Low anisotropy of the upper critical field in a strongly anisotropic layered cuprate: Evidence for paramagnetically limited superconductivity

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We study angular-dependent magnetoresistance in a layered Bi$_{2}$Sr$_{1.9}$CuO$_{6+x}$ cuprate. The low $T_{c} \sim 4$ K allows complete suppression of superconductivity by modest magnetic fields and facilitates an accurate analysis of the fluctuation conductivity and a confident extraction of the upper critical field $H_{c2}(T)$. We observe that the anisotropy of $H_{c2}$ is surprisingly low, much smaller than the effective mass anisotropy of the material $\sim 300$. We show that the anisotropy is decreasing with increasing field and saturates at a small value when the field reaches the paramagnetic limit. We argue that the dramatic discrepancy of high field and low field anisotropies is a clear evidence for paramagnetically limited superconductivity.

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The upper critical field $H_{c2}$ is one of the key parameters of type-II superconductors [1]. The knowledge of $H_{c2}$ is particularly important for understanding unconventional superconductivity [2]. However, estimation of $H_{c2}$ for high-temperature superconductors is a notoriously difficult task. The high $T_{c}$ leads to an extended thermally activated flux-flow region at $T < T_{c}$ otherwise difficult to sufficiently accurately obtain $H_{c2}$ from flux-flow characteristics.

The high $T_{c}$ in combination with a strong coupling lead to a large superconducting energy gap $\Delta \sim 20 - 50$ meV [3, 4] and $H_{c2}(0) \sim 10^{2}$ T [5, 10]. Such strong fields alter the ground state of the material. Even relatively weak fields can induce a canting ferromagnetic order in strongly underdoped cuprates [11]. Furthermore, the normal state of underdoped cuprates is characterised by the presence of the pseudogap (PG), which probably represents a charge/spin or orbital density wave order coexisting and competing with superconductivity [3, 12, 13]. Suppression of superconductivity by magnetic field may enhance the competing PG, as follows from observation of the charge density wave order in vortex cores [4]. But even stronger magnetic fields of several hundred tesla suppress the PG [14, 15]. Thus, both superconducting and normal states respond significantly to magnetic fields and separation of the two contributions is highly non-trivial and controversial. Disentanglement of superconducting and PG magnetoresistances is difficult even above $T_{c}$ due to presence of profound superconducting fluctuations well above $T_{c}$ [16]. Therefore, principal new questions, which do not appear for low-$T_{c}$ superconductors, are to what extent $H \sim H_{c2}$ alters the abnormal normal state of high-$T_{c}$ superconductors and how to define the non-superconducting background in measured ($T, H$)-dependent characteristics.

For many unconventional superconductors the measured $H_{c2}$ exceeds the paramagnetic limit of the BCS theory [1]. This has been reported for organic [17, 18], cuprate [7, 8], pnictide [19, 20] and heavy fermion [21, 22] superconductors. Such an overshooting is an important hint in a long standing search for exotic spin-triplet and Fulde Ferrell Larkin Ovchinnikov states (for review see e.g. Ref. [23]). Yet, the overshooting is not a proof of unconventional pairing because the paramagnetic limit is rather flexible. It is increasing in the presence of spin-orbit interaction [1], significantly enhancing in the two-dimensional (2D) and is lifted in the one-dimensional (1D) case [22, 23]. Unconventional superconductors are usually anisotropic. Some of them have layered quasi-2D, or possibly even quasi-1D structure (cuprates and organic superconductors). Many have significant spin-orbit interaction between localized spins and itinerant charge carriers. Consequently, one needs a more robust criterion for the paramagnetically (un)limited superconductivity in search for exotic states of matter.

Here we investigate the anisotropy of $H_{c2}$ in an extremely anisotropic layered Bi$_{2}$Sr$_{1.9}$CuO$_{6+x}$ (Bi-2201) cuprate with a very low $T_{c} \sim 4$ K. The low $T_{c}$ and the associated large disparity of superconducting and pseudogap scales [15] in this compound allow simple and accurate estimation of $H_{c2}$ without complications typical for high-$T_{c}$ cuprates. Remarkably, we observed that $H_{c2}^{\perp}/H_{c2}^{\parallel}(T \rightarrow 0) \sim 2$ is much smaller than the anisotropy of effective mass $\gamma_{m} \approx 300$. This discrepancy clearly indicates that $H_{c2}^{\parallel}$ parallel to the CuO planes is cut-off by the paramagnetic limit.

Bi-based cuprates, including Bi-2201, represent natural stacks of atomic-scale intrinsic Josephson junctions [15, 24]. This reflects the 2D nature of superconductivity, confined within the CuO (ab) planes. The out-of-plane (c-axis) resistivity $\rho_{c} \sim 10$-100 $\Omega$cm is much larger than $\rho_{ab} \sim 10^{-4}$ $\Omega$cm [25, 26] resulting in the extremely large resistive anisotropy $\gamma_{R} = \rho_{c}/\rho_{ab} \sim 10^{5}$-$10^{6}$ and an effective mass anisotropy $\gamma_{m} = \sqrt{\gamma_{R}} \sim 100$-$1000$.

We present data for a nearly optimally doped crys-
with $T_c \simeq 4.3$ K [OP(4.3)] and a slightly overdoped crystal with $T_c \simeq 4.0$ K [OD(4.0)]. Studied samples had several contact pads (mesas) micro-fabricated on top of a freshly cleaved Bi-2201 single crystal. Details of sample fabrication, sample characterization, and the experimental setup can be found in Ref. [15]. The growth and characterization of crystals is described in Ref. [27].

Figures 1 (a) and (b) show $T$-dependencies of the in-plane resistance $R_{ab}$ at different fields perpendicular (a) and parallel (b) to $ab$ planes. There is a clear anisotropy with respect to field orientation. It is seen that for $H \perp ab$, $R_{ab}$ reaches the normal state value at $H_{c2} \simeq 10$ T. For $H \parallel ab$ the field of 17 T still does not completely suppress the superconductivity. Those fields are much smaller than the PG closing field $\sim 300$ T [15]. This allows accurate analysis of superconducting characteristics not affected by interferences with the co-existing PG.

Fig. 1 (c) shows angular dependencies of the in-plane resistance $R_{ab}(\Theta)$ at $T = 2$ K for rotation around two orthogonal axes in the ab-plane. In both cases $\Theta = 90^\circ$ corresponds to $H \perp ab$, $H \perp I$. But $\Theta = 0^\circ$ corresponds to the Lorentz force-free configuration $H \parallel ab \parallel I$ (dashed lines) or to the case $H \parallel ab$, $H \perp I$ (solid lines). In the latter case the Lorentz force is perpendicular to layers and is acting on Josephson-like vortices. It is seen that the behavior in both cases is very similar. Therefore, at $H \parallel ab$ the flux-flow contribution to $R_{ab}$ is small due to either zero Lorentz force or a strong intrinsic pinning in the layered superconductor [28,30], which prevents motion of Josephson vortices across the planes. The $R_{ab}(\Theta)$ exhibits a clear cusp-like dip at $\Theta = 0$, indicating the 2D-nature of superconductivity in the CuO planes. The dip becomes narrower with increasing field. This is particularly pronounced at $H > H_{c2} \simeq 10$ T. In this case the sample is in the normal state with a flat $R_{ab}(\Theta) \simeq R_n$, where $R_n$ is the normal state resistance, for angles $\Theta \sim 90^\circ$ at which $H_{c2}(\Theta) < H$. As the field approaches the highest $H_{c2}^\parallel$, superconductivity survives only in a narrow range of angles around $\Theta \sim 0^\circ$ where $H_c(\Theta) > H$. Therefore, a significant narrowing of $R(\Theta = 0)$ dip at $H = 17$ T in Fig. 1 (c) indicates that $H_{c2}^\parallel$ is close to 17 T.

The data in Figs. 1 (a-c) does contain information about $H_{c2}$. However, its extraction is complicated by the lack of criteria for the value of resistance at $H = H_{c2}$. Indeed, in the flux-flow case the resistivity can be approximately estimated as

$$R = R_n \frac{H}{H_{c2}}.$$  

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In this case $H = H_{c2}$ would correspond to the end of the resistive transition $R = R_s$. In the absence of flux-flow, however, $H_{c2}$ would correspond to the beginning of the resistive transition $R(H = H_{c2}) \geq 0$. This is also the case for strong pinning, as demonstrated for Nb films [9]. Thus, $R(H = H_{c2})$ depends on pinning of vortices and is almost impossible to predict a priori. This is particularly relevant for layered superconductors with a large anisotropy of pinning. Without exact knowledge of the flux-flow contribution it is impossible to confidently extract $H_{c2}$ from $R(T, H)$ characteristics at $T < T_c$. Therefore, the main focus of our analysis will be on the fluctuation part at $T > T_c$, which does not suffer from ambiguity associated with vortex pinning.

The fluctuation para-conductivity [10] is apparently seen as a tail in $R_{ab}(T)$ at $T_c < T \lesssim 10$ K. Figures 1 (d) and (e) represent normalized excess conductivities $\Delta \sigma_{ab}(T) = 1/R_{ab}(T) - 1/R_{n}(T)$, in perpendicular and parallel to ab-planes magnetic fields, obtained from the data in Figs. 1 (a) and (b), respectively. In obtaining those plots we assumed $R_{n}(T) = R_{ab}(T, H^\perp = 14$ T) and $R_{ab}(T) = R_{ab}(T, H^\parallel = 17$ T). It is seen that for both field orientations the fluctuation conductivity at $T > T_c$ decreases approximately linearly in the semi-logarithmic scale with almost field-independent slopes. This implies

$$\Delta \sigma_{ab}(T, H) \propto \exp[-a(T - T_c(H))]$$

where $a$ is some constant. A similar exponential decay of superconducting fluctuations has been reported for other cuprates [9][31]. Even though an exponential decay does not follow explicitly from theoretical analysis of fluctuation conductivity [10], it allows an unambiguous determination of the characteristic temperature scale $T_c(H)$ from the relative shift of the curves along the $T$-axis with respect to the known $T_c(H = 0)$. Since the $\Delta \sigma_{ab}(T)$ curves in Figs. 1 (d) and (e) remain almost parallel at different H, such determination of $T_c(H)$ does not suffer from widening of the resistive transition, as in the flux-flow case at $T < T_c$ in Fig. 1 (a). Therefore, thus obtained $T_c(H)$ has the same degree of certainty as $T_c(H = 0)$.

Fig. 1 (f) represents a semi-logarithmic plot of $\Delta \sigma_{ab} R_n$ vs $H \perp ab$ for the OD(4.0) sample at $T = 1.8$ K and slightly above $T_c$ at $T = 4.2$ K. It is seen that $\Delta \sigma_{ab}(H)$ decays almost exponentially also as a function of field at constant $T$. In this case the relative shift along the horizontal axis provides the characteristic magnetic field scale for suppression of superconductivity $\sim H_{c2}$. Assuming that $H_{c2} = 0$ at $T = 4.2$ K $\sim T_c$, we estimate from the relative shift of the two curves that $H_{c2}(T = 2$ K) $\approx 6$ T. This is consistent with $T_c(H^\perp = 6$ T) $\approx 2$ K, estimated from $\Delta \sigma_{ab}(T)$ scaling in Fig. 1 (d). Thus, from the analysis of fluctuation conductivity we obtain a confident estimation of $T_c(H)$ or equivalently $H_{c2}(T)$.

Figure 2 (a) contains the main result of this work: obtained $T$-dependencies of $H_{c2}$. Blue and red squares represent $H_{c2}^\parallel(T)$ for OD(4.0) and OP(4.3) crystals, respectively. Horizontal and vertical error bars correspond to the accuracy of scaling of $\Delta \sigma(T, H)$ curves according to Eq. (2). Filled circles represent $H_{c2}^\parallel(T)$ for the OP(4.3) crystal. At low $T$ estimation of $H_{c2}^\parallel$ is complicated by the lack of confident knowledge of $R_n(T)$. In Fig. 2 (e) we assumed $R_n(T) = R_{ab}(H = 17$ T). Alternatively we approximated $R_n$ from the linear extrapolation of $R_{ab}(H = 17$ T) at $T > 8$ K $[R_n(T) \approx 2.1$ Ω]. $H_{c2}^\parallel$ obtained this way is shown by stars. Up to $H^\parallel = 10$ T both approximations give the same $H_{c2}^\parallel(T)$. Therefore, those values are confident. However, at $H > 12$ T results start to depend on the choice of $R_n(T)$. Unfortunately, none of the two approximations is good enough at $T \to 0$. Qualitatively, $R_n = R_{ab}(H^\parallel = 17$ T) tends to underestimate $H_{c2}$ because it assumes $H_{c2}^\parallel(T = 0) = 17$ T. The linear extrapolation of $R_n(T > T_c)$ tends to overestimate $H_{c2}^\parallel(T = 0)$ because it assumes that $R(H = H_{c2}) = R_n$. However, without the flux-flow phenomenon $R(H = H_{c2}) \approx 0$ [9]. This is what we expect for our Lorentz force free data at $H \parallel ab$. In the absence of a better way to define $H_{c2}^\parallel$ at low $T$, in Fig. 2 (a) we also show fields $H_{50\%}(T)$ at which middle points of resistive transitions occurs for in-plane (open circles) and c-axis (open rhombuses) resistances. Those points fall inbetween the underestimating (solid circles) and overestimating (stars) analysis of fluctuation conductivity. Therefore, they provide a reasonable estimate of $H_{c2}^\parallel$ at low $T$.

From Fig. 2 (a) it is seen that $H_{c2}^\parallel(T)$ and $H_{c2}^\parallel(T)$ are qualitatively different. The $H_{c2}^\parallel(T)$ is almost linear in the whole $T$-range $H_{c2}^\parallel(T) \propto T_c - T$. Such a behavior is consistent with a conventional orbital upper critical field,

$$H_{c2}^\parallel = \frac{\Phi_0}{2\pi \xi_{ab}^\parallel},$$

where $\Phi_0$ is the flux quantum and $\xi_{ab}$ is the in-plane coherence length, $\xi_{ab}(0) = 5.5 \pm 0.2$ nm.

The $H_{c2}^\parallel(T)$ is clearly non-linear. The dashed line in Fig. 2 (a) demonstrates that $H_{c2}^\parallel(T) \propto \sqrt{T_c - T}$. At the first glance, it resembles the behavior of $H_{c2}^\parallel(T)$ in thin film multilayers [28][30],

$$H_{c2}^\parallel = \frac{3\Phi_0}{\pi \xi_{ab}^d},$$

where $d$ is the thickness of superconducting layers. However, the corresponding $d = 9.3 \pm 0.5$ nm is much larger than the thickness of CuO layers $\sim 0.2$ nm and is not connected to any geometrical length scale of the sample. Consequently, there is no agreement with Eq. (4).

The upper limit of $H_{c2}$ is determined by Pauli paramagnetism. The spin-singlet pairing is destroyed when the Zeeman spin-split energy becomes comparable to the
superconducting energy gap $\Delta$. This gives \[ H_p = \frac{\sqrt{2}\Delta}{g\mu_B}, \] (5)
where $g$ is the gyromagnetic ratio and $\mu_B$ is the Bohr magneton. In case of negligible spin-orbit coupling $g \approx 2$ this yields $dH_c^2/dT(T = T_c) = -2.25\, \text{T/K}$ for d-wave superconductors [32]. Our values $H_c^2(T)/T_c \approx 2.5\, \text{T/K}$ and $|dH_c^2/dT| (T = T_c) \approx 5\, \text{T/K}$ and especially $H_c^2(T)/T_c \approx 5\, \text{T/K}$ and $|dH_c^2/dT| (T = T_c) > 40\, \text{T/K}$ clearly exceed this limit. Most importantly, $H_p$ does not depend on orientation of the field. Therefore, paramagnetically limited $H_{c2}$ should be isotropic, irrespective of the underlying effective mass anisotropy.

According to Eq. (3), $H_p$ is determined solely by $\Delta$. Open triangles in Fig. 2 (a) represent $\Delta(T)$-dependence measured by intrinsic tunneling spectroscopy [3] on a slightly underdoped crystal from the same batch [15]. It matches nicely $H_c^2(T)$. Therefore, we conclude that the observed $H_c^2(T) \propto \sqrt{T_c-T}$ dependence is not originating from the geometrical confinement, Eq. (1), but follows the corresponding $\Delta(T)$ dependence of $H_p$, Eq. (5).

Fig. 2 (b) shows the anisotropy of the upper critical field $\gamma_H = H_{c2}/H_{c1}$. Close to $T_c$ it diverges due to different $T$-dependencies of the two fields. However, at $T \ll T_c$ it shows a tendency for saturation at $\gamma_H(T \rightarrow 0) \approx 2$. Such a low anisotropy of $H_{c2}$ is remarkable for the layered Bi-2201 compound with $\gamma_m \sim 300$. Observation of the low anisotropy of $H_{c2}$ in the extremely anisotropic Bi-2201 cuprate is the central new result of this work.

In Fig. 2 (c) we show magnetic field dependence of the angular anisotropy $[R_{ab}(90^\circ)/R_{ab}(0^\circ)]^{1/2}$ obtained from the data in Fig. 1 (c). The anisotropy is large at low fields, but rapidly decreases at $H > 7\, \text{T}$ when the paramagnetic limitation starts to play a role. At high fields it tends to saturate at $\sim 2$, consistent with $\gamma_H$ in Fig. 2 (b). As mentioned above, paramagnetically limited $H_{c2}$ should be isotropic. Therefore, a finite residual anisotropy $\gamma_H(T \rightarrow 0) \sim 2$ indicates that only $H_{c2}^1$ is paramagnetically limited, while $H_{c2}^2$ is still governed by orbital effects. Finally we note that $\gamma_H < \gamma_m$ was reported for several unconventional superconductors [6, 17, 18]. In particular, a nearly isotropic $H_{c2}$ was reported for the (Ba,K)Fe$_2$As$_2$ pnictide [33] despite a quasi-2D electronic structure. It is likely that all those observations have the same origin.

To conclude, we estimated $H_{c2}$ in the low-$T_c$ layered cuprate Bi$_2$Sr$_2$Ti$_1$$_3$CuO$_{6+\delta}$, based on analysis of the fluctuation paraconductivity. Our main result is the observation of a low anisotropy of the upper critical field $\gamma_H(T \rightarrow 0) \sim 2$, which is much smaller than the effective mass anisotropy $\gamma_m \sim 300$. This demonstrates that the anisotropy of $H_{c2}$ in unconventional superconductors may have nothing to do with the actual anisotropy of superconductivity at zero field. The large discrepancy in anisotropies serves instead as a robust evidence for paramagnetically limited superconductivity.

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