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Zheng Li, Vladimir Koval , Amit Mahajan, Zhipeng Gao, Carlo Vecchini, Mark Stewart, Markys G. Cain , Kun Tao, Chenglong Jia , Giuseppe Viola, and Haixue Yan 



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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> Graduate School of Materials Science and Engineering, Beijing University of Aeronautics and Astronautics, Beijing 100191, China

<sup>2</sup> Institute of Materials Science and Engineering, A\*STAR, Singapore 117602, Singapore

<sup>3</sup> Department of Materials Science and Engineering, University of California, Los Angeles, California 90095, USA

<sup>4</sup> National Key Laboratory of Materials Physics, Institute of Materials Physics and Chemistry, Chinese Academy of Sciences, Beijing 100883, China

<sup>5</sup> Materials Science Center, University of Cambridge, Cambridge CB2 3RQ, UK

<sup>6</sup> Department of Materials Science and Engineering, University of California, Los Angeles, California 90095, USA

<sup>7</sup> Department of Materials Science and Engineering, Beijing University of Aeronautics and Astronautics, Beijing 100191, China

a) Email: [cjia@ucla.edu](mailto:cjia@ucla.edu)

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

## ABSTRACT

Multiferroic Aurivillius phase (AP) ceramics exhibit room-temperature ferroelectric (FE) and ferromagnetic (FM) behaviors. However, the origin of the room-temperature multiferroic behavior in APs remains unclear. In this work, we study the room-temperature multiferroic behavior in APs with the general formula  $B_{5.25}L_{0.75}F_{1-x}C_xO_{18}$  ( $B = \text{Bi, Pb, Sr, Ba}$ ;  $L = \text{La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Y, Lu}$ ). The room-temperature multiferroic behavior is observed in APs with  $x = 0.5$  and  $x = 0.75$ . The room-temperature multiferroic behavior is attributed to the presence of  $F^{3+}$  and  $C^{3+}$  ions in the AP structure. The room-temperature multiferroic behavior is observed in APs with  $x = 0.5$  and  $x = 0.75$ . The room-temperature multiferroic behavior is attributed to the presence of  $F^{3+}$  and  $C^{3+}$  ions in the AP structure.

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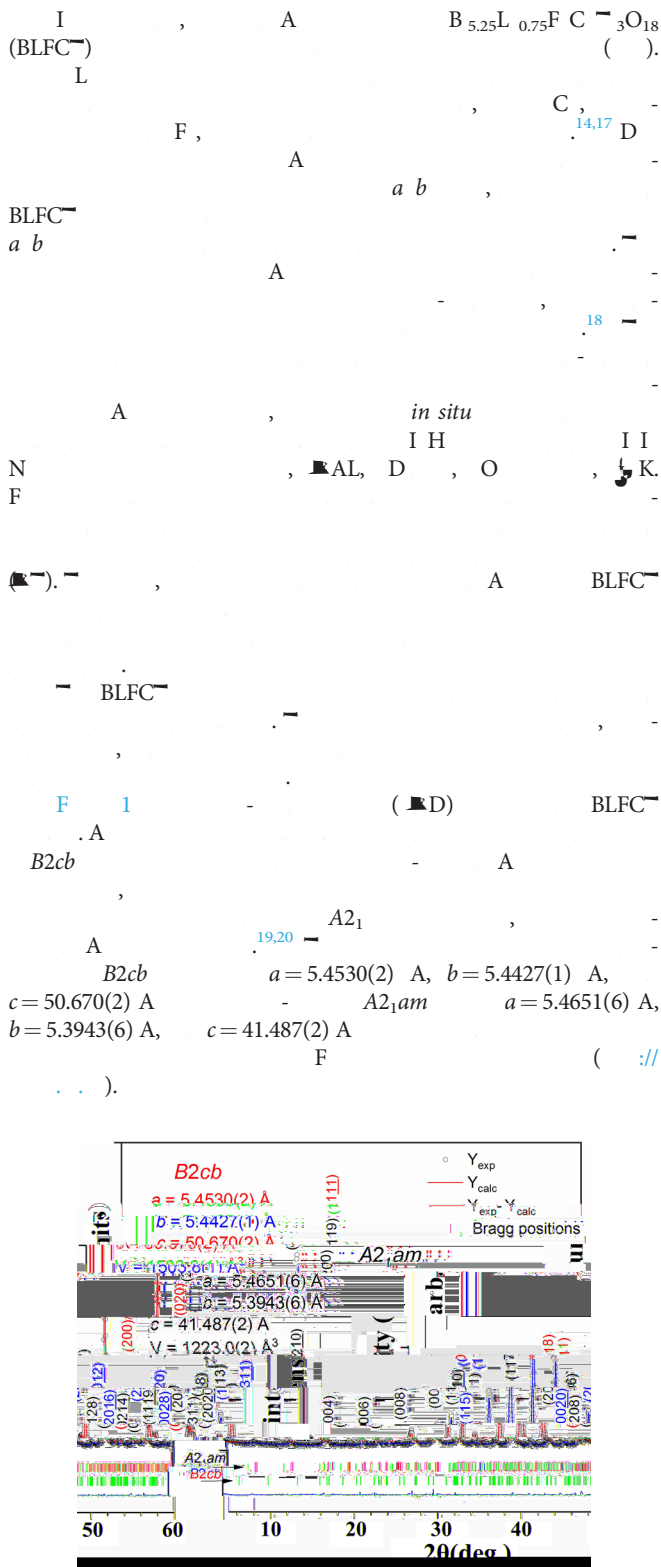


FIG. 1. XRD patterns and unit cell parameters for BLFC.

BLFC  $B_{5.25}L_{0.75}F_1C_2O_{18}$

$B_{5.25}L_{0.75}F_1C_2O_{18}$  (BLFC)  $B_{5.25}L_{0.75}F_1C_2O_{18}$

$B_{5.25}L_{0.75}F_1C_2O_{18}$  (BLFC)  $B_{5.25}L_{0.75}F_1C_2O_{18}$

$B_{5.25}L_{0.75}F_1C_2O_{18}$  (BLFC)  $B_{5.25}L_{0.75}F_1C_2O_{18}$

$B_{5.25}L_{0.75}F_1C_2O_{18}$  (BLFC)  $B_{5.25}L_{0.75}F_1C_2O_{18}$

$B_{5.25}L_{0.75}F_1C_2O_{18}$  (BLFC)  $B_{5.25}L_{0.75}F_1C_2O_{18}$

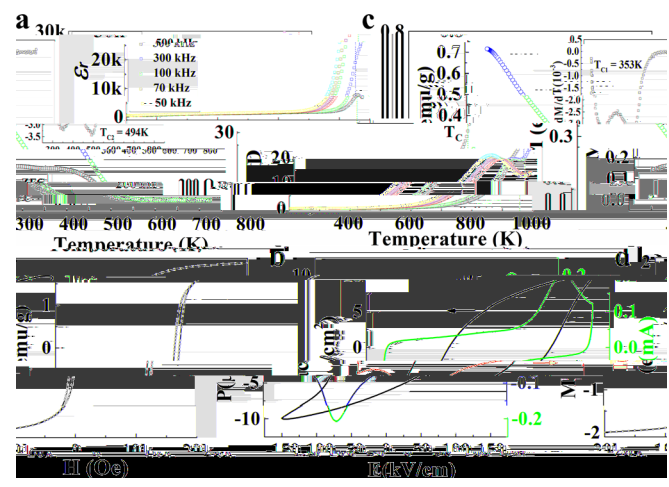


FIG. 2. (a) Raman spectra for BLFC at 494 K. (b) Raman spectra for BLFC at 353 K. (c) Raman spectra for BLFC at different temperatures. (d) Raman spectra for BLFC under different electric fields.

$\sim 494$  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  
 FC  
 $\sim 353$  K  
 $C_2F_4O_4$   
 $C_2F_4O_4$  (460 K)  
 (M)  $C_2F_4O_4$  16 23.5 / .<sup>25</sup> , 1.4 .%  
 $C_2F_4O_4$  0.22 0.32 / , BLFC  
 $M = 1.85$  / ,  $F = 2$  ( ). I  
 M H  
 $\sim 425$  K 1.58 / .  
 0.27 / , ED  
 BLFC  
 A  
 $F = 3$   
 $(DF)$   $F^{3+} O C^{3+}$  *ab initio*  
 $(A)$   $H$   
 $\mu_F = 2$   $\mu_C = 3$   $F$   $C$  ,  
 $(GGA)+\mu$  . I  
 BLFC  
 $F = 3$  ( ),  $F^{3+}$   $C^{3+}$  (3.1 2.1  $\mu_B$  / , ),  
 $(0.1 \mu_B / )$  .  
 $F O_6$   $C O_6$   
 $F/C$  .  $F = 3$  ( ).  
 $F$   $O$  /  
 $F^{3+}$   $C^{3+}$  ,  
 $(\dots)$  ,  
 $(\dots)$  ,  
 $E_{FM} - E_{AFM}$   
 $= -144.1$  .  
 $H$  , (FM)  
 $43.5$  ( , 504.6 K), FM  
 $\sim 1$  FC/FC .  $F = 2$  ( )  
 $a b$   
 $010$   
 BLFC  
 $F = 4$   
 I  
 $399$  O .  
 $F$  .  
 $F$  -

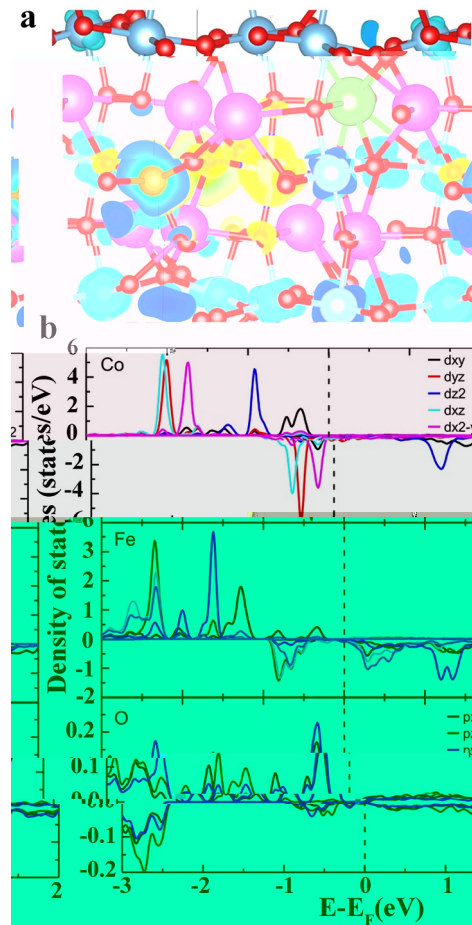


FIG. 3. (a) Crystal structure of BLFC. (b) Density of states (DOS) for Co, Fe, and O atoms. The DOS is calculated using the GGA+U method with  $U = 0.005$  eV. The legend indicates the contributions from different orbitals: dxy (black), dyz (red), dz2 (blue), dxz (green), and dx2-y2 (magenta).

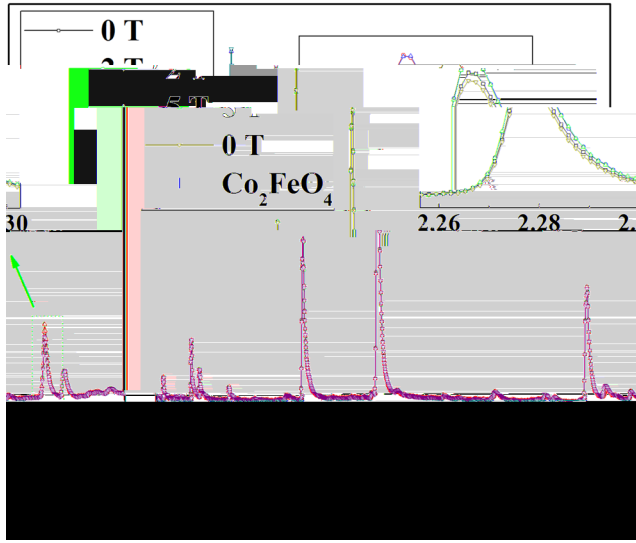


FIG. 4. XRD patterns of  $\text{Co}_2\text{FeO}_4$  at 0 T and 2 T. The inset shows the magnified view of the peaks at 2.26 and 2.28 Å.

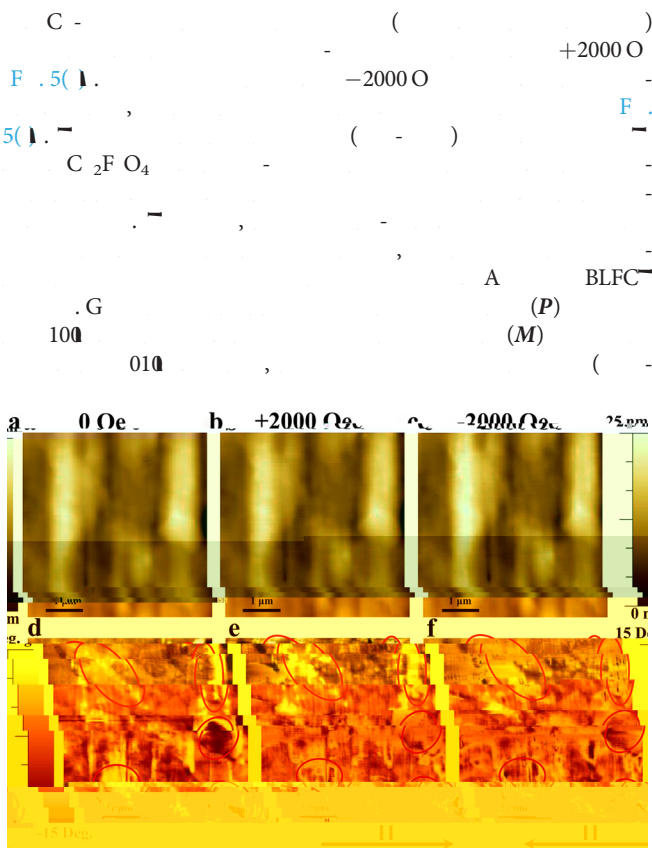


FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe, +2000 Oe, and -2000 Oe. The red circles in the bottom row highlight the magnetic domains.

$T = P \times M$   
 $\text{BLFC}^-$   
 $\text{F}^{3+} \text{O} \text{F}^{3+}$   
 $\text{C}^{3+} \text{O} \text{C}^{3+}, \text{F}^{3+} \text{O} \text{C}^{3+}$   
 $\text{A}, \text{C} / \text{F}$   
 $\text{EM} \text{ (ED)}$   
 $\text{BLFC}^-$   
 $\text{D. M}, \text{D. K}$   
 $\text{D. H. I. I. N}$   
 $\text{D. O. K.}$   
 $\text{A. E. D. F.}$   
 $\text{G. A. A. (G. N. 2/0038/20), C. (G. N. K2015-0602006), N. FC (G. N. 11474138 11834005). A. E. M. (EM)}$   
 $\text{IND54 N. EM}$   
 $\text{EM. E. AME}^- \text{ E}$

DATA AVAILABILITY

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