

Supporting Information

Homochiral Dy₂ single-molecule magnets with strong magneto-optical Faraday effect and strong third-harmonic generation

Cai-Ming Liu^{*a}, Rong Sun^b, Xiang Hao^a, Xi-Li Li^c and Bing-Wu Wang^{*c}

^a Beijing National Laboratory for Molecular Sciences, CAS Key Laboratory for Organic Solids, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China. Email: cmliu@iccas.ac.cn

^b State Key Laboratory of Rare Earth Materials Chemistry and Applications, College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, China. Email: wangbw@pku.edu.cn

^c Henan Provincial Key Laboratory of Surface and Interface Science, Zhengzhou University of Light Industry, Zhengzhou 450002, China.

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Table S1. Crystal data and structural refinement parameters for *D-1/L-1*, *D-2/L-2*, *D-3/L-3*, *D-4/L-4*.

	<i>D-1</i>	<i>L-1</i>	<i>D-2</i>	<i>L-2</i>	<i>D-3</i>	<i>L-3</i>	<i>D-4</i>	<i>L-4</i>
formula	C ₅₆ H ₆₈ F ₆ N ₁₀ O ₁₄ Dy ₂	C ₅₆ H ₆₈ F ₆ N ₁₀ O ₁₄ Dy ₂	C ₅₈ H ₇₀ Dy ₂ F ₆ N ₈ O ₁₄	C ₅₈ H ₇₀ Dy ₂ F ₆ N ₈ O ₁₄	C ₆₀ H ₆₂ Dy ₂ F ₁₄ N ₁₀ O ₁₂	C ₆₀ H ₆₂ Dy ₂ F ₁₄ N ₁₀ O ₁₂	C ₆₂ H ₆₉ Dy ₂ F ₁₄ N ₈ O ₁₄	C ₆₂ H ₆₉ Dy ₂ F ₁₄ N ₈ O ₁₄
F_w	1544.20	1544.20	1542.22	1542.22	1706.19	1706.19	1741.25	1741.25
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	triclinic	triclinic	monoclinic	monoclinic
space group	$P2_1$	$P2_1$	$P2_1$	$P2_1$	$P1$	$P1$	$P2_1$	$P2_1$
a [Å]	11.9091(2)	11.91880(16)	11.9571(2)	11.94360(18)	11.3419(2)	11.3542(2)	18.0384(5)	18.0064(4)
b [Å]	18.6155(2)	18.5949(3)	18.4949(3)	18.4933(3)	16.6925(3)	16.7025(3)	10.5248(2)	10.5352(2)
c [Å]	13.9636(2)	13.9673(2)	14.1630(2)	14.1704(2)	18.8529(4)	18.8605(4)	18.9442(5)	18.9592(4)
α [°]	90.00	90.00	90.00	90.00	70.521(2)	70.275(2)	90.00	90.00
β [°]	93.0150(10)	92.9810(12)	93.4050(10)	93.2910(14)	89.7460(10)	89.9510(10)	108.627(3)	108.460(2)
γ [°]	90.00	90.00	90.00	90.00	82.7060(10)	82.8120(10)	90.00	90.00
V [Å ³]	3091.36(8)	3091.37(8)	3126.55(9)	3124.75(8)	3335.05(12)	3337.16(12)	3408.17(16)	3411.52(13)
Z	2	2	2	2	2	2	2	2
ρ_{calc} [g cm ⁻³]	1.659	1.659	1.638	1.639	1.701	1.698	1.697	1.692
μ [mm ⁻¹]	2.487	2.487	2.458	2.460	2.329	2.327	2.282	2.280
T [K]	170	170	170	170	170	170	170	170
λ [Å]	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073
reflections collected	53168	53508	49359	62716	89540	89477	50504	46717
unique reflections	16564	16571	16769	16650	24878	24868	18259	12053
observed reflections	16145	15756	16048	15584	23781	22026	16389	11445
parameters	806	806	805	805	1700	1700	913	915
GoF[$I \geq 2\sigma(I)$]	1.024	1.047	1.016	1.027	1.068	1.030	1.045	1.035
R_1 [$I \geq 2\sigma(I)$]	0.0177	0.0222	0.0211	0.0254	0.0628	0.0548	0.0398	0.0317
wR_2 [$I \geq 2\sigma(I)$]	0.0429	0.0526	0.0507	0.0598	0.1722	0.1455	0.1029	0.0804
Flack value	-0.007(2)	0.016(4)	-0.006(4)	-0.019(6)	0.018(17)	0.037(6)	0.011(7)	0.012(9)
CCDC	2308016	2308017	2308018	2308019	2308020	2308021	2308022	2308023

Table S2. Dy (III) ion geometry analysis by SHAPE 2.1 software for *D-1*.

Configuration	ABOXIY Dy1	ABOXIY Dy2
Octagon(D_{8h})	32.232	32.403
Heptagonal pyramid(C_{7v})	21.806	21.495
Hexagonal bipyramid(D_{6h})	11.660	12.296
Cube (O_h)	7.722	8.277
Square antiprism (D_{4d})	2.585	2.999
Triangular dodecahedron (D_{2d})	1.726	1.487
Johnson gyrobifastigium J26 (D_{2d})	12.057	12.518
Johnson elongated triangular bipyramid J14 (D_{3h})	26.094	26.098
Biaugmented trigonal prism J50 (C_{2v})	3.482	3.654
Biaugmented trigonal prism (C_{2v})	2.291	2.420
Snub diphenoid J84 (D_{2d})	5.024	4.685
Triakis tetrahedron (T_d)	8.591	9.072
Elongated trigonal bipyramid (D_{3h})	23.298	23.074

Table S3. Dy (III) ion geometry analysis by SHAPE 2.1 software for *D-2*.

Configuration	ABOXIY Dy1	ABOXIY Dy2
Octagon(D_{8h})	32.235	32.327
Heptagonal pyramid(C_{7v})	21.879	21.434
Hexagonal bipyramid(D_{6h})	11.763	12.297
Cube (O_h)	7.903	8.356
Square antiprism (D_{4d})	2.596	3.059
Triangular dodecahedron (D_{2d})	1.784	1.530
Johnson gyrobifastigium J26 (D_{2d})	11.944	12.438
Johnson elongated triangular bipyramid J14 (D_{3h})	26.665	26.256
Biaugmented trigonal prism J50 (C_{2v})	3.370	3.655
Biaugmented trigonal prism (C_{2v})	2.130	2.396
Snub diphenoid J84 (D_{2d})	5.106	4.719
Triakis tetrahedron (T_d)	8.745	9.157
Elongated trigonal bipyramid (D_{3h})	23.274	23.229

Table S4. Dy (III) ion geometry analysis by SHAPE 2.1 software for *D-3*.

Configuration	ABOXIY Dy1	ABOXIY Dy2	ABOXIY Dy3	ABOXIY Dy4
Octagon(D_{8h})	30.500	30.053	30.942	31.076
Heptagonal pyramid(C_{7v})	21.402	22.185	22.268	22.092
Hexagonal bipyramid(D_{6h})	14.775	14.809	14.455	12.365
Cube (O_h)	12.478	12.788	11.974	10.412
Square antiprism (D_{4d})	4.622	5.003	4.285	3.944
Triangular dodecahedron (D_{2d})	2.589	2.590	2.406	2.844
Johnson gyrobifastigium J26 (D_{2d})	11.770	11.154	10.884	10.337
Johnson elongated triangular bipyramid J14 (D_{3h})	22.146	21.740	22.250	22.052
Biaugmented trigonal prism J50 (C_{2v})	3.040	2.589	3.043	3.014
Biaugmented trigonal prism (C_{2v})	2.896	2.600	2.955	2.564
Snub diphenoid J84 (D_{2d})	3.463	3.058	2.888	3.525
Triakis tetrahedron (T_d)	13.269	13.587	12.746	11.224
Elongated trigonal bipyramid (D_{3h})	20.409	20.089	20.721	20.406

Table S5. Dy (III) ion geometry analysis by SHAPE 2.1 software for *D-4*.

Configuration	ABOXIY Dy1	ABOXIY Dy2
Octagon(D_{8h})	32.525	33.076
Heptagonal pyramid(C_{7v})	21.233	22.566
Hexagonal bipyramid(D_{6h})	10.882	10.357
Cube (O_h)	7.678	7.297
Square antiprism (D_{4d})	3.020	2.824
Triangular dodecahedron (D_{2d})	2.193	2.365
Johnson gyrobifastigium J26 (D_{2d})	11.150	10.941
Johnson elongated triangular bipyramid J14 (D_{3h})	25.509	26.361
Biaugmented trigonal prism J50 (C_{2v})	3.801	3.710
Biaugmented trigonal prism (C_{2v})	2.496	2.451
Snub diphenoid J84 (D_{2d})	5.605	6.007
Triakis tetrahedron (T_d)	8.541	8.098
Elongated trigonal bipyramid (D_{3h})	22.925	22.986

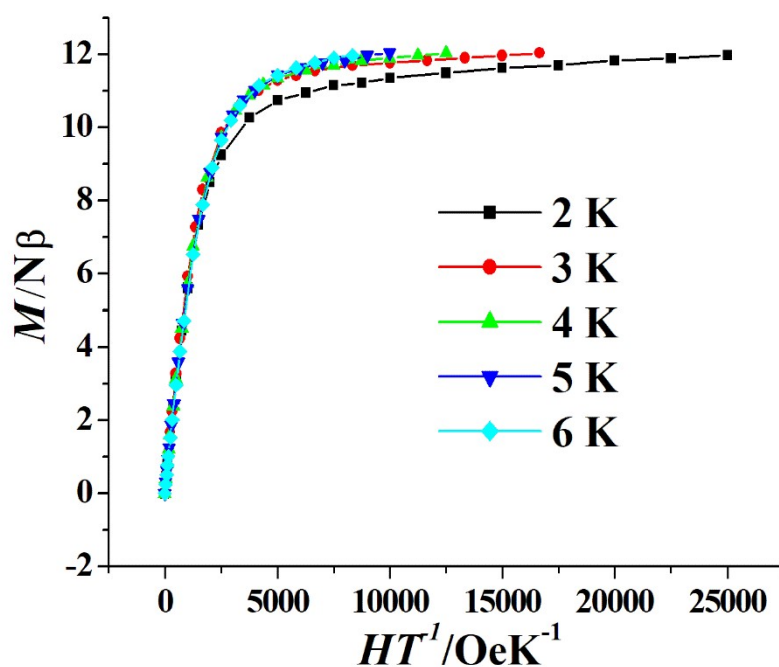


Fig. S1. M versus H/T plots at 2-6 K of *D-1*.

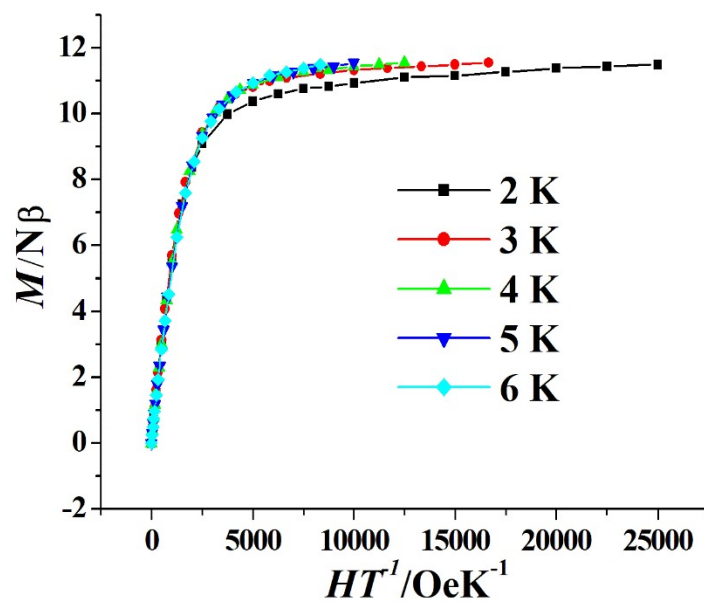


Fig. S2. M versus H/T plots at 2-6 K of $D-2$.

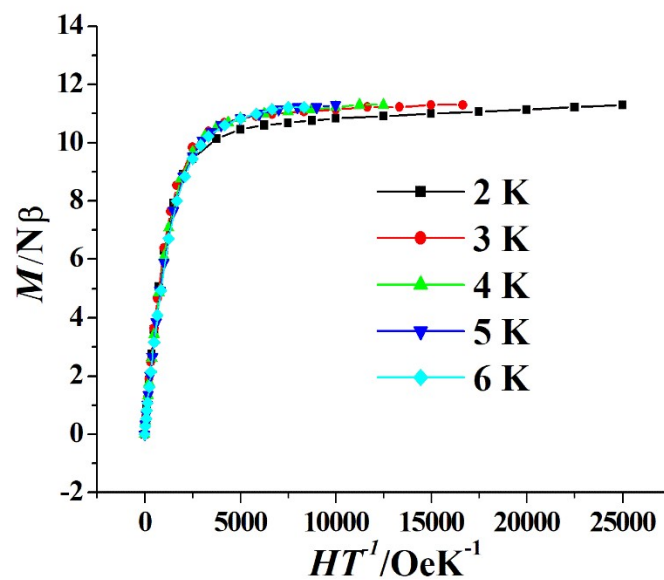


Fig. S3. M versus H/T plots at 2-6 K of $D-3$.

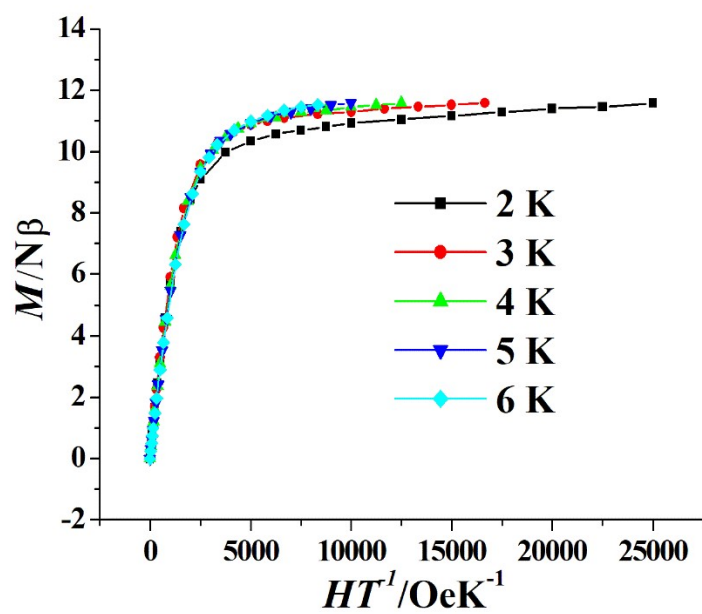


Fig. S4. M versus H/T plots at 2-6 K of $D-4$.

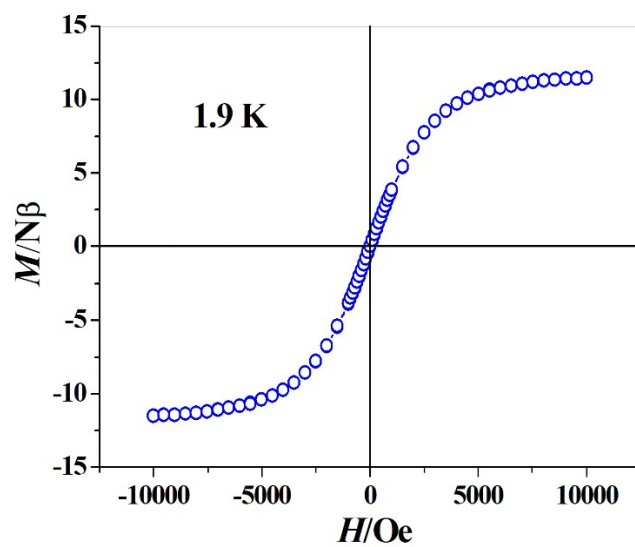


Fig. S5. Hysteresis loop for $D-1$ at 1.9 K.

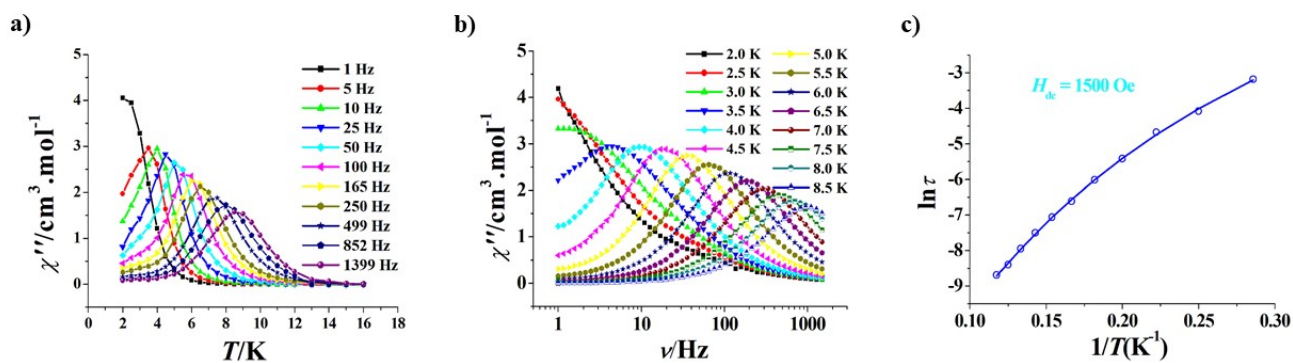


Fig. S6. Plots of χ'' versus T for $D-1$ ($H_{dc} = 1500$ Oe) (a). Frequency dependence of χ'' for $D-1$ at 1500 Oe (b). Plot of $\ln(\tau)$ versus $1/T$ for $D-1$ ($H_{dc} = 1500$ Oe), the solid line represents the best fitting with Orbach plus Raman (c).

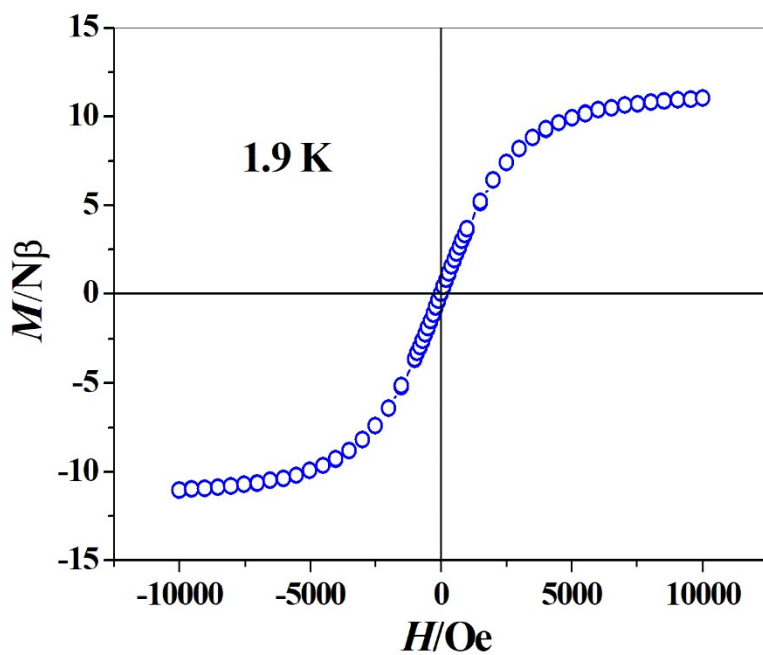


Fig. S7. Hysteresis loop for $D-2$ at 1.9 K.

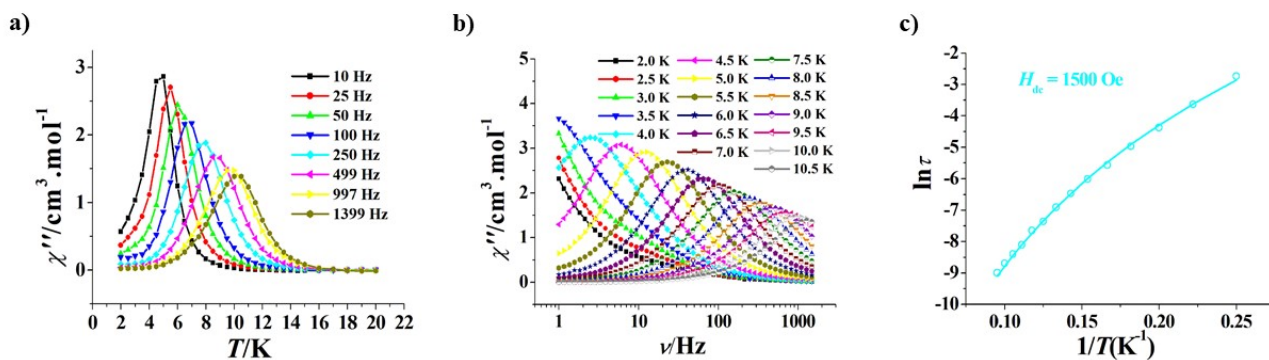


Fig. S8. Plots of χ'' versus T for $D-2$ ($H_{dc} = 1500$ Oe) (a). Frequency dependence of χ'' for $D-2$ at 1500 Oe (b). Plot of $\ln(\tau)$ versus $1/T$ for $D-2$ ($H_{dc} = 1500$ Oe), the solid line represents the best fitting with Orbach plus Raman (c).

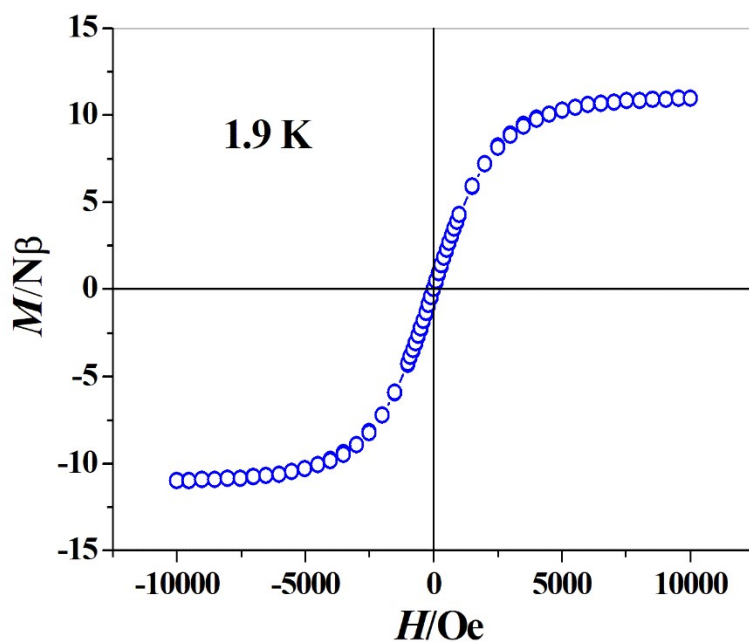


Fig. S9. Hysteresis loop for $D-3$ at 1.9 K.

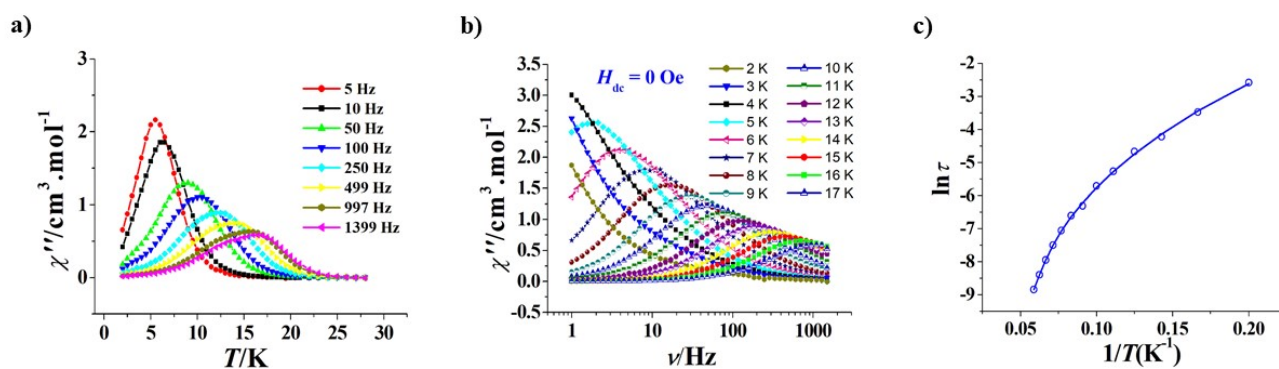


Fig. S10. Plots of χ'' versus T for $D-3$ ($H_{dc} = 1500$ Oe) (a). Frequency dependence of χ'' for $D-3$ at 1500 Oe (b). Plot of $\ln(\tau)$ versus $1/T$ for $D-3$ ($H_{dc} = 1500$ Oe), the solid line represents the best fitting with Orbach plus Raman (c).

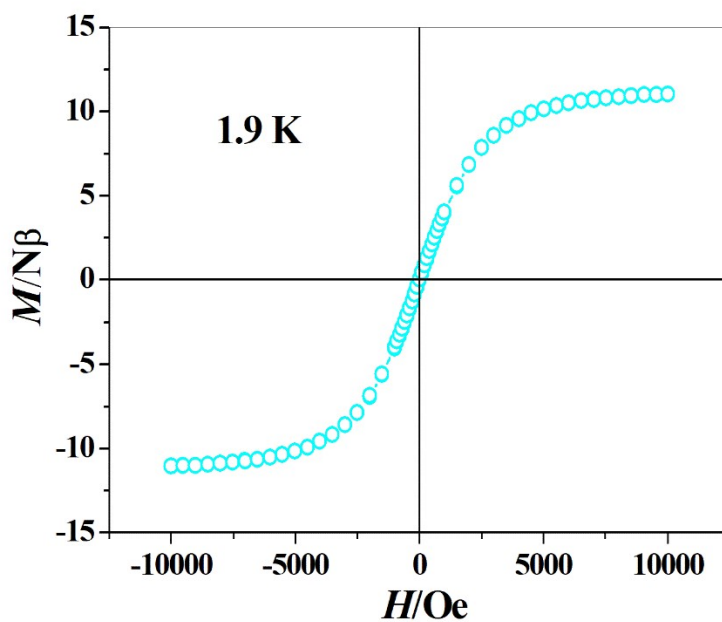


Fig. S11. Hysteresis loop for $D-4$ at 1.9 K.

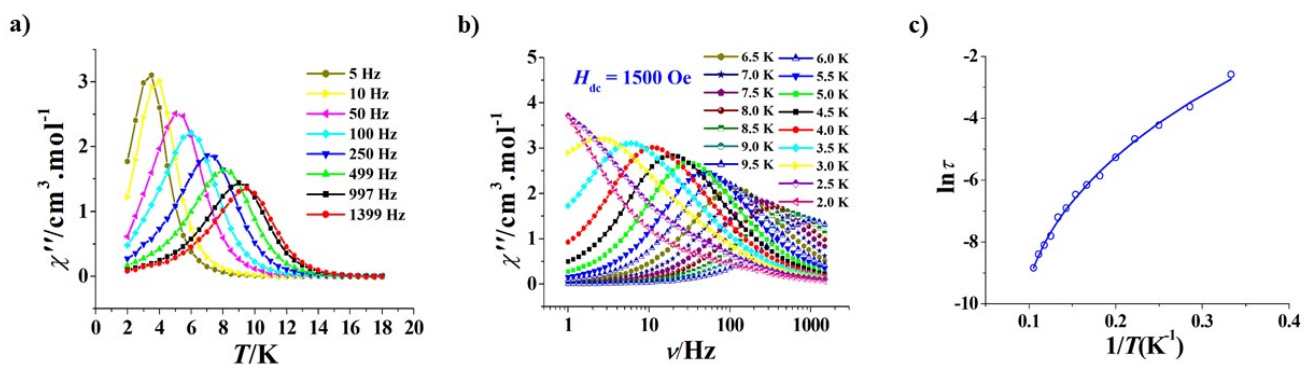


Fig. S12. Plots of χ'' versus T for D-4 ($H_{dc} = 1500$ Oe) (a). Frequency dependence of χ'' for D-4 at 1500 Oe (b). Plot of $\ln(\tau)$ versus $1/T$ for D-4 ($H_{dc} = 1500$ Oe), the solid line represents the best fitting with Orbach plus Raman (c).

Supporting Information for theoretical calculations

Ab initio calculations were performed with MOLCAS 8.1 program.¹ All atoms are described by relativistic atomic natural orbital (ANO) basis sets from the MOLCAS ANO-RCC library: ANO-RCC-VTZP for Dy (III) ion; VTZ for coordinated O and N; VDZ for other atoms, accounting scalar relativistic contractions. A second-order Douglas-Kroll-Hess Hamiltonian was employed. The spin-orbit coupling was handled separately through the restricted-active-space state interaction procedure (RASSI-SO). Active electrons in seven active spaces include all *f* electrons (CAS (9, 7)) of Dy (III) in the CASSCF calculation.

The intramolecular interaction between two Dy (III) ions was also calculated using theoretical method. Firstly, each Dy (III) fragment was calculated using CASSCF to obtain the corresponding magnetic properties. Then, the exchange interaction between the magnetic centers is considered within the Lines model², while the account of the dipole-dipole magnetic coupling is treated exactly. The Lines model is effective and has been successfully used widely in the research field of f-element single-molecule magnets. For this compound, there is only one type of *J* (Dy-Dy). The Hamiltonian is as follows,

$$\hat{H}_{exch} = -J_{total} \hat{S}_{Dy1} \hat{S}_{Dy2}$$

Where the J_{total} is the parameter of the total magnetic interaction ($J_{total} = J_{dip} + J_{exch}$) between magnetic center ions. The $\hat{S}_{Dy1} = \pm 1/2$ are the ground pseudospin on the Dy (III) sites. The dipolar magnetic coupling can be calculated in Ising approximation, while the exchange coupling constant was fitted through comparison of the computed and measured magnetic susceptibility and molar magnetization using the POLY_ANISO program.³

Table S6. Calculated energy levels (K) and g (g_x, g_y, g_z) tensors of the lowest Kramers doublets (KDs) of individual Dy(III) fragment for *D-1*.

KDs	Dy1		Dy2	
	E / K	g	E / K	g
1	0.0	0.019	0.0	0.023
		0.024		0.029
		19.541		19.543
2	183.6	0.193	192.2	0.095
		0.213		0.111
		16.634		16.548
3	374.2	1.886	363.2	9.733
		2.845		7.516
		12.211		3.113
4	436.3	1.168	404.8	2.129
		3.142		3.964
		15.323		11.844
5	509.3	3.257	488.2	4.501
		5.958		6.612
		9.981		9.347
6	621.7	0.286	580.8	0.728
		1.346		1.127
		16.056		16.968
7	712.0	0.069	697.0	0.061
		1.252		0.734
		17.545		18.464
8	766.5	0.349	775.5	0.162
		0.802		0.413
		19.056		19.342

Table S7. Calculated energy levels (K) and g (g_x, g_y, g_z) tensors of the lowest Kramers doublets (KDs) of individual Dy(III) fragment for *D-2*.

KDs	Dy1		Dy2	
	E / K	g	E / K	g
1	0.0	0.024	0.0	0.014
		0.037		0.020
		19.504		19.460
2	180.3	0.250	193.0	0.098
		0.310		0.107
		16.551		16.484
3	344.6	3.115	384.3	9.387
		4.819		7.514
		11.361		2.725
4	394.5	0.429	419.2	2.998
		2.705		3.908
		14.270		10.927
5	470.1	2.664	502.7	4.029
		5.531		5.605
		9.825		10.888
6	602.2	0.278	583.0	0.969
		1.745		1.296
		15.339		17.113
7	683.6	0.272	711.1	0.548
		1.393		1.633
		17.214		16.758
8	735.9	0.327	740.7	0.374
		0.643		2.100
		19.072		17.329

Table S8. Calculated energy levels (K) and g (g_x, g_y, g_z) tensors of the lowest Kramers doublets (KDs) of individual Dy(III) fragment for *D-3*.

KDs	Dy1		Dy2		Dy3		Dy4	
	E/K	g	E/K	g	E/K	g	E/K	g
1	0.0	0.007	0.0	0.003	0.0	0.006	0.0	0.006
		0.010		0.005		0.009		0.012
		19.679		19.641		19.691		19.485
2	294.5	0.099	294.5	0.032	304.5	0.064	252.8	0.107
		0.296		0.084		0.161		0.273
		16.916		16.820		16.896		16.445
3	419.4	0.210	429.5	0.182	472.1	0.580	356.7	0.243
		0.607		0.364		1.483		0.743
		18.284		19.145		17.207		18.380
4	498.5	0.998	519.8	1.350	533.6	0.818	431.8	0.843
		4.254		1.581		4.336		2.099
		11.017		12.469		11.894		14.541
5	532.1	0.501	577.4	0.164	582.0	1.681	505.0	0.316
		4.115		2.918		4.046		3.453
		12.213		10.674		8.333		9.222
6	601.6	2.800	660.4	4.968	681.8	2.475	593.3	7.489
		3.554		5.471		6.196		7.151
		10.470		8.243		11.742		3.723
7	688.5	2.821	701.6	1.709	708.4	1.240	634.8	1.546
		4.631		1.980		3.760		2.266
		11.148		15.113		10.484		16.608
8	774.4	0.665	764.4	0.745	776.1	0.726	660.9	1.050
		1.950		1.060		1.555		1.300
		17.893		17.931		17.993		16.866

Table S9. Calculated energy levels (K) and g (g_x, g_y, g_z) tensors of the lowest Kramers doublets (KDs) of individual Dy(III) fragment for *D-4*.

KDs	Dy1		Dy2	
	E / K	g	E / K	g
1	0.0	0.006	0.0	0.001
		0.029		0.016
		19.454		19.532
2	161.0	0.163	174.5	0.194
		0.238		0.249
		16.575		16.674
3	314.0	10.126	367.5	2.863
		7.358		4.567
		3.178		11.503
4	347.8	1.823	426.8	0.511
		3.767		3.423
		12.664		14.697
5	432.5	3.859	510.6	3.834
		6.354		6.194
		9.369		9.615
6	506.5	1.461	620.2	0.731
		1.902		1.638
		16.543		15.657
7	656.4	0.445	724.3	11.030
		0.622		6.506
		18.447		1.556
8	713.7	0.252	745.7	12.466
		0.961		7.569
		18.679		1.046

Table S10. Wave functions with definite projection of the total moment $|m_J\rangle$ for the lowest three Kramers doublets (KDs) of individual Dy^{3+} fragments in *D-1*.

	E / K	wave functions
	0.0	95.4% $ \pm 15/2\rangle$
Dy1	183.6	88.4% $ \pm 13/2\rangle$ +4.7% $ \pm 7/2\rangle$
	374.2	64.8% $ \pm 11/2\rangle$ +6.9% $ \pm 9/2\rangle$ +8.0% $ \pm 3/2\rangle$ +8.8% $ \pm 1/2\rangle$
<hr/>		
	0.0	95.7% $ \pm 15/2\rangle$
Dy2	192.2	88.2% $ \pm 13/2\rangle$ +4.8% $ \pm 7/2\rangle$
	363.2	54.2% $ \pm 11/2\rangle$ +9.7% $ \pm 9/2\rangle$ +14.5% $ \pm 3/2\rangle$ +10.5% $ \pm 1/2\rangle$
		>

Table S11. Wave functions with definite projection of the total moment $|m_J\rangle$ for the lowest three Kramers doublets (KDs) of individual Dy^{3+} fragments in *D-2*.

	E / K	wave functions
	0.0	95.3% $ \pm 15/2\rangle$
Dy1	180.3	84.5% $ \pm 13/2\rangle$ +4.7% $ \pm 11/2\rangle$ +4.4% $ \pm 7/2\rangle$
	344.6	45.7% $ \pm 11/2\rangle$ +10.0% $ \pm 9/2\rangle$ +4.6% $ \pm 7/2\rangle$ +16.3% $ \pm 3/2\rangle$ +17.9% $ \pm 1/2\rangle$
		>
<hr/>		
	0.0	93.5% $ \pm 15/2\rangle$
Dy2	192.2	88.8% $ \pm 13/2\rangle$ +3.5% $ \pm 9/2\rangle$ +4.7% $ \pm 7/2\rangle$
	363.2	48.2% $ \pm 11/2\rangle$ +8.4% $ \pm 9/2\rangle$ +6.3% $ \pm 7/2\rangle$ +15.6% $ \pm 3/2\rangle$ +14.5% $ \pm 1/2\rangle$

Table S12. Wave functions with definite projection of the total moment $|m_J\rangle$ for the lowest three Kramers doublets (KDs) of individual Dy^{3+} fragments in *D-3*.

	E / K	wave functions
	0.0	97.0% $ \pm 15/2\rangle$
Dy1	294.5	80.0% $ \pm 13/2\rangle$ +8.3% $ \pm 11/2\rangle$ +7.6% $ \pm 9/2\rangle$
	419.4	41.6% $ \pm 1/2\rangle$ +33.3% $ \pm 3/2\rangle$ +16.3% $ \pm 5/2\rangle$ +4.5% $ \pm 7/2\rangle$
<hr/>		
	0.0	95.8% $ \pm 15/2\rangle$
Dy2	294.5	80.9% $ \pm 13/2\rangle$ +5.4% $ \pm 11/2\rangle$ +9.2% $ \pm 9/2\rangle$
	429.5	29.7% $ \pm 1/2\rangle$ +29.8% $ \pm 3/2\rangle$ +22.8% $ \pm 5/2\rangle$ +11.0% $ \pm 7/2\rangle$ +4.0% $ \pm 9/2\rangle$
<hr/>		
	0.0	96.2% $ \pm 15/2\rangle$
Dy3	304.5	82.5% $ \pm 13/2\rangle$ +6.5% $ \pm 11/2\rangle$ +7.9% $ \pm 9/2\rangle$
	472.1	52.6% $ \pm 1/2\rangle$ +28.6% $ \pm 3/2\rangle$ +7.7% $ \pm 5/2\rangle$ +4.6% $ \pm 7/2\rangle$
<hr/>		
Dy4	0.0	93.6% $ \pm 15/2\rangle$
	294.5	83.4% $ \pm 13/2\rangle$ +9.3% $ \pm 11/2\rangle$ +3.5% $ \pm 9/2\rangle$

429.5 45.4% $|\pm 1/2\rangle$ +30.1% $|\pm 3/2\rangle$ +14.9% $|\pm 5/2\rangle$ +6.0% $|\pm 7/2\rangle$ +1.1% $|\pm 9/2\rangle$

Table S13. Wave functions with definite projection of the total moment $|m_J\rangle$ for the lowest three Kramers doublets (KDs) of individual Dy³⁺ fragments in *D-4*.

	E / K	wave functions
Dy1	0.0	94.5% $ \pm 15/2\rangle$
	161.0	81.5% $ \pm 13/2\rangle$ +7.5% $ \pm 11/2\rangle$ +5.4% $ 7/2\rangle$
	314.0	44.8% $ \pm 11/2\rangle$ +17.8% $ \pm 9/2\rangle$ +17.0% $ \pm 3/2\rangle$ +10.4% $ \pm 1/2\rangle$
		>
Dy2	0.0	95.1% $ \pm 15/2\rangle$
	174.5	86.5% $ \pm 13/2\rangle$ +4.6% $ \pm 11/2\rangle$ +4.5% $ 7/2\rangle$
	363.2	59.8% $ \pm 11/2\rangle$ +8.1% $ \pm 9/2\rangle$ +10.8% $ \pm 3/2\rangle$ +7.5% $ \pm 1/2\rangle$

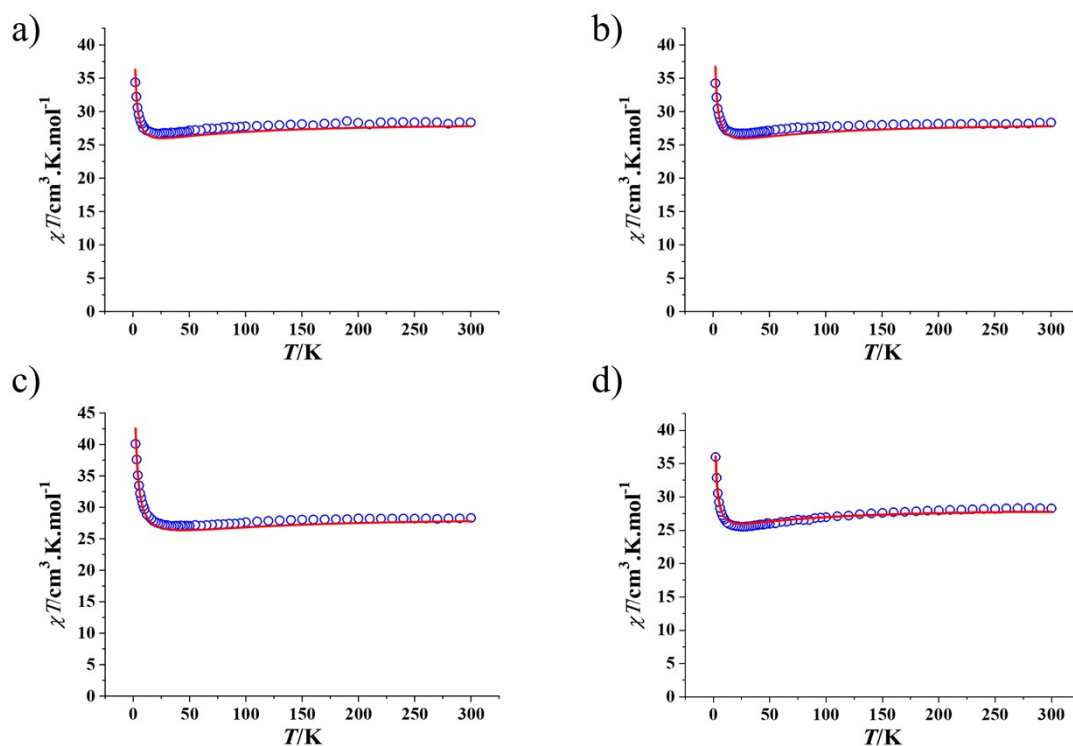


Fig. S13. Plot of χT versus T of *D-1* ($H_{dc} = 1000$ Oe) (a); Plot of χT versus T of *D-2* ($H_{dc} = 1000$ Oe) (b); Plot of χT versus T of *D-3* ($H_{dc} = 1000$ Oe), and the simulated results are obtained from Dy1 and Dy2. (c); Plot of χT versus T of *D-4* ($H_{dc} = 1000$ Oe) (d); The red line corresponds to the simulated results.

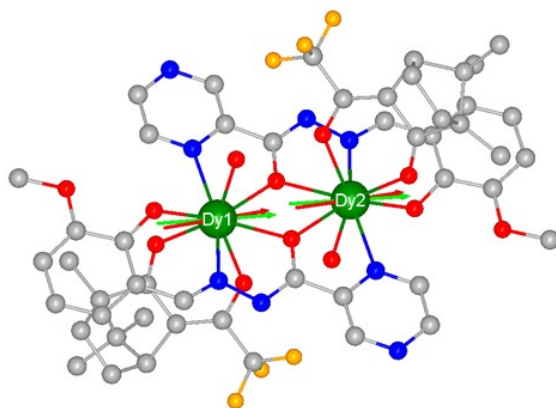


Fig. S14. The orientations of easy axis of the KDs on Dy^{3+} for *D-1*. The green arrow represents the ground KDs and the red stands for the first excited KDs. Color codes: Dy, green; N, blue; O, red; C, grey; F, light orange. All hydrogens are omitted for clarity.

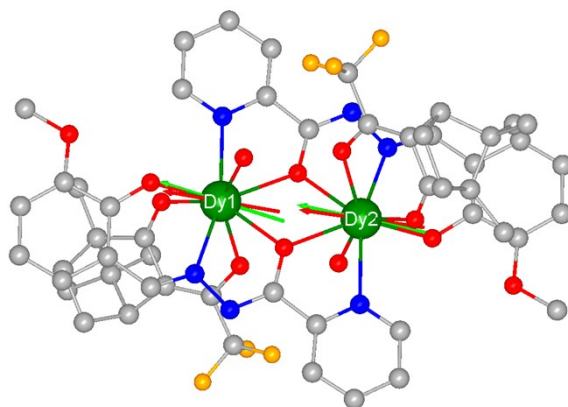


Fig. S15. The orientations of easy axis of the KDs on Dy^{3+} for *D-2*. The green arrow represents the ground KDs and the red stands for the first excited KDs. Color codes: Dy, green; N, blue; O, red; C, grey; F, light orange. All hydrogens are omitted for clarity.

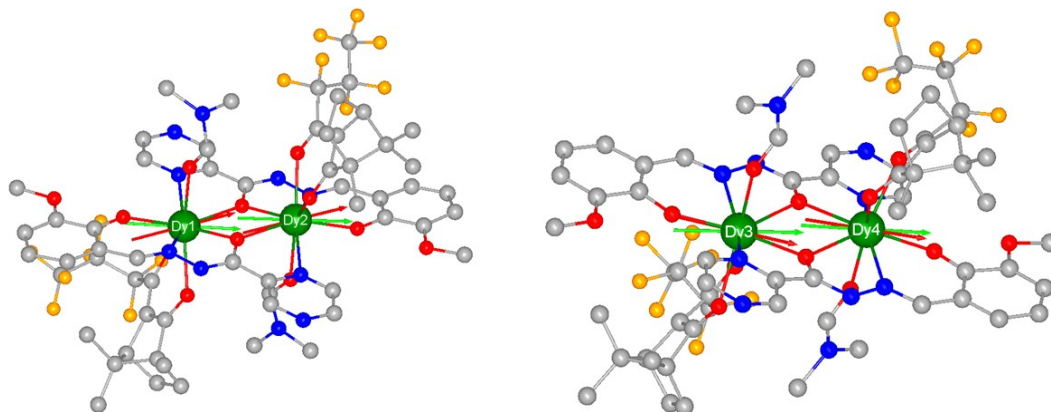


Fig. S16. The orientations of easy axis of the KDs on Dy^{3+} for *D-3*. The green arrow represents the ground KDs and the red stands for the first excited KDs. Color codes: Dy, green; N, blue; O, red; C, grey; F, light orange. All hydrogens are omitted for clarity.

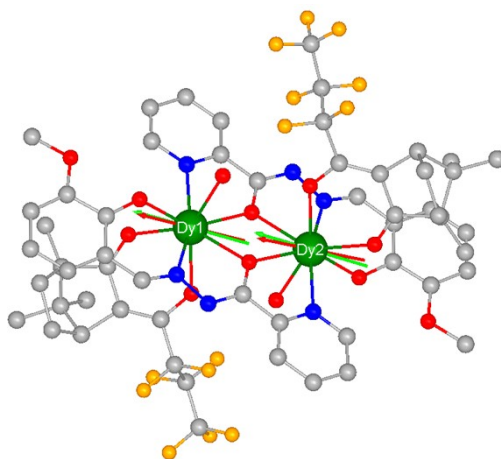


Fig. S17. The orientations of easy axis of the KDs on Dy^{3+} for *D-4*. The green arrow represents the ground KDs and the red stands for the first excited KDs. Color codes: Dy, green; N, blue; O, red; C, grey; F, light orange. All hydrogens are omitted for clarity.

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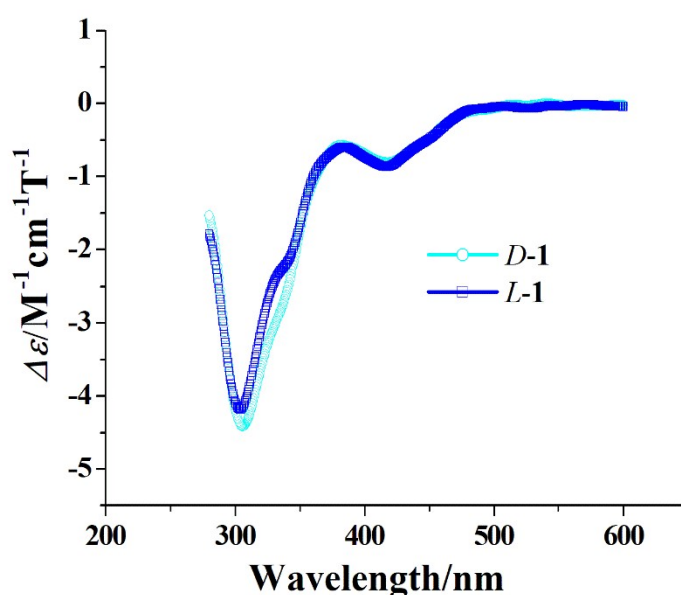


Fig. S18. MCD spectra of *D-1* and *L-1*.

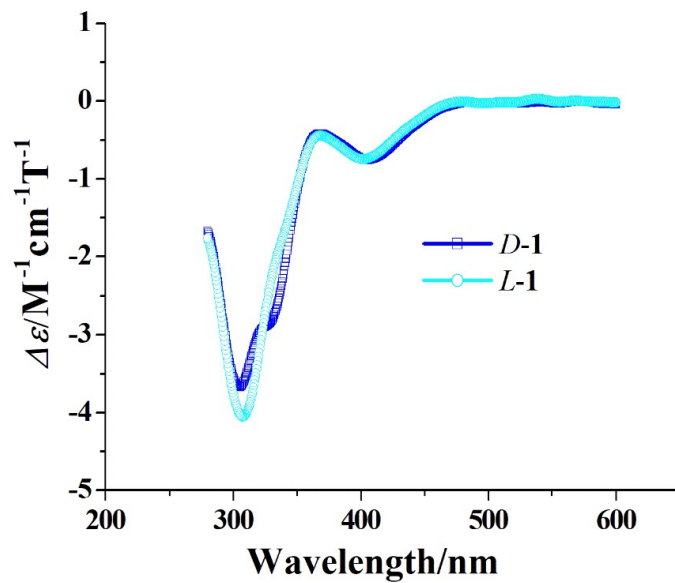


Fig. S19. MCD spectra of *D-2* and *L-2*.

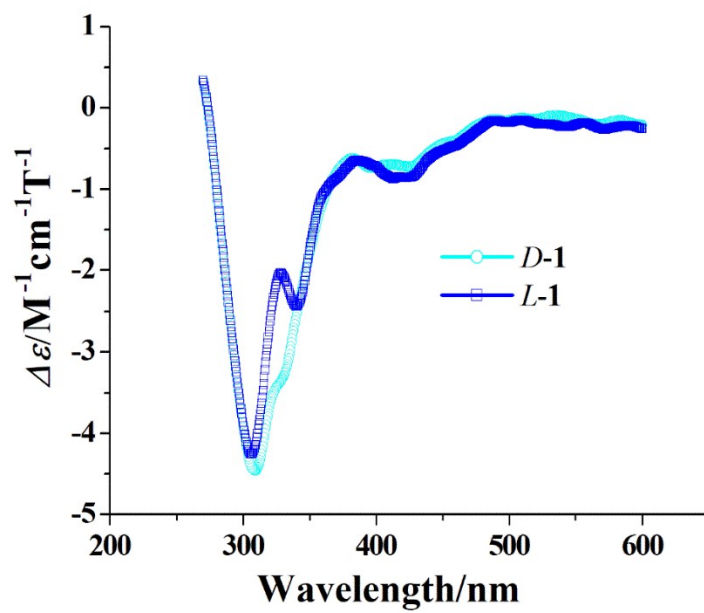


Fig. S20. MCD spectra of *D-3* and *L-3*.

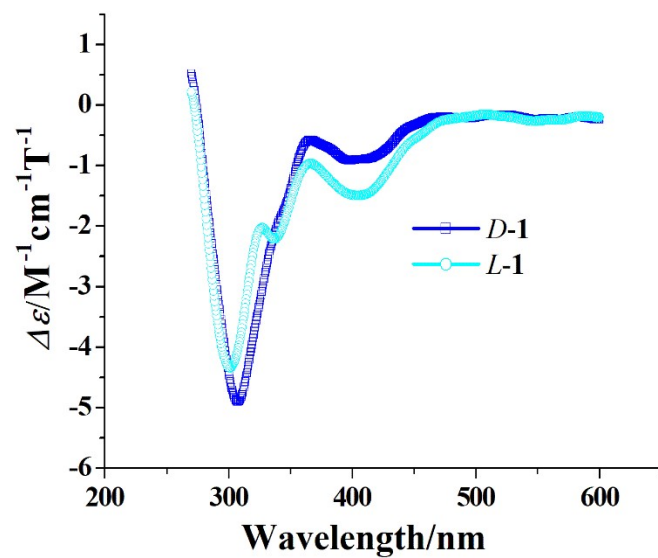


Fig. S21. MCD spectra of *D-4* and *L-4*.

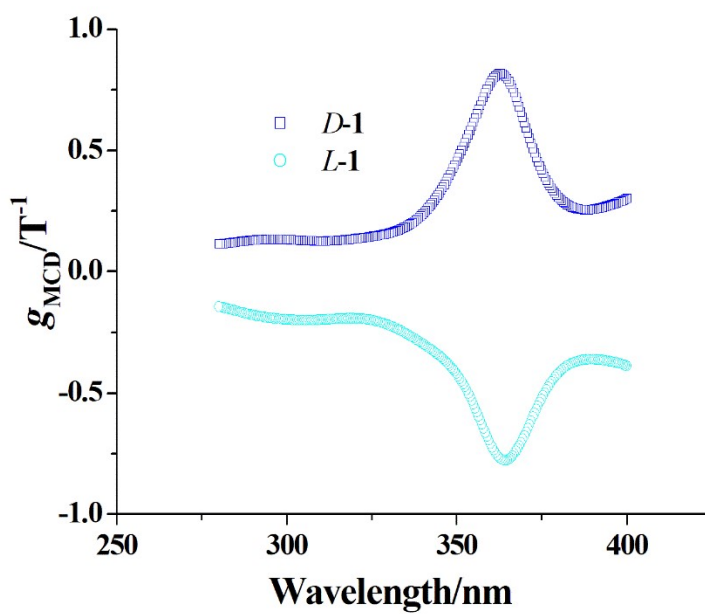


Fig. S22. The wavelength-dependent g_{MCD} of *D-1* and *L-1*.

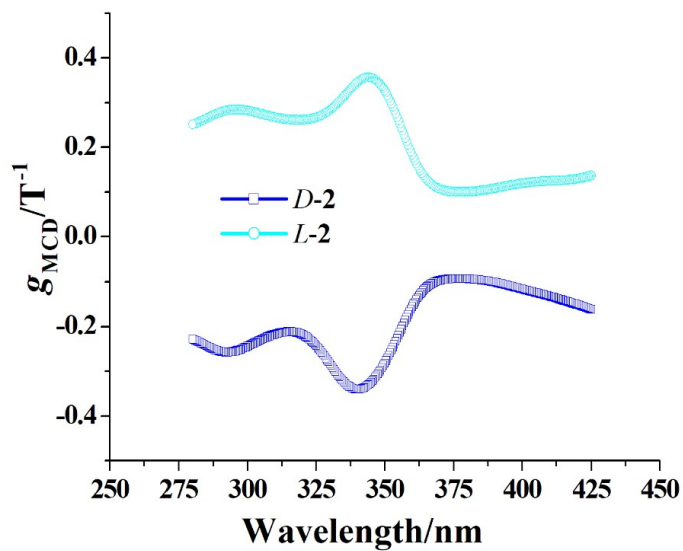


Fig. S23. The wavelength-dependent g_{MCD} of *D-2* and *L-2*.

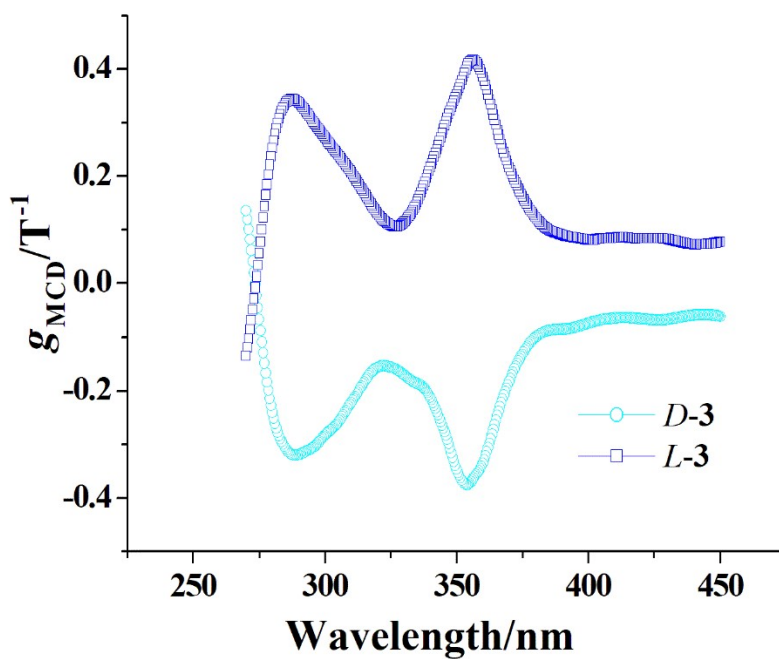


Fig. S24. The wavelength-dependent g_{MCD} of *D-3* and *L-3*.

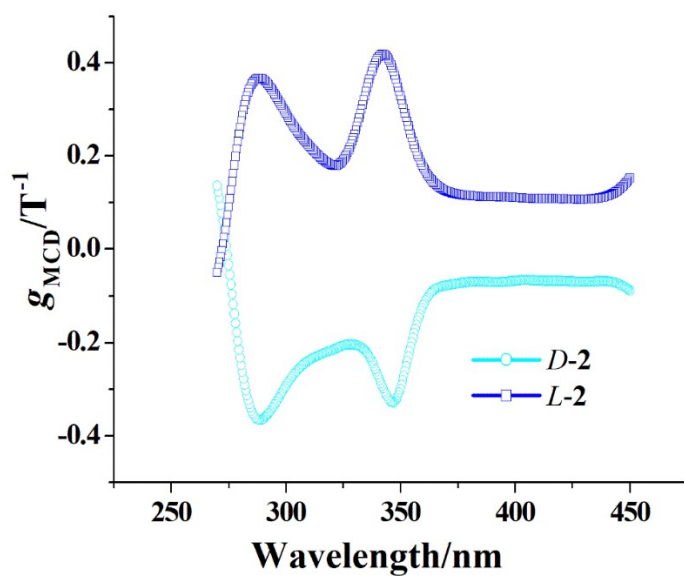


Fig. S25. The wavelength-dependent g_{MCD} of *D-4* and *L-4*.

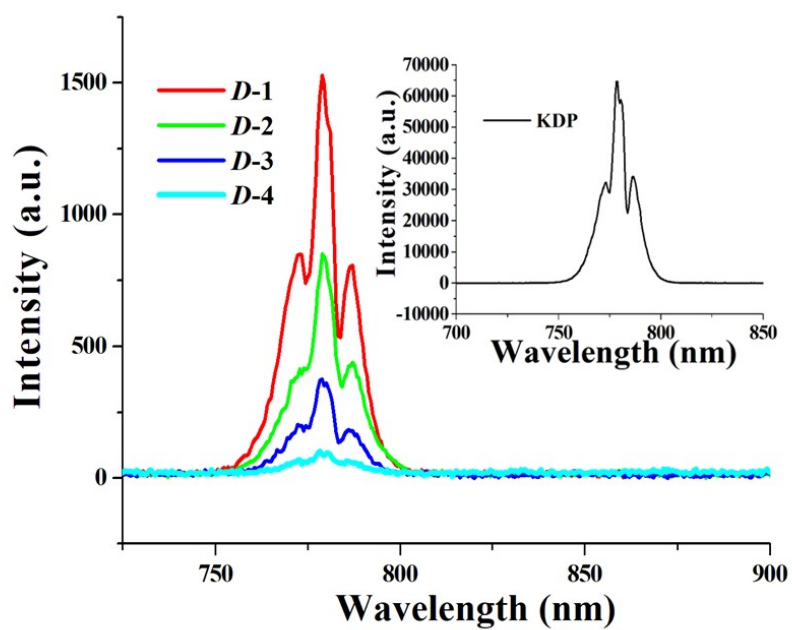


Fig. S26. SHG spectra of crystalline samples of *D-1*, *D-2*, *D-3*, *D-4* and KDP (illustration) under

excitation at $\lambda = 1550$ nm ($T_{\text{int}} = 0.5$ s).