Atomically Thin
Magnetic Microstructures

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Christian Stamm
Dipl. Phys. ETH
born on January 3rd, 1970
citizen of Winterthur (ZH) and Thayngen (SH)

accepted on the recommendation of

Prof. Dr. D. Pescia, examiner
Prof. Dr. H.-C. Siegmann, co-examiner

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Abstract

In this thesis the magnetic properties of atomically thin microstructures are investigated. By drastically decreasing the size of a ferromagnet along one space dimension a two-dimensional particle results, which exhibits new magnetic behaviour.

Domains in ferromagnetic bodies are the result of two competing interactions. The magnetostatic self-energy is reduced by the formation of magnetic domains with opposite magnetization. On the other hand exchange and anisotropy energy increase with each additional domain wall. The reason why in three-dimensional ferromagnets domains occur is that the magnetostatic energy is a volume energy, whereas the wall energy is a surface energy. Therefore, only small magnets are expected to be in a single-domain state. This is not necessarily true for two-dimensional magnets, where two different situations must be considered.

In the first case, the magnetization is oriented parallel to the film plane. The physical systems investigated in this thesis are Co on a Cu(001) surface and Fe on a W(110) surface. The main result for in-plane magnetized structures is that they do not exhibit domains, irrespective of their size and shape. The smallest particle with remanent magnetization at room temperature was 130 nm wide and 2 atomic layers thick. The shape anisotropy of stripes with in-plane aspect ratio $R = 80$ is negligible compared to the crystalline anisotropy. The mutual interaction between atomically thin magnets is too small to determine the magnetic configuration.

In the second case, the magnetization stands perpendicular to the film plane, realized in this work by a few layers of Fe on Cu(001). Continuous films exhibit a striped domain configuration with local orientational order. When confining its lateral size on the micron scale a transition becomes visible, where magnetic particles below a critical size become single-domain. The phase transition of a perpendicular magnetized thin film to the paramagnetic state is found to happen in two steps, which is reminiscent of a two-dimensional melting process.
Zusammenfassung

In dieser Arbeit werden die magnetischen Eigenschaften von atomar dünnen Mikrostrukturen untersucht. Durch die starke Verkleinerung der Abmessung eines Ferrismagneten entlang einer räumlichen Dimension zeigt das so entstandene zweidimensionale Teilchen neuartiges magnetisches Verhalten.


In der zweiten Situation steht die Magnetisierung senkrecht auf die Ebene, was in dieser Arbeit durch wenige Lagen Fe auf einer Cu(001) Oberfläche realisiert wurde. Kontinuierliche Filme zeigen in Streifen angeordnete Domänen mit lokaler Ordnung der Orientierung. Durch Eingrenzung auf Mikrometer grosse Bereiche wird ein Übergang sichtbar: Magnetische Teilchen unterhalb einer kritischen Größe werden eindomänig. Der Phasenübergang von senkrecht magnetisierten Filmen findet in zwei Schritten statt, dies erinnert an den Prozess des zweidimensionalen Schmelzens.
Chapter 1
Introduction

A ferromagnetic body contains permanent magnetic dipoles that are spontaneously oriented parallel to a common axis even in the absence of an external field. It is not the magnetostatic interaction between two dipoles that produces the parallel alignment; a quantum mechanical effect known as exchange interaction is needed to align the dipoles and thus make a substance ferromagnetic. Furthermore, the magnetic dipole of an electron originates from its spin, a quantity not present in classical physics.

Within the last decades much work has been done in the field of two-dimensional magnetism. A thin magnetic film — only a few atoms in thickness — is produced by molecular beam epitaxy and analysed in an ultra-high vacuum environment. Because of the small amount of magnetic material, surface sensitive measurement methods are needed. They are found in techniques using light and electrons, both polarized to be sensitive to the spin direction.

A great challenge is presented by the desire to construct magnetic objects of controlled size, shape, crystal structure, and composition on the nanometre scale. One possible technique is to confine an atomically thin film laterally. This can be achieved by molecular beam epitaxy with a suitable diaphragm in front of the substrate crystal which has specially designed holes defining the geometry of the magnetic structure. The resulting particle has well defined crystal structure and willingly designed size and shape and a thickness of only a few atomic layers (AL). It is an ideal model system to test and enlarge our knowledge of magnetism.

A common observation is that for a large ferromagnetic crystal the homogeneously magnetized state is unstable against domain formation. The energy due to the magnetic field is minimized by the formation of magnetic domains — regions of uniform magnetization along an individual direction — trying to keep the magnetic field confined to the body. What will the properties of two-dimensional magnets be regarding magnetic domains? There are two different geometries to consider: the magnetization vector can either lie in the film plane, or it can stand perpendicular to it. Which of these situations occurs for a certain magnetic system is dependent on the physical parameters of the materials as well as on the size of the magnet. Calculations of the magnetic energy suggest that only perpendicular magnetized bodies will exhibit domains, whereas the magnetization parallel to the plane will be uni-
form throughout the whole body, irrespective of their size. This is a new effect not present in three-dimensional magnetism.

Today, one main application of ferromagnetism is the storage of information. The two remanent states — positive and negative magnetization, magnetic north and magnetic south — code the information in a binary — “yes” or “no”, “1” or “0” — manner. For this application, miniaturization and long-term conservation of the information are essential. At least two questions arise: How small can the magnetic particles be made without loosing long-term stability against thermal fluctuations? And how near can these magnets be placed together while they can still be treated as independent magnetic particles?

From the scientific point of view it is challenging to construct a small magnet on the nanometre scale, too. To what extent does a two-dimensional magnetic particle behave differently from a three-dimensional one? Where will be the ultimate limit when ferromagnetic long range order breaks down? Will new quantum mechanical effects appear approaching atomic dimensions? Another challenge is the need for new measurement methods able to access the nanomagnets on an atomic scale, like magnetic force microscopy and spin-dependent tunnelling.

The outline of this thesis is as follows: in chapter 2 the experimental apparatus with the fabrication of magnetic microstructures and the SEMPA measurement technique is introduced. Chapter 3 discusses theoretically the various energies in a ferromagnetic body, domain formation, stray fields, and the stability of the remanent magnetization against thermal fluctuations. The experimental results start in chapter 4 with the description of in-plane magnetized two-dimensional microstructures of Co on a Cu(001) surface and Fe on a W(110) surface. Finally, in chapter 5 measurements on magnetic structures of Fe/Cu(001) with perpendicular magnetization are shown.
Chapter 2

The Experimental Apparatus

2.1 The Ultra-High Vacuum System

Experiments with atomically thin samples need to be done in an ultra-high vacuum environment (UHV, base pressure below $1 \times 10^{-10}$ mbar) in order to prevent contamination and oxidation. The apparatus used for this study consists of a preparation chamber and a measurement chamber. Sample transfers between the chambers always take place under UHV conditions.

The preparation chamber is equipped with sample preparation tools (ion sputter gun and annealing/heating stage) and surface analysis tools (Auger electron spectrometer and LEED). A load-lock permits the introduction of a new sample within one hour. The most important and not commercially available part of the preparation chamber is the evaporator for microstructures, which is sketched in the next chapter.

The main part of the measurement chamber is the scanning electron microscope (SEM). A beam of primary electrons is focused on the specimen. Images are produced by rasterizing the beam across the region of interest and detecting the amount of secondary electrons emerging from the sample. In addition, the secondary electrons are spin analysed in a Mott detector (SEM with polarization analysis: SEMPA). The measurement chamber can also hold a special constructed scanning tunnelling microscope (STM). Then it is possible to take SEMPA and STM images of the same spot on the sample.

2.2 Producing Microstructures

Magnetic microstructures are fabricated by placing a diaphragm between evaporation source and sample. The setup is sketched in figure 2.1, see also the dissertation by Ramsperger [1]. The mask consists of a $1 \mu$m thick titanium foil mounted on a stainless steel holder. Through this thin foil micrometre sized holes are etched using a focused ion beam machine (either at the PSI in Villigen, Switzerland, or at the NRIM in Tsukuba, Japan). Only these holes let magnetic materials get through to
the sample surface, thus defining the geometry of the microstructure. The smallest holes are about 10nm in diameter.

Figure 2.1: Schematic view of the evaporator setup. The mask can be positioned in front of the sample with piezo inertial sliders to minimize the distance D.

Structures below one micron demand for a highly parallel molecular beam. This is the reason for the rather large distance $L$ between source and sample. Another necessity for sharp structures is a minimal distance $D$ between mask and sample. It can be achieved with the help of piezo inertial sliders which drive the mask to the right position. The mask is manipulated very close to the sample surface, actually until making contact. The resulting thickness profile can be simulated with Monte Carlo techniques. The best agreement to the measurement shown in figure 2.2 was with $D = 20 \mu m$. However, this value varies from sample to sample, which is experienced by measuring “accidentally unsharp evaporated structures”.

Figure 2.2: Scanning Electron Microscope cross section through a 63nm wide hole in a mask (white dots) and through a Co island grown through this hole (black dots). The smooth line is a Monte Carlo simulation of the growth assuming a mask to sample distance of $D = 20 \mu m$.

It is also possible to fabricate wedge-like structures by choosing a large mask to sample distance of $D = 10 \text{mm}$. The result is a micro-wedge where the coverage...
ranges from zero to the nominal thickness within a length of 70\textmu m. Thus it becomes easy to do thickness dependent measurements with one SEMPA image.

2.3 Measuring with the SEMPA

The microscope used in these studies is a Hitachi 4100S UHV field emission SEM. Accelerator voltages for the primary electron beam were chosen ranging from 2 – 10 kV. The achievable resolution in our setup is about 10 nm. The secondary electrons emitted from the sample surface are detected by a channeltron-type electron multiplier and simultaneously collected with electrostatic lenses for spin-polarization analysis in a Mott detector (SEMPA). Magnetic coils are used to magnetize the sample, however the low-energy secondary electrons do not allow a field during the measurements. A \mu-metal shield in the SEMPA chamber and around the electrostatic lenses screens external magnetic fields.

SEMPA was first introduced by Koike and Hayakawa in 1984 [2,3]. In the meantime several others have built such microscopes with various types of spin detectors [4,5,6]. Our Mott detector was built in a collaboration with the St. Petersburg Technical University [7]. Secondary electrons are collected near the sample surface by a system of electrostatic lenses. A 90° deflection selects electrons with energy of about 2 – 6 eV which is, according to Oepen and Kirschner [5], the energy range with the highest polarization. Afterwards the electrons are accelerated by 60 kV (“medium voltage”) onto a 80 nm thick gold foil with four detectors counting the electrons scattered backwards. Two electron polarization components are measured simultaneously, one parallel and one perpendicular to the sample surface. After rotating the sample 90° around the surface normal one can measure the remaining in-plane direction to finally get the magnetization vector on each point of the sample. The experimental setup is sketched in figure 2.3.

If the incident beam on the gold foil is polarized (“up” or “down”), there will be an asymmetry in the detector counts (detector “left” and “right”). The polarization of the secondary electrons can be calculated by the number of counts \( N^+ \) and \( N^- \) in two detectors lying opposite to each other. The constant \( S \) is called Sherman factor and is determined only by the detector features.

\[
P = \frac{1}{S} \frac{N^+ - N^-}{N^+ + N^-}
\]

The polarization of the secondary electrons emitted from the sample is proportional to the magnetization. The polarization images are displayed using a gray-scale where white and black stand for positive and negative polarization, gray corre-
The SEMPA setup: left the SEM column with the sample crystal tilted 45° to the horizontal, right the Mott detector. Secondary electrons are emitted from the sample surface as the beam is scanned over the sample. They are collected by electrostatic lenses, deflected 90° for energy selection and accelerated to 60 keV. Four SSB detectors count the back-scattered electrons.

This corresponds to zero polarization. In addition to the two components of the magnetization the Mott detector gives an image with topographic information, calculated as the sum of all detector counts, see figure 2.4.

The acquisition time per pixel in SEMPA measurements was typically between 20 and 100 ms, equivalent to 2000 – 10000 counts per pixel (at 10^5 counts per second). A typical image size is 128×128 pixels, which takes about 7 minutes (at 25 ms/pixel) to acquire.

### 2.4 The SEMPA-STM

The standard SEM manipulator in the measurement chamber can be exchanged with a special miniature STM setup. The sample surface still has a 45° angle to the horizontal, so it is possible to do SEMPA measurements as well. The photograph in figure 2.5 shows the STM setup.

The STM piezo was gauged with a Si(111) sample. Heating to 1400 K for about two minutes gave a beautiful 7x7 reconstruction, see figure 2.6. Every seventh atom on the hexagonal surface is missing, the distance between the holes is 2.688 nm.
Figure 2.4: SEMPA measurements of a “butterfly” structure. Left column imaged with magnification 10’000, right column is a close-up with magnification 50’000. The scale bars beneath the first image row applies to all three images of the corresponding column. Topography, in-plane polarization along the vertical image axis and out of plane polarization are displayed. The polarization range is ± 30%. In the topography image intensity flickering is visible, this is a side effect emerging from the field emission cathode and cancels when calculating the polarization images.

As the construction of the STM was finished only recently, only few experiments have been possible by now. Nevertheless, the STM can be used to check the sharpness of the evaporated microstructures. The image in figure 2.7 displays a scan over a “butterfly” structure. The vertical lines are steps of the Cu substrate, which has a miscut of 2° with respect to the (001) plane. The magnification of the centre shows a gap of about 150nm width, see the line section in figure 2.7.
Figure 2.5: Photograph of the SEMPA-STM. The tip points towards the sample which is mounted on a 5×5×20mm large holder in the middle (with two holes on each side). The whole construction hangs on three springs.

Figure 2.6: STM images of 7×7 reconstructed Si(111). Image size is 19.6nm by 22.8nm at left and 5.9nm by 6.8nm at right. The images were used to calibrate the lateral sensitivity of the piezo scanner. Tunnelling parameters: $U = 800$mV, $I = 0.5$nA.
Figure 2.7: STM Measurements of a Co “butterfly” structure with thickness 7.8AL on a Cu(001) substrate with 2° miscut. Top left image is 6.9μm wide, right is a 1.3μm wide magnification of the centre. Tunnelling parameters: 0.8V voltage and 0.5nA current. Below the right image two line sections along the horizontal direction are plotted with vertical offset to each other. The middle islands is about 450nm wide, the gap is about 150nm (both measured at half maximum). The island rises from zero to maximum coverage within 120nm.
Chapter 3

Theory

3.1 Energies in Ferromagnetic Solids

The configuration of a ferromagnetic body is determined by the minimization of its energy $E$. Assuming that no magnetic field is applied, the total energy is given by the sum of the exchange energy $E_{ex}$, the magnetocrystalline or anisotropy energy $E_a$, and the magnetostatic energy $E_m$:

$$E = E_{ex} + E_a + E_m$$

Sometimes the magnetoelastic energy is also added, like in Kittel’s review [8]. However, it can often be included in the anisotropy term and is omitted here. In the following, each of these energies will be discussed shortly. Later on, the scaling properties of the energies are exploited to estimate the dependence of the magnetic configuration on the geometry of the body.

3.1.1 The Exchange Energy

Suppose to take two bound electrons with coulomb interaction and calculate their quantum-mechanical energies. A physical realisation is the $\text{H}_2$ molecule. To satisfy the Pauli exclusion principle only antisymmetric wave functions are accepted as solutions. Two different solutions exist, one for parallel spin and one for antiparallel spin of the two electrons. Their energy difference is called exchange splitting, it is a matrix element between two states that have their two electron coordinates exchanged. Only regions where the electron wave functions overlap contribute to the exchange integral, making the interaction short-ranged. In the example of the $\text{H}_2$ molecule antiparallel alignment of the spins has lower energy. The conclusion is that a spin-independent coupling together with Pauli’s principle can lead to an interaction which is sensitive to the spin.

In ferromagnetism the exchange interaction favours parallel alignment of neighbouring spins. The energy difference can be described with the following formula,
where $J_{ij}$ are the exchange constants of the spins $S_i$ and $S_j$. If only nearest neighbours interaction is regarded, then $J$ is constant and the sum becomes much easier, where $\phi_{ij}$ denotes the angle between spin $i$ and $j$ (see for example Ashcroft and Mermin, chapter 32 [9]):

$$E_{ex} = -\sum_{ij} J_{ij} S_i \cdot S_j = -JS^2 \sum_{\text{neighbours}} \cos \phi_{ij}$$

### 3.1.2 The Magnetocrystalline Energy

The previous formula for the exchange energy does not account for the various symmetries found in crystals, which act via the $\mathbf{L} \cdot \mathbf{S}$ coupling on the spin system. This is corrected by introducing the magnetocrystalline energy, often called crystalline anisotropy. A ferromagnetic body will be magnetized along an easy magnetization axis, any deviation from this axis costs energy proportional to the volume of the body. The magnetocrystalline energy density can be expressed as a series in even powers of $\cos(\theta)$, where $K_i$ stand for the anisotropy constants and $\theta$ is the angle between axis and magnetization (shown up to fourth order):

$$\frac{E_a}{V} = K_1 \cos^2 \theta + K_2 \cos^4 \theta$$

The above formula is a prototype for uniaxial anisotropy. Examples are ultrathin films of Fe on W(110) with in-plane magnetization along [1\bar{1}0] and Fe on Cu(001) with easy axis perpendicular to the film plane.

In thin magnetic films the presence of a surface changes the local electronic structure [10]. This leads via the spin-orbit coupling to another symmetry-induced anisotropy, called surface anisotropy, with an energy contribution proportional to the size of the surface. Depending on its sign and strength, it can lead to perpendicular magnetization in atomically thin films like in Fe/Cu(001), although the magnetostatic energy favours a magnetization parallel to the film plane. According to calculations by Bruno the anisotropy of the magnetic moment can be as large as $0.1 \mu_B$ [11].

Surface anisotropies also affect the anisotropy constants in the film plane. An experimentally important model system is a thin film of Co on a Cu(001) surface. For this system, Krams et al. [12] measured the four-fold in-plane anisotropy as a function of film thickness. It can be divided into a volume part and a surface part, their values are $K_p = -2.3 \times 10^6 \text{erg/cm}^3 = -1600 \text{Oe} \cdot M_S$ for the volume anisotropy and $k_p = 0.034 \text{erg/cm}^2 = 1400 \text{Oe/ÅL} \cdot M_S$ for the surface anisotropy. The angle
\( \phi \) defines the in-plane orientation with respect to the [110] direction, the energy density for polar angle \( \theta = \pi / 2 \) is

\[
\frac{E_a}{V} = \frac{1}{4} K_{\text{in-plane}} \sin^2(2\phi) + \frac{2}{d} k_s
\]

with the out-of-plane surface anisotropy constant \( k_s \) and the total in-plane anisotropy

\[
K_{\text{in-plane}} = K_p + \frac{2}{d} k_p
\]

where \( d \) is the film thickness and the factor 2 counts the two surfaces of the film.

### 3.1.3 The Magnetostatic Energy

The magnetostatic energy is a self energy, arising from the interaction among magnetic dipoles. Each dipole produces a magnetic field, and the sum of all these fields acts on each dipole. The resulting energy is given by integrating \( M \cdot H \) over the magnetic body (see for example the book by Aharoni, chapter 6 [13]),

\[
E_m = -\frac{1}{2} \int_{\text{body}} M \cdot H \ dV = \frac{1}{8\pi} \int_{\text{space}} H^2 \ dV
\]

The equivalence with the integration of \( H^2 \) over the whole space is called pole avoidance principle. As the integrand is always positive, \( E_m \) is greater than zero except when \( H = 0 \) everywhere.

To calculate the energy stored in a ferromagnetic body one starts from a single dipole and gets the magnetic field by summing the field of all dipoles in the volume \( V \) of the magnet,

\[
H(r) = \int \frac{3v(M(r') \cdot v - M(r'))}{|r - r'|^3} dV
\]

where \( v = \frac{r - r'}{|r - r'|} \).

The magnetostatic energy is obtained by another integration over \( V \):
\[ E_m = \frac{1}{2} \int \int \frac{M(r) \cdot M(r') - 3(M(r) \cdot v)(M(r') \cdot v)}{|r - r'|^3} dV dV' \]

According to a theorem by Maxwell the internal field of a body whose surface is of second degree (an ellipsoid) is uniform [14]. In this case the field can be written as

\[ H_{in} = -NM = -4\pi DM \]

where \( N \) and \( D \) are tensors known as demagnetization factors, the trace of \( D \) is equal to 1. The magnetostatic energy of an ellipsoid with volume \( V \) then becomes

\[ E_m = \frac{1}{2} V(N_xM_x^2 + N_yM_y^2 + N_zM_z^2) \]

The demagnetization factors \( N \) depend only on the ratios of the ellipsis axes. Sometimes this energy is also called shape anisotropy, because a general ellipsoid has different energies along different axes. Demagnetizing factors of the general ellipsoid were calculated by Osborn [15]. For ellipses with two equal axes there are two special cases. The prolate spheroid looks like a long needle, the oblate spheroid is formed like a flat disk. These approximations for \( N \) are displayed in table 3.1.

<table>
<thead>
<tr>
<th></th>
<th>Prolate Spheroid</th>
<th>Oblate Spheroid</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( a \approx b = c )</td>
<td>( a = b \approx c )</td>
</tr>
<tr>
<td>( N_a )</td>
<td>( \frac{\ln(2m) - 1}{m^2} )</td>
<td>( \frac{\pi}{4m} - \frac{1}{m^2} )</td>
</tr>
<tr>
<td>( N_b )</td>
<td>( \frac{1}{2} - \frac{\ln(2m) - 1}{2m^2} )</td>
<td>( \frac{\pi}{4m} - \frac{1}{m^2} )</td>
</tr>
<tr>
<td>( N_c )</td>
<td>( \frac{1}{2} - \frac{\ln(2m) - 1}{2m^2} )</td>
<td>( 1 - \frac{\pi}{2m} + \frac{2}{m^2} )</td>
</tr>
</tbody>
</table>

*Table 3.1: Approximations of the demagnetization factors for prolate and oblate spheroids in the limit \( m = a/c \approx 1 \) (after [15]).*

Can the above results be used for the calculation of the magnetostatic energy of atomically flat particles? With the exception of magnetic domains, SEMPA measurements show a magnetization that is constant over the particle within experimen-
tal uncertainty. The magnetocrystalline anisotropy seems to be strong enough to keep the spins aligned along the easy axis. On the other hand, the approximation of a two-dimensional particle with an ellipsoid is not the best one can think of.

3.2 Energy Minimization and Domain Formation

The creation of domains in a magnetic body leads to lowered magnetostatic energy. On the other hand this introduces domain walls, which cost magnetocrystalline and exchange energy. So the question arises, how many domains will there be in a ferromagnet of given size and shape? A fundamental theorem by Brown proves that “the state of lowest free energy of a ferromagnetic particle is one of uniform magnetization if the particle is sufficiently small and one of nonuniform magnetization if it is sufficiently large” [16].

Kittel proposed scaling arguments to predict the domain configuration in thin films and small particles [17]. The various energies in a ferromagnetic body are calculated as a function of the body’s size and shape. The total energies of different configurations — single-domain and domains with various geometries — are compared with each other. Then the actual configuration is found by choosing the domain structure with the lowest total energy.

3.2.1 The Energy Cost of a Domain Wall

The structure of a domain wall is mainly influenced by the exchange energy, trying to make it infinitely wide, and the magnetocrystalline anisotropy, trying to minimize the wall width. The spin configuration which minimizes the sum of the two energies was calculated first by Landau and Lifshitz [18]. Kittel simply assumes a constant wall energy $\sigma_w$ per unit area of 3erg/cm² [17]. The wall energy constant for the case of an Ising system is given by $\sigma_w = 2J$ and for a Heisenberg system by $\sigma_w = 2\sqrt{JK}$ (see for example Politi and Pini [19]).

The above calculations neglect magnetostatic energy and thus assume an infinite large crystal. However, in a crystal of finite size the domain wall will reach the surface causing magnetostatic energy. This is important in thin films, where the spin orientation in the domain wall changes from Bloch-type (rotation in the plane of the wall) to Néel-type (rotation in the plane of the film) [13].

Suppose to observe a particle with a certain geometry. What happens with the domain wall energy if the particle is scaled by a factor $L$? A three-dimensional particle is enlarged along all space axes, so the wall energy becomes $L^2$ as much. For a two-dimensional particle only the in-plane coordinates are multiplied by $L$, its thickness
(a few atomic layers for a physical system) stays constant. Therefore the wall energy of a two-dimensional particle scales with $L$.

### 3.2.2 Magnetostatic Energy via Demagnetization Factors

For the general ellipsoid, the magnetostatic energy is proportional to the volume times the demagnetization factor. Enlarging all three ellipsoid axes by a factor $L$ does not change the demagnetization factors, because they depend only on the axis ratios. Thus the magnetostatic Energy scales with $L^3$, like the volume of the body.

A two dimensional body can be approximated by the oblate spheroid of width $a$ and thickness $c$. The magnetostatic energy is (to the lowest order, see table 3.1) proportional to $ac^2$ for in-plane magnetization and $a^2c$ for out-of-plane magnetization. Scaling $a$ with a factor $L$ leads to the following result: Two-dimensional magnets show a different scaling of the magnetostatic energy for in-plane ($\propto L$) and perpendicular ($\propto L^2$) magnetized systems.

### 3.2.3 Numerical Calculation of the Dipolar Sum

The dipolar energy of a monolayer of localized spins $S$ (measured in units of $\hbar$) is a discrete sum and can be calculated with techniques introduced by Politi and Pini [19], where $n$ and $m$ are (in-plane) coordinates in units of lattice constants, $r_{nm} = n - m$ is their difference, and $\Omega = (g\mu_B S)^2/a^3$:

$$E_m = \frac{\Omega}{2} \sum_{n \neq m} \frac{1}{|r_{nm}|^3} \left[ 1 - 3 \frac{(e_S \cdot r_{nm})^2}{|r_{nm}|^2} \right]$$

The spins $S$ are assumed to be aligned along a common direction $e_S = S/|S|$.

### 3.2.4 Introducing a Domain Wall in a Square Particle

The gain in magnetostatic energy by cutting a square particle of length $L$ into two rectangles of size $L$ by $L/2$ is calculated as follows: The summation of the dipoles is split in one part where both $n$ and $m$ are in the same rectangle and one part where $n$ belongs to one and $m$ belongs to the other rectangle. The latter changes its sign in the two-domains configuration. If $E(L, M)$ is the dipolar energy of a rectangle with $L$ by $M$ spins, the energies of the single domain particle $E_1$ and the two-domains configuration $E_2$ are

$$E_1 = E(L, L) = 2E\left(L, \frac{L}{2}\right) + [E(L, L) - 2E\left(L, \frac{L}{2}\right)]$$
and

\[ E_2 = 2E\left(L, \frac{L}{2}\right) - \left[ E(L, L) - 2E\left(L, \frac{L}{2}\right) \right] = 4E\left(L, \frac{L}{2}\right) - E(L, L) \]

Now the gain in magnetostatic energy calculates straight forward as the double of the square bracket,

\[ \Delta E_m = E_1 - E_2 = 2E(L, L) - 4E\left(L, \frac{L}{2}\right) \]

Figure 3.1 shows the calculated dependence of the energy gain from the size \( L \). Perpendicular magnetized squares gain energy according to a \( L \cdot \ln(L) \) law. For the in-plane magnetized geometry, the difference is proportional to \( L \). Comparing with the results of the scaling arguments given above, this leads to a logarithmic correction in the case of perpendicular magnetization.

Figure 3.1: Gain of magnetostatic energy of a 2-d square with \( L \cdot L \) atoms by introducing a domain wall in the middle. The circles are the result of the numerical summation of the dipoles. Open circles are for perpendicular magnetization, the line is a fit with \( L \cdot \ln(L) \). Filled circles are for parallel magnetization, the line is the exact calculation.
3.2.5 Formal Calculation of the Magnetostatic Energy

In the formal calculation the double integration over the volume is difficult to carry out. However, doing two partial integrations and assuming $\nabla \cdot \mathbf{M} = 0$ only a double integration over the surface $S$ of the volume remains:

$$E_m = \frac{1}{2} \iint_{SS} \frac{(\mathbf{M}(\mathbf{r}) \cdot \mathbf{n}(\mathbf{r}))(\mathbf{M}(\mathbf{r}') \cdot \mathbf{n}(\mathbf{r}'))}{|\mathbf{r} - \mathbf{r}'|} dSdS$$

This result is formal similar to the electrostatic energy, where $\mathbf{M}(\mathbf{r}) \cdot \mathbf{n}(\mathbf{r})$ plays the role of a surface charge. The integration of the magnetostatic energy can be carried out in the case of a two-dimensional rectangle with uniform in-plane magnetization parallel to one of the edges. It is reduced to the integration along the two lines with magnetization perpendicular to the line. The result for the energy gain by domain formation is $\Delta E_m \propto L$, it is shown in figure 3.1.

3.2.6 No Domains for In-Plane Particles

The above result for the gain in magnetostatic energy has to be compared with the cost in wall energy, which goes with the wall surface and thus is proportional to $L$. In-plane magnetized particles have the same functional dependence on the particle size of wall energy and magnetostatic energy. Thus one can conclude that the particle size does not influence domain formation. Only for very large values of $\Omega = E_{DW}$ domains can lower the total energy. The conclusion is that there will be no formation of magnetic domains in atomically thin in-plane magnetized particles. This contradicts Brown’s theorem. However, one could argue that a two-dimensional particle is always “small” in the real three-dimensional world.

3.2.7 Domain Configurations in Perpendicular Magnetized Films

In perpendicular magnetized particles the situation is quite different. The magnetostatic energy rises faster (proportional to $L \cdot \ln(L)$) than the wall energy. There is a critical length where it is favourable to create a domain wall and lower the dipolar energy. Exactly this behaviour is found in SEPA measurements of perpendicular magnetized structures of Fe/Cu(001). Particles with $L > 1 \mu m$ show domains.

There are numerous calculations which domain configuration is energetically favoured. Yafet and Gyorgy found that for certain ratios between surface anisotropy and dipolar interaction the magnetization is organized in stripe domains of alternating up and down oriented spins [20]. With different parameters Czech and Villain found that square domains are favoured [21]. Another paper by Kaplan and Gehring concludes that the energy difference between the checkerboard and the stripe con-
figuration is very small [22]. In the SEMPA experiments of perpendicular magnet-
ized Fe/Cu(001) a stripe configuration was found (see Chapter 5).

### 3.3 Stray Field of Magnetic Particles

One main application of small single-domain ferromagnetic particles is their possi-
ble use to store information. For this purpose it is essential to have many magnets
packed close together. But how close can they be put together without influencing
each other? Experiments were carried out to test the mutual interaction of atomical-
ly thin in-plane magnetized particles.

The interaction of magnetic particles is via the dipolar field. Special designed struc-
tures with “ellipse” and “butterfly” shapes (drawn in figure 3.2) are investigated in
SEMPA experiments. The two larger bodies act as a source of the stray field, the
smaller one in between is the “test charge” which measures the field.

![Figure 3.2: Geometry of the “ellipse” and “butterfly” structures. The probe island
has diameter of 1 μm in the left and side length 500 nm in the right structure.](image)

The strength of the dipolar stray field is calculated by summing the atomic dipoles,
assuming parallel alignment of all spins. A map of the stray fields is shown in figure
3.3 for the “butterfly” structure. The calculated fields at the place of the small test
particle are shown in figure 3.4 for the “ellipse” and “butterfly” shapes as a function
of the gap.

### 3.4 Thermal Fluctuations of the Magnetization

Let us assume a small ferromagnetic particle with uniform magnetization. Crystal-
line as well as shape anisotropy act as an energy barrier to prevent a change of the
magnetization direction. To overcome this barrier usually an external magnetic field
is needed, but rising the temperature results in a lowered barrier. The model of Néel
[23] and Brown [24] assumes thermal fluctuations of the magnetization direction of
a homogeneous magnetized particle with uniaxial anisotropy similar to Brownian
Figure 3.3: Calculated stray fields of a “butterfly” structure magnetized along +x. Only the two large particles (“wings”, marked in the left figure) are considered to be ferromagnetic for the calculation. Both in-plane components of the field are shown, $B_x$ (left) and $B_y$ (right). The field between the two “wings” points versus -x. The grayscale is white (black) for field along the positive (negative) axis.

Figure 3.4: Dipolar fields of “Ellipses” and “Butterflies” for one atomic layer. The field is calculated by summing the dipolar moments on the large particles averaged over the area of the test particle.
motion. The probability that the magnetization direction has not switched after the time \( t \) is given by

\[
P(t) = e^{-t/\tau}
\]

where \( \tau \) can be expressed by an Arrhenius law of the form

\[
\tau = \tau_0 e^{E/kT}
\]

The energy barrier \( E \) is given by the anisotropy of the particle via \( E = KV \). The parameter \( \tau_0 \) is of the order of \( 10^{9} \) s, which is also confirmed experimentally by Wernsdorfer et al. [25]. During measurement a magnetic particle behaves different depending on the ratio of \( \tau \) (calculated for given \( K, V, T \)) to the observation time \( t \):

In the large particle, low temperature limit \( \tau \gg t \) the magnetization direction is stable during the measurement (\( P(t) = 1 \)). If we request constant magnetization over \( \tau = 10^9 \) s (about 30 years) then \( E \) has to be at least \( 40kT = 1 \) eV. An estimation for the smallest stable Co particle on a Cu(001) surface which is 10 Å thick leads to a lateral size of \( \approx 60 \) nm (\( K = 1/4 \cdot 630 \text{Oe} \cdot 1400 \text{G} \)). When requesting only a time constant of \( \tau = 1 \) s a stable particle is \( \approx 40 \) nm large.

In the case \( \tau < t \) the magnetization flips back and forth during the measurement (\( P(t) \approx 0 \)). Measurements of an ensemble of particles will give an averaged magnetization equal to zero without applied field, so the remanence is zero. However, if a field is applied, the system is easily saturated. Each particle behaves like an atom of a paramagnetic substance but with a spin many times larger. This phenomenon is called superparamagnetism.

For intermediate conditions when \( \tau \approx t \), changes of the magnetization orientation occur on time scales of the measuring time or slightly slower. This effect is called magnetic after effect or magnetic viscosity. Because of the exponential law, only particles of a narrow size region fall into this category.

Fluctuations of the magnetization direction could become a limiting factor for getting stable ferromagnetic particles in two dimensions, because their volume is so small. On the other hand, only in atomically thin films one can expect new sources of stability, like a strong surface anisotropy.

Another description of changes of the magnetization direction in a microscopic system is quantum tunnelling as described by Awschalom, DiVincenzo and Smyth.
[26]. At low temperatures a particle can coherently move from one magnetic orientation to another by tunnelling. However, quantum effects are expected only for structures below a size of $\approx 20\text{nm}$, the length scale being dependent on the physical parameters of the material. With this effect, even at $T = 0$ changes of the magnetization direction are possible.
Chapter 4

In-Plane Magnetized Two-Dimensional Microstructures

4.1 Introduction

In this chapter two different magnetic systems are investigated: cobalt on a copper (001) surface and iron on a tungsten (110) surface. They are well known from studies of atomically thin films and have an easy magnetization axis parallel to the film plane. For this reason, only the in-plane component of the magnetization will be shown in the following SEMPA measurements.

4.1.1 Continuous Films of Co on Cu(001)

When evaporating atomically thin films of Co onto a Cu(001) single crystal surface, there is a certain thickness where ferromagnetic long range order will start. Combined magnetic (magneto-optic Kerr effect) and structural (STM, Auger spectroscopy) measurements by Ramsperger [1] give a thickness of 1.7 Å for the onset of long range order at room temperature. The magnetization lies in the film plane along a <110> direction, it has a four-fold symmetry induced by the substrate. Cobalt on copper grows in the fcc phase, whereas bulk Co has a hcp structure. As reported by Ramsperger et al. [27] the growth proceeds in an almost ideal layer by layer mode. This makes Co/Cu(001) an ideal model system for two-dimensional magnetism.

The structural quality of the substrate crystal plays an important role, too. On a millimetre sized sample there are certainly monoatomic steps present at the surface. If the steps lie parallel to each other with a more or less equidistant spacing, it is possible to assign a miscut angle to the Cu crystal or to adapt the crystallographic notation. Berger et al. [28] measured Co on a Cu(1 1 1 3) crystal, which corresponds to a (001) surface with a miscut angle of 6.2°. The steps induce an uniaxial anisotropy and the easy magnetization axis lies parallel to the step edges. For some measurements in this chapter a Cu crystal with 2° miscut was used, this will be mentioned in the figure caption.

Figure 4.1 shows an atomically thin Co film imaged over the whole Cu crystal with a surface of approximately 2 by 3 mm. This film is homogeneously magnetized, and
can be destabilized into a state with domains by applying a field close to the coercive field antiparallel to the magnetization direction. However, the interpretation of the images is not straightforward, as the edges of the substrate crystal are not well defined and scans over millimetre regions are difficult with the SEM (the secondary electron signal in the Mott detector gets weak when deviating the beam this far from the middle). The images of figure 4.1 are in accordance with domain structure measurements on Co films done by Oepen [29]. He found that the as grown state has a single domain configuration with small oppositely magnetized domains at the film edges.

Figure 4.1: Co film with thickness 4.4AL and coercive field 43.5Oe, recorded at the lowest magnification of the SEM. Left after applying a field of 340Oe, right after applying -43.3Oe. The images were recorded in remanence. The magnetization direction lies in the film plane along the horizontal image direction.

4.1.2 Fe Films on W(110) with Variable Thickness

Thin films of Fe on W(110) have been studied intensively by Gradmann et al. [30,31,32]. Both Fe and W have bcc structure in the bulk phase, and Fe films grow in the bcc structure on the (110) surface. However, the large lattice mismatch of \( a_W = 3.16\text{Å} \) to \( a_{Fe} = 2.87\text{Å} \) leads to a complicated growth mode for thicknesses larger than 2AL. A STM study reveals perfect layer by layer growth only for the initial 1.5AL, after that growth proceeds in bilayer mode [1]. The (110) surface of the W crystal defines a two-fold symmetry, causing thin Fe films on W(110) to have an uniaxial anisotropy, in contrast to the Co/Cu system mentioned above. The easy magnetization axis lies along the [110] direction.

A micro-wedge structure of Fe/W(110) was grown using shadow deposition. The result is shown in figure 4.2. Within a distance of 70μm the coverage increases from zero to the nominal thickness of 4.4AL. In the range from 2.2 to 3.5AL the film has
domains when measured as grown and even after applying a rather large magnetic field of 590Oe.

Figure 4.2: Micro-wedge of Fe/W(110), maximum thickness 4.4AL. The magnetization is measured along the [110] direction which lies horizontally in the image. In the thickness region of about 2.2 to 3.5AL domains are present even after magnetization of the film with 590Oe. In the right image the onset of ferromagnetic ordering at 1.7AL [1] is visible.

4.2 Particles of Variable Size and Shape

Much effort is put into the fabrication and characterization of small magnetic particles as well as their theoretical treatment. One important question is if magnetic domains will occur in a particle with known geometry and material constants. Kittel calculated domain structures of films and small particles using the scaling of the magnetic energies [17]. Brown’s theorem predicts a critical radius for a uniform magnetized sphere, dividing the single-domain region — valid for smaller particles — and the multi-domain regime for sufficiently large spheres [16]. Aharoni extended the validity of this theorem for prolate spheres [33].

Numerous results come from experiments [34-39]. Hehn et al. [38] fabricated cobalt squares with side length 500nm and thickness 25 – 150nm which are magnetized perpendicular to the plane. They concluded that the domain patterns are controlled by the details of the dot geometry.
The easiest two dimensional system is an atomically thin film of magnetic material. For the case of magnetization in the film plane a single domain state is predicted, as the demagnetizing factor approaches zero. When going to laterally confined geometries, the question arises, if the approaching boundaries can create magnetic domains. The measurement by Gu et al. [39] found that in 15nm thick epitaxial Fe (001) elements the transition from single-domain to multi-domain occurs if the size gets smaller than ≈ 50µm.

For these measurements, well defined ferromagnets are produced by placing a mask in front of the substrate during the growth process as described in chapter 2.2. These particles are ideal candidates to test the predictions made by the theory of two-dimensional magnets, because knowing the exact shape and size of the magnetic body is crucial. In the following, experimental results of different structures and materials are presented.

### 4.2.1 Atomically Thin Co Stripes

A first experiment addresses the question of the influence of the geometry upon the magnetization direction in flat particles. In other words, how large is the shape anisotropy in two-dimensional magnets? Figure 4.3 shows a set of 50µm long and 1µm wide Co stripes (in-plane aspect ratio $R = 50$). Each stripe has a different angle $\alpha$ with respect to the vertical image direction, which is the easy axis along which the magnetization is measured. The polarization of two sets of stripes with 3 and 6AL thickness ($R = 80$) is plotted as a function of $\alpha$. The polarization stays constant, that is the magnetization remains always parallel to the easy axis and does not lie along the stripe axis. The four-fold in-plane crystalline anisotropy of Co/Cu(001) is in the range of 600Oe [40] to 800Oe [12]. The shape anisotropy induced by $R$ is of the order of $6\text{Oe/AL} \cdot \sin^2(\alpha)$ when calculating according the techniques of Politi and Pini [19].

### 4.2.2 Micron-Sized Flat Co Particles

Figure 4.4 shows the result of fabricating Co particles of different size and shape. The SEMPA measurement of the in-plane magnetization was done “as grown”, that is without having applied any magnetic field (besides the earth magnetic field of 0.5Oe). The magnetization of the Co particles is uniform over the whole volume, irrespective of form and size. It is fixed during the growth process when long range order sets in. This is the reason why not all of the particles have their magnetization in the same direction.

Figure 4.5 is an image of the two smallest Co dots which were found for structures grown and measured at room temperature. They are 300nm and 130nm in diameter. Magnetic fields were applied before measurement to demonstrate the ability to store information in these particles. The islands remained stable at least during the measurement which took 10 minutes.
Figure 4.3: Co stripes magnetized along the vertical image axis. Top image is a set of stripes with variable angle $\alpha$, 23AL thick, 50µm long, and 1µm wide. In the separate image a stripe with magnetization perpendicular to its axis is shown (10AL thick, 120µm by 2µm). Bottom is a plot of polarizations obtained from two different sets of stripes, 3 and 6AL thick (both 40µm by 0.5µm).

4.2.3 Two-Dimensional Fe Microstructures

Small Fe/W(110) islands reveal the same behaviour as the Co islands. In figure 4.6 square and round islands are measured as grown. Again some of the magnetic particles are magnetized “positive” (white) and some “negative” (black). The two smallest Fe structures are displayed in figure 4.7. Their sizes are about 320nm and 160nm in diameter, comparable to the smallest Co/Cu(001) particles. This leads to the conjecture that the size of the smallest island is determined by the sharpness of the evaporation process rather than by fluctuations of the remanent magnetization. Further improvements of the evaporation process are necessary to finally find the limit where small magnetic structures loose their stability against thermal fluctuations at room temperature.
Figure 4.4: Round and square Co particles imaged as grown, 10 AL thick. A map of the magnetization along the vertical in-plane direction is shown, white and black correspond to a magnetization along +y and -y respectively.

Figure 4.5: The two smallest islands, magnetized with a field of $H = 340\,\text{Oe}$ (top, white) and $H = -340\,\text{Oe}$ (bottom, black) are 300nm (2.5AL) and 130nm (2AL) in diameter. The magnetization lies along the vertical image axis, indicated by the arrows. For details of the thickness evaluation see chapter 4.2.4 (thickness according to evaporation time: 12AL)
Figure 4.6: Fe/W(110) islands, 25AL thick, measured as grown. The islands are magnetized at random, but are in a single domain configuration. The magnetization direction is in-plane along the vertical image axis.

Figure 4.7: The smallest islands of Fe/W(110) have diameters 320nm and 160nm, measured as grown. Secondary electron polarizations are 24.5% (left) and -18.7% (right). The effective thickness is 4.6AL and 2.9AL (25AL according to evaporation time).

4.2.4 Thickness-Dependent Polarization

While measuring microstructures the polarization on the smallest islands was less than for larger structures or continuous films. Most probably this is due to shadow
effects during the growth of the structure. A mask which is not oriented exactly perpendicular to the molecular beam diminishes the maximum thickness for the smallest islands. The polarization as a function of the film thickness was evaluated from SEMPA images of structures larger than 1 μm, it is shown in figure 4.8. An exponential rise with the thickness $d$ to a maximum value is fitted to the measured points, where $\lambda$ is the inelastic mean free path of the electrons. This formula was used to calculate the thickness of the islands in figures 4.5 and 4.7.

$$P(d) = P_0(1 - e^{-d/\lambda})$$

![Graph showing polarization as a function of film thickness for Fe and Co](image)

**Figure 4.8: Polarization of the secondary electrons as a function of the film thickness for Fe and Co. The fit parameters are $P_0 = 34.5\%$, $\lambda = 3.7\text{AL}$ for Fe and $P_0 = 25.4\%$, $\lambda = 3.1\text{AL}$ for Co.**

### 4.2.5 Discussion

The experiments on atomically thin in-plane magnetized structures reveal a different behaviour than expected for three-dimensional magnets. The shape anisotropy is much weaker than the crystalline anisotropy. Domains do not penetrate inside the particle irrespective of their size. Both systems under investigation, Co/Cu(001) and Fe/W(110), behave very similar. The smallest particles are about 130 nm in diameter and stable at room temperature on the time-scale of the experiment.
4.3 Occurrence of Domains

4.3.1 Domains in Co/Cu(001)

Very seldom, 90° domains are found in the “as grown” state of Co particles, as shown in figure 4.9. To understand their origin one can argue that the substrate morphology plays an important role. A local concentration of monoatomic steps can introduce a local uniaxial anisotropy additional to the four-fold crystalline anisotropy so that the 90° rotated axis becomes the easy magnetization direction [28]. Once magnetized in an external field, the particles of figure 4.9 remained in the single-domain state.

![Topography Mx My](image)

**Figure 4.9:** Domains in Co/Cu(001): Topography and both in-plane components of the magnetization of three islands (thickness 6AL) are shown. The sample was imaged as grown, without having applied a magnetic field.

A completely different situation reveals the measurement on a 82 AL thick Co structure shown in figure 4.10. The domains clearly differ from those in thinner particles. There are straight domain walls, running along high-symmetry directions of the substrate crystal. The magnetization tries to be parallel to the borders to minimize the magnetostatic energy. Only the middle island (width 500nm) is single-domain. Because of these differences and because of its thickness, this structure will not be called two-dimensional. With a structure comparable to the “butterfly” of figure 4.10, one has entered the regime of three-dimensional magnetism.
Figure 4.10: 82Å thick Co “butterfly” structure. Topography and both in-plane components of the magnetizations are shown. Walls along <100> directions separate 90° rotated domains, one wall along <110> separates the magnetization rotation of 180° in the right part. The measurement was done on a 2° miscut Cu(001) with steps along the vertical image axis which corresponds to [110].

4.3.2 Domains in Fe/W(110)

Gradmann et al. [31] found that when the film thickness in the Fe/W(110) system is enlarged, the easy magnetization axis rotates from the [110] direction found in thin films to the [001] direction, the easy axis of bulk Fe. The transition takes place at a critical thickness of about 100Å, which is equal to 49Å thick. This behaviour is a consequence of competing in-plane anisotropies of surface and bulk. In figure 4.11 Fe structures are shown which are 42Å thick, however this thickness has an error of about 20%. The magnetization of the inner part on the three larger islands has rotated 90° to [001], while the border region and the smaller islands stay magnetized along the [110] direction, because they have a smaller thickness due to shadow effects.

Figure 4.12 demonstrates another complication when producing Fe microstructures. Round Fe islands with a diameter of 15μm are fabricated with thickness of 2.3Å and 3Å. In the as-grown state they have domains, like the microwedge on the location with corresponding thickness in figure 4.2. The domain walls are curved, in contrast to the walls of thick structures like in figures 4.10 and 4.11. A magnetic field was applied to force them into a single domain state. While the 2.3Å islands behave as expected, the 3Å thick structure could only be magnetized in one direction (black), but the opposite magnetic field of the same strength was too weak to
Figure 4.11: 42Å thick Fe structures on W(110). The easy axis is rotated 90° in the film plane to the [001] direction. Both images show the magnetization component along the vertical image direction. In the upper image only the border regions are magnetized along [110], probably because they are thinner due to shadow effects of the mask. The signal is $P = P_y$. The lower image was taken after rotating the sample ($\alpha = 30°$): both magnetization directions are visible in the polarization measurement, $P = P_x \sin(\alpha) + P_y \cos(\alpha)$.

The coercive field which has a peak as a function of thickness at about 2.5Å [41,42]. This peak is due to lattice distortions reaching a maximum before the Fe film finally relaxes into its own lattice constant at larger thickness. The domain walls are not straight, they possibly lie along imperfections caused by the relaxation process.

4.3.3 Conclusions

Domain formation cannot be completely excluded in two-dimensional systems with in-plane easy axis because atomically thin magnets are very sensitive to structural
Figure 4.12: Fe islands on W(110) with different thickness, as grown and after applying external fields. The 3AL thick islands have a coercive field larger than 590 Oe.

details like substrate imperfections (in the case of 90° domains in Co/Cu) and imperfect growth (Fe film on W(110) with ~2.5 AL thickness). Making the structures too thick is another limit where the no-domains rule does not apply any more.

4.4 Mutual Interaction via Dipolar Fields

Imagine to put small 2-d magnets close together on a crystal surface. Can they still be treated as independent, or will there be a mutual interaction? The stray field due to the dipolar moment of the atoms in a magnetic dot decays with the distance like $r^{-3}$. When placing structures very close together, the dipolar interaction could become a limiting factor for getting independent particles.

A first experiment investigates the switching behaviour of two-dimensional magnets in an external field. In figure 4.13 Co dots arranged in a circle were imaged after applying various fields. The result is that no domains are formed inside the individual dots, and that there is a certain interval of field strength (ranging from about 60 Oe to about 160 Oe), in which the magnetization of a Co dot is changed. The variation of this “reversing field” can be attributed to the non-perfect substrate surface, which certainly has many atomic steps and defects in a 3 μm wide area. Fur-
ther experiments led to the conclusion that the field necessary for the reversal of the magnetization does not depend on size or shape of the particles.

Figure 4.13: Magnetic dots with diameter 3μm and thickness 10ÅL arranged in a circle, after applying various magnetic fields. First, a large positive field saturated all dots in the positive direction (white, H = 210 Oe), see top left. Consecutively, a negative field of variable strength was applied and then the corresponding image was taken. The magnetization lies parallel to the vertical image direction.

The structure in figure 4.14 is designed to measure the strength of the dipolar interaction outside a magnetic particle. A small island is sandwiched between two larger ones. It acts as a probe for the stray field of the elliptically shaped particles. The magnetization lies vertically, so that the configuration in the middle image (with magnetizations “plus”, “minus”, “plus”) has lower energy than the configuration when all particles are magnetized in the same direction (top and bottom image in the figure, all islands “plus”).
The dipolar field of particles was calculated in chapter 3.3. The following measurement tries to quantify this field. It can only be done if the probe island switches at a smaller field than the larger particles. This was the case for the measurement shown in figure 4.14, where the external field did not change the elliptical particles. The superposition of the external and the dipolar field acts on the probe island. As the two ellipses do not change their magnetization direction, the field necessary to reverse the small island is larger when switching from black to white then from white to black. However, in the measurement shown in figure 4.14 no asymmetry in the reversing field was found within the experimental uncertainty of about 20e. The calculated value for a 4AL thick structure is 0.56Oe, which is too low to be detected in this experiment.

Figure 4.14: The small Co dot in the middle acts as a sensor for the stray field of the elliptical particles (Co thickness 4AL). The magnetizations lie in the vertical image direction. At the beginning the system was saturated in a field of \( H = +82\,\text{Oe} \), then three consecutive measurements were done: after applying \(-31\,\text{Oe}\) (top), after applying \(-33\,\text{Oe}\) (middle), and after applying \(+33\,\text{Oe}\).

Another geometry which produces a higher dipolar field was also tested. It consists of butterfly shaped structures as shown in figure 4.15. The “butterflies” have different gaps between the “wings” producing the stray field and the “body” acting as the probe. The measurement shown in figure 4.15 was done as grown, without applying any field. Note that only the butterfly with the smallest gap (100nm) has a centre which is magnetized oppositely to the “wings”.
The stray field acting on the small part in the middle of the butterfly can be evaluated from figure 4.16. Image 1 was recorded after having applied first +340 Oe and then -61.7 Oe, image 2 after +340 Oe and -82 Oe. This means that the field to switch the middle island from white to black lies somewhere between 61.7 and 82 Oe. Between image 2 and 3 a field of +85.3 Oe was applied to switch the middle island back to white, but nothing happened. Only after a field of +92.5 Oe the middle island switched back to white, see image 4. This means, that for changing from “black” to “white” at least an additional field of 3.3 Oe is needed. This extra field corresponds to twice the stray field of the white magnetized “wings”. With experimental errors included, the lower bound for the dipolar field in the middle of the butterfly is 1.7 ± 1 Oe, the upper bound is 15 Oe. Calculations give a value of 7.9 Oe for the stray field due to the “wings”.

The following conclusions can be drawn from the above experiments: dipolar stray fields can be measured on a sub-micron scale, their strength is in agreement with calculations. While in these structures the mutual interaction is rather weak compared to the coercive field of the particles, packing particles closer together will certainly change the situation.

4.5 Outlook: STM-SEMPA Measurements

Structural details play an important role in thin film magnetism. Only a scanning tunnelling microscope can give information on an atomic scale, so it is very desirable to do STM measurements simultaneously to the SEMPA investigations on the same magnetic island. However, this task is not an easy one, and by now only the very first measurements could be done.
Figure 4.16: “Butterfly” shape with 250nm gap, 10AL thick. Various fields were applied prior to the measurement: +340Oe and -61.7Oe for image 1, +340Oe and -82Oe for image 2, +85.3Oe immediately after the second image for image 3, and +92.5Oe immediately after the third image for image 4. The asymmetry in the switching field for the middle island is greater than 3.3±20e. The magnetization as well as the applied field lies along the horizontal parallel to the [110] direction. The Cu crystal used in this measurement has a miscut of 2° along the [\bar{1}10] direction.

On the structure of figure 4.17 SEMPA and STM measurements were done consecutively on the same spot on the sample. It is a “storage array” consisting of sixteen squares of length 200nm and 7.8AL thickness. A Cu substrate with 2° miscut was used for this measurement. The steps of the substrate are visible in the STM image as parallel lines.
Figure 4.17: SEMPA topography and in-plane magnetization along the vertical image axis and STM measurements of the same Co structure (4×4 array of 200nm squares with thickness 7.8AL). Tunnelling parameters are $U = 800\text{mV}$, $I = 0.5\text{nA}$, the STM image has some piezo drift.
Chapter 5
Atomically Thin Magnetic Structures
with Perpendicular Magnetization

5.1 Introduction

Perpendicular magnetized atomically thin films of Fe on Cu(001) possess an alternating up and down stripe-domain structure, see figure 5.1. The stripes show local orientational order and sustain fluctuations such as meandering and topological excitations. Confining the lateral size to structures with variable shape on the micron scale does not affect the width and the orientation of the stripes. Approaching the Curie-temperature, the domain configuration enters an orientationally melted phase before disordering completely. This two-step disordering is reminiscent of a two-dimensional melting process.

5.1.1 Structural and Magnetic Properties of Fe films on Cu(001)

Atomically thin films of Fe on Cu(001) deposited at room-temperature grow in the fcc phase, also known as γ-Fe. They have a perpendicular remanent magnetization at room temperature (Pescia, Stampanoni et al.[43,44]) in a thickness range from 2.5 to 5.5Å. (Liu et al.[45]). Outside this region, the Curie-temperature lies below 300K. At about 11Å, the film structure changes to bcc and the magnetization switches to in-plane orientation (Thomassen et al. [46]). These remarkable structural and magnetic properties are due to structural instabilities which can have two origins: first, lattice mismatch between fcc-Fe and Cu, and second, bulk bcc-Fe is energetically more favourable than fcc-Fe. Müller et al. [47] report that films below 5Å grow epitaxially on the substrate in a tetragonal distorted fcc phase with a complex reconstruction pattern (sinusodial shifts and vertical buckling).

According to a study by Giergel et al. [48] the temperature during growth is a crucial parameter for the morphology of the resulting Fe film. If evaporated at room temperature, the growth is almost ideal layer-by-layer throughout the thickness range from 0 to 11Å. At 5Å a martensitic transformation from fcc to bcc starts, with the coexistence of both phases, which reaches the bcc phase at about 11Å. When Fe is deposited at 100K, it exhibits a transformation to the bcc phase at a thickness of about 5Å. Simultaneously, the remanent magnetization direction
switches from perpendicular to parallel to the film plane. Although low-temperature grown films have a much less perfect morphology, they seem to be perfect to measure the reorientation of the magnetization direction at this thickness. This measurement was done by Pappas et al. [49] and Allenspach and Bischof [50].

All measurements presented here deal with room temperature grown films of Fe on Cu(001) with a thickness below 5 Å. In the following SEMPA measurements only the out-of-plane magnetization component was non-zero, so only this component will be shown in the images.

5.1.2 The Striped Domain Configuration

The long-ranged dipolar interaction leads to domains in perpendicularly magnetized films. Although being much weaker than the exchange interaction, which acts essentially only between nearest neighbours, it wins at distances in the μm range. Yafet and Gyorgy calculated the energy gain when domains are formed [20]. The continuous film breaks into domains consisting of stripes with mesoscopic width of up and down aligned spins. The width of the domain stripes is a trade-off between the gain in magnetostatic energy and the cost in exchange energy, which favours parallel alignment of neighbouring spins.

What happens with the domains if the temperature increases and finally reaches the Curie temperature? Monte Carlo simulations of ultrathin magnetic films by Booth...
et al. predict three distinct phases [51]. The ground state for low temperatures is a phase with striped domains. As the temperature is raised, fluctuations of the domain boundaries increase until the domain configuration loses the directional and translational symmetry of the stripes. This loss of orientational order is the first transition of a two-step melting process. Only if the temperature is increased further, the second transition to a fully disordered system occurs. The Hamiltonian used in the simulations is given by the sum of exchange interaction $J$ (restricted over nearest-neighbours’ spins) and the dipolar interaction $g$ (summed over all pairs)[51]:

$$H = -J\sum_{(ij)} S_i S_j + g \sum_{i \neq j} \frac{S_i S_j}{R_{ij}^3}$$

There are several other studies of competing interactions. In a theoretical analysis of a Hamiltonian similar to the above Löw et al. concluded that even a simple Ising model with a combination of long-ranged dipolar and short-ranged exchange interaction has a remarkably complex phase diagram [52].

The arising stripe geometry is somehow similar to a liquid crystal. The domain stripes play the role of the building block, whereas a conventional liquid crystal is made up of elongated organic molecules. A liquid crystal is also called a mesomorphic phase (of intermediate form) with mechanical and symmetry properties between a solid and a liquid. Depending on the dimension of the positional order, there are three different types (see for example the book by DeGennes and Proust [53]):

1. **Nematic phases** have only directional, but no positional order. It corresponds to a liquid of elongated molecules which tend to be oriented parallel to a common axis. With the liquid phase in mind, this behaviour is best described by the term orientational freezing.

2. **Smectics** can be described as nematics with one-dimensional positional order. In one direction, the molecules are stacked in layers with a well defined spacing constant.

3. **Columnar phases** are two-dimensionally ordered systems in three dimensions. They can be described as a two-dimensional array of liquid tubes without order along the tube axis.

The analogy of the domain stripes to a liquid crystal is used for instance by Abanov et al. [54], where the authors calculate domain structures of ultrathin ferromagnetic films and their phase transitions. They predict a low-temperature smectic phase with stripes and a tetragonal liquid phase (liquid with tetragonal symmetry which is induced by the substrate crystal surface) at higher temperature. The transition between the two phases is either of first order or there exists a third phase, which the authors call Ising nematic.
5.2 Perpendicular Magnetized Ultrathin Particles

Fe particles with thickness of about 3ÅL and variable lateral size and shape were produced with the technique of mask deposition. The magnetic structure consists of stripe domains as found in atomically thin films. Size and shape of the particles do not influence the stripe width and orientation, as shown in figure 5.2. This is in contrast to measurements by Hehn et al., where the domain pattern is controlled by details of the particle geometry [38]. Thus it is possible to use these geometrical stripe properties as a two-component order parameter.

As their size is getting smaller, the particles of Fe/Cu(001) shown in figure 5.3 exhibit a transition to a single-domain state. According to theoretical estimations there will be a critical length $L_{\text{crit}}$ where the ground state of smaller particles is the single domain configuration. Like in calculations for an infinite long stripe of magnetic material by Politi [19], the length $L_{\text{crit}}$ is given by the following function of dipolar
interaction strength $\Omega$, domain wall energy $E_{DW}$, and domain wall width $w$, only the numerical constant is changed slightly [55]:

$$L_{crit} = 3.72 \cdot w \cdot e^{E_{pw}/2\Omega}$$

Figure 5.3: Top: magnification of the two smallest islands of figure 5.2 (round particles at left, square ones at right). Bottom: slightly thicker particles with thickness 2.8AL Fe. The result is the occurrence of wider stripes and thus a transition to a single-domain configuration for the smaller particles.

A rough estimate for the critical length assuming $w = 10$ nm ($w < 20$ nm is found in our SEMPA measurements), $2\Omega = 1$ K, and $E_{DW} = 5$ K is $L_{crit} = 5 \mu m$. In the experiment a value of $L_{crit} = 2 \mu m$ is found, it varies between different samples and particle thickness (compare figure 5.2 with bottom image of figure 5.3).

### 5.3 Continuous Films of Fe/Cu(001)

Atomically thin Fe Films are evaporated onto a Cu(001) substrate at room temperature. Consecutively, SEMPA measurements of the remanent magnetization are done without having applied a magnetic field. The film is magnetized perpendicular to the plane, with striped domains oriented along a certain preferential direction, see figure 5.4.

The domains show pronounced meandering around the preferential stripe direction. A Fourier transform shown in figure 5.5 allows to quantify the amount of local orientational order. Transforming many images shows that the preferential stripe orientation lies between the [010] and [\overline{1}10] directions with respect to the fcc Cu(001)
substrate surface. This meandering is one type of excitation predicted by the theory of Abanov et al. [54].

There are also topological excitations present, that is defects localized in space: in figure 5.6 it is possible to identify the four different types a – d introduced in [54]. Occasionally, further dislocations are also detected, with circular- and kink-like defects (figure 5.7). These do not need to be new types of dislocations, but could be combinations of defects a–d and extreme meandering. Simultaneously acquired SEM images show no irregularities in the topology to account for these excitations. However, structural defects cannot be completely excluded without checking on an atomic scale, for instance by using a STM.

5.4 Wedge-Shaped Films of Fe on Cu(001)

Using the technique of shadow-deposition it is possible to produce wedge-shaped Fe films. A wire of diameter 0.125 mm is positioned about 10 mm in front of the Cu substrate. Evaporating Fe across this wire results in a film with an unsharp shadow of the wire. The thickness profile of the structure can be obtained by Monte-Carlo simulation, see the upper part of figure 5.8. With the chosen evaporator geometry, the wedge rises from zero to maximum coverage within $\approx 65 \mu$m. The lower part of
Figure 5.5: Top: power spectrum of the Fourier transform of figure 5.1 (left) and figure 5.4 (right). Bottom: polar plot summarizing the propagation directions found in Fourier transforms of many images.

Figure 5.8 shows the polarization image of a wedge structure with maximum thickness of 4.7 AL Fe. It reveals two windows where the perpendicular component is non-vanishing. Inside it there are domains with a stripe configuration similar to those of continuous films.

After magnifying the magnetic active region, it is possible to examine the thickness dependence of the domain stripes by moving across the image in figure 5.9. Having in mind the connection between Curie temperature and film thickness [45], this is equivalent — regarding the phase transition — to changing the temperature. On the wedge sample, it is possible to have a “look” on the phase transition in a single image.

Approaching Curie temperature at the edges of the window, the domain width becomes smaller, causing more domains to appear. Simultaneously the local orientational order of the stripes is disturbed, as can be seen in the magnification in figure 5.10. In a narrow region there is magnetic contrast without formation of stripes but of irregularly shaped domains. Going further on, one reaches the paramagnetic regime, where there is no remanent magnetization at 300K.

Cooling the wedge structure gives a similar image, but now the magnetic active window is enlarged, see the lower image in figure 5.9. Even at maximum thickness of the wedge the perpendicular magnetization is still present in the right image of figure 5.10. However, for this thickness of 4.1 AL the stripe order has vanished, as found in the room temperature measurement only in a narrow region. Now it is pos-
Figure 5.6: 2.6AL Fe/Cu(001) with dislocations in the stripe-domain structure. The inset shows the definition of the four types of dislocations [54]: a inserted from above, b inserted from below, c passage due to two dislocations inserted from above and from below, d island due to two dislocations, one ending above and one ending below the island centre. The [100] direction of the Cu surface lies along the horizontal image axis.

Figure 5.7: Two other types of excitations. Left: circular-like defect, 2.6AL Fe. Right: kink-like defects, 2.1AL Fe. The orientation of the Cu substrate is indicated on the side.

It is possible to analyse this phase more quantitatively. The Fourier transform shows no preferential direction, but a tetragonal symmetry corresponding to the four-fold symmetry of the Cu(001) surface. This phase can be described as a tetragonal liquid in accordance to theoretical prediction [54]. Another hint comes from Monte Carlo
Figure 5.8: Fe wedge on Cu(001). Top: Monte-Carlo simulated thickness profile of a 4.7ÅL thick wedge. Bottom: SEMPA image of a wedge with maximum thickness 4.7ÅL (same length scale as top graph). The perpendicular magnetization is non-vanishing in a range of about 2.5 – 4ÅL (±20%).

Figure 5.9: Fe wedge on Cu(001): The thickness increases from 1.6ÅL on the right to 4ÅL on the left. Top and bottom images measured at 300K and 190K respectively. The horizontal is the [110] direction of the Cu surface.
Figure 5.10: Left: magnification of the marked area of figure 5.9, measured at $T = 300\,\text{K}$. Right: region of maximum thickness 4.1AL, measured at $T = 190\,\text{K}$ and its Fourier transform. The Fourier spectrum reveals tetragonal symmetry.

Simulations of domain structures. Booth et al. [51] show in their figure 1 the calculated magnetic domain configuration for various temperatures. One image (1d) is redrawn in figure 5.11, along with its Fourier transform. Both resemble very much the measured configuration as displayed on the right side in figure 5.10.

Figure 5.11: Left: Monte Carlo simulation of a system consisting of 64·64 spins for “temperature” $J/g = 10$ (figure 1d of [51]). Right: its Fourier spectrum.

A still open question is how the transition from striped to disordered domains exactly takes place. Neither simulations [51] nor our experiments are able to clearly identify if there is a first order phase transition or a new phase — the Ising nematic — that separates the low temperature smectic phase from the tetragonal liquid. According to theory [54] this depends on the sign of a parameter. In the mean-field approximation, this parameter was found to be negative, which would cause a first order transition.

The experiments described here are in line with the theory of dislocation-mediated two-dimensional melting of Kosterlitz and Thouless [56]. As Nelson and Halperin point out [57], one consequence of this theory is that the melting process takes place in two steps. First, the low-temperature solid phase is brought into a liquid-crystal phase with exponential decay of translational order due to dissociation of disloca-
tion pairs. At higher temperatures, dissociations of disclination pairs produces a liquid where the orientational order is destroyed, too.

What is the reason that it is possible to observe a disordered liquid in these measurements, which take several minutes for a single image? The domain boundaries with all the described fluctuations must have been pinned at least for this time. In an atomically thin film pinning is possible due to defects on the surface, like monoatomic steps, as well as impurities. If there is no pinning, the disordering can vary in time making it impossible to measure domain patterns. What about the paramagnetic region following the tetragonal liquid phase? Can it be explained due to depinning and therefore fluctuating during measurement, resulting in an image without polarization? At present, this question cannot be answered, as well as the question if there are domains smaller than the spatial resolution of the SEMPA (about 10 nm).
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Curriculum Vitae

Personal Data
Name                     Christian Stamm
Date of birth            January 3rd, 1970
Place of birth           Davos, Switzerland
Nationality              Swiss
Citizen of               Winterthur (ZH) and Thayngen (SH)

Schools
1976 - 1980              Primary School (Volksschule) in Vienna, Austria
1980 - 1988              High School (Bundesgymnasium 18) in Vienna, Austria
June 8th, 1998           High School Diploma (Matura)

University Studies
1989 - 1994              Physics student at the Swiss Federal Institute of Technology (ETH) in Zürich, Switzerland
Summer 1992              Trainee at the Paul Scherrer Institut (Villigen, Switzerland) at the Low Energy Muon Project.
April 28th, 1995          Physics Diploma of the ETH Zürich
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