

Supplementary Materials

A New {Dy₅} Single-Molecule Magnet Bearing the Schiff Base Ligand *N*-Naphthalidene-2-amino-5-chlorophenol

Table S1. Selected interatomic distances (Å) and angles (°) for complex 1·3MeOH·CH₂Cl₂.

Dy1-O3	2.284(4)	Dy3-O21	2.391(5)
Dy1-O4	2.551(4)	Dy3-O22	2.310(4)
Dy1-O7	2.270(4)	Dy3-O23	2.355(4)
Dy1-O8	2.378(4)	Dy3-N4	2.486(5)
Dy1-O24	2.327(4)	Dy4-O11	2.346(4)
Dy1-O25	2.389(5)	Dy4-O13	2.459(4)
Dy1-O26	2.313(4)	Dy4-O14	2.318(4)
Dy1-N1	2.451(5)	Dy4-O15	2.228(5)
Dy2-O2	2.447(5)	Dy4-O16	2.415(4)
Dy2-O4	2.434(4)	Dy4-O18	2.394(5)
Dy2-O5	2.276(4)	Dy4-O19	2.418(4)
Dy2-O6	2.292(4)	Dy4-N5	2.476(5)
Dy2-O9	2.365(4)	Dy5-O8	2.348(4)
Dy2-O10	2.415(5)	Dy5-O9	2.331(4)
Dy2-O24	2.348(4)	Dy5-O11	2.356(4)
Dy2-O26	2.409(4)	Dy5-O12	2.415(4)
Dy3-O12	2.507(4)	Dy5-O13	2.300(4)
Dy3-O13	2.323(4)	Dy5-O26	2.320(4)
Dy3-O14	2.331(4)	Dy5-N2	2.495(5)
Dy3-O19	2.383(4)	Dy5-N3	2.476(5)
Dy2-O4-Dy1	92.1(1)	Dy5-O13-Dy4	106.7(2)
Dy5-O8-Dy1	107.9(2)	Dy4-O14-Dy3	100.8(2)
Dy5-O9-Dy2	109.9(2)	Dy1-O26-Dy2	98.9(2)
Dy4-O11-Dy5	108.6(2)	Dy1-O26-Dy5	111.2(2)
Dy5-O12-Dy3	103.0(2)	Dy5-O26-Dy2	108.8(2)
Dy3-O13-Dy4	97.0(2)	Dy3-O19-Dy4	96.5(2)
Dy5-O13-Dy3	112.9(2)	Dy1-O24-Dy2	100.3(2)
Dy1...Dy2...Dy5	61.8(2)	Dy3...Dy4...Dy5	55.7(2)
Dy2...Dy5...Dy1	55.8(2)	Dy3...Dy5...Dy4	62.6(2)
Dy5...Dy1...Dy2	62.4(2)	Dy5...Dy3...Dy4	61.7(2)

Table S2. Continuous shape measures (CShM) of the 8-coordinate Dy(1-5) coordination polyhedra in complex **1**.^a

Polyhedron ^b	Dy1	Dy2	Dy3	Dy4	Dy5
OP-8	29.89	31.53	29.46	30.61	34.32
HPY-8	24.36	21.09	21.77	24.03	22.68
HBPY-8	14.85	15.93	14.07	16.17	14.51
CU-8	10.59	13.00	8.06	12.33	7.79
SAPR-8	1.41	4.31	1.53	2.57	1.10
TDD-8	1.40	2.86	1.55	1.03	2.19
JGBF-8	13.15	13.48	14.30	12.30	15.47
JETBPY-8	29.10	26.83	27.13	28.50	29.11
JBTPR-8	1.92	2.22	3.22	2.47	3.54
BTPR-8	1.82	1.52	2.43	2.09	3.00
JSD-8	3.11	4.58	4.34	2.92	5.97
TT-8	10.82	13.70	8.65	13.09	8.48
ETBPY-8	25.19	22.69	22.14	24.69	24.27

^a The values in boldface indicate the closest polyhedron according to the CShM. ^b Abbreviations: OP-8, octagon; HPY-8, heptagonal pyramid; HBPY-8, hexagonal bipyramid; CU-8, cube; SAPR-8, square antiprism; TDD-8, triangular dodecahedron; JGBF-8, Johnson gyrobifastigium; JETBPY-8, Johnson elongated triangular bipyramid; JBTPR-8, Johnson biaugmented trigonal prism; BTPR-8, biaugmented trigonal prism; JSD-8, Johnson snub diphendoid; TT-8, triakis tetrahedron; ETBPY-8, elongated trigonal bipyramid.

Table S3. Crystallographic data for complex **1**·3MeOH·CH₂Cl₂.

Parameter	1 ·3MeOH·CH ₂ Cl ₂
Empirical formula	C ₁₁₅ H ₁₁₈ N ₅ O ₂₆ Dy ₅ Cl ₇
FW / g mol ⁻¹	3046.79
Temperature / K	100(1)
Crystal system	triclinic
Space group	<i>P</i> -1
<i>a</i> / Å	14.4539(3)
<i>b</i> / Å	16.7348(3)
<i>c</i> / Å	27.4249(4)
α / °	79.7590(10)
β / °	79.6500(10)
γ / °	65.134(2)
Volume / Å ³	5881.7(2)
<i>Z</i>	2
ρ_{calc} / g cm ⁻³	1.720
μ / mm ⁻¹	3.369
F(000)	3000
Radiation	Mo K α (λ = 0.71073)
Index ranges	-17 ≤ <i>h</i> ≤ 17
	-19 ≤ <i>k</i> ≤ 19
	-32 ≤ <i>l</i> ≤ 32
Reflections collected	66721
Independent reflections	17139 (<i>R</i> _{int} = 0.0610)
Goodness-of-fit on <i>F</i> ²	1.043
Final <i>R</i> indexes [<i>I</i> ≥ 2σ(<i>I</i>)] ^{a,b}	<i>R</i> ₁ = 0.0462
	w <i>R</i> ₂ = 0.1143
Final <i>R</i> indexes [all data]	<i>R</i> ₁ = 0.0590
	w <i>R</i> ₂ = 0.1223
(Δρ) _{max,min} / e Å ⁻³	3.651 and -1.743

^a $R_1 = \Sigma(|F_o| - |F_c|) / \Sigma|F_o|$. ^b $wR_2 = [\Sigma[w(F_o^2 - F_c^2)^2] / \Sigma[w(F_o^2)^2]]^{1/2}$, $w = 1 / [\sigma^2(F_o^2) + (ap)^2 + bp]$, where $p = [\max(F_o^2, 0) + 2F_c^2] / 3$.

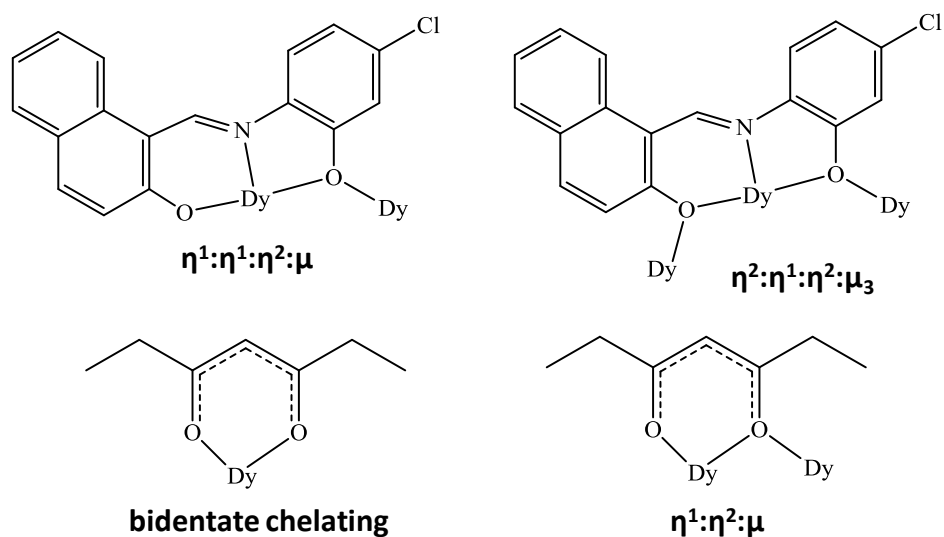


Figure S1. Crystallographically established coordination modes of the ligands nacp^{2-} (top) and hpd^- (bottom) present in **1**.

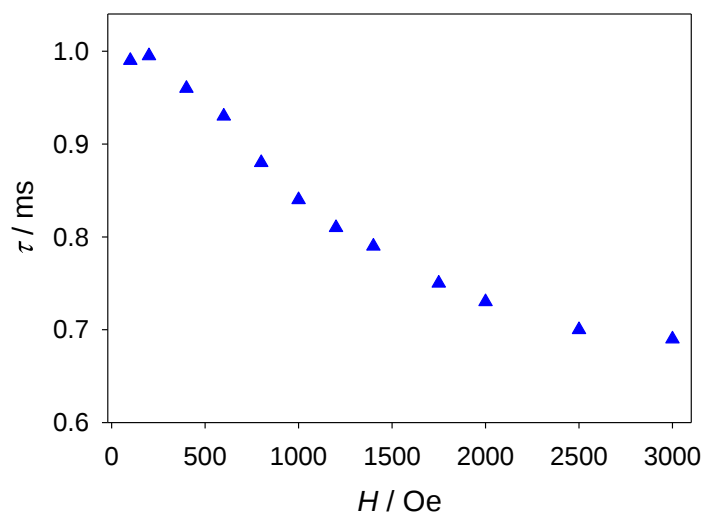


Figure S2. Field (H) dependence of the relaxation time (τ) of $1:3\text{MeOH}:\text{CH}_2\text{Cl}_2$ measured at 2.0 K. The peak maximum corresponds to the optimum dc field.

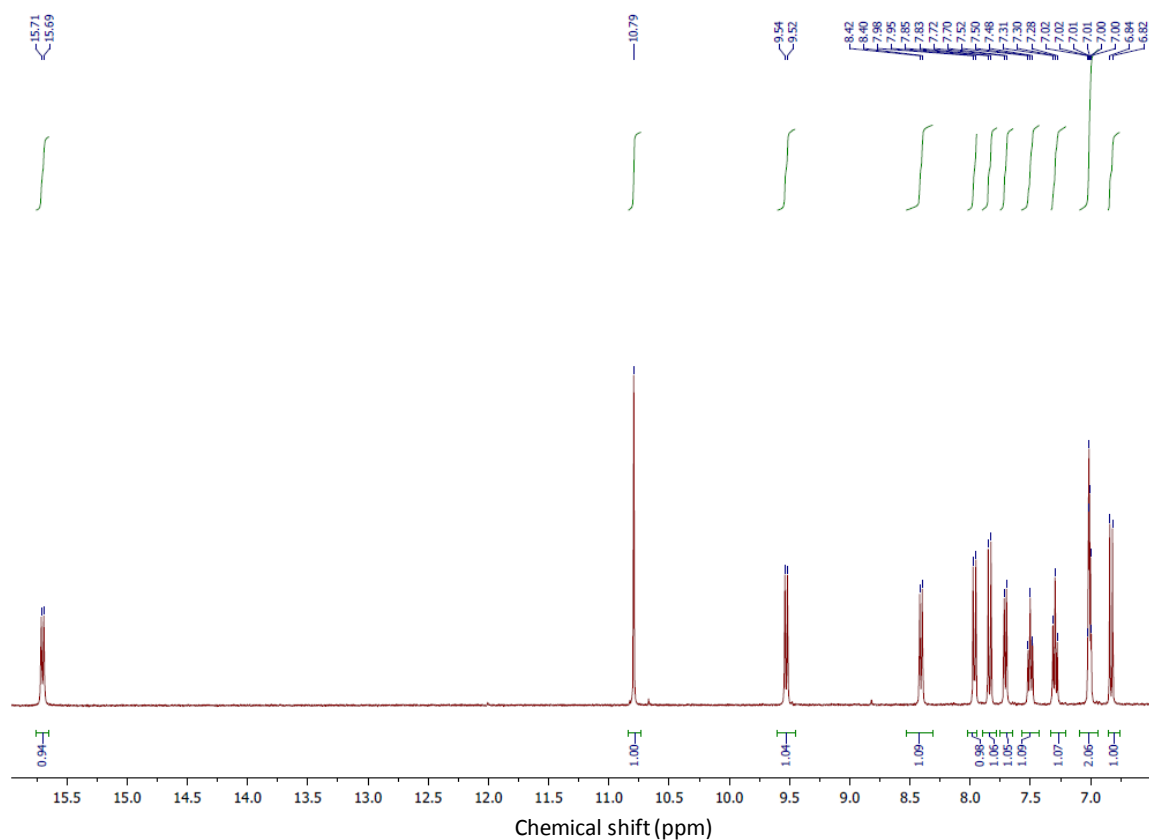


Figure S3. ¹H NMR spectrum of the ligand nacpH₂ in DMSO-*d*₆.

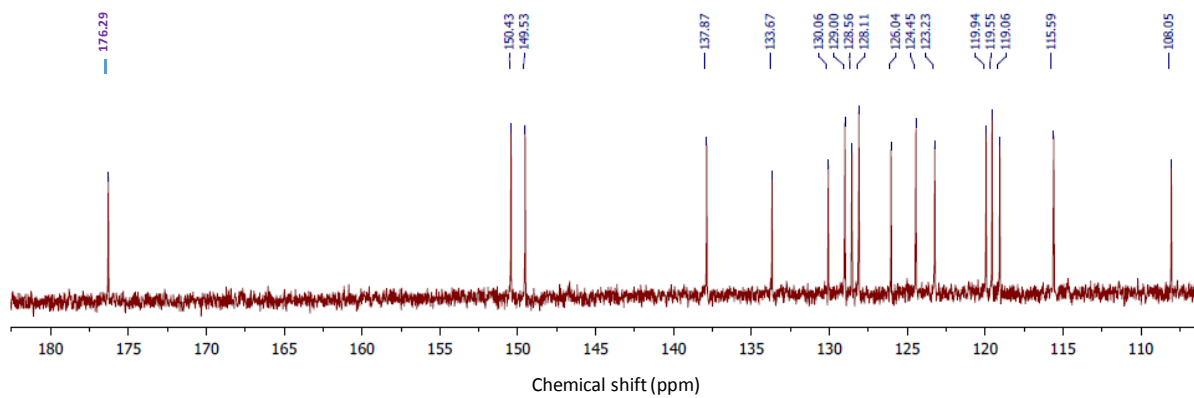


Figure S4. ¹³C NMR spectrum of the ligand nacpH₂ in DMSO-*d*₆.

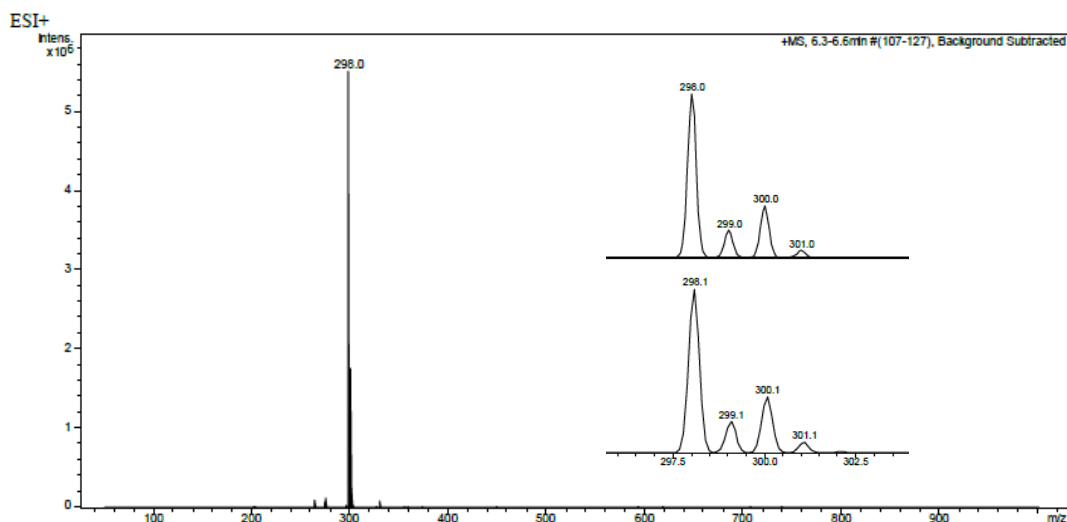


Figure S5. Positive ESI-MS spectra of the ligand nacpH₂ in MeCN. The inset spectra show a zoomed picture of the experimental data (**top**) and the theoretical simulation of the isotope model of the protonated ion of nacpH₂ (**bottom**).

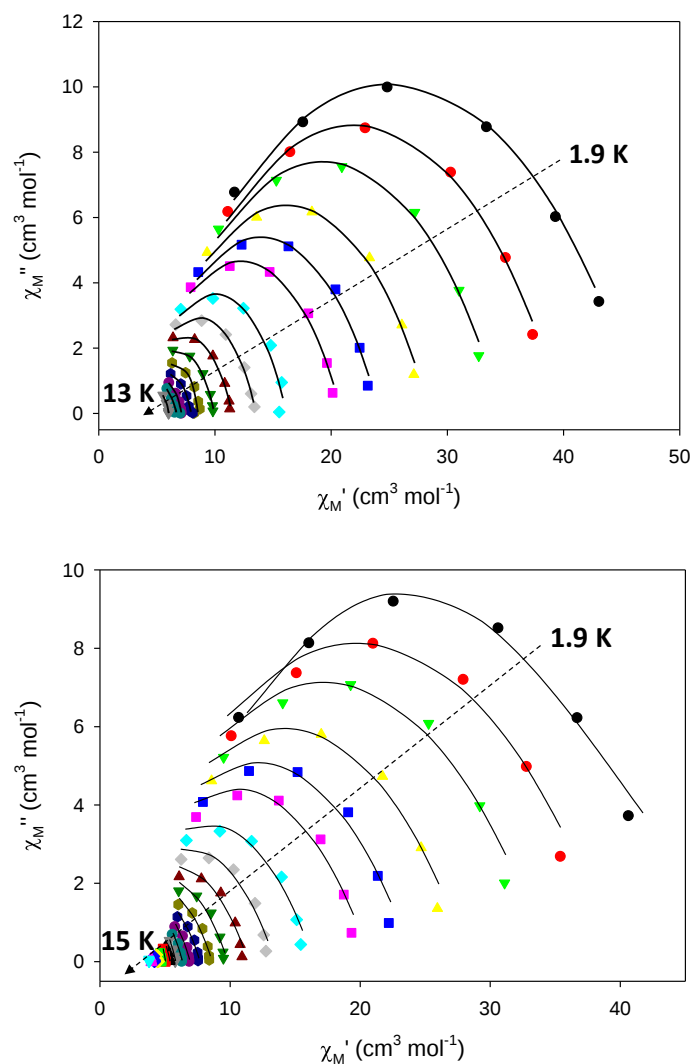


Figure S6. Cole-Cole plots for 1:3MeOH-CH₂Cl₂ obtained using the ac susceptibility data at zero (**top**) and 200 Oe (**bottom**) applied dc fields. The solid lines are the best fit obtained from a generalized Debye model.