THE EFFECTS OF HEAVY-ION IRRADIATION ON SUPERCONDUCTIVITY IN YBa₂Cu₃O₇ EPITAXIAL FILMS

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Abstract --We report a comparative study of the superconducting behavior in YBa₂Cu₃O₇ epitaxial films irradiated with Ag and Au ions of several hundred MeV energy. The irradiation results in columnar defects of average diameter ~55 and ~110 Å for Ag⁺²¹ and Au⁺²⁴ ions respectively. A universal linear scaling is observed between the fractional areal damage vs the superconducting transition temperature and normal state resistivity. Our results also show that in these films with intrinsically strong pinning, a marginal enhancement in the critical current density (J_c) occurs when the defect density is $\leq 5 \times 10^{10}/\text{cm}^2$ and the defect diameter is of the order of coherence length. This criterion is satisfied by the silver ions. The damage due to Au⁺²⁴ ions is much too severe to improve the J_c .

I. INTRODUCTION

In addition to the challenges involved in fabricating crystallographically well-oriented materials of desired shape, a major hurdle in the technological applications of the copper-oxide based superconductors is their insignificantly low critical current density (J_c) in the presence of moderate magnetic fields. Irradiation with energetic electrons, γ -rays and nucleon (protons and neutrons) has been used since the early days of type-II superconductors for selective creation of defects which provide pinning sites for the magnetic flux [1]. A large body of literature also exists on the use of energetic electrons, protons and neutrons for pinning enhancement in high T_c cuprate superconductor [2]. However, depending on the defect density and the crystalline quality of the starting material, the consequences of irradiation have varied from marginal enhancement to degradation of the superconducting parameters. For example, after irradiation with neutrons, the enhancement in the J_c of YBa₂Cu₃O₇ (YBCO) single crystals [3], which had a low J_c to start with, is higher as compared to films with intrinsically high J_c (10^6 A/cm^2 at 77 K) [4]. As compared to the marginal effects of irradiation with nucleon on J_c , much larger improvements in the critical current, particularly at high temperatures and magnetic fields have been realized in YBCO crystals by irradiation with heavy ions of penetration depth much larger than the crystal thickness [5].

We have carried out a systematic study of the mixed state transport in epitaxial films of YBCO irradiated with the ions of silver and gold. In the unirradiated (called pristine henceforth) state, these films show a high degree of intrinsic pinning (J_c at 77 K $\sim 10^6 \text{ A/cm}^2$). The gold-ion irradiation at a dose as low as $5.6 \times 10^{10} \text{ ions/cm}^2$, leads to a

significant loss in the superconducting volume fraction which counteracts any enhancement in J_c due to additional pinning. These consequences of the radiation-induced loss of the superconducting volume are also seen in the behavior of the normal state resistivity and T_c of the films. For the silver ions, a marginal increase in the pinning force is seen at 77K.

II. EXPERIMENTAL DETAILS

Predominantly c-axis oriented superconducting thin films of thickness 300 to 400 nm were deposited, *in situ*, onto (100) cut SrTiO₃ substrates using the standard dc off-axis magnetron sputtering of a YBCO target [6]. The films were photolithographically patterned in the form of 1 mm long and 100 μm wide bridges for a four probe measurement of resistivity and J_c . Low resistivity contacts were achieved by depositing 200 nm silver pads onto which the current and voltage leads were attached with indium solder. Resistivity measurements were performed with a 11.6 Hz excitation current of magnitude low enough to ensure linear response and the voltage was measured with a lock-in amplifier. Current - Voltage (I-V) characteristics and hence the J_c , were measured with a pulsed, dc technique. The maximum transport current that we could use in the I-V measurements was limited to 100 mA.

The thin film samples and pre-thinned ceramic samples of YBCO prepared for dosimetry using a Transmission Electron Microscope (TEM), were irradiated at room temperature with gold and silver ions produced in a Tandem Van de Graaff accelerator. The ion flux during irradiation, as measured with an annular Faraday cup, was typically $\sim 10^7$ and 10^8 ions/cm^2 for Au⁺²⁴ and Ag⁺²¹ ions respectively. With the energy of 300 and 276 MeV for Au⁺²⁴ and Ag⁺²¹ ions, their mean penetration depth in YBCO is $\sim 15 \mu\text{m}$. Since the film thickness is negligibly small compared to the range of these ions, the Linear Energy Transfer (LET) of the projectiles in the film volume can be regarded as constant. This ensures a uniform damaged track along the film thickness.

III. RESULTS AND DISCUSSION

In Fig. 1 we show TEM micrographs of a YBCO sample irradiated with Au⁺²⁴ ions at a dose of $5.6 \times 10^{10} \text{ ions/cm}^2$. The ion-induced damaged regions are clearly visible in the form of circular features whose density corresponds directly to the applied dose as measured during irradiation. A high resolution image of such a feature is

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Fig. 1 Transmission electron micrograph of a sample irradiated at a dose of 5.6×10^{10} ions/cm² of 300 MeV gold ions. Inset is a high resolution TEM image of a defect.

shown in the inset of the figure. A detailed analysis [7] of the TEM images has revealed that these defects consist of amorphised material with a Gaussian size distribution which peaks at ~ 110 Å for the gold ions of 300 MeV energy measurement of Electron-Energy-Loss-Spectrum of the oxygen K-edge using a nano-meter size electron beam, also revealed a region of oxygen disorder surrounding the amorphous tracks. The diameter of amorphous tracks is nearly half for the silver ions. Furthermore, in the case of silver, the actual defect density in the material is about half of the measured fluence during irradiation. This discrepancy in the density and size of the defects produced by the Au and Ag-ions is intimately connected with the way the ions lose energy in the medium. A fast, heavy, charged particle transfers its energy to the atoms of the solid by means of direct nuclear collisions and inelastic collisions with the atomic electrons [8]. The latter mechanism, which is proportional to the effective charge on the projectile, dominated overwhelmingly in the early stages of its passage through the stopping medium. Since we are dealing with a thin film, the inelastic collisions with the electrons and dissipation of this electronic energy through electron - lattice interactions would be the primary cause of damage in the present case. The creation of amorphous tracks, as observed in Fig. 1, may result from a localized melting and subsequent rapid solidification of the material [9].

Table I(a)
Ag⁺²¹ irradiation

Fluence (TEM)	Areal Damage (A) %	Defect Separation (A)	Matching Field (T)
2.5×10^{10}	0.6	632	0.6
5×10^{10}	1.2	447	1.2
1×10^{11}	2.4	316	2.3
2×10^{11}	4.8	224	4.6

In Table I we summarize the value of T_c , normal state resistivity ($\rho(290 \text{ K})$), TEM-measured defect density and the fractional areal density (A) of the films occupied by these defects. The loss of conducting volume fraction of the films, as a result of the amorphous tracks created by the gold and silver ions, is reflected in the normal state resistivity of the material. In Fig. 2 we show the resistivity at 290 K plotted as a function of the damaged area, A, which was estimated from the size of the defects. A universal linear increase in the resistivity is seen with the increasing areal damage.

Table I(b)
Au⁺²⁴ irradiation

Fluence (TEM)	Areal Damage (A) %	Defect Separation (A)	Matching Field (T)
5.6×10^{10}	4.8	422	1.3
8.8×10^{10}	7.5	337	2.0

Fig. 2 Percentile change in the normal state resistivity at 290 K after irradiation plotted as a function the fractional areal damage.

The percentile change in T_c , defined as $[(T_c - T_{irr})/T_c] \times 100$, as a function of the areal density of the damage is shown in Fig. 3. The zero field T_c has been defined as the temperature at which the extrapolated normal state resistivity above 90 K fall to half of its value. The change in the superconducting transition temperature also scales linearly with the damaged area irrespective of the ion type used. Although this correspondence is striking, some degree of caution needs to be exercised in attributing the change in T_c directly to a macroscopic loss of the superconducting volume. This change must affect the density of states at the Fermi level in some way to reduce the T_c . It is clear from the Table, and Figs. 2 and 3 that irradiation with gold ions at a dose as low as 5.6×10^{10} ions/cm², which results in a defect separation equal to the flux line lattice spacing at ~ 1.3 Tesla, causes nearly the same change in T_c , $\rho(290 \text{ K})$ and A as due to the Ag-ion

irradiation at 2×10^{11} ions/cm². This result clearly indicates that from the view-point of optimization of the pin density vs. the loss in superconducting volume, the silver ions are much more effective than the ions of gold.

Fig. 3 Percentile change in transition temperature vs. the areal damage due to irradiation.

While discussing the issue of dissipation in the mixed state, the important question to ask is whether the irradiation leads to pinning of the magnetic flux in the material. Any additional pinning contribution would be reflected in a sensitive measurement of the resistivity in the mixed state.

Fig. 4 Resistivity of two films measured in 2 Tesla field ($H \parallel c$ -axis) before and after irradiation with gold and silver ions.

In Fig. 4 we compare the resistivity of two films measured at 2 Tesla before and after irradiation. The magnetic field in these measurement was directed perpendicular to the plane of the films. For the film irradiated at 5.6×10^{10} ions/cm² of

Au⁺²⁴ ions, the dissipation becomes detectable at ~ 80 K, which is at a slightly lower temperature as compared to the unirradiated sample. Clearly, in the Thermally Activated Flux Flow (TAFF) regime of dissipation, which is most sensitive to pinning, no improvement is seen after irradiation. However, in the case of film irradiated with the Ag-ions, the tail of the TAFF resistivity shows a small but distinct and progressive shift to higher temperatures on irradiation at 2.5×10^{10} and 5×10^{10} ions/cm². On increasing the fluence further, the resistivity curve moves back towards the curve for the unirradiated state. This data has not been shown in the figure for the sake of clarity.

Fig. 5 Pinning force density of a film at 77 K in the pristine form and after irradiation with Ag⁺²¹ ions at several fluence.

The enhancement in pinning due to silver-ions induced linear defects is also seen in the measurement of critical current density. Figure 5 shows the pinning force, $F_p = \mu_0 J_c \times H$, data taken at 77 K. The voltage criterion for J_c -measurement was 1 μ V/cm. In the pristine state, the pinning force peaks at 1 Tesla. The F_p increases on irradiation with the silver ions up to a dose of 5×10^{10} ions/cm². The maximum increase of a factor of two over the unirradiated value is reached at this dose. A further increase in the fluence reduces the pinning force density. At a fluence of 2×10^{11} ions/cm², the F_p is the same as for the unirradiated sample. Since the T_c of the material at this dose changes only by ~ 2 percent, a renormalization of the data in terms of the reduced field (B/B_{c2}) would not alter this decreasing trend in F_p .

As seen in Fig. 1, the irradiation leads to the formation of well-defined disordered regions in the material. For the gold-ion irradiation at 5.6×10^{10} and 8.8×10^{10} fluence, the average spacing between the defects is equal to the flux-line-lattice parameter at 1.3 and ~ 2 Tesla respectively. While in the case of single crystals, a similar areal density

of defects causes a significant enhancement in J_c at 77 K, the null result in the case of films irradiated with Au ions points towards two possibilities; (a) the intrinsic defects, which are responsible for the large critical current at 77 K in the pristine form, dominate the transport, and (b) any gain in pinning due to the radiation-induced defects is counteracted by the loss in superconducting volume fraction in the films. However, for the silver ions, a finite increase in pinning is evident in the critical current density data. This observation shows that the smaller diameter of the radiation-tracks created by Ag^{+21} ions allows a more efficient accommodation of the vortex cores.

In summary, we have investigated the effects of gold and silver ion irradiation on the resistivity and transport critical current density of $YBa_2Cu_3O_7$ thin films. The irradiation results in columnar defects of average diameter 55 and 110 Å for Ag^{+21} and Au^{+24} ions respectively. In the case of the silver ions, a marginal enhancement in the critical current density is seen at fluence $< 5 \times 10^{10}$ ions/cm². However, irradiation at the same fluence with Au ions results in a significant loss of the superconducting volume that nulls any gain in the pinning strength due to the columnar defects.

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