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Ferromagnetism in (Ga, Mn) as Synthesized by Mn⁺ Ion Implantation and 5 MeV Si⁺⁺ Ion Beam induced Recrystallization

S K Dubey^{*}

Department of Global Physics, University of Mumbai, Mumba, India

Abstract

In this study, gallium arsenide samples were first implanted with $325 keV Mn^+$ ions for the fluence of $2 \times 10^{16} ions cm^{-2}$. These implanted samples were further irradiated using $5MeVSi^{2+}$ ion beams for the fluence of $1 \times 10^{16} ions cm^{-2}$ at a substrate temperature of 350 0C for recrystallization. Super conducting quantum interface device (SQUID) measurements on asimplanted sample revealed the paramagnetic behavior. While, after irradiation with $5MeVSi^{2+}$ ions, SQUID measurements showed the hysteresis loop indicative 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.

Keywords: Media • Mn+ implantation • Si2+ irradiation • SQUID

Introduction

Dilute magnetic semiconductors from III-V group semiconductors when doped with small concentration of magnetic elements showed the ferromagnetism with high Curie temperatures [1-3]. 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 [4]. In the past, fewer studies on recrystallization on (Ga,Mn)As dilute magnetic semiconductors by energetic ion irradiation have been reported in the literatures. Mn ions were implanted into p-GaAs substrates at room temperature. Post-annealing was performed using $350keV He^+$ ion irradiation at a temperature of $325keV Mn^+$. 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 epitaxial re-grown without formation of 2nd phase [5]. In this work, gallium arsenide sample was first implanted with 325keV Mn⁺ ions for the fluence of $2 \times 10^{16} \textit{ions cm}^{-2}$. This implanted sample was further irradiated with 5 MeV Si2+ ion beams for the fluence of $1 \times 10^{16} ions \, cm^{-2}$ at a substrate temperature of 350°C. Manganese provides the localized spins in

*Address for Correspondence: S K Dubey, Department of Global Physics, University of Mumbai, Mumba, India, E-mail: skdubey@physics.mu.ac.in

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gallium arsenide. Magnetic properties of this samplehave been investigated using superconducting quantum interference device (SQUID) measurements.

Experimental Details

In the present work, single crystal, undoped semi insulating gallium arsenide wafer with resistivity of $3 \times 10^7 \Omega - cm$ 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 $2.0 \times 10^{16} ions \, cm^{-2}$. 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 $1cm \times 1cm$ by electronic scanner. Theprojected range of 325 keV manganese ions in gallium aresinide estimated from the Stopping and Range of Ionsin Matter (SRIM) was found to be $0.16 \mu m$ [4]. After implantation, the sample was further irradiated with $350^{\circ}C$ ions for re-crystallization at $350^{\circ}C$ 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. td.). For this measurement samples were mounted plane parallel to the applied field. Zerofield-cooled (ZFC) and field-cooled (FC) magnetization curves as function of temperature were recorded for Curie temperature measurement.

Results

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

After irradiation with silicon ions sample showed the hysteresis loop figure 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 depending on the ion nature and target structures (Figure 3). Temperature dependence zero fields cooled and field cooled (ZFCFC) magnetization curves after irradiation with 5 MeV Si2+ ions. The Curie temperatureestimated from the intercept of ZFC and FC curves figure 3 was found to be 292 Kelvin (close to

room temperature). The high value of Curie temperature may be vacancies created by $5MeVSt^{++}$ ions in gallium arsenide



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 1016 ions cm-2.



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 1016 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.

Discussion

The ion fluence required to form the amorphous layer in gallium aresinide can be calculated using the following equation assuming no vacancy diffusion [5].

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

Where D is ion fluence to drive the target amorphous in ions cm⁻²

, Ed is displacement energy for amorphous (~18 eV), N is the number of target atoms per cm-3



Figure 4. Variation of electronic energy loss, nuclear energy loss and number of vacancies per μ m per ion as function of depth for 5 MeV Si2+ ions projected on gallium arsenide.

 $2.21 \times 10^{22} atoms/cm^2$ and $\frac{dE}{dx} = 8.90 \times 10^9 eV/cm$ nuclear energy loss per unit length. Ion fluence estimated from the above equation is found to be $4.46 \times 10^{13} ions cm^{-2}$. In our experiments Mn^+ ion implantation fluence, $2.0 \times 10^{16} ions cm^{-2}$ is higher than the fluence required forming 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 figure 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 $3.16 \times 10^3 keV/\mu m$, $4.12 \times 10^1 keV/\mu m$ and $2.42 \mu m$ respectively. The

variation of nuclear energy loss (Sn) and electronic energy los (Se) with energy in gallium arsenide for 5 MeV Si2+ ions estimated by SRIM is shown in Figure 4. The Se / Sn ratio for $5MeVSi^{2+}$ 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 burried 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 of caused by the combined effects of nuclear energy loss and electronic energy 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 OC 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 5MeVSi⁺⁺ ions in gallium arsenide can be estimated with the modified Kinchin relation [6];

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

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

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

$$2 \times 10^{16} ions \, cm^{-2}$$
 ------(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) [7-10]. 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.

Conclusion

In the present work, crystalline gallium arsenide sample implanted with Mn+ ions for the fluence of $2 \times 10^{16} ions \, cm^{-2}$. SQUID measurements on as implanted sample showed the paramagnetic behavior. After irradiation with $5MeVSi^{2+}$ 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 $5MeVSi^{++}$ ion beam indicated that almost all Mn^+ ions contribute to ferromagnetic behavior in the sample.

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Conflict of Interest

None

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