Abstract—Elongated, discontinuous defects provide strong vortex pinning and a corresponding large increase in critical current density in highly anisotropic Tl2Ba2CaCu2Ox-Tl-2212 films. We generate controlled, amorphous damage regions in Ti-2212 thin films ranging from short, discontinuous defects to continuous linear tracks using Au ion irradiation with incident energies of 30, 60 and 88 MeV. High-resolution TEM images and magnetization data allow correlation of microstructural damage with changes in the superconducting properties. Furnace annealing of an irradiated film shows that the enhanced pinning is stable to above 600°C.

I. INTRODUCTION

Shortly after the discovery of the high-temperature superconducting (HTS) cuprates [1], it was recognized that magnetic vortices are weakly pinned, particularly at elevated temperatures [2],[3]. Hence, there has been a large effort to understand and enhance vortex pinning by introducing lattice defects or secondary phases. Early work showed modest improvements in pinning at localized defects produced by irradiation with neutrons [4],[5] or light ions [6]. In contrast, strong vortex pinning in HTS materials was demonstrated for extended defects introduced in YBa2Cu3O7 single crystals by high-energy, heavy-ion irradiation [7],[8]. Detailed structural studies confirmed that the pinning sites were amorphous, linear damage tracks produced by these heavy ions [9].

For Tl-Ba-Ca-Cu-O superconductors, the localized defects generated by proton or neutron irradiation of a Tl2Ba2CaCu2Ox (Tl-2223) single crystal [10],[11] produced modest enhancements in magnetization critical current density Jc and vortex pinning. Considerably smaller improvements were observed for Tl2Ba2CaCu2Ox (Tl-2212) thin films [11],[12]. Extended defects provide strongly enhanced vortex pinning and Jc in both bulk ceramics [13] and thin films [14]-[16]. In this paper we vary the defect microstructure in Ti-2212 films from short, discontinuous, elongated defects to continuous, linear, amorphous tracks using Au ion irradiation at 30, 60 and 88 MeV. Magnetization data demonstrate that all types of extended defects are very effective for vortex pinning in highly anisotropic Ti-2212 films and that these defects are stable to temperatures above 600°C.

II. EXPERIMENTAL DETAILS

Ti-2212 thin films were grown by off-axis sputtering of a Ba-Ca-Cu-O precursor film onto an unheated (100) LaAlO3 single crystal substrate followed by a closed-crucible anneal in air at 850°C in the presence of Tl-Ba-Ca-Cu-O and Tl2O3 powders [17]. Superconducting properties include transition temperatures of about 102 K, low microwave surface resistance and high transport critical current densities of 2×106 A/cm² at 77 K [17],[18]. The ~600 nm-thick Ti-2212 films were cleaved into ~2×2 mm² samples and irradiated at ambient temperature to a fluence of 1.0×1011 Au ions/cm² with incident energies of 30, 60 and 88 MeV. The low dose rate of 10⁷ ions/sec/cm² caused negligible heating and the projected range for the primary ions was an order of magnitude greater than the film thickness. The incident ion direction was slightly off the normal to the film surface (the crystallographic c-axis).

High-energy heavy-ion irradiation has two major effects on the superconducting properties of HTS materials, a decrease and broadening of the Meissner transition and the generation of extended structural defects that may substantially enhance vortex pinning. While the degradation in the Meissner transition increases with the irradiation dose, there is an optimum ion fluence that maximizes pinning and Jc for a given operating temperature and magnetic field strength. To compare the effects of different incident energies and average defect lengths, all fluences in this study were equal to 1×10¹¹ Au ions/cm², corresponding to a matching field of 2 T if each ion produced a linear track.

The microstructural damage due to the Au ion irradiation was analyzed using high-resolution transmission electron microscopy (HRTEM) on a JEOL model 2010 in the Earth and Planetary Science Department at the University of New Mexico. As-grown and irradiated films were cross-sectioned and thinned to ~200 nm for electron transparency. Images were collected using a Gatan 694 slow-scan camera with Digital Micrograph imaging and analysis software. Magnetic measurements were performed with a commercial SQUID magnetometer (Quantum Design MPMS) to determine the onset of superconductivity from the Meissner transition and the critical current density from isothermal hysteresis loops.

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III. Microstructural Damage

The energy loss rates dE/dx for Au ions traversing TI-2212 films were calculated using Monte-Carlo based simulation codes TRIM [19,20] and TRIMRC [20]. These codes do not model amorphous track formation but do include both electronic and nuclear interactions between the incident ions and target atoms in the film. In particular, they are used to calculate recoil histories of the target atoms for correlation with the observed lateral damage adjacent to the amorphous tracks. The calculations employed the measured crystallographic density for TI-2212 and default atomic displacement energies. TRIMRC shows dE/dx of 9.0 keV/nm for 30 MeV Au ions, 15.2 keV/nm for 60 MeV Au and 19.5 keV/nm for 88 MeV Au. The primary ions lose less than 5% of their incident energy through the film thickness. Loss rates in this range have been shown to generate extended defects in other complex oxides [21].

The HRTEM results for Au-irradiated TI-2212 films are discussed in detail elsewhere [22]. Using prior studies on ion irradiation of yttrium iron garnet [23] as a guide, we expected that an energy loss rate above 5 keV/nm would produce elongated defects. The average length of these defects would increase with increasing loss rate; dE/dx above 12 keV/nm should yield continuous, cylindrical, amorphous damage tracks. Cross-sectional HRTEM images of the irradiated TI-2212 films reveal amorphous damage regions ~10 nm wide for all three incident energies. The average length of the defects ranges from a relatively short ~20 nm for 30 MeV Au, increasing to ~80 nm for 60 MeV Au and nearly continuous, linear tracks for 88 MeV Au. The core damage region (elongated amorphous defect) for 60 MeV Au is similar to that reported previously for 60 MeV Cu (dE/dx ~ 12.5 keV/nm) [16,24].

An important distinction between damage from Cu and Au ions is the amount of collateral damage due to secondary (recoil) ions from primary ion collisions with host atoms: the Au ions generate an order of magnitude more secondary ions than the Cu ions. Surrounding the amorphous cores from Au irradiation are dark-contrast strain lobes extending outward across 15 to 25 nm and attributed to secondary ion damage. The elongated defects from Cu ions have a thin ring of light-contrast variation attributed to point defects from secondary recoils, but strain was not observed. In contrast, HRTEM images for the microstructural damage by 30, 45, 60 and 88 MeV Au ions show that more recoil damage occurs at lower incident energy and that all Au energies introduce considerable strain adjacent to the amorphous core defects. The region surrounding the short defects from 30 MeV Au ions shows the most strain with stacking faults. However, the relatively short lengths of the 30 MeV Au defects lead to a strain volume/film volume ratio that is much smaller than for Au ions with higher incident energies and longer defect length.

IV. Magnetic Studies

1) Effects of Au irradiation: The effects of irradiation on the Meissner transition were determined from field cooling data in a 0.2 mT field applied normal to the film (along the crystallographic c-axis). Fig. 1 compares the high-temperature portion of the transition in an as-grown TI-2212 film and after irradiation at the three incident energies to a common fluence of 1.0 x 10^11 Au ions/cm^2. T_c is decreased by ~2 K (from 101 to 99 K) by the 30 MeV Au, by ~4 K for 60 MeV Au and by ~6 K for 88 MeV Au. More importantly, the transition is increasingly broadened for higher irradiation energies. The screening supercurrents in the Meissner state flow through the superconducting regions between the amorphized extended defects, so the suppression of T_c and the broadened transition reflect damage throughout the film. The greater degradation for the higher energy Au ions is attributed to the much greater strain volume/film volume shown by HRTEM images. This secondary damage is presumably nonuniform, resulting in the observed broadening.

![Fig. 1. Meissner transition in 0.2 mT (field cooling) for TI-2212 films as-grown (open triangles) and irradiated by 1x10^11 Au ions/cm^2 incident at 30 (solid triangles), 60 (open squares) and 88 (solid squares) MeV.](image.png)

The effectiveness of extended defects as vortex pinning sites is shown by comparing the magnetic critical current densities J_c in the TI-2212 films before and after irradiation. Isothermal hysteresis loops were measured to determine the magnetic field dependence of J_c. The semilog plot in Fig. 2 compares J_c at 40 K versus magnetic field applied normal to the TI-2212 film surface for an as-grown film and films irradiated with 30, 60 and 88 MeV Au ions. Note that all fields are below the matching field of 2 T. J_c was extracted from the hysteresis data using the Bean critical state model and the dimensions of the entire sample with the appropriate geometric correction [25].

At low fields J_c is decreased ~20% by the 30 MeV Au, ~30% by the 60 MeV Au and ~50% by the 88 MeV Au.
Fig. 2. Critical current density at 40 K versus applied magnetic field for Ti-2212 films as-grown and irradiated with 30, 60 and 88 MeV Au ions.

compared to the as-grown film. Vortex pinning is not the limiting factor for \( J_{\text{cm}} \) at low fields, and the decreases after irradiation reflect the smaller superconducting film volume due to the extended defects and the adjacent damaged regions. As the magnetic field increases, the increasing Lorentz force on the vortices exposes the relatively weak pinning in the as-grown film; \( J_{\text{cm}} \) drops by four orders of magnitude in a field of 0.3 T.

In contrast, the extended defects resulting from the Au irradiations are strong pinning sites for the vortices. \( J_{\text{cm}} \) decreases by less than an order of magnitude in a 0.3 T field for all three irradiated films. Interestingly, the short, discontinuous defects from the 30 MeV Au ions provide vortex pinning that is just as effective as the continuous, linear tracks from the 88 MeV Au ions. The longer discontinuous defects from the 60 MeV Au ions are the best at higher fields. Similar \( J_{\text{cm}} \) data at 20 K (not shown) indicate that the continuous tracks from the 88 MeV Au ions offer the strongest pinning in fields above 3 T. Despite the broad Meissner transition and the greater suppression of \( T_c \) by the higher energy Au ions, the continuous defects provide better pinning at low temperatures.

2) Furnace annealing and damage stability: We have recently demonstrated that the superconducting properties of Ti-Ba-Ca-Cu-O thin films are not changed significantly by one-hour furnace anneals in flowing oxygen at temperatures up to 650°C [26]. Hence, we annealed the irradiated films at 625°C to determine the stability of both the elongated defects generated by the primary Au ions and the secondary ion damage that appears as dark-contrast strain lobes. Fig. 3 compares the high-temperature portion of the Meissner transition, measured in 0.2 mT, for an as-grown Ti-2212 film, a film irradiated by \( 1.0 \times 10^{11} \) 88 MeV Au ions/cm², and an irradiated film after annealing for one hour at 625°C in flowing oxygen. \( T_c \) is decreased by \( \sim 6 \) K by the 88 MeV Au irradiation, but the oxygen anneal raises \( T_c \) to within \( \sim 2 \)K of its initial value. More importantly, the broadened transition after irradiation is narrowed to roughly the starting width. This recovery in the Meissner transition suggests that the oxygen anneal removes some of the secondary damage from the irradiation. HRTEM studies of annealed films are in progress to test this explanation.

Fig. 3. Meissner transition in 0.2 mT for Ti-2212 films as-grown (open triangles), irradiated by \( 1.0 \times 10^{11} \) 88 MeV Au ions/cm², and irradiated plus annealed at 625°C for one hour in flowing oxygen.

The effect of annealing on the damage produced by the 88 MeV Au ions is shown in Fig. 4 which compares \( J_{\text{cm}} \) at 40 K versus field for an as-grown film, an irradiated film and an irradiated plus annealed film. The annealing has two prominent effects on \( J_{\text{cm}} \). At low fields where the 88 MeV Au irradiation reduces \( J_{\text{cm}} \) by \( \sim 50 \)%, annealing at 625°C restores \( J_{\text{cm}} \) to within \( \sim 10 \)% of its value in the as-grown film. This also supports the suggestion that annealing removes secondary damage and, thus, increases the film volume supporting supercurrents at 40 K. At higher fields
there is a significant, further increase in $J_{\text{m}}$ upon annealing.

V. CONCLUDING REMARKS

Irradiation of Ti-2212 thin films by $1 \times 10^{11}$ Au ions/cm$^2$ with incident energies of 30, 60 and 88 MeV produces extended defects that act as strong pinning sites for magnetic vortices. The relatively short, discontinuous defects created by 30 MeV Au are just as effective as the continuous, linear amorphous tracks generated by 88 MeV Au. HRTEM images show dark-contrast strain lobes surrounding these defects that are attributed to secondary ion damage. These strained regions occupy a significant volume fraction of the film for 88 MeV Au irradiation, leading to a ~50% reduction in the low-field $J_{\text{m}}$ and a lowering and broadening of the Meissner transition. Annealing of a film irradiated by 88 MeV Au causes a significant recovery in both the onset temperature and width of the Meissner transition. In addition, annealing restores the low-field $J_{\text{m}}$ to within ~10% of its initial value and actually enhances $J_{\text{m}}$ at higher fields, indicating that the continuous, linear tracks remain. The improved vortex pinning after annealing may arise from a "sharpening" or localization of the pinning potential. The dark-contrast strain lobes observed by HRTEM in the as-irradiated films suggest a "smear" potential well to pin vortices. Annealing may remove some strain and cause the well to localize and, perhaps, become deeper. Further irradiation and annealing experiments as well as detailed HRTEM studies are in progress to test this explanation.

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REFERENCES