Supporting Information

Anion Dependent Self-Assembly of Sandwich 13-metal Ni-Ln Nanoclusters with a long-chain Schiff base ligand

Shiqing Wang, Xiaoping Yang, Jinjie Qian, Qipeng Li, Zhongning Chen, Lijie Zhang, Shaoming Huang, Chengri Wang and Richard A. Jones

Contents

1. General Procedures

All reactions were performed under dry oxygen-free dinitrogen atmospheres using standard Schlenk techniques. The H_2salen ligand H_2L was prepared according to well-established procedures.[1] Physical measurements: NMR: VARIAN UNITY-plus. 600 spectrometer (^{1}H, 600 MHz) at 298 K; Powder XRD: SMART APE II DUO; IR: FTIR-650 spectrometer. Melting points were obtained in sealed glass capillaries and are uncorrected. Elemental analyses (C, H, N) were carried out on a EA1112 elemental analysis. Transmission electron microscopy (TEM) images were recorded on a JEOL JEM-1200EX transmission electron microscope. Field emission scanning electron microscopy (FESEM) images were recorded on a Nova NanoSEM 200 scanning electron microscope. Magnetic measurements were performed on a Quantum Design MPMS (SQUID)-XL magnetometer and PPMS-9T system.

Ref. (1) F. Lam, J.-X. Xu, K.-S. Chan, *J. Org. Chem.*, **1996**, *61*, 8414-8418.

2. Synthesis of complexes 1-4

$[Gd_4Ni_3L_3(OAc)_6(NO_3)_3(OH)_3]$ (**1**). $Ni(OAc)_2 \cdot 4H_2O$ (0.4 mmol, 0.0707 g), $Gd(NO_3)_3 \cdot 6H_2O$ (0.4 mmol, 0.1805g) and H_2L (0.4 mmol, 0.1696g) were dissolved in 5 mL MeOH and 10mL EtOH at room temperature, and a solution of NEt_3 in EtOH (0.35 mol/L, 2 ml) was then added. The resulting solution was stirred and heated under reflux for 15 mins. It was allowed to cool and was then filtered. Diethyl ether was allowed to diffuse slowly into the filtrate at room temperature and pale green crystals were obtained after two weeks. The crystals were filtered off, washed with EtOH (5 ml) and dried in the air for one week. Yield (based on $Gd(NO_3)_3 \cdot 6H_2O$): 0.0589 g (21 %). m. p. > 221°C (dec.). Elemental analysis: Found: C, 35.67; H, 4.96; N, 7.44 %. Calc. for $C_{80}H_{102}Ni_3N_{18}Gd_4O_{41}(EtOH)_7(MeOH)(H_2O)_5$: C, 35.31; H, 4.94; N, 7.80 %. IR (cm^{-1}): 1630 (m), 1570 (m), 1470 (s), 1410 (s), 1300 (m), 1240 (s), 1080 (m), 971 (w), 855(w), 739(s).

$[Tb_4Ni_3L_3(OAc)_6(NO_3)_3(OH)_3]$ (**2**). The procedure was the same as that for **1** using $Tb(NO)_3 \cdot H_2O$ (0.4 mmol, 0.1380g). Pale green single crystals of **2** were formed after two weeks. Yield (based on $Tb(NO_3)_3 \cdot 6H_2O$): 0.0685 g (22 %). m. p. > 224°C (dec.). Elemental analysis: Found: C, 35.565; H, 4.637; N, 7.423 %. Calc. for $C_{80}H_{102}Ni_3N_{18}Tb_4O_{41}(EtOH)_7(MeOH)(H_2O)_5$: C, 35.21; H, 4.62; N, 7.79 %. IR (cm^{-1}): 1630 (s), 1580 (m), 1470 (s), 1410 (s), 1300 (m), 1220 (s), 1080 (m), 970 (w), 849(w), 742(s).

$[Gd_6Ni_7L_6(OAc)_{12}(OH)_6](OH)_2$ (**3**). The procedure was the same as that for **1** using $Gd(OAc)_3 \cdot 6H_2O$ (0.4 mmol,0.1770g). Pale green single crystals of **3** were formed after two weeks. Yield (based on $Gd(OAc)_3 \cdot 6H_2O$): 0.0748 g (15 %). m. p. > 188 °C (dec.). Elemental analysis: Found: C, 39.08; H, 5.13; N, 3.62 %. Calc. for $C_{168}H_{210}Ni_7N_{12}Gd_6O_{54}(EtOH)_2(MeOH)_2(H_2O)_{10}$: C, 39.20; H, 4.76; N, 3.17 %. IR (cm^{-1}): 1630 (s), 1580 (m), 1470 (s), 1410 (s), 1300 (m), 1250 (m), 1240 (m), 1130 (w), 1110 (m), 1080 (m), 966 (w),850(w),742(s).

$[Dy_6Ni_7L_6(OAc)_{12}(OH)_6](OH)_2$ (**4**). The procedure was the same as that for **4** using $Dy(OAc)_3 \cdot 4H_2O$ (0.4 mmol, 0.1647g). Pale green single crystals of **4** were formed after two weeks. Yield (based on $Dy(OAc)_3 \cdot 4H_2O$) : 0.0932 g (17 %). m. p. > 189 °C (dec.). Elemental analysis: Found: C, 38.86; H, 5.02; N, 3.61 %. Calc. for $C_{168}H_{210}Ni_7N_{12}Gd_6O_{54}(EtOH)_2(MeOH)_2(H_2O)_{10}$: C, 38.97; H, 4.74; N, 3.15 %. IR (cm^{-1}): 1630 (s), 1580 (m), 1470 (s), 1410 (s), 1300 (m), 1220 (s), 1100 (m), 973 (w), 858(w),739(s).

3. ^{1}H NMR spectra of the ligand H_2L

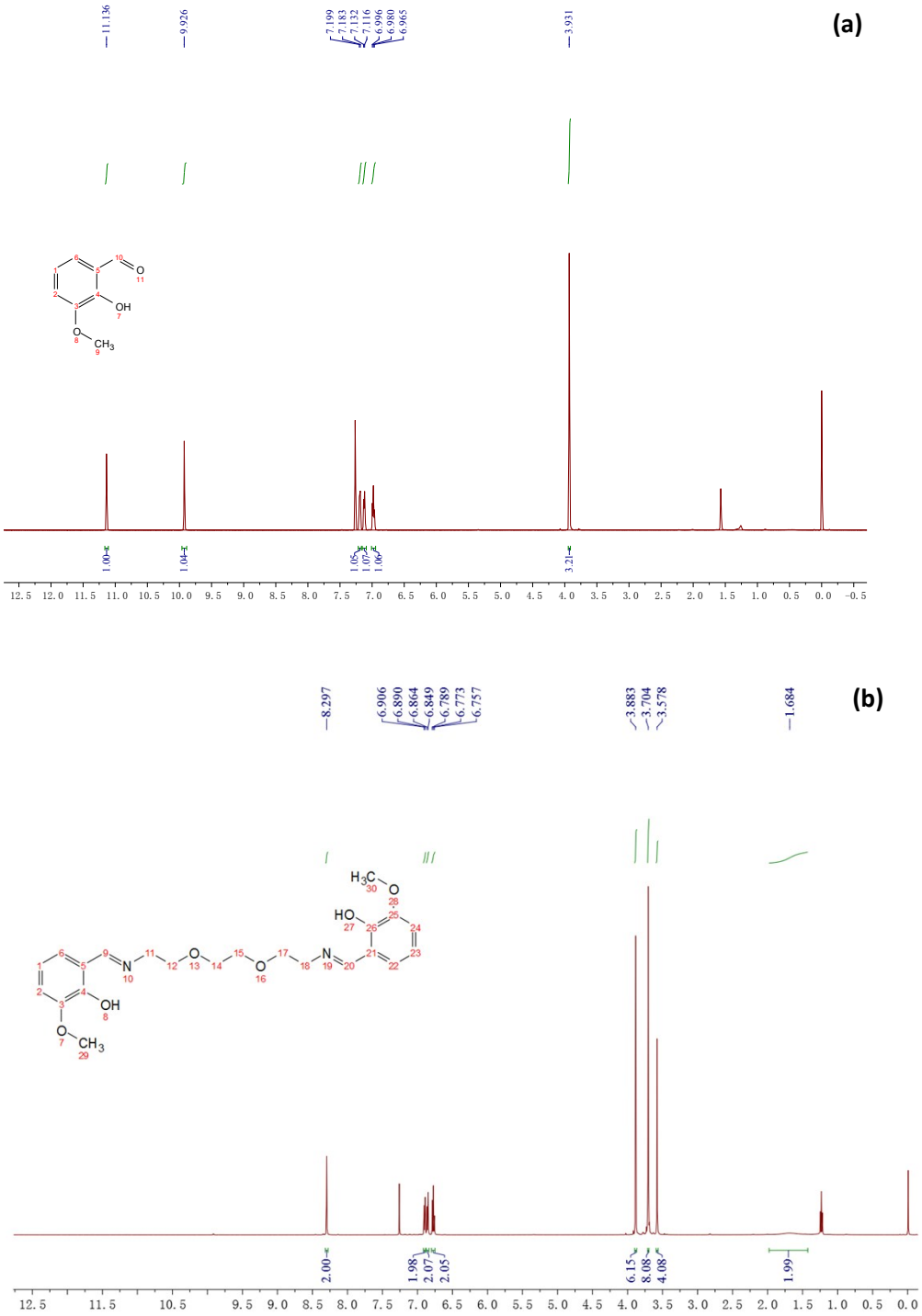
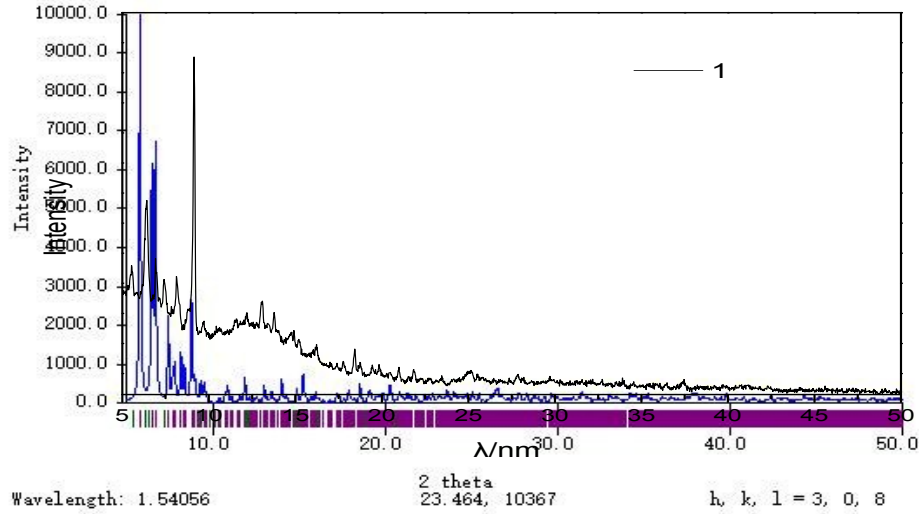


Fig. S1. ^{1}H NMR spectra of reactant 3-methoxyl-2-hydroxyl-benzaldehyde (a) and H_2L (b) in $CDCl_3$ at 298K.

4. Powder XRD patterns of 1 and 3



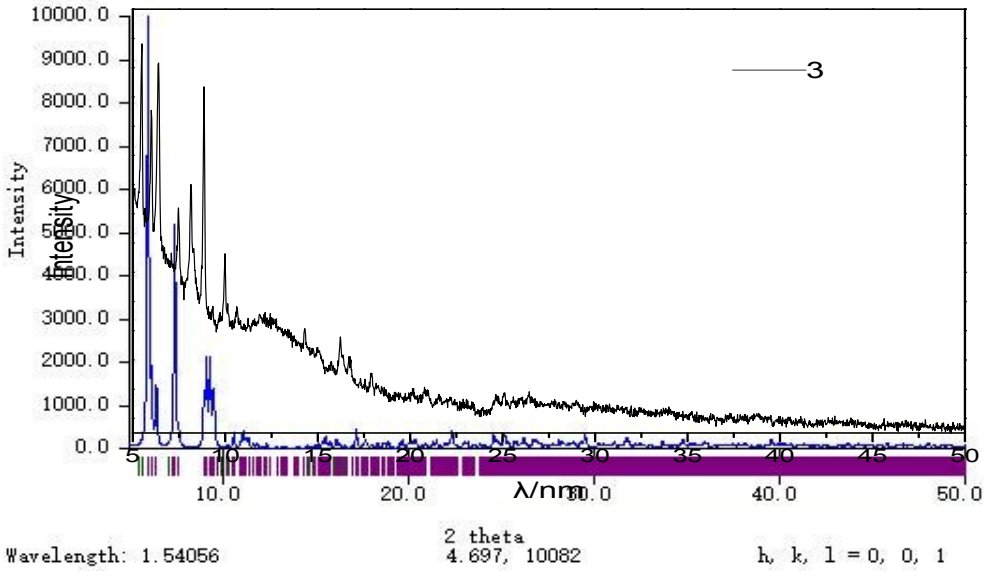


Fig. S2. Powder XRD patterns of **1** and **3** (the insert pictures were obtained from experiments).

5. The thermogravimetric analysis of 1 and 3

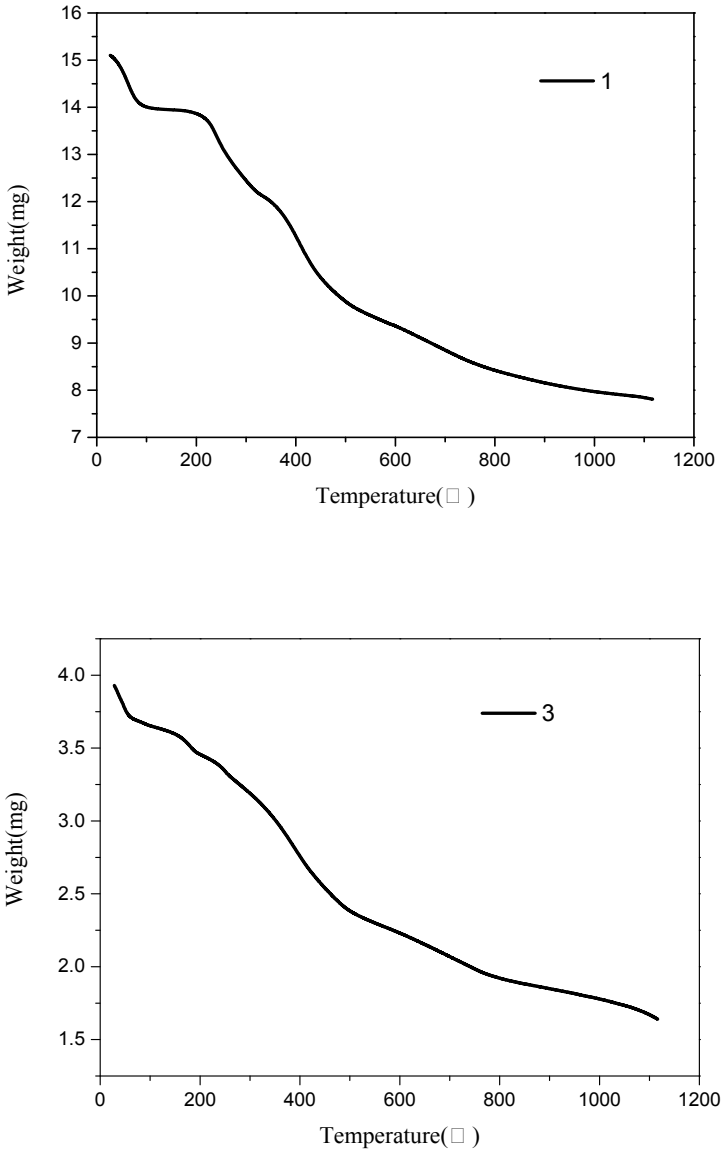
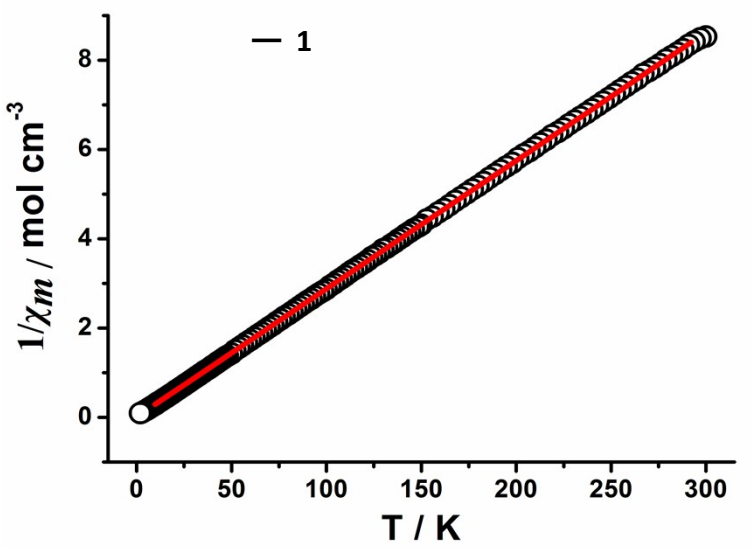
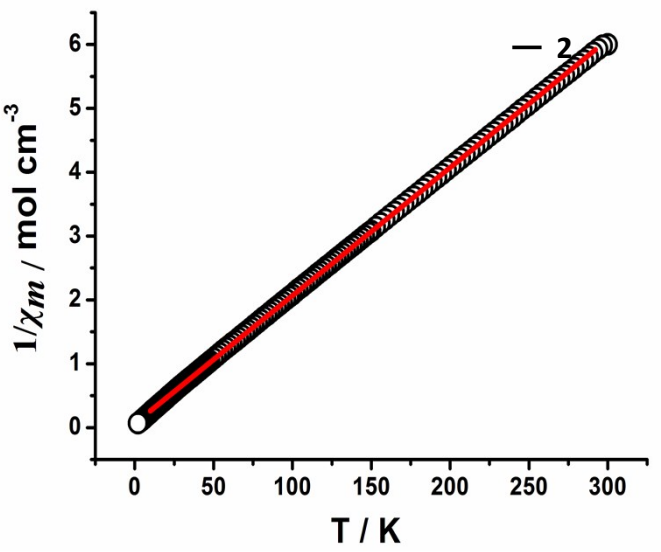


Fig. S3. The thermogravimetric analysis of **1** and **3**.

6. The magnetic properties of 1-4





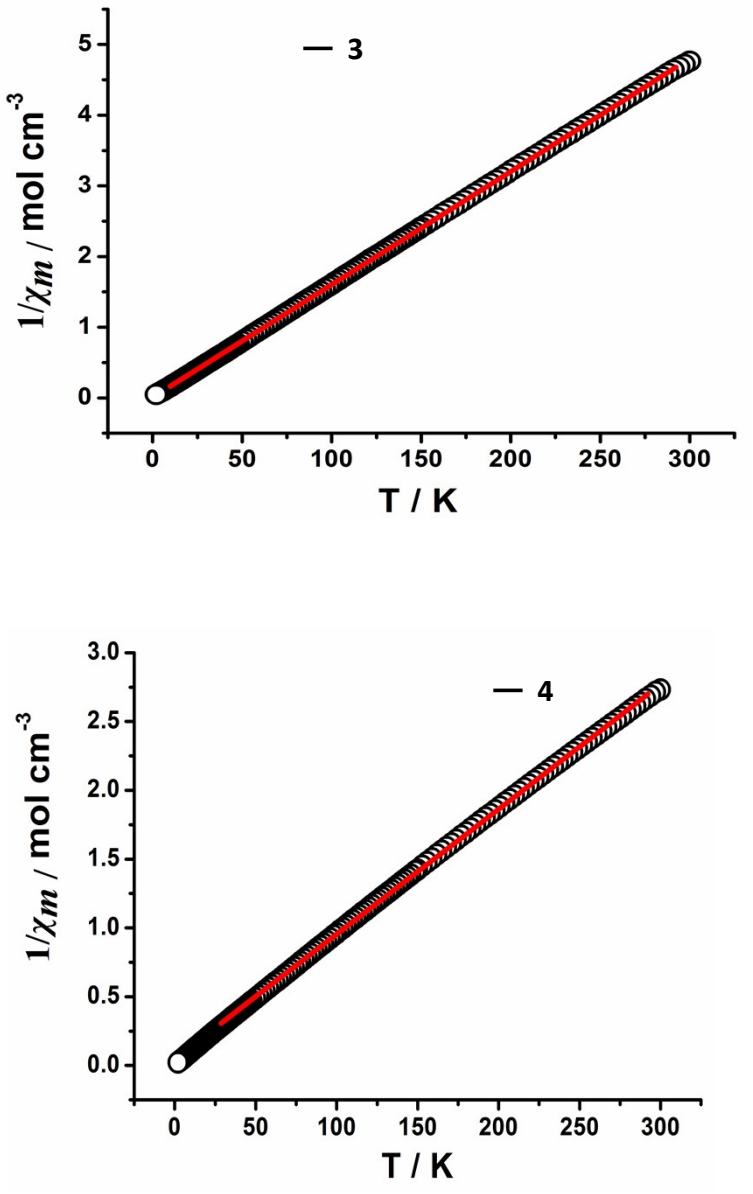


Fig. S4. The Curie–Weiss fitting for **1**-**4** (red lines).

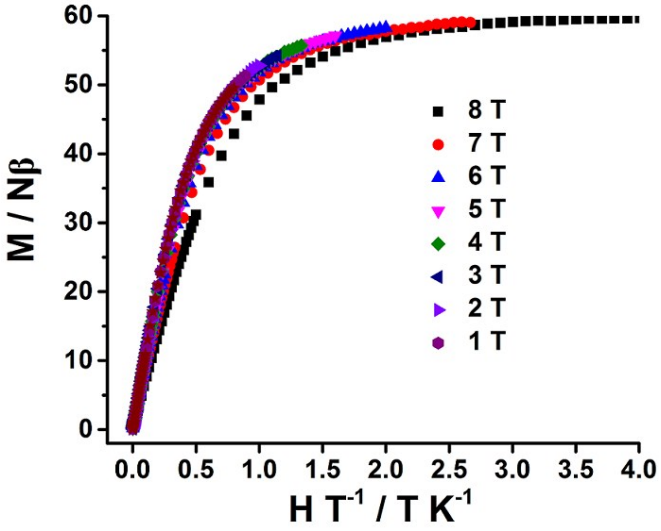


Fig. S5. Plots of reduced magnetization (M/Nβ) vs. H/T in the ranges 2-8 K and 1-8 T for **4**.

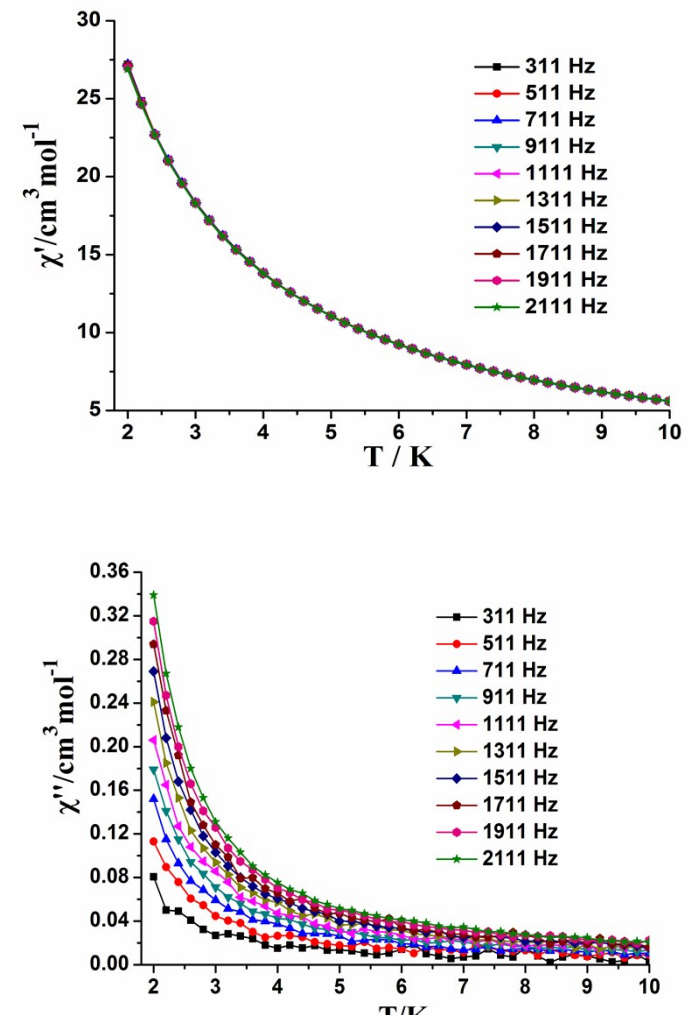


Fig. S6. Frequency dependence of the in-phase and out-of-phase ac susceptibility for **4** measured under zero static field.

7. X-Ray Crystallography

Data were collected on a Smart APEX CCD diffractometer with graphite monochromated Mo-Kα radiation (λ = 0.71073 Å) at 190 K. The data set was corrected for absorption based on multiple scans and reduced using standard methods. Data reduction was performed using DENZO-SMN.[1] The structures were solved by direct methods and refined anisotropically using full-matrix least-squares methods with the SHELX 97 program package.[2] Coordinates of the non-hydrogen atoms were refined anisotropically, while hydrogen atoms were included in the calculation isotropically but not refined. Neutral atom scattering factors were taken from Cromer and Waber.[3] PLATON98 was used as incorporated in WinGX (Farrugia, 1999).

ISOR was applied to all non-hydrogen atoms to allow their Uij components approximate to isotropic behavior. Hydrogen atoms of water molecules were not added. Crystallographic data for **1-4** are presented in Table S1 and selected bond lengths are given in Tables S2-S5. (CCDC reference numbers 1499598-1499601. See http://www.rsc.org/suppdata/cc/ for crystallographic data in CIF format.)

Ref. (1) DENZO-SMN. (**1997**). Z. Otwinowski, W. Minor, *Methods in Enzymology*, 276: *Macromolecular Crystallography, Part A*, 307 – 326, C. W. J. Carter, M. I. Simon, R. M. Sweet, Editors, Academic Press.

(2) G. H. Sheldrick, SHELX 97, *A software package for the solution and refinement of X-ray data*; University of Göttingen: Göttingen, Germany, **1997**.

(3) D. T. Cromer, J. T. Waber, *International Tables for X-Ray Crystallography*, Kynoch Press, Birmingham, vol. 4, **1974**, Table 2.2A.

Table S1. Crystal data and structure refinement for **1-4**.

	1	**2**	**3**	**4**
Formula	$C_{86}H_{118}Gd_4N_8Ni_3O_{45}$	$C_{86}H_{118}Tb_4N_8Ni_3O_{47}$	$C_{164}H_{220}Gd_6N_{12}Ni_7O_{80}$	$C_{160}H_{208}Dy_6N_{12}Ni_7O_{81}$
Fw	2787.01	2813.69	4993.99	4981.35
Crystal system	Monoclinic	Monoclinic	Monoclinic	Monoclinic
Space group	P2(1)/c	P2(1)/c	P2(1)/n	P2(1)/n
a [Å]	19.202(3)	18.9474(17)	17.0972(10)	17.0562(9)
b [Å]	14.379(2)	14.2777(13)	37.019(2)	37.109(2)
c [Å]	42.671(7)	42.613(4)	17.7843(10)	17.9813(10)
α [deg]	90	90	90	90
β [deg]	98.352(2)	97.8690(10)	115.1690(10)	114.9780(10)
γ [deg]	90	90	90	90
V / [Å^3]	11657(3)	11419.3(18)	10187.4(10)	10316.6(10)
d / [g/cm^3]	1.588	1.637	1.628	1.604
Z	4	4	2	2
T [K]	190(1)	190(1)	190(1)	190(1)
F(000)	5556	5612	5016	4984
μ, mm^{-1}	2.800	3.014	2.643	2.854
θ rang, deg	0.96-25.00	1.35-25.00	1.38-25.00	1.38-25.00
reflns meads	86141	41749	76403	63077
reflns used	20450	14053	17879	18037
params	1351	1369	1224	1226
R1[a], wR2[a] [$I > 2\sigma(I)$]	0.0466, 0.1229	0.0550, 0.1731	0.0956, 0.2613	0.0981, 0.2614
R1, wR2 (all data)	0.0635, 0.1389	0.0707, 0.2184	0.1148, 0.2823	0.1203, 0.2855
Quality of fit	1.125	1.054	1.037	1.018

[a] $R1 = \Sigma||F_o| - |F_c||\Sigma|F_o|$. $wR2 = [\Sigma w[(F_o^2 - F_c^2)^2]/\Sigma|[w(F_o^2)^2]]^{1/2}$. $w=1/[\sigma^2(F_o^2)+(0.075P)^2]$, where $P = [\max(F_o^2,0)+2F_c^2]/3$.

Table S2. Selected Bond Lengths (Å) and Angles (°) for **1**.

Gd(1)-O(34)	2.329(5)	Ni(3)-O(41)	2.243(5)
Gd(1)-O(31)	2.329(6)	O(34)-Gd(1)-O(31)	77.8(2)
Gd(1)-O(32)	2.376(6)	O(34)-Gd(1)-O(32)	74.6(2)
Gd(1)-O(25)	2.420(6)	O(31)-Gd(1)-O(32)	76.5(2)
Gd(1)-O(22)	2.441(6)	O(34)-Gd(1)-O(25)	128.8(2)
Gd(1)-O(28)	2.447(6)	O(31)-Gd(1)-O(25)	147.9(2)
Gd(1)-O(29)	2.506(6)	O(32)-Gd(1)-O(25)	92.5(2)
Gd(1)-O(26)	2.537(6)	O(34)-Gd(1)-O(22)	87.3(2)
Gd(1)-O(23)	2.565(7)	O(31)-Gd(1)-O(22)	126.5(2)
Gd(1)-N(8)	2.904(7)	O(32)-Gd(1)-O(22)	147.4(2)
Gd(1)-N(7)	2.911(8)	O(25)-Gd(1)-O(22)	77.8(2)
Gd(2)-O(33)	2.262(5)	O(34)-Gd(1)-O(28)	147.3(2)
Gd(2)-O(19)	2.365(5)	O(31)-Gd(1)-O(28)	88.8(2)
Gd(2)-O(20)	2.384(5)	O(32)-Gd(1)-O(28)	131.4(2)
Gd(2)-O(38)	2.386(5)	O(25)-Gd(1)-O(28)	75.9(2)
Gd(2)-O(8)	2.387(5)	O(22)-Gd(1)-O(28)	76.8(2)
Gd(2)-O(5)	2.440(5)	O(34)-Gd(1)-O(29)	143.9(2)
Gd(2)-O(36)	2.447(5)	O(31)-Gd(1)-O(29)	74.1(2)
Gd(2)-O(7)	2.517(5)	O(32)-Gd(1)-O(29)	77.06(19)
Gd(3)-O(35)	2.292(5)	O(25)-Gd(1)-O(29)	74.0(2)
Gd(3)-O(19)	2.311(5)	O(22)-Gd(1)-O(29)	127.8(2)
Gd(3)-O(21)	2.386(5)	O(28)-Gd(1)-O(29)	54.3(2)
Gd(3)-O(37)	2.406(5)	O(34)-Gd(1)-O(26)	75.64(19)
Gd(3)-O(17)	2.437(5)	O(31)-Gd(1)-O(26)	145.40(19)
Gd(3)-O(41)	2.440(5)	O(32)-Gd(1)-O(26)	75.3(2)
Gd(3)-O(2)	2.450(5)	O(25)-Gd(1)-O(26)	53.22(19)
Gd(3)-O(1)	2.489(5)	O(22)-Gd(1)-O(26)	74.0(2)
Gd(4)-O(30)	2.302(5)	O(28)-Gd(1)-O(26)	125.17(19)
Gd(4)-O(20)	2.343(5)	O(29)-Gd(1)-O(26)	117.9(2)
Gd(4)-O(21)	2.364(5)	O(34)-Gd(1)-O(23)	74.9(2)
Gd(4)-O(40)	2.378(5)	O(31)-Gd(1)-O(23)	74.0(2)
Gd(4)-O(11)	2.413(5)	O(32)-Gd(1)-O(23)	141.14(19)
Gd(4)-O(14)	2.427(6)	O(25)-Gd(1)-O(23)	125.5(2)
Gd(4)-O(39)	2.463(6)	O(22)-Gd(1)-O(23)	52.51(19)
Gd(4)-O(13)	2.508(6)	O(28)-Gd(1)-O(23)	72.7(2)
Ni(1)-O(5)	2.016(5)	O(29)-Gd(1)-O(23)	117.4(2)
Ni(1)-O(2)	2.044(5)	O(26)-Gd(1)-O(23)	119.0(2)
Ni(1)-N(2)	2.060(6)	O(34)-Gd(1)-N(8)	102.0(2)
Ni(1)-N(1)	2.107(6)	O(31)-Gd(1)-N(8)	158.9(2)
Ni(1)-O(19)	2.154(5)	O(32)-Gd(1)-N(8)	83.0(2)
Ni(1)-O(36)	2.234(5)	O(25)-Gd(1)-N(8)	26.9(2)
Ni(2)-N(4)	2.051(7)	O(22)-Gd(1)-N(8)	74.3(2)
Ni(2)-O(11)	2.062(6)	O(28)-Gd(1)-N(8)	101.0(2)
Ni(2)-N(3)	2.093(7)	O(29)-Gd(1)-N(8)	96.3(2)
Ni(2)-O(8)	2.094(5)	O(26)-Gd(1)-N(8)	26.3(2)
Ni(2)-O(20)	2.146(5)	O(23)-Gd(1)-N(8)	126.7(2)
Ni(2)-O(39)	2.191(6)	C(68)-Gd(1)-N(8)	99.5(2)
Ni(3)-O(17)	2.057(6)	O(34)-Gd(1)-N(7)	79.6(2)
Ni(3)-N(5)	2.060(7)	O(31)-Gd(1)-N(7)	100.2(2)
Ni(3)-O(14)	2.061(6)	O(32)-Gd(1)-N(7)	154.2(2)
Ni(3)-N(6)	2.075(7)	O(25)-Gd(1)-N(7)	102.1(2)
Ni(3)-O(21)	2.108(5)	O(22)-Gd(1)-N(7)	26.3(2)

O(28)-Gd(1)-N(7)	73.5(2)	N(2)-Ni(1)-N(1)	97.3(2)
O(29)-Gd(1)-N(7)	127.3(2)	O(5)-Ni(1)-O(19)	85.8(2)
O(26)-Gd(1)-N(7)	96.5(2)	O(2)-Ni(1)-O(19)	84.2(2)
O(23)-Gd(1)-N(7)	26.2(2)	N(2)-Ni(1)-O(19)	96.2(2)
C(68)-Gd(1)-N(7)	100.3(3)	N(1)-Ni(1)-O(19)	166.2(2)
N(8)-Gd(1)-N(7)	100.5(3)	O(5)-Ni(1)-O(36)	78.0(2)
O(5)-Ni(1)-O(2)	166.3(2)	O(2)-Ni(1)-O(36)	90.6(2)
O(5)-Ni(1)-N(2)	93.2(2)	N(2)-Ni(1)-O(36)	169.5(2)
O(2)-Ni(1)-N(2)	97.2(2)	N(1)-Ni(1)-O(36)	89.3(2)
O(5)-Ni(1)-N(1)	95.7(2)	O(19)-Ni(1)-O(36)	77.62(19)
O(2)-Ni(1)-N(1)	91.9(2)		

Table S3. Selected Bond Lengths (Å) and Angles (°) for **2**.

Tb(1)-O(32)	2.405(7)	Ni(3)-O(41)	2.213(7)
Tb(1)-O(34)	2.414(7)	O(32)-Tb(1)-O(34)	77.0(3)
Tb(1)-O(31)	2.421(8)	O(32)-Tb(1)-O(31)	77.2(3)
Tb(1)-O(28)	2.533(7)	O(34)-Tb(1)-O(31)	79.8(3)
Tb(1)-O(22)	2.537(8)	O(32)-Tb(1)-O(28)	130.6(3)
Tb(1)-O(29)	2.541(8)	O(34)-Tb(1)-O(28)	149.0(3)
Tb(1)-O(25)	2.579(8)	O(31)-Tb(1)-O(28)	92.0(3)
Tb(1)-O(26)	2.582(8)	O(32)-Tb(1)-O(22)	148.4(3)
Tb(1)-O(23)	2.598(8)	O(34)-Tb(1)-O(22)	84.3(3)
Tb(1)-N(7)	2.940(9)	O(31)-Tb(1)-O(22)	124.5(3)
Tb(1)-N(8)	2.957(8)	O(28)-Tb(1)-O(22)	75.8(3)
Tb(2)-O(20)	2.384(6)	O(32)-Tb(1)-O(29)	78.6(3)
Tb(2)-O(33)	2.405(7)	O(34)-Tb(1)-O(29)	149.8(3)
Tb(2)-O(5)	2.448(6)	O(31)-Tb(1)-O(29)	77.6(3)
Tb(2)-O(8)	2.457(7)	O(28)-Tb(1)-O(29)	52.0(3)
Tb(2)-O(38)	2.471(7)	O(22)-Tb(1)-O(29)	125.2(3)
Tb(2)-O(19)	2.474(6)	O(32)-Tb(1)-O(25)	94.6(3)
Tb(2)-O(36)	2.548(7)	O(34)-Tb(1)-O(25)	126.7(3)
Tb(2)-O(7)	2.558(7)	O(31)-Tb(1)-O(25)	150.3(3)
Tb(3)-O(19)	2.388(6)	O(28)-Tb(1)-O(25)	71.5(3)
Tb(3)-O(35)	2.402(7)	O(22)-Tb(1)-O(25)	76.4(3)
Tb(3)-O(17)	2.444(7)	O(29)-Tb(1)-O(25)	72.8(2)
Tb(3)-O(21)	2.475(7)	O(32)-Tb(1)-O(26)	77.5(3)
Tb(3)-O(37)	2.480(7)	O(34)-Tb(1)-O(26)	76.9(3)
Tb(3)-O(41)	2.493(7)	O(31)-Tb(1)-O(26)	148.8(3)
Tb(3)-O(2)	2.495(6)	O(28)-Tb(1)-O(26)	118.5(3)
Tb(3)-O(1)	2.578(7)	O(22)-Tb(1)-O(26)	73.6(3)
Tb(4)-O(30)	2.391(7)	O(29)-Tb(1)-O(26)	114.7(3)
Tb(4)-O(21)	2.403(7)	O(25)-Tb(1)-O(26)	50.1(3)
Tb(4)-O(20)	2.450(7)	O(32)-Tb(1)-O(23)	142.4(3)
Tb(4)-O(40)	2.470(7)	O(34)-Tb(1)-O(23)	73.7(3)
Tb(4)-O(11)	2.476(7)	O(31)-Tb(1)-O(23)	74.8(3)
Tb(4)-O(14)	2.532(7)	O(28)-Tb(1)-O(23)	75.3(3)
Tb(4)-O(39)	2.566(7)	O(22)-Tb(1)-O(23)	49.6(3)
Tb(4)-O(13)	2.589(8)	O(29)-Tb(1)-O(23)	118.5(3)
Ni(1)-O(5)	2.003(6)	O(25)-Tb(1)-O(23)	121.8(3)
Ni(1)-N(2)	2.034(8)	O(26)-Tb(1)-O(23)	117.3(3)
Ni(1)-N(1)	2.039(9)	O(32)-Tb(1)-N(7)	153.0(3)
Ni(1)-O(2)	2.048(6)	O(34)-Tb(1)-N(7)	76.0(3)
Ni(1)-O(19)	2.134(7)	O(31)-Tb(1)-N(7)	99.7(3)
Ni(1)-O(36)	2.216(7)	O(28)-Tb(1)-N(7)	76.0(3)
Ni(2)-O(8)	2.032(7)	O(22)-Tb(1)-N(7)	24.9(3)
Ni(2)-N(4)	2.043(9)	O(29)-Tb(1)-N(7)	127.4(3)
Ni(2)-O(11)	2.056(7)	O(25)-Tb(1)-N(7)	100.0(3)
Ni(2)-N(3)	2.064(8)	O(26)-Tb(1)-N(7)	94.6(3)
Ni(2)-O(20)	2.136(7)	O(23)-Tb(1)-N(7)	24.9(3)
Ni(2)-O(39)	2.186(7)	C(68)-Tb(1)-N(7)	101.9(3)
Ni(3)-O(17)	2.028(7)	O(32)-Tb(1)-N(8)	85.7(3)
Ni(3)-O(14)	2.025(8)	O(34)-Tb(1)-N(8)	101.9(3)
Ni(3)-N(5)	2.050(10)	O(31)-Tb(1)-N(8)	162.0(3)
Ni(3)-N(6)	2.057(10)	O(28)-Tb(1)-N(8)	95.0(3)
Ni(3)-O(21)	2.109(7)	O(22)-Tb(1)-N(8)	73.4(3)

O(29)-Tb(1)-N(8)	93.8(3)	N(1)-Ni(1)-O(2)	90.7(3)
O(25)-Tb(1)-N(8)	25.0(3)	O(5)-Ni(1)-O(19)	85.2(3)
O(26)-Tb(1)-N(8)	25.2(3)	N(2)-Ni(1)-O(19)	94.4(3)
O(23)-Tb(1)-N(8)	123.0(3)	N(1)-Ni(1)-O(19)	167.9(3)
N(7)-Tb(1)-N(8)	98.1(3)	O(2)-Ni(1)-O(19)	86.3(3)
O(5)-Ni(1)-N(2)	90.8(3)	O(5)-Ni(1)-O(36)	79.8(2)
O(5)-Ni(1)-N(1)	95.8(3)	N(2)-Ni(1)-O(36)	169.0(3)
N(2)-Ni(1)-N(1)	97.6(3)	N(1)-Ni(1)-O(36)	89.2(3)
O(5)-Ni(1)-O(2)	167.7(3)	O(2)-Ni(1)-O(36)	89.8(3)
N(2)-Ni(1)-O(2)	98.7(3)	O(19)-Ni(1)-O(36)	79.1(3)

Table S4. Selected Bond Lengths (Å) and Angles (°) for **3**.

Gd(1)-O(30)	2.345(6)	Ni(4)-O(21)	2.108(5)
Gd(1)-O(21)	2.356(5)	Ni(4)-O(26)	2.180(5)
Gd(1)-O(19)	2.417(5)	O(30)-Gd(1)-O(21)	73.82(19)
Gd(1)-O(8)	2.420(5)	O(30)-Gd(1)-O(19)	85.43(18)
Gd(1)-O(27)	2.421(5)	O(21)-Gd(1)-O(19)	80.90(17)
Gd(1)-O(5)	2.447(5)	O(30)-Gd(1)-O(8)	74.04(19)
Gd(1)-O(25)	2.473(5)	O(21)-Gd(1)-O(8)	138.05(18)
Gd(1)-O(6)	2.517(5)	O(19)-Gd(1)-O(8)	70.34(16)
Gd(2)-O(33)	2.361(7)	O(30)-Gd(1)-O(27)	147.8(2)
Gd(2)-O(19)	2.380(4)	O(21)-Gd(1)-O(27)	77.19(17)
Gd(2)-O(24)	2.418(6)	O(19)-Gd(1)-O(27)	103.61(16)
Gd(2)-O(20)	2.420(5)	O(8)-Gd(1)-O(27)	138.14(17)
Gd(2)-O(14)	2.437(5)	O(30)-Gd(1)-O(5)	88.32(19)
Gd(2)-O(11)	2.446(6)	O(21)-Gd(1)-O(5)	72.12(17)
Gd(2)-O(23)	2.489(5)	O(19)-Gd(1)-O(5)	152.98(17)
Gd(2)-O(12)	2.523(5)	O(8)-Gd(1)-O(5)	132.62(15)
Gd(3)-O(28)	2.357(7)	O(27)-Gd(1)-O(5)	69.52(17)
Gd(3)-O(20)	2.389(6)	O(30)-Gd(1)-O(25)	138.4(2)
Gd(3)-O(21)	2.408(5)	O(21)-Gd(1)-O(25)	128.57(15)
Gd(3)-O(22)	2.427(5)	O(19)-Gd(1)-O(25)	67.93(16)
Gd(3)-O(2)	2.437(5)	O(8)-Gd(1)-O(25)	67.32(16)
Gd(3)-O(17)	2.459(4)	O(27)-Gd(1)-O(25)	72.09(18)
Gd(3)-O(26)	2.500(5)	O(5)-Gd(1)-O(25)	129.38(17)
Gd(3)-O(18)	2.541(6)	O(30)-Gd(1)-O(6)	116.46(18)
Ni(1)-O(29)#1	2.278(11)	O(21)-Gd(1)-O(6)	132.06(19)
Ni(1)-O(29)	2.278(11)	O(19)-Gd(1)-O(6)	143.00(17)
Ni(1)-O(32)#1	2.288(7)	O(8)-Gd(1)-O(6)	86.70(18)
Ni(1)-O(32)	2.288(7)	O(27)-Gd(1)-O(6)	74.26(16)
Ni(1)-O(31)	2.317(7)	O(5)-Gd(1)-O(6)	62.15(17)
Ni(1)-O(31)#1	2.317(7)	O(25)-Gd(1)-O(6)	76.66(17)
Ni(2)-O(8)	2.021(4)	O(29)#1-Ni(1)-O(29)	179.997(1)
Ni(2)-O(11)	2.031(4)	O(29)#1-Ni(1)-O(32)#1	83.2(3)
Ni(2)-N(4)	2.050(7)	O(29)-Ni(1)-O(32)#1	96.8(3)
Ni(2)-N(3)	2.051(7)	O(29)#1-Ni(1)-O(32)	96.8(3)
Ni(2)-O(19)	2.118(5)	O(29)-Ni(1)-O(32)	83.2(3)
Ni(2)-O(25)	2.190(6)	O(32)#1-Ni(1)-O(32)	179.999(1)
Ni(3)-O(14)	2.022(6)	O(29)#1-Ni(1)-O(31)	101.4(4)
Ni(3)-N(5)	2.050(7)	O(29)-Ni(1)-O(31)	78.6(4)
Ni(3)-N(6)	2.056(6)	O(32)#1-Ni(1)-O(31)	98.1(3)
Ni(3)-O(17)	2.067(6)	O(32)-Ni(1)-O(31)	81.9(3)
Ni(3)-O(20)	2.107(4)	O(29)#1-Ni(1)-O(31)#	78.6(4)
Ni(3)-O(23)	2.177(6)	O(29)-Ni(1)-O(31)#1	101.4(4)
Ni(4)-O(2)	2.014(5)	O(32)#1-Ni(1)-O(31)#1	81.9(3)
Ni(4)-O(5)	2.039(5)	O(32)-Ni(1)-O(31)#1	98.1(3)
Ni(4)-N(1)	2.046(6)	O(31)-Ni(1)-O(31)#1	179.999(1)
Ni(4)-N(2)	2.067(7)		

Table S5. Selected Bond Lengths (Å) and Angles (°) for **4**.

Dy(1)-O(21)	2.380(8)	O(21)-Dy(1)-O(8)	137.4(3)
Dy(1)-O(30)	2.384(10)	O(30)-Dy(1)-O(8)	74.3(3)
Dy(1)-O(8)	2.437(8)	O(21)-Dy(1)-O(27)	77.2(3)
Dy(1)-O(27)	2.450(8)	O(30)-Dy(1)-O(27)	147.5(3)
Dy(1)-O(19)	2.462(8)	O(8)-Dy(1)-O(27)	138.2(3)
Dy(1)-O(5)	2.473(9)	O(21)-Dy(1)-O(19)	80.9(3)
Dy(1)-O(25)	2.505(8)	O(30)-Dy(1)-O(19)	85.9(3)
Dy(1)-O(6)	2.553(9)	O(8)-Dy(1)-O(19)	69.4(3)
Dy(2)-O(33)	2.380(12)	O(27)-Dy(1)-O(19)	103.6(3)
Dy(2)-O(19)	2.403(8)	O(21)-Dy(1)-O(5)	71.8(3)
Dy(2)-O(24)	2.441(9)	O(30)-Dy(1)-O(5)	88.0(3)
Dy(2)-O(14)	2.458(9)	O(8)-Dy(1)-O(5)	133.9(3)
Dy(2)-O(20)	2.459(8)	O(27)-Dy(1)-O(5)	69.2(3)
Dy(2)-O(11)	2.469(9)	O(19)-Dy(1)-O(5)	152.7(3)
Dy(2)-O(23)	2.511(9)	O(21)-Dy(1)-O(25)	128.1(3)
Dy(2)-O(12)	2.550(10)	O(30)-Dy(1)-O(25)	138.5(3)
Dy(3)-O(28)	2.395(14)	O(8)-Dy(1)-O(25)	67.0(3)
Dy(3)-O(20)	2.408(9)	O(27)-Dy(1)-O(25)	72.4(3)
Dy(3)-O(21)	2.434(8)	O(19)-Dy(1)-O(25)	67.1(3)
Dy(3)-O(22)	2.457(9)	O(5)-Dy(1)-O(25)	130.0(3)
Dy(3)-O(2)	2.458(9)	O(21)-Dy(1)-O(6)	131.8(3)
Dy(3)-O(17)	2.509(9)	O(30)-Dy(1)-O(6)	115.9(4)
Dy(3)-O(26)	2.558(9)	O(8)-Dy(1)-O(6)	87.7(3)
Dy(3)-O(18)	2.582(10)	O(27)-Dy(1)-O(6)	74.3(3)
Ni(2)-O(8)	2.028(9)	O(19)-Dy(1)-O(6)	143.3(3)
Ni(2)-N(3)	2.043(11)	O(5)-Dy(1)-O(6)	62.1(3)
Ni(2)-O(11)	2.053(9)	O(25)-Dy(1)-O(6)	77.8(3)
Ni(2)-N(4)	2.053(12)	O(8)-Ni(2)-N(3)	90.9(4)
Ni(2)-O(19)	2.111(8)	O(8)-Ni(2)-O(11)	168.3(3)
Ni(2)-O(25)	2.199(8)	N(3)-Ni(2)-O(11)	97.2(4)
Ni(3)-O(14)	2.008(9)	O(8)-Ni(2)-N(4)	96.3(4)
Ni(3)-N(5)	2.046(12)	N(3)-Ni(2)-N(4)	96.2(5)
Ni(3)-O(17)	2.058(9)	O(11)-Ni(2)-N(4)	91.2(4)
Ni(3)-N(6)	2.060(11)	O(8)-Ni(2)-O(19)	84.7(3)
Ni(3)-O(20)	2.107(9)	N(3)-Ni(2)-O(19)	97.3(4)
Ni(3)-O(23)	2.173(9)	O(11)-Ni(2)-O(19)	85.9(3)
Ni(4)-O(2)	2.017(10)	N(4)-Ni(2)-O(19)	166.5(4)
Ni(4)-N(1)	2.030(12)	O(8)-Ni(2)-O(25)	80.3(3)
Ni(4)-O(5)	2.046(9)	N(3)-Ni(2)-O(25)	170.7(4)
Ni(4)-N(2)	2.067(11)	O(11)-Ni(2)-O(25)	91.1(3)
Ni(4)-O(21)	2.121(8)	N(4)-Ni(2)-O(25)	87.8(4)
Ni(4)-O(26)	2.192(9)	O(19)-Ni(2)-O(25)	79.1(3)
O(21)-Dy(1)-O(30)	73.7(3)		