Supporting Information

A Nonsymmetric Dy_2 Single-Molecule Magnet with two Relaxation Processes Triggered by External Magnetic Field: Theoretical and Integrated EPR study of the Role of Magnetic-Site Dilution

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Tables

Figures

Table S1. Crystal data and structure refinement for **1**·MeOH at 106.8(2) and **Dy@Y2** at 107.7(6) K.

Empirical formula	$C_{74}H_{62}Dy_2O_{14}$	$C_{74}H_{62}Dy_{0.14}O_{14}Y_{1.86}$
Formula weight	1500.23	1363.18
Temperature	106.8(2) K	107.7(6) K
Wavelength	1.54184 Å	1.54184 Å
Crystal system	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$
Unit cell dimensions	a = 11.2128(4) Å b = 16.0056(7) Å c = 18.6715(9) Å α = 78.035(4)° β = 75.762(4)° γ = 72.617(4)°	a = 11.1868(4) Å b = 16.0404(6) Å c = 18.6620(6) Å α = 78.186(3)° β = 75.862(3)° γ = 72.605(3)°
Volume	3067.0(2) Å^3	3067.2(2) Å^3
Z	2	2
Density (calculated)	1.624 g/cm^3	1.476 g/cm^3
Absorption coefficient	13.456 mm^{-1}	3.813 mm^{-1}
F(000)	1500	1399
Crystal size	0.09 x 0.06 x 0.05 mm	0.05x 0.03 x 0.03 mm
θ range for data collection	3.54 to 73.17°	3.54 to 73.55°
Index ranges	-8<=h<=13, -17<=k<=19, -22<=l<=22	-13<=h<=11, -19<=k<=19, -22<=l<=20
Reflections collected	20441	20366
Independent reflections	11046 [R_{int} = 0.0322]	11053 [R_{int} = 0.0391]
θ/completeness	67.5°/99.9%	67.5°/99.9%
Refinement method	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2
Data / restraints / parameters	11046 / 44 / 825	11053 / 50 / 827
Goodness-of-fit on F^2	1.037	1.019
Final R indices [I > 2σ(I)]	R_1 = 0.0382, wR_2 = 0.0969	R_1 = 0.044, wR_2 = 0.113
R indices [all data]	R_1 = 0.0454, wR_2 = 0.1022	R_1 = 0.056, wR_2 = 0.123
Largest diff. peak and hole	1.892 and -0.670 e·Å^{-3}	1.673 and -0.703 e·Å^{-3}

$R_1 = \Sigma||F_o|-|F_c|| / \Sigma|F_o|$, $wR_2 = \{\Sigma[w(F_o^2 - F_c^2)^2] / \Sigma[w(F_o^2)^2]\}^{1/2}$ and $w=1/[\sigma^2(F_o^2)+(aP)^2+bP]$ where $P=(F_o^2+2F_c^2)/3$ and a and b are the two weighting parameters suggested by the SHELXL refinement software.

Table S2. Coordination geometry analysis for the 8-coordinated metal atoms Dy1 and Dy2 of **1**·MeOH by the SHAPE v2.1 software.

	CShM value		Symmetry	Polyhedron
1·MeOH	**Dy1**	**Dy2**		
	28.802	28.284	D_{8h}	Octagon
	23.095	22.121	C_{7v}	Heptagonal pyramid
	15.634	16.787	D_{6h}	Hexagonal bipyramid
	9.179	9.596	O_h	Cube
	0.607	**0.494**	$\boldsymbol{D_{4d}}$	**Square antiprism**
	1.959	1.781	D_{2d}	Triangular dodecahedron
	14.268	15.472	D_{2d}	Johnson gyrobifastigium J26
	27.337	27.330	D_{3h}	Johnson elongated triangular bipyramid J14
	2.502	2.535	C_{2v}	Biaugmented trigonal prism J50
	1.894	1.788	C_{2v}	Biaugmented trigonal prism
	4.921	4.484	D_{2d}	Snub diphenoid J84
	9.808	10.354	T_d	Triakis tetrahedron
	22.661	22.574	D_{3h}	Elongated trigonal bipyramid

Table S3. Selected Bond lengths [Å] for **1**·MeOH at 106.8(2) K with estimated standard deviations in parentheses.

Distance	Value
O1-DY2	2.342(3)
O1-DY1	2.360(3)
O1M-DY2	2.383(3)
O2-DY1	2.428(3)
O2M-H2M	0.852(10)
O3-DY1	2.302(3)
O4-DY1	2.362(3)
O5-DY1	2.255(3)
O6-DY1	2.336(3)
O7-DY1	2.365(3)
O7-DY2	2.376(3)
O8-DY2	2.402(3)
O9-DY2	2.360(3)
O10-DY2	2.337(3)
O10-DY1	2.403(3)
O11-DY2	2.266(3)
O12-DY2	2.289(3)
DY1-DY2	3.5669(3)

Table S4. Bond angles [°] for **1**·MeOH at 106.8(2) K with estimated standard deviations in parentheses.

Angle	Value
DY2-O1-DY1	98.69(10)
DY1-O7-DY2	97.57(10)
DY2-O10-DY1	97.63(10)
O5-DY1-O3	82.52(11)
O5-DY1-O6	70.62(10)
O3-DY1-O6	141.43(11)
O5-DY1-O1	142.54(10)
O3-DY1-O1	78.54(10)
O6-DY1-O1	138.66(10)
O5-DY1-O4	81.89(12)
O3-DY1-O4	69.98(12)
O6-DY1-O4	79.03(12)
O1-DY1-O4	120.29(12)
O5-DY1-O7	149.07(10)
O3-DY1-O7	108.92(11)
O6-DY1-O7	84.18(10)
O1-DY1-O7	68.30(10)
O4-DY1-O7	75.79(11)
O5-DY1-O10	117.15(11)
O3-DY1-O10	144.78(10)
O6-DY1-O10	73.65(10)
O1-DY1-O10	68.30(10)
O4-DY1-O10	137.76(11)
O7-DY1-O10	69.99(10)
O5-DY1-O2	76.84(10)
O3-DY1-O2	83.22(10)
O6-DY1-O2	115.44(10)
O1-DY1-O2	69.10(10)
O4-DY1-O2	147.66(11)
O7-DY1-O2	131.87(10)
O10-DY1-O2	74.32(10)
O5-DY1-DY2	157.48(8)
O3-DY1-DY2	115.11(8)
O6-DY1-DY2	98.82(7)
O1-DY1-DY2	40.47(7)
O4-DY1-DY2	116.42(9)

O7-DY1-DY2	41.33(7)
O10-DY1-DY2	40.49(7)
O2-DY1-DY2	90.87(7)
O11-DY2-O12	70.91(11)
O11-DY2-O10	77.11(11)
O12-DY2-O10	121.79(11)
O11-DY2-O1	110.84(11)
O12-DY2-O1	78.04(10)
O10-DY2-O1	69.70(10)
O11-DY2-O9	83.46(11)
O12-DY2-O9	146.14(11)
O10-DY2-O9	70.91(11)
O1-DY2-O9	133.23(10)
O11-DY2-O7	146.07(11)
O12-DY2-O7	136.91(11)
O10-DY2-O7	70.93(10)
O1-DY2-O7	68.40(10)
O9-DY2-O7	75.70(11)
O11-DY2-O1M	80.78(12)
O12-DY2-O1M	77.99(11)
O10-DY2-O1M	142.09(11)
O1-DY2-O1M	147.88(11)
O9-DY2-O1M	76.33(11)
O7-DY2-O1M	118.73(10)
O11-DY2-O8	144.88(11)
O12-DY2-O8	80.20(11)
O10-DY2-O8	136.76(10)
O1-DY2-O8	81.21(10)
O9-DY2-O8	112.88(11)
O7-DY2-O8	68.91(10)
O1M-DY2-O8	73.89(12)
O11-DY2-DY1	115.24(8)
O12-DY2-DY1	118.10(8)
O10-DY2-DY1	41.88(7)
O1-DY2-DY1	40.84(7)
O9-DY2-DY1	92.45(8)
O7-DY2-DY1	41.10(7)
O1M-DY2-DY1	159.64(8)

O8-DY2-DY1	95.64(7)

Table S5. Geometry (Å, °) of the strong hydrogen-bonding motifs in compound **1**•MeOH

D–H⋯*A*	*D*–H	H⋯*A*	*D*⋯*A*	*D*–H⋯*A*
O1M-H1M⋯O2M^{i}	0.82(6)	1.88(6)	2.681(5)	163(5)
O2M-H2M⋯O3	0.85(5)	1.88(5)	2.727(5)	176(6)

Symmetry codes: (i) 1+x, y, z.

Table S6. Bond lengths [Å] for **Y@Dy2** at 107.7(6) K with estimated standard deviations in parentheses.

Distance	Value
Y1-O5	2.254(3)
Y1-O3	2.286(3)
Y1-O6	2.313(3)
Y1-O10	2.390(3)
Y1-O1	2.354(3)
Y1-O4	2.343(3)
Y1-O7	2.331(3)
Y1-O2	2.427(3)
Y1-DY2	3.490(7)
Y1-Y2	3.549(1)
DY1-O5	2.199(9)
DY1-O6	2.401(11)
DY1-O7	2.461(7)
DY1-O2	2.266(8)
DY1-O3	2.237(14)
DY1-O10	2.388(12)
DY1-O1	2.313(9)

DY1-O4	2.460(10)
DY1-DY2	3.526(10)
DY1-Y2	3.585(9)
Y2-O11	2.248(3)
Y2-O12	2.284(3)
Y2-O7	2.383(3)
Y2-O1	2.330(2)
Y2-O8	2.414(3)
Y2-O1M	2.369(3)
Y2-O10	2.315(3)
Y2-O9	2.329(3)
DY2-O1	2.163(10)
DY2-O12	2.202(9)
DY2-O10	2.450(9)
DY2-O9	2.536(10)
DY2-O7	2.291(9)
DY2-O11	2.466(10)
DY2-O8	2.180(10)
DY2-O1M	2.434(9)

Table S7. Selected Bond angles [°] for Dy@Y2 at 107.7(6) K with estimated standard deviations in parentheses.

Distance	Value
O5-Y1-O3	81.90(10)
O5-Y1-O6	70.99(10)
O3-Y1-O6	141.47(11)
O5-Y1-O10	116.84(11)
O3-Y1-O10	144.31(10)
O6-Y1-O10	73.93(10)
O5-Y1-O1	141.66(10)
O3-Y1-O1	78.64(10)
O6-Y1-O1	138.75(10)
O10-Y1-O1	68.06(9)
O5-Y1-O4	81.81(11)
O3-Y1-O4	70.29(11)
O6-Y1-O4	78.99(11)
O10-Y1-O4	138.61(10)
O1-Y1-O4	121.01(11)

O5-Y1-O7	149.97(10)
O3-Y1-O7	109.25(11)
O6-Y1-O7	84.63(9)
O10-Y1-O7	70.48(9)
O1-Y1-O7	68.30(9)
O4-Y1-O7	76.40(10)
O5-Y1-O2	76.03(9)
O3-Y1-O2	82.83(9)
O6-Y1-O2	115.07(11)
O10-Y1-O2	73.78(9)
O1-Y1-O2	69.02(9)
O4-Y1-O2	147.25(10)
O7-Y1-O2	131.85(10)
O5-Y1-DY2	160.76(17)
O3-Y1-DY2	111.29(18)
O6-Y1-DY2	102.47(19)
O10-Y1-DY2	44.55(17)
O1-Y1-DY2	37.46(17)
O4-Y1-DY2	115.32(19)
O2-Y1-DY2	91.35(18)
O7-Y1-DY2	40.53(19)
O2-Y1-Y2	90.51(7)
O5-Y1-Y2	156.85(10)
O3-Y1-Y2	115.50(8)
O6-Y1-Y2	98.92(7)
O10-Y1-Y2	40.26(6)
O1-Y1-Y2	40.48(6)
O4-Y1-Y2	117.50(8)
O7-Y1-Y2	41.71(7)
O5-DY1-O6	70.2(3)
O5-DY1-O7	143.6(4)
O6-DY1-O7	80.0(3)
O5-DY1-O2	80.5(3)
O6-DY1-O2	117.9(5)
O7-DY1-O2	133.5(4)
O5-DY1-O3	84.2(4)
O6-DY1-O3	138.7(4)
O7-DY1-O3	106.4(5)

O2-DY1-O3	87.7(3)
O5-DY1-O10	119.1(6)
O6-DY1-O10	72.4(4)
O7-DY1-O10	68.3(3)
O2-DY1-O10	76.8(3)
O3-DY1-O10	148.5(4)
O5-DY1-O1	149.4(4)
O6-DY1-O1	135.8(5)
O7-DY1-O1	66.8(2)
O2-DY1-O1	72.5(2)
O3-DY1-O1	80.5(4)
O10-DY1-O1	68.8(3)
O5-DY1-O4	80.3(3)
O6-DY1-O4	75.0(2)
O7-DY1-O4	71.9(2)
O2-DY1-O4	151.0(5)
O3-DY1-O4	68.9(4)
O10-DY1-O4	131.9(3)
O1-DY1-O4	117.8(5)
O5-DY1-DY2	163.1(6)
O6-DY1-DY2	99.6(3)
O7-DY1-DY2	40.3(2)
O2-DY1-DY2	93.2(4)
O3-DY1-DY2	111.4(4)
O10-DY1-DY2	43.9(2)
O1-DY1-DY2	36.5(3)
O4-DY1-DY2	110.8(4)
O5-DY1-Y2	158.7(6)
O6-DY1-Y2	96.2(3)
O7-DY1-Y2	41.43(15)
O2-DY1-Y2	92.3(3)
O3-DY1-Y2	115.7(4)
O10-DY1-Y2	39.61(14)
O1-DY1-Y2	39.62(15)
O4-DY1-Y2	112.9(3)
O11-Y2-O12	71.22(10)
O11-Y2-O7	146.58(10)
O12-Y2-O7	135.90(10)

O11-Y2-O1	110.93(10)
O12-Y2-O1	77.82(9)
O7-Y2-O1	67.84(9)
O11-Y2-O8	144.61(10)
O12-Y2-O8	79.40(10)
O7-Y2-O8	68.73(9)
O1-Y2-O8	80.68(9)
O11-Y2-O1M	80.97(10)
O12-Y2-O1M	77.71(10)
O7-Y2-O1M	118.95(10)
O1-Y2-O1M	147.29(11)
O8-Y2-O1M	73.76(10)
O11-Y2-O10	77.47(10)
O12-Y2-O10	122.16(10)
O7-Y2-O10	70.87(9)
O1-Y2-O10	69.73(9)
O8-Y2-O10	136.52(9)
O1M-Y2-O10	142.68(11)
O11-Y2-O9	84.43(11)
O12-Y2-O9	146.71(10)
O7-Y2-O9	75.82(10)
O1-Y2-O9	133.49(9)
O8-Y2-O9	112.35(10)
O1M-Y2-O9	76.29(10)
O10-Y2-O9	71.63(9)
O11-Y2-Y1	115.59(8)
O12-Y2-Y1	118.13(7)
O7-Y2-Y1	40.61(6)
O1-Y2-Y1	41.00(6)
O8-Y2-Y1	95.31(7)
O1M-Y2-Y1	159.39(8)
O10-Y2-Y1	41.85(7)
O9-Y2-Y1	92.52(7)
O11-Y2-DY1	113.75(18)
O12-Y2-DY1	116.02(17)
O7-Y2-DY1	43.12(14)
O1-Y2-DY1	39.29(19)
O8-Y2-DY1	96.5(2)

O1M-Y2-DY1	161.97(14)
O10-Y2-DY1	41.1(2)
O9-Y2-DY1	94.20(19)
O1-DY2-O12	83.2(4)
O1-DY2-O10	70.0(3)
O12-DY2-O10	119.8(5)
O1-DY2-O9	131.1(3)
O12-DY2-O9	137.7(4)
O10-DY2-O9	66.0(2)
O1-DY2-O7	72.3(2)
O12-DY2-O7	148.7(5)
O10-DY2-O7	70.1(2)
O9-DY2-O7	73.5(3)
O1-DY2-O11	108.9(4)
O12-DY2-O11	68.6(3)
O10-DY2-O11	71.0(3)
O9-DY2-O11	75.9(3)
O7-DY2-O11	137.6(4)
O1-DY2-O8	90.0(4)
O12-DY2-O8	86.5(3)
O10-DY2-O8	143.1(4)
O9-DY2-O8	113.1(5)
O7-DY2-O8	74.5(3)
O11-DY2-O8	145.8(3)
O1-DY2-O1M	157.4(4)
O12-DY2-O1M	77.9(2)
O10-DY2-O1M	130.7(4)
O9-DY2-O1M	71.4(3)
O7-DY2-O1M	120.0(5)
O11-DY2-O1M	75.5(3)
O8-DY2-O1M	76.7(3)
O1-DY2-Y1	41.45(14)
O12-DY2-Y1	123.2(4)
O10-DY2-Y1	43.19(13)
O9-DY2-Y1	90.4(2)
O7-DY2-Y1	41.39(14)
O11-DY2-Y1	111.4(3)
O8-DY2-Y1	101.7(3)

O1M-DY2-Y1	158.8(4)
O1-DY2-DY1	39.5(2)
O12-DY2-DY1	120.9(3)
O10-DY2-DY1	42.5(3)
O9-DY2-DY1	92.0(3)
O7-DY2-DY1	43.98(18)
O11-DY2-DY1	109.8(5)
O8-DY2-DY1	102.9(4)
O1M-DY2-DY1	161.2(3)
DY2-O1-Y1	101.1(2)
Y2-O1-Y1	98.52(9)
DY2-O1-DY1	103.9(4)
Y2-O1-DY1	101.1(3)
DY1-O7-Y2	95.5(2)
DY1-O7-DY2	95.7(3)
Y2-O7-Y1	97.68(9)
DY2-O7-Y1	98.1(3)
DY2-O10-Y1	92.3(2)
Y1-O10-Y2	97.88(10)
DY2-O10-DY1	93.6(3)
Y2-O10-DY1	99.3(3)

Table S8. Parameters obtained by fitting the Cole-Cole plots (1500 dc field, Figure 3e) of 1·MeOH using the sum of two generalized Debye functions.

T	Δχ1	Δχ2	α1	α2	τ1	τ2
1.8	3.33313	6.609	0.11239	0.48806	0.08699	0.00183
1.9	3.20179	6.66823	0.14812	0.50379	0.0789	0.0018
2	2.93556	6.66126	0.15095	0.50456	0.07479	0.00185
2.5	2.21439	5.82446	0.1884	0.47805	0.04587	0.00164
3	1.81643	4.9345	0.21725	0.45188	0.02688	0.00132
3.5	1.82428	3.82966	0.24748	0.40776	0.01508	8.84003E-4
4	1.76695	3.04706	0.24002	0.37069	0.00921	6.02288E-4
4.5	1.51216	2.5898	0.21808	0.33713	0.00665	4.88965E-4
5	1.35823	2.18504	0.21401	0.31361	0.00475	3.90499E-4
6	0.90384	1.6648	0.14422	0.23779	0.00333	3.26681E-4
7	0.61128	1.28968	0.10199	0.19307	0.00237	2.78417E-4
8	0.41084	0.94096	0.03841	0.11414	0.00187	2.62749E-4
9	0.28782	0.68026	0.02576	0.06072	0.00142	2.48878E-4
10	0.22378	0.48196	0.01827	0	0.00105	2.25511E-4

Table S9. Parameters obtained by fitting the out-of-phase (Figure **5b**) versus T ac magnetic susceptibility signals for **Dy@Y2** in a 3.0 G ac field oscillating between 1488 and 10 Hz using the sum of two generalized Debye functions.

T	**α1**	**α2**	**τ1**	**τ2 (fixed) taken from Tab. S7**
1.8	0.50666	0.47611	0.12037	0.00183
1.9	0.52619	0.47155	0.12157	0.0018
2	0.548	0.48988	0.1174	0.00185
2.5	0.56303	0.49591	0.0973	0.00164
3	0.53468	0.46686	0.07734	0.00132
3.5	0.54144	0.47294	0.04837	8.84003E-4
4	0.52138	0.37567	0.02837	6.02288E-4
4.5	0.51715	0.3994	0.02073	4.88965E-4
5	0.49233	0.41928	0.01481	3.90499E-4
6	0.46589	0.40545	0.00803	3.26681E-4
7	0.41853	0.35795	0.0061	2.78417E-4
8	0.38227	0.30308	0.0041	2.62749E-4
9	0.40699	0.26806	0.0027	2.48878E-4

Table S10. Ab Initio CASSCF/RASSI-SO calculations of the energy barrier of the lowest 8 KD of both Dy centers in **1**·MeOH along with the deviation in the angle of the excited state anisotropic axis with respect to the ground state.

KD	Dy-1(cm^{-1})	Angle(°)	Dy-2(cm^{-1})	Angle	Dy-1 $g_{xx}/g_{yy}/g_{zz}$	Dy-2 $g_{xx}/g_{yy}/g_{zz}$
1	0	0	0	0	0.387/2.827/14.944	0.009/0.022/19.688
2	14.00	87.4	120.47	15.7	0.542/2.253/14.907	0.702/1.591/15.910
3	42.40	27.2	168.88	73.6	1.764/3.015/14.254	2.482/3.313/13.451
4	80.89	40.6	227.75	115.1	2.486/5.517/12.962	7.416/6.259/2.518
5	126.04	86.4	286.85	84.9	1.720/4.467/10.367	1.574/2.679/11.096
6	176.21	114.1	316.75	83.5	1.567/2.110/15.847	0.749/1.035/15.450
7	278.48	89.2	377.73	74.5	0.061/0.080/19.802	0.169/0.419/18.719
8	523.36	131.6	695.87	119.9	0.004/0.009/19.923	0.001/0.002/19.913

Table S11. *Ab initio* computed crystal field parameters for Dy-1 and Dy-2 centres of **1**·MeOH

Dy-1			Dy-2		
k	q	B(k,q)	k	q	B(k,q)
2	-2	-5.23E+00	2	-2	7.00E+00
	-1	8.99E+00		-1	-1.12E+01
	0	**2.85E+00**		**0**	**-3.28E+00**
	1	-4.98E+00		1	-8.78E+00
	2	-2.24E-01		2	-1.94E+00
4	-4	-3.66E-02	4	-4	-5.15E-03
	-3	1.34E-02		-3	-4.31E-01
	-2	-2.15E-02		-2	1.34E-01
	-1	2.58E-01		-1	-5.97E-02
	0	**4.21E-02**		**0**	**-3.81E-02**
	1	-7.04E-02		1	-5.44E-02
	2	8.44E-03		2	-3.87E-03
	3	1.48E-01		3	3.73E-01
	4	-3.50E-02		4	-1.54E-02
6	-6	-1.96E-03	6	-6	-2.34E-03
	-5	5.33E-03		-5	1.32E-02
	-4	-1.06E-03		-4	-9.31E-04
	-3	-3.80E-03		-3	-4.91E-03
	-2	-4.22E-03		-2	-1.83E-03
	-1	6.65E-03		-1	4.05E-03
	0	**2.33E-04**		**0**	**1.23E-04**
	1	-3.34E-04		1	3.90E-03
	2	-1.05E-03		2	-3.54E-03
	3	9.74E-04		3	4.98E-03
	4	4.09E-03		4	-3.79E-03
	5	-6.78E-03		5	6.70E-03
	6	-1.68E-03		6	4.10E-03

The following Hamiltonian is used for calculating the crystal field parameters.

$$H_{CF} = \sum_{k,q} B_k^q O_k^q$$

Where $O_k^q = extended\ stevens\ operator$

k=rank of ITO=2,4,6

q=component of the ITO=-k,-k+1,….,0,….,k

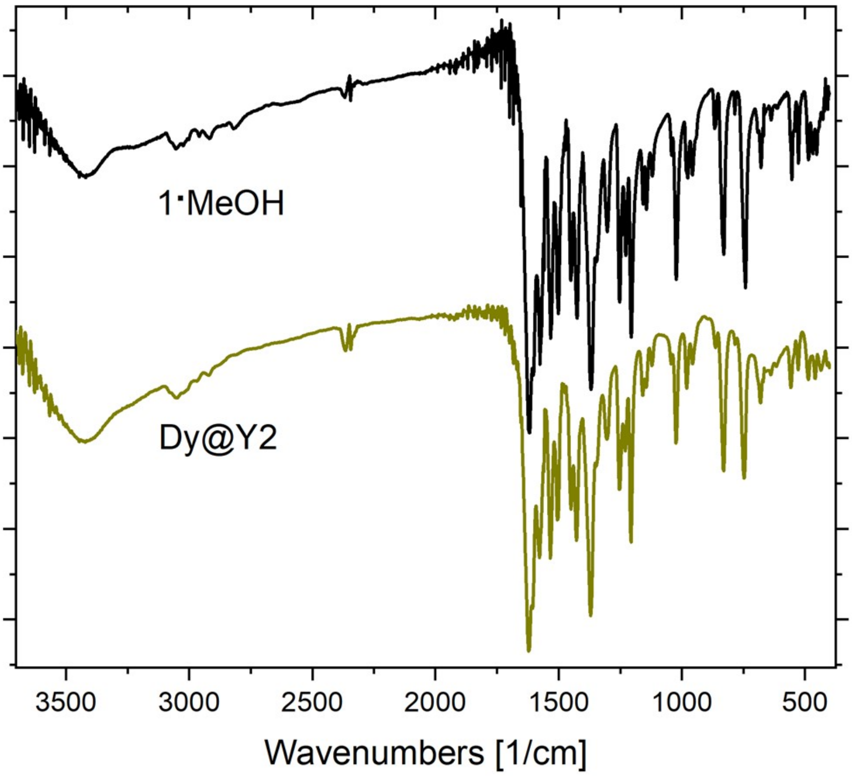

***Figure S 1**. The IR spectra (KBr, cm^{-1}) of **1**·MeOH and **Dy@Y2**.*

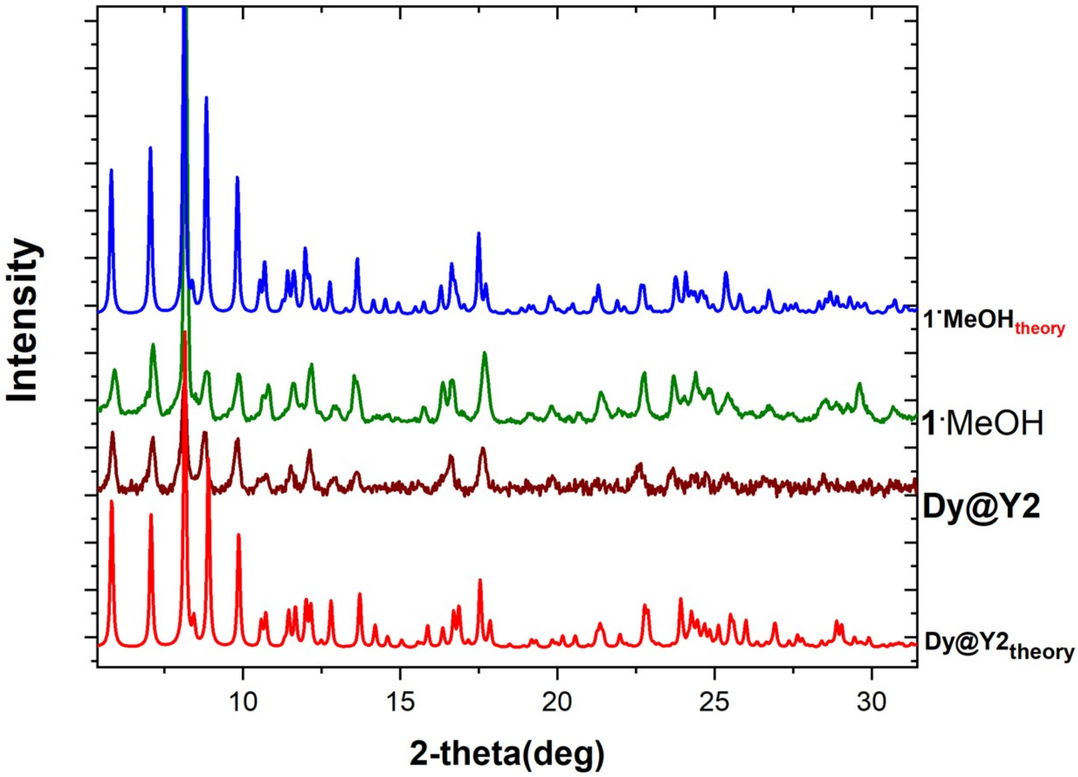

Figure S 2. *Experimental and theoretical X-ray powder patterns of **1**·MeOH and **Dy@Y2**.*

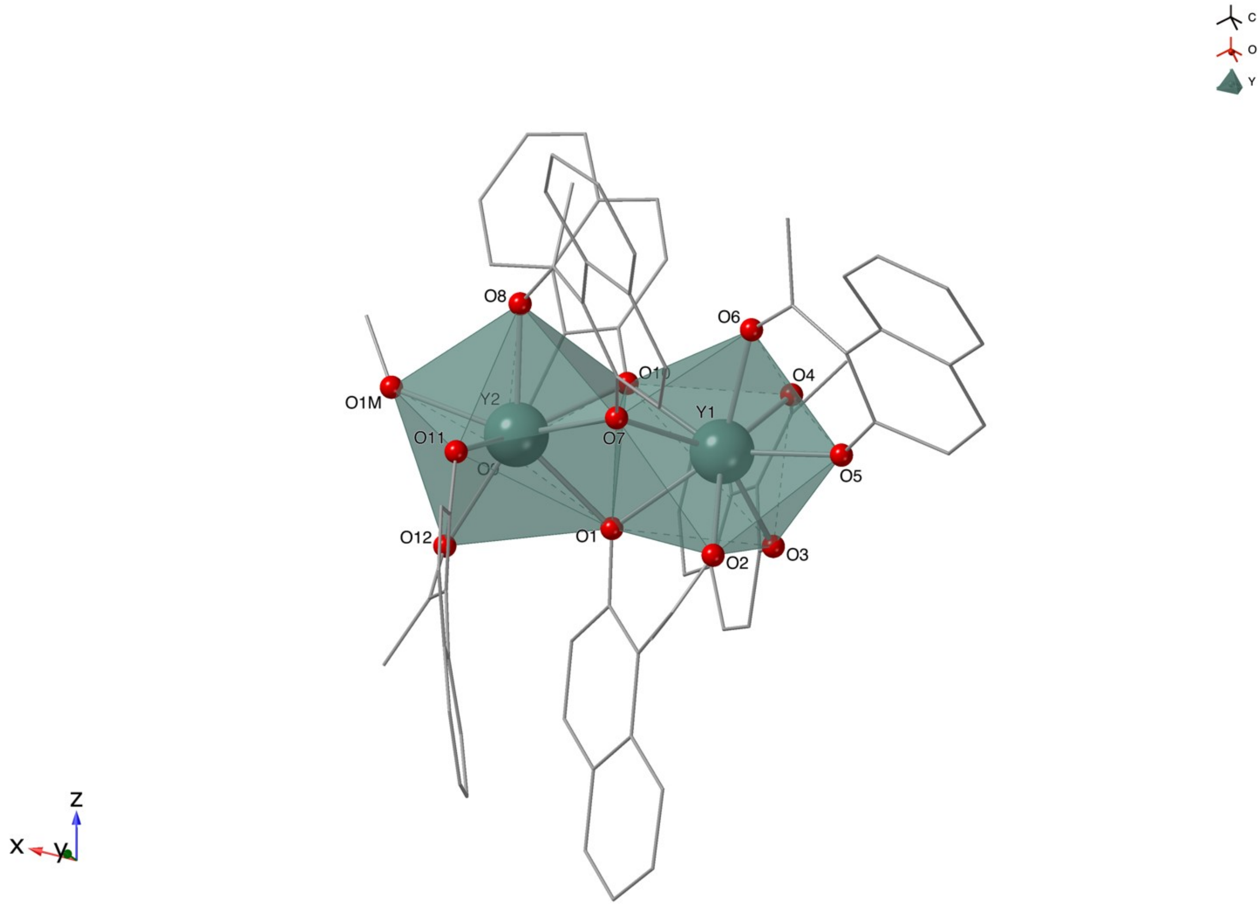

Figure S 3. *Partially labelled plot of the crystal structure of* ***Dy@Y2****. The polyhedral representations of the Y^{III} atoms are also shown. Hydrogen atoms and the dopant Dy^{III} ions have been omitted for the shake of clarity.*

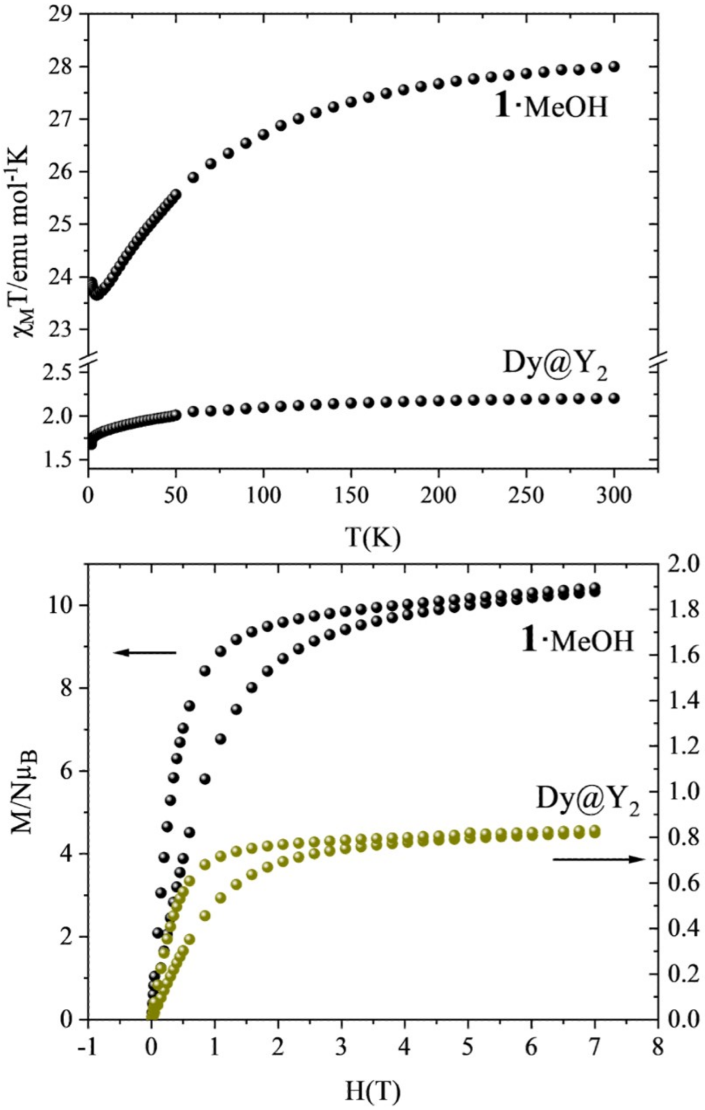

Figure S 4. ***(upper)****Dc magnetic susceptibility of of* ***1****·MeOH and Dy@Y2 in the 300-2 K temperature range under an applied field of 0.1 T****.(lower)*** *Isothermal magnetization curves of* ***1****·MeOH and Dy@Y2 at 2K and 5K.*

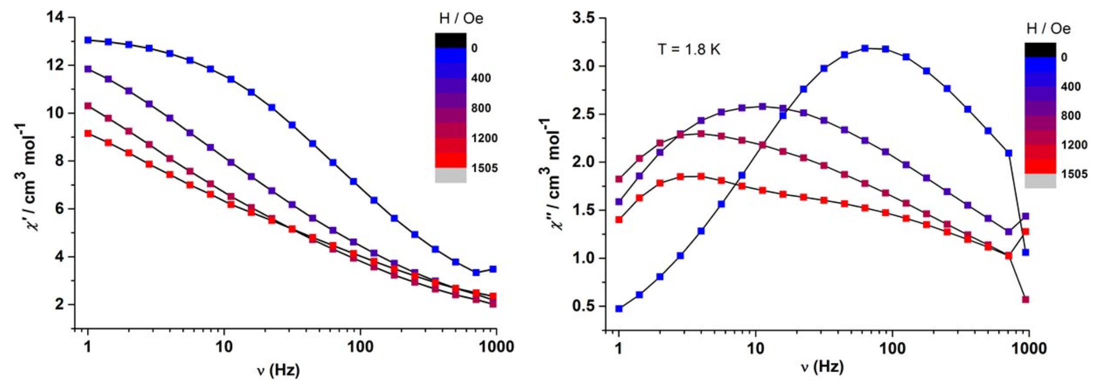

Figure S 5. *Field dependence of the out-of-phase ac susceptibility signals for* ***1***·*MeOH*

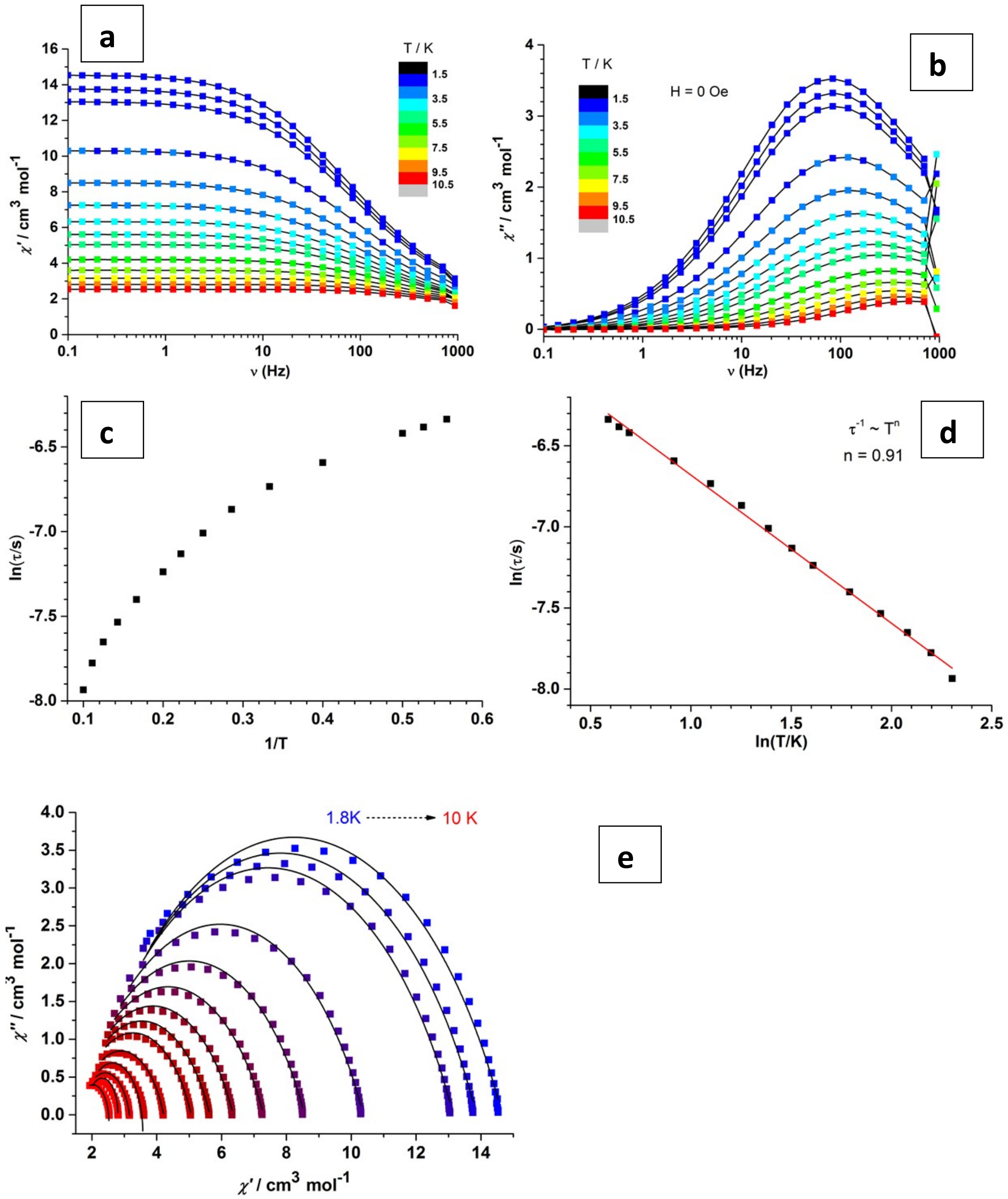

***Figure S 6. (a)-(b)** The weak temperature dependent χ'' signals were observed under zero dc field, indicating a small effective barrier.**(c)-(d)** No linear region is observed in Arrhenius fitting, while an exponential (T^n) fitting gives n value close to 1, suggesting the direct relaxation process present here.**(e)** The fitting of Cole-Cole plots with a generalized Debye model gives α values between 0.12 and 0.34. All curves are refer to **1**·MeOH*

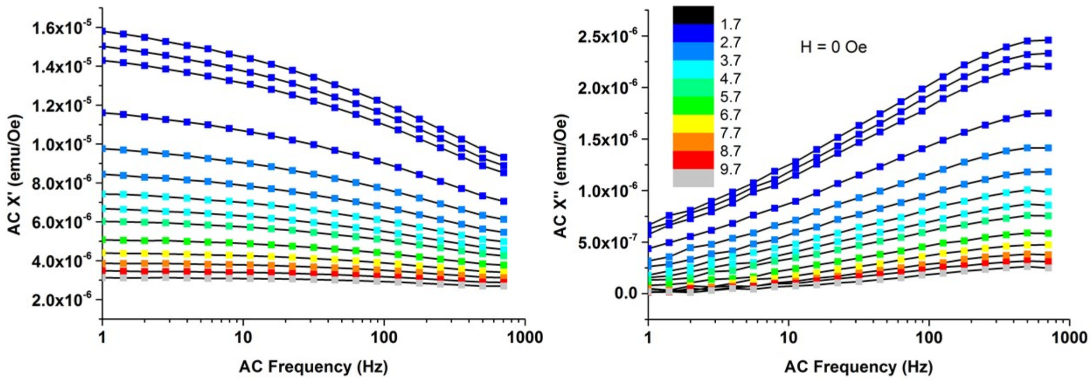

Figure S 7. *Under zero field, no χ_M'' peaks were observed for the sample* ***Dy@Y2***

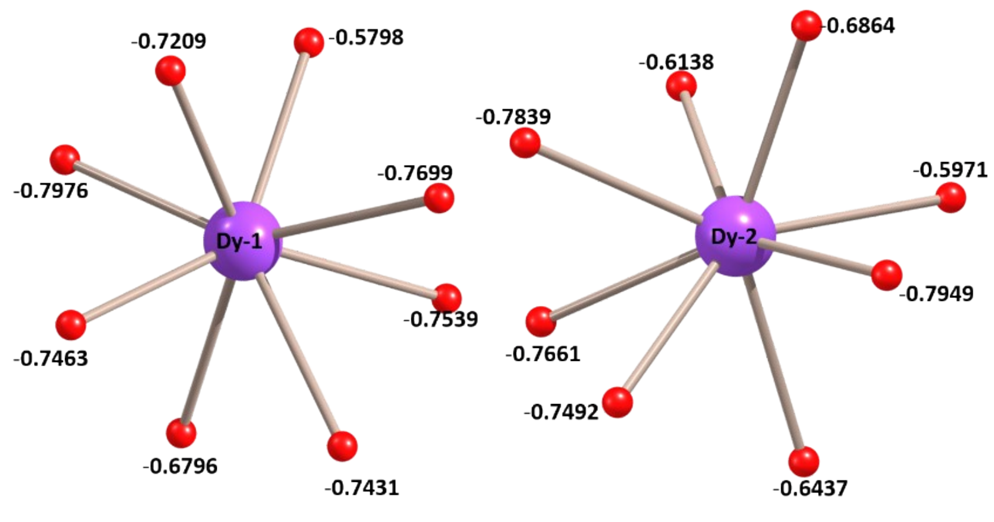

Figure S 8. *LoProp charge on the surrounding O of both Dy-1 and Dy-2 centres of* ***1****·MeOH*

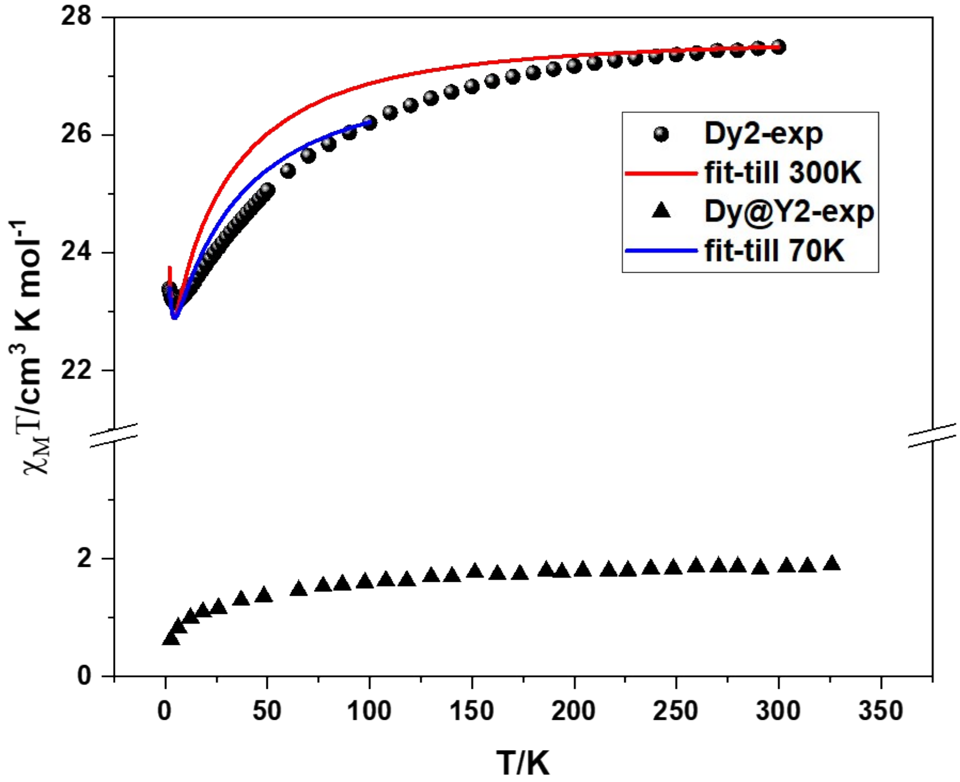

***Figure S 9**. Poly_aniso fitted magnetic susceptibility for sample **1**·MeOH (solid spheres). The magnetic susceptibility of **Dy@Y2** (solid triangles) is also presented for comparison reasons*

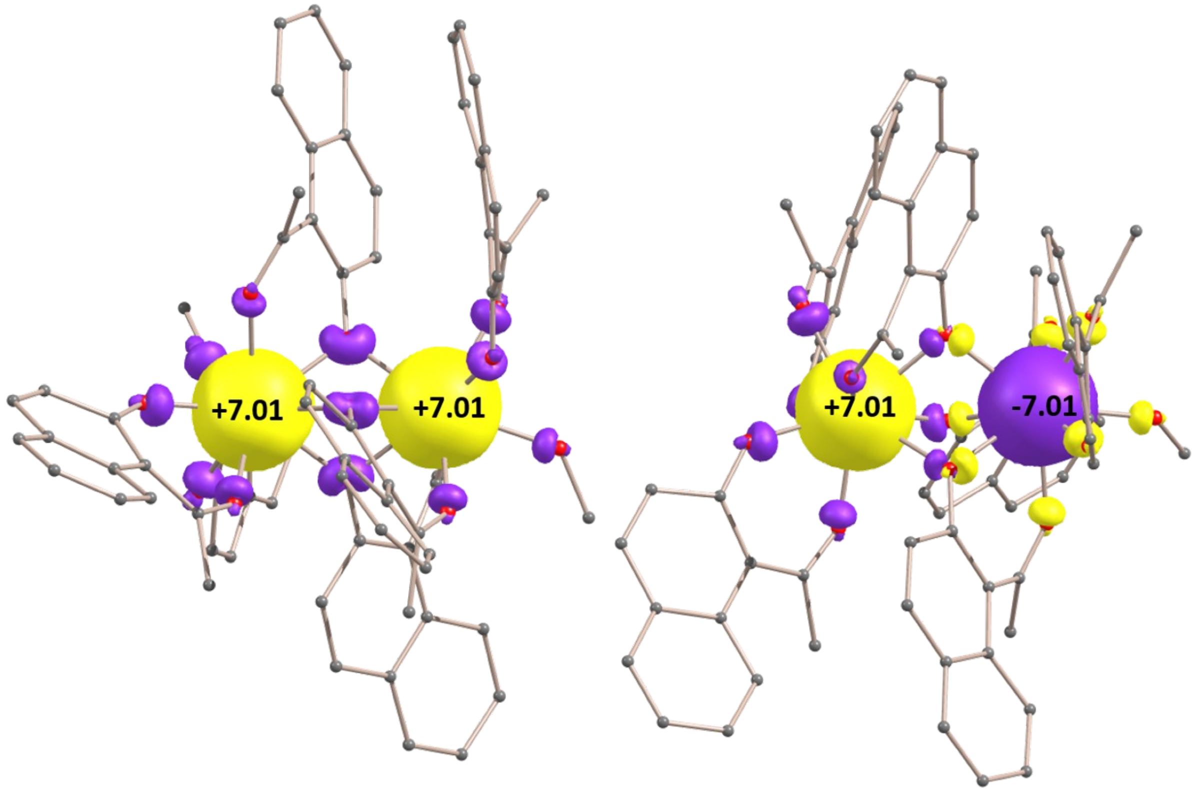

Figure S 10. *Spin density plots for the high spin and broken symmetry states, obtained from the density function calculations using broken symmetry approach for **1**·MeOH*

The exchange has been calculated using the following equation,

$$J_{exch} = \frac{E_{BS} - E_{HS}}{2S_1S_2 + S_2}$$

$E_{BS} = energy\ of\ broken\ symmetry\ state,\ E_{HS} = Enegry\ of\ high\ spin\ state$