Supporting Information

Radii-Dependent Self-Assembly of Chiral Lanthanide Complexes: Synthetic, Chiroptical, and Single-molecule Magnet Behavior

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Table S1 Crystal Data and Structure Refinement Parameters for Complexes.

	Complex **1a**	Complex **1b**	Complex **2a**	Complex **2b**
Formula	$C_{104}H_{116}La_3N_{17}O_{34}$	$C_{104}H_{114}La_3N_{17}O_{34}$	$C_{108}H_{120}Dy_5N_{17}O_{36}$	$C_{112}H_{126}Dy_5N_{19}O_{36}$
Formula Weight	2564.86	2562.85	3044.70	3126.81
Crystal System	Orthorhombic	Orthorhombic	Tetragonal	Tetragonal
Space Group	$P2_12_12_1$	$P2_12_12_1$	$P4_22_12$	$P4_22_12$
a (Å)	14.6618(10)	14.6403(11)	28.2725(12)	28.2054(4)
b (Å)	26.2233 (10)	26.197(2)	28.2725(12)	28.2054(4)
c (Å)	29.3764(10)	29.383(2)	18.1870(7)	18.1449(3)
α (°)	90	90	90	90
β (°)	90	90	90	90
γ (°)	90	90	90	90
Z	4	4	4	4
Flack parameter	-0.0055(14)	0.002(4)	-0.009(9)	-0.003(3)
Volume (Å 3)	11294.66(10)	11269.5(15)	14537.5(14)	14435.1(5)
ρ_{calc} (g/cm^3)	1.508	1.511	1.391	1.439
Absorption coefficient (mm^{-1})	9.335	1.203	2.609	14.212
F(000)	5208	5200	6020	6196
Temperature	293 K	296.15 K	298 K	120 K
Reflections collected	142478	62056	31737	49240
Independent ref.	22207	22165	12403	14157
Data/restraints/parameters	22207/97/1470	22165/80/1461	12403/249/783	14157/201/811
R_{int}	0.0752	0.0348	0.0644	0.0538
Final R indices [I>2.0σ(I)]	R_1 = 0.0387, wR_2 = 0.1022	R_1 = 0.0327, wR_2 = 0.0812	R_1 = 0.0699, wR_2 = 0.1331	R_1 = 0.0587, wR_2 = 0.1466

Final R indexes [all data]	R_1 = 0.0398, wR_2 = 0.1028	R_1 = 0.0369, wR_2 = 0.0833	R_1 = 0.0984, wR_2 = 0.1484	R_1 = 0.0888, wR_2 = 0.1661
GOF	1.022	1.062	1.046	0.969

Table S2 Selected Bond Lengths (Å) and Angles (°) for All Complexes.

Complex **1a**					
La(1)-O(2)	2.428(4)	La(2)-O(5)	2.764(5)	La(3)-O(9)	2.616(5)
La(1)-O(3)	2.578(5)	La(2)-O(6)	2.466(4)	La(3)-O(10)	2.562(4)
La(1)-O(6)	2.470(4)	La(2)-O(10)	2.689(4)	La(3)-O(15)	2.618(4)
La(1)-O(7)	2.588(5)	La(2)-O(11)	2.591(4)	La(3)-O(16)	2.597(5)
La(1)-O(11)	2.613(5)	La(2)-O(14)	2.665(5)	La(3)-O(18)	2.465(4)
La(1)-O(12)	2.672(4)	La(2)-O(15)	2.595(4)	La(3)-O(19)	2.585(4)
La(1)-O(13)	2.639(5)	La(2)-O(17)	2.748(4)	La(3)-O(22)	2.462(4)
La(1)-O(14)	2.574(5)	La(2)-O(18)	2.453(4)	La(3)-O(23)	2.599(4)
La(1)-N(1)	2.855(6)	La(2)-N(5)	2.637(6)	La(3)-N(9)	2.782(6)
La(1)-N(3)	2.804(5)	La(2)-N(8)	2.613(6)	La(3)-N(11)	2.809(6)
O(2)-La(1)-O(3)	111.62(16)	O(6)-La(2)-O(5)	59.32(14)	O(9)-La(3)-O(15)	110.51(14)
O(2)-La(1)-O(6)	168.40(17)	O(6)-La(2)-O(10)	138.75(14)	O(9)-La(3)-N(9)	144.14(16)
O(2)-La(1)-O(7)	70.97(18)	O(6)-La(2)-O(11)	71.88(14)	O(9)-La(3)-N(11)	86.87(15)
O(2)-La(1)-O(11)	110.05(15)	O(6)-La(2)-O(14)	65.67(14)	O(10)-La(3)-O(9)	61.20(14)
O(2)-La(1)-O(12)	57.97(15)	O(6)-La(2)-O(15)	120.78(14)	O(10)-La(3)-O(15)	61.28(14)
O(2)-La(1)-O(13)	75.74(17)	O(6)-La(2)-O(17)	73.42(14)	O(10)-La(3)-O(16)	78.06(15)
O(2)-La(1)-O(14)	124.17(16)	O(6)-La(2)-N(5)	132.29(15)	O(10)-La(3)-O(19)	137.96(14)
O(2)-La(1)-N(1)	61.37(16)	O(6)-La(2)-N(8)	123.82(17)	O(10)-La(3)-O(23)	80.97(14)
O(2)-La(1)-N(3)	107.58(16)	O(10)-La(2)-O(5)	118.24(13)	O(10)-La(3)-N(9)	129.15(15)
O(3)-La(1)-O(7)	128.08(16)	O(10)-La(2)-O(17)	74.75(14)	O(10)-La(3)-N(11)	136.37(15)
O(3)-La(1)-O(11)	136.76(15)	O(11)-La(2)-O(5)	124.17(14)	O(15)-La(3)-N(9)	102.47(15)
O(3)-La(1)-O(12)	141.17(15)	O(11)-La(2)-O(10)	116.04(14)	O(15)-La(3)-N(11)	161.43(15)
O(3)-La(1)-O(13)	69.28(15)	O(11)-La(2)-O(14)	61.08(14)	O(16)-La(3)-O(9)	73.07(16)
O(3)-La(1)-N(1)	57.57(16)	O(11)-La(2)-O(15)	165.73(13)	O(16)-La(3)-O(15)	59.97(14)
O(3)-La(1)-N(3)	73.92(16)	O(11)-La(2)-O(17)	64.72(13)	O(16)-La(3)-O(23)	139.92(15)
O(6)-La(1)-O(3)	70.13(15)	O(11)-La(2)-N(5)	60.86(15)	O(16)-La(3)-N(9)	138.60(16)
O(6)-La(1)-O(7)	98.71(16)	O(11)-La(2)-N(8)	107.45(15)	O(16)-La(3)-N(11)	122.63(15)
O(6)-La(1)-O(11)	71.45(14)	O(14)-La(2)-O(5)	74.66(14)	O(18)-La(3)-O(9)	116.81(15)
O(6)-La(1)-O(12)	128.25(14)	O(14)-La(2)-O(10)	155.24(14)	O(18)-La(3)-O(10)	67.86(14)
O(6)-La(1)-O(13)	114.87(15)	O(14)-La(2)-O(17)	119.26(14)	O(18)-La(3)-O(15)	70.84(14)
O(6)-La(1)-O(14)	67.06(14)	O(15)-La(2)-O(5)	64.59(13)	O(18)-La(3)-O(16)	129.29(14)
O(6)-La(1)-N(1)	113.67(15)	O(15)-La(2)-O(10)	59.93(13)	O(18)-La(3)-O(19)	100.08(14)

O(6)-La(1)-N(3)	61.38(15)	O(15)-La(2)-O(14)	116.05(14)	O(18)-La(3)-O(23)	70.63(14)
O(7)-La(1)-O(11)	76.75(15)	O(15)-La(2)-O(17)	123.10(14)	O(18)-La(3)-N(9)	61.36(15)
O(7)-La(1)-O(12)	86.34(15)	O(15)-La(2)-N(5)	106.93(15)	O(18)-La(3)-N(11)	107.87(15)
O(7)-La(1)-O(13)	146.37(15)	O(15)-La(2)-N(8)	60.82(15)	O(19)-La(3)-O(9)	142.89(15)
O(7)-La(1)-N(1)	85.42(16)	O(17)-La(2)-O(5)	119.07(13)	O(19)-La(3)-O(15)	76.68(14)
O(7)-La(1)-N(3)	57.54(15)	O(18)-La(2)-O(5)	72.74(14)	O(19)-La(3)-O(16)	80.81(15)
O(11)-La(1)-O(12)	59.66(13)	O(18)-La(2)-O(6)	75.37(14)	O(19)-La(3)-O(23)	134.88(15)
O(11)-La(1)-O(13)	111.22(14)	O(18)-La(2)-O(10)	66.00(14)	O(19)-La(3)-N(9)	58.04(15)
O(11)-La(1)-N(1)	162.09(15)	O(18)-La(2)-O(11)	120.69(13)	O(19)-La(3)-N(11)	85.49(15)
O(11)-La(1)-N(3)	103.93(14)	O(18)-La(2)-O(14)	138.17(14)	O(22)-La(3)-O(9)	73.24(15)
O(12)-La(1)-N(1)	118.08(15)	O(18)-La(2)-O(15)	71.44(14)	O(22)-La(3)-O(10)	125.84(14)
O(12)-La(1)-N(3)	143.68(15)	O(18)-La(2)-O(17)	59.16(13)	O(22)-La(3)-O(15)	115.91(15)
O(13)-La(1)-O(12)	71.89(15)	O(18)-La(2)-N(5)	123.26(16)	O(22)-La(3)-O(16)	61.19(15)
O(13)-La(1)-N(1)	82.84(16)	O(18)-La(2)-N(8)	131.85(15)	O(22)-La(3)-O(18)	166.16(14)
O(13)-La(1)-N(3)	141.13(15)	N(5)-La(2)-O(5)	160.06(15)	O(22)-La(3)-O(19)	71.18(15)
O(14)-La(1)-O(3)	85.06(16)	N(5)-La(2)-O(10)	65.72(15)	O(22)-La(3)-O(23)	107.50(15)
O(14)-La(1)-O(7)	138.68(16)	N(5)-La(2)-O(14)	95.03(15)	O(22)-La(3)-N(9)	104.85(16)
O(14)-La(1)-O(11)	62.01(14)	N(5)-La(2)-O(17)	80.83(15)	O(22)-La(3)-N(11)	61.63(16)
O(14)-La(1)-O(12)	75.56(14)	N(8)-La(2)-O(5)	81.35(16)	O(23)-La(3)-O(9)	66.91(15)
O(14)-La(1)-O(13)	60.77(15)	N(8)-La(2)-O(10)	93.64(16)	O(23)-La(3)-O(15)	133.70(14)
O(14)-La(1)-N(1)	135.88(16)	N(8)-La(2)-O(14)	66.29(16)	O(23)-La(3)-N(9)	80.35(15)
O(14)-La(1)-N(3)	128.23(15)	N(8)-La(2)-O(17)	159.39(16)	O(23)-La(3)-N(11)	58.32(15)

D—H···A	**Distance(D—H)**	**Distance(H···A)**	**Distance (D···A)**	**Angles(D—H···A)**
O(4)-H(4)...O(28)	0.82	1.86	2.68(2)	175.0
O(8)-H(8)...O(22) [i]	0.82	1.92	2.736(7)	172.7
O(20)-H(20)...N(6) [ii]	0.82	2.04	2.852(7)	172.0
O(24)-H(24)...O(33)	0.82	1.92	2.714(9)	164.0
N(10)-H(10)...O(31) [iii]	0.86	2.03	2.844(11)	157.1
O(25)-H(25B)...O(26)	0.83	2.05	2.838(12)	158.4
O(26)-H(26A)...O(35)	0.86	2.21	2.989(17)	151.4
O(27)-H(27A)...N(16) [iv]	0.90	2.04	2.821(19)	144.5
O(31)-H(31A)...O(27) [v]	0.85	2.27	2.802(17)	120.5
O(31)-H(31B)...O(32)	0.81	1.89	2.675(17)	161.8
O(32)-H(32D)...N(14) [iii]	0.88	1.96	2.81(3)	161.9
O(32)-H(32E)...N(7) [i]	0.90	1.89	2.785(10)	176.8
O(33)-H(33A)...O(35)	0.85	2.18	2.840(13)	134.0
O(34)-H(34A)...O(36)	0.85	1.78	2.47(4)	136.8

O(35)-H(35A)...O(28)^{v}	0.86	2.02	2.81(2)	153.0
O(36)-H(36A)...O(32)	0.80	2.11	2.88(2)	162.2

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

Complex **1b**

La(1)-O(2)	2.428(4)	La(2)-O(5)	2.774(4)	La(3)-O(9)	2.609(4)
La(1)-O(3)	2.589(4)	La(2)-O(6)	2.466(4)	La(3)-O(10)	2.554(4)
La(1)-O(6)	2.469(4)	La(2)-O(10)	2.690(4)	La(3)-O(15)	2.632(4)
La(1)-O(7)	2.600(4)	La(2)-O(11)	2.584(4)	La(3)-O(16)	2.600(4)
La(1)-O(11)	2.618(4)	La(2)-O(14)	2.668(4)	La(3)-O(18)	2.466(4)
La(1)-O(12)	2.670(4)	La(2)-O(15)	2.605(4)	La(3)-O(19)	2.590(4)
La(1)-O(13)	2.639(4)	La(2)-O(17)	2.747(4)	La(3)-O(22)	2.472(4)
La(1)-O(14)	2.582(4)	La(2)-O(18)	2.462(4)	La(3)-O(23)	2.598(4)
La(1)-N(1)	2.854(5)	La(2)-N(5)	2.637(5)	La(3)-N(9)	2.782(5)
La(1)-N(3)	2.804(5)	La(2)-N(8)	2.626(5)	La(3)-N(11)	2.801(5)

O(2)-La(1)-O(3)	111.84(15)	O(6)-La(2)-O(5)	59.28(12)	O(9)-La(3)-O(15)	110.75(13)
O(2)-La(1)-O(6)	168.18(15)	O(6)-La(2)-O(10)	138.84(13)	O(9)-La(3)-N(9)	144.77(14)
O(2)-La(1)-O(7)	70.76(16)	O(6)-La(2)-O(11)	71.64(13)	O(9)-La(3)-N(11)	86.86(14)
O(2)-La(1)-O(11)	110.28(14)	O(6)-La(2)-O(14)	65.84(13)	O(10)-La(3)-O(9)	61.71(13)
O(2)-La(1)-O(12)	58.21(13)	O(6)-La(2)-O(15)	120.96(13)	O(10)-La(3)-O(15)	61.43(12)
O(2)-La(1)-O(13)	75.46(15)	O(6)-La(2)-O(17)	73.01(13)	O(10)-La(3)-O(16)	77.99(13)
O(2)-La(1)-O(14)	124.23(14)	O(6)-La(2)-N(5)	131.96(14)	O(10)-La(3)-O(19)	137.36(13)
O(2)-La(1)-N(1)	61.36(15)	O(6)-La(2)-N(8)	123.66(15)	O(10)-La(3)-O(23)	80.71(13)
O(2)-La(1)-N(3)	107.15(15)	O(10)-La(2)-O(5)	118.02(12)	O(10)-La(3)-N(9)	129.34(13)
O(3)-La(1)-O(7)	128.60(14)	O(10)-La(2)-O(17)	75.03(13)	O(10)-La(3)-N(11)	136.24(14)
O(3)-La(1)-O(11)	136.36(13)	O(11)-La(2)-O(5)	124.21(12)	O(15)-La(3)-N(9)	102.04(13)
O(3)-La(1)-O(12)	141.13(14)	O(11)-La(2)-O(10)	116.09(12)	O(15)-La(3)-N(11)	161.25(14)
O(3)-La(1)-O(13)	69.74(14)	O(11)-La(2)-O(14)	61.18(13)	O(16)-La(3)-O(9)	72.70(15)
O(3)-La(1)-N(1)	57.64(15)	O(11)-La(2)-O(15)	165.70(12)	O(16)-La(3)-O(15)	59.85(13)
O(3)-La(1)-N(3)	74.37(15)	O(11)-La(2)-O(17)	64.78(12)	O(16)-La(3)-N(9)	138.08(15)
O(6)-La(1)-O(3)	70.19(14)	O(11)-La(2)-N(5)	60.74(14)	O(16)-La(3)-N(11)	123.20(14)
O(6)-La(1)-O(7)	98.66(14)	O(11)-La(2)-N(8)	107.67(14)	O(18)-La(3)-O(9)	117.77(13)
O(6)-La(1)-O(11)	71.01(13)	O(14)-La(2)-O(5)	75.08(13)	O(18)-La(3)-O(10)	68.10(13)
O(6)-La(1)-O(12)	127.95(13)	O(14)-La(2)-O(10)	155.00(12)	O(18)-La(3)-O(15)	70.85(12)
O(6)-La(1)-O(13)	115.46(13)	O(14)-La(2)-O(17)	119.35(12)	O(18)-La(3)-O(16)	129.22(13)
O(6)-La(1)-O(14)	67.14(13)	O(15)-La(2)-O(5)	64.57(12)	O(18)-La(3)-O(19)	100.03(13)
O(6)-La(1)-N(1)	113.83(14)	O(15)-La(2)-O(10)	60.03(13)	O(18)-La(3)-O(22)	165.93(13)

O(6)-La(1)-N(3)	61.65(14)	O(15)-La(2)-O(14)	115.73(13)	O(18)-La(3)-O(23)	70.65(13)
O(7)-La(1)-O(11)	76.45(13)	O(15)-La(2)-O(17)	123.27(13)	O(18)-La(3)-N(9)	61.28(13)
O(7)-La(1)-O(12)	86.06(14)	O(15)-La(2)-N(5)	107.08(14)	O(18)-La(3)-N(11)	107.35(14)
O(7)-La(1)-O(13)	145.78(14)	O(15)-La(2)-N(8)	60.53(14)	O(19)-La(3)-O(9)	141.90(13)
O(7)-La(1)-N(1)	85.94(14)	O(17)-La(2)-O(5)	118.23(12)	O(19)-La(3)-O(15)	75.94(13)
O(7)-La(1)-N(3)	57.46(14)	O(18)-La(2)-O(5)	72.15(13)	O(19)-La(3)-O(16)	80.10(14)
O(11)-La(1)-O(12)	59.80(12)	O(18)-La(2)-O(6)	75.58(13)	O(19)-La(3)-O(23)	135.86(14)
O(11)-La(1)-O(13)	111.18(13)	O(18)-La(2)-O(10)	65.97(12)	O(19)-La(3)-N(9)	58.23(14)
O(11)-La(1)-N(1)	162.33(14)	O(18)-La(2)-O(11)	120.86(13)	O(19)-La(3)-N(11)	86.22(14)
O(11)-La(1)-N(3)	103.58(14)	O(18)-La(2)-O(14)	138.38(13)	O(22)-La(3)-O(9)	72.57(14)
O(12)-La(1)-N(1)	118.21(14)	O(18)-La(2)-O(15)	71.39(13)	O(22)-La(3)-O(10)	125.85(13)
O(12)-La(1)-N(3)	143.33(14)	O(18)-La(2)-O(17)	59.15(13)	O(22)-La(3)-O(15)	115.86(13)
O(13)-La(1)-O(12)	71.41(13)	O(18)-La(2)-N(5)	123.42(14)	O(22)-La(3)-O(16)	61.32(13)
O(13)-La(1)-N(1)	82.70(14)	O(18)-La(2)-N(8)	131.47(14)	O(22)-La(3)-O(19)	71.11(14)
O(13)-La(1)-N(3)	141.81(14)	N(5)-La(2)-O(5)	160.56(14)	O(22)-La(3)-O(23)	107.67(14)
O(14)-La(1)-O(3)	85.00(14)	N(5)-La(2)-O(10)	65.88(14)	O(22)-La(3)-N(9)	104.69(14)
O(14)-La(1)-O(7)	138.28(14)	N(5)-La(2)-O(14)	94.79(14)	O(22)-La(3)-N(11)	62.06(15)
O(14)-La(1)-O(11)	61.88(13)	N(5)-La(2)-O(17)	81.17(14)	O(23)-La(3)-O(9)	67.27(14)
O(14)-La(1)-O(12)	75.28(13)	N(8)-La(2)-O(5)	81.53(15)	O(23)-La(3)-O(15)	133.58(13)
O(14)-La(1)-O(13)	60.96(13)	N(8)-La(2)-O(10)	93.72(14)	O(23)-La(3)-O(16)	139.86(14)
O(14)-La(1)-N(1)	135.76(14)	N(8)-La(2)-O(14)	66.00(14)	O(23)-La(3)-N(9)	81.02(14)
O(14)-La(1)-N(3)	128.60(14)	N(8)-La(2)-O(17)	160.04(15)	O(23)-La(3)-N(11)	58.30(14)

D—H···A	**Distance(D—H)**	**Distance(H···A)**	**Distance (D···A)**	**Angles(D—H···A)**
O(8)-H(8)...O(22)^{i}	0.82	1.92	2.738(6)	173.6
O(20)-H(20)...N(6)^{ii}	0.82	2.03	2.844(7)	174.0
O(24)-H(24)...O(33)	0.82	1.99	2.733(9)	149.8
N(10)-H(10)...O(31)^{iii}	0.86	2.00	2.811(9)	155.9
O(25)-H(25B)...O(26)	0.83	1.92	2.743(17)	169.7
O(26)-H(26A)...O(35)	0.85	2.03	2.83(2)	156.2
O(27)-H(27A)...N(16)^{iv}	0.87	2.17	2.790(17)	128.0
O(31)-H(31A)...O(27)^{v}	0.85	2.34	2.862(15)	120.5
O(31)-H(31B)...O(32)	0.81	1.95	2.738(13)	162.0
O(32)-H(32E)...N(7)^{i}	0.90	1.89	2.789(9)	176.2
O(33)-H(33A)...O(35)	0.85	2.13	2.775(15)	132.4
O(34)-H(34A)...O(36)	0.85	1.97	2.71(3)	144.4
O(35)-H(35A)...O(28)^{v}	0.86	1.98	2.80(2)	160.3
O(35)-H(35B)...O(34)^{iii}	0.85	2.26	2.87(2)	128.3

O(36)-H(36A)...O(32)	0.80	2.03	2.815(16)	166.3

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

Complex **2a**

Dy(1)-O(7)	2.380(12)	Dy(2)-O(11)	2.195(12)	Dy(3)-O(10)	2.342(13)
Dy(1)-O(2)	2.319(13)	Dy(2)-O(7)	2.411(13)	Dy(3)-O(10)	2.342(13)
Dy(1)-O(13)	2.378(12)	Dy(2)-O(10)	2.317(11)	Dy(3)-O(16)	2.323(12)
Dy(1)-O(6)	2.411(12)	Dy(2)-O(16)	2.253(13)	Dy(3)-O(16)	2.323(12)
Dy(1)-O(3)	2.232(13)	Dy(2)-O(2)	2.312(13)	Dy(3)-O(9)	2.220(13)
Dy(1)-O(17)	2.315(12)	Dy(2)-O(8)	2.454(15)	Dy(3)-O(9)	2.220(13)
Dy(1)-N(2)	2.503(16)	Dy(2)-O(1)	2.398(14)	Dy(3)-O(15)	2.448(14)
Dy(1)-N(4)	2.473(16)	Dy(2)-N(6)	2.496(16)	Dy(3)-O(15)	2.448(14)

O(7)-Dy(1)-O(6)	124.0(4)	O(11)-Dy(2)-O(7)	87.4(4)	O(10)-Dy(3)-O(10)	143.5(6)
O(7)-Dy(1)-N(2)	72.6(5)	O(11)-Dy(2)-O(10)	138.5(4)	O(10)-Dy(3)-O(15)	119.1(4)
O(7)-Dy(1)-N(4)	137.3(5)	O(11)-Dy(2)-O(16)	155.7(4)	O(10)-Dy(3)-O(15)	88.3(4)
O(2)-Dy(1)-O(7)	73.5(4)	O(11)-Dy(2)-O(2)	82.9(4)	O(10)-Dy(3)-O(15)	119.1(4)
O(2)-Dy(1)-O(13)	75.7(4)	O(11)-Dy(2)-O(8)	82.8(5)	O(10)-Dy(3)-O(15)	88.3(4)
O(2)-Dy(1)-O(6)	142.6(5)	O(11)-Dy(2)-O(1)	104.1(4)	O(16)-Dy(3)-O(10)	64.2(4)
O(2)-Dy(1)-N(2)	145.8(5)	O(11)-Dy(2)-N(6)	74.7(5)	O(16)-Dy(3)-O(10)	64.2(4)
O(2)-Dy(1)-N(4)	64.9(5)	O(7)-Dy(2)-O(8)	65.6(4)	O(16)-Dy(3)-O(10)	136.2(5)
O(13)-Dy(1)-O(7)	70.6(4)	O(7)-Dy(2)-N(6)	143.7(5)	O(16)-Dy(3)-O(10)	136.2(5)
O(13)-Dy(1)-O(6)	138.9(4)	O(10)-Dy(2)-O(7)	119.3(4)	O(16)-Dy(3)-O(16)	125.4(6)
O(13)-Dy(1)-N(2)	89.1(5)	O(10)-Dy(2)-O(8)	81.0(5)	O(16)-Dy(3)-O(15)	84.7(4)
O(13)-Dy(1)-N(4)	106.4(5)	O(10)-Dy(2)-O(1)	78.9(4)	O(16)-Dy(3)-O(15)	84.7(4)
O(6)-Dy(1)-N(2)	64.7(5)	O(10)-Dy(2)-N(6)	65.1(4)	O(16)-Dy(3)-O(15)	54.8(4)
O(6)-Dy(1)-N(4)	87.2(5)	O(16)-Dy(2)-O(7)	74.0(4)	O(16)-Dy(3)-O(15)	54.8(4)
O(3)-Dy(1)-O(7)	141.1(4)	O(16)-Dy(2)-O(10)	65.7(4)	O(9)-Dy(3)-O(10)	68.6(4)
O(3)-Dy(1)-O(2)	116.6(5)	O(16)-Dy(2)-O(2)	76.8(4)	O(9)-Dy(3)-O(10)	84.6(5)
O(3)-Dy(1)-O(13)	75.9(4)	O(16)-Dy(2)-O(8)	103.0(5)	O(9)-Dy(3)-O(10)	84.6(5)
O(3)-Dy(1)-O(6)	72.4(5)	O(16)-Dy(2)-O(1)	80.3(5)	O(9)-Dy(3)-O(10)	68.6(4)
O(3)-Dy(1)-O(17)	134.6(5)	O(16)-Dy(2)-N(6)	129.3(5)	O(9)-Dy(3)-O(16)	132.9(5)
O(3)-Dy(1)-N(2)	88.1(6)	O(2)-Dy(2)-O(7)	73.0(4)	O(9)-Dy(3)-O(16)	89.4(5)
O(3)-Dy(1)-N(4)	70.8(5)	O(2)-Dy(2)-O(10)	132.9(5)	O(9)-Dy(3)-O(16)	89.4(5)
O(17)-Dy(1)-O(7)	82.8(4)	O(2)-Dy(2)-O(8)	136.6(4)	O(9)-Dy(3)-O(16)	132.9(5)
O(17)-Dy(1)-O(2)	79.7(4)	O(2)-Dy(2)-O(1)	67.2(4)	O(9)-Dy(3)-O(9)	86.2(7)
O(17)-Dy(1)-O(13)	147.8(4)	O(2)-Dy(2)-N(6)	133.4(5)	O(9)-Dy(3)-O(15)	94.2(5)
O(17)-Dy(1)-O(6)	71.4(4)	O(8)-Dy(2)-N(6)	80.7(5)	O(9)-Dy(3)-O(15)	94.2(5)

O(17)-Dy(1)-N(2)	100.2(5)	O(1)-Dy(2)-O(7)	136.5(4)	O(9)-Dy(3)-O(15)	172.3(4)
O(17)-Dy(1)-N(4)	80.9(5)	O(1)-Dy(2)-O(8)	156.2(4)	O(9)-Dy(3)-O(15)	172.3(5)
N(4)-Dy(1)-N(2)	149.3(5)	O(1)-Dy(2)-N(6)	79.3(5)	O(15)-Dy(3)-O(15)	86.5(6)
O(7)-Dy(1)-O(6)	124.0(4)	O(11)-Dy(2)-O(7)	87.4(4)	O(10)-Dy(3)-O(10)	143.5(6)
O(7)-Dy(1)-N(2)	72.6(5)	O(11)-Dy(2)-O(10)	138.5(4)	O(10)-Dy(3)-O(15)	119.1(4)
O(7)-Dy(1)-N(4)	137.3(5)	O(11)-Dy(2)-O(16)	155.7(4)	O(10)-Dy(3)-O(15)	88.3(4)
O(2)-Dy(1)-O(7)	73.5(4)	O(11)-Dy(2)-O(2)	82.9(4)	O(10)-Dy(3)-O(15)	119.1(4)
O(2)-Dy(1)-O(13)	75.7(4)	O(11)-Dy(2)-O(8)	82.8(5)	O(10)-Dy(3)-O(15)	88.3(4)
O(2)-Dy(1)-O(6)	142.6(5)	O(11)-Dy(2)-O(1)	104.1(4)	O(16)-Dy(3)-O(10)	64.2(4)

D—H···A	**Distance(D—H)**	**Distance(H···A)**	**Distance (D···A)**	**Angles(D—H···A)**
O(13)-H(13A)...O(11)	0.87	2.25	3.061(18)	154.6
O(13)-H(13A)...O(12)	0.87	2.40	2.965(18)	123.3
O(13)-H(13B)...O(14)	0.87	1.86	2.665(14)	152.4
O(1)-H(1)...O(9)^{i}	0.861(14)	1.82(10)	2.489(17)	134(11)
O(5)-H(5)...N(1)^{iii}	0.80	2.17	2.95(3)	163.2
N(7)-H(7)...O(15)^{i}	0.98	1.66	2.63(2)	166.7
N(7)-H(7)...O(17)^{i}	0.98	2.47	3.14(2)	125.5
N(7)-H(7)...N(8)^{i}	0.98	2.31	3.22(3)	155.0
O(14)-H(14)...O(3)	0.86	2.08	2.681(13)	126.8
O(14)-H(14)...O(4)	0.86	2.39	3.21(3)	160.4

Symmetry codes: (i) *y*, *x*, −*z*−1; (ii) y,x,-z-2 (iii) −*x*−1, −*y*−1, *z*.

Complex **2b**

Dy(1)-O(7)	2.406(8)	Dy(2)-O(11)	2.204(7)	Dy(3)-O(10)	2.361(8)
Dy(1)-O(2)	2.336(7)	Dy(2)-O(7)	2.407(8)	Dy(3)-O(10)	2.361(8)
Dy(1)-O(13)	2.399(8)	Dy(2)-O(10)	2.329(7)	Dy(3)-O(16)	2.306(7)
Dy(1)-O(6)	2.412(8)	Dy(2)-O(16)	2.285(8)	Dy(3)-O(16)	2.306(7)
Dy(1)-O(3)	2.253(7)	Dy(2)-O(2)	2.331(9)	Dy(3)-O(9)	2.255(9)
Dy(1)-O(17)	2.327(7)	Dy(2)-O(8)	2.470(9)	Dy(3)-O(9)	2.255(9)
Dy(1)-N(2)	2.491(11)	Dy(2)-O(1)	2.427(9)	Dy(3)-O(15)	2.471(9)

O(7)-Dy(1)-O(6)	123.0(3)	O(11)-Dy(2)-O(7)	87.1(3)	O(10) -Dy(3)-O(10)	141.8(4)
O(7)-Dy(1)-N(2)	72.1(3)	O(11)-Dy(2)-O(10)	137.9(3)	O(10) -Dy(3)-O(15)	88.5(3)
O(7)-Dy(1)-N(4)	137.1(3)	O(11)-Dy(2)-O(16)	155.4(3)	O(10)-Dy(3)-O(15)	88.5(3)
O(2)-Dy(1)-O(7)	74.4(3)	O(11)-Dy(2)-O(2)	83.2(3)	O(10) -Dy(3)-O(15)	120.0(3)
O(2)-Dy(1)-O(13)	75.4(3)	O(11)-Dy(2)-O(8)	82.1(3)	O(10)-Dy(3)-O(15)	120.0(3)
O(2)-Dy(1)-O(6)	142.7(3)	O(11)-Dy(2)-O(1)	104.6(3)	O(16)-Dy(3)-O(10)	136.0(3)
O(2)-Dy(1)-N(2)	146.1(3)	O(11)-Dy(2)-N(6)	74.8(3)	O(16)-Dy(3)-O(10)	65.7(3)
O(2)-Dy(1)-N(4)	63.8(3)	O(7)-Dy(2)-O(8)	65.4(3)	O(16) -Dy(3)-O(10)	136.0(3)

O(13)-Dy(1)-O(7)	70.2(3)	O(7)-Dy(2)-O(1)	137.0(3)	O(16) -Dy(3)-O(10)	65.7(3)
O(13)-Dy(1)-O(6)	139.4(3)	O(7)-Dy(2)-N(6)	142.8(4)	O(16)-Dy(3)-O(16)	124.0(4)
O(13)-Dy(1)-N(2)	88.9(3)	O(10)-Dy(2)-O(7)	119.5(3)	O(16) -Dy(3)-O(15)	54.3(3)
O(13)-Dy(1)-N(4)	106.5(3)	O(10)-Dy(2)-O(2)	132.7(3)	O(16)-Dy(3)-O(15)	54.3(3)
O(6)-Dy(1)-N(2)	64.5(3)	O(10)-Dy(2)-O(8)	81.1(3)	O(16)-Dy(3)-O(15)	84.4(3)
O(6)-Dy(1)-N(4)	88.2(4)	O(10)-Dy(2)-O(1)	78.7(3)	O(16) -Dy(3)-O(15)	84.4(3)
O(3)-Dy(1)-O(7)	141.0(3)	O(10)-Dy(2)-N(6)	64.3(3)	O(9)-Dy(3)-O(10)	83.8(3)
O(3)-Dy(1)-O(2)	115.8(3)	O(16)-Dy(2)-O(7)	73.9(3)	O(9)-Dy(3)-O(10)	68.2(3)
O(3)-Dy(1)-O(13)	76.2(3)	O(16)-Dy(2)-O(10)	66.5(3)	O(9) -Dy(3)-O(10)	68.2(3)
O(3)-Dy(1)-O(6)	73.1(3)	O(16)-Dy(2)-O(2)	76.8(3)	O(9) -Dy(3)-O(10)	83.8(3)
O(3)-Dy(1)-O(17)	135.0(3)	O(16)-Dy(2)-O(8)	103.1(3)	O(9)-Dy(3)-O(16)	133.8(3)
O(3)-Dy(1)-N(2)	88.2(3)	O(16)-Dy(2)-O(1)	80.5(3)	O(9)-Dy(3)-O(16)	89.9(3)
O(3)-Dy(1)-N(4)	71.3(3)	O(16)-Dy(2)-N(6)	129.6(3)	O(9) -Dy(3)-O(16)	133.8(3)
O(17)-Dy(1)-O(7)	82.7(3)	O(2)-Dy(2)-O(7)	74.5(3)	O(9) -Dy(3)-O(16)	89.9(3)
O(17)-Dy(1)-O(2)	79.1(3)	O(2)-Dy(2)-O(8)	137.7(3)	O(9) -Dy(3)-O(9)	85.3(5)
O(17)-Dy(1)-O(13)	146.8(3)	O(2)-Dy(2)-O(1)	66.3(3)	O(9) -Dy(3)-O(15)	171.8(3)
O(17)-Dy(1)-O(6)	71.9(3)	O(2)-Dy(2)-N(6)	133.1(3)	O(9) -Dy(3)-O(15)	94.3(3)
O(17)-Dy(1)-N(2)	101.1(3)	O(8)-Dy(2)-N(6)	79.9(4)	O(9)-Dy(3)-O(15)	171.8(3)
O(17)-Dy(1)-N(4)	80.3(3)	O(1)-Dy(2)-O(8)	156.0(3)	O(9)-Dy(3)-O(15)	94.3(3)
N(4)-Dy(1)-N(2)	150.0(4)	O(1)-Dy(2)-N(6)	79.7(4)	O(15)-Dy(3)-O(15)	87.2(4)
O(7)-Dy(1)-O(6)	123.0(3)	O(11)-Dy(2)-O(7)	87.1(3)	O(10) -Dy(3)-O(10)	141.8(4)
O(7)-Dy(1)-N(2)	72.1(3)	O(11)-Dy(2)-O(10)	137.9(3)	O(10) -Dy(3)-O(15)	88.5(3)
O(7)-Dy(1)-N(4)	137.1(3)	O(11)-Dy(2)-O(16)	155.4(3)	O(10)-Dy(3)-O(15)	88.5(3)
O(2)-Dy(1)-O(7)	74.4(3)	O(11)-Dy(2)-O(2)	83.2(3)	O(10) -Dy(3)-O(15)	120.0(3)
O(2)-Dy(1)-O(13)	75.4(3)	O(11)-Dy(2)-O(8)	82.1(3)	O(10)-Dy(3)-O(15)	120.0(3)
O(2)-Dy(1)-O(6)	142.7(3)	O(11)-Dy(2)-O(1)	104.6(3)	O(16)-Dy(3)-O(10)	136.0(3)

D—H···A	**Distance(D—H)**	**Distance(H···A)**	**Distance (D···A)**	**Angles(D—H···A)**
O(13)-H(13A)...O(11)	0.87	2.21	3.012(12)	153.9
O(13)-H(13A)...O(12)	0.87	2.35	2.916(12)	122.9
O(13)-H(13B)...O(14)	0.87	1.87	2.668(9)	151.7
N(7)-H(7)...O(15)^{i}	1.00	1.65	2.632(16)	167.0
N(7)-H(7)...O(17)^{i}	1.00	2.44	3.158(14)	128.1
N(7)-H(7)...N(8)^{i}	1.00	2.33	3.273(19)	157.4
O(14)-H(14)...O(3)	0.86	2.06	2.661(9)	127.0
O(14)-H(14)...O(4)	0.86	2.38	3.209(17)	161.2

Symmetry code: (i) *y*, *x*, −*z*+1; (ii) y,x,-z+2

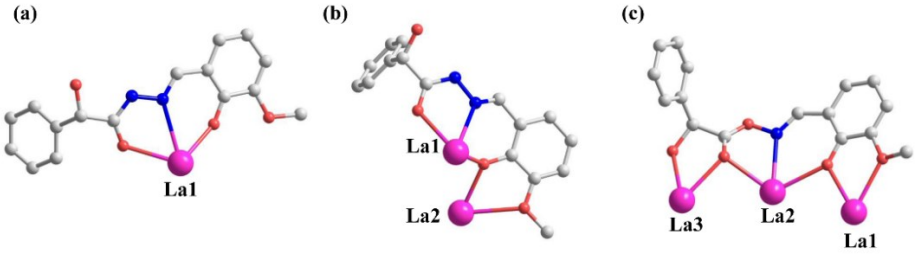

Figure S1 Coordination modes of ligands in complex **1a**.

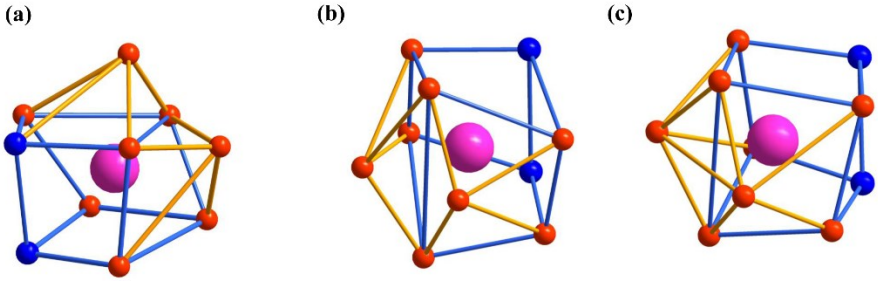

Figure S2 Coordination polyhedron of (a) La1, (b) La2 and (c) La3 in complex **1a**.

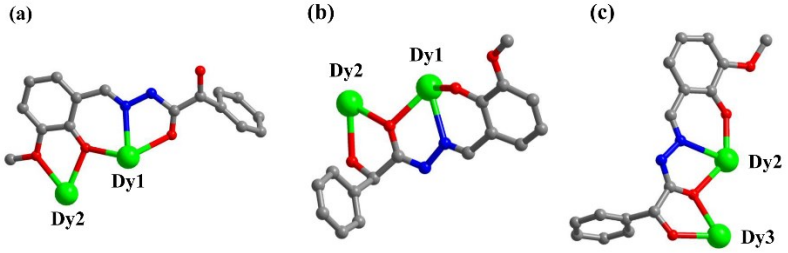

Figure S3 Coordination modes of ligands in complex **2a**.

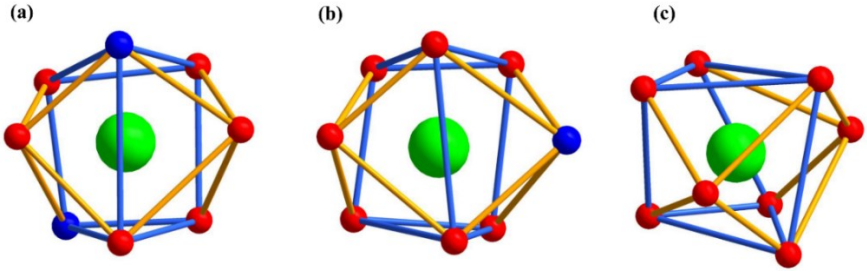

Figure S4 Coordination polyhedron of (a) La1, (b) La2 and (c) La3 in complex **2a**.

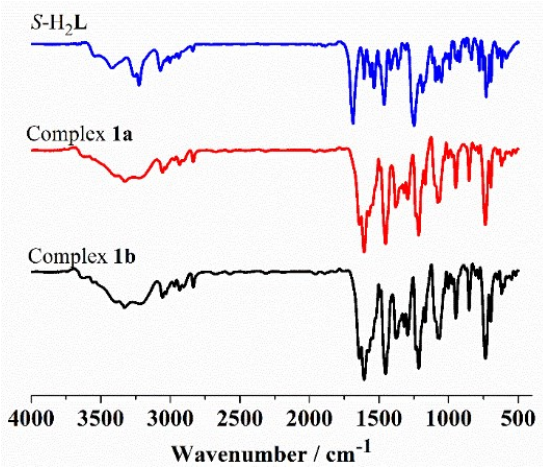

Figure S5 IR spectrum of complex **1a** and **1b**.

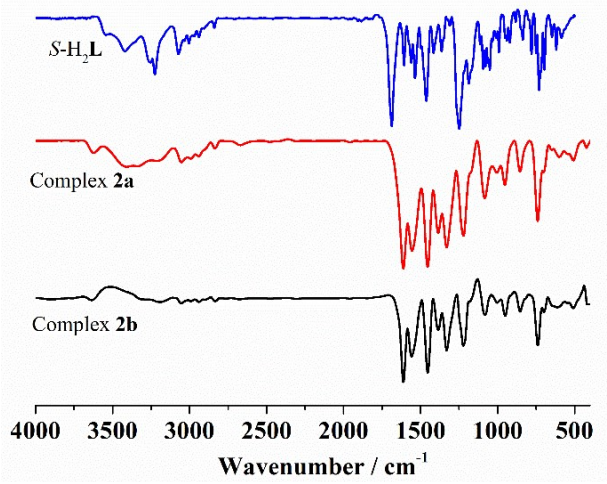

Figure S6 IR spectrum of complex **2a** and **2b**.

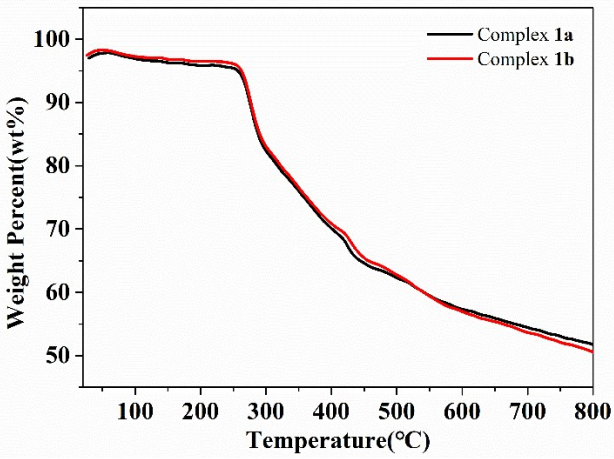

Figure S7 TGA spectra of complex **1a** and **1b.**

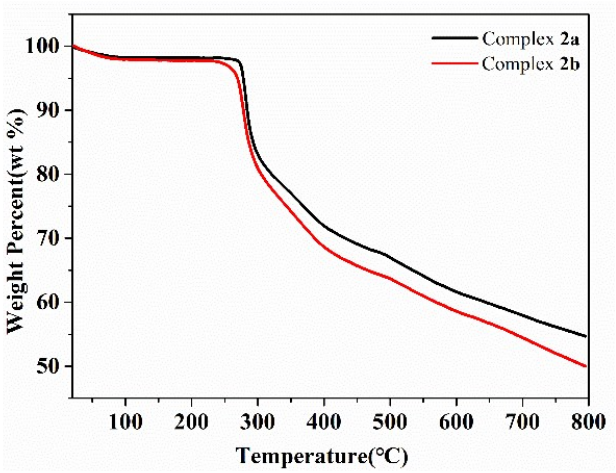

Figure S8 TGA spectra of complex **2a** and **2b.**

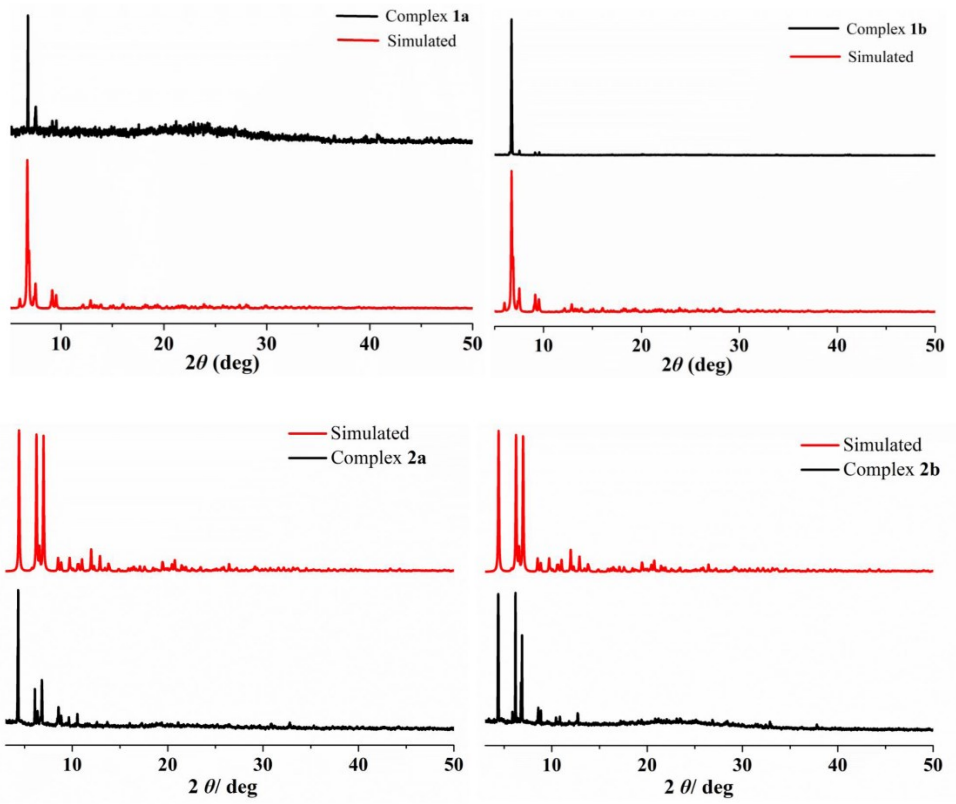

Figure S9 Comparing the simulated PXRD with experimental patterns of complexes.

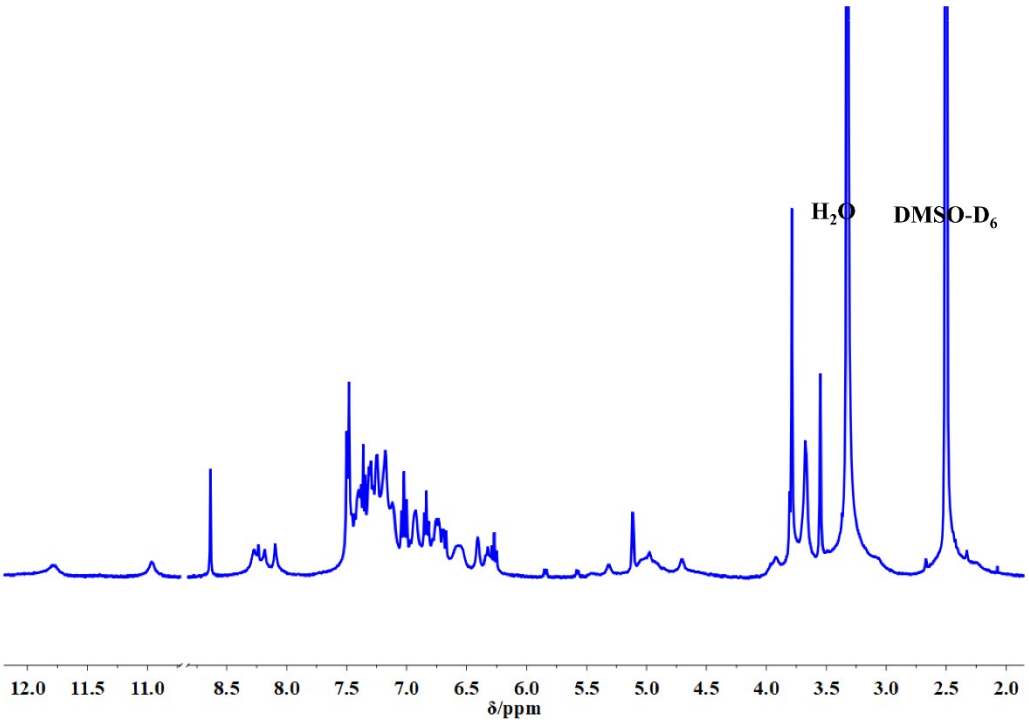

Figure S10 ^{1}H-NMR spectrum (400 MHz, DMSO-d_6) of **1a**.

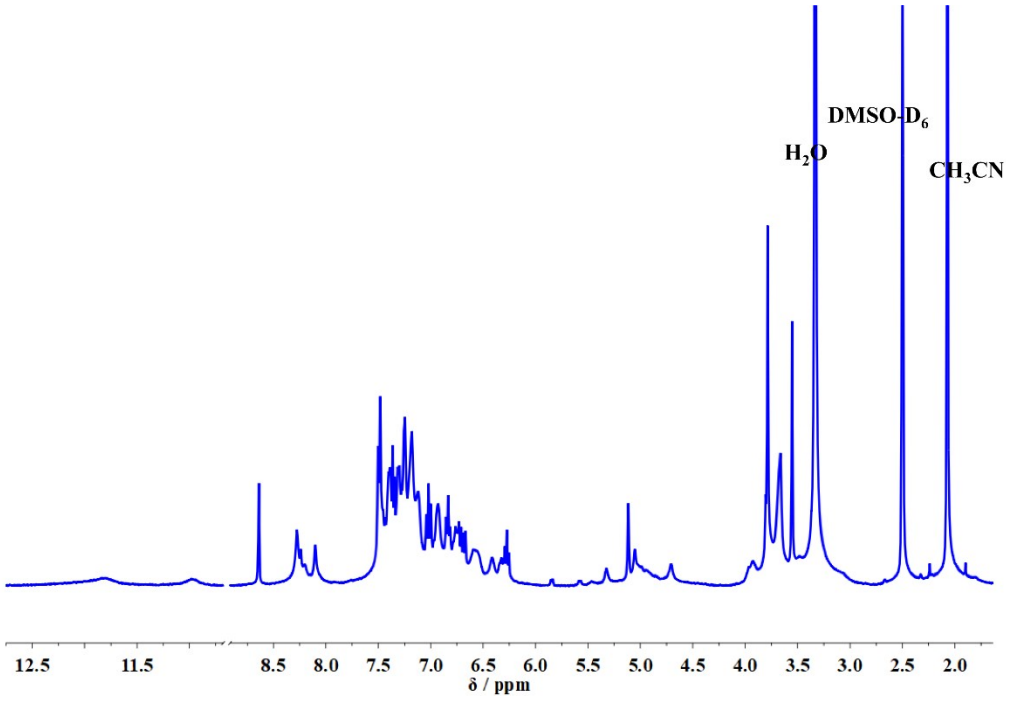

Figure S11 ^{1}H-NMR spectrum (400 MHz, DMSO-d_6) of **1b**.

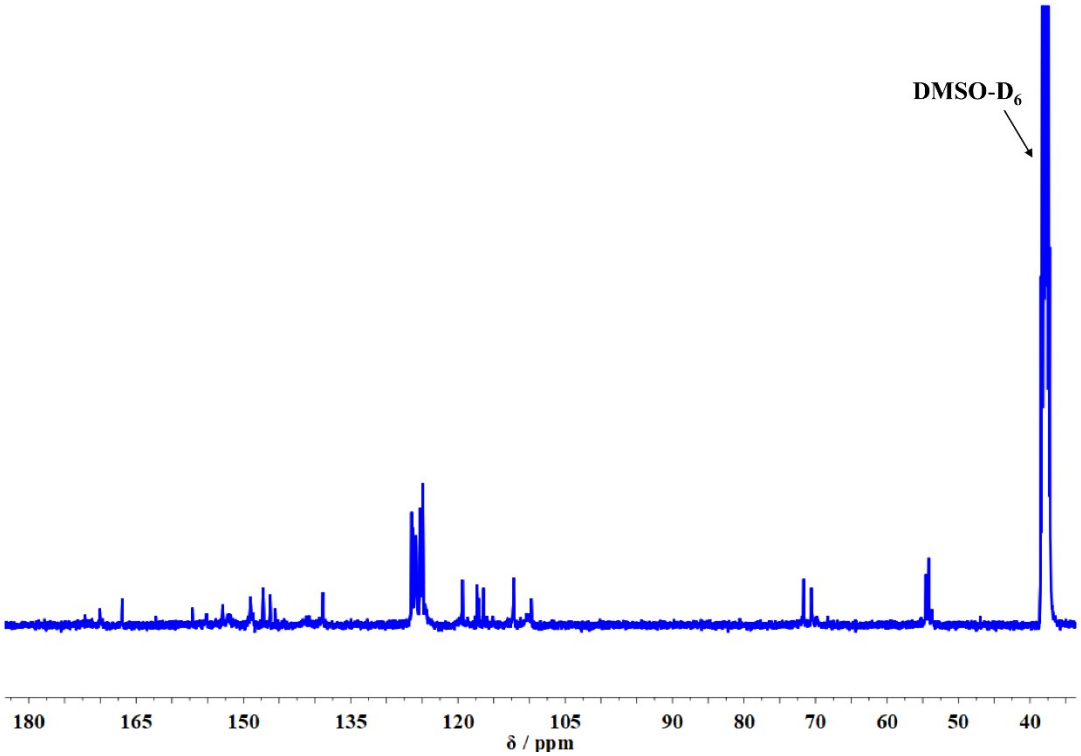

Figure S12 ^{13}C-NMR spectrum (400 MHz, DMSO-d_6) of **1a**.

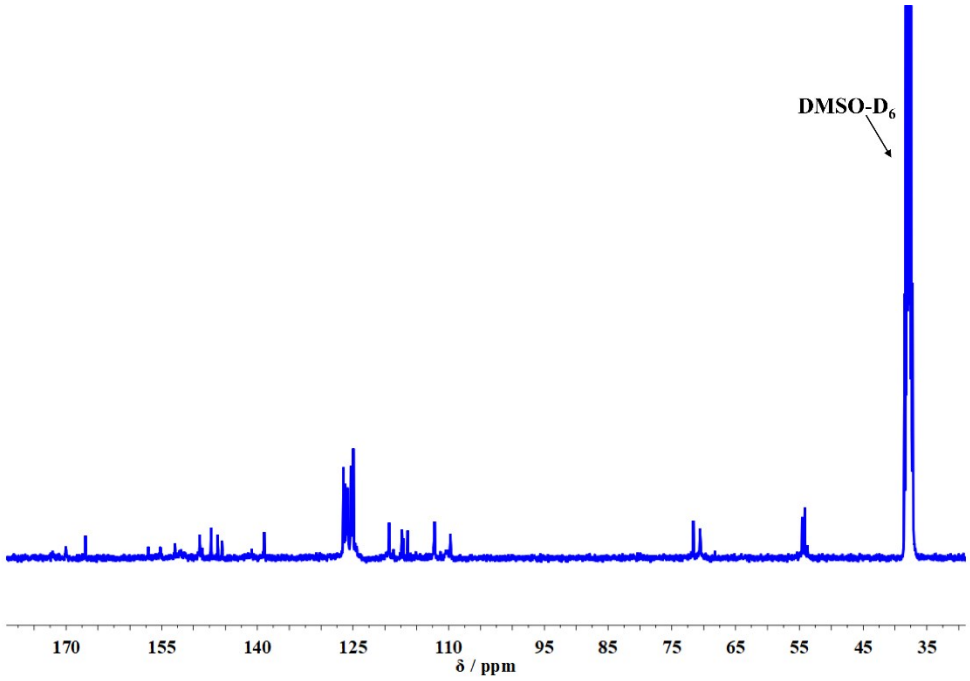

Figure S13 ^{13}C-NMR spectrum (400 MHz, DMSO-d_6) of **1b**.

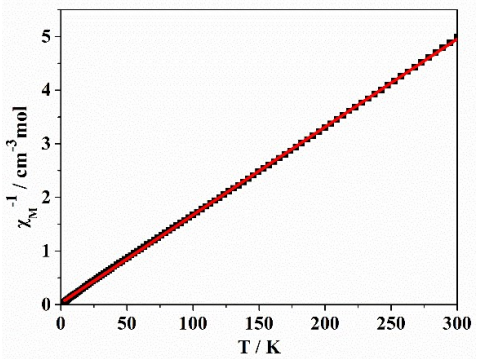

Figure S14 The temperature dependence of χ_M^{-1} for **2a**. The red line represents the best fit with Curie-Weiss law.

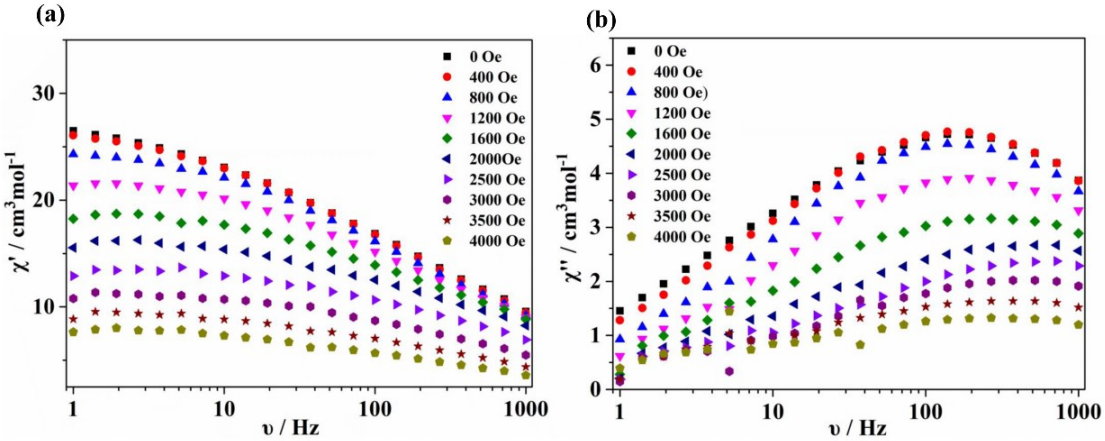

Figure S15 Frequency dependence of the (a) in-phase (χ′) and (b) out-of-phase (χ″) magnetic susceptibilities of complex **2a** measured at 2.0 K in various applied fields from 0 to 4000 Oe.

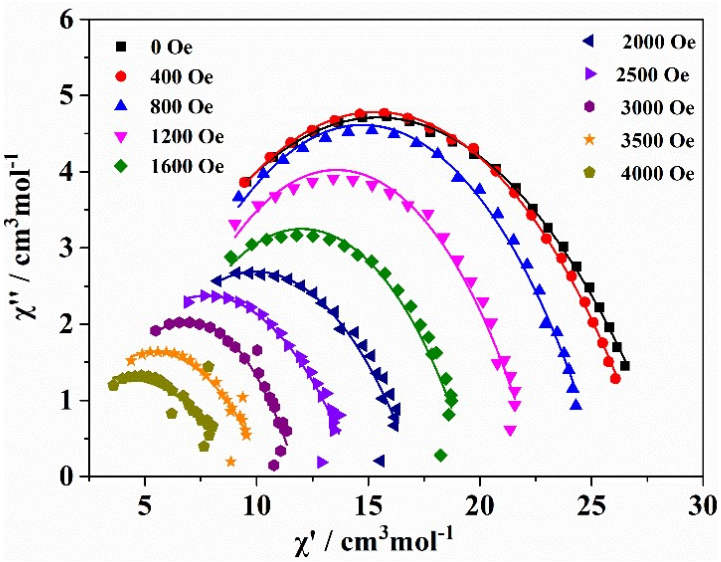

Figure S16 Cole-Cole plots of **2a** at various applied fields from 0 to 4000 Oe.

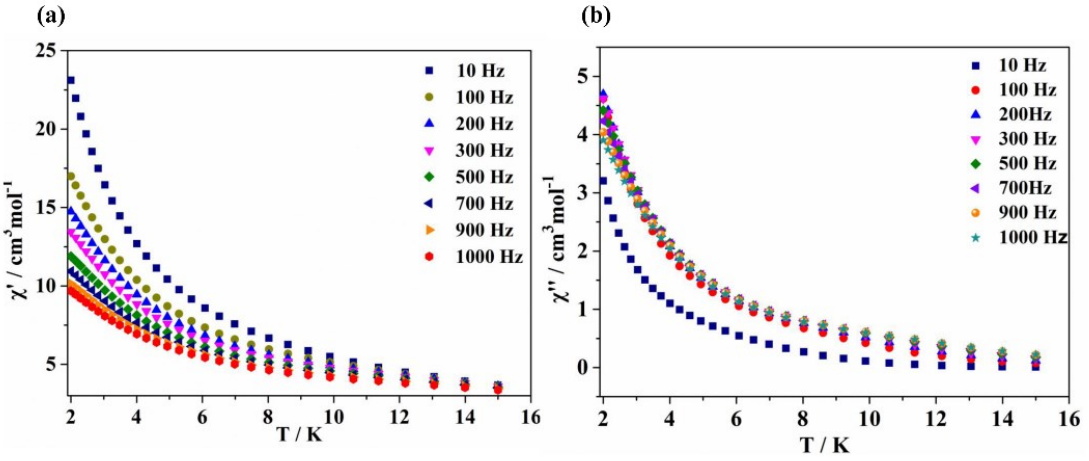

Figure S17 Temperature dependence of ac susceptibility data for **2a** collected under 0 Oe dc field over the frequency from 1 to 999 Hz.

Table S3 Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **2a** under 2.0 K-15.0 K according to the generalized Debye model.

T/K	χ_S	χ_T	τ	α
2.0	2.14E+00	2.85E+01	9.68E-04	5.65E-01
2.2	2.13E+00	2.60E+01	7.92E-04	5.60E-01
2.4	2.29E+00	2.38E+01	6.87E-04	5.52E-01
2.6	2.29E+00	2.20E+01	6.09E-04	5.51E-01
2.8	2.41E+00	2.04E+01	5.60E-04	5.47E-01
3.0	2.47E+00	1.91E+01	5.20E-04	5.45E-01
3.5	2.63E+00	1.63E+01	4.58E-04	5.42E-01
4.0	2.85E+00	1.42E+01	4.35E-04	5.39E-01
4.5	3.02E+00	1.26E+01	4.25E-04	5.37E-01
5.0	3.27E+00	1.13E+01	4.40E-04	5.25E-01
5.5	3.40E+00	1.02E+01	4.48E-04	5.15E-01
6.0	3.50E+00	9.36E+00	4.56E-04	5.02E-01
6.5	3.54E+00	8.61E+00	4.51E-04	4.85E-01
7.0	3.56E+00	7.97E+00	4.40E-04	4.67E-01
7.5	3.58E+00	7.40E+00	4.26E-04	4.43E-01
8.0	3.52E+00	6.93E+00	3.90E-04	4.29E-01
8.5	3.49E+00	6.51E+00	3.56E-04	4.10E-01
9.0	3.45E+00	6.13E+00	3.23E-04	3.88E-01
9.5	3.41E+00	5.81E+00	2.87E-04	3.73E-01
10	3.36E+00	5.51E+00	2.54E-04	3.56E-01
11	3.29E+00	5.00E+00	2.05E-04	3.20E-01
12	3.25E+00	4.59E+00	1.73E-04	2.82E-01
13	3.21E+00	4.24E+00	1.54E-04	2.46E-01
14	3.18E+00	3.94E+00	1.50E-04	2.06E-01
15	3.12E+00	3.68E+00	1.51E-04	1.64E-01