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On the current transport limitations in Bi-based high temperature superconducting tapes

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The transport critical current densities, J_c, of superconducting $(\mathrm{Bi,Pb})_2\mathrm{Sr}_2\mathrm{Ca}_2\mathrm{Cu}_3\mathrm{O}_x/\mathrm{Ag}$ tapes were measured before and after employing a special radiation technique, which leaves the grain boundary properties largely unaffected. We identify two regions separated by a temperature dependent crossover field $H_{\mathrm{gb}-p}$. In the low field region, J_c is limited by the transport currents across the grain boundaries, which remain unchanged after irradiation. Above $H_{\mathrm{gb}-p}$, J_c is limited by flux pinning. In this field region, the artificial defects optimize flux pinning and enhance J_c. [DOI: 10.1063/1.1378797]

Tremendous efforts have been made over the past few years to improve the current carrying capability of $(\mathrm{Bi,Pb})_2\mathrm{Sr}_2\mathrm{Ca}_2\mathrm{Cu}_3\mathrm{O}_x$ (Bi-2223) high temperature superconducting tapes. As the current transport is entirely determined by the percolative carrier transfer between adjacent grains (''intergrain'' currents), the main thrust of research has been directed towards an improvement of the grain connectivity and, in particular, the grain alignment, since the physical properties of the superconductor are strongly dependent on the relative orientation of current flow or magnetic field with respect to the main crystallographic directions. It turned out, however, that the *in-field* current transport properties remained largely unchanged at elevated temperatures, an observation that is related to the unchanged flux pinning behavior within the grains (''intragrain currents''), which finally limits the current carrying capability of the superconductor in external magnetic fields.

Improvements of the in-field transport behavior by orders of magnitude have so far only been achieved by radiation techniques. Fast neutrons, e.g., were reported to enhance the critical current densities and to shift the irreversibility lines[1] to higher fields quite early on,[2] but the accompanying damage of the intergrain properties, the limited pinning capability[3] of the radiation-induced collision cascades[4] for the magnetic ''pancake''[5] microstructure as well as a certain reduction of the transition temperature due to oxygen disorder at higher neutron fluences showed-up the limitations of the method. An alternative radiation technique consists of bombarding the tape with very high energy (800 MeV) protons, which leads to the fission of the ^{209}Bi nuclei and to the production of two fission products with energies of the order of 100 MeV each.[6] Magnetic measurements of the intragrain currents showed tremendous enhancements of the critical current densities and large shifts of the irreversibility lines for both major orientations of the magnetic field, which are related to the random orientation of these extended defects. It can be expected, however, that the intergranular properties will severely degrade due to damage by these energetic particles. An alternative method, the thermal neutron induced fission of uranium admixtures to the material,[7] avoids the latter problem, takes advantage of the introduction of these extended defects and, in addition, allows us to identify from transport measurements those regions in the (H,T) plane that are dominated by the grain boundary properties and those that are dominated by flux pinning.

Uranium[8,7] (and more recently, ''pure'' ^{235}U),[9] is added in the form of uranium oxide powder to the starting powders. As a consequence of thermal neutron irradiation, fission of the ^{235}U nuclei is induced resulting in the production of two fission products (e.g., ^{147}La and ^{87}Br) with a total kinetic energy of 162 MeV, propagating from the U nucleus in opposite directions and thus creating a (discontinuous) amorphous track with a diameter of approximately 10 nm and a total length of up to 10 μm. The orientation of the track is completely random. The concentration of these defects depends on the uranium content and the thermal neutron fluence. Since only thermal neutrons are needed for this process, the irradiation can be done in such a way that the contribution of higher energy neutrons is very small. As a consequence, radiation damage of the grains or the intergrain properties by nuclear displacements is very small and the contribution of fast neutron induced collision cascades to flux pinning is completely negligible.

The superconducting tapes were produced by the standard powder-in-tube technique,[10,11] but small amounts of uranium oxide (containing highly enriched ^{235}U, $\sim$96%) were added to the precursor powders prior to processing. The processing details described in Refs. 10 and 11 remained unchanged. Monocore tapes in a standard silver sheath (filling factor: 20%–30%; superconducting cross section $1.0-1.8\times10^{-7}$ m^2) and containing four different concentrations of ^{235}U (0.15, 0.4, 0.6, and 1.0 wt %) were prepared

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and their properties in the unirradiated state compared with undoped samples. All of the tapes showed self-field critical current densities of about 1.5×10^8 Am^{-2} at 77 K, but the in-field transport properties degraded slightly (by about 12% and 25%, $H\|a,b$, 77 K) at the highest two U concentrations.[12] Transmission electron microscopy images[13] confirmed very clean grain boundary structures. Pieces of 5 cm length and with a width of 3 mm (overall tape thickness: 150 μm) were cut from the tapes for the transport current measurements, whereas small sections (3×3 mm^2) were used for the magnetic measurements[14,15] in a superconducting quantum interference device.

The transport properties of the samples in the superconducting state were determined by dc transport measurements in two magnet systems. In the first device (for fields up to 6 T), the samples could be rotated in the horizontal field of a split-coil magnet to assess the angular dependence of J_c (1 μV cm^{-1} criterion) at temperatures between 30 and 77 K. In the other device, the measurements could be taken up to 15 T, but only for $H\|c$ and $H\|a,b$. The irreversibility lines were measured resistively (transport current: 30 mA, 1 μV cm^{-1}, corresponding to a J_c criterion of 3×10^5 Am^{-2}).

The irradiation of the materials was made in the TRIGA reactor Vienna[16] in a specially designed irradiation position with a highly thermalized neutron spectrum (thermal neutron flux density: 2.7×10^{15} m^{-2} s^{-1}, fast neutron flux density: 5.3×10^{14} m^{-2} s^{-1}). The temperature during irradiation was below 80 °C, and the samples were kept under ambient atmosphere. The samples were irradiated sequentially to thermal neutron fluences between 5×10^{18} and 6×10^{19} m^{-2}, corresponding to fission track densities between 3×10^{19} and 3×10^{20} m^{-3}, depending on the uranium content. Some care had to be taken after the irradiation because of the induced radioactivity of the silver sheath (note that the radioactivity of the superconductor and of the fission products typically only amounts to ~0.2% and 0.9% of the silver activity, respectively).

We observe enhancements of J_c at 77 K by factors of the order of 100 for fields oriented perpendicular to the tape surface ($H\|c$), and by factors of the order of 10 for $H\|a,b$,[17] in particular at intermediate and high magnetic fields. This anisotropic response to a certain spatial distribution of extended defects can be assessed more accurately by measuring the angular dependence of J_c at fixed temperature and magnetic field. The results demonstrate that the original J_c anisotropy, i.e., the ratio $J_c(H\|a,b)/J_c(H\|c)$, is strongly reduced, e.g., from 40 in the unirradiated state to 4 (77 K, 0.5 T), mainly because flux pinning for $H\|c$ is enhanced in such a drastic way. This strong pinning character of the tracks also manifests itself in the irreversibility lines. We note in particular that the irreversibility fields move up systematically with the density of tracks for both major orientations of the field and reach, e.g., at 77 K, ~2 T for $H\|c$ and very high fields for $H\|a,b$ (~ 12 T, extrapolated). This smooth variation also emphasizes the fact that the intrinsic superconducting properties, such as the transition temperature T_c, remain basically unaffected by the irradiation procedure, since the percentage of high energy neutrons and their effect on the oxygen positions are negligibly small (T_c varies from 105 K in the unirradiated state to 103 K at the highest fluence).

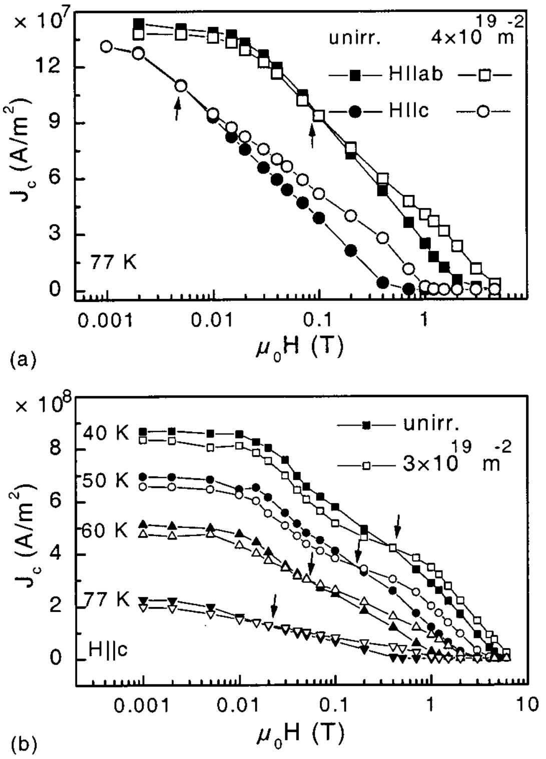

FIG. 1. J_c of a sample containing 0.15 wt % ^{235}U (a) and of a sample containing 0.4 wt % ^{235}U (b) before (full symbols) and after irradiation (open symbols).

The results presented so far and pertaining to the high temperature regime can be readily discussed in terms of flux pinning by extended defects. Although most of the published work refers to heavy ion irradiated single crystals (for an overview, cf. Ref. 18), the enhanced flux pinning action of these defects, in particular for $H\|c$, can be safely related to the improved confinement of the flux lines or pancakes at the defect location. The role of a certain limited splay of the tracks, introduced by irradiating the single crystals under certain angles with respect to the c axis, was discussed in terms of vortex entanglement[19,20] and the geometrical orientation of the defects with respect to the vortex direction.[21] The latter argument applies directly to the "full" splay prevailing in samples with fission-induced tracks[6,22,23] and confirms the role of strong core pinning under all orientations of the magnetic field. As a consequence, the critical current densities are enhanced in all grains, particularly also in those that are not fully aligned, resulting in the overall improvement of the transport J_c's. The newly established defect structure is, of course, most effective, whenever the pre-existing pinning landscape is weak, as in Bi-2223 for $H\|c$.

A more detailed inspection of the J_c–B dependence, particularly at low fields and at lower temperatures, allows us to further elucidate the interplay between inter- and intragranular current flow, as shown in Figs. 1(a) and 1(b). They present a plot of the J_c–B data on a semilogarithmic scale, thus emphasizing the low field regime. It turns out that the pre- and the postirradiation data are completely identical (or just slightly different) in the mT field regime, and that the enhancements, discussed earlier, set in (arrowed) at a certain magnetic field that clearly depends on the field orientation. At fixed field orientation, but at lower temperatures, these

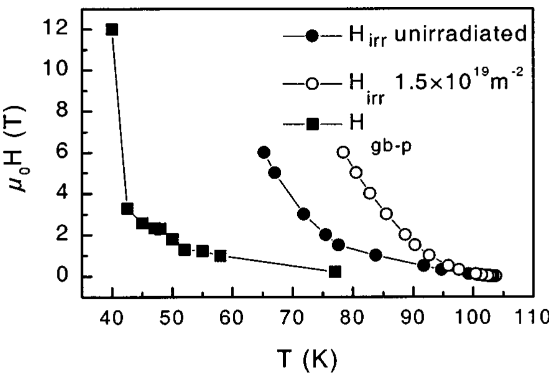

FIG. 2. Crossover field H_{gb-p} and irreversibility lines for a sample containing 0.6 wt % ^{235}U for $H\|ab$.

"crossover" fields are found to shift to the right, i.e., flux pinning becomes dominating at higher magnetic fields, when the temperature is lowered. Since the small decrease of J_c in the low field range is readily attributed to residual damage of the grain boundary structure, which leads to an increase of the normal state resistivity[24] and, consequently, to a decrease of the percolative coupling strength,[5] these results indicate that the current transport in the low field regime is clearly and fully dominated by the intergrain properties of the superconductor and that additional strong pinning defects within the grains are irrelevant in this (temperature dependent) field range. A summary of results on the crossover fields H_{gb-p} from grain boundary to flux pinning dominated current transport is presented in Fig. 2. We observe a smooth temperature variation of H_{gb-p} at high temperatures and a sudden increase at 40 K for $H\|a,b$ and at 20 K for $H\|c$ (not shown). The same behavior was found in all tapes with different uranium content. Although we have obviously no means of deciding about the transport mechanisms below these temperatures, it is reasonable to assume that the as-grown (preirradiation) defects exert sufficiently strong pinning forces at these temperatures to override the contribution of the extended defects of comparatively low density.

Additional evidence for this analysis is provided by the magnetic measurements made on the tapes. According to Müller *et al.*,[14] a shoulder (or a peak in its derivative)[15] should be observed when plotting the remnant magnetic moments as a function of the maximum applied field prior to field reversal, when the intergranular currents become saturated. However, the remnant moments of the tapes in the *unirradiated* state did not provide evidence of distinguishable differences between inter- and intragranular current flow at 5 K, thus confirming the excellent connectivity between the grains. After irradiation of a highly doped tape (1 wt % ^{235}U) to a fluence of 6×10^{18} m^{-2} we find a decrease in the intergranular peak and the appearance of a significant intragranular peak, i.e., the difference between these two currents became larger due to damage of the grain boundary structure on the one hand and due to improved flux pinning on the other hand.

In summary, we have shown that uranium doping of Bi-2223 tapes and subsequent thermal neutron irradiation, resulting in the formation of randomly oriented amorphous fission tracks, lead to substantial improvements of the current carrying capability of this material at elevated temperatures. Order of magnitude enhancements of the critical transport current densities, reductions of the J_c anisotropy as well as very significant shifts of the irreversibility lines (up to 2 T, $H\|c$) were found for optimized defect configurations. Because of the fact that thermal neutrons do not produce pinning centers and that the contribution of higher energy neutrons can be kept very small, this radiation technique leaves the superconductor and, in particular, the grain boundary structure largely unaffected, but introduces extended defects into the grains starting from atomically dispersed locations. As a consequence, regions in the $H-T$ plane could be identified, where enhanced pinning does not play a role, i.e., the grain boundary structure dominated regime, and others where flux pinning is essential. This could help devise strategies for improving this superconductor and pave the way for more widely spread applications of this material.

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