Measurement of the upper critical field of optimally-doped YBa$_2$Cu$_3$O$_{7-\delta}$ in megagauss magnetic fields

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Abstract. We have observed crossover from superconducting state to normal state in optimally doped YBa$_2$Cu$_3$O$_{7-\delta}$ ($T_c \sim 90\,\text{K}$) in very high magnetic fields ($B$) up to 600 T applied parallel to CuO$_2$ planes ($B_{\parallel \text{CuO}_2}$). This is the first systematic critical field measurement at low temperatures in the geometry of the $B_{\parallel \text{CuO}_2}$-planes using superconductors with $T_c$ greater than 90 K. In order to perform high-sensitivity magneto-conductivity measurements in short-pulsed high magnetic fields, we developed a contactless radio frequency transmission technique. Ultrahigh magnetic fields higher than 200 T are generated by using electromagnetic flux compression, while a single-turn coil system is used for generating magnetic fields up to 200 T. We observed clearly the crossover from the superconducting to normal state conductivity at 250 T at a temperature of 5 K, and a systematic decrease of the critical field with increasing temperature. The field–temperature phase diagram was constructed over a wide temperature range between 5 K and 300 K.

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1. Introduction

It is generally accepted that high-$T_c$ superconductors will open up a vast area of novel applications, such as processors, storages, sensors, wires and applications for levitated trains [1]–[3]. In particular, optimally-doped YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO: $T_c \sim 90$ K) is one of the most promising superconductors since its superconductivity transition temperature is higher than 90 K, thereby exceeding the liquid nitrogen temperature. Although optimally-doped YBCO is a promising superconducting material for generating high magnetic fields [4]–[6], its upper critical fields (crossover field from superconducting to normal state) and normal state behaviours have not yet been fully understood at low temperatures. This is mainly due to the fact that the normal state of YBCO at low temperatures can be accessed only by magnetic fields higher than 100 T. Ultrahigh magnetic fields above 100 T can be generated only for a short duration (less than 10 $\mu$s) using the single turn coil technique or the electromagnetic flux compression system [7]–[9]. Because of the very short rise time of the magnetic field of the order of several microseconds, conventional resistivity measurements using the four-probe method are extremely difficult because of the induction of a huge voltage in the lead wires. Moreover, several types of electrical noise resulting from large-current switching or mechanical vibration of magnets deteriorate the signal. In addition, we have to take into account intrinsic heating due to eddy currents within the sample and contacts. Therefore, so far, experiments performed in short-pulsed magnetic fields exceeding 100 T have mainly been limited to optical measurements in low conductivity semiconductors [10]–[12].

In early days soon after the discovery of the high-$T_c$ superconductors, we have succeeded in measuring the critical fields for irreversible magnetization in both configurations $\mathbf{B}\parallel_{\text{CuO}_2}$ planes, hereafter we denote $\mathbf{B}\perp_{\text{CuO}_2}$ and $\mathbf{B}\perp_{\text{c}}$, and $\mathbf{B}\parallel_{\text{CuO}_2}$ planes, hereafter we denote $\mathbf{B}\parallel_{\text{CuO}_2}$ [13, 14]. Moreover, for $\mathbf{B}\perp_{\text{CuO}_2}$, the magnetoresistance measurements were also performed by using the four contacts method under the magnetic field up to 120 T produced by the single turn coil technique by an extremely careful sample setting [15, 16]. However, we still need a more systematic study of the upper critical field for $\mathbf{B}\parallel_{\text{CuO}_2}$ not only for basic understanding of the material but also for practical applications.

In this study, we have succeeded in systematic measurements of the critical field for optimally-doped YBCO in ultrahigh magnetic fields ($\mathbf{B}$) up to 600 T. From the measurements over a wide range of temperature between 5 and 300 K, we have obtained the critical field versus temperature phase diagram for $\mathbf{B}\parallel_{\text{CuO}_2}$-plane. In order to overcome the difficulty in resistivity measurement, we developed a contactless radio frequency (RF) transmission technique that can be used in short-pulsed high magnetic fields. Using this technique, we clearly observed the crossover from the superconducting state to the normal state, referred to as ‘$H_{c2}$’ in the following sections.
Figure 1. Schematic of contactless RF transmission apparatus and its dimensions. (a) Diagram of the apparatus, (b) cross-section of the sample, (c) micrograph of Au thin-film microcoils, and (d) detailed illustration of sample assembly including microcoils. Thick arrows represent the directions of the external pulsed magnetic field and of the RF-field generated by microcoils.

A prototype of the contactless RF transmission technique devised by Sakakibara et al was incorporated with the single turn coil technique, and its efficiency was clearly demonstrated in high field experiments up to 100 T [17, 18]. We further developed the system for experiments in higher magnetic fields generated by electromagnetic flux compression up to 600 T [9]. A preliminary report has been given in [19], but here we place the main focus more on this new experimental technique.

2. Experimental procedure

The set-up and dimensions of a contactless RF transmission technique are shown in figure 1. For the limited space available in high magnetic fields above 100 T and to reduce the eddy current heating, we set the dimension of the sample assembly as small as possible—less than 500 µm. The system comprises a set of two flat microcoils sandwiching a thin YBCO sample and the entire coil system is fixed by thermally and electrically insulating GE-varnish (figures 1(b) and (c)). The inductance of the microcoils is 100 nH. One microcoil is connected to a RF oscillator of 60 MHz that generates a RF field. The transmitted RF-field is picked up by the other microcoil that is connected to a band-pass filter, a detector, and a digital recorder, as shown in figure 1(a). Extreme care was taken so that the external pulsed magnetic field can be applied exactly parallel to the thin-film plane. By use of thin-film microcoils and by reducing the size of effective metallic area perpendicular to pulsed-high magnetic fields as much as possible, we minimized the heating effects. In fact, the exact alignment of the sample is crucial not only for measuring the highly anisotropic sample but also for reducing the area perpendicular to the external...
The samples were high-quality single-crystalline YBCO with a thickness of 30 µm and their critical temperature \( T_c \) was \( \sim 90 \) K. These samples were manufactured using molecular beam epitaxy (MBE) at the International Superconductivity Technology Center (ISTEC). The \( c \)-axis was perpendicular to the sample plane and the thickness of sample was \( \sim 30 \) µm; the sample was made to have a thickness which is smaller than the skin depth of the sample at 60 MHz. Therefore, the eddy current could penetrate through the sample and the induced RF-field on the opposite surface of the sample. Figure 2(a) shows resistivity as a function of temperature, measured using a conventional four-probe method.

Figure 2. (a) Resistivity as a function of temperature. Sample is an optimally doped YBCO single-crystal thin-film (\( T_c \sim 90 \) K, thickness \( \sim 30 \) µm). Measurements are performed by a conventional DC current by using the four probe method. (b) Temperature dependence of the transmission (real part) and (c) imaginary part output obtained by the RF method of 60 MHz.
Figure 3. (a) Experimental trace of a magnetic field generated by the single turn coil system. One trace can be divided into six sections because of the increasing and decreasing part of the field pulse and second peak due to electromagnetic induction. Inset shows the schematic of the single turn coil system. (b) Transmission data with hysteresis due to the misalignment of sample orientation, and (c) without hysteresis from exact alignment with applied magnetic fields parallel to the CuO2.

Figures 2(b) and (c) show the temperature dependence of the real part (in-phase component) and imaginary part (out of phase component) of the signal obtained from a wireless RF transmission technique respectively, for a YBCO thin film in the absence of a magnetic field. The transmission described as the real-part output shows a sharp drop at $T_c$ of 90 K as the RF-field is completely screened below $T_c$, as shown in figure 2(b). On the other hand, the transmission does not show significant changes above $T_c$. In addition, the imaginary part also shows a sharp spike-like change at $T_c$ (figure 2(c)). The data obtained from the RF method were analysed in terms of the magnetic permeability of the sample.

Figure 3(a) shows a trace of the magnetic field generated by the single turn coil technique [8, 20, 21]. In preliminary experiments with fields up to 120 T by using the single turn coil
system, we have confirmed negligible eddy current heating and perfect alignment of the applied field parallel to the CuO₂ planes. Measurements were performed at 30 K. When the face plane of the YBCO thin sample was slightly tilted from the exactly parallel direction with respect to the external magnetic field or if the YBCO was not separated from the metallic part of the microcoils, a large hysteresis was observed between the up-sweep and the down-sweep of pulsed field, as shown in figure 3(b). This is due to the interaction of eddy current heating during the up-sweep of the field pulse and cooling of the sample by heat conduction. On the other hand, an YBCO thin sample with a perfectly aligned applied field parallel to the CuO₂ planes did not exhibit any significant hysteresis. This fact implies that the eddy current heating is negligibly small even in the case of short-pulsed magnetic fields above 100 T (figure 3(c)). Furthermore, the eddy current heating increases remarkably with increasing sample thickness. On the other hand, if a sample is too thin, the difference in the transmission intensity between the superconducting state and the normal state would become too small to be measured. Therefore, in order to obtain reliable data, a choice of optimal sample thickness (30 µm in this experiment) and measuring frequency is very important. In the case of the single turn coil measurements, experimental traces are disturbed by a big trigger noise at the beginning of the current to the coil and the resulting noise lasts during the rise time of the field pulse. However, in the decreasing part of the field pulse, reliable measurements can be performed because the influence of the trigger noise dies out completely beforehand.

Figure 4 shows transmission over a temperature range of 91 to 30 K in the fields up to 110 T; these fields were obtained using the single turn coil system. \( H_{c2} \) has been determined as the point where transmission merges with the normal state level just above \( T_c \); thus we obtained \( H_{c2} \) as 110 T at 73 K, 85 T at 75 K, 70 T at 80 K, 62 T at 86 K, 42 T at 87 K, 25 T at 88 K, 9 T at 89 K, and 5 T at 89.5 K. However, at temperatures below 70 K we need a higher magnetic field. We measured transmission at lower temperatures using the electromagnetic flux compression, which can generate very high fields up to 622 T [9]. The sample assemblies were made so that
Figure 5. (a) Coil system for the electromagnetic flux compression. (b) High speed photographs of the motion of the liner. The liner is squeezed in about 50 μs. The video is made from a number of framing pictures which are taken by an image converter camera. The pictures are displayed successively as if it is a moving object [22]. (c) Experimental trace of a magnetic field generated by the electromagnetic flux compression system.

the microcoils system was interchangeable between the measurements by both the single turn coil system and the electromagnetically driven flux compression system. In this way, the exact alignment of the sample for the field $B \parallel \text{CuO}_2$ is made in the single turn coil system before the system is set in the flux compression system.

Figure 5 (a) shows the coil system used for the electromagnetic flux compression system. A large pulsed current (3–4 MA) is applied to the primary coil from a capacitor bank of 5 MJ (40 kV). Then the secondary current is induced in the liner in the opposite direction. The repulsive force between the two opposite currents rapidly squeezes the liner in the inward direction.
Figure 6. Normalized transmission measured at temperatures of 5 K, 30 K, and 45 K as a function of magnetic fields up to 300 T. Solid lines representing superconducting (0) and normal state levels (1) were determined in the absence of magnetic field below and above $T_c$, respectively.

The motion of the liner is seen in figure 5(b). This motion compresses the seed magnetic flux ($\sim 3$ T) which is injected in the liner beforehand by another set of coils [22]. In this way, a very high magnetic field is generated almost in proportion to the inverse of the cross-section of the liner.

The magnetic field trace is shown in figure 5(c) as a function of time. The maximum field recorded by the pick-up probe is different from shot to shot because it depends very much on the time of the break of the probe, but we can obtain a field 400–600 T before the break point.

3. Results and discussion

Figure 6 shows transmission as a function of magnetic fields up to 400 T at 5 K, 30 K, and 45 K as an example. The transmission level below 60 T corresponds to the superconducting state that we measured below $T_c$ before the application of magnetic fields. With increasing field above 60 T, the transmission starts increasing rapidly; we found that the transmission reached the level corresponding to the normal state at a field of 250 T at 5 K, 220 T at 30 K, and 190 T at 45 K.

It should be noted that transmission saturates at the transmission level of the normal state that was measured above $T_c$ before the application of magnetic fields. This indicates that the magnetic field where transmission saturates corresponds to the crossover from the superconducting state.
Figure 7. Magnetic phase diagram of the critical field for optimally-doped YBCO in the field for $B \parallel \text{CuO}_2$. Lines represent the Werthamer–Helfand–Hohenberg (WHH) theory including the spin Zeeman effect ($\alpha$) and the spin-orbit effect ($\lambda_{SO}$) as parameters. ($\alpha, \lambda_{SO}$) = (0, 0) represents the WHH phase boundary, which is governed by the pair breaking for the orbital effect only.

to the normal state although it has a spread of about $\pm 10$ T. We defined this field as the upper critical field parallel to the CuO$_2$ plane. These characteristics of $H_{c2}$ in the transmission were reproducible at different temperatures and were also observed at 250 T at 5 K, 235 T at 18 K, and 140 T at 65 K. Thus, the $H_{c2}$-$T$ phase diagram for $B \parallel \text{CuO}_2$ can be constructed as shown in figure 7.

We observed a large amount of noise above 260 T. In the electromagnetic-driven flux compression system, the metal tube (Cu-liner) that is imploded and compresses the seed-field (flux) leads to higher magnetic fields above 200 T, while the approach of the imploding metal tube results in the destruction of the measuring probe. The maximum available field depends on the outermost diameter of the sample holder. More details of the electromagnetic flux compression are described in [8, 9].

In our previous work on magnetic field direction perpendicular to the CuO$_2$ ($B \perp \text{CuO}_2$), we have demonstrated $H_{c2}$ for YBCO to be 120 T at a measurement temperature of 5 K; the measurements were performed using a conventional four probe method in addition to the single turn coil technique [7, 8]. However, because of a two-dimensional characteristic involved, a much higher magnetic field is needed to suppress superconductivity in the field direction parallel to the CuO$_2$ plane ($B \parallel \text{CuO}_2$). Therefore, there have been no systematic measurements of $H_{c2}$ for optimally doped YBCO in $B \parallel \text{CuO}_2$.

There have been a few experiments with regard to $H_{c2}$ for optimally-doped YBCO with $B \parallel \text{CuO}_2$, as reported by Goettee et al [23], Dzurak et al [24], and O’Brien et al [25]. Experimental results by Goettee et al using explosive-driven flux compression indicated a broad crossover from the superconducting state to the normal state in the wide range from 75 T to 340 T. The results by Dzurak et al were obtained using the GHz strip-line transmission technique in the same explosive-driven system. This result indicated the crossover at 240 T at a low temperature of 1.6 K. These previous results are in reasonably good agreement with our results at low temperatures.
Our previous measurements of $H_{c2}$ in the case of $B$ perpendicular to the CuO$_2$ plane provided a $H_{c2} \perp T$ phase diagram over a wide temperature range [7]. It provided convincing evidence for adopting a conventional theory of the upper critical field known as the Werthamer–Helfand–Hohenberg (WHH) formalism [26]. In the case of $B$ perpendicular to the CuO$_2$ plane, quenching of the superconductivity was well explained solely by the orbital effect. In contrast, we found that the $H_{c2}$ measured for $B \parallel$CuO$_2$ exhibits a large discrepancy with the WHH theory taking account of only an orbital effect; this predicts a crossover at around 600 T, as shown by a dashed line ($\alpha, \lambda_{SO} = (0, 0)$) in figure 6. A good agreement between the WHH model and the experimental result for $B \perp$CuO$_2$ and the discrepancy for $B \parallel$CuO$_2$ suggests that a different mechanism should be responsible for quenching the superconductivity in the latter case. One possible explanation for the origin of this large discrepancy is the spin–orbit effect ($\lambda_{SO}$) and the Clogston–Chandrasekhar paramagnetic limit—generally referred to as the Pauli limit—which arises when the Zeeman energy exceeds the superconducting gap, resulting in a destruction of the Cooper pair singlet state [27]. The best fit to the experimental results for $B \parallel$CuO$_2$ was obtained when we included the spin–orbit effect ($\lambda_{SO}$) and the spin Zeeman effect ($\alpha$). The value of $\alpha$ is defined from Maki’s equation, $\alpha = -0.52758(dH_{c2}/dT)_{T=T_c}$ [28], based on which $\alpha$ is evaluated to be 5.2 because the value of $dH_{c2}/dT$ near $T_c$ is around $-10$ T/K in our measurements. This is in good agreement with that from the magnetization measurement by Welp et al [29]. In the $H_{c2}$–$T$ curve, there is a specific temperature $T^*$ ($< T_c$) at which the $H_{c2}$ trace deviates from the dashed line ($\alpha, \lambda_{SO} = (0, 0)$) in figure 6. In the present experiment, $T^*/T_c$ is found to be 0.9. For $T^*/T_c < 0.9$, the spin Zeeman and the spin–orbit effects are more responsible for the quenching of superconductivity.

As is well known, there is a large controversy as to the definition of the upper critical field $H_{c2}$ because of the existence of a vortex-liquid phase and large superconducting fluctuations. Recently, in order to investigate the intrinsic upper critical field, extensive efforts toward the development of measuring techniques and systems have been made [30–32]. Therefore, we could not exclude the possibility of an underestimation for $H_{c2}$ in our experimental result. Further investigations are needed into the geometry of $B \parallel$ CuO$_2$ at low temperatures although measuring techniques are limited in the short-pulsed high magnetic fields. However, based on our definition, we can propose that superconductivity for optimally-doped YBCO can be maintained at least up to 250 T for $B \parallel$CuO$_2$ at low temperatures, which is important for practical use of optimally doped YBCO.

To summarize, we clearly demonstrated that the contactless RF transmission technique is very useful for measuring crossover from the superconducting state to normal state conductivities even in short-pulsed fields. Such systematic measurements of low-temperature normal state characteristics under high magnetic fields can facilitate a better understanding of the superconducting mechanism and then enable us to realize reliable and sophisticated applications comprising high-$T_c$ superconductors.

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New Journal of Physics 9 (2007) 47 (http://www.njp.org/)