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Zheng Li,<sup>1</sup> Vladimir Koval,<sup>2</sup> Amit Mahajan,<sup>3</sup> Zhipeng Gao,<sup>4</sup> Carlo Vecchini,<sup>5</sup> Mark Stewart,<sup>5</sup> Markys G. Cain,<sup>6</sup> Kun Tao,<sup>7</sup> Chenglong Jia,<sup>7,a)</sup> Giuseppe Viola,<sup>3</sup> and Haixue Yan<sup>3,b)</sup>

## AFFILIATIONS

<sup>1</sup>Guangdong Provincial Key Laboratory of Applied Superconductivity, Institute of Superconducting Ceramics, Institute of Materials, Chinese Academy of Sciences, Shenzhen 518007, China  
<sup>2</sup>Department of Materials Science and Engineering, University of Illinois at Chicago, Chicago, Illinois 60607, USA  
<sup>3</sup>Department of Physics, University of California, San Diego, La Jolla, California 92037, USA  
<sup>4</sup>National Key Laboratory of Applied Superconductivity, Institute of Materials, Chinese Academy of Sciences, Shenzhen 518007, China  
<sup>5</sup>Department of Materials Science and Engineering, University of Illinois at Chicago, Chicago, Illinois 60607, USA  
<sup>6</sup>Department of Physics, University of California, San Diego, La Jolla, California 92037, USA  
<sup>7</sup>Department of Materials Science and Engineering, University of Illinois at Chicago, Chicago, Illinois 60607, USA

a)Email: [cljia@uic.edu](mailto:cljia@uic.edu)

b)Author to whom correspondence should be addressed: [hyan@uic.edu](mailto:hyan@uic.edu)

## ABSTRACT

Multiferroic Aurivillius phase ceramics (APCs) have attracted significant attention due to their potential applications in spintronics and memory devices. However, the room-temperature multiferroic behavior in layer-structured APCs is still under investigation. In this work, we report the synthesis and characterization of a series of layer-structured APCs with the general formula  $B_{5.25}L_{0.75}F_{1-x}C_xO_{18}$  ( $x = 0, 0.25, 0.5, 0.75, 1$ ). The structural, magnetic, and ferroelectric properties are investigated. The results show that the room-temperature multiferroic behavior is significantly enhanced in the  $B_{5.25}L_{0.75}F_{0.5}C_{0.5}O_{18}$  sample, which exhibits a magnetic transition at  $T_M \approx 120$  K and a ferroelectric transition at  $T_F \approx 100$  K. The *in situ* X-ray diffraction (XRD) measurements reveal the structural changes during the phase transitions. The results indicate that the layer-structured APCs with a high degree of layer ordering exhibit enhanced room-temperature multiferroic behavior.

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I A B<sub>5.25</sub>L<sub>0.75</sub>F C<sub>3</sub>O<sub>18</sub>  
 (BLFC) P P (P).  
 Pa La  
 F, C, D<sup>14,17</sup>  
 A a b P  
 BLFC A  
 a b A  
 A  
 in situ  
 I H I I  
 N AL, D, O K  
 F  
 ( ) A BLFC  
 P A  
 BLFC P  
 F 1 (D) BLFC  
 A (FC)  
 B2cb A  
 A A<sub>21</sub>  
 A A<sub>21am</sub>  
 B2cb a = 5.4530(2) Å, b = 5.4427(1) Å, c = 50.670(2) Å  
 A A<sub>21am</sub> a = 5.4651(6) Å, b = 5.3943(6) Å, c = 41.487(2) Å  
 F P ( //

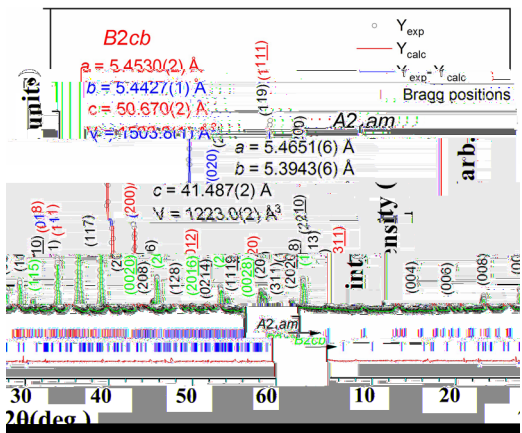


FIG. 1. XRD patterns of B2cb and A21am phases.

BLFC A  
 = 4 = 5 . N  
 D'  
 BLFC F 1 EM  
 (a-b) )  
 P . M  
 F . 1  
 . %  
 D. ED (F . 2  
 1) )  
 F, C, O, )  
 C<sub>2</sub>F O<sub>4</sub>  
 A B<sub>5</sub>F<sub>0.5</sub>C<sub>0.5</sub>O<sub>15</sub>.<sup>16</sup>  
 BLFC  
 P (50, 70 100,  
 300, 500 H).  
 1060 K FE T BLFC . H  
 BLFC B<sub>6</sub> F<sub>2</sub> O<sub>18</sub>  
 ( 973 K).<sup>13</sup> F 2( ) P-E I-E  
 BLFC P-I-E  
 P  
 21,22  
 BLFC 10 μC/ .  
 F 2( )  
 (FC) (FC)  
 200 O BLFC BLFC

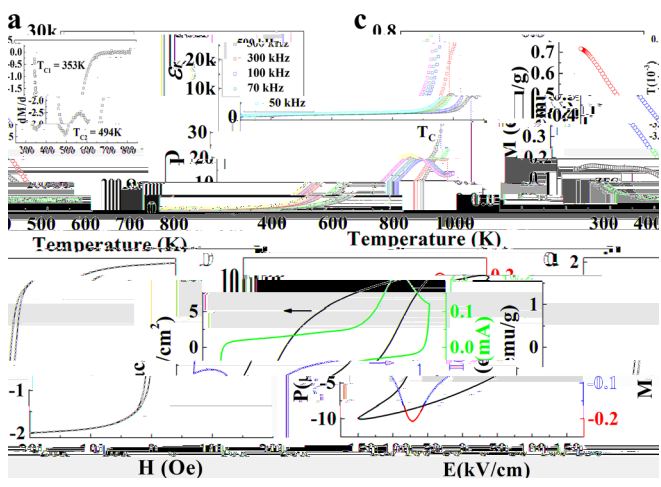


FIG. 2. Temperature dependence of dielectric permittivity (ε'), loss tangent (tan δ), and piezoelectric coefficient (d<sub>33</sub>) for BLFC. (a) ε' and tan δ vs. temperature for various frequencies. (b) d<sub>33</sub> and piezoelectric coefficient vs. temperature. (c) P-E loop and piezoelectric coefficient vs. magnetic field.

$\sim 494$  K (M/),  
 $B_6FC_3O_{18}$  (526 K).<sup>23</sup>  
 BLFC  
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$  (.).<sup>24</sup>  
 ED  
 $FC$   $\sim 353$  K  
 $C_2F_4O_4$  (460 K)  $16.25$  / .<sup>25</sup>  
 $C_2F_4O_4$  (M)  $C_2F_4O_4$  1.4 .%  
 $16.235$  / .<sup>25</sup>,  $0.22$   $0.32$  / ,  
 $C_2F_4O_4$  BLFC  
 $M = 1.85$  / ,  $F_2$  ( ) . I  
 $M H$   
 $2$  (F . 3).  
 $425$  K  $1.58$  / .  
 $0.27$  / ,  
 ED  
 BLFC  
 $F_3$   
 $F^{3+} O C^{3+}$   
 (DF) *ab initio*  
 (A P)  
 $F = 2$   $C = 3$   $F$   $C$  ,  
 (GGA) . I  
 BLFC  
 $F_3(a)$ ,  $F^{3+}$   $C^{3+}$  ( $3.1$   $2.1 \mu_B/a$  , ),  
 $O$   $0.1 \mu_B/a$  ).  
 $F O_6$   $C O_6$   
 ( ) F/C  
 $F$   $O$  /  $F_3$  ( ) .  
 $F^{3+}$   $C^{3+}$   
 ( . , )  
 $E_{FM} - E_{AFM} = -144.1$  .  
 $H$  (FM)  
 $43.5$  ( . , 504.6 K),  
 $FC/FC$  .  $F_2$  ( ) .  
 $a b$   
 $010$   
 $F_4$   
 BLFC . I  
 $399$  O .  
 $F_5$  .  
 $5$  ) . A

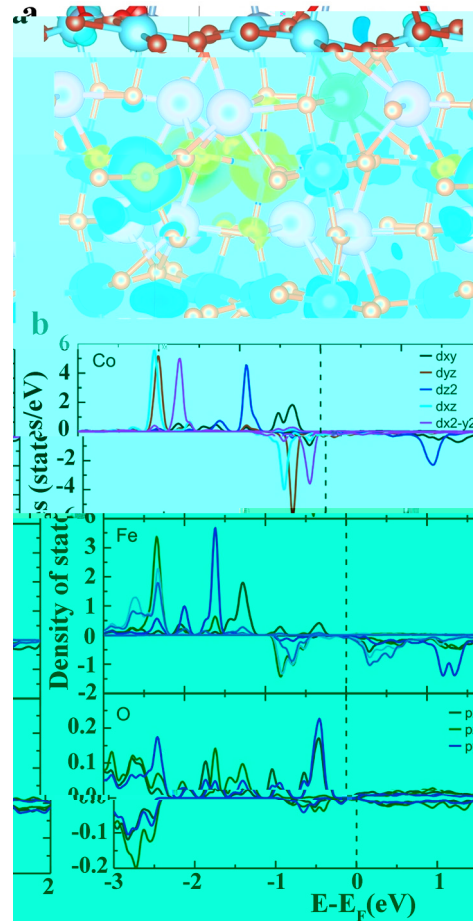


FIG. 3. (a) Crystal structure of BLFC. (b) Density of states (DOS) for Co, Fe, and O atoms. The DOS is calculated using the GGA+U method with  $U = 0.005$  eV. The DOS is shown for different orbitals:  $d_{xy}$ ,  $d_{yz}$ ,  $d_{z^2}$ ,  $d_{xz}$ ,  $d_{x^2-y^2}$  for Co;  $p_x$ ,  $p_y$ ,  $p_z$  for O. The energy  $E - E_f$  is in eV.

. N  
 . I  $F_4$  (0 1 20)  
 $2 < H < 5$  ,  
 $M H$   $F_2$  ( )  $3. F$  ,  
 $F_5$   
 BLFC P F M  
 $399$  O .  
 $F_5$  .  
 5 ) . A

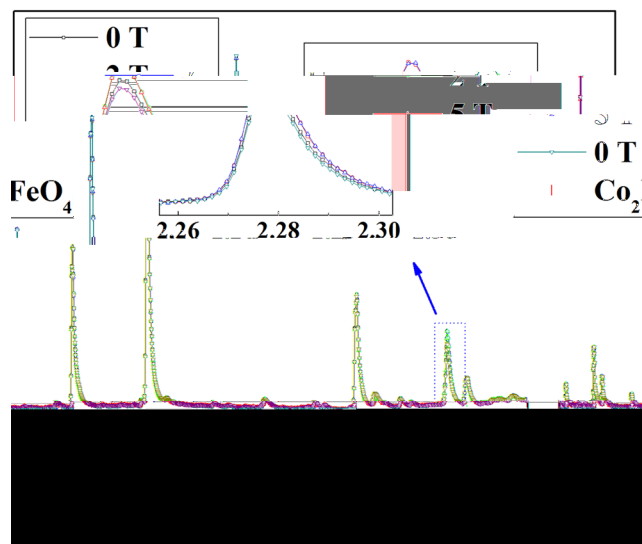


FIG. 4. XRD patterns of BLFC (001) and BLFC/FeO4 (001) at 0 T. The inset shows the schematic of the BLFC structure.

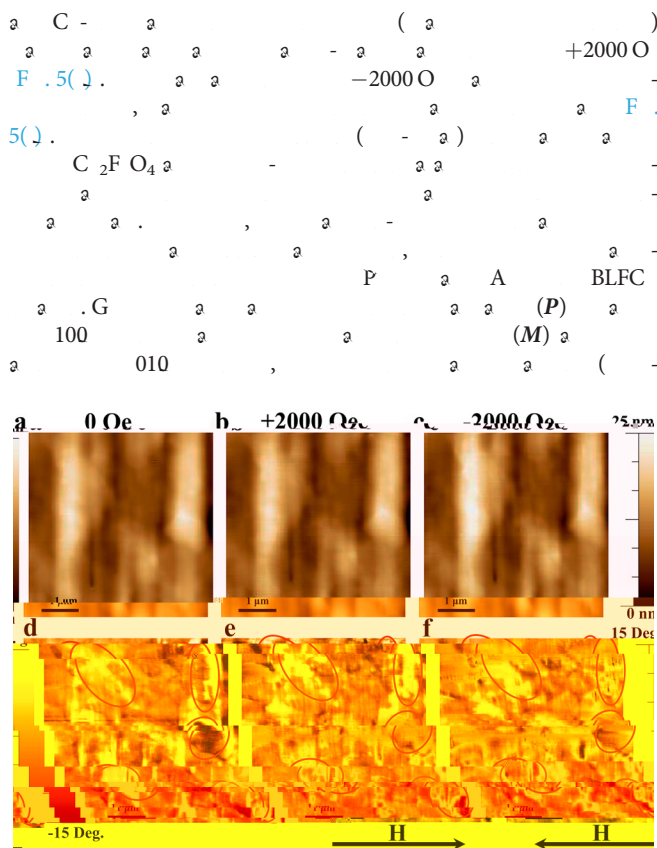


FIG. 5. TEM images of BLFC grains. (a) 0 Oe, (b) +2000 Oe, (c) -2000 Oe. (d-f) HRTEM images at -15, 0, and 15 degrees respectively.

$T = P \times M$   
 BLFC  
 I, A BLFC  
 F  
 $C^{3+} O C^{3+}, F^{3+} O C^{3+}$   
 $F^{3+} O F^{3+}$   
 A, C/F  
 EM (ED)  
 BLFC

D. Ma, P. D. K.  
 I H I I N, AL,  
 D, O K.  
 A E D F  
 G A A A (G N. 2/  
 0038/20), C (G N. K2015-0602006), N FC (G  
 N. 11474138 11834005). A  
 E M P (EM P)  
 P IND54 Na EM P  
 EM P AME E

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon request.

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