Upper critical field of electron-doped \( \Pr_{2-x} \text{Ce}_x \text{CuO}_{4-\delta} \) in parallel magnetic fields

Pengcheng Li\textsuperscript{1}, F. F. Balakirev\textsuperscript{2}, and R. L. Greene\textsuperscript{1}

\textsuperscript{1}Center for Superconductivity Research and Department of Physics, University of Maryland, College Park, Maryland 20742-4111
\textsuperscript{2}NHMFL, Los Alamos National Laboratory, Los Alamos, NM 87545

(Dated: February 1, 2008)

We report a systematic study of the resistive superconducting transition in the electron-doped cuprates \( \Pr_{2-x} \text{Ce}_x \text{CuO}_{4-\delta} \) down to 1.5 K for magnetic field up to 58 T applied parallel to the conducting \textit{ab}-planes. We find that the zero temperature parallel critical field \( (H_{c2}(0)) \) 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 \( \mu_B H_{c2}\textit{ab}(0) \) reaches the superconducting gap \( \Delta_0 \), i.e. \( \mu_B H_{c2}\textit{ab}(0) \approx \Delta_0 \), for all the dopings, strongly suggesting that the parallel critical field is determined by the Pauli paramagnetic limit in electron-doped cuprates.

PACS numbers: 74.25. Ha, 74.25.Op, 74.72.-h

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.\textsuperscript{1,2} In past years, numerous transport experiments\textsuperscript{3,4} on high-\( T_c \) cuprates in the \( \textit{H} \perp \textit{ab} \) 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.\textsuperscript{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.\textsuperscript{5} These properties are detrimental to the determination of \( H_{c2} \) from resistivity measurements. Recent high-field Nernst effect measurements\textsuperscript{6} 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 non-vanishing 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 \( \textit{H} \perp \textit{ab} \) configuration. The strong anisotropy, which would result in a much higher \( H_{c2} \) for magnetic field parallel to the conducting plane (\textit{ab}-plane), and the limitation of laboratory accessible magnetic fields makes the \( H_{c2} \) determination impossible for most of the cuprates. Nevertheless, a few \( H_{c2}\textit{ab} \) data have been reported.\textsuperscript{6,7,8} An early work\textsuperscript{9} predicted \( H_{c2}\textit{ab}(T = 0) \) for \( \text{YBa}_2\text{Cu}_3\text{O}_7-\delta \) based the initial slope, \(-dH_{c2}/dT \) near \( T_c \), was shown to be an overestimation by recent measurements.\textsuperscript{6,7} The reason for this is that WHH theory only accounts for the orbital pair breaking, but in the \( \textit{H} \parallel \textit{ab} \) orientation, the Pauli spin pair breaking effect could also be important. In fact, a recent measurement\textsuperscript{10} on an underdoped \( \text{Bi}_2\text{Sr}_2\text{CuO}_{6+\delta} \) 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 are distinctive for having a much lower \( H_{c2}\textit{ab} \). This implies a larger in-plane coherence length, and thus a smaller orbital critical field for \( \text{H} \parallel \text{CuO}_2 \) planes is expected. In addition, Nernst effect measurements have shown that electron-doped cuprates have much weaker SC fluctuations\textsuperscript{11} compared to the hole-doped. In this paper, we present systematic parallel critical field measurements in the electron-doped \( \Pr_{2-x} \text{Ce}_x \text{CuO}_{4-\delta} \) (PCCO) for doping \( (x) \) throughout the SC region and establish the \( H_{c2}\textit{ab}-T \) 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 \( \mu_B H_{c2}\textit{ab}(0) \) approaches the SC gap. Therefore, we conclude that the paramagnetic limit is the cause of the suppression of superconductivity in the \( \text{H} \parallel \textit{ab} \) configuration.

Five PCCO films with various doping \( (x=0.13, 0.15, 0.16, 0.17, 0.19) \) with thickness about 2500 Å were fabricated by pulsed laser deposition on \( \text{SrTiO}_3 \) substrates.\textsuperscript{12} Since the oxygen content has an influence on both the SC and normal state properties of the material,\textsuperscript{14} we optimized the annealing process for each Ce concentration. The sharp transition and low residual resistivity are similar to our previous report\textsuperscript{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 Lab (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 \( \pm 10^\circ \) with respect to the...
the normal state can not be completely recovered in the
ab-plane and we found no signs of eddy current heating
in the data.

Fig. 1 shows the in-plane resistivity ($\rho_{ab}$) versus tem-
perature in zero field and in 58 T for $H||ab$ for all the
films. The zero field transition temperatures are 10.8 K,
21.3 K, 16.9 K, 14 K, and 10.4 K for $x=0.13$, 0.15, 0.16,
0.17 and 0.19 respectively. In the $H\perp ab$ field orienta-
tion, a field of order $H_{c2}\perp ab<10$ T is enough to suppress the super-
conductivity, similar to previously work. However, when
the field is aligned in the ab-plane, the superconductiv-
ity is not completely destroyed in the underdoped $x=0.13$
and optimally doped $x=0.15$ 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 can not be completely recovered in the
optimally doped $x=0.15$ for $T\leq 10$ K. However, for the
overdoped film $x \geq 0.16$, 58 T is sufficient to destroy the superconductivity even at the lowest temperature (1.5 K)
measured. Compared to the $H\perp ab$ geometry, a broader
transition in $\rho_{ab}(H)$ is observed for the parallel field orien-
tation. A similar behavior was found for the other
dopings (not shown).

From the $\rho_{ab}(H)$ traces in Fig. 2, we can determine the resis-
tive parallel critical field. However, the choice of a
criterion remains arbitrary, mainly because of the cur-
vature of the high-field flux-flow resistivity typical of all
high-$T_c$ superconductors. Following the schemes in the
prior work 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 corre-
sponding to the complete recovery of the normal state
($H_{100}$). In Fig. 3(a), we show $H_{onset}$ and $H_{100}$ as a func-
tion 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 $H_{c2||ab}$.

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 $H_{c2||ab}(T)$, no low temperature divergence
or positive curvature is observed in the $H||ab$ configura-
tion for most of the films. Although the low tempera-
ture $H_{c2||ab}(T)$ 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. From the H-
T plots in Fig. 3, we can roughly extrapolate the curves
to get $H_{c2||ab}(0)$ 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 dra-
matic decrease of $H_{c2||ab}(0)$ is observed for the overdoped
films. A similar trend was found in the doping depen-
dence of $H_{c2||ab}(0)$, both $H_{c2||ab}(0)$ and $H_{c2||ab}(0)$
...
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 $H_{||ab}$ 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. This implies that the diamagnetic orbital effect dominates the paramagnetic spin effect in the destruction of the superconductivity.

In the $H_{||ab}$ 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

$$H_{c2}(0) = 0.693(\frac{-dH_{c2}}{dT})_{T=T_c}$$

is about 170 T (using the initial slope value at $T_c$, $dH_{c2}/dT \approx -11.5$ T/K), which is much larger than the extrapolated 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. predicted that the upper critical field would diverge for temperature below a certain value $T^*$ where the out-of-plane coherence length $\xi_z$ decreases to the value $d/\sqrt{2}(d$ is the distance between the conducting layers) and a dimensional crossover from 3D to 2D would occur at low temperature. The critical magnetic field to decouple the layers at $T^*$ was predicted to be $H_c = \phi_0 / d^2 \gamma$ ($\gamma = H_{c2||ab} / H_{c21||ab}$).

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 $\gamma \approx 8$), 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 $H_{c'} = \sqrt{3} \phi_0 / nt \xi_{ab}$. From our perpendicular critical field data\(^{29}\) we can get the in-plane coherence length $\xi_{ab}$ via the Ginzburg-Landau equation $H_{c2||ab} = \phi_0 / 2t \xi_{ab}^2$. 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 coupled with the applied field and when the spin Zeeman energy reaches the pairbreaking energy, the Cooper pair singlet state is destroyed. An early theory by Clogston and Chandrasekhar 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

$$H_P(0) = 1.84T_c^2$$

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\(^{16,17}\), 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 estimated the paramagnetic limit for a d-wave superconductor in a purely 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.

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.\(^{17}\) However, it has been shown\(^{22}\) 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 $\Delta_0$. 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., $k_BT_c = 4g\mu_B H_{c2||ab}$, where $g=2$ is the electronic g factor, $\mu_B$ 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 is 18 K and 24 K for 0.13 and 0.15, respectively. This temperature is slightly higher than the resistive transition temperature. For the overdoped films, both tunneling 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, we find that \( g\mu_B H_{c2}(0) \approx 2\Delta_0 \), i.e. \( \mu_B H_{c2}(0)/\Delta_0 \approx 1 \), 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 \( \frac{1}{2}\Delta \leq \mu_B H_{c2}(0) \leq 2\Delta \). Therefore, our results strongly suggest the Pauli paramagnetic limit is responsible for the high field deparing 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 one. 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 \( \text{Pr}_2-x\text{Ce}_x\text{CuO}_4-\delta \) from the underdoped to the overdoped region. We found that the critical field anisotropy, \( H_{c2}(ab)/H_{c2}(ab) \) is about 8. We also found that the Zeeman energy \( \mu_B H_{c2}(ab) \) reaches the superconducting gap \( \Delta_0 \), which strongly suggests that the Pauli paramagnetic limit is responsible for quenching superconductivity in electron-doped cuprates for \( H \parallel \text{the CuO}_2 \) planes.

PL and RLG acknowledge the support of NSF under Grant DMR-0352735. The work in NHMFL is supported by NSF and DOE.

1. M. Tinkham, *Introduction to Superconductivity*, 2nd edition (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, 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, G. Czjzek, Physica(Amsterdam) 297C, 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. Ovchinnikov, JETP Lett. 75, 195 (2002).
20. A. Clogston, Phys. Rev. Lett. 3, 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, 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).