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Magnetization and characteristic fields in $Nd_{0.8}Ba_{0.2}FeAsO_{0.6}F_{0.4}$ with binary doping

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ABSTRACT

In the present work, the iron oxypnictide superconductor of the namely $Nd_{0.8}Ba_{0.2}FeAsO_{0.6}F_{0.4}$ is prepared with respect to the observation of distinct magnetization characteristics arising from both electron and hole doping. A magnetothermal phase diagram is given for the present iron oxypnictide system, based on the irreversibility fields (H_{irr}) and the upper critical fields (H_{c2}) obtained from magnetotransport measurements, as well as the lower critical fields (H_{c1}) evaluated by magnetization loops at various temperatures. High $H_{c2}(0)$ are revealed at low temperature range, which is in consistent with the observations of comparatively high critical current (J_c) and the temperature dependent peak effect in the J_c vs H curves.

1. Introduction

Among the family of oxypnictide based superconductors, the $RFeAsO_{1-y}$ (FeAs-1111, R is the rare earth element) exhibits a relatively high superconducting critical temperature, reaching 55 K [1] in the case of electron doping such as $SmFeAsO_{1-x}F_x$. The parent compounds of both 1111 and 122 type, normally show the magnetic order phase of spin density wave (SDW), but no superconductivity [2–4]. They may evolve into superconductors as a consequence of the normative symmetry breaking of 1111 type SDW due to the element doping, such as $SmFe_{0.9}Co_{0.1}AsO$ [5], $Ca_{1-x}Na_xFe_2As_2$ [6], and so on. Noting the iron site doping generally suppresses the critical transition temperature, and enhances the disorder of magnetism as well.

Apart from one element or one site substitution, people also suggest that the superconductivity is tailorable by the second element doping in other site simultaneously [7–9], such as $Ce_{1-x}Gd_xFeAsO_{0.84}F_{0.16}$ [7], and $SmFe_{1-x}Ru_xAsO_{0.85}F_{0.1}$ [8]. It is reported that the second site of doping could enhance transition temperature as well as the upper critical field [10].

We recently look into the effect of binary doping on the performances of the oxypnictide superconductors, $Nd_{1-x}Ba_xFeAsO_{1-2x}F_{2x}$ at a series of doping contents (x = 0.02, 0.05, 0.1, 0.15, and 0.2) [11]. Temperature dependences of electric resistivity and magnetic susceptibility reveal that the superconductivity for the studied system emerges at $x = 0.1$, and enhances together with $H_{c2}(0)$ as the doping content x increases further. In case of $x = 0.2$, the superconducting critical temperature reaches as high as 50 K, which is the first demonstration of superconductivity with a high fluorine doping induced by both electron and hole doping in this family.

In the present paper, we focus on the sample of $x = 0.2$, which exhibits the highest T_c in $Nd_{1-x}Ba_xFeAsO_{1-2x}F_{2x}$ series. A lot of efforts have been made to understand its distinct magnetization behaviors and magnetothermal phase diagrams, together with various characteristic magnetic fields. Note that such a special system with both electron-doping and hole-doping may allow more breaking of original symmetry in composite and structure, giving rise to more complicated superconducting and magnetic behaviors.

2. Experimental

Polycrystalline bulk of nominal $Nd_{0.8}Ba_{0.2}FeAsO_{0.6}F_{0.4}$ was synthesized by a conventional solid state reaction. Firstly, the intermediate phases of Fe_2As, NdAs and FeO were prepared by the reactions of the mixed commercial powders of Nd, Fe, As and Fe_2O_3, sealed in a vacuum quartz tube, annealed at 600 °C for 6 h and then 850 °C for 12 h. After that, the pressed pellets were sealed in a vacuum quartz tube for final annealing at 1160 °C. More details for sample preparation can be found in the Ref. [12].

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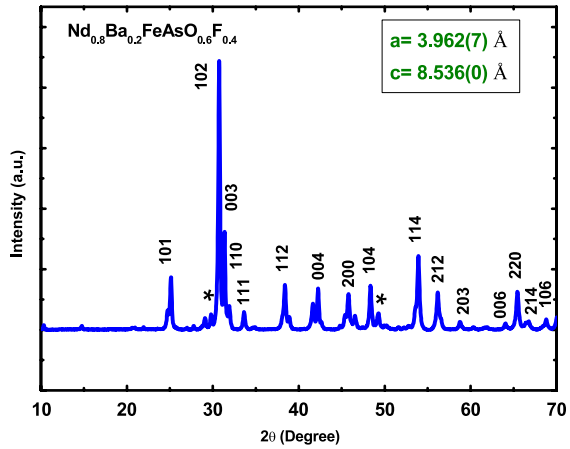

Fig. 1. (Color online) X-ray diffraction pattern for the $Nd_{0.8}Ba_{0.2}FeAsO_{0.6}F_{0.4}$ compound with its lattice parameters.

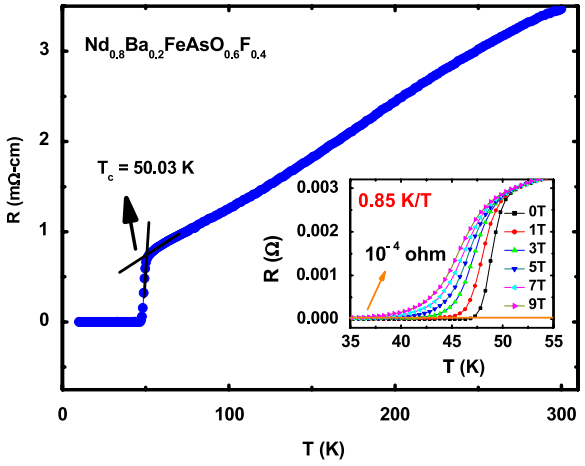

Fig. 2. (Color online) Temperature dependence of the resistivity for the present sample of $Nd_{0.8}Ba_{0.2}FeAsO_{0.6}F_{0.4}$, the arrow indicates the critical transition temperature. The inset shows the temperature dependence of the resistance in various applied magnetic fields up to 9 T.

The superconducting properties were measured by using a physical property measurement system (Quantum Design PPMS) in magnetic fields up to 9 T. For the magnetization measurements, rectangular specimens were prepared nearly in the same dimension of about $6 \times 2 \times 1$ mm^3, and the applied magnetic field was normal to the long axis direction of the specimen.

3. Result and discussion

3.1. Magnetotransport performances

Fig. 1 shows the room temperature X-ray diffraction (XRD) patterns for the present sample of $Nd_{0.8}Ba_{0.2}FeAsO_{0.6}F_{0.4}$ alone with their Rietveld refinement. It is observed that all main peaks are well indexed based on their basis of space group P4/nmm, although little extra peaks at around 30° and 49° of low intensity (marked with $*$). Based on the index, the lattice parameters for such a compound, is calculated to be $a = 3.962(7)$ and $c = 8.536(0)$.

Fig. 2 shows the superconducting critical temperature, identified from the R vs. T curves. It is revealed that the present sample exhibits the superconductivity 50 K at $x = 0.2$. Moreover, the superconducting transition width ΔT, determined by the temperatures corresponding to 90% and 10% of the normal state resistivity, decreases with increasing doping content, being as low as 1.86 K at $x = 0.2$. The resistivity exhibits the linear behavior above T_c, and the residual resistance ratio ($RRR = R(300\text{ K})/R(T_c)$) reaches 5.5, demonstrating the good quality of the present sample.

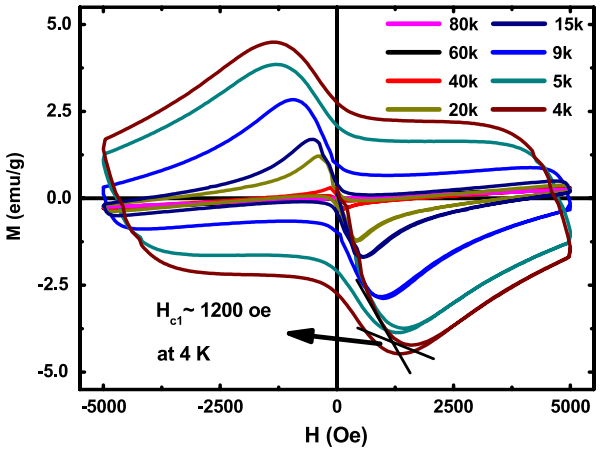

Fig. 3. (Color online) The magnetization curves $M(H)$ of the present sample of $Nd_{0.8}Ba_{0.2}FeAsO_{0.6}F_{0.4}$ at various temperatures from 4 to 80 K.

The inset of Fig. 2 shows the temperature dependences of electric resistance in various applied magnetic fields. The superconducting transition temperature is determined by using a criterion [13] at which the resistivity becomes negligible (about 10^{-4} Ohm). It is found that the transition temperature decreases with applied magnetic field, roughly at a slope rate of 0.85 K/T, i.e. dT_c/dH−0.85 K/T, in contrast to 1 K/T of dT_c/dH for $SmFe_{0.85}Co_{0.15}AsO$ [13]. Obviously, the latter is larger than the former, implying the higher H_{c2} and stronger pinning effect probably existing in the present sample.

As well, this value is much lower than that of YBCO (dT_c/dH−4 K/T) and MgB_2 (dT_c/dH−1.5 K/T) samples [14,15], suggesting a high value of upper critical field H_{c2} in these compounds [16]. One may suppose that the oxypnictides appear as a new class of high field superconductors with H_{c2} surpassing the corresponding values of Nb_3Sn and MgB_2. In reality, the upper critical magnetic fields of the iron oxypnictides superconductors may surpass the 100 T, a benchmark of the high T_c cuprate superconductors. The values of H_{c2} and H_{irr} for the present sample will be evaluated and discussed in the section below.

3.2. Critical field and critical current

Fig. 3 shows the magnetic hysteresis curves measured at temperatures of from 4 to 80 K in the field of $-0.5\text{ T} < H < 0.5$ T. The experimental $M(H)$ curves can be understood as the superposition of a superconducting contribution and a paramagnetic background emerges above T_c. The arrow indicates the characteristic magnetic fields, H_{c1}, which are about 1200 Oe at 4 K. Note that the peak of magnetization is broadened with the decreasing of the temperature.

It is nontrivial to notice that the M vs. H is not symmetric, with the peak deviation from the y axis. This scenario is frequently observed in iron oxypnictides superconductors, not only in single crystal [17] but also in polycrystalline samples [13,18,19]. Another feature is the peak deflection from the origin point [13,20] after deleting the paramagnetic background determined by M vs. H at the temperatures above T_c.

Fig. 4 shows the temperature dependence of H_{c1}. One can find the value surpasses 0.1 T at 4 K taken above. The values of H_{c1} decreasing with the increasing of temperature can be fitted well by using the expression of $H_{irr}(T) = H_{irr}(0)^*(1 - T/T_c)^{\beta}$.

Based on the magnetotransport measurements (shown in the inset of Fig. 2), the other two characteristic magnetic fields,

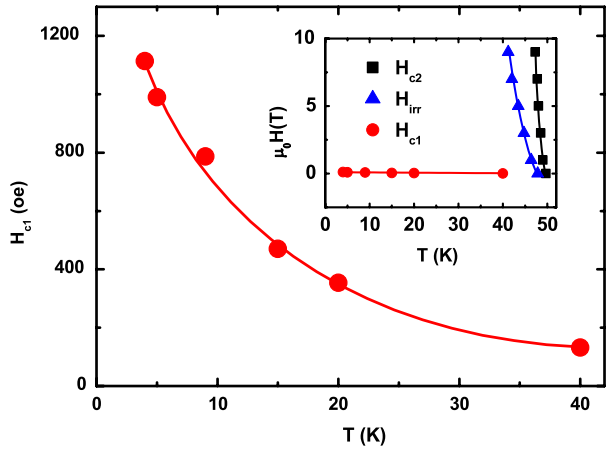

Fig. 4. (Color online) The temperature dependence of H_{c1} evaluated from $M(H)$. The inset shows the phase diagram characterized by the temperature dependences of H_{c2}, $H_{\rm irr}$ and H_{c1}.

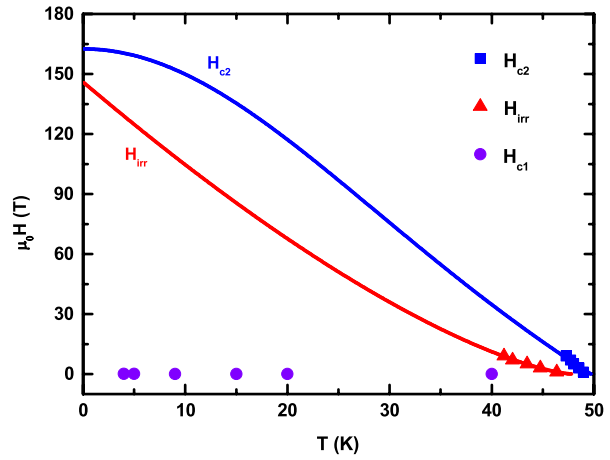

Fig. 5. (Color online) The extended temperature dependences of H_{c1}, $H_{\rm irr}$ and H_{c2} at a wide range of temperatures, based on the values fitted by the formula of $H_{\rm irr}(T) = H_{\rm irr}(0)^*(1 - T/T_c)^\beta$ and the G–L formula of $H_{c2}(T) = H_{c2}(0)\frac{1-(T/T_c)^2}{1+(T/T_c)^2}$.

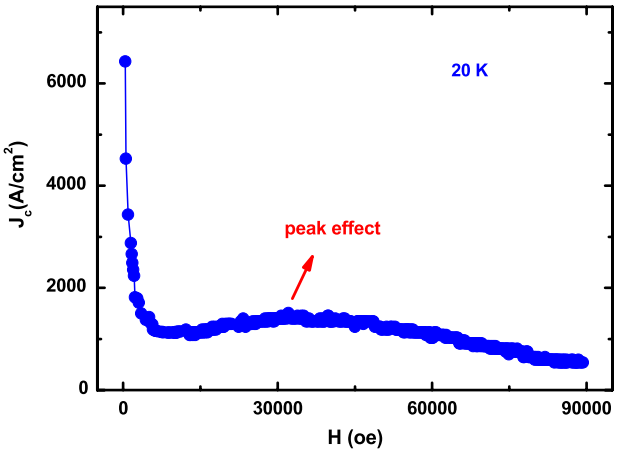

Fig. 6. (Color online) The field dependence of the critical current density at 20 K estimated from magnetization curves. Peak effect is marked with arrow.

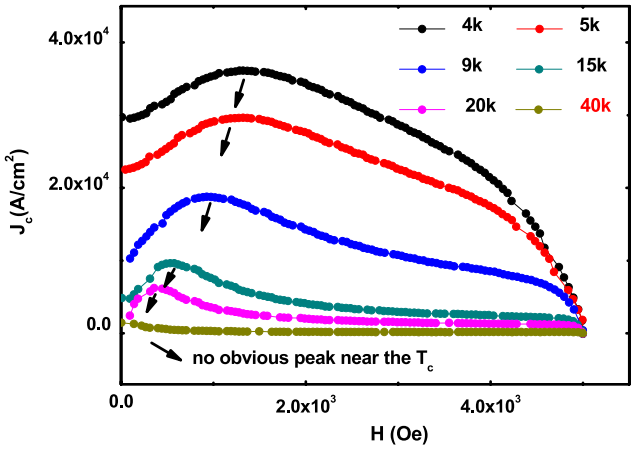

Fig. 7. (Color online) The critical current density vs. the applied magnetic field up to 5000 Oe at several given temperatures. It is revealed that the peak position shifts towards the low magnetic field with increasing the temperature, until it disappears near the T_c.

irreversibility fields ($H_{\rm irr}$) and upper critical fields (H_{c2}) are determined by the criterion of 10% and 90% of normal state resistance, respectively. Noting that similar evaluation is widely applied to YBaCuO, MgB_2, the Fluorin doped LaFeAsO and Strontium doped PrFeAsO polycrystalline samples [2,21,22]. The inset shows the relations between the reduced temperature and characteristic magnetic fields including H_{c1}, $H_{\rm irr}$ and H_{c2}. It is revealed that the $H_{\rm irr}(T)$ curve is close to the $H_{c2}(T)$ curve, implying that the strong flux pinning effect hinders the dissipating resistance due to flux jumping.

To further investigate the various characteristic magnetic fields and their relation, temperature dependences are taken into account through the whole temperature range below the critical transition temperature. The results are shown in Fig. 5, where the $H_{\rm irr}(0)$ and $H_{c2}(0)$ are evaluated by the formula below, respectively.

$$H_{c2}(T) = H_{c2}(0)\frac{1-(T/T_c)^2}{1+(T/T_c)^2} \quad (1)$$

$$H_{c2}(0) = -0.693T_c\left(\frac{{\rm d}H_{c2}}{{\rm d}T}\right)_{T=T_c} \quad (2)$$

$$H_{\rm irr}(T) = H_{\rm irr}(0)^*(1 - T/T_c)^\beta. \quad (3)$$

The first one follows the Ginzburg–Landau theory, with which the value of upper critical field $H_{c2}(0)$ is evaluated to be 162.5 T. This value is very close to the 162.8 T deduced by the Werthamer–Helfand–Hohenberg theory, the second formula. This suggests that the present sample exhibits good quality, regardless of lower than corresponding values for iron oxypnictides 42 622 family [23] and single step synthesized $NdFeAsO_{0.8}F_{0.2}$ [24]. However, the slope of $dH_{c2}(T)/dT|_{T_c}$ for the present sample is much larger than that of hole doped $Pr_{1-x}Sr_xFeAsO$ [2], and the electron doped $LaFeASO_{1-x}F_x$ [21,22].

Also one can get $H_{\rm irr}(0)$ of 146 T from the third formula with $\beta = 1.42$ which corresponds to the value (β) of YBaCuO [25,26], and this value is higher than that of $Sr(Fe_{1-x}Co_x)_2As_{2-y}$ with $H_{\rm irr}(0)$ of 70 T, but the value of β is consistent, while remaining lower than that ($\beta \sim 2$) predicted by the melting theory [27].

To further understand the pinning characteristic of present sample, the critical current density in various magnetic fields is studied as well. Fig. 6 shows the magnetic field dependence of magnetization critical current (J_c) estimated from magnetization curves and the conventional critical state model, i.e. Bean model. The center deviated peaks occur for all the magnetization curves. As marked by an arrow, a typical peak appears near 3.2 T in the case of the applied temperature 20 K.

Fig. 7 shows the magnetic J_c derived from the magnetic hysteresis vs. the applied magnetic field up to 5000 Oe at several given temperatures. Noting that the maximum applied field is lower than that of at 20 K in Fig. 6, to look into the low field

effect purposely. It is obvious that a distinct peak is present, and its position shifts towards the low magnetic field with increasing the temperatures, until it disappears near the T_c. This should be reasonable as the irreversibility fields decreases, and thus the pinging effect occurs at a relatively low field with increasing temperatures.

As for the origination of peak effects, one may attribute it to the flux pinning, either from the coexisting weak magnetic heterogeneous phases which are probably most effective at the intermediate fields or from the high density defects or nonsuperconducting phases being enhanced at matching induced magnetic fields. For the other oxypnictide based superconductors, there are few reports with regard to the peak effect. At present it is unclear whether or not this is due to the peak effect as observed in most bulks of high T_c cuprates [28].

It is noted that the present J_c at 5 K is more than $3 * 10^4$ A/cm^2, which is similar to the J_c of $2.5*10^4$ A/cm^2 at the same temperature for the polycrystalline sample of 122 type $Sr_{0.6}K_{0.4}Fe_2As_2$ with Ag addition reported by Lei Wang et al. [9]. This value of J_c is comparable with the single crystal single crystal $Ba(Fe_{1-x}Co_x)_2As_2$, its J_c (1.8 K) reaches $1.8 * 10^4$ A/cm^2 [29].

The direct result of the nonsymmetrical M vs. H curve is that the main peaks of J_c move to the right from the center. In reality, the second peak effect in the magnetization loop is frequently observed for the iron oxypnictide superconductors. For instance, single crystal $Ba_{0.6}K_{0.4}Fe_2As_2$ [18] in a magnetic field of 1000 Oe, more details should be given in polycrystalline $SmFeAsO_{0.85}F_{0.15}$, and $Sr_{1-x}Nd_xFeAsF$ [15,16,30]. The underlying antiferromagnetic phases probably make the main contribution to asymmetric magnetization, leading to the additional pinning effect at immediate field range. Further studies are required to understand the related vortex pinning mechanism [31].

It is interesting to note that the main peak moves to the left with increasing temperature, until it disappears at temperatures close to T_c such as 40 K. This implies that the effective pinning centers are subject to applied magnetic fields and are visualized at relatively low temperatures.

4. Conclusions

A special iron oxypnictide based system, namely $Nd_{0.8}Ba_{0.2}FeAsO_{0.6}F_{0.4}$ with both electron and hole doping is investigated with respect to the distinct magnetization behaviors and magnetothermal phases. While a high $H_{c2}(0)$ of about 163 T is estimated by both GL theory and WHH formula, a large slope of $dH_{c2}(T)/dT|_{T_c}$ is obtained as well, implying that the present system with both electron doping and hole doping gives rise to more breaking of original symmetry, and then more modification for magnetic and superconducting order parameters. The comparatively high critical current (J_c) and the temperature dependent peak effect suggest the complicated flux pinning for the present sample.

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References

[1] Zhi-An Ren, Wei Lu, Jie Yang, Wei Yi, Xiao Li Shen, Li Zheng Cai, Guang Can Che, Xiao li Dong, Li Ling Sun, Fang Zhou, Zhong Xian Zhao, Chin. Phys. Lett. 25 (2008) 2215.
[2] Gang Mu, Bin Zeng, Xiyu Zhu, Fei Han, Peng Cheng, Bing Shen, Hai Hu Wen, Phys. Rev. B 79 (2008) 104501.
[3] A.J. Drew, Ch. Niedermayer, P.J. Baker, F.L. Pratt, S.J. Blundell, T. Lancaster, R.H. Liu, G. Wu, X.H. Chen, I. Watanabe, V.K. Malik, A. Dubroka, M. Rössel, K.W. Kim, C. Baines, C. Bernhard, Nat. Mater. (2009) 2396.
[4] Marianne Rotter, Marcus Tegel, Dirk Johrendt, Phys. Rev. Lett. 101 (2008) 107006.
[5] V.P.S. Awana, Arpita Vajpayee, Anand Pal, Mnika Mudgel, R.S. Meena, H. Kishan, J. Supercond. Novel Magn. 22 (2009) 623–626.
[6] G. Wu, H. Chen, T. Wu, Y.L. Xie, Y.J. Yan, R.H. Liu, X.F. Wang, J.J. Ying, X.H. Chen, J. Phys.: Condens. Matter 20 (2008) 422201.
[7] J.L. Yang, W.J. Ren, D. Li, W.J. Hu, B. Li, Z.D. Zhang, Supercond. Sci. Technol. 23 (2010) 025003.
[8] M. Tropeano, M.R. Cimberle, C. Ferdeghini, G. Lamura, A. Martinelli, A. Palenzona, I. Pallecchi, A. Sala, I. Sheikin, F. Bernardini, M. Monni, S. Massidda, M. Putti, Phys. Rev. B 81 (2010) 184054.
[9] Lei Wang, Yanpeng Qi, Zhaoshun Gao, Dongliang Wang, Xianping Zhang, Yanwei Ma, Supercond. Sci. Technol. 23 (2010) 025027.
[10] J. Prakash, S.J. Singh, A. Banerjee, S. Patnaik, A.K. Ganguli, Appl. Phys. Lett. 95 (2009) 262507.
[11] C.Q. Qu, Z.Y. Liu, Y.M. Lu, C.Z. Chen, C.B. Cai, A.H. Fang, F.Q. Huang, M.F. Wang, X.M. Xie, Journal of Modern Physics (in press).
[12] Ai Hua Fang, Fu Qiang Wang, Xiao Ming Xie, J. Am. Chem. Soc. 132 (2010) 3260–3261.
[13] V.P.S. Awana, Anand Pal, Arpita Vajpayee, R.S. Meena, H. Kishan, Mushahid Husain, R. Zeng, S. Yu, K. Yamaura, E. Takayama-Muromachi, J. Appl. Phys. 107 (2010) 09E146.
[14] Arpita Vajpayee, R. Jha, A.K. Srivastava, H. Kishan, M. Tropeano, C. Ferdeghini, V.P.S. Awana, 2011. arXiv:1101.5206.
[15] Monika Mudgel, L.S. Sharath Chandra, V. Ganesan, G.L. Bhalla, H. Kishan, V.P.S. Awana, J. Appl. Phys. 106 (2009) 033904.
[16] X. Wang, S.R. Ghorbani, G. Peleckis, S.X. Dou, Adv. Mater. 21 (2009) 236.
[17] Cong Ren, Zhao Sheng Wang, Hui Qian Luo, Huan Yang, Lei Shan, Hai Hu Wen, Phys. Rev. Lett. 101 (2008) 257006.
[18] C. Senatore, R. Flukiger, Phys. Rev. B 78 (2008) 054514.
[19] A. Yamamoto, A.A. Polyanskii, J. Jiang, F. Kametani, C. Tarantini, F. Hunte, J. Jaroszynski, E.E. Hellstrom, P.J. Lee, A. Gurevich, D.C. Larbalestier, Z.A. Ren, J. Yang, X.L. Dong, W. Lu, Z.X. Zhao, Supercond. Sci. Technol. 21 (2008) 095008.
[20] S.V. Chong, T. Goya, H. Yamaguchi, K. Kadowakia, 2009. arXiv:0908.3285.
[21] Xiyu Zhu, Huan Yang, Lei Fang, Gang Mu, Hai Hu Wen, Supercond. Sci. Technol. 21 (2008) 105001.
[22] G.F. Chen, Z. Li, G. Li, J. Zhou, D. Wu, J. Dong, W.Z. Hu, P. Zheng, Z.J. Chen, H.Q. Yuan, J. Singleton, J.L. Luo, N.L. Wang, Phys. Rev. Lett. 101 (2008) 057007.
[23] XiYu Zhu, Fei Han, Mu Gang, Peng Cheng, Bing Zeng, Hai Hu Wen, 2009. arXiv:0904.1732v2.
[24] V.P.S. Awana, R.S. Meena, A. Pal, A. Vajpayee, K.V.R. Rao, H. Kishan, Eur. Phys. J. B 79 (2011) 139–146.
[25] C. Cai, B. Hozapfel, J. Hänisch, L. Fernandez, L. Schultz, Phys. Rev. B 69 (2004) 104531.
[26] C.B. Cai, J.L. Liu, Z.Y. Liu, L.L. Ying, B. Gao, L. Peng, C.Z. Chen, J. Appl. Phys. 104 (2008) 23913.
[27] M. Tinkham, Introduction to Superconductivity, McGraw-Hill, New York, 1996.
[28] B. Khaykovich, E. Zeldov, D. Majer, T.W. Li, P.H. Kes, M. Konczykowski, Phys. Rev. Lett. 76 (1996) 2555.
[29] Athena S. Sefat, Rongying Jin, Michael A. McGuire, Brian C. Sales, David J. Sing, David Mandrus, Phys. Rev. Lett. 101 (2008) 117004.
[30] Huan Yang, Huiqian Luo, Zhaosheng Wang, Hai-Hu Wen, Appl. Phys. Lett. 93 (2008) 142506.
[31] Hyeong-Jin Kim, Yong Liu, Yoon Seok Oh, Seunghyun Khim, Ingyu Kim, G.R. Stewart, Kee Hoon Kim, Phys. Rev. B 79 (2009) 014514.