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Supplementary material for

Magnetoelectric Coupling induced by Jahn-Teller Cu^{2+} in $SrFe_{12}O_{19}$ Ceramics

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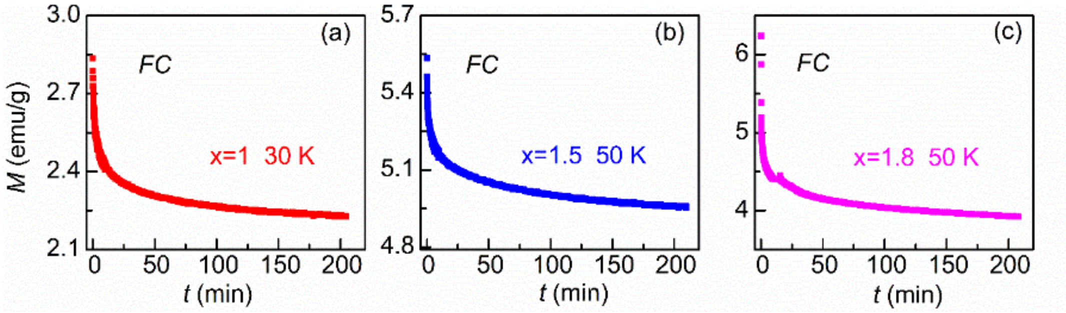


Figure S1 The isothermal remanent magnetization (IRM) curves of $Sr(CuTi)_xFe_{12-2x}O_{19}$ ceramics.

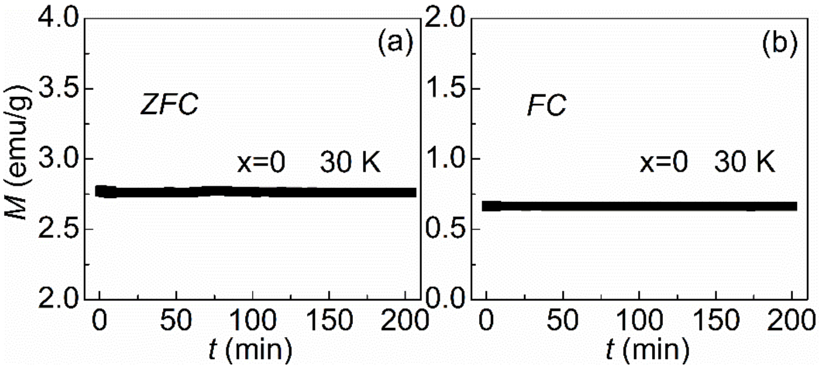


Figure S2 Time dependence of (a) isothermal magnetization measured with 200 Oe after ZFC cooling and (b) remanent magnetization measured under zero magnetic field after FC cooling with 200 Oe for $SrFe_{12}O_{19}$ ceramics.

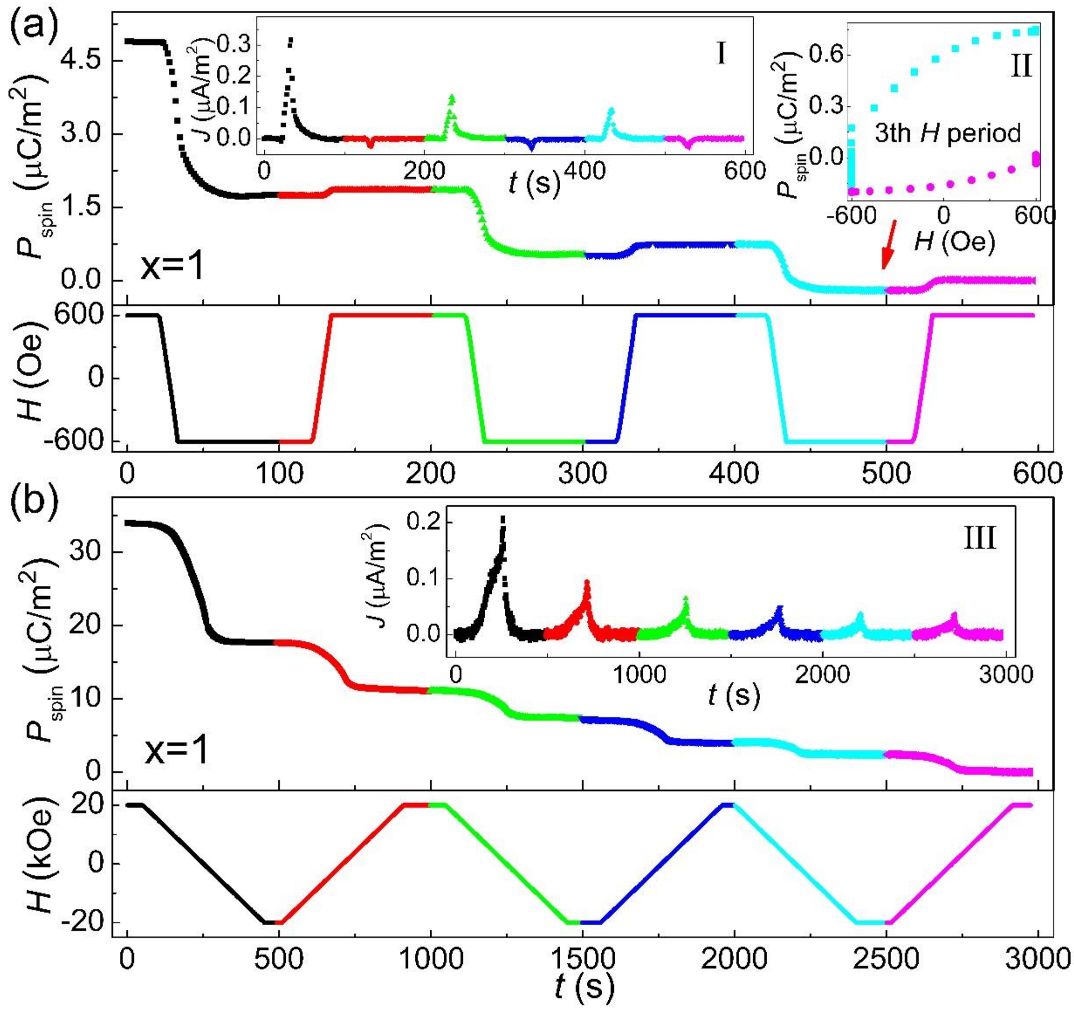


Figure S3. The evolution of P_{spin} with periodic H of the ceramic with x=1 under $H \perp E$ and 10 K, with H equals (a) 600 Oe and (b) 20 kOe. The insets I and III show the corresponding variation of current density J with periodic H. The inset II in (a) is the P_{spin}-H curves under $H \perp E$ and 10 K.

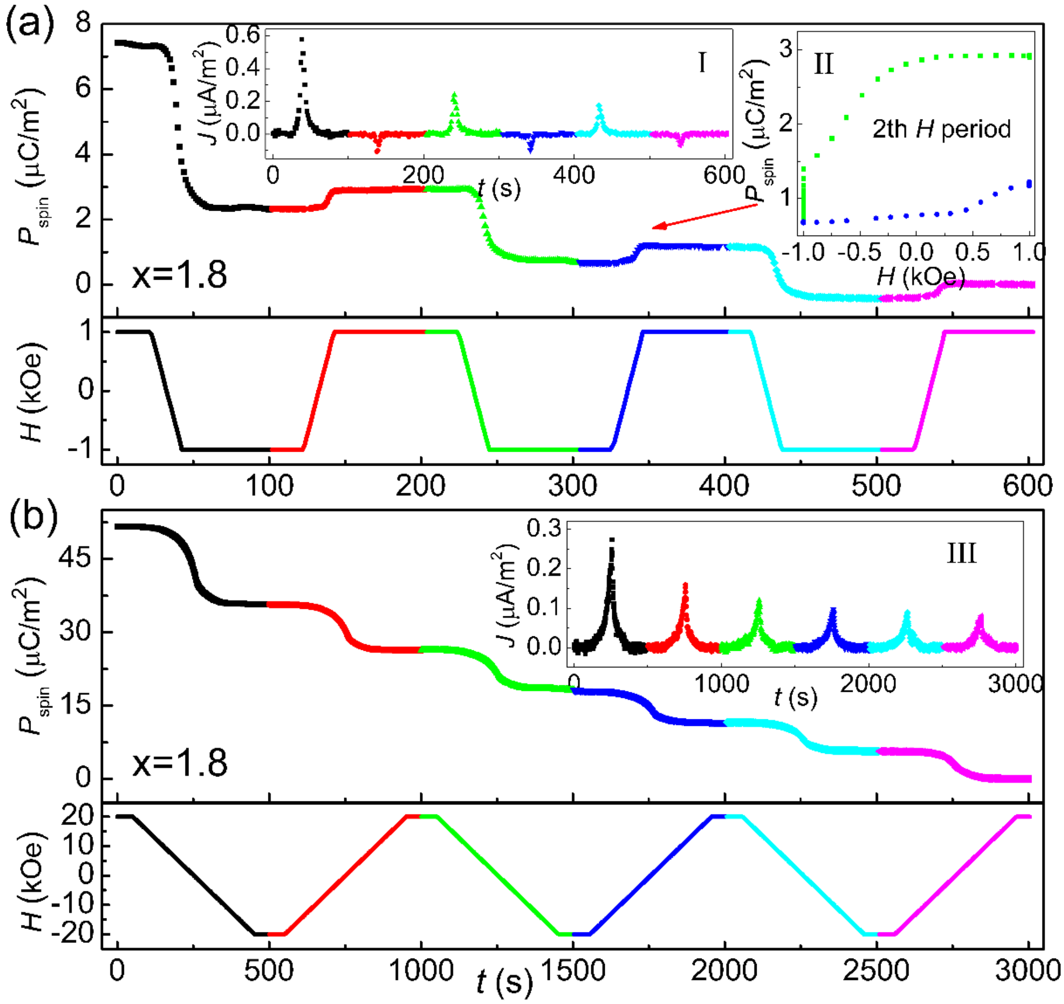


Figure S4. The evolution of P_{spin} with periodic H of the ceramic with x=1.8 under $H \perp E$ and 10 K, with H equals (a) 1 kOe and (b) 20 kOe. The insets I and III show the corresponding variation of current density J with periodic H. The inset II in (a) is the P_{spin}-H curve under $H \perp E$ and 10 K.

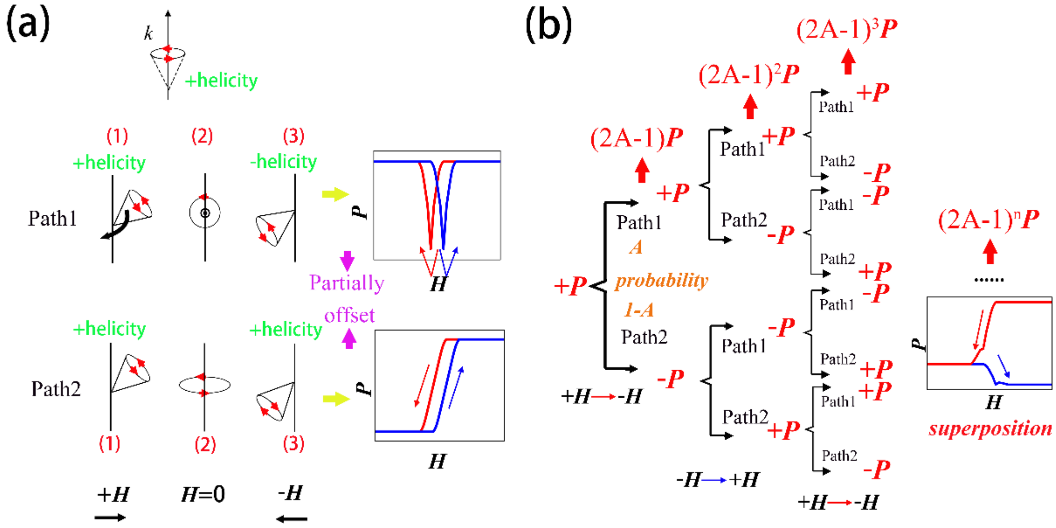
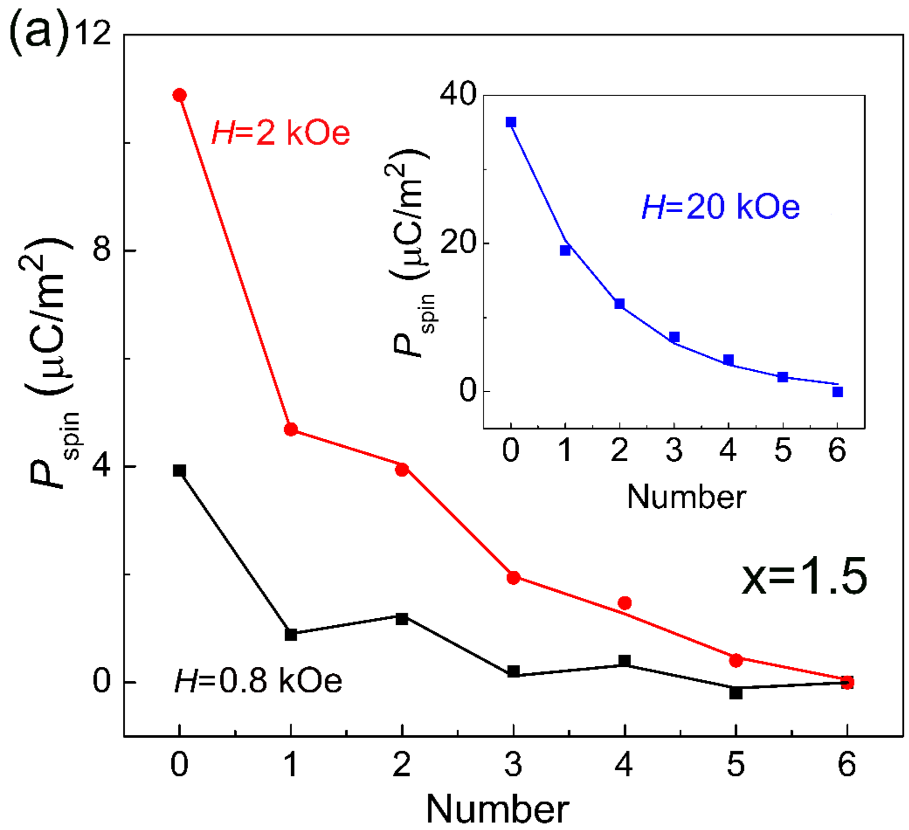


Figure S5 (a) Schematic diagram of the two spin flipping processes and their corresponding P-H curves, red arrows represent the conical arrangement. (b) Schematic diagram of P_{spin} evolution with magnetic field reversal.



(b)

	0.8 kOe	2 kOe	20 kOe
P_{free} ($\mu C/m^2$)	3.05	9.67	36.16
P_{pin} ($\mu C/m^2$)	1.04	2.20	——
η_{free}	0.57	0.69	0.57
A_{free}	0.785	0.845	0.785
η_{pin}	-0.64	-0.45	——
A_{pin}	0.18	0.275	——

Figure S6 (a) The evolution of P_{spin} with the number of H reversal and the fitting results by Equ. S1, and (b) fitting parameters of P_{spin} of $Sr(CuTi)_{1.5}Fe_9O_{19}$ ceramics.

Based on the spin flipping process shown in Fig. S5(b), the evolution of P_{spin} can be described by the following formula:

$$P_{spin}=P_s+P_{free}\eta_{free}{}^{n}+P_{pin}\eta_{pin}{}^{n} \tag{S1}$$

Here, P_s is a constant to compensate for the unreleased polarization. P_{free} and η_{free} respectively represent the polarization and attenuation factors of unpinned spin, and their flipping usually be realized through domain wall movement. P_{pin} and η_{pin} are the polarization and attenuation factors of the pinned spin, respectively. $\eta_{free/pin}$=2$A_{free/pin}$-1, and n is the number of H reversal. Based on such formula we proposed, the evolution process of P_{spin} are well fitted. As shown in Figure S6, P_{free} and P_{pin} coexist in $Sr(CuTi)_{1.5}Fe_9O_{19}$ ceramics under small H. The domain wall pining and the spin glass freezing prevent the corresponding spin flipping achieved through domain wall movement. This part of spin flipping is mainly realized by path 2, contributing to the negative J and P_{spin}, and thus the atypical hysteresis P_{spin}-H curve. While with the increase of H, the depinning of domain wall increases, and therefore the $\frac{P_{pin}}{P_{free}}$ ratio decreases from 0.34 to 0.23 as H increases from 800 Oe to 2 kOe, resulting in the disappearance of negative peak of J. Moreover, for H = 20 kOe, the evolution of P_{spin} can be fitted well only by P_{free}. In addition, A_{free} is distributed among 0.785 and 0.845, while A_{pin} is between 0.18 and 0.275, confirming the free and pinned spin flipping processes are dominated by path 1 and path 2, respectively.

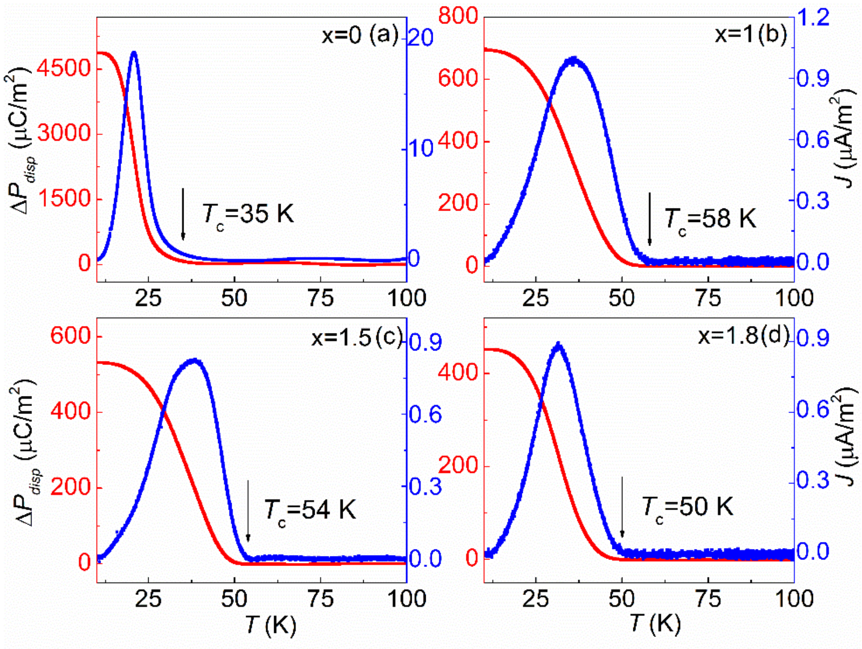


Figure S7 Temperature dependent ΔP_{disp} and J of $Sr(CuTi)_xFe_{12-2x}O_{19}$ ceramics. It should be mentioned that the value of ΔP_{disp} is incomparable among different samples because of their different degree of polarization under the same small polarized electric field (~1 MV/m).

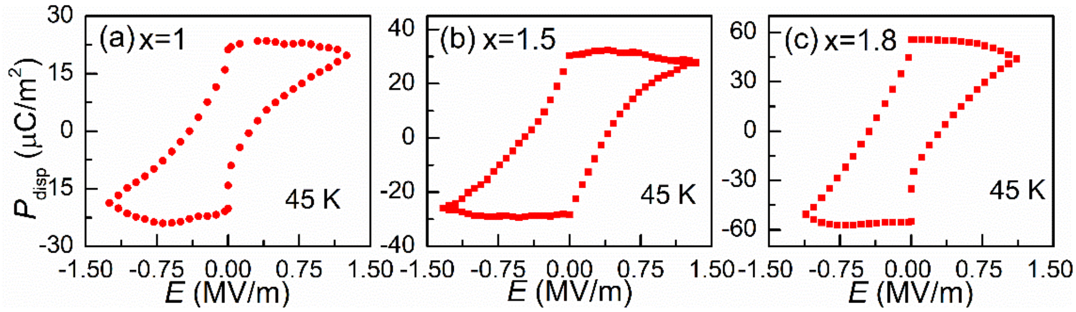


Figure S8 The ferroelectric hysteresis loops measured at 45 K by PUND method of $Sr(CuTi)_xFe_{12-2x}O_{19}$ ceramics. Here, the applied electric field corresponds to the voltage of 200 V, the largest acceptable voltage of PPMS. The poling electric field (~1MV/m) of pyroelectric measurement is applied from the temperature (~300 K) larger than T_C, and therefore higher degree of polarization and thus larger polarization are obtained in pyroelectric response.