Applied Physics Letters

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

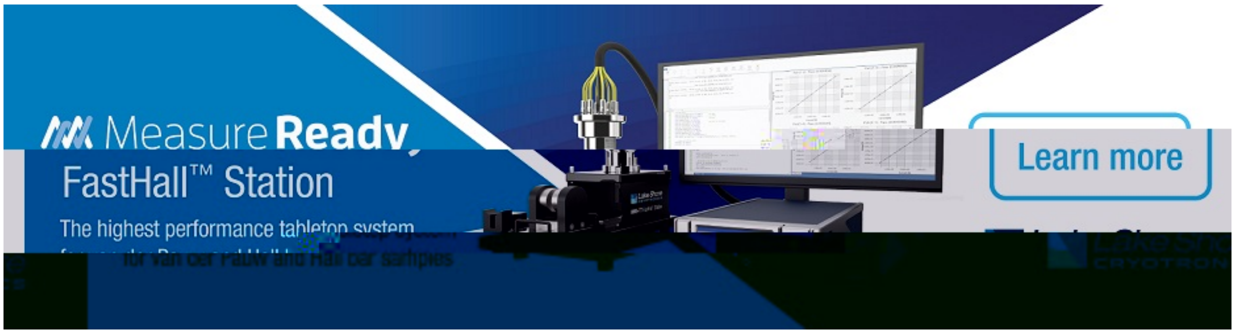
ARTICLES YOU MAY BE INTERESTED IN

Intrinsic piezoelectricity in (K,Na)NbO_3-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 $CrNb_3S_6$
Applied Physics Letters **117**, 052408 (2020); https://doi.org/10.1063/5.0017882

Magnetic transition behavior and large topological Hall effect in hexagonal $Mn_{2-x}Fe_{1+x}Sn$ ($x = 0.1$) magnet
Applied Physics Letters **117**, 052407 (2020); https://doi.org/10.1063/5.0011570

AIP Publishing



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

Zheng Li,[1] Vladimir Koval,[2] Amit Mahajan,[3] Zhipeng Gao,[4] Carlo Vecchini,[5] Mark Stewart,[5] Markys G. Cain,[6] Kun Tao,[7] Chenglong Jia,[7,a)] Giuseppe Viola,[3] and Haixue Yan[3,b)]

AFFILIATIONS

[1] G I , C f G , 430074, C
[2] I f M , A f , 47, K 04001,
[3] f E M , M , f L , L E1 4N , K
[4] N K L f D I f F , C A f E , M 621900, C
[5] N L , H , 11 0L , K
[6] E L , F , G 9 9 , K
[7] f , L , L 730000, C

[a)] Email: @ . .
[b)] Author to whom correspondence should be addressed: . . @ . .

ABSTRACT

M . H , - - . , - A , . D . H , A $B_{5.25}L_{0.75}F\ C\ _3O_{18}$. - A , *in situ* . F $^{3+}$ O F $^{3+}$, C $^{3+}$ O C $^{3+}$, F $^{3+}$ O C $^{3+}$ - . - . A , C /F - , - .

M (FE) (FM) , , .[1–4] H , - - .[5] , - A $(B_2O_2)^{2+}(A_{-1}B\ O_{3+1})^{2-}$ (A -, -, 12- - , B - - ,)[6] B- - B F O_3 - .[7–11]

A $B_5F\ _3O_{15}$ ($= 4$) $B_6F_2\ _3O_{18}$ ($= 5$), $B_4\ _3O_{12}$ B F O_3, FE FM .[12,13] B - C F B- , $B_5F_{0.5}C_{0.5}\ _3O_{15}$ ($= 4$) $B_6F\ C\ _3O_{18}$ ($= 5$) - .[14,15] H , D - .[16] , - A .

I , A B $_{5.25}$L $_{0.75}$F C $_3$O$_{18}$ (BLFC) ( ). L , C , F , [14,17] D A a b , BLFC a b . A - - , [18] - - A , *in situ* I H I I N , AL, D , O , K. F - ( ). , A BLFC

. BLFC . , - , .

F 1 - (D) BLFC . A *B2cb* - A , $A2_1$, - A [19,20] . - *B2cb* $a = 5.4530(2)$ A, $b = 5.4427(1)$ A, $c = 50.670(2)$ A - $A2_1am$ $a = 5.4651(6)$ A, $b = 5.3943(6)$ A, $c = 41.487(2)$ A F (:// . .).

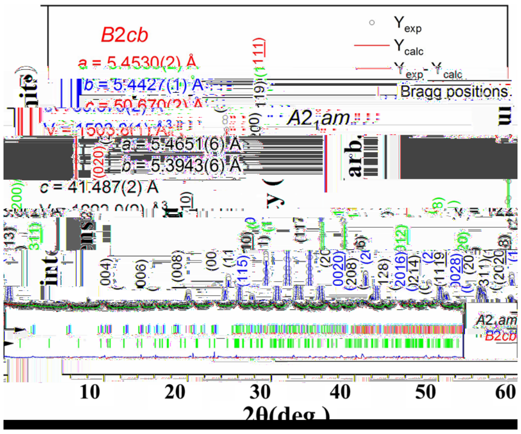


FIG. 1. - w .

BLFC - A = 4 = 5 . N D' . F 1 EM - BLFC . - (a–b) . M , F . 1 . - 1.4 . %, D. ED (F . 2 1) F , C , O, . , C $_2$F O$_4$ - .

- A B $_5$F $_{0.5}$C $_{0.5}$ $_3$O$_{15}$.[16] F 2( ) BLFC (50, 70 100, 300, 500 H). 1060 K FE T BLFC . H , BLFC B $_6$ $_3$F $_2$O$_{18}$ (973 K).[13] F 2( ) P–E I–E BLFC . . I–E [21,22] , . BLFC 10 μC/ ^{2}. F 2( ) - (FC) - (FC) 200 O BLFC . BLFC

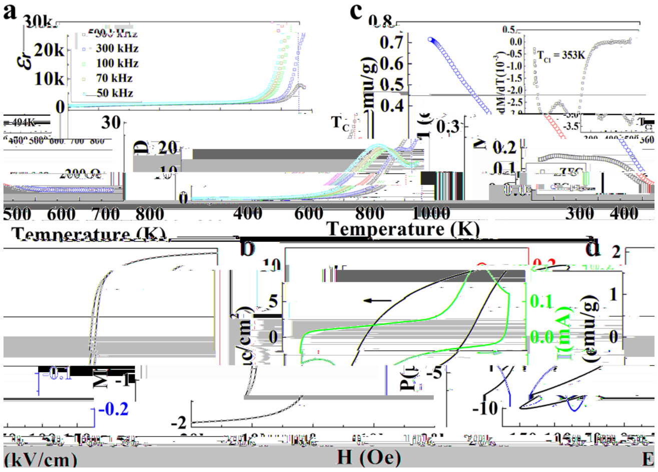


FIG. 2. ( ) = 200 . W M/ . ( ) t 300 .

$_1 \sim 494$ K (M/), B $_6$F C $_3$O$_{18}$ (526 K).[23] BLFC F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+} (.).[24] - ED . A FC $_2 \sim 353$ K C $_2$F O$_4$. $_2$ - C $_2$F O$_4$ (460 K) [16,25] . (M) C $_2$F O$_4$ 16 23.5 / .[25] , 1.4 . % C $_{2-}$ F O$_4$ 0.22 0.32 / , BLFC M $= 1.85$ / , F . 2( ). I - , M H - $_1$ $_2$ (F . 3). - 425 K 1.58 / . 0.27 / , ED . , BLFC A .

F 3 F^{3+} O C^{3+} - (DF) *ab initio* (A) . H $U_F = 2$ $U_C = 3$ F C , , (GGA)+U . I - BLFC F . 3( ), F^{3+} C^{3+} (3.1 2.1 μ_B/ ,), O (0.1 μ_B/). - F O$_6$ C O$_6$ ( ) F /C - F 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 . F 4 BLFC . I - , - ,

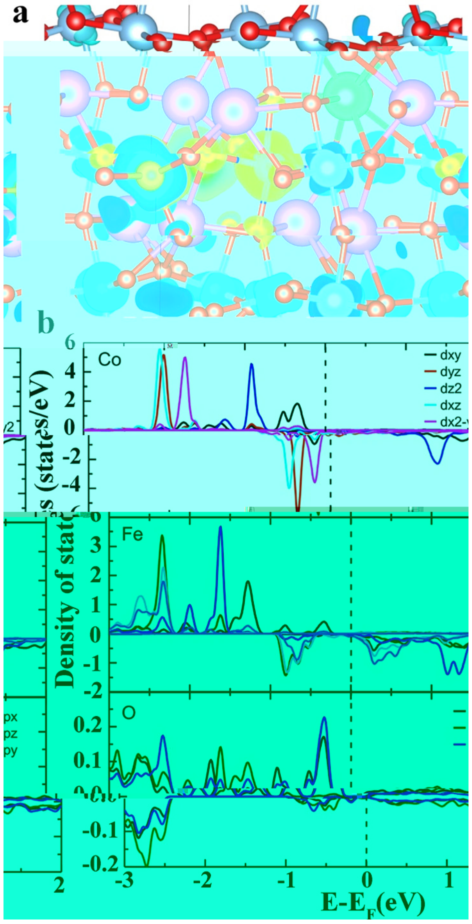


FIG. 3. ( ) $= 0.005$) ( ), ( ), ( ), ( ), ( ), ($_W$) . ( ) $^{3+}$ $^{3+}$.

. . 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 399 O . FM BLFC , F . 5( ). A F -

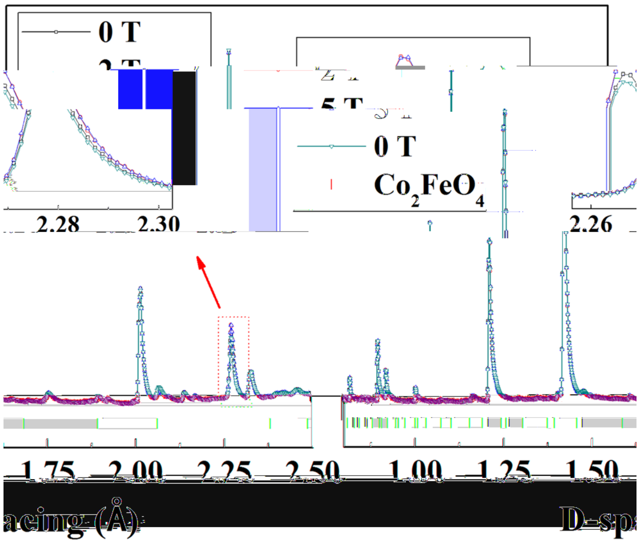


FIG. 4. 0, 2, 5 0).

C - ( ) - +2000 O F . 5( ). −2000 O - , F . 5( ). (-) C $_2$F O$_4$ - - - . , - , - , A BLFC . G (P) 100 (M) 010 , (-

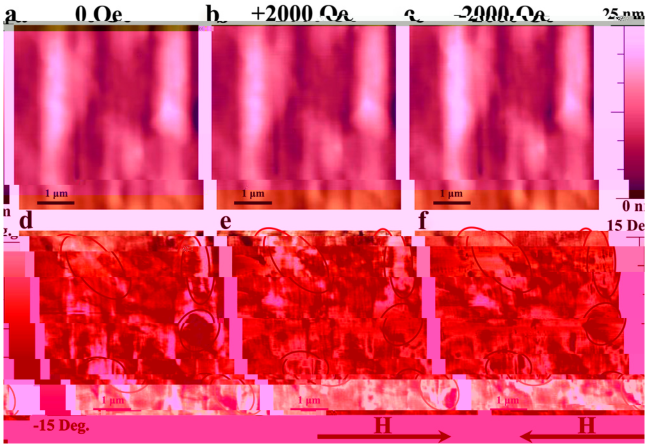


FIG. 5. ( ). ( ) ( ) M 0 , ( ) ( ) M +2000 , ( ) ( ) M −2000 .

) $T = P \times M$ BLFC . I , A BLFC - . F F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+} - . - . A , C /F , - - .

, EM - (ED) , BLFC .

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 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 : II. B $_4$ $_3$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 G A 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).

[16] M. , . . C , M. B. , A. . B , J. . H , . K , L. K , . , M. , . , C. , . H. K , A. J. B , J. A . . **112**, 073919 (2012).

[17] J. L , . , H. , M. J. , . K , . , J. A . . **102**, 104107 (2007).

[18] M. G. C , *Characterisation of Ferroelectric Bulk Materials and Thin Films* (, 2014), . 2.

[19] . L , 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).

[20] . K , I. , G. , M. , C. J , H. , J. . C . C **122**, 15733 (2018).

[21] L. J , F. L , . , J. A . C . . **97**, 1 (2014).

[22] H. , F. I , G. , H. N , H. , . J , . , . G , M. J. , J. A . D . **1**, 107 (2011).

[23] J. , L. , . L , . , . , J. D , . , . , A . . L . **101**, 012402 (2012).

[24] B. , J. , J. C , . , L. , . , . , J. D , . , . , A . . L . **104**, 062413 (2014).

[25] I. . M . N. B , . . . **11**, 719 (2009).