ELECTRONIC SUPPLEMENTARY INFORMATION

A [Ce21] Keplerate †

Angelos B. Canaj,^a Milosz Siczek,^b Tadeusz Lis,^b Mark Murrie,^{c,*} Euan K. Brechin^{d,*} and Constantinos J. Milios^{a, *}

^a Department Of Chemistry, University of Crete, Voutes 71003, Herakleion, Greece. Fax: +30-2810-545001; Tel: +30-2810-545099; E-mail: <u>komil@uoc.gr</u>

^b Faculty of Chemistry, University of Wroclaw, Joliot-Curie 14, Wroclaw 50-383, Poland.

^c WestCHEM, Department of Chemistry, University of Glasgow, University Avenue, Glasgow, UK G12 8QQ; <u>Mark.Murrie@glasgow.ac.uk</u>

^d EaStCHEM School of Chemistry, The University of Edinburgh, David Brewster Road, EH9 3FJ, Edinburgh, UK; <u>E.Brechin@ed.ac.uk</u>

Materials and physical measurements

All manipulations were performed under aerobic conditions, using materials as received. Elemental analyses (C, H, N) were performed by the University of Ioannina microanalysis service. Variable-temperature, solid-state direct current (dc) magnetic susceptibility data down to 2.0 K were collected on a Quantum Design MPMS-XL SQUID magnetometer equipped with a 5 T DC magnet at the University of Glasgow. Diamagnetic corrections were applied to the observed paramagnetic susceptibilities using Pascal's constants.

Synthesis

To a methanolic solution (10 ml) of 2-amino-isobutyric acid (1 mmol, 104 mg) were added Ce(NO₃)₃·6H₂O (1mmol, 434 mg) and NEt₃ (1.81 mmol), the resulting colourless solution left to stir. After 24 hours, 2-hydroxy-l-naphthaldehyde (1 mmol, 172 mg) and 2-amino-2-methyl-1,3-propanediol (1 mmol, 105 mg) were added and the brown reaction mixture transferred to a Teflon-lined autoclave and heated at 95 °C for 24 hours. Slow cooling to room temperature afforded dark brown crystals in ~15 % yield. Elemental Anal. calcd (found) for 1·2H₂O: C 29.84 (30.1), H 2.85 (2.61), N 4.00 (3.83) %. IR (KBr pellet): 1595 (vs), 1543 (s), 1512 (s), 1466 (vs), 1402 (vs), 1350 (vs), 1297 (vs), 1203 (vs), 1145 (vs), 1092 (w), 1032 (w), 831 (m), 735 (m), 615 (w), 471 (vs).

	1 ·9MeOH·4.2H ₂ O
Formula ^a	$C_{192}H_{248.60}Ce_{21}N_{21}O_{118.80}$
$M_{ m W}$	7694.02
Crystal System	Trigonal
Space group	<i>R</i> -3
a/Å	20.830 (5)
α/ο	106.55 (3)
V/Å ³	7617 (6)
Ζ	1
T/K	100
λ ^b /Å	0.71073
$D_{\rm c}/{\rm g~cm^{-3}}$	1.677
μ (Mo-K _{α})/ mm ⁻¹	3.15
Meas./indep.(<i>R</i> _{int}) refl.	35039/14429 (0.054)
Obs. refl. $[I > 2\sigma(I)]$	8254
wR2 ^{c,d}	0.198
$R1^{d,e}$	0.068
Goodness of fit on F^2	1.11
$\Delta ho_{ m max,min}/ { m e}{ m \AA}^{-3}$	1.75 -1.27

 Table S1. Crystallographic data for complex 1.

^aIncluding solvate molecules, ^bMo-K α radiation(graphite monochromator), ^cwR2= $[\Sigma w(F_o^2 - F_c^2)^2 / \Sigma wF_o^2]^{1/2}$, ^dFor observed data, ^eR1= $\Sigma ||F_o| - |F_c|| / \Sigma |F_o|$.



Figure S1. Plot of the in-phase (χ_M') susceptibility versus temperature (*T*) for **1** measured with a 3.5 G field oscillating at the indicated frequencies.