Electronic Supplementary Material (ESI) for RSC Advances.


# A Breakthrough in the Intrinsic Multiferroic Temperature Region in Prussian Blue Analogues

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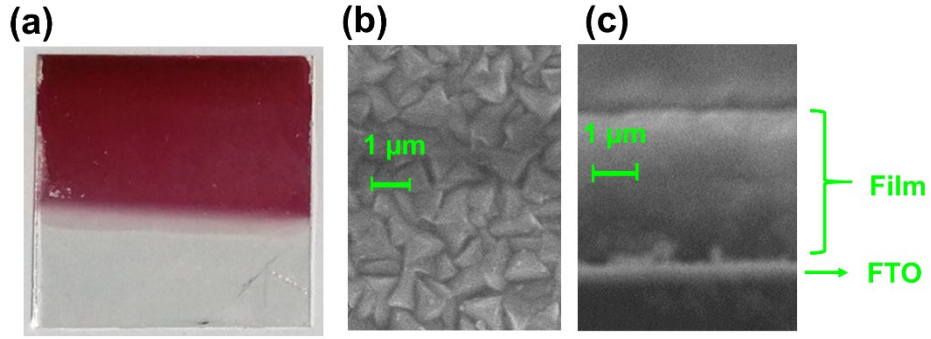


**Figure S1.** (a) The photograph of colored film **1**. (b)The topographic image of the film surface. (c)The thickness for the film **1**.

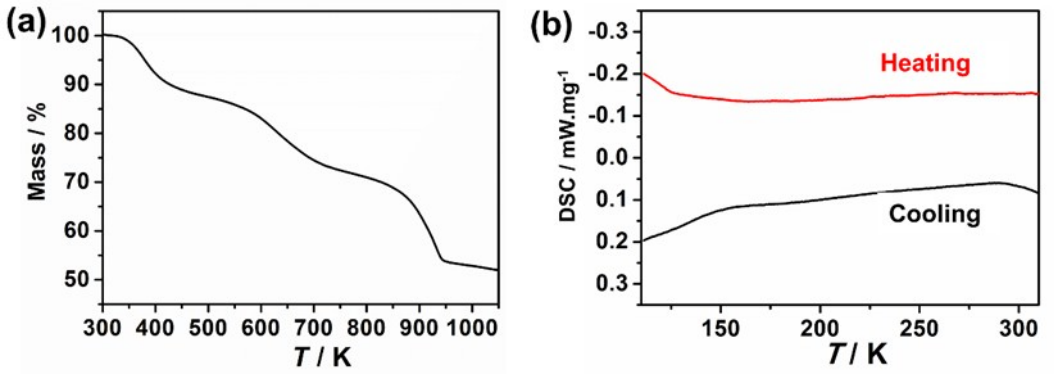


**Figure S2. (a)** TGA curve of crushed film **1**. (b) DSC data of crushed film **1** from 100 K to 310 K.

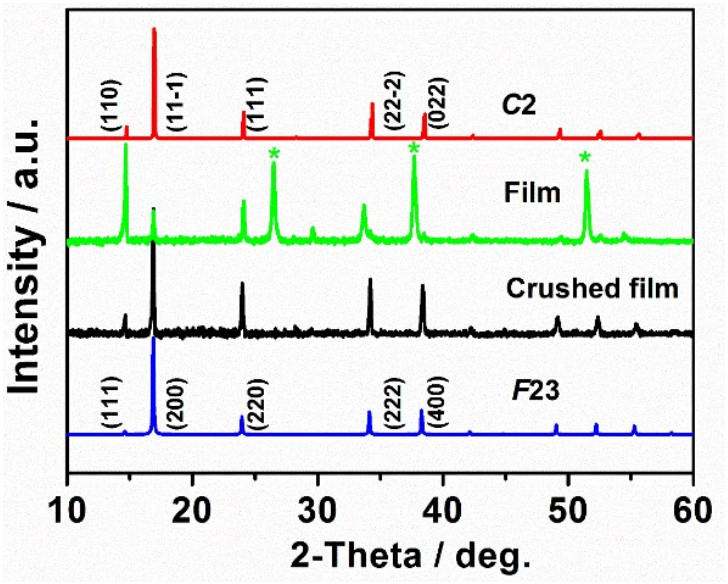


**Figure S3.** The XRD patterns of film, and crushed film for compound **1**. The peaks marked with * is the diffraction peaks of FTO substrate.

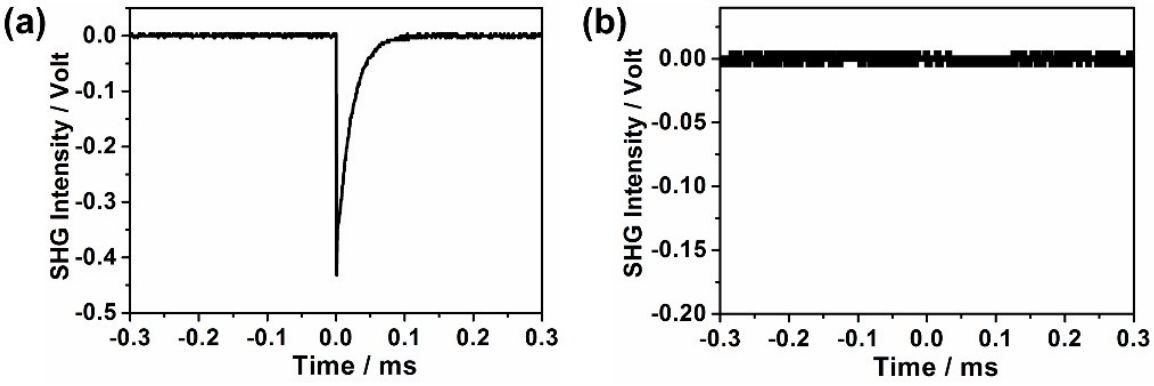


**Figure S4.** (a). The film **1** SHG intensity oscilloscope traces at room temperature. (b)

SHG signals detected for the crushed film **1** at room temperature.

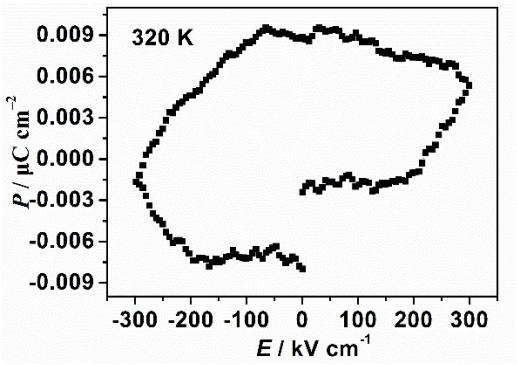


**Figure S5.** Polarization versus electric field curve (*P*–*E* hysteresis loop) at 320 K of film **1**.

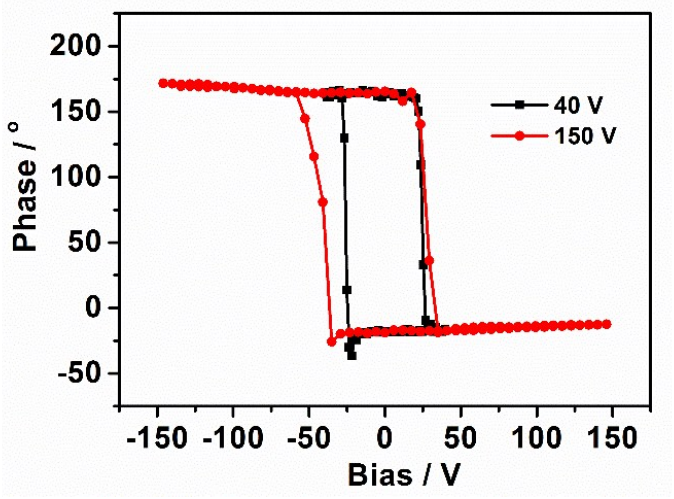


**Figure S6.** PFM phase hysteresis loop at the bias voltage value of 40 V and 150 V of film **1**.

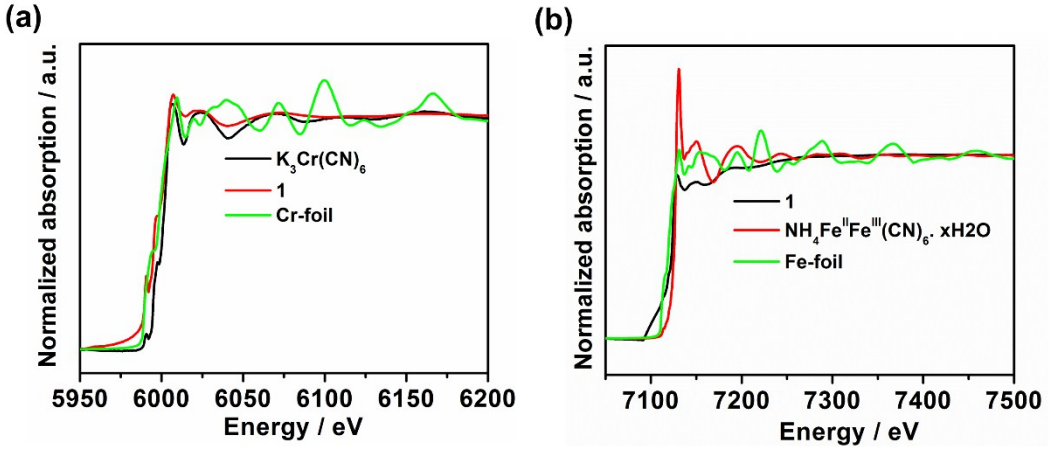


**Figure S7.** The normalized X-ray adsorption spectrum of Cr and Fe samples.

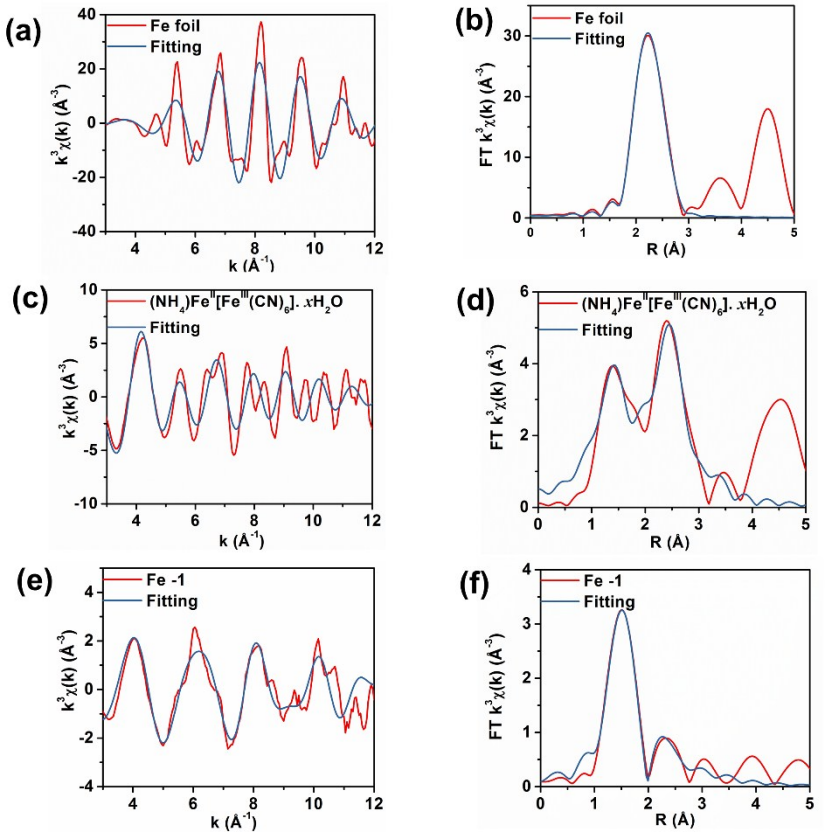


**Figure S8.** The EXAFS fitting results of Fe samples.

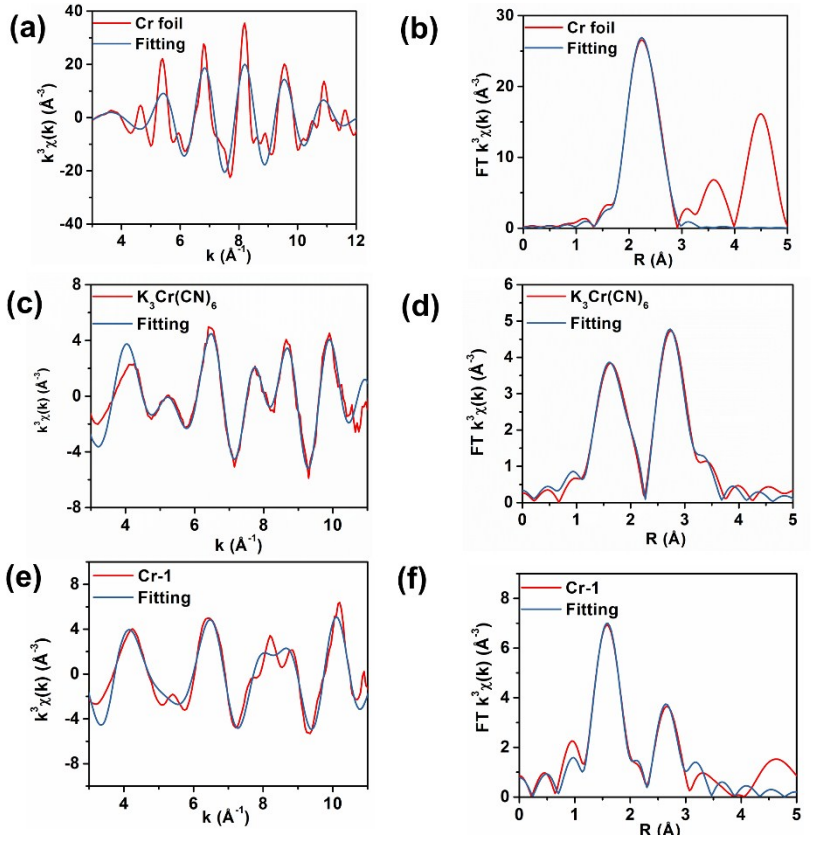

**Figure S9.** The EXAFS fitting results of Cr samples.

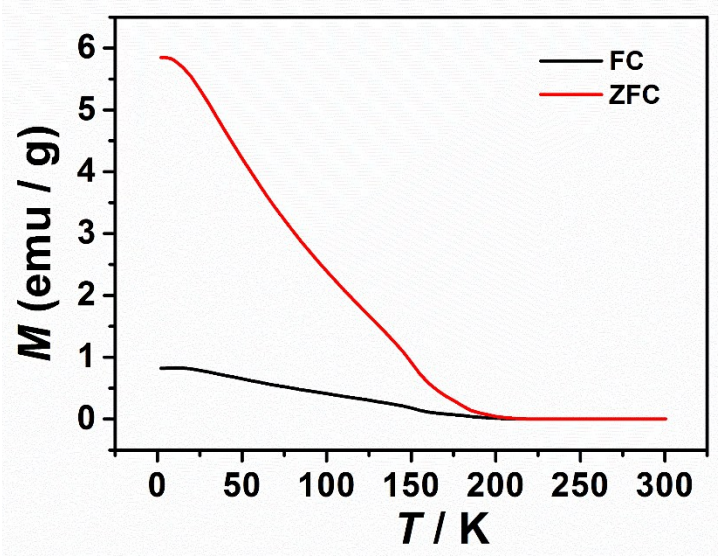


**Figure S10.** ZFC/FC magnetization curves of crushed film **1** under an applied magnetic field of 10 Oe.

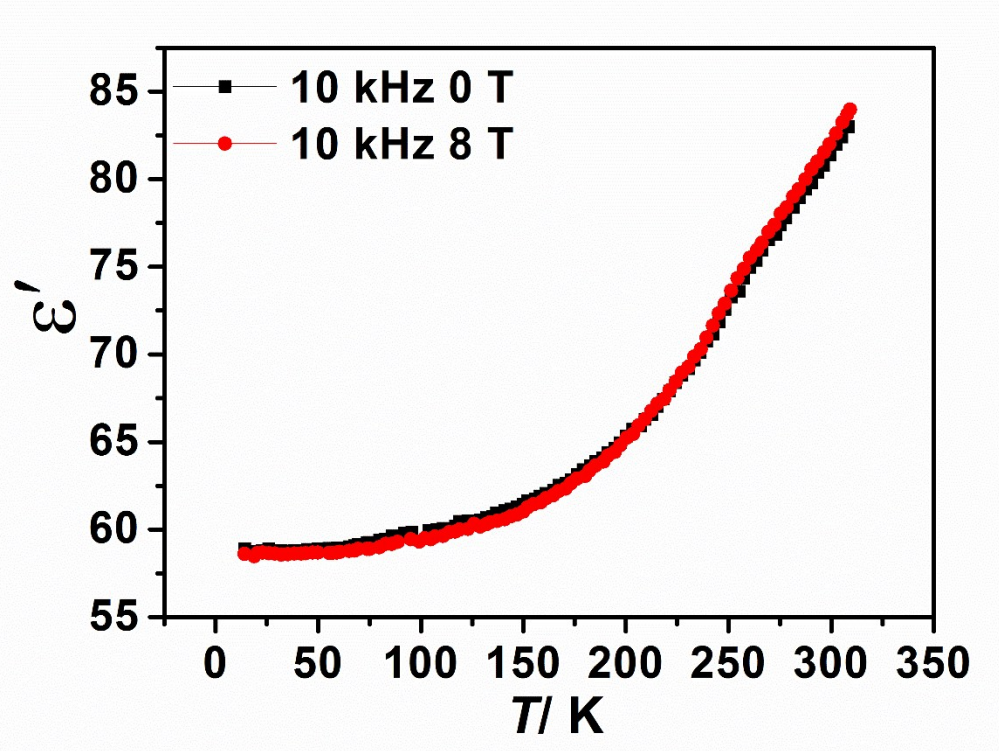


**Figure11.** Dielectric constant versus temperature at 10 kHz measured in zero field and in an external magnetic field of 8 T for film **1**

**Table S1.** The results of ICP-OES of **1**.

| Sample 1 | Fe (μg/L) | Cr (μg/L) | n (Fe) :n (Cr) |
|---|---|---|---|
| Film 1-1 | 164 | 669 | 1 : 4.436 |
| Film 1-2 | 168 | 623 | 1 : 3.980 |
| Film 1-3 | 201 | 751 | 1 : 4.023 |
| Film 1-4 | 253 | 893 | 1 : 3.787 |
| Film 1-5 | 183 | 742 | 1 : 4.368 |
| Film 1-6 | 259 | 920 | 1 : 3.825 |

**Table S2.** EXAFS fitting parameters at the Cr and Fe K-edge various samples（$S_0^2$=0.72, 0.70）

| Sample | Path | C.N. | R (Å) | $\sigma^2\times10^3$ (Å$^2$) | ΔE (eV) | R factor |
|---|---|---|---|---|---|---|
| Cr foil | Cr-Cr | 8* | 2.48±0.01 | 5.2±1.3 | 3.8±1.6 | 0.002 |
| | Cr-Cr | 6* | 2.87±0.01 | 4.0±1.7 | 4.1±2.3 | |
| $K_3Cr(CN)_6$ | Cr-C/N | 4.7±1.0 | 2.05±0.01 | 3.1±1.6 | -3.3±3.6 | 0.005 |
| | Cr-C/N | 7.1±1.5 | 3.25±0.01 | 4.8±1.4 | -1.8±1.8 | |
| Cr - **1** | Cr- C/N | 4.9±1.3 | 2.03±0.02 | 6.7±2.1 | 3.9±3.6 | 0.005 |
| | Cr- C/N | 3.8±2.3 | 3.03±0.03 | 13.2±4.4 | 15.9±4.6 | |
| Fe foil | Fe-Fe | 8* | 2.46±0.01 | 4.7±1.1 | 6.1±1.6 | 0.002 |
| | Fe-Fe | 6* | 2.84±0.01 | 5.0±2.1 | 4.6±2.9 | |
| $(NH_4)Fe[Fe(CN)_6]\cdot xH_2O$ | Fe- C/N | 6.7±2.6 | 1.97±0.03 | 12.0±3.0 | -8.0±2.2 | 0.018 |
| | Fe- C/N | 12.7±3.8 | 3.18±0.02 | 7.5±3.0 | | |
| Fe - **1** | Fe- C/N | 4.3±1.2 | 2.02±0.02 | 6.3±2.5 | -5.5±3.5 | 0.018 |
| | Fe- C/N | 1.7±2.0 | 3.17±0.06 | 10.6±8.7 | -5.9±5.3 | |

[a]*N*: coordination numbers; [b]*R*: bond distance; [c]$\sigma^2$: Debye-Waller factors; [d] $\Delta E_0$: the inner potential correction. *R* factor: goodness of fit. * the experimental EXAFS fit of metal foil by fixing CN as the known crystallographic value.