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## AFFILIATIONS

<sup>1</sup>G... <sup>2</sup>I... <sup>3</sup>E... <sup>4</sup>N... <sup>5</sup>N... <sup>6</sup>E... <sup>7</sup>

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## ABSTRACT

M... H... A... B<sub>5.25</sub>L<sub>0.75</sub>F<sub>2</sub>C<sub>3</sub>O<sub>18</sub>... *in situ*... F<sup>3+</sup> O F<sup>3+</sup>, C<sup>3+</sup> O C<sup>3+</sup>, F<sup>3+</sup> O C<sup>3+</sup>... A... C/F...

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M... (FM) (FE) A... B<sub>5</sub>F<sub>2</sub>O<sub>15</sub> (=4) B<sub>6</sub>F<sub>2</sub>O<sub>18</sub>... B<sub>4</sub>O<sub>12</sub>... B<sub>5</sub>F<sub>0.5</sub>C<sub>0.5</sub>O<sub>15</sub>... (B<sub>2</sub>O<sub>2</sub>)<sup>2+</sup>(A<sub>-1</sub>B<sub>1</sub>O<sub>3</sub>)<sup>2-</sup>... A... H... B... D... B-... B F O<sub>3</sub>... 7 11... A...

$B_{5.25}L_{0.75}F_1C_{3.0}O_{18}$   
 (BLFC) P L  
 F, A, C, D  
 $a b$ , P  
 BLFC  
 $a b$   
 A  
 in situ  
 I H, I I  
 N F, AL, D, O, U K.  
 (P), A BLFC  
 P  
 BLFC  
 P.  
 F 1 (D) BLFC  
 A  
 $B2cb$   
 A  
 $A2_1$   
 $B2cb$   
 $a = 5.4530(2) \text{ \AA}$ ,  $b = 5.4427(1) \text{ \AA}$ ,  
 $c = 50.670(2) \text{ \AA}$ ,  $A2_1am$   $a = 5.4651(6) \text{ \AA}$ ,  
 $b = 5.3943(6) \text{ \AA}$ ,  $c = 41.487(2) \text{ \AA}$   
 F P ( //

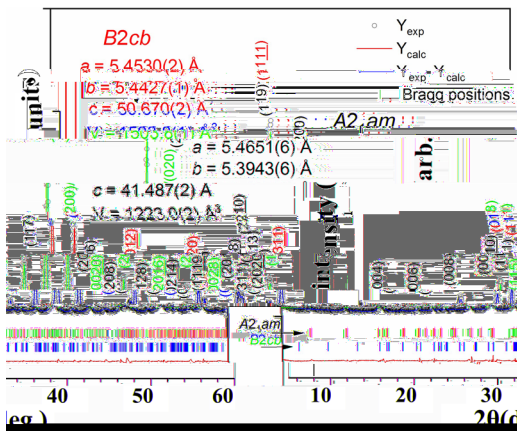


FIG. 1. XRD patterns of B2cb and A2<sub>1</sub>am phases.

BLFC = 4 = 5 A N  
 BLFC F 1 EM (a-b) M  
 F 1  
 D. ED 1.4 %, (F 2  
 1)  
 F, C, O, C<sub>2</sub>F O<sub>4</sub>  
 A B<sub>5</sub>F<sub>0.5</sub>C<sub>0.5</sub>O<sub>15</sub><sup>16</sup>  
 BLFC (50, 70 100,  
 300, 500 H).  
 1060 K FE T BLFC H,  
 ( 973 K).<sup>13</sup> F BLFC P-E I-E B<sub>6</sub> F<sub>3</sub> O<sub>18</sub>  
 BLFC 2() I-E  
 P 21,22  
 BLFC 10 μC/ F 2() (FC) BLFC  
 ( FC) 200 O BLFC BLFC

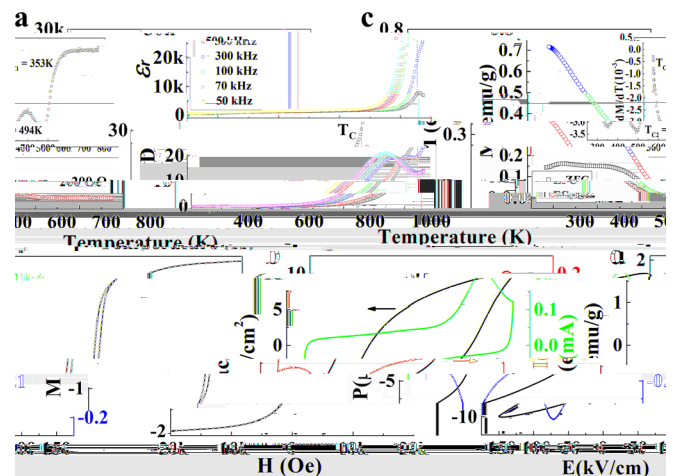
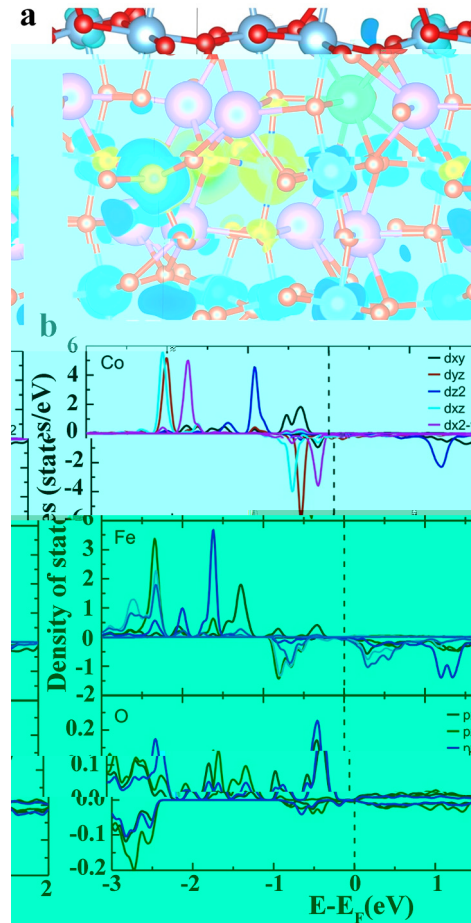


FIG. 2. (a) Temperature dependence of dielectric loss (tan δ) for BLFC at various frequencies. (b) Magnetization (M) and magnetoelectric coefficient (ME) vs magnetic field (H) and electric field (E) at various temperatures. (c) Temperature dependence of the magnetoelectric coefficient (ME) for BLFC at various frequencies.

$\sim 494$  K  
 $M/$  ),  
 $B_6F C_3O_{18}$  (526 K).<sup>23</sup>  
 BLFC  
 $F^{3+} O F^{3+}, C^{3+} O C^{3+}, F^{3+} O C^{3+}$  ( .  
 $ED$  ).<sup>24</sup>  
 $A$  FC  $2 \sim 353$  K  
 $C_2F O_4$   $2$   $16,25$   
 $C_2F O_4$  (460 K) (M)  $C_2F O_4$   
 $16 \ 23.5 \ /$  .<sup>25</sup> ,  $0.22 \ 0.32 \ /$  ,  $1.4 \ .\%$   
 $C_{2-} F O_4$  BLFC  
 $M = 1.85 \ /$  ,  $F . 2( ) . I$  ,  $M H$   
 $2 (F . 3)$   $1$   
 $425$  K  $1.58 \ /$  .  $0.27 \ /$  ,  $ED$   
 $BLFC$   
 $A$   
 $F 3$   $F^{3+} O C^{3+}$   
 $(DF)$   $ab initio$   
 $(A P)$   $H$   
 $U_F = 2$   $U_C = 3$   $F C$  ,  
 $(GGA)$   $I$   
 $BLFC$   
 $F . 3( )$  ,  $F^{3+} C^{3+}$  (3.1  $2.1 \mu_B/$  , ) ,  
 $O$   
 $( 0.1 \mu_B/ )$  .  
 $F O_6 C O_6$   
 $( )$   $F/C$  -  $F . 3( )$  .  
 $F$   $O$  -  $/$   $F . 3( )$  .  
 $F^{3+} C^{3+}$  ,  
 $( . , )$   $( . , )$  ,  
 $= -144.1$  .  
 $H$  , (FM)  
 $43.5$  ( . , 504.6 K),  $FM$  -  
 $1$  FC/FC  $. F . 2( )$  .  $2$  ,  
 $a b$   
 $010$  .  
 $BLFC$   $F 4$   $I$   
 $5( ) . A$  PFM  $BLFC$  ,  $399 O$  .  
 $F$  -



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

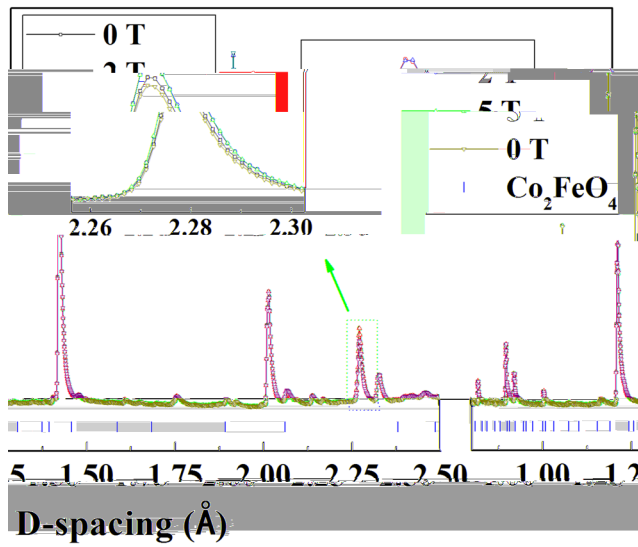


FIG. 4. XRD patterns of  $\text{Co}_2\text{FeO}_4$  at 0 T and 2 T. The inset shows the magnified view of the peak at  $2.25 \text{ \AA}$ .

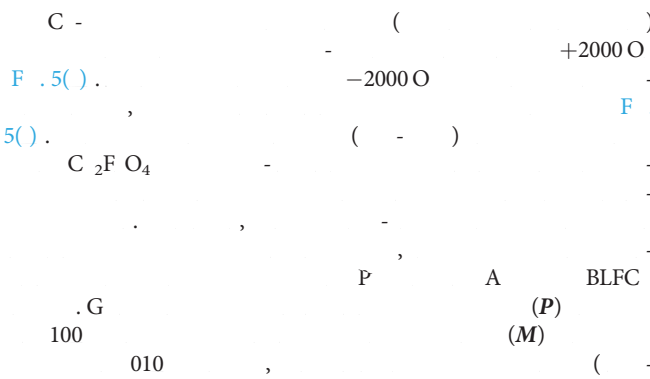


FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe, +2000 Oe, and -2000 Oe. The inset shows the magnified view of the phase image.

FIG. 5. MFM images of  $\text{Co}_2\text{FeO}_4$  at 0 Oe, +2000 Oe, and -2000 Oe. The inset shows the magnified view of the phase image.

$T = P \times M$   
 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 P 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. 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. 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., 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).

- <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>L. P. M. ..., N. B. ..., **11**, 719 (2009).