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Transport measurements of in-plane critical fields in $YBa_2Cu_3O_{7-\delta}$ to 300 T

A. S. Dzurak, B. E. Kane, R. G. Clark, N. E. Lumpkin, J. O'Brien, G. R. Facer, R. P. Starrett, and A. Skougarevsky
National Pulsed Magnet Laboratory and Semiconductor Nanofabrication Facility, School of Physics, University of New South Wales, Sydney 2052, Australia

H. Nakagawa and N. Miura
Institute for Solid State Physics, University of Tokyo, 7-22-1 Roppongi, 106 Tokyo, Japan

Y. Enomoto
Superconductivity Research Laboratory, International Technology Center, Shinome, Koto-ku, 135 Tokyo, Japan

D. G. Rickel, J. D. Goettee, L. J. Campbell, C. M. Fowler, C. Mielke, J. C. King, W. D. Zerwekh, D. Clark, and B. D. Bartram
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

A. I. Bykov, O. M. Tatsenko, and V. V. Platonov
All-Russian Research Institute of Experimental Physics, Arzamas-16, 607200, Russia

E. E. Mitchell, J. Herrmann, and K.-H. Müller
CSIRO, Division of Telecommunications and Industrial Physics, Epping 2121, Australia

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Thin-film $YBa_2Cu_3O_{7-\delta}$ has been studied with in-plane magnetic fields B to 850 T, with good signal-to-noise to 300 T, using a GHz-transmission-line technique for electrical transport measurements in explosive-driven microsecond pulsed magnetic fields. At an ambient temperature of 1.6 K with $B\perp c$ axis we find an onset of dissipation at 150 T, the highest field reported, with a saturation of resistivity at 240 T. Comparison with the Pauli limit $B_p = 155$ T expected for this material suggests that the critical field is limited by spin paramagnetism. [S0163-1829(98)50422-5]

At low temperatures the upper critical magnetic fields B_{c2} for high-T_c cuprates exceed the maximum B available using steady-field magnets. Recent transport measurements[1] using ms-duration 60 T pulsed magnets have provided significant information on the normal state properties of cuprates having $T_c < 40$ K, however, for materials with higher T_c, critical fields can only be attained using *destructive* field generators. Such measurements are difficult, because fields above ~70 T can only be maintained for a few μs. The resulting dB/dt can reach 10^9 T/s, creating voltage up to 1 kV in a conducting loop of area 1 mm^2.

Despite the considerable technical challenges, measurements in intense fields provide the possibility of exploring new physics.[2] Reentrant superconductivity at very high B has been proposed for electron systems near the quantum limit,[3] which is in the range 100–1000 T for cuprates. Additionally, recent data[4] for low-T_c organic superconductors give B_{c2} values which exceed the paramagnetic (or Pauli) limit B_p,[5] beyond which singlet pairing of Cooper pairs is not possible within BCS theory. In the case of layered superconductors such as high-T_c cuprates, if B is directed within the layers it is possible that B_p will be reached before the conventional superconductor-normal transition associated with the collapse of the vortex lattice. The only previous low temperature measurement on $YBa_2Cu_3O_{7-\delta}$ (YBCO) with $B\perp c$ was obtained at 94 GHz in B to ~800 T and indicated a broad superconductor-normal transition in the range 75–340 T.[6] Further measurements at lower frequencies, allowing a more straightforward comparison with theory, are clearly desirable.

Here we report data to 300 T obtained in pulsed magnetic field shots to 850 T on thin-film YBCO samples with $T_c = 84$ K and a transition width ~4 K at $B = 0$.[7] Probe frequencies ν=0.9–1.9 GHz were used with the maximum $h\nu = 8$ μeV corresponding to thermal energies below 1 K. In our measurements B was directed *perpendicular* to the **c** axis. Previous measurements on similar samples in fields to ~150 T with B *parallel* to the **c** axis gave $B_{c2}^c = 110$ T (Ref. 7) and 135 T (Ref. 8) at $T = 2-4$ K. A much larger critical field ${B_{c2}}^{ab} = 674$ T (Ref. 9) is expected for $B\perp c$ due to the shorter coherence length ξ_{ab} in this orientation, however, before the magnetic length falls below ξ_{ab}, the paramagnetic limit B_p should be reached. Based upon simple BCS theory $B_p(T=0) = \gamma T_c(B=0)$ with γ=1.84 T/K,[5] yielding $B_p = 155$ T for our samples, close to the field at which we observe an onset of dissipation.

The measurement technology developed for this experiment has been fully described elsewhere.[10] Magnetic fields up to 850 T were produced using implosive flux compression in a triple-liner MC1-class generator.[11] An initial magnetic flux was compressed by chemical explosives to produce a peak B in ~10 μs, after which the generator, cryostat, and samples were destroyed. The experiment comprised two destructive pulses, in which a total of five YBCO samples were

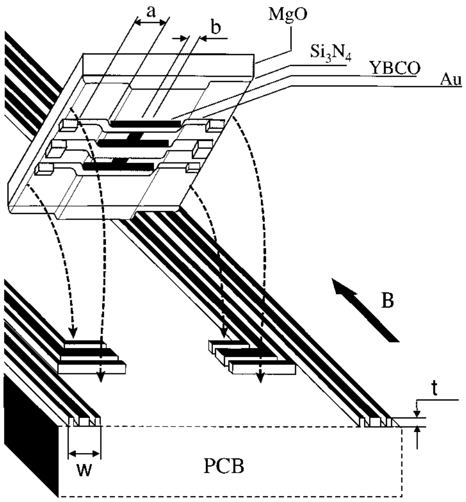

FIG. 1. Flip-chip sample mounting. Thin Au transmission lines (80 nm) bridged gaps in the thicker lines on the PCB. Six Au pads (300 nm) provided reliable contacts and the chips were held in place using a heat curable epoxy. Here $a=150$ μm, $b=10$ μm, $t=9$ μm, and $w=410$ μm.

monitored at various power levels, along with a number of semiconductor samples. Here we describe results obtained at the highest power level only (~1 mW), since lower powers gave poor signal-to-noise. Electrical connections to the samples were made using Au/Cu coplanar transmission lines (CTL's) patterned on a microwave printed circuit board (PCB) designed to avoid Faraday pickup.

The experimental configuration is shown in Fig. 1 and probes the in-plane resistivity ρ_{ab} of YBCO through its modulation of the transmission S of GHz radiation. Care was taken to ensure sample alignment within the MC1 generator to better than 1°, minimizing the contribution from any component of B along the **c** axis. If the sample is superconducting the inner and outer conductors of the CTL triplet are shorted so that S is zero, except for a small contribution from cross-talk discussed below, whereas a perfect insulator has no effect on transmission. Although the sample impedance at 1 GHz is a complex quantity the measurement provides no phase information so, for simplicity, we assume that the impedance corresponds to a scalar resistivity ρ.[12]

The YBCO films, of thickness 100 nm, were prepared by pulsed laser deposition on MgO (001) substrates with the **c** axis in the growth direction. Thermal isolation was achieved by patterning thin (80 nm) Au CTL's directly onto the samples (Fig. 1), bridging a gap in the thick (9 μm) CTL's on the PCB, with an estimated maximum T increase at the sample of only 2 K from heating in the adjacent thick CTL's on the time scale of the field pulse.[10] To avoid the problem of erratic Ohmic contacts at large B, a 50 nm layer of Si_3N_4 dielectric was sandwiched between the YBCO mesa and the metal CTL's, so that the coupling between sample and CTL's was capacitive. To estimate T at the sample, $Ge_{1-x}Au_x$ thin-film thermometers with large T-dependent resistivities were fabricated in a manner analogous with the YBCO samples. During MC1 pulses the thermometers allowed an upper bound of 10 K to be placed on the sample temperature for $B<300$ T.

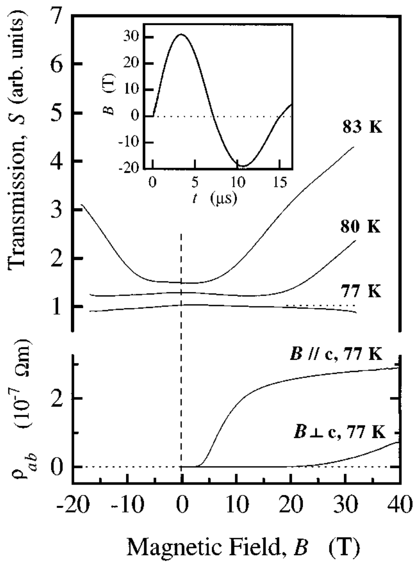

FIG. 2. Trials in μs and ms pulsed B fields prior to MC1 shots. *Upper panel:* S (in *arbitrary units)* plotted as a function of B with $B\perp c$ and $\nu=1.1$ GHz in μs pulsed fields generated using a single-turn coil technique in Tokyo. The curves have been offset for clarity and show data obtained in one field swing from 30 T to -17 T. *Inset:* Time profile of the single-turn coil field pulse. *Lower panel:* In-plane resistivity ρ_{ab} of the YBCO films obtained from four-terminal dc measurements using ms pulsed fields in Sydney.

Prior to the MC1 shots, the YBCO samples and measurement system were characterized using a single-turn coil pulsed field system[13] to assess the importance of dynamic effects. This system produces a 30 T nondestructive field with a rise time ~3 μs (inset to Fig. 2), giving a peak $dB/dt \sim 10^7$ T/s which, while around an order of magnitude smaller than the MC1 generators up to 300 T, provides indicative information. T was controlled between 77 K and $T_c=84$ K, so that the superconductor-normal transition could be observed for $B\perp c$. With a pulse cycle time of a few minutes it was possible to carry out a detailed study and the data was found to be reproducible after thermal cycling. The transmission $S(B)$ is plotted at three temperatures approaching T_c in Fig. 2 (upper panel). The onset of dissipation at $B_{on}{}^{ab}$ is evident in the two higher T traces, although S does not saturate, indicating $B_{c2}{}^{ab}>30$ T in both cases.

The lower panel in Fig. 2 shows $\rho_{ab}(B)$ at $T=77$ K obtained in a *millisecond* pulsed B, where dc resistance measurements are straightforward. With $B\perp \mathbf{c}$ we find a nonzero resistance for $B>20$ T. The GHz data at 77 K shows a decrease in S consistent with this, resulting from coupling of signal input to output along the 300 mm long tail of the PCB, so that $S>0$ even when $\rho=0$, as seen in Fig. 2. Interference of the transmitted and reflected modes along the CTL's then produces an initial *decrease* in S as ρ *increases,* although S will always approach unity as $\rho\rightarrow\infty$.[14] The full response function $S(\rho)$ is plotted as an inset to Fig. 3.

Nonequilibrium effects in the YBCO samples are unimportant on ms time scales or longer, since our dc resistance data in ms pulsed B to 50 T showed negligible hysteresis between field sweeps with B increasing and decreasing. However, single-turn coil measurements on μs time scales with $B\|\mathbf{c}$ indicate that $B_{c2}{}^{c}$ and $B_{on}{}^{c}$ can occur at *lower*

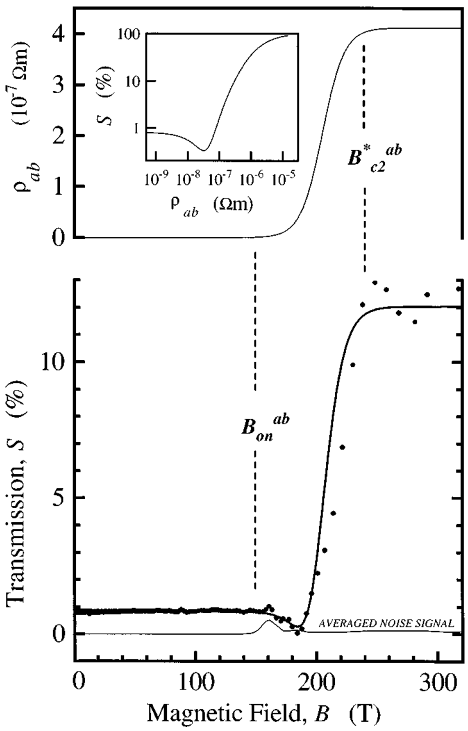

FIG. 3. Measured normalized transmission S data (•) for YBCO obtained in an MC1 pulse with $T=1.6$ K, $\nu=0.9$ GHz, and a power $\sim$1 mW at the sample. The bold line is a fit to this S data. A small averaged noise background (fine line) has been subtracted from the data. The upper curve is the calculated resistivity, assuming the transmission function $S(\rho_{ab})$ shown in the inset.

fields than when measured in ms pulses, probably due to eddy current heating.[7] In our measurements any time dependence resulting from eddy current heating or flux flow dynamics should be much smaller, since B is directed in the plane of the YBCO film. Significantly, the results in Fig. 2 confirm this, with the μs data being almost perfectly symmetric about $B=0$, inferring only minor time-dependent effects.

With this check in place, Fig. 3 shows results for $T=1.6$ K, i.e., $T/T_c\sim0.02$, in an 850 T MC1 field pulse. Only data to 320 T is plotted, since results above this field were obscured by noise, discussed below. We observe the onset of a dissipative ($\rho>0$) state at $B_{on}{}^{ab}=(150\pm20)$ T and define $B^{*}{}_{c2}{}^{ab}=(240\pm30)$ T as the field at which S, and therefore ρ, saturates, using an asterisk to indicate that the transition may result from paramagnetic limiting. The uncertainties in B reflect the noise-limited accuracy with which we can define inflection points in the S data, together with an estimate of timing accuracy.

To obtain *resistivity* from the transmission data in Fig. 3 we calculated the response function $S(\rho)$ (inset to Fig. 3) using the known capacitance between CTL's and sample, and the fraction of the signal that directly leaks from input to output, determined from S at $B=0$. The resistivity $\rho_{ab}(B)$ was then determined using an inverse response after smoothing the raw $S(B)$ data and the result is plotted in Fig. 3. The normal state resistivity $\rho_{ab}=4.1\times10^{-7}$ Ωm in Fig. 3 is 30% above the resistivity at $T=77$ K and $B=40$ T obtained from dc measurements with $B\|c$ (Fig. 2). Although a reduced ρ_{ab} is expected at lower T, this has probably been offset by positive magnetoresistance in the film.

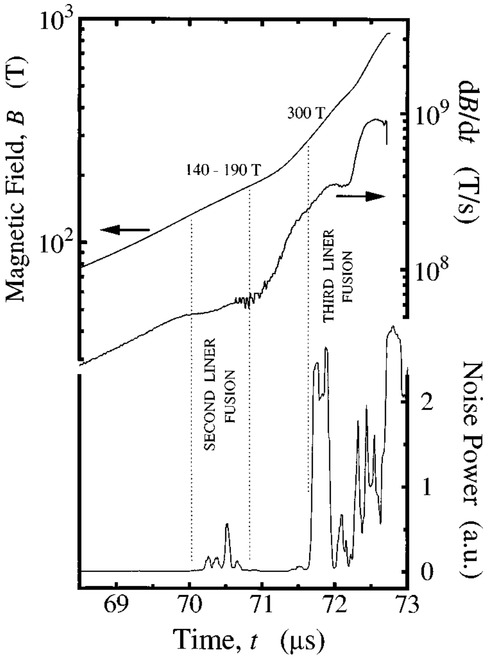

FIG. 4. B and dB/dt plotted as a function of time for an MC1 generator field pulse. Also plotted is the noise power on a bare transmission line with no sample or input signal. Fusion occurs when the shock wave hits a liner in the generator, causing electrical noise and a slowdown in the rise of dB/dt.

To determine the YBCO response in Fig. 3 accurately it was necessary to subtract from the raw data a noise background (as shown), obtained by monitoring the CTL's with no input power. Below 300 T the noise is small and occurred in the interval 140–190 T, due to the fusion of metal wires comprising the second of the three generator liners. Above 300 T wire fusion proceeded in the third liner with the noise creating severe problems for transport measurements. The origin of the noise is evident in Fig. 4. When the implosive shock wave fused a liner in the generator, the increase in dB/dt slowed as kinetic energy was absorbed and electrical noise greatly increased. To obtain measurements at higher B, single-liner generators with no electrical fusion noise will be a necessary compromise.

To our knowledge $B_{on}{}^{ab}\sim150$ T is the largest field in which a superconducting phase has been observed. A broader superconductor-normal transition than observed here, spanning 75–340 T, has been observed in previous measurements,[6] possibly due to the high frequency (94 GHz) used. We also note that the strongest saturation feature present in this data[6] is near our critical field of 240 T.

The importance of spin paramagnetism in a type-II superconductor can be assessed by comparing B_p with the critical field $B^0{}_{c2}$ determined by orbital effects alone, given by $B^0{}_{c2}(T=0)=0.70\ T_c(\partial B_{c2}/\partial T)|_{T_c}$.[15] The paramagnetic limit B_p is determined within BCS theory from $\Delta_0=\sqrt{2}\mu_B B_p$, which relates the energy gap Δ_0 with the Zeeman energy, where μ_B is the Bohr magneton. This then gives $B_p=\gamma T_c$ with $\gamma=1.84$ T/K.[5] A treatment which goes beyond BCS theory by including strong coupling effects for d-wave superconductivity predicts $1.5<\gamma<1.8$ over a wide parameter range.[16] For most superconductors $B_p > B^0{}_{c2}$, although there are exceptions. For example, the upper critical fields of Nb_3Sn and some related compounds have been known to be determined by paramagnetic limiting for some time.[17] More recent measurements have also demonstrated this for the heavy-fermion compound UPd_2Al_3.[18]

In high-T_c cuprates $B^0{}_{c2}$, as determined by measurement of $(\partial B_{c2}/\partial T)$ close to T_c, is generally much larger for $B \perp \mathbf{c}$ than for $B \parallel \mathbf{c}$, due to the shorter coherence lengths perpendicular to the CuO planes. In YBCO, $B^0{}_{c2}{}^{c} = 122$ T and $B^0{}_{c2}{}^{ab} = 674$ T, corresponding to $\xi_c = 16$ Å and $\xi_{ab} = 3$ Å.[9] Assuming the BCS value of $\gamma = 1.84$, for our samples $B_p = 155$ T and so paramagnetic effects are not significant when $B \parallel c$, as confirmed experimentally,[7,8] but should be important when $B \perp c$.

The observations that $B^*{}_{c2}{}^{ab}$ is well below $B^0{}_{c2}{}^{ab}$ and that $B_{on}{}^{ab}$ coincides with B_p, within experimental uncertainty, suggest that paramagnetic limiting determines the critical field in YBCO for $B \perp \mathbf{c}$. The fact that $B^*{}_{c2}{}^{ab}$ exceeds the BCS value of B_p by 50% in our measurements means that more detailed models may be necessary to determine B_p in cuprates, or possibly, that spin relaxation dynamics may be relevant, since B increases from 150 T to 240 T in only ~ 1 μs. More speculatively, Fulde-Ferrell[19] or triplet superconducting phases may exist in the range 150–240 T. Finally, it has been pointed out that resistivity data does not always give an accurate value for the critical field,[20] with saturation values often exceeding B_{c2} determined from specific heat or magnetization measurements, although this discrepancy is thought to be associated with vortex lattice melting and may not necessarily occur when paramagnetic limiting dominates.

In summary, via a clear transport signature we have measured $B^*{}_{c2}{}^{ab} = 240$ T for thin-film $YBa_2Cu_3O_{7-\delta}$, with an onset of dissipation at 150 T, representing a new upper bound for superconductivity in any material. From a comparison with $B^0{}_{c2}{}^{ab} = 674$ T, assuming no spin paramagnetism, and $B_p = 155$ T, it would appear that the critical field with $B \perp c$ is primarily determined by paramagnetic limiting, in contrast with the case for $B \parallel c$. This may also be the case for other cuprate superconductors, particularly those with greater anisotropy than YBCO. Although electrical noise remains a problem at low signal levels, alternative magnetic generators should open the way for measurements at even higher fields, where phenomena such as reentrant superconductivity may occur.

The flux compression measurements to 850 T reported here took place at Los Alamos National Laboratory as part of the *Dirac Series* collaboration. We thank M. von Ortenberg, F. Herlach, J. S. Brooks, and H. Yokoi for assistance and advice during the Dirac Series and R. H. McKenzie and G. S. Boebinger for useful discussions. This work was supported by the U.S. Department of Energy, the U.S. National High Magnetic Field Laboratory (NSF Cooperative Agreement No. DMR-9527035), and the Australian Research Council (Grant No. A69702465).

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