



# Room-temperature multiferroic behavior in layer-structured Aurivillius phase ceramics

Cite as: Appl. Phys. Lett. **117**, 052903 (2020); <https://doi.org/10.1063/5.0017781>

Submitted: 09 June 2020 . Accepted: 25 July 2020 . Published Online: 07 August 2020

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



View Online



Export Citation



CrossMark

## ARTICLES YOU MAY BE INTERESTED IN

[Intrinsic piezoelectricity in \(K,Na\)NbO<sub>3</sub>-based lead-free single crystal: Piezoelectric anisotropy and its evolution with temperature](#)

Applied Physics Letters **117**, 052904 (2020); <https://doi.org/10.1063/5.0012124>

[Current-induced bulk magnetization of a chiral crystal CrNb<sub>3</sub>S<sub>6</sub>](#)

Applied Physics Letters **117**, 052408 (2020); <https://doi.org/10.1063/5.0017882>

[Magnetic transition behavior and large topological Hall effect in hexagonal Mn<sub>2-x</sub>Fe<sub>1+x</sub>Sn \(x = 0.1\) magnet](#)

Applied Physics Letters **117**, 052407 (2020); <https://doi.org/10.1063/5.0011570>



**Measure Ready**  
**FastHall™ Station**

The highest performance tabletop system  
for van der Pauw and Hall bar samples

[Learn more](#)

LakeShore  
CRYOTRONICS

# Room-temperature multiferroic behavior in layer-structured Aurivillius phase ceramics

Cite as: Appl. Phys. Lett. **117**, 052903 (2020); doi: 10.1063/5.0017781

Submitted: 9 June 2020 · Accepted: 25 July 2020 ·

Published Online: 7 August 2020 · Corrected: 11 August 2020



View Online



Export Citation



CrossMark

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 100083, 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 Physics, Chinese Academy of Sciences, Beijing 100080, China

<sup>5</sup> MRL, NIST, Gaithersburg, Maryland 20899, USA

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

<sup>7</sup> School of Materials Science and Engineering, Beijing University of Aeronautics and Astronautics, Beijing 100083, China

a)Email: jia@ucla.edu

b)Author to whom correspondence should be addressed: yan@ucla.edu

## ABSTRACT

Multiferroic Aurivillius phase ceramics (APCs) are a class of layered perovskite materials with a unique structure consisting of alternating layers of perovskite and BiFeO<sub>3</sub> (BFO) layers. The BFO layers are known for their ferroelectric (FE) and antiferromagnetic (AFM) properties, while the perovskite layers provide structural stability. In this study, we investigate the room-temperature multiferroic behavior in layer-structured Aurivillius phase ceramics. We focus on the structure and properties of Bi<sub>5.25</sub>La<sub>0.75</sub>FeC<sub>3</sub>O<sub>18</sub> (B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub>), which is a member of the Aurivillius phase family. The structure of B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub> is characterized by the presence of Bi<sup>3+</sup>, La<sup>3+</sup>, Fe<sup>3+</sup>, and O<sup>2-</sup> ions. The BFO layers are known for their ferroelectric (FE) and antiferromagnetic (AFM) properties, while the perovskite layers provide structural stability. We investigate the room-temperature multiferroic behavior in layer-structured Aurivillius phase ceramics. We focus on the structure and properties of Bi<sub>5.25</sub>La<sub>0.75</sub>FeC<sub>3</sub>O<sub>18</sub> (B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub>), which is a member of the Aurivillius phase family. The structure of B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub> is characterized by the presence of Bi<sup>3+</sup>, La<sup>3+</sup>, Fe<sup>3+</sup>, and O<sup>2-</sup> ions. We investigate the room-temperature multiferroic behavior in layer-structured Aurivillius phase ceramics. We focus on the structure and properties of Bi<sub>5.25</sub>La<sub>0.75</sub>FeC<sub>3</sub>O<sub>18</sub> (B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub>), which is a member of the Aurivillius phase family. The structure of B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub> is characterized by the presence of Bi<sup>3+</sup>, La<sup>3+</sup>, Fe<sup>3+</sup>, and O<sup>2-</sup> ions.

Published under license by AIP Publishing. <https://doi.org/10.1063/5.0017781>

Multiferroic Aurivillius phase ceramics (APCs) are a class of layered perovskite materials with a unique structure consisting of alternating layers of perovskite and BiFeO<sub>3</sub> (BFO) layers. The BFO layers are known for their ferroelectric (FE) and antiferromagnetic (AFM) properties, while the perovskite layers provide structural stability. In this study, we investigate the room-temperature multiferroic behavior in layer-structured Aurivillius phase ceramics. We focus on the structure and properties of Bi<sub>5.25</sub>La<sub>0.75</sub>FeC<sub>3</sub>O<sub>18</sub> (B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub>), which is a member of the Aurivillius phase family. The structure of B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub> is characterized by the presence of Bi<sup>3+</sup>, La<sup>3+</sup>, Fe<sup>3+</sup>, and O<sup>2-</sup> ions. We investigate the room-temperature multiferroic behavior in layer-structured Aurivillius phase ceramics. We focus on the structure and properties of Bi<sub>5.25</sub>La<sub>0.75</sub>FeC<sub>3</sub>O<sub>18</sub> (B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub>), which is a member of the Aurivillius phase family. The structure of B<sub>5.25</sub>L<sub>0.75</sub>FC<sub>3</sub>O<sub>18</sub> is characterized by the presence of Bi<sup>3+</sup>, La<sup>3+</sup>, Fe<sup>3+</sup>, and O<sup>2-</sup> ions.

I, A, B<sub>5.25</sub>L<sub>0.75</sub>F<sub>1.0</sub>C<sub>3.0</sub>O<sub>18</sub> (BLFC) P<sub>a</sub> La<sub>a</sub> F, A, C, D<sup>14,17</sup> BLFC a b A a b A A<sup>18</sup> A, in situ I H, O, K. N F AL, D, O, K. ( ). A BLFC BLFC P. A B<sub>2</sub>cb A A<sub>2</sub>am<sup>19,20</sup> B<sub>2</sub>cb a = 5.4530(2) Å, b = 5.4427(1) Å, c = 50.670(2) Å A<sub>2</sub>am a = 5.4651(6) Å, b = 5.3943(6) Å, c = 41.487(2) Å F P ( // . . . ).

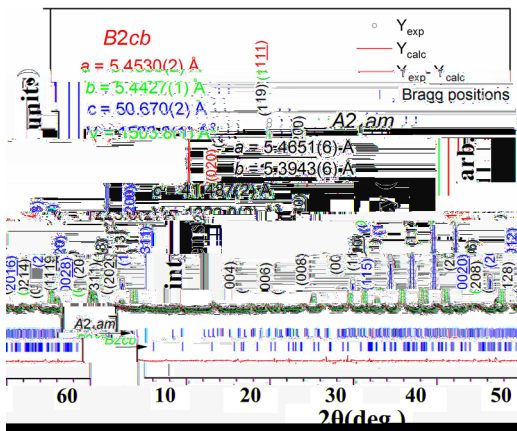


FIG. 1. - B

BLFC A = 4 = 5 . N D' BLFC F 1 EM (a-b a ) M , a F . 1 a . a 1.4 . %, D. ED a a (F . 2 a a 1) a a a a F, C, O, a a a a C<sub>2</sub>F O<sub>4</sub> a a a A B<sub>5</sub>F<sub>0.5</sub>C<sub>0.5</sub>O<sub>15</sub>.<sup>16</sup> BLFC a a (50, 70 100, 300, a 500 H ). a FE T BLFC . H , BLFC BLFC B<sub>6</sub> F<sub>2</sub> O<sub>18</sub> a (a 973 K).<sup>13</sup> F 2( ) P-E a I-E BLFC a a I-E P a .<sup>21,22</sup> BLFC 10 μC/ . F 2( ) (FC) a a a (FC) a a 200 O BLFC . a BLFC

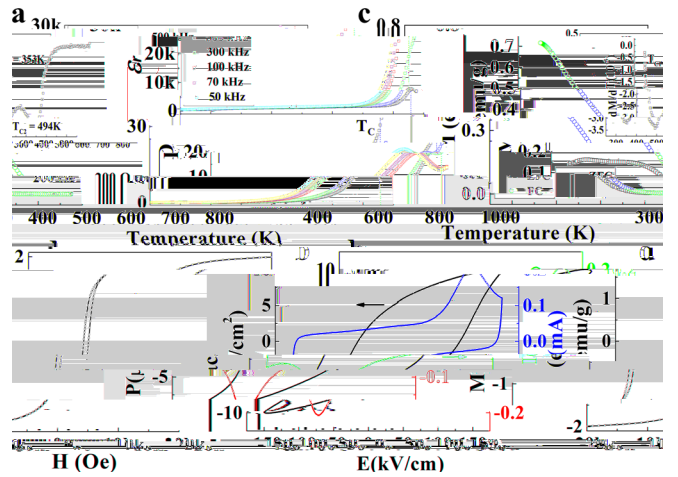


FIG. 2. ( ) ( ) B P ( ) P B P B = 200 ( ) M/ 300 B . ( )

$\sim 494$  K (M/),  
 $B_6FC_{3O_{18}}$  (526 K).<sup>23</sup>  
 BLFC  
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$  (.  
 ED  
 $FC$   $\sim 353$  K  
 $C_2FO_4$  (460 K)  $16.235 / .25$ ,  $0.22 0.32 /$ ,  $1.4 .\%$   
 $C_{2-} F O_4$  (M)  $C_2FO_4$   $16,25$   
 $M = 1.85 / , F . 2( ) . I$  BLFC  
 $M H$   
 $2 (F . 3)$   $425$  K  $1.58 / .$   $0.27 /$ , ED  
 $BLFC$   
 $F 3$   $F^{3+} O C^{3+}$   
 $(DF)$   $ab initio$   
 $(A P)$   $H$   
 $F = 2$   $C = 3$   $F$   $C$ ,  
 $(GGA)$   $I$   
 $BLFC$   
 $F . 3(a)$ ,  $F^{3+}$   $C^{3+}$  ( $3.1$   $2.1 \mu_B/a$ ),  
 $O$   $0.1 \mu_B/a$ ).  
 $F O_6$   $C O_6$   
 $F / C$   
 $F$   $O$   $F . 3( )$   
 $F^{3+}$   $C^{3+}$   
 $(. , )$   $(. , )$   
 $E_{FM} - E_{AFM}$   
 $= -144.1$   
 $H$  (FM)  
 $43.5$  ( $\dots, 504.6$  K), FM  
 $FC/FC$   $F . 2( )$   
 $a b$   
 $010$   
 $F 4$   
 $BLFC$   $I$   
 $399 O$   $F$   
 $5( ) . A$   $P F M$   $BLFC$   $F$

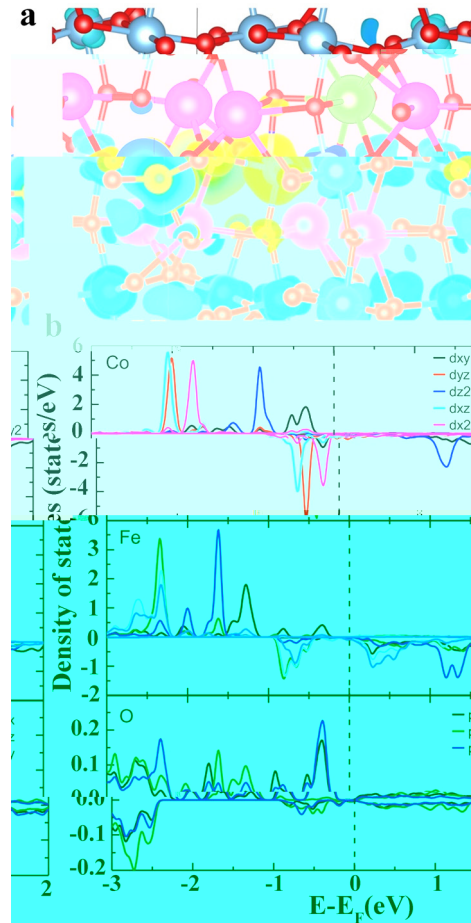


FIG. 3. (a) Crystal structure of BLFC (space group  $R\bar{3}m$ ) showing layers of  $Co^{2+}$ ,  $Fe^{3+}$ , and  $O^{2-}$  ions. (b) Calculated density of states (DOS) for  $Co$ ,  $Fe$ , and  $O$  atoms. The DOS is shown in units of states/eV versus energy  $E - E_f$  (eV). The contributions from different orbitals ( $d_{xy}$ ,  $d_{yz}$ ,  $d_{z^2}$ ,  $d_{xz}$ , and  $d_{x^2-y^2}$ ) are indicated by different colors. The Fermi level  $E_f$  is set at 0 eV.

$N$   
 $I$   $F . 4$   
 $A$   $(0 1 20)$   
 $2 < H < 5$ ,  
 $M H$   $F . 2( )$   $3. F$ ,  
 $F 5$   
 $BLFC$   $P$   $F M$   
 $P F M$   $BLFC$ ,  
 $5( ) . A$   $F$

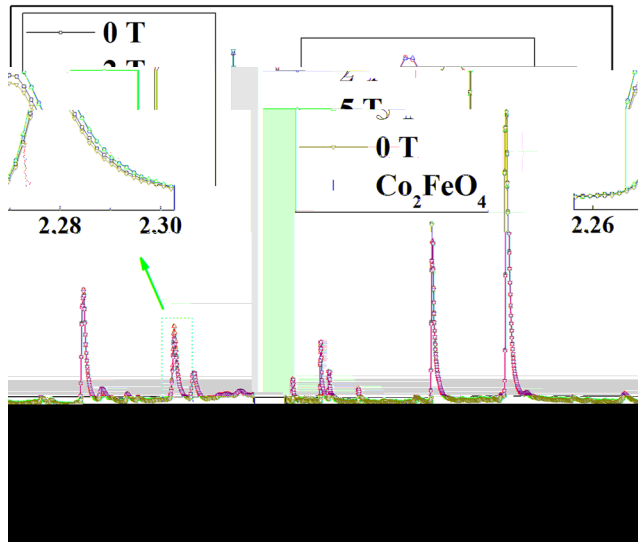


FIG. 4. XRD patterns of  $\text{Co}_2\text{FeO}_4$  at 0 T and 5 T. The inset shows the zoomed-in view of the 0 T peak at  $2\theta = 2.28$ – $2.30$ .

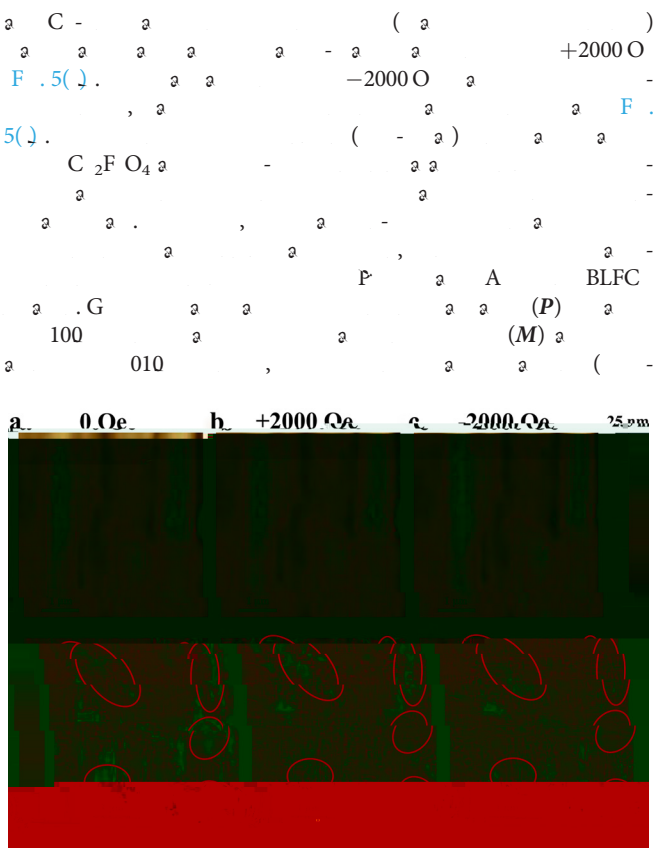


FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe (a), +2000 Oe (b), and -2000 Oe (c). The scale bar is 25 nm.

$T = P \times M$   
 BLFC  
 $\text{F}^{3+} \text{O} \text{F}^{3+}$   
 $\text{C}^{3+} \text{O} \text{C}^{3+}, \text{F}^{3+} \text{O} \text{C}^{3+}$   
 $\text{A} \text{C} / \text{F}$   
 EM  
 (ED)  
 BLFC  
 D. Ma, P. Ma, D. K.  
 D. H. I I N  
 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

REFERENCES

1. E. N. D. Ma, J. F., *Nature* **442**, 759 (2006).
2. N. A., *Nature Materials* **6**, 21 (2007).
3. J. Ma, J. H., L., C., *Nature Materials* **23**, 1062 (2011).
4. L. F. H., O. C., J. B., J. L., C. H., H., O. G., D. C. L., H., K., A. J. B., *Nature Materials* **26**, 2111 (2016).
5. N. A. H., *J. Phys. Chem. B* **104**, 6694 (2000).
6. B. A., M. : II.  $\text{B}_4\text{O}_3\text{O}_{12}$ , A. K. **1**(58), 499–512 (1949).
7. A., G. K., M. M. K., *J. Phys. Chem. B* **11**, 3335 (1999).
8. N., P., G., K., *Nature Materials* **108**, 194 (2004).
9. L. K., M., A. A., N. D., N. P., M., E. P., D. J., *J. Am. Chem. Soc.* **96**, 2339 (2013).
10. L., J. Ma, G., K., A. Ma, L., C. J., C. Na, H., *ACS Nano* **45**, 14049 (2016).
11. J. F., *NPG Asia Materials* **5**, 72 (2013).
12. A., B., C. E., *Phys. Rev. B* **90**, 214109 (2014).
13. J. B. L., P. H., G. H., G. L., J. L., J. C., J. K. L., *Phys. Rev. Lett.* **96**, 222903 (2010).
14. M., C., L., *Phys. Rev. Lett.* **95**, 082901 (2009).
15. L., J., L., J. D., *Phys. Rev. Lett.* **101**, 122402 (2012).

- <sup>16</sup>M. P. ... P. C. ..., M. B. ..., A. P. B. ..., J. P. H. ..., K. ..., L. K. ..., M. P. ..., C. ..., H. K. ..., A. J. B. ..., *J. A. P.* **112**, 073919 (2012).
- <sup>17</sup>J. L. ..., H. ..., M. J. ..., K. ..., P. ..., *J. A. P.* **102**, 104107 (2007).
- <sup>18</sup>M. G. C. ..., *Characterisation of Ferroelectric Bulk Materials and Thin Films* (..., 2014), ... 2.
- <sup>19</sup>..., K. ..., J. M. ..., G. ..., K. ..., C. J. ..., G. ..., H. ..., A. M. ..., J. C. ..., M. C. ..., I. A. ..., C. N. ..., C. J. ..., H. ..., *J. M. C. C.* **6**, 2733 (2018).
- <sup>20</sup>..., K. ..., I. ..., G. ..., M. ..., C. J. ..., H. ..., *J. P. C.* **122**, 15733 (2018).
- <sup>21</sup>L. J. ..., F. L. ..., ..., *J. A. C.* **97**, 1 (2014).
- <sup>22</sup>H. ..., F. I. ..., G. ..., H. N. ..., H. ..., J. ..., G. ..., M. J. ..., *J. A. D.* **1**, 107 (2011).
- <sup>23</sup>J. ..., L. ..., L. ..., ..., J. D. ..., ..., *A. P. L.* **101**, 012402 (2012).
- <sup>24</sup>B. ..., J. ..., J. C. ..., ..., L. ..., ..., J. D. ..., ..., *A. P. L.* **104**, 062413 (2014).
- <sup>25</sup>I. P. M. ..., ..., N. B. ..., ..., **11**, 719 (2009).