# Anomalous behaviour in the critical current density of a $(Bi_{2-x}, Pb_x)Sr_2Ca_2Cu_3O_{10-\delta}$ tape below 10 K

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Abstract. The critical current density of a  $(Bi_{2-x}, Pb_x)Sr_2Ca_2Cu_3O_{10-\delta}$  silver-sheathed tape has been measured in fields up to 15 T and at temperatures from 2 K up to  $T_c$ . Measurements were made for three different field-current orientations of the tape. Below 10 K there is a dramatic anomalous rise in the critical current as the magnetic field is decreased below a value  $B_A$ . The value of  $B_A$  is temperature dependent but orientation independent. At 4.2 K and 2 K,  $B_A$  is 1.5 T and 2 T respectively. At 10 K and above this anomalous behaviour disappears. The effect is attributed to the existence of a low- $T_c$  (~10 K) superconducting phase at the grain boundaries in the tape and clearly demonstrates the importance of intergrain connectivity in obtaining high critical current densities in  $(Bi_{2-x}, Pb_x)Sr_2Ca_2Cu_3O_{10-\delta}$  tapes.

#### 1. Introduction

The  $(Bi_{2-x}, Pb_x)Sr_2Ca_2Cu_3O_{10-\delta}$  (Bi-2223) compound remains the most promising high-temperature superconductor for high-current applications. Unfortunately, when fabricated in the tape form necessary for such applications, the critical current density,  $J_c(B, T)$ , of Bi-2223 is significantly smaller than that found in thin films of the same material. The reasons for the low measured values and strong field dependence of  $J_c(B, T)$  in Bi-2223 tapes are still unclear.

In this paper, detailed measurements of  $J_c(B,T)$  for a Bi-2223 tape are presented. Data were obtained for three different sample orientations in the temperature range between 2 K and  $T_c$  in magnetic fields up to 15 T. These data provide direct evidence that a low- $T_c$  phase with a  $T_c$  of about 10 K at the grain boundaries may be limiting the critical current density in this tape.

## 2. Sample preparation

The monocore tape was made using a standard powder in tube method by BICC Cables. The original precursor powder was similar to the 'Endo' composition [1] which was then doped with 10% Ag. The heat treatment consisted of a total of 120 hours at 840°C in air with two intermediate pressings of the tape. After the final treatment the tape was cooled to room temperature at 100°C h<sup>-1</sup> incorporating a hold at 500°C for 5 hours. From optical micrographs, the superconducting cross-sectional area was found to be 0.0466 mm<sup>2</sup>. Preliminary measurements showed that  $J_c$  was above  $10^4$  A cm<sup>-2</sup> at 77 K in zero field—typical of good quality superconducting tapes.

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## 3. Experimental procedure

Critical current measurements were made using the Durham  $J_c(B,T)$  probe. For high current measurements in high fields, accurate temperature control of the sample space is necessary. In particular, measurements between 4.2 and 30 K are most difficult because the heat capacity of the materials is small but the transport currents are high. Above 4.2 K, the sample is enclosed in a variable-temperature gas-cooled environment which incorporates a heater, a Rh-Fe standard thermometer and a field-independent capacitance thermometer, all in intimate thermal contact with the sample. Measurements at 4.2 K and below were made with the sample in direct contact with liquid helium. The exact experimental procedure and equipment design are detailed elsewhere [2]. Critical current values quoted are accurate to the equivalent of  $\pm 70$  mK at 10 K,  $\pm 100$  mK at 80 K and  $\pm 50$  mK at 4.2 K and below. The measured sample was a short section (approximately 10 mm long) cut from a tape of 30 mm length. A criterion of 2  $\mu$ V cm<sup>-1</sup> was used to determine  $J_c$ .

Initially,  $J_c(B, T)$  was measured from 4.2 K up to  $T_c$  for three different orientations of the magnetic field with respect to the tape (in all cases the macroscopic transport current was parallel to the plane of the tape):

- (i) the field perpendicular to the plane of the tape (B parallel to the c-axis);
- (ii) the field parallel to the plane of the tape but perpendicular to the transport current (B parallel to the ab-plane) and
- (iii) the field parallel to the plane of the tape and also to the transport current (the Lorentz force-free orientation).

The measurements were taken in increasing magnetic fields. Before and after each change of sample orientation the critical current of the tape was measured in liquid nitrogen. No change in the critical current was observed. This ensured that the sample had not been damaged during handling or from thermal cycling.

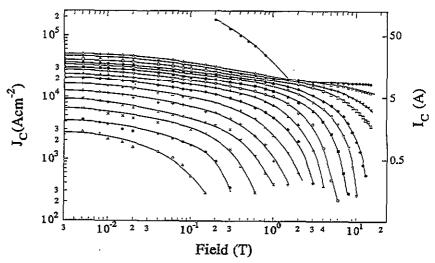


Figure 1. The critical current density for the *B*-field parallel to the *c*-axis of the tape on a log-log scale. The curves are measurements at constant temperatures of (from the top down) 4.2 K, 10 K, 20 K, 30 K, 40 K, 45 K, 50 K, 55 K, 60 K, 65 K, 70 K, 75 K, 80 K, 85 K, 90 K, and 95 K respectively.

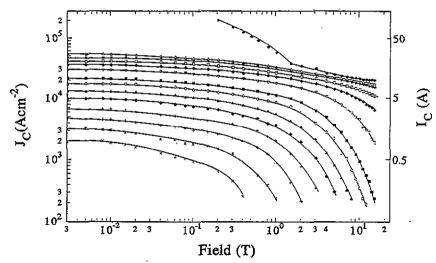


Figure 2. The critical current density of the tape for the *B*-field parallel to the surface of the tape and orthogonal to the direction of macroscopic current flow on a log-log scale. The curves are measurements at constant temperatures of (from the top down) 4.2 K, 10 K, 20 K, 30 K, 40 K, 50 K, 60 K, 65 K, 70 K, 75 K, 80 K, 85 K, 90 K, and 95 K respectively.

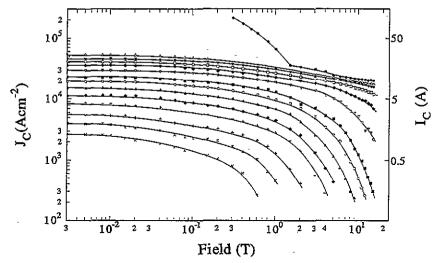


Figure 3. The critical current density of the tape for the *B*-field parallel to the surface of the tape and parallel to the direction of macroscopic current flow on a log-log scale. The curves are measurements at constant temperatures of (from the top down) 4.2 K, 10 K, 20 K, 30 K, 40 K, 50 K, 60 K, 65 K, 70 K, 75 K, 80 K, 85 K, 90 K, and 95 K respectively.

## 4. Experimental results

The  $J_c(B, T)$  data are plotted for the three orientations on a log-log scale in figures 1-3. In all three figures there is a marked anomaly in  $J_c(B)$  at 4.2 K at a field, denoted  $B_A$ , of 1.5 T. At higher temperatures this sharp change in field dependence has disappeared.  $J_c$  was found to be zero in the self-field at 110 K.

Additional data were later taken in liquid helium at 4.2 K and 2 K in a second series of measurements. The magnitude of  $J_c$  was 10-20% smaller than in the initial series of experiments; indicating a small degradation in the sample but the magnetic field and orientation dependence are the same. We attribute the degradation to limited microcracking on thermal cycling after the initial set of measurements.

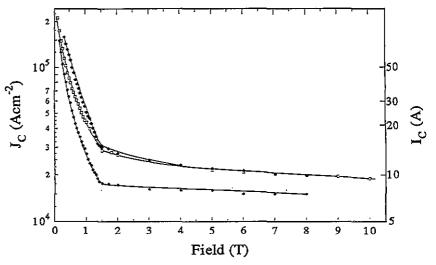


Figure 4. A comparison of the critical current density at 4.2 K in increasing field for the three orientations:  $\bullet$ , B-field parallel to the c-axis;  $\square$ , B-field parallel to the surface of the tape and orthogonal to the direction of macroscopic current flow;  $\blacklozenge$ , B-field parallel to the surface of the tape and parallel to the direction of macroscopic current flow.

In figure 4 the data obtained at 4.2 K for all three orientations in increasing field are presented. It can be seen that a sharp change in the field dependence of  $J_c$  occurs at the same field ( $B_A = 1.5$  T) for all three orientations. In figure 5, the critical current density at 4.2 K and 2 K in increasing and decreasing fields is presented for the B-field parallel to the surface of the tape and orthogonal to the direction of macroscopic current flow. Similar hysteresis in  $J_c(B)$  has been found for the other two orientations at 4.2 K. On decreasing the field,  $J_c(B)$  is typically 20-40% higher than in an increasing field. The data in figure 5 demonstrate that the field  $B_A$  has increased from 1.5 T at 4.2 K to 2 T at 2 K. A more detailed description of the behaviour of  $J_c$  above 10 K will be presented elsewhere [10] but it should be pointed out that, apart from the value of the anomaly field  $B_A$ ,  $J_c$  is highly dependent on the sample orientation. This is typical for Bi-2223 tapes [11] indicating their high degree of texture.

We have also completed a preliminary phase composition analysis using x-rays. Data on a Bi-2223 core of a tape made from the same batch of wire as the measured sample show small amounts of 2212 phase and 2201 phase.

#### 5. Discussion

An anomalous increase in  $J_c$  at low magnetic fields at 4.2 K can be found in other published data on Bi-2223 tapes. Gurevich *et al* [3] have measured the  $J_c$  of a tape (with the field perpendicular to the tape surface) at temperatures of 4.2 K and 77 K. In this work, at 4.2 K the anomalous change in  $J_c$  occurred at 1 T. No such behaviour was observed at 77 K.

Critical current measurements at 4.2 K on another Bi-2223 tape from BICC Cables show similar anomalous behaviour with  $B_A = 100$  mT. With this tape,  $J_c(B_A) = 0.13$   $J_c(B = 0)$ . Frost et al [4] have also observed such a value for  $B_A \sim 100$  mT. In such cases however, the abrupt disappearance of the anomalous behaviour above 4.2 K may not have been widely reported because of the experimental difficulty of measuring the transport  $J_c$  of tapes at temperatures just above 4.2 K where critical currents are high. It should be stressed also that not all Bi-2223 tapes measured at 4.2 K have shown this anomaly. Its existence appears to depend on the tape fabrication procedure. The data presented here clearly demonstrate that  $B_A$  is independent of the orientation of the sample with respect to the applied field but is temperature dependent.

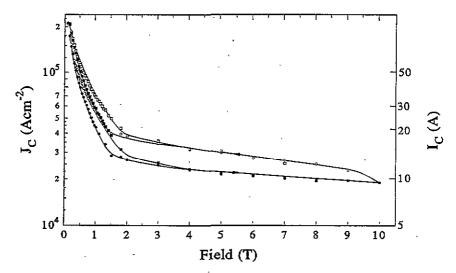


Figure 5. The critical current density at 4.2 K (circles) and 2 K (squares) in increasing and decreasing fields for the *B*-field parallel to the surface of the tape and orthogonal to the direction of macroscopic current flow.

Several theories [5–8] have predicted a phase change for layered structures at low fields from 3D single flux lines to coupled 2D pancake vortices. For magnetically coupled layers Koshelev and Kes [5] suggest that Bi-2223 compounds would have such a phase change between 1 T and 5 T at 4.2 K (depending on the magnitude of  $J_c$ ). From considering Josephson coupled layers [6–8], the flux lines are expected to decompose into pancakes at a field given approximately by  $B_{2D} = \phi_0/s^2\Gamma$ , where s is the spacing between the Cu-O<sub>2</sub> planes and  $\Gamma$  is the anisotropy. Using reasonable values of s = 18 Å and  $\Gamma = 960$  for Bi-2223,  $B_{2D} = 670$  mT. Cubitt et al [9] claim to have observed this 3D to 2D phase transition at 60 mT from SANS measurements on a Bi-2212 crystal. As the theory predicts, they found  $B_{2D}$  to be temperature independent which is not consistent with our measured values of  $B_A$ .

Other predicted phase transitions include changes in collective pinning. Such phase transitions are a consequence of the highly anisotropic structure of the Bi-2223 compound. The position of predicted transition lines in the field/temperature phase diagram are strongly dependent on temperature and/or the component of the magnetic field parallel to the c-axis of the superconductor. The anomalous behaviour in  $J_c$  that we have observed is clearly independent of the orientation of the applied field. For this reason we do not believe that it is due to a phase change in the flux line lattice in the tape.

Although it is clear that Bi-2223 is a granular material, little is understood about the superconducting properties at or near the grain boundaries. The 2223 phase of BiSrCaCuO can coexist in phase equilibrium with the superconducting 2212 and 2201 phases and other non-superconducting phases [12]. Almost all Bi-2223 tapes contain some 2212 and/or 2201 phase and previous work has shown how the amount of secondary phase in a tape can control the value of  $J_c$  [13, 14]. It is very difficult however to determine how the second-phase material affects the local electronic and pinning properties and therefore to understand how it may affect  $J_c$ . The amount of 2212 and 2201 phase found in a fully reacted tape depends sensitively on the composition of the precursor powder, the presence of silver and the heat treatment (temperature, atmosphere and reaction times) [12, 15]. Umezawa et al [13] have investigated the microstructure of Bi-2223 tapes (which were not silver doped) for different heat treatments. Even after very long heat treatments they found a few layers of the lower- $T_c$  Bi-2212 phase at the c-axis twist boundaries. They demonstrated that these layers of Bi-2212 controlled the critical current in the range 75-105 K. No other BiSrCaCuO phases were observed at the boundaries. Däumling et al [14] and Yoshida et al [16] have shown that at 840°C (the temperature at which the BICC tape was fabricated), in the presence of silver, the 2201 phase coexists with the 2223 phase. During postannealing at lower temperatures and slow cooling the 2201 phase is gradually converted to 2212. Both groups conclude that this 2212 phase material plays an important role in determining intergrain connectivity. These results and our x-ray data are consistent with small amounts of 2201 phase remaining after the final heat treatment.

We suggest the following explanation for the anomaly in  $J_c$  below 10 K. In the tape studied here not all of the 2201 phase has been converted to 2212 and a small proportion of 2201 still remains at some of the grain boundaries. When this phase is not superconducting a poor current carrying link exists between these grains. Current flow across these links is only possible by Josephson tunnelling and is low. The current now mostly flows via grains that are connected by different mechanisms (such as links of the 2212 phase). The observed rapid increase in  $J_c$  as the applied field is decreased below  $B_A$ , the independence of  $B_A$  on orientation and the relatively field-independent  $J_c$  behaviour above  $B_A$  is consistent with this description. By linearly extrapolating the measured  $B_A$  to zero, a  $T_c$  of  $\sim$  10 K is found for the intergrain phase. This value is within the range of  $T_c$  expected for the single-layer 2201 phase which has a maximum  $T_c$  of 22 K.

In summary we attribute the marked increase in  $J_c(B)$  below 10 K in low fields to some of the grain boundaries containing a low- $T_c$  superconducting phase. When the grain boundaries are superconducting, there are strong superconducting paths between the grains and there is a marked increase in  $J_c$ . These results clearly demonstrate the important role of intergrain connectivity in obtaining high- $J_c$  Bi-2223 tapes.

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