Electronic Supplementary Information

Employment of methyl 2-pyridyl ketone oxime in 3d/4f-metal chemistry: dinuclear nickel(II)/lanthanide(III) species and complexes containing the metals in separate ions[+]

Christina D. Polyzou,[a,b] Helen Nikolaou,[a] Constantina Papatriantafyllopoulou,[c] Vassilis Psycharis,[d] Aris Terzis,[d] Catherine Raptopoulou,[d] Albert Escuer*[,b] and Spyros P. Perlepes*[,a,e]

[a] *Department of Chemistry, University of Patras, 265 04 Patras, Greece. Tel: +30 2610 996730; E-mail: perlepes@patreas.upatras.gr*

[b] *Departament de Quimica Inorgànica and Institut de Nanociéncia i Nanotecnologia, Universitat de Barcelona(IN^{2}UB), Av.Diagonal 645, 08028 Barcelona, Spain. Tel: +34 93 4039138; E-mail:albert.escuer@qi.ub.es*

[c] *Department of Chemistry, University of Cyprus, 1678 Nicosia, Cyprus*

[d] *Institute of Advanced Materials, Physicochemical Processes, Nanotechnology and Microsystems, Department of Materials Science, NCSR "Demokritos", 153 10 Aghia Paraskevi Attikis, Greece*

[e] *Institute of Chemical Engineering Sciences, Foundation for Research and Technology – Hellas (FORTH/ICE-HT), Platani, P.O. Box 1414, 265 04 Patras, Greece*

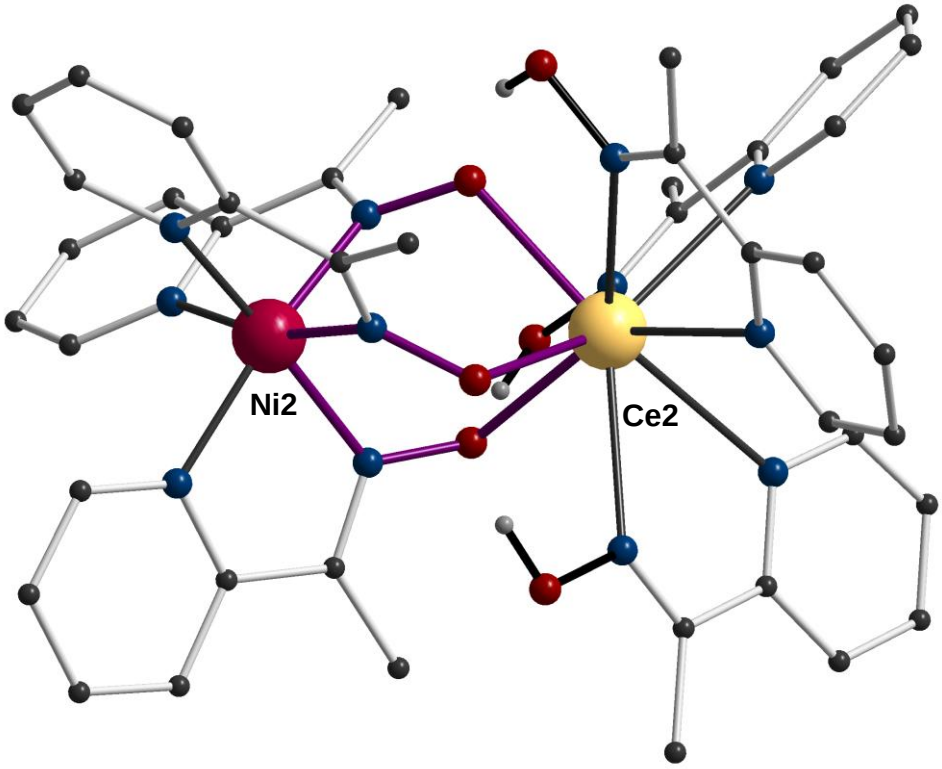

Fig. S1 Plot of the second crystallographically independent, heterometallic cation that is present in **1**·1.2MeOH·0.6H_2O; the first one is shown in Fig. 1. Only the two metal ions are labelled. Colour scheme: Ni, dark red; Ce, yellow; O, red; N, blue; C, black; H, gray.

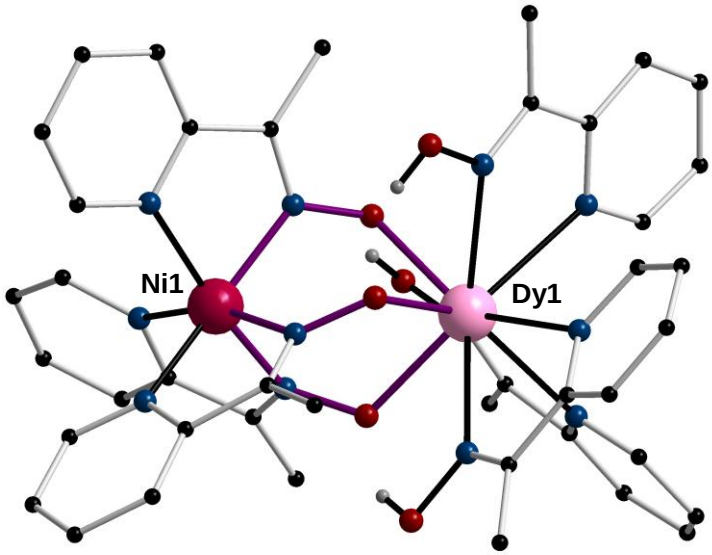

Fig. S2 Plot of the second crystallographically independent, heterometallic cation that is present in **8**·1.2MeOH·0.6H_2O; the first one is shown in Fig. 3. Only the two metal ions are labelled. Colour scheme: Ni, dark red; Dy, pink; O, red; N, blue; C, black; H, gray.

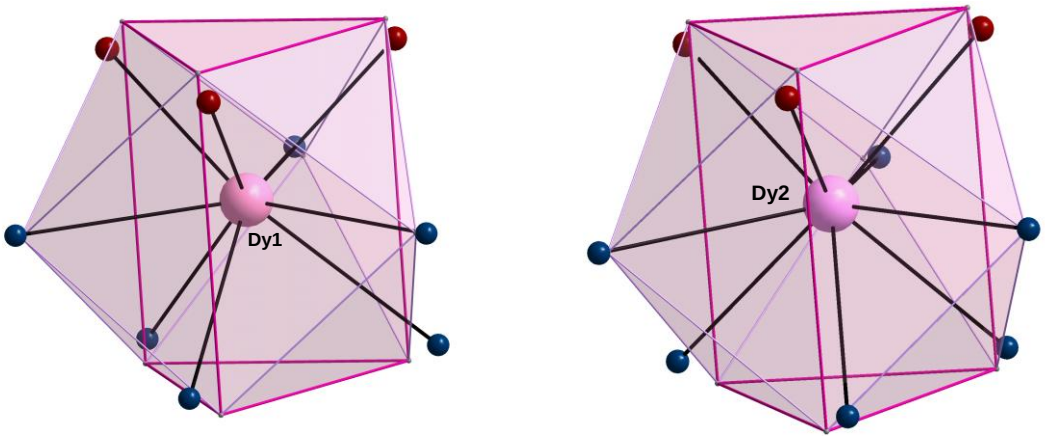

Fig. S3 The tricapped trigonal prismatic coordination spheres of Dy1(left) and Dy2 (right) in the structure of **8**·1.2MeOH·0.6H_2O. The very small spheres define the vertices of the ideal polyhedron. Colour scheme: Dy, pink; O, red; N, blue.

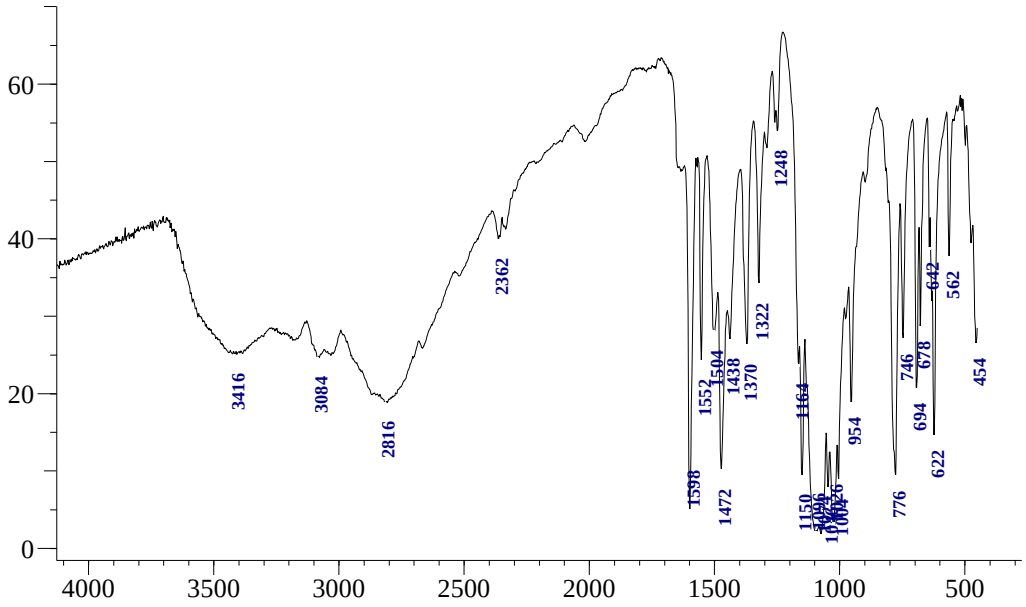

Fig. S4 The IR spectrum (KBr, cm^{-1}) of complex $[NiCe(mpko)_3(mpkoH)_3](ClO_4)_2$ (**1**).

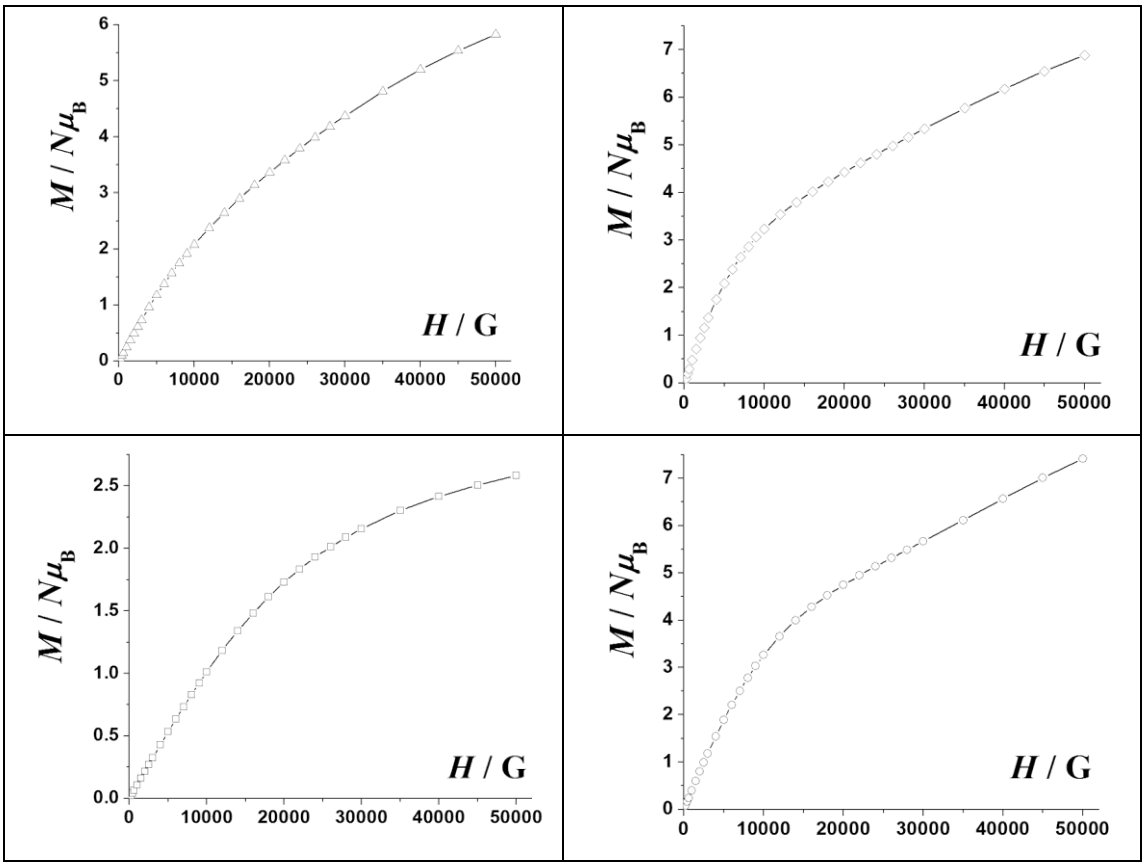

Fig. S5 Reduced magnetization *vs*. magnetic field plots for complexes **2** (squares), **6** (circles), **7** (triangles) and **8** (diamonds) at 2 K. Solid lines are guides to the eye.

Table S1 Shape measures of the 9-coordinate Ce1[a], Ce2[a], Dy1[b], and Dy[b] coordination polyhedra in complexes **1**·1.2MeOH·0.6H_2O and **8**·1.2MeOH·0.6H_2O. The values in boldface indicate the closest polyhedron according to the continuous shape measures

Polyhedron [c]	Ce1	Ce2	Dy1	Dy2
EP-9	31.25	32.13	31.95	32.62
OPY-9	24.21	24.29	23.78	24.13
HBPY-9	17.52	17.51	18.13	18.39
JTC-9	12.64	13.09	12.82	13.36
JCCU-9	9.57	9.55	9.83	9.99
CCU-9	8.43	8.39	8.75	8.88
JCSAPR-9	2.86	2.63	2.65	2.36
CSAPR-9	2.12	**1.75**	1.78	1.48
JTCTPR-9	2.07	2.11	1.72	1.76
TCTPR-9	**2.02**	1.79	**1.65**	**1.46**
JTDIC-9	11.28	11.22	12.03	11.93
HH-9	11.61	11.90	11.68	11.92
MFF-9	2.61	2.27	2.31	2.10

[a] See Fig. 2. [b] See Fig. S3. [c] Abbreviations: EP-9, enneagon; OPY-9, octagonal pyramid; HBPY-9, heptagonal bipyramid; JTC-9, Johnson triangular cupola; JCCU-9, capped cube-Johnson; CCU-9, spherical-relaxed capped cube; JCSAPR-9, capped square antiprism-Johnson; CSAPR-9, spherical capped square antiprism; JTCTPR-9, tricapped trigonal prism; TCTPR-9, spherical tricapped trigonal prism; JTDIC-9, tridiminished icosahedron; HH-9, hula-hoop; MFF-9, muffin.

Table S2 Shape measures of the 12-coordinate La coordination polyhedron[a] in complex **11**·2MeOH. The value in boldface indicates the closest polyhedron according to the continuous shape measures

Polyhedron[b]	La
DP-12	34.89
HPY-12	30.72
DBPY-12	19.35
HPR-12	9.90
HARP-12	16.30
TT-12	10.97
COC-12	**2.04**
ACOC-12	6.38
IC-12	3.60
JSC-12	20.84
JEPBPY-12	9.89
JBAPPR-12	15.37
JSPMC-12	19.08

[a] See Fig. 5. [b] Abbreviations: DP-12, dodecagon; HPY-12, Hendecagonal pyramid; DBPY-12, decagonal bipyramid; HPR-12, hexagonal prism; HAPR-12, hexagonal antiprism; TT-12, truncated tetrahedron; COC-12, cuboctahedron; ACOC-12, anticuboctahedron (triangular orthobicupola); IC-12, icosahedron; JSC-12, square cupola; JEPBY-12, elongated pentagonal bipyramid; JBAPPR-12, biaugmented pentagonal prism; JSPMC-12, sphenomegacorona.