Supplementary Material

Dy_4, Dy_5 and Ho_2 Complexes of an N_3O_2 Aminophenol Donor: A Dy_5-μ_3-Peroxide Single Molecule Magnet

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Table S1. Crystal data and structure refinement for **1**·2EtOH·H$_2$O-**3**·2.5H$_2$O

	1·2EtOH·H$_2$O	**2**·2H$_2$O	**3**·2.5H$_2$O
Empirical formula	$C_{65}H_{81}Cl_4Dy_4N_9O_{14}$	$C_{76}H_{70}Dy_5N_{16}O_{26}$	$C_{38}H_{39}Ho_2N_9O_{17.5}$
Molecular weight	2004.18	2435.98	1231.64
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	*I*2/*a*	*C*2/*c*	*C*2/*c*
Wavelength (Å)	0.71073	0.71073	0.71073
Crystal size (mm^3)	0.14 x 0.13 x 0.13	0.08 x 0.05 x 0.03	0.450 x 0.189 x 0.032
Color, shape	Yellow, block	Orange, plate	Yellow, plate
T (K)	100	100	100
a (Å)	21.2306(16)	20.628(2)	31.657(2)
b (Å)	16.4877(10)	20.1771(17)	11.4734(6)
c (Å)	49.810(4)	23.198(2)	25.8050(16)
α (º)	90	90	90
β (º)	93.267(3)	99.533(4)	108.408(3)
γ (º)	90	90	90
Volume (Å^3)	17407(2)	9522.2(15)	8893.0(9)
Z	8	4	4
Absorpt. coef. (mm^{-1})	3.573	3.952	3.617
Reflections collected	402343	98996	195235
Independent reflections	26570 [R_{int} = 0.0711]	11335 [R_{int}= 0.0676]	13578 [R_{int} = 0.0647]
Data / restraints / param.	26570 / 0 / 928	11335 / 0 / 558	13578 / 2 / 625
Final R indices [$I>2\sigma(I)$]	R_1 = 0.0302 wR_2 = 0.0660	R_1 = 0.0301 wR_2 = 0.0624	R_1 = 0.0282 wR_2 = 0.0674
R indices (all data)	R_1 = 0.0480 wR_2 = 0.0731	R_1 = 0.0467 wR_2 = 0.0683	R_1 = 0.0402 wR_2 = 0.0732

Table S2. Main bond distances (Å) and angles (º) for **1**·2EtOH·H_2O

Dy1-O1	2.342(2)	Dy3-O1	2.437(2)
Dy1-O12	2.355(2)	Dy3-O32	2.462(2)
Dy1-O11	2.380(2)	Dy3-O11	2.327(2)
Dy1-O1W	2.386(3)	Dy3-O31	2.352(2)
Dy1-N11	2.546(3)	Dy3-O2W	2.386(3)
Dy1-N13	2.546(3)	Dy3-N32	2.563(3)
Dy1-N12	2.563(3)	Dy3-N33	2.612(3)
Dy1-Cl1	2.6960(8)	Dy3-N31	2.638(3)
Dy2-O21	2.318(2)	Dy3-Cl2	2.8532(8)
Dy2-O22	2.351(2)	Dy4-O32	2.286(2)
Dy2-O3S	2.438(2)	Dy4-O22	2.294(2)
Dy2-N21	2.499(3)	Dy4-O1	2.297(2)
Dy2-N22	2.499(3)	Dy4-O31	2.332(2)
Dy2-N23	2.544(3)	Dy4-O21	2.336(2)
Dy2-Cl32	2.6633(9)	Dy4-O12	2.349(2)
Dy2-Cl31	2.7546(9)	Dy4-O4S	2.369(2)
Dy1···Dy3	3.8693(4)	Dy2···Dy4	3.8259(3)
Dy1···Dy4	3.7890(3)	Dy3···Dy4	3.5410(3)
N11-Dy1-N12	63.199	O32-Dy3-N33	63.529
O12-Dy1-N11	164.668	O1-Dy3-N32	152.228
N22-Dy2-N23	65.759	O22-Dy4-O21	67.218
N23-Dy2-Cl21	149.467	O22-Dy4-O1	162.108

Table S3. SHAPE v2.1. Continuous Shape Measures Calculation (c) 2013 Electronic Structure Group, Universitat de Barcelona

Geometries Coordination number 7

JETPY-7	7	C3v	Johnson elongated triangular pyramid J7
JPBPY-7	6	D5h	Johnson pentagonal bipyramid J13
CTPR-7	5	C2v	Capped trigonal prism
COC-7	4	C3v	Capped octahedron
PBPY-7	3	D5h	Pentagonal bipyramid
HPY-7	2	C6v	Hexagonal pyramid
HP-7	1	D7h	Heptagon

Geometries Coordination number 8

ETBPY-8	13	D3h	Elongated trigonal bipyramid
TT-8	12	Td	Triakis tetrahedron
JSD-8	11	D2d	Snub diphenoid J84
BTPR-8	10	C2v	Biaugmented trigonal prism
JBTPR-8	9	C2v	Biaugmented trigonal prism J50
JETBPY-8	8	D3h	Johnson elongated triangular bipyramid J14
JGBF-8	7	D2d	Johnson gyrobifastigium J26
TDD-8	6	D2d	Triangular dodecahedron
SAPR-8	5	D4d	Square antiprism
CU-8	4	Oh	Cube
HBPY-8	3	D6h	Hexagonal bipyramid
HPY-8	2	C7v	Heptagonal pyramid
OP-8	1	D8h	Octagon

Geometries Coordination number 9

MFF-9	13	Cs	Muffin
HH-9	12	C2v	Hula-hoop
JTDIC-9	11	C3v	Tridiminished icosahedron J63
TCTPR-9	10	D3h	Spherical tricapped trigonal prism
JTCTPR-9	9	D3h	Tricapped trigonal prism J51
CSAPR-9	8	C4v	Spherical capped square antiprism
JCSAPR-9	7	C4v	Capped square antiprism J10
CCU-9	6	C4v	Spherical-relaxed capped cube
JCCU-9	5	C4v	Capped cube J8
JTC-9	4	C3v	Johnson triangular cupola J3
HBPY-9	3	D7h	Heptagonal bipyramid
OPY-9	2	C8v	Octagonal pyramid
EP-9	1	D9h	Enneagon

1·2EtOH·H₂O

Dy1

Structure [ML8] ETBPY-8 TT-8 JSD-8 **BTPR-8** JBTPR-8 JETBPY-8
22.544, 8.607, 6.418, **3.797**, 4.603, 26.599,
JGBF-8 **TDD-8** SAPR-8 CU-8 HBPY-8 HPY-8 OP-8
8.619, **3.346**, 4.325, 7.849, 7.951, 22.154, 34.851

Dy2

Structure [ML8] ETBPY-8 TT-8 JSD-8 **BTPR-8** JBTPR-8 JETBPY-8
22.047, 12.094, 5.465, **3.212**, 4.183, 26.802,
JGBF-8 **TDD-8** SAPR-8 CU-8 HBPY-8 HPY-8 OP-8
12.963, **2.270**, 3.633, 11.584, 14.022, 22.159, 32.870

Dy3

Structure [ML9] **MFF-9** HH-9 JTDIC-9 TCTPR-9 JTCTPR-9 **CSAPR-9**
1.522, 9.720, 12.180, 3.169, 4.414, **2.544**,
JCSAPR-9 CCU-9 JCCU-9 JTC-9 HBPY-9 OPY-9 EP-9
3.110, 6.910, 7.860, 15.304, 15.770, 22.449, 35.291

Dy4

Structure [ML7] JETPY-7 JPBPY-7 **CTPR-7** **COC-7** PBPY-7
17.621, 10.669, **1.916**, **1.214**, 7.239,
HPY-7 HP-7
18.940, 33.080

2·2H₂O

Dy1

Structure [ML9] **MFF-9** HH-9 JTDIC-9 TCTPR-9 JTCTPR-9 **CSAPR-9**
2.703, 8.516, 11.653, 3.801, 4.882, **2.814**,
JCSAPR-9 CCU-9 JCCU-9 JTC-9 HBPY-9 OPY-9 EP-9
3.832, 8.572, 9.907, 14.411, 18.263, 21.994, 37.134

Dy2

Structure [ML9] **MFF-9** HH-9 JTDIC-9 TCTPR-9 JTCTPR-9 **CSAPR-9**
2.805, 8.292, 11.714, 3.637, 5.043, **2.788**,
JCSAPR-9 CCU-9 JCCU-9 JTC-9 HBPY-9 OPY-9 EP-9
3.788, 8.555, 9.950, 14.535, 18.071, 21.712, 37.261

Dy3

Structure [ML8] ETBPY-8 TT-8 JSD-8 **BTPR-8** JBTPR-8 JETBPY-8
22.968, 13.254, 5.593, **5.570**, 6.423, 26.800,
JGBF-8 **TDD-8** SAPR-8 CU-8 HBPY-8 HPY-8 OP-8
12.202, **5.460**, 7.197, 12.466, 12.034, 23.141, 27.530

3·2.5H₂O

Ho1

Structure [ML9] MFF-9 HH-9 JTDIC-9 **TCTPR-9** JTCTPR-9 **CSAPR-9**
1.809, 11.445, 11.026, **1.480**, 3.099, **1.214**,
JCSAPR-9 CCU-9 JCCU-9 JTC-9 HBPY-9 OPY-9 EP-9
1.975, 9.909, 10.999, 13.680, 19.597, 19.366, 36.809

Ho2

Structure [ML9] **MFF-9** HH-9 JTDIC-9 TCTPR-9 JTCTPR-9 **CSAPR-9**
1.931, 7.708, 12.027, 2.848, 3.205, **2.069**,
JCSAPR-9 CCU-9 JCCU-9 JTC-9 HBPY-9 OPY-9 EP-9
2.977, 8.299, 9.165, 13.934, 16.651, 21.585, 33.208

Table S4. Main bond distances (Å) and angles (º) for **2**·2H_2O

Dy1-O1	2.438(3)	Dy2-O1	2.295(2)	Dy3-O1	2.364(2)
Dy1-O2	2.312(2)	Dy2-O2	2.402(3)	Dy3-O2	2.370(3)
Dy1-O11	2.336(3)	Dy2-O21	2.322(3)	Dy3-O11	2.301(2)
Dy1-O12	2.274(3)	Dy2-O22	2.305(3)	Dy3-O21	2.323(3)
Dy1-O13	2.464(3)	Dy2-O23	2.496(3)	Dy1···Dy3	3.6317(4)
Dy1-O14	2.469(3)	Dy2-O24	2.461(3)	Dy2···Dy3	3.5923(4)
Dy1-N11	2.512(3)	Dy2-N21	2.514(4)	Dy1···Dy2	4.2636(6)
Dy1-N12	2.515(3)	Dy2-N22	2.483(3)		
Dy1-N13	2.557(3)	Dy2-N23	2.541(3)		
Dy1-N10	2.890(3)	Dy2-N20	2.895(4)		
O12-Dy1-O13	151.95(9)	O2-Dy2-N22	152.80(10)	$O1^{\#1}$-Dy3-O2	157.97(9)
O13-Dy1-O14	52.17(9)	O24-Dy2-O23	51.70(10)	O1-Dy3-O2	37.56(8)

$^{\#1}$ = -x+1,y,-z+1/2

Table S5. Main bond distances (Å) and angles (º) for **3**·2.5H_2O.

Ho1-O11	2.314(2)	Ho2-O11	2.346(2)
Ho1-O21	2.350(2)	Ho2-O21	2.306(2)
Ho1-O12	2.385(2)	Ho2-O22	2.355(2)
Ho1-O1W	2.401(2)	Ho2-O2W	2.390(2)
Ho1-O14	2.459(2)	Ho2-O23	2.436(2)
Ho1-O13	2.491(2)	Ho2-O24	2.504(2)
Ho1-N12	2.504(2)	Ho2-N22	2.528(3)
Ho1-N11	2.531(3)	Ho2-N23	2.529(3)
Ho1-N13	2.535(2)	Ho2-N21	2.529(3)
Ho1···Ho2	3.8091(2)		
O14-Ho1-O13	51.79(8)	O23-Ho2-O24	51.88(7)
O11-Ho1-N13	145.88(8)	O22-Ho2-O24	148.43(7)

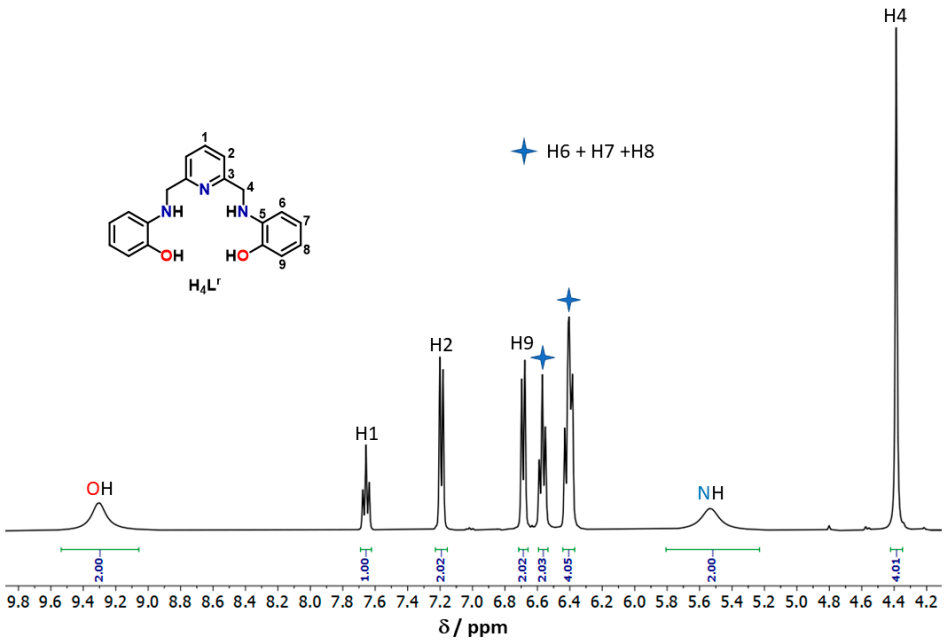

Figure S1. ^{1}H NMR spectrum for H_4L^r in DMSO-d_6 in the range 4.2-9.8 ppm.

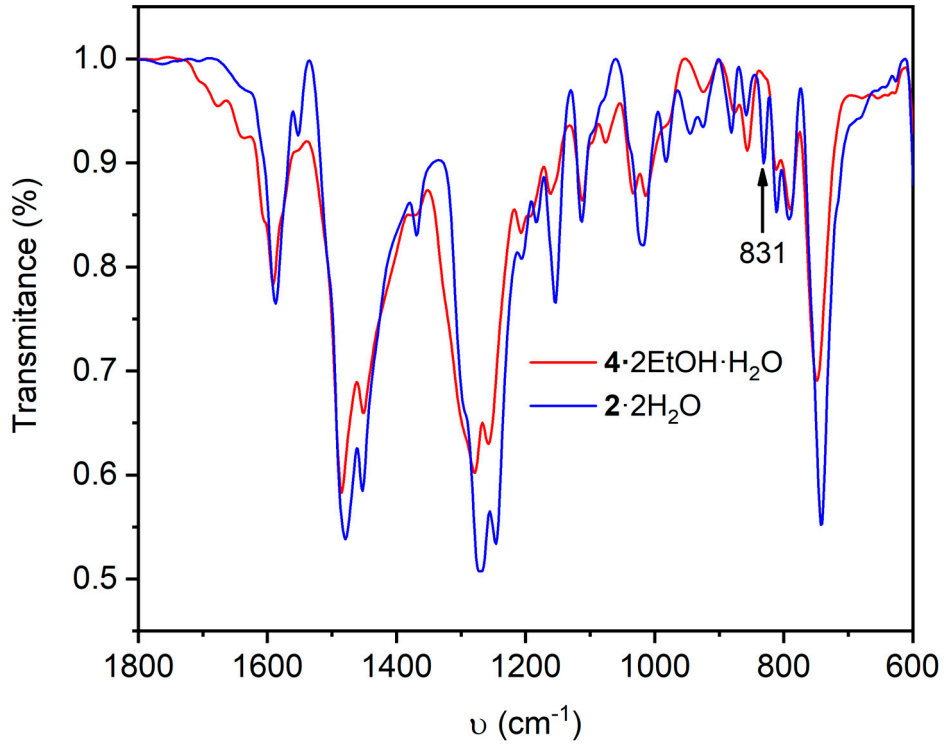

Figure S2. Comparative IR spectra for **2**·2H_2O and **4**·2EtOH·H_2O in the 1800-600 cm^{-1} region.

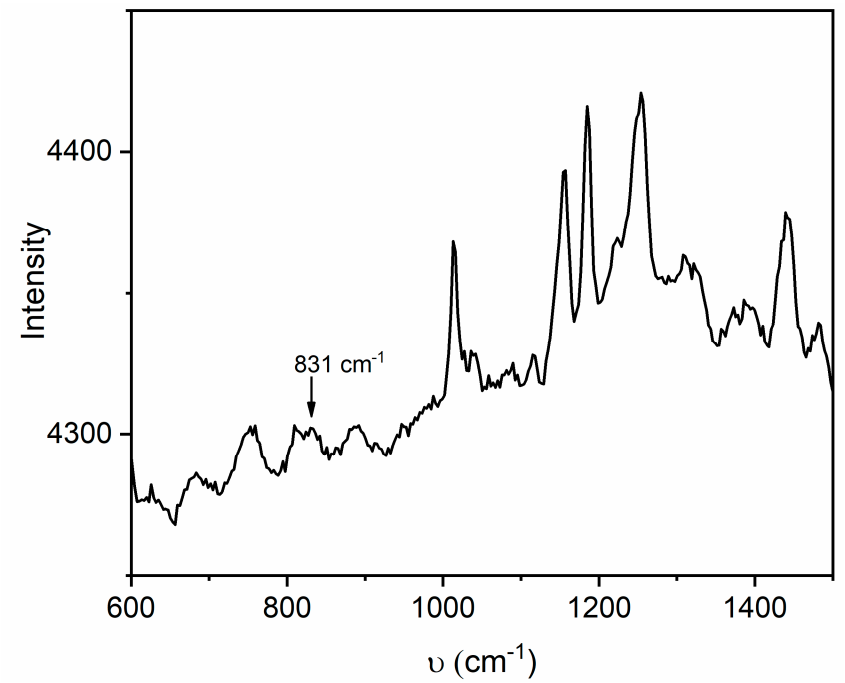

Figure S3. Raman spectrum for **2**·2H₂O in the 600-1500 cm⁻¹ region.

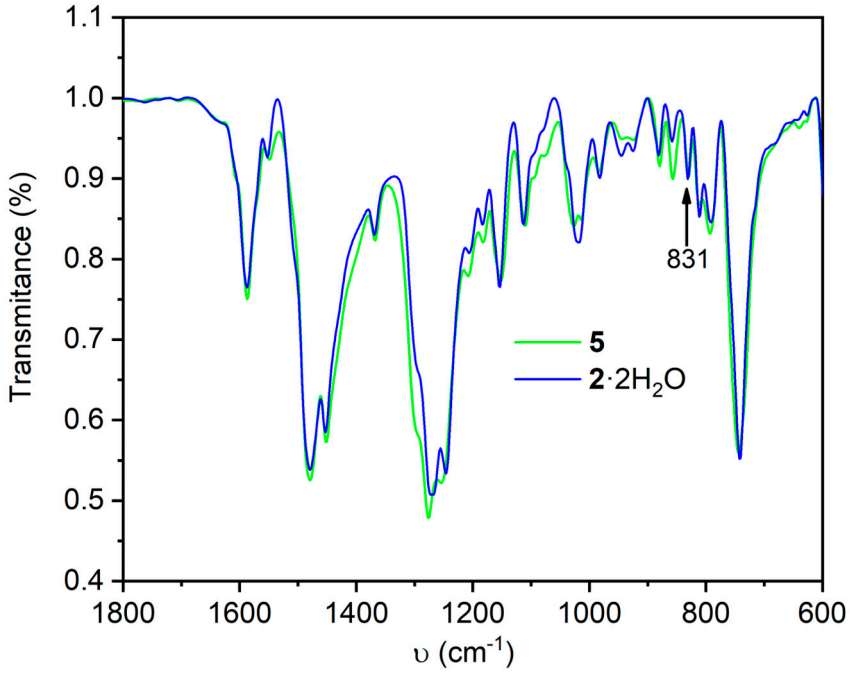

Figure S4. Comparative IR spectra for **2**·2H₂O and **5** in the 1800-600 cm⁻¹ region.

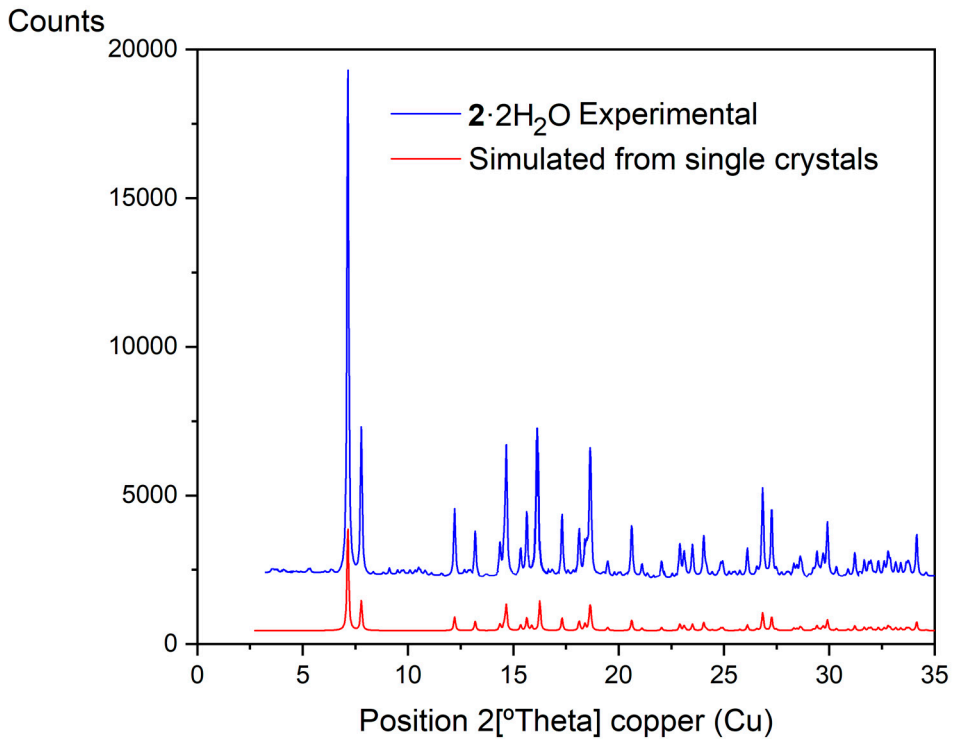

Figure S5. Comparison of X-powder diffractogram for the crystalline sample of **2**·2H2O with the simulated one from single X-ray data.

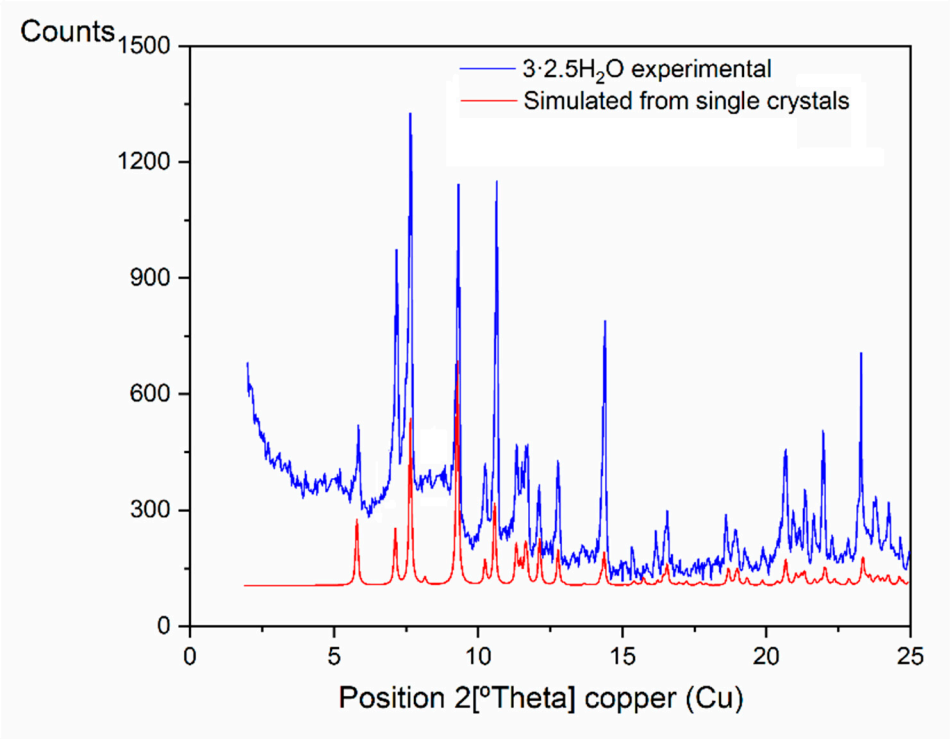

Figure S6. Comparison of X-powder diffractogram for the crystalline sample of **3**·2.5H2O with the simulated one from single X-ray data.

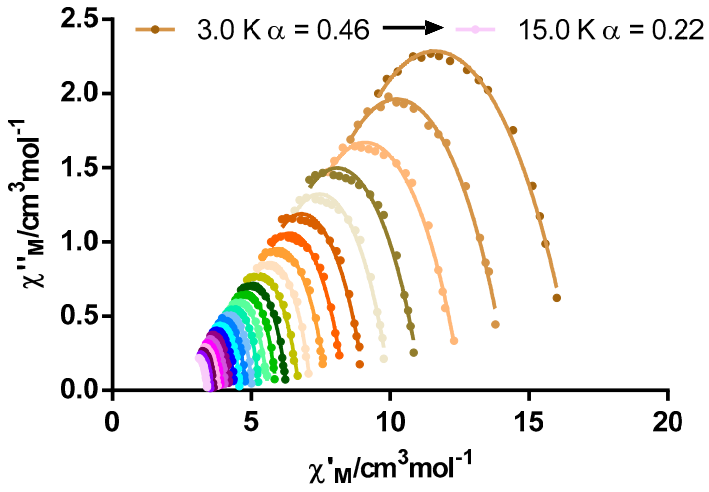

Figure S7. Cole-Cole plot for **2**·2H2O in a zero dc field

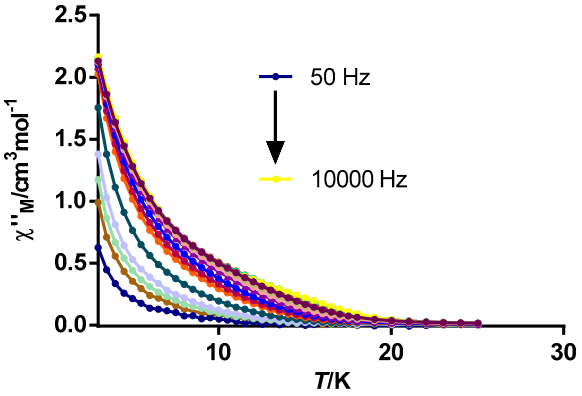

Figure S8. Temperature dependence of χ''_M for **2**·2H2O in a zero dc field at different frequencies.

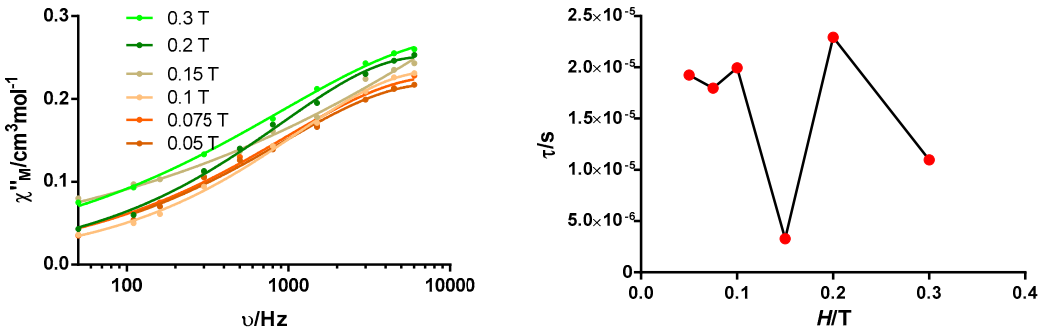

Figure S9. Left) frequency dependence of χ''_M for **1**·5H2O at 3 K under different magnetic fields; right) dependence of the relaxation time with the field.

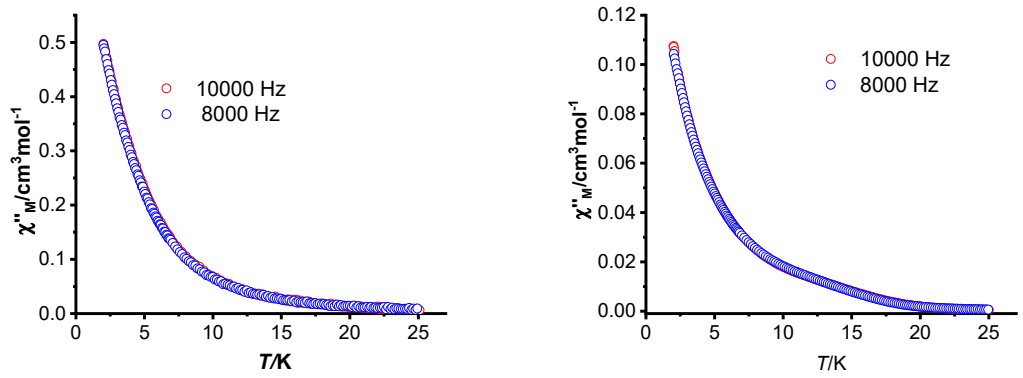

Figure S10. Temperature dependence of χ''_M in H_{dc} = 2000 Oe at two different frequencies for: left) **1**·5H₂O; right) **3**·2.5H₂O.

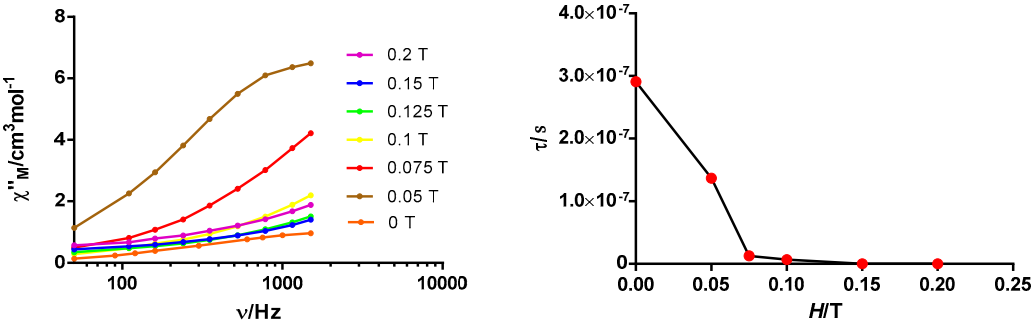

Figure S11. Left) frequency dependence of χ''_M for **2**·2H₂O at 6 K under different magnetic fields; right) dependence of the relaxation time with the field.