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**High Pressure Synthesis, Structure, and Multiferroic Properties of Two Perovskite Compounds  $\text{Y}_2\text{FeMnO}_6$  and  $\text{Y}_2\text{CrMnO}_6$**

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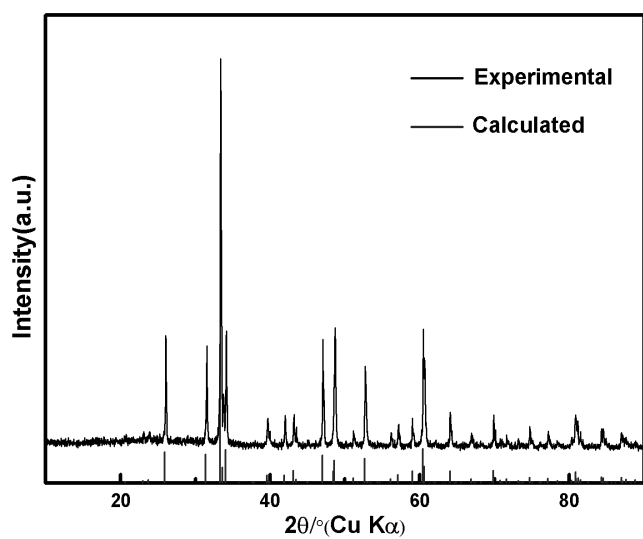
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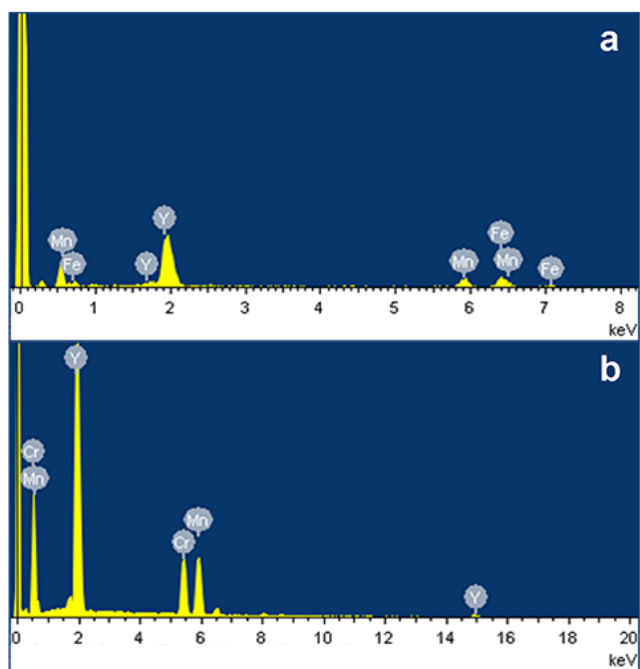
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**Figure S01. Powder XRD pattern of YFMO**



**Figure S01.** The experimental pattern was in close agreement with that calculated from the single-crystal data, and no secondary phase was detected.

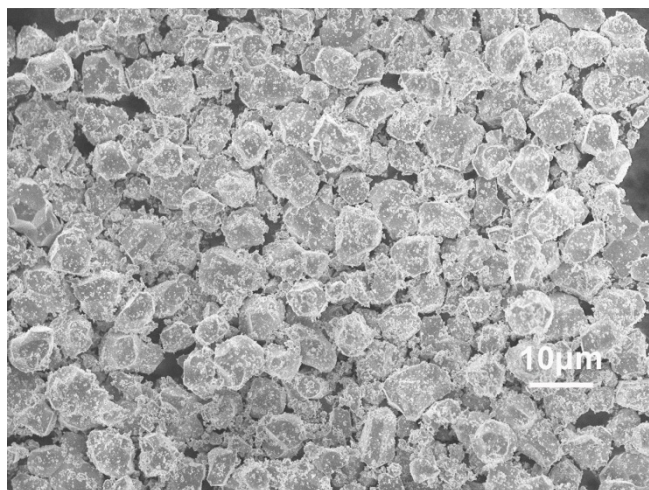
**Figure S02.** The EDS spectrum of: (a)  $\text{Y}_2\text{CrMnO}_6$  and (b)  $\text{Y}_2\text{FeMnO}_6$



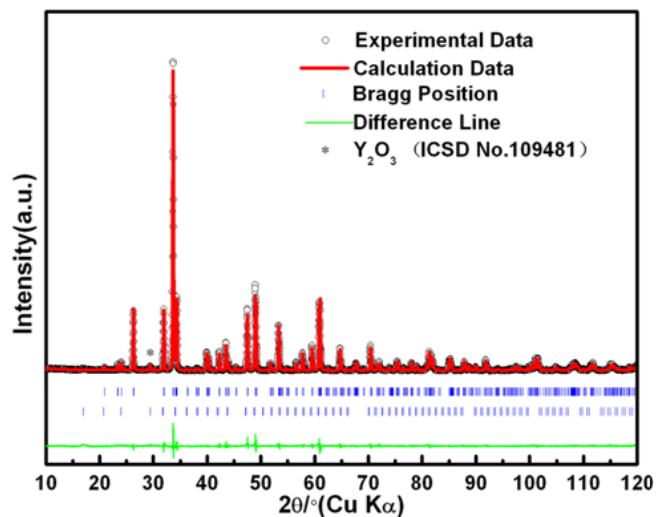
**Figure S02.** EDS spectrum of: (a)  $\text{Y}_2\text{CrMnO}_6$  (at%: Y/Cr/Mn= 1.01/0.5/0.49) and (b)  $\text{Y}_2\text{FeMnO}_6$  (at%: Y/Fe/Mn= 1.13/0.45/0.42). The EDS of YCMO and YFMO were observed using SEM (JSM-6700F, JEOL, Japan) equipped with energy dispersive X-ray spectroscopy.

**Figure S03. SEM image of YCMO synthesized under ambient pressure**

The YCMO samples obtained under ambient pressure (AP-YCMO) was examined. The impurities can be seen in the SEM image and the powder X-ray diffraction pattern. Though there was no other extra peaks appeared in the X-Ray Diffraction (XRD) result, we have sufficient reason to believe that a trace amount of  $\text{Mn}_2\text{O}_3$  and  $\text{Cr}_2\text{O}_3$  is left over which have not be examined by XRD method. The unreasonable inflection point at 25K in the ZFC curve and the wasp-waist hysteresis loop at 5K suggest that the impurities which have not been detected by the XRD method seriously affect the magnetism data.

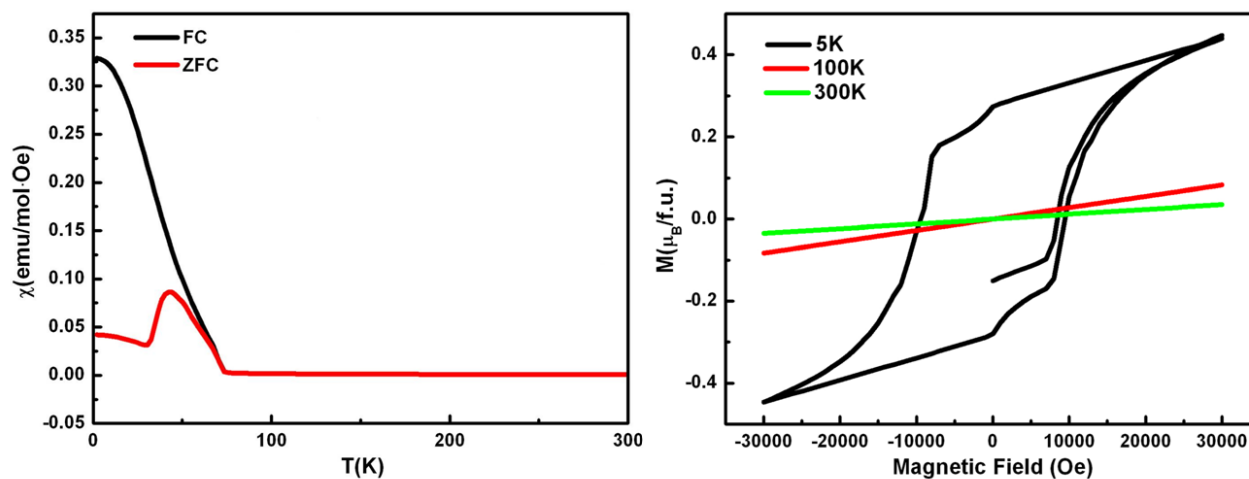


**Figure S04.** Observed and calculated X-ray diffraction pattern of AP-YCMO at room temperature



**Figure S04.** Observed and calculated X-ray diffraction pattern of AP-YCMO at room temperature.  $\text{Y}_2\text{O}_3$  (neither ferroelectric nor magnetic) impurity phase was indicated by an asterisk.

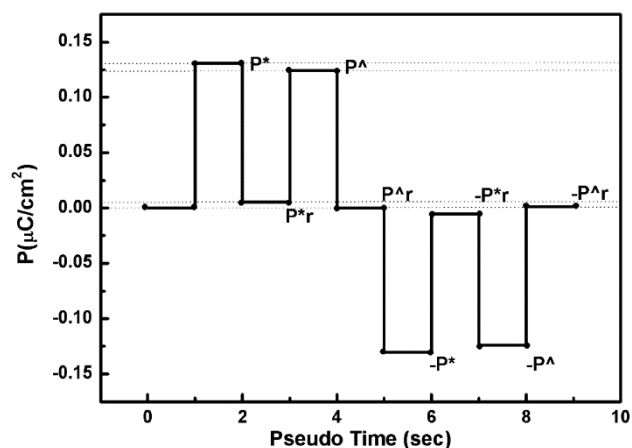
**Figure S05.** Temperature-dependent magnetization curves and hysteresis loops of AP-YCMO



**Figure S05.** Temperature-dependent magnetization curves (a) and hysteresis loops (b) recorded of AP-YCMO. Molar susceptibility versus temperature curves showing three anomalies at 25, 50 and 70 K. The unreasonable inflection point at 25K in the ZFC curve and the wasp-waist hysteresis loop at 5K suggest that the impurities which have not been detected by the XRD method seriously affect the magnetism data.



**Figure S06. PUND (positive-up & negative-down) pulse result of  $\text{Y}_2\text{FeMnO}_6$  at 77 K**



**Figure S06.** PUND (positive-up & negative-down) pulse result of YFMO at 77 K. The PUND result was obtained by employing the pulse width of 10  $\mu\text{s}$  and the delay time of 1000 msec. The saturation value of the polarization was obtained by applying an electric field of 50 kV/cm. The net switching polarization ( $2P_r$ ) was evaluated using the following relation:  $(2P_r) = (\pm P^*) - (\pm P^A)$ . Remanent polarization  $P_r$  value obtained from the PUND study is  $\sim 0.006 \mu\text{C}/\text{cm}^2$ .

**Table S01. Structure Parameters for Orthorhombic YFMO**

atom	Wyckoff site	x	y	z	$U_{iso}(\text{\AA}^2)$
Y	4c	0.576(0)	0.250(0)	1.017(9)	0.007(7)
Mn (Fe)	4a	1.000(0)	0.500(0)	1.000(0)	0.005(0)
O1	4c	1.038(0)	0.750(0)	1.110(9)	0.009(4)
O2	8d	1.184(0)	0.555(3)	0.696(7)	0.009(3)