Table S1. Main crystallographic data for the crystal of complex **1**.

Empirical formula	$C_{42}H_{54}Cl_6N_2O_{14}Dy_2$ (**1**)
Formula weight	1348.56
Temperature	100.00(10) K
Wavelength	0.7107 Å
Crystal system, space group	Monoclinic, P 21/c
a, Å	10.3832(4)
b, Å	18.1959(10)
c, Å	13.7949(4)
alpha, deg.	90
beta, deg.	92.590(3)
gamma, deg.	90
Volume	2603.63(19) $Å^3$
Z, Calculated density	4, 1.720 Mg/m^3
Absorption coefficient	3.217 mm^{-1}
F(000)	1332
Crystal size	0.10 x 0.05 x 0.02 mm
Theta range for data collection	2.956 to 26.069 deg.
Limiting indices	-12<=h<=12, -15<=k<=22, -17<=l<=9
Reflections collected / unique	10218 / 5131 [R(int) = 0.0742]
Completeness to theta	25.242 99.8 %
Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	1.00000 and 0.54734
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	5131 / 12 / 298
Goodness-of-fit on F^2	0.818
Final R indices [I>2sigma(I)]	R1 = 0.0503, wR2 = 0.0734
R indices (all data)	R1 = 0.1010, wR2 = 0.0921
Extinction coefficient	n/a
Largest diff. peak and hole	1.604 and -1.334 $e.Å^{-3}$

Table S2. Selected bond lengths and valence angles in complex **1**.

Bond	d, Å	Bond	d, Å
Dy(1)-O(2)	2.312(4)	Dy(1)-O(5)	2.360(5)
Dy(1)-O(4)	2.321(5)	Dy(1)-O(7)#1	2.425(5)
Dy(1)-O(1)	2.335(5)	Dy(1)-O(7)	2.514(5)
Dy(1)-O(3)	2.340(5)	Dy(1)-O(6)	2.313(5)
Угол	ω, degree	Угол	ω, degree
O(6)-Dy(1)-O(2)	141.30(18)	O(3)-Dy(1)-O(5)	75.70(17)
O(6)-Dy(1)-O(4)	76.94(17)	O(6)-Dy(1)-O(7)#1	92.75(17)
O(2)-Dy(1)-O(4)	73.75(18)	O(2)-Dy(1)-O(7)#1	90.70(16)
O(6)-Dy(1)-O(1)	145.88(16)	O(4)-Dy(1)-O(7)#1	135.36(18)
O(2)-Dy(1)-O(1)	72.48(16)	O(1)-Dy(1)-O(7)#1	79.43(17)
O(4)-Dy(1)-O(1)	130.88(17)	O(3)-Dy(1)-O(7)#1	151.38(17)
O(6)-Dy(1)-O(3)	92.77(17)	O(5)-Dy(1)-O(7)#1	79.17(17)
O(2)-Dy(1)-O(3)	102.21(17)	O(6)-Dy(1)-O(7)	74.26(16)
O(4)-Dy(1)-O(3)	73.18(19)	O(2)-Dy(1)-O(7)	74.10(16)
O(1)-Dy(1)-O(3)	80.31(18)	O(4)-Dy(1)-O(7)	74.85(17)
O(6)-Dy(1)-O(5)	72.89(17)	O(1)-Dy(1)-O(7)	126.62(16)
O(2)-Dy(1)-O(5)	145.27(17)	O(3)-Dy(1)-O(7)	147.48(17)
O(4)-Dy(1)-O(5)	134.97(18)	O(5)-Dy(1)-O(7)	125.72(15)
O(1)-Dy(1)-O(5)	73.03(16)	O(7)#1-Dy(1)-O(7)	60.60(19)

Table S3. The local symmetry of Dy(III) ions for **1** defined by the continuous shape measure (CShM) analysis with SHAPE 2.1 software [54].

			Structure	CShM
1	D_{8h}	OP-8	Octagon	32.054
2	C_{7v}	HPY-8	Heptagonal pyramid	24.336
3	D_{6h}	HBPY-8	Hexagonal bipyramid	16.100
4	O_h	CU-8	Cube	9.942
5	D_{4d}	SAPR-8	Square antiprism	2.529
6	**D_{2d}**	**TDD-8**	**Triangular dodecahedron**	**0.492**
7	D_{2d}	JGBF-8	Johnson gyrobifastigium J26	13.453
8	D_{3h}	JETBPY-8	Johnson elongated triangular bipyramid J14	29.524
9	C_{2v}	JBTPR-8	Biaugmented trigonal prism J50	2.623
10	C_{2v}	BTPR-8	Biaugmented trigonal prism	2.183
11	D_{2d}	JSD-8	Snub diphenoid J84	2.802
12	T_d	TT-8	Triakis tetrahedron	10.614
13	D_{3h}	ETBPY-8	Elongated trigonal bipyramid	24.076

Table S4. SINGLE_ANISO computed wave function decomposition analysis for the lowest KDs of Dy(III) ion in **1**. Only main contributions (>10%) are shown

KD	RASSI wave function composition
1	**0.945 |±15/2›**
2	**0.795 |±13/2›**+0.176 |±9/2›
3	**0.225 |±11/2›**+ 0.211 |±1/2›+ 0.207 |±3/2›+ 0.175 |±7/2›+ 0.125 |±5/2›
4	**0.266 |±11/2›**+ 0.265 |±1/2›+0.233 |±7/2›+0.124 |±3/2›
5	**0.388 |±5/2›**+ 0.272 |±3/2›+0.148 |±1/2›+ 0.119|±9/2›
6	**0.298 |±9/2›**+0.234 |±7/2›+0.195 |±11/2›
7	**0.276 |±1/2›** + 0.256 |±3/2› +0.132 |±9/2›+ 0.122 |±5/2› +0.102 |±11/2›
8	**0.274 |±7/2›** +0.223 |±9/2› +0.186 |±5/2›+ 0.124|±11/2›

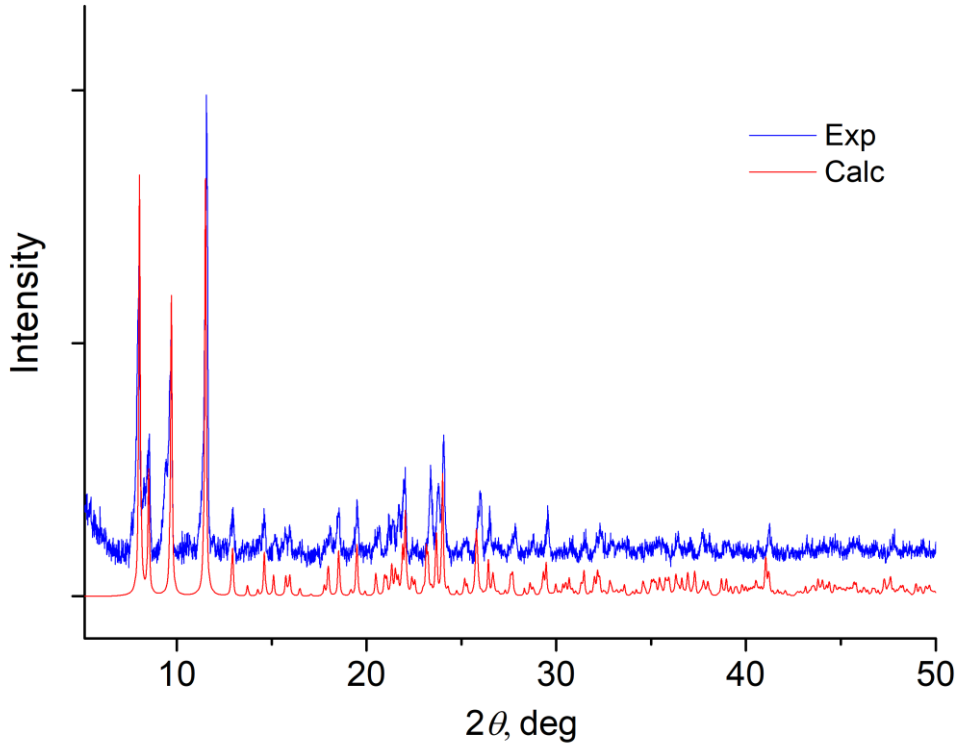


Figure S1. Powder X-ray diffraction pattern of polycrystalline sample of **1**: experimental (blue), and calculated from single crystal data (red).

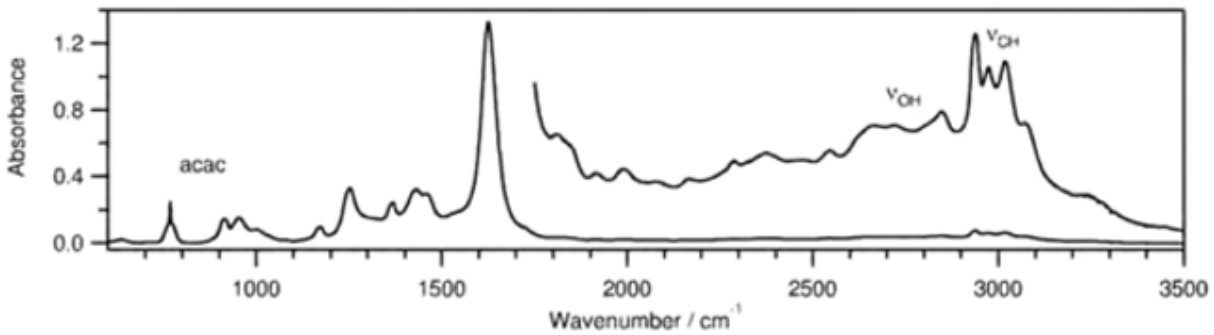


Figure S2. Experimental IR spectrum of acac **Error! Reference source not found.**.

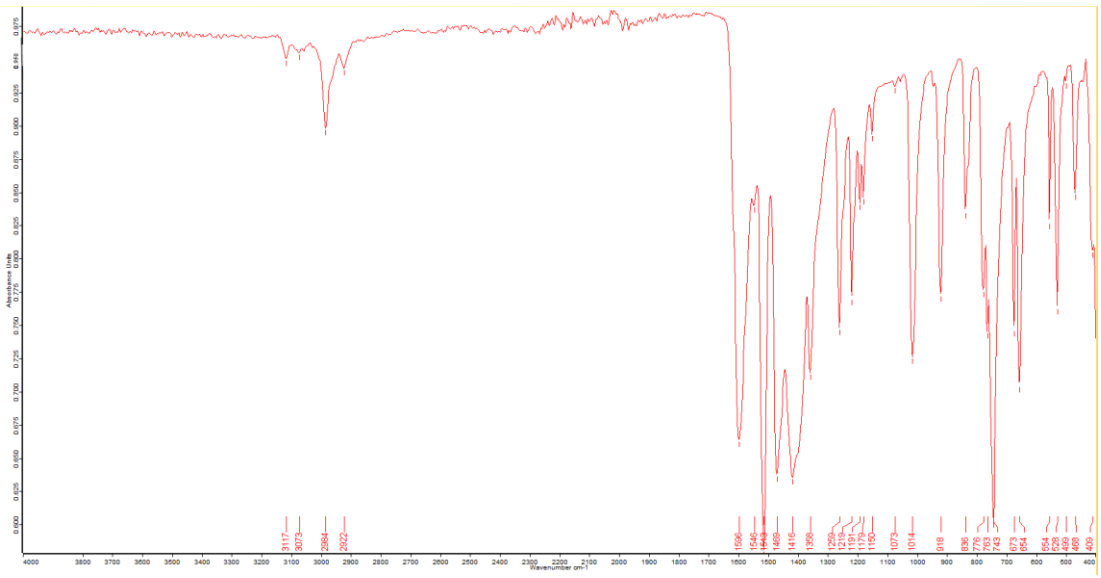


Figure S3. Experimental IR spectrum of **1**.

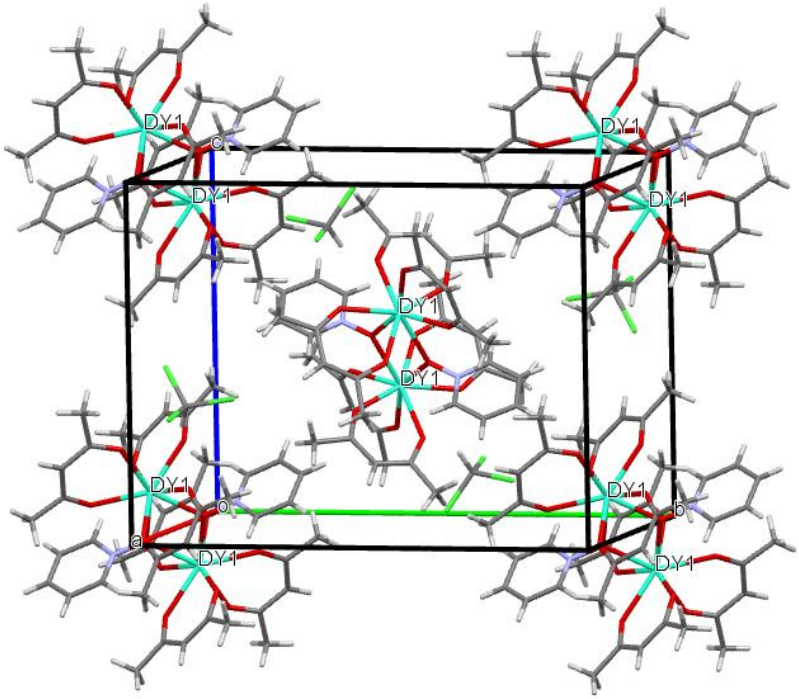


Figure S4. Crystalline packing of **1**.

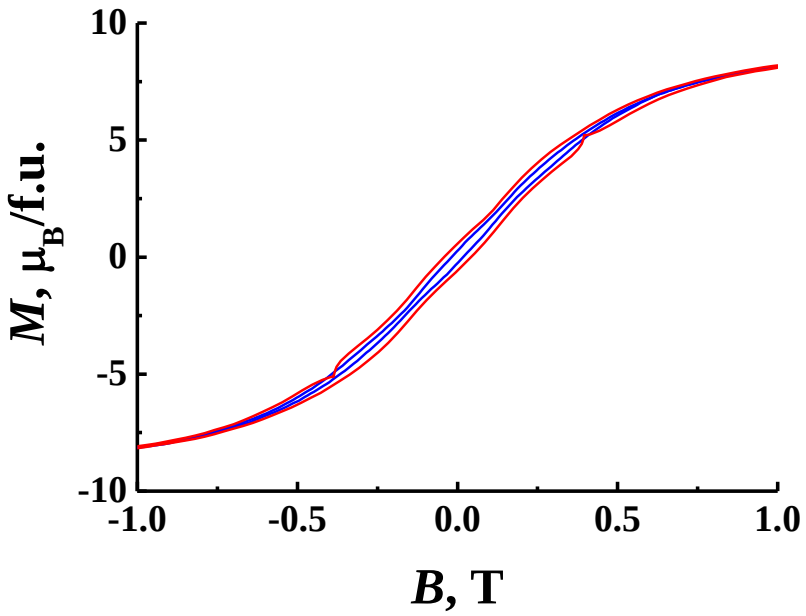
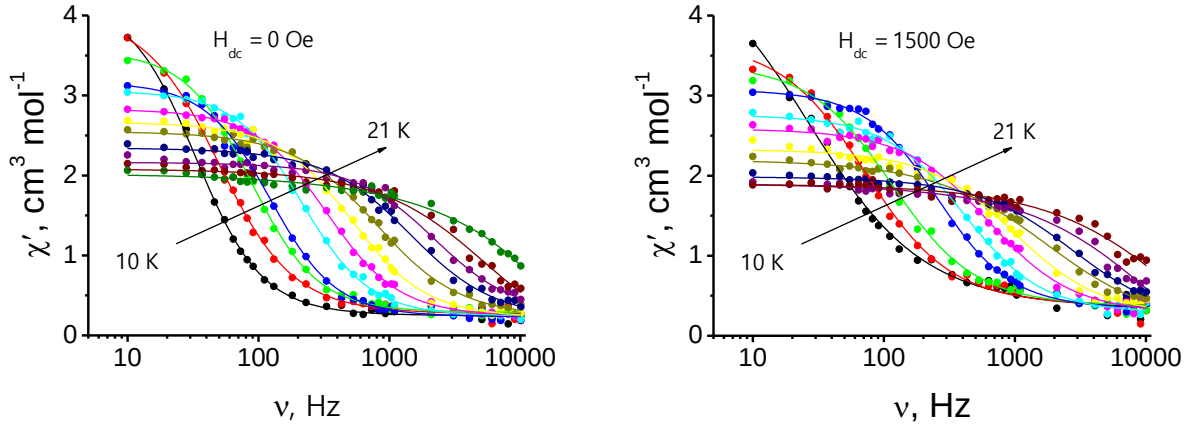


Figure S5. Magnetic hysteresis loops at 2 K with magnetic field sweep rates of 0.3 T/min (blue line) and 0.9 T/min (red line).



(a) (b)

Figure S6. Frequency dependences of the in-phase ac susceptibility signals for complex 1 at zero (a) and 1500 Oe (b) dc fields and temperatures from 10 to 21 K.

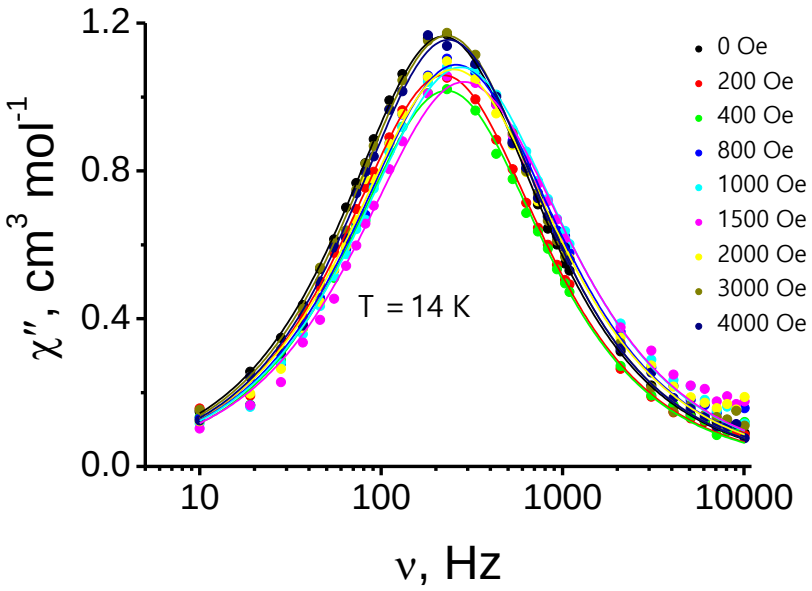


Figure S7. Frequency dependences of the out-of-phase ac susceptibility for **1** at temperature 14 K and applied dc fields from 0 to 4000 Oe.