Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Inorganic-organic Hybrid Switchable Dielectric Material with the Coexistence of Magnetic Anomalies Induced by Reversible High-Temperature Phase Transition.

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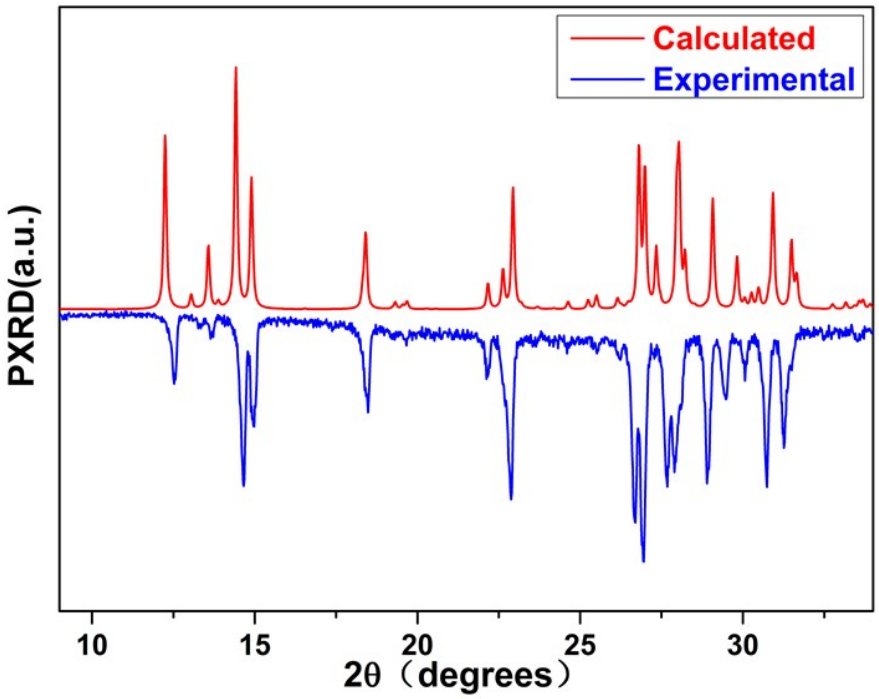

Figure S1. Experimental and calculated powder X-ray diffraction patterns of **1** at room temperature.

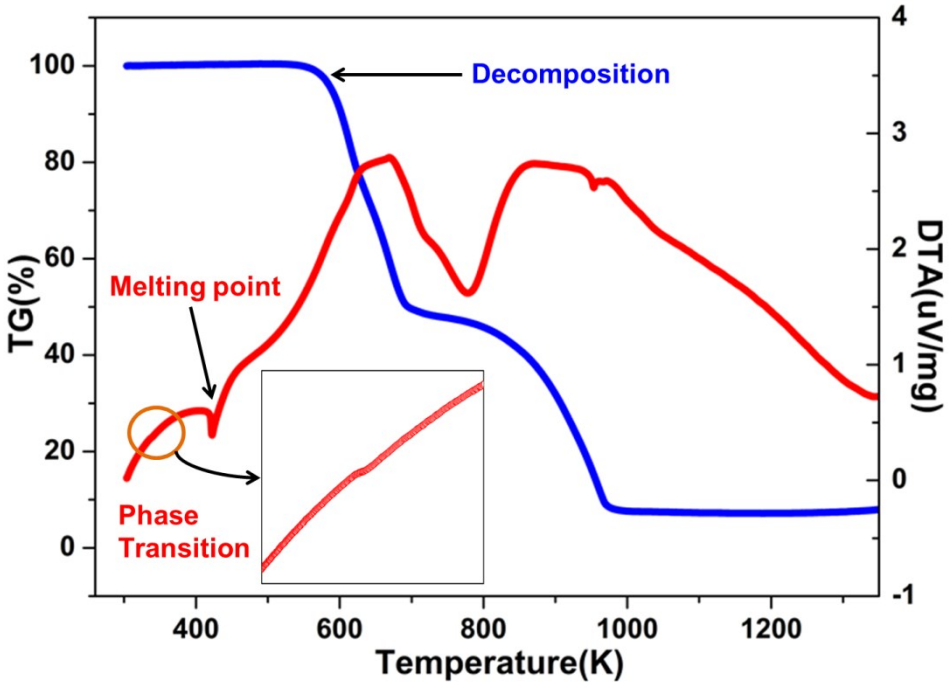

Figure S2. TG-DTA curves of **1** in the heating mode.

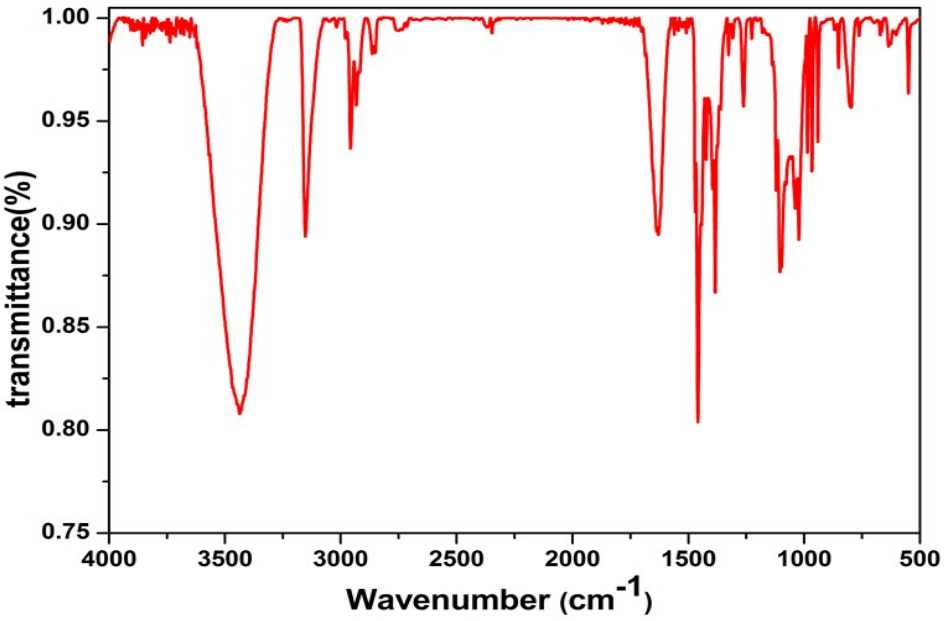

Figure S3. IR spectra of **1** in KBr pellets were recorded on a Shimadzu model IR-60 spectrometer at room temperature.

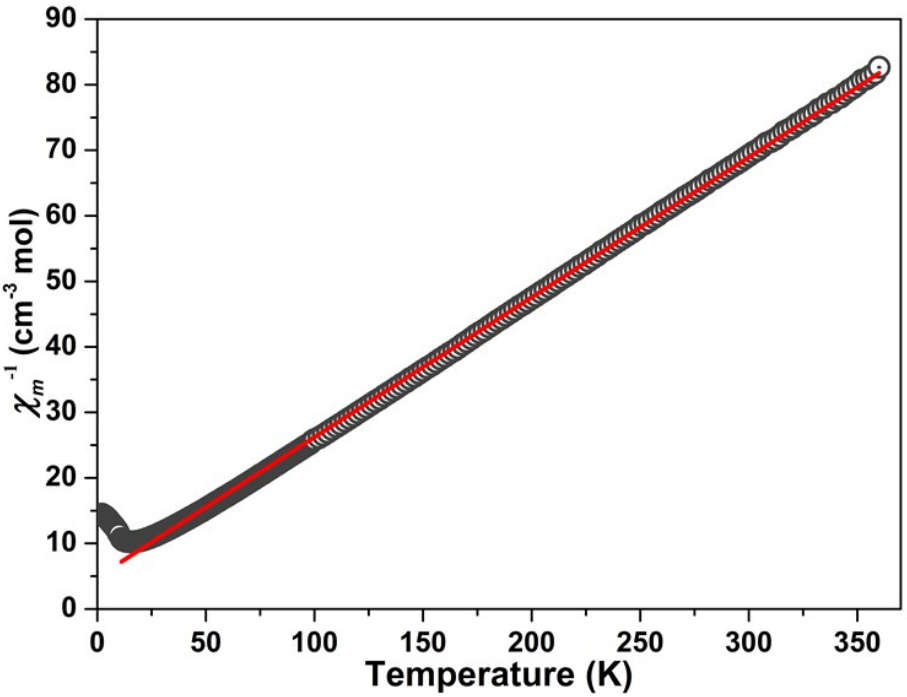

Figure S4. Temperature dependences of χ_m^{-1} of **1** measured in the temperature range of 2-360 K. The red solid line is fitted by the Curie-Weiss law.

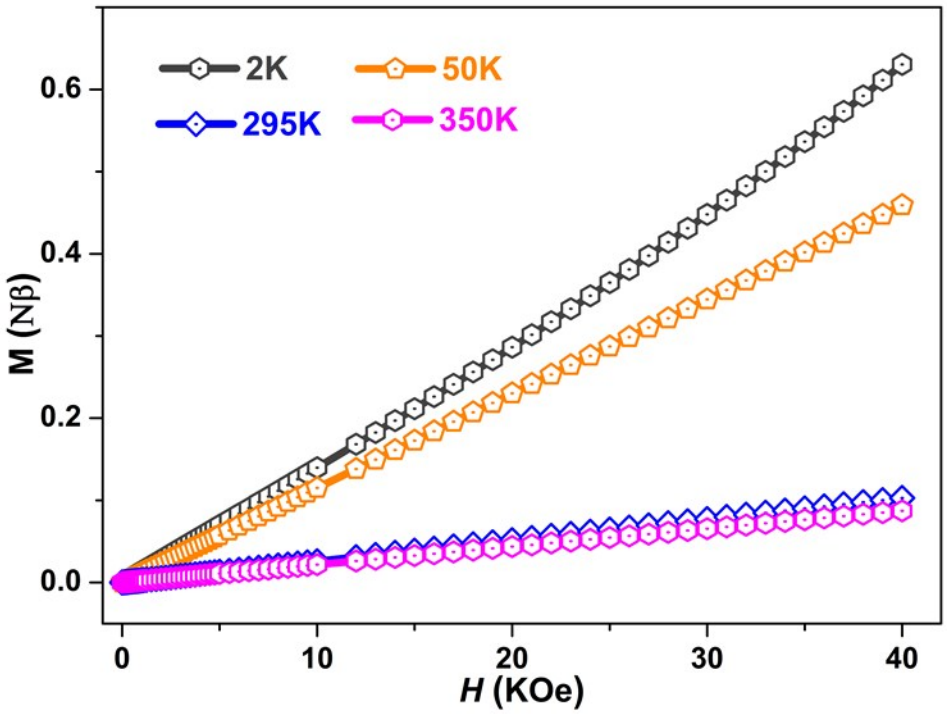

Figure S5. Plots of field-dependent isothermal magnetizations of **1** measured at 2 K, 50 K, 295 K and 350 K.

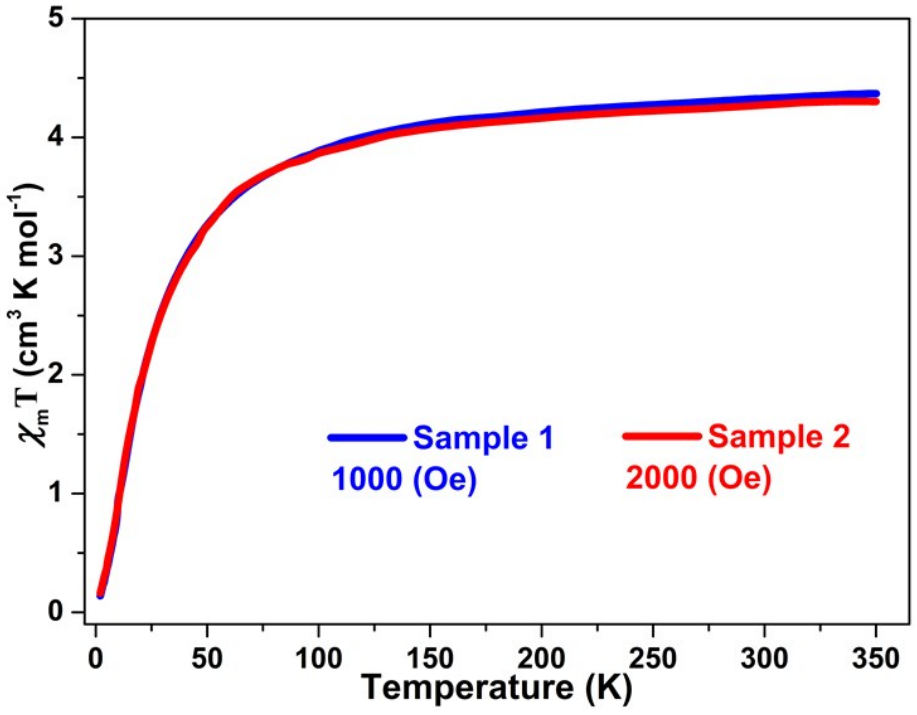

Figure S6. Temperature-dependence of $\chi_m T$ for **1** measured adopting two samples under different applied direct-current fields (sample **1** for 1000 Oe and sample **2** for 2000 Oe).

Table S1. Crystal data and structure refinement for compound 1.

Empirical formula	$C_6H_{14}NFeBr_4$	$C_6H_{14}NFeBr_4$
Temperature	100 K, below T_c	345 K, above T_c
Formula weight	475.67	475.67
Crystallographic system,	monoclinic	orthorhombic
Space group	$P2_1/c$	*Cmcm*
Unit cell dimensions	a = 6.5974(1)	a = 9.9932(4)
	b = 13.7002(3)	b = 9.5392(4)
	c = 14.6244(2)	c = 14.8097(5)
	V= 1320.49(4)	V= 1411.77(9)
Z, Calculated density	4, 2.393g/cm^3	4, 2.238g/cm^3
F(000)	892.0	892.0
Theta range for data collection	8.84 – 175.16º	11.952 – 145.404º
Limiting indices	-7<=h<=8	-12<=h<=8
	-17<=k<=17	-11<=k<=6
	-16<=l<=11	-12<=l<=18
Reflections collected / unique	6995 / 2522 [R_{int} = 0.0813]	2189 / 761 [R_{int} = 0.0607]
Completeness	99.4 %	99.3 %
Data / restraints / parameters	2522/0/110	761/50/50
Goodness-of-fit	1.090	1.024
Final R indices [I>2sigma(I)]	R_1 = 0.0889 wR$_2$ =0.2314	R_1 = 0.0540 wR$_2$ =0.1477

Table S2. Fe-Br Bond Lengths of compound 1 at LTP and HTP.

LTP			
Bond	(Å)	Bond	(Å)
Fe(1)-Br(1)	2.5020(15)	Fe(1)-Br(2)	2.2692(15)
Fe(1)-Br(3)	2.3482(16)	Fe(1)-Br(4)	2.2432(14)
HTP			
Bond	(Å)	Bond	(Å)
Fe(1)-Br(1)	2.3333(17)	Fe(1)-Br(2)	2.3096(15)

Table S3. Br-Fe-Br Bond angles of compound 1 at LTP and HTP.

LTP			
Bond Angle	(˚)	Bond Angle	(˚)
Br(1)-Fe(1)-Br(2)	111.80(6)	Br(2)-Fe(1)-Br(3)	110.66(6)
Br(1)-Fe(1)-Br(3)	105.86(6)	Br(2)-Fe(1)-Br(4)	106.80(6)
Br(1)-Fe(1)-Br(4)	113.59(6)	Br(3)-Fe(1)-Br(4)	108.10(6)
HTP			
Bond Angle	(˚)	Bond Angle	(˚)
Br(1)-Fe(1)-Br(1)	106.74(11)	Br(2)-Fe(1)-Br(2)	112.16(11)

Br(1)-Fe(1)-Br(2)	109.44(3)

Table S4. N-H···Br Hydrogen bonds of 1 at LTP.

D-H···A	d(D-H)	d(H···A)	< DHA	d(D..A)
N1-H1···Br1	0.930	2.743	136.35	3.475
N1-H1···Br3	0.930	2.877	135.10	3.596

Table S5. N-H···Br Hydrogen bonds of 1 at HTP.

D-H···A	d(D-H)	d(H···A)	< DHA	d(D..A)
N1-H1A···Br1$^{\#1}$	0.900	3.140	135.80	3.842
N1-H1A···Br1$^{\#2}$	0.900	3.140	135.80	3.842

Symmetry transformations used to generate equivalent atoms:

$^{\#1}$ –X, 1-Y, 1-Z $^{\#2}$ +X, 1-Y, ½+Z