Magnetic Circular Dichroism Measurements of (Ga,Mn)As Epilayers

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Abstract

The III-V diluted magnetic semiconductor (Ga,Mn)As is of interest in the field of spintronics due to its semiconducting and ferromagnetic properties. This research probes the optical behavior of (Ga,Mn)As under the influence of an external magnetic field. The Zeeman splitting of the band structure as a result of the applied magnetic field is enhanced by the magnetic moment of Mn^{2+} *through the sp-d exchange interaction in (Ga,Mn)As, however it can be difficult to probe the Zeeman splitting directly in this material. Magnetic Circular Dichroism (MCD) is a powerful method to investigate the field-induced modification of the electronic band structure of (Ga,Mn)As. This study presents MCD data as well as SQUID data of the temperature dependence of magnetization for several (Ga,Mn)As samples.*

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1 Introduction

The electron has two fundamental properties: charge, and spin. In the field of semiconductor physics, scientists often manipulate the electron's charge in order to process, carry, and store information. This is so common and so useful that many modern-day conveniences are based on it; cellular phones, laptops, and mp3 players are just a few of the devices which rely on semiconductors. If control of the electron's charge can lead to many beneficial applications, it is only natural that scientists have begun to ask whether it is possible to manipulate the electron's spin, along with its associated magnetic moment. This question led to the development of the field of spintronics, which studies semiconductors with magnetic properties.[4] Ultimately, being able to control both charge and spin will lead to faster information processing, multiple functionalities and greater data storage capacities.

The applications of such magnetic semiconductors, however, have been limited thus far due to the lack of a semiconductor compound which retains its magnetic properties (e.g., ferromagnetism) at room temperature. Diluted magnetic semiconductors (DMS)[1], and particularly ferromagnetic semiconductors, have attributes which make them promising candidates for future research in this respect. Gallium Manganese Arsenide – (Ga,Mn)As -- which is the primary focus of this research, is a good example of a compound that falls into this class.

Fig 1.1 (Left) A (Ga,Mn)As sample that has undergone chemical wet etching and a coating process. (Right) A (Ga,Mn)As sample as grown.

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[1] Diluted magnetic semiconductors are compounds in which a fraction of nonmagnetic cations are substituted by magnetic ions, which are typically transition metals. [5] A common notation for such semiconductors includes the magnetic ion concentration as a variable, i.e. GaMnAs would be denoted $Ga_{1-x}Mn_xAs$.

2 Theoretical Model of Semiconductors

Semiconductors are neither good conductors of electricity, nor are they good insulators. These compounds do, however, have characteristics which make their electrical resistivity highly sensitive to external factors such as light exposure, heat, or the application of electric and magnetic fields. The electronic band structure[2] of a given compound determines its conductivity: a smaller energy gap between the valence band and the conduction band corresponds to a better electrical conductivity. This is explained in more detail below.

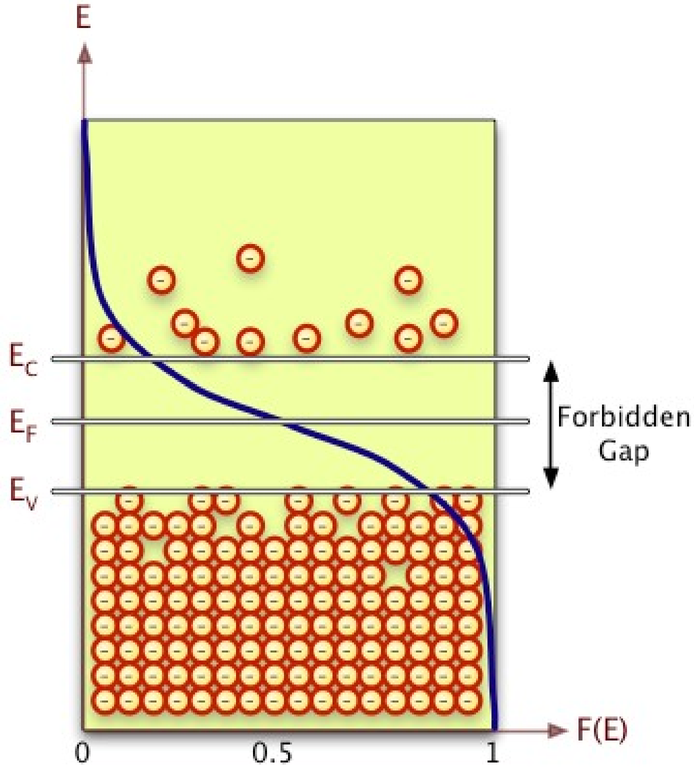

Fig 1.2[6] A simple diagram of energy levels in a semiconductor. The maximum energy of the valence band is denoted E_v, while the minimum energy of the conduction band is denoted E_c. Between the two regions is a forbidden energy zone where no electrons reside. The blue function is the Fermi Function, F(E), which is the probability that a state with a particular energy is occupied by an electron, given that the energy is allowed. Note that the Fermi energy lies at the midpoint of the bandgap only for intrinsic semiconductors.

If an electron in the valence band receives enough energy to jump the energy gap (E_C-E_V in the diagram above), it is able to transition into the relatively empty conduction band. The flow of electrons in the conduction band constitutes a current.

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[2] In solid compounds, there exist a large number of atoms with numerous energy states that are very near each other; these states can be approximated by a region of allowed energy called an energy band. In a semiconductor, the valence band is the last energy band that is filled with electrons, whereas the conduction band is the next highest band in terms of energy which is completely devoid of electrons.

When an electron leaves its place in the valence band, it leaves behind it a hole. A valence electron from a nearby atom is then free to take its place, and thus it in turn will leave a hole in its wake. It is fairly easy to envision this hole, which behaves much like a positive charge, moving through the valence band of the semiconductor compound, with the electrons flowing in the opposite direction. For simplification, physicists treat these holes as positive charges.

With this simple model of a semiconductor in mind, there are two very straightforward ways of improving the conductivity of a sample: either increase the number of loosely bound electrons in the valence band, or increase the hole concentration. Scientists achieve this by doping[3] semiconductor compounds with electron donors or electron acceptors. In p-type (or positive-type) doping, an element with fewer electrons in its valence shell than the valence shell of the semiconductor compound is substituted in place of one of the elements of the crystal lattice. This creates an excess hole concentration. In n-type (or negative-type) doping, an element with more electrons in its valence shell than the valence shell of the semiconductor compound is substituted. This creates an excess of weakly-bound outer electrons that can easily be excited into the conduction band.

The GaAs in this study has been doped with Mn^{2+} ions in a p-type doping process. These Mn ions play a dual role in altering the properties of GaAs: they improve the electrical conductivity of the material by increasing hole concentration, and they introduce a localized magnetic moment of $^{5}/_{2}\,\mu_B$.[2] Although the dominant interactions between the Mn^{2+} ions are themselves antiferromagnetic, they are able to pair with free charge carriers, which are holes in the case of (Ga,Mn)As. This coupling leads to ferromagnetic order via the p-d exchange interaction.[4]

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[3] Doping is a process that adds impurities to intrinsic (pure) semiconductor compounds in order to either increase or decrease hole concentration.

[4] See Titova LV. 2004. Optical Studies of Low-Dimensional Magnetic and Non-Magnetic Semiconductor Structures. p. 93-95. [2] for a more thorough discussion of RKKY Theory and the p-d exchange coupling.

3 Experimental Setup

The experiment was performed in two distinct phases. The preparation of the (Ga,Mn)As samples in this study involved molecular beam epitaxy (MBE) growth, an annealing process, and chemical wet etching. The analysis of the samples involved magnetic circular dichroism measurements and absorption spectrum measurements.

MBE Crystal Growth

Molecular beam epitaxy is a method of growing thin crystalline films that allows scientists a high degree of control over the growth conditions. In the process, molecular beams of the desired elements are incident on a heated substrate in an ultra-high vacuum of around 10^{-11} torr.[1] This vacuum is achieved through the use of the combination of vacuum pump and cryotrapping.[5] Layers are deposited on the substrate epitaxially at a rate of around 1 μm per hour, or about 1 atomic monolayer per second.[1]

(Ga,Mn)As crystallizes in the zinc-blende structure. This consists of two interpenetrating face-centered cubic lattices dispersed from one another by one-quarter of a body diagonal.

When incorporating Mn into the GaAs lattice, Mn^{2+} ions are generally substituted in the place of Ga atoms, to form Mn_{Ga}. However, some fractional Mn can also form interstitials[6], denoted Mn_I, or can form Mn clusters. Post-growth annealing has been found to reduce the number of Mn_I, leading to an increase in the Curie temperatures of the samples and improved transport properties.[2]

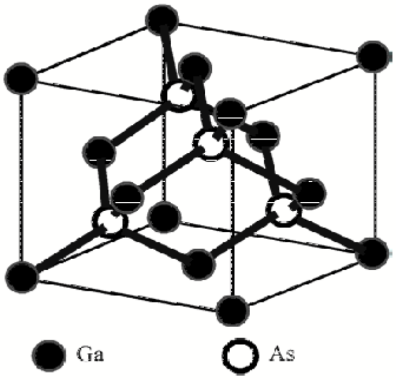

Fig. 3.1[2] The unit cell for GaAs

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[5] Cryotrapping refers to a process of trapping molecules on a cold surface as a result of the loss of kinetic energy due to collisions with that surface. In MBE, the walls of the chamber are cooled using liquid Nitrogen.

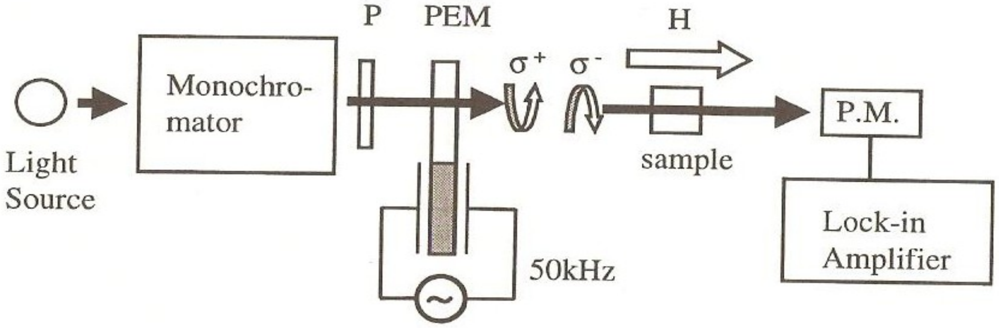
[6] Interstitials form either when Mn substitutes for an As atom rather than Ga, or when Mn occurs at the center of a Ga or As face. These generally occur with less than ~20% Mn.

The (Ga,Mn)As samples grown for this experiment use low-temperature MBE technique. First, the (100) GaAs substrate was heated to approximately 600ºC to allow desorption of the oxide layer. Next, a 100 nm GaAs buffer layer was grown on the substrate to smooth the surface, followed by a 250 nm (Ga,Al)As layer (Al~30%) as an etching stop layer at 600ºC. Finally, the substrate was cooled to 250ºC and a layer of (Ga,Mn)As was grown to a thickness of between 100 nm and 1 μm. Note that some samples were co-doped by Beryllium (Be) in order to further tune the hole concentration.

Magnetic Circular Dichroism Setup

MCD spectroscopy measures the difference in intensity between σ^+ and σ^- (or left and right) circularly polarized light transmitted through a DMS under the influence of a strong external magnetic field. The setup for this is shown in the schematic below.

Fig. 3.2[3] Schematic of MCD setup



The monochromator in the diagram selected single wavelengths of light which then passed through a linear polarizer (P) and a photoelastic modulator (PEM). The (Ga,Mn)As samples were mounted on a sample holder and placed into an Oxford cryostat which also housed a superconducting magnet. This system was cooled using liquid He and an insulating layer of liquid N_2 to a temperature of 1.7K. An external magnetic field (H) of maximum strength 5T was applied in the direction of propagation of the circularly polarized light. The light transmitted through the sample passed through a photomultiplier (P.M.) and the signal was sent to a lock-in amplifier, at which point our computer would measure the signal as a function of the wavelength of light.

4 Data

The SQUID[7] data of the temperature dependence of magnetization for four different samples of (Ga,Mn)As is presented below. Observe that the Curie temperatures (T_C) of the samples can be found at the point where the magnetization reaches zero.

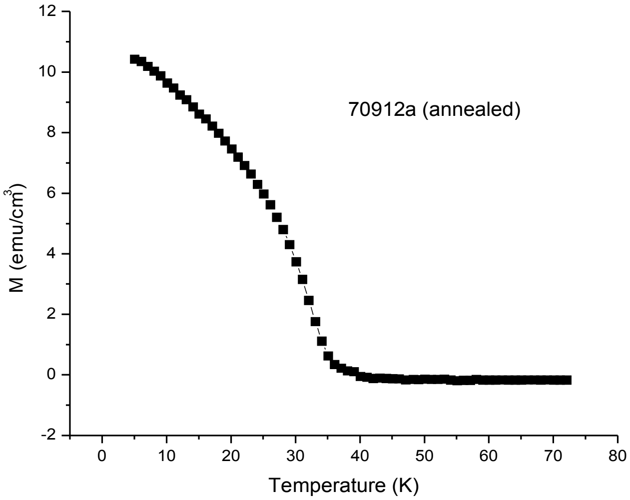


Fig 4.1 $T_C \sim 34K$, $T_{Be} \sim 1020$ºC

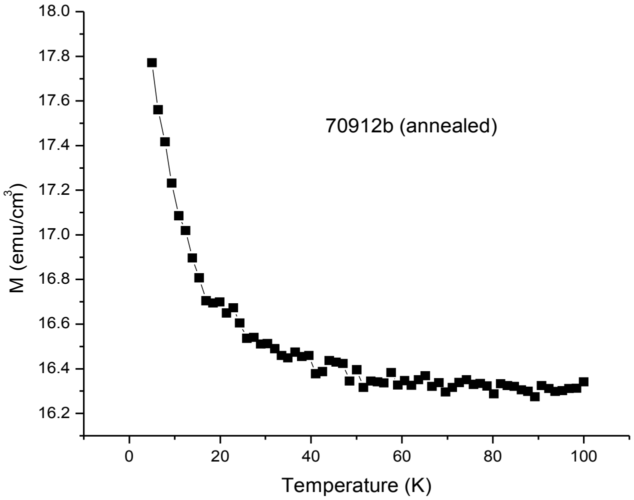


Fig 4.2 $T_C \sim 25K$, sample was not codoped with Be

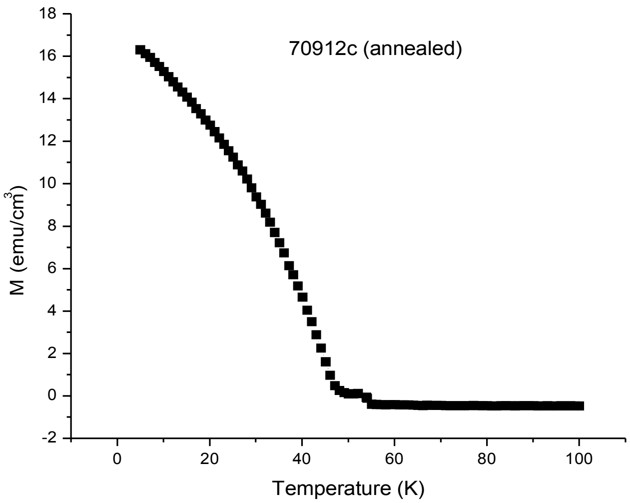


Fig 4.3 $T_C \sim 43K$, $T_{Be} \sim 950$ºC

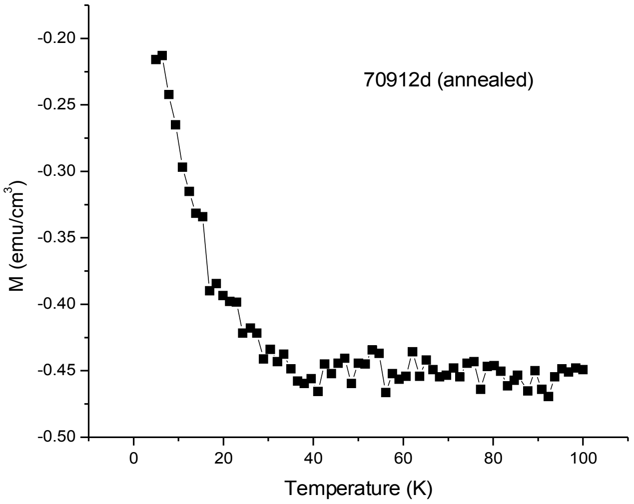


Fig 4.4 $T_C \sim 27K$, $T_{Be} \sim 1040$ºC

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[7] Superconducting Quantum Interference Device

Sample MCD Data is presented below. The sample series 70912 from the data above was coated with an anti-reflection material for MCD measurements to reduce the effects of thin-film interference.

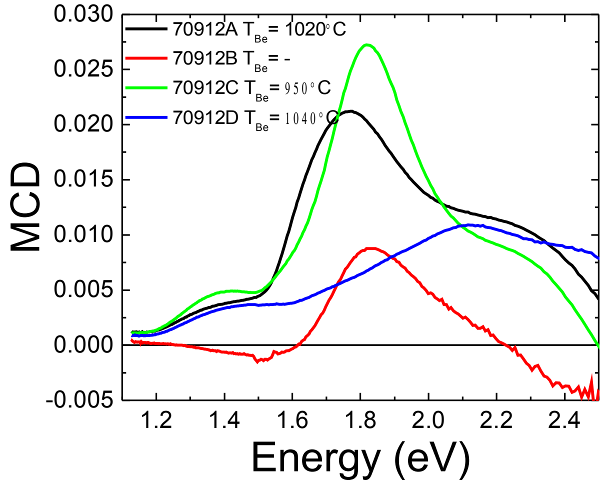

Fig 4.5 MCD for the 70912 series, taken at B = 5T. The MCD is the difference in transmission intensity between left- and right- circularly polarized light; it is in arbitrary units. Initially MCD is measured as a function of the wavelength of light generated by the monochromator, however it is more useful to put this into terms of energy using the relation E = hc/λ.

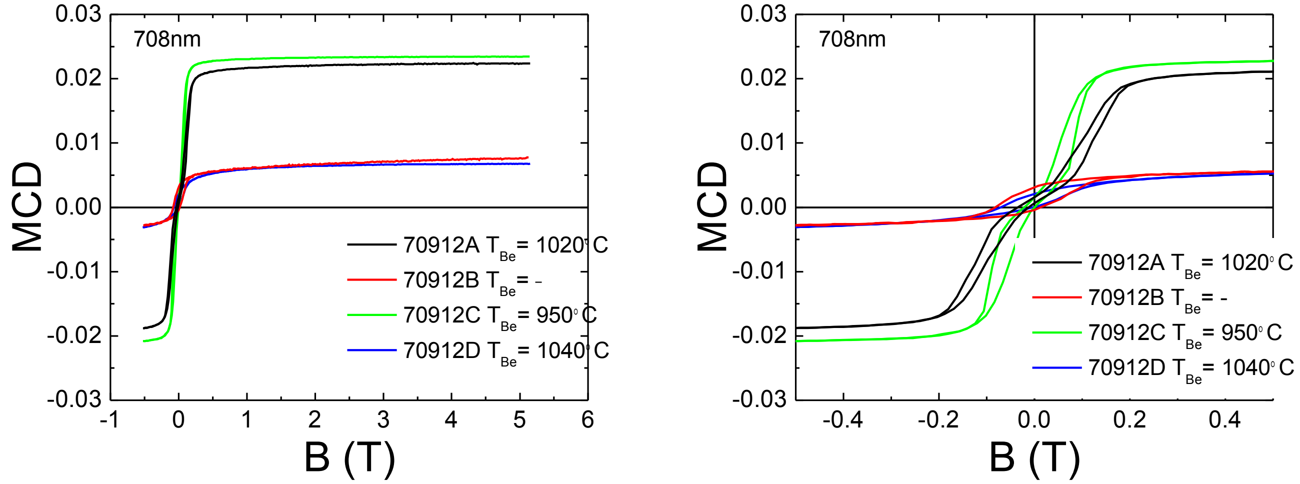

Fig 4.6 The hysteresis loops[8] for the 70912 series, measured between -0.5T and 5T.

Fig 4.7 A closer look at the hysteresis loops for the samples, measured between -0.5T and 0.5T.

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[8] A hysteresis loop measures magnetization in the sample as a function of the externally applied magnetic field.

5 Discussion

The Curie temperatures of the four samples in the SQUID measurements appear to correspond closely to Tbe (a higher temperature indicates a higher Be concentration in the sample). Sample 70912B, which was not codoped with Be, and sample 70912D, which has the highest concentration of Be, have lower Curie temperatures than the other samples. It would seem that increasing the Be concentration improves the Curie temperature of (Ga,Mn)As:Be only up to a certain point, and further increasing of the Be concentration will decrease the T_C. Further experimental verification is needed for this hypothesis.

Fig 4.5 reveals several interesting properties of the samples studied. First, it appears that the peak MCD intensity corresponds to the Be concentration in a way that is analogous to the discussion above. Second, all of the samples except 70912D have peak intensities at approximately 1.8 eV. This suggests that the bandgap occurs near this energy. It is possible that the Be concentration in the sample 70912D is too high for the sample to retain the electronic band structure of (Ga,Mn)As.

In Fig 4.6 and Fig 4.7, hysteresis loops were measured at 708 nm, near where the MCD peaks at 1.8 eV. The width of each of the hysteresis loops contains information about the ferromagnetic properties of the samples. A narrow loop indicated weak ferromagnetism, as displayed by samples 70912B and 70912D. A wider loop indicates stronger ferromagnetism, as in 70912A and 70912C.

6 Future Work

Future work will involve a thorough analysis of the MCD, SQUID, and absorption spectrum measurement data from several samples of (Ga,Mn)As. It is hoped that a thorough characterization of the optical behavior and transport properties will lead to a better understanding of the electronic band structure and ferromagnetic properties of these semiconductors, ultimately yielding many applications in material science.

7 Acknowledgements

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