Electronic Supplementary Information

Self-assembly of Schiff Base Anions and Trapping of HO^-, O^{2-} and Piv^- Bridges in a Family of Ni_3Ln_4 Complexes: Synthesis, Structures and Magnetic Properties

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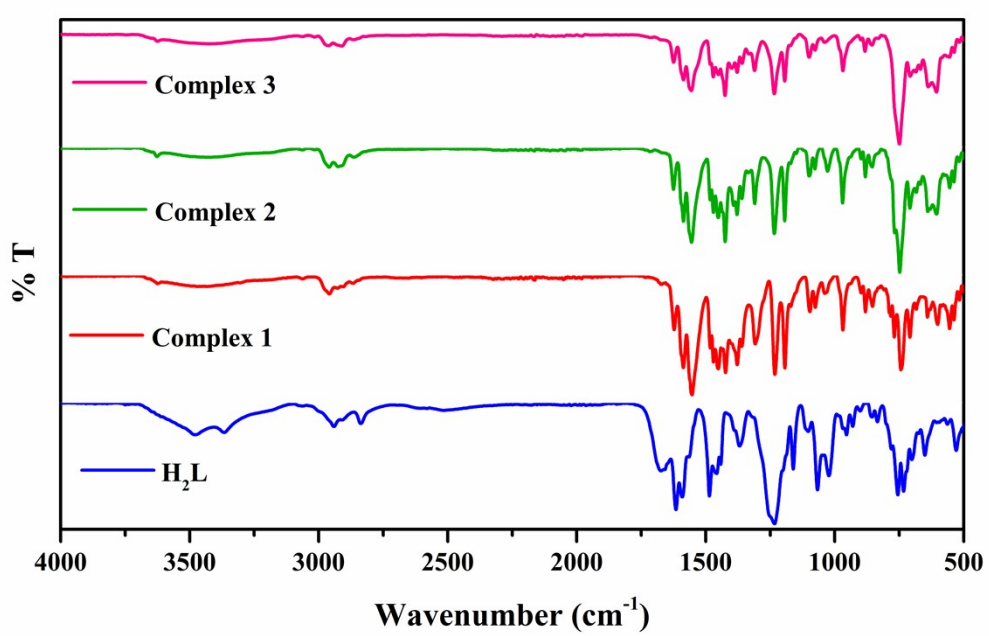
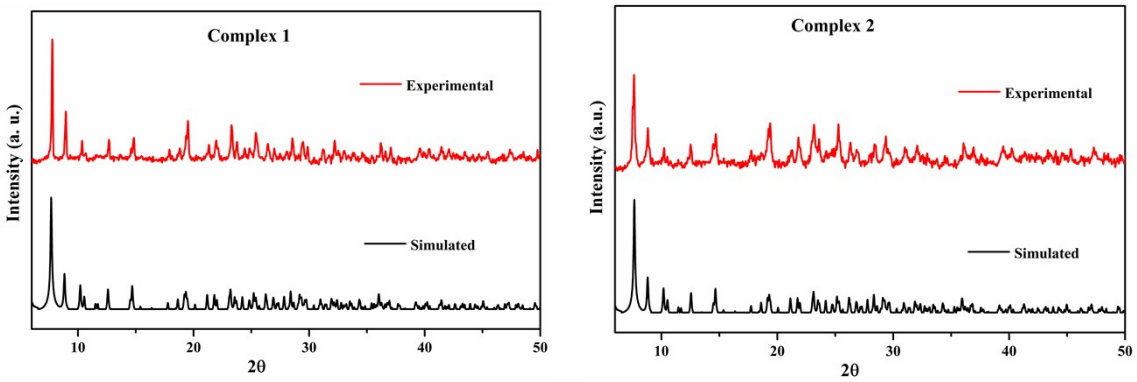


Fig. S1 FT-IR spectra of H_2L and complexes **1-3**



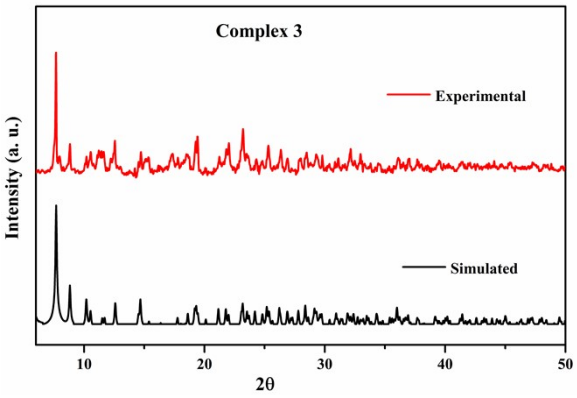


Fig. S2 PXRD patterns of Complexes **1**−**3**

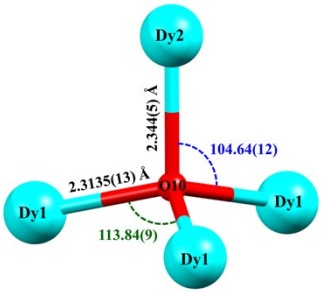


Fig. S3 Tetrahedral (3+1 type) arrangement of four Dy^{III} ions

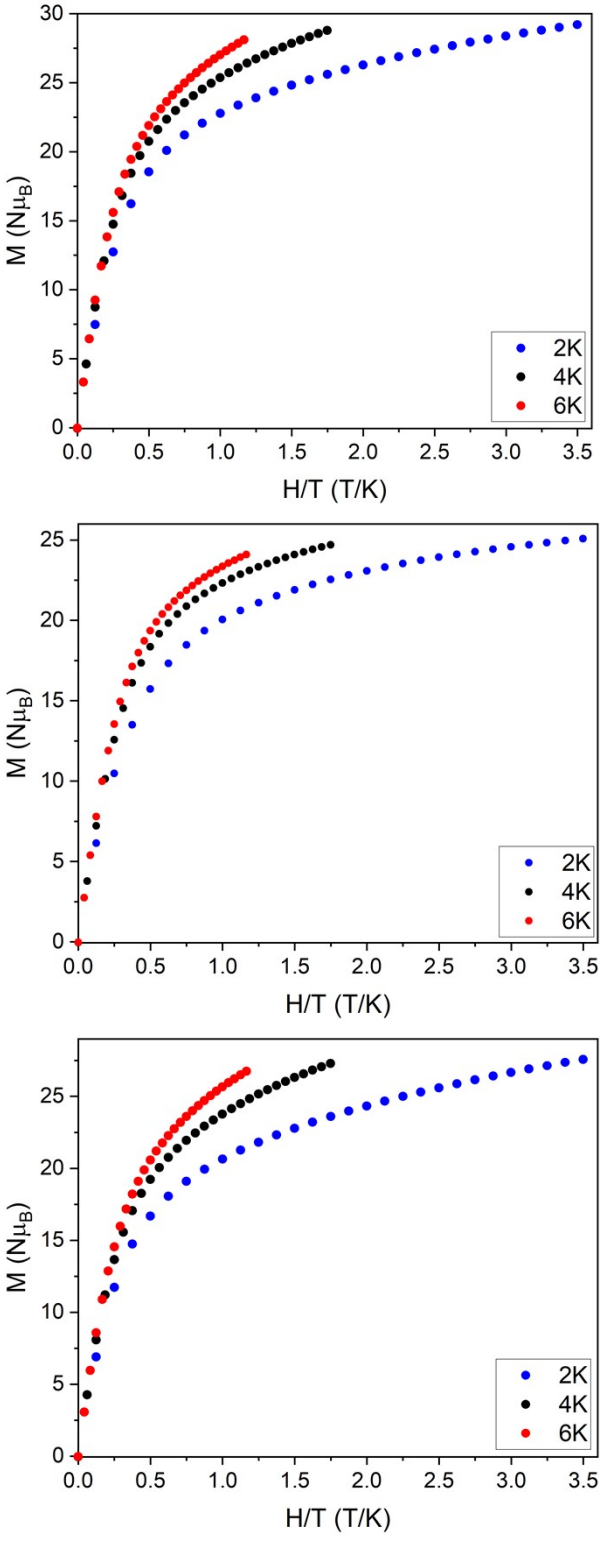


Fig. S4. M vs. H/T data for **1** (upper), **2** (middle) and **3** (lower).

Table S1. *Continuous Shape Measures (CShM) for Ni^{II} centres using SHAPE

Metal centre	Structure				
	HP-6	PPY-6	**OC-6**	TPR-6	JPPY-6
1-Ni	30.423	26.569	**0.534**	13.950	30.041
2-Ni	30.589	26.810	**0.484**	14.014	30.148
3-Ni	30.368	26.524	**0.542**	13.857	29.946

*HP-6 = Hexagon (*D6h*); PPY-6 = Pentagonal pyramid (*C5v*); OC-6 = Octahedron (*Oh*); TPR-6 = Trigonal prism (*D3h*); JPPY-6 = Johnson pentagonal pyramid J2 (*C5v*)

Table S2. #Continuous Shape Measures (CShM) for Ln^{III} centres using SHAPE

	Structure											
$Ln1^{3+}$	HPY-8	HBPY-8	CU-8	SAPR-8	**TDD-8**	JGBF-8	JETBPY-8	JBTPR-8	BTPR-8	JSD-8	TT-8	ETBPY-8
Dy1	22.765	12.869	8.177	4.520	**2.152**	13.802	24.854	2.798	2.965	4.345	8.649	21.013
Tb1	22.792	12.674	8.072	4.646	**2.196**	13.797	24.848	2.910	3.046	4.421	8.545	20.775
Ho1	22.758	13.010	8.254	4.414	**2.124**	13.869	24.909	2.718	2.895	4.275	8.712	21.242

#HPY-8 = (*C7v*) Heptagonal pyramid; HBPY-8 = (*D6h*) Hexagonal bipyramid; CU-8 = (*Oh*) Cube; SAPR-8 = (*D4d*) Square antiprism; TDD-8 = (*D2d*) Triangular dodecahedron; JGBF-8 = (*D2d*) Johnson gyrobifastigium J26; JETBPY-8 = (*D3h*) Johnson elongated triangular bipyramid J14; JBTPR-8 = (*C2v*) Biaugmented trigonal prism J50; BTPR-8 = (*C2v*) Biaugmented trigonal prism; JSD-8 = (*D2d*) Snub diphenoid J84; TT-8 = (*Td*) Triakis tetrahedron; ETBPY-8 = (*D3h*) Elongated trigonal bipyramid

$Ln2^{3+}$	†HP-7	HPY-7	PBPY-7	**COC-7**	CTPR-7	JPBPY-7	JETPY-7
Dy2	36.490	19.366	8.303	**0.343**	2.168	12.071	18.251
Tb2	36.606	19.109	8.319	**0.362**	2.188	12.108	18.422
Ho2	36.429	19.510	8.293	**0.313**	2.140	12.037	18.220

†HP-7 = (*D7h*) Heptagon; HPY-7 = (*C6v*) Hexagonal pyramid; PBPY-7 = (*D5h*) Pentagonal bipyramid; COC-7 = (*C3v*) Capped octahedron; CTPR-7 = (*C2v*) Capped trigonal prism; JPBPY-7 = (*D5h*) Johnson pentagonal bipyramid J13; JETPY-7 = (*C3v*) Johnson elongated triangular pyramid J7

Table S3. Important bond lengths (Å) of complexes **1-3**

	Complex 1 (Dy)	**Complex 2 (Tb)**	**Complex 3 (Ho)**
Ni1–O1	2.016(2)	2.024(2)	2.018(2)
Ni1–O3	2.073(2)	2.078(2)	2.074(2)
Ni1–O5	2.013(2)	2.020(2)	2.018(2)

Ni1–O9	2.034(2)	2.033(2)	2.032(2)
Ni1–O11	2.122(3)	2.130(4)	2.122(3)
Ni1–N1	2.017(3)	2.022(3)	2.020(3)
Ln1–O1	2.327(2)	2.340(2)	2.319(2)
Ln1–O2	2.810(2)	2.813(2)	2.813(2)
Ln1–O3	2.565(2)	2.579(2)	2.554(2)
Ln1–O4	2.440(2)	2.455(2)	2.431(2)
Ln1–O7	2.266(3)	2.287(3)	2.259(2)
Ln1–O8	2.330(2)	2.350(3)	2.318(2)
Ln1–O9	2.357(2)	2.378(2)	2.350(2)
Ln1–O10	2.3133(10)	2.3240(10)	2.3069(9)
Ln2–O6	2.320(2)	2.331(2)	2.313(2)
Ln2–O9	2.315(2)	2.333(2)	2.304(2)
Ln2–O10	2.344(4)	2.357(4)	2.342(3)

Table S4. Important bond angles (º) of complexes **1-3**

	Complex 1 (Dy)	**Complex 2 (Tb)**	**Complex 3 (Ho)**
O1–Ln1–O2	58.79(7)	58.67(7)	58.87(7)
O1–Ln1–O3	131.15(7)	131.04(8)	131.21(7)
O1–Ln1–O4	90.21(8)	90.47(8)	89.92(8)
O1–Ln1–O8	129.54(9)	129.21(9)	129.76(8)
O1–Ln1–O9	69.10(7)	68.83(7)	69.26(7)
O3–Ln1–O2	117.70(7)	117.90(7)	117.35(7)
O4–Ln1–O2	69.77(8)	70.05(8)	69.35(8)
O4–Ln1–O3	51.78(7)	51.55(7)	51.97(7)
O7–Ln1–O1	79.64(9)	79.60(9)	79.50(9)
O7–Ln1–O2	78.80(9)	79.53(10)	78.21(9)
O7–Ln1–O3	148.99(9)	149.01(9)	149.17(8)
O7–Ln1–O4	147.65(10)	148.70(10)	146.66(9)
O7–Ln1–O8	82.63(10)	83.32(11)	82.19(10)
O7–Ln1–O9	125.82(9)	125.19(10)	126.23(9)

O7–Ln1–O10	82.37(10)	81.83(11)	82.93(10)
O8–Ln1–O2	71.60(8)	71.36(9)	71.75(8)
O8–Ln1–O3	78.85(8)	79.20(9)	78.53(8)
O8–Ln1–O4	80.47(9)	80.24(9)	80.64(9)
O8–Ln1–O9	150.81(8)	150.85(9)	150.79(8)
O9–Ln1–O2	116.37(7)	115.89(7)	116.62(7)
O9–Ln1–O3	72.70(7)	72.58(7)	72.89(7)
O9–Ln1–O4	76.95(8)	76.65(8)	77.19(8)
O10–Ln1–O1	115.81(6)	115.54(6)	116.07(6)
O10–Ln1–O2	161.07(8)	161.23(8)	161.06 (8)
O10–Ln1–O3	80.01(6)	79.84(6)	80.22(6)
O10–Ln1–O4	129.15(8)	128.72(8)	129.56(8)
O10–Ln1–O8	107.97(9)	108.70(9)	107.42(9)
O10–Ln1–O9	73.69(9)	73.68(10)	73.76(9)
O6–Ln2–O6	85.34(10)	85.84(10)	84.96(10)
O6–Ln2–O9	81.56(8)	81.33(8)	81.48(8)
O6–Ln2–O9	75.11(8)	74.91(9)	75.37(8)
O6–Ln2–O9	157.20(8)	157.53(8)	156.93(8)
O9–Ln2–O9	112.62(4)	112.63(5)	112.67(4)
O6–Ln2–O10	128.50(7)	128.16(7)	128.76(7)
O9–Ln2–O10	73.89(5)	73.90(5)	73.96(5)
O1–Ni1–O3	89.39(9)	89.29(9)	89.44(9)
O1–Ni1–O5	174.39(10)	174.52(10)	174.31(10)
O1–Ni1–O9	81.97(8)	82.20(9)	81.85(8)
O1–Ni1–O11	90.35(11)	89.03(11)	90.20(11)
O1–Ni1–N1	87.86(10)	87.62(16)	87.97(10)
O5–Ni1–O3	96.18(10)	96.13(10)	96.22(10)
O9–Ni1–O3	88.96(8)	89.25(8)	88.82(8)
O3–Ni1–O11	178.76(11)	178.09(18)	179.13(12)
N1–Ni1–O3	84.12(10)	84.13(10)	84.11(10)
O5–Ni1–O9	98.70(9)	98.62(10)	98.71(9)
O5–Ni1–O11	84.07(11)	84.54(16)	84.12(12)

O5–Ni1–N1	92.09(11)	92.09(11)	92.11(11)
O9–Ni1–O11	92.20(11)	91.43(19)	91.91(11)
N1–Ni1–O9	167.76(10)	167.96(10)	167.67(10)
N1–Ni1–O11	94.66(12)	94.9(2)	95.08(12)