Supporting Information

Modulating the magnetization dynamics in Ln-Cu-Rad hetero-tri-spin complexes through *cis/trans* coordination of nitronyl nitroxide radicals around metal center

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Contents

Table S1. Selected bond lengths [Å] and angles [°] for **1**.

Bond distances			
Gd(1)-O(5)	2.364(5)	Cu(1)-O(13)	2.348(5)
Gd(1)-O(10)	2.455(6)	Cu(1)-O(12)	2.279(5)
Gd(1)-O(2)	2.366(5)	Cu(1)-O(11)	1.974(5)
Gd(1)-O(4)	2.370(5)	Cu(1)-N(5)	2.014(6)
Gd(1)-O(9)	2.374(5)	Cu(1)-N(6)	2.019(6)
Gd(1)-O(8)	2.422(6)	O(2)-N(2)	1.319(9)
Gd(1)-O(6)	2.381(5)	O(3)-N(3)	1.270(10)
Gd(1)-O(7)	2.341(5)	O(4)-N(4)	1.310(8)
Cu(1)-O(14)	1.974(5)	O(1)-N(1)	1.275(9)
Angles			
O(5)-Gd(1)-O(10)	69.7(2)	O(7)-Gd(1)-O(2)	145.7(2)
O(5)-Gd(1)-O(2)	82.91(18)	O(7)-Gd(1)-O(4)	144.9(2)
O(5)-Gd(1)-O(4)	120.36(18)	O(7)-Gd(1)-O(9)	83.93(19)
O(5)-Gd(1)-O(9)	138.8(2)	O(7)-Gd(1)-O(8)	71.2(2)
O(5)-Gd(1)-O(8)	134.41(19)	O(7)-Gd(1)-O(6)	76.35(19)
O(5)-Gd(1)-O(6)	70.5(2)	O(14)-Cu(1)-O(13)	85.12(19)
O(2)-Gd(1)-O(10)	74.88(19)	O(14)-Cu(1)-O(12)	86.9(2)
O(2)-Gd(1)-O(4)	69.40(19)	O(14)-Cu(1)-N(5)	175.8(2)
O(2)-Gd(1)-O(9)	92.50(18)	O(14)-Cu(1)-N(6)	87.3(2)
O(2)-Gd(1)-O(8)	139.08(18)	O(12)-Cu(1)-O(13)	166.93(18)
O(2)-Gd(1)-O(6)	123.10(19)	O(11)-Cu(1)-O(14)	87.6(2)
O(4)-Gd(1)-O(10)	140.57(18)	O(11)-Cu(1)-O(13)	84.6(2)
O(4)-Gd(1)-O(9)	95.48(18)	O(11)-Cu(1)-O(12)	84.8(2)
O(4)-Gd(1)-O(8)	75.52(19)	O(11)-Cu(1)-N(5)	89.0(2)
O(4)-Gd(1)-O(6)	82.07(17)	O(11)-Cu(1)-N(6)	174.7(2)
O(9)-Gd(1)-O(10)	69.6(2)	N(5)-Cu(1)-O(13)	92.1(2)
O(9)-Gd(1)-O(8)	70.2(2)	N(5)-Cu(1)-O(12)	95.2(2)
O(9)-Gd(1)-O(6)	139.7(2)	N(5)-Cu(1)-N(6)	96.0(2)
O(8)-Gd(1)-O(10)	127.3(2)	N(6)-Cu(1)-O(13)	93.2(2)
O(6)-Gd(1)-O(10)	133.04(18)	N(6)-Cu(1)-O(12)	96.8(2)
O(6)-Gd(1)-O(8)	70.24(19)	N(2)-O(2)-Gd(1)	131.9(4)
O(7)-Gd(1)-O(5)	77.8(2)	N(4)-O(4)-Gd(1)	133.8(4)
O(7)-Gd(1)-O(10)	71.9(2)		

Table S2. Selected bond lengths [Å] and angles [°] for **2**.

Bond distances			
Dy(1)-O(5)	2.329(7)	Cu(1)-O(13)	2.344(6)
Dy(1)-O(10)	2.452(8)	Cu(1)-O(12)	2.282(6)
Dy(1)-O(2)	2.340(7)	Cu(1)-O(11)	1.971(7)
Dy(1)-O(4)	2.354(6)	Cu(1)-N(5)	2.015(8)
Dy(1)-O(9)	2.338(6)	Cu(1)-N(6)	2.013(8)
Dy(1)-O(8)	2.414(8)	O(2)-N(2)	1.322(11)

Dy(1)-O(6)	2.350(7)	O(3)-N(3)	1.261(12)
Dy(1)-O(7)	2.323(7)	O(4)-N(4)	1.295(11)
Cu(1)-O(14)	1.977(7)	O(1)-N(1)	1.279(12)
Angles			
O(5)-Dy(1)-O(10)	69.9(3)	O(7)-Dy(1)-O(2)	146.5(3)
O(5)-Dy(1)-O(2)	82.6(2)	O(7)-Dy(1)-O(4)	144.7(3)
O(5)-Dy(1)-O(4)	119.6(2)	O(7)-Dy(1)-O(9)	84.5(2)
O(5)-Dy(1)-O(9)	138.8(3)	O(7)-Dy(1)-O(8)	71.3(3)
O(5)-Dy(1)-O(8)	134.7(2)	O(7)-Dy(1)-O(6)	76.7(2)
O(5)-Dy(1)-O(6)	70.5(3)	O(14)-Cu(1)-O(13)	85.0(3)
O(2)-Dy(1)-O(10)	75.2(3)	O(14)-Cu(1)-O(12)	87.6(3)
O(2)-Dy(1)-O(4)	68.9(2)	O(14)-Cu(1)-N(5)	175.4(3)
O(2)-Dy(1)-O(8)	138.6(2)	O(14)-Cu(1)-N(6)	87.2(3)
O(2)-Dy(1)-O(6)	122.2(2)	O(12)-Cu(1)-O(13)	167.4(2)
O(4)-Dy(1)-O(10)	140.5(3)	O(11)-Cu(1)-O(14)	87.9(3)
O(4)-Dy(1)-O(8)	75.4(2)	O(11)-Cu(1)-O(13)	84.9(3)
O(9)-Dy(1)-O(10)	69.3(3)	O(11)-Cu(1)-O(12)	84.7(3)
O(9)-Dy(1)-O(2)	92.1(2)	O(11)-Cu(1)-N(5)	88.5(3)
O(9)-Dy(1)-O(4)	95.7(2)	O(11)-Cu(1)-N(6)	174.8(3)
O(9)-Dy(1)-O(8)	70.9(3)	N(5)-Cu(1)-O(13)	91.8(3)
O(9)-Dy(1)-O(6)	140.6(3)	N(5)-Cu(1)-O(12)	94.9(3)
O(8)-Dy(1)-O(10)	127.7(3)	N(6)-Cu(1)-O(13)	93.0(3)
O(6)-Dy(1)-O(10)	133.5(2)	N(6)-Cu(1)-O(12)	96.8(3)
O(6)-Dy(1)-O(4)	81.3(2)	N(6)-Cu(1)-N(5)	96.3(3)
O(6)-Dy(1)-O(8)	70.4(2)	N(2)-O(2)-Dy(1)	131.4(5)
O(7)-Dy(1)-O(5)	78.4(3)	N(4)-O(4)-Dy(1)	134.2(6)
O(7)-Dy(1)-O(10)	72.4(3)		

Table S3. Selected bond lengths [Å] and angles [°] for **3**.

Bond distances			
Gd(1)-O(18)	2.392(5)	Cu(2)-N(8)#1	2.001(6)
Gd(1)-O(10)	2.384(5)	Cu(1)-O(4)	1.947(5)
Gd(1)-O(17)	2.411(5)	Cu(1)-O(2)	1.950(5)
Gd(1)-O(12)	2.372(5)	Cu(1)-N(3)	2.145(6)
Gd(1)-O(13)	2.404(6)	Cu(1)-O(3)	2.266(6)
Gd(1)-O(15)	2.400(5)	Cu(1)-O(1)	2.021(6)
Gd(1)-O(14)	2.375(5)	Cu(1)-N(7)#2	2.407(6)
Gd(1)-O(16)	2.377(5)	N(2)-O(10)	1.306(8)
Cu(2)-O(6)	2.018(6)	O(12)-N(6)	1.312(8)
Cu(2)-O(5)	2.279(6)	N(5)-O(11)	1.286(8)
Cu(2)-O(8)	2.029(5)	O(9)-N(1)	1.276(8)
Cu(2)-O(7)	2.273(6)	N(8)-Cu(2)#3	2.001(6)
Cu(2)-N(4)	2.006(6)	N(7)-Cu(1)#4	2.407(6)
Angles			
O(18)-Gd(1)-O(17)	73.71(18)	O(6)-Cu(2)-O(7)	92.5(2)

O(18)-Gd(1)-O(13)	128.78(18)	O(8)-Cu(2)-O(5)	93.0(2)
O(18)-Gd(1)-O(15)	129.28(19)	O(8)-Cu(2)-O(7)	87.1(2)
O(10)-Gd(1)-O(18)	74.46(17)	O(7)-Cu(2)-O(5)	179.7(2)
O(10)-Gd(1)-O(17)	72.74(18)	N(4)-Cu(2)-O(6)	90.7(2)
O(10)-Gd(1)-O(13)	148.87(19)	N(4)-Cu(2)-O(5)	89.8(2)
O(10)-Gd(1)-O(15)	72.17(18)	N(4)-Cu(2)-O(8)	89.9(2)
O(12)-Gd(1)-O(18)	71.82(18)	N(4)-Cu(2)-O(7)	90.5(2)
O(12)-Gd(1)-O(10)	138.85(19)	N(8)#1-Cu(2)-O(6)	89.9(2)
O(12)-Gd(1)-O(17)	75.69(18)	N(8)#1-Cu(2)-O(5)	89.6(2)
O(12)-Gd(1)-O(13)	72.27(19)	N(8)#1-Cu(2)-O(8)	89.6(2)
O(12)-Gd(1)-O(15)	148.89(19)	N(8)#1-Cu(2)-O(7)	90.1(2)
O(12)-Gd(1)-O(14)	95.76(19)	N(8)#1-Cu(2)-N(4)	179.2(3)
O(12)-Gd(1)-O(16)	99.37(18)	O(4)-Cu(1)-O(2)	177.7(2)
O(13)-Gd(1)-O(17)	129.11(19)	O(4)-Cu(1)-N(3)	92.5(2)
O(15)-Gd(1)-O(17)	128.28(18)	O(4)-Cu(1)-O(3)	86.6(2)
O(15)-Gd(1)-O(13)	76.72(19)	O(4)-Cu(1)-O(1)	90.4(2)
O(14)-Gd(1)-O(18)	75.87(18)	O(4)-Cu(1)-N(7)#2	82.7(2)
O(14)-Gd(1)-O(10)	98.22(19)	O(2)-Cu(1)-N(3)	86.6(2)
O(14)-Gd(1)-O(17)	149.56(19)	O(2)-Cu(1)-O(3)	95.6(2)
O(14)-Gd(1)-O(13)	72.7(2)	O(2)-Cu(1)-O(1)	90.2(2)
O(14)-Gd(1)-O(15)	72.42(19)	O(2)-Cu(1)-N(7)#2	95.1(2)
O(14)-Gd(1)-O(16)	134.59(19)	N(3)-Cu(1)-O(3)	96.4(2)
O(16)-Gd(1)-O(18)	149.53(18)	N(3)-Cu(1)-N(7)#2	82.1(2)
O(16)-Gd(1)-O(10)	97.81(18)	O(3)-Cu(1)-N(7)#2	169.1(2)
O(16)-Gd(1)-O(17)	75.85(18)	O(1)-Cu(1)-N(3)	171.0(3)
O(16)-Gd(1)-O(13)	71.69(19)	O(1)-Cu(1)-O(3)	92.3(3)
O(16)-Gd(1)-O(15)	72.67(19)	O(1)-Cu(1)-N(7)#2	89.7(3)
O(6)-Cu(2)-O(5)	87.4(2)	N(2)-O(10)-Gd(1)	138.3(4)
O(6)-Cu(2)-O(8)	179.3(2)	N(6)-O(12)-Gd(1)	137.3(5)

#1 x-1, -y+1/2, z-1/2 #2 x-1, y, z

#3 x+1, -y+1/2, z+1/2 #4 x+1, y, z

Table S4. Selected bond lengths [Å] and angles [°] for **4**.

Bond distances			
Tb(1)-O(18)	2.367(6)	Cu(2)-N(8)#1	1.993(7)
Tb(1)-O(10)	2.366(6)	Cu(1)-O(4)	1.938(6)
Tb(1)-O(17)	2.382(6)	Cu(1)-O(2)	1.944(6)
Tb(1)-O(12)	2.348(6)	Cu(1)-N(3)	2.132(7)
Tb(1)-O(13)	2.380(6)	Cu(1)-O(3)	2.252(7)
Tb(1)-O(15)	2.365(6)	Cu(1)-O(1)	2.009(7)
Tb(1)-O(14)	2.355(6)	Cu(1)-N(7)#2	2.393(8)
Tb(1)-O(16)	2.353(6)	N(2)-O(10)	1.301(8)
Cu(2)-O(6)	1.996(6)	O(12)-N(6)	1.306(8)
Cu(2)-O(5)	2.272(7)	N(5)-O(11)	1.289(10)

Cu(2)-O(8)	2.023(6)	O(9)-N(1)	1.281(9)
Cu(2)-O(7)	2.258(7)	N(8)-Cu(2)#3	1.993(7)
Cu(2)-N(4)	1.996(7)	N(7)-Cu(1)#4	2.393(8)
Angles			
O(18)-Tb(1)-O(10)	74.6(2)	O(6)-Cu(2)-O(7)	92.3(3)
O(18)-Tb(1)-O(17)	73.4(2)	O(6)-Cu(2)-N(4)	91.0(3)
O(18)-Tb(1)-O(13)	128.8(2)	O(8)-Cu(2)-O(5)	92.9(3)
O(10)-Tb(1)-O(17)	72.4(2)	O(8)-Cu(2)-O(7)	87.2(3)
O(10)-Tb(1)-O(13)	148.6(2)	O(7)-Cu(2)-O(5)	179.8(3)
O(12)-Tb(1)-O(18)	71.2(2)	N(4)-Cu(2)-O(5)	90.2(3)
O(12)-Tb(1)-O(10)	138.2(2)	N(4)-Cu(2)-O(8)	89.9(3)
O(12)-Tb(1)-O(17)	75.4(2)	N(4)-Cu(2)-O(7)	90.0(3)
O(12)-Tb(1)-O(13)	73.1(2)	N(8)#1-Cu(2)-O(6)	89.8(3)
O(12)-Tb(1)-O(15)	148.9(2)	N(8)#1-Cu(2)-O(5)	88.8(3)
O(12)-Tb(1)-O(14)	96.1(2)	N(8)#1-Cu(2)-O(8)	89.4(3)
O(12)-Tb(1)-O(16)	99.4(2)	N(8)#1-Cu(2)-O(7)	91.0(3)
O(13)-Tb(1)-O(17)	129.7(2)	N(8)#1-Cu(2)-N(4)	178.7(3)
O(15)-Tb(1)-O(18)	129.9(2)	O(4)-Cu(1)-O(2)	177.8(3)
O(15)-Tb(1)-O(10)	72.8(2)	O(4)-Cu(1)-N(3)	92.1(3)
O(15)-Tb(1)-O(17)	128.6(2)	O(4)-Cu(1)-O(3)	86.7(3)
O(15)-Tb(1)-O(13)	75.9(2)	O(4)-Cu(1)-O(1)	90.8(3)
O(14)-Tb(1)-O(18)	76.0(2)	O(4)-Cu(1)-N(7)#2	82.3(3)
O(14)-Tb(1)-O(10)	98.2(2)	O(2)-Cu(1)-N(3)	87.1(3)
O(14)-Tb(1)-O(17)	149.4(2)	O(2)-Cu(1)-O(3)	95.4(3)
O(14)-Tb(1)-O(13)	72.6(2)	O(2)-Cu(1)-O(1)	89.7(3)
O(14)-Tb(1)-O(15)	72.2(2)	O(2)-Cu(1)-N(7)#2	95.5(3)
O(16)-Tb(1)-O(18)	149.3(2)	N(3)-Cu(1)-O(3)	96.4(3)
O(16)-Tb(1)-O(10)	97.9(2)	N(3)-Cu(1)-N(7)#2	82.1(3)
O(16)-Tb(1)-O(17)	75.9(2)	O(3)-Cu(1)-N(7)#2	168.9(3)
O(16)-Tb(1)-O(13)	71.7(2)	O(1)-Cu(1)-N(3)	171.4(3)
O(16)-Tb(1)-O(15)	72.7(2)	O(1)-Cu(1)-O(3)	91.9(3)
O(16)-Tb(1)-O(14)	134.6(2)	O(1)-Cu(1)-N(7)#2	90.2(3)
O(6)-Cu(2)-O(5)	87.6(3)	N(2)-O(10)-Tb(1)	137.2(5)
O(6)-Cu(2)-O(8)	179.0(3)	N(6)-O(12)-Tb(1)	136.6(5)

#1 x-1, -y+1/2, z-1/2 #2 x-1, y, z

#3 x+1, -y+1/2, z+1/2 #4 x+1, y, z

Table S5. Selected bond lengths [Å] and angles [°] for **5**.

Bond distances			
Dy(1)-O(18)	2.356(5)	Cu(2)-N(8)#1	1.984(6)
Dy(1)-O(10)	2.356(5)	Cu(1)-O(4)	1.939(5)
Dy(1)-O(17)	2.386(5)	Cu(1)-O(2)	1.936(5)
Dy(1)-O(12)	2.346(5)	Cu(1)-N(3)	2.132(6)
Dy(1)-O(13)	2.373(6)	Cu(1)-O(3)	2.258(6)

Dy(1)-O(15)	2.358(5)	Cu(1)-O(1)	2.017(6)
Dy(1)-O(14)	2.341(5)	Cu(1)-N(7)#2	2.409(6)
Dy(1)-O(16)	2.340(5)	N(2)-O(10)	1.286(8)
Cu(2)-O(6)	2.005(5)	O(12)-N(6)	1.304(8)
Cu(2)-O(5)	2.270(6)	N(5)-O(11)	1.279(9)
Cu(2)-O(8)	2.015(6)	O(9)-N(1)	1.281(8)
Cu(2)-O(7)	2.262(7)	N(8)-Cu(2)#3	1.984(6)
Cu(2)-N(4)	1.995(6)	N(7)-Cu(1)#4	2.409(6)
Angles			
O(18)-Dy(1)-O(10)	74.89(17)	O(6)-Cu(2)-O(7)	92.3(2)
O(18)-Dy(1)-O(17)	74.16(18)	O(8)-Cu(2)-O(5)	93.1(2)
O(18)-Dy(1)-O(13)	128.82(18)	O(8)-Cu(2)-O(7)	87.1(2)
O(18)-Dy(1)-O(15)	129.72(19)	O(7)-Cu(2)-O(5)	179.8(2)
O(10)-Dy(1)-O(17)	73.01(18)	N(4)-Cu(2)-O(6)	90.7(2)
O(10)-Dy(1)-O(13)	148.03(19)	N(4)-Cu(2)-O(5)	90.4(2)
O(10)-Dy(1)-O(15)	72.26(19)	N(4)-Cu(2)-O(8)	89.8(2)
O(12)-Dy(1)-O(18)	71.64(18)	N(4)-Cu(2)-O(7)	89.8(2)
O(12)-Dy(1)-O(10)	138.95(19)	N(8)#1-Cu(2)-O(6)	89.8(2)
O(12)-Dy(1)-O(17)	75.52(18)	N(8)#1-Cu(2)-O(5)	88.9(2)
O(12)-Dy(1)-O(13)	73.01(19)	N(8)#1-Cu(2)-O(8)	89.7(2)
O(12)-Dy(1)-O(15)	148.72(19)	N(8)#1-Cu(2)-O(7)	91.0(3)
O(13)-Dy(1)-O(17)	129.38(19)	N(8)#1-Cu(2)-N(4)	179.1(3)
O(15)-Dy(1)-O(17)	128.18(17)	O(4)-Cu(1)-N(3)	92.7(2)
O(15)-Dy(1)-O(13)	75.8(2)	O(4)-Cu(1)-O(3)	86.7(2)
O(14)-Dy(1)-O(18)	75.34(18)	O(4)-Cu(1)-O(1)	90.4(2)
O(14)-Dy(1)-O(10)	97.63(19)	O(4)-Cu(1)-N(7)#2	82.5(2)
O(14)-Dy(1)-O(17)	149.47(18)	O(2)-Cu(1)-O(4)	178.1(2)
O(14)-Dy(1)-O(12)	96.23(18)	O(2)-Cu(1)-N(3)	86.2(2)
O(14)-Dy(1)-O(13)	73.0(2)	O(2)-Cu(1)-O(3)	95.1(2)
O(14)-Dy(1)-O(15)	72.56(18)	O(2)-Cu(1)-O(1)	90.4(2)
O(16)-Dy(1)-O(18)	149.06(18)	O(2)-Cu(1)-N(7)#2	95.7(2)
O(16)-Dy(1)-O(10)	98.11(18)	N(3)-Cu(1)-O(3)	97.1(2)
O(16)-Dy(1)-O(17)	74.98(18)	N(3)-Cu(1)-N(7)#2	81.8(2)
O(16)-Dy(1)-O(12)	98.50(18)	O(3)-Cu(1)-N(7)#2	169.0(2)
O(16)-Dy(1)-O(13)	71.72(19)	O(1)-Cu(1)-N(3)	170.5(2)
O(16)-Dy(1)-O(15)	73.23(19)	O(1)-Cu(1)-O(3)	92.1(2)
O(16)-Dy(1)-O(14)	135.55(19)	O(1)-Cu(1)-N(7)#2	89.7(2)
O(6)-Cu(2)-O(5)	87.5(2)	N(2)-O(10)-Dy(1)	138.9(4)
O(6)-Cu(2)-O(8)	179.2(2)	N(6)-O(12)-Dy(1)	136.8(4)

#1 x-1, -y+1/2, z-1/2 #2 x-1, y, z

#3 x+1, -y+1/2, z+1/2 #4 x+1, y, z

Table S6. SHAPE analysis for the Ln coordination spheres for **1** and **2**.

Complex	**BTPR-8**	JBTPR-8	TDD-8	SAPR-8
1 Gd	**0.945**	1.648	2.144	2.678
2 Dy	**1.003**	1.586	2.072	2.623

BTPR-8: Biaugmented trigonal prism; JBTPR-8: Biaugmented trigonal prism J50; TDD-8: Triangular dodecahedron; SAPR-8: Square antiprism.

Table S7. SHAPE analysis for the Cu coordination spheres for **1** and **2**.

Complex	**OC-6**	TPR-6	PPY-6
1 Gd	**0.861**	15.503	28.278
2 Dy	**0.850**	15.559	28.354

OC-6: Octahedron; TPR-6: Trigonal prism; PPY-6: Pentagonal pyramid.

Table S8. SHAPE analysis for the Ln coordination spheres for **3–5**.

Complex	**TDD-8**	BTPR-8	SAPR-8	JSD-8
3 Gd	**0.073**	2.747	2.802	2.943
4 Tb	**0.068**	2.754	2.838	2.951
5 Dy	**0.052**	2.727	2.908	2.904

TDD-8: Triangular dodecahedron; BTPR-8: Biaugmented trigonal prism; SAPR-8: Square antiprism; JSD-8: Snub diphenoid J84.

Table S9. SHAPE analysis for the Cu coordination spheres for **3–5**.

Complex		**OC-6**	TPR-6	PPY-6
3 Gd	**Cu1**	**1.027**	14.395	26.029
	Cu2	**0.388**	16.455	29.209
4 Tb	**Cu1**	**1.024**	14.291	26.018
	Cu2	**0.396**	16.275	29.027
5 Dy	**Cu1**	**1.093**	14.182	25.652
	Cu2	**0.412**	16.301	29.021

OC-6: Octahedron; TPR-6: Trigonal prism; PPY-6: Pentagonal pyramid.

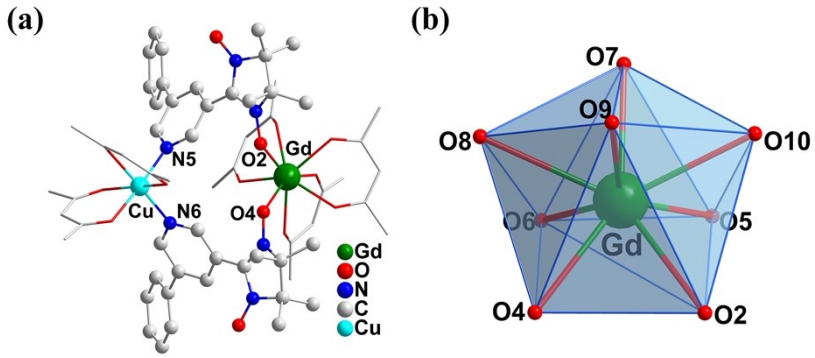


Fig. S1 (a) The dinuclear structure of complex **1**. H and F atoms are not shown for the sake of clarity. (b) The coordination polyhedron of Gd^{III} ion.

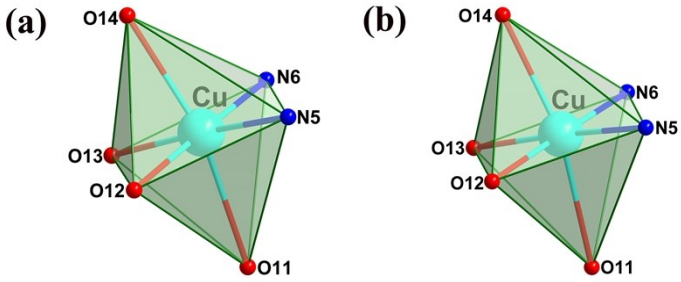


Fig. S2 Coordination polyhedrons of the Cu ions in complex **1** (a) and complex **2** (b).

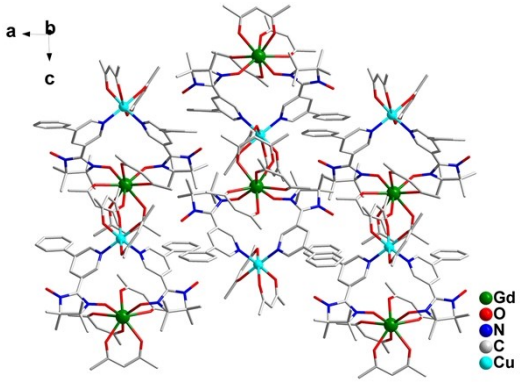


Fig. S3 Packing diagram of **1** (H and F atoms are omitted for clarity).

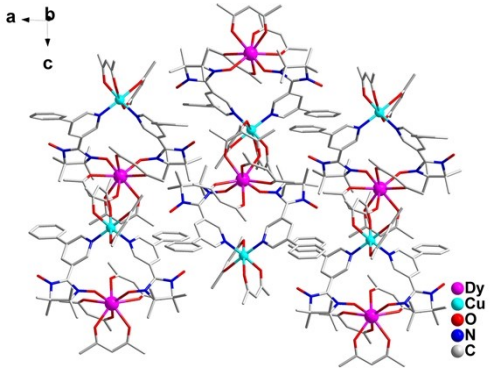


Fig. S4 Packing diagram of **2** (H and F atoms are omitted for clarity).

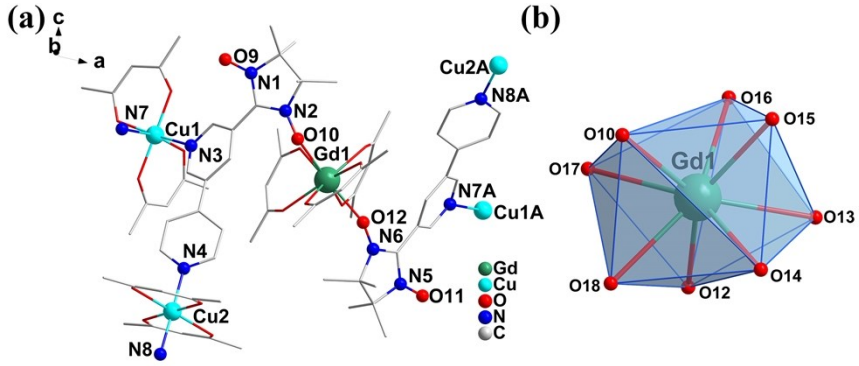


Fig. S5 (a) The asymmetric unit found in complex **3**. H and F atoms are not shown for the sake of clarity. (b) The coordination polyhedron of Gd^{III} ion.

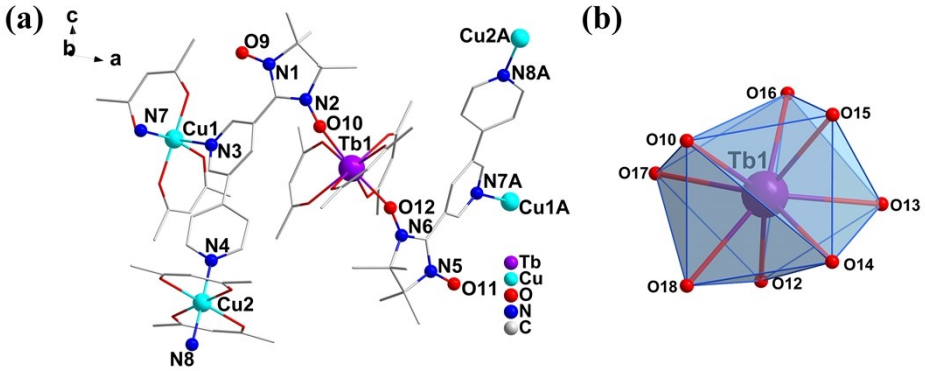


Fig. S6 (a) The asymmetric unit found in complex **4**. H and F atoms are not shown for the sake of clarity. (b) The coordination polyhedron of Tb^{III} ion.

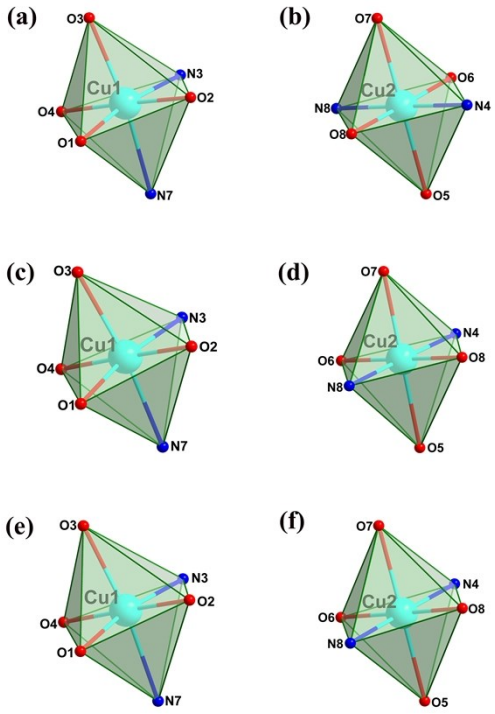


Fig. S7 Coordination polyhedrons of Cu1 ion (a) and Cu2 ion (b) in complex **1**, Cu1 ion (c) and Cu2 ion (d) in complex **2** and Cu1 ion (e) and Cu2 ion (f) in complex **3**.

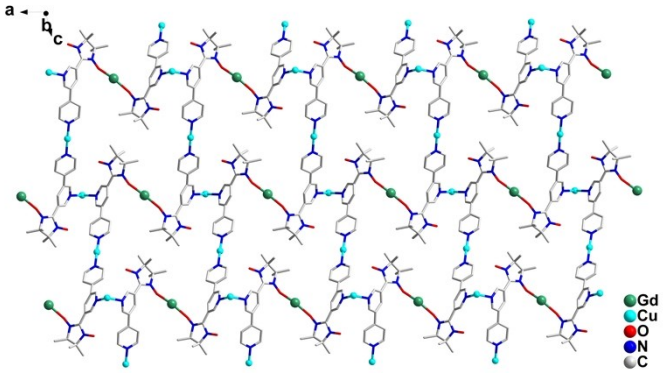


Fig. S8 2D structure of complex **3** (H atoms and hfac ligands are not shown for the sake of clarity).

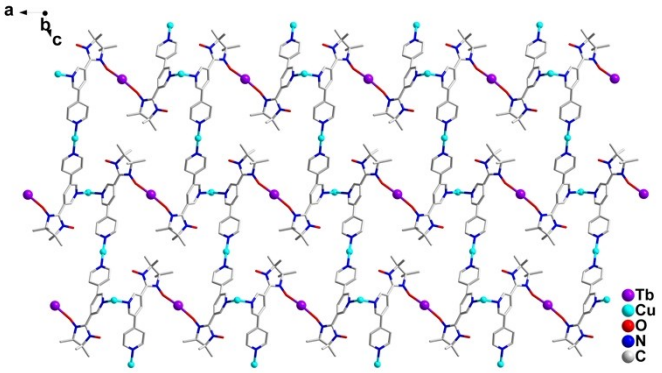


Fig. S9 2D structure of complex **4** (H atoms and hfac ligands are not shown for the sake of clarity).

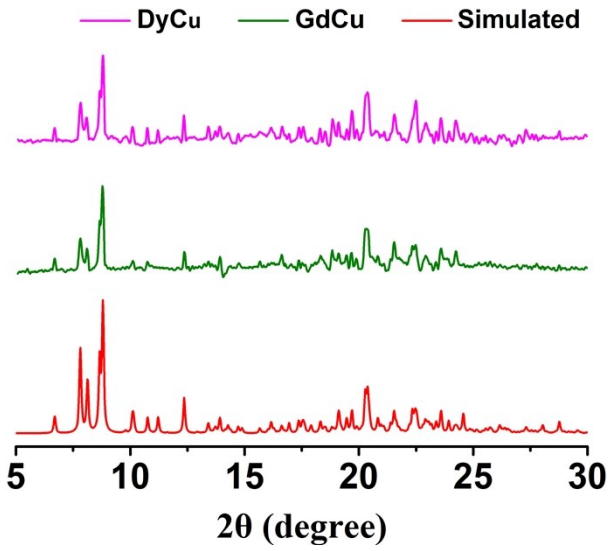


Fig. S10 Powder X-ray diffraction patterns of complexes **1** and **2**.

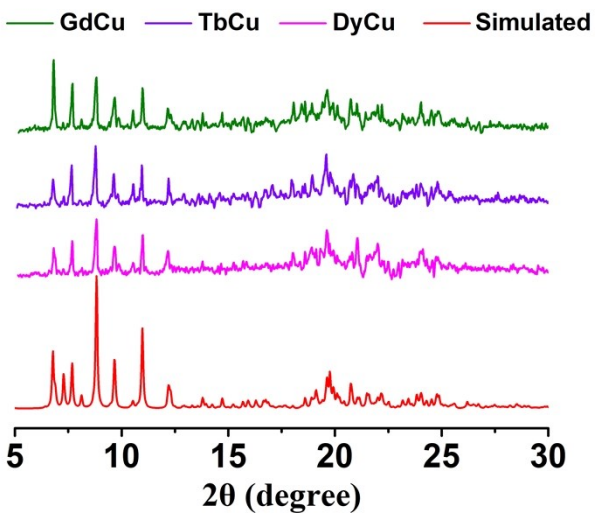


Fig. S11 Powder X-ray diffraction patterns of complexes **3–5**.

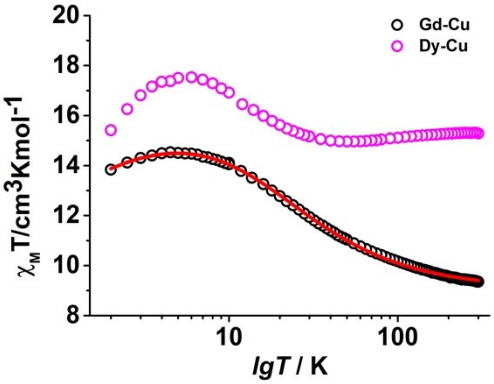


Fig. S12 Plot of $\chi_M T$ versus lg$T$ for **1** and **2**. The red line is the simulation curve by *PHI* for **1**.

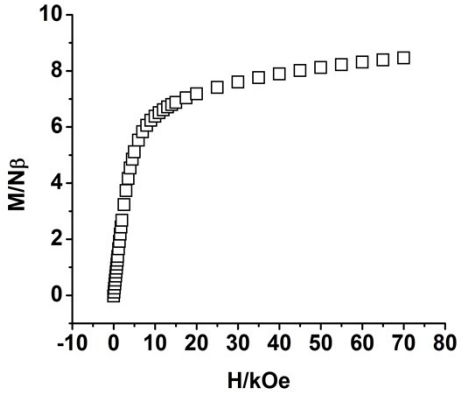
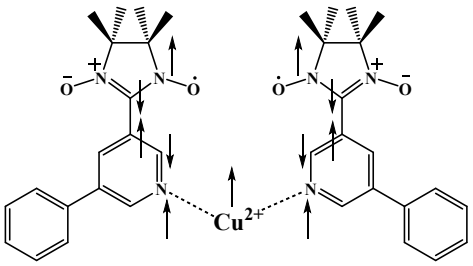


Fig. S13 Plot of magnetization *vs* field for **2** at 2 K.



Scheme S1. Spin polarization mechanism for the magnetic coupling mediated by the *m*-pyridyl ring.

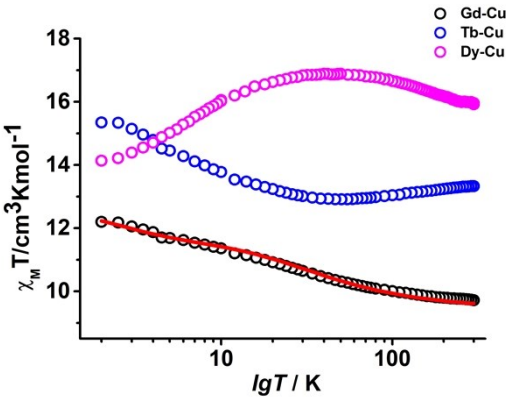


Fig. S14 Plot of $\chi_M T$ versus lg$T$ for **3**–**5**. The red line is the simulation curve by *PHI* for **3**.

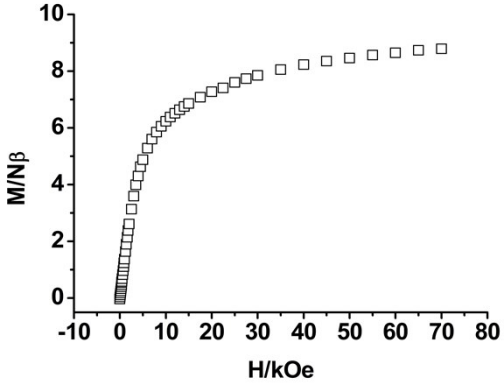


Fig. S15 Plot of magnetization *vs* field for **4** at 2 K.

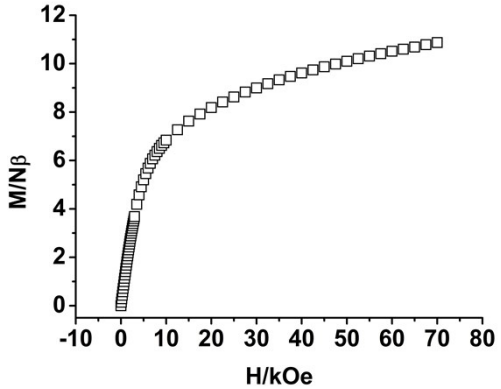


Fig. S16 Plot of magnetization *vs* field for **5** at 2 K.

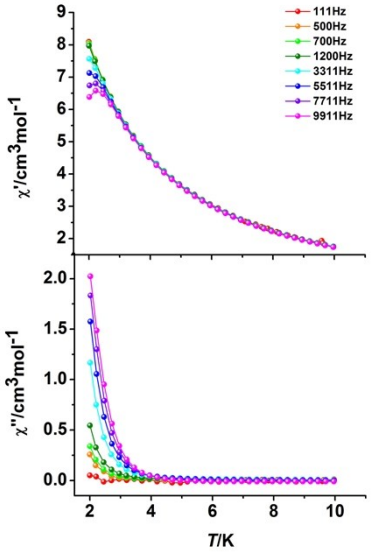


Fig. S17 Temperature-dependent ac signals of the χ′ (top) and χ″ (bottom) under zero dc field for compound **2**.

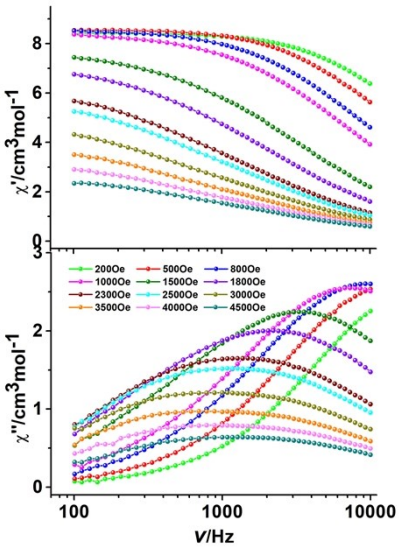


Fig. S18 Frequency dependence of the χ′ (top) and χ″ (bottom) components of the ac susceptibility for **2** at 2 K under different dc fields.

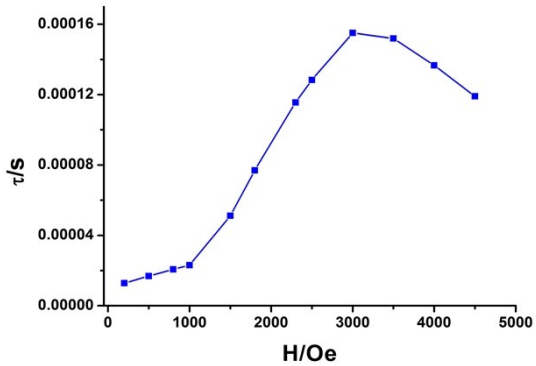


Fig. S19 The τ versus H plot for complex **2** at 2.0 K under the applied dc field.

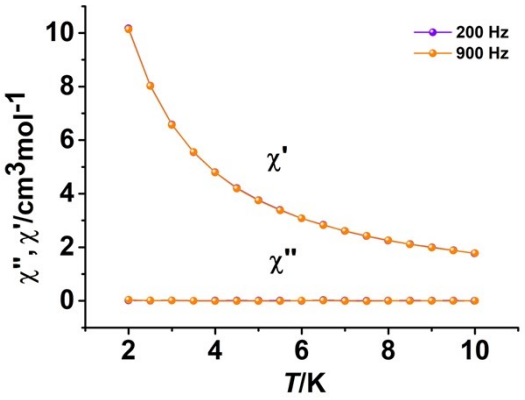


Fig. S20 Temperature-dependent ac signals of the χ′ (top) and χ″ (bottom) under zero dc field for compound **4**.

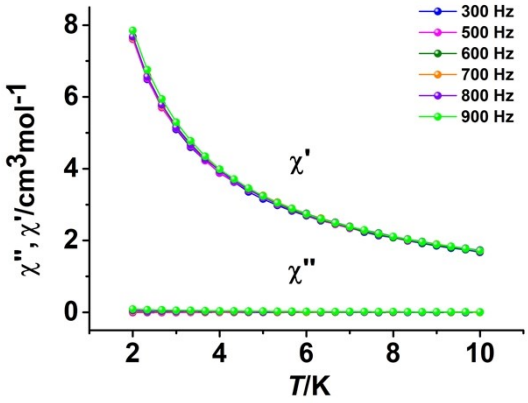


Fig. S21 Temperature-dependent ac signals of the χ′ (top) and χ″ (bottom) under zero dc field for compound **5**.

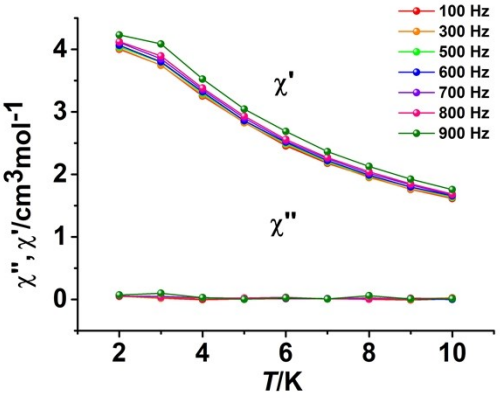


Fig. S22 Temperature-dependent ac signals of the χ′ (top) and χ″ (bottom) under 3000 Oe dc field for compound **5**.