

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

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ABSTRACT

Multiferroic Aurivillius phase ceramics are a class of layered perovskites with the general formula $B_{5-2x}L_xO_{15}$, where B is a divalent or trivalent B-site cation, L is a monovalent or divalent L-site cation, and x is the amount of L-site cation. The Aurivillius phase ceramics exhibit a variety of physical properties, including ferroelectricity, piezoelectricity, and magnetostriction. In this work, we report the synthesis and characterization of a new Aurivillius phase ceramic, $B_5F_{0.75}C_{0.25}O_{18}$. This material exhibits room-temperature multiferroic behavior, showing both ferroelectric and magnetic properties. The ferroelectricity is observed as a hysteresis loop in the polarization-electric field ($P-E$) loop, and the magnetic properties are observed as a hysteresis loop in the magnetization-field ($M-H$) loop. The origin of the multiferroic behavior is discussed in terms of the structural distortions and the interactions between the different structural units.

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Keywords: Aurivillius phase, multiferroic, room-temperature, ferroelectric, magnetic.

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$B_6F_2O_{18}$ (BLFC) $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$, $c = 50.670(2) \text{ \AA}$, $Z = 4$, $V = 1503.38(1) \text{ \AA}^3$, $T_c = 354 \text{ K}$, $T_N = 494 \text{ K}$.
 Crystallographic parameters for $B_6F_2O_{18}$ in the $B2cb$ space group. The lattice constants are $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$, and $c = 50.670(2) \text{ \AA}$. The volume is $V = 1503.38(1) \text{ \AA}^3$. The structure is refined with $R = 0.018$ and $wR = 0.025$. The space group is $B2cb$. The structure is shown in the $in situ$ neutron diffraction experiment. The structure is refined with $R = 0.018$ and $wR = 0.025$. The space group is $B2cb$. The structure is refined with $R = 0.018$ and $wR = 0.025$. The space group is $B2cb$.

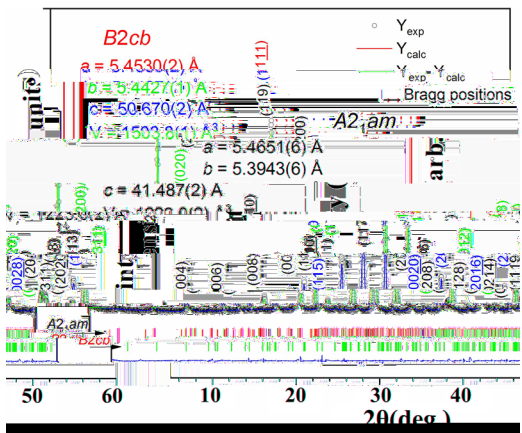


FIG. 1. XRD pattern of $B_6F_2O_{18}$ in the $B2cb$ space group.

Crystallographic parameters for $B_6F_2O_{18}$ in the $B2cb$ space group. The lattice constants are $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$, and $c = 50.670(2) \text{ \AA}$. The volume is $V = 1503.38(1) \text{ \AA}^3$. The structure is refined with $R = 0.018$ and $wR = 0.025$. The space group is $B2cb$. The structure is refined with $R = 0.018$ and $wR = 0.025$. The space group is $B2cb$. The structure is refined with $R = 0.018$ and $wR = 0.025$. The space group is $B2cb$.

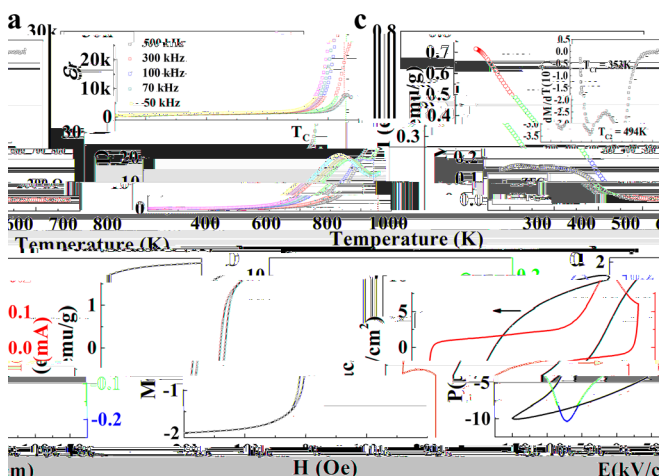


FIG. 2. (a) Dielectric loss $\tan \delta$ vs temperature (K) for BLFC at various frequencies. (b) Temperature dependence of the dielectric constant ϵ' . (c) Temperature dependence of the magnetic susceptibility χ . (d) Magnetization M vs magnetic field H at various temperatures. (e) Field-cooled (FC) and zero-field-cooled (ZFC) magnetization curves. (f) P-E hysteresis loops for BLFC at different temperatures.

~ 494 K
 $B_6FC_3O_{18}$ (526 K).²³
 BLFC
 $F^{3+} O F^{3+}, C_a^{3+} O C_a^{3+}, F^{3+} O C^{3+}$ (\dots).²⁴
 ED
 FC ~ 353 K
 $C_2F_2O_4$ (460 K)
 (M) $C_2F_2O_4$.^{16,25}
 $16.235 / \dots$
 $C_2F_2O_4$ $0.22, 0.32 / \dots$ BLFC
 $M = 1.85 / \dots$ BLFC
 $2(F_a \cdot 3)$ 425 K $1.58 / \dots$ ED
 $0.27 / \dots$ BLFC
 F_a^3
 (DF) $F^{3+} O C^{3+}$ *ab initio*
 (A)
 $F = 2$ $C = 3$ $F_a C_a$
 $(GGA) + \dots$
 $F \cdot 3(a)$ BLFC $F^{3+} C^{3+}$ (3.1 $2.1 \mu_B/a$)
 $0.1 \mu_B/a$
 F/C $F \cdot 3(a)$
 $F_a O_a$
 $F^{3+} C^{3+}$
 (\dots)
 $E_{FM} - E_{AFM}$
 $= -144.1$
 H_a 43.5 ($\dots, 504.6$ K), (FM) FM
 FC/FC $F \cdot 2(a)$
 010
 BLFC F_a^4
 I

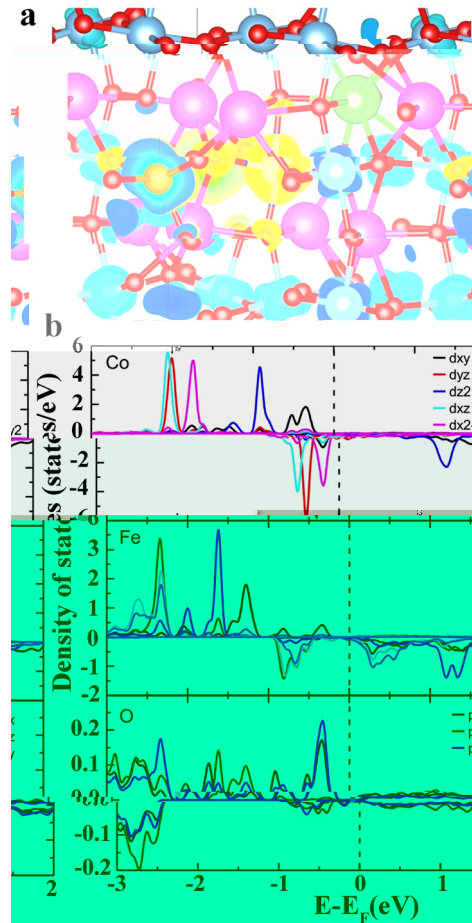


FIG. 3. (a) Crystal structure of BLFC. (b) Density of states (DOS) plots for Co, Fe, and O orbitals. The Co plot shows dxy, dyz, dz2, dxz, and dx2-y2 orbitals. The Fe plot shows d orbitals. The O plot shows p orbitals. The x-axis is E - Ef (eV) from -3 to 1, and the y-axis is Density of states (states/eV).

N
 I F_a^4 $(0, 1, 20)$
 $(2 \leq H < 5)$
 $M H$ $F_a^2(a)$ $3 F_a$
 F_a^5 BLFC $F M$
 $5(a)$ A FM BLFC $399 O$ F

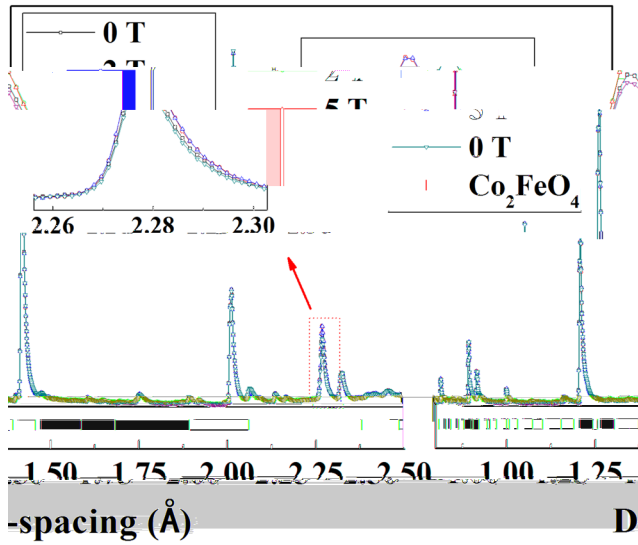


FIG. 4. XRD patterns of Co_2FeO_4 at 0 T (blue) and 5 T (red). The inset shows a zoomed-in view of the peaks between 2.26 and 2.30 Å. A red arrow points to a peak at approximately 2.27 Å.

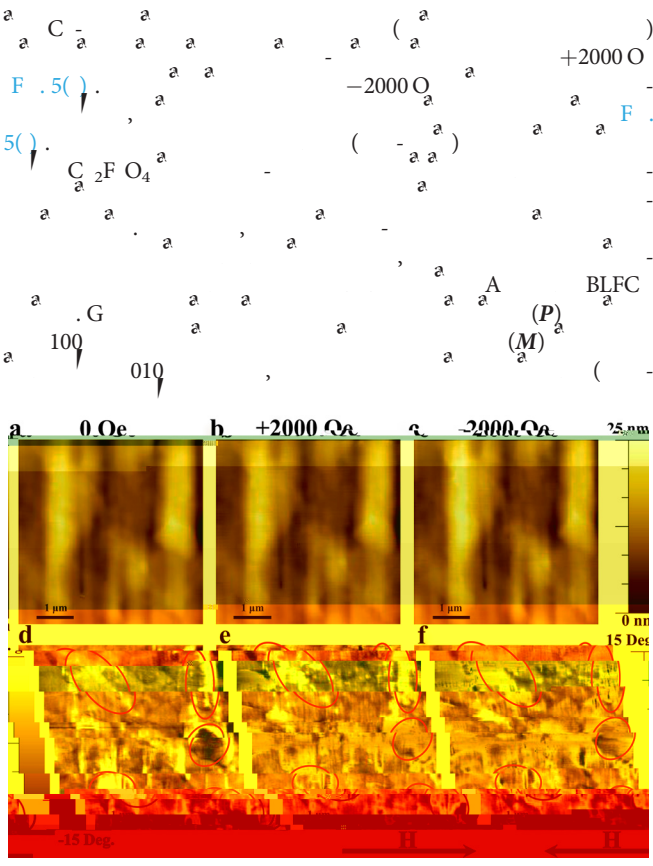


FIG. 5. MFM images of BLFC at different magnetic fields: (a) 0 Oe, (b) +2000 Oe, (c) -2000 Oe. Panels (d), (e), and (f) show corresponding phase images. Scale bars are 1 μm and 25 nm.

$T = P \times M$
 BLFC
 $\text{C}^{3+} \text{O}_2 \text{C}^{3+}$, $\text{F}^{3+} \text{O}_2 \text{C}^{3+}$, $\text{F}^{3+} \text{O} \text{F}^{3+}$,
 C_2/F
 EM (ED)
 BLFC
 D.M., D.K., D.
 I.H., I.I.N., AL,
 D., O., K.
 A.A.E., D.A.F.,
 G.A.A., N.K2015-0602006), N.F.C. (G
 0038/20), C (G.N. 11834005), N. 11474138
 N. 11834005). A (EM)
 IND54 N.M. EM
 E.A.M.E. E.

DATA AVAILABILITY

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