ESI for

Three new mononuclear tri-spin lanthanide-nitronyl nitroxide radical compounds: syntheses, structures and magnetic properties

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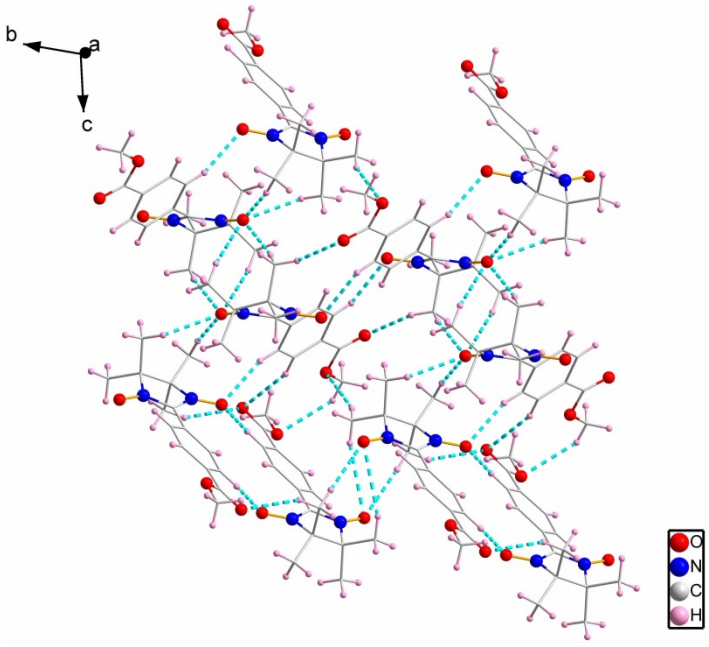

Fig. S1 Viewing of intermolecular hydrogen bonds of **1** in *bc* plane. Blue dashed lines indicate the intermolecular hydrogen bonds.

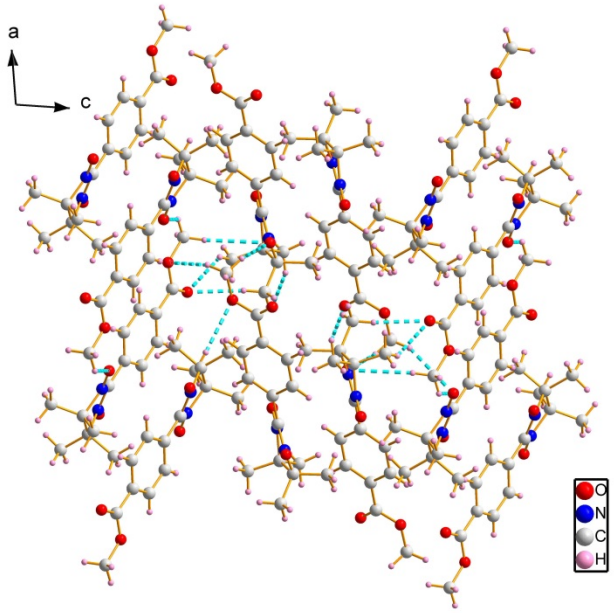

Fig. S2 Viewing of intermolecular hydrogen bonds of **1** parallel along *a* direction. Blue dashed lines indicate the intermolecular hydrogen bonds.

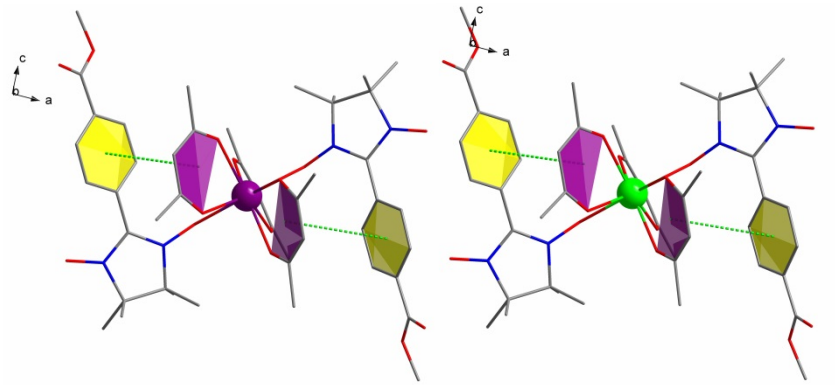

Fig. S3 Representation of the π···π stacking interactions in the molecule of **2** (left) and **3** (right).

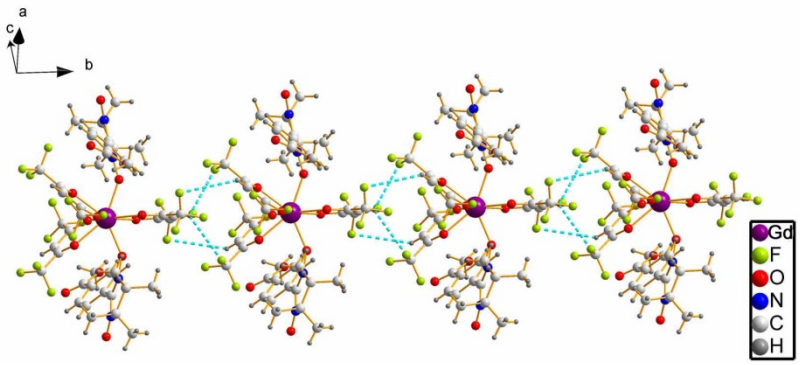

Fig. S4 Viewing of the supermolecular chain along *b* direction of **2**. Blue dashed lines represent the hydrogen bonds.

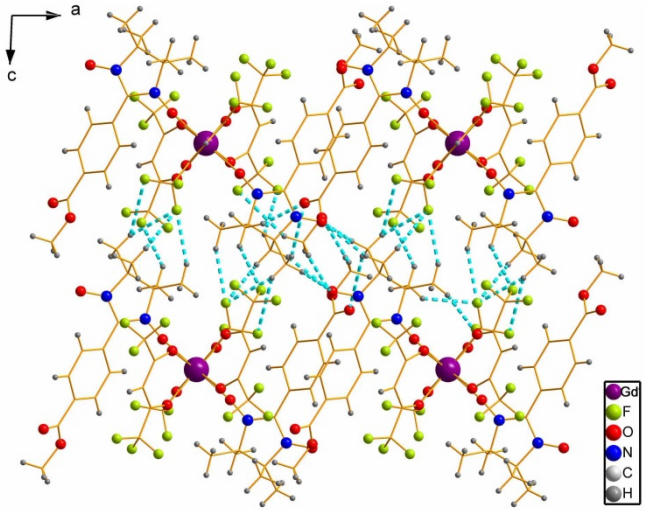

Fig. S5 Viewing of the crystal structure perpendicular *b* direction of **2**. Blue dashed lines represent the hydrogen bonds.

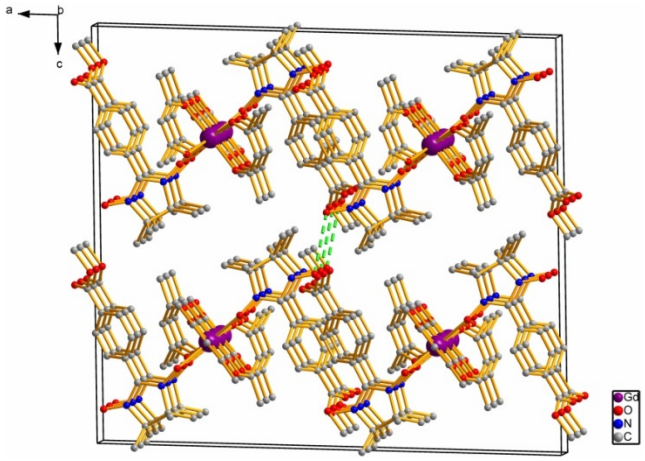

Fig. S6 Viewing of the molecular packing diagram of **2**. The green dashed lines represent the shortest contacts between NO groups.

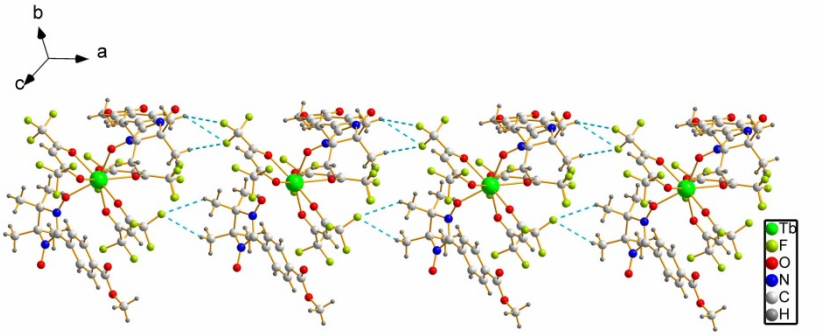

Fig. S7 Viewing of the supermolecular chain along a direction of **3**. Blue dashed lines represent the hydrogen bonds between isolated molecules.

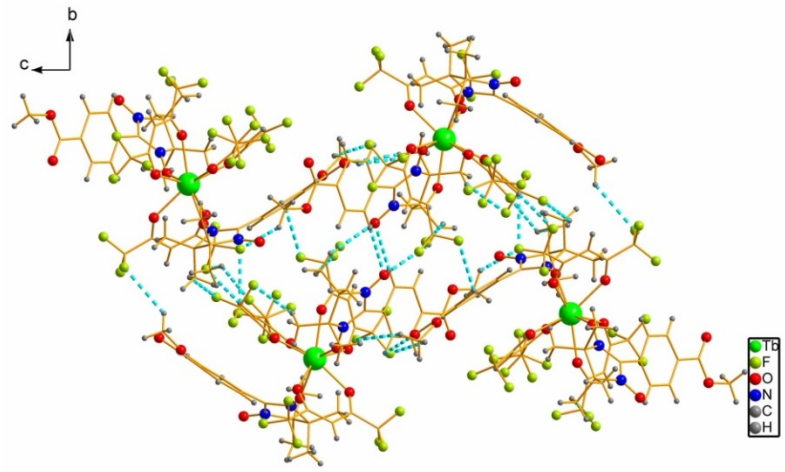

Fig. S8 Viewing of the crystal structure perpendicular *a* direction of **3**. Blue dashed lines represent the hydrogen bonds between neighboring supermolecular chains.

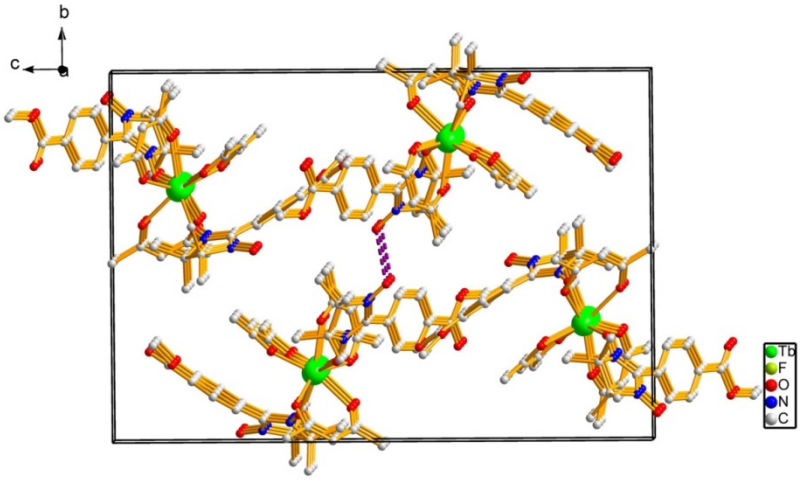

Fig. S9 Viewing of the molecular packing diagram of **3**. The violet dashed lines represent the shortest contacts between NO groups.

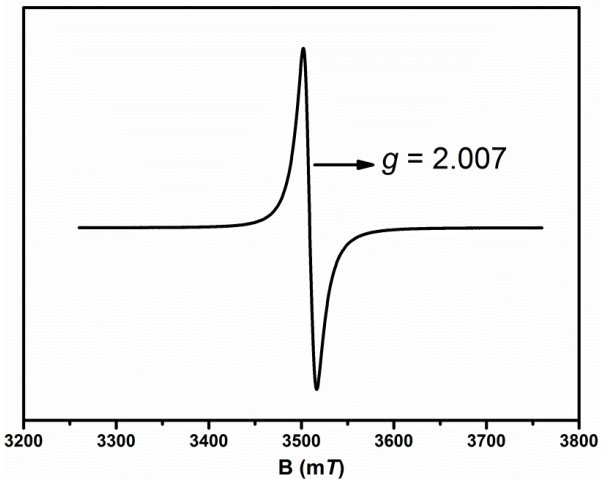

Fig. S10 EPR spectrum of organic radical **1**.

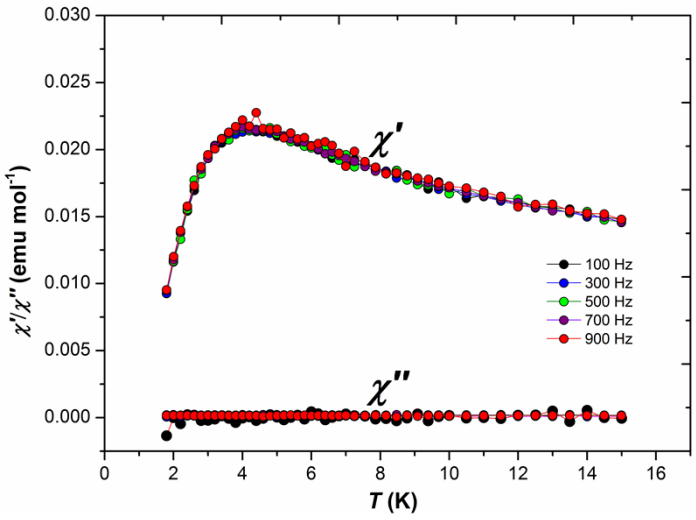

Fig. S11 The real and imaginary components of ac magnetic susceptibility for **1** under zero applied external dc field.

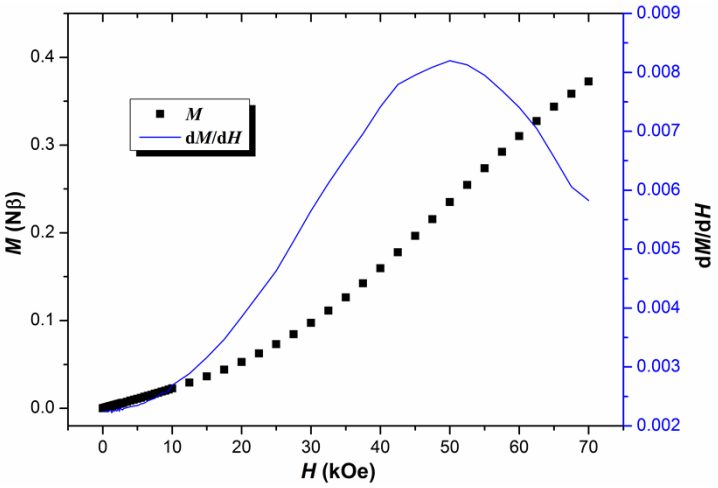

Fig. S12 *M* versus *H* and d*M*/d*H* versus *H* plots for **1** at 2.0 K.

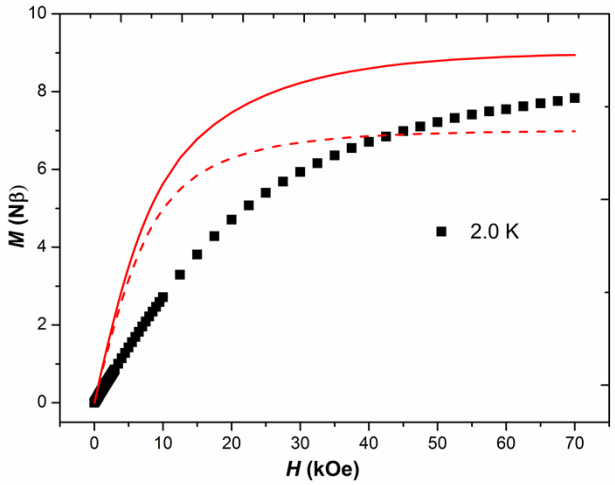

Fig. S13 Field dependence of magnetization for **2** at 2.0 K. The red solid line represents the fitting result by Brillouin function with $J = 0$ cm^{-1} and the red dashed line is the fitting result for only one Gd^{III} ion.

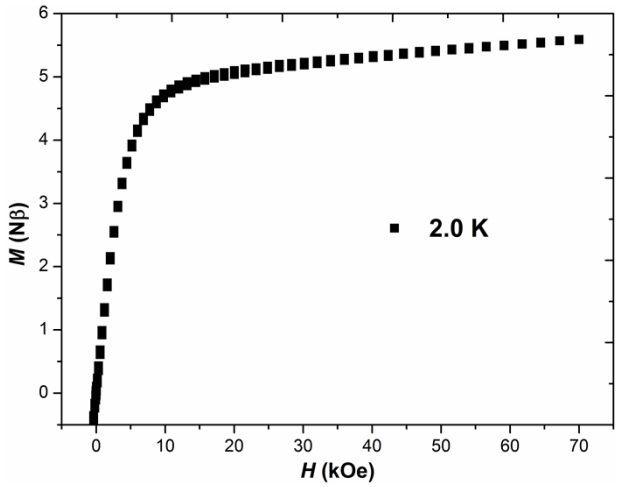

Fig. S14 Field dependence of magnetization for **3** under 2.0 K.

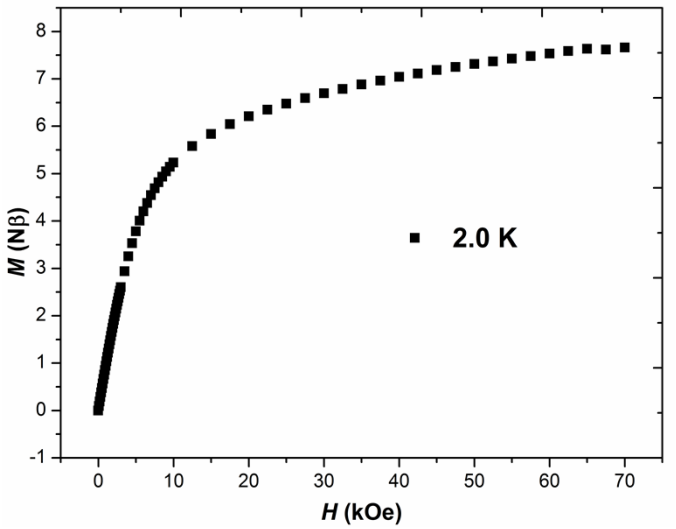

Fig. S15 Field dependence of magnetization for **4** under 2.0 K.

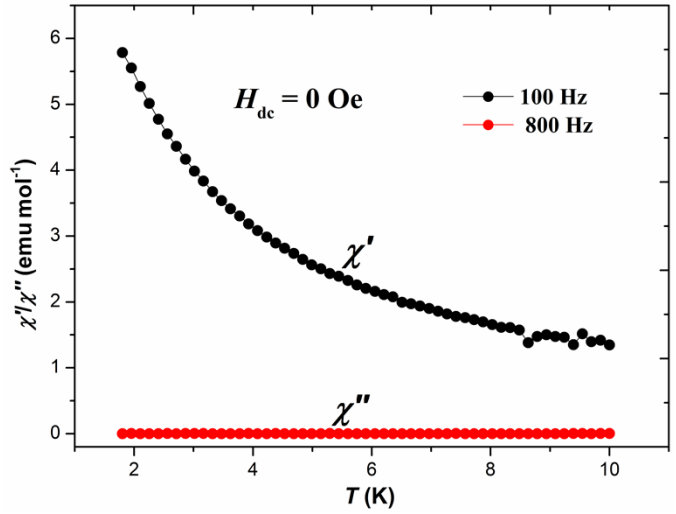

Fig. S16 The real and imaginary components of ac magnetic susceptibility for **4** under zero applied external dc field.

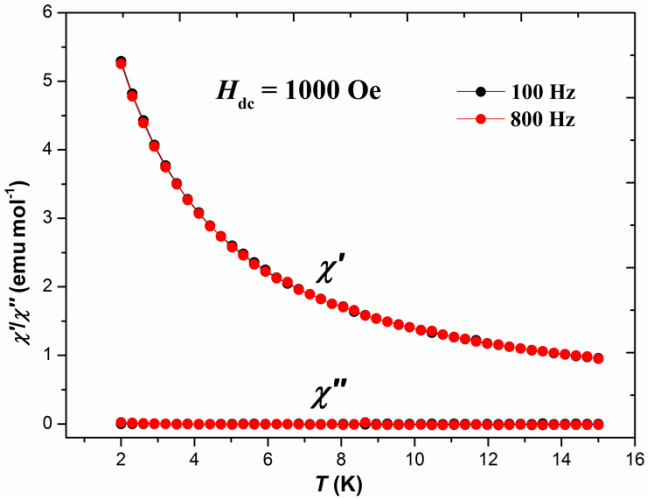

Fig. S17 The real and imaginary components of ac magnetic susceptibility for **4** under 1000 Oe applied external dc field.

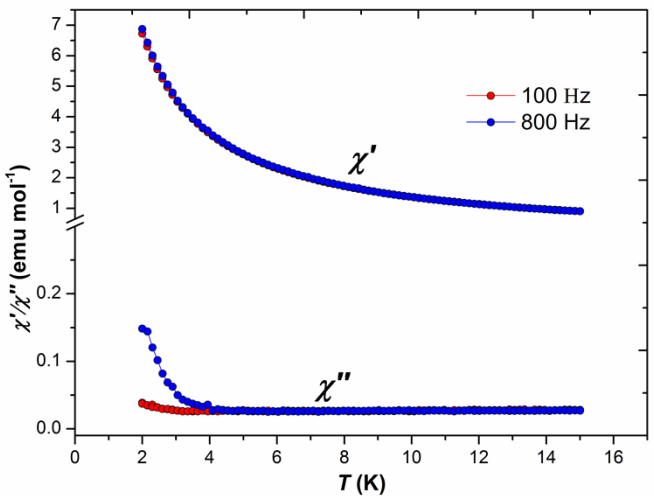

Fig. S18 The real and imaginary components of ac magnetic susceptibility for **3** under zero applied external dc field**.**

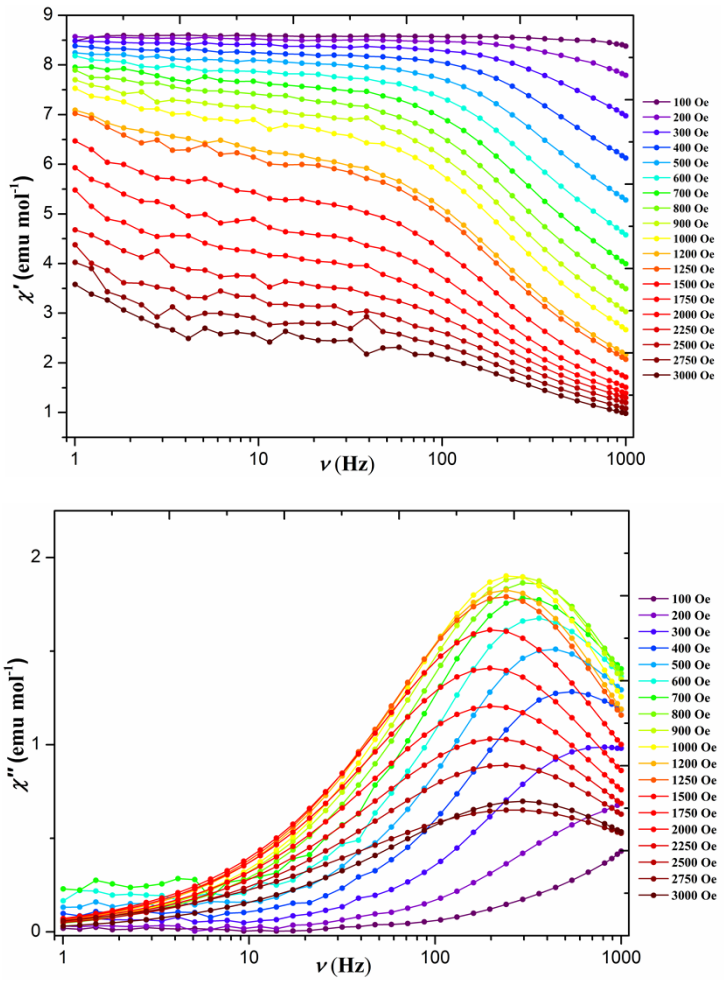

Fig. S19 The real (top) and imaginary (down) components of ac magnetic susceptibility for **3** under dc fields up to 3000 Oe at 1.8 K.

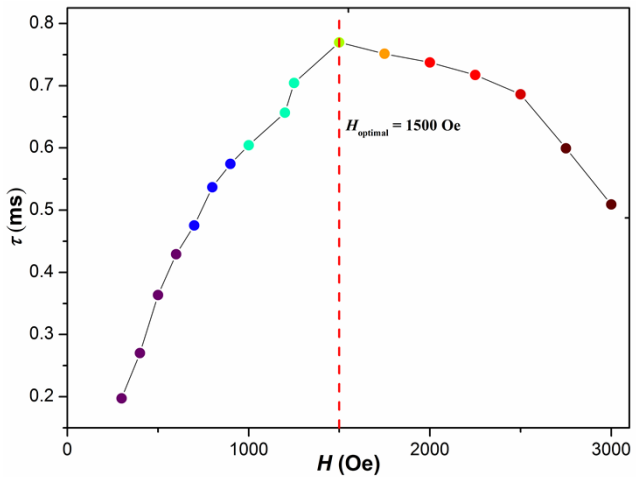

Fig. S20 Relaxation times τ under different dc fields at 1.8 K for **3**.

Table S1 Fitting parameters of Cole-Cole plots of for **3** under 1500 Oe applied dc field.

T (K)	χ_0	χ_∞	α
1.80	1.10	6.16	0.24
1.85	1.20	6.02	0.19
1.90	1.18	5.92	0.17
1.95	1.14	5.84	0.15
2.00	1.12	5.76	0.14
2.05	0.94	5.72	0.14
2.10	0.63	5.69	0.16
2.15	0.62	5.66	0.14
2.20	0.75	5.60	0.13
2.40	0.86	5.45	0.11

Table S2 Selected bonds and angles for **2** and **4.** Symmetry code:#1 -x, y, -z+1/2; #2 x+1/2, y+1/2, z; #3 -x+1/2, y+1/2, -z+1/2.

$[Gd(hfac)_3(NITPhCOOMe)_2]$ (2)		**$[Dy(hfac)_3(NITPhCOOMe)_2]$ (4)**	
Bonds/Angles	Å / °	Bonds/Angles	Å / °
Gd(1)-O(1)	2.3475(19)	Dy(1)-O(1)	2.3124(2)
Gd(1)-O(2)	2.390(2)	Dy(1)-O(2)	2.3447(3)
Gd(1)-O(3)	2.3564(17)	Dy(1)-O(3)	2.3132(2)
Gd(1)-O(4)	2.3875(19)	Dy(1)-O(4)	2.3473(3)
O(1)-Gd(1)-O(1)#1	138.54(11)	O(1)#1-Dy(1)-O(1)	138.21(14)
O(1)-Gd(1)-O(3)	98.60(6)	O(1)#1-Dy(1)-O(3)#1	98.82(8)
O(1)#1-Gd(1)-O(3)	95.29(6)	O(1)-Dy(1)-O(3)#1	94.80(8)
O(1)-Gd(1)-O(3)#1	95.29(6)	O(1)#1-Dy(1)-O(3)	94.80(8)
O(1)#1-Gd(1)-O(3)#1	98.60(6)	O(1)-Dy(1)-O(3)	98.82(8)
O(3)-Gd(1)-O(3)#1	140.06(10)	O(3)#1-Dy(1)-O(3)	141.18(14)
O(1)-Gd(1)-O(4)	73.45(7)	O(1)#1-Dy(1)-O(2)#1	73.30(9)
O(1)#1-Gd(1)-O(4)	73.50(7)	O(1)-Dy(1)-O(2)#1	73.66(9)
O(3)-Gd(1)-O(4)	73.47(7)	O(3)#1-Dy(1)-O(2)#1	72.31(9)
O(3)#1-Gd(1)-O(4)	146.46(7)	O(3)-Dy(1)-O(2)#1	146.49(9)
O(1)-Gd(1)-O(4)#1	73.50(7)	O(1)#1-Dy(1)-O(2)	73.66(9)
O(1)#1-Gd(1)-O(4)#1	73.45(7)	O(1)-Dy(1)-O(2)	73.30(9)
O(3)-Gd(1)-O(4)#1	146.46(7)	O(3)#1-Dy(1)-O(2)	146.49(9)
O(3)#1-Gd(1)-O(4)#1	73.47(7)	O(3)-Dy(1)-O(2)	72.31(9)
O(4)-Gd(1)-O(4)#1	73.04(9)	O(2)#1-Dy(1)-O(2)	74.27(12)
O(1)-Gd(1)-O(2)#1	148.95(8)	O(1)#1-Dy(1)-O(4)	72.52(9)
O(1)#1-Gd(1)-O(2)#1	72.39(7)	O(1)-Dy(1)-O(4)	149.20(10)
O(3)-Gd(1)-O(2)#1	76.38(7)	O(3)#1-Dy(1)-O(4)	73.87(9)
O(3)#1-Gd(1)-O(2)#1	72.58(7)	O(3)-Dy(1)-O(4)	75.95(9)
O(4)-Gd(1)-O(2)#1	131.43(6)	O(2)#1-Dy(1)-O(4)	126.60(8)
O(4)#1-Gd(1)-O(2)#1	126.64(7)	O(2)-Dy(1)-O(4)	130.72(8)
O(1)-Gd(1)-O(2)	72.39(7)	O(1)#1-Dy(1)-O(4)#1	149.20(10)
O(1)#1-Gd(1)-O(2)	148.95(8)	O(1)-Dy(1)-O(4)#1	72.52(9)
O(3)-Gd(1)-O(2)	72.58(7)	O(3)#1-Dy(1)-O(4)#1	75.95(9)
O(3)#1-Gd(1)-O(2)	76.38(7)	O(3)-Dy(1)-O(4)#1	73.87(9)
O(4)-Gd(1)-O(2)	126.64(7)	O(2)#1-Dy(1)-O(4)#1	130.72(8)
O(4)#1-Gd(1)-O(2)	131.43(6)	O(2)-Dy(1)-O(4)#1	126.60

Table S3 Selected bonds and angles for **3**. # 1 -x, y+1/2, -z+1/2**.**

[Tb(hfac)$_3$(NITPhCOOMe)$_2$]			
Bonds/Angles	Å / °	Bonds/Angles	Å / °
Tb(1)-O(1)	2.350(2)	Tb(1)-O(5)	2.375(2)
Tb(1)-O(2)	2.319(2)	Tb(1)-O(6)	2.333(2)
Tb(1)-O(3)	2.371(2)	Tb(1)-O(7)	2.397(3)
Tb(1)-O(4)	2.357(3)	Tb(1)-O(8)	2.331(3)
O(2)-Tb(1)-O(8)	103.84(9)	O(4)-Tb(1)-O(3)	73.82(9)
O(2)-Tb(1)-O(6)	90.19(9)	O(2)-Tb(1)-O(5)	148.74(9)
O(8)-Tb(1)-O(6)	136.60(9)	O(8)-Tb(1)-O(5)	73.49(9)
O(2)-Tb(1)-O(1)	137.15(8)	O(6)-Tb(1)-O(5)	73.69(9)
O(8)-Tb(1)-O(1)	92.85(9)	O(1)-Tb(1)-O(5)	73.74(8)
O(6)-Tb(1)-O(1)	104.32(9)	O(4)-Tb(1)-O(5)	125.17(9)
O(2)-Tb(1)-O(4)	73.40(9)	O(3)-Tb(1)-O(5)	132.99(9)
O(8)-Tb(1)-O(4)	148.30(9)	O(2)-Tb(1)-O(7)	73.55(8)
O(6)-Tb(1)-O(4)	74.98(9)	O(8)-Tb(1)-O(7)	73.04(10)
O(1)-Tb(1)-O(4)	72.00(8)	O(6)-Tb(1)-O(7)	72.07(9)
O(2)-Tb(1)-O(3)	72.97(8)	O(1)-Tb(1)-O(7)	149.22(8)
O(8)-Tb(1)-O(3)	75.24(9)	O(4)-Tb(1)-O(7)	132.55(9)
O(6)-Tb(1)-O(3)	147.67(9)	O(3)-Tb(1)-O(7)	125.85(9)
O(1)-Tb(1)-O(3)	73.72(8)	O(5)-Tb(1)-O(7)	76.01(9)