



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

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ABSTRACT

Multiferroic behavior is observed in layer-structured Aurivillius phase ceramics $B_{5.25}L_{0.75}F_{1-x}C_{1-x}O_{18}$ ($x = 0.1, 0.2, 0.3, 0.4, 0.5$) at room temperature. The $B_{5.25}L_{0.75}F_{1-x}C_{1-x}O_{18}$ system is a layered structure consisting of $F^{3+}O$, $F^{3+}C$, and O layers. The $F^{3+}O$ and $F^{3+}C$ layers are antiferromagnetic (AFM) and the O layer is nonmagnetic. The $B_{5.25}L_{0.75}F_{1-x}C_{1-x}O_{18}$ system shows a transition from a paramagnetic (PM) state to a multiferroic (MF) state at room temperature. The MF behavior is attributed to the presence of the $F^{3+}O$ and $F^{3+}C$ layers. The $B_{5.25}L_{0.75}F_{1-x}C_{1-x}O_{18}$ system shows a transition from a PM state to a MF state at room temperature. The MF behavior is attributed to the presence of the $F^{3+}O$ and $F^{3+}C$ layers.

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Multiferroic (MF) behavior is observed in layer-structured Aurivillius phase ceramics $B_{5.25}L_{0.75}F_{1-x}C_{1-x}O_{18}$ ($x = 0.1, 0.2, 0.3, 0.4, 0.5$) at room temperature. The $B_{5.25}L_{0.75}F_{1-x}C_{1-x}O_{18}$ system is a layered structure consisting of $F^{3+}O$, $F^{3+}C$, and O layers. The $F^{3+}O$ and $F^{3+}C$ layers are antiferromagnetic (AFM) and the O layer is nonmagnetic. The $B_{5.25}L_{0.75}F_{1-x}C_{1-x}O_{18}$ system shows a transition from a paramagnetic (PM) state to a multiferroic (MF) state at room temperature. The MF behavior is attributed to the presence of the $F^{3+}O$ and $F^{3+}C$ layers. The $B_{5.25}L_{0.75}F_{1-x}C_{1-x}O_{18}$ system shows a transition from a PM state to a MF state at room temperature. The MF behavior is attributed to the presence of the $F^{3+}O$ and $F^{3+}C$ layers.

$B_{5.25}L_{0.75}F_{1-x}C_{2x-3}O_{18}$
 (BLFC) $a b$
 $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$,
 $c = 50.670(2) \text{ \AA}$, $c = 41.487(2) \text{ \AA}$
 $A_{21}am$

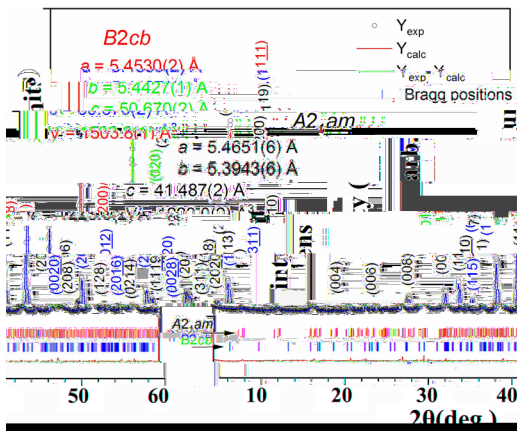


FIG. 1. XRD pattern of BLFC.

$B_{5.25}L_{0.75}F_{1-x}C_{2x-3}O_{18}$
 BLFC $a b$
 $a = 5.4530(2) \text{ \AA}$, $b = 5.4427(1) \text{ \AA}$,
 $c = 50.670(2) \text{ \AA}$, $c = 41.487(2) \text{ \AA}$
 $A_{21}am$

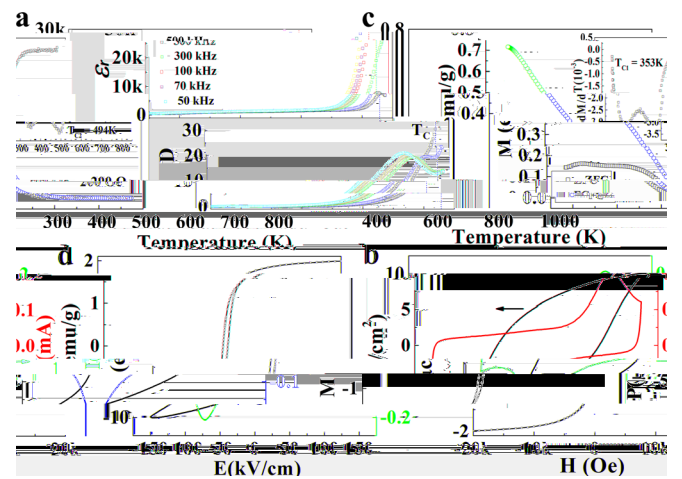


FIG. 2. (a) Dielectric loss ϵ'' vs Temperature (K) for BLFC at frequencies of 50 kHz, 70 kHz, 100 kHz, 300 kHz, and 500 kHz. (b) Temperature dependence of the dielectric constant ϵ' and loss tangent $\tan \delta$. (c) Temperature dependence of the magnetic moment M (emu/g) and Curie temperature T_c (K). (d) Field-cooled (FC) and zero-field-cooled (ZFC) magnetization M (emu/g) vs magnetic field H (Oe) at various temperatures.

$T_1 \sim 494 \text{ K}$
 $\text{BLFC} \text{ } B_6\text{F}_2\text{C}_{18}\text{O}_{18} \text{ (526 K)}^{23}$
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$ (. . .)²⁴
 ED
 $T_2 \sim 353 \text{ K}$
 $C_2F_4 O_4$
 $C_2F_4 \text{ (460 K)}$ (M) $C_2F_4 O_4$ ^{16,25}
 $16 \text{ } 23.5 \text{ } /$ ²⁵ , $0.22 \text{ } 0.32 \text{ } /$, $1.4 \text{ } .\%$
 $C_2 - F O_4$ BLFC
 $M = 1.85 \text{ } /$, $F . 2(\text{ })$, $M H$
 $T_2 \text{ (F . 3) } .$ 425 K $1.58 \text{ } /$. $0.27 \text{ } /$, ED
 BLFC
 $F . 3$ A
 (DF^-) $F^{3+} O C^{3+}$ *ab initio*
 (A) H
 $\downarrow_F = 2$ $\downarrow_C = 3$ F C , ,
 $(GGA)+\downarrow$. I
 BLFC
 $F . 3(\text{ })$, F^{3+} C^{3+} (3.1 $2.1 \mu_B/$, ,) ,
 $(0.1 \mu_B/)$.
 $F O_6$ $C O_6$
 F / C -
 F O - / $F . 3(\text{ })$.
 F^{3+} C^{3+} ,
 $(\text{ } , \text{ })$ (. ,)
 $E_{FM} - E_{AFM}$
 $= -144.1$.
 H , (FM)
 $43.5 \text{ } (\text{ } , 504.6 \text{ K})$ FM
 FC/FC $F . 2(\text{ })$ $a b$
 010
 BLFC $F . 4$. I
 399 O .
 F .
 F -

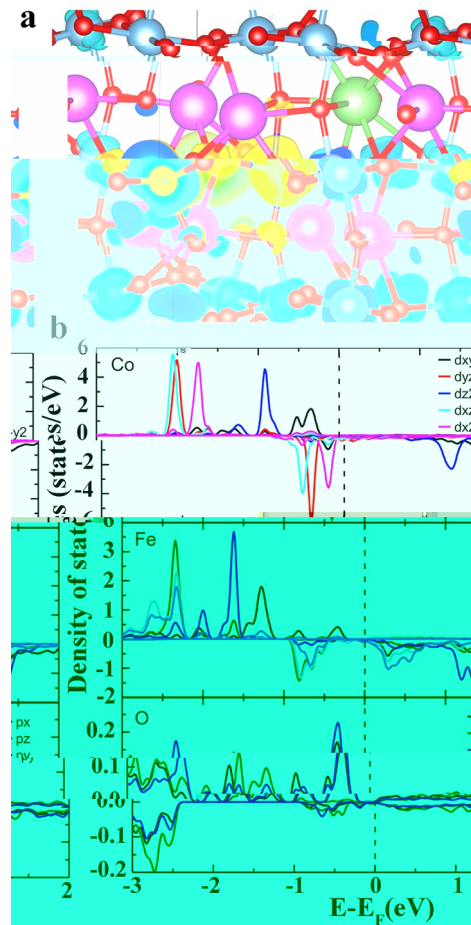


FIG. 3. () () $= 0.005$ () , B () , () , () , $3+$, $3+$ () . () A

N
 $(2^- < H < 5^-)$,
 $M H$ $F . 2()$ $3. F$,
 $F . 4$,
 $(0 \text{ } 1 \text{ } 20)$
 $2^- . F$
 $F . 5$
 BLFC F M
 399 O .
 F .
 F -
 $5() . A$ BLFC ,

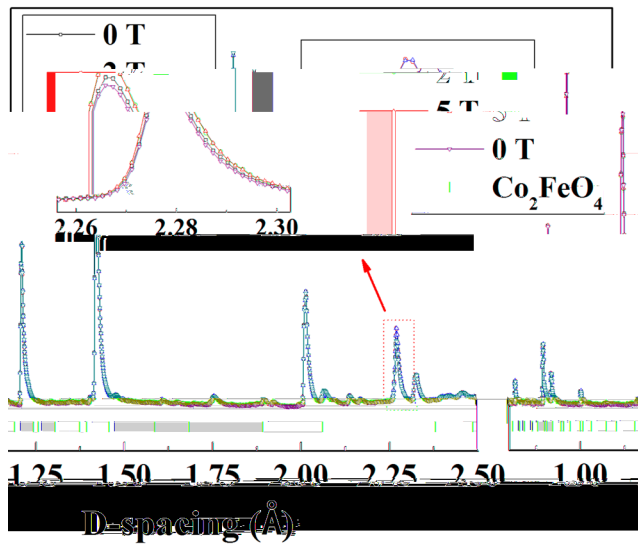


FIG. 4. XRD patterns of Co_2FeO_4 at 0 T and 5 T. The inset shows the zoomed-in view of the 2.25–2.50 Å region.

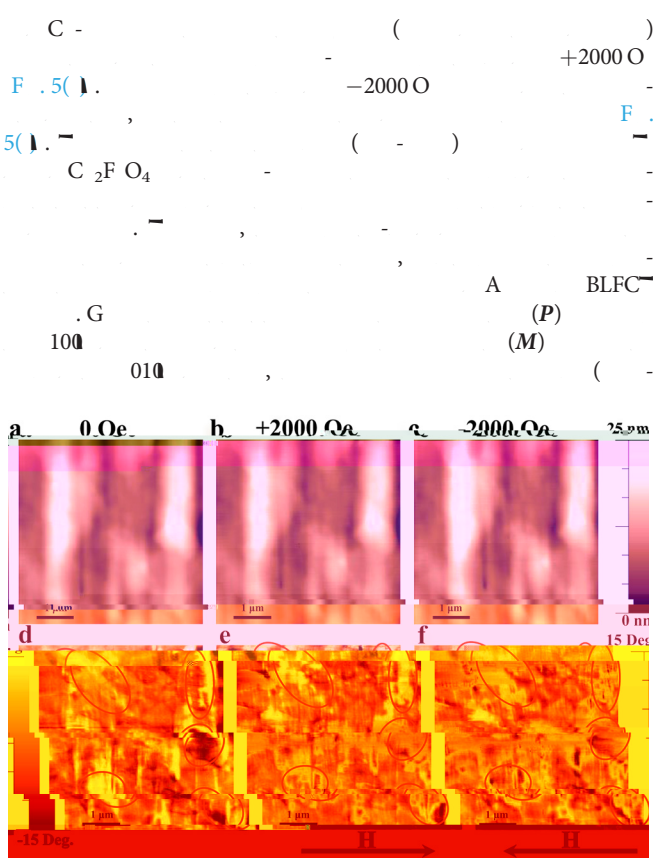


FIG. 5. MFM images of Co_2FeO_4 at 0 Oe, +2000 Oe, and -2000 Oe. The inset shows the corresponding magnetic field maps.

$T = P \times M$
 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 , 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 E₂AME⁻ E

DATA AVAILABILITY

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