ABSTRACT:

MAGNETIC DAMPING IN Fe$_3$O$_4$ THROUGH THE VERWEY TRANSITION FOR VARIABLE AG THICKNESSES

by Daniel Stanley

The temperature dependence of damping in epitaxial Fe$_3$O$_4$/Ag bilayers grown on [001] MgO substrates was measured by Ag thickness. The Fe$_3$O$_4$ was deposited to 350 nm while the Ag thickness ranged from 0 to 500 nm. Measurements using ferromagnetic resonance (FMR) at 9.5 GHz with the sample film normal to the applied magnetic field were taken over a temperature range of 80 to 295 K. In-plane FMR was also done at 35GHz to determine the anisotropy. From these measurements the effective magnetization was calculated and with it the anisotropy parameters K$_4$ and K$_u$. The samples exhibited easy-plane anisotropy consistent with thin-film effects. Line widths did not change substantially until the Verway transition ($T_V \sim$ 110 K), after which the damping increases dramatically at lower temperatures. This phenomenon will be discussed in terms of spin pumping into the Ag in conjunction with the properties of Fe$_3$O$_4$ changing at the Verway transition.
MAGNETIC DAMPING IN FE$_3$O$_4$ THROUGH THE VERWEY TRANSITION FOR VARIABLE AG THICKNESSES

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1 Introduction:

The earliest discovered sources of magnetic phenomenon were found in naturally occurring mineral deposits [1]. In 6th century BC, the Greek philosopher Thales of Miletus discovered an attraction between certain materials. This naturally occurring magnetic material was called a lodestone. These lodestones were composed of magnetite or Fe₃O₄. From these early discoveries came the observation of the Earth’s magnetic field as well as effect of magnetite on other materials, such as the finding by early natural philosophers that rubbing magnetite against them could transfer some of this property to certain metals. This eventually led to the use of refined Fe ore for compasses which were made in the 11th century. Without the discovery and subsequent application of this simple magnetic device exploration of the planet and the mercantile trade that came from this would have been severely curtailed, as the implementation of compasses preceded and effectively allowed the great naval exploration if the coming centuries. Remarkably, the first known magnetic material still has properties being discovered even to this day which help to further our understanding of the very basics of magnetic phenomena, as well as lead to new applications for research and commercial interest.

Besides their continued use in compasses and navigational systems magnetic materials also form the basis by which modern computers retain data, and have served a similar function for nearly half a century with the wide use of magnetic tape. However, the continued need for memory storage that is fast, large capacity, and nonvolatile leads new research and development of magnetic memory storage. All current hard drives owe their speed and the density of information that can be stored to the discovery of the giant magnetoresistance effect (GMR), and in this case the application to construct superior magnetic field sensors [2]. As the previous hard drive sensor geometry eventually proved insufficient for the growing demands of storage density, it was changed from current in-plane (CIP) to current-perpendicular-to-the-plane (CPP) [3]. These developments represent the continued applications that come about from investigating new methods to manipulate electrons by way of their intrinsic spin instead of using the charge, called spintronics [4]. This provides an additional degree of freedom for logic operations and memory storage [5]. GMR is an example of spintronic phenomena, as it requires different
resistances dependent on the spins of the electrons in order to function. The spin-dependent resistance is caused by the relative orientations of the ferromagnetic layers, where parallel produces the smallest amount of scattering and the least resistance, while anti-parallel alignments produce the most scattering and the greatest resistance [6].

These spin-polarized currents are generated by ferromagnetic materials as the alignment of band structure inside of them produces additional resistance for electrons of one spin than the other [5]. Since the magnetic domains of the ferromagnet produce a spin polarized current, altering those same domains with an applied magnetic field allows for the relative resistance applied to the differing spins to be controlled [8]. These alignments can be altered by an external magnetic field thus producing spin polarized currents as required [7]. This in turn can be used to control the absolute strength of the generated current as well as the relative proportion of spins that make it up. Additional layers of nonmagnetic conductors and ferromagnets in a multilayer can then be used to further control the magnitude of the currents and relative spin polarization, such as in the example of GMR [8,9]. In these more complicated cases, the additional ferromagnetic layers can have their magnetic domains altered to further tune the resulting current passing through the device to the specifications desired. Still, to generate a spin polarized current a material needs to have a net magnetization like a ferromagnet or ferrimagnet. Magnetite provides such a state and serves as a natural source of spin-polarized currents, as its conduction electrons are nearly a hundred percent polarized.
Figure 1: Crystalline Structure of Fe$_3$O$_4$, where large dots are O, small dark ones tetrahedral Fe positions, and the small grey ones are the octahedral Fe atoms. The oxygen atoms form a distorted face-centered-cubic structure [10,11].

Magnetite or Fe$_3$O$_4$ itself is not actually a ferromagnetic material though [10]. It is instead a very closely related type called ferrimagnetic. This type of magnetic material does not have all of its magnetic moments aligned like a ferromagnet. Instead they exist as interacting sublattices within the structure of the magnetite sample, which will have aligned themselves anti-parallel to each other with one sublattice dominating in magnetization over the other [10,14]. The interesting structure of magnetite, shown in figure 1 is the source of this property. While the predominant Fe atoms orient in such a way as to produce a ferromagnetic orientation, a sublattice with a smaller moment within the lattice will be oriented to oppose that majority. Figure 2 below shows a closer look at one octant of the magnetite lattice [10]. This complex structure prevented its proper identification until the mid twentieth century, and ferrimagnetism itself was first discovered in attempts to explain the unusual properties witnessed in magnetite [14]. This ferrimagnetic material also possesses a high curie temperature of 860K, which further increases its interest as a material for investigation in spintronics devices, as this means that it could have high operational temperatures. The material itself takes the form of
FeOFe$_2$O$_3$ in a cubic structure. Nonmagnetic, diamagnetic and paramagnetic materials may be readily applied to Fe$_3$O$_4$ in order to construct spin-based devices [12]. Finally and of significant importance is the transition magnetite undergoes at ~120K [14].

Figure 2: The Fe$^{+2}$ and Fe$^{+3}$ ions in the octahedral positions from the {100} and {110} as well as a diagram of the one eight of the spinal structure of octahedral Fe atoms [12].
Verwey Transition

The Verwey transition is a metal conductor to insulator transition at 120K, as can be readily observed in Figure 3. The sharp change in conductivity is typical of good quality magnetite samples, as they will readily undergo fast and significant changes in their measured properties at the Verwey temperature. The exact temperature at which this transition occurred proved to be very difficult to pin down exactly, because of how even small impurities or imperfections in the structure of magnetite caused this transition temperature to be much lower [13,14]. The Verwey transition itself is caused by the crystalline structure of Fe$_3$O$_4$ going through a geometric shift, changing from cubic to monoclinic, with the accompany change in electrical conductivity, as well as the magnetic properties of the sample [14]. In Figure 3 this transition is shown, though not the change in the angular change of $.2^\circ$ [15]. While this transition does place a lower limit on the use of Fe3O4 as a source of spin polarized currents for low temperature systems, it also provides a useful method of discerning the stoichiometry of Fe$_3$O$_4$. This is especially important in the case of thin films as obtaining the correct concentration of oxygen atoms in the material can even be more difficult while growing the films can be very difficult. In cases of even minor errors in film growth the temperature at which the Verwey transition occurs can be substantially lowered [13]. This dependence on good stoichiometry for the Verwey transition can be considered to be the effect that that the geometric transition that the crystal goes through at the critical temperature. In instances of too much or too little O, the lattice will not be able to uniformly shift into the monoclinic structure. When this transition occurs it results in an insulator, so measuring
the resistance v. temperature will clearly show the Verwey transition if it is present in a sample. Figure 4 shows the resistance dependence on temperature for our samples, and it is clearly a better quality magnetite sample. This result comes about from the better stoichiometry in our samples, as the concentration and position of the oxygen atoms is correct for high quality magnetite. The use of temperature dependence in identifying the quality of magnetite samples already exists as a diagnostic technique [1], but in our case we will be focusing entirely on the changing magnetic properties about the Verwey temperature.

This transition is of particular interest given the effect that it has on the magnetic damping of the magnetite samples. In the context of a ferromagnetic material, damping is the effect of the decay of spin excitation. About the orientation of the ferromagnet’s magnetization equilibrium orientation this excitation will precess, and the damping is then the measure of the rate at which this precessions amplitude will decay [16]. This was first determined by observing the scattering rates for spin-up and spin-down electrons in the itinerant electrons of transition metal ferromagnets [17]. The proportion of spins that will all align also depends on the material, which as previously stated, in magnetite is nearly 100%. If magnetite is in contact with a nonmagnetic conductor the precessing magnetization induces a spin polarization in the nonmagnetic conductor [18]. The samples studied in this instance used Ag, though Au and Pt are also common elements used in multilayers.
Ag was chosen because it has a very long spin coherence length, meaning that spin polarized currents from the source magnetic material will travel further through this material before they lose their polarization [19,20]. The spin diffusion length for Ag has been found to be as great as 180nm [19]. Past this length the spins of the component electrons will no longer be dominated by the net polarization of the source.

This in turn can be considered as a decrease of the relaxation time or increase in the damping rate of the magnetic moments in the magnetite itself. This is caused by the spin polarized currents being produced in the magnetic material serving to transfer a small amount of angular momentum from the donor material into the nonmagnetic conductor or semiconductor material. The transfer of momentum can be considered to occur because of the spin currents created in the magnetic material from the application of an external magnetic field, it causes the magnetic moments to precess. In a sense they spin like tiny tops. With a nonmagnetic sink material adhered to the outside surface, these generated currents may leave the source material, in turn transferring energy out of the precession of the magnetic moment. In the case of these magnetite samples, amount of magnetic damping was studied both as a function of temperature and Ag layer thickness. In addition to the magnetic properties of magnetite, I have made significant contributions to several other experiments on magnetic materials while working in Dr. Pechan’s lab. The results of these studies are summarized in the appendix.
2 Background Theory:

Before speaking on theoretical background of ferromagnetic resonance imaging, it is important to first define ferromagnetic materials in more detail than previously approached. A ferromagnetic material is one in which the magnetic moments have aligned in one direction and will remain locked in that orientation to produce a net magnetization of the material. This distinction separates it from anti-ferromagnets where there are still distinct magnetic domains, yet they are aligned in such as way as to produce no net magnetization and paramagnet where there is no domain structure at all as magnetic moments remain random and disordered in the absence of an external field. Within a ferromagnetic material there may exist more than one domain, and this is often the result in bulk material. However, in FMR the magnetic material will be saturated by an external field, so that all domains orient in the same direction.

2.1 Anisotropy:

Anisotropy refers to the effect that orientation of a material can have on the results of a measurement [21]. This is caused by the material existing in a sublattice structure in which the net magnetization energy is not spherically symmetric producing an angular dependence. Some directions will naturally allow for a weaker external magnetic field to align them than in other orientations. As such rotating the magnetic moments requires greater field strength in one direction than in another. This essentially defines the difference between the easy axes as the orientation that requires the minimum field strength and the hard axes as the direction that requires the maximum field strength to achieve the same affect. Figure 6 shows the orientation at which this occurs, as well as the angle $\phi_H$ is the deflection from the easy axis. The difference between the magnetization observed along these directions is shown in Figure 7 [22]. The magnetization of out-of-plane was also measured for these films.
Still, the actual source of anisotropy can be more clearly defined. The magnetic moments interact in two specific methods to achieve this effect. The first is dipole-to-dipole interaction which is minor contributor in Fe based ferromagnets as it is negligible in a cubic crystal [23]. Instead, the primary source of anisotropy will be spin orbit coupling. This couples the spin to the charge densities within the crystal lattice. As this ties the anisotropy to the relative positions of crystalline structure, the anisotropy energy $E$ can be defined geometric position of the atoms in that lattice. Where $\alpha_i$ is the series expansion of direction cosines.
and k is the anisotropy constants, the saturation magnetization for Fe relative to axes of the crystal lattice is [21, 2424]:

\[ E = k_1(a_1^2a_2^2 + a_2^2a_3^2 + a_3^2a_4^2) + k_2(a_1^2a_2^2a_3^2) \]  

(1)

While this expansion may be carried to higher terms, it does not apply to any known ferromagnet. The effect of anisotropy on the resulting FMR spectra cannot be modeled using only equation 1. Of particular importance is the shape of the samples themselves. Thin films have radically different properties when studied with FMR then in bulk or other configurations. This effect is called shape anisotropy. The demagnetizing energy for a thin film in-plane is:

\[ E_D = \frac{1}{2}MH_D = 0 \text{ where } H_D = 0 \]  

(2)

While the same quantity for out-of-plane orientation is [25,26]:

\[ E_D = \frac{1}{2}MH_D = 2\pi M^2 \text{ where } H_D = 4\pi M \]  

(3)

The total energy for a ferromagnet is given in equation 5 below:

\[ E = -MH[\sin \theta \cos(\varphi - \varphi_H)] - (2\pi M^2 + K_u) \sin^2 \theta + K_4 \sin 2\varphi^2 \sin \theta^2 \]  

(4)

Angles \( \varphi \) and \( \varphi_H \) measure the distance that the external magnetic field \( H \) and the direction of the material’s magnetization make to the easy axis. The remaining angle \( \theta \) is for the out-of-plane anisotropy. The constants \( K_u \) and \( K_4 \) are additional anisotropy terms, with the first for out-of-plane and the second four-fold in-plane anisotropy. The Landau-Lifshitz equation of motion can model the anisotropy of four-fold materials like magnetite [25,27]:

\[ \frac{dM}{dt} = \gamma M \times H_E + \lambda M \times (M \times H_E) \]  

(5)

Figure 8: Sample orientation for in-plane measurements.
In equation 5, $\gamma$ is the gyromagnetic ratio and $\lambda$ is a constant characterizing the damping of the material. It is determined using the Bohr magneton $\mu_B$, the reduced Planck constant $\hbar/2\pi$, and g-factor which is measured for magnetite to be 2.12 from literature [28]. $H_E$ is the effective $H$ as the energy gradient to the magnetization. The resonance equation for the in-plane anisotropy is derived from this equation and the following expression:

$$\left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{(M \sin \varphi_H)^2} \left[ \frac{d^2 E}{d\theta^2} \frac{d^2 E}{d\varphi^2} - \left(\frac{d^2 E}{d\theta d\varphi}\right)^2 \right], \text{where } \gamma = \frac{2\pi g \mu_B}{\hbar} \tag{6}$$

The new quantity $\omega$ is the frequency $f$ by multiplied $2\pi$. $F$ is the radio frequency (RF) field applied transverse to the sample. The in-plane resonance is observed with a microwave source of 35GHz, as this aligns the magnetization at all angles, which is important as the four-fold anisotropy is observed as an angular dependence in resonance position and the best measurements of it coming from the rotation of the sample in a stationary field about $360^\circ$ [27]. Expanding the second order derivatives in equation 6 and the energy expression in equation 4 gives:

$$\left(\frac{\omega}{\gamma}\right)^2 = \left[ H + \frac{8K_A}{M} \cos 4\varphi_H \right] \left[ \left( H + \frac{K_A}{M} (\cos 4\varphi_H - 1) + 4\pi M + \frac{2K_u}{M} \right) \right] \tag{7}$$

This equation holds assuming that the magnetization is saturated, which means $M$ is aligned with the applied field $H$. When no crystal field anisotropy is present, equation 8 reduces to the form below:

$$\left(\frac{\omega}{\gamma}\right)^2 = H \times (H + 4\pi M) \tag{8}$$

When reduced to the thin film geometry, the demagnetizing fields arise from magnetic poles will then have a much greater influence on the anisotropy than in the bulk.
material. In-plane the distance between the poles will be the largest, so the field lines will exist mostly on the outside of the sample. In contrast, when measured from the out-of-plane orientation, the poles will be tightly packed together and the field lines will pass through the sample. This generates a strong demagnetizing effect. Thin films are also used to explore interface effects by layering additional ferromagnetic layers or even nonmagnetic metals onto the surface [24].

The out-of-plane resonance condition is given by following equation:

\[
\frac{\omega}{\gamma} = H - 4\pi M, \tag{9}
\]

where M is the magnetization of the material and H is the external field being applied. Measurements taken in the out-of-plane orientation do not have any angular component, as the applied field is kept completely perpendicular to the surface of the thin film itself during all measurements. By doing this the damping measured by this method is the average across all potential measurement orientations [29]. This, along with the lower field at which the resonance will be observed in the 10GHz system used, is the primary reason that all out-of-plane measurements were taken at this frequency.

While magnetite is ferrimagnetic instead of ferromagnetic, the anti-parallel domains are minor enough that the specifics of FMR do not need to be modified to explain how we achieve measure the FMR spectra for our samples. First the sample will be placed in an external magnetic field sufficient to saturate the net magnetization of the magnetite samples. At this point an oscillating microwave field is applied perpendicular to the first that serves to kick the magnetic moments. This causes the magnetization to precess about the applied external field with a frequency proportional to the dc magnetic field. When the microwave source is set to the resonant frequency the sample will absorb some of this energy to excite the precession [30]. By measuring energy lost to the absorption the applied field at which this occurs as well as its strength can be observed. An example of the derivative of the FMR absorption signal generated is provided below in Figure 8. The derivative of the signal is used in order that a lock-in-amplifier might be used so that a better signal-to-noise ratio can be obtained.
Figure 10: FMR spectra of magnetite film with 50nm of Ag taken at room temperature sample taken in-plane.

2.2 Damping.

Earlier in this paper the use of thin films to explore the result of having a nonmagnetic metal layered onto a ferromagnetic film and how this could affect the anisotropy of the sample was brought up as an important research interest in both this study and others. The more interesting effect is one of damping, namely by increasing the potential effect of this phenomena on the line width. This quantity is the half width of the resonance at its maximum power, or the field range between the two peaks of the derivative of this graph shown above [26]. This is caused by the loss of energy in the precession of the magnetic moments, which decreases’ the relaxation time. This will show up has a modified resonance in the FMR spectra having larger line-width. The most critical point of this phenomenon is that the injected currents will in turn be spin polarized when a material like magnetite is used. This spin-pumping into a nonmagnetic conductor is of critical importance in the development of new forms of magnetic memory and other micro magnetic devices.
However, this damping effect is effectively limited to the surface of a material. For this reason ultra thin films are required to observe it in the FMR, otherwise the primary spin coupling effects will be between neighboring moments within the magnetic material itself and unaffected by the additional damping layer adhered on top. Should both conditions be met, significant difference in the resonant spectra would be seen between a damped and undamped sample of the same material. It is also important that the sink material have a long spin coherence length to spin currents produced by source material, allowing them to propagate further before losing their polarization. This is why Ag was selected as the nonmagnetic layer in this study.
Experimental Details:

The samples used in this study were single crystals of Fe$_3$O$_4$ grown by reactive sputtering to a thickness of 350nm. In this method the prepared substrate, MgO in this case, is placed in a vacuum chamber. Once a high vacuum as been achieved, a metal boat containing the film material is heated until vaporization occurs. At the same time controlled injection of oxygen is allowed into the chamber so that the Fe will react with it while depositing onto the film. No external field was applied to the films during deposition that would have a Ag bilayer placed on them. Surface properties were verified with X-ray spectroscopy to make sure that good quality magnetite had been grown. Temperature v. resistivity measurements were also taken on the samples at this time to provide further proof of the quality of the

Figure 12: Schematic of FMR apparatus used in this study.

Figure 11: FMR apparatus with 35GHz system in place.
film growth given the difficulty of fabricating magnetite with the proper stoichiometry, as shown in figure 3. The clearly visible Verwey transition was further evidence of the quality of these samples. Later, once growth was completed they were broken into uniform pieces and Ag layers were placed on top of the magnetite surface of varying thicknesses of 10nm, 50nm, 100nm, 200nm, and 500nm. The Ag layer was placed on the samples in order to construct a spin pumping multilayer system of a ferromagnetic, or in this case ferrimagnetic, and nonmagnetic conductor.

The experimental apparatus in used in this study is shown in figure 10 above. The water cooled electromagnet allows for the generation of strong applied H fields over 2T. The orientation of the samples can be changed by exchanging cavities of different frequencies in and out of the magnet. This is a simple procedure, and the microwave source can readily be changed from 10GHz to 35GHz dependent on whether out-of-plane or in-plane measurements are required. The cavity is centered in the magnet adjacent to the Hall probe so that the strength of the applied field against the sample is always known. Both the 35GHz in-plane cavity and the 10GHz have cryostats, though temperature dependent measurements were only taken with the 10GHz. The control of both the magnet and the cryostat are performed using LabVIEW programs, and all data is first recorded through this same method [7].

The in-pane measurements of the magnetite samples verified that the stoichiometry was good, as the four-fold anisotropy was clearly visible in the samples.
The out-of-plane measurement was of more importance as this was the orientation that would allow for the measuring the possible damping effect that the Ag layer had on the magnetite samples. The 10GHz cavity used in this experiment is shown above. Unlike the 35GHz cavity, it has internal modulation coils, and can have samples mounted in either the in-plane or out-of-plane orientation, though this feature was not used during data collection.

Figure 14: 35GHz cavity used for in-plane measurements.
4 Results and Discussion:

The primary topic of interest is the potentially increased damping caused by the additional Ag layers. Before the temperature runs were done however, measurements of the in-plane anisotropy were made to determine if the expected four-fold angular dependence was present. These measurements were taken at room temperature and over 160° arc. Once these measurements had been concluded the samples were then mounted perpendicular on thin quartz rods for the out-of-plane measurements to be taken. These used full effect of the 10GHz cavity available as low temperature measurements were taken so as to observe possible temperature changes in the FMR spectra. There were a total of six samples examined to measure the damping effects of the different Ag thicknesses, ranging from 0nm of Ag to 500nm.

4.1 In-plane at 35GHz

The in-plane measurements did in fact show the expected four-fold symmetric anisotropy. In figure 13, the quality of the in-plane data can be seen, as there is good signal to noise and clear resonances. Three in-plane spectras are shown, with off sets along the y axis so that they may be distinguished from each other. The effect of increasing film thickness on signal strength for our 35GHz cavity can also be seen, as the samples with larger Ag thicknesses were harder to get strong signals from. Still samples all gave fairly strong signals with clearly visible resonances across a 150° or more. Figure 14 contains the graphs with this data plotted. Those graphs show that the optimum oxygen stoichiometry is present in these samples, as their anisotropy in-plane is like that of the bulk magnetite sample. As all samples of magnetite came from the same deposition, the similarities between in-plane anisotropy measurements are expected, and
further show the quality of the samples grown.

Figure 15: FMR spectra for in-plane samples at variable Ag thicknesses. The spectra are offset by 50uV so as to be more distinct.
Figure 16: Resonance v. Angle for in-plane orientation. Graph A is 10nm Ag resonance position v. angle, B is the 50nm Ag, and C is the 200nm Ag.
The graphs in figure 14 show the resonance positions as they vary by angle, from which the four-fold anisotropy of magnetite can be determined. It is even possible to determine the anisotropy constant $K_4$ and $K_u$ from these graphs. Using equation 7 and solving for $H$, which is the applied field at which the resonance occurs, the following equation is obtained:

\[ \frac{1}{2M^2} \left( KM - M^2 M_E - 9KM \cos 4\varphi \right) - \sqrt{(-KM + M^2 N9KM \cos 4\varphi)^2 - 4M^2 \left( 4 K^2 - M^2 \left( \frac{2}{9} \right)^2 - 8K^2 \cos 4\varphi + 8KMN \cos 4\varphi + 4K^2 \cos 8\varphi \right)} = H \]  

(10)

The quantity $N$ in equation 10 is defined below in equation 11:

\[ N = 4\pi M_E = 4\pi M + \frac{2K_u}{M}. \]  

(11)

The resulting fit of in-plane resonance positions is shown figure 15 with experimental data. In this fit, changing $K_4$ alters the amount of anisotropy in the sample, meaning the minimum and maximum resonance position. $K_u$ on the other hand changes the mean at mean at which this is seen. In the table below the anisotropy parameters used to fit the experimental data are listed.
Figure 17: Graphs of in-plane resonance positions with the fit from equation 10.
Looking at the fit in figure 14, it is clear that the magnetite samples show correspondence with the expected four-fold symmetry. There might also be a two-fold contribution as well, but without a full 360° it is hard to say if that is why graph C is so different from the fit line. Using equation 11 and solving for $K_u$ gives:

\[
\frac{M(4\pi M_E-4\pi M)}{2} = K_u, \tag{12}
\]

where $K_u$ is $-0.67\text{Mergs/cm}^3$. The effective magnetization is caused by the sublattice trying to orient the field out-of-plane, with $K_u$ being the parameter that gauges the strength of this effect. Compared to the anisotropy energy associated with demagnetizing field of $2\pi M^2$, which is $1.22\text{Mergs/cm}^3$. Unfortunately, no significant trend is seen in the anisotropy parameters for the increasing Ag thicknesses. The increasing sample thicknesses also produce weaker signals, making it harder to fit them.

### 4.2 Out-of-plane at 10GHz:

In the out-of-plane orientation temperature dependent measurements were taken of all six samples. At room temperature, the different samples gave similar results for FMR spectra, with the line width and resonance position showing little dependence upon the thickness of the Ag layer. However, once the samples were lowered to the Verwey temperature this trend no longer held. The resonance position changed dramatically at this point, dropping to almost half the original field strength in all cases. This can be seen

<table>
<thead>
<tr>
<th>Ag Thickness</th>
<th>In-plane Anisotropy Parameter K4 (erg/cm³)</th>
<th>Magnetization (emu/cm³)</th>
<th>$4\pi M_E=N$ (Oe)</th>
<th>$\omega/\gamma$(Oe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10nm</td>
<td>11600</td>
<td>440</td>
<td>2450</td>
<td>11700</td>
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<tr>
<td>50nm</td>
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<td>13500</td>
<td>440</td>
<td>2400</td>
<td>11700</td>
</tr>
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</table>

Table 1: Values calculated for the fitting of in-plane resonance positions for 10nm, 50nm, and 200nm Ag layers.
in figures 16 and 17 below. Using this data it is also possible to find the effective magnetization of the magnetite samples as a function of temperature. Using the equation 9, and solving for the magnetization constant M:

\[ M = \frac{H \cdot \alpha}{4\pi} \]  

(13)

The results of the magnetization calculation are in figure 18. Magnetization will briefly increase with decreasing temperature. First the effective magnetization gradually increased, as expected because the magnetization M will increase with decreasing temperatures. At the Verwey temperature this trend reversed, and the magnetization decreased in all samples with the thickness of the Ag layer not showing any effect on this or the resonance position. Substantial fall in magnetization at the Verwey temperature is caused by an increasing out-of-plane anisotropy component \( K_u \).

Using equation 12, it is possible to solve for \( K_u \) in the out-of-plane orientation and to compare it with the value obtained in the in-plane orientation. Using the magnetization values graphed in figure 20, the value of \( K_u \) in the out-of-plane orientation is -0.25Mergs/cm\(^3\). The difference between 9.5GHz and 35GHz values for the anisotropy contribution in the out-of-plane direction is interesting. It would be expected for both values to be equal, and not for one to be almost three times the other. The different frequencies used to measure the in-plane and out-of-plane orientations might be the source of this discrepancy, but will remain unresolved issue in this thesis.

<table>
<thead>
<tr>
<th>Temp.</th>
<th>( K_u ) (Mergs/cm(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>~295K</td>
<td>-0.25</td>
</tr>
<tr>
<td>~100K</td>
<td>-0.80</td>
</tr>
</tbody>
</table>

Table 2: Calculated \( K_u \) values at room temperature and after the Verwey transition.
Figure 18: Resonance positions for Ag thicknesses varying with temperature.
Figure 19: All resonance positions v. temperature.

Figure 20: Magnetization values v. temperature for all values. Note that the bulk magnetization for magnetite is 440emu/cm$^3$. 
The out-of-plane orientation is expected to give the best indication of the enhanced damping in these samples from the Ag layer. As such, it is important to note that in figure 21 and 22, there is almost no difference in the line width of any sample at room temperature or for a substantial range from that going towards the Verwey transition. The most likely cause of this is the relative thicknesses of the Ag and magnetite layers. At 350nm of magnetite deposited, these films are not particularly thin. The damping is then not dominated by the surface effects but instead by that of the bulk magnetite itself. This can be seen by comparing the line width of magnetite without Ag to the following five samples. This trend continues as the samples are reduced in temperature towards the Verwey transition.

At the Verwey temperature the resulting change in conductivity and the geometric shift in the lattice structure causes a dramatic increase the damping present in all samples, including those without Ag. The temperature at which this occurs as good correspondence to the change in resonance position as well as historically measured properties of magnetite [14]. The amount that the line width increases was significantly affected by the thickness of the Ag layer. This effect was quite significant in all cases, but particularly the 50nm sample were the line width after the Verwey transition was six times that of a sample with no Ag layer. Looking at figure 22, it is also interesting to note that there is some optimum thickness of Ag to generate the most dramatic increase in damping at the Verwey transition. The rate at which the line width increases at the Verwey transition increases until 100nm of Ag, where after drops down with greater Ag thickness. At 500nm of Ag the line widths after the Verwey transitions are almost the same as when there was no Ag layer at all. Thus the amount of damping present in a sample of magnetite after the Verwey transition has completed is highly dependent on the thickness of the Ag layer.

The increased line width at the Verwey transition shows that there is substantial increase in spin injection and the resulting magnetic damping at this temperature. The change in the magnetic structure and conductivity present at the Verwey temperature is the most likely cause of the increased damping reflected in the larger increase in line width for certain Ag thicknesses. Given the optimum Ag thickness for maximum
damping observed, there must be substantially greater amount of spin injection donating energy to the sink from the magnetic precessions to give this effect. The long Ag spin diffusion length means that those injected spins could propagate to the edge of the sample. As the thickness of the Ag layer is increased, the damping is reduced back to what it was when there was no Ag layer at all. Given this result, the increased Ag thickness must prevent the injection of spins once it grows to a sufficient length, which does not even have to be significant proportion of the 1000nm spin diffusion length of Ag.
Figure 21: Measured line width v. temperature for all samples. Graph A shows the line width without Ag, while graphs B through F show it in cases of increasing Ag thicknesses.
Figure 22: Comparison of line widths for all samples.

Figure 23: Estimated relationship between Ag thickness and line width for magnetite samples.
5 Conclusion:

In this study the magnetic properties of magnetite were the primary topic of interest, with added Ag layer’s effect on damping also serving as an interesting feature. Magnetite’s novel lattice structure has been known for almost half a century, though that is but a fraction of the time the magnetite has been a material of interest in the research of magnetic properties. Magnetite serves as a source of 100% spin polarized currents and its high Curie temperature of 860K. In fact the discovery of ferrimagnetism itself was first seen when studying magnetite. These are not the only interesting property of magnetite, as the temperature dependent transition discovered by Verwey accompanies a host of changing properties related to the now altered lattice structure and conductivity. We have seen in this study that the conductivity change is combined with substantially increased damping.

We also looked at the in-plane anisotropy of magnetite as another method of determining the quality of our samples before continuing to temperature dependent analysis. The measured resonance data and subsequent fitting show good correspondence to the expected four-fold anisotropy. It also allowed for the determination of the out-of-plane anisotropy parameter $K_u$ as well as the in-plane anisotropy parameter $K_4$. Out-of-plane measurements also allowed for another calculation of the out-of-plane anisotropy parameter, as well as the determination of the magnetic damping of our samples.

While magnetite’s potential use as a source of spin-polarized currents was not observed at room temperature, our results show thickness dependent magnetic damping effects at the Verwey transition. There is a substantial increase in damping beyond the Verwey temperature, and a significant effect from the Ag layer on the rate at which damping increased. That there should be an optimum thickness is also interesting, and a potential avenue for further research in spin injection with magnetite multilayers. The lack of contrast between samples above the Verwey temperature is most likely caused by the substantial thickness of the magnetite layers, which causes the bulk properties to dominate over the interface effect that we wished to observe. Still, the clear effect of the Verwey transition on all samples shows that the magnetite that was grown was of high
quality, a significant achievement given the difficulty of producing even bulk magnetite with the correct stoichiometry.
A Appendix: Other Research Projects.

1. Heusler Alloys:

Heusler alloys are complicated magnetic multilayers. In the case of the samples investigated they were Co$_2$MnSi grown on GaAs to varying thicknesses and different complexities. In one of the four samples it was actually to Heusler layers separated by a nonmagnetic conductor. The in-plane and out-of-plane anisotropies were studied in detail at room temperature, with a substantial amount of in-plane measurements taken in order to obtain the damping parameters from our model. This study was done in collaboration with Chris Palmstrom at U.C. Santa Barbara and has been presented at the 2013 APS March Meeting.

2. Fe Films:

Two samples of single crystal Fe films grown to 300nm and 1000nm thicknesses respectively, were studied on a stripline. Room temperature measurements were taken for variable frequency from 10GHz to 24.5GHz. From this data the dispersion relation was obtained and the intrinsic damping. This study was in collaboration with Dan Dahlberg at University of Minnesota and Casey Miller at University of South Florida, and was done to consider the viability of further experiments on similar samples.

3. Core/Shell Magnetic Nanoparticles

Multiple magnetic nanoparticles have been investigated, with core/shell interface effects between magnetically hard and magnetically soft materials. This work was done in collaboration with Josep Nogues from Barcelona. The initial set of two samples alternated between whether the magnetically soft or hard material was the core or shell respectively. In all cases, measurements were conducted in the 10GHz cavity to make use of the superior cryostat for extremely low temperatures. The magnetic nanoparticles were adhered to the end of the sample rods with varnish and data taken down below 10K. A new set of samples was recently obtained, but results have only been of room temperature with new data taken considering the effect of the power used by the microwave source. This has been shown to be a substantial effect on the shape of the resulting FMR spectra.

4. Nickel Films
Ni single film crystals grown on MgO by our collaborators Carl Thompson and Carolyn Ross at MIT had experimental data obtained for in-plane anisotropy and damping with varying temperature down to 50K. Results from these samples were presented at the 2012 March Meeting, and are currently being worked on for publication.
Bibliography


