Ferromagnetism in (Ga, Mn) As synthesized by Mn^{+} ion implantation and 5 MeV Si^{++} ion beam induced recrystallization

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*Abstract:*In this study, gallium arsenide samples were first implanted with 325 keV Mn^{+} ions for the fluenceof 2 x 10^{16} ions cm^{-2}. These implanted samples were further irradiated using 5 MeV Si^{2+} ion beams for the fluence of 1 x 10^{16} ions cm^{-2} at a substrate temperature of 350^{0}C for recrystallization. Super conducting quantum interface device (SQUID) measurements on as-implanted sample revealed the paramagnetic behavior. While, after irradiation with 5 MeV Si^{2+} ions, SQUID measurementsshowed the hysteresis loopindicative of the ferromagnetic behavior. Ferromagnetic transition temperature after irradiation of(Ga,Mn)As samples measured from zero field cool and field cool measurements were found to be 292 Kelvin .

***IndexTerms* - GaAs; Mn^{+} implantation; Si^{2+} irradiation; SQUID.**

I. INTRODUCTION

Dilute magnetic semiconductors from III-V group semiconductors when doped with small concentration of magnetic elements showed the ferromagnetism with high Curie temperatures [1-9]. Such materials have attracted much attention in spintronic devices. In order to satisfy the need of spintronic devices, it is also necessary that these materials must exhibit the ferromagnetism at room temperature. Most of the past work on (Ga,Mn)As reported the highest Curie Temperature was about 250 K. The main impediment in the fabrication of dilute magnetic semiconductors for the spintronic application is the solubility limit of transition metal impurities in gallium arsenide. The way to increase the Curie temperature is to increase the manganese concentration. Ion implantation is a versatile non equilibrium technique that can achieve concentration more than their equilibrium solubility limit by means of energetic ion beams. However, in the ion implantation processes create various types of defects in the implanted layers and produce the amorphous layer beyond threshold fluence. The conversion of amorphous layer to crystalline form high temperature annealing is required. Many research groups were met with difficulty in recrystallizing the implanted gallium arsenide samples during annealing processes. Thus ion beam gives a better alternative for converting the amorphous to crystalline form at lower temperature [10]. In the past, fewer studies on recrystallization on (Ga,Mn)As dilute magnetic semiconductors by energetic ion irradiation have been reported in the literatures [11-13]. Mn ions were implanted into p-GaAs substrates at room temperature. Post-annealing was performed using 350 keV He^{+} ion irradiation at a temperature of 150-250 °C. The structure of the films before and after annealing was characterized by X-ray diffraction. The depth profiles of the implanted Mn^{+} were measured by secondary ion mass spectrometry. The results indicated that the Mn^{+} implanted gallium arsenide layer had been epitaxially re-grown without formation of 2nd phase [12]. In this work, gallium arsenide sample was first implanted with 325 keV Mn^{+} ions for the fluence of 2 x 10^{16} ions cm^{-2}. This implanted sample was further irradiated with 5 MeV Si^{2+} ion beams for the fluence of 1 x 10^{16} ions cm^{-2} at a substrate temperature of $350\ ^{0}$C. Manganese provides the localized spins in gallium arsenide. Magnetic properties of this sample have been investigated using superconducting quantum interference device (SQUID) measurements.

II EXPERIMENTAL DETAILS

In the present work, single crystal, undoped semi insulating gallium arsenide wafer with resistivity of $3x10^{7}\Omega$-cm was used as substrate materials. The wafer was cut into pieces typically 1 cm x 1 cm sample. After cleaning, the sample was first implanted with 325 keV manganese ions for the fluence of 2.0 x 10^{16} ions cm^{-2}.During implantation, the ion beam current density was about 50 nA cm^{-2} and the vacuum in the target chamber was maintained at about 10^{-7} mbar. The ion beam was scanned in area of 1 cm x 1 cm by electronic scanner. The projected range of 325 keV manganese ions in gallium aresinide estimated from the Stopping and Range of Ions in Matter (SRIM) was found to be 0.16 µm [14]. After implantation, the sample was further irradiated with 5 MeV Si^{2+} ions for re-crystallization at $350\ ^{0}$C substrate temperature using 1.7 MV Tandetron accelerators at IGCAR, Kalpakkam. It is commonly supposed that on Ga sites silicon form donor and on arsenic site acceptor. The magnetic properties of the as implanted and irradiated samples have been investigated using superconducting quantum interference device magnetometer (MPMSXL-Quantum design Co. Ltd.). For this measurement samples were mounted plane parallel to the applied field. Zero-field-cooled (ZFC) and field-cooled (FC) magnetization curves as function of temperature were recorded for Curie temperature measurement.

III. RESULTS

Magnetization versus magnetic field (M-H) curves of gallium arsenide sample implanted with 325 keV manganese ions for the fluence of 2.0 x 10^{16} ions cm^{-2} and after irradiation with 5 MeV Si^{2+} ions at $350\ ^{0}$C substrate temperature were shown in Figures 1 & 2 respectively. It is seen from the Figure 1 that as implanted sample showed the paramagnetic behavior.

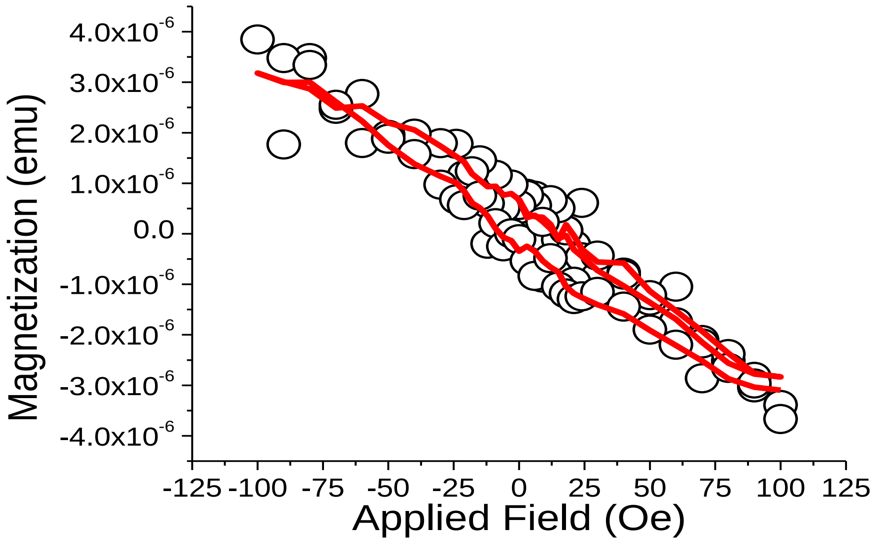

Figure 1: Magnetization (M-H) curve of (Ga, Mn) As prepared with 325 keV Mn^+ ion implantation in gallium arsenide to the fluence of 2.0 x 10^{16} ions cm^{-2}

After irradiation with silicon ions sample showed the hysteresis loop (Fig. 2). This result indicated the magnetic ordering in the sample. It is well known that both the implantation and irradiation processes caused the amorphization and create the various types of defects in the crystals. The characteristics of these defects were found to be depend on the ion nature and target structures. Figure 3 shows the temperature dependence zero fields cooled and field cooled (ZFC-FC) magnetization curves after irradiation with 5 MeV Si^{2+} ions. The Curie temperature estimated from the intercept of ZFC and FC curves [Fig. 3] was found to be 292 Kelvin (close to room temperature). The high value of Curie temperature may be vacancies created by 5 MeV Si^{++} ions in gallium arsenide

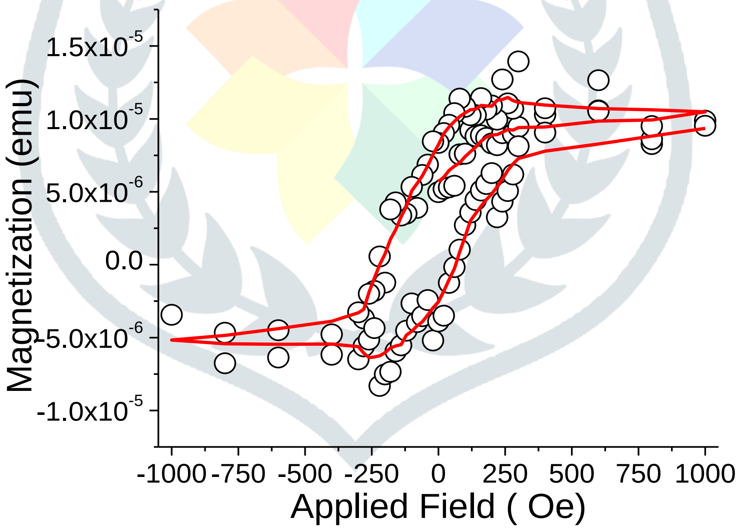

Figure 2: Magnetization (M-H) curves of Ga,Mn)As prepared with 325 keV Mn+ ions implantation in gallium arsenide for the fluence of 2.0 x 10^{16} ions cm^{-2} after irradiation with 5 MeV silicon ions.

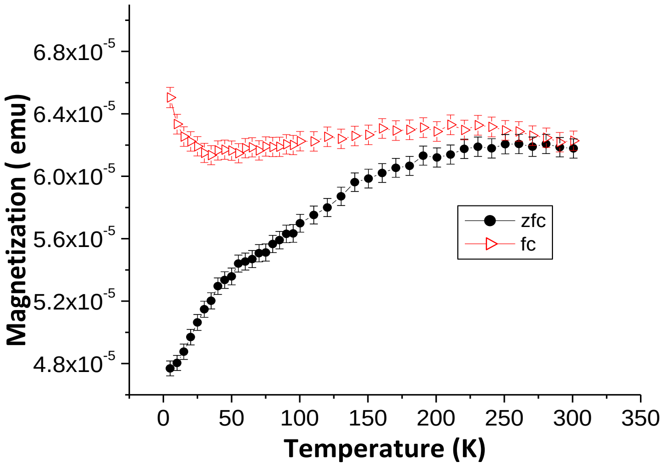

Figure 3: Zero Field Cooled and Field Cooled magnetization curve of (Ga,Mn)As sample after irradiation with 5 MeV silicon ions.

IV. DISCUSSION

The ion fluence required to form the amorphous layer in gallium arsenide can be calculated using the following equation assuming no vacancy diffusion [15]

$$D = E_d N \frac{dE}{dx} cm^{-2} \text{ ---------------------(1)}$$

Where D is ion fluence to drive the target amorphous in ions cm^{-2}, E_d is displacement energy for amorphous (~18 eV), N is the number of target atoms per cm^{-3} (2.21×10^{22} atoms /cm^2) and dE/ dx = 8.90×10^9 eV / cm nuclear energy loss per unit length. Ion fluence estimated from the above equation is found to be 4.46×10^{13} ions cm^{-2}. In our experiments Mn^+ ion implantation fluence, 2.0×10^{16} ions cm^{-2} is higher than the fluence required to form the amorphous layer in gallium arsenide. The formation of amorphous layer and presence of large number of defect complexes showed the paramagnetic behavior in as implanted sample (Fig. 1). In case of 5 MeV silicon ions irradiation in gallium arsenide, the electronic energy loss, nuclear energy loss and projected rage estimated from the SRIM code were found to be 3.16×10^3keV/ µm, 4.12×10^1keV/ µm and 2.42 µm respectively. The variation of nuclear energy loss (S_n) and electronic energy los (S_e) with energy in gallium arsenide for 5 MeV Si^{2+} ions estimated by SRIM is shown in Fig. 4.

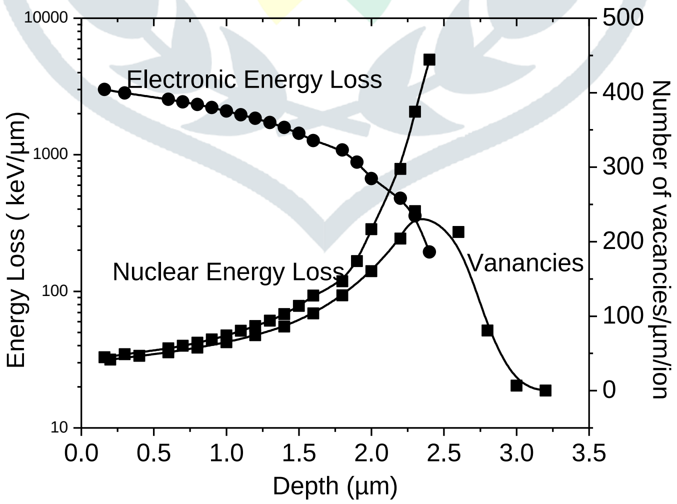

Figure 4: Variation of electronic energy loss, nuclear energy loss and number of vacancies per µm per ion as function of depth for 5 MeV Si^{2+} ions projected on gallium arsenide.

The S_e / S_n ratio for 5 MeV Si^{2+} ions was found to be ~77, suggesting that the mostly crystallization created due to electronic energy loss processes. The range of 5 MeV silicon ions is much higher than the 325 keV manganese ions in gallium arsenide. So that the silicon ions pass through the (Ga, Mn) As and buried in gallium arsenide substrate. The silicon ions during their traverse transfer the electronic energy and nuclear energy to the (Ga, Mn) As surface and deposit the large kinetic energy on the (Ga, Mn) As surface. Ion beam recrystallization is induced due to the migration and recombination caused by the combined effects of nuclear energy loss and electronic energy loss processes. The observed changes in B-H curve (Figure 2) from paramagnetic to ferromagnetic behavior, was created due recrystallization of (Ga,Mn) As. In our experiment irradiation performed at 350 ^{O}C using high currents caused the increase in sample temperature and self-annealing of the implanted layers took place, which leads to the recrystallization of the implanted samples. This may be another reason for recrystallization of the implanted layer. Further, the number of vacancies created by 5 MeV Si^{++} ions in gallium arsenide can be estimated with the modified Kinchin relation [15];

$$N_d(x) = \frac{0.8U(E,x)}{2E_d} \quad \text{----------(2)}$$

Here U (E, x) is the energy transferred to the recoil atoms at depth x from the surface of target. Total number of displacements in the implanted volume is given by;

$$N_d = 0.8\int \frac{U(E,x)}{2E_d} \quad \text{----------(3)}$$

The dependence of displace per atom (DPA(x)) versus depth is given by the following relation;

$$DPA(x) = \frac{0.8U(E,x)}{2E_d N} F \quad \text{--------(4)}$$

where N is the atomic density (atoms cm^{-3}) and F is the ion fluence (ions cm^{-2}). Figure 4 showed the vacancy distribution profile for silicon ion implanted gallium arsenide sample. This displays the number of vacancies produced per unit depth per ion versus depth inside of the target material. The simulation has been performed using Monto Carlo simulation program, stopping and ranges of ions in matter (SRIM) [14]. It is clear that a reasonably good number of vacancies are created after ion irradiation. However, all the vacancies cannot survive because of recombination / trapping process. These vacancies worked as holes in which ferromagnetic coupling are mediated in the valance band.

CONCLUSIONS

In the present work, crystalline gallium arsenide sample implanted with Mn^{+} ions for the fluence of 2 x 10^{16} ions cm^{-2}. SQUID measurements on as implanted sample showed the paramagnetic behavior. After irradiation with 5 MeV Si^{2+} ions, Mn ion implanted gallium arsenide sample showed the transformation from Paramagnetic to ferromagnetic indicated the magnetic ordering of the spin. Ion induced recrystallization is attributed to the migration and recombination of vacancies caused by energy loss processes. The Curie temperature estimated from the intercept of ZFC and FC curves was found to be 292 Kelvin. The result after irradiation with 5 MeV Si++ ion beam, indicated that almost all Mn^{+} ions contribute to ferromagnetic behavior in the sample.

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