Evidence for antiferromagnetic order in $La_{2-x}Ce_xCuO_4$ from angular magnetoresistance measurements

K. Jin, X. H. Zhang, P. Bach, and R. L. Greene

Department of Physics and Center for Nanophysics & Advanced Materials, University of Maryland, College Park, Maryland 20742, USA

(Received 13 April 2009; revised manuscript received 8 June 2009; published 2 July 2009)

We investigated the in-plane angular magnetoresistance (AMR) of T'-phase $La_{2-x}Ce_xCuO_4$ thin films ($x=0.06-0.15$) fabricated by a pulsed laser deposition technique. The in-plane AMR with $\mathbf{H}\|ab$ shows a twofold symmetry instead of the fourfold behavior found in other electron-doped cuprates such as $Pr_{2-x}Ce_xCuO_4$ and $Nd_{2-x}Ce_xCuO_4$. The twofold AMR disappears above a certain temperature, T_D. The $T_D(x)$ is well above $T_c(x)$ for $x=0.06$ ($\sim$110 K), and decreases with increasing doping until it is no longer observed above $T_c(x)$ at $x=0.15$. This twofold AMR below $T_D(x)$ is suggested to originate from an antiferromagnetic or spin-density-wave order.

DOI: 10.1103/PhysRevB.80.012501 PACS number(s): 74.78.Bz, 73.43.Qt, 74.25.Fy, 74.72.−h

High-T_c superconductivity in the cuprates can be induced in the parent antiferromagnetic (AFM) insulator by the doping of either holes or electrons, corresponding to the formation of so-called hole- or electron-doped high-T_c cuprates. Neutron-scattering experiments have revealed that the Néel ordering is rapidly suppressed in the hole-doped cuprates[1] but persists to much higher doping levels in the electron-doped systems.[2,3] For the frequently studied hole-doped $La_{2-x}Sr_xCuO_4$ (LSCO) system, the long-range AFM ordering is completely destroyed at $x\sim0.02$ and the superconductivity appears above $x\sim0.05$.[1] It is reported that the long-range AFM ordering in electron-doped $Nd_{2-x}Ce_xCuO_4$ (NCCO) can extend up to $x\sim0.13$ (Ref. 3) or 0.15.[2] In electron-doped $Pr_{2-x}Ce_xCuO_4$ (PCCO) thin films, extensive transport studies strongly suggest a quantum phase transition at $x\sim0.16$ (Refs. 4–6) and a signature of static or quasistatic antiferromagnetism up to $x\sim0.15$.[7]

For NCCO and PCCO, the optimal doping is around 0.15 while for electron-doped $La_{2-x}Ce_xCuO_4$ (LCCO), the optimally doped region shifts to $x\sim0.09-0.11$.[8–10] This difference has been suggested to originate from a smaller antiferromagnetic exchange interaction in LCCO than that in NCCO and PCCO.[9] However, the magnetic nature of LCCO has never been determined experimentally because the T'-phase LCCO has only been synthesized in thin-film form, where neutron-scattering techniques cannot be used. In this Brief Report, we show that it is possible to determine the magnetic order in LCCO by using transport measurements.

In films, it has been shown that in-plane angular magnetoresistance (AMR) measurements can shed light on the magnetic order by probing the spin-charge coupling.[7,11–17] In lightly electron-doped $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ (PLCCO) ($x=0.01$) crystals, a fourfold in-plane AMR has been observed due to a magnetic-field-induced transition from a noncollinear to collinear Cu-spin arrangement in adjacent CuO_2 planes, with the "spin-flop" easy axis along the Cu-Cu ([110]) direction and hard axis along the Cu-O-Cu ([100]) direction.[11] The fourfold AMR has also been reported in underdoped NCCO (Refs. 12 and 13) and in PCCO up to $x\sim0.15$.[7] For PCCO, the temperature at which the fourfold AMR disappears is consistent with the static or quasistatic AFM ordering temperature determined by neutron scattering on large crystals.[7]

In other systems such as underdoped LSCO,[14] $YBa_2Cu_3O_{6+x}$ (YBCO) (Ref. 15) and the newly discovered $BaFe_{2-x}Co_xAs_2$ (Ba122),[16] a twofold in-plane AMR has been observed. Additionally, the coexistence of twofold and fourfold AMRs has been found in $Y_{0.2}Pr_{0.8}Ba_2Cu_3O_{7-\delta}$.[17] The twofold behavior in these systems has also been suggested to be associated with spin ordering. For Ba122, the temperature at which the twofold AMR disappears coincides with the spin-density-wave (SDW) ordering temperature.[16]

Here, we report the in-plane AMR of LCCO thin films with $x=0.06-0.15$. The in-plane AMR in LCCO shows a twofold symmetry, which is distinct from other electron-doped cuprates where a fourfold symmetry is found. This suggests that the spin-flop transition does not occur in LCCO. This twofold AMR disappears at a certain temperature, T_D. The $T_D(x)$ is well above $T_c(x)$ for $x=0.06$ ($\sim$110 K), and decreases with increasing doping until it is no longer observed above $T_c(x)$ at $x=0.15$. This characteristic $T_D(x)$ is suggested to originate from static AFM ordering or a SDW transition.

The c-axis-oriented LCCO films were deposited directly on (100) $SrTiO_3$ substrates by a pulsed laser deposition (PLD) technique utilizing a KrF excimer laser as the exciting light source. The films were deposited in an oxygen pressure of $\sim$230 mTorr at 700–750 °C. After the deposition, the films were annealed in vacuum between 10^{-5} and 10^{-6} Torr for 15–30 min to achieve the highest T_{c0} (zero-resistance superconducting transition temperature) and sharpest transition width for each doping. The samples used for this study are $\sim$2000 Å and patterned into a Hall-bar shape with the bridge typically along the a axis. The measurements were carried out using a Quantum Design PPMS 14 T magnet and the AMR was measured at temperatures above T_c due to the large in-plane upper critical field.

As shown in Fig. 1(a), the resistivity of LCCO films decreases with increasing x from 0.06 to 0.15. All the films are superconducting at low temperatures with the optimal doping around $x=0.10-0.11$. Compared with the LCCO films prepared by a dc magnetron sputtering (MS) method,[10] a PLD technique employing $BaTiO_3$ as buffer layer (PLD +BUFFER),[9] and a molecular-beam epitaxy (MBE) technique,[18] our samples show a broader superconducting

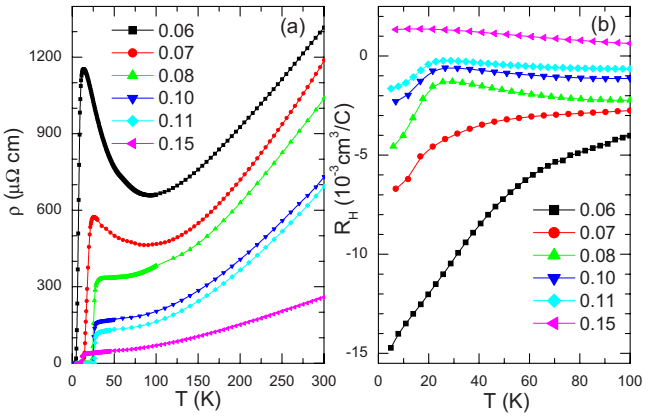

FIG. 1. (Color online) The temperature dependence of resistivity in (a) zero magnetic field and (b) Hall coefficient in 14 T of LCCO thin films with $x=0.06-0.15$.

region and a lower resistivity in the underdoped region. The T_{c0} of the optimal doping ($\sim$25 K) in our films is comparable to that found by the PLD+BUFFER method but slightly lower than that found by MS and MBE methods. Although different techniques and preparation processes result in these slight differences, the optimal doping region and the temperature dependence of resistivity do not change. To verify the doping concentrations, we also measured the Hall coefficient, obtained by subtracting the transverse Hall voltage in -14 T from that in 14 T ($\mathbf{H}\perp ab$ plane). The R_H gradually changes from negative to positive with increasing Ce concentration as seen in Fig. 1(b), consistent with previously reported behavior.[9,19]

For the in-plane AMR measurements, the film was rotated around the c axis with $\mathbf{H}\|ab$. The configurations $\mathbf{H}\|\mathbf{I}$ and $\mathbf{H}\perp\mathbf{I}$ are referred to $\theta=90°$ and 0°, respectively. Here θ is the angle between $\mathbf{H}$ and the direction normal to $\mathbf{I}$ [see inset of Fig. 4(a)]. We define the AMR as $[\rho(\theta)-\rho_{\min}]/\rho_{\min}$ and plot it as a function of θ for the films with $x=0.06-0.15$ at 14 T in Fig. 2, where the $\rho_{\min}$ represents the minimum resistivity when θ changes from 0° to 360°. Three main features are clearly seen: (i) only twofold symmetry exists in the underdoped and optimal doped LCCO films [Figs. 2(a)–2(e)], and it is not observed above T_c at $x=0.15$ [Fig. 2(f)]; (ii) the peak of AMR for $x=0.06$ appears at $\theta=90°$ and 270° corresponding to $\mathbf{H}\|\mathbf{I}$ at certain temperatures (discussed below) while it shifts 90° for other doping levels, appearing when $\mathbf{H}\perp\mathbf{I}$; (iii) the magnitude of the anisotropic in-plane AMR, $(\rho_{\max}-\rho_{\min})/\rho_{\min}$, is $\sim$0.01–0.1 % and decreases with increasing temperature. In PCCO, the anisotropic AMR is of the same magnitude as in LCCO but shows fourfold symmetry due to the anisotropic (fourfold) spin-flop field.[7]

It should be noted that the data shown in Fig. 2 are measured at 14 T but the twofold AMR is found with smaller magnitude at lower magnetic fields. In Fig. 3(a), magnetoresistivity curves at $\theta=90°$ ($\mathbf{H}\|\mathbf{I}$, defined as longitudinal MR, LMR) and 0° ($\mathbf{H}\perp\mathbf{I}$, defined as transversal MR, TMR) at 35 K for three dopings are plotted as a function of the magnetic field. The difference between TMR and LMR increases with increasing H so that the maximal AMR signal appears at 14 T. The twofold behavior is clear at lower temperatures, whereas at higher temperatures, it is not distinguishable due to smaller signal and much higher temperature fluctuations in the Quantum Design rotator. To probe the temperature at which the twofold behavior disappears (T_D), we used a Quantum Design resistivity sample stage and measured the magnetoresistivity of the samples with $x=0.06-0.11$ under the fixed configurations of $\mathbf{H}\perp\mathbf{I}$ and $\mathbf{H}\|\mathbf{I}$.

In Fig. 3(b), we show the field dependence of the TMR (solid symbols) and the LMR (open symbols) for $x=0.06$ sample at different temperatures. At low temperatures, the TMR is more negative than the LMR. With increasing temperature, the TMR becomes larger than the LMR and finally they overlap with each other. The difference between TMR and LMR at 14 T, $\delta\rho(14T)=[\rho_{\perp}(14T)-\rho_{//}(14T)]/\rho_0$ (where ρ_0 is the zero-field resistivity), is shown in the inset of Fig. 3(c). At $\sim$80 K, the $\delta\rho(14T)$ changes from negative to positive. Thus, the peak of the AMR of $x=0.06$ sample at $T>80$ K shifts 90°, showing a similar shape as that for higher doping levels. The $\delta\rho(14T)$ almost reaches zero above $T_D\sim110$ K, i.e., the twofold AMR disappears above 110 K.

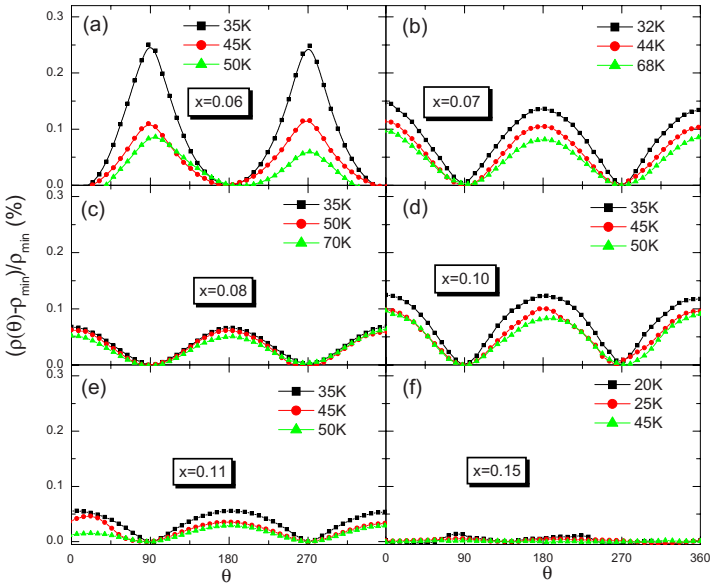

FIG. 2. (Color online) The in-plane angular magnetoresistivity of LCCO films with $x=0.06-0.15$ at 14 T. θ is the angle between $\mathbf{H}$ and the direction normal to the current ($\theta=0°$ corresponds to $\mathbf{H}\perp\mathbf{I}$). Note that here $\mathbf{I}$ is along the a axis.

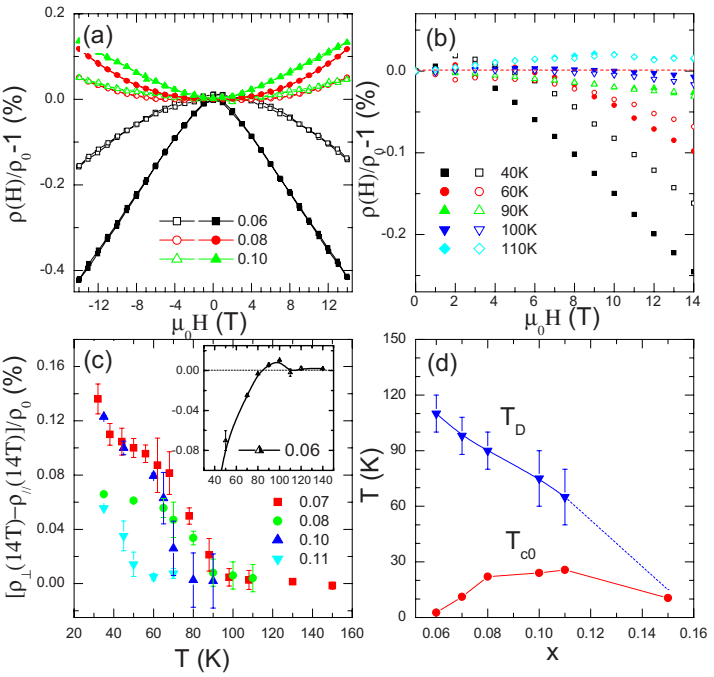

FIG. 3. (Color online) (a) The field dependence of the in-plane magnetoresistivity [open symbols LMR ($\mathbf{H}\|\mathbf{I}$), solid symbols TMR ($\mathbf{H}\perp\mathbf{I}$)] of LCCO with $x=0.06$, 0.08, and 0.10 at 35 K. (b) The LMR and TMR of $x=0.06$ at different temperatures. (c) The difference between LMR and TMR at 14 T. (d) $T_D(x)$ and $T_{c0}(x)$ of the LCCO. T_D and T_{c0} represent the temperature where the twofold AMR disappears and the zero-resistance superconducting transition temperature, respectively.

Using the same method, we also obtained T_D for other LCCO films. As shown in Fig. 3(c), the samples with $x=0.07$, 0.08, 0.10, and 0.11 show the same behavior: the difference between TMR and LMR gradually decreases with increasing temperature and almost disappears above $T_D(x)$. The $T_D(x)$ decreases with increasing doping until it is not observed above $T_{c0}(x)$ at $x=0.15$ as seen in Fig. 3(d). *This is the most important finding in this study.*

We shall now discuss the possible origin of this $T_D(x)$. In Ba122, the twofold AMR has been ascribed to SDW ordering.[16] We note that although a fourfold AMR has been reported in PCCO, it is likely that a twofold AMR also exists, as seen in the Fig. 4 of Ref. 7. Moreover, the twofold and fourfold AMRs seem to disappear roughly at the same temperature so the twofold AMR in PCCO may also be caused by the static or quasistatic AFM ordering.[7] Therefore, it is most likely that the twofold AMR in LCCO originates from an AFM or SDW order.

Other explanations based on magnetic ordering should also be considered. Ando *et al.*[15] suggested that field-induced ordering of stripes in YBCO resulted in the anisotropy of the in-plane magnetoresistivity. However, there is no clear evidence for stripes in electron-doped cuprates. Another possibility is based on the orthorhombic distortion. In the AFM ordered state, the crystal structure of lightly doped YBCO has a small orthorhombic distortion, which may lead to the in-plane anisotropic magnetoresistance in a magnetic field.[20] In cuprates containing only CuO_2 planes, a perfect tetragonal structure forbids this anisotropy.[21] For LSCO, there is a tetragonal-to-orthorhombic transition and the structure is orthorhombic in the AFM-ordered state.[1] The electron-doped cuprates are known to be in tetragonal structure. If the apical oxygens are not fully removed after annealing,[22] a local orthorhombic distortion may occur in LCCO as in LSCO, resulting in a twofold AMR. The LCCO films containing apical oxygens should be in T-phase structure.[18] The magnitude of AMR in underdoped LCCO is comparable to that in underdoped YBCO and LSCO. If the twofold AMR is caused by the orthorhombic distortion, the T-phase peaks should be detectable by x-ray diffraction, However, our x-ray diffraction data only show strong T'-phase ($00l$) peaks.

We should also discuss possible explanations without magnetic ordering. First, for our thin films, in which the in-plane anisotropy is of magnitude ~0.1%, one may expect that, if there is a small angle between the field and the ab plane, the c-axis field component can cause the difference. However, no asymmetric component can be resolved from the field sweep as shown in Fig. 3(a) so we can rule out this possibility.

Second, it is possible that the twofold AMR is caused by an extrinsic difference of resistivity for $\mathbf{H}\|\mathbf{I}$ vs $\mathbf{H}\perp\mathbf{I}$. However, if this was true, we should see twofold AMR in the films with $x=0.15$ as well and we do not. Moreover, we patterned two bridges, one along the a axis and another 45° offset, on the same sample and measured their in-plane magnetoresistivity simultaneously. As seen in Fig. 4, the θ is defined as the angle between $\mathbf{H}$ and the a axis. It is clear at 35 K that for both configurations, the magnetoresistivity of the $x=0.08$ sample at $\theta=45°$ is located between that at $\theta=0°$ and 90° as seen in Figs. 4(a) and 4(b). From the corresponding AMR [Figs. 4(c) and 4(d)], we can see that for the second configuration (bridge along [110]), the AMR at $\theta=45°$ ($\mathbf{H}\perp\mathbf{I}$) is almost equal to that at $\theta=135°$ ($\mathbf{H}\|\mathbf{I}$). We find the same result for other dopings and at other temperatures below T_D. So the origin of the twofold AMR is not due to the angle between $\mathbf{H}$ and $\mathbf{I}$.

Third, recent Nernst experiments have revealed that a large Nernst signal can exist at temperatures above T_c in both electron- and hole-doped cuprates, suggesting an extended phase fluctuation region.[23,24] However, this fluctuation region is dome shaped and does not monotonously decrease as doping increases. Moreover, in electron-doped PCCO and

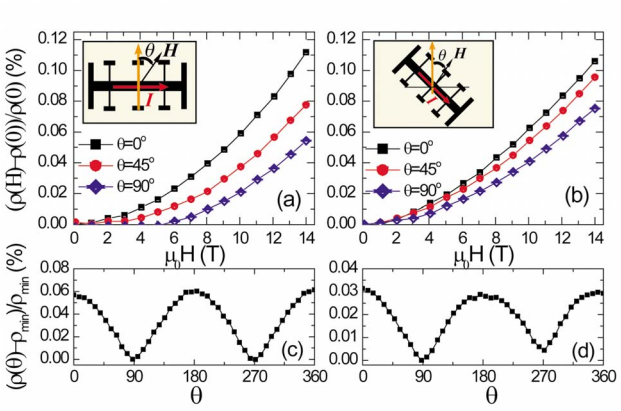

FIG. 4. (Color online) Magnetoresistivity measured at $\theta=0°$, 45°, and 90° with **I** (a) along the *a* axis and (b) along the diagonal direction for the samples with $x=0.08$ at 35 K. (c) and (d) are the corresponding AMR for these two configurations.

NCCO, the phase fluctuation temperature extends only up to ~30 K.[23,24] Thus, the $T_D(x)$ does not originate from superconducting phase fluctuations. Tunneling experiments in PCCO observed that a normal-state gap vanished at a certain temperature T^*.[25] This characteristic T^* is greater than T_c for the underdoped region and follows T_c on the overdoped side. However, the T^* is lower than 30 K so it is not responsible for the twofold AMR.

Fourth, there are two kinds of charge carriers in LCCO.[26] However, even if this could result in the in-plane anisotropy, it has been found that the magnetoresistance caused by the two-band feature is strongest near the optimal doping,[19] similar to PCCO.[23] So two kinds of charge carriers are unlikely to be the origin of the twofold AMR.

Thus, we suggest that the $T_D(x)$ originates from a static or quasistatic AFM or SDW ordering. We note that the $R_H(T)$ of underdoped and optimally doped LCCO films shows a downturn at certain doping-dependent temperatures but these temperatures are much lower than the $T_D(x)$. In PCCO films, the $R_H(T)$ also shows a downturn in underdoped and optimally doped regions at certain temperatures,[4] and these temperatures are also lower than the AFM temperatures.[7] The origin of the downturn of $R_H(T)$ is not understood at this time but it must be something other than the AMF ordering. In the LCCO films, the disappearance of $T_D(x)$, the insulator-to-metal transition,[27] and the formation of a large holelike Fermi surface[19] all occur at $x\sim0.15$, suggesting a relation among these behaviors. According to the above discussion, we infer that the disappearance of an AFM or SDW ordering causes all these behaviors.

In summary, we investigated the in-plane AMR of LCCO thin films ($x=0.06-0.15$) and observed a twofold symmetry. Unlike other electron-doped cuprates, a fourfold AMR caused by a spin-flop transition is not observed in the LCCO system. The twofold AMR disappears above a certain temperature, T_D. The $T_D(x)$ decreases with increasing doping ($T_D\sim110$ K for $x=0.06$), falling below $T_{c0}(x)$ at $x=0.15$. This newly disclosed characteristic $T_D(x)$ is suggested to originate from a static AFM or SDW ordering.

The authors would like to thank N. Butch, S. Saha, and J. Paglione for fruitful discussions. K.J. acknowledges W. Yu for technical help and useful discussions. This work is supported by the NSF under Grant No. DMR-0653535.

[1] M. A. Kastner *et al.*, Rev. Mod. Phys. **70**, 897 (1998).
[2] H. J. Kang *et al.*, Nature (London) **423**, 522 (2003).
[3] E. Motoyama *et al.*, Nature (London) **445**, 186 (2007).
[4] Y. Dagan, M. M. Qazilbash, C. P. Hill, V. N. Kulkarni, and R. L. Greene, Phys. Rev. Lett. **92**, 167001 (2004).
[5] Y. Dagan, M. C. Barr, W. M. Fisher, R. Beck, T. Dhakal, A. Biswas, and R. L. Greene, Phys. Rev. Lett. **94**, 057005 (2005).
[6] P. C. Li, F. F. Balakirev, and R. L. Greene, Phys. Rev. Lett. **99**, 047003 (2007).
[7] W. Yu, J. S. Higgins, P. Bach, and R. L. Greene, Phys. Rev. B **76**, 020503(R) (2007).
[8] Y. Krockenberger, J. Kurian, A. Winkler, A. Tsukada, M. Naito, and L. Alff, Phys. Rev. B **77**, 060505(R) (2008).
[9] A. Sawa, M. Kawasaki, H. Takagi, and Y. Tokura, Phys. Rev. B **66**, 014531 (2002).
[10] H. Wu *et al.*, Phys. Rev. B **73**, 104512 (2006).
[11] A. N. Lavrov, H. J. Kang, Y. Kurita, T. Suzuki, S. Komiya, J. W. Lynn, S.-H. Lee, P. Dai, and Y. Ando, Phys. Rev. Lett. **92**, 227003 (2004).
[12] A. I. Ponomarev *et al.*, JETP Lett. **81**, 394 (2005).
[13] T. Wu *et al.*, J. Phys.: Condens. Matter **20**, 275226 (2008).
[14] A. N. Lavrov, I. Tsukada, and Y. Ando, Phys. Rev. B **68**, 094506 (2003); Y. Ando, A. N. Lavrov, and S. Komiya, Phys. Rev. Lett. **90**, 247003 (2003).
[15] Y. Ando, A. N. Lavrov, and K. Segawa, Phys. Rev. Lett. **83**, 2813 (1999).
[16] X. F. Wang, T. Wu, G. Wu, R. H. Liu, H. Chen, Y. L. Xie, and X. H. Chen, New J. Phys. **11**, 045003 (2009); X. F. Wang, T. Wu, G. Wu, H. Chen, Y. L. Xie, J. J. Ying, Y. J. Yan, R. H. Liu, and X. H. Chen, Phys. Rev. Lett. **102**, 117005 (2009).
[17] V. Sandu *et al.*, J. Phys.: Conf. Ser. **51**, 231 (2006).
[18] M. Naito, S. Karimoto, and A. Tsukada, Supercond. Sci. Technol. **15**, 1663 (2002).
[19] K. Jin, B. Y. Zhu, B. X. Wu, L. J. Gao, and B. R. Zhao, Phys. Rev. B **78**, 174521 (2008).
[20] A. Jánossy, F. Simon, and T. Fehér, Phys. Rev. Lett. **85**, 474 (2000).
[21] F. C. Chou *et al.*, Phys. Rev. Lett. **78**, 535 (1997).
[22] P. Richard, G. Riou, I. Hetel, S. Jandl, M. Poirier, and P. Fournier, Phys. Rev. B **70**, 064513 (2004), and references therein.
[23] Y. Wang, L. Li, and N. P. Ong, Phys. Rev. B **73**, 024510 (2006).
[24] P. Li and R. L. Greene, Phys. Rev. B **76**, 174512 (2007).
[25] Y. Dagan, M. M. Qazilbash, and R. L. Greene, Phys. Rev. Lett. **94**, 187003 (2005).
[26] K. Jin *et al.*, Phys. Rev. B **75**, 214501 (2007).
[27] K. Jin, B. Y. Zhu, B. X. Wu, J. Vanacken, V. V. Moshchalkov, B. Xu, L. X. Cao, X. G. Qiu, and B. R. Zhao, Phys. Rev. B **77**, 172503 (2008).