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Strain buildup in GaAs due to 100 MeV Ag ion irradiation

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BEAM INTERACTIONS WITH MATERIALS AND ATOMS

Shramana Mishra^a, Sudipta Bhaumik^a, Jaya Kumar Panda^a, Sunil Ojha^b, Achintya Dhar^a, D. Kabiraj^b, Anushree Roy^{a,*}

^a Department of Physics and Meteorology, Indian Institute of Technology, Kharagpur 721 302, India ^b Inter-University Accelerator Center, Aruna Asaf Ali Marg, New Delhi 110 067, India

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ABSTRACT

The formation of strained layers and a non-monotonic evolution of strain in high energy (100 MeV) silver ion (Ag⁷⁺) irradiated undoped semi-insulating GaAs are observed and analyzed using Raman scattering and high resolution X-ray diffraction (HRXRD) measurements. At low fluence, compressively strained layers are formed, whereas, with increase in fluence both compressive and tensile strains appear as observed from HRXRD measurements. Further, at low fluence, the change in compressive strain with increase in fluence is found to be sharper than what is observed at higher fluence, thereby suggesting a critical fluence value, beyond which there is a simultaneous generation and annihilation of vacancy type defects. The initial blue shift and subsequent relative red shift beyond above critical fluence in the Raman peak also qualitatively reveal non-monotonic evolution of strain in this case. Finally, we demonstrate the sensitivity of Raman spectroscopy in detecting the decrease in lattice ordering in the crystal in the low fluence regime, below the detection limit of Rutherford back-scattering channeling (c-RBS) measurements. © 2013 Elsevier B.V. All rights reserved.

1. Introduction

Group III–V semiconductors find wide scale applications in modern technology. The optical and the structural properties of the semiconductors play a crucial role in device engineering. For tuning the characteristics of an as-grown semiconductor, a variety of techniques are used. High energy ion irradiation is one such tool [1–4]. When penetrating a solid, swift heavy ions (SHI) lose their energy predominantly through inelastic interactions with the target electrons. The process is fast (the time-scale is of the order of few pico-seconds) and is confined in a space of a narrow cylindrical region (of diameter few nanometers and of length several micro-meters). SHI modify materials structurally, chemically or electrically along their path of motion [1–8]. This prompted researchers to tailor material modifications by SHI irradiation in order to achieve novel functionalities.

There are quite a few reports in the literature, which characterize the structural evolution in SHI irradiated semiconductors. It is observed that in SHI irradiated III–V binary semiconductors; crystalline to amorphous phase transformation occurs [4,7,8]. Small angle X-ray scattering (SAXS) [5] and infra-red absorption spectroscopy [6] reveal that SHI irradiation results in the breaking of

* Corresponding author. Tel.: +91 3222283856. E-mail address: anushree@phy.iitkgp.ernet.in (A. Roy). atomic bonds. It has been shown that a nanometer sized damaged zones are formed by single ion impact during SHI irradiation [7,8]. Various theoretical models are available in the literature which explains the synergy of electronic and nuclear energy loss (ε_e and ε_n , respectively) during the process of irradiation in a semiconductor [6,9]. The newer and finer experiments on SHI irradiated semiconductors would enable the researcher to refine the existing models for better understanding of the microscopic nature of the process.

In this study, we have used room temperature Raman scattering, high resolution X-ray diffraction (HRXRD) and Rutherford back scattering channeling (c-RBS) measurements to understand the microscopic nature of the defects, which are formed in GaAs due to 100 MeV silver ion (Ag⁷⁺) irradiation. We exploit the sensitivity of Raman spectroscopy to study the structural evolution at lower fluences (below the detection limit of c-RBS) of irradiation. At higher fluence range, the irradiation induced defect-disorder in the material, as obtained from Raman measurement, could be correlated with the same as revealed from c-RBS measurements. We have observed an evolution of non-monotonic compressive strain in the irradiated sample by HRXRD measurements. The change in the slope of the plot of compressive strain in SHI irradiated GaAs vs. ion fluence indicates that beyond a critical fluence of irradiation, there is a simultaneous generation and annihilation of defect states. Furthermore, we show that along with the compressive strain, the tensile strain develops beyond this critical fluence of irradiation.

2. Experimental details

High resistive ($\sim 10^6 \ \Omega \ cm$) undoped semi-insulating GaAs (SI-GaAs) single crystal wafers of orientation (001) and thickness $\sim 150 \ \mu m$, grown by liquid encapsulated Czochralski (LEC) technique, were subjected to a 100 MeV Ag⁷⁺ irradiation in the fluence range of 6×10^{11} – $6 \times 10^{13} \ ions/cm^2$ using 15UD Pelletron at IUAC, New Delhi. In case of 100 MeV Ag ions irradiation in GaAs, ε_n is $\sim 0.11 \ keV/nm$, whereas ε_e is $\sim 17 \ keV/nm$, as calculated using Monte–Carlo TRIM (Transport of Ions in Matter) simulation [10]. It is to be noted that in our case the electron energy loss is less than the threshold energy ($\varepsilon_e \sim 33 \ keV/nm$) required for track formation in GaAs [2]. The range of the ions in the material is $\sim 10.9 \ \mu m$.

HRXRD measurements were carried out using X-pert pro MRD diffractometer with $CuK_{\alpha 1}$ radiation. Diffraction patterns were measured at the GaAs (004) reflection, which is sensitive to the strain in the vertical layer structure. The X-ray penetration depth in this case is larger than 5 μ m [11]. The c-RBS measurements were performed at IUAC, New Delhi using 1.7MV Pelletron facility. 2 MeV Helium (He) was used for c-RBS experiment, where backscattered He ions were detected at an angle of 160°. A four-axis goniometer was used to align the crystals with respect to the collimated incoming He ions. Micro-Raman spectra were collected in backscattering geometry using 488 nm argon ion laser as an excitation source with a power of 10 mW. The incident beam was focused on the sample to a $\sim 2 \,\mu m$ diameter spot using a $50 \times$ objective lens. The spectrometer was equipped with a single monochromator (model TRIAX 550, make JY, France) with an edge filter and a charge coupled device (CCD) detector.

3. Results and discussion

3.1. Strain in the lattice

Fig. 1 shows GaAs (004) HRXRD curves for undoped SI-GaAs irradiated with high energy Ag ions at different fluences. The main peak centered at 33.03° is from the undamaged portion of the sample. The absence of diffraction fringes may due to the building up of non-uniform strain originating from high energy heavy ion irradiation. We observe that for the lowest fluence of irradiation (6×10^{11}



Fig. 1. HRXRD pattern of undoped SI-GaAs wafers, irradiated with Ag⁺ ions for different fluences (6×10^{11} ions/cm² (black), 3×10^{12} ions/cm² (red), 1×10^{13} ions/cm² (green), 3×10^{13} ions/cm² (blue) and 6×10^{13} ions/cm² (cyan)). Inset shows the compressive and tensile strain percentage obtained from HRXRD analysis. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

ions/cm²) the line-shape of the diffraction pattern is Gaussian without any extra peak, suggesting absence of any strain in GaAs on irradiation at this level. With increase in fluence, the main peak is broadened and a new peak appears at lower angle. These two new characteristics of diffraction patterns are signatures of the increase in disorder of the bulk material and the formation of compressively strained layer with a lattice constant larger than that of unirradiated crystal respectively. With further increase in fluence, diffraction features both at lower and higher angles appear. The diffraction peak at lower angles can be attributed to the irradiated regions, which has a larger lattice constant than the substrate. Similarly, the one at higher angle may be originating from some other regions of smaller lattice constant than the substrate. It is to be noted that these additional diffraction peaks are appreciably broad, which indicates the distribution of strain in the material. Here we would like to mention that as the projected range of ions and the penetration depth of X-ray in the samples are \sim 10.9 µm and \sim 5 µm respectively, there is possibility of HRXRD to probe the disorder due to both electronic and nuclear energy loss (ε_e and ε_n) of the incident ions in the material. However, as $\varepsilon_e \gg \varepsilon_n$ (refer to Section 2), the former is expected to have the dominant effect in the sample, which we probe by the above measurements.

From each diffraction pattern, an approximate measure of the maximum effective perpendicular strain ε_{max} was obtained from the angular separation between the Bragg peak of the unstrained and the subsidiary strained peaks from the expression [12]:

$$\varepsilon_{\max} = -\cot(\theta_{\rm B})\Delta\theta_{\rm 0},\tag{1}$$

where $\theta_{\rm B}$ is the Bragg angle for unstrained GaAs and $\Delta\theta_0$ is the largest peak separation from the unstrained peak. $\varepsilon_{\rm max}$ is positive or negative depending on whether there is a net effective compressive or tensile strain in the lattice in the perpendicular direction. The values of percentage of $\varepsilon_{\rm max}$ as obtained from this analysis are shown in the inset of Fig. 1 (filled circles) for different fluences of irradiation.

A large number of reports are available in literature, where compressive and tensile strains were observed independently on ion irradiation in III-V semiconductors [11-19]. For low energy (keV) ion-implantation [11,12] or high energy light ion (HELI) irradiation [13–17], only compressive strain was observed. The origin of compressive strain in irradiated GaAs is explained in view of generation of vacancy-type defects during irradiation [13-15]. In a vacancy-type defect, a missing atom in the lattice causes inward displacements of the neighboring atoms. The magnitude of the displacement decreases as the neighboring atoms are away from the center of the vacancy. Thus, unequal-distance displacements of the neighboring shell atoms result in an expansion of the lattice spacing [13–15]. Considering diffusion of defects during irradiation, it has been suggested that the predominant defects in the near-surface disorder region in MeV-ion-irradiated GaAs are vacancies and their complexes [15].

The tensile strain had been reported in InP on SHI irradiation [18,19]. Interstitials (i.e., displaced atoms) in a damaged crystal produce an opposite displacement direction of the surrounding atoms compared to what is observed for a vacancy type defects in the system. However, for this particular case compressive strain was not observed. The appearance of both compressive and tensile stain in irradiated samples, as observed by us, is most likely due to the simultaneous formation of vacancy and interstitial defects in Ag irradiated GaAs beyond a certain fluence of irradiation. The appearance of both types of defects in Ag ion irradiated GaAs may be due to the fact that the mass of Ag ions and energy of irradiation, used by us, are much higher than those ions and energy reported in the above references [13–16,18,19]. Here, the tensile strain appears at the fluence of $1 \times 10^{13} \text{ ions/cm}^2$. At this fluence

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