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FLUX PINNING BY HEAVY-ION-IRRADIATION INDUCED LINEAR DEFECTS IN $YBa_2Cu_3O_7$ EPITAXIAL FILMS

R. C. Budhani, Y. Zhu and M. Suenaga
Materials Science Division
Brookhaven National Laboratory
Upton, NY 11973.

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ABSTRACT

We report some transport measurements carried out to study flux pinning by heavy-ion-irradiation induced linear defects in $Y_1Ba_2Cu_3O_7$ films. Our results show that in these *in situ* deposited films containing a large concentration of defects frozen-in at the time of film growth, a marginal enhancement in critical current density occurs when the density of linear defects $< 5 \times 10^{10}/cm^2$, and their diameter of the order of coherence length. This criterion is satisfied by Ag^{+21} ions. The damage due to Au^{+24} ions is much too severe to improve the J_c.

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 J_c of $YBa_2Cu_3O_7$ (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 have been realized in YBCO crystals by irradiation with heavy ions whose linear energy transfer in the medium exceeds the threshold value for amorphisation.[5] The traversing ion thus leaves a linear track of amorphised material as it passes through the medium. These columns in the material where the superconducting order parameter (ψ) is essentially zero, provide effective pinning of the flux lines. We can estimate the pinning energy per unit length of a flux line trapped in a linear defect as follows:[6,7] the energy of a vortex per unit length is a sum of the core condensation energy (E_c) and the kinetic and magnetic energy (E_e) of the circulating superfluid;

$$E_v = E_c + E_e$$

$$= (\mu_0 H_c^2 \pi \xi^2_{ab})/2 + (\phi^2_0/4\pi\mu_0\lambda^2_{ab}) \ln (\lambda_{ab}/\xi_{ab}) \quad \text{------} \quad (1)$$

The net energy saved by the system when a flux line is trapped in a defect is;

$$= E_c + (\phi^2_0/4\pi\mu_0\lambda^2_{ab}) \ln(R/\xi_{ab})$$

$$= (\phi^2_0/4\pi\mu_0\lambda^2_{ab}) [0.5 + \ln(R/\xi_{ab})] \quad \text{------} \quad (2)$$

where R is the radius of a linear defect and other symbols have their usual meaning. This is the pinning potential per unit length of the line. For YBCO with $\lambda(0) \sim 150$ nm, $\xi_{ab}(0) \sim 1.8$ nm and $R \sim 2.8$ nm, the pinning potential for a 0.78 nm segment (spacing between CuO_2 planes of two consecutive unit cells of $YBa_2Cu_3O_7$) of the line is ~ 54 and ~ 12 meV at zero and 77 K, respectively. This should be compared with the thermal energy at 77 K (6.6 meV), a temperature at and above which possible usage of the high T_c cuprates are conceived.

In a clean system there are two intrinsic contribution to pinning that come from flux-line tension and cohesion of the Abrikosov lattice. Brandt[7] has estimated the relative strengths of the pinning forces due to lattice cohesion, line tension and linear defects denoted as F_g, F_p, and F_t respectively; their ratios vary as $F_g/F_p \sim 4B/B_{c2}$ and $F_t/F_p \sim 2\xi_c^2/d^2$, where B_{c2} is the upper critical field, ξ_c the coherence length along c and d the interlayer spacing. Since $B/B_{c2} << 1$ in high T_c cuprates, the restoring force due to lattice cohesion is negligible as compared to F_p. The line-tension contribution on the other hand, depends on ξ_c and the interlayer spacing d. Since the Tl and Bi-based cuprates containing twin Tl-O or Bi-O planes per unit cell, are highly anisotropic ($\xi_c << d$, except for $T \sim T_c$), the line tension term also does not contribute to pinning. The depinning in this case occurs by sliding of pancake vortices on the CuO_2 planes. Some recent measurements[6,8] of critical current density and decay of remanent magnetization in $Bi_2Sr_2Ca_1Cu_2O_8$ crystals irradiated with heavy ions strongly suggest the motion of pancake vortices as being the cause of dissipation in this system. In the case of YBCO however, the interlayer Josephson coupling is stronger and the contribution of line tension can not be neglected. The dissipation in this case occurs by nucleation of the vortex loops of a critical size decided by the balance of F_p, F_t and thermal energies. These loops of the critical size grow spontaneously under the action of the Lorentz force.

However, this type of description is strictly valid only when (a) the pinning is solely caused by the linear defects, and (b) the defects out-number flux lines in the system as in the high temperature regime, where lateral displacements of a flux line due to thermal fluctuations become significant, more unoccupied defects would be required as nearest neighbors for effective pinning of *individual* lines in the system.[9] In situations where the number of flux lines exceeds defects, the above description breaks down and the transport must be understood in the framework of collective pinning theories. The role of columnar defects in materials where the vortex transport is significantly affected by structural defects (such as oxygen vacancies, interstitials, twins and screw dislocations) present before irradiation is not well understood. The *in situ* deposited thin films of YBCO which show much larger critical current density in the high temperature, high field regime than single crystals, are examples of such materials.

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 of several hundred MeV energy. In the unirradiated state, these films show a high degree of flux pinning (J_c at 77 K ~ 10^6 A/cm^2) due to the structural defects frozen-in at the time of film growth. The gold-ion irradiation at a dose as low as 5.6 x 10^{10} 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 77 K. These results show that vortex dynamics in issitu deposited YBCO films is controlled primarily by preexisting defects.

EXPERIMENTAL DETAILS

Predominantly c-axis oriented superconducting thin films of thickness 300 to 400 nm were deposited, *in situ*, onto (100) cut $SrTiO_3$ substrates using the standard dc off-axis magnetron sputtering of a YBCO target.[10] 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. 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 ~10^7 and 10^8 ions/cm^2/sec 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 ~ 15 μm and the linear energy transfer rate 25 keV/nm. Since the film thickness is negligibly small compared to the range of these ions we expect uniform damaged track along the film thickness.

RESULTS AND DISCUSSION

Our detailed transmission electron microscopy studies on the irradiated samples[10,11] have shown that the radiation induced defects consist of amorphised material with a Gaussian size distribution which peaks at ~11 nm for the gold ions of 300 MeV energy. Electron energy loss spectroscopy studies reveal a significant damage in the oxygen sub-lattice in regions surrounding the amorphous tracks. The size of such defects is nearly half for the silver ions.

The loss of superconducting volume fraction in 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. 1 we show the resistivity at 290 K plotted as a function of the amorphised area, A, estimated from the size of the defects. A universal linear increase in the resistivity is seen with the increasing areal damage

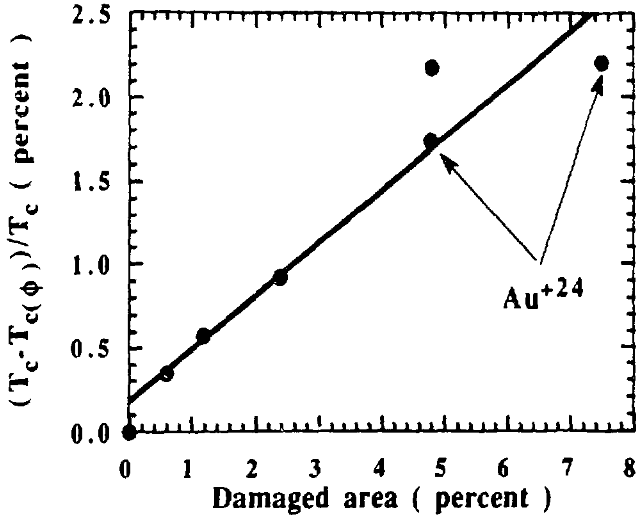

Fig. 1. Percentile change in the normal state resistivity at 290 K after irradiation plotted as a function the fractional areal damage.

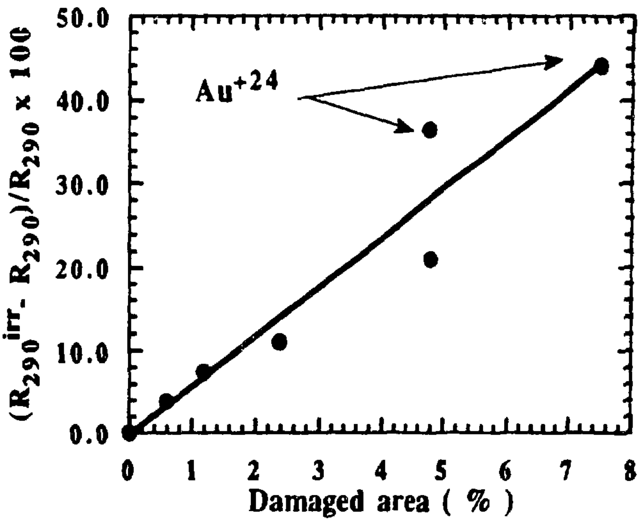

Fig. 2. Percentile change in transition temperature vs the areal damage due to irradiation.

in the films. The percentile change in T_c , defined as $[(T_c - T_{c(\phi)})/T_c] \times 100$, as a function of the areal density of the damage is shown in Fig. 2. The zero-field critical temperatures before (T_c) and after irradiation at fluence ϕ ($T_{c(\phi)}$) have 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 in the superconducting volume. The radiation must lead to a global depression of the superconducting order parameter in the material in order to affect T_c. It is clear from Figs. 1 and 2 that irradiation with gold ions at a dose as low as 5.6×10^{10} ions/cm^2, 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 , ρ(290 K) and A as due to the Ag-ion irradiation at 2×10^{11} ions/cm^2. 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.

While discussing the issue of dissipation in the mixed state, the important question to ask here is whether the irradiation leads to any increase in pinning of flux lines in the material. Any additional pinning contribution would be reflected in a sensitive measurement of the resistivity in the mixed state. In Fig. 3 we compare the resistivity of two films measured at 2 Tesla before and after

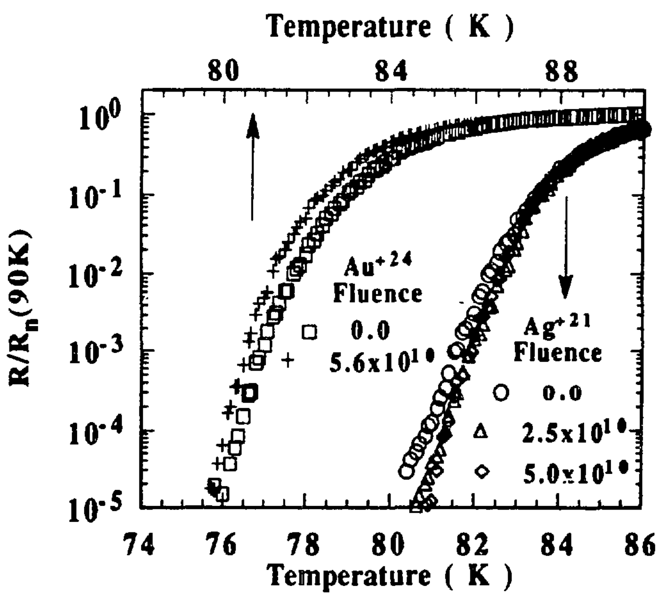

Fig. 3. Resistivity of two films measured in 2 Tesla field (H // c-axis) before and after irradiation with gold and silver ions.

irradiation. The magnetic field in these measurement was directed perpendicular to the plane of the films. For the film irradiated at 5.6 x 10^{10} ions/cm^2 of Au^{+24} ions, the dissipation becomes detectable at ~ 80 K, which is at a slightly lower temperature as compared to the data for the unirradiated sample. Clearly, in the Thermally Activated Flux Flow (TAFF) regime of dissipation, no improvement in pinning is seen after irradiation. However, in the case of film irradiated with the Ag-ions, the tail of the TAFF resistivity shows a distinct and progressive shift to higher temperatures on irradiation at 2.5 x 10^{10} and 5 x 10^{10} ions/cm^2. 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.

The enhancement in pinning due to silver-ion induced linear defects is also seen in the measurement of critical current density. In Fig. 4 we show the J_c (voltage criterion 1 μV/cm) vs applied field at 77 K. Consistent with the TAFF data shown in Fig. 3, a distinct increase in J_c occurs after irradiation at the lower fluences. At 1 Tesla for example, the J_c increases by a factor of two after irradiation at 5x10^{10} ions/cm^2. A further increase in the fluence degrades the J_c of the material. In order to fully establish the role of linear defects in controlling J_c of the material at 77 K, we need to know the relative concentrations of defects and vortices in the system. Moreover, to account for the degradation in T_c after irradiation, we need to normalize this data with respect to the upper critical field. In Fig. 5 we replot the J_c data of Fig. 4 in terms of the pinning force $F_p = (\mu_o J_c \times H)$ and the reduced field B/B_{c2}. Here we have used the B_{c2} data of Welp et al.[12]

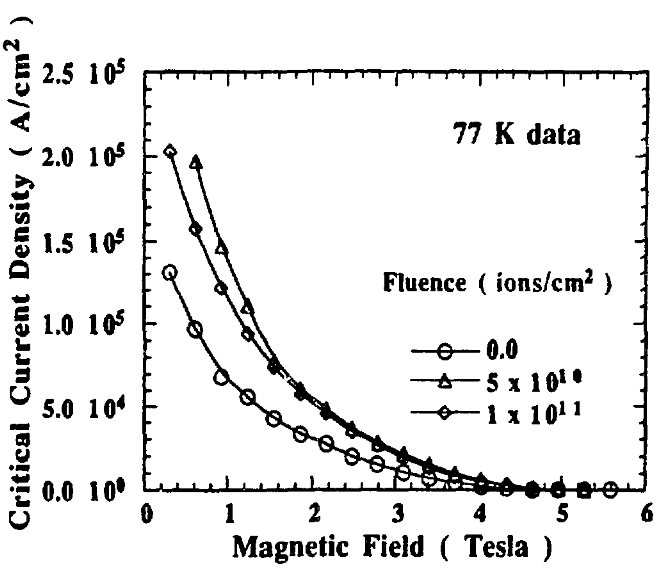

Fig. 4. Critical current density of a film at 77 K in the pristine form and after irradiation with Ag^{+21} ions at several fluence.

with the assumption that the slope dB_{c2}/dT (-1.9 Tesla/K) remains the same after irradiation. With this assumption , the B_{c2} at 77 K is ~ $1.9(T_c(\phi) - 77)$ Tesla, where $T_c(\phi)$ is the critical temperature after irradiation at dose ϕ. The F_p vs B/B_{c2} curve of Fig. 5 brings forth a very interesting feature of the vortex dynamics in this system. It shows that at higher fields the pinning force increases continuously as the concentration of linear defects is raised in the sample.

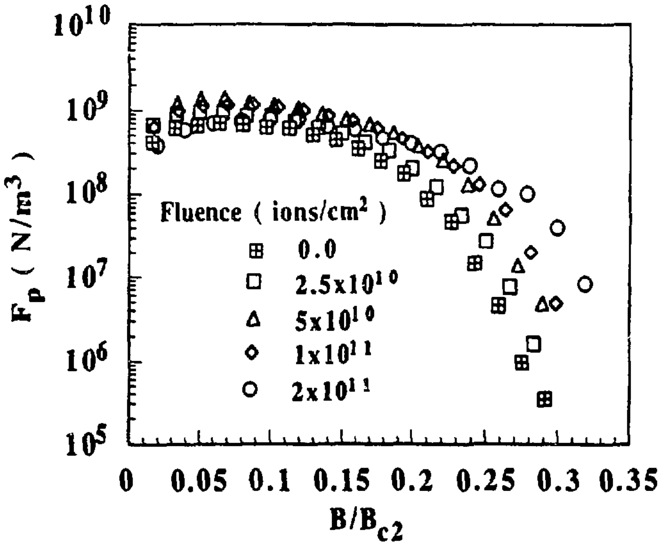

Fig. 5. Pinning force density vs B/B_{c2} at 77 K before and after irradiation with the silver ions.

As shown by our TEM studies, the irradiation leads to formation of well-defined disordered regions in the material. For the gold-ion irradiation at 5.6 x 10^{10} and 8.8 x 10^{10} ions/cm^2 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,[5] 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, in the case of silver ions a distinct increase in J_c is observed despite a strong flux pinning by the preexisting defects in the material. Behavior of the pinning force vs fluence curve suggest that a one-to-one matching between defects and vortices is required for the maximum gain in F_p. This observation shows that the linear defects created by Ag^{+21} ions allow a more efficient accommodation of the vortex cores as compared to the defects created by Au^{+24} ions.

In summary, we have investigated the effects of gold and silver ion irradiation on resistivity and transport critical current density of $YBa_2Cu_3O_7$ thin films. The irradiation results in columnar defects of average diameter 5.5 and 11 nm 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/cm^2. 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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