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## ABSTRACT

Multiferroic behavior is observed in layer-structured Aurivillius phase ceramics  $B_{5.25}L_{0.75}F_{1-x}C_xO_{18}$  ( $x = 0, 1, 2, 3, 4, 5$ ) at room temperature. The  $B_{5.25}L_{0.75}F_{1-x}C_xO_{18}$  ceramics exhibit a transition from a paraelectric (PE) state to a ferroelectric (FE) state at room temperature. The transition temperature  $T_C$  increases with the  $x$  value. The  $B_{5.25}L_{0.75}F_{1-x}C_xO_{18}$  ceramics exhibit a transition from a paraelectric (PE) state to a ferroelectric (FE) state at room temperature. The transition temperature  $T_C$  increases with the  $x$  value. The  $B_{5.25}L_{0.75}F_{1-x}C_xO_{18}$  ceramics exhibit a transition from a paraelectric (PE) state to a ferroelectric (FE) state at room temperature. The transition temperature  $T_C$  increases with the  $x$  value.

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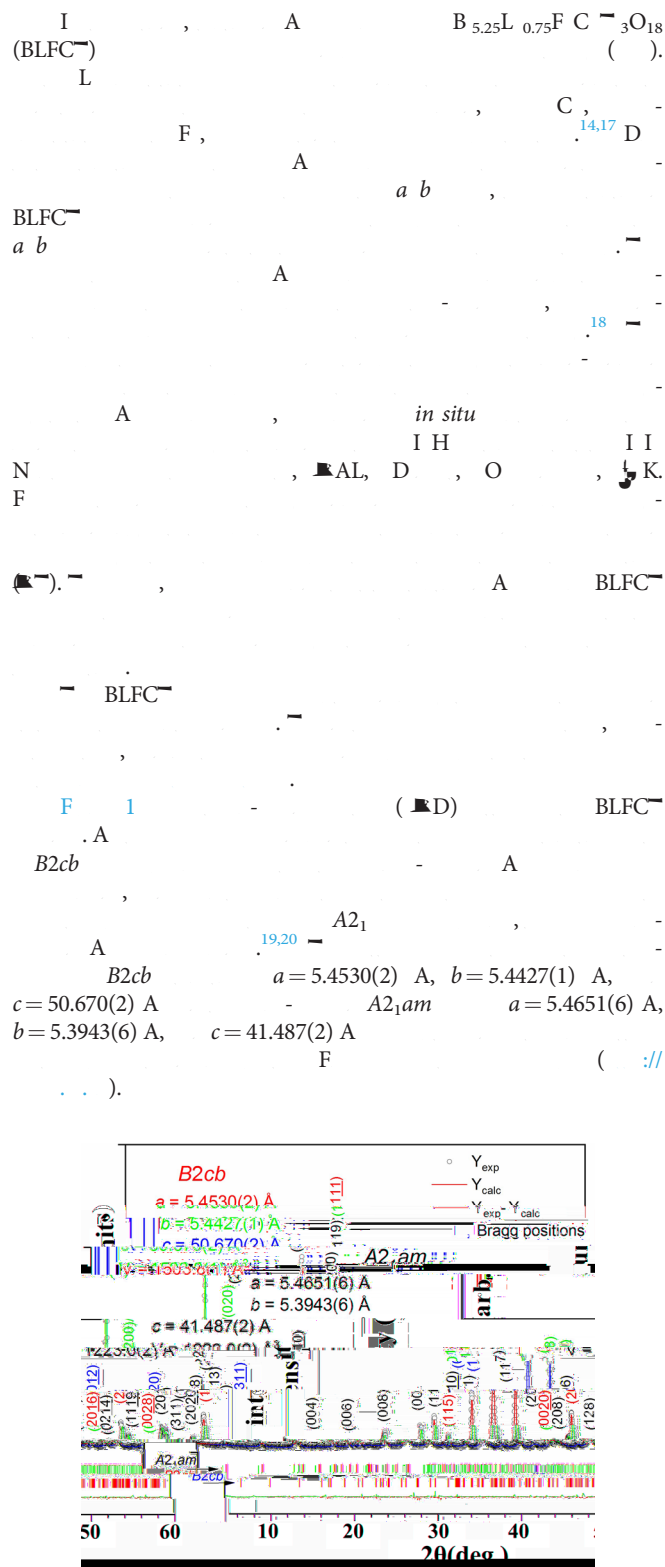


FIG. 1. XRD patterns of B2cb and A2,am phases.

BLFC (B<sub>5.25</sub>F<sub>0.75</sub>C<sub>18</sub>O<sub>15</sub>)  
 BLFC (B<sub>5</sub>F<sub>0.5</sub>C<sub>0.5</sub>O<sub>15</sub>)  
 BLFC (B<sub>6</sub>F<sub>3</sub>O<sub>18</sub>)  
 BLFC (B<sub>5</sub>F<sub>0.5</sub>C<sub>0.5</sub>O<sub>15</sub>)  
 BLFC (B<sub>6</sub>F<sub>3</sub>O<sub>18</sub>)  
 BLFC (B<sub>5</sub>F<sub>0.5</sub>C<sub>0.5</sub>O<sub>15</sub>)  
 BLFC (B<sub>6</sub>F<sub>3</sub>O<sub>18</sub>)

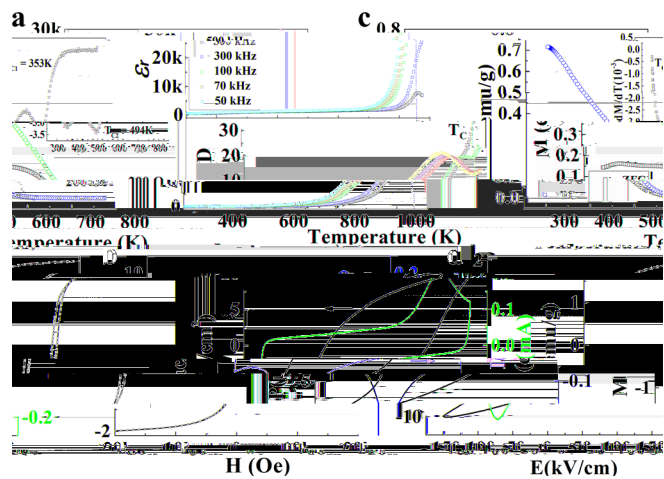
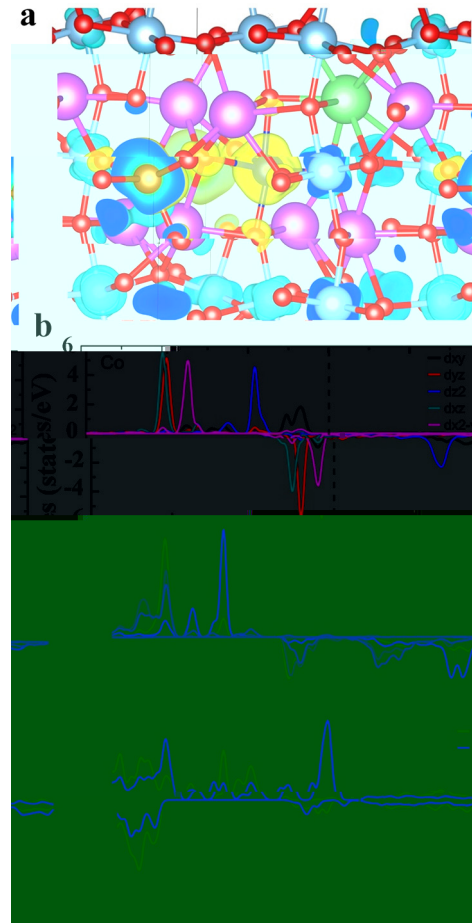


FIG. 2. (a) Raman spectra of BLFC at various temperatures. (b) Temperature dependence of Raman intensity. (c) Structural diagrams of BLFC.

$\sim 494$  K  
 $M/\mu_B$ ,  
 $B_6F_2C_{18}O_{18}$  (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  
 $\sim 353$  K  
 $FC$   
 $C_2F_4O_4$  (460 K)  
 $(M) C_2F_4O_4$  16.25%  
 $16.235 / 0.22$  0.32 / 1.4 %  
 $C_2F_4O_4$   
 $M = 1.85 / , F = 2(1.1)$   
 $M H$   
 $\sim 425$  K 1.58 / 0.27 / ,  
 ED  
 $BLFC$   
 $F^{3+} O C^{3+}$   
 (DF) *ab initio*  
 $(A)$   $H$   
 $\mu_F = 2$   $\mu_C = 3$   $F$   $C$  ,  
 (GGA)+  
 $BLFC$   
 $F = 3(1), F^{3+} C^{3+} (3.1 \ 2.1 \mu_B/)$  ,  
 $(0.1 \mu_B/)$  .  
 $F O_6 C O_6$   $F/C$  .  
 $F$   $O$  /  $F = 3(1)$  .  
 $F^{3+} C^{3+}$  ,  
 $(\dots)$  ,  
 $E_{FM} - E_{AFM}$   
 $= -144.1$  .  
 $H$  ,  
 $43.5$  ( , 504.6 K), (FM)  
 $\sim 1$  FC/FC  $F = 2(1)$  .  
 $a b$   
 $010$   
 $BLFC$   $F = 4$  .  
 $I$



**FIG. 3.** (a) Crystal structure of BLFC showing layers of fluorine (red), carbon (blue), and oxygen (green) atoms. (b) Density of states (DOS) plot showing energy levels (states/eV) versus energy (eV) for different components: dxy, dxz, dyz, and dz2.

$\sim 399$  O  
 $F = 5$   
 $BLFC$   $F M$   
 $FM$   $BLFC$  ,  
 $5(1)$  . A  $F = 2(1)$  3. F ,  
 $(2^{\sim} < H < 5^{\sim})$ ,  
 $M H$   $F = 2(1)$  3. F ,  
 $F = 4$ ,  
 $(0 \ 1 \ 20)$   
 $2^{\sim} F$   
 $I$  A  
 $N$

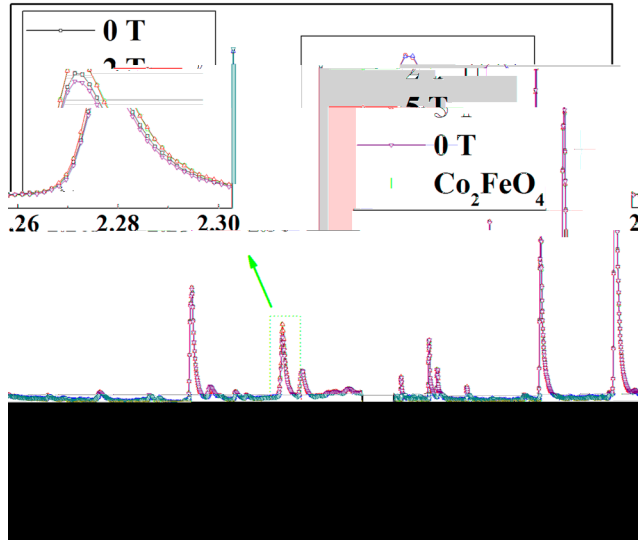


FIG. 4. XRD patterns of  $\text{Co}_2\text{FeO}_4$  at 0 T and 2 T. The inset shows the schematic of the sample and the XRD setup. A green arrow points to a peak at approximately  $2.285^\circ$ .

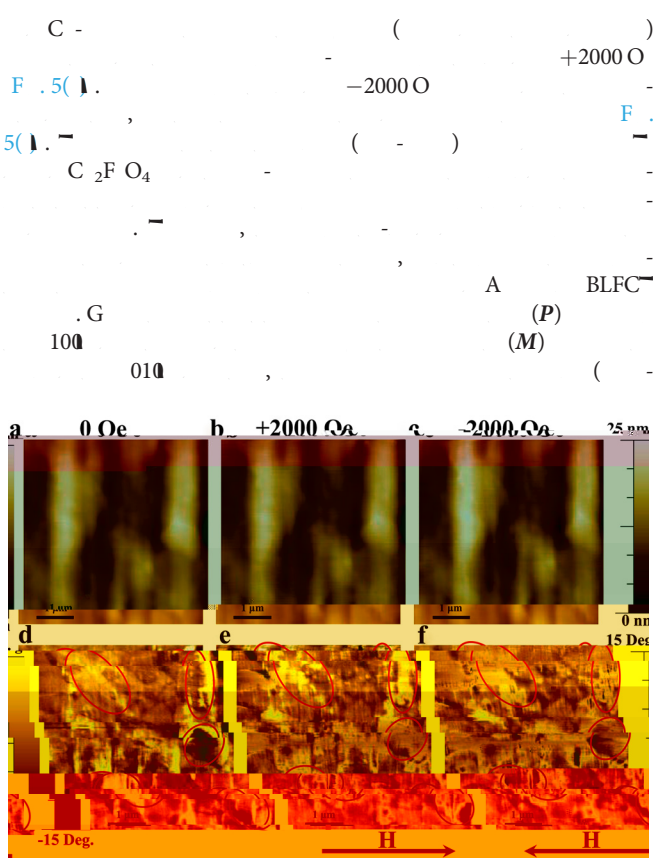


FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe, +2000 Oe, and -2000 Oe. Panels a, b, and c show the top surface, while d, e, and f show the bottom surface. Arrows labeled 'H' indicate the magnetic field direction.

$T = P \times M$   
 BLFC<sup>-</sup>  
 I , A BLFC<sup>-</sup>  
 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<sup>-</sup>  
 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 E<sub>2</sub>AME<sup>-</sup> E

DATA AVAILABILITY

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