Electronic Supplementary Information:

Heteroleptic, polynuclear dysprosium(III)-carbamato complexes through in-situ carbon dioxide capture

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Table S1: Continuous Shape Measures for complex **1** obtained using Shape v. 2.1.

	Dy(1)	Dy(2)
OP-8	31.517	32.639
HPY-8	22.439	21.874
HBY-8	12.374	13.777
CU-8	11.376	7.759
SAPR-8	4.848	1.813
TDD-8	3.228	1.791
JGBF-8	11.631	15.403
JETBPY-8	25.579	25.919
JBTPR-8	4.146	2.876
BTPR-8	3.769	2.343
JSD-8	3.923	4.778
TT-8	12.072	8.251
ETBPY-8	21.600	23.990

Table S2: Continuous Shape Measures for complex **2** obtained using Shape v. 2.1.

	Dy(1)	Dy(2)	Dy(3)
OP-8	31.327	32.150	32.975
HPY-8	22.405	21.249	20.891
HBY-8	16.067	14.435	12.368
CU-8	14.291	9.588	12.024
SAPR-8	5.288	0.994	4.231
TDD-8	2.976	2.338	3.106
JGBF-8	12.219	13.195	11.526
JETBPY-8	24.056	28.597	27.556
JBTPR-8	2.969	2.457	2.942
BTPR-8	2.847	2.273	2.620
JSD-8	4.314	4.154	4.187
TT-8	14.918	10.320	12.529
ETBPY-8	22.372	24.043	23.276

Table S3: Continuous Shape Measures for complex **3** obtained using Shape v. 2.1.

	Dy(1)	Dy(2)	Dy(3)
OP-8	31.866	32.274	32.341
HPY-8	22.490	20.790	20.054
HBY-8	15.309	14.887	14.665
CU-8	12.494	9.819	12.805
SAPR-8	4.361	0.997	4.665
TDD-8	2.493	2.448	2.276
JGBF-8	12.024	13.596	12.558
JETBPY-8	24.379	28.690	27.733
JBTPR-8	3.465	2.652	3.583
BTPR-8	3.470	2.374	3.113
JSD-8	3.860	4.430	3.717
TT-8	13.272	10.531	13.266
ETBPY-8	23.100	23.892	21.675

Table S4: Energy separation and g-factors for compounds **(1)**, **(2)** and **(3)** obtained from CASSCF.

	ΔE [cm^{-1}]	ΔE [K]	g_x	g_y	g_z
(1) Dy(1)	23.35	33.60	1.0417	6.5595	14.0246
(1) Dy(2)	42.18	60.69	0.0679	0.1846	19.5099
(2) Dy(1)	91.86	132.16	0.0300	0.0596	19.7198
(2) Dy(2)	57.98	83.42	0.3206	0.4356	18.7902
(2) Dy(3)	85.79	123.43	0.1981	0.5220	18.9881
(3) Dy(1)	97.27	139.95	0.0244	0.0447	19.6749
(3) Dy(2)	95.56	137.49	0.1099	0.2193	19.1709
(3) Dy(3)	93.04	133.86	0.0426	0.1239	19.2214

Table S5: Calculated dipolar interactions.

	(1)	(2)	(3)
J_1 [cm^{-1}]	-0.858	-0.196	-1.782
J_2 [cm^{-1}]	-0.681	-0.061	-0.263
J_3 [cm^{-1}]	-0.103	-0.178	-0.194
J_4 [cm^{-1}]	--------	-0.124	-0.330

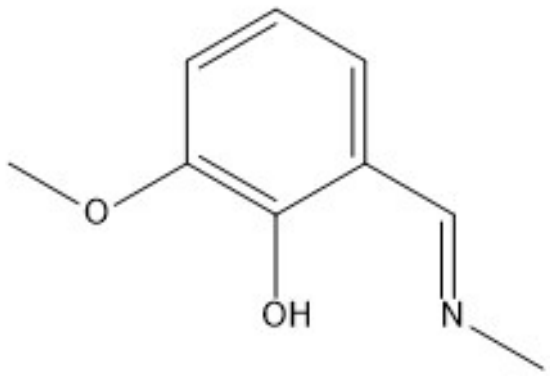

Fig. S1: Structure of 2-Hydroxy-3-methoxybenzaldehyde-N-methylimine

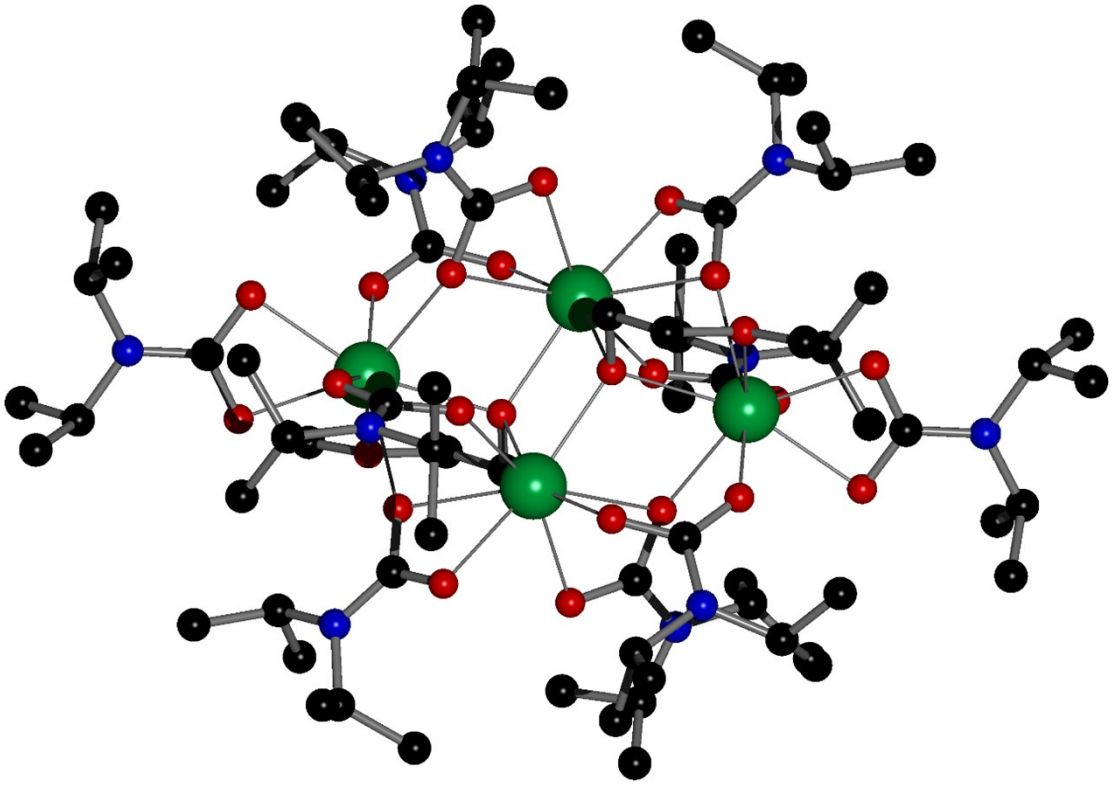

Fig. S2: Full structure of $[Dy_4(O_2CN^iPr_2)_{10}(O\text{-}C_2H_4\text{-}OMe)_2]$ (**1**); Dy: green; O: red; N: blue; C: black; H omitted for clarity.

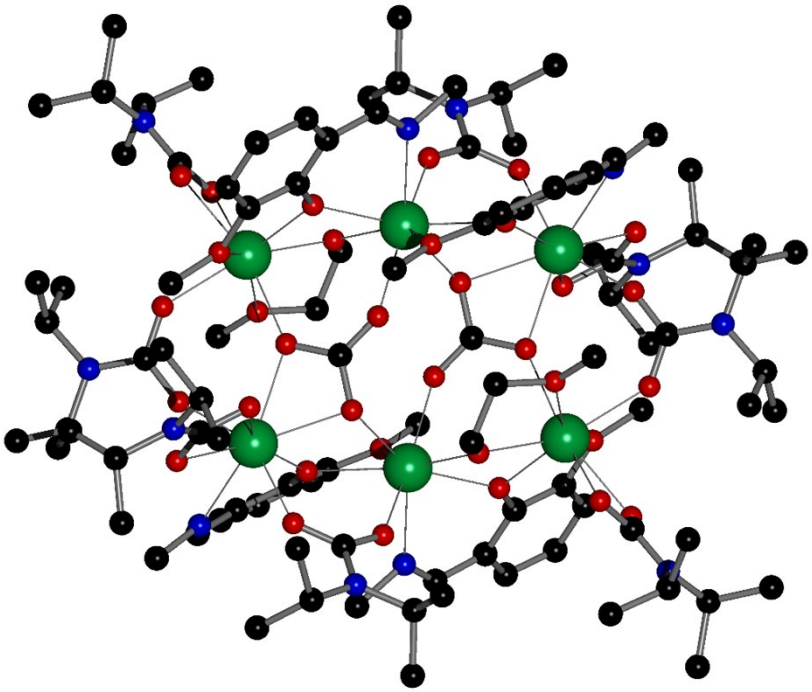

Fig. S3: Full structure of $[Dy_6(O_2CN^iPr_2)_8(O\text{-}C_2H_4\text{-}OMe)_2(CO_3)_2(C_9O_2NH_{10})_4]$ (**2**); Dy: green; O: red; N: blue; C: black; H omitted for clarity.

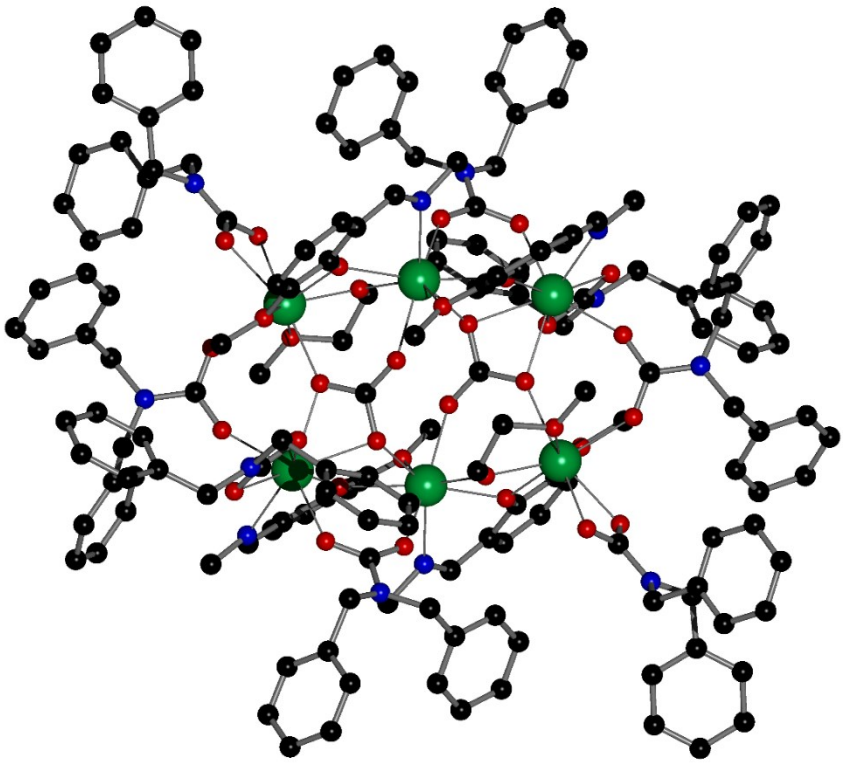

Fig. S4: Full structure of $[Dy_6(O_2CNBz_2)_8(O\text{-}C_2H_4\text{-}OMe)_2(CO_3)_2(C_9O_2NH_{10})_4]$ (**3**); Dy: green; O: red; N: blue; C: black; H omitted for clarity.

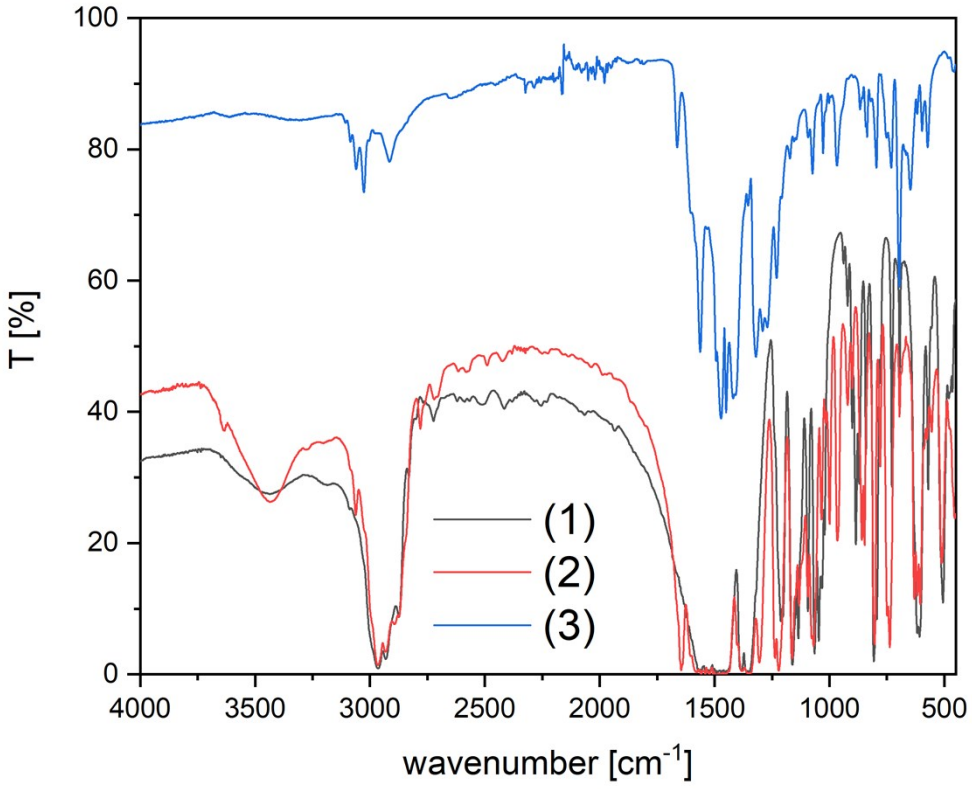

Fig. S5: Infrared transmission spectrum for compounds **(1)** , **(2)** and **(3)**.

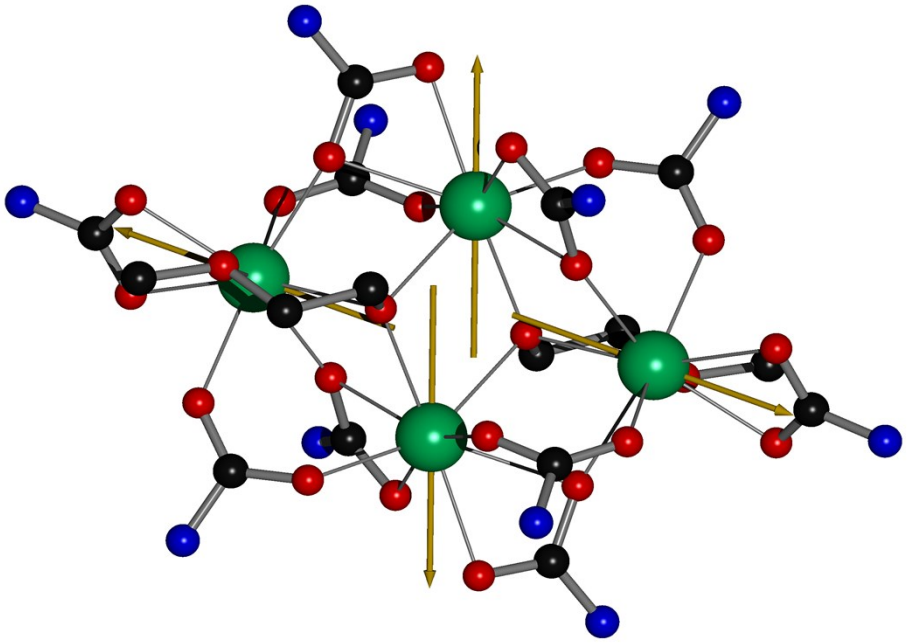

Fig. S6: Orientation of the magnetic z-axes in (**1**).

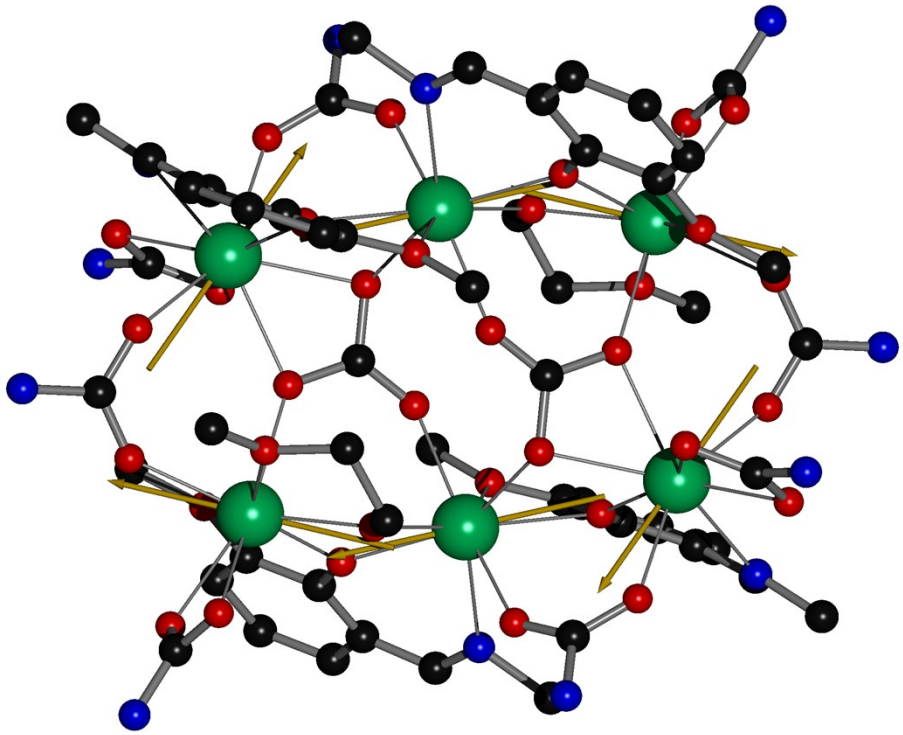

Fig. S7: Orientation of the magnetic z-axes in (**2**).

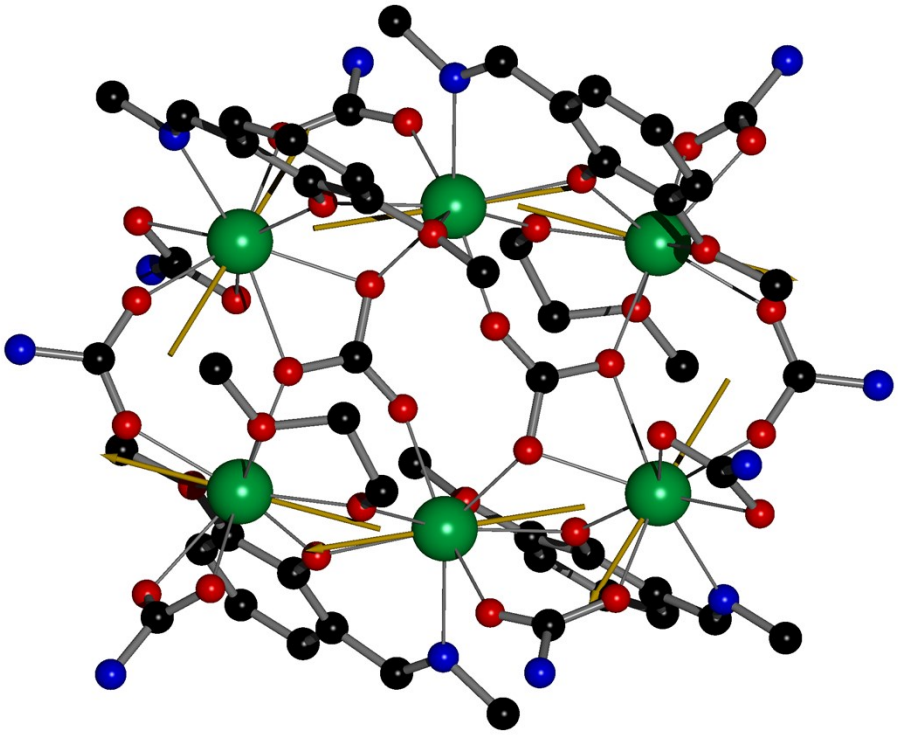

Fig. S8: Orientation of the magnetic z-axes in (**3**).