Doctoral Thesis

Anisotropy and phase transitions in atomically thin magnetic microstructures

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Publication Date:
2000

Permanent Link:
https://doi.org/10.3929/ethz-a-004003698

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Anisotropy and Phase Transitions in Atomically Thin Magnetic Microstructures

A dissertation submitted to the
SWISS FEDERAL INSTITUTE OF TECHNOLOGY ZURICH
for the degree of
Doctor of Natural Sciences

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2000
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Abstract

A recently developed technique allows the in situ fabrication of real two dimensional ferromagnetic particles. In this thesis a Scanning Kerr Microscope (SKEM) with a lateral resolution of about 1\(\mu\)m is used to study the in-plane magnetization of such ultrathin particles on the micrometer size scale.

The SKEM provides the possibility to measure the temperature dependent magnetization not only in zero magnetic field, but also in an applied external field. Magnetization loops M(H) can be collected at different spots on the sample. In addition, by scanning an interesting area we are able to map the magnetization in any external magnetic field along an in-plane direction.

In the first part of this thesis a detailed study of the magnetization of ultrathin Co particles evaporated on a Cu(100) single crystal is presented. The outstanding property is their single domain remanent state. They show square hysteresis loops along the easy magnetization axis with slightly varying fields H_{C}. The single domain state is stable and independent on the lateral extent of the structures, at least down to 1\(\mu\)m. Hysteresis loops are collected on particles of various lateral sizes and shapes to measure the switching behavior and the influence of their shape on their magnetization. We found that local changes in the substrate dominates over eventual size or shape dependence. No mutual interaction was observed for particles with a distance between each other down to 2\(\mu\)m.

In the second part of this thesis the critical behavior of in-plane magnetized Co/Cu(100) films and particles are investigated. The system exhibits a second-order phase transition from a ferromagnetic phase to a paramagnetic phase. In addition to the fourfold symmetry breaking field, a misfit in the Cu substrate causes a weak twofold anisotropy field. Both anisotropies scales differently with temperature and the transition is probably dominated by the uniaxial crystal field. However, the fourfold symmetry breaking cannot be neglected and causes non-universal behavior. Simultaneously collected M(H) curves parallel and perpendicular to the easy magnetization axis during phase transition reveal an anisotropic behavior up to about 10 Kelvin above T_{C} and pronounced peaks in both susceptibilities at T_{C}. An analysis of the shape of the magnetization curves indicates that the fourfold symmetry breaking vanish slightly below T_{C} while the twofold field persists.

The relevant length at phase transition - the correlation length of the electron spins - develops to the maximum of about 1\(\mu\)m in our system. This is about the maximal particle
size detectable with the SKEM. Finite size effects are therefore hard to observe. The values for the critical exponents \( \beta, \delta \) and \( \gamma \) measured at 10x6\( \mu \)m\(^2\) and 4x4\( \mu \)m\(^2\) particles are slightly enhanced compared with the extended film values. In addition they exhibit a slightly different scaling function.
Zusammenfassung

Eine kürzlich entwickelte Technik erlaubt die Herstellung von zweidimensionalen ferromagnetischen Teilchen im Ultrahochvakuum. In dieser Arbeit wird ein Scanning Kerr Mikroskope (SKEM) mit 1µm Auflösung benutzt, um die in-plane Magnetisierung dieser Teilchen auf Mikrometer Skala zu untersuchen.

Mit dem SKEM kann man die temperaturabhängige Magnetisierung sowohl ohne, als auch mit einem extern angelegten Feld messen. Auf diese Weise können Magnetisierungskurven M(H) an beliebigen Stellen auf der Probe gemessen werden. Zusätzlich kann durch rastern ein Bild der in-plane Magnetisierung entlang einer beliebigen Richtung in beliebigem Feld erstellt werden.


Die relevante Länge beim Phasenübergang - die Korrelationslänge der Elektronen spins - beträgt in unserem System maximal etwa 1µm. Unglücklicherweise ist 1µm auch ungefähr die Partikelgrösse, die man mit dem SKEM noch detektieren kann. Ein eventueller
Effekt, der von der Teilchengrösse herrührt, ist deshalb schwer nachzuweisen. Die an 10x6\(\mu\)m\(^2\) und 4x4\(\mu\)m\(^2\) Teilchen gemessenen Werte für die kritischen Exponenten \(\beta, \delta\) und \(\gamma\) sind leicht grösser verglichen mit den Werten gemessen an einem ausgedehnten Film. Die Teilchen zeigen auch ein leicht andere Scaling Funktion.
Introduction

Chapter 1 gives a short overview of the experimental facilities. Aside from common UHV facilities our experimental apparatus features a modified MBE evaporation stage, which allows production of the 2D particles and a Scanning Kerr Effect Microscope SKEM for the magnetic measurements.

In chapter 2 we address the question what happens with the magnetization when the lateral dimensions of an ultrathin in-plane magnetized film are reduced. We will measure the magnetization of particles with different sizes (down to $\approx 1\mu m$) and different shapes. Thereby the SKEM is used to measure magnetization loops or to collect magnetization maps of an area. We designed different structure geometries and arrangements to investigate shape anisotropy, mutual interaction or simple size effects.

Magnetic order is a collective phenomenon. As such, the dimensionality of the system plays a crucial role. Close to the Curie temperature the correlation length of the electron spin in a two dimensional magnet reaches values of about one micrometer. The influence of this diverging correlation length on the behavior of magnetic islands with a lateral size of the same magnitude is investigated with the SKEM. In chapter 3 we first measure magnetic phase transition on atomically thin Co films grown on a Cu(100) crystal to learn more about the magnetic system. Especially as our system combines both a fourfold and a twofold in-plane symmetry breaking field. After knowing the behavior of the extended system we repeat the same measurements on structured film. Thus by comparing both measurements one can assign a different behavior to finite size effects.

In chapter 4 we proceed measuring phase transitions. But this time both magnetic in-plane components are measured during phase transition. Thus by comparing both components we will directly see an isotropic (at least with respect to the film plane) or anisotropic behavior.

Useful insight in our system can be gained from the shifted hysteresis loops. In the case of a single magnetic film with two- and fourfold magnetic anisotropy, the shiftfield $H_S$ is proportional to the twofold magnetic anisotropy. In addition the inverse of the initial slope is proportional to the sum of the two and the fourfold anisotropy. An analysis of the measured perpendicular loops near $T_C$ is given in chapter 4.
1. Experimental Setup

Magnetic properties of atomically thin films are very sensitive to contamination of residual gas [1.1]. It is therefore crucial to work under ultra high vacuum (UHV) conditions. Our UHV system consists of three interconnected chambers, namely a preparation chamber, a Kerr measurement chamber and a STM chamber. These chambers are independently pumped with a base pressure better then $5 \times 10^{-11}$ mbar. The UHV system is equipped with a load lock which allows to introduce samples, crucibles for evaporation and STM-tips into the UHV without breaking the vacuum [1.2-1.4]. Substrates, STM-tips and masks (see next section) are mounted onto tetragonal bodies (transporters) [1.3]. These can be handled within and transferred between the chambers with a specially designed transfer system.

Surface cleaning, film and structured layer growth, surface characterization with low energy electron diffraction (LEED) and Auger electron spectroscopy (AES), as well as tip preparation for the scanning tunneling microscope (STM) are done in the preparation chamber. The most important tool in the preparation chamber is a modified MBE evaporation source. It allows to grow ultrathin films with lateral extensions below 1μm (see next section).

Once prepared, the samples are transferred to the second chamber. There, the magnetic measurements are done with a Scanning Kerr Microscope SKEM.

Finally the third chamber contains a Scanning Tunneling Microscope STM, which is used to characterize the sample’s surface.

1.1. Evaporation stage

Our magnetic metals, which are of 99,999% purity, are evaporated from a water cooled Knudsen cell-type evaporation source. The substrate is held at room temperature (RT) and the pressure never exceeds $5 \times 10^{-10}$ mbar. In order to grow well defined and laterally limited films, a mask technique is used.

Figure 1-1 shows a schematic view of the evaporation stage. The mask basically consists
of a 1μm thick titanium foil which is mounted onto a transporter. With a commercial Focussed Ion Beam (FIB) system a structure is etched through the foil. Next the transporter with the foil is charged into the UHV system through the load lock and annealed in situ. The magnetic films/structures are evaporated onto a Cu(100) single crystal substrate. Both, the crystal and the mask are placed in front of the evaporation source. In addition, the transporter with the diaphragm can be moved with an inertial slider [1.4] allowing accurate positioning relative to the substrate. To evaporate continuous films the mask is removed.

As the sharpness of the structures is determined by the ratio of the distance between source and mask on one hand and between mask and substrate on the other hand, the source has to be placed far away from the mask, whereas the mask and the substrate should be as close as possible. This leads to a very small evaporation rate, i.e. the amount of deposited material per minute is typically 0.03 monolayer per minute. The evaporation rate is determined with Auger Electron Spectroscopy AES and cross-checked with scanning tunneling microscopy STM. Therefore a magnetic film is evaporated (e.g. Co on Cu(100)) and the time is monitored. Then the number of atomic layers is calculated from the ratio of Auger peaks, e.g. the height of the 656eV Co-peak relative to the height of the 920eV Cu-peak. Previously, the AES has been calibrated with the help of the STM [1.5]. With the evaporation time and the film thickness, the evaporation rate can be calculated. Structures are evaporated by controlling the evaporation rate, previously determined on the extended film. With this combination of MBE and mask technique we are able to fabricate atomically thin microstructures and control their number, thickness, size, and shape at will.
1.2. Scanning Kerr Microscope

As the magnetic signal originating from size limited microstructures is extremely small, a scanning technique with focused light is employed to measure the magnetization locally. Therefore, we use the magneto optic Kerr effect [1,6].

FIGURE 1-2. Schematic view of the Kerr Setup.

In our experiment, we use a modified Kerr setup. The light from a stabilized laser diode ($\lambda = 675\text{nm}$) outside the vacuum is directed through a linear polarizer and a focusing optics. This optics is located in the tube of a reentrant UHV view port. Both, optics as well as the UHV view port were specially designed to reach a diffraction limited laser focus. This focused beam is then reflected on the sample and with an identical optical arrange-
1.2. Scanning Kerr Microscope

ment directed to another polarizer outside the vacuum and finally detected with a photodiode. We use the transversal Kerr geometry [1.6], where the polarization and intensity of the reflected light depends on the magnetization of the sample perpendicular to the scattering plane. To switch the magnetization a magnetic field of up to 150 Oe can be applied with a Helmholtz coil. Before each measurement the sample is transfered from the preparation chamber and placed on a specially designed stage. The stage basically consists of two tables on linear roller bearings, which allow horizontal scanning. Each of the tables is moved independently from ex situ with a micrometer screw and a dc-motor coupled to a bellows. In addition, the sample can be rotated around its vertical axis. For temperature dependent measurements the sample is cooled down with liquid nitrogen or heated with the help of a tungsten filament. The whole optical setup has to be removed from the chamber while baking the UHV system.

The SKEM is mainly used in two different ways. In one mode, the ‘imaging’ mode, a map of the magnetization $M(H)$ is simultaneously collected together with a reflectivity image. Therefore at each image pixel a field $+H$ is applied to magnetize the sample along the field direction. Next the field is either turned off or another field $+H'$ is applied and the Kerr intensity $I(+H')$ is measured. Subsequently, a field $-H$ is applied in the opposite direction. Again this field is either switched off or reduced to $-H'$, and the intensity $I(-H')$ is measured. The sum of these two intensities $I(+H')+I(-H')$ is proportional to the total reflectivity while the difference $I(+H')-I(-H')$ is proportional to the magnetization at the field $H'$ within the laser focus [1.6].

**Figure 1-3.** A typical SKEM image of 40x40 pixel and 40x40 $\mu$m$^2$ size. (a) $I_{\text{Kerr}}(+0)+I_{\text{Kerr}}(-0)$, i.e. the total reflectivity and (b) $I_{\text{Kerr}}(+0)-I_{\text{Kerr}}(-0)$, the remanent magnetization of the same area. It takes about 5 minutes to record such an image.
Figure 1-3 shows a typical SKEM image collected in this manner. While Fig. 1-3(a) displays the total reflectivity, Fig. 1-3(b) reports the remanent (+H' = -H' = 0) magnetization. The area is 40x40μm² and the image size 40x40pix. The magnetic dots visible in (b) are circular with a diameter of 3μm. 

Note: With this technique, it is possible to collect a map not only of the remanent magnetization but at any applied field within the fieldrange of the helmholtz coil.

The lateral resolution is defined by the laser spot size. It is minimized by scanning the edge of a well defined magnetic feature and varying the vertical position of the whole stage to achieve maximum edge sharpness. Figure 1-4 show the resolution limits of the SKEM. The plot in (a) shows the coverage at a stripe edge vs. the lateral distance. It is obtained by analyzing STM pictures. The edge sharpness of the grown structure is 150nm. In (b) a similar edge is scanned with the SKEM after optimizing the focus. Here the edge is about 3μm. The full width at half maximum is about 1.5μm [1,2].

The other way to use the SKEM is to continuously monitoring the intensity I(H) while sweeping the magnetic field. A plot of the collected signal vs. the applied field is shown in Figure 1-5. It shows a square shaped hysteresis loop typical for a magnetically easy axis. The signal I(+H') at zero applied field H'=0 Oe, i.e. the remanent magnetization, is the same as in a saturation field of H'=20 Oe.

To collect such a hysteresis it takes about one second.
A home built scanning tunneling microscope STM situated in the third vacuum chamber is used for structural characterization. It is damped with a coil spring suspension with an eigenfrequency of 1.3 Hz. For STM imaging the sample is placed on a stage with its surface pointing to the horizontal tube scanner. This scanner holds an exchangeable tungsten tip, which was chemically etched and subsequently processed in the preparation chamber by electron bombardment and characterized by field emission. The piezotube scanner is calibrated by imaging a cleaved GaAs (110) surface with atomic resolution [1.2]. In addition, single atomic steps on the same surface were used to calibrate the horizontal axis i.e. the step height. Samples can be imaged at room temperature.

In this work the STM is mainly used to characterize the samples surfaces.

**Figure 1-5.** A typical easy axis magnetization loop collected with the SKEM.
2. Single domain particles

Ultrathin ferromagnetic layers have been studied experimentally since more than 10 years. Co/Cu(100) e.g., has become a model system for investigating the properties of two dimensional systems [2.1-2.4].

Face-centered-cubic (fcc) Co has a small lattice mismatch (1.9%) with respect to Cu(100), and thus grows pseudomorphically, thereby matching the fcc structure of the substrate. In addition, the distortion along the surface normal is also very small [1.5, 2.5, 2.6]. This results in a nearly layer by layer growth on a flat Cu(001) surface. Deposited onto a stepped Cu surface, the Co films reproduce largely but not exactly the steps of the underlying substrate. These ultrathin films reveal long-range ferromagnetic order at room temperature (RT) down to a thickness of ~1.7ÅL. In the ferromagnetic phase, these layers are homogeneously magnetized in the film plane with a single domain remanent state.

An epitaxial grown film on flat Cu(100) crystal substrate exhibits a fourfold magnetic anisotropy with the energetically most favored axis, i.e. the magnetic easy axis, along the [1±10] directions. Grown on a stepped surface, Co exhibits a breaking of the magnetic fourfold symmetry due to the steps, resulting in a magnetic easy axis along the steps [2.7-2.11].

In the following, we investigate how reducing the lateral size and thereby introducing boundaries influences the magnetic properties of atomically thin Co layers, i.e. the remanent state, coercivity field and mutual interaction. In addition, some measurements are done to investigate the influence not only of the size but also of the shape of these structures on their magnetic behavior.

2.1. Sample preparation

The surface of the Cu(100) substrate is cleaned by Ar ion sputtering, using a beam energy of 1kV and a target current of 7μA. Subsequently, the copper crystal is annealed up to 400K and left for some hours to reach room temperature again. Then, the substrate is placed onto the evaporation stage in front of a structured mask and the magnetic material is evaporated. After growth, the sample is transferred to the SKEM, where the structures are located by collecting hysteresis loops along lines over the hole sample.
2.2. Single domain remanent state

The most dominant feature of Co microstructures is their single domain remanent state.

![Graph showing remanent magnetization](image)

**Figure 2-1.** (top) SKEM image showing the remanent magnetization of a set of circular Co dots (4 x L) with diameter of 1, 2, 4, 6, 8, 10μm (left to right). (bottom) Linescan through the same dots. The full circles report the remanent magnetization deduced from hysteresis loops. The open circles represent the saturation magnetization from the same loops. Both parts give no indication on domains penetrating the particles. The particles are in a single domain remanent state.

Figure 2-1 illustrates this fact. The upper part displays a SKEM image of a series of cir-
2.3. Exception to the single domain rule

There are exceptions to the no-domains rule. Figure 2-2 reports on such an exception. The plots in (a), (b) and (c) are SKEM images all taken on the same 5μm wide and 2.5AL thick Co stripe. It is evaporated on a nominally flat Cu(100) single crystal. (a) presents a map of the total intensity, which correspond to the surface topography. Even on this length scale a surface structure nearly parallel to the stripe is visible. Figure 2-2(b) and (c) display magnetization maps collected at the same location. The figures show the Kerr signal along the easy magnetization [1-10] direction, indicated with an arrow in (c). While in (b) the external field was switched off completely, (c) was collected with a small external field (4 Oe) applied. The remanent picture contains within the fully magnetized section (white) two grey sections in the middle of the stripe. Hysteresis loops taken in the corresponding areas clarify the findings. They are shown in Fig. 2-2(d-f). Loop (d) taken in the white section represents an easy axis loop indicating that M_R points along [1-10]. (e) and (f) display loops from the “dark grey” region and from the “light grey” region respectively. These loops are shifted with different fields H_S separating the two minor loops. Shifted hysteresis loops in the Co/Cu(100) system regularly occur, when monatomic steps induce locally an uniaxial anisotropy. This uniaxial anisotropy together with the already present...
FIGURE 2-2. Magnetic domains in two-dimensional particles. The figure shows SKEM images of the same section of a 5 μm wide and 2.5 AL thick Co stripe. (a) Reflectivity image, (b) magnetization measured in remanence, (c) magnetization measured in an applied field of 40 Oe. (d), (e), (f) show hysteresis loops collected at different locations of the stripe. The middle section of the stripe contains a 90° domain (arrows show the direction of the remanent magnetization).

Fourfold crystalline anisotropy might be able to rotate the easy axis in the region surrounding the steps [2.12]. Thus in Figure 2-2 the local disturbance of the fourfold anisotropy induces a 90° domain in the middle region of the stripe.

2.4. Magnetization switching

The reorientation of the magnetization from an easy axis into the opposite direction by applying an external field is expected to happen through the formation of domains and
subsequent domain wall motion. The timescale for this process is nanoseconds. This is beyond the time resolution achieved in our experiment, which is about 1 millisecond. Thus, we see this reorientation as a sudden change in the Kerr signal at a certain field $H_C$. This coercivity field $H_C$ is known to be dependent on the thickness of the magnetic layer. In addition pinning of domain walls by defects in the underlying substrate makes the coercivity field very sensitive to substrate inhomogeneities. As shown by Weber et al. [2,12], variations in the magnetic anisotropies influence $H_C$. Here we investigate whether $H_C$ depends on the size of laterally structured ultrathin particles or not. In Figure 2-3 the coercivity field values $H_C$ of the particles shown in Fig. 2-1 are plotted. No significant dependence of $H_C$ on the particle size is detected. In most cases, the influence of the substrate is too strong and dominates any eventual size dependence. Note that $H_C$ varies within the same particle up to about 10 Oe. In the following $H_C$ is a mean value with an error of ±10 Oe.

As the switching is expected to happen through domain formation we try to induce domains in these particles by bringing them very close to the transition point. Therefore, we apply a reversing field very close to $H_C$ and map the magnetization of a formerly homogenously magnetized particle with the SKEM. Figure 2-4 reports a series of SKEM ima-

![Figure 2-3. Coercivity field $H_C$ of a set of circular Co dots (4AL) with diameter of 2, 4, 6, 8, 10 μm (left to right). $H_C$ is determined from square shaped hysteresis loops collected in a linescan through the center of the dots. No significant dependence of $H_C$ on the particle size can be observed.](image)
2.4. Magnetization switching

Images of a 10μm square shaped Co island. At each pixel the particle is saturated, then a reversing field is applied and the signal I(-H_{rev}) detected, then the particle is saturated in the negative direction and again a reversing field is applied and the signal I(+H_{rev}) measured. These signals are subtracted from each other. Therefore a white pixel means that the magnetization has not switched upon applying the reversing field, whereas at a black pixel the magnetization has switched. Grey pixels indicate no difference between the two signals. From one picture to the next, the reversing field is increased until the particle has completely switched. As one approaches H_{C}, the number of black pixels increases. However, there is no field value for which only a part of the island is homogeneously black or grey, i.e. the formation of domains might have occurred. Repeating the same measurement al-

**Figure 2-4.** Switching of a 10μm, 4 AL thick, square shaped island. Except the most left image all the other SKEM images where collected in an applied field close to H_{C}. In the white pixels the magnetization still lays in the initial direction, whereas in the black pixels it has already switched in the opposite direction. Grey means no magnetic signal at all.
ways results in an other pixel distribution. We conclude that the observed distribution is statistically and may result from the “chittering” of $H_C$ described in the former section and not from domain formation.

In SEMPA images (Scanning Electron Microscopy with Polarisation Analysis), the same samples, previously exposed to a reversing field of 99.9% of $H_C$, show no domain formation even at a resolution of 10nm [2.13-2.14].

### 2.5. Mutual interaction

Responsible for the formation of domains in a magnetic system is the demagnetizing field.

![Figure 2-5](image)

**Figure 2-5.** *(top) Remanent SMEK image of two circular dots with a distance of 2µm. The stray field of one particle is not sufficient to switch the remanent state of the other. *(bottom) The same dots in an external field. The left dot has switched its magnetization.*

In an in-plane magnetized two dimensional film this field approaches zero [2.15-2.17]. As
demonstrated in the former section, the demagnetizing field of in-plane magnetized particles is too weak to introduce domains in this particles. However, the stray field of an homogeneously magnetized particle could influence the behavior of a neighboring particle. The experiment to test the mutual interaction between two particles is reported in Figure 2-5. Two circular particles with a diameter of 2μm, thickness 3 AL and a distance between each other of 2μm were fabricated. The upper image is taken in remanence and shows both particles homogeneously magnetized along the [1-10] axis. Considering one particle as a magnetic dipole, the stray field of this dipole produces a negative field at the location of the other particle. The upper plot of Figure 2-5 clearly demonstrate that this stray field is smaller then the coercivity field $H_C$ of the particle. Thus both particles are magnetized in the same direction. However, as $H_C$ of the left-hand particle is slightly smaller then $H_C$ of the right-hand one, it is possible to switch the left-hand particle without switching the right-hand one. This is shown in the lower part of Figure 2-5.

The stray field of the right-hand particle — if sizable — helps the switching process of the left-hand particle. This should be seen as an asymmetry in the hysteresis loops of the left-hand particle. Within the accuracy of our experiment (±10e) no such asymmetry was detected.

### 2.6. Shape anisotropy

The dipolar - or magnetostatic - energy of a magnetic particle can be described in analogy to the electrostatic energy of a particle with a non vanishing surface charge density, where the surface charge density is proportional to the magnetization along the surface normal [2.15]. In this description, north and south pole of a magnetic particle are considered as magnetic charges. In a homogeneously magnetized particle with finite size there are always some magnetic charges at the boundaries. These are responsible for the stray field. In order to reduce this stray field the system tries to reduce the magnetic charges at its boundaries. This fact can be described as an additional anisotropy, the shape anisotropy.

In the case of a narrow stripe, the magnetization tends to be along the stripes long axis (magnetic needle), assuming that there are no other anisotropies present. When evaporating Co stripes onto a fourfold Cu(100) crystal, the shape anisotropy adds an uniaxial anisotropy to the magnetocrystalline anisotropy. It can be calculated according a techniques described by Politi et al. [2.18].

A SKEM experiment to probe the shape anisotropy of narrow stripes is represented in Figure 2-6. Figure 2-6(a) shows a remanent SKEM image of 9 stripes with a length of 40μm, a width of 1μm (aspect ratio R = 40) and a thickness of 7.7 AL each. The stripes are evaporated through a mask onto a Cu(100) with regular monoatomic steps along one of the [1-10] directions. In image 2-6(a) the [1-10] crystal axis and the steps are oriented vertically. $\alpha$ describes the angle between the steps in the substrate surface and the long axis of the stripes. From right to left, $\alpha$ increases 3° between two stripes. For all stripes, the remanent
FIGURE 2-6. A set of stripes with an aspect ratio of 40. (a) Remanent SKEM image of the stripes. (b) Linescan through the stripes taken with the field applied along the steps. Shown are the remanent magnetization (open circles) and $M$ at 600 Oe (full circles). (c) Corresponding $H_C$ determined from these easy axis loops. (d) The field $H_S$ determined from shifted loops collected during a linescan with the field applied perpendicular to the steps. No significant dependence of $H_S$ from the stripes orientation relative to the steps is found.
magnetization $M_R$ points along the steps in the substrate, irrespective of the relative orientation $\alpha$. In (b) an easy axis line-scan through a set of 20 stripes is plotted. Starting from a stripe with its long axis parallel to the steps (left) the angle $\alpha$ is increased up to 60° (most right stripe). Although there is a variation in the absolute magnetic signal, we find no stripe were $M_R$ points away from the step direction. Even stripes with their long axis perpendicular to the steps are magnetized along the steps. Thus, the shape anisotropy must be much smaller than the uniaxial anisotropy caused by the steps.

An estimation of the uniaxial anisotropy strength can be made from hysteresis loops collected perpendicular to the steps. The shift field $H_S$ is a good measure of the uniaxial anisotropy [1,1]. Therefore, changes in the uniaxial anisotropy should also be seen in $H_S$. In Figure 2-6(d) $H_S$ for the set of stripes in (a) is plotted. The substrate miscut cause an offset in $H_S$ in the range of 700e, whereas a calculation by Politi et al. [2,18] gives a value for the shape anisotropy of $H_S = 6 \sin^2 \alpha$ Oe/AL (R=40) leading for a $\Delta\alpha=60^\circ$ and 7.7AL to a $\Delta H_S$ of about 400e. Unfortunately the change of $H_S$ caused by inhomogenities of the step density is of the same magnitude as the expected shape anisotropy changes. Therefore a significant dependence of $H_S$ on the orientation $\alpha$ of the stripes could not be found.

For comparison, we plot the switching field $H_C$ determined from easy axis loops in Figure 2-6(c).
3. Phase transition on Co/Cu(100)

Magnetic phase transitions are of interest as they reveal much of the symmetry in a system [3.1-3.7]. It is well known that the magnetization exhibits a second-order phase transition from a ferromagnetic phase to a paramagnetic phase. The transition temperature, the Curie temperature $T_C$, is thickness-dependent [3.8-3.10] and is far lower in two dimensional films than in the corresponding bulks. A 1.5ÅL thick Co/Cu(100) film has a $T_C$ of $\approx 300$ Kelvin. This makes Co/Cu(100) suitable for experiments, as phase transitions can be measured without extensive heating or cooling, which might be connected with structural changes.

Most phase transition experiments have been performed on magnetic systems with only one symmetry breaking crystal field. This field can be either an uniaxial anisotropy field, like in the case of in-plane magnetized Fe/W(110), or a fourfold anisotropy, like in-plane magnetized Co films on a flat Cu(100) surface. Here we provide results of a system which combines both a fourfold symmetry breaking crystal field and a twofold one.

3.1. Concept of universality and scaling

In this section a brief summary of the concept of universality and scaling is given.

Near a phase transition the spontaneous magnetization $M_{SP}(T)$, the susceptibility $\chi(T)$ and the dependence of the magnetization $M(H)$ on external applied fields at $T_C$ follow a power law with specific exponents, the critical exponents [3.11]. These exponents can be used to separate real physical systems into so-called universality classes. According to the concept of universality, a class is specified by few properties, like e.g. the dimensionality, the range of the microscopic interactions and the dimensionality of the order parameter (e.g. the magnetization $M$). Two ferromagnetic systems which belong to the same universality class have the same critical exponents. Or vice versa, by determining the critical exponents experimentally one has a strong indication to which class the system belongs.

Three of these exponents are relatively easy to determine experimentally, namely $\beta$, describing the behavior of the spontaneous magnetization $M_{SP} = M(T,H=0)$ with $T \rightarrow T_C$,.
δ the exponent of the dependence of the magnetization \( M(T=T_C, H) \) on the magnetic field at the Curie temperature \( T_C \) and \( \gamma \), the exponent for the static susceptibility \( \partial M(T, H)/\partial H \). For a number of systems these exponents are either theoretically exactly known (e.g. \( \beta \) for

$$
\begin{array}{|c|c|c|}
\hline
 & 2D \text{ Ising} & 3D \text{ Ising} & \text{Mean Field Theory} \\
\hline
\beta & 1/8 & 0.3125 & 1/2 \\
\delta & 15 & 5 & 3 \\
\gamma & 1.75 & 1.25 & 1 \\
\hline
\end{array}
$$

Table 3-1. Theoretical values for critical exponents: \( \beta \), the exponent of the zero field magnetization, \( \gamma \), the exponent for the static susceptibility below and above the Curie temperature and \( \delta \), the exponent for the dependence of the magnetization on the magnetic field at the Curie temperature.

2D Ising [3.12]) or conjectured from numerical calculations (e.g. \( \delta \) for the 2D Ising model [3.13]). Table 3-1 summarizes their values for some important systems: the 2D Ising model, the 3D Ising model and the mean field theory.

Thermodynamically, a system is fully described by the equation of state. Therefore in addition to the critical exponents one has to find its equation of state. In the case of a ferromagnetic system the equation of state depicts the magnetization \( M \) as a function of the temperature \( T \) and the external applied field: \( M(T, H) \). The scaling hypothesis predicts that this function can be expressed in a specific form [3.14-3.17],

$$
M(t, H) = H^{1/\delta} \cdot M(\text{sgn}(t), H/|t|^{\beta\delta})
$$

where \( t = (T/T_C - 1) \) is the reduced temperature and \( M(\text{sgn}[t], H/|t|^\beta\delta) \) is the so called scaling function.

Experimentally \( M, t \) and \( H \) are measured.

By suitable plotting the measured \( M(t, H) \) data, i.e. using \textit{scaled} thermodynamic variables, one can graphically find the function \( M^\pm(x) \), i.e. determine the equation of state in the critical region. Here \( M^+ (M^-) \) represents the function for \( t > 0 (t < 0) \).

In a standard representation, \( M \) and \( t \) are scaled with \( H^{1/\delta} \) and \( H^{1/\beta\delta} \), respectively. By
using the variables $y = M/H^{1/\delta}$ and $x = t/H^{1/\beta}$, all measured $M(t,H)$ points should collapse onto one curve $y = f(x)$, thus defining the equation of state and the scaling function.

Another representation uses the scaled variables $H/M^\delta = h(x)$ with $h(x)$ being a function of $x = t/M^{1/\beta}$ [3.15-3.18]. The function $h(x)$ - the scaling function - is conjectured to be specific for a universality class and has been constructed (numerically) by D. S. Gaunt et al. [3.19], e.g. for the two dimensional Ising model.

We will plot our data points using both representations.

The concept of scaling gives also a relation between the critical exponents. They can be expressed in terms of two independent parameters, i.e. the scaling parameters and therefore only two independent critical exponents exist. The connections between the various critical exponents are given by the so-called scaling equations [3.15-3.18]. The relevant scaling equation in our experiment is

\[ \gamma = \beta \cdot (\delta - 1) \]

The predictions from the concept of universality as well as those from the concept of scaling can be confirmed experimentally. First the critical exponents $\beta$, $\delta$ and $\gamma$ have to be determined and compared with theoretical values. Second the experimental values should fulfill the scaling equation and the measured $M(t,H)$ data should collapse onto one curve when scaled and plotted according to the guidelines described above. Third, provided the values for the exponents can be assigned to a theoretical model and the theoretical scaling function is known, the scaled data should coincide with the theoretical scaling function.

Both the concept of universality as well as the scaling hypothesis have been tested on in-plane magnetized Fe/W(110) [3.20] with a strong inplane uniaxial anisotropy. Although microscopically Fe/W(110) does not fulfill the assumption of the ideal Ising model, i.e. only next neighbor interaction between localized spins with a infinite strong uniaxial anisotropy, it follows closely the theoretically predicted behavior for a 2D Ising model in the critical regime.

Here we test scaling and universality for a Co/Cu(100) system with a much smaller uniaxial anisotropy and an additional fourfold anisotropy.

In the second part of this chapter, we will focus on the question what happens when the lateral sizes of these films are reduced, i.e. wether finite size effects occur or not. The single domain particle presented in the former chapter are used to probe these finite size effects.
3.2. Symmetry breaking fields in Co/Cu(100)

Co evaporated on a perfectly flat Cu(100) single crystal exhibit fourfold anisotropy with the two [1-10] directions being equivalent magnetic easy axis [3,3].

A polished Cu(100) crystal with a miscut of $= 3^\circ$ with respect to the [100] direction has regular steps in its surface running along one of the [1-10] direction with an average spacing of 35 Å. This step arrangement breaks the fourfold symmetry [2,12].

It is known that Co films grown on such a substrate show a different magnetic response along the two perpendicular [1-10] directions [3.22-3.24]. This is also documented in Figure 3-1, which reports two magnetization loops taken parallel (a) and perpendicular to the steps (b). Both loops are taken on the same 5.5ÅL thick Co film. The hysteresis in Figure 3-1(a) displays a square shaped easy axis loop and indicates an easy magnetization axis...
running along the steps. In contrast, the loop collected perpendicular to the steps, see Fig.3-1(b), consists of two loops, which are separated by a field $2H_S$. The difference in the magnetic response originates from the presence of an uniaxial anisotropy [3.23]. While the direction parallel to the steps combines both the easy axis character of the fourfold cubic anisotropy with the easy axis character of the uniaxial anisotropy, the perpendicular direction unites an easy axis character with a hard axis character. An estimation of the symmetry breaking fields can be obtained from the shifted loop in Figure 3-1(b).

For small external fields the magnetization in (b) depends linearly from the external field $H$. In this regime the anisotropy field $H_{ani}$ (comprising the fourfold $H_U$ and the twofold crystal field $H_U$) equals $M_{sat}/s$. From the slope $s$ we estimate this anisotropy field to be about $H_{ani} = 1kOe$. In addition the shift field $H_S$ is linear in the uniaxial anisotropy and therefore a direct measure of the uniaxial field $H_U = H_S = 37.7Oe$ [2.7, 2.12]. Thus the main contribution to the in-plane anisotropy field $H_{ani}$ arises from the fourfold crystal field $H_U = H_{ani} = 1kOe$.

Compared to the uniaxial field of Fe/W(110): $H_U \approx 10kOe$ [3.25] the step induced twofold anisotropy in our system is very small. It will be interesting to see how 4-fold and 2-fold anisotropies influence the critical behavior and whether we observe a temperature at which one of the symmetry breaking fields vanishes.

### 3.3. Experiment

The aim is to measure the magnetization $M(T,H)$ for different temperatures and in various applied fields in the region of the magnetic phase transition.

As mentioned in the introduction, $T_C$ is thickness dependent. Small changes in the film thickness results in a rather large $\Delta T_C$. Typically for these ultrathin films near the onset of long-range order, changes in the thickness of 0.25ÅL causes a $\Delta T_C$ of about 80 Kelvin. However, a sudden jump at 1.8ÅL observed by Bovensiepen et al. [3.10] was not observed.

This strong thickness dependence of $T_C$ restricts the thickness range of the films and structures we can investigate. As in our set-up, temperatures between 177 K and 440 K can be achieved, the experimental thickness limits are 1.5ÅL and 2.5ÅL, respectively.

In the preparation chamber the copper crystal is cleaned and annealed and a Co film is evaporated onto it. The thickness is controlled by monitoring the evaporation time. For a Co film thickness of $\approx 1.5ÅL$ the transition occurs at about 300 Kelvin. For such a film the transition point can be reached without excessive heating or cooling. Thus we stop evaporation at this thickness. Next, the sample is transferred to the SKEM chamber where it is placed onto the Kerr stage and the laser spot is roughly minimized at a prominent feature. Then the sample is cooled with liquid nitrogen and the Kerr signal along the step direction is collected during reheating. The heating rate is kept constant at about 20 Kelvin
FIGURE 3-2. Typical magnetic loops collected near $T_C$ on a continuous Co/Cu(100) film. They are recorded along the [1-10] direction and parallel to steps, i.e., along the magnetic easy axis. An additional loop at $T_C$ is measured with a finer field resolution to obtain a better accuracy in determine the critical temperature $T_C$. 

$T = 0.97 \ T_C$

$T \approx T_C$

$T = 1.03 \ T_C$

magnetic field H (Oe)
per hour, while magnetization loops $M(H)$ and temperature are continuously measured. In order to avoid residual magnetic fields compensating coils are used to ensure that the hysteresis loops are symmetric. After each measurement the sample is cleaned and a new film or structure is evaporated in order to reduce effects which adsorbed residual gas might have on the magnetization.

Figure 3-2 shows typical loops collected near $T_C$. When approaching the critical point, $H_C$ gets smaller. For a better accuracy in determining $T_C$, additional loops with a finer field resolution are collected. Such an additional loop at $T \approx T_C$ is shown in the Figure 3-2. From these data, $M(T,H)$ curves can be extracted for every field value within the field range.

Figure 3-3 reports six $M(T,H)$ curves derived from a set of hysteresis loops. The remanent magnetization as well as the magnetization in five different applied fields is reported over a wide temperature range.

The following features are obvious from the plot:

**FIGURE 3-3.** A family of $M(T,H)$ curves obtained simultaneously during a phase transition. The data are extracted from hysteresis loops collected while heating up the sample.
(i) The magnetization at low temperatures decreases almost linear in $T$ independent of the applied field.

(ii) The remanent magnetization $M_R$ vanishes sharply at a well-defined temperature which is identified with the Curie temperature $T_C$.

(iii) Upon application of small magnetic field, the magnetization attains sizeable values even well above $T_C$.

The sharp drop of $M_R$ at $T_C$ as well as the strong response of the magnetization to small applied fields above $T_C$ is typical for 2D systems (see also on page 34).

The linear temperature dependence of the magnetization $M(T)$ at low temperature suggests to be dominated by spin-wave excitations. In this regime the zero field $M_R(T)$ curve can therefore be described with a spin wave derived formula [3.26]:

$$\frac{M_R(T)}{M(0)} = 1 - \frac{T}{4\pi\Gamma} \ln \frac{T}{\sqrt{\lambda} \cdot \Lambda}$$

Here, $\Gamma$ is the exchange coupling between the spins, $\Lambda$ is the easy plane anisotropy causing the magnetization to lay within the film plane and $\lambda$ is an anisotropy in the film plane. According to this formula the low temperatures data are fitted and the values for $\Gamma$, $\Lambda$ and $\lambda$ determined. The fit for the data in Figure 3-3 yields $\Gamma \approx 250K$ and $\sqrt{\lambda} \cdot \Lambda \approx 1.5K$.

With $H_{ani} = M_{sat}/s = \lambda/g\mu_B S = 1kOe$ (see previous section) follows for $\lambda = 0.2K$ and $\Lambda = 10K$.

The extrapolated value for the magnetization $M$ at $T=0$ Kelvin is used in the following to normalize the Kerr signal data.

### 3.4. Critical Exponents

From the data we will now determine the critical exponents $\beta$, $\delta$ and $\gamma$ and compare them with known values of models with similar symmetry and dimension.

One important task in determining the critical exponents is to precisely localize $T_C$. It is defined as the temperature at which the spontaneous magnetization $M_{SP}$ vanishes. A priori it is not clear that we can identify the remanent magnetization $M_R$ measured with the SKEM with this spontaneous magnetization. However SEMPA measurements on ultrathin magnetic systems during phase transition including flat Co/Cu(100), exclude domain formation as the reason for the vanishing $M_R$. In addition D. Kerkmann et al. [3.3] show that $M_R$ does not approach zero because the system overstep the blocking tempera-
3.4. Critical Exponents

ture of a superparamagnetic state. In a superparamagnetic state the response of the mag-

\begin{figure*}
\centering
\includegraphics[width=\textwidth]{figure3-4}
\caption{Data from Figure 3-3 replotted to determine the critical exponents $\beta$, $\delta$ and $\gamma$. The exponents $\beta = 0.18$, $\gamma = 2$ and $\delta = 12$ are provided by power law fits through the data (solid lines).}
\end{figure*}
3.4. Critical Exponents

Netization to applied fields is strongly time dependent. Changing the sweeping rate of the applied field by a factor 10 does not alter the behavior of the system in our experiment. We identify the temperature were MR vanishes as $T_C$.

The next step is to determine the critical exponents $\beta$, $\delta$ and $\gamma$.

Therefore the data from Figure 3-3 are plotted in a suitable way. Figure 3-4(a-c) reports the corresponding plots.

Near the critical point the $M_R(T)$ curve can be described with a power law using an exponent $\beta$. Consequently, we fit our data in the critical region $0.95T_C < T < 1.05T_C$ according to

$$M_R(T) \propto \left(1 - \frac{T}{T_C}\right)^\beta = (-t)^\beta$$

with $t = (T/T_C - 1)$ the reduced temperature.

$T_C$ is determined accurately according a procedure described by Dürr et al. [3.1]. The data points are plotted in a log($M$) versus log($1-T/T_C$) representation and the best value of $T_C$ maximize the range over which the data points form a straight line, i.e. the range over which $M(T)$ follows a power law. With this procedure $T_C$ is restricted to $237 \pm 1$ K. In addition the slope in the log-log plot provide the value for $\beta = 0.18 \pm 0.03$. The error in $\beta$ is mainly caused by the error of $T_C$. Figure 3-4(a) show the remanent magnetization vs. the reduced temperature together with the power law fit.

Next we plot $[M(T,H=0.1Oe)-M(T,0)]/M(T=0,H=0)$. The plot is shown in Figure 3-4(b). In the limes $H \to 0$, this quantity corresponds to the static susceptibility $\chi$. The corresponding power law has the form

$$\chi_-(T) = \Gamma_-(1 - \frac{T}{T_C})^{-\gamma'} \quad \text{for } T < T_C$$

$$\chi_+(T) = \Gamma_+(\frac{T}{T_C} - 1)^{-\gamma} \quad \text{for } T > T_C$$

Following the scaling hypotesis we set $\gamma = \gamma'$. The solid line in (b) displays the best fit to the data yielding $\gamma = 2$ and $\Gamma_+ / \Gamma_- = 4.3$. We will see in the next section that $\gamma$ and $\Gamma_+ / \Gamma_-$ can be determined more precisely simply by replotting the data in a suitable representation according to the scaling hypotesis.
Also shown in Figure 3-4 is the critical isotherm $M(T=T_C,H)$. The plot (c) includes the data and a fit corresponding to the power law

$$M(H) \propto H^{1/\delta}$$

The data are best fitted with $\delta = 12$.

In summary we found the critical exponents $\beta = 0.18$, $\gamma = 2$ and $\delta = 12$. These values fulfill the scaling equation. However their interpretation is not straightforward.

A number of experiments [3.28-3.30] on ultrathin films have determined the critical exponent $\beta$. Mostly, a $\beta=1/8$, characteristic of 2D Ising model, was found for systems with uniaxial anisotropy and a $\beta=0.23$ for those systems with a 4-fold symmetry breaking field. There are some exceptions with $\beta$ being in the range 0.15-0.18. These systems had a very weak anisotropy and are generally ascribed to the Ising model with a nonuniversal correction to $\beta$ [3.29].

Theoretically the situation is unclear. A renormalization group analysis by Jose et al. [3.31] shows that the presence of a fourfold field perturbation to an infinite XY model gives rise to nonuniversal exponents. In contrast, Bramwell et al. [3.32-3.33] calculate that the effect of a 4-fold perturbation on an idealized finite size XY model is irrelevant to its critical behavior. They demonstrate by means of Monte Carlo simulation that for real films with fourfold or sixfold in-plane anisotropy the finite size XY model is adaptable, leading to a $\beta = 0.23$.

Obviously our system belongs neither to the Ising universality class nor to the finite size XY-model. A pure Ising like transition is excluded by the values of the critical exponents and an isotropic state (XY-model) is neither reached above nor below $T_C$ (as we will see in the next chapter). The transition is probably dominated by the uniaxial crystal field, but the fourfold symmetry breaking cannot be neglected and causes a non-universal behavior. Additional information can be obtained by measuring the complementary in-plane component, i.e. the magnetization perpendicular to the easy axis during a phase transition. The corresponding experiments are described in the next chapter.

### 3.5. Scaling/Equation of state

In this section we will represent the data using the exponents $\beta$, $\delta$ and $\gamma$ to construct suitable scaled variables and compare the resulting plot with the equation of state of an appropriate model.
According to the scaling hypothesis, the data $M(T,H)$ should collapse onto one single curve when suitable scaled thermodynamic variables are chosen. The data from Figure 3-3 are replotted in Figure 3-5. (a) and (b) show two different representations of the scaling function. In (a) the variables $t$ and $M$ are scaled with $H^{1/\delta}$ and $H^{1/\beta}$, respectively. The best data collapsing is realized using the exponents $\beta=0.18$ and a value for $\delta$ between 10-14.

The representation in (b) is displayed as a log-log plot with the variables $x=t/M^{1/\beta}$ and $y=H/M^{\delta}$ already mentioned in the introduction and the values of $\beta$ and $\delta$ from plot (a). In this representation the point $h(x=0)$ represents the critical isotherm $M(t=0,H)$ of Fig. 3-4(c) and the point $x_0$ the remanent magnetization $M_R(t)$ of Fig. 3-4(a). For $x \to \infty$ the scaling function is proportional to $\gamma \cdot x$. A fit through the linear part of the plot yields $\gamma=2.1$, which confirms the finding $\gamma=2$ in the former section. The solid line displays the numerically constructed function $h(x)$ by D. S. Gaunt et al. [3.19] for the 2D Ising model. From Figure 3-5 two main conclusions can be drawn. First, data collapsing is realised with $\beta$, $\delta$ and $\gamma$ fulfilling the scaling equation. Second the equation of state is determined graphically. Yet, the experimentally found scaling function does not coincide with the theoretical scaling function $h(x)$ of the 2D Ising model. This is not surprising as the critical exponents do not match with the Ising exponents.
3.6. Limiting the lateral size

Theoretically the correlation length $\xi$ between spins diverges to infinity as a 2D system approaches the critical temperature $T_C$ [3.34-3.37]. Thus the region of correlated spins grows infinitely large at $T_C$. Nevertheless, within these blocks of correlated spins, fluctuations on every length scale cause the remanent magnetization $M_R$ to vanish at $T_C$.

In real physical systems the spin block will have a finite size even at $T_C$. The question is whether the real maximum spin block size is larger than the size of our smallest achievable particles. If so, the particles boundaries would hinder the spin blocks in growing and one would expect a finite size effect in the behavior near the critical region.

An estimation of the spin block size can be made with the help of the static susceptibility $\chi = \lim_{H \to 0} \frac{[M(H) - M(H = 0)]}{H}$. According to arguments based on the Renormalization Group method, $\chi$ and $\xi$ are related by the equation [3.36-3.37]:

$$\chi(T) \propto \xi^2(T) \frac{C}{T_C}$$

The limit $H \to 0$ is experimentally not realized ($H = 0.10$Oe). However, we can use the data for a rough estimation of $\xi$.

The magnetic field $H$ is expressed in units of $4\pi M_S$, where $M_S$ is the spontaneous magnetization of bulk Co in Gauss: $4\pi M_S = 18532$ Gauss. With $H = 5.4 \cdot 10^{-6} \cdot 4\pi M_S$ (corresponding to 0.1 Gauss) this results in a maximal value for $\chi_{\text{max}} = 26088$. Taking the Curie constant $C = 1\text{K}$ results in $\xi_{\text{max}} = 2500$ lattice constant, $a$. For a Cu lattice with $a = 3.61\text{Å}$ this results in a block size of 0.9µm. This value is in good agreement with previous findings of 1µm maximal correlation length [3.34]. Yet, this value is below the achievable sizes in our experiment. However, our estimation is very rough, especially as $H = 0.1$Oe is a rather large field to evaluate $\chi$ and it will still be interesting to see if our particles will show a different behavior near $T_C$ compared to the extended film.

To investigate the critical behavior of ultrathin particles, we use the same procedure which we have already used on extended films. First, the structures have to be localized on the crystal before a phase transition measurement on such a particle can be accomplished. This is performed by collecting a reflectivity and a magnetization image of the interesting area with the SKEM. Figure 3-6 shows typical particles on which phase transitions are measured. Visible are particles with three different geometries, namely 4x4µm² and 10x6µm² islands in (a) and (b) and 44x16µm² islands in (c) and (d). The arrangement shown in (a) and (b) was originally designed to study the mutual interaction between neighboring particles. Thus the distances between the islands decreases from 8µm down to 2µm from the top of the image to the bottom. To exclude effects from mutual interactions, phase transitions were performed only on islands with a distance larger than 4µm.
3.6. Limiting the lateral size

FIGURE 3-6. SKEM image of particles with three different geometries. (a) and (b) show four sets consisting of three islands each. Thereby a 4x4μm² is sandwiched between two 10x6μm² island with decreasing distance from 8μm to 2μm in 2μm steps (top to bottom). (c) and (d) reports two 4x4x16μm³ islands. (a) and (c) display the remanent magnetization. (b) and (d) are reflectivity images, showing the topographic contrast. The structures are 1.7AL thick and the images are collected at 290 Kelvin.

All investigated particles exhibit a single domain remanent state in the ferromagnetic phase. This fact is well documented by the remanent images in Fig. 3-6(a) and (c). In addition, square shaped hysteresis loop are collected on these particles. Structural defects, which can alter dramatically the magnetic behavior, are easily localized and avoided. In Fig. 3-6(c) and (d) such a defect appears in the lower part of the right particle.
3.6. Limiting the lateral size

Figure 3-7 reports the data evaluated from hysteresis loops. Shown are the results for a 10x6μm² island (a) and a 4x4μm² island (b). At first glance the particles exhibit the same features as reported in Figure 3-3 for extended films. We evaluate Tₐ for these measurements and normalize the data with respect to M(T=0,H=0) using the procedure discussed earlier. For comparison the scaled magnetization curves Mₐ are plotted together with the Mₐ curve from an extended film. The result is shown in Figure 3-8. Although the data from the particles include more noise, there might be a short tail in the remanent magnetization curve of the 4x4μm² particle above Tₐ. Such a tail is ascribed to a finite size ef-
3.6. Limiting the lateral size

Figure 3-8. Comparison between the zero field magnetization curves $M_R(T)$ of an extended film and laterally limited particles. The Kerr data are normalized with respect to $M(T=0,H=0)$, (see text). A very small tail might be visible above $T_C$ in the $4\times4\mu m^2$ particle $M_R(T)$ curve. Such a tail is typical for a finite size effect.

fect, since the finite size of the system suppresses the development of the correlation length $\xi$ and thus the missing long range fluctuations in the excitation spectrum causes a finite magnetization at $T_c$.

In a more detailed analysis we determine $\beta$ for the $4\times4\mu m^2$ and $10\times6\mu m^2$ islands by fitting the data. The fits yield $\beta=0.18$ for the larger and $\beta=0.20$ for the smaller island. Again the data are replotted in two different representations. Thereby, $\delta$ was varied to realize best data collapsing. The results are shown in Figure 3-9. In (a) the data are those of the $10\times6\mu m^2$ island and in (b) those from the $4\times4\mu m^2$ island. The exponents $\beta$ and $\delta$ yielding the best data collapsing on the left representation are slightly different from the continuous film values although within the error. The exact values are in (a): $\beta=0.18$, $\delta=18-20$ and in (b): $\beta=0.2$ and $\delta=18$. Using these values for the representation on the right and fitting the linear part of the plots in (a) and (b) yields $\gamma=3$ and $\gamma=3.2$ respectively. Again the exponents fulfill the scaling equation and the scaling hypothesis is positively tested. The full dots represent a scaled $M(t,H)$ curve from Figure 3-5, i.e. the equation of state of the extended film.

Figure 3-9 provides the graphic solution for the equation of state of the particles. Comparing this result with the curve obtained for an extended film (Figure 3-5) two conclusions
can be drawn. The film as well as the particles scale as predicted by the scaling hypothesis. However, their behavior near $T_C$ is not universal. Considering the large noise in the particle data, the graphical solution for the equation of state might be the same for particles and film. The larger value for $\gamma$ in Figure 3-9 might be a result of a larger error in determining $\beta$ and $\delta$ from the particles data.
4. Two- and fourfold symmetry breaking fields near phase transition

Again we consider the magnetic system Co/Cu(100) with a 3° misfit with respect to the [100] direction. In the previous chapter we have measured the easy axis magnetization \( M_{//} \) during a magnetic phase transition to classify the system. In this chapter we will mainly focus on the magnetic component perpendicular to the easy axis, \( M_{\perp} \).

From magnetization loops collected along this 'intermediate' axis it is possible to deduce the fourfold \( K_1 \) and the uniaxial \( K_U \) anisotropy [4.1], which are both present in our magnetic system. Thus, by measuring perpendicular loops during a magnetic phase transition, we also record the temperature behavior of these anisotropies. Moreover, an eventual transition to an isotropic behavior would immediately manifest, as both in-plane components become equal in the isotropic phase.

In chapter 3 we have already determined \( K_1 \) and \( K_U \) for a 5.5 AL thick Co film at room temperature. The strength of the anisotropy fields were \( H_1 = 1kOe \) and \( H_U = H_S = 50Oe \) and the corresponding anisotropy constants were \( K_1 = 0.1K \) and \( K_U = 0.01K \).

4.1. Experiment

During evaporation of Co onto a Cu(100) crystal, magnetic hysteresis loops are collected simultaneously along the monoatomic steps resulting from the misfit and perpendicular to the steps. We use two identical Kerr setups to measure both components. The spot sizes for both components are about 40\( \mu \)m and the laser beams are roughly aligned on the same spot on the crystal. Magnetic fields can be applied with two perpendicular oriented helmholtz coils. The time is monitored during evaporation and the thickness of the resulting film is determined with Auger electron spectroscopy (AES). Then the thickness scale is calculated. Figure 4-1 reports such a measurement. The graph shows twenty loops, ten displayed for each direction in the thickness range 1.4AL - 1.95AL. From left to right and from top to bottom the thickness is increased by \( \approx 0.7AL \) per step.

The easy axis behavior of the magnetization near the onset of ferromagnetic order is ex-
FIGURE 4-1. Hysteresis loops collected perpendicular and parallel to periodic monoatomic steps in the substrate. Both components are recorded simultaneously during film growth using two independent Kerr setups in transversal mode.
tensively discussed in the previous chapter. Long range order manifests itself by a non vanishing remanent magnetization $M_R$. From the lower part of Figure 4-1, showing the easy axis loops, we estimate the onset of ferromagnetic long range order at a thickness of $\approx 1.5\text{AL}$. At the same thickness the slope $s$, indicated in the upper part of Figure 4-1 (showing the $M_\perp$-loops), reaches its maximal value. This slope $s$ provides the linear response of $M_\perp(H)$ to small applied fields $H$ and corresponds to the perpendicular static susceptibility $\chi_\perp = \lim_{H \to 0} [M(H) - M(0)]/H$.

In Figure 4-2 the slope $s$, i.e. the susceptibility $\chi_\perp$ (open circles) is plotted versus film thickness. Also shown in Fig. 4-2 is the quantity $[M_\parallel(H) - M_\parallel(0)]/H$ with $H=50\text{e}$ (full circles). In the limit $H \to 0$ this quantity corresponds to the easy axis susceptibility $\chi_\parallel$. To compare $M_\perp$ and $M_\parallel$ (resp. $\chi_\perp$ and $\chi_\parallel$), which are measured with two different Kerr set-ups, we normalize both Kerr signals with their corresponding saturation signal at a film thickness of $7.7\text{AL}$.

Both components reveal a pronounced peak in susceptibility at a thickness $d_C$. Below, as well as above $d_C$ both susceptibilities vanishes but their decline is different. Thus in this range the response to small applied fields is anisotropic. We conclude that in the critical region there is an uniaxial magnetic anisotropy present, which persists over the phase tran-

**Figure 4-2.** Thickness dependence of the susceptibility along the easy in-plane axis $\chi_\parallel$ (full circles) and perpendicular to the easy axis $\chi_\perp$ (open circles). The peak in $\chi_\parallel$ indicates the onset of ferromagnetic order. $\chi_\perp$ corresponds to the initial slope $s$ of perpendicular $M_\perp(H)$ loops and $\chi_\parallel$ corresponds $[M_\parallel(H) - M_\parallel(0)]/H$ evaluated in a field $H=50\text{e}$. 
In the former chapter, phase transition were measured by varying the temperature at a given film thickness. This represents a more accurate way to measure the critical behavior of a ferromagnetic system because experimentally it is much simpler to tune the temperature than to tune the film thickness. Thus, we stopped evaporation at or slightly before the onset of ferromagnetic order so that the resulting film thickness was about 1.5AL. Subsequently, we cooled the film with liquid nitrogen until it reached ferromagnetic order and collected both magnetic components while reheating the film. Unfortunately we picked up too much noise and the data could not be analyzed. Thus, we changed to the SKEM setup already proved useful in measuring phase transitions. However, in this setup the magnetic component has to be measured one after the other. After measuring one component the sample has to be rotated by 90° in order to measure the second component.

![Graph](image)

**Figure 4-3.** Effect of film aging on the Curie temperature. Shown are the remanent $M_H$ (open circles) and $M_H$ in 50e applied field (full circles) for two phase transitions recorded on the same Co/Cu(100) film. One transition is measured directly after film growth and the other one after repeated cooling and heating in the range 180K<T<320K. Thereby, the pressure never exceeds $1x10^{-8}$mbar. $T_C$ drops slightly.

We start with the easy magnetization axis and, after cooling the sample a second time, measure the component perpendicular to the steps. In doing so the transition temperature $T_C$ varies slightly but apart from that the cooling-reheating cycles do not affect the critical
FIGURE 4.4. Magnetization curves vs temperature on CuCf(100) substrate.

The shift field $H_s$ is no longer defined and the magnetization approaches $M^\infty$ as the shift field $H_s$ is no longer defined and the magnetization approaches $M^\infty$. Figure 4.4 reports the temperature dependence of the magnetization and (c) shows the field dependence of the magnetization component perpendicular to the easy axis. The insets in (b) show typical loops from above and below the transition temperature. (a) shows the in-plane component parallel to the easy axis, with a $k$-axis inset and (b) shows the in-plane component perpendicular to the easy axis. The inset shows the field dependence of the magnetization component parallel to the easy axis.
behavior of the film. This is demonstrated in Figure 4-3, showing two easy axis transitions recorded on the same film. One is measured directly after film growth and the other is collected after three cooling-reheating cycles. At this time the Co film was about one day in a pressure of $\approx 5 \times 10^{-11}$ mbar, whereby $T_C$ drops $= 15$ Kelvin.

Data obtained from the analysis of hysteresis loops are presented in Figure 4-4. (a) represents the data along the magnetic easy axis and (b) those perpendicular to it. Both measurements were performed at the same 1.7AL thick film. Along the easy axis the transition is characterized by a sharp drop of $M_R$ (open circles) at $T_C$. Fitting leads to a critical exponent $\beta = 0.19$ in accordance with the findings in the previous section. The graph in (b) displays $M_L(30\text{Oe})$ (full circles) and $M_L(10\text{Oe})$ (open circles), with the insets showing typical loops collected below and above the transition temperature. Eventually $M_L(H)$ shows no distinguishable shifted loops but continuously approaches the saturation value. At low temperatures, the applied field was not sufficient to saturate the sample perpendicular to the steps and therefore the $M_L(300\text{e})$ curve remains nearly constant.

In addition, Figure 4-4(c) shows a plot of the shift field $H_S$ evaluated below the transition temperature. A linear extrapolation of $H_S$ (solid line) suggest that $H_S$ vanishes clearly above the transition temperature. The connection between $H_S$ and the anisotropies $K_U$ and $K_I$ is discussed in section (see chapter 4.3, on page 45) First we will focus on $\chi_{\perp}$ and $\chi_{//}$.

### 4.2. Susceptibility

As mentioned in the former section, the susceptibility $\chi_{\perp}$ is represented by the slope $s$ in the linear part of the $M_L(H)$-curves whereas the corresponding value along the easy axis is given by $[M_{//}(H,T)-M_{//}(0,T)]/H$ in the limit $H \to 0$. We plotted $\chi_{\perp}(T)$ (open circles) together with $[M_{//}(H,T)-M_{//}(0,T)]/H$ evaluated in a field of 50e (full circles). The result is presented in Figure 4-5. Both curves show a pronounced peak at a slightly different temperature. Such a shift in $T_C$ is also observed when subsequently measure two easy axis transitions at the same film (compare Figure 4-3). Thus, we consider this temperature shift to be caused by film aging and not significant for comparing the susceptibilities. Obviously the shape of the curves are different both below the transition temperature as well as above. While $[M_{//}(H,T)-M_{//}(0,T)]/H$ develops a sharp narrow peak, the susceptibility $\chi_{\perp}$ develops slower and over a wider temperature range. It is only at about $= 10$ Kelvin above $T_C$ that both susceptibilities become equal. This is about the temperature where $H_S$ vanishes in Figure 4-4(c). Again the system shows anisotropic behavior in the critical region. An explanation of the peak in $\chi_{\perp}$ is given in [4.2-4.4]. The author attribute the peak to topological excitation, i.e. a combined effects of domain walls and vortices.
**4.3. Anisotropies**

It is possible to quantitatively determine the anisotropies in a system from the magnetization curve $M(H)$ along the intermediate axis.

Minimizing the Gibbs free energy with respect to the magnetization $M$ leads to the equilibrium magnetization $M(H)$ in any applied field. The case of an in-plane magnetized system with a fourfold anisotropy $K_4 > 0$ plus a twofold anisotropy $K_U > 0$ is extensively studied in [4.1]. The authors classify the hysteresis according to the ratio of the anisotropies $K_4/K_U$. Choosing the intermediate axis, i.e. the axis perpendicular to the uniaxial anisotropy as the quantization axis, a dominant $K_4$ leads to shifted $M(H)$ loops characterized by an initial slope $s$ and a shift field $H_S$.

In contrast a dominant $K_U$ results in a second order transition, where the magnetization continuously approaches the saturation value. Here again the $M(H)$ curve is characterized...
by the slope $s$ and a field $H_{\text{sat}}$ where the magnetization reaches saturation.

The authors provide a formula for the initial slope: $s = \frac{M_{\text{sat}}}{2(K_U + K_I)}$ in both regimes.

As the ratio $K_I/K_U$ changes the interpretation of $H_S$ and $H_{\text{sat}}$ in terms of $K_U$ and $K_I$ changes also. For dominant $K_I$, $H_S$ is to first order proportional to $K_U$. In the regime $K_I \approx K_U$, the shift field $H_S$ is no longer proportional to $K_U$. For $K_I/K_U < 0.2$, (dominant $K_I$) a simple relation is given for $H_{\text{sat}}$: $H_{\text{sat}} = 2(K_U - K_I)$.

![Figure 4-6. Anisotropy vs. temperature in the critical region. The data points are obtained by analysing the saturation field $H_{\text{sat}}$ and the initial slope $s$ of hysteresis collected perpendicular to the easy axis. $s$ as well as $H_{\text{Sat}}$ are not defined over the whole measured temperature range. We used the relation $K_U + K_I = M_{\text{sat}}/2s$ for the low temperatures and $K_U - K_I = H_{\text{sat}}/2$ for elevated temperatures (see also Figure 4-4).](image)

We will now apply this findings to our data. Obviously in our experiment we observe a transition from shifted loops to continuous loops. We separate three temperature regimes. At low temperatures shifted loops are measured (dominant $K_I$). For higher temperatures the loops are no longer shifted (dominant $K_U$) and finally above $T_C$ we measure paramagnetic loops.

Accordingly we determine from the slope $s$ the quantity

$$K_U + K_I = \frac{M_{\text{sat}}}{2s}$$
In the middle temperature regime, where the M(H) loops suggest a continuous approach of the saturation magnetization $M_{\text{sat}}$ at a field $H_{\text{sat}}$, we evaluate the quantity

$$K_U - K_1 = \frac{H_{\text{sat}}}{2}$$

Figure 4-6 shows the evaluated quantities vs. temperature. The temperature range used to determine $K_U - K_1$ is quite small. Nevertheless, there seems to be a linear dependence on temperature in this temperature range. The solid lines represent a linear fit through the data. At the point where both curves cross each other the fourfold anisotropy vanishes. This is also documented in Figure 4-7, where $K_1$ and $K_U$ are evaluated from the curves in Fig.4-6 and plotted vs the temperature. The plot suggest that $K_1$ vanishes really close to the critical point, yet $K_U$ persist over the phase transition.

This findings are in line with the interpretation of the phase transition in chapter 3.4. The system seems to undergo a crossover from a fourfold symmetry phase to a twofold symmetry phase slightly below the Curie temperature $T_C$. However, the uniaxial symmetry dominates the critical behavior.

**Figure 4-7.** Calculated values for the uniaxial anisotropy $K_U$ and the fourfold anisotropy $K_1$ in the critical region using the fit in Figure 4-6. The temperature where the fourfold anisotropy $K_1$ vanishes is close to the critical temperature $T_C = 265K$ (peak in $\chi_{\perp}$ in Fig. 4-5).
References

1 Chapter


2 Chapter


3 Chapter


4 Chapter

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Dank


Ohne URS MAIER und ANDREAS VATERLAUS wäre die UHV-Kammer wohl immer noch ein Buch mit sieben Siegeln für mich. Ihr habt mich in die Geheimnisse der Experimentalphysik eingeführt und auch später seit ihr immer mit Rat und Tat da gewesen, wenn Probleme physikalischer (Andreas) oder technischer Art (Urs) auftauchten. Euch beiden ein ganz herzliches Danke!


Meinen Mitstreitern um die begehrte Doktortrophäe STEFAN, CHRISTIAN STAMM und YVES möchte ich auch herzlich danken. Die Konferenzen und anschliessenden 'Bildungsaufenthalte' mit Stefan und Christian waren immer sehr kurzweilig, auch wenn die Harmonie zwi schendurch etwas verloren ging. Yves, dank deinen Tips in Sachen Elektronik sahen die Hysteresen etwas weniger verrauscht aus!

Wertvoll und abwechslungsreich waren auch immer die Kontakte mit dem 'F-Stock'. MEHMET, BENI und ANDREAS vielen Dank für eure Tips und Anregungen. Auch wenn die Erkenntnisse aus unseren Gesprächen nicht immer direkte Anwendung in meinem physikalischen Schaffen fanden, so waren sie doch sehr aufschlussreich.

Last but not least, möchte ich mich noch bei meinen beiden Diplomanden VOLKER und SAMI bedanken. Viele der Messungen in dieser Arbeit werden euch bekannt vorkommen. Ihr habt mich immer wieder mit eurer Hingabe und Begeisterung überrascht. Egal ob zum x-ten mal die Kammer belüftet, eingepackt und ausgeheizt wurde oder wochenlang dieselben verrauschten Hysteresen gemessen wurden - nie war eine Spur von 'Tristesse' zu spüren..... ! Es hat mir mit Euch Spass gemacht!