

## Critical fields and fundamental lengths in a superconducting electron-doped $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$ single crystal

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Both critical fields  $H_{c1}$  and  $H_{c2}$ , together with the intrinsic superconducting parameters  $\lambda$  and  $\xi$ , have been obtained from a set of careful systematic measurements of isothermal magnetization curves in a  $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  single crystal with  $\mathbf{H}\parallel c$ . The analysis of the low-field regime has been performed, taking into account demagnetizing effects. A zone of complete diamagnetic screening is observed, followed by a transition region in which magnetic-flux penetration starts according to the predictions of Bean's critical-state model. The  $H_{c2}(T)$  values, as well as a  $\kappa \leq 10$ , have been obtained from the reversible part of the magnetization curve, in the high-field region, by using Abrikosov's model. The estimated values at  $T=0$  K for the different intrinsic superconducting parameters are  $H_{c1}(0) \approx 920$  Oe,  $H_{c2}(0) \approx 70$  kOe,  $\lambda_{ab}(0) \approx 700$  Å, and  $\xi_{ab}(0) \approx 70$  Å. The significance of these intrinsic superconducting parameters, the rather low  $\kappa$  value, and a large Sommerfeld constant are discussed and compared with the corresponding values of other cuprates.

### INTRODUCTION

$L_{2-x}\text{Ce}_x\text{CuO}_{4-y}$  superconducting cuprates<sup>1</sup> are singular within the cuprates family because of the sign of the charge carriers, which is negative at high temperatures. Their relatively low critical temperature ( $\sim 20$  K) is an indication of longer coherence length and consequently the reported upper critical fields [i.e.,  $H_{c2}(0) = 67$  kOe (Ref. 2)] are much smaller than for most of  $p$ -type superconductors and lie well within the available experimental field range. Therefore, the electron-doped materials offer a unique opportunity to explore completely the  $H$ - $T$  phase diagram of cuprate superconductors.

Measurements of  $H_{c1}(T)$ ,  $H_{c2}(T)$ , and the characteristic lengths  $\xi_{ab}(T)$  and  $\lambda_{ab}(T)$  on electron-doped superconducting oxides are scarce and sometimes contradictory. This is specially true in the lower critical-field case, for which values ranging from 1.8 kOe (Ref. 3) to 26 Oe (Ref. 4) have been reported. It is clear that magnetic determinations of  $H_{c1}(T)$  are severely affected by the material's quality and thus good single crystals are needed. Alternative methods to determine  $\lambda_{ab}(T)$ , such as muon-spin rotation, have failed because of the strong paramagnetism of the rare-earth ions.<sup>5</sup>

On the other hand, in high-temperature superconductor (HTSC) cuprates, measurements of  $H_{c2}(T)$  are somewhat obscured by the existence of the "irreversibility line." It is now well established that in HTSC's measurements of the resistivity in a magnetic field do not provide  $H_{c2}(T)$  but are more related to the irreversibility line ( $H_{irr}, T$ ). An experimental manifestation of the crossing of the ( $H_{irr}, T$ ) line when increasing temperature is a pronounced broadening of the resistive transition in a magnetic field, which is commonly observed in the HTSC cuprates.<sup>6</sup> It has been argued that the important broadening of  $R(T, H)$  is caused by the "giant flux creep" which takes place at high temperature because of the ratio

$U/kT$  (where  $U$  is a measure of the flux-pinning strength) is small.<sup>6</sup> In electron-doped materials, the resistance curves  $R(T, H)$  shift essentially parallel<sup>2</sup> without a significant broadening, thus indicating that flux creep occurs at slower rates and therefore  $U/kT$  is higher. Indeed, ac susceptibility measurements on  $\text{Nd}_{1.85}\text{Ce}_{0.85}\text{CuO}_4$  single crystals<sup>7</sup> provide indications that in these materials  $U$  is notably higher than the values typically found in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  (BSCCO) and even  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO).<sup>8</sup> Consequently, one might speculate that in the electron-doped materials, the irreversibility line and the upper critical field should lie close together. However, de Andrade *et al.*<sup>9</sup> have reported that for a  $\text{Sm}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$  single crystal, the resistively determined  $H_{c2}(T)$  is placed well above the irreversibility line determined from zero-field-cooling (ZFC) and field-cooling (FC) magnetization measurements. In addition, both  $H_{c2}(T)$  and ( $H_{irr}, T$ ) show a similar upward curvature close to  $T_c$ , which can not be explained within a Ginzburg-Landau theory. Therefore, it is of prime importance to determine  $H_{c2}(T)$  from the reversible part of the magnetization curve at constant temperature because it reflects an equilibrium state not affected by flux motion effects.<sup>10</sup>

In this paper we report a complete set of measurements of  $H_{c1}(T)$  and  $H_{c2}(T)$  in a superconducting single crystal of  $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$ , for  $\mathbf{H}\parallel c$ . Specifically, we have measured the magnetization  $M(H, T)$  at several temperatures and fields. From the low-field region data  $H_{c1}(T)$  has been extracted, whereas the reversible region close to  $M=0$  has been analyzed in the framework of the Abrikosov model, which provides a measure of  $H_{c2}(T)$  and the Ginzburg-Landau parameter  $\kappa$ . We show that all the available experimental data point to a rather low ( $\leq 10$ )  $\kappa$  value, in contrast to most HTSC  $p$ -type cuprates, which are extreme type-II superconductors [ $\kappa \geq 55$  (Ref. 11)]; we will see that this lower  $\kappa$  results from larger coherence

length and shorter penetration depth values. Comparison of  $H_{c2}(T)$  and  $(H_{irr}, T)$  will also be provided.

The measured  $H_{c1}(T)$  and  $H_{c2}(T)$  data allow an evaluation of the thermodynamic critical field  $H_c(0)$  and thus through BCS theory the Sommerfeld constant  $\gamma$ . It turns out that  $\gamma$  is rather large ( $\approx 60$  mJ/mol K<sup>2</sup>), placing the electron-doped cuprates close to the strongly coupled A15 alloys and the C<sub>60</sub> fullerene in the  $T_c$ - $\gamma$  diagram.<sup>12</sup>

### EXPERIMENTAL

The field-dependent dc magnetization of a Pr<sub>2-x</sub>Ce<sub>x</sub>CuO<sub>4-y</sub> single crystal has been studied by using a Quantum Design superconducting quantum interference device (SQUID) magnetometer with a scan length of 3 cm, under magnetic fields up to 55 kOe.

A platelet shaped single crystal with dimensions 1.80 × 0.95 × 0.03 mm<sup>3</sup>, grown by the self-flux method and with an estimated Ce content around  $x = 0.15$ ,<sup>13</sup> has been used. A critical temperature of  $T_c = 19.2$  K was obtained from the diamagnetic onset of a ZFC excursion at  $H_a = 1.6$  Oe. A transition width of less than 2 K was also determined from these data.

All data reported here correspond to a field  $\mathbf{H}$  applied perpendicular to the Cu-O planes, which are parallel to the largest faces of the crystals. Severe effects associated to the demagnetizing field can be present for this particular geometry and the internal field  $\mathbf{H}_i$  in the crystal may differ considerably from the applied field  $\mathbf{H}_a$ . This effect is especially important for lower critical-field determinations and it should be carefully considered. In the simplest case of a uniformly magnetized system and for  $\mathbf{H}_a$  applied along a principal axis it is  $\mathbf{H}_i = \mathbf{H}_a - 4\pi D\mathbf{M}$ , where  $\mathbf{M}$  is the magnetization and  $D$  is the demagnetizing factor.

The estimation of the demagnetizing factor has been carried out in three different ways. An effective  $D$  value can be determined by assuming a complete magnetic shielding at the lowest fields. With this assumption, and using the theoretical density of the crystal ( $\rho = 7.36$  g/cm<sup>3</sup>) and the volume deduced from its mass ( $m = 0.43$  mg), it turns out that  $D = 0.946$ . According to Kunchur and Poon,<sup>14</sup>  $D$  can also be extracted from the fit of the  $M(H)$  curve for  $H \leq H^*$ , where  $H^*$  is the field at which full flux penetration occurs. By using this method of analysis, we deduced  $D = 0.944$  in good agreement with the value estimated from the previous method. Finally, from the sample geometry, by using the ellipsoidal approximation, a value of  $D = 0.97$  is obtained. Because of the fact that the geometrically evaluated  $D$  values are not reliable for nonellipsoidal and nonisotropic materials, in the following we will use  $D = 0.946$  as a value of the demagnetizing factor.

The remanent magnetic field in the superconducting magnet is a serious problem when performing zero-field-cooling (ZFC) measurements at very low applied magnetic fields on samples with large demagnetization factors. Consequently, all low-field data have been obtained after cancellation of the remanent field ( $\mathbf{H}_{rem} \leq 0.2$  Oe), measured by a Pb shot.

On the other hand, the paramagnetic rare-earth contri-

bution to the measured magnetization can be easily evaluated by measuring the differential susceptibility  $\chi_i$  in the high-field region ( $30 \text{ kOe} < \mathbf{H} < 55 \text{ kOe}$ ), where the material becomes normal. It is straightforward to note that this contribution is essentially temperature independent in Pr<sub>2-x</sub>Ce<sub>x</sub>CuO<sub>4-y</sub>, because of the singlet ground state of the Pr<sup>3+</sup> ions,<sup>15</sup> and much lower than in Nd<sub>2-x</sub>Ce<sub>x</sub>CuO<sub>4-y</sub>. Thus, the raw magnetization data have been corrected by using  $M(T, H) = M_{raw}(T, H) - \chi_i(T)H$ . Notice that this method can be used because  $H_{c2}(5 \text{ K}) < 30 \text{ kOe}$  (see below).

### RESULTS

Figure 1 shows the temperature dependence of the dc susceptibility. Both ZFC (zero-field-cooled) and FC (field-cooled) were measured with the magnetic field ( $H_a = 1.6$  Oe) perpendicular to the Cu-O planes. The onset of diamagnetism occurs at  $T_c = 19.2$  K. The ZFC curve shows full diamagnetic screening when the demagnetizing factor is taken into account. A small Meissner effect is evidenced in the FC magnetization curve. However, flux pinning may lead to an inhomogeneous flux distribution within the crystal and, consequently, the use of a unique  $D$  factor may not be appropriate to describe the actual demagnetizing effects which take place inside the sample. If the rough estimation  $D = 0.946$  is used, as deduced from the ZFC data, then the flux expulsion amounts about 7.4%. Although small, such an effect is a clear signature of bulk superconductivity.

In the inset of Fig. 2 we show a typical  $M(H_a)$  curve, obtained at  $T = 7$  K.  $H_a$  is the applied field and  $M$  is the value of the superconducting contribution to the magnetization, as described above. The low-field part of this curve, before the full flux penetration ( $H_a^* \approx 2000$  Oe) is used to determine  $H_{c1}$ . It is known<sup>16,17</sup> that determination of the first flux penetration can be better done if the departure from the linearity  $\Delta M = M - \chi_0(T)H_i$  is plotted versus the internal field  $H_i = H_a - 4\pi D\mathbf{M}$ . The initial slope  $\chi_0(T) = dM(T)/dH_i$  has been determined from the

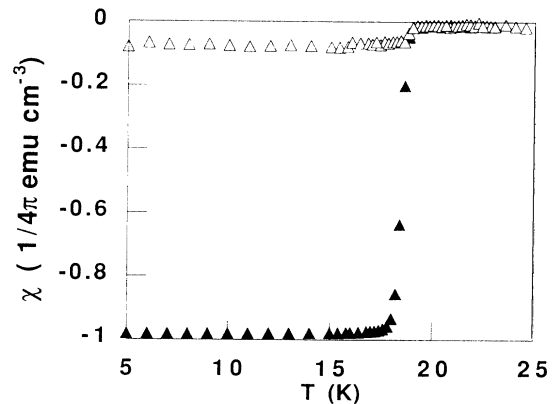


FIG. 1. ZFC (closed symbols) and FC (open symbols) susceptibility  $\chi = M/H_i$  obtained with an applied field  $\mathbf{H}_a = 1.6$  Oe perpendicular to the Cu-O planes. The internal field  $H_i = H_a - 4\pi D\mathbf{M}$  has been calculated using  $D = 0.946$ .

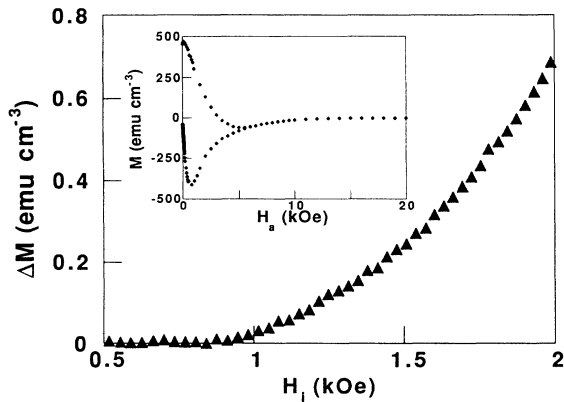


FIG. 2. Deviation of the linearity  $\Delta M = M - \chi_o H_i$  (see text) of the first magnetization curve at  $T = 7$  K, vs the internal field  $H_i$ . Inset: Magnetization vs applied field  $H_a$  at  $T = 7$  K, for increasing and decreasing magnetic field.

data in the Meissner state, at the lowest fields. In Fig. 2 we show  $\Delta M$  versus  $H_i$  at 7 K. A sudden flux penetration at  $H_i \sim 900$  Oe signals  $H_{c1}$  at this particular temperature. According to Bean's critical-state model,<sup>18</sup>  $\Delta M \sim (H_i - H_{c1})^2$  for  $H_{c1} < H_i < H^*$ . Therefore, a plot of  $(\Delta M)^{1/2}$  versus  $H_i$  should be a straight line, allowing a more accurate determination of  $H_{c1}$ . As shown in Fig. 3, the Bean model predictions are reasonably well verified. This observation is of capital importance because it allows us to extrapolate  $H_{c1}(T)$  from the bulk flux penetration and gives confidence that the results are not compromised by edge effects or surface barriers. These effects are at the origin of the slight deviation from the theoretical curve just below  $H_{c1}$ .<sup>19</sup>

Figure 4 summarizes the temperature dependence of the lower critical field as obtained via this procedure. The observation of a null remanent magnetization for

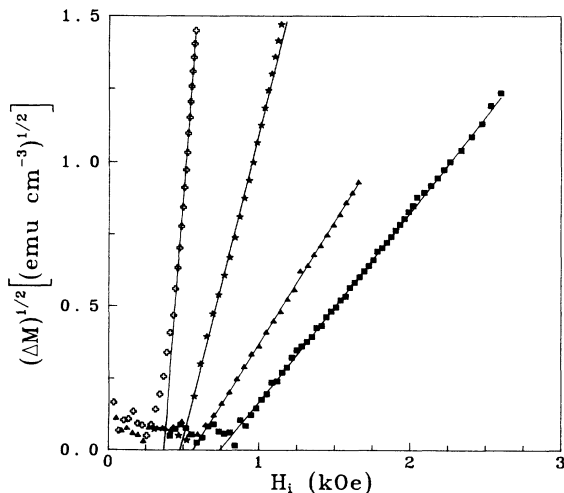


FIG. 3. Square root of the deviation of linearity  $\Delta M$  against the internal field  $H_i$ , for  $T = 7$  K (squares), 8 K (triangles), 10 K (stars), and 11 K (crosses). A linear dependence is observed at every temperature, according to Bean's model.

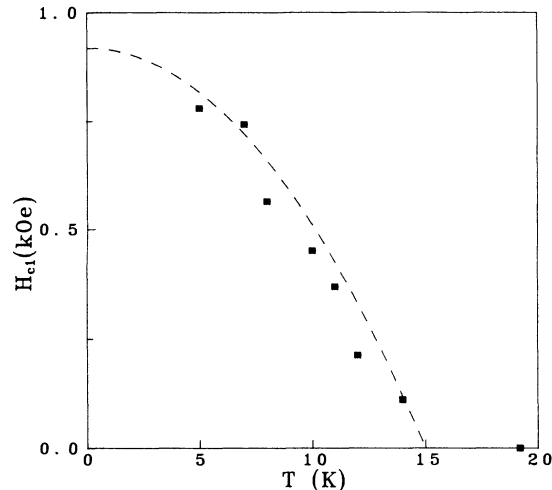


FIG. 4. Lower critical fields obtained from the extrapolation to  $\Delta M = 0$  of the data in Fig. 3. The dashed line is the fit of the data to Eq. (1) in the text.

magnetic fields smaller than  $H_{c1}(T)$  provides a further measure of the lower critical field.<sup>20</sup> There is no sign of any anomalous enhancement of  $H_{c1}$  at low temperatures as previously observed in some  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$  and  $\text{YBaCuO}$  crystals.<sup>3,17</sup> The temperature dependence of the lower critical field follows within a few percent the empirical law:<sup>21</sup>

$$H_{c1}(T) = H_{c1}(0) [1 - (T/T_c)^2]. \quad (1)$$

From the fit we extrapolate the lower critical field at zero temperature  $H_{c1}(0) \approx 920$  Oe by using a critical temperature of about 15 K. Linear extrapolation of the low-temperature values of  $H_{c1}(T)$  toward zero shows that the lower critical field is vanishingly small at  $T \approx 16$  K, below the  $T_c$  value deduced from the low-field ZFC measurement. As mentioned above, our  $H_{c1}(T)$  values reflect a bulk property of the material and thus do not preclude that some slight variation of the superconducting parameters may exist within the crystal. It is to be noted that the ZFC curve of Fig. 1 shows a very sharp transition at  $T_c = 19.2$  K and complete shielding well above the temperature ( $\sim 15$  K) where  $H_{c1}$  extrapolates to a zero value. It is well known<sup>22</sup> that the reduction process necessary to induce superconductivity in electron-doped cuprates typically produces materials having compositional gradients. Therefore, the existence of a surface sheath, even of negligible volume, may easily lead to a complete field screening at a temperature slightly higher than the overall, bulk critical temperature.

We turn now to the high-field region of the  $M(H)$  curves. In Fig. 5 we show several magnetization curves after correction of the differential susceptibility  $\chi_i$ . Both the increasing and the decreasing parts of the  $M(H_i)$  curves are shown, and a narrow reversible region can be clearly appreciated before the magnetization is reduced to zero at the normal state. We have taken the onset of diamagnetism in  $M(H)$  as a first estimation of the upper critical field  $H_{c2}(T)$ . In Fig. 6 we show the resulting

values.

A deeper insight into the fundamental superconducting parameters can be obtained from the analysis of the  $M(H)$  curves. In the Abrikosov regime at high fields and temperatures, the reversible magnetization near  $H_{c2}(T)$  is given by

$$4\pi\mathbf{M} = -\frac{1}{(2\kappa^2 - 1)\beta_A} (H_{c2} - H), \quad (2)$$

where  $H = H_i$  is the applied field and  $\beta_A = 1.16$  for a hexagonal flux lattice. Equation (2) predicts a linear  $M \sim H$  behavior close to  $H_{c2}(T)$  and allows to determine  $\kappa(T)$  and  $H_{c2}(T)$  from the slope and the intercept of the linear part of the  $M(H)$  curve. This procedure avoids the arbitrariness of the definition of the diamagnetic onset, which may be affected by fluctuations<sup>21</sup> and any inhomogeneous superconductivity, which is very sensitive to the subtraction of the nonsuperconducting (sample holder and rare-earth) background.

Data in Fig. 7(a) reveal the existence of a linear  $M \sim H$  region. According to Eq. (2) extrapolation of  $M(H)$  towards  $M = 0$  defines  $H_{c2}(T)$ . In Fig. 6 we have also plotted the  $H_{c2}(T)$  values obtained by this method. These values are very similar to the ones previously obtained; at low temperatures ( $t < 0.6$ ) they can be fitted to a power law  $H_{c2}(T) = H_{c2}(0)(1-t)^n$ , with  $n = 3.3$  and  $H_{c2}(0) \approx 70 \pm 1$  kOe. For higher temperatures ( $t > 0.6$ ) a change to an exponent  $n = 2.3$  is observed. A similar behavior has been reported by Dalichaouch *et al.*<sup>23</sup> and will be discussed below. For  $H_{c2}(0) = 70$  kOe the corresponding in-plane coherence length is  $\xi_{ab}(0) \approx 70$  Å which, together with the previously obtained  $H_{c1}(0) \approx 920$  Oe, gives a penetration depth  $\lambda_{ab}(0) \approx 700$  Å parallel to the Cu-O layers. Consequently, a Ginzburg-Landau parameter of the order of  $\kappa \approx 10$  is obtained.

An independent estimation of  $\kappa$  can be obtained from the slopes of  $M \sim H$  [Fig. 7(a)]. It is clear that the slopes

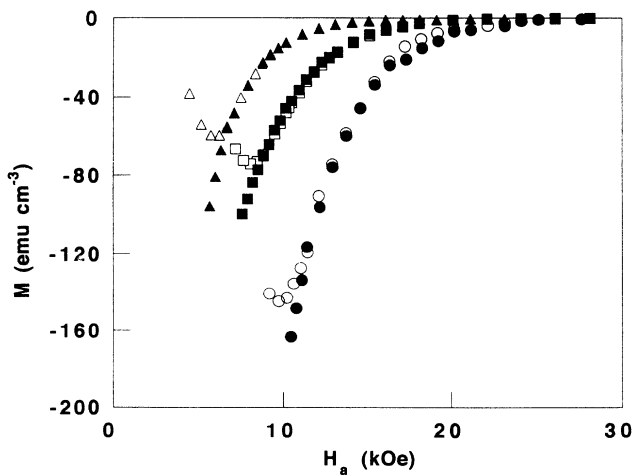


FIG. 5. Field dependence of the reversible magnetization at  $T = 5$  K (circles), 6 K (squares), and 7 K (triangles). Closed and open symbols are used for increasing and decreasing fields, respectively.

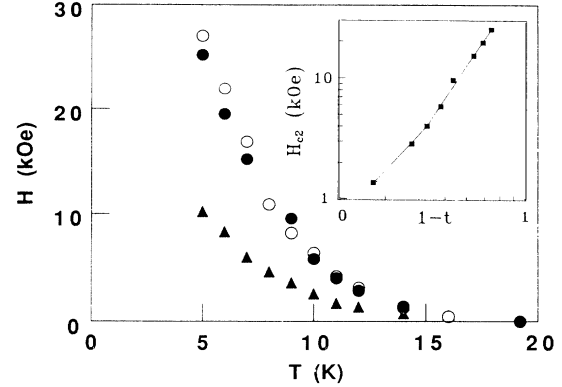


FIG. 6. Upper critical-field values obtained from the condition  $M = 0$  (open circles) and from the Abrikosov fits near  $M = 0$  (closed circles); both sets of data follow a power-law behavior, with  $n = 3.3$  for  $t < 0.6$  (see text). The irreversibility line from Ref. 25 is also shown for comparison (triangles). Inset:  $\ln H$  vs  $\ln[1 - (T/T_c)]$  for the  $H_{c2}(T)$  data obtained from the Abrikosov model.

are slightly temperature dependent. Within the scope of the Abrikosov model [Eq. (2)] the dependence of  $dM/dH$  with temperature comes through the variation of  $\kappa$ . In Fig. 7(a) (inset) we have included the obtained  $\kappa(T)$ , which decreases when increasing the temperature, giving values ranging from 2.6 at  $T = 16$  K to 5 at  $T = 5$  K. This relative variation of  $\kappa$  is similar to the one observed in classical superconductors<sup>24</sup> and expected from theory.<sup>21</sup> The absolute values are slightly smaller than the ones determined from  $H_{c1}(0)$  and  $H_{c2}(0)$ . This discrepancy may be due to the rather arbitrary extrapolation to  $T = 0$  K of the obtained  $H_{c1}(T)$  and  $H_{c2}(T)$  data. If one wishes to compare the values estimated from both methods it could be better to evaluate  $\kappa$  from the determined  $H_{c1}$  and  $H_{c2}$  values at a finite temperature; this procedure gives, for instance,  $\kappa(5 \text{ K}) = 6.3$ , much closer to the value obtained from the Abrikosov fit.

According to the London model, in the reversible intermediate-field region ( $H_{c1} \ll H \ll H_{c2}$ ), the magnetization is approximated by

$$-4\pi\mathbf{M} = \frac{\phi_0}{8\pi\lambda^2} \ln \left[ \eta \frac{H_{c2}(T)}{H} \right], \quad (3)$$

where  $\eta$  is a constant of order unit.<sup>25</sup> Therefore, an approximately linear relation  $M \sim \ln H$  should be observed at moderate field intensities, and its slope provides an independent way to estimate  $\lambda(T)$ . In Fig. 7(b) we show the  $M(H_i)$  curves of Fig. 5 plotted as  $M$  versus  $\ln H_i$ . At the lowest temperatures ( $T \leq 9$  K) the data display a linear behavior as predicted by Eq. (3), in a narrow field range. At higher temperatures, this linear part is washed out. From the slopes  $dM/d \ln H$  in Fig. 7(b) we have obtained penetration depth values ranging from 170 to 400 Å, which correspond to  $T = 5$  and  $T = 9$  K, respectively.

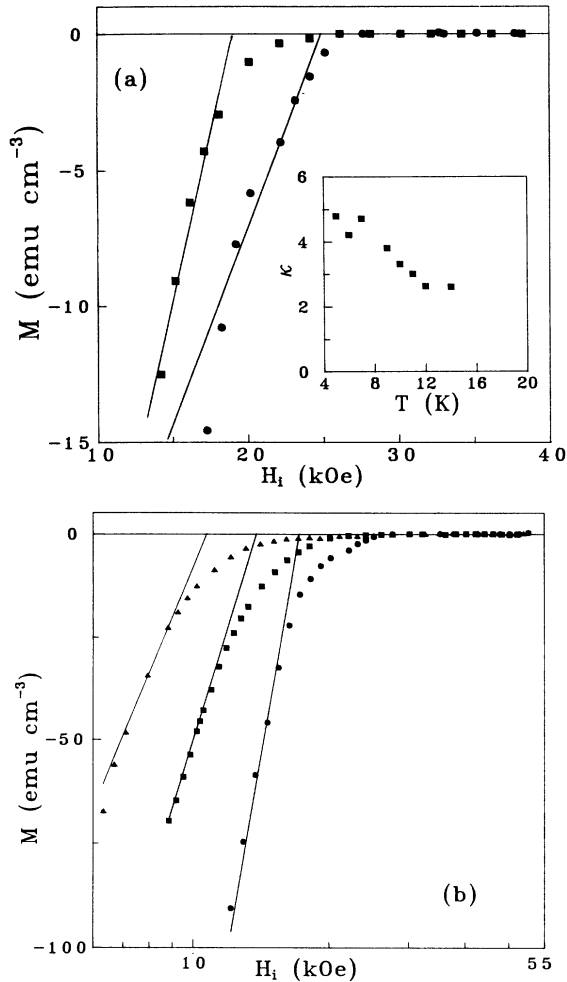


FIG. 7. (a) Magnetization vs internal field near  $H_{c2}(T)$  for  $T=5$  K (circles) and 6 K (squares). The solid lines are fits to the Abrikosov expression. Inset: Dependence of the Ginzburg-Landau parameter  $\kappa$  on temperature. (b) London plots  $M$  vs  $\ln(H_i)$  at  $T=5$  K (circles), 6 K (squares), and 7 K (triangles). The solid lines show the fits of the data to Eq. (2) in the text, for  $H_{\text{irr}}(T) < H_i \ll H_{c2}(T)$ .

## DISCUSSION

The most striking experimental result so far reported is the low value of the Ginzburg-Landau parameter  $\kappa$ . Indeed, all the estimations of  $\kappa$  presented above lead to values smaller than 10, thus placing this  $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  single crystal rather far from most  $p$ -type cuprates, with estimated  $\kappa > 55$ .<sup>12</sup>

This result by itself is important enough and should be contrasted with the available data and the experimental procedures carefully checked. We will discuss first the upper critical-field results and the corresponding coherence lengths. Next, we will focus our attention on the lower critical field and penetration depth values.

From the magnetoresistance data Hidaka and Suzuki<sup>2</sup> reported,  $H_{c2}(0)=67$  kOe for  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  single crystals, whereas for a  $\text{Sm}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  single crystal Dalichaouch *et al.* obtained  $H_{c2}(0)=53$  kOe.<sup>23</sup> As it is

not clear if the magnetoresistance measurements provide a determination of  $H_{c2}(T)$  or the irreversibility line in these low-temperature cuprates, it may be worth comparing our data with the magnetically determined  $H_{c2}(T)$ . Almasan *et al.*<sup>4</sup> have recently reported  $M(H)$  measurements on magnetically aligned particles of  $\text{Sm}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$ ; the data were analyzed in the framework of the model proposed by Hao *et al.*,<sup>26</sup> and  $H_c(T)$  and  $\kappa=77$  were extracted. From these data a value of  $H_{c2}(0)=65$  kOe was computed. Therefore, our  $H_{c2}(0)$  data are similar to the available data for similar materials, and consequently  $\xi_{ab}=70$  Å is also close to the data so far reported.

Dalichaouch *et al.*<sup>23</sup> reported a change in the power-law  $H_{c2}(T)=H_0(1-t)^m$  data of a  $\text{Sm}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  single crystal for  $\mathbf{H}\parallel c$ , at  $t=T/T_c=0.5$ . The enhancement of the upper critical field at low temperatures was interpreted in terms of magnetic ordering of the  $\text{Sm}^{3+}$  ions. However, latter measurements on other grain-aligned  $\text{Sm}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  samples having different  $T_c$  but similar rare-earth Néel temperatures revealed that the change of the exponent in the  $H_{c2}(T)$  law is not related to the magnetic ordering.<sup>9</sup> Our results also show indication of a change of the exponent, at  $t \approx 0.6$  (see inset in Fig. 6). It is important to recall that the magnetically determined irreversibility line (defined by the merging point of the increasing and decreasing parts of the hysteresis loop) measured on the very same crystal<sup>27</sup> also shows a power-law behavior but it lies well below  $H_{c2}(T)$ . We thus conclude that the upward curvature of  $H_{c2}(T)$  is not reminiscent of the irreversibility line, i.e., is not related in any way to flux-pinning effects and it reflects an equilibrium property of the measured crystal. Similar behavior had been observed in the early reports on  $H_{c2}(T)$  in YBCO and it was suggested that it might result from superconducting fluctuations.<sup>28</sup> Later, it became clear that better quality single crystals displayed a linear variation of  $H_{c2}(T)$  at  $T \approx T_c$ .<sup>29</sup>

The linear region  $M$  vs  $H$  predicted by the Abrikosov model for  $H=H_{c2}$  has been identified [see Fig. 7(a)]. Before going further it is important to assess if the used  $M(H)$  points are close enough to  $H_{c2}(T)$  for the Abrikosov expression to hold. As shown by Hao *et al.*,<sup>26</sup> the linear behavior  $M \sim H$  predicted by the Abrikosov model can be observed for  $h=H/H_{c2}(T) \geq 0.3$ . Our experimental data are well within the expected field range.

Very close to  $M=0$  some rounding of the  $M(H)$  curves can be appreciated. A related experimental observation in BSCCO and YBCO has been interpreted in terms of field induced superconducting fluctuations.<sup>30</sup> We have evaluated the field range  $\Delta H$  in the  $H$ - $T$  diagram where diamagnetic fluctuations could be observed. An estimation of the fluctuation diamagnetism above  $T_c$  is given by<sup>21</sup>

$$\chi_D = -\frac{\pi kT}{6\pi\phi_0^2} \xi(T) \approx -10^{-7} [T_c(H)/\Delta T]^{1/2},$$

where  $\Delta T=T-T_c(H)$ . If we take a magnetic-moment

resolution of  $10^{-6}$  emu it turns out that  $\Delta T < 0.1$  K. The associated field intensity interval is about  $\Delta H = 0.6$  kOe and thus the corresponding magnetization is clearly hidden by our measurement noise. This evidence, together with the fact that for higher temperatures the rounding becomes more apparent, seems to rule out the interpretation of the mentioned rounding as a fluctuation effect. Instead of that, we can attribute it to experimental uncertainties caused by the background subtraction, which is obviously more important for  $M \sim 0$ . However, the existence of some compositional inhomogeneity within the crystal having slightly higher  $T_c$  cannot be excluded and it may easily give rise to the observed rounding.

Let us discuss now the London model fit shown in Fig. 7(b). Hao and Clem<sup>26</sup> have pointed out that the London model in the intermediate-field region is quantitatively incorrect because it ignores the contribution of the vortex cores to the free-energy density. It turns out that  $M(\ln H)$  is no longer linear in the  $H_{c1} \ll H \ll H_{c2}$  field range; however, if one still wishes to use the  $M \sim \ln H$  expression, then field-dependent fitting factors should be included in the logarithm and in the prefactor. In any case, it must be  $h = H/H_{c2} < 0.3$  for the approximate  $M \sim \ln H$  behavior to be observed.<sup>26</sup>

Our data of Fig. 7(b), showing an approximate  $M \sim \ln H$  law, are close to the upper limit for  $h$ . Indeed, for some temperatures  $h$  lies above the field range where the logarithm term should be observed, i.e., for  $T = 6$  K it is  $h = 0.5$ . Therefore, we conclude that the field range in the London  $M \sim \ln H$  plots is too narrow and in some cases extends to excessively high fields. Consequently, the penetration depth values extracted from these fits may not be significant.

We turn now to the determination of the lower critical field  $H_{c1}(T)$ . To our knowledge direct estimations of  $H_{c1}(T)$  from  $M(H)$  curves in  $L_{2-x}\text{Ce}_x\text{CuO}_{4-y}$  single crystals have only been attempted by Balakrishnan *et al.*<sup>3</sup> for  $L = \text{Nd}$ . From the analysis of the isothermal magnetization data they concluded that an upper limit for  $H_{c1}(0)$  is  $1800 \pm 100$  Oe, well above our  $H_{c1}(0) \approx 920$  Oe. However, these authors extracted  $H_{c1}$  from the deviation of  $\Delta M(H)$  from zero and obviously the observed  $H_{c1}(T)$  values depend strongly on the experimental resolution. Almasan *et al.*<sup>4</sup> have measured the magnetization for a grain-aligned polycrystalline  $\text{Sm}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  sample; the  $M(H)$  data in the reversible region were analyzed by using the Hao and Clem model.<sup>26</sup> From this analysis they extracted the Ginzburg-Landau parameter and the thermodynamical critical field  $H_c(T)$ , which were used to evaluate  $H_{c1}(0) = 26.5$  Oe and  $H_{c2}(0) = 65$  kOe. This  $H_{c1}(0)$  value is much smaller ( $\sim \frac{1}{30}$ ) than our estimation. It is not clear whether this low  $H_{c1}$  reflects different electronic properties or it stems from the distinct procedure used to analyze their data. It should be mentioned that the polycrystalline particles that these authors have used to perform such analysis might have a dense weak-link structure which can provide inadequate first flux penetration values for bulk materials.

The close similarity of the  $\kappa$  values we have deduced from the slopes of the linear part of  $M(H)$ , which are not

subject to serious demagnetizing field-dependent correction, and the  $\kappa$  values deduced from the measured upper and lower critical fields strongly supports our experimentally determined  $H_{c1}(0) \approx 920$  Oe and signals that this large value is not a consequence of sample shape or surface barrier effects; in fact, the absence of significant surface-barrier effects could also be inferred from the fact that the decreasing irreversible part of the  $M(H)$  curve is not close to  $M = 0$ , as it is usually found.<sup>31</sup>

A comment on this low  $\kappa$  value and short penetration depth is in order. It is important to recognize that other attempts to determine  $\lambda_{ab}$  by muon-spin relaxation, for instance, have been unsuccessful because the rare-earth paramagnetism produces a spin relaxation which is much greater than the expected from the field inhomogeneity caused by the formation of a flux lattice.<sup>5</sup> We would like to recall that it has been reported in YBCO, for  $\mathbf{H} \parallel c$ ,  $H_{c1}(0) = 780$  Oe from high-field and high-temperature flux penetration experiments,<sup>19</sup> and it has been argued<sup>26</sup> that low-temperature experiments which provide higher  $H_{c1}(0)$  values may be significantly affected by Bean-Livingstone type surface barriers; these are expected to play a minor role in this  $n$ -type material because of its smaller  $\kappa$  value.<sup>32</sup>

Based on these  $H_{c1}(0)$  and  $H_{c2}(0)$  measurements, one can calculate the numerical value of the density of states  $N(0)$ , or equivalently, the corresponding Sommerfeld parameter  $\gamma$  (the coefficient of the linear-in- $T$  electronic specific heat). The results is  $\gamma \approx 60$  mJ/mol K<sup>2</sup>. This value is similar to that reported by Ghamaty *et al.*<sup>33</sup> for  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  from specific heat measurements ( $\gamma = 53$  mJ/mol K<sup>2</sup>), but larger than the one reported by Sanders, Hyun, and Finnemore<sup>34</sup> from the analysis of the  $M(H)$  curves ( $\gamma = 2-3$  mJ/mol K<sup>2</sup>). In our estimation of  $\gamma$  we have used the weak-coupling BCS relations. Indeed, Huang *et al.*<sup>35</sup> have recently found from tunneling data that  $2\Delta(0)/kT_c = 3.9 \pm 0.4$  in  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ , which would signal a moderate coupling strength. On the other hand, our estimation of  $\gamma$  points to a rather high density of states and places the electron-doped superconductors ( $L_{2-x}\text{Ce}_x\text{CuO}_{4-y}$ ) close to the  $A15$  alloys [for instance,  $\text{V}_3\text{Ga}$  has  $T_c \approx 15$  K and  $\gamma \approx 97$  mJ/mol K<sup>2</sup> (Ref. 26)] and the new fullerenes in the  $T_c \sim \gamma$  plot.<sup>12</sup> In contrast, it may be significant that other  $p$ -type HTSC [ $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO), YBCO, BSCCO, . . .] are placed well above a separating line in the  $T_c$ - $\gamma$  plot, thus suggesting that an asymmetry other than the sign of the charge carriers<sup>1</sup> and pressure effects on  $T_c$  (Ref. 37) exist in the electron-doped superconductors.

In summary, magnetization measurements on a  $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  single crystal have been performed for  $\mathbf{H} \parallel c$ . Low-field ZFC and FC data reveal a perfect diamagnetism and bulk superconductivity with a remarkable flux pinning. The demagnetizing field has been evaluated by using several distinct approaches and the experimental data  $M(H)$  have been accordingly corrected. The flux penetration into the material at  $H > H_{c1}(T)$  has been observed to closely follow the predictions of the Bean critical-state model, and  $H_{c1}(T)$  has been extracted. In the high-field reversible region of the  $M(H)$  curve the ex-

perimental data are well described by the Abrikosov model, thus providing a measure of  $H_{c2}(T)$  and  $\kappa$ . It turns out that the estimates of the lower and upper critical fields, as well as the coherence length and the penetration depth, are  $H_{c1}(0) \approx 920$  Oe,  $H_{c2}(0) \approx 70$  kOe,  $\xi_{ab}(0) \approx 70$  Å, and  $\lambda_{ab} \approx 700$  Å. Consequently, this electron-doped superconductor has a rather low  $\kappa \leq 10$  value.

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