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IRRADIATION-INDUCED PINNING IN SINGLE-CRYSTAL AND THIN-FILM $Tl_2Ca_2Ba_2Cu_3O_{10}$ SUPERCONDUCTORS

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Abstract

Magnetization relaxation measurements were used to directly measure the effects of ion irradiation on magnetic flux pinning. The pinning potential (U) in a $Tl_2Ca_2Ba_2Cu_3O_y$ single crystal irradiated with 4.5 MeV protons, to a damage level of 4×10^{-4} dpa, increased by ≈25% at 25 K. In contrast, U for a highly oriented polycrystalline thin film irradiated with 300 keV He ions to a damage level of 2×10^{-4} dpa increased by only 5% at 25 K (and less at higher temperatures). Enhancement of the magnetization critical current density (J_{cm}) is determined as a function of temperature and damage in the single crystal and thin film. High energy proton irradiation increased J_{cm} in the single crystal by an order of magnitude at 25 K but had only a small effect on J_{cm} in the thin film.

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One of the most important aspects of ion-beam modification of high temperature superconductors (HTSs) is the possibility of enhancing the critical current density (J_c). Defects introduced from ion irradiation can act as pinning centers for magnetic flux lines or vortices. The critical current density in as-grown HTSs is typically limited by weak pinning energies for these 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. Our previous work [1] showed that high-energy proton irradiation of a $Tl_2Ca_2Ba_2Cu_3O_y$ (Tl-2223) single crystal reduced the magnetic flux creep and quadrupled the pinning energy at 20 K.

The effects of ion and neutron irradiation on flux pinning and critical current density in $YBa_2Cu_3O_{7-x}$ (YBCO) single crystals and thin films has been studied by several groups [2-5]. Umezawa et al. 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. 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 4 and 5 modelled the effects of proton irradiation on YBCO single crystals to show that irradiation-induced defects act as fluxoid-core pinning centers. In this paper, we will compare the effects of ion irradiation on Tl-2223 single crystals and thin films, and examine the relative increases in J_c.

Experiment

The Tl-Ca-Ba-Cu-O system is an important class of HTS materials because of the high superconducting transition temperatures (T_cs) and J_cs found in oriented polycrystalline films. In particular, the Tl-2223 phase has T_c values up to 125 K [6] and J_c values up to 9×10^5 A/cm^2 at 76 K [7]. The thin film employed for this experiment was a 300 nm thick highly-oriented polycrystalline Tl-2223 film on a 3 mm x 3 mm square $LaAlO_3$ substrate. The Tl-2223 single crystal was a thin plate 50 μm thick and 0.7 mm^2 in area. The film was prepared by sequential electron beam evaporation of the elements onto the substrate in an oxygen overpressure of approximately 1×10^{-5} mbar. The film was then sintered in air with careful control of the Tl and O partial pressures, as described previously [7]. Processed films had T_cs to 114 K and the thin film examined in this experiment had a T_c of 107 K. The single crystal was grown [8] from a starting melt composition of $Tl_4Ca_3BaCu_4O_{14}$ and an x-ray precession diffraction pattern was used to choose a single crystalline plate with the Tl-2223 structure.

The effects of low- and high-fluence ion irradiation on the Meissner effect and resistivity behavior of thin-film Tl-2223 on $SrTiO_3$ substrates were previously studied [9,10]. These results showed that the damage mechanism in these films is predominantly due to nuclear collisions which causes a decrease in T_c. Further, the quantitative agreement between the decrease in T_c measured from the transport and magnetization measurements suggested that the dominant effect of irradiation is a decrease in the superconducting order

parameter. The rate of decrease in T_c was 1 K per $2x10^{-4}$ dpa. Both the thin film on $LaAlO_3$ and the single crystal used in this experiment exhibited a similar rate of decrease in T_c.

In order to model the previous experiments in which a uniform level of damage was produced throughout the film, the single crystal was irradiated at room temperature with 4.5 MeV protons which have a projected range of 72 μm in Tl-2223. Only 0.1% of the protons come to rest in the crystal (primarily in the back third of the crystal). Monte Carlo simulations [11] show that a fluence of $4.7x10^{15}$ H/cm^2 yield a damage level of $2x10^{-4}$ dpa in the crystal. Since the concentration of defects is 10^4 times greater than the concentration of implanted protons, chemical doping effects are thought to be negligible in comparison to ion-beam damage effects. Magnetic flux creep measurements (described below) were made as a function of temperature before and after irradiation in order to determine the pinning energy and the effective shielding magnetization. The single crystal was irradiated to the following fluences: $4.7x10^{15}$ H/cm^2, $1x10^{16}$ H/cm^2, and $2x10^{16}$ H/cm^2. Similarly, the thin film sample was irradiated with 300 keV He^+ ions to a fluence of $2.5x10^{13}$ He/cm^2 in order to create a damage level of $2x10^{-4}$ dpa. The range of these He ions is 870 nm.

Magnetization Relaxation and Pinning Energies

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 the 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 [12] 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 [1]).

Figure 1a and 1b show the flux pinning energy for the single crystal and thin film, respectively, as a function of temperature calculated using $\tau=10^{-9}$ sec and t=200 sec in equation 2. Comparing the unirradiated state (open triangles) to the irradiated state (closed triangles) shows that room temperature ion irradiation can significantly increase pinning (≈25% at 25 K) in the single crystal over the entire temperature range. In contrast, the pinning in the 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 1×10^{16} H/cm^2 (damage level of 4×10^{-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. Further, the magnitude and the dependence of pinning energy on temperature are nearly equal for the irradiated single crystal and the unirradiated thin film (which differs little from the irradiated film behavior). Therefore, 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 thin films.

The increased pinning for the 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).

Magnetization Critical Current Density

The strength of the diamagnetic shielding signal M(t,T) is proportional to the superconducting current flow. Under the conditions described for the flux creep measurement, the sample is in the critical state with essentially uniform critical current density in the a-b plane. Hence, we can use M(t=200, T) with the appropriate geometrical factors [15] to determine the magnetization

critical current density, $J_{cm}(T)$, in a field of 50 mT for relative comparisons of J_{cm} as a function of ion irradiation.

Figures 2a and 2b show the dependence of J_{cm} on temperature as a function of ion irradiation. The magnetization critical current density for the unirradiated thin-film sample (Fig. 2b) decreases a factor of 17 with increasing temperature from 5 K to 50 K. As with the pinning energy, J_{cm} is changed very little after ion irradiation to a damage level of $2x10^{-4}$ dpa. 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 decreases by a factor of 25 in 20 K (from 5 K to 25 K); whereas, the rate of decrease in J_{cm} for the crystal irradiated to a damage level of $4x10^{-4}$ dpa is lowered such that J_{cm} only decreases by a factor of 25 over a temperature range from 5 K to 50 K. Further irradiation of the sample to a level of $8x10^{-4}$ dpa causes a slight increase in J_{cm}, however it is plausible to significantly change the rate of decrease in J_{cm} at higher temperatures if stronger pinning centers could be introduced into the sample.

Conclusion

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 Tl-2223 material, such as unirradiated single crystals. However, a large enhancement of the flux pinning in a defective structure requires pinning centers with energy barriers greater than those introduced by ion irradiation. A strong pinning center with a pinning barrier greater than 150 meV at 60 K is needed to greatly enhance the high temperature pinning and critical current density in an oriented polycrystalline Tl-2223 film.

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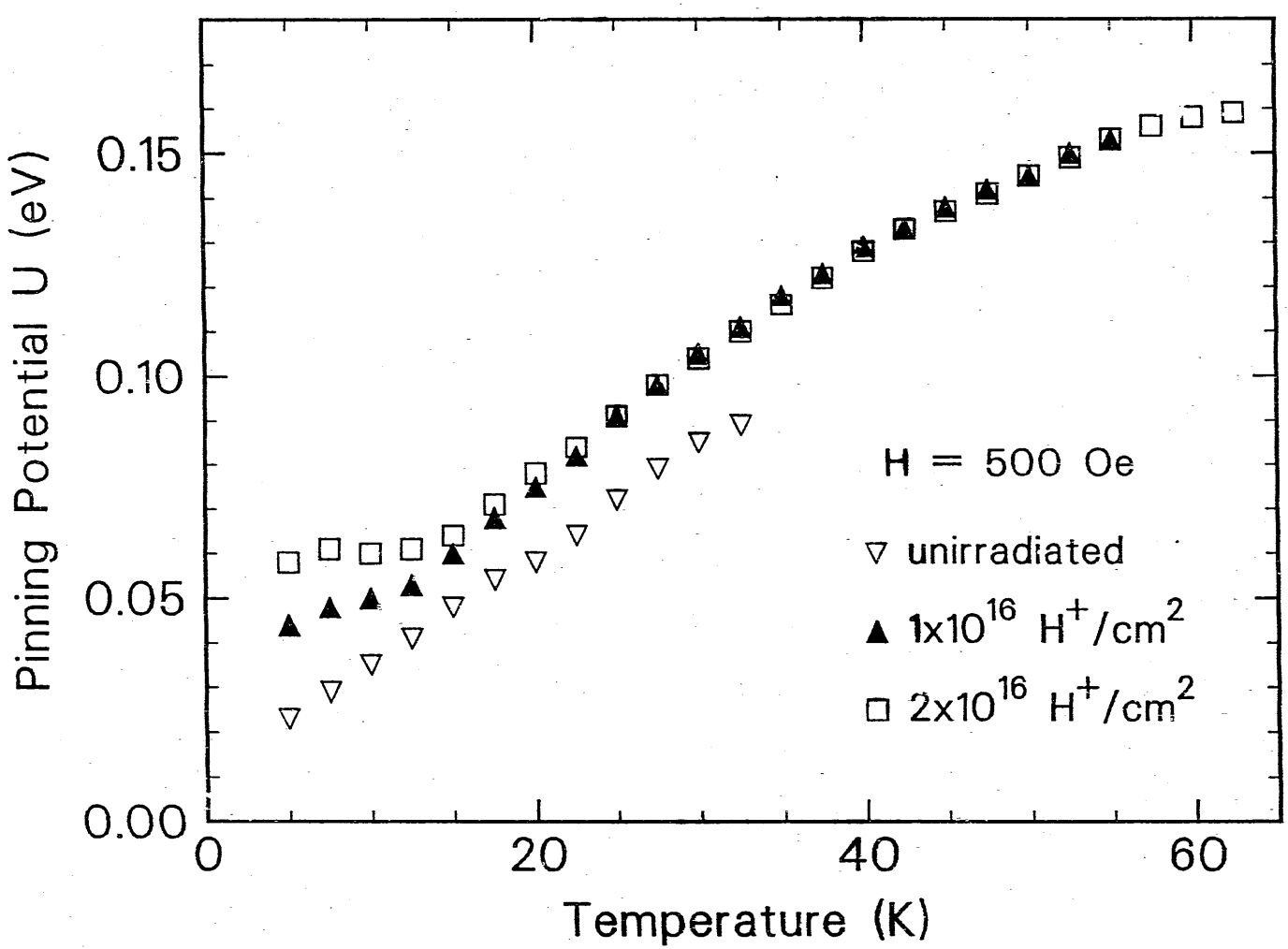
Figure Captions

Fig. 1a. 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 to damage levels of: 4×10^{-4} dpa (1×10^{16} H/cm^2), and 8×10^{-4} dpa (2×10^{16} H/cm^2).

Fig. 1b. The flux pinning potential as a function of temperature for a 300 nm thick Tl-2223 thin film before and after irradiation with 300 keV He ions to a damage level of 2×10^{-4} dpa (2.5×10^{13} He/cm^2).

Fig. 2a. 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. 2b. Ion irradiation has little effect on the magnetization critical current density for a highly oriented polycrystalline Tl-2223 thin film.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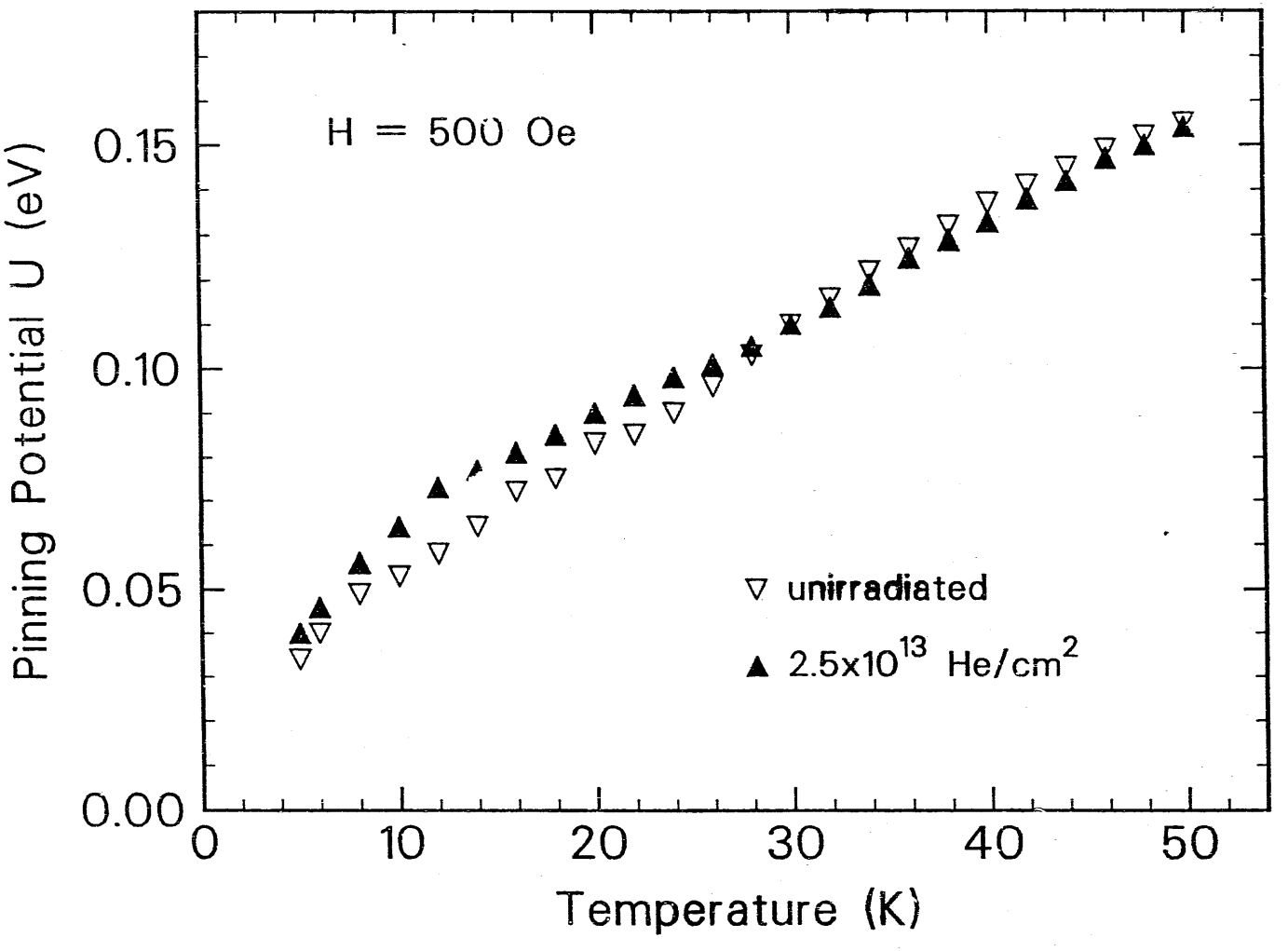
0.10
0.05
0.00
0
20
40
60
Temperature (K)
H = 500 Oe
unirradiated
1x10^16 H+/cm^2
2x10^16 H+/cm^2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Pinning Potential U (eV)
0.15
0.10
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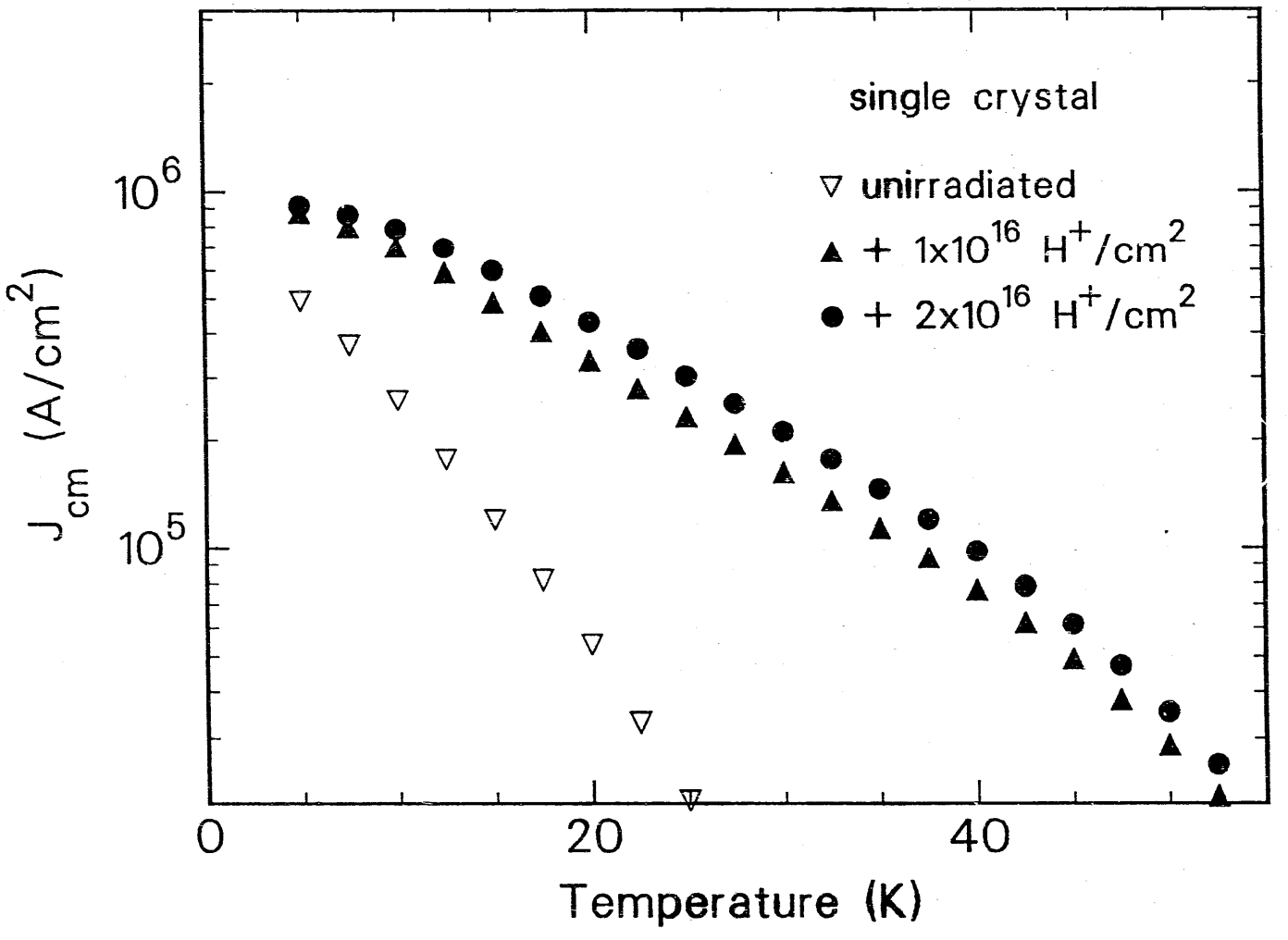
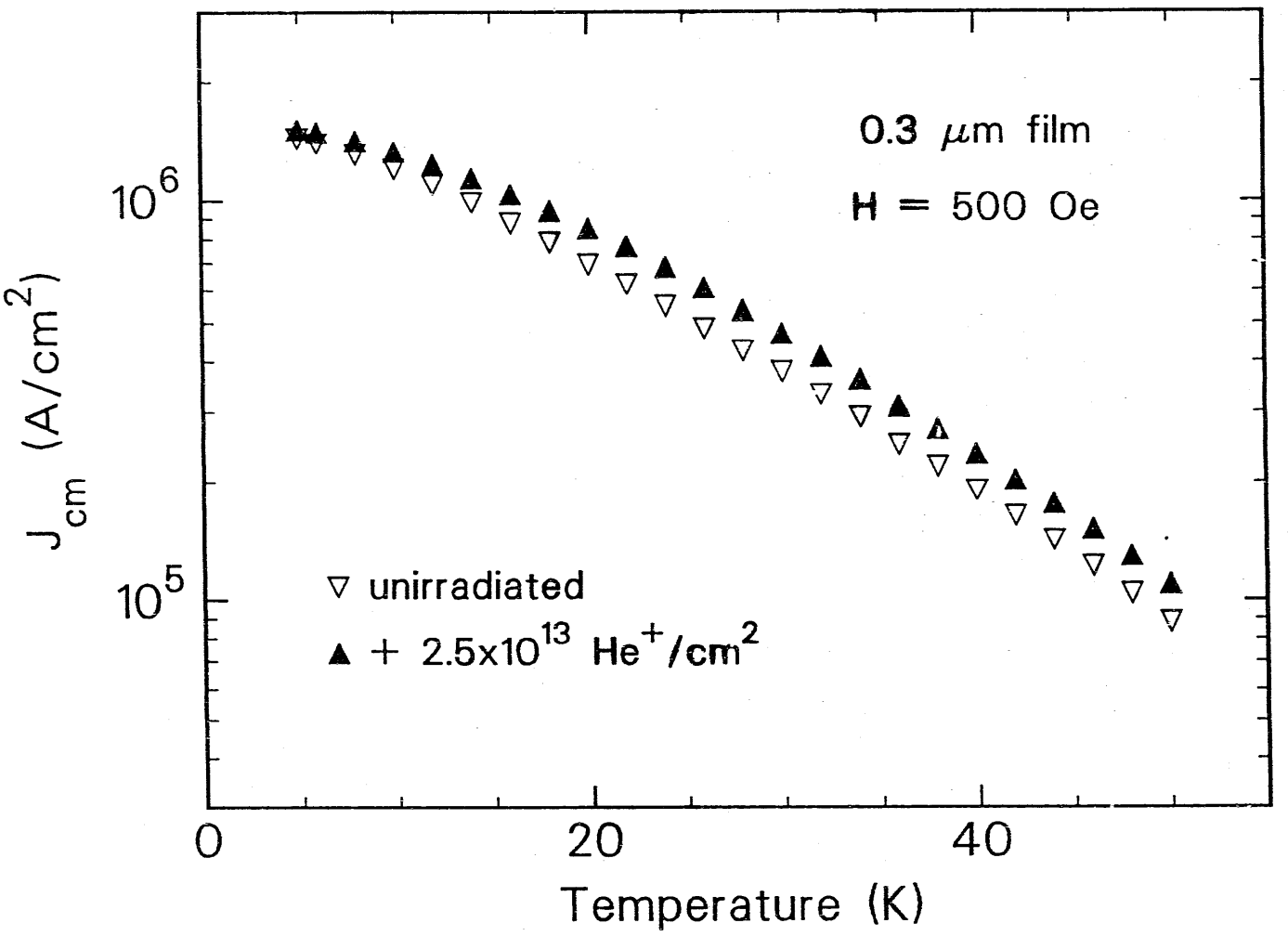
2.5x10^13 He/cm^2


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)


0.3 μm film
H = 500 Oe
▽ unirradiated
▲ + 2.5x10^13 He+/cm^2
10^6
10^5
J_cm (A/cm^2)
0
20
40
Temperature (K)

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