Experimental determination of $B$-$T$ phase diagram of YBa$_2$Cu$_3$O$_{7−δ}$ to 150T for $B \perp c$

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The $B$-$T$ phase diagram for thin film YBa$_2$Cu$_3$O$_{7−δ}$ with $B$ parallel to the superconducting layers has been constructed from GHz transport measurements to 150T. Evidence for a transition from a high $T$ regime dominated by orbital effects, to a low $T$ regime where paramagnetic limiting drives the quenching of superconductivity, is seen. Up to 110T the upper critical field is found to be linear in $T$ and in remarkable agreement with extrapolation of the longstanding result of Welp et al arising from magnetisation measurements to 6T. Beyond this a departure from linear behaviour occurs at $T$=74K, where a 3D-2D crossover is expected to occur.

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Recent magneto-transport measurements on high-$T_c$ cuprates have provided invaluable information in building a complete picture of these materials, essential to the development of a rigorous theory for the phenomenon of high temperature superconductivity. For example, divergence in the upper critical field at low $T$ has been reported in overdoped Tl$_2$Ba$_2$CuO$_6$ [1], Bi$_2$Sr$_2$CuO$_y$ [2] and La$_{2−x}$Sr$_x$CuO$_4$ [3]. Such results have provided the impetus for considerations of unconventional behaviour in the anisotropic high-$T_c$ cuprates such as reentrant superconductivity and the possibility to exceed the paramagnetic limit [4]. These measurements have typically relied on millisecond pulsed fields to observe upper critical fields which exceed the range (~35T) of steady field magnets. For higher $T_c$ materials such as optimally oxygen-doped YBCO ($T_c$~90K) access to the normal state requires magnetic fields well in excess of those generated by ms pulsed systems except for $T$ very near $T_c$. Explosive flux compression technology has been used to access this high field regime [5,6], in one case providing evidence for paramagnetic limiting of the upper critical field in YBCO for $B$ parallel to the superconducting layers ($B \perp c$) [6]. However, such measurements are extremely difficult and opportunities to make them are few. Single-turn coil magnetic field generators have also been used to make transport measurements on thin film YBCO [7]. While these systems do not produce fields in the range of flux compression techniques, they do allow for systematic, repeatable measurements to be made as the destruction of the coil does not damage the sample or cryostat. Nevertheless, transport measurements remain difficult since the peak field is reached in a few $\mu$s and the maximum $dB/dt$ exceeds $10^9$T/s.

Previous flux compression measurements on thin film YBCO with $B \perp c$ [8] gave an onset of dissipation $B_{ons}$=150T and an upper critical field $B_{c2}$=240T at 1.6K. This $B_{c2}$ is significantly smaller than $B_{c2}(0)$=674T predicted by Welp et al [9] who applied the Werthamer-Helfand-Hohenberg (WHH) [10] formalism, accounting for orbital effects only, following measurements of the slope $dB_{c2}/dT$ near $T_c$. This large discrepancy has been interpreted [11] in terms of the Clogston-Chandrasekhar paramagnetic limit $B_p$ [12,13] which arises when the Zeeman energy exceeds the superconducting energy gap $\Delta_0$, thus destroying the Cooper pair singlet state. Within BCS theory $B_p$=$\gamma T_c$, with $\gamma$=1.84T/K [12] which, for optimally-doped YBCO gives $B_p$~170T. In stark contrast to these results for in-plane magnetic fields, measurements [14] in the alternative and more widely-studied configuration, $B||c$, have mapped out the entire phase diagram in good agreement with the WHH model.

Here we report transport measurements which have allowed the $B$-$T$ phase diagram of YBCO to be constructed for $B$<150T in the $B \perp c$ orientation, greatly extending previous magnetisation measurements to 6T [10]. Although explosive flux compression techniques have previously been used to access this regime [5,6], the single turn coil system allows systematic measurements to be made on a single sample with both rising and falling magnetic field. Measurements on thin-film samples were made using a GHz technique [14] in a single-turn coil system [15] generating fields to 150T. The superconducting-normal transition was observed to be an equilibrium process, evidenced by the absence of any measurable hysteresis between up and down $B$ sweeps. Above $T$=74K $B_{c2}$ is found to be linear in $T$ with the slope $\alpha$=$dB_{c2}/dT$ corresponding closely to that found in magnetisation measurements [14]. Below 74K a departure from this slope is observed and is understood as arising from a transition from 3D behaviour where orbital effects quench superconductivity to 2D behaviour where paramagnetic limiting dominates.
The experimental configuration is shown in the inset to Fig. 1. A symmetric triplet coplanar transmission line (CTL) carries a microwave signal, $\nu=0.8$GHz-1GHz, past the sample and the transmission $S$ is modulated by the resistivity $\rho$ of the sample. Full details of the experimental arrangement are set out in Ref. [14]. A flow of cold He gas gives access to $T$ in the range 7K-300K with sample $T$ monitored by a AuFe-Cromel thermocouple mounted on the back side of the substrate. Two thermocouples mounted side by side on the same sample gave consistent readings to within 0.5K. Discharge of a 40kV capacitor bank into a 10mm diameter single turn copper coil generated fields to 150T and the copper coil was vaporised in the process, leaving the sample and cryostat intact [15].

A YBCO film, thickness 250nm, $T_c=87.2$K and critical current $J_c=3.14$MA/cm$^2$ at 77K, was grown by on-axis dc magnetron sputtering on a MgO (001) substrate, with the $c$-axis oriented in the growth direction, at $T=770$C in an Argon-Oxygen atmosphere with a deposition time $\sim90$min. The film was etched to produce a 20$\mu$m strip perpendicular to the CTL to match the sample resistance $\rho$ resistivity across a thin dielectric layer to a 2D sheet of electrons [12]. Sharp noise spikes in the data are attributed to GHz emission from the plasma produced in vapourisation of the single turn coil and are predominantly in the direction of increasing $S$ since the technique measures transmitted power (as opposed to voltage).

In the superconducting state the sample acts as an equipotential across the CTL, completely attenuating the microwave signal, resulting in zero transmission. As the applied field $B$ drives the sample normal, $S$ increases with increasing $\rho$. A model of this response is shown in the inset to Fig. 2, calculated assuming capacitive coupling across a thin dielectric layer to a 2D sheet of electrons [13]. Sharp noise spikes in the data are attributed to GHz emission from the plasma produced in vapourisation of the single turn coil and are predominantly in the direction of increasing $S$ since the technique measures transmitted power (as opposed to voltage).

FIG. 1. Raw GHz transmission $S$, with $B\perp c$, shown for times after the point marked $a$ (b) for $T=70$K (60K). Before these times, data are obscured by electrical noise arising from the discharge of the capacitor bank. Arrows indicate the direction of the field sweep. The insets show the experimental configuration for measurements and the field profile of the single turn coil.

FIG. 2. Fits to raw magneto-response $S(B)$ data for $T=80$K, 78K, 74K, 72K, 66K, 65K and 60K with onset fields increasing respectively. Traces have been offset vertically for clarity. Raw data are shown for comparison for $T=80$K and 74K. Definitions of $B_{ons}$ and $B_{c2}$ are indicated. The $S$ response to sample sheet resistivity $\rho$ (in $\Omega$) is shown in the inset.

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Fits to the raw data for the decreasing $B$ sweep which take into account the positive-going nature of the noise spikes are shown in Fig. 2. We define $B_{c2}$ as the intersection of the tangent to the transmission curve in the transition region and that immediately after it, following Ref. [10] on the basis that this gives values close to the 90% criterion and in good agreement with tunnelling data. For the lowest $T$ measurements, determining $B_{c2}$ becomes more difficult because there is less saturation region. We find, however, that the form of the transition is essentially the same for all $T$. $B_{ons}$ is defined as the point at which the transmission departs from $S=0$.

Fig. 3 shows the $B$-$T$ phase diagram for $T>60$K, with $B_{ons}$ and $B_{c2}$ values determined from the complete data set, a subset of which is shown in Fig. 4. In magnetisation measurements up to 6T on single crystal YBCO with $B\perp c$, $B_{c2}$ was found to be linear in $T$ with $dB_{c2}/dT=10.5$T/K [10]. The extrapolation of this slope $\alpha$ is plotted in Fig. 4 and we find that our data follow it closely down to $T=74$K where $B_{c2}$~100T. Note that in Ref. [10] this line intersects the $T$ axis slightly below $T_c$ as do the $B_{ons}$ data. This is allowed for in Fig. 4 below.
Discrepancies between $B_{c2}$ determined from resistive measurements and either magnetisation or specific heat data have been reported, and it has been argued that the latter two better probe the mean-field $B_{c2}$ than do resistivity measurements. With the definition of $B_{c2}$ used here for GHz measurements in the high field regime the good agreement we find with $dB_{c2}/dT$ determined from low field magnetisation measurements and the dashed line is simply a guide to the eye.

Although WHH theory includes paramagnetic and spin-orbit effects, in this Letter we consider the model arising from orbital effects only, as applied by Welp et al. Whereas this model predicts a departure from the slope $\alpha$ only at low $T$, we see clear evidence for a departure below 74K. Previous measurements by Nakagawa et al. provide convincing evidence for the applicability of this model to YBCO for $B||c$ (Fig. 3 inset). Near $T_c$ their data agree well with the $dB_{c2}/dT$ slope determined previously and application of the WHH result:

$$B_{c2}(0) = 0.7T_c(dB_{c2}/dT)|_{T_c}$$

(1)
gives $B_{c2}||c(0)=112T$, in close agreement with that measured. Indeed, this agreement extends over the entire phase boundary (dotted line in Fig. 3 inset). A larger $dB_{c2}/dT$ for the case $B\perp c$, due to anisotropy in coherence lengths $\xi_c$ out of plane and $\xi_{ab}$ in plane, leads to a significantly larger $B_{c2}||c(0)>600T$ in this model. A deviation well below this value is clear in Fig. 3, however. The good agreement between WHH and experiment for $B||c$ and the departure from expected behaviour for $B\perp c$ suggests that a different mechanism may be responsible for the quenching of superconductivity in the latter case.

FIG. 3. $B$-$T$ phase diagram for YBCO with $B\perp c$. Solid circles and triangles, represent $B_{c2}$ and $B_{on}$, respectively. Open symbols arise from measurements on a second sample fabricated from the same film. The inset shows the equivalent phase diagram taken from Nakagawa et al [4] with $B||c$. In both cases the solid line shows the slope $\alpha=dB_{c2}/dT$ determined from magnetisation measurements and the dashed line is simply a guide to the eye.

FIG. 4. Full phase diagram for YBCO for $B\perp c$. The $B_{c2}$ data from Fig. 3 is replotted along with low $T$ data from Refs. [4,6]. The dotted line is the WHH phase boundary calculated considering orbital effects only. The solid line is the second order phase boundary, and the dashed line, the first order BCS-FLL phase boundary taken from Ref. [2] which considers only coupling between the spins and the applied $B$. The reduced field is calculated with the energy gap defined as $\Delta_0=2.14T_c$. Misalignment of $B$ has been considered but cannot explain the low $T$ results of Ref. [6] or the deviation observed here.

Paramagnetic-limited upper critical fields have been observed in UPd$_2$Al$_3$ and $B_{c2}>B_p$ has recently been seen in (TMTSF)$_2$PF$_6$. The results in Ref. [4] provided the first evidence for paramagnetic limiting in a high $T_c$ material. The paramagnetic limit in YBCO is expected to occur at $B_p\sim 170T$, well above $B_{c2}||c(0)=110T$ measured for $B||c$ [4], and well below $B_{c2}||c(0)=460T$ predicted for $B\perp c$ with $T_c=87K$ (see Ref. [4]). The difference between WHH and experiment for $B\perp c$ is shown clearly in Fig. 4 where the WHH phase boundary and the data from Fig. 3 are plotted on a full phase diagram. The departure from this phase boundary is consistent with the results from flux compression measurements [4] (also plotted) and provides further evidence for paramagnetic limiting of $B_{c2}$ for $B\perp c$.

A possible fit to the data of Refs. [4,6] for $B\perp c$ has been obtained by including spin-orbit and paramagnetic parameters in the WHH and Maki models. Whilst it is instructive to apply these models, the value of the parameter giving rise to paramagnetic effects is tending to be unphysical, and spin-orbit scattering should be negligible in YBCO above a few Tesla since it is in the clean limit. Furthermore, the applicability of these 3D models to in-plane critical fields in layered superconductors is brought into question by analysis which has found that there is a non-zero temperature $T^*=T_c$ below which the normal cores of the vortices are smaller than the lattice constant $d$, defined by $\xi_c(T^*)=d/\sqrt{2}=8.5A$. Below $T^*$ orbital effects should no longer provide a mechanism for quenching of superconductivity and, in the absence of paramagnetic and
spin-orbit effects, $B_{c2}$ would be infinite.

A 3D-2D crossover is expected to occur near $T^*$ when $\xi_c$ becomes smaller than the inter-plane spacing. $\xi_c(70K) \sim 8\AA$ is the separation between pairs of CuO planes [24], and $\xi_c(80K) \sim d = 12\AA$ is the lattice constant [24]. The departure from the linear behaviour of $B_{c2}(T)$ observed here at $T = 74K$ is almost midway between these two characteristic temperatures. A theory which includes the finite thickness of the superconducting layers in the cuprates, but neglects paramagnetic effects [23], predicts a crossover in $B_{c2}(T)$ from linear to non-linear behaviour at $T = 0.9T^* \sim 78K$, close to the departure observed here. The fact that we see $B_{c2}$ increase less rapidly with $T$ in the nonlinear regime, in contrast to this theory [23], is most likely due to paramagnetic limiting.

For systems in which paramagnetic effects are important a finite momentum or Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) superconducting state may exist. An extension of the original theory suggests that a large ratio of orbital to paramagnetic terms, $\beta = \sqrt{2}B_{c2}(0)/B_p$, is favourable to the formation of such a state, provided the superconductor is in the clean limit [23]. Here $B_{c2}(0)$ is that defined in Eq. 1. The first reported observation of the FFLO state was in UPd$_2$Al$_3$ with $\beta = 2.4$ [18]. More recently it has been suggested that the FFLO state should be enhanced in quasi-2D superconductors [29], making YBCO, which is in the clean limit with $\beta = 5.7$ for $B \perp c$, an ideal candidate.

The phase diagram predicted [21] for $d$-wave superconductivity in layered materials with $B \perp c$ which accounts for coupling between the spins and the applied $B$ includes a FFLO phase. A comparison with experiment is shown in Fig. 2 where the only free parameter is the $g$-factor, which is set equal to 2. The agreement between this theory [21] and the low $T$ data [21] is surprisingly good, supporting the conjecture that quenching of superconductivity is driven by paramagnetic effects at low $T$. In contrast, the data close to $T_c$ is better fitted by WHH, which is probably due to the fact that the theory in Ref. [21] does not consider orbital effects. We note that $B_{ons}$ for the data of Ref. [21] coincides with the position of the first order phase transition between the zero momentum and FFLO state. This may not be accidental since the superfluid density and $J_c$ may be lower in the FFLO phase [21].

In conclusion, we have found that for $B \perp c$ the upper critical field in YBCO follows the WHH phase boundary for $T > 74K$. A clear departure is observed below 74K near the expected position of a 3D-2D crossover. The low $T$ data below the crossover is consistent with a theory [24] which accounts for coupling of the spins to the applied $B$. This suggests a transition from a high $T$ regime where superconductivity is governed by orbital effects to a low $T$ regime where paramagnetic effects dominate. Combined with the data of Refs. [26] this constitutes strong evidence for the experimental realisation of paramagnetic limiting in a high-$T_c$ cuprate superconductor.

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