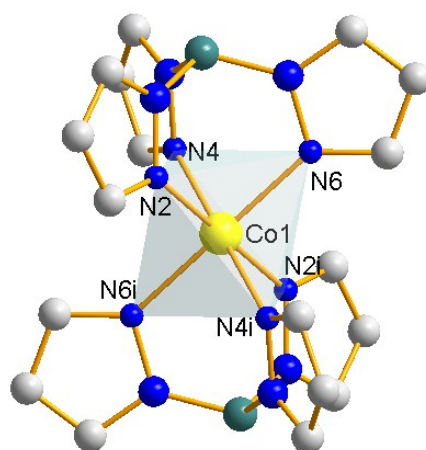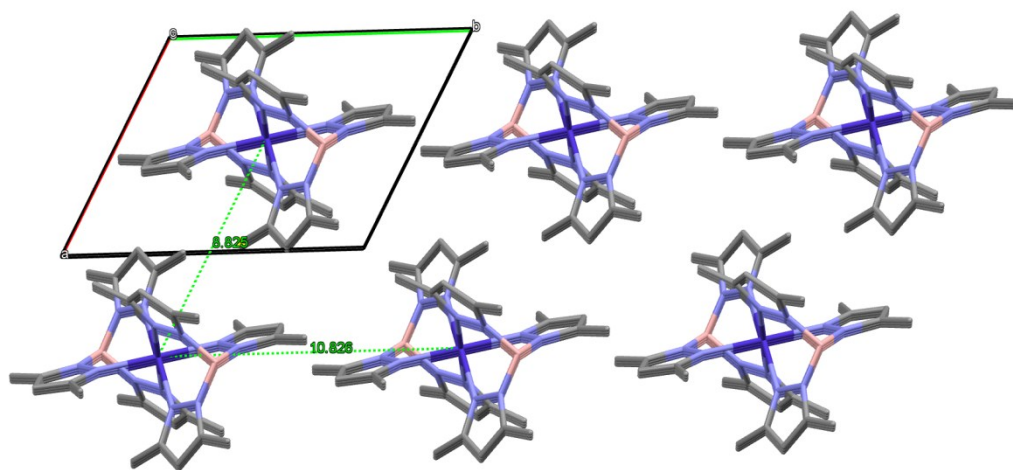# Electronic Supplementary Information

Two Field-Induced Slow Magnetic Relaxation Processes in a Mononuclear Co(II) Complex with Trigonal Anti-prism Geometry

Jing Li,[a] Yuan Han,[a] Fan Cao,[a] Rong-Min Wei,[a] Yi-Quan Zhang*[b] and You Song*[a]

(a)

(b)
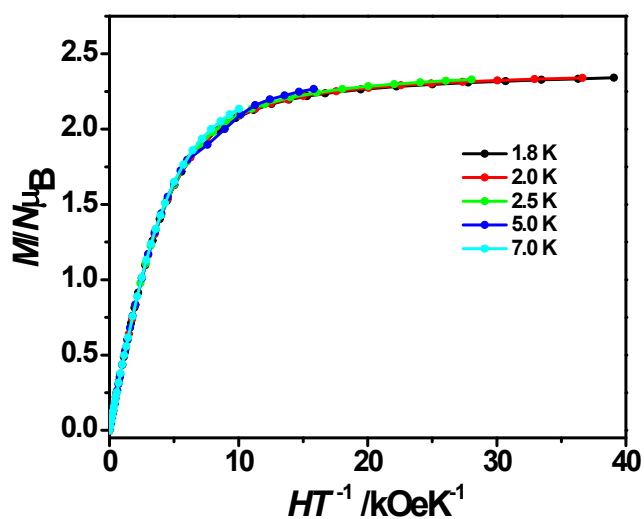
Fig. S1 (a) Structures of complex **1**. Hydrogen atoms and methyl group in 3,5-dimethylpyrazole have been omitted for clarity. (b) The molecular packing of **1** in 1.5×2.5×2.5 at the *c* orientation, which demonstrates the arrangements of anisotropic axis of Co^{II} are in a same orientations. The shortest distance of paramagnetic Co^{II} ions between neighbor clusters are 8.825 Å.

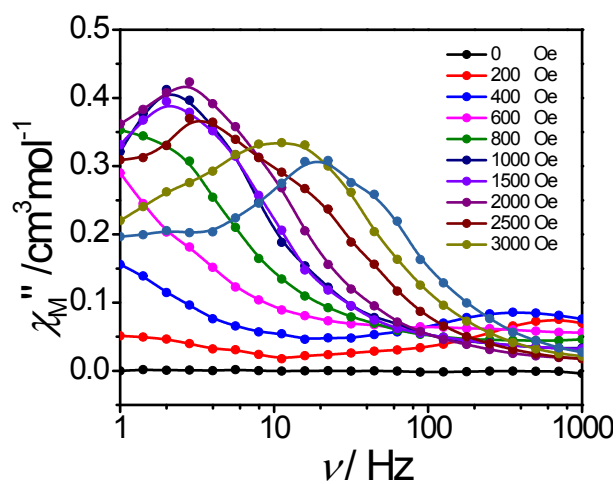**Table S1**. Selected distance [Å] and angle [º] for **1**.

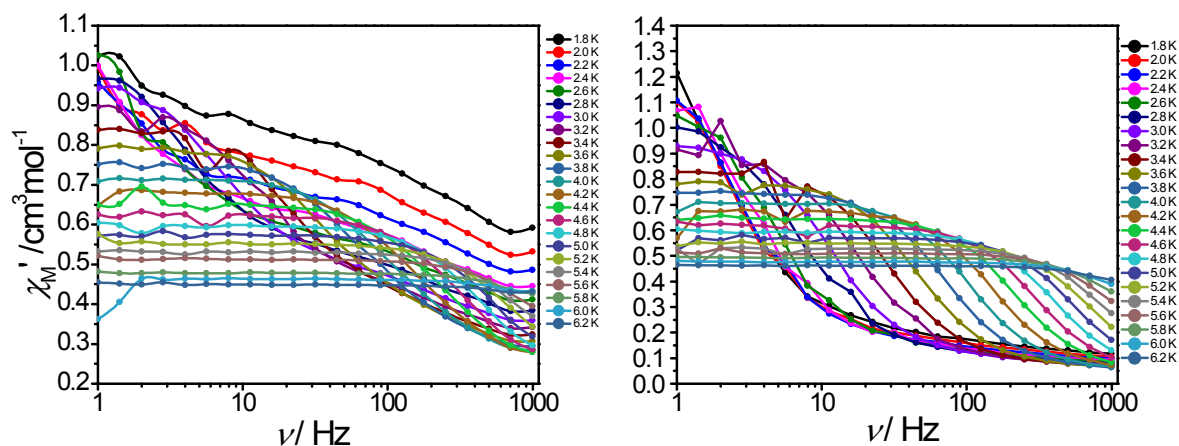| Co1-N2	2.146	Co1-N4	2.151	Co1-N6	2.133
N6-Co1-N6i	180.0	N6-Co1-N2	86.61(7)	N6-Co1-N2i	93.39(7)
N2-Co1-N2i	180.0	N6-Co1-N4i	93.51(3)	N6-Co1-N4	84.49(2)
N2-Co1-N4	87.51(7)	N2-Co1-N4i	92.49(1)	N4-Co1-N4i	180.0

Symmetry code: (i) -x, -y, -z.

Fig. S2 Experimental M vs H/T plots at different temperatures for complex **1**.

Fig. S3 Variable-frequency out-of-phase χ_M'' components of the ac magnetic susceptibility collected for a microcrystalline sample of **1** (at 1.8 K) under different applied dc fields.
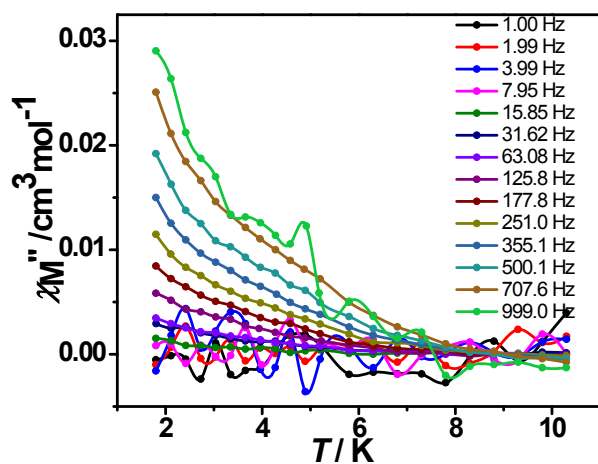
Fig. S4 Frequency dependence of in-phase (χ_M') ac susceptibilities under 400 Oe (left) and 1 kOe (right) dc field (1-1000 Hz, by MPMS VSM) at indicated temperatures for complex **1**, respectively.

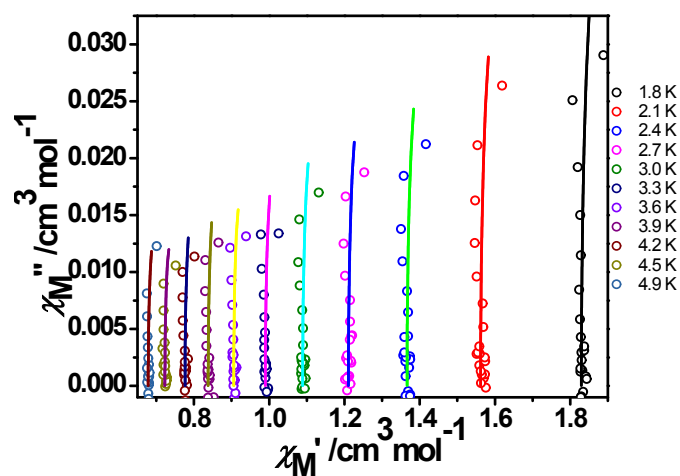**Table S2**. Analysis of Cole-Cole plot of complex **1** under 400 Oe dc field.

T/K	χ_S	$\Delta\chi_1$	τ_1	α_1	$\Delta\chi_2$	τ_2	α_2	R
1.8	0.4979	0.3458	0.6399E-3	0.2795	0.8644	0.4813	0.2503	0.3183E-2
2.0	0.4324	0.3511	0.6523E-3	0.3398	0.7147	0.3241	0.1690	0.2988E-2
2.2	0.4111	0.2873	0.6400E-3	0.2721	0.6363	0.1992	0.1575	0.1083E-2
2.4	0.3681	0.2956	0.6876E-3	0.3316	0.5334	0.1195	0.0741	0.4404E-3
2.6	0.3097	0.3570	0.8116E-3	0.4553	0.4521	0.0784	0	0.4405E-2
2.8	0.3050	0.3007	0.7972E-3	0.3916	0.4071	0.0407	0	0.6984E-3
3.0	0.2930	0.2819	0.8661E-3	0.3653	0.3787	0.0236	0	0.5348E-3
3.2	0.2673	0.3082	0.1022E-2	0.4111	0.3198	0.0132	0	0.2365E-2
3.4	0.2576	0.2986	0.1037E-2	0.3711	0.2856	0.0079	0	0.5105E-2
3.6	0.2542	0.2853	0.9770E-3	0.2895	0.2598	0.0051	0	0.3879E-3
3.8	0.2536	0.2377	0.7213E-3	0.1754	0.2608	0.0033	0	0.5637E-3
4.0	0.2419	0.2322	0.5861E-3	0.1319	0.2406	0.0022	0	0.2459E-3
4.2	0.2301	0.2061	0.4186E-3	0.0828	0.2420	0.0015	0	0.1526E-2
4.4	0.2085	0.2815	0.4495E-3	0.1709	0.1662	0.9115E-3	0	0.2159E-2
4.6	0.1960	0.2106	0.2693E-3	0.1599	0.2158	0.6219E-3	0	0.9635E-3
4.8	0.1831	0.1872	0.1807E-3	0.1307	0.2255	0.4617E-3	0	0.5259E-3
5.0	0.1130	0.2209	0.7201E-4	0.2326	0.2405	0.3274E-3	0	0.2519E-3
5.2	0.0654	0.2798	0.5098E-4	0.2499	0.2100	0.2494E-3	0	0.5950E-3
5.4	0.0797	0.2884	0.6029E-4	0.1077	0.1628	0.2261E-3	0	0.1551E-3
5.6	0.0923	0.3273	0.6744E-4	0.0329	0.0929	0.2431E-3	0	0.1706E-3
5.8	0.0009	0.3706	0.4896E-4	0.0990	0.1072	0.3515E-4	0	0.1172E-3
6.0	0	0.3487	0.4098E-4	0	0.1049	0.4099E-4	0	0.1272E-1
6.2	0	0.2765	0.4875E-4	0	0.1722	0.1141E-4	0	0.2970E-3

Table S3. Analysis of Cole-Cole plot of complex **1** under 400 Oe dc field.

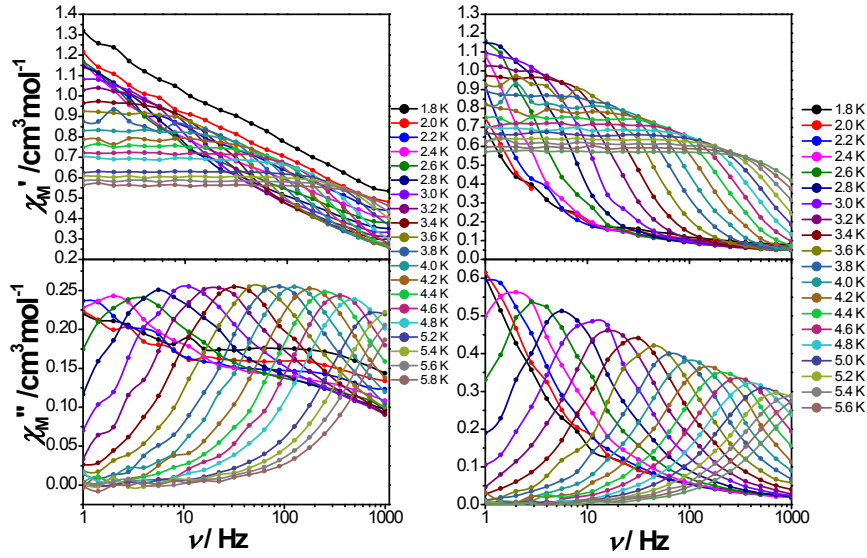| T /K	χ_S	χ_t	τ	α	R
1.8	0.1430	1.7128	0.0928	0.1983	0.0115
2.0	0.1264	1.5972	0.0857	0.2103	0.0106
2.2	0.1213	1.3844	0.0626	0.1637	0.0117
2.4	0.1183	1.2311	0.0450	0.1223	0.0180
2.6	0.1079	1.1229	0.0329	0.0983	0.0121
2.8	0.0931	1.0484	0.0217	0.1112	0.0058
3.0	0.0857	0.9521	0.0135	0.0755	0.0022
3.2	0.0771	0.9300	0.0087	0.0849	0.0213
3.4	0.0689	0.8284	0.0052	0.0579	0.0268
3.6	0.0658	0.7849	0.0034	0.0414	0.0049
3.8	0.0599	0.7480	0.0022	0.0371	0.0006
4.0	0.0595	0.7060	0.0015	0.0201	0.0015
4.2	0.0567	0.6630	0.0010	0.0058	0.0120
4.4	0.0506	0.6474	0.0007	0.0213	0.0003
4.6	0.0477	0.6166	0.0005	0.0153	0.0023
4.8	0.0434	0.5943	0.0004	0.0197	0.0002
5.0	0.0485	0.5651	0.0003	0.0018	0.0026
5.2	0.0373	0.5488	0.0002	0.0201	0.0003
5.4	0.0459	0.5240	0.1701E-3	0	0.0002
5.6	0.0505	0.5101	0.1136E-3	0	0.0002
5.8	0.0418	0.4920	0.1061E-3	0	0.0002
6.0	0.0284	0.4760	0.8232E-4	0	0.0001
6.2	0.0083	0.4611	0.6367E-4	0	0.0001

Fig. S5 Temperature dependence of the out-of-phase (χ″) ac susceptibility under zero dc field for complex **1** with 10 times magnetic site dilution.
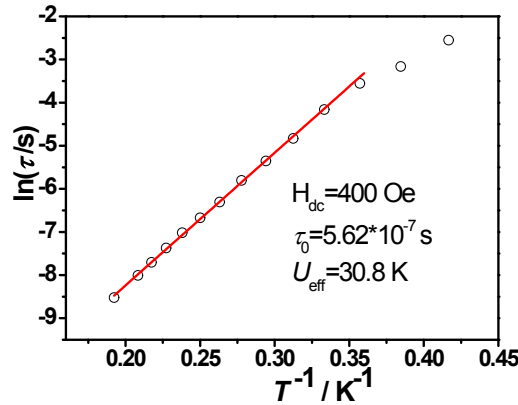

Fig. S6 Variable temperature Cole-Cole plots of Complex **1** with 10 times magnetic site dilution under zero dc field (1-1000 Hz, by MPMS VSM). Fitted parameters are compiled in supplementary Table S3.

Table S4. Analysis of Cole-Cole plot of complex **1** with 10 times magnetic site dilution under zero dc field.
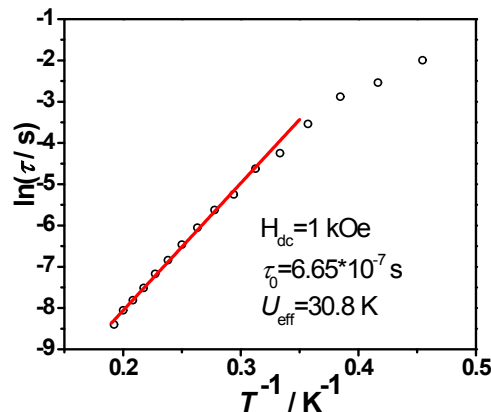
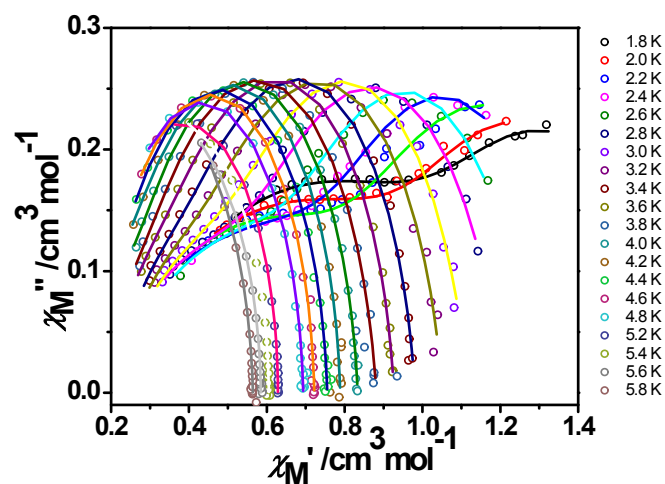| T /K	χ_S	χ_t	τ	α	R
1.8	1.9018	1.8297	0.1019E-3	0.2048E-14	0.3706E-2
2.1	1.6228	1.5619	0.1145E-3	0.2793E-14	0.3818E-2
2.4	1.4178	1.3664	0.1135E-3	0.4366E-14	0.3117E-2
2.7	1.2555	1.2106	0.1166E-3	0.5652E-14	0.2020E-2
3.0	1.1297	1.0890	0.1184E-3	0.7379E-14	0.1849E-2
3.3	1.0249	0.9896	0.1133E-3	0.9292E-14	0.1356E-2
3.6	0.9378	0.9056	0.1198E-3	0.1205E-13	0.1022E-2
3.9	0.8691	0.8378	0.1041E-3	0.1567E-13	0.1007E-2
4.2	0.8043	0.7762	0.1073E-3	0.1567E-13	0.7180E-3
4.5	0.7470	0.7228	0.1368E-3	0.1532E-13	0.7741E-3
4.9	0.7032	0.6784	0.1175E-3	0.2068E-13	0.4318E-3

Fig. S7 Frequency dependence of the in-phase (χ') and out-of-phase (χ'') ac susceptibility under 400 Oe (left) and 1000 Oe (right) dc field for complex **1** with 10 times magnetic site dilution.

Fig. S8 Arrhenius plots of $\ln(\tau)$ vs the inverse temperature T^{-1}, calculated from data at dc field of 400 Oe for complex **1** with 10 times magnetic site dilution. Red lines show fit of the data to the Arrhenius expression $\tau = \tau_0 \exp(U_{eff}/kT)$.
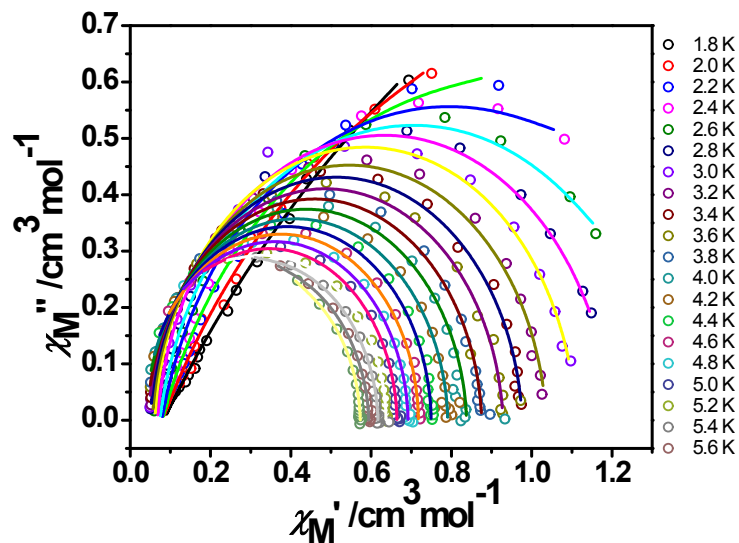

Fig. S9 Arrhenius plots of $\ln(\tau)$ vs the inverse temperature T^{-1}, calculated from data at dc field of 1kOe for complex **1** with 10 times magnetic site dilution. Red lines show fit of the data to the Arrhenius expression $\tau = \tau_0 \exp(U_{eff}/kT)$.

Fig. S10 Variable temperature Cole-Cole plots of Complex **1** with 10 times magnetic site dilution under 400 Oe dc field (1-1000 Hz, by MPMS VSM). Fitted parameters are compiled in supplementary Table S4.

Table S5. Analysis of Cole-Cole plot of complex **1** with 10 times magnetic site dilution under 400 Oe dc field.

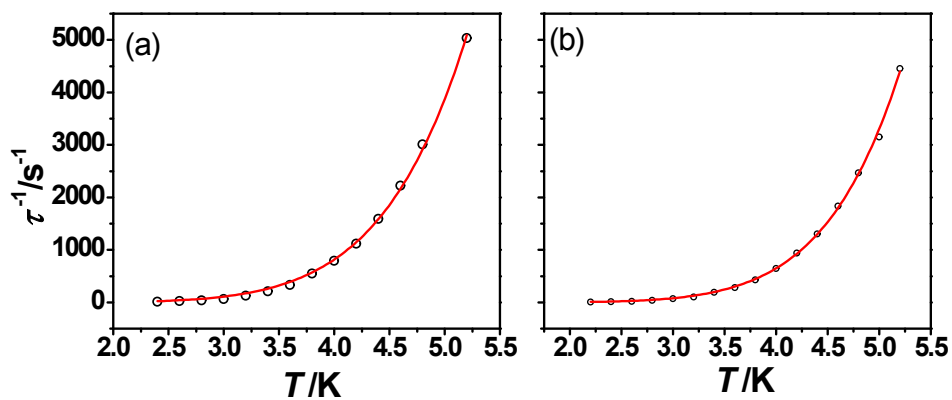| T/K	χ_S	$\Delta\chi_1$	τ_1	α_1	$\Delta\chi_2$	τ_2	α_2	R
1.8	0.2089	0.9794	0.8966E-3	0.5936	0.5043	0.1953	0.2894	0.1234E-2
2.0	0.1639	0.9759	0.1111E-2	0.6219	0.4826	0.2480	0.2439	0.1096E-2
2.2	0.1700	0.7934	0.8279E-3	0.5897	0.5855	0.1940	0.2613	0.2298E-2
2.4	0.1363	0.8822	0.1533E-2	0.6386	0.4107	0.1055	0.1221	0.5303E-3
2.6	0.1886	0.6814	0.1415E-2	0.5671	0.4267	0.0578	0.0987	0.1004E-2
2.8	0.1817	0.54408	0.8561E-3	0.5247	0.4878	0.0308	0.1314	0.7076E-3
3.0	0.1925	0.5372	0.1259E-2	0.5029	0.4017	0.0171	0.0568	0.7440E-3
3.2	0.2039	0.3500	0.6121E-3	0.3926	0.4991	0.0095	0.1054	0.1060E-2
3.4	0.2034	0.2821	0.4704E-3	0.3281	0.4958	0.0055	0.0807	0.9081E-3
3.6	0.1842	0.2417	0.3350E-3	0.3023	0.5014	0.0033	0.0790	0.3564E-3
3.8	0.1457	0.2348	0.1952E-3	0.3975	0.5021	0.0020	0.0786	0.5811E-2
4.0	0.1347	0.1515	0.8007E-4	0.2183	0.5474	0.0013	0.0820	0.3637E-3
4.2	0.1509	0.1244	0.1076E-3	0.1168	0.5131	0.0009	0.0535	0.1140E-2
4.4	0	0.2419	0.1680E-4	0.2614	0.5135	0.0006	0.0568	0.3975E-3
4.6	0	0.2179	0.2151E-4	0	0.5039	0.0005	0.0440	0.2177E-3
4.8	0	0.2032	0.1845E-4	0	0.4902	0.0004	0.0464	0.5122E-3
5.2	0	0.2729	0.3754E-4	0	0.3557	0.0003	0.0101	0.1381E-3
5.4	0	0.2859	0.4093E-4	0	0.3191	0.0002	0.0179	0.2298E-3
5.6	0	0.3215	0.4244E-4	0	0.2630	0.0002	0.0302	0.2186E-3
5.8	0	0.3049	0.4239E-4	0	0.2591	0.0001	0.00590	0.2969E-3

Fig. S11 Variable temperature Cole-Cole plots of Complex **1** with 10 times magnetic site dilution under 1000 Oe dc field(1-1000 Hz, by MPMS VSM). Fitted parameters are compiled in supplementary Table S5.

Table S6. Analysis of Cole-Cole plot of complex **1** with 10 times magnetic site dilution under 1 kOe dc field.

T /K	χ_S	χ_t	τ	α	R
1.8	0.0832	3.9538	1.4851	0.3777	0.5762E-2
2.0	0.0798	2.5472	0.4792	0.3089	0.5633E-2
2.2	0.0794	1.8449	0.1894	0.2290	0.9471E-2
2.4	0.0782	1.5123	0.0911	0.1528	0.7464E-2
2.6	0.0732	1.3379	0.0486	0.1117	0.08132E-2
2.8	0.0677	1.1985	0.0261	0.0614	0.6362E-2
3.0	0.0612	1.1101	0.0154	0.0399	0.6516E-2
3.2	0.0552	1.0356	0.0090	0.0401	0.2253E-2
3.4	0.0500	0.9759	0.0055	0.0346	0.1937E-2
3.6	0.0446	0.9284	0.0035	0.0360	0.9038E-2
3.8	0.0422	0.8754	0.0022	0.0269	0.1188E-2
4.0	0.0375	0.8378	0.0015	0.0313	0.1183E-2
4.2	0.0368	0.7909	0.0010	0.0226	0.3349E-2
4.4	0.0363	0.7493	0.0007	0.0120	0.1625E-2
4.6	0.0357	0.7174	0.0005	0.0088	0.8178E-3
4.8	0.0280	0.6919	0.0004	0.0184	0.6047E-3
5.0	0.0287	0.6642	0.0003	0.0161	0.4523E-3
5.2	0.0299	0.6371	0.0002	0.0121	0.4039E-3
5.4	0.0097	0.6174	0.0001	0.0322	0.1166E-2
5.6	0	0.5942	0.0001	0.0322	0.4126E-3

Fig. S12 τ^{-1} vs temperature T, calculated from data at dc field of 400 Oe (a) and 1 kOe (b) for complex **1** with 10 times magnetic site dilution. Red lines show fit of the data to eq. **2**.

Table S7. Calculated spin-free energies (cm^{-1}) of the lowest terms of complex **1**.

	Spin mult	4	4	4	4	4	4	4	4	4	4
1	*E*	0.0	32.0	1995.4	9540.2	9678.7	10603.3	21638.8	22053.2	22255.7	22844.0

Table S8. Calculated spin-orbit energies (cm^{-1}) and **g** tensors (x, y, z) of the lowest Kramers doublets of complex **1**.

		1	2	3	4	5	6
1	*E*	0.0	217.6	467.9	763.2	2364.1	2450.6
	g	1.015	0.146	0.806	0.124	0.258	3.857
		1.041	1.075	0.634	0.161	0.275	3.349
		8.685	4.583	0.330	3.201	6.407	2.121