ELECTRONIC SUPPLEMENTARY INFORMATION

In search for molecules displaying ferromagnetic exchange: multiple-decker Ni_{12} and Ni_{16} complexes from the use of pyridine-2-amidoxime

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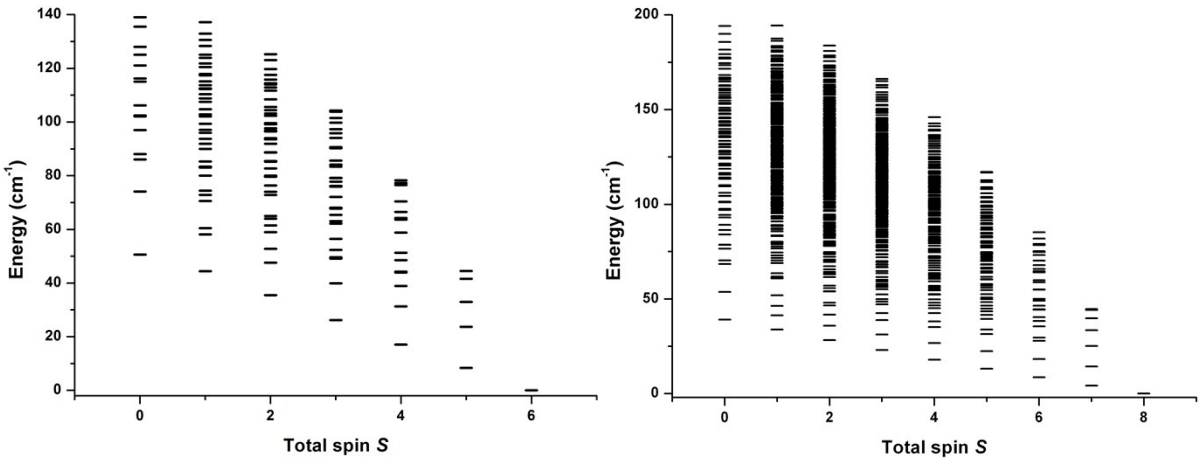

Fig. S1. Plot of energy versus total spin state for **1** (left) and **2** (right), derived from the isotropic fit of the susceptibility.

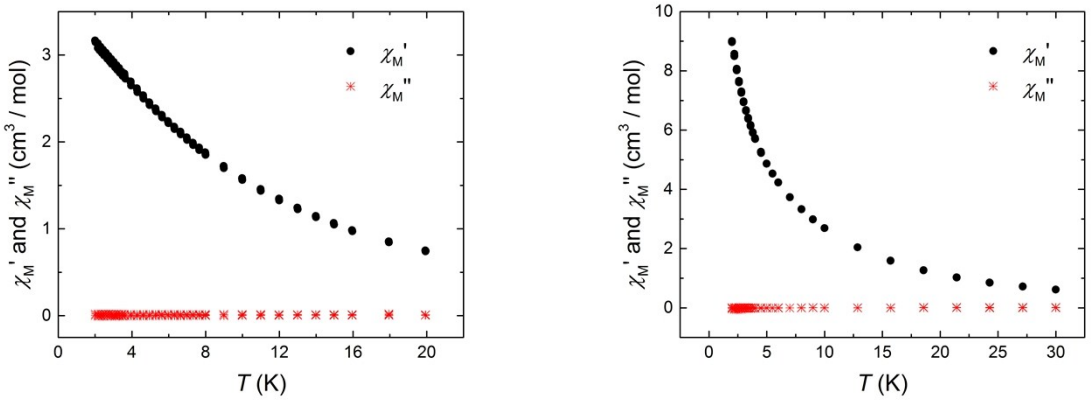

Fig. S2. Plot of the *ac* susceptibility for **1** (left) and **2** (right). The data were collected in zero applied *dc* field, a drive amplitude of 4.5 Oe and for 33, 333 and 1333 Hz, though they show no dependence on the frequency, at least down to $T = 2$ K.

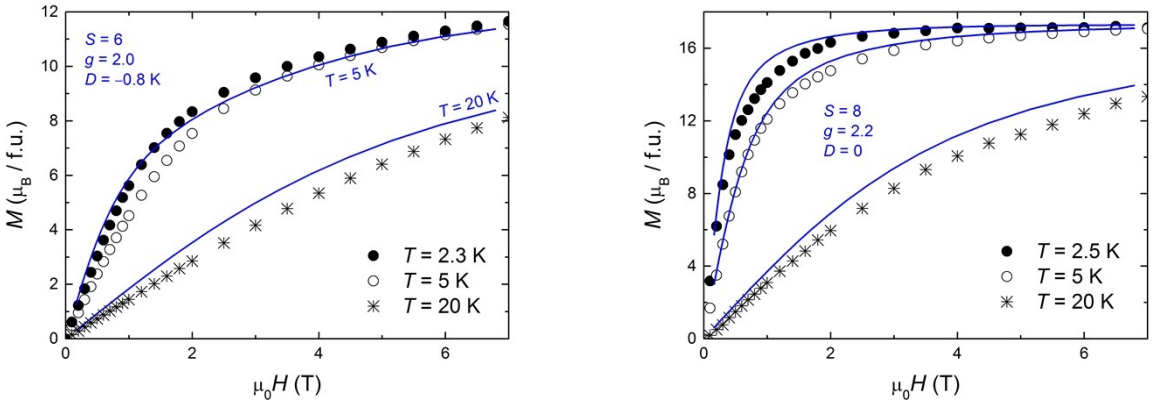

Fig. S3. Magnetization *vs.* field data for **1** (left) and **2** (right) at three different temperatures. The solid lines represent calculated curves for uncorrelated cations at the corresponding temperatures, assuming $S = 6$, $g = 2.0$ and uniaxial anisotropy $D = -0.8$ K for **1** and $S = 8$, $g = 2.2$ and $D = 0$ for **2**.

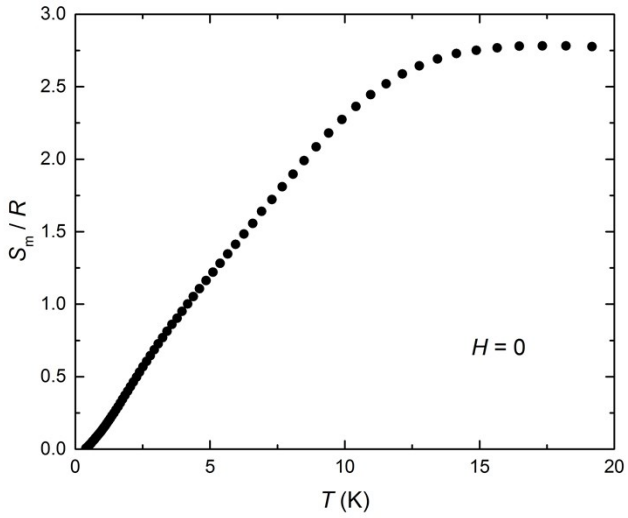

Fig. S4. Zero-applied-field magnetic entropy S_m, normalised to the gas constant R, *vs.* T in the indicated temperature range for **1**.

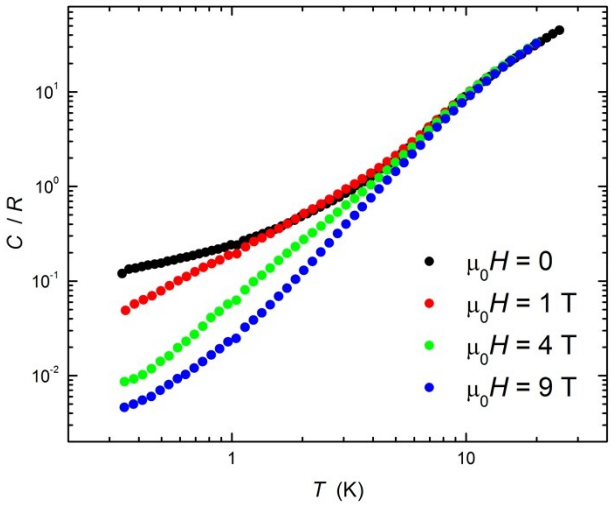

Fig. S5. Specific heat *C*, normalised to the gas constant *R*, *vs. T* in the indicated temperature and field ranges for **2**.

Table S1. Selective interatomic distances and angles in complex **2**·10H_2O·26MeOH [a]

Ni1-O9	1.983(3)	Ni4-O4	2.104(3)
Ni1-O6′	2.014(3)	Ni4-N1	2.237(3)
Ni1-N18	2.041(3)	Ni5-O5′	1.826(2)
Ni1-N16	2.069(3)	Ni5-N19	1.847(3)
Ni1-O1	2.158(3)	Ni5-N2′	1.854(3)
Ni1-O10	2.170(3)	Ni5-N21	1.890(3)
Ni2-O5′	2.019(2)	Ni6-O6′	1.823(3)
Ni2-N15	2.041(3)	Ni6-N5′	1.843(3)
Ni2-O1	2.042(2)	Ni6-N22	1.864(3)
Ni2-N13	2.096(3)	Ni6-N24	1.884(3)
Ni2-O2	2.113(2)	Ni7-O8′	1.822(3)
Ni2-N16	2.374(3)	Ni7-N7	1.844(3)
Ni3-O7′	2.023(2)	Ni7-N17′	1.847(3)
Ni3-O2	2.037(2)	Ni7-N8	1.890(3)
Ni3-N3	2.066(3)	Ni8-O7′	1.824(2)
Ni3-N1	2.076(3)	Ni8-N10	1.846(3)
Ni3-O3	2.164(2)	Ni8-N14′	1.860(3)
Ni3-N13	2.249(3)	Ni8-N12	1.903(3)
Ni4-N4	2.022(3)	Ni1···Ni2	3.189(2)

Ni4-O8′	2.032(2)	Ni1···Ni6	3.312(2)
Ni4-O3	2.046(2)	Ni1···Ni4′	3.956(2)
Ni4-N6	2.077(3)	Ni1···Ni7′	4.671(2)
O6′-Ni1-N18	172.84(12)	N19-Ni5-N2′	173.48(14)
O9-Ni1-N16	170.19(11)	O5′-Ni5-N21	173.10(12)
O1-Ni1-O10	174.22(11)	N2′-Ni5-N21	101.87(14)
O5′-Ni2-N15	176.44(11)	O6′-Ni6-N22	90.66(12)
O1-Ni2-N13	173.36(10)	N5′-Ni6-N22	173.56(15)
O2-Ni2-N16	172.79(10)	O6′-Ni6-N24	173.29(13)
O7′-Ni3-N3	178.81(11)	N7-Ni7-N17′	174.21(14)
O2-Ni3-N1	173.16(11)	O8′-Ni7-N8	171.63(12)
O3-Ni3-N13	176.65(10)	N17′-Ni7-N8	102.36(14)
N4-Ni4-O3	171.99(11)	N10-Ni8-N14′	173.17(13)
O8′-Ni4-N6	173.26(11)	O7′-Ni8-N12	171.24(12)
O4-Ni4-N1	175.38(11)	N14′-Ni8-N12	103.89(13)

[a] Symmetry transformations used to generate equivalent atoms:

′: -x+1,-y+1,-z+1

Table S2. Hydrogen Bonds for Complex **2**·10H_2O·26MeOH [a]

D–H···A	D···A [Å]	D-H···A [deg]	Symmetry Operator of A
N9–H9A···O14	2.805(2)	167.4(1)	*x*, *y*, *z*
N11–H11A···O11	2.880(2)	162.5(1)	*x*, *y*, *z*
N23-H23A··· O13	2.908(2)	162.7(1)	*x, y,* 1+*z*
N14-H14A···O17	2.935(2)	163.0(1)	*x*, *y*, *z*
O4-H4A···O24	2.778(2)	177.4(1)	*x*, *y*, *z*
N5-H5A···O27	3.051(2)	170.5(1)	*x*, *y*, *z*
O10-H10A···O24	2.704(2)	169.8(1)	*x*, *y*, *z*
O32-H32A···O12	2.736(2)	170.8(1)	*x*, *y*, *z*
O24-H24A···O13	2.605(2)	176.3(1)	*x*, *y*, *z*
O33-H33A···O12	2.920(2)	158.8(1)	*x*, *y*, *z*
O28-H28A···O18	2.758(2)	158.9(1)	*x*, *y*, *z*
O27-H27···O15	2.683(2)	169.8(1)	*x*, *y*, *z*
O26-H26···O17	2.793(2)	166.6(1)	*x*, *y*, *z*
O31-H31A···O33A	2.710(2)	120.4(1)	*x*, *y*, *z*
O20-H20···O15	2.717(2)	178.2(1)	*x*, *y*, *z*
O29-H29A···O4W	2.796(2)	164.9(1)	*x*, *y*, *z*

[a] A = acceptor atom, D = donor atom.