Intragranular and Intergranular Superconducting Properties of Bulk Melt-Textured YBCO

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***Abstract*—We have investigated the superconducting properties of bulk melt-processed YBCO using different experimental techniques. First, single domain materials (i.e. containing no grain boundaries) have been studied. Electrical resistivity, current-voltage characteristics, M-H loops, and A.C. susceptibility have been measured for different field and current directions. The intragranular properties are characteristic of high-quality melt-processed YBCO material, with a critical current density J_c(T=77K, B=1T//c) > 10^4 A/cm^2. The anisotropy ratio $J_c(ab)$ / $J_c(c)$ is found to be close to 3. Both magnetic and transport measurements show that defects in the microstructure do not significantly impede the current flow inside the single domain. These results were compared to those measured on samples containing one "natural" single grain boundary which sometimes appears during the grain growth process. The intergranular properties show a much stronger current and magnetic field dependence than that measured within the grain and these differences are discussed.**

I. Introduction

Bulk melt-textured $YBa_2Cu_3O_7$ (Y-123) specimens have been shown to be promising candidates for a wide range of engineering applications including motors, magnetic bearings and flywheels [1]. The properties which make these materials attractive are (i) a high critical current density J_c at 77K, (ii) a large irreversibility field H_{irr} , which results in a moderate decrease of J_c in external magnetic fields and (iii) the ability to produce large grains, called single domains. From a microscopic point of view, a single domain consists of a pseudo-crystalline matrix composed of large parallel Y-123 crystal plates (platelets) having common c-axes, but which may be slightly rotated in the ab-plane. A single domain includes numerous defects, such as Y_2BaCuO_5 (Y-211) inclusions, stacking faults, twins and microcracks. In this paper, we analyse the anisotropy of transport and magnetic measurements carried out on single grain specimens in order to investigate the general influence of defects parallel to ab planes, i.e. cracks and platelet boundaries, on the current flow uniformity.

On the other hand, bulk materials, and especially those of the largest size, frequently contain a few large single domains which are known to be separated by so-called grain boundaries. At this time, systematic investigations of single grain boundaries have been already reported for thin films [2], intergrown bicrystals [3], dual-seeded bicrystals [4] or melt-textured YBCO [5]; these studies show different types of intergranular coupling which depend strongly on the boundary microstructure. In the present work we concentrate only on the behaviour of high-angle, Josephson-like, grain boundaries which may form during the YBCO growth process. These *intergranular* properties will be compared to the *intragranular* characteristics mentioned above.

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II. Experimental Procedure

Large grain melt-processed Y-123 specimens were prepared by a top seeding technique. The details of the synthesis process as well as microstructural characterizations of the single grain material have been described in a previous paper [6].

Transport measurements were carried out on bar-shaped samples cut from the initial melt-textured specimen using a wire saw. Their typical size was 2mm x 0.3mm x 0.3mm. Each sample contained either one single grain or two adjacent grains. In the latter, the boundary was approximately perpendicular to the bar length and its microstructure could be observed on all 4 faces of the sample after polishing. Contacts were deposited by thermal evaporation of silver through masks. Then 25 µm diameter gold leads were attached with silver epoxy annealed at 420°C in flowing O_2 for 5 minutes. The resistance of all contact pairs (including wires) was less than 3Ω. A standard 4-point technique was used for electrical resistivity and DC I-V measurements. In samples containing grain boundaries, voltage contacts were placed within the grains as well as across the grain boundary. For the R(T) measurements, an alternating current (72 Hz) was injected through the sample and the inter- and intra- grain potential differences were recorded simultaneously using low-noise transformers and lock-in amplifiers.

DC magnetic measurements were made using a commercial Quantum Design SQUID magnetometer. A scan length of 5cm was chosen to ensure a reasonable field uniformity. AC magnetic susceptibility and flux profiles were performed in a home-made susceptometer based on a cryocooler [7]. Parallel

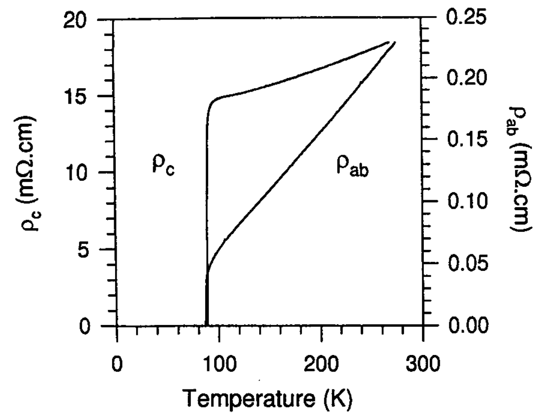

Fig.1. Electrical resistivities ρ_{ab} and ρ_c versus temperature for single grain samples

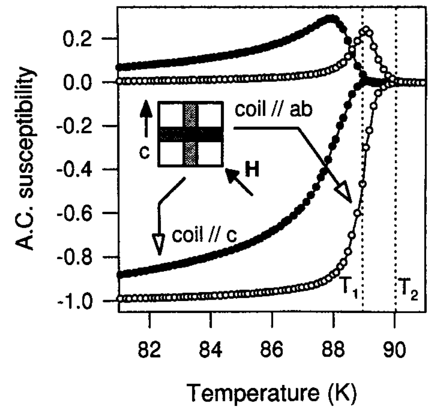

Fig. 2. AC magnetic susceptibility versus temperature measured by two orthogonal sensing coils wounded on the single grain. The DC and AC applied inductions are 50 mT and 3 mT respectively, the frequency is fixed at 1kHz.

AC and DC magnetic fields were generated by an electromagnet which can be rotated in an horizontal plane whereas the sample's magnetic response was measured by one or two orthogonal pick-up coils made of fine copper wire (50μm diameter) and positioned close to the sample surface. Each pick-up coil signal was applied to an EGG 5210 lock-in amplifier which allows the real (χ') and the imaginary (χ'') components of AC susceptibility to be determined.

III. INTRAGRANULAR PROPERTIES

First, the quality of the single grain material was probed by *measuring* the ab-plane electrical resistivity, ρ_{ab}, as a function of temperature. Results (Fig. 1) clearly show the following properties : (i) ρ_{ab} is comparable with single crystals values [ρ_{ab}(300K) ≈ 250 μΩ.cm], (ii) the normal-state $\rho_{ab}(T)$ is quasi-linear and extrapolates nearly through the origin (iii) the transition is very sharp (<1K). These features are characteristic of high-quality melt-textured samples [8].

The c-axis resistivity ρ_c as a function of temperature is plotted on the same graph. In this case, the injection current is perpendicular to the ab planes and crosses microcracks and platelets boundaries. As can be seen, ρ_c is much larger than ρ_{ab} ; the anisotropy ratio ρ_c / ρ_{ab} is 80 at T = 300K. Since this value is comparable to those measured on Y-123 single crystals (30 to 100) [9], this would suggest that platelet boundaries and microcracks do not significantly impede the flow of current parallel to the c-axis.

The anisotropy of AC magnetic susceptibility has been investigated using the technique described above. A square-shaped sample was subjected to a 3mT alternating magnetic field (1 kHz) superimposed on a static 50 mT magnetic field. Both AC and DC magnetic fields were parallel and directed at 45° with respect to the c-axis of the single grain, as illustrated in the inset of Fig. 2. The shielding signals were recorded using a pair of perpendicular coils mounted, respectively, parallel and perpendicular to the sample's c-axis. Results are presented in Fig. 2. For $T_1 < T < T_2$, only the c-axis component of the magnetic flux (measured by the coil // ab) is shown to be screened, whereas the ab component (measured by the coil // c) is not modified. Shielding currents circulating parallel to the c-axis are seen to be efficient for $T < T_1$ only. The 1K shift between both onset temperatures, T_1 and T_2, suggests an anisotropic T_c distribution within the sample. This may be due to cation contamination at the platelet boundaries or might be explained by considering different oxygenation levels within the single grain [10]. Indeed, microcracks are known to favour the oxygen diffusion throughout the specimen. Thus one expects the better reoxygenated zones to be generally aligned along the ab planes. When decreasing temperature, shielding currents are first confined in oxygen-rich regions and therefore only contribute to the magnetic shielding for the signal measured by the coil parallel to the ab planes. As temperature is decreased further, the oxygen-poor regions become superconducting as well; consequently induced currents flow along the whole cross-section and allow for an efficient magnetic shielding in both directions. In conclusion, this kind of measurement shows weakly anisotropic T_c and J_c inhomonogeneities in the melt-textured YBCO single grains.

Fig. 3 shows the M-H loops measured on a cubic single grain sample at T = 77K for H // ab and H // c. The well-known "fishtail" is clearly visible at $\mu_0 H \approx 0.9$T for H // c. A correct use of the Bean model [11] for determination of J_c from magnetisation measurements requires a knowledge of the characteristic diameter of the induced current loops. For H // c, shielding currents circulate in the ab-planes and the current loops size is expected to be macroscopic. For H // ab however, it is of interest to know whether shielding currents circulate on a macroscopic lengthscale, crossing cracks and platelet boundaries, or whether they are limited by defects and flow in significantly smaller subregions compared to the

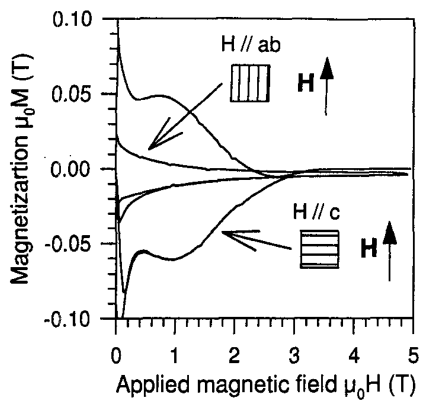

Fig. 3. Magnetization versus magnetic field for single grain samples with H//ab and H//c at a temperature of 77K.

grain size, as suggested in ref. [12]. In order to investigate this, AC magnetic flux profiles, determined following the Campbell procedure [13], were measured first on a single grain sample with H // ab. Then this specimen was then cleaved along the ab planes, both identical parts were repositioned side by side, glued with insulating GE varnish, and the same measurement repeated. The sample geometries and results are summarized in Fig. 4. The first notable feature is that both flux profiles are curved. Such a deviation from linearity has also been observed in melt-textured Nd-123 specimens [14]. In our case, it can probably be attributed to a slightly higher critical current density near the sample's surface. When comparing both curves, it turns out that for a given magnetic field value, the AC penetration depth is clearly higher in the case of the cleaved sample than for the original large specimen. Both mean slopes differ approximately by 12% which is in agreement with analytical calculations [15] which assume that induced current flows in macroscopic loops and take into account a critical current density anisotropy. On the other hand, if the induced current loops were microscopic, the flux profiles would not be modified by the cutting and rejoining processes. Consequently, this measurement provides an indirect but reasonable proof that shielding currents flow over a length scale comparable to the sample size. Note that M-H loops measurements using a similar configuration lead to the same conclusions [15].

The critical current densities $J_c(ab)$ and $J_c(c)$ were determined from the above magnetisation measurements using the Bean formula [11] and assuming current flows over the whole sample dimensions. This leads to a value of $J_c(ab)$ at $T = 77K$ and $B = 1T$ equal to 1.3 10^4 A/cm^2. The anisotropy ratio $J_c(c) / J_c(ab)$ lies between 3 and 4 at low fields. Such a value agrees with independent trapped flux measurements carried out on the same material using a miniature Hall probe [15].

From the above set of experiments, it can be concluded that the single grain properties are characteristic of high-quality melt-processed YBCO and that defects parallel to the ab-planes do not significantly influence the current flow uniformity. However, despite the long oxygenation treatment applied to these samples, some sub-regions parallel to the ab planes appear to be less reoxygenated than others. Therefore the quality of the single grain may probably be further enhanced by improving the oxygenation uniformity.

IV. Intergranular Properties

It is of interest to compare the intragranular characteristics to the properties of grain boundaries naturally occurring during the pellet synthesis. As pointed out by Küpfer [16], single grain boundaries cannot be easily detected using magnetic measurements because of the small intergrain volume. Therefore only transport measurements were carried out. Fig. 5 compares the electrical resistance measured within single grain (Gr.) and that measured across a high-angle grain boundary (G.B.). The injected current was 3 mA

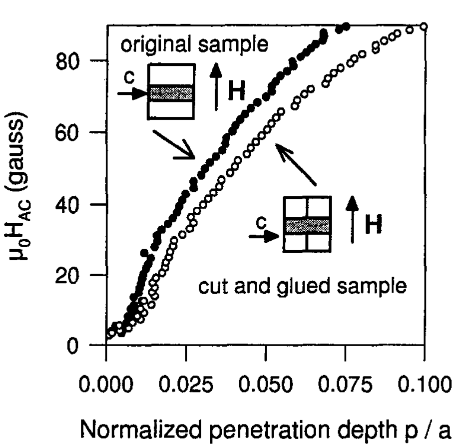

Fig. 4. Flux profiles measured at T = 77K and for an applied DC induction of 0.15T.

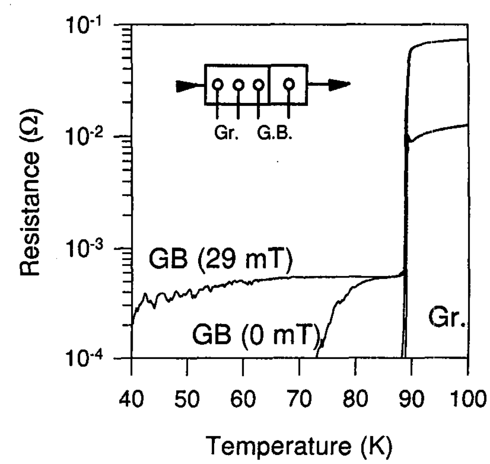

Fig. 5. Comparison of the resistance measured across a high-angle grain boundary (GB) and within an YBCO grain (Gr.) for two magnetic fields.

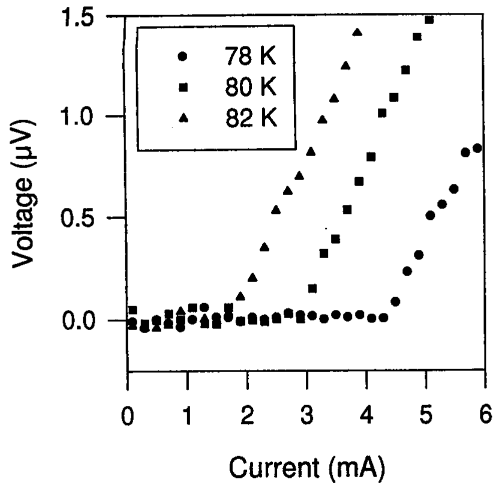

Fig. 6. I-V curves measured across the grain boundary at several temperatures.

(corresponding to a current density of 1.5 A/cm^2) and the applied magnetic field was either zero (strictly speaking, the self field) or 29 mT. The grain transition is seen to be very sharp and falls monotonically from the onset of T_c to below the noise-resolution of the voltmeter. The grain boundary characteristic, however, breaks off from the grain curve to exhibit a large resistive tail. The plateau region formed by this tail extends to significantly lower temperatures when a small 29mT magnetic field is applied parallel to the boundary plane, whereas the grain curve remains almost unaffected. Further, I-V curves (Fig.6) recorded at different temperatures exhibit a sharp onset of voltage at I_c. These features are a characteristic of a clear Josephson-like behaviour for this kind of grain boundary. The intergranular current density extracted from the I-V curves at 78K and zero field was approximately 2 A/cm^2, which is 4 orders of magnitude lower than the intragranular J_c. Therefore we can conclude that the trapped induction generated by a multi-grain pellet is strongly limited by high-angle grain boundaries and is therefore mainly due to intragranular currents.

V. Conclusions

From the above study we can conclude that
(i) the YBCO single grains exhibit characteristics of high-quality melt-textured material
(ii) the intragranular defects parallel to ab-planes do not significantly alter the current flow parallel to the c-axis
(iii) both T_c and J_c within the melt-textured sample appear to be non-uniform and anisotropic
(iv) the naturally occuring high-angle grain boundaries are characterized by an extremely low critical current density, sensitive to low magnetic fields.

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