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Surface modification of gallium phosphide caused by swift (200 MeV) silver ions

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ABSTRACT

In the present work, the effects of swift silver ion irradiation in crystalline gallium phosphide samples with various fluences ranging between 1×10^{11} and 2×10^{13} ions cm^{-2} have been described. Atomic force microscopy images of the samples irradiated with different fluences showed the existence of hillocks at the surface, the diameter and density of these clusters were found to be depend on the ion fluence. As the ion fluence increased ($\geqslant 1 \times 10^{13}$ ions cm^{-2}), the big size hillocks having arbitrary shapes were observed due to outflow of the molten material to the sample surface or defect induced swelling of track areas accumulated during the track overlapping. Phonon confinement model employed to first order Raman scattering from longitudinal optical phonon mode revealed the decrease in phonon coherence length from 73.0 nm to 23.7 nm with the increase in ion fluence from 1×10^{12} to 2×10^{13} ion cm^{-2}.

1. Introduction

When a swift heavy ion penetrates through a solid, it produces the electronic excitation of the atoms in the material with negligible contribution from nuclear energy loss and the effective electronic energy loss that brings out interesting change in the materials properties [1–4]. Generally, swift heavy ion irradiation of semiconductors with any kind of species in the electronic energy loss regime produces the point defects and point defect clusters. In addition to the defects mentioned above, beyond a threshold of energy deposition to the electronic system, ion tracks of different size (few nanometers in diameter) depending on the target materials and ion nature were also observed [5]. However, the reports on experimental evidence of the formation of ion tracks in semiconductors are limited. In the literature, several models such as Coulomb explosion, shock wave, inelastic thermal spike etc. have been developed in details to describe the damage induced in the solids by swift heavy ions. Among them thermal spike model is most widely used to discuss the ion track formation in all kind of materials irradiated by swift heavy ions [5–11]. Gallium phosphide is a very promising semiconductor material suitable for various electronic and optoelectronic applications. It is being used in the manufacture of red, orange and green light emitting diodes and semiconductor lasers [12]. The irradiation of gallium phosphide with different species i.e. N^+, Kr^+, Ar^+ etc. at high energy (<10 MeV) have previously reported [13–16]. To the best of our knowledge, the irradiation of gallium phosphide with swift heavy ions, having velocities comparable to or higher than the orbital electron velocity, is much less investigated [5,17,18]. The modification occurred in gallium phosphide after interaction with 100 MeV iron ions for various ion fluences varying between 1×10^{11} and 1×10^{14} ions cm^{-2} have been studied using the atomic force microscopy, Raman scattering, X-ray diffraction and Fourier transforms infrared measurements [17,18]. The transmission electron microscopic studies of gallium phosphide irradiated with 573 MeV gold ions for the fluence of 1×10^{12} ions cm^{-2} have shown the formation of discontinuous ion tracks [5]. The main purpose of this study is to investigate the changes which occurs in gallium phosphide surface on interacting with swift (200 MeV) silver (Ag^{9+}) ions for various ion fluences varying between 1×10^{11} and 2×10^{13} ions cm^{-2} by atomic force microscopy and Raman scattering measurements.

2. Experimental details

In the present work, we have used undoped, <1 1 1> orientation, single crystalline gallium phosphide wafers, which were cut into pieces typically 1 cm × 1 cm samples. These samples were irradiated with Ag^{9+} ions for various ion fluences ranging between 1×10^{11} and 2×10^{13} ions cm^{-2} at 200 MeV using 15 UD Tandem Pelletron Accelerator at Inter University Accelerator Centre, New

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Delhi. The samples were mounted on copper target ladder with silver paste to have good thermal conduction between them. During the ion irradiation, the beam was scanned over 1 cm × 1 cm area on the sample surface for uniform irradiation and the beam current was maintained between 3 and 4 pnA (particle nano ampere). The charge collected by the sample holder was measured by a current integrator. The vacuum inside the irradiation chamber was maintained about 10^{-4} Pa. The projected range of 200 MeV Ag^{9+} ions in gallium phosphide estimated from SRIM code was found to be 20.05 μm with a lateral straggling value of the order of 1.08 μm [19]. The values of electronic energy loss (S_e) along with nuclear energy loss (S_n) for silver ions in gallium phosphide as a function of beam energy varying between 1 keV and 1000 MeV estimated from the SRIM code have shown in Fig. 1. Both S_e and S_n increases with increasing the beam energy, reaching a maximum value and then start to decrease. The peak in electronic energy loss is known as Bragg peak. It lies at ~350 MeV and the contributions of S_e and S_n at the Bragg peak are 1.75×10^4 keV/μm and 2.85×10^1 keV/μm respectively. In our experiment, we have chosen the energy of Ag^+ ions to 200 MeV, which is near to the Bragg peak. Therefore maximum modifications which take place at the surface of gallium phosphide are expected due to electronic energy deposition. The surface morphology of samples before and after irradiation with different ion fluences was examined using atomic force microscope (Nanoscope III). A cantilever with a silicon nitride tip was scanned over the samples. The measurements were carried out in tapping mode. The scanned AFM images had 512 × 512 point resolution. First order Raman scattering spectra were recorded using 514.50 nm line of argon ion laser in the wave number range between 320 cm^{-1} and 440 cm^{-1} using RENISHAW in Via microscope.

3. Results and discussion

3.1. Atomic force microscopy studies

The two dimensional atomic force microscopy (AFM) images (3 μm × 3 μm) of gallium phosphide samples before and after irradiation with 200 MeV Ag^{9+} ions for various ion fluences varying between 1×10^{11} and 2×10^{13} ions cm^{-2} were recorded. The representative images of the non irradiated and samples irradiated for the fluence of 1×10^{11}, 5×10^{12} and 2×10^{13} ions cm^{-2} are shown in Fig. 2. From the Figure, it is clear that the non irradiated sample has very smooth surface (Fig. 2(a)). AFM image (Fig. 2(b)) of the sample irradiated with low ion fluence of 1×10^{11} cm^{-2} showed the pit like features on the surface of gallium phosphide.

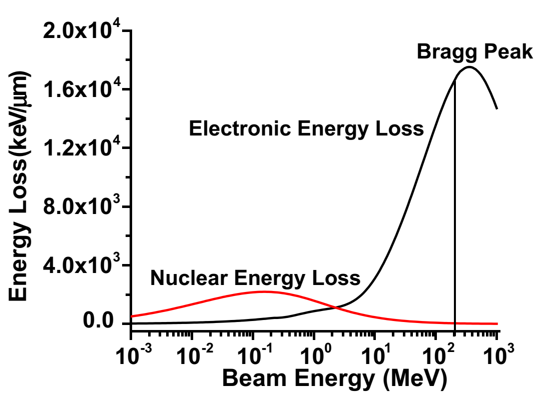

Fig. 1. Variation of electronic energy loss along with nuclear energy loss for silver ions in gallium phosphide as a function of beam energy.

These pits may be due to ion tracks in gallium phosphide created from swift (200 MeV) Ag^{9+} ion impact. However, the samples irradiated with ⩾5×10^{12} ions cm^{-2} AFM measurements revealed the existence of hillocks (Fig. 2(b) and (c)). The average diameters of the hillocks were found between 50 nm and 100 nm. As the ion fluence increased further (⩾1×10^{13} ions cm^{-2}), the big size hillocks having arbitrary shape (Fig. 1(d)) were formed. It is well known that the electronic energy loss released by swift heavy ions beyond threshold value transferred to the lattice, increases the temperature of the material leading to a cylindrical molten zone of few nanometers radius. As a result, an outflow of the molten material transported to the sample surface or defect induced swelling of track areas accumulated during the track overlapping to form the hillocks on the surface of gallium phosphide [20–23]. In order to compare our results with the Kamarou et al. [5], we have plotted the variation of electronic energy loss and nuclear energy loss of 573 MeV Au^+ ions and 200 MeV Ag $^+$ ions incident in gallium phosphide as function of depth from the surface (Fig. 3). We note that the value of S_e for 200 MeV Ag^+ ion irradiation (~1.65×10^4 keV/μm) is lower by a factor of ~1.6 than the S_e of 573 MeV Au^+ ions (~2.73×10^4 keV/μm). Transmission electron microscopic studies of gallium phosphide irradiated with 573 MeV gold ions for the fluence of 1×10^{12} ions cm^{-2} have shown that the gallium phosphide does not melt along the ion trajectory i.e. the threshold value of the electronic energy deposition for the onset of melting was not reached in case of 573 MeV Au $^+$ ion irradiation [5]. In principal, such difference is a clear indication that gallium phosphide does not melt along the ion trajectory at lower S_e (1.65×10^4 keV/μm) value in the present study. The value of nuclear energy loss (4.50×10^1 keV/μm) in the sample surface is three orders of magnitude smaller than the electronic energy loss (1.65×10^4 keV/μm). This result suggests that the electronic energy transferred to lattice via electron-phonon coupling is the mechanism involved in production of hillocks. Other possible reason of elevations could be the lateral mass transport and dewetting induced by ion irradiation on gallium phosphide surface [24]. The density of hillocks evaluated from the AFM data using image analysis software for various ion fluencies is given in Table 1. It is seen from the Table 1 that the density of the hillocks increases with respect to the increase of ion fluence up to 5×10^{12} ions cm^{-2}, after that its value starts decreasing. It is also evident from the table that the density of these hillocks is smaller than the ion fluence. This result suggests that a part of projectiles creates hillocks on the gallium phosphide surface at the places of ion entries into target. Earlier, similar experimental observations of swift heavy ions tracks on the surface of GaAs and Al_2O_3 were reported in literature [25,26]. The change in the surface morphology was also measured in terms of root mean square surface roughness. Variation of root mean square surface roughness with respect to the increase of ion fluence is also given in Table 1. It is evident from the Table 1 that the value of root mean square surface roughness increases gradually up to fluence of 5×10^{12} ions cm^{-2}, after this fluence, it decreased. The increase in the root mean square surface roughness is due to swift heavy ion induced mass transport phenomena through atomic displacements in the surface region, whereas the decrease in root mean square surface roughness at higher ion fluences (⩾1×10^{13} ions cm^{-2}) may be due to the electronic sputtering where the atoms/molecules of the material is electronically excited by incident swift heavy ions [27].

3.2. Raman scattering studies

Fig. 4 showed the first order Raman spectra of non-irradiated and as irradiated samples in the spectral region between 320 cm^{-1} and 440 cm^{-1}. For the non irradiated gallium phosphide sample, the peaks appeared at 402.45 cm^{-1} and 365.26 cm^{-1}