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Synthesis, Structures and Magnetic Properties of Chiral 3d-3d'-4f Heterobimetallic Complexes Based on $[(Tp^*)Fe(CN)_3]^-$

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Support Information

Table S1 Summary of crystallographic data for all complexes.

	1-RR	**1-SS**	**2-RR**	**2-SS**	**3-RR**	**3-SS**
formula	$C_{60}H_{92}B_2CuGdFe_2N_{21}O_{17}$	$C_{60}H_{92}B_2CuGdFe_2N_{21}O_{17}$	$C_{60}H_{92}B_2CuTbFe_2N_{21}O_{17}$	$C_{60}H_{92}B_2CuTbFe_2N_{21}O_{17}$	$C_{60}H_{92}B_2CuDyFe_2N_{21}O_{17}$	$C_{60}H_{92}B_2CuDyFe_2N_{21}O_{17}$
fw	1733.61	1733.61	1735.29	1735.29	1738.86	1738.86
crystal system	Monoclinic	Monoclinic	Monoclinic	Monoclinic	Monoclinic	Monoclinic
space group	$P2_1$	$P2_1$	$P2_1$	$P2_1$	$P2_1$	$P2_1$
a, Å	13.4352(9)	13.5975(11)	13.559(7)	13.5669(13)	13.585(3)	13.4347(11)
b, Å	16.0651(11)	16.1208(13)	16.024(9)	16.1004(16)	16.218(3)	16.0670(13)
c, Å	17.6194(12)	17.9767(15)	17.767(10)	17.8246(18)	18.401(4)	17.6509(14)
a, deg	90	90	90	90	90	90
β, deg	103.5480(10)	103.4320(10)	103.229(9)	103.2710(10)	103.848(4)	103.5890(10)
γ, deg	90	90	90	90	90	90
V, Å^3	3697.1(4)	3832.8(5)	3758(4)	3789.5(6)	3936.3(14)	3703.4(5)
Z	2	2	2	2	2	2
ρ_{calcd}, g cm^{-3}	1.419	1.369	1.397	1.384	1.337	1.421
T / K	296(2)	296(2)	296(2)	296(2)	296(2)	296(2)
μ, mm^{-1}	1.619	1.561	1.651	1.637	1.627	1.729
θ, deg	1.725 to 26.00	1.718 to 26.00	1.177 to 26.00	1.726 to 26.00	1.544 to 26.00	1.724 to 27.54

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$F(000)$	1608	1608	1608	1606	1612	1612
index ranges	-16<=h<=16 -19<=k<=19 -21<=l<=10	-16<=h<=14 -19<=k<=16 -18<=l<=22	−16 ≤ h ≤ 10 −19 ≤ k ≤ 19 −19 ≤ l ≤ 21	-16<=h<=11 -19<=k<=16 -21<=l<=21	−16 ≤ h ≤ 14 −18 ≤ k ≤ 20 −17 ≤ l ≤ 22	−16 ≤ h ≤ 17 −18 ≤ k ≤ 20 −21 ≤ l ≤ 22
data/restraints /parameters	13678/ 2 / 871	12347/ 3 / 871	14555/ 1/ 871	12803 / 4 / 871	14503 / 1/871	13795 / 1/ 870
GOF (F^2)	0.982	0.951	0.934	0.939	0.909	0.941
$R_1{}^a$,$wR_2{}^b$	0.0421	0.0457	0.0418	0.0484	0.0450	0.941
(I>2σ(I))	0.0773	0.1038	0.0752	0.1099	0.0807	0.0630
$R_1{}^a$, $wR_2{}^b$	0.0537	0.0618	0.0642	0.0702	0.0676	0.0379
(all data)	0.0813	0.1097	0.0813	0.1179	0.0880	0.0647
Flack χ	0.045(11)	-0.009(16)	-0.012(11)	0.013(15)	0.035(11)	0.006(6)

$R_1{}^a = \Sigma||F_o| - |F_c||/\Sigma F_o|$. $wR_2{}^b = [\Sigma w(F_o{}^2 - F_c{}^2)^2/\Sigma w(F_o{}^2)^2]^1$

Table S2 Selected bond lengths (Å) and angles (°) for **1-RR**.

Cu1-O2	1.909(6)	Cu1-O3	1.931(5)
Cu1-N10	1.917(6)	Cu1-N11	1.922(7)
Cu1-N9	2.582(7)	Gd1-O1	2.597(5)
Gd1-O2	2.362 (5)	Gd1-O3	2.357 (6)
Gd1-O4	2.578(5)	Gd1-O5	2.513 (5)
Gd1-O6	2.515(5)	Gd1-O1W	2.383(5)
Gd1-O2W	2.372 (5)	Gd1-O3W	2.369(6)
Cu1-Gd1	3.413 (1)	Fe1-N1	2.019 (6)
Fe1-N3	1.989(7)	Fe1-N5	1.981 (8)
Fe1-C16	1.921(9)	Fe1-C17	1.921 (9)
Fe1-C18	1.924 (7)	Fe2-N12	1.989 (6)
Fe2-N14	1.999(7)	Fe2-N16	1.986 (7)
Fe2-C56	1.943 (9)	Fe2-C57	1.936 (8)
Fe2-C58	1.909(8)	O1-Gd1-O2	61.48 (2)
O1-Gd1-O3	126.96(2)	O1-Gd1-O4	155.24(1)
O1-Gd1-O5	67.94 (2)	O1-Gd1-O6	116.93(1)
O1-Gd1-O1W	100.81 (2)	O1-Gd1-O2W	84.79 (2)
O1-Gd1-O3W	78.12(1)	O2-Gd1-O3	65.64 (2)
O2-Gd1-O4	123.47(2)	O2-Gd1-O5	113.93(2)
O2-Gd1-O6	153.16(2)	O2-Gd1-O1W	79.41(2)
O2-Gd1-O2W	73.34 (2)	O2-Gd1-O3W	128.14 (2)
O3-Gd1-O4	62.21(2)	O3-Gd1-O5	143.12 (2)
O3-Gd1-O6	111.78(2)	O3-Gd1-O1W	72.73 (2)
O3-Gd1-O2W	76.62 (2)	O3-Gd1-O3W	137.33 (2)
O5-Gd1-O6	50.75 (2)	O5-Gd1-O1W	71.15 (2)
O5-Gd1-O2W	140.09 (2)	O5-Gd1-O3W	74.32 (2)
O6-Gd1-O1W	74.62(2)	O6-Gd1-O2W	133.25(2)
O6-Gd1-O3W	73.17 (2)	O2W-Gd1-O3W	71.85(2)

O2-Cu1-O3	83.56 (2)	O2-Cu1-N10	95.78 (2)
O2-Cu1-N11	170.44(3)	O2-Cu1-N9	96.64(2)
O3-Cu1-N9	88.92 (2)	O3-Cu1-N10	178.54 (2)
O3-Cu1-N11	94.04(2)	N9-Cu-N10	92.45(2)
N9-Cu-N11	92.56 (2)	N10-Cu-N11	86.40(3)
N1-Fe1-N3	89.28 (3)	N1-Fe1-N5	90.38(3)
N3-Fe1-N5	88.55 (3)	C16-Fe1-C17	90.36 (4)
C16-Fe1-C18	85.70(4)	C17-Fe1-C18	85.62(3)
N7-C16-Fe1	177.63 (9)	N9-C18-Fe1	175.09 (7)
N8-C17-Fe1	177.78(8)	N12-Fe2-N14	89.21(3)
N12-Fe2-N16	88.85(3)	N14-Fe2-N16	89.10(3)
C56-Fe2-C57	84.82(4)	C56-Fe2-C58	85.54(3)
C57-Fe1-C58	85.60(3)	N18-C56-Fe2	175.34(7)
N19-C57-Fe2	176.22(8)	N20-C58-Fe2	178.05 (7)

Table S3 Selected bond lengths (Å) and angles (°) for **1-SS**.

Cu1-O2	1.925(5)	Cu1-O3	1.916(6)
Cu1-N9	2.638(9)	Cu1-N19	1.926(9)
Cu1-N20	1.922(6)	Gd1-O1	2.591(6)
Gd1-O2	2.366(6)	Gd1-O3	2.374(5)
Gd1-O4	2.605(7)	Gd1-O5	2.512(6)
Gd1-O6	2.518(6)	Gd1-O1W	2.440(7)
Gd1-O2W	2.397(5)	Gd1-O3W	2.415(8)
Cu1-Gd1	3.419(1)	Fe1-N2	2.009(7)
Fe1-N4	2.004(8)	Fe1-N6	1.999(9)
Fe1-C16	1.963(1)	Fe1-C17	1.932(1)
Fe1-C18	1.947(9)	Fe2-N10	1.980(9)
Fe2-N12	2.005(8)	Fe2-N14	2.011(7)
Fe2-C34	1.922(1)	Fe2-C35	1.892(1)
Fe2-C36	1.952(1)	O1-Gd1-O2	62.19(2)
O1-Gd1-O3	123.79 (2)	O1-Gd1-O4	156.28(2)
O1-Gd1-O5	119.74(2)	O1-Gd1-O6	69.92(2)
O1-Gd1-O1W	103.6(22)	O1-Gd1-O2W	75.69 (2)
O1-Gd1-O3W	82.77(2)	O2-Gd1-O3	65.57(2)
O2-Gd1-O4	126.38(2)	O2-Gd1-O5	144.17 (2)
O2-Gd1-O6	112.22(2)	O2-Gd1-O1W	72.75 (2)
O2-Gd1-O2W	76.78 (2)	O2-Gd1-O3W	136.65 (2)
O3-Gd1-O4	60.97(2)	O3-Gd1-O5	114.38 (2)
O3-Gd1-O6	153.25(2)	O3-Gd1-O1W	78.77 (2)
O3-Gd1-O2W	73.69 (2)	O3-Gd1-O3W	126.78 (2)
O4-Gd1-O5	68.02(2)	O4-Gd1-O6	117.04(2)
O4-Gd1-O1W	100.09(2)	O4-Gd1-O2W	84.76(2)
O4-Gd1-O3W	77.96(2)	O5-Gd1-O6	50.50(2)

O5-Gd1-O1W	72.30(2)	O5-Gd1-O2W	138.94(2)
O5-Gd1-O3W	74.46(2)	O6-Gd1-O1W	75.39(2)
O6-Gd1-O2W	132.86(2)	O6-Gd1-O3W	74.45(2)
O1W-Gd1-O2W	145.27 (2)	O1W-Gd1-O3W	144.67 (2)
O2W-Gd1-O3W	70.06 (2)	O2-Cu-O3	83.85 (2)
O2-Cu-N9	89.28(3)	O2-Cu-N19	93.25 (3)
O2-Cu-N20	177.86 (2)	O3-Cu-N9	96.95(2)
O3-Cu- N19	169.68 (3)	O3-Cu- N20	95.59 (3)
N9-Cu-N19	92.90(3)	N9-Cu-N20	92.84(3)
N19-Cu-N20	86.93 (3)	N2-Fe1-N4	88.73(3)
N2-Fe1-N6	90.05(3)	N4-Fe1-N6	88.02 (3)
C16-Fe1-C17	92.37(5)	C16-Fe1-C18	85.83(4)
C17-Fe1-C18	85.24(4)	N7-C16-Fe1	179.50(1)
N8-C17-Fe1	179.64(9)	N9-C18-Fe1	176.83 (9)
C34-Fe2-C36	87.01(48)	N16-C34-Fe2	177.49(1)
N17-C35-Fe2	174.97(1)	N18-C36-Fe2	175.78(1)

Table S4 Selected bond lengths (Å) and angles (°) for **2-RR**.

Cu1-O2	1.887 (6)	Cu1-O3	1.909 (5)
Cu1-N19	1.921(6)	Cu1-N20	1.913(8)
Cu1-N17	2.632(7)	Tb1-O1	2.585(5)
Tb 1-O2	2.355(5)	Tb 1-O3	2.349 (5)
Tb 1-O4	2.571(5)	Tb 1-O6	2.487(5)
Tb 1-O7	2.486(6)	Tb 1-O1W	2.360(6)
Tb 1-O2W	2.373(5)	Tb 1-O3W	2.333(6)
Cu1-Tb1	3.394(2)	Fe2-N10	1.983(7)
Fe2-N12	1.987(8)	Fe2-N14	1.993(6)
Fe2-C34	1.894(9)	Fe2-C35	1.917 (7)
Fe2-C36	1.930(9)	Fe1-N1	1.996(8)
Fe1-N3	1.983(8)	Fe1-N5	1.996(6)
Fe1-C16	1.916(1)	Fe1-C17	1.919 (9)
Fe1-C18	1.907(9)	O1-Tb1-O2	62.02(2)
O1-Tb1-O3	127.04(2)	O1-Tb1-O4	155.67 (1)
O1-Tb1-O6	116.52(2)	O1-Tb1-O7	67.42 (2)
O1-Tb1-O1W	100.35(2)	O1- Tb1-O2W	84.58(2)
O1- Tb1-O3W	77.45(2)	O2-Tb1-O3	65.17(2)
O2-Tb1-O4	123.59(2)	O2-Tb1-O6	152.68 (2)
O2-Tb1-O7	113.87(2)	O2-Tb1-O1W	78.19 (2)
O2-Tb1-O2W	73.37(2)	O2-Tb1-O3W	127.61 (2)
O3-Tb1-O4	62.49(2)	O3-Tb1-O6	111.96 (2)
O3-Tb1-O7	143.31(2)	O3-Tb1-O1W	72.27(2)
O3-Tb1-O2W	76.90(2)	O3-Tb1-O3W	137.25(2)
O4-Tb1-O6	70.14 (1)	O4-Tb1-O7	120.27 (2)

O4-Tb1-O1W	103.97 (2)	O4-Tb1-O2W	75.84(2)
O4-Tb1-O3W	82.55(2)	O6-Tb1-O7	50.79(2)
O6-Tb1-O1W	75.32(2)	O6-Tb1-O2W	133.69(2)
O6-Tb1-O3W	74.26(2)	O7-Tb1-O1W	71.83 (2)
O7-Tb1-O2W	139.56(2)	O7-Tb1-O3W	74.75(2)
O1W-Tb1-O2W	144.63(2)	O1W-Tb1-O3W	144.40(2)
O2W-Tb1-O3W	70.96(2)	O2-Cu-O3	83.71(2)
O2-Cu-N17	97.37(2)	O2-Cu-N19	96.03(3)
O2-Cu-N20	169.62(3)	O3-Cu-N17	89.44(2)
O3-Cu-N19	178.04(2)	O3-Cu-N20	93.81 (3)
N17-Cu-N19	92.52(3)	N17-Cu-N20	92.69(3)
N19-Cu-N20	86.04(3)	N10-Fe2-N12	88.09 (3)
N10-Fe2-N14	88.92(3)	N12-Fe2-N14	91.24(3)
C34-Fe2-C35	85.55(4)	C34-Fe2-C36	90.62 (4)
C35-Fe2-C36	85.41(4)	N16-C34-Fe2	179.15(8)
N17-C35-Fe2	176.81(8)	N18-C36-Fe2	178.76(9)
C17-Fe1-C18	86.11(4)	N7-C16-Fe1	176.03(9)
N8-C17-Fe1	176.83(9)	N9-C18-Fe1	178.99(8)

Table S5 Selected bond lengths (Å) and angles (°) for **2-SS**.

Cu1-O1	1.903(73)	Cu1-O2	1.934(6)
Cu1-N17	2.635(9)	Cu1-N19	1.924(9)
Cu1-N20	1.958(1)	Tb1-O1	2.359(6)
Tb1-O2	2.359(7)	Tb1-O3	2.578(9)
Tb1-O4	2.582(2)	Tb1-O6	2.501(8)
Tb1-O7	2.493(7)	Tb1-O1W	2.377(7)
Tb1-O2W	2.379 (6)	Tb1-O3W	2.357(8)
Cu1-Tb1	3.396(1)	Fe3-N1	1.989(1)
Fe3-N3	2.006(1)	Fe3-N5	2.009(9)
Fe3-C16	1.910(1)	Fe3-C17	1.925(1)
Fe3-C18	1.937(1)	Fe4-N10	1.979(9)
Fe4-N12	2.001(9)	Fe4-N14	1.988(9)
Fe4-C34	1.964(1)	Fe4-C35	1.917(9)
Fe4-C36	1.895(1)	O1-Tb1-O2	66.23(2)
O1-Tb1-O3	124.18(2)	O1-Tb1-O4	61.51(2)
O1-Tb1-O6	114.17(3)	O1-Tb1-O7	152.93(2)
O1-Tb1-O1W	78.40(2)	O1-Tb1-O2W	73.44(2)
O1-Tb1-O3W	127.48(2)	O2-Tb1-O3	61.94(2)
O2-Tb1-O4	127.56(2)	O2-Tb1-O6	143.78(2)
O2-Tb1-O7	111.56(2)	O2-Tb1-O1W	72.92(2)
O2-Tb1-O2W	76.34(2)	O2-Tb1-O3W	137.19(2)
O3-Tb1-O4	155.89(2)	O3-Tb1-O6	119.50(3)
O3-Tb1-O7	69.75(2)	O3-Tb1-O1W	104.09(2)

O3-Tb1-O2W	75.67(2)	O3-Tb1-O3W	82.81(2)
O4-Tb1-O6	67.77(3)	O4-Tb1-O7	116.62(2)
O4-Tb1-O1W	100.02(2)	O4-Tb1-O2W	85.07(2)
O4-Tb1-O3W	77.39(2)	O6-Tb1-O7	50.44(3)
O6-Tb1-O1W	71.92(3)	O6-Tb1-O2W	139.79(3)
O6-Tb1-O3W	73.71(2)	O7-Tb1-O1W	75.41(2)
O7-Tb1-O2W	133.39(2)	O7-Tb1-O3W	73.76(2)
O1W-Tb1-O2W	144.62(2)	O1W-Tb1-O3W	143.63(2)
O2W-Tb1-O3W	71.73(2)	O1-Cu1-O2	84.40(3)
O1-Cu1-N17	97.22(3)	O1-Cu1-N19	95.76(3)
O1-Cu1-N20	170.79(4)	O2-Cu1-N17	88.67(3)
O2-Cu1-N19	178.36(3)	O2-Cu1-N20	93.45(4)
N17-Cu1-N19	92.92(4)	N17-Cu1-N20	91.68(4)
N19-Cu1-N20	86.13(3)	N10-Fe4-N12	89.29(4)
N10-Fe4-N14	90.92(4)	N12-Fe4-N14	89.11(4)
C34-Fe4-C35	85.58(5)	C34-Fe4-C36	90.33(6)
C35-Fe4-C36	84.92(5)	N16-C34-Fe4	179.08 (1)
N17-C35-Fe4	177.17(9)	N18-C36-Fe4	176.37(1)
C16-Fe3-C17	86.39(5)	N7-C16-Fe3	177.56(1)
N8-C17-Fe3	174.26(1)	N9-C18-Fe3	174.77(1)

Table S6 Selected bond lengths (Å) and angles (°) for **3-RR**.

Cu1-O2	1.906(6)	Cu1-O3	1.915(5)
Cu1-N7	2.595(7)	Cu1-N10	1.944(8)
Cu1-N11	1.922(8)	Dy1-O1	2.583(6)
Dy1-O2	2.349(6)	Dy1-O3	2.339(6)
Dy1-O4	2.558(6)	Dy1-O6	2.444(6)
Dy1-O7	2.486(6)	Dy1-O1W	2.342(7)
Dy1-O2W	2.356(5)	Dy1-O3W	2.331(6)
Cu1-Dy1	3.401(2)	Fe1-N1	1.993 (8)
Fe1-N3	1.990(6)	Fe1-N5	2.006(8)
Fe1-C16	1.917(8)	Fe1-C17	1.915(1)
Fe1-C18	1.886(9)	Fe2-N13	1.999(7)
Fe2-N15	1.989(8)	Fe2-N17	1.998(9)
Fe2-C56	1.922(1)	Fe2-C57	1.923(9)
Fe2-C58	1.923(1)	O1-Dy1-O2	61.67(2)
O1-Dy1-O3	126.73(2)	O1-Dy1-O4	157.17(2)
O1-Dy1-O6	66.97(2)	O1-Dy1-O7	116.79(2)
O1-Dy1-O1W	100.62(2)	O1-Dy1-O2W	84.61(2)
O1-Dy1-O3W	77.45(2)	O2-Dy1-O3	65.43(2)
O2-Dy1-O4	124.71(2)	O2-Dy1-O6	112.51(2)
O2-Dy1-O7	152.54(2)	O2-Dy1-O1W	77.43(2)
O2-Dy1-O2W	74.85(2)	O2-Dy1-O3W	128.09(2)

O3-Dy1-O4	62.02(2)	O3-Dy1-O6	144.64(2)
O3-Dy1-O7	112.73(2)	O3-Dy1-O1W	73.15(2)
O3-Dy1-O2W	76.11(2)	O3-Dy1-O3W	136.41(2)
O4-Dy1-O6	119.88(2)	O4-Dy1-O7	68.55(2)
O4-Dy1-O1W	102.19(2)	O4-Dy1-O2W	77.19(2)
O4-Dy1-O3W	83.49(2)	O6-Dy1-O7	51.73(2)
O6-Dy1-O1W	72.12(2)	O6-Dy1-O2W	139.00(2)
O6-Dy1-O3W	74.70(2)	O7-Dy1-O1W	76.06(2)
O7-Dy1-O2W	132.41(2)	O7-Dy1-O3W	73.57(2)
O1W-Dy1-O2W	144.99(2)	O1W-Dy1-O3W	144.45(2)
O2W-Dy1-O3W	70.55(2)	O2-Cu-O3	83.10(2)
O2-Cu-N7	96.84(2)	O2-Cu-N10	95.95(3)
O2-Cu-N11	169.49(3)	O3-Cu-N7	89.21(2)
O3-Cu-N10	177.76(3)	O3-Cu-N11	94.61(3)
N7-Cu-N10	92.92(3)	N7-Cu-N11	93.37(3)
N10-Cu-N11	85.95(3)	N1-Fe1-N3	90.14(3)
N1-Fe1-N5	89.18(3)	N3-Fe1-N5	89.45(3)
C16-Fe1-C17	85.21(4)	C16-Fe1-C18	85.29(4)
C17-Fe1-C18	89.94(5)	N7-C16-Fe1	176.74(8)
N8-C17-Fe1	178.48(1)	N9-C18-Fe1	178.90(8)
C56-Fe2-C57	85.33(4)	N19-C56-Fe2	177.83(1)
N20-C57-Fe2	179.35(9)	N21-C58-Fe2	174.70(1)

Table S7 Selected bond lengths (Å) and angles (°) for **3-SS**.

Cu1-O2	1.924(3)	Cu1-O3	1.905(4)
Cu1-N7	2.605(4)	Cu1-N10	1.926(5)
Cu1-N11	1.921(5)	Dy1-O1	2.567(3)
Dy1-O2	2.345(3)	Dy1-O3	2.342(3)
Dy1-O4	2.581(3)	Dy1-O5	2.481(4)
Dy1-O6	2.494(3)	Dy1-O1W	2.331(3)
Dy1-O2W	2.339(4)	Dy1-O3W	2.350 (3)
Cu1-Dy1	3.396(8)	Fe1-N1	1.983(5)
Fe1-N3	2.015(4)	Fe1-N5	2.003(5)
Fe1-C16	1.927(5)	Fe1-C17	1.932(6)
Fe1-C18	1.934(6)	Fe2-N13	1.995(5)
Fe2-N15	1.997(4)	Fe2-N17	1.991(5)
Fe2-C56	1.922(5)	Fe2-C57	1.934(6)
Fe2-C58	1.917(6)	O1-Dy1-O2	62.62(1)
O1-Dy1-O3	123.94(1)	O1-Dy1-O4	155.15(1)
O1-Dy1-O5	120.12(1)	O1-Dy1-O6	69.57(1)
O1-Dy1-O1W	103.90(1)	O1-Dy1-O2W	81.84(1)
O1-Dy1-O3W	75.31(1)	O2-Dy1-O3	65.83(1)
O2-Dy1-O4	127.58(1)	O2-Dy1-O5	142.88(1)

O2-Dy1-O6	111.32(1)	O2-Dy1-O1W	72.26(1)
O2-Dy1-O2W	137.63(1)	O2-Dy1-O3W	77.25(1)
O3-Dy1-O4	61.87(1)	O3-Dy1-O5	113.93(1)
O3-Dy1-O6	153.59(1)	O3-Dy1-O1W	79.45(1)
O3-Dy1-O2W	128.06(1)	O3-Dy1-O3W	73.33(1)
O4-Dy1-O5	67.44(1)	O4-Dy1-O6	116.79(1)
O4-Dy1-O1W	100.93(1)	O4-Dy1-O2W	77.78 (1)
O4-Dy1-O3W	84.84(1)	O5-Dy1-O6	51.21(1)
O5-Dy1-O1W	71.39(1)	O5-Dy1-O2W	74.12(1)
O5-Dy1-O3W	139.71(1)	O6-Dy1-O1W	74.96(1)
O6-Dy1-O2W	72.98(1)	O6-Dy1-O3W	132.81(1)
O1W-Dy1-O2W	142.91(1)	O1W-Dy1-O3W	145.38(1)
O2W-Dy1-O3W	71.70(1)	O2-Cu-O3	83.40(1)
O2-Cu-N7	88.50(1)	O2-Cu-N10	94.00(1)
O2-Cu-N11	178.28(2)	O3-Cu-N7	96.05(1)
O3-Cu-N10	170.82(2)	O3-Cu-N11	95.91(2)
N7-Cu-N10	92.67(2)	N7-Cu-N11	93.14(1)
N10-Cu-N11	86.44(2)	N1-Fe1-N3	89.11(2)
N1-Fe1-N5	88.62(2)	N3-Fe1-N5	90.57(2)
C16-Fe1-C17	85.42(2)	C16-Fe1-C18	85.64(2)
C17-Fe1-C18	90.64(3)	N7-C16-Fe1	176.61(5)
N8-C17-Fe1	179.37 (5)	N9-C18-Fe1	178.96 (6)
C56-Fe2-C57	84.62(2)	N19-C56-Fe2	175.95(5)
N20-C57-Fe2	175.24(5)	N21-C58-Fe2	178.31(5)

Table S8. Results of the Continuous Shape Measure Analysisa geometry[a]

Geometry	CSAPR-9	JCSAPR-9	CCU-9
1-RR	**3.77**	4.54	6.79
1-SS	**3.94**	4.74	6.68
2-RR	**3.83**	4.61	6.88
2-SS	**3.80**	4.58	6.84
3-RR	**4.06**	4.82	6.56
3-SS	**3.74**	4.51	6.78

[a]CSAPR-9 is the shape measure relative to the spherical capped square antiprism; JCSAPR-9 is the shape measure relative to the capped square antiprism; CCU-9 is the shape measure relative to the spherical-relaxed capped cube. The number in bold corresponds to the closer ideal geometry to the real complexes.

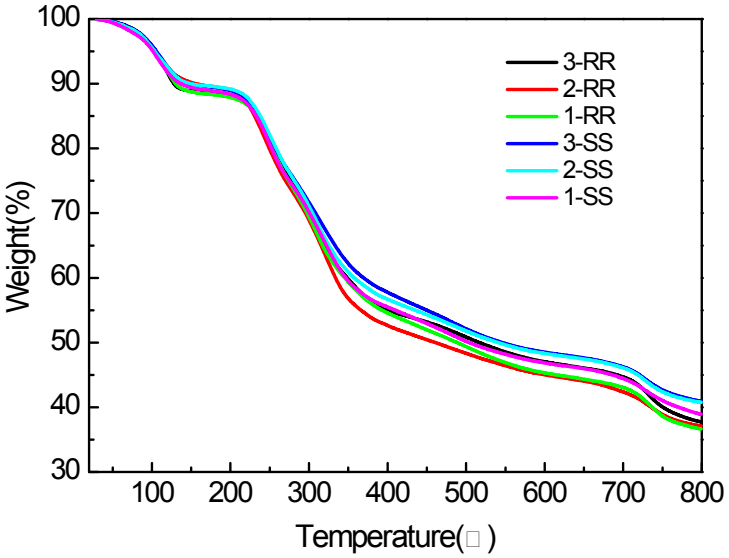
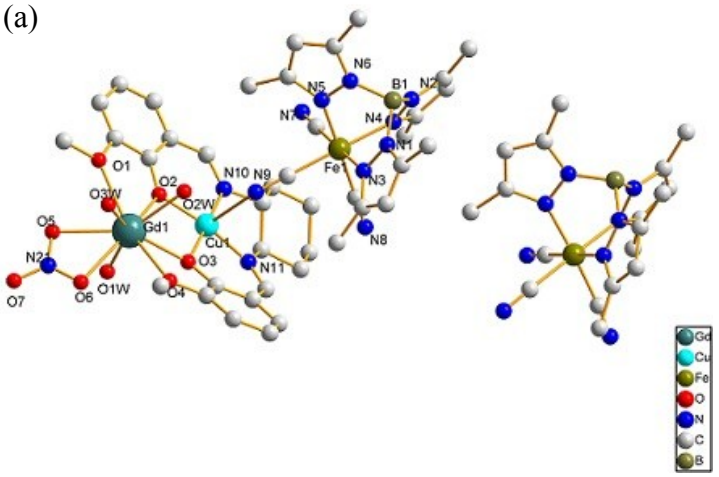
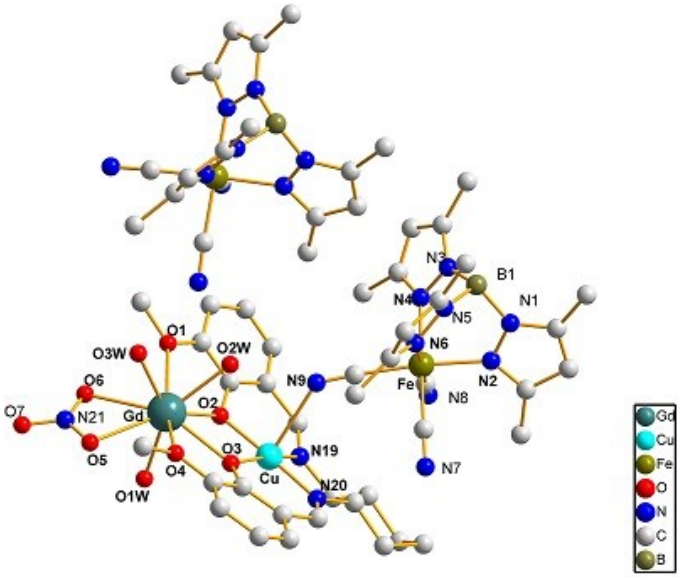


Figure S1. The thermogravimetry curves of all complexes in an N_2 atmosphere at a heating rate of 10 °C/min.

(a)



(b)



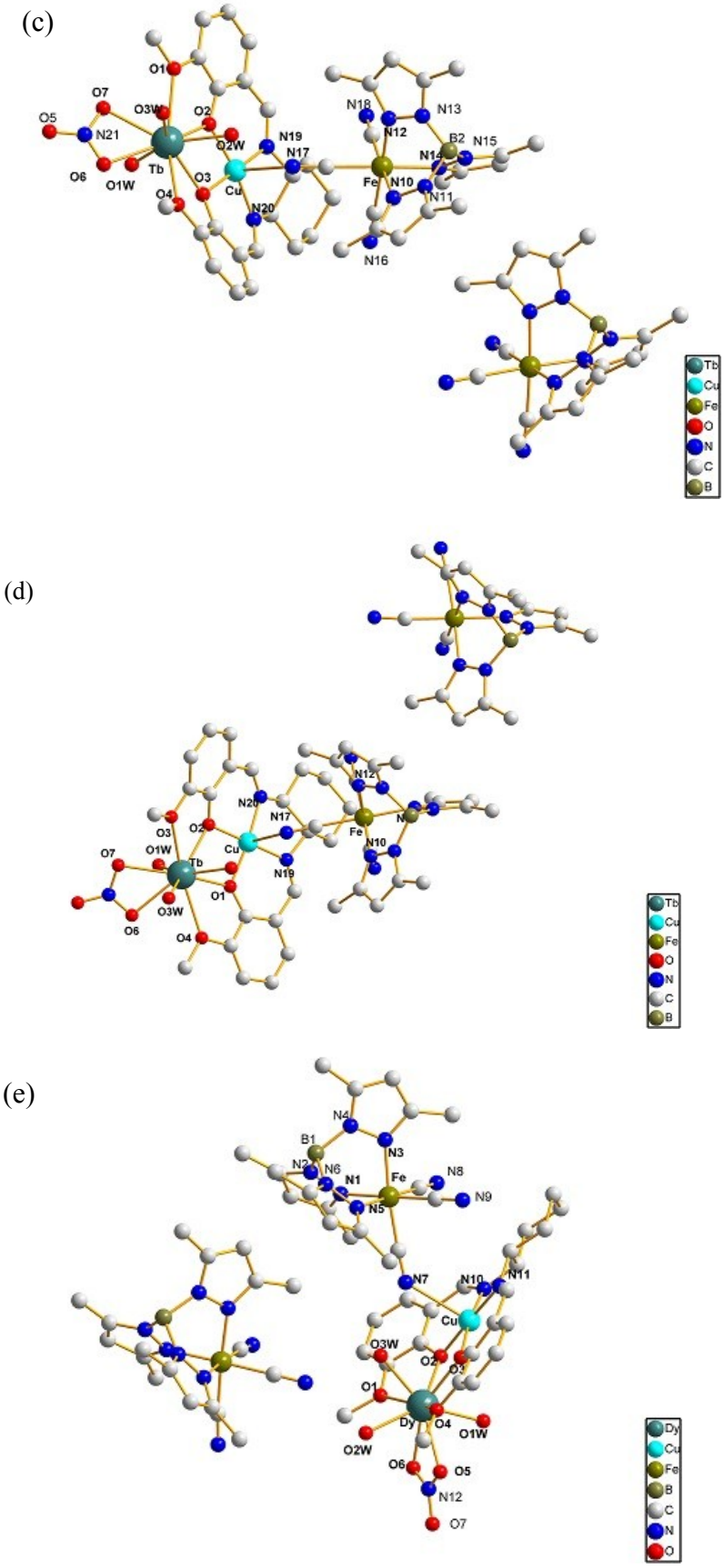


Figure S2. Perspective drawing of the crystallographic structural unit of **1-RR** (a), **1-SS** (b), **2-RR** (c), **2-SS** (d), and **3-SS** (e), respectively, showing the atom numbering. H atoms are omitted for

clarity.

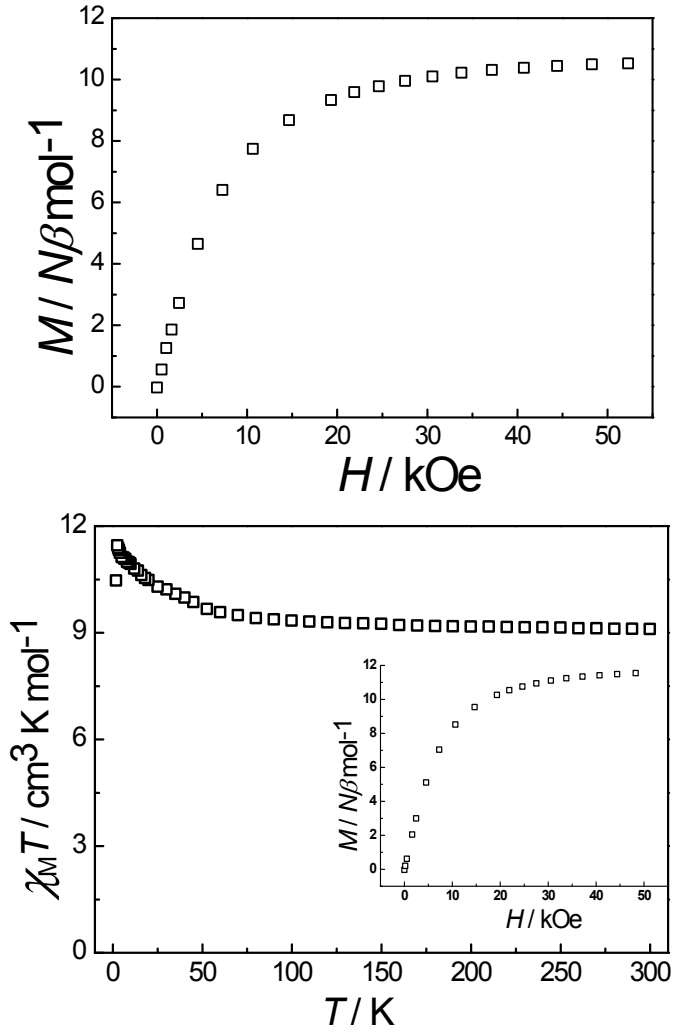


Figure S3. Up: Field dependence of magnetization for **1-RR** at 1.8 K. Down: Temperature dependence of the $\chi_M T$ product for **1-SS** under an applied magnetic fields of 100 KOe. Inset: Field dependence of magnetization for **1-SS** at low temperature..

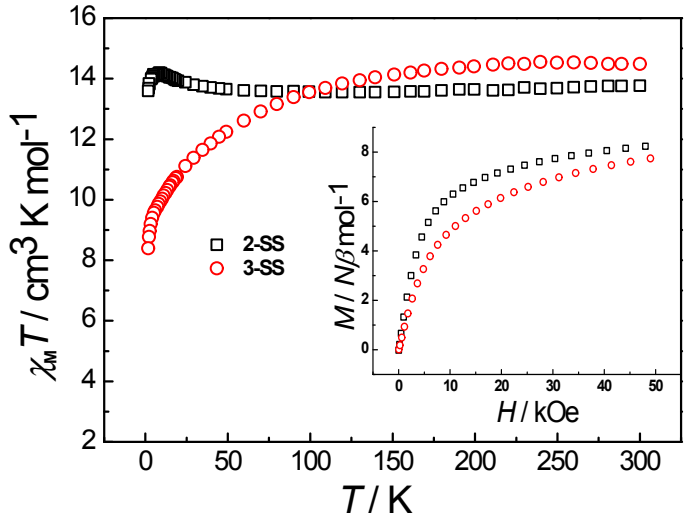


Figure S4. Temperature dependence of the $\chi_M T$ product for **2-SS**, **3-SS** at 1 kOe. Inset: Field dependence of magnetization for **2-SS**, **3-SS** at 1.8 K.

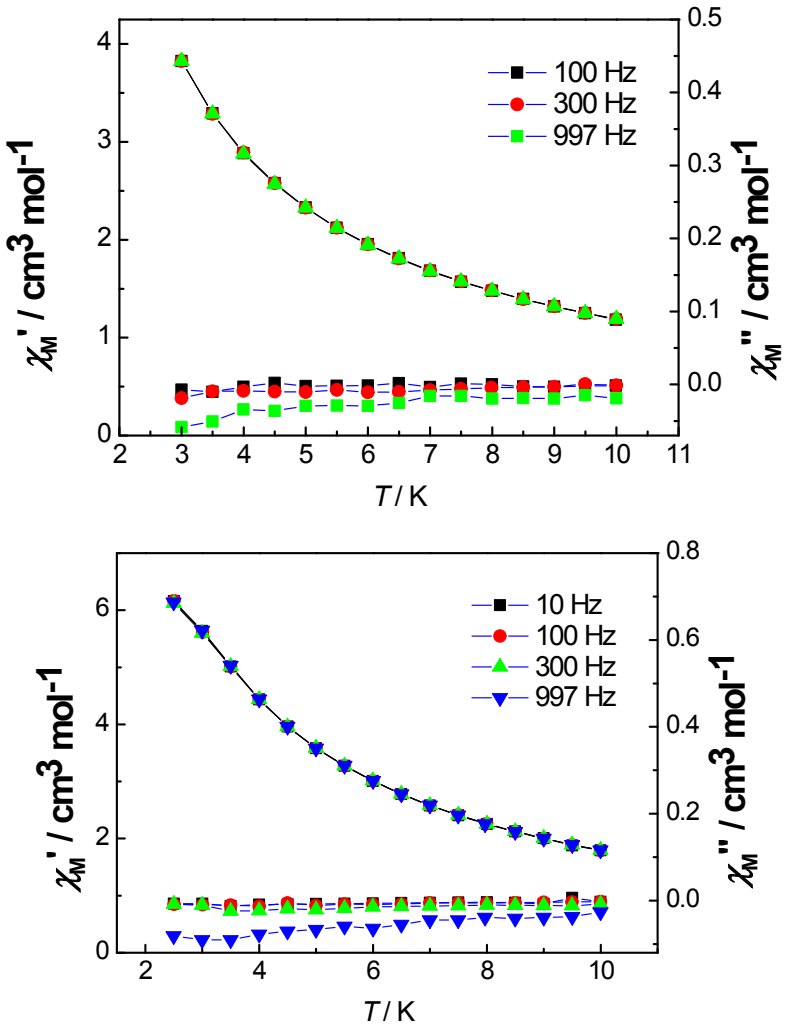


Figure S5. Temperature dependence of the in-phase χ' and out-of-phase χ'' at different frequencies in a 2.5 Oe ac field oscillating at 100–997 Hz with a dc field of 0 Oe for **2-RR** (up) and **3-RR** (down).