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HEAVY ION IRRADIATION OF Bi-2223 SILVER -CLAD TAPES FOR SUPERCONDUCTING CURRENT DENSITY ENHANCEMENT

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ABSTRACT

Silver-clad composite tapes of Bi(Pb)SrCaCuO-2223 were irradiated with 1 GeV Au²³⁺ ions perpendicular to the tape plane, creating columnar tracks with 10 nm diameter. Detailed transmission electron microscopy shows continuous columns, but with variations in the track thickness of ± 2 nm. Magnetic hysteresis measurements show substantial enhancements of superconducting current density at most temperatures and fields. Granular and nongranular models for the origin of the magnetic signals are discussed. The results indicate the potential for further performance improvement in high temperature superconducting wire technology, as well as limits in the high-field performance at 77 K.

INTRODUCTION

In a recent paper,¹ the authors have described the effect of heavy ion irradiation on the current density of Bi(Pb)SrCaCuO-2223 ("BSCCO-2223") silver-clad composite tape-shaped wires. In this paper, these results are reviewed, and new data are presented on the transmission electron microscopy of the irradiated wires, showing some deviations from ideal columnar damage. Also, the magnetic measurements of critical current density are discussed in the context of both granular and nongranular models. The conclusions indicate opportunities for improving performance of existing superconducting wire technology but also limits on the possibility of high-field (several Tesla) performance of BSCCO-2223 wires at 77 K.

The presently leading approach to making long-length flexible high-temperature superconducting wires is the oxide-powder-in-tube method with the BSCCO-2223 superconductor in a composite wire configuration with silver or silver alloys.²⁻⁴ Recent progress has pushed the 77 K filament current density in short samples (1 cm) to almost 70,000 A/cm² at 77 K and self-field, with correspondingly higher values at lower temperatures. In longer wires, values of 10,500 A/cm² (measured with a 10⁻¹¹ Ω cm criterion) at 77 K and self field over 300 m have been reached.⁵ While these current densities persist to high fields at temperatures below about 35 K, the field range at 77 K is somewhat limited.⁶ It is generally believed that this field limitation arises from thermal activation of "pancake vortices" in the

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highly anisotropic BSCCO materials.⁷ Since such thermal activation can be reduced in the presence of appropriate defects which constitute vortex pinning centers, many efforts have been made to introduce such centers.

Among the most promising such defects are the narrow columnar amorphous tracks made by the irradiation of heavy ions above a certain threshold energy.⁸ This is because the damage perimeter around the track is sharp and the diameter of order 10 nm is well matched to the coherence length of these superconductors in the *ab* plane. Furthermore, even though these materials are highly anisotropic and the vortices are to a first approximation just pancakes confined to the separate CuO blocks of the structure, there are still Josephson and magnetic couplings between them which make the pancakes tend to arrange into linear stacks or columns. Thus, columnar damage rather than the uncorrelated random damage from lower energy ions or neutrons should constitute the most effective possible pinning center in these materials.

SAMPLE PREPARATION, IRRADIATION AND MICROSCOPIC CHARACTERIZATION

The samples were monofilament silver-clad tape-shaped wires of width 2.5 to 3 mm and total thickness 0.1 to 0.15 mm. The superconducting core thickness was approximately 50 and 75 μm in the two cases, respectively. They were prepared by the now standard oxide-powder-in-tube technique, which has been described in detail elsewhere.^{5,9} The samples were characterized by transport measurements at 77 K, giving filament current densities (1 $\mu\text{V}/\text{cm}$ criterion) of about 15,000 A/cm².

The irradiation has also been described earlier.^{1,10} Briefly, the samples were irradiated with 1.08 GeV Au²³⁺ ions at the TASCC (tandem accelerator superconducting cyclotron) facility at the Chalk River Laboratories of AECL Research, Canada. The irradiation dose was 5.35×10^{10} ions/cm² on the initially 0.1 mm thick sample and 1.7×10^{11} ions/cm² on the initially 0.15 mm thick sample. To reduce attenuation by the silver, the samples were mechanically polished prior to irradiation on one surface. Since each heavy ion creates a damage track, the irradiations can be characterized by a magnetic flux density B_0 corresponding to the magnetic vortex density required to fill all the tracks. Thus, the doses correspond to $B_0 = 1.1$ and 3.5 T respectively.

Fig. 1. Transmission electron micrograph viewed along the irradiation direction in a BSCCO-2223 tape, showing columnar tracks induced by irradiation of 1.08 GeV Au²³⁺ ions. Dark lines are dislocations gliding on the (001) basal plane.

Transmission electron microscopy (TEM) on thin flakes prepared from these samples has been carried out at the Electron Microscopy Center for Materials Research at Argonne National Laboratory. Here we present new and more complete TEM results on the damage tracks inside the BSCCO-2223 superconductor, in particular on the sample irradiated to $B_0 = 1.1$ T. Fig. 1 shows a view approximately along the irradiation direction perpendicular to the plane of the tape. Since the wires are highly textured, this means that the BSCCO-2223 grains also lie predominantly with their ab-planes in the tape plane and that the view looks down approximately along the c-axis of the grains. The tracks appear as circular dots on a grey background. Analysis of the damage cores shows them to be amorphous in structure.

Also visible in the micrograph are numerous dislocations, which lie in the plane of the thin flake specimen. As will be discussed further below, the dislocations are believed to result from mechanical deformation during the TEM sample preparation, which involved coating the surface of the irradiated tape with an acetone soluble plastic film and mechanically removing small thin flakes for direct TEM analysis. Due to the highly textured nature of the tape and ease of deformation on the (001) basal plane in BSCCO-2223, the flakes were always oriented as shown in Fig. 1, and the dislocations lie normal to the c-axis, in the plane of the flake. It is interesting to note that when these dislocations intersect the amorphous track cores, they sometimes become pinned. This is evidenced by the numerous bowing dislocations extending from track to track throughout the microstructure. There is a crude analogy here with the way vortices also become pinned at the tracks.

Also of interest is the evaluation of the depth dependence of the damage character in these samples. Cross-sectional views have shown that for certain heavy ion irradiations of YBaCuO-123, the tracks are discontinuous.^{8,11,12} In the present BSCCO-2223 case, the microstructure of a cross-sectional sample was imaged at different distances from the top or entry surface of the tape to evaluate the damage character along the length of the tracks. Fig. 2 illustrates the track morphologies at three different penetration depths: a) 0.2 μm , b) 5 μm and c) 18 μm .

Several typical features can be seen in Fig. 2a. The tracks are highly aligned with one another but are not aligned parallel to the c-axis of the local grains (note the 1.8 nm c-axis fringes). The c-axis in the local grain is measured to be 16 degrees from the irradiation direction. Since the sample was irradiated approximately perpendicular to the plane of the tape, it seems likely that the misalignment in the figure is simply due to the fact that the texture is not perfect, and some of the grain ab-plane orientations deviate from the tape plane.

Another noticeable effect is the regular rightward displacement by about 10 nm of all tracks above the dark band diagonally crossing the middle of the micrograph. The dark band represents a grain with its ab-orientation rotated from that of the surrounding grains, but with its c-axis in perfect registry. And so, we interpret the track offsets in terms of mechanical deformation caused by a dislocation slip plane at the c-axis twist grain boundary. This deformation must have occurred after irradiation, during TEM sample preparation. While not showing the same area, Fig. 1 images what we believe are this kind of dislocations. It is further interesting in Fig. 2b that a small offset of about 0.3 nm occurs at an intergrowth 2/5 of the distance from the bottom of the figure, while there is no offset at the prominent grain boundary 2/5 of the distance from the top of the figure, where there is a several degree c-axis misalignment. It is perhaps not surprising that ab-dislocation slip should be easier when there is perfect registry of the c-axis, hence less misfit grain-boundary structure.

The fact that some tracks appear to stop part-way through the micrographs of Fig. 2 can be attributed to tracks terminating at the irregular surface of the TEM sample. This has been confirmed by tilting studies. An important conclusion, therefore, is that these tracks are entirely continuous throughout the depth range up to 20 μm accessed in this TEM study. However, calculations based on the threshold energy loss rate¹ indicate that the continuous columnar tracks should be only 29 μm deep and the total projected range only

34 μm deep, while the superconductor thickness in the center of the tape is 50 or 75 μm in the two samples, respectively (see further discussion of the edges below).

A further significant observation concerns the track diameter, which, as evident from Fig. 2, varies along its length from roughly 8 to 11 nm, with an average diameter of 10 nm. Thus we can conclude that the damage is nonideal in two respects: First the columns are not perfect columns but show diameter variations even in the range where they are continuous. Second they do not penetrate through the full sample. It is likely the second problem simply reduces the magnitude of the current density enhancement in proportion to the unirradiated volume of the sample. The first problem should be considered more carefully from a theoretical perspective: how strong an effect could such diameter variations have on thermal activation of pancake vortices?

Fig. 2. Bright-field TEM micrographs of a cross-sectional sample of the irradiated BSCCO-2223 tape (view along tape plane) at a) 0.2 μm , b) 5 μm and c) 18 μm depth from the entry surface. The trajectory of the high energy Au ions lies approximately in the plane of the figures and illustrates the continuous morphology of the track damage.

MAGNETIC CHARACTERIZATION AND INTERPRETATION

Both unirradiated and irradiated samples were measured in a Quantum Design SQUID (superconducting quantum interference device) magnetometer, as described earlier.¹ They were cut to 0.3 cm lengths and had core widths of about 0.23 cm. Examples of the hysteresis loops taken with field applied perpendicular to the sample plane are shown in Fig. 3 comparing unirradiated and irradiated magnetizations (moment per volume) at three different temperatures: 5, 35 and 77 K. According to the Bean critical state model, the current density is proportional to the width of the hysteresis loop. The data indicate a substantial increase in the current density at most temperatures and fields (except for very low fields at 77 K) as well as a substantial shift of the irreversibility line (the boundary where the magnetic loops stay open) to higher temperatures and fields, as shown in Fig. 4. Experiments cutting the samples into 16 smaller pieces indicate that the signals are granular in nature,¹ coming from regions which are less than 500 μm in lateral dimension. This granularity is believed to arise from the polishing procedure used to remove silver from the tape surface for irradiation; earlier studies on similar but unpolished BSCCO-2223 tape samples^{6,14} showed non-granular behavior.

Fig. 3. Isothermal magnetic hysteresis loops of unirradiated and Au-irradiated BSCCO-2223 tape samples at three temperatures (from Ref. 1).

Fig. 4. Irreversibility line (H_{irrev}) of unirradiated and Au-irradiated BSCCO-2223 tapes from DC magnetic measurements, determined from data such as Fig. 3, from the field where the hysteresis loops close to within 5×10^{-5} emu.

An important question in the interpretation of these results is whether the signals represent a flow of supercurrent which is confined to single grains (*intragranular*) or whether it crosses grain boundaries (*intergranular*). This is because the transport critical current density of interest for wire applications must cross grain boundaries (*intergranular* transport), and it would be of considerable practical significance if the above results could be interpreted to imply that this current density increases with heavy ion irradiation. The concern is that irradiation, while adding pinning centers to the bulk of the grains, might actually decrease the currents across the grain boundaries because of damage to the grain boundaries.

Here we extend our earlier discussion¹ of this question. First, the transmission electron microscopy shows good coupling of grain boundaries in these samples. These means that the mechanical polishing has not so damaged the specimens as to limit current flow totally to the intragranular regions of size d_g (= approx. 10 μ m in our case). The cracking from polishing presumably creates regions of an intermediate size d_c . However, it is important to recognize that the magnetic signal in such samples can arise both from intragranular and from intergranular critical currents simultaneously.¹³ Calling these $J_{c,t}$ and $J_{c,g}$ respectively, and assuming approximately equal lateral dimensions and no anisotropy in the *ab*-plane, the Bean model predicts that the net magnetization (in emu/cc) is given by¹³

$$M = (J_{c,t}d_c + J_{c,g}d_g)/30 \quad (1)$$

with J in A/cm and d in cm. Thus, even though d_c may be substantially larger than d_g , the magnetization signal can be dominated by the intragranular contribution $J_{c,g}d_g$ if $J_{c,g}$ is sufficiently large compared to $J_{c,t}$.

In earlier work,^{6,14} Maley et al. and McHenry et al. established that the magnetic signals in good quality uncracked BSCCO tapes (including samples of the type studied here) are nongranular in nature, that is, that the intergranular $J_{c,t}d_c$ term in Eq. 1 dominates. Our hysteresis loop shapes on cracked but unirradiated samples in Fig. 3 are similar to theirs, suggesting that the first term of Eq. 1 still dominates. If irradiation enhances the second term rather than the first, one would expect M not to change until a crossover point as a function of irradiation. Thus a study with more detailed dependence on irradiation dose would distinguish between these two interpretations. Of course, direct transport measurements would also help clarify whether heavy ion irradiations can truly enhance the transport critical current. These are topics for future work.

Further insight into the origin of the magnetic signal comes from considering the mechanism of current density enhancement by irradiation in these polycrystalline materials. The simplest hypothesis is that irradiation creates pinning centers which enhance *intragranular* current density but that it damages grain

boundaries and so reduces the *intergranular* current density. From this perspective, it would be natural to interpret the observed magnetic enhancement as coming from the intragranular $J_{c,g}d_g$ term in Eq. 1.

However we argue here that irradiation can actually enhance *intergranular* current density, particularly in the textured polycrystalline microstructure characteristic of the BSCCO-2223 tapes. This structure has been likened to a brick wall,¹⁵⁻¹⁶ with the bricks representing grains. As shown in Fig. 2, the grains have partially or fully aligned c-axes and randomly oriented ab directions creating c-axis twist boundaries in the overlapping sections of the bricks. Because of the large aspect ratios of the BSCCO grains, it has been proposed that the dominant transport current weaves through these large area boundaries. The observed low-temperature high-field current density plateau has been interpreted in this model¹⁵ in terms of nonuniformities in the Josephson coupling as a function of position inside the c-axis twist boundaries. Irradiation could enhance this nonuniformity and so increase the current density. Furthermore, Bulaevsky et al.⁷ recently calculated the temperature dependence of the critical current density based on the thermal excitation of pancake vortices in the vicinity of the junction. Again, the effect of heavy ion irradiation would be to create pinning centers suppressing such excitations and thus increasing the current density.

In summary, these arguments make plausible the interpretation of the magnetic data in terms of the intergranular (transport) current density. However further work is required to test and confirm this interpretation with direct transport current density measurements.

The pinning created by heavy ion irradiation is near ideal. This is especially true if one considers the recent results of Larbalestier et al.,⁴ who sectioned BSCCO-2223 monofilament tapes of the type we study here and who found that current flow was concentrated at the edges of the tape. These edges are tapered and thin (a typical cross-section is shown in Ref. 2), and so here the heavy ions can fully penetrate the superconducting filament thickness, creating excellent pinning throughout. As suggested by the Bulaevsky et al. pancake-vortex excitation theory,⁷ the pinning should determine the critical-current temperature dependence and hence the irreversibility line of Fig. 4. Especially in the high temperature end where the line falls below several Tesla, increasing the heavy ion dosage is not expected to further enhance the irreversibility line because there are enough tracks to pin every vortex.

This points to another important conclusion: With near-ideal pinning, the irreversibility line we observe in Fig. 4 should be close to the maximum possible for BSCCO-2223 tapes. Unfortunately, at 77 K the irreversibility field (perpendicular to the tape plane) is only about 0.5 T. This implies that moderate-to-high-field (several Tesla) applications working at 77 K may remain out-of-reach for BSCCO-2223 tapes.

This conclusion by no means eliminates the impact of these tapes: they remain attractive for lower-field power transmission applications at 77 K, and they also have good performance in several Tesla below about 35 K.⁶ However, the search for high-field performance at 77 K should probably turn to systems like the recently discovered Hg high temperature superconductors, where first results already show an even higher enhancement of the irreversibility line under irradiation.¹⁷

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