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Ion velocity dependent secondary electron induced point defects in SHI irradiated solids

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Abstract Swift Heavy Ion (SHI) mostly dissipates its energy in electron excitations rather than nuclear collisions inside a target material It is known that about half of the energy deposited into the electron excitation contributes to track formation, the remaining energy is transported far away by the energetic secondary electrons A certain fraction of the secondary electrons are finally ejected from the surface We report the effect of the remaining fraction left behind in the target in modifying the target properties We show that the energy of these secondary electrons is too low to induce defects by elastic scattering process We therefore consider the inelastic interaction of the low energy electrons in $YBa_2Cu_3O_{7-y}$ (YBCO) As a consequence the secondary electrons induce dissociative recombination in YBCO by breaking some of the Cu O bonds We estimate the maximum energy of the emanated secondary electrons from the ion tracks and their corresponding ranges in YBCO for different initial ion energies In contrast to the velocity effect that leads to smaller diameter tracks for ions with higher energies, we show that the diameter of the defected zone due to ion induced secondary electrons increases with increasing energy of the ion

Keywords Swift heavy ion irradiation cuprate superconductor secondary electrons velocity effect

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1. Introduction

An energetic ion traversing through a solid transfers its energy by two nearly independent processes . the nuclear and the electronic energy losses. The former process dominates in the keV range of ion energy and leads to creation of atomic size point defects and clusters of defects For the latter process, inelastic collision is the dominant mechanism for energy transfer through electronic excitations and ionisations of the target atoms [1]

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This process leads to a coherent excitation and ionisation of electrons along the ion path When the electronic energy loss, $S_e\left(=(dE/dX)_e\right)$ exceeds a threshold value, S_{eth} amorphized latent tracks are created along the ion path [2,3]

Recent studies have shown that SHI induced track registration, in addition to depending on S_e, also depends on the projectile's energy and hence the velocity of the projectile [4] Even with the same S_e, the track diameter has been found to scale inversely with ion velocity This phenomenon is known as the velocity effect

Confining S_e to values greater than S_{eth}, Bourgault *et al* [5] have shown that at a fluence $\leq 3 \times 10^{12}$ ions cm^{-2} where tracks do not overlap, the T_c onset remains almost constant in curprate superconductors due to availability of percolating paths for supercurrent flow In the present study, however we show that the T_c decrease by about 0 35 – 1 55 K in thin film of $YBa_2Cu_3O_{7-y}$ (YBCO) when irradiated at 200 MeV Ag ions at 82 K at fluences three orders of magnitude less than that observed earlier [5] To explain this unusual result, we consider the secondary electrons (SE) induced dissociative recombination in YBCO, which create point defects around the latent track and causes the observed T_c decrease

We estimate the maximum energy of the emanated SE from the ion tracks and their corresponding ranges in YBCO for different initial ion energies In contrast to the velocity effect that leads to smaller diameter tracks for ions with higher energies, we show that the diameter of the defected zone due to ion induced SE increases with increasing energy of the ion The experimental value of the radius of the defect zone at low fluence for 200 MeV Ag ions was found to be 53 nm, which agrees with the calculated value

2. Experimental

The YBCO thin films of thickness 150 nm were prepared by pulsed laser deposition technique on $LaAlO_3$ substrates The films were irradiated with 200 MeV Ag ions using the 15 MV tandem Pelletron accelerator at the IUAC, New Delhi Irradiation was done in a slightly off-normal condition (~5°) to avoid channelling effect The fluence was estimated by integrating the charges of ions impinging on the samples kept inside a cylindrical electron suppressor The ion beam was magnetically scanned over a 1 × 0 5 cm^2 area covering the complete sample surface for uniform irradiation The sampls were mounted on a copper target ladder using silver paste To prevent sample heating during irradiation and to acquire *in-situ* resistance data in the low fluence regime, a low ion beam current (0 027–0 1 pnA) was maintained The temperature during each irradiation was kept at 82 K using liquid N_2 as coolant *In-situ* temperature dependent resistance was measured after irradiating the sample with ion beams at different fluences up to a maximum temperature of 150 K in the heating cycle The temperature dependent resistance, $\rho(T)$ data was acquired using four-probe technique with a computer controlled data acquisition system Under current reversal mode the measurement gives a resolution of 100μ Ohm, which amounts to an error of ~ 0 001% Details of the synthesis routine and resistivity measurement can be found elsewhere [6]

3. Results and discussion

The electronic energy loss S_e, nuclear energy loss S_n, and range of 200 MeV Ag ion in YBCO calculated from SRIM-98 are 25.22 keV.nm^{-1}, 70 eV nm^{-1} and 12 1 μm respectively Thickness of the sample being much less than the range of the ion beam, energy deposited in the film is uniform and is mostly due to S_e. Since the S_e far exceeds S_{eth} (14 4 keV nm^{-1}) [7], amorphized latent tracks are created in YBCO About half of the energy deposited by the projectile ion in a range of a few nanometers around its path effectively contributes to thermal spike resulting in track formation [3] The rest of the energy, not used in the track formation process, is taken away by the SE These electrons are sufficiently energetic to escape the track region A fraction of these electrons finally leave the solid Direct experimental evidence of the ejected electrons do exist in the literature [8, 9] In these experiments, thin target with thickness of the order of 50 nm or less (thickness ≤ range of SE) are used to determine the electrons yield [9] In relatively thicker targets, as is used in the present study where thickness exceeds the range fo the electrons some of the SE are trapped in the target medium The effect of these trapped electrons in materials modification is studied through the evolution of superconducting transition of YBCO with irradiation fluence

The $\rho(T)$ characteristics of the sample irradiated with various fluences (Figure 1) show superconducting transition at low fluences ($\Phi < 1\ 71\times10^{11}$ ions cm^{-2}) At higher fluences up to 6 17 × 10^{12} ions.cm^{-2}, the sample showed superconducting transition almost with same mean field transition temperature, T_c but the zero resistance temperature, T_{c0} could not be achieved up to the lowest temperature (82 K) of the target ladder The room temperature resistance interestingly shows metallic behaviour even at these high

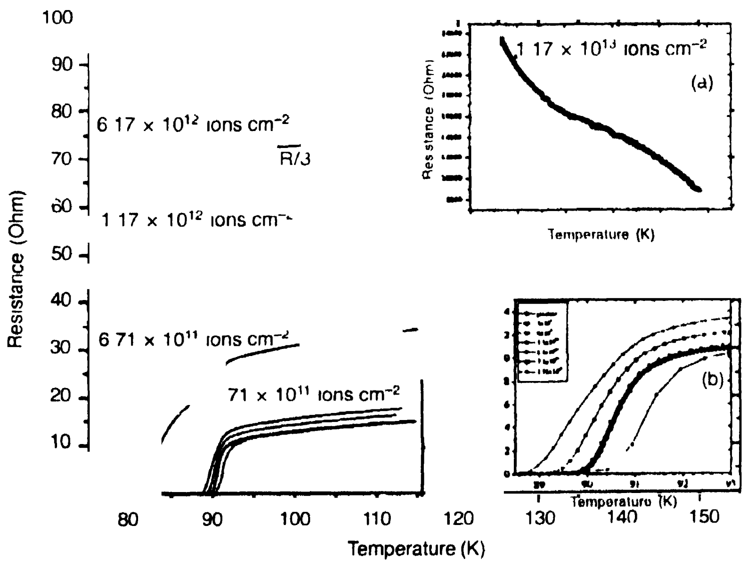

Figure 1. Evolution of superconducting transition with irradiation fluence for thin film of $YBa_2Cu_3O_{7-y}$ irradiated at 82 K by 200 MeV Ag ions Inset (a) shows $\rho(T)$ for fluence 1 × 10^{13} ions cm^{-2} Inset (b) Magnified view of the $\rho(T)$ characteristics up to fluence 1 71 × 10^{11} ions cm^{-2}

fluences. At the highest fluence of 1.1 × 10^{13} ions.cm^{-2} used in the present study, the $\rho(T)$ shows a semiconducting behaviour with complete loss of superconductivity. Figure 2 shows the fluence dependence of T_c and T_{c0}. Both T_c and T_{c0} continuously decrease with fluence up to 1.7 × 10^{11} ions.cm^{-2}.

Going by the earlier reports that SHI with $S_e > S_{eth}$ creates only latent tracks of a few (~ 5) nm diameter, it is not expected that T_c should decrease up to a fluence of 3 × 10^{12} ions.cm^{-2} [5] where tracks start overlapping and pecolative superconducting paths are blocked. We however observed a sharp decrease in T_c even in a fluence interval of 10^9 ions.cm^{-2}. The fraction of the YBCO thin film covered by latent tracks at this fluence is ~ 0.1%. This clearly indicates that the amorphized latent tracks created at this fluence alone cannot account for the observed T_c decrease, since 99.9% area of the sample is still undamaged and can provide percolating supercurrent paths.

Irradiation induced cylindrical amorphized track in a crystalline target medium is expected to impose a high radial pressure on the surrounding material. Since hydrostatic pressure is known to result into T_c increase in high T_c superconductors [10], the observed T_c decrease in the present case cannot be explained by the track induced radial pressure Further, since the amorphized latent tracks do not anneal out at room temperature, and modification induced by these tracks in the materials medium will be a permanent effect However, our earlier observations on SHI irradiated YBCO thin film showed the recovery of T_c and T_{c0} to its pre-irradiated value after annealing the sample at room temperature [11] So it is unlikely that pressure induced by latent tracks play any role in the observed T_c and T_{c0} decrease.

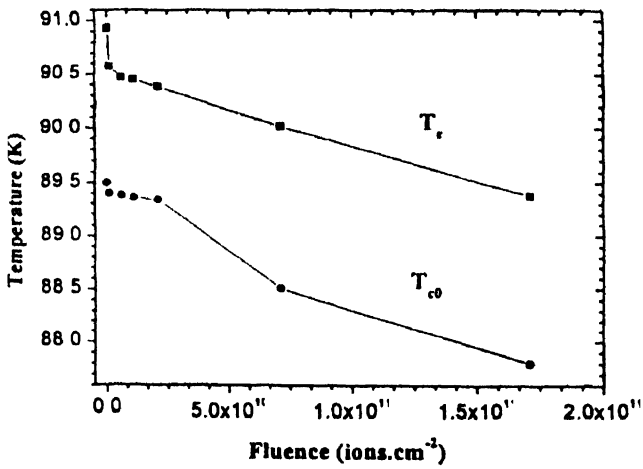

Figure 2. Variation of the mean field transition temperature, T_c and the zero resistance temperature, T_{c0} with fluence.

The T_c decrease by about 0.35–1.55 K and T_{c0} by about 0.1–1.7 K with increasing fluence up to 1.71 × 10^{11} ions.cm^{-2} (Figure 2) suggests that along with the latent tracks there must be a large concentration of point defects created due to irradiation, which anneal out at room temperature, but contribute to the $\rho(T)$ data at low temperature

These point defects are in fact created by the SE which emerge in the wake of the swift heavy ions passing through the medium [11] As discussed later, SE cannot create any other defects at more strongly bound atoms in the YBCO structure Even if some of these defects are created during SHI irradiation, they would have much shorter life time as compared to the defects created by displacement of the most loosely bound chain oxygen and hence would anneal out even at low temperature.

The maximum energy of the SE, E_0, assuming that the electrons were initially at rest, is calculated by using Rutherford scattering formula as E_0 = 4mE/M, where E is the kinetic energy of SHI and m, M are the masses of electron and SHI respectively. For 200 MeV, 250 MeV and 300 MeV Ag ions, the maximum energy of the SE are found to be ~ 4 keV, ~ 5.1 keV and ~ 6.1 keV respectively. The SE of energy E_0 will be contained within a region who radial extent from the ion tracks corresponds to the range of the electrons. The radial extent of the SE can be reasonably well described by a function of the form $R = a\ E_0{}^n$, where 'a' and 'n' are constants and were taken as 0.01 and 1.35 respectively in various stopping media [12] Here R is in mg/cm^2 when E_0 is taken in keV

For 4 keV, 5.1 keV and 6.1 keV SE corresponding to different ion energies in a medium like YBCO the maximum radial extent were found to be 6 63 × 10^{-2} mg/cm^2, 8 99 × 10^{-2} mg/cm^2 and 11.48 × 10^{-2} mg/cm^2, which correspond to ~ 110 nm, ~ 150 nm and ~ 191 nm respectively. By virtue of multiple scattering however, only a small fraction of electrons penetrate a distance comparable with the range. In the absence of a detailed form of the density distribution of the delta electrons in the YBCO medium, we assume it to peak at a value midway between the points of origin and R, *i.e.*, at R/2, which corresponds to ~ 55 nm for 200 MeV Ag ion, which was used in the present study.

One of the experimental techniques, which has been successfully used to measure diameter of latent tracks created by SHI irradiation, is to follow the fractional change in the property of a medium with ion fluence, which satisfies a Poisson relation

$$\Delta \boldsymbol{P} = \boldsymbol{P}_0\left(1 - e^{\sigma\,\phi}\right) \quad (1)$$

where ϕ is the fluence, σ is the cross sectional area of the track and P_0 represents the property of the material when it is fully covered with ion tracks. Fluence dependence of resistance has shown that for track dia of 5 nm, tracks overlap at a fluence of ~ 3 × 10^{12} ions.cm^{-2} [5]. However, if the defected zone were of much larger radius as in the present case, the overlap of the defected regions would occur at a much lower fluence. In the low fluence regime, the fluence *vs* resistance plot at constant temperature 90 K (Figure 3) shows that the value of σ is about 8919 nm^2, which corresponds to radius of about 53.3 nm. This experimental value of the radius of the defect zone is in agreement with the calculated value.

In the electronic stopping power regime, it is known that dE/dX is only the linear energy transfer and does not characterize the energy density distribution in matter From calculation performed by several author [13], it has been found that the energy density in the track region is higher at low velocity then at high velocity of the projectile ion. At constant dE/dX, this velocity effect thus predicts that the damage cross section is higher for low-velocity ions [4, 14]. However our calculation shows that the diameter of the defected zone due to ion induced SE increases with increasing energy and hence velocity of the ion, which is in contrast to the velocity effect that leads to smaller diameter tracks for ions with higher energies.

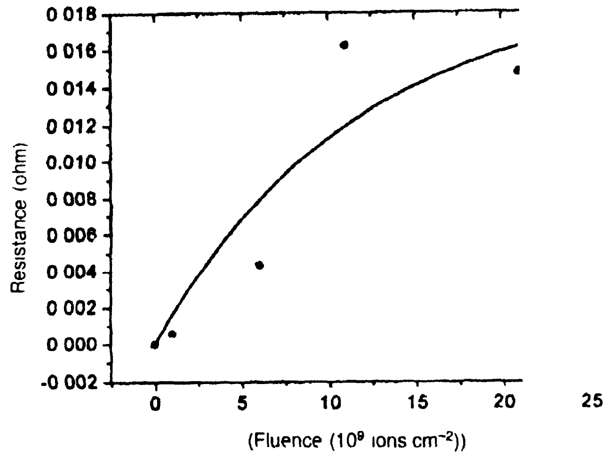

Figure 3. The data point (solid circle) shows the variation of resistance with fluence at 90 K The solid line is the fitted curve with poisson's function matter

Electron irradiation in the past has been shown to create defects in YBCO mostly in the oxygen sub-lattices. Because of the complex crystal chemistry of the cuprates, there has been a lot of controversy on the threshold energy for the creation oxygen defects The fixed five-fold oxygen coordination of Cu(2) ions makes the plane and apical oxygen ions strongly bound to the Cu ions. The chain oxygen however is loosely bound and can have lower displacement energy. The displacement energy, E_d, per ion for plain and chain oxygen was evaluated to be 8.4 eV and 2.8 eV, respectively [15]. The threshold energy, E_{th} of the electrons of mass '*m*' to displace an atom of mass '*M*' by elastic electron-atom collision is found as

$$E_d = 2\ E_{th}\left(E_{th} + 2mc^2\right)/Mc^2 . \tag{2}$$

Even with a low displacement energy (2.8 eV) of the chain oxygen, eq. (2) gives a threshold electron energy of ~ 20 keV for defect creation in YBCO. Since the maximum energy of the SE induced by Ag ions is far too low than E_{th}, a simple elastic knock-on process cannot create defects in YBCO medium. We therefore invoke the inelastic interaction of the low energy electrons with the target atoms.

Low energy electrons have been shown in the past to break bonds and cause fragmentation of molecules in hydrogen bonded molecules and organic medium by a process called dissociative recombination (DR) [16] even though its mass is 2000 times smaller than that of a hydrogen atom In this process an electron is captured by a molecular ion causing the molecule to fragment as in eq (3)

$$AB^{+} + e^{-} \rightarrow A + B \tag{3}$$

where AB^{+} is a molecular ion and A and B are neutral fragments In inorganic however, these electrons mostly lead to scintillation [12] and colour centre formation In the present study, though YBCO is an inorganic ceramic oxide, it undergoes DR as organic molecules. The DR in this system arises due to its capability of accommodating wide range of oxygen vacancies, which induces different crystal structure and metastable oxygen coordination for Cu ions in the basal planes [17]

4. Conclusion

In YBCO thin film irradiated by 200 MeV Ag ions at low temperature, we observed suppression of T_c and T_{c0} even when the fraction of latent tracks is much less than the percolation threshold for prohibiting supercurrent conduction This result points to an enhanced interaction cross-section of SHI in the medium due to creation of the point defects by the radially emitted SE around the tracks at low temperature The experimentally determined value of the radius of the defected zone is in agreement with the calculated value In contrast to the velocity effect, where diameter of the amorphized latent tracks decreases with increasing ion velocity at the same S_e value, we show that the diameter of the defected zone around the latent tracks due to ion induced SE scales with increasing ion velocity

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