MATERIALS MODIFICATION WITH SWIFTS HEAVY IONS

D.K.Avasthi, D.Kanjilal and G.K.Mehta Nuclear Science Centre, Post Box 10502, New Delhi-110067, India

Abstract

Swift Heavy Ions (SHI) of various species in the energy range of 50 MeV to 250 Mev provided by the 15 million volt Pelletron at NSC are being used for materials modification and characterization. Irradiation of materials at these high energies gives rise to highly excited states of lattice atoms with negligible contribution from elastic collisions. Atomic displacements and structural modifications of such a lattice due to deexcitation process are studied. SHI induced defects in semiconductors and modification of electrical characteristics are investigated. SiC formation at the interface has been observed due to electronic excitation induced ion beam mixing. SHI irradiation of organic crystals show significant changes in dielectric constant providing a possibility of making buried optical wave guide structures. Ion track diameters have been estimated from the monitoring of hydrogen release, using Elastic Recoil Detection, during ion irradiation of polymers.

1 INTRODUCTION

Energetic ion beams have been exploited by researchers in different ways in the field of materials science. Its effect on the materials depends on the ion energy, fluence and ion species. More specifically, the interaction of ion with materials is the deciding factor in the ion beam induced materials modification. The ions lose energy as they traverse through the material which is either spent in displacing atoms (of the sample) by elastic collisions or in exciting or ionizing the atoms by inelastic collision. The former is the dominant process at low energies the inelastic collisions dominate whereas at high energies where the displacement of atoms due to elastic collisions are insignificant. However, even at high energies the displacement of lattice atoms has been known to occur in insulating materials in a cylindrical core along the ion path. During the passage of the ion, a long cylinder containing charged ions is produced which explodes radially due to the conversion of electrostatic energy to coherent radial atomic movements under Coulomb forces until the ions are screened by conduction electrons. Due to the resulting cylindrical shock wave columnar defects are formed. This is known as Coulomb explosion [1]. The other competing process which can lead to the formation of the columns is thermal spike [2]. According to this model during the passage of SHI the kinetic energy of the ejected electrons is transmitted to the lattice by electron-phonon interaction in a way efficient enough to increase the local lattice temperature above the melting point of the material. The temperature increase is then followed by a rapid quenching $(10^{13}-10^{14} \text{ K/s})$ that results in an amorphous columnar structure when the melt

solidifies. Beyond certain threshold of electronic energy deposition, such ion track formation has also been observed in metals [3-5]. Swift heavy ions at NSC Pelletron [6] are being used [7] to probe into exotic effects of large electronic excitation in different types of materials e.g. metals, semiconductors, superconductors, polymers, organic crystals etc. The salient features have been defect production and annealing of defects in semiconductors, flux pinning in high Tc superconductors, desorption of H in polymers, ion beam mixing in the metal/Si interface and optical waveguide formation. The swift heavy ions are also used for on-line monitoring of light elements by elastic recoil detection (ERD) [8]. The present paper gives a brief account of these studies.

2 ELECTRONIC EXCITATION INDUCED MIXING AT INTERFACE

The ion beam mixing is widely used for generating new phases specially silicides with the help of low energy ion beams. The ion energy and species are chosen in such a way that they impart a large amount of energy to the atoms at the interface of thin film and the substrate by the process of elastic collisions. It has been observed in recent years that the electronic energy deposition beyond a certain threshold can cause the movements of atoms, which can lead to the mixing at the interface.

The mixing in Ti/Si [9] and Fe/Si [10] system has been observed by 200 MeV Ag ions. The electronic energy deposition in these cases is above 1 keV/A. The amount of mixing is found [10] to increase in Fe/Si at higher value of Se obtained by 243 MeV Au ions. In the irradiation of a diamond like carbon (DLC) film deposited on Si substrate using 50 MeV Si ions, the formation of SiC [11] at the interface has been observed. These provide evidences of the ion beam mixing mediated by the swift heavy ions passing through the material without any direct displacement of atoms through elastic collisions.

3 TAILORING OF MINORITY CARRIERS LIFETIME BY SWIFT HEAVY IONS

The variation of the lifetime of the minority carriers, τ , in crystalline silicon along depth was studied after irradiation with 60 MeV and 80 MeV silicon ions [12]. The values of τ on unirradiated surface was 19µs, which changed to 10µs and 8µs for 60 MeV and 80 MeV Si ion irradiation with fluence of about 10¹³ ions/cm². The rate of variation of τ with ion fluence reduces drastically after a fluence of about 10¹³ ions/cm² indicating that the tailoring of value of τ requires a small fluence of SHI. The variation of τ with thickness of etched out silicon

irradiated to a fluence of 10^{13} ions/cm² using 60MeV and 80MeV silicon shows that the rate of increase of τ is slow with the increase in the total thickness of the etched out silicon on the irradiated side for both the energies. A steep increase in the value of τ is observed when the total thickness of the etched out layers varied from 18 to 24µm for 60 MeV ion irradiated side and from 23 to 28µm for 80 MeV ion irradiated side of the sample. These depths correspond to the range of the ions. The value of τ of the unirradiated side remained unchanged at each stage of etching indicating that etching does not introduce defects in the sample. This was also verified by recording x-ray diffraction spectra of the irradiated and unirradiated sides before and after each stage of etching.

4 FLUX PINNING IN HTS MATERIALS

Based on the calculations of transport of ions into solids [13] TRIM-95 it is seen that the $(dE/dx)_e$ of Ag-beam in YBCO due to the inelastic ionizing collisions which produces the columnar defects peaks (~ 2keV/A) at about 200 MeV. The $(dE/dx)_n$ due to elastic collision of the Ag ion which gives rise to the point defects is negligible at this energy. The irradiation of High Temperature Superconducting (HTS) materials by this swift heavy ion causes mainly electronic excitation and ionization of the atoms of the materials through inelastic collisions up to a depth of several micro meters. Enhancement of critical current density takes place with these columnar defects

Silver beam having energy of 200 MeV was used for irradiation of single crystals of Bi2Sr2CaCu2O8+y (BSCCO) cleaved into dimension of ~1.5x1x0.02 at room temperature (~ 295K) for formation of the required number of columnar defects. Irradiation was done at an angle θ_c of 5° to avoid channeling. The resulting matching field ranged from 1 to 25 T. Investigation on the same crystal was conducted before and after irradiation. The magnetization measurements of both as-grown and irradiated crystals were carried out using a SQUID magnetometer (Quantum Design MPMS 5) at Centre of Advance Technology, Indore. The aligned columns of amorphized tracks of dimensions of about 50 Å in diameter distributed randomly in the plane normal to the direction of the beam are formed. The defects of continuous cylinders provide core pinning sites for the flux lines along the length of the defects. Figure 1 shows the magnetically determined irreversibility lines (IL) of as grown and irradiated single crystals of BSCCO [14]. The IL shows a strong shift to higher temperature after irradiation. The enhanced pinning of vortices at the columnar defects causes the shift of IL to higher temperature. The linear amorphous channels act as very effective pinning centers.



Figure: 1 Variation of magnetic field with the temperature in irradiated and as prepared BSCCO sample.

5 OPTICAL WAVEGUIDE FORMATION IN ORGANIC CRYSTALS

An optical waveguide is a layer of material whose refractive index is significantly higher than its surrounding so that the light rays remain confined in this region during its transmission through it. The possibility of the formation of such waveguides in some organic crystals have been shown by the irradiation of the organic crystals by 100 MeV Ag ion irradiation. Significant changes in the refractive index (from 1.521 to 1.564) in the irradiated region have been observed [15]. The dielectric constant of the irradiated region also increases by about an order of magnitude. The on-line H measurement by ERD technique indicated that these canges are corelated with the loss of H in the irradiated region.

6 ION TRACK RADIUS BY ON-LINE ERD MEASUREMENT

There is growing interest in nanopores and micropores in polymers generated by swift heavy ions due to vast variety of applications. These pores are made by controlled chemical etching of ion irradiated thin polymer foil. The energetic ion creates damage along its path due to its large electronic energy deposition. The track diameter is a quantity of interest for the understanding of basic ion insulator interaction. There have been a few attempts to measure the track diameters by scanning force microscopy and other state of the art surface morphology probing equipment. We have demonstrated a novel approach to determine the track diameters in polymers by on-line measurement of H loss [16] during ion irradiation by on-line elastic recoil detection analysis.

Hydrogen is liberated due to electronic excitation of constituent atoms causing the breaking of hydrogen bonds. Free H atoms combine with each other to form hydrogen molecule. Being lightest gaseous molecule having high diffusivity, these molecules escape from the polymer causing reduction in H content due to ion irradiation. Thus incident ion along its path releases H. Each ion is effective in much larger area releasing H from a cylindrical zone of damaged polymer referred as ion track. The variation of H content with ion dose is shown in Figure 2. The concentration of H [16] is plotted as a function of ion fluence (of 110 MeV Ni ions) in two polymers Polystrene and Poly vinyl di flouride(PVDF). The track radius can be estimated by the following relation.

 $H(\phi) = H_{in} \exp(-\rho. \phi)$

where H_{in} represents initial content of H in the sample, ρ the cross section of release of H and ϕ the ion fluence.



Figure: 2 H concentration as a function of ion fluence.

The initial slope gives the cross section of H release, which is equivalent to πr^2 where r is the track radius. The track radius in polyster and PVDF are found to be around 3 nm and 6nm respectively, which are consistent with other available reports [17] on track radius measurements.

7 CONCLUSION

The swift heavy ions have vast potential in the field of modification of materials by extremely large energy transferred through the electronic excitation but negligible damage due to elastic collisions. The metal/Si interface can be transformed into silicide in the case of some specific metas by swift heavy ions. Generation of controlled modification in Si and columnar defects in high Tc materials for incorporation of effective pinning by high energy heavy ions is providing centers possibilities for applications in devices. The damage generated by ion beams in organic crystals can be exploited for the formation of optical waveguides. The on-line ERD measurements of H release provides a nice way to measure the ion track radius. It is clear that the swift heavy ions in materials have several interesting and unique aspects with the possibilities of applications.

8 REFERENCES

- D. Lesueur and A. Dunlop, Rad. Eff. and Def. in Solids, 126, (1993) 163.
- [2] G. Szenes, Phys. Rev., **B51**, (1995) 8026.
- [3] D.Lesueur, Rad. Eff. and Def. in Solids, 126, (1993) 123.
- [4] T.A.Tombrello, Nucl. Instr. Meth. B103, (1995) 318.
- [5] A.Dunlop, D.Lesueur and H.Dammak, Nucl. Instr. Meth., B90,(1994) 330.
- [6] D. Kanjilal, S. Chopra, M. M. Narayanan, I. S. Iyer, V. Jha, R. Joshi and S. K. Datta, Nucl. Instr. Meth. A238, (1993) 97.
- [7] G. K. Mehta, Nucl. Instr. Meth. A382, (1996) 335.
- [8] D.K.Avasthi, Nucl. Istr. Meth. B (in press).
- [9] D.K.Avasthi et al, Proceeding of DAE Symp. on Solid State Physics, (1996).
- [10] W.Assmann, D.K.Avasthi, M.Dobler, S.Kruijer, H.D.Mieskes and H.Nolte, To be presented at SHIM-98 Berlin.
- [11] Nita Dilawar, S.Sah,B.R.Mehta, V.D.Vankar, D.K.Avasthi and G.K.Mehta, Vacuum 47 (1996) 1269.
- [12] S. T. Chavan, S. D. Dhole, V. N. Bhoraskar, D. Kanjilal and G. K. Mehta, J. Appl. Phys. 82 (10), (1997) 4805.
- [13] J. F. Ziegler, J. P. Biersack and U. Littmark, Stopping and Ranges of Ions in Matter, (Pergamon, New York, 1985).
- [14] A. K. Pradhan, S. B. Roy, P. Chaddah, D. Kanjilal, C. Chen and B. M. Wanklyn, Phys. Rev. B53, (1996) 2269.
- [15] S.Aithal, H.S.Nagaraja, P.Mohan Rao, D.K.Avasthi, A.Sarma, J. of Appl. Phys. 81 (1997) 7526.
- [16] V.K.Mittal, S.Lotha and D.K.Avasthi, Rad. Eff. & Def. in Solids (in press).
- [17] C.Trautmann, Nucl. Instr. Meth. B105 (1995) 81.