Supporting Information for

Air-stable pentagonal-bipyramidal dysprosium(III) single-molecule magnets with sulfur-containing macrocycle equatorial ligand

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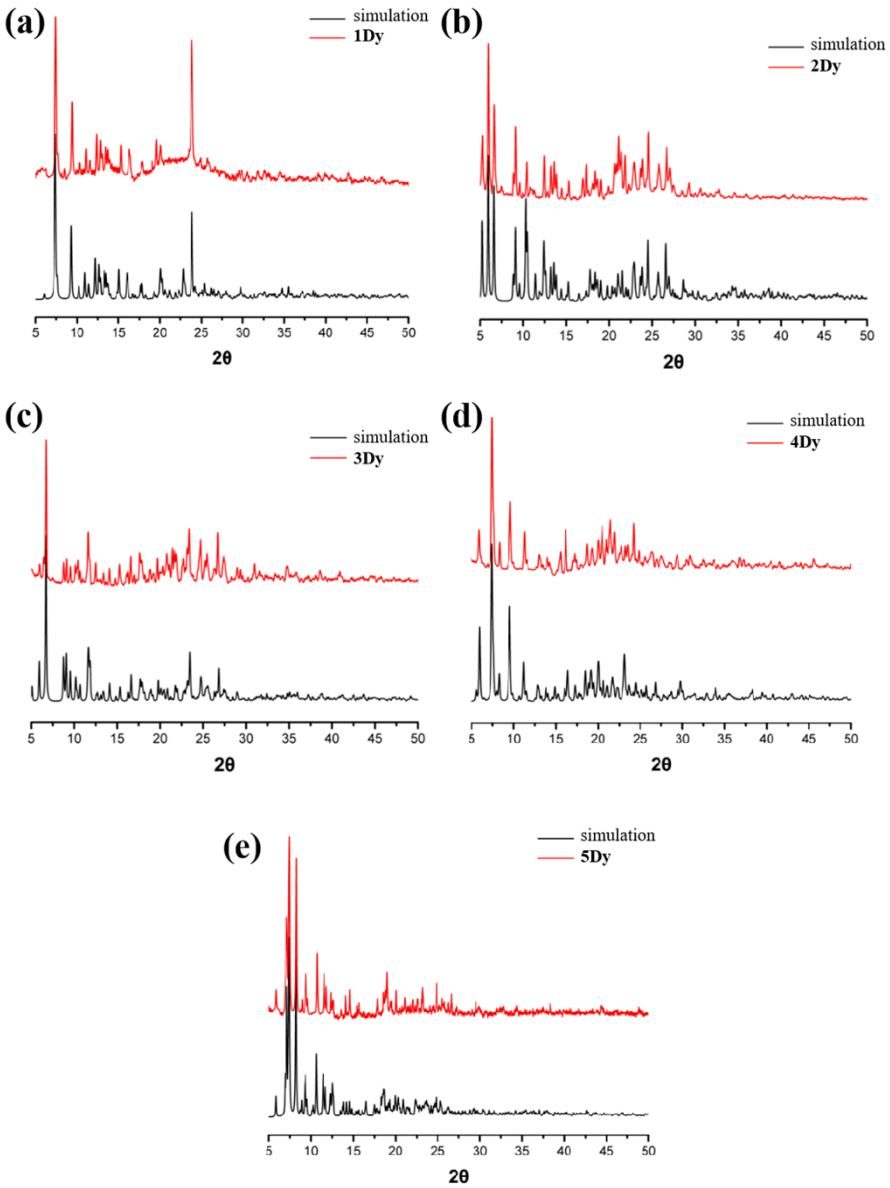

Figure S1 PXRD patterns of compounds **1Dy** to **5Dy**.

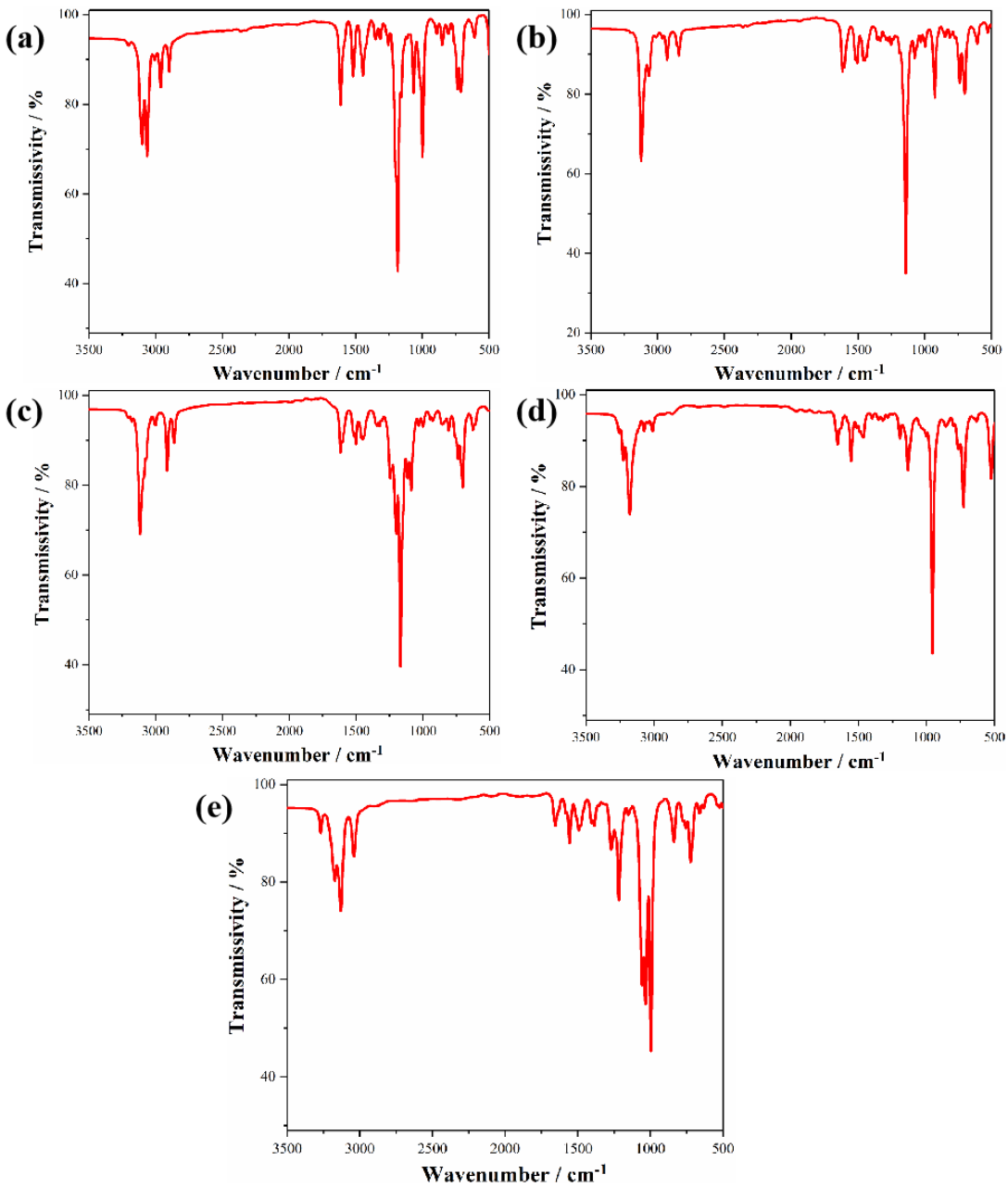

Figure S2 Infrared spectra of compounds **1Dy** to **5Dy**.

Table S1. Crystal data and structure refinement for **1Dy** to **5Dy**.

Compound	**1Dy**	**2Dy**	**3Dy**	**4Dy**	**5Dy**
Chemical formula	$C_{57}H_{59}BDyN_5O_3S_2$	$C_{65}H_{59}BDyN_5O_3S_2$	$C_{65}H_{53}BDyF_6N_5O_3S_2$	$C_{97}H_{101}BDyN_5O_6S_2Si_2$	$C_{69}H_{87}BDyN_5O_8S_2Si_2$
Formula Mass	1099.52	1195.60	1303.55	1726.48	1408.04
Temperature (K)	150	150	150	150	150
Crystal system	triclinic	triclinic	triclinic	orthorhombic	triclinic
Space group	*P*-1	*P*-1	*P*-1	$P2_12_12_1$	*P*-1
a (Å)	13.7632(15)	10.4761(13)	10.737(2)	15.382(2)	15.174(7)
b (Å)	14.1873(15)	16.413(2)	16.291(4)	18.785(3)	23.960(11)
c (Å)	16.1154(17)	17.823(2)	18.447(4)	29.655(4)	24.117(11)
α (°)	108.3830(10)	71.160(2)	71.730(3)	90	64.181(6)
β (°)	99.0650(10)	84.308(2)	77.574(3)	90	86.473(6)
γ (°)	112.6230(10)	72.163(2)	71.364(3)	90	89.051(6)
Unit cell volume (Å^3)	2613.0(5)	2760.9(6)	2879.4(11)	8569(2)	7877(6)
Z	2	2	2	4	4
ρ_{calc} (g/cm^3)	1.397	1.438	1.503	1.330	1.187
Absorption coefficient, μ(mm^{-1})	1.558	1.481	1.442	1.006	1.081
F(000)	1126.0	1222.0	1318.0	3544.0	2924.0
Reflections collected	25133	29593	27249	83933	75190
Independent reflections	9174	11358	10242	15841	27889
R_{int}	0.0316	0.0309	0.0560	0.0467	0.0497
GOF on F^2	1.056	1.041	1.007	1.081	1.020
R_1 (I>2σ/all data)	0.0398/0.0478	0.0315/0.0419	0.0432/0.0689	0.0402/0.0487	0.0430/0.0740
wR_2 (I>2σ/all data)	0.1016/0.1071	0.0720/0.0760	0.0996/0.1104	0.1007/0.1056	0.0971/0.1094

Table S2. Selected bond lengths (Å) and angles (°) for **1Dy**.

Dy(1)-O(1)	2.153(3)	O(1)-Dy(1)-N(1)	94.35(12)	O(2)-Dy(1)-N(4)	80.50(13)
Dy(1)-O(2)	2.080(4)	O(1)-Dy(1)-N(2)	95.37(12)	O(2)-Dy(1)-N(5)	89.73(14)
Dy(1)-N(1)	2.602(4)	O(1)-Dy(1)-N(3)	91.26(12)	N(1)-Dy(1)-N(2)	62.72(12)
Dy(1)-N(2)	2.462(4)	O(1)-Dy(1)-N(4)	85.57(12)	N(2)-Dy(1)-N(3)	63.35(12)
Dy(1)-N(3)	2.676(4)	O(1)-Dy(1)-N(5)	90.82(12)	N(3)-Dy(1)-N(4)	107.57(11)
Dy(1)-N(4)	2.646(4)	O(2)-Dy(1)-N(1)	100.01(14)	N(4)-Dy(1)-N(5)	63.44(12)
Dy(1)-N(5)	2.462(4)	O(2)-Dy(1)-N(2)	97.20(13)	N(5)-Dy(1)-N(1)	62.90(12)
O(1)-Dy(1)-O(2)	164.13(14)	O(2)-Dy(1)-N(3)	63.35(12)		

Table S3. Selected bond lengths (Å) and angles (°) for **2Dy**.

Dy(1)-O(1)	2.125(2)	O(1)-Dy(1)-N(1)	92.67(8)	O(2)-Dy(1)-N(4)	87.31(8)
Dy(1)-O(2)	2.081(2)	O(1)-Dy(1)-N(2)	84.51(8)	O(2)-Dy(1)-N(5)	93.06(8)
Dy(1)-N(1)	2.584(2)	O(1)-Dy(1)-N(3)	82.82(8)	N(1)-Dy(1)-N(2)	63.34(8)
Dy(1)-N(2)	2.442(2)	O(1)-Dy(1)-N(4)	92.09(8)	N(2)-Dy(1)-N(3)	64.29(8)
Dy(1)-N(3)	2.585(2)	O(1)-Dy(1)-N(5)	99.72(8)	N(3)-Dy(1)-N(4)	106.43(8)
Dy(1)-N(4)	2.654(2)	O(2)-Dy(1)-N(1)	99.52(8)	N(4)-Dy(1)-N(5)	63.17(8)
Dy(1)-N(5)	2.469(2)	O(2)-Dy(1)-N(2)	93.72(8)	N(5)-Dy(1)-N(1)	62.92(8)
O(1)-Dy(1)-O(2)	165.37(8)	O(2)-Dy(1)-N(3)	83.33(8)		

Table S4. Selected bond lengths (Å) and angles (°) for **3Dy**.

Dy(1)-O(1)	2.156(3)	O(1)-Dy(1)-N(1)	94.10(13)	O(2)-Dy(1)-N(4)	88.42(14)
Dy(1)-O(2)	2.098(3)	O(1)-Dy(1)-N(2)	86.94(14)	O(2)-Dy(1)-N(5)	94.25(14)
Dy(1)-N(1)	2.582(4)	O(1)-Dy(1)-N(3)	82.83(13)	N(1)-Dy(1)-N(2)	63.15(13)
Dy(1)-N(2)	2.433(4)	O(1)-Dy(1)-N(4)	87.29(13)	N(2)-Dy(1)-N(3)	64.20(13)
Dy(1)-N(3)	2.591(4)	O(1)-Dy(1)-N(5)	96.12(13)	N(3)-Dy(1)-N(4)	106.06(13)
Dy(1)-N(4)	2.626(4)	O(2)-Dy(1)-N(1)	99.54(13)	N(4)-Dy(1)-N(5)	63.28(13)
Dy(1)-N(5)	2.459(4)	O(2)-Dy(1)-N(2)	94.96(14)	N(5)-Dy(1)-N(1)	63.27(13)
O(1)-Dy(1)-O(2)	165.56(13)	O(2)-Dy(1)-N(3)	85.12(14)		

Table S5. Selected bond lengths (Å) and angles (°) for **4Dy**.

Dy(1)-O(1)	2.129(5)	O(1)-Dy(1)-N(1)	100.00(18)	O(2)-Dy(1)-N(4)	88.31(19)
Dy(1)-O(2)	2.126(5)	O(1)-Dy(1)-N(2)	88.10(19)	O(2)-Dy(1)-N(5)	90.52(19)
Dy(1)-N(1)	2.560(5)	O(1)-Dy(1)-N(3)	81.58(19)	N(1)-Dy(1)-N(2)	63.38(18)
Dy(1)-N(2)	2.429(5)	O(1)-Dy(1)-N(4)	80.77(19)	N(2)-Dy(1)-N(3)	63.89(19)
Dy(1)-N(3)	2.610(6)	O(1)-Dy(1)-N(5)	98.57(19)	N(3)-Dy(1)-N(4)	106.16(19)
Dy(1)-N(4)	2.596(6)	O(2)-Dy(1)-N(1)	99.32(19)	N(4)-Dy(1)-N(5)	63.15(19)
Dy(1)-N(5)	2.440(6)	O(2)-Dy(1)-N(2)	100.00(19)	N(5)-Dy(1)-N(1)	63.79(18)
O(1)-Dy(1)-O(2)	160.67(19)	O(2)-Dy(1)-N(3)	86.27(19)		

Table S6. Selected bond lengths (Å) and angles (°) for **5Dy**.

Dy(1)-O(1)	2.119(3)	O(1)-Dy(1)-N(1)	85.37(13)	O(2)-Dy(1)-N(4)	95.25(12)
Dy(1)-O(2)	2.116(3)	O(1)-Dy(1)-N(2)	91.80(12)	O(2)-Dy(1)-N(5)	88.41(12)
Dy(1)-N(1)	2.597(4)	O(1)-Dy(1)-N(3)	97.89(13)	N(1)-Dy(1)-N(2)	63.96(13)
Dy(1)-N(2)	2.452(4)	O(1)-Dy(1)-N(4)	93.66(13)	N(2)-Dy(1)-N(3)	62.85(12)
Dy(1)-N(3)	2.584(4)	O(1)-Dy(1)-N(5)	84.27(13)	N(3)-Dy(1)-N(4)	62.96(12)
Dy(1)-N(4)	2.442(4)	O(2)-Dy(1)-N(1)	83.74(13)	N(4)-Dy(1)-N(5)	64.19(13)
Dy(1)-N(5)	2.625(4)	O(2)-Dy(1)-N(2)	93.08(12)	N(5)-Dy(1)-N(1)	106.08(13)
O(1)-Dy(1)-O(2)	164.67(14)	O(2)-Dy(1)-N(3)	97.27(13)		
Dy(2)-O(3)	2.127(3)	O(3)-Dy(2)-N(6)	81.85(12)	O(2)-Dy(1)-N(9)	91.89(12)
Dy(2)-O(4)	2.125(3)	O(3)-Dy(2)-N(7)	95.84(11)	O(2)-Dy(1)-N(10)	83.13(12)
Dy(2)-N(6)	2.635(4)	O(3)-Dy(2)-N(8)	106.69(11)	N(6)-Dy(1)-N(7)	62.86(12)
Dy(2)-N(7)	2.475(4)	O(3)-Dy(2)-N(9)	102.47(12)	N(7)-Dy(1)-N(8)	62.59(12)
Dy(2)-N(8)	2.624(4)	O(3)-Dy(2)-N(10)	80.71(11)	N(8)-Dy(1)-N(9)	62.17(11)
Dy(2)-N(9)	2.467(4)	O(2)-Dy(2)-N(6)	80.52(12)	N(9)-Dy(1)-N(10)	63.52(11)
Dy(2)-N(10)	2.578(4)	O(2)-Dy(2)-N(7)	96.75(12)	N(10)-Dy(1)-N(6)	109.12(11)
O(3)-Dy(2)-O(4)	150.64(12)	O(2)-Dy(2)-N(8)	102.65(12)		

Table S7 Continuous Shape Measures (CShM) calculations for the potential coordination geometries of **1Dy**-**5Dy**.

Compound	HP-7	HPY-7	PBPY-7	COC-7	CTPR-7	JPBPY-7	JETPY-7
1Dy	30.723	14.599	4.574	8.220	7.468	5.234	19.142
2Dy	30.236	15.620	4.230	8.113	6.555	4.922	18.607
3Dy	31.546	15.891	4.038	8.149	6.880	4.966	18.801
4Dy	31.424	14.360	4.608	8.211	7.077	5.509	16.775
5Dy	32.254	15.498	4.163	8.003	7.418	5.002	19.321
	32.289	12.560	6.109	8.298	7.798	6.600	16.488

Note: HP-7 = Heptagon (D_{7h}); HPY-7 = Hexagonal pyramid (C_{6v}); PBPY-7 = Pentagonal bipyramid (D_{5h}); COC-7 = Capped octahedron (C_{3v}); CTPR-7 = Capped trigonal prism (C_{2v}); JPBPY-7 = Johnson pentagonal bipyramid J13 (D_{5h}); JETPY-7 = Johnson elongated triangular pyramid J7 (C_{3v}). Two sets of CShM values for **5Dy** refer to both diverse complexes in the unit cell, which correspond to Dy(1) and Dy(2) in Table S6 respectively.

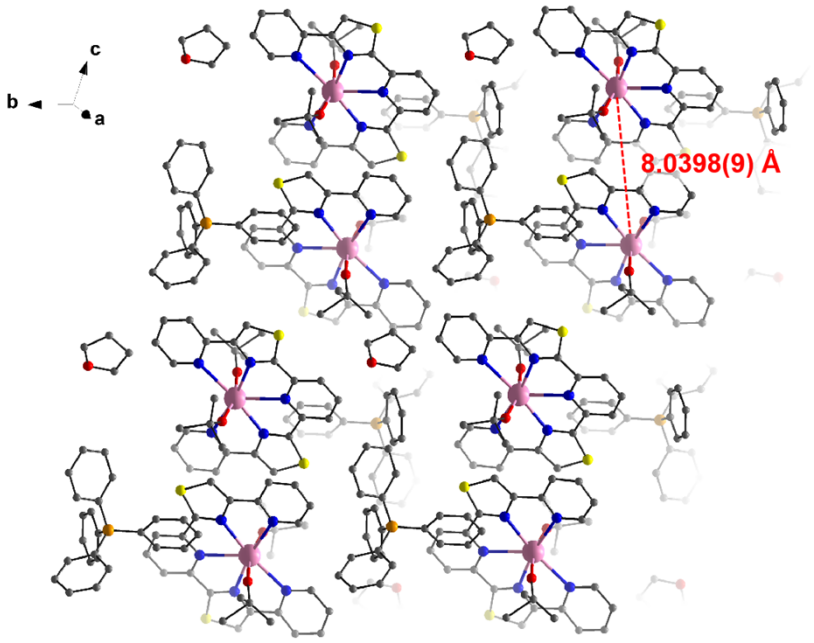

Figure S3 Packing arrangement of **1Dy**. The red dashed lines represent the nearest intermolecular Dy···Dy separation. For clarity, hydrogen atoms are omitted. Colour codes: Dy, pink; S, yellow; B, orange; C, gray; O, red; N, blue.

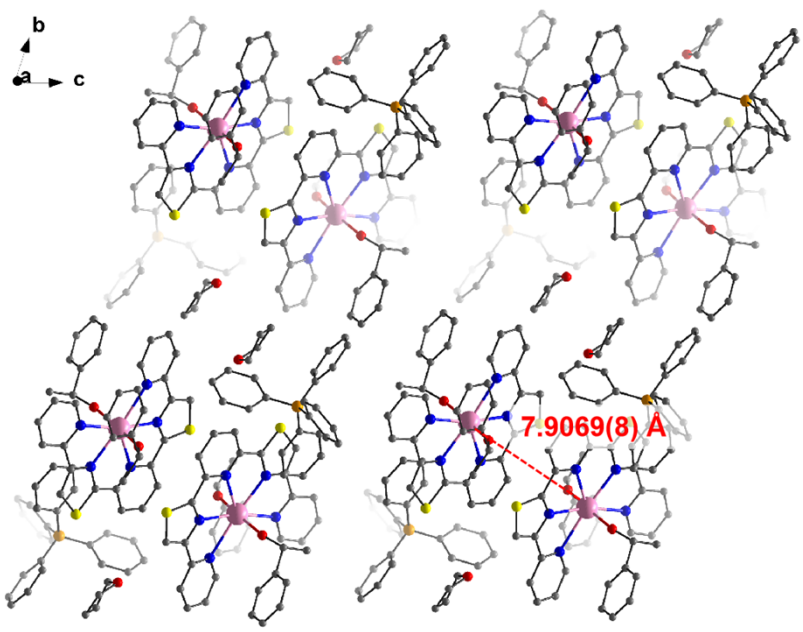

Figure S4 Packing arrangement of **2Dy**. The red dashed lines represent the nearest intermolecular Dy···Dy separation. For clarity, hydrogen atoms are omitted. Colour codes: Dy, pink; S, yellow; B, orange; C, gray; O, red; N, blue.

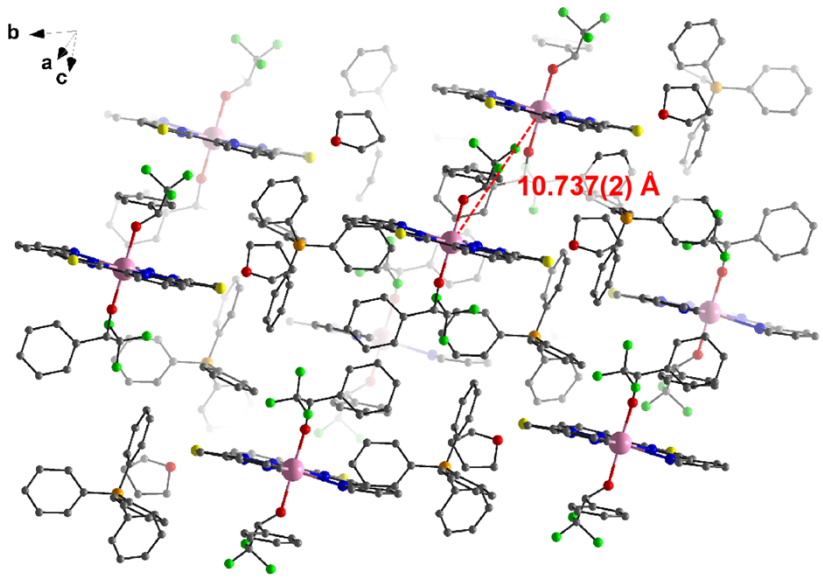

Figure S5 Packing arrangement of **3Dy**. The red dashed lines represent the nearest intermolecular Dy···Dy separation. For clarity, hydrogen atoms are omitted. Colour codes: Dy, pink; S, yellow; B, orange; C, gray; F, green; O, red; N, blue.

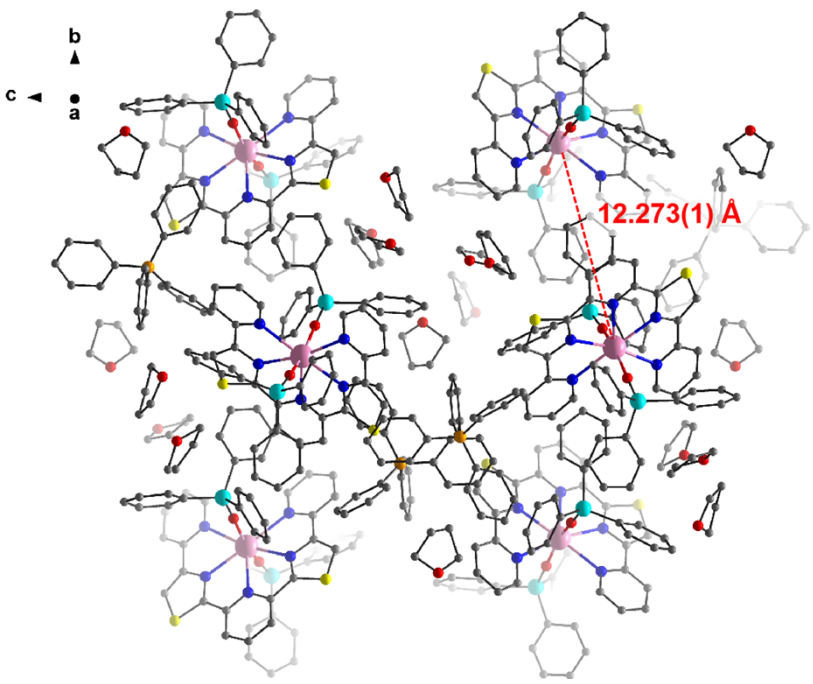

Figure S6 Packing arrangement of **4Dy**. The red dashed lines represent the nearest intermolecular Dy···Dy separation. For clarity, hydrogen atoms are omitted. Colour codes: Dy, pink; S, yellow; B, orange; C, gray; Si, turquoise; O, red; N, blue.

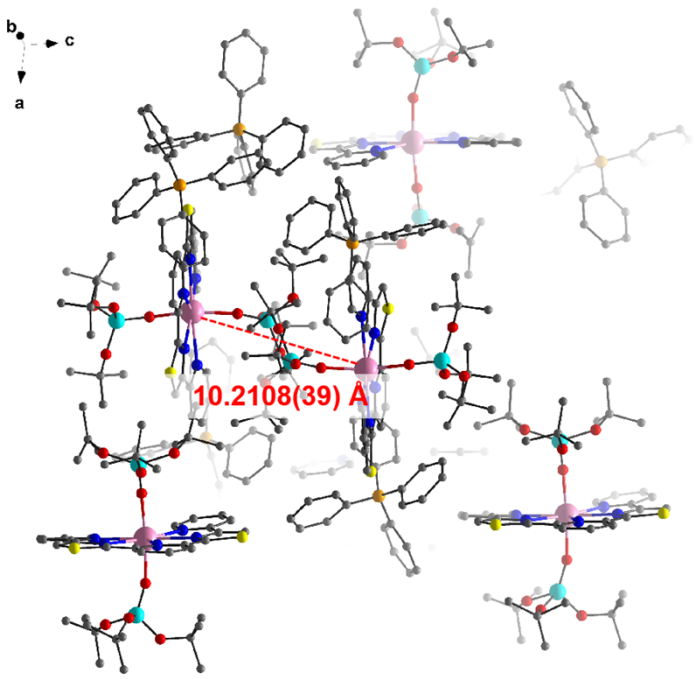

Figure S7 Packing arrangement of **5Dy**. The red dashed lines represent the nearest intermolecular Dy···Dy separation. For clarity, hydrogen atoms are omitted. Colour codes: Dy, pink; S, yellow; B, orange; C, gray; Si, turquoise; O, red; N, blue.

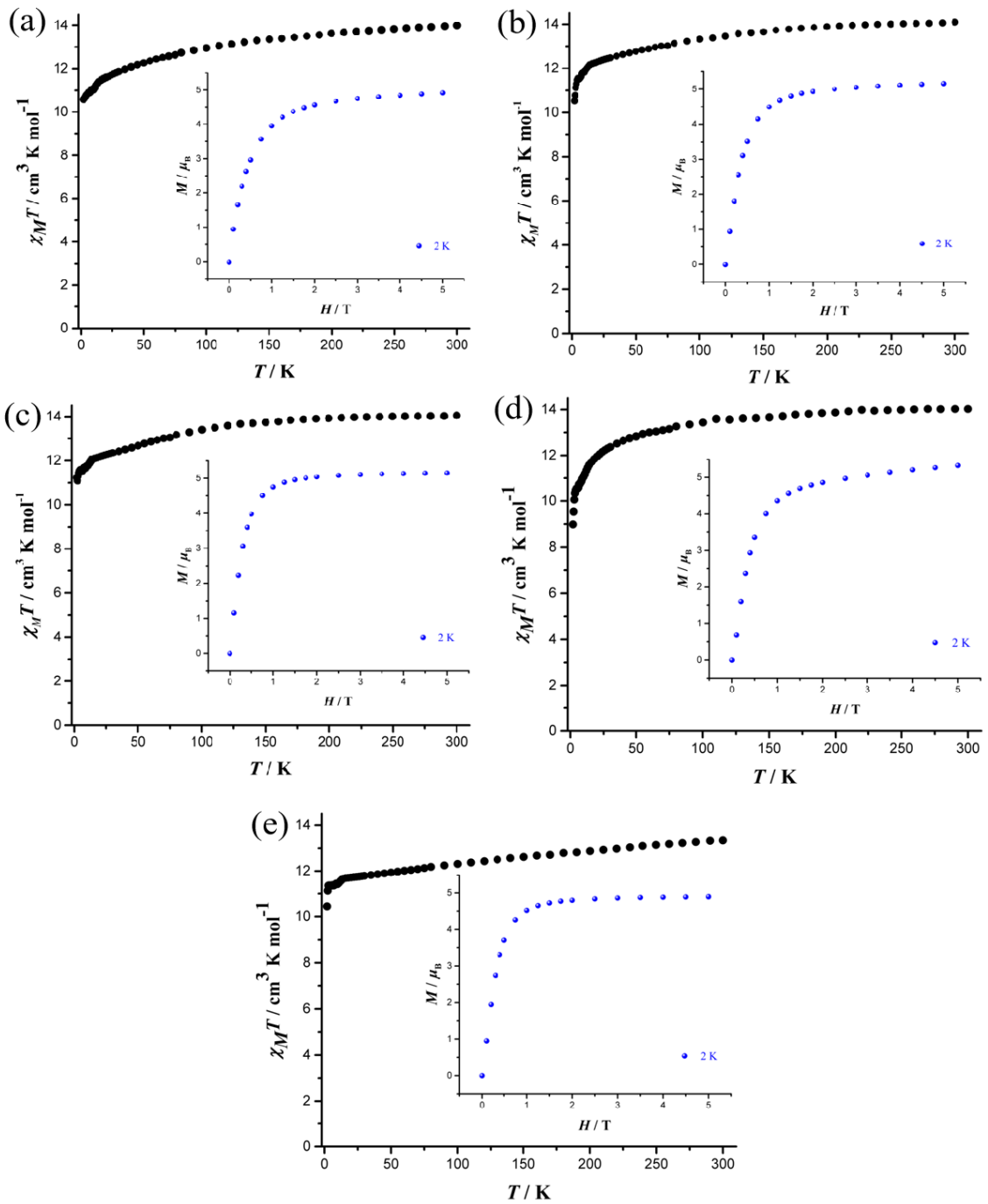

Figure S8 Temperature dependence of the product $\chi_M T$ with an applied field of 1 kOe and field dependent magnetization at 2 K up to 5 T (insert) for **1Dy** (a)**, 2Dy** (b), **3Dy** (c), **4Dy** (d) and **5Dy** (e), respectively.

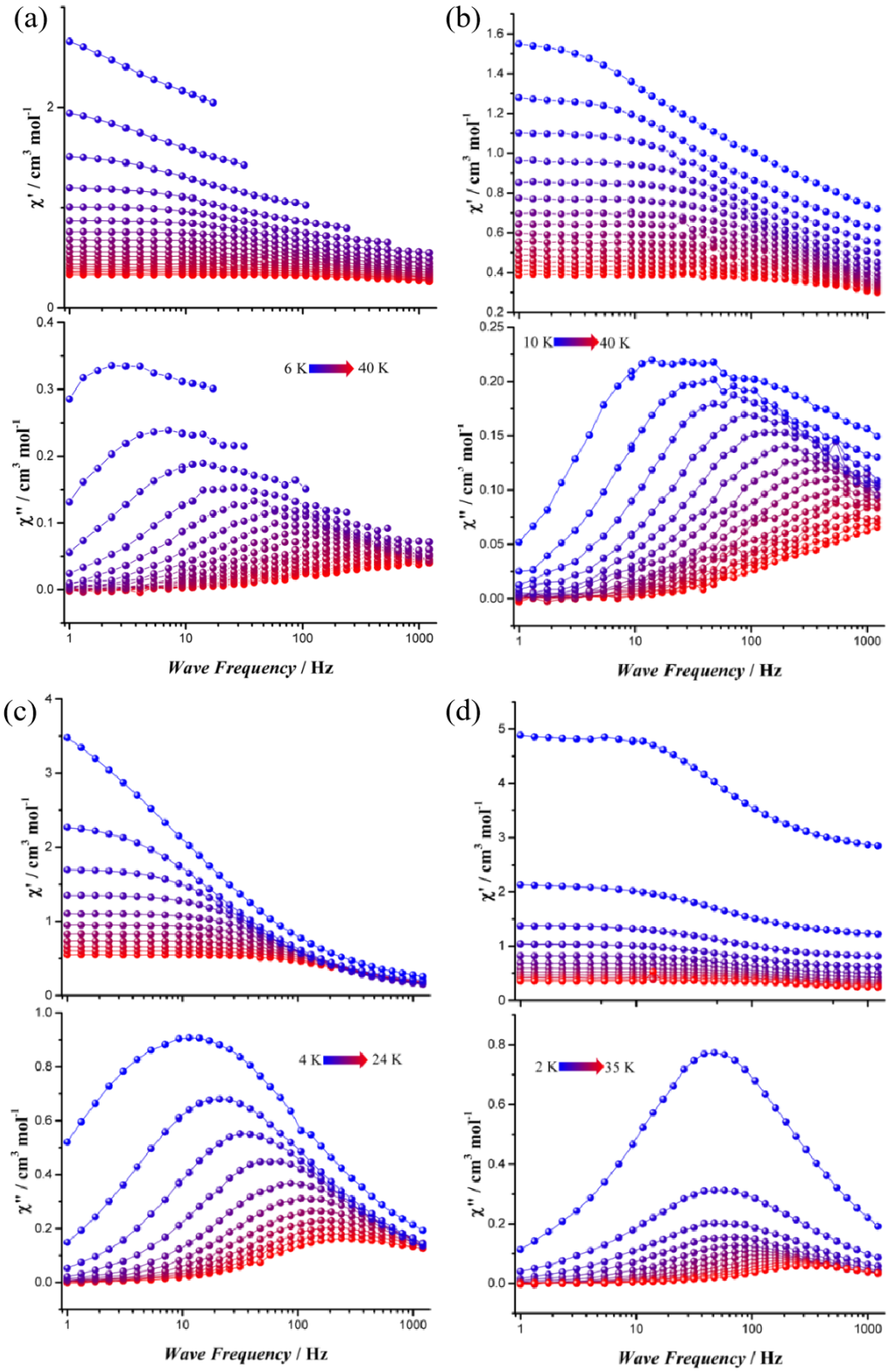

Figure S9 Frequency dependence of the in-phase χ' (top) and out-of-phase χ'' (bottom) components of the ac susceptibility for **1Dy** (a)**, 2Dy** (b), **3Dy** (c) and **4Dy** (d) measured under zero dc field. Solid lines represent the fits using the generalized Debye model.

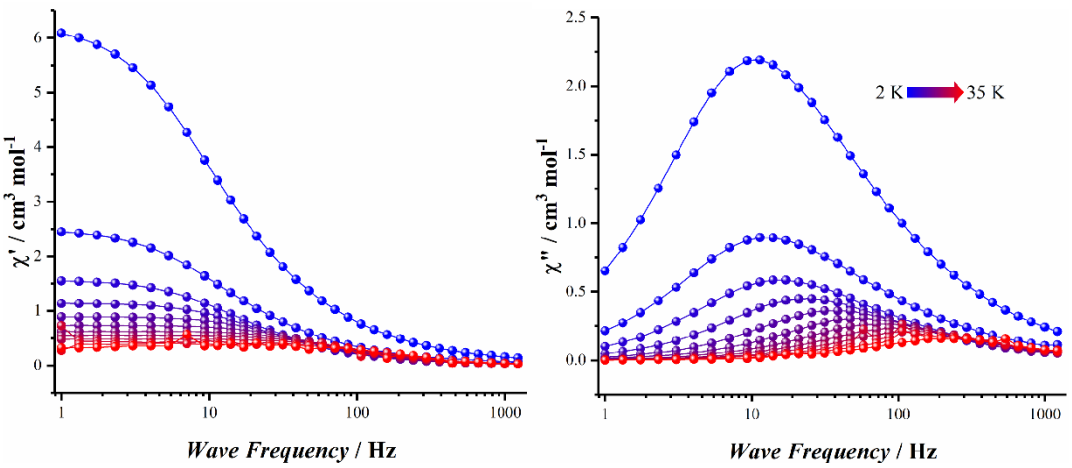

Figure S10 Frequency dependence of the in-phase χ' (left) and out-of-phase χ'' (right) components of the ac susceptibility for **5Dy** measured under zero dc field. Solid lines represent the fits using the generalized Debye model.

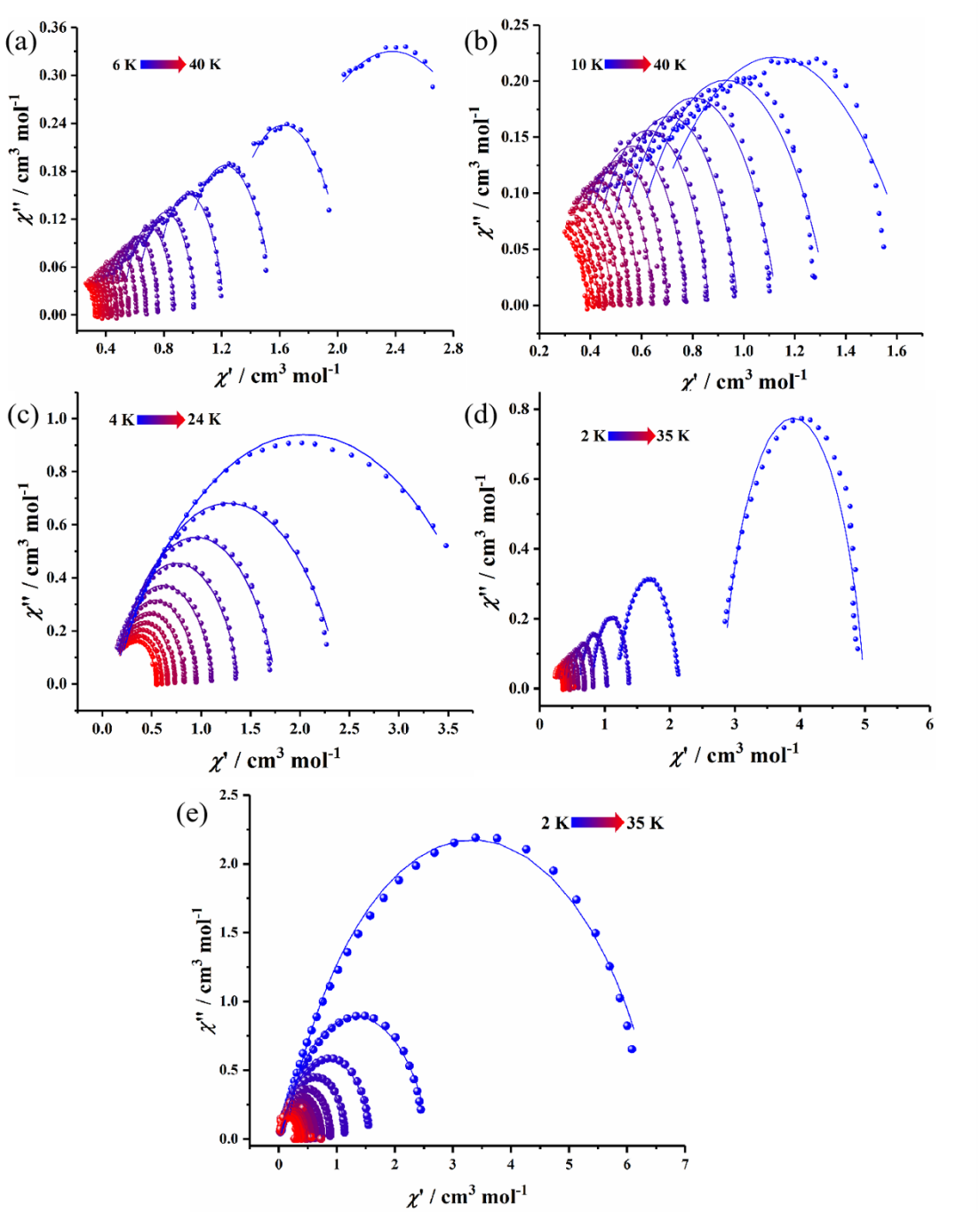

Figure S11 Cole-Cole plots for **1Dy** (a)**, 2Dy** (b), **3Dy** (c), **4Dy** (d) and **5Dy** (e) at zero dc field. The solid lines are the best fits obtained with a generalized Debye model.

Table S8 Relaxation fitting parameters obtained with a generalized Debye model for **1Dy**.

T / K	χ_S	χ_T	τ / s	α
6	1.42630E+00	3.32912E+00	4.50823E-02	5.74741E-01
8	1.15564E+00	2.09930E+00	1.90369E-02	4.05091E-01
10	8.74607E-01	1.57512E+00	8.59792E-03	3.74702E-01
12	7.05529E-01	1.22749E+00	4.60246E-03	3.30486E-01
14	5.94426E-01	1.03135E+00	2.87111E-03	3.21987E-01
16	5.10342E-01	8.85497E-01	1.85173E-03	3.24069E-01
18	4.62112E-01	7.69570E-01	1.42892E-03	2.74971E-01
20	4.20626E-01	6.84265E-01	1.12916E-03	2.53200E-01
22	3.82242E-01	6.15399E-01	8.65705E-04	2.41695E-01
24	3.51560E-01	5.60626E-01	6.87563E-04	2.47791E-01
26	3.27580E-01	5.14994E-01	5.80070E-04	2.28537E-01
28	3.02699E-01	4.76716E-01	4.58427E-04	2.36552E-01
30	2.86881E-01	4.42331E-01	4.01392E-04	2.19815E-01
32	2.65361E-01	4.14199E-01	3.26295E-04	2.45670E-01
34	2.52314E-01	3.89564E-01	2.80004E-04	2.50936E-01
36	2.39419E-01	3.67773E-01	2.48019E-04	2.50681E-01
38	2.20014E-01	3.47838E-01	1.87270E-04	2.80127E-01
40	2.10797E-01	3.30331E-01	1.71488E-04	2.84374E-01

Table S9 Relaxation fitting parameters obtained with a generalized Debye model for **2Dy**.

T / K	χ_S	χ_T	τ / s	α
10	5.48746E-01	1.69447E+00	3.36291E-03	5.30151E-01
12	5.25225E-01	1.33325E+00	2.24145E-03	4.12675E-01
14	4.77639E-01	1.13226E+00	1.60412E-03	3.44247E-01
16	4.35356E-01	9.78763E-01	1.18366E-03	2.92285E-01
18	3.87353E-01	8.66293E-01	8.67954E-04	2.63934E-01
20	3.47191E-01	7.78961E-01	6.52235E-04	2.58671E-01
22	3.21234E-01	7.04653E-01	5.21082E-04	2.43423E-01
24	2.92156E-01	6.49188E-01	4.11149E-04	2.48396E-01
26	2.67733E-01	5.97894E-01	3.31177E-04	2.52888E-01
28	2.49798E-01	5.56736E-01	2.72813E-04	2.58032E-01
30	2.38003E-01	5.19272E-01	2.34853E-04	2.55808E-01
32	2.18502E-01	4.87601E-01	1.92342E-04	2.72524E-01
34	2.07224E-01	4.59023E-01	1.61857E-04	2.73415E-01
36	2.00278E-01	4.35006E-01	1.41514E-04	2.85455E-01
38	1.91318E-01	4.13236E-01	1.23110E-04	2.98218E-01
40	1.90832E-01	3.91825E-01	1.14693E-04	2.85512E-01

Table S10 Relaxation fitting parameters obtained with a generalized Debye model for **3Dy**.

T / K	χ_S	χ_T	τ / s	α
4	8.70855E-02	4.02183E+00	1.44133E-02	4.32249E-01
6	1.40376E-01	2.41619E+00	6.64584E-03	3.12331E-01
8	1.34855E-01	1.75722E+00	3.78734E-03	2.38226E-01
10	1.18559E-01	1.38575E+00	2.37756E-03	2.06191E-01
12	1.00728E-01	1.12871E+00	1.57507E-03	2.01651E-01
14	9.25251E-02	9.68784E-01	1.17180E-03	2.03669E-01
16	9.08489E-02	8.46316E-01	9.18063E-04	2.07385E-01
18	9.02728E-02	7.49121E-01	7.49514E-04	2.10220E-01
20	9.72442E-02	6.72191E-01	6.46808E-04	2.02978E-01
22	1.02739E-01	6.11340E-01	5.75728E-04	1.99209E-01
24	1.09893E-01	5.57847E-01	5.24492E-04	1.88494E-01

Table S11 Relaxation fitting parameters obtained with a generalized Debye model for **4Dy**.

T / K	χ_S	χ_T	τ / s	α
2	2.80436E+00	4.99166E+00	2.78756E-03	2.15006E-01
5	1.18737E+00	2.14844E+00	2.86565E-03	2.62084E-01
8	7.89276E-01	1.38780E+00	2.58841E-03	2.38670E-01
11	6.49436E-01	1.01538E+00	2.40283E-03	1.69983E-05
14	5.22080E-01	8.13379E-01	1.95096E-03	2.44229E-05
17	4.21266E-01	6.93663E-01	1.47975E-03	1.32623E-01
20	3.66641E-01	5.96943E-01	1.18071E-03	1.09953E-01
23	3.27093E-01	5.24421E-01	9.73946E-04	8.42846E-02
26	2.94035E-01	4.68883E-01	7.90778E-04	7.31095E-02
29	2.67535E-01	4.23548E-01	6.60828E-04	5.27468E-02
32	2.44946E-01	3.86854E-01	5.68237E-04	5.19620E-02
35	2.26631E-01	3.64934E-01	5.15458E-04	7.21730E-02

Table S12 Relaxation fitting parameters obtained with a generalized Debye model for **5Dy**.

T / K	χ_S	χ_T	τ / s	α
2	1.12006E-01	6.57645E+00	1.31088E-02	2.45858E-01
5	5.42612E-02	2.60376E+00	1.09236E-02	2.23848E-01
8	4.26272E-02	1.62148E+00	8.78822E-03	1.90236E-01
11	3.38753E-02	1.17379E+00	6.04060E-03	1.48259E-01
14	2.49396E-02	9.08192E-01	3.91177E-03	1.20856E-01
17	2.15343E-02	7.45393E-01	2.71392E-03	9.97197E-02
20	1.75171E-02	6.31839E-01	2.00176E-03	8.72382E-02
23	1.53796E-02	5.50263E-01	1.54786E-03	7.88501E-02
26	1.25817E-02	4.86482E-01	1.22956E-03	7.28056E-02
29	1.98852E-14	4.60692E-01	1.11776E-03	1.06487E-01
32	3.87315E-15	3.91410E-01	8.71604E-04	7.97316E-02
35	4.99384E-15	3.71408E-01	7.57481E-04	8.73723E-02

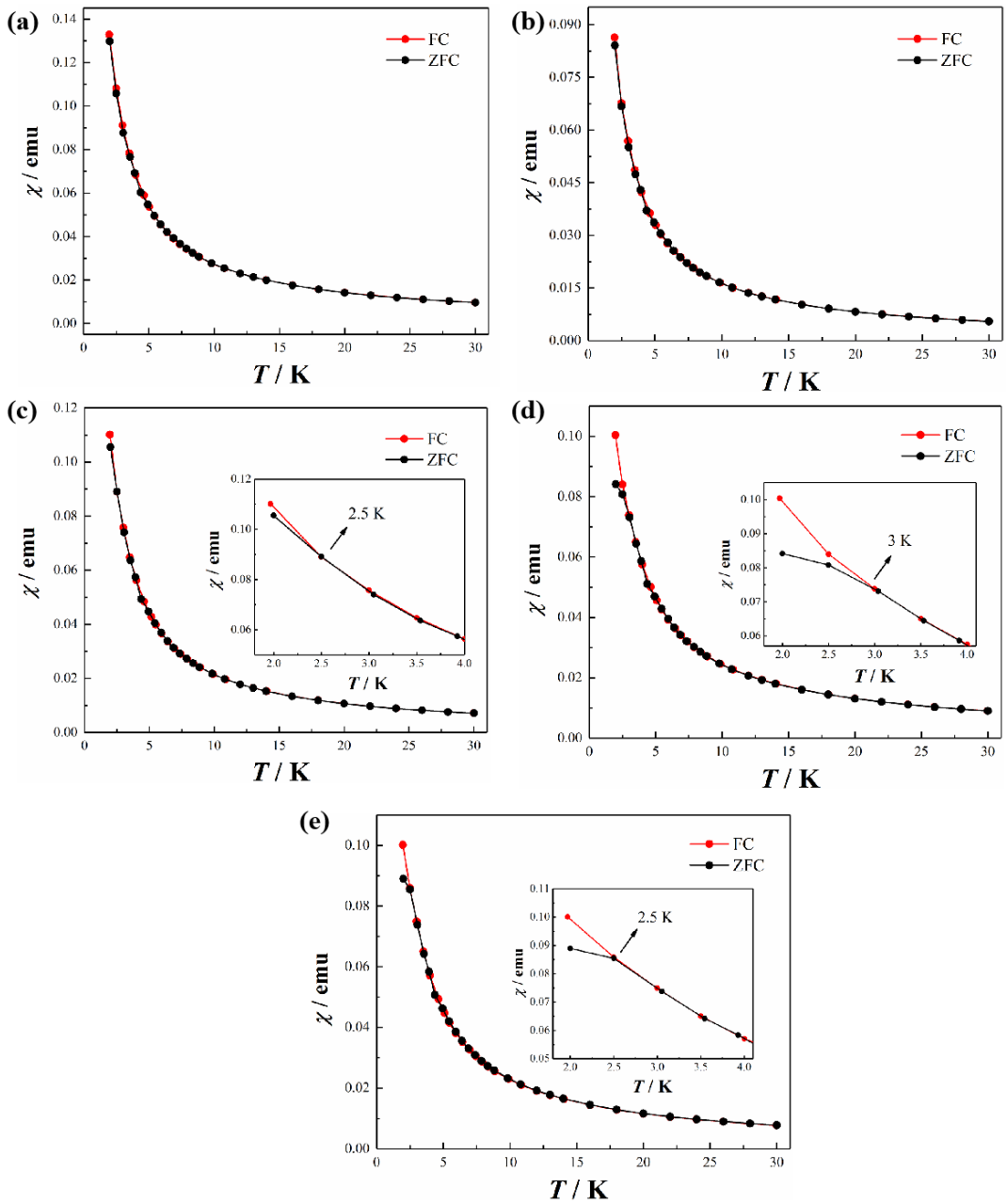

Figure S12 Field-cooled (FC) and zero-field-cooled (ZFC) magnetic susceptibilities for **1Dy** (a), **2Dy** (b), **3Dy** (c), **4Dy** (d) and **5Dy** (e) under a dc magnetic field of 2000 Oe.

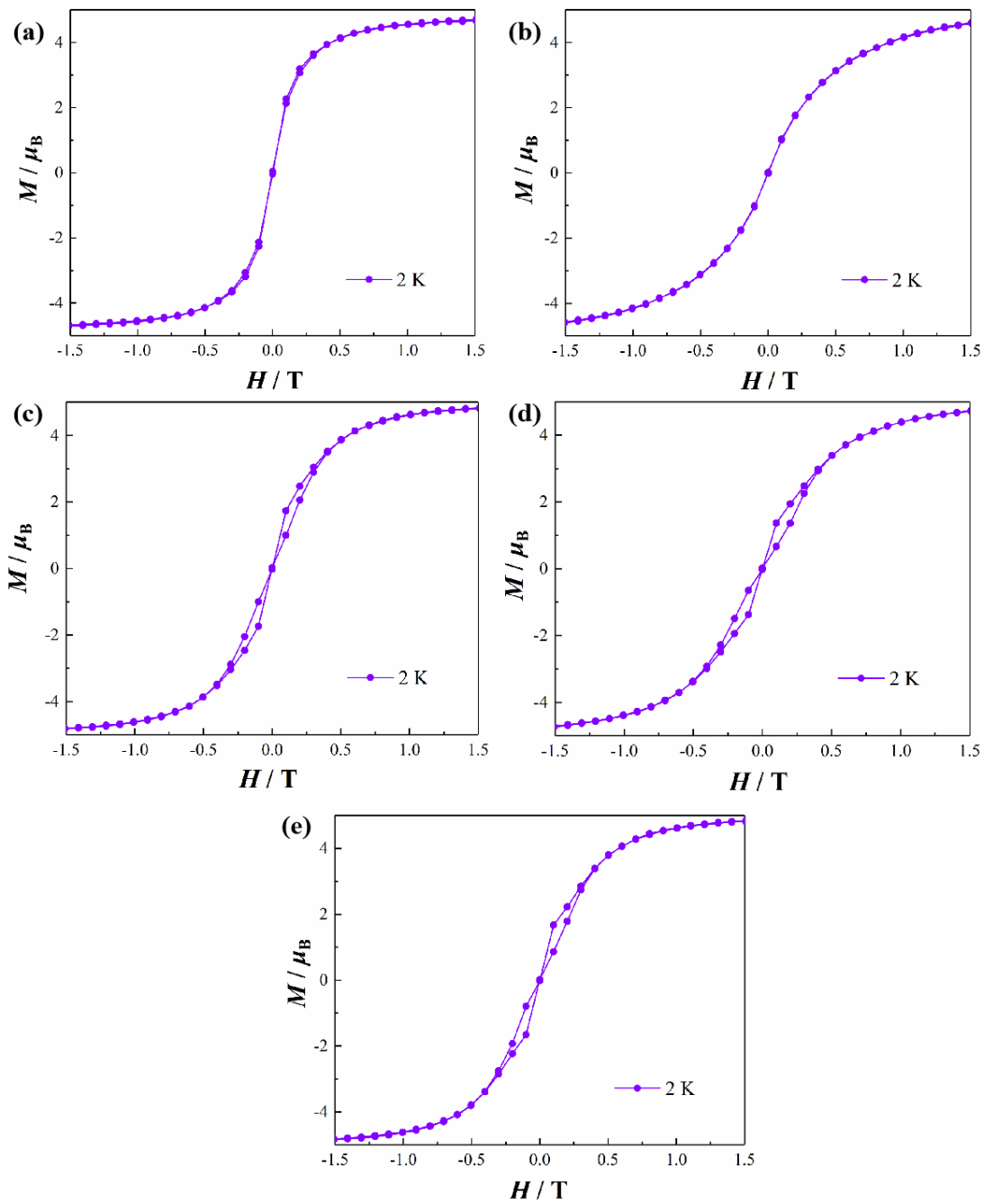

Figure S13 Magnetic hysteresis loop for **1Dy** (a), **2Dy** (b), **3Dy** (c), **4Dy** (d) and **5Dy** (e) at 2 K with an average scan rate of 30 Oe s^{-1}.

Table S13 SA-CASSCF/RASSI calculated electronic states for **1Dy**.

Ab initio **Energy (cm $^{-1}$)**	***Ab initio*** **Energy (K)**	g_x	g_y	g_z	g_z **Angle (°)**	**Crystal field Wavefunction**
0	0	0.001	0.001	19.882	-	99.9%| ± 15/2>
576	828	0.046	0.056	16.920	1.13	99.7%| ± 13/2>
982	1412	0.237	0.403	13.863	2.50	96.2%| ± 11/2>
1177	1692	4.474	5.249	11.800	89.45	32.0%| ± 9/2>+10.9%| ± 5/2>+11.7 %| ∓ 3/2>+15.9%| ∓ 9/2>
1285	1847	3.764	5.211	7.390	85.59	10.1%| ± 7/2>+13.6%| ± 3/2>+25.2 %| ∓ 1/2>+43.2%| ∓ 9/2>
1366	1964	3.639	5.420	9.429	89.76	52.0%| ± 7/2>+29.1%| ∓ 1/2>
1418	2039	0.651	1.610	16.342	88.21	46.2%| ± 5/2>+35.3%| ∓ 3/2>+11.0 %| ∓ 7/2>
1665	2394	0.012	0.020	19.701	88.79	23.0%| ± 5/2>+30.1%| ± 1/2>+27.8 %| ∓ 3/2>+13.1%| ∓ 7/2>

Only components with > 10% contribution are given, rounded to the nearest percent.

Table S14 SA-CASSCF/RASSI calculated electronic states for **2Dy**.

Ab initio **Energy (cm $^{-1}$)**	***Ab initio*** **Energy (K)**	g_x	g_y	g_z	g_z **Angle (°)**	**Crystal field Wavefunction**
0	0	0.001	0.001	19.880	-	99.8%| ± 15/2>
581	835	0.045	0.059	16.914	2.92	99.3%| ± 13/2>
978	1406	0.380	0.619	13.794	9.63	92.6%| ± 11/2>
1159	1666	4.272	4.460	12.225	79.66	32.7%| ± 9/2>+15.6%| ± 5/2>+15. 7%| ∓ 3/2>
1272	1829	1.979	4.891	9.299	84.69	33.2%| ± 9/2>+16.6%| ± 1/2>+11. 4%| ∓ 1/2>+12.4%| ∓ 9/2>
1349	1939	2.784	6.140	10.526	89.59	44.1%| ± 7/2>+20.8%| ∓ 1/2>+13. 2%| ∓ 5/2>
1419	2040	0.124	0.184	17.314	89.69	37.6%| ± 5/2>+27.3%| ∓ 3/2>+17. 4%| ∓ 7/2>
1672	2404	0.017	0.027	19.668	88.60	22.1%| ± 5/2>+27.9%| ± 1/2>+26. 8%| ∓ 3/2>+13.8%| ∓ 7/2>

Only components with > 10% contribution are given, rounded to the nearest percent.

Table S15 SA-CASSCF/RASSI calculated electronic states for **3Dy**.

Ab initio Energy (cm^{-1})	*Ab initio* Energy (K)	g_x	g_y	g_z	g_z Angle (°)	Crystal field Wavefunction
0	0	0.002	0.002	19.876	-	99.8%| ± 15/2>
507	729	0.073	0.094	16.941	3.27	99.3%| ± 13/2>
856	1231	0.244	0.866	13.350	12.77	60.9%| ± 11/2>+29.3%| ∓ 11/2>
971	1396	0.912	2.775	15.398	85.55	11.2%| ± 9/2>+11.3%| ± 5/2>+12.7%| ± 3/2>+13.4%| ± 1/2>+17.3%| ∓ 3/2>
1084	1558	8.336	6.992	1.077	77.35	61.2%| ± 9/2>+29.1%| ± 1/2>
1148	1650	2.295	4.867	13.215	87.24	29.8%| ± 7/2>+16.2%| ± 3/2>+14.5%| ∓ 5/2>
1187	1706	0.649	1.370	16.644	85.86	34.4%| ± 5/2>+17.6%| ∓ 3/2>+29.4%| ∓ 7/2>
1435	2063	0.057	0.080	19.524	88.74	24.2%| ± 5/2>+25.0%| ± 1/2>+24.7%| ∓ 3/2>+16.4%| ∓ 7/2>

Only components with > 10% contribution are given, rounded to the nearest percent.

Table S16 SA-CASSCF/RASSI calculated electronic states for **4Dy**.

Ab initio Energy (cm^{-1})	*Ab initio* Energy (K)	g_x	g_y	g_z	g_z Angle (°)	Crystal field Wavefunction
0	0	0.002	0.002	19.876	-	99.9%| ± 15/2>
531	763	0.069	0.089	16.922	1.28	99.5%| ± 13/2>
921	1324	0.509	1.248	13.318	6.42	87.4%| ± 11/2>
1064	1530	1.478	3.553	14.722	85.10	15.9%| ± 9/2>+13.5%| ± 5/2>+16.0%| ± 1/2>+23.1%| ∓ 3/2>
1195	1718	8.047	6.377	0.938	84.19	33.9%| ± 9/2>+15.0%| ± 1/2>+13.7%| ∓ 1/2>+29.4%| ∓ 9/2>
1264	1817	2.763	6.213	12.398	88.99	41.3%| ± 7/2>+19.8%| ± 3/2>+18.0%| ∓ 1/2>+16.1%| ∓ 5/2>
1321	1899	0.577	1.398	16.756	89.13	37.8%| ± 5/2>+20.2%| ∓ 3/2>+28.9%| ∓ 7/2>
1581	2273	0.064	0.079	19.486	89.59	24.7%| ± 5/2>+24.1%| ± 1/2>+24.9%| ∓ 3/2>+17.0%| ∓ 7/2>

Only components with > 10% contribution are given, rounded to the nearest percent.

Table S17 SA-CASSCF/RASSI calculated electronic states for **5Dy-1**.

Ab initio Energy (cm^{-1})	*Ab initio* Energy (K)	g_x	g_y	g_z	g_z Angle (°)	Crystal field Wavefunction
0	0	0.001	0.002	19.879	-	100%|±15/2>
560	805	0.053	0.066	16.917	0.65	99.3%|±13/2>
998	1435	0.300	0.672	13.597	1.98	94.4%|±11/2>
1185	1704	2.024	3.213	15.387	88.90	17.9%|±9/2>+14.2%|±5/2>+ 17.1%|±1/2>+21.3%|∓3/2>
1302	1872	9.067	6.580	0.420	86.58	31.1%|±9/2>+14.2%|±1/2>+ 17.2%|∓1/2>+26.3%|∓9/2>
1383	1988	2.125	5.149	13.052	89.62	33.5%|±7/2>+18.4%|±3/2>+ 15.1%|∓1/2>+12.3%|∓5/2>
1450	2085	0.640	1.517	16.873	89.96	35.2%|±5/2>+18.4%|∓3/2>+ 29.5%|∓7/2>
1691	2431	0.131	0.141	19.308	88.61	26.7%|±5/2>+21.5%|±1/2>+ 23.8%|∓3/2>+19.4%|∓7/2>

Only components with > 10% contribution are given, rounded to the nearest percent.

Table S18 SA-CASSCF/RASSI calculated electronic states for **5Dy-2**.

Ab initio Energy (cm^{-1})	*Ab initio* Energy (K)	g_x	g_y	g_z	g_z Angle (°)	Crystal field Wavefunction
0	0	0.002	0.003	19.874	-	99.8%|±15/2>
478	687	0.080	0.103	16.940	1.79	99.3%|±13/2>
816	1173	0.536	1.488	13.136	6.67	89.1%|±11/2>
922	1325	0.849	3.327	15.067	85.81	12.3%|±9/2>+12.5%|±5/2>+1 6.6%|±1/2>+22.5%|∓3/2>
1044	1501	8.138	6.876	0.149	72.35	63.0%|±9/2>+22.1%|±1/2>
1111	1597	2.988	6.624	10.985	89.76	11.4%|±7/2>+15.3%|±3/2>+1 1.6%|±1/2>+16.1%|∓5/2>+32. 0%|∓7/2>
1162	1670	0.235	0.669	16.646	83.67	35.4%|±5/2>+21.2%|∓3/2>+1 8.5%|∓7/2>
1425	2049	0.035	0.044	19.592	87.17	22.7%|±5/2>+24.8%|±1/2>+2 2.4%|∓3/2>+13.3%|∓7/2>

Only components with > 10% contribution are given, rounded to the nearest percent.

Table S19 *Ab initio* calculated crystal field parameters for Dy(III) ion in **1Dy**.

Crystal Field Parameter B_k^q		**Value / cm^{-1}**
k	q	
2	-2	4.6620677005176E-02
2	-1	-2.3361979946498E-02
2	0	-8.2481059744994E+00
2	1	-2.0027888993166E-01
2	2	3.2186009055183E+00
4	-4	3.3085849128835E-03
4	-3	1.4036866224199E-03
4	-2	-3.3817755692414E-03
4	-1	-1.3016027068508E-03
4	0	-1.2737168663571E-02
4	1	3.6534191145802E-03
4	2	-1.1495237255478E-02
4	3	2.5637381339191E-03
4	4	1.4431008318631E-02
6	-6	2.3653847850098E-04
6	-5	9.5506649993301E-05
6	-4	2.4600431351059E-05
6	-3	-1.3252716263402E-05
6	-2	4.4714814963711E-05
6	-1	-3.9963227397008E-05
6	0	1.8014285265054E-05
6	1	3.7734505478509E-06
6	2	-3.4012965962612E-05
6	3	1.9497448143109E-05
6	4	9.0643823138061E-05
6	5	1.1054745699851E-04
6	6	5.8482241587784E-04

Table S20 *Ab initio* calculated crystal field parameters for Dy(III) ion in **2Dy**.

Crystal Field Parameter B_k^q		**Value / cm^{-1}**
k	q	
2	-2	6.4356857675733E-02
2	-1	-3.0537949362411E-01
2	0	-8.1355830410379E+00
2	1	1.8031779485620E-01
2	2	3.5915941900407E+00
4	-4	5.6019101243105E-03
4	-3	1.4230553431214E-03
4	-2	-3.4606425839751E-03
4	-1	8.3233429683023E-03
4	0	-1.3045823675671E-02
4	1	-2.6633978352190E-03
4	2	-9.6940383285547E-03
4	3	4.8080764683993E-03
4	4	1.4860026888366E-02
6	-6	3.9808953168030E-04
6	-5	-3.6086645685270E-04
6	-4	3.7279358897288E-05
6	-3	1.3947599534307E-06
6	-2	3.0350512743697E-05
6	-1	8.3101106657047E-05
6	0	1.6950012488346E-05
6	1	3.7048574042991E-05
6	2	-4.8180267127492E-05
6	3	1.0592974380977E-05
6	4	8.9230732600843E-05
6	5	1.3293444480626E-04
6	6	5.0819824065758E-04

Table S21 *Ab initio* calculated crystal field parameters for Dy(III) ion in **3Dy**.

Crystal Field Parameter B_k^q		**Value / cm^{-1}**
k	q	
2	-2	4.7373043683041E-02
2	-1	2.7257804757000E-01
2	0	-6.7112404492439E+00
2	1	-1.4973536626038E-01
2	2	3.1240473457342E+00
4	-4	3.4708060314549E-03
4	-3	-3.6986415812722E-04
4	-2	-2.5571610076168E-03
4	-1	-7.1725295301111E-03
4	0	-1.3004631711568E-02
4	1	3.5960702074648E-03
4	2	-1.3419399019280E-02
4	3	4.2585750514633E-04
4	4	1.6258246505976E-02
6	-6	2.5341890624327E-04
6	-5	3.5966670354779E-04
6	-4	2.8201934074169E-05
6	-3	-1.0982049017024E-05
6	-2	4.0268321374175E-05
6	-1	-9.3254895765430E-05
6	0	2.2741182535657E-05
6	1	-2.7306214768466E-05
6	2	1.1266231239867E-06
6	3	-2.2955224135342E-05
6	4	9.8245758671465E-05
6	5	3.6288617126914E-05
6	6	5.9884597428077E-04

Table S22 *Ab initio* calculated crystal field parameters for Dy(III) ion in **4Dy**.

Crystal Field Parameter B_k^q		**Value / cm^{-1}**
k	q	
2	-2	-1.7743199785102E-02
2	-1	2.8463934245465E-01
2	0	-7.5209216708090E+00
2	1	4.9201401184416E-02
2	2	3.6076099599012E+00
4	-4	-3.1901231853271E-03
4	-3	-1.7784630057028E-03
4	-2	6.3017746205170E-04
4	-1	-6.0723391702676E-03
4	0	-1.3637891387170E-02
4	1	-1.4731202756930E-03
4	2	-1.2272197958430E-02
4	3	-4.5054575421793E-03
4	4	1.5109368700128E-02
6	-6	-1.5501599191762E-04
6	-5	2.3919120899272E-04
6	-4	-1.7375830551732E-05
6	-3	-6.1085745353623E-06
6	-2	2.4408200928710E-07
6	-1	-1.1004648954647E-05
6	0	3.2556731815086E-05
6	1	-5.4183925098563E-06
6	2	-3.2645118284637E-05
6	3	-2.7716855857223E-05
6	4	9.3812474611214E-05
6	5	4.5152480661534E-05
6	6	6.7755278189766E-04

Table S23 *Ab initio* calculated crystal field parameters for Dy(III) ion in **5Dy-1**.

Crystal Field Parameter B_k^q		**Value / cm^{-1}**
k	q	
2	-2	-5.8499299182348E-02
2	-1	1.0407234952738E-01
2	0	-8.2456732475606E+00
2	1	-4.1722392538986E-02
2	2	3.5635968917130E+00
4	-4	-6.1123961277988E-03
4	-3	-4.9980068447594E-04
4	-2	1.9927516725302E-03
4	-1	-1.7503938349341E-03
4	0	-1.4969641991355E-02
4	1	3.5067755272825E-03
4	2	-1.0203198770836E-02
4	3	4.6266532149306E-03
4	4	1.4981028469320E-02
6	-6	-3.4391438719356E-04
6	-5	4.5203112990471E-05
6	-4	-3.9643171747695E-05
6	-3	-5.4836887761843E-08
6	-2	-1.1957631551092E-05
6	-1	-1.5765338250783E-05
6	0	4.5860706157264E-05
6	1	-2.1167204419227E-05
6	2	-6.2967490989270E-05
6	3	3.8408127171451E-05
6	4	9.7177457907388E-05
6	5	1.0926146083568E-04
6	6	5.8807917478033E-04

Table S24 *Ab initio* calculated crystal field parameters for Dy(III) ion in **5Dy-2**.

Crystal Field Parameter B_k^q		**Value / cm^{-1}**
k	q	
2	-2	9.1376484262735E-03
2	-1	1.4880592918488E-01
2	0	-6.6137795042511E+00
2	1	2.3051328215063E-01
2	2	3.3807581676403E+00
4	-4	-4.5203013523336E-03
4	-3	2.8188672260513E-03
4	-2	3.3128446962853E-04
4	-1	-5.7772077381156E-03
4	0	-1.1875613296339E-02
4	1	-9.8175428529672E-03
4	2	-1.5551269704463E-02
4	3	-1.7054353274672E-02
4	4	1.6149353561323E-02
6	-6	-2.0374602995645E-04
6	-5	2.5669193871025E-04
6	-4	-4.4383475147162E-05
6	-3	1.3758581841988E-05
6	-2	-2.1219740379605E-05
6	-1	9.4068894051747E-07
6	0	2.2865780549029E-05
6	1	1.7354321163453E-05
6	2	3.6514105558555E-06
6	3	-9.6956155414787E-05
6	4	9.9359428330312E-05
6	5	-2.0323488158797E-04
6	6	6.4317479925746E-04

Table S25 Average transition magnetic moment elements between the states in **1Dy**, given in μ_B.

Matrix Element < Mult. *i* | Mult. *j* >	**Average Value**
< 1.1+ | 1.1- >	3.41E-04
< 2.1+ | 2.1- >	1.70E-02
< 3.1+ | 3.1- >	1.07E-01
< 4.1+ | 4.1- >	2.72E+00
< 5.1+ | 5.1- >	2.09E+00
< 6.1+ | 6.1- >	2.48E+00
< 7.1+ | 7.1- >	2.73E+00
< 8.1+ | 8.1- >	1.02E-01
< 1.1+ | 2.1+ >	1.78E+00
< 1.1+ | 2.1- >	1.01E-03
< 2.1+ | 3.1+ >	2.38E+00
< 2.1+ | 3.1- >	2.41E-02
< 3.1+ | 4.1+ >	2.33E+00
< 3.1+ | 4.1- >	2.92E-01
< 4.1+ | 5.1+ >	1.20E+00
< 4.1+ | 5.1- >	1.76E+00
< 5.1+ | 6.1+ >	2.84E+00
< 5.1+ | 6.1- >	6.92E-01
< 6.1+ | 7.1+ >	2.30E+00
< 6.1+ | 7.1- >	1.24E+00
< 7.1+ | 8.1+ >	1.27E+00
< 7.1+ | 8.1- >	5.85E-01
< 1.1+ | 3.1+ >	4.55E-02
< 1.1+ | 3.1- >	7.49E-03
< 2.1+ | 4.1+ >	8.31E-02
< 2.1+ | 4.1- >	2.14E-01
< 3.1+ | 5.1+ >	1.58E+00
< 3.1+ | 5.1- >	2.61E-01
< 4.1+ | 6.1+ >	8.83E-01
< 4.1+ | 6.1- >	6.63E-01
< 5.1+ | 7.1+ >	4.89E-01
< 5.1+ | 7.1- >	5.75E-01
< 6.1+ | 8.1+ >	5.11E-01
< 6.1+ | 8.1- >	5.18E-01
< 1.1+ | 4.1+ >	5.31E-02
< 1.1+ | 4.1- >	1.16E-02
< 2.1+ | 5.1+ >	7.07E-02
< 2.1+ | 5.1- >	1.93E-01
< 3.1+ | 6.1+ >	2.25E-01
< 3.1+ | 6.1- >	2.44E-01
< 4.1+ | 7.1+ >	3.43E-01
< 4.1+ | 7.1- >	5.75E-01
< 5.1+ | 8.1+ >	5.53E-01
< 5.1+ | 8.1- >	6.00E-01
< 1.1+ | 5.1+ >	3.93E-02
< 1.1+ | 5.1- >	1.15E-02
< 2.1+ | 6.1+ >	1.07E-01
< 2.1+ | 6.1- >	5.72E-02
< 3.1+ | 7.1+ >	2.73E-01
< 3.1+ | 7.1- >	1.96E-01
< 4.1+ | 8.1+ >	1.31E-01
< 4.1+ | 8.1- >	1.15E-01
< 1.1+ | 6.1+ >	1.75E-02
< 1.1+ | 6.1- >	2.49E-02
< 2.1+ | 7.1+ >	8.75E-02
< 2.1+ | 7.1- >	1.28E-01
< 3.1+ | 8.1+ >	2.34E-01
< 3.1+ | 8.1- >	2.24E-01
< 1.1+ | 7.1+ >	3.23E-02
< 1.1+ | 7.1- >	3.44E-02
< 2.1+ | 8.1+ >	1.02E-01
< 2.1+ | 8.1- >	9.70E-02
< 1.1+ | 8.1+ >	2.96E-02
< 1.1+ | 8.1- >	2.98E-02

Table S26 Average transition magnetic moment elements between the states in **2Dy**, given in μ_B.

Matrix Element < Mult. *i* | Mult. *j* >	**Average Value**
< 1.1+ | 1.1- >	4.42E-04
< 2.1+ | 2.1- >	1.74E-02
< 3.1+ | 3.1- >	1.68E-01
< 4.1+ | 4.1- >	2.60E+00
< 5.1+ | 5.1- >	2.21E+00
< 6.1+ | 6.1- >	2.82E+00
< 7.1+ | 7.1- >	3.17E+00
< 8.1+ | 8.1- >	1.56E-01
< 1.1+ | 2.1+ >	1.78E+00
< 1.1+ | 2.1- >	1.44E-03
< 2.1+ | 3.1+ >	2.38E+00
< 2.1+ | 3.1- >	2.51E-02
< 3.1+ | 4.1+ >	2.49E+00
< 3.1+ | 4.1- >	3.30E-01
< 4.1+ | 5.1+ >	2.01E+00
< 4.1+ | 5.1- >	1.71E+00
< 5.1+ | 6.1+ >	2.96E+00
< 5.1+ | 6.1- >	9.07E-01
< 6.1+ | 7.1+ >	2.08E+00
< 6.1+ | 7.1- >	1.17E+00
< 7.1+ | 8.1+ >	1.26E+00
< 7.1+ | 8.1- >	6.31E-01
< 1.1+ | 3.1+ >	1.23E-01
< 1.1+ | 3.1- >	8.13E-03
< 2.1+ | 4.1+ >	3.35E-01
< 2.1+ | 4.1- >	2.43E-01
< 3.1+ | 5.1+ >	1.59E+00
< 3.1+ | 5.1- >	3.32E-01
< 4.1+ | 6.1+ >	9.73E-01
< 4.1+ | 6.1- >	8.35E-01
< 5.1+ | 7.1+ >	4.88E-01
< 5.1+ | 7.1- >	6.58E-01
< 6.1+ | 8.1+ >	6.91E-01
< 6.1+ | 8.1- >	6.08E-01
< 1.1+ | 4.1+ >	4.94E-02
< 1.1+ | 4.1- >	2.04E-02
< 2.1+ | 5.1+ >	9.92E-02
< 2.1+ | 5.1- >	1.71E-01
< 3.1+ | 6.1+ >	2.53E-01
< 3.1+ | 6.1- >	4.15E-01
< 4.1+ | 7.1+ >	2.75E-01
< 4.1+ | 7.1- >	5.95E-01
< 5.1+ | 8.1+ >	4.73E-01
< 5.1+ | 8.1- >	5.20E-01
< 1.1+ | 5.1+ >	3.41E-02
< 1.1+ | 5.1- >	1.50E-02
< 2.1+ | 6.1+ >	1.41E-01
< 2.1+ | 6.1- >	7.72E-02
< 3.1+ | 7.1+ >	3.24E-01
< 3.1+ | 7.1- >	2.29E-01
< 4.1+ | 8.1+ >	1.69E-01
< 4.1+ | 8.1- >	9.94E-02
< 1.1+ | 6.1+ >	2.69E-02
< 1.1+ | 6.1- >	3.48E-02
< 2.1+ | 7.1+ >	1.01E-01
< 2.1+ | 7.1- >	1.31E-01
< 3.1+ | 8.1+ >	2.46E-01
< 3.1+ | 8.1- >	2.09E-01
< 1.1+ | 7.1+ >	3.67E-02
< 1.1+ | 7.1- >	4.14E-02
< 2.1+ | 8.1+ >	1.09E-01
< 2.1+ | 8.1- >	1.01E-01
< 1.1+ | 8.1+ >	3.11E-02
< 1.1+ | 8.1- >	3.18E-02

Table S27 Average transition magnetic moment elements between the states in **3Dy**, given in μ_B.

Matrix Element < Mult. *i* | Mult. *j* >	**Average Value**
< 1.1+ | 1.1- >	6.73E-04
< 2.1+ | 2.1- >	2.79E-02
< 3.1+ | 3.1- >	1.96E-01
< 4.1+ | 4.1- >	2.04E+00
< 5.1+ | 5.1- >	1.67E+00
< 6.1+ | 6.1- >	3.01E+00
< 7.1+ | 7.1- >	1.72E+00
< 8.1+ | 8.1- >	5.38E-01
< 1.1+ | 2.1+ >	1.77E+00
< 1.1+ | 2.1- >	2.38E-03
< 2.1+ | 3.1+ >	2.35E+00
< 2.1+ | 3.1- >	4.79E-02
< 3.1+ | 4.1+ >	2.25E+00
< 3.1+ | 4.1- >	7.53E-01
< 4.1+ | 5.1+ >	1.86E+00
< 4.1+ | 5.1- >	1.34E+00
< 5.1+ | 6.1+ >	2.66E+00
< 5.1+ | 6.1- >	9.96E-01
< 6.1+ | 7.1+ >	1.67E+00
< 6.1+ | 7.1- >	1.29E+00
< 7.1+ | 8.1+ >	1.49E+00
< 7.1+ | 8.1- >	4.42E-01
< 1.1+ | 3.1+ >	1.32E-01
< 1.1+ | 3.1- >	1.49E-02
< 2.1+ | 4.1+ >	5.17E-01
< 2.1+ | 4.1- >	3.00E-01
< 3.1+ | 5.1+ >	2.07E+00
< 3.1+ | 5.1- >	3.28E-01
< 4.1+ | 6.1+ >	8.61E-01
< 4.1+ | 6.1- >	1.21E+00
< 5.1+ | 7.1+ >	1.39E+00
< 5.1+ | 7.1- >	7.98E-01
< 6.1+ | 8.1+ >	5.95E-01
< 6.1+ | 8.1- >	5.48E-01
< 1.1+ | 4.1+ >	2.95E-02
< 1.1+ | 4.1- >	3.56E-02
< 2.1+ | 5.1+ >	1.69E-01
< 2.1+ | 5.1- >	1.53E-01
< 3.1+ | 6.1+ >	4.09E-01
< 3.1+ | 6.1- >	5.27E-01
< 4.1+ | 7.1+ >	5.77E-01
< 4.1+ | 7.1- >	6.56E-01
< 5.1+ | 8.1+ >	4.60E-01
< 5.1+ | 8.1- >	5.19E-01
< 1.1+ | 5.1+ >	4.56E-02
< 1.1+ | 5.1- >	2.03E-02
< 2.1+ | 6.1+ >	1.26E-01
< 2.1+ | 6.1- >	8.31E-02
< 3.1+ | 7.1+ >	3.31E-01
< 3.1+ | 7.1- >	5.68E-01
< 4.1+ | 8.1+ >	1.33E-01
< 4.1+ | 8.1- >	8.74E-02
< 1.1+ | 6.1+ >	2.46E-02
< 1.1+ | 6.1- >	3.34E-02
< 2.1+ | 7.1+ >	1.47E-01
< 2.1+ | 7.1- >	1.59E-01
< 3.1+ | 8.1+ >	2.62E-01
< 3.1+ | 8.1- >	2.59E-01
< 1.1+ | 7.1+ >	4.65E-02
< 1.1+ | 7.1- >	5.17E-02
< 2.1+ | 8.1+ >	1.23E-01
< 2.1+ | 8.1- >	1.21E-01
< 1.1+ | 8.1+ >	3.91E-02
< 1.1+ | 8.1- >	3.92E-02

Table S28 Average transition magnetic moment elements between the states in **4Dy**, given in μ_B.

Matrix Element < Mult. *i* | Mult. *j* >	**Average Value**
< 1.1+ | 1.1- >	6.17E-04
< 2.1+ | 2.1- >	2.64E-02
< 3.1+ | 3.1- >	2.95E-01
< 4.1+ | 4.1- >	2.49E+00
< 5.1+ | 5.1- >	1.62E+00
< 6.1+ | 6.1- >	3.11E+00
< 7.1+ | 7.1- >	3.16E+00
< 8.1+ | 8.1- >	1.67E+00
< 1.1+ | 2.1+ >	1.77E+00
< 1.1+ | 2.1- >	1.70E-03
< 2.1+ | 3.1+ >	2.35E+00
< 2.1+ | 3.1- >	4.48E-02
< 3.1+ | 4.1+ >	2.21E+00
< 3.1+ | 4.1- >	8.47E-01
< 4.1+ | 5.1+ >	1.72E+00
< 4.1+ | 5.1- >	1.73E+00
< 5.1+ | 6.1+ >	2.87E+00
< 5.1+ | 6.1- >	5.66E-01
< 6.1+ | 7.1+ >	1.38E+00
< 6.1+ | 7.1- >	1.46E+00
< 7.1+ | 8.1+ >	1.30E+00
< 7.1+ | 8.1- >	7.69E-01
< 1.1+ | 3.1+ >	5.52E-02
< 1.1+ | 3.1- >	1.54E-02
< 2.1+ | 4.1+ >	3.55E-01
< 2.1+ | 4.1- >	3.78E-01
< 3.1+ | 5.1+ >	1.94E+00
< 3.1+ | 5.1- >	2.18E-01
< 4.1+ | 6.1+ >	6.95E-01
< 4.1+ | 6.1- >	9.20E-01
< 5.1+ | 7.1+ >	1.21E+00
< 5.1+ | 7.1- >	6.56E-01
< 6.1+ | 8.1+ >	4.89E-01
< 6.1+ | 8.1- >	4.98E-01
< 1.1+ | 4.1+ >	5.57E-02
< 1.1+ | 4.1- >	3.09E-02
< 2.1+ | 5.1+ >	1.04E-01
< 2.1+ | 5.1- >	1.52E-01
< 3.1+ | 6.1+ >	4.12E-01
< 3.1+ | 6.1- >	3.46E-01
< 4.1+ | 7.1+ >	6.26E-01
< 4.1+ | 7.1- >	4.74E-01
< 5.1+ | 8.1+ >	5.57E-01
< 5.1+ | 8.1- >	4.81E-01
< 1.1+ | 5.1+ >	3.50E-02
< 1.1+ | 5.1- >	1.30E-02
< 2.1+ | 6.1+ >	8.91E-02
< 2.1+ | 6.1- >	4.71E-02
< 3.1+ | 7.1+ >	2.47E-01
< 3.1+ | 7.1- >	5.14E-01
< 4.1+ | 8.1+ >	7.86E-02
< 4.1+ | 8.1- >	5.04E-02
< 1.1+ | 6.1+ >	1.10E-02
< 1.1+ | 6.1- >	2.66E-02
< 2.1+ | 7.1+ >	1.38E-01
< 2.1+ | 7.1- >	1.08E-01
< 3.1+ | 8.1+ >	2.43E-01
< 3.1+ | 8.1- >	2.71E-01
< 1.1+ | 7.1+ >	3.67E-02
< 1.1+ | 7.1- >	4.47E-02
< 2.1+ | 8.1+ >	1.22E-01
< 2.1+ | 8.1- >	1.06E-01
< 1.1+ | 8.1+ >	3.45E-02
< 1.1+ | 8.1- >	3.66E-02

Table S29 Average transition magnetic moment elements between the states in **5Dy-1**, given in μ_B.

Matrix Element < Mult. *i* | Mult. *j* >	**Average Value**
< 1.1+ | 1.1- >	4.70E-04
< 2.1+ | 2.1- >	1.98E-02
< 3.1+ | 3.1- >	1.62E-01
< 4.1+ | 4.1- >	3.19E+00
< 5.1+ | 5.1- >	2.10E+00
< 6.1+ | 6.1- >	3.12E+00
< 7.1+ | 7.1- >	3.70E+00
< 8.1+ | 8.1- >	1.02E+00
< 1.1+ | 2.1+ >	1.78E+00
< 1.1+ | 2.1- >	1.23E-03
< 2.1+ | 3.1+ >	2.37E+00
< 2.1+ | 3.1- >	3.04E-02
< 3.1+ | 4.1+ >	1.94E+00
< 3.1+ | 4.1- >	4.25E-01
< 4.1+ | 5.1+ >	1.01E+00
< 4.1+ | 5.1- >	1.63E+00
< 5.1+ | 6.1+ >	2.65E+00
< 5.1+ | 6.1- >	5.67E-01
< 6.1+ | 7.1+ >	7.80E-01
< 6.1+ | 7.1- >	1.53E+00
< 7.1+ | 8.1+ >	1.13E+00
< 7.1+ | 8.1- >	9.92E-01
< 1.1+ | 3.1+ >	2.97E-02
< 1.1+ | 3.1- >	1.01E-02
< 2.1+ | 4.1+ >	1.22E-01
< 2.1+ | 4.1- >	3.12E-01
< 3.1+ | 5.1+ >	1.98E+00
< 3.1+ | 5.1- >	2.20E-01
< 4.1+ | 6.1+ >	7.64E-01
< 4.1+ | 6.1- >	8.72E-01
< 5.1+ | 7.1+ >	1.16E+00
< 5.1+ | 7.1- >	4.13E-01
< 6.1+ | 8.1+ >	6.72E-01
< 6.1+ | 8.1- >	7.48E-01
< 1.1+ | 4.1+ >	5.07E-02
< 1.1+ | 4.1- >	1.91E-02
< 2.1+ | 5.1+ >	8.35E-02
< 2.1+ | 5.1- >	1.22E-01
< 3.1+ | 6.1+ >	2.97E-01
< 3.1+ | 6.1- >	5.49E-01
< 4.1+ | 7.1+ >	8.45E-01
< 4.1+ | 7.1- >	2.24E-01
< 5.1+ | 8.1+ >	5.46E-01
< 5.1+ | 8.1- >	4.92E-01
< 1.1+ | 5.1+ >	3.53E-02
< 1.1+ | 5.1- >	1.11E-02
< 2.1+ | 6.1+ >	1.15E-01
< 2.1+ | 6.1- >	4.28E-02
< 3.1+ | 7.1+ >	1.51E-01
< 3.1+ | 7.1- >	5.28E-01
< 4.1+ | 8.1+ >	2.96E-02
< 4.1+ | 8.1- >	5.83E-02
< 1.1+ | 6.1+ >	1.16E-02
< 1.1+ | 6.1- >	3.07E-02
< 2.1+ | 7.1+ >	1.31E-01
< 2.1+ | 7.1- >	7.68E-02
< 3.1+ | 8.1+ >	2.85E-01
< 3.1+ | 8.1- >	2.89E-01
< 1.1+ | 7.1+ >	2.90E-02
< 1.1+ | 7.1- >	3.68E-02
< 2.1+ | 8.1+ >	1.22E-01
< 2.1+ | 8.1- >	1.08E-01
< 1.1+ | 8.1+ >	3.42E-02
< 1.1+ | 8.1- >	3.56E-02

Table S30 Average transition magnetic moment elements between the states in **5Dy-2**, given in μ_B.

Matrix Element < Mult. *i* | Mult. *j* >	**Average Value**
< 1.1+ | 1.1- >	7.78E-04
< 2.1+ | 2.1- >	3.04E-02
< 3.1+ | 3.1- >	3.39E-01
< 4.1+ | 4.1- >	2.13E+00
< 5.1+ | 5.1- >	1.61E+00
< 6.1+ | 6.1- >	2.99E+00
< 7.1+ | 7.1- >	5.39E-01
< 8.1+ | 8.1- >	1.46E-01
< 1.1+ | 2.1+ >	1.77E+00
< 1.1+ | 2.1- >	2.35E-03
< 2.1+ | 3.1+ >	2.33E+00
< 2.1+ | 3.1- >	5.32E-02
< 3.1+ | 4.1+ >	2.15E+00
< 3.1+ | 4.1- >	1.10E+00
< 4.1+ | 5.1+ >	1.75E+00
< 4.1+ | 5.1- >	1.67E+00
< 5.1+ | 6.1+ >	2.85E+00
< 5.1+ | 6.1- >	8.06E-01
< 6.1+ | 7.1+ >	1.92E+00
< 6.1+ | 7.1- >	1.48E+00
< 7.1+ | 8.1+ >	1.55E+00
< 7.1+ | 8.1- >	1.78E-01
< 1.1+ | 3.1+ >	6.78E-02
< 1.1+ | 3.1- >	1.80E-02
< 2.1+ | 4.1+ >	3.94E-01
< 2.1+ | 4.1- >	4.26E-01
< 3.1+ | 5.1+ >	2.07E+00
< 3.1+ | 5.1- >	3.27E-01
< 4.1+ | 6.1+ >	9.57E-01
< 4.1+ | 6.1- >	9.57E-01
< 5.1+ | 7.1+ >	1.14E+00
< 5.1+ | 7.1- >	7.49E-01
< 6.1+ | 8.1+ >	5.77E-01
< 6.1+ | 8.1- >	5.62E-01
< 1.1+ | 4.1+ >	5.10E-02
< 1.1+ | 4.1- >	4.18E-02
< 2.1+ | 5.1+ >	1.56E-01
< 2.1+ | 5.1- >	1.71E-01
< 3.1+ | 6.1+ >	3.79E-01
< 3.1+ | 6.1- >	5.24E-01
< 4.1+ | 7.1+ >	4.44E-01
< 4.1+ | 7.1- >	6.55E-01
< 5.1+ | 8.1+ >	4.38E-01
< 5.1+ | 8.1- >	5.27E-01
< 1.1+ | 5.1+ >	6.30E-02
< 1.1+ | 5.1- >	1.88E-02
< 2.1+ | 6.1+ >	1.66E-01
< 2.1+ | 6.1- >	7.92E-02
< 3.1+ | 7.1+ >	3.95E-01
< 3.1+ | 7.1- >	5.00E-01
< 4.1+ | 8.1+ >	9.79E-02
< 4.1+ | 8.1- >	5.20E-02
< 1.1+ | 6.1+ >	2.78E-02
< 1.1+ | 6.1- >	4.27E-02
< 2.1+ | 7.1+ >	1.29E-01
< 2.1+ | 7.1- >	1.93E-01
< 3.1+ | 8.1+ >	2.68E-01
< 3.1+ | 8.1- >	2.45E-01
< 1.1+ | 7.1+ >	4.33E-02
< 1.1+ | 7.1- >	5.63E-02
< 2.1+ | 8.1+ >	1.25E-01
< 2.1+ | 8.1- >	1.24E-01
< 1.1+ | 8.1+ >	3.66E-02
< 1.1+ | 8.1- >	4.11E-02

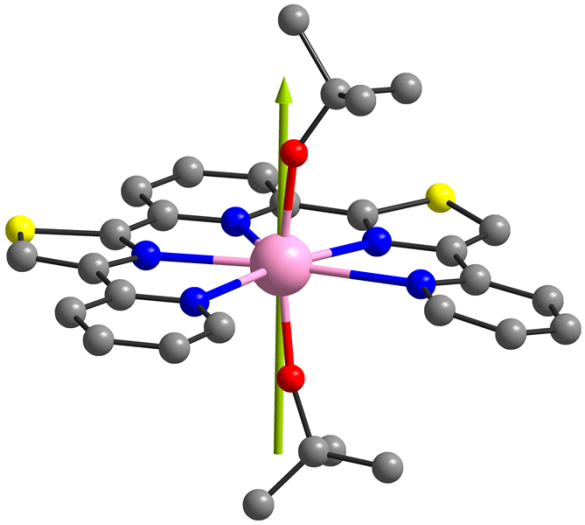

Figure S14 *Ab initio* calculated principal magnetic axis (depicted with lime arrow) for the ground doublet of Dy(III) ion in **1Dy**. Colour codes: Dy, rose; O, red; S, yellow; N, blue and C, grey. All hydrogen atoms are omitted for clarity.

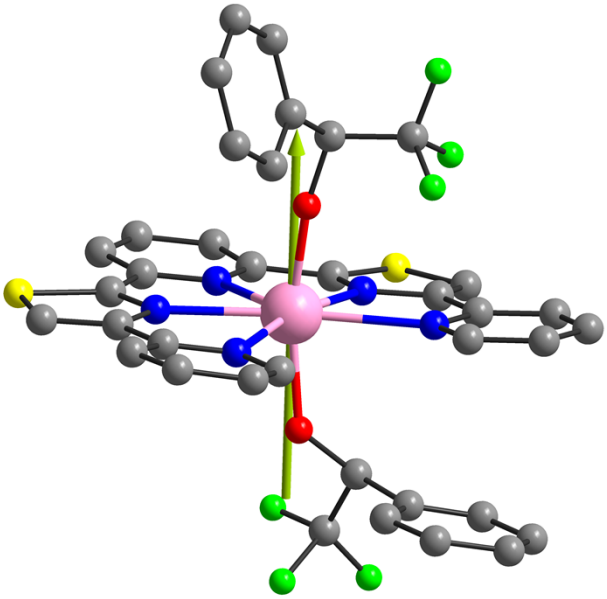

Figure S15 *Ab initio* calculated principal magnetic axis (depicted with lime arrow) for the ground doublet of Dy(III) ion in **3Dy**. Colour codes: Dy, rose; O, red; S, yellow; F, green; N, blue and C, grey. All hydrogen atoms are omitted for clarity.

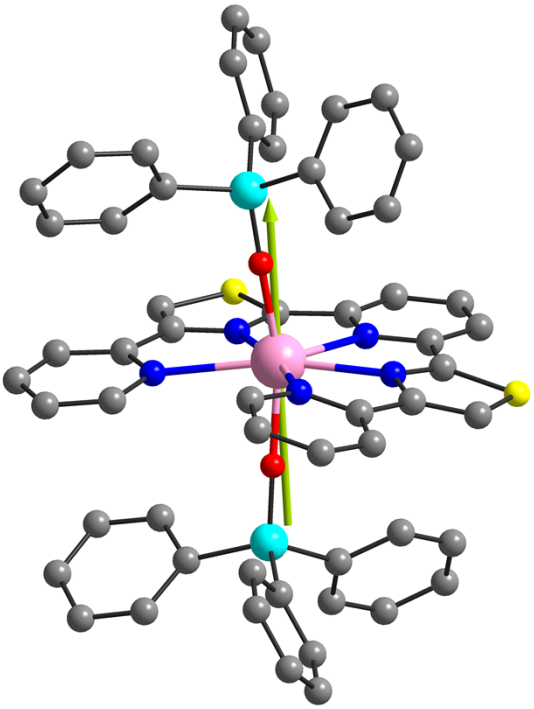

Figure S16 *Ab initio* calculated principal magnetic axis (depicted with lime arrow) for the ground doublet of Dy(III) ion in **4Dy**. Colour codes: Dy, rose; O, red; S, yellow; Si, turquoise; N, blue and C, grey. All hydrogen atoms are omitted for clarity.

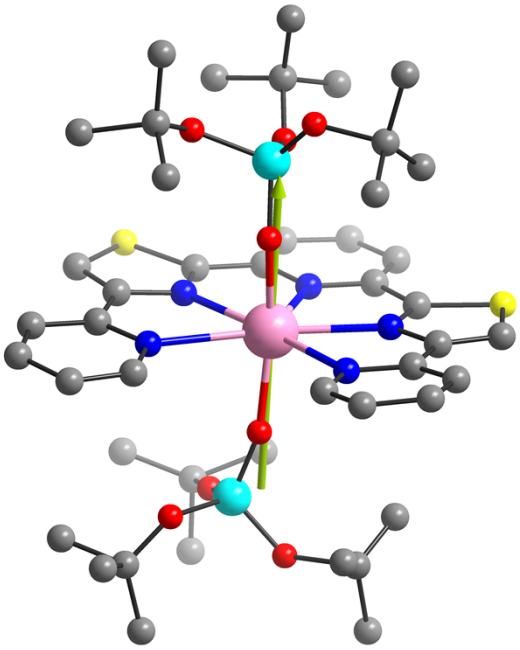

Figure S17 *Ab initio* calculated principal magnetic axis (depicted with lime arrow) for the ground doublet of Dy(III) ion in **5Dy-1**. Colour codes: Dy, rose; O, red; S, yellow; Si, turquoise; N, blue and C, grey. All hydrogen atoms are omitted for clarity.

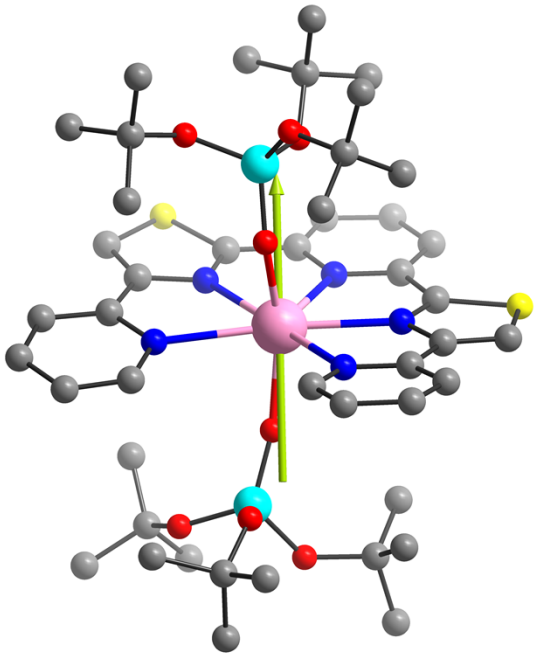

Figure S18 *Ab initio* calculated principal magnetic axis (depicted with lime arrow) for the ground doublet of Dy(III) ion in **5Dy-2**. Colour codes: Dy, rose; O, red; S, yellow; Si, turquoise; N, blue and C, grey. All hydrogen atoms are omitted for clarity.

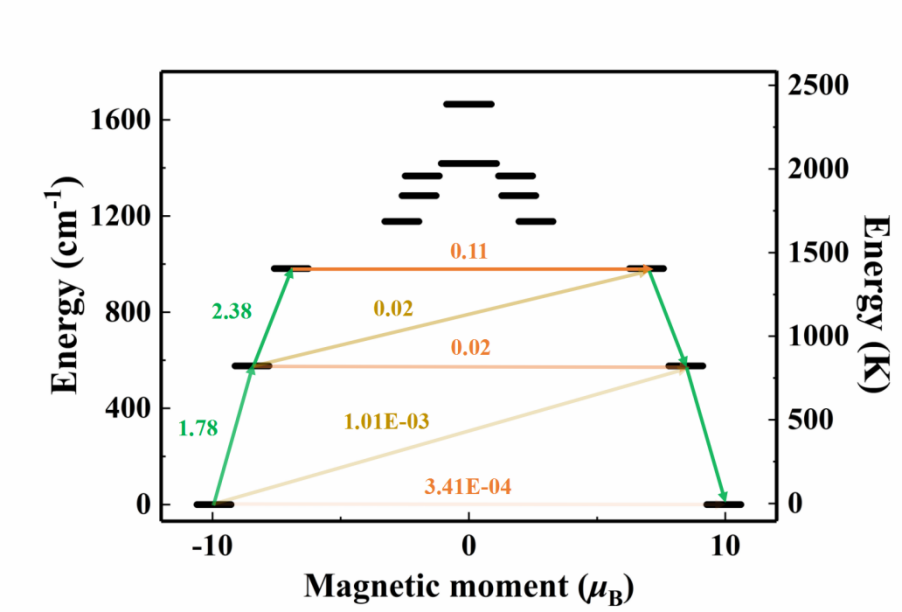

Figure S19 *Ab initio* calculated possible magnetic relaxation path diagram for **1Dy**. The horizontal orange arrows represent the QTM/TA-QTM processes, while the non-horizontal ones show the spin-phonon transition paths. The numbers next to the arrows are averaged transition magnetic moments ($(|\mu_X|+|\mu_Y|+|\mu_Z|)/3$) in μ_B between the connecting pairs.

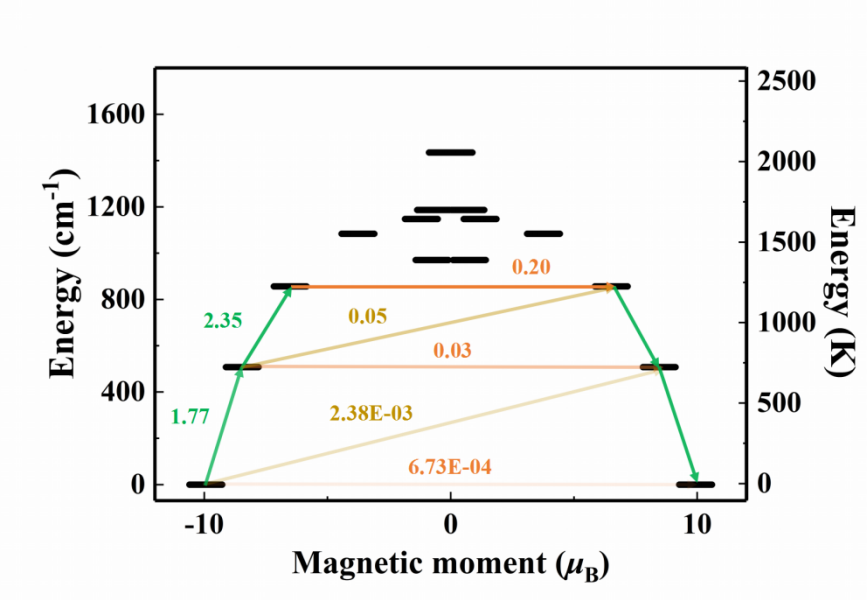

Figure S20 *Ab initio* calculated possible magnetic relaxation path diagram for **3Dy**. The horizontal orange arrows represent the QTM/TA-QTM processes, while the non-horizontal ones show the spin-phonon transition paths. The numbers next to the arrows are averaged transition magnetic moments ($(|\mu_X|+|\mu_Y|+|\mu_Z|)/3$) in μ_B between the connecting pairs.

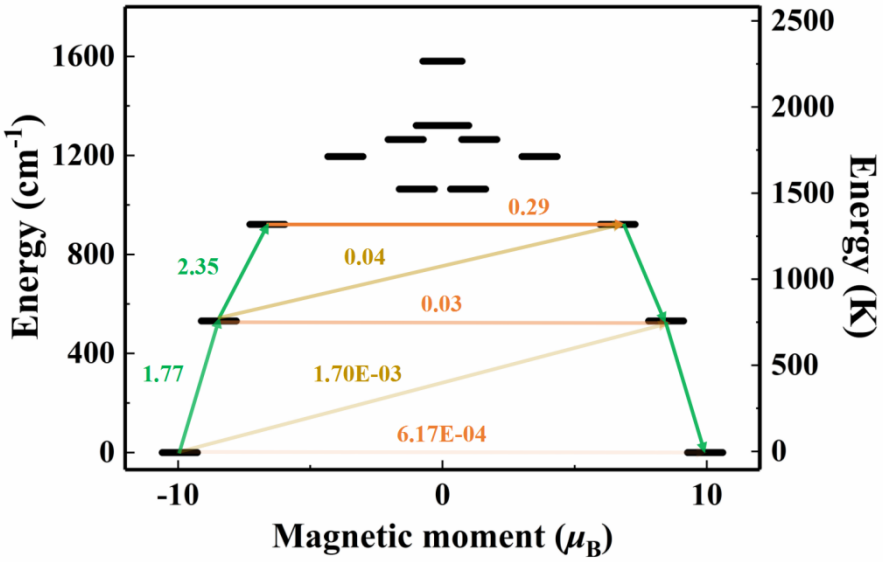

Figure S21 *Ab initio* calculated possible magnetic relaxation path diagram for **4Dy**. The horizontal orange arrows represent the QTM/TA-QTM processes, while the non-horizontal ones show the spin-phonon transition paths. The numbers next to the arrows are averaged transition magnetic moments ($(|\mu_X|+|\mu_Y|+|\mu_Z|)/3$) in μ_B between the connecting pairs.

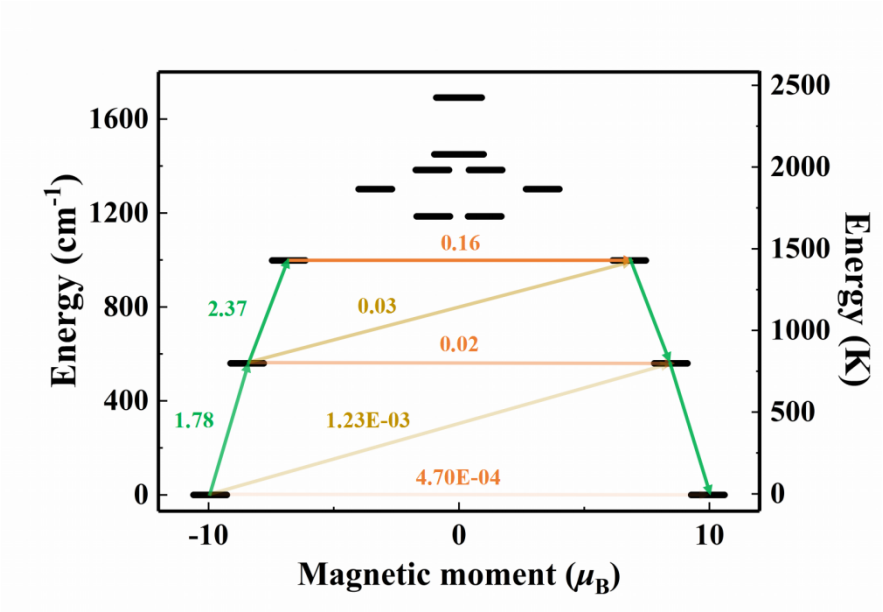

Figure S22 *Ab initio* calculated possible magnetic relaxation path diagram for **5Dy-1**. The horizontal orange arrows represent the QTM/TA-QTM processes, while the non-horizontal ones show the spin-phonon transition paths. The numbers next to the arrows are averaged transition magnetic moments ($(|\mu_X|+|\mu_Y|+|\mu_Z|)/3$) in μ_B between the connecting pairs.

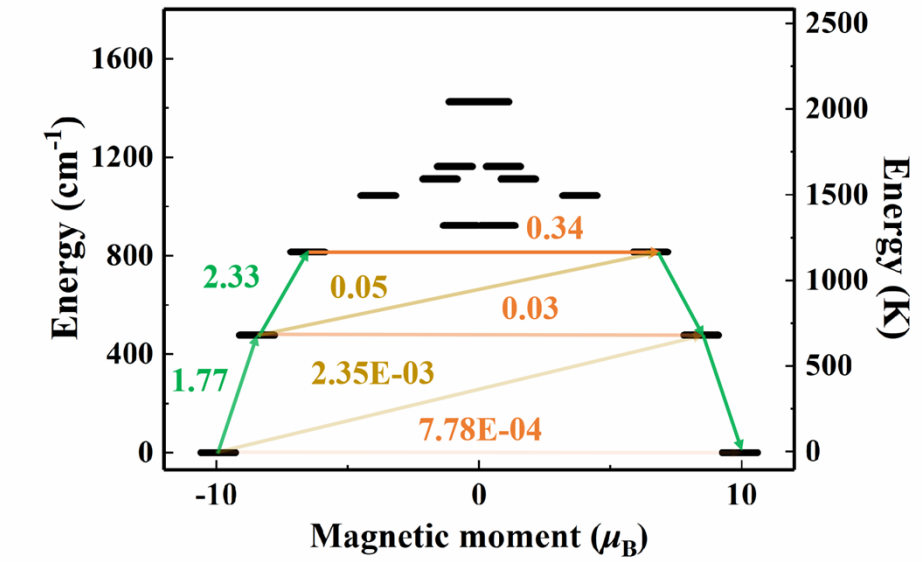

Figure S23 *Ab initio* calculated possible magnetic relaxation path diagram for **5Dy-2**. The horizontal orange arrows represent the QTM/TA-QTM processes, while the non-horizontal ones show the spin-phonon transition paths. The numbers next to the arrows are averaged transition magnetic moments ($(|\mu_X|+|\mu_Y|+|\mu_Z|)/3$) in μ_B between the connecting pairs.

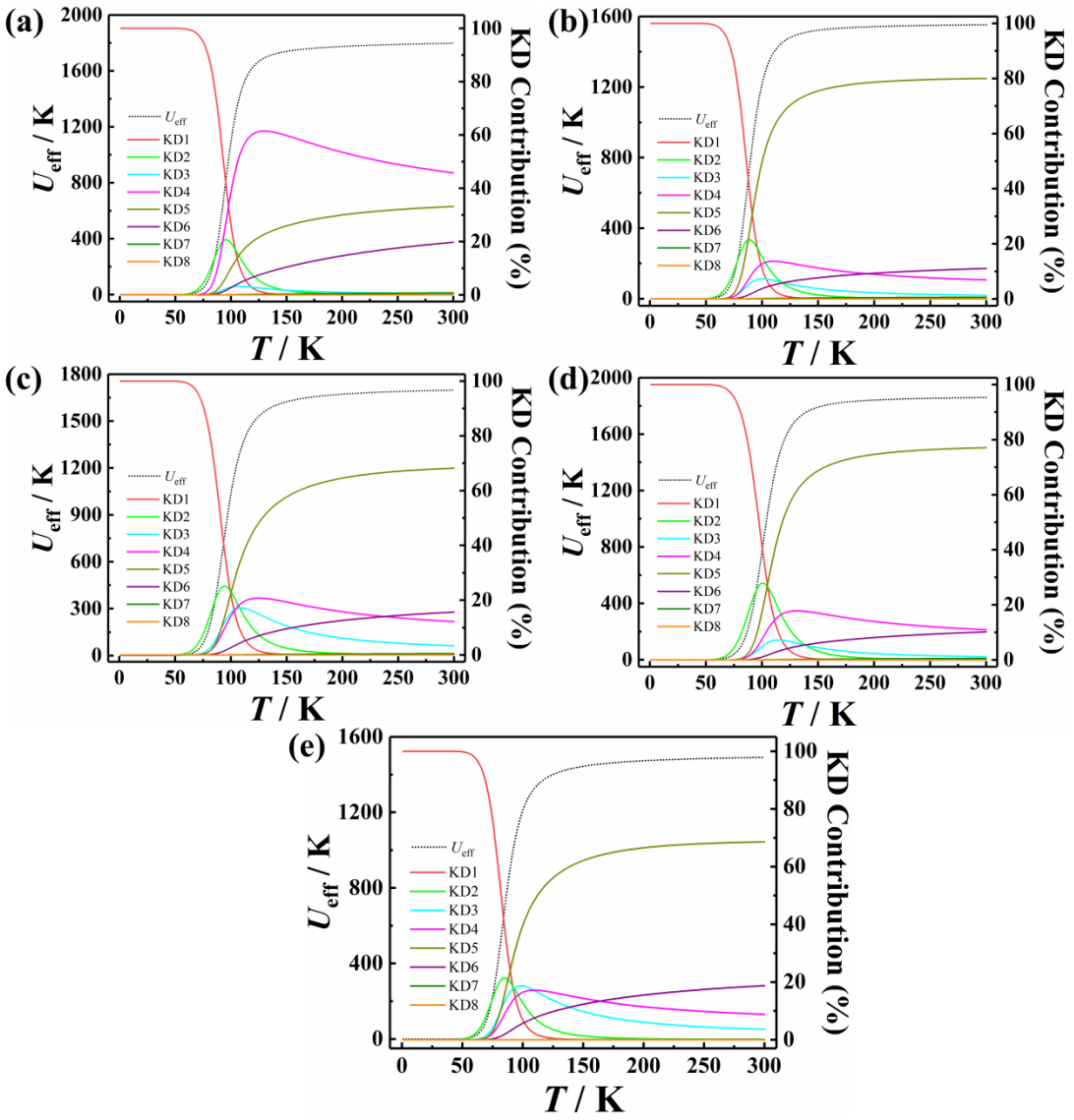

Figure S24 Predicted U_{eff} (black dotted lines) and relaxation contributions from various KDs (coloured solid lines) for **1Dy** (a), **3Dy** (b), **4Dy** (c), **5Dy-1** (d) and **5Dy-2** (e).