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J. Giapintzakis[1], M. A. Kirk[2] W. C. Lee[1], J. P. Rice[1], D. M. Ginsberg,[1]
I. M. Robertson[1] and R. Wheeler[2]
[1]Department of Physics and Materials Research Laboratory
University of Illinois at Urbana-Champaign, Urbana, IL 61801

[2]Materials Science Division
Argonne National Laboratory, Argonne, IL 60439

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FLUX PINNING DEFECTS INDUCED BY ELECTRON IRRADIATION IN $Y_1Ba_2Cu_3O_{7-\delta}$ SINGLE CRYSTALS

J. GIAPINTZAKIS,* M. A. KIRK,+ W. C. LEE,* J. P. RICE,* D. M. GINSBERG,* I. M. ROBERTSON,* R. WHEELER+
*Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, 1110 W. Green Street, Urbana, Illinois 61801
+Materials Science Division, Argonne National Laboratory, 9700 S. Cass Avenue, Argonne, Illinois 60439

ABSTRACT

Single crystals of $R_1Ba_2Cu_3O_{7-\delta}$, (R=Y, Eu and Gd), have been irradiated with 0.4-1.0 MeV electrons in directions near the c-axis. An incident threshold electron energy for producing flux pinning defects has been found. In-situ TEM studies found no visible defects induced by electron irradiation. This means that point defects or small clusters ($\leq$ 20 Å) are responsible for the extra pinning. A consistent interpretation of the data suggests that the most likely pinning defect is the displacement of a Cu atom from the CuO_2 planes.

1. INTRODUCTION

J_c can be increased by introducing defects into materials. There are many techniques to accomplish this, but irradiation by particles has proved to be one of the most effective methods for the materials at hand. Moreover, electron irradiation has the advantages over other types of particle irradiation, in that: a) it produces the simplest type of defects, point defects, which are uniformly distributed throughout the sample and b) it enables us to study the effect of certain lattice defects on superconducting properties, because the good control of the transferred energy enables us to selectively produce defects on the different sublattices.

The focus of our work has been on the enhancement of J_c and most importantly on the understanding of the underlying mechanism of flux pinning in single crystals of $YBa_2Cu_3O_{7-\delta}$ (YBCO). To that end we have studied the dependence of J_c in single crystals on the irradiation conditions: energy, fluence and temperature, in association with TEM information on the microstructure created by the electron irradiation. More specifically, we have: (i) performed in-situ TEM experiments to determine the size and concentration of the radiation-induced defects and (ii) identified the sublattice responsible for pinning with H//c.

2. EXPERIMENTAL TECHNIQUES

Single crystals were used in this work to avoid the complication of the weak links present in sintered materials and in particular single crystals of YBCO because by far the best quality HTSC crystals available at present can be made of this compound. The single crystals were grown by a "self flux" method in yttrium-stabilized zirconia crucibles [1]. The samples used in this study included twinned and untwinned crystals of nearly square shape with dimensions up to 0.5x0.5 mm^2 and thickness of 10-60 μm. All of the crystals had initial transition temperatures (T_c) in the range of 90-91 K and transition widths $\Delta T_c \sim 2$ K. Preirradiation J_c values in a magnetic field of 1 T at 10 K and 40 K were $1x10^6$ and $1x10^5$ Acm^{-2}, respectively, for H//c-axis.

The electron irradiations were performed at the HVEM/accelerator facility at Argonne National Laboratory. Much effort was expended to ensure that the temperature of the sample did not exceed a nominal value of 300 K. This was accomplished by rigidly mounting the bulk crystal on a copper grid with silver paint. The irradiations were performed at 100 and 300K using 0.4 - 1.0 MeV electrons at a flux of $\sim 2x10^{15}$ cm^{-2} sec^{-1} in a vacuum of $\sim 1x10^{-6}$ Torr. The crystals were flat, and were irradiated 8-10 degrees off the c-axis to avoid channeling effects.

Measurements of magnetization as a function of applied field, M(H), at 10 K, 40 K and 70 K were made at fields up to 5 T for the H//c-axis orientation in a Quantum Design SQUID magnetometer. No attempt was made to correct the M(H) data for demagnetizing effects, since we were interested only in the relative changes of the J_c.

The HVEM was employed to examine in-situ the defect microstructure produced in thin samples under various electron irradiation conditions. Bulk single crystals of YBCO were thinned by electropolishing, and were irradiated with 1 MeV electrons to doses ranging from 10^{18}-10^{20} cm^{-2} and at rates of 10^{15}-10^{18} cm^{-2} sec^{-1}. In-situ imaging conditions in the HVEM employed diffraction contrast using g=200 dark field at electron voltages of 1 MeV and 100 keV.

3. RESULTS

3.1 J_c(H,T) Measurements

A significant effect of electron irradiation on the magnetic properties of YBCO single crystals can be observed in Figures 1a-1b. M(H) curves for crystal #1 at T=10 K

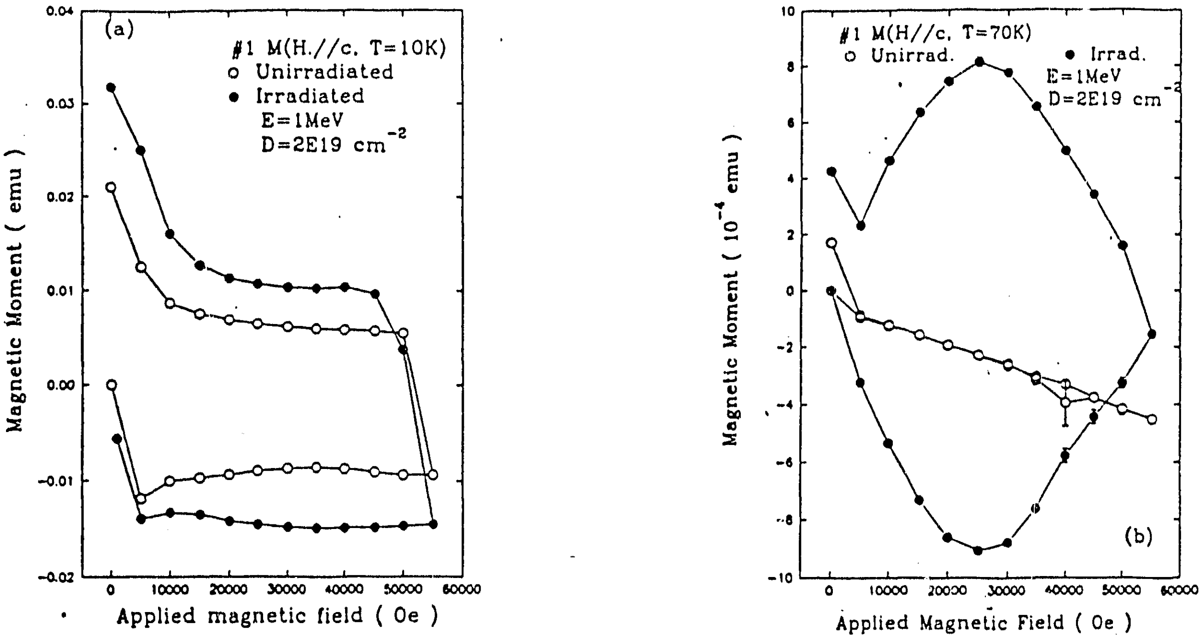

Figure 1. Magnetic moment as a function of applied magnetic field for crystal #1 at (a) 10 K and (b) 70 K, before and after irradiation by 1 MeV electrons.

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and 70 K before and after irradiation by a 1 MeV electron beam to a dose $\Phi=2x10^{19}$ cm^{-2} are shown. The Figures clearly indicate an increase of the width of the hysteresis loop (ΔM) following irradiation. According to the Bean critical-state model [2] $J_c \propto \Delta M$, where ΔM is the difference between the upper and lower branches of the M(H) loop. Hence J_c has been enhanced as a consequence of extra pinning generated by the irradiation-induced defects.

The M(H) curves at T=10 K for crystal #2, before and after irradiation by a 0.4 MeV electron beam to a dose $\Phi=1.4x10^{19}$ cm^{-2}, are illustrated in Figure 2. The data indicate that instead of enhancing the flux pinning capability of the crystal we have actually reduced it. Shown in Figure 3 are plots of M(H) for crystal #3 at T=10 K both before and after irradiation by a 0.6 MeV electron beam to $\Phi=$ $2.9x10^{18}$ cm^{-2} and $6.2x10^{18}$ cm^{-2}. The Figure clearly indicates that defects produced by 0.6 MeV electrons are effective flux pinning sites. From Figures 2 and 3 we therefore conclude that there is a threshold incident electron energy for enhancing flux pinning.

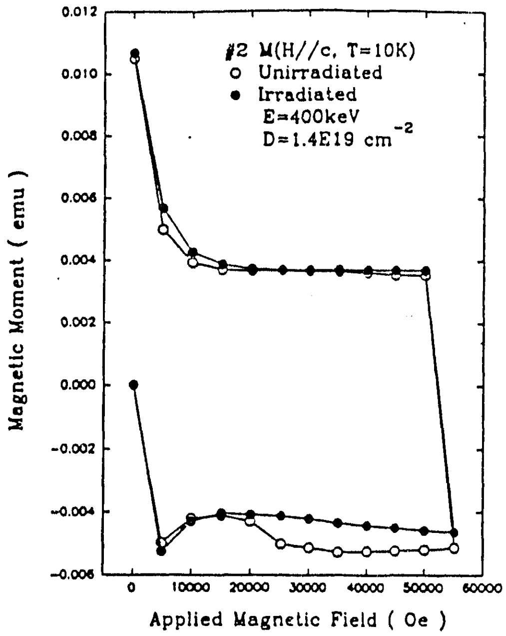

Figure 2. Magnetic moment as a function of applied magnetic field for crystal #2 at 10 K before and after irradiation by 400 keV electrons.

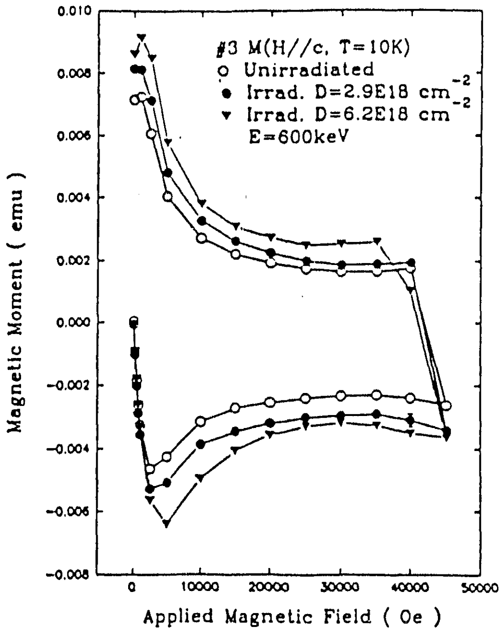

Figure 3. Magnetic moment as a function of applied magnetic field for crystal #3 at 10 K before and after irradiation by 600 keV electrons.

3.2 Annealing Studies

Crystal #4 was irradiated by 1 MeV electron beam to $\Phi=1.55x10^{19}$ cm^{-2} at 100 K and then annealed at 300 K over a period of a few days before its T_c and M(H) curves were measured. As shown in Figures 4a and 4b, we observed an increase of T_c by about a degree, as well as an enhancement of J_c by 22%.

After we irradiated crystal #5 by a 1 MeV electron beam to $\Phi=7x10^{19}$ cm^{-2} at 300 K, it exhibited an observable decrease of T_c and an enhancement of J_c. Then it was annealed in 100% dried oxygen atmosphere at 200 °C for 4 hours. Figures 5a and 5b show that during annealing we recovered 100% of T_c but only about 25% of J_c.

3.3 TEM observations

No defect structure could be observed in the in-situ HVEM experiments under electron irradiation conditions similar to those of the bulk crystal irradiations. The same was observed under other irradiations up to higher doses and employing higher dose rates. Under the HVEM imaging conditions employed, the resolution limit was ≤ 20 Å. No defect structure larger than this resolution limit was observed.

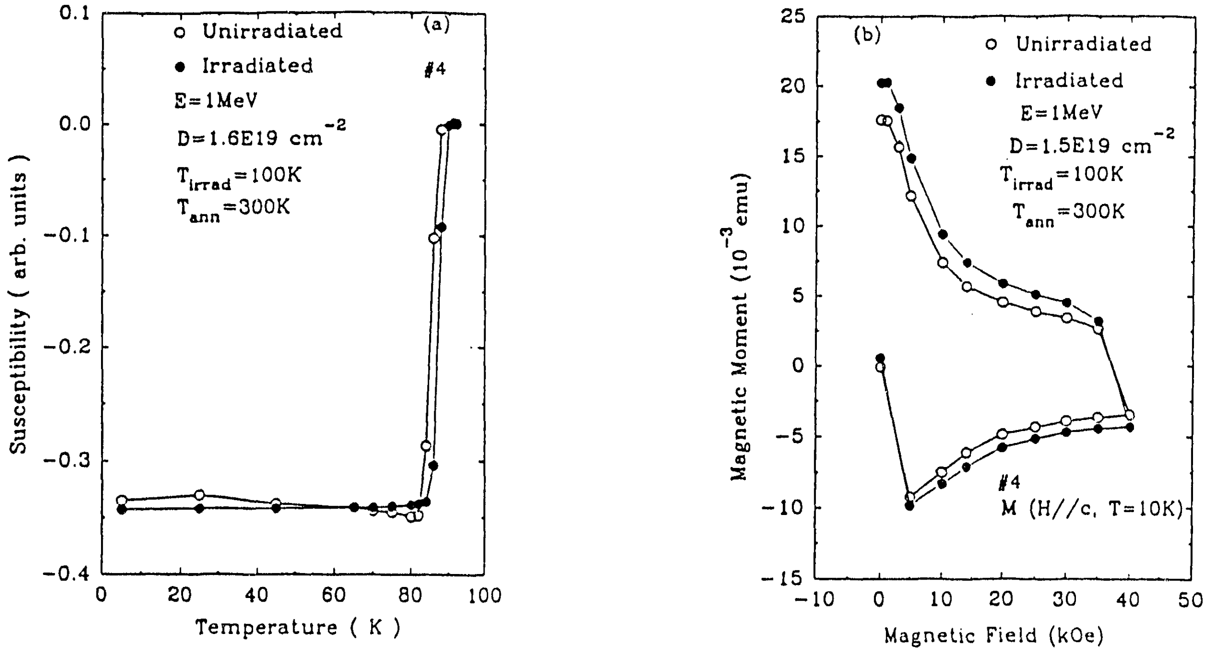

Figure 4. (a) Susceptibility as a function of temperature at 10 Oe and (b) magnetic moment as a function of applied magnetic field at 10 K for crystal #4, in the unirradiated state and following irradiation at 100 K with a subsequent annealing at 300 K for a few days.

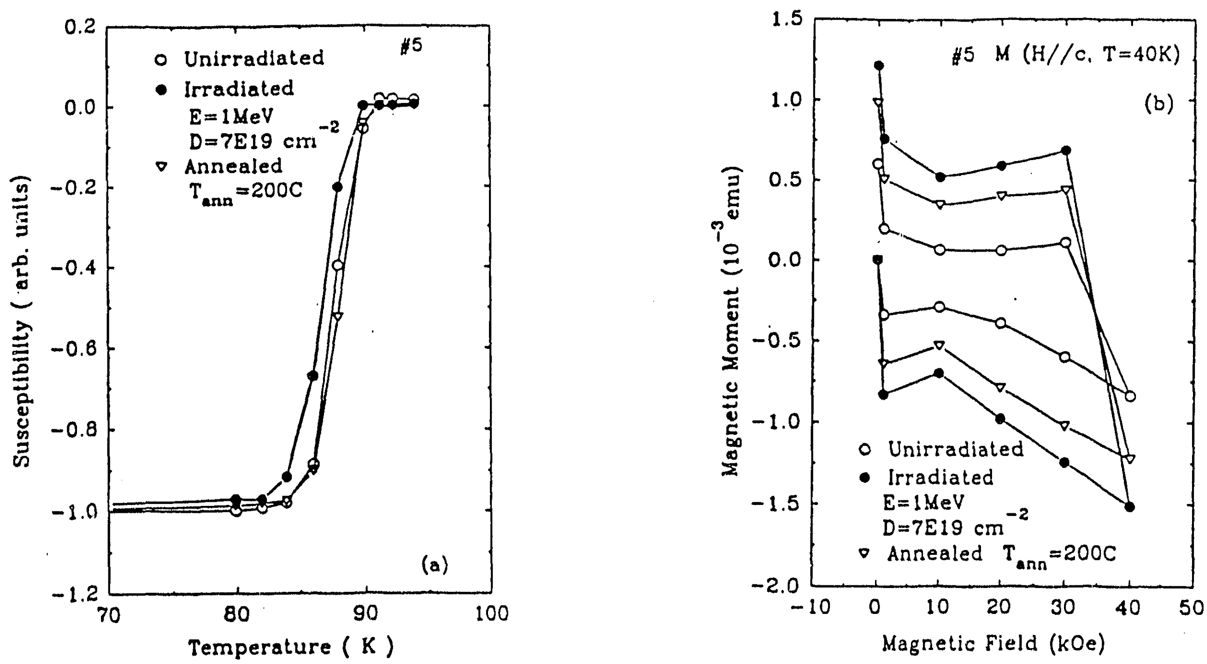

Figure 5. (a) Susceptibility as a function of temperature at 10 Oe and (b) magnetic moment as a function of applied magnetic field at 40 K for crystal #5, in the unirradiated state, after electron irradiation at 300 K, and after annealing at 200 °C.

4. DISCUSSION

4.1 Radiation Damage

The data tell us about the effects of displacement damage on the superconducting properties, such as J_c and T_c, produced by the electron irradiation of YBCO single crystals. During irradiation the incident electrons collide with the target atoms, transferring part of their energy, producing primary knock-on atoms (PKAs). These PKAs in turn are able to produce defects if their energy exceeds the threshold for producing a stable displaced atom. The primary defect is thus usually considered to be a Frenkel pair, although variations on this simple concept should be considered in a complex structure. The final stable defect structure might also consist of point defect clusters if either the vacancy or interstitial is mobile at the irradiation temperature. Depending on the defect's location within the unit cell as well as the size and concentration of the point or small cluster defects, the superconducting properties of the material can be altered, possibly increasing J_c, if these defects can effectively pin magnetic flux lines by locally depresing the superconducting order parameter.

Making the simple assumption that it requires about 20 eV to permanently displace any of the four atom types, this threshold is exceeded by 1 MeV electrons for all atoms, since the minimum electron energies required for displacement are 129, 413, 532, and 730 keV for O, Cu, Y and Ba, respectively.

4.2 Identification of Primary Pinning Defect

The possible pinning sites in the YBCO structure are defects in the Y-site, Ba-site, Cu- and O-sites in the chains, Cu- and O- sites in the CuO_2 planes. The threshold incident electron energy of ~0.6 MeV suggests that the candidates can be reduced to O, Cu, and Y sites, since making the assumption that E_d~20 eV a stable defect in the Ba site would require an incident energy of 0.73 MeV. Substituting the Y by much heavier rare-earth elements such as Eu and Gd, which would require incident electron energies of 0.784 and 0.802 MeV for direct displacements, we observed a similar flux pinning enhancement for these compounds as for the YBCO ones following irradiation by a 0.6 MeV electron beam. This suggests that the Y site is not an effective pinning site in agreement with the recent results of K. Sickafus et al.[3]. So the remaining candidates are the Cu and O sites at the chains and CuO_2 planes.

The annealing data for crystals #4 and #5 suggest that oxygen ordering in the chains can be achieved by electron irradiation and annealing, while an increase in J_c can be produced following the same irradiation and annealing. This argues for a primary pinning defect (for H//c) not associated with oxygen disorder in the chains on a local or unit-cell scale. However, in agreement with the results of the 0.4 MeV irradiation (Figure 2), the removal of some extended regions of oxygen deficiency, possible pinning defects, by radiation-enhanced oxygen diffusion appears possible. In addition the copper at the chains can also be eliminated as a primary pinning site based on the argument that its position in unfavorable for creating stable defects that can pin strongly. Hence the data suggest that the primary pinning defects must be displacements of Cu and/or O atoms form the CuO_2 planes. This is in agreement with the argument that defects on the CuO_2 planes pin most effectively because they would produce a strong disruption of the local electronic structure in the strongly superconducting planes [4].

Based on calculations of cross sections for the Cu and O displacements, using the threshold incident electron energy for producing pinning, we can calculate the areal density of Cu and O defects on the CuO_2 planes. Then we can calculate the J_c that these point defects are responsible for by using: (i) the Thuneberg's electron scattering formalism [5] to calculate the elementary pinning force between a single vacancy and a single vortex and (ii) the 1-D limit of the Collective Weak Pinning Theory [4] to calculate the macroscopic pinning force. The results were that $J_c \sim 2 \times 10^5$ Acm^{-2} due to Cu point defects and $J_c \sim 8 \times 10^4$ Acm^{-2} due to O point defects. Hence the results slightly favor the Cu vacancy as the primary pinner. When these numbers are compared with the experimental result of $\Delta J_c = J_c$(after irradiation)-J_c(before irradiation)$\sim 1 \times 10^6$ Acm^{-2} we find that in addition to point defect pinning we must consider another pinning mechanism such as small point defect clusters.

5. CONCLUSION

We have shown that 1 MeV electron irradiation results in an enhancement of J_c in YBCO single crystals. In-situ TEM studies in the HVEM suggest that the pinning centers must be small ($\leq$ 20 Å). A consistent interpretation of our data suggests that the primary pinning defect is most likely the displacement of a copper atom from the CuO_2 plane. But in order to account for the entire enhancement of J_c other pinning mechanisms aside from point defects should be considered, such as small point defect clusters.

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