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Enhanced critical magnetization currents due to fast neutron irradiation in single-crystal $YBa_2Cu_3O_{7-\delta}$

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Magnetization measurements were performed on single crystals of $YBa_2Cu_3O_{7-\delta}$ to study the effect of fast neutron irradiation on flux pinning and the critical magnetization current. Neutron irradiation up to a fluence of $8.16\times10^{17}\,n/\mathrm{cm}^2$ systematically enhanced the magnitude and reduced the anisotropy of the critical magnetization current compared to unirradiated samples. The largest increases in the critical magnetization current occurred at 77 K. There was a slight linear decrease in the transition temperature with neutron fluence.

A major problem in developing applications of the high-T_c superconductors $La_{2-x}M_xCuO_4$(M = Ca, Sr, Ba) and $YBa_2Cu_3O_{7-\delta}$ is the low superconducting critical current found in polycrystalline samples of these materials as they are commonly prepared.[1-3] The low critical currents are often attributed to the granular nature of the sintered polycrystalline samples. To make full use of the high transition temperature available in these materials, applications must be carried out near liquid-nitrogen temperatures. At these temperatures, the critical current in bulk polycrystalline and single-crystal samples has been found to be lower by two orders of magnitude than its value at liquid-helium temperatures.[1,2,4-6] In single-crystal samples the critical current has been found to be highly anisotropic with the a-b plane supporting currents a factor of 10 greater than those in the c direction.[4,5,7] For applications, it is desirable to achieve isotropic critical currents of the order of 10^6 A/cm^2 at liquid-nitrogen temperatures. To this end we have investigated the effect of fast neutron irradiation on single crystals of $YBa_2Cu_3O_{7-\delta}$ where the intrinsic behavior, uncomplicated by the question of granularity, can be studied. We find a systematic increase in the critical magnetization current with increasing neutron fluence accompanied by a slight decrease in T_c. The anisotropy usually found in single-crystal samples is reduced.

The single-crystal samples used in this experiment were prepared by the same technique[8] as those used for our previous study of the anisotropy of the critical magnetization current.[4] The crystals ranged in mass from 175–497 μg with typical dimensions of 0.4 mm in the a-b plane and 0.2 mm along the c direction. The c direction was easily recognized with optical microscopy and confirmed by x-ray diffraction. The transition temperature of these crystals as determined by shielding-effect measurements described below is above 91 K without oxygen annealing. The four crystals used in this study were not of equal quality. Although their transition temperatures were all above 91 K before irradiation, their shielding curves showed varying degrees of structure with decreasing temperatures. Typically, the shielding curve showed a sharp superconducting transition at about 91.5 K followed by a shoulder at approximately 75 K and a second decrease at around 60 K. This structure is present for both irradiated and unirradiated samples. Typical transition curves for irradiated samples are shown in Fig. 1.

The neutron irradiation was done at the H2 position of the Intense Pulse Neutron Source (IPNS) at Argonne National Laboratory. Three crystals were irradiated at 30 °C with fast neutrons ($E > 0.1$ MeV) to fluences of 1.80×10^{17}, 2.98×10^{17}, and $8.16\times10^{17}\,n/\mathrm{cm}^2$. The superconducting transition temperatures were determined from the temperature dependence of the magnetic shielding curve measured in a special superconducting quantum interference device (SQUID) magnetometer designed to operate in fields less than 100 Oe where data can be taken continuously as a function of temperature and field. The

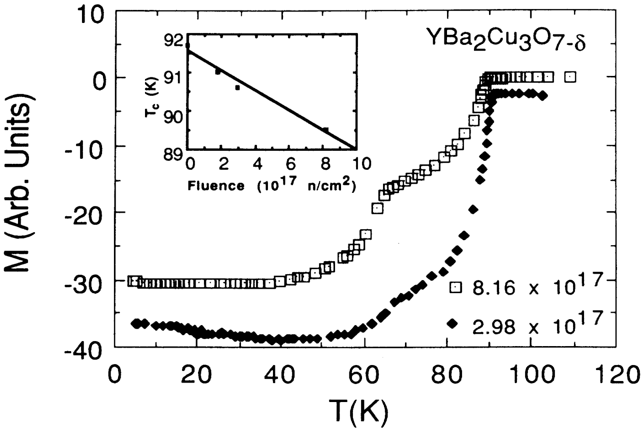

FIG. 1. Temperature-dependent shielding curves for samples irradiated with 2.98×10^{17} and $8.16\times10^{17}\,n/\mathrm{cm}^2$. The superconducting transition temperature falls linearly with neutron fluence as shown in the inset.

transition curves shown in Fig. 1 were obtained by cooling the sample in zero field to 4.2 K, applying a field of 20–50 Oe, and measuring the magnetization with increasing temperature. The field dependence of the magnetization of each of the three samples were taken with a commercial SQUID magnetometer at 6, 45, and 77 K for fields along the *c* direction, and at 6 K for fields along the *a* direction. For all directions and temperatures, magnetizations were taken up to 1 T. The data on irradiated samples were compared with those of an unirradiated sample reported earlier.[4]

Magnetization curves for the crystal with the highest neutron fluence are shown in Figs. 2 and 3. Due to the stabilization requirements of the SQUID magnetometer, data at each field were taken 15 min after the field change. This time interval prevented any influence on our measurements of flux creep as reported in Ref. 9. At 6 K, for the field along the *c* direction, the magnetization curve continues to increase at 1 T even though H_{c1} is less than 0.1 T. The return magnetization curve is nearly straight with a slope approximately equal to the initial magnetization curve indicating that little flux leaves the sample as the field is decreased. At 1 T, 71% of the applied flux is shielded from the sample. Of the flux that has entered at 1 T, 67% remains trapped within the sample at zero field. In contrast, the magnetization of the unirradiated sample reported earlier[4] reached a maximum at 0.9 T. Furthermore, the unirradiated sample shielded 65% of the applied flux at 1 T and retained, at zero field, 55% of the flux which entered at 1 T. Although higher field magnetization data are required to assess the full extent of the improvement in the irradiated sample, it is clear that neutron irradiation has increased the flux pinning and raised the critical magnetization current at 6 K.

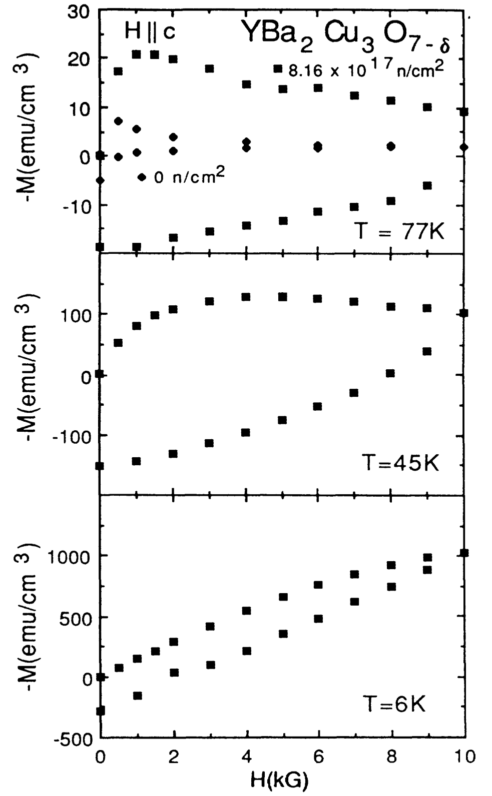

FIG. 2. The magnetization curves for the sample irradiated with 8.16×10^{17} n/cm^2 for fields along the *c* direction. At 77 K the magnetization for the unirradiated sample is shown for comparison.

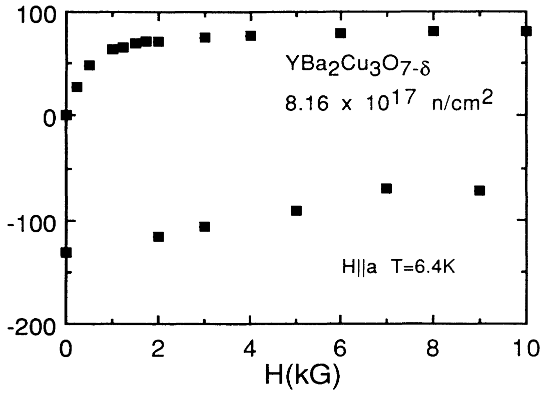

FIG. 3. The magnetization curve for the sample irradiated with 8.16×10^{17} n/cm^2 for fields along the *a* direction.

At 45 and 77 K, the magnetization for the field along the *c* direction reaches maxima at 0.3 and 0.1 T, respectively. These maxima occur at substantially higher fields compared to unirradiated samples indicating stronger flux pinning at these temperatures. In particular, at 77 K there is a significant hysteresis in the magnetization curve. This contrasts with the unirradiated sample[4] whose magnetization curve was nearly reversible at 77 K above 0.4 T.

The magnetization curve for fields along the *a* direction at 6 K showed a sharp increase for fields up to 0.2 T followed by a gentle rise up to 1 T, as shown in Fig. 3. This behavior is significantly different from that of the unirradiated sample[4] where the magnetization reached a maximum below 0.1 T and decreased by a factor of four between 0.1 and 1 T.

The magnetization curves were analyzed to give critical magnetization currents using the Bean model.[10] For simplicity, we use the field independent critical current formulation of the Bean model to obtain our critical magnetization currents. This gives sufficient accuracy to examine the overall trends in our results. This approximation will be valid for fields well above the maximum in the magnetization curve. Since our samples have rectangular rather than circular cross sections, we use a generalization of the original Bean formulas due to Clem.[11] This allows us to compare the critical magnetization currents for samples of different dimensions. For a sample of rectangular cross section $2a_1 \times 2a_2$ where $a_1 > a_2$, the magnetization is given by

$$M = \frac{a_2}{20} J_c \left(1 - \frac{a_2}{3a_1} \right) ,$$

where J_c is the critical magnetization current in A/cm^2, M is the magnetization in emu/cm^3, and a_1 and a_2 are in cm. This formula reduces to the cylindrical and slab forms given by Bean in the limits $a_1 = a_2 = R$ and $a_1 \rightarrow \infty$, respectively.

The critical magnetization current derived from the magnetization curves on the irradiated and the unirradiated samples are shown in Figs. 4 and 5. We show no critical magnetization current for the field in the *c* direction at $T = 6$ K because the magnetization curve did not reach a maximum below 1 T and the Bean model cannot be applied. At 45 and 77 K there is a general increase in J_c with neutron fluence. Between the sample with the maximum irradiation and the unirradiated sample,[4] there is a factor of 1.6 increase in J_c at 45 K and 2.4 at 77 K in a field of 1 T. The strong decrease in J_c with temperature observed in unirradiated samples is not as serious in the irradiated samples. Between 45 and 77 K in a field of 1 T the unirradiated sample showed a decrease in J_c of a factor of 16. In the irradiated sample, the decrease is reduced to a factor of 11, 30% lower than in the unirradiated sample. Apart from the increase in the magnitude of the critical magnetization current, neutron irradiation does not significantly affect the field dependence of J_c at 45 and 77 K for the field along the *c* direction.

For fields along the *a* direction, both the magnitude and the field dependence of the critical magnetization currents are improved by irradiation as shown in Fig. 5. In the sample with maximum irradiation, the magnetization is still increasing with field at 1 T at 6 K making application of the Bean model questionable. Nevertheless, in order to make an approximate comparison, we use the Bean model on the slowly rising part of the magnetization curve only at 1 T. The value of the critical magnetization current derived in this way is, therefore, a lower limit. The field dependence of J_c in the irradiated sample is quite different from that of the unirradiated sample where a reduction of almost a factor of 4 occurs between 0.1 T and 1 T. At 1 T the improvement of J_c with maximum irradiation is a factor of 5, the largest improvement observed in our experiment. This large increase for the field along the *a* direction compared to the smaller increase for the field along the *c* direction indicates that the strong anisotropy of J_c observed in unirradiated crystals is reduced by neutron irradiation.

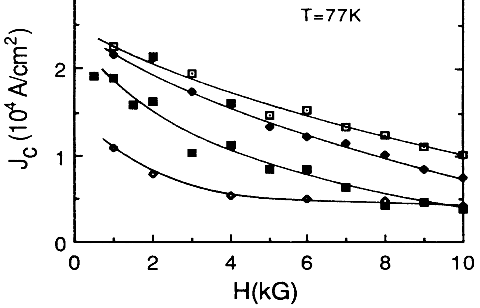

FIG. 4. Critical magnetization current derived from the Bean model for the unirradiated and irradiated samples for the field along the *c* direction at 45 and 77 K.

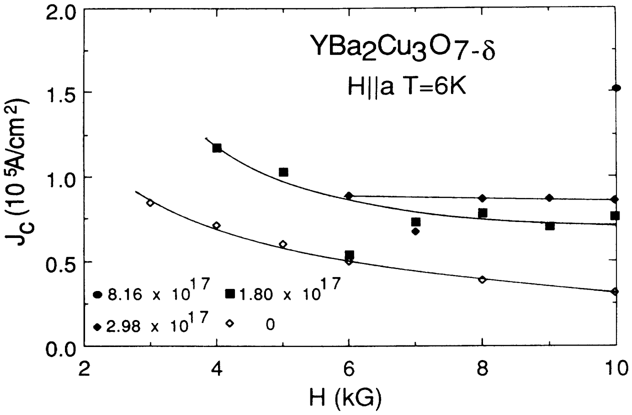

FIG. 5. Critical magnetization current derived from the Bean model for the unirradiated and irradiated samples for the field along the *a* direction at 6 K.

There is a slight decrease in superconducting transition temperature with irradiation as shown in Fig. 1, where T_c has been defined as the onset of diamagnetism. The average decrease obtained from the initial slope of a least squares fit to the data is 2.6 K/10^{18} $n\,\mathrm{cm}^{-2}$. This is to be compared with 1.7 K/10^{18} $n\,\mathrm{cm}^{-2}$ for polycrystalline YBa$_2$Cu$_3$O$_{7-\delta}$ at similar fluences (Ref. 6) and 2.1 K/10^{18} $n\,\mathrm{cm}^{-2}$ for polycrystalline La$_{1.85}$Sr$_{0.15}$CuO$_4$ (Ref. 9). In all cases the slope was obtained from a small number of points making precise comparisons difficult. The percentage decrease in the transition temperature of YBa$_2$Cu$_3$O$_{7-\delta}$ with neutron fluence is comparable but slightly less than that typically observed in the *A*15 superconductors.[12]

The structure in the shielding curves illustrated in Fig. 1 indicates that the temperature dependence of the shielding characteristics varies from sample to sample, especially in the temperature range above ~45 K. Thus, some of the irregularities in the critical magnetization currents of the irradiated samples in this temperature range may be related to variations in the shielding properties among the samples.

Our data show that neutron irradiation is a viable method for improving the strength of the flux pinning and raising the critical magnetization current of YBa$_2$Cu$_3$O$_{7-\delta}$. The larger increase in the critical magnetization current at 77 K compared to 45 K makes neutron irradiation an attractive method for increasing the critical current for applications intended to operate at liquid-nitrogen temperatures. The fact that the largest improvements in the critical magnetization currents were observed for fields along the *a* direction suggests that the anisotropy in the critical magnetization currents observed in unirradiated crystals may not be intrinsic. The systematic increase of the critical magnetization current with neutron fluence indicates that further significant increases can be achieved with higher fluences without severely depressing the transition temperature.

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