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## ABSTRACT

Multiferroic Aurivillius phase ceramics (APCs) exhibit a rich variety of magnetic and ferroelectric (FE) properties. In this work, we report the synthesis and characterization of a new class of APCs,  $B_{5.25}L_{0.75}F_2C_3O_{18}$ , which shows a room-temperature magnetic transition and a ferroelectric-like behavior. The structure of the APCs is analyzed by *in situ* X-ray diffraction (XRD) and electron diffraction (ED). The structure is determined to be  $B_{5.25}L_{0.75}F_2C_3O_{18}$  with a space group of  $R\bar{3}m$ . The structure is similar to that of  $B_5F_2C_3O_{18}$  (space group  $R\bar{3}m$ ), but with the presence of  $L$  ions. The structure is analyzed by XRD and ED. The structure is determined to be  $B_{5.25}L_{0.75}F_2C_3O_{18}$  with a space group of  $R\bar{3}m$ . The structure is similar to that of  $B_5F_2C_3O_{18}$  (space group  $R\bar{3}m$ ), but with the presence of  $L$  ions.

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Multiferroic Aurivillius phase ceramics (APCs) exhibit a rich variety of magnetic and ferroelectric (FE) properties. In this work, we report the synthesis and characterization of a new class of APCs,  $B_{5.25}L_{0.75}F_2C_3O_{18}$ , which shows a room-temperature magnetic transition and a ferroelectric-like behavior. The structure of the APCs is analyzed by *in situ* X-ray diffraction (XRD) and electron diffraction (ED). The structure is determined to be  $B_{5.25}L_{0.75}F_2C_3O_{18}$  with a space group of  $R\bar{3}m$ . The structure is similar to that of  $B_5F_2C_3O_{18}$  (space group  $R\bar{3}m$ ), but with the presence of  $L$  ions. The structure is analyzed by XRD and ED. The structure is determined to be  $B_{5.25}L_{0.75}F_2C_3O_{18}$  with a space group of  $R\bar{3}m$ . The structure is similar to that of  $B_5F_2C_3O_{18}$  (space group  $R\bar{3}m$ ), but with the presence of  $L$  ions.

$B_{5.25}L_{0.75}F_{1.0}C_{3.0}O_{18}$   
 (BLFC) P L  
 F, A C, D  
 $a b$ , P  
 BLFC  
 $a b$   
 A  
 in situ  
 I H I I  
 N F AL, D O U K  
 (P), P A BLFC  
 BLFC P  
 F 1 (D) BLFC  
 A  
 $B2cb$  A  
 A  
 $B2cb$   $A2_1$   
 $a = 5.4530(2) \text{ \AA}$ ,  $b = 5.4427(1) \text{ \AA}$ ,  
 $c = 50.670(2) \text{ \AA}$ ,  $c = 41.487(2) \text{ \AA}$ ,  
 $b = 5.3943(6) \text{ \AA}$ ,  $c = 41.487(2) \text{ \AA}$   
 F P ( // )

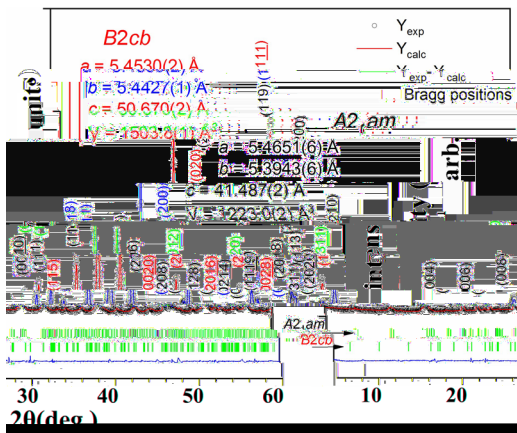


FIG. 1. XRD pattern of BLFC showing Bragg positions and unit cell parameters.

BLFC = 4 = 5 A N  
 BLFC F 1 EM (a-b) M  
 F 1  
 D. ED 1.4 %, (F 2  
 1)  
 $F, C, O, C_2F_2O_4$   
 A  $B_{5F_{0.5}C_{0.5}O_{15}}$ <sup>16</sup>  
 BLFC (50, 70 100,  
 300, 500 H).  
 1060 K FE T BLFC H,  
 ( 973 K).<sup>13</sup> F BLFC 2( ) P-E I-E  
 BLFC P I-E  
<sup>21,22</sup>  
 BLFC  $10 \mu C / 2$ .  
 F 2( ) (FC)  
 200 O BLFC BLFC

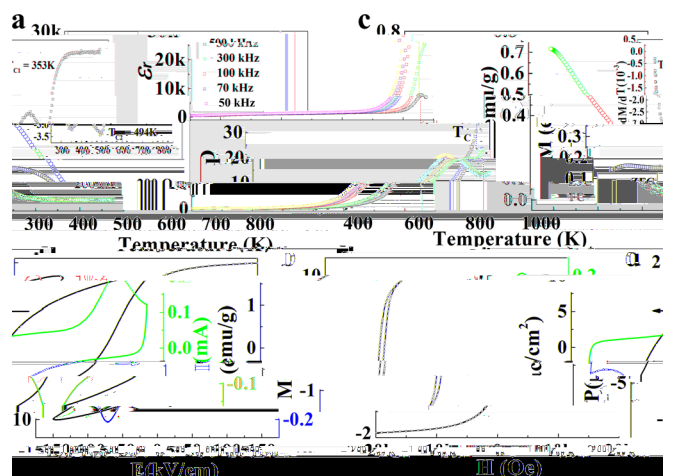


FIG. 2. (a) Dielectric loss tangent (tan δ) vs Temperature (K) for BLFC at frequencies of 300 kHz, 100 kHz, 70 kHz, and 50 kHz. (b) Magnetization (M) vs Temperature (K) showing hysteresis loops at various frequencies. (c) P-E and I-E loops showing ferroelectric and ferroelectric-like behavior.

$\sim 494$  K  
 $M/$  ),  
 $B_6F C_3O_{18}$  (526 K).<sup>23</sup>  
 BLFC  
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$  ( .  
 $ED$  .<sup>24</sup>  
 $A$  FC  $2 \sim 353$  K  
 $C_2F O_4$  .  
 $C_2F O_4$  (460 K) .<sup>16,25</sup>  
 $(M)$   $C_2F O_4$  .  
 $16 \ 23.5$  / .<sup>25</sup> ,  $1.4$  . %  
 $C_{2-} F O_4$   $0.22 \ 0.32$  / , BLFC  
 $M = 1.85$  / ,  $F . 2( ) . I$  ,  $M H$   
 $2 (F . 3)$  .  
 $425$  K  $1.58$  / .  $0.27$  / , ED  
 $BLFC$  ED  
 $A$  .  
 $F$   $3$  .  
 $(DF)$   $F^{3+} O C^{3+}$  *ab initio*  
 $(A P)$  .  $H$   
 $U_F = 2$   $U_C = 3$   $F C$  ,  
 $(GGA)$  .  $I$   
 $BLFC$   
 $F . 3( )$  ,  $F^{3+} C^{3+}$  ( $3.1 \ 2.1 \ \mu_B/$  , ) ,  
 $O$  .  
 $(0.1 \ \mu_B/)$  .  
 $F O_6$   $C O_6$   $F/C$  -  
 $( )$   $O$  - /  $F . 3( )$  .  
 $F$   $F^{3+} C^{3+}$  ,  
 $( . , )$   $( . , )$  .  
 $E_{FM} - E_{AFM}$   
 $= -144.1$  .  
 $H$  , (FM)  
 $43.5$  ( . , 504.6 K), FM  
 $1$  FC/FC  $F . 2( )$  .  
 $a b$   
 $010$  .  
 $BLFC$   $F$   $4$  .  $I$   
 $BLFC$  .  $I$   
 $5( ) . A$  PFM BLFC ,  $399 O$  .  
 $F$  .  
 $P$  F M  
 $5( ) . A$  PFM BLFC ,  $399 O$  .  
 $F$  .  
 $P$  F M

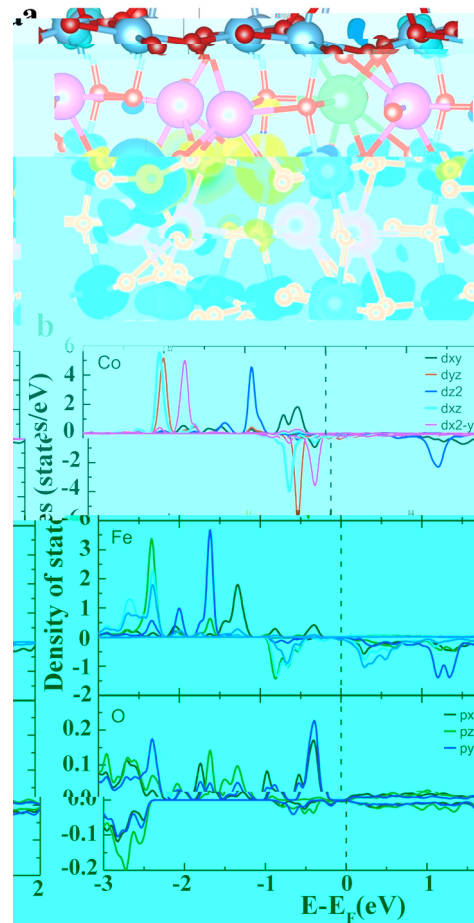


FIG. 3. (a) Crystal structure of BLFC showing layers of Co, Fe, and O atoms. (b) Density of states (DOS) plots for Co, Fe, and O atoms, showing contributions from various orbitals (dxy, dyz, dz2, dxz, dx2-y2, px, py, pz) across the energy range from -3 eV to 1 eV.

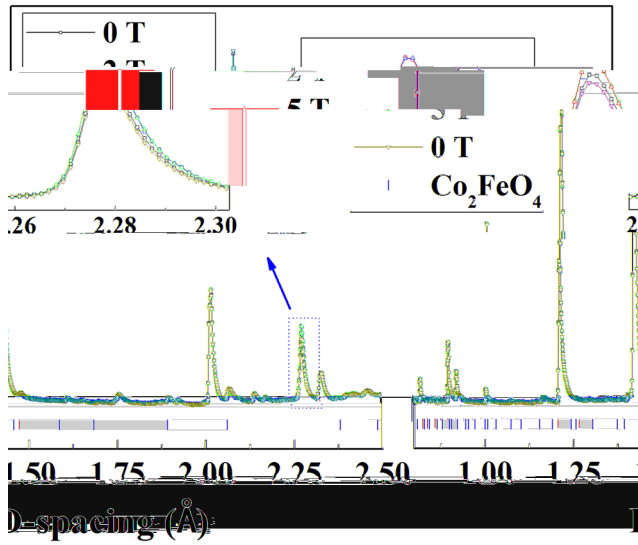


FIG. 4. XRD patterns of  $\text{Co}_2\text{FeO}_4$  at 0 T (red) and 5 T (black). The inset shows the XRD pattern at 0 T with a blue arrow pointing to the peak at  $d \approx 2.25$  Å.

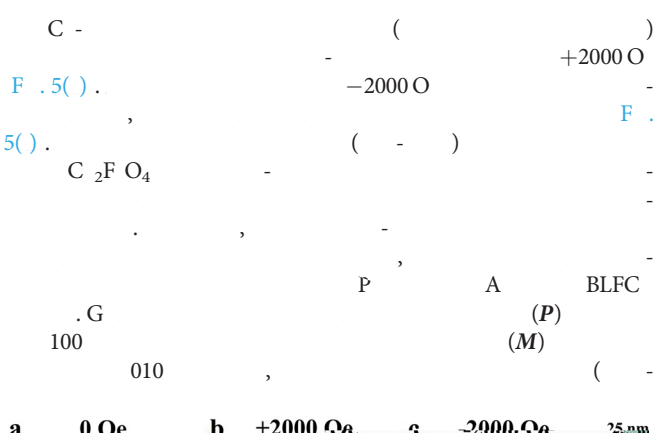


FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe (a), +2000 Oe (b), and -2000 Oe (c). Scale bar is 25 nm.

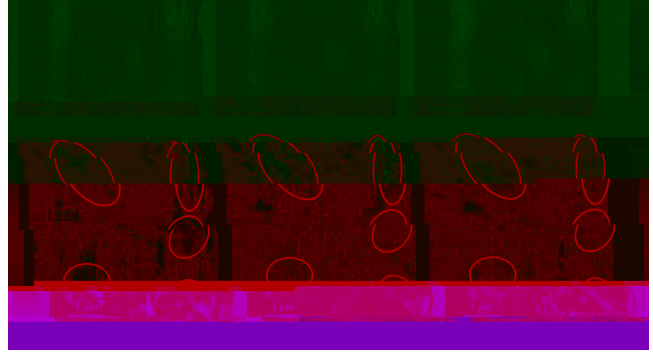


FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe (a), +2000 Oe (b), and -2000 Oe (c). Red circles highlight magnetic features.

$T = P \times M$   
 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 , P 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 P (EM P)  
 P IND54 N EM P  
 EM P E P AME E

DATA AVAILABILITY

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