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IRRADIATION EFFECTS IN HIGH TEMPERATURE SUPERCONDUCTORS

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ABSTRACT

Irradiation effects on Tl- and Y-based high temperature superconductors (HTS) are compared. Ion irradiation can enhance the critical current density (J_c) at low fluences through the interaction of defects with the flux lattice, and at higher fluences ion irradiation can degrade the superconducting transition temperature (T_c) and increase the normal state resistivity (ρ). Low-fluence irradiation of $Tl_2Ca_2Ba_2Cu_3O_{10}$ (Tl-2223) single crystals was shown to increase J_c by an order of magnitude over J_c for unirradiated crystals. Similarly, irradiated $YBa_2Cu_3O_{7-\delta}$ (YBCO) crystals have increased J_cs by factors of 2 to 100; however the effects of irradiation on thin-film HTS from these two materials depends greatly on their crystalline quality. Ion irradiation of millimeter grain-size Tl-2223 films was shown to increase J_c while similar irradiation of ~10 μm grain-size Tl-2223 films had little effect on J_c. For ions with energies less than ~MeV/amu, the dominant mechanism causing irradiation-induced degradation of T_c is collisional damage. Both Tl-based and YBCO superconductors behave similarly in that T_c decreases linearly with the level of damage; yet, the rate of decrease in T_c for Tl-based superconductors (5000 K/dpa) was approximately twice that for irradiated YBCO. An examination of the temperature behavior and rate of damage recovery of ρ yielded an activation energy of 0.36 eV for annealing defects.

INTRODUCTION

Irradiation of the older "oxygen-free" superconductors such as the A15 compound Nb_3Sn has long been known [1] to cause a steady decrease in the transition temperature (T_c) proportional to the mean energy transfer per lattice atom (the number of displacements per atom - dpa) resulting from nuclear elastic collisions. Also, fast neutron irradiation of Nb_3Sn at 6 K increased the critical current density (J_c) by ≈30% in a 33 kOe field [1]. Although previous data had suggested that grain boundaries were the predominant flux-pinning mechanism in Nb_3Sn, the irradiation results indicated that defect cascades along with the existing microstructure are effective in the pinning of fluxoids. In general, the older A15 superconductors were microcrystalline metallic-like materials with rather large coherence lengths (~100 nm). By contrast, the new oxide superconductors have much smaller coherence lengths (~1 nm) and for the $YBa_2Cu_3O_{7-\delta}$ (YBCO) system the best transport properties are found for grain-boundary free materials. Dimos et al. [2] have shown that several types of grain boundaries in YBCO bicrystals cause a reduction of J_c at the grain boundary, perhaps by a local reduction in the order parameter.

This paper will examine the effects of irradiation on oxide superconductors of the type $Tl_2Ca_2Ba_2Cu_3O_{10-\varepsilon}$ (Tl-2223) and YBCO, and in particular contrast the results from thin-film polycrystalline materials with those from single crystalline materials. One might think that the ionizing energy loss which creates defects in many other oxides would play a similar role in the superconducting oxides and thereby strongly effect their electrical properties. Yet, several previous authors [3-8] have shown that in the YBCO system the predominant mechanism creating damage is

nuclear elastic collisions. Recent work [9,10], however, has shown that the electronic energy loss component cannot be completely neglected in that very high-energy ion irradiation which has primarily electronic energy loss can effect J_c and T_c. Basic investigations of ion-irradiation effects on superconducting properties are thus important to better understand these materials as well as to develop a technique to modify the electronic and magnetic properties in beneficial ways.

The effects of irradiation on high T_c superconductors (HTS) can be thought of in terms of several different damage regimes which each have different benefits. For low fluences, ion-irradiation can play an important role in enhancing J_c through the interaction of irradiation-induced defects with the flux lattice. One of the most important areas in irradiation of HTS materials is the investigation of irradiation effects on flux pinning. The J_c in as-grown HTS crystals and films is typically limited by weak pinning energies for fluxoids in comparison to thermal energies for activating flux motion; therefore, an irradiation-induced increase in pinning can give rise to an increase in J_c. For example, low-fluence irradiation of Tl-2223 single crystals increases flux-lattice melting temperatures [11] and increases J_cs by an order of magnitude over unirradiated crystals [12]. Similarly, the effects of ion and neutron irradiation on flux pinning and critical current density in YBCO single crystals and thin films has been studied by several groups [13-16]. Umezawa et al. [13] found that neutron irradiation of YBCO single crystals doubled the magnetization critical current density (J_{cm}) to $1x10^4$ A/cm^2 in 1 T at 77 K, and Roas et al. [14] showed that 25 MeV O ion irradiation of epitaxial YBCO films on $SrTiO_3$ substrates could increase J_c from $3x10^6$ A/cm^2 to $4x10^6$ A/cm^2 in 1 T at 60 K. References [15] and [16] modelled the

effects of proton irradiation on YBCO single crystals to show that irradiation-induced defects act as fluxoid-core pinning centers.

At higher fluences both T_c and J_c can be controllably reduced [3-6,8,17] until the point where the material undergoes a superconducting to nonsuperconducting transition. In fact, the loss of superconductivity also correlates to a dramatic change in the normal state resistivity which changes from a metallic behavior to a semiconducting behavior [18]. Reports on the irradiation of YBCO films have shown that displacements by elastic collisions are predominantly responsible for the irradiation-induced decrease in the normal-state conductivity [3]. In this damage regime, ion irradiation can play an important role in producing thin-film devices (e.g., by ion-beam patterning). In fact, ion beams have been used to destroy superconductivity in 123 films in order to pattern SQUIDs [7] and also to reduce the critical current density (J_c) in order to make Josephson junction devices [8]. In this paper, we will characterize the effects of ion irradiation on the transport and magnetic properties of Tl-2223 and YBCO for both the low and high-fluence regimes.

EXPERIMENT

The Tl-Ca-Ba-Cu-O system is an important class of HTS materials because of the high T_cs and J_cs found in oriented polycrystalline films. Also, the Tl-Ca-Ba-Cu-O system contains at least five superconducting crystal structures leading to compositional tolerance in processing. The Tl-2223 phase has the highest T_c [19,20] (up to 125 K) and J_c [21] (up to 9×10^5 A/cm^2 at 76 K) of the polycrystalline thin film HTSs, and can thus

provide a basis for thin-film device development. Therefore, our work has concentrated on the Tl-2223 phase.

Thin films with a nominal metal atom ratio of $Tl_2Ca_2Ba_2Cu_3$ were prepared by sequential electron beam evaporation of the metals onto $SrTiO_3$ and $LaAlO_3$ substrates in an oxygen overpressure of $1x10^{-5}$ mbar. The film was then sintered in air with careful control of the Tl and O partial pressures, as described previously [21]. The thin-film sample thicknesses varied from 200 nm to 700 nm. These polycrystalline (pc) films contain highly oriented (c-axis normal to the substrate) rectangular grains 10 to 20 μm in size. Processed pc films have T_cs to 114 K and the pc films examined in these experiments had T_cs (zero resistance) of 99-107 K. Single crystals were grown [22] from a starting melt composition of $Tl_4Ca_3BaCu_4O_{14}$ and x-ray precession diffraction patterns were used to choose the crystals with the Tl-2223 structure. The Tl-2223 single crystals were thin plates typically 50 μm thick and about 1 mm^2 in area; and had T_cs (from Meissner measurements) of 108-115 K.

The effects of ion bombardment were studied by irradiating the samples at room temperature in increments of fluence up to a damage level which caused a loss of superconductivity. The resistivity, ρ, and the magnetization (for He and H irradiated samples) were measured at each increment of fluence as a function of temperature. In order to determine the damage mechanism, the incident ion species were varied from H to Au. The ion energies were chosen such that the ions would traverse the entire film (or crystal) and come to rest in the substrate, in order to avoid possible chemical effects in the film due to ion implantation of the incident species. Further, the ion energies were chosen to yield an approximately constant damage profile throughout the superconducting material. The range and deposited energy profiles for the bombarding ions

were determined using a Monte Carlo simulation [23] in which $Tl_2Ca_2Ba_2Cu_3O_{10}$ was approximated by $Tl_2Ba_2Cu_3Ne_{12}$ with an atomic displacement energy of 20 eV. A summary of the film thickness and irradiation conditions are given in table I.

The resistance transition temperature, $T_c(\rho=0)$, was determined from low frequency AC four-point probe measurements and the magnetization transition temperature T_c(M-onset) was determined from the Meissner effect as the temperature at which the diamagnetic signal dominated the normal state paramagnetism. The fluxoid pinning barrier and effective shielding magnetization (J_{cm}) were determined over the temperature range from 5 to 60 K by monitoring the magnetization relaxation in a field applied normal to the film (or crystalline plate). The magnetization relaxation rate (flux creep) was determined by measuring the decrease in the diamagnetic shielding signal M(t,T) versus time t at temperature T. The sample was warmed above 60 K and a field of 1 T was applied such that H_a was parallel to the c-axis. Flux motion is rapid under these conditions, and the sample quickly reaches an equilibrium vortex state. The sample was then cooled to the measurement temperature, the field was decreased to 50 mT ($H_a>H_{c1}$), and M(t,T) was measured over a time interval from 200 to 4200 seconds. A logarithmic decrease in M as a function of time was found for all temperatures (5-60 K), in agreement with thermally activated flux motion, and M can be parameterized by the following equation:

$$M(t,T) = M_0(T) + S(T)\ \ln(t), \qquad (1)$$

where M_0 is the initial effective shielding magnetization and $S=dM/d[\ln(t)]$ is the relaxation rate. The ratio of M over S can be used to determine the flux pinning energy U [24] assuming a single activation barrier:

$$U(T)/kT = -M/S + \ln(t/\tau), \qquad (2)$$

where τ is the inverse of the fluxoid hopping attempt frequency (τ is typically 10^{-9} sec [11]).

RESULTS

Irradiation-induced reduction of T_c and increase in ρ

In order to compare the decrease in T_c of the Tl-2223 pc-films for the different irradiations, the decrease in T_c normalized by T_c^o (unirradiated) was plotted (figure 1) as a function of dpa created by nuclear collisions, in a fashion analogous to that stated above for the Nb_3Sn compound. T_c/T_c^o decreases approximately linearly as a function of collisional damage although heavier ion irradiations cause a slightly more negative deviation from linearity at the higher damage levels. The rate of decrease in T_c was 5000 K per dpa which is a much faster rate than that for the A15 superconductors [17]. Each value of T_c(M-onset) for the irradiated Tl-2223 films corresponded well with the value of $T_c(\rho=0)$ for the same fluence, and both measures of T_c decreased at the same rate as a function of dpa. The quantitative agreement between the decrease in T_c for Tl-2223 measured from the transport and magnetization measurements suggests that the dominant effect of irradiation is an intragrain decrease in the superconducting order parameter rather than disruption at the grain boundaries. A fluence of 1.45×10^{16} He/cm^2 (0.020 dpa) caused complete suppression of T_c(M-onset) as measured down to 5 K.

Data for irradiated YBCO films (taken from references [3] and [4]) are also shown to the right on figure 1 in the cross-hatched area. The fact that this data appears to the right of the data for the Tl-2223 material indicates that the T_c for Tl-2223 is more strongly effected by irradiation

than the T_c for YBCO. In fact, the rate of decrease in T_c/T_c^o for the Tl-2223 films (and single crystals) is two to three times that of the YBCO films. Both sets of data from the YBCO and Tl-2223 films show the general result that the decrease in T_c/T_c^o depends predominantly on displacements created by nuclear collision events. The Au ions (nuclear stopping power ≈ 55 eV/A) incident on Tl-2223 have a greater fraction of their energy loss in the form of nuclear collisions than the He ions (nuclear stopping power ≈ 0.022 eV/A) by more than three orders of magnitude. Similarly, the data for the irradiation of the YBCO films cover approximately two orders of magnitude in nuclear stopping power (0.22 eV/A to 19 eV/A). Still, the rate of decrease in T_c/T_c^o for the different irradiating species as a function of dpa from atomic collisions is nearly equal.

The predominant dependence on collisional damage has also been observed [25,26] for changes in the normal state resistivity (ρ) of both YBCO and Tl-2223 thin films. The increase in the room temperature ρ compared to the unirradiated state [$\Delta\rho=\rho_{RT}$(irradiated)-ρ_{RT}(unirradiated)] approximately scales with the collisional damage level at low fluence levels. However as the defect level in the films increased, the change in ρ_{RT} increased more rapidly than that for a simple linear relationship. Initially, $\Delta\rho_{RT}$ increases approximately linearly with deposited energy for the Tl-2223 films up to a damage level of 0.007 dpa, but for higher damage levels the resistivity increase is superlinear. A linear dependence of $\Delta\rho$ on deposited energy would be expected for metallic-like behavior in which the resistivity increases proportionally to the increase in carrier scattering centers (defects created by ion irradiation), assuming the carrier concentration is not altered. Detailed studies of the resistivity in YBCO materials [25,18] have shown that the increase after ion-irradiation is more complex than a simple linear behavior, even at low

fluences, and that a change in the carrier concentration is not sufficient to explain the change in the resistivity. This deviation from linearity greater than expected from a change in carrier concentration may be related to the film microstructure and the creation of a percolation network (such as small regions of insulating material imbedded in a metal matrix) or to a steady change to a more semiconducting-like material. When Tl-2223 samples were irradiated with oxygen ions to 0.028 dpa, a level at which superconductivity was completely suppressed, the ρ-T data exhibited semiconducting behavior at low temperatures.

Both sets of data for Tl-2223 and YBCO in figure 1 (indicated by cross-hatched and dotted background areas) show deviations from linearity as a function of dpa and a range of T_c/T_c^o reduction rates for different ion beams. These results may reflect the variations in the density of the defects in the cascades. A three-dimensional Monte Carlo analysis [26] of the cascades created by 20 MeV Au and 2 MeV He irradiation of Tl-2223 showed that the cascades for the 2 MeV He ions generally do not progress beyond one secondary event whereas the cascades for the 20 MeV Au ions do produce defects from higher order events. The lighter target atoms are recoiled farther than the heavier target atoms for both Au and He irradiation. The light target atoms recoiled by the He ions come to rest at a distance of 1-1.5 nm from the point of vacancy production and the heavy target atoms come to rest at a distance of 0.2-0.4 nm from the point of vacancy production. In comparison, the light atoms recoiled by Au ions travel from 2 nm to 10's of nm before coming to rest and the heavier atoms recoiled by the Au ions can travel 1-8 nm before coming to rest. Therefore, a heavier ion with many secondary cascades would be expected to show a more rapid reduction rate as small insulating areas in each track begin to overlap. High resolution transmission electron microscopy of ion and

neutron irradiated YBCO [27,28] has shown the production of isolated homogeneously distributed defect clusters.

In order to separate-out the non-linear effects at higher dpa, the initial rate of decrease in T_c as a function of fluence (Φ) can be examined where the slopes of $dT_c/d\Phi$ are approximately linear. The damage level in dpa is, to first order: $dpa = S_n\Phi/D$; where S_n is the nuclear stopping power (approximately constant throughout the material), and D is the atomic density. Summers et al. [29] have demonstrated that $dT_c/d\Phi$ is directly proportional to the nuclear stopping power over seven orders of magnitude. Their calculation of S_n included only the effects of primary knock-on atoms, but for the low fluence regime the secondary cascades are expected to be less important to the reduction in T_c. Figure 2 shows this proportionality holds true for electron, proton, and heavy ion irradiation of YBCO with a proportionality constant of $\approx 4x10^{-21}$ K-g/eV ($\approx$ 2600 K/dpa for YBCO). This constant is in good agreement with the YBCO results shown above. This figure also shows that $dT_c/d\Phi$ for Tl-2223 is greater than that for YBCO but $dT_c/d\Phi$ for Tl-2212 is nearly the same (or slightly less) than that for YBCO.

A striking contrast to the results presented in figures 1 and 2 are the recent results [9,10] from high-energy (several MeV/amu) heavy-ion irradiation of YBCO in which the electronic energy loss (S_e) was shown to contribute to the reduction in T_c and increase in ρ when S_e surpasses a threshold value. Figure 3 is data taken from [9] and shows that irradiation of YBCO by 3.5 GeV Xe ions at 105 K causes a substantial decrease in T_c for a fluence regime where the number of displacements from nuclear energy deposition are small. For this energy, S_e ($\approx$ 2100 eV/A) is more than three orders of magnitude greater than S_n. This behavior was explained in terms of the layer structure of the HTS material in which some layers (with more

ionic behavior) are more sensitive to ionization processes while the other layers (with more metallic behavior) depend primarily on nuclear collision processes. Figure 3 also shows the effect on J_c of 3.5 GeV Xe ion irradiation at 300 K. This is typical behavior for irradiated superconductors in that as the damage level increases for low fluences J_c increases, and at higher fluences the degradation in T_c causes a degradation in J_c.

Thermal recovery of ion-beam damage

Another effect which can cause deviations from the expected rate of decrease of T_c/T_c^o and rate of increase of ρ as a function of dpa is thermal recovery of defects during irradiation. Significant recovery of ρ was found [5,25] for ion-irradiated YBCO films at low temperatures and earlier results [30] from Tl-2223 films showed that both ρ_{RT} and T_c recovered by 3-5% as a result of mobile defects at 300 K. Several factors can influence the amount of annealing which can occur: the temperature of the anneal, different defect densities from a single cascade can lengthen the distance over which defects must diffuse in order to annihilate damage, and increased flux densities can increase the amount of radiation enhanced diffusion.

Understanding the rate and mechanisms of damage recovery are important in device processing and material applications; e.g., a sample irradiated with the Au ion beam to a damage level of 0.010 dpa was annealed for 15 min. at 600°C and nearly recovered its original ρ-T characteristics. In order to get a measure of the activation energy for the damage recovery, a detailed examination of the recovery of ρ was done [26]. Samples from adjacent sections of a thin film were irradiated with 500 keV He ions to a

level 0.001 dpa for temperatures from 100 K to 375 K, and the recovery of ρ as a function of time was monitored in-situ immediately after the irradiating beam was stopped. Annealing of defects did not occur in measurable time below 250 K; above 375 K the oxygen loss from the vacuum annealing became significant. Therefore the temperature range of the experiment was limited to 275-375 K.

For metallic-like behavior in the normal state, the increased damage would yield an increase in the number of carrier scattering centers. Assuming the carrier density is not drastically changing for this low damage level, then ρ should increase in proportion to dpa. By monitoring the rate of decrease in ρ as a function of time after the irradiation, the annealing of damage can then be monitored. If the kinetics of the recovery process are diffusion controlled then the fraction (f) of material which remains in the defective state at time t is given by [31]: $f=\exp[-(t/\tau)^n]$, where τ is the time constant characteristic of the annealing process and n is the order of the kinetics. The value of f can be determined from the resistivity by the equation: $f = \rho(t)-\rho(0)/(\rho^o-\rho(0)$, where $\rho(0)$ is ρ at time zero after irradiation and ρ^o is ρ for the sample before irradiation. Simple thermal annealing of defects by random diffusion to a uniform distribution of sinks would follow first-order reaction kinetics, n=1. The activation energy (Q) for the recovery process can be determined from the temperature behavior of τ. An Arrhenius behavior (figure 4) for $\tau(T)$ was observed which is characteristic of a thermally activated process with a single activation energy = 0.36 ± .05 eV. This small value for Q is consistent with the idea that a defect formation energy is not needed under irradiation conditions, and it indicates that a relatively small energy is needed for atomic motion.

Flux Pinning Energy in Tl-2223

As crystalline quality of thin-film HTS materials improves, the pinning barrier has been found to decrease and therefore the use of ion irradiation to controllably introduce pinning centers is desirable. (An increase in pinning at temperatures approaching 77 K is of particular importance.) Figure 5 shows the flux pinning energy for a Tl-2223 single crystal irradiated with 4.5 MeV protons and Tl-2223 pc-film irradiated with 300 keV He^+ as a function of temperature. The room temperature ion irradiation significantly increased pinning (≈25% at 25 K) in the single crystal over the entire temperature range, whereas the pinning in the pc-thin film is increased by 10-15% only in the temperature range from 10 to 20 K (and is unaffected above approximately 25 K). While a fluence of $1x10^{16}$ H/cm^2 (damage level of $4x10^{-4}$ dpa) caused the pinning energy in the single crystal to increase by 20% or more, continued irradiation to $2x10^{16}$ H/cm^2 did not increase pinning above a temperature of 15 K. In fact, the rate of increase in irradiation-induced pinning in the single crystal appears to saturate (with increasing damage levels) at approximately the pinning energy which existed in the unirradiated pc-thin films.

The increased pinning for the pc-thin film at temperatures between 10 and 20 K (and the further increase in pinning for the single crystal below 15 K) suggests that the pinning mechanisms at higher temperatures are dominated by pre-existing pinning centers which are stronger than those introduced by continued ion irradiation. Further increases in pinning energy at higher temperatures would then require the introduction of a stronger pinning center (e.g., with an activation barrier greater than 150 meV above 50 K).

Critical Current Density

The strength of the diamagnetic shielding signal M(t,T) is proportional to the critical current, and under the conditions described for the flux creep measurement the sample is in the critical state with essentially uniform J_c in the a-b plane. For relative comparisons of ion irradiation on critical current density, M(t=200, T) was used to determine the magnetization critical current density, $J_{cm}(T)$, in a field of 50 mT as shown in figures 6 and 7. J_{cm} for the unirradiated thin-film sample (Fig. 7) decreased a factor of 17 with increasing temperature from 5 K to 50 K, and changed very little after ion irradiation, as expected from the result for the pinning energy of the film.

In contrast, the single crystal J_{cm}(irradiated) is a factor of 2 times greater than J_{cm}(unirradiated) at 5 K, and the enhancement of J_{cm} after irradiation is even more dramatic at higher temperatures as a result of the different rates of decrease in J_{cm} with temperature. For example, J_{cm}(irradiated) is an order of magnitude higher than J_{cm}(unirradiated) at 25 K. (These data are in good agreement with isothermal magnetization hysteresis loop determinations of J_{cm} in crystals before and after ion irradiation.) The magnetization critical current density for the unirradiated crystal decreased by a factor of 25 over a temperature range from from 5 K to 25 K; whereas, the rate of decrease in J_{cm} for the crystal irradiated to a damage level of 4×10^{-4} dpa is lowered such that J_{cm} only decreased by a factor of 25 over a temperature range from 5 K to 50 K. Further irradiation of the sample to 8×10^{-4} dpa caused only a slight increase in J_{cm}.

As stated above, ion irradiation will become increasingly important for controlling flux pinning as the quality of HTS materials increases. Figure 7 shows J_{cm} decreases in Tl-2223 films as a function of applied magnetic field at a temperature of 25 K. Before irradiation, the decrease in J_{cm} is much faster for the more weakly pinned millimeter-size epi-grain film (epitaxial over millimeter-size regions of the substrate) than for the polycrystalline film (two orders of magnitude smaller grain size). Irradiation of the pc film had a negligible effect on U and J_{cm}. In contrast, ion irradiation substantially enhanced J_{cm} for this epi-film particularly at higher applied fields where the flux-motion driving force is stronger, although irradiation did not increase J_{cm} of the better quality epi-film up to that of the pc film. Therefore, ion irradiation will find use as a technique to enhance pinning and J_c in selective regions of high quality thin-film high T_c superconductors.

CONCLUSIONS

Ion irradiation of HTS can be used for different beneficial results in different fluence regimes. Low-fluence ion irradiation can be used to significantly enhance the pinning energy and critical current density (with only a modest decrease in T_c) for low defect density material, such as unirradiated Tl-2223 single crystals. This method for enhanced pinning is also becoming more important for thin films as the crystalline quality of the material increases. High-fluence ion irradiation can be used to controllably decrease T_c and thus provides a technique for patterning HTS devices. The rate of decrease in T_c is a material property which is independent of the grain morphology and depends primarily on nuclear energy

loss, except for MeV/amu energy ions which have a significant electronic energy loss component. The decrease in T_c(M-onset) determined from Meissner measurements showed the suppression of T_c occurs throughout the superconducting material rather than at discrete isolated regions such as grain boundaries. Finally, the defects introduced through ion irradiation were shown to be highly mobile with a damage-recovery activation energy of 0.36 eV.

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REFERENCES

[1] S. T. Sekula, J. Nucl. Materials 72, 91 (1978), and references therein.

[2] D. Dimos, P. Chaudari, J. Mannhart, and F.K. LeGoues, Phys. Rev. Lett 81, 219 (1988).

[3] G. J. Clark, A. D. Marwick, R. H. Koch, and R. B. Laibowitz, Appl. Phys. Lett. 51, 139 (1987).

[4] 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).

[5] G. C. Xiong, H. C. Li, G. Linker, and O. Meyer, Phys. Rev. B 38, 240 (1988).

[6] G. P. Summers, E. A. Burke, D. B. Chrisey, M. Nastasi, and J. R. Tesmer, Appl. Phys. Lett. 55, 1469 (1989).

[7] R. H. Koch, C. P. Umbach, G. J. Clark, P. Chaudari, and R. B. Laibowitz, Appl. Phys. Lett. 51, 200 (1987).

[8] A. E. White, K. T. Short, R. C. Dynes, A. F. J. Levi, M. Anzolwar, K.W. Baldwin, P. A. Polakos, T. A. Fulton, and L. N. Dunkleberger, Appl. Phys. Lett. 53, 1010 (1988).

[9] D. Bourgault, S. Bouffard, M. Toulemonde, D. Groult, J. Provost, F. Studer, N. Nguyen, and B. Raveau, Phys. Rev. B39, 6549 (1989).

[10] B. Hensel, B. Roas, S. Henke, R. Hopfengartner, M. Lippert, J. P. Strobel, M. Vildic, G. Saemann-Ischenko, and S. Klaumunzer, Phys. Rev. B42, 4135 (1990).

[11] E. L. Venturini, J. C. Barbour, D. S. Ginley, R. J. Baughman, and B. Morosin, Appl. Phys. Lett. 56, 2456 (1990).

[12] J. C. Barbour, E. L. Venturini, and D. S. Ginley, Proceedings of the 1990 Ion-Beam Modification of Materials Conference, Knoxville, TN, September 9-14, 1990.

[13] 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).

[14] B. Roas, B. Hensel, G. Saemann-Ischenko, and L. Schultz, Appl. Phys. Lett. 54, 1051 (1989).

[15] L. Civale, A. D. Marwick, M. W. McElfresh, T. K. Worthington, A. P. Malozemoff, F. H. Holtzberg, J. R. Thompson, and M. A. Kirk, Phys. Rev. Lett. 65, 1164 (1990).

[16] R. B. van Dover, E. M. Gyorgy, A. E. White, L. F. Schneemeyer, R. J. Felder, J. V. Waszczak, Appl. Phys. Lett. 56, 2681 (1990).

[17] O. Meyer, in Studies of High Temperature Superconductors Vol. 3, ed. by A. V. Narlikar (Nova Science, New York, 1989).

[18] J. M. Valles, Jr., A. E. White, K. T. Short, R. C. Dynes, J. P. Garno, A. F. J. Levi, M. Anzlowar, K. Baldwin, Phys. Rev. B39, 11599 (1989).

[19] S. S. P. Parkin, V. Y. Lee, E. M. Engler, A. I. Nazzal, T. C. Huang, G. Gorman, R. Savoy, and R. Beyers, Phys. Rev. Lett. 60, 2539 (1988).

[20] Z. Z. Sheng and A. M. Hermann, Nature 332, 138 (1988).

[21] D. S. Ginley, J. F. Kwak, E. L. Venturini, B. Morosin, and R. J. Baughman, Physica C 160, 42 (1989).

[22] D. S. Ginley, B. Morosin, R. J. Baughman, E. L. Venturini, J. E. Schirber, and J. F. Kwak, J. Crystal Growth 91, 456 (1988).

[23] J. F. Ziegler and J. P. Biersack, The Stopping and Range of Ions in Solids, (Pergamon, New York, 1985); see also TRIM87 computer code by J. F. Ziegler, IBM Yorktown Heights, New York.

[24] C. W. Hagen and R. Griessen in Studies of High Temperature Superconductors, Vol. 3. ed. by A. V. Narlikar (Nova Science, New York, 1989), pp. 159-195.

[25] A. D. Marwick, G. J. Clark, D. S. Yee, R. B. Laibowitz, G. Coleman, and J. J. Cuomo, Phys. Rev. B 39, 9061 (1989).

[26] J. C. Barbour, E. L. Venturini, J. F. Kwak, D. S. Ginley, R. J. Baughman, and D. K. Brice, submitted to J. Appl. Phys.

[27] M-O. Ruault, M. Barrachin, J. Lesueur, L. Dumoulin, H. Bernas, M. Gasgnier, J. C. Levet, H. Noel, R. Gougeon, and M. Potel, Physica C 162-164, 77 (1989).

[28] M. A. Kirk, M. C. Frischherz, J. Z. Liu, L. L. Funk, L. J. Thompson, E. A. Ryan, S. T. Ockers, and H. W. Weber, Physica C 162-164, 532 (1989).

[29] G. P. Summers, B. D. Weaver, E. Jackson, J. M. Pond, H. Newman, D. B. Chrisey, J. S. Horwitz, E. A. Burke, Proceedings of the IEEE Nuclear and Space Radiation Effects Conference, San Diego, 1991.

[30] J. C. Barbour, J. F. Kwak, D. S. Ginley, and P. S. Peercy, Appl. Phys. Lett. 55, 507 (1989).

[31] J. W. Christian, The Theory of Transformations in Metals and Alloys Part I, 2nd. ed. (Pergamon, New York, 1975), p. 542.

TABLE I

Ion	E (kev)	Thickness (nm)	R_p* (nm)	Φ(dpa=.001)* ions/cm^2	Inc. Angle
			Tl-2223 FILMS		
He	300	300	870	1.3×10^{14}	5°
He	2000	700	3280	7.1×10^{14}	45°
O	740	200	560	7.1×10^{12}	10°
Au	20000	700	1760	2.9×10^{11}	5°
			Tl-2223 CRYSTAL		
H	4500	50000	72000	2.4×10^{16}	5°

* R_p=Projected Range; Φ=incident ion fluence

FIGURE CAPTIONS

Fig. 1. The transition temperature (T_c) normalized to the unirradiated state (T_c^o) as a function of damage from atomic collisions in Tl-2223 and YBCO. The YBCO data were taken from [3] and [4]. The symbols for the Tl-2223 films correspond to the following irradiation conditions: Δ=20 MeV Au ions; O=740 keV O ions; and +=2 MeV He ions. The symbols for the YBCO films correspond to the following irradiation conditions: X=500 keV O ions [3]; diamond=2 MeV Ar ions [4]; stars=1 MeV Ne ions [4]; filled square=800 keV N ions [4]; and filled circle=3.5 MeV Be ions [4].

Fig. 2. The rate of decrease in T_c with fluence (Φ) scales with energy loss due to nuclear stopping over seven orders of magnitude. This plot was taken from [29].

Fig. 3. The reduced transition temperature T_c/T_c^o (left axis) for a YBCO sample irradiated by 3.5 GeV Xe ions at 105 K as a function of fluence. The corresponding magnetization critical current J_c/J_c^o (right axis) as a function of Xe fluence. (Data taken from Bourgault [9])

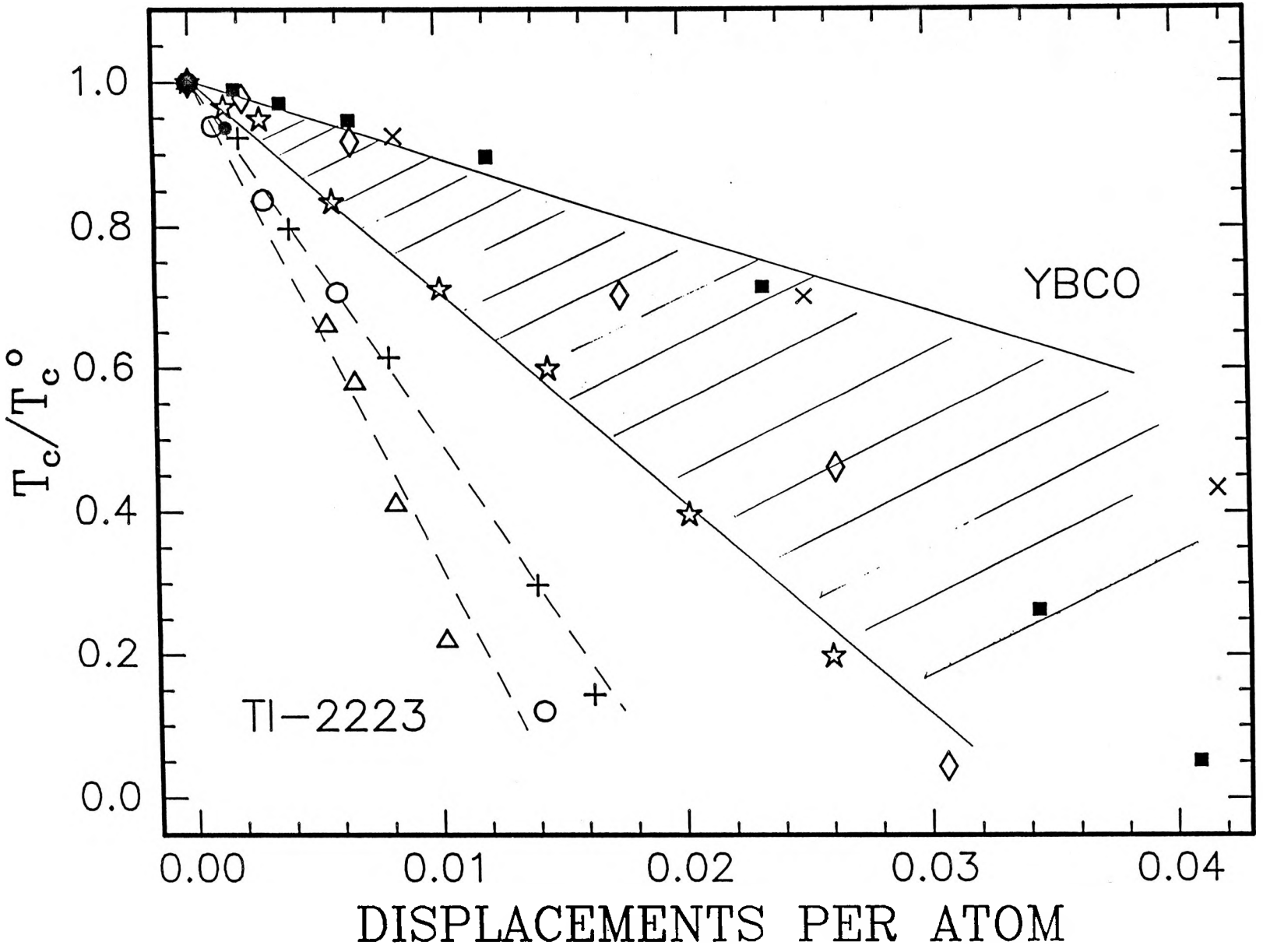
Fig. 4. The damage-recovery time constant (τ) for annealing of irradiation damage was determined from the time rate of change in ρ at temperature T immediately after the ion beam was stopped. Q is the activation energy for the recovery process.

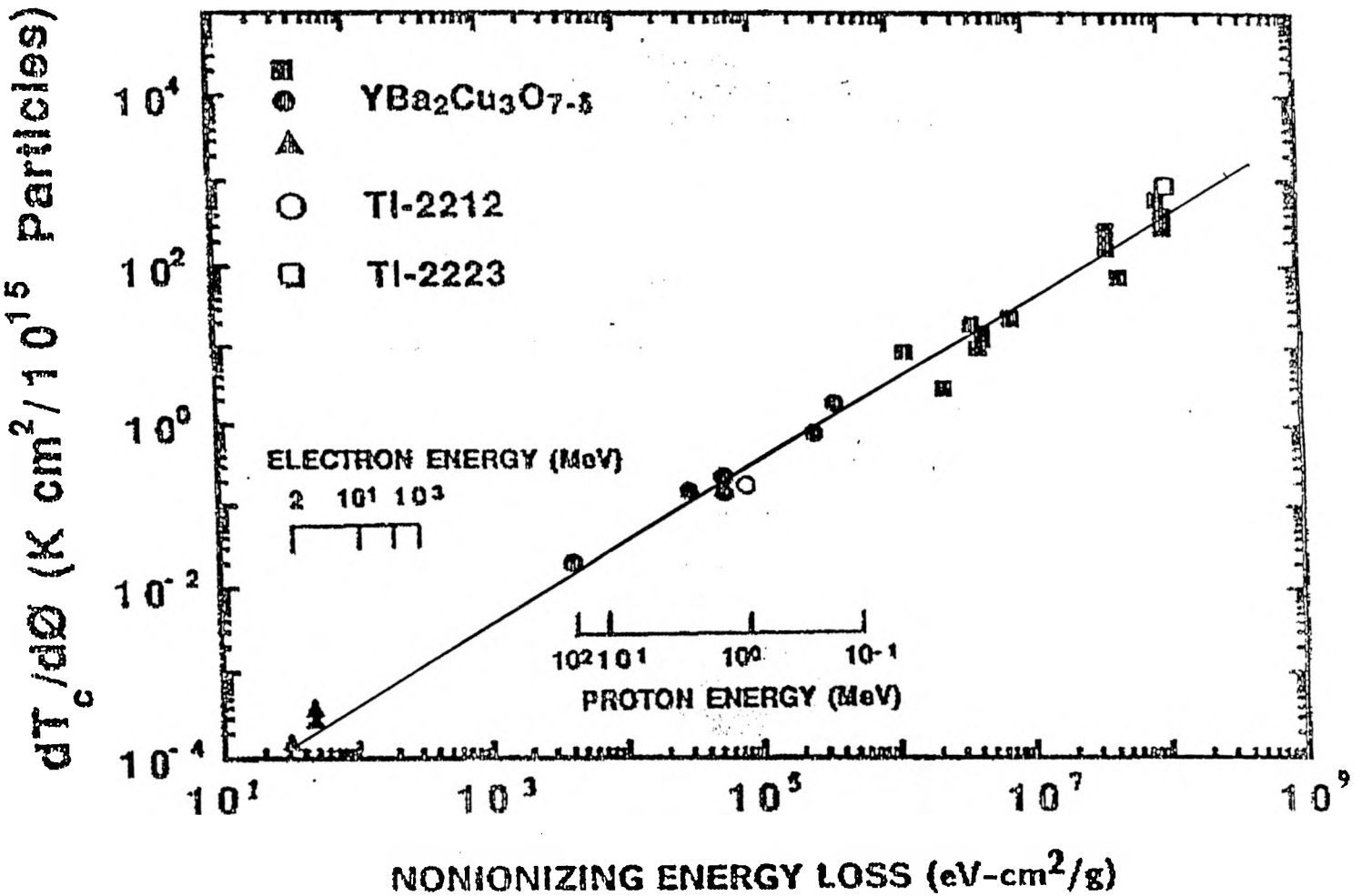
Fig. 5. The flux pinning potential as a function of temperature for a 50 μm thick Tl-2223 single crystal before and after irradiation with 4.5 MeV

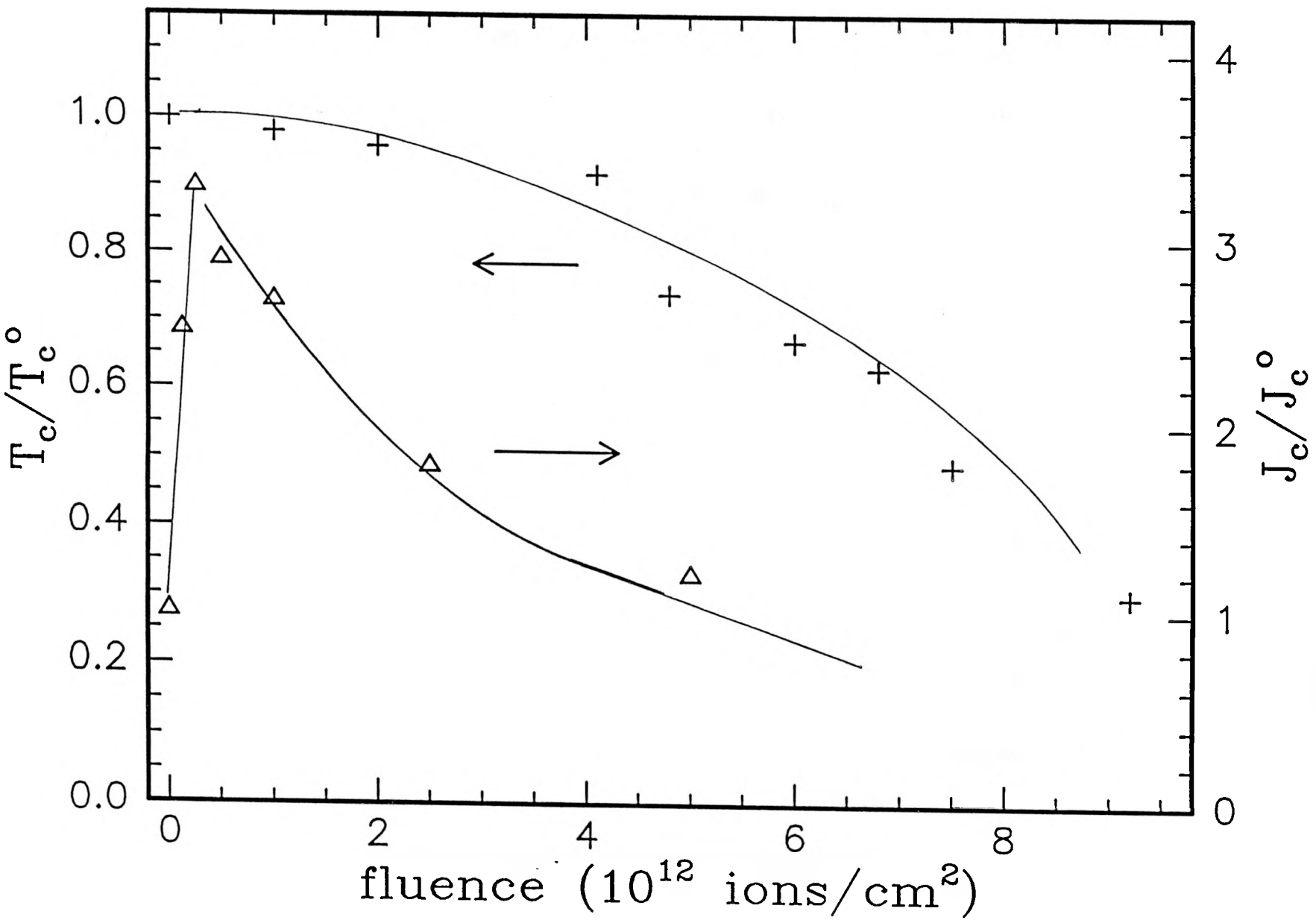
protons and for a 300 nm thick Tl-2223 film before and after irradiation with 300 keV He^+.

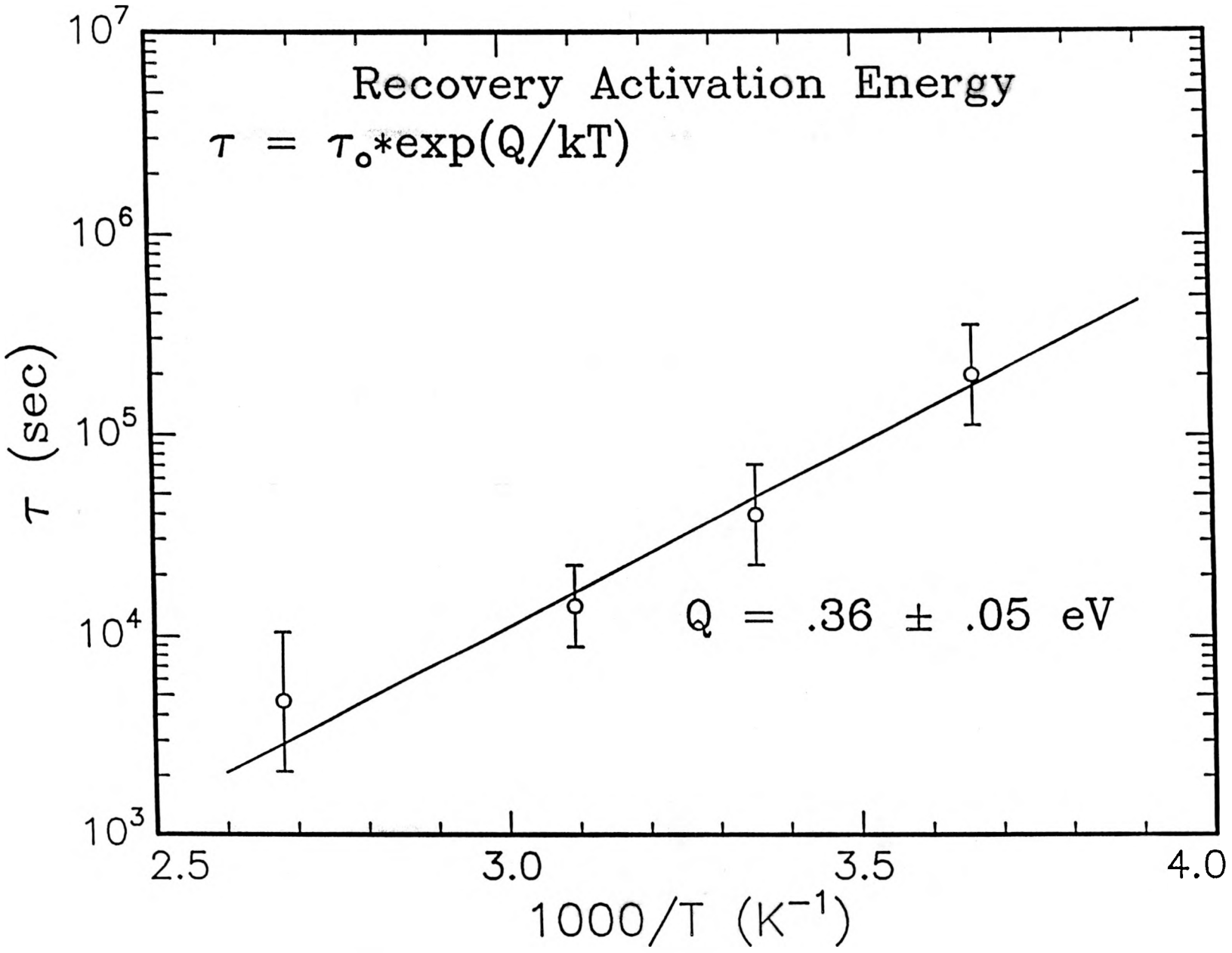
Fig. 6. Irradiation enhancement of the magnetization critical current density for a Tl-2223 single crystal (determined from the initial value of magnetization in flux creep data).

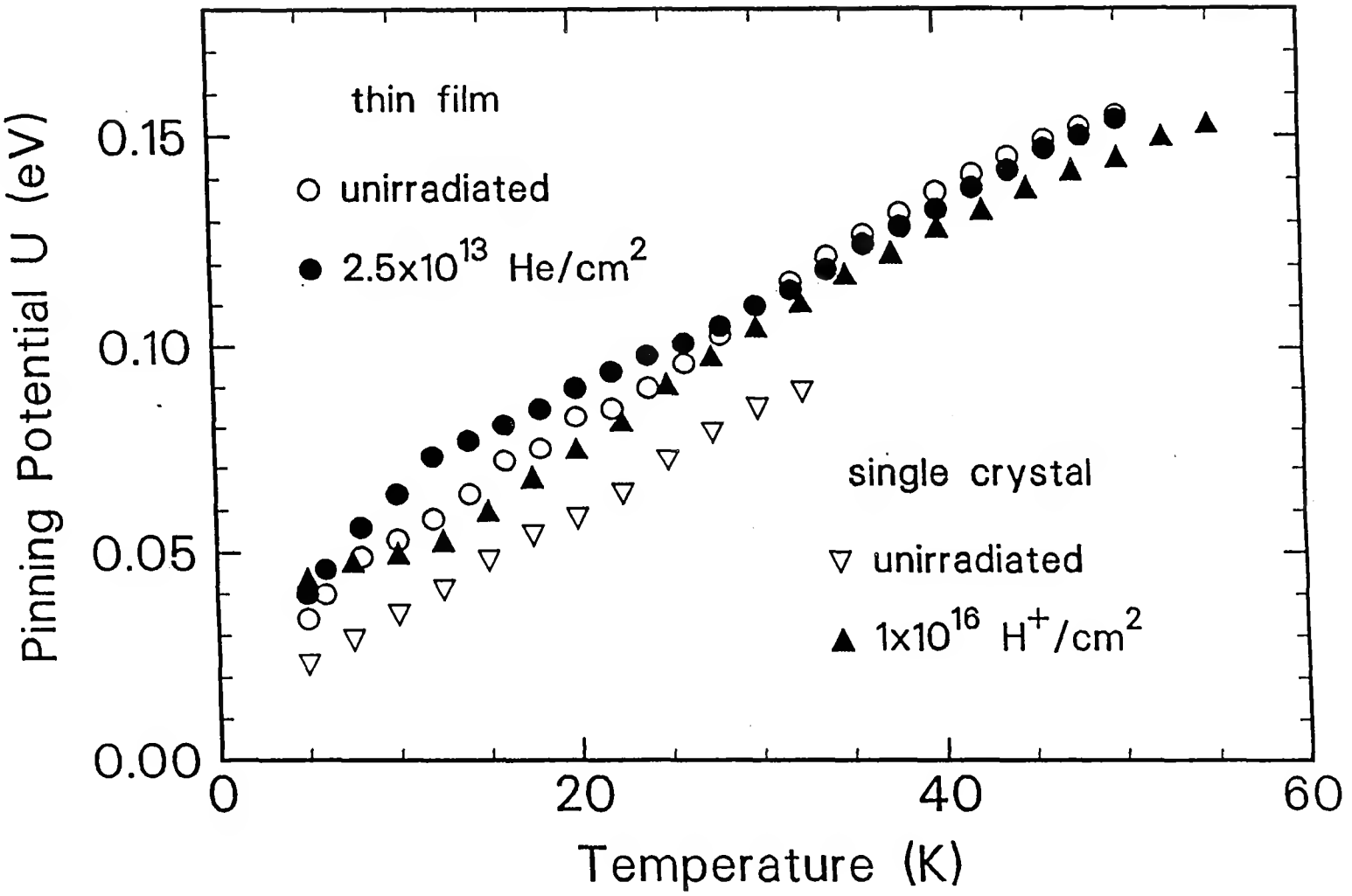
Fig. 7. The magnetization critical current density (J_{cm}) as a function of applied magnetic field was determined from hysteresis loops. The "poly" film has a grain size of 10 μm while the "epi" film was epitaxial over millimeter-size regions of the substrate.

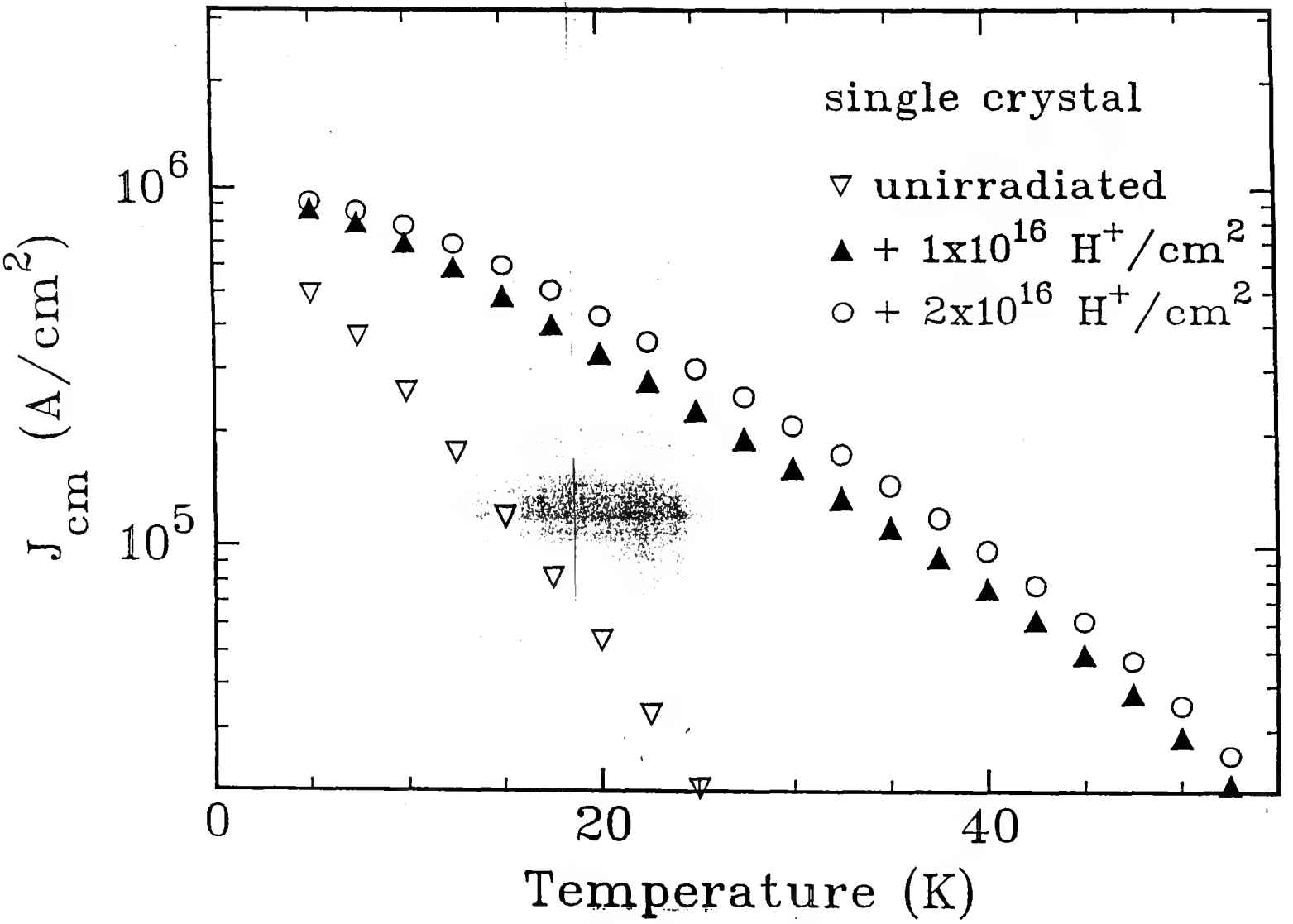

1.0
0.8
0.6
0.4
0.2
0.0
$T_c/T_c°$
YBCO
Tl−2223
0.00
0.01
0.02
0.03
0.04
DISPLACEMENTS PER ATOM

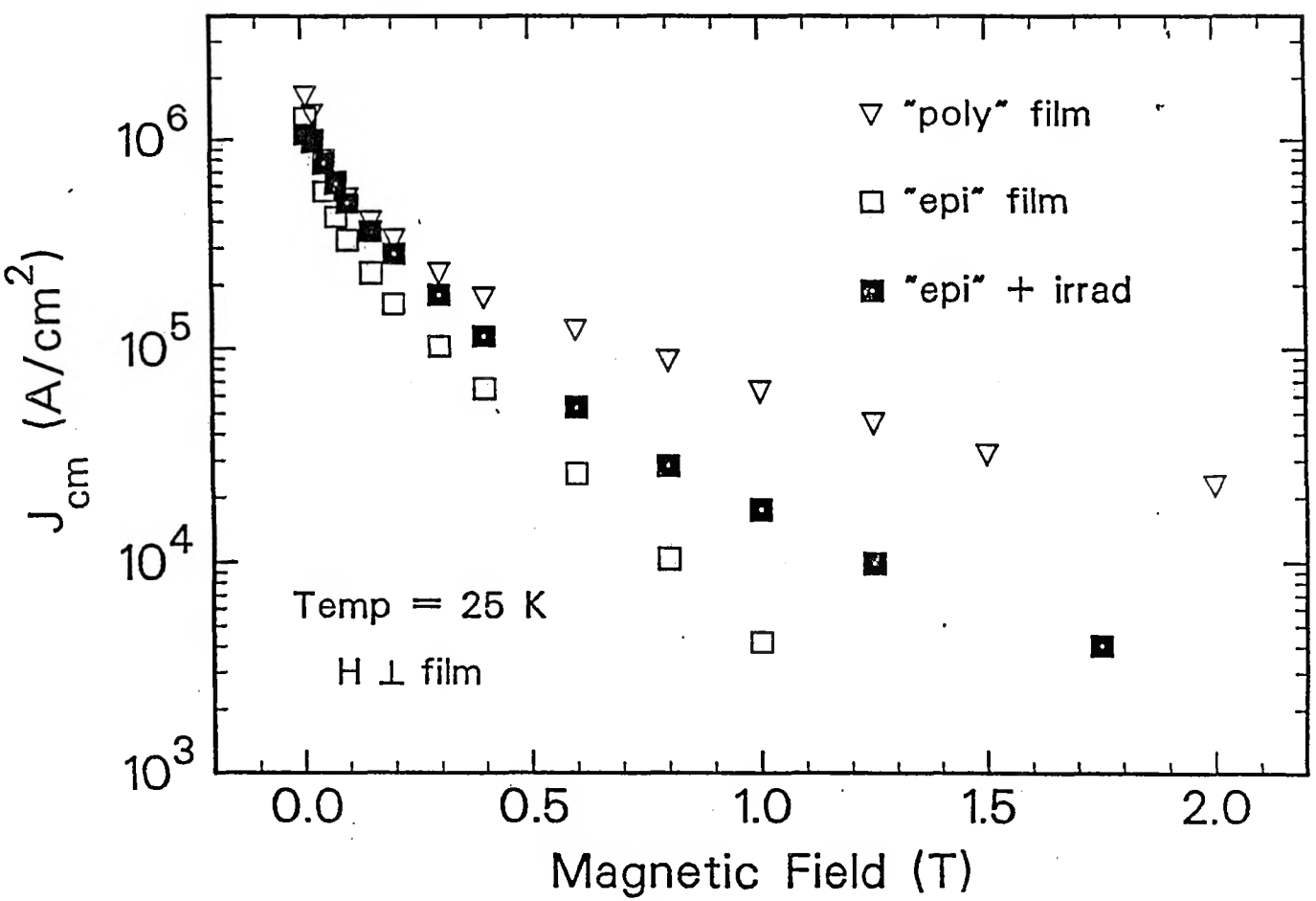

$dT_c/d\Phi$ (K cm^2/10^{15} Particles)
10^4
10^2
10^0
10^{-2}
10^{-4}
$YBa_2Cu_3O_{7-\delta}$
Tl-2212
Tl-2223
ELECTRON ENERGY (MeV)
2 10^1 10^3
10^2 10^1 10^0 10^{-1}
PROTON ENERGY (MeV)
10^1
10^3
10^5
10^7
10^9
NONIONIZING ENERGY LOSS (eV-cm^2/g)


1.0
0.8
0.6
0.4
0.2
0.0
$T_c/T_c°$
0
2
4
6
8
fluence (10^{12} ions/cm^2)
4
3
2
1
0
$J_c/J_c°$


Recovery Activation Energy
$\tau = \tau_o * \exp(Q/kT)$
$Q = .36 \pm .05$ eV
τ (sec)
10^7
10^6
10^5
10^4
10^3
2.5
3.0
3.5
4.0
1000/T (K^{-1})


thin film
unirradiated
2.5x10^13 He/cm^2
single crystal
unirradiated
1x10^16 H+/cm^2
Pinning Potential U (eV)
0.00
0.05
0.10
0.15
0
20
40
60
Temperature (K)


single crystal
unirradiated
+ 1x10^16 H+/cm^2
+ 2x10^16 H+/cm^2
10^6
10^5
J_cm (A/cm^2)
0
20
40
Temperature (K)


10^6
10^5
10^4
10^3
J_{cm} (A/cm^2)
0.0
0.5
1.0
1.5
2.0
Magnetic Field (T)
"poly" film
"epi" film
"epi" + irrad
Temp = 25 K
H ⊥ film