Swift Iron Ion Irradiation Induced Disorder in Crystalline Silicon

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Abstract: The p-type <111> silicon wafers have been irradiated with 70 MeV $^{56}\text{Fe}^{5+}$ ions for various fluences varying from 1×10^{12} to 5×10^{14} ions cm^{-2} at room temperature. Irradiated samples were characterized by Fourier transform infrared, ultraviolet-visible and X-ray diffraction techniques. Near infrared studies showed the presence of irradiation induced defect state in the band gap which traps the free carriers and causes the decrease in optical band gap and free carrier concentration after irradiation. The interference fringe observed in the reflectance spectra indicates the existence of irradiated layer with different refractive index as of bulk silicon. Optical depth profile of sample irradiated with ion fluence 3×10^{14} cm^{-2} showed that the irradiation induced damage/defects which vary with depth of the sample. The XRD studies revealed the presence of strain in the irradiated samples at higher ion fluence. The UV reflectance spectra revealed that the intensity of reflectance peak at 364 nm and 274 nm decreases with increasing ion fluence, indicating the increase of surface roughness after irradiation.

Key words: Swift heavy ion, iron, silicon, near infrared, optical depth profile, X-ray studies, UV reflectance spectra.

1. Introduction

Swift heavy ions with energies greater than 1 MeV/amu penetrate into a solid and lose their energy during short time interval of 10^{-16} second within a small cylindrical volume of material surrounding the ion trajectory. More than ninety percent energy loss of such projectiles is due to electronic excitations. Above a critical value of electronic energy deposition, melting around the ion trajectories followed by fast cooling and re-solidification causes the formation of ion tracks in the materials [1]. Subsequent energy and momentum transfer to the electrons and ion systems of the lattices produces disorder in the near surface region of the wafer leading to strain/stress in the materials. This provides a remarkable flexibility and adequate

opportunities to engineer properties of the materials to acquire desired structural, optical and electrical properties. The implantation of silicon with transition metal ions including iron are known to give a sequence of deep levels in silicon band gap [2]. These kinds of impurities, lattice defects and impurity defect complexes are referred to as deep centers. Several studies on low energy (keV to few MeV) iron ion implantation in silicon have been reported in the literature [3-6]. However, very few reports on the effects of swift heavy iron ions into silicon are available [7, 8]. Deep level transient spectroscopy was used to investigate the defect distribution as function of depth in silicon irradiated with low fluences of H, He, and O ions at MeV energy [5]. The damage produced by 4 MeV Ag ions in silicon for different substrate temperatures of 210, 350 and 400 K have been studied by electron paramagnetic resonance and was found to be fluence dependent [6]. The electron spin resonance

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studies of the 70 MeV $^{56}\text{Fe}^{5+}$ irradiated silicon carried by Bhole, et al. (8) showed the presence of Pa and Pb centers with gyromagnetic ratio (g) values 2.0063 and 2.0015, respectively, at room temperature. In addition to these defects, other defects were also observed at g values 2.0071 and 1.9947 are due to complex defects produced by high-energy ion implantation. The dominant signal obtained at gyromagnetic ratio value of 2.0063 was attributed to the dangling bond states of silicon. The aim of the present study is to investigate the effect of 70 MeV $^{56}\text{Fe}^{5+}$ ions on the optical and structural properties of crystalline silicon irradiated for various ion fluences varying from 1×10^{12} to 5×10^{14} ions.

2. Experiments

The p-type <111> silicon wafers having sheet resistivity $0.05 \Omega\text{-cm}$ were irradiated with 70 MeV $^{56}\text{Fe}^{5+}$ ions for different ion fluences varying from 1×10^{12} to 5×10^{14} ions at room temperature, using the 15 UD Pelletron facility at Inter University Accelerator (IUAC) Centre, New Delhi. The irradiation energy was chosen on the basis of SRIM calculation, which showed that the electronic stopping of $^{56}\text{Fe}^{5+}$ ions in silicon has an approximate maximum value of $6.60 \times 10^2 \text{ eV/\AA}$ at about 70 MeV with a projected range of $14.87 \mu\text{m}$ [9]. The samples were mounted on a copper target ladder with silver paste. The beam was magnetically scanned over $8 \text{ mm} \times 8 \text{ mm}$ area on sample surface for uniform irradiation. During the irradiation, the Fe flux was varied between 20 to 30 nA/cm^2 . The vacuum inside the irradiation chamber was maintained at about 10^{-6} mbar. The near infrared (NIR) transmission spectra of non-irradiated and irradiated samples were recorded in the spectral region $14,000$ to $4,000 \text{ cm}^{-1}$. In order to study the depth profile, sample irradiated for fluence $3 \times 10^{14} \text{ ions cm}^{-2}$ was etched in $\text{HF:HNO}_3:\text{H}_2\text{O}$ solution in the volume ratio of 8:1:1. The mid-infrared Fourier transform reflectance spectrum was recorded at each depth using Fourier transform infrared spectrometer (JASCO-610) in the spectral region $1,400$ to 400 cm^{-1} . To study the

irradiation effect on the silicon surface, reflectance spectra of non-irradiated and irradiated samples were recorded in the spectral region 150 - 2200 nm using the UV-VIS-NIR spectrometer (Shimadzu UV-3600) with standard aluminum mirror as reference. The structural properties of the irradiated samples was investigated using a JEOL Model JDX 8030 X-ray diffractometer equipped with $\text{Cu K}\alpha$ ($1.54\text{\AA}$) source.

3. Results and Discussion

3.1 Optical Studies

3.1.1 Near Infrared Studies

The optical density (αd) as a function of photon energy ($h\nu$) for the irradiated samples was calculated from the measured transmission spectra using the following equation [10]

$$T = \frac{I_t}{I_0} = \frac{(1 - R)^2 e^{-\alpha d}}{1 - R^2 e^{-2\alpha d}} \quad (1)$$

where T is the transmittance, I_0 and I_t are incident and transmitted intensities respectively, d is the thickness of the sample ($250 \mu\text{m}$), α is the absorption coefficient of the material and R is the reflectivity. In this measurement, non-irradiated silicon sample was used as reference for background intensity correction. The variation of optical density ($\alpha \cdot d$) as a function of photon energy ($h\nu$) for the samples irradiated with different ion fluences 1×10^{12} , 3×10^{14} and 5×10^{14} ions are shown in Fig. 1. The optical density curves for irradiated sample show the two effects; first the irradiation increases absorption and second the absorption edge shifted towards the lower photon energy (from 1.3 eV to 1.1 eV) with ion fluence. The increase in absorption is attributed to increase in the defect centers produced by the ion irradiation, while the shift of absorption edge indicate that the presence of irradiation induced defect states below the conduction band, which reduces the optical band gap of irradiated silicon with ion fluence. The spectral range of 1.0 - 1.3 eV corresponds to the band gap energy of silicon. The optical band gap was determined from the following expression for the indirect band to band transitions [10].

Fig. 1 Optical density vs photon energy plot for silicon samples irradiated with different ion fluences.

$$\alpha h\nu = C(E - E_g)^2 \quad (2)$$

where α is the absorption coefficient, E is the photon energy, C is a constant related to the special features of the band structure and E_g is the optical band gap. The value of band gap energy (E_g) has been calculated by taking the intersection of the linear fit of the data beyond the band gap on the X axis. The optical band gap of the samples irradiated for ion fluence 1×10^{12} , 3×10^{14} and $5 \times 10^{14} \text{ cm}^{-2}$ was found to be 1.089 ± 0.004 , 1.084 ± 0.004 and $1.045 \pm 0.004 \text{ eV}$, respectively. Where as, the corresponding estimate of optical band gap for non-irradiated silicon was $1.120 \pm 0.004 \text{ eV}$. The decrease in optical band gap with increasing ion fluence indicates the presence of irradiation induced defect states in the band gap of the silicon.

The carrier density for samples irradiated with different fluences was determined using the formula based on the classical Drude theory [10]:

$$\alpha = \frac{n e^2 \lambda^2}{4 \pi m_p^* \epsilon_0 \mu c^3 \tau} \quad (3)$$

where α is the absorption coefficient, n is the carrier concentration, μ is the refractive index of silicon, e is the electronic charge, λ is the wavelength, m_p^* is the effective mass of hole, ϵ_0 is the permittivity of free

space, c is the velocity of light and τ is the relaxation time. The carrier density estimated from Eq. (3) is given in Table 1. The carrier density decreases up to ion fluence $3 \times 10^{14} \text{ cm}^{-2}$, where as it increased slightly for ion fluence $5 \times 10^{14} \text{ cm}^{-2}$. Swift heavy ions when passing through the target material creates different types of defects at the end of the range such as point defects, defect complexes, etc. These defects trap the free charge carriers which in turn results in the decrease in the carrier density.

Below the fundamental optical absorption edge of silicon, low energy exponential absorption tails that are known as Urbach tails, was observed. This observation is caused by defects and can be described by the following empirical formula [11]:

$$\alpha = \alpha_0 \exp\left(\frac{E - E_g}{\Delta E}\right) \quad (4)$$

where α_0 is a constant, E is the incident photon energy, E_g is comparable to the band gap energy and ΔE is the inverse logarithmic slope of the Urbach tail. The calculated values of ΔE for all samples are also given in Table 1. It is evident from this Table that the value of ΔE increases from 45.6 meV for non-irradiated sample to 98.59 MeV for fluence $5 \times 10^{14} \text{ ions cm}^{-2}$. The increase of ΔE indicates that both static structural disorders and dynamic thermal disorders contribute to the absorption at the photon energies smaller than the band gap. It has been shown that structural disorders due to grain boundaries and other structural defects contribute predominantly to the Urbach tail [12].

3.1.2 Mid Infrared Studies

Fourier transform infrared reflection spectra of non-irradiated silicon sample and samples irradiated for ion fluences of 1×10^{12} , 1×10^{13} , 5×10^{13} , 3×10^{14} and $5 \times 10^{14} \text{ cm}^{-2}$ were recorded in the spectral region 1,400 to 400 cm^{-1} . Fourier transform infrared reflection spectra of silicon sample irradiated with higher ion

Table 1 Optical band gap and tail width of silicon samples irradiated with 70 MeV ^{56}Fe ions for different fluences.

Fluence (ions cm^{-2})	Non-irradiated	1×10^{12}	1×10^{13}	5×10^{13}	3×10^{14}	5×10^{14}
Tail width (meV)	45.60	55.00	67.15	73.09	75.11	98.59
Carrier Density (cm^{-3})	-	6.64×10^{19}	6.32×10^{19}	5.97×10^{19}	4.33×10^{19}	4.89×10^{19}

Fig. 2 Optical depth profiles of silicon irradiated with 70 MeV ^{56}Fe ions for fluence of 3×10^{14} ions cm^{-2} .

fluences ($> 1 \times 10^{13} \text{ cm}^{-2}$) in the spectral region 1,000 to 400 cm^{-1} exhibit a large number of interferences fringes. With increase in ion fluence, fringe amplitude increased and position of maxima and minima were shifted. The presence of interference fringes indicates the existence of thin irradiated layer with different refractive index as silicon. Because the mean layer thickness is nearly constant (projected range = $14.87 \mu\text{m}$) with ion fluence [9], it follows that the refractive index of the irradiated layer increases with increase in ion fluence [13]. The representative spectra of the sample irradiated with ion fluence of $3 \times 10^{14} \text{ cm}^{-2}$ (Fig. 2a) and etched at different depths (Figs. 2a-2h) are shown in Fig. 2. The reflectance spectrum of non-irradiated silicon sample (Fig. 2i) is also shown in Fig. 2 for comparison. The decrease in amplitude of fringes with increasing wave number is due to non-constant dielectric constant. It can be seen from the Fig. 2 that the number of fringes and amplitude decreases with etching depth (Figs. 2b-2g). At the depth of $15 \mu\text{m}$ the fringes disappeared (Fig. 2h) and

the reflectance spectrum is comparable to non-irradiated silicon (Fig. 2i). This indicates that the irradiation induced defects has been etched out at $15 \mu\text{m}$ which is comparable to the projected range ($14.87 \mu\text{m}$) of 70 MeV $^{56}\text{Fe}^{5+}$ ions in silicon.

3.1.3 UV-Visible Studies

Fig. 3 shows the UV-visible reflectance spectra of non-irradiated and irradiated samples. The reflectance

Fig. 3 Reflectance spectra of non-irradiated sample and samples irradiated for various fluence.

Fig. 4 XRD spectra of 70 MeV $^{56}\text{Fe}^{5+}$ ion irradiated silicon (a) non-irradiated (b) 1×10^{12} , (c) 5×10^{13} and (d) 3×10^{14} ions cm^{-2} .

spectra of non-irradiated sample shows two peak at 364 nm and 274 nm. The intensity of these two peaks decreased with increase in ion fluence. This indicates the increase in surface roughness due to the presence disorder in the surface region after ion irradiation.

3.2 X-Ray Diffraction Studies

The variation of intensity of the XRD peak at $2\theta = 28.4$ with different fluences is shown in Fig. 4. The crystal retains its crystalline behavior up to the ion fluence of $1 \times 10^{12} \text{ cm}^{-2}$. However, for higher fluences ($> 1 \times 10^{12} \text{ ions-cm}^{-2}$) peak intensity decreased and the XRD peak was found to shift towards the lower 2θ value. The reduction of peak intensity and broadening of XRD peak indicate the presence of amorphous silicon. The shift of peak position towards lower Bragg's angle indicates that there is a strain on the crystal during irradiation causing changes in the lattice constant.

4. Conclusions

The irradiation of silicon samples with 70 MeV $^{56}\text{Fe}^{5+}$ ions was carried out for various ion fluences ranging from 1×10^{12} to $5 \times 10^{14} \text{ ions cm}^{-2}$. Near

infrared studies showed the presence of irradiation induced defect state in the band gap which traps the free carriers and causes the decrease in optical band gap and free carrier concentration after irradiation. The interference fringe observed in the reflectance spectra indicates the existence of irradiated layer with different refractive index as of bulk silicon. Optical depth profile studies revealed that the concentration of the irradiation induced damage/defects varied continuously with depth up to the projected range. The XRD studies revealed presence of strain and non crystalline region in the silicon crystal which increases with ion fluence.

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References

- [1] G. Szenes, Z. Horváth, B. Pécz, F. Pászti, L. Tóth, Tracks induced by swift heavy ions in semiconductors, *Phy. Rev. B* 65 (2002) 045206-1-5.
- [2] A.M. Stoneham, *Semiconductor Physics*, Adam Hilger,

- Bristol and Boston, 1986.
- [3] M. Behar, H. Bernas, J. Desimoni, X.W. Lin, R.L. Maltez, Sequential phase formation by ion-induced epitaxy in Fe-implanted Si(001): Study of their properties and thermal behavior, *J. Appl. Phys.* 79 (1996) 752-762.
- [4] M. Dobler, H. Reuther, CEMS study of iron disilicide formation by iron ion implantation into silicon, *Nucl. Instrum. Methods Phys. Res. B* 155 (1999) 468-478.
- [5] A. Hallen, N. Keskitalo, Defect distributions in silicon implanted with low doses of MeV ions, *Nucl. Instrum. Methods Phys. Res. B* 186 (2002) 344-348.
- [6] C.O. Raifeartaigh, R.C. Barklie, J.K. Lindner, EPR study of defects produced by MeV Ag ion implantation into silicon, *Nucl. Instrum. Methods Phys. Res. B* 217 (2004) 442-448.
- [7] B.A. Kamalapurkar, S.K. Dubey, A.D. Yadav, K.G. Bhole, T. Mohanty, D. Kanjilal, Investigation of 70 MeV iron irradiation induced defects in C-silicon, *Nucl. Instrum. Methods Phys. Res. B* 211 (2003) 383-388.
- [8] K.G. Bhole, B.A. Kamalapurkar, S.K. Dubey, A.D. Yadav, T. Mohanty, D. Kanjilal, High-energy ion implantation of iron in silicon, *Nucl. Instrum. Methods Phys. Res. B* 212 (2003) 525-529.
- [9] J.F. Ziegler, *Handbook of Ion Implantation Technology*, North Holland, New York, 1992.
- [10] J.R. Ferraro, K. Krishnan, *Practical Fourier Transform Infrared Spectroscopy*, Academic Press Inc. San Diego, 1990.
- [11] F. Urbach, The long-wavelength edge of photographic sensitivity and of the electronic absorption of solids, *Phys. Rev.* 92 (1953) 1324.
- [12] Z. Yang, K.P. Homewood, Effect of annealing temperature on optical and structural properties of ion-beam-synthesized semiconducting FeSi₂ layers, *J. Appl. Phys.* 79 (1996) 4312-4317.
- [13] E. Wendler, G. Peiter, Optical properties of partially damaged ion implanted SiC layers by use of effective medium models *J. Appl. Phys.* 87 (2000) 7679-7684.