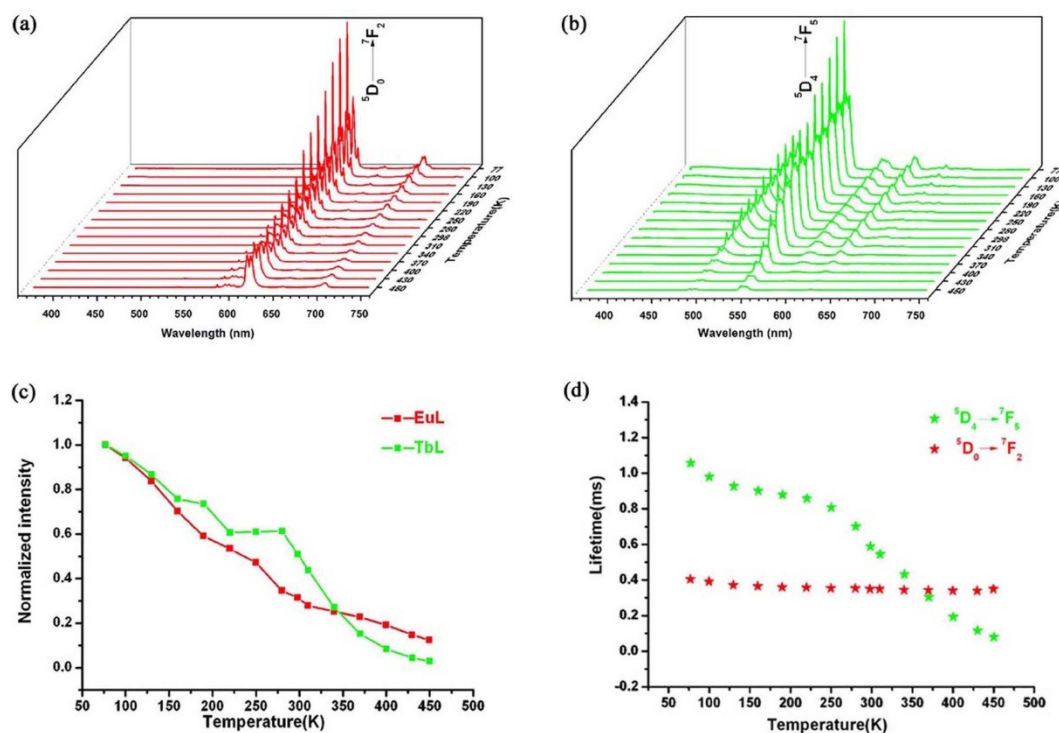


Fig. S10 Solid-state emission spectra of (a) **EuL** and (b) **TbL** recorded between 77 K and 450 K excited at 335 nm. (c) Temperature-dependent normalized emission intensity of the $^5D_0 \rightarrow ^7F_2$ transitions (612 nm) for **EuL** and $^5D_4 \rightarrow ^7F_5$ transitions (542 nm) for **TbL**. (d) Temperature-dependent lifetimes of the $^5D_0 \rightarrow ^7F_2$ transitions (612 nm) for **EuL** and $^5D_4 \rightarrow ^7F_5$ transitions (542 nm) for **TbL** excited at 335 nm.

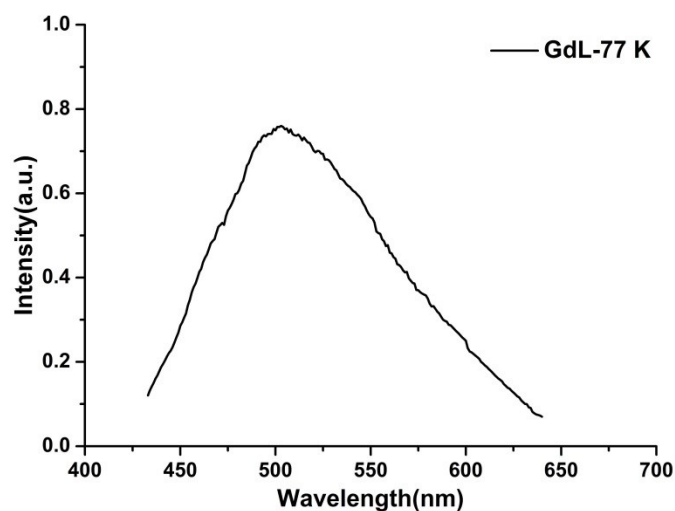


Fig. S11 The phosphorescence spectra of compound [Me₂NH₂][GdL(H₂O)₂] (**GdL**) at 77 K. The ligand-centered triplet state T₁ was calculated from the shortest wavelength emission edge of the phosphorescence spectrum of compound [Me₂NH₂][GdL(H₂O)₂]

(GdL) at 77 K. The phosphorescence spectrum of **GdL** reveals that the triplet state energy level T_1 of the ligand is 23753 cm^{-1} (421 nm).

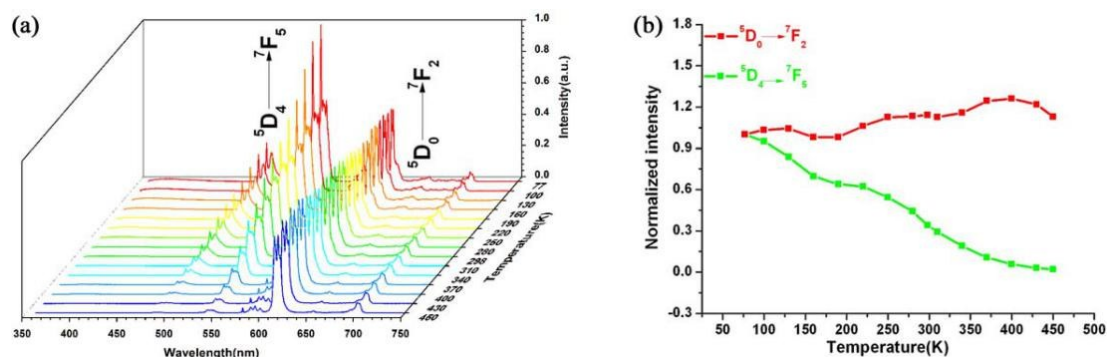


Fig. S12 (a) Solid-state emission spectra of $\text{Eu}_{0.060}\text{Tb}_{0.94}\text{L}$ recorded between 77 K and 450 K excited at 335 nm. (b) Temperature-dependent normalized emission intensity of the $^5\text{D}_4 \rightarrow ^7\text{F}_5$ and $^5\text{D}_0 \rightarrow ^7\text{F}_2$ transitions of $\text{Eu}_{0.060}\text{Tb}_{0.94}\text{L}$.

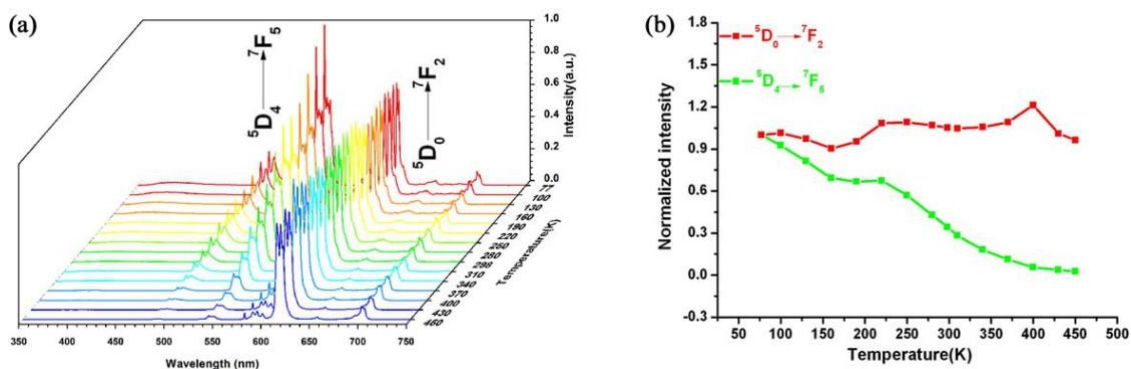


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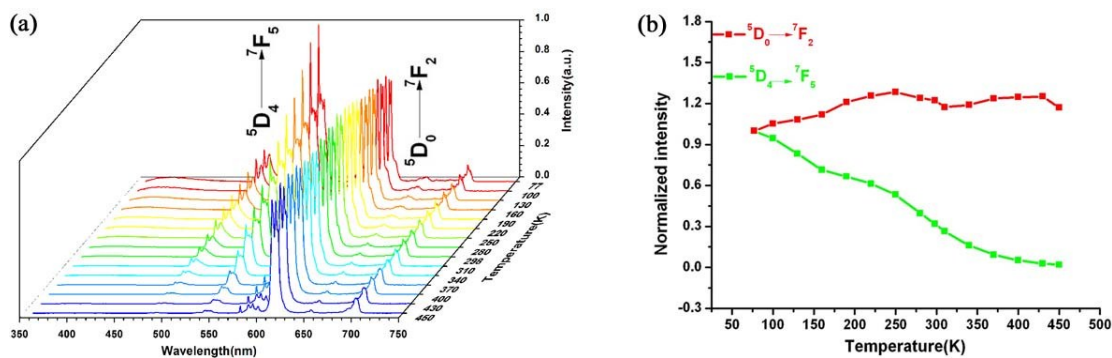


Fig. S14 (a) Solid-state emission spectra of $\text{Eu}_{0.035}\text{Tb}_{0.965}\text{L}$ recorded between 77 K and

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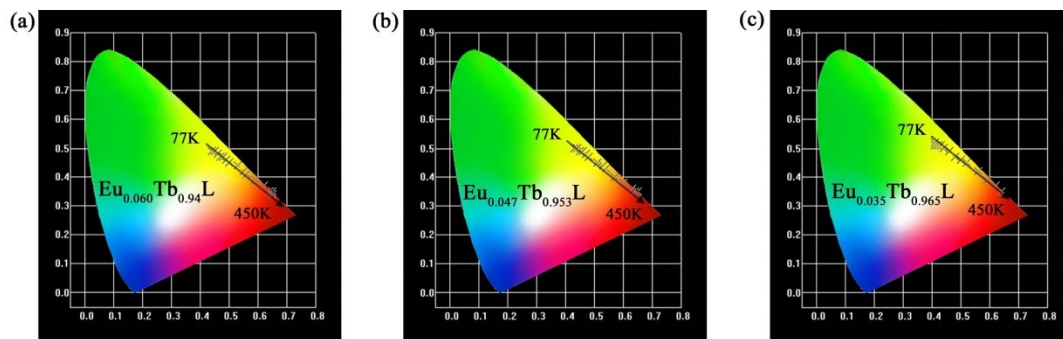


Fig. S15 CIE-1931 chromaticity diagram showing the luminescent colors of the binary mixed **Eu_xTb_yL** ($x=0.060, 0.047, 0.035, 0.0066$) at different temperatures from 77 K to 450 K.

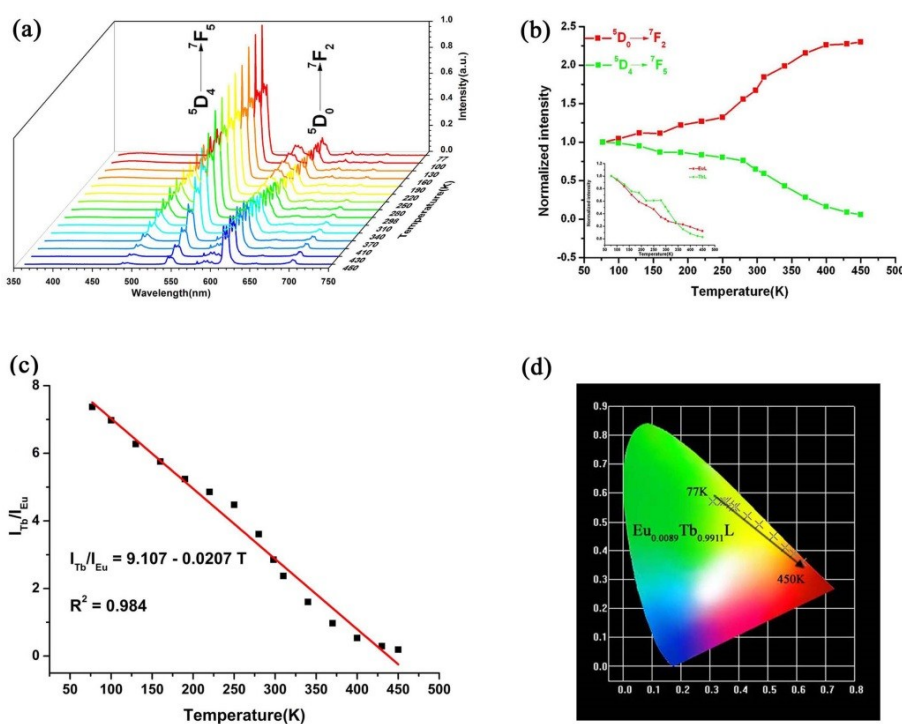


Fig. S16 (a) Solid-state emission spectra of **Eu_{0.0089}Tb_{0.9911}L** recorded between 77 K and 450 K excited at 335 nm. (b) Temperature-dependent normalized emission intensity of the $^5D_4 \rightarrow ^7F_5$ and $^5D_0 \rightarrow ^7F_2$ transitions of **Eu_{0.0089}Tb_{0.9911}L**. (c) Temperature-dependent intensity ratio of Tb³⁺ (542 nm) to Eu³⁺ (612 nm) for binary mixed **Eu_{0.0089}Tb_{0.9911}L** at different temperatures from 77 to 450 K. (d) CIE-1931 chromaticity

diagram showing the luminescent colors of the binary mixed $\text{Eu}_{0.0089}\text{Tb}_{0.9911}\text{L}$.

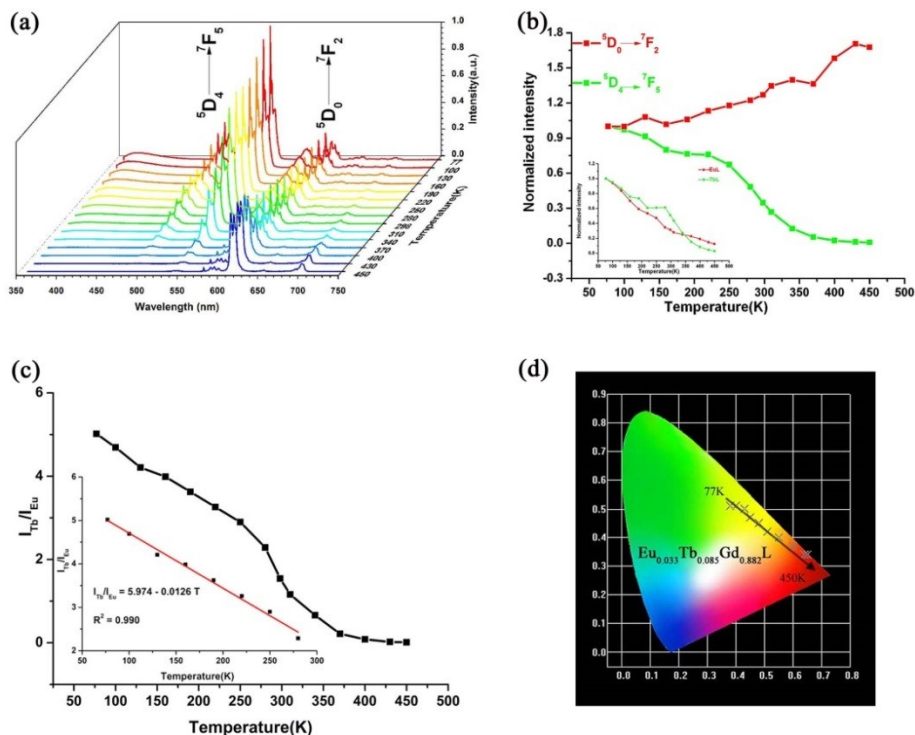


Fig. S17 (a) Solid-state emission spectra of $\text{Eu}_{0.033}\text{Tb}_{0.085}\text{Gd}_{0.882}\text{L}$ recorded between 77 K and 450 K excited at 335 nm. (b) Temperature-dependent normalized emission intensity of the $^5\text{D}_4 \rightarrow ^7\text{F}_5$ and $^5\text{D}_0 \rightarrow ^7\text{F}_2$ transitions of $\text{Eu}_{0.033}\text{Tb}_{0.085}\text{Gd}_{0.882}\text{L}$. (c) Temperature-dependent intensity ratio of Tb^{3+} (542 nm) to Eu^{3+} (612 nm) for ternary mixed $\text{Eu}_{0.033}\text{Tb}_{0.085}\text{Gd}_{0.882}\text{L}$ at different temperatures from 77 to 450 K. (d) CIE-1931 chromaticity diagram showing the luminescent colors of the ternary mixed $\text{Eu}_{0.033}\text{Tb}_{0.085}\text{Gd}_{0.882}\text{L}$.

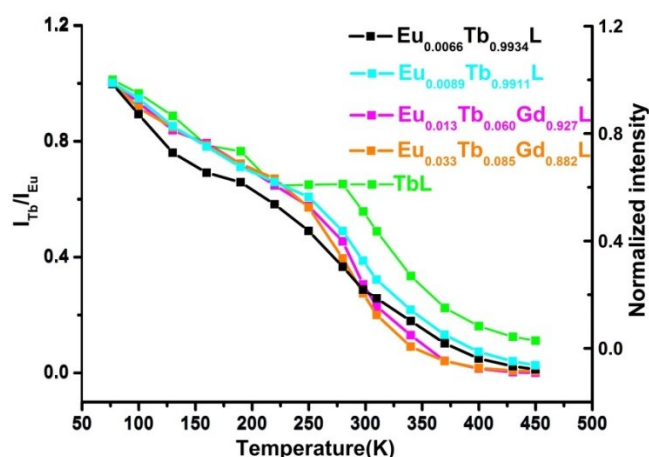


Fig. S18 Comparison of the temperature-dependent intensity ratio ($I_{\text{Tb}}/I_{\text{Eu}}$) of $\text{Eu}_{0.0089}\text{Tb}_{0.9911}\text{L}$, $\text{Eu}_{0.0066}\text{Tb}_{0.9934}\text{L}$, $\text{Eu}_{0.013}\text{Tb}_{0.060}\text{Gd}_{0.927}\text{L}$, $\text{Eu}_{0.033}\text{Tb}_{0.085}\text{Gd}_{0.882}\text{L}$ and temperature-dependent intensity of Tb^{3+} (542 nm) for TbL .

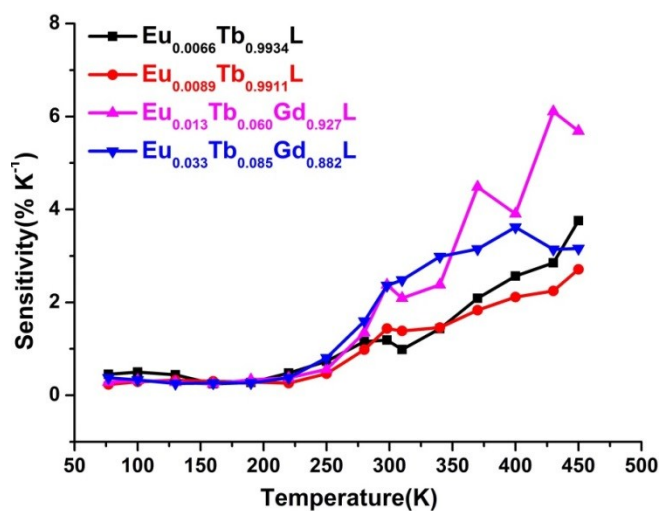


Fig. S19 The relative thermometric sensitivity values for mixed LnMOFs $\text{Eu}_{0.0089}\text{Tb}_{0.9911}\text{L}$, $\text{Eu}_{0.0066}\text{Tb}_{0.9934}\text{L}$, $\text{Eu}_{0.013}\text{Tb}_{0.060}\text{Gd}_{0.927}\text{L}$ and $\text{Eu}_{0.033}\text{Tb}_{0.085}\text{Gd}_{0.882}\text{L}$ determined from the curves plotting of $I_{\text{Tb}}/I_{\text{Eu}}$ vs. temperature.

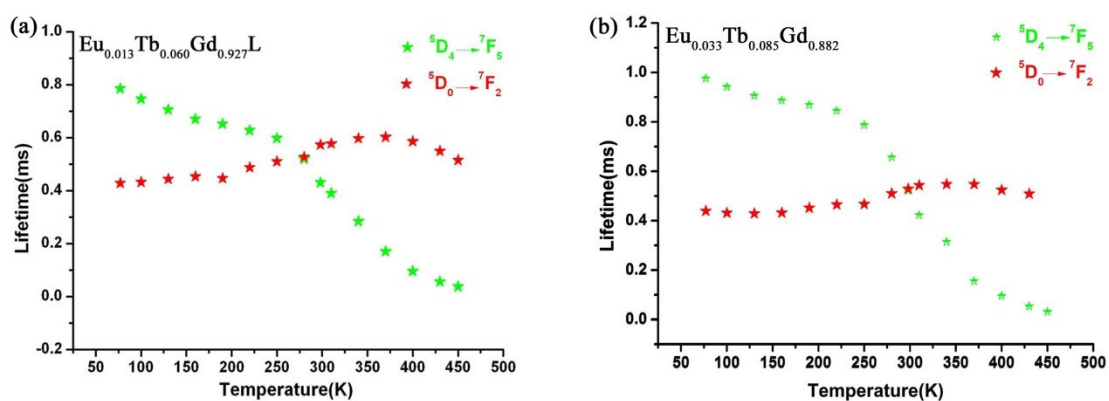
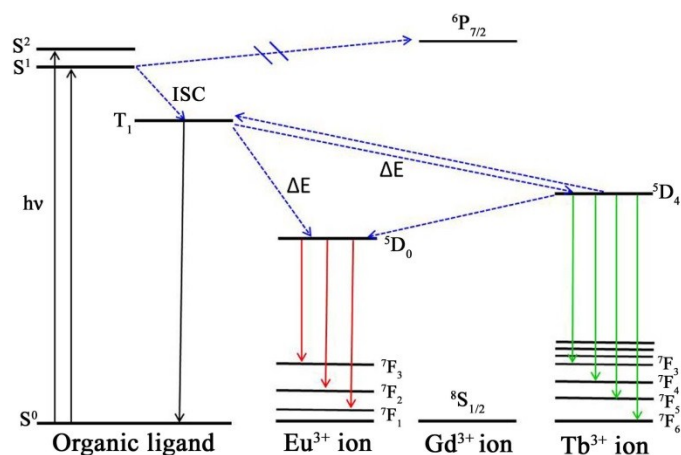


Fig. S20 Temperature-dependent lifetimes of the $^5D_0 \rightarrow ^7F_2$ transitions (612 nm) and $^5D_4 \rightarrow ^7F_5$ transitions (542 nm) for the ternary mixed (a) $\text{Eu}_{0.013}\text{Tb}_{0.060}\text{Gd}_{0.882}\text{L}$ and (b) $\text{Eu}_{0.033}\text{Tb}_{0.085}\text{Gd}_{0.882}\text{L}$ between 77 K and 450 K excited at 335 nm.

Scheme S1. Simplified schematic diagram of the ligand–metal energy transfer in the ternary mixed lanthanide MOFs (S_0 is the ground state of the ligand; S_1 and T_1 are the singlet state and triplet state of the ligand, respectively.).



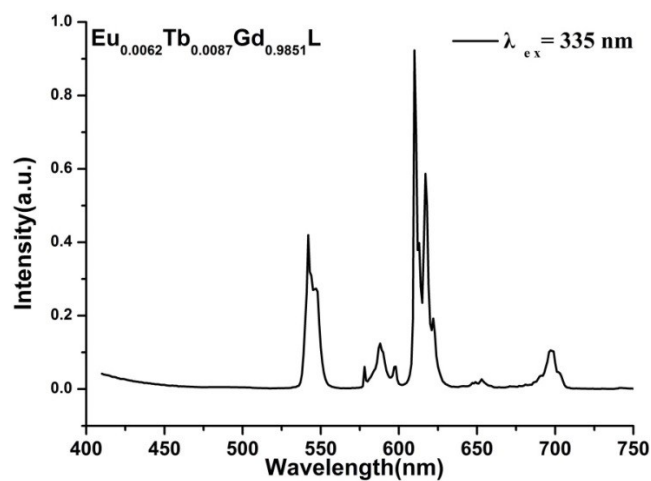


Fig. S21 Solid-state emission spectrum of the $\text{Eu}_{0.0062}\text{Tb}_{0.0087}\text{Gd}_{0.9851}\text{L}$ at room temperature excited at 335 nm.

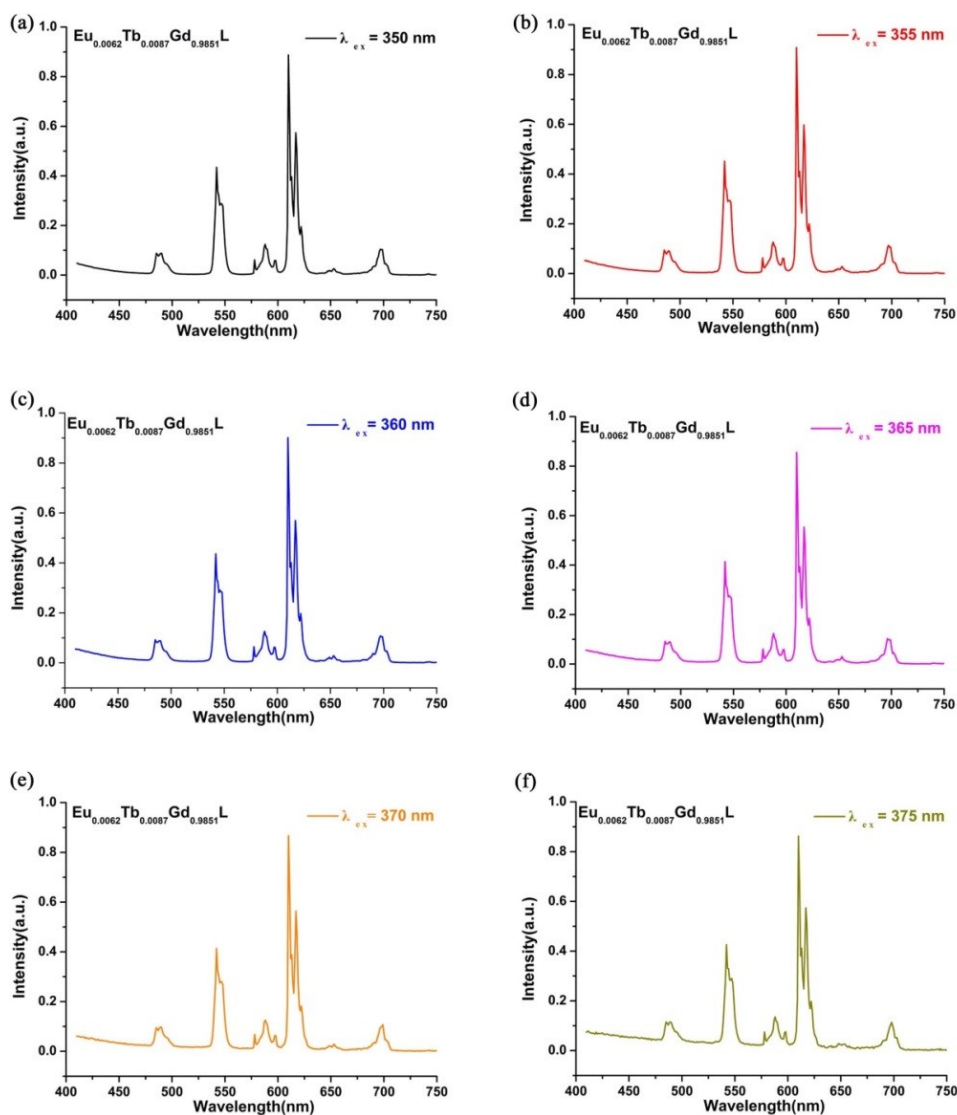


Fig. S22 Solid-state emission spectra of the **Eu_{0.0062}Tb_{0.0087}Gd_{0.9851}L** at room temperature excited at (a) 350 nm, (b) 355 nm, (c) 360 nm, (d) 365 nm, (e) 370 nm, (f) 375 nm.

Table S1. The original ratios of different lanthanide metal salts and the corresponding ICP results.

	Experimental			ICP Results		
Eu_xTb_yL	Eu(x)	Tb(y)		Eu(x)	Tb(y)	
Eu_{0.060}Tb_{0.94}L	0.1	0.9		0.060	0.94	
Eu_{0.047}Tb_{0.953}L	0.08	0.92		0.047	0.953	
Eu_{0.035}Tb_{0.965}L	0.05	0.95		0.035	0.965	
Eu_{0.0089}Tb_{0.9911}L	0.02	0.98		0.0089	0.9911	
Eu_{0.0066}Tb_{0.9934}L	0.01	0.99		0.0066	0.9934	
Eu_xTb_yGd_{1-x-y}L	Eu(x)	Tb(y)	Gd(1-x-y)	Eu(x)	Tb(y)	Gd(1-x-y)
Eu_{0.033}Tb_{0.085}Gd_{0.882}L	0.03	0.07	0.90	0.033	0.085	0.882
Eu_{0.013}Tb_{0.060}Gd_{0.927}L	0.01	0.045	0.945	0.013	0.060	0.927
Eu_{0.0062}Tb_{0.0087}Gd_{0.9851}L	0.005	0.005	0.99	0.0062	0.0087	0.9851

Table S2. The parameters containing composition, temperature ranges (K), maximum relative sensitivity values (S_m , % K⁻¹), the temperature at which S_m is maximum (T_m , K) of several selected ratiometric luminescent MOF thermometers.

Luminescent M'LnMOF Thermometers	Temperature range (K)	S_m (% K ⁻¹)	T_m (K)	Ref.
Eu _{0.0069} Tb _{0.9931} -DMBDC	50 - 200	1.15	200	7d
Tb _{0.9} Eu _{0.1} -PIA	100 - 300	3.27	300	4a
Tb _{0.98} Eu _{0.02} -OA-DSTP	77 - 275	2.4	275	21c
Tb _{0.98} Eu _{0.02} -BDC-DSTP	77 - 225	2.8	225	
Tb _{0.99} Eu _{0.01} -BDC-DSTP	77 - 200	3.9	200	
[Eu _{0.102} Tb _{0.898} (notpH ₄)(NO ₃)(H ₂ O)]·8H ₂ O	18 - 300	3.9	38	21d
Tb 0.914 Eu 0.086 -PDA Tb 0.914 Eu 0.086 -PDA Tb 0.914 Eu 0.086 -PDA Tb _{0.914} Eu _{0.086} -PDA	10 - 325	5.96	25	21e
Eu _{0.2} Tb _{0.8} L Eu _{0.1} Tb _{0.9} L Eu _{0.3} Tb _{0.7} L	40 - 300	0.15 0.11 0.17	300	22a
(Eu ₁ Tb ₉₉ -HFA)@LA	197 - 287	1.107	287	22b
ZJU-88⌐perylene	293.15 – 353.15	1.28	193.15	22c
Tb _{0.957} Eu _{0.043} cpda	40 - 300	1.77	250	22d
d-U(600)-Eu _{0.25} Tb _{0.75} (btfa) ₃ (bpeta)	10 - 330	4.9	150	22e
Tb _{0.99} Eu _{0.01} (BDC) _{1.5} ·(H ₂ O) ₂	298 – 320	0.31	318	20b

Eu_{0.0089}Tb_{0.9911}L	77-450	2.71	450	This work
Eu_{0.0066}Tb_{0.9934}L	77-450	3.76	450	This work
Eu_{0.033}Tb_{0.085}Gd_{0.882}L	77-450	3.62	400	This work
Eu_{0.013}Tb_{0.060}Gd_{0.927}L	77-450	6.11	430	This work

DMBDC = 2,5-dimethoxy-1,4-benzenedicarboxylate, HPIA = 5-(pyridin-4-yl)isophthalic acid, H₂DSTP = 2,4-(2,2':6',2''-terpyridin-4'-yl)-benzenedisulfonic acid, OA = oxalic acid, BDC = 1,4-benzene dicarboxylic acid, notpH₆ = 1,4,7-triazacyclononane-1,4,7-triyl-tris(methylene-phosphonic acid), PDA = 1,4-phenylenediacetic acid, L = 1,3-bis(4-carboxyphenyl)imidazolium, cpda = 5-(4-carboxyphenyl)-2,6-pyridinedicarboxylate, HFA = hexafluoroacetylacetone, ZJU-88 = [Eu₂(QPTCA)(NO₃)₂(DMF)₄]·(CH₃CH₂OH)₃ (H₄QPTCA = 1,1':4',1'':4'',1'''-quaterphenyl-3,3'',5,5'''-tetracarboxylic acid), H₃cpda = 5-(4-carboxyphenyl)-2,6-pyridinedicarboxylic acid, bpeta = 1,2-bis(4-pyridyl)ethane.

Table S3. Crystal data and refinement results for **EuL**, **GdL** and **TbL**.

	EuL	GdL	TbL
Formula	C ₂₃ H ₂₁ EuN ₂ O ₁₀	C ₂₃ H ₂₁ GdN ₂ O ₁₀	C ₂₃ H ₂₁ TbN ₂ O ₁₀
Formula weight	637.39	642.68	652.36
Crystal system	orthorhombic	orthorhombic	orthorhombic
space group	<i>Pnma</i>	<i>Pnma</i>	<i>Pnma</i>
<i>a</i> (Å)	8.5023 (2)	8.5011 (3)	8.5052 (5)
<i>b</i> (Å)	17.3319 (4)	17.2958 (5)	17.3256 (4)
<i>c</i> (Å)	12.1180 (3)	12.0726 (3)	12.0864 (3)
α (°)	90	90	90
β (°)	90	90	90
γ (°)	90	90	90
Volume (Å ³)	1785.72 (7)	1775.08 (9)	1781.02 (7)
T (K)	100	100	100
Z	4	4	4
F (000)	1140	1144	1148
R ₁ (I>2σ(I))	0.0360	0.0467	0.0382
wR ₂ (reflections)	0.1488	0.1064	0.1014
Goodness of fit on <i>F</i> ²	1.214	1.157	1.071

Table S4. Selected bond lengths (Å) and bond angles (°) for **EuL**, **GdL** and **TbL**.

Bond	Dist.	Bond	Dist.
Eu1—O6 ⁱ	2.355 (3)	Eu1—O4	2.450 (5)
Eu1—O6 ⁱⁱ	2.355 (3)	Eu1—O4 ^v	2.450 (5)
Eu1—O3 ⁱⁱⁱ	2.394 (4)	Eu1—O2	2.465 (5)
Eu1—O3 ^{iv}	2.394 (4)	Eu1—O1	2.547 (4)
Angle	(°)	Angle	(°)
O6 ⁱ —Eu1—O6 ⁱⁱ	125.4 (2)	O4—Eu1—O4 ^v	71.3 (2)
O6 ⁱ —Eu1—O3 ⁱⁱⁱ	74.31 (14)	O6 ⁱ —Eu1—O2	65.67 (11)
O6 ⁱⁱ —Eu1—O3 ⁱⁱⁱ	137.57 (14)	O6 ⁱⁱ —Eu1—O2	65.67 (11)
O6 ⁱ —Eu1—O3 ^{iv}	137.57 (14)	O3 ⁱⁱⁱ —Eu1—O2	137.92 (11)
O6 ⁱⁱ —Eu1—O3 ^{iv}	74.31 (14)	O3 ^{iv} —Eu1—O2	137.92 (11)
O3 ⁱⁱⁱ —Eu1—O3 ^{iv}	69.14 (18)	O4—Eu1—O2	109.38 (14)
O6 ⁱ —Eu1—O4	143.52 (14)	O4 ^v —Eu1—O2	109.38 (14)
O6 ⁱⁱ —Eu1—O4	76.63 (14)	O6 ⁱ —Eu1—O1	73.52 (10)
O3 ⁱⁱⁱ —Eu1—O4	110.42 (12)	O6 ⁱⁱ —Eu1—O1	73.52 (10)
O3 ^{iv} —Eu1—O4	71.80 (12)	O3 ⁱⁱⁱ —Eu1—O1	78.87 (11)
O6 ⁱ —Eu1—O4 ^v	76.63 (14)	O3 ^{iv} —Eu1—O1	78.87 (11)
O6 ⁱⁱ —Eu1—O4 ^v	143.52 (14)	O4—Eu1—O1	142.66 (12)
O3 ⁱⁱⁱ —Eu1—O4 ^v	71.80 (12)	O4 ^v —Eu1—O1	142.66 (12)
O3 ^{iv} —Eu1—O4 ^v	110.42 (12)	O2—Eu1—O1	77.90 (16)

Symmetry codes: (i) 1-x, -1/2+y, 1-z; (ii) 1-x, 1-y, 1-z; (iii) x, 1/2-y, z; (iv) 1/2+x, y, 3/2-z; (v) 1/2+x, 1/2-y, 3/2-z.

Bond	Dist.	Bond	Dist.
Gd1—O6 ⁱ	2.322 (6)	Gd1—O3	2.425 (5)
Gd1—O6 ⁱⁱ	2.322 (6)	Gd1—O3 ^v	2.425 (5)
Gd1—O4 ⁱⁱⁱ	2.367 (6)	Gd1—O1	2.465 (10)
Gd1—O4 ^{iv}	2.367 (6)	Gd1—O2	2.517 (6)
Angle	(°)	Angle	(°)
O6 ⁱ —Gd1—O6 ⁱⁱ	123.0 (4)	O3—Gd1—O3 ^v	71.5 (2)
O6 ⁱ —Gd1—O4 ⁱⁱⁱ	138.8 (2)	O6 ⁱ —Gd1—O1	64.3 (2)
O6 ⁱⁱ —Gd1—O4 ⁱⁱⁱ	75.5 (3)	O6 ⁱⁱ —Gd1—O1	64.3 (2)
O6 ⁱ —Gd1—O4 ^{iv}	75.5 (3)	O4 ⁱⁱⁱ —Gd1—O1	137.7 (2)
O6 ⁱⁱ —Gd1—O4 ^{iv}	138.8 (2)	O4 ^{iv} —Gd1—O1	137.7 (2)
O4 ⁱⁱⁱ —Gd1—O4 ^{iv}	69.6 (3)	O3—Gd1—O1	108.7 (2)
O6 ⁱ —Gd1—O3	143.2 (2)	O3 ^v —Gd1—O1	108.7 (2)
O6 ⁱⁱ —Gd1—O3	76.9 (2)	O6 ⁱ —Gd1—O2	73.72 (16)
O4 ⁱⁱⁱ —Gd1—O3	72.21 (19)	O6 ⁱⁱ —Gd1—O2	73.72 (16)
O4 ^{iv} —Gd1—O3	111.2 (2)	O4 ⁱⁱⁱ —Gd1—O2	78.32 (18)
O6 ⁱ —Gd1—O3 ^v	76.9 (2)	O4 ^{iv} —Gd1—O2	78.32 (18)
O6 ⁱⁱ —Gd1—O3 ^v	143.2 (2)	O3—Gd1—O2	142.59 (13)
O4 ⁱⁱⁱ —Gd1—O3 ^v	111.2 (2)	O3 ^v —Gd1—O2	142.59 (13)

O4 ^{iv} —Gd1—O3 ^v	72.21 (19)	O1—Gd1—O2	78.6 (3)
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Symmetry codes: (i) 1-x, -1/2+y, 1-z; (ii) 1-x, 1-y, 1-z; (iii) x, 1/2-y, z; (iv) 1/2+x, y, 3/2-z; (v) 1/2+x, 1/2-y, 3/2-z.

Bond	Dist.	Bond	Dist.
Tb1—O6 ⁱ	2.339 (4)	Tb1—O3 ^{iv}	2.434 (4)
Tb1—O6 ⁱⁱ	2.339 (4)	Tb1—O3 ^v	2.434 (4)
Tb1—O4	2.366 (4)	Tb1—O1	2.462 (7)
Tb1—O4 ⁱⁱⁱ	2.366 (4)	Tb1—O2	2.522 (5)
Angle	(°)	Angle	(°)
O6 ⁱ —Tb1—O6 ⁱⁱ	123.5 (3)	O3 ^{iv} —Tb1—O3 ^v	71.55 (19)
O6 ⁱ —Tb1—O4	138.66 (16)	O6 ⁱ —Tb1—O1	64.64 (14)
O6 ⁱⁱ —Tb1—O4	75.04 (17)	O6 ⁱⁱ —Tb1—O1	64.64 (14)
O6 ⁱ —Tb1—O4 ⁱⁱⁱ	75.04 (17)	O4—Tb1—O1	137.75 (13)
O6 ⁱⁱ —Tb1—O4 ⁱⁱⁱ	138.66 (16)	O4 ⁱⁱⁱ —Tb1—O1	137.75 (13)
O4—Tb1—O4 ⁱⁱⁱ	69.9 (2)	O3 ^{iv} —Tb1—O1	108.31 (16)
O6 ⁱ —Tb1—O3 ^{iv}	143.11 (14)	O3 ^v —Tb1—O1	108.31 (16)
O6 ⁱⁱ —Tb1—O3 ^{iv}	76.68 (14)	O6 ⁱ —Tb1—O2	73.61 (11)
O4—Tb1—O3 ^{iv}	72.33 (13)	O6 ⁱⁱ —Tb1—O2	73.61 (11)
O4 ⁱⁱⁱ —Tb1—O3 ^{iv}	111.46 (13)	O4—Tb1—O2	78.59 (12)
O6 ⁱ —Tb1—O3 ^v	76.68 (14)	O4 ⁱⁱⁱ —Tb1—O2	78.59 (12)
O6 ⁱⁱ —Tb1—O3 ^v	143.11 (14)	O3 ^{iv} —Tb1—O2	142.72 (10)
O4—Tb1—O3 ^v	111.46 (13)	O3 ^v —Tb1—O2	142.72 (10)
O4 ⁱⁱⁱ —Tb1—O3 ^v	72.33 (13)	O1—Tb1—O2	78.5 (2)

Symmetry codes: (i) 1-x, -1/2+y, 1-z; (ii) 1-x, 1-y, 1-z; (iii) x, 1/2-y, z; (iv) 1/2+x, y, 3/2-z; (v) 1/2+x, 1/2-y, 3/2-z.