CORRELATION OF INTERMEDIATE ION ENERGY INDUCED EXTENDED DEFECT CONTINUITY TO ENHANCED PINNING POTENTIAL IN Tl-2212 FILMS

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ABSTRACT

OSTI

Lattice defects are introduced into the structure to suppress the motion of magnetic vortices and enhance the critical current density in high temperature superconductors. Point defects are not very effective pinning sites for the cuprate superconductors; however, extended defects, such as linear tracks, have been shown to be strong pinning sites. We study the superconducting cuprate Tl-2212 (the numbers designate Tl-Ba-Ca-Cu stoichiometry). Large enhancements of vortex pinning potential were observed in Tl-2212 after high-intermediate energy heavy-ion irradiations where non-continuous extended defects were induced at dE/dx of 9 to 15.2 keV/nm (60 MeV Au, 60 MeV Cu, and 30 MeV Au) and continuous linear defects were induced at 19.5 keV/nm (88 MeV Au). Our research addresses the question of pinning in highly anisotropic materials like Tl-2212 where the vortices are "pancakes" rather than "rods" and suitable defect structures may be discontinuous extended damage domains. The defect microstructure and the effectiveness of the pinning potential in Tl-2212 after irradiation by intermediate energy Au at lower dE/dx of 5-15 keV/nm, where recoils are more significant, is studied using high resolution transmission electron microscopy digital imaging and a SQUID magnetometer. The nature of the ion irradiation damage at these intermediate dE/dx will be correlated to the average vortex pinning potential and the TRIMRC calculations for recoils.

INTRODUCTION

Microstructural defects are an important aspect in the performance of many high-temperature superconductors, HTS. Many applications for HTS require high critical current densities, J_c , in high magnetic fields, B , where magnetic vortices penetrate the materials and strong Lorentz forces cause dissipation [1]. One of the goals of this study is to achieve suitable defect formation in the HTS's at ion velocities that are obtainable at most accelerator facilities. The radial and longitudinal damage profile along a heavy ion track depends strongly on the rate of energy loss (electronic and nuclear stopping power), dE/dx , in the sample. The rate of energy loss is proportional to the square of the atomic number, Z^2 . The velocity dependence of the incident ions on dE/dx can be summarized as follows. For very small ion velocities, v , of a few keV/amu (or more accurately for $v \ll v_0$; v_0 =Bohr velocity) nuclear collisions dominate dE/dx , creating a non-contiguous path of randomly displaced target ions. At a higher velocity of $0.1v_0$ to $Z^{2/3}v_0$ nuclear stopping power becomes insignificant and decreases with $1/v^2$ while the electronic stopping power increases proportional to v until it reaches a maximum. At ion velocities much larger than $Z^{2/3}v_0$, the electronic stopping power decreases again at an approximate rate of $\ln(v^2)/v^2$. The regime where extended defects and tracks can be created is near the maximum electronic stopping power.

Early work has shown some improvement in vortex pinning at localized structural defects induced by neutron or ion irradiation [2, 3]. Significant enhancement in J_c and vortex pinning was shown in a $Tl_2Ba_2Ca_2Cu_3O_{10-\delta}$, Tl-2223, single crystal after irradiation with 4.5 MeV protons. The superconducting transition temperature, T_c , decreased, but the J_c increased by an order of magnitude [4]. Irradiation that generates point defects can enhance J_c at low fluences; however, as fluence is increased T_c decreases and an increase normal state resistivity results [5, 6]. Considerable less improvement was observed in Tl-2212 films for 2 MeV protons [7]. A microstructural study with HRTEM showed damage by 1.5 MeV Kr ions in Tl-2212 single crystals consisted of clustered point defects in localized amorphized regions [8]. Strong vortex

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pinning was observed in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, YBCO, single crystals after high energy heavy ion irradiation [9, 10]. HRTEM has shown that pinning sites are amorphous, linear damage tracks [11]. Tl-based HTS ceramics [12, 13] and thin films [14, 15] showed strongly enhanced pinning when linear damage tracks were incorporated. Recent work has shown that an intermediate energy ion irradiation, 60 MeV Cu [16] and higher energy 88 MeV Au [17], in Tl-2212 thin films resulted in extended defects which greatly enhanced the vortex pinning and J_c .

The goal of this study is to induce the formation of suitable defects in the HTS phase, $\text{Tl}_2\text{Ba}_2\text{CaCu}_3\text{O}_{8-\delta}$, Tl-2212, thin films at ion velocities which are widely available at accelerator facilities; and correlate the resulting microstructural defects to the HTS properties. Loss rates in this range have been shown to generate extended defects in other complex oxides [18,19,20]. For dE/dx below ~ 12 keV/nm the damage regions are elongated but discontinuous along the incident ion path; above this threshold continuous amorphous tracks are generated [17]. It is possible that suitable defects for efficient enhancement of Tl-2212 may consist of short *c-axis* extended defects in Tl-2212. Ion irradiations, produced by a Tandem Ion Accelerator at 88, 60, and 30 MeV Au on Tl-2212 films are compared.

EXPERIMENTAL METHODS

Thin films of Tl-2212, 600 nm thick, were grown by Dupont [21]. These films typically exhibit strong *c-axis* normal orientation and are highly oriented in the plane as well. The films have superconducting onsets greater than 100K. The films have low microwave surface resistance, and high J_c 's of 2×10^6 A/cm² at 77K [22].

The Tl-2212 films were cleaved into $\sim 2 \times 2$ mm² samples and irradiated at ambient temperature to a fluence of 1.0×10^{11} Au ions/cm² with incident energies of 30, 60 and 88 MeV at the Sandia National Laboratory Tandem Accelerator. The low dose rate of 10^9 ions/sec/cm² caused negligible heating during the irradiation. The incident ion direction was slightly off the normal to the film surface (the crystallographic *c-axis*). The energy loss rate decreases by $\sim 5\%$ through the film thickness. The projected range for these ions is more than an order of magnitude greater than the 600 nm film thickness.

The dE/dx in Tl-2212 was calculated using Monte-Carlo based simulation codes, TRIM [23,24] and TRIMRC [24]. These codes do not model track formation, but are used to correlate recoil histories with the observed extent of recoil damage in the microstructure. The estimated dE/dx for these incident ions in Tl-2212 was compared to detailed damage morphology and dE/dx studies of irradiated yttrium iron garnet, YIG, [18,19,20,]. TRIMRC calculations show dE/dx in Tl-2212 increasing from 9.0 keV/nm for 30 MeV Au to 19.5 keV/nm for 88 MeV Au.

HRTEM was done on the JEOL 2010 equipped with a Gatan 694 slow-scan camera in the Earth and Planetary Sciences Department at University of New Mexico using phase contrast imaging with the (100) diffracting beams. The data were analyzed quantitatively using the Gatan's Digital Micrograph software. Superconducting properties were measured with a Quantum Design MPMS SQUID magnetometer.

RESULTS

Using the YIG studies [18,19,20] as a guide, we estimate a dE/dx of more than 4.5 keV/nm is needed to form elongated defects. Approximately 20 keV/nm is needed to produce continuous cylindrical linear damage tracks in a complex oxide. TRIMRC calculations show dE/dx of 12.5 keV/nm for 60 MeV Cu, 19.5 keV/nm for 88 MeV Au, 15.2 keV/nm for 60 MeV Au, 9.0 keV/nm for 30 MeV Au in Tl-2212 using a crystallographically determined density and default displacement energies. Electronic and nuclear interactions between ion and target atoms are taken into account. The calculated dE/dx of 12.5 keV/nm for 60 MeV Cu in the previous study [17] and 15.2 keV/nm for 60 MeV Au in this study is below the linear track formation in previous YIG studies. Cross-section HRTEM shows the continuous to discontinuous cylindrical shape of the extended amorphous domains along the ion beam direction are similar in the 60 MeV Au to the 60 MeV Cu irradiated samples with the exception there is strain surrounding the defects generated by the Au irradiation. The calculated dE/dx value of 19.5 keV/nm for 88 MeV

Au is near the 20 keV/nm threshold for forming linear tracks. Cross-section HRTEM of the resulting damage morphologies show continuous cylindrical extended damage.

HRTEM observation and comparison of the 88, 60, 30 MeV Au, and 60 MeV Cu irradiation damaged microstructures shows that more recoil damage occurred after the Au irradiations, as shown by strain surrounded defects, than the 60 MeV Cu, where only point defects surround the damage. Figure 1 shows the comparison of the damaged microstructures after the 3 Au irradiations. The 30 MeV Au irradiation generated more recoil damage than at higher energy resulting in strain surrounding the short extended defects; however the total strain volume/volume of film is much lower than that generated by 88 MeV Au. HRTEM of the region surrounding the 30 MeV Au induced defects shows stacking faults with damage along 2 to 3 planes. Dark-contrast strain lobes were also observed around the 88 MeV Au induced tracks. This strain could be caused by stacking faults in many layers surrounding the extended defects. The tracks lengths are nearly half the thickness of the film; therefore, the strain volume/volume of film is quite large resulting in broadening of the superconducting transition. Strain is also observed surrounding the 60 MeV Au induced extended amorphous defects, but was not observed after 60 MeV Cu. The extended defects are short compared to the thickness of the film and the strain volume/volume of film is much less in the 60 MeV Au than in the 88 MeV Au.

Figure 1 HRTEM observation and comparison of the 88, 60, 30 MeV Au, and 60 MeV Cu irradiation damaged microstructures shows that more recoil damage occurred after the Au irradiations than the Cu. This shows the comparison of the damaged microstructures.

The onset of superconductivity (T_C) was determined from the Meissner transition (field cooling) in a 0.2 mT field applied normal to the film (along the crystallographic c-axis). Figure 2 compares the high-temperature portion of the transition in an as-grown Tl-2212 film and after irradiation at three incident energies to a common fluence of 1.0×10^{11} Au ions/cm². T_C is decreased by ~2 K (from 101 to 99 K) by the 30 MeV Au, by ~3 K for 60 MeV Au and by 7-8 K for 88 MeV Au. The transition is increasingly broadened for higher irradiation energies. Isothermal hysteresis loops were measured to determine the magnetic field dependence of J_c . The semilog plot in Figure 3 compares J_c at 20 K versus magnetic field applied normal to the Tl-2212 film surface for an as-grown film and films irradiated with 30, 60 and 88 MeV Au ions. 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 40 K all three Au irradiations result in a dramatic, but similar, improvement in vortex pinning. The 60 MeV Au produced the greatest improvement at higher magnetic fields. At 80 K (approaching T_C) the damage at all three irradiation energies reduces J_c compared to the as-grown values, and the degradation increases with increasing incident ion energy.

Figure 2 Comparison the high-temperature portion of the transition in an as-grown Tl-2212 film and after irradiation at three incident Au ion energies at 1.0×10^{11} Au ions/cm².

Figure 3 This semilog plot compares J_c at 20 K versus magnetic field applied normal to the Tl-2212 film surface.

Figure 4 Comparison of J_c versus field at 80 K for the as-grown and irradiated films.

DISCUSSION

The decrease in T_c by ~ 2 K (from 101 to 99 K) by the 30 MeV Au, by ~ 3 K for 60 MeV Au and by 7-8 K for 88 MeV Au at a fluence of 1.0×10^{11} Au ions/cm², shown in figure 2, correlates to strain surrounding the defects. The increasingly broadened transition for 88 MeV irradiated sample correlates to the larger volume of strained microstructure surrounding the tracks. 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 suggest damage throughout the film from secondary ions generated by collisions with the incident Au ions. This secondary damage is presumably nonuniform, resulting in the observed broadening of the Meissner transition and dark contrast regions observed by TEM.

The relatively weak vortex pinning in the as-grown film, as shown by the semilog plot in figure 3, is reflected in the decrease in J_c with increasing field. J_c drops by nearly three orders of magnitude in a field of only 0.2 tesla. In contrast, the extended defects resulting from the Cu and Au irradiations provide strong pinning sites for the vortices. J_c decreases by only an order of magnitude in a field of 1 tesla following the Cu irradiation and two orders of magnitude after Au irradiation.

At low fields J_c is similar in the as-grown and all three irradiated films, suggesting that vortex pinning is not the dominant factor that limits J_c . As the magnetic field increases, the relatively weak vortex pinning in the as-grown film is reflected in the rapid decrease in J_c which drops by four orders of magnitude in a field of 2 tesla. In contrast, the extended defects resulting

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from the Au irradiations provide strong pinning sites for the vortices; J_c decreases by only an order of magnitude in the same field for all three irradiated films. At this relatively low temperature, the continuous, amorphous tracks produced by the 88 MeV Au ions offer significantly stronger vortex pinning sites as shown by the data in fields above 3 tesla. Despite the broadened Meissner transition and greater suppression of T_c for the higher energy Au ions, the continuity of the extended defects dominates the pinning force.

This is not true at higher temperatures, however. At 40 K all three Au irradiations result in a dramatic, but similar, improvement in vortex pinning with the 60 MeV Au producing the greatest improvement at higher magnetic fields. The comparison in figure 4 of J_c versus field at 80 K for the as-grown and irradiated films shows that at this relatively high temperature (approaching T_c) the damage at all three irradiation energies reduces J_c compared to the as-grown values, and the degradation increases with increasing incident ion energy. At 80 K J_c is not limited by vortex pinning and the secondary ion damage between the extended defects is very detrimental to supercurrent flow. The greater suppression of T_c and the much broader Meissner transition at higher irradiation energies, reflecting a nonuniform damage profile, are clearly dominant in reducing J_c . Hence, although the continuous amorphous tracks from the 88 MeV Au ions provide the strongest vortex pinning sites, the additional damage outside these tracks dominates the supercurrent flow. Strain free, perhaps short, extended damage with very few point defects throughout would be more efficient.

TRIM and TRIMRC alone can not predict the morphology of the damage, as this would require the calculation of time dependent electro-mechanical forces of more than 10^6 ionized target atoms for each incident ion. Using the YIG damage morphology results from the different dE/dx regimes as a guide, we determined that we need a dE/dx of approximately 20 keV/nm to produce continuous tracks, a dE/dx of approximately 10 keV/nm for extended continuous to noncontinuous. Although there is some agreement with the HRTEM observed data, the highly anisotropic layered nature of the Tl-2212 results in slightly different thresholds and damage morphologies for continuous track formation.

Since the radial and longitudinal damage profile along a heavy ion track depends strongly on the rate of energy loss dE/dx in the sample, intermediate energies can be used to obtain continuous tracks as long as the dE/dx is sufficient. There is a low and a high velocity regime where dE/dx can have the same value but differing attributes. Obviously, at low velocity the range of the ions is greatly reduced; however, this does not pose any problems for the 600 nm thick films used in this study. Secondly, and less obvious, is an increase in the track radius over the high velocity track at the same dE/dx . This can be easily explained by the increase in the impact parameter dependent ionization cross section of the target atoms as predicted in the Plain-Wave-Born approximation. At the same time, the average energy transferred to the electrons is reduced. This effect has been seen in previous studies [20].

The capability of creating continuous columnar amorphous cores in Tl-2212 films with 88 MeV Au on a Tandem Accelerator at lower energies is significant because previous track formation studies [11, 12, 13, 14, 15] were done at much higher energies with heavy ions at less accessible facilities. The fact that the continuous tracks created by the 88 MeV Au caused a broadening of the Meissner transition and breakdown at higher temperature is in part due to the strain surrounding the length of the track. The volume of the long strained cylinders is great compared to the volume of the film, while the volume of the strained damage regions from the 30 MeV Au irradiation is much less because it only involves a few planes. The 30 MeV Au induced elongate damage is odd shaped and short while the 88 MeV Au induced track lengths approach the thickness of the film and strain surrounds them. The damage volume for the 60 MeV Cu irradiated samples is much less in comparison to the 60 MeV Au irradiated samples because there is no lattice mismatch due to strain surrounding the tracks.

SUMMARY

One of the goals of this study was to induce suitable defects in Tl-2212 at ion velocities that are obtainable at most accelerator facilities. Another was to tailor the shape of defects. We have shown that extended defects are created with 30 MeV Au, 60 MeV Au, and 88 MeV Au irradiations in Tl-2212. It appears that the greater strain resulting in larger damage regions

accompanying the Au irradiation severely limits the improvement in J_c . The strain is a result of the significant recoil damage, which can be correlated to the TRIMRC calculations. Suitable defects for enhancement of magnetic pinning potential may not necessarily be continuous linear defects; however shorter extended defects with insignificant surrounding damage are desirable.

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