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Susanne Tönies, Harald W. Weber, Yuan C. Guo, Shi X. Dou, Ravi Sawh, Roy Weinstein

Abstract— **Uranium doped BiSCCO 2223 tapes were irradiated by thermal neutrons. The resulting fission-induced defects improve flux pinning and shift the irreversibility line to higher fields. Significant enhancements of the transport critical current density as well as a reduction of the J_c-anisotropy are found for the irradiated samples. Furthermore, inter- and intragranular critical current densities were determined from the remanent magnetic moments by SQUID magnetometry.**

Keywords— **BiSCCO Tapes, Flux Pinning, Neutron Irradiation, Transport Measurements.**

I. Introduction

THE critical current densities J_c in $Bi_2Sr_2Ca_2Cu_3O_x$ (BiSCCO) tapes still need to be improved and the mechanisms limiting them to be explored. In view of the structure of BiSCCO tapes, which consist of small platelet-like grains [1], [2] with a diameter of $\sim$ 20 μm and a height of $\sim$ 1 μm, different limiting factors for the critical current density can be expected, i.e. the connectivity between the grains (intergranular critical currents) or the pinning within the grains (intragranular critical currents) can limit the critical current density.

Radiation-induced defects improve the pinning properties within the grains. Therefore, we can distinguish between regions in the (H,T)-plane, where the critical current density is limited by the grain boundaries, and regions, where J_c is limited by pinning within the grains.

II. Samples

BiSCCO 2223 tapes were produced by the powder in tube process [3], [4], but small amounts of uranium oxide were added to the precursor powders prior to processing [5]. Samples with four different uranium concentrations were produced (0.15 wt%, 0.4 wt%, 0.6 wt% and 1 wt%) and compared to undoped samples. TEM-pictures show that the grain boundaries are very clean.

For transport measurements, samples with 5 cm length and a width of 3 mm were used. For SQUID-measurements, the samples were cut to pieces of 3 mm· 3mm. With a filling factor of 20-30 %, the superconducting cross section of the samples is 1 to $1.8 \cdot 10^{-7}$ m^2.

III. Methods

A. Measurements

The superconducting properties of the samples were determined by dc-transport measurements in fields up to 6 T at temperatures between 30 K and 77 K. The standard 1 μV/cm criterion was used to assess the critical current density. By rotating the sample in the horizontal field of a split-coil magnet, the angular dependence of the critical current density was measured. The irreversibility line was measured resistively at a transport current of 30 mA. A criterion of 1 μV/cm was used to determine the irreversibility fields (J_c criterion: $3 \cdot 10^5$ Am^{-2}).

To determine the inter- and intragranular critical currents, the samples were measured in a MPMS SQUID in fields up to 1 T. Magnetisation loops with ascending maximum applied fields were measured. The inter- and intragranular critical currents can be calculated from the remanent magnetic moments [2]. This moment changes, when the network of grain boundaries becomes fully penetrated and the field starts to penetrate into the grains, because intragranular currents will also contribute to the remanent moment. This leads to a "shoulder" in the remanent magnetic moment versus maximum applied fields curve, which is better visible in the logarithmic derivative of this curve [6], where two peaks occur, the intergranular and the intragranular peak (at higher fields). The area under the peaks is proportional to the critical current densities. Furthermore, the grain sizes can be calculated from the slope of the reverse legs of the magnetisation loops [7].

B. Irradiation

The samples were irradiated in the TRIGA reactor in Vienna at a reactor power of 250 kW using a special irradiation position outside the graphite reflector. This irradiation position has a low total flux density and a highly thermal spectrum (thermal flux density $2.7 \cdot 10^{15}$ m^{-2}s^{-1}, fast flux density $5.3 \cdot 10^{14}$ m^{-2}s^{-1}). The temperature during the irradiation is below 80°C. From the fission cross section of ^{235}U, the density of uranium atoms in the sample and the thermal neutron fluence the density of the defects was calculated. All quoted fluences are thermal neutron fluences.

Thermal neutron irradiation leads to fission of the ^{235}U nuclei. The two fission products have kinetic energies of about 80 and 100 MeV and move in opposite directions. On their way through the sample the fission products trans-

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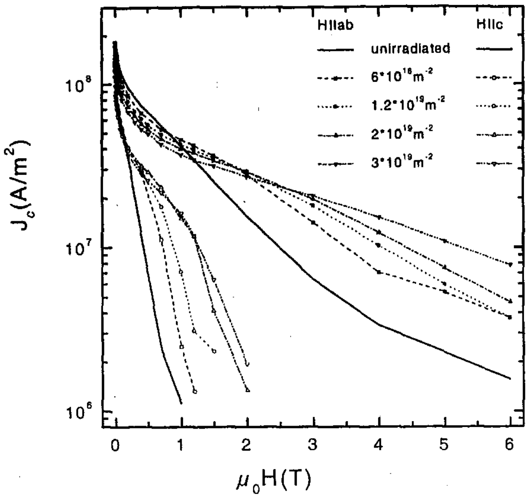

Fig. 1. Critical current densities at 77 K of a sample containing 1 wt% ^{235}U prior to irradiation and after four irradiation steps.

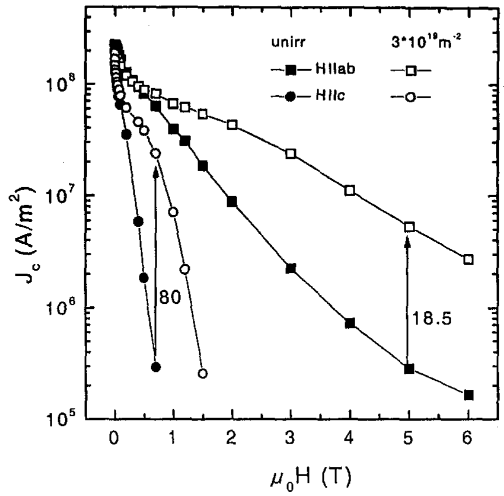

Fig. 2. Critical current densities at 77 K before (full symbols) and after (open symbols) irradiation (0.4 wt% ^{235}U).

fer their energy to the electronic system, which results in local melting of the lattice. The resulting defects, fission tracks, have a length of ~ 5 μm each and a width of a few nanometer [5]. They are randomly oriented in the sample and consist of amorphous material. Their shape is not a homogeneous cylinder [8], as in the case of GeV ion-induced columnar defects, but the cross section varies along its length because of the rather low energy of the fission products [9]. These defects are optimally suited for flux pinning.

Fast neutron irradiation induces spherical cascade defects with a diameter of 3 nm, surrounded by a strain field of the same size [10]. They do not seem to be as effective as pinning centers in two dimensional superconductors, since they are too small to pin more than two pancakes [11].

IV. Results

A. Critical currents

Prior to irradiation the samples have critical current densities of about $1.6 \cdot 10^8$ A/m^2 at 77 K in zero field. The unirradiated samples show only a small influence of ^{235}U doping on their superconducting properties. Samples with less than 0.6 wt% ^{235}U have the same critical current density as undoped samples. Samples with higher doping levels have an up to 25 % reduced critical current density compared with samples with a lower uranium content. No influence of the uranium content is found on the critical temperature and the irreversibility line, which indicates that the uranium atoms do not move to positions in the BiSCCO lattice. The uranium forms, together with other elements, small clusters. After irradiation of samples with different amounts of uranium to the same defect density of $6 \cdot 10^{19}$ m^{-3}, those with higher amounts of uranium show smaller enhancements of the critical current density. This is probably due to some clustering of uranium in the samples. The uranium atoms and, therefore, the defects are then concentrated in some areas and not as effective as homogeneously distributed defects. Thus the effective defect density does not depend linearly on the fluence. After further irradiation we find large enhancements also for samples with higher doping levels. Uranium doped tapes were irradiated up to a maximum defect density of $3 \cdot 10^{20}$ m^{-3}. Samples without uranium do not show any influence of irradiation to this low fluence on any of their superconducting properties.

The largest enhancements of the critical current density were found for a sample with 0.4 wt% ^{235}U. The critical current densities before and after irradiation in both field directions are shown in Fig. 2. For H||c (circles) an enhancement by up to a factor of 80 was found at a field of 0.7 T, and for H||a,b (squares) the enhancement factor is 18 at 5 T. The sample was irradiated to a thermal fluence of $3 \cdot 10^{19}$ m^{-2}, which corresponds to a defect density of $1.2 \cdot 10^{20}$ m^{-3}.

The irreversibility lines shift to very high fields after irradiation due to improved flux pinning. Values of 1.8 T were reached for H||c at 77 K. At this temperature the irreversibility field for H||a,b was above 6 T. This field was reached at 83 K. (Fig. 3). The critical temperature of the unirradiated tapes was ~ 106 K and it was reduced by only 0.2 K after irradiation.

After irradiation we find large enhancements of the critical current density only at higher fields. At low fields the critical current density is limited first by the self field and then by the currents across the grain boundaries, the intergranular currents, which cannot be improved by irradiation. In this low field regime, the critical current density stays the same after irradiation for samples with low amounts of uranium (0.15 wt%) or shifts down due to damage to the grain boundaries in samples with more uranium. At higher fields pinning within the grains limits the transport critical current density, which is improved significantly by the irradiation. The position of the cross-over from the low field to the high field regime depends on the temper-

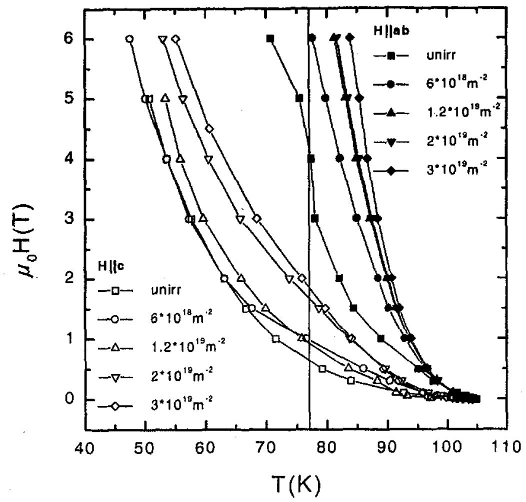

Fig. 3. Irreversibility line of a sample containing 1 wt% ^{235}U prior to irradiation and after four irradiation steps.

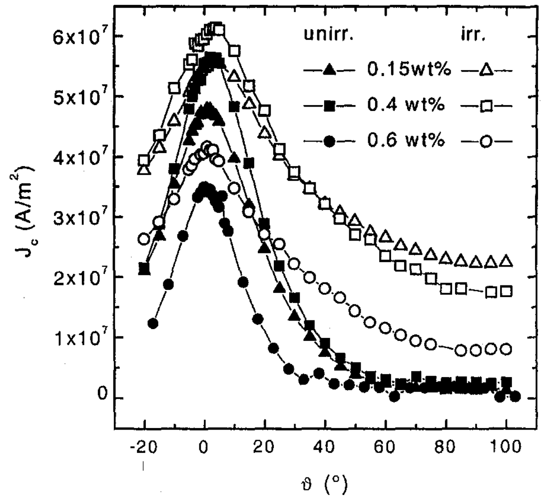

Fig. 4. Critical current densities versus angle between the ab-planes and the field direction before (full symbols) and after (open symbols) irradiation.

ature, the direction of the applied field and also on the neutron fluence.

Some samples were irradiated sequentially to find the optimal fluence. A sample with 0.6 wt% ^{235}U was irradiated first to a fluence of $1.5 \cdot 10^{19}$ m^{-2}. The critical current density was enhanced by a factor of 7 for H||a,b at 5 T and a factor of 6 at 1 T for H||c at 77 K. The irreversibility fields were doubled for both field orientations after this irradiation. Then the sample was irradiated to a total fluence of $2 \cdot 10^{19}$ m^{-2}. The enhancements of the critical current densities were higher (a factor of 13 for H||a,b at 5 T and a factor of 8 at 1 T for H||c) and also the irreversibility line was shifted up. After the third irradiation to a total fluence of $3.5 \cdot 10^{19}$ m^{-2}, J_c was enhanced by a factor of 23 for H||a,b and 3 T and a factor of 11 at 1 T for H||c. The irreversibility line shifted down after the third irradiation to the values after the second irradiation, which is attributed to an excess number of defects compared to the flux line density. The critical current densities also start to decrease after irradiation to higher fluences. The decrease of J_c starts at low fields: e.g. J_c was enhanced by 20% at 1.2 T for H||a,b and 77 K after an irradiation to $6 \cdot 10^{18}$ m^{-2} and went down to the values of the unirradiated sample after a total fluence of $3 \cdot 10^{19}$ m^{-2}. But at high fields, J_c was enhanced by a factor of 20 at the high fluence. The enhancements of the critical current densities are lower at lower temperatures.

Apart from the strong field dependence, also the J_c-anisotropy of BiSCCO tapes limits their application in coils, where the field is always parallel to the ab-planes in the centre, but partly parallel to the c-axis at the ends of the coil due to stray fields. The J_c-anisotropy decreases with decreasing temperature, has a minimum at $\sim$ 40 K and goes up again at lower temperatures. It is reduced by irradiation by an order of magnitude at 77 K and 0.5 T (from 40 to 4). The angular dependence of J_c is plotted for samples with different amounts of uranium at 77 K and 0.5 T before (full symbols) and after (open symbols) irradiation in Fig. 4.

B. *Intergranular and intragranular critical currents*

The irradiation effects on the intergranular and intragranular critical currents were qualitatively deduced from transport measurements, but can be determined more accurately from SQUID measurements [2]. Samples with different amounts of uranium were investigated before and after irradiation. The inter- and the intragranular critical currents are quite similar in all samples prior to irradiation, i.e. a shoulder in the remanent magnetic moment versus maximum applied field curve does not occur. The two currents cannot be distinguished, because the connectivity between the grains is very good and only one peak appears in the logarithmic derivative (open symbols in Fig. 5). The influence of irradiation depends on the uranium content of the samples. Samples with only small amounts of uranium (0.15 wt%) do not show any influence of irradiation. After irradiation the curve still does not show a shoulder and the two current densities still cannot be distinguished. This effect is also visible in the low field region in transport measurements, where the current is limited by the intergranular currents. For samples with 0.15 wt% ^{235}U the critical current density in this field region stays the same after irradiation. In samples with higher amounts of uranium (see Fig. 5) the difference between the two critical currents becomes larger due to irradiation, even at the same defect density, and the inter- and the intragranular critical currents can be distinguished. The intergranular peak becomes smaller due to the damage of the grain boundaries and the intragranular peak is higher because of improved pinning. Note that the critical current density of the transport measurements was reduced in the low field region in these samples after irradiation. The intragranular critical current density is determined to be $1.5 \cdot 10^{10}$A/m^2 at 5 K

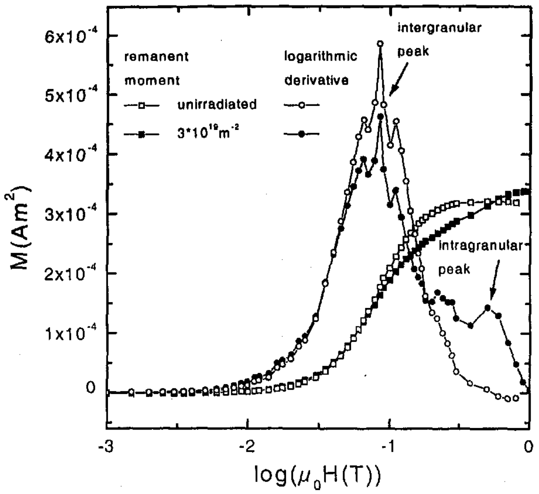

Fig. 5. Remanent magnetic moments at 5 K versus maximum applied field before and after irradiation (squares) and the logarithmic derivatives (circles).

and zero field. The grain sizes are $\sim 25\mu m$ before and after irradiation for all samples, i.e. the grains themselves were not damaged.

V. Conclusions

Thermal neutron irradiation of uranium doped BiSCCO tapes is demonstrated to be an excellent method for enhancing the critical current density at high fields and high temperatures. We find J_c enhancements by factors of 80 for H||c (77 K and 0.7 T) and by factors of 16 at 5 T for H||a,b. The J_c-anisotropy is reduced by an order of magnitude at 77 K and 0.5 T. The irreversibility field at 77 K was shifted to 1.8 T for H||c. The critical temperature was reduced by only 0.2 K.

Furthermore, we can distinguish between the regions in the (H,T)-plane, where the critical current density is limited by the grain boundaries and those, where J_c is limited by pinning within the grains. In the grain boundary dominated low field region, the critical current density remains unchanged after irradiation, whereas very significant enhancements of J_c occur in the high field region.

The intergranular critical current density, which is reduced by irradiation due to damage of the grain boundaries, and the intragranular critical current density, which is enhanced due to improved pinning after irradiation, were evaluated separately from SQUID measurements.

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