Effect of Field Cooling on Domain Memory in Exchange Coupled Magnetic Thin Films

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ABSTRACT

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Combining ferromagnetic and antiferromagnetic materials has been shown to improve magnetic domain memory at low temperature. We explored this effect when these materials are brought to low temperature in the presence of an applied magnetic field. Our goal is to determine if this Field Cooling would affect the degree of magnetic domain memory. Our sample consists of alternating layers of Co and Pd paired with an antiferromagnetic IrMn alloy. This combination leads to exchange coupling. The method of X-ray Resonant Magnetic Scattering allows us to get an image specific to the domain morphology. We analyzed the data using a cross-correlation technique. The results of our study show that there is actually less memory when the sample has undergone Field Cooling as well as that the region of highest memory is shifted due to exchange bias.

Keywords: magnetic domain memory, exchange bias, x-ray scattering, cross-correlation
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Chapter 1

Introduction

1.1 Motivation

Data memory is cheaper than ever before, yet the demand continues to grow. Beyond the need for storage for data for companies, government, and personal use, there is currently a proliferation of hand-held devices which require denser memory storage. These demands have led to the investigation of the properties of thin magnetic films. In such films, when the thickness of magnetic layers is finely adjusted, the magnetization tends to prefer perpendicular orientation rather than in-plane. [1] Such systems exhibit ferromagnetic domains, i.e. regions where magnetic moments are aligned as explained in Section 1.2. The size of these domains is a crucial parameter for the potential storage density of the material, and make thin films attractive objects of study.

Most common magnetic memory storage devices currently have two main limitations. First, they are limited in their spatial memory density. As technological devices become increasingly compact, the limiting factor is quickly becoming data storage density, particularly in magnetic-based data storage. [2] The second limitation is the susceptibility to modification or erasure by external magnetic fields. Stray magnetic fields eventually corrupt magnetic data storage devices,
1.2 Ferromagnetism and Hysteresis

so long-term archives must be rewritten periodically (typically every four years). Technology based on our research has significant promise to overcome both of these limitations by both creating a substrate for data storage impervious to external magnetic fields of virtually any strength and one capable of storing many orders of magnitude more data per unit area than the current models. Typically, the smallest magnetic data storing structure possible on ferromagnetic films is the size of a single magnetic domain; however, to be useful for storage, the reversal of domains must be reproducible. This reproducibility of specific domain morphology is what we call magnetic domain memory (MDM). We propose to study the reversal and MDM properties in such films when they have been Field Cooled.

1.2 Ferromagnetism and Hysteresis

Ferromagnetic materials have the property that their domains align to applied fields. One specificity of ferromagnetic materials is the formation of domains. These domains are characterized by the constituent atomic spins all pointing in the same direction; in the absence of other magnetic fields, the domains are normally oriented randomly. However, when an external field is applied, the domains align to the applied field and merge together to form larger domains. With a strong enough field, all of the domains in the sample essentially become one large domain with all the spins aligned to the applied field. This condition is what we call saturating the material.

Another characteristic of ferromagnetic materials is the possible occurrence of hysteresis. As one applies a varying field to the sample, the net magnetization of the sample changes as the domains align to the field. This changing field is often done in a loop. First one would apply a stronger field in one direction, and then apply a field in the other direction. In samples like ours, when one begins to saturate the sample, the net magnetization no longer changes the same way for the ascending and descending branches of the loop. This is hysteresis; the net magnetization of the
1.2 Ferromagnetism and Hysteresis

Figure 1.1 A sample hysteresis loop. Note that the ascending and descending branches have distinct shapes. Points of interest: a) Nucleation is where the domains begin to rapidly change from saturation to match the applied field. b) Remanence is the remaining net magnetization when the applied field is zero. c) The coercive point is when the net magnetization of the sample is zero. d) Saturation is when essentially all of the domains are aligned to the applied field.
sample is path dependent. This is best visualized by Fig. 1.1. The net magnetization of the sample is different for the ascending and descending branches, though it is fairly symmetrical. Note that for the same applied field, such as $H = 0$, the net magnetization of the sample is distinct. This remaining magnetization when there is no applied field is referred to as remanence.

1.3 Magnetic Domain Memory

One may also note that in a given loop there are two points with the same net magnetization. While the net magnetization is the same, the shape of individual up and down domains, or the morphology, may be different. For an example of a standard morphology of this sample refer to Fig. 1.2. If the domains assume the same physical morphology at a given net magnetization after being saturated and then returned to the previous magnetization, then the sample displays memory. In our sample, this memory is generally highest in a "maze state" when the sample has zero net magnetization.

![Figure 1.2](image.png)

**Figure 1.2** An image taken via magnetic force microscopy of our sample with no applied field. Here there are equal amounts of up and down domains visualized as the light and dark lines.
1.4 The Sample

Our sample consists of both ferromagnetic and antiferromagnetic material. Unlike with ferromagnetism, antiferromagnetic domains are characterized not by atomic spins which are aligned in parallel, but by exactly spins which are aligned anti-parallel as shown in Fig. 1.3. While in the bulk of the antiferromagnetic material the spins are anti-parallel so that the net magnetization is zero, at the edge the spins align in parallel. This causes some net magnetization and thus these spins are referred to as uncompensated spins.

The ferromagnetic material in our sample are thin layers of cobalt four angstroms thick. This thickness was chosen to force the spins to point into or out of the plane of the sample rather than in plane. [1] These Co layers are paired with seven angstrom thick layers of palladium. There are then 12 Co/Pd pairs stacked together. An antiferromagnetic layer of an IrMn alloy is then inserted which is 24 angstroms thick. This combination of Co/Pd multilayers and IrMn is repeated four times. This can be seen in Fig. 1.4.
1.5 Exchange Coupling

In general, the alternating spin direction in the antiferromagnetic material creates a net-zero magnetization for the sample. The only place of distinction is at the domain boundaries where there are uncompensated spins. These uncompensated spins interact with the ferromagnetic layers in a process called exchange coupling.

At the interface between the ferromagnetic and antiferromagnetic layers, exchange coupling causes the ferromagnetic spins to align with the uncompensated antiferromagnetic spins, i.e., the "down" domains in the ferromagnetic layer appear adjacent to the uncompensated "down" spins in the antiferromagnetic layer. However, this is only a consistent process when the antiferromagnetic material is brought to low temperature. The uncompensated spins in the IrMn still respond and align to an external magnetic field, but when the sample is brought below its blocking temperature (approximately 300 K for this sample), the spins lock and no longer respond to an external field. The locking of spins gets stronger at lower temperatures. Exchange coupling serves as the basis for creating materials with high MDM.

The cooling of the sample below the blocking temperature can be done either with or without

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**Figure 1.4** Our sample contains 12 pairs of Co and Pd layers combined with an IrMn layer. These are stacked together.
Figure 1.5 This is a zoomed in image of Fig. 1.1 showing the bias introduced from the Field Cooling. The applied field during cooling as applied toward the right of the graph causing the bias to the left.

an external magnetic field. When an external field is present during cooling, this process is called Field Cooling, while Zero Field Cooling (ZFC) would then imply that there is no external field during cooling. The effect of Field Cooling is to introduce a bias to the hysteresis loop. This exchange bias is an effect of both exchange coupling and the field cooling. This effect is clearly seen in Fig. 1.5. This is actually the same graph as Fig. 1.1 only zoomed in. The applied field during cooling as applied toward the right of the graph causing the bias to the left (in Fig. 1.5). The bias is in the opposite direction of the cooling field because the uncompensated spins of the antiferromagnetic layers were locked with a majority of them in the direction of cooling field. This means that there is a net magnetization for the sample in that direction. Thus, it takes a greater external field in the opposite direction to create the same net magnetization in the sample.

1.6 Previous work

Our study of this material began with quantifying the degree of MDM when it was under ZFC conditions. Two former students, Brian Wilcken and Joseph Nelson helped develop some of the
analysis tools used for my study, and applied them to the ZFC case. It was shown that exchange coupling was effective at creating high MDM. [3]

Brian Wilcken focused on creating interfield maps of the MDM in the ZFC state. Data was collected for several hysteresis loops, and interfield maps show the amount of memory when images are compared between applied field ($H_1$) in one loop and the applied field ($H_2$) in another loop. This creates a two dimensional map of the memory. In order to do this, Brian developed a Matlab code to determine the similarity between images. His program isolates the parts of the image which are necessary for determining the degree of MDM, compares the images, and outputs maps of the various memory values. [4] The program will be explained in more detail in Sec. 2.2 and 2.3. Joseph focused on Q-selective memory where Q stands for a given radius of the pattern in the images. [5] These images and how they were obtained will be explained further in Section 2.2.

1.7 Goals

For my study, I have used two techniques: Magnetic Force Microscopy (MFM) and X-Ray Resonant Magnetic Scattering (XRMS). These are complimentary tools. MFM allows us to get a visualization of the physical domain pattern in real space, while XRMS gives a visualization of the domains in scattering space. We have primarily used the data collected via XRMS to quantify the degree of MDM. We have done this by cross-correlating the images. Through this process we determined the effect of Field Cooling on memory and have shown that it actually lowers the degree of memory relative to ZFC, although there is still significant MDM.
Chapter 2

Imaging Techniques

2.1 Magnetic Force Microscopy

Magnetic Force Microscopy (MFM) is a scanning microscopy technique to visualize the size and pattern of the domains in magnetic materials. MFM is an imaging technique closely related to Atomic Force Microscopy (AFM). In AFM, a very small tip, only a few nanometers across at its tip, is vibrated at high frequency (around 100 kHz) near the surface of the sample. A laser is reflected off the tip, and when the tip interacts with the atoms of the sample, the deflection of the laser is measured and used to create a visual image of the height and width of the sample’s surface features. The tip scans the surface horizontally before moving a few nanometers over and traversing the sample again. These nearly one-dimensional passes are combined to create a two dimensional image. MFM builds on this by using a cobalt doped tip so that magnetic interactions can also be measured. After an AFM pass is conducted, the tip raised to a lift height of 30 nm and moves in such a way to counteract the physical topography of the sample so that the only deflections of the tip come from magnetic forces.

MFM has limitations as a tool for measuring magnetic memory because it is difficult to com-
Figure 2.1 This is a diagram of the MFM process showing the lifting of tip to eliminate the effect of the physical features to leave only the magnetic forces.
pare the images easily for similarity and it is hard to consistently measure the exact same location over the course of several images. However, MFM is great for visualizing what the domains look like in real space, so I began with this tool to visualize how the sample looked and behaved physically (for the structure of the sample refer back to Fig. 1.4). Extra *in situ* magnetic imaging capability was added thanks to another member of our group, Andrew Westover. He placed a set of permanent magnets on a translational stage so that they could be raised and lowered beneath the sample. This allowed us to control the field the sample was in and to see not only what the sample

![MFM images](image)

**Figure 2.2** The series of MFM images shows the progression of the sample’s domains as the field goes from saturation toward zero. Image (a) shows the beginning nucleation with just a few domains. Images (b) - (d) show the growth of the original domains. Not that you can see the opening structure throughout.
2.2 X-Ray Resonant Magnetic Scattering

looked like with no applied magnetic field but how the domain pattern changes with applied field. A sample of such images are included in Fig 2.2.

2.2 X-Ray Resonant Magnetic Scattering

X-Ray Resonant Magnetic Scattering (XRMS) is a technique developed in the late 1990s which uses x rays generated by a synchrotron to study magnetic domain morphology. [6] To do this the energy of the x rays are tuned to a resonant edge of the magnetic material in the sample. This means that the energy is such to excite electrons from the (2p) to the (3d) orbital after which the photon is scattered. In our case the resonant edge was the $L_3$ edge of Cobalt at about 780eV. [7] These scattered photons are captured by a CCD camera to create a 2-D image. In order to get the specific energy needed, and of sufficient intensity, these experiments can only be performed at a synchrotron and ours was done at the ALS at Berkeley.

The images we obtain are a transform of the physical features into scattering space. Thus, the radius of the ring corresponds to the period $d$ of the domains according to the transmission geometry version of Bragg’s law

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

where $\lambda$ is the wavelength of the incoming coherent x rays, $n$ is an integer, and $\theta$ is the scattered angle. The angle can be found knowing the distance between the sample and the camera (about 1 m) and the radius of the image(about 5.2 mm). Since our light has a wavelength of 1.59 nm, the periodicity of the domains is found to be about 300 nm; this agrees with the measurements of period obtained using MFM. The image is in a ring shape because of the isotropic nature of the domains(the domains are in a maze state like in Fig. 1.2 rather than ordered stripes) which causes isotropic scattering. These images contain both magnetic and charge scattering. The charge
**Figure 2.3** A diagram of the experiment at the synchrotron showing how the light enters the sample and is captured. The x rays transmit through the sample and are scattered. The scattered photons are captured by a CCD camera. [8]

**Figure 2.4** The angle of the scattered photons can be found by knowing the distances between and sample and the camera and the radius of the image.
2.3 Cross-Correlation Technique

Figure 2.5 A sample XRMS image showing the strong ring feature of the scattering. Red indicates high intensity while blue is low. The shape in the middle is caused by a blocker which catches the unscattered x rays.

scattering comes from the atomic structure of the sample while the magnetic scattering comes solely from the domains in the cobalt. As such, the charge scattering is not beneficial in a study of memory, because one would expect the overall structure of the sample to remain unchanged. Thus the charge scattering would be constant and artificially inflate the degree of similarity between images. The images were processed to isolate the magnetic scattering which gives us the true "shape" of the magnetic structure of the sample. The images were smoothed to approximate the charge scattering envelope in the image and then subtracting the envelope from the raw images to leave only the magnetic "speckle". Fig 2.6 shows why we call it that.

2.3 Cross-Correlation Technique

The images were compared using a cross-correlation technique. In cross-correlation, the images undergo a Fourier transform and are then compared pixel by pixel to determine the degree of similarity between the images. The resulting correlation pattern image has a peak as its main feature. This peak is only a few pixels large. This actually corresponds to the average size of each speckle, because for most of the image comparison shifts there is a low degree of similarity
2.3 Cross-Correlation Technique

Figure 2.6 An image of the isolated speckle. This is zoomed in from a processed image so that the speckle are more visible.

Figure 2.7 A graph showing how the images were processed to isolate the speckle. This is a slice of the intensity of an image such as Fig. 2.5 taken vertically in the middle. The area of zero intensity is where the slice crosses the blocker. By subtracting the smoothed curve from the raw data, the speckle is what remains.
2.3 Cross-Correlation Technique

The process of comparing two images pixel by pixel constitutes the cross-correlation process. The result of which gives us a large central peak as shown. Note that the peak image is zoomed in around the peak since the peak is so small compared to the total image.

except when the images are actually over the top of each other. The resulting similarity tells us how similar the domain morphology of the sample was at the time the images were captured. This is crucial to determine if there is MDM.

Once the peak was created we integrated it to obtain the degree of memory. Since the only region of interest is right around the peak, we integrate only a selected ellipse which surrounds the peak. This integrated value is normalized by dividing it by the correlation values of the images making it up. For instance, if comparing images $A$ and $B$, then $AxB$ is divided by the square root of $AxA$ and $BxB$ to normalize and obtain a value between 0 and 1. This value is called $\rho$ and is indicative of the degree of MDM.
2.3 Cross-Correlation Technique

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Table 2.1 Values of rho for a correlation between a pair of images with given ellipse parameters as we sought to optimize the integration area. Here $a$ stands for the semi-major axes and $b$ for the semi-minor axis of the ellipse. We found that the larger the integration size the better value for $\rho$. This leads us to believe the program was doing something different than we thought it was since we would expect to find an optimum ellipse size.
Chapter 3

Results and Conclusions

3.1 Interfield Maps of Magnetic Domain Memory

The goal of all this optimization of the images and their correlation is ultimately to allow us to characterize the degree of MDM in the sample. This research was to determine to effect applying an external magnetic field during cooling affects MDM. Thus, with these values of $\rho$ at our disposal, we can organize them in order to visualize the degree of memory.

The first method of organization is to characterize the memory only at the return point and the conjugate point of the hysteresis loop (as a reminder of what the return point and conjugate point are see Fig. 1.1). Our analysis of the memory initially lends it self to being analyzed by the type of branch, RPM vs CPM, the amount of separation between loops, and the value of the applied field during cooling. The decrease we see in the CPM vs the RPM was expected in Fig. 3.1. The reason that we would expect this is that the bias introduced by FC is is less than the step size of measurements (150 Oe vs 300 Oe). Thus the conjugate points do not fully line up. Even in ZFC we found that the CPM was slightly lower than the RPM, but this bias leads to a greater difference between the two points. Further, it is interesting to note that the memory decreases, both in RPM
3.1 Interfield Maps of Magnetic Domain Memory

These graphs of $\rho$ at return points and conjugate points show that the memory decreases as one goes further from the initial loop.

Figure 3.2 These interfield maps compare the values of $\rho$ at 1280 Oe for $H_C$ on the left and at 2240 Oe for $H_C$ on the right. Note that the maximum for 1280 Oe map is higher and the plateau of relatively high MDM is larger.

and CPM as the applied field during FC is increased.

The next analysis we performed consisted of creating two-dimensional maps of $\rho$. This analysis is more robust than the one-dimensional graphs and consists of finding $\rho$ not only at the return point or conjugate point, but finding the memory between all pairs of fields on the hysteresis loop. The applied field during the loop is referred to as $H$, thus these $H \times H$ interfield maps consist of values of $\rho$ at every field combination.
Figure 3.3 This graph shows that the greater the field applied during cooling ($H_C$), the less memory there is. It also shows that there is a trend for the maximum $\rho$ to occur at a greater field values in the loop. The curves also become less symmetrical. Finally, it is important to know that eventually the MDM effectively disappears.
There are some distinctive characteristics of the maps in Fig. 3.2. The first point of interest is the low degree of memory at nucleation and saturation. Both of these points have a similar process occurring of the sample transitioning to or away from a single domain direction. The fact that the memory is so low implies that this process is quite random. Another prominent feature is that the highest memory is indeed along the axis of the return points where $H = H$. This shows that RPM is higher than any other. The final point of interest is the high memory in the coercive region evident by the plateau of higher memory in region. This is where the net magnetization of the sample is near zero, i.e. there are approximately equal numbers of up and down domains. Note that this does not occur when the applied field is zero, but further up the loop at the coercive point (refer to Fig. 1.1 for visual clarification). This is caused by the exchange coupling between the antiferromagnetic and ferromagnetic layers. The antiferromagnetic layer serves as a template which, due to the low temperature, does not respond to the varying external field. Thus, the sample is encouraged to return to a domain morphology similar to the state it had when the antiferromagnetic spins were locked in place.

The fact that the MDM goes to only 10% or so with the greatest $H_C$ illustrates that MDM is indeed a consequence of exchange coupling. When that large of a field (3200 Oe or 0.32 T) is applied during cooling, the antiferromagnetic layer's uncompensated spins are largely saturated. This effectively removes the template the ferromagnetic domains were using to return to the same morphology and eliminates the MDM.

### 3.2 Comparison Between Zero Field Cooling and Field Cooling States

When these results found in Field Cooling are compared to the ZFC results found previously, it is quickly apparent that there is lower memory in FC. When the sample underwent ZFC, it exhibited
3.3 Conclusions

MDM of over 90%. This high memory even persisted after several loops. Our FC sample still shows high memory, but only in the range of 50% to 60%. There is also a larger decrease in MDM between after several loops. It is also interesting to note that the greater the field applied during cooling ($H_C$), the greater the decrease in memory.

3.3 Conclusions

Exchange coupling provides a good method for inducing high memory in the coercive region. This has been shown through both ZFC and Field Cooling measurements. It had been previously shown that ferromagnetic material can exhibit high MDM through the use of defects. [9] That was usually in the nucleation stage, however. Using antiferromagnetic materials and the induced exchange coupling provides a template for the ferromagnetic layer to exhibit high MDM in the coercive region where the net magnetization of the sample is near zero. It appears, however, that Field Cooling reduces the benefit of the exchange coupling and lowers the MDM of the material.

3.4 Future Directions

These results, while significant, are really only the start of what we can do. In order to really determine the optimal configuration for magnetic domain memory, we will need to vary more parameters. We plan to do further experiments in which the step size in the applied field is smaller to produce a fuller range of data. We also have already begun to do some experiments at higher temperatures and will continue to explore that variable. We also will explore the effect of film thickness on memory. And finally, we would like to expand this research into other types of magnetic material other than cobalt. This area of research offers to greatly improve magnetic data storage as a viable long-term option; we want to add to the fundamental knowledge of these materials so that the best choices can be made.
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