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Synthesis, Structures and Magnetism of series of Binuclear and one-dimensional Ni-Ln Complexes: Single-molecule Magnetic Behavior in one-dimensional Nitrate-bridged Dy Analogue

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Supporting information

Table S1 Selected bond lengths (Å) and angles (°) for **1**.

Gd(1)-O(2)	2.3414(18)	Gd(1)-O(5)	2.3494(19)
Gd(1)-O(14)	2.484(2)	Gd(1)-O(7)	2.485(2)
Gd(1)-O(10)	2.495(3)	Gd(1)-O(13)	2.511(3)
Gd(1)-O(6)	2.546(2)	Gd(1)-O(11)	2.549(3)
Gd(1)-O(8)	2.555(3)	Gd(1)-O(1)	2.653(2)
Gd(1)-Ni(1)	3.4798(6)	Ni(1)-N(1)	2.011(2)
Ni(1)-O(5)	2.0213(19)	Ni(1)-O(2)	2.0357(18)
Ni(1)-N(2)	2.053(2)	Ni(1)-N(4)	2.088(3)
Ni(1)-N(3)	2.134(3)	O(2)-Gd(1)-O(5)	68.46(6)
O(2)-Gd(1)-O(14)	88.22(7)	O(5)-Gd(1)-O(14)	77.53(7)
O(2)-Gd(1)-O(7)	79.30(7)	O(5)-Gd(1)-O(7)	90.91(7)
O(14)-Gd(1)-O(7)	165.49(8)	O(2)-Gd(1)-O(10)	134.07(9)
O(5)-Gd(1)-O(10)	146.45(9)	O(14)-Gd(1)-O(10)	120.65(9)
O(7)-Gd(1)-O(10)	73.76(9)	O(2)-Gd(1)-O(13)	76.27(8)
O(5)-Gd(1)-O(13)	117.39(8)	O(14)-Gd(1)-O(13)	50.83(8)
O(7)-Gd(1)-O(13)	131.23(8)	O(10)-Gd(1)-O(13)	94.49(10)

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O(2)-Gd(1)-O(6)	130.63(6)	O(5)-Gd(1)-O(6)	64.09(7)
O(14)-Gd(1)-O(6)	69.34(7)	O(7)-Gd(1)-O(6)	113.61(8)
O(10)-Gd(1)-O(6)	94.45(9)	O(13)-Gd(1)-O(6)	114.40(9)
O(2)-Gd(1)-O(11)	144.04(8)	O(5)-Gd(1)-O(11)	131.32(10)
O(14)-Gd(1)-O(11)	71.53(9)	O(7)-Gd(1)-O(11)	122.97(10)
O(10)-Gd(1)-O(11)	49.65(10)	O(13)-Gd(1)-O(11)	67.88(9)
O(6)-Gd(1)-O(11)	70.16(10)	O(2)-Gd(1)-O(8)	117.28(8)
O(5)-Gd(1)-O(8)	77.03(9)	O(14)-Gd(1)-O(8)	133.16(8)
O(7)-Gd(1)-O(8)	50.05(7)	O(10)-Gd(1)-O(8)	70.22(10)
O(13)-Gd(1)-O(8)	164.10(9)	O(6)-Gd(1)-O(8)	64.27(7)
O(11)-Gd(1)-O(8)	97.87(10)	O(2)-Gd(1)-O(1)	62.42(7)
O(5)-Gd(1)-O(1)	128.81(7)	O(14)-Gd(1)-O(1)	113.35(8)
O(7)-Gd(1)-O(1)	67.27(8)	O(10)-Gd(1)-O(1)	72.99(9)
O(13)-Gd(1)-O(1)	64.05(9)	O(6)-Gd(1)-O(1)	166.86(7)
O(11)-Gd(1)-O(1)	98.07(10)	O(8)-Gd(1)-O(1)	113.29(8)
N(1)-Ni(1)-O(5)	170.31(9)	N(1)-Ni(1)-O(2)	90.22(8)
O(5)-Ni(1)-O(2)	81.14(7)	N(1)-Ni(1)-N(2)	98.71(9)
O(5)-Ni(1)-N(2)	89.98(8)	O(2)-Ni(1)-N(2)	171.06(8)
N(1)-Ni(1)-N(4)	87.67(9)	O(5)-Ni(1)-N(4)	88.36(9)
O(2)-Ni(1)-N(4)	92.71(9)	N(2)-Ni(1)-N(4)	88.05(9)
N(1)-Ni(1)-N(3)	93.30(9)	O(5)-Ni(1)-N(3)	90.39(9)
O(2)-Ni(1)-N(3)	85.40(9)	N(2)-Ni(1)-N(3)	93.68(9)
N(4)-Ni(1)-N(3)	177.87(9)		

Table S2 Selected bond lengths (Å) and angles (°) for **2**.

Tb(1)-O(4)	2.3283(15)	Tb(1)-O(1)	2.3394(16)
Tb(1)-O(11)	2.4703(17)	Tb(1)-O(7)	2.4730(19)
Tb(1)-O(13)	2.478(2)	Tb(1)-O(8)	2.503(2)
Tb(1)-O(5)	2.5395(19)	Tb(1)-O(14)	2.544(2)
Tb(1)-O(10)	2.549(2)	Tb(1)-O(6)	2.649(2)
Tb(1)-Ni(1)	3.4695(7)	Ni(1)-N(2)	2.0109(19)
Ni(1)-O(1)	2.0158(15)	Ni(1)-O(4)	2.0332(16)
Ni(1)-N(1)	2.0562(19)	Ni(1)-N(3)	2.090(2)
Ni(1)-N(4)	2.133(2)	O(4)-Tb(1)-O(1)	68.50(5)
O(4)-Tb(1)-O(11)	79.23(6)	O(1)-Tb(1)-O(11)	90.77(6)
O(4)-Tb(1)-O(7)	88.14(6)	O(1)-Tb(1)-O(7)	77.66(6)
O(11)-Tb(1)-O(7)	165.39(6)	O(4)-Tb(1)-O(13)	134.16(8)
O(1)-Tb(1)-O(13)	146.22(7)	O(11)-Tb(1)-O(13)	73.79(8)

O(7)-Tb(1)-O(13)	120.71(8)	O(4)-Tb(1)-O(8)	76.28(6)
O(1)-Tb(1)-O(8)	117.66(7)	O(11)-Tb(1)-O(8)	131.07(7)
O(7)-Tb(1)-O(8)50.92(7)	O(13)-Tb(1)-O(8)	94.47(8)	
O(4)-Tb(1)-O(5)	130.99(5)	O(1)-Tb(1)-O(5)	64.43(5)
O(11)-Tb(1)-O(5)	113.66(6)	O(7)-Tb(1)-O(5)	69.52(6)
O(13)-Tb(1)-O(5)	94.00(7)	O(8)-Tb(1)-O(5)	114.45(7)
O(4)-Tb(1)-O(14)	143.70(7)	O(1)-Tb(1)-O(14)	131.64(8)
O(11)-Tb(1)-O(14)	123.07(8)	O(7)-Tb(1)-O(14)	71.54(8)
O(13)-Tb(1)-O(14)	49.64(8)	O(8)-Tb(1)-O(14)	67.53(8)
O(5)-Tb(1)-O(14)	70.20(9)	O(4)-Tb(1)-O(10)	117.39(6)
O(1)-Tb(1)-O(10)	76.92(7)	O(11)-Tb(1)-O(10)	50.22(6)
O(7)-Tb(1)-O(10)	133.15(7)	O(13)-Tb(1)-O(10)	70.08(8)
O(8)-Tb(1)-O(10)	163.95(8)	O(5)-Tb(1)-O(10)	64.11(6)
O(14)-Tb(1)-O(10)	98.07(8)	O(4)-Tb(1)-O(6)	62.85(6)
O(1)-Tb(1)-O(6)	129.22(5)	O(11)-Tb(1)-O(6)	67.23(7)
O(7)-Tb(1)-O(6)	113.40(7)	O(13)-Tb(1)-O(6)	72.68(8)
O(8)-Tb(1)-O(6)	63.95(7)	O(5)-Tb(1)-O(6)	166.07(6)
O(14)-Tb(1)-O(6)	97.38(9)	O(10)-Tb(1)-O(6)	113.22(6)
N(2)-Ni(1)-O(1)	170.16(7)	N(2)-Ni(1)-O(4)	90.25(7)
O(1)-Ni(1)-O(4)	80.91(6)	N(2)-Ni(1)-N(1)	98.83(8)
O(1)-Ni(1)-N(1)	90.05(7)	O(4)-Ni(1)-N(1)	170.92(7)
N(2)-Ni(1)-N(3)	87.71(8)	O(1)-Ni(1)-N(3)	88.37(7)
O(4)-Ni(1)-N(3)	92.50(8)	N(1)-Ni(1)-N(3)	88.13(8)
N(2)-Ni(1)-N(4)	93.19(8)	O(1)-Ni(1)-N(4)	90.45(7)
O(4)-Ni(1)-N(4)	85.69(7)	N(1)-Ni(1)-N(4)	93.52(8)
N(3)-Ni(1)-N(4)	177.98(8)		

Table S3 Selected bond lengths (Å) and angles (°) for **3**.

Dy(1)-O(2)	2.283(3)	Dy(1)-O(5)	2.293(3)
Dy(1)-O(15)	2.372(3)	Dy(1)-O(9)	2.429(3)
Dy(1)-O(12)	2.430(3)	Dy(1)-O(11)	2.476(3)
Dy(1)-O(1)	2.488(3)	Dy(1)-O(6)	2.497(3)
Dy(1)-O(8)	2.517(3)	Dy(1)-Ni(1)	3.4538(6)
Ni(1)-O(5)	2.005(3)	Ni(1)-O(2)	2.010(3)
Ni(1)-N(1)	2.023(4)	Ni(1)-N(2)	2.037(4)
Ni(1)-N(6)	2.117(4)	Ni(1)-O(13)	2.161(3)
O(2)-Dy(1)-O(5)	67.27(9)	O(2)-Dy(1)-O(15)	148.23(10)
O(5)-Dy(1)-O(15)	135.16(10)	O(2)-Dy(1)-O(9)	91.95(11)

O(5)-Dy(1)-O(9)	80.22(10)	O(15)-Dy(1)-O(9)	74.50(10)
O(2)-Dy(1)-O(12)	124.36(11)	O(5)-Dy(1)-O(12)	121.80(11)
O(15)-Dy(1)-O(12)	68.21(10)	O(9)-Dy(1)-O(12)	141.85(11)
O(2)-Dy(1)-O(11)	81.77(11)	O(5)-Dy(1)-O(11)	80.32(10)
O(15)-Dy(1)-O(11)	119.57(11)	O(9)-Dy(1)-O(11)	160.49(11)
O(12)-Dy(1)-O(11)	51.74(11)	O(2)-Dy(1)-O(1)	65.67(10)
O(5)-Dy(1)-O(1)	129.22(10)	O(15)-Dy(1)-O(1)	95.53(10)
O(9)-Dy(1)-O(1)	118.97(11)	O(12)-Dy(1)-O(1)	73.38(11)
O(11)-Dy(1)-O(1)	75.23(12)	O(2)-Dy(1)-O(6)	131.23(9)
O(5)-Dy(1)-O(6)	64.48(9)	O(15)-Dy(1)-O(6)	77.53(10)
O(9)-Dy(1)-O(6)	86.73(11)	O(12)-Dy(1)-O(6)	77.87(11)
O(11)-Dy(1)-O(6)	83.71(11)	O(1)-Dy(1)-O(6)	150.93(10)
O(2)-Dy(1)-O(8)	73.36(10)	O(5)-Dy(1)-O(8)	114.99(10)
O(15)-Dy(1)-O(8)	75.82(10)	O(9)-Dy(1)-O(8)	51.39(11)
O(12)-Dy(1)-O(8)	123.00(11)	O(11)-Dy(1)-O(8)	141.27(12)
O(1)-Dy(1)-O(8)	67.67(10)	O(6)-Dy(1)-O(8)	134.94(10)
O(5)-Ni(1)-O(2)	78.28(10)	O(5)-Ni(1)-N(1)	165.19(14)
O(2)-Ni(1)-N(1)	88.03(14)	O(5)-Ni(1)-N(2)	91.37(13)
O(2)-Ni(1)-N(2)	168.75(13)	N(1)-Ni(1)-N(2)	102.67(16)
O(5)-Ni(1)-N(6)	87.78(13)	O(2)-Ni(1)-N(6)	95.75(13)
N(1)-Ni(1)-N(6)	87.98(14)	N(2)-Ni(1)-N(6)	88.19(14)
O(5)-Ni(1)-O(13)	98.37(11)	O(2)-Ni(1)-O(13)	84.83(12)
N(1)-Ni(1)-O(13)	85.88(13)	N(2)-Ni(1)-O(13)	92.40(13)
N(6)-Ni(1)-O(13)	173.81(13)		

Table S4 Selected bond lengths (Å) and angles (°) for **4**.

Tb(1)-O(5)	2.240(2)	Tb(1)-O(2)	2.293(2)
Tb(1)-O(14)	2.399(3)	Tb(1)-O(12)	2.407(3)
Tb(1)-O(9)	2.427(3)	Tb(1)-O(15)	2.428(3)
Tb(1)-O(1)	2.443(3)	Tb(1)-O(11)	2.483(3)
Tb(1)-O(6)	2.548(3)	Tb(1)-Ni(1)	3.3479(6)
Ni(1)-O(5)	1.999(2)	Ni(1)-N(1)	2.015(3)
Ni(1)-O(2)	2.024(2)	Ni(1)-N(2)	2.067(3)
Ni(1)-O(16)	2.070(3)	Ni(1)-O(8)	2.122(2)
O(5)-Tb(1)-O(2)	70.82(8)	O(5)-Tb(1)-O(14)	131.94(11)
O(2)-Tb(1)-O(14)	118.17(11)	O(5)-Tb(1)-O(12)	133.67(10)
O(2)-Tb(1)-O(12)	135.18(10)	O(14)-Tb(1)-O(12)	75.96(13)
O(5)-Tb(1)-O(9)	81.38(9)	O(2)-Tb(1)-O(9)	73.35(9)

O(14)-Tb(1)-O(9)	146.21(11)	O(12)-Tb(1)-O(9)	75.05(11)
O(5)-Tb(1)-O(15)	91.13(12)	O(2)-Tb(1)-O(15)	75.87(11)
O(14)-Tb(1)-O(15)	52.31(13)	O(12)-Tb(1)-O(15)	127.99(13)
O(9)-Tb(1)-O(15)	149.10(12)	O(5)-Tb(1)-O(1)	136.30(9)
O(2)-Tb(1)-O(1)	66.29(8)	O(14)-Tb(1)-O(1)	78.05(11)
O(12)-Tb(1)-O(1)	77.09(10)	O(9)-Tb(1)-O(1)	78.88(10)
O(15)-Tb(1)-O(1)	86.45(12)	O(5)-Tb(1)-O(11)	83.03(10)
O(2)-Tb(1)-O(11)	138.38(10)	O(14)-Tb(1)-O(11)	103.42(13)
O(12)-Tb(1)-O(11)	51.83(11)	O(9)-Tb(1)-O(11)	71.11(11)
O(15)-Tb(1)-O(11)	137.98(13)	O(1)-Tb(1)-O(11)	125.34(10)
O(5)-Tb(1)-O(6)	64.91(9)	O(2)-Tb(1)-O(6)	124.41(9)
O(14)-Tb(1)-O(6)	74.17(11)	O(12)-Tb(1)-O(6)	100.07(11)
O(9)-Tb(1)-O(6)	127.97(10)	O(15)-Tb(1)-O(6)	73.19(12)
O(1)-Tb(1)-O(6)	151.87(10)	O(11)-Tb(1)-O(6)	66.65(11)
O(5)-Ni(1)-N(1)	169.94(12)	O(5)-Ni(1)-O(2)	81.52(10)
N(1)-Ni(1)-O(2)	88.52(12)	O(5)-Ni(1)-N(2)	87.24(12)
N(1)-Ni(1)-N(2)	102.76(13)	O(2)-Ni(1)-N(2)	168.56(11)
O(5)-Ni(1)-O(16)	89.45(12)	N(1)-Ni(1)-O(16)	89.53(13)
O(2)-Ni(1)-O(16)	92.33(11)	N(2)-Ni(1)-O(16)	89.71(12)
O(5)-Ni(1)-O(8)	92.22(10)	N(1)-Ni(1)-O(8)	88.71(12)
O(2)-Ni(1)-O(8)	87.31(10)	N(2)-Ni(1)-O(8)	90.99(12)
O(16)-Ni(1)-O(8)	178.21(12)		

Table S5 Selected bond lengths (Å) and angles (°) for **5**.

Dy(1)-O(4)	2.239(3)	Dy(1)-O(1)	2.293(3)
Dy(1)-O(9)	2.407(4)	Dy(1)-O(14)	2.415(3)
Dy(1)-O(11)	2.417(3)	Dy(1)-O(8)	2.434(4)
Dy(1)-O(5)	2.436(3)	Dy(1)-O(12)	2.482(4)
Dy(1)-O(6)	2.542(3)	Dy(1)-Ni(1)	3.3435(7)
Ni(1)-O(4)	2.002(3)	Ni(1)-O(1)	2.020(3)
Ni(1)-N(1)	2.029(4)	Ni(1)-N(2)	2.063(4)
Ni(1)-O(16)	2.071(3)	Ni(1)-O(15)	2.120(3)
O(4)-Dy(1)-O(1)	70.89(10)	O(4)-Dy(1)-O(9)	131.95(13)
O(1)-Dy(1)-O(9)	118.01(14)	O(4)-Dy(1)-O(14)	81.41(11)
O(1)-Dy(1)-O(14)	73.74(10)	O(9)-Dy(1)-O(14)	146.13(13)
O(4)-Dy(1)-O(11)	133.07(12)	O(1)-Dy(1)-O(11)	135.67(12)
O(9)-Dy(1)-O(11)	76.20(15)	O(14)-Dy(1)-O(11)	74.82(13)
O(4)-Dy(1)-O(8)	91.54(14)	O(1)-Dy(1)-O(8)	75.59(13)

O(9)-Dy(1)-O(8)	52.14(15)	O(14)-Dy(1)-O(8)	149.17(13)
O(11)-Dy(1)-O(8)	128.07(15)	O(4)-Dy(1)-O(5)	136.35(10)
O(1)-Dy(1)-O(5)	66.23(10)	O(9)-Dy(1)-O(5)	77.83(14)
O(14)-Dy(1)-O(5)	79.28(12)	O(11)-Dy(1)-O(5)	77.86(12)
O(8)-Dy(1)-O(5)	85.68(15)	O(4)-Dy(1)-O(12)	83.29(12)
O(1)-Dy(1)-O(12)	138.63(11)	O(9)-Dy(1)-O(12)	103.32(15)
O(14)-Dy(1)-O(12)	70.80(13)	O(11)-Dy(1)-O(12)	50.91(13)
O(8)-Dy(1)-O(12)	138.45(15)	O(5)-Dy(1)-O(12)	125.28(12)
O(4)-Dy(1)-O(6)	65.11(11)	O(1)-Dy(1)-O(6)	124.65(10)
O(9)-Dy(1)-O(6)	74.09(14)	O(14)-Dy(1)-O(6)	127.76(12)
O(11)-Dy(1)-O(6)	99.32(13)	O(8)-Dy(1)-O(6)	73.79(15)
O(5)-Dy(1)-O(6)	151.59(12)	O(12)-Dy(1)-O(6)	66.65(13)
O(4)-Ni(1)-O(1)	81.62(12)	O(4)-Ni(1)-N(1)	169.97(14)
O(1)-Ni(1)-N(1)	88.50(14)	O(4)-Ni(1)-N(2)	87.09(13)
O(1)-Ni(1)-N(2)	168.51(13)	N(1)-Ni(1)-N(2)	102.84(15)
O(4)-Ni(1)-O(16)	89.40(14)	O(1)-Ni(1)-O(16)	92.75(13)
N(1)-Ni(1)-O(16)	89.30(16)	N(2)-Ni(1)-O(16)	89.24(14)
O(4)-Ni(1)-O(15)	92.49(12)	O(1)-Ni(1)-O(15)	87.53(12)
N(1)-Ni(1)-O(15)	88.84(15)	N(2)-Ni(1)-O(15)	90.85(14)
O(16)-Ni(1)-O(15)	178.11(14)		

Table S6. Results of the Continuous Shape Measure Analysisa geometry[a]

Geometry	JSPC-10	SDD-10	TD-10
1	3.00	**1.82**	1.94
2	2.94	**1.86**	1.96

[a]JSPC-10 is the shape measure relative to the sphenocorona J87; SDD-10 is the shape measure relative to the staggered dodecahedron (2:6:2); TD-10 is the shape measure relative to the tetradecahedron (2:6:2).

Geometry	MFF-9	CSAPR-9	JCSAPR-9	TCTPR-9
3	2.64	2.72	3.90	**2.38**
4	2.00	**1.70**	3.02	2.82
5	2.09	**1.77**	3.08	2.86

[a]MFF-9 is the shape measure relative to the muffin; 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; TCTPR-9 is the shape measure relative to the spherical tricapped trigonal prism. The number in bold corresponds to the closer ideal

geometry to the real complexes.

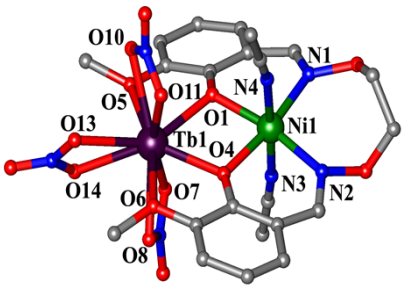

Figure S1. Perspective drawing of the crystallographically structural unit of **2** showing the atom numbering. H atoms are omitted for clarity.

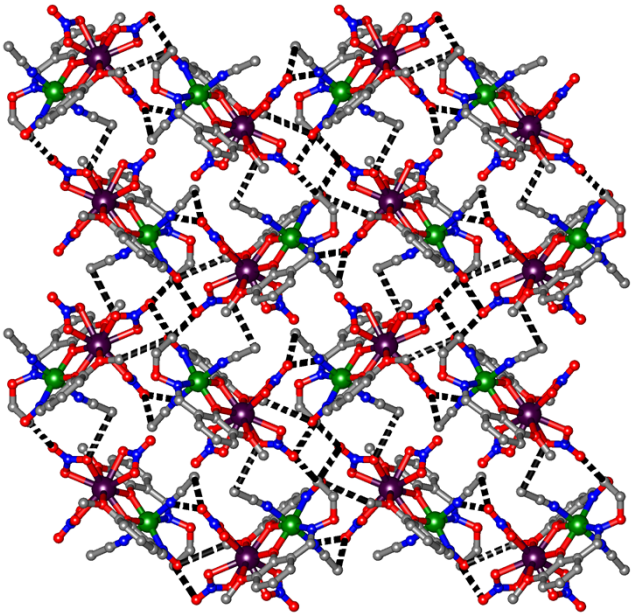

Figure S2. A view showing 3D structure formed by weak H-bonding interactions (black dash lines) in **1**.

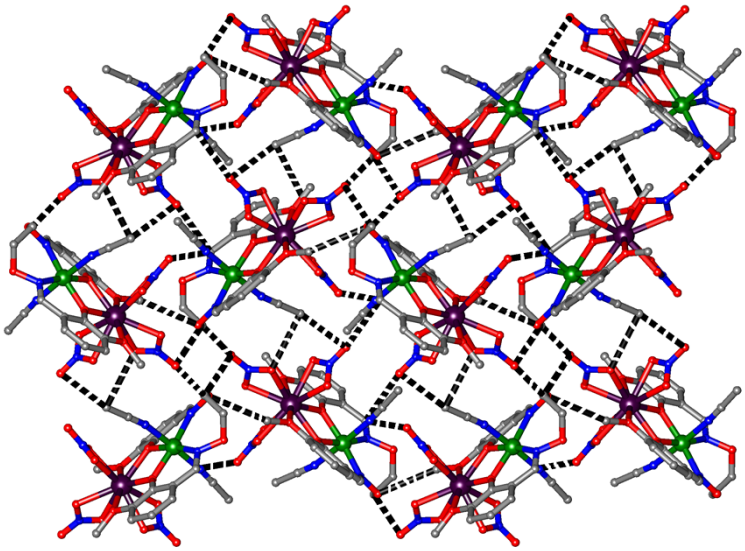

Figure S3. A view showing 3D structure formed by weak H-bonding interactions (black dash lines) in **2**.

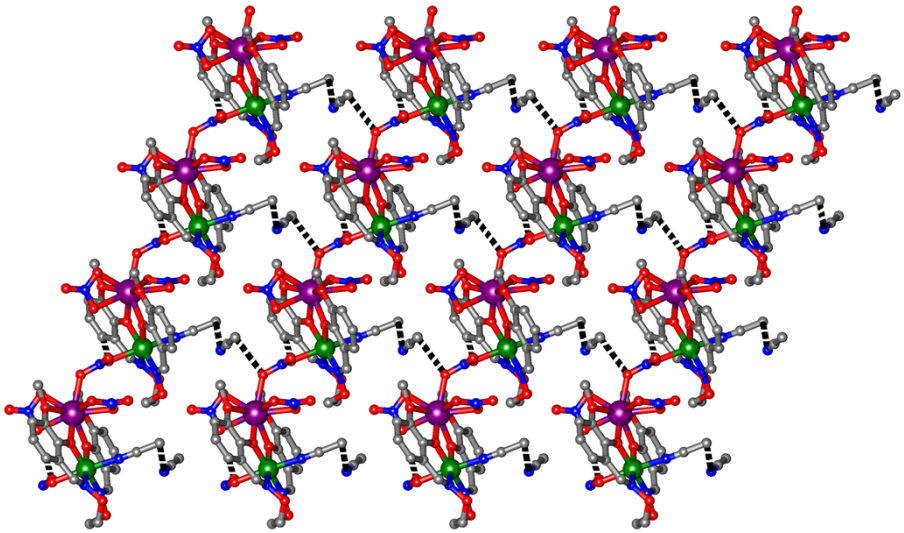

Figure S4. A view showing 2D structure formed by weak H-bonding interactions (black dash lines) in **3**.

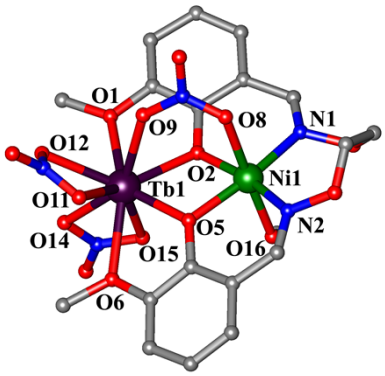

Figure S5. Perspective drawing of the crystallographically structural unit of **4** showing the atom numbering. H atoms and one solvated diethyl ether molecule are omitted for clarity.

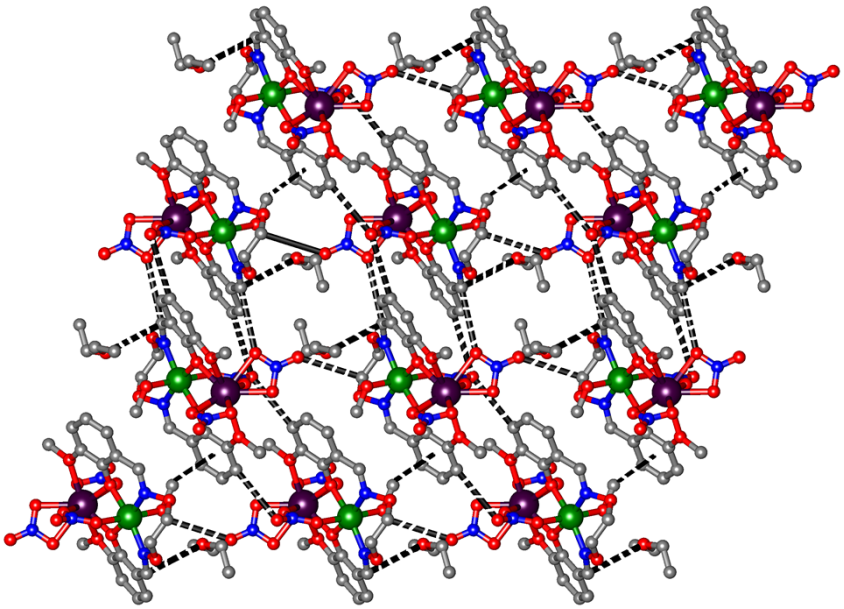

Figure S6. A view showing 3D structure formed by weak H-bonding interactions and CH···π interactions (black dash lines) in **4**.

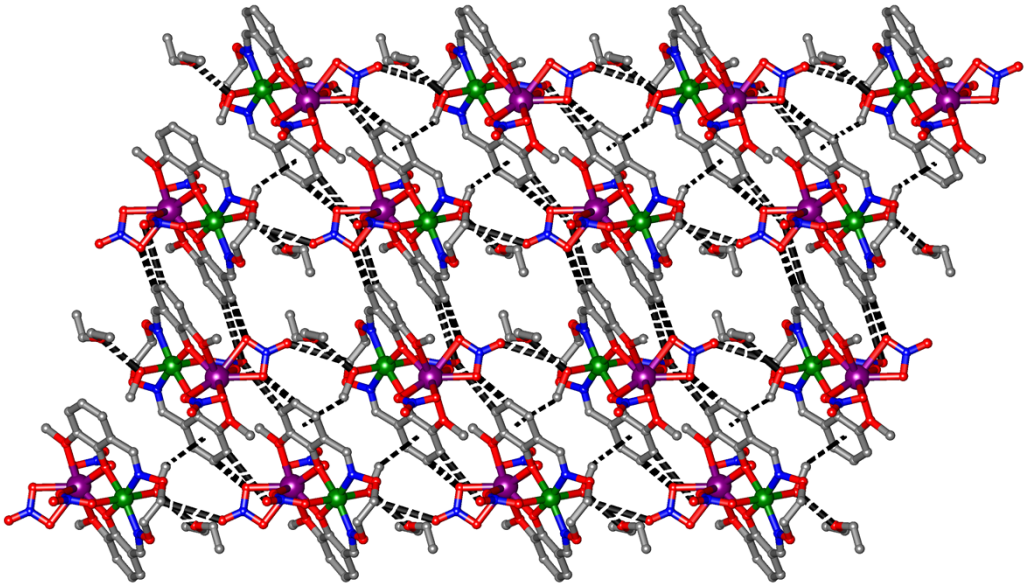

Figure S7. A view showing 3D structure formed by weak H-bonding interactions and CH···π interactions (black dash lines) in **5**.

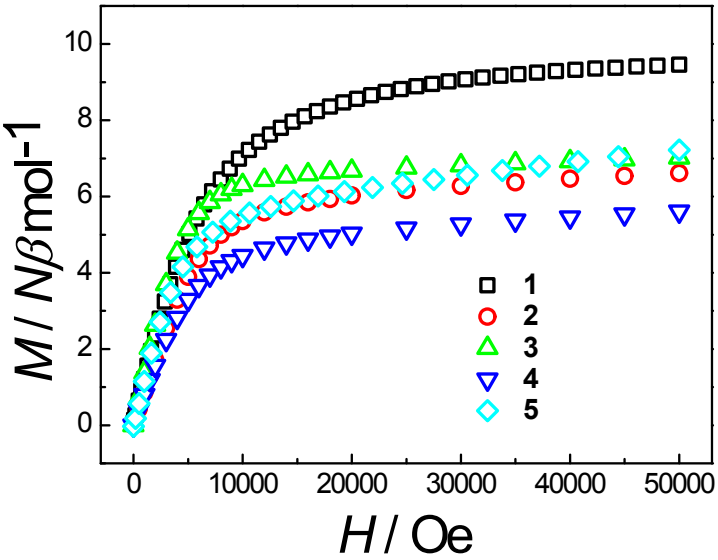

Figure S8. Field dependence of magnetization for **1**-**5** at low temperatures.

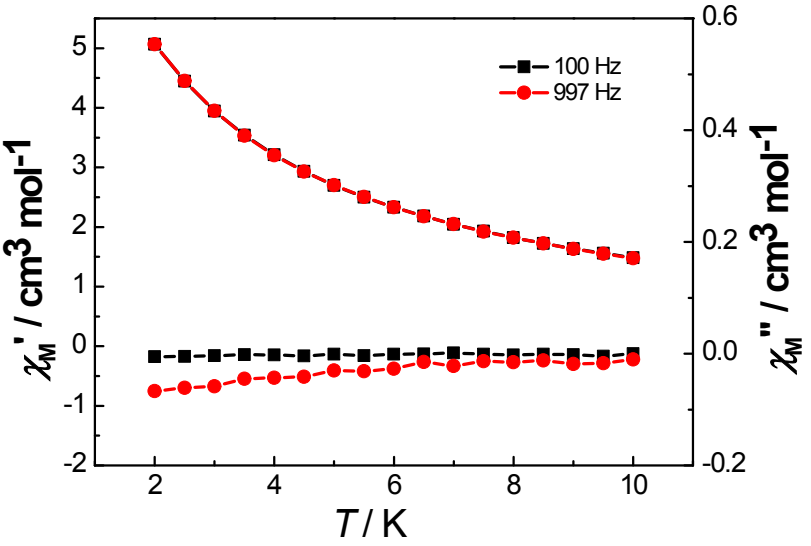

Figure S9. Temperature dependence of the in-phase χ' and out-of-phase χ'' in a 2 Oe ac field oscillating at 100 and 999 Hz with a zero applied dc field for **2**.

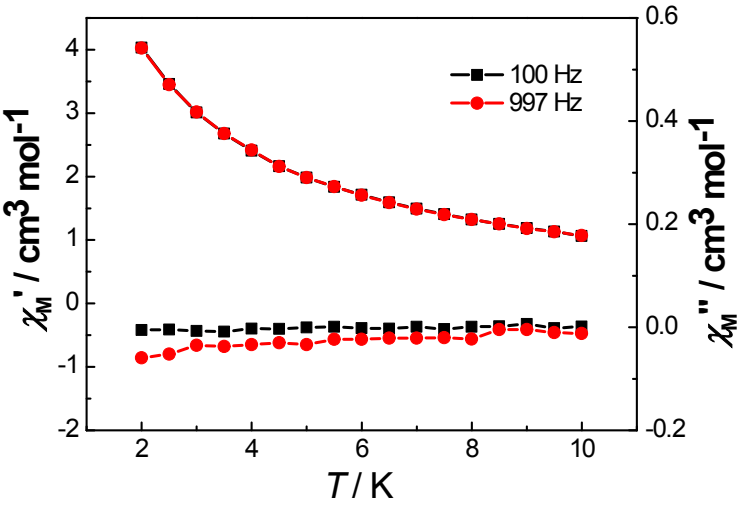

Figure S10. Temperature dependence of the in-phase χ' and out-of-phase χ'' in a 2 Oe ac field oscillating at 100 and 999 Hz with a zero applied dc field for **4**.

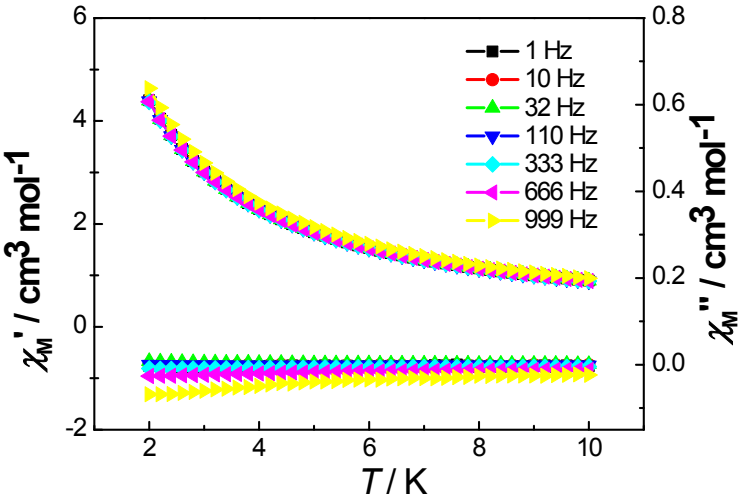

Figure S11. Temperature dependence of the in-phase χ' and out-of-phase χ'' at different frequencies in a 2 Oe ac field oscillating at 1–999 Hz with a zero applied dc field for **5**.

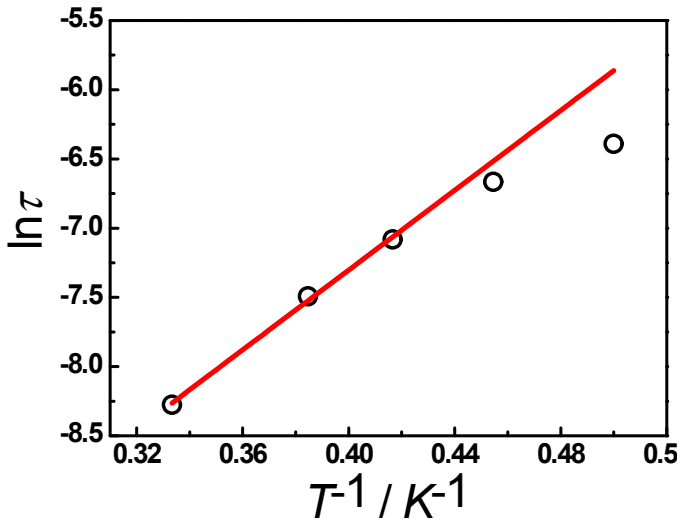

Figure S12. the Arrhenius fit for the $\ln\tau$ vs T^{-1} plot from ac-f data of **3**. The red solid line represents the best fits of the data.

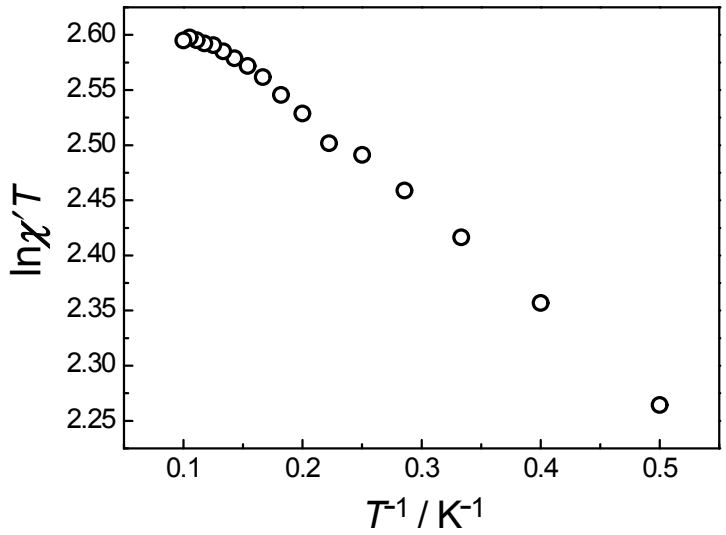

Figure S13. ln($\chi'T$) vs T^{-1} plots on a semi-logarithmic scale from polycrystalline samples of **3**.