Electronic Supplementary Information

Construction of $Ni^{II}Ln^{III}M^{III}$ (Ln = Gd^{III}, Tb^{III}; M = Fe^{III}, Cr^{III}) Clusters Showing Slow Magnetic Relaxations

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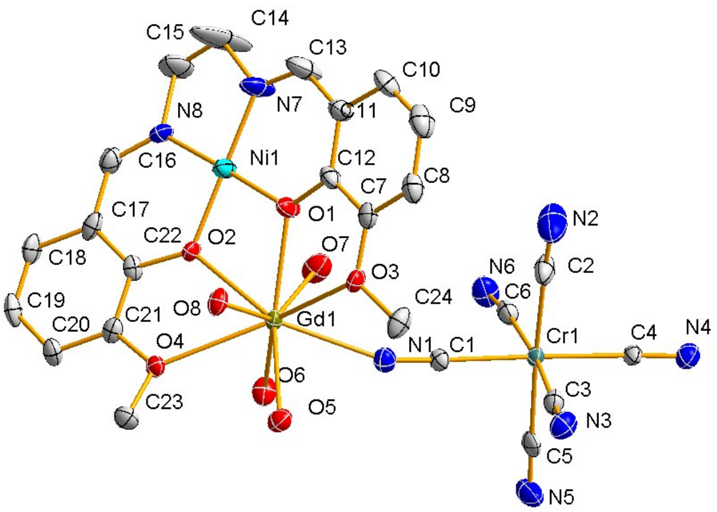
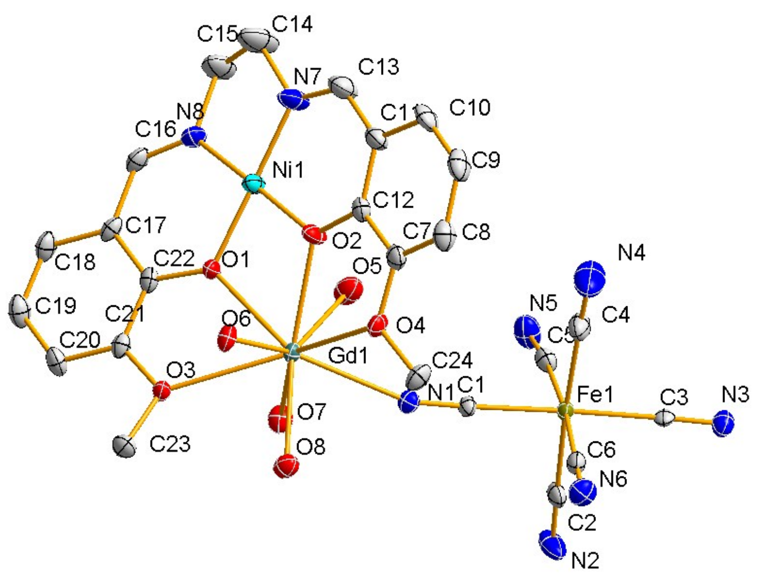


(a)



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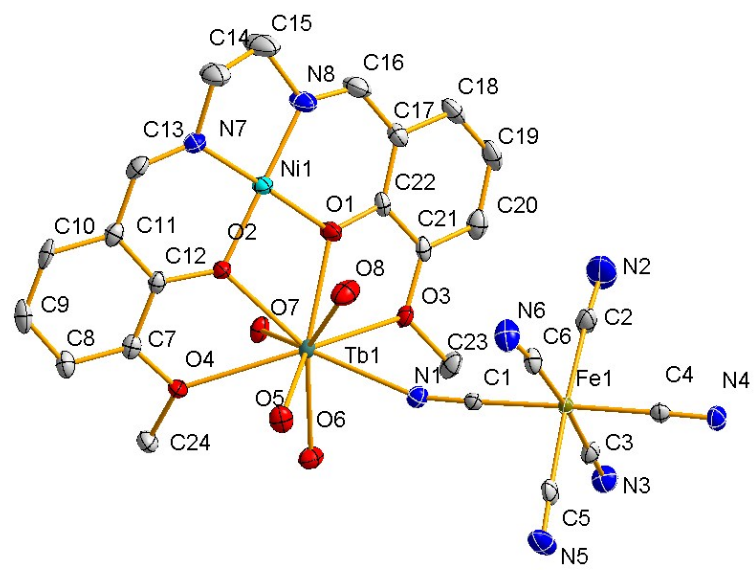
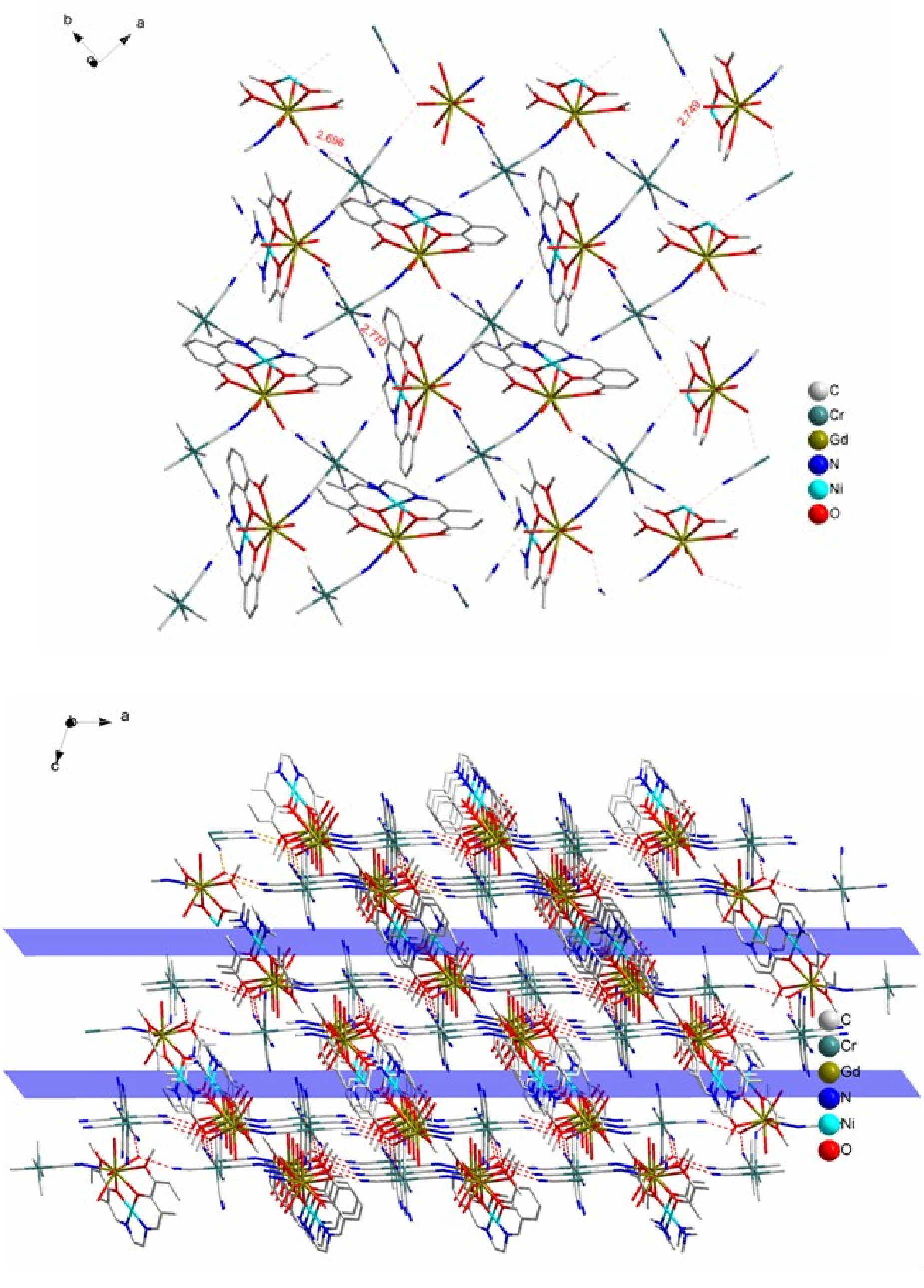
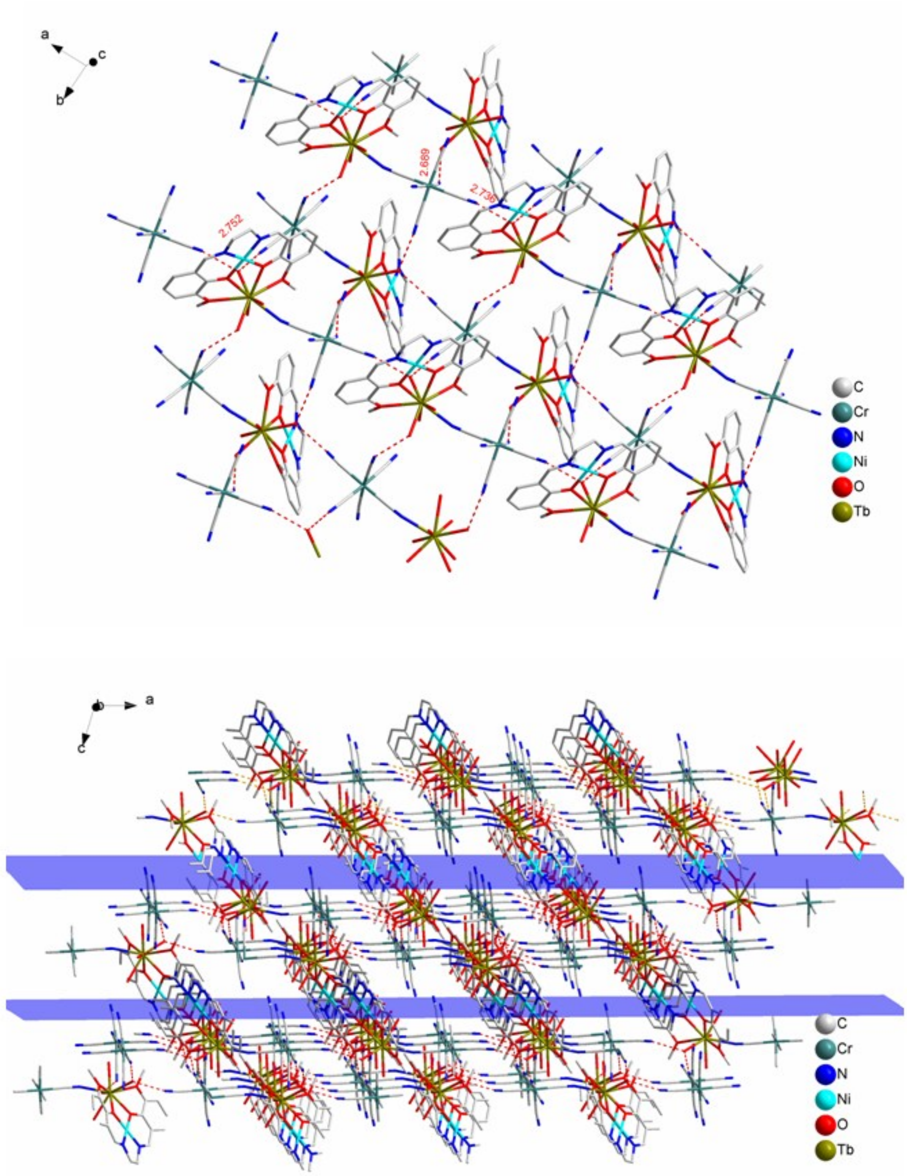


(c)

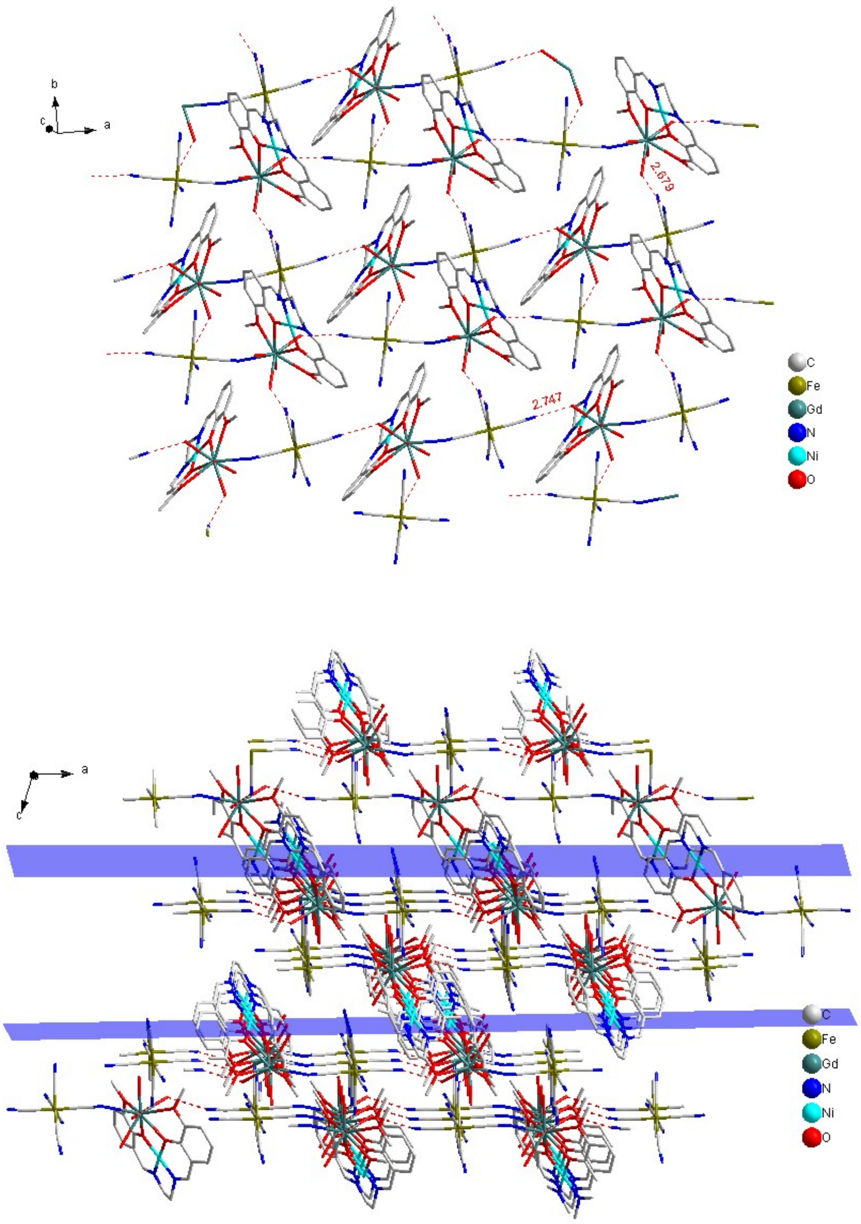
Figure S1 ORTEP (30%) diagrams of asymmetric units with selected atom-labeling schemes for complex **2**(a), **3**(b) and **4**(c).



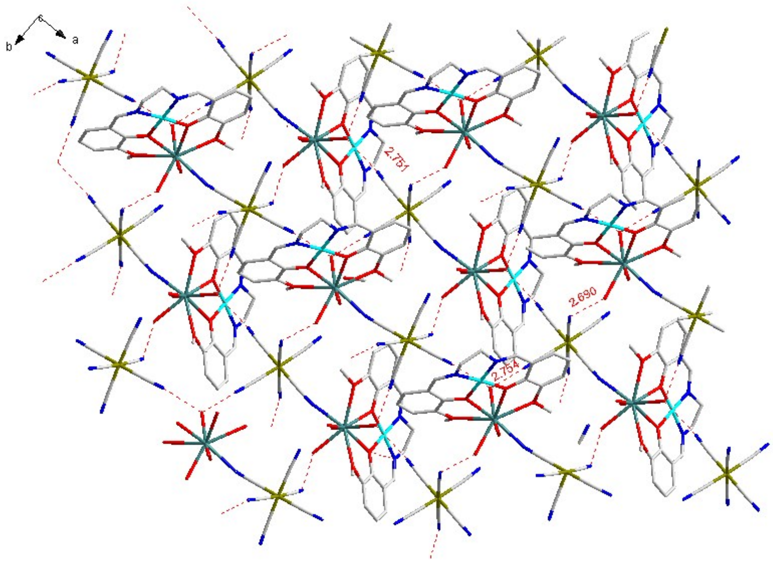
(a)

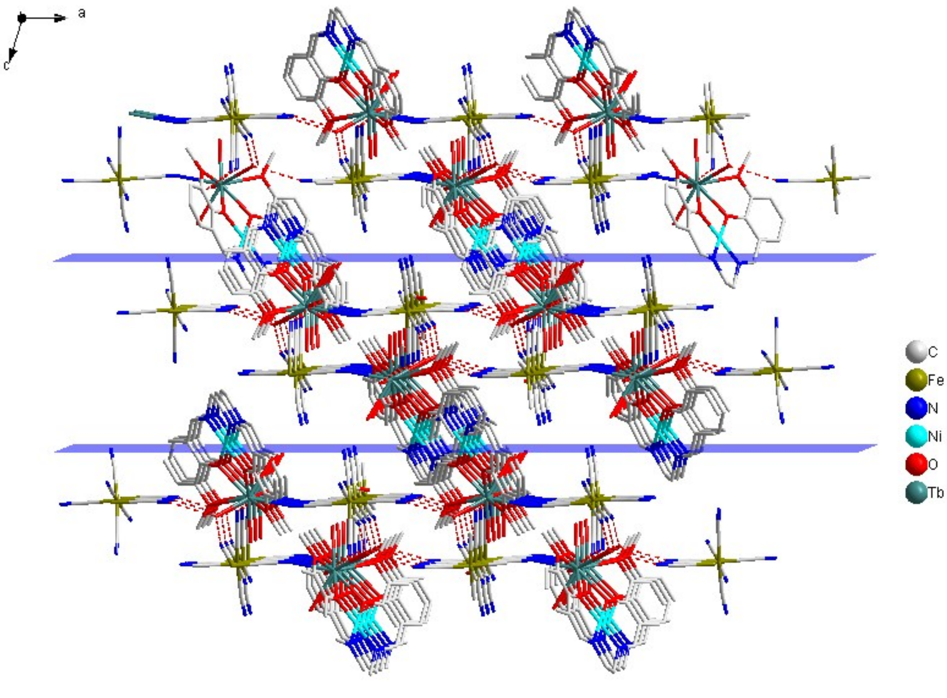


(b)



(c)





(d)

Figure S2 The extended molecular structures of complex **1**(a), **2**(b), **3**(c) and **4**(d).

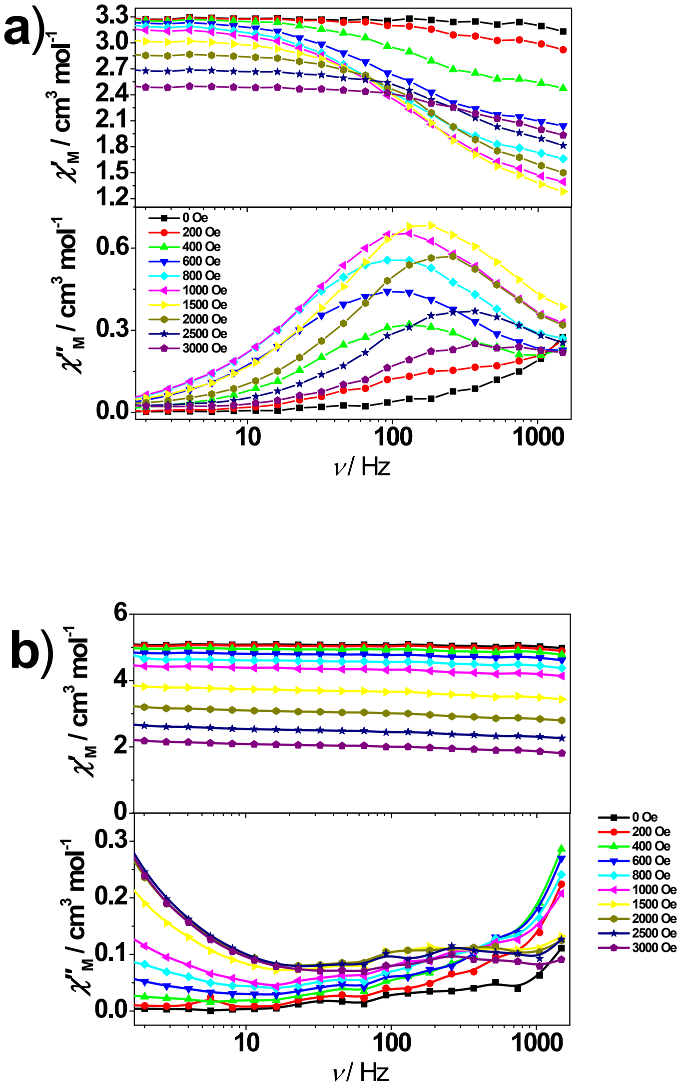


Figure S3 Field dependence of the in-phase (χ_M') and out-of-phase (χ_M'') *ac* susceptibilities of **2**(a) and **4**(b) in different *dc* field and 3 Oe *ac* field at 1.8 K (Solid lines are guides for the eyes).

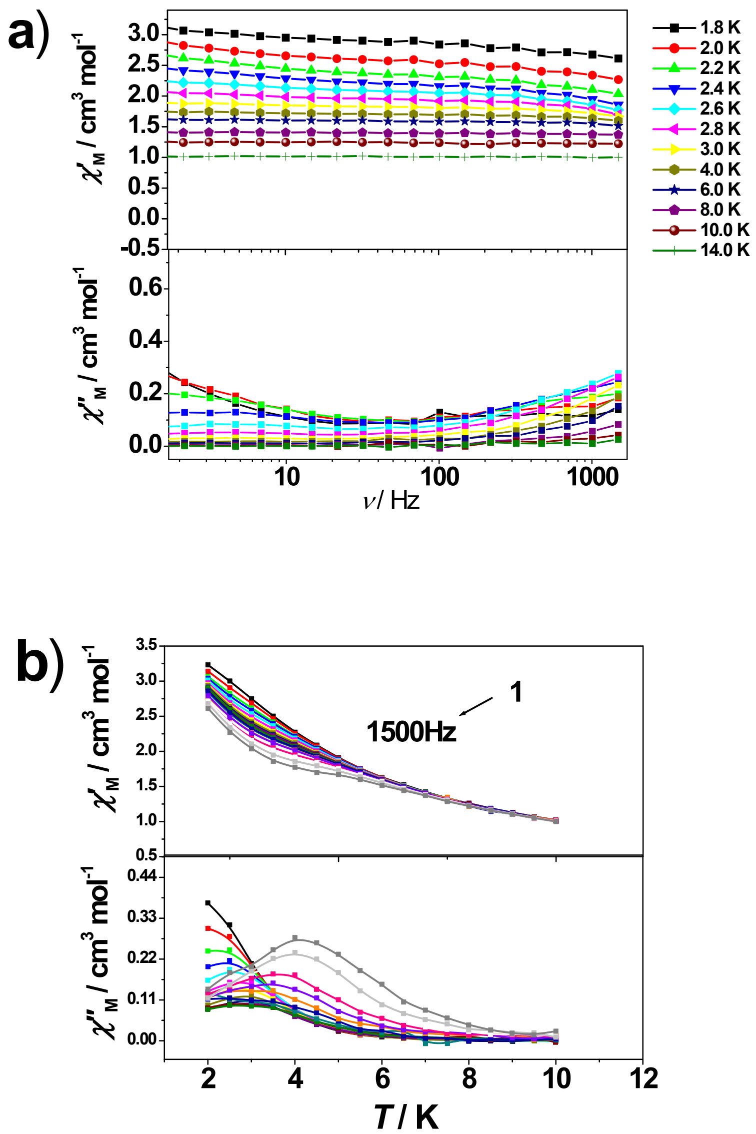


Figure S4 Frequency (a) and temperature (b) dependence of the in-phase (χ_M') and out-of-phase (χ_M'') *ac* susceptibilities of **4** in 2000 Oe *dc* field and 3 Oe *ac* field (Solid lines are guides for the eyes).

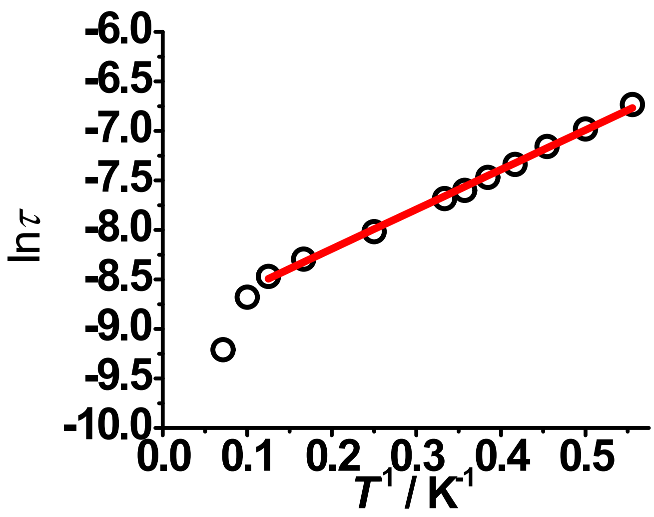


Figure. S5 The fitting of the relaxation time (τ) using Arrhenius law for complex **2**

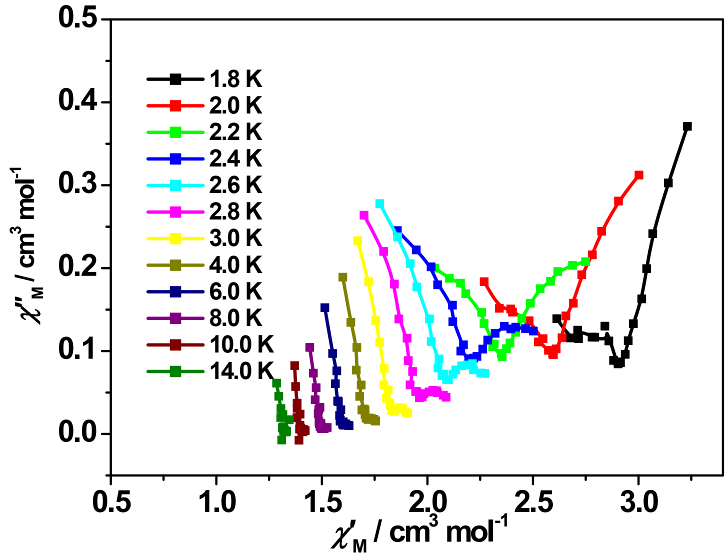


Figure. S6 The Cole-Cole plots of complex **4** measured under 2 kOe *dc* field and 3 Oe *ac* field**.** (Solid lines are guides for the eyes)

Table S1. Selected bond lengths [Å] and angles [deg°] for **1**-**4**.

1		**2**		**3**		**4**	
C1-Cr1	2.063(6)	C1-Cr1	2.066(4)	C1-Fe1	1.930(4)	C1-Fe1	1.931(4)
C2-Cr1	2.076(7)	C2-Cr1	2.079(5)	C2-Fe1	1.936(4)	C2-Fe1	1.953(5)
C3-Cr1	2.045(7)	C3-Cr1	2.070(5)	C3-Fe1	1.937(4)	C3-Fe1	1.922(4)
C4-Cr1	2.052(6)	C4-Cr1	2.057(5)	C4-Fe1	1.949(5)	C4-Fe1	1.945(4)
C5-Cr1	2.060(7)	C5-Cr1	2.069(5)	C5-Fe1	1.934(4)	C5-Fe1	1.942(5)
C6-Cr1	2.080(7)	C6-Cr1	2.058(4)	C6-Fe1	1.924(5)	C6-Fe1	1.940(4)

Gd1-O8	2.359(4)	N1-Tb1	2.475(4)	Gd1-O1	2.364(3)	N1-Tb1	2.486(3)
Gd1-O1	2.372(4)	N7-Ni1	1.830(4)	Gd1-O6	2.371(3)	N7-Ni1	1.844(4)
Gd1-O6	2.374(4)	N8-Ni1	1.838(4)	Gd1-O7	2.375(3)	N8-Ni1	1.840(4)
Gd1-O2	2.374(4)	Ni1-O2	1.845(3)	Gd1-O2	2.378(3)	Ni1-O2	1.848(3)
Gd1-O5	2.423(4)	Ni1-O1	1.851(3)	Gd1-O8	2.425(3)	Ni1-O1	1.850(3)
Gd1-N1	2.496(5)	Ni1-Tb1	3.3706(12)	Gd1-N1	2.489(4)	Ni1-Tb1	3.3861(12)
Gd1-O7	2.520(4)	O1-Tb1	2.357(3)	Gd1-O5	2.521(3)	O1-Tb1	2.378(3)
Gd1-O3	2.607(4)	O2-Tb1	2.355(3)	Gd1-O4	2.598(3)	O2-Tb1	2.366(3)
Gd1-O4	2.656(4)	O3-Tb1	2.663(3)	Gd1-O3	2.664(3)	O3-Tb1	2.598(3)
Gd1-Ni1	3.3890(13)	O4-Tb1	2.608(3)	Gd1-Ni1	3.3864(12)	O4-Tb1	2.668(3)
N7-Ni1	1.833(6)	O5-Tb1	2.403(3)	N7-Ni1	1.841(4)	O5-Tb1	2.369(3)
N8-Ni1	1.836(5)	O6-Tb1	2.335(3)	N8-Ni1	1.833(4)	O6-Tb1	2.423(3)
Ni1-O2	1.845(4)	O7-Tb1	2.355(3)	Ni1-O2	1.844(3)	O7-Tb1	2.372(3)
Ni1-O1	1.852(4)	O8-Tb1	2.516(3)	Ni1-O1	1.850(3)	O8-Tb1	2.528(3)
N1-C1-Cr1	177.7(5)	N1-C1-Cr1	176.6(4)	N1-C1-Fe1	177.9(4)	N1-C1-Fe1	177.9(4)
N2-C2-Cr1	175.7(7)	N2-C2-Cr1	176.2(6)	N2-C2-Fe1	178.9(4)	N2-C2-Fe1	175.9(5)
N3-C3-Cr1	176.8(6)	N3-C3-Cr1	172.6(4)	N3-C3-Fe1	175.4(4)	N3-C3-Fe1	177.7(4)
N4-C4-Cr1	174.2(6)	N4-C4-Cr1	176.3(4)	N4-C4-Fe1	176.5(5)	N4-C4-Fe1	175.3(4)
N5-C5-Cr1	179.1(7)	N5-C5-Cr1	178.8(4)	N5-C5-Fe1	173.5(4)	N5-C5-Fe1	178.5(4)
N6-C6-Cr1	172.5(6)	N6-C6-Cr1	174.0(4)	N6-C6-Fe1	177.5(4)	N6-C6-Fe1	173.7(4)
C1-N1-Gd1	160.6(5)	C1-N1-Tb1	162.9(3)	C1-N1-Gd1	162.1(3)	C1-N1-Tb1	162.3(3)

Fit of Cole-Cole plots. The derivation of Debye model mentioned in the text is applied and displayed here:

$$\chi'(\omega)=\beta\left(\chi_S+\frac{(\chi_T-\chi_S)[1+(\omega\tau_1)^{1-\alpha_1}\sin{}^1/_2\,\alpha_1\pi]}{1+2(\omega\tau_1)^{1-\alpha_1}\sin{}^1/_2\,\alpha_1\pi+(\omega\tau_1)^{2(1-\alpha_1)}}\right)+(1-\beta)\left(\chi_S+\frac{(\chi_T-\chi_S)[1+(\omega\tau_2)^{1-\alpha_2}\sin{}^1/_2\,\alpha_2\pi]}{1+2(\omega\tau_2)^{1-\alpha_2}\sin{}^1/_2\,\alpha_2\pi+(\omega\tau_2)^{2(1-\alpha_2)}}\right)$$

$$\chi''(\omega)=\beta\left(\frac{(\chi_T-\chi_S)(\omega\tau_1)^{1-\alpha_1}\cos{}^1/_2\,\alpha_1\pi}{1+2(\omega\tau_1)^{1-\alpha_1}\sin{}^1/_2\,\alpha_1\pi+(\omega\tau_1)^{2(1-\alpha_1)}}\right)+(1-\beta)\left(\frac{(\chi_T-\chi_S)(\omega\tau_2)^{1-\alpha_2}\cos{}^1/_2\,\alpha_2\pi}{1+2(\omega\tau_2)^{1-\alpha_2}\sin{}^1/_2\,\alpha_2\pi+(\omega\tau_2)^{2(1-\alpha_2)}}\right)$$

Table S2 Parameters in double magnetic relaxations for complex **2**

T / K	α		τ		χ		β
	α_1	α_2	τ_1	τ_2	χ_T	χ_s	
1.8	0.14	0.11	0.002	0.00033	1.3	3.2	0.66
2.0	0.11	0.13	0.00028	0.00156	1.8	3.0	0.35
2.2	0.09	0.13	0.00027	0.00133	1.1	2.9	0.37
2.4	0.13	0.08	0.00111	0.00023	1.1	2.7	0.62
2.6	0.13	0.1	0.00106	0.00024	1.0	2.6	0.55
2.8	0.13	0.09	0.00087	0.00021	1.0	2.5	0.60
3.0	0.13	0.05	0.00079	0.00018	0.96	2.36	0.6
4.0	0.13	0.095	0.00063	0.00017	0.79	1.97	0.5
6.0	0.13	0.07	0.00051	0.00013	0.61	1.48	0.46

8.0	0.10	0.04	0.00041	0.0001	0.48	1.18	0.50
10.0	0.10	0.08	0.00052	0.00013	0.42	0.98	0.21
14.0	0.001	0.05	0.00076	0.0001	0.32	0.73	0.07