Interaction of Stress and Magnetic Properties in Patterned Copper-Nickel-Copper Thin Films

by

Elizabeth Friend
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Author: ____________________________________________________________
Department of Materials Science and Engineering
September 5, 2006

Certified by: _______________________________________________________
Caroline A. Ross
Professor of Materials Science and Engineering
Thesis Supervisor

Certified by: _______________________________________________________
Robert C. O’Handley
Senior Research Scientist
Thesis Supervisor

Accepted by: _______________________________________________________
Samuel M. Allen
POSCO Professor of Physical Metallurgy
Chair, Departmental Committee on Graduate Students
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ABSTRACT

In recent years, the epitaxial Cu/Ni/Cu thin film system has been extensively studied, due to its wide range of perpendicular magnetization. It has proved to be a model system to explore the interactions of strain, surface energies and magnetic properties. For that reason, is also an ideal system to explore the effects of patterning. It is expected that the miniaturization of patterned magnetic devices will be accompanied by a transition from polycrystalline to epitaxial films. This transition will require a detailed theoretical understanding of the interaction of strain and magnetic properties in patterned epitaxial magnetic thin films. The Cu/Ni/Cu film system is used in this work to explore a triaxial model for an orthorhombic symmetry of strain. By patterning the Cu/Ni into nanolines and measuring the resulting magnetic anisotropy, the validity of the model has been tested.

It has been shown that upon patterning certain thicknesses of nickel into nanolines, the easy axis of magnetization shifts from out of the film plane to in-plane, transverse to the line direction, an observation at odds with the direction of magnetization predicted by shape considerations alone. This transition is explained by the dominant magnetoelastic energy for the Cu/Ni/Cu nanoline system. The resulting anisotropy values are consistent with strain relief values predicted by finite element modeling.

In addition, the low temperature properties of the Cu/Ni/Cu epitaxial film system have been explored. The variation of the overall magnetic anisotropy as a function of temperature is found to be proportional to the cube of the reduced magnetization. In addition, the easy axis of magnetization for certain thicknesses of nickel has been found to shift from in-plane to perpendicular with the reduction of temperature.

Thesis Supervisor: Caroline A. Ross
Title: Professor of Materials Science and Engineering

Thesis Supervisor: Robert C. O’Handley
Title: Senior Research Scientist
To Nick,
without whose love, support and patience
none of this would have been possible.

To my parents,
who have been there for me from day 1.

And to my grandfather (in memoriam),
who never failed to tell me that he was proud of me – repeatedly.
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To my husband Nick for the proofreading help. You may not have understood what you were reading, but you prevented many a grammatical error from slipping into the text.
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Chapter 1
Introduction

Every year, the amount of information that can be stored in a given square inch on a computer’s hard drive approximately doubles. As the information density increases, however, it will eventually reach a limiting value using existing conventional media technology. This limit is set by thermal fluctuations that destroy the magnetically stored information. New technologies must be developed to overcome this limit, probably technologies that use three-dimensional, patterned magnetic structures instead of the traditional two-dimensional thin-film hard disk media. Further, advanced read heads and emerging technologies such as magnetic random access memory (MRAM) rely on thin films patterned to nanometer dimensions. Key to developing these technologies is a fundamental understanding of how the magnetic properties of these films are affected as they are patterned into nanostructures.

Most films used for magnetic nanostructures have traditionally been polycrystalline, due to the ease of growth. Researchers studying the magnetic properties of nanostructures have focused primarily on the effects of shape and crystalline anisotropy. While polycrystalline films do have some strain, due to the growth conditions, the effects of strain have not been studied extensively. The alloys popularly chosen are often of zero or low magnetostriction compositions so that any strain effects are minimal. In studying patterned structures with nonzero magnetostriction, it would be expected that the effect on the overall anisotropy should be characterized empirically.

Polycrystalline films have remained popular in industry, even as the shrinking lateral dimensions have forced development of new alloys and growth conditions to permit smaller grain sizes with the desired magnetic properties. In the not too distant future, however, miniaturization will require structures made from epitaxial films, which will no longer have the problems caused by multiple-grain architecture. Transitioning to epitaxial structures will result in advantages, due to the efficiencies of having a single grain of known orientation, yet will require an improvement in our understanding of the magnetic properties of epitaxial nanostructures.
Epitaxial films often have large biaxial strains. Small area structures such as nanolines can relax at the surfaces, so the strain of a nanostructure can be significantly reduced in the narrower dimension from the thin film value. This altered state of strain will, in turn, affect the magnetic properties of the patterned epitaxial thin film.

This thesis studies a model epitaxial system, Cu / Ni / Cu / Si (100) in order to improve our understanding of the complex interplay of all the factors that influence the magnetic properties of thin films in nanoscale lateral dimensions. These factors include, first and foremost, a reduction in the symmetry of the in-plane strain from the biaxial film to the patterned structure and the effects of this on the magnetoelastic anisotropy. Also to be considered are possible changes in magnetostatic energy and surface magnetic energies (magnetocrystalline and magnetoelastic). These changes have to be considered in the total magnetic energy of the patterned structure which also includes intrinsic magnetocrystalline anisotropy and applied field (Zeeman) energy. Finally, the different temperature dependence of each of these terms needs to be accounted for because most magnetic devices work over a significant temperature range.

Since the mid-19th century, researchers have observed that samples placed in a magnetic field undergo a small strain, due to the magnetostrictive effect. The effect of strain on magnetic properties (the inverse magnetostrictive effect) was observed by Buckley and McKeehan in 1925 [1], who noted that the magnetic hysteresis loops of nickel and nickel-iron alloys (68% Ni, 32% Fe) changed drastically upon application of strain to the samples. In thin epitaxial films, whose internal stresses can be large, the magnetic properties often change dramatically due to inverse magnetostriction (Figure 1).

\[ \sigma = 0 \]
\[ \sigma \neq 0 \]

**Figure 1:** Illustration of inverse magnetostriction (after O'Handley [2]) – a sample has no preferred direction of magnetization when the stress is zero; under compressive stress, the easy axis of magnetization rotates perpendicular to the applied stress (for \( \lambda_S > 0 \)).
Another major effect unique to thin films is that of the surfaces. At an interface, the crystal structure will often deviate from the bulk structure. Reordering of atoms, and a corresponding reorientation of their spins, will result in changed magnetic properties. In a bulk solid, this effect will be negligible, but in a thin film system, it can be significant [3]. The various energy terms mentioned in this section are defined below.

1.1 Cu/Ni/Cu Thin Film Properties

One magnetic thin film system that has been studied extensively for the past 16 years is the epitaxial Cu / Ni / Cu (001) system. At thicknesses of Ni below \( \sim 120 \) Å, the magnetization is essentially perpendicular, which is atypical of thin films, where magnetostatic considerations usually dictate in-plane magnetization. Other thin film systems, such as Fe/Ag (001), also have shown perpendicular magnetization, [4] but only at extremely small film thicknesses, making extensive quantitative study more difficult. Researchers studying Cu/Ni/Cu have offered conflicting theories to explain the origins of the perpendicular magnetization. A review of the relevant literature is in order.

In the late 1970s and early 1980s, the magnetization of Ni/Cu superlattices was studied in detail by researchers hoping to observe novel effects due to coupling between neighboring layers [see 5]. Early reports of enhanced magnetization were contradicted by later studies and the study of Ni/Cu superlattices was supplanted by studies of single layer Ni/Cu films. In 1990, Chang [5] observed a reversal in magnetic anisotropy between polycrystalline Cu (1000 Å) / Ni (\( t_{Ni} \)) / Cu (1000 Å) structures with \( t_{Ni} = 50 \) and 1000 Å. Films with \( t_{Ni} = 100 \) Å had near-isotropic magnetization states in-plane and out-of-plane. Chang’s x-ray data of the lattice constant of the Ni, showing tetragonal distortion, suggested that the magnetic properties were correlated to the Ni strain state.

Perpendicular magnetization at other nickel film thicknesses in the Ni/Cu/Si single layer epitaxial thin film system was later observed by other researchers [6, 7]. Bochi et al [8] grew Ni (\( t_{Ni} \)) / Cu (3000 Å) / Si films, observing a transition at \( t_{Ni} = 60 \) Å from perpendicular to in-plane magnetization in situ using surface magnetooptic Kerr effect (SMOKE) magnetometry. TEM microscopy images showed misfit dislocations above \( t_{Ni} = 15 \) Å, and also indicated that the Ni film strain was much higher than its predicted equilibrium value. The anisotropy of the system was modeled as a balance between the bulk magnetoelastic and magnetostatic...
energies and the surface magnetocrystalline anisotropy, with an estimated surface magnetocrystalline constant, $K_s$, of 0.3 erg/cm$^2$.

The oxidation of the Ni films made *ex situ* study of the Ni/Cu films difficult, so research turned to Cu/Ni/Cu sandwiches. Perpendicular magnetization in epitaxial Cu (50 Å) / Ni / Cu (1500 Å) sandwiches was observed by ferromagnetic resonance (FMR) by Naik et al [9] for $t_{Ni} = 50$ Å. The films with $t_{Ni} \geq 100$ Å had magnetization in-plane. Jungblut et al [10] studied wedge samples of Au (25 Å) / Cu (10 Å) / Ni (0 < $t_{Ni} < 130$ Å) on Cu (100), observing perpendicular magnetization for $t_{Ni} < 115$ Å. The magnetic anisotropy was modeled as a sum of volumetric (magnetoelastic, magnetostatic and magnetocrystalline) and surface anisotropy terms, with the surface anisotropy term – a sum of the positive magnetocrystalline [$K_N$] and negative magnetoelastic [$K_\lambda$] (for $t_{Ni} > t_c$)$^1$ terms – creating the peak in the perpendicular anisotropy at the critical thickness for dislocation formation, $t_c$.

![Figure 2: Jungblut - total magnetic anisotropy * $t_{Ni}$ vs. $t_{Ni}$. Left slope: 2$K_N$ (Néel surface anisotropy). Right slope: 2$K_S$ ($K_S = K_N + K_\lambda$ [magnetoelastic interface anisotropy]) [10]](image)

Bochi et al [11] measured the anisotropy of Cu (50 Å) / Ni ($t_{Ni}$) / Cu (2000 Å) / Si (100) films, observing perpendicular magnetic anisotropy below 140 Å, with the perpendicular anisotropy peaking well above the critical thickness, indicating that the Jungblut model did not sufficiently describe the system [12]. They modeled the anisotropy as a sum of magnetostatic, magnetoelastic (bulk and surface) and magnetocrystalline (surface) energies:

1 Below $t_c$, strain does not vary with thickness, so $K_\lambda = 0$
where $B_S = -50$ and $K_S = 0.85$ erg/cm$^2$ [13]. Further, the variation of nickel strain with film thickness was found by them to fit the empirical relation $\varepsilon(t_{Ni}) = (0.18/t_{Ni})^{0.7}$ using optical interferometry to measure wafer curvature changes after film deposition [12].

Figure 3: Bochi - total magnetic anisotropy $^* \ t_{Ni}$ vs. $t_{Ni}$. Solid line – model, including $B_S$. Dashed line – model excluding $B_S$. Ha et al [14], arguing that Bochi’s value of $B_S$ was overly large for a surface energy, sought to fit his torque magnetometer anisotropy data to a model containing second order bulk and surface magnetoelastic energy terms:

$$K_{Eff} = -2\pi M_s^2 + 2 \left( B_i + \frac{B_S}{t_{Ni}} \right) \varepsilon(t_{Ni}) + \frac{2K_S}{t_{Ni}}$$

where $D_{Eff}$ and $D_S$ are the bulk and surface second order magnetoelastic anisotropy parameters, respectively. Ha calculated the value of $B_S$ to be -1.22 erg/cm$^2$, and extracted fit values for $K_S$, $D_S$ and $D_{Eff}$ of 0.72, -1.94 erg/cm$^2$ and $1.1 \times 10^{10}$ erg/cm$^3$, respectively, using a phenomenological strain-thickness dependence of $\varepsilon = 0.26 \ast (27/t)^{2/3}$ derived from synchrotron x-ray studies [15].
Figure 4: $H_a - K_{Eff}*t_{Ni}$ vs. $t_{Ni}$. Solid line: model with 2nd order magnetoelastic anisotropy

Overall, the Cu/Ni/Cu system has been ideal for detailed study of the interaction between film strain and magnetic properties, highlighting the crucial role played by strain, interfaces microstructure and magnetic domain structure. Because the magnetic properties of the Cu/Ni/Cu epitaxial film system depend strongly upon strain, any changes to the strain, such as are expected to occur upon patterning, will significantly alter the thin film properties.

1.2 Small-Area Strain Effects

Epitaxial thin films have been used for decades in the electronics and optics industries, due to their high quality. Many novel devices have required materials difficult to grow epitaxially, due to lattice constants dissimilar from the substrate materials currently in use. When grown in small thicknesses, the film, if the mismatch is less than a few percent, may deposit pseudomorphically, matching its crystal structure to that of the substrate below it, but above a critical thickness, the volumetric strain energy outweighs the surface energy required to introduce dislocations, permitting the relief of strain by dislocation formation [ref - 16].

Many methods have been explored to grow high quality lattice-mismatched films, such as buffer layers, compositional grading, pseudomorphic multilayer films. One method, briefly alluded to by Matthews et al [17], was the notion of reducing the substrate size: “there are circumstances in which the elastic strain in a film is sensitive to the lateral dimensions of the film and to its dislocation content. … many and perhaps all of the dislocations which extend from the substrate to the surface of the [film] and escape at the specimen edge.”
The concept of small-area dislocation growth was extended to the growth of Ge$_x$Si$_{1-x}$ alloys on small-area pads of Si by Luryi and Suhir [18]. They found that for a given mismatch, the critical thickness for dislocation formation strongly depended on the lateral dimensions of the pads. For sufficiently small pads, dislocation-free alloy films could be grown (Figure 5).

Figure 5: Relation between misfit and critical lateral size for Ge$_x$Si$_{1-x}$ alloys on Si (Luryi and Suhir [18]), $l$ = pad size, $h$ = film thickness

Growth on patterned substrates has been explored in detail for many film systems, including SiGe [16], and, more recently, for GaN [19] and GaN/AlN multilayers [20]. These small-area epitaxial structures have stimulated study of the complementary question, namely, what is the relationship between the in-plane strain and the lateral dimensions.

Figure 6: Left, Raman spectra at different locations across 20x20 µm$^2$ squares. Right, stress vs. lateral position for micro-squares of various sizes [Wang et al, 19]

As indicated in Figure 6, the stress in a patterned structure is strongly dependant upon the lateral position within the line cross-section and structure size. The center of a large structure
has a minimal amount of strain relief, while the corner of a small structure is nearly totally relaxed. This effect was measured for tungsten lines using x-ray diffraction by Maniguet et al [21], who found that the longitudinal strain was nearly unchanged, while transverse strain and both longitudinal and transverse stress were reduced by an amount that was greatly dependent upon the aspect ratio of the lines. Stoica and Vescan proposed a simple exponential model for the strain as a function of position for a line cross-section [22]:

$$\varepsilon(y,z) = -f e^{-\lambda z} \cos(\pi y^{0.5}/L)$$

where $f$ is a material-dependent prefactor, $y$ and $z$ are the horizontal and vertical distances from the center point of the substrate-line interface and $L$ is the width of the line. Similar models have also been proposed for mesa structures [23] and thin- or compliant-substrate systems [24]. UV-Raman measurements of strain by Lei et al [25] confirmed that biaxially strained films patterned into 90 nm wide (200 nm period) gratings are under uniaxial strain, with 80% transverse strain relaxation and ~5% longitudinal strain relaxation, in agreement with finite-element simulation and the models proposed by Stoica and Vescan.

### 1.3 Small-Structure Magnetic Properties

To understand the effect of patterning Cu/Ni/Cu epitaxial thin films into nanolines, it is necessary to first discuss the forces affecting the magnetic properties of nanoscale structures. The previous section discussed changes to the strain upon patterning, which will have an effect on the magnetoelastic energy. Nanoscale patterning will also result in size-dependent effects independent of the strain, namely magnetostatic and surface energy changes.

#### 1.3.a - Magnetostatic Energy

One important factor contributing to the magnetic anisotropy in any non-spherical system is magnetostatic energy. The magnetostatic energy density is $u_{MS} = -M \cdot H_D$, where $M$ is the magnetization vector and $H_D$ is the demagnetization field. Therefore for a given magnetization, the magnetostatic energy is dependent on the field originating from magnetization discontinuities at the surface of the sample.

A magnetized sample can be thought of as an array of microscopic magnetic dipoles, as illustrated in Figure 7, where each dipole has a magnetic field emanating from its north pole and terminating at its south pole. If the dipoles are aligned side to side in parallel ($a$ – second
from top), the dipoles’ magnetic fields will add together. If the dipoles are arrayed end-to-end in parallel (a – second from bottom), the magnetic field emitted by the bottom dipole will flow into the top dipole, reducing the net field emitted. If the dipoles are aligned side-to-side antiparallel (a - bottom), much of the magnetic field emanating from one dipole will flow into its neighbor and vice-versa. Only at the sides will there be a stray magnetic field.

Figure 7: Illustration of a) relative dipole energy and b) magnetostatic energy for a thin film (after O’Handley)

Figure 7b shows the difference between magnetizing a thin film system parallel and perpendicular to the sample plane. A film magnetized in-plane with an array of parallel dipoles (b – top left) will have a few uncompensated dipoles at the right and left surfaces, a moderately high energy state. If the same array of dipoles is aligned in antiparallel rows (b – bottom left), the dipoles at the right and left surface will cancel each other out far from the surfaces, lowering the magnetostatic energy. Perpendicular magnetization will always be a high magnetostatic energy state, due to the greater number of uncompensated spins at the top and bottom surfaces. If the array is antiparallel (b – bottom right), the energy will be moderately low, whereas an array of perpendicular, parallel dipoles (b – top right) will be a high energy state.

By analogy, magnetization of a patterned magnetic line along the length of the line will give a low magnetostatic energy state, while magnetization transverse to the line length will result in a high energy state, and magnetization normal to the closest surfaces will give the highest magnetostatic energy. The difference between the low and high energy states is called the shape anisotropy.
1.3.b - Surfaces

As mentioned in a previous section, another mechanism by which thin films may have different properties from bulk samples comes from the effect of interfaces on the magnetic anisotropy (both magnetocrystalline and magnetoelastic). This can (in many cases) be understood as an outgrowth of the exchange energy, which causes neighboring spins to line up in parallel. If the surface layer of spins is pinned (for whatever reason) in a direction different from the easy axis of the material’s interior, the successive layers of magnetic spins will have to transition to align with the interior magnetization direction. Figure 8 illustrates one example of interface anisotropy – the pinning of a ferromagnet by an antiferromagnet (exchange bias).

![Figure 8: Illustration of a surface pinning effect - exchange bias](image)

Nanoscale structures will be surrounded by interfaces which have chemical and/or mechanical states different from those in the sample interior. These states will cause alterations to the magnetic properties, the magnitude of which depends upon the strength of the interfacial interactions and the extent to which the surface effect propagates into the sample interior. The surfaces of the nanolines should thus have properties significantly altered from the properties of the interior – an effect that will be stronger for the thinner films and narrower lines.

1.4 Organization of Thesis Text

This thesis covers two main topics: the effect on the magnetic properties of patterning thin epitaxial strained films into nanolines and the thermal properties of Cu/Ni/Cu. Chapters 2, 3, 4, 5 and 6 will treat the first topic via discussions of the magnetic anisotropy model, simulations of strain for nanoline cross sections, experimental methodology of nanopatterning, data and discussion. Chapter 7 will cover the low temperature Cu/Ni/Cu data and discussion.
Finally, a note on the conventions used in the text. I have used Å for film thicknesses and nm for longitudinal distances (ex: nanoline width). CGS units have been used for the magnetic properties, except as noted.
Chapter 2
Triaxial Anisotropy Model

This chapter develops the three-dimensional model of the Cu / Ni / Cu nanolines’ energy. First, the uniaxial model is revisited, with parameters updated. The strain state of patterned nanolines is briefly examined. Patterning will break the biaxial in-plane symmetry from tetragonal to orthorhombic, necessitating a triaxial anisotropy model, and will introduce new surface terms and demagnetization factors. The second-order magnetoelastic energy is presented in its generalized form. Finally, the triaxial anisotropy model is presented in full.

2.1 Uniaxial Model Parameters

As discussed in Chapter 1, previous researchers developed an anisotropy model to treat the Cu / Ni / Cu continuous thin film energy state, specifically the anisotropy between the in-plane and out-of-plane magnetization states. This uniaxial model contains magnetostatic, magnetoelastic (1st and 2nd order) and magneto-crystalline surface energy terms [26]:

\[ E = -2\pi M_S^2 + \left( B_1 + \frac{2c_{11}}{t_{Ni}} \right) \varepsilon_0 + \tilde{D}_{eff} \varepsilon_0^2 + \frac{2K_s}{t_{Ni}} \]

where \( M_S \) is the saturation magnetization, \( B_1 \) is the bulk first order magnetoelastic energy coefficient, \( c_{11} \) and \( c_{12} \) are the bulk elastic energy coefficients, \( B_s \) and \( K_s \) are the surface magnetoelastic and surface magneto-crystalline terms, respectively, \( t_{Ni} \) is the nickel film thickness, \( \tilde{D}_{eff} \) is the bulk second order magnetoelastic anisotropy coefficient and \( \varepsilon_0 \) is the biaxial in-plane strain. The uniaxial energy equation omits terms of negligible magnitude, namely the 2nd order magnetoelastic surface and bulk magneto-crystalline anisotropy terms.

The surface and second-order parameters, \( B_s, K_s \) and \( \tilde{D}_{eff} \), will be crucial in any calculations of the nanolines’ energy, so it is important to revisit their derivation to establish accurate values before continuing.

\(^2 \tilde{D}_{eff} \) should not be confused with \( D_{eff} \), the nonlinear magnetoelastic coupling coefficient, defined for magnetoelastic cantilever bending experiments as:

\[ \Delta \sigma = B_{eff} \varepsilon = (B_1 + D_{eff} \varepsilon) \varepsilon \]
Ha [26] used the above equation and his measured torque magnetometer data to determine the magnitudes of the parameters $\tilde{D}_{\text{Eff}}$ and $K_s$. The other unknown parameters, the first and second order magnetoelastic surface parameters $B_s$ and $D_s$, were calculated using Néel’s second-order spin-pair model. As can be seen in Figure 9, this model fits Ha’s measured data for a wide range of thicknesses. But Ha’s torque magnetometer data was significantly lower in magnitude than the data from previous researchers, including Bochi [27] and Jungblut [28]. Ha’s fit value of $\tilde{D}_{\text{Eff}}: -11.0 \times 10^9$ erg/cm$^3$ is significantly greater in magnitude than subsequent directly measured values of $D_{\text{eff}}$: $-2.34 \times 10^9$ erg/cm$^3$ (Gutjahr-Löser et. al. [29]) and $2.50 \times 10^9$ erg/cm$^3$ (Ciria et. al. [30]).

Figure 9: Effective uniaxial anisotropy times $t_{\text{Ni}}$ versus $t_{\text{Ni}}$, after O'Handley [31]

One question that arises is how Ha’s lower-magnitude torque data affected his fit to the parameters extracted in his model. Table 1 lists the model parameters for both the Bochi and the Ha fits. There is only a small discrepancy between the $K_s$ values, but a large difference between the values of $B_s$. [The Bochi model was first-order only, relying on a large surface magnetoelastic energy constant, $B_s$, to give the correct curvature to fit the data.]

---

3 This is an apples-to-oranges comparison, although Ha’s extracted $\tilde{D}_{\text{Eff}}, -11.0 \times 10^9$ erg/cm$^3$, can be divided by $(1+2c_{12}/c_{11})$ to permit an approximate comparison value of $-4.8 \times 10^9$ erg/cm$^3$. 26
Table 1: Bochi and Ha Cu / Ni / Cu thin film model parameters (*calculated* and fit)

<table>
<thead>
<tr>
<th></th>
<th>$K_s$ (erg/cm$^2$)</th>
<th>$B_s$ (erg/cm$^2$)</th>
<th>$\tilde{D}_{Eff}$ (erg/cm$^3$)</th>
<th>Fit Error$^4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bochi 1996</td>
<td>0.88</td>
<td>-52</td>
<td>-</td>
<td>0.14</td>
</tr>
<tr>
<td>Ha 1999</td>
<td>0.72</td>
<td>-1.22</td>
<td>-11.0 x10$^9$</td>
<td>0.95</td>
</tr>
</tbody>
</table>

To determine accurate values for $K_s$ and $B_s$, it was necessary to repeat the fit of Ha’s second-order uniaxial equation to data, using a simulated value of $\tilde{D}_{Eff}$ $^5$ [32] and newly measured torque magnetometer data of continuous Cu / Ni / Cu thin films (see Chapter 5 for discussion of torque magnetometry). Figure 10 shows the new torque magnetometer data for the series of Cu / Ni / Cu continuous thin films with the revised-parameter model.

Figure 10: Effective anisotropy times Ni thickness versus Ni thickness, with newly measured torque magnetometer data and revised-parameter model

The newly extracted parameters are given in Table 2. For details on the mathematical procedures for the fit, see Appendix 1.

---

$^4$ Fit error is given with respect to the average of the Friend and Bochi $K_{2eff} \ast t$ data sets.

$^5$ Per $\tilde{D}_{Eff}$ equation described in section 5 of this chapter.
Table 2: Revised model parameters

<table>
<thead>
<tr>
<th>$K_s$ (erg/cm$^2$)</th>
<th>$B_s$ (erg/cm$^2$)</th>
<th>$\tilde{D}_{\text{eff}}$ (erg/cm$^3$)</th>
<th>Fit Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.506</td>
<td>-17.95</td>
<td>-1.85 x10$^9$</td>
<td>0.15</td>
</tr>
</tbody>
</table>

Although the resulting values for $K_s$ are slightly smaller than either Ha’s or Bochi’s, the value for $B_s$ is in between Ha’s and Bochi’s and the fit error is low, permitting good confidence in the extracted parameters. As can be seen in Figure 10, the revised parameters give a good fit to both the new data and the data measured by Bochi et al.

### 2.2 Nanoline Strain State

For continuous Cu/Ni/Cu thin films, the strain state is biaxial:

$$
\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_0 = \eta(t_{Ni})
$$

$$
\varepsilon_{zz} = \frac{-2c_{12}\varepsilon_{xx}}{c_{11}}
$$

The two in-plane directions ($x$ and $y$) have equal magnitudes of tensile strain, equal to the misfit strain $\eta$ for that film thickness value. Out of plane ($z$), the strain is compressive, related by the Poisson ratio to the in-plane strain.

Once patterned into nanolines, the strain state in-plane is changed, due to transverse strain relaxation at the nanoline edges (the longitudinal strain should be largely unchanged). To capture the average strain in the $y$ (in-plane, transverse to lines) and $z$ directions, the average stress in the $y$ direction is estimated as $f$ times the unrelaxed stress in the $x$ (parallel to lines) direction, where $0 \leq f \leq 1$.

$$
\sigma_y = c_{ij}\varepsilon_{jj}
$$

$$
\sigma_y = c_{11}\varepsilon_{yy} + c_{12}(\varepsilon_{xx} + \varepsilon_{zz}) = f\sigma_x
$$

$$
\sigma_z = c_{11}\varepsilon_{zz} + c_{12}(\varepsilon_{yy} + \varepsilon_{xx}) = 0
$$

Solving for $\varepsilon_{yy}$ and $\varepsilon_{zz}$ in terms of the $\varepsilon_{xx}$ gives:

$$
\varepsilon_{yy} = \frac{\varepsilon_{xx}(fc_{11} - (1-f)c_{12})}{c_{11} + (1-f)c_{12}}
$$
\[ \varepsilon_{zz} = \varepsilon_{xx} \left( \frac{-c_{12}(1 + f)}{c_{11} + (1 - f)c_{12}} \right) \]

In other words, if \( f = 0 \) (complete transverse strain relief), the strain state will be uniaxial:

\[ \varepsilon_{yy} = \varepsilon_{zz} = \varepsilon_{xx} \left( \frac{-c_{12}}{c_{11} + c_{12}} \right) \]

while if \( f = 1 \) (no strain relief), the strains are the same as the original biaxial strain state.

It is expected that the nanolines’ strain state will be intermediate between the biaxial and uniaxial extremes. This is explored further in Chapter 3 via simulations.

### 2.3 Surface Energies

The continuous thin film model dealt with two surfaces – the Ni-Cu interfaces on top and below the nickel thin film, separated by a distance \( t_{Ni} \), as shown in Figure 11.

![Figure 11: Ni-Cu top and bottom interfaces](image)

Each surface contributes both to the magnetocrystalline and to the magnetoelastic energy in the form:

\[ E_{\text{surface},Z} = \left[ \frac{K_{\text{surf,Cu-Ni}}}{t} + \frac{B_{\text{surf,Cu-Ni}}}{t} \varepsilon_{xx} \right] \alpha_3^2 \]

For nanolines, two additional surfaces are added (Figure 12) – the two sides of the nickel nanolines. Assuming that these side surfaces oxidize after patterning, this will add Ni-NiO interfaces separated by a distance \( w \).
These surfaces also alter the magnetocrystalline and magnetoelastic energy of the system, in an analogous manner:

\[ E_{\text{surface},y} = \left[ \frac{K_{\text{surf}, \text{Ni-NiO}}}{w} + \frac{B_{\text{surf}, \text{Ni-NiO}}}{w} \varepsilon_{xx} \right] \alpha_z^2 \]

These energy terms are expected to be much smaller in magnitude, given that \( w \gg t_{\text{Ni}} \), but further add to the system anisotropy. Additionally, due to the lowered symmetry and changes to the strain state in the nanolines, the \( B_s \) terms for the nanolines may not be equal to those for the continuous thin films. This is discussed further in Appendix 2.

### 2.4 Demagnetization Factors

The magnetostatic energy of a continuous thin film is calculated using the demagnetization factor for perpendicular magnetization of a thin sheet, which is simply \(-4\pi\). In a system with more complicated shapes, calculation of the magnetostatic energy requires evaluation of the demagnetization factors.

For a line with thickness \( t \) and width \( w \), a point in the interior at \((y, z)\) is subject to demagnetization fields from the top and bottom surfaces and from the side surfaces. These are calculated using the angles shown in Figure 13.
Figure 13: Line cross-section illustration for demagnetization factor calculations

where the angles $\theta$ are given by the relations:

$$
\theta_{11} = \arctan \frac{z}{y}, \quad \theta_{12} = \arctan \frac{z}{w-y}, \quad \theta_{21} = \arctan \frac{t-z}{y}, \quad \theta_{22} = \arctan \frac{t-z}{w-y}
$$

The demagnetization field at a given point due to a charged\textsuperscript{6} surface of infinite extent in one direction has two components, one perpendicular to the surface and one parallel to the surface, given by:

$$
H_\perp = \frac{\Sigma}{2\pi} \theta \quad \text{and} \quad H_\parallel = \frac{\Sigma}{2\pi} \ln \frac{r_2}{r_1}
$$

where $\Sigma$ is the concentration of magnetic dipoles per unit volume at the surface, $\theta$ is the angle subtended by the surface from the point of interest, and $r_1$ and $r_2$ are the distances from the point to the left and right edges of the surface. To estimate the demagnetization factor, the perpendicular demagnetization field, $H_\perp$, is averaged over the thickness and width of the nanoline cross-section. Details are given in Appendix 3.

### Table 3: Demagnetization Factors for Nanolines

<table>
<thead>
<tr>
<th>t_{Ni} (Å)</th>
<th>w (nm)</th>
<th>$N_Z/4\pi$</th>
<th>$N_Y/4\pi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>69</td>
<td>250</td>
<td>0.955</td>
<td>0.045</td>
</tr>
<tr>
<td>69</td>
<td>100</td>
<td>0.907</td>
<td>0.093</td>
</tr>
<tr>
<td>206</td>
<td>250</td>
<td>0.895</td>
<td>0.105</td>
</tr>
<tr>
<td>206</td>
<td>100</td>
<td>0.798</td>
<td>0.202</td>
</tr>
</tbody>
</table>

2.5 Second Order Magnetoelastic Energy

In the uniaxial anisotropy model developed by Ha et al, the second order magnetoelastic energy was simply modelled as:

$$
E_{ME,2} = D_{eff} \varepsilon_o^2
$$

\textsuperscript{6} “Charged” refers to surface magnetic dipoles, not electrical charge.
where it was understood that $D_{\text{Eff}}$ encompassed a more complicated expression with one numerical value. The full second order magnetoelastic expression, developed by du Trémolet de Lacheisserie [33] and expanded by Komelj and Fähnle [34] is:

$$E_{\text{ME,2}} = \frac{1}{2} \left( B_1 + m^{t,2}_1 \right) \left[ e_1^2 \alpha_1^2 + e_2^2 \alpha_2^2 + e_3^2 \alpha_3^2 \right] + \frac{1}{2} m^{t,2}_2 \left[ e_1 e_2 \alpha_3^2 + e_1 e_3 \alpha_2^2 + e_3 e_2 \alpha_1^2 \right]$$

$$+ m^{t,2}_1 \left[ e_1 e_3 \alpha_2 \alpha_3 + e_2 e_3 \alpha_1 \alpha_3 + e_3 e_1 \alpha_2 \alpha_3 \right] + \frac{1}{2} \left( m^{t,2}_3 - B_1 \right) \left[ e_2^2 \alpha_1^2 + e_3^2 \alpha_2^2 + e_1^2 \alpha_3^2 \right]$$

$$+ \left( B_2 + m^{t,2}_2 \right) \left[ (\varepsilon_{11} + \varepsilon_{22}) e_{1,2} \alpha_1 \alpha_2 + (\varepsilon_{11} + \varepsilon_{33}) e_{1,3} \alpha_1 \alpha_3 + (\varepsilon_{33} + \varepsilon_{22}) e_{2,3} \alpha_2 \alpha_3 \right]$$

$$+ \left( B_2 + m^{t,2}_3 \right) \left[ e_{1,2} e_{2,3} \alpha_1 \alpha_3 + e_{1,3} e_{2,3} \alpha_2 \alpha_3 + e_{1,3} e_{3,2} \alpha_1 \alpha_2 \right]$$

Removing the (zero value) shear strain terms gives:

$$E_{\text{ME,2}} = \frac{1}{2} \left( B_1 + m^{t,2}_1 \right) \left[ e_1^2 \alpha_1^2 + e_2^2 \alpha_2^2 + e_3^2 \alpha_3^2 \right] + \frac{1}{2} m^{t,2}_2 \left[ e_1 e_2 \alpha_3^2 + e_1 e_3 \alpha_2^2 + e_3 e_2 \alpha_1^2 \right]$$

In other words, the $D_{\text{Eff}}$ represented in the uniaxial equation, above, is:

$$D_{\text{Eff}} = \left[ \frac{1}{2} \left( B_1 + m^{t,2}_1 \right) \left[ 1 - \frac{e_{22}^2}{e_{xx}^2} \right] + \frac{1}{2} m^{t,2}_2 \left[ \frac{e_{12} e_{23} - e_{13} e_{22}}{e_{xx}} \right] \right]$$

A value of $D_{\text{Eff}}$ for the $xz$ plane will not be equivalent to that of the $xy$ or $yz$ plane. Once the strains are changed by patterning into nanolines, the more general equation for $D_{\text{Eff}}$ is needed for accurate calculation of the second order magnetoelastic energy.

### 2.6 Triaxial Anisotropy Model

Combining the magnetostatic, magnetocrystalline (bulk and surface), magnetoelastic (first order bulk, first order surface and second order bulk) for the nanoline system gives the following expression for the overall energy:

$$E_{\text{overall}} = E_0 + \frac{\mu_0}{2} N_z M_s^2 \alpha_3^2 + \frac{\mu_0}{2} N_s M_s^2 \alpha_2^2 + K_1 \left[ \alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2 \right] + K_2 \left[ \alpha_1^2 \alpha_2^2 \alpha_3^2 \right]$$

$$- \frac{2K_{\text{sCa-Ni}}} {t} \alpha_2^2 \frac{2K_{\text{sNi-NiO}}} {w} \alpha_2^2 + B_1 \left[ \alpha_1^2 \varepsilon_{11} + \alpha_2^2 \varepsilon_{22} + \alpha_3^2 \varepsilon_{33} \right] - \frac{2B_{\text{sNi-NiO}}}{t} \varepsilon_0 \alpha_3^2 - \frac{2B_{\text{sNi-NiO}}}{w} \varepsilon_0 \alpha_2^2$$

$$+ \frac{1}{2} \left( B_1 + m^{t,2}_1 \right) \left[ e_1^2 \alpha_1^2 + e_2^2 \alpha_2^2 + e_3^2 \alpha_3^2 \right] + \frac{1}{2} m^{t,2}_2 \left[ e_1 e_2 \alpha_3^2 + e_1 e_3 \alpha_2^2 + e_3 e_2 \alpha_1^2 \right]$$

where all shear terms were removed, and angle-invariant terms were consolidated into $E_0$. 

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The magnetostatic term has now been split into two components, due to the nonzero $N_y$ demagnetization term. The side surfaces introduce magnetocrystalline and magnetoelastic surface terms, which should be small, $w$ usually being large. Finally, the changed strain state necessitates a more general expression for $D_{\text{eff}}$ and, when the strain relief is sufficiently large, general expressions for the $B_s$ terms.

2.7 Summary

Reexamination of the uniaxial anisotropy model using new anisotropy data and second-order magnetoelastic energy constants gives more accurate values for the magnetocrystalline surface anisotropy constant, $K_s$, and the magnetoelastic surface anisotropy constant, $B_s$. Patterning of the continuous Cu/Ni/Cu thin film system into nanolines will lower the symmetry from tetragonal to orthorhombic by introducing new surfaces (the nanoline sides) and changing the out-of-plane and transverse strain states. Due to the reduction in symmetry, more general expressions are needed for the magnetostatic and the surface and second-order magnetoelastic energy terms. The new demagnetization values are calculated. Finally, the triaxial anisotropy model is presented to describe the Cu/Ni/Cu nanoline energy.
Chapter 3

Strain Simulation by by Object-Oriented Finite Element Analysis (OOF)

This chapter presents simulation of the Cu-Ni-Cu nanoline strain state via Object-Oriented Finite Element Analysis (OOF).

3.1 Description and Procedures

In order to better understand the strain state within the nanolines, OOF 1.1.20 (in conjunction with PPM2OOF\(^7\) 1.1.28) was used to model the strain state of nanolines in cross-section. OOF (Object-Oriented Finite Element Analysis) is a program developed at NIST and available to the public. It is designed to calculate materials properties (stress and strain) using images of real or simulated microstructures.\(^8\)

To model the strain state of the nanolines, a simple, 2-dimensional representation of the nanolines’ cross-sections was created. These 2-dimensional representations are accurate for almost the entire length of the nanolines, though of course at the nanoline ends, they no longer represent the true state of the sample. This end volume represents a negligibly small volume of the material, so is not treated in this model.

Images of each sample to be modeled were created using rectangular blocks scaled to the appropriate size. PPM2OOF 1.1.28 was used to convert the images for use in OOF. Each rectangular block was assigned appropriate materials properties, and a dense triangular mesh was created.

The boundary conditions in OOF were set such that there could be no displacement, either vertical or horizontal (\(y\) and \(x\), respectively), on the bottom of the structure (representing the interface with the Cu “substrate”). A temperature (chosen so that the resulting thermal expansion strain would be equal to the equivalent Cu-Ni lattice misfit strain) was applied to the system and then the system was equilibrated. From there, the stress and strain statistics were

\(^7\)“ppm to OOF”, a program designed to transform ppm graphics files for use in the OOF program.
\(^8\)http://www.ctcms.nist.gov/oof/
computed and saved. Images of the stress and strain, both $xx$ and $yy$, were generated. These images could not be saved to disk, but screen captures were performed.

It is important to understand that the strain values calculated in OOF do not correspond to the strain in the nanolines, rather to the strain relaxation in the nanolines. This is because the simulation models the nanoline system moving from a fully strained state to a partially relaxed state. Hence, an element in the simulations with a “strain” (really strain relaxation) value of -0.026 corresponds to, in an actual sample (with a base strain of 0.026), a region with a strain of 0. To convert from the strain relaxation values calculated in OOF to the strain state in the nanolines, one simply adds to the OOF values the base strain state of the corresponding continuous thin film. In this chapter, the term “strain” will be used to refer to the values of strain relaxation calculated in OOF, not the absolute strain state of the nanolines, unless stated otherwise.

### 3.2 Strain Simulation in Rectangular Blocks of Varying Aspect Ratio

To quantify the strain effect in the line cross-section caused by changes to the aspect ratio, images were created of simple rectangular blocks, with a variety of aspect ratios (height : width, aka film thickness : nanoline width) of 1:1, 1:1.5, 1:2, 1:3, 1:4, 1:5, 1:7.5, 1:10, 1:15 and 1:20.

The materials properties of these structures were chosen to be the same as the nickel properties as used for the Cu/Ni/Cu films simulated later. A simple mesh of approximately 2500 elements was created, with the number of elements in the $x$ and $y$ directions chosen to give a uniform density of elements. In other words, the 1:1 block had 50 x 50 elements, the 1:4 block had 25 x 100 elements, etc.

![Image of 1:2 Structure](image)

**Figure 14:** Image of 1:2 Structure, showing coordinate axes and fixed (zero-displacement) edge (shaded)
In OOF, the temperature change was chosen to be -1°C, so that the strain would be, at maximum, -0.026, to match the maximum nickel lattice misfit strain. The generated images of strain ($\varepsilon_{xx}$) relaxation are pictured in the following three figures:

![Figure 15: Strain ($\varepsilon_{xx}$) map, aspect ratio 1:1](image1)

![Figure 16: Strain ($\varepsilon_{xx}$) map, aspect ratio 1:5](image2)

![Figure 17: Strain ($\varepsilon_{xx}$) map, aspect ratio 1:20, with enlarged detail section of left end](image3)

Each image is plotted to the same scale, with white indicating the pinned (strain relief = 0) areas and black indicating images of maximum strain relief ($\varepsilon_{xx} = -0.026$), and each color gradation representing a 0.002 strain change. As the images indicate, increasing the aspect ratio results in a greater “pinning” effect from the bottom surface. At high aspect ratios, the interior of the structure is almost completely pinned: very little strain relaxation is permitted. Only towards the edges (within about 1 ½ thickness units of the edge) is strain relief observed. This effect is quantified in Table 4, which shows a monotonic decrease in the average strain with increasing
aspect ratio. Unrelaxed areas ($\varepsilon_{xx}=0$) represent regions at the maximum strain for continuous thin films of nickel at the given thickness.

**Table 4: Strain Relaxation (Average and Standard Deviation) and Aspect Ratios**

<table>
<thead>
<tr>
<th>Aspect Ratio</th>
<th>Average $\Delta \varepsilon_{xx}$</th>
<th>StDev($\Delta \varepsilon_{xx}$)</th>
<th>Average $\varepsilon_{xx}$</th>
<th>% Strain Relaxed$^9$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>-0.0224</td>
<td>0.0066</td>
<td>0.0006</td>
<td>86.2</td>
</tr>
<tr>
<td>1.5</td>
<td>-0.0207</td>
<td>0.0076</td>
<td>0.0053</td>
<td>79.6</td>
</tr>
<tr>
<td>2.0</td>
<td>-0.0190</td>
<td>0.0081</td>
<td>0.0070</td>
<td>73.1</td>
</tr>
<tr>
<td>3.0</td>
<td>-0.0155</td>
<td>0.0080</td>
<td>0.0105</td>
<td>59.7</td>
</tr>
<tr>
<td>4.0</td>
<td>-0.0126</td>
<td>0.0079</td>
<td>0.0134</td>
<td>48.3</td>
</tr>
<tr>
<td>5.0</td>
<td>-0.0103</td>
<td>0.0079</td>
<td>0.0157</td>
<td>39.7</td>
</tr>
<tr>
<td>7.5</td>
<td>-0.0069</td>
<td>0.0077</td>
<td>0.0191</td>
<td>26.4</td>
</tr>
<tr>
<td>10.0</td>
<td>-0.0051</td>
<td>0.0072</td>
<td>0.0209</td>
<td>19.5</td>
</tr>
<tr>
<td>15.0</td>
<td>-0.0034</td>
<td>0.0063</td>
<td>0.0226</td>
<td>12.9</td>
</tr>
<tr>
<td>20.0</td>
<td>-0.0024</td>
<td>0.0055</td>
<td></td>
<td>9.1</td>
</tr>
</tbody>
</table>

These values are plotted graphically, below, in Figure 18.

![Figure 18: Average Strain vs. Aspect Ratio (width/thickness)](image)

These simple simulations shed light on the effect of aspect ratio on the strain state, but the Cu-Ni-Cu system has three additional complications: 1) different thicknesses of nickel have different base strain states, as dislocation density increases with increasing thickness, further decreasing the average strain; 2) the presence of the copper capping layer, which will serve as a restraint on the contraction of the nickel layer, to some extent; and 3) the possibility of overetch or underetch relative to misfit strain = 0.026.
of the nanolines, which will change either the effective nickel thickness (underetch) or act to separate the nickel from the substrate pinning (overetch).

3.3 Strain Simulation for Cu/Ni/Cu Films

The Cu-Ni-Cu samples were created using blocks of appropriate size and differing colors for the nickel and copper. The copper capping layer was of a uniform height of 5.0 nm. The 12 configurations modeled are shown in Table 5:

Table 5: Cu/Ni/Cu Simulation Configurations

<table>
<thead>
<tr>
<th>Nanoline Width (nm)</th>
<th>Ni Thickness (regular), nm</th>
<th>Ni Thickness (underetch), nm</th>
<th>Cu substrate overetch, nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>6.9</td>
<td>6.2</td>
<td>0.7</td>
</tr>
<tr>
<td>250</td>
<td>6.9</td>
<td>6.2</td>
<td>0.7</td>
</tr>
<tr>
<td>100</td>
<td>20.6</td>
<td>18.5</td>
<td>2.1</td>
</tr>
<tr>
<td>250</td>
<td>20.6</td>
<td>18.5</td>
<td>2.1</td>
</tr>
</tbody>
</table>

In the regular and underetch conditions, the only blocks created were the nickel and the copper capping layer blocks (Figure 19 - regular case). The nickel thickness was the nominal thickness of 6.9 or 20.6 nm in the regular case, and 90% of the nominal thickness in the underetch case.

Figure 19: Image of 20.6 nm nickel (blue), 100 nm wide, regular etch, showing copper substrate underneath (not included in model)

In the overetch condition, an additional block of copper is inserted below the nickel, representing an overetch of 10% of the nickel thickness into the copper below.
Figure 20: Image of overetched 20.6 nm nickel, 100 nm wide, showing copper substrate underneath (not included in model)

In OOF, the 6.9 nm cases were subjected to a temperature change of -0.535°, and the 20.6 nm cases were given a temperature change of -0.258°, corresponding to maximum strains of 0.0139 and 0.0067, respectively, in keeping with the measured strains for those nickel film thicknesses, as continuous thin films under biaxial strain.

Two strain images comparing strain distributions for regular vs. overetch cases are shown in Figure 21 and Figure 22, where each color step represents a strain change of 0.0005.

Figure 21: 20.6 nm Ni, regular etch case, showing mesh (top) and strain map (bottom)
Figure 22: 20.6 nm Ni, overetch case, showing mesh (top) and strain map (bottom)

At the opposite extreme lies the 6.9 nm Ni, 250 nm linewidth sample, in Figure 23, where each color step represents a strain change of 0.001.

Figure 23: 6.9 nm Ni, regular case, showing mesh (top), strain map (middle), and enlarged view of left corner (bottom)

The same “pinning” effects (preventing strain relaxation) as were seen in the aspect ratio simulations are observed in the images of the Cu-Ni-Cu layers, with the greatest amount seen in the wider (250 nm) lines, and the thinner Ni films. The thicker Ni film shows pervasive strain relaxation penetrating deep into the interior of the narrower width (100 nm) line. Though the underetch effect is subtle, the addition of the thin layer of Cu underneath the Ni in the overetch cases is significant. Each overetch case shows added strain relaxation compared to the regular or underetch cases. This effect is quantified in Table 6, below:
Table 6: Strains for Cu-Ni-Cu Cases

<table>
<thead>
<tr>
<th>(nom.) $t_{Ni}$ nm</th>
<th>Width, nm</th>
<th>Etch cond.</th>
<th>Aspect Ratio (1:X)</th>
<th>Average $\Delta \varepsilon_{xx}$</th>
<th>StDev $(\Delta \varepsilon_{xx})$</th>
<th>Average $\varepsilon_{xx}$</th>
<th>% strain relax$^{10}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.9</td>
<td>100</td>
<td>reg</td>
<td>14.5</td>
<td>-0.0015</td>
<td>0.0027</td>
<td>0.0124</td>
<td>10.7</td>
</tr>
<tr>
<td>6.9</td>
<td>100</td>
<td>over</td>
<td>14.5</td>
<td>-0.0021</td>
<td>0.0030</td>
<td>0.0118</td>
<td>15.4</td>
</tr>
<tr>
<td>6.9</td>
<td>100</td>
<td>under</td>
<td>16.1</td>
<td>-0.0013</td>
<td>0.0026</td>
<td>0.0126</td>
<td>9.6</td>
</tr>
<tr>
<td>6.9</td>
<td>250</td>
<td>reg</td>
<td>36.2</td>
<td>-0.0005</td>
<td>0.0017</td>
<td>0.0134</td>
<td>3.8</td>
</tr>
<tr>
<td>6.9</td>
<td>250</td>
<td>over</td>
<td>36.2</td>
<td>-0.0008</td>
<td>0.0021</td>
<td>0.0131</td>
<td>5.7</td>
</tr>
<tr>
<td>6.9</td>
<td>250</td>
<td>under</td>
<td>40.3</td>
<td>-0.0005</td>
<td>0.0017</td>
<td>0.0134</td>
<td>3.5</td>
</tr>
<tr>
<td>20.6</td>
<td>100</td>
<td>reg</td>
<td>4.9</td>
<td>-0.0025</td>
<td>0.0018</td>
<td>0.0042</td>
<td>36.7</td>
</tr>
<tr>
<td>20.6</td>
<td>100</td>
<td>over</td>
<td>4.9</td>
<td>-0.0029</td>
<td>0.0017</td>
<td>0.0038</td>
<td>43.0</td>
</tr>
<tr>
<td>20.6</td>
<td>100</td>
<td>under</td>
<td>5.4</td>
<td>-0.0022</td>
<td>0.0018</td>
<td>0.0045</td>
<td>32.9</td>
</tr>
<tr>
<td>20.6</td>
<td>250</td>
<td>reg</td>
<td>12.1</td>
<td>-0.0010</td>
<td>0.0015</td>
<td>0.0057</td>
<td>14.8</td>
</tr>
<tr>
<td>20.6</td>
<td>250</td>
<td>over</td>
<td>12.1</td>
<td>-0.0013</td>
<td>0.0016</td>
<td>0.0054</td>
<td>19.9</td>
</tr>
<tr>
<td>20.6</td>
<td>250</td>
<td>under</td>
<td>13.5</td>
<td>-0.0009</td>
<td>0.0014</td>
<td>0.0058</td>
<td>12.8</td>
</tr>
</tbody>
</table>

As can be seen in Table 6, the Ni film strain is largely a function of aspect ratio, with values closely tracking the results shown in Table 4, for strain as a function of aspect ratio. The close dependance on aspect ratio explains the small differences between the regular and underetch cases. The case of overetched Ni is much more dramatic. An overetch of a mere 10% increases the nickel’s strain relaxation by as much as 5% of the target value. Small variations in processing at the ion mill stage thus could have a large impact on the properties of the finished nanolines.

### 3.4 OOF Modeling Limitations

Using thermal expansion to simulate biaxial strain is not optimal, as thermal expansion is three-dimensional, which will result in the sample contracting both in-plane and out of plane. This means that the out-of-plane strain components of the simulation are incorrect, because they show the effects of a strain of -0.026, instead of the Poisson effect strain from the in-plane biaxial strain of 0.026. It is unlikely that the out-of-plane inaccuracies affect the in-plane values to any significant degree, or, if there is any affect, this modeling at least captures the important aspects of the in-plane strain effects in an internally consistent manner. The absolute values of each configuration’s stress/strain distribution may be slightly incorrect, but it will still be a useful

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$^{10}$ Relative to the misfit strain for a given nickel thickness: 0.0139 for 6.9 nm, 0.0067 for 20.6 nm.
way to compare the relative effects of different aspect ratios and configurations. Other modeling programs may be better able to capture biaxial strain distributions.

Another major limitation of this modeling is the choice of rectangles to model the structures. Ion milling may result in erosion of the copper capping layer, making its optimal shape more likely to be a trapezoid. Since the effect of ion milling on this layer is not quantified, it would be difficult to determine the dimensions of such a trapezoid. Also, the surfaces are not perfectly flat, possessing some inherent roughness. This, too, is difficult to model accurately, given the lack of specific information about the roughness of each interface.

3.5 Summary

In this section, the strain state of the Cu/Ni/Cu nanolines has been modeled. Strain relaxation is largely a function of the nanoline cross section’s aspect ratio, so thicker films and narrower linewidths show large strain relaxation, in contrast with thinner films and wider linewidths, which show very little strain relaxation. In these cases (thin films, wide linewidths), the outer 1 ½ thickness units relax their strain, while the interior is pinned (zero strain relaxation) by the substrate-film interface. Underetching by 10% of the nickel thickness does not significantly impact the degree of strain relaxation, but overetching by 10% will result in a significant increase in strain relaxation.
Chapter 4
Fabrication of Cu/Ni/Cu/Si (100) Thin Films and Nanolines

4.1 Fabrication of Cu/Ni/Cu/Si (100) Thin Films

The growth of Cu/Ni/Cu/Si (100) epitaxial thin films has been documented by previous researchers [35] and is reviewed here. The growth of Cu/Ni/Cu/Si (100) polycrystalline thin films by ion beam sputter deposition has not been previously documented in detail.

a. Fabrication of Epitaxial Cu/Ni/Cu/Si (100) Thin Films

Although Cu (001) and Si (001) have widely different lattice parameters (3.61 Å vs. 5.43 Å, respectively, giving a mismatch of ~40%) [36,37], rotation of the Cu lattice 45° in-plane (so that the Cu [110] is parallel to the Si [100] and vice-versa) results in a more closely matched lattice (Cu [110]: 5.105 Å, a mismatch of ~6%), making heteroepitaxial growth possible. Observations of Cu/Si (001) growth have shown that the Cu, grown at room temperature, forms a diffuse interface (~100 Å thick) with the Si due to rapid interdiffusion and energetically favorable compound formation. Beyond the interface, the copper is epitaxial, with a (001) surface.

Molecular beam epitaxy is a valuable technique used for growing specialized epitaxial films. Using evaporation by effusion cells or electron beams in an ultra-high vacuum, MBE is capable of growing films at the slow rates needed to ensure that arriving atoms find their energetically favored positions on the substrate lattice. Thus, high purity films of good crystalline quality and sharp interfaces can be grown. [38,39] Electron-beam evaporation makes use of a water-cooled crucible containing the desired metal, heated by an incoming beam of electrons from a tungsten filament (Figure 24). Typically the filament is located adjacent to the crucible, with the electron beam bent by magnetic fields to impinge upon the crucible contents, thus preventing evaporation of metal onto the filament itself.
Films of Cu (50 Å) / Ni (t_{Ni}) / Cu (2000 Å) / Si(001), where t_{Ni} = 57, 69, 92, 103, 115, 138, 161, 172, 206, 229 and 1835 Å, were grown using a Perkin-Elmer molecular beam epitaxy tool (Figure 25).

Prior to loading the silicon wafers into the chamber, the silicon wafers were immersed in a 5% solution of HF for 10 seconds to remove native oxide at the surface, then were rinsed with deionized water for 2 minutes, dried with nitrogen gas and immediately inserted into the chamber, to prevent oxidation. The base pressure was \( \sim 2 \times 10^{-10} \) Torr, with chamber pressure rising to \( 2 \times 10^{-8} \) Torr and \( 1.2 \times 10^{-9} \) Torr during deposition of Cu and Ni, respectively. Depositions occurred at room temperature, with substrate temperatures rising to as much as 40°C during...
depositions. The deposition rate for Ni was 0.8 Å/sec; for Cu, it was 1.15 Å/sec. Deposition rates were monitored in situ using a quartz crystal, and ex situ using a calibration sample measured by a step height profilometer. To ensure uniform film thickness, the substrate was rotated 45° every 100 Å during deposition of the thick copper, while during deposition of the thin copper and nickel films, the substrate was rotated continuously. Reflection high energy electron diffraction spectroscopy (RHEED) shows a 1x1 streak pattern for the thick copper films, indicating successful epitaxial growth of the Cu, in agreement with previous results [40].

**b. Fabrication of Polycrystalline Cu/Ni/Cu/Si (100) Thin Films**

In order to compare the properties of strained Cu/Ni/Cu thin films with unstrained thin films of similar thicknesses, polycrystalline films of Cu (2000 Å) / Ni (t) / Cu (50 Å), with t_{Ni} = 69 Å and 206 Å were grown. These films would have the same composition and interface characteristics of their epitaxial counterparts, but without epitaxy, there would be little to no film strain, allowing isolation of the strain contribution. These polycrystalline films were grown in a custom-built ultrahigh vacuum deposition chamber (Superior Vacuum Technology #4000176, see Figure 26), using triode / DC magnetron sputtering (LM Simard PD-250) to grow the copper and ion beam sputtering (Commonwealth Scientific 3” ion source) to grow the nickel films.

![Figure 26: UHV Sputter Deposition Tool](image)

Both DC magnetron and ion beam sputtering use argon ions to sputter material from the target, but there are crucial differences (Figure 27). Both sputter techniques, because of the broad area
of the target (unlike the point-type source from evaporation), create a variety of entry angles at the substrate, resulting in good planarity, though step coverage is poor at the low pressures used in this UHV system. The high energy of incoming atoms and high rate of resputtering often precludes placement of atoms at their energetically favored sites, making epitaxy unlikely. Due to the large size of the target relative to the substrates (3”), no rotation was necessary for film thickness uniformity, although in production, with larger substrates, rotation would be used.

Figure 27: Schematic of DC triode magnetron sputtering (left) and ion beam sputtering (right), after Plummer (39) and Smith (37), respectively (not to scale)

In DC magnetron sputtering, the target and substrate are placed opposite each other. Argon gas forms a plasma. The positive argon ions strike the negatively charged target, physically sputtering material. Adding a magnetron increases the process efficiency, as the electrons in the plasma are given spiral trajectories due to the magnetic field. The spiral trajectories increase the probability of collision with argon gas molecules, increasing the plasma efficiency and decreasing the power necessary, which, in turn decreases the heat dissipated into the substrate. A triode adds the extra feature of having the plasma created using an extra filament and anode, which results in a more uniform, near-cylindrical, flux of atoms from the target. DC magnetron sputtering in general is popular due to its excellent step coverage, moderately high growth rates and relatively low contamination. [39, 41]

Ion beam sputtering, by contrast, is a lesser used technique, though often favored by researchers. This is because the flux of ions from a separate ion gun striking the target is controlled by the cathode current, and the ion energy is set by the velocity imparted by the ion beam’s acceleration grid (or the bias between gun and the target), making the ion flux and energy
parameters separable, unlike a typical sputter process, in which the two parameters would be coupled. Ion beam sputtering uses a lower argon pressure, because it does not need to form a plasma, and because sputtered material has farther to travel between target and substrate (due to the geometry of the system), so needs a longer mean free path. Ion beam sputtering has a lower growth rate, and the apparatus is complicated, so is not a popular manufacturing technique. [37]

Before loading the samples into the chamber, the silicon wafers were blown with nitrogen gas to remove any particulates. The base pressure of the system was approximately $2 \times 10^{-8}$ Torr. For DC magnetron sputtering of the copper, argon plasma at a pressure of 1 mTorr was used for growth rates of $2.1 \, \text{Å/s}$. For ion beam sputtering of the nickel, argon at $3.8 \times 10^{-5}$ Torr was used for growth rates of $0.24 \, \text{Å/s}$.

Additional deposition parameters are in Appendix 5.

### 4.2 Fabrication of Cu/Ni/Cu/Si (100) Nanolines

Once the films were grown, subtractive lithographic patterning was done using a Lloyd’s mirror lithography tool to create 200 and 500 nm period nanolines of Ni ($t$) / Cu (50 Å), with $t_{Ni} = 69 \, \text{Å}$ and 206 Å on Cu (2000 Å)/Si (100).

**a. Interferometric Lithography**

Interferometric lithography is a maskless lithography technique used to pattern simple, periodic elements such as dots, ellipses or lines over a large area. By intersecting two coherent beams, a standing wave is generated in a photoresist layer, forming an array of parallel lines after development. The line period of the array is related to the wavelength of the light ($\lambda$) and the angle between the two beams, $\theta$, by: [42]

$$p = \frac{\lambda}{2\sin\left(\frac{\theta}{2}\right)}$$

The two beams can be generated in one of two ways: a beam splitter can split a laser beam, which is then recombinated at an angle to the sample surface, or a laser beam can impact on both a sample and a mirror, known as a Lloyd’s mirror, invented by Lloyd in 1837 [43]. Unlike a traditional interferometer, a Lloyd’s mirror is a very simple structure, and is resistant to vibrations in the system, as the mirror and substrate are physically joined. The only disadvantages of the Lloyd’s mirror are that the mirror must be perfectly flat, any dust particles will act as sources of diffuse scattering, causing patterning defects, and the contrast at the sample
will never be perfectly sharp, as the intensity of the reflected beam will be slightly smaller than the intensity of the incoming beam. These disadvantages can be overcome by using a flat mirror, blown clear of particles, made of a material highly reflective of the incoming light. Adjustments of the line period only require adjustment of either the mirror-substrate angle, or, keeping the mirror-substrate angle fixed, adjusting the angle of the mirror-substrate array with respect to the incoming beam. The latter further stabilizes the mirror-substrate array from vibrations. [44]

**Figure 28: Lloyd's mirror schematic (after Walsh [44])**

The Lloyd’s mirror is a custom-built tool in the Nanostructures Laboratory (NSL) which uses a Kimmon IK3501R-G 52 mW HeCd laser emitting a 325 nm (UV) beam. The laser beam is reflected off two mirrors, then passed through a spatial filter (to remove any beam noise). The light travels ~1.7 m to the substrate-mirror assembly. This length causes the beam to spread out to approximately 36 cm in diameter. Though the intensity drops with the distance, the resulting dose is sufficient to expose photoresist in approximately ten minutes, which, given the system’s low sensitivity to vibrations, is feasible. The increase in diameter also permits patterning of large areas (4” wafers), and causes the beam radius to increase to the point where the incoming beam is effectively a plane wave. The mirror-substrate angle is fixed; the line period is controlled by adjusting the angle of the mirror-substrate assembly at the base. The mirror is made of aluminum, which is an efficient reflector of 325 nm light at a wide range of angles. [44]
b. Trilayer Stack

At its simplest, subtractive patterning would only need photoresist spun onto the surface of the material to be etched. Three factors preclude the use of such a simple procedure: the inability to etch most magnetic thin films to submicron features using photoresist as a mask, the need to prevent reflections from the substrate surface and the need for a hard mask to protect the magnetic films against an ion milling procedure (discussed in greater detail later). A tungsten film is used as the hard mask. Because tungsten is not photoreactive, other films are needed to transfer the line pattern into the tungsten. And since photoresist and tungsten both etch in oxygen, an intermediate material - SiO₂ - is needed to transfer the pattern into the W.

Reflection prevention is done by using a trilayer stack – composed of antireflective coating (ARC), SiO₂ and photoresist. Trilayer stacks, when composed of films of the correct thickness, prevent standing waves from forming in the photoresist when exposed to the UV light used to pattern the films. These standing waves would, at minimum, cause “scalloping” – vertical waves - in the patterned photoresist, but could, if sufficiently severe, narrow the patterned photoresist, causing the structures to collapse. [45] Dr. Michael Walsh developed an algorithm to calculate optical impedances of a stack of multiple films and plot the reflectivity as a function of any unknown parameter (such as the thickness of an ARC layer, for example). [44] Parameters used for stack calculations are listed in Appendix #O.

c. Lithographic Processing

Evaporation was used to deposit W (t = 30 Å) and SiO₂ (t = 30 Å) films at the NSL (Nanostructures Laboratory). Clariant BARLi antireflective coating (ARC) was then spun onto
the surface at a revolution speed chosen to ensure the desired ARC film thickness of 2200 Å, and then the top layer of SiO$_2$ was evaporated, to a thickness of 30 Å.

A thin layer of NMP (n-methyl 2-pyrrolidone) was poured onto the wafer and allowed to stand for 1 minute, then was spun off. This layer of NMP acts to passivate the surface, promoting adhesion of photoresist. Following the NMP treatment, the photoresist, Sumitomo PFI-88, was deposited, and spun for 1 minute at a revolution speed chosen to form a 2000 Å thick layer. This photoresist was softbaked at 90° C for 1 minute to remove excess solvents and prepare it for exposure. [Figure 30] [This temperature is less than the recommended temperature for that photoresist, due to the need to minimize the heat exposure to the sample. This did not adversely affect the processing.]

![Figure 30: Schematic of Cu/Ni/Cu patterning layers, pre-lithography](image)

Exposure was done in the Lloyd’s mirror tool, as discussed above, with angles adjusted to give line periods of either 200 nm or 500 nm. Exposure times ranged from 6-12 minutes. Following exposure, the wafers were developed in Rohm & Haas CD26 developer solution for 1 minute to remove the unexposed photoresist. Typically, line width was ~40-50% of the period, with line width influenced by processing parameters, including exposure dose and etching. [Figure 31]

![Figure 31: SEM of post-exposure sample (left) and schematic of Cu/Ni/Cu patterning layers, post-exposure (right) polycrystalline 206 Å Ni, 500 nm line period](image)
d. Reactive Ion Etching

Following exposure and development, the patterned wafers were processed in a reactive ion etcher (Plasma-Therm 790) to transfer the pattern from the photoresist into the underlying layers. A reactive ion etcher is a parallel plate plasma etching system (Figure 32), using plasma to enhance chemical reactions which are chosen to preferentially etch a desired species. The technique, popular in industry, is a synergistic mixture of both physical (ions accelerated at a target) and chemical etching techniques. The result is an etch with good selectivity and anisotropy, assuming there exists a reactant byproduct that is volatile at room temperature. [39]

![Figure 32: Schematic of reactive ion etcher (after Plummer [39])](image)

The steps used in processing were:

- Etch with CHF$_3$ to transfer the pattern into the top layer of SiO$_2$ [Figure 33]
- Etch with O$_2$ to transfer the pattern into the ARC (and remove the surface photoresist) [Figure 34]
- Etch with CHF$_3$ to transfer the pattern into the bottom layer of SiO$_2$ (and remove the top layer of SiO$_2$) [Figure 35]
- Etch with CF$_4$ and O$_2$ to transfer the pattern into the W layer (and remove the ARC). [Figure 36]
- Etch with CHF$_3$ to remove the remaining SiO$_2$. [Figure 36]

Etch recipes and parameters are given in Appendix # 5. Etch rates were calibrated by either ellipsometer measurements before and after etch (SiO$_2$ and ARC) or step height profilometer
measurements before and after etch (W) of calibration samples. Etch time of samples was adjusted for actual machine power output. Etch success was verified by SEM imaging.

Figure 33: SEM of post-RIE1 sample (left) and schematic of Cu/Ni/Cu patterning layers, post-RIE 1 (right), polycrystalline 206 Å Ni, 500 nm line period

Figure 34: SEM of post-RIE2 sample (left) and schematic of Cu/Ni/Cu patterning layers, post-RIE 2 (right), epitaxial 206 Å Ni, 500 nm line period

Figure 35: SEM of post-RIE3 sample (left) and schematic of Cu/Ni/Cu patterning layers, post-RIE 3 (right), polycrystalline 69Å Ni, 200 nm line period
Figure 36: SEM of post-RIE5 sample (left) and schematic of Cu/Ni/Cu patterning layers, post-RIE 4 & 5 (right, top and bottom), epitaxial 206 Å Ni, 200 nm line period

**e. Ion Milling**

Ideally, the magnetic materials would be patterned using a reactive ion etch, which could remove the desired material in a both selective (without etching the mask) and highly anisotropic (directional) manner. Unfortunately, there do not currently exist reactant gases whose products are volatile enough for use with typical magnetic materials, and wet chemistry processes are not well suited for submicron processing, due to their isotropic etch profiles. This means that a highly physical process is needed to remove magnetic materials. Ion milling – physical sputtering of the layers with charged ions – is used to abrade away the magnetic materials.

Due to its highly physical nature, however, ion milling is not very selective. This would logically suggest that any mask should therefore be thick, to minimize the probability of its wearing away due to faceting before the etch is complete. Unfortunately, material sputtered from a thick mask tends to land in the trenches between, further slowing the etch rate of the material and reducing selectivity to a minimum. Redeposition can also coat the sides of the mask, leading to undesirable “wings” at the end of the process. Therefore, a method was found to maximize the selectivity of the process using a thin hard mask. [46]

Selectivity of physical sputter processes is proportional to the energy transfer parameter,
\[ \gamma = \frac{4M_1M_2}{(M_1 + M_2)^2} \]

where \(M_1\) = mass of species 1 (etchant gas) and \(M_2\) = mass of species 2 (material to be etched).

This parameter is at its maximum when the two masses are equal, i.e. an incoming gas molecule will most efficiently transfer energy to an atom of comparable mass. So it can be seen
that for maximum selectivity, there should be a large difference in mass between gas and mask, and a small difference in mass between the gas and the material to be etched. This way, the etch rate for the material will dominate over the mask etch rate.

Tungsten, with its high atomic mass and easy process compatibility with reactive ion etching, was the ideal mask choice. And by using a low-mass noble gas, Ne, the mass difference can be increased, while still resulting in a high etch rate. This process resulted in great stability, allowing for complete etching without the risk of removing features. [46]

![Figure 37: Ion miller schematic (after Hao [45])]()

An ion miller is essentially a Kaufmann ion source, similar to the ion gun used in the ion beam sputter system, with a shutter to separate it from the water-cooled substrate [Figure 37]. Ion energy and flux are independently controlled. The cathode generates electrons, which ionize the gas. Magnets are used to improve the efficiency of ionization. Once the ions are generated, a high voltage grid accelerates them towards the substrate (though the beam is passed by the neutralizer). This is not an efficient process: the vast majority of the input energy is wasted in heat. To minimize the amount of sample heating, processing was done 30 seconds on and 30 seconds off, and the samples were mounted to the water-cooled sample block using Apiezon L (vacuum-safe thermal grease).

Etch parameters are given in Appendix # 5. Etch rates were calibrated for the CHA Industries/Ion-Tech, Inc ion miller using continuous thin films, measuring magnetic signal (in an Alternating Gradient Magnetometer [AGM], Princeton Measurement Corp. Micromag #2900) as a function of etch time. SEM images of the milled samples show some tungsten remaining on the copper surface [Figure 38, Figure 39].

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Figure 38: SEM of milled sample (left) and schematic of Cu/Ni/Cu patterning layers, post-ion mill (right), epitaxial 206 Å Ni, 500 nm line period

Figure 39: SEM image of epitaxial 206 Å Ni, 200 nm line period, post-ion mill

### 4.3 Summary

Epitaxial Cu/Ni/Cu/Si films with nickel thicknesses of 57, 69, 92, 103, 115, 138, 161, 172, 206, 229 and 1835 Å were grown. Polycrystalline Cu/Ni/Cu/Si films with nickel thicknesses of 69 and 206 Å were grown. Epitaxial and polycrystalline Cu/Ni/Cu/Si films with nickel thicknesses of 69 and 206 Å were patterned into Cu/Ni nanolines with line periods of 500 and 200 nm (Table 7). These nanolines had line widths approximately 40-50% of line periods, and had good line quality over a large area, as seen in SEM images.

#### Table 7: Summary of Nanoline Samples Made

<table>
<thead>
<tr>
<th>$t_{\text{Ni}}$ (Å)</th>
<th>Type</th>
<th>Line period (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>69</td>
<td>Epi</td>
<td>500</td>
</tr>
<tr>
<td>69</td>
<td>Epi</td>
<td>200</td>
</tr>
<tr>
<td>69</td>
<td>Poly</td>
<td>500</td>
</tr>
<tr>
<td>69</td>
<td>Poly</td>
<td>200</td>
</tr>
<tr>
<td>206</td>
<td>Epi</td>
<td>500</td>
</tr>
<tr>
<td>206</td>
<td>Epi</td>
<td>200</td>
</tr>
<tr>
<td>206</td>
<td>Poly</td>
<td>200</td>
</tr>
</tbody>
</table>
Chapter 5

Room Temperature Characterization

This chapter presents room temperature characterization results of the Cu/Ni/Cu nanolines and continuous thin films via vibrating sample magnetometry (VSM) and torque magnetometry. It also presents characterization results of the strain state in continuous Cu/Ni/Cu thin films.

5.1 Vibrating Sample Magnetometry

a. Description

A vibrating sample magnetometer is one of the most common tools used for measuring $M-H$ [magnetization vs. magnetic field] hysteresis loops. It consists of a computer-controlled electromagnet, a vibrator unit (often embedded within a rotation stage) and a set of detection (or pick-up) coils (Figure 40). The sample is mounted on a nonmagnetic (glass) sample holder. It is vibrated vertically at a low frequency (75 Hz) while a DC (static) magnetic field is applied using the electromagnet. The sample’s magnetic flux is detected by the pick-up coils as a voltage. Typical VSMs can measure magnetization as small as $10^{-5}$ emu in 0.01 Oe field increments. [47]

![Figure 40: VSM diagram (after Speliotis [47])](image)

A Digital Measurement Systems Model 1660 Vibrating Sample Magnetometer was used to measure all the Cu/Ni/Cu samples. The magnetic field was varied in a series of programmed steps, with emphasis on the +/- 2000 Oe region. Multiple readings were taken at each step to decrease sample noise. Background subtraction (mathematic removal of signal linear with magnetic field) was performed to remove the diamagnetic substrate and sample holder signals – signals which are negligible for higher volume samples, but are significant for thin films.
**b. Continuous Thin Film Data**

VSM $M$-$H$ loops are presented for the 69 and 206 Å epitaxial and polycrystalline continuous thin film samples. Each sample was measured both with the magnetic field applied in plane and normal to the film plane between ± 10 kOe, although the charts below are presented between ± 4 kOe. Each measurement point represents an average of 50 readings. Magnetization for each sample is normalized relative to the saturation magnetization ($M$ at 10 kOe).

Both the 69 and the 206 Å polycrystalline thin films display typical behavior for thin films – easy axes in-plane and hard axis out-of-plane. The out-of-plane loops for both thicknesss have no hysteresis, suggesting that magnetization reversal occurs by magnetization rotation. The 69 Å polycrystalline films (Figure 42) have smaller remanence in-plane and smaller saturation fields out-of-plane. This suggests the effect of surface energies, which follow a $1/t$ relation, stabilizing out-of-plane magnetization. The in-plane loop has significant hysteresis (but remanence only ~0.6 $M_s$). The 206 Å polycrystalline thin films (Figure 44) have a remanence of approximately 0.8 $M_s$. Compared to the 69 Å polycrystalline thin films, saturation of the out-of-plane loop occurs at a higher field, indicating that hard-axis magnetization requires more energy.

The 69 and 206 Å epitaxial films, on the other hand, do not behave like typical thin films, where magnetization behavior is dominated by shape anisotropy. The 69 Å epitaxial film (Figure 41) shows atypical thin film behavior, with its easy axis out of plane and its hard axes in-plane. Even in an out-of-plane field, however, the remanance is not unity, indicating partial demagnetization. The 206 Å epitaxial thin films (Figure 43) show interesting behavior, with hysteresis both in-plane and out-of-plane. Although the 206 Å epitaxial film has a nominally easy in-plane hysteresis loop, the out-of-plane loop shows remanence and a low saturation field, suggesting low anisotropy between the two directions. The in-plane loops show higher remanance, but complete saturation only at high fields. The out-of-plane loops have less remanence, but saturate at lower fields.

Apart from the 206 Å epitaxial film, the thin films display unambiguous easy axes –in-plane (polycrystalline) or out-of-plane (69 Å epitaxial).
Figure 41: 69 Å epitaxial thin film $M-H$ loop, normalized

Figure 42: 69 Å polycrystalline thin film $M-H$ loop, normalized

Figure 43: 206 Å epitaxial thin film $M-H$ loop, normalized

Figure 44: 206 Å polycrystalline thin film $M-H$ loop, normalized
c. Nanoline Data

VSM $M-H$ loops are presented for each nanoline sample: lines with periods of 500 and 200 nm made from epitaxial and polycrystalline, 69 and 206 Å Ni films. Sample measurements were performed in the same manner as for the continuous thin films. The easy, medium and hard axes of magnetization for each sample are summarized in Table 8.

The 69 Å epitaxial 500 nm period nanolines hysteresis loops, Figure 45, appear at first very similar to the 69 Å epitaxial continuous thin film loops, Figure 41. Out-of-plane magnetization is easy, while in-plane magnetization is isotropic (as for the thin films) - an unexpected result, given the shape anisotropy introduced by patterning. Compared to the continuous thin film, the 500 nm nanolines show a drop in the remanence for the out-of-plane loops, and a reduction in the saturation field for the in-plane loops, pointing to a reduced anisotropy. Compared to the 500 nm period nanolines, the 69 Å epitaxial 200 nm period nanolines, Figure 46, represent a drastic change in anisotropy. The hysteresis loops are very clearly split into 3 distinct loops – the out of plane loop is now a hard axis, while the in-plane transverse loop has become the easy axis, with high remanence. The in-plane parallel loop is intermediate – showing lower remanence than the transverse loop, but saturating below the out-of-plane loop’s saturation field.

The 69 Å polycrystalline nanoline loops represent a dramatic departure from the 69 Å polycrystalline thin film loops (Figure 42), which had the easy axes in-plane. The 69 Å polycrystalline 500 nm period nanolines, Figure 47, show hard-axis behavior for all orientations, with no hysteresis and low coercivity, though the transverse loop is somewhat easier, with a lower saturation field. The 69 Å polycrystalline 200 nm period nanolines, Figure 48, behave in a similar manner as the 500 nm period nanolines. All loops have hard-axis character, with low coercivity and no hysteresis, though the out-of-plane loop has a slightly lower saturation field.
Figure 45: 69 Å epi 500 nm period nanolines $M-H$ loop

Figure 46: 69 Å epi 200 nm period nanolines $M-H$ loop

Figure 47: 69 Å poly 500 nm period nanolines $M-H$ loop

Figure 48: 69 Å poly 200 nm period nanolines $M-H$ loop
Figure 49: 206 Å epi 500 nm period nanolines $M$-$H$ loop

Figure 50: 206 Å epi 200 nm period nanolines $M$-$H$ loop

Figure 51: 206 Å poly 200 nm period nanolines $M$-$H$ loop
Like the loops for the 69 Å epitaxial 200 nm nanolines (Figure 46), the 206 Å epitaxial 500 nm period nanolines (Figure 49) are split into 3 distinct loops. The transverse loop displays easy axis magnetization, with the parallel loop a medium axis and the out-of-plane loop the hard axis. Like its thin film counterpart, Figure 43, the loops are similar in shape, with only small differences between each. The transverse direction is the easy axis, as can be inferred from its lowered saturation field. The 206 Å epitaxial 200 nm period nanolines, Figure 50, like the 206 Å epitaxial 500 nm period nanolines, are also split into 3 distinct loops, though these loops are more spread out, indicating increased anisotropy between the measurement directions. The out-of-plane loop is now a hard axis with no remanence and an extremely high saturation field.

The 206 Å polycrystalline 200 nm period nanolines, Figure 51, closely resemble their continuous thin film counterparts, Figure 44. The in-plane loops are nearly isotropic and the out-of-plane loop is the hard axis, albeit with a lower saturation field than in the continuous thin film.

In summary, the hysteresis loops for the Cu-Ni-Cu nanolines show that patterning has a strong effect upon the magnetic properties of the films. As the line width decreases, the epitaxial Cu-Ni-Cu nanolines’ out-of-plane axis becomes increasingly hard in character, while the in-plane $y$ axis tends to become the easy axis. The 206 Å epitaxial Ni immediately displays 3 distinct loops upon patterning, while the 69 Å epitaxial Ni transitions more gradually, not splitting into 3 loops until patterned into 200 nm period nanolines. The polycrystalline Cu-Ni-Cu nanoline loops show a more subtle effect of the patterning: the 69 Å polycrystalline nanoline loops are all hard-axis in character, while the 206 Å polycrystalline nanoline loops resemble their thin-film counterparts.

**Table 8: Summary of VSM nanoline data**

<table>
<thead>
<tr>
<th>$t_{Ni}$</th>
<th>Line Period</th>
<th>Type</th>
<th>Easy Axis</th>
<th>Med. Axis</th>
<th>Hard Axis</th>
</tr>
</thead>
<tbody>
<tr>
<td>69</td>
<td>500</td>
<td>Epi</td>
<td>Z</td>
<td>-</td>
<td>X, Y</td>
</tr>
<tr>
<td>69</td>
<td>200</td>
<td>Epi</td>
<td>Y</td>
<td>X</td>
<td>Z</td>
</tr>
<tr>
<td>69</td>
<td>500</td>
<td>Poly</td>
<td>-</td>
<td>Y</td>
<td>X, Z</td>
</tr>
<tr>
<td>69</td>
<td>200</td>
<td>Poly</td>
<td>-</td>
<td>Z</td>
<td>Y, X</td>
</tr>
<tr>
<td>206</td>
<td>500</td>
<td>Epi</td>
<td>Y</td>
<td>X</td>
<td>Z</td>
</tr>
<tr>
<td>206</td>
<td>200</td>
<td>Epi</td>
<td>Y</td>
<td>X</td>
<td>Z</td>
</tr>
<tr>
<td>206</td>
<td>200</td>
<td>Poly</td>
<td>X</td>
<td>Y</td>
<td>Z</td>
</tr>
</tbody>
</table>
5.2 Torque Magnetometry

a. Description

A magnetic material placed in a magnetic field will try to rotate, due to a torque on the sample’s magnetic moment, acting to align it with the field. The torque, $L$, is equal to the cross product of the magnetization and the magnetic field: $L = -M \times H$. This effect is not only useful for compasses, but forms the basis of the most widely used method of measuring magnetic anisotropy. Similar to the VSM, a torque magnetometer consists of a computer-controlled electromagnet, a sample mounted on a glass sample holder and a sample rotation stage / torque sensor above (Figure 52). The torque magnetometer, an ADE Technologies Digital Measurement Systems Torque / VSM Model 1660, uses a feedback loop to maintain a net zero torque. The amount of force required to counteract the torque is detected as a voltage. By measuring the torque as a function of angle and extracting the sinusoidal terms, the sample anisotropy energy may be determined. [48]

![Figure 52: Torque magnetometer diagram](image)

A sample’s magnetic anisotropy energy, $E$, can be written as a power series of sine terms. The torque is the negative derivative of energy by angle, so torque curves may be represented as a series of sine terms. For the first two terms, the torque (for a uniaxial magnetic sample) is $^{11}$:

$^{11}$ Note: the $K_n$ terms here refer to generalized energy terms, not the magnetocrystalline anisotropy terms.
In a mixed-anisotropy system with a sixth-power sine term (due to an anisotropy term of cubic symmetry), the torque in a representative plane (ex: a plane including the [100], [110] and [111] directions) will be:

\[ E = K_0 + K_2 \sin^2 \theta + K_4 \sin^4 \theta \]

\[ L = \frac{-\partial E}{\partial \theta} = \left[ -K_2 - K_4 \right] \sin 2\theta + \frac{K_4}{2} \sin 4\theta \]

Ideally, a torque measurement of such a sample would consist of such a series of sine terms, but measurement noise and other effects contribute error that can be captured as cosine terms. So, in a simple (2\textsuperscript{nd} and 4\textsuperscript{th} order only) mixed-anistopy system, torque curves are modeled as [49]:

\[ L = P_1 \sin 2\theta + P_2 \cos 2\theta + P_3 \sin 4\theta + P_4 \cos 4\theta + P_5 \]

For the system with the 6\textsuperscript{th} order terms, the modeling equation for the torque is:

\[ L = P_1 \sin 2\theta + P_2 \cos 2\theta + P_3 \sin 4\theta + P_4 \cos 4\theta + P_5 \sin 6\theta + P_6 \cos 6\theta + P_7 \]

A sample torque curve is shown in Figure 53. This represents the torque curve at 13,500 Oe minus the torque curve at 0 Oe (which represents the effects of gravity on the data). The measured torque is well described by \( \sin(2\theta) \) and \( \sin(4\theta) \) terms, with near-zero \( \cos(n\theta) \) terms.
Figure 53: Torque magnetometer curve (69 Å epitaxial continuous thin film, out-of-plane). $P_1 = 1.07$, $P_2 = -0.05$, $P_3 = 0.25$ and $P_4 = -0.03$, giving $K_2^* t = -0.98$ and $K_4^* t = 0.31$ erg/cm$^2$.

Multiple measurements of each sample were performed at fields well above saturation, 13,500, and at 0 Oe, in 10 Oe steps, with 20 averages per step. The zero-field curve was subtracted from the high-field curve to remove the gravity effects. Due to the low signal-to-noise ratios, the samples required additional measurement processing to extract meaningful parameters. Data sets were smoothed and multiple curves were averaged to further increase the signal to noise ratio. The $\sin(n\theta)$ and $\cos(n\theta)$ terms [2$^{nd}$ and 4$^{th}$ order$^{12}$] were deconvoluted ex situ using Origin. These procedures are described in Appendix 6.

**b. Continuous Thin Film Data**

Torque measurements of the continuous thin film samples were performed on nearly square pieces ~0.5 x 0.5 cm cleaved along the silicon <110>. In-plane measurements were performed with the in-plane sample holder, mounted so that one set of the sample edges was parallel to the magnetic field. Out-of-plane measurements were performed with the out-of-plane sample holder. Samples were mounted square to the sample holder, so that one set of edges was parallel

$^{12}$ There should be no $\sin 6\theta$ term for the out-of-plane measurements taken in this thesis, as a rotation from the Ni (100) through the (101) to the (001) will not pick up any 6$^{th}$ order terms (which would show up on a rotation through the <111>). However, small misalignments (resulting in $\alpha_2 \neq 0$) may result in 6$^{th}$ order components. These are not treated here.
to the ground. The sample holder was mounted so that at 0 degrees the field was parallel to the plane of the film, as shown in Figure 54.

**Figure 54: Diagram of torque measurement directions**

The extracted anisotropy terms are presented in Table 9. The $R^2$ values indicate quality of fit of the simulated torque curves to the data, where 1.00 represents a perfect fit. The lower $R^2$ values indicate noisy data sets, due to near-zero anisotropies.

**Table 9: Continuous Thin Film Torque Data**

<table>
<thead>
<tr>
<th>$t_{\text{Ni}}$ (Å)</th>
<th>Type</th>
<th>Direction(IP or OOP)$^{13}$</th>
<th>$K_2$</th>
<th>$K_4$</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>69</td>
<td>Epitaxial</td>
<td>IP</td>
<td>0.11 ± 0.01</td>
<td>0.06 ± 0.01</td>
<td>.97</td>
</tr>
<tr>
<td>69</td>
<td>Epitaxial</td>
<td>OOP</td>
<td>-0.61 ± 0.04</td>
<td>0.19 ± 0.03</td>
<td>.96</td>
</tr>
<tr>
<td>69</td>
<td>Polycrystalline</td>
<td>IP</td>
<td>0.59 ± 0.13</td>
<td>-0.49 ± 0.12</td>
<td>.86</td>
</tr>
<tr>
<td>69</td>
<td>Polycrystalline</td>
<td>OOP</td>
<td>0.37 ± 0.05</td>
<td>0.03 ± 0.04</td>
<td>.86</td>
</tr>
<tr>
<td>206</td>
<td>Epitaxial</td>
<td>IP</td>
<td>0.39 ± 0.28</td>
<td>0.07 ± 0.25</td>
<td>.87</td>
</tr>
<tr>
<td>206</td>
<td>Epitaxial</td>
<td>OOP</td>
<td>-0.22 ± 0.08</td>
<td>0.29 ± 0.07</td>
<td>.70</td>
</tr>
<tr>
<td>206</td>
<td>Polycrystalline</td>
<td>IP</td>
<td>0.55 ± 0.10</td>
<td>-0.42 ± 0.09</td>
<td>.41</td>
</tr>
<tr>
<td>206</td>
<td>Polycrystalline</td>
<td>OOP</td>
<td>1.31 ± 0.04</td>
<td>0.45 ± 0.03</td>
<td>.99</td>
</tr>
</tbody>
</table>

The torque magnetometer data will be discussed in the following chapter.

---

$^{13}$ $IP$ = In-Plane (measured within sample plane), $OOP$ = Out-of-plane (measurement begins in plane, rotates to out of plane).
c. Nanoline Data

Torque data (Table 10) was measured for the patterned nanoline samples in the same manner as for the continuous thin film samples. Measurement directions are diagrammed in Figure 55.

Figure 55: Torque measurement direction diagram for nanoline samples
Table 10: Nanoline Torque Data

<table>
<thead>
<tr>
<th>t_{Ni}(Å)</th>
<th>Type</th>
<th>Period (nm)</th>
<th>Direction (XY, XZ or YZ)</th>
<th>K_2</th>
<th>K_4</th>
<th>R^2</th>
</tr>
</thead>
<tbody>
<tr>
<td>69</td>
<td>Epitaxial</td>
<td>500</td>
<td>xy</td>
<td>0.42 ± 0.09</td>
<td>-0.51 ± 0.08</td>
<td>.91</td>
</tr>
<tr>
<td>69</td>
<td>Epitaxial</td>
<td>500</td>
<td>xz</td>
<td>0.45 ± 0.05</td>
<td>-0.11 ± 0.04</td>
<td>.89</td>
</tr>
<tr>
<td>69</td>
<td>Epitaxial</td>
<td>500</td>
<td>yz</td>
<td>0.04 ± 0.03</td>
<td>0.02 ± 0.02</td>
<td>.69</td>
</tr>
<tr>
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<td>Epitaxial</td>
<td>200</td>
<td>xy</td>
<td>-1.13 ± 0.10</td>
<td>0.51 ± 0.09</td>
<td>.92</td>
</tr>
<tr>
<td>69</td>
<td>Epitaxial</td>
<td>200</td>
<td>xz</td>
<td>0.27 ± 0.07</td>
<td>0.23 ± 0.06</td>
<td>.92</td>
</tr>
<tr>
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<td>Epitaxial</td>
<td>200</td>
<td>yz</td>
<td>0.43 ± 0.07</td>
<td>0.28 ± 0.06</td>
<td>.95</td>
</tr>
<tr>
<td>69</td>
<td>Polycrystalline</td>
<td>500</td>
<td>xy</td>
<td>0.30 ± 0.04</td>
<td>0.07 ± 0.04</td>
<td>.97</td>
</tr>
<tr>
<td>69</td>
<td>Polycrystalline</td>
<td>500</td>
<td>xz</td>
<td>0.24 ± 0.08</td>
<td>0.12 ± 0.07</td>
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</tr>
<tr>
<td>69</td>
<td>Polycrystalline</td>
<td>500</td>
<td>yz</td>
<td>0.25 ± 0.06</td>
<td>0.02 ± 0.05</td>
<td>.84</td>
</tr>
<tr>
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<td>Polycrystalline</td>
<td>200</td>
<td>xy</td>
<td>-0.72 ± 0.16</td>
<td>0.20 ± 0.14</td>
<td>.79</td>
</tr>
<tr>
<td>69</td>
<td>Polycrystalline</td>
<td>200</td>
<td>xz</td>
<td>0.30 ± 0.08</td>
<td>0.11 ± 0.07</td>
<td>.88</td>
</tr>
<tr>
<td>69</td>
<td>Polycrystalline</td>
<td>200</td>
<td>yz</td>
<td>0.30 ± 0.05</td>
<td>-0.06 ± 0.05</td>
<td>.86</td>
</tr>
<tr>
<td>206</td>
<td>Epitaxial</td>
<td>500</td>
<td>xy</td>
<td>-1.15 ± 0.16</td>
<td>0.19 ± 0.14</td>
<td>.86</td>
</tr>
<tr>
<td>206</td>
<td>Epitaxial</td>
<td>500</td>
<td>xz</td>
<td>0.30 ± 0.06</td>
<td>0.30 ± 0.06</td>
<td>.95</td>
</tr>
<tr>
<td>206</td>
<td>Epitaxial</td>
<td>500</td>
<td>yz</td>
<td>0.39 ± 0.07</td>
<td>0.17 ± 0.06</td>
<td>.93</td>
</tr>
<tr>
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<td>Epitaxial</td>
<td>200</td>
<td>xy</td>
<td>-1.22 ± 0.14</td>
<td>0.50 ± 0.13</td>
<td>.88</td>
</tr>
<tr>
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<td>200</td>
<td>xz</td>
<td>1.60 ± 0.13</td>
<td>1.32 ± 0.12</td>
<td>.99</td>
</tr>
<tr>
<td>206</td>
<td>Epitaxial</td>
<td>200</td>
<td>yz</td>
<td>1.33 ± 0.09</td>
<td>1.05 ± 0.08</td>
<td>.99</td>
</tr>
<tr>
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<td>Polycrystalline</td>
<td>200</td>
<td>xy</td>
<td>-0.57 ± 0.12</td>
<td>0.23 ± 0.11</td>
<td>.91</td>
</tr>
<tr>
<td>206</td>
<td>Polycrystalline</td>
<td>200</td>
<td>xz</td>
<td>0.47 ± 0.05</td>
<td>0.13 ± 0.05</td>
<td>.96</td>
</tr>
<tr>
<td>206</td>
<td>Polycrystalline</td>
<td>200</td>
<td>yz</td>
<td>0.42 ± 0.06</td>
<td>0.15 ± 0.06</td>
<td>.93</td>
</tr>
</tbody>
</table>

The torque magnetometer data will be discussed in the following chapter.
5.3 Strain Characterization

a. Epitaxial Cu/Ni/Cu Continuous Films (interpolation from previous work)

The strain state of epitaxial Cu/Ni/Cu continuous thin films deposited in the exact same manner and on the same equipment as the samples for this work was examined by Ha & Ciria [50] using Bragg diffraction and grazing-incidence diffraction with synchrotron x-rays. Average in-plane biaxial strains show a phenomenological dependence of:

\[ \varepsilon_{IP} = \varepsilon_{\text{misfit}} \left( \frac{27}{t_{Ni}} \right)^{2/3} \]

with \( t_{Ni} \) given in Å. Out-of-plane strains closely follow the predicted bulk value of:

\[ \varepsilon_{OOP} = -2 \frac{c_{12}}{c_{11}} \varepsilon_{IP} = -1.28 \varepsilon_{IP} \]

Table 11 shows the in-plane strains predicted for the 69 and 206 Å epitaxial Ni films. Because only select thicknesses of Ni were measured by Ha & Ciria using x-ray synchrotron, the strain values for these thicknesses of Ni were interpolated using the phenomenological equation derived from the data. [Because epitaxial growth requires clean surfaces, ex situ measurements of film strain for the epitaxial films in between layer depositions could not be performed.]

**Table 11: Interpolated values for in-plane strain of epitaxial Ni films (data from Ha [50])**

<table>
<thead>
<tr>
<th>Nickel Thickness (Å)</th>
<th>Strain</th>
</tr>
</thead>
<tbody>
<tr>
<td>69</td>
<td>0.0139</td>
</tr>
<tr>
<td>206</td>
<td>0.0067</td>
</tr>
</tbody>
</table>

The maximum strain, the lattice mismatch strain, \( \eta \), for Ni on Cu is 0.026. Both 69 and 206 Å are well over the critical thickness (~18 Å), yet significant tensile strain with respect to \( \eta \) is expected to remain in the films. This is largely due to the copper capping layer.

b. Polycrystalline Cu/Ni/Cu Continuous Films

To measure the strain of the polycrystalline continuous Cu/Ni/Cu films, two 3” wafers were deposited with 2000 Å of Cu in the UHV sputter using DC magnetron sputtering. The wafer curvature was measured *ex situ* using a Tencor FLX-2320 Thin Film Stress Measurement
System, which uses a laser to measure the curvature along the wafer diameter (which was consistently chosen to be the diameter intersecting the wafer flat). Nickel films of thickness 69 and 206 Å were deposited using ion beam sputtering, followed by another 50 Å of Cu deposited via DC magnetron sputtering. The wafer curvatures were remeasured to obtain the new radii of curvature. Since each deposited layer contributes independently to the wafer curvature, the resulting strain could be calculated using the Stoney equation [51,52]:

$$
\sigma_f = \left( \frac{E}{1-v} \right) \frac{t_s^2}{6Rf} \left( \frac{E}{1-v} \right) = 1.805 \times 10^{11} \text{Pa} \quad t_s = 350 \mu\text{m}
$$

The measured stresses were converted to strains by dividing by the reduced modulus ($E/(1-v)$) of Nickel, 320 GPa [53]. These strains are presented in Table 12.

<table>
<thead>
<tr>
<th>Nickel thickness (Å)</th>
<th>Stress (GPa)</th>
<th>Strain</th>
</tr>
</thead>
<tbody>
<tr>
<td>69</td>
<td>-2.94</td>
<td>-0.009</td>
</tr>
<tr>
<td>206</td>
<td>-1.53</td>
<td>-0.005</td>
</tr>
</tbody>
</table>

Unlike the tensile mismatch stresses predicted for the epitaxial films, these films are under compressive stress, though with magnitudes lower than those of the epitaxial films. Film stress is highly dependent upon deposition conditions, due to microstructural dependence upon deposition rate, pressure, incoming adatom energy, etc. Researchers have observed transitions from compressive to tensile stress in Ni films with increasing sputter deposition pressure [54]. So for these polycrystalline Ni films, deposited at extremely low pressures, it is entirely reasonable to observe compressive stress. Further, increasing film thickness decreases the strain, as is expected.

**5.4 Summary**

VSM, Torque magnetometry and strain data have been presented for the Cu/Ni/Cu nanolines. The torque magnetometer data will be discussed in subsequent chapters. Strain was measured directly for the polycrystalline films, using wafer curvature measurements before and after film deposition. Strain values for the epitaxial films were interpolated from previous researchers’ x-ray synchrotron data. The strain data from Ha et. al. show that the epitaxial Ni films are in tension, with the strain related to the film thickness with a $(1/t)^{2/3}$ power dependence. The
polycrystalline Ni films, on the other hand, are in compression, with strain values smaller in magnitude than the epitaxial strain values at a given thickness.
Chapter 6
Anisotropy Data Discussion and Analysis

This chapter discusses the Cu/Ni/Cu nanoline magnetic data measured in Chapter 5 in terms of the triaxial anisotropy model developed in Chapter 2 and strain calculations from Chapter 3.

6.1 Comparison of VSM and torque data

Both $M-H$ loops and torque magnetometer curves give important information about the magnetic properties of a sample, yet they have different emphases. The $M-H$ loops are performed at varying fields but fixed field orientation. The magnetization energy to saturate a given field’s orientation may be calculated (if the zero-field magnetization state is known), and the energies at differing orientations compared, yet such comparisons cannot capture the energy of intermediate orientations of potential significance. Torque magnetometer curves are performed at varying orientations but fixed field. In this work, the field for the torque curves was chosen to saturate the sample, thus creating a known magnetization state at every orientation, so that a direct comparison could be made between the energies of fully magnetized states.

Although these two techniques may seem equivalent, there is a subtle difference between the energy at a given orientation and the energy needed to magnetize the sample to an orientation – the former being independent of the magnetization process. If a magnetic sample was homogeneous in its magnetization distribution, the magnetization process would be independent of orientation. In practice, the zero-field magnetization distribution of a thin film or nanostructure is anisotropic, making the magnetization energy likewise anisotropic.

6.1.a Cu/Ni/Cu Thin films

With these differences in mind, let us revisit the VSM and torque magnetometer data presented in the previous chapter. The torque magnetometer data tabulated in Chapter 5 for the thin films is plotted in Figure 56, with estimated error values. The anisotropy terms $K_{ij}$ are positive when $E_j > E_i$ - for example: $K_{XZ} < 0$ for out-of-plane (Z) magnetization ($E_X > E_Z$).
Figure 56: Thin Film Torque magnetometer data, $K_2*t$ and $K_4*t$ (erg/cm$^2$). $K_{n,ij} = E_j - E_i$.

$K > 0 : X$ low energy. $K < 0 : Y$ or $Z$ low energy.

The large, positive $K_{2XY}$ values for the polycrystalline and 206 Å epitaxial films in are puzzling, given that the thin films should have no in-plane anisotropy save for crystal anisotropy (which is a fourth order term), though the non-circular sample shape may introduce a small anisotropy term (due to shape anisotropy and/or anisotropic strain changes), especially in the thicker films. The $K_{4XY}$ values are near-zero for the epitaxial films, as is expected given the small crystal anisotropy term, but unexpectedly large and negative for the polycrystalline films. Both the polycrystalline films had low confidence ($R^2$) values for the in-plane data. Adding the positive $K_{2XY}$ and negative $K_{4XY}$ terms for the polycrystalline films gives an approximate total anisotropy ($K_{Eff}$) of near-zero, which is appropriate, suggesting that high $K_{2XY}$ and $K_{4XY}$ values are an artifact of the low-confidence torque data.

Given the negative $K_{2XZ}$ terms in Figure 56, both epitaxial films might be expected to have perpendicular magnetic anisotropy. Negative $XZ$ anisotropy is consistent with the VSM results for the 69 Å film, which show clear perpendicular anisotropy, but opposite the VSM results for the 206 Å film, which suggest a near-isotropic system with slight preference for in-plane magnetization. Of course, the 206 Å epitaxial film has a positive $K_{4XZ}$ term, which
acts to counterbalance the negative $K_{2XZ}$ term. The total anisotropy for the 206 Å film will be slightly positive, which is consistent with the VSM data. Both polycrystalline films have positive $K_{2XZ}$, with magnitudes increasing with film thickness, indicating easy in-plane magnetization, as would be expected, given the magnetostatic energy considerations. All $K_{4XZ}$ terms are positive, with magnitudes for the 69 Å epitaxial and 206 Å epitaxial and polycrystalline films that are higher than expected from magnetocrystalline anisotropy alone.

**Figure 57: Thin Film $K_{\text{Eff}} \ast t$ - VSM (closed circle) and Torque (open circle)**

The torque $K_2$ and $K_4$ terms are added together (and labeled as $K_{\text{Eff}}$) to make an approximate comparison\(^{14}\) to the $K_{\text{Eff}}$ extracted from the VSM graphs\(^{15}\) for the thin films in Figure 57. The two sets of thin film anisotropy data track fairly closely, given the estimated

\[ E = K_0 + K_2 \sin^2 \theta + K_4 \sin^4 \theta \ldots \]

The difference between the energy at 90° and 0° is given as

\[ \Delta E = K_2 \left[ \sin^2(90) - \sin^2(0) \right] + K_4 \left[ \sin^4(90) - \sin^4(0) \right] + \ldots = K_2 + K_4 \ldots = K_{\text{Eff}}. \]

Thus, adding $K_4$ and $K_2$ to give $K_{\text{Eff}}$ will allow more direct comparison of torque and VSM anisotropy data.

\(^{14}\) The torque magnetometer directly extracts $K_2$ and $K_4$ terms, as discussed in Chapter 5. The VSM can only extract $K_{\text{Eff}}$, the total anisotropy. Since $E = K_0 + K_2 \sin^2 \theta + K_4 \sin^4 \theta \ldots$, the difference between the energy at 90° and 0° is given as

\[ \Delta E = K_2 \left[ \sin^2(90) - \sin^2(0) \right] + K_4 \left[ \sin^4(90) - \sin^4(0) \right] + \ldots = K_2 + K_4 \ldots = K_{\text{Eff}}. \]

\(^{15}\) Each $M-H$ loop was converted to an anhysteretic loop (decreasing-field and increasing-field loops were averaged at equal magnetization values), and the area to the side of the curves, $\int MdH$, was calculated using $\text{Area} = \sum_i (M_i + M_{i+1})(H_{i+1} - H_i)/2$ to obtain the magnetization energy.

The difference between the energy of the in-plane and out-of-plane anhysteretic loops gives the anisotropy.
margins of error. These error estimates may be smaller than the actual error, especially for low-magnitude torque data, such as that for the 206 Å epitaxial film data, which has very low in-plane to out-of-plane anisotropy. For all films except the 206 Å epitaxial film, the VSM anisotropy data has smaller magnitude than the torque data, pointing to systematic differences between the two measurement techniques.

6.1.b Cu/Ni/Cu Nanolines

The torque magnetometer data tabulated in Chapter 5 for the 69 Å nanolines is plotted on the following page, with estimated error values. As for the thin films, the anisotropy terms $K_{ij}$ are positive when $E_j > E_i$. Example: $K_{XY} < 0$ for transverse ($Y$) magnetization ($E_X > E_Y$).

Note: Nanolines are labeled by their nickel film thickness and line period (not line width), except as noted.
Figure 58: 69 Å nanoline XY torque magnetometer data

Figure 59: 69 Å nanoline XZ torque magnetometer data

Figure 60: 69 Å nanoline YZ torque magnetometer data

Figure 61: 69 Å nanoline $K_{Eff} \times t$ torque vs. VSM
Both the VSM and torque show decreases in $XY$ anisotropy for the 69 Å epitaxial nanolines from the thin film to the 200 nm period nanoline samples. The $K_{2XY}$ terms measured by torque magnetometer for the 69 Å nanolines, Figure 58, decrease from the thin film to the 200 nm nanolines, though the 500 nm nanoline value is larger than the thin film value. The $K_{4XY}$ terms are large in magnitude for the epitaxial nanolines, suggesting the effects of noise, not actual signal. The total $XY$ anisotropy decreases monotonically for the epitaxial nanolines. For the polycrystalline nanolines, the $K_{2XY}$ terms show a near-monotonic decrease with decreasing line width, with large-magnitude $K_{4XY}$ terms for the thin film and 200 nm samples. The VSM terms (Figure 61) show the $K_{\text{Eff,}XY}$ for the nanolines decreasing from near-zero to negative at the 200 nm nanoline sample., while the polycrystalline nanolines have near-zero values for the thin film and 200 nm samples, decreasing to negative at the 500 nm samples.

Figure 59 shows the torque $XZ$ anisotropy for the 69 Å nanolines. Both the VSM and torque data (Figure 61) show a rise in $K_{\text{Eff,XZ}}$ as the epitaxial films are patterned into decreasing widths. The torque data shows near-constant positive $K_{2XZ}$ for the polycrystalline samples, while the VSM data shows monotonic decreases in $K_{\text{Eff,XZ}}$, going from positive to negative with decreasing line width. The trends in the $YZ$ anisotropy, Figure 60, mirror those of the $XZ$ anisotropy, with increasing $K_{2YZ}$ and $K_{\text{Eff,YZ}}$ for the epitaxial lines and flat and decreasing $K_{2YZ}$ and $K_{\text{Eff,YZ}}$ for the polycrystalline torque and VSM data, respectively. The $K_{4YZ}$ terms are near-zero, suggesting lower-error torque data.

The sum of the torque $K_2$ and $K_4$ terms is compared to the $K_{\text{Eff}}$ extracted from the VSM graphs (in the same manner as for the thin films) for the 69 Å nanolines in Figure 61. As observed for the thin film data, the torque magnetometer data gives $K_{\text{Eff}}$ values equal or greater in magnitude than the values extracted from VSM anisotropy data, with a couple of exceptions where the two are equal within experimental error. About half of the values are within experimental error of each other, especially for $|K_{\text{Eff}}| > 0.5$ erg/cm$^2$ (as measured by the VSM). For small anisotropy values, the torque magnetometer error is expected to grow, so some of the lack of agreement between VSM and torque is due to the torque’s inherent difficulty at low anisotropies. Other discrepancies are more puzzling, namely the large negative $K_{2XY}$ terms, as discussed earlier and the difference between the $XZ$ and $YZ$ polycrystalline $K_{\text{Eff}}$. 
Figure 62: 206 Å nanoline $XY$ torque magnetometer data

Figure 63: 206 Å nanoline $XZ$ torque magnetometer data

Figure 64: 206 Å nanoline $YZ$ torque magnetometer data

Figure 65: 206 Å nanoline $K_{Eff} * t$ torque vs. VSM
The torque magnetometer data tabulated in Chapter 5 for the 206 Å nanolines is plotted on the previous page, with estimated error values. The sum of the $K_2$ and $K_4$ torque terms is compared to the $K_{\text{Eff}}$ extracted from the VSM graphs (in the same manner as for the thin films & 69 Å nanolines) for the 206 Å nanolines, Figure 65. The $XY$ anisotropy data, Figure 62, show decreases in $K_{2XY}$ for both epitaxial and polycrystalline nanolines with decreasing line width, reaching provocatively large negative values ($K_{2XY} < 0$: $Y$ direction easy axis) for the nanolines. The torque $K_{\text{Eff}}$ data shows similar trends, though skewed by the large $K_{4xy}$ terms. These negative trends to the $K_{\text{Eff}}$ are echoed by the VSM data (albeit with lower magnitudes) for the epitaxial nanolines (Figure 65), but the VSM data indicates increases to the positive $K_{\text{Eff}}$ for the polycrystalline nanolines.

The torque data shows large increases to both $K_{2XZ}$ and $K_{2YZ}$ (Figure 63 and Figure 64) for the epitaxial nanolines, and decreases in both $K_{2XZ}$ and $K_{2YZ}$ for the polycrystalline nanolines. These trends are followed by the $K_{\text{Eff}}XZ$ and $YZ$ data for both torque and VSM (Figure 65). The 200 nm epitaxial samples have anomalously large $XZ$ and $YZ$ terms.

As with the thin film and 69 Å nanoline data, the torque magnetometer data for the 206 Å nanolines gives $K_{\text{Eff}}$ values equal or greater in magnitude than the values extracted from VSM anisotropy data. About a third of the torque and VSM $K_{\text{Eff}}$ values are within experimental error of each other. Much of the variation occurs for $|K_{\text{Eff}}| < 1.0$ erg/cm$^2$ (as measured by the VSM), due to increased torque error at low anisotropy values. Other discrepancies are more puzzling, namely the large negative $K_{2XY}$ terms, as discussed earlier, and the 206epi200 $XZ$ terms.

Overall, the anisotropy data shows that, upon patterning, the out-of-plane ($XZ$ and $YZ$) anisotropy terms increase with decreasing line width for the epitaxial nanolines, transitioning from negative to positive. This indicates that the $X$ and $Y$ directions become more favored upon patterning to narrower line widths. The transition of the epitaxial nanolines from zero- (or positive) to negative $XY$ anisotropy once patterned, increasingly favoring magnetization transverse to the line direction with decreasing line width. The in-plane anisotropy trends with line width are not as clear as the out-of-plane trends, though this is most likely due to uncertainty in the torque magnetometer measurements.
The effect of patterning on the polycrystalline films depends on the measurement technique. The torque data shows no alteration in anisotropy for the 69 Å nanolines, but the in-plane preference decreases for the 206 Å nanolines, while the VSM data shows decreases to both, indicating that perpendicular anisotropy will be favored for the 69 Å 200 nm polycrystalline nanolines. Overall, the polycrystalline nanoline trends are ambiguous.

### 6.2 Calculated epitaxial nanoline energy values

The triaxial nanoline energy model developed in Chapter 2 is restated here:

\[
E_{overall} = E_0 + \mu_0 \frac{N_s M_s^2 \alpha_3^2}{2} + \mu_0 \frac{N_y M_y^2 \alpha_2^2}{2} + K_1 \left[ \alpha_1^2 \alpha_2^2 + \alpha_1^2 \alpha_3^2 + \alpha_2^2 \alpha_3^2 \right] + K_2 \left[ \alpha_1^2 \alpha_2^2 \alpha_3^2 \right] - \frac{2K_{sCu-Ni}}{t} \alpha_3^2 - \frac{2K_{sNi-NiO}}{w} \alpha_2^2 + B \left[ \alpha_1^2 \varepsilon_{11} + \alpha_2^2 \varepsilon_{22} + \alpha_3^2 \varepsilon_{33} \right] - \frac{2B_{sCu-Ni}}{t} \varepsilon_{11}^2 \alpha_3^2 - \frac{2B_{sNi-NiO}}{w} \varepsilon_{11}^2 \alpha_2^2 + \frac{1}{2} \left[ B_1 + m_1 \gamma \right] \left[ \varepsilon_{11}^2 \alpha_1^2 + \varepsilon_{22}^2 \alpha_2^2 + \varepsilon_{33}^2 \alpha_3^2 \right] + \frac{1}{2} \left[ m_2 \gamma \right] \left[ \varepsilon_{11} \varepsilon_{22} \alpha_1^2 + \varepsilon_{11} \varepsilon_{33} \alpha_2^2 + \varepsilon_{33} \varepsilon_{22} \alpha_3^2 \right]
\]

The calculated values \( N_Y \) and \( N_Z \) for \( t = 69 \) and 206 Å and \( w = 100 \) and 250 nm were tabulated in Chapter 2. The average transverse strain, \( \varepsilon_{22} \), calculated for the nanolines was tabulated in Chapter 3. The material parameters for this model are presented in Table 13:

**Table 13: Anisotropy Constants and Material Parameter Values for Nanoline Model**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \mu_0 )</td>
<td>1</td>
<td>( K_{S Cu-Ni} )</td>
<td>0.506 erg/cm(^2)</td>
</tr>
<tr>
<td>( M_S )</td>
<td>485 emu/cm(^3)</td>
<td>( K_{S Ni-NiO} )</td>
<td>0.36 erg/cm(^2)</td>
</tr>
<tr>
<td>( K_1 )</td>
<td>-4.5x10(^4) erg/cm(^3)</td>
<td>( B_{S Cu-Ni} )</td>
<td>-17.95 erg/cm(^2)</td>
</tr>
<tr>
<td>( B_1 )</td>
<td>6.2 \times 10(^7) erg/cm(^3)</td>
<td>( B_{S Ni-NiO} )</td>
<td>-1.22 erg/cm(^2)</td>
</tr>
</tbody>
</table>

Note - ◆: Ref. [55], ◆: Ref. [56].

The values for \( K_S \) and \( B_S \) were extracted from thin film torque magnetometer data in Chapter 2. Missing are values for the two second-order magnetoelastic energy parameters, \( m_1 \gamma \) and \( m_2 \gamma \). Komelj and Fähnle calculated values for these parameters using ab initio density functional electronic theory [57]. Their average values, \(-1.06 \times 10^9\) and \(4.69 \times 10^8\) erg/cm\(^3\), for \( m_1 \gamma \) and \( m_2 \gamma \), respectively, do not agree with the measured \( D_{Eff} \) from Gutjahr-
Löser [58] (whose $D_{\text{Eff}}$ fit assumed no surface magnetoelastic terms), but give a reasonable fit to the data, as described in Chapter 2.

Using the estimated parameter values, as described above, the energy for each orientation was calculated. Comparison of the calculated values to the epitaxial data shows large discrepancies between most of the measurements and the calculated values. The best agreement is found for the VSM $XY$ anisotropy data, plotted in Figure 66. Due to the poorer quality of the torque data, analysis henceforth will work with the VSM data.

Since one of the most crucial parameters in the energy model is the strain state, a comparison was made between the data and calculations made with variations to the strain relaxation parameter, $f$, defined as $\sigma_y = f\sigma_x$ [such that $f=1$ corresponds to a biaxial strain state and $f=0$ corresponds to a uniaxial strain state]. The calculated anisotropy as a function of $f$ (curved solid lines), against the data (constant value points) is shown in Figure 67, Figure 68, Figure 69 and Figure 70.
Figure 67: 69 Å 500 nm calculated and measured $K_{\text{Eff}}$ vs. $f$

Figure 68: 69 Å 200 nm calculated and measured $K_{\text{Eff}}$ vs. $f$

Figure 69: 206 Å 500 nm calculated and measured $K_{\text{Eff}}$ vs. $f$

Figure 70: 206 Å 200 nm calculated and measured $K_{\text{Eff}}$ vs. $f$
The fits for the within-plane ($XY$) anisotropy data yield reasonable $f$ values for all samples. For the 69 Å 500 nm lines, which should have little transverse strain relief, the model intersects with the data at $f = 0.99$. For the 69 Å 200 nm lines and 206 Å 500 nm lines, which should have some transverse strain relief, the model intersects the data at $f \sim 0.6$. And for the 206 Å 200 nm lines, which should have significant strain relief, the model intersects the data at $f \sim 0.25$.

The model yields poor fits for the $XZ$ and $YZ$ data for most samples, with the exception of the 206 Å 500 nm lines, which have reasonable fits for all three anisotropies. It is unclear why the model and the measured data are not in agreement. Of course, the model parameters have not all been measured directly – the $m_1^{\gamma^2}$ and $m_2^{\gamma^2}$ were simulated and the $B_S$ and $K_S$ parameters were extracted using the thin film anisotropy data, fit to torque magnetometer data. Because the fit to the $XY$ anisotropy data is good, while the fit to the $XZ$ and $YZ$ anisotropies is poor, it is likely that there is a systematic error in the $Z$ energy value of the model, which suggests erroneous surface terms, which greatly affect the $Z$ energy.

### 6.3 Summary

The anisotropy data for the epitaxial nanolines shows that the out-of-plane anisotropy terms, $K_{XZ}$ and $K_{YZ}$, increase upon patterning, even transitioning from negative (perpendicular magnetization) to positive (in-plane magnetization) with decreasing line width. This is contrasted with polycrystalline nanolines, whose positive out-of-plane anisotropy terms decrease upon patterning. The in-plane anisotropy trends show transitions from zero anisotropy (no preferred direction) to negative anisotropy (easy axis transverse to the lines) with decreasing line width for the epitaxial nanolines. The polycrystalline nanolines show no clear in-plane anisotropy trends.

Though the anisotropy model did not fit the measured data using the OOF-calculated strain relaxation values, a fit to the data using adjustable strain relaxation values yielded reasonable fits to the $XY$ anisotropy. The transverse strain relaxation values fit to the data were consistent with the expected strain relaxation for the system – with large relaxation predicted for the 206 Å 200 nm wide nanolines, little relaxation predicted for the 69 Å 500 nm nanolines. Compare to Lei et al [ref 25 – Chapter 1], who observed 80% strain relaxation in the transverse direction for 90 nm line width.
nanolines, and moderate relaxation predicted for the 69 Å 200 nm and 206 Å 500 nm nanolines. That the fit of model to data worked well for the $XY$ but not the $XZ$ or $YZ$ anisotropies suggests that further measurements are needed to obtain the values of the surface energy parameters, which would disproportionately skew the $Z$ energies.
Chapter 7
Low Temperature Cu-Ni-Cu Thin Film Properties

This chapter discusses the low-temperature magnetic properties of the Cu-Ni-Cu continuous thin films.

7.1 Introduction

As mentioned in previous chapters, the Cu-Ni-Cu thin film system magnetic properties are governed by many energies, including magnetostatic, magnetocrystalline, magnetoelastic and surface energies. The interplay between these factors causes the system to be perpendicularly magnetized. But is perpendicular magnetization to be expected at all temperatures? The answer depends on how these energies change as a function of temperature. The temperature dependence of some of these factors is well understood, while others’ are not.

7.1.a Magnetostatic Energy vs. Temperature

One of the best-known properties of magnetic materials is the Curie temperature, i.e. the temperature above which a magnetic material transitions from the ferromagnetic state to the paramagnetic state. This transition represents the point where the exchange energy keeping the atomic magnetic moments aligned is overcome by thermal fluctuations. As shown in Figure 71, a ferromagnetic material’s saturation magnetization gradually drops as temperature increases, in a manner that can be approximated with the Brillouin function, $J_{1/2}(x)$ [59].
The magnetostatic energy is strongly dependent upon the saturation magnetization, $M_s$:

$$E_{MS} = \mu_0 \frac{\Delta N}{2} M_s^2 \cos^2 \theta$$

where $\Delta N$ is the demagnetization factor ($4\pi$ for a thin film in a perpendicular, saturating field) and $\theta$ is the angle between the magnetization and the applied magnetic field. It is important to examine the temperature dependence of the saturation magnetization, because variations in the saturation magnetization can cause a dramatic shift in the magnetostatic energy. Nickel’s Curie temperature is 627 K [59], so at room temperature ($0.47 T/T_C$), (see Figure 71) the saturation magnetization will be $\sim 0.95 M_s(0 K)$. Further decreases in temperature to 0 K result in quite small changes to the saturation magnetization. With increasing temperature, the saturation magnetization and magnetostatic energy will decrease.

Interestingly, Bovensiepen et al. have observed that the Curie temperature in ultrathin (<20 Å) Ni/Cu (001) films is strongly dependent upon the Ni film thickness [61]. At extremely low thicknesses, the Curie temperature decreases significantly below the bulk value (Figure 72), due to a transition from 3-dimensional “Heisenberg” behavior to 2-dimensional “Ising” behavior [62].

Figure 71: Reduced magnetization vs. reduced temperature for nickel after Weiss and Forrer [60], reprinted in O’Handley [59], with Brillouin function, $J_{1/2}(\alpha)$ (solid line).
Figure 72: $T_C$ (solid circles) vs. Ni film thickness (in monolayers: 1 ML ~ 1.7 Å), measured by Bovensiepen et. al. [61] using a magnetooptical kerr effect (MOKE) magnetometer

Their range of film thicknesses, < 17 Å, is mostly below the perpendicular regime studied in this thesis. Once the nickel film thickness is $\geq$ 40 Å, the Curie temperature increases to its bulk value of 627 K [63].

7.1.b Magnetocrystalline Anisotropy vs. Temperature

Another well-characterized temperature dependence is that of the bulk magnetocrystalline anisotropy. The magnetocrystalline anisotropy of a cubic material is given as [59]:

$$E_{MC} = K_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_1^2\alpha_3^2 + \alpha_2^2\alpha_3^2) + K_2(\alpha_1^2\alpha_2^2\alpha_3^2)$$

where $K_1$ and $K_2$ are the first two cubic magnetocrystalline anisotropy constants and the $\alpha_i$ are the direction cosines ($K_0$ represents the angle-invariant energy of the system).
Figure 73: Temperature dependence of Ni anisotropy constants after Franse [64]

As can be seen in Figure 73, the values of $K_1$ and $K_2$ are small at room temperature ($-4.5$ and $-2.3 \times 10^4$ erg/cm$^3$, respectively), giving negligible values for the bulk magnetocrystalline energy compared to other energies (as will be seen later) [59]. But below room temperature, $K_1$ increases significantly in magnitude, to $-12 \times 10^5$ erg/cm$^3$. This is now on the same order as the magnetostatic and magnetoelastic energy terms.

The question remains whether the surface magnetocrystalline anisotropy energy will follow the same temperature dependence as the bulk magnetocrystalline anisotropy. Farle et al. measured the temperature dependence of magnetic anisotropy in ultrathin Ni (001) films on Cu (001) using ferromagnetic resonance in situ. Below the critical thickness, no dislocations form, so there is no variation of strain with thickness. The magnetic anisotropy values could be divided into a volumetric (values not a function of film thickness) and a surface component (values proportional to $1/t$). The second-order surface anisotropy values of a 7.6 monolayer ($\sim 13$ Å) Ni film as a function of the reduced temperature $T/T_C$ are presented below (Figure 74) [65].
This graph of the surface magnetocrystalline anisotropy constant, $K_S$, gives an energy value of 0.2 erg/cm$^2$ at room temperature ($T/T_C = 0.47$), on the same order as the 0.62 erg/cm$^2$ value of $K_S$ calculated in Chapter 2. It is unclear whether the graphed values of $K_S$ are generally applicable for the Ni/Cu system, given that the measurement was performed for only one sample. It is also unclear whether $K_S$ changes at lower temperatures than shown in Figure 74, or has reached a plateau.

One possibility for the variation of $K_S$ with lower temperatures will be as a function of the reduced magnetization, $m$. For uniaxial symmetries (such as for the surfaces), the anisotropy will vary as the cube of the reduced magnetization, while for cubic symmetries (bulk crystalline anisotropy), the anisotropy will vary as the tenth power of the reduced magnetization [59]. This will be revisited in the discussion section of this chapter.

### 7.1.c Magnetoelastic Energy vs. Temperature

Two factors will independently influence the temperature dependence of the magnetoelastic energy, a function of both strain ($\varepsilon_{ii}$) and magnetoelastic parameters ($B_i$) [59]:

$$E_{ME,1} = B_1 \left( \alpha_1^2 \varepsilon_{xx} + \alpha_2^2 \varepsilon_{yy} + \alpha_3^2 \varepsilon_{zz} \right) + B_2 \left( \alpha_1 \alpha_2 \varepsilon_{xy} + \alpha_1 \alpha_3 \varepsilon_{xz} + \alpha_2 \alpha_3 \varepsilon_{yz} \right)$$

First, let us consider the variation of strain with temperature. Due to thermal expansion, both nickel and silicon will tend to contract with decreases in temperature. Because silicon’s
thermal expansion coefficient, $\alpha$, is lower than nickel’s [66] (2.6 vs. $13.4 \times 10^{-6}$/K at 293 K), it will contract to a lesser degree. The nickel will therefore be under tension from the substrate, due to the differential contractions of the two materials. The exact magnitude of this thermal tensile stress can be found by calculations via the Vilms-Kerps formula [67]:

$$\varepsilon_i = \frac{\langle \varepsilon_c \rangle \Delta \left[ 1 + 3(1 + \delta)(1 + 2[\delta_1 + \delta_2 + \ldots \xi \delta]) \right]}{1 + \Delta + 3\Delta (1 + \delta)^3} - \varepsilon_{ci}$$

where $\xi$ is the position parameter $r_i / t_i$ within each layer ($r_i$ the distance from the bottom of the film), $\varepsilon_i$ the strain of film $i$ at position $\xi$, $\varepsilon_{ci}$ the difference between the freestanding thermal strain of film $i$ and silicon, $<\varepsilon_{ci}>$ the average differential thermal strain of all the films, $\delta_i = t_i / t_s$, $\delta = \sum_i t_i / t_s$, $\Delta = \sum_i (t_i E_i / (1 - \nu_i))(1 - \nu_s) / E_s$. The calculated strain change (relative to its strain at 293 K) for Ni is plotted in Figure 75, below.

![Freestanding vs. multilayer thermal strains, Ni and Si](image)

**Figure 75: Freestanding vs. multilayer thermal strains, Ni and Si**

Though freestanding nickel will contract at lower temperatures, when it is part of a multilayer system with copper and silicon, the nickel layer will be under tension.

The magnetoelastic constants at low temperatures have been measured for bulk nickel crystals by Lee and Asgar using a cantilever bending method [68], as shown in Figure 76.
Figure 76: Ni magnetostriction constants vs. temperature by Lee & Asgar [68], where

\[ h_1 = -b_1/(c_{11} - c_{12}) \] and \[ h_2 = b_2/c_{44} \], measured on 2 samples (open & closed circles)

The magnetoelastic coefficient of Cu/Ni/Cu has also been measured directly as a function of temperature by Ciria et. al., using a cantilever method [69], as shown in Figure 77.

Figure 77: Magnetoelastic stress \( B_{\text{eff}} \) vs. \( T \) for Cu/Ni/Cu by Ciria et. al. [69]

They observed that the magnetoelastic parameter, \( B_{\text{eff}} \), of biaxially strained Ni increases at low temperature. \( B_{\text{eff}} \) includes first order, second order [70] and surface magnetoelastic effects. It is difficult to determine which parameters contribute substantively to the measured temperature variation. However it is clear that in the low temperature range (<200 K), \( B_{\text{eff}} \) is nearly constant. From 300 K to 200 K, \( B_{\text{eff}} \) for Cu/Ni/Cu varies more strongly than the corresponding magnetoelastic constants \( h_1 \) and \( h_2 \) for pure Ni measured by Lee & Asgar, pointing to other parameters’ influence.
7.2 VSM data

For an overview of how the properties of the epitaxial Cu/Ni/Cu thin film system vary with lowered temperature, $M-H$ hysteresis loops were measured by VSM for $t_{Ni} = 57, 92, 103, 115, 161, 172, 206, 226$ and $1635$ Å. Measurements were performed between +/- 10 kOe at 293 and 173 K, in the film plane and perpendicular to the film plane. The anisotropy was determined from the difference between the saturation energy at each orientation. The graphs, below, show normalized, centered data between +/- 1 kOe, corrected to remove slopes above 6 kOe (an artifact of the diamagnetic substrate signal).
Figure 78: Ni 57 Å In-Plane Hysteresis Loops, 173 and 293 K

Figure 79: Ni 57 Å Out-of-Plane Hysteresis Loops, 173 & 293 K

Figure 80: Ni 92 Å In-Plane Hysteresis Loops, 173 and 293 K

Figure 81: Ni 92 Å Out-of-Plane Hysteresis Loops 173, 293 K
Figure 82: Ni 103 Å In-Plane Hysteresis Loops, 173 and 293 K

Figure 83: Ni 103 Å Out-of-Plane Hysteresis Loops 173, 293 K

Figure 84: Ni 115 Å In-Plane Hysteresis Loops, 173 and 293 K

Figure 85: Ni 115 Å Out-of-Plane Hysteresis Loops 173, 293 K
Figure 86: Ni 161 Å In-Plane Hysteresis Loops, 173 & 293 K

Figure 87: Ni 161 Å Out-of-Plane Hysteresis Loops 173, 293 K

Figure 88: Ni 172 Å In-Plane Hysteresis Loops, 173 and 293 K

Figure 89: Ni 172 Å Out-of-Plane Hysteresis Loops 173, 293 K
Figure 90: Ni 206 Å In-Plane Hysteresis Loops, 173 and 293 K

Figure 91: Ni 206 Å Out-of-Plane Hysteresis Loops 173, 293 K

Figure 92: Ni 226 Å In-Plane Hysteresis Loops, 173 and 293 K

Figure 93: Ni 226 Å Out-of-Plane Hysteresis Loops 173, 293 K
Figure 94: Ni 1635 Å In-Plane Hysteresis Loop, 173 and 293 K

Figure 95: Ni 1635 Å Out-of-Plane Hysteresis Loops 173, 293 K

Figure 96: $M_r/M_S$ vs $t_N$, 173 and 293 K, IP and OOP

Figure 97: $H_c$ vs $t_N$, 173 and 293 K, IP and OOP
The $M-H$ loops show the expected trends with thickness: at low thicknesses, in-plane axes are hard and the out-of-plane axis is easy (based on the remanences). As thickness increases, the in-plane axes become progressively less hard, with the formerly linear hard loops becoming hysteretic, remanent magnetization and coercivity increasing steadily and saturation fields decreasing. At the same time, the out of plane $M-H$ curve transitions from an upright and square easy loop, shearing over, with gradual drops in the remanent magnetization and coercivity and increasing saturation field, eventually becoming a completely nonhysteretic hard axis loop.

The temperature trends (tabulated in Figure 96 and Figure 97) are less obvious, given the noisy loops, but some observations can be made. As temperature increases, the coercive field decreases slightly in nearly all cases. For many loops, as the temperature increases, the saturation field decreases, while the loop area decreases and the remanent magnetization decreases (ex: Figure 87, Figure 88 and Figure 94).

### 7.3 DC-SQUID Data

To gain a more detailed picture of how a given sample varies its properties as a function of temperature, measurements were performed using SQUID magnetometry on two samples – Ni 92 and 226 Å – in-plane and out-of-plane, between +/- 10 kOe at 75, 125, 175 and 225 K. The DC-SQUID magnetometer (Direct Current Superconducting Quantum Interference Device) measures $M-H$ loops at temperatures down to 4 K.

The graphs, below, show data that has been centered (with respect to magnetization) and levelled (diamagnetic slope subtracted), with outliers removed. In each $M-H$ loop series, there is a distinct trend. The coercive field, $H_c$, and the remanent magnetization, $M_r$ (the magnetization at zero magnetic field), decrease slightly with increasing temperature. The loops also become smaller in area (representing less energy loss due to hysteretic effects) as temperature increases. This suggests that with additional ambient thermal energy, the magnetic energy needed to magnetize the sample, overcoming its internal energy barriers, is reduced. The anisotropy field $H_A$ of the samples\(^\text{17}\) – with the exception of the Ni 92 Å in-

\(^{17}\) $H_A$ is the field at which the (linear) magnetization reaches saturation. For samples with significant nonlinear magnetization curves, $H_A$ will be below the actual saturation field.
plane sample - did not significantly vary with temperature. This is shown in Figure 102, Figure 103 and Figure 104.

Each $M-H$ loop was converted to an anhysteretic loop (decreasing-field and increasing-field loops were averaged at equal magnetization values), and the area to the side of the curves ($\int HdM$) was calculated$^{18}$ to obtain the magnetization energy. The difference between the energy of the in-plane and out-of-plane anhysteretic loops at each thickness, which is a reasonable measure of the total anisotropy, is plotted in Figure 105 as a function of temperature. Both thicknesses show a distinct decrease in anisotropy as a function of increasing temperature. The sign of the anisotropy for the 226 Å samples at low temperatures is positive, which does not agree with the clear easy in-plane axes observable in the $M-H$ loops. This discrepancy will be discussed in the subsequent section.

$^{18}$ Area $= \sum_i \frac{M_i + M_{i-1}}{2} (H_i - H_{i-1})$
Figure 98: In-Plane $M-H$ loops, Ni 92Å: 75, 125, 175 & 225 K

Figure 99: OOP $M-H$ loops, Ni 92Å: 75, 125, 175 & 225 K

Figure 100: In-Plane $M-H$ loops, Ni 226Å: 75, 125, 175 & 225 K

Figure 101: OOP $M-H$ loops, Ni 226 Å: 75, 125, 175 & 225 K
Figure 102: SQUID $H_c$ vs $T$ for Ni 92, 226 Å

Figure 103: SQUID $M_r/M_S$ vs $T$ for Ni 92, 226 Å

Figure 104: SQUID $H_a$ vs $T$ for Ni 92, 226 Å

Figure 105: SQUID $K_{eff}$ vs $T$ for Ni 92 and 226 Å (293 K points from torque magnetometer data)
7.4 Discussion

a. Temperature trends in remanence and coercivity

The $M-H$ loops measured by both VSM and by SQUID magnetometers show clear decreases in both $H_c$ and $M_r$ with increasing temperature. To understand this behavior, it is important to discuss the magnetic domain structure of the Cu/Ni/Cu thin films.

The drop in remanence with increasing temperatures may be due, in part, to increases in domain wall widths. Domain wall widths are controlled by a balance of exchange and anisotropy energies\(^{19}\), so when increases in temperature result in decreases in anisotropy, then exchange energy (much less sensitive to temperature) will result in widened walls.

The coercivity in most magnetic samples (excluding single-domain particles) comes from their magnetic domains. A soft magnetic material will often appear to be demagnetized, due to its tendency to spontaneously form domains. These domains reduce the magnetostatic energy, which would otherwise be quite large, with uncompensated spins at the surfaces. Magnetization of soft magnetic materials in easy directions is accomplished by sweeping domain walls through the material. In materials with defects, domain wall motion requires overcoming energy barriers. This results in $M-H$ loops with hysteretic loss and coercivity.

It is reasonable to expect that any process involving energy barriers will be sensitive to temperature. Added thermal energy will decrease the magnetic energy needed to overcome pinning sites, thus resulting in reduced coercive fields. Similarly, magnetic remanence is also a function of thermal energy – with additional energy, the magnetic spins precess at greater angles from the magnetization vector and with less coherency, increasing the probability of spontaneous demagnetization. Increasing temperature may result in destabilizing some of the smaller magnetic domains, with corresponding reductions to the remanent magnetization.

A more detailed understanding of how the Cu/Ni/Cu magnetic properties vary with nickel film thickness requires examination of the domain structure. Bochi et. al. used magnetic

\[ \delta_{dw} = \frac{\pi \sqrt{A/K_u}}{A} \]  

\(^{19}\) example- 180° domain wall width for a sample with uniaxial anisotropy $K_u$: $\delta_{dw} = \frac{\pi \sqrt{A/K_u}}{A}$ (A is the material-specific exchange stiffness constant)
force microscopy (MFM) to examine the magnetic domain structure of Cu/Ni/Cu films grown in similar conditions [71]. Examining as-grown Cu/Ni/Cu thin films (Figure 106), they found a refinement in domain structure with increasing thickness.

**Figure 106:** 12x12 µm MFM images of as-grown Cu/Ni/Cu, \( t_{\text{Ni}} = 85, 100 \) and 125 Å, showing transition from perpendicular domains (85 Å, left) to perpendicular domains mixed with “bubble” domains (100 Å, center) to bubble domains (125 Å, right) [71].

As illustrated in Figure 107 (left), below ~90 Å, the domains (as-grown) are exclusively perpendicularly magnetized, mesoscopic (1-4 µm diameter) domains of irregular shape. Between the perpendicular domains are in-plane Bloch walls with thicknesses \( \geq 30 \text{ nm} \) (magnetization parallel to wall length to minimize magnetostatic energy). Above ~90 Å (Figure 107, right), a different set of domains evolves – the small, cylindrical “bubble” domains (~0.2 µm diameter), with the cylinder’s magnetization out-of-plane, probably
surrounded by relatively broad in-plane (or complex-orientation) domain walls [72]. As film thickness increases, the in-plane remanence increases, indicating that the easy axis returns to the film plane. The bubble domains (shown in Figure 106 for the 125 Å Ni) represent the equilibrium domain structure\(^{20}\). At extremely large film thicknesses (~2000 Å), the domain structure appears maze-like, ~125 nm wide, with quantitative MFM suggesting that the perpendicular domains are capped by closure domains whose magnetization is canted by 51° and surrounded by in-plane Bloch walls [73].

This transition in magnetic domain structure accounts for the different coercivity behavior between the 92 and 226 Å samples shown in Figure 102. We see near-linear decreases in coercivity with increasing temperature, with weak temperature dependence for the high-coercivity 92 Å samples, moderate temperature dependence for the high in-plane coercivity of the 226 Å Ni and strong temperature dependence for the low-coercivity out-of-plane 226 Å Ni sample.

The 92 Å Ni film is composed of perpendicular domains with a few bubble domains. Out-of-plane magnetization – along the film’s easy axis – is likely accomplished both by sweeping domain walls through the material (to grow the domains with magnetization parallel to the applied field) and by rotating the magnetization of the in-plane domain walls out of plane. The observed \(M-H\) loop (Figure 99) shows features of both effects – the near-square center loop (from domain wall motion) and the gradual (~linear) approach to saturation (from rotation of the small volume of in-plane domain wall moments). The in-plane magnetization (Figure 98) is much more linear (likely due to rotation of the magnetization of the dominant perpendicular domains), with a much smaller central loop (the hysteresis most likely due to the domain walls’ straightening into stripes).

The 226 Å Ni films are composed entirely of bubble domains. Out-of-plane magnetization requires short-range domain wall motion to grow the domains with magnetization parallel to the field – a relatively reversible process due to the small distances travelled by the domain walls (Figure 101). In-plane magnetization is accomplished by rotating the perpendicular

\(^{20}\) NB: The large-scale domains visible in Figure 106 for the 125 Å Ni vanish upon AC demagnetization.
domains and aligning the in-plane domain walls with the field direction, which requires moving them over a longer range than in the perpendicular process.

Comparing the coercivity values as a function of film thickness and temperature (Figure 102), additional observations may be made. Assuming that the defect density is independent of direction within the nickel films, the amount of energy required to move a domain wall will be a function of the distance the domain wall moves during the magnetization process. The highest coercivities should be found in samples where the domain walls have the farthest distances to move. These are the 92 Å IP, 92 Å OOP and 226 Å IP samples.

The 92 Å domains are large, so magnetization, whether in-plane or out-of-plane, requires long-range domain wall motion. Although the 226 Å domains are small, in-plane magnetization will require long-range domain wall motion. The 226 Å OOP sample shows the lowest coercivity because its magnetization reversal is dominated by short-range domain wall motion and rotation of the domain wall magnetization (a zero-coercivity process). As coercivities increase, the slope with temperature decreases. This would suggest that the coercivities are a function of the energy barriers to magnetization – small energy barriers would give lower coercivity and correspondingly larger variation with temperature, given the smaller Arrhenius rate term $E_a/k_B T$.

b. Temperature trends in anisotropy

The $K_{\text{eff}}$ data extracted from the SQUID $M$-$H$ loops (Figure 105) for the 92 and the 226 Å Ni films show intriguing trends in anisotropy. The 92 Å film shows positive anisotropy ($M$ out of plane) over the entire temperature range studied while the 226 Å film data suggest a possible transition from positive anisotropy to negative anisotropy with increasing temperature. The smaller anisotropy values for the 226 Å film have larger error, suggesting that the anisotropy trends must be treated cautiously, given that the axes of magnetization from the DC-SQUID loops suggest unambiguously that the preferred axis of magnetization is in-plane. From 293 K down to 75 K, the 92 Å film anisotropy increases by $\sim2.0 \times 10^5$ erg/cm$^3$. The question is: which factors account for the observed $K_{\text{eff}}$ changes?

First, let us consider the magnetostatic energy. As discussed earlier, the saturation magnetization at 293 K is 485 emu, increasing to 510 emu at 4.2 K. At 75 K, $M_s \sim 0.99$
\( M_s(0) \), so decreasing from 293 to 75 K gives an increase in magnitude of the magnetostatic energy, -2\( \pi M_s^2 \), of ~1.2x10^5 erg/cm^3.

Next, let us consider the magnetocrystalline anisotropy energy. The bulk magnetocrystalline anisotropy term \( K_1 \) increases by over an order of magnitude from room temperature to 0 K, from -4.5 to -120 x10^5 erg/cm^3. \( K_2 \) also increases in magnitude, from -2.3x10^4 to +10x10^4 erg/cm^3 over the same temperature range [64]. It is not obvious how the bulk magnetocrystalline anisotropy will contribute to the net anisotropy, given that, at saturation, the angular terms for in-plane and out-of-plane magnetization are both zero.\(^{21}\) The data from Farle et. al. (Figure 74) suggest that the surface magnetocrystalline energy does not vary significantly with temperature below 0.5 \( T/T_C \) (room temperature).

Finally, let us look at the magnetoelastic energy. As discussed in Chapter 2, the magnetoelastic energy consists of first- and second-order bulk and first-order surface magnetoelastic terms interacting with the film strain. The first-order magnetoelastic parameter, \( B_1 \), is well characterized as a function of temperature, rising from 6.2x10^7 erg/cm^3 at room temperature to ~6.6x10^7 erg/cm^3 at 75 K \({[68]}\), scaling \( B_1 \) (T=293) by \( h_1(75)/h_1(293) \). The remaining magnetoelastic parameters, \( D_{Eff} \) and \( B_S \), are uncharacterized with respect to temperature, although Ciria et. al. [70] have characterized \( B_{Eff} \) (which should encompass first-order, second-order and surface terms) as a function of temperature, showing changes from ~6.2 x10^7 to ~8.5 x10^7 erg/cm^3 between 293 and 73 K. Assuming the strain changes are well-characterized by Figure 75, and using Ciria et. al.’s values of \( B_{Eff}(T) \), a reduction in temperature from 293 to 75 K should give a magnetoelastic energy change of +9.8 and +7.1 x10^5 erg/cm^3 for the 92 and 226 Å Ni films, respectively.

Together, the predicted total changes in the magnetostatic and magnetoelastic energies (+8.6 and −5.8 x10^5 erg/cm^3 for the 92 and 226 Å Ni films, respectively) from 293 to 75 K overestimate the observed changes to \( K_{eff} \). The source of this discrepancy is unclear. Regardless, the disagreement between predicted and measured anisotropy points to gaps in our understanding of how the Cu/Ni/Cu system varies as a function of temperature.

\(^{21}\) Though the bulk magnetocrystalline energy at \( \alpha_1=1 \) and at \( \alpha_3=1 \) when the sample is saturated should be zero, the anisotropy surface itself has the full contribution from the cubic magnetocrystalline anistropy term. The magnetization process involves states below saturation, so off-axis contributions must be considered. Such a treatment is beyond the scope of this thesis.
A more general approach than relying on measured parameter values of Cu/Ni/Cu may be to characterize thermal variations as a function of the reduced magnetization, $m(T)$. This approach will better lend itself to model calculations of anisotropy and $M-H$ loops at various temperatures. For uniaxial symmetries, $K(T)$ varies as $K(0) m(T)^3$ below $0.6 \frac{T}{T_C}$. For cubic symmetries, $K(T)$ varies as $K(0) m(T)^{10}$. The temperature dependence of the anisotropy is a function of the overall anisotropy surface: high curvature of energy with respect to angle will result in steeper temperature dependence [59].

In the thin films, most terms, with the exception of the bulk magnetocrystalline energy, have uniaxial symmetry, due to the tetragonal film distortion, the 2-dimensional shape and the Cu/Ni interfaces. I will assume that the bulk magnetocrystalline anisotropy terms can be neglected to first order. Thus, $K_{\text{Eff}}(T)$ may be modeled as:

$$K_{\text{Eff}} = MS(T) + ME_1(T) + ME_2(T) + ME_S(T) + MC_S(T)$$

where $N^*$ is a thin film scaling factor decreasing the magnetostatic energy. The inclusion of such a factor can be justified by observing that the saturation magnetization is affected by the film thickness. Previous researchers [71] have observed $M_S$ for Cu/Ni/Cu thin films to be 435 emu, rather than the bulk value of 485 emu. This corresponds to a $N^*$ factor of 0.9.
The resulting simulations of $K_{\text{Eff}}(t_{\text{Ni}}, T)$ give reasonably good agreement to the data for both the Ni 92 and 226 Å films, although future refinement to the model is in order. The $N^*$ value chosen, 0.89, is close to 0.9, the value predicted.

### 7.5 Summary

Low temperature $M$-$H$ loops for the Cu/Ni/Cu films have been measured by VSM and SQUID. Coercivity and remanence decrease with increasing temperature, consistent with the behavior of a thermally-activated processes, namely domain wall motion in an applied magnetic field. The magnetization behavior is strongly dependent upon the nickel film thickness, due to the different domain states that result from the change in easy axis from out-of-plane to in-plane, with corresponding changes to magnetization processes.

The effective anisotropy, $K_{\text{Eff}}$, has been plotted from SQUID magnetometer data. The plots show anisotropy moving towards more negative values with increasing temperature for both the 92 and 226 Å Ni films. The anisotropy remains positive (M out of plane) for the 92 Å film over the entire temperature range studied. The anisotropy value for the 226 Å Ni is smaller in magnitude and hence is less certain; it is difficult to determine whether it changes sign from negative to positive at lower temperatures. The observed $K_{\text{Eff}}$ can not be entirely

![Figure 108: Simulated $K_{\text{Eff}}$ vs. $T$, with $N^* = 0.89$](image)
explained by the few measured values for the parameters affecting the magnetic anisotropy. A model of $K_{\text{Eff}}(T)$ as a function of the cube of the reduced magnetization, $m(T)^3$, gives reasonable agreement to the data, although inclusion of the bulk anisotropy term should improve the fit.

In sum, the anisotropy observed at room temperature has been found to change with temperature. Due to the interplay between thermal strain and the changes to the energy parameters, the anisotropy becomes more positive as the temperature decreases, causing positive anisotropy films to become even stronger in their perpendicular anisotropy and possibly causing low-anisotropy in-plane magnetized films to become perpendicularly magnetized. These anisotropy changes are governed by the dependence of effective anisotropy on different powers of the temperature-dependent reduced magnetization, $M(T)/M(0)$.

The effective anisotropy, $K_{\text{Eff}}$, has been plotted from SQUID magnetometer data. The plots show decreases in anisotropy with temperature for both the 92 and 226 Å Ni films. The observed $K_{\text{Eff}}$ can not be entirely explained by the few measured values for the parameters affecting the magnetic anisotropy, suggesting that more complete information about how the strain, surface parameters and second-order magnetoelastic energy parameters vary with temperature is needed. A model of $K_{\text{Eff}}(T)$ as a function of the cube of the reduced magnetization, $m(T)^3$, gives good agreement for the film data.
Chapter 8
Summary, conclusions and future Work

8.1 Summary

a. Cu/Ni/Cu nanolines

The Cu / Ni / Cu thin film system has been studied in detail at room temperature by previous researchers, giving insights on how surface anisotropy, magnetoelastic energy and magnetostatic energy interact to affect the magnetic properties of an epitaxial, strained thin film system. Until now, researchers had explored a purely uniaxial system, treating the differences between the out-of-plane and in-plane (without angular dependance) properties, which are governed by the top and bottom interfaces, by the two-dimensional shape and by the biaxial film strain. Theorists had generalized the equations for a cubic system predicting that the surface anisotropy and magnetoelastic energy, but there was limited data for the higher order relations, especially surface anisotropy. In these uniaxial models, the only strain relief is that due to misfit dislocations which relieve the biaxial in-plane strain.

In Chapter 2, the uniaxial thin film model was expanded to a triaxial model for Cu/Ni/Cu nanolines. The triaxial model considers transverse within-line strain relief (due to the proximity of the surfaces defining the line width), necessitating two second-order magnetoelastic terms instead of the single term used in the thin film model. Using a measured value for the second-order magnetoelastic constant, \( D_{\text{eff}} \), new values of \( B_S \) and \( K_S \) were fit to thin film torque magnetometer data. Demagnetization factors were calculated for the nanoline cross sections used in this work.

The strain state for the Cu/Ni/Cu nanolines, simulated by finite element modeling using the OOF program, was described in Chapter 3. The transverse line strain relief is largely a function of cross-section aspect ratio, although any removal of the Cu “substrate” due to overetching results in increased strain relief in the Ni layer. The outer 1-1 \( \frac{1}{2} \) thickness units of a given cross section relax their strain completely, while the center regions retain their biaxial strain state.
Chapter 4 described the fabrication procedures for the Cu/Ni/Cu nanolines. Epitaxial and polycrystalline Cu/Ni/Cu films with nickel thicknesses of 69 and 206 Å were grown using molecular beam epitaxy and DC magnetron / ion beam sputtering, respectively. Nanolines of line period 200 and 500 nm were fabricated using interferometric lithography and subtractive patterning. SEM images show good long-range line quality, with line width approximately 40% of line period.

The strain data for the Cu/Ni/Cu thin films, and $M-H$ and torque data for the Cu/Ni/Cu nanolines were presented in Chapter 5. The epitaxial film strain data (interpolated from previous researchers’ x-ray synchrotron data) shows the nickel to be under biaxial tension, while the polycrystalline film strain data shows the nickel under biaxial compressive strain. VSM data showed that the narrower epitaxial Cu/Ni/Cu nanolines have greater misfit strain relief across their width and show easy magnetization axes transverse to the line width, contrary to the directions predicted by magnetostatic energy considerations alone. Patterning the epitaxial Cu/Ni/Cu films into nanolines reduced or removed the perpendicular magnetic anisotropy.

In Chapter 6, the anisotropy data (from VSM $M-H$ loops and torque magnetometer data) for the epitaxial nanolines was analyzed and discussed. In large, the data confirmed the trends observed for the $M-H$ loops in Chapter 5. The epitaxial out-of-plane anisotropy terms ($K_{XZ}$ and $K_{YZ}$) all became increasingly positive upon patterning, confirming a transition from out-of-plane ($K<0$) to in-plane magnetization ($K>0$) as the line width decreases. The epitaxial in-plane anisotropy terms ($K_{XY}$) changed to favor transverse magnetization. This was contrasted to the polycrystalline nanolines, whose positive out-of-plane anisotropy reduced in magnitude and whose in-plane anisotropy showed little change upon patterning.

Quantitative analysis was performed in Chapter 6 by fitting to the triaxial anisotropy model developed in Chapter 2. A good fit was achieved for the $XY$ anisotropy terms when the misfit strain relaxation parameter was allowed to vary, but the model fit was poor for the $XZ$ and $YZ$ anisotropy terms, suggesting that better values for the surface magnetocrystalline and magnetoelastic parameters are needed.
b. Low temperature properties

Previous researchers had explored the magnetic properties of bulk nickel as a function of temperature, but few researchers had explored the Cu / Ni / Cu properties as a function of temperature, with the exception of the magnetoelastic coupling coefficients. No study to date has explored the overall magnetic anisotropy of Cu / Ni / Cu thin films as a function of temperature.

Chapter 7 presented Cu/Ni/Cu VSM data for a variety of nickel film thicknesses at two representative temperatures and SQUID data for two nickel film thicknesses at several temperatures. The anisotropy data as a function of temperature was modeled as a function of the reduced magnetization cubed. The model used the room temperature values for the magnetoelastic parameters (first order, second order and first order surface) and the surface magnetocrystalline anisotropy parameter together with calculated values for the nickel film strain. Overall, this model for the anisotropy as a function of temperature gives good agreement to the data.

8.2 Conclusions

In the past, researchers developing nanoscale patterned magnetic devices had worked almost exclusively with polycrystalline thin films. The effects of shape and crystalline anisotropy have been well understood, while the effects of strain have been either sidestepped (through the choice of low-magnetostriction materials or low-strain growth conditions) or characterized empirically. Working with epitaxial films, which will prove advantageous from the standpoint of film quality, will require both a sound understanding of the effects of strain and a willingness not merely to characterize these effects but to use strain as an additional design variable to attain the desired properties.

If developers add strain engineering to their design toolkits, they will be able to broaden their range of materials having a given set of properties, or broaden the range of properties exhibited by a given material. This flexibility will enable the creation of novel magnetic devices whose properties will be controllable not just through materials choice and shape but also through strain. A nanostructure could have its strain state set through choice of an appropriate growth layer. Alternatively, if a zero-anisotropy structure were desired, the
designers could choose an orientation, shape and strain state so that the magnetostatic, magnetoelastic and magnetocrystalline effects cancelled each other out.

This work has demonstrated that magnetoelastic energy can substantially affect the properties of a nanostructure, even to the point of changing the preferred axis of magnetization upon patterning. This effect can even outweigh the magnetic effects of shape, which is traditionally assumed to dominate the preferred direction of magnetization in zero field. For perspective, consider a compass needle, which uses shape anisotropy to set the magnetization along the length of the needle. A nanoline with magnetization transverse to the line direction is as unexpected as a compass needle pointing east!

The analysis developed in this work for Cu/Ni/Cu nanolines can be generalized for nanolines with other materials, or can be modified (with the addition of appropriate surface and demagnetization terms) to treat finite nanostructures instead of nanolines. Of course, some of the material parameters needed for accurate calculation of magnetic energies are unknown. It is possible, however, that researchers can use nanolines – of a sufficiently wide range of thicknesses and widths – to determine these unknown parameters.

Similarly, the model developed in this work for the low temperature properties is expected to apply to other thin film material systems, provided that the same uniaxial symmetry holds.

### 8.3 Future Work

- This work used a basic 2-factorial design – 2 nickel film thicknesses, 2 line widths, epitaxial vs. polycrystalline – to explore the triaxial model, which contains quadratic terms, whose fitting will require at least 3, not 2, points. Although the model fit may be qualitatively evaluated using just the sample data taken, error (whether due to fabrication or measurement) makes quantitative fit difficult. Additional epitaxial nanoline widths and film thicknesses would help evaluate the model applicability, and help in the extraction of valid surface and bulk quadratic terms.
- Chapter 7 featured SQUID data on two Ni thin film samples – 92 and 226 Å. Data from additional film thicknesses as well as replication of data taken for these samples will help validate the accuracy of the low temperature model.
• The bulk cubic magnetocrystalline anisotropy term was not included in the low temperature magnetic properties model developed in chapter 7. For a sample magnetized to saturation, the bulk magnetocrystalline anisotropy value at (001) is equivalent to the value at (100) and (010). An $M-H$ curve (which begins from a poorly defined, initially demagnetized state), however, does not merely measure the magnetization at saturation, but at low fields as well. The magnetization in the (001) direction should follow a different path along the overall anisotropy surface than magnetization in the (100) and (010) directions, resulting in a possible nonzero bulk anisotropy term ($M-H$ curves of different shapes). The exact effect of this $K_I$ term is unknown, but additional analytical work should establish its influence on the $M-H$ anisotropy values.

• Although magnetic force microscopy has been done on thin film samples, it has not previously been performed on Cu/Ni/Cu nanolines. The domain structure of the nanolines is expected to differ significantly from that of the thin films because of the significant change in magnetic anisotropy induced by patterning. A problem arises in doing MFM on lithographically patterned nanolines due to the uneven residual W (hard mask) left on top of the 50 Å Cu capping layer. Collaborators have patterned Cu/Ni/Cu thin films into nanolines using focused-ion-beam etching (which leaves a smooth, clean top surface for the nanolines), and will shortly explore the domain state of these structures.

• This work has focused exclusively on Cu/Ni/Cu. Other systems will likely have magnetostriction coefficients large enough to show altered anisotropies once patterned. A useful experiment in strain engineering might involve using strain, crystal direction and shape to create a zero-anisotropy patterned structure. One candidate material might be BCC-FeCo. Its magnetostriction coefficients are nearly 3x larger than that of Ni. Of course, both Co and Fe have saturation magnetizations that are also 3x larger than that of Ni, leading to magnetostatic energy values that will, correspondingly, be 9x larger than that of Ni, and crystalline anisotropy values 3x larger than Ni. It would not be likely to see anisotropy reversal in patterned structures of BCC-FeCo, given the large magnetostatic and magnetocrystalline energies, but use of buffer layers to create a
strained state for the material could result in anisotropy values that are altered from those predicted by magnetostatic and magnetocrystalline energies alone.

- The low temperature properties explored in chapter 7 are strongly dependent upon the domain structure of Cu/Ni/Cu. Previous MFM work has all been done at room temperature for this system. These MFM images had shown that the domain structure for Cu/Ni/Cu thin films is complex, especially for nickel thicknesses above 100 Å. The domain evolution of Cu/Ni/Cu as a function of temperature and film thickness can be performed by a cryo-MFM. This will help further our understanding of the magnetic properties as a function of temperature.

- The analysis assumed thin film surface terms, including magnetoelastic surface terms assumed to be uniaxial. Theoretically, however, they are triaxial, and must include all three strain terms, and 2 independent magnetoelastic surface parameters. Further analytical work should explore this, with the aim of extracting values for the two parameters. One possible method to explore the surface terms will be to decouple the strain from the thickness. It would be interesting to explore nickel films deposited on Cu$_{1-x}$Ni$_x$ buffer layers. The strain would be considerably reduced, but assuming small $x$ values, the value of the surface terms would remain roughly constant. Accurate values for the surface and magnetoelastic parameters could then be extracted.

- The OOF strain simulations assumed rectangular cross-sections. In reality, ion milling of the lines may result in erosion of the Cu capping layer, which will change the strain relief profile. Additional simulations could characterize the magnitude of this effect. Similarly, the influence to the strain state caused by the tungsten remaining on the copper surface is unknown, and could also be explored via simulations.

- The OOF simulations also assumed that the longitudinal strain was unchanged. Further, the vertical strain was inaccurate, because the simulations were based on volumetric thermal expansion strain. A different strain package, such as ANSYS, could capture the three-dimensional strain profile as a function of line width and nickel film thickness.
Appendix 1
Maple Calculation of \( B_s, K_s \)

Below is the Maple worksheet used for calculation of \( B_s \) and \( K_s \) as described in Chapter 2. The data is a compilation of the Bochi data [74] and new torque magnetometer data, fit to the second-order continuous thin film energy model via least-squares. The strains used are calculated using the phenomenological equation fit to synchrotron x-ray data for Cu/Ni/Cu continuous thin films. This equation uses Ciria’s measured values of \( D_B \). In addition, the second order magnetoelastic surface energy, \( D_s \), is calculated.

```maple
> unassign('t');
> unassign('K2');
> unassign('Db');
> unassign('Bs');
> Tvalues:=
[30,50,50,60,60,75,80,85,90,100,100,120,125,150,150,180]:

> K2effvalues:=
[0.28,0.42,0.67,0.65,0.65,0.7,0.72,0.58,0.5,0.58,0.4,0.2,-
0.27,0.1,-0.37]:

> e:=.026*(27/t)^(2/3):
> MS:=-2*Pi*485^2:
> K:=2*Ks/(t*10^(-8)):
> Beff:=(620000000*2.28)+(2*(-Bs/(t*10^(-8)))):
> ME1:=Beff*e:
> Db:=-1850000000:
> Di:=((3.5241*10^(-8))/4)*((1-2*1.28+(1.28)^2)/(3+2*1.28-
(1.28)^2))*Db:
>
> Deff:=(Db-(2*Di/(t*10^(-8)))):
> ME2:=Deff*e^2:
> K2efft:=(t*10^(-8))*(MS+ME1+K+ME2);
```
K_{eff}t = \frac{1}{100000000} \left[ \frac{-470450\pi + 0.026\left(141360000 - \frac{200000000B_s}{t}\right)27^{2/3}\left(\frac{1}{t}\right)^{2/3}}{t^2} + \frac{200000000K_s}{t^2} + 0.018252\left(-185000000 + \frac{65169250.32}{t}\right)27^{1/3}\left(\frac{1}{t}\right)^{4/3} \right]

> with(stats):
> fit[leastsquare][[t,K2],K2=(t*10^(-8))*(MS+ME1+K+ME2),{Ks,Bs}]([[30,50,50,60,60,75,80,85,90,100,100,120,125,150,150,180],[0.28,0.42,0.67,0.65,0.65,0.7,0.72,0.58,0.5,0.58,0.4,0.2,-0.27,0.1,-0.37]]);

K_{eff}t = \frac{1}{100000000} \left[ \frac{-470450\pi + 0.026\left(141360000 - \frac{3591238566}{t}\right)27^{2/3}\left(\frac{1}{t}\right)^{2/3}}{t^2} + \frac{101156797.4}{t^2} + 0.018252\left(-185000000 + \frac{65169250.32}{t}\right)27^{1/3}\left(\frac{1}{t}\right)^{4/3} \right]

Thus, B_s is calculated to be -17.95 erg/cm³ and K_s is calculated to be 0.506 erg/cm³.
Appendix 2

Surface Magnetoelastic Energy

Like the second-order magnetoelastic energy terms, the surface magnetoelastic energy terms may need to be given in generalized forms when the system symmetry is reduced. Du Trémolet de Lacheisserie proposed the general equation for a magnetic thin film sandwiched between two nonmagnetic layers [75]:

\[
E = \frac{1}{2} \mu_0 M_s^2 - \frac{2k^s}{t_f} \left[ \alpha^2 - \frac{1}{3} \right] + B_0 + \left( \frac{\sqrt{2} b_{1}^{a_s}}{t_f} \right) (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) + \left( B_{12}^{y_s} + \frac{2b_2^{a_s}}{t_f} \right) \left[ \varepsilon_{zz} - \frac{\varepsilon_{yy} + \varepsilon_{xx}}{2} \right] \left[ \alpha_3^2 - \frac{1}{3} \right] 
\]

\[
+ \left( B_{12}^{y_s} + \frac{2b_2^{a_s}}{t_f} \right) \left[ \frac{1}{2} (\varepsilon_{xx} - \varepsilon_{yy}) (\alpha_1^2 - \alpha_2^2) + 2\varepsilon_{xy} \alpha_1 \alpha_2 \right] + \left( B_{12}^{y_s} + \frac{2b_2^{a_s}}{t_f} \right) \left[ \varepsilon_{zz} + \varepsilon_{yy} \alpha_3 + \varepsilon_{xx} \alpha_3 \right] + E_{\text{elastic}} 
\]

\[
B_0 = \frac{1}{3} \left( B^{a_s} + \frac{2b_0^{a_s}}{t_f} \right) (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) + \frac{\sqrt{2}}{3} \left( \frac{2b_0^{a_s}}{t_f} \right) \left[ \varepsilon_{zz} - \frac{\varepsilon_{yy} + \varepsilon_{xx}}{2} \right]
\]

Removing the shear and angle-independent terms and assuming biaxial strain (\(\varepsilon_{xx} = \varepsilon_{yy}\)) gives:

\[
E = \frac{1}{2} \mu_0 M_s^2 - \frac{2k^s}{t_f} \left[ \alpha^2 - \frac{1}{3} \right] + \left( B_{12}^{y_s} - \frac{2B_{eff}^{y_s}}{t_f} \right) \varepsilon_{xx} \alpha_3^2
\]

\[
B_{eff}^{y_s} = b_2^{a_s} \left( 1 + \frac{2c_{12}}{c_{11}} \right) - \sqrt{2} b_1^{a_s} \left( 1 - \frac{c_{12}}{c_{11}} \right)
\]

This \(B_{eff}^{y_s}\) is the familiar thin film surface magnetoelastic constant, but it is now shown to contain two surface magnetoelastic energy constants, \(b_2^{a_s}\) and \(b_1^{a_s}\). For a non-biaxial strain case we have:

\[
E = \frac{1}{2} \mu_0 N_s M_s^2 - \frac{2k^s}{t_f} \left[ \alpha^2 - \frac{1}{3} \right] + B_{12}^{y_s} \left[ \varepsilon_{xx} \alpha_1^2 + \varepsilon_{yy} \alpha_2^2 + \varepsilon_{zz} \alpha_3^2 \right]
\]

\[
+ \frac{b_1^{a_s}}{t_f} (\varepsilon_{xx} - \varepsilon_{yy}) \alpha_1^2 - \frac{b_2^{a_s}}{t_f} (\varepsilon_{xx} - \varepsilon_{yy}) \alpha_2^2 + \left[ \frac{\sqrt{2} b_1^{a_s}}{t_f} (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) + \frac{2b_2^{a_s}}{t_f} \varepsilon_{zz} - \frac{\varepsilon_{yy} + \varepsilon_{xx}}{2} \right] \left[ \alpha_3^2 - \frac{1}{3} \right]
\]

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Now the surface magnetoelastic term contains three unknown constants, $b_{2}^{a,s}$, $b_{1}^{a,s}$ and $b^{y,s}$ and is no longer confined only to the $a_{3}$ direction, but also to the $a_{2}$ and $a_{1}$. Extending this to the sides of the nanolines, by analogy, gives:

$$E_{sides} = \left[ \frac{1}{2} \mu_{0} N_{y} M_{s}^{2} - \frac{2k_{Ni-NiO}^{w}}{w} \right] \alpha_{2}^{2} + b^{y,s} \left[ e_{xx} \alpha_{1}^{2} + e_{yy} \alpha_{2}^{2} + e_{zz} \alpha_{3}^{2} \right]$$

$$+ b^{y,s} \left( e_{xx} - e_{zz} \right) \alpha_{1}^{2} - b^{y,s} \left( e_{xx} - e_{zz} \right) \alpha_{3}^{2} + \left[ \sqrt{2}b_{1}^{a,s} \left( 1 + \frac{e_{yy} + e_{zz}}{e_{xx}} \right) + 2b_{2}^{a,s} \left[ \frac{2e_{yy} - e_{zz}}{2e_{xx}} + 1 \right] \right] \alpha_{3}^{2}$$

These additional side surfaces introduce not only new magnetocrystalline terms, but also new magnetoelastic terms. Though these terms are small (due to $w \ll t_{Ni}$), they are included for completeness.

The modified $B_{5}$ terms for the top and side surfaces are given below:

$$B_{5}^{s}_{top/bottom} = b^{y,s} \left( 1 - \frac{e_{yy}}{e_{xx}} \right) \alpha_{1}^{2} - b^{y,s} \left( 1 - \frac{e_{yy}}{e_{xx}} \right) \alpha_{2}^{2} + \left[ \sqrt{2}b_{1}^{a,s} \left( 1 + \frac{e_{yy} + e_{zz}}{e_{xx}} \right) + 2b_{2}^{a,s} \left[ \frac{2e_{yy} - e_{zz}}{2e_{xx}} + 1 \right] \right] \alpha_{3}^{2}$$

$$B_{5}^{s}_{sides} = b^{y,s} \left( 1 - \frac{e_{zz}}{e_{xx}} \right) \alpha_{1}^{2} - b^{y,s} \left( 1 - \frac{e_{zz}}{e_{xx}} \right) \alpha_{3}^{2} + \left[ \sqrt{2}b_{1}^{a,s} \left( 1 + \frac{e_{yy} + e_{zz}}{e_{xx}} \right) + 2b_{2}^{a,s} \left[ \frac{2e_{yy} - e_{zz}}{2e_{xx}} + 1 \right] \right] \alpha_{2}^{2}$$

For nanoline strain states where $e_{yy} \approx e_{xx}$, the $\alpha_{1}$ and $\alpha_{2}$ terms in the top and bottom surface equation will approach zero. The $\alpha_{3}$ term will then be the same as the $B_{5}$ term for the continuous thin films. Since the 3 $b$ parameters are all unknown, as a first order approximation, the continuous thin film value of $B_{5}$ will be used here.
Appendix 3

Maple Calculation of Demagnetization Factors

Below is the Maple worksheet used for calculation of \( N_Y \) and \( N_Z \) as described in Chapter 2. For ease in computation, averages were performed between 0 and \( t/2, w/2 \), then multiplied by 4 to obtain the correct value. For a thin film, the demagnetization factor, \( N_Z \), should be \( 4\pi \) (in cgs units). This is replicated in the following example computation, below, where the aspect ratio (width to thickness) is a million to one:

```maple
> unassign('t');
> unassign('w');
> unassign('y');
> unassign('z');
> t:=1;
    t := 1
> w:=1000000;
    w := 1000000

> theta11:=(arctan(z/y));
> theta12:=(arctan(z/abs((y-w))));
> theta21:=(arctan(abs((z-t)/y)));
> theta22:=(arctan(abs((z-t)/(y-w))));

> Fy1:=int(-1*(Pi-theta11-theta12),y=0..w/2):
> Fyz1:=int(Fy1,z=0..t/2):
> Fy3:=int(-1*(Pi-theta21-theta22),y=0..w/2) assuming y > 0:
> Fyz3:=int(Fy3,z=0..t/2):
> Nz:=8*evalf(Fyz1+Fyz3)/t/w;
    Nz := -12.56000000

> Fy2:=int(-1*(theta11+theta21),y=0..w/2):
> Fyz2:=int(Fy2,z=0..t/2):
> Fy4:=int(-1*(theta12+theta22),y=0..w/2):
> Fyz4:=int(Fy4,z=0..t/2):
> Ny:=8*evalf(Fyz2+Fyz4)/t/w;
    Ny := 0.008008000000

> NZ:=evalf(Nz/4/Pi);
    NZ := -0.9994930424

> NY:=evalf(Ny/4/Pi);
    NY := 0.0006372563920
```

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Appendix 4

OOF Simulation Details

The rectangular block images were created in Microsoft Powerpoint using monochromatic rectangular blocks scaled to the appropriate size. These images were saved as jpg files. The jpg files were opened in Gimp\(^{22}\). To remove the white “empty” spaces, the rectangular blocks were selected using the “fuzzy select” wand. All other elements were removed by cropping the image. The images were then saved in ppm format.

In PPM2OOF 1.1.28, the ppm files were opened. The rectangular blocks were selected, and materials properties (including a thermal expansion coefficient, \(\alpha\), of 0.026 – not the value of nickel’s thermal expansion coefficient, but chosen to give the desired misfit strain of 0.026 with a temperature change of 0\(^\circ\)) were assigned. Then a mesh was created, and the resulting image was saved as a goof file, ready for use in OOF simulations.

Table 14: PPM2OOF Materials Properties, Aspect Ratio Cases

<table>
<thead>
<tr>
<th>Material Type</th>
<th>Gray Level</th>
<th>Plane Strain</th>
<th>Orientation</th>
<th>Young</th>
<th>Poisson</th>
<th>Alpha</th>
<th>Anisotropy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cubic</td>
<td>0.5</td>
<td>False</td>
<td>0</td>
<td>225</td>
<td>0.297</td>
<td>0.026</td>
<td>1</td>
</tr>
</tbody>
</table>

Table 15: PPM2OOF Materials Properties, Cu/Ni/Cu Cases

(all cases: Orientation =0 and Anisotropy =1)

<table>
<thead>
<tr>
<th>Layer</th>
<th>Material Type</th>
<th>Gray Level</th>
<th>Plane Strain</th>
<th>Young</th>
<th>Poisson</th>
<th>Alpha</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>Cubic</td>
<td>0.8</td>
<td>False</td>
<td>225</td>
<td>0.297</td>
<td>0.026</td>
</tr>
<tr>
<td>Cu</td>
<td>Cubic</td>
<td>0.2</td>
<td>False</td>
<td>129</td>
<td>0.3435</td>
<td>0</td>
</tr>
</tbody>
</table>

The Young’s modulus values in Table 15 are in GPa, and correspond to the moduli of nickel and copper. The simulation’s resulting stress values are in GPa.

Due to the complexity of the Cu/Ni/Cu structures, a simple mesh could not be used. Instead, an adaptive mesh was used. Each adaptive mesh was at least 10-25 elements high and 50-75 elements wide. Following creation of the initial grid mesh, the system was repeatedly annealed.

\(^{22}\) http://www.gimp.org/ “GIMP is the GNU Image Manipulation Program. It is a freely distributed piece of software for such tasks as photo retouching, image composition and image authoring.”
In an anneal step, small displacements are made to the mesh and tested to see whether the displacements improve the homogeneity (presence of more than one material) and shape uniformity of the elements. This mesh was annealed for at least 20 iterations at first, then the \textit{swap\_worst} procedure was applied. This sequence was repeated (with annealings of smaller step size, from the start size of 1 down to 0.1 as needed, and setting increasing preference for element homogeneity from the starting value of $\alpha = 0.33$ increasing to as much as 0.9). If needed, the smooth function was applied, as was \textit{Refine – Interface} (subdivides blocks at the interface) or \textit{Refine – E} (subdivides high-“energy” blocks). Using increasingly stringent conditions, annealing continued until the interface was smooth (to the limits of the pixel resolution).
Appendix 5
Fabrication Recipes and Parameters

Table 16: DC magnetron sputtering parameters

<table>
<thead>
<tr>
<th>Target Power</th>
<th>DC Bias (Voltage)</th>
<th>DC Bias (Current)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 W</td>
<td>169 V</td>
<td>0.62 A</td>
</tr>
</tbody>
</table>

Table 17: Ion beam sputter deposition parameters

<table>
<thead>
<tr>
<th></th>
<th>Voltage (V)</th>
<th>Current (A)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cathode</td>
<td>5.8</td>
<td>6.9</td>
</tr>
<tr>
<td>Discharge</td>
<td>39.9</td>
<td>1.44</td>
</tr>
<tr>
<td>Beam</td>
<td>1002</td>
<td>.035</td>
</tr>
<tr>
<td>Acceleration</td>
<td>200</td>
<td>.0013</td>
</tr>
<tr>
<td>Argon Flow Rate</td>
<td></td>
<td>5 sccm</td>
</tr>
<tr>
<td>Main Gate Valve Position</td>
<td></td>
<td>Open</td>
</tr>
</tbody>
</table>

Table 18: Reactive ion etch recipe parameters

<table>
<thead>
<tr>
<th>Etch Step</th>
<th>Pressure (mTorr)</th>
<th>Voltage (V)</th>
<th>Power (W)</th>
<th>Gas(es)</th>
<th>Etch Rate (Å/min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>7</td>
<td>200</td>
<td>~110</td>
<td>10 sccm CHF₃</td>
<td>170</td>
</tr>
<tr>
<td>ARC</td>
<td>7</td>
<td>250</td>
<td>~130</td>
<td>5 sccm O₂, 10 sccm He</td>
<td>770</td>
</tr>
<tr>
<td>W</td>
<td>7</td>
<td>100</td>
<td>~50</td>
<td>3 sccm O₂, 19.5 sccm CF₄</td>
<td>70</td>
</tr>
</tbody>
</table>

Table 19: Reflectivity model parameters

<table>
<thead>
<tr>
<th>Material</th>
<th>Index of Refraction – Real portion</th>
<th>Index of Refraction – Imaginary portion</th>
<th>Thickness (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>1</td>
<td>0</td>
<td>Infinite</td>
</tr>
<tr>
<td>PFI-88 Resist</td>
<td>1.79</td>
<td>0.02</td>
<td>2000</td>
</tr>
<tr>
<td>SiO₂</td>
<td>1.48</td>
<td>0</td>
<td>300</td>
</tr>
<tr>
<td>BARLi ARC</td>
<td>1.55</td>
<td>0.14</td>
<td>TBD</td>
</tr>
<tr>
<td>SiO₂</td>
<td>1.48</td>
<td>0</td>
<td>300</td>
</tr>
<tr>
<td>W</td>
<td>2.99</td>
<td>2.56</td>
<td>300</td>
</tr>
<tr>
<td>Cu</td>
<td>1.34</td>
<td>1.81</td>
<td>50</td>
</tr>
<tr>
<td>Ni</td>
<td>1.69</td>
<td>1.99</td>
<td>69 or 206</td>
</tr>
<tr>
<td>Cu</td>
<td>1.34</td>
<td>1.81</td>
<td>2000</td>
</tr>
<tr>
<td>Si</td>
<td>4.68</td>
<td>2.03</td>
<td>Infinite</td>
</tr>
</tbody>
</table>
Table 20: Ion miller process parameters

<table>
<thead>
<tr>
<th><strong>Parameter</strong></th>
<th><strong>Setting</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas Flow</td>
<td>30 sccm Ne</td>
</tr>
<tr>
<td>Base Pressure</td>
<td>$3 \times 10^{-6}$ Torr</td>
</tr>
<tr>
<td>Process Pressure</td>
<td>$3 \times 10^{-4}$ Torr</td>
</tr>
<tr>
<td>Beam</td>
<td>500 V</td>
</tr>
<tr>
<td>Acceleration</td>
<td>150 V, &lt; 5 mA</td>
</tr>
<tr>
<td>Discharge</td>
<td>49 V, 1.9 A</td>
</tr>
<tr>
<td>Cathode</td>
<td>14.5 A</td>
</tr>
<tr>
<td>Neutralizer</td>
<td>50 mA</td>
</tr>
<tr>
<td>Beam Current</td>
<td>40 mA</td>
</tr>
</tbody>
</table>
Appendix 6
Torque Measurement Data Processing

Due to the low signal-to-noise ratio present in many of the patterned sample measurements, additional processing was needed to extract the parameters from the torque magnetometer measurements. The torque magnetometer raw data consists of two curves: the zero field and the high field (13,500 Oe) curves. To obtain the magnetic signal absent the effects of gravity, the zero field curve is automatically subtracted from the high field curve. Any noise present in the zero field curve is directly transferred to the high field data. For high signal samples, this noise is negligible, but can be a substantial contribution of additional error for low signal samples.

Figure 109 shows a zero field curve for one measurement. Significant noise is present, due to measurement error.

![Figure 109: Zero field curve (blue) and simulated fit (black), 69 Å epitaxial 200 nm period nanolines, XY direction](image)

A simulated fit of the zero field curve was extracted in Origin using the equation:

\[ L = P_1 \sin(\theta - P_2) - P_3 \]
The noise is reduced by replacing the original zero field curve with the simulated fit (shown in black). Each simulated zero field curve was then subtracted from its corresponding raw 13,500 Oe curve to generate a lower-noise torque curve.

Further noise reduction was gained by averaging multiple samples. As shown in Figure 110, individual curves contain significant noise. No individual measurement curve resembles a \( \sin(2\theta) \) or \( \sin(4\theta) \) curve, as predicted for uniaxial and cubic anisotropy samples.

**Figure 110:** 4 Individual measurement curves, 69 Å polycrystalline, 500 nm period nanolines, XZ direction

Once these measurement curves were averaged, however, as shown in Figure 111, the resulting curve has two distinct peaks, making accurate modeling possible.
These curves were fit in Origin to the following equation:

\[ L = P_1 \sin 2\theta + P_2 \cos 2\theta + P_3 \sin 4\theta + P_4 \cos 4\theta + P_5 \sin 6\theta + P_6 \cos 6\theta + P_7 \]

with:

<table>
<thead>
<tr>
<th>$P_1$</th>
<th>$P_2$</th>
<th>$P_3$</th>
<th>$P_4$</th>
<th>$P_5$</th>
<th>$P_6$</th>
<th>$P_7$</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>-0.147</td>
<td>-0.111</td>
<td>0.011</td>
<td>0.029</td>
<td>0.002</td>
<td>0.009</td>
<td>0.077</td>
<td>0.87</td>
</tr>
</tbody>
</table>

The fit contains significant cosine terms, and the overall correlation, $R^2=0.87$, is mediocre, but this represents an improvement over the original data, allowing good confidence in the extracted sine parameters.
References