




# 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](#)

Lake Shore  
CRYOTRONICS

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

Cite as: Appl. Phys. Lett. **117**, 052903 (2020); doi: [10.1063/5.0017781](https://doi.org/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>Gemological Institute, China University of Geosciences, Wuhan 430074, China

<sup>2</sup>Institute of Materials Research, Slovak Academy of Sciences, Watsonova 47, Kosice O4001, Slovakia

<sup>3</sup>School of Engineering and Materials Science, Queen Mary, University of London, London E1 4NS, United Kingdom

<sup>4</sup>National Key Laboratory of Shock Wave and Detonation Physics Institute of Fluid Physics, China Academy of Engineering Physics, Mianyang 621900, China

<sup>5</sup>National Physical Laboratory, Hampton Road, Teddington TW11 0LW, United Kingdom

<sup>6</sup>Electrosiences Ltd, Farnham, Surrey GU9 9QT, United Kingdom

<sup>7</sup>School of Physical Science and Technology, Lanzhou University, Lanzhou 730000, China

<sup>a)</sup>Email: [cjlja@lzu.edu.cn](mailto:cjlja@lzu.edu.cn)

<sup>b)</sup>Author to whom correspondence should be addressed: [h.x.yan@qmul.ac.uk](mailto:h.x.yan@qmul.ac.uk)

## ABSTRACT

M

... H ... A ...  
 ... D ... H ... A ...  $B_{5.25}L_{0.75}F_2C_3O_{18}$  ...  
 ... A ... *in situ* ...  
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$  ...  
 ... A ... C / F ...

Published under license by AIP Publishing. [:// . /10.1063/5.0017781](https://doi.org/10.1063/5.0017781)

M (FM) (FE) ...  $B_5F_2O_{15}$  ( = 4)  $B_6F_2O_{18}$  ...  
 ...  $B_4O_{12}$  ...  $B_5F_{0.5}C_{0.5}O_{15}$  ...  
 ...  $(B_2O_2)^{2+}(A_{-1}B O_3 +1)^{2-}$  ...  
 ...  $B F O_3$  ...  $B F O_3$  ...  
 ...  $B F O_3$  ...  $B F O_3$  ...

$B_{5.25}F_{0.75}C_{-3}O_{18}$   
 $(BLFC)$   
 $L$   
 $F$ ,  $A$ ,  $C$ ,  $D$   
 $a$ ,  $b$   
 $BLFC$   
 $a$ ,  $b$   
 $A$   
 $in situ$   
 $I$ ,  $H$ ,  $I$ ,  $I$   
 $N$ ,  $F$ ,  $A$ ,  $L$ ,  $D$ ,  $O$ ,  $K$   
 $A$ ,  $BLFC$   
 $BLFC$   
 $F$ ,  $1$ ,  $(D)$ ,  $BLFC$   
 $A$ ,  $B2cb$ ,  $A$   
 $A$ ,  $A_{21}$   
 $B2cb$ ,  $a = 5.4530(2)$  Å,  $b = 5.4427(1)$  Å,  
 $c = 50.670(2)$  Å,  $A_{21}am$ ,  $a = 5.4651(6)$  Å,  
 $b = 5.3943(6)$  Å,  $c = 41.487(2)$  Å  
 $F$  ( // )

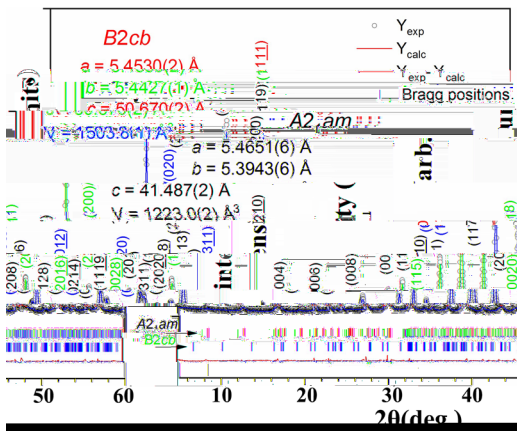


FIG. 1. XRD patterns of B2cb and A21am phases.

$BLFC$ ,  $F$ ,  $1$ ,  $EM$ ,  $(a-b)$ ,  $N$ ,  $D$ ,  $M$ ,  $F$ ,  $1$ ,  $1.4$  %,  $(F$ ,  $2$   
 $BLFC$ ,  $D$ ,  $ED$ ,  $1$ ,  $F$ ,  $C$ ,  $O$ ,  $C$ ,  $2F$ ,  $O_4$ ,  $A$ ,  $B_{5F_{0.5}C_{0.5}O_{15}}$ ,  $16$   
 $BLFC$ ,  $(50, 70, 100,$   
 $300, 500$  H),  $1060$  K,  $FE$ ,  $T$ ,  $BLFC$ ,  $H$ ,  $B_6F_3O_{18}$ ,  $BLFC$ ,  $2()$ ,  $P-E$ ,  $I-E$ ,  $I-E$ ,  $21,22$   
 $BLFC$ ,  $10 \mu C /$ ,  $F$ ,  $2()$ ,  $(FC)$ ,  $(FC)$ ,  $200$  O,  $BLFC$ ,  $BLFC$

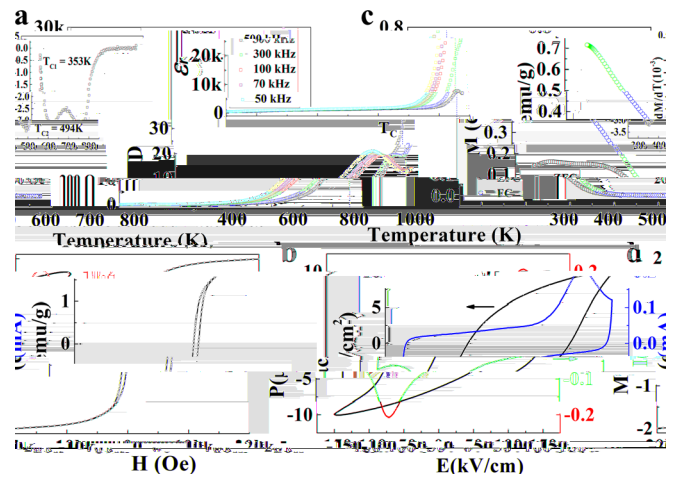


FIG. 2. Temperature dependence of dielectric loss (a), piezoelectric coefficient (b), and polarization (c) for BLFC.

$B_6F_2C_{18}O_{18}$  (526 K).<sup>23</sup>  $\sim 494$  K  
 BLFC  $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$  ( . )<sup>24</sup>  $\sim 353$  K  
 ED FC  $\sim 2 \sim 353$  K  
 $C_2F_4O_4$  (460 K)  $\sim 2$   $\sim 16,25$   
 (M)  $C_2F_4O_4$  1.4 %  
 $16 \text{ } 23.5 \text{ } / \text{ } .^{25}$  , 0.22 0.32 / , BLFC  
 $C_2F_4O_4$  M = 1.85 / , F . 2 ( ) . I  
 M H  
 $\sim 2$  (F . 3) .  $\sim 1$   
 425 K 1.58 / .  $\sim 0.27$  / , ED  
 BLFC  
 A  
 $F^{3+} O C^{3+}$  *ab initio*  
 (DF) ( A ) F 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 F O - /  $F . 3$  ( ) .  
 $F^{3+} C^{3+}$  ,  
 ( . , ) ( . , )  
 $E_{FM} - E_{AFM}$   
 $= -144.1$  .  
 H , (FM)  
 43.5 ( . , 504.6 K), FM  
 $\sim 1$  FC/FC  $F . 2$  ( ) .  
 a b  
 070  
 $F . 4$   
 BLFC . I  
 . I  
 . I  
 . N  
 . I  $F . 4$   
 A (0 1 20)  
 $2^-$  F  
 (  $2^- < H < 5^-$  ) ,  
 2^- , M H  $F . 2$  ( ) 3. F ,  
 $F . 5$   
 BLFC F M  
 FM BLFC , 399 O .  
 $5$  ( ) . A F .  
 F -

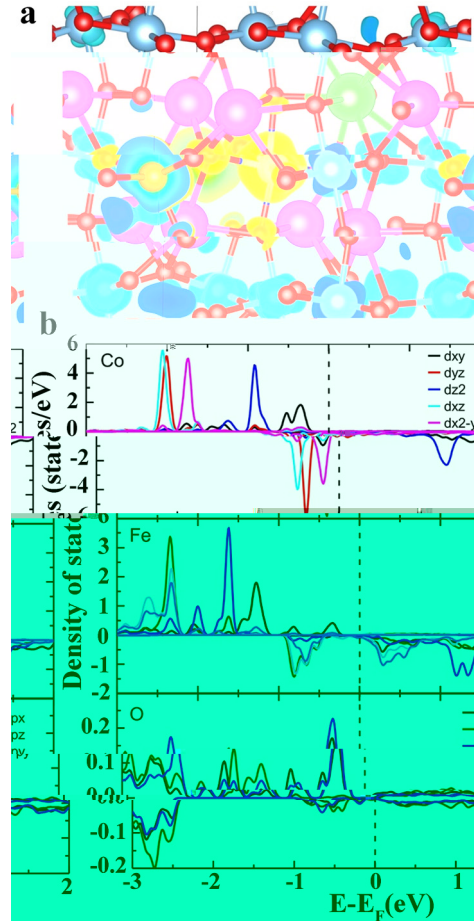


FIG. 3. (a) Crystal structure of BLFC. (b) Density of states (DOS) for Co, Fe, and O atoms. The DOS is calculated using the GGA+U method with U = 0.005 eV. The x-axis is E - E<sub>F</sub> (eV) and the y-axis is Density of states (states/eV).

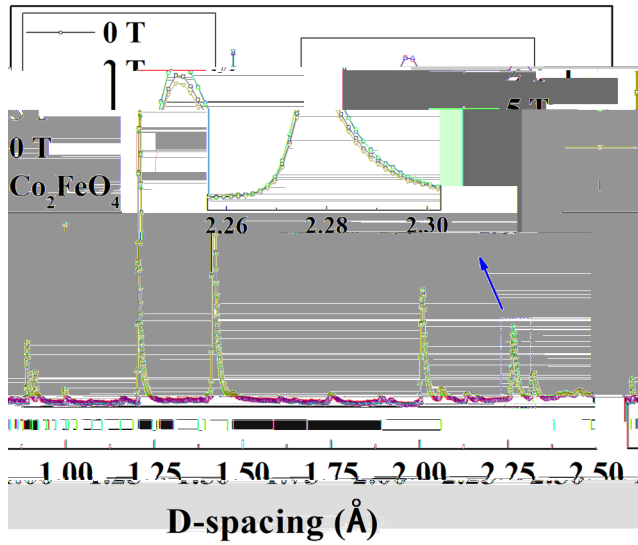


FIG. 4. XRD patterns of  $\text{Co}_2\text{FeO}_4$  at 0 T and 5 T. The inset shows the schematic of the sample and measurement setup.

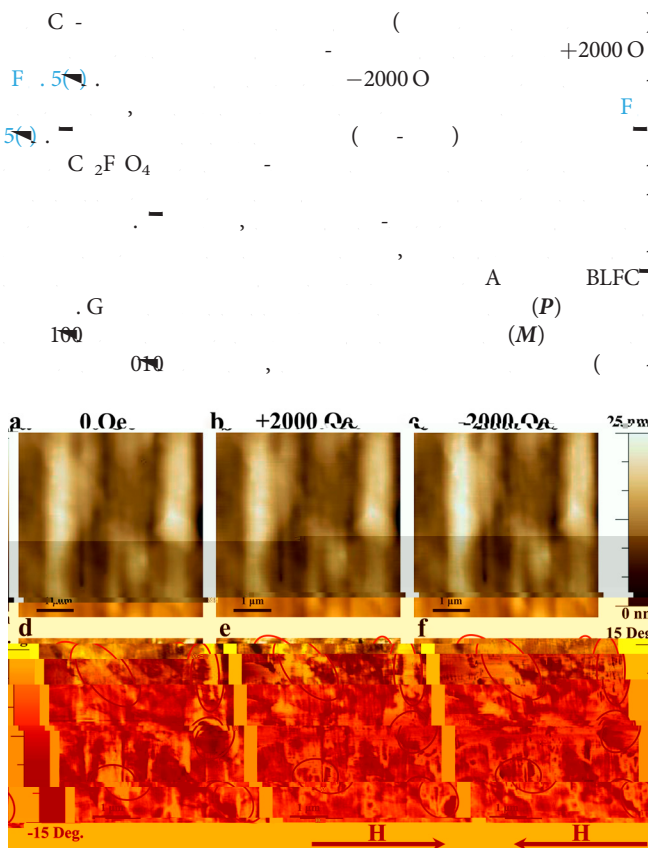


FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe, +2000 Oe, and -2000 Oe. The top row shows MFM images and the bottom row shows phase images. Arrows labeled 'H' indicate the magnetic field direction.

$T = P \times M$   
 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 U AME E

DATA AVAILABILITY

REFERENCES

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

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