Upper critical field of electron-doped $Pr_{2-x}Ce_xCuO_{4-\delta}$ in parallel magnetic fields

Pengcheng Li,[1] F. F. Balakirev,[2] and R. L. Greene[1]
[1]*Center for Superconductivity Research and Department of Physics, University of Maryland, College Park, Maryland 20742-4111, USA*
[2]*NHMFL, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
(Received 7 May 2007; published 25 May 2007)

We report a systematic study of the resistive superconducting transition in the electron-doped cuprates $Pr_{2-x}Ce_xCuO_{4-\delta}$ down to 1.5 K for magnetic field up to 58 T applied parallel to the conducting *ab* planes. We find that the zero-temperature parallel critical field $[H_{c2\|ab}(0)]$ exceeds 58 T for the underdoped and optimally doped films. For the overdoped films, 58 T is sufficient to suppress the superconductivity. We also find that the Zeeman energy $\mu_B H_{c2\|ab}(0)$ reaches the superconducting gap (Δ_0), i.e., $\mu_B H_{c2\|ab}(0) \simeq \Delta_0$, for all the dopings, strongly suggesting that the parallel critical field is determined by the Pauli paramagnetic limit in electron-doped cuprates.

DOI: 10.1103/PhysRevB.75.172508 PACS number(s): 74.72.Jt, 74.25.Ha, 74.25.Op

The upper critical field H_{c2} is a crucial parameter for high-T_c superconductors. It provides important information about the superconducting (SC) parameters, such as coherence length, SC gap, etc.[1] In past years, numerous transport experiments[2,3] on high-T_c cuprates in the $H \perp ab$ configuration have been reported and the H_{c2}-T diagrams have been established. A positive curvature in both cases was observed from the resistivity measurements, which is in contradiction to the expected low-temperature saturation in the Werthamer-Helfand-Hohenberg (WHH) theory.[4] The most likely reason for this is that the complicated H-T phase diagram of high-T_c superconductors includes a broad region of a vortex liquid state and strong SC fluctuations.[5] These properties are detrimental to the determination of H_{c2} from resistivity measurements. Recent high-field Nernst effect measurements[5] in hole-doped cuprates revealed a different H-T diagram when H_{c2} is determined by a loss of vorticity. A significant increase of H_{c2} and an extrapolation of $H_{c2}(T)$ to well above T_c were found. This observation was explained by the existence of a nonvanishing pairing amplitude well above T_c, while long-range phase coherence emerges only at T_c. H_{c2} could then be a measure of the onset of pairing amplitude.

Most of the H_{c2} results obtained so far on the cuprate superconductors are in the $H \perp ab$ configuration. The strong anisotropy, which would result in a much higher H_{c2} for magnetic field parallel to the conducting plane (*ab* plane), and the limitation of laboratory accessible magnetic fields make the $H_{c2\|ab}$ determination impossible for most of the cuprates. Nevertheless, a few $H_{c2\|ab}$ data have been reported.[6–9] An early work[10] that predicted $H_{c2\|ab}(T=0)$ for $YBa_2Cu_3O_{7-\delta}$ based on the initial slope $-dH_{c2}/dT$ near T_c was shown to be an overestimation by recent measurements.[6,7] The reason for this is that WHH theory only accounts for the orbital pair breaking, but in the $H \| ab$ orientation, the Pauli spin-pair-breaking effect could also be important. In fact, a recent measurement[11] on an underdoped $Bi_2Sr_2CuO_{6+\delta}$ in a pulsed magnetic field up to 52 T found that the Pauli paramagnetic limit could explain the H_{c2} for field parallel to the conducting layers.

Compared to the hole-doped cuprates, the electron-doped cuprates are distinctive for having a much lower $H_{c2\perp ab}$.[3] This implies a larger in-plane coherence length, and thus a smaller orbital critical field for H parallel to CuO_2 planes is expected. In addition, Nernst effect measurements have shown that electron-doped cuprates have much weaker SC fluctuations[12] compared to the hole-doped ones. In this Brief Report, we present systematic parallel critical-field measurements in the electron-doped $Pr_{2-x}Ce_xCuO_{4-\delta}$ (PCCO) for doping (x) throughout the SC region and establish the $H_{c2\|ab}$-T phase diagram. We find that the low-temperature parallel critical field is large (above 58 T at 4 K) for the underdoped and optimally doped films, while it is below 58 T for the overdoped films. We also find that the Zeeman splitting energy $\mu_B H_{c2\|ab}$ approaches the SC gap. Therefore, we conclude that the paramagnetic limit is the cause of the suppression of superconductivity in the $H \| ab$ configuration.

Five PCCO films with various dopings ($x = 0.13, 0.15, 0.16, 0.17, 0.19$) and with thickness of about 2500 Å were fabricated by pulsed laser deposition on $SrTiO_3$ substrates.[13] Since the oxygen content has an influence on both the SC and normal-state properties of the material,[14] we optimized the annealing process for each Ce concentration. The sharp transition and low residual resistivity are similar to our previous report,[15] which implies the high quality and well-defined doping and oxygen homogeneity of our films. Photolithography and ion-mill techniques were used to pattern the films into a standard six-probe Hall bar. Parallel field resistivity measurements were carried out using a 60 T pulsed magnetic field at the National High Magnetic Field Laboratory (NHMFL) in Los Alamos. Resistivity data traces were recorded on a computer using a high-resolution low-noise synchronous lock-in technique developed at NHMFL. The films were carefully aligned to ensure a parallel field (within ±1° with respect to the *ab* plane), and we found no signs of eddy current heating in the data.

Figure 1 shows the in-plane resistivity (ρ_{ab}) versus temperature in zero field and in 58 T for $H \| ab$ for all the films. The zero-field transition temperatures are 10.8, 21.3, 16.9, 14, and 10.4 K for x=0.13, 0.15, 0.16, 0.17, and 0.19, respectively. In the $H \perp ab$ field orientation, a field of order $H \leqslant 10$ T is enough to suppress the superconductivity, similar to a previous work.[3] However, when the field is aligned in the *ab* plane, the superconductivity is not completely destroyed in the underdoped x=0.13 and optimally doped

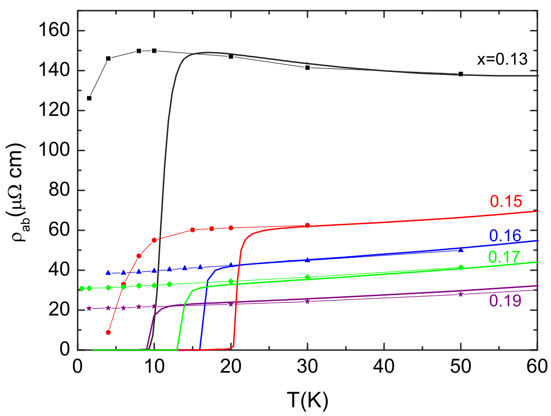

FIG. 1. (Color online) In-plane resistivity versus temperature in zero field (solid lines) and $H=58$ T applied parallel to the *ab* planes (filled symbols) in PCCO films with various Ce concentrations.

$x=0.15$ films even at 58 T, as seen in Fig. 1. In Fig. 2, we show $\rho_{ab}(H)$ for H parallel to the *ab* plane for the films $x=0.15$ and 0.16. Apparently, the normal state cannot be completely recovered in the optimally doped $x=0.15$ for $T \leqslant 10$ K. However, for the overdoped film $x \geqslant 0.16$, 58 T is sufficient to destroy the superconductivity even at the lowest temperature (1.5 K) measured. Compared to the $H \perp ab$ geometry,[3] a broader transition in $\rho_{ab}(H)$ is observed for the parallel field orientation. A similar behavior was found for the other dopings (not shown).

From the $\rho_{ab}(H)$ traces in Fig. 2, we can determine the resistive parallel critical field. However, the choice of a criterion remains arbitrary, mainly because of the curvature of the high-field flux-flow resistivity typical of all high-T_c superconductors. Following the schemes in the prior work[2,3] as presented in Fig. 2(b), we can determine the characteristic fields corresponding approximately to the onset of flux flow (H_{onset}) and a higher field corresponding to the complete recovery of the normal state (H_{100}). In Fig. 3(a), we show H_{onset} and H_{100} as a function of the reduced temperature (T/T_c) for $x=0.16$. The larger uncertainty of H_{100} is marked with larger error bars. In this figure, we also show the extracted value (H_{ext}) at the extrapolation point of the flux-flow region and the normal-state asymptote. We find that H_{ext} lies between H_{onset} and H_{100} and it is close to the field value determined from 90% of the normal-state resistivity. We note that the H_{ext} criterion has been regularly used as representing an acceptable determination of H_{c2} and we will adopt H_{ext} values as our estimate of $H_{c2\parallel ab}$.

In Fig. 3(b), we plot the characteristic field H_{ext} as a function of T/T_c for the other films (we note that T_c is taken from resistivity in a procedure similar to H_{ext}). In contrast to $H_{c2\perp ab}(T)$,[3] no low-temperature divergence or positive curvature is observed in the $H \parallel ab$ configuration for most of the films. Although the low-temperature $H_{c2\parallel ab}(T)$ behavior is unknown for $x=0.13$ and 0.15 due to the limit of our field, from the overdoped films data, a saturation seems to emerge at low temperature, which is similar to hole-doped cuprates.[7,11] From the *H*-*T* plots in Fig. 3, we can roughly extrapolate the curves to get $H_{c2\parallel ab}(0)$, and its doping dependence is shown in Fig. 4(a). A large zero-temperature critical field is found in the underdoped and optimally doped films, and a dramatic decrease of $H_{c2\parallel ab}(0)$ is observed for the overdoped films. A similar trend was found in the doping dependence of $H_{c2\perp ab}(0)$,[3,16] both $H_{c2\parallel ab}(0)$ and $H_{c2\perp ab}(0)$ decrease rapidly in the overdoped region compared to the underdoped, although the T_c of underdoped films drops even faster.

We have established an experimental parallel field *H*-*T* diagram for PCCO. Now, let us compare our data with theory. For most conventional superconductors, WHH theory can quantitatively explain the temperature dependence of the upper critical field. For the layered high-T_c cuprates, in the $H \perp ab$ configuration, it is found that the upper critical field is in good agreement with the WHH theory except for some unexplained low-temperature upward curvature.[11] This implies that the diamagnetic orbital effect dominates the paramagnetic spin effect in the destruction of the superconductivity. In the $H \parallel ab$ geometry, we attempted to compare our data with WHH theory (dotted lines in Fig. 3) by using the initial slopes of the *H*-*T* plots. As shown in Fig. 3, for the films near optimal doping ($x=0.15$ and 0.16), we found that WHH curves depart strongly from the experimental data at low temperatures. To show this here, we take $x=0.15$ as an example. The zero-temperature critical field obtained from the WHH formula $H_{c2}(0)=0.693(-dH_{c2}/dT)|_{T=T_c}T_c$ is about 170 T (using the initial slope value at T_c, $dH_{c2}/dT|_{T=T_c}=-11.5$ T/K), which is much larger than the extrapolated

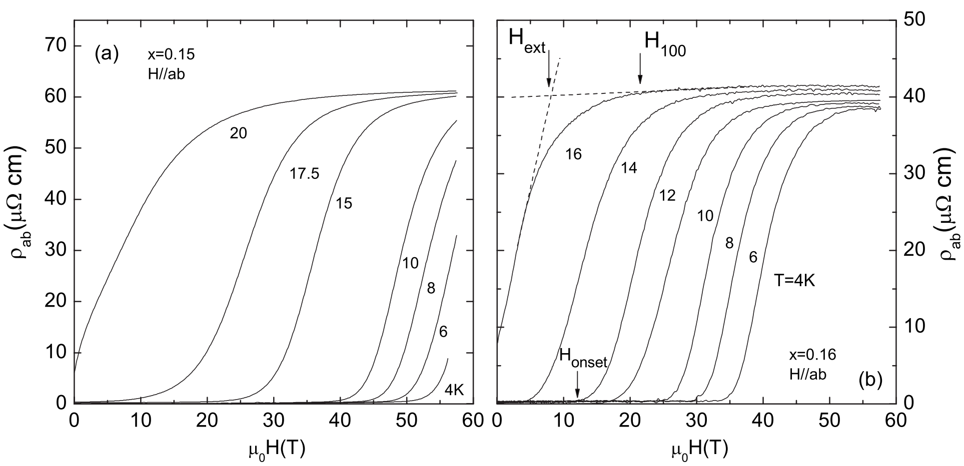

FIG. 2. In-plane resistivity versus magnetic field for $H \parallel ab$ plane for (a) $x=0.15$ ($T_c=21.3$ K) and (b) $x=0.16$ ($T_c=16.9$ K).

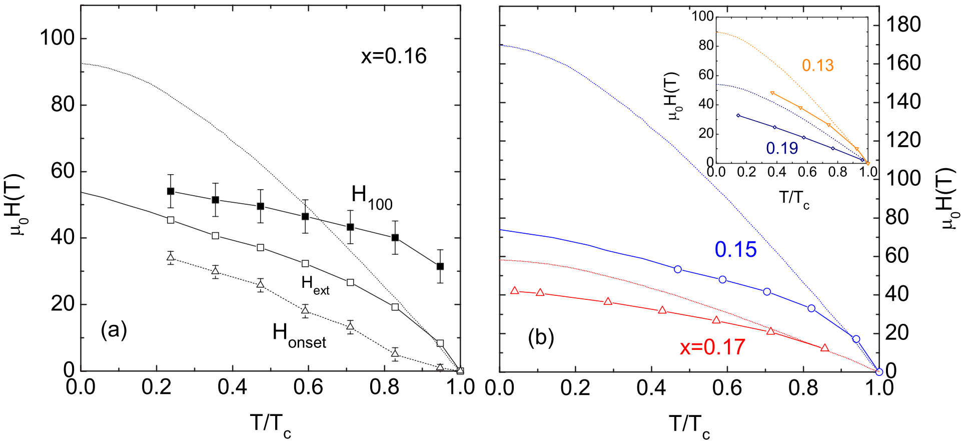

FIG. 3. (Color online) (a) Resistive characteristic fields H_{onset}, H_{ext}, and H_{100} for $H\|ab$ as a function of reduced temperature T/T_c for $x=0.16$, and (b) H_{ext} versus T/T_c for $x=0.15$ and 0.17. Inset shows the data for $x=0.13$ and 0.19. Dotted lines are fits to the WHH theory (Ref. 4). Solid lines are extrapolation based on a smooth $H(T)$ behavior.

value of 73 T. As seen in Fig. 3, the WHH value of $H_{c2}(0)$ is also larger than the experimental number for $x=0.13$ and 0.16. It appears that the WHH orbital theory only sets the upper bound of $H_{c2}(0)$ for these dopings. However, we find that for the overdoped films, $x=0.17$ and 0.19, the $H_{c2\|ab}(0)$ values are close to the WHH theoretical estimation.

For a layered superconductor, by neglecting the thickness of the conducting layers, Klemm *et al.*[17] predicted that the upper critical field would diverge for temperature below a certain value T^* where the out-of-plane coherence length ξ_c decreases to the value $d/\sqrt{2}$ (d is the distance between the conducting layers) and a dimensional crossover from three dimensions to two-dimensions would occur at low temperature. The critical magnetic field to decouple the layers at T^* was predicted to be $H_c=\phi_0/d^2\gamma$ ($\gamma=H_{c2\|ab}/H_{c2\perp ab}$). Experimentally, the low-temperature saturation in the H-T phase diagram for $H\|ab$ is contrary to this prediction and no trace of a dimensional crossover is observed. The predicted H_c, which is about 765 T for $x=0.15$ ($d=6$ Å and $\gamma\sim 8$, a similar number is found for the other dopings), is also very large. By considering the thickness (t) of the conducting layers, it has been found[18,19] that the parallel critical field can be rewritten as $H_{c'}=\sqrt{3}\phi_0/\pi t\xi_{ab}$. From our perpendicular critical-field data,[3] we can get the in-plane coherence length ξ_{ab} via the Ginzburg-Landau equation $H_{c2\perp ab}=\phi_0/2\pi\xi_{ab}^2$. Setting the corresponding values of $x=0.15$ [$t=3$ Å, $\xi_{ab}(0)=60$ Å], we find $H_{c'}=582$ T, which is still much higher than our measured value.

We now discuss paramagnetic (Pauli) limitation of the parallel critical field. In this case, the electron spins couple with the applied field, and when the spin Zeeman energy reaches the pair-breaking energy, the Cooper pair singlet state is destroyed. An early theory by Clogston and Chandrasekhar[20] estimated the paramagnetic limit based on the isotropic BCS theory and predicted the Pauli paramagnetic limit $H_P=\Delta_0/\mu_B\sqrt{2}$. Under the assumption $2\Delta_0=3.5k_BT_c$, we have $H_P(0)=1.84T_c\frac{T}{K}$. Applying this to our $x=0.15$ doping ($T_c=21.3$ K), we get $H_P(0)=39$ T. This is much smaller than our experimental value of 73 T. If we take $\Delta_0=4.3$ meV (maximum gap value) from the optics results,[16,21] then $H_P'(0)=53$ T. For the other dopings, we find that the Clogston theory also underestimates the measured values. This suggests that a simple BCS s-wave model for the paramagnetic limit is not valid for PCCO. This is not surprising since PCCO is believed to be a quasi-two-dimensional d-wave superconductor. Recent work by Yang and Sondhi[22] estimated the paramagnetic limit for a d-wave superconductor in a purely two-dimensional (2D) system by only considering the coupling of the spins of the electrons and the applied field and found that $H_P(0)=0.56\Delta_0/\mu_B$. This is even smaller than the s-wave case due to the existence of nodes in the gap function.

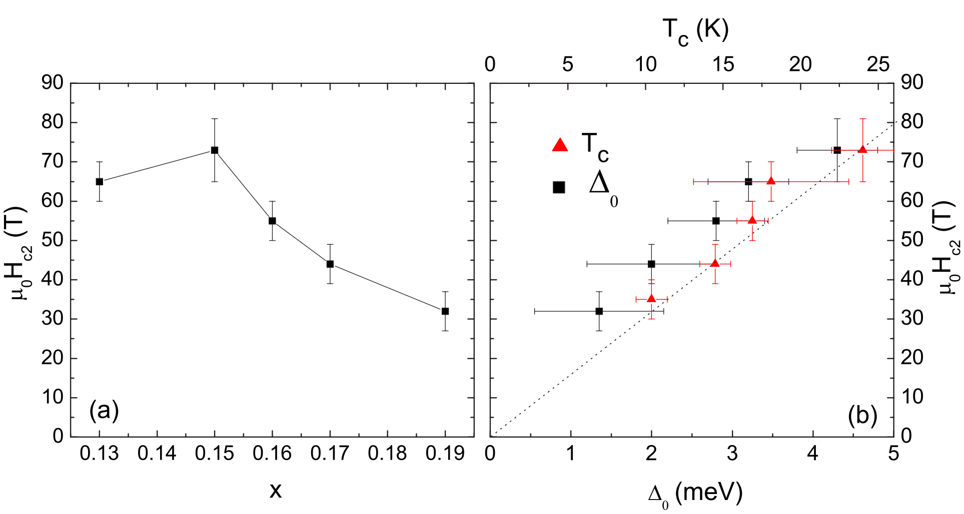

FIG. 4. (Color online) (a) Doping dependence of extrapolated $H_{c2\|ab}(0)$. (b) $H_{c2\|ab}(0)$ as a function of T_c and superconducting gap Δ_0.

The experimental critical field often exceeds the theoretical predictions for the Pauli limit, even in some conventional *s*-wave superconductors. To explain this, some other possibilities were introduced, such as spin-orbit coupling to impurities. It was found that the spin-orbit scattering enhances the Pauli critical field over the spin-only value for *s*-wave symmetry.[4,17] However, it has been shown[23] that the spin-orbit interaction significantly lowers the critical field for *d*-wave symmetry. Therefore, the enhancement of the parallel critical field in PCCO is most unlikely caused by the spin-orbit coupling.

Despite the discrepancy between theory and data, we find that our extrapolated $H_{c2\|ab}(0)$ can be scaled with both T_c and SC gap Δ_0. As seen in Fig. 4(b), $H_{c2\|ab}$ is linearly proportional to T_c and can be written in a Zeeman-like way, i.e., $k_B T_c = \frac{1}{4} g \mu_B H_{c2\|ab}(0)$, where $g=2$ is the electronic g factor and μ_B is the Bohr magneton. This suggests that the thermal energy at T_c and the electronic Zeeman energy at $H_{c2\|ab}(0)$ give the single energy scale required to destroy the phase coherence. We note that, for underdoped $x=0.13$ and optimally doped $x=0.15$, due to the SC fluctuation, we determined T_c from the temperatures at which the vortex Nernst effect disappears, which are 18 and 24 K for 0.13 and 0.15, respectively. This temperature is slightly higher than the resistive transition temperature.[12] For the overdoped films, both tunneling[24] and Nernst effect measurements show that the fluctuation is much weaker; therefore, T_c can be reliably taken from resistivity measurement. Meanwhile, if we compare the Zeeman energy and the maximum SC gap values obtained from optics,[16,21] we find that $g\mu_B H_{c2\|ab}(0) \simeq 2\Delta_0$, i.e., $\mu_B H_{c2\|ab}(0)/\Delta_0 \simeq 1$, as shown in Fig. 4. This strongly suggests that the magnetic Zeeman energy reaches the SC gap, and thus the superconductivity is destroyed. It has been shown that due to possible quantum fluctuations, the superconductivity can be destroyed within a Zeeman energy interval,[25] $\frac{1}{2}\Delta \leqslant \mu_B H_{c2\|ab} \leqslant 2\Delta$. Therefore, our results strongly suggest that the Pauli paramagnetic limit is responsible for the high-field depairing process.

Finally, it is worth mentioning that the SC gap to parallel critical-field ratio in some hole-doped cuprates was also found to be roughly 1.[6,11] It seems that in the layered quasi-2D cuprate superconductors, the parallel critical field is universally determined by the paramagnetic limit, suggesting that diamagnetic orbital pair-breaking effect is negligible compared to the spin effect due to a much shorter out-of-plane coherence length.

In summary, we measured $H_{c2\|ab}$ in electron-doped cuprates $Pr_{2-x}Ce_xCuO_{4-\delta}$ from the underdoped to the overdoped region. We found that the critical-field anisotropy $H_{c2\|ab}/H_{c2\perp ab}$ is about 8. We also found that the Zeeman energy $\mu_B H_{c2\|ab}(0)$ reaches the superconducting gap Δ_0, which strongly suggests that the Pauli paramagnetic limit is responsible for quenching superconductivity in electron-doped cuprates for H parallel to the CuO_2 planes.

P.C.L. and R.L.G. acknowledge the support of NSF under Grant No. DMR-0352735. The work in NHMFL is supported by NSF and DOE.

[1] M. Tinkham, *Introduction to Superconductivity*, 2nd ed. (McGraw-Hill, New York, 1996).

[2] Y. Ando, G. S. Boebinger, A. Passner, L. F. Schneemeyer, T. Kimura, M. Okuya, S. Watauchi, J. Shimoyama, K. Kishio, K. Tamasaku, N. Ichikawa, and S. Uchida, Phys. Rev. B **60**, 12475 (1999).

[3] P. Fournier and R. L. Greene, Phys. Rev. B **68**, 094507 (2003).

[4] N. Werthamer, E. Helfand, and P. Hohenberg, Phys. Rev. **147**, 295 (1966).

[5] Y. Wang, L. Li, and N. P. Ong, Phys. Rev. B **73**, 024510 (2006), and references therein.

[6] J. L. O'Brien, H. Nakagawa, A. S. Dzurak, R. G. Clark, B. E. Kane, N. E. Lumpkin, N. Miura, E. E. Mitchell, J. D. Goettee, J. S. Brooks, D. G. Rickel, and R. P. Starrett, Phys. Rev. B **61**, 1584 (2000).

[7] T. Sekitani, N. Miura, S. Ikeda, Y. H. Matsuda, and Y. Shiohara, Physica B **346**, 319 (2004).

[8] S. I. Vedeneev, A. G. M. Jansen, E. Haanappel, and P. Wyder, Phys. Rev. B **60**, 12467 (1999).

[9] A. S. Dzurak *et al.*, Phys. Rev. B **57**, R14084 (1998).

[10] U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, Phys. Rev. Lett. **62**, 1908 (1989).

[11] S. I. Vedeneev, Cyril Proust, V. P. Mineev, M. Nardone, and G. L. J. A. Rikken, Phys. Rev. B **73**, 014528 (2006).

[12] H. Balci, C. P. Hill, M. M. Qazilbash, and R. L. Greene, Phys. Rev. B **68**, 054520 (2003); Pengcheng Li and R. L. Greene (unpublished).

[13] E. Maiser, P. Fournier, J. Peng, F. Araujo-Moreira, T. Venkatesan, R. L. Greene, and G. Czjzek, Physica C **297**, 15 (1998); J. L. Peng, E. Maiser, T. Venkatesan, R. L. Greene, and G. Czjzek, Phys. Rev. B **55**, R6145 (1997).

[14] W. Jiang, S. N. Mao, X. X. Xi, X. Jiang, J. L. Peng, T. Venkatesan, C. J. Lobb, and R. L. Greene, Phys. Rev. Lett. **73**, 1291 (1994).

[15] Y. Dagan, M. M. Qazilbash, C. P. Hill, V. N. Kulkarni, and R. L. Greene, Phys. Rev. Lett. **92**, 167001 (2004).

[16] M. M. Qazilbash, A. Koitzsch, B. S. Dennis, A. Gozar, H. Balci, C. A. Kendziora, R. L. Greene, and G. Blumberg, Phys. Rev. B **72**, 214510 (2005).

[17] R. A. Klemm, A. Luther, and M. R. Beasley, Phys. Rev. B **12**, 877 (1975).

[18] F. Harper and M. Tinkham, Phys. Rev. **172**, 441 (1968).

[19] S. Vedeneev and Y. N. Ovichinnikov, JETP Lett. **75**, 195 (2002).

[20] A. Clogston, Phys. Rev. Lett. **9**, 266 (1962); B. Chandrasekhar, Appl. Phys. Lett. **1**, 7 (1962).

[21] C. C. Homes, R. P. S. M. Lobo, P. Fournier, A. Zimmers, and R. L. Greene, Phys. Rev. B **74**, 214515 (2006).

[22] K. Yang and S. L. Sondhi, Phys. Rev. B **57**, 8566 (1998).

[23] C. Grimaldi, J. Phys.: Condens. Matter **12**, 1329 (2000).

[24] Y. Dagan, M. M. Qazilbash, and R. L. Greene, Phys. Rev. Lett. **94**, 187003 (2005).

[25] I. L. Aleiner and B. L. Altshuler, Phys. Rev. Lett. **79**, 4242 (1997).