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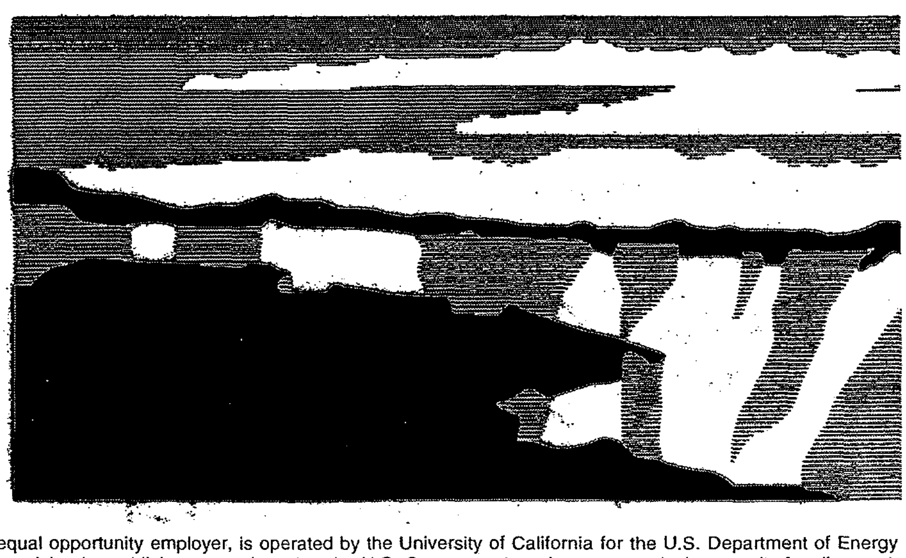

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High Energy Proton Irradiation Induced Pinning Centers in Bi-2212 and Bi-2223 Superconductors

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ABSTRACT

Bi-2212 single crystals and Bi-2223/Ag-sheathed tapes were irradiated with high energy protons. TEM images reveal the production of randomly oriented (splayed) columnar defects with an amorphous core of ~10 nm diameter caused by the fissioning of Bi nuclei. The critical current density J_c and irreversibility line both substantially increased with the proton dose for both crystals and tapes, especially for the magnetic field parallel to the c axis. An irradiated tape had a J_c value ~100 times greater than that of an unirradiated one at 1 T and 75 K.

KEYWORDS: Bi-2223/Ag tape, proton irradiation induced pinning, splayed columnar defects, critical current density

INTRODUCTION

High temperature superconductivity (HTS) has been proposed as a new technology for commercial applications such as magnets, motors, and generators. These devices require that the HTS conductors be available in long lengths, that they have sufficient flexibility to be handled and wound into the device, and that they maintain substantial critical current densities (J_c's) in the presence of a large magnetic field at elevated temperatures. The only HTS materials that can be manufactured in long lengths (1000 m) [1] of strongly linked material (grains) capable of transporting large dissipationless currents are the bismuth-based superconductors $Bi_2Sr_2CaCu_2O_x$ (Bi-2212) and $(Bi,Pb)_2Sr_2Ca_2Cu_3O_y$ (Bi-2223) manufactured by the oxide powder in tube process [2]. Of these two, only Bi-2223, because of its significantly higher superconducting transition temperature (T_c) of 110 K, can be considered for use at liquid nitrogen temperature, 77 K. However, at these temperatures thermally activated magnetic flux motion is severe in the bismuth based superconductors, which are electronically highly anisotropic and nearly two dimensional, and tends to reduce J_c in the presence of a magnetic field [3]. As a result, the useful operating range of Bi-2223 at 77 K is less than 0.5 T.

The introduction of columnar defects by high energy heavy ion irradiation has been shown to be capable of producing large enhancements in the magnetization J_c and in raising the irreversibility line in single crystal Bi-2212 [4]. Both of these factors increase the useful operating field at high temperatures. However, the heavy ions penetrate only 20-30 µm into the HTS material, a distance insufficient to penetrate a typical OPIT tape of thickness 100-200 µm, and thus impractical for generating pinning centers throughout the volume of manufacturable conductor geometries.

Recently, a method using high energy protons to produce columnar defects *in situ* has been developed.[5,6] In this technique, the protons, which have a 0.5 m penetration length in matter, interact with high atomic number elements (Bi and Pb for the Bi-2223 superconductor) causing a fraction of these nuclei to fission into two roughly equal mass fragments with ~200 MeV of energy available. These fragments recoil through the lattice producing randomly oriented (splayed) long columnar defects of 5-10 nm diameter. Previous work reported magnetization measurements on Bi-2212 thick films on Ag foil [5]. We have more recently reported transport critical current measurements on Bi-2223/Ag sheathed tape [6]. It has also been theoretically predicted that splayed columnar defects should be more effective at pinning flux vortices than the parallel columnar defects [7].

In the following sections, the results of high energy proton irradiation experiments on Bi-2212 single crystals and Bi-2223/Ag tapes are discussed. The microscopic structure and orientation of the defects are examined by transmission electron microscopy. The effects on the superconducting properties of the two types of samples analyzed are compared, and finally, some implications for device applications are discussed.

EXPERIMENTAL PROCEDURE

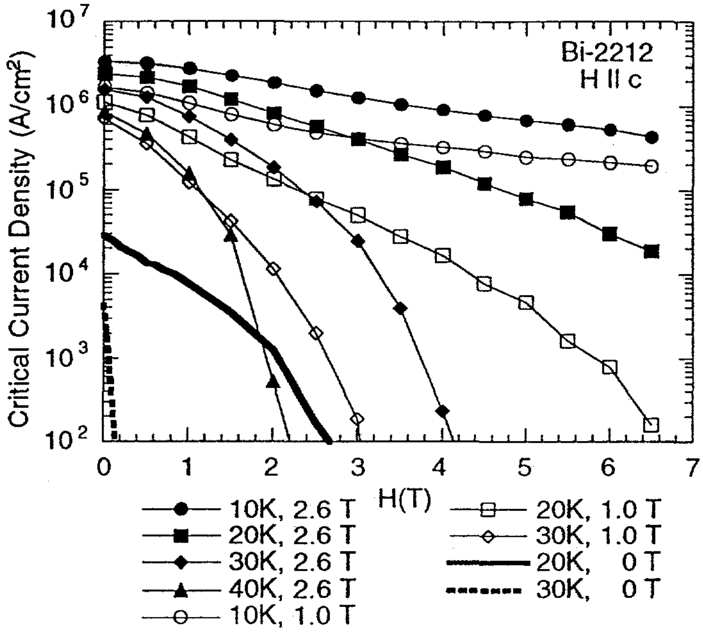
Bi-2212 single crystal samples were flux grown at Los Alamos in air with a 20% excess of Bi_2O_3. Partial melting was done at 990°C followed by rapid ramping to 930°C. The samples were then slow cooled to 840°C over approximately 120 h. Post annealing in air raised the superconducting critical temperature T_c to ~87 K. Mono- and 85-filament Bi-2223/Ag tape samples were manufactured by American Superconductor Corporation (ASC). The tapes were prepared at ASC by a standard OPIT process [8]. Tapes were prepared by packing the powder into cylindrical silver billets, which were deformed into tape using standard deformation techniques, including wire drawing to a hexagonal shape, repacking an 85 filament bundle into a silver tube, wire drawing, and rolling. For monofilament tapes the rebundling step was omitted. The samples were given multiple heat treatments at temperatures between 800 and 830°C in 7.5% oxygen with intermediate deformations consisting of pressing for the monofilament tapes and rolling for the multifilament tapes. The final dimensions of the monofilament tape are 0.1 x 3 mm with a ~35-40 µm thick core and a 20% fill factor. The 85 filament tape has dimensions of 0.25 x 2.5 mm with filament dimensions of 15 µm thick and 120 µm wide and a 28% fill factor.

The samples were irradiated in vacuum at the WNR Facility of the Los Alamos Meson Physics Facility (LAMPF) with 0.8 GeV protons at an average beam current of 1.2 µA. The beam has an axially symmetric Gaussian distribution in the density of incident protons with a full width at half maximum of 1.2 cm. The tape samples were given approximately equal doses, which were determined by measuring the ^{22}Na activation levels on Al foils placed immediately behind the samples. Other irradiated Al foils were used to determine the distribution of the proton beam intensity more precisely.

The number of fissions per unit volume per proton was determined as the product of the proton fluence, the density of Bi nuclei in Bi-2212 (8.89×10^{21} nuclei/cm^3) or Bi-2223 (7.33×10^{21} nuclei/cm^3), and the Bi fission cross section value of 155 mb at 0.8 GeV interpolated from data at 1.0 and 0.6 GeV [9]. Assuming an average track length of 6 µm [5] for each fission, an areal density of tracks was calculated. The track density was then multiplied by the flux quantum ϕ_0 (= 2.07×10^{-11} Tesla-cm^2), to convert it to an equivalent magnetic field B_ϕ, at which the vortex density equals the track density. Bi-2212 crystals were irradiated to calculated track densities of 1.0 and 2.6 T. The crystals were evaluated by magnetization measurements using a Quantum Design Model MPMS. Four voltage contacts were placed along the length of each Bi-2223/Ag tape encompassing three regions of varying defect density because of the spatial variation of the beam intensity. The fluences in these regions were 1.6, 2.9 and 7.6×10^{16} protons/cm^2. For the multifilamentary tape measurements were made only on the region that received the highest fluence. These values correspond to defect densities B_ϕ of 0.2, 0.4 and 1.1 T for the monocore tape and 1.1 T for the multifilament tape. The *I-V* characteristics of the samples were measured with the samples immersed in liquid nitrogen to avoid heating at the contacts. The critical current I_c was determined at an electric field criterion of 1 µV/cm.

RESULTS AND DISCUSSION

Bi-2212 crystals irradiated to nominal defect densities B_ϕ of 0, 1.0, and 2.6 T had measured T_c's of 87, 85.5, and 83 K, respectively, thus indicating a small depression of T_c with proton fluence or defect density. Figure 1 compares the magnetization critical current density J_c, determined from the width of the hysteresis loops using the Bean model, for all of the crystals. Compared to the unirradiated sample, both irradiated crystals show large enhancements of the zero field J_c and of the field at which J_c drops below a small finite value. The 2.6 T sample clearly shows improved performance at all measured values of magnetic field. This is also evident from the irreversibility line, determined from the closing of the magnetic hysteresis loops, shown in Fig. 2. At 2 T, for example, the irreversibility temperature T_{irr} for the unirradiated specimen more than doubles from 18 K to 38 K for the B_ϕ = 1.0 T sample, and increases by another 3 K to reach 41 K for the B_ϕ = 2.6 T specimen. The J_c and irreversibility line of this latter sample are quite comparable to that of a sample with twice the density, B_ϕ = 5 T, of parallel columnar defects. [4]



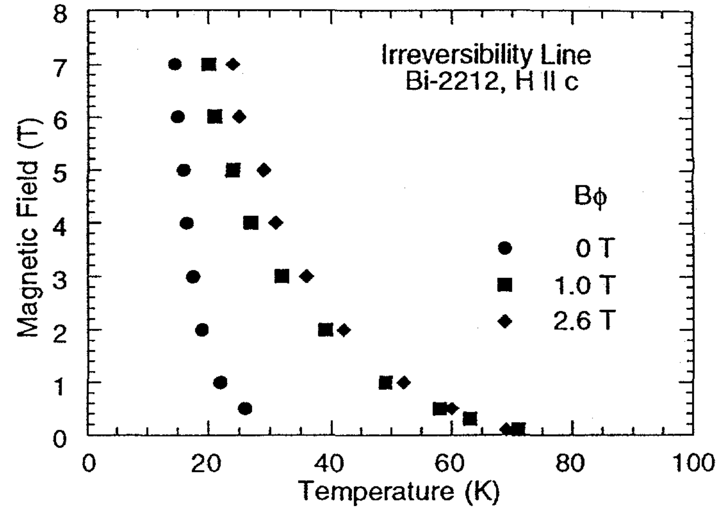


Fig. 1. Magnetization critical current density *vs.* magnetic field for irradiated and unirradiated Bi-2212 crystals. The curves are labeled by measurement temperature and calculated defect density B_ϕ.

Fig. 2. Irreversibility line for unirradiated and irradiated Bi-2212 single crystals. Points are labeled by the calculated defect density B_ϕ. The irreversibility temperature T_{irr} is enhanced approximately 20 K at 2 T.

Figure 3 shows a transmission electron microscope (TEM) image of the splayed columnar defects produced by the fission fragments in the $B_\phi = 1.0$ T crystal. Unirradiated crystals show no defects of this linear nature. The defects are quite long, approximately 0.3 μm for this sample, and believed to be limited in length by the sample thickness, estimated to be less than 1 μm. An analysis of these defects shows that the in-plane (azimuthal with respect to the proton beam) angular distribution of defects is isotropic; there is additional weaker evidence, obtained by analyzing images obtained by tilting the sample at 0, 5, and 10° from the normal, that the distribution with respect to the polar angle is also isotropic. The defect densities determined from micrographs of the irradiated crystals are only about 10% of the calculated values. Some of this discrepancy is the result of the inability to account for defect tracks nearly perpendicular to the plane of the sample since they are not readily visible in images such as Fig. 3, typical of images from which the track density was determined. Other sources of discrepancies could be shorter average track lengths than used in the calculations. This aspect of the analysis continues to be under investigation.

Figure 4 shows a high resolution TEM image of a defect practically parallel to the c axis viewed end-on. The diameter of the defect is about 10 nm, which is similar to the size of the superconducting coherence length ξ, and therefore, nearly optimum for pinning magnetic flux lines. The core of the defect is also amorphous, presumably a result of the large energy deposited by the passage of the fission fragment. Just to the left of the main defect is the remnant of another fission track recrystallized during the ion milling step of the TEM specimen preparation or in response to the electron beam. This spot is apparent because its *ab* lattice orientation is nearly orthogonal to that of the host crystal. Atomic planes and the incommensurate modulation along the *b*-axis (010) direction are also visible in Fig. 4.

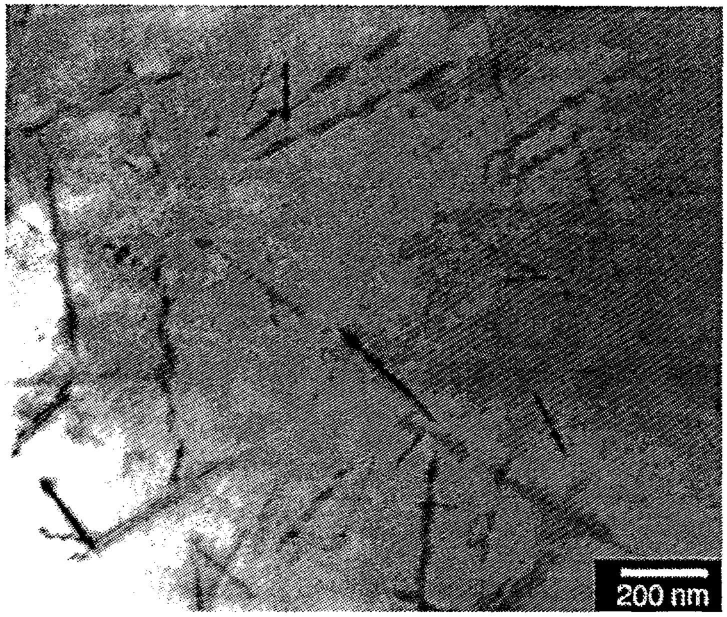


Fig. 3. Transmission electron microscope TEM image of the *ab* plane of crystal with B_ϕ = 1.0 T showing the randomly oriented, splayed columnar defects.

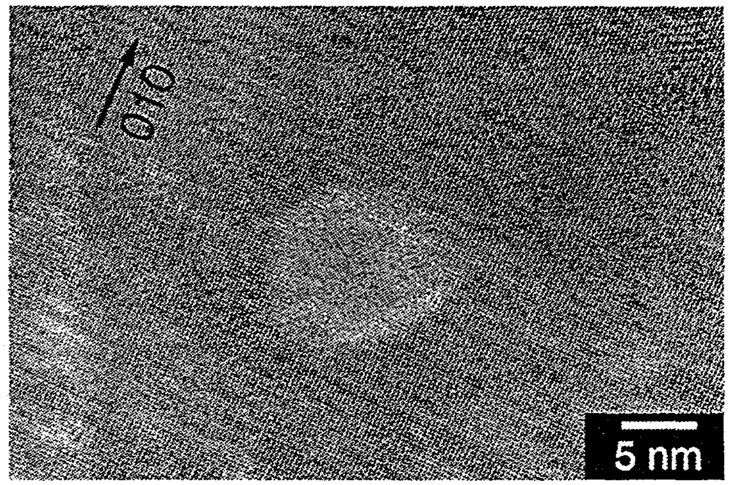


Fig. 4. High resolution TEM image of a columnar defect in the crystal with B_ϕ = 2.6 T. The core of the defect is amorphous. The incommensurate modulation and recrystallized defects are also visible.

The results of critical current measurements on multi- and monofilamentary tapes are shown in Figs. 5 and 6, respectively. Both tapes show a remarkable improvement of the transport critical current density after irradiation. This effect is more notable at the higher fields. For example, at 1 T, Fig. 5 shows that for the multifilamentary tape J_c has increased from an immeasurably low value to ~ 5 x 10^3 A/cm^2 at 75 K.

At 64 K, the fall off in J_c shifts to ~1.5 T. Furthermore, the $J_c(H)$ at 64 K is less field dependent before the fall off, consistent with a larger pinning activation energy at the lower temperature. The zero field values of J_c (J_{co}) after irradiation of the monocore tape were different for the different segments and were totally uncorrelated with the fluences B_ϕ. This may have been caused by accidental mechanical damage during handling of this fragile tape. There was no indication of damage incurred by the more mechanically robust multifilament tape. Figure 6 shows the transport J_c normalized to J_{co} for three different regions of the irradiated monocore tape and for an unirradiated ASC monocore tape at 75 K. Clearly evident is the progressive improvement of the field dependence of J_c with defect density, and the functional form of J_c(H) at the highest fluence is similar to that of the multifilament tape.

Although not shown here, the irreversibility line was determined approximately as the locus of magnetic field/temperature values for which the sample exhibits a voltage of 5 nV/cm for an ac current density of 50 A/cm^2. This criterion is equivalent to the very low current density 1 kA/cm^2 at 1 μV/cm. These data show that both mono- and multifilamentary tapes irradiated to the same defect density B_ϕ of 1 T show equivalent increases of T_{irr} of about 15 K at 2 T compared to the unirradiated samples. This result is slightly smaller, but very comparable to the increases of observed T_{irr} for the Bi-2212 crystals under similar conditions.

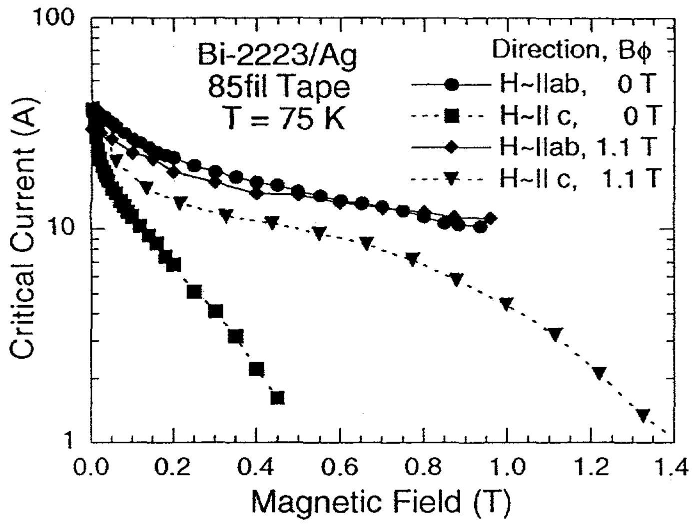


Fig. 5. Critical current *versus* magnetic field for a multifilamentary tape at 75 K.

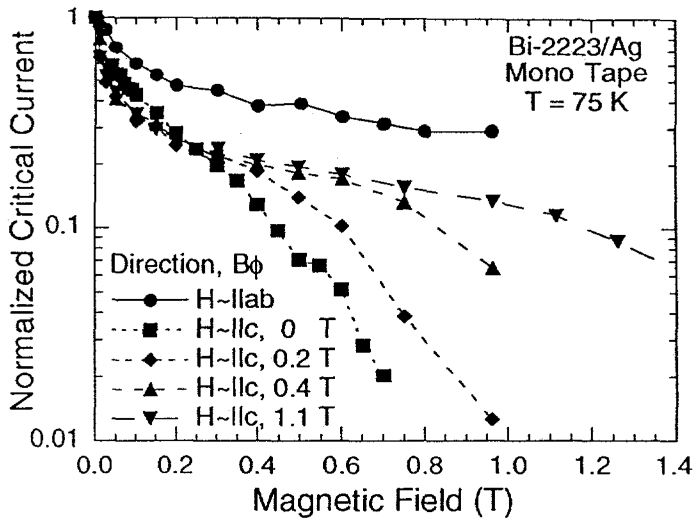


Fig. 6. Normalized critical current *versus* magnetic field for a monofilamentary tape at 75 K.

CONCLUSIONS

We have shown that high energy proton irradiation of Bi-2212 crystals and Bi-2223/Ag tapes results in fissioning of the Bi nuclei producing high energy fission fragments which produce splayed long columnar defects with amorphous cores ~10 nm in diameter. These defects substantially increase the magnetization J_c in the Bi-2212 crystals and shift the irreversibility line to higher temperatures. We have also shown that it is possible to greatly increase the high temperature (64 K and 75 K) transport J_c in the presence of applied fields for industrial state of the art mono- and multifilamentary Bi-2223/Ag tapes. The large penetration depth of high energy protons (~0.5 m) coupled with the improvements in J_c at high fields suggest the possibility of irradiating completed pancake coils to improve their high temperature performance.

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