Supporting Information

Structural cutting and recombining in a layered sodium dysprosium phosphonate: key roles of flexible pyrazinyl hydrazone molecular tools†

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Table S1. Selected bond lengths (Å) and angles (º) for **1**.

Compound 1							
Dy1–O1	2.334(9)	Dy6···Na3	3.7346(1)	Dy5···Na3	3.7524(1)	Dy3···Na1	9.9315(3)
Dy1–O2	2.247(10)	Dy6···Na3	7.8328(3)	Dy5···Na1	7.3023(2)	Dy4···Na5	3.9729(1)
Dy1–O13	2.378(8)	Dy2–N5	3.1187(2)	Dy6···Na1	3.7841(1)	Dy4···Na3	9.4972(3)
Dy1–O19	2.493(8)	Dy3–O4	2.347(9)	Dy6···Na4	8.1641(3)	Dy5···Dy6	3.9638(1)
Dy1–O20	2.288(9)	Dy3–O5	2.274(10)	Dy4–N16	2.505(12)	Dy5···Na2	9.2912(3)
Dy1–O22	2.303(9)	Dy3–O7	2.340(8)	Dy5–O16	2.392(9)	Dy6···Na5	6.5693(2)
Dy1–N4	2.514(13)	Dy3–O26	2.271(9)	Dy5–O21	2.343(10)	Na1···Na3	7.3825(3)
Dy1–N17	2.532(11)	Dy3–O27	2.462(8)	Dy5–O23	2.460(9)	Na1···Na2	9.0417(3)
Dy2–O1	2.828(9)	Dy3–O28	2.342(10)	Dy5–O24	2.461(9)	Na2···Na4	7.0537(2)
Dy2–O4	2.758(9)	Dy3–N8	2.561(10)	Dy5–O30	2.196(9)	Na3···Na4	9.5088(3)
Dy2–O20	3.1070(1)	Dy3–N9	2.596(11)	Dy5–O33	2.366(10)	Na1···Na3	9.0770(3)
Dy2–O22	2.9914(1)	Dy4–O7	2.484(8)	Dy5–N21	2.598(13)	Na1···Na5	9.4702(3)
Dy2–O23	2.8837(2)	Dy4–O8	2.250(10)	Dy6–O13	2.487(8)	Na3···Na5	6.4419(2)
Dy2–O26	3.0751(3)	Dy4–O10	2.393(9)	Dy6–O14	2.276(8)	Na2···Na5	7.1192(2)
Dy2–O28	2.8712(1)	Dy4–O11	2.259(10)	Dy6–O16	2.416(7)	Na4···Na5	3.0644(1)
Dy2–N1	3.1168(2)	Dy4–O27	2.380(8)	Dy6–O17	2.239(10)	Dy1–O1–Dy2	103.2(3)
Na1–O2	2.415(11)	Dy4–O29	2.298(9)	Dy6–O19	2.323(8)	Dy1–O2–Na1	105.1(4)
Na1–O3	2.375(13)	Dy4–N12	2.570(11)	Dy6–O24	2.348(8)	Dy3–O7–Na2	99.9(3)
Na1–O13	2.506(11)	Na3–O36	2.378(19)	Dy6–N20	2.557(13)	Dy4–O10–Na4	89.2(4)
Na1–O17	2.319(11)	Na4–O8	2.397(12)	Dy6–N24	2.556(11)	Dy4–O11–Na2	108.7(4)
Na1–O18	2.519(12)	Na4–O10	2.490(13)	P2–O23	1.524(10)	Dy6–O13–Na1	98.6(3)
Na1–O34	2.292(12)	Na4–O29	2.453(11)	P2–O24	1.531(9)	Dy5–O16–Na3	100.5(4)
Na2–O5	2.380(11)	Na4–O31	2.600(15)	P2–C89	1.789(15)	Dy1–O19–Dy6	114.0(4)
Na2–O6	2.335(13)	Na4–O37	2.69(3)	P3–O25	1.459(10)	Dy3–O27–Dy4	111.6(3)
Na2–O7	2.535(10)	Na5–O10	2.291(12)	P3–O26	1.555(11)	Dy5–O31–Na5	97.0(5)
Na2–O11	2.299(11)	Na5–O23	2.302(12)	P3–O27	1.540(8)	Dy2–O4–Dy3	102.8(3)
Na2–O12	2.400(13)	Na5–O25	2.543(13)	P3–C99	1.878(19)	Dy4–O7–Na2	95.1(3)
Na2–O35	2.295(12)	Na5–O31	2.127(14)	P4–O28	1.521(11)	Dy4–O10–Na5	116.0(4)
Na3–O14	2.401(11)	Na5–N13	2.500(16)	P4–O29	1.516(9)	Dy1–O13–Dy6	112.3(3)
Na3–O15	2.235(12)	P1–O19	1.545(10)	P4–O30	1.526(10)	Dy6–O14–Na3	105.9(4)
Na3–O16	2.488(9)	P1–O20	1.522(11)	P4–C109	1.809(15)	Dy6–O16–Na3	99.2(3)
Na3–O33	2.238(14)	P1–O21	1.502(10)	P5–O31	1.540(13)	Dy5–O23–Na5	90.6(3)
Dy1···Dy6	4.0403(1)	P1–C79	1.785(15)	P5–O32	1.458(14)	Dy4–O29–Na4	92.3(4)
Dy1···Na5	6.8952(2)	P2–O22	1.514(9)	P5–O33	1.548(12)	Na4–O31–Na5	80.2(5)
Dy1···Na4	9.3144(3)	Dy1···Dy2	4.0562(1)	P5–C119	1.856(18)	Dy3–O5–Na2	106.7(4)
Dy2···Dy5	4.2016(1)	Dy1···Na3	7.3664(3)	Dy1···Na1	3.7016(1)	Dy4–O8–Na4	95.0(4)
Dy2···Dy4	6.1982(2)	Dy1···Na2	9.9431(3)	Dy1···Dy5	5.4053(2)	Na4–O10–Na5	79.6(4)
Dy2···Na2	7.5940(2)	Dy2···Na5	4.2297(1)	Dy1···Dy3	8.0166(2)	Dy1–O13–Na1	98.6(3)
Dy3···Dy4	4.0040(1)	Dy2···Na4	6.5205(2)	Dy2···Dy3	3.9970(1)	Dy5–O16–Dy6	111.1(3)
Dy3···Dy5	6.6637(2)	Dy2···Na3	7.7451(2)	Dy2···Dy6	6.0670(2)	Dy6–O17–Na1	112.2(5)
Dy4···Na4	3.4268(1)	Dy3···Na5	5.1605(2)	Dy2···Na1	7.5794(2)	Dy5–O24–Dy6	111.0(3)
Dy4···Dy5	6.3877(2)	Dy3···Dy6	9.7190(3)	Dy3···Na2	3.7341(1)	Dy5–O31–Na4	124.3(5)
Dy5···Na5	3.3855(1)	Dy4···Na2	3.7043(1)	Dy3···Na4	6.3243(2)	Dy5–O33–Na3	109.1(5)
Dy5···Na4	4.4111(1)	Dy4···Na4	9.4243(3)				

Table S2. Selected bond lengths (Å) and angles (º) for **2**.

Compound 2							
Dy1–O20	2.293(13)	Dy4–O46	2.405(15)	P3–O39	1.522(16)	Na1–O43	2.60(2)
Dy1–O22	2.327(13)	Dy4–O47	2.383(13)	P3–O40	1.49(2)	Na1–O49	2.39(3)
Dy1–O27	2.314(17)	Dy5–O17	2.33(2)	P3–C22	1.87(2)	Na2–O7	2.37(2)
Dy1–O28	2.290(16)	Dy5–O18	2.280(14)	P4–O22	1.522(15)	Na2–O8	2.513(17)
Dy1–O44	2.421(16)	Dy5–O28	2.465(15)	P4–O23	1.484(17)	Na2–O29	2.493(18)
Dy1–O46	2.332(14)	Dy5–O29	2.408(14)	P4–O24	1.528(14)	Na2–O31	2.711(18)
Dy1–O47	2.334(14)	Dy5–O31	2.237(14)	P4–C32	1.810(16)	Na2–O33	2.520(19)
Dy2–O9	2.456(15)	Dy5–O36	2.259(14)	P5–O35	1.533(15)	Na2–O34	2.38(3)
Dy2–O10	2.267(17)	Dy5–O47	2.431(14)	P5–O36	1.510(17)	Na2–N6b	2.72(2)
Dy2–O12	2.382(17)	Dy6–O3	2.470(17)	P5–O37	1.479(17)	Na3–O4	2.41(2)
Dy2–O24	2.290(14)	Dy6–O5	2.264(17)	Na3–N2d	2.67(2)	Na4–O15	2.64(2)
Dy2–O39	2.230(14)	Dy6–O6	2.40(2)	Na4–O10	2.430(17)	Na4–O23	2.692(17)
Dy2–O48	2.472(12)	Dy6–O19	2.280(15)	Na4–O11	2.36(2)	Na4–O24	2.594(19)
Dy2–N1	2.601(16)	Dy6–O21	2.473(15)	Na4–O40	2.49(2)	Dy7–O15–Na4	86.6(7)
Dy2–N8	2.501(17)	Dy6–O37	2.247(14)	Dy1···Dy2	5.7425(13)	Dy6–O6–Dy8	104.2(7)
Dy3–O8	2.230(17)	Dy6–N11	2.645(19)	Dy1···Dy3	5.7731(13)	Dy6–O19–Na3	96.2(6)
Dy3–O9	2.370(16)	Dy6–N15	2.49(2)	Dy1···Dy4	3.8799(13)	Dy6–O21–Dy8	98.7(4)
Dy3–O12	2.433(15)	Dy7–O14	2.66(2)	Dy1···Dy5	3.8094(13)	Dy5–O36–Na3	96.6(6)
Dy3–O30	2.258(13)	Dy7–O15	2.43(2)	Dy1···Dy6	5.8121(14)	Dy7–O40–Na4	94.0(6)
Dy3–O33	2.310(15)	Dy7–O23	2.327(14)	Dy1···Dy7	3.8596(14)	Dy2–O9–Dy3	103.2(5)
Dy3–O48	2.472(12)	Dy7–O26	2.426(16)	Dy1···Dy8	5.8359(14)	Dy2–O10–Na4	99.1(6)
Dy3–N4	2.472(17)	Dy7–O27	2.480(15)	Dy2···Dy3	3.7839(13)	Dy2–O12–Dy3	103.6(5)
Dy3–N5	2.609(17)	Dy7–O40	2.271(16)	Dy2···Dy4	5.6291(13)	Dy7–O14–Na1	88.2(8)
Dy4–O32	2.274(14)	Dy7–O43	2.254(16)	Dy2···Dy5	7.3551(18)	Dy6–O5–Na3	98.6(6)
Dy4–O35	2.312(16)	Dy7–O46	2.494(17)	Dy2···Dy6	9.3316(16)	Dy5–O18–Na3	94.9(6)
Dy4–O38	2.273(16)	Dy8–O2	2.22(2)	Dy2···Dy7	5.4327(13)	Dy3–O33–Na2	96.0(6)
Dy4–O41	2.257(16)	Dy8–O3	2.344(16)	Dy3···Dy4	6.0339(13)	Dy7–O23–Na4	87.7(5)
Dy4–O45	2.347(16)	Dy8–O6	2.39(2)	Dy3···Dy5	5.4486(14)	Dy2–O24–Na4	94.0(5)
Dy8–O21	2.505(16)	P6–C52	1.839(16)	Dy3···Dy7	7.6393(18)	Dy7–O26–Na1	96.4(7)
Dy8–O25	2.268(17)	P7–O41	1.54(2)	Dy4···Dy5	4.2939(13)	Dy1–O27–Dy7	107.2(5)
Dy8–O42	2.217(17)	P7–O42	1.506(17)	Dy4···Dy6	5.6412(14)	Dy1–O28–Dy5	106.4(5)
Dy8–N9	2.43(2)	P7–O43	1.48(2)	Dy4···Dy7	4.3789(13)	Dy5–O29–Na2	103.6(6)
Dy8–N12	2.62(3)	P7–C62	1.767(18)	Dy4···Dy8	5.9576(14)	Dy1–O46–Dy7	106.2(5)
P1–O31	1.498(15)	P8–O25	1.490(15)	Dy5···Dy6	5.4598(14)	Dy5–O31–Na2	101.8(6)
P1–O32	1.540(19)	P8–O26	1.553(19)	Dy5···Dy7	7.1309(18)	Dy1–O47–Dy5	106.1(5)
P1–O33	1.495(14)	P8–O27	1.543(16)	Dy5···Dy8	7.6029(18)	Dy8–O42–Na1	95.2(7)
P1–C1	1.803(19)	P8–C72	1.843(18)	Dy6···Dy7	7.3901(18)	Dy7–O43–Na1	96.7(7)
P2–O28	1.568(16)	Na1–O1	2.44(3)	Dy6···Dy8	3.7774(18)	Dy1–O46–Dy4	110.0(6)
P2–O29	1.521(18)	Na1–O2	2.48(2)	Dy7···Dy8	5.3935(16)	Dy4–O46–Dy7	126.7(6)
P2–O30	1.500(14)	Na1–O14	2.56(2)	Dy3–O8–C93	137.9(16)	Dy1–O47–Dy4	110.7(6)
P2–C11	1.817(17)	Na1–O26	2.44(2)	Dy8–O2–Na1	100.8(8)	Dy4–O47–Dy5	126.3(6)
P3–O38	1.515(16)	Na1–O42	2.68(2)	Dy6–O3–Dy8	103.4(6)	Dy2–O48–Dy3	99.9(3)

Table S3. Hydrogen bonds in **1**.

Compound 1				
D–H···A	d(D–H) (Å)	d(H···A) (Å)	d(D···A) (Å)	<(DHA) (°)
O34–H34b···N10	0.8500	2.1500	2.81(2)	134.00
O34–H34c···N2	0.8500	2.5200	2.944(16)	112.00
O35–H35b···N18	0.8500	2.5600	2.851(19)	101.00
O35–H35c···N6	0.8500	2.1700	2.912(17)	145.00
O37–H37b···O32	0.8500	2.3000	2.77(3)	115.00
C1–H1···O26	0.9300	2.3800	3.081(19)	132.00
C129–H12a···O36	0.9600	1.9600	2.78(5)	142.00
C129–H12c···O32	0.9600	2.0200	2.71(5)	126.00
C14–H14···O20	0.9300	2.3100	3.01(2)	132.00
C42–H42···N15	0.9300	2.4400	2.77(2)	101.00
C55–H55···N19	0.9300	2.4900	2.80(2)	100.00
C80–H80···O21	0.9300	2.4900	2.899(19)	107.00
C82–H82···N15	0.9300	2.6100	3.36(3)	139.00
C87–H87···O19	0.9300	2.4400	3.016(19)	120.00
C90–H90···O24	0.9300	2.5200	2.95(2)	108.00
C97–H97···O22	0.9300	2.2700	2.96(2)	131.00
C100–H100···O27	0.9300	2.3800	2.87(2)	112.00
C110–H110···O29	0.9300	2.5500	2.96(2)	107.00
C117–H117···O28	0.9300	2.3700	3.07(2)	132.00
C120–H120···O33	0.9300	2.5200	2.94(2)	108.00
C127–H127···O32	0.9300	2.3700	3.04(3)	130.00

Symmetry codes: (a) x, y, -1+z; (b) -x, -y, -z; (c) 1+x, y, z.

Table S4. Hydrogen bonds in **2**.

Compound 2				
D–H···A	d(D–H) (Å)	d(H···A) (Å)	d(D···A) (Å)	<(DHA) (°)
O13–H13b···N16	0.8500	2.2600	2.82(3)	124.00
O34–H34a···O17	0.8500	2.2200	2.78(3)	124.00
C2–H2···O31	0.9300	2.4200	2.86(2)	109.00
C12–H12···O30	0.9300	2.4600	2.89(2)	108.00
C120–H12a···O16	0.9600	2.4000	3.21(4)	141.00
C17–H17···O29	0.9300	2.2500	3.00(3)	137.00
C23–H23···O39	0.9300	2.5400	2.92(3)	104.00
C28–H28···O40	0.9300	2.2800	3.05(3)	140.00
C33–H33···O23	0.9300	2.3600	2.84(2)	112.00
C40–H40···O9	0.9300	2.6000	3.36(2)	139.00
C43–H43···O36	0.9300	2.4500	2.86(3)	107.00
C50–H50···O37	0.9300	2.3900	3.13(3)	136.00
C53–H53···O18	0.9300	2.4800	2.92(2)	109.00
C60–H60···O20	0.9300	2.5500	3.18(2)	125.00
C63–H63···O43	0.9300	2.4000	2.85(3)	110.00
C73–H73···O27	0.9300	2.5100	2.96(2)	110.00
C8–H80···O25	0.9300	2.4000	3.01(3)	123.00
C82–H82···O10	0.9300	2.4900	3.07(2)	121.00
C94–H94a···O34	0.9600	2.3900	3.15(4)	135.00
C96–H96···O33	0.9300	2.5400	3.27(3)	136.00
C121–H121···O5	0.9300	2.5800	3.14(3)	119.00

Symmetry codes: (a) -x, 2-y, -z; (b) x, -1+y, z.

Table S5. Dy^{III} and Na^{I} geometry analysis of **1** by SHAPE 2.1 software.

Dy							
Geometry (CN = 8)	**Dy1**	**Dy3**	**Dy4**	**Dy5**	**Dy6**	**Geometry (CN = 10)**	**Dy2**
TDD-8	3.061	2.881	3.125	2.712	2.573	**PARP-10**	3.431
BTPR-8	3.110	3.090	2.123	2.679	2.472	**SDD-10**	5.613
JSD-8	3.485	3.538	3.282	5.988	2.319	**HD-10**	6.591
JBTPR-8	4.248	4.312	2.670	3.571	3.149	**TD-10**	7.416
SAPR-8	5.341	4.772	3.942	4.377	5.007	**JMBIC-10**	7.547
JGBF-8	9.787	9.786	10.082	12.443	9.223	**JBCCU-10**	9.655
CU-8	11.616	12.003	9.334	10.367	12.614	**PPR-10**	9.834
TT-8	12.189	12.582	9.774	10.998	13.262	**JSPC-10**	12.936
HBPY-8	12.885	12.723	12.507	13.921	14.016	**JBCSAPR-10**	13.388
ETBPY-8	21.041	21.636	20.191	21.739	20.115	**JATDI-10**	14.600
JETBPY-8	25.982	26.470	24.577	24.200	24.309	**OBPY-10**	15.740
HPY-8	24.390	25.016	24.114	21.173	24.088	**EPY-10**	24.156
OP-8	31.937	30.971	31.950	34.252	32.016	**DP-10**	25.993

Na							
Geometry (CN = 6)	**Na1**	**Na2**	**Na5**	**Geometry (CN =5)**	**Na3**	**Geometry (CN = 7)**	**Na4**
HP-6	26.359	27.495	34.288	**PP-5**	18.038	**HP-7**	21.108
PPY-6	12.386	12.507	16.956	**vOC-5**	5.122	**HPY-7**	15.043
OC-6	8.447	8.638	7.886	**TBPY-5**	8.825	**PBPY-7**	7.626
TPR-6	8.871	7.109	8.880	**SPY-5**	5.402	**COC-7**	7.270
JPPY-6	15.606	15.995	20.965	**JTBPY-5**	11.374	**CTPR-7**	4.875
						JPBPY-7	10.479
						JETPY-7	15.860

Lable	Shape	Lable	Shape	Lable	Shape	Lable	Shape	Lable	Shape
PP-5	Pentagon (D_{5h})	JPPY-6	Johnson pentagonal pyramid (C_{5V})	HPY-8	Heptagonal pyramid (C_{7v})	JSD-8	Snub diphenoid J84 (D_{2d})	JMBIC-10	Metabidiminished icosahedron (J62) (C_{2v})
vOC-5	Vacant octahedron‡ (Johnson square pyramid, J1) (C_{4v})	HP-7	Heptagon	HBPY-8	Hexagonal bipyramid (D_{6h})	TT-8	Triakis tetrahedron (T_d)	JATDI-10	Augmented tridiminished icosahedron (J64) (C_{3v})
TBPY-5	Trigonal bipyramid (D_{3h})	HPY-7	Hexagonal pyramid (C_{6v})	CU-8	Cube (O_h)	DP-10	Decagon (D_{10h})	JSPC-10	Sphenocorona (J87) (C_{2v})
SPY-5	Square pyramid§ (C_{4v})	PBPY-7	Pentagonal bipyramid (D_{5h})	SAPR-8	Square antiprism (D_{4d})	EPY-10	Enneagonal pyramid (C_{9v})	SDD-10	Staggered dodecahedron (2:6:2) # (D_2)
JTBPY-5	Johnson trigonal bipyramid (J12) (D_{3h})	COC-7	Capped octahedron (C_{3v})	TDD-8	Triangular dodecahedron (D_{2d})	OBPY-10	Octagonal bipyramid (D_{8h})	TD-10	Tetradecahedron (2:6:2) (C_{3v})
HP-6	Hexagon (D_{6h})	CTPR-7	Capped trigonal prism (C_{2v})	JGBF-8	Johnson gyrobifastigium J26 (D_{2d})	PPR-10	Pentagonal prism (D_{5h})	HD-10	Hexadecahedron (2:6:2, or 1:4:4:1) (D_{4h})
PPY-6	Pentagonal pyramid (C_{5V})	JPBPY-7	Johnson pentagonal bipyramid J13 (D_{5h})	JETBPY-8	Johnson elongated triangular bipyramid J14 (D_{3h})	PAPR-10	Pentagonal antiprism (D_{5d})		
OC-6	Octahedron (O_h)	JETPY-7	Elongated triangular pyramid J7 (C_{3v})	JBTPR-8	Biaugmented trigonal prism J50 (C_{2v})	JBCCU-10	Bicapped cube (Elongated square bipyramid J15) (D_{4h})		
TPR-6	Trigonal prism (D_{3h})	OP-8	Octagon (D_{8h})	BTPR-8	Biaugmented trigonal prism (C_{2v})	JBCSAPR-10	Bicapped square antiprism (Gyroelongated square bipyramid J17) (D_{4d})		

Table S6. Dy^{III} and Na^{I} geometry analysis of **2** by SHAPE 2.1 software.

Dy									
Geometry (CN = 7)	**Dy1**	**Dy4**	**Dy5**	**Geometry (CN = 8)**	**Dy2**	**Dy3**	**Dy6**	**Dy7**	**Dy8**
HP-7	35.140	33.262	30.045	**OP-8**	32.863	34.037	32.526	29.539	32.257
HPY-7	19.787	23.957	22.079	**HPY-8**	23.889	23.451	23.807	20.769	23.056
PBPY-7	4.526	0.358	2.156	**HBPY-8**	15.735	15.840	15.359	10.740	15.858
COC-7	2.285	7.096	3.866	**CU-8**	12.397	12.223	12.212	12.137	11.049
CTPR-7	1.462	5.262	2.228	**SAPR-8**	3.183	3.668	3.146	6.316	3.462
JPBPY-7	8.185	3.336	4.986	**TDD-8**	2.379	2.515	2.184	5.059	1.990
JETPY-7	20.286	22.968	20.255	**JGBF-8**	14.038	13.728	13.488	7.332	13.413
				JETBPY-8	28.107	28.080	28.195	20.574	27.329
				JBTPR-8	2.721	3.138	2.720	3.891	2.892
				BTPR-8	2.195	2.541	2.135	3.892	2.424
				JSD-8	4.441	4.662	4.161	4.432	4.067
				TT-8	13.141	13.012	12.903	12.695	11.606
				ETBPY-8	24.918	24.641	24.854	17.863	23.820

Na				
Geometry (CN = 7)	**Na1**	**Na2**	**Na3**	**Na4**
HP-7	30.889	31.509	32.008	29.778
HPY-7	15.241	21.702	21.498	15.571
PBPY-7	6.061	2.851	2.381	4.913
COC-7	5.580	5.065	5.748	5.567
CTPR-7	4.625	3.240	4.420	4.760
JPBPY-7	8.505	7.280	7.153	7.080
JETPY-7	16.747	19.396	20.232	16.235

Lable	**Shape**	**Lable**	**Shape**	**Lable**	**Shape**	**Lable**	**Shape**	**Lable**	**Shape**
HP-7	Heptagon	CTPR-7	Capped trigonal prism (C_{2v})	OP-8	Octagon (D_{8h})	SAPR-8	Square antiprism (D_{4d})	JBTPR-8	Biaugmented trigonal prism J50 (C_{2v})
HPY-7	Hexagonal pyramid (C_{6v})	JPBPY-7	Johnson pentagonal bipyramid J13 (D_{5h})	HPY-8	Heptagonal pyramid (C_{7v})	TDD-8	Triangular dodecahedron (D_{2d})	BTPR-8	Biaugmented trigonal prism (C_{2v})
PBPY-7	Pentagonal bipyramid (D_{5h})	JETPY-7	Elongated triangular pyramid J7 (C_{3v})	HBPY-8	Hexagonal bipyramid (D_{6h})	JGBF-8	Johnson gyrobifastigium J26 (D_{2d})	JSD-8	Snub diphenoid J84 (D_{2d})
COC-7	Capped octahedron (C_{3v})			CU-8	Cube (O_h)	JETBPY-8	Johnson elongated triangular bipyramid J14 (D_{3h})	TT-8	Triakis tetrahedron (T_d)

Table S7. Relaxation fitting parameters from least-squares fitting of $\chi(\omega)$ data under zero *dc* field for **1** (1.0-1000 Hz).

Ln(χ''/χ')									
T (k) / *V* (Hz)	**1.8**	**2.0**	**2.3**	**2.6**	**2.9**	**3.2**	**3.5**	**3.8**	**4.1**
1.000(5)	-5.049(9)	-5.183(5)	-5.263(1)	-5.337(5)	-5.392(8)	-5.429(8)	-5.483(2)	-5.521(4)	-5.546(1)
17.193(1)	-4.972(1)	-5.095(3)	-5.196(1)	-5.256(7)	-5.324(5)	-5.382(7)	-5.396(8)	-5.457(1)	-5.512(3)
21.046(6)	-4.851(4)	-4.989(2)	-5.090(6)	-5.162(8)	-5.203(7)	-5.279(4)	-5.314(1)	-5.356(4)	-5.371(2)
25.807(6)	-4.730(7)	-4.854(3)	-4.936(5)	-5.014(4)	-5.075(2)	-5.123(9)	-5.166(7)	-5.196(3)	-5.239(1)
31.624(4)	-4.608(9)	-4.728(7)	-4.809(6)	-4.876(2)	-4.939(8)	-4.979(1)	-5.010(6)	-5.067(2)	-5.096(8)
38.752(4)	-4.507(2)	-4.621(7)	-4.702(2)	-4.782(4)	-4.834(2)	-4.880(2)	-4.931(8)	-4.962(2)	-5.001(1)
47.498(1)	-4.436(7)	-4.549(9)	-4.643(6)	-4.717(8)	-4.766(5)	-4.811(1)	-4.869(6)	-4.899(5)	-4.927(1)
58.128(7)	-4.369(8)	-4.492(1)	-4.581(4)	-4.648(8)	-4.722(1)	-4.768(1)	-4.807(1)	-4.855(6)	-4.898(9)
71.178(1)	-4.302(2)	-4.423(6)	-4.513(1)	-4.589(7)	-4.649(4)	-4.703(1)	-4.754(1)	-4.792(1)	-4.839(1)
87.349(1)	-4.202(2)	-4.326(9)	-4.424(5)	-4.501(9)	-4.566(9)	-4.617(2)	-4.658(9)	-4.703(6)	-4.743(1)
107.079(2)	-4.095(6)	-4.216(8)	-4.314(4)	-4.390(6)	-4.452(5)	-4.509(4)	-4.549(1)	-4.591(1)	-4.626(8)
130.906(4)	-3.978(2)	-4.103(3)	-4.192(2)	-4.263(9)	-4.330(1)	-4.381(1)	-4.428(1)	-4.464(9)	-4.505(9)
160.618(8)	-3.860(7)	-3.982(1)	-4.070(7)	-4.138(5)	-4.201(8)	-4.257(1)	-4.292(9)	-4.333(2)	-4.366(2)
196.887(6)	-3.748(5)	-3.872(1)	-3.961(6)	-4.026(7)	-4.088(7)	-4.136(9)	-4.178(1)	-4.214(1)	-4.249(1)
241.126(5)	-3.647(1)	-3.772(4)	-3.856(4)	-3.923(8)	-3.985(4)	-4.037(3)	-4.074(5)	-4.110(4)	-4.141(7)
295.480(3)	-3.553(4)	-3.675(3)	-3.766(6)	-3.845(6)	-3.897(8)	-3.950(1)	-3.983(1)	-4.020(9)	-4.056(1)
362.360(8)	-3.462(8)	-3.586(2)	-3.683(6)	-3.753(9)	-3.809(9)	-3.864(8)	-3.906(7)	-3.938(2)	-3.972(6)
443.892(1)	-3.375(4)	-3.504(1)	-3.595(4)	-3.679(1)	-3.731(5)	-3.785(9)	-3.826(8)	-3.861(6)	-3.896(6)
544.045(9)	-3.287(3)	-3.422(8)	-3.518(3)	-3.594(6)	-3.658(5)	-3.708(4)	-3.750(7)	-3.784(9)	-3.817(2)
666.595(5)	-3.203(8)	-3.338(6)	-3.435(2)	-3.514(5)	-3.574(2)	-3.620(6)	-3.662(6)	-3.697(1)	-3.734(1)
815.501(1)	-3.129(4)	-3.266(1)	-3.366(5)	-3.449(9)	-3.513(2)	-3.567(5)	-3.607(2)	-3.635(9)	-3.682(1)
999.040(9)	-3.076(2)	-3.219(6)	-3.322(1)	-3.407(1)	-3.469(2)	-3.522(1)	-3.564(6)	-3.601(6)	-3.632(4)

Table S8. Relaxation fitting parameters from least-squares fitting of $\chi(\omega)$ data under zero *dc* field for **1** (10-10000 Hz).

T	χ_T	χ_s	α	τ / s
1.9	63.710(2)	36.288(3)	0.320(9)	1.995(9)E-5
2.5	60.250(7)	39.749(2)	0.299(7)	1.862(1)E-5
3.0	58.435(3)	41.564(6)	0.291(9)	1.707(1)E-5
3.5	56.968(1)	43.031(9)	0.282(9)	1.621(3)E-5
4.0	55.808(6)	44.191(3)	0.271(9)	1.586(6)E-5
4.5	55.190(7)	44.809(2)	0.287(6)	1.484(7)E-5
5.0	54.155(5)	45.844(4)	0.255(3)	1.416(2)E-5
7.0	51.908(5)	47.091(4)	0.242(7)	1.216(9)E-5
8.0	50.385(1)	48.614(8)	0.221(1)	1.081(2)E-5
9.0	48.834(5)	50.165(4)	0.206(8)	9.622(2)E-6
10.0	46.148(8)	52.148(8)	0.191(3)	9.046(1)E-6
11.0	44.690(7)	53.690(7)	0.179(1)	8.377(6)E-6
12.0	42.359(1)	54.640(9)	0.169(1)	7.375(1)E-6
13.0	40.385(4)	56.034(7)	0.151(6)	6.238(5)E-6
14.0	38.680(8)	58.059(2)	0.144(9)	5.411(7)E-6
15.0	36.561(5)	60.084(7)	0.128(4)	4.058(1)E-6
16.0	34.768(2)	61.809(2)	0.118(6)	3.112(8)E-6
17.0	32.757(3)	63.127(9)	0.106(9)	2.861(2)E-6
18.0	31.647(3)	64.182(4)	0.083(3)	2.346(1)E-6
19.0	30.013(6)	65.187(1)	0.073(6)	2.088(1)E-6
20.0	28.761(1)	66.152(8)	0.059(6)	1.954(4)E-6

Table S9. Relaxation fitting parameters from least-squares fitting of $\chi(\omega)$ data under zero *dc* field for **2** (1.0-1000 Hz).

T	χ_T	χ_s	α	τ / s
1.8	38.146(3)	23.853(6)	0.229(1)	1.185(1)E-4
2.0	36.780(1)	25.219(9)	0.219(1)	1.051(1)E-4
2.3	35.837(2)	26.162(8)	0.212(4)	8.950(7)E-5
2.6	35.139(7)	26.860(2)	0.207(1)	8.064(9)E-5
2.9	34.564(2)	27.435(7)	0.200(2)	7.532(4)E-5
3.2	34.238(8)	27.761(1)	0.202(9)	6.614(1)E-5
3.5	33.865(2)	28.134(7)	0.199(3)	6.272(9)E-5
4	33.533(6)	28.466(3)	0.194(3)	6.058(3)E-5
4.3	33.319(9)	28.680(1)	0.194(2)	5.617(5)E-5
4.6	32.927(5)	29.072(4)	0.189(8)	5.269(7)E-5
4.9	32.787(1)	29.212(9)	0.197(2)	4.491(1)E-5
5.5	32.600(3)	29.399(7)	0.199(5)	3.885(4)E-5
6.0	32.424(7)	29.575(2)	0.199(1)	3.422(4)E-5
6.5	32.239(1)	29.760(8)	0.196(5)	2.876(4)E-5
7.0	32.516(1)	29.483(9)	0.213(8)	2.222(1)E-5
7.5	32.206(6)	29.793(3)	0.198(3)	1.735(6)E-5
8.0	32.308(8)	29.691(1)	0.204(5)	1.397(2)E-5
8.5	32.316(6)	29.683(3)	0.203(3)	1.164(6)E-5
9.0	32.234(5)	29.765(4)	0.189(3)	9.352(8)E-6
10.0	31.719(7)	29.480(2)	0.126(5)	7.063(4)E-6
11.0	31.558(7)	29.241(2)	0.167(9)	6.014(5)E-6
12.0	31.492(5)	30.507(4)	0.099(9)	5.079(5)E-6
13.0	31.421(5)	30.578(4)	0.083(8)	4.659(6)E-6
14.0	35.771(4)	26.228(5)	0.115(3)	4.026(2)E-6
15.0	35.687(8)	26.312(1)	0.102(3)	3.530(7)E-6
16.0	35.783(5)	26.216(4)	0.071(9)	3.005(9)E-6
17.0	35.672(7)	26.327(2)	0.067(7)	2.665(2)E-6
18.0	35.457(1)	26.542(9)	0.076(6)	2.310(7)E-6
19.0	35.322(8)	26.677(2)	0.080(4)	2.036(1)E-6
20.0	35.733(3)	26.266(6)	0.038(5)	1.813(3)E-6

Table S10. Relaxation fitting parameters from least-squares fitting of $\chi(\omega)$ data under zero *dc* field for **2** (10-10000 Hz).

T	χ_T	χ_s	α	τ / s
1.9	69.004(4)	30.995(5)	0.312(1)	5.847(2)E-5
2.5	63.456(8)	36.543(1)	0.293(1)	3.911(3)E-5
3.0	60.665(1)	39.334(8)	0.283(7)	3.129(7)E-5
3.5	58.692(2)	41.307(7)	0.277(7)	2.649(7)E-5
4.0	57.146(2)	42.853(7)	0.265(1)	2.325(4)E-5
4.5	56.006(3)	43.993(7)	0.259(1)	2.049(3)E-5
5.0	55.230(3)	44.769(6)	0.266(1)	1.736(5)E-5
7.0	54.071(3)	45.928(6)	0.278(1)	1.218(6)E-5
8.0	53.060(1)	46.939(8)	0.268(4)	9.450(6)E-6
9.0	52.130(6)	47.869(3)	0.240(6)	8.342(4)E-6
10.0	51.584(2)	48.415(7)	0.234(2)	7.528(6)E-6
11.0	51.037(1)	48.962(9)	0.193(2)	6.331(5)E-6
12.0	51.693(8)	48.306(1)	0.312(6)	5.252(4)E-6
13.0	50.957(3)	49.042(6)	0.263(9)	3.874(5)E-6
14.0	51.494(7)	48.505(2)	0.289(5)	2.879(3)E-7
15.0	50.421(4)	49.578(5)	0.192(1)	2.429(4)E-6
16.0	50.275(2)	49.724(7)	0.192(9)	1.831(6)E-6
17.0	50.347(9)	49.652(1)	0.408(6)	1.360(2)E-6
18.0	50.309(7)	49.690(2)	0.669(5)	1.005(1)E-6
19.0	50.099(9)	49.900(1)	0.210(3)	8.169(1)E-7
20.0	50.052(5)	49.947(4)	0.193(3)	6.166(5)E-7

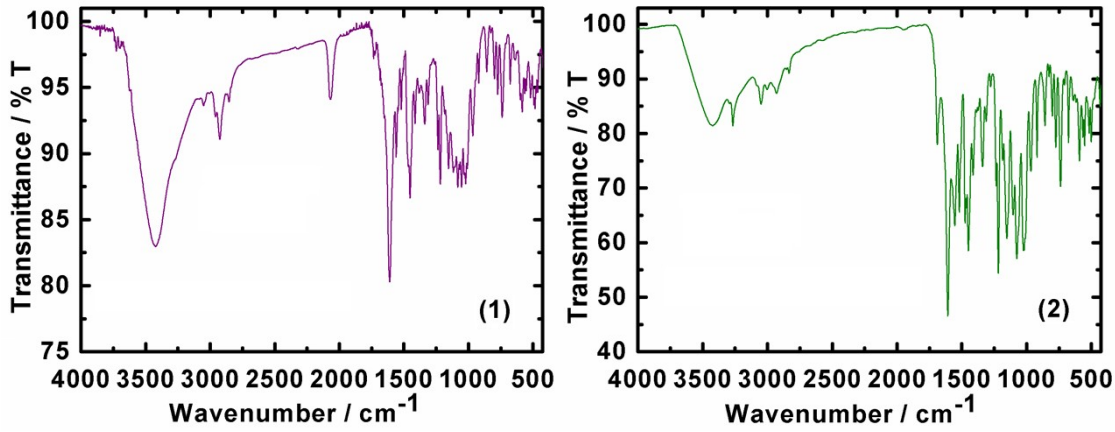


Figure S1. Infrared spectra of **1** (left) and **2** (right).

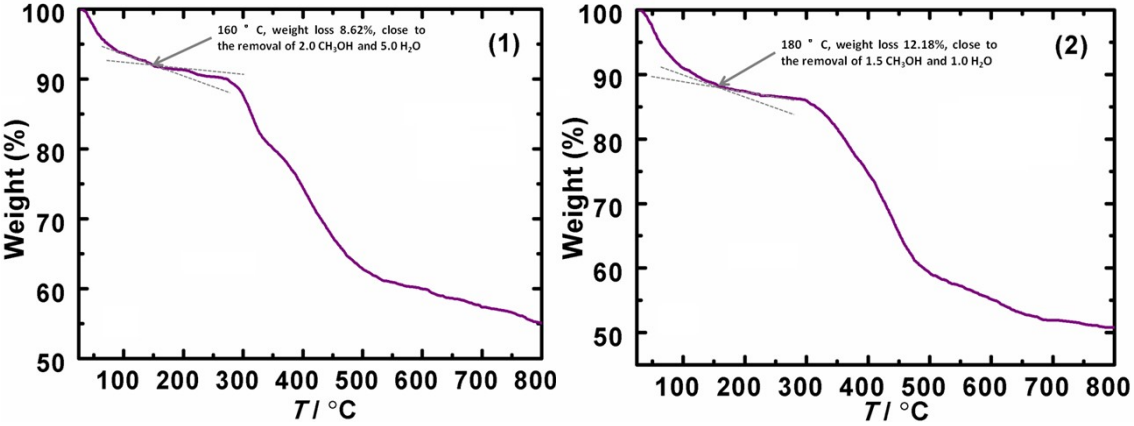


Figure S2. TG analyses of **1** (left) and **2** (right).

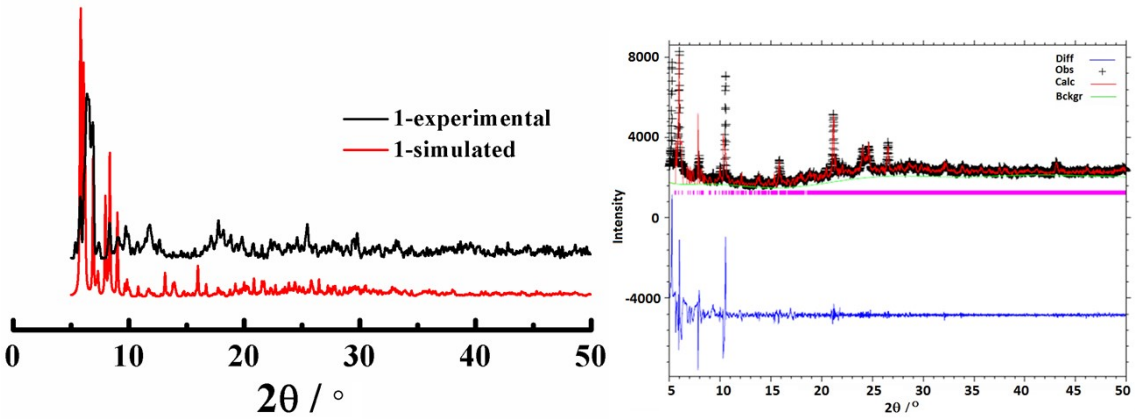


Figure S3. Left: the powder XRD patterns for compound **1**. Right: the black circles are for the observed data. The red solid line is for the calculated data. The grey solid curve is for the difference. The vertical bars are the positions of Bragg peaks. Cell parameters: *P*-1, a = 15.19 Å, b = 19.96 Å, c = 26.24 Å, α = 89.5°, β = 79.02°, γ = 84.28°, V = 7356.6 Å^3 (wRp = 0.114).

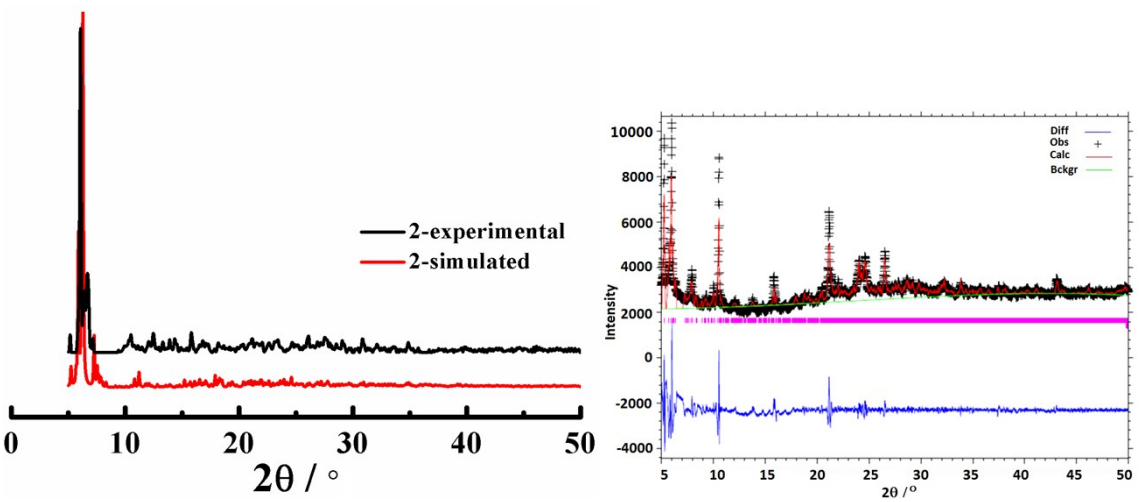
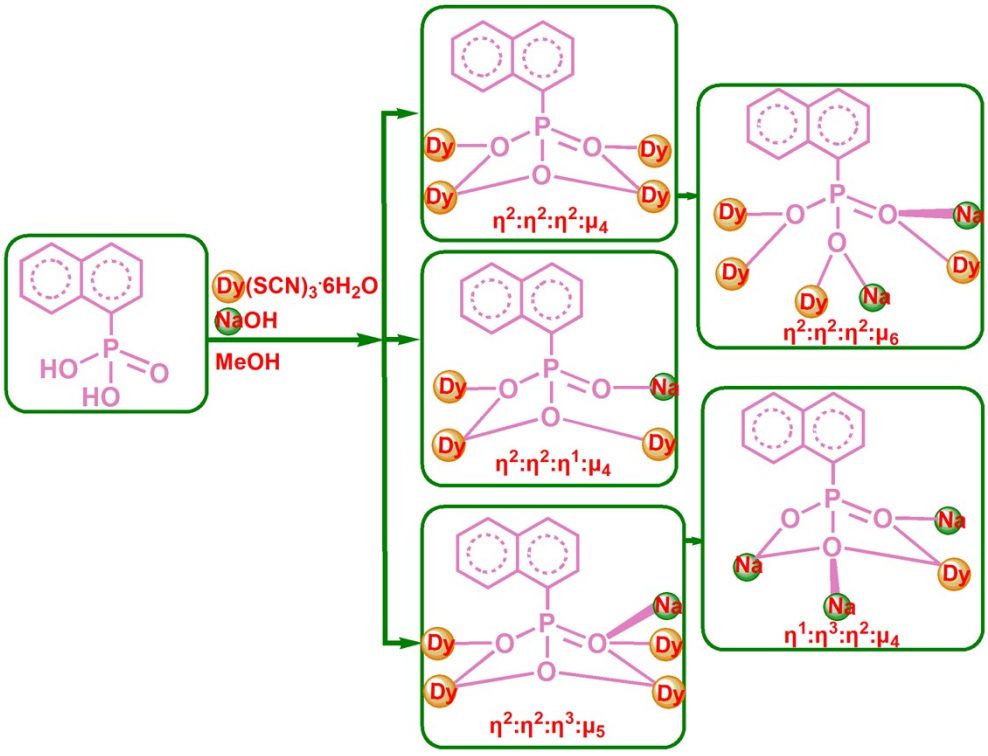
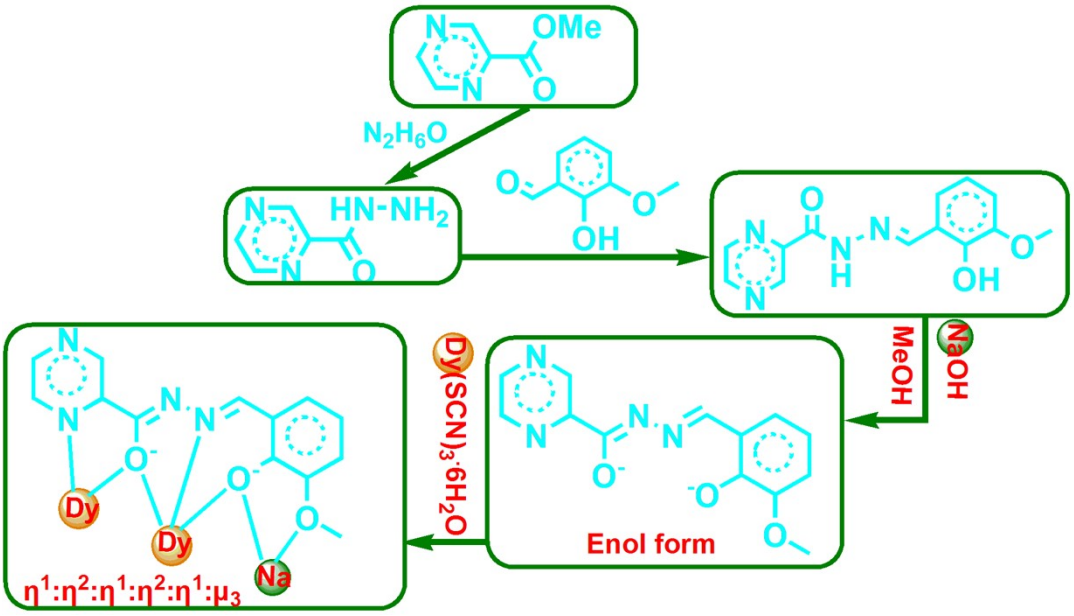


Figure S4. Left: the powder XRD patterns for compound **2**. Right: the black circles are for the observed data. The red solid line is for the calculated data. The grey solid curve is for the difference. The vertical bars are the positions of Bragg peaks. Cell parameters: $P2_1/c$, a = 32.55 Å, b = 16.45 Å, c = 33.81 Å, β = 113.8°, V = 16355.1 Å^3 (wRp = 0.069).



Scheme S1. H_2opch, di-deprotonated opch^{2-} and corresponding coordination mode in **1**.



Scheme S2. H_2opch, di-deprotonated opch^{2-} and corresponding coordination mode in **1**.

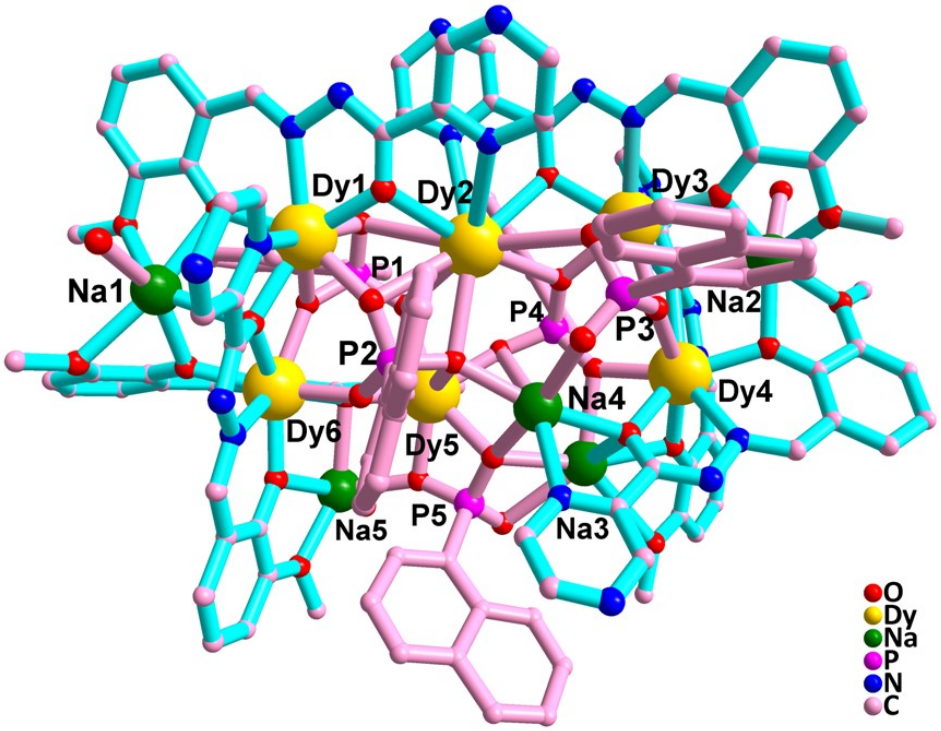


Figure S5. Top-view of **1**. Color scheme: the opch^{2-} fragments in turquoise, the $C_{10}H_7PO_3^{2-}$ fragments in rose, Dy in gold, O in red, N in blue, C in rose, P in pink.

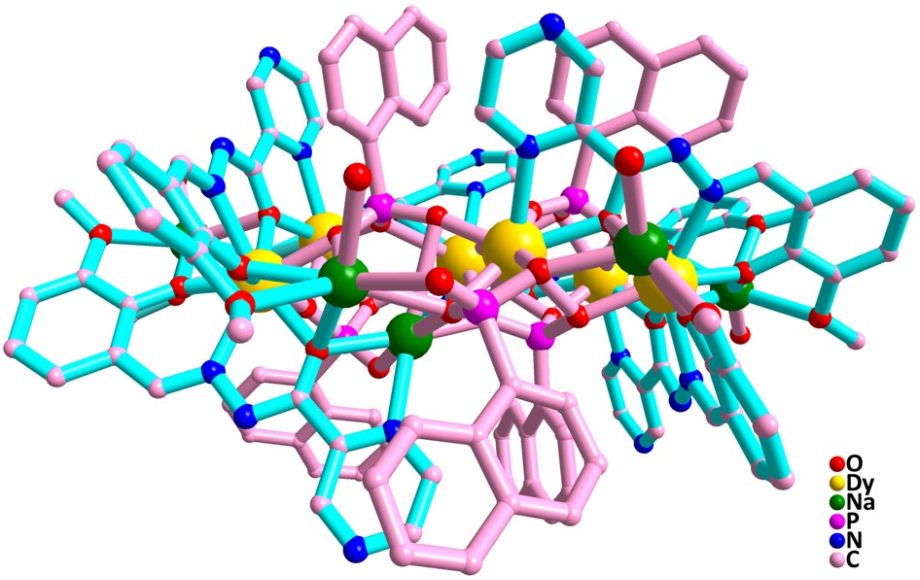


Figure S6. Side-view of **1**. Color scheme: the $opch^{2-}$ fragments in turquoise, the $C_{10}H_7PO_3^{2-}$ fragments in rose, Dy in gold, O in red, N in blue, C in rose, P in pink.

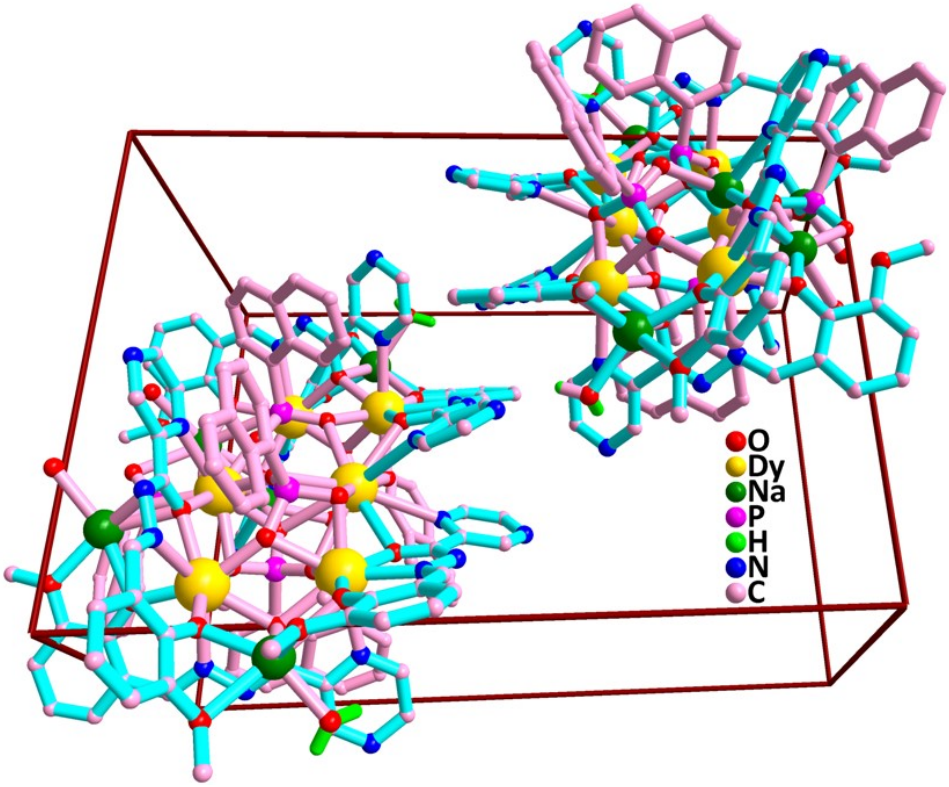


Figure S7. Unit cell of **1** showing the presence of two isomers.

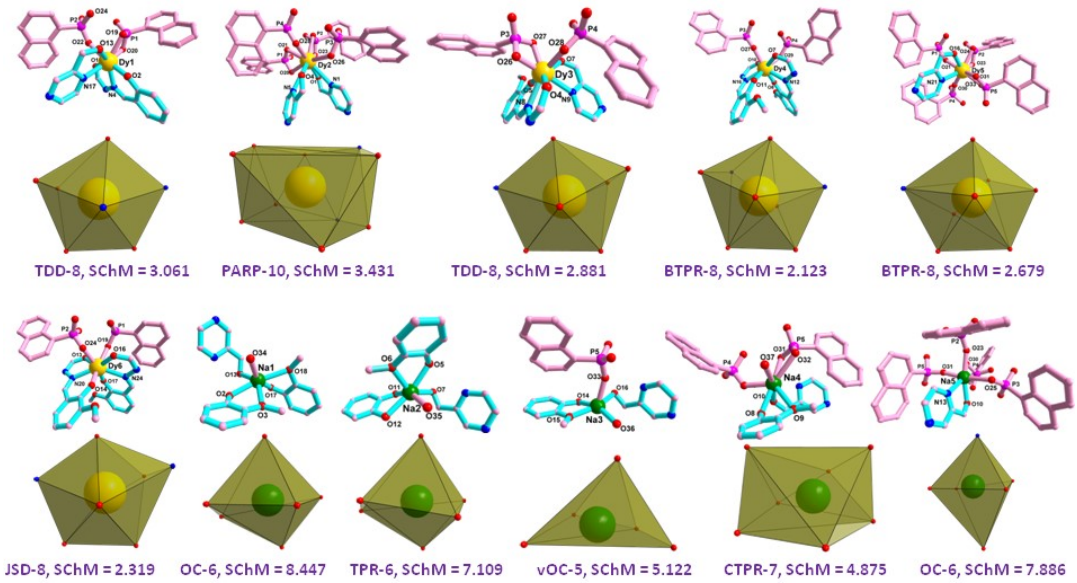


Figure S8. Coordination environments of six crystallographically independent Dy^{III} ions and five Na^{I} ions in **1**. Color scheme: the $opch^{2-}$ fragments in turquoise, the $C_{10}H_7PO_3^{2-}$ fragments in rose, Dy in gold, O in red, N in blue, C in rose, P in pink.

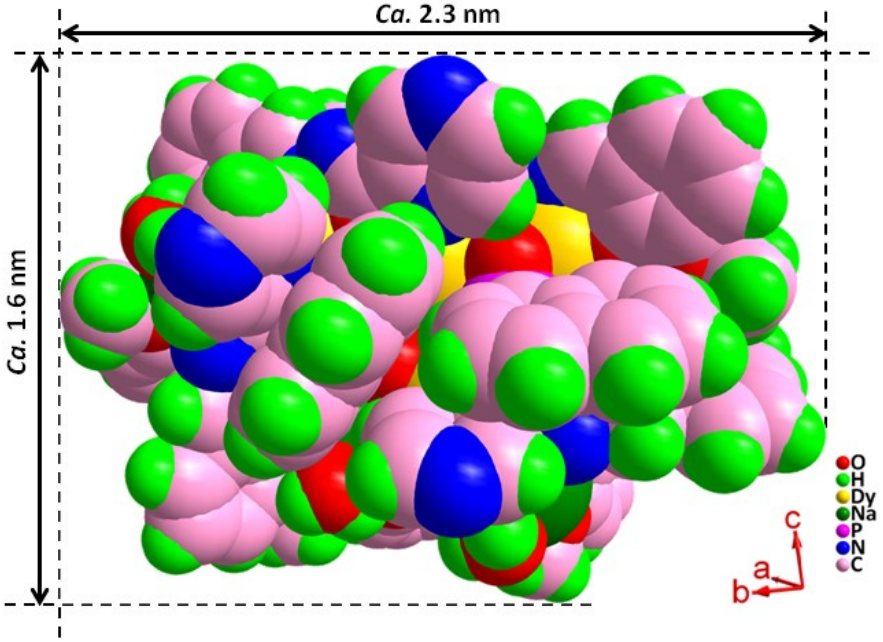


Figure S9. Space-filling representation of **1**. Color scheme: Dy in gold, Na in green, O in red, N in blue, P in pink, C in rose, H in bright green.

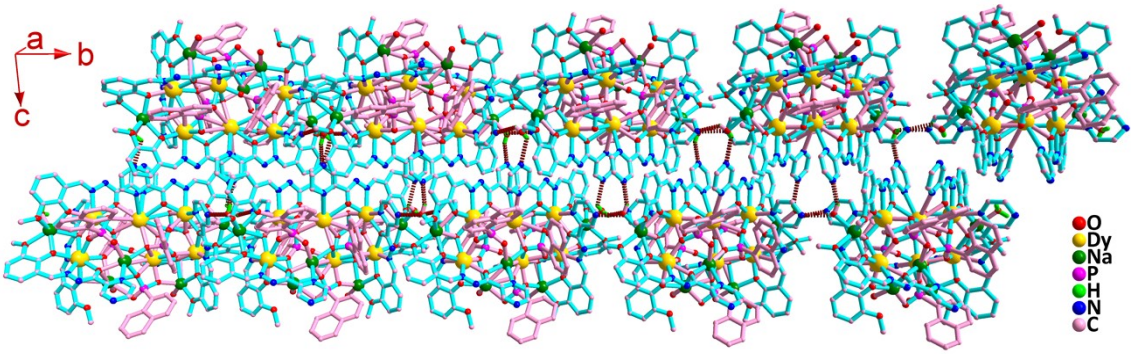
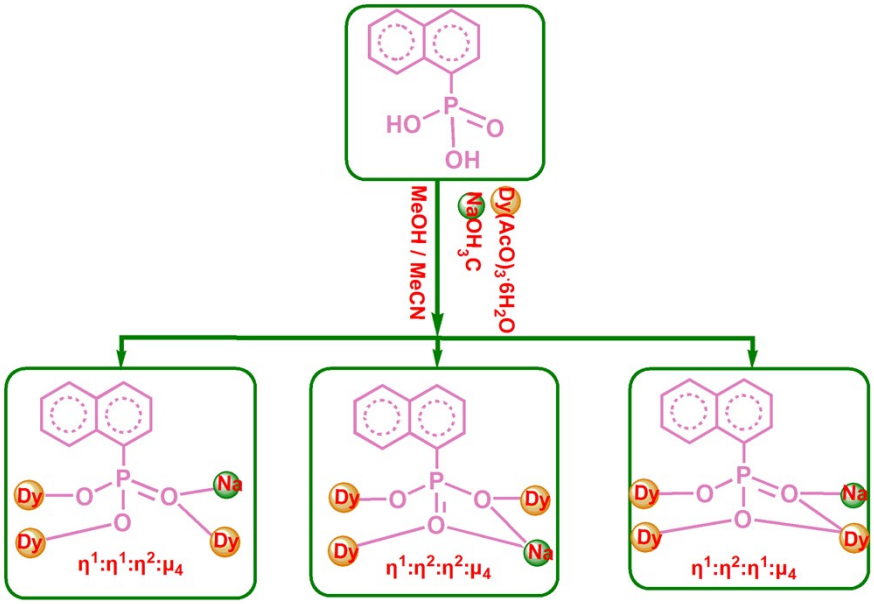
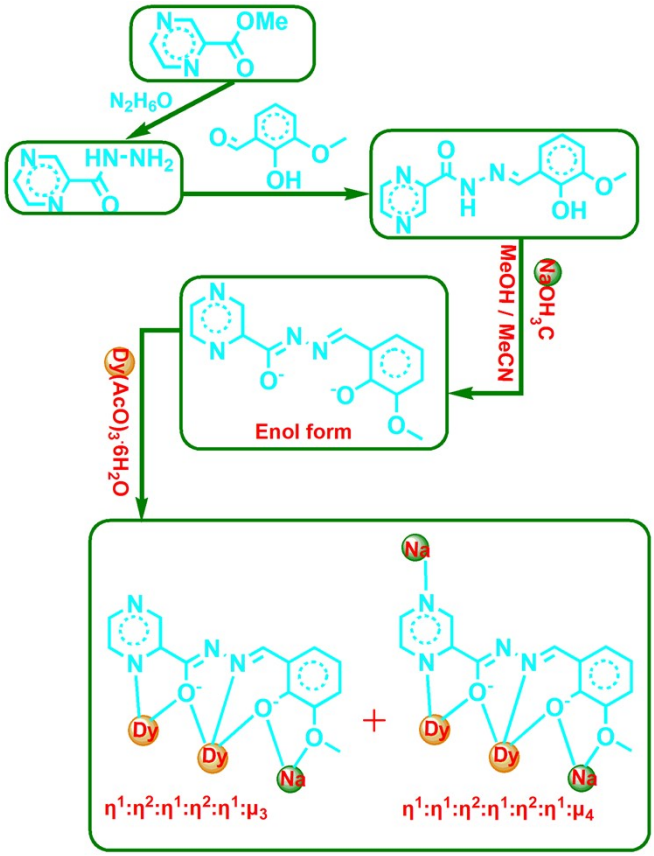


Figure S10. Illustration showing the hydrogen-bonding interactions (brown dash lines) and the double-chain supramolecular arrangement of the molecules in **1**.



Scheme S3. H_2opch, di-deprotonated opch^{2-} and corresponding coordination mode in **2**.



Scheme S4. H_2opch, di-deprotonated opch^{2-} and corresponding coordination mode in **2**.

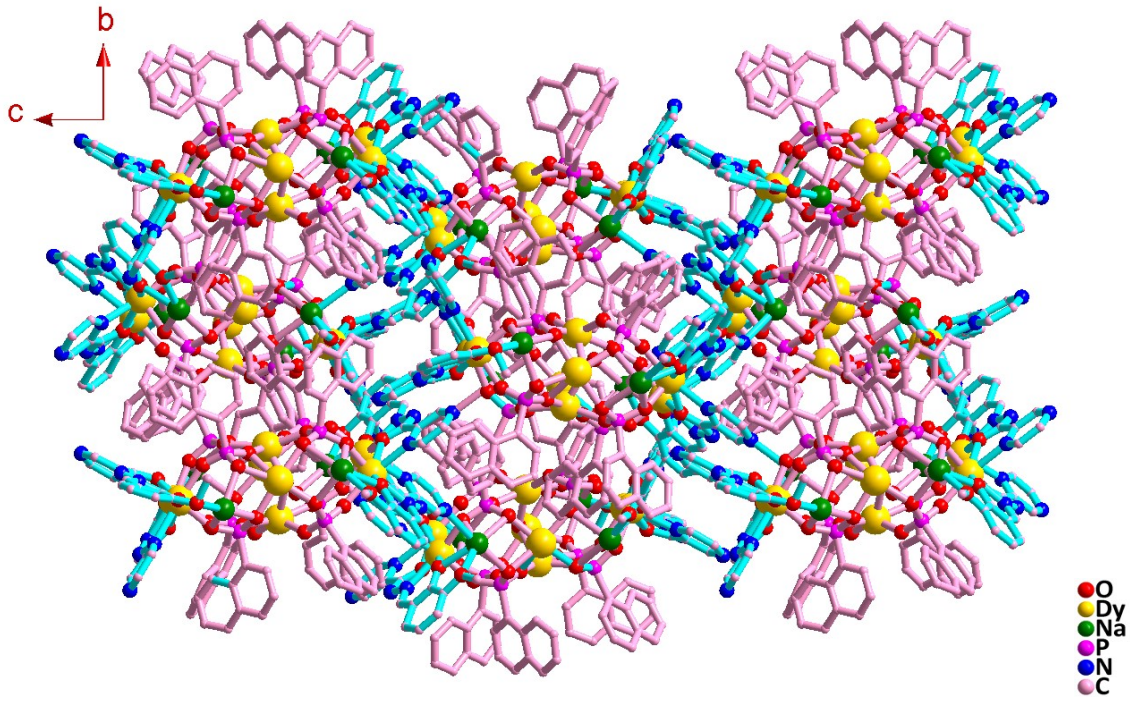


Figure S11. Single-crystal structure of **2**. Color scheme: the opch^{2-} fragments in turquoise, the $C_{10}H_7PO_3^{2-}$ fragments in rose, Dy in gold, O in red, N in blue, C in rose, P in pink.

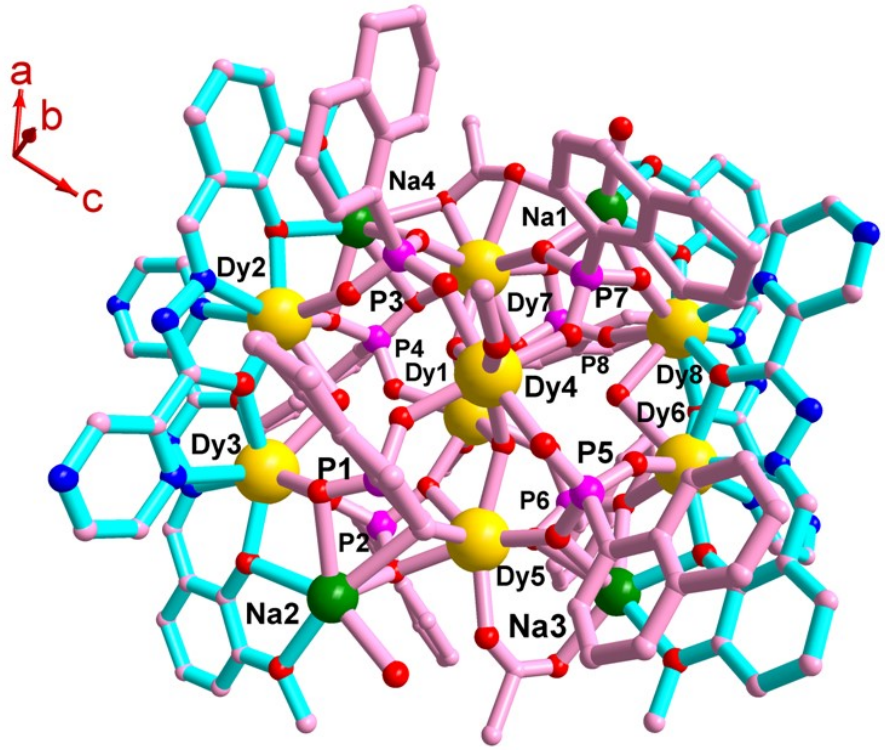


Figure S12. Top-view of **2**. Color scheme: the $opch^{2-}$ fragments in turquoise, the $C_{10}H_7PO_3^{2-}$ fragments in rose, Dy in gold, O in red, N in blue, C in rose, P in pink.

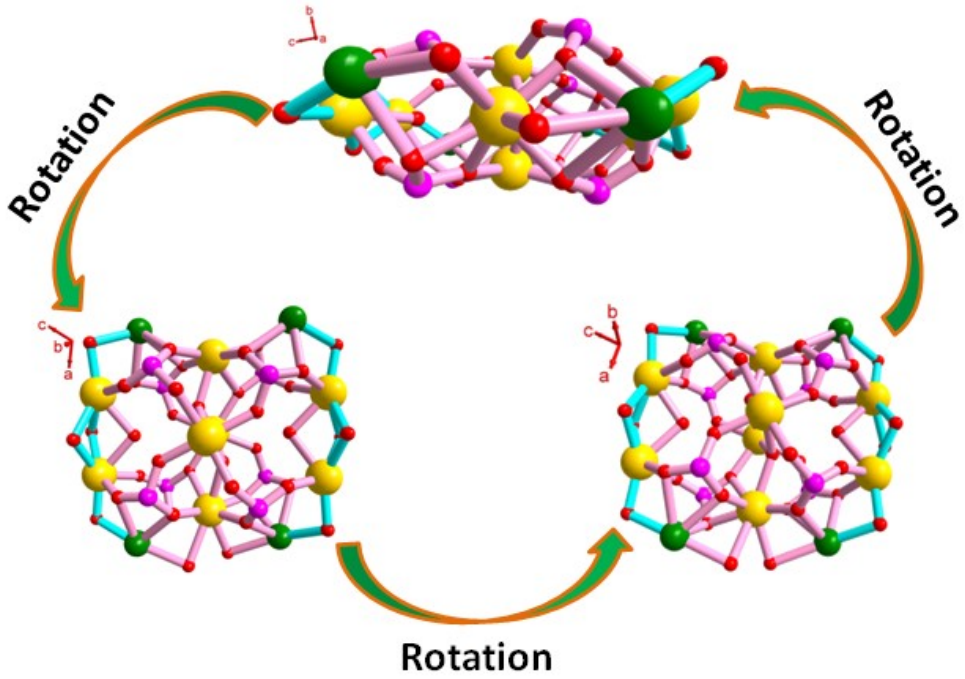


Figure S13. Continuous observation of the Na_4Dy_8 core from top- to side-view in **2**.

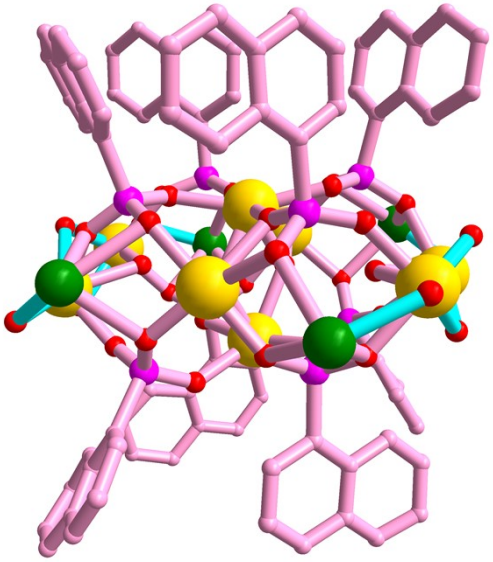


Figure S14. The $[Na_4Dy_8(C_{10}H_7PO_3)_8]^{12+}$ core for **2**. Color scheme: the $C_{10}H_7PO_3^{2-}$ fragments in rose, Dy in gold, O in red, C in rose, P in pink.

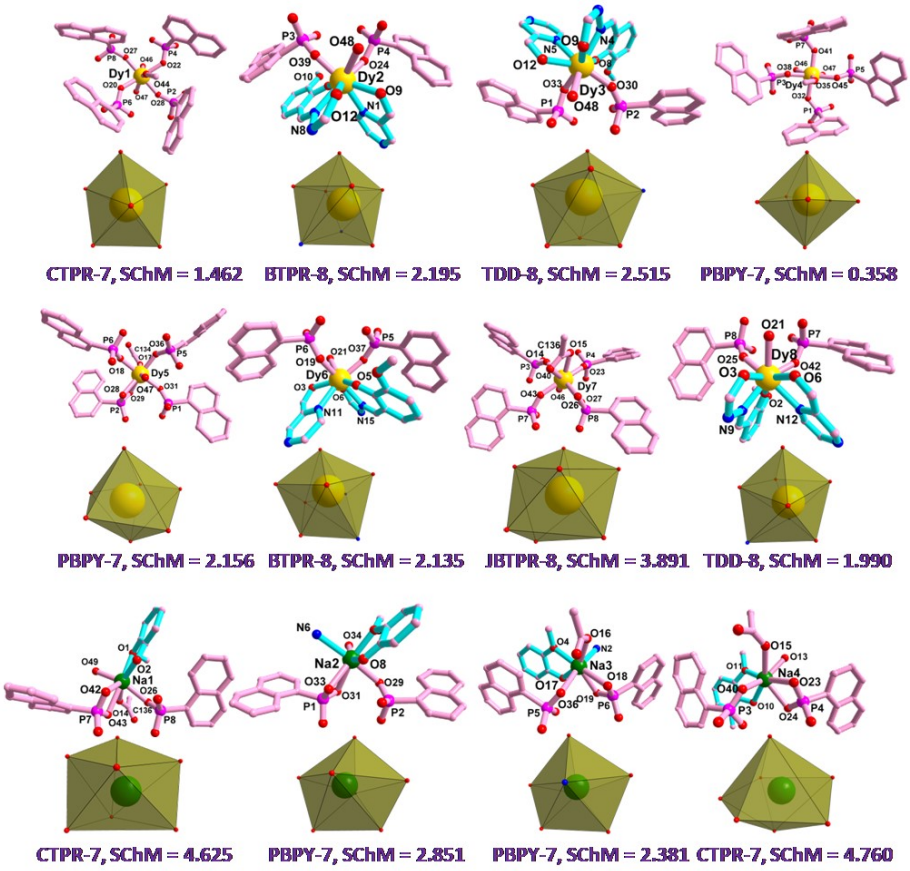


Figure S15. Coordination environments of eight crystallographically independent Dy^{III} ions and four Na^{I} ions in **2**. Color scheme: the $opch^{2-}$ fragments in turquoise, the $C_{10}H_7PO_3^{2-}$ fragments in rose, Dy in gold, O in red, N in blue, C in rose, P in pink.

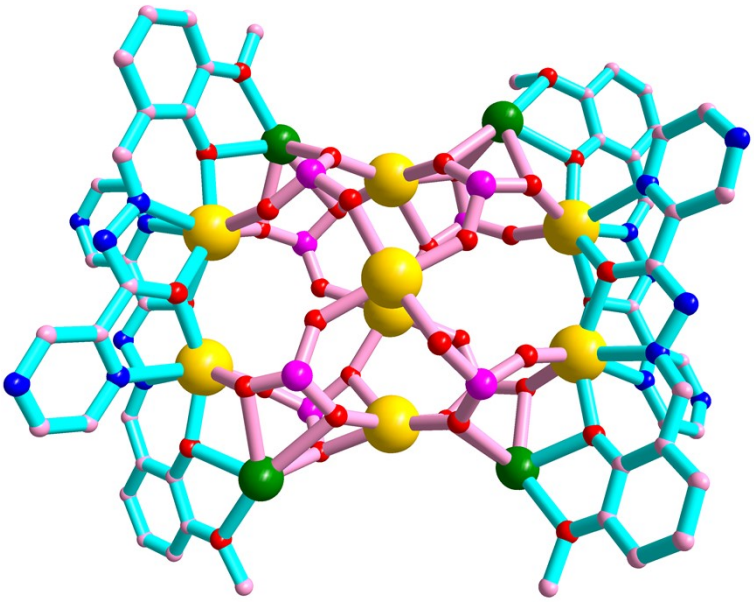


Figure S16. The $[Na_4Dy_8(opch)_4(PO_3)_8]^{4+}$ core for **2**. Color scheme: the $opch^{2-}$ fragments in turquoise, the PO_3^{2-} fragments in rose, Dy in gold, O in red, N in blue, C in rose, P in pink.

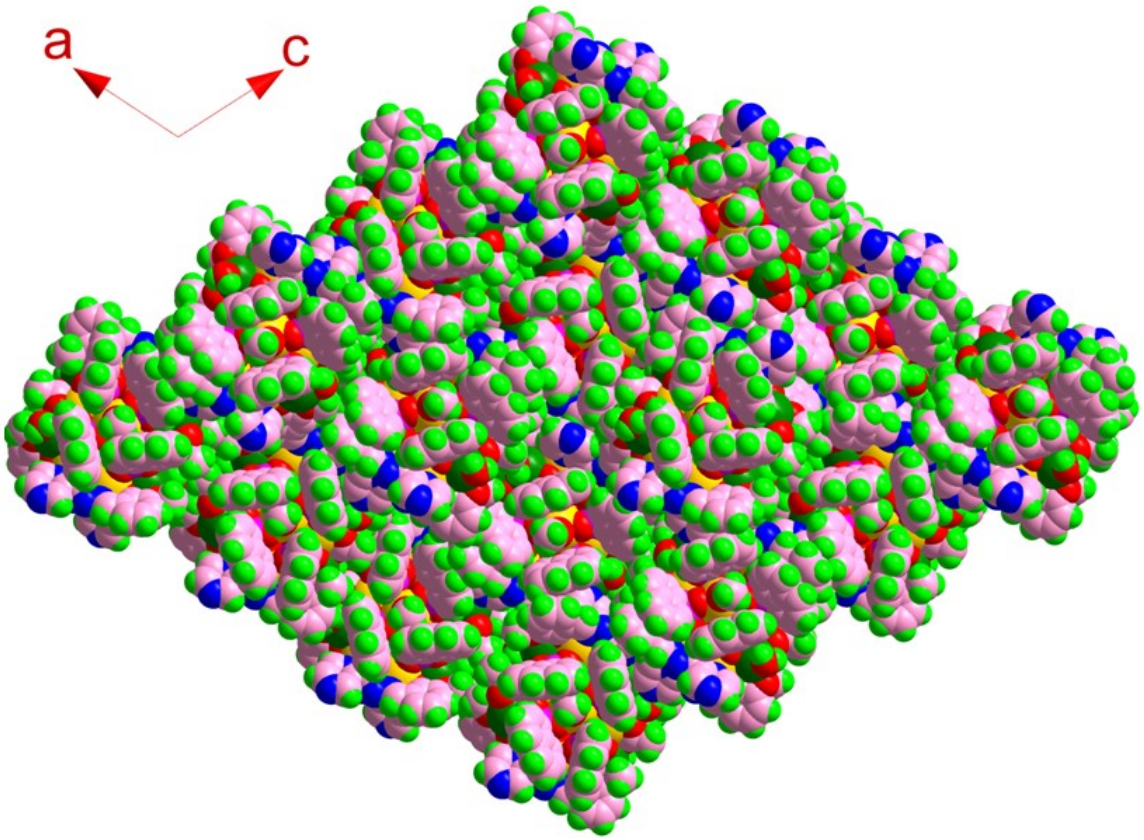


Figure S17. Space-filling representation of **2**. Color scheme: Dy in gold, Na in green, O in red, N in blue, P in pink, C in rose, H in bright green.

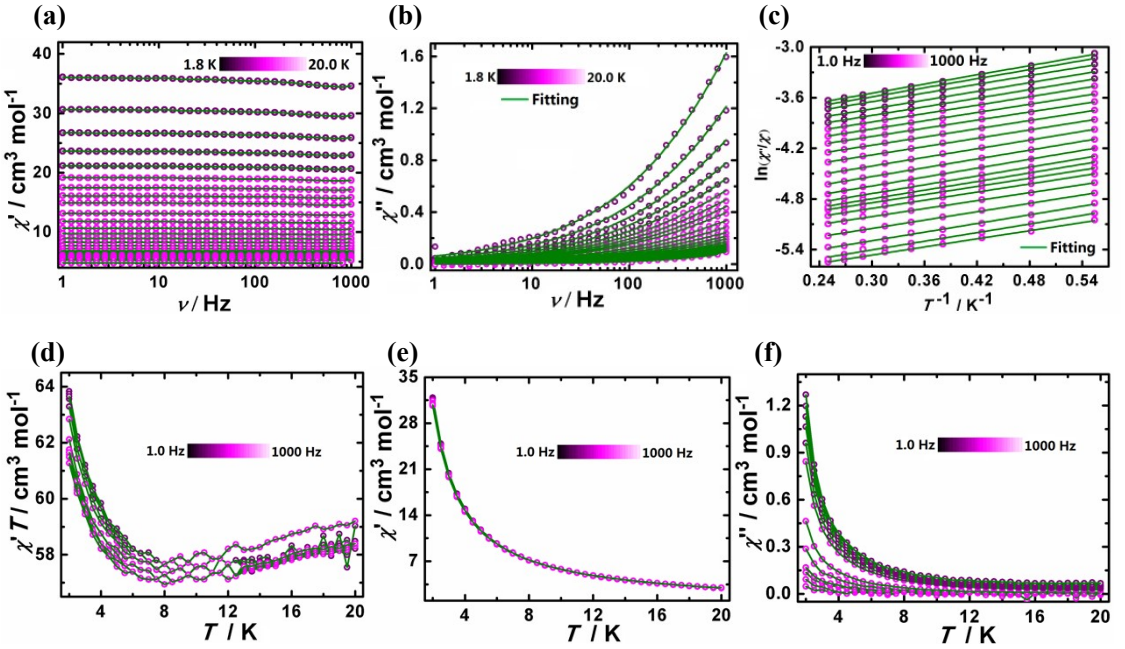


Figure S18. Frequency dependence of the χ' (a) and χ'' (b) products, *ac* susceptibilities under zero *dc* field for **1**. The ln(χ''/χ') *versus* T^{-1} plots (c). Temperature dependence of the $\chi'T$ (d), χ' (e) and χ'' (f) products, *ac* susceptibilities (MPMS3).

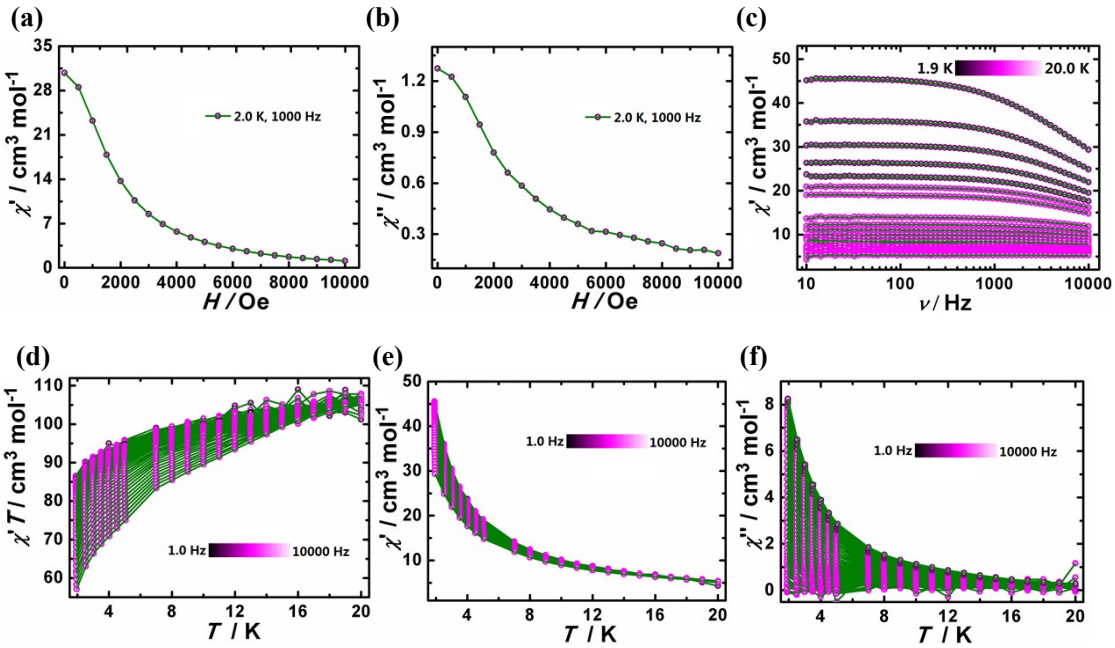


Figure S19. Field dependence of the χ' (a) and χ'' (b) products, ac susceptibility of **1** at 1000 Hz and 2 K. Frequency dependence of the χ' (c) product, *ac* susceptibilities under zero *dc* field. Temperature dependence of the $\chi'T$ (d), χ' (e) and χ'' (f) products, *ac* susceptibilities (PPMS).

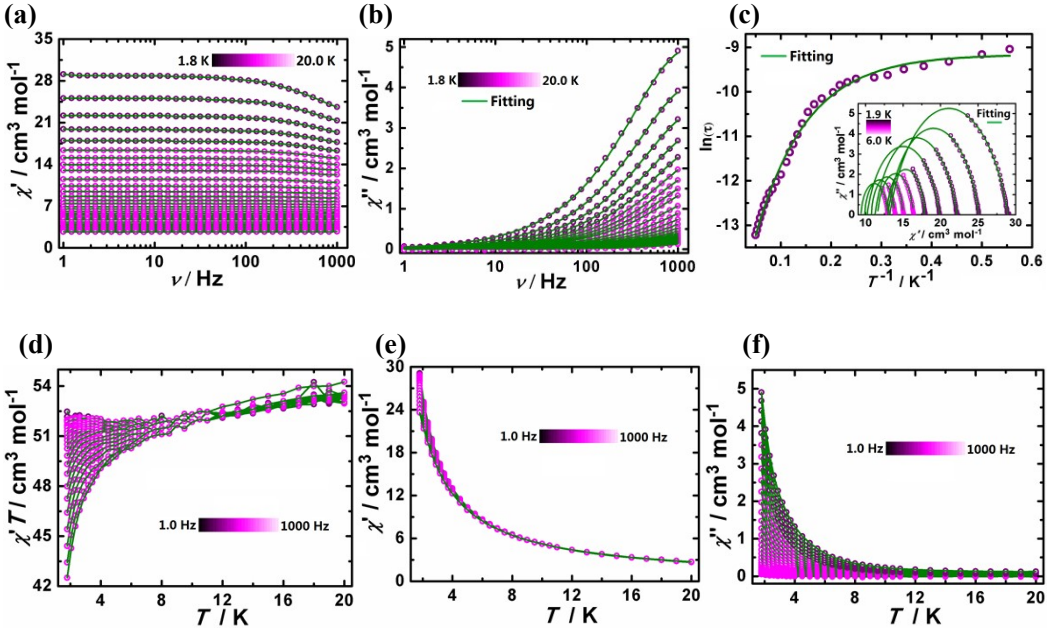


Figure S20. Frequency dependence of the χ' (a) and χ'' (b) products, *ac* susceptibilities under zero *dc* field for **2** (MPMS3). The ln(τ) *versus* T^{-1} plots (c). Temperature dependence of the $\chi'T$ (d), χ' (e) and χ'' (f) products, *ac* susceptibilities (PPMS).

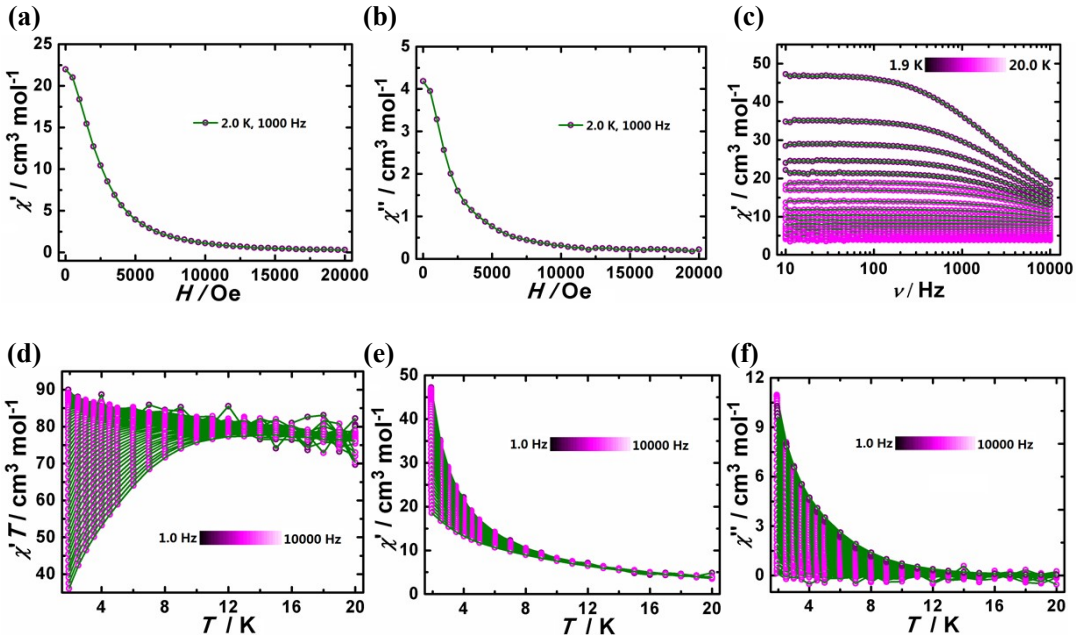


Figure S21. Field dependence of the χ' (a) and χ'' (b) products, ac susceptibility of **2** at 1000 Hz and 2 K. Frequency dependence of the χ' (c) product, *ac* susceptibilities under zero *dc* field. Temperature dependence of the $\chi'T$ (d), χ' (e) and χ'' (f) products, *ac* susceptibilities (PPMS).