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Supporting Information

Supplementary Information

Cobalt-Lanthanide Coordination Polymers Constructed with Metalloligands: A Ferromagnetic Coupled Quasi-1D Dy^{3+} Chain Showing Slow Relaxation

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Supplementary Index

Experimental Section:

Materials and Methods. All of the chemicals were purchased from commercial suppliers and were used without further purification. Elemental analyses were carried out by the Elemental Analysis Lab of our Institute. The IR spectra were recorded as KBr pellets on a Spectrum One FT-IR spectrometer (PerkinElmer Instruments). X-ray powder diffraction data were recorded on a Rigaku MultiFlex diffractometer at 40 kV, 40 mA for Cu Kα (λ = 1.5406 Å), with a scan speed of 0.05-0.2 deg/min. Simulated XRD patterns were calculated with the SHELXTL-XPOW program using the single-crystal data. TGA experiments were carried out at a heating rate of 10 °C/min in N_2 atmosphere. Magnetic measurements for powder samples of **1** and **2** were carried out with a Quantum Design PPMS model 6000 magnetometer.

X-ray crystallography: Data collections were performed at 293(2) K on a Mercury CCD diffractometer with graphite monochromated Mo-Kα radiation (λ = 0.71073 Å). The structures were solved by direct methods and all calculations were performed using the SHELXL package [S1]. The structures were refined by full matrix least squares with anisotropic displacement parameters for non-hydrogen atoms. In addition, all hydrogen atoms belonging to the water molecules were found in the electron density map and refined isotropically. The other H atoms were generated geometrically and treated as riding. The crystallographic data are summarized in Table 1, and selected bond lengths and angles of all compounds are listed in Table S1-S6 in the Supplementary Information.

Table S1. Selected bond lengths [Å] and angles [°]for metalloligand.

bond			
Co1—O9	1.890 (3)	Co1—N3	1.913 (4)
Co1—O5	1.893 (3)	Co1—N2	1.919 (4)
Co1—O1	1.896 (4)	Co1—N1	1.935 (4)
angle			
O9—Co1—O5	178.68 (14)	O1—Co1—N2	176.62 (15)
O9—Co1—O1	92.04 (15)	N3—Co1—N2	92.77 (17)
O5—Co1—O1	88.78 (15)	O9—Co1—N1	89.80 (15)
O9—Co1—N3	94.10 (15)	O5—Co1—N1	91.30 (15)
O5—Co1—N3	84.91 (15)	O1—Co1—N1	84.84 (17)
O1—Co1—N3	87.07 (17)	N3—Co1—N1	171.14 (17)
O9—Co1—N2	84.60 (15)	N2—Co1—N1	95.52 (17)
O5—Co1—N2	94.57 (15)		

Table S2. Selected bond lengths [Å] and angles [°]for **1a**.

bond			
Tb1—O17	2.325 (3)	Tb2—O7^{iii}	2.316 (4)
Tb1—O25	2.330 (4)	Tb2—O3	2.320 (3)
Tb1—O4^{i}	2.345 (3)	Tb2—O14^{ii}	2.341 (3)
Tb1—O13^{ii}	2.354 (3)	Tb2—O18	2.364 (4)
Tb1—O11^{i}	2.416 (4)	Tb2—O27	2.364 (4)
Tb1—O26	2.431 (4)	Tb2—O28	2.401 (4)
Tb1—O22^{i}	2.437 (3)	Tb2—O12^{i}	2.454 (4)
Tb1—O21^{i}	2.447 (4)	Tb2—O11^{i}	2.551 (4)
angle			
O17—Tb1—O25	145.15 (13)	O7^{iii}—Tb2—O3	74.30 (12)
O17—Tb1—O4^{i}	133.08 (12)	O7^{iii}—Tb2—O14^{ii}	132.06 (13)
O25—Tb1—O4^{i}	77.52 (13)	O3—Tb2—O14^{ii}	138.35 (13)
O17—Tb1—O13^{ii}	73.86 (13)	O7^{iii}—Tb2—O18	149.29 (13)
O25—Tb1—O13^{ii}	85.74 (13)	O3—Tb2—O18	75.16 (12)
O4^{i}—Tb1—O13^{ii}	147.08 (12)	O14^{ii}—Tb2—O18	75.37 (13)
O17—Tb1—O11^{i}	76.14 (12)	O7^{iii}—Tb2—O27	82.99 (14)
O25—Tb1—O11^{i}	72.95 (13)	O3—Tb2—O27	79.15 (13)
O4^{i}—Tb1—O11^{i}	119.50 (12)	O14^{ii}—Tb2—O27	74.48 (13)
O13^{ii}—Tb1—O11^{i}	81.09 (12)	O18—Tb2—O27	94.40 (14)
O17—Tb1—O26	73.53 (12)	O7^{iii}—Tb2—O28	70.71 (14)
O25—Tb1—O26	114.28 (13)	O3—Tb2—O28	144.76 (14)
O4^{i}—Tb1—O26	68.38 (12)	O14^{ii}—Tb2—O28	68.67 (14)
O13^{ii}—Tb1—O26	144.43 (12)	O18—Tb2—O28	139.98 (14)
O11^{i}—Tb1—O26	77.68 (13)	O27—Tb2—O28	92.45 (17)
O17—Tb1—O22^{i}	121.27 (12)	O7^{iii}—Tb2—O12^{i}	69.36 (14)

O25—Tb1—O22^{i}	76.27 (13)	O3—Tb2—O12^{i}	87.14 (13)
O4^{i}—Tb1—O22^{i}	76.80 (12)	O14^{ii}—Tb2—O12^{i}	129.21 (13)
O13^{ii}—Tb1—O22^{i}	71.75 (13)	O18—Tb2—O12^{i}	106.02 (15)
O11^{i}—Tb1—O22^{i}	140.06 (13)	O27—Tb2—O12^{i}	151.63 (14)
O26—Tb1—O22^{i}	139.37 (13)	O28—Tb2—O12^{i}	84.51 (17)
O17—Tb1—O21^{i}	77.84 (13)	O7^{iii}—Tb2—O11^{i}	114.35 (12)
O25—Tb1—O21^{i}	129.01 (13)	O3—Tb2—O11^{i}	121.82 (12)
O4^{i}—Tb1—O21^{i}	83.94 (13)	O14^{ii}—Tb2—O11^{i}	80.43 (12)
O13^{ii}—Tb1—O21^{i}	85.00 (14)	O18—Tb2—O11^{i}	79.68 (12)
O11^{i}—Tb1—O21^{i}	153.03 (12)	O27—Tb2—O11^{i}	154.90 (13)
O26—Tb1—O21^{i}	101.61 (13)	O28—Tb2—O11^{i}	77.64 (16)

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

Table S3. Selected bond lengths [Å] and angles [°]for **1b**.

bond			
Dy1—O20	2.297 (7)	Dy2—O3^{iii}	2.296 (7)
Dy1—O26	2.319 (7)	Dy2—O7^{iv}	2.302 (7)
Dy1—O24^{i}	2.333 (7)	Dy2—O23^{i}	2.324 (8)
Dy1—O8	2.342 (7)	Dy2—O19	2.327 (7)
Dy1—O11	2.395 (7)	Dy2—O27	2.364 (8)
Dy1—O25	2.407 (8)	Dy2—O28	2.403 (9)
Dy1—O15^{ii}	2.415 (8)	Dy2—O12	2.428 (9)
Dy1—O16^{ii}	2.419 (8)	Dy2—O11	2.560 (8)
		angle	
O20—Dy1—O26	144.3 (3)	O3^{iii}—Dy2—O7^{iv}	74.5 (3)
O20—Dy1—O24^{i}	74.4 (3)	O3^{iii}—Dy2—O23^{i}	132.4 (3)
O26—Dy1—O24^{i}	85.9 (3)	O7^{iv}—Dy2—O23^{i}	138.2 (3)
O20—Dy1—O8	133.5 (3)	O3^{iii}—Dy2—O19	149.1 (3)
O26—Dy1—O8	77.1 (3)	O7^{iv}—Dy2—O19	74.9 (3)
O24^{i}—Dy1—O8	146.5 (3)	O23^{i}—Dy2—O19	75.6 (3)
O20—Dy1—O11	75.3 (3)	O3^{iii}—Dy2—O27	83.1 (3)
O26—Dy1—O11	72.3 (3)	O7^{iv}—Dy2—O27	78.7 (3)
O24^{i}—Dy1—O11	80.8 (3)	O23^{i}—Dy2—O27	75.1 (3)
O8—Dy1—O11	119.7 (3)	O19—Dy2—O27	95.0 (3)
O20—Dy1—O25	73.5 (3)	O3^{iii}—Dy2—O28	71.2 (3)
O26—Dy1—O25	112.9 (3)	O7^{iv}—Dy2—O28	145.3 (3)
O24^{i}—Dy1—O25	144.9 (3)	O23^{i}—Dy2—O28	68.1 (3)
O8—Dy1—O25	68.5 (3)	O19—Dy2—O28	139.6 (3)
O11—Dy1—O25	77.6 (3)	O27—Dy2—O28	92.4 (4)
O20—Dy1—O15^{ii}	78.7 (3)	O3^{iii}—Dy2—O12	69.5 (3)
O26—Dy1—O15^{ii}	129.8 (3)	O7^{iv}—Dy2—O12	86.9 (3)
O24^{i}—Dy1—O15^{ii}	85.3 (3)	O23^{i}—Dy2—O12	129.2 (3)
O8—Dy1—O15^{ii}	83.9 (3)	O19—Dy2—O12	104.7 (3)

O11—Dy1—O15^{ii}	153.0 (3)	O27—Dy2—O12	151.6 (3)
O25—Dy1—O15^{ii}	101.9 (3)	O28—Dy2—O12	85.7 (4)
O20—Dy1—O16^{ii}	122.5 (3)	O3^{iii}—Dy2—O11	114.5 (2)
O26—Dy1—O16^{ii}	76.7 (3)	O7^{iv}—Dy2—O11	121.9 (3)
O24^{i}—Dy1—O16^{ii}	71.7 (3)	O23^{i}—Dy2—O11	80.0 (3)
O8—Dy1—O16^{ii}	76.4 (3)	O19—Dy2—O11	78.8 (3)
O11—Dy1—O16^{ii}	139.7 (3)	O27—Dy2—O11	155.0 (3)
O25—Dy1—O16^{ii}	139.5 (3)	O28—Dy2—O11	78.0 (3)

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

Table S4. Selected bond lengths [Å] and angles [°]for **2**.

bond			
Tb1—O8^{i}	2.279 (8)	Tb2—O23^{iii}	2.286 (9)
Tb1—O7^{ii}	2.329 (8)	Tb2—O3	2.313 (8)
Tb1—O4	2.338 (8)	Tb2—O20^{iv}	2.335 (8)
Tb1—O15	2.407 (9)	Tb2—O19	2.355 (9)
Tb1—O24^{iii}	2.402 (10)	Tb2—O11^{iv}	2.370 (10)
Tb1—O16	2.433 (9)	Tb2—O28	2.464 (8)
Tb1—O25	2.453 (9)	Tb2—O12^{iv}	2.470 (10)
Tb1—O26	2.500 (10)	Tb2—O27	2.570 (8)
angle			
O8^{i}—Tb1—O7^{ii}	95.8 (3)	O23^{iii}—Tb2—O3	87.8 (3)
O8^{i}—Tb1—O4	98.5 (3)	O23^{iii}—Tb2—O20^{iv}	139.3 (3)
O7^{ii}—Tb1—O4	140.4 (3)	O3—Tb2—O20^{iv}	89.2 (3)
O8^{i}—Tb1—O15	163.7 (3)	O23^{iii}—Tb2—O19	145.0 (3)
O7^{ii}—Tb1—O15	85.9 (4)	O3—Tb2—O19	94.9 (4)
O4—Tb1—O15	90.2 (3)	O20^{iv}—Tb2—O19	75.7 (3)
O8^{i}—Tb1—O24^{iii}	80.5 (3)	O23^{iii}—Tb2—O11^{iv}	86.0 (3)
O7^{ii}—Tb1—O24^{iii}	140.1 (3)	O3—Tb2—O11^{iv}	147.4 (3)
O4—Tb1—O24^{iii}	78.9 (3)	O20^{iv}—Tb2—O11^{iv}	75.3 (4)
O15—Tb1—O24^{iii}	87.7 (3)	O19—Tb2—O11^{iv}	108.3 (3)
O8^{i}—Tb1—O16	141.8 (3)	O23^{iii}—Tb2—O28	71.3 (3)
O7^{ii}—Tb1—O16	73.0 (3)	O3—Tb2—O28	83.5 (3)
O4—Tb1—O16	73.0 (3)	O20^{iv}—Tb2—O28	148.4 (3)
O15—Tb1—O16	54.1 (3)	O19—Tb2—O28	74.3 (3)
O24^{iii}—Tb1—O16	131.2 (3)	O11^{iv}—Tb2—O28	124.1 (4)
O8^{i}—Tb1—O25	81.0 (4)	O23^{iii}—Tb2—O12^{iv}	88.7 (4)
O7^{ii}—Tb1—O25	70.9 (3)	O3—Tb2—O12^{iv}	158.5 (3)
O4—Tb1—O25	147.9 (3)	O20^{iv}—Tb2—O12^{iv}	107.0 (4)
O15—Tb1—O25	84.3 (3)	O19—Tb2—O12^{iv}	76.3 (4)
O24^{iii}—Tb1—O25	69.3 (3)	O11^{iv}—Tb2—O12^{iv}	53.2 (3)
O16—Tb1—O25	125.8 (4)	O28—Tb2—O12^{iv}	75.3 (3)

O8^{i}—Tb1—O26	72.6 (3)	O23^{iii}—Tb2—O27	69.4 (3)
O7^{ii}—Tb1—O26	72.4 (3)	O3—Tb2—O27	77.2 (3)
O4—Tb1—O26	77.0 (3)	O20^{iv}—Tb2—O27	70.4 (3)
O15—Tb1—O26	123.1 (3)	O19—Tb2—O27	145.1 (3)
O24^{iii}—Tb1—O26	140.4 (3)	O11^{iv}—Tb2—O27	70.7 (3)
O16—Tb1—O26	69.2 (3)	O28—Tb2—O27	136.5 (3)

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

Table S5. Selected bond lengths [Å] and angles [°]for **3**.

bond			
Tb1—O2	2.300 (5)	Tb2—O19^{ii}	2.308 (7)
Tb1—O15	2.332 (5)	Tb2—O12^{iii}	2.300 (8)
Tb1—O16^{i}	2.329 (5)	Tb2—O33	2.336 (7)
Tb1—O28	2.389 (6)	Tb2—O31	2.365 (9)
Tb1—O25	2.413 (6)	Tb2—O24	2.397 (7)
Tb1—O26	2.417 (5)	Tb2—O30	2.407 (10)
Tb1—O29	2.443 (5)	Tb2—O32	2.450 (8)
Tb1—O27	2.466 (6)	Tb2—O23	2.482 (7)
angle			
O2—Tb1—O15	147.3 (2)	O19^{ii}—Tb2—O12^{iii}	103.6 (3)
O2—Tb1—O16^{i}	83.6 (2)	O19^{ii}—Tb2—O33	74.2 (2)
O15—Tb1—O16^{i}	106.2 (2)	O12^{iii}—Tb2—O33	79.4 (3)
O2—Tb1—O28	110.6 (2)	O19^{ii}—Tb2—O31	78.0 (3)
O15—Tb1—O28	79.4 (2)	O12^{iii}—Tb2—O31	76.6 (4)
O16^{i}—Tb1—O28	145.1 (2)	O33—Tb2—O31	137.5 (4)
O2—Tb1—O25	73.4 (2)	O19^{ii}—Tb2—O24	85.5 (3)
O15—Tb1—O25	80.6 (2)	O12^{iii}—Tb2—O24	153.4 (3)
O16^{i}—Tb1—O25	141.6 (2)	O33—Tb2—O24	79.2 (3)
O28—Tb1—O25	73.0 (2)	O31—Tb2—O24	130.0 (3)
O2—Tb1—O26	77.79 (19)	O19^{ii}—Tb2—O30	142.2 (4)
O15—Tb1—O26	75.5 (2)	O12^{iii}—Tb2—O30	100.2 (4)
O16^{i}—Tb1—O26	73.5 (2)	O33—Tb2—O30	139.6 (4)
O28—Tb1—O26	139.4 (2)	O31—Tb2—O30	79.5 (5)
O25—Tb1—O26	71.8 (2)	O24—Tb2—O30	86.1 (4)
O2—Tb1—O29	139.1 (2)	O19^{ii}—Tb2—O32	142.5 (3)
O15—Tb1—O29	73.1 (2)	O12^{iii}—Tb2—O32	76.1 (4)
O16^{i}—Tb1—O29	74.64 (18)	O33—Tb2—O32	68.9 (3)
O28—Tb1—O29	74.3 (2)	O31—Tb2—O32	135.7 (3)
O25—Tb1—O29	141.1 (2)	O24—Tb2—O32	81.5 (3)
O26—Tb1—O29	126.05 (19)	O30—Tb2—O32	71.8 (4)
O2—Tb1—O27	70.37 (19)	O19^{ii}—Tb2—O23	73.6 (3)
O15—Tb1—O27	141.6 (2)	O12^{iii}—Tb2—O23	153.1 (3)

O16^{i}—Tb1—O27	77.9 (2)	O33—Tb2—O23	123.6 (2)
O28—Tb1—O27	77.4 (2)	O31—Tb2—O23	76.7 (3)
O25—Tb1—O27	120.3 (2)	O24—Tb2—O23	53.3 (2)
O26—Tb1—O27	139.1 (2)	O30—Tb2—O23	71.9 (4)
O29—Tb1—O27	71.41 (19)	O32—Tb2—O23	122.7 (3)

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

Table S6. Selected bond lengths [Å] and angles [°]for **4**.

bond			
La1—O3^{i}	2.439 (3)	La1—O14	2.547 (4)
La1—O10^{ii}	2.485 (4)	La1—O11^{iv}	2.562 (4)
La1—O8	2.536 (4)	La1—O12^{iv}	2.598 (4)
La1—O8^{iii}	2.536 (4)	La1—O7	2.693 (4)
La1—O13	2.544 (4)	La1—O1^{v}	2.718 (4)
angle			
O3^{i}—La1—O10^{ii}	82.51 (14)	O8—La1—O12^{iv}	73.39 (13)
O3^{i}—La1—O8	82.40 (14)	O8^{iii}—La1—O12^{iv}	73.39 (13)
O10^{ii}—La1—O8	126.67 (13)	O13—La1—O12^{iv}	78.61 (13)
O3^{i}—La1—O8^{iii}	82.40 (14)	O14—La1—O12^{iv}	127.98 (12)
O10^{ii}—La1—O8^{iii}	126.67 (13)	O11^{iv}—La1—O12^{iv}	50.52 (11)
O8—La1—O8^{iii}	0.0 (2)	O3^{i}—La1—O7	69.96 (13)
O3^{i}—La1—O13	71.13 (13)	O10^{ii}—La1—O7	76.89 (13)
O10^{ii}—La1—O13	76.00 (13)	O8—La1—O7	49.90 (12)
O8—La1—O13	142.87 (14)	O8^{iii}—La1—O7	49.90 (12)
O8^{iii}—La1—O13	142.87 (14)	O13—La1—O7	134.75 (12)
O3^{i}—La1—O14	138.35 (13)	O14—La1—O7	71.17 (13)
O10^{ii}—La1—O14	74.66 (13)	O11^{iv}—La1—O7	114.71 (12)
O8—La1—O14	84.01 (14)	O12^{iv}—La1—O7	119.39 (13)
O8^{iii}—La1—O14	84.01 (14)	O3^{i}—La1—O1^{v}	139.32 (12)
O13—La1—O14	132.96 (13)	O10^{ii}—La1—O1^{v}	86.28 (12)
O3^{i}—La1—O11^{iv}	132.56 (13)	O8—La1—O1^{v}	133.67 (13)
O10^{ii}—La1—O11^{iv}	144.72 (12)	O8^{iii}—La1—O1^{v}	133.67 (13)
O8—La1—O11^{iv}	71.18 (13)	O13—La1—O1^{v}	68.22 (12)
O8^{iii}—La1—O11^{iv}	71.18 (13)	O14—La1—O1^{v}	73.93 (12)
O13—La1—O11^{iv}	108.38 (13)	O11^{iv}—La1—O1^{v}	64.74 (12)
O14—La1—O11^{iv}	78.09 (12)	O12^{iv}—La1—O1^{v}	88.79 (12)
O3^{i}—La1—O12^{iv}	84.84 (13)	O7—La1—O1^{v}	144.14 (12)
O10^{ii}—La1—O12^{iv}	154.15 (12)		

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

Equation S1:

$$\chi''(\upsilon_{ac}) = \frac{(\chi_0 - \chi_\infty)(2\pi\upsilon_{ac}\tau)^{1-\alpha}\cos(\alpha\pi/2)}{1 + 2(2\pi\upsilon_{ac}\tau)^{1-\alpha}\sin(\alpha\pi/2)(2\pi\upsilon_{ac}\tau)^{2(1-\alpha)}} \qquad \text{(S1)}$$

Comment: in this equation, χ_∞ is the adiabatic susceptibility extrapolated when $\nu_{ac}\rightarrow\infty$, χ_0 is the isothermal susceptibility extrapolated when $\nu_{ac}\rightarrow 0$, and τ is the average relaxation time of magnetization. The α parameter, which ranges between 0 and 1, quantifies the width of the τ distribution.[S2]

Table S7 Relaxation fitting parameters from least-squares fitting of $\chi''(\nu_{ac})$ data for a microcrystalline sample of complex **1b.**

(χ_0 was fixed as 0, $R = \Sigma(\chi_m''_{obs} - \chi_m''_{calc})^2 / \Sigma\chi_m''^2_{obs}$).

T/K	T^{-1}/K^{-1}	χ_∞/cm^3mol^{-1}	$\tau/10^{-7}$	$ln\tau$	α	*R*
1.9	0.53	1.70	10.043	-13.81	0.200	4.10×10^{-7}
2.0	0.50	1.78	8.426	-13.99	0.193	4.12×10^{-7}
2.2	0.45	1.82	6.780	-14.20	0.189	1.78×10^{-7}
2.4	0.42	1.72	5.853	-14.35	0.182	9.39×10^{-8}

Reference:

(S1) G. M. Sheldrick, SHELXTL-97, Program for the Solution of Crystal Structures, University of Göttingen, Germany **1997.**

(S2) (a) K. S. Cole, R. H. Cole, *J. Chem. Phys.* **1941**, *9*, 341; (b) S. M. Aubin, Z. Sun, L. Pardi, J. Krzysteck, K. Folting, L. J. Brunel, A. L. Rheingold, G. Christou, D. N, Hendrickson, *Inorg. Chem*. **1999**, *38*, 5329; (c) h. miyasaka, r. cleric, k. mizushima, k. sugiura, m. yamashita, w. wernsdorfer, c. coulon, *Inorg. Chem*. **2003**, *42*, 8203.

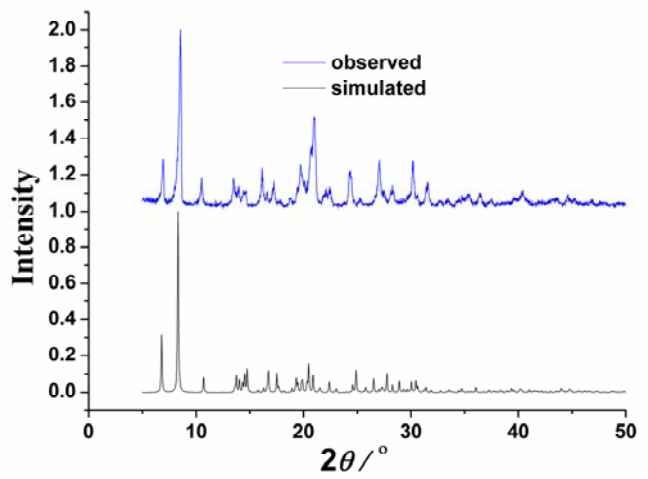

Figure S1. Powder XRD patterns of **metalloligand L**.

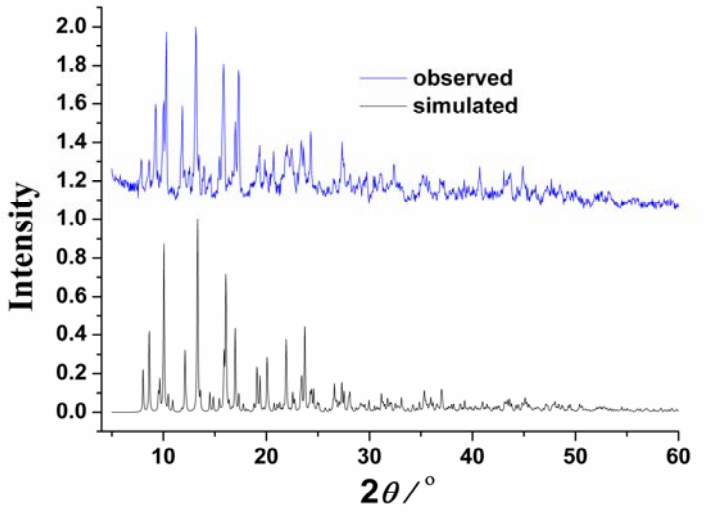

Figure S2. Powder XRD patterns of **1**.

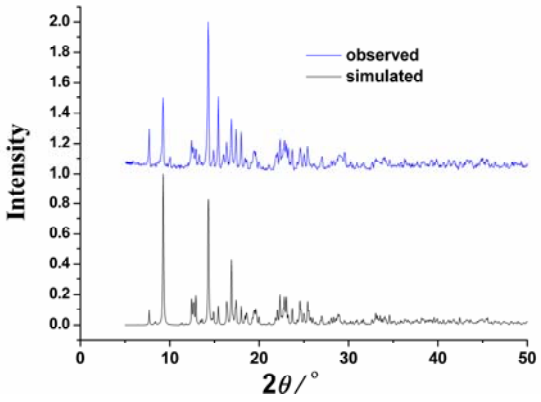

Figure S3. Powder XRD patterns of **2**.

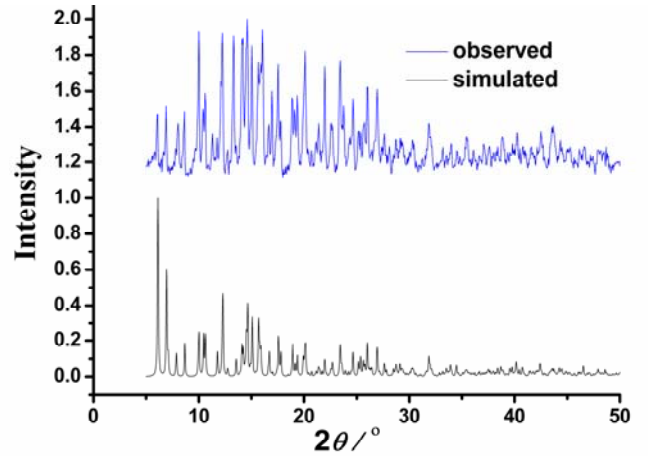

Figure S4. Powder XRD patterns of **3**.

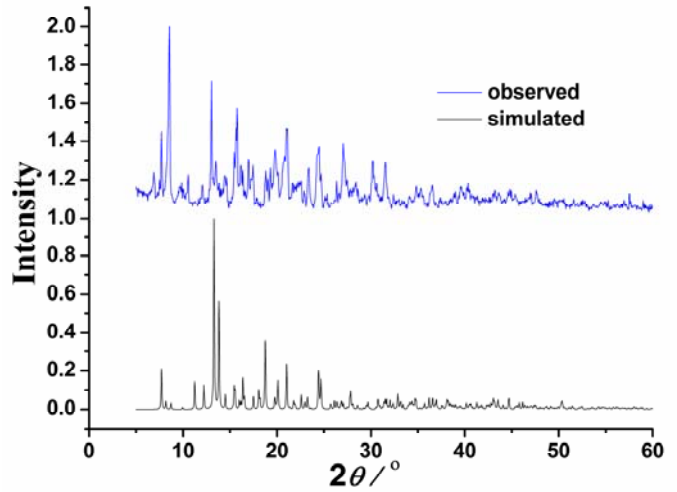

Figure S5. Powder XRD patterns of **4**.

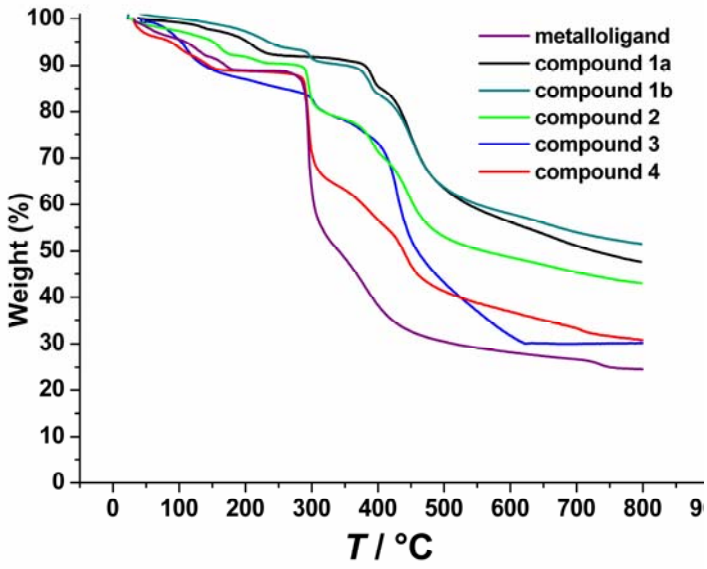

Figure S6.TGA Curves of metalloligand, **1**, **2**, **3**, **4**.

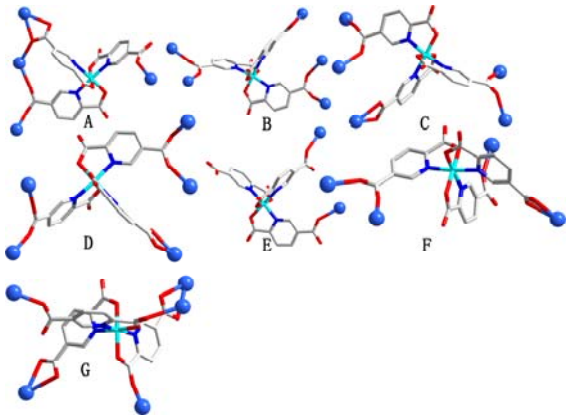

Figure S7. Coordination modes of metalloligand L. (A, B in **1**, C, D in **2**, E, F in **3**, and G in **4**)

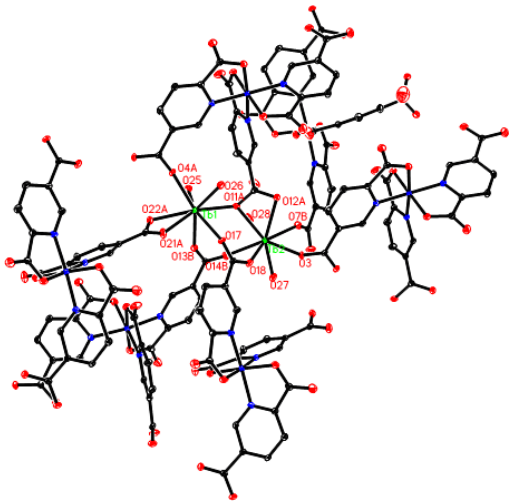

Figure S8. The coordination environments of Tb1 and Tb2 atoms in **1a**. Symmetry codes: A: x, y-1, z; B: 1–x, y-1/2, 3/2-z respectively.

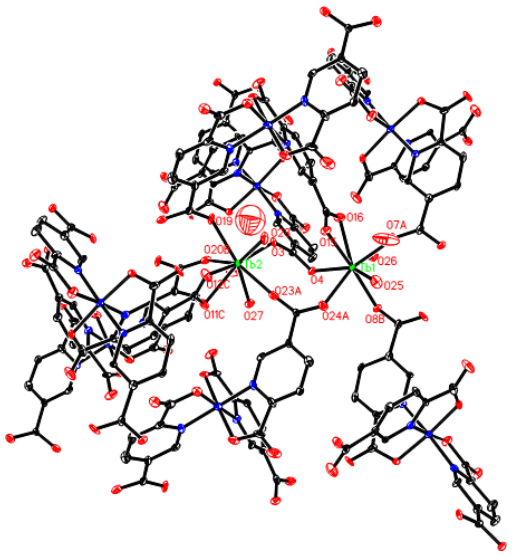

Figure S9. The coordination environments of Tb1 and Tb2 atoms in **2**. Symmetry codes: A:1+ x, y, z; B: 1+x, y-1, z; C: 1-x, 1-y, -z respectively.

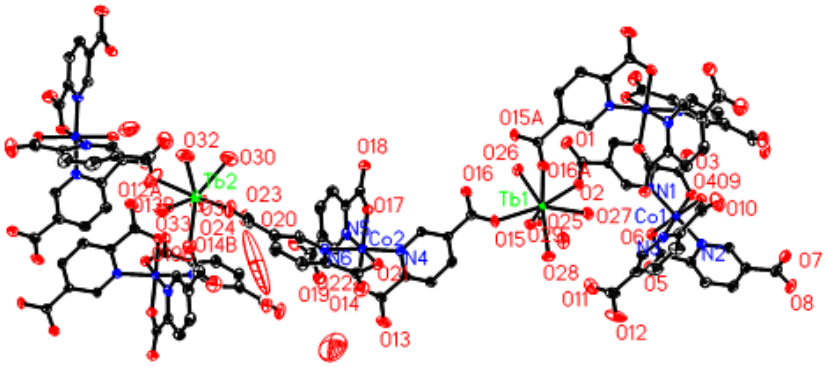

Figure S10. The coordination environments of Tb1 and Tb2 atoms in **3**. Symmetry codes: A: 1-x, 1-y, 1-z; B: 1-x, 2-y, 1-z respectively.

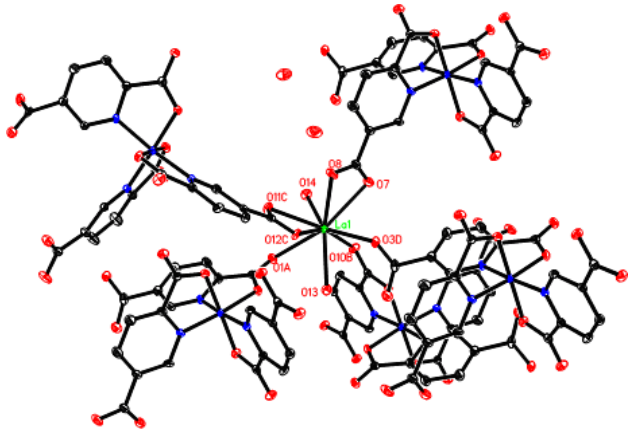

Figure S11. The coordination environments of La atom in **4**. Symmetry codes: A: x-1, y, z-1; B: 1-x, y, 1-z; C: x-1, 1/2-y, z-1/2; D: x, y, z-1; respectively.

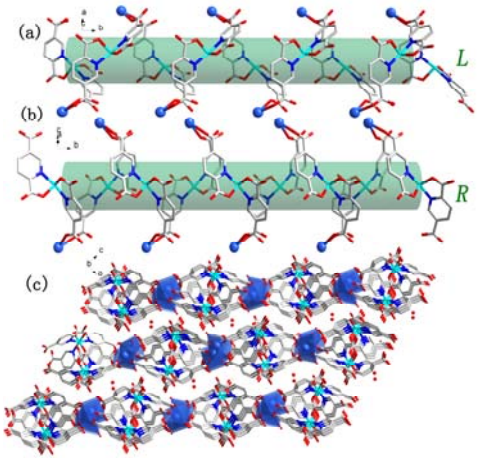

Figure S12. (a) The left handed helical chain formed by Co^{3+} ions and two 2,5-pydc moieties of L^A in **1**. (b) The right handed helical chain formed by Co^{3+} ions and two 2,5-pydc moieties of L^B in **1**. (c) The stacking fashion of **1**.

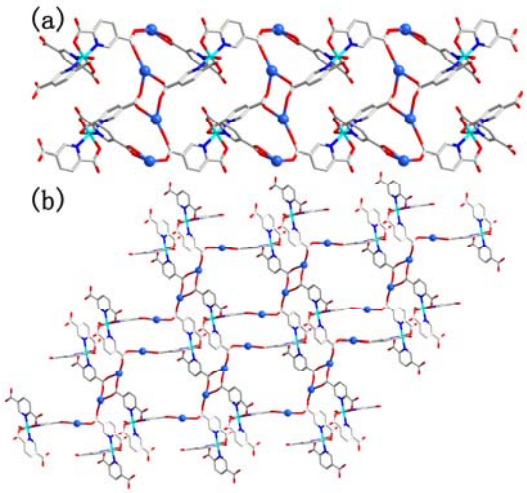

Figure S13. (a) The tape based upon linear $[Tb^{3+}]_4$ units in **2**. (b) The layer based upon linear $[Tb^{3+}]_4$ units in **2**.

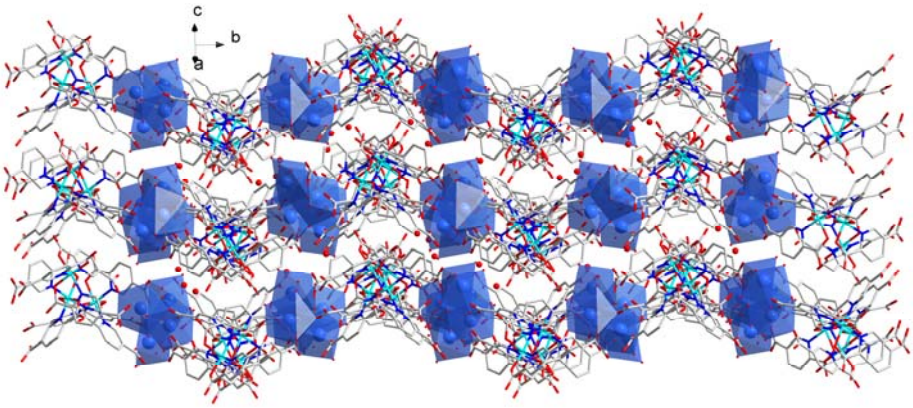

Figure S14. The stacking fashion of **3**.

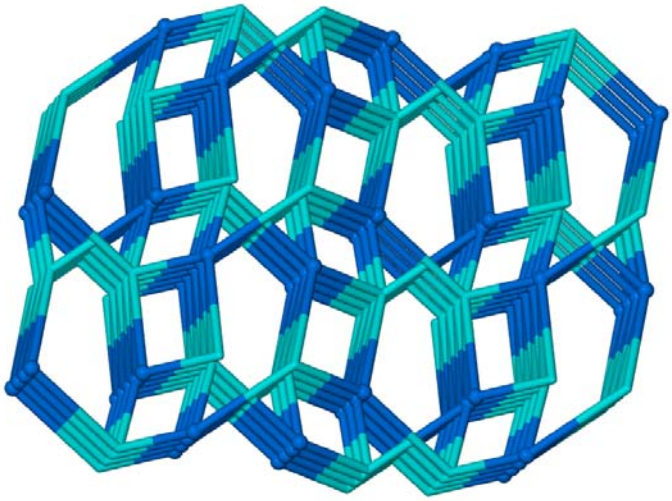

Figure S15. Topological view of the three-dimensional network of **4** with BN topology. La nodes are represented by blue balls and L^G nodes are represented by grin balls.

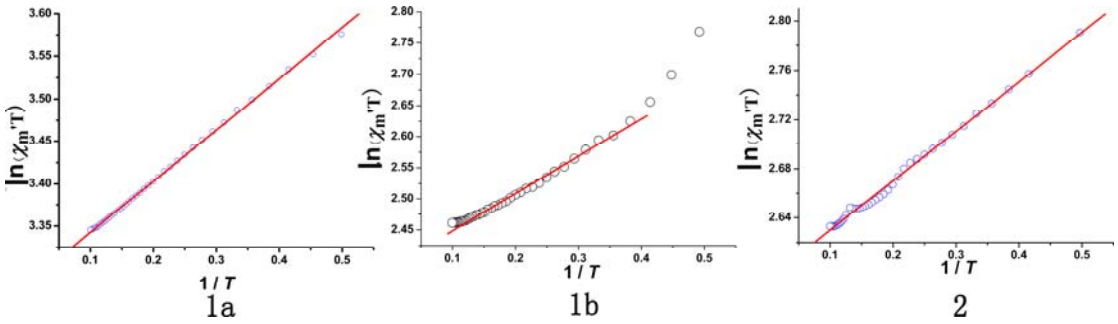

Figure S16. ln$\chi_m{}'T$ versus $1/T$ plots for **1a**, **1b** and **2**.

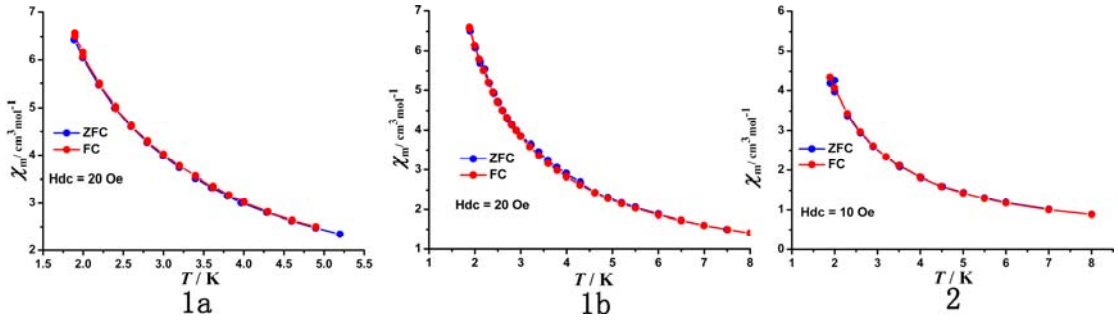

Figure S17. Field-cooled (FC) and zero-field-cooled (ZFC) magnetization for **1a**, **1b**, **2**.

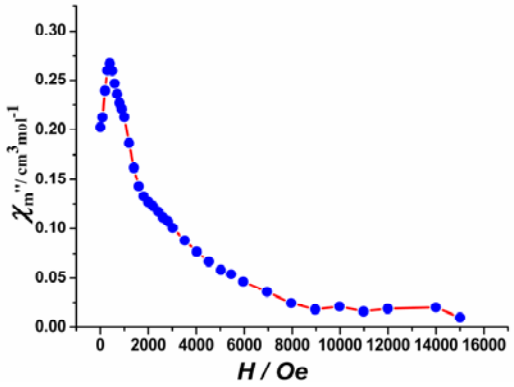

Figure S18. Field dependence of the product of the imaginary $\chi_m{}''$ component of the ac-susceptibility for **1b** at 2.0 K.

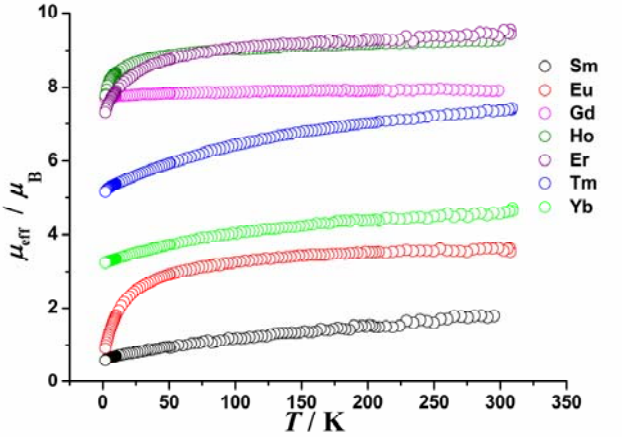

Figure S19. Temperature dependence of μ_{eff} for the Sm, Eu, Gd, Ho, Er, Tm, Yb, analogous to **1** at 1000 Oe.

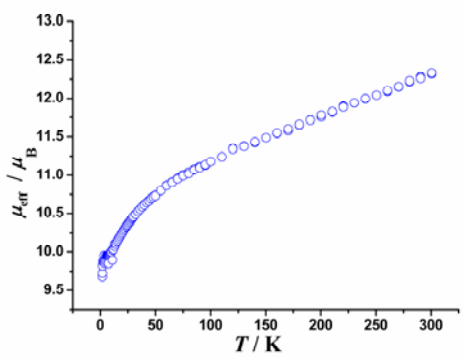

Figure S20. Temperature dependence of μ_{eff} for the Dy analogou to **2** at 100 Oe.