Supporting Information

TABLE OF CONTENTS

S1

Table S1: Crystallographic data and refinement for complexes **1-4**.

	1	2	3	4
formula	$C_{74}H_{90}BFeN_4O_2$	$C_{78}H_{88}BFeN_4O$	$C_{78}H_{72}BF_{24}FeN_4$	$C_{86}H_{80}BF_{24}FeN_4$
Mr	1134.15	1164.18	1588.05	1692.20
cryst syst	monoclinic	monoclinic	triclinic	monoclinic
space group	$P2_1$	$P2_1/c$	$P\bar{1}$	$P2_1$
a, Å	13.0956(15)	19.071(3)	12.520(2)	18.7708(15)
b, Å	18.1551(19)	16.663(2)	15.717(3)	16.9182(14)
c, Å	13.6087(15)	21.656(3)	19.186(3)	26.489(2)
α, deg	90	90	79.488(3)	90
β, deg	91.873(2)	106.455(3)	84.243(3)	106.1770(10)
γ, deg	90	90	89.168(3)	90
V, Å^3	3233.8(6)	6600.0(16)	3693.1(10)	8079.1(11)
Z	2	4	2	4
T, K	130	130	130	130
μ, mm^{-1}	0.281	0.276	0.312	0.290
λ, Å	0.71073	0.71073	0.71073	0.71073
Cryst size, mm^3	0.20 0.05 0.03	0.38 0.28 0.20	0.22 0.20 0.16	0.32 0.20 0.18
GOF	0.962	1.020	0.957	0.993
R_{int}	0.0656	0.0827	0.0591	0.0516
R_1, $wR_2[I > 2\sigma(I)]$	0.0702, 0.1122	0.0589, 0.1124	0.0920, 0.2481	0.0719, 0.1751
R_1, wR_2[all data]	0.173, 0.1383	0.1317, 01404	0.1543, 0.2852	0.1761, 0.2275
CCDC code	1426589	1882118	1426588	1426587

Table S2: Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **1** according to the generalized Debye model.

T / K	χ_S / cm^3 mol^{-1} K	χ_T / cm^3 mol^{-1} K	α	τ / s
2	0.0214	1.2359	0.1928	0.00884
2.25	0.02159	0.90554	0.14772	0.00338
2.5	0.02129	0.82909	0.12568	0.00191
2.75	0.02075	0.79103	0.1157	0.00119
3	0.01992	0.74734	0.10794	0.000747
3.25	0.02011	0.69197	0.09133	0.000474
3.5	0.02053	0.64204	0.07395	0.00311
3.75	0.02104	0.6018	0.06229	0.000212
4	0.02184	0.56517	0.05151	0.000148
4.25	0.02284	0.53275	0.042	0.0001067
4.5	0.02213	0.50605	0.04285	0.00007848
4.75	0.02174	0.4804	0.0408	0.0000585
5	0.02055	0.45618	0.03966	0.0000439
5.25	0.0169	0.43615	0.04654	0.0000334
5.5	0.01537	0.41605	0.04216	0.0000258
5.75	0.01424	0.39919	0.04266	0.0000202
6	0.0154	0.38268	0.03969	0.0000161

Table S3: Relaxation fitting parameters from the least-square fitting of the Cole-Cole plots of **2** according to the generalized Debye model under 2 kOe dc field.

T / K	χ_S / cm^3 mol^{-1} K	χ_T / cm^3 mol^{-1} K	α	τ / s
3	0.01495	1.86093	0.18645	0.01553
3.25	0.01505	1.24621	0.13108	0.00485
3.5	0.01454	1.06609	0.1014	0.00239
3.75	0.01418	0.9626	0.07956	0.00137
4	0.01343	0.90131	0.07036	0.000855
4.25	0.01327	0.84131	0.05763	0.000553
4.5	0.01282	0.79205	0.05108	0.000376
4.75	0.01244	0.75242	0.04791	0.000266
5	0.01246	0.71373	0.04252	0.000192
5.25	0.01286	0.67946	0.03782	0.000142
5.5	0.01157	0.65017	0.04181	0.0001065
5.75	0.01293	0.62292	0.03655	0.0000821
6	0.00981	0.06195	0.05836	0.0000634
6.25	0.00498	0.57501	0.04345	0.0000503
6.5	0	0.55546	0.06065	0.0000393
6.75	0	0.53467	0.04551	0.0000326
7	0	0.51584	0.05233	0.0000262
7.25	0	0.49824	0.05482	0.0000214
7.5	0	0.48163	0.058	0.0000177
7.75	0	0.46603	0.05019	0.0000149
8	0	0.45185	0.05543	0.0000125

Table S4: Calculated low-lying spin-orbit energy levels based on CASSCF method.

	1	2	3	4
	Calculated low-lying spin-orbit energy levels based on CASSCF method			
1	0	0	0	0
2	119.475	226	103	132
3	1055	581	732	260
4	1189	815	935	496
5	1601	1319	1251	634
6	1721	1403	1369	762
7	2353	2093	1601	848
8	2510	2223	1782	1075
Calculated zero-field splitting (ZFS) parameters: the axial D and rhombic E parameters of the ground manifold (pseudospin S=3/2)				
D	-58.7	-112.9	46.6	-65.3
E	6.3	2.5	12.7	5.6

Table S4: Calculated low-lying spin-orbit energy levels based on DDCI3 method.

	1	2	3	4
	Calculated low-lying spin-orbit energy levels based on DDCI3 method			
1	0	0	0	0
2	76.2	126	71	127
3	1614	1066	1486	412
4	1704	1198	1669	711
5	2161	1684	1939	771
6	2338	1798	2022	848
7	2775	2350	2245	1024
8	2984	2513	2391	1134
Calculated zero-field splitting (ZFS) parameters: the axial D and rhombic E parameters of the ground manifold (pseudospin S=3/2)				
D	-38.3	-62.9	34.0	-62.4
E	6.5	5.5	3.1	11.7

Figure S1: Variable-field-variable-temperature magnetization measurement for **1-4** (a-d)
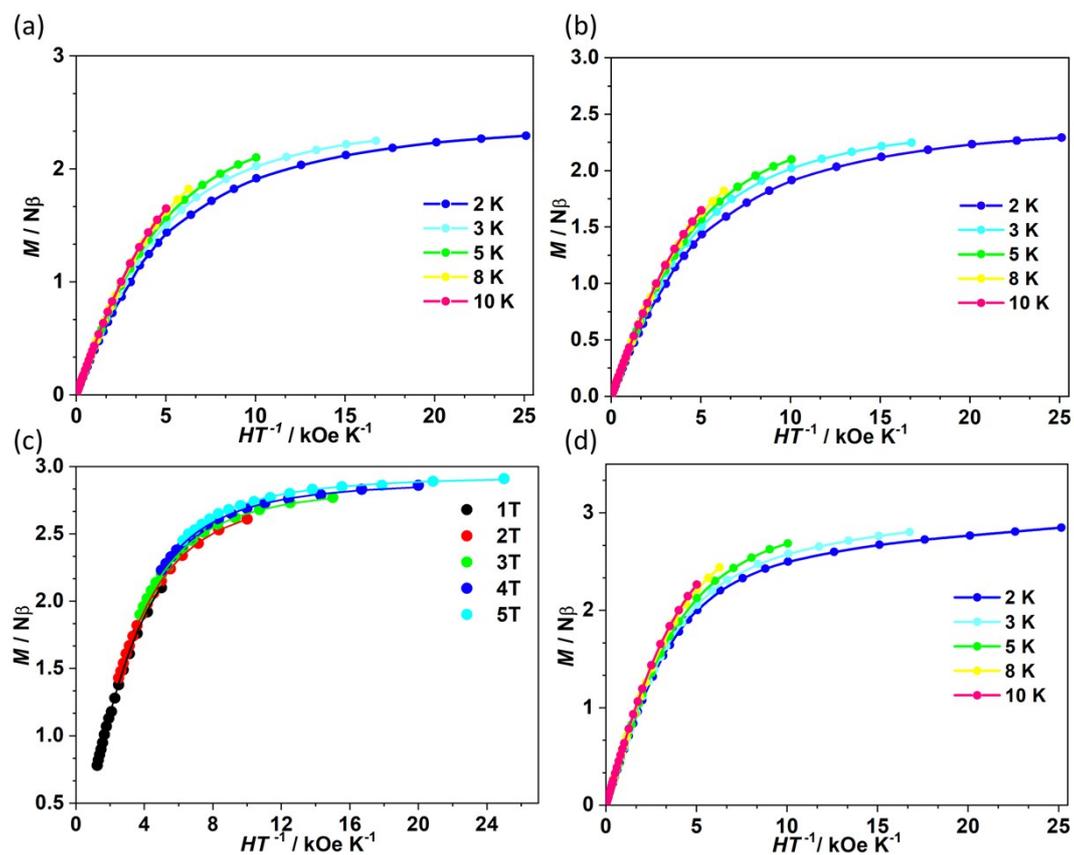
Figure S2. Temperature dependence ac susceptibility in the absence of dc field for **1** and **2**.
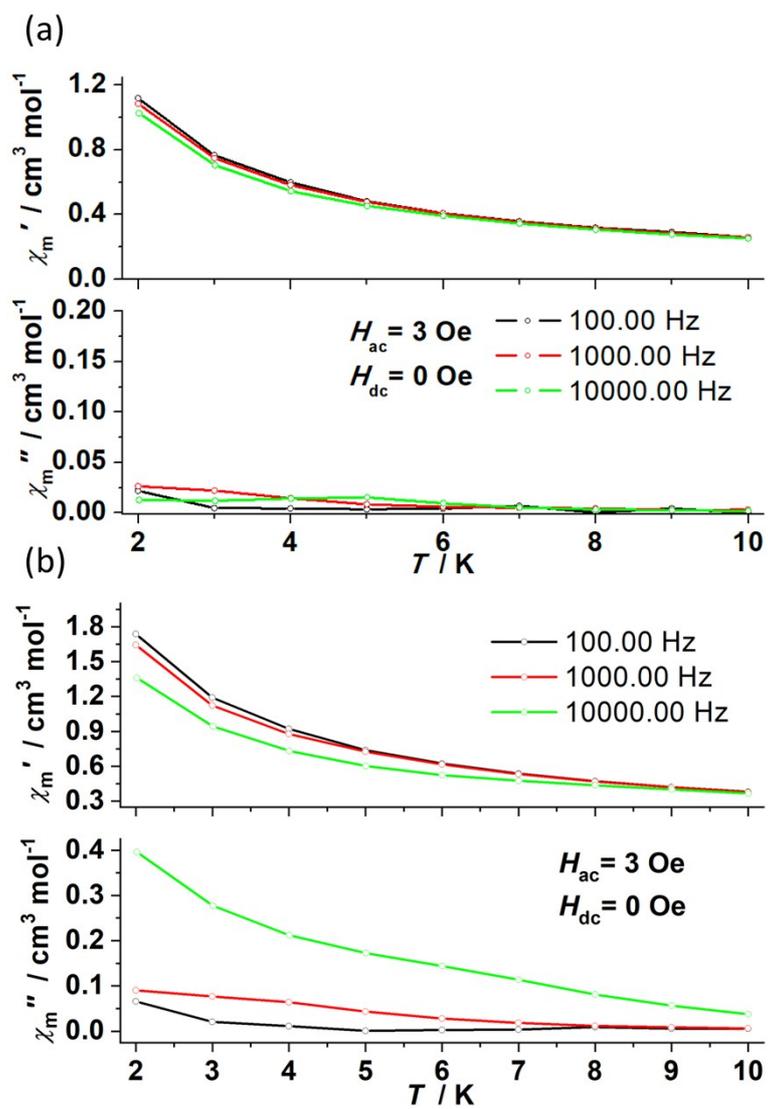
Figure S3: Cole-Cole plots fitting for the determination of the temperature dependence of τ for **1** (a) and **2** (b) under 2 kOe dc field
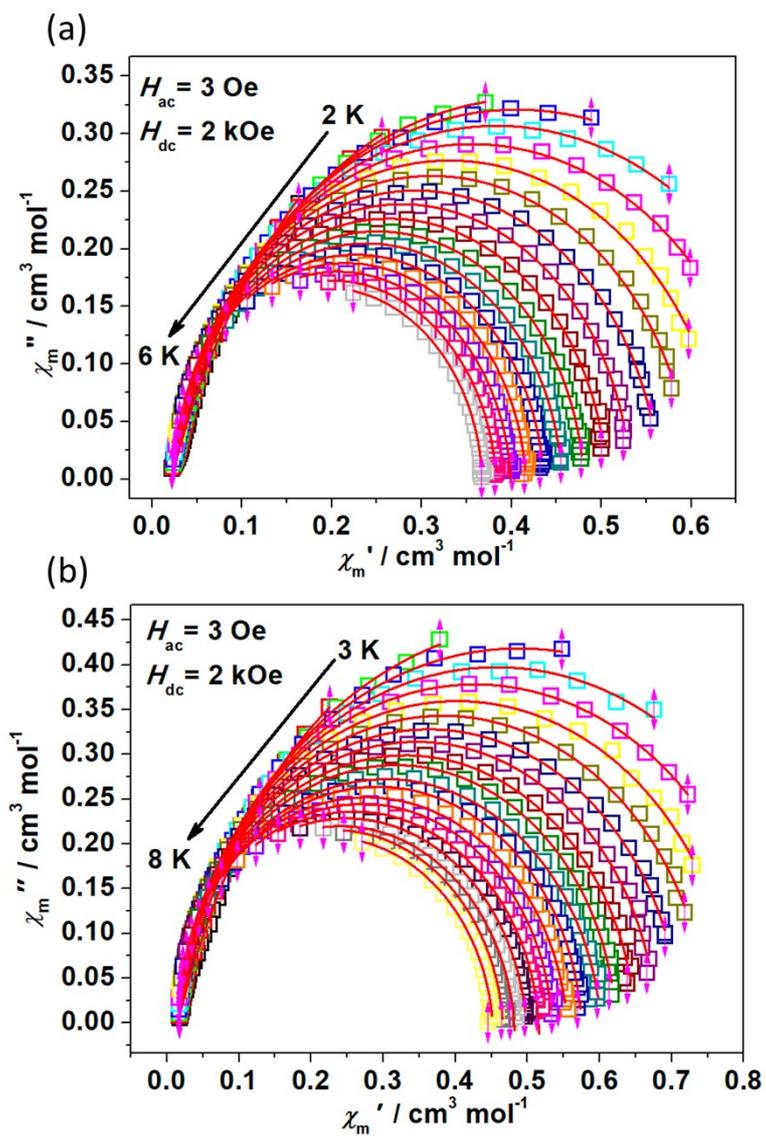
Figure S4. Temperature dependence ac susceptibility for **3**.
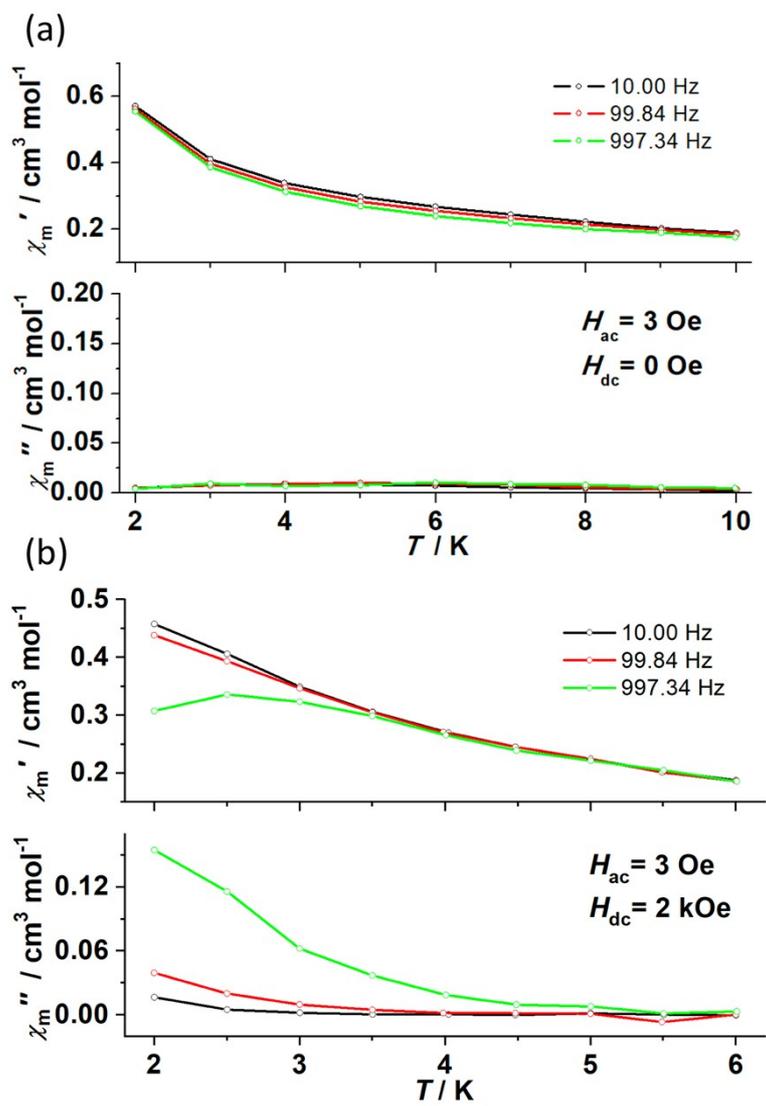
Figure S5. Temperature dependence ac susceptibility for **4**.
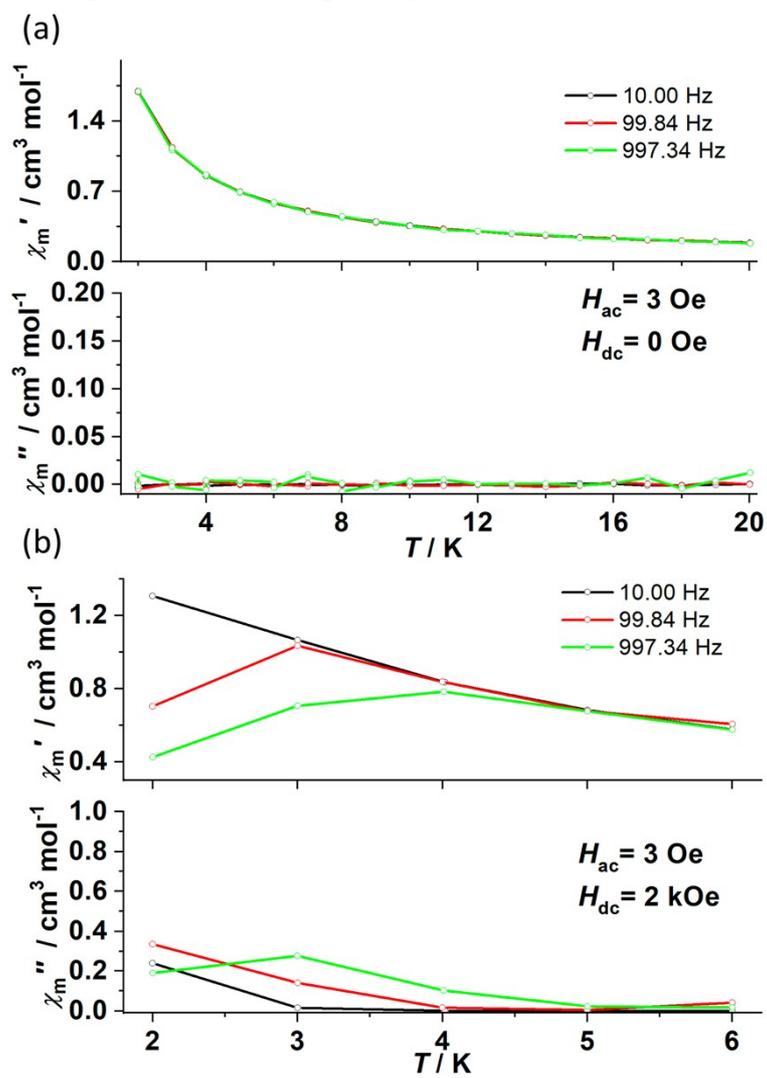
Figure S6. (a) Field dependence of ac susceptibility for **1** at 4 K. (b) Field dependence of relaxation times τ. The solid lines represented the global fitting.
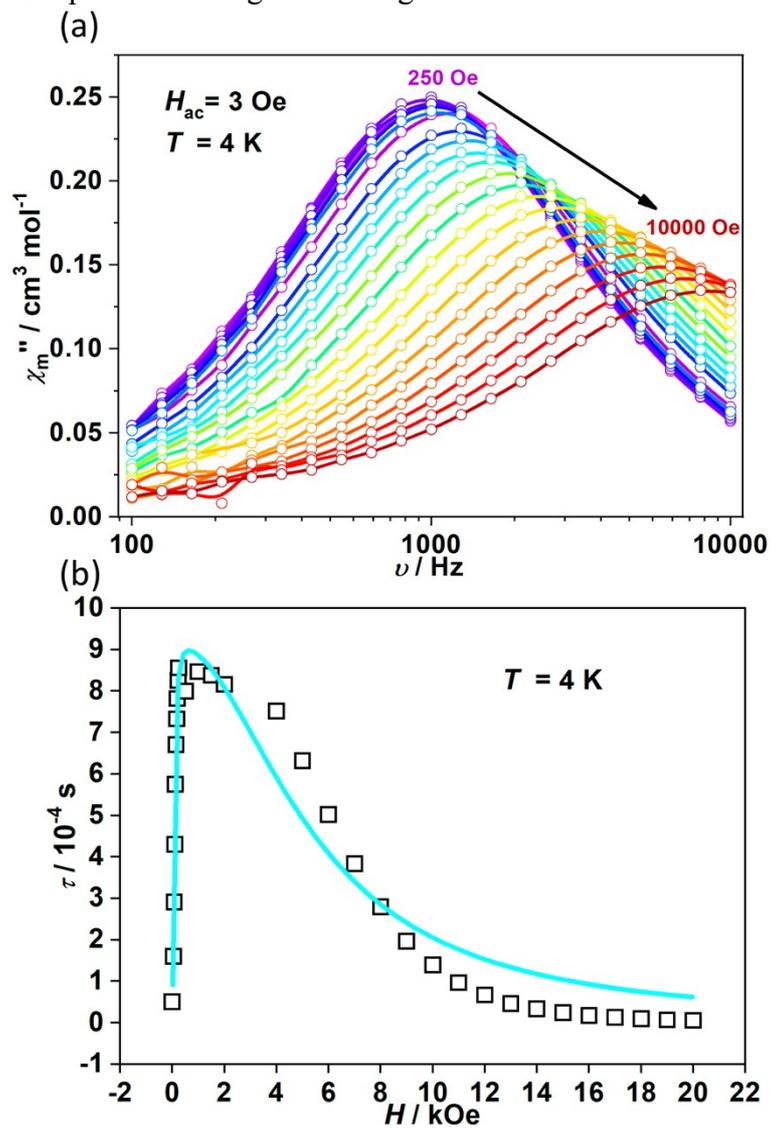
Figure S7. (a) Field dependence of ac susceptibility for **2** at 3 K. (b) Field dependence of relaxation times τ. The solid lines represented the global fitting
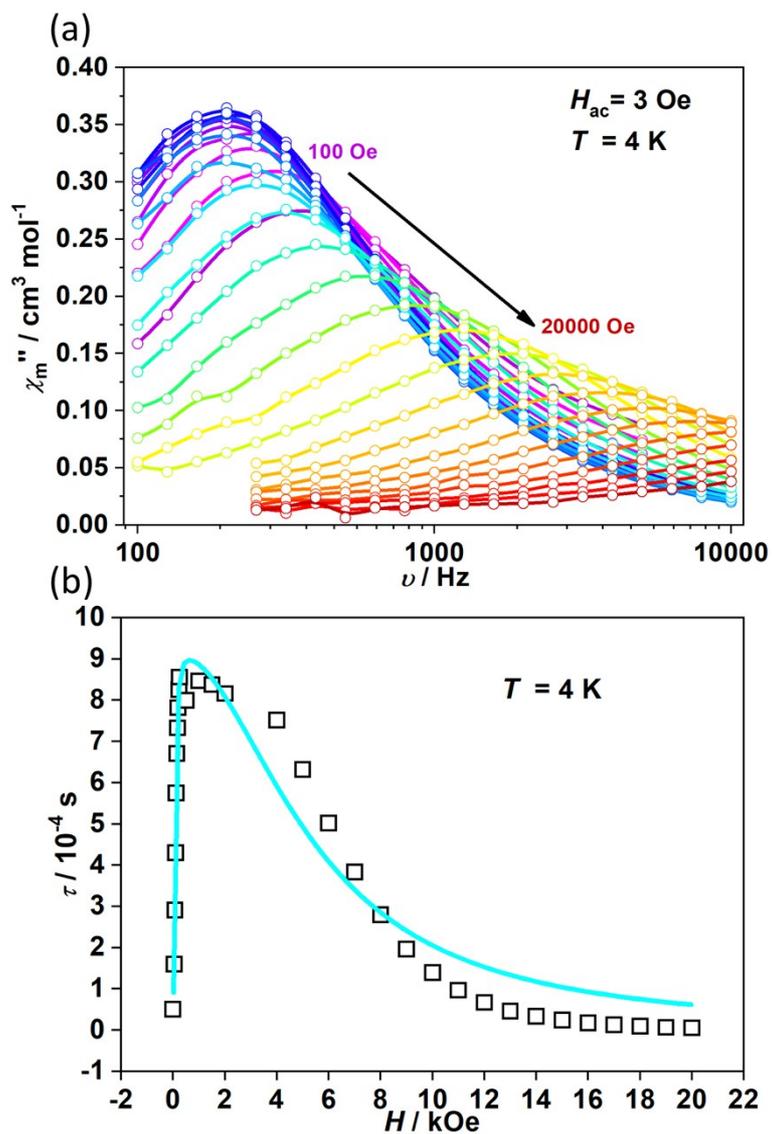
Figure S8. (a) Field dependence of ac susceptibility for **2** at 5 K. (b) Field dependence of relaxation times τ. The solid lines represented the global fitting
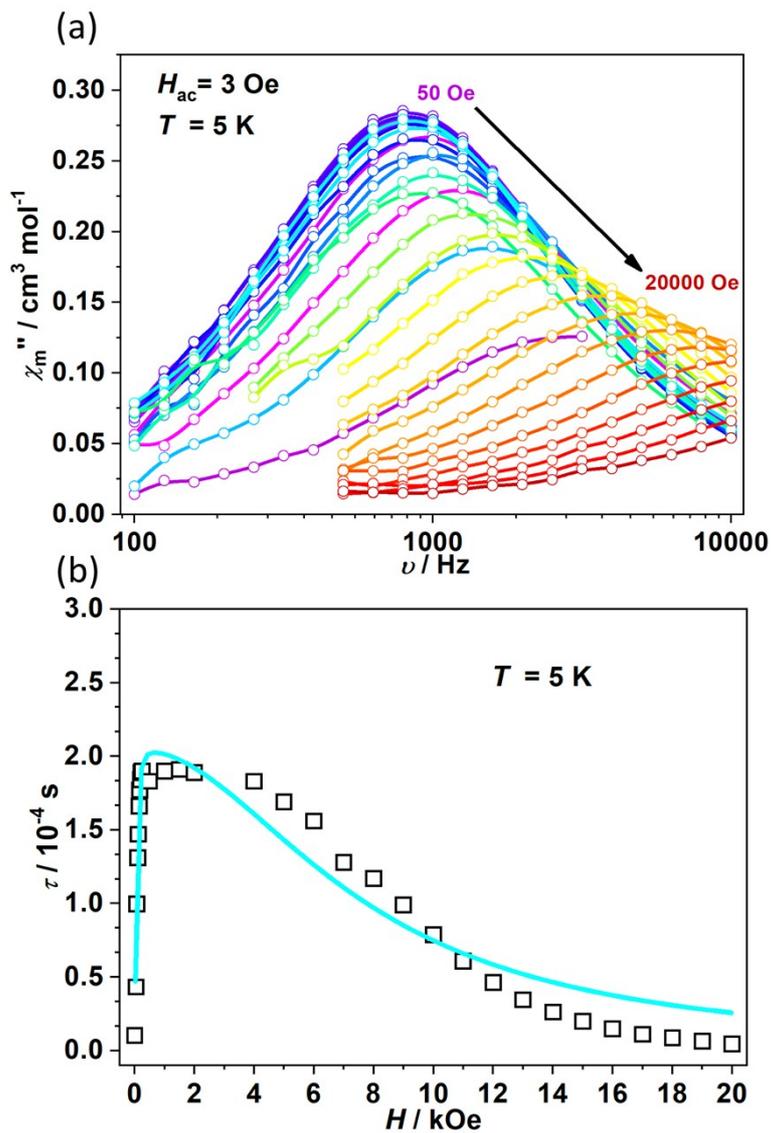
Figure S9. Calculated $\chi_m T$ vs. T plots under 1 kOe dc field