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AFFILIATIONS

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

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ABSTRACT

M
. H , -
. , - A
,
D . H , A B 5.25L 0.75F C 3O18
.
A , *in situ*
.
F 3+ O F 3+, C 3+ O C 3+, F 3+ O C 3+ - .
. A , C /F -
, - .

M (FE) A B 5F 3O15 (=4) B 6F 2 3O18 (FM) (=5), B 4 3O12
, , BF O3, FE FM .[12,13] B -
, .[1 4] H , - C F B- , B 5F 0.5C 0.5 3O15
- (=4) B 6F C 3O18 (=5)
.[5] , - A
(B 2O2)2+(A -1B O 3 +1)2− (A -, .[14,15] H ,
-, 12- - , B
- ,
D -
)[6] .[16] , -
B- - A
BF O3 - .[7 11] .

I , A $B_{5.25}L_{0.75}FC_3O_{18}$ (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 (:// . .).

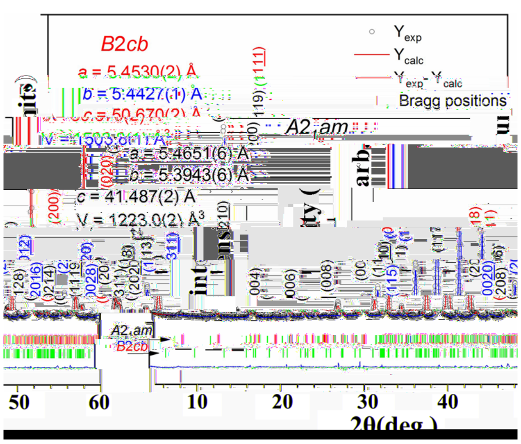

FIG. 1.

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 2F O4 .

- A $B_5F_{0.5}C_{0.5}{}_3O_{15}$.[16] F 2( ) BLFC (50, 70 100, 300, 500 H). 1060 K FE T BLFC. H , BLFC $B_6{}_3F_2O_{18}$ (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

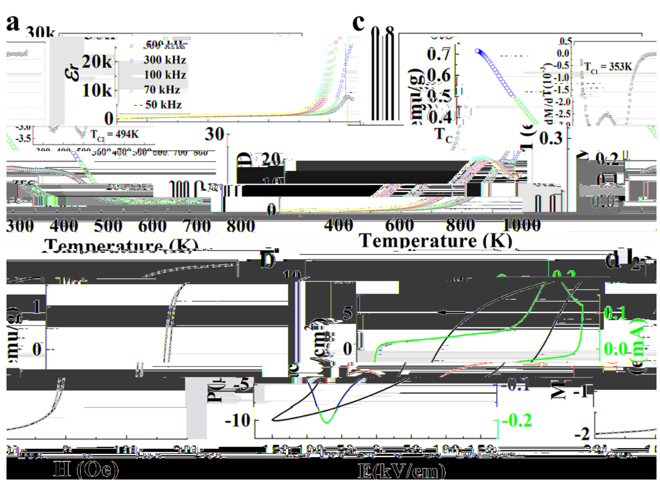

FIG. 2. ( ) = 200 . ( ) 300 .

$_1 \sim 494$ K
(M/),
B_6FC $_3O_{18}$ (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_2FO_4. $_2$
C_2FO_4 (460 K) .[16,25]
(M) C_2FO_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
$_F = 2$ $_C = 3$ F C , ,
(GGA)+ . 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
- , - ,

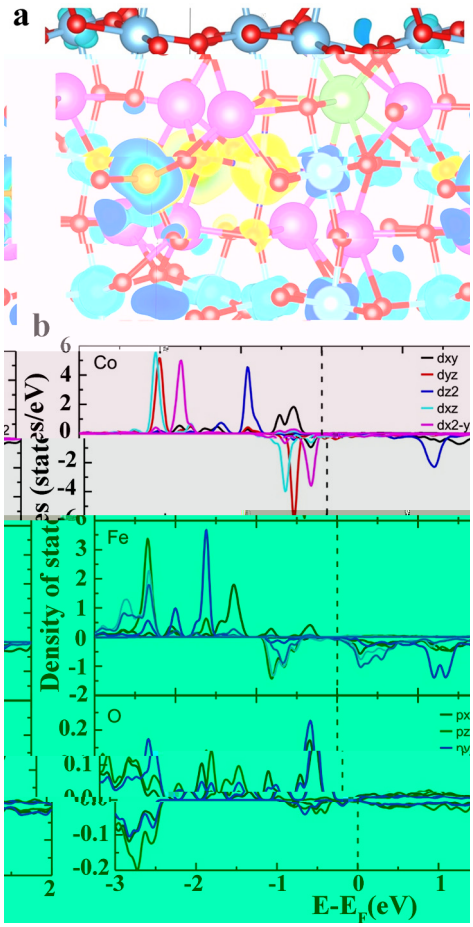

FIG. 3. ( ) t n t n (= 0.005) ( ), ( ), ( ), ( ), ( ), ( ) t n . ( ) t t n t t $^{3+}$ $^{3+}$.

.
. 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 -

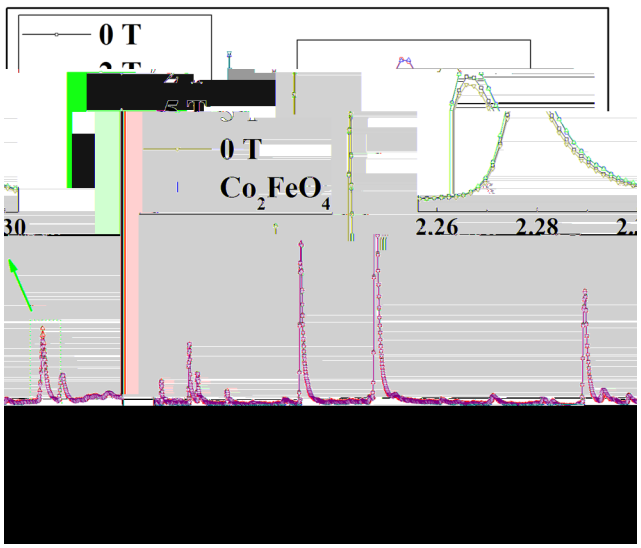

FIG. 4. [illegible]

C - ( ) - +2000 O F . 5( ). −2000 O - , F . 5( ). (-) C 2F O4 - - . , - , - A BLFC . G (P) 100 (M) 010 , (-

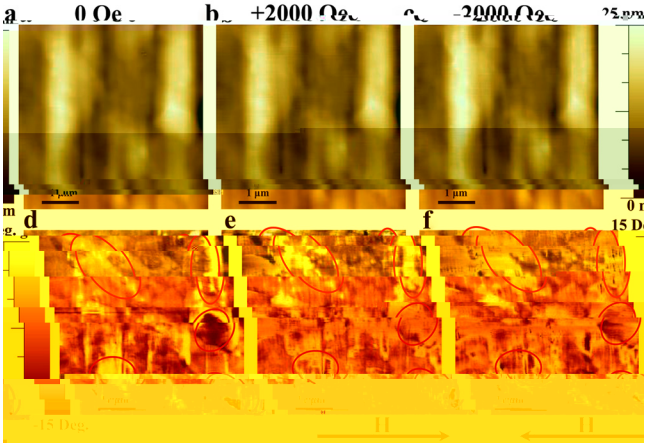

FIG. 5. [illegible]

) $T = P \times M$ 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 3O12, 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).