Reaching the Pauli limit in the cuprate $Bi_2Sr_2CuO_{6+\delta}$ in high parallel magnetic fields

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We present a careful study of the resistive superconducting transition in $Bi_{2+x}Sr_{2-x}CuO_{6+\delta}$ down to $T/T_c = 0.04$ for magnetic field applied parallel to the conducting planes. We find that 52 *T* is enough to destroy superconductivity at low temperature. Based on a Ginzburg-Landau calculation, the paramagnetic limitation of superconductivity for the field parallel to the layers is considered as an explanation of the observed behavior.

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I. INTRODUCTION

Since the discovery of high T_c superconductors (HTSC), the determination of their upper critical field H_{c2} is controversial. Nevertheless this parameter is of crucial importance since it reflects the coherence length of the Cooper pair. Owing to the large value of H_{c2} in the cuprates, many experiments have been devoted to resistive measurements at high magnetic field perpendicular to the conducting planes¹⁻⁷ and have shown an anomalous positive curvature of H_{c2} at low temperature. This result is in contradiction with the expected low temperature saturation described by the conventional Werthamer, Helfand, and Hohenberg (WHH) theory.⁸ More recently, a careful study of H_{c2} in electron-doped thin films of $Pr_{2-x}Ce_xCuO_4$ by using resistive and susceptibility measurements⁹ has shown a direct correlation between the low temperature behavior of H_{c2} measured by resistivity and the irreversibility line. Another support of the inadequacy of the resistivity as a probe of H_{c2} is given by Nernst measurements¹⁰ in Bi₂Sr₂CaCu₂O₈ and Bi₂Sr₂CuO₆, which suggest that H_{c2} is much higher that the upper critical field deduced from resistivity measurements. A possible explanation is the small coherence length giving rise to strong fluctuation effects¹¹ and a phase diagram occupied by a large region of vortex liquid.¹² Another approach considers that HTSC possess intrinsic inhomogeneities and that the superconducting transition corresponds to a percolation among different superconducting regions.¹³

In contrast, few measurements have been performed at high magnetic field parallel to the conducting planes. In this configuration, the magnetic field required exceeds the maximum field available in magnetic field facilities. O'Brien *et al.*¹⁴ and Sekitani *et al.*¹⁵ have used single-turn coil and explosive flux compression with a contactless resistivity measurement technique in the radio-frequency range in order to construct the *H*-*T* phase diagram in the **H**||**ab** configuration in YBa₂Cu₃0_{7+ δ} (YBCO). In Ref. 14, a paramagnetic limitation of superconductivity was deduced, while in Ref. 15 a phase diagram consistent with the WHH theory was found, taking into account the Zeeman and spin-orbit effects. Such measurements are extremely difficult since the rise time of the field is typically a few microseconds, generating a *dB/dt* of more than 10⁸ T/s. Besides a poor signal-to-noise ratio, the eddy currents generated by the field pulse may also strongly increase the temperature of the sample during the pulse. It seems to be rather difficult to draw a definitive conclusion from these measurements.

Nevertheless, HTSC are suitable systems in order to test theories of parallel upper critical field in layered superconductors. Historically, the first attempt to calculate $H_{c2||ab}$ of layered superconductors was carried out by Kats¹⁶ and by Lawrence and Doniach.¹⁷ Afterwards, Klemm et al.¹⁸ have shown that the upper critical field diverges at the temperature T^* when the coherence length perpendicular to the *ab*-plane, $\xi_c(T)$, approaches the value $s/\sqrt{2}$ where s is the distance between the conducting layers. In this limit, the vortex cores fit between the superconducting layers and supercurrents do not quench the superconductivity. However, this conclusion is only valid when the thickness of the superconducting layers is neglected.¹⁹ When one takes into account the flow of supercurrents into the layers (which flow in opposite direction along the upper and the lower surfaces of the layers), the upper critical field is determined by the Cooper pair critical velocity and is given by $H_{c2\parallel ab} = \sqrt{3}\Phi_0 / \pi d\xi_{ab}$.^{20,21} The temperature driven transition from a three-dimensional (3D) to a two-dimensional (2D) situation has been shown experimentally for Nb-Ge multilayers by Ruggiero et al.22 They found a *H*-*T* phase diagram where $H_{c2\parallel ab}$ starts as $(T_c - T)$ close to T_c (the anisotropic 3D behavior predicted by Refs. 16 and 17) and crosses over to the $(T_c - T)^{1/2}$ behavior when $\xi_c(T)$ $\approx s/\sqrt{2}$. The extension of the theory of $H_{c2\parallel ab}$ to low temperatures has been derived by Lebed and Yamaji.²³ It was found that below the temperature $T_L \approx v_F s H/\Phi_0$ there is another divergence of the upper critical field which has a quantum origin. This type of divergence has never been observed experimentally, most likely because it can occur only in ultraclean materials.²⁴ Recently, an analytic expression of the angular and temperature dependencies of the upper critical field in layered superconductors has been derived.²⁵ However, this derivation did not take into account the paramagnetic limitation of the upper critical field $H_{c2\parallel ab}$ which can be important in layered materials, like the HTSC.

In this paper, we present a careful study of the *H*-*T* phase diagram for magnetic fields applied parallel and perpendicular to the conducting planes of $\text{Bi}_{2+x}\text{Sr}_{2-x}\text{CuO}_{6+\delta}$ (Bi-2201).

We have measured the magnetoresistance using the standard four probe geometry in pulsed magnetic fields up to 55 T over a wide range of temperature, down to 0.38 K in a home-made ³He refrigerator designed for pulsed magnetic field.²⁶ Owing to the low critical temperature of Bi-2201, 52 T is sufficient to suppress superconductivity at $T/T_c=0.04$ in the **H**||**ab** geometry. Based on a Ginzburg-Landau approach appropriate for Bi-2201, we estimate the paramagnetic Pauli limit to be in agreement with the experimental value of $H^*_{c2||ab}$.

II. EXPERIMENT

Two slightly underdoped single-phase $Bi_{2+x}Sr_{2-x}CuO_{6+\delta}$ crystals with a carrier concentration per Cu atom of p =0.15-0.16, are investigated in this study. They were grown without intentional doping by a KCl-solution-melt free growth method detailed elsewhere.²⁷ Typical dimensions of the crystals are $(2-10) \times (400-800) \times (600-900) \ \mu m^3$. The zero-field critical temperatures defined by 10% and 90% of the resistive transition equal 7.0-8.8, and 7.0-8.4 K for samples No. 3 and No. 5, respectively. We estimate p in our samples by using the empirical (nearly linear) relation between the excess Bi, x, and p.^{28,29} Optimum doping takes place around $p \simeq 0.17^{29}$ In the four-probe resistivity measurements the current is injected along the *ab*-plane of the crystals and transverse to the field. The resistivity of each sample was measured at a given fixed temperature during the magnetic field pulse³⁰ using a lockin amplifier working at 50 kHz. The onset of the transition was independent of current density in our range of measurements and no frequency dependence was observed in the data. Results obtained during the rising (26 ms) and the falling (110 ms) edge of the pulsed magnetic field, at the same B but at different dB/dt, agree, which excludes any heating effects due to eddy currents. In order to check the stability of the temperature of the sample during the pulse, the signal measured during the rising and the falling of the pulse must coincide. This is shown in Fig. 1 for the data obtained in the ³He refrigerator at T=0.38 K. Moreover, if the sample is not a superconductor, the resistance of the sample recorded via a pretrigger before the shot must be the same after the shot (see the curve of Fig. 1 obtained at T=8.2 K).

III. RESULTS

Figure 2 shows the in-plane magnetoresistivity (MR) ρ_{ab} versus *H* for sample No. 3 at different temperatures for field direction parallel to the *ab*-plane of the crystal. For comparison, we represent also the resistive superconducting transition of the same sample at T=0.46 K with the field direction perpendicular to the *ab*-plane of the crystal. The field induced resistive transitions for sample No. 5 are similar to those shown in Fig. 2. At low temperature, the traces of $\rho_{ab}(H)$ are roughly parallel with respect to each other in the transition region. When the temperature is close but below T_c , the superconducting transition broadens. The rapid increase of the magnetoresistance at very low field observed at T=7 K comes from the fact that this point lies in the sharp



FIG. 1. Magnetoresistance of the Bi-2201 crystal No. 3 at two temperatures measured during the rising (dash lines) and the falling (solid lines) of the magnetic pulse. It can be noticed that the curves are superimposed.

superconducting transition of the sample. Beyond this fluxflow regime, the resistivity reaches an asymptotic value close to its normal-state value, which matches exactly for the two orientations of the magnetic field.

Before addressing the $H_{c2}^*(T)$ data, it is instructive to present the temperature dependence of the in-plane resistivity of the samples at low temperatures for the magnetic field



FIG. 2. Selected traces of ρ_{ab} as a function of magnetic field applied parallel to the *ab*-plane for different temperatures (solid lines). For comparison, we represent also the resistive superconducting transition of the same sample at T=0.46 K with the field direction perpendicular to the *ab*-plane of the crystal (symbols).



FIG. 3. Semilog plot of ρ_{ab} versus temperature for various magnetic fields applied in the *ab*-plane. The inset shows our data for the Bi-2201 single crystal with p=0.16 at various fixed magnetic fields applied perpendicular to the *ab*-plane (Ref. 29).

parallel to the *ab*-plane. Figure 3 (main panel) shows a semilogarithmic plot of $\rho_{ab}(T)$ at different magnetic fields applied parallel to the *ab*-plane for sample No. 3 extracted from the curves in Fig. 2. It is worth to notice that the 50 T and 55 T data are almost identical. This is a further proof that we are measuring the truly normal-state resistivity at high magnetic fields in the parallel configuration. For comparison, the inset of Fig. 3 represents data for Bi-2201 single crystal with p =0.16 at various fixed magnetic fields applied perpendicular to the *ab*-plane.²⁹ Since these samples have nearly the same carrier concentration and the same T_c , we conclude that the behavior of $\rho_{ab}(T)$ for the slightly underdoped samples in the normal state is nearly independent of the field orientation (perpendicular or parallel to the CuO_2 planes). That is to say, the slight upturn of the resistivity at low temperature which becomes much more pronounced for heavily underdoped samples^{29,31} (the so-called metal-to-insulator transition) is not affected by the orientation of the magnetic field and the Zeeman effect does not seem to play any role in this anomaly.

Let us now turn our attention to the upper critical fields. The temperature dependencies of the resistive upper critical fields $H_{c2\parallel ab}^*$ ($H_{c2\perp ab}^*$) in the **H** || **ab** (**H** \perp **ab**) geometry are shown in the main panel of Fig. 4 for the two investigated samples together with the theoretical WHH curves.⁸ The resistive H_{c2}^* deduced from the curves of Fig. 2 corresponds to the field where the normal state resistivity value is completely recovered. The latter has been obtained by subtracting the magnetoresistance in the normal state, as demonstrated in Ref. 5. For the **H** \perp **ab** configuration, except at very low temperature,⁵ the experimental data can be described by the conventional WHH theory, which is based on an orbital mechanism of quenching of superconductivity. The slight upturn of $H_{c2\perp ab}^*(T)$ at low temperature moves to higher tem-



FIG. 4. Main panel, the resistive upper critical fields H_{c2}^* deduced from 100% of the normal state resistivity as a function of the reduced temperature T/T_c for two investigated samples (No. 3, squares and circles; No.5, diamonds) at two field orientations **H**||**ab** and **H** \perp **ab**. T_c =10.2 K (9.0 K) for sample No. 3 (No. 5). Dashed lines correspond to the theoretical WHH curves (Ref. 8). Inset, the resistive upper critical fields H_{c2}^* deduced from 90% of the normal state resistivity as a function of the reduced temperature T/T_c , where $T_c(90\%)$ =8.8 K (8.4 K) for sample No. 3 (No. 5) (see text).

perature when one use a criterion distinct from the complete normal state resistivity value, as shown in the inset of Fig. 4 for which $H_{c2}^{*}(T)$ has been deduced from 90% of the normal state resistivity (open circles). However, in this configuration, we know that fluctuation effects and the presence of a vortex liquid in a sizeable part of the phase diagram may lead to discrepancies.^{5,9}

In contrast, for the $H \parallel ab$ configuration, the data show saturation at low temperature. It is important to emphasize that this saturation is robust against any criterion chosen for the extrapolation of $H_{c2\parallel ab}^*$, in particular when H_{c2}^* is deduced from 90% of the normal state resistivity as depicted in the inset of Fig. 4 (open squares and diamonds). The curve corresponding to the WHH theory departs strongly from the experimental data points for the H ab configuration. The WHH formula predicts $H_{c2}^*(0) = 0.693(-dH_{c2}^*/dT)T_c'$ where dH_{c2}^*/dT is the tangent of $H_{c2}^*(T)$ when $T \rightarrow T_c$ and T_c' the intersection of this tangent with the temperature axis. From the linear part of the $H_{c2}^{*}(T)$ dependency near T_{c} , we obtained the slopes $dH_{c2}^*/dT_c = -13.3$ T/K and -3.3 T/K in the **H**||**ab** and $\mathbf{H} \perp \mathbf{ab}$ geometry, respectively. Setting these values into the WHH formula leads to $H_{c2\parallel ab}^{*}(0)=92$ T and $H_{c2\perp ab}^{*}(0)$ =24 T. Note that the $H_{c2}^{*}(T)$ line in **H** || **ab** geometry has a negative curvature close to T_c (Ref. 21) and therefore, the $H_{c2}^{*}(0)$ value deduced from WHH theory is certainly a lower bound. It should be recognized that the strong broadening of the superconducting transition when the temperature is close to T_c leads to different temperature dependence of H_{c2}^* versus *T*. The slope of H_{c2}^* near T_c will thus depend on the method of evaluation of H_{c2}^* .⁶ Although the doping level of the



FIG. 5. Coherence lengths vs reduced temperature T/T_c as derived from experimental measurements of critical fields and derived using the AGL expressions for ξ_{ab} (squares) and ξ_c (circles). The solid and dashed curves are the best fit of the AGL expression $\xi_{ab}(T)$ and $\xi_c(T)$ (see text).

samples in the present study is slightly lower than the one in Ref. 21, it has been shown that the temperature dependence of the reduced upper critical field as a function of the reduced temperature for $\mathbf{H} \perp \mathbf{ab}$ does not depend on the magnitude of T_c for Bi-2201 single crystal providing that T_c (midpoint) lies between 3.7 K and 9 K and in the temperature range $T/T_c = 0.04 - 1.5$ In previous studies of Bi-2201 single crystals in static magnetic fields up to 28 T, the critical field $H^*_{c2\parallel ab}(0)$ has been estimated to 43 T (T_c =6-7.7 K) when deduced from 50% of the normal state resistivity⁵ and to 65 T (T_c =8.1–9.8 K) when deduced from 80% of the normal state resistivity.²¹ A rough extrapolation gives $H^*_{c2\parallel ab}(0) = 86$ T and 81 T, respectively. Considering a drastic broadening of the MR curves in the upper part of the superconducting transition, these magnitudes should be somewhat higher and we can say that 92 T shown in Fig. 4 is consistent with preliminary data. The fact that our observations strongly deviate from the WHH theory for the case **H||ab** shows that the quenching of the superconductivity is not only due to orbital effect for this field orientation.

Another way to illustrate this departure is to translate the upper critical field in terms of a coherence length via the anisotropic Ginzburg-Landau (AGL) theory. The AGL relations are given by $H_{c2\perp ab}=\Phi_0/2\pi\xi_{ab}^2$ and $H_{c2\parallel ab}=\Phi_0/2\pi\xi_{ab}\xi_c$, where $\Phi_0=hc/2e$ is a flux quantum. Under the assumption that the relations are valid over a wide range of temperature, the temperature dependence of $\xi_{ab}(T)$ is obtained from experimental values of $H_{c2\perp ab}^*(T)$ using the first AGL expression and $\xi_c(T)$ is deduced from the second AGL expression together with the $H_{c2\parallel ab}^*(T)$ data. These results are shown in Fig. 5 for sample No. 3. The solid curve is the best fit of the AGL expression $\xi_{ab}(T) = \xi_{ab}(0)(1-T/T_c)^{-1/2}$ to the

data using $\xi_{ab}(0)=32$ Å and T_c as adjustable parameter. The dashed curve is a fit of the AGL expression $\xi_c(T) = \xi_c(0)(1 - T/T_c)^{-1/2}$ to the low temperature points using again $\xi_c(0)$ and T_c as adjustable parameters. The same analysis has been done for the other sample. Since the experimental $H_{c2}^*(T)$ dependencies are almost identical, the values of $\xi_{ab}(T)$ and $\xi_c(T)$ are also in close agreement. Although there is a good agreement with AGL theory for the temperature dependence of the in-plane coherence length ξ_{ab} , the data deduced from $H_{c2\parallel ab}^*(T)$ give a temperature-independent value of ξ_c ≈ 17 Å. Note that the values of $\xi_{ab}(0)$ and $\xi_c(0)$ are in good agreement with previous measurement.⁵ We can also estimate the anisotropy parameter $\gamma \equiv H_{c2\parallel ab}/H_{c2\perp ab} = 1.9$ in the low temperature range. This parameter is unusually low compared to other HTSC.

IV. COMPARISON WITH THEORIES

For a layered superconductor in a parallel magnetic field, a dimensional crossover from 3D to 2D with decoupled layers is predicted. At a temperature T^* corresponding to $\xi_c(T^*) \approx s/\sqrt{2}$, the upper critical field should diverge. However, no sign of divergence and or even of discontinuity in the *H*-*T* phase diagram for $\mathbf{H} \| \mathbf{ab}$ is observed in the experimental data. This is in contradiction with the model proposed by Klem *et al.*¹⁸ which predicts at T^* a 2D situation with decoupled layers for magnetic fields higher than H_{KLR} $=\Phi_0/s^2\gamma$. Setting in the corresponding values for Bi-2201 s=12.3 Å and $\gamma=1.9$, we find $H_{KLB}=720$ T, much higher than the experimental value of $H_{c2\parallel ab}^*$ for $T \rightarrow 0$. Moreover, if one tries to take into account the finite thickness d of the layers (which in principle should remove the divergence of H_{c2}), the parallel critical field can be written as $H_{c2\parallel ab}$ $=\sqrt{3}\Phi_0/\pi d\xi_{ab}$ ²⁰ Then the upper critical field $H_{c2||ab}$ at T =0.4 K is estimated to be approximately 1100 T for a thickness of the superconducting layers d=3 Å, which is much larger than the measured value.

To explain the moderate values of $H_{c2\|ab}^*$ and of the anisotropic parameter γ , we propose that the quenching of superconductivity for $\mathbf{H} \| \mathbf{ab}$ is due to paramagnetic limitation. Indeed, if the paramagnetic limit in Bi-2201 is of the order of 50 Tesla, then for $\mathbf{H} \perp \mathbf{ab}$ we do not reach it. In contrast, for $\mathbf{H} \| \mathbf{ab}$, by lowering the temperature, we reach the region where the critical field determined by the orbital effects $H_{c2\|ab} = \Phi_0 / 2\pi \xi_{ab} \xi_c$ starts to be larger than the paramagnetic limiting field. Then, at low temperatures, the H_{c2} stops to grow up and saturates at $H \approx H_p$. This effect finds its origin in the quasi-two-dimensionality of the system.

A crude estimate of the paramagnetic field is given by the Clogston-Chandrasekhar formula:^{32,33} $H_P = \Delta_0 / \mu_B \sqrt{2}$. Under the assumption that $2\Delta_0 = 3.5k_BT_c$, we can rewrite $H_P(0) = 1.84T_c$, which give $H_P(0) = 18$ T for Bi-2201, smaller than the experimental value. If we take the experimental values of the superconducting gap deduced from tunneling³⁴ or ARPES³⁵ experiments, then $H_P(0) = 48-82$ T. Owing to the uncertainty of the paramagnetic limit given by the simple Clogston–Chandrasekhar formula, it is necessary to do a proper calculation of $H_P(0)$ for a layered superconductor,

which has not been considered in the previous calculations of the angle and temperature dependencies of the upper critical field in layered superconductors.^{19,25} Since we are not interested in the very low temperature region of the phase diagram, where impurities and defects dominate the upper critical field behavior,²⁴ we have chosen a Ginzburg–Landau approach. In this approach, the upper critical field is an eigenvalue of the following equation, which has been directly derived from an integral equation in Ref. 23:

$$\frac{7\zeta(3)}{(2\pi T_c)^2} \left\{ -\frac{\hbar^2 v_F^2}{4} \frac{\partial^2 \Delta}{\partial x^2} + 2\left(\frac{etsHx}{\hbar c}\right)^2 \Delta + (\mu H)^2 \Delta \right\} = \tau \Delta.$$
(1)

Here $\tau = 1 - T/T_c$, *t* is the interlayer hopping integral and *s* is the distance between the centers of adjacent conducting layers, taken as the period of the structure. For simplicity we neglect the higher order harmonics in the dispersion law corresponding to the actual period of Bi-2201 which is equal to 2*s*. Finally, $\mu = g\mu_B/2$ is the magnetic moment of the electron.

By solving analytically equation (1), we find that $H_{c2\parallel ab}$ is a solution of the equation

$$\frac{H}{H_{\rm orb}} + \left(\frac{H}{H_p}\right)^2 = \tau.$$
 (2)

Here

$$H_{\rm orb} = \frac{(2\pi T_c)^2 c}{7\zeta(3)\sqrt{2}etsv_F}$$
(3)

and

$$H_p = \frac{2\pi T_c}{\sqrt{7\zeta(3)\mu}}.$$
(4)

When $\tau \rightarrow 0$ $(T \rightarrow T_c)$ we recover the Ginzburg-Landau formula

$$H_{c2}(T) = H_{\rm orb}\tau.$$
 (5)

The initial slope $-dH_{c2}^*/dT$ is very large due to the quasi-2D character of the material. If one assumes that

$$H_p < H_{\rm orb}.\tag{6}$$

Then at low enough temperatures, where τ has a finite value of about 1/2, we have from (2)

$$H_{c2}(T) \approx H_p \sqrt{\tau}.$$
 (7)

Hence, when $T \rightarrow 0$, the $H_{c2}(T)$ curve tends to saturate towards the Pauli limit.

Taking $T_c \approx 10$ K for Bi-2201, one can obtain the estimate

$$H_p(0) \approx \frac{64}{g}$$
 tesla. (8)

If one takes the free electron *g*-factor value g=2, we find $H_P(0) \approx 32$ T, smaller than the experimental value. However, if we take into account spin-orbital coupling, the value of the *g*-factor could be smaller than 2. Unfortunately, to our knowledge, there is no electron paramagnetic resonance result on HTSC which gives an estimate of the *g*-factor. Anyway, for **H**||**ab**, we find a paramagnetic limit H_P smaller than the orbital limit, given by the WHH extrapolation at low temperature (see Fig. 4) and we can conclude that superconductivity is quenched at moderate fields due to the Zeeman effect.

V. CONCLUSION

In summary, we have measured the resistive superconducting transition in Bi-2201 for $\mathbf{H} \| \mathbf{ab}$. By drawing the *H*-*T* phase diagram, we compare the experimental results with several theories which estimate the upper critical field in layered superconductors. We did not observe any temperature driven transition from a 3D to a 2D situation with decoupled layers. We conclude by using a Ginzburg-Landau approach, that superconductivity is quenched by paramagnetism at low temperature.

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