Controllable reduction of critical currents in $YBa_2Cu_3O_{7-\delta}$ films

Alice E. White, K. T. Short, R. C. Dynes, A. F. J. Levi, M. Anzlowar, K. W. Baldwin, P. A. Polakos, T. A. Fulton, and L. N. Dunkleberger
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

(Received 6 June 1988; accepted for publication 21 July 1988)

The critical currents in high quality thin films of the high T_c superconductor, $YBa_2Cu_3O_{7-\delta}$, can be controllably reduced by orders of magnitude using ion irradiation. This reduction in critical current occurs without substantial decrease in T_c or increase in room-temperature resistivity. Using this technique, we have fabricated weak links that exhibit an ac Josephson effect at 77 K.

Most technological applications for the high T_c superconductors require critical current densities in excess of 10^5 A/cm^2 at 77 K. However, for applications which involve a Josephson "weak link" between two regions of good superconductor, the ability to controllably reduce the critical current (I_c) would be very convenient. For example, in order to optimize the sensitivity of a superconducting quantum interference device (SQUID),[1] the parameters should be chosen to satisfy

$$2LI_c/\phi_0 \sim 1,$$

where L is the loop inductance and ϕ_0 is the fundamental flux quantum.[2] It is difficult to make structures small enough to lower the L below 10^{-9} H, so often I_c must also be reduced. By introducing defects with an ion beam, we show that the critical currents in $YBa_2Cu_3O_{7-\delta}$ films can be reduced by orders of magnitude without substantial changes in T_c or the room-temperature resistivity. The use of ion beams for this purpose is particularly appealing because it is compatible with conventional integrated circuit fabrication techniques.

In earlier work on $YBa_2Cu_3O_{7-\delta}$ films,[3] ion bombardment resulted in resistance (R) transitions that showed a signature of the original onset even at high fluences, but broadened continuously. We interpreted this as evidence for a loss of phase coherence between regions of good superconductor. In fact, ion-beam-induced damage in these films starts to degrade the zero resistance temperature [$T(R=0)$] before changes in the crystal structure, as measured by ion channeling or x-ray diffraction, can be observed.[3,4] Much progress has been made in the fabrication of epitaxial $YBa_2Cu_3O_{7-\delta}$ films since then,[5] with concomitant changes in the radiation effects.

The films studied in this work were fabricated by simultaneous deposition of Y, Cu, and BaF_2 in a partial pressure (1×10^{-5} Torr) of O_2 on (100) $SrTiO_3$ substrates.[5] The Y was evaporated using electron beam heating, and the Cu and BaF_2 were evaporated resistively. Annealing was performed at temperatures of 800 °C in flowing O_2 that had been bubbled through H_2O. The presence of H_2O facilitates the removal of the F from the as-deposited film and accelerates the formation of the pure superconducting phase. The stoichiometry of the as-deposited films was confirmed using Rutherford backscattering spectrometry (RBS) and the axial alignment of the reacted films with the $SrTiO_3$ substrate was determined with ion channeling. After annealing, the films were lithographically patterned and etched in dilute HCl to form a 50-μm-wide bridge geometry which enabled four-terminal resistance measurements. All the films discussed here had sharp superconducting transitions (<3 K) with $T(R=0)$'s of >90 K and axial alignments of $>70\%$ before irradiation. We chose an energy of 1 MeV for the Ne^+ bombardment to ensure that the range of the ions was much greater than the ~ 2000 Å film thickness.

In this study, I_c was defined as the current at the break in the current-voltage (I-V) characteristic of a bridge (which corresponds to developing $\sim 0.5\,\mu V$ between the voltage probes). Critical current densities (J_c) in these oriented films are typically 10^5–10^6 A/cm^2 at 77 K. No sign of an enhancement of I_c (Ref. 6) was observed at the lowest fluence ($1\times10^{11}/cm^2$). Decreases in I_c were observed at fluences as low as 5×10^{11} Ne^+/cm^2, even before changes in the R vs T plots could be identified. As shown in Fig. 1(a), the decrease in I_c with ion fluence is monotonic and nonlinear. Although the fluence scale is slightly different for each film, the shape of the curves is similar. In general, once a section of the film has been characterized, the reduction of I_c with irradiation is exactly reproduced in other sections. As illustrated in Figs. 1(a) and 1(b), we were able to achieve reductions of three orders of magnitude in I_c while only raising ρ(300 K) 50% and maintaining T_c above 77 K. This suggests that the type of defects being introduced by the ion beam may also be partially responsible for the low critical current densities in polycrystalline ceramics which have T_c's and ρ(300 K)'s that are similar to those of the films.

Identifying the exact nature of these defects is difficult, but the density of displacements can be estimated by doing a Monte Carlo calculation of the nuclear energy deposited in the film,[7] assuming that the energy to displace an atom averages 20 eV and the energy to break a bond averages 1 eV. When recoils are included, the calculation gives $\sim 1\times10^{19}$ displacements/cm^3 for a fluence of 1×10^{12} Ne^+/cm^2 at 1 MeV. Although the calculation is oversimplified, this order of magnitude estimate of displacements can provide pinning centers every 50 Å. Although some pinning centers are necessary to stop flux flow and maintain high critical currents, when they start to overlap on the size scale of the coherence length (~ 30 Å), the fluxoids can move easily between pinning centers and the critical current is greatly reduced. It is not until the defect density approaches interatomic length

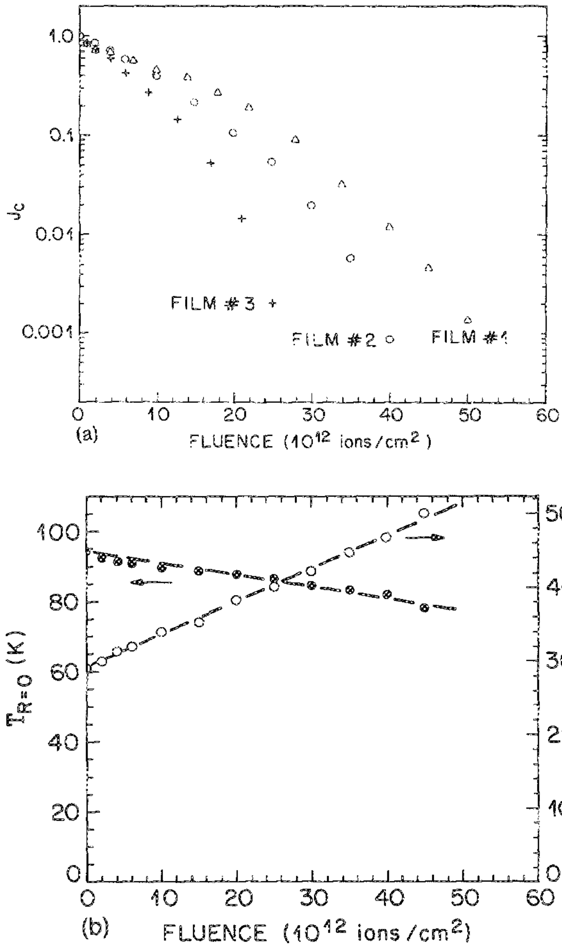

FIG. 1. (a) Reduction of J_c (77 K) in three oriented thin films of $YBa_2Cu_3O_{7-\delta}$ as a result of ion bombardment with 1 MeV Ne^+. The data for each film are normalized to J_c before bombardment: 0.06, 1.16, and 0.42×10^6 A/cm^2 for films No. 1, 2, and 3. (b) Decrease of $T(R=0)$ and increase in ρ(300 K) for film 2 from (a) over the same fluence range. Although J_c drops three orders of magnitude after a total fluence of 4×10^{13} Ne^+/cm^2, ρ(300 K) and $T(R=0)$ have not changed significantly.

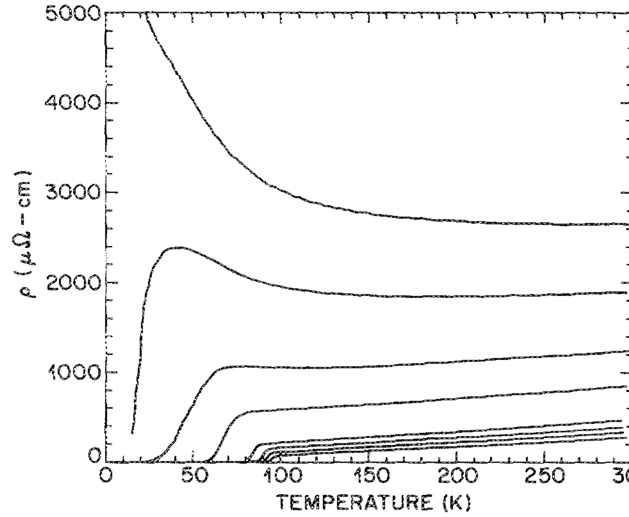

FIG. 2. Series of R vs T plots for a high quality $YBa_2Cu_3O_{7-\delta}$ film (No. 2 from Fig. 1) showing the transition from superconducting to insulating behavior as a function of 1 MeV Ne^+ fluence. For increasing resistivity, the fluences are (from the bottom curve up): undamaged, 1.0, 2.5, 4.0, 10.0, 15.0, 20.0, and 22.0×10^{13}/cm^2.

scales at fluences that are two orders or magnitude higher, that superconductivity is *completely* destroyed (T_c goes to 0 K).

Since the ion-beam-induced changes in $T(R=0)$ and ρ(300 K) have been shown to depend on the nuclear energy deposited in the film,[3,4] we expected the changes in I_c to scale with the deposited nuclear energy also. In order to test this hypothesis, lower energy (200 keV) F^+ irradiations were performed. We chose a fluence of 7×10^{12}/cm^2 in order to approximate a 1 MeV Ne^+ fluence of 3×10^{13}/cm^2. This irradiation resulted in a reduction of greater than 80% in the I_c of the film, similar to that caused by the Ne^+ irradiation, indicating that lower energy irradiation may also be effective in tuning I_c.

We also studied the destruction of superconductivity in these films. A series of R vs T plots at several fluences for a representative film is presented in Fig. 2. Although ρ(300 K) does increase linearly, this series differs in several significant respects from the previous films.[3] In particular, the onset of superconductivity moves to lower temperatures with increasing fluence and the transitions do not broaden substantially. This is more reminiscent of destroying superconductivity by reducing the amplitude rather than the phase coherence of the pair wave function.[8] Nonetheless, the fluence required to eliminate bulk superconductivity in the films is $\sim2\times10^{14}$/cm^2, similar to the previous work.[3]

A possible explanation for the differences between the two sets of films is that inhomogeneities in the early films provided sinks for the ion-beam-induced defects. Several groups have observed that those defects are mobile at room temperature[3,9] which lead us to speculate that the effect of irradiation is to generate disorder on the oxygen sublattice. Regions of high defect concentration will then be insulating,[10] leading to the "granular" behavior observed in the resistive transitions. High-resolution transmission electron microscope (TEM) studies of these films do show mottled contrast on a 100 Å scale, indicative of this sort of granularity.[11] In the absence of TEM studies of the newer, higher quality films, we can only assume that the as-grown films have fewer inhomogeneities, so the ion-beam-induced defects are distributed more uniformly. Estimating the density of vacancies at the fluence required to destroy superconductivity indicates that they occur on a length scale of 8 Å. This uniform, atomic scale disorder can result in reduction of the amplitude of the pair wave function, giving rise to the resistive transitions shown in Fig. 2.

The shape of the R vs T characteristics provide additional evidence for this model. The slope of the linear region from 300 K to just above the transition at 100 K changes very little with damage up to a fluence of 1.5×10^{14}/cm^2, which implies that the defects are simply acting as additional scattering centers and thus increasing the residual resistivity. This behavior is consistent with our model of beam-induced point defects.

As a first step in demonstrating that ion irradiation might be useful for tuning critical currents in Josephson devices, we have irradiated selected areas of thin-film structures with 2.5 μm linewidths which initially did not show any ac Josephson effect. This is in contrast to the work of

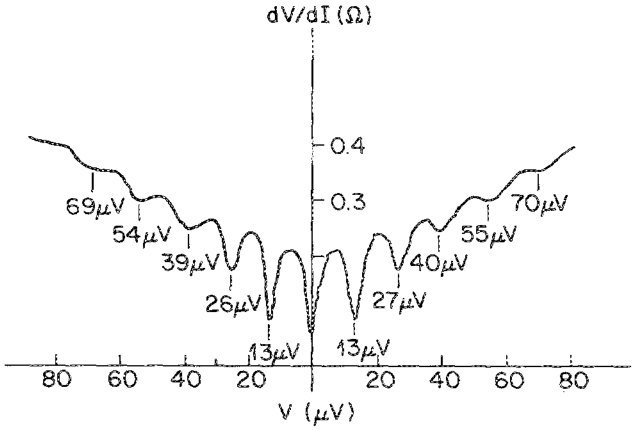

FIG. 3. Plot of dV/dI vs V for a 2.5 μm linewidth $YBa_2Cu_3O_{7-\delta}$ film after bombardment with 1 MeV Ne^+ to a fluence of $3.5\times10^{13}/cm^2$ 1 MeV Ne^+. Shapiro steps induced in the I-V characteristic of the film by 6.5 GHz microwave radiation (ac Josephson effect) are clearly visible as minima in the curve at $V = n(h\nu/2e)$, where n is an integer, h is Planck's constant, and ν is the microwave frequency.

Koch *et al.*,[12] who used ion irradiation to electrically isolate their SQUID patterns. A copper stencil mask with a ~300-μm-diam hole was used to delineate the irradiated region and the fluences of 1 MeV Ne^+ were chosen to reduce the critical currents by two orders of magnitude. Figure 3 shows the microwave response of such a film after irradiation. The critical current was initially 10.5 mA and a fluence of 3.5×10^{13} ions/cm^2 reduced it to 0.095 mA. Shapiro steps appear at a voltage spacing of 13 μV in the I-V characteristics, clearly illustrating the ac Josephson effect and showing that a weak link has been formed.[13]

In conclusion, we have demonstrated that the critical currents of high quality $YBa_2Cu_3O_{7-\delta}$ films can be controllably reduced by ion bombardment. This reduction in I_c is accompanied by relatively small changes in T_c and ρ(300 K). A model which assumes that the ion-beam-induced damage creates point defects on a length scale that results in vortex depinning can explain the observations. At higher fluences, the density of the defects increases, creating disorder on an atomic scale and reducing the amplitude of the pair wave function. This is reflected in the resistive transitions which remain sharp while T(onset) decreases with increasing fluence. The use of ion irradiation to create a weak link which is sensitive to microwave radiation has been demonstrated. This technique has potential application to the fabrication of superconducting weak link devices such as SQUID's and current limiters.

We would like to thank Walter Brown, Bob Cava, Roger Koch, Gene Pacher, John Poate, and Catherine Rice for stimulating discussions.

[1]For a review, see J. Clarke, IEEE Trans. Electron Devices **ED-27**, 1896 (1980).
[2]T. A. Fulton and R. C. Dynes, Phys. Rev. Lett. **25**, 794 (1970).
[3]A. E. White, K. T. Short, D. C. Jacobson, J. M. Poate, R. C. Dynes, P.M. Mankiewich, W. J. Skocpol, R. E. Howard, M. Anzlowar, K. W. Baldwin, A. F. J. Levi, J. R. Kwo, T. Hsieh, and M. Hong, Phys. Rev. B **37**, 3755 (1988).
[4]G. J. Clark, A. D. Marwick, R. H. Koch, and R. B. Laibowitz, Appl. Phys. Lett. **51**, 1753 (1987).
[5]A. F. J. Levi, C. E. Rice, A. Ramirez, J. M. Vandenburg, K. W. Baldwin, M. Anzlowar, A. E. White, and K. T. Short, J. Cryst. Growth (to be published).
[6]A. Umezawa, G. W. Crabtree, J. Z. Liu, H. W. Weber, W. K. Kwok, L. H. Nunez, T. J. Moran, and C. H. Sowers, Phys. Rev. B **36**, 7151 (1987).
[7]J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Range of Ions in Solids* (Pergamon, New York, 1985), Vol. I, p. 109f.
[8]A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. B **33**, 3549 (1986); R. C. Dynes, A. E. White, J. M. Graybeal, and J. P. Garno, Phys. Rev. Lett. **57**, 2195 (1986).
[9]G. C. Xiong, H. C. Li, G. Linker, and O. Meyer, Phys. Rev. B **38**, 240 (1988).
[10]R. J. Cava, B. Batlogg, C. H. Chen, E. A. Reitman, S. M. Zahurak, and D. Werder, Phys. Rev. B **36**, 5719 (1987).
[11]C. H. Chen, D. W. Werder, A. E. White, K. T. Short, P. M. Mankiewich, W. J. Skocpol, and R. E. Howard (unpublished).
[12]R. H. Koch, C. P. Umbach, G. J. Clark, P. Chaudhari, and R. B. Laibowitz, Appl. Phys. Lett. **51**, 200 (1987).
[13]M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1985), p. 203.