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Synthesis, Structures and Magnetism of Heterobinuclear Ni-Ln Complexes: Field-induced Single-molecule Magnetic Behavior in the Dysprosium Analogue

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Supporting information

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

Gd(1)-O(2)	2.336(4)	Gd(1)-O(3)	2.349(5)
Gd(1)-O(6)	2.395(5)	Gd(1)-O(8)	2.401(5)
Gd(1)-O(7)	2.415(5)	Gd(1)-O(5)	2.461(5)
Gd(1)-O(2W)	2.461(4)	Gd(1)-O(4)	2.582(4)
Gd(1)-O(1)	2.631(4)	Gd(1)-Ni(1)	3.491(1)
N(1)-Ni(1)	2.019(5)	N(2)-Ni(1)	2.022(6)
N(3)-Ni(1)	2.074(6)	Ni(1)-O(2)	2.039(5)
Ni(1)-O(3)	2.040(4)	Ni(1)-O(1W)	2.125(4)
O(2)-Gd(1)-O(3)	68.17(2)	O(2)-Gd(1)-O(6)	134.25(2)
O(3)-Gd(1)-O(6)	128.88(2)	O(2)-Gd(1)-O(8)	97.94(2)
O(3)-Gd(1)-O(8)	150.89(2)	O(6)-Gd(1)-O(8)	79.42(2)
O(2)-Gd(1)-O(7)	144.22(2)	O(3)-Gd(1)-O(7)	105.49(2)
O(6)-Gd(1)-O(7)	78.27(2)	O(8)-Gd(1)-O(7)	70.27(2)
O(2)-Gd(1)-O(5)	78.50(2)	O(3)-Gd(1)-O(5)	72.84(2)
O(6)-Gd(1)-O(5)	70.53(2)	O(8)-Gd(1)-O(5)	131.26(2)
O(7)-Gd(1)-O(5)	135.03(2)	O(2)-Gd(1)-O(2W)	73.85(2)
O(3)-Gd(1)-O(2W)	74.77(2)	O(6)-Gd(1)-O(2W)	145.67(2)

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O(8)-Gd(1)-O(2W)	76.80(2)	O(7)-Gd(1)-O(2W)	70.60(2)
O(5)-Gd(1)-O(2W)	143.33(2)	O(2)-Gd(1)-O(4)	128.15(2)
O(3)-Gd(1)-O(4)	62.83(2)	O(6)-Gd(1)-O(4)	72.67(2)
O(8)-Gd(1)-O(4)	133.49(2)	O(7)-Gd(1)-O(4)	68.15(2)
O(5)-Gd(1)-O(4)	72.08(2)	O(2W)-Gd(1)-O(4)	107.37(2)
O(2)-Gd(1)-O(1)	62.52(2)	O(3)-Gd(1)-O(1)	123.17(1)
O(6)-Gd(1)-O(1)	75.91(2)	O(8)-Gd(1)-O(1)	64.52(2)
O(7)-Gd(1)-O(1)	130.99(2)	O(5)-Gd(1)-O(1)	71.37(1)
O(2W)-Gd(1)-O(1)	114.44(1)	O(4)-Gd(1)-O(1)	137.84(2)
N(1)-Ni(1)-N(2)	97.6(2)	N(1)-Ni(1)-O(2)	91.1(2)
N(2)-Ni(1)-O(2)	167.67(2)	N(1)-Ni(1)-O(3)	171.3(2)
N(2)-Ni(1)-O(3)	91.0(2)	O(2)-Ni(1)-O(3)	80.14(2)
N(1)-Ni(1)-N(3)	92.2(2)	N(2)-Ni(1)-N(3)	92.2(2)
O(2)-Ni(1)-N(3)	96.1(2)	O(3)-Ni(1)-N(3)	88.9(2)
N(1)-Ni(1)-O(1W)	88.78(2)	N(2)-Ni(1)-O(1W)	88.15(2)
O(2)-Ni(1)-O(1W)	83.35(2)	O(3)-Ni(1)-O(1W)	90.11(2)
N(3)-Ni(1)-O(1W)	178.9(2)		

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

N(1)-Ni(1)	2.001(5)	N(2)-Ni(1)	2.000(5)
N(5)-Ni(1)	2.285(6)	Ni(1)-O(1)	2.016(4)
Ni(1)-O(2)	2.022(4)	Ni(1)-O(1W)	2.211(4)
Ni(1)-Tb(1)	3.490(8)	O(1)-Tb(1)	2.343(4)
O(2)-Tb(1)	2.326(4)	O(3)-Tb(1)	2.565(4)
O(4)-Tb(1)	2.593(4)	O(5)-Tb(1)	2.393(4)
O(6)-Tb(1)	2.423(4)	O(7)-Tb(1)	2.348(5)
O(8)-Tb(1)	2.448(4)	O(2W)-Tb(1)	2.388(4)
N(1)-Ni(1)-N(2)	97.0(2)	N(1)-Ni(1)-O(1)	91.65(2)
N(2)-Ni(1)-O(1)	171.3(2)	N(1)-Ni(1)-O(2)	166.4(2)
N(2)-Ni(1)-O(2)	92.0(2)	O(1)-Ni(1)-O(2)	79.33(2)
N(1)-Ni(1)-O(1W)	86.11(2)	N(2)-Ni(1)-O(1W)	88.4(2)
O(1)-Ni(1)-O(1W)	91.29(2)	O(2)-Ni(1)-O(1W)	83.91(2)
N(1)-Ni(1)-N(5)	89.3(2)	N(2)-Ni(1)-N(5)	90.4(2)
O(1)-Ni(1)-N(5)	90.60(2)	O(2)-Ni(1)-N(5)	100.9(2)
O(1W)-Ni(1)-N(5)	175.04(2)	O(2)-Tb(1)-O(1)	67.00(1)
O(2)-Tb(1)-O(7)	134.66(2)	O(1)-Tb(1)-O(7)	129.41(2)
O(2)-Tb(1)-O(2W)	74.90(2)	O(1)-Tb(1)-O(2W)	74.75(2)
O(7)-Tb(1)-O(2W)	144.64(2)	O(2)-Tb(1)-O(5)	96.43(2)

O(1)-Tb(1)-O(5)	149.34(2)	O(7)-Tb(1)-O(5)	80.81(2)
O(2W)-Tb(1)-O(5)	76.06(2)	O(2)-Tb(1)-O(6)	145.39(2)
O(1)-Tb(1)-O(6)	107.87(2)	O(7)-Tb(1)-O(6)	76.41(2)
O(2W)-Tb(1)-O(6)	70.88(2)	O(5)-Tb(1)-O(6)	70.59(2)
O(2)-Tb(1)-O(8)	78.35(1)	O(1)-Tb(1)-O(8)	72.02(1)
O(7)-Tb(1)-O(8)	71.16(1)	O(2W)-Tb(1)-O(8)	143.40(1)
O(5)-Tb(1)-O(8)	131.95(2)	O(6)-Tb(1)-O(8)	134.42(2)
O(2)-Tb(1)-O(3)	127.59(1)	O(1)-Tb(1)-O(3)	63.10(1)
O(7)-Tb(1)-O(3)	73.37(2)	O(2W)-Tb(1)-O(3)	105.60(2)
O(5)-Tb(1)-O(3)	135.34(2)	O(6)-Tb(1)-O(3)	68.31(2)
O(8)-Tb(1)-O(3)	72.24(1)	O(2)-Tb(1)-O(4)	63.40(2)
O(1)-Tb(1)-O(4)	122.05(1)	O(7)-Tb(1)-O(4)	75.00(2)
O(2W)-Tb(1)-O(4)	117.06(2)	O(5)-Tb(1)-O(4)	64.96(2)
O(6)-Tb(1)-O(4)	129.96(2)	O(8)-Tb(1)-O(4)	70.42(2)
O(3)-Tb(1)-O(4)	136.99(2)		

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

Dy(1)-O(2)	2.310(4)	Dy(1)-O(3)	2.320(4)
Dy(1)-O(7)	2.326(5)	Dy(1)-O(5)	2.376(4)
Dy(1)-O(2W)	2.377(5)	Dy(1)-O(6)	2.397(4)
Dy(1)-O(8)	2.450(4)	Dy(1)-O(1)	2.566(4)
Dy(1)-O(4)	2.615(4)	Dy(1)-Ni(1)	3.482(9)
N(1)-Ni(1)	1.994(5)	N(2)-Ni(1)	2.004(6)
N(3)-Ni(1)	2.204(5)	Ni(1)-O(3)	2.053(4)
Ni(1)-O(2)	2.056(4)	Ni(1)-O(1W)	2.146(4)
O(2)-Dy(1)-O(3)	69.00(1)	O(2)-Dy(1)-O(7)	129.20(2)
O(3)-Dy(1)-O(7)	135.49(2)	O(2)-Dy(1)-O(5)	149.69(2)
O(3)-Dy(1)-O(5)	94.74(1)	O(7)-Dy(1)-O(5)	80.48(2)
O(2)-Dy(1)-O(2W)	74.80(2)	O(3)-Dy(1)-O(2W)	74.33(2)
O(7)-Dy(1)-O(2W)	143.78(1)	O(5)-Dy(1)-O(2W)	76.22(2)
O(2)-Dy(1)-O(6)	107.50(1)	O(3)-Dy(1)-O(6)	144.70(2)
O(7)-Dy(1)-O(6)	75.27(1)	O(5)-Dy(1)-O(6)	70.68(1)
O(2W)-Dy(1)-O(6)	71.05(1)	O(2)-Dy(1)-O(8)	72.25(1)
O(3)-Dy(1)-O(8)	79.95(1)	O(7)-Dy(1)-O(8)	71.60(1)
O(5)-Dy(1)-O(8)	131.78(2)	O(2W)-Dy(1)-O(8)	143.69(1)
O(6)-Dy(1)-O(8)	133.91(1)	O(2)-Dy(1)-O(1)	62.91(1)
O(3)-Dy(1)-O(1)	129.41(1)	O(7)-Dy(1)-O(1)	72.52(1)
O(5)-Dy(1)-O(1)	135.30(1)	O(2W)-Dy(1)-O(1)	106.10(2)

O(6)-Dy(1)-O(1)	68.41(1)	O(8)-Dy(1)-O(1)	71.62(1)
O(2)-Dy(1)-O(4)	123.04(1)	O(3)-Dy(1)-O(4)	63.53(2)
O(7)-Dy(1)-O(4)	74.80(2)	O(5)-Dy(1)-O(4)	64.86(1)
O(2W)-Dy(1)-O(4)	117.93(2)	O(6)-Dy(1)-O(4)	129.42(1)
O(8)-Dy(1)-O(4)	70.21(1)	O(1)-Dy(1)-O(4)	135.71(1)
N(1)-Ni(1)-N(2)	97.2(2)	N(1)-Ni(1)-O(3)	167.48(2)
N(2)-Ni(1)-O(3)	91.4(2)	N(1)-Ni(1)-O(2)	91.98(2)
N(2)-Ni(1)-O(2)	170.7(2)	O(3)-Ni(1)-O(2)	79.33(2)
N(1)-Ni(1)-O(1W)	88.05(2)	N(2)-Ni(1)-O(1W)	89.9(2)
O(3)-Ni(1)-O(1W)	82.89(2)	O(2)-Ni(1)-O(1W)	89.30(2)
N(1)-Ni(1)-N(3)	88.5(2)	N(2)-Ni(1)-N(3)	90.8(2)
O(3)-Ni(1)-N(3)	100.5(2)	O(2)-Ni(1)-N(3)	90.60(2)
O(1W)-Ni(1)-N(3)	176.50(2)		

Table S4. Results of the Continuous Shape Measure Analysisa geometry[a]

Geometry	MFF-9	CSAPR-9	JCSAPR-9	TCTPR-9
1	**0.70**	0.90	1.42	1.98
2	**0.81**	1.24	1.78	2.10
3	**0.73**	1.01	1.58	2.06
18a-Dy	1.53	**1.22**	2.86	**1.32**

[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.

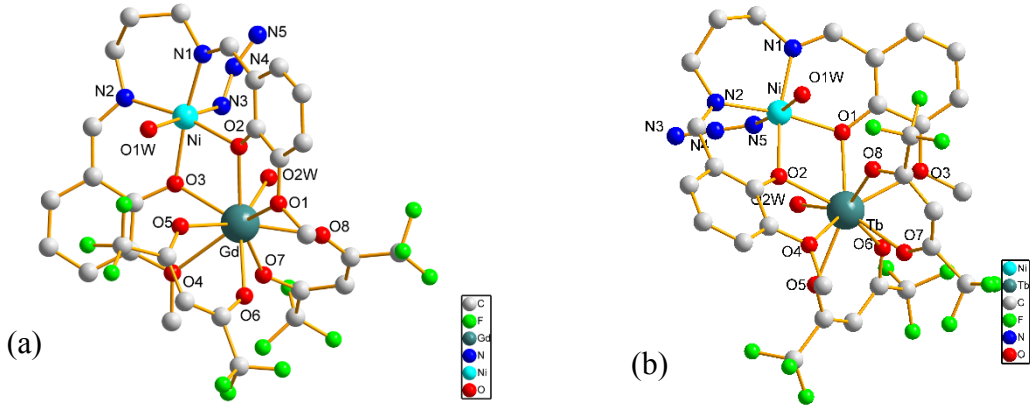

Figure S1. (a) Perspective drawing of the crystallographically structural unit of **1** showing the atom numbering. (b) Perspective drawing of the crystallographically structural unit of **2** showing the atom numbering. H atoms are omitted for clarity.

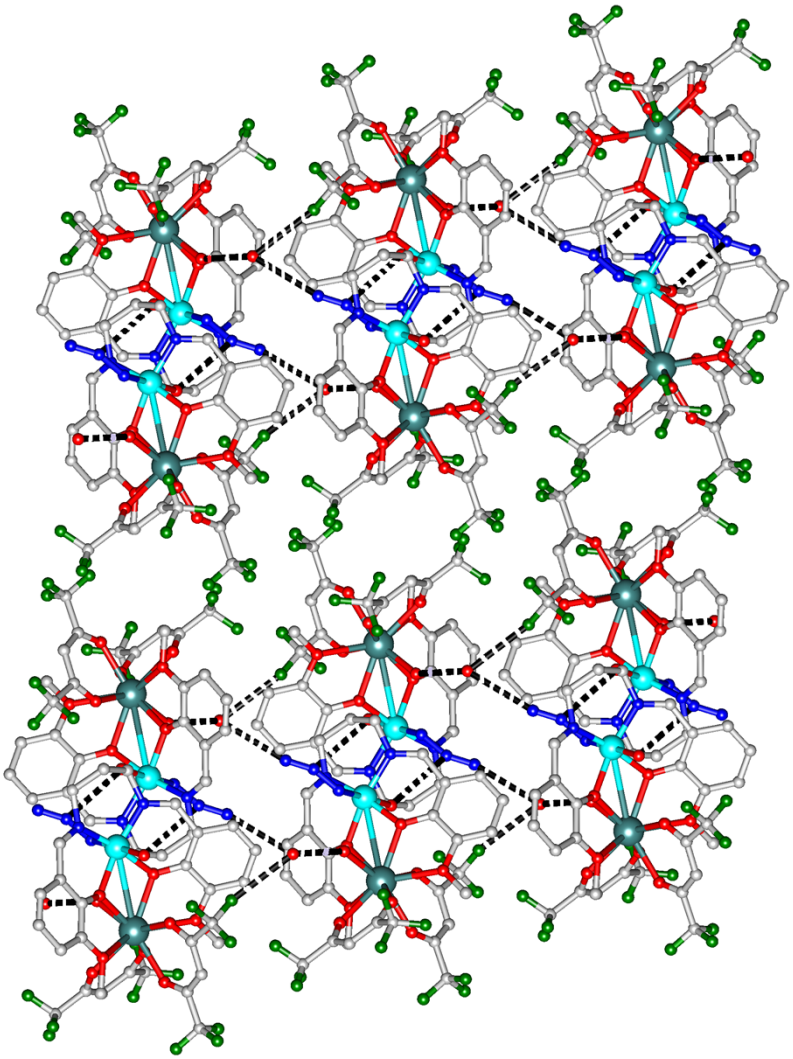

Figure S2. A view showing 3D structure formed by weak H-bonding interactions (black dash lines) and Van der Waals' interactions in **1–3**.

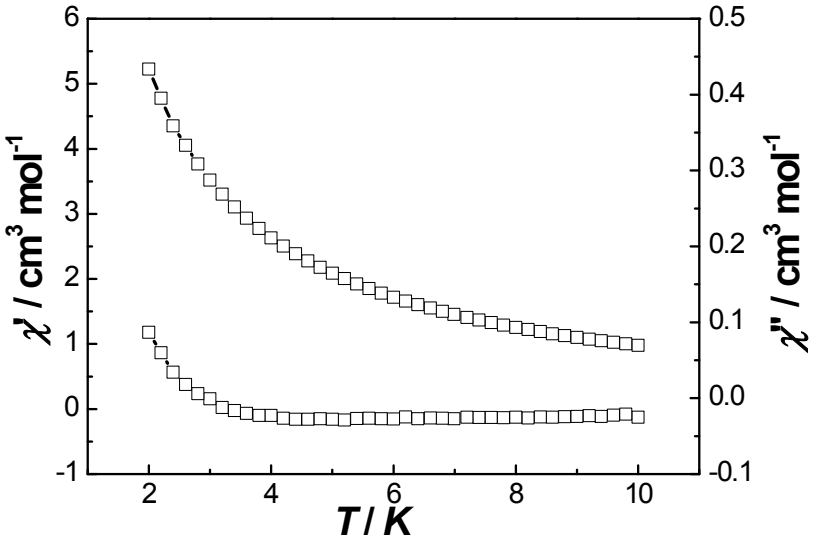

Figure S3. Temperature dependence of the in-phase χ' and out-of-phase χ'' in a 2 Oe ac field oscillating at 1000 Hz with a zero applied dc field for **3**.