Electronic Supporting Information

Structures and magnetic properties of novel Ln(III)-based pentanuclear clusters: magnetic refrigeration and single-molecule magnet behavior

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Table S1 The important bond lengths and angles for cluster **1**.

Bond lengths			
Sm(4)-O(5)	2.316(4)	Sm(3)-O(26)	2.369(4)
Sm(4)-O(3)	2.377(4)	Sm(3)-O(13)	2.350(4)
Sm(4)-O(24)	2.489(4)	Sm(3)-N(7)	2.750(5)
Sm(4)-O(15)	2.452(4)	Sm(3)-N(1)	2.742(5)
Sm(4)-O(23)	2.348(4)	Sm(2)-O(11)	2.390(4)
Sm(4)-N(10)	2.660(6)	Sm(2)-O(1)	2.344(4)
Sm(4)-O(9)	2.348(4)	Sm(2)-O(13)	2.325(4)
Sm(4)-N(3)	2.649(5)	Sm(2)-O(21)	2.493(4)
Sm(3)-O(25)	2.363(4)	Sm(2)-O(7)	2.481(4)
Sm(3)-O(5)	2.347(4)	Sm(2)-O(22)	2.343(4)
Sm(3)-O(3)	2.422(4)	Sm(2)-N(4)	2.641(5)
Sm(3)-O(11)	2.412(4)	Sm(2)-N(9)	2.649(6)
Sm(1)-O(1)	2.292(5)	Sm(1)-O(21)	2.617(4)
Sm(1)-O(17)	2.352(5)	Sm(1)-O(7)	2.458(4)
Sm(1)-O(20)	2.389(5)	Sm(1)-O(18)	2.362(5)
Sm(1)-N(6)	2.558(6)	Sm(1)-O(19)	2.392(6)
Sm(5)-O(29)	2.376(5)	Sm(5)-O(24)	2.605(5)
Sm(5)-O(15)	2.461(4)	Sm(5)-O(27)	2.367(5)
Sm(5)-O(28)	2.335(5)	Sm(5)-O(30)	2.412(5)
Sm(5)-O(9)	2.306(4)	Sm(5)-N(12)	2.552(6)
Bond angles			
O(5)-Sm(4)-O(3)	68.22(14)	O(5)-Sm(4)-O(15)	91.05(14)
O(5)-Sm(4)-O(24)	154.18(15)	O(5)-Sm(4)-O(23)	129.39(15)
O(5)-Sm(4)-O(9)	90.22(15)	O(3)-Sm(4)-O(24)	134.78(14)
O(3)-Sm(4)-O(24)	134.78(14)	O(3)-Sm(4)-O(15)	157.03(14)

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O(15)-Sm(4)-O(24)	63.90(14)	O(23)-Sm(4)-O(3)	82.88(15)
O(23)-Sm(4)-O(24)	72.54(15)	O(23)-Sm(4)-O(15)	119.15(15)
O(23)-Sm(4)-O(9)	137.03(15)	O(9)-Sm(4)-O(3)	101.08(15)
O(9)-Sm(4)-O(24)	75.14(15)	O(9)-Sm(4)-O(15)	67.99(15)
O(25)-Sm(3)-O(3)	133.18(14)	O(25)-Sm(3)-O(11)	76.11(14)
O(25)-Sm(3)-O(26)	72.00(16)	O(5)-Sm(3)-O(25)	152.73(15)
O(5)-Sm(3)-O(3)	66.98(13)	O(5)-Sm(3)-O(11)	90.99(14)
O(5)-Sm(3)-O(26)	103.01(15)	O(5)-Sm(3)-O(13)	90.96(14)
O(11)-Sm(3)-O(3)	148.21(14)	O(26)-Sm(3)-O(3)	74.89(15)
O(26)-Sm(3)-O(11)	134.64(15)	O(13)-Sm(3)-O(25)	105.18(16)
O(13)-Sm(3)-O(3)	90.24(14)	O(13)-Sm(3)-O(11)	66.52(14)
O(13)-Sm(3)-O(26)	153.23(15)	O(11)-Sm(2)-O(21)	133.59(14)
O(11)-Sm(2)-O(7)	159.46(14)	O(1)-Sm(2)-O(11)	102.50(15)
O(1)-Sm(2)-O(21)	73.87(14)	O(1)-Sm(2)-O(7)	68.68(15)
O(13)-Sm(2)-O(11)	67.27(14)	O(13)-Sm(2)-O(1)	89.18(15)
O(13)-Sm(2)-O(21)	154.98(15)	O(13)-Sm(2)-O(7)	93.51(14)
O(13)-Sm(2)-O(22)	130.32(16)	O(7)-Sm(2)-O(21)	63.46(14)
O(22)-Sm(2)-O(11)	84.01(15)	O(22)-Sm(2)-O(1)	137.80(16)
O(22)-Sm(2)-O(21)	72.08(15)	O(22)-Sm(2)-O(7)	115.23(15)
O(1)-Sm(1)-O(21)	72.33(14)	O(1)-Sm(1)-O(17)	87.75(19)
O(1)-Sm(1)-O(7)	69.89(15)	O(1)-Sm(1)-O(20)	79.84(17)
O(1)-Sm(1)-O(18)	106.56(18)	O(1)-Sm(1)-O(19)	152.02(18)
O(17)-Sm(1)-O(21)	142.89(17)	O(17)-Sm(1)-O(7)	139.77(17)
O(17)-Sm(1)-O(20)	76.31(18)	O(17)-Sm(1)-O(18)	72.03(18)
O(17)-Sm(1)-O(19)	81.4(2)	O(7)-Sm(1)-O(21)	61.95(14)
O(20)-Sm(1)-O(21)	69.67(16)	O(20)-Sm(1)-O(7)	128.45(16)
O(20)-Sm(1)-O(19)	72.56(19)	O(18)-Sm(1)-O(21)	143.03(15)
O(18)-Sm(1)-O(7)	82.59(16)	O(18)-Sm(1)-O(20)	147.31(17)
O(18)-Sm(1)-O(19)	94.57(19)	O(19)-Sm(1)-O(21)	101.55(17)
O(19)-Sm(1)-O(7)	132.36(19)	O(29)-Sm(5)-O(24)	69.42(15)
O(29)-Sm(5)-O(15)	129.52(16)	O(29)-Sm(5)-O(30)	72.93(18)
O(15)-Sm(5)-O(24)	62.07(13)	O(27)-Sm(5)-O(29)	145.94(18)
O(27)-Sm(5)-O(24)	139.90(17)	O(27)-Sm(5)-O(15)	78.11(18)
O(27)-Sm(5)-O(30)	108.1(2)	O(28)-Sm(5)-O(29)	73.7(2)
O(28)-Sm(5)-O(24)	141.0(2)	O(28)-Sm(5)-O(15)	145.32(18)
O(28)-Sm(5)-O(27)	72.9(2)	O(28)-Sm(5)-O(30)	80.5(2)
O(30)-Sm(5)-O(24)	100.13(16)	O(30)-Sm(5)-O(15)	127.38(16)
O(9)-Sm(5)-O(29)	85.78(16)	O(9)-Sm(5)-O(24)	73.60(14)
O(9)-Sm(5)-O(27)	88.24(18)	O(9)-Sm(5)-O(28)	91.7(2)
O(9)-Sm(5)-O(30)	158.60(17)		

Table S2 The important bond lengths and angles for cluster **2**.

Bond lengths			
Eu(4)-O(5)	2.311(4)	Eu(4)-O(25)	2.340(5)
Eu(4)-O(9)	2.339(4)	Eu(4)-O(26)	2.466(4)
Eu(4)-O(15)	2.444(5)	Eu(4)-N(3)	2.634(6)
Eu(4)-N(10)	2.651(6)	Eu(4)-O(3)	2.363(4)
Eu(3)-O(5)	2.337(4)	Eu(3)-O(11)	2.408(4)
Eu(3)-O(21)	2.346(4)	Eu(3)-O(13)	2.349(4)
Eu(3)-O(22)	2.362(4)	Eu(3)-N(1)	2.741(6)
Eu(3)-N(7)	2.734(6)	Eu(3)-O(3)	2.409(4)
Eu(2)-O(7)	2.471(5)	Eu(2)-O(11)	2.379(4)
Eu(2)-O(13)	2.311(4)	Eu(2)-O(24)	2.334(5)
Eu(2)-O(1)	2.321(5)	Eu(2)-O(23)	2.483(4)
Eu(2)-N(4)	2.632(5)	Eu(2)-N(9)	2.628(6)
Eu(5)-O(9)	2.294(4)	Eu(5)-O(27) Eu(5)-	2.365(5)
Eu(5)-O(26) Eu(5)-	2.594(5)	O(15)	2.449(4)
O(30)	2.321(5)	Eu(5)-O(28)	2.406(5)
Eu(5)-O(29)	2.349(5)	Eu(5)-N(12)	2.542(6)
Eu(1)-O(17)	2.339(5)	Eu(1)-O(20) Eu(1)-	2.379(5)
Eu(1)-O(7) Eu(1)-	2.442(4)	O(1)	2.289(5)
O(18)	2.350(5)	Eu(1)-O(23)	2.595(5)
Eu(1)-N(6)	2.548(6)	Eu(1)-N(6)	2.548(6)
Eu(1)-O(19)	2.383(6)		
Bond angles			
O(5)-Eu(4)-O(25)	128.57(16)	O(5)-Eu(4)-O(9)	90.72(15)
O(5)-Eu(4)-O(26)	154.10(16)	O(5)-Eu(4)-O(15)	90.88(15)
O(5)-Eu(4)-O(3)	68.16(15)	O(25)-Eu(4)-O(26)	73.21(16)
O(25)-Eu(4)-O(15)	119.75(16)	O(25)-Eu(4)-O(3)	81.82(16)
O(9)-Eu(4)-O(25)	137.23(16)	O(9)-Eu(4)-O(26)	75.04(15)
O(9)-Eu(4)-O(15)	68.05(15)	O(9)-Eu(4)-O(3)	102.00(16)
O(15)-Eu(4)-O(26)	63.87(15)	O(3)-Eu(4)-O(26)	135.34(15)
O(3)-Eu(4)-O(15)	157.18(14)	O(3)-Eu(4)-N(3)	62.24(16)
O(5)-Eu(3)-O(11)	91.17(14)	O(5)-Eu(3)-O(13)	92.55(15)
O(5)-Eu(3)-O(22)	102.51(15)	O(5)-Eu(3)-O(3)	66.97(14)
O(11)-Eu(3)-O(3)	148.16(14)	O(21)-Eu(3)-O(13)	104.09(16)
O(21)-Eu(3)-O(22)	72.12(16)	O(21)-Eu(3)-O(3)	133.63(15)
O(13)-Eu(3)-O(11)	66.55(14)	O(13)-Eu(3)-O(22)	152.96(16)
O(13)-Eu(3)-O(3)	90.61(15)	O(22)-Eu(3)-O(11)	134.27(15)
O(22)-Eu(3)-O(3)	75.26(15)	O(7)-Eu(2)-O(23)	63.30(15)
O(11)-Eu(2)-O(7)	159.38(14)	O(11)-Eu(2)-O(23)	134.36(14)
O(13)-Eu(2)-O(7)	92.81(15)	O(13)-Eu(2)-O(11)	67.63(14)
O(13)-Eu(2)-O(24)	130.21(16)	O(13)-Eu(2)-O(1)	89.36(16)
O(13)-Eu(2)-O(23)	154.35(16)	O(24)-Eu(2)-O(7)	115.88(15)
O(24)-Eu(2)-O(11)	83.15(16)	O(24)-Eu(2)-O(23)	72.51(16)

O(1)-Eu(2)-O(7)	68.83(15)	O(1)-Eu(2)-O(11)	103.13(16)
O(1)-Eu(2)-O(24)	137.63(16)	O(1)-Eu(2)-O(23)	73.86(15)
O(9)-Eu(5)-O(27)	85.18(17)	O(9)-Eu(5)-O(26)	73.32(15)
O(9)-Eu(5)-O(15)	68.67(15)	O(9)-Eu(5)-O(30)	91.9(2)
O(9)-Eu(5)-O(28)	157.29(18)	O(9)-Eu(5)-O(29)	87.69(18)
O(27)-Eu(5)-O(26)	69.28(16)	O(27)-Eu(5)-O(15)	129.21(17)
O(27)-Eu(5)-O(28)	72.25(18)	O(15)-Eu(5)-O(26)	61.93(14)
O(30)-Eu(5)-O(27)	74.4(2)	O(30)-Eu(5)-O(26)	141.6(2)
O(30)-Eu(5)-O(15)	145.07(17)	O(28)-Eu(5)-O(15)	128.21(16)
O(28)-Eu(5)-O(26)	100.13(16)	O(30)-Eu(5)-O(28)	79.9(2)
O(29)-Eu(5)-O(27)	146.23(18)	O(29)-Eu(5)-O(26)	139.05(17)
O(29)-Eu(5)-O(15)	77.50(18)	O(29)-Eu(5)-O(28)	109.60(19)
O(17)-Eu(1)-O(20)	76.34(18)	O(17)-Eu(1)-O(7)	139.42(18)
O(17)-Eu(1)-O(18)	72.51(18)	O(17)-Eu(1)-O(23)	142.86(18)
O(17)-Eu(1)-O(19)	81.3(2)	O(20)-Eu(1)-O(7)	128.41(17)
O(20)-Eu(1)-O(23)	69.65(16)	O(20)-Eu(1)-O(19)	73.12(18)
O(7)-Eu(1)-O(23) O(1)-	62.04(14)	O(1)-Eu(1)-O(20) O(1)-	79.54(17)
Eu(1)-O(7)	69.83(15)	Eu(1)-O(17)	87.6(2)
O(1)-Eu(1)-O(18)	106.40(18)	O(1)-Eu(1)-O(23) O(18)-	72.22(15)
O(1)-Eu(1)-O(19)	152.20(17)	Eu(1)-O(20)	147.90(18) 142.43(15)
O(18)-Eu(1)-O(7)	81.90(16)	O(18)-Eu(1)-O(23)	132.64(18)
O(18)-Eu(1)-O(19)	94.46(19)	O(19)-Eu(1)-O(7)	
O(19)-Eu(1)-O(23)	102.15(17)		

Table S3 The important bond lengths and angles for cluster **3**.

Bond lengths			
Gd(2)-O(16)	2.326(7)	Gd(2)-O(8)	2.301(6)
Gd(2)-O(10)	2.357(6)	Gd(2)-O(2)	2.433(7)
Gd(2)-O(22)	2.319(7)	Gd(2)-O(21)	2.450(7)
Gd(2)-N(7)	2.619(8)	Gd(2)-N(3)	2.638(9)
Gd(4)-O(14)	2.375(7)	Gd(4)-O(4)	2.308(6)
Gd(4)-O(12)	2.306(7)	Gd(4)-O(6)	2.457(7)
Gd(4)-O(30)	2.472(6)	Gd(4)-O(29)	2.322(7)
Gd(4)-N(6)	2.635(8)	Gd(4)-N(10)	2.624(9)
Gd(3)-O(14)	2.396(6)	Gd(3)-O(24)	2.342(7)
Gd(3)-O(8)	2.333(6)	Gd(3)-O(10)	2.398(6)
Gd(3)-O(4)	2.332(7)	Gd(3)-O(23)	2.347(7)
Gd(3)-N(9)	2.722(8)	Gd(3)-N(12)	2.737(9)
Gd(5)-O(25)	2.371(7)	Gd(5)-O(12)	2.279(7)
Gd(5)-O(6)	2.432(6)	Gd(5)-O(30)	2.580(7)
Gd(5)-O(28)	2.310(7)	Gd(5)-O(26)	2.388(8)
Gd(5)-O(27)	2.337(7)	Gd(5)-N(4)	2.539(9)
Gd(1)-O(16)	2.293(7)	Gd(1)-O(20)	2.378(7)
Gd(1)-O(2)	2.440(7)	Gd(1)-O(18)	2.347(7)

Gd(1)-O(21)	2.582(7)	Gd(1)-O(17)	2.303(8)
Gd(1)-O(19)	2.394(7)	Gd(1)-N(1)	2.536(9)
Bond angles			
O(16)-Gd(2)-O(10)	102.3(2)	O(16)-Gd(2)-O(2)	68.5(2)
O(16)-Gd(2)-O(21)	75.3(2)	O(8)-Gd(2)-O(16)	91.2(2)
O(8)-Gd(2)-O(10)	68.2(2)	O(8)-Gd(2)-O(2)	90.7(2)
O(8)-Gd(2)-O(22)	128.1(2)	O(8)-Gd(2)-O(21)	154.0(2)
O(10)-Gd(2)-O(2)	157.3(2)	O(10)-Gd(2)-O(21)	135.8(2)
O(2)-Gd(2)-O(21)	63.9(2)	O(14)-Gd(4)-O(6)	159.3(2)
O(22)-Gd(2)-O(16)	137.1(2)	O(14)-Gd(4)-O(30)	134.8(2)
O(22)-Gd(2)-O(10)	81.2(2)	O(4)-Gd(4)-O(14)	67.2(2)
O(22)-Gd(2)-O(2)	120.0(2)	O(4)-Gd(4)-O(6)	92.9(2)
O(22)-Gd(2)-O(21)	73.4(2)	O(4)-Gd(4)-O(30)	154.7(2)
O(4)-Gd(4)-O(29)	129.5(3)	O(12)-Gd(4)-O(14)	103.8(2)
O(12)-Gd(4)-O(4)	90.0(2)	O(12)-Gd(4)-O(6)	69.0(2)
O(12)-Gd(4)-O(30)	73.8(2)	O(12)-Gd(4)-O(29)	137.5(2)
O(6)-Gd(4)-O(30)	63.4(2)	O(29)-Gd(4)-O(14)	82.6(2)
O(29)-Gd(4)-O(6)	116.2(2)	O(29)-Gd(4)-O(30)	72.7(2)
O(14)-Gd(3)-O(10)	148.4(2)	O(24)-Gd(3)-O(14)	75.3(2)
O(24)-Gd(3)-O(10)	133.9(2)	O(24)-Gd(3)-O(23)	72.6(3)
O(8)-Gd(3)-O(14)	91.4(2)	O(8)-Gd(3)-O(23)	102.0(2)
O(8)-Gd(3)-O(24)	151.6(2)	O(4)-Gd(3)-O(14)	66.5(2)
O(8)-Gd(3)-O(10)	67.0(2)	O(4)-Gd(3)-O(24)	103.4(2)
O(4)-Gd(3)-O(8)	93.6(2)	O(4)-Gd(3)-O(10)	91.2(2)
O(4)-Gd(3)-O(23)	152.6(2)	O(23)-Gd(3)-O(14)	134.4(2)
O(23)-Gd(3)-O(10)	74.8(2)	O(25)-Gd(5)-O(30)	69.9(2)
O(25)-Gd(5)-O(6)	128.7(2)	O(25)-Gd(5)-O(26)	73.2(3)
O(12)-Gd(5)-O(25)	79.4(3)	O(12)-Gd(5)-O(30)	72.2(2)
O(12)-Gd(5)-O(6)	69.9(2)	O(12)-Gd(5)-O(28)	87.2(3)
O(12)-Gd(5)-O(26)	152.1(3)	O(12)-Gd(5)-O(27)	106.6(3)
O(6)-Gd(5)-O(30)	62.2(2)	O(28)-Gd(5)-O(6)	139.1(3)
O(28)-Gd(5)-O(25)	76.0(3)	O(28)-Gd(5)-O(30)	142.6(3)
O(28)-Gd(5)-O(26)	81.5(3)	O(26)-Gd(5)-O(6)	132.8(3)
O(28)-Gd(5)-O(27)	73.0(3)	O(26)-Gd(5)-O(30)	102.2(2)
O(27)-Gd(5)-O(25)	148.0(3)	O(27)-Gd(5)-O(30)	142.1(2)
O(27)-Gd(5)-O(6)	81.5(2)	O(27)-Gd(5)-O(26)	94.5(3)
O(16)-Gd(1)-O(20)	84.2(3)	O(16)-Gd(1)-O(21)	73.3(2)
O(16)-Gd(1)-O(2)	68.9(2)	O(16)-Gd(1)-O(17)	92.5(3)
O(16)-Gd(1)-O(18)	87.6(3)	O(16)-Gd(1)-O(19)	156.7(2)
O(20)-Gd(1)-O(2)	129.2(2)	O(20)-Gd(1)-O(21)	69.6(2)
O(20)-Gd(1)-O(19)	72.7(3)	O(2)-Gd(1)-O(21)	61.8(2)
O(18)-Gd(1)-O(20)	146.1(3)	O(18)-Gd(1)-O(2)	76.8(3)
O(18)-Gd(1)-O(21)	138.3(3)	O(17)-Gd(1)-O(21)	142.2(3)
O(18)-Gd(1)-O(19)	110.2(3)	O(17)-Gd(1)-O(19)	79.1(3)

O(17)-Gd(1)-O(20)	74.3(3)	O(19)-Gd(1)-O(2)	128.4(2)
O(17)-Gd(1)-O(2)	145.4(3)	O(19)-Gd(1)-O(21)	100.2(2)
O(17)-Gd(1)-O(18)	73.3(3)		

Table S4 The important bond lengths and angles for cluster **4**.

Bond lengths			
Tb(4)-O(11)	2.426(5)	Tb(4)-O(13)	2.290(4)
Tb(4)-O(25)	2.318(5)	Tb(4)-O(25)	2.318(5)
Tb(4)-O(26)	2.446(4)	Tb(4)-N(3)	2.598(5)
Tb(4)-N(7)	2.631(6)	Tb(4)-O(3)	2.345(4)
Tb(4)-O(5)	2.314(5)	Tb(2)-O(22)	2.311(5)
Tb(2)-O(9)	2.283(4)	Tb(2)-O(7)	2.360(4)
Tb(2)-O(1)	2.301(4)	Tb(2)-O(21)	2.451(4)
Tb(2)-O(15)	2.435(5)	Tb(2)-N(10)	2.626(5)
Tb(2)-N(6)	2.591(6)	Tb(3)-O(9)	2.318(4)
Tb(3)-O(13)	2.309(4)	Tb(3)-O(7)	2.390(4)
Tb(3)-O(23)	2.316(4)	Tb(3)-N(1)	2.730(6)
Tb(3)-O(24)	2.333(5)	Tb(3)-N(4)	2.723(6)
Tb(3)-O(3)	2.381(4)	Tb(1)-O(18)	2.351(5)
Tb(1)-O(1)	2.260(5)	Tb(1)-O(20)	2.319(5)
Tb(1)-O(21)	2.562(5)	Tb(1)-O(15)	2.411(4)
Tb(1)-O(19)	2.324(5)	Tb(1)-O(17)	2.359(5)
Tb(1)-N(12)	2.531(6)	Tb(5)-O(11)	2.416(4)
Tb(5)-O(27)	2.330(5)	Tb(5)-O(29)	2.340(5)
Tb(5)-O(28)	2.297(5)	Tb(5)-O(26)	2.573(5)
Tb(5)-O(26)	2.573(5)	Tb(5)-N(9)	2.515(6)
Tb(5)-O(30)	2.375(5)	Tb(5)-O(5)	2.275(4)
Bond angles			
O(11)-Tb(4)-O(26)	63.82(14)	O(13)-Tb(4)-O(11)	90.56(15)
O(13)-Tb(4)-O(25)	127.90(16)	O(13)-Tb(4)-O(26)	153.82(16)
O(13)-Tb(4)-O(3)	67.75(15)	O(13)-Tb(4)-O(5)	90.75(15)
O(25)-Tb(4)-O(11)	119.71(16)	O(25)-Tb(4)-O(26)	73.66(16)
O(25)-Tb(4)-O(3)	81.60(16)	O(5)-Tb(4)-O(11)	68.13(16)
O(5)-Tb(4)-O(25)	138.07(15)	O(5)-Tb(4)-O(26)	75.31(15)
O(5)-Tb(4)-O(3)	102.89(16)	O(22)-Tb(2)-O(7)	82.41(16)
O(22)-Tb(2)-O(21)	73.32(16)	O(22)-Tb(2)-O(15)	116.52(16)
O(9)-Tb(2)-O(22)	129.47(16)	O(9)-Tb(2)-O(7)	67.20(15)
O(9)-Tb(2)-O(1)	89.39(15)	O(9)-Tb(2)-O(21)	154.25(16)
O(9)-Tb(2)-O(15)	92.50(15)	O(7)-Tb(2)-O(21)	135.09(14)
O(7)-Tb(2)-O(15)	158.94(14)	O(1)-Tb(2)-O(22)	138.31(15)
O(1)-Tb(2)-O(7)	104.09(16)	O(1)-Tb(2)-O(21)	73.74(15)
O(1)-Tb(2)-O(15)	68.64(15)	O(15)-Tb(2)-O(21)	63.50(14)
O(9)-Tb(3)-O(7)	66.17(15)	O(9)-Tb(3)-O(24)	153.03(16)
O(9)-Tb(3)-O(3)	91.59(15)	O(13)-Tb(3)-O(9)	94.26(16)

O(13)-Tb(3)-O(7)	91.61(14)	O(13)-Tb(3)-O(23)	151.83(16)
O(13)-Tb(3)-O(24)	101.41(16)	O(13)-Tb(3)-O(3)	66.82(14)
O(23)-Tb(3)-O(9)	102.47(16)	O(23)-Tb(3)-O(7)	75.10(15)
O(23)-Tb(3)-O(24)	73.06(16)	O(23)-Tb(3)-O(3)	134.05(15)
O(24)-Tb(3)-O(7)	134.23(15)	O(24)-Tb(3)-O(3)	74.97(15)
O(3)-Tb(3)-O(7)	148.36(14)	O(18)-Tb(1)-O(21)	69.85(16)
O(18)-Tb(1)-O(15)	128.62(17)	O(18)-Tb(1)-O(17)	73.34(18)
O(1)-Tb(1)-O(18)	79.47(16)	O(1)-Tb(1)-O(20)	87.24(19)
O(1)-Tb(1)-O(21)	72.26(15)	O(1)-Tb(1)-O(15)	69.72(15)
O(1)-Tb(1)-O(19)	106.54(18)	O(1)-Tb(1)-O(17)	129.21(17)
O(20)-Tb(1)-O(18)	75.70(18)	O(20)-Tb(1)-O(21)	142.38(17)
O(20)-Tb(1)-O(15)	139.34(16)	O(20)-Tb(1)-O(19)	73.11(18)
O(20)-Tb(1)-O(17)	80.6(2)	O(15)-Tb(1)-O(21)	62.18(14)
O(19)-Tb(1)-O(18)	147.81(18)	O(19)-Tb(1)-O(21)	142.33(15)
O(19)-Tb(1)-O(15)	81.74(16)	O(19)-Tb(1)-O(17)	93.77(19) 133.41(17)
O(17)-Tb(1)-O(21)	103.00(17)	O(17)-Tb(1)-O(15)	76.00(17)
O(11)-Tb(5)-O(26)	62.06(14)	O(27)-Tb(5)-O(11)	137.69(17)
O(27)-Tb(5)-O(29)	146.70(19)	O(27)-Tb(5)-O(26)	129.13(17)
O(27)-Tb(5)-O(30)	110.71(19)	O(29)-Tb(5)-O(11)	72.71(18)
O(29)-Tb(5)-O(26)	69.38(16)	O(29)-Tb(5)-O(30)	73.5(2)
O(28)-Tb(5)-O(11)	145.00(17)	O(28)-Tb(5)-O(27)	142.60(19)
O(28)-Tb(5)-O(29)	74.9(2)	O(28)-Tb(5)-O(26)	128.76(16)
O(28)-Tb(5)-O(30)	78.85(19)	O(30)-Tb(5)-O(11) O(5)-	68.91(15) 84.10(16)
O(30)-Tb(5)-O(26)	100.18(16)	Tb(5)-O(11)	73.48(15)
O(5)-Tb(5)-O(27)	87.10(18)	O(5)-Tb(5)-O(29)	
O(5)-Tb(5)-O(28)	92.6(2)	O(5)-Tb(5)-O(26)	
O(5)-Tb(5)-O(30)	156.63(17)		

Table S5 The important bond lengths and angles for cluster **5**.

Bond lengths			
Dy(4)-O(9)	2.267(6)	Dy(4)-O(25)	2.284(7)
Dy(4)-O(5)	2.299(7)	Dy(4)-O(26)	2.456(7)
Dy(4)-O(3)	2.349(7)	Dy(4)-O(15)	2.421(7)
Dy(2)-O(13)	2.275(6)	Dy(2)-O(1)	2.310(7)
Dy(2)-O(22)	2.297(7)	Dy(2)-O(22)	2.297(7)
Dy(2)-O(21)	2.427(6)	Dy(2)-O(7)	2.335(7)
Dy(2)-O(11)	2.406(7)	Dy(3)-O(13)	2.308(6)
Dy(3)-O(9)	2.330(7)	Dy(3)-O(24)	2.296(7)
Dy(3)-O(23)	2.321(6)	Dy(3)-O(3)	2.373(6)
Dy(3)-O(7)	2.366(6)	Dy(1)-O(1)	2.252(7)
Dy(4)-N(10)	2.614(8)	Dy(4)-N(3)	2.581(8)
Dy(2)-N(6)	2.581(9)	Dy(2)-N(7)	2.632(8)
Dy(3)-N(4)	2.714(8)	Dy(3)-N(1)	2.721(8)
Dy(1)-O(18)	2.322(7)	Dy(1)-O(19)	2.364(7)

Dy(1)-O(17)	2.284(8)	Dy(1)-O(21)	2.572(7)
Dy(1)-O(20)	2.340(7)	Dy(1)-N(9)	2.501(9)
Dy(1)-O(11)	2.412(6)	Dy(5)-O(28)	2.351(7)
Dy(5)-O(27)	2.342(7)	Dy(5)-O(5)	2.244(7)
Dy(5)-O(30)	2.310(7)	Dy(5)-N(12)	2.512(9)
Dy(5)-O(29)	2.307(7)	Dy(5)-O(26)	2.553(7)
Dy(5)-O(15)	2.412(6)		

Bond angles

O(9)-Dy(4)-O(25)	129.9(2)	O(9)-Dy(4)-O(5)	89.4(2)
O(9)-Dy(4)-O(26)	153.7(2)	O(9)-Dy(4)-O(3)	67.5(2)
O(9)-Dy(4)-O(15)	91.4(2)	O(25)-Dy(4)-O(5)	138.0(2)
O(25)-Dy(4)-O(26)	73.1(2)	O(25)-Dy(4)-O(3)	82.2(2)
O(25)-Dy(4)-O(15)	116.5(2)	O(5)-Dy(4)-O(26)	73.7(2)
O(5)-Dy(4)-O(3)	105.0(2)	O(5)-Dy(4)-O(15)	69.0(2)
O(3)-Dy(4)-O(26)	135.7(2)	O(3)-Dy(4)-O(15)	158.5(2)
O(15)-Dy(4)-O(26)	64.0(2)	O(13)-Dy(2)-O(1)	91.1(2)
O(13)-Dy(2)-O(22)	128.0(2)	O(13)-Dy(2)-O(21)	153.7(2)
O(13)-Dy(2)-O(7)	68.0(2)	O(13)-Dy(2)-O(11)	90.0(2)
O(1)-Dy(2)-O(21)	75.0(2)	O(1)-Dy(2)-O(7)	103.4(2)
O(1)-Dy(2)-O(11)	68.4(2)	O(22)-Dy(2)-O(1)	137.5(2)
O(22)-Dy(2)-O(21)	73.7(2)	O(22)-Dy(2)-O(7)	81.0(3)
O(22)-Dy(2)-O(11)	120.2(2)	O(7)-Dy(2)-O(21)	136.5(2)
O(7)-Dy(2)-O(11)	156.7(2)	O(11)-Dy(2)-O(21)	64.2(2)
O(13)-Dy(3)-O(9)	95.7(2)	O(13)-Dy(3)-O(23)	100.8(2)
O(13)-Dy(3)-O(3)	91.5(2)	O(13)-Dy(3)-O(7)	66.9(2)
O(9)-Dy(3)-O(3)	66.1(2)	O(9)-Dy(3)-O(7)	92.1(2)
O(24)-Dy(3)-O(13)	151.5(2)	O(24)-Dy(3)-O(9)	101.4(2)
O(24)-Dy(3)-O(23)	73.4(2)	O(24)-Dy(3)-O(3)	75.2(2)
O(24)-Dy(3)-O(7)	134.1(2)	O(23)-Dy(3)-O(9)	152.7(2)
O(23)-Dy(3)-O(3)	134.3(2)	O(23)-Dy(3)-O(7)	74.8(2)
O(7)-Dy(3)-O(3)	148.2(2)	O(1)-Dy(1)-O(18)	87.9(3)
O(1)-Dy(1)-O(19)	155.8(3)	O(1)-Dy(1)-O(17)	92.0(3)
O(1)-Dy(1)-O(21)	73.2(2)	O(1)-Dy(1)-O(20)	83.2(2)
O(1)-Dy(1)-O(11)	69.2(2)	O(18)-Dy(1)-O(19)	110.2(3)
O(18)-Dy(1)-O(21)	138.0(3)	O(18)-Dy(1)-O(20)	146.5(3)
O(18)-Dy(1)-O(11)	76.4(3)	O(19)-Dy(1)-O(21)	100.7(2)
O(19)-Dy(1)-O(11)	129.4(2)	O(17)-Dy(1)-O(18)	73.5(3)
O(17)-Dy(1)-O(19)	78.6(3)	O(17)-Dy(1)-O(21)	142.1(3)
O(17)-Dy(1)-O(20)	74.5(3)	O(17)-Dy(1)-O(11)	145.0(3)
O(20)-Dy(1)-O(19)	72.8(3)	O(20)-Dy(1)-O(21)	69.3(2)
O(20)-Dy(1)-O(11)	128.9(2)	O(11)-Dy(1)-O(21)	62.0(2)
O(28)-Dy(5)-O(26)	103.1(2)	O(28)-Dy(5)-O(15)	133.8(3)
O(27)-Dy(5)-O(28)	73.6(3)	O(27)-Dy(5)-O(26)	69.6(2)
O(27)-Dy(5)-O(15)	128.6(2)	O(5)-Dy(5)-O(28)	151.9(3)

O(5)-Dy(5)-O(27)	79.0(2)	O(5)-Dy(5)-O(30)	86.0(3)
O(5)-Dy(5)-O(29)	106.8(3)	O(5)-Dy(5)-O(26)	72.7(2)
O(5)-Dy(5)-O(15)	70.0(2)	O(30)-Dy(5)-O(28)	81.1(3)
O(30)-Dy(5)-O(27)	75.7(3)	O(30)-Dy(5)-O(26)	142.0(2)
O(30)-Dy(5)-O(15)	138.5(2)	O(29)-Dy(5)-O(28)	93.4(3)
O(29)-Dy(5)-O(27)	148.1(2)	O(29)-Dy(5)-O(30)	73.5(3)
O(29)-Dy(5)-O(26)	142.3(2)	O(29)-Dy(5)-O(15)	81.4(2)
O(15)-Dy(5)-O(26)	62.6(2)		

Table S6 The important bond lengths and angles for cluster **6**.

Bond lengths			
Ho(2)-O(15)	2.328(7)	Ho(2)-O(7)	2.410(8)
Ho(2)-O(22)	2.293(7)	Ho(2)-O(21)	2.435(7)
Ho(2)-O(9)	2.275(7)	Ho(2)-N(4)	2.619(8)
Ho(2)-N(12)	2.553(9)	Ho(2)-O(1)	2.296(7)
Ho(3)-O(15)	2.357(7)	Ho(3)-O(5)	2.294(7)
Ho(3)-O(3)	2.366(7)	Ho(3)-O(24)	2.300(7)
Ho(3)-O(23)	2.303(7)	Ho(3)-O(9)	2.295(7)
Ho(3)-N(10)	2.700(9)	Ho(3)-N(1)	2.726(10)
Ho(4)-O(27)	2.292(8)	Ho(4)-O(3)	2.341(7)
Ho(4)-O(5)	2.271(7)	Ho(4)-O(28)	2.429(7)
Ho(4)-O(11)	2.394(8)	Ho(4)-N(3)	2.564(9)
Ho(4)-O(13)	2.294(8)	Ho(5)-O(29)	2.340(8)
Ho(4)-N(7)	2.586(8)	Ho(5)-O(25)	2.289(8)
Ho(5)-O(11)	2.386(7)	Ho(5)-O(28)	2.568(8)
Ho(5)-O(13)	2.239(8)	Ho(5)-O(30)	2.320(8)
Ho(5)-O(26)	2.282(9)	Ho(1)-O(18)	2.306(9)
Ho(5)-N(9)	2.532(10)	Ho(1)-O(7)	2.402(7)
Ho(1)-O(19)	2.349(8)	Ho(1)-O(21)	2.527(8)
Ho(1)-O(20)	2.324(9)	Ho(1)-N(6)	2.491(10)
Ho(1)-O(1)	2.234(7)	Ho(1)-O(17)	2.298(10)
Bond angles			
O(15)-Ho(2)-O(7)	156.5(2)	O(15)-Ho(2)-O(21)	138.3(2)
O(22)-Ho(2)-O(15)	81.8(3)	O(22)-Ho(2)-O(7)	118.9(3)
O(22)-Ho(2)-O(21)	74.5(3)	O(22)-Ho(2)-O(1)	138.2(3)
O(7)-Ho(2)-O(21)	63.1(2)	O(9)-Ho(2)-O(15)	67.1(2)
O(9)-Ho(2)-O(22)	128.8(3)	O(9)-Ho(2)-O(7)	90.2(2)
O(9)-Ho(2)-O(21)	152.4(3)	O(9)-Ho(2)-O(1)	90.2(3)
O(1)-Ho(2)-O(15)	104.8(3)	O(1)-Ho(2)-O(21)	74.0(3)
O(1)-Ho(2)-O(7)	68.1(3)	O(15)-Ho(3)-O(3)	148.7(2)
O(5)-Ho(3)-O(15)	92.3(2)	O(24)-Ho(3)-O(15)	135.1(3)
O(5)-Ho(3)-O(3)	66.1(2)	O(24)-Ho(3)-O(3)	73.7(3)
O(5)-Ho(3)-O(24)	100.4(3)	O(24)-Ho(3)-O(23)	73.6(3)
O(5)-Ho(3)-O(23)	152.6(3)	O(23)-Ho(3)-O(15)	75.6(3)

O(5)-Ho(3)-O(9)	96.9(2)	O(23)-Ho(3)-O(3)	133.4(3)
O(9)-Ho(3)-O(15)	66.3(2)	O(27)-Ho(4)-O(11)	120.0(3)
O(9)-Ho(3)-O(3)	92.8(2)	O(27)-Ho(4)-O(3)	79.4(3)
O(9)-Ho(3)-O(24)	151.2(3)	O(27)-Ho(4)-O(28)	73.7(3)
O(9)-Ho(3)-O(23)	100.3(3)	O(27)-Ho(4)-O(13)	137.2(3)
O(5)-Ho(4)-O(27)	128.0(3)	O(18)-Ho(1)-O(20)	76.0(3)
O(5)-Ho(4)-O(11)	91.7(2)	O(7)-Ho(1)-O(21)	61.9(3)
O(5)-Ho(4)-O(3)	66.9(2)	O(19)-Ho(1)-O(7)	132.3(3)
O(5)-Ho(4)-O(28)	155.0(3)	O(19)-Ho(1)-O(21)	104.2(3)
O(5)-Ho(4)-O(13)	91.0(3)	O(20)-Ho(1)-O(7)	129.4(3)
O(25)-Ho(5)-O(30)	81.8(3)	O(20)-Ho(1)-O(19)	73.0(3)
O(13)-Ho(5)-O(29)	80.5(3)	O(20)-Ho(1)-O(21)	70.1(3)
O(13)-Ho(5)-O(11)	69.0(3)	O(1)-Ho(1)-O(18)	85.9(3)
O(13)-Ho(5)-O(25)	86.6(3)	O(1)-Ho(1)-O(7)	69.2(2)
O(13)-Ho(5)-O(28)	72.3(3)	O(1)-Ho(1)-O(19)	155.1(3)
O(13)-Ho(5)-O(26)	104.6(3)	O(1)-Ho(1)-O(21)	73.2(3)
O(13)-Ho(5)-O(30)	153.6(3)	O(1)-Ho(1)-O(20)	83.1(3)
O(26)-Ho(5)-O(29)	147.4(3)	O(1)-Ho(1)-O(17)	103.7(3)
O(26)-Ho(5)-O(11)	81.4(3)	O(17)-Ho(1)-O(18)	72.5(3)
O(26)-Ho(5)-O(25)	73.4(3)	O(17)-Ho(1)-O(7)	82.1(3)
O(26)-Ho(5)-O(28)	142.5(3)	O(17)-Ho(1)-O(19)	92.9(4)
O(26)-Ho(5)-O(30)	94.7(3)	O(17)-Ho(1)-O(21)	142.8(3)
O(30)-Ho(5)-O(29)	73.7(3)	O(17)-Ho(1)-O(20)	147.1(3)
O(30)-Ho(5)-O(11)	132.7(3)	O(18)-Ho(1)-O(19)	81.6(4)
O(30)-Ho(5)-O(28)	102.9(3)	O(18)-Ho(1)-O(21)	141.8(3)
O(18)-Ho(1)-O(7)	139.0(3)		

Table S7 The Gd^{III} geometry analysis by SHAPE 2.0 for cluster **3**.

Cluster 3	D_{4d}**SAPR**	D_{2d}**TDD**	C_{2v}**JBTPR**	C_{2v}**BTPR**	D_{2d}**JSD**
$Gd1^{III}$	4.845	2.661	3.022	1.969	4.273
$Gd2^{III}$	5.291	2.813	3.243	2.542	5.800
$Gd4^{III}$	5.201	2.832	3.694	3.035	6.024
$Gd5^{III}$	2.378	1.564	3.105	2.406	3.472
	D_{2d}**JGBF**	D_{3h}**JETBPY**	C_{2v}**JBTPR**	D_{2d}**JSD**	T_d**TT**
$Gd3^{III}$	42.878	33.909	41.000	42.127	43.649

SAPR-8 = Square antiprism; **TDD-8 =** Triangular dodecahedron; **JBTPR-8 =** Biaugmented trigonal prism J50; **BTPR-8 =** Biaugmented trigonal prism; **JSD-8 =** Snub diphenoid J84; **JGBF-8 =** Johnson gyrobifastigium J26; **JETBPY-8 =** Johnson elongated triangular bipyramid J14; **TT-8 =** Triakis tetrahedron.

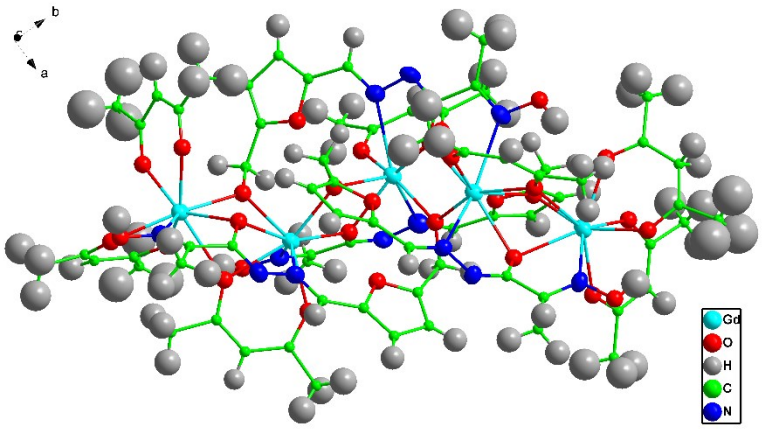

Fig. S1 The ellipsoidal model of cluster **3**.

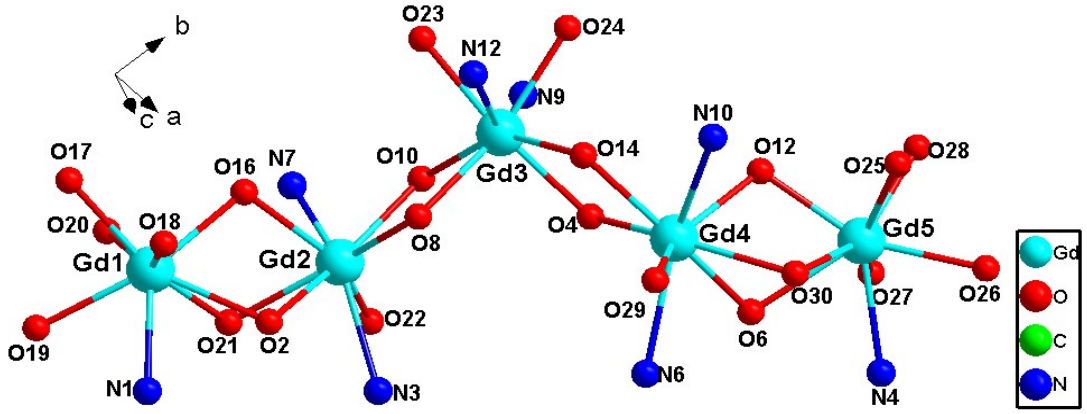

Fig. S2 The coordinate atom labels of central Gd(III) ions in cluster **3**.

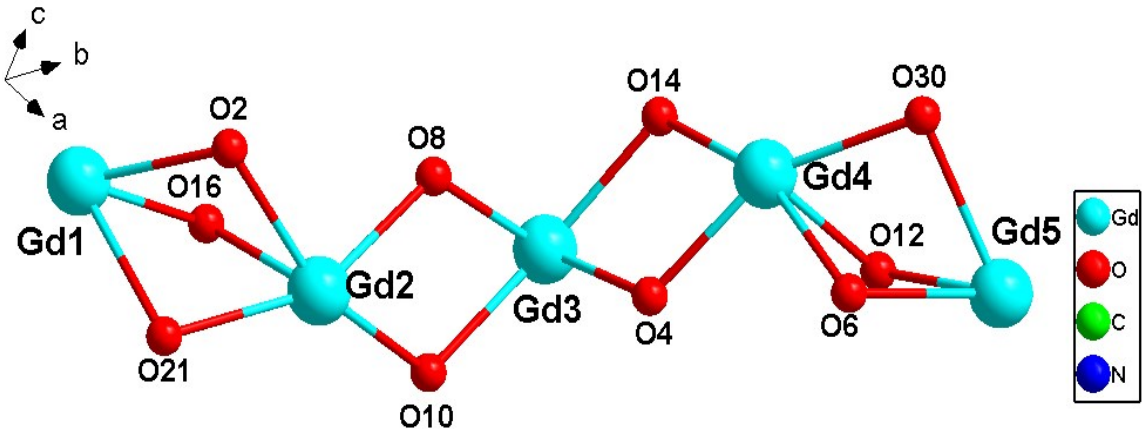

Fig. S3 The five central Gd atoms are bridged by ten μ_2-O atoms forming the Gd_5 core.

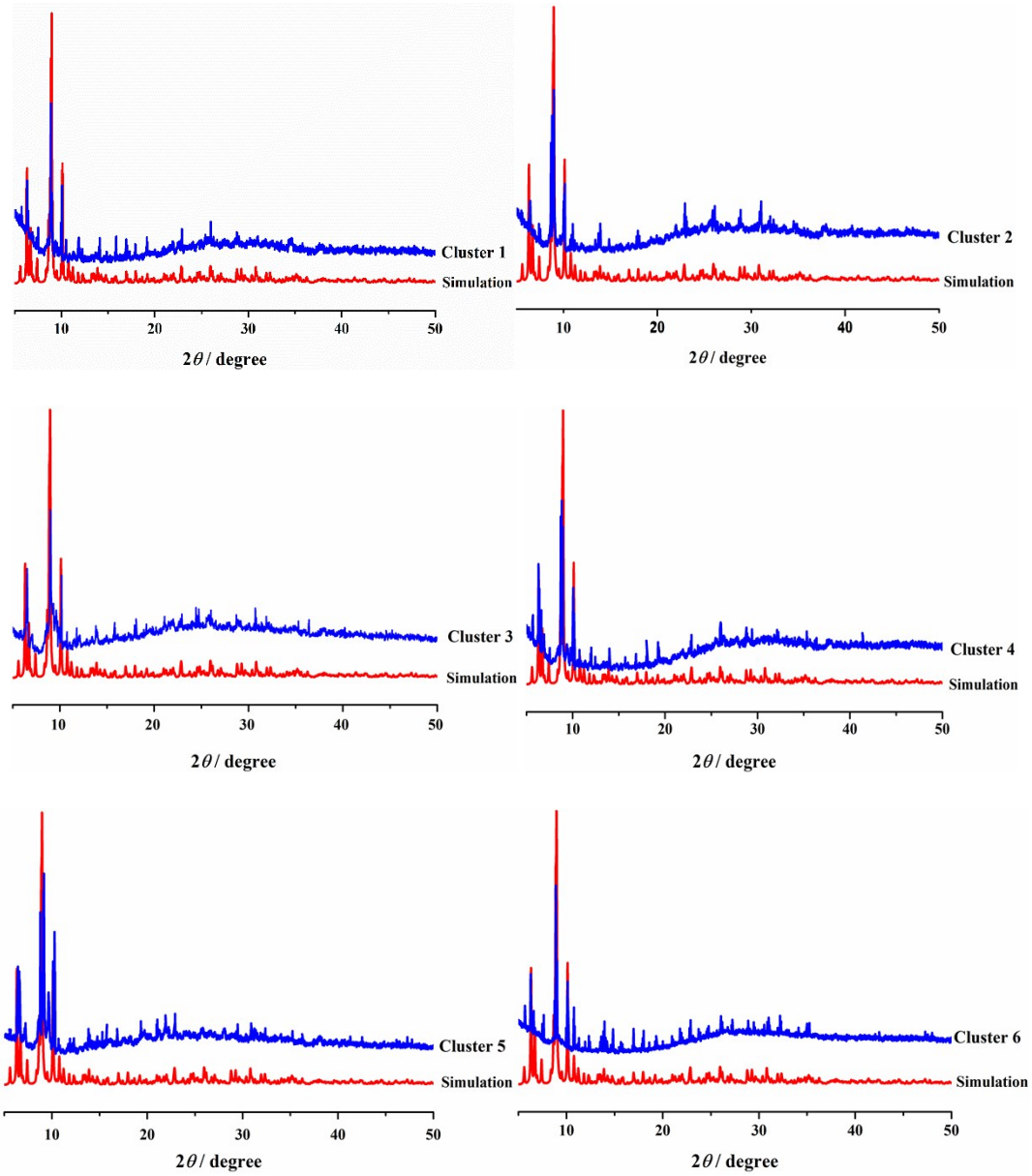

Fig. S4 PXRD patterns for clusters **1**-**6**.

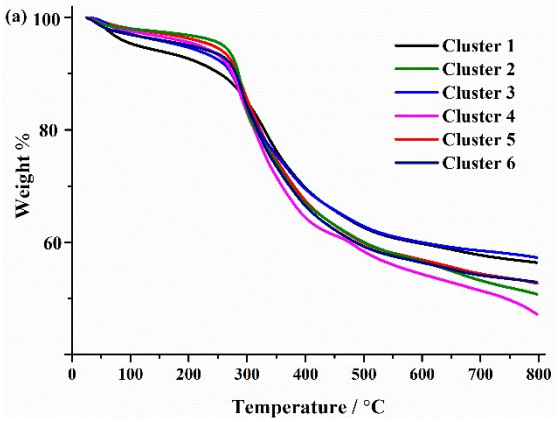

Fig. S5 The TGA curves of clusters **1**-**6**.

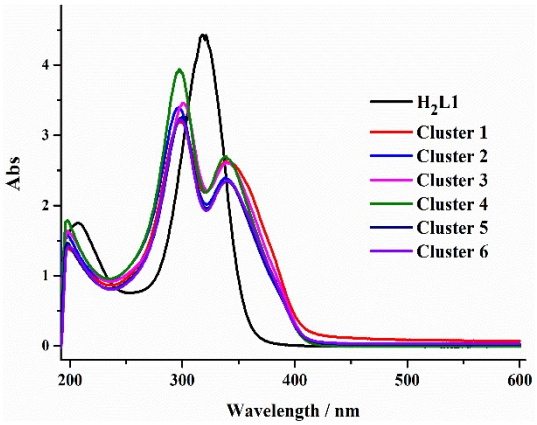

Fig. S6 The UV-vis spectra of ligand H_2L1 and clusters **1-6** at room temperature in ethanol solution.

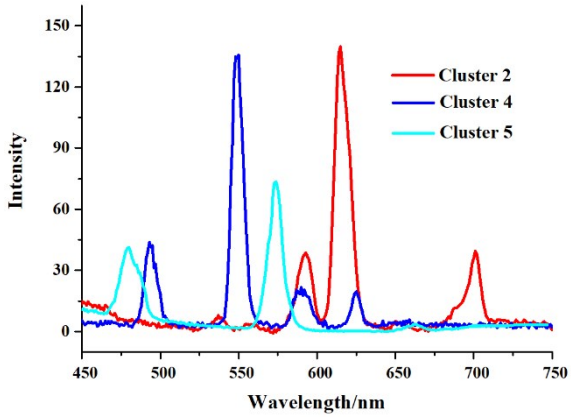

Fig. S7 The solid-state fluorescent spectra of **2**, **4** and **5** at room temperature.

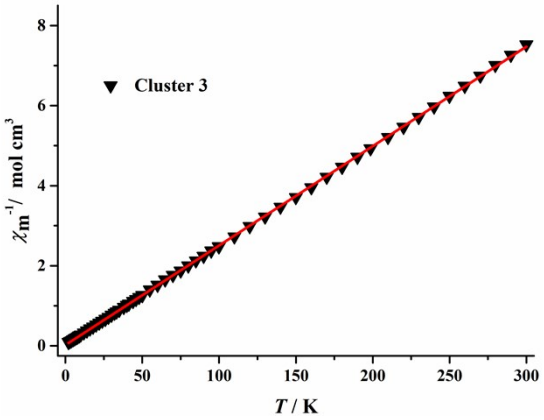

Fig. S8 Plots of χ_M^{-1} vs T for cluster **3**. The solid lines were generated from the best fit by the Curie-Weiss expression.

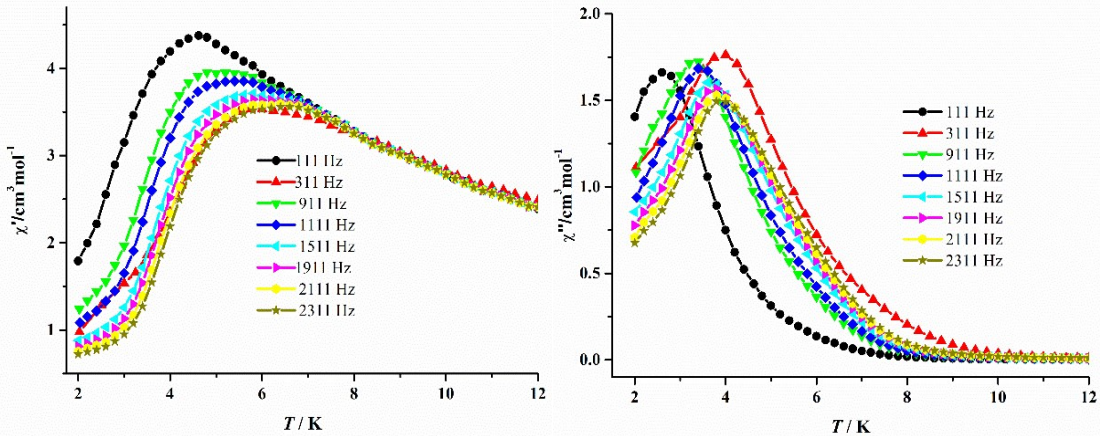

Fig. S9 Temperature dependence of the in-phase (left) and out-of-phase (right) components of the ac magnetic susceptibility for **5** in zero dc field with an oscillation of 3.0 Oe.

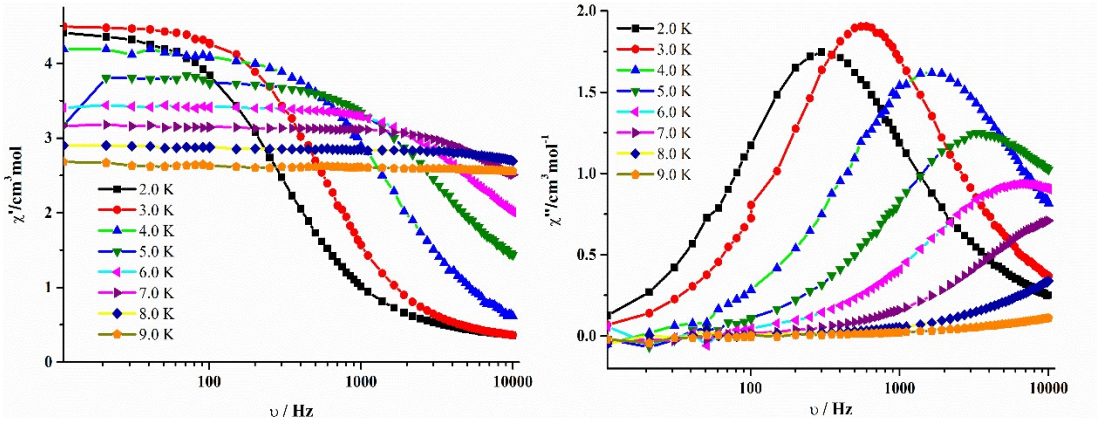

Fig. S10 Frequency dependence of χ' and χ'' for **5** at 2.0−9.0 K under a H_{dc} = 0 Oe field.

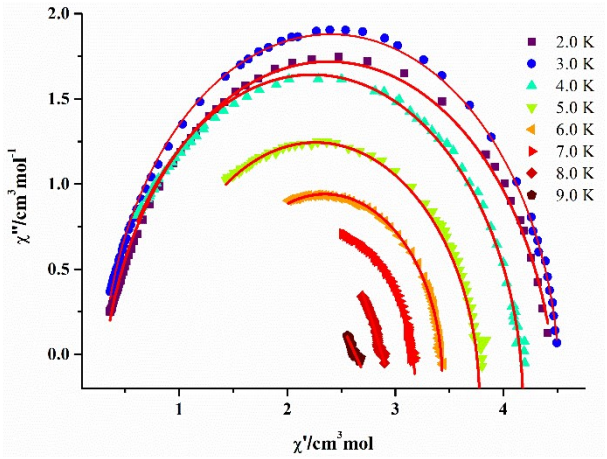

Fig. S11 Cole–Cole plots for **5** measured in zero-dc field. The solid lines are the best fit to the experimental data, obtained with the generalized Debye model with α = 0.07–0.17.