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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

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

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



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ABSTRACT

Multiferroic Aurivillius phase ceramics (APCs) have attracted significant attention due to their potential applications in spintronics and memory devices. However, the room-temperature multiferroic behavior in layer-structured APCs remains elusive. In this work, we report the synthesis and characterization of a series of layer-structured APCs with the general formula $B_{5-2x}L_{0.75}F_{0.75}C_{1-x}O_{18}$ ($x = 0, 0.1, 0.2, 0.3, 0.4$). The structure of these APCs is based on the Aurivillius phase structure, which consists of a perovskite layer (A) sandwiched between two layers of B_2O_2 and BO_3 groups. The presence of the L and F ions in the perovskite layer leads to the formation of a layered structure. The room-temperature multiferroic behavior is observed in these APCs, which is attributed to the presence of the F^{3+} and C^{3+} ions in the perovskite layer. The multiferroic behavior is characterized by the presence of a magnetic transition at room temperature and a ferroelectric transition at a higher temperature. The multiferroic behavior is observed in all the APCs studied here, indicating that the room-temperature multiferroic behavior is a general property of layer-structured APCs.

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Multiferroic Aurivillius phase ceramics (APCs) have attracted significant attention due to their potential applications in spintronics and memory devices. However, the room-temperature multiferroic behavior in layer-structured APCs remains elusive. In this work, we report the synthesis and characterization of a series of layer-structured APCs with the general formula $B_{5-2x}L_{0.75}F_{0.75}C_{1-x}O_{18}$ ($x = 0, 0.1, 0.2, 0.3, 0.4$). The structure of these APCs is based on the Aurivillius phase structure, which consists of a perovskite layer (A) sandwiched between two layers of B_2O_2 and BO_3 groups. The presence of the L and F ions in the perovskite layer leads to the formation of a layered structure. The room-temperature multiferroic behavior is observed in these APCs, which is attributed to the presence of the F^{3+} and C^{3+} ions in the perovskite layer. The multiferroic behavior is characterized by the presence of a magnetic transition at room temperature and a ferroelectric transition at a higher temperature. The multiferroic behavior is observed in all the APCs studied here, indicating that the room-temperature multiferroic behavior is a general property of layer-structured APCs.

BLFC $a b$ $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$, $c = 50.670(2) \text{ \AA}$, $b = 5.3943(6) \text{ \AA}$, $c = 41.487(2) \text{ \AA}$

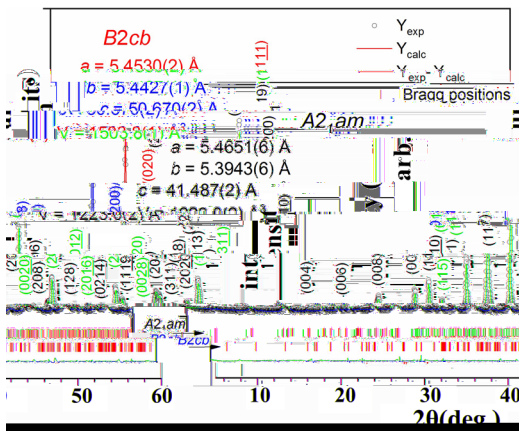


FIG. 1. XRD pattern of BLFC showing intensity versus 2θ (deg).

BLFC $a b$ $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$, $c = 50.670(2) \text{ \AA}$, $b = 5.3943(6) \text{ \AA}$, $c = 41.487(2) \text{ \AA}$

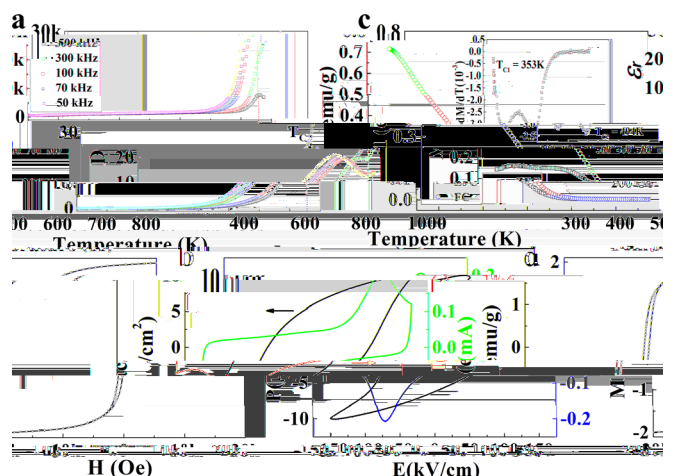


FIG. 2. (a) Temperature dependence of the dielectric constant ϵ_r for BLFC at frequencies of 300 kHz, 100 kHz, 70 kHz, and 50 kHz. (b) Temperature dependence of the dielectric loss tangent $\tan \delta$ for BLFC at the same frequencies. (c) Temperature dependence of the piezoelectric coefficient d_{33} (pm/V) for BLFC, showing a peak around 800 K. (d) Temperature dependence of the pyroelectric coefficient P_r ($\mu\text{C}/\text{cm}^2$) for BLFC, showing a peak around 800 K. The temperature range is from 300 K to 1000 K.

~ 494 K
 $M/$),
 $B_6F C_3O_{18}$ (526 K).²³
 BLFC
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$ (.
 ED .²⁴
 A FC $2 \sim 353$ K
 $C_2F O_4$.
 $C_2F O_4$ (460 K) .^{16,25}
 (M) $C_2F O_4$.
 $16 \ 23.5 \ /$.²⁵ , $1.4 \ .\%$
 $C_{2-} F O_4$ $0.22 \ 0.32 \ /$, BLFC
 $M = 1.85 \ /$, $F . 2() . I$
 $M H$.
 $2 (F . 3)$.
 425 K $1.58 \ /$.
 $0.27 \ /$, ED
 $BLFC$.
 A .
 $F 3$.
 (DF) $F^{3+} O C^{3+}$ *ab initio*
 $(A P)$.
 $U_F = 2$ $U_C = 3$ $F C$,
 (GGA) .
 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),
 1 FC/FC . $F . 2()$.
 $a b$
 010 .
 $BLFC$ $F 4$.
 I .
 $BLFC$, $399 O$.
 PFM $BLFC$, $F .$
 $5() . A$ P $F M$
 $F 5$.
 $5() . A$ PFM $BLFC$, $F .$
 P $F M$

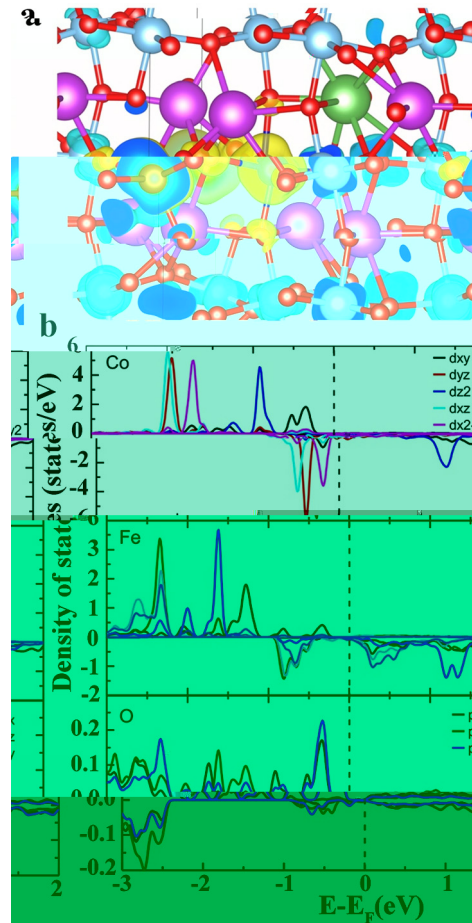


FIG. 3. (a) Crystal structure of BLFC showing Co, Fe, and O atoms. (b) Density of states (DOS) for Co, Fe, and O atoms, showing contributions from dxy, dyz, dz2, dxz, and dx2-y2 orbitals. The x-axis is E-E_F (eV) from -2 to 1, and the y-axis is Density of states (states/eV).

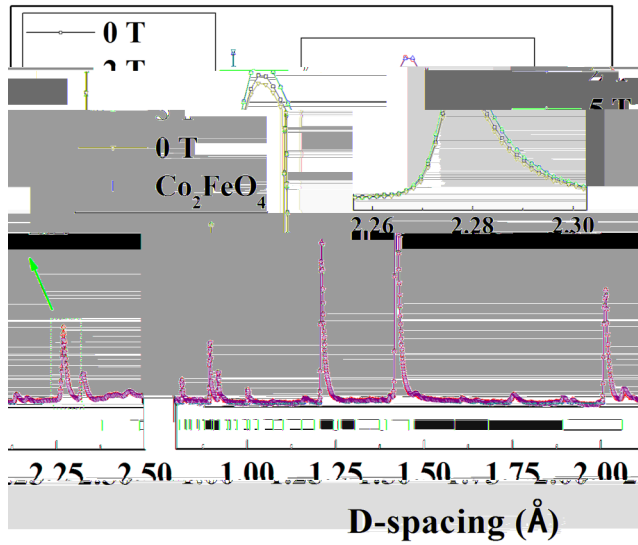


FIG. 4. XRD patterns of Co_2FeO_4 at various magnetic fields (0, 2, 5 T) at $\theta = 0^\circ$.

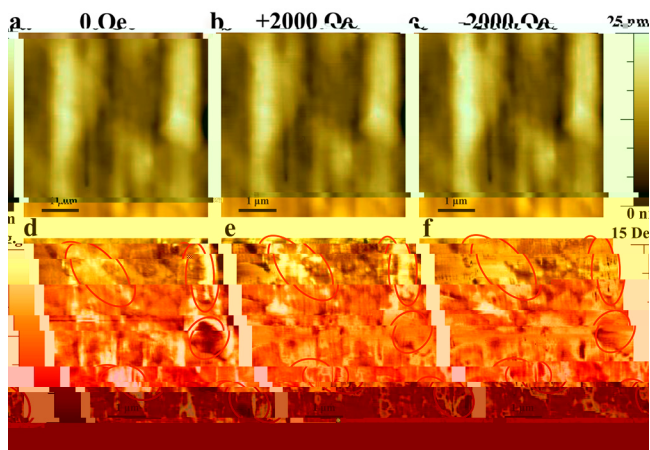
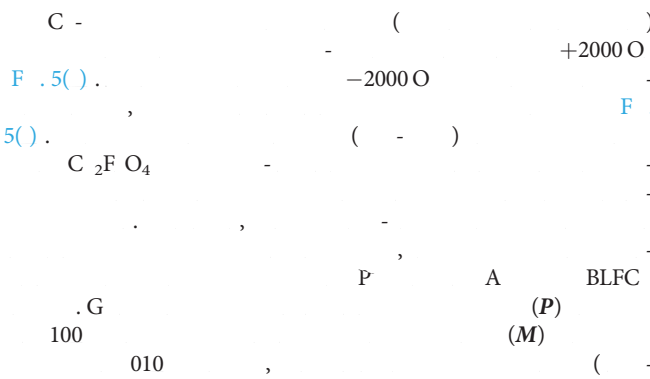


FIG. 5. MFM images of Co_2FeO_4 at various magnetic fields (0, +2000, -2000 Oe).

$T = P \times M$
 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

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., O. G., D. C. L., H., K., A. J. B., A. F. M. 26, 2111 (2016).
5. N. A. H., J. P. C. B 104, 6694 (2000).
6. B. A., M. : IL.
7. B. 4 3O₁₂, A. K. I(58), 499-512 (1949).
7. A., G. K., M. M. K., J. P. C. M. 11, 3335 (1999).
8. N. P., G. K., M. E. B 108, 194 (2004).
9. L. K., M., M., A. A., N. D., N. P., E. P., 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., NPGA M. 5, 72 (2013).
12. A. B., C. E., P. B 90, 214109 (2014).
13. J. B. L., P. H., G. H., G. L., J. L., J. C., J. K. L., A. P. L. 96, 222903 (2010).
14. M., C., L., A. P. L. 95, 082901 (2009).
15. L., J., L., J. D., A. P. L. 101, 122402 (2012).

- ¹⁶M. 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).
- ¹⁷J. L. , H. , M. J. , K. , P. , *J. A. P.* **102**, 104107 (2007).
- ¹⁸M. 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. P. C.* **122**, 15733 (2018).
- ²¹L. J. , F. L. , , *J. A. C.* **97**, 1 (2014).
- ²²H. , F. I. , G. , H. N. , H. , J. , .G. , M. J. , *J. A. D.* **1**, 107 (2011).
- ²³J. , L. , .L. , . , J. D. , . , A. . *P. L.* **101**, 012402 (2012).
- ²⁴B. , J. , J. C. , .L. , . , J. D. , . , A. *P. L.* **104**, 062413 (2014).
- ²⁵I. P. M. , N. B. , . , **11**, 719 (2009).