Eur. J. Inorg. Chem. **2010** · © WILEY-VCH Verlag GmbH & Co. KGaA, 69451 Weinheim, 2011 · ISSN 1434–1948

SUPPORTING INFORMATION

DOI: 10.1002/ejic.201100038

Title: A Rare μ_4-O Centred Dy_4 Tetrahedron with Coordination-Induced Local Chirality and Single-Molecule Magnet Behaviour

Author(s): Po-Heng Lin, Ilia Korobkov, Wolfgang Wernsdorfer, Liviu Ungur, Liviu F. Chibotaru, Muralee Murugesu*

X-Ray Crystallography:

Single crystals of $[Dy_4(\mu_4\text{-}O)(\mu\text{-}OMe)_2(beh)_2(esh)_4]\cdot 3MeOH$, **1**·3MeOH were grown from MeOH. Suitable needle shaped crystal was mounted in inert oil and transferred to the cold gas stream of the diffractometer. Unit cell measurements and intensity data collections were performed on a Bruker-AXS SMART 1 k CCD diffractometer using graphite monochromated Mo Kα radiation (λ = 0.71073 Å). The data reduction included a correction for Lorentz and polarization effects, with an applied multi-scan absorption correction (SADABS).
The crystal structure was solved and refined using the SHELXTL program suite.[1] Direct methods yielded all non-hydrogen atoms, which were refined with anisotropic thermal parameters. All hydrogen atom positions were calculated geometrically and were riding on their respective atoms.

Magnetic properties:

The magnetic susceptibility measurements were obtained using a Quantum Design SQUID magnetometer MPMS-XL7 at the University of Ottawa. This magnetometer operates between 1.8 and 400 K for applied dc fields ranging from –7 to 7 T. The measurements were performed on freshly filtered 5.00 mg of ground polycrystalline sample. The obtained magnetisation data were corrected for the sample holder and the diamagnetic contribution. Before measurements were carried out, the sample was checked for the presence of ferromagnetic impurities by measuring the magnetization as a function of the field at 100 K. For pure paramagnetic or diamagnetic systems, a perfect straight line is expected and is indeed observed for this compound, indicating the absence of any ferromagnetic impurities.

Computational details:

Basis Sets: All employed basis sets were taken from the standard ANO-RCC basis set library from MOLCAS. The following contractions were used for the atoms:

Dy – 8s7p5d4f2g1h

La –. La.ECP.deGraaf.0s.0s.0e-La(LaMnO3).

N, O – 4s3p2d. (only for the first coordinated atoms, which make a bond with Dy)

N,O, C – 3s2p. (for distant atoms)

H – 2s.

Active space of the CASSCF method included 9 electrons in 7 orbitals.

The **spin orbit interaction** was computed by mixing of 21 sextets, 128 quartets and 130 doublet spin free states.

([1]) Sheldrick, G. M.; *Acta Cryst*. **2008**, *A64*, 112-122.

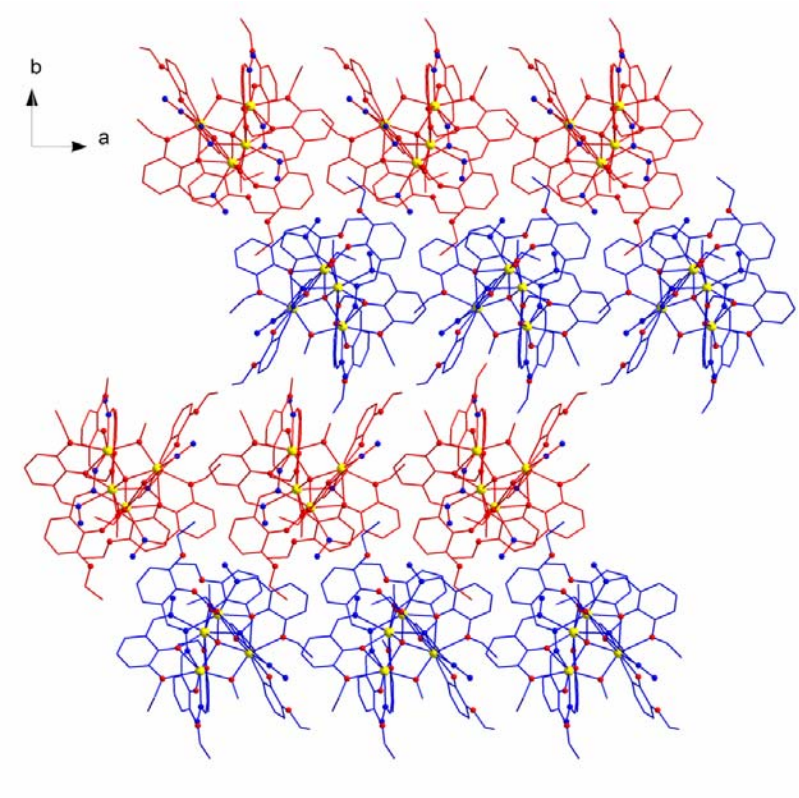

Figure S1. Crystal packing diagram of the along the crystallographic *c*-axis showing the different stereoisomers

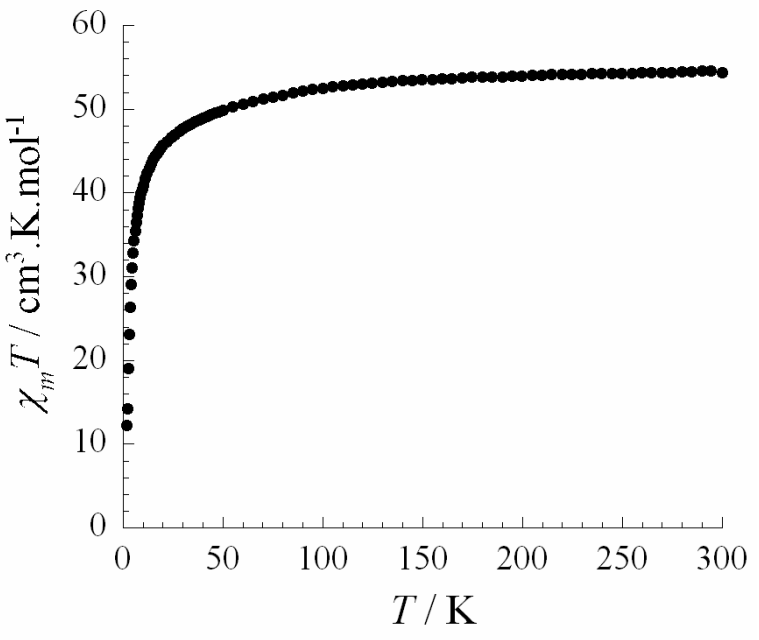

Figure S2. Temperature dependence of the $\chi_M T$ product at 0.1 T for **1** (with $\chi = M/H$ normalized per mol).

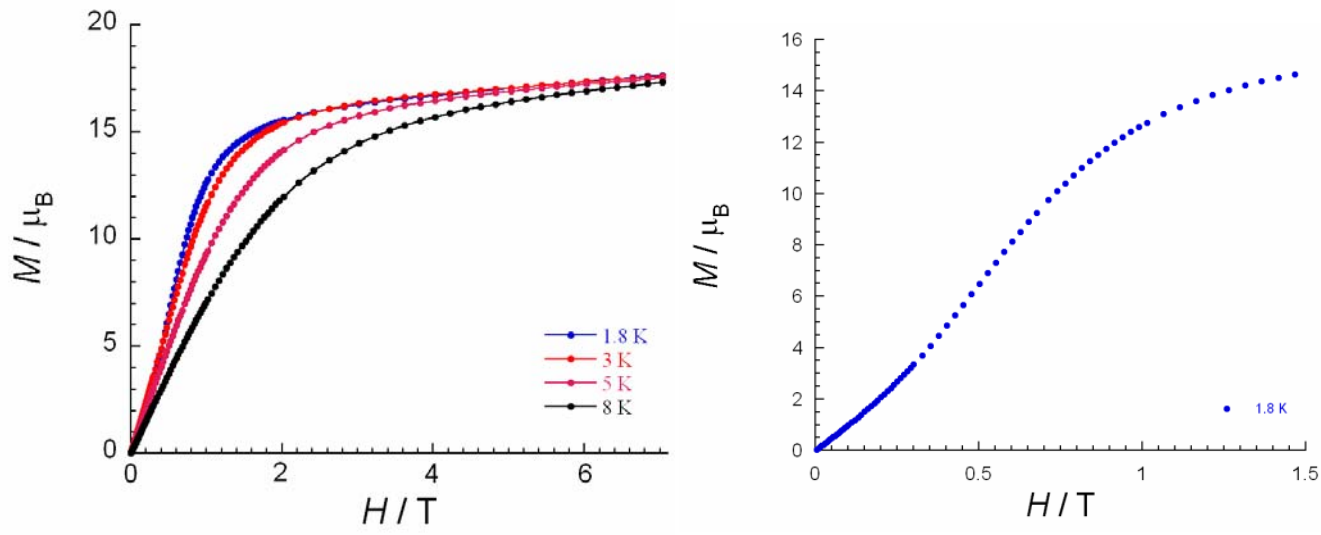

Figure S3. Left: Field dependence of the magnetization of **1** below 8 K. Right: Zoom in section of the magnetization of 1 at 1.8 K.

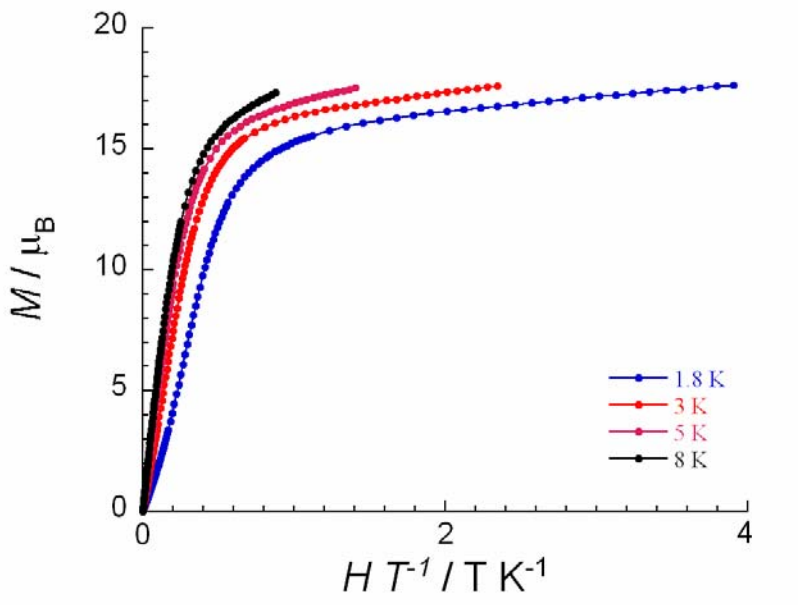

Figure S4. Field dependence of the reduced magnetization of **1** below 8 K.

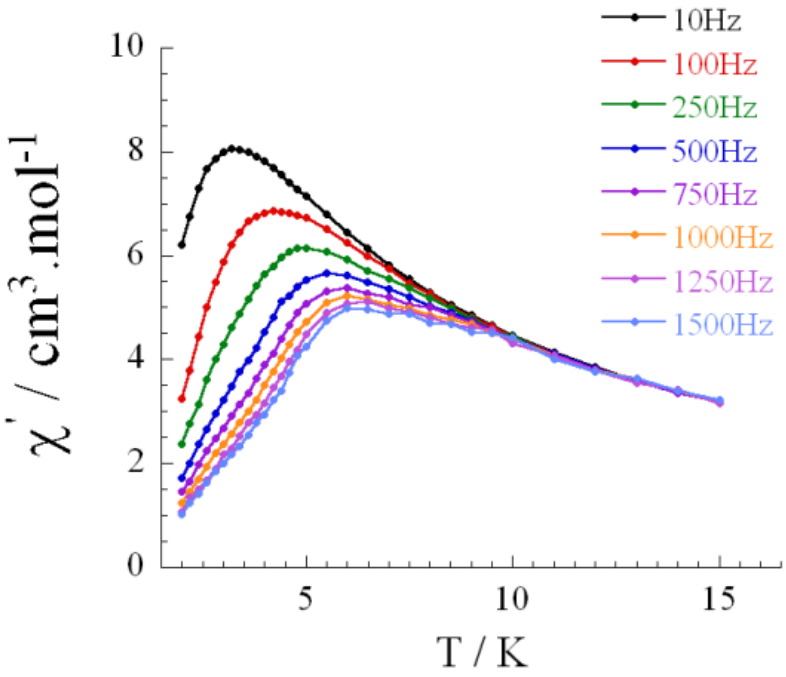

Figure S5. Frequency dependence of the in-phase (χ') ac susceptibility between 10 and 1500 Hz at H_{dc} = 0 T.

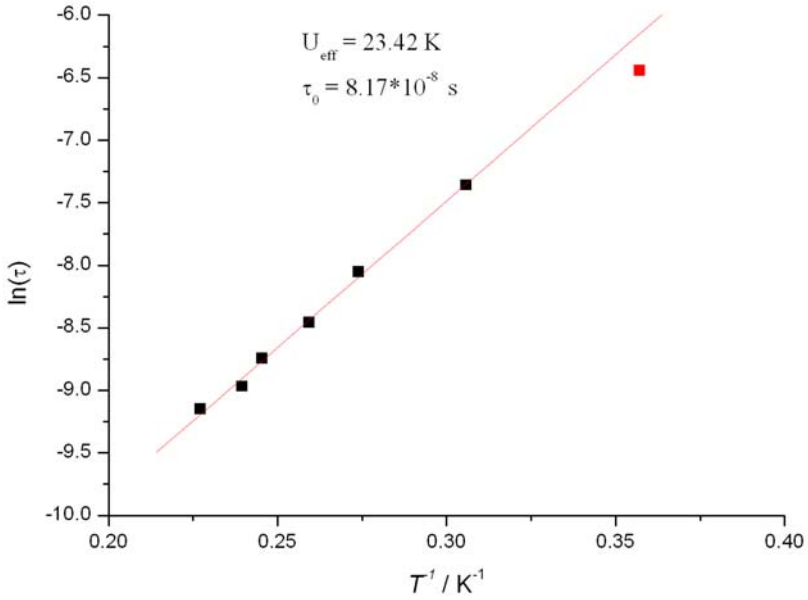

Figure S6. Relaxation time of the magnetization ln(τ) *vs*. T^{-1} (Arrhenius Plot using temperature dependent ac data). The solid line corresponds to the fit.

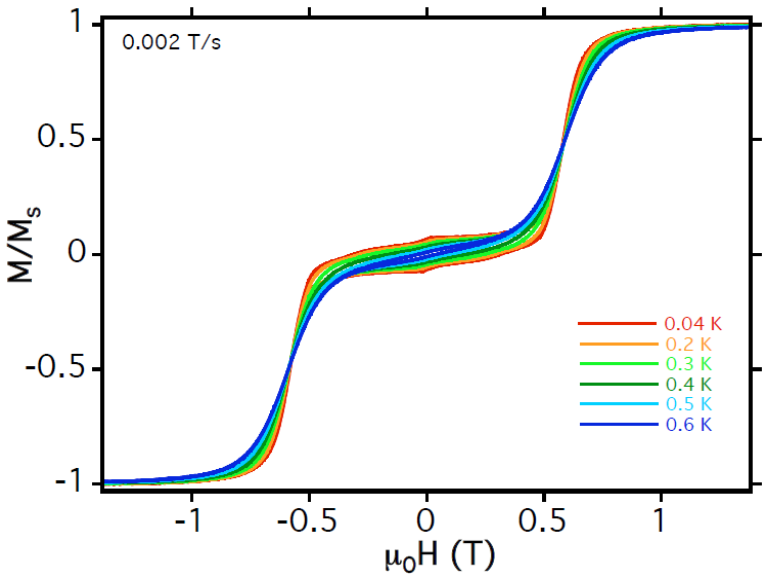

Figure S7. Magnetization (M) vs. applied dc field sweeps at the indicated temperatures and field-sweeping rate of 0.002 T/s.

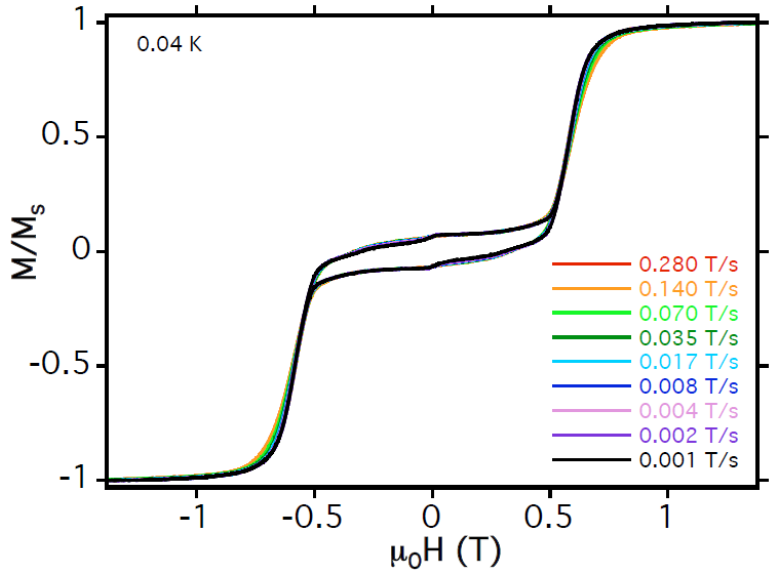

Figure S8. Magnetization (M) *vs.* applied dc field sweeps at the indicated sweep rate and 0.04 K.

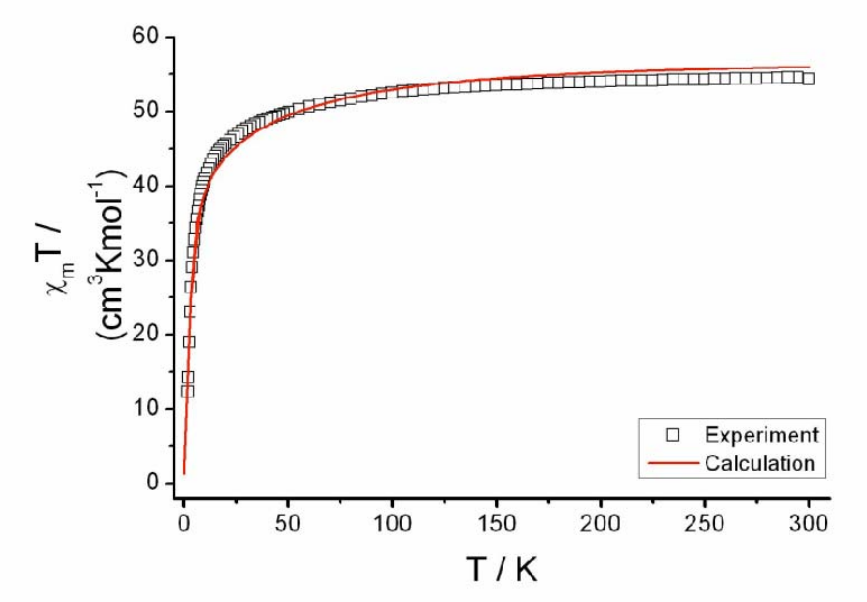

Figure S9. A comparison between measured (empty squares) and calculated (solid line) magnetic susceptibility.

Table S1. Lowest exchange Ising doublets (arising from interaction of the lowest Kramers doublets on Dy sites), the corresponding energy difference, and g tensor.

	Exchange Ising doublets (cm^{-1})	Energy difference (cm^{-1})		Main values of the *g* tensor
1	0.000000000 0.000000606	0.000000606	g_x g_y g_z	0.00000000000 0.00000000148 6.26665932554
2	1.968070024 1.968085032	0.000015008	g_x g_y g_z	0.00000000000 0.00000038726 29.03259779981
3	2.626979385 2.627002588	0.000023203	g_x g_y g_z	0.00000000000 0.00000017890 33.71601804671
4	3.189897427 3.189963472	0.000066045	g_x g_y g_z	0.00000000000 0.00000007279 36.83560801316
5	3.371614505 3.371677563	0.000063058	g_x g_y g_z	0.00000000000 0.00000006427 38.05238967367
6	3.737914219 3.737940634	0.000026415	g_x g_y g_z	0.00000033713 0.00000047687 39.78258976500
7	4.246628308 4.246642665	0.000014357	g_x g_y g_z	0.00000000000 0.00000038327 42.59751775497
8	6.720190691 6.720192285	0.000001594	g_x g_y g_z	0.00000000000 0.00000018799 53.20988514345