Supporting Information for the Manuscript:

Three Series of Heterometallic Ni^{II}-Ln^{III} Schiff Base Complexes: Synthesis, Crystal Structures, and Magnetic Characterization

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Supplementary Information Contents:

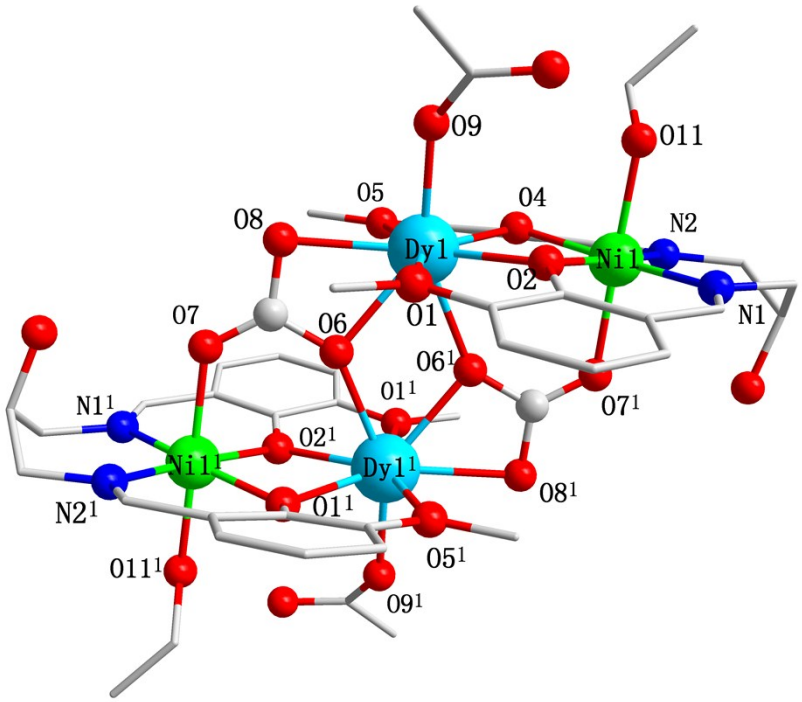

Fig. S**1** View of the Molecular Structure for [{(μ_3-CO_3)$_2$[Ni(H**L**)(CH_3CH_2OH)Dy(CH_3COO)]$_2$}·2CH_3CH_2OH (**2**). Solvent molecules and H atoms are omitted for clarity.

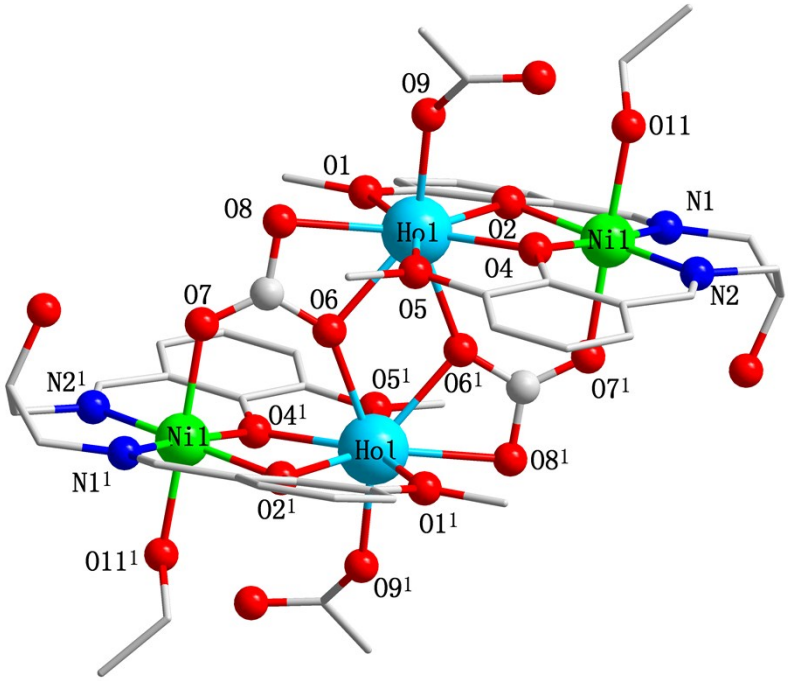

Fig. S**2** View of the Molecular Structure for [{(μ_3-CO_3)$_2$[Ni(H**L**)(CH_3CH_2OH)Ho(CH_3COO)]$_2$}·2CH_3CH_2OH (**3**). Solvent molecules and H atoms are omitted for clarity.

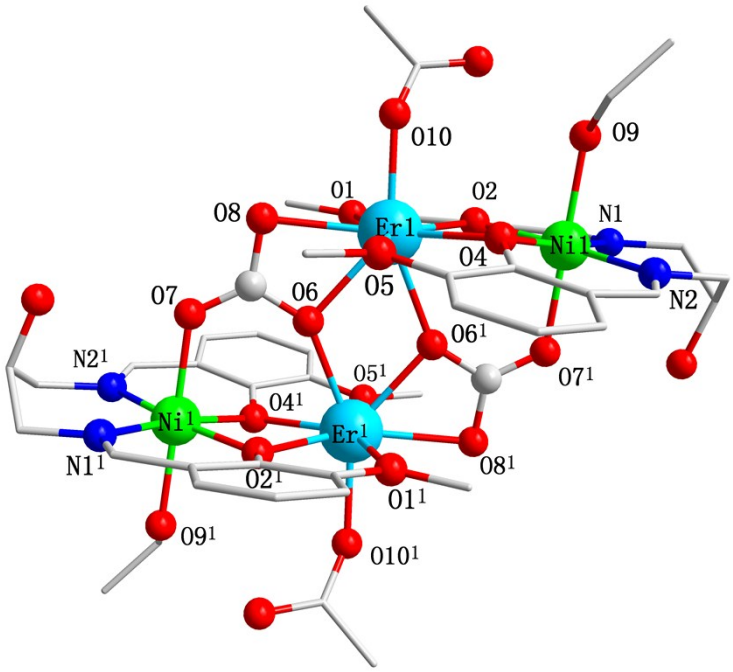

Fig. S**3** View of the Molecular Structure for [{(μ_3-CO_3)$_2$[Ni(H**L**)(CH_3CH_2OH)Er(CH_3COO)]$_2$}·2CH_3CH_2OH (**4**). Solvent molecules and H atoms are omitted for clarity.

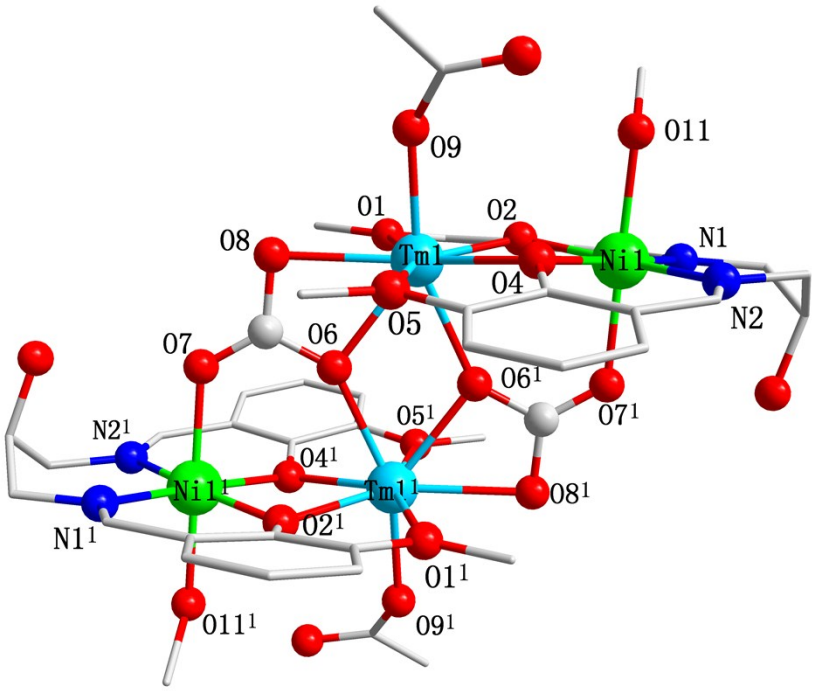

Fig. S**4** View of the Molecular Structure for [{(μ_3-CO_3)$_2$[Ni(H**L**)(CH_3CH_2OH)Tm (CH_3COO)]$_2$}·2CH_3CH_2OH (**5**). Solvent molecules and H atoms are omitted for clarity.

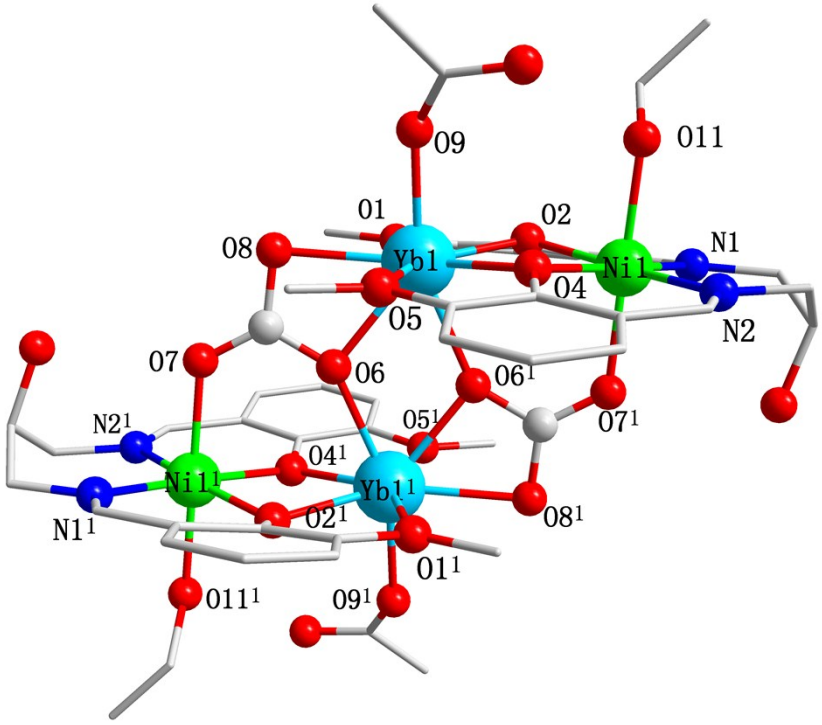

Fig. S**5** View of the Molecular Structure for [{(μ_3-CO_3)$_2$[Ni(H**L**)(CH_3CH_2OH)Yb (CH_3COO)]$_2$}·2CH_3CH_2OH (**6**). Solvent molecules and H atoms are omitted for clarity.

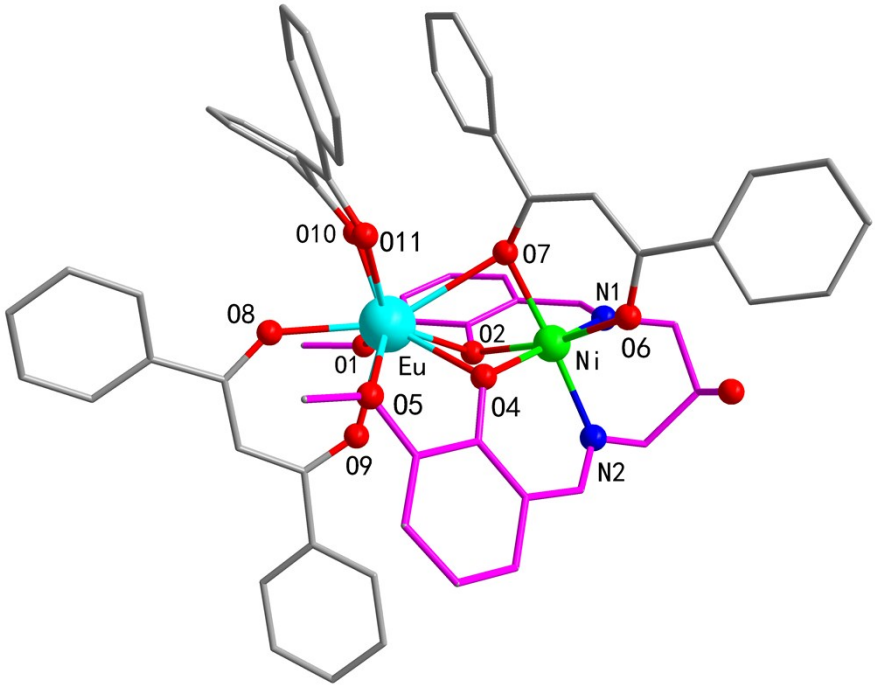

Fig. S**6** View of the Molecular Structure for [Ni(H**L**)Eu(dbm)$_3$] ·CH_3OH·$2CH_2Cl_2$ (**8**). Solvent molecules and H atoms are omitted for clarity.

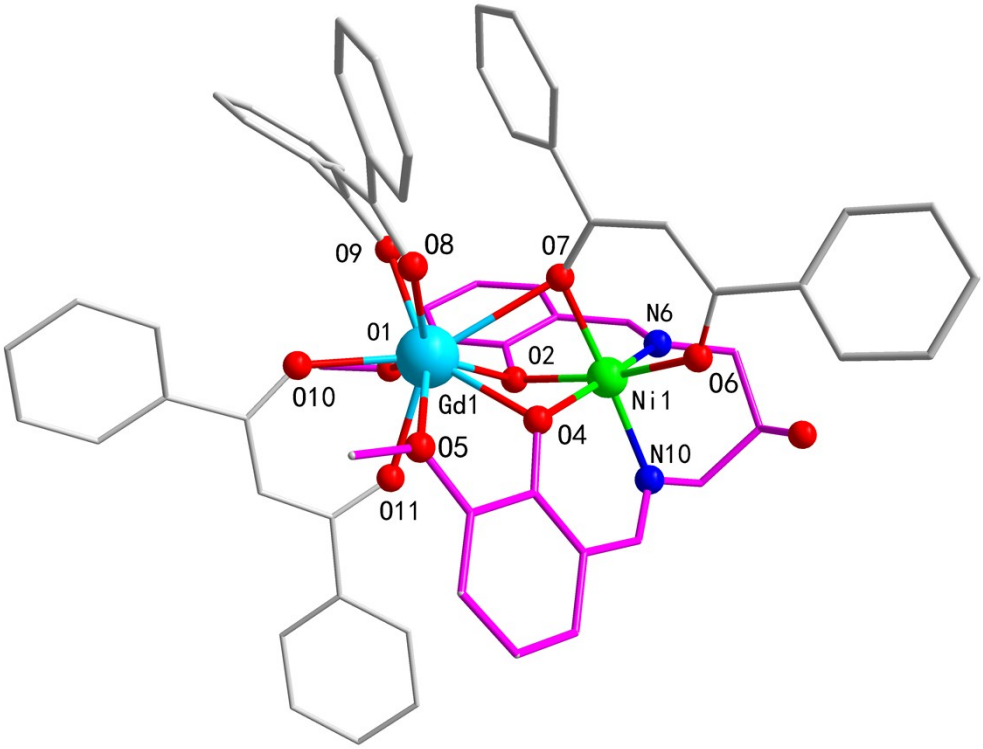

Fig. S**7** View of the Molecular Structure for [Ni(H**L**)Gd(dbm)$_3$] ·CH_3OH·$2CH_2Cl_2$ (**9**). Solvent molecules and H atoms are omitted for clarity.

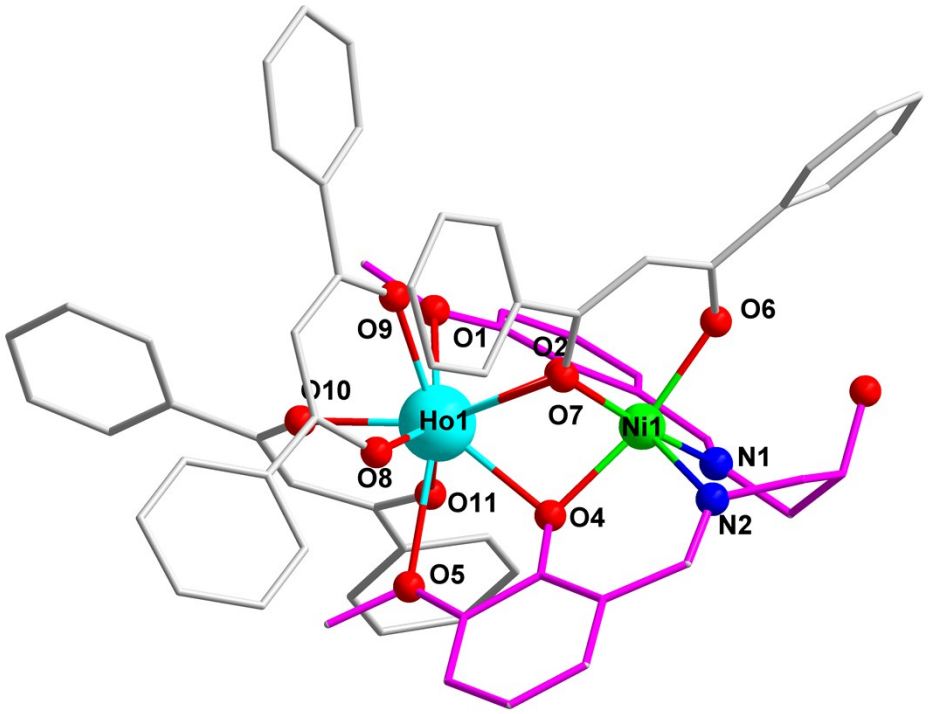

Fig. S**8** View of the Molecular Structure for [Ni(H**L**)Ho(dbm)$_3$] ·CH_3OH·$2CH_2Cl_2$ (**10**). Solvent molecules and H atoms are omitted for clarity.

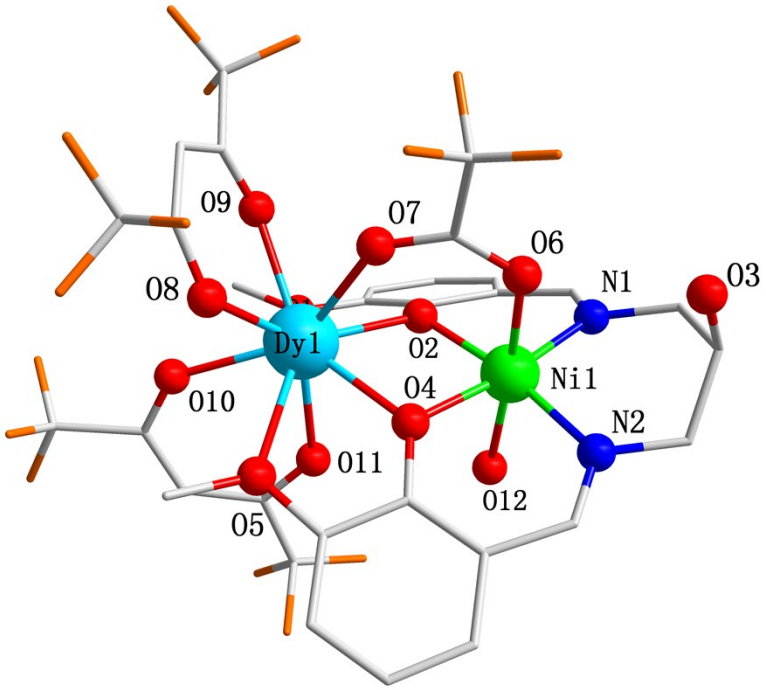

Fig. S**9** View of the Molecular Structure for [Ni(HL)(H_2O)(tfa)Dy(hfac)$_2$] (**12**). Solvent molecules and H atoms are omitted for clarity.

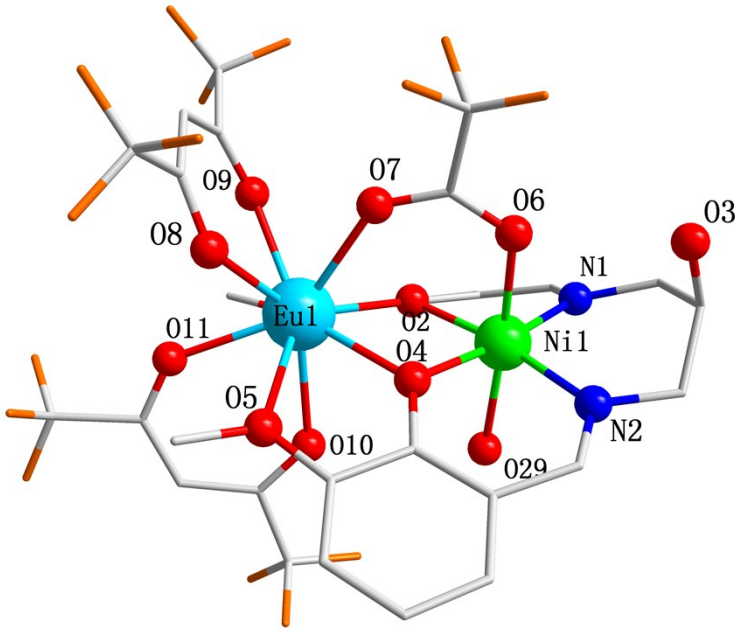

Fig. S**10** View of the Molecular Structure for [Ni(HL)(H_2O)(tfa)Eu(hfac)$_2$] (**13**). Solvent molecules and H atoms are omitted for clarity.

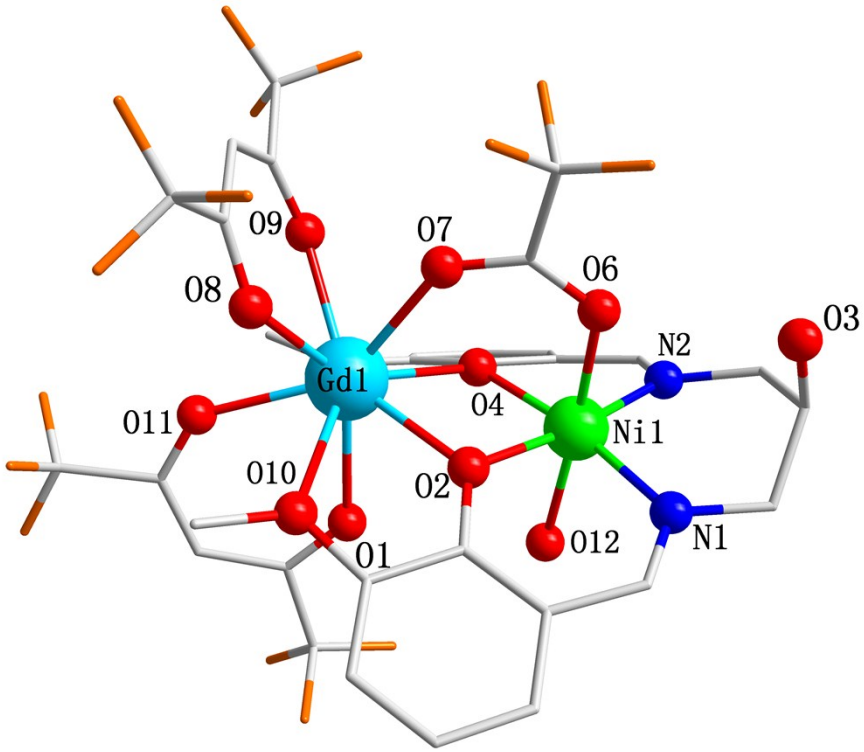

Fig. S**11** View of the Molecular Structure for [Ni(HL)(H_2O)(tfa)Gd(hfac)$_2$] (**14**). Solvent molecules and H atoms are omitted for clarity.

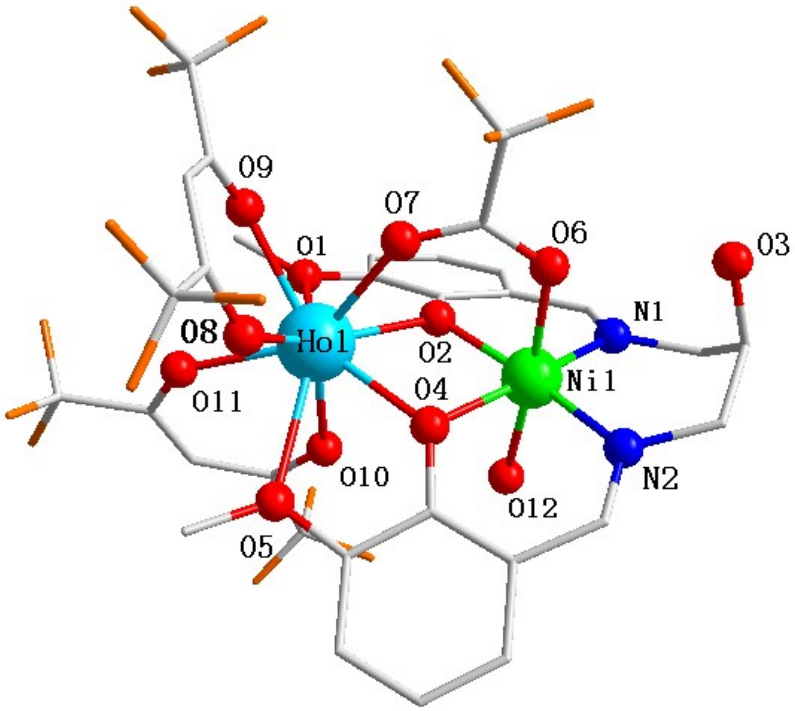

Fig. S**12** View of the Molecular Structure for [Ni(HL)(H_2O)(tfa)Ho(hfac)$_2$] (**15**). Solvent molecules and H atoms are omitted for clarity.

Table S**1** Selected coordination bond distances (Å) and angles (°) for complex **1-4**.

1		**2**		**3**		**4**	
Selected bond distances around Ni^{II}							
Tb1-Ni1	3.4176(10)	Dy1-Ni1	3.3919(10)	Ho1-Ni1	3.3908(9)	Er1-Ni1	3.3699(5)
Ni1-O2	2.051(3)	O7-Ni1^1	2.107(3)	O7-Ni1^1	2.112(3)	Ni1-O2	2.022(2)
Ni1-O1^1	2.105(3)	Ni1-O4	2.043(3)	Ni1-O4	2.032(3)	Ni1-O4	2.035(3)
Ni1-O4	2.042(3)	Ni1-O2	2.031(2)	Ni1-O2	2.037(3)	Ni1-O7^1	2.101(2)
Ni1-N2	2.048(4)	Ni1-O11	2.099(3)	Ni1-N2	2.041(3)	Ni1-O9	2.094(2)
Ni1-N1	2.059(4)	Ni1-N1	2.042(3)	Ni1-N1	2.052(3)	Ni1-N2	2.035(3)
O7-Ni1^1	2.108(3)	Ni1-N2	2.042(3)	Ni1-O11	2.106(3)	Ni1-N1	2.035(3)
Selected bond distances around Tb^{III}		Selected bond distances around Dy^{III}		Selected bond distances around Ho^{III}		Selected bond distances around Er^{III}	
Tb1-O2	2.309(3)	O6-Dy1	2.315(2)	Ho1-O5	2.489 (3)	Er1-O2	2.260(2)
Tb1-O5	2.514(3)	O6-Dy1^1	2.352(2)	Ho1-O4	2.276(2)	Er1-O10	2.199(3)
Tb1-O6	2.471(3)	Dy1-O4	2.293(2)	Ho1-O8	2.436(2)	Er1-O1	2.476(2)
Tb1-O1	2.513(3)	Dy1-O2	2.287(3)	Ho1-O2	2.282(3)	Er1-O4	2.265(2)
Tb1-O4	2.309(3)	Dy1-O1	2.497(2)	Ho1-O1	2.479 (3)	Er1-O5	2.467(2)
Tb1-O8^1	2.376(3)	Dy1-O5	2.493(2)	Ho1-O6	2.306(3)	Er1-O8	2.417(2)
Tb1-O8	2.330(3)	Dy1-O9	2.226(3)	Ho1-O6^1	2.343(3)	Er1-O6	2.299(2)
Tb1-O9	2.250(4)	Dy1-O8	2.446(3)	Ho1-O9	2.222(3)	Er1-O6^1	2.321(2)
1		**2**		**3**		**4**	
Bond angles containing both Tb^{III} and Ni^{II}		Bond angles containing both Dy^{III} and Ni^{II}		Bond angles containing both both Ho^{III} and Ni^{II}		Bond angles containing both Er^{III} and Ni^{II}	
Ni1-O2-Tb1	103.06(13)	Ni1-O4-Dy1	102.78(10)	Ni1-O4-Ho1	103.68(11)	Ni1-O2-Er1	103.65(10)
Ni1-O4-Tb1	103.31(13)	Ni1-O2-Dy1	103.38(10)	Ni1-O2-Ho1	103.33(12)	Ni1-O4-Er1	103.09(10)
Bond angles around Ni^{II}							
1		**2**		**3**		**4**	
N1-Ni1-N2	98.93(16)	O4-Ni1-O7^1	93.63(10)	O7^1-Ni1-O11	177.64(12)	O4-Ni1-O2	80.56(10)
O2-Ni1-O7^1	93.77(13)	O2-Ni1-O7^1	91.19(10)	O4-Ni1-O7^1	91.21(11)	O7^1-Ni1-O2	91.10(10)
O11-Ni1-O7^1	177.56(14)	O2-Ni1-O4	81.28(10)	O4-Ni1-N2	89.65(12)	O7^1-Ni1-O4	93.93(10)
O11-Ni1-O2	87.70(12)	O11-Ni1-O7^1	177.83(11)	O4-Ni1-N1	171.26(12)	O9-Ni1-O2	87.83(10)
O4-Ni1-O7^1	91.04(13)	O11-Ni1-O4	88.00(10)	O4-Ni1-O11	87.67(11)	O9-Ni1-O4	87.83(10)

O4-Ni1-O2	81.36(12)	O11-Ni1-O2	87.63(12)	O2-Ni1-O7^{1}	94.05(11)	O9-Ni1-O7^{1}	177.70(11)
O4-Ni1-O11	87.70(12)	N1-Ni1-O7^{1}	89.73(12)	O2-Ni1-O4	80.61(10)	N2-Ni1-O2	171.09(12)
N2-Ni1-O7^{1}	89.63(15)	N1-Ni1-O4	170.22 (11)	O2-Ni1-N2	169.74(12)	N2-Ni1-O4	90.68(12)
N2-Ni1-O2	170.19(13)	N1-Ni1-O2	89.48(12)	O2-Ni1-N1	90.73(12)	N2-Ni1-O7^{1}	87.92(11)
N2-Ni1-O11	88.27(14)	N1-Ni1-O11	88.44(12)	O2-Ni1-O11	87.81(11)	N2-Ni1-O9	93.51(13)
N2-Ni1-O4	89.39 (14)	N2-Ni1-O7^{1}	87.89(12)	N2-Ni1-O7^{1}	89.35(13)	N1-Ni1-O2	89.60(11)
N1-Ni1-O7^{1}	89.63 (15)	N2-Ni1-O4	90.37(12)	N2-Ni1-N1	99.06(13)	N1-Ni1-O4	169.61(10)
N1-Ni1-O2	90.25(16)	N2-Ni1-O2	171.55(14)	N2-Ni1-O11	88.57(13)	N1-Ni1-O7^{1}	89.57(11)
N1-Ni1-O11	93.06(14)	N2-Ni1-O11	93.54(12)	N1-Ni1-O7^{1}	88.18(13)	N1-Ni1-O9	88.45(11)
N1-Ni1-O4	170.19(13)	N2-Ni1-N1	98.94(13)	N1-Ni1-O11	93.24(13)	N1-Ni1-N2	99.24(12)
Bond angles around Tb^{III}		Bond angles around Dy^{III}		Bond angles around Ho^{III}		Bond angles around Er^{III}	
O6-Tb1-O2	143.59(11)	O4-Dy1-O6^{1}	74.13(9)	O4-Ho1-O5	65.61(8)	O10-Er1-O2	83.22(9)
O6-Tb1-O5	80.11(11)	O4-Dy1-O6	125.46(9)	O4-Ho1-O8	144.58(10)	O1-Er1-O2	65.75(8)
O1-Tb1-O2	65.31(11)	O2-Dy1-O6^{1}	73.73 (9)	O4-Ho1-O2	70.55(9)	O1-Er1-O10	82.48(8)
O1-Tb1-O5	158.91(11)	O2-Dy1-O6	121.28(9)	O4-Ho1-O1	136.09(9)	O4-Er1-O2	70.85(9)
O1-Tb1-O6	78.91(11)	O2-Dy1-O4	70.80(9)	O4-Ho1-O6	121.19(10)	O4-Er1-O10	90.53(9)
O4-Tb1-O2	70.59(11)	O1-Dy1-O6^{1}	94.45(9)	O4-Ho1-O6^{1}	73.83(10)	O4-Er1-O1	136.55(8)
O4-Tb1-O5	65.21(10)	O1-Dy1-O6	80.74(9)	O8-Ho1-O5	79.46(9)	O5-Er1-O2	136.55(8)
O4-Tb1-O6	144.98(11)	O1-Dy1-O4	136.08(9)	O8-Ho1-O1	78.94(9)	O5-Er1-O10	98.24(8)
O4-Tb1-O1	135.87(10)	O1-Dy1-O2	65.30(8)	O2-Ho1-O5	136.12(9)	O5-Er1-O1	157.69(8)
O8^{1}-Tb1-O2	73.48(11)	O5-Dy1-O6	85.04(9)	O2-Ho1-O8	143.66(9)	O5-Er1-O4	65.75(8)
O8-Tb1-O2	125.98(11)	O5-Dy1-O6^{1}	93.00(9)	O2-Ho1-O1	65.56(9)	O8-Er1-O2	144.50(9)
O8^{1}-Tb1-O5	94.76(11)	O5-Dy1-O4	65.30(9)	O2-Ho1-O6	126.02(10)	O8-Er1-O10	86.32(8)
O8-Tb1-O5	80.08(11)	O5-Dy1-O2	136.09(8)	O2-Ho1-O6^{1}	74.24(10)	O8-Er1-O1	79.31(8)
O8^{1}-Tb1-O6	116.42(10)	O5-Dy1-O1	158.61(9)	O1-Ho1-O5	158.30(8)	O8-Er1-O4	143.23(9)
O8-Tb1-O6	53.78(10)	O9-Dy1-O6	140.14(9)	O6^{1}-Ho1-O5	94.97(10)	O8-Er1-O5	78.48(8)
O8-Tb1-O1	85.80(11)	O9-Dy1-O6^{1}	155.28(9)	O6-Ho1-O5	80.51(10)	O6^{1}-Er1-O2	74.07(9)
O8^{1}-Tb1-O1	92.61(11)	O9-Dy1-O4	90.93(9)	O6^{1}-Ho1-O8	116.71(9)	O6-Er1-O2	121.15(9)
O8^{1}-Tb1-O4	73.17(11)	O9-Dy1-O2	82.87(9)	O6-Ho1-O8	54.41(9)	O6-Er1-O10	139.50(8)
O8-Tb1-O4	120.45(12)	O9-Dy1-O1	82.63 (9)	O6^{1}-Ho1-O1	92.86(10)	O6^{1}-Er1-O10	155.56(9)
O9-Tb1-O2	90.61(11)	O9-Dy1-O5	98.55(9)	O6-Ho1-O1	85.27(10)	O6^{1}-Er1-O1	95.59 (9)
O9-Tb1-O5	82.57 (11)	O8-Dy1-O6^{1}	116.54(9)	O6-Ho1-O6^{1}	62.45(10)	O6-Er1-O1	80.18(9)
O9-Tb1-O6	88.40(11)	O8-Dy1-O6	54.29(9)	O9-Ho1-O5	82.50(10)	O6^{1}-Er1-O4	74.07(9)
O9-Tb1-O1	98.96(11)	O8-Dy1-O4	143.58(9)	O9-Ho1-O4	83.16(10)	O6-Er1-O4	126.47(9)
O9-Tb1-O4	82.67(12)	O8-Dy1-O2	144.53(9)	O9-Ho1-O8	86.80(9)	O6^{1}-Er1-O5	92.64(9)
O9-Tb1-O8^{1}	154.32(12)	O8-Dy1-O1	79.72(9)	O9-Ho1-O2	90.75(10)	O6-Er1-O5	85.22(9)
O9-Tb1-O8	140.46 (11)	O8-Dy1-O5	79.01(9)	O9-Ho1-O1	98.32(9)	O6^{1}-Er1-O8	117.41(8)
		O8-Dy1-O9	87.22(9)	O9-Ho1-O6^{1}	155.61(9)	O6-Er1-O8	54.64(8)
				O9-Ho1-O6	139.81(9)	O9-Er1-O2	83.22(9)
^{1}1-X,1-Y,2-Z		^{1}1-X,1-Y,1-Z		^{1}1-X,1-Y,2-Z		^{1}-X,2-Y,1-Z	

Table S**2** Selected coordination distances (Å) and angles (°) for complex **5-8**.

5		**6**		**7**		**8**	
Selected bond distances around Ni^{II}							
Tm1-Ni1	3.3835(10)	Yb1-Ni1	3.3645(10)	Tb-Ni	3.3048(5)	Eu-Ni1	3.3180(8)
Ni1-O4	2.031(3)	O7-Ni1^{1}	2.116(3)	Ni-O2	2.013(3)	O2-Ni1	2.048(4)
Ni1-O2	2.035(3)	Ni1-O4	2.022(3)	Ni-O4	2.035(3)	O4-Ni1	2.011(4)
Ni1-N1	2.029(4)	Ni1-O2	2.036(3)	Ni-O6	2.047(3)	O7-Ni1	2.056(4)
Ni1-N2	2.049(4)	Ni1-N1	2.038(4)	Ni-O7	2.045(3)	O6-Ni1	2.051(5)
Ni1-O11	2.126(3)	Ni1-N2	2.043 (4)	Ni-N1	2.052(3)	N1-Ni1	2.032(6)
O7^{1}-Ni1	2.120(3)	Ni1-O11	2.108(3)	Ni-N2	2.023(3)	N2-Ni1	2.053(6)
Selected bond distances around Tm^{III}		Selected bond distances around europium Yb^{III}		Selected bond distances around Tb^{III}		Selected bond distances around Eu^{III}	
Tm1-O1	2.491(3)	Yb1-O4	2.248(3)	Tb-O1	2.744(3)	Eu-O2	2.430(4)
Tm1-O5	2.463(3)	Yb1-O8	2.395(3)	Tb-O2	2.328(2)	Eu-O10	2.307(4)
Tm1-O4	2.277(3)	Yb1-O5	2.473(3)	Tb-O4	2.385(2)	Eu-O9	2.353(4)
Tm1-O2	2.255(3)	Yb1-O1	2.466(3)	Tb-O5	2.674(3)	Eu-O5	2.751(5)
Tm1-O8	2.409(3)	Yb1-O2	2.260(3)	Tb-O7	2.766(3)	Eu-O4	2.363(4)
Tm1-O6^{1}	2.320(3)	Yb1-O6^{1}	2.307(3)	Tb-O8	2.326(3)	Eu-O11	2.337(4)
Tm1-O6	2.296(3)	Yb1-O6	2.268(3)	Tb-O9	2.277(2)	Eu-O8	2.333(4)
Tm1-O9	2.199(3)	Yb1-O9	2.184(3)	Tb-O10	2.308(2)	Eu-O7	2.738(4)
				Tb-O11	2.277(2)	Eu-O1	2.679(4)
5		**6**		**7**		**8**	
Bond angles containing both Tm^{III} and Ni^{II}		Bond angles containing both Yb^{III} and Ni^{II}		Bond angles containing both Tb^{III} and Ni^{II}		Bond angles containing both Eu^{III} and Ni^{II}	
Ni1-O4-Tm1	103.41(12)	Ni1-O4-Yb1	103.84(12)	Ni-O2-Tb	96.46(9)	Ni1-O2-Eu	95.22(16)
Ni1-O2-Tm1	104.04 (13)	Ni1-O2-Yb1	102.95(12)	Ni-O4-Tb	98.91(10)	Ni1-O4-Eu	98.35(17)

				Ni-O7-Tb	85.34(9)	Ni1-O7-Eu	86.35(15)
Bond angles around Ni^{II}							
5		**6**		**7**		**8**	
O4-Ni1-O7[1]	91.66(13)	O4-Ni1-O7[1]	91.17(12)	O4-Ni1-O7	167.96(10)	O2-Ni1-O7	79.85(17)
O2-Ni1-O7[1]	93.00(13)	O2-Ni1-O7[1]	93.52(12)	O4-Ni1-O6	79.48(10)	O2-Ni1-O6	168.41(19)
O2-Ni1-O4	80.04(12)	O2-Ni1-O4	80.33(12)	O4-Ni1-N1	102.75(12)	O2-Ni1-N2	102.0(2)
N1-Ni1-O7[1]	88.02(15)	N1-Ni1-O7[1]	87. 98(14)	O2-Ni1-O4	80.65(10)	O4-Ni1-O2	81.65(17)
N1-Ni1-O4	171.53(15)	N1-Ni1-O4	170.90(14)	O2-Ni1-O7	94.24(11)	O4-Ni1-O7	82.50(17)
N1-Ni1-O2	91.53(15)	N1-Ni1-O2	90.67(14)	O2-Ni1-O6	82.74(10)	O4-Ni1-O6	93.90(18)
N2-Ni1-O7[1]	89.22(14)	N2-Ni1-O7[1]	170.90(14)	O2-Ni1-N2	169.98(12)	O4-Ni1-N1	170.8(2)
N2-Ni1-O4	90.11(14)	N2-Ni1-O4	89.90(12)	N2-Ni1-O4	89.80(12)	O4-Ni1-N2	90.2(2)
N2-Ni1-O2	169.95(14)	N2-Ni1-O2	169.75(14)	O7-Ni1-N1	88.09(12)	O6-Ni1-O7	88.99(18)
N2-Ni1-N1	98.35(17)	N2-Ni1-N1	99.16(16)	O7-Ni1-O6	89.09(10)	O6-Ni1-N2	88.6(2)
O11-Ni1-O7[1]	177.71(15)	O11-Ni1-O7[1]	178.06(13)	O6-Ni1-N1	172.18(12)	N1-Ni1-O2	89.4(2)
O11-Ni1-O4	86.85(13)	O11-Ni1-Yb1	94.37(9)	O2-Ni1-N1	90.19(12)	N1-Ni1-O7	98.3(2)
O11-Ni1-O2	88.46(13)	O11-Ni1-O4	87.89(12)	N2-Ni1-O7	95.71(13)	N1-Ni1-O6	95.3(2)
O11-Ni1-N1	93.71(15)	O11-Ni1-O2	88.10(12)	N2-Ni1-O6	98.62(12)	N1-Ni1-N2	89.3(2)
Tm1-Ni1-O7[1]	85.70(9)	O11-Ni1-N1	93.21(14)	N2-Ni1-N1	88.93(14)	N2-Ni1-O7	172.2(2)
Bond angles around Tm^{III}		Bond angles around Yb^{III}		Bond angles around Tb^{III}		Bond angles around Eu^{III}	
O5-Tm1-O1	158.19(12)	O8-Yb1-O4	144.22(11)	O11-Tb1-O5	66.64(8)	O2-Eu-O5	121.33(14)
O4-Tm1-O1	135.65(11)	O5-Yb1-O4	66.02(10)	O11-Tb1-O4	94.21(9)	O2-Eu-O7	60.93(14)
O4-Tm1-O5	65.94(10)	O5-Yb1-O8	78.92(10)	O11-Tb1-O10	73.94(9)	O2-Eu-O1	59.04(13)
O2-Tm1-O1	65.37(11)	O1-Yb1-O4	136.86(11)	O11-Tb1-O9	80.33(9)	O10-Eu-O2	95.68(15)
O2-Tm1-O5	136.37(11)	O1-Yb1-O8	78.25(11)	O11-Tb1-O1	142.31(8)	O10-Eu-O9	129.02(15)
O2-Tm1-O4	70.46(11)	O1-Yb1-O5	157.09(11)	O11-Tb1-O2	142.86(9)	O10-Eu-O5	142.01(14)
O8-Tm1-O1	78.63(14)	O2-Yb1-O4	70.98(11)	O11-Tb1-O6	80.00(8)	O10-Eu-O4	143.22(15)
O8-Tm1-O5	79.63(11)	O2-Yb1-O8	143.04(11)	O11-Tb1-O8	129.75(9)	O10-Eu-O11	73.00(15)
O8-Tm1-O4	144.45(12)	O2-Yb1-O5	136.95(10)	O5-Tb1-O1	137.99(8)	O10-Eu-O8	80.21(15)
O8-Tm1-O2	143.52(12)	O2-Yb1-O1	65.88(11)	O5-Tb1-O6	106.54(8)	O10-Eu-O7	80.29(14)
O6-Tm1-O1	84.22(12)	O6[1]-Yb1-O4	74.78(11)	O4-Tb1-O5	59.53(8)	O10-Eu-O1	66.97(14)
O6[1]-Tm1-O1	97.93(12)	O6-Yb1-O4	121.71(11)	O4-Tb1-O1	122.24(8)	O9-Eu-O2	81.93(15)
O6[1]-Tm1-O5	91.25(13)	O6-Yb1-O8	55.28(10)	O4-Tb1-O6	60.33(8)	O9-Eu-O5	69.75(15)
O6-Tm1-O5	82.64(12)	O6[1]-Yb1-O8	117.61(10)	O10-Tb1-O5	139.07(8)	O9-Eu-O4	82.40(14)
O6[1]-Tm1-O4	74.00(11)	O6-Yb1-O5	80.55(11)	O10-Tb1-O4	136.23(9)	O9-Eu-O7	136.13(14)
O6-Tm1-O4	124.92(12)	O6[1]-Yb1-O5	96.11(11)	O10-Tb1-O1	72.81(8)	O9-Eu-O1	68.64(14)
O6[1]-Tm1-O2	75.24(12)	O6-Yb1-O1	84.94(11)	O10-Tb1-O2	96.67(9)	O4-Eu-O2	67.23(14)
O6-Tm1-O2	123.10(12)	O6[1]-Yb1-O1	92.76(11)	O10-Tb1-O6	76.03(9)	O4-Eu-O5	59.09(14)
O6[1]-Tm1-O8	116.98(10)	O6-Yb1-O2	126.19(11)	O10-Tb1-O8	137.19(9)	O4-Eu-O7	62.93(14)
O6-Tm1-O8	54.81(10)	O6[1]-Yb1-O2	74.35(11)	O9-Tb1-O5	84.49(9)	O4-Eu-O1	121.16(14)
O9-Tm1-O1	94.94(10)	O9-Yb1-O4	83.81(11)	O9-Tb1-O4	142.19(9)	O11-Eu-O2	137.73(15)
O9-Tm1-O5	83.84(11)	O9-Yb1-O8	84.90(11)	O9-Tb1-O10	78.43(9)	O11-Eu-O9	137.00(15)
O9-Tm1-O4	83.00(11)	O9-Yb1-O5	82.66(10)	O9-Tb1-O1	76.10(9)	O11-Eu-O5	73.52(15)
O9-Tm1-O2	92.52 (12)	O9-Yb1-O1	97.15(10)	O9-Tb1-O2	133.96(9)	O11-Eu-O4	97.43(15)
O9-Tm1-O8	84.86(12)	O9-Yb1-O2	90.77(11)	O9-Tb1-O6	151.19(8)	O11-Eu-O7	76.91(14)
O9-Tm1-O6	139.12 (15)	O9-Yb1-O6[1]	156.93(12)	O9-Tb1-O8	73.06(9)	O11-Eu-O1	138.50(14)
O9-Tm1-O6[1]	156.44(13)	O9-Yb1-O6	138.99(12)	O1-Tb1-O6	108.32(8)	O8-Eu-O2	141.19(15)
				O2-Tb1-O5	121.34(8)	O8-Eu-O9	71.82(15)
				O2-Tb1-O4	67.51(8)	O8-Eu-O5	75.88(15)
				O2-Tb1-O1	59.10(8)	O8-Eu-O4	133.69(15)
				O2-Tb1-O6	62.87(8)	O8-Eu-O11	78.27(15)
				O8-Tb1-O5	69.00(8)	O8-Eu-O7	151.99(14)
				O8-Tb1-O4	82.72(9)	O8-Eu-O1	84.54(15)
				O8-Tb1-O1	69.81(8)	O7-Eu-O5	108.88(13)
				O8-Tb1-O2	81.57(9)	O1-Eu-O5	137.73(14)
				O8-Tb1-O6	135.64(8)	O1-Eu-O7	105.92(13)
[1]1-X,1-Y,2-Z		[1]2-X,-Y,1-Z					

Table S**3** Selected coordination bond distances (Å) and angles (°) for complex **9-12**.

9		**10**		**11**		**12**	
Selected bond distances around Ni^{II}							
Gd1-Ni1	3.3141(6)	Ho1-Ni1	3.2841(5)	Tb1-Ni1	3.4285(12)	Dy1-Ni1	3.4159(11)
Ni1-O2	2.035(2)	Ni1-O4	2.040(3)	Ni1-O4	1.997(5)	Ni1-O4	2.005(5)
Ni1-O4	2.009(2)	Ni1-O2	2.022(2)	Ni1-O12	2.164(6)	Ni1-N1	2.019(7)
Ni1-O7	2.053(2)	Ni1-O7	2.042(3)	Ni1-O2	2.032(5)	Ni1-O2	2.025(5)
Ni1-O6	2.044(2)	Ni1-O6	2.049(3)	Ni1-O6	2.080(6)	Ni1-O12	2.151(6)
Ni1-N2	2.022(2)	Ni1-N2	2.024(3)	Ni1-N2	2.022(7)	Ni1-N2	2.021(7)
Ni1-N1	2.048(3)	Ni1-N1	2.049(3)	Ni1-N1	2.026(7)	Ni1-O6	2.087(6)
Selected bond distances around Gd^{III}		Selected bond distances around europium Ho^{III}		Selected bond distances around Tb^{III}		Selected bond distances around Dy^{III}	

Gd1-O2	2.4028(19)	Ho1-O4	2.358(2)	Tb1-O1	2.626(5)	Dy1-O4	2.321(5)
Gd1-O9	2.2861(19)	Ho1-O1	2.732(3)	Tb1-O4	2.338(5)	Dy1-O5	2.593(5)
Gd1-O11	2.345(2)	Ho1-O2	2.306(3)	Tb1-O2	2.304(5)	Dy1-O2	2.304(5)
Gd1-O5	2.725(2)	Ho1-O10	2.272(2)	Tb1-O8	2.364(5)	Dy1-O1	2.625(5)
Gd1-O4	2.3525(19)	Ho1-O9	2.293(2)	Tb1-O9	2.357(6)	Dy1-O11	2.447(6)
Gd1-O8	2.3271(19)	Ho1-O11	2.307(2)	Tb1-O7	2.379(6)	Dy1-O8	2.349(6)
Gd1-O10	2.3190(19)	Ho1-O5	2.670(3)	Tb1-O11	2.464(5)	Dy1-O9	2.352(5)
Gd1-O7	2.755(2)	Ho1-O7	2.770(3)	Tb1-O5	2.598(6)	Dy1-O7	2.358(7)
Gd1-O1	2.664(2)	Ho1-O8	2.257(3)	Tb1-O10	2.346(6)	Dy1-O10	2.329(6)
9		**10**		**11**		**12**	
Bond angles containing both Gd^{III} and Ni^{II}		Bond angles containing both Er^{III} and Ni^{II}		Bond angles containing both Tb^{III} and Ni^{II}		Bond angles containing both Dy^{III} and Ni^{II}	
Ni1-O2-Gd1	96.27(7)	Ni1-O4-Ho1	96.34(10)	Ni1-O4-Tb1	104.3(2)	Ni1-O4-Dy1	104.0(2)
Ni1-O4-Gd1	98.61(8)	Ni1-O2-Ho1	98.51(10)	Ni1-O2-Tb1	104.3(2)	Ni1-O2-Dy1	104.0(2)
Ni1-O7-Gd1	85.82(7)	Ni1-O7-Ho1	84.65(9)				
Bond angles around Ni^{II}							
9		**10**		**11**		**12**	
O2-Ni1-O7	79.50(8)	O4-Ni1-O7	79.54(11)	O4-Ni1-O12	86.6(2)	O4-Ni1-N1	172.1(2)
O2-Ni1-O6	168.06(8)	O4-Ni1-O6	167.94(10)	O4-Ni1-O2	81.3(2)	O4-Ni1-O2	81.3(2)
O2-Ni1-N1	102.45(9)	O4-Ni1-N1	102.68(12)	O4-Ni1-O6	88.2(2)	O4-Ni1-O12	86.1(2)
O4-Ni1-O2	81.17(8)	O2-Ni1-O4	80.53(10)	O4-Ni1-N2	91.9(3)	O4-Ni1-N2	92.2(3)
O4-Ni1-O7	82.63(8)	O2-Ni1-O7	82.93(10)	O4-Ni1-N1	172.2(3)	O4-Ni1-O6	89.0(2)
O4-Ni1-O6	94.00(8)	O2-Ni1-O6	94.68(10)	O2-Ni1-O12	87.5(2)	N1-Ni1-O2	91.0(2)
O4-Ni1-N2	170.31(10)	O2-Ni1-N2	169.40(13)	O2-Ni1-O6	92.0(2)	N1-Ni1-O12	95.4(3)
O4-Ni1-N1	90.25(9)	O2-Ni1-N1	90.14(11)	O6-Ni1-O12	174.7(2)	N1-Ni1-N2	95.7(3)
O6-Ni1-O7	89.08(8)	O7-Ni1-O6	88.93(11)	N2-Ni1-O12	86.7(3)	N1-Ni1-O6	89.5(3)
O6-Ni1-N1	88.42(9)	O7-Ni1-N1	172.31(11)	N2-Ni1-O2	171.3(3)	O2-Ni1-O12	87.4(2)
N2-Ni1-O2	89.62(9)	N2-Ni1-O4	88.32(13)	N2-Ni1-O6	93.1(3)	O2-Ni1-O6	92.4(2)
N2-Ni1-O7	98.71(9)	N2-Ni1-O7	89.39(12)	N2-Ni1-N1	95.8(3)	N2-Ni1-O2	171.3(3)
N2-Ni1-O6	95.61(10)	N2-Ni1-O6	98.55(12)	N1-Ni1-O12	95.3(3)	N2-Ni1-O12	86.4(2)
N2-Ni1-N1	88.81(10)	N2-Ni1-N1	95.84(13)	N1-Ni1-O2	91.2(3)	N2-Ni1-O6	93.3(3)
N1-Ni1-O7	172.27(9)	N1-Ni1-O6	88.87(13)	N1-Ni1-O6	90.0(3)	O6-Ni1-O12	175.1(2)
Bond angles around Gd^{III}		Bond angles around Ho^{III}		Bond angles around Tb^{III}		Bond angles around Dy^{III}	
O2-Gd1-O5	121.70(6)	O4-Ho1-O1	123.35(8)	O4-Tb1-O1	121.77(17)	O4-Dy1-O5	62.35(17)
O2-Gd1-O7	60.45(6)	O4-Ho1-O5	59.75(9)	O4-Tb1-O8	144.15(19)	O4-Dy1-O1	121.90(18)
O2-Gd1-O1	59.52(6)	O4-Ho1-O7	60.59(8)	O4-Tb1-O9	106.94(19)	O4-Dy1-O11	73.93(18)
O9-Gd1-O2	94.86(7)	O1-Ho1-O7	108.60(8)	O4-Tb1-O7	73.43(19)	O4-Dy1-O8	106.39(19)
O9-Gd1-O11	129.43(7)	O2-Ho1-O4	68.52(9)	O4-Tb1-O11	74.15(17)	O4-Dy1-O9	144.29(19)
O9-Gd1-O5	142.20(6)	O2-Ho1-O1	59.17(8)	O4-Tb1-O5	62.26(19)	O4-Dy1-O7	74.0(2)
O9-Gd1-O4	142.68(7)	O2-Ho1-O11	81.84(9)	O4-Tb1-O10	132.05(19)	O4-Dy1-O10	131.74(19)
O9-Gd1-O8	73.29(7)	O2-Ho1-O5	122.30(8)	O2-Tb1-O1	62.73(18)	O5-Dy1-O1	134.78(19)
O9-Gd1-O10	80.10(7)	O2-Ho1-O7	63.27(8)	O2-Tb1-O4	68.83(18)	O2-Dy1-O4	69.20(17)
O9-Gd1-O7	79.92(7)	O10-Ho1-O4	142.06(10)	O2-Tb1-O8	90.85(18)	O2-Dy1-O5	128.83(17)
O9-Gd1-O1	66.97(7)	O10-Ho1-O1	76.05(9)	O2-Tb1-O9	143.97(19)	O2-Dy1-O1	62.64(18)
O11-Gd1-O2	82.82(7)	O10-Ho1-O2	134.07(9)	O2-Tb1-O7	74.72(19)	O2-Dy1-O11	80.27(19)
O11-Gd1-O5	69.82(7)	O10-Ho1-O9	78.85(9)	O2-Tb1-O11	80.48(18)	O2-Dy1-O8	143.7(2)
O11-Gd1-O4	82.36(7)	O10-Ho1-O11	73.54(9)	O2-Tb1-O5	128.28(18)	O2-Dy1-O9	90.45(18)
O11-Gd1-O7	136.26(6)	O10-Ho1-O5	83.82(9)	O2-Tb1-O10	135.9(2)	O2-Dy1-O7	74.9(2)
O11-Gd1-O1	68.78(6)	O10-Ho1-O7	150.37(8)	O8-Tb1-O1	67.21(17)	O2-Dy1-O10	135.8(2)
O5-Gd1-O7	108.42(6)	O9-Ho1-O4	135.65(9)	O8-Tb1-O7	72.8(2)	O11-Dy1-O5	71.78(19)
O4-Gd1-O2	67.17(7)	O9-Ho1-O1	72.09(8)	O8-Tb1-O11	133.14(18)	O11-Dy1-O1	67.87(17)
O4-Gd1-O5	59.10(6)	O9-Ho1-O2	95.43(9)	O8-Tb1-O5	139.67(18)	O8-Dy1-O5	69.19(19)
O4-Gd1-O7	62.76(6)	O9-Ho1-O11	137.46(8)	O9-Tb1-O1	131.21(19)	O8-Dy1-O1	131.63(19)
O4-Gd1-O1	121.24(6)	O9-Ho1-O5	139.49(9)	O9-Tb1-O8	72.06(18)	O8-Dy1-O11	134.6(2)
O8-Gd1-O2	136.43(7)	O9-Ho1-O7	75.17(8)	O9-Tb1-O7	70.0(2)	O8-Dy1-O9	72.61(19)
O8-Gd1-O11	137.17(7)	O11-Ho1-O4	82.87(8)	O9-Tb1-O11	134.26(19)	O8-Dy1-O7	69.5(2)
O8-Gd1-O5	73.29(7)	O11-Ho1-O1	70.27(8)	O9-Tb1-O5	69.76(18)	O9-Dy1-O5	139.52(18)
O8-Gd1-O4	96.91(7)	O11-Ho1-O5	68.64(8)	O7-Tb1-O1	119.5(2)	O9-Dy1-O1	67.00(18)
O8-Gd1-O7	76.10(6)	O11-Ho1-O7	136.01(8)	O7-Tb1-O11	144.51(19)	O9-Dy1-O11	132.99(18)
O8-Gd1-O1	138.81(6)	O5-Ho1-O1	137.86(8)	O7-Tb1-O5	105.2(2)	O9-Dy1-O7	72.5(2)
O10-Gd1-O2	142.11(7)	O5-Ho1-O7	107.13(7)	O11-Tb1-O1	67.96(18)	O7-Dy1-O5	105.3(2)
O10-Gd1-O11	72.51(7)	O8-Ho1-O4	93.04(8)	O11-Tb1-O5	71.32(18)	O7-Dy1-O1	119.3(2)
O10-Gd1-O5	76.33(7)	O8-Ho1-O1	142.20(8)	O5-Tb1-O1	134.67(19)	O7-Dy1-O11	144.65(19)
O10-Gd1-O4	134.24(7)	O8-Ho1-O2	142.08(9)	O10-Tb1-O1	74.9(2)	O10-Dy1-O5	74.5(2)
O10-Gd1-O8	78.35(7)	O8-Ho1-O10	80.72(9)	O10-Tb1-O8	83.2(2)	O10-Dy1-O1	74.8(2)
O10-Gd1-O7	151.15(6)	O8-Ho1-O9	74.47(9)	O10-Tb1-O9	74.7(2)	O10-Dy1-O11	72.9(2)
O10-Gd1-O1	84.53(7)	O8-Ho1-O11	130.16(10)	O10-Tb1-O7	141.9(2)	O10-Dy1-O8	75.2(2)
O1-Gd1-O5	137.93(6)	O8-Ho1-O5	66.72(9)	O10-Tb1-O11	72.7(2)	O10-Dy1-O9	83.4(2)
O1-Gd1-O7	106.33(6)	O8-Ho1-O7	78.82(9)	O10-Tb1-O5	74.8(2)	O10-Dy1-O7	141.7(2)

Table S**4** Selected coordination bond distances (Å) and angles (°) for complex **13-15**.

13		**14**		**15**	
Selected bond distances around Ni^{II}					
Eu1-Ni1	3.4469(14)	Gd1-Ni1	3.4411(14)	Ho1-Ni1	3.4082(13)
Ni1-O4	1.999(7)	Ni1-O2	2.003(6)	Ni1-O2	1.989(6)
Ni1-O2	2.023(6)	Ni1-O12	2.157(7)	Ni1-N2	2.021(8)
Ni1-O12	2.175(7)	Ni1-O4	2.026(6)	Ni1-N1	2.022(8)
Ni1-O7	2.079(6)	Ni1-O6	2.090(6)	Ni1-O6	2.089(6)
Ni1-N2	2.021(9)	Ni1-N1	2.023(9)	Ni1-O4	2.030(6)
Ni1-N1	2.022(10)	Ni1-N2	2.029(9)	Ni1-O12	2.154(6)
Selected bond distances around Eu^{II}		Selected bond distances around europium Gd^{III}		Selected bond distances around Ho^{III}	
Eu1-O8	2.390(6)	Gd1-O2	2.348(6)	Ho1-O10	2.439(6)
Eu1-O4	2.367(7)	Gd1-O5	2.618(6)	Ho1-O2	2.310(6)
Eu1-O1	2.620(6)	Gd1-O7	2.392(8)	Ho1-O8	2.344(6)
Eu1-O2	2.332(6)	Gd1-O11	2.477(6)	Ho1-O5	2.632(6)
Eu1-O5	2.610(7)	Gd1-O8	2.382(7)	Ho1-O1	2.593(6)
Eu1-O6	2.411(8)	Gd1-O4	2.333(6)	Ho1-O11	2.320(7)
Eu1-O11	2.494(6)	Gd1-O9	2.381(6)	Ho1-O4	2.288(6)
Eu1-O9	2.392(7)	Gd1-O1	2.606(7)	Ho1-O7	2.351(7)
Eu1-O10	2.383(7)	Gd1-O10	2.352(7)	Ho1-O9	2.351(6)
13		**14**		**15**	
Bond angles containing both Eu^{III} and Ni^{II}		Bond angles containing both Gd^{III} and Ni^{II}		Bond angles containing both Ho^{III} and Ni^{II}	
Ni1-O4-Eu1	103.9(3)	Ni1-O2-Gd1	104.3(2)	Ni1-O2-Ho1	104.7(2)
Ni1-O2-Eu1	104.4(2)	Ni1-O4-Gd1	104.0(2)	Ni1-O4-Ho1	104.1(2)
Bond angles around nickel					
13		**14**		**15**	
O4-Ni1-O2	82.0(3)	O2-Ni1-O12	85.9(2)	O2-Ni1-N2	171.9(3)
O4-Ni1-O12	86.3(2)	O2-Ni1-O4	81.8(3)	O2-Ni1-N1	92.4(3)
O4-Ni1-O7	88.2(2)	O2-Ni1-O6	88.5(2)	O2-Ni1-O6	88.8(2)
O4-Ni1-N2	91.8(3)	O2-Ni1-N1	91.8(3)	O2-Ni1-O4	80.8(2)
O4-Ni1-N1	172.6(3)	O2-Ni1-N2	172.3(3)	O2-Ni1-O12	86.2(2)
O2-Ni1-O12	87.7(2)	O4-Ni1-O12	87.2(2)	N2-Ni1-O6	89.8(3)
O2-Ni1-O7	91.9(2)	O4-Ni1-O6	92.3(2)	N2-Ni1-O12	95.2(3)
O7-Ni1-O12	174.5(2)	O4-Ni1-N2	90.8(3)	N1-Ni1-N2	95.7(4)
N2-Ni1-O2	171.8(3)	O6-Ni1-O12	174.4(3)	N1-Ni1-O6	93.2(3)
N2-Ni1-O12	86.4(3)	N1-Ni1-O12	86.2(3)	N1-Ni1-O4	171.2(3)
N2-Ni1-O7	93.3(3)	N1-Ni1-O4	171.1(3)	N1-Ni1-O12	86.5(3)
N2-Ni1-N1	95.4(4)	N1-Ni1-O6	93.6(3)	O6-Ni1-O12	175.0(3)
N1-Ni1-O2	91.0(3)	N1-Ni1-N2	95.9(4)	O4-Ni1-N2	91.2(3)
N1-Ni1-O12	95.8(3)	N2-Ni1-O12	95.8(3)	O4-Ni1-O6	92.3(2)
N1-Ni1-O7	89.7(3)	N2-Ni1-O6	89.8(3)	O4-Ni1-O12	87.4(2)
Bond angles around Eu^{III}		Bond angles around Gd^{III}		Bond angles around Ho^{III}	
O8-Eu1-O1	67.6(2)	O2-Gd1-O5	121.5(2)	O10-Ho1-O5	67.84(19)
O8-Eu1-O5	140.2(2)	O2-Gd1-O7	73.4(2)	O10-Ho1-O1	71.7(2)
O8-Eu1-O6	72.7(2)	O2-Gd1-O11	74.1(2)	O2-Ho1-O10	74.0(2)
O8-Eu1-O11	133.6(2)	O2-Gd1-O8	106.8(2)	O2-Ho1-O8	144.0(2)
O8-Eu1-O9	71.4(2)	O2-Gd1-O9	143.3(2)	O2-Ho1-O5	121.88(19)
O4-Eu1-O8	143.8(2)	O2-Gd1-O1	62.0(2)	O2-Ho1-O1	63.0(2)
O4-Eu1-O1	121.2(2)	O2-Gd1-O10	131.6(2)	O2-Ho1-O11	132.5(2)
O4-Eu1-O5	61.9(2)	O7-Gd1-O5	118.5(2)	O2-Ho1-O7	73.8(2)
O4-Eu1-O6	73.2(2)	O7-Gd1-O11	144.3(2)	O2-Ho1-O9	106.1(2)
O4-Eu1-O11	74.1(2)	O7-Gd1-O1	105.1(2)	O8-Ho1-O10	132.9(2)
O4-Eu1-O9	107.5(2)	O11-Gd1-O5	68.5(2)	O8-Ho1-O5	66.87(19)
O4-Eu1-O10	131.2(2)	O11-Gd1-O1	71.3(2)	O8-Ho1-O1	139.5(2)
O2-Eu1-O8	91.0(2)	O8-Gd1-O5	131.5(2)	O8-Ho1-O7	72.4(2)
O2-Eu1-O4	68.3(2)	O8-Gd1-O7	70.0(2)	O8-Ho1-O9	73.0(2)
O2-Eu1-O1	62.6(2)	O8-Gd1-O11	134.4(2)	O1-Ho1-O5	134.4(2)
O2-Eu1-O5	127.4(2)	O8-Gd1-O9	71.9(2)	O11-Ho1-O10	73.2(2)
O2-Eu1-O6	74.4(2)	O8-Gd1-O1	69.9(2)	O11-Ho1-O8	83.0(2)
O2-Eu1-O11	81.0(2)	O4-Gd1-O2	68.6(2)	O11-Ho1-O5	74.4(2)
O2-Eu1-O9	143.5(2)	O4-Gd1-O5	62.3(2)	O11-Ho1-O1	74.6(2)
O2-Eu1-O10	135.9(2)	O4-Gd1-O7	74.4(2)	O11-Ho1-O7	141.8(2)
O5-Eu1-O1	134.8(2)	O4-Gd1-O11	80.6(2)	O11-Ho1-O9	75.6(2)
O6-Eu1-O1	119.3(2)	O4-Gd1-O8	143.6(2)	O4-Ho1-O10	80.0(2)
O6-Eu1-O5	105.0(2)	O4-Gd1-O9	90.6(2)	O4-Ho1-O2	69.1(2)

O6-Eu1-O11	144.5(2)	O4-Gd1-O1	127.9(2)	O4-Ho1-O8	90.3(2)
O11-Eu1-O1	68.3(2)	O4-Gd1-O10	136.1(2)	O4-Ho1-O5	62.6(2)
O11-Eu1-O5	70.6(2)	O9-Gd1-O5	67.4(2)	O4-Ho1-O1	129.2(2)
O9-Eu1-O1	131.2(2)	O9-Gd1-O7	72.0(2)	O4-Ho1-O11	135.4(2)
O9-Eu1-O5	70.7(2)	O9-Gd1-O11	133.9(2)	O4-Ho1-O7	74.7(2)
O9-Eu1-O6	70.0(2)	O9-Gd1-O1	140.0(2)	O4-Ho1-O9	143.6(2)
O9-Eu1-O11	134.2(2)	O1-Gd1-O5	135.6(2)	O7-Ho1-O10	144.4(2)
O10-Eu1-O8	84.4(2)	O10-Gd1-O5	75.7(2)	O7-Ho1-O5	119.1(2)
O10-Eu1-O1	75.3(2)	O10-Gd1-O7	142.5(3)	O7-Ho1-O1	105.9(2)
O10-Eu1-O5	74.6(2)	O10-Gd1-O11	72.3(2)	O9-Ho1-O10	134.8(2)
O10-Eu1-O6	143.1(2)	O10-Gd1-O8	75.2(3)	O9-Ho1-O5	132.0(2)
O10-Eu1-O11	71.4(2)	O10-Gd1-O9	84.5(2)	O9-Ho1-O1	69.0(2)
O10-Eu1-O9	75.5(2)	O10-Gd1-O1	74.8(2)	O9-Ho1-O7	69.5(2)

Table S**5** Hydrogen bonds of complexes **1-15**.

	D−H···A	**D(D-H)**	**D(D···A)**	**D(H···A)**	**∠(D-H-A)**
1	O6−H3···O7[1]	0.8204(45)	2.8269(51)	2.0320(28)	163.068(29)
	O11−H11···O10	0.865537(26)	2.6334(46)	1.8176(25)	156.761(24)
2	O6−H3···O7[1]	0.8191 (39)	2.8037(47)	1.9880(26)	173.730(27)
	O11−H11···O10	0.8639(24)	2.6303(37)	1.8112(24)	157.567(23)
3	O6−H3···O7[1]	0.8186(38)	2.8039(47)	2.0040(27)	165.456(28)
	O11−H11···O10	0.8695(23)	2.6329(37)	1.8002(23)	1579.726(22)
4	O6−H3···O7[1]	0.8187(28)	2.7962(38)	1.9797(25)	174.943(20)
	O11−H11···O10	0.8689(22)	2.6201(35)	1.7894(22)	159.238(21)
5	O6−H3···O7[1]	0.8201(52)	2.8131(65)	2.2642 (38)	124.697(37)
	O11−H11···O10	0.8618(34)	2.6411(55)	1.8458(37)	152.657(31)
6	O6−H3···O7[1]	0.8196(47)	2.8092(51)	2.0562(27)	152.586(30)
	O11−H11···O10	0.8720(25)	2.6273(45)	1.7992(25)	157.785(24)
7	O3−H3···O6	0.8199(32)	2.7044(40)	1.9187(26)	160.220 (23)
8	O3−H3···O6	0.8186(47)	2.7127(66)	2.0363(44)	139.679(36)
9	O3−H3···O6	0.8192(24)	2.7061(37)	1.9251(20)	159.067(17)
10	O3−H3···O6	0.8200(29)	2.7015(38)	1.9252(24)	157.616(22)
11	O3−H3···O6	0.8203(98)	2.7427(11)	1.9430(56)	164.670 (67)
12	O3−H3···O6	0.8192(10)	2.7456 (11)	1.9345(56)	170.419(67)
13	O3−H3···O6	0.8187(10)	2.7308(13)	1.9463(74)	160.217(71)
14	O3−H3···O6	0.8193(99)	2.7455(12)	1.9412(75)	166.900(68)
15	O3−H3···O6	0.8200(10)	2.7456 (12)	1.9341(63)	170.169(70)

Table S**6** $Ln^{III}\cdots M^{II}$ Distances(Å) and Selected Coordination Bond Distances (Å) and angles(°).

	Ni···Ln	Ln···Ln	$(Ln\text{-}O_{phenoxy})_{avg}$	$(Ln\text{-}O_{methoxy})_{avg}$	$(Ln\text{-}O_{carbonate})_{avg}$	Ln-OO-Ni dihedral
1 (Tb)	3.4176	4.0160	2.3092	2.5143	2.3926	12.333

2 (Dy)	3.3919	3.9934	2.2901	2.4951	2.3692	12.680
3 (Ho)	3.3907	3.9755	2.2788	2.4841	2.3616	12.893
4 (Er)	3.3699	3.9395	2.2625	2.4715	2.3453	12.783
5 (Tm)	3.3847	3.9523	2.2660	2.4767	2.3418	13.311
6 (Yb)	3.3644	3.9109	2.2543	2.4692	2.3233	12.905
	Ni…Ln	$(Ln\text{-}O_{dbm})_{avg}$	$(Ln\text{-}O_{phenoxy})_{avg}$	Ln-O7	$(Ln\text{-}O_{methoxy})_{avg}$	
8 (Eu)	3.3179	2.3322	2.3967	2.7383	2.7148	40.394
9 (Gd)	3.3142	2.3192	2.3777	2.7554	2.6943	39.330
7 (Tb)	3.3046	2.3041	2.3565	2.7662	2.7087	38.910
10 (Ho)	3.2841	2.2821	2.3319	2.7692	2.7012	38.571
	Ni…Ln	$(Ln\text{-}O_{hfac})_{avg}$	$Ln\text{-}O_{phenoxy})_{avg}$	$Ln\text{-}O_{tfa}$	$(Ln\text{-}O_{methoxy})_{avg}$	
13 (Eu)	3.4468	2.4147	2.3494	2.4108	2.6153	10.198
14 (Gd)	3.4411	2.3980	2.3405	2.3913	2.6115	10.882
11 (Tb)	3.4285	2.3828	2.3207	2.3784	2.6122	10.615
12 (Dy)	3.4159	2.3697	2.3133	2.3583	2.6086	11.156
15 (Ho)	3.4083	2.3633	2.2989	2.3513	2.6121	10.945

Table S7 Geometry Analysis using the Shape v 2.0 Software for Lanthanide Ion in Complex **1-6**.

```
-------------------------------------------------------------------------------
S H A P E   v2.0         Continuous Shape Measures calculation
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Contact:  llunell@ub.edu
-------------------------------------------------------------------------------

OP-8             1 D8h   Octagon
HPY-8            2 C7v   Heptagonal pyramid
HBPY-8           3 D6h   Hexagonal bipyramid
CU-8             4 Oh    Cube
SAPR-8           5 D4d   Square antiprism
TDD-8            6 D2d   Triangular dodecahedron
JGBF-8           7 D2d   Johnson gyrobifastigium J26
JETBPY-8         8 D3h   Johnson elongated triangular bipyramid J14
JBTPR-8          9 C2v   Biaugmented trigonal prism J50
BTPR-8          10 C2v   Biaugmented trigonal prism
JSD-8           11 D2d   Snub diphenoid J84
TT-8            12 Td    Triakis tetrahedron
ETBPY-8         13 D3h   Elongated trigonal bipyramid
```

[ML8]	OP-8	HPY-8	HBPY-8	CU-8	SAPR-8	TDD-8	JGBF-8	JETBPY-8	JBTPR-8	BTPR-8	JSD-8	TT-8	ETBPY-8
1 (Tb)	31.812	23.663	12.455	11.796	4.858	3.391	11.876	26.360	4.777	4.125	5.128	12.617	22.919
2 (Dy)	31.937	23.915	12.541	11.847	4.884	3.334	11.891	26.349	4.777	4.116	5.085	12.651	22.787
3 (Ho)	31.844	24.043	12.607	11.814	4.830	3.296	11.865	26.314	4.687	4.031	5.018	12.620	22.778

4 (Er)	31.729	23.909	12.623	11.744	4.655	3.181	11.856	26.380	4.529	3.859	4.921	12.555	22.857
5 (Tm)	33.119	24.084	13.164	12.831	5.768	3.355	11.769	25.653	4.484	3.651	5.023	13.454	21.860
6 (Yb)	31.876	24.128	12.702	11.823	4.699	3.148	11.882	26.386	4.489	3.802	4.938	12.604	22.784

Table S**8** Geometry Analysis using the Shape v 2.0 Software for Lanthanide Ion in Complex **7-15**.

```
S H A P E   v2.0          Continuous Shape Measures calculation

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                    Contact:  llunell@ub.edu
-------------------------------------------------------------------------------

EP-9              1 D9h   Enneagon
OPY-9             2 C8v   Octagonal pyramid
HBPY-9            3 D7h   Heptagonal bipyramid
JTC-9             4 C3v   Johnson triangular cupola J3
JCCU-9            5 C4v   Capped cube J8
CCU-9             6 C4v   Spherical-relaxed capped cube
JCSAPR-9          7 C4v   Capped square antiprism J10
CSAPR-9           8 C4v   Spherical capped square antiprism
JTCTPR-9          9 D3h   Tricapped trigonal prism J51
TCTPR-9          10 D3h   Spherical tricapped trigonal prism
JTDIC-9          11 C3v   Tridiminished icosahedron J63
HH-9             12 C2v   Hula-hoop
MFF-9            13 Cs    Muffin
```

[ML9]	EP-9	OPY-9	HBPY-9	JTC-9	JCCU-9	CCU-9	JCSAPR-9	CSAPR-9	JTCTPR-9	TCTPR-9	JTDIC-9	HH-9	MFF-9
7 (Tb)	35.004	23.005	17.917	13.691	10.486	10.175	2.690	2.412	2.664	2.952	12.538	10.036	1.669
8 (Eu)	35.337	22.938	17.787	13.838	10.605	10.064	2.658	2.431	2.906	2.958	12.461	9.692	1.680
9 (Gd)	35.143	22.994	17.781	13.696	10.537	10.065	2.717	2.395	2.803	2.928	12.436	9.893	1.629
10 (Ho)	34.602	23.280	18.123	13.535	10.380	10.187	2.595	2.346	2.388	2.878	12.734	10.324	1.656
13 (Eu)	36.800	22.780	18.389	15.658	10.196	9.214	1.374	0.761	3.071	1.722	12.390	9.952	0.534
14 (Gd)	36.898	22.856	18.261	15.844	10.113	9.278	1.363	0.758	3.006	1.774	12.573	9.960	0.537
11 (Tb)	36.690	22.929	18.213	15.735	9.985	9.312	1.258	0.725	2.891	1.793	12.655	10.086	0.551
12 (Dy)	36.794	23.053	18.264	15.857	10.001	9.360	1.234	0.723	2.828	1.821	12.792	10.131	0.553
15 (Ho)	36.825	23.100	18.360	15.983	9.927	9.344	1.177	0.718	2.711	1.873	12.972	10.311	0.561

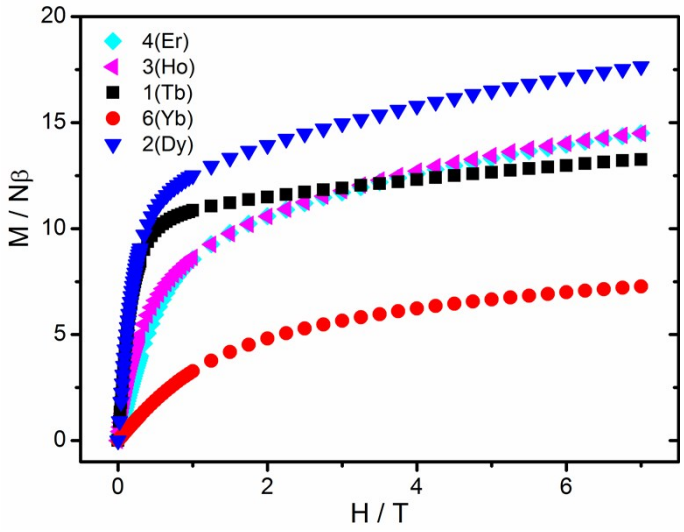

Fig. S**13** Field Dependence of Magnetization for Complex **1**-**4** and **6** at 2.0 K in the field range of 0-70 kOe.

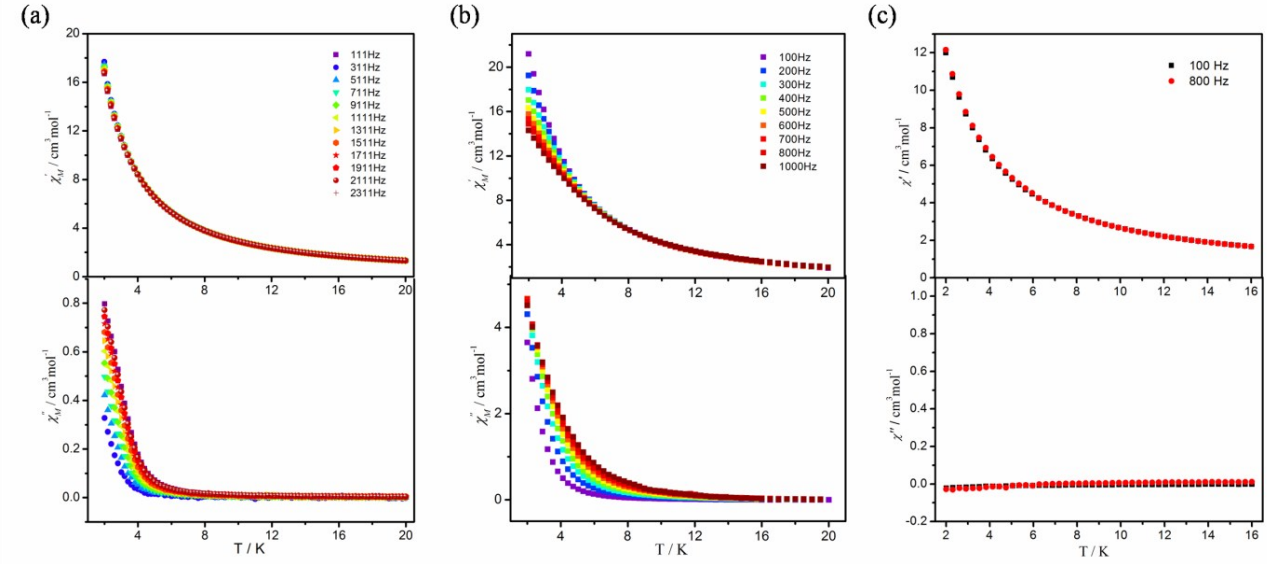

Fig. S**14** Temperature-dependent ac Susceptibility Data for **1**(Tb), **2** (Dy) and **3** (Ho) measured under 0 Oe dc Field at the indicated frequencies.

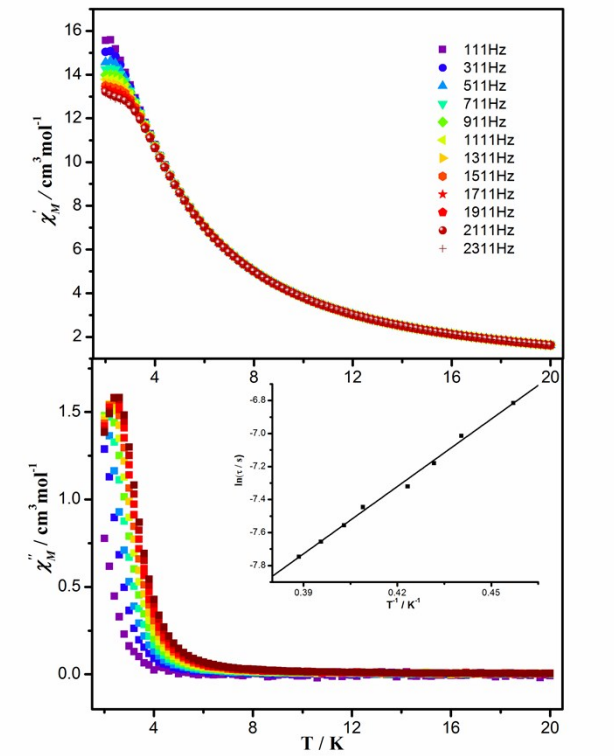

Fig. S**15** Temperature-dependent ac susceptibility data for **1**(Tb) measured under 1200 Oe dc field at the indicated frequencies. Inset: Arrhenius fitting of logarithmic relaxation time vs. reciprocal temperature.

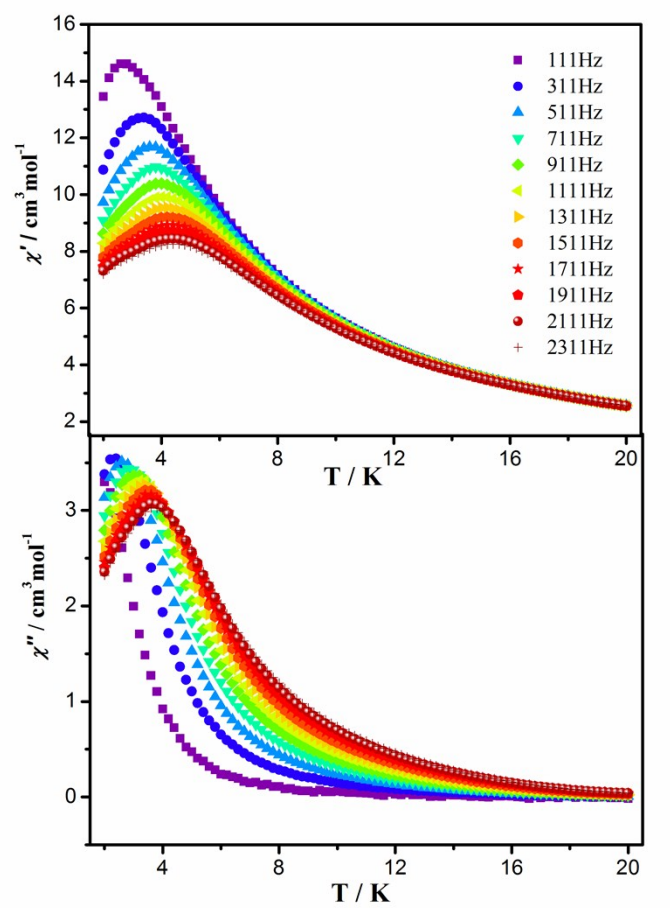

Fig. S**16** Temperature-dependent ac susceptibility data for **2** (Dy) measured under 1200 Oe dc field at the indicated frequencies.

Table S**9** Calculated α values for compound **2**(Dy).

T / K	α	T	α
2	0.370	3.7	0.249
2.5	0.342	3.9	0.246
2.8	0.299	4.0	0.238
3.0	0.285	4.5	0.233
3.1	0.260	5.0	0.207
3.3	0.252	5.5	0.205
3.5	0.251	6.0	0.172

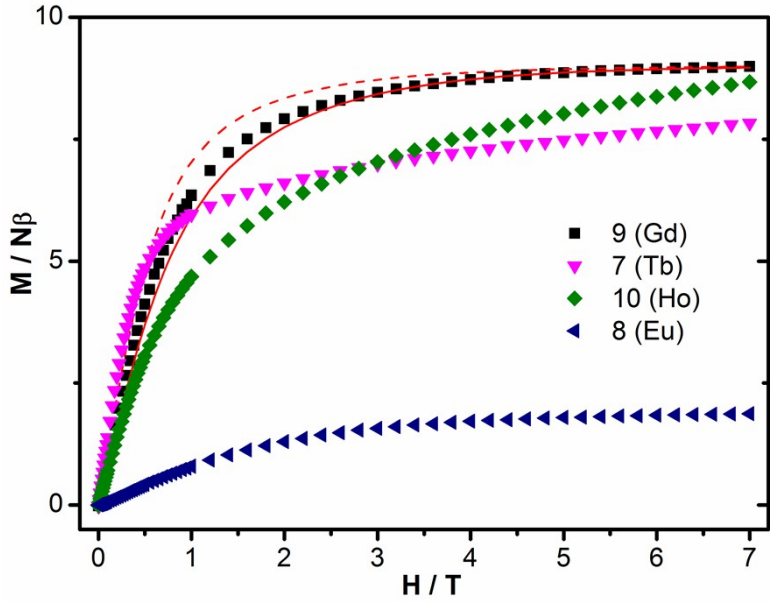

Fig. S**17** Field dependence of magnetization at 2.0 K in the field range of 0-7 T. The red solid line represents the Brillouin function for two isolated Ni^{II} and Gd^{III} centers, and red dashed line represents an S = 9/2 state.

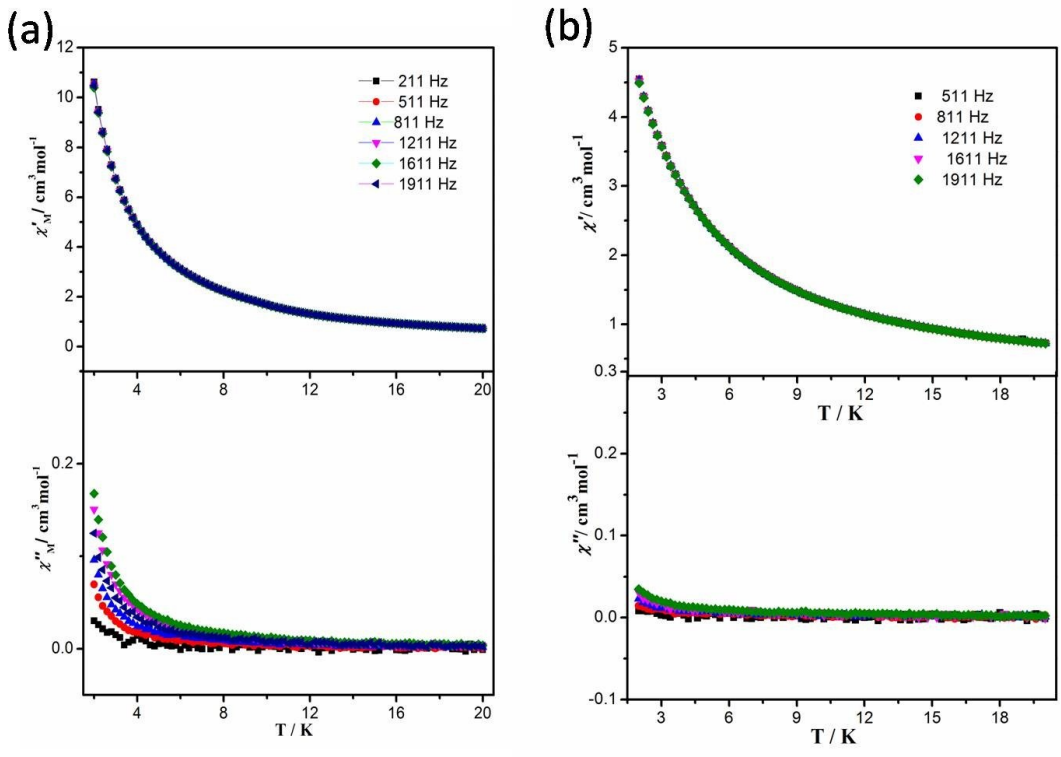

Fig. S**18** Temperature-dependent ac susceptibility data for **7** (a) and **10** (b) measured under 0 Oe dc field at the indicated frequencies.

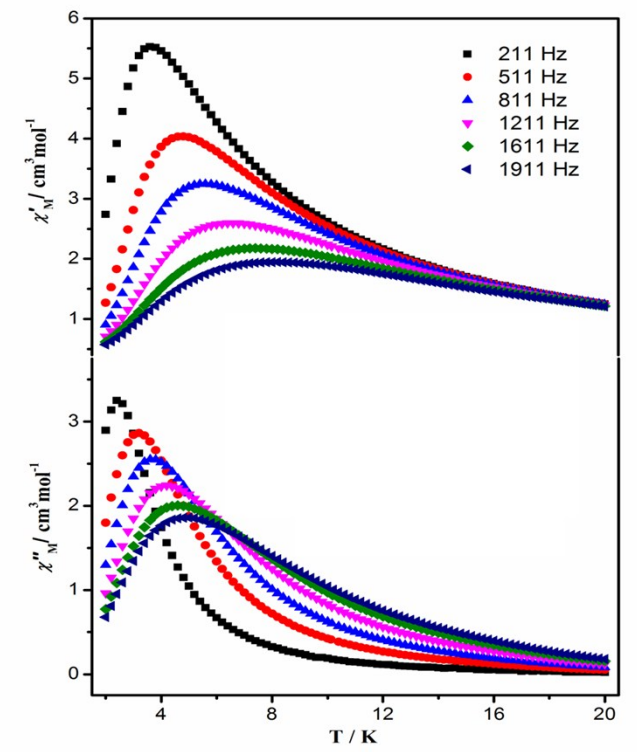

Fig. **S19** Temperature dependent ac susceptibility for **7** (Tb) at the indicated frequencies under 2000 dc field.

Table S**10** Calculated *α* values for compound **7** (Tb).

T / K	α	T	α
2	0.139	3.8	0.121
2.5	0.134	4.0	0.118
3.0	0.138	4.2	0.112
3.5	0126	4.4	0.108

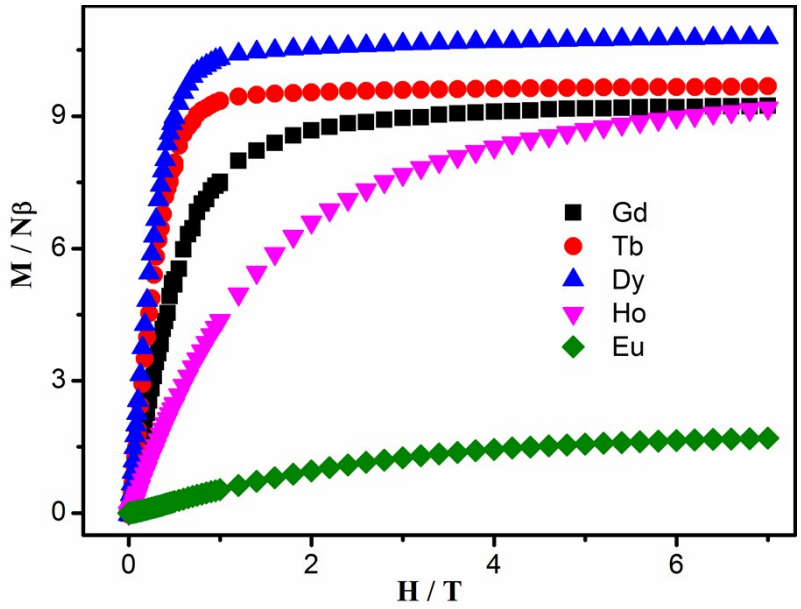

Fig. S**20** Field dependence of magnetization at 2.0 K for complex **11-15** in the field range of 0-7 KOe.

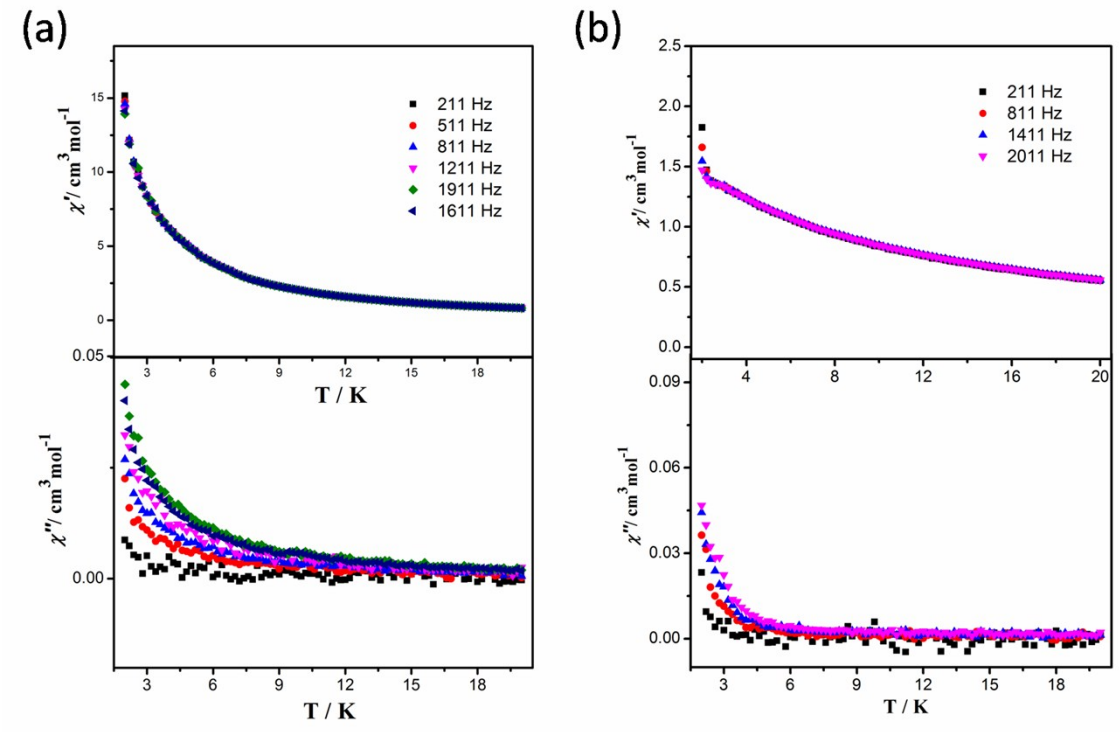

Fig. S**21** Temperature-dependent ac susceptibility data for **11** (a**)** and **15** (b) measured under 0 Oe dc field at the indicated frequencies.

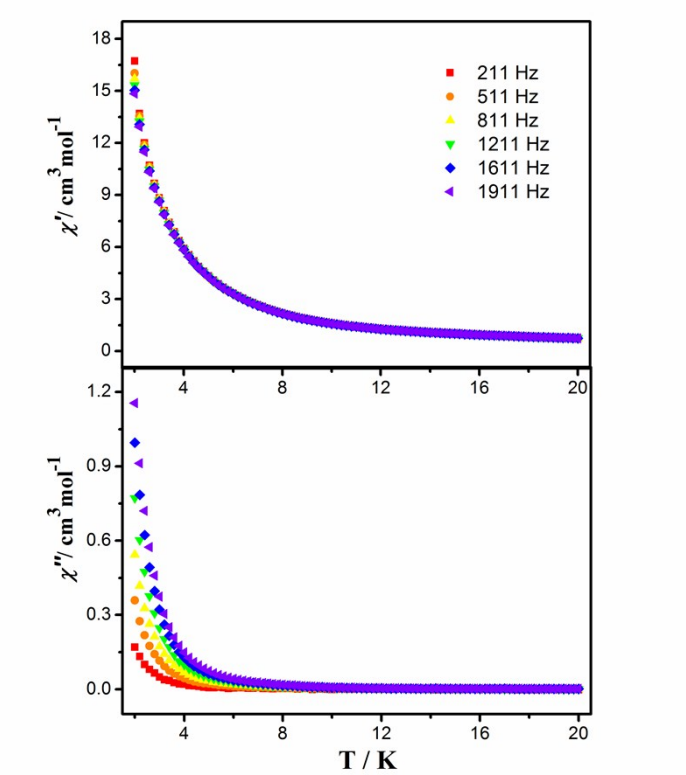

Fig. S**22** Temperature dependent ac susceptibility for **12** (Dy) at the indicated temperatures under 0 Oe dc field. at the indicated frequencies.

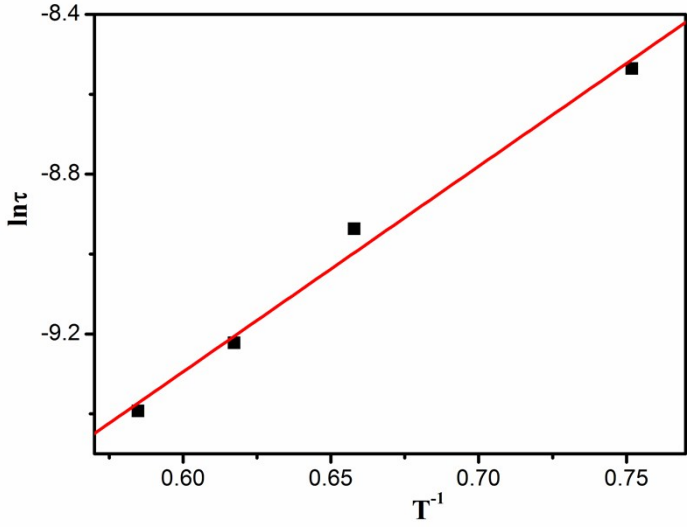

Fig.S23 Plots of lnτ vs. T^{-1} of complex **12** extracted from temperature dependent ac susceptibility data under 2000 Oe.

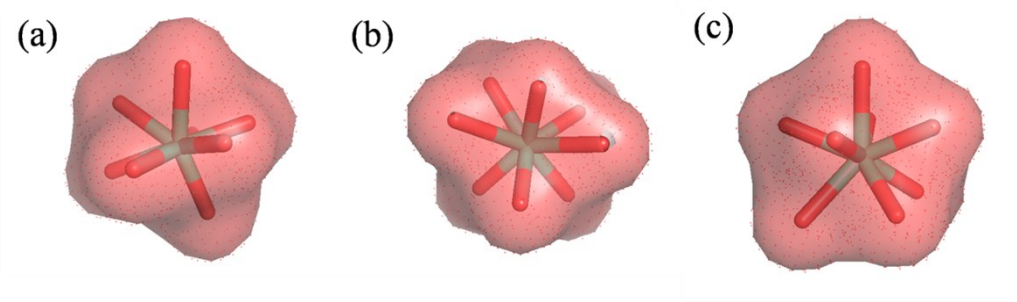

Fig. S24 Electronic environment around Tb^{III} ion in complex **1** (a), complex **7** (b) and complex **11** (c).