


Room-temperature multiferroic behavior in layer-structured Aurivillius phase ceramics

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ABSTRACT

M

... H , - A ... B_{5.25}L_{0.75}F₃O₁₈ ... A , *in situ* ... F³⁺ O F³⁺, C³⁺ O C³⁺, F³⁺ O C³⁺ ... A , C /F ...

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M (FM) (FE) - A B₅F₃O₁₅ (= 4) B₆F₂₃O₁₈ (= 5), B₄F₃O₁₂ ... H , - , B F O₃, FE FM ... C F B- , 12,13 B B₅F_{0.5}C_{0.5}₃O₁₅ ... (= 4) B₆F₃O₁₈ (= 5) ... (B₂O₂)²⁺(A₋₁B₀O₃₊₁)²⁻ ... A , 14,15 H , ...)⁶ ... 16 - ... B- B F O₃ ... 7 11 ... A

$B_{5.25}L_{0.75}F_{1.0}C_{1.0}O_{18}$
 $(BLFC)_{1-x}A_x$
 $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$, $c = 50.670(2) \text{ \AA}$
 $b = 5.3943(6) \text{ \AA}$, $c = 41.487(2) \text{ \AA}$

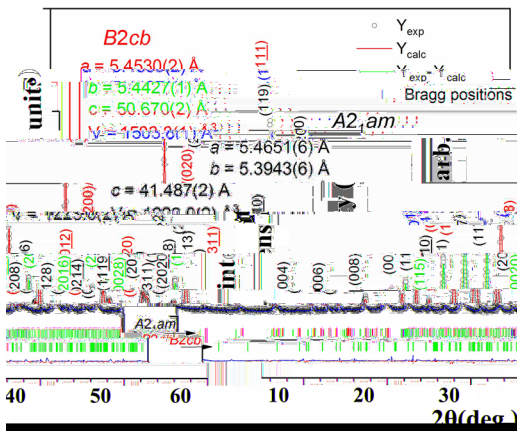


FIG. 1. XRD patterns of B2cb and A21am phases.

$B_{5F_{0.5}C_{0.5}O_{15}}$
 $(BLFC)_{1-x}A_x$
 $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$, $c = 50.670(2) \text{ \AA}$
 $b = 5.3943(6) \text{ \AA}$, $c = 41.487(2) \text{ \AA}$

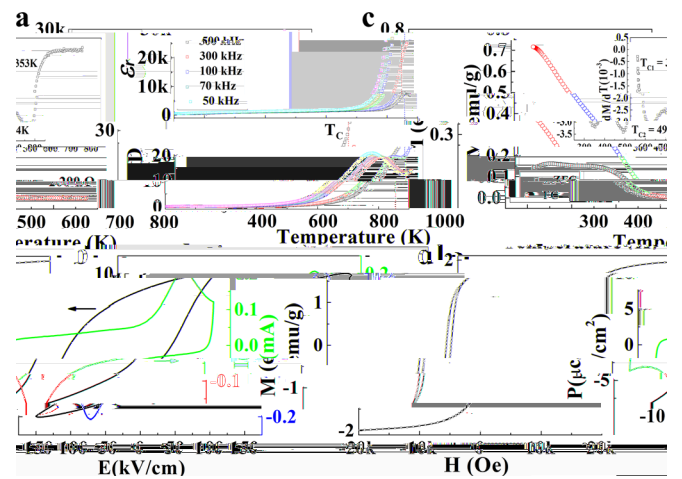


FIG. 2. (a) Temperature dependence of dielectric loss, (b) temperature dependence of the Curie temperature, and (c) temperature dependence of the piezoelectric coefficient for BLFC_{1-x}A_x.

~ 494 K
 M/μ_B ,
 $B_6F_2C_{18}O_{18}$ (526 K).²³
 BLFC
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$ (.
 ED
 $FC \sim 353$ K
 $C_2F_4O_4$ (460 K)
 $(M) C_2F_4O_4$ 16,25
 $16.235 / .^{25}$, 1.4 . %
 $C_2F_4O_4$ 0.22 0.32 / , BLFC
 $M = 1.85 / , F . 2(1.1$
 $M H$
 ~ 425 K 1.58 / .
 $0.27 / ,$ ED
 BLFC
 $F^{3+} O C^{3+}$
 (DF)
 (A) *ab initio*
 $\mu_F = 2$ $\mu_C = 3$ F C ,
 (GGA)+ μ . I
 BLFC
 $F . 3(1, F^{3+} C^{3+} (3.1 2.1 \mu_B/ ,)$,
 $(0.1 \mu_B/)$.
 $F O_6 C O_6$ F/C -
 $F O - / . F . 3(1.1$
 $F^{3+} C^{3+}$,
 $(. ,)$,
 $E_{FM} - E_{AFM}$
 $= -144.1$.
 H , (FM)
 $43.5 (. . , 504.6$ K), FM
 ~ 1 FC/FC . F . 2(1.1 .
 $a b$
 010
 BLFC
 $F . 4$
 I
 399 O .
 $F .$
 $F -$

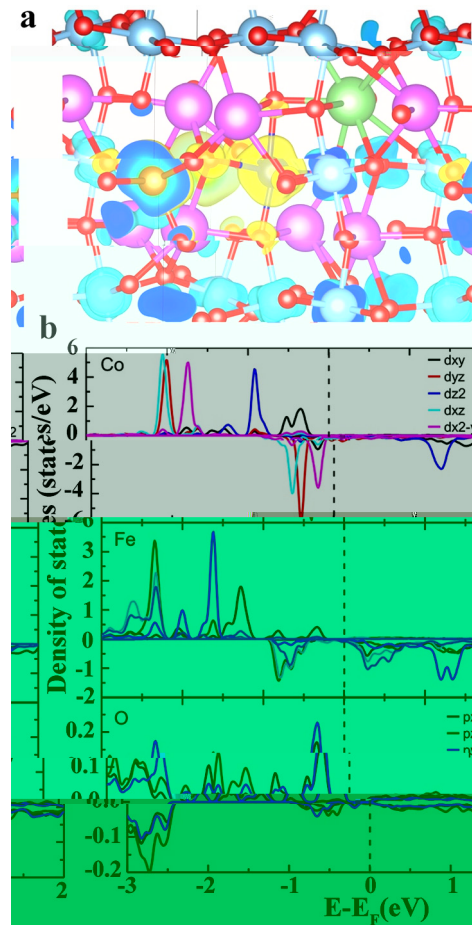


FIG. 3. (a) Crystal structure of BLFC with lattice parameters $a = b = 0.354$ nm, $c = 0.399$ nm, and $\beta = 90^\circ$. (b) Density of states (DOS) for Co, Fe, and O atoms. The DOS is shown in units of states/eV versus energy $E - E_f$ (eV). The legend indicates the contributions of different orbitals: d_{xy} (black), d_{yz} (red), d_{z^2} (blue), d_{xz} (green), $d_{x^2-y^2}$ (purple) for Co; p_x (red), p_y (green), p_z (blue) for Fe; and p_x (red), p_y (green), p_z (blue) for O.

~ 494 K
 M/μ_B ,
 $B_6F_2C_{18}O_{18}$ (526 K).²³
 BLFC
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$ (.
 ED
 $FC \sim 353$ K
 $C_2F_4O_4$ (460 K)
 $(M) C_2F_4O_4$ 16,25
 $16.235 / .^{25}$, 1.4 . %
 $C_2F_4O_4$ 0.22 0.32 / , BLFC
 $M = 1.85 / , F . 2(1.1$
 $M H$
 ~ 425 K 1.58 / .
 $0.27 / ,$ ED
 BLFC
 $F^{3+} O C^{3+}$
 (DF)
 (A) *ab initio*
 $\mu_F = 2$ $\mu_C = 3$ F C ,
 (GGA)+ μ . I
 BLFC
 $F . 3(1, F^{3+} C^{3+} (3.1 2.1 \mu_B/ ,)$,
 $(0.1 \mu_B/)$.
 $F O_6 C O_6$ F/C -
 $F O - / . F . 3(1.1$
 $F^{3+} C^{3+}$,
 $(. ,)$,
 $E_{FM} - E_{AFM}$
 $= -144.1$.
 H , (FM)
 $43.5 (. . , 504.6$ K), FM
 ~ 1 FC/FC . F . 2(1.1 .
 $a b$
 010
 BLFC
 $F . 4$
 I
 399 O .
 $F .$
 $F -$

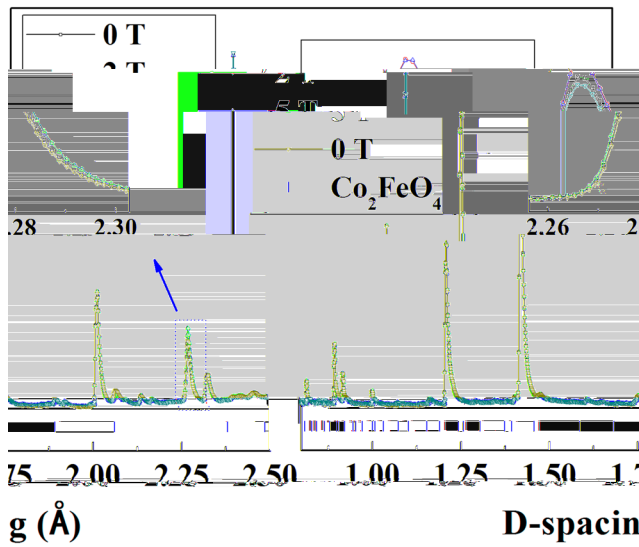


FIG. 4. XRD patterns of Co_2FeO_4 at 0 T and 2 T. The inset shows the schematic of the sample and magnetometer. The bottom panel shows the Rietveld refinement with Bragg peak positions marked.

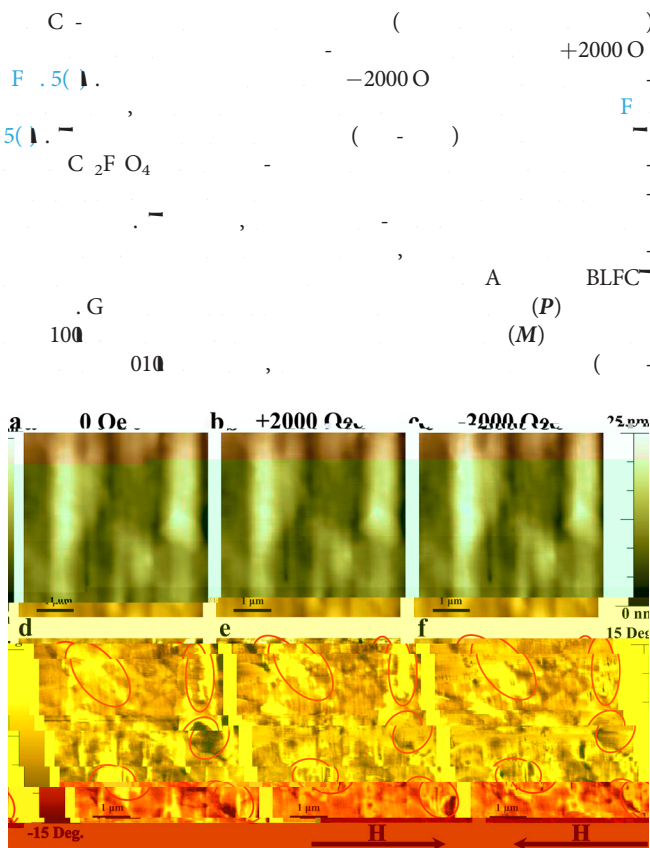


FIG. 5. MFM images of Co_2FeO_4 at different magnetic fields: (a) 0 Oe, (b) +2000 Oe, (c) -2000 Oe, (d) 0 Oe, (e) +2000 Oe, (f) -2000 Oe. The images show magnetic domains with red and blue contrast. Scale bars are 1 μm. The bottom panel shows the Rietveld refinement with Bragg peak positions marked.

$T = P \times M$
 BLFC⁻
 I , A BLFC⁻
 F
 $\text{C}^{3+} \text{O} \text{C}^{3+}, \text{F}^{3+} \text{O} \text{C}^{3+}$
 $\text{F}^{3+} \text{O} \text{F}^{3+}$
 A , C / F
 EM (ED)
 BLFC⁻
 D . M , D . K , D .
 D I H I I N , AL,
 D , O , K .
 A E D F
 G A A (G N . 2/
 0038/20), C (G N . K2015-0602006), N FC (G
 N . 11474138 11834005). A
 E M (EM)
 IND54 N EM
 EM E₂AME⁻ E

DATA AVAILABILITY

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