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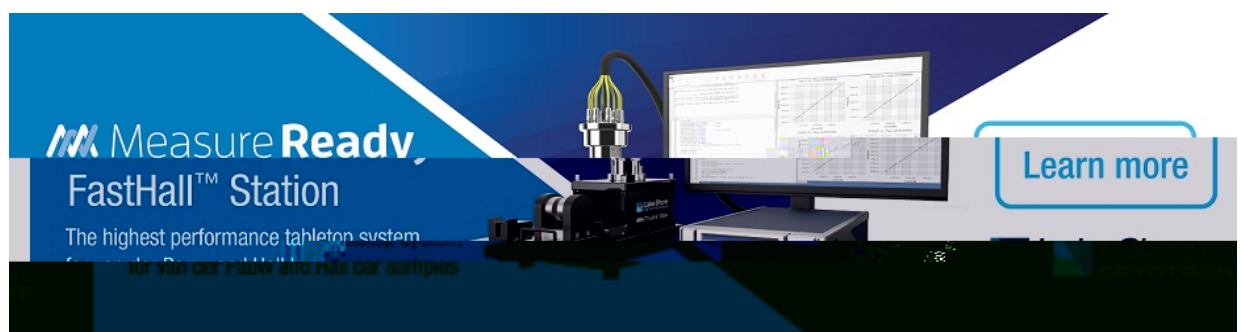
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## ABSTRACT

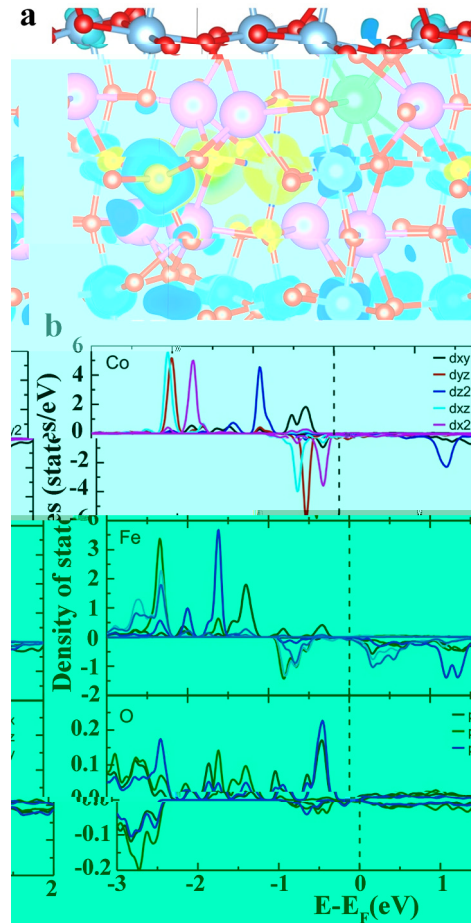
Multiferroic Aurivillius phase ceramics (APCs) are a class of layered perovskite materials with a unique structure consisting of alternating layers of perovskite and Bi<sub>2</sub>O<sub>2</sub> layers. The perovskite layers are composed of B<sub>5.25</sub>L<sub>0.75</sub>F<sub>1</sub>C<sub>3</sub>O<sub>18</sub> units, where B is a divalent cation (Ca, Sr, Ba, Pb, Bi, etc.), L is a monovalent cation (Na, K, Ag, etc.), and F and C are divalent and trivalent cations, respectively. The Bi<sub>2</sub>O<sub>2</sub> layers are composed of Bi<sup>3+</sup> and O<sup>2-</sup> ions. The structure of APCs is highly tunable by varying the chemical composition of the perovskite layers. This tunability allows for the design of materials with a wide range of properties, including ferroelectricity, ferromagnetism, and multiferroicity. In this work, we report on the synthesis and characterization of a new class of APCs with a unique structure consisting of alternating layers of perovskite and Bi<sub>2</sub>O<sub>2</sub> layers. The perovskite layers are composed of B<sub>5.25</sub>L<sub>0.75</sub>F<sub>1</sub>C<sub>3</sub>O<sub>18</sub> units, where B is a divalent cation (Ca, Sr, Ba, Pb, Bi, etc.), L is a monovalent cation (Na, K, Ag, etc.), and F and C are divalent and trivalent cations, respectively. The Bi<sub>2</sub>O<sub>2</sub> layers are composed of Bi<sup>3+</sup> and O<sup>2-</sup> ions. The structure of APCs is highly tunable by varying the chemical composition of the perovskite layers. This tunability allows for the design of materials with a wide range of properties, including ferroelectricity, ferromagnetism, and multiferroicity.

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



**FIG. 3.** (a) Crystal structure of BLFC (a = 0.38 nm, b = 0.38 nm, c = 0.38 nm,  $\beta = 90^\circ$ ), (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 x-axis is E-E<sub>F</sub> (eV) and the y-axis is Density of states (states/eV).

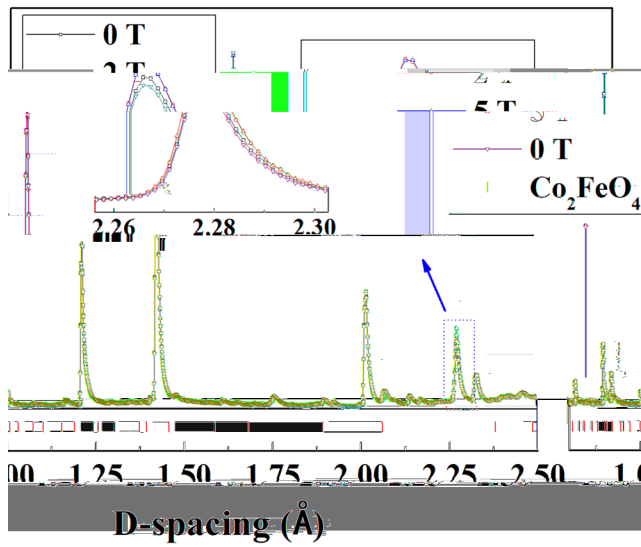


FIG. 4. XRD patterns of  $\text{Co}_2\text{FeO}_4$  at 0 T and 5 T. The inset shows the XRD patterns at 0 T and 5 T. The main plot shows the XRD patterns at 0 T and 5 T. The x-axis is D-spacing (Å) and the y-axis is intensity.

$\text{Co}_2\text{FeO}_4$  (P) (M) 100 010  
 (P) (M) 100 010  
 (P) (M) 100 010

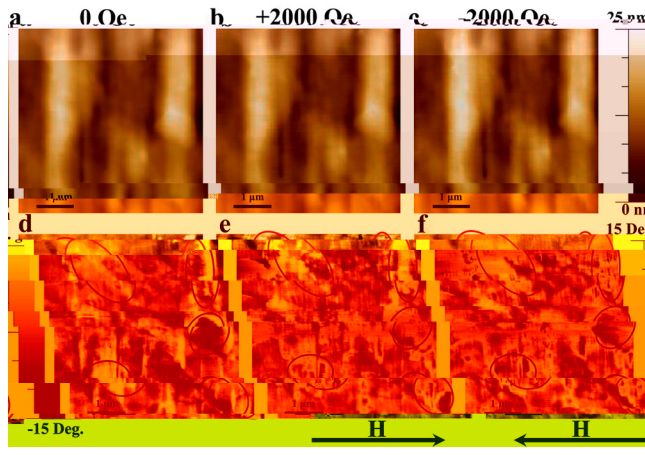


FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe, +2000 Oe, and -2000 Oe. The inset shows the MFM images at 0 Oe, +2000 Oe, and -2000 Oe. The main plot shows the MFM images at 0 Oe, +2000 Oe, and -2000 Oe. The x-axis is D-spacing (Å) and the y-axis is intensity.

$T = P \times M$   
 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 PAME E

DATA AVAILABILITY

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