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## ENHANCEMENTS OF THE CRITICAL CURRENTS OF YBaCuO SINGLE CRYSTALS BY NEUTRON (n) AND PROTON (p) IRRADIATION

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Abstract--We present the results of magnetization hysteresis and T<sub>c</sub> measurements of neutron and proton irradiated YBaCuO single crystals. The crystals used for comparison were irradiated to a fluence of  $2x10^{17}$  n/cm<sup>2</sup> (n, E>0.1MeV) and 1x10<sup>16</sup>p/cm<sup>2</sup> (p, E=3.5MeV). The critical currents at 1T and 10K are enhanced by a factor of 5 for the neutron irradiated and a factor of 9 for the proton irradiated sample respectively. After irradiation the crystals were annealed at 100, 200 and 300°C for 8h each in air. Following each annealing step the critical temperature and the magnetization hysteresis at 10 and 70K was measured. Upon annealing, we observe a decrease of the critical currents, which is more pronounced for the proton irradiated sample. This decrease is related to the removal of point defects or their small clusters. Thus, their contribution to pinning can be studied. The critical temperature decreases after both types of irradiation by about 0.5K and is fully recovered after annealing.

#### **1. INTRODUCTION**

It is well established that the critical currents of YBaCuO single crystals increase upon irradiation with neutrons[1], protons [2], electrons [3]. The defects created are different, depending on the kind of irradiation imposed on the material. Whereas with electron irradiation only point defects and point defect clusters are created, neutron and proton irradiation creates point defects, point defect clusters and cascade defects. Since proton irradiation possesses a larger fraction of low energy recoils than neutron irradiation at the chosen fluences [4], more point defects and less defect cascades compared to neutron irradiation are created. In earlier work, [5] it has been shown, that cascade defects are effective pinning centers. The role of point defects, in contrast has not yet been fully clarified, partly because they are invisible in conventional TEM. It has been shown in the study of electron irradiated crystals, that point defects are effective pinning centers [3], [6], and that they are removed by annealing [7]. Cascade defects, in contrast are stable at annealing temperatures up to 400°C [5], [8]. Thus, annealing experiments provide the possibility to distinguish between point defects or their clusters and defect cascades.

In this paper we present the results of the critical current and critical temperature measurements on two YBaCuO single crystals, which were irradiated with neutrons and protons respectively. After irradiation they were annealed at 100°C, 200°C and 300°C in air for 8h each. The fluences for irradiation were chosen so that the maximum of the critical current is reached. After irradiation the samples exhibit comparable critical currents. The critical current at 10K and 1T is enhanced by a factor of 5 for the neutron irradiated sample and a factor of 9 for the proton irradiated sample. The enhancement of the critical current of the proton irradiated sample is mainly due to the creation of point defects or their clusters, whereas for the neutron irradiated sample it is caused by a combination of point defects or their clusters and defect cascades. As expected, annealing effects in the proton irradiated sample are stronger than in the neutron irradiated sample. The critical temperatures are decreased after irradiation with neutrons and protons by about 0.5K and 1K respectively and recover fully after annealing.

#### 2. EXPERIMENT

 $T_c$  measurements were carried out in a field of 10e with Hlk, by cooling in zero field. Magnetization hysteresis measurements were carried out in a Quantum Design SQUID magnetometer in fields up to 5Tesla at temperatures of 10 and 70K.

Neutron irradiations were performed at the Missouri University Research Reactor (MURR) in the H1 position to a fluence of  $2x10^{17}$ n/cm<sup>2</sup> (E>0.1MeV). Proton irradiation was carried out at room temperature in the TANDEM accelerator at Argonne National Laboratories. The crystal was irradiated to a fluence of  $1x10^{16}$ p/cm<sup>2</sup> with 3.5MeV protons. The thickness of the crystal (30µm) was less than the range of the protons at this energy (55µm).

In the course of this experiment 6 neutron irradiated samples and 3 proton irradiated samples have been studied. The results are similar in all cases, the crystals shown here represent typical examples of the results.

#### 3. RESULTS

The critical currents were calculated from the magnetization hysteresis measurements by using the anisotropic critical state model [9], [10]. In order to characterize the annealing results, the following definition for the reduction of the critical currents is used:

decrease(%) = 
$$\frac{J_{c}(after irr.) - J_{c}(after annealing)}{J_{c}(after irr.) - J_{c}(before irr.)} \times 100$$
 (1)

Fig. 1 shows the results of the critical currents after irradiation and annealing for the neutron irradiated sample.



Fig.1. Critical current density as a function of field for Hllc at 10K (top) and 70K (bottom) before neutron irradiation, after neutron irradiation and after annealing at 100°C, 200°C and 300°C

Upon irradiation to a fluence of  $2 \times 10^{17}$  n/cm<sup>2</sup> we observe an increase in the critical current by a factor of 5 at 1Tesla and 10K. The increase of the critical current at 70K would be infinite, since essentially no hysteresis was observed before irradiation. After annealing at 100°C and 200°C we observe a slight decrease of the critical current at 10K (using equation 1, less than 10% at each step). After annealing at 300°C the critical current is reduced by about 30%. At 70K, each annealing at 100°C has the smallest impact, leading to a decrease of about 10%, annealing at 200°C and 300°C reduces the critical current by about 30% and 50% respectively.

Fig. 2 shows the results of annealing of the proton irradiated sample. Proton irradiation to a fluence of  $1 \times 10^{16}$  p/cm<sup>2</sup> leads to critical currents comparable to that of the neutron irradiated sample. A variation in critical currents by a factor of two is within the statistical variation of several crystals irradiated under identical conditions. The decrease of the critical current following annealing, however, seems now to be independent of measuring temperature. Each annealing step leads roughly to about the same decrease of the critical current of about 20%. After the final annealing step the critical current is decreased by about 70% at 10K. A summary of the decrease of the critical currents as a function of annealing temperature at a field of 2T is given in table1.



Fig.2. Critical current density as a function of field for Hllc at 10K (top) and 70K (bottom) before proton irradiation, after proton irradiation and after annealing at 100°C, 200°C and 300°C

Fig. 3 and Fig. 4 show the results of the measurements of the critical temperature of the neutron and proton irradiated sample respectively.

Neutron irr.	100°C	200°C	300°C
10K	0%	9%	33%
70K	9%	34%	52%
Proton irr.	100°C	200°C	300°C
10K	23%	47%	68%
70K	15%	32%	60%

Table.1 recovery of the critical current at a field of 2Tesla

Neutron irradiation reduces the critical temperature by about 0.5K. Each annealing step increases  $T_c$ , after annealing at 300°C the critical temperature is even slightly higher than before irradiation.

Proton irradiation leads to a reduction of the critical temperature by about 1K. Again it increases with each annealing step, after annealing at 300°C the critical temperature is again higher than before irradiation.



Fig.3. T<sub>c</sub> before irradiation, after irradiation and annealing at 100°C, 200°C, 300°C of the neutron irradiated sample



Fig.4.  $T_C$  before irradiation, after irradiation and annealing at 100°C, 200°C, 300°C of the proton irradiated sample

## 4. DISCUSSION

Neutron irradiation creates defect cascades, point defects and clusters, whereas proton irradiation creates predominantly point defects and clusters. Both types of irradiation lead to an increase of the critical currents. Annealing the crystals at 100°C, 200°C and 300°C reduces the critical currents for both, the neutron and proton irradiated samples and leads to a recovery of  $T_c$ . It was shown in a previous publication [5], that annealing at these temperatures

leaves the cascade defects unchanged, the decrease of the critical currents after annealing is therefore attributed to the removal of point defects or their clusters. The larger effect of annealing on the critical currents of the proton irradiated samples can be explained by the higher amount of point defects present after this type of irradiation. Since each annealing step leads to about the same decrease of the critical currents at 10 and 70K, these point defects or clusters are contributing to pinning by the same amount at each measuring temperature. About 70% of the critical current at 10K and 60% of the critical current at 70K is provided by point defects or their clusters. The pinning remaining after irradiation might be related to a more complex defect structure, which is described in detail in a recent publication [4].

In contrast, the critical currents of the neutron irradiated sample are more reduced at 70K than at 10K. This suggests that in this case, the point defects or their clusters removed by annealing contribute more to pinning at higher measuring temperatures. This is surprising, since the defect cascades which are visible in TEM, are of a bigger size than the point defects or their clusters. The pinning left after annealing is then provided by the cascade defects which are very efficient pinning centers at 10K. The point defects or clusters are responsible for about 50% of the critical current at 70K, and only 30% at 10K.

Our data do not give any direct indication for the specific point defects or clusters responsible for pinning, however, based on former results on electron [3] as well as neutron irradiation [11], and taking the relatively low annealing temperatures into account, we believe that the point defects or clusters responsible for our annealing results are O displacements on the basal plane and Cu displacements on the CuO<sub>2</sub> plane. The latter is believed to recover at annealing temperatures starting at 300°C, and therefore responsible for the additional pinning in the neutron irradiated sample at 10 and 70K.

The reduction in  $T_c$  is caused by oxygen disorder, which recovers during annealing at 100°C and 200°C. These defects are not contributing significantly to pinning in the case of neutron irradiation. at 10K. It is interesting to note, that the critical temperature recovers fully after annealing, in both cases even slightly higher values than before irradiation are observed.

#### 5. SUMMARY

After irradiating two YBaCuO single crystals with neutrons and protons at fluences where the maximum enhancement of the critical currents occurs, we observe comparable critical currents after irradiation. The depression of the critical temperature at these fluences is small, being about 0.5K for the neutron irradiated and 1K for the proton irradiated sample. Annealing experiments carried out at 100°C, 200°C and 300°C reveal that both types of irradiation create point defects or their small clusters, which contribute significantly to pinning. In the case of the proton irradiated sample these defects act as the main pinning centers being responsible for about 70% of the critical current after irradiation. Neutron irradiated samples are less susceptible to annealing, due to an additional defect cascade structure, which is stable upon annealing. Point defects or clusters in this case contribute to 50% of the critical currents at 70K, and 30% of the critical current at 10K.

# induced flux pinning in Gd-doped $Y_1Ba_2Cu_3O_{7-x}$ and $Gd_1Ba_2Cu_3O_{7-x}$ " submitted to Phys. Rev. B

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#### REFERENCES

[1] A. Umezawa, G. W. Crabtree, J. Z. Liu, H. W. Weber, W. K. Kwok, L. H. Nunez, T. J. Moran, C. H. Sowers, "Enhanced critical magnetization currents due to fast neutron irradiation in single crystal YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub>", Phys. Rev. B., vol.36, pp.7151, November 1987.

[2] L. Cevale, A. D. Marwick, M. W. Mc Elfresh, T. K. Worthington, A. P. Malozemoff, F. H. Holtzberg, M. A. Kirk,"Defect Independence of the irreversibility line in proton-irradiated Y-Ba-Cu-O Crystals", Phys. Rev. Lett., vol. 65, pp.1164, August 1990

[3] J. Giapintzakis, W. C. Lee, J. P. Rice, D. M. Ginsberg, I. M. Robertson, M. A. Kirk, R. Wheeler, "The production and identification of flux pinning defects by electron irradiation in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> single crystals", Phys. Rev. B, vol.45, pp.1677

[4] M. A. Kirk, "Structure and flux pinning properties of irradiation defects in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub>" in *Critical Currents in High Tc Superconductors*, Vienna, Austria, April 1992, to be published in Cryogenics

[5] B. M. Vlcek, M. C. Frischherz, S. Fleshler, U. Welp, J. Z. Liu, J. Downey, K. G. Vandervoort, G. W. Crabtree, M. A. Kirk, J. Giapintzakis, J. Farmer, "Flux pinning in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals: Neutron irradiation and annealing", Phys. Rev. B. vol. 46, pp.6441, Sept. 1992

[6] M. Konczykowski, "Flux pinning enhancement after electron irradiation in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>", Physica A., vol. 168, pp.291,

[7] J. Giapintzakis, M. A. Kirk, W. C. Lee, J. P. Rice, D. M. Ginsberg, I. M. Robertson, R. Wheeler, "Flux pinning induced by electron irradiation in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals" to be published in Proc. Mat. Res. Soc. Symp. S, April 1992

[8] M. C. Frischherz, "Transmission electron microscopy studies of radiation induced defects in the high temperature superconductor YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>", Technical University of Vienna, Vienna, Austria, Ph. D. dissertation, 1992

[9] E. M. Gyorgy, R. B. van Dover, K. A. Jackson, L. F. Schneemeyer, J. V. Waszcak, "Anisotropic critical currents in Ba<sub>2</sub>YCu<sub>3</sub>O<sub>7</sub> analyzed using an extended Bean model", Appl. Phys. Lett, vol. 55, pp..283, July 1989

[10] F. M. Sauerzopf, H. P. Wiesinger, H. W. Weber, "Anisotropic current flow and demagnetization corrections in the Bean model", Cryogenics, vol.30, pp. 650, July 1990

[11]K. E. Sickafus, J. O. Willis, P. J. Kung, W. B. Wilson, D. M. Parkin, M. P. Maley, F. W. Clinard, "Neutron irradiation







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