ABSTRACT

GROWTH, MAGNETIZATION AND TRANSPORT PROPERTIES OF Co$_2$FeAl-BASED MULTILAYERS

by Rabindra Pahari

Circular transfer line method was used to study sputtered Co$_2$FeAl (CFA) full Heusler alloy ferromagnetic films. These features were used to study various effects such as tunneling magnetoresistance (TMR), giant magnetoresistance (GMR), anisotropic magnetoresistance (AMR). X-Ray Diffraction (XRD) data show that magnetron-sputtered CFA is poly-crystalline in structure. Scanning electron microscope and energy dispersive x-ray were used to characterize the topology of sample and the concentrations of the elements present in the sample. The magnetization was observed at different temperatures and a large MR of 13% was observed at 5K in TMR structured sample. The magnetization switching in two electrodes separated by a thick oxide spacer used in TMR structure shows no coupling between electrodes whereas typical thickness of oxide spacer for TMR study shows trace of coupling between electrodes. 36% spin valve giant magnetoresistance was observed in CFA/Aluminum/Permalloy multilayers and 19% AMR was observed in CFA/Al bilayer. The multilayer was further examined for in plane and perpendicular to plane magnetoresistance.
GROWTH, MAGNETIZATION AND TRANSPORT PROPERTIES OF Co$_2$FeAl-BASED MULTILAYERS

A Thesis

Submitted to the

Faculty of Miami University

In partial fulfillment of

The requirements for the degree of

Master of Science

Department of Physics

By

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2015

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Acknowledgements

I humbly acknowledge those who directly or indirectly inspired me to do research and Miami University. Specially, Dr. Khalid Eid who gave me platform to horn my experimental skill and provided constant guidance throughout this work. It is worth to mention the contribution from Dr. Mahmud Khan who helped me to collect the data in this work. I cannot forget contribution from my wife who was also a part of my encouragement and constant support throughout this work and my son with whom I used to enjoy after I got tired of work to refresh myself for next day.

Rabindra Pahari
Chapter 1

Introduction

1.1 Spin Polarization

The Spin is inseparable from magnetism and plays a crucial role in the study of magnetic materials. Spin is a fundamental property of elementary particles which constitute the atom and hence matter. Basically, the spin gives rise to the magnetic moment and so magnetization of the material. The spin of an electron can be either positive (taken as up) or negative (taken as down) relative to an applied magnetic field. Spin polarization is a degree by which more of the spins of atoms and/or electrons are aligned in either the up or the down direction relative to the applied magnetic field.

Different methods to measure spin polarization are proposed and the spin polarization definition also varies\(^1\). The most popular method to measure the ‘transport’ spin polarization is

\[
P = \frac{D_{\uparrow}(E_F) - D_{\downarrow}(E_F)}{D_{\uparrow}(E_F) + D_{\downarrow}(E_F)}
\]

With, \(D_{\uparrow}(E_F)\) is the density of states (DOS) of spin up electrons at the Fermi level \((E_F)\) and \(D_{\downarrow}(E_F)\) is the DOS of spin down electrons at \(E_F\).

The alignment of magnetic moments in matter gives rise to the different types of magnetic materials. Whenever the magnetic moments of constituent atoms are aligned randomly due to thermal agitation then such material will not have a magnetic moment in any particular direction so that the net spin polarization is zero. This type of magnetic materials is called paramagnetic materials [fig. 1 (a)]. The next category of magnetic materials that plays an important role in magnetism is ferromagnetic materials. In this type, the electron spins are aligned in a particular direction without applied field giving rise to a non-zero total magnetic moment in a particular direction. This happens when the density of states corresponding to spin up and spin down electrons are different at the Fermi level [fig.1.b]. A unique type of materials showing interesting spin behavior is the half metals. What makes this kind of metal interesting is that the density of states available at the Fermi level is nonzero for only one type of spin, either up or down. This case can be understood as the material is a metal or conductor for one kind of spin and is an insulator for the other kind of spin [fig. 1.c]
Fig. 1: Cartoon for density of states in a) paramagnetic b) ferromagnetic and c) half-metal material.²

1.2 Two promising materials for Spin Polarization

1.3 Co₂FeAl Heusler alloy

A New era of spintronics started with the discovery of Heusler alloys³. Their electronic properties vary depending upon the structure of the alloy itself. They are found to exist as half metallic ferromagnetic, antiferromagnetic, non-ferromagnetic metals, semiconductors⁴, as well as superconductors. Heusler alloys, ternary intermetallic compounds, are classified into two categories: half-Heusler alloys and full-Heusler alloys. Those alloys having structure XYZ are referred to as Half-Heusler (HF) alloys and those with structure X₂YZ are referred to as Full-Heusler (FH) alloys⁵. Where X and Y are transition metals and Z is main block element, in some cases Y can be a rare earth element or an alkaline earth metal. The metal which comes twice is usually put at the beginning of the formula and the main group element at the end. Following the color scheme in fig. 2, a large number of Heusler alloys can be formed. Half Heusler compounds crystalize in non-centrosymmetric cubic structure with space group 216 and Wyckoff positions are (0,0,0), (0.5, 0.5, 0.5) and (0.25, 0.25, 0.25 ) respectively for X, Y and Z atoms. This structure is
more known as three interpenetrating face centered cubic sublattices each of which is occupied by X, Y and Z atoms respectively. Half-Heusler crystal structure can be visualized as ZnS structure of which the octahedral site is occupied. The electronic properties of this kind of materials are dominated by the covalent bonding between two of the atoms. On the other hand, Full Heusler compounds crystallize in cubic space group 225 and corresponding Wyckoff positions are (0.25, 0.25, 0.25), (0, 0, 0) and (0.5, 0.5, 0.5) for X, Y and Z elements respectively. Full-Heusler structures consist of four interpenetrating FCC sublattices, of which two are occupied by X atom and NaCl type lattice structure is formed by Y and Z atoms. Due to the Ionic character of their interaction, these atoms occupy octahedral positions and X atoms occupy tetrahedral positions. The Co₂FeAl Heusler alloy consists of four FCC sublattices with space group Fm3m and occupying Wyckoff positions as follows: Cobalt atoms at (0, 0, 0) as well as (0.5, 0.5, 0.5), Iron atoms at (0.25, 0.25, 0.25) and Aluminum atoms at (0.75, 0.75, 0.75) respectively.

The magnetic moments of Heusler alloys are calculated from the Slater-Pauling rule. The magnetic moment of the binary alloys and ternary alloys of 3d elements are predicted by the valence electron per atom (Nᵥ). Slater-Pauling model divides materials into two groups. First with Nᵥ ≤8, having localized magnetism. Second with Nᵥ ≥8, having itinerant magnetism. Heusler compounds fall in the category of localized magnetism. Slater-Pauling curve for some transition metals and alloys is as shown in fig. 3. According to this rule, the magnetic moment of compounds in integer multiples of Bohr magneton (μᵥ), per unit formula unit is given by,

\[ m = Nᵥ - 2n \]

Where, n is the number of minority electrons. Neglecting s and p electrons, the number of minority electrons in the d band is forced to be 3 due to a minimum in minority density of states. So, the above formula for the magnetic moment per unit formula unit can be reduced to

\[ m = Nᵥ - 6 \]

Now for XYZ type Heusler alloys, having three atoms in the formula unit, this moment can be given as

\[ m = Nᵥ - 18 \]

and for X₂YZ type Heusler alloys, having four atoms in each formula unit, this moment will be

\[ m = Nᵥ - 24 \] (2)
1.4 Bi$_2$Se$_3$ Topological Insulator

The insulators, which are materials that do not conduct electricity, can be divided into two types. First are those whose valence band and conduction band are separated by large energy gap and hence no electrons can be made available to flow through the conduction band, like diamond. Second are those who permit electrons to flow only through their surface and not through their bulk and are called topological insulators. In the second kind of insulators the energy gap between the two bands is modified due to the spin-orbit interaction. This interaction in these insulators is so strong that the insulating energy gap is inverted, meaning that those states which should be above the gap will appear to be below the gap connected by a ‘Dirac cone’. Furthermore, the energy level of a normal insulator consists of a parabola in momentum space with the maximum occupied energy level known as Fermi level. Two electrons in such state can occupy the same energy level subject to Pauli Exclusion Principle. In contrary with normal insulators, the topological insulator energy levels form two bands that are like a cone in nature and are connected by tip to tip (fig. 4)
Fig. 4: Energy band diagram of a Topological insulator. The valence band and conduction band of the surface states are connected by edge states and hence the surface is conductive while the bulk is not.

Topological insulators are the materials whose surface is a good conductor of electricity whereas the bulk is insulator. This is possible due to the edge states effect. Those kinds of materials are predicted to have a wide range of weird applications such as hiding submarines, quantum computers, fully efficient spintronic devices.

To understand edge effects we have to go back to the late 1980’s and dig into Integer Quantum Hall Effect. When an electron in a semiconductor is subjected to a strong magnetic field, the quantum Hall conductance becomes $\nu e^2/h$, where $\nu$ is an integer quantity called filling factor. The motion of such an electron in uniform magnetic fields can be compared with a harmonic oscillator in quantum mechanics with quantized energy levels as, $(n+1/2)h\omega_c$, where $\omega_c = eB/2\pi m$ is the cyclotron frequency. Those energy levels are called landau levels and are degenerate. Filled landau levels occur at $\nu=1$. The radius of such a circular electron orbit becomes, $R_n = \sqrt{\frac{h}{2\nu eB}} (2n + 1)$.

When an electron reaches the boundary it bounces back forming a conducting channel along the boundary called edge state. Each filled landau level forms one conducting channel. So for discrete landau levels, there will be a series of conducting channels, which is the characteristic behavior of a topological phase.

The electron spin plays a crucial role in magnetism. In magnetism applications, when the net spin in small region is in a given direction it corresponds to storing a ‘0’ and if it is in the opposite
direction that stores a ‘1’ in the computer’s hard disc. When we flow an un-polarized current on the surface of topological insulator, the spin up electrons flow towards one side of the material and spin down electrons flow towards the other side, due to spin momentum locking. The time reversal symmetry prevents edge spin current from backscattering.

1.5 Methods to measure spin polarization
Depending upon the nature of the material in magnetic multilayers, there are different ways to measure spin polarization (SP). For ferromagnetic metal /Insulator/ferromagnetic metal (F-I-F) type of multilayer the most famous way to measure SP is the tunneling magneto-resistance (TMR). For metal/Superconductor (N-S) type of multilayer Andreev Reflection (AR) is an easy and powerful way to extract the SP. Below we will focus on TMR to measure spin polarization. There are still other ways to measure SP depending on the type of multilayer and sample, which are out of the scope of this thesis.

1.6 Tunneling Magneto-resistance (TMR)
A well-known application of magnetic materials in spintronics is the magnetic sensor device, which can be realized with the multilayers separated by a non-magnetic metal. This picture can be well understood as tunneling magneto-resistance (TMR), defined as,

\[ TMR = \frac{\Delta R}{R} = \frac{R_{ap} - R_p}{R_p} = \frac{2P_1P_2}{1-P_1P_2} \]  

(3)

Where \( R_{ap} \) and \( R_p \) are the resistance of the tunnel junction when the spins of the two magnetic films are antiparallel and parallel respectively. The terms \( P_1 \) and \( P_2 \) are polarization of each electrodes defined in equation (1)\(^\text{12}\).

Studies of Tunneling magneto-resistance of molecular beam epitaxy (MBE)-grown full Heusler alloy electrodes with MgO barrier\(^\text{13}\) found high values of TMR ratio. The temperature dependent TMR ratio was 386\% and 832\% at 300K and 9K respectively. Tunneling properties of Sputter deposited Heusler alloys have also been studied\(^\text{14}\). The Tunneling magneto-resistance (TMR) ratio in Co\(_2\)FeAl (CFA)/MgO/CoFe electrode was found to be 700\% at 10K and 330\% at room temperature. The study concluded that the high value to TMR is due to coherent tunneling. The spin polarization of CFA was found to be 0.75.
Chapter 2

Experimental setup and Methods

2.1 Sample preparation for Andreev Reflection (AR) measurement

Co$_2$FeAl Sputtering target with desired stoichiometry was made in the lab of Prof. Mahmud Khan. The target had an oval shape of dimensions at the widest circle diameter of 1”. The sample is then cut through major axis of the oval to get two halves. Those halves are then polished with sand paper. The 3 mm thick half oval was brought down to a 0.45 mm thickness disc. We checked the target for sputtering process with different thickness but we could not establish sputtering process with thick sample, but 0.45 mm thick sample worked fine in the magnetic sputtering process. Comparison of two halves is as shown in fig. 5. A disc of Silicon wafer with 5 inch diameter and 0.5 mm thickness was cut into pieces and placed inside furnace inside hollow cylinder of alumina. Oxygen is allowed to flow into the furnace and pure silicon is oxidized at 1100°C for one hour to get Silicon oxide at the top of Silicon. Thickness of the thermally grown oxide of silicon was verified to be 180 nanometer using Scanning Electron Microscopy (SEM), See Fig. 9 below.

Fig. 5: Two halves of Co$_2$FeAl target. The target on the right has been polished and thinned-down in order to fit into the 1” magnetron sputtering gun.

The standard process of cleaning and photolithography in the sample is followed as follows. Silicon oxide coated sample was cleaned with Acetone, Isopropanol and then Water to get rid of organic and inorganic contaminants present at the surface. It was then dried with nitrogen. The sample was then spun with Lift-Off photoresist (LOR) in two steps: at 500 rpm for 10 seconds followed by 6000 rpm for 40 seconds to get a uniform thin layer of LOR. The sample was baked at 150°C for 10 minutes to harden the LOR. The sample was then again spun with S1813 (positive
photoresist) in the same fashion as in LOR, and then baked at 105\(^0\)C for 1 minute. The sample with the bilayer of photoresist was then exposed to ultra-violet light for 18 seconds using a mask. The developing process uses CD26 (Tetramethylammonium hydroxide) for 40 seconds and then sample is rinsed with water and dried with Nitrogen. The photoresist which is not exposed to light stays on the sample while the exposed parts of the resist get washed out leaving the desired pattern to deposit metal on it. The sample was then transferred to a sputtering chamber and then pumped down to 5\(\times\)10\(^{-9}\) mbar pressure to get sputter deposited Co\(_2\)FeAl (CFA). The problem of adhesion in depositing metal on oxide layer was removed by depositing a Chromium (2.5nm) layer. The chromium makes a metallic bonding with CFA and its strong oxidizing property allows it to make bonding with the silicon oxide layer as well. A CFA film was then deposited without breaking the vacuum. The Sample is then immersed in acetone for 10 minutes with sonication to remove S1813 and 1165 remover to remove LOR. Then second step of photolithography and sputtering of Niobium follows in the same way except the exposure time with UV radiation is 30 Seconds. The final sample structure was Si substrate/ siliconoxide(180)/Chromium(2.5)/CFA(70)/Nb(61) multilayer is was fabricated, Thickness measured in nanometer. Due to the lack of time in this project we could not measure this sample but will be worth measuring in the future.

2.2 Samples for Tunneling Magneto-Resistance (TMR) Measurement

A plane silicon substrate was used to deposit a trilayer of CFA/AlO\(_2\)/CFA. The Si substrate was cleaned using acetone, IPA, DI water and followed by nitrogen drying. The sample was then hit with oxygen plasma for 5 minutes so as to introduce roughness on the surface of silicon which was found to be required for adhesion with photoresist layer. The LOR was spun in two steps: at 500rpm for 10 seconds and at 4000 rpm for 40 seconds, which is baked for 10 minutes at 180 \(^0\)C to make it dry. Next layer of photoresist (S1813) was applied on substrate and again spun in the same way as LOR and is baked for 1 minute at 105 \(^0\)C to harden it. The sample was then exposed for 8 seconds, developed in CD-26 for 40 seconds and then rinsed with DI water followed by nitrogen dry. The sample was loaded into the metal growth chamber until the vacuum became 5\(\times\)10\(^{-9}\) mbar. A 50nm CFA film was deposited using magnetron sputtering at 2.5\(\times\)10\(^{-3}\) mbar base pressure. The Argon gas used in sputtering was stopped and 1 nm of Aluminum was deposited using thermal evaporation. The oxide layer of Aluminum on the top of CFA was observed by flowing Oxygen at base pressure of 2\(\times\)10\(^{-2}\) mbar for an hour. The chamber was pumped down
again to get a base pressure of $5 \times 10^{-9}$ mbar and another layer of CFA was deposited with thickness of 25nm. Finally the sample was cleaned to remove the photoresist.

![Diagram of TMR multilayers](image)

Fig. 6: Cartoon of TMR multilayers (side view)

### 2.3 Measurement Techniques

During this thesis, the following methods were used to take the measurement.

#### 2.4 X-Ray Diffraction (XRD)

X-Rays are electromagnetic waves ranging in energy from 100eV to 100MeV. Thus the wavelength of x-ray ranges from 10nm to 1pm. When a high voltage (tens of kilovolts) is applied between two electrodes, high speed electrons will come out of cathode and are attracted towards the anode (Coper anode used in this study) to collide. These electrons lose energy and the loss in energy of de-accelerating electrons is radiated as x-rays.

X-ray diffraction is the phenomena in which x-rays of wavelength ($\lambda$) are diffracted by atoms on different planes of a crystal. The planes of the crystal contain a periodic array of atoms with inter-planar spacing of $d$. In this case a high intensity of diffracted x-ray is detected when satisfying the Bragg condition.

$$2d \sin \theta = n\lambda$$

Where $\theta$ is incident angle of x-rays on the plane of crystal and $n$ is the order of diffraction.

Inter-planar spacing $d$ can be expressed in terms of the Miller indices (hkl) and lattice constant (a) as
\[ d = \frac{a}{\sqrt{h^2 + k^2 + l^2}}. \]

Fig. 7: Diffraction spectra of X-rays by silicon crystal.

The sputtered-deposited CFA film on top of the natural silicon oxide just before second step of photolithography was characterized further using XRD analysis. The crystal of CFA was found to be crystalline in structure and with a lattice constant of 5.73 Å for (022) plane using Cu-Kά X-Ray [fig. 8]. This is in good agreement with literature\textsuperscript{16}.

Fig. 8: XRD pattern a) Si/SiO\textsubscript{2} substrate without any metal films. b) CFA/Si. c) CFA/SiO\textsubscript{2}/Si
2.5 Scanning Electron Microscopy

Scanning electron microscopy (SEM) was used extensively to produce and characterize the image of the sample. It produces and electron beam using Field effect which is then incident onto the sample after accelerating through a high potential (several tens of kilovolt). The primary and secondary beams of electrons were collected by the collector to produce the image of the sample. Since it uses the electron to scan the sample, SEM has a very fine resolution. The resolution of the image is given by: \[ d = \frac{\lambda}{2}, \]
where \( \lambda \) is the wavelength of the electrons used and \( d \) is the resolution of the image. The wavelength of an electron accelerated by a potential of 3 KV (used to produce fig. 9) is 0.224Å, using De Broglie equations, thus producing a nice and crisp image with high resolution. The thicknesses of the oxide film at the top of silicon wafer and the CFA film deposited on the top of it were found using SEM as shown in fig. 9. This method thus helped us to estimate the geometrical factor used in the magnetron sputtering process and in thermal deposition as well.

Fig. 9: SEM micrograph of CFA/SiO\textsubscript{2}/Si multilayer
2.6 Magnetization
The magnetization study was performed in multilayer of CFA and Perm-alloy at various temperatures ranging from room temperature to 5K. We were able to magnetize the sample with the field perpendicular and parallel to the plane of sample. This measurement involves the use of Physical Property Measurement System (PPMS) to get the hysteresis loop of multilayer. The independent switching hysteresis loop involves the hysteresis loop of two different layers switching separately which is important to study magnetoresistance in multilayer system.

2.7 Magnetoresistance
Magnetoresistance measurement was carried using PPMS system in three steps. The first step involves the TMR measurement in CFA/AlO₂/CFA(Py) multilayer system. The second step involves the GMR measurement in CFA/Al/Py multilayer and thirdly AMR measurement in CFA single layer. Also we were able to study the effect of the field applied perpendicular and parallel to the plane of sample.
Chapter 3
Theory

3.1 Two-Current Series-Resistance model

In the case of multilayers the electron spin orientation in these multilayers play crucial role in determining the resistance of the structure. Assuming the case of electron spins aligning parallel with each other within multilayers (say up). The spin up electron traveling across these multilayers will not suffer too much scattering. This is because there are electronic states available to the free spin up electrons to occupy and travel through. Whereas spin down electrons will not find enough states to occupy and hence they will be scattered more. The situation is illustrated by fig. 10.a. On the other hand, in the case of anti-parallel spin alignment within the multilayers both spin up and spin down electrons will get strongly scattered. Spin up electrons get scattered heavily whenever they encounter spin down layers and vice versa. This situation is well illustrated in fig. 10.b. The scattering of the electrons can be modeled as if there is large resistance (R) present and electron travelling with little scattering can be modeled as if there is a small resistance (r) present.

Fig. 10: Two-current series-resistor model for different spin orientations in multilayers. The electron with spin up is represented by an arrow with a dot and pointing upward whereas spin down electrons are represented by an arrow with a dot and pointing downward.
Whenever there is an increase in the resistance of a material with the application of an external magnetic field, we get magnetoresistance. This is the general case of magnetoresistance. Magnetoresistance occurs even in non-magnetic metals as a result of the Lorentz force. According to Lorentz force the electron in a magnetic field starts to move in a circular path so that the mean free path between two collisions gets reduced and hence the resistance increases as predicted by following the equation of resistivity.

$$\rho = \frac{m}{ne^2\tau} = \frac{\tau E}{nel}$$

Where E is electric field, ne is total charge concentration, l is mean free path, m is the mass of an electron, and \(\tau\) is the relaxation time.

The electric transport in ferromagnetic 3d metals is carried out by 4s electrons due to their reduced effective mass compared to that of 3d electrons. Higher density of states of 3d electrons is the cause for scattering of 4s electrons into 3d states resulting in high resistivity. As a result of exchange splitting of the density for 3d electrons the scattering probability of 3s electrons is higher when two thin films are antiparallel. On the other hand the probability will be a minimum when they are parallel.

### 3.2 Tunneling Magnetoresistance

The situation, where there are two electrodes separated by an insulating layer that is small enough to allow electrons to tunnel across it with the application of an electric current is called quantum tunneling. Quantum mechanics predicts the tunneling probability depends on the barrier thickness (L) as, \(e^{-2\sqrt{kL}}\). Where k is the electron wave vector in the barrier region and L is the thickness of the insulating layer. Now we choose two electrodes to be magnetic and the observed magnetoresistance is called tunneling magnetoresistance. As seen from the two-resistor model discussed above, the total resistivity of parallel alignment is less than that of anti-parallel alignment, thus TMR represents the band structure effect that relies on the spin resolved Density of States at the Fermi level.

The tunneling current between two electrodes plays an important role in tunneling magnetoresistance and is calculated from Fermi’s golden rule as:\(^{17}\);
\[ I = 2e \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} |M(E)|^2 D_1(E - eU)D(E) \left[ f(E - eU) - f(E) \right] dE \]

Where \( E \) is the energy of the electron with respect to the Fermi energy \( E_F \), \( U \) is applied bias voltage, \( M \) is the tunneling matrix, \( D_1 \) and \( D_2 \) are the density of states of first and second electrode respectively and \( f(E) \) is Fermi Dirac distribution function given by

\[ f(E) = \frac{1}{1 + \exp \left( \frac{E}{K_B T} \right)} \]

Where \( E \) is the energy with respect to Fermi energy \( E_F \), again, \( K_B \) is Boltzmann’s constant, \( T \) is absolute temperature.

Following the Julliere model\(^\text{12}\), the assumptions used in the tunneling process here are that the tunneling process conserves spin and that the tunneling current is proportional to the density of states of the corresponding spin orientation in the two electrodes across the insulating layer.

Therefore, the tunneling current for a parallel magnetization is,

\[ I^{\uparrow \uparrow} \propto D_1(E_F)^\dagger D_2(E_F)^\dagger + D_1(E_F)^\dagger D_2(E_F)^\dagger \]

Where \( D_1(E_F)^\dagger \), \( D_1(E_F)^\dagger \) are the densities of states of electrode 1 at fermi level \( E_F \) with spin up and spin down electrons respectively. Similarly \( D_2(E_F)^\dagger \), \( D_2(E_F)^\dagger \) are the densities of states of electrode 2 with spin up and spin down electrons respectively. For antiparallel alignment the tunneling current will be,

\[ I^{\uparrow \downarrow} \propto D_1(E_F)^\dagger D_2(E_F)^\dagger + D_1(E_F)^\dagger D_2(E_F)^\dagger \]

Defining the fraction of majority electrons and minority electrons for \( i \) th electrode as \( a_i \) and \((1 - a_i)\) as,

\[ a_i = \frac{D_i(E_F)^\dagger}{D_i(E_F)^\dagger + D_i(E_F)^\dagger} \]

And

\[ 1 - a_i = \frac{D_i(E_F)^\dagger}{D_i(E_F)^\dagger + D_i(E_F)^\dagger} \]

The spin polarization equation (1) can be written as

\[ P_l = \frac{D_l(E_F)^\dagger - D_l(E_F)^\dagger}{D_l(E_F)^\dagger + D_l(E_F)^\dagger} = 2a_l - 1 \]

Thus the differential conductance for parallel alignment can be expressed as
\[ G^{\uparrow\uparrow} = G_p \propto a_1a_2 + (1 - a_1)(1 - a_2) = \frac{1 + P_1P_2}{2} \]

And for antiparallel alignment as,

\[ G^{\uparrow\downarrow} = G_{aP} \propto a_1(1 - a_2) + (1 - a_1)a_2 = \frac{1 - P_1P_2}{2} \]

So, equation (3) for tunneling magnetoresistance can be written as,

\[ TMR = \frac{G^{\uparrow\uparrow} - G^{\uparrow\downarrow}}{G^{\uparrow\downarrow}} = \frac{G_p - G_{aP}}{G_{aP}} = \frac{R_{aP} - R_p}{R_p} = \frac{2P_1P_2}{1 - P_1P_2} \]

3.3 Anisotropic magnetoresistance (AMR)

Anisotropic magnetoresistance is the current induced effect in which resistance of a ferromagnetic material changes in an external magnetic field. This effect is caused by spin-orbit interaction on the 3d orbitals as a result of the external magnetic field. For a finite value of spin orbit interaction there occurs a spin flip scattering. This is to say that majority s-state electrons scatter into minority d states, which is the reason for the increased resistance. For parallel and perpendicular orientation between the current and magnetization directions, the scattering cross section of s electrons is different due to the orbital anisotropy of the empty d states. Higher scattering of electrons occurs when the current and magnetization directions are parallel, so we get high resistance. Similarly there will be lower scattering of electrons when the current and magnetization directions are perpendicular to each other. This leads to the change in resistance of the sample in accordance with the angle between electric current and magnetization direction. Thus the angular dependence of AMR is written as,

\[ R(\theta) = R_\perp + (R_\perp - R_\parallel)\cos^2(\theta_{IM}) \]

Where, \( R_\perp \) is the resistance when the current and magnetization are perpendicular, \( R_\parallel \) is the resistance when current and magnetization are parallel to each other and \( \theta_{IM} \) is angle between current and magnetization.

3.4 Giant magnetoresistance (GMR)

GMR effect is applicable to multilayer systems in which two ferromagnetic layers are separated by an antiferromagnetic or non-magnetic spacer. We used our ferromagnetic layers to be
Co$_2$FeAl and perm-alloy and the non-magnetic layer to be aluminum. The origin of GMR is the interlayer is spin flip scattering between two ferromagnetic layers. Similar to TMR, the resistance of the interlayer is high if two spins in ferromagnetic layers are antiparallel to each other and low if they are parallel to each other. The GMR is thus written as

$$GMR = \frac{\rho_{ap} - \rho_p}{\rho_p}$$

Where, $\rho_{ap}$ and $\rho_p$ are resistivity of sample in antiparallel and parallel alignment of two ferromagnetic layers.
Chapter 4

Data and Analysis

4.1 Circular transfer line method

We used circular transfer line method\(^{18}\) (CTLM) to measure our sample. CTLM samples have the geometry of concentric circular rings. The inner circular ring of multilayer is separated by a gap of varying width 80, 40, 20, 15, 10, 8, 6, 4, 2 micrometers respectively. The inner circular rings of 600 and 400 micrometer diameters were used. We apply positive current across the inner rings and take a negative current out from the surface of the substrate using PPMS. Similarly the voltage is measured across the internal ring and outer rings in a four-probe configuration. The circular rings used in the measurement are shown in fig. 11.

Fig. 11: Image of the sample used for CTLM showing inner circles (600, 400, 300 μm from bottom to top respectively) separated by a gap (or break in the multilayer) with varying width (80, 40, 20, 15 μm from right to left respectively).
4.2 Van Der Pauw measurement

We used the well-known four probe Van Der Pauw\textsuperscript{19} method to measure the sheet resistivity of our sample. The sample used was CFA grown on silicon oxide-coated Si wafer, as shown in fig. 12. The accuracy of this method is dependent on several factors, such as sample being flat, much thinner as compared to its length and width, sample is continuous i.e. there are no holes in sample, and the electrical contacts are made at the corners of the sample. The geometry of the sample used is as shown in fig. 12 below.

![Van der Pauw geometry for resistivity measurement.](image)

The four corners of sample are marked 1 through 4 in counterclockwise direction. Standard DC four probe technique uses positive DC current from corner 1 and negative current from 2 ($I_{12}$). Similarly, positive DC voltage from corner 3 and negative DC voltage from corner 4 ($V_{34}$). So that the resistance of the sample is given by, $R_1 = \frac{V_{34}}{I_{12}}$.

Similarly DC current supplied across corners 2 and 3 gives the $I_{23}$. Also the DC voltage measured across corners 4 and 1 gives $V_{41}$, for which the resistance of the sample is given by, $R_2 = \frac{V_{41}}{I_{23}}$. Now the resistance of sample is given by $R = (R_1 + R_2)/2$.

Van der Pauw\textsuperscript{19} showed that (apart from a geometrical factor) the sheet resistance of the sample is given by:
Thus the resistivity of the sample was found to be 13μΩcm. Fig. 13 shows the metallic behavior of CFA thin film deposited on SiO₂. An extra layer (2.5nm) of Chromium was deposited in this geometry so that chromium being highly reactive with oxide it makes covalent bonding with Silicon oxide layer and makes metallic bonding with CFA film on its top. In other words, chromium was used to make an adhesive layer for CFA with oxide.

![Graph showing Voltage (V) Vs Temperature (K) relationship in CFA surface, measured using two probe.](image)

**4.3 Resistance measurement**

It is important to ensure that we have a good oxide layer in TMR structure. If there is direct contact between the two electrodes across the tunnel barrier then one should expect to get metallic behavior in resistance with temperature. On the other hand, in the absence of such ‘pinholes’, one should expect to get resistance as in semiconductor or oxide. The resistance of normal metals decreases with decreasing the temperature because at low temperature there will be less scattering of electrons with an increased mean free path whereas at high temperature there will be higher scattering of electrons as a result of phonon scattering and hence increased resistance. But the case of oxide layer sandwiched between two metal electrodes, will be
different than that of normal metals: It is purely determined by quantum mechanical tunneling phenomena. In this case resistance of the oxide rises with reducing the temperature. We used this fact to verify that we have good oxide layers inserted between the two ferromagnetic metal electrodes in the TMR structures. The temperature variation of the resistance of some of the devices is shown in fig. 14 below.

Fig. 14: a) Resistance of multilayer CFA(25)/Al(1.2)O₂/Py(20) on silicon substrate after annealing for 1 hour at 200°C and b) Resistance of multilayer CFA(30)/Al(0.6)O₂/Al(0.6)O₂/Py(25) in
Buffer oxide etched silicon substrate. The labeling numbers are for diameter of circular feature and gap in micrometer.

The resistance of the samples grown without removing the native oxide on the silicon substrate were observed to be higher, which is due to the oxide of silicon present in the substrate. The buffered oxide etch (diluted hydrofluoric acid, HF) cleans that oxide and gives clean silicon interface. This method of buffer oxide etch cleaning is explained in next section. Since there are both the Si native oxide and portions of the Si semiconductor present in our sample so it is hard to tell which one contributes more to the behavior of the resistance, but for sure it is not due to contribution from metal.

4.4 Magnetization measurement

Depending on the nature of a magnetic material the magnetic moment of the sample varies. The Magnetization is the total magnetic moment per-unit volume and is dependent on the magnetic material. The magnetization loop, sometimes called hysteresis loop, is the measure of the total power loss in one cycle of magnetization and demagnetization of the material. With the application of a magnetic field, the different magnetic domains inside the material start to respond with the field. The magnetic domain which has its moment initially aligned in the applied field direction starts to increase in size. This process continues until all moments in all domains are aligned in the applied field direction. This explains the increasing magnetization in the first quadrant. Once all the domains are aligned in the same direction, the magnetization obtained is called saturation magnetization and it is not going to increase much more with the increase in field. From this point, the magnetization starts to decrease with decreasing field and does not follow the same path with decreasing field. The magnetization remaining in the sample at zero field is called remnant magnetization (M_r). At this point the magnetic moments in individual domains start to orient randomly and hence the magnetization decreases. The field required to make the magnetization value zero in a sample is called the coercive field (H_c). This explains the second quadrant magnetization loop. The magnetization then starts to increase in opposite direction which is the case that the magnetic moment starts to align in the field direction, which is in the negative direction now. Once all magnetic moments become parallel to the field direction we get saturation.
magnetization but in the negative direction in accordance with the field direction in the third quadrant. After reaching the magnetization saturation point the same phenomena happen as in the first quadrant which is followed by remnant magnetization in negative direction and then coercive field in positive direction. This completes a loop called the hysteresis loop.

![Magnetization loop of Perm-alloy (9nm) on Silicon wafer.](image)

Fig. 15. Magnetization loop of Perm-alloy (9nm) on Silicon wafer.

The shift in the center of the magnetization curve from origin is due to exchange bias\textsuperscript{20,21}.

4.5 Independent switching effect in multilayer

Magnetoresistance study in multilayers is more importantly dependent on the magnetization of two or more layers. If a multilayer is coupled together then the magnetization in different layers will no longer be independent. In such case the magnetization will be dominated by the soft magnetic material. The resistance of the sample is dependent on achieving the parallel and antiparallel alignment of the two layers as seen from the two-resistor model. So it is important to get independent switching of the two layers before expecting magnetoresistance. Magnetization in CFA/AIO/CFA was measured at different temperatures. The purpose of this study is to find the switching field at which the two layers of CFA switches its magnetization direction. The following graphs are for the magnetization of the multilayer at different temperatures. The two layers of CFA
have different thickness, bottom layer has 50 nm and the top one is 25 nm. The switching of the moment in thin films is dependent on their thickness\textsuperscript{22}. So we expect those two layers to switch their moment independently i.e. at different applied field. But the observed hysteresis loop reveals the fact that they are no longer switching independently but are together. The loop below was taken at different temperatures of 5K, 50K, 100K, and 300K respectively.
Fig. 16: Magnetization measurement in CFA(50)/Al(1)O$_2$/CFA(25) multilayer at temperatures 5, 50, 100, 300 Kelvin respectively.

As seen from these loops, the two layers of CFA are switching their field together. This situation may have resulted from coupling of the two CFA layers at some point. This is to say that there may be some channel present in multilayer which connects the bottom to the top CFA, or that the switching of the two layers does not vary much with the different thickness of the sample in CFA. It is just a speculation.
Fig. 17: Variation of Resistance of the sample with magnetic field at room temperature. The sample with multilayer structure CFA(50)/Al(1.4)O₂/CFA(25). The numbers in bracket are thicknesses in nanometer.

The sample shows no TMR as shown in fig. 17. This might be because there was already high resistance coming from of the Si used as a substrate (few Kilo Ohms) so that the small variation in the resistance of the multilayer with change in field was negligible and was not showing any TMR effect. To resolve this problem, we changed our method to prepare the sample. We did thermal deposition to deposit 104 nm of copper on the Silicon sample before photolithography and then followed by CFA/AlO₂/Py multilayer after photolithography. We chose the top layer in the TMR structure to be perm-alloy so that CFA and perm-alloy switches their magnetizations at different fields. Furthermore it is a soft magnetic material so that it will not take high magnetic field to be magnetized completely. The oxidation of the sandwiched aluminum layer to make insulating barrier was done as described²³. The thickness of oxide layer is controlled as shown in fig. 18.

Fig. 18: Aluminum oxide thickness at different pressure and different oxidation time²³. Oxide thickness in our sample is at the point shown by green arrow at 0.225 torr pressure of 99.999% pure oxygen and flow time was 40 second.
The resistance of the copper thus deposited was measured to be few milli-Ohm. The morphology of the sample was studied with the Electron Dispersive X-ray (EDX) analysis. SEM was used to produce the image of the sample and was analyzed with EDX. The results of EDX show that our sample does not have other impurities. The only observed peaks in EDX spectrum were for Silicon, Copper, Nickel, Cobalt, Iron, Aluminum that were used to produce the sample. Fig. 19 shows that there is 76.7% of Silicon as expected because we used Si as the substrate. Copper in our sample was 16.6%, which also makes sense because there was 104 nm of Copper deposited on the sample. Thirdly Nickel, because there was 80% of nickel in perm-alloy which carries 20 nm of thickness in our multilayer followed by cobalt, aluminum and Iron. Aluminum concentration is slightly higher than iron because in addition to CFA, where iron and aluminum was in equal amount, there was insulation barrier of aluminum oxide as a heart of TMR structure.
To solve the problem of the CFA in both electrodes switches its magnetization at the same field, we prepared two types of samples of trilayers of CFA/AI$_2$O$_3$/Py as explained above. One sample without lithography which is used to check independent switching of the two magnetic layers while another was featured with circle of different diameter and with varying gap. The magnetization observed in the un-patterned sample is as shown in fig. 20. This structure clearly shows that the two magnetic layers separated by aluminum oxide are switching their magnetization independently.
Fig. 20: Magnetization observed in CFA/AlO$_2$/Py Structure a) Measured at room temperature (298K) and b) at cold temperature (10K). Red and black arrows indicate the spin orientation of the two metals at different fields.

This graph is actually a juxtaposition of hysteresis loop for CFA and Perm-alloy. Those two materials switches their magnetization at different fields. The observed plateau between remnant magnetization and coercive field is the transition region between the fields required to switch magnetization in either of the layers. Higher field at low temperature is due to the fact that electrons will have less energy at low temperature so higher torque has to be applied to align
spins in field direction than at room temperature. This graph explains the fact that Perm-alloy switches its magnetization at low field first and then after the field is increased the CFA switches its magnetization, leading to a plateau in the region between remnant magnetization and coercive field.

4.6 Magnetoresistance measurements

TMR sample prepared as explained in previous section was used for magnetoresistance measurements with different features. Circular features having 600 and 400 µm diameter and varying gap width were used. No TMR effect was observed in these features either. It is noteworthy to mention that this sample showed some interesting result coming from the 400 µm diameter circular channel with 10 µm gap in it. It is interesting in the sense that it has large percentage (13.58%) of anisotropic magnetoresistance (AMR). This effect is shown in fig. 21. The reason for such large AMR could be resulting from the pinholes through the oxide layer.
Fig. 21: Large (13.58%) of AMR obtained in TMR structure. Sample measured had structure of CFA(25)/Al(1.4)O$_2$/Py(30) with 400 µm diameter feature and 6 micron gap on it. a) TMR in terms of resistance b) TMR in terms of percentage.

To further reduce the effect of the contact of the Heusler alloy with the silicon substrate, the native silicon oxide present at the surface of Si was cleaned by immersing the substrate diluted hydrofluoric acid (HF) etch. We did photolithography as described earlier using LOR and S1813. The sample was further etched with oxygen plasma followed by etching in 2 percent of HF acid in deionized water for 10 seconds while attaching the sample in sample holder. Finally sample is rinsed in water to stop further etching, followed by nitrogen gas drying. This sample was then loaded into the sputtering chamber, which was pumped down immediately. In this cleaning the parts of Si substrate that are not covered with photoresist are exposed directly to HF and hence the oxide present on such parts gets etched away where we want to deposit our TMR multilayer. But unfortunately the sample did not show any measurable TMR effect as shown in fig. 22 below.
Fig. 22: TMR measurement across tri-layer electrodes in BOF etched silicon substrate. 400x15 stands for 400 micron channel and a gap of 15 micron.

A new set of samples was then made using the silicon oxide substrate as shown in fig. 9. We were able to fabricate three samples, multilayer of CFA(30)/Al(10)/Py(40), bilayer of CFA(30)/Al(15), and bilayer of Al(15)/Py(40). We used the first multilayer for GMR studies and the latter two for AMR measurements.

The multilayer was first checked to find out whether it has a metallic temperature dependence or not. The variation of resistance with temperature was measured by supplying current across a long 100 µm channel and taking voltage across any two points between the two current terminals. Resistance variation with temperature is shown in fig. 23. While we did not observe a straight line, the resistance decreased monotonically with decreasing the temperature.
Fig. 23: Resistance variation in GMR sample showing metallic behavior.

Results of the GMR measurements on this sample are included in fig. 24. We applied magnetic field perpendicular to the plane of sample and current along the plane of the sample. Magnetic moments in multilayers have to rotate in a direction perpendicular to the plane of sample which takes more anisotropic energy to rotate. For that reason we applied large fields of 2kOe and 3kOe respectively. The resistance follows the same pattern in both fields. We measured 2 samples each of them in both field values. All of the measurements show the same pattern. The resistance of the sample first decreases with increasing field. After a certain value of field, the resistance stays
almost the same. But following the negative loop, the resistance does not follow the same path and does not increase back to the initial value. Rather it shows a small hysteretic field dependence that is consistent with Py anisotropic magnetoresistance.\textsuperscript{24}.

Fig. 24: Large magnetoresistance obtained in CFA/Al/Py (30/10/40) multilayer, Thickness in
nanometer inside brackets. a) 36.5% MR with 1.6% AMR at 3kOe. b) 36.8% MR with 2.39% AMR at 3kOe. Right graph in each of case is enlargement of the AMR due to Py.

We also checked the magnetoresistance of the same sample with the field in a direction parallel to the field. The observed MR curves are as shown in fig. 25.

Fig. 25: Magnetoresistance measurement in CFA(30)/Al(10)/Py(40) at 10K with field parallel to the plane. The two identical channels were used but the length of two channels were different in fig. a) and b).

The minimization of total energy within each domain needs the magnetic moment to be aligned in direction parallel to the plane of sample. This is to say that each domain minimizes its energy by making antiferromagnetic coupling within it. So whenever we want to magnetize the layer perpendicular to its plane it requires more energy to do it. As seen from fig. 24 that we have to apply high field to get all moments to align in field direction and hence get parallel orientation of moments within the layers. In contrast to fig. 24, we get both of the ferromagnetic layers magnetized in field direction at low field when field is in direction parallel to the plane as in fig. 25. Similarly whenever the field is applied in a direction parallel to the plane the spin structure
which explains the minimization of total energy by antiferromagnetic coupling between any two consecutive domains in vertical direction, is not present. This leads to small MR value (2.1% and 1.8% in fig. 25). The highest resistance in the sample comes from the state which favors the antiferromagnetic coupling between consecutive vertical domains and gives 36 percentage of MR as shown fig. 24. Where as in parallel field case, due to lack of such coupling effect, the highest resistance comes when all the moments are aligned parallel to each other within single layer and aligned antiparallel within different layers which is the source of secondary high resistance in field perpendicular to the plane case.

The fact that we need to apply high field to completely magnetize the sample in direction perpendicular to the plane direction is supported by fig. 26. We used the same sample as we used to produce fig. 20 where we get saturation moment around 50 Oe. But in parallel case the saturation magnetization is not observed even at 5kOe.

Fig. 26. Magnetization in CFA(30)/Al(15)O\textsubscript{2}/Py(25) with field perpendicular to the field. Inset is zoomed in portion of center of loop.
The same sample was then used for AMR measurement with varying the angle between current and magnetization direction. The observed result is as shown in fig. 27 below. The resistance of the sample is maximum when the current and magnetization are parallel to each other and is minimum when the current and magnetization are perpendicular to each other. This gives us AMR of 2.2% and 1.9%.

![Graph](image)

**Fig. 27.** AMR measurement in two samples featuring CFA(30)/Al(10)/Py(40) at 10K. Sample is rotated from 0° to 360° which changes the magnetization angle keeping current angle constant. The only difference between fig. a) and b) is that we measured resistance across different distance.

We further measured the second sample which has CFA and Aluminum only on it. This is an interesting result in the sense that curve that we get with bilayer is showing the same pattern as in tri-layer at the beginning which gives rise to 10.2%, 19% and 18.89% of magnetoresistance. The occurrence of less MR percentage in one of the samples is attributed to the fact that we were missing resistance data values up to the field value of 200Oe. This work is similar to the result in reference25.
Fig. 28: Magnetoresistance obtained in CFA(30)/Al(15). a) Magnetoresistance of 10.2% b) magnetoresistance of 19% c) magnetoresistance of 18.9%.
Chapter 5

Conclusion and Future works

Co$_2$FeAl and Perm-alloy were prepared using arc melting. Thin layers of CFA were then deposited using magnetron sputtering whereas Py was deposited using physical/thermal deposition. Both the magnetic and transport properties of these thin-film samples were studied with the PPMS. We checked for the TMR effect using various techniques to make the trilayer. A large value of AMR (13.58%) was observed at 5K in TMR structure. Strong coupling between the two electrodes in TMR sample was dominant. Annealing of the samples that showed a high value of AMR in ambient environment at 200C for 1 hour showed no effect. Interestingly an in-plane MR of 36.8% was observed in CFA/Al/Py multilayers. We have also developed a technique to etch the Bismuth Selenide (Bi$_2$Se$_3$) (see appendix B). Bi$_2$Se$_3$ sample was grown by Dr. Jacek Furgyna group at University of Notre Dame. We etched Bi$_2$Se$_3$ with this technique. This sample can be further used to study spin polarization using Andreev reflection or using TMR. Further we have made CFA/Nb contact as shown in fig. 29, which can be further analyzed for Andreev reflection spectroscopy.
Fig. 29: Top view of CFA/Nb contact for AR study.
Appendix A

1. Samples prepared for project

<table>
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<th>SN</th>
<th>Symbol</th>
<th>Structure</th>
<th>Thickness (nm)</th>
<th>Date Sample was prepared (mm/dd/yy)</th>
<th>Fig. Produced</th>
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<td>1</td>
<td>S2</td>
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<td>(25/1.2/20)</td>
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<td>05/27/2015</td>
<td>15</td>
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<td>05/04/15</td>
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<td>CFA/Al</td>
<td>(30/15)</td>
<td>07/10/15</td>
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CFA = Co$_2$FeAl  
Al = Aluminum  
Cu = Copper  
Py = Perm-alloy
Appendix B

**Bi₂Se₃ etching process to make hall bar**

Following steps were used to make Hall bar by etching Bi₂Se₃ sample

1. Clean sample with Acetone for 2 minutes followed by cleaning in Isopropyl alcohol for 2 minutes and finally dry with nitrogen
2. Spin and bake LOR for 10 minute at 150 °C
3. Spin and bake S1813 for 1 minute at 105 °C
4. Expose for 7 seconds.
5. Develop in CD 26 for 40 Seconds, clean with De-Ionized (DI) water and dry with nitrogen
6. Oxygen plasma etch for 2 minutes
7. Make etchant as follows
   a. Take 40 ml of DI water in a beaker
   b. Add 40 ml of Acetic acid
   c. Add 20 ml of Nitric acid
   d. Add 5 ml of Hydrogen peroxide
   e. Add next 40 ml of DI water and stir well
8. Dip the sample made in step 6 in etchant made in step 7 for 150 seconds.
9. Remove the sample from acid in step 8 and rinse in DI water to stop reaction and dry sample with nitrogen.
10. Remove photoresist.

Using this method we were able to etch 27 nm of Bi₂Se₃.
Chapter 6
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