ABSTRACT

For the evolution of magnetic storage devices, the conventional charge-based devices were superseded by spin-based devices. This new generation of devices involve spin which is the root cause of magnetism and an intrinsic angular momentum that a particle cannot lose or gain. Magnetic materials have the ability to remember their spin state for a very long time. On the other hand, semiconducting materials with their charge carriers could help in the transfer of information. This prompted to combine the spin and charge property of electron onto a single material that would simultaneously help in the processing of information as well as storing them for a considerable time. In short, the material incorporates features of both semiconductor and magnetism giving rise to a new class of materials known as Dilute Magnetic Semiconductor (DMS). They consist of a semiconductor, where fraction of their original atoms are substituted by magnetic elements making them semi magnetic in nature. This leads to a merger of magnetic properties along with semiconducting properties.

The main objective of developing and studying DMS materials is to achieve Curie temperature T_C greater than the room temperature as it is desirable in order to sustain ferromagnetism for a longer duration in practical device applications. Care must be taken to enhance the magnetic properties without affecting the semiconducting properties.

Magnetic semiconducting materials can be used in a variety of electronic devices incorporating the spin property of charge carrier [33-35]. Semiconductor memory devices like Magneto-resistive random access memory (MRAM), computer hard disk, spin transistors, spin valves, etc. are some areas of its application where conventional ferromagnetic metals have failed to deliver desired spin polarized carriers.

Taking into consideration the requirement a DMS system serves, it becomes imperative to come up with new materials that can have high T_C that

would help sustain ferromagnetism for as long as possible. Since research on elemental and group IV-VI semiconductors are found to be limited in the sense of being a DMS material and with a view to realize room temperature ferromagnetism, for the present work, elemental semiconductor tellurium and two compound semiconductors, SnSe and SnTe, belonging to group IV-VI are chosen as the materials for research. Transition metal Fe doped bulk alloys of these semiconductors having the form $(Te)_{1-x}Sb_x$, $(SnSe)_{1-x}Sb_x$ and $(SnTe)_{1-x}Sb_x$ are made wherein antimony, a group VI element is substituted which acts as the source of supplementary charge carriers in the system.

The thesis consists of chapters that covers the background information, motivation and objectives of the present work, synthesis methodology and experimental techniques used, the obtained results, and conclusions drawn from them. The chapters are as follows:

Chapter I Introduction:

In this chapter, a brief introduction of elemental and compound semiconductors that belong to the groups II-VI, III-V and IV-VI, with a specific focus on IV-VI group along with an introduction to dilute magnetic semiconductors (DMS) has been presented. Additionally, various theoretical models predicting the origin of magnetism in the DMS system is also discussed. Further, along with the description of the materials under consideration, the motivation and objectives of the present work has also been outlined.

Chapter II Experimental Techniques utilized for characterizing the materials:

This chapter describes the methodology of bulk sample preparation for the present study. Also, details of experimental techniques adopted to characterize the samples are also explicitly explained.

Chapter III Study on hole impurity triggered magnetism in Fe doped $(Te)_{1-x}Sb_x$ bulk alloys:

This chapter focuses on elemental semiconductor Tellurium which is doped with dilute amount of transition element Fe. Its functionality as a DMS system has been explored alongside the substitution of group V element Sb which acts as a hole dopant/acceptor impurity. The doping amount of Fe is limited to 0.05 in order to avoid formation of Fe clusters or other undesirable magnetic phases. The general sample composition is represented as $Fe_{0.05}(Te)_{1-x}Sb_x$ where x = 0, 0.01, 0.03 and 0.05. Hexagonal structure of Te is confirmed from the X-ray Diffraction spectra with presence of no additional peaks. The electrical resistivity data and plot of temperature coefficient of resistance confirms the semiconducting nature of the samples. Transport property of the samples are explained on the basis of small polaron hopping (SPH) model and variable range hopping (VRH) model. A crossover from negative to positive trend is observed in the magnetoresistance plot of the samples as temperature reaches 100 K for x = 0, 0.01 and 0.03 samples. Since Tellurium is diamagnetic in nature, doping of Fe brings about paramagnetic feature in the sample. However, with the substitution of Sb, presence of weak ferromagnetic ordering is perceived from the magnetic studies which can be regarded as an interesting behaviour. Further interpretation of magnetic properties is also discussed in the chapter.

Chapter IV Effect of substitution of non-magnetic impurity Sb on ferromagnetism in dilute Fe doped SnSe:

In this chapter, work on group IV-VI semiconductor SnSe, doped with Fe followed by a co-doping of Sb having a general form $Fe_{0.05}(SnSe)_{1-x}Sb_x$ where x = 0, 0.03 and 0.05 is presented. The samples are prepared by sealing the quartz tube under vacuum and then melting the contents in an oxy-butane flame. An elaborate explanation and illustration of different studies carried out to get an understanding of the underlying properties has been elucidated in the chapter. Studies include structural, surface morphology, optical, R-T measurements in the presence of applied magnetic field, M-T and M-H measurements, magnetic memory effect, etc. A slight variation in the direct and indirect band gap values is observed from that of pristine SnSe due to Fe doping and addition of varying concentration of Sb. Resistivity measurements show a transition from metallic to semiconducting nature in x = 0 sample whereas the other two samples show a purely metallic nature. In addition to discussing results from magnetization measurements, the findings of magnetic memory effect measurement is also presented that is carried out to check whether the samples could remember their past spin.

Chapter V An interplay between ferromagnetic and magnetic glassy state in Fe doped $(SnTe)_{1-x}Sb_x$ bulk alloys:

This chapter discusses work on dilute Fe doped SnTe bulk alloys having general form $Fe_{0.05}(SnTe)_{1-x}Sb_x$ where x = 0, 0.01, 0.03 and 0.05. The alloys are prepared by varying the concentration of Sb to see the effect of donor impurities on the properties of samples. The samples are characterized for their structural, optical, electrical resistivity, magneto-resistance and magnetic properties. The XRD pattern of all samples show Rock salt crystal structure. The band gap values calculated from the FTIR spectrum also showed unusual variations with a decreasing as well as increasing trend. The electrical resistivity data show a transition from metallic to insulating nature in x = 0 sample. However, with the substitution of Sb, the resistivity values are found to decrease with temperature thereby showing semiconducting nature. Magnetoresistance plot of all the samples at different temperature values is seen to show a positive trend with the maximum value undergoing a decrease with temperature. In addition, detailed analysis of magnetic studies is also done in this chapter where x = 0.03 sample shows magnetic glassy behaviour. This glassy behaviour is confirmed through

AC resistivity measurements and calculations pertaining to the confirmation has been presented. The samples are also found to showcase ferromagnetic behaviour at room temperature.

Chapter VI Summary:

In this chapter, an overall summary and conclusion derived from the work carried out on Fe doped $(Te)_{1-x}Sb_x$ bulk alloys, $(SnSe)_{1-x}Sb_x$ bulk alloys and $(SnTe)_{1-x}Sb_x$ bulk alloys is presented. The prospects of extending the above work in terms of device making that could be utilized for different applications is also discussed.