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Heavy ion irradiation induced transient behaviour studies in cuprate superconductors.

T. Mohanty, A.K. Mohapatra, P.K. Samal, B.N. Dash,
D.Behera, H.P.Mohapatra, S. Misra[*], N.C. Mishra, K.
Patnaik, L. Senapati[+], D. Kanjilal[+] and G.K. Mehta[+].

Deptt. of Physics, Utkal University, Bhubaneswar-4
*Deptt. of Physics, OUAT, Bhubaneswar-3.
+Nuclear Science Centre, Post Box 10502, New Delhi.

Abstract: The present work aims at understanding the mechanism of creation and annealing of defects in cuprate superconductors by heavy ion irradiation. A study of the transient behaviour of resistivity during irradiation of $Y/GdBa_2Cu_3O_7$ thick and thin films by 100 MeV oxygen ion at 100K and at room temperature has been undertaken. The changes in resistivity with time during and between irradiations give an insight into such aspects as the effect of irradiation on grain and grain boundaries, various competing processes involved in defect creation, annihilation and interstitial oxygen diffusion.

Key words: Ion irradiation, Metastability, Oxygen diffusion

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1. Introduction

The high concentration of various defects in high Tc superconductors[1] lend these systems a metastable state[2]. Of the many means of introducing defects for inducing metastability, irradiation with high energy particles provides one with the advantage of gaining insight into the formation / annihilation of intrinsic disorders/defects. The present work investigates the effect of such defects in 123 type systems, created by high energy heavy ions of oxygen.

2. Experimental

Thick films of Gd123 were screen printed on Gd_2BaCuO_5 (Gd211) insulating substrates doped with 10% Gd123. This doping reduced the brittleness of the substrate. Thin films of Y123 deposited on $LaAlO_3$ were procured from the SSCU, IISc, Bangalore. The films were irradiated by 100MeV $^{16}O^{7+}$ beam at room temperature and at 100K using a cold-finger type LN_2 cryostat at the NSC, New Delhi. Oxygen ion flux ϕ of 2.232×10^{10} O^{7+}/cm^2-sec of $(0.2 \times 0.8) cm^2$ size, with particle currents varying from 4.0 to 17.5 enA was aimed at the middle part of the 2mm wide sample, between the two voltage probes. Pulsed beams of durations varying from 2 to 100 seconds were made to strike the samples. Sample resistance with time was continually measured on line at seconds interval using a home developed data acquisition system switching the beam off for pre-assigned periods.

3. Results and Discussion

Figure 1 shows variations of the normalized resistance R_n with time. In the case of thin film fig1(i), the R_n increases during the beam on period both at room temperature and at 100K and then equilibrates over a period of time to its pre-irradiation value. For the thick films fig1(ii), the R_n shows an opposite behaviour during the beam on period. For prolonged irradiation by the beam, a plateau and a valley were observed during beam on period for thin and thick films respectively in the $R_n(t)$ plot figs2(i) and (ii). At low temperature, similar features occur, but at a higher fluence. In case of irradiation at room temperature of both thin and thick films, the resistance transiently shoots up and drops when the beam is switched off. It

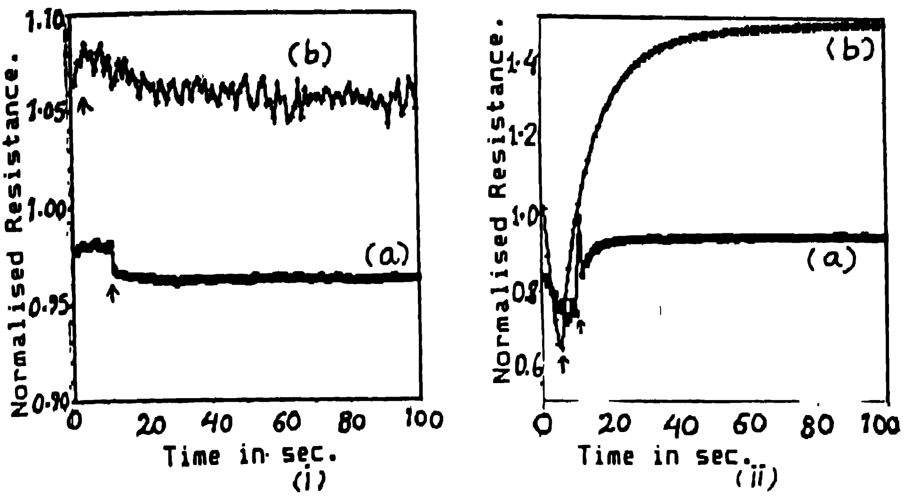

Fig.1 Variation of normalized resistance vs time during beam on and off period for (i) thin films(ii) thick films(a) data taken at room temperature(b)data taken at 100K.(^ indicates beam off)

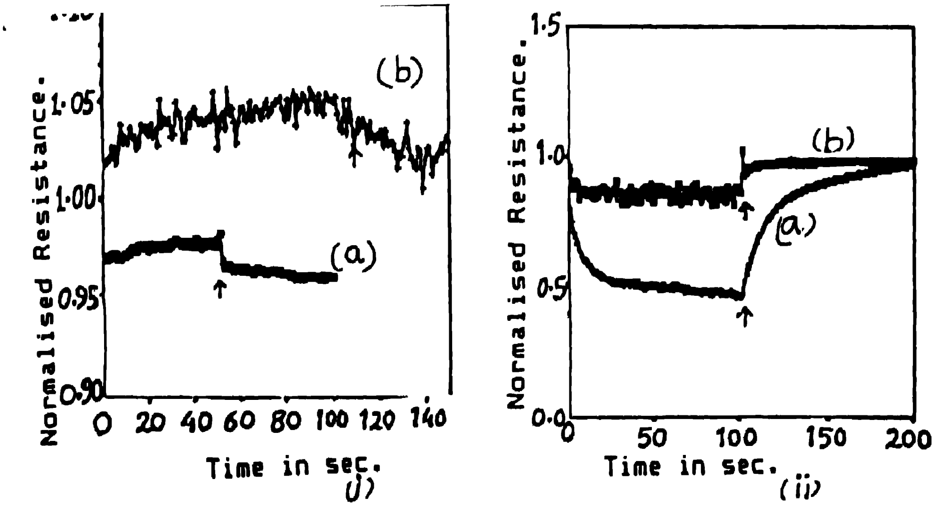

Fig.2 (i) The platue in thin film (ii)the valley in thick film observed in normalised resistance vs time atVariation of normalized resistance vs time (a)at room temperature (b)at 100K (^ indicates beam off) was observed that the system takes a longer time to equilibrate at low temperature than at room temperature.

Irradiation with heavy ions at high energies cause local thermal excitations and disorder in the structure leading to short lived or permanent point and extended defects[3]. The permanent defects increase the resistivity with increasing fluence[4]. The transient defects arising during beam on period and annealing in seconds after the beam is switched off are also expected to affect the resistance similarly. Our observations in the thin films confirm it fig(1). Unlike in the case of epitaxially grown thin films with minimal grain boundaries, the effect of irradiation on thick films having randomly oriented grains and correspondingly large grain boundaries is not well understood. Our observation of the variation of resistivity during the beam on period shows a decrease instead of the expected increase fig(2). To understand this unusual feature we note: (a)during beam on period the system is progressively driven to a far-from-equilibrium state; (b)the recovery time of the resistance to its pre-irradiation value shows a temperature dependence indicating that a diffusion process leading to equilibration is operating; (c)time for starting of the valley/plateau in the $R_n(t)$ plot showing a temperature dependence also implies that a diffusion is involved; (d)the grain boundaries consist of electron depleted CuO_2 terminating layers of grains and insulating phases like Gd211, $BaCO_3$, CuO etc.

Thus, for a system with randomly oriented grains and grain boundaries like the thick films, it is expected that several competing processes occurring in different regions of the system driven far-from-equilibrium would decide the overall resistivity. The insulating grain boundaries behave differently from the conducting grains. Hence, changes in the observed resistivity during beam on period must be accounted for by the various competing processes like creation and annihilation of charge carriers, modification of the mean-free-path etc. both in the grains and grain boundaries. Local excitations due to irradiation cause the mean-free-path to decrease contributing to increase of the resistivity as observed in the thin films. The observed decrease of the resistivity of the thick films, therefore, must take into account a process by which charge carriers can be created increasing the density of states especially at the insulating grain boundaries providing a path for current through the disjointed grains. Electronic processes as local polarization etc arising due to primary and secondary electron emission increase the hole concentration leading to a transient decrease of resistivity during irradiation. Such a process, however, cannot account for the long time scales ≅ 100 secs of post-irradiation recovery of resistance. These relaxation time-scales can be long due to restoration by diffusion of oxygen ions physically displaced by high energy beams.

In the systems of the 123 genre, the apical and the plane oxygens are far too more stably bonded than the chain oxygens [5]. The relaxation time of the thermally disordered chain oxygens at room temperature has been reported to be of the order of hours for $YBa_2Cu_3O_{6.45}$ samples quenched from 600C[6]. In the present experiment, on the other hand, the short time scale of only a few seconds of equilibration figs1(i) and (ii) from irradiation induced oxygen displacement/disorder is possible only if the system is in a far-from-equilibrium state arising due to the displacement of strongly bound apical and plane oxygens. It follows therefore that high energy ion beams, contrary to thermal excitations cause local excitations through electronic interaction processes displacing the plane and apical oxygens in addition to the chain oxygens. The slow recovery of resistivity at low temperature as compared to room temperature also supports the view that recovery may be due to oxygen diffusion process.

The valley and the plateau type features figs2(i) and (ii) indicate evolution to a steady state due to the competing processes such as creation of vacancies by high energy ions through Coulomb explosion and/or thermal spikes and trapping of the displaced oxygen ions into these vacancies through diffusion that should be slower at lower temperature as observed. We note that, during irradiation, Coulomb explosion can lead to displacement of oxygens in zero or even positive charge states liberating electrons.These electrons act as excess carriers in the otherwise insulating grain boundaries of the .thick films reducing resistivity, but in the grains, they reduce the net carrier concentration by hole electron recombination. In the thin films, epitaxial with minimal grain boundaries, this recombination decreases carrier concentration and in consequence increases resistivity.

The abrupt rise and fall of resistivity on switching the beam off, figs1(ii), 2(i) and (ii) takes place because of the relaxation of the system from a highly nonequilibrium state. This feature,seen prominent only at room temperature indicates that phonons which scatter the charge carriers, are involved in relaxation. After the beam was shut off, the metastable state self-organizes systematically restoring the preirradiation resistivity of the system indicating reorganization of the displaced plane and apical oxygens. This dependence may be rationalized in terms of well known phenomenological theories of self-organization and domain growth, provided the nature of the domain walls is taken into account[7].

4. Conclusion

The study of the transient behaviour of resistivity when a 123 system gets driven to a metastable state by high energy heavy ion irradiation and when the system equilibrates after the beam is shut off has thrown light on the effect of irradiation (i)on the grains as well as on the grain boundaries, (ii)at the strongly bound plane and apical oxygen sites causing transitory vacancies and/or altering their charge states, as well as on the recovery of the system from a metastable state induced by ion irradiation through diffusion of interstitial oxygens and on the dynamic scaling of these oxygen ordering as the system self-organizes.

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