Doping dependence of the upper critical field of electron-doped $Pr_{2-x}Ce_xCuO_4$ thin films

P. Fournier*

Canadian Institute of Advanced Research, Centre de recherche sur les propriétés électroniques de matériaux avancés Département de Physique, Université de Sherbrooke, Sherbrooke, Québec, Canada, JIK 2R1

R. L. Greene

Center for Superconductivity Research, Department of Physics, University of Maryland, College Park, Maryland 20742, USA (Received 25 October 2002; published 16 September 2003)

Using resistivity measurements as a function of applied magnetic field down to 38 mK, we evaluate the temperature dependence of the *c*-axis upper critical field $(H_{c2,\rho})$ for electron-doped $Pr_{2-x}Ce_xCuO_4$ thin films. We compare its temperature dependence to the irreversibility line as determined by ac susceptibility $[H_{IL}(T)]$ between 2 K and the transition temperature. For all Ce contents, $H_{c2,\rho}(T)$ presents an upward curvature, with no sign of the expected conventional saturation at low temperature, even down to 38 mK. The onset of resistivity follows closely the irreversibility line, and the general trend in temperature for $H_{c2,\rho}(T)$ is rather insensitive to the criterion used for its determination. Only a rough criterion corresponding to a full recovery of the normal-state resistivity for x=0.15 is bringing the characteristic field temperature dependence close to the expected description by Werthamer, Helfand and Hohenberg. Doping affects mainly the zero-temperature value of $H_{c2,\rho}$ and H_{IL} which are scaling with the critical temperature, but not the superconducting gap. The temperature dependence is very similar to that observed with the hole-doped cuprates, and underlines a similar physical origin related to the properties in the vortex-liquid phase and contributions of superconducting fluctuations.

DOI: 10.1103/PhysRevB.68.094507

PACS number(s): 74.25.Op, 74.25.Dw, 74.72.Jt, 74.25.Fy

I. INTRODUCTION

The determination of the upper critical field (H_{c2}) and many other parameters characterizing the superconducting state of high-temperature superconductors (HTSC's), similar to the magnitude of the gap, its anisotropy, and the magnitude of the penetration depth λ , is a crucial test for the validation of any theory trying to explain the phenomenon. A knowledge of the evolution of these parameters with carrier concentration is likely to be a stringent test of competing theories.¹⁻⁶ However, the strong anisotropy and the small in-plane coherence length of HTSC have a major impact on their field-temperature (H-T) phase diagram.⁷ It modifies significantly the nature of the phases observed with respect to the conventional picture.⁸ For the HTSC, a broad region of the phase diagram is occupied by a vortex liquid, its relative importance in the H-T plane being tuned mainly by the anisotropy⁹ and defects.^{7,10} Furthermore, superconducting fluctuations along the $T_c(H)$ line are also expected to contribute.7 The presence of the vortex liquid phase and these fluctuations in the hole-doped cuprates might be detrimental to the determination of H_{c2} .

Indeed, most reports using resistivity as a probe of the transition between the normal and the superconducting states have shown an anomalous positive curvature of $H_{c2,\rho}(T)$ for the hole-doped cuprates, even with the indications of possible divergence at low temperatures,^{11,12} in apparent contradiction with the expected low temperature saturation described theoretically by Werthamer, Helfand, and Hohenberg.¹³ Based on these observations, several theories, such as melting in proximity to a quantum critical point,¹⁴

scattering by magnetic impurities,¹⁵ localization of charged bosons,¹⁶ antiferromagnetic correlations in *d*-wave superconductors,¹⁷ mixing of $d_{x^2-y^2}$ and *s* or d_{xy} order parameters,¹⁸ or some granularity or inhomogeneity effects¹⁹ have been proposed to explain this apparent positive curvature of $H_{c2}(T)$. However, different approaches to the interpretation of the experimental data²⁰ and several other experiments^{21–24} seem to contradict this positive curvature and generate larger values of H_{c2} at a fixed temperature. This fact alone might indicate simply that resistivity is not an appropriate probe for the determination of H_{c2} .

Recent reports on the measurement of the high-field Nernst coefficient^{25,26} have revived the interest in the significance of these very different values of the measured H_{c2} . The Nernst effect shows that $H_{c2}(T)$ determined by a loss of vorticity (i.e., loss of entropy carried per vortex) is significantly higher than that measured by resistivity.²⁵ Moreover, $H_{c2}(T)$ extrapolates to a much higher temperature than the actual critical temperature T_c . This observation might indicate a nonzero (short-range) pairing amplitude at much higher temperature than T_c , while phase coherence is only setting in at T_c , i.e., H_{c2} would then be a measure of the actual onset of pairing amplitude, and not phase coherence (which occurs at T_c).

For hole-doped HTSC, the low-temperature behavior of $H_{c2}(T)$ extracted from resistivity mimics the temperature dependence of the irreversibility line close to the vortex solid to liquid crossover (transition). It was actually shown recently that the temperature dependence of $H_{c2,\rho}$, determined by resistivity measurements, can be correlated to the changes in anisotropy obtained by varying the carrier concentration,

as probed by the irreversibility line defined at the onset of flux flow.²⁷ It was further shown that the criterion used to determine H_{c2} , usually picked either by extrapolation or by using the field corresponding to a certain percentage of the normal state resistivity, has an impact on the actual temperature dependence deduced. This fact clearly underlines our limited understanding of the flux-flow regime deep into the vortex liquid state of these systems, and our need to clarify the nature of the vortex liquid for large driving (Lorentz) forces (at high fields) and the possible impact of superconducting fluctuations extending on a significant part of the phase diagram around the $T_c(H)$ line.

In the case of the electron-doped (n-type) cuprates R_{2-x} Ce_xCuO_{4+ δ} (with R=Pr, Nd, Sm), it was initially observed that the temperature dependence of H_{c2} seemed to be more conventional,²⁸ despite their huge anisotropy, of the order of $\rho_c / \rho_{xx} \approx 10^3 - 10^4$ [comparable to overdoped Bi₂Sr₂CaCu₂O₈ (Bi-2212)]. However, more recent reports show that the temperature dependence of H_{c2} is very similar to the *p*-type cuprates.^{29,30} All reports showed, however, that the absolute value of $H_{c2,\rho}$ at low temperatures is low com-pared to its closest hole-doped counterparts $La_{2-x}Sr_xCuO_4$ (LSCO)²⁷ For this reason, the *n*-type cuprates are members of a unique family of HTSC for which a moderate external dc magnetic field applied along the c axis, in the vicinity of 10 T, is apparently sufficient to suppress completely superconductivity for all dopings, i.e., from underdoping to overdoping. Reaching the normal state of these materials for all dopings becomes a powerful tool for probing the ground state (T=0) from which, we assume, arises the superconductivity.31,32

In this work, we present a systematic study of the doping dependence of the upper critical field of $Pr_{2-x}Ce_xCuO_4$ (PCCO) thin films down to 38 mK. We show that the choice of the criterion affects the temperature dependence of $H_{c2,\rho}$. This fact is further emphasized by the similarity of the $H_{c2,\rho}(T)$ lines and the irreversibility line determined by ac susceptibility. Only by choosing a very rough 100% criterion are we able to approach the expected Werthamer, Helfand, and Hohenberg (WWH) behavior. We use the extrapolated zero temperature value of H_{c2} to evaluate the doping dependence of the in-plane coherence length.

II. EXPERIMENTAL SETUP AND PROCEDURES

Our measurements were made on c-axis oriented PCCO thin films made by pulsed-laser deposition (PLD)³³ on three different substrates, namely, LaAlO₃ (LAO), SrTiO₃ (STO), and yttria-stabilized ZrO₂ (YSZ). Contrary to single crystals, it is possible to control very accurately the cerium concentration and even the oxygen content in thin films. It is still the only reliable and reproducible growth technique to target composition at will very accurately and thus studying in great details the phase diagram of these compounds.^{31,33} We should mention that similar measurements done on high quality single crystals with x=0.15 grown by the directional flux growth technique result in very similar behavior, comparable to data previously published.³⁴ However, because of the limited ability to vary efficiently the cerium content,

these optimal crystals give an incomplete dataset in view of our goal to span over the whole phase diagram. Despite the fact that all the results presented here were obtained for films on LAO, data obtained with different substrates do not differ significantly for the few compositions studied in more details in the temperature range from 1.5 K to T_c . Most of the $Pr_{2-x}Ce_xCuO_4$ (PCCO) films were patterned into Hall bars using ion milling (width=275 μ m, length=2.17 mm, and thickness=3000 to 6000 Å) allowing a battery of transport measurements in the normal and superconducting states.^{35,36} Unpatterned films were also studied, giving the same behavior as patterned ones, thus ruling out ion milling as a source of modification of the properties of the films.

All the films were mounted on a rotating probe which allowed angular-dependent measurements in a variabletemperature insert (1.5 to 300 K) with magnetic fields up to 10 T. All the data presented here correspond to field always applied parallel to the c axis (i.e., perpendicular to the copper-oxygen planes). Our measurements for fields along the planes revealed a large anisotropy in H_{c2} similar to pre-vious reports.^{37–39} A few of the thin film samples were also measured in a dilution refrigerator down to 38 mK, and up to 12 T. Particular care was taken to avoid self-heating of the samples below 2 K by varying the applied currents (at least by one order of magnitude). Susceptibility with an ac field of 5 Oe parallel to the *c* axis (normal to the surface of the films) as a function of an applied dc field also parallel to the c axis at fixed temperature was measured using a physical properties measurement system (PPMS) from Quantum Design. We use the imaginary part of the susceptibility to determine the irreversibility field (see below).

III. RESULTS AND DISCUSSION

To introduce our results and analysis, we first focus on our data for the x=0.15 thin films. In Fig. 1(a), we show the in-plane resistivity (ρ_{xx}) as a function of applied magnetic field at several temperatures down to 38 mK. These traces of $\rho_{xx}(H)$ are roughly parallel with respect to each other in the transition region (flux-flow regime).^{28–30} The specific temperature dependence of this flux-flow regime will be discussed further below. One can see anomalies at high fields and very low temperatures which suggest the possible presence of a slight distribution of critical temperatures. This might have an influence on the exact determination of H_{c2} at very low temperature depending on the criterion (see below). In the case of single crystals, these anomalies would be attributed to inhomogeneity of the composition, which is controlled by both cerium and oxygen contents. Since we are working with thin films, in which case compositional inhomogeneity should be ruled out,³³ another likely candidate is strain induced by lattice mismatch with the substrate. This could lead to a thin layer at the substrate-film interface with slightly different properties than the rest of the film. This has yet to be confirmed by exploring more thoroughly the data obtained with films on different substrates with different lattice mismatches and various thicknesses at these low temperatures.40

From the $\rho_{xx}(H)$ traces, the choice of a criterion to deter-



FIG. 1. (a) Field dependence of the resistivity at various temperatures down to 38 mK for PCCO x = 0.15 thin film (T = 38, 605 mK, 1.7, 3, 5, 7.5, 10, 12.5, 15, 17.5, 20 K). (b) Definition of the various characteristic fields for a fixed temperature. The double-arrow represents the uncertainty on H_{100} .

mine the upper critical field ($H_{c2,\rho}$) remains arbitrary, mainly because of the very wide rounded curvature of the high field flux-flow resistivity typical of all HTSC's. The onset of resistivity (ρ =0) is *independent* of current density in our range of measurements, and the corresponding field should follow closely the irreversibility line (see below). Beyond the onset, a flux-flow regime is established until the resistivity reaches asymptotically a value close to its normal-state limit.

In order to clarify the role of the criterion in the determination of H_{c2} and its possible link to the irreversibility line, we adopt a scheme introduced by Ando *et al.*²⁷ for $La_{2-x}Sr_xCuO_4$ and also presented in Fig. 1(b). The same procedure was also used in another report on Bi₂Sr₂CuO₆ (Bi-2201).¹² We choose the field values corresponding approximately to the onset of flux flow (H_0) and to 50% (H_{50}) of the high field normal state value. We also add the extracted value (H_{ext}) at the extrapolation point of the fluxflow regime and the normal-state asymptote, and a value at



FIG. 2. (a) Temperature dependence of the characteristic fields for x=0.15. (b) Same data plotted on a log-log scale to show the temperature dependence close to T_c . H_0 : solid circles, H_{50} : solid squares, H_{ext} : solid diamonds, H_{100} : solid triangles, H_{IL} : open circles, and H_R : open squares. Solid lines are guides to the eye. Dashed line is the expected *s*-wave clean limit temperature dependence deduced from the value of $H_{c2}(0)$ (see text).

much higher field corresponding to our estimate of the total recovery of the normal state (H_{100}) . Here, H_{100} presents a fairly large uncertainty which we have underlined by the hatched region (about 1 T wide for x=0.15). H_{ext} and H_{50} have been used regularly as representing an acceptable determination of H_{c2} in HTSC's.²⁸ Here, we argue that this assumption is wrong because H_{ext} and H_{50} correlates with the irreversibility line (and the onset of resistivity), in agreement with previous observations in *p*-type cuprates.^{12,27}

In Fig. 2(a), we show the temperature dependence of these characteristic fields. Except for H_{100} at temperatures close to T_c , the temperature dependence of these characteristic fields show an upward (positive) curvature, similar to most reports.²⁹ In a sense, this is not surprising since the reported resistivity anisotropy of the electron-doped cuprates is in the vicinity of $\rho_c/\rho_{ab} \approx 100-1000$ (see Ref. 41), rela-



FIG. 3. (a) Field dependence of the imaginary part of ac susceptibility at various temperatures down to 2 K for x=0.15. (b) A close-up allowing the determination of $H_{\rm IL}$.

tively close to that of overdoped Bi-2212.⁴² We should underline that $H_{100}(T)$ tends to zero at a temperature very close to T_c for x = 0.15.

To complete our analysis of the x=0.15 films, we present in Fig. 3(a) the imaginary part of the ac susceptibility data for a comparable film (same T_c , same normal-state resistivity and Hall coefficient). On a close-up scale [Fig. 3(b)], we show that the imaginary part of the susceptibility reaches a field-independent value at a field which we define as the irreversibility field $H_{\rm IL}$ (our criterion corresponds roughly to the field at which the susceptibility reaches the noise level of the PPMS setup). The extracted irreversibility fields ($H_{\rm IL}$) for several temperatures down to 2 K are shown in Fig. 2(a). In the same figure, we show the field H_R corresponding to the vanishing point (within our noise level) of the real part of the susceptibility (data not shown here).⁴³ One can see that the temperature dependence of the irreversibility line (and H_R) follows closely the resistivity onset line (H_0).⁴⁴

Actually, the temperature dependence observed for $H_{\rm IL}$ is very similar to that observed for *all* the characteristic fields determined from resistivity, except H_{100} . The latter observa-



FIG. 4. Field dependence of the first derivative of the resistivity with respect to the field at various temperatures for PCCO x = 0.15. Same temperatures as Fig. 1(a).

tion can be confirmed simply by plotting these characteristic fields as a function of $(1 - T/T_c)$ on a log-log scale as seen in Fig. 2(b). In the case of hole-doped cuprates, this plot is often used to extract the power-law temperature dependence of the irreversibility line close to T_c .⁴⁵ In our case, this particular figure clearly demonstrates that, except for H_{100} , all the other characteristic fields, including H_{ext} , are following closely the irreversibility line. In fact, the temperature dependence of all these "characteristic" fields close to T_c is following $H \approx H_0 (1 - T/T_c)^n$ with $n \approx 1.5$. This value of n is consistent with previous report of exponent related with the temperature dependence of the vortex glass transition and the critical behavior in the transition³⁹ in NdCeCuO. Because the irreversibility line is sensitive to the out-of-plane vs in-plane anisotropy⁷ and that H_{c2} should not be (with H remaining parallel to the c axis), these characteristic fields are unlikely to be linked to the real value of H_{c2} . Thus, we conclude that the determination of H_{c2} is severely affected by the peculiar vortex state and probably by the fluctuation region around $H_{c2}(T)$.

Interestingly, it seems that our $H_0(T)$ [and $H_{II}(T)$] and $H_{100}(T)$ lines are converging at low temperatures and close to T_c . If we assume that the $H_{c2}(T)$ is very close to $H_{100}(T)$, the behavior observed in our resistivity data should simply reflect the fact that the largest field interval between the irreversibility field and the real value of H_{c2} is observed for intermediate temperatures $(T \approx T_c/2)$. This is confirmed by another systematic behavior observed for all our high quality thin films with narrow transitions, independent of the doping level. In Fig. 4, we can observe a clear evolution of the sharpness of the transition in the flux-flow regime as a function of temperature: the maximum slopes are observed for temperatures close to T_c and towards T=0. This observation suggests that the irreversibility line is approaching H_{c2} at $T \rightarrow 0$ and T_c , and that it sets apart strongly at intermediate temperatures (as $H_{\rm IL}$ gets significantly smaller than H_{c2}). From this data, we conclude once again that our ability to determine the upper critical field in the intermediate temperature range is affected severely by the special nature of vortex dynamics, in particular for the field range over which a vortex liquid phase is possible and beyond into the critical fluctuation region. Because the anisotropy in PCCO is of the order of 10^3 (see Ref. 41), the liquid phase must surely span over a wide field range in the *H*-*T* diagram.⁴⁶ Figure 4 highlights also the presence of the anomalies at low temperatures and high magnetic fields also observed in the $\rho_{xx}(H)$ traces at 38 and 605 mK (Fig. 1). This might be indicative of a slight T_c distribution which could not be detected at higher temperatures. An obvious conclusion from this figure is that the value of H_{80} (or H_{ext}) extrapolated at T=0 is likely to be very close to the real $H_{c2}(T=0)$.

In Fig. 5, we show the field dependence of the in-plane resistivity for several compositions at fixed temperatures. From this data, we can extract the same characteristic fields as in the x = 0.15 case. These fields are shown also in Fig. 5. Data for x = 0.13 and 0.20 were taken only down to 1.6 K and data for several temperatures had to be ignored because we did not reach large enough fields. A very similar field dependence is found for all compositions, including the variation of the maximum flux-flow slope with temperature (not shown). However, one can see for example in the case of x = 0.17 that the transition to the normal state occurs on a wider range of applied magnetic field than for x = 0.15. Using the same criteria as before, one can estimate the characteristic fields described above. However, it is difficult to evaluate a "well-behaved" value of H_{100} for all Ce contents apart from x=0.15. In fact, this is best illustrated for x = 0.17 [Fig. 5(f)] for which $H_{100}(T)$ seems to converge to a much higher T_c value than the one measured by ac susceptibility and resistivity.⁴⁷ This trend could be the same as the one observed in LSCO by Wang et al. using the Nernst effect.²⁵ We should mention here that our measurements done on several PCCO x=0.17 thin films annealed in situ with different procedures could not clarify the possible contribution from inhomogeneity which could lead to a distribution of T_c 's and broadened transition, and that this feature seems to be very robust whatever is done to affect the quality of the films (including using different substrates).

All the data for all Ce contents seem to provide a clear connection between the irreversibility field and the characteristic fields determined by resistivity. In fact, if one plots the characteristic fields on log-log scales as in Fig. 2(b), we conclude that all of the characteristic fields are linked to the temperature dependence of $H_{\rm IL}$. In Ref. 27, a clear correlation was shown between the line defined by the onset of resistivity (our H_0) and the corresponding anisotropy, with respect to the effect on the various characteristic fields determined in a similar fashion. In our case, it is impossible to estimate the doping dependence of the anisotropy with our thin films, and only a rough estimate of the anisotropy using the existing data⁴¹ on single crystals can be achieved. Because doping proceeds by varying both cerium and oxygen contents, we prefer to avoid a direct parallel between single crystal and thin film data, even though their cerium content is the same.

If one uses instead the irreversibility line (or the related

value of H_{ext}) as a measure of the anisotropy (cast into the prefactor of the temperature dependence close to T_c), one cannot really conclude anything about a change of anisotropy from Figs. 5(b), 5(d), 5(f), and 5(h). In fact, our evaluation of the irreversibility line for x = 0.13 and 0.17 seems to actually show that the prefactor H_0 [in $H \approx H_0 (1 - T/T_c)^n$], and thus the anisotropy, does not vary significantly over the doping range from x = 0.13 to 0.17. This is also understandable from the fact that the narrow range of doping over which we are studying the phenomenon is likely not sufficient to observe a significant change in anisotropy. We conclude that a definite test of a possible correlation between the anisotropy and the irreversibility line (and the characteristic fields) has yet to be done, but it will remain a difficult task because of the narrow range of doping for which superconductivity is observed combined to severe materials issues with single crystals.

One might use the zero-temperature H_{c2} evaluated from extrapolation of the irreversibility line or any of the characteristic fields [assuming they all converge to $H_{c2}(0)$] to follow its Ce concentration dependence. This is of considerable interest since, within the previous assumption, the in-plane coherence length can be evaluated as a function of cerium doping. These values and their trend with doping can then be compared to competing theories. In Table I, we present several parameters one can evaluate from our data and combine them with data from a recent report on point-contact tunneling.⁴⁸

Our measurements down to very low temperatures allow us to pinpoint (with an uncertainty of about 0.5 T) the value of $H_{c2}(0)$ for a few Ce contents, while it can only be done with a larger uncertainty (about 1 T) for x=0.13 and 0.20 (measured only down to 1.5 K). In Fig. 6, we show $H_{c2}(0)$, T_c and the gap (normalized using the BCS weak-coupling expression $2\Delta_0/3.52k_B$) as a function of cerium content. One can see a convincing correlation between T_c and $H_{c2}(0)$. However, there is no such agreement between H_{c2} and the gap. This is in complete contradiction with the expectations from the clean limit ($H_{c2} \propto \Delta_0^2$) or the dirty limit (H_{c2} $\propto \Delta_0/l$, where l is the mean-free path). One should mention here that the doping dependence of the superconducting gap measured by point-contact tunneling⁴⁸ is reminescent of the behavior observed in hole-doped cuprates.^{49,50}

In Table I, we include the in-plane coherence length evaluated within two different scenarios: (1) clean *s*-wave $(H_{c2} = \phi_0/2\pi\xi_0^2)$,⁸ and (2) dirty *s*-wave $(H_{c2} = \phi_0/2\pi\xi_0 l)$, where *l* is the mean-free path).^{8,51} Here, ϕ_0 is the quantum of flux and ξ_0 is the in-plane coherence length at T=0. One could also add to the previous list the expression for the clean *d*-wave case.^{52,53} However, we did not include it since the corresponding correction deduced from the linearized gap equation is small and our data cannot help us to discriminate realistically between the clean *s*- and *d*-wave cases, in particular with our error bars as large as 10%. From our measurements, coherence lengths as low as 56 Å can be estimated (x=0.14, clean *s*-wave scenario).

One can also link the zero-temperature value of H_{c2} and the slope $\partial H_{c2}/\partial T$ at T_c . In the s-wave clean limit



FIG. 5. Resistivity as a function of applied magnetic field at fixed temperatures and the characteristic fields for x = 0.13 in (a) and (b), x = 0.14 in (c) and (d), x = 0.17 in (e) and (f), and x = 0.20 in (g) and (h). Solid lines are guides to the eye. Arrows point toward the value of T_c corresponding to zero resistance and the peak center of the imaginary part of the ac susceptibility.

TABLE I. Values of several parameters measured on these $Pr_{2-x}Ce_xCuO_4$ thin films as a function of cerium content.

x	<i>T</i> _c [K]	<i>H</i> _{c2} (0) [T]	ξ ₀ clean s [Å]	$\rho_{xx}(0)$ $\mu\Omega$ -cm	k _F l _{el}	MFP [Å]	ξ ₀ dirty s [Å]	Δ_0 [meV]
0.13	10	6.5	70	475	3.2	4.5	1085	6.5
0.14	21	10	56	125	12	17	185	
0.15	23	9	60	70	21.5	31	115	4.4
0.17	13	7	67	40	37.5	54	85	1.75
0.20	6	2.5	113	35	42.9	61	208	

for example, we have $\partial H_{c2}/\partial T|_{T \to T_c} = -\phi_0/C\xi_0^2 T_c = -2\pi H_{c2}(0)/CT_c$, where C = 3.44 (see Ref. 8). In principle, the same value for ξ_0 should be obtained from both evaluations. Of course, this is not expected from our data for all the criteria, *except* maybe for H_{100} . In Fig. 2(a), we show the expected $H_{c2}(T)$ close to T_c in the *s*-wave clean limit as a dashed line assuming $H_{c2}(0) \sim 9$ T. It approaches the value of H_{100} and is another hint at the inappropriate criteria one might use to determine H_{c2} from resistivity.

Serious questions remain, however, on the validity of our 100% criterion and if our estimate is really in proximity to the real H_{c2} , in particular when considering the behavior observed for x = 0.17. Moreover, it is not quite sure that we can use the determination of H_{c2} in the clean limit in our case since the estimated mean-free path one can determine from the low-temperature resistivity³¹ for x = 0.15 is about 30 Å [$k_F l \approx 23$, with $k_F \approx 0.70$ Å⁻¹ (Ref. 54)]. For this reason, we have included in Table I the estimation of the coherence length in the dirty limit as a reference.

It is clear from the above interpretations that resistivity is far from being the appropriate probe for the determination of a thermodynamic property such as H_{c2} for the HTSC. This underlines the need for experimental techniques measuring bulk thermodynamic properties such as the specific heat, the Nernst effect or thermal conductivity.

IV. SUMMARY

In summary, we observe the same temperature dependence for the upper critical field of *n*-type cuprate superconductors as found in p-type cuprates provided one uses a criterion of extrapolation from the flux-flow and normal-state regimes. However, we show clear evidence of a direct correlation between the irreversibility line and this particular characteristic field and conclude that it is unlikely to be the true upper critical field. We show that a rough estimate of the field at which the material reaches its true normal state leads



FIG. 6. Comparison of the cerium content dependence of the measured parameters characterizing the superconducting state of PCCO. Solid circles: T_c , open circles: $H_{c2}(0)$, and solid squares: Δ_0 . Solid and dashed lines are guides to the eye.

to a temperature dependence approaching the conventional behavior described by Werthamer, Helfand, and Hohenberg, giving us an in-plane coherence length as low as 56 Å in the electron-doped cuprates. Thermodynamic, force-free experiments which can help determine the upper critical field are needed to confirm our interpretation.

Note added in proof. Since our first submittal, new reports on the Nernst effect clearly demonstrate the need for better probe than resistivity for H_{c2} determination.^{55,56} The data on similar PCCO thin films clearly indicates traces of vortices at fields up to about H_{100} ,⁵⁵ not H_{ext} . It is even showed that $H_{c2}(0)$ pursue its rise for underdoping contrary to our observation extracted from the resistivity data. This shows once again the inadequacy of the resistivity as a probe of H_{c2} . Additional work by Yang *et al.*⁵⁶ supports further this observation.

ACKNOWLEDGMENTS

We thank J.-L. Peng and Z.Y. Li for the PCCO target preparation, P. Mohanty and R.A. Webb for the high-field and low temperature measurements, and J. Gauthier and M.-E. Gosselin for some additional recent measurements on thin films. The work in Maryland was supported by the NSF Condensed Matter Physics Division under Grant No. DMR 01-02350. The work in Sherbrooke is supported by the Canadian Institute of Advanced Research, the Canadian Foundation for Innovation, the Natural Sciences and Engineering Research Council of Canada, and the Fondation of the Université de Sherbrooke.

⁶V. Emery, S. Kivelson, and O. Zachar, Phys. Rev. B **56**, 6120 (1997).

^{*}Electronic address: patrick.fournier@physique.usherb.ca

¹P.A. Lee and X.G. Wen, Phys. Rev. Lett. 78, 4111 (1997).

²A.S. Alexandrov, Phys. Rev. B 53, 2863 (1996).

³P.W. Anderson, *The Theory of High Tc Superconductivity* (Princeton University Press, Princeton, New Jersey, 1997).

⁴C.M. Varma, Phys. Rev. B 55, 14 554 (1997).

⁵P. Monthoux and D. Pines, Phys. Rev. B **47**, 6069 (1993).

⁷G. Blatter, M.V. Feigel'man, V.B. Geshkenbein, A.I. Larkin, and V.M. Vinokur, Rev. Mod. Phys. 66, 1125 (1994).

⁸M. Tinkham, *Introduction to Superconductivity* 2nd ed. (McGraw-Hill, New York, 1996).

⁹This neglects the changes of the coherence length as a function of

doping for the hole-doped cuprates.

- ¹⁰L. Civale, A.D. Marwick, T.K. Worthington, M.A. Kirk, J.R. Thompson, L. Krusin-Elbaum, Y. Sun, J.R. Clem, and F. Holtzberg, Phys. Rev. Lett. **67**, 648 (1991).
- ¹¹C.C. Kim, E.F. Skelton, S.B. Qadri, V.M. Browning, M.S. Osofsky, M.E. Reeves, and D.H. Liebenberg, Phys. Rev. B 49, 13 075 (1994).
- ¹²S.I. Vedeneev, A.G.M. Jansen, E. Haanappel, and P. Wyder, Phys. Rev. B **60**, 12 467 (1999).
- ¹³N.R. Werthamer, E. Helfand, and O.C. Hohenberg, Phys. Rev. 147, 295 (1966).
- ¹⁴G. Kotliar and C.M. Varma, Phys. Rev. Lett. 77, 2296 (1996).

¹⁵Y.N. Ovchinnikov and V.Z. Kresin, Phys. Rev. B **52**, 3075 (1995).

- ¹⁶A.S. Alexandrov, V.N. Zavaritsky, W.Y. Liang, and P.L. Nevsky, Phys. Rev. Lett. **76**, 983 (1996).
- ¹⁷A. Pérez-González and R.A. Brito-Orta, Phys. Rev. B **64**, 214502 (2001).
- ¹⁸T. Koyama and M. Tachiki, Physica C 263, 25 (1993).
- ¹⁹ V.B. Geshkenbein, L.B. Ioffe, and A.J. Millis, Phys. Rev. Lett. **80**, 5778 (1998).
- ²⁰S.H. Han, C.C. Almasan, M.C. de Andrade, Y. Dalichaouch, and M.B. Maple, Phys. Rev. B 46, 14 290 (1992).
- ²¹G. Blumberg, M. Kang, and M.V. Klein, Phys. Rev. Lett. **78**, 2461 (1997).
- ²²A. Carrington, A. Mackenzie, and A. Tyler, Phys. Rev. B 54, 3788 (1996).
- ²³J.L. O'Brien et al., Phys. Rev. B 61, 1584 (2000).
- ²⁴U. Welp, W.K. Kwok, G.W. Crabtree, K.G. Vandervoort, and J.Z. Liu, Phys. Rev. Lett. **62**, 1908 (1989).
- ²⁵ Y. Wang, N.P. Ong, Z.A. Xu, T. Kakeshita, S. Uchida, D.A. Bonn, R. Liang, and W.N. Hardy, Phys. Rev. Lett. 88, 257003 (2002).
- ²⁶C. Capan, K. Behnia, J. Hinderer, A.G.M. Jensen, W. Lang, C. Marcenat, C. Marin, and J. Flouquet, Phys. Rev. Lett. 88, 056601 (2002).
- ²⁷Y. Ando, G.S. Boebinger, A. Passner, L.F. Schneemeyer, T. Kimura, M. Okuya, S. Watauchi, J. Shimoyama, N.I.S.U.K. Kishio, and K. Tamasaku, Phys. Rev. B **60**, 12 475 (1999).
- ²⁸Y. Hidaka and M. Suzuki, Nature (London) **338**, 635 (1989).
- ²⁹I.W. Sumarlin, S. Skanthakumar, J.W. Lynn, J.L. Peng, Z.Y. Li, W. Jiang, and R.L. Greene, Phys. Rev. Lett. 68, 2228 (1992).
- ³⁰F. Gollnik and M. Naito, Phys. Rev. B 58, 11 734 (1998).
- ³¹P. Fournier, P. Mohanty, E. Maiser, S. Darzens, T. Venkatesan, C.J. Lobb, G. Czjzek, R.A. Webb, and R.L. Greene, Phys. Rev. Lett. **81**, 4720 (1998).
- ³²R.H. Hill, C. Proust, L. Taillefer, P. Fournier, and R.L. Greene, Nature (London) **414**, 711 (2001).
- ³³E. Maiser, P. Fournier, J.L. Peng, F.M. Araujo-Moreira, T. Venkatesan, R.L. Greene, and G. Czjzek, Physica C 297, 15 (1998).
- ³⁴M. Suzuki and M. Hikita, Phys. Rev. B **41**, 9566 (1990).
- ³⁵P. Fournier, E. Maiser, and R.L. Greene, in *The Gap Symmetry and Fluctuations in High Temperature Superconductors*, edited by G. Deutscher, J. Bok, D. Pavuna, and S. Wolf (Plenum Press, New York, 1998), p. 145.
- ³⁶P. Fournier (unpublished).
- ³⁷Y. Dalichaouch, B.W. Lee, C.L. Seaman, J.T. Markert, and M.B. Maple, Phys. Rev. Lett. 64, 599 (1990).
- ³⁸M. Suzuki and M. Hikita, Phys. Rev. B **41**, 9566 (1990).
- ³⁹J. Hermann, M.C. de Andrade, C.C. Almasan, R.P. Dickey, M.B.

Maple, W. Jiang, S.N. Mao, and R. Greene, Phys. Rev. B 54, 3610 (1996).

- ⁴⁰Our numerous tests have not yet clarified this issue. Using Rutherford backscattering, a thin layer of materials, ~100 Å with a different oxygen content could be detected from time to time on top of the films. A bridge made of a 4000 Å thick film which was progressively thinned down showed no obvious dependence of the critical temperature and properties with thickness down to 300 Å. However, we have observed a significant dependence of T_c with the thickness of the as-grown films, which could be just an indication that the in-situ annealing conditions at the end of the growth might have to be optimized for every single thicknesses. This problem is now under more thorough investigation.
- ⁴¹M. Brinkmann, H. Somnitz, H. Back, and K. Westerholt, Physica C 217, 418 (1993).
- ⁴²M.F. Crommie and A. Zettl, Phys. Rev. B 43, 408 (1991).
- ⁴³We use the imaginary part to determine the irreversibility field because its nonzero value in the mixed state is an indication that minor hysteresis loops are obtained at the corresponding fields. The imaginary part of the susceptibility will reach (and remain at) zero at the closure (and beyond) of the major hysteresis loop, i.e., at the irreversibility field.
- ⁴⁴Because the irreversibility line approaches H_0 , we expect the flux-flow regime to be totally independent of the shape of the sample (no demagnetization effects) since pinning is irrelevant beyond $H_{\rm II}$.
- ⁴⁵A. Houghton, R.A. Pelcovits, and A. Sudbo, Phys. Rev. B 40, 6763 (1989).
- ⁴⁶The impact of the small coherence length and the strong anisotropy on the phase diagram can be cast into a single parameter of reference, namely the Ginzburg number Gi. One can evaluate Ginzburg number for PCCO to the be Gi = $[T_c/H_c^2(0)\epsilon\xi^3(0)]^2/2\approx 10^{-2}$ where the anisotropy factor is $\epsilon \approx 1/30$ to 1/100 in the case of the electron-doped cuprates. This value of Gi is comparable to the ones obtained with the other high temperature superconductors and implies that thermal fluctuations are relevant for the determination of the magnetic properties. Even in the case of the 2D Ginzburg number (Gi^{2D}), the value obtained is comparable to that of YBCO. This emphasizes the impact of thermal fluctuations on the actual phase diagram. For more details, see Ref. 7.
- ⁴⁷The observations of this apparent larger T_c value was observed on several PLD x = 0.17 thin films from two different sources. This underlines the great reproducibility of our results and the easy control on stoichiometry, which is a function of both cerium and oxygen contents.
- ⁴⁸A. Biswas, P. Fournier, M.M. Qazilbash, V.N. Smolyaninova, H. Balci, and R.L. Greene, Phys. Rev. Lett. 88, 207004 (2002).
- ⁴⁹N. Miyakawa, J.F. Zasadzinski, L. Ozyuzer, P. Guptasarma, D.G. Hinks, C. Kendziora, and K.E. Gray, Phys. Rev. Lett. 83, 1018 (1999).
- ⁵⁰C. Kendziora and A. Rosenbergh, Phys. Rev. B **52**, 9867 (1993).
- ⁵¹The mean-free path (MFP) *l* was evaluated assuming a cylindrical Fermi surface and conventional Boltzmann transport. The residual resistivity, obtained from extrapolation from the high temperature metallic resistivity was used to evaluate $k_F l = hc_0 / \rho_{xx} e^2$ where $c_0 \approx 6.05$ Å. The extrapolation neglects the corrections on conductivity at low temperatures giving a nonmetallic behavior. Using $k_F \sim 0.70$ Å⁻¹ extracted from data of Ref.

54, one gets the values of MFP and coherence lengths shown in Table I.

- ⁵² W. Kim, J.-X. Zhu, and C.S. Ting, Phys. Rev. B 58, 607 (1998).
 ⁵³ H. Won and K. Maki, Phys. Rev. B 53, 5927 (1996).
- ⁵⁴N.P. Armitage et al., Phys. Rev. Lett. 87 147003 (1993).
- ⁵⁵H. Balci, R. Buhdani, and R. Greene, cond-mat/0303469.
- ⁵⁶Y. Wang, S. Ono, Y. Onose, G. Gu, Y. Ando, Y. Tokura, S. Uchida, and N.P. Ong, Science 299, 86 (2003).