Micromagnetism in the Ultrathin Limit

A dissertation submitted to the
SWISS FEDERAL INSTITUTE OF TECHNOLOGY ZURICH
for the degree of
Doctor of Sciences
presented by
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2005
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ZUSAMMENFASSUNG


Teil I

Eine starke uniaxiale Anisotropie sorgt bei ultradünnen Eisenfilmen auf Cu(100) dafür, dass die Magnetisierung senkrecht zur Filmebene gerichtet ist. Durch das gleichzeitige Vorhandensein von Austausch- und Dipolwechselwirkung zerfällt die Magnetisierung der Filme in entgegengesetzt gerichtete Domänen. Der Grundzustand besteht aus Streifendomänen.


In Modellrechnungen zur Entwicklung von Streifendomänen wird die Temperaturabhängigkeit der Domänenbreite oft vernachlässigt. Das Experiment zeigt, dass die Änderung der Domänenbreite nahe der Curie-Temperatur $T_c$ proportional zu $(T_c - T)^2$ ist. Einfache Modellrechnungen vermögen dieses Verhalten zu reproduzieren. Die grösse Domänendichte bei höheren effektiven Temperaturen stellt möglicherweise einen der zusätzlich erforderlichen Freiheitsgrade dar.


In Filmen mit einer Dicke, bei der $T_c$ grösser oder gleich der Raumtemperatur ist, werden die Hochtemperaturstreifen bei einer Temperatur knapp unterhalb von $T_c$ beweglich. Dieser dynamische Übergang erinnert an einem Glasübergang. Das Auftreten eines Glasübergangs innerhalb eines engen Temperaturbereichs um $T_c$ ist für Coulomb-frustrierte Ferromagneten vorausgesagt worden. Be- findet sich in Filmen $T_c$ unterhalb der Raumtemperatur, so trennt ein stark ungeordnetes, wenn auch stationäres Domänennuster die Hochtemperaturstreifen von der paramagnetischen Phase. In dieser Phase sind Orientierungs- und Positionsordnung kurzreichweitig. Ein Zusammenhang mit der beweglichen oder der paramagnetischen Phase bleibt zu untersuchen.

Teil II

Die Domänenbreite in ultradünnen Eisenfilmen auf Cu(001) kann genau eingestellt werden. Dies wird für die Untersuchung des Übergangs vom mehrdomänen zum eindomänen Zustand in seitlich begrenzten Strukturen benutzt. Es bestätigt sich, dass der eindomäne Zustand eintritt, sobald die seitlichen Abmessungen einer Struktur in der Größenordnung der Domänenbreite liegen.
Teil III

ABSTRACT

This thesis deals with thin magnetic films, either extended or laterally confined. A Scanning Electron Microscope with Polarization Analysis (SEMPA) is used to image the magnetization vector in the films. The thesis consists of three parts. In Part I, the magnetic domain pattern in extended ultrathin Fe films on Cu(001) is imaged. Laterally confined Fe films on Cu(100) are the subject of Part II, and the profile of charged walls in thin Fe films on W(110) is investigated in Part III.

Part I

The system of ultrathin Fe films on Cu(001) is characterized by a strong uniaxial anisotropy that forces the magnetization to align along the axis perpendicular to the film plane. Due to the competition of exchange and dipolar interactions, the magnetization of the films breaks up into oppositely magnetized domains. The ground state consists of stripe domains.

Here, it is shown that the stripe domains are only one of a series of domain configurations. The domain pattern varies both as a function of film thickness and temperature. A unified description is obtained with the concept of an effective temperature. As the effective temperature increases, the stripe domains with two-fold symmetry give way to a labyrinthine pattern with six-fold symmetry. At even higher effective temperatures, however, the labyrinthine pattern is replaced by another stripe domain pattern. This decrease of the symmetry as the effective temperature is increased has to be balanced by additional degrees of freedom in the high-temperature stripe phase. A quantity often neglected in model calculations for the evolution of stripe domains is the variation of the domain width with temperature. Experimentally, the variation of the domain width near the Curie temperature $T_C$ is found to be proportional to $(T_C - T)^2$. This behavior is reproduced by a simple model calculation. The higher domain density at high effective temperatures may provide one of the additional degrees of freedom required.

A number of theoretical models have suggested that transitions between different phases in two-dimensional modulated systems be mediated by defects. Here, the transitions are followed step by step and on a microscopic level. Indeed, defects are found to play an important role. The loss of the orientational order of the low-temperature stripe phase during the transition to the labyrinthine phase is initiated by the creation of new domain segments perpendicular to the existing stripe domains. This process can also be described as the creation of disclination pairs that then dissociate. The creation of disclination pairs is only possible because the mean domain width is reduced as the effective temperature is raised. The ensuing transition to the high-temperature stripes is mediated by knee-bend instabilities.
New domain segments are formed at knee bends and then proceed along a common direction to increase the overall domain length and thus reduce the mean domain width even further. The process can be described as the decay of higher-order dislocations into simple dislocations with a lower total energy. The individual phases are characterized with respect to their positional and orientational order. The basis for calculating orientational correlations is the director field of the magnetic domains. Despite the meandering evidenced by bands in the director field, the stripe-domain phases have long-range orientational order. The positional order is short- to (quasi-)long-ranged. In the labyrinthine phase, both orientational and positional orders are short-ranged. The director field of the labyrinthine phase is composed of patches within which the director field is uniform. The patches are separated by phase and amplitude grain boundaries.

In films with a thickness where $T_c$ is on the order of room temperature or above, the high-temperature stripes become mobile at a temperature just below $T_c$. This dynamic transition is reminiscent of a glass transition. A glass transition has indeed been predicted to occur in Coulomb-frustrated ferromagnets within a narrow temperature range around $T_c$. In films where $T_c$ is below room temperature, a highly disordered though stationary domain pattern separates the high-temperature stripes from the paramagnetic phase. In this phase, both orientational and positional orders are short-ranged. The connection of this phase with the mobile phase or the paramagnetic phase remains to be explored.

**Part II**

The tunability of the domain width in ultrathin Fe films on Cu(001) is used to investigate the transition from a multi-domain to a single-domain state in laterally confined structures. It is confirmed that the single-domain state occurs as soon as the lateral dimensions of a structure are of the order of the domain width.

**Part III**

Thin Fe films on W(110) have a strong uniaxial anisotropy in the plane of the film. The direction of the anisotropy rotates by $90^\circ$ at a thickness of around 80 Å. The high spatial resolution of SEMP A is used to image the transition region. It is found that the transition region on a wedge is characterized by asymmetric zig-zag domain walls. Across these walls, the magnetization rotates by $90^\circ$. In thicker films, symmetric zig-zag walls are found. There the magnetization rotates by $180^\circ$ across a wall. Both wall types are magnetically charged. In the case of $180^\circ$ walls, the walls are wide enough for their internal structure to be resolved in great detail. The distribution of the magnetic charge across these walls is determined.
Part I

Magnetic Domains in Ultrathin Iron Films on Cu(100)
Chapter 1

Introduction

Ultrathin Fe films on Cu(001) have long been the subject of extensive studies, both experimental and theoretical. Structural instabilities give rise to a highly complex behavior of the magnetization in this, at first sight, simple epitaxial system. The focus in most previous studies has been directed at the intricate interplay between the film structure and the magnetic properties of the individual layers of the films. Here, a Scanning Electron Microscope with Polarization Analysis (SEMPA) is used to shed light on the magnetic domains and on how they evolve as temperature or film thickness is varied. Due to the high spatial resolution achievable with this technique, detailed images of the domain patterns can be obtained.

In the limit of ultrathin films, a strong uniaxial anisotropy forces the magnetization in these films to be oriented along a direction perpendicular to the film plane. The ground state of such a system has been predicted to consist of an array of parallel stripes with alternating orientations of the magnetization [1–4]. Experimentally, this stripe-domain pattern has been confirmed by Allenspach and Bischof [5]. The formation of stripe domains is the result of competing interactions. The short-ranged exchange interaction favors a parallel alignment of neighboring spins. At long distances, however, the strong but short-ranged exchange interaction is counteracted by the weak but long-ranged dipolar interaction, which favors an antiparallel alignment of the spins. Under the influence of this frustration, the magnetization of a film with a strong perpendicular anisotropy breaks up into domains with opposite magnetizations, the stripe domains.

Such modulated phases due to competing interactions are ubiquitous in nature, both in equilibrium and nonequilibrium systems [6–8]. They have been found in systems as diverse as Langmuir monolayers, Rayleigh-Bénard convection cells, chemical reaction-diffusion systems, ferrofluids, liquid crystals, the optical cortex of primates, or show up in the skins of animals. Stripe order has also been found in high-temperature superconductors [9]. Striking similarities of these various modulated phases suggest a common frame-
work for their description [6]. Thus, insight gained in one of these systems may be relevant for other modulated phases. Indeed, it can be shown that the elementary description of perpendicularly magnetized films in terms of exchange and dipolar interactions can be transformed into a continuum model based on elastic constants [10, 11]. This continuum model is also used to describe smectics, and an analogy has been established between smectics and superconductors [12].

The order of two-dimensional systems is eventually lost at sufficiently high temperatures. A large number of models have been put forward to explain the melting of such two-dimensional systems [13]. While some highlight the role of topological defects and predict the occurrence of intermediate states and second-order transitions, other models suggest a direct first-order transition to the disordered high-temperature state. However, experimental evidence is still rare.

Due to its high spatial resolution and surface sensitivity, the Scanning Electron Microscope with Polarization Analysis represents an ideal tool to investigate the disordering of the perpendicularly magnetized domains in ultrathin Fe films on Cu(001) on a microscopic scale. Indeed, it is found that the stripe domains are only one of a number of different domain configurations that occur as temperature or film thickness is varied. On the way to the disordered paramagnetic phase, the system undergoes a series of transitions through intermediate states with different symmetries. These transitions are found to be mediated by topological defects. However, the mechanisms are not those predicted theoretically. Quite surprisingly, a reentrant high-temperature stripe phase is observed just below the transition to the disordered state [14]. It evolves out of a labyrinthine phase with higher symmetry. Such cases where the symmetry of the system is reduced as the temperature is increased are rare. The reduced symmetry of the reentrant phase has to be balanced by some disorder in other aspects so that the entropy does not decrease with temperature [15].

A possible ingredient mostly neglected in theoretical models is the domain width. A model reproducing the experimental results near the transition temperature is presented. However, an explanation for the reentrance phenomenon is still missing.

Throughout most of the phase diagram, the magnetic domains form stationary patterns, but close to the transition to the paramagnetic state the high-temperature stripe domains become highly mobile. This is true for a range of thicknesses where the Curie temperature is on the order of room temperature or above. The dynamic transition of the high-temperature stripes is reminiscent of a glass transition. A glass transition has indeed been predicted to occur in a Coulomb-frustrated ferromagnet [16–18]. The glass transition is thereby expected to take place at a temperature in close vicinity of the Curie temperature, as is the case for the dynamic transition in the system of ultrathin Fe films on Cu(001).

For thicknesses where the Curie temperature is below room temperature, the high-temperature stripe phase is separated from the paramagnetic state by a highly disordered but stationary domain pattern. A similar domain pattern has already been
observed in a previous study of this system [19]. It is a puzzle why this liquid state is only found in films where the Curie temperature is low. Whether this phase represents some kind of a frozen paramagnetic state remains speculative and further experiments are required to determine the nature of this disordered state.

In order to characterize the various phases that occur in ultrathin Fe films, both positional and orientational correlations are calculated from the images. These experimentally determined correlations reveal the type of order present in the individual phases and are compared to model calculations. The possible types of topological defects in a lamellar two-dimensional system and their combination laws are determined using concepts of homotopy theory. Various mechanisms that mediate transitions between different phases can be described as mergers or dissociations of such topological defects.

The system of ultrathin Fe films on Cu(001) is characterized by a number of structural instabilities due to the lattice mismatch between the Fe film and the copper substrate. The film structure is sensitive to film thickness, temperature, and preparation conditions. It has an impact on the magnetic properties of a film, including the magnetic domain patterns. Great care has thus to be taken to control the preparation conditions. Structural differences are monitored by means of electron diffraction. A more comprehensive characterization can be expected by means of Scanning Tunneling Microscopy, which images a film on an atomic level and in direct space. Such an investigation is especially desirable in order to understand the nonnegligible influence of substrate corrugation.

Note that the whole experiment is carried out without applying magnetic fields. The influence of magnetic fields on stripe domains has been investigated in previous experiments, although in slightly thicker films [20–22].
Starting from the elementary exchange and dipolar interactions, the theory of magnetic domain patterns in perpendicularly magnetized ultrathin films is developed step by step. First, the mean-field free energy is calculated. This free energy itself already reveals some interesting aspects of the domain pattern, notably the temperature dependence of the domain width. An analysis using Fourier transforms of both energy and entropy investigates the free energy as a function of symmetry. In a second step, a continuum model is developed out of the mean-field free energy. It expresses the free energy in terms of elastic constants and deviations of the domain pattern from its equilibrium configuration and is used to tackle such issues as the energy of topological defects or the type of positional and orientational order present in the domain pattern. The topological defects are classified by means of group-theoretical methods. Finally, previous work concerning the topological disordering of two-dimensional ordered media is reviewed.

While failing to deliver a description of the phase diagram of the domain pattern in ultrathin perpendicularly magnetized films, the theory presented in this chapter provides a framework for describing the individual magnetic phases and produces important ingredients for understanding the mechanisms mediating the transitions between the individual phases.
2.1 Relevant Energies

The magnetic configuration of a ferromagnetic body is governed by the interplay of various interactions. The relevant energies are the exchange energy $E_{\text{exch}}$, the magnetostatic or dipolar energy $E_d$, and the anisotropy energy $E_a$. In the presence of a magnetic field, the energy of the magnetized body in the field has to be included in the considerations. As the experiments discussed in this thesis have been performed without the application of a magnetic field, the corresponding energy will, however, be neglected in the further discussion.

2.1.1 Exchange Energy

The exchange interaction is a consequence of the antisymmetrization of the wave function due to Pauli’s exclusion principle. It arises whenever there is an overlap of the wave functions of neighboring electrons. As the overlap decreases quickly with increasing distance, the exchange interaction is short-ranged and can be written as

$$E_{\text{exch}} = -\frac{1}{2} \sum_{\langle i,j \rangle} J \vec{m}_i \cdot \vec{m}_j,$$

(2.1)

where the sum is over neighboring sites $i$ and $j$. The dimensionless quantities $\vec{m}_i$ are proportional to the spin assigned to the lattice site $i$, $J$ is the exchange energy between neighboring spins. In the case of ferromagnetism, $J > 0$, which favors a uniform parallel alignment of the spins. The exchange energy is independent of the direction along which the spins are aligned.

2.1.2 Dipolar Energy

The magnetostatic or dipolar energy of a magnetized body is due to the energy of its individual magnetic moments in the magnetic field created by the other magnetic moments. It is given by

$$E_d = -\frac{1}{2} \mu_0 \iiint \vec{M} \cdot \vec{H}_d \ d^3r,$$

(2.2)

where $\vec{M}$ is the magnetization and $\vec{H}_d$ the so-called demagnetizing field. The demagnetizing field $\vec{H}_d$ is the magnetic field created by the magnetization $\vec{M}$ and can

\footnote{For other formulations of the dipolar energy of a magnetization distribution $\vec{M}(\vec{r})$, see [23].}
be written as the negative gradient of a magnetic scalar potential $\Phi$. The sources of the magnetic scalar potential are the magnetic charges $\rho_m = - \text{div} \vec{M}$ [24]:

$$\Phi(\vec{r}) = \frac{1}{4\pi} \iiint \frac{\rho_m(\vec{r}')}{|\vec{r} - \vec{r}'|} \, d^3r' ,$$

(2.3)

where the integral extends over all space.

When the magnetic charges are restricted to the surface of the magnetic body, the magnetic scalar potential can also be expressed as a surface integral:

$$\Phi(\vec{r}) = \frac{1}{4\pi} \int_S \frac{\sigma(\vec{r}')}{|\vec{r} - \vec{r}'|} \, dS ,$$

(2.4)

$\sigma = \vec{M} \cdot \vec{n}$ being the magnetic surface charge density, and $\vec{n}$ being a unit vector perpendicular to the surface.

For a magnetization $\vec{M}$ of a given magnitude, the dipolar energy is dependent on the relative orientation of the magnetization axis and the body. Provided the magnetization is uniform throughout the body, the demagnetizing field $\vec{H}_d$ can be written as

$$\vec{H}_d = -N \cdot \vec{M} ,$$

(2.5)

where $N$ is the so-called demagnetizing tensor. In the case of an extended film with vanishing thickness, the demagnetizing fields associated with a uniform magnetization parallel or perpendicular to the film plane are given by

$$\vec{H}_{d,||} = 0 \quad \text{and} \quad \vec{H}_{d,\perp} = -\vec{M} .$$

(2.6)

The dipolar energy thus favors a magnetization parallel to the film plane.\(^2\)

### 2.1.3 Bulk Anisotropy Energy

In bulk crystalline materials, the energy of a uniform magnetization is sensitive to the relative orientation of the magnetization vector and the crystal lattice. Van Vleck pointed out that magnetic anisotropy is due to spin-orbit coupling [26]. The density of the anisotropy energy can be written as an expansion in the magnetization components along different axes. In a cubic crystal it reads:

$$\frac{E_a}{V} = K_{c1} \left\{ M_x^2 M_y^2 + M_x^2 M_z^2 + M_y^2 M_z^2 \right\} + K_{c2} M_x^2 M_y^2 M_z^2 ,$$

(2.7)

where the term proportional to the constant $K_{c2}$ and higher order terms are small compared to the term proportional to $K_{c1}$. The sign of the constant $K_{c1}$ basically determines whether the $\langle 100 \rangle$ or the $\langle 111 \rangle$ directions are the easy directions for the magnetization.\(^3\) For iron, $K_{c1}$ is positive and the easy magnetization direction is

---

\(^2\)A general method for computing the demagnetizing tensor of uniformly magnetized bodies is given in [25].

\(^3\)For more detailed information, see [27].
This bulk anisotropy contribution neither favors in-plane, nor out-of-plane magnetization.

### 2.1.4 Surface Anisotropy Energy

In the system of ultrathin Fe films on Cu(100), the magnetization is perpendicular to the film plane \([28]\). This has to be attributed to the surface anisotropy energy. Néel \([29, 30]\) has shown that due to the broken symmetry at the surfaces of a film an additional surface anisotropy term has to be considered. If the anisotropy is reduced to pairwise interactions, it becomes clear that contributions which cancel for a site in the bulk are no longer compensated at the surface. The energy density of the surface anisotropy can be written as:

\[
\frac{E_{s,a}}{A} = K_s \cos^2(\theta),
\]

where \(\theta\) is the angle between the magnetization and the surface normal. If \(K_s\) is negative, a perpendicular orientation of the magnetization is favored. As the surface anisotropy energy is only proportional to the surface area of the film, the demagnetizing energy will dominate in thicker films and force the magnetization to lie in the film plane.

A first calculation of the surface anisotropy of a monolayer was carried out by Gay and Richter \([31]\). In a first step, the spin-polarized electronic structure is calculated by the self-consistent local-orbital method. The associated energy is independent of the magnetization direction. In a second step, spin-orbit coupling is introduced as a small perturbation. Gay and Richter find a surface anisotropy of about \(-0.4\) meV/atom, which is 100 times as large as the anisotropy of bulk Fe (4 \(\mu\)eV/atom).

In more recent calculations, Lorenz and Hafner \([32]\) considered Fe films of various thicknesses on Cu(100). In self-consistent real-space recursion calculations using a tight-binding-linear-muffin-tin-orbital Hubbard Hamiltonian, the energy associated with a certain direction of the magnetization can be determined. As the calculations are carried out in real space, problems occurring in calculations in the reciprocal space can be avoided, e.g. metastable states can be identified as such. Their method allows the determination of the magnetization for each individual layer in the film, which is especially interesting for thicker films. In films with a thickness of up to three atomic layers of interest here, the magnetization direction across the film is uniform with enhanced magnetic moments at the surface due to the lower coordination number. The anisotropy energies are of the order of \(-(1–2)\) meV.\(^4\)

\(^4\) References for further calculations can be found in \([32]\).
2.2 Ground State of Perpendicularly Magnetized Films

As ultrathin Fe films on Cu(100) are magnetized along an axis perpendicular to the film plane, a uniform magnetization throughout the film would create a large dipolar energy, Eq. (2.6). This energy is reduced when the films break up into domains of opposite magnetization. The insertion of oppositely magnetized domains, however, increases the domain wall length and thus the domain wall energy. At a domain wall, neighboring spins are no longer parallel, which increases the exchange energy.

As will be shown in Subsec. 2.3.3, the domain width $L$ is determined by a minimization of the free energy and proportional to the ratio of exchange and dipolar energy. The case of relatively thick films where the thickness $D$ clearly exceeds the domain width $L$ was discussed by Kittel [33, 34]. Later, films where $D$ and $L$ are of the same order were analyzed by Málek and Kamberský [35]. The case relevant for ultrathin Fe films on Cu(100) where the domain width $L$ exceeds the domain thickness $D$ by a factor of several thousands was first treated by Yafet and Gyorgy [1]. They found that ultrathin films with perpendicular anisotropy would break up into domains when the perpendicular surface anisotropy was larger than a critical value. According to their results, the domain configuration with the lowest energy is given by stripe domains. Considering a monolayer with strong Ising anisotropy, Czech and Villain [37] argued that a checkerboard domain pattern would have a lower energy. This controversy was settled by Kaplan and Gehring [2] in favor of the stripe pattern. The ground state was also calculated by Kashuba and Pokrovsky [3] and MacIsaac et al. [4]. Their analyses yield the same conclusion.

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5 The domain wall width is governed by the ratio of exchange and bulk anisotropy energy.
6 As an isotropic two-dimensional system has no long-range order [36], some kind of anisotropy is necessary for magnetic ordering.
2.3 Mean-Field Theory

In this section, a simple mean-field model including both the short-range exchange interaction and the long-range dipolar interaction is explored. The considerations are based on a free energy according to Villain [37, 38]. This free energy delivers the well-known mean-field dependence of the magnetization on temperature near the Curie temperature $T_c$. A Fourier transform of the energy will then be used to calculate the stripe width at $T_c$. Although the temperature dependence of the stripe width cannot be calculated over the whole temperature range, the model permits a prediction of the temperature dependence in the vicinity of $T_c$. The experimental verification in Subsec. 3.7.2 will show that the formula obtained describes the temperature dependence adequately. As a by-product, we will obtain the variation of the magnetization profile near $T_c$. Finally, using a Fourier transform of the entropy, the influence of the domain pattern symmetry on the free energy of the system will be evaluated. The free energies of systems with two-fold, four-fold, and six-fold symmetry are compared.

The mean-field theory developed in this section will form the basis for a continuum model developed in Sec. 2.4. This continuum model will then be used to calculate the energies of topological defects in stripe domains in Subsec. 2.5.3. It will also be used to determine the type of positional and orientational order present in the system, cf. Sec. 2.6.

2.3.1 Free Energy

We consider a two-dimensional square lattice of spins. Although, at a given time, every site of the lattice will be occupied by a spin with a definite value, which in this model can either be 1 or $-1$, every site will also be characterized by the average value of the spin at that site.

In order to determine the entropy of the system, we follow the ideas of Bragg and Williams [39] by establishing an analogy with alloys. We divide the system into areas of equal average spin, which in an alloy would correspond to areas occupied by one type of atoms at low temperatures. These areas are not restricted in their geometry and not necessarily connected. We assume the system to be large enough so that each such area will still contain a large number of spins.

Let $N_i$ be the number of spins in area $i$ with average spin $\tilde{m}_i$. The number of spins 1 in area $i$ will be given by $\frac{N_i}{2}(1 + \tilde{m}_i)$, the number of opposite spins will be $\frac{N_i}{2}(1 - \tilde{m}_i)$. The number of configurations consistent with an average spin of $\tilde{m}_i$ will thus be given by

$$\frac{N_i!}{\left(\frac{N_i}{2}(1 + \tilde{m}_i)\right)! \left(\frac{N_i}{2}(1 - \tilde{m}_i)\right)!}.$$ (2.9)
The total number of configurations of the system $W$ is obtained by multiplying this factor for every area $i$. The entropy is proportional to the logarithm of $W$:

\[
\ln(W) = \ln \left( \prod_{i=1}^{n} \frac{N_i!}{\left(\frac{N_i}{2}(1 + \tilde{m}_i)\right)! \left(\frac{N_i}{2}(1 - \tilde{m}_i)\right)!} \right) \\
= \sum_{i=1}^{n} \ln \left( \frac{N_i!}{\left(\frac{N_i}{2}(1 + \tilde{m}_i)\right)! \left(\frac{N_i}{2}(1 - \tilde{m}_i)\right)!} \right) \\
\approx \sum_{i=1}^{n} \left\{ N_i \ln(N_i) - \left(\frac{N_i}{2}(1 + \tilde{m}_i)\right) \ln \left(\frac{N_i}{2}(1 + \tilde{m}_i)\right) \\
- \left(\frac{N_i}{2}(1 - \tilde{m}_i)\right) \ln \left(\frac{N_i}{2}(1 - \tilde{m}_i)\right) \right\} \\
= \sum_{i=1}^{n} \left\{ N_i \ln(2) - \frac{N_i}{2} \left[ (1 + \tilde{m}_i) \ln(1 + \tilde{m}_i) + (1 - \tilde{m}_i) \ln(1 - \tilde{m}_i) \right] \right\} \\
= \sum_{i=1}^{N} \left\{ \ln(2) - \frac{1}{2} \left[ (1 + m_i) \ln(1 + m_i) + (1 - m_i) \ln(1 - m_i) \right] \right\}. \tag{2.10}
\]

Here, $n$ is the number of areas the system consists of, and $N$ is the total number of lattice sites present in the system. Note that the final summation is over the lattice sites and not over the areas which no longer appear in the formula. The partition of the system into areas of equal average spin will not be needed any more in the forthcoming calculations.

Using Eq. (2.10), the free energy of the systems reads:

\[
F = -\frac{1}{2} \sum_{i,j} J_{ij} m_i m_j + \frac{1}{2} k_B T \sum_i \left[ (1 + m_i) \ln(1 + m_i) + (1 - m_i) \ln(1 - m_i) \right], \tag{2.11}
\]

where

\[
J_{ij} = \begin{cases} 
J - g \left( \frac{a}{r_{ij}} \right)^3 & \text{if } i \text{ and } j \text{ are neighbours,} \\
- g \left( \frac{a}{r_{ij}} \right)^3 & \text{otherwise.} \end{cases} \tag{2.12}\]

The energy of the exchange interaction between parallel neighboring spins is given by $J$ whereas $g$ denotes the energy of the dipolar interaction, $r_{ij}$ is the distance between sites $i$ and $j$, $a$ the lattice parameter. The constant contribution of the term proportional to $\ln(2)$ in Eq. (2.10) to the free energy will be neglected.
2.3.2 Magnetization as a Function of Temperature

In order to determine the temperature dependence of the magnetization near $T_c$, we assume a homogeneously magnetized system where the magnetization is given by $m$. Neglecting the dipolar interaction, we can write the free energy as:

$$ F(m) = -\frac{1}{2}NzJm^2 + \frac{1}{2}k_B T N [(1 + m) \ln(1 + m) + (1 - m) \ln(1 - m)], \quad (2.14) $$

where $z$ denotes the number of nearest neighbors. The value of the magnetization assumed at a certain temperature $T$ is obtained by minimizing the free energy with respect to $m$. This yields an implicit equation for $m$:

$$ \frac{1}{N} \frac{dF}{dm} = -\frac{zJ}{k_B T} m + \frac{1}{2}k_B T \ln \left( \frac{1 + m}{1 - m} \right) = 0 \Rightarrow m = \tanh \left( \frac{zJ}{k_B T} m \right). \quad (2.15) $$

If $\frac{zJ}{k_B T} < 1$, the only solution for $m$ is $m = 0$. For $\frac{zJ}{k_B T} > 1$, however, two non-vanishing solutions for $m$ exist. The Curie temperature $T_c$ is thus given by

$$ T_c = \frac{zJ}{k_B}. \quad (2.16) $$

Near $T_c$, where $m$ is small, we can expand $\tanh x$ as $\tanh x \approx x - \frac{1}{3}x^3$ and get

$$ m = \sqrt{3} \left( \frac{T_c - T}{T_c} \right)^{\frac{1}{2}}. \quad (2.17) $$

This is the well-known mean-field behavior with the critical exponent $\beta = \frac{1}{2}$.

2.3.3 Stripe Width as a Function of Temperature

The stripe width $L$ as a function of temperature is obtained by Fourier transforming the energy term in the free energy, Eq. (2.11), and minimizing it with respect to the wave vector $\vec{k}$. For this purpose, we introduce the Fourier transforms of $m_i$ and $J_{ij}$. For the Fourier transform of $m_i$, we use the convention:

$$ m_j = \sum_k m_k e^{i\vec{k}\vec{r}_j}, \quad m_k = \frac{1}{N} \sum_j m_j e^{-i\vec{k}\vec{r}_j}. \quad (2.18) $$

In this convention, $m_{\vec{k}}$ is an intensive quantity proportional to the magnetization of the system. For the Fourier transform of $J_{ij}$, a different scheme is adopted:

$$ J_{ij} = \frac{1}{N} \sum_{\vec{k}} J_{\vec{k}} e^{i\vec{k}(\vec{r}_j - \vec{r}_i)}, \quad J_{\vec{k}} = \sum_j J_{ij} e^{-i\vec{k}(\vec{r}_i - \vec{r}_j)}. \quad (2.19) $$
Although the normalization is different, $J_{\vec{k}}$ is also virtually independent of system size. We now express the energy term in Eq. (2.11) in terms of the Fourier transforms $m_{\vec{k}}$ and $J_{\vec{k}}$:

\[
\sum_{i,j} J_{ij} m_i m_j = \sum_{i,j} \left( \sum_{\vec{k}} m_{\vec{k}} e^{i\vec{k}\cdot\vec{r}_{ij}} \right) \left( \sum_{\vec{k}'} m_{\vec{k}'} e^{i\vec{k}'\cdot\vec{r}'_j} \right) \left( \frac{1}{N} \sum_{\vec{k}''} J_{\vec{k}''} e^{i\vec{k}''(\vec{r}_j - \vec{r}_i)} \right)
\]

\[
= \sum_{\vec{k},\vec{k}',\vec{k}''} m_{\vec{k}} m_{\vec{k}'} J_{\vec{k}''} \cdot \sum_i e^{i(\vec{k}'' - \vec{k}'')\vec{r}_i} \cdot \left\{ \frac{1}{N} \sum_j e^{i(\vec{k}'' + \vec{k}'')\vec{r}_j} \right\}
\]

\[
= \sum_{\vec{k},\vec{k}',\vec{k}''} m_{\vec{k}} m_{\vec{k}'} J_{\vec{k}''} \cdot \sum_i e^{i(\vec{k}'' - \vec{k}'')\vec{r}_i} \cdot \delta(\vec{k}' + \vec{k}'')
\]

\[
= N \sum_{\vec{k},\vec{k}'} m_{\vec{k}} m_{\vec{k}'} J_{\vec{k}'} \cdot \left\{ \frac{1}{N} \sum_i e^{i(\vec{k} - \vec{k}')\vec{r}_i} \right\}
\]

\[
= N \sum_{\vec{k},\vec{k}'} m_{\vec{k}} m_{\vec{k}'} J_{\vec{k} - \vec{k}'} \cdot \delta(\vec{k} - \vec{k}') = N \sum_{\vec{k}} m_{\vec{k}} m_{-\vec{k}} J_{\vec{k}} . \quad (2.20)
\]

The Fourier transform $J_{\vec{k}}$ can be evaluated in a series of steps. We thereby follow the line of Czech and Villain [37]. First, we note that:

\[
J_{\vec{k}} = \sum_j J_{ij} e^{-i\vec{k}\cdot\vec{r}_{ij}} = \sum_j J_{ij} \cos(\vec{k}\cdot\vec{r}_{ij})
\]

\[
= 2J \{ \cos(k_x a) + \cos(k_y a) \} - g \sum_j \cos(\vec{k}\cdot\vec{r}_{ij}) \left( \frac{a}{r_{ij}} \right)^3 . \quad (2.21)
\]

We now concentrate on the sum in Eq. (2.21). As a typical domain width will comprise many lattice constants, the relevant values of $\vec{k}$ will be small, and therefore, the sum will be nearly isotropic. For convenience, we choose $\vec{k}$ to be parallel to a side of the square unit cell, e.g. the $x$-axis, and measure distances in units of the lattice parameter $a$. Then we have:

\[
S(\vec{k}) = \sum_{\vec{r} \neq 0} \cos(\vec{k}\cdot\vec{r}) \frac{1}{r_{ij}^3} = 2 \sum_{\vec{x}=1}^{\infty} \cos(ka\vec{x}) R(\vec{x}) + 2 \sum_{\vec{y}=1}^{\infty} \frac{1}{y^3} , \quad (2.22)
\]

where $R(\vec{x})$ is the sum over an atomic row at distance $\vec{x}$ perpendicular to the direction of $\vec{k}$. If $\vec{x}$ is not too small, this sum can be replaced by an integral:

\[
R(\vec{x}) = \sum_{\vec{y}=\infty}^{\infty} \frac{1}{(\vec{x}^2 + \vec{y}^2)^{3/2}} \approx \int_{-\infty}^{\infty} \frac{d\vec{y}}{(\vec{x}^2 + \vec{y}^2)^{3/2}} = \frac{2}{\vec{x}^2} . \quad (2.23)
\]
This approximation has been discussed by Yafet and Gyorgy [1]. The corresponding error is only of the order of 1% for the smallest possible $\bar{x} = 1$. The sum over $\bar{x}$ in Eq. (2.22) can also be replaced by an integral provided an appropriate lower integration boundary $\bar{x}_0$ is introduced:

$$
\sum_{\bar{x}=1}^{\infty} \cos(ka\bar{x}) \frac{1}{\bar{x}^2} \to \int_{\bar{x}_0}^{\infty} \cos(ka\bar{x}_0) \frac{d\bar{x}}{\bar{x}^2} = \frac{\cos(ka\bar{x}_0)}{\bar{x}_0} + ka \text{Si}(ka\bar{x}_0) - ka \frac{\pi}{2} .
$$

(2.24)

Since we are interested in small values of $k$, it is reasonable to choose the boundary $\bar{x}_0 = 6/\pi^2$ which yields the correct result for $k = 0$ [1]. With $c = \sum_{j=1}^{\infty} \frac{1}{j^3} \approx 1.202$, we finally obtain the following expression for the Fourier transform $J_\vec{k}$:

$$
J_\vec{k} = 2J\{\cos(kx) + \cos(ky)\} - 4g\left\{\frac{\cos(ka\bar{x}_0)}{\bar{x}_0} + ka \text{Si}(ka\bar{x}_0)\right\} + 2\pi gka - 2gc
\approx 4J - 2g\left\{\frac{2}{\bar{x}_0} + c\right\} + 2\pi gka - \{J + 2g\bar{x}_0\}k^2a^2 .
$$

(2.25)

In order to determine the stripe width at temperatures lower than $T_c$, we approximate the magnetization profile by a series of cosines and evaluate the free energy in this approximation. The stripe width is then given by the value of $k$ that leads to a minimum of the free energy. Quite generally, a one-dimensional modulation with a periodicity of $2L(T) = \frac{2\pi}{k_0(T)}$ can be described by

$$
m_i = \sum_{n=0}^{\infty} a_n(k_0, T) \cos(nk_0x_i) .
$$

(2.26)

If we require that the magnetization at locations separated by $L$ solely differs by a factor of $-1$, we can restrict ourselves to terms with $n$ odd. The expansion then reads

$$
m_i = \sum_{n=0}^{\infty} a_n(k_0, T) \cos((2n + 1)k_0x_i) .
$$

(2.27)

To simplify the further calculations, we limit the expansion to the third harmonic:

$$
m_i = A \cos(k_0x_i) + B \cos(3k_0x_i) .
$$

(2.28)

Substituting Eq. (2.28) into Eq. (2.20), the free energy, Eq. (2.11), reads:

$$
\frac{F}{N} = -\frac{1}{4} [A^2J_{k_0} + B^2J_{3k_0}]
+ \frac{k_BT}{2\pi} \int_{0}^{\pi} [(1 + m(\varphi)) \ln(1 + m(\varphi)) + (1 - m(\varphi)) \ln(1 - m(\varphi))] d\varphi .
$$

(2.29)

7The simple expression $T_c = \frac{4J}{k_B}$ is but the first approximation. How the Curie temperature is affected by the dipolar interaction is discussed in Subsec. 2.3.6.
where
\[ m(\varphi) = A \cos(\varphi) + B \cos(3\varphi) \]  \hspace{1cm} (2.30)

The entropy does not depend on \( k_0 \) whence the minimization of the free energy with respect to \( k_0 \) is given by
\[- \frac{4}{N} \frac{\partial F}{\partial k_0}(k_0) = A^2 \frac{\partial J_k}{\partial k}(k_0) + B^2 \frac{\partial J_k}{\partial k}(3k_0) \equiv 0 \]  \hspace{1cm} (2.31)

At this point, the temperature dependence of the coefficients \( A \) and \( B \) has to be taken into account. Near \( T_c \), the coefficients can be expanded in a series as follows [38]:
\[ a_n(k_0, T) = \sum_{p=n}^{\infty} \alpha_n^p(k_0) \left( \frac{T_c - T}{T_c} \right)^{\frac{2p+1}{2}} \]  \hspace{1cm} (2.32)

If only the first-order terms are retained, the temperature dependence of the coefficients \( A \) and \( B \) is given by:
\[ A = \alpha_0^0 \left( \frac{T_c - T}{T_c} \right)^{\frac{1}{2}}, \quad B = \alpha_1^1 \left( \frac{T_c - T}{T_c} \right)^{\frac{3}{2}} \]  \hspace{1cm} (2.33)

In this approximation, we consider the coefficients to be independent of \( k_0 \). Substituting Eq. (2.33) into Eq. (2.31), and solving Eq. (2.31) for \( k_0 \), we have the following expression for the stripe width at a temperature \( T \) lower than \( T_c \):
\[ L_0(T) = \frac{\pi}{k_0(T)} \approx \frac{J + 2g\tilde{r}_0}{g} \left\{ 1 + 2 \left( \frac{\alpha_1^1}{\alpha_0^0} \right)^2 \left( \frac{T_c - T}{T_c} \right)^2 \right\} a \]  \hspace{1cm} (2.34)

As the expansion of \( m_i \) has been limited to the third harmonic, this expression only holds for temperatures near \( T_c \).

### 2.3.4 Magnetization Profile as a Function of Temperature

Knowing both the temperature dependence of the coefficients \( a \) and \( b \) in the expansion of \( m_i \) and the temperature dependence of the stripe width \( L_0 \), the magnetization profile can be written as:
\[ m_i(T) = \alpha_0^0 \left( \frac{T_c - T}{T_c} \right)^{\frac{1}{2}} \cos(k_0(T)x_i) + \alpha_1^1 \left( \frac{T_c - T}{T_c} \right)^{\frac{3}{2}} \cos(3k_0(T)x_i) \]  \hspace{1cm} (2.35)

Very close to \( T_c \), the profile is described by a cosine. For slightly lower temperatures, however, the profile soon assumes a more rectangular form.
2.3.5 Free Energy as a Function of Symmetry

In Sec. 2.2, it was shown that the ground state of perpendicularly magnetized ultrathin films is characterized by stripe domains. The assumption of stripe domains was the basis for investigating the domain width as a function of temperature in Subsec. 2.3.3. However, domain configurations with a different geometry are conceivable and indeed found in experiments as will be shown in Chapter 3.

The subject of this subsection is to calculate the free energy as a function of symmetry within the model introduced in Subsec. 2.3.1. For this purpose, we expand the logarithms in the free energy of Eq. (2.11):

\[
(1 + m_i) \ln(1 + m_i) + (1 - m_i) \ln(1 - m_i) = \sum_{n=1}^{\infty} \alpha_n m_i^{2n},
\]

(2.36)

where

\[
\alpha_n = \frac{1}{n(2n - 1)}.
\]

(2.37)

Thus we have:

\[
\sum_i [(1 + m_i) \ln(1 + m_i) + (1 - m_i) \ln(1 - m_i)] \approx \sum_i m_i^2 + \frac{1}{6} \sum_i m_i^4 + \frac{1}{15} \sum_i m_i^6.
\]

(2.38)

We can now express the first sum in Eq. (2.38) using the Fourier transform \( m_{\vec{k}} \):

\[
\sum_i m_i^2 = \sum_i \left\{ \sum_{\vec{k}, \vec{k}'} e^{i \vec{k} \cdot \vec{r}_i} m_{\vec{k}} \cdot e^{i \vec{k} \cdot \vec{r}_i} m_{\vec{k}'} \right\}
\]

\[
= N \sum_{\vec{k}, \vec{k}'} m_{\vec{k}} m_{\vec{k}'} \left\{ \frac{1}{N} \sum_i e^{i(\vec{k} + \vec{k}' \cdot \vec{r}_i) \cdot \vec{r}_i} \right\}
\]

\[
= N \sum_{\vec{k}, \vec{k}'} m_{\vec{k}} m_{\vec{k}'} \delta(\vec{k} + \vec{k}') = N \sum_{\vec{k}} m_{-\vec{k}} m_{\vec{k}}.
\]

(2.39)

The Fourier transforms of the other sums read:

\[
\sum_i m_i^4 = N \sum_{\vec{k}, \vec{k}', \vec{k}''} m_{\vec{k}} m_{\vec{k}'} m_{\vec{k}''} m_{-\vec{k} + \vec{k}''}.
\]

(2.40)

\[
\sum_i m_i^6 = N \sum_{\vec{k}, \vec{k}', \vec{k}'', \vec{k}''' \vec{k}'''} m_{\vec{k}} m_{\vec{k}'} m_{\vec{k}''} m_{\vec{k}'''} m_{\vec{k}''' - (\vec{k} + \vec{k}' + \vec{k}'' + \vec{k}''')}.
\]

(2.41)
Table 2.1: Symmetry-dependent wave-vector distributions occurring in the calculation of the sums in the expansion of the entropy. The wave vectors occupy sites on a ring with radius $k_0$. As the vectorial sum of the wave vectors has to vanish in each configuration, only a limited number of configurations are possible. The total number of wave vectors in each configuration is equal to the power of $m_i$ in the sums, and the number of possible sites is determined by the symmetry of the domain pattern. Occupied sites are marked with solid circles.
Regardless of symmetry, the domain configuration will be considered to be characterized by a typical domain width $L_0$. The Fourier transforms will thus have maxima on a circle with radius $k_0 = \frac{\pi}{L_0}$.

We will now distinguish the cases where the Fourier transform has $C_2$, $C_4$ or $C_6$ rotational symmetry. For simplicity, we assume that, in the case of $C_2$ symmetry, the Fourier transform is given by two $\delta$ peaks at the intersections of a line through the origin with the circle with radius $k_0$. The Fourier transforms in the cases of $C_4$ and $C_6$ symmetry are assumed to be given by four $\delta$ peaks on the corners of a square and six $\delta$ peaks on the corners of a regular hexagon respectively.

The sums on the right-hand side of Eqs. (2.39), (2.40) and (2.41) are now a mere function of symmetry and can be calculated using combinatorics. The distributions of wave vectors $\mathbf{k}$ that make a contribution to the sums are depicted in Tab. 2.1.

Each such distribution has to be weighted with the number of possible realizations. Thus we have for each symmetry $C_\alpha$

$$\sum_i m_i^j = NC_j^\alpha m_{k_0}^j$$

(2.42)

The coefficients $C_j^\alpha$ are calculated in Tab. 2.2 and allow to write the free energy in a simple form:

$$\frac{F^\alpha}{N} = -\frac{1}{2} J_{k_0} C_2^\alpha m_{k_0}^2 + \frac{1}{2} k_B T \left\{ C_2^\alpha m_{k_0}^2 + \frac{1}{6} C_4^\alpha m_{k_0}^4 + \frac{1}{15} C_6^\alpha m_{k_0}^6 \right\} .$$

(2.43)

It is a simple polynomial in $m_{k_0}$. The value of $m_{k_0}$ is obtained by minimizing the free energy:

$$\frac{1}{N} \frac{\partial F^\alpha}{\partial m_{k_0}} = -(J_{k_0} - k_B T) C_2^\alpha m_{k_0} + \frac{1}{3} k_B T C_4^\alpha m_{k_0}^3 + \frac{1}{5} k_B T C_6^\alpha m_{k_0}^5 = 0 .$$

(2.44)
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Figure 2.1: Temperature dependence of the magnetization as a function of symmetry. The Fourier transform $m_{k_0}$ of a system with $C_2$ symmetry (solid line) is highest at all temperatures. The Fourier transform $m_{k_0}$ in a system with $C_6$ symmetry (dotted line) is slightly smaller than in a system with $C_4$ symmetry (dashed line). The curves displayed were calculated using the approximation of Eq. (2.45).

Solving Eq. (2.44) for $m_{k_0}$ and retaining the lowest order, we obtain an expression for $m_{k_0}$ with a familiar temperature dependence, Eq. (2.17), and a magnitude which is dependent on symmetry:

$$m_{k_0} \approx \sqrt{\frac{3}{C_4}} \frac{C_0}{C_4} \sqrt{\frac{T_c - T}{T_c}}.$$

The temperature dependence of the magnetization as a function of symmetry is displayed in Fig. 2.1. Substituting $m_{k_0}$ into Eq. (2.43), we obtain the temperature dependence of the free energy. It is shown in Fig. 2.2. The system with $C_2$ symmetry has the lowest free energy over the whole temperature range up to the Curie temperature $T_c$. We would expect the system to be characterized by a $C_2$ symmetry for all temperatures lower than $T_c$. The differences between the free energies, however, become smaller and smaller as $T_c$ is approached. Apparently, the mean-field model considered here cannot predict a transition to a domain configuration with a higher symmetry.
2.3.6 Curie Temperature in the Presence of Dipolar Interaction

In a simple model where only nearest-neighbor exchange interactions are considered, the Curie temperature is given by $T_c = \frac{J}{k_B}$ with $z$ the number of nearest neighbors as discussed in Subsec. 2.3.2. In a model with the free energy of Eq. (2.11), however, the expression for $T_c$ is more complicated and includes terms due to the dipolar interaction.

Combining Eqs. (2.20), (2.38), and (2.39), we obtain the following expression for the free energy:

$$F = \frac{N}{2} \sum_k \left( k_B T - J_k \right) m_k m_{-k} + \frac{k_B T}{12} \sum_i m_i^4 + \cdots .$$  \hspace{1cm} (2.46)
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The minimum of the free energy corresponds to \( m_{k} = 0 \) for \( k_{B}T > J_{k_{\text{max}}} \), where \( k_{\text{max}} \) is the value of \( k \) for which \( J_{k} \) takes its maximum.\(^8\) The Curie temperature \( T_{c} \) is thus given by

\[
T_{c} = \frac{J_{k_{\text{max}}}}{k_{B}}. \tag{2.47}
\]

As \( J \gg g, J_{k} \), Eq. (2.25), is to first order given by \( zJ \) where \( z \) is the number of nearest neighbors. Since there are no third-order terms in Eq. (2.46) and the fourth-order terms have positive coefficients, this transition is continuous.

2.3.7 Modifications for Films Consisting of Several Layers

If films consisting of several atomic layers are to be considered, the thickness of the films has to be included in the calculations. The number of nearest neighbors in the bulk is higher than the number of nearest neighbors in a monolayer. In a cubic lattice, the average number of nearest neighbors as a function of film thickness \( D \) is given by

\[
z(D) = \frac{4D + 2(D - a)}{D} = \left\{ 6 - \frac{2a}{D} \right\} \quad \text{.} \tag{2.48}
\]

Due to the higher coordination number in thicker films, the Curie temperature is increased:

\[
T_{c}(D) = \frac{z(D)J}{k_{B}} = \frac{J}{k_{B}} \left\{ 6 - \frac{2a}{D} \right\} \quad \text{.} \tag{2.49}
\]

If the energy and entropy terms in Eq. (2.46) are taken to be proportional to the thickness \( D \), we obtain the following expression for the free energy

\[
F(D) = \left( \frac{D}{a} \right) \frac{N}{2} \sum_{k} \left\{ k_{B}T - J_{k}(D) \right\} m_{k} m_{-k} + \left( \frac{D}{a} \right) \frac{k_{B}T}{12} \sum_{i} m_{i}^{4} + \cdots. \tag{2.50}
\]

\(^8\) The maximum value of \( J_{k} \) is reached for

\[
k_{\text{max}} = \frac{\pi g}{a(J + 2g x_{0})}. \quad \text{ }
\]

Assuming a one-dimensional modulation of the magnetization, the stripe width at \( T_{c} \) is thus given by:

\[
L(T_{c}) = \frac{\pi}{k_{\text{max}}} = \frac{J + 2g x_{0}}{g} a \approx \frac{J}{g} a, \quad \text{which is the value obtained in Eq. (2.34).}
\]
Note that $J_{\vec{k}}$ is also a function of the thickness. We must pay attention to the thickness dependence of the coordination number, and Eq. (2.21) suggests that terms in $J_{\vec{k}}$ proportional to $g$ should vary linearly with the thickness. An analysis of the acoustic mode of the dispersion, however, shows that the terms that are independent of $k$ do not vary with thickness [40]. Those terms correspond to an effective easy-plane anisotropy independent of the film thickness. Thus $J_{\vec{k}}$ is given by

$$J_{\vec{k}}(D) \approx z(D)J - 2g \left\{ \frac{2}{\bar{x}_0} + c \right\} + \left( \frac{D}{a} \right) 2\pi gka - \{ J + 2g\bar{x}_0 \} k^2a^2 . \quad (2.51)$$

As a consequence of the different scaling of the various terms, the domain width $L$ will be a function of thickness:

$$L_0(T,D) = \frac{\pi}{k_0(T,D)} \approx \frac{J + 2g\bar{x}_0}{g} \left\{ 1 + 2 \left( \frac{\alpha_1}{\alpha_0} \right)^2 \left( \frac{T_c - T}{T_c} \right)^2 \right\} \frac{\alpha^2}{D} . \quad (2.52)$$

For thicker films, the domain width becomes smaller. This is consistent with previous results for the domain period as a function of thickness in the ultrathin limit [1, 41, 42].

### 2.4 Continuum Model

In Subsec. 2.3.5, an attempt was made to predict a change in symmetry by analyzing the free energy developed in Sec. 2.3. However, no hint to such a change has been found. In this section, we will elaborate a continuum model which expresses the free energy by a set of elastic constants describing the energy associated with bending and compression of domains. This model will be used in Subsec. 2.5.3 to calculate the energy of topological defects, and in Sec. 2.6 to discuss the type of positional and orientational order present in the domain pattern. The continuum model has served as a basis for calculations suggesting a multi-step disordering process as the Curie temperature is approached [43]. During this process, the two-fold symmetry of the stripe phase is predicted to be replaced with a four-fold symmetry in an intermediate state. A more comprehensive discussion of models for topological disordering is given in Sec. 2.7.

As in Sec. 2.3, a Fourier transform of the free energy density will be used to single out the wave vector that minimizes the free energy. Then the free energy associated with deviations from this optimal wave vector will be calculated. We will thereby follow the line of Garel and Doniach [10]. The deviations will be taken into account by means of a phase approximation which will lead us to a formulation of the free
energy in terms of elastic constants [11]. We will assume a free energy of the form

\[ F = F_{\text{exch}} + F_d \]

(2.53)

where \( F_{\text{exch}} \) denotes the exchange energy including entropy, and \( F_d \) denotes the dipolar energy.\(^9\) For \( F_{\text{exch}} \), we take a Landau-Ginzburg expansion in the magnetization \( m \):

\[ F_{\text{exch}} = \int \int \left\{ \frac{D}{2a} J(\nabla m)^2 + \frac{D}{2a^3} k_B (T - T_c) m^2 + \frac{D}{12a^3} k_B T_c m^4 \right\} d^2r \]

(2.54)

where the integral extends over the film area. \( D \) is the thickness of the film, the lattice parameter of which is represented by \( a \). The Curie temperature is here again given by

\[ T_c = \frac{zJ}{k_B} \]

(2.55)

where \( z \) is the coordination number. For the ensuing calculations, we adopt the following definition of the Fourier transform:

\[ m(\vec{r}) = \frac{1}{2\pi} \int \int m(\vec{q}) e^{i\vec{q} \cdot \vec{r}} d^2q \quad \text{and} \quad m(\vec{q}) = \frac{1}{2\pi} \int \int m(\vec{r}) e^{-i\vec{q} \cdot \vec{r}} d^2r \]

(2.56)

\(^9\) The model of the free energy according to Villain used in Sec. 2.3 is equivalent to the free energy by Garel and Doniach analyzed in Sec. 2.4 provided the correct thickness dependence of the various terms is heeded (cf. Subsec. 2.3.7).

Based on Eqs. (2.50) and (2.51), we obtain the following expression for the density of the free energy according to Villain:

\[ \frac{F(D)}{Na^2} = \sum_{\vec{k}} \left\{ \frac{D}{2a} \left\{ J + 2g \bar{x}_0 \right\} k^2 + \frac{D}{2a^3} k_B (T - T_c) + \frac{D}{a^3} g \left\{ \frac{2}{\bar{x}_0} + c \right\} - \frac{D^2}{a^3} \pi g k \right\} m_{\vec{k}} m_{-\vec{k}} \]

\[ + \frac{1}{N} \frac{D}{12a^3} k_B T \sum_i m_i^4 + \cdots \]

The term proportional to \( m^4 \) is identical to the corresponding term in Eq. (2.54), and with \( J \gg 2g \bar{x}_0 \), the correspondence to Eq. (2.70) is apparent. The equivalence of the terms that are due to the dipolar interaction becomes clear when the approximation \( (1 - e^{-qD}) \approx qD - \frac{1}{2} q^2 D^2 \) is combined with the correspondence

\[ g_{\text{Villain}} = \frac{\mu_0 (g_{\mu_B})^2}{4\pi a^3} \]

The only discrepancy is due to the choice of the boundary \( \bar{x}_0 \), which was chosen such as to give the correct result for \( k = 0 \). A complete equivalence would be established with \( \bar{x}_0 = \frac{2}{\pi - c} \) instead of \( \bar{x}_0 = \frac{4}{\pi} \). However, the exact value of \( \bar{x}_0 \) is not essential for the calculations.
Using this definition, the first term in Eq. (2.54) can be transformed as follows:

\[
\int\int (\nabla m(\vec{r})) \cdot d^2r = -\int\int\int m(\vec{q}) m(\vec{q}') \vec{q} \vec{q}' \left( \int\int \frac{1}{(2\pi)^2} e^{i(\vec{q} + \vec{q}') \cdot \vec{r}} \, d^2q \, d^2q' \right) \, d^2q \, d^2q' = -\int\int\int m(\vec{q}) m(\vec{q}') \vec{q} \vec{q}' \delta(\vec{q} + \vec{q}') \, d^2q \, d^2q' = \int\int m(\vec{q}) m(-\vec{q}) \, d^2q \ . \quad (2.57)
\]

A similar calculation yields the corresponding expression for the second term in Eq. (2.54):

\[
\int\int m^2(\vec{r}) \, d^2r = \int\int m(\vec{q}) m(-\vec{q}) \, d^2q \ . \quad (2.58)
\]

A Fourier transform of the term in Eq. (2.54) proportional to \(m^4\) will not be needed.

For the dipolar energy, we take the magnetization within the film to be constant along the axis perpendicular to the film. Then, the magnetic scalar potential is due to magnetic charges residing on the two surfaces of the film (cf. 2.1.2). If \(\vec{m}\) is the magnetization within the film, the magnetic surface charge \(\sigma\) is given by \(\vec{n} \cdot \vec{m}\) where \(\vec{n}\) is a unit vector perpendicular to the surface.

Now we can calculate the scalar potential due to the magnetic charges on surface \(i\) at a location \(\vec{r}_j\) on surface \(j\):

\[
\Phi_i(\vec{r}_j) = \frac{1}{4\pi} \int_{S_i} \frac{\sigma(\vec{r}_i)}{|\vec{r}_j - \vec{r}_i|} \, dS_i \ . \quad (2.59)
\]

The total dipolar energy comprises both interactions between the charges on one surface with the charges on the other surface and interactions between charges on a single surface. It reads:

\[
F_d = \frac{\mu_0}{4\pi} \int_{S_1} \sigma(\vec{r}_1) \Phi_2(\vec{r}_1) \, dS_1 + \frac{\mu_0}{24\pi} \int_{S_1} \sigma(\vec{r}_1) \Phi_1(\vec{r}_1) \, dS_1 + \frac{\mu_0}{4\pi} \int_{S_2} \sigma(\vec{r}_2) \Phi_2(\vec{r}_2) \, dS_2
\]

\[
= \frac{\mu_0}{4\pi} \int_{S_1} \sigma(\vec{r}_1) \Phi_2(\vec{r}_1) \, dS_1 + \frac{\mu_0}{4\pi} \int_{S_1} \sigma(\vec{r}_1) \Phi_1(\vec{r}_1) \, dS_1 \ . \quad (2.60)
\]

In the further calculations, we will take advantage of the equation\(^{11}\)

\[
\frac{1}{|\vec{r}_i - \vec{r}_j|} = \frac{1}{2\pi} \int\int \frac{e^{i\vec{q} \cdot (\vec{r}_i - \vec{r}_j)}}{q} \, d^2q \ , \quad (2.61)
\]

\(^{10}\)This assumption is justified as the perpendicular anisotropy in the system Fe/Cu(100) is strong.

\(^{11}\) The formula can be found in [44]. It can be derived from the formula

\[
\frac{1}{|\vec{r} - \vec{r}'|} = \frac{1}{2\pi^2} \int\int\int \frac{e^{i\vec{q} \cdot (\vec{r} - \vec{r}')}}{q^2} \, d^3q \ .
\]
where \( \vec{\rho}_i \) is the in-plane component and \( z_i \) the out-of-plane component of \( \vec{r}_i \). As only ultrathin films are considered here, \( \vec{\rho}_i \) can be replaced with \( \vec{r}_i \). Using Eq. (2.61) and

\[
\sigma(\vec{r}) = \frac{g \mu_B}{a^3} m(\vec{r}) ,
\]

we can write the dipolar energy as

\[
F_d = \frac{1}{2\pi} \iiint m(\vec{r}) g(\vec{r} - \vec{r}') m(\vec{r}') d^2r d^2r' ,
\]

where

\[
g(\vec{r} - \vec{r}') = \frac{\mu_0 (g \mu_B)^2}{4\pi a^6} \iint \frac{1}{q} \left( 1 - e^{-qD} \right) e^{i\vec{q} \cdot (\vec{r} - \vec{r}')} d^2q .
\]

The integrations in Eq. (2.63) extend over one surface only. Hence, the subscript for distinguishing the two surfaces of the film is no longer needed. The factor \( 1 - e^{-qD} \) in Eq. (2.64) takes into account both interactions within the individual surfaces (1) and the interaction between the two surfaces (\( -e^{-qD} \)). The minus sign in front of the exponential function results from the fact that the magnetic charge on the lower surface is the negative of the charge on the upper surface. With

\[
g(q) = \frac{\mu_0 (g \mu_B)^2}{4\pi a^6} \frac{1}{q} \left( 1 - e^{-qD} \right) ,
\]

cited, for example, in [24]. The derivation goes as follows

\[
\frac{1}{|\vec{r} - \vec{r}'|} = \frac{1}{2\pi^2} \iiint e^{i\vec{q} \cdot (\vec{r} - \vec{r}') + i\vec{q}_z (z - z')} \frac{d^3q}{q_x^2 + q_y^2 + q_z^2} = \frac{1}{2\pi^2} \iiint e^{i\vec{q}_1 \cdot (\vec{r} - \vec{r}') + i\vec{q}_z (z - z')} \frac{d^3q}{1 + \left( \frac{q_z}{q_1} \right)^2} = \frac{1}{2\pi^2} \iiint e^{i\vec{q}_1 \cdot (\vec{r} - \vec{r}') + i\vec{q}_z (z - z')} \frac{d^2q}{q_1} d\vec{q}_z .
\]
the first integration in the direct space of Eq. (2.63) is given by
\[
\int \int m(\vec{r})g(\vec{r}-\vec{r}')d^2r = \int \int \int m(\vec{r})g(\vec{r})e^{i\vec{q} \cdot (\vec{r}-\vec{r}')}d^2r d^2q d^2q' d^2q''
\]
\[
= \frac{1}{2\pi} \int \int \int \int m(\vec{q}'')e^{i\vec{q}'' \cdot \vec{r}'} g(q) e^{i\vec{q} \cdot (\vec{r}-\vec{r}')} d^2r d^2q d^2q' d^2q''
\]
\[
= 2\pi \int \int \int m(\vec{q}'')g(q)e^{-i\vec{q} \cdot \vec{r}'} \left( \frac{1}{2\pi} \int \int e^{i(\vec{q}''+\vec{q}) \cdot \vec{r}'} d^2r \right) d^2q' d^2q''
\]
\[
= 2\pi \int \int \int m(\vec{q}'')g(q)e^{-i\vec{q} \cdot \vec{r}'} \delta(\vec{q}' + \vec{q}) d^2q' d^2q''
\]
\[
= 2\pi \int \int m(-\vec{q}'')g(q)e^{-i\vec{q} \cdot \vec{r}'} d^2q' d^2q'' .
\]

The dipolar energy as a function of \( q \) is obtained after performing the second integration in the direct space:
\[
F_d = \int \int \int m(\vec{q}')m(-\vec{q}'')g(q)e^{-i\vec{q} \cdot \vec{r}'} d^2q' d^2q'' .
\]
\[
= \frac{1}{2\pi} \int \int \int \int m(\vec{q}'')e^{i\vec{q}'' \cdot \vec{r}'} m(-\vec{q}'')g(q)e^{-i\vec{q} \cdot \vec{r}'} d^2q d^2q'' d^2q' d^2q'''
\]
\[
= 2\pi \int \int \int m(\vec{q}'')m(-\vec{q}'')g(q) \left( \frac{1}{2\pi} \int \int e^{i(\vec{q}''-\vec{q}) \cdot \vec{r}'} d^2r \right) d^2q d^2q''
\]
\[
= 2\pi \int \int \int m(\vec{q}'')m(-\vec{q}'')g(q) \delta(\vec{q}'' - \vec{q}) d^2q d^2q''
\]
\[
= 2\pi \int \int m(\vec{q}'')m(-\vec{q}'')g(q) d^2q .
\]

Neglecting the term proportional to \( n^4 \) in Eq. (2.54), the free energy can be written as
\[
F = \int \int m(\vec{q}')m(-\vec{q}')f(q) d^2q ,
\]
where
\[
f(q) = \frac{D}{2a} J q^2 + \frac{D}{2a^3} k_B (T - T_c) + \frac{\mu_0 (g\mu_B)^2}{4\pi} \frac{2\pi}{q} \left( 1 - e^{-qD} \right) .
\]

The wave vector \( q_0 \) that minimizes the free energy is determined by
\[
\frac{df}{dq}(q_0) = 0 .
\]
2.4. CONTINUUM MODEL

It reads

\[ q_0 = \frac{3\pi a D}{2\pi a D^2 + 3 \frac{\mu_0}{4\pi} \frac{(g \mu_B)^2}{J}} \approx \frac{\mu_0}{4\pi} \frac{(g \mu_B)^2}{J} \cdot \frac{D}{a^2}, \]  

(2.72)

which is equivalent to the expression obtained in Eq. (2.52). The transition temperature is a function of \( q_0 \) and given by

\[ T_c(q_0) = T_c - q_0^2 a^2 \frac{J}{k_B} - 4\pi \frac{\mu_0}{4\pi} \frac{(g \mu_B)^2}{a} \cdot \frac{(1 - e^{-q_0 D})}{q_0 D} \approx T_c - q_0^2 a^2 \frac{J}{k_B} - 4\pi \frac{\mu_0}{4\pi} \frac{(g \mu_B)^2}{a} . \]

(2.73)

With \( T_c(q_0) \), the Fourier transform of the free energy density can be written in a simple form:

\[ f(q_0) = \frac{D}{2a^3} k_B \{ T - T_c(q_0) \} , \]

(2.74)

\[ f''(q_0) = -\frac{D}{2a^3} k_B T_c''(q_0) . \]

(2.75)

For small deviations from \( q_0 \), a Taylor expansion of \( f(q) \) can be introduced in the free energy:

\[ F = \int \int m(\vec{q}) m(-\vec{q}) \left( f(q_0) + \frac{(q - q_0)^2}{2} f''(q_0) \right) d^2q . \]

(2.76)

We are now interested in the difference in energy associated with a deviation of the wave vector from \( q_0 \):

\[ \delta F = \int \int m(\vec{q}) m(-\vec{q}) \left( \frac{(q - q_0)^2}{2} f''(q_0) \right) d^2q \approx \int \int m(\vec{q}) m(-\vec{q}) \left( \frac{(q - q_0)^2}{2} \cdot \frac{(q + q_0)^2}{(2q_0)^2} \right) f''(q_0) d^2q \]

(2.77)

\[ \approx \int \int m(\vec{q}) m(-\vec{q}) \left( \frac{q^2 - q_0^2}{2(2q_0)^2} \right) f''(q_0) d^2q \]

(2.78)

\[ \approx \frac{f''(q_0)}{8q_0^2} \int [(\nabla^2 m(\vec{r}))^2 - 2q_0^2 (\nabla m(\vec{r}))^2 + q_0^4 m^2(\vec{r})] d^3r \]

(2.79)

For small deformations, the factor introduced in Eq. (2.77) is close to unity. An inverse Fourier transform is used to obtain Eq. (2.79) from Eq. (2.78). We now
evaluate the free energy density in Eq. (2.79) by assuming a magnetization of the form

\[ m(x, y) = m_s \cos(q[x - u(x, y)]) , \tag{2.80} \]

where the local coordinate system is supposed to be oriented in such a way that the \( x \) axis is parallel to the wave vector and that the \( y \) axis is parallel to the domain wall. The quantity \( u(x, y) \) describes the displacement of the domain at \((x, y)\) along the \( x \) axis. The magnitude \( m_s \) will be taken as constant. This phase-only approximation has proven innocuous for a two-dimensional \( x-y \) model [45]. Substituting Eq. (2.80) into the free energy density of Eq. (2.79), we obtain the following expression

\[
\left[ (\nabla^2 m)^2 - 2q_0^2(\nabla m)^2 + q_0^4m^2 \right] = \\
\frac{1}{2}m_s^2 \left\{ (q^2 - q_0^2)^2 + 2(q^2 - q_0^2) \left[-2q^2u_x + q^2(\nabla u)^2 \right] \\
+ 4q^4u_x^2 + q^2(\nabla^2 u)^2 - 4q^4u_x(\nabla u)^2 + q^4(\nabla u)^4 \right\} , \tag{2.81}
\]

where the prefactor \( \frac{1}{2} \) is due to averaging \( \cos^2(q[x - u(x, y)]) \) over a period, and \( u_x \) denotes the derivative of \( u \) with respect to \( x \). The magnitude is obtained by minimizing the free energy density including the term proportional to \( m^4 \) (cf. Eq. (2.54)):

\[
f = \frac{1}{2}f(q_0)m_s^2 + \frac{3}{8}\frac{D}{12a^3}k_B T_c m_s^4 , \tag{2.82}
\]

where the prefactors are again due to averaging. The magnitude reads

\[ m_s = 2\sqrt{\frac{T_c(q_0) - T}{T_c}} . \tag{2.83} \]

In order to ensure that the expectation value of \( u_x \) vanishes, \( q \) must equal \( q_0 \). Under this condition and considering only terms quadratic in \( u \), we can rewrite the free
energy in terms of elastic constants $B$ and $K$: \(^{12}\)

$$\delta F = \int \int \left\{ \frac{1}{2} B \left( \frac{\partial u}{\partial x} \right)^2 + \frac{1}{2} K \left( \frac{\partial^2 u}{\partial y^2} \right)^2 \right\} \, dx \, dy \, , \quad (2.84)$$

where

$$B = \frac{2\pi^2}{a^2} \frac{T_c(q_0) - T}{T_c} \frac{\left( \frac{\mu_0}{4\pi} \frac{(g\mu_B)^2}{a^3} \right)^2}{J} \left( \frac{D}{a} \right)^3 \, , \quad (2.85)$$

$$K = \frac{1}{2} \frac{T_c(q_0) - T}{T_c} J \frac{D}{a} \, . \quad (2.86)$$

The result of Eq. (2.84) has been obtained by Sornette [11]. Note, however, that the elastic coefficients in [11] are slightly different. They correspond to the values expected in thicker films where the exponential $e^{-qD}$ in Eq. (2.64) is negligible.

### 2.5 Topological Defects

The ground state of an ultrathin magnetic film with a sufficiently strong perpendicular anisotropy is given by a stripe domain pattern (cf. Sec. 2.2). Although associated with an increased energy due to bending and compression, Eq. (2.84), defects can enter the perfect stripe pattern at higher temperatures (cf. Sec. 2.7).

Topological defects are singularities of the order-parameter space. The types of defects that can occur in a system with a given order-parameter space and their interactions depend on the symmetry of the order-parameter space. The theory of homotopy is capable of dealing with these issues. After an introduction to the main concepts of homotopy theory in the context of ordered media, homotopy theory is applied to two-dimensional nematics and smectics. Due to the Abelian nature of the fundamental group of the order-parameter space of nematics, the classification of defects in nematics is simple and straightforward. In the case of two-dimensional smectics, however, the non-Abelian nature of the fundamental group leads to a more

\(^{12}\) The free energy of Eq. (2.84) is not invariant under a global rotation of the sample. Under such an operation, the stripe width along the $x$ axis will change although the real stripe width is not altered. In order to account for this effect, higher order terms can be introduced into the free energy of smectics:

$$\delta F = \int \int \left\{ \frac{1}{2} B \left[ \frac{\partial u}{\partial x} - \frac{1}{2} \left( \frac{\partial u}{\partial y} \right)^2 \right]^2 + \frac{1}{2} K \left( \frac{\partial^2 u}{\partial y^2} \right)^2 \right\} \, dx \, dy \, .$$

Note that these higher order terms are already present in Eq. (2.81). A detailed discussion of this issue is given in [11].
complicated structure. Individual defects and their interactions have to be analyzed in terms of conjugacy classes. To the best knowledge of the author, an analysis of two-dimensional smectics in the framework of homotopy theory has not been presented before.

Finally, the energies associated with the defects are calculated within the continuum model presented in Sec. 2.4. The elementary defects of nematics and smectics are disclinations and dislocations. Their energy determines their stability. The knowledge of their energy is important for understanding the behavior of the domain pattern in ultrathin Fe films on Cu(001).

2.5.1 Homotopy Theory

In order to capture topological order we can use the concept of an order parameter introduced by Landau [46]. The order parameter is a continuous field describing the system at each point. The topology of the order-parameter space, also called manifold of internal states, now determines the classes of defects that can occur in the system. The adequate framework for the analysis of defects in ordered media is the theory of homotopy. It was first used for the description of ordered media in the studies by Toulouse and Kléman [47], and independently by Volovik and Mineev [48]. In this subsection, the main concepts and results of the application of the theory of homotopy to ordered media shall be presented, a more comprehensive treatment can be found in a review paper by Mermin [49].

It is assumed that the order parameter field is well defined and continuous throughout the medium with the exception of some singular points which characterize defects. Due to the continuity of the order parameter field around defects, the class of a defect can be deduced from the behavior of the order parameter field in the vicinity of the defect. A central role is played by mappings from closed contours around a defect in the real space into the order-parameter space. Two such mappings are said to be homotopic if they can be deformed into one another continuously. Any explicit construction of such a deformation is called a homotopy.

The possible mappings of a closed contour in the real space into the order-parameter space are given by the topology of the order-parameter space. Information about defects can thus be gained from an analysis of the order-parameter space. Loops in

\[13\] Note that some singularities of the order-parameter field can be removed by a continuous variation of the order-parameter field and a homogeneous configuration is attained. These singularities are said to be removable or topologically unstable. When such an elimination of a singularity is not possible, the singularity is called topologically stable. It is then a so-called topological defect. Topological stability is not necessarily equivalent to physical stability. Energetic barriers can hamper configurational changes even in the case of topologically unstable singularities. Conversely, topologically stable singularities cannot be removed by a local fluctuation of the order parameter. They are physically stable.
the order-parameter space can be described in terms of continuous maps $f$ of the real interval $0 \leq z \leq 1$, with $f(0)=f(1)$. Loops starting and ending at $x$ are called loops based at $x$. We can define a product $f \circ g$ of two loops $f$ and $g$ based at $x$ as the loop given by

$$f \circ g(z) = \begin{cases} f(2z), & 0 \leq z \leq \frac{1}{2} \\ g(2z-1), & \frac{1}{2} \leq z \leq 1 \end{cases} \quad (2.87)$$

The set of loops based at $x$ with this product does not form a group. A group structure is, however, obtained if the individual loops are replaced by homotopy classes of loops, that is sets of loops the members of which can be continuously transformed into each other.\(^{14}\) Homotopy classes will be denoted by $[f]$ where $f$ is a representative of the corresponding class. The group product is thus given by

$$[f \circ g] = [f] \circ [g] \quad (2.88)$$

The set of homotopy classes with this product is called the fundamental group at $x$, $\pi_1(R,x)$, where $R$ is the order-parameter space. It can be shown that fundamental groups of a connected space based at different points in the order-parameter space are isomorphic. There is thus a single abstract group $\pi_1(R)$, of which the based fundamental groups are based isomorphic copies. It is called the fundamental group of $R$, or the first homotopy group of $R$.

In general, there is no reason why mappings of closed contours into the order-parameter space should all share a single common point $x$ in the order-parameter space. A defect is therefore characterized by a set of loops in the order-parameter space that are freely homotopic, and not based at a certain point $x$. It can now be shown that classes of freely homotopic loops in $R$ can be labeled by the conjugacy classes of $\pi_1(R)$. When the fundamental group of an order-parameter space is Abelian, the conjugacy classes consist of single group elements. Then each mapping of a closed contour in the real space into the order-parameter space determines a unique element of the fundamental group. In non-Abelian groups, the situation is somewhat more difficult.

Defects can combine to form a new defect. Since the loop in the order-parameter space given by a real-space contour surrounding two defects is freely homotopic to the product of loops given by contours surrounding each defect separately, the defect resulting from the combination of the two defects can only be characterized

\(^{14}\)Defects belonging to the same homotopy class are said to be topologically equivalent, i.e. they can be transformed into each other by a local surgery. Such a local surgery does not affect the order parameter beyond a certain small distance from the defect. The homotopy class of the defect is thus preserved.
by conjugacy classes of the fundamental group whose members are the products of members of the classes characterizing the original pair of defects. The combination laws are thus given by the multiplication table of the conjugacy classes.

The computation of the fundamental group of an order-parameter space can be reduced to a simple algorithm when the symmetry of the order parameter space is taken into account. Let \( G \) be a continuous group of transformations acting transitively on the order-parameter space \( R \), i.e. for each pair \( f_1 \) and \( f_2 \) of possible values of the order parameter there is (at least) one element \( g \) in \( G \) which takes \( f_1 \) into \( f_2: f_2 = gf_1. \) The set of elements \( g \) of \( G \) that leave a given value \( f \) of the order parameter unchanged, i.e. \( gf = f \), forms a group \( H_f \) called isotropy subgroup of \( f \) or little group of \( f \). The space of cosets \( G/H_f \) of \( H_f \) in \( G \) is then equivalent to the order-parameter space itself [50]. The fundamental theorem on the fundamental group now states that the fundamental group \( \pi_1(G/H_f) \) of the coset space \( G/H_f \) of a connected, simply connected continuous group \( G \) is isomorphic to the quotient group \( H_f/H_0^f \) where \( H_0^f \) is the set of elements in \( H_f \) that are connected to the identity by continuous paths lying entirely in \( H_f \). The requirement that the group \( G \) be simply connected poses no restriction on the applicability of the fundamental theorem as any continuous group can be embedded in a larger group known as the universal covering group that is simply connected.

### 2.5.2 Defects in Two-Dimensional Nematics and Smectics

For a topological analysis of the stripe-domain pattern, the magnetization is not an adequate order parameter. The local orientation of a domain pattern can, however, be described by a director field indicating the axis along which the stripes are oriented at each point. The corresponding order-parameter space can be mapped onto a unit circle where diametrically opposite points are identified. In contrast to the vector field of the magnetization, the director field does not distinguish between opposite directions.

The order-parameter space \( R \) of a two-dimensional director field can thus be expressed as \( R = S_1/Z_2 \), where \( S_1 \) is the one-dimensional sphere describing the rotations and \( Z_2 \) is the two-element group due to the identification of opposite points in \( S_1 \). The corresponding fundamental group is \( \pi_1(R) = \mathbb{Z} \), i.e. it is isomorphic to the group of integers [51]. Consequently, the fundamental group of the two-dimensional director field is Abelian and each defect is unambiguously represented by one element of the fundamental group. Each element can be labeled with the corresponding winding number \( k \). Two defects with winding numbers \( k_1 \) and \( k_2 \) can combine to form a defect with winding number \( k_3 = k_1 + k_2 \). The winding number \( k \) indicates the amount by which the angle of a director field changes when a closed contour in the real space is followed. For each closed contour, the change in angle has to be a
multiple of $\pi$. The winding number $k$ now describes this change in angle as a fraction of $2\pi$, it can thus assume the values of integer and half-integer numbers. Defects in a two-dimensional director field are called disclinations. A two-dimensional director field corresponds to a nematic liquid crystal.

A stripe pattern such as the magnetic domains in perpendicularly magnetized ultrathin films considered here, however, is not only characterized by the orientation of the stripes. If the fact that the stripe domains are equally spaced is taken into account, we have the situation of a two-dimensional smectic A liquid crystal.\footnote{Note that due to the inequivalence of spin-up and spin-down domains, this analogy only applies when one of these components is considered as the boundary separating the branches of the other component that make up the lamellar structure of two-dimensional smectic A liquid crystal.} In contrast to a nematic, which has a continuous translational symmetry, such a system can only be brought into itself by translations perpendicular to the stripes that are multiples of the stripe width $L$. The order-parameter space is thus a rectangle \{(t, r), 0 \leq t \leq L, 0 \leq r \leq \pi\}. The sides of the rectangle are identified by the rules $(0, r) \equiv (L, r)$ and $(t, 0) \equiv (L - t, \pi)$. A translation about $t$ corresponds to a translation about $L - t$ in the system rotated by $\pi$. Hence, the order-parameter space is given by the Klein bottle, and not by a torus. The fundamental group of the Klein bottle is isomorphic to the semidirect product of the group $\mathbb{Z}_{\text{trans}}$ describing translations by the group $\mathbb{Z}_{\text{rot}}$ describing rotations, which are both isomorphic to $\mathbb{Z}$\footnote{An overview of the fundamental groups for three-dimensional mesomorphic states is given in [53] and [54].}:

$$\pi_1(R) = \mathbb{Z}_{\text{trans}} \wedge \mathbb{Z}_{\text{rot}}.$$  \tag{2.89}$$

Every point defect in a smectic A liquid crystal is thus characterized by a pair of numbers $(n, m)$. Elements $(n, 0)$ correspond to point dislocations with a Burgers vector $nL$, and elements $(0, m)$ correspond to point disclinations of integer (even $m$’s) and half-integer strength (odd $m$’s), the winding number $k$ being given by $k = \frac{1}{2}m$. The fundamental group as a semidirect product is not Abelian, which makes the possible results of combining two defects more intricate than in the case of a director field with unbroken translational symmetry. The combination of two defects can result in any element of the conjugacy classes formed by the product of the individual conjugacy classes the defects belong to.
The associative multiplication law for two elements \((n_1, m_1)\) and \((n_2, m_2)\) of the fundamental group of the Klein bottle is given by:

\[
(n_1, m_1)(n_2, m_2) = (n_1 + (-1)^{m_1} \cdot n_2, m_1 + m_2)
\]

(2.90)

The identity element is:

\[
(0, 0)
\]

(2.91)

The inverse of \((n, m)\) is:

\[
((-1)^m \cdot (-n), -m)
\]

(2.92)

The conjugacy classes can be classified as follows:

\[
E = \{(0, 0)\} \\
E_g = \{(0, g)\} \quad , \quad g \in 2\mathbb{Z}/\{0\} \\
G^\gamma_g = \{(\gamma, g), (-\gamma, g)\} \quad , \quad \gamma \in 2\mathbb{N} \quad , \quad g \in 2\mathbb{Z} \\
U^u_g = \{(u, g), (-u, g)\} \quad , \quad u \in (2\mathbb{N} + 1) \quad , \quad g \in 2\mathbb{Z} \\
G_a = \{(p, u) \quad , \quad p \in 2\mathbb{Z}\} \quad , \quad u \in (2\mathbb{Z} + 1) \\
U_a = \{(p, u) \quad , \quad p \in (2\mathbb{Z} + 1)\} \quad , \quad u \in (2\mathbb{Z} + 1)
\]

(2.93)\text{ - } (2.98)

The multiplication table of the conjugacy classes is given in Tab. 2.3. The multiplication table is important for analyzing the outcome of the merger of two defects.

In the absence of further nearby defects, two defects \((n_1, m_1)\) and \((n_2, m_2)\) can merge directly to form the defect characterized by \((n_1, m_1)(n_2, m_2)\). The situation is more complicated when one of the defects first circumnavigates another defect.

\footnote{Let \(G\) be an arbitrary group. Let \(\Omega\) be a group of automorphisms of \(G\). Then the composite \((g, \omega)\) where \(g \in G\) and \(\omega \in \Omega\) can be given a group structure. The associative multiplication law reads [55]:

\[
(g, \omega)(\bar{g}, \bar{\omega}) = (g\omega(\bar{g}), \omega\bar{\omega})
\]

The identity element is the composite \((e, \epsilon)\) of the identity elements \(e\) of \(G\) and \(\epsilon\) of \(\Omega\). The inverse of \((g, \omega)\) is given by \((\omega^{-1}(g^{-1}), \omega^{-1})\). The composite of an arbitrary group \(G\) and a group \(\Omega\) of its automorphisms with the law of multiplication defined above is called the semidirect product of \(G\) and \(\Omega\).

In the case of smectics, the group \(G\) is \(Z_{\text{trans}}\), which is isomorphic to \(Z\). The only automorphisms of \(Z\) are the multiplications with 1 or \(-1\). Now there are two homomorphisms associating to each element of \(Z_{\text{rot}}\) an element of the multiplicative group consisting of 1 and \(-1\). The first homomorphism is the trivial operation that maps every element of \(Z_{\text{rot}}\) onto 1. In this case, the semidirect product is equal to the direct product of \(Z_{\text{trans}}\) and \(Z_{\text{rot}}\). The second homomorphism associates to each element \(m \in Z_{\text{rot}}\) the multiplication with \((-1)^m\). Hence, the multiplication law of the fundamental group of the Klein bottle is \((n_1, m_1)(n_2, m_2) = (n_1 + (-1)^{m_1} \cdot n_2, m_1 + m_2)\).}
Table 2.3: Multiplication table for conjugacy classes of defects in two-dimensional smectics. The fundamental group of two-dimensional smectics is \( \pi_1(R) = \mathbb{Z}_{\text{trans}} \times \mathbb{Z}_{\text{rot}} \). The corresponding conjugacy classes are explained in the text. The result of the merger of two defects \( a \in A \) and \( b \in B \) can be any element \( c \) in any conjugacy class \( C \) contained in the product of \( A \) and \( B \). The resultant defect \( c \) is determined by the interplay with additional defects.

A defect \((n_1, m_1)\) that circumnavigates a defect \((n_3, m_3)\) is transformed into a defect \((n_4, m_4)\) in turn transforms into \((n_1, m_1)^{-1}(n_3, m_3)\). The defect \((n_3, m_3)\) circumnavigated by defect \((n_1, m_1)\) in turn transforms into \((n_3, m_3)^{-1}(n_1, m_1)\) [56]. Defect \((n_4, m_4)\) can now merge with defect \((n_2, m_2)\) to form \((n_4, m_4)(n_2, m_2)\), which in general is unlike \((n_1, m_1)(n_2, m_2)\). The defects \((n_1, m_1)\) and \((n_4, m_4)\) belong to the same conjugacy class, but the defects \((n_1, m_1)(n_2, m_2)\) and \((n_4, m_4)(n_2, m_2)\) need not be in the same conjugacy class.

The effect of circumnavigating another defect is illustrated in Fig. 2.3 for the homotopy class of the circumnavigating defect. But as only the relative motion of the defects is important, the results also apply for the homotopy class of the circumnavigated defect.

The influence of nearby defects is illustrated in Fig. 2.4. The two dislocation defects \((1, 0)\) and \((-1, 0)\) of Fig. 2.4 a can annihilate each other to form \((0, 0)\), Fig. 2.4 b. The two defects \((1, 0)\) and \((-1, 0)\) are in the same conjugacy class \(U_0^1\). The product of \(U_0^1\) with \(U_0^1\) is the sum of the classes \(G_0^0 = E \) and \(G_0^2\). The ‘defect’ \((0, 0)\) is the only element in \(E\). The two defects \((2, 0)\) and \((-2, 0)\) of \(G_0^2\) are obtained when one of the defects circumnavigates a \((0, 1)\) disclination before merging with the other defect. Fig. 2.4 c shows defect \((1, 0)\) after it has circumnavigated the \((0, 1)\) disclination. It is now described by \((0, 1)^{-1}(1, 0)(0, 1) = (-1, 0)\) and can merge with the other \((-1, 0)\).
Figure 2.3: Homotopy class of a defect after circumnavigating another defect. a, Initial configuration in the real space with defects $\alpha$ and $\beta$. Two oriented loops $\Gamma_\alpha$ and $\Gamma_\beta$ around $\alpha$ and $\beta$ respectively are shown. They are based at $x$. Defect $\alpha$ is now supposed to circumnavigate defect $\beta$. Different stages of this process are depicted in Figs. e–h. As the configuration in the real space is varied in a continuous way, the loop around defect $\alpha$ follows the moving defect. The final loop in Fig. h is now homotopic to $\Gamma_{\beta-1}\Gamma_\alpha\Gamma_\beta$. The reason for this is shown in Figs. a–d. In Fig. b, an additional loop homotopic to $\Gamma_{\beta-1}$ is added. The orientation of this loop is opposite to the orientation of loop $\Gamma_\beta$. If the loops $\Gamma_\beta$, $\Gamma_\alpha$ and $\Gamma_{\beta-1}$ are combined in this order and if only the beginning of $\Gamma_\beta$ and the end of $\Gamma_{\beta-1}$ remain fixed at $x$, a loop like the one in Fig. c can be obtained. It is homotopic to the final loop in Fig. d. As the mapping from the real space onto the order-parameter space is continuous, we find the same relation between the corresponding loops in the order-parameter space.
2.5. TOPOLOGICAL DEFECTS

**Figure 2.4:** Example for the merger of defects in smectics. **a.** Initial configuration with two dislocations $(1,0)$ and $(-1,0)$ next to each other. These two defects can merge and annihilate each other to form a $(0,0)$ configuration (**b**). If, however, the dislocation $(1,0)$ circumnavigates a $(0,1)$ disclination as indicated by the arrow in Fig. **a**, it becomes a $(-1,0)$ dislocation (**c**). The dislocations $(1,0)$ and $(-1,0)$ are thus in the same conjugacy class. The two $(-1,0)$ dislocations in Fig. **c** can now merge to form a $(-2,0)$ dislocation (**d**). The movement of the lower $(-1,0)$ dislocation thereby follows the arrow in Fig. **c** and is called a glide movement. A glide movement involves the breaking of layers, which can be suppressed by energy barriers.

A defect to form a $(-2,0)$ defect, which is an element of $G_0^2$. The $(-2,0)$ defect is depicted in Fig. **2.4d**.

Obviously, circumnavigating another defect does not change the conjugacy classes of the defects involved. The defects may remain the same as the $(0,1)$ disclination in the example above, or become a different member of the same conjugacy class as the $(1,0)$ dislocation which transforms into a $(-1,0)$ dislocation. But the defects $(0,0)$ and $(-2,0)$ obtained on different paths are not in the same conjugacy class. For energetic reasons, not all conceivable defects $(n,m)$ will be found in a real system. The energies associated with defects will be dealt with in Subsec. 2.5.3.
2.5.3 Energies Associated with Disclinations and Dislocations

Disclinations are primarily defects of the director field. In the calculation of the energy associated with a disclination, the contribution to the free energy proportional to the elastic constant $B$ will thus be neglected. In the derivation of the elastic free energy of Eq. (2.84) from Eq. (2.81), higher-order derivatives of the displacement field $u(x, y)$ along the $x$ axis perpendicular to the stripe direction were disregarded. Without the term proportional to $B$ but including the remaining lowest-order derivatives with respect to $x$, the free energy of a smectic reads:

\[ \delta F_{\text{nematic}} = \int \int \frac{1}{2} K \left\{ \left( \frac{\partial^2 u}{\partial x^2} \right)^2 + 2 \left( \frac{\partial^2 u}{\partial x^2} \right) \cdot \left( \frac{\partial^2 u}{\partial y^2} \right) \right\} \, dx \, dy . \tag{2.99} \]

The second term in this symmetric free energy can be integrated by parts so that the free energy reads:

\[ \delta F_{\text{nematic}} = \int \int \frac{1}{2} K \left\{ \left( \frac{\partial^2 u}{\partial x^2} \right)^2 + 2 \left( \frac{\partial^2 u}{\partial x \partial y} \right)^2 + \left( \frac{\partial^2 u}{\partial y^2} \right)^2 \right\} \, dx \, dy . \tag{2.100} \]

Introducing a unit vector $\vec{n}(\vec{r})$ parallel to the local director field and using

\[ n_x \approx \frac{\partial u}{\partial x} \, , \quad n_y \approx \frac{\partial u}{\partial y} \, , \tag{2.101} \]

we obtain the free energy in terms of the unit vector $\vec{n}$:

\[ \delta F_{\text{nematic}} = \int \int \frac{1}{2} K \left\{ \left( \frac{\partial^2 u}{\partial x} \vec{n} \right)^2 + \left( \frac{\partial^2 u}{\partial y} \vec{n} \right)^2 \right\} \, dx \, dy . \tag{2.102} \]

When the unit vector field $\vec{n}(\vec{r})$ is parameterized by an angular field $\theta(\vec{r})$, i.e.

\[ \vec{n}(x, y) = \begin{pmatrix} \cos(\theta(x, y)) \\ \sin(\theta(x, y)) \end{pmatrix} , \tag{2.103} \]

the free energy assumes a particularly simple form:

\[ \delta F_{\text{nematic}} = \int \int \frac{1}{2} K (\nabla \theta)^2 \, dx \, dy . \tag{2.104} \]

The corresponding Euler-Lagrange equation is given by

\[ \Delta \theta = 0 \, . \tag{2.105} \]
The solutions of the Euler-Lagrange equation are thus of the form

\[
\theta(r, \varphi) = A\varphi + B(r, \varphi) , \tag{2.106}
\]

\[
\theta(r, \varphi) = C \ln(r) + D(r, \varphi) , \tag{2.107}
\]

where \( A \) and \( C \) are two constants, and \( B(r, \varphi) \) and \( D(r, \varphi) \) are harmonic functions \[51\]. We will now look at singular solutions of the form

\[
\theta(r, \varphi) = S\varphi + \theta_0 , \quad S \in \frac{1}{2} \mathbb{Z} . \tag{2.108}
\]

The restriction on \( S \) ensures that \( \theta(r, \varphi) \) is a continuous director field, i.e. \( \theta(r, \varphi + 2\pi) = \theta(r, \varphi) + n\pi \) where \( n \) is an integer number. Applying the Laplacian, we obtain the following free-energy density:

\[
f_{\text{disclination}} = \frac{K S^2}{2 r^2} . \tag{2.109}
\]

The energy associated with a single disclination of strength \( S \) is now obtained after integrating the free-energy density over the plane:

\[
W_{\text{disclination}} = \pi KS^2 \ln \left( \frac{R}{r_{\text{core}}} \right) + W_{\text{core}} . \tag{2.110}
\]

\( W_{\text{core}} \) denotes the energy of the singular core of radius \( r_{\text{core}} \), and \( R \) is the radius of the film. The energy of a single disclination in an infinitely extended director field is thus infinite. The energy of a pair of disclinations of opposite sign, however, is finite. As the energy of a disclination is proportional to the square of its strength \( S \), a disclination with a high value of \( S \) will decay into a number of disclinations with a lower strength \( S_i \) so that \( S = \sum_i S_i \). In a nematic, we thus expect to find mainly \( S = \pm \frac{1}{2} \) disclinations.

Dislocations are defects of the translational order. The term proportional to \( B \) in the free energy can thus no longer be neglected. The free energy of a smectic, Eq. (2.84), reads:

\[
\delta F_{\text{smectic}} = \iint \left\{ \frac{1}{2} B \left( \frac{\partial u}{\partial x} \right)^2 + \frac{1}{2} K \left( \frac{\partial^2 u}{\partial y^2} \right)^2 \right\} \, dx \, dy . \tag{2.111}
\]

In the half-plane \( x > 0 \), the corresponding Euler-Lagrange equation is given by

\[
\lambda^2 \frac{\partial^4 u}{\partial y^4} = \frac{\partial^2 u}{\partial x^2} , \quad \text{where} \quad \lambda = \sqrt{\frac{K}{B}} \tag{2.112}
\]
is the so-called penetration depth related to the stripe width via\(^\text{18}\)

\[ L = 4\pi \lambda \] \hspace{1cm} (2.113)

The Euler-Lagrange equation has solutions of the form [57]:\(^\text{19}\)

\[ u(x, y) = \frac{b}{4} + \frac{b}{4\pi} \int_{-\infty}^{\infty} \frac{e^{iq_y y}}{iq_y} g_{q_y}(x) dq_y \hspace{1cm} (2.114) \]

where \( b \) is the modulus of the Burgers vector and

\[ g_{q_y}(x) = e^{-\lambda q_y^2 x} \] \hspace{1cm} (2.115)

Such solutions represent dislocations centered at the origin and symmetric with respect to a mirror transformation \( x \to -x \).

The energy of a dislocation with strength \( b \) is obtained by integrating over the plane:\(^\text{20}\)

\[ W_{\text{dislocation}} = \frac{K}{2} \frac{b^2}{\lambda r_{\text{core}}} + W_{\text{core}} \] \hspace{1cm} (2.116)

As in the case of disclinations, the energy of a dislocation is proportional to the square of the strength of the defect, here the modulus \( b \) of the Burgers vector. A decay into dislocations with smaller \( b \)'s is energetically favorable. The modulus of most Burgers vectors found in a sample should thus be equivalent to one period, i.e. the width of two stripes. In contrast to disclinations, however, the energy of a single dislocation in an infinitely extended medium is finite.

\(^{18}\) This relation follows directly from Eq. (2.81). The ratio of the two elastic constants \( K \) and \( B \) is accordingly given by \( \frac{K}{B} = \frac{1}{4\pi} \).

\(^{19}\) Let the boundary condition along the \( y \) axis be given by

\[ u(0, y < 0) \equiv 0 \hspace{1cm} u(0, y > 0) \equiv \frac{b}{2} \hspace{1cm} . \]

With the step function \( H(y) = \frac{1}{2} \int_{-\infty}^{\infty} \frac{\sin(q_y y)}{i q_y} dq_y \), the boundary condition can then be written as

\[ u(x, 0) = \frac{b}{4} + \frac{b}{4\pi} \int_{-\infty}^{\infty} \frac{e^{iq_y y}}{iq_y} dq_y \hspace{1cm} . \]

The form of the additional factor \( g_{q_y}(x) \), which takes into account the dependence on the \( x \) coordinate, is obtained by substituting the ansatz into the Euler-Lagrange equation.

\(^{20}\) This result is identical to the energy per unit length of a dislocation in three-dimensional media [58].
2.6 Order in Two Dimensions

While long-range order is present in three-dimensional crystals, thermal fluctuations prevent the same type of order in one- or two-dimensional crystals. This lack of long-range order was first noticed by Peierls and Bethe. They gave a qualitative argument for the one-dimensional case [59] and a quantitative argument based on the harmonic approximation for the one-dimensional system that can be extended to the two-dimensional case [60]. Calculating the fluctuations of the displacement field, Landau [61] showed that the fluctuations diverge in one- and two-dimensional crystals whereas fluctuations in three-dimensional crystals remain finite. Based on Bogoliubov’s inequality [62], Mermin [63] later gave a rigorous prove that the conventional crystalline long-range order is impossible in two-dimensional crystals with power-law potentials. Similarly, Mermin and Wagner [36] excluded ferromagnetic or antiferromagnetic ordering in one- or two-dimensional isotropic Heisenberg models. Again based on Bogoliubov’s inequality, Hohenberg [64] showed the absence of conventional long-range order in two-dimensional superconductors and superfluids. Calculating the asymptotic behavior of the correlation function, Berezinskii [65, 66] finds a logarithmic divergence for a number of systems such as two-dimensional crystals, the $x$-$y$ model of magnetism, the two-dimensional isotropic Heisenberg ferromagnet and the two-dimensional Bose liquid. The analysis is extended to arbitrary two-dimensional systems characterized by commutative continuous symmetry groups.

Not all two-dimensional systems, however, are marked by the lack of conventional long-range order. The two-dimensional Ising ferromagnet is such a system [67]. Moreover, even two-dimensional systems with no long-range (positional) order can be characterized by long-ranged orientational order of their elementary units [61].

The type of order present in a system is reflected in its correlation function. In a two-dimensional system, three types of order are distinguished: long-range, quasi-long-range, and short-range order. In a system with long-range order, the correlation function may decay initially but reaches a finite value at infinity. The corresponding fluctuations are finite even in an infinitely extended sample. When the fluctuations diverge with system size, however, the correlation function will vanish at infinity. A logarithmic divergence of the fluctuations leads to an algebraic (power-law) decay of the correlation function. This slow decay is the characteristic of quasi-long-range order. Faster divergences of the fluctuations bring about an exponential decay of the correlation function. This is the case of short-range order.

In the following subsections, both positional and orientational correlation functions of two-dimensional nematics and smectics will be determined. The basis is the

\[ \text{In three dimensions, an algebraic decay of the correlation function only occurs at isolated second order critical points [68]. An algebraic decay is not usual in three-dimensional melting as such transitions tend to be of first order.} \]
calculation of the mean-square fluctuations.

### 2.6.1 Two-Dimensional Nematics

Nematics are characterized by a director field. The only possible correlation in a nematic is that between the orientations of the director field at different locations. A positional correlation cannot be defined. Using the Fourier transform of the free energy density, the free energy of a two-dimensional nematic, Eq. (2.104), can be written as

\[
\delta F_{\text{nematic}} = \frac{1}{(2\pi)^2} \int \frac{1}{2} Kq^2 |\hat{\theta}(\mathbf{q})|^2 d^2q .
\]  

(2.117)

Due to the equipartition theorem, each degree of freedom will be associated with an energy of \(\frac{k_B T}{2} \). Thus we have

\[
\langle |\hat{\theta}(\mathbf{q})|^2 \rangle = \frac{k_B T}{Kn^2} .
\]  

(2.118)

The mean-square of the angular fluctuation can now be calculated by integrating Eq. (2.118) over the wave-vector space:

\[
\langle \theta^2(0,0) \rangle = \frac{1}{(2\pi)^2} k_B T \int \frac{1}{Kq^2} d^2q = \frac{1}{2\pi} \frac{k_B T}{K} \ln \left( \frac{R}{r_{\text{cutoff}}} \right) ,
\]  

(2.119)

where \(R\) is the radius of the sample, and \(r_{\text{cutoff}}\) a lower cutoff. While there is a natural lower cutoff for smectics, such a cutoff in a continuum model of a nematic is arbitrary.

Due to the logarithmic divergence of the mean-square fluctuation, the orientational order parameter defined as \(^{22}\)

\[
G_{n}^{\text{nematic}} = \langle e^{in\theta(r,\varphi)} \rangle = e^{-\frac{1}{2}n^2 \langle \theta^2(0,0) \rangle} ,
\]  

(2.120)

where \(n\) is an even integer, will decay algebraically with system size:

\[
G_{n}^{\text{nematic}} = \left( \frac{r_{\text{cutoff}}}{R} \right)^{\frac{n^2 k_B T}{K}} .
\]  

(2.121)

\(^{22}\)For a variable \(x\) with a Gaussian probability distribution of variance \(\sigma^2\), \(\langle e^{iqx} \rangle\) is the Fourier transform of the Gaussian distribution:

\[
\langle e^{iqx} \rangle = \int_{-\infty}^{\infty} e^{iqx} \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{x^2}{2\sigma^2}} dx = e^{-\frac{q^2}{2\sigma^2}} .
\]
Note that the requirement that \( n \) be an even integer reflects the character of a director field. Angles differing by \( \pi \) have to be identified. Odd integers \( n \) are not meaningful; they would depend on the stipulated range of angles \( \theta \). In order to determine the correlation function defined as

\[
g_n^{\text{nematic}}(r, \varphi) = \langle e^\{i[\theta(r, \varphi) - \theta(0, 0)]\} \rangle (2.122)
\]

\[
e^{\{-\frac{1}{2}n^2[\theta(r, \varphi) - \theta(0, 0)]^2\}} (2.123)
\]

\[
e^{\{-\frac{1}{2}n^2[\theta^2(r, \varphi) - 2\theta(r, \varphi)\theta(0, 0) + \theta^2(0, 0)]\}} (2.124)
\]

\[
e^{-n^2[\theta^2(0, 0) - \theta(r, \varphi)\theta(0, 0)]}, (2.125)
\]

we need to calculate:

\[
\langle \theta^2(0, 0) - \theta(r, \varphi)\theta(0, 0) \rangle = \frac{1}{(2\pi)^2} \frac{k_B T}{K} \int_0^{q_{\text{max}}} \int_0^{2\pi} \frac{1 - e^{-iqr\cos(\varphi)}}{q^2 \varphi} d\varphi dq (2.126)
\]

\[
= \frac{1}{(2\pi)^2} \frac{k_B T}{K} \int_0^{q_{\text{max}}} \int_0^{2\pi} \frac{1 - e^{-iqr\sin(\varphi)}}{q} d\varphi dq (2.127)
\]

\[
= \frac{1}{(2\pi)^2} \frac{k_B T}{K} \int_0^{q_{\text{max}}} \int_0^{2\pi} \frac{1 - \cos(qr\sin(\varphi))}{q} d\varphi dq (2.128)
\]

\[
= \frac{1}{\pi^2} \frac{k_B T}{K} \int_0^{q_{\text{max}}} \frac{2\pi}{q} - \int_0^{\pi} \cos(qr\sin(\varphi)) d\varphi dq (2.129)
\]

\[
= \frac{1}{2\pi} \frac{k_B T}{K} \int_0^{q_{\text{max}}} 1 - Jo(qr) dq (2.130)
\]

\[
\approx \frac{1}{2\pi} \frac{k_B T}{K} \ln \left( \frac{r}{r_{\text{cutoff}}} \right). (2.131)
\]

The approximation from Eq. (2.130) to Eq. (2.131) is explained in [69]. An alternative way of calculating this integral uses the properties of two-dimensional Green functions. We define

\[
\phi(\vec{r}) = \iint \frac{1 - e^{iq\vec{r}}}{q^2} d^2q (2.132)
\]

and note that

\[
\Delta \phi(\vec{r}) = \Delta \iint \frac{1 - e^{iq\vec{r}}}{q^2} d^2q = \int \int \frac{q^2 e^{iq\vec{r}}}{q^2} d^2q = \int \int e^{iq\vec{r}} d^2q = (2\pi)^2 \delta(\vec{r}). (2.133)
\]

The solution of this differential equation is given by the two-dimensional Green function, i.e.

\[
\phi(\vec{r}) = -(2\pi)^2 \left\{ -\frac{1}{2\pi} \ln \left( \frac{r}{r_{\text{cutoff}}} \right) \right\} = 2\pi \ln \left( \frac{r}{r_{\text{cutoff}}} \right). (2.134)
\]
The orientational correlation function of nematics thus decays algebraically:

\[ g_{n}(r, \varphi) \propto \left( \frac{r}{r_{\text{cutoff}}} \right)^{-n}. \tag{2.135} \]

### 2.6.2 Two-Dimensional Smectics

In contrast to nematics, smectics are not only characterized by orientational correlations but also by positional correlations. Using the Fourier transform of the free energy density, the free energy of a two-dimensional smectic, Eq. (2.84), can be written as

\[ \delta F_{\text{smectic}} = \frac{1}{(2\pi)^2} \int \int \left\{ \frac{1}{2} Bq_x^2 + \frac{1}{2} Kq_y^4 \right\} |\hat{u}(\vec{q})|^2 dq_x dq_y. \tag{2.136} \]

Each degree of freedom is again associated with an energy of \( \frac{1}{2} k_B T \). Thus we have

\[ \langle |\hat{u}(\vec{q})|^2 \rangle = \frac{k_B T}{Bq_x^2 + Kq_y^4}. \tag{2.137} \]

The mean-square fluctuation of the displacement field \( u(x, y) \) is obtained by integrating Eq. (2.137) over the wave-vector space:

\[
\langle u^2(0,0) \rangle = \frac{1}{(2\pi)^2} \int \int \frac{1}{Bq_x^2 + Kq_y^4} \frac{dq_x dq_y}{q_x^2 + \lambda^2 q_y^4} \tag{2.138}
\]

\[
= \frac{1}{(2\pi)^2} \frac{k_B T}{B} \int_{q_{\text{min}}}^{q_{\text{max}}} \int_{q_{\text{min}}}^{q_{\text{max}}} \frac{1}{q_x^2 + \lambda^2 q_y^4} dq_x dq_y \tag{2.139}
\]

\[
= \frac{1}{(2\pi)^2} \frac{k_B T}{B} \int_{q_{\text{min}}}^{q_{\text{max}}} \arctan \left( \frac{q_x}{\lambda q_y^2} \right) dq_y \tag{2.140}
\]

\[
= \frac{1}{(2\pi)^2} \frac{k_B T}{B} \left\{ \lambda \left( \frac{\sqrt{2} \sqrt{q_{\text{min}}}}{\sqrt{2} \sqrt{q_{\text{min}}}} + \lambda + O(q_{\text{min}}) \right) \right\} \tag{2.141}
\]

\[
\approx \frac{
\frac{1}{(2\pi)^2} \frac{k_B T}{B} \left\{ \lambda \left( \frac{\sqrt{2} \sqrt{q_{\text{min}}}}{\sqrt{2} \sqrt{q_{\text{min}}}} + \lambda + O(q_{\text{min}}) \right) \right\} \right\}
\]

\[
\approx \frac{1}{(2\pi)^2} \frac{k_B T}{B} \left( \frac{\sqrt{\lambda}}{\lambda} + \lambda + O(q_{\text{min}}) \right) \tag{2.142}
\]

\[
\approx \frac{1}{(2\pi)^2} \frac{k_B T}{B} \sqrt{\frac{\lambda}{2\pi^2}} \sqrt{\lambda} \tag{2.143}
\]
2.6. ORDER IN TWO DIMENSIONS

The mean-square fluctuation diverges as the square root of the system size $L$.\footnote{A similar although not identical result was obtained by Salditt et al. [70]. Their calculation leads to a divergence linear in the system size. The difference has to be attributed to the fact that in [70] the limit $q_{\text{max}} \to \infty$ is already calculated after the first integration. Here, however, the limit $q_{\text{max}} \to \infty$ is only calculated after the two integrations have been completed.} The correlation function

$$g^{\text{smectic}}(x, y) = \langle e^{i\mathbf{q}_0 \cdot \{u(x,y)-u(0,0)\}} \rangle$$

(2.144)

$$= e^{-\frac{1}{2} q_0^2 \{u(x,y)-u(0,0)^2\}}$$

(2.145)

$$= e^{-\frac{1}{2} q_0^2 \{u^2(x,y)-2u(x,y)u(0,0)+u^2(0,0)\}}$$

(2.146)

$$= e^{-\frac{1}{2} q_0^2 \{u^2(0,0)-u(x,y)u(0,0)\}}$$

(2.147)

is obtained by calculating

$$\langle u^2(0,0) - u(x,y)u(0,0) \rangle = \frac{1}{(2\pi)^2} k_B T \int \int \frac{1 - e^{-i\mathbf{q} \cdot \mathbf{r}}}{Bq_x^2 + Kq_y^4} dq_x dq_y$$

$$= k_B T \left\{ \frac{1}{2\sqrt{\pi}} \frac{1}{\lambda} e^{-\frac{x^2}{4\lambda^2}} + \frac{|y|}{4\lambda} \text{erf} \left( \frac{|y|}{2\sqrt{\lambda|x|}} \right) \right\} ,$$

(2.148)

a result first obtained by Graham \footnote[24]{Details of the calculation can be found in [70].} \footnote[25]{In three-dimensional smectics, the positional correlation function decays algebraically [73].}. The behavior of the correlation function

$$g^{\text{smectic}}(x, y) = e^{-q_0 \frac{k_B T}{2\sqrt{\pi}} \left\{ \frac{1}{2\sqrt{\pi}} \frac{1}{\lambda} e^{-\frac{x^2}{4\lambda^2}} + \frac{|y|}{4\lambda} \text{erf} \left( \frac{|y|}{2\sqrt{\lambda|x|}} \right) \right\} }$$

(2.149)

along the $x$ axis perpendicular to the stripes is given by

$$g^{\text{smectic}}(x, 0) = e^{-\frac{k_B T}{4\pi} \left\{ \frac{1}{2\sqrt{\pi}} \frac{1}{\lambda} e^{-\frac{x^2}{4\lambda^2}} + \frac{|y|}{4\lambda} \text{erf} \left( \frac{|y|}{2\sqrt{\lambda|x|}} \right) \right\} }$$

(2.150)

where $\xi_\perp = 64\pi \left( \frac{B}{k_B T} \right)^2 \lambda^5$.

The decay of the correlation function along the $y$ axis parallel to the stripes reads

$$g^{\text{smectic}}(0, y) = e^{-\frac{K}{4\pi} \left\{ \frac{1}{2\sqrt{\pi}} \frac{1}{\lambda} e^{-\frac{x^2}{4\lambda^2}} + \frac{|y|}{4\lambda} \text{erf} \left( \frac{|y|}{2\sqrt{\lambda|x|}} \right) \right\} }$$

(2.151)

where $\xi_\parallel = 16 \frac{B}{k_B T} \lambda^3$.

The knowledge of the two correlation lengths $\xi_\perp$ and $\xi_\parallel$ allows calculating the values of the elastic constants $B$ and $K$ and thus the penetration depth $\lambda$:

$$\frac{B}{k_B T} = \frac{4}{\pi^3} \frac{\xi_\parallel^3}{\xi_\perp^5} ,$$

(2.152)

$$\frac{K}{k_B T} = \frac{1}{4\pi} \frac{\xi_\perp}{\xi_\parallel} ,$$

(2.153)

$$\lambda = \frac{\pi}{4} \frac{\xi_\parallel^2}{\xi_\perp} .$$

(2.154)
The correlation function of the displacement field \( u(x, y) \) is closely related to the correlation function of the magnetization field \( m(x, y) \):

\[
g_m(x, y) = \frac{q_0}{2\pi} \int_0^{2\pi} \left\langle m(x + x_0, y) \cdot m(x_0, 0) \right\rangle dx_0
\]

\[
= \frac{1}{2} m_s^2 \cdot \cos(q_0 x) \cdot g^{\text{smectic}}(x, y) .
\]

Unlike the mean-square positional fluctuation, the mean-square orientational fluctuation is finite even in infinitely extended systems. As there is no infrared divergence, we extend the integrals to \( q_{\min} = 0 \) and obtain

\[
\left\langle \theta^2(0, 0) \right\rangle = \left\langle \left( \frac{\partial u}{\partial y} \right)^2(0, 0) \right\rangle
\]

\[
= \frac{1}{8\pi^2} \frac{k_B T}{B} \frac{q_{\max}}{\lambda} \left\{ A(q_{\max}, \lambda) + 2 B(q_{\max}, \lambda) + 4 C(q_{\max}, \lambda) \right\}
\]

\[
< \infty .
\]

where we have introduced the functions

\[
A(q_{\max}, \lambda) = \sqrt{\frac{2}{q_{\max} \lambda}} \ln \left( \frac{1 - \sqrt{2q_{\max} \lambda} + q_{\max} \lambda}{1 + \sqrt{2q_{\max} \lambda} + q_{\max} \lambda} \right),
\]

\[
B(q_{\max}, \lambda) = \sqrt{\frac{2}{q_{\max} \lambda}} \left\{ \arctan \left( \sqrt{2q_{\max} \lambda} + 1 \right) + \arctan \left( \sqrt{2q_{\max} \lambda} - 1 \right) \right\},
\]

\[
C(q_{\max}, \lambda) = \arctan \left( \frac{1}{q_{\max} \lambda} \right) .
\]

\[\text{Using Eq. (2.80), Eq. (2.156) is obtained after a trigonometric transformation and averaging over the fluctuations of the displacement field. Putting}
\]

\[
r = q_0 x \quad , \quad s = q_0 x_0 \quad , \quad \alpha = q_0 u(x + x_0, y) \quad \text{and} \quad \beta = q_0 u(x_0, 0) ,
\]

\[
\text{we have}
\]

\[
\left\langle \cos(r + s - \alpha) \cos(s - \beta) \right\rangle
\]

\[
= \left\langle \left\{ \cos(r + s) \cos(\alpha) + \sin(r + s) \sin(\alpha) \right\} \left\{ \cos(s) \cos(\beta) + \sin(s) \sin(\beta) \right\} \right\rangle
\]

\[
= \left\langle \cos(r + s) \cos(s) \cos(\alpha) \cos(\beta) \right\rangle = \cos(r + s) \cos(s) \cos(\alpha) \cos(\beta) .
\]

Finally,

\[
\left\langle \cos(\alpha) \cos(\beta) \right\rangle
\]

\[
= \left\langle e^{i\alpha} e^{-i\beta} + i \cos(\alpha) \sin(\beta) - i \sin(\alpha) \cos(\beta) - \sin(\alpha) \sin(\beta) \right\rangle
\]

\[
= \left\langle e^{i\alpha} e^{-i\beta} \right\rangle = \left\langle e^{iq_0 u(x + x_0, y)} e^{-iq_0 u(x_0, 0)} \right\rangle = \left\langle e^{iq_0 u(x, y)} e^{-iq_0 u(0, 0)} \right\rangle ,
\]

which is the correlation function of the displacement field \( u(x, y) \).
2.6. ORDER IN TWO DIMENSIONS

These functions only depend on the product of $q_{\text{max}}$ and $\lambda$ and return pure numbers. For

$$q_{\text{max}} = \frac{2\pi}{L} = \frac{1}{2\lambda},$$

we have

$$A\left(\frac{1}{2\lambda}, \lambda\right) = -2 \ln(5) \approx -3.22,$$

$$B\left(\frac{1}{2\lambda}, \lambda\right) = 2 \arctan(2) \approx 2.21,$$

$$C\left(\frac{1}{2\lambda}, \lambda\right) = \arctan(2) \approx 1.11.$$ (2.165)

Due to the finite value of the mean-square orientational fluctuations, the orientational order parameter

$$G_{\text{smectic}} = \langle e^{i\theta(x,y)} \rangle = e^{-\frac{1}{2}n^2\langle \theta^2(0,0) \rangle}$$

is finite too. It is given by

$$G_{\text{smectic}} = \exp\left(-\frac{n^2}{16\pi^2} \frac{k_{\text{B}} T q_{\text{max}}}{B \lambda} \{A(q_{\text{max}}, \lambda) + 2B(q_{\text{max}}, \lambda) + 4C(q_{\text{max}}, \lambda)\}\right).$$ (2.169)

The orientational correlation function is related to the mean-square orientational fluctuations in a simple way:

$$g_{\text{smectic}}(x, y) = \langle e^{in(\theta(x,y) - \theta(0,0))} \rangle$$

$$= e^{-\frac{1}{2}n^2\{\langle \theta(x,y) - \theta(0,0) \rangle^2\}}$$

$$= e^{-\frac{1}{2}n^2\{\langle \theta^2(x,y) - 2\langle \theta(x,y) \theta(0,0) \rangle + \langle \theta^2(0,0) \rangle \}}$$

$$= e^{-n^2\langle \theta^2(0,0) \rangle} e^{n^2\langle \theta(x,y) \theta(0,0) \rangle}$$

$$= (G_{\text{smectic}}^n)^2 \cdot e^{n^2\langle \theta(x,y) \theta(0,0) \rangle}.$$ (2.172)

We will show that $\langle \theta(x,y) \cdot \theta(0,0) \rangle$ vanishes over infinite distances. Therefore, the value of the correlation function at infinity is equal to the square of the orientational order parameter $G_{\text{smectic}}$. As the orientational correlation function cannot be calculated analytically, a two-step procedure is chosen. In a first step, the first integral in the calculation of $\langle \theta(x,y) \cdot \theta(0,0) \rangle$ is extended from $q_{\text{max}}$ to infinity. This facilitates the calculation as an integration from $-\infty$ to $\infty$ corresponds to an inverse Fourier transform. In a second step, an approximation of the error introduced in the first step is evaluated. The correction will be incorporated in functions $F_x(x)$.
for the correlation perpendicular to the stripes and $F_y(y)$ for the correlation along the stripe direction respectively. Due to the absence of an infrared divergence, $q_{\text{min}}$ is chosen equal to zero in all the integrals. Along the $x$ axis perpendicular to the stripes, we have

$$\langle \theta(x, 0) \cdot \theta(0, 0) \rangle - F_x(x) = \left\langle \frac{\partial u}{\partial y}(x, 0) \cdot \frac{\partial u}{\partial y}(0, 0) \right\rangle - F_x(x) \quad (2.175)$$

$$= \frac{1}{(2\pi)^2} \frac{k_B T}{B} \int_{q_{\text{max}}}^{q_{\text{min}}} e^{-iq_{\text{max}}x} dq_y \int_{-q_{\text{max}}}^{q_{\text{max}}} \frac{q_y^2 + \lambda^2 q_y^4}{2\lambda} dq_x dq_y \quad (2.176)$$

$$x > 0 = \frac{k_B T}{B} \frac{\text{erf}(q_{\text{max}} \sqrt{\lambda x})}{4\sqrt{\pi \lambda x}} \quad (2.177)$$

where (2.177) is obtained from (2.176) via an inverse Fourier transform. A similar calculation can be done for the correlation along the $y$ axis parallel to the stripes:

$$\langle \theta(0, y) \cdot \theta(0, 0) \rangle - F_y(y) = \left\langle \frac{\partial u}{\partial y}(0, y) \cdot \frac{\partial u}{\partial y}(0, 0) \right\rangle - F_y(y) \quad (2.179)$$

$$= \frac{1}{(2\pi)^2} \frac{k_B T}{B} \int_{q_{\text{max}}}^{q_{\text{min}}} e^{-iq_{\text{max}}y} dq_x \int_{-q_{\text{max}}}^{q_{\text{max}}} \frac{q_x^2 + \lambda^2 q_x^4}{8\lambda^2 \alpha} dq_x dq_y \quad (2.180)$$

$$= \frac{1}{2\pi} \frac{k_B T}{B} \sum_{\alpha} \frac{i e^{i \alpha y} (1 + \text{sgn}(\text{Im}(\alpha)))}{2\pi \lambda y} \quad (2.181)$$

$$= \frac{k_B T}{B} \frac{\sin\left(\sqrt{\frac{q_{\text{max}}^2}{2\lambda}} y\right)}{2\pi \lambda y} e^{-\sqrt{\frac{q_{\text{max}}^2}{\lambda}} y} \quad , \quad (2.182)$$

where Eq. (2.181) is again obtained from Eq. (2.180) via an inverse Fourier transform. The sum in Eq. (2.181) runs over the roots $\alpha$ of

$$q_x^2 + \lambda^2 q_x^4 = 0 \quad . \quad (2.183)$$

In order to obtain the corrections, we have to calculate the integrals from $q_{\text{max}}$ to infinity. After an integration by parts, we will make use of the fact that for large distances $r$ the second term obtained in the integration will be sufficiently small to be neglected.\(^{27}\)

$$\int_{-q_{\text{max}}}^{q_{\text{max}}} f(q) e^{-iqr} dq + \int_{q_{\text{max}}}^{\infty} f(q) e^{-iqr} dq = 2 \int_{q_{\text{max}}}^{\infty} f(q) \cos(qr) \ dq$$

$$= 2 f(q) \frac{\sin(qr)}{r} \bigg|_{q_{\text{max}}}^{\infty} - 2 \int_{q_{\text{max}}}^{\infty} \frac{f'(q)}{r} \ dq \approx -2 f(q_{\text{max}}) \frac{\sin(q_{\text{max}} r)}{r} . \quad (2.184)$$

\(^{27}\)A similar approach is used in the calculation of the RKKY interaction.
The finite cutoff $q_{\text{max}}$ leads to an oscillatory dependence of the correction on the distance $r$.

Along the $x$ axis perpendicular to the stripes, we have

$$\mathcal{F}_x(x) = \frac{-1}{(2\pi)^2} \frac{k_B T}{B} \int_{-q_{\text{max}}}^{q_{\text{max}}} \left\{ \int_{-\infty}^{-q_{\text{max}}} \frac{q_y^2}{q_x^2 + \lambda^2 q_y^2} e^{-iq_y x} dq_y + \int_{q_{\text{max}}}^{\infty} \frac{q_y^2}{q_x^2 + \lambda^2 q_y^2} e^{-iq_y x} dq_y \right\} dq_x \approx \frac{1}{(2\pi)^2} \frac{k_B T}{B} \int_{-q_{\text{max}}}^{q_{\text{max}}} 2 \frac{q_y^2}{q_{\text{max}}^2 + \lambda^2 q_y^2} \frac{\sin(q_{\text{max}} x)}{x} dq_y \approx \frac{k_B T}{B} \frac{\sin(q_{\text{max}} x)}{8\pi^2 \lambda x} \left\{ A(q_{\text{max}}, \lambda) + 2 B(q_{\text{max}}, \lambda) \right\}, \quad (2.185)$$

and along the $y$ axis parallel to the stripes we have

$$\mathcal{F}_y(y) = \frac{-1}{(2\pi)^2} \frac{k_B T}{B} \int_{-q_{\text{max}}}^{q_{\text{max}}} \left\{ \int_{-\infty}^{-q_{\text{max}}} \frac{q_y^2}{q_x^2 + \lambda^2 q_y^2} e^{-iq_y y} dq_y + \int_{q_{\text{max}}}^{\infty} \frac{q_y^2}{q_x^2 + \lambda^2 q_y^2} e^{-iq_y y} dq_y \right\} dq_x \approx \frac{1}{(2\pi)^2} \frac{k_B T}{B} \int_{-q_{\text{max}}}^{q_{\text{max}}} 2 \frac{q_{\text{max}}^2}{q_x^2 + \lambda^2 q_{\text{max}}^4} \frac{\sin(q_{\text{max}} y)}{y} dq_x \approx \frac{k_B T}{B} \frac{\sin(q_{\text{max}} y)}{\pi^2 \lambda y} \left\{ C(q_{\text{max}}, \lambda) \right\}. \quad (2.186)$$

In both cases the correction is inversely proportional to the distance. For large distances, an expansion of the exponential in Eq. (2.174) to lowest orders exhibits the algebraic decay of the orientational correlation function. The correlation function along the $x$ axis perpendicular to the stripes is dominated by a non-oscillatory term with an exponent of $-\frac{1}{2}$:

$$g_{\text{smectic}}(x, 0) \overset{x \to \infty}{\approx} \left( C_{\text{n}}^{\text{smectic}} \right)^2 \cdot \left\{ 1 + \sqrt{\frac{\xi_{n, \perp}}{x}} + \frac{\zeta_{n, \perp}}{x} \cdot \sin(q_{\text{max}} x) \right\}, \quad (2.187)$$

where

$$\xi_{n, \perp} = \frac{n^4}{16\pi} \left( \frac{k_B T}{B} \right)^2 \frac{1}{\lambda^3}, \quad (2.188)$$

$$\zeta_{n, \perp} = \frac{n^2}{8\pi^2} \frac{k_B T}{B} \frac{1}{\lambda} \left\{ A(q_{\text{max}}, \lambda) + 2 B(q_{\text{max}}, \lambda) \right\}. \quad (2.189)$$

The correlation along the $y$ axis parallel to the stripes is to first order given by the term due to the correction function $\mathcal{F}_y(y)$:

$$g_{\text{smectic}}(0, y) \overset{y \to \infty}{\approx} \left( C_{\text{n}}^{\text{smectic}} \right)^2 \cdot \left\{ 1 + \frac{\zeta_{n, \parallel}}{y} \cdot \sin(q_{\text{max}} y) \right\}, \quad (2.190)$$
where
\[ \zeta_{n\parallel} = \frac{n^2}{\pi^2} \frac{k_B T}{B} \frac{1}{\lambda} C(q_{\text{max}}, \lambda) \quad . \quad (2.191) \]

As in the case of the positional correlation function, we can use the correlation lengths to extract the elastic moduli \( K \) and \( B \) and hence the penetration depth \( \lambda \):
\[ \frac{B}{k_B T} = \frac{16n^2}{\pi^5} C^6(q_{\text{max}}, \lambda) \frac{\xi_{n\perp}}{\zeta_{n\parallel}} \quad , \quad (2.192) \]
\[ \frac{K}{k_B T} = \frac{n^2}{16} \frac{C^2(q_{\text{max}}, \lambda)}{\pi^3} \frac{\zeta_{n\parallel}}{\xi_{n\perp}} \quad , \quad (2.193) \]
\[ \lambda = \frac{\pi^3}{16} \frac{1}{C^2(q_{\text{max}}, \lambda)} \frac{\zeta_{n\parallel}^2}{\zeta_{n\perp}} \quad . \quad (2.194) \]

### 2.6.3 Two-Dimensional Smectics with Anisotropy

The positional correlation function of smectics, Eq. (2.149), decays exponentially. If an additional anisotropy energy for the orientation of the domain walls is assumed, however, the positional correlation function decays algebraically, changing the positional order from short-range to quasi-long-range. An additional anisotropy energy could be due to oriented steps on the surface of a substrate but such an anisotropy energy is also an intrinsic property of ultrathin iron films on Cu(100), as has been shown by Kashuba and Pokrovsky [74]. They find that higher-order gradient terms of the exchange energy yield a four-fold anisotropy for the orientation of the domain walls. According to their analysis, stripe domains in ultrathin iron films on Cu(100) should be aligned along a \( \langle 100 \rangle \) direction. This has indeed been observed in experiments by Allenspach and Bischof [5] and is also supported by our experiments.

Following Kashuba and Pokrovsky [74], we introduce a new term into the free energy of smectics, Eq. (2.84):
\[ \delta F = \int \int \left\{ \frac{1}{2} B \left( \frac{\partial u}{\partial x} \right)^2 + \frac{1}{2} \nu \left( \frac{\partial u}{\partial y} \right)^2 + \frac{1}{2} K \left( \frac{\partial^2 u}{\partial y^2} \right)^2 \right\} \, dx \, dy \quad . \quad (2.195) \]

Due to the equipartition theorem, each degree of freedom is associated with an energy of \( \frac{1}{2} k_B T \), hence
\[ \langle |\hat{u}(\vec{q})|^2 \rangle = \frac{k_B T}{Bq_x^2 + \nu q_y^2 + Kq_y^4} \quad . \quad (2.196) \]

We now introduce a dimensionless constant \( \kappa \) via
\[ \kappa = \sqrt{\frac{B}{\nu}} \quad . \quad (2.197) \]
Looking at the scaling behavior of the terms \( \left( \frac{\partial u}{\partial y} \right)^2 \) and \( \left( \frac{\partial^2 u}{\partial y^2} \right) \) in Eq. (2.195) respectively, we see that beyond a distance of

\[
r_0 = \sqrt{\frac{K}{\nu}}
\]

(2.198)

the term associated with bending, i.e. the term proportional to \( K \), becomes negligible with respect to the anisotropy term. As anisotropy will only become important for large distances, we will henceforth concentrate on contributions to the correlation function at large distances and neglect the bending term.

The mean-square positional fluctuation of smectics with anisotropy diverges logarithmically with system size \( L_{\text{system}} \):

\[
\langle u^2(0,0) \rangle = \frac{1}{(2\pi)^2} k_B T \int \int \frac{1}{Bq_x^2 + \nu q_y^2} dq_x dq_y = \frac{1}{2\pi} \frac{k_B T}{B} \kappa \ln \left( \frac{L_{\text{system}}}{L} \right).
\]

(2.199)

In order to obtain the positional correlation function, we have to calculate

\[
\langle u^2(0,0) - u(x,y)u(0,0) \rangle \approx \frac{1}{2\pi} \frac{k_B T}{B} \kappa \ln \left( \frac{\sqrt{x^2 + \kappa^2 y^2}}{L\sqrt{1 + \kappa^2}} \right).
\]

(2.200)

The calculation is analogous to the calculation from Eq. (2.126) to Eq. (2.131) for the orientational correlation function of nematics. The resulting positional correlation function

\[
g_{\text{sm.an.}}(x,y) \approx \left( \frac{\sqrt{x^2 + \kappa^2 y^2}}{L\sqrt{1 + \kappa^2}} \right)^{\frac{3}{2} \nu \frac{k_B T}{B} \kappa}.
\]

(2.201)

decays algebraically, which is tantamount to quasi-long-range positional order. Note that for weak anisotropies \( \kappa \) will be large. Therefore, the correlation along the \( x \) axis perpendicular to the stripes will exceed the correlation along the \( y \) axis parallel to the stripe direction.

The mean-square orientational fluctuations exhibit no infrared divergence. Extending the integrals from \( q_{\min} \) to zero, we obtain

\[
\langle \theta^2(0,0) \rangle = \frac{1}{(2\pi)^2 \nu} \int_{-q_{\max}}^{q_{\max}} \int_{-q_{\max}}^{q_{\max}} \frac{q_y^2}{\kappa^2 q_x^2 + q_y^2} dq_x dq_y
\]

\[
= \frac{1}{2\pi^2 \nu} q_{\max}^2 \left\{ 1 + \frac{1}{\kappa} \arctan(\kappa) - \kappa \arctan \left( \frac{1}{\kappa} \right) \right\}.
\]

(2.202)

Consequently, the orientational order parameter is given by

\[
G_{\text{n.sm.an.}} = \exp \left( -\frac{n^2 k_B T}{4\pi^2 \nu} q_{\max}^2 \left\{ 1 + \frac{1}{\kappa} \arctan(\kappa) - \kappa \arctan \left( \frac{1}{\kappa} \right) \right\} \right).
\]

(2.204)
As in the case of smectics without anisotropy, the orientational correlation function of smectics with anisotropy cannot be calculated analytically. We will therefore adopt the procedure described in Subsec. 2.6.2. Along the $x$ axis, we have

$$\langle \theta(x,0) \cdot \theta(0,0) \rangle - G_x(x) = \left\langle \frac{\partial u}{\partial y}(x,0) \cdot \frac{\partial u}{\partial y}(0,0) \right\rangle - G_x(x) \quad (2.205)$$

$$= \frac{1}{(2\pi)^2} \frac{k_B T}{\nu} \int_{-q_{\text{max}}}^{q_{\text{max}}} \int_{-\infty}^{\infty} \frac{q_x^2 e^{-iq_x x}}{\kappa^2 q_x^2 + q_y^2} dq_x dq_y \quad (2.206)$$

$$= \frac{1}{(2\pi)^2} \frac{k_B T}{\nu} \int_{-q_{\text{max}}}^{q_{\text{max}}} \frac{q_y}{\kappa} e^{-\frac{q_y^2}{\kappa}} dq_y \quad (2.207)$$

$$= \frac{k_B T}{\nu} \frac{\kappa}{\pi} \left\{ 1 - \left\{ 1 + \frac{q_{\text{max}} x}{\kappa} \right\} e^{-\frac{q_{\text{max}} x}{\kappa}} \right\} \quad (2.208)$$

and along the $y$ axis we have

$$\langle \theta(0,y) \cdot \theta(0,0) \rangle - G_y(y) = \left\langle \frac{\partial u}{\partial y}(0,y) \cdot \frac{\partial u}{\partial y}(0,0) \right\rangle - G_y(y) \quad (2.209)$$

$$= \frac{1}{(2\pi)^2} \frac{k_B T}{\nu} \int_{-q_{\text{max}}}^{q_{\text{max}}} \int_{-\infty}^{\infty} \frac{q_y^2 e^{-iq_y y}}{\kappa^2 q_x^2 + q_y^2} dq_x dq_y \quad (2.210)$$

$$= \frac{1}{(2\pi)^2} \frac{k_B T}{\nu} \int_{-q_{\text{max}}}^{q_{\text{max}}} \frac{-\kappa q_x e^{-\kappa q_y y}}{q_y} dq_x \quad (2.211)$$

$$= \frac{k_B T}{\nu} \frac{1}{\kappa \pi} \left\{ -1 + \left\{ 1 + \frac{q_{\text{max}} y}{\kappa} \right\} e^{-\kappa q_{\text{max}} y} \right\} \quad (2.212)$$

The correction for the correlation function along the $x$ axis is

$$G_x(x) = \frac{k_B T}{\nu} \frac{\sin(q_{\text{max}} x)}{\pi^2 x} q_{\text{max}} \left\{ 1 - \kappa \arctan \left( \frac{1}{\kappa} \right) \right\} \quad , \quad (2.213)$$

and the correction for the correlation function along the $y$ axis is

$$G_y(y) = \frac{k_B T}{\nu} \frac{\sin(q_{\text{max}} y)}{\pi^2 y} \frac{\arctan(\kappa)}{\kappa} \quad . \quad (2.214)$$

For large distances, the orientational correlation function along the $x$ axis is therefore given by

$$g_{n,\text{sm.an.}}(x,0) \approx (G_{n,\text{sm.an.}}^2)^2 \left\{ 1 + \frac{\zeta_{n,\perp}}{x} \sin(q_{\text{max}} x) + \frac{\xi_{n,\perp}^2}{x^2} \right\} \quad , \quad (2.215)$$

where

$$\zeta_{n,\perp} = \frac{n^2}{\pi^2} \frac{k_B T}{\nu} q_{\text{max}} \left\{ 1 - \kappa \arctan \left( \frac{1}{\kappa} \right) \right\} \quad , \quad (2.216)$$

$$\xi_{n,\perp} = \frac{n}{\sqrt{\pi}} \sqrt{\frac{k_B T}{\nu} \sqrt{\kappa}} \quad . \quad (2.217)$$
Along the $y$ axis, the orientational correlation function is given by

$$g_{n}^{\text{sm.an.}}(0, y) \approx (G_{n}^{\text{sm.an.}})^{2} \cdot \left\{ 1 + \frac{\zeta_{n}}{y} \sin(q_{\text{max}} y) - \frac{\xi_{n}}{y^{2}} \right\},$$

(2.218)

where

$$\zeta_{n} = \frac{n^{2} k_{B} T}{\pi^{2} \nu} q_{\text{max}} \frac{\arctan(\kappa)}{\kappa},$$

(2.219)

$$\xi_{n} = \frac{n}{\sqrt{\pi}} \sqrt{\frac{k_{B} T}{\nu}} \frac{1}{\sqrt{\kappa}}.$$

(2.220)

### 2.7 Topological Disordering

The route taken by two-dimensional ordered media on their way to their high-temperature isotropic state has been the subject of numerous studies. While some models emphasize the role of defects and suggest a multi-step process with distinct intermediate phases and second-order transitions\[43, 75–79\], other studies predict a direct first-order transition\[10, 80–85\]. In many cases, the disordering scenario is sensitive to the energies associated with the defects, and without knowledge of their precise values, predictions of the order of the transition are often inconclusive. A number of computer simulations have been carried out to shed some light on the disordering process and evidence for either scenario has been found\[81, 86–94\]. An overview of the diverse models is given in\[13\], with a special emphasis on computer simulations.

#### 2.7.1 Models for a Second-Order Transition

Mizushima\[95, 96\] has put forward a theory of melting in three-dimensional crystals based on dislocations. According to this theory, the entropy of the system rises faster than the energy when the density of dislocations is increased. Consequently, there will be a temperature above which the dislocations rare at low temperatures will start to proliferate and initiate the melting process. Kosterlitz and Thouless\[75\] have developed an analogous theory for two-dimensional systems. It applies to a range of systems where individual defects have an energy that grows with the logarithm of the system size. As the entropy is proportional to the logarithm of the defect density, there will be a well-defined transition temperature. Below the transition temperature, the defects are expected to be bound in pairs of oppositely

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28 This dislocation-based melting procedure of three-dimensional crystals represents a first-order transition.
‘charged’ defects. Such pairs will have a finite energy. Above the transition temperature, however, the defect pairs are expected to unbind.

The systems analyzed by Kosterlitz and Thouless comprise the two-dimensional $x$-$y$ model of magnetism, the two-dimensional crystal with a square lattice, and the two-dimensional neutral superfluid. The defects are magnetic vortices (disclinations), dislocations and superfluid vortices respectively. The analysis has been extended to two-dimensional crystals with a triangular lattice by Halperin and Nelson \[76, 78\], and Young \[77\]. Nelson and Halperin find that the melting of such a system could proceed in two steps, the defects mediating the first transition being dislocations and the second transition being mediated by disclinations. At low temperatures, the solid has quasi-long-range positional order and long-range orientational order. After the dislocation unbinding process, the system is believed to be in a state with short-range positional order and quasi-long-range orientational order. This new phase is called the hexatic state. The dissociation of disclination pairs in the second transition is then believed to result in the isotropic state.

Applying the same principles to layered liquid-crystal films, Toner and Nelson \[79\] find that the first transition mediated by dislocations should occur at zero Kelvin. This is due to the finite energy of dislocations in such systems. Accordingly, Toner and Nelson expect individual dislocations to be present at any finite temperature and the type of order at sufficiently large distances to be that of a nematic. The transition to the isotropic state is thought to be mediated by disclinations. Each single disclination has an energy proportional to the logarithm of the system size, but a pair of oppositely charged disclinations has a finite energy. The isotropic state is reached after an unbinding process of disclination pairs. The smectic state is only predicted for zero Kelvin or patches of limited extent.

Considering the stripe domains in ultrathin ferromagnetic films with perpendicular anisotropy as equivalent to the layers of a two-dimensional smectic, Abanov, Kalatsky, Pokrovsky, and Saslow \[43\] elaborated phase diagrams for such magnetic systems. An additional ingredient as compared to the smectic studied by Toner and Nelson \[79\] arises from fourth-order gradient terms in the exchange energy. They lead to a four-fold stripe-orientation anisotropy \[74\]. For small values of the anisotropy, it is assumed that finite-energy dislocations can cause exponentially decaying order. Larger values of the anisotropy, however, make individual dislocations have infinite energies. In this case, the system is characterized by quasi-long-range positional order and long-range orientational order. It is predicted to undergo a Kosterlitz-Thouless like transition to an Ising nematic state with short-range positional order. The orientational order of the Ising nematic phase is assumed to be lost in a subsequent Ising-like transition mediated by so-called rotation domain walls. The resulting state will have domain sections along any of the two easy axes of the stripe-orientation anisotropy and is therefore named a tetragonal liquid. Note that this scenario depends on the precise value of elastic constants. A direct first-order
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transition from the smectic to the tetragonal liquid is not excluded.

2.7.2 Models for a First-Order Transition

Several models indicating a first-order transition have been proposed. They are based on various arguments and accordingly predict a number of different routes towards the isotropic state.

Brazovskii investigated nearly isotropic systems characterized by a spectrum of fluctuations with a minimum at a nonzero wave vector $q_0$. A low minimum of the spectrum will lead to strong fluctuations, which, eventually, will establish the isotropic state. The transition is predicted to be of first order. Such considerations were applied to magnetic films with perpendicular anisotropy by Garel and Doniach [10]. The free energy of Eq. (2.76) reaches its minimum at $q = q_0$ and should thus allow a first-order transition driven by fluctuations. The Ginzburg-Landau mean-field transition would be suppressed. However, Garel and Doniach raise some doubts as to the applicability of the first-order perturbation theory by Brazovskii to systems without long-range order.

Neglecting fluctuations, Ramakrishnan [82] elaborated a density-wave theory of melting for two-dimensional crystals. Due to the presence of a third-order term in the expansion of the free energy, the transition is first order. Note that such a behavior is found for hexagonal lattices but not for square lattices.

A model for a first-order transition based on defects has been proposed by Chui [83, 84]. The type of defects considered by Chui is grain boundaries, which may be viewed as a string of dislocations. Chui finds that as the temperature is increased grain boundaries are generated before dislocations unbind. A first-order transition results when the core energy of the dislocations is small enough. Another reason for a first-order transition would be a coupling to a finite density change. The formation of grain boundaries is accompanied by a strong increase of the dislocation density. In contrast, the second-order transitions of the KTHNY (Kosterlitz-Thouless-Halperin-Nelson-Young) type are characterized by a constant density of dislocations around the transition temperature.

In the KTHNY scenario the dislocation interactions in the solid phase are treated independently of the disclination interactions in the hexatic phase. If the disclination interaction is sufficiently screened as the dislocations unbind, a simultaneous unbinding of dislocations and disclinations is conceivable. Such a theory has been put forward by Kleinert [85]. Kleinert argues that the core energy of dislocations as-

\[29\] Previously, Fisher, Halperin, and Morf had pointed out that the energy required for the formation of a single grain boundary vanishes as the dislocation-unbinding transition temperature is approached [97]. However, they predicted that the sequence of phases occurring during a transition process due to grain boundaries would be identical to the Kosterlitz-Thouless-Halperin-Nelson-Young scenario with an intermediate hexatic phase. Chui’s analysis is more detailed.
sumed by Halperin and Nelson [76] is unphysically high. More realistic values would enable the formation of new configurations and lead to a first-order transition.

2.7.3 Simulations

Systems with competing long-range and short-range interactions have been investigated in various Monte Carlo simulations. Hurley and Singer [86] examined the melting of a two-dimensional dipolar lattice gas on a triangular lattice. At half filling, the low-temperature phase is found to be given by oriented stripe domains. As the temperature is increased, the overall orientational order is lost. Locally, however, the system is still characterized by homogeneous stripe segments. It is only at a higher temperature that this local order is found to disappear. A similar sequence of phases was observed in simulations by Booth et al. [87]. They investigated the properties of a uniaxial spin system on a square lattice supplemented with a long-range dipole-dipole interaction. The low-temperature stripe configuration is again found to undergo a transition to a state with no long-range orientational order but locally still well-defined domains. Due to the symmetry of the underlying square lattice, Booth et al. call the resultant state with four-fold symmetry the tetragonal phase. The transition to the tetragonal phase is marked by a relatively sharp peak in the specific heat but the behavior of the orientational order parameter suggests a continuous phase transition. A weak first-order transition, however, is not ruled out on the basis of the Monte Carlo simulations. At still higher temperatures, the tetragonal phase decays into a fully disordered, paramagnetic state. The transition between the tetragonal and the paramagnetic phase is gradual; the corresponding peak in the specific heat is finite and broad. The individual phases are found to be reflected in the structure factor. The stripes are characterized by maxima along a line through the origin; the maxima of the tetragonal phase are located at the corners of a square centered at the origin. With increasing temperature, the structure factor of the tetragonal phase gradually transforms into an isotropic ring which is assumed to characterize the paramagnetic phase. The effect of a magnetic field on the domain pattern was investigated by Arlett et al. [88]. The transition line between the stripe phase and the tetragonal phase is observed to decrease with increasing field strengths. Some evidence for a first-order transition is found for sufficiently high fields in contrast to the continuous, or at most, very weak first-order transition found at zero field. The prediction as to the occurrence of a tetragonal phase at sufficiently high fields is at variance with the prediction by Garel and Doniach [10] about a bubble phase. Such a bubble phase is, however, supported by the simulations of Hurley and Singer [86] of the two-dimensional dipolar lattice gas. Sampaio, de Albuquerque, and de Menezes [89] used Monte Carlo simulations to track the magnetic relaxation process of a classical two-dimensional Ising model with
long-range dipolar interaction after the application of a magnetic field. Depending on the ratio of the strengths of dipolar and exchange interactions, two scenarios can be distinguished. For weak dipolar interactions, the magnetization decays exponentially when the magnetic field is switched off. The relaxation process is initiated by the nucleation of a small number of oppositely magnetized domains which then grow rapidly to reduce the magnetization. Stronger dipolar interactions lead to the simultaneous nucleation of a large number of oppositely magnetized domains. In such a case, the magnetization decays algebraically when the magnetic field is switched off. The sequence of domain patterns observed during the relaxation process is reminiscent of that found by Booth et al. [87] upon lowering the temperature in their uniaxial spin system on a square lattice with dipole-dipole interaction.

Lyuksyutov and Bauer [90] used Monte Carlo simulations to study the effect of nonuniformities in the film structure. The island structure of films is incorporated into their model by means of two interpenetrating sublattices. One sublattice has a fixed perpendicular anisotropy whereas the other sublattice allows some in-plane oriented spins. These in-plane spins represent a perturbation of the system. But the authors also investigated the influence of the relative strength of dipolar and exchange interaction on the magnetic domain pattern. It turned out that the sequence of phases that evolves as the strength of the dipolar interaction is increased again resembles that found by Booth et al. [87] upon cooling down from high temperatures.

In order to minimize effects due to the symmetry of the discrete underlying lattice, Stoycheva and Singer [92, 93] performed Monte Carlo simulations for a model with nearest-neighbor attractive interactions and long-range repulsive dipolar interactions on a triangular lattice. Due to the long-range nature of the dipolar interaction and the increase of the stripe width with decreasing strength of the dipolar interaction, only systems with relatively strong dipolar interactions are accessible to Monte Carlo simulations. For strong dipolar interactions, Stoycheva and Singer find a transition from the stripe phase at low temperatures to a phase without long-range orientational order at higher temperature. Locally, well-defined stripe segments are still present above the transition temperature. However, disclinations destroy the long-range order found in the stripe phase. This is seen as evidence supporting a defect-mediated melting involving an unbinding of dislocations into disclinations. Direct images showing the disordering process in a step-by-step manner are not provided. The transition temperature is predicted to increase to the critical temperature of the Ising model as the strength of the dipolar interaction is reduced. For weak dipolar interactions, the defect-mediated melting is expected to be replaced by an Ising-like disordering of the spins within the domains. Note that the transition to this regime is already predicted for ratios between the dipolar and the

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\[98, 99\]

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nearest-neighbor attractive interaction orders of magnitude larger than those found in ultrathin Fe films on Cu(001). The sequence of phases predicted for a variation of the relative strengths of repulsive dipolar and attractive nearest-neighbor interactions is just the opposite of what Lyuksyutov and Bauer [90] have found in their Monte Carlo simulations.

An alternative model for a system with competing interactions has been put forward by Iglesias et al. [94]. It is based on a three-state spin-1 Hamiltonian equipped with a long-range RKKY-like potential and produces a sequence of domain patterns already familiar from the other Monte Carlo simulations. However, due to the special form of the long-range repulsive interaction, the applicability of these results to ultrathin Fe films on Cu(001) is questionable.

In contrast to previous Monte Carlo simulations, simulations by Cannas et al. [81] suggest a weak first-order transition between the stripe phase and a tetragonal phase. The nature of the transition is concluded from the energy distribution of the spins which, in the transition region, shows two distinct peaks. The lower-energy peak is associated with the stripe phase, and the higher-energy peak with the tetragonal phase. The authors support their numerical findings with an analytic calculation based on the ideas of Brazowskii [80]. However, the first-order nature of the transition disappears gradually as the relative strength of the dipolar energy is reduced. The first-order transition is only present in systems with very strong dipolar interactions.
Chapter 3

Experiment

The system of ultrathin Fe films on Cu(001) is extremely rich in its structural and magnetic properties and has been the subject of a huge number of theoretical and experimental studies. To quote Asada and Blügel \[100\]: ‘A few atomic layers of Fe on Cu(100) make up the single most complex and complicated ultrathin magnetic system of all.’ Although many studies have been concerned with the magnetic properties of ultrathin Fe films, only few have focused on the magnetic domain pattern \[5, 14, 19\]. Much more attention has been directed towards the magnetic moments in the individual layers and the type of ordering between the different layers of a film.

In this study, the focus is on the various domain patterns that occur in this system. The magnetic domain patterns are imaged using a Scanning Electron Microscope with Polarization Analysis (SEMPA). Due to its high surface sensitivity and its high lateral resolution, SEMPA is particularly well suited for investigating magnetic domain patterns in ultrathin films and the transitions between them.

The magnetic domain patterns turn out to depend on the preparation conditions, which affect the structural properties of the films. Low-Energy Electron Diffraction (LEED) allows determining the interlayer distance in the films. This information is used to set the films examined in this study in the context of previous studies, both experimental and theoretical.

The magnetic domain patterns, however, are also a function of film thickness and temperature. In order to determine their influence, the temperature of the samples is varied, and films are grown as wedges. Wedges provide a simple way to compare the magnetic domain patterns at different thicknesses. Due to the uniform preparation conditions throughout a wedge, varying preparation conditions can be ruled out as a cause for different domain patterns at different thicknesses.

An important factor influencing the magnetic domain pattern at a given thickness and a given temperature is the value of the Curie temperature. Its dependence on film thickness has already been the subject of previous studies. In the thickness
range investigated in this study, the Curie temperature increases monotonously with film thickness. The same sequence of phases as upon increasing the temperature can be observed on a wedge on the way to smaller thicknesses. This suggests the introduction of an effective temperature

\[ \tau = T/T_c \]  

where \( T_c \) is the Curie temperature given by the film thickness. The concept of an effective temperature allows a unified description of the sequence of phases observed upon varying the actual temperature or moving along a wedge.

The various phases are characterized by their symmetry, but more specifically also by their positional and orientational correlations. These correlations are calculated for each phase. Some of these correlations can be described by the formulae developed in Subsec. 2.6.2. But in many cases, an adequate model has still to be found. The experimentally determined correlations can serve as a testing ground for future theoretical models.

The mechanisms mediating the transitions between the different phases are identified by examining the evolution of the domain patterns in successive SEMPA images. SEMPA images are also used to verify the mean-field predictions of Subsec. 2.3.3 concerning the temperature dependence of the stripe width near the transition to the paramagnetic state. The knowledge of the variation of the domain width is instrumental to understanding the various transitions between the different phases.
3.1 Sample Preparation

The samples used in this experiment have been prepared under UHV conditions\(^1\) and consist of ultrathin Fe films evaporated onto a single-crystalline Cu(001) substrate. The preparation of the substrate involves sputtering and annealing. The subsequent evaporation of the Fe film is done by Molecular Beam Epitaxy. The delay time between annealing and evaporation defines the substrate temperature during evaporation.

3.1.1 Sputtering

Before a new Fe film is evaporated, the Cu(001) substrate is cleaned thoroughly. Usually, a bombardment with argon ions for 30 min at a pressure of \(2.5 \cdot 10^{-5}\) mbar and a voltage of 1 kV is sufficient to remove previous Fe films and contaminations. The effectiveness of the sputtering procedure is checked with Auger Electron Spectroscopy (AES). However, in order to minimize contamination, no analysis with AES is made when the substrate is to be used for a new sample.

3.1.2 Annealing

The bombardment with argon ions in the sputtering process results in a clean but rough surface of the substrate. Subsequent thermal treatment reestablishes a smooth Cu(001) surface. In the annealing procedure, the Cu(001) substrate crystal is heated to 700 K for 30 min. The effect of annealing is apparent from the quality of Low-Energy Electron Diffraction (LEED) patterns before and after annealing. In order to prevent contamination, no LEED analysis is made when the substrate is to be used for a new sample.

3.1.3 Delay

The temperature of the substrate during the evaporation of an Fe film is decisive for the magnetic domain pattern that develops in the film. The temperature is varied by adjusting the delay time after the annealing process. The temperature dependence of the substrate as a function of the delay time after the annealing process is given in Fig. 3.1. Note that the temperatures were measured with the evaporation source switched off. Radiation from the evaporation source may increase the temperature of the substrate during the evaporation process.

\(^1\)The base pressure in the preparation chamber is below \(8 \cdot 10^{-11}\) mbar.
After annealing, the Cu(100) substrate has a temperature significantly higher than room temperature. The temperature of the substrate during the evaporation process can be adjusted by selecting the appropriate delay time. The temperature curve was measured on the evaporation stage while the evaporation source was off. The temperature decays exponentially as indicated by the fit.

### 3.1.4 Molecular Beam Epitaxy

The Fe films are deposited by means of Molecular Beam Epitaxy. A sketch of the evaporation stage is given in Fig. 3.2. The parameters for heating the Fe wires in the evaporation source are chosen such as to obtain a deposition rate of 0.09 ML/min. In order to make sure that the evaporation rate is stable when the shutter is opened, the heating of the Fe wires is started 20 min prior to the actual evaporation process. At first, the voltage is set to 1000 kV and the emission current is set to somewhat below the required current (about 65 mA). After 5 min, the current is permanently adjusted to the required current as determined in previous experiments (about 77.2 mA). About 5 min before the shutter is opened, the current has normally reached a stable state.
3.2 Types of Samples

The samples investigated in this experiment are of two types: homogeneously thick films and wedges. Wedges provide an overview of the magnetic phases as a function of film thickness. The combined use of Scanning Electron Microscopy with Polarization Analysis (SEMPA) and AES allows assigning magnetic properties to films of a certain thickness. However, structural investigations by means of LEED measure the properties of areas with a diameter on the order of 1.5 mm. In such a case, homogeneously thick films have to be used. Moreover, the use of homogeneously thick films is required to rule out effects due to the shape of the wedge. Due to the long-range dipolar interaction, such effects cannot be ruled out a priori. Images of extended homogeneously thick films are also needed for a statistical analysis of the magnetic domain patterns.

3.2.1 Wedges

Wedges are created by moving a mask in front of the substrate during the evaporation process, cf. Fig. 3.3. The sooner a given area is covered by the shutter, the smaller the local thickness of the Fe film. Although a cross section of such an Fe film represents a wedge, the evaporation rate is the same throughout the wedge and deposition starts everywhere at the same time. The only difference between different spots on the wedge is the local thickness of the film. For practical purposes, the mask is only moved after about 1–2 ML of Fe have been deposited on the substrate. The beginning of a wedge the thickness of which increases continuously from zero is hardly visible in the Scanning Electron Microscope (SEM); a step of 1–2 ML, however, can be identified easily. This is important for
Homogeneously thick samples and wedges.

A mask placed between the evaporation source and the substrate allows producing laterally confined samples of homogeneous thickness. If the mask is continuously shifted during the evaporation process, the film will have a wedge-shaped thickness profile.

establishing the correlation between magnetic properties and film thickness.

A cross section of a typical wedge as determined by means of AES is depicted in Fig. 3.4. The step and the wedge just described can be seen on the left hand side. Adjacent to the wedge, there is a small area of constant thickness, which permits an accurate determination of both maximum thickness and evaporation rate.

3.2.2 Homogeneously Thick Films

If the mask in front of the substrate is not moved during the evaporation process, a homogeneously thick film will be created. Under a number of circumstances, however, it will be more useful to modify a wedge in such a way that the flat area extends almost throughout the whole sample. The small wedge at the edge can then be used to image the magnetic properties of slightly thinner films. This is especially beneficial if the targeted thickness range is very small.

3.3 Types of Substrates

All the experiments reported in Part I of this thesis have been carried out on ultrathin Fe films deposited onto either flat Cu(001) substrates or a slightly miscut Cu(001) substrate. The miscut angle of the miscut substrate is 3°. The miscut results in a dense array of parallel steps that run along one of the \langle 110 \rangle directions. While the overwhelming majority of the experiments on flat substrates were done on substrates with a corrugation of less than 30 nm, some experiments were also performed with films deposited onto a less perfect Cu(001) substrate. Wherever this less perfect crystal is used, this is mentioned explicitly.
3.4 Experimental Characterization of Films

The main focus of this experiment on ultrathin Fe films on Cu(001) is on the magnetic domain pattern. The domain patterns are recorded using a Scanning Electron Microscope with Polarization Analysis (SEMPA). With the experimental setup, the out-of-plane component and one in-plane component of the polarization can be measured simultaneously. In the thickness range investigated in this experiment, only the out-of-plane component of the polarization is relevant. The in-plane component is found to vanish in all samples investigated.

The magnetic domain patterns not only depend on temperature but also on film thickness and preparation conditions. In order to examine the influence of the preparation conditions on the structural properties of the samples, Low-Energy Electron Diffraction (LEED) is used. It not only enables a qualitative assessment of film or substrate quality but also allows determining the interlayer distance.

Due to its capability to distinguish contributions from different elements, Auger Electron Spectroscopy (AES) is used to determine the film thickness after the magnetic characterization has been completed.
3.4.1 Magnetic Characterization with SEMP A

The magnetic characterization is made in an ultra-high vacuum chamber\(^2\) equipped with a Scanning Electron Microscope with Polarization Analysis (SEMPA).\(^3\) The SEMPA consists of a Scanning Electron Microscope (SEM) combined with a Mott detector. In order to obtain a map of the magnetization in a given area of a sample, the unpolarized primary electron beam of the SEM is scanned over the area where it excites a large number of secondary electrons. The polarization of these secondary electrons is determined by the magnetization at their origin. The polarization of the secondary electrons thereby depends on their energy. It is highest for low-energy secondary electrons. Moreover, the intensity of secondary electrons is concentrated at low energies. Therefore, these low-energy secondary electrons are selected for the spin analysis in the Mott detector. There the secondary electrons are accelerated onto a gold foil where spin-orbit coupling leads to spin-dependent scattering. The spin asymmetry of the secondary electrons results in a spatial asymmetry, which is determined by comparing the counts in detectors collecting primarily electrons with a given spin orientation. The component of the polarization of the secondary electrons along a direction \(i\) is given by

\[
P_i = \frac{1}{S} \frac{N_+ - N_-}{N_+ + N_-},
\]

where \(N_+\) and \(N_-\) are the numbers of electrons measured in two detectors placed in the plane containing the gold foil and perpendicular to the direction \(i\), and \(S\) is the Sherman factor determined by the setup of the Mott detector.\(^4\)

Fig. 3.5 shows a sketch of the experimental setup. The use of two mutually perpendicular pairs of detectors provides information on both the out-of-plane component of the polarization as well as one in-plane component. Due to the short mean-free path of low-energy electrons, the use of the low-energy secondary electrons makes SEMPA a highly surface-sensitive technique. This is important for the characterization of films consisting of only a small number of monolayers. The spatial resolution of SEMPA is given by the spatial resolution of the scanning electron microscope. The best resolution achievable with the present setup is on the order of 20 nm. High acceleration voltages in the SEM are favorable from the point of view of lateral resolution, but due to the larger penetration depth of high-energy electrons, the number of secondary electrons is reduced at higher acceleration voltages. In this

\(^2\) The pressure in the SEMPA chamber is kept below \(5 \times 10^{-11}\) mbar.

\(^3\) Exploiting the fact that secondary electrons ejected from a ferromagnetic sample are spin-polarized [101–103], the first SEMPAs were built by Koike and Hayakawa [104, 105] and Unguris et al. [106]. Overviews of SEMPA and its applications are given by Scheinfein et al. [107] and Allenspach [108].

\(^4\) The type of Mott detector used in this study is described in [109]. For the Sherman factor, see [110].
3.4. EXPERIMENTAL CHARACTERIZATION OF FILMS

Figure 3.5: Scanning Electron Microscope with Polarization Analysis. The beam of electrons from a Scanning Electron Microscope (SEM) is directed at the surface of the sample and creates numerous secondary electrons that leave the sample and carry the spin polarization of the sample. The secondary electrons are collected and accelerated to a high voltage. Due to spin-orbit coupling, the scattering at the gold foil is sensitive to the orientation of the spin component in the plane of the gold film. A setup with four detectors allows the detection of both the out-of-plane component of the spin polarization and the in-plane component parallel to the plane of the gold film.

study of ultrathin Fe films on Cu(001), the SEM acceleration voltage is set to 2 kV. Larger acceleration voltages tend to modify the magnetic domain pattern in this sensitive system. The same voltage of 2 kV was chosen by Allenspach and Bischof to avoid structural and magnetic changes in their experiments [5].

Standard SEMP A images are made of 256 × 256 pixels. The typical measuring time per pixel is 10 ms.

At each location on a sample where a magnetic image is taken, additional SEM topography images of different scales are made. These topography images permit to assign unusual features of the domain pattern to irregularities of the substrate. In the case of wedges, the topography images are used to map magnetic domain patterns to the correct film thickness.

3.4.2 Structural Characterization with LEED

LEED is mainly used for two purposes: first, LEED patterns provide a simple qualitative criterion for film or substrate quality; second, LEED patterns permit the identification of superstructures and \( I(E) \) curves are used to detect structural
differences between films grown on substrates at different temperatures. \( I(E) \) curves show the intensity \( I \) of a given beam of elastically scattered electrons as a function of electron energy \( E \).

LEED patterns are recorded using a CCD camera. \( I(E) \) curves are measured in two slightly different ways. One method consists of identifying the maximum intensity of the specularly reflected beam in a CCD image. In another method, a photo diode is used to measure the integral intensity of the specularly reflected beam on the LEED screen directly. In both cases, a small deviation of 5.5° from normal incidence is necessary to ensure the visibility of the specularly reflected beam on the fluorescent LEED screen.

As the primary electron beam has a diameter on the order of 1.5 mm, which is relatively large compared to the width of the Fe films of about 2 mm, the position of the sample has to be adjusted very carefully.

**3.4.2.1 Measuring \( I(E) \) Curves with a CCD Camera**

A CCD camera permits both the recording of diffraction patterns and the investigation of \( I(E) \) curves. Diffraction patterns taken at different electron energies are used to determine the energy dependence of the intensity in the center of the specularly reflected (00) beam. As taking CCD images is a rather time-costly endeavor, recordings are only made on one carefully chosen spot per sample. \( I(E) \) curves measured using a photo diode are labeled as \( I_{\text{max}}(E) \) curves.

**3.4.2.2 Measuring \( I(E) \) Curves with a Photo Diode**

Measuring \( I(E) \) curves using a photo diode is a relatively quick method. The photo diode is focused on the specularly reflected (00) beam and measures the integral intensity of the spot. \( I(E) \) curves are taken at the center of the Fe film and on the four branches of a cross centered there. Any contribution by diffraction at the free Cu(100) surface surrounding the Fe film should thus be discernable by comparing \( I(E) \) curves taken at different spots. \( I(E) \) curves measured using a photo diode are labeled as \( I_{\text{int}}(E) \) curves.

**3.4.3 Determination of Film Thickness Using AES**

Spatially resolved information on film thickness is gained by moving the sample under the AES electron beam and taking spectra every 0.1 mm. The thickness of the Fe film is calculated from the relative intensities of the main Fe and Cu peaks. A profile of a wedge-shaped Fe film obtained in this way is depicted in Fig. 3.4. Based on a direct comparison of the relative intensities of the main Fe and Cu peaks in Auger electron spectra and the Fe coverage observed in scanning tunneling
micrographs of Fe films on Cu(100), an empirical formula has been established to determine the film thickness as a function of the intensity ratio. It is given by

$$D[\text{ML}] = 4.1 \cdot \ln\left(1 + 1.2 \cdot \frac{I_{\text{Fe}}(654 \text{ eV})}{I_{\text{Cu}}(922 \text{ eV})}\right), \quad (3.3)$$

where $I_{\text{Fe}}(654 \text{ eV})$ is the intensity of the Fe peak at 654 eV and $I_{\text{Cu}}(922 \text{ eV})$ is the intensity of the Cu peak at 922 eV. This is the formula that has been used to determine the thickness of Fe films on Cu(100). However, the film thickness as a function of the intensity ratio can also be calculated directly under the assumption that the electron intensity decays exponentially with distance. The intensities of the Fe and Cu peaks are then given by

$$I_{\text{Fe}} = I_0 S_{\text{Fe}} \int_{-D}^{0} e^{z\left(\frac{1}{\lambda_{\text{Fe}}} - \frac{1}{\lambda_{0\text{Fe}}\text{Fe}}\right)} dz$$

$$= I_0 S_{\text{Fe}} \frac{\lambda_{\text{Fe}}\lambda_{0\text{Fe}}}{\lambda_{\text{Fe}} + \lambda_{0\text{Fe}}} \left\{1 - e^{-D\left(\frac{1}{\lambda_{\text{Fe}}} + \frac{1}{\lambda_{0\text{Fe}}\text{Fe}}\right)}\right\}, \quad (3.4)$$

$$I_{\text{Cu}} = I_0 S_{\text{Cu}} \int_{-\infty}^{0} e^{z\left(\frac{1}{\lambda_{\text{Cu}}} - \frac{1}{\lambda_{0\text{Cu}}\text{Cu}}\right)} dz \cdot e^{-D\left(\frac{1}{\lambda_{0\text{Cu}}} + \frac{1}{\lambda_{\text{Cu}}\text{Cu}}\right)}$$

$$= I_0 S_{\text{Cu}} \frac{\lambda_{\text{Cu}}\lambda_{0\text{Cu}}}{\lambda_{\text{Cu}} + \lambda_{0\text{Cu}}} e^{-D\left(\frac{1}{\lambda_{0\text{Cu}}} + \frac{1}{\lambda_{\text{Cu}}\text{Cu}}\right)}, \quad (3.5)$$

where $I_0$ is the intensity of the incident electron beam, $S_{\text{Fe}}$ and $S_{\text{Cu}}$ are sensitivity factors, and the $\lambda$'s are mean free paths. $\lambda_{0\text{Fe}}$ and $\lambda_{0\text{Cu}}$ are the mean free paths of the incident electrons in the Fe film and the Cu substrate respectively, $\lambda_{\text{Fe}}$ and $\lambda_{\text{Cu}}$ denote the mean free paths of Fe and Cu Auger electrons in their respective environment. The mean free path of Cu Auger electrons within the Fe film is denoted by $\lambda_{\text{Cu}/\text{Fe}}$.

The mean free paths can be calculated using an empirical formula [111]:

$$\lambda[\text{ML}] = \frac{538}{(E[\text{eV}])^2} + 0.41 \sqrt{a[\text{ML}]} \cdot E[\text{eV}] \quad (3.6)$$

$E$ is the electron energy, and $a$ the interlayer distance in the material. The film thickness can now be extracted from the ratio of the Auger electron intensities:

$$\frac{I_{\text{Fe}}}{I_{\text{Cu}}} = R \left\{e^{D\left(\frac{1}{\lambda_{0\text{Fe}}} + \frac{1}{\lambda_{\text{Cu}}\text{Fe}}\right)} - e^{-D\left(\frac{1}{\lambda_{\text{Fe}}} + \frac{1}{\lambda_{\text{Cu}}\text{Cu}}\right)}\right\}, \quad (3.7)$$

where the prefactor

$$R = \frac{S_{\text{Fe}}}{S_{\text{Cu}}} \frac{\lambda_{\text{Fe}}\lambda_{0\text{Cu}}}{\lambda_{\text{Fe}} + \lambda_{0\text{Cu}} \lambda_{\text{Cu}}}, \quad (3.8)$$
can be obtained experimentally by comparing the Auger electron intensities measured for sufficiently thick Fe films and clean Cu substrates. The values for the film thickness extracted from Eq. (3.7) are in good agreement with the values calculated using the empirical formula of Eq. (3.3).

AES measurements are always done after all the other characterisations have been completed; this is normally around 20 hours after the evaporation. Typically, some contamination of the Fe film with oxygen and carbon is found. The contamination increases with time. Hardly any contamination is found on the Cu substrate.

If Auger electron spectra are taken too late, the thickness of the Fe film tends to be overestimated. An increase of the measured thickness by 4.5% has been found after 2.5 days.

3.5 Structural Properties of Ultrathin Fe Films on Cu(100)

Numerous previous experiments and theoretical studies have shown that magnetic properties are tightly linked to the structure of a film. We will seize the opportunity and review some of the previous work on the system of ultrathin Fe films on Cu(100) before reporting the results of the present study.

3.5.1 Previous Studies

In an early experiment by Jesser and Matthews, transmission electron micrographs and diffraction patterns were used to determine the crystalline structure of iron films on Cu(100) \[112\]. Films of less than 20 Å revealed an fcc structure while misfit dislocations and nuclei of bcc iron were discerned in thicker films. The dislocation lines were found to be parallel to the [011] and [01\(\overline{1}\)] directions in the (100) film plane. During the growth of thicker films, the initial fcc structure has to undergo a transformation to achieve the bcc structure stable in thicker films. Jesser and Matthews identified three possible orientations of the bcc crystal with respect to the initial fcc structure: Nishiyama, Pitsch, and Bain orientations \[112\]. Later STM studies by Wuttig et al. showed that the transformation is connected to the appearance of stripes running in the [011] direction on the surface. These stripes were often found in the vicinity of dislocation lines, which serve as nucleation centers for the growth of bcc iron with a (110) orientation on the Cu(100) surface \[113\]. Atomically resolved scanning tunneling micrographs by Biedermann et al. revealed the arrangement of atoms in needle-shaped bcc nucleation centers \[114\]. It is similar to the Pitsch orientation but the bcc regions remain strained. The strained bcc configuration corresponds to a shear deformation of the fcc lattice. The instability of fcc iron on Cu(100) against a monoclinic shear deformation producing an almost bcc-like structure was confirmed by Spiššák and Hafner using local-spin-density calculations \[115\]. The instability is predicted to lead to the formation of a striped pattern of near-bcc domains, which is in agreement with the experimental observations.
tion of fcc iron of Cu(100) is facilitated by the close match of the respective lattice constants.\footnote{Bulk iron exists in a variety of crystal structures. Under normal conditions, bulk iron is characterized by a bcc structure and ferromagnetic. At 1041 K, this so-called $\alpha$ iron transforms into paramagnetic $\beta$ iron which has a bcc structure with a slightly increased lattice constant. At 1185 K, a structural transformation to an fcc structure is observed. Before melting at 1811 K, however, this so-called $\gamma$ iron transforms into another bcc phase, $\delta$ iron \cite{116}.}

Assuming an fcc structure, Fu and Freeman \cite{117} performed all-electron total-energy local-spin-density studies to calculate electronic and magnetic properties of one and two monolayers of iron on Cu(100). The magnetic moment at the surface was found to be enhanced.\footnote{The magnetic moments of closely packed surfaces of iron, cobalt, and nickel have been calculated by Aldén et al. by means of a spin-polarized Green-function technique based on the linear muffin-tin orbitals method within the tight-binding and atomic sphere approximations \cite{118}. Enhanced magnetic moments are obtained for all the surfaces considered with the exception of the Ni fcc(111) surface.}

It was slightly reduced by a copper overlayer. Surface and subsurface layers were predicted to couple ferromagnetically in contrast to the antiferromagnetic coupling in bulk fcc iron.\footnote{The magnetic properties of bulk bcc and bulk fcc iron were studied by Moruzzi et al. using the local-density approximation \cite{119}. Based on total-energy surfaces in the moment-volume parameter space, the magnetic moment of fcc iron is predicted to vanish whereas a ferromagnetic ordering is predicted for bcc iron. At expanded lattice parameters, however, the total energy of fcc iron has its minimum at a high moment, and at intermediate lattice parameters a local minimum of the total energy is found for a low-moment state. Dropping the restriction of ferromagnetic ordering, Moruzzi et al. later identified the antiferromagnetic state to be the ground state and the state with the lowest total energy over a considerable range of volumes \cite{120}. For sufficiently large lattice parameters, the antiferromagnetic ordering is replaced by the high-moment ferromagnetic state. Using the self-consistent linearized augmented-plane-wave method, Zhou et al. showed the coexistence of a ferrimagnetic state for lattice parameters in the range between 3.59 Å and 3.67 Å. \cite{121}.}

The distance between the copper substrate and the first iron layer was found to assume a reduced value as compared to the fcc copper lattice parameter. Very similar results were obtained by Fernando and Cooper using the self-consistent film linearized muffin-tin-orbital method \cite{122}. The preponderance of antibonding majority states at the surface was expected to favor an expanded lattice near the surface. Such an expansion of the lattice was indeed obtained by Kraft et al. applying the full-potential linear muffin-tin orbital method \cite{123}. The interlayer distance between the surface and subsurface layers was predicted to be expanded by 3.9%. The calculation of four to eleven monolayers of iron on Cu(100) established ferromagnetic ordering within each layer and between the surface and subsurface layers but antiferromagnetic coupling between the lower layers. Note that ferromagnetic intralayer coupling was presupposed in the studies by Fu and Freeman \cite{117} and Fernando and Cooper \cite{122}. The case of thinner films was examined by Lorenz and Hafner using self-consistent real-space
recursion calculations using a tight-binding-muffin-tin-obital Hubbard Hamiltonian [32]. Under the assumption of ideally pseudomorphic films, ferromagnetic coupling was observed for films consisting of up to three monolayers. In thicker films, ferromagnetic coupling was restricted to surface and subsurface layers while lower layers again coupled antiferromagnetically. Magnetic moments at the interface with copper were reduced with respect to magnetic moments at a free surface due to hybridization of the $d$ states. Iron films of up to three monolayers were allowed to relax in full-potential linearized-augmented-plane-wave calculations using generalized gradient Perdew-Burke-Ernzerhof functionals by Yin and Hermann [124]. The whole films were found to be ferromagnetic and the interlayer distances as well as the magnetic moments were increased. The increase was largest for the topmost layer.\(^9\)

These calculations show the interdependence of magnetic and structural properties of ultrathin fcc iron films on Cu(100). Ferromagnetic ordering is restricted to layers near the surface and coupled to an increased interlayer distance. Hybridization of the $d$ states of iron with $d$ states of an overlayer reduces the magnetic moments. The magnetic properties of ultrathin iron films on Cu(100) can thus be expected to be very sensitive to growth conditions, which can have a strong impact on both film morphology and film structure.

Basically two preparation procedures have been applied to produce ultrathin iron films on Cu(100) by MBE. The two procedures result in different morphological and structural properties and thus entail different magnetic properties. One procedure consists in growing the films on substrates held at a low temperature of about 100 K and subsequent annealing at room temperature. In the other procedure, the iron films are directly grown on a substrate held at room temperature. We will label films grown under these conditions as low-temperature- and room-temperature-grown films respectively.

Low-temperature-grown films were investigated by Pappas et al. [125, 126]. LEED patterns of films of 5–7 ML right after the evaporation showed a $1 \times 1$ structure, which transformed into a $5 \times 1$ superstructure after annealing to room temperature. Spin-polarized secondary-electron spectroscopy was used to measure the remanent magnetization of such annealed films. While films thinner than 5 ML were characterized by a perpendicular magnetization, films in the range of 5–7 ML exhibited a reversible transition from a perpendicular magnetization at low temperatures to

\(^9\) Using the generalized gradient approximation combined with the full-potential linearized augmented-plane-wave method, Asada and Blügel calculated the energies of all collinear states of fcc iron films of up to six monolayers on Cu(100) [100]. Ferromagnetism was found for films of up to three monolayers, bilayer antiferromagnetism in films consisting of four or six layers and the coexistence of several spin states for five monolayers. Note that these results apply to unrelaxed films. Interlayer relaxation was then considered for films of one, five, or seven monolayers. Ferromagnetic coupling at the surface was accompanied by an expansion of the interlayer distance at the surface by 2%.
an in-plane magnetization at higher temperatures. Films of 10 ML were always magnetized in the plane. The reversible switching of the magnetization direction in films of 5–7 ML was accompanied by a puzzling absence of magnetization in a finite temperature range of about 30 K. This puzzle was resolved by SEMPA measurements by Allenspach and Bischof [5]. They showed that the transition region is marked by the appearance of perpendicularly magnetized stripe domains so that the overall magnetization vanishes although ferromagnetic ordering persists within the stripe domains.

The difference in growth mode between low-temperature grown films and films grown at room temperature has been investigated in many studies. Steigerwald et al. estimated the surface energies of copper and fcc iron [127]. The values obtained for the surface energy of copper and fcc iron are 1.85 J/m$^2$ and 2.15 J/m$^2$ respectively. Hence, layer-by-layer growth is energetically unfavorable. Moreover, the heat of mixing is positive. Steigerwald et al. distinguish three temperature ranges for the substrate during iron deposition: temperatures below 200 K, temperatures between 200 K and 300 K, and temperatures above 300 K. The film quality was determined from XPS spectra, XPS forward scattering, a CO-titration technique and LEED patterns. Iron films grown at low temperatures were found to be poorly ordered, films grown at room temperature showed agglomeration. For temperatures above room temperature, copper segregation led to intermixing. LEED spot sizes decreased considerably when the substrate temperature during deposition was increased. A high initial nucleation density of islands was found at both room-temperature and low-temperature deposition by Giergiel et al. using scanning tunneling microscopy [128]. While the number of islands decreased rapidly in room-temperature-grown films due to coalescence upon further deposition, the number of islands in low-temperature-grown films remained almost constant. Low-temperature-grown films were thus characterized by a much higher roughness. The percolation threshold in low-temperature-grown films was increased with respect to room-temperature grown films. In another STM study by Chambliss et al. [129], small height variations were detected at the initial stages in room-temperature-grown films and interpreted as due to intermixing. Patches of iron were found embedded in the surface layer of the copper substrate, and copper patches were found to form part of the islands. Intermixing was also observed by Noh et al. for the first monolayer deposited on a copper substrate [130].

**10** This might explain RHEED oscillations indicating initial bilayer growth at room temperature [132]. Three-dimensional growth was deduced for deposition at low temperatures. MEED and RHEED oscillations have often been used to characterize the growth mode of the films. The studies generally agree that

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10 Intermixing did not occur in subsequent layers which grew in the layer-by-layer mode up to a thickness of 5 ML. Hydrogen adsorption measurements by Egawa et al., however, either indicate that the copper substrate is not completely covered by films of up to 3 ML or signal that intermixing persists up to this coverage [131].
layer-by-layer growth is present in the thickness range between 5 ML and 11 ML in room-temperature-grown films as evidenced by regular oscillations. In thicker films, the oscillations disappear. The situation of thinner films, however, is somewhat more controversial. Oscillations in this thickness range are less regular. A minimum at a coverage of 1 ML was observed in some studies [133, 134] but rather regular oscillations were found in another study [135]. However, the appearance of RHEED oscillations in very thin films has been shown to depend on the choice of the azimuthal angle [136]. An interesting dependence of the growth mode on the deposition rate was observed by Man et al. [137]. The LEED and RHEED intensity oscillations corresponding to the completion of the first iron layer virtually absent when the deposition rate exceeded 0.5 ML/min were found to be strongly enhanced when the deposition rate was lowered below 0.5 ML/min. The simultaneous growth of the first and second atomic layers was converted to a more perfect layer-by-layer growth when the deposition rate was lowered below 0.5 ML/min.\textsuperscript{11} Based on the dependence of Auger-electron intensity on film thickness, other studies report layer-by-layer growth already at the initial stages for substrates at room temperature or 350 K [28, 140].

Using spin-polarized photoelectron emission, Pescia et al. showed that room-temperature-grown fcc iron films on Cu(100) were ferromagnetic with a perpendicular anisotropy for thicknesses of up to five monolayers [28]. A detailed structural and magnetic study of room-temperature-grown films was carried out by Thomassen et al. by combining LEED and SMOKE [133]. It allowed dividing ultrathin iron films on Cu(100) into three distinct thickness regions. Region I comprises films up to 4 ML. These films have a perpendicular anisotropy, and the Kerr ellipticity increases linearly with thickness. Films in the thickness range between 4 ML and 11 ML (region II) show a reduced but constant Kerr ellipticity whereas thicker films (region III) again show a linearly increasing Kerr ellipticity and in-plane anisotropy. The crystalline structure in regions I and II is fcc; in region III it is bcc. The magnetic reorientation transition at the border of regions II and III is thus directly related to a structural transformation, in contrast to the case of low-temperature-grown films where the reorientation transition is purely magnetic. The reduced but constant Kerr ellipticity of the films in region II has been associated with ferromagnetic coupling between the surface and subsurface layers.

\textsuperscript{11} This observation is relevant for the ultrathin iron films investigated in this thesis. While most previous experiments involved films grown at deposition rates larger than 0.5 ML/min, the films investigated in this thesis were grown at a deposition rate of about 0.1 ML/min. Interestingly enough, ultrathin iron on Cu(100) produced by pulsed laser deposition as well showed layer-by-layer growth from the very beginning [138, 139]. This was not only reflected in clear RHEED intensity oscillations but also confirmed by scanning tunneling microscopy. The instantaneous deposition rate during each pulse was orders of magnitude larger than for MBE-grown films. However, the average deposition rate was 0.1 ML/min.
Exposure to CO lowered the Curie temperature considerably, which supports the idea of a surface 'live' layer. As theoretically expected [122, 123], the interlayer distance between the surface and subsurface layers was found to be expanded, the increase amounting to 0.1 Å.

Thomassen et al., however, did not detect any antiferromagnetic ordering in the interior of the films for temperatures as low as 100 K. These findings by Thomassen et al. concerning the magnetic ordering in the interior are in contradiction to SMOKE measurements by Li et al. which show an antiferromagnetic ordering in the interior [135]. The period of the antiferromagnetic ordering is found to be 2.6 ML, which suggests a role for spin-density waves. The Néel temperature of the antiferromagnetic interior was around 200 K, i.e. somewhat lower than the Curie temperature of the ferromagnetically coupling live surface layers, which is around 250 K. The results of Li et al. were confirmed by MOKE measurements by Qian et al. [136]. The magnetic moment per atom in the spin-density wave is not reduced with respect to the magnetic moment in the ferromagnetically coupling surface and subsurface layers but is estimated to amount to 60 % of that of the ferromagnetic fcc phase at 4 ML [141].

Like films grown at low temperatures, films grown at room temperature show LEED superstructures. Egawa et al. found four different thickness regimes [131]: a 1×1 pattern was observed for films of up to two monolayers and in the thickness range between 6 and 10 ML, a 4×1 superstructure for films of 3–5 ML, and finally a streaky 3×1 superstructure for thicker films with the bcc structure. A slightly different sequence was observed by Thomassen et al. [133] and confirmed by Heinz et al. [142]. According to their results, films of a thickness of 1 or 2 ML have a 4×1 superstructure, a 5×1 superstructure is present at 4 ML, and a 2×1 superstructure is found for films of 5–8 ML. The bcc phase was again associated with a 3×1-like superstructure in films thicker than 12 ML. A 1×1 pattern instead of the 2×1 superstructure was, however, observed by Li et al. [135].

Fcc iron films on Cu(100) experience some strain due to the lattice mismatch between fcc iron and copper. Interlayer distances will thus be relaxed. A quantitative LEED intensity-versus-energy analysis by Wuttig and Thomassen yielded an unrelaxed in-plane lattice constant of 3.61 Å and a reduced lattice constant perpendicular to the film plane of 3.54 Å [134]. The interlayer distance between the surface and subsurface layers was found to be increased by 0.1 Å. This was attributed to the ferromagnetic coupling between the surface and subsurface layers. The interlayer distances of a 2-ML film were determined by Heinz et al. using the tensor LEED technique to fit experimental intensity-versus-energy curves [142]. The values obtained were 1.87 Å for the distance between surface and subsurface layers and 1.84 Å for the interface. Using helium atom scattering, Dastoor and Allison obtained a sim-
ilar value for 2 ML of 1.86 Å [143]. Thinner films were found to be expanded even more with an interlayer distance of 2.1 Å; films in the thickness range between 2 and 4 ML had an interlayer spacing of about 1.9 ML.

In their analysis of LEED patterns of films of 2 ML, Heinz et al. noticed that 1/4 spots were missing [142]. Randomly distributed micro-domains with a 4×1 superstructure were suggested as an explanation. An analysis of the 5×1 superstructure of 5 ML yielded sinusoidal shifts and vertical buckling [146]. Recent atomically resolved STM images by Biedermann et al. indeed revealed a complex structure of such ultrathin films [147]. Bcc-like crystal structures coexisting with remnants of fcc structure were discovered in films of 2–4 ML with the highest bcc-like content at 3 ML. The bcc-like crystal structure was restricted to elongated stripes 2–4 atoms wide.

3.5.2 Present Study

The iron films studied in this thesis were deposited at various substrate temperatures. In this subsection, we will investigate the structural properties of films prepared at 300 K and 315 K respectively. LEED patterns will be used to discuss the symmetry, and LEED I(E) curves will reveal interlayer distances. Although films prepared at 300 K or 315 K have very similar properties, some differences do occur.

From the point of view of magnetic domains, the thickness range around 2 ML is of particular interest as changes of the domain pattern occur in this thickness range. The precise value of the thickness at which a certain domain pattern occurs is dependent on growth conditions. As some minor variations of the growth conditions over years of experiments are inevitable, the thickness at which the labyrinthine pattern occurs at room temperature has also been subject to minor variations. Under the growth conditions present during the LEED measurements, the labyrinthine pattern was found at 2.3 ML in films grown on a substrate at 315 K. It was already replaced by a stripe domain pattern in films deviating from this thickness by as little as 0.1 ML. The labyrinthine pattern is absent in films grown at 300 K. In order to correlate the domain pattern with the film structure, homogeneously thick films of 2.2 ML, 2.3 ML, and 2.4 ML were deposited onto substrates at 315 K and 300 K. The diffraction pattern of both films grown at 315 K and 300 K show a 4×1 superstructure. This is in agreement with results obtained in previous experiments on films of about 2 ML [133, 135]. No systematic trend is observed for films grown at

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12 Based on the simpler kinematic analysis of LEED intensity-versus-energy curves a set of two lattice spacings was obtained by Zharnikow et al. for each thickness [144, 145]. For 2 ML, the dominant lattice spacing was 1.9 ML, the other spacing being 1.8 ML. For films of 5 ML, the value of the dominant lattice spacing was reduced to 1.85 ML while the other lattice spacing remained unchanged.
either 315 K or 300 K, or films with slightly different thicknesses. A diffraction pattern recorded at 111 eV is depicted in Fig. 3.6 along with a secondary-electron image. The clear 4×1 superstructure is superimposed with a faint p(2×2) superstructure. Such a p(2×2) superstructure was also observed in LT-grown films annealed at temperatures above 400 K \[126\].

Fig. 3.7 shows the integral intensity of the specularly reflected (00) beam as a function of the applied voltage for both a 2.3-ML film grown at 315 K and the copper substrate. The \(I_{\text{int}}(E)\) curve of the iron film is characterized by the presence of two sets of Bragg peaks. One set of peaks occurs at voltages near which peaks appear for the copper substrate. These peaks are encircled in blue. The other much more prominent set encircled in red corresponds to a structure with an increased interlayer distance. It is shifted toward smaller voltages. We will call these peaks fcc and fct (face-centered-tetragonal) peaks respectively.\(^\text{14}\)

Bragg peaks in \(I(E)\) curves occur whenever there is constructive interference of

\(^{13}\text{Further annealing to 650 K led to a transition to a c(2×2) superstructure with an enhanced copper content in the film [126]. A c(2×2) superstructure was also observed in RT-grown films of 3–5 ML annealed to 490 K [148]. It was tentatively associated with the formation of an iron-copper alloy.}\)

\(^{14}\text{In a previous experiment [147], a considerable content of bcc-like iron was detected in this thickness range. However, due to the basically fct-like diffraction pattern (Fig. 3.6), we will retain the term of face-centered-tetragonal to describe this phase.}\)
Figure 3.7: LEED $I_{\text{int}}(E)$: Fct and fcc peaks. $I_{\text{int}}(E)$ curves of ultrathin iron films on Cu(100) in the thickness range around 2 ML are characterized by two sets of peaks. The dependence of the integral intensity of the (00) spot of a 2.3-ML film grown at 315 K as a function of voltage is shown in black, the corresponding curve of the copper substrate in blue. Blue circles indicate peaks of the iron film that occur near the Bragg peaks of the copper substrate and are thus indicative of an fcc structure. The red circles denote peaks that can be associated with a lattice with an expanded interlayer distance. They correspond to the Bragg peaks of an fct structure. Additional peaks of the copper substrate where no equivalent peaks are marked on the $I_{\text{int}}(E)$ curve of the iron film are due to multiple scattering and correspond to fractional values of $n$ (cf. [149]).

Electrons scattered at different lattice planes, i.e. whenever the Bragg condition $\vec{K} \cdot \vec{D} = n \cdot 2\pi$ is met. $\vec{K}$ is the change in wave vector between the scattered and the initial electrons, $\vec{D}$ is a vector perpendicular to the surface with a length equal to the interlayer distance $D$, and $n$ is an integer number. The change in wave vector $\vec{K}$ is connected to the wave vector of the incident beam $\vec{k}$. When the angle between the surface normal and the incident wave vector is $\theta$, the modulus of the change in wave vector for specularly reflected electrons is given by $|\vec{K}| = 2|\vec{k}| \cos \theta$. Setting the kinetic energy of the impinging electrons equal to the energy gained by passing the voltage difference $V$ and assuming specularly reflected electrons, we obtain the following formula for maximum intensity:

$$V_n = \frac{Mn^2}{4D^2\cos^2\theta} - V_0 \quad ,$$

(3.9)
Figure 3.8: LEED $I_{\text{int}}(E)$: Influence of growth conditions. $I_{\text{int}}(E)$ curves of films of 2.4 ML (black), 2.3 ML (blue), and 2.2 ML (red) are shown. 

a, Films grown at 315 K. b, Films grown at 300 K.

Films grown at 315 K are marked by an increased contribution of fcc peaks.
where
\[ M = \frac{\hbar^2}{2q_e m_e} = 1.5 \text{ Vnm}^2. \] (3.10)

When electrons enter a material, their kinetic energy is increased by the inner potential. Here, this fact is taken into account of by introducing a constant \( V_0 \). Bragg peaks are characterized by integer values of \( n \), peaks due to multiple scattering have fractional values of \( n \).

Fig. 3.8 shows \( I_{\text{int}}(E) \) curves for films of 2.2 ML, 2.3 ML, and 2.4 ML prepared at 315 K and 300 K. The 2.3-ML film prepared at 315 K is the only film with a labyrinthine domain pattern; all the other films have stripe domains. A comparison of the \( I_{\text{int}}(E) \) curves of the films grown at 315 K makes clear that the occurrence of the labyrinthine pattern is not related to special features of the structure. All the \( I_{\text{int}}(E) \) curves of films grown at 315 K are very similar. There is, however, a difference between films grown at 315 K and films grown at 300 K. Films grown at 315 K are marked by a slightly larger contribution of fcc peaks. This enhanced contribution is especially conspicuous in the vicinity of the main fct peak near 150 V. The fcc peak around 180 V is much more prominent in films grown at 315 K. An intensity dip present just below the main fct peak in films grown at 300 K is filled by the strong \( n = 3.5 \) fcc peak at 135 V in films grown at 315 K.

The interlayer distances of the fct and fcc lattices can be obtained from the linear dependence of the Bragg peak positions on \( n^2 \), cf. Eq. (3.9). In order to obtain reliable quantitative results, the LEED setup was calibrated on a clean Cu(100) surface. The interlayer spacing of the copper crystal was assumed to be equal to its bulk value of 1.805 Å. Although surface reconstructions do occur, the error introduced by this assumption can be estimated to be below 1%.\(^ {15} \) The peak positions of the fct and fcc lattices as a function of \( n^2 \) are plotted in Fig. 3.9 for 2.3-ML films grown at 315 K and

\(^ {15} \) By means of a dynamical LEED analysis which included both multiple scattering and a complex inner potential, Davis and Noonan had determined a multilayer reconstruction at the Cu(100) surface [150]. While the first two interlayer distances at the surface were found to be reduced by about 1%, an increase of 1% was observed for the third interlayer distance. Deeper interlayer spacings were not varied. Such an involved dynamical analysis would be beyond the scope of this work. We will restrict ourselves to the kinematic approach, which neglects multiple scattering and attenuation, and make use of Eq. (3.9). Other faces of copper were investigated in a further study by Davis and Noonan [151].

\(^ {16} \) Based on qualitative considerations, Finnis and Heine provided arguments for a reduced interlayer distance at the surface of metals [152]. Due to the high surface tension of the electron fluid, an electron redistribution can be expected at the surface. Such a redistribution will lead to electrostatic forces on the ion cores at the surface and induce a contraction of the interlayer spacing near the surface. The magnitude of the contraction will depend on both surface orientation and crystal structure. While Finnis and Heine used a simple step function to describe the electron density at the surface, Landmann et al. used three different, more realistic models of the electron density [153]. According to their calculations, multilayer relaxations at metal surfaces can be
Figure 3.9: LEED $I_{\text{int}}(E)$: Peak energy versus $n^2$. The energy of fct (squares) and fcc (circles) Bragg peaks is plotted as a function of $n^2$ for films of 2.3 ML.

a, Film grown at 315 K. b, Film grown at 300 K.
Temperature during Growth: 315 K

<table>
<thead>
<tr>
<th>Thickness</th>
<th>(d_{\text{fcc}}) [Å]</th>
<th>(d_{\text{fct}}) [Å]</th>
</tr>
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<tbody>
<tr>
<td>2.4 ML</td>
<td>1.82±0.03</td>
<td>1.91±0.03</td>
</tr>
<tr>
<td>2.3 ML</td>
<td>1.82±0.03</td>
<td>1.91±0.03</td>
</tr>
<tr>
<td>2.2 ML</td>
<td>1.81±0.03</td>
<td>1.90±0.03</td>
</tr>
</tbody>
</table>

Temperature during Growth: 300 K

<table>
<thead>
<tr>
<th>Thickness</th>
<th>(d_{\text{fcc}}) [Å]</th>
<th>(d_{\text{fct}}) [Å]</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.4 ML</td>
<td>1.81±0.03</td>
<td>1.89±0.03</td>
</tr>
<tr>
<td>2.3 ML</td>
<td>1.81±0.03</td>
<td>1.89±0.03</td>
</tr>
<tr>
<td>2.2 ML</td>
<td>1.81±0.03</td>
<td>1.90±0.03</td>
</tr>
</tbody>
</table>

**Table 3.1:** Interlayer distances obtained from fcc and fct peaks in \(I_{\text{int}}(E)\) curves. The interlayer distances of films grown at 315 K tend to be somewhat larger. The interlayer distance of the fcc structure slightly exceeds the interlayer distance of bulk copper.

300 K. The expected linear dependence is experimentally verified. The values of the interlayer distances extracted from the proportionality factor are listed in Tab. 3.1. The fcc interlayer distance of about 1.81 Å is very close to the interlayer distance of bulk copper of 1.805 Å. The fcc peaks are slightly shifted toward smaller energies with respect to the Bragg peaks of the copper substrate, cf. Fig. 3.7. This suggests a larger interlayer distance than in the copper substrate. The values obtained for the fcc lattice indeed tend to be slightly larger than the interlayer distance in the copper substrate. The interlayer distance of the fct lattice is on the order of 1.90 Å, which is in very good agreement with values obtained in previous studies using this kinematic analysis of LEED \(I(E)\) curves or helium atom scattering [143, 145]. It is slightly larger than the distance between the surface and subsurface layers of 1.87 Å or the interface interlayer distance of 1.84 Å obtained in a tensor LEED analysis [142].

The inner potential obtained from the kinematic analysis is different for fct and fcc peaks. It is on the order of 16 V for the fct peaks. The average of the inner potential of films of 2.4 ML, 2.3 ML, and 2.2 ML grown at 315 K is \((15.2±0.5)\) V, which is only slightly smaller than the average of films grown at 300 K of \((16.7±0.9)\) V. The inner potential associated with the fcc peaks, however, differs clearly for films grown at 315 K or 300 K. The former have an average inner potential of \((6.2±0.2)\) V while the average inner potential of films grown at 300 K is \((8.8±0.4)\) V.

Previous experimental and theoretical studies produced inner potentials ranging from 6.6 V to 15 V [145, 149, 154–156]. It was argued that intralayer distances have

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expected to comprise 3–6 layers at least-densely packed faces (fcc(110) and bcc(100)) and 1–3 layers at other faces. This finding is supported by calculations by Fernando and Cooper which showed that the density of states of copper two layers below the (100) surface is very similar to the density of states of bulk copper [122].
Figure 3.10: LEED $I_{\text{max}}(E)$: Influence of growth conditions. $I_{\text{max}}(E)$ curves of films of 2.4 ML (black), 2.3 ML (blue), and 2.2 ML (red) are shown. 

a, Films grown at 315 K. b, Films grown at 300 K.
a considerable impact on the inner potential [154]. Moreover, the inner potential is a function of the energy of the impinging electron [155]. The adjustment of the electrons in the solid depends on the energy of the impinging electron. If this energy dependence is not neglected, a correction of the interlayer distance on the order of 0.6 % can be expected [156].

The enhanced contribution of fcc peaks in films grown at the elevated temperature of 315 K together with an inner potential close to the inner potential of the Cu(100) substrate of about 6.4 V may suggest intermixing of the iron film with the copper substrate, a copper overlayer, or the formation of pits in the iron films but an fcc structure of the iron film itself cannot be excluded. A comparison of the Auger spectra of films prepared at 315 K and 300 K with identical MBE settings reveals no enhancement of the Auger peaks of copper in films prepared at 315 K. However, due to the higher energy of the electrons involved, Auger electron spectroscopy is less sensitive to details of the surface than LEED. In order to get a better understanding, scanning tunneling micrographs of the various films would be desirable. The larger inner potential of the fcc structure in films prepared at 300 K remains a puzzle.

In conclusion, both films deposited at 315 K and 300 K are characterized by a predominantly fct structure. The interlayer distance perpendicular to the surface is on the order of 1.9 Å. In both cases, some fcc contribution is seen, it is slightly more important in films grown at 315 K. The origin of this fcc contribution is not clear. The determination of the inner potential suggests a role for copper in films grown at 315 K. However, no enhanced copper contribution is observed in Auger-electron spectra. The blurred distribution of the intensity in LEED spots due to specular reflection at the fcc structure makes an exposure of the copper substrate unlikely. The LEED patterns show a 4×1 superstructure. No evidence for an impact of the magnetic domain pattern on the crystalline structure has been found.

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17 A reversible transition of an fct to an fcc structure in films of 4 ML was observed by Zharnikov et al. as the temperature was varied [144, 145]. A theoretical explanation for this behavior was given by Spišák and Hafner [157].

18 In an STM study, Shen et al. discovered the formation of rectangular pits in films annealed at 490 K [148]. The pits were identified as channels for the diffusion of copper from the substrate to the film surface where a surface alloy formed.
### 3.6 Curie Temperature as a Function of Film Thickness

Knowledge of the thickness dependence of the Curie temperature $T_c$ is instrumental for understanding the behavior of magnetic domains in ultrathin iron films on Cu(100). In the thickness range relevant for the experiments reported in Part I, the Curie temperature increases monotonously with film thickness. Moving along a wedge towards smaller thicknesses at a given temperature thus results in an increase of the effective temperature. That the effective temperature is indeed the appropriate parameter for describing the sequence of phases is expounded in Sec. 3.10. The thickness dependence of $T_c$ has been determined in previous experiments.\(^{19,20}\)

Whereas the previous experiments relied on extrapolating to zero saturation, SEMPA images allow determining the Curie temperature in a more direct way. The Curie temperature is here defined as the temperature at which the magnetic domains in images covering the transition vanish.\(^{21}\)

Fig. 3.11 shows the Curie temperature in the thickness range between 1.3 ML and 2.9 ML for a film grown at 315 K. Qualitatively, the Curie temperature determined here is in good agreement with previous results. Quantitatively, some deviations from the results found by Thomassen et al. do occur. While the maximum Curie temperature obtained by Thomassen et al. was above 370 K, the maximum Curie temperature observed here is around 325 K. Moreover, the maximum Curie temperature is shifted toward smaller thicknesses. Man et al. have noticed that reducing the deposition rate results in a suppressed Curie temperature [137]. Indeed, the

\(^{19}\) Thomassen et al. measured Kerr ellipticities at saturation at various temperatures and determined $T_c$ by extrapolating to zero saturation ellipticities [133]. They covered the thickness range of 2–10 ML and found that after a small initial increase $T_c$ dropped quite dramatically as the film thickness was further increased. This drop has to be associated with magnetic and structural transformations, cf. Sec. 3.5. Due to the higher coordination number in thicker films, an increase of $T_c$ would be expected. The maximum of $T_c$ was located at around 3 ML. Beyond 5 ML, $T_c$ remained constant.

\(^{20}\) By extrapolating the linear dependence of the saturation magnetization on temperature to zero saturation magnetization, Pescia et al. obtained Curie temperatures of (230±30) K for 1 ML, and of (390±30) K for films of 3 and 5 ML [28].

\(^{21}\) In films with a Curie temperature on the order of room temperature or higher, the magnetic domains become highly mobile just before the paramagnetic state is reached, cf. Subsec. 3.12.3. When the velocity of the domains is such that the magnetization per pixel averages to zero within 10 ms, which is the typical measuring time per pixel, the magnetic contrast is lost even though the magnetization within the quickly moving domains may still be there. However, an extrapolation of the magnetization as a function of temperature, see Subsec. 3.7.1, indicates that the value for the Curie temperature obtained by visual inspection of SEMPA images underestimates the Curie temperature only marginally. The mobile phase is restricted to a temperature interval on the order of 1–2 K. Where not indicated otherwise, the term Curie temperature will be understood as the transition temperature determined from visual inspection of SEMPA images.
Figure 3.11: Curie temperature as a function of film thickness. $T_c$ was measured for a number of locations on a wedge grown at 315 K. $T_c$ first increases, then drops again slowly as the film thickness is increased. The experiments reported in Part I were performed on films with a thickness smaller than that for which $T_c$ reaches its maximum.

deposition rate used here is an order of magnitude lower than the deposition rate used by Thomassen et al. As a small variation of the substrate temperature during deposition has a similar effect as a change in deposition rate, the Curie temperature of films grown at other temperatures can be expected to deviate from the curve in Fig. 3.11. This will be discussed below. The thickness range relevant for Part I is the thickness range below the maximum of the Curie temperature curve. It is depicted in Fig. 3.12. The thickness dependence of the Curie temperature according to the mean-field theory is given by Eq. (2.49):

$$T_c(D) = \frac{J}{k_B} \left\{ 6 - \frac{2a}{D} \right\} = T_c(\infty) \left\{ 1 - \frac{1}{3} \left( \frac{a}{D} \right) \right\} .$$

The best fit to the data achievable with this formula is indicated by the dotted line. It yields $J = (60 \pm 5) k_B$, which corresponds to a bulk Curie temperature of $T_c(\infty) = (360 \pm 30)$ K. Obviously, the thickness dependence of the Curie temperature in ultrathin Fe films on Cu(001) cannot be adequately described within the mean-field theory.
Using a high-temperature series expansion of the zero-field susceptibility, Allan calculated \( T_c \) for films of 2, 3, 4, and 5 ML \([158]\). For thick films, he expected a power-law thickness dependence:

\[
T_c(D) = T_c(\infty) \left\{ 1 - c \left( \frac{a}{D} \right)^\lambda \right\}.
\]  

(3.11)

A fit to the calculated values of \( T_c \) for films of 2, 3, 4 and 5 ML yielded \( \lambda = 1.27 \pm 0.05 \). However, the applicability of the extrapolations used by Allan was seriously questioned by Domb \([159]\). Nonetheless, the experimental values of \( T_c \) obtained here can be fitted quite accurately using this more general power law conceived for thicker films. The solid line in Fig. 3.12 represents the best fit using Eq. (3.11). The bulk Curie temperature obtained in the fit is \((357 \pm 3) \text{ K}\), the other parameters are \( \lambda = 4.25 \pm 0.14 \) and \( c = 1.92 \pm 0.06 \). \( T_c \) decreases much faster than expected from the mean-field theory. If this more general power law is indeed applicable here, films as thin as 1.2 ML will become ferromagnetic at low enough temperatures.

In a more recent study, Zhang and Willis presented a model of spin-spin coupling which showed that the power-law dependence of \( T_c \) was replaced with a linear depen-
Figure 3.13: Influence of growth conditions on the Curie temperature. The maximum of $T_c$ increases slightly and the magnetic thickness range at a given temperature becomes broader when the substrate temperature during evaporation is lowered. $T_c$ is plotted for films grown with a delay after annealing of 35 min (solid circles), 45 min (open circles), 55 min (solid triangles), and 65 min (open triangles). The Curie temperatures were measured on wedges. While thickness differences can be determined quite accurately, the absolute thicknesses are less certain. This is due to the problem of correlating Auger electron spectra with SEMP A images. The curves have been shifted by up to 0.15 ML in order to achieve a better coincidence.

The thickness dependence of $T_c$ of ultrathin Ni films grown on Cu substrates as well as previous results for other systems could be explained with this model.\footnote{A close correlation between the percolation of the first monolayer and the transition between superparamagnetic and ferromagnetic behavior was observed in ultrathin Fe films on W(110) by Elmers et al. [161]. There, the transition was observed at a coverage of 0.6 ML.}

Assuming the general validity of such a behavior and extrapolating $T_c$ linearly below room temperature (dashed line in Fig. 3.12), we find that even films of less than one monolayer should become ferromagnetic at low temperatures. In an STM study, Giergiel et al. determined the percolation threshold of ultrathin Fe films grown under very similar conditions as 0.9 ML [128]. This suggests that ultrathin Fe films on Cu(100) become ferromagnetic as soon as the first atomic layer of Fe percolates.\footnote{The thickness dependence of $T_c$ of ultrathin Ni films grown on Cu substrates as well as previous results for other systems could be explained with this model.}
The Curie temperatures for films grown at slightly different temperatures are plotted in Fig. 3.13. For films grown at higher temperatures, the magnetic thickness range at a given temperature is reduced, and $T_c$ is diminished. As shown in Sec. 3.5, films grown at higher temperatures are characterized by an increased contribution of fcc peaks. Such an increased contribution of fcc peaks has been found to be correlated with a reduced magnetization \[144\]. Along with a reduced magnetization, a reduced $T_c$ can be expected. Note, however, that a variation of the fcc content in \[144\] was obtained by varying the temperature of the film during the measurement and not by varying the substrate temperature during the deposition of the film.

### 3.7 Mean-Field Aspects

The Scanning Electron Microscope with Polarization Analysis (SEMPA) is an ideal tool to analyze geometrical features of magnetic domain patterns. But being based on measuring the spin polarization of the secondary electrons, it also offers a way of tracking the spontaneous magnetization within the domains as a function of temperature. Under the assumption that the magnetization is proportional to the spin polarization of the secondary electrons, the critical exponent $\beta$ can be determined directly. The result of this analysis is presented in Subsec. 3.7.1. A feature of magnetic domain patterns the knowledge of which is instrumental in understanding geometrical transformations is the average domain width. A number of theoretical works have been concerned with the stripe-domain width of perpendicularly magnetized ultrathin films \[3, 37, 162\]. They predict the stripe domain width to decrease as the temperature is increased. This has indeed been observed \[5\]. In Sec. 3.7.2, a detailed analysis of the domain width near the transition to the paramagnetic phase will reveal a good agreement with Eq. (2.52).

#### 3.7.1 Magnetization as a Function of Temperature

In this subsection, the critical exponent $\beta$ will be determined based on the assumption that the spontaneous magnetization is proportional to the spin polarization of the secondary electrons. In such an approach, the contribution to the spontaneous magnetization due to the orbital moments is neglected. The spin polarization at a given temperature can be extracted from SEMPA images by fitting a Gaussian to the histogram of polarizations present in a single domain. While this is easily feasible for large domains, this procedure is less practical near the transition to the paramagnetic state where the domain width becomes small. Therefore, two Gaussians are simultaneously fitted to the histogram of an entire SEMPA image. Fig. 3.14 shows the polarization as a function of temperature. The polarization...
Figure 3.14: Polarization as a function of temperature. The polarization is obtained by fitting two Gaussians to the polarization histograms of SEMPA images. The solid line represents a power-law fit to the data. The critical exponent $\beta$ is found to be $0.25 \pm 0.05$. For the Curie temperature a value of $T_c = 325.7 \pm 1.3$ K is obtained. The film thickness is 1.95 ML.

decreases as the transition to the paramagnetic phase is approached. A fit with

$$P(T) = P_0 \left( \frac{T_c - T}{T_c} \right)^\beta$$ (3.12)

yields an exponent $\beta$ of $0.25 \pm 0.05$, which is slightly larger than the critical exponent $\beta$ of $0.17 \pm 0.03$ found by Thomassen et al. in their analysis of thicker films [133]. The exponent thus deviates substantially from the value of $\frac{1}{2}$ predicted by mean-field theory and is larger than the value of $\frac{1}{8}$ obtained for the two-dimensional Ising model. The Curie temperature obtained by the fit is $T_c = 325.7 \pm 1.3$ K, which exceeds the Curie temperature obtained by visual inspection of the domain images by 1–2 K. Note that near the Curie temperature determined by visual inspection, the stationary domain pattern has given way to rapidly moving domains. Hence, it is not surprising to find a higher Curie temperature upon analyzing the behavior of the polarization. The fit is represented by the solid curve in Fig. 3.14. If $T_c$ is set to 324.2 K, which is the Curie temperature as determined by visual inspection of SEMPA images, a slightly smaller value of $\beta = 0.19 \pm 0.05$ is obtained.
3.7. MEAN-FIELD ASPECTS

3.7.2 Domain Width as a Function of Temperature

A strong dependence of the width of stripe domains on temperature was noticed by Allenspach and Bischof in their investigation of the domain pattern of low-temperature-grown ultrathin Fe films on Cu(001) \cite{5}. On approaching the reorientation temperature where the magnetization turns into the plane of the film, a marked decrease of the stripe width was detected. Czech and Villain calculated the domain width at 0 K and at the Curie temperature and found that the domain width at the Curie temperature was reduced to a finite value given by the exchange energy and the dipolar energy \cite{37}. Gehring and Keskin obtained an expression for the domain width over the whole temperature range \cite{162}. While the domain width was found to vary little at low temperatures, a sudden decrease appeared at higher temperatures. An exponential decrease of the domain width and a finite width at the Curie temperature were predicted by Kashuba and Pokrovsky \cite{3}.

Here, the domain width of ultrathin Fe films on Cu(001) will be examined both as a function of temperature and as a function of film thickness. Allenspach has noticed that the domain width depends crucially on preparation conditions \cite{163}. The films analyzed here were grown at 315 K. It will be shown that the domain width as a function of temperature near the Curie temperature is well described by Eq. (2.52). The variation of the domain width with film thickness can be explained with the concept of the effective temperature.

Fig. 3.15 shows the average domain width as a function of temperature in the vicinity of the Curie temperature. The domain width decreases with temperature and reaches a finite value on the order of half a micron at the Curie temperature. Such a finite value has been predicted by Czech and Villain \cite{37} and Kashuba and Pokrovsky \cite{74}. The solid line in Fig. 3.15 represents the best fit using Eq. (2.52):

\[
L_0(T, D) = \frac{\pi}{k_0(T, D)} \approx \frac{J + 2g\bar{\alpha}_0}{g} \left\{ 1 + 2 \left( \frac{\alpha_T}{\alpha_0} \right)^2 \left( \frac{T_c - T}{T_c} \right)^2 \right\} \frac{a^2}{D} .
\]

The fit was restricted to the data marked as black circles in Fig. 3.15, which cover the temperature range between approximately 0.9\cdot T_c and T_c. It yields a good agreement with the experimental data. The finite domain width at the Curie temperature obtained by the fit is 0.56\pm0.04 \mu m and corresponds to the domain width observed in the SEMPA images.\textsuperscript{24} The domain width at lower temperatures, however, deviates significantly from the fit. The value for the ratio \frac{T_c}{T} obtained is 6350 \pm 400 and the Curie temperature determined by the fit is \(T_c = 324.2 \pm 0.6 \) K, which coincides

\textsuperscript{24} Kashuba and Pokrovsky derived the expression \(4\pi\Gamma/\Omega\) for the finite domain width at the transition temperature \cite{74}. \(\Gamma\) is a measure of the exchange interaction and \(\Omega a\) is the strength of the dipolar interaction where \(a\) is the in-plane nearest-neighbor distance. Setting \(a\) to 0.25 nm, \(\Gamma\) to \(T_c \approx 300 \) K and \(\Omega a\) to 2.5 K, we obtain a domain width of approximately 300 nm.
perfectly with the Curie temperature obtained by visual inspection of the SEMPA image at the transition to the paramagnetic state. This is the temperature at which the coherent stripe domain pattern is lost. Above this temperature, some individual patches with stripe domains of the same period are still found in a paramagnetic matrix. The coefficient $\alpha_1$ substantially exceeds the coefficient $\alpha_0$, the ratio found by the fit is $\frac{\alpha_1}{\alpha_0} = -30.9 \pm 0.8$.

A fit to the data marked with black circles with the more general function

$$L_0(T) = a \cdot (T_c - T)^b + c$$  \hspace{1cm} (3.13)

yields an exponent of $b = 2.2 \pm 0.7$, which compares favorably with the value of 2 expected by mean-field theory, cf. Eq. (2.52). The domain width at $T_c$ calculated using the parameters determined in the fit with this more general function deviates only slightly from the domain width obtained using the fit with Eq. (2.52).

The variation of the domain width across a wedge at room temperature is displayed in Fig. 3.16. The domain width first increases and then decreases again as the film thickness in increased. A comparison with Fig. 3.11 reveals that the maximum domain width at room temperature occurs at the film thickness where the Curie temperature is highest. The graph in Fig. 3.16 does not comprise the whole
3.7. MEAN-FIELD ASPECTS

Figure 3.16: Domain width as a function of thickness. The domain width decreases as the effective temperature is increased. The average domain width at room temperature reaches its maximum where the Curie temperature is highest, cf. Fig. 3.11. In thinner or thicker films, room temperature corresponds to a higher effective temperature and the domain width decreases. The domain-width determination was carried out on a wedge. The whole magnetic thickness range that is magnetic at room temperature. While the whole magnetic thickness range is almost completely covered at the thin end, the domain width at room temperature decreases even further for larger film thicknesses at the thick end. In both cases, the domain width finally assumes a value on the order of half a micrometer, which is the value also found at the Curie temperature in Fig. 3.15. To summarize, the domain width decreases whenever the effective temperature is increased, be it by increasing the actual temperature or by moving along a wedge. It can thus be expected that these two procedures will produce similar effects. That this is indeed true will become apparent in Secs. 3.10 and 3.12.

25 In the multilayer systems (Fe/Ni)/Cu(001) and Co/Cu/(Fe/Ni)/Cu(001), the stripe domain width was determined to decay exponentially towards the spin reorientation transition [164, 165].
3.8 Domain-Width Adjustment by Dislocations

Due to their finite energy, cf. Subsec. 2.5.3, dislocations will be present in a stripe domain pattern at any finite temperature. They predominantly occur in pairs, Fig. 3.17a. In contrast, isolated disclination have a huge energy and only occur at elevated temperatures. Dislocations have been suggested as possible mediators for pattern transformations Sec. 2.7. While some transformations can indeed be interpreted in terms of dislocations, the role of dislocations in these transformations differs considerably from the one predicted theoretically. The mechanisms mediating pattern transformations are the subject of Sec. 3.12. Here, another important role of dislocations is analyzed.

Fig. 3.17b shows stripe domains on a wedge. The effective temperature rises from left to right. As reported in Subsec. 3.7.2, the stripe-domain width decreases as the effective temperature is increased. This reduction of the average domain width is mediated by dislocations. They allow for new stripe domains to be inserted as the Curie temperature is approached. A truly continuous variation of each individual stripe width is hampered by the geometry of the wedge and the intrinsic anisotropy favoring an orientation along one of the (100) directions. But even without the restriction of the wedge, the width of individual domains does not vary continuously when the temperature is changed.

Figs. 3.17c and 3.17d identify the important role dislocations play in adjusting the stripe-domain width as the temperature of an extended homogeneously thick film is lowered from a temperature close to the Curie temperature. The top of the images has the highest temperature. While the average domain width varies continuously, such a continuous variation of each single stripe domain in an extended sample would entail a continuous sideward migration over large distances of almost all the stripes present in the sample. Such a massive migration of domain walls is only thinkable in ideal, defect-free samples where the stripe pattern would form a true floating solid. Instead, the adjustment of the average domain width is again mediated by dislocations. Moving dislocations lead to the annihilation of stripe domains. Such a process is highlighted in Fig. 3.17c. Two stripe domains highlighted in red merge to form a single stripe domain as the temperature is lowered. This merger (or annihilation) occurs on a short time scale and is accompanied by a rapid zig-zag movement of the neighboring stripes. A number of such processes can be seen in Fig. 3.17d. The result of these annihilation processes is an increased stripe-domain width, as required by the free energy. Thus, dislocations are essential for adjusting the average domain width to that required by the free energy at a given temperature.

The important role of dislocations in adjusting the domain width has already been noticed by Kooy and Enz [20], and Seul and Wolfe [21].
3.8. DOMAIN-WIDTH ADJUSTMENT BY DISLOCATIONS

Figure 3.17: Domain-width adjustment by dislocations. Dislocations are essential for adjusting the average stripe-domain width as required by the free energy. a, Various defects composed of dislocations. Dislocations are present in stripe domain patterns at any finite temperature. b, Dislocations on a wedge. The effective temperature increases from left to right. Dislocations mediate the increase of the stripe density. c, Detailed image showing the annihilation of a stripe domain by a moving dislocation. The temperature decreases from top to bottom. The vertical axis of the image is stretched. d, Overview image showing the result of stripe annihilation by moving dislocations. The temperature decreases from top to bottom. The area enlarged in Fig. c is again highlighted. The moving dislocations reduce the number of stripes as the sample is cooled down (compare the number of stripes at the top and at the bottom).

\( w_a = 46 \, \mu m, \, w_b = 65 \, \mu m, \, w_c = 12 \, \mu m, \, w_d = 18.4 \, \mu m, \, D_a = 2.44 \, ML, \, D_b \) from 2.33 ML (left) to 2.12 ML (right), \( D_{c,d} = 196 \, ML, \, T_a = 293 \, K, \, T_b = 286 \, K, \, T_{c,d} \approx 295 \, K \)
Figure 3.18: Orientation of stripe domains on a flat substrate. An intrinsic anisotropy of ultrathin Fe films on Cu(001) orients stripe domains along a \( \langle 100 \rangle \) direction \([5, 74]\). All the subsequent domain images will have their vertical axis parallel to the \([110]\) direction. \((w = 92 \mu m, D = 2.12 \text{ ML}, T = 293 K)\)

3.9 Influence of the Substrate

Ultrathin Fe films deposited onto Cu(001) substrates at 300 K are characterized by stripe domains when observed at room temperature. This has already been noticed in experiments by Allenspach and Bischof \([5]\). Fig. 3.18 shows magnetic stripe domains on a flat Cu(001) substrate. The stripes are nicely aligned along a \( \langle 100 \rangle \) direction. The preferential orientation along a \( \langle 100 \rangle \) direction can be understood as due to higher order gradient terms of the exchange interaction \([74]\). The same preference was noticed by Allenspach and Bischof in their experiments \([5]\). In another experiment, however, a more uniform distribution of the stripe directions was found \([19]\). This wider distribution can be attributed to the use of a less perfect Cu(001) substrate.

The orientation of stripe domains in ultrathin Fe films is indeed very sensitive to topographic features of the underlying substrate. Fig. 3.19a shows the magnetic domain structure on a less perfect Cu(001) substrate. A comparison with the topography image in Fig. 3.19b reveals a rather strong correlation between the magnetic domain structure and topography. The magnetic stripe domains tend to be aligned along lines of equal height and encircle local elevations and depressions. Fig. 3.19c shows magnetic stripe domains in an ultrathin Fe film deposited onto a slightly miscut Cu(001) substrate. The miscut angle is 3° and produces a dense array of steps along the [110] direction. Fig. 3.19d shows some of the neatly aligned steps along the [110] direction. The magnetic stripe domains tend to follow the orientation of the substrate steps.

All the experiments reported in Part I of this thesis were done either on flat substrates or slightly miscut substrates, cf. Sec. 3.3. In the course of the experiments, it became soon clear that other orientations of magnetic stripe domains are possible. Stripe domains on the miscut substrate cannot only be oriented along the steps but also perpendicular to them. Moreover, under certain conditions, domain configu-
3.9. INFLUENCE OF THE SUBSTRATE

Figure 3.19: Influence of the substrate on stripe domains. In many cases, inhomogeneities of the substrate are mirrored in the domain pattern. 

- a, Domain pattern on a less perfect crystal. The domain walls follow lines of equal height.
- b, Topography of the area imaged in a.
- c, Domain pattern on a miscut crystal. The domain walls are parallel to the steps induced by the miscut.
- d, Topography of the area imaged in c.

(w_{a,b} = 46 \text{ ML}, w_{c,d} = 23 \text{ ML}, D_{a,b} = 2.67 \text{ ML}, D_{c,d} = 3.07 \text{ ML}, T_{a,b} = 293 \text{ K}, T_{c,d} = 294 \text{ K})

Rations more complicated than the simple stripe domains appear. These different phases will be introduced in Sec. 3.10. We will find that the intrinsic anisotropy favoring a domain wall orientation along a \langle 100 \rangle direction can lose its dominant role.
3.10 Sequence of Phases

The system of ultrathin Fe films on Cu(001) is highly sensitive to variations of
the preparation conditions, both with respect to ‘mean-field’ properties such as the
magnetization or the average domain width and with respect to the geometrical
aspects of the magnetic domain patterns. This section provides an overview of the
various domain patterns that occur in these films. A more detailed analysis of the
individual phases is given in Sec. 3.11 and the mechanisms mediating the transitions
between the individual phases are dealt with in Sec. 3.12.

The phases presented here occur in films with a thickness up to approximately
two monolayers. Due to the dependence of the Curie temperature on film thickness
discussed in Sec. 3.6, the effective temperature can be raised either by moving along a
wedge toward smaller thicknesses or by increasing the actual temperature. It will be
shown that the two ways of approaching the Curie temperature lead to an identical
sequence of domain patterns. While the overall features of the domain patterns occur
irrespective of whether the substrate crystal is flat or stepped, a stepped substrate
does impose certain domain wall orientations. The use of a stepped substrate allows
distinguishing phases that have identical properties on a flat substrate.

Ultrathin films grown at relatively low temperatures caused by long delay times
between annealing and evaporation are characterized by stripe domains throughout
the thickness range magnetic at room temperature. The films investigated here
have been produced with delay times on the order of 35–45 min and show more
complicated domain patterns.

3.10.1 Phases in Ultrathin Fe Films on a Flat Cu(001) Sub-
strate

The sequence of phases found on a wedge grown with a delay time of 45 min is shown
in Figs. 3.20a–d. The images were taken at room temperature. At low effective tem-
peratures, i.e. at a thickness where the Curie temperature is high, they show a stripe-
domain pattern, cf. Fig. 3.20a. Toward higher effective temperatures, i.e. smaller
thicknesses, the stripe-domain pattern transforms into a disordered pattern which we
will call the labyrinthine phase, cf. Fig. 3.20b. The domains are still characterized by
a well-defined domain width but the orientational order of the stripe-domain pattern
is lost. At higher effective temperatures, i.e. even smaller thicknesses, a reentrance of
the more ordered stripe-domain pattern occurs, cf. Fig. 3.20c. Two stripe-domain
patterns separated by a labyrinthine pattern can thus be distinguished. We will
call them the low-temperature and the high-temperature stripe-domain patterns
respectively. The terms ‘low-temperature’ and ‘high-temperature’ thereby refer to
the effective temperature rather than to the actual temperature. On increasing the
effective temperature even further, the stationary magnetic domain pattern is lost;
3.10. SEQUENCE OF PHASES

Figure 3.20: Sequence of phases in ultrathin Fe films on a flat Cu(001) substrate. The occurrence of the individual phases is governed by the effective temperature. The images show magnetic domain patterns found in films grown with a delay time between annealing and evaporation of 45 min. a–d, Sequence of phases on a wedge at room temperature. With increasing effective temperature, the low-temperature stripes (a) transform into the labyrinthine phase (b), which is then replaced by high-temperature stripes (c) before the paramagnetic phase (d) is reached. e–h, Sequence of phases upon heating. The domain pattern at a given location is shown at different temperatures. i–m, Sequence of phases upon cooling. This sequence is the reverse of the sequence observed upon heating. The images i–k display the domain pattern at a given location at different temperatures. The transition to the low-temperature stripe phase is rare and depicted in the images l and m. Again, a given location is imaged at different temperatures.

\[ \begin{align*}
    w_a &= 92 \text{ } \mu \text{m},
    w_b &= 46 \text{ } \mu \text{m},
    w_{c,d} &= 23 \text{ } \mu \text{m},
    w_e &= 46 \text{ } \mu \text{m},
    w_{f-g} &= 23 \text{ } \mu \text{m},
    w_h &= 23 \text{ } \mu \text{m},
    w_{i-j} &= 44 \text{ } \mu \text{m},
    w_{k,l} &= 33 \text{ } \mu \text{m},
    w_n &= 11.5 \text{ } \mu \text{m},
    D_a &= 2.1 \text{ ML},
    D_b &= 1.91 \text{ ML},
    D_c &= 1.84 \text{ ML},
    D_d &= 1.73 \text{ ML},
    D_{e-h} &= 1.89 \text{ ML},
    D_{l-k} &= 1.97 \text{ ML},
    D_{l,m} &= 1.94 \text{ ML},
    D_n &= 1.56 \text{ ML},
    T_{a-d} &= 293 \text{ K},
    T_e &= 210 \text{ K},
    T_f &= 268 \text{ K},
    T_g &= 283 \text{ K},
    T_h &= 313 \text{ K},
    T_i &= 321 \text{ K},
    T_j &= 300 \text{ K},
    T_k &= 296 \text{ K},
    T_l &= 294 \text{ K},
    T_m &= 281 \text{ K},
    T_n &= 154 \text{ K}
\end{align*} \]
it gives way to a contrastless phase which we identify as the paramagnetic phase. The image appears contrastless because the polarization at each pixel averages to zero within the measuring time of 10 ms.

Figs. 3.20 e–h reveal that the same sequence of phases occurs upon varying the actual temperature. A fixed location on a film is imaged as the temperature is increased. Again, a labyrinthine phase is found to separate high-temperature stripes from low-temperature stripes. The reverse sequence of phases is observed upon decreasing the temperature from a temperature above the Curie temperature. The first domain pattern that emerges as a film is cooled down from a temperature above the Curie temperature is the high-temperature stripe phase, cf. Fig. 3.20 i and j. It is followed by the labyrinthine phase, cf. Fig. 3.20 k. The formation of the labyrinthine pattern out of the high-temperature stripe phase is hampered by the large number of microscopic transformations necessary to establish the labyrinthine pattern and requires several hours. The transition from the labyrinthine phase to the low-temperature stripe-domain phase is rare. Figs. 3.20 l and m are evidence that this transition does occur occasionally. It is probably hindered by the slow kinetics of domain walls at low effective temperatures.

At low temperatures, a phase even more disordered than the labyrinthine phase is observed between the high-temperature stripes and the paramagnetic phase. This liquid phase is shown in Fig. 3.20 n. As in the labyrinthine phase, the orientational order is lost. Moreover, the domain width, which is well defined in the case of labyrinthine patterns, varies substantially and isolated domains are not rare. Such a liquid phase is not observed at room-temperature or even higher temperatures. The occurrence of the liquid phase is independent of the substrate temperature during evaporation.

3.10.2 Phases in Ultrathin Fe Films on a Slightly Miscut Cu(001) Substrate

The overall sequence of phases in ultrathin Fe films grown on a slightly miscut Cu(001) substrate corresponds to the sequence found in films grown on a flat Cu(001) substrate. The magnetic phases, however, are shifted towards larger thicknesses. Fig. 3.21 shows the evolution of the magnetic domains as a function of thickness. As in the case of films grown on a flat substrate, low-temperature stripe domains are followed by a labyrinthine pattern which then gives way to high-temperature stripes before the Curie temperature is reached. The uniaxial anisotropy of the stepped substrate is reflected in the orientation of the domain walls. Low-temperature stripes are

\[26\] The kinetics of domain walls seems to depend on the effective temperature rather than the actual temperature. Domains are found to become mobile both at elevated temperatures and room temperature whenever the effective temperature is close to unity.
Figure 3.21: Sequence of phases in ultrathin Fe films on a slightly miscut Cu(001) substrate I. a–c, Sequence of phases on a wedge. All the images were taken on the same wedge; they can be linked to form a continuous image showing the low-temperature stripes in thicker areas, the labyrinthine pattern, and the high-temperature stripes in thinner areas. The only exception is the area shaded in gray in a. The low-temperature stripes run along the steps of the substrate. The high-temperature stripes, however, are aligned perpendicular to the steps. The wedge was produced with a delay time of 45 min.

\[ w_{a-c} = 140 \, \mu m, \; D_a = 2.78 \, \text{ML} \text{ (left)} - 2.60 \, \text{ML} \text{ (right)}, \; D_b = 2.60 \, \text{ML} \text{ (left)} - 2.43 \, \text{ML} \text{ (right)}, \; D_c = 2.43 \, \text{ML} \text{ (left)} - 2.25 \, \text{ML} \text{ (right)}, \; T_{a-c} = 294 \, \text{K} \]

always oriented along the steps. The high-temperature stripes in Fig. 3.21c, however, run along the direction perpendicular to the steps. Such a difference in stripe orientation between low-temperature and high-temperature stripes does not occur on a flat substrate where the stripe orientation is given by an intrinsic anisotropy due to higher-order terms in the exchange energy [74].

With the orientation of low-temperature and high-temperature stripes in mind, the labyrinthine pattern could be interpreted as a transition region where the anisotropies favoring a parallel or perpendicular orientation of the domain walls are of the same order. In the labyrinthine phase, domain walls are aligned along one of the two mutually perpendicular directions. This view of the labyrinthine pattern, however, is refuted by the sequence of phases displayed in Fig. 3.22. The high-temperature
Figure 3.22: Sequence of phases in ultrathin Fe films on a slightly miscut Cu(001) substrate. The image shows the low-temperature stripes in thicker areas, the labyrinthine pattern, and the high-temperature stripes in thinner areas. Unlike in other overview images, the whole thickness range magnetic at room temperature is shown. The region of interest here is the region to the right of where the stripe domains have the largest period. Both low-temperature and high-temperature stripe domains are aligned along the steps of the substrate. The wedge was produced with a delay time of 85 min. ($w = 181 \, \mu m, D = 4.10 \, ML \text{ (left)} - 2.16 \, ML \text{ (right)}, T = 293 \, K$)

stripes on this wedge grown at slightly lower substrate temperatures are aligned parallel to the steps. In this case, the labyrinthine pattern separates two stripe-domain patterns with the same orientation. The two domain-wall directions occurring in the labyrinthine pattern can no longer be ascribed to a transition region sharing properties of both the low-temperature stripes and the high-temperature stripes. The origin for the occurrence of the labyrinthine pattern remains unresolved. Note that the orientation of the high-temperature stripes on a stepped substrate is reproducible. Elevated temperatures during the evaporation lead to an orientation perpendicular to the steps but growth at temperatures close to room temperature yield high-temperature stripes parallel to the steps. At sufficiently low substrate temperatures, the labyrinthine pattern vanishes altogether leaving behind a stripe domain pattern with uniform stripe orientation throughout the thickness range magnetic at room temperature. The use of a stepped substrate thus introduces a further distinction between low-temperature and high-temperature stripes. Not only are they separated by the labyrinthine pattern but they can also have a different orientation with respect to steps on the substrate.
3.11 Analysis of the Individual Phases

Like ultrathin Fe films grown on a flat substrate, films grown on a stepped substrate exhibit a liquid phase at low temperatures. This liquid evolves in the transition region between the high-temperature stripes and the paramagnetic phase. The transition from the high-temperature stripes to the liquid phase is depicted in Fig. 3.23. While the domain-wall orientations in all the other phases are dictated by the steps of the substrate, such an influence is absent in the liquid phase. The liquid phase occurs irrespective of the preparation conditions.

3.11 Analysis of the Individual Phases

While the individual phases occurring in ultrathin Fe films on Cu(001) can easily be distinguished by visual inspection alone, a more quantitative analysis is necessary when the individual phases are to be compared with phases predicted by theory. The overall symmetry of the phases is reflected in the Fourier transforms. But more specific information is obtained by calculating the positional and orientational correlations of the phases.

Both positional and orientational correlations are calculated along different directions. As a first step, the polarization of a given image is binarized, that is, a value of ±1 is assigned to each pixel of the image depending on the local orientation of...
Figure 3.24: Director field of magnetic domain patterns. In order to obtain the director field over a whole image area, the direction of the domain boundaries is determined and interpolated into the interior of the domains. 

(a) Director field of a labyrinthine pattern. For the sake of clarity, the director field is only shown for one of the two perpendicular magnetization directions. The color code is given in (d). 

(b) Director field of a $(0,-1)$ disclination of the labyrinthine pattern. The calculation of the director field yields an excellent agreement with the actual director field. 

(c) Director field of a $(0,1)$ disclination of the labyrinthine pattern. At the tip of the domain, the calculated director field rotates. This is due to the interpolation of the director field in the interior from the orientation of the domain wall. The actual director field does not rotate. 

(d) Color code. The orientation of the director field is color-coded. In contrast to vector fields, opposite directions are identified in director fields. 

(e) Stripe pattern with dislocations. 

(f) Director field of a stripe pattern with dislocations. The area shown is the same as the area depicted in (e). The dislocations are reflected in a characteristic configuration of the director field that stands out against the slowly varying director field of the undisturbed stripe pattern surrounding the dislocations. ($w_a = 46 \mu m$, $w_{b,c} = 4.5 \mu m$, $w_{e,f} = 8 \mu m$, $D_{a-c} = 1.97$ ML, $D_{e,f} = 2.17$ ML, $T_{a-c,e,f} = 293$ K)
the polarization. This reduces the attenuation of the positional correlation function
due to fluctuations of the primary electron beam of the scanning electron micro-
scope, and is necessary to compute the director field, which yields the orientational
correlation function. The binarization is obtained by applying a series of interdigi-
tated Gaussian convolutions and thresholding operations. In order to facilitate the
calculation of the director field, the number of pixels per image is increased from
$256 \times 256$ to $512 \times 512$ by interpolation.

The procedure for calculating the director field has been developed by Knowles and
is described in more detail in [166]. The angle of the director field in the interior of
the domains is interpolated from the orientation of the domain boundaries, which is
obtained by a generalized least-square algorithm. This algorithm minimizes the per-
pendicular distance between pixels belonging to the domain boundary and a straight
line fitted to the boundary. The angle of the director field at the boundary is then
given by the angle between the fitted line and a given reference axis.

The direction of the director field is visualized by using a color code. Fig. 3.24a
shows the director field of one of the two polarization orientations of a labyrinthine
pattern; the color code is presented in Fig. 3.24d. The direction of the domains
in the labyrinthine pattern is well reflected by the angle of the director field. The
director field of $(0,-1)$ and $(0,1)$ disclinations is depicted in Figs. 3.24c and d re-
spectively. Whereas the calculated director field gives an excellent representation of
the domain orientation at $(0,-1)$ disclinations, the interpolation of the director field
in the interior of the domains from the orientation of the domain boundaries induces
an artificial rotation of the director field at domain tips, that is at $(0,1)$ disclinations.
However, the total area of $(0,-1)$ disclinations in an image is small even in the case
of the labyrinthine pattern. The effect of those incorrect director-field configurations
at $(0,1)$ disclinations on the order parameter $G_n$ and the orientational correlations
seems negligible. Figs. 3.24e and f show the polarization and the director field of a
stripe pattern. The dislocations in the stripe pattern appear as conspicuous features
in the director field, cf. Fig. 3.24f.

The director field forms the basis for the calculation of the orientational correla-
tion function. Both positional and orientational correlation functions are calculated
in Cartesian and polar coordinates. Cartesian coordinates are used to give a two-
dimensional representation of the correlation, and polar coordinates are used to
obtain the correlation function along different directions. While a lot of pixel pairs
enter the determination of the correlation at small distances, the correlation at larger
distances is determined by the average of a smaller number of pairs. Consequently,
the error of calculated correlations increases with distance. The correlations in polar
coordinates are averaged over an angle range of approximately $6^\circ$ centered at the
chosen direction. The effect of this procedure on the positional correlation perpen-
dicular to the stripes is negligible. However, this procedure causes the positional
correlation along the stripes to decay with the inverse of the distance even in the
case of perfectly aligned stripe domains. This effect has to be considered when discussing the experimentally determined positional correlations. Orientational correlations are largely unaffected by this averaging procedure.

In order to distinguish true positional correlations from noise, the statistical error due to the limited size of the images has to be calculated. Each correlation $c$ in a given direction and at a given distance is obtained by counting the pixel pairs with positive and negative correlations. If this procedure is repeated in a large number of different images showing the same physical system, the different values for the correlation $c$ will be distributed around the expectation value $\bar{c}$. The distribution will be a binomial distribution.

Image pixels are arbitrary units. The statistical error cannot be reduced simply by multiplying the number of pixels per image. The calculation of the statistical error has to be based on some natural units. A natural choice for these natural units are squares with the side length of a domain width. Let $N$ be the number of pixel pairs and $L$ be the number of pixels per domain width. Then the number of natural units entering the calculation of the correlation is given by $n = N/L^2$. The binomial distribution for $p = \frac{\bar{c} + 1}{2}$ will be centered at $n \cdot \bar{p} = n \cdot \frac{\bar{c} + 1}{2}$ with a variance of $\sigma_p^2 = n \cdot p \cdot (1 - p)$. The standard deviation of $c$ will thus be

$$\sigma_c = \frac{2\sigma_p}{n} = \sqrt{\frac{1 - c^2}{n}} = \sqrt{\frac{1 - c^2}{N/L^2}}.$$  \hspace{1cm} (3.14)

It is largest for $c = 0$. The number of pixel pairs $N$ depends on the number $N$ of pixels per image side and the number $d$ of pixels across which the correlation is determined. For correlations parallel to a side of an image, $N$ is given by:

$$N(N, d) = N(N - d).$$  \hspace{1cm} (3.15)

For correlations along the diagonal of an image, $N$ is given by:

$$N(N, d) = \left( N - \frac{d}{\sqrt{2}} \right)^2.$$  \hspace{1cm} (3.16)

### 3.11.1 Low-Temperature Stripe Phase

Low-temperature stripes in films grown on flat Cu(001) substrates are aligned along one of the $\langle 100 \rangle$ directions. The polarization image and the director field of such stripes are depicted in Figs. 3.25a and c. The dislocations in the stripe pattern are clearly visible as conspicuous features in the director field. Moreover, the director field reveals the meandering of the stripes. This is evidenced by the bands of homogeneous orientation running perpendicular to the average stripe direction. In films grown on slightly miscut substrates, low-temperature stripes are aligned along the
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Figure 3.25: Low-temperature stripe phase. a, Low-temperature stripe phase on a flat Cu(001) substrate. b, Low-temperature stripe phase on a flat Cu(001) substrate with additional anisotropy. c, Director field of a low-temperature stripe phase on a flat Cu(001) substrate. The area shown is the same as the area depicted in a. d, Director field of a low-temperature stripe phase on a flat Cu(001) substrate with additional anisotropy. The area shown is the same as the area depicted in b. \( w_{a-d} = 46 \, \mu m, \, D_{a,c} = 2.05 \, \text{ML}, \, D_{b,d} = 2.44 \, \text{ML}, \, T_{a-d} = 293 \, \text{K} \)

direction of the steps induced by the miscut. Such an in-plane anisotropy for the orientation of domain walls is occasionally also found locally in films grown on flat substrates. An example is shown in Figs. 3.25 b and d. Again, the dislocations in the
The Fourier transforms of Figs. 3.25a and b are depicted in Figs. 3.26a and b respectively. In both cases, the Fourier transforms show two sharp spots. The limited extent of the spots is an indication that the stripe domain width is well-defined. Positional and orientational correlations calculated from the images in Fig. 3.25 are given in Fig. 3.27. The modulation of the positional correlation is preserved over large distances. After an initial drop, the orientational correlation reaches a finite value.

The positional correlations perpendicular and parallel to the stripe direction of the film grown on a flat substrate without additional in-plane anisotropy are presented in Figs. 3.28a and b respectively. The modulation of the oscillation amplitude of the perpendicular correlation is indicative of a beat. Indeed, the Fourier transform of the positional correlation perpendicular to the stripe direction shows two narrowly spaced sharp peaks. Using the two corresponding wavelengths, the experimental
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Figure 3.27: Correlations in the low-temperature stripe phase. a, Positional correlation of Fig. 3.25a. b, Positional correlation of Fig. 3.25b. c, Orientational correlation of Fig. 3.25a. d, Orientational correlation of Fig. 3.25b. (w_{a-d} = 46 \mu\text{m}, D_{a,c} = 2.05 ML, D_{b,d} = 2.44 ML, T_{a-d} = 293 K)

Data are fitted with a modified version of Eq. (2.156):

\[ g_m(x, 0) = A \left\{ a_1 \cos \left( 2\pi \frac{x}{\lambda_1} + \varphi_1 \right) + a_2 \cos \left( 2\pi \frac{x}{\lambda_2} + \varphi_2 \right) \right\} \cdot e^{-\sqrt{x_\perp}}. \]  

(3.17)
perpendicular & parallel \\
\hline
\(a_1\) & 0.0494 & \(A\) & 10.5 ± 0.5 & \(A\) & 1.101 ± 0.005 \\
\(a_2\) & 0.0695 & \(\xi_\perp [\mu m]\) & 4.00 ± 0.03 & \(\xi_\parallel [\mu m]\) & 5.44 ± 0.05 \\
\(\varphi_1\) & 0.363 & & & & \\
\(\varphi_2\) & 0.282 & & & & \\
\(\lambda_1 [\mu m]\) & 2.65 & & \(\lambda_3 [\mu m]\) & 32.4 ± 0.2 & \\
\(\lambda_2 [\mu m]\) & 2.25 & & & & \\
\hline
\(R^2\) & 0.905 & & \(R^2\) & 0.995 & \\
\hline

Table 3.2: Fits to positional correlations in low-temperature stripes. The parameters are obtained by fitting Eqs. (3.17) and (3.18) to the experimental data shown in Figs. 3.28a and b respectively.

The amplitudes \(a_1\) and \(a_2\), the wavelengths \(\lambda_1\) and \(\lambda_2\), as well as the phases \(\varphi_1\) and \(\varphi_2\) are obtained from the Fourier transform of the positional correlation perpendicular to the stripe direction. The only parameters that need to be fitted are the amplitude \(A\) and the correlation length \(\xi_\perp\), which is found to amount to 4.00 ± 0.03 \(\mu m\). An overview of the parameter values obtained in the fit is given in Tab. 3.2. The fitted curve is displayed in Fig. 3.28a as a gray line. It is in good agreement with the experimental data, given the fact that only the two most prominent contributions to the Fourier transform are retained. The presence of two slightly different wavelengths can be attributed to the meandering of the stripes which leads to varying periodicities along the average stripe direction. If the square-root dependence of the exponent in Eq. (3.17) is relaxed to the more general power law of \(-x^d\), the fit yields an exponent \(d = 0.59 ± 0.04\), which is close to the theoretically expected value of 0.5. The quality of the fit is only insignificantly improved by this adjustment. The positional correlation perpendicular to the stripes can be described by the functional form predicted for a two-dimensional smectic.

The positional correlation parallel to the stripes is given in Fig. 3.28b. It decays more quickly than the positional correlation perpendicular to the stripes in Fig. 3.28a. This is to be expected from Eq. (2.151), which predicts an exponential decay of the correlation function parallel to the stripes. However, the experimentally observed decay is not monotonous. Instead, the decay is modulated. This modulation is not
apparent from Eq. (2.156), which relies on the ansatz of Eq. (2.80). This ansatz
with a well-defined direction of the magnetization modulation is too simple to yield
the beat observed in the positional correlation perpendicular to the stripes, which
is caused by the meandering of the stripes. Meandering means a slowly varying
direction of the magnetization modulation and thus also leads to a modulation of
the positional correlation parallel to the stripes. It turns out that this modulation
of the correlation can be accounted for by the following modification of Eq. (2.151):

\[ g_m(0, y) = A \cos^2 \left( \frac{2\pi y}{\lambda_3} \right) \cdot e^{-\frac{y}{\xi_\parallel}}. \]  

(3.18)

The corresponding fit is shown as a gray line in Fig. 3.28b. The fit yields \( \xi_\parallel = 5.44 \pm 0.05 \) \( \mu \)m. If the linear dependence of the exponent in Eq. (3.18) is replaced
with the more general power law of \( -y^d \), the fit improves slightly and yields an
exponent \( d = 1.13 \pm 0.02 \) \( \mu \)m. The positional correlation parallel to the stripes
can be described by the functional form predicted for a two-dimensional smectic
supplemented with a modulation. Note, however, that the correlation lengths \( \xi_\perp \)
and \( \xi_\parallel \) obtained in the fit do not fulfill Eq. (2.154). This equation correlates
\( \xi_\perp \) and \( \xi_\parallel \) with \( \lambda \). Using the experimentally found values for the correlation lengths,
a value of \( \lambda \approx 5.7 \) \( \mu \)m is obtained. Such a value corresponds to a stripe width of
\( L = 4\pi \lambda = 72 \) \( \mu \)m, which is nearly two orders of magnitude larger than the actual
stripe width.

In Subsec. 2.6.3, the influence of an anisotropy energy due to higher order gradi-
ent terms of the exchange energy on the positional correlation is investigated. The
anisotropy energy is found to become effective at large distances; the positional
correlation functions are then given by power laws. According to Eq. (2.201), the
correlation along the direction perpendicular to the stripes is stronger than the cor-
relation parallel to the stripes for not too large anisotropies. This coincides with the
experimental findings. However, the functional form of the positional correlation
functions for smectics without anisotropy is sufficient to describe the correlations
over the distances covered experimentally. But after the strong initial decay, the
functional form expected for a smectic without anisotropy decays somewhat faster
than the experimentally determined correlation. The positional correlation can be
fitted slightly better by a power law, the functional form expected for a smectic
with anisotropy. A definite statement is difficult because of the irregularities of the
oscillations. Anisotropy should become important at larger distances.

Fits to the orientational correlations perpendicular and parallel to the stripes us-
ing the approximations developed in Subsec. 2.6.2 are presented as gray lines in
Figs. 3.29a and b respectively. The function fitted to the orientational correlation
Table 3.3: Fits to orientational correlations in low-temperature stripes. The parameters are obtained by fitting Eqs. (3.19) and (3.20) to the experimental data shown in Figs. 3.29a and b respectively.

<table>
<thead>
<tr>
<th></th>
<th>perpendicular</th>
<th>parallel</th>
</tr>
</thead>
<tbody>
<tr>
<td>$G_2$</td>
<td>0.901 ± 0.001</td>
<td>0.907 ± 0.001</td>
</tr>
<tr>
<td>$k_B T / B [\mu m^2]$</td>
<td>0.0020 ± 0.0003</td>
<td>14 ± 2</td>
</tr>
<tr>
<td>$\lambda [\mu m]$</td>
<td>0.046 ± 0.005</td>
<td>30 ± 5</td>
</tr>
<tr>
<td>$q_{max} [\mu m^{-1}]$</td>
<td>7.9 ± 0.4</td>
<td>35 ± 5</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.912</td>
<td>0.893</td>
</tr>
</tbody>
</table>

perpendicular to the stripes is

$$g_2(x, 0) = G_2^2 \cdot \exp \left( \frac{k_B T}{B} \left\{ \frac{\text{erf} \left( \frac{q_{max} \sqrt{x}}{\sqrt{\pi} \lambda \sqrt{x}} \right)}{\sqrt{\pi} \lambda \sqrt{x}} + \frac{\sin(q_{max})}{2\pi^2 \lambda x} \{A(q_{max}, \lambda) + 2B(q_{max}, \lambda)\} \right\} \right),$$

and the function fitted to the orientational correlation parallel to the stripes is

$$g_2(0, y) = G_2^2 \cdot \exp \left( \frac{k_B T}{B} \left\{ \frac{2 \sin \left( \frac{q_{max} \sqrt{y}}{\pi \lambda y} \right)}{\pi \lambda y} e^{-\frac{q_{max}^2 y}{2\pi^2 \lambda y}} + \frac{4 \sin(q_{max} y)}{\pi^2 \lambda y} C(q_{max}, \lambda) \right\} \right).$$

In both cases the approximated orientational correlation functions yield a good agreement with the experimental data. The value of $G_2$ obtained from the perpendicular correlation is 0.901, the value obtained from the parallel correlation is 0.907. Both values are close to 0.904, the value of $G_2$ obtained directly by averaging over the director field. However, the two fits do not agree on the other parameters that appear in the correlation functions. Tab. 3.3 gives an overview of the parameters obtained by the fits. While the correlation perpendicular to the stripes yields a reasonable value for $\lambda$ of 0.046 ± 0.005 µm, the correlation parallel to the stripes gives the much larger value of 30 ± 5 µm. The correlation perpendicular to the stripes gives a reasonable value for $q_{max}$ of 7.9 ± 0.4 µm⁻¹. The product $\lambda \cdot q_{max} = 0.36$ justifies the choice of the cut-off $q_{max}$ made in Subsec. 2.6.2. The orientational correlation perpendicular to the stripes is thus well described by Eq. (3.19).
The values of $\lambda = 30 \pm 5 \mu m$ and $q_{\text{max}} = 35 \pm 5 \mu m^{-1}$ obtained from the correlation parallel to the stripes, however, yield a product of more than 1000. Finally, orientational correlations perpendicular and parallel to the stripes are also at odds concerning the value of $k_B T/B$, the correlation perpendicular yields a value of $0.002 J/\mu m^{-2}$, the correlation parallel to the stripes a value of $14 J/\mu m^{-2}$.

Figs. 3.30 and 3.31 show the positional and orientational correlations in a film grown onto a flat substrate that, locally, has an additional anisotropy for the orientation of the domain walls. The Fourier transform of the positional correlation perpendicular to the stripes, Fig. 3.30 a, shows a large number of peaks, which contrasts with the two well-defined peaks found in films grown onto a flat substrate without additional anisotropy for the domain walls. Consequently, the positional correlation perpendicular to the stripes cannot be fitted with an approximation only containing a small number of Fourier modes. The wiggles in the positional correlation parallel to the stripes produced by meandering do not show up in films grown onto a substrate with an additional anisotropy for the orientation of the domain walls. Note that the positional correlation perpendicular to the stripes is significantly larger than the standard deviation $\sigma$ obtained for the case of zero correlation. The positional correlation parallel to the stripes, however, quickly falls below $\sigma$. On a flat substrate, the parallel correlation falls below $\sigma$ after the first bump, on a stepped substrate it is below $\sigma$ right after the initial drop. When the parallel correlation is divided by the corresponding correlation of perfect stripes, it becomes of the order of $\sigma$. It is very noisy.

In summary, the functional form of the correlation functions derived in Subsec. 2.6.2 for two-dimensional smectics can be fitted to the experimental data. However, the fits yield parameters that conflict with each other. The elastic free energy of Subsec. 2.6.2 thus fails to describe the stripe domains adequately. At larger distances, the positional correlation perpendicular to the stripes seems to decay more slowly than expected for a smectic without anisotropy. Meandering leads to the presence of two narrowly spaced wavelengths for the positional correlation perpendicular to the stripes. In addition, meandering causes modulations in the positional correlation parallel to the stripes. If the substrate provides an additional anisotropy for the orientation of the domain walls, meandering is hindered.
Figure 3.28: Positional correlations in the low-temperature stripe phase on a flat substrate. a, Positional correlation of Fig. 3.25a perpendicular to the stripe direction. b, Positional correlation of Fig. 3.25a parallel to the stripe direction. The black lines represent the experimental data; the gray lines are fits to the experimental data using Eqs. (3.17) and (3.18) respectively. The dotted line represents the correlation of perfect stripes of the same width. The standard deviation $\sigma_0$ for zero correlation is given by the dashed lines. ($D_{a,b} = 2.05 \text{ ML}, T_{a,b} = 293 \text{ K}$)
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Figure 3.29: Orientational correlations in the low-temperature stripe phase on a flat substrate. a, Orientational correlation of Fig. 3.25a perpendicular to the stripe direction. b, Orientational correlation of Fig. 3.25a parallel to the stripe direction. The black lines represent the experimental data; the gray lines are fits to the experimental data using Eqs. (3.19) and (3.20). The dotted lines indicate the value of $G_2^2$. ($D_{a,c} = 2.05$ ML, $D_{b,d} = 2.44$ ML, $T_{a-d} = 293$ K)
Figure 3.30: Positional correlations in the low-temperature stripe phase with additional anisotropy. a, Positional correlation of Fig. 3.25b perpendicular to the stripe direction. b, Positional correlation of Fig. 3.25b parallel to the stripe direction. The dotted line represents the correlation of perfect stripes of the same width. The standard deviation $\sigma_0$ for zero correlation is given by the dashed lines. ($D_{a,b} = 2.44$ ML, $T_{a,b} = 293$ K)
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Figure 3.31: Orientational correlations in the low-temperature stripe phase with additional anisotropy. a, Orientational correlation of Fig. 3.25b perpendicular to the stripe direction. b, Orientational correlation of Fig. 3.25b parallel to the stripe direction. The dotted lines indicate the value of $G_2^2$. ($D_{a,c} = 2.44$ ML, $D_{b,d} = 2.44$ ML, $T_{a-d} = 293$ K)
3.11.2 Labyrinthine Phase

The long-range orientational order of both low-temperature and high-temperature stripes is lost in the labyrinthine phase. A characteristic feature of the labyrinthine phase are disclinations. Unlike dislocations, which are present in the stripe phases, disclinations break long-range orientational order. Figs. 3.32a and c show the domain configuration and the director field of a labyrinthine phase grown on a flat Cu(001) substrate. The branches of (0, −1) disclinations in such films tend to enclose angles of $2\pi/3$, a typical (0, −1) disclination is highlighted in the inset of Fig. 3.32a. The direction of one of the branches typically coincides with the direction of stripes grown onto the same substrate, i.e. the ⟨100⟩ direction. The symmetry of the labyrinthine phase is revealed by the Fourier transform in Fig. 3.33a. The intensity of the Fourier transform is concentrated within a narrow, slightly hexagonally distorted ring. The two-fold symmetry of the stripe phases is replaced by a weak six-fold symmetry in the labyrinthine phase. The domain width, however, remains well-defined.

The domain configuration and the director field of a labyrinthine phase grown onto a stepped Cu(001) substrate are displayed in Figs. 3.32b and d. The effect of the substrate-induced anisotropy is that possible directions for domain segments are restricted to the directions perpendicular and parallel to the steps of the substrate. Consequently, the branches of (0, −1) disclinations enclose either an angle of $\pi/2$ or $\pi$. A typical (0, −1) disclination is shown in the inset of Fig. 3.32b. The Fourier transform reflects this anisotropy. The intensity is concentrated in the corners of a square, cf. Fig. 3.33b. Again, the domain width remains well-defined.

The director field of the labyrinthine pattern breaks up into patches within which the director field is uniform. It changes abruptly across the boundaries separating the patches. The order within such patches in a lamellar system can be described by a local wave vector $\vec{k}$ defined as the local phase gradient $\nabla \theta$ [167]. Within this framework, two types of boundaries can be distinguished. Phase grain boundaries are boundaries across which the phase is continuous but the wave vector changes its direction. Such a case is shown in Fig. 3.34 and marked by a black line. No disclinations are involved in forming such a boundary. The other type of patch boundaries are amplitude grain boundaries. Such boundaries consist of arrays of disclinations that separate areas of different wave vectors. An example is marked by a white line in Fig. 3.34. The patch boundaries experimentally observed can, however, also be a mixture of the two types.

Positional and orientational correlations of the labyrinthine pattern are given in Fig. 3.35. They have been calculated from the images in Fig. 3.32. The symmetry of the labyrinthine phase, weakly six-fold on a flat substrate and four-fold on a stepped substrate, is also reflected in the positional correlation. In the labyrinthine phase grown on the flat substrate, the preferential direction of stripes grown on the
3.11. Analysis of the Individual Phases

Figure 3.32: Labyrinthine phase. The labyrinthine pattern is characterized by a high density of disclinations. 

a, Labyrinthine phase on a flat Cu(001) substrate. The branches of $(0, -1)$ disclinations typically enclose an angle of $\approx 2\pi/3$ (see inset). 
b, Labyrinthine phase on a stepped Cu(001) substrate. The branches of $(0, -1)$ disclinations typically enclose an angle of $\approx \pi/2$ or $\approx \pi$ (see inset). 
c, Director field of a labyrinthine phase on a flat Cu(001) substrate. The area shown is the same as the area depicted in a. 
d, Director field of a labyrinthine phase on a stepped Cu(001) substrate. The area shown is the same as the area depicted in b. ($w_{a-d} = 92 \mu m$, $D_{a,c} = 1.97$ ML, $D_{b,d} = 2.15$ ML, $T_{a-d} = 293$ K)

same substrate is still a preferential direction for domain segments. This becomes apparent when correlations along different directions are compared. Figs. 3.36 a and
Figure 3.33: Fourier transform of the labyrinthine phase. The labyrinthine pattern is characterized by a well-defined domain width. This is reflected in the Fourier transform, where the intensity is concentrated in a ring of limited width. On a stepped substrate, the domains are either parallel or perpendicular to the miscut-induced steps of the substrate. Consequently, the ring assumes the shape of a square. On a flat substrate, however, no such limitations apply, and the ring resembles a hexagon. a, Modulus of the Fourier transform of Fig. 3.32a. b, Modulus of the Fourier transform of Fig. 3.32b. \(D_a = 1.97\) ML, \(D_b = 2.15\) ML, \(T_{a,b} = 293\) K.

b display the positional correlations perpendicular and parallel to the preferential direction of stripes. As can be expected from the symmetry of the labyrinthine phase, positional correlations along either direction are characterized by a rapidly decaying oscillation. The dip in the positional correlation perpendicular to the preferential direction of stripes, however, is clearly more pronounced. It is not surprising that the intrinsic anisotropy for the orientation of domain walls along a \(\langle 100 \rangle\) direction in stripe phases plays a residual role in the formation of the labyrinthine pattern. On the stepped substrate, a similar difference is found between positional correlations along one of the two preferential directions and positional correlations along the bisector. These correlations are displayed in Figs. 3.39a and b. The orientational correlations in films grown either on flat or stepped substrates, however, seem to be isotropic. They vanish already after a short distance. Orientational correlations in the film grown on the flat substrate are shown in Figs. 3.37a and b, the corresponding curves for the film grown on the stepped substrate are displayed in Figs. 3.40a and b.

Due to the strong noise found in both positional and orientational correlations in the
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Figure 3.34: Amplitude and phase grain boundaries. The image shows a section of Fig. 3.32c overlaid on a black-and-white image of the domain configuration. The patches in the director field of the labyrinthine pattern are separated by amplitude grain boundaries (white line) and phase grain boundaries (black line). \( w = 18 \, \mu m, \, D = 1.97 \, ML, \, T = 293 \, K \)

labyrinthine phase along a given direction, angle-averaged correlations are used to determine the kind of decay that best describes positional and orientational correlations. Angle-averaged positional and orientational correlations for the film grown on the flat substrate are shown in Figs. 3.38a and b; angle-averaged correlations for the film grown on the stepped substrate are given in Figs. 3.41a and b.

A number of approaches have been used in an attempt to characterize correlations in the labyrinthine phase. The positional correlations have been fitted with modulated power-laws:

\[
g_m(r) = \left( \frac{A}{r + r_0} \right)^\kappa \cdot \cos \left( 2\pi \frac{r}{R} + \varphi_0 \right) + M , \tag{3.21}
\]

modulated exponential functions:

\[
g_m(r) = A \cdot \exp \left( -\frac{r}{r_0} \right) \cdot \cos \left( 2\pi \frac{r}{R} + \varphi_0 \right) + M , \tag{3.22}
\]

and modulated modified exponential functions of the kind used in the analysis of the positional correlations in the stripe phase:

\[
g_m(r) = A \cdot \exp \left( - \left( \frac{r}{r_0} \right)^\kappa \right) \cdot \cos \left( 2\pi \frac{r}{R} + \varphi_0 \right) + M . \tag{3.23}
\]
Figure 3.35: Correlations in the labyrinthine phase. The order of labyrinthine patterns on flat and stepped Cu(001) substrates is similar, but the stepped substrate only allows domains to be oriented parallel or perpendicular to the miscut-induced steps of the substrate. a, Positional correlation of Fig. 3.32a. b, Positional correlation of Fig. 3.32b. c, Orientational correlation of Fig. 3.32a. d, Orientational correlation of Fig. 3.32b. \((w_{a-d} = 92 \text{ } \mu\text{m}, D_{a,c} = 1.97 \text{ } \text{ML}, D_{b,d} = 2.15 \text{ } \text{ML}, T_{a-d} = 293 \text{ } \text{K})\)
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<table>
<thead>
<tr>
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<th>flat substrate</th>
<th>stepped substrate</th>
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<tbody>
<tr>
<td>$A$</td>
<td>$2.13 \pm 0.03$</td>
<td>$1.58 \pm 0.02$</td>
</tr>
<tr>
<td>$r_0$ [$\mu$m]</td>
<td>$0.77 \pm 0.02$</td>
<td>$1.03 \pm 0.02$</td>
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<tr>
<td>$\kappa$</td>
<td>$0.60 \pm 0.01$</td>
<td>$0.80 \pm 0.01$</td>
</tr>
<tr>
<td>$R$ [$\mu$m]</td>
<td>$3.68 \pm 0.01$</td>
<td>$3.53 \pm 0.02$</td>
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<tr>
<td>$\varphi_0$</td>
<td>$-1.08 \pm 0.01$</td>
<td>$-0.93 \pm 0.01$</td>
</tr>
<tr>
<td>$M$</td>
<td>$0.0020 \pm 0.0002$</td>
<td>$0.0316 \pm 0.0004$</td>
</tr>
<tr>
<td>$R^2$</td>
<td>$0.998$</td>
<td>$0.998$</td>
</tr>
</tbody>
</table>

Table 3.4: Fits to angle-averaged positional correlations in labyrinthine patterns. The parameters are obtained by fitting Eq. (3.23) to the experimental data shown in Figs. 3.38a and 3.41a respectively.

It turns out that Eq. (3.23) yields the best fit, whether a phase $\varphi_0$ is used or not. Without a phase $\varphi_0$, Eq. (3.23) is clearly the best choice. The power-law ansatz decays too slowly and the unmodified modulated exponential cannot reproduce more than the first minimum. However, the goodness of the fits with a phase $\varphi_0$ is close to each other so that a definite distinction is difficult. Figs. 3.38a and 3.41a show the fitted curves obtained using Eq. (3.23) as gray lines. The fitted curves nicely reproduce the experimentally determined positional correlations. The values of the various parameters obtained by means of the fits are given in Tab. 3.4 and of the same order for labyrinthine phases both on a flat substrate and a stepped substrate. Note that in both cases the phase $\varphi_0$ deviates substantially from zero.

Angle-averaged orientational correlations for the labyrinthine phase on a flat and a stepped substrate are shown in Figs. 3.38b and 3.41b respectively. The experimentally obtained angle-averaged orientational correlations have been fitted with a similar set of functions, i.e. a power law decay:

$$g_2(r) = \left( \frac{A}{r + r_0} \right)^\mu + c,$$

an exponential decay:

$$g_2(r) = A \cdot \exp \left( -\frac{r}{r_0} \right) + c,$$
Table 3.5: Fits to angle-averaged orientational correlations in labyrinthine patterns. The parameters are obtained by fitting Eq. (3.26) to the experimental data shown in Figs. 3.38b and 3.41b respectively.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Flat Substrate</th>
<th>Stepped Substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$0.977 \pm 0.006$</td>
<td>$0.951 \pm 0.005$</td>
</tr>
<tr>
<td>$r_0$ [$\mu$m]</td>
<td>$1.94 \pm 0.01$</td>
<td>$1.37 \pm 0.01$</td>
</tr>
<tr>
<td>$\mu$</td>
<td>$1.40 \pm 0.02$</td>
<td>$1.40 \pm 0.02$</td>
</tr>
<tr>
<td>$c$</td>
<td>$0.0070 \pm 0.0003$</td>
<td>$0.0322 \pm 0.0003$</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.997</td>
<td>0.996</td>
</tr>
</tbody>
</table>

In contrast to the case of angle-averaged positional correlations, no cosine-like modulation is observed. Again, the modified exponential function delivers the best fit to the experimental data. The values for the parameters obtained by the fit are given in Tab. 3.5. Both in the labyrinthine phase on the flat substrate and the labyrinthine phase on the stepped substrate, the exponent $\mu$ is found to be $1.40 \pm 0.02$. A very similar exponent is found in the liquid phase, cf. Subsec. 3.11.4. The orientational correlations of these orientationally disordered phases seem to be described by the same function. However, a model free energy producing this functional form for the orientational correlation is not known to the author.

In summary, the orientational order of the labyrinthine phase is short-ranged. However, the labyrinthine phase is not isotropic. Depending on the substrate, the symmetry of the labyrinthine phase is either slightly six-fold or fourfold. In either case the domain width remains well defined. Both angle-averaged positional and orientational correlations are best described by a modified exponential function. The orientational correlation is very similar to the orientational correlation found in the liquid phase. The domain pattern of the labyrinthine phase breaks up into patches of uniformly oriented domain sections separated by either phase grain boundaries or amplitude grain boundaries.
3.11. ANALYSIS OF THE INDIVIDUAL PHASES

Figure 3.36: Positional correlations in the labyrinthine phase on a flat substrate. a, Positional correlation of Fig. 3.32a perpendicular to the preferential direction of stripe domains. b, Positional correlation of Fig. 3.32a parallel to the preferential direction of stripe domains. The standard deviation $\sigma_0$ for zero correlation is given by the dashed lines. ($D_{a,b} = 1.97$ ML, $T_{a,b} = 293$ K)
Figure 3.37: Orientational correlations in the labyrinthine phase on a flat substrate. a, Orientational correlation of Fig. 3.32a perpendicular to the preferential direction of stripe domains. b, Orientational correlation of Fig. 3.32a parallel to the preferential direction of stripe domains. The dotted lines indicate the value of $G_2^2$. ($D_{a,b} = 1.97$ ML, $T_{a,b} = 293$ K)
Figure 3.38: Angle-averaged correlations in the labyrinthine phase on a flat substrate. 
(a) Positional correlation of Fig. 3.32a perpendicular to the preferential direction of stripe domains. b, Orientational correlation of Fig. 3.32a parallel to the preferential direction of stripe domains. The black lines represent the experimental data; the gray lines are fits to the experimental data. The dotted line indicates the value of $G_2^2$. ($D_{a,b} = 1.97$ ML, $T_{a,b} = 293$ K)
Figure 3.39: Positional correlations in the labyrinthine phase on a stepped substrate. 
a. Positional correlation of Fig. 3.32b an angle of $\frac{\pi}{4}$ off the direction of low-temperature stripes. b. Positional correlation of Fig. 3.32b parallel to the direction of low-temperature stripes. The standard deviation $\sigma_0$ for zero correlation is given by the dashed lines. ($D_{a,b} = 1.97$ ML, $T_{a,b} = 293$ K)
3.11. ANALYSIS OF THE INDIVIDUAL PHASES

Figure 3.40: Orientational correlations in the labyrinthine phase on a stepped substrate. a, Orientational correlation of Fig. 3.32b an angle of $\frac{\pi}{4}$ off the direction of low-temperature stripes. b, Orientational correlation of Fig. 3.32b parallel to the direction of low-temperature stripes. The dotted lines indicate the value of $G_2^2$. ($D_{a,b} = 1.97$ ML, $T_{a,b} = 293$ K)
Figure 3.41: Angle-averaged correlations in the labyrinthine phase on a stepped substrate. a, Positional correlation of Fig. 3.32b an angle of $\frac{\pi}{4}$ off the direction of low-temperature stripes. b, Orientational correlation of Fig. 3.32b parallel to the direction of low-temperature stripes. The black lines represent the experimental data; the gray lines are fits to the experimental data. The dotted line indicates the value of $G_2^2$. ($D_{a,b} = 1.97$ ML, $T_{a,b} = 293$ K)
Figure 3.42: High-temperature stripe phase. a, High-temperature stripe phase on a flat Cu(001) substrate. b, High-temperature stripe phase on a stepped Cu(001) substrate. c, Director field of a high-temperature stripe phase on a flat Cu(001) substrate. The area shown is the same as the area depicted in a. d, Director field of a high-temperature stripe phase on a stepped Cu(001) substrate. The area shown is the same as the area depicted in b. (w_{a,c} = 46 \, \mu m, w_{b,d} = 23 \, \mu m, D_{a,c} = 1.95 \, \text{ML}, D_{b,d} = 2.11 \, \text{ML}, T_{a,c} = 320 \, \text{K}, T_{b,d} = 293 \, \text{K})

3.11.3 High-Temperature Stripe Phase

The high-temperature stripe phase is very similar to the low-temperature stripe phase. High-temperature as well as low-temperature stripes in films grown on a
flat substrate run along a \(\langle 100 \rangle\) direction. While low-temperature stripes in films grown on a stepped substrate are aligned parallel to the miscut-induced steps along the \([110]\) direction, the orientation of high-temperature stripes depends on the substrate temperature during the growth of the film. Elevated temperatures produce stripes that run perpendicular to the steps of the substrate. When the substrate is kept at a temperature near room-temperature during evaporation, however, the high-temperature stripes assume the same orientation as low-temperature stripes, i.e. they are parallel to the miscut-induced steps of the substrate. Figs. 3.42a and b show the high-temperature stripe phase on a flat and a stepped substrate respectively. The Fe film on the stepped substrate has been grown at an elevated temperature. Hence, the stripes are perpendicular to the substrate induced steps. Figs. 3.42c and d display the director field of the images in Figs. 3.42a and b. The banded structure of the director field of low-temperature stripes grown on a flat substrate is also present in the high-temperature stripe phase in films grown on a flat substrate, cf. Fig. 3.42c. No such structure is, however, found in the director field of high-temperature stripes grown on a stepped substrate, cf. Fig. 3.42d. Thus, the substrate-induced anisotropy for the orientation of domain walls prevents meandering in the high-temperature stripe phase in the same way as it does in the low-temperature stripe phase.

The Fourier transforms of Figs. 3.42a and b are depicted in Figs. 3.43a and b respectively. As in the case of low-temperature stripes, the Fourier transform shows

**Figure 3.43:** Fourier transform of the high-temperature stripe phase. a, Modulus of the Fourier transform of Fig. 3.42a. b, Modulus of the Fourier transform of Fig. 3.42b. \((D_a = 1.95 \text{ ML}, D_b = 2.11 \text{ ML}, T_a = 320 \text{ K}, T_b = 293 \text{ K})\)
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Figure 3.44: Correlations in the high-temperature stripe phase. a, Positional correlation of Fig. 3.42a. b, Positional correlation of Fig. 3.42b. c, Orientational correlation of Fig. 3.42a. d, Orientational correlation of Fig. 3.42b. ($w_{a,c} = 46 \, \mu m$, $w_{b,d} = 23 \, \mu m$, $D_{a,c} = 1.95 \, \text{ML}$, $D_{b,d} = 2.11 \, \text{ML}$, $T_{a,c} = 320 \, \text{K}$, $T_{b,d} = 293 \, \text{K}$)

two well-defined peaks reflecting the two-fold order of the stripe phase.
Figs. 3.44a and b show the positional correlations calculated from the images in
Table 3.6: Fits to positional correlations in high-temperature stripes. The parameters are obtained by fitting Eqs. (3.17) and (3.18) to the experimental data shown in Figs. 3.45a and b respectively.

<table>
<thead>
<tr>
<th></th>
<th>perpendicular</th>
<th></th>
<th>parallel</th>
</tr>
</thead>
<tbody>
<tr>
<td>( a_1 )</td>
<td>0.0376</td>
<td>( A )</td>
<td>11.0 ± 0.3</td>
</tr>
<tr>
<td>( a_2 )</td>
<td>0.0790</td>
<td>( \xi_\perp [\mu m] )</td>
<td>4.00 ± 0.02</td>
</tr>
<tr>
<td>( \varphi_1 )</td>
<td>0.236</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \varphi_2 )</td>
<td>-0.145</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \lambda_1 [\mu m] )</td>
<td>1.73</td>
<td></td>
<td>( \lambda_3 [\mu m] )</td>
</tr>
<tr>
<td>( \lambda_2 [\mu m] )</td>
<td>1.55</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>( R^2 )</td>
<td>0.907</td>
<td>( R^2 )</td>
</tr>
</tbody>
</table>

Figs. 3.42a and b. The corresponding orientational correlations are presented in Figs. 3.44c and d respectively. The modulation of the positional correlation is preserved over large distances. The orientational correlation decays quickly but soon assumes a finite value. The orientational correlation is described by the order parameter \( G_2 \) directly calculated from the director field. In the case of high-temperature stripes, the order parameter \( G_2 \) amounts to approximately 0.8. This is somewhat smaller than the value of approximately 0.9 found for the low-temperature stripes. Positional and orientational correlations perpendicular and parallel to the stripe direction calculated from the image in Fig. 3.42a are shown in Figs. 3.45a and b, and 3.46a and b. As in the case of low-temperature stripes, the positional correlation perpendicular to the stripes displayed in Fig. 3.45a shows a modulation of the oscillation amplitude. A Fourier transform of the positional correlation perpendicular to the stripes reveals two closely spaced peaks responsible for the beat. The situation is thus identical to the case of low-temperature stripes in films grown on a flat substrate. A fit with Eq. (3.17) describing two-dimensional smectics and the two most prominent Fourier components yields a good agreement with the experimental data. The fitted curve is shown as a gray line in Fig. 3.45a. If the square-root dependence of the exponent in Eq. (3.17) is relaxed to the more general power law of \(-x^d\), an exponent of \( d = 0.50 ± 0.02 \) is found. As in the case of low-temperature stripes grown on a flat substrate, the exponential decay predicted for a two-dimensional
smectic supplemented with a modulation, Eq. (3.18), is capable of describing the positional correlation parallel to the stripes. The fitted curve is shown as a gray line in Fig. 3.45b. The values of the parameters in Eqs. (3.17) and (3.18) as obtained by the fits are summarized in Tab. 3.6. While the functional forms of the correlations for smectics can be used to fit the positional correlations, the parameters obtained in the fits along different directions are again not compatible with each other.

As in the case of low-temperature stripes, the positional correlations of high-temperature stripes after the initial decay can equally well be described by the power-law predicted for a smectic with anisotropy. Again, a definite statement is difficult.

The orientational correlations both perpendicular and parallel to the stripe direction can be fitted by means of Eqs. (3.19) and (3.20). The fitted curves are shown as gray lines in Figs. 3.46a and b. The values of the parameters obtained by the fits are given in Tab. 3.7. As with low-temperature stripes, the functional forms of Eqs. (3.19) and (3.20) are suitable to describe the experimentally observed orientational correlations but the values of the parameters obtained are at odds with each other. Again, the fit to the orientational correlation perpendicular to the stripe direction is more reliable, and the values of the parameters obtained in this fit are more in line with the values expected from the theory. The value of \( \lambda \) is of the expected order of magnitude (\( \lambda = L/(4\pi) \), Eq. (2.113)), and the product \( \lambda \cdot q_{\text{max}} \approx 0.38 \) is near the value of 0.5 expected according to Eq. (2.164).

Figs. 3.47 and 3.48 show the positional and orientational correlations extracted from the image in Fig. 3.42b, which shows the high-temperature stripe domains in a film grown on a slightly miscut substrate. As in the case of low-temperature stripes with a local anisotropy for the stripe orientation, meandering is hindered in high-
temperature stripes grown on a stepped substrate. The Fourier transform of the positional correlation perpendicular to the stripe direction, Fig. 3.47a, reveals the presence of a large number of peaks. The modulation of the correlation cannot be described as a simple beat.

In summary, the high-temperature stripes show the same features as the low-temperature stripes. Meandering present in films on a flat substrate is absent in films with an additional anisotropy introduced by a stepped substrate. Again, the formulae calculated for two-dimensional smectics can be fitted to the experimental data but the parameters obtained in the various fits do not match. The orientational order parameter $G_2$ in high-temperature stripes is smaller than in low-temperature stripes.
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Figure 3.45: Positional correlations in the high-temperature stripe phase on a flat substrate. a, Positional correlation of Fig. 3.42a perpendicular to the stripe direction. b, Positional correlation of Fig. 3.42a parallel to the stripe direction. The black lines represent the experimental data; the gray lines are fits to the experimental data. The dotted line represents the correlation of perfect stripes of the same width. The standard deviation $\sigma_0$ for zero correlation is given by the dashed lines. ($D_{a,b} = 1.95$ ML, $T_{a,b} = 320$ K)
Figure 3.46: Orientational correlations in the high-temperature stripe phase on a flat substrate. 

(a) Orientational correlation of Fig. 3.42 as perpendicular to the stripe direction. 
(b) Orientational correlation of Fig. 3.42 as parallel to the stripe direction. The black lines represent the experimental data; the gray lines are fits to the experimental data. The dotted lines indicate the value of \( G_2 \). (\( D_{a,b} = 1.95 \) ML, \( T_{a,b} = 320 \) K)
Figure 3.47: Positional correlations in the high-temperature stripe phase on a stepped substrate. a, Positional correlation of Fig. 3.42b perpendicular to the stripe direction. b, Positional correlation of Fig. 3.42b parallel to the stripe direction. The dotted line represents the correlation of perfect stripes of the same width. The standard deviation $\sigma_0$ for zero correlation is given by the dashed lines. ($D_{\text{a,b}} = 2.11 \text{ ML, } T_{\text{a,b}} = 293 \text{ K}$)
Figure 3.48: Orientational correlations in the high-temperature stripe phase on a stepped substrate. a, Orientational correlation of Fig. 3.42 b perpendicular to the stripe direction. b, Orientational correlation of Fig. 3.42 b parallel to the stripe direction. The dotted lines indicate the value of $G_2^2$. ($D_{a,b} = 2.11$ ML, $T_{a,b} = 293$ K)
3.11.4 Liquid Phase

The liquid phase is only found at temperatures below room temperature. It occurs between the high-temperature stripe phase and the paramagnetic phase. It is thus found in the region of the phase diagram where a tetragonal liquid has been
The distribution of the domain width in the liquid phase is wide. The intensity of the Fourier transform is smeared out around a ring and can fill the whole interior of the ring. a, Modulus of the Fourier transform of Fig. 3.32. b, Modulus of the Fourier transform of Fig. 3.32. (\(D_a = 1.56\) ML, \(D_b = 1.85\) ML, \(T_a = 154\) K, \(T_b = 163\) K)

Predicted by Abanov et al. [43]. Such a tetragonal liquid at the transition to the paramagnetic phase has also been found in computer simulations by Booth et al. [87].

Figs. 3.49 a and b show the liquid phase on a flat substrate and a stepped substrate respectively. As in the labyrinthine phase, the orientational order of the stripe phase is lost. However, the labyrinthine phase and the liquid phase differ in some important properties. Whereas the domain width in the labyrinthine phase is well-defined, it varies substantially in the liquid phase. In contrast to the labyrinthine phase, the domain pattern in the liquid phase decays into a large number of irregular, unconnected domains. The labyrinthine phase, however, consists only of one spin-up and one spin-down domain. All equally polarized regions are connected in the labyrinthine phase.

The orientational disorder is reflected in the director field, Figs. 3.49 c and d, and the Fourier transforms, Figs. 3.50 a and b. The patches in the director field are more irregular than in the labyrinthine phase. The intensity in the Fourier transform of the liquid phase is distributed over a wider ring. The Fourier transform of the image in Fig. 3.49 b shown in Fig. 3.50 b retains some degree of the two-fold symmetry of the high-temperature stripe phase. This residual two-fold symmetry is lost at higher effective temperatures.
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Figure 3.51: Correlations in the liquid phase. a, Positional correlation of Fig. 3.49a. b, Positional correlation of Fig. 3.49b. c, Orientational correlation of Fig. 3.49a. d, Orientational correlation of Fig. 3.49b. ($w_{a,c} = 23 \mu m$, $w_{b,d} = 11.5 \mu m$, $D_{a,c} = 1.56$ ML, $D_{b,d} = 1.85$ ML, $T_{a,c} = 154$ K, $T_{b,d} = 163$ K)

Positional and orientational correlations of the liquid phase are presented in Fig. 3.51. Both correlations rapidly decay to zero. In order to characterize the correlations, the
**Table 3.8:** Fits to angle-averaged positional correlations in the liquid phase. The parameters are obtained by fitting Eq. (3.29) to the experimental data shown in Figs. 3.52a and 3.53a respectively.

correlations are averaged over all the directions. The corresponding angle-averaged positional and orientational correlations for the liquid phase on a flat substrate and a stepped substrate are given in Figs. 3.52a and b and 3.53a and b respectively. While the angle-averaged orientational correlation is very similar to the angle-averaged orientational correlation of the labyrinthine phase, the angle-averaged positional correlation of the liquid phase decays even faster than in the labyrinthine phase. The correlation virtually vanishes already after the first minimum. The positional order is very short-ranged. The angle-averaged positional order has been fitted with a modulated power law:

\[
g_m(r) = \left( \frac{A}{r + r_0} \right)^\kappa \cdot \cos \left( 2\pi \frac{r}{R} \right) + M ,
\]

a modulated exponential function:

\[
g_m(r) = A \cdot \exp \left( - \frac{r}{r_0} \right) \cdot \cos \left( 2\pi \frac{r}{R} \right) + M ,
\]

and a modulated modified exponential function:

\[
g_m(r) = A \cdot \exp \left( - \left( \frac{r}{r_0} \right)^\kappa \right) \cdot \cos \left( 2\pi \frac{r}{R} \right) + M .
\]

As in the case of the labyrinthine phase, the modulated modified exponential function, Eq. (3.29), yields the best agreement with the experimentally determined
angle-averaged positional correlation. However, no phase \( \varphi_0 \) is needed to achieve a satisfactory agreement. The parameters obtained by fitting Eq. (3.29) to the experimental data in Figs. 3.52a and 3.53a are presented in Tab. 3.8. No universal value for the exponent \( \kappa \) is found but the value of the exponent \( \kappa \) in the liquid phase is substantially larger than in the labyrinthine phase.

A universal exponent \( \mu \), however, seems to govern the angle-averaged orientational correlation in both the labyrinthine phase and the liquid phase. When fits with a power law, Eq. (3.24), an exponential function, Eq. (3.25), or a modified exponential function, Eq. (3.26), are compared, the fit with the modified exponential function again yields the best results. The values of the parameters obtained by fitting Eq. (3.26) to the experimentally determined angle-averaged orientational correlations in Figs. 3.52b and 3.53b are presented in Tab. 3.9. Most striking are the values obtained for the exponent \( \mu \). They are 1.31 and 1.36 for the liquid phase on a flat substrate and a stepped substrate respectively. This is close to the value of 1.40 obtained for the labyrinthine phase.

In summary, the domains in the liquid phase are much more irregular than in the labyrinthine phase. The domain width is no longer well defined. The positional correlation decays faster than in the labyrinthine phase but a universal exponent \( \mu \) seems to describe the angle-averaged orientational correlation in both the labyrinthine phase and the liquid phase.

<table>
<thead>
<tr>
<th></th>
<th>flat substrate</th>
<th>stepped substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>( A )</td>
<td>0.998 ± 0.005</td>
<td>0.928 ± 0.006</td>
</tr>
<tr>
<td>( r_0 [\mu m] )</td>
<td>0.390 ± 0.002</td>
<td>0.442 ± 0.003</td>
</tr>
<tr>
<td>( \mu )</td>
<td>1.31 ± 0.01</td>
<td>1.36 ± 0.02</td>
</tr>
<tr>
<td>( c )</td>
<td>0.0019 ± 0.0003</td>
<td>0.0754 ± 0.0005</td>
</tr>
<tr>
<td>( R^2 )</td>
<td>0.997</td>
<td>0.994</td>
</tr>
</tbody>
</table>

**Table 3.9:** Fits to angle-averaged orientational correlations in the liquid phase. The parameters are obtained by fitting Eq. (3.26) to the experimental data shown in Figs. 3.52b and 3.53b respectively.
Figure 3.52: Angle-averaged correlations in the liquid phase on a flat substrate. a, Positional correlation of Fig. 3.49a. b, Orientational correlation of Fig. 3.49a. The black lines represent the experimental data; the gray lines are fits to the experimental data. The dotted line indicates the value of $G_2^2$. ($D_{a,b} = 1.56$ ML, $T_{a,b} = 154$ K)
3.11. ANALYSIS OF THE INDIVIDUAL PHASES

Figure 3.53: Angle-averaged correlations in the liquid phase on a stepped substrate. a, Positional correlation of Fig. 3.49b. b, Orientational correlation of Fig. 3.49b. The black lines represent the experimental data; the gray lines are fits to the experimental data. The dotted line indicates the value of $G_2^2$. ($D_{ab} = 1.85$ ML, $T_{ab} = 163$ K)
3.12 Analysis of the Mechanisms Mediating the Transitions

The mechanisms mediating the various transitions can be identified by comparing subsequent images across a transition. Unlike predicted in theoretical works [43, 79], the driving mechanisms are not the dissociations of dislocation or disclination pairs. The transition from the low-temperature stripe pattern to the labyrinthine pattern is mediated by transversal instabilities, and the ensuing transition to the high-temperature stripe pattern is characterized by knee-bend and bridge instabilities. In both processes, the variation of the average domain width as discussed in Subsec. 3.7.2 plays a vital role.

3.12.1 Mechanisms Mediating the Transition between the Low-Temperature Stripes and the Labyrinthine Phase

The transition from the low-temperature stripes to the labyrinthine pattern occurs as the effective temperature is increased. Fig. 3.54a shows the transition on a wedge. Toward smaller film thicknesses, the stripe domains emit transversal fingers which then start to bend. Two stripe segments with their fingers are highlighted. This fingering instability is made possible by the fact that the average domain width has to decrease as the effective temperature increases. When the average domain width becomes smaller, the total length of the domains within a given area has to grow. This is achieved by the emission of fingers. The increase of the total domain length could also be realized by the dissociation of dislocation pairs as discussed in Sec. 3.8. The effect would be an increase in the number of stripes but the overall orientation of the stripes would remain unaltered. This, however, is not what occurs during the transition from the low-temperature stripes to the labyrinthine pattern. The emission of transversal fingers and domain bending destroy the orientational order of the stripe domain pattern. The result is the labyrinthine pattern. The same process can, occasionally, also be observed upon variation of the actual temperature. Figs. 3.54b–d show how the domain pattern evolves as the temperature is increased. Again the stripe domains emit fingers that then bend to initiate the labyrinthine pattern. New domain segments tend to be emitted at locations where stripe domains are bent. This becomes apparent when Figs. 3.54b and c are compared. As required, the density of domains in Fig. 3.54d has grown with respect to Fig. 3.54b.

From a topological point of view, the formation of new domain segments via the transversal instability can be interpreted as the creation of a \((0,1)\) disclination together with a \((0,-1)\) disclination. A \((0,0)\) ‘defect’ decays into \((0,1)\) and \((0,-1)\)
3.12. ANALYSIS OF THE MECHANISMS

Figure 3.54: Transversal instability. The transition from the low-temperature stripes to the labyrinthine pattern is mediated by transversal instabilities. As the effective temperature is increased, domain segments emit new segments with an orientation off the original stripe direction. These new segments then bend, and the labyrinthine pattern is formed. 

a. Transition from the low-temperature stripes to the labyrinthine pattern on a wedge. The thickness of the wedge decreases from left to right. The decrease of the thickness goes along with an increase of the effective temperature. 

b–d, Transversal instabilities followed step by step. This sequence shows the same area at increasing temperatures. 

\( w_a = 155 \mu m, \quad w_{b,c,d} = 36 \mu m, \quad D_a = 2.06 \text{ ML (left)} - 1.98 \text{ ML (right)}, \quad D_{b,c,d} = 2.19 \text{ ML,} \quad T_a = 294 \text{ K,} \quad T_b = 298 \text{ K,} \quad T_c = 304 \text{ K,} \quad T_d = 305 \text{ K} \)

defects. The disclinations then unbind and destroy the orientational order of the low-temperature stripes.

Seul and Wolfe have observed the appearance of a labyrinthine pattern in ferromagnetic garnets [21, 22]. It is formed when a stripe domain pattern obtained by cooling down from a temperature above the Curie temperature is heated. In order to decrease the average domain width, the domains first assume a regular chevron pattern. At even higher temperatures, disclination dipoles similar to the transversal instability occur at the tips of the chevron structure. Once the disclination dipoles have been formed, the domain length grows as the disclination dipoles unbind. Seul
and Wolfe argue that the labyrinthine pattern formed by means of this procedure represents a metastable state obtained by minimizing the free energy under the constraint of a fixed number of domains. This is indeed substantiated by the fact that no labyrinthine pattern is formed when the garnet film is cooled down from a temperature above the Curie temperature. In the case of ultrathin Fe films on Cu(001), however, the labyrinthine pattern also occurs when the sample is cooled down from the paramagnetic state, cf. Subsec. 3.12.2. There are thus a number of similarities between the magnetic domains in ferrimagnetic garnets and ultrathin Fe films on Cu(001) but some differences remain.

The formation of labyrinthine patterns has not only been observed upon varying the temperature but also as a result of demagnetizing. The equilibrium width of perpendicularly magnetized domains is reduced when a perpendicular field is applied to the sample [20]. When the magnetic field is switched off, the average domain width has to decrease. In cases where domain nucleation is inhibited, this can be achieved by the growth of existing domains. In a study on ferrimagnetic garnet films, Molho et al. discovered that the overall domain length per unit area was increased by bending of stripe domains and the creation of disclination dipoles at bent domain sections [168]. For sufficiently high initial fields, the final domain pattern resembled the labyrinthine pattern found here. Seul and Wolfe found that the type of disordered pattern created by demagnetizing depended on the rate by which the magnetic field was reduced [22]. When the field was lowered slowly enough, the increase of the domain length per unit area was achieved by the bending of the few initial minority domains. As no branching occurred in the minority domains, all the disclinations present in the final labyrinthine phase were restricted to the original majority domains. Domain bending seemed to be hampered when the magnetic field was reduced within a short time. Then, a kind of transversal instability occurred and the final pattern consisted of stripe domains with chains of closely bound disclination pairs. The disclinations were now equally distributed among the original minority and majority domains. Due to the limited time resolution of the experiment, the way by which this so-called ‘comb’ pattern was formed remained a puzzle.

It is interesting to note that earlier experiments by Kooy and Enz had led to results that conflict with the observations by Seul and Wolfe, and Molho et al. presented here [20]. As in ultrathin Fe films on Cu(001), a labyrinthine pattern is formed in BaFe\(_{12}\)O\(_{19}\) films cooled down from the paramagnetic state. Contrary to the findings of Seul and Wolfe, and Molho et al. presented above, Kooy and Enz describe the domain pattern that evolves after demagnetizing as that of stripe domains.
3.12.2 Mechanisms Mediating the Transition between the Labyrinthine Phase and the High-Temperature Stripes

Like the transition from the low-temperature stripes to the labyrinthine pattern, the ensuing transition to the high-temperature stripes is accompanied by a decrease of the average domain width. In the former transition, the newly created domain segments are oriented off the original stripe-domain direction and eventually destroy orientational order. In the latter transition considered in more detail here, however, the newly created domain segments preferentially align along the original stripe-domain direction, which results in a nicely ordered high-temperature stripe pattern. Fig. 3.55 shows three stages on the way from the labyrinthine pattern to the high-temperature stripe domains. The labyrinthine pattern is characterized by a large density of disclinations, but also a lot of bent domain sections. It is exactly from these bent sections that new domain segments are created during the transition. Such new segments emanating from bent sections are highlighted in Fig. 3.55b. They are all aligned along a single direction that coincides with the orientation of the low-temperature stripe domains. Such a uniform alignment is necessary to establish a stripe-domain pattern.

The process leading to the formation of new domains is exemplified by two sequences of SEMP A images in Fig. 3.56. The bell-shaped domain section highlighted in Fig. 3.56a emits a new segment that cuts into the surrounding domain of opposite magnetization, Fig. 3.56b. As a consequence, domain sections adjacent to the new domain segment are aligned along the direction of the new segment. In a further step, the white domain enclosed by the bell-shaped domain is cut by the moving tip of a black domain, Fig. 3.56c, and another bell-shaped domain configuration inside the original bell is formed. This new bell then as well emits a new segment which cuts into the segment created by the original bell Fig. 3.56d. Apart from reducing the average domain width, these processes align more and more domain sections along a single direction. The basic process in which a new segment is emitted from a bent domain section has also been called a knee-bend instability [169]. Such knee-bend instabilities are abundant during the transition to the high-temperature stripes. Another example of a knee-bend instability is shown in the sequence of Figs. 3.56e–h.

Knee-bend instabilities have been found as solutions to the Cross-Newell pattern-formation equation [169]. As soon as the angle between the two sides of a knee-bend falls short of a certain value, the knee-bend becomes unstable with respect to the emission of a new segment at the knee-bend. The formation of the new segment has been analyzed as the creation of a pair of (0,1) and (0,−1) disclinations. From a topological point of view, we thus have a decay of a (0,0) ‘defect’ into (0,1) and
Figure 3.55: From the labyrinthine pattern to high-temperature stripes. A characteristic feature of the transition to the high-temperature stripes are new domain segments that are emitted from bent domain sections. a, Labyrinthine pattern. b, New domain segments emanating from bent domains. The new domains are all aligned along a single direction and grow continuously. c, High-temperature stripe domains. All the images were taken at the same location of the same film. ($w_a = 260 \mu m$, $w_b = 130 \mu m$, $w_c = 65 \mu m$, $D_{a,b,c} = 1.95$ ML, $T_a \approx 310$ K, $T_b \approx 316$ K, $T_c \approx 320$ K)
3.12. ANALYSIS OF THE MECHANISMS

Figure 3.56: Knee-bend instability. New domain segments are created at bent domain sections as the temperature is increased during the transition to the high-temperature stripe domains. a–d and e–h, Sequences showing the evolution of knee-bend instabilities. The temperature rises from left to right. A bridge instability is highlighted in blue in g and h. i–j The knee-bend instability from a topological point of view. The knee-bend instability can be considered as the decay of a (2,0) dislocation into two (1,0) dislocations. (w_{a–d} = 23 \, \mu m, w_{e–h} = 33 \, \mu m, D_{a–h} = 1.95 \, ML, T \text{ increases from } T_a \approx 311 \, K \text{ to } T_d \approx 315 \, K \text{ and from } T_e \approx 311 \, K \text{ to } T_h \approx 316 \, K)

(0, -1) disclinations. On a truly local level, such an analysis is also applicable here, but with regard to the stripe-domain pattern forming in the surrounding area, it is more appropriate to identify the knee-bend instability as the decay of a (2,0) dislocation into two (1,0) dislocations. This is illustrated with the sketches in Figs. 3.56i and j. According to Eq. (2.116), the energy of a dislocation is proportional to the square of its strength given by its Burgers vector. A decay of a (2,0) dislocation into two (1,0) dislocations is thus energetically favorable since the total energy is
Figure 3.57: Orientation of newly created domain areas. As the temperature is increased, new domain segments have to be created for the average domain width to be reduced. New domain segments grow at the tips of domains that already exist and via knee-bend instabilities. a, Labyrinthine pattern at a lower temperature. b, Labyrinthine pattern at a higher temperature. The domains highlighted in a have been transformed into the domains still highlighted in yellow in b. The domains highlighted in green in b are new domain segments created by domains in the area highlighted in yellow. (\(w_{a,b} = 92 \mu m, D_{a,b} = 1.95 \text{ ML}, T_a \approx 311 \text{ K}, T_b \approx 316 \text{ K}\))
3.12. ANALYSIS OF THE MECHANISMS

Figure 3.58: Return to the labyrinthine phase. Before the labyrinthine phase is established, the high-temperature stripe phase is characterized by bubble-like domains. 

a, High-temperature stripe phase with bubble-like domains. b, Labyrinthine phase created from the high-temperature stripe phase. The two images were taken at the same location of the same film. (w = 92 µm, \(D_{a,b} = 1.97\) ML, \(T_a = 321\) K, \(T_b = 296\) K)

temperature stripe patterns prior to the transformation to labyrinthine pattern is the formation of small bubble-like domains. Such bubble-like domains are highlighted in Fig. 3.58 a. The final labyrinthine pattern is only established after several hours. It is depicted in Fig. 3.58 b. While the continuous decrease of the average domain width leaves enough space for bending and new domains to be created by transversal instabilities during the transition from the low-temperature stripe phase to the labyrinthine phase, more complicated rearrangements are needed when the labyrinthine pattern is to be formed out of the high-temperature stripe phase. The reordering process is thus hampered and requires a lot of time.

3.12.3 Mechanisms Mediating the Transition between the High-Temperature Stripes and the Paramagnetic Phase

In sufficiently thick ultrathin Fe films on Cu(001), the Curie temperature is on the order of room temperature or higher. In this case, the ferromagnetic-paramagnetic phase transition is preempted by a dynamic transition. When the temperature approaches the Curie temperature, the stationary stripe domains of the high-temperature stripe phase start to become mobile. The same mobile phase is seen when the temperature is lowered from a temperature above the Curie temperature. Figs. 3.59 a and b show the domain pattern as it evolves as the temperature falls below the Curie temperature. Immediately below the Curie temperature, the domains are highly mobile. This is seen by the abrupt displacements of the domains between successive scan lines. However, there are isolated patches of stationary domains within which the domain pattern consists of the same stripes as in the high-temperature stripe phase. The stripes have the same width and the same orientation. This is an indi-
Figure 3.59: Transition to the paramagnetic phase. In sufficiently thick films, a mobile stripe phase occurs between the high-temperature stripe phase and the paramagnetic phase. The images show the intervening mobile phase both as a function of temperature and as a function of film thickness, and hence effective temperature. The images are scanned from left to right, with the first scan line taken at the top of the images. 

a, Transition from the paramagnetic phase to high-temperature stripes upon cooling down. 
b, Transition from the paramagnetic phase to high-temperature stripes upon cooling down. 
c, Transition on a wedge grown onto a flat substrate. 
d, Transition on a wedge grown onto a stepped substrate.

\( w_{a,b} = 18.4 \, \mu m, \, w_{c,d} = 23 \, \mu m, \, D_a = 1.94 \, ML, \, D_b = 1.96 \, ML, \, D_c = 2.06 \, ML \) (left) \(- 2.13 \, ML \) (right), 
\( D_d = 2.17 \, ML \) (left) \(- 2.24 \, ML \) (right), 
\( \Delta T_a \approx 3 \, K, \, \Delta T_b \approx 3 \, K, \, T_{c,d} = 293 \, K \)
cation that the domain pattern in the mobile phase also consists of stripe domains. Faster imaging techniques are required to obtain more information on the domain pattern in the mobile phase and its dynamics. The mobile phase is restricted to a temperature range on the order of 1–2 K below the Curie temperature. Unlike the domain displacements due to moving dislocations discussed in Sec. 3.8, the domain displacements observed here in a narrow temperature range just below the Curie temperature are not needed to adjust the mean domain width. The experimentally determined temperature dependence of the mean domain width near the Curie temperature is well described by the calculations in the framework of mean-field theory, cf. Subsec. 2.3.3. The variation of the mean domain width is proportional to \((T_c - T)^2\), cf. Subsec. 3.7.2, and thus vanishingly small just below the Curie temperature. That the displacements are not induced by the need to establish a different equilibrium width is substantiated by images taken on wedges kept at a constant temperature, cf. Figs. 3.59c and d. There, the transition line between the stationary high-temperature stripes and the mobile phase is blurred. Whereas abrupt displacements are seen in some areas, the domain pattern remains stationary in other areas of the same film thickness. As in Figs. 3.59a and b, isolated patches with a stationary domain pattern are observed inside the mobile phase.

In [14], the stationary domain pattern in ultrathin Fe films on Cu(001) was interpreted as a ‘floating solid’ pinned by defects of the substrate. But the systematic occurrence of a mobile phase just 1–2 K below the Curie temperature irrespective of the exact value of the Curie temperature (on the order of room temperature or higher) suggests that the mobile phase is an intrinsic property of the magnetic domain pattern in ultrathin Fe films on Cu(001).

In their analysis of supercooled liquids, Kivelson et al. find that a glass transition occurs at a temperature slightly smaller than a narrowly avoided thermodynamic phase transition [16]. Such a phase transition can be avoided due to an additional frustrating interaction. In the case of thin magnetic films with an easy axis perpendicular to the film plane, the Ising transition is avoided due to the long-ranged dipolar interaction, cf. Subsec. 2.3.6. In such a situation, Kivelson et al. expect exponentially long relaxation times in a temperature range where the correlation length associated with the avoided transition is still felt but smaller than the domain width. From this point of view, the stationary domain pattern can be interpreted as the result of a self-generated glass transition. No pinning centers are needed to produce a stationary domain pattern. Given the fact that the temperature range of the mobile phase scales with the Curie temperature, such an interpretation seems reasonable. A self-generated stripe glass for frustrated systems has also been predicted by Schmalian and Wolynes using the mean-field replica formalism with the self-consistent screening approximation [17]. They find that in a system with competing interactions on different length scales an exponentially large number of metastable states will emerge and lead to long, landscape-dominated relaxation times. The Langevin
dynamics of a continuum version of the Coulomb-frustrated Ising ferromagnet has been analyzed in simulations by Grousson et al. [18]. Using either the mode-coupling approximation or a dynamical self-consistent screening approximation, they find a transition from an ergodic to a nonergodic behavior. The transition temperature is found to coincide with the temperature at which Schmalian and Wolynes predict the occurrence of an exponentially large number of metastable states. The ergodicity-breaking transition is observed close to the fluctuation-induced first-order transition from the paramagnetic to the modulated phase predicted by Brazovskii [80]. For most values of the frustration parameter $Q$, which describes the relative strength of the frustrating interaction, Grousson et al. find the ergodicity-breaking transition to occur at slightly higher temperatures than the fluctuation-induced first-order transition from the paramagnetic to the modulated phase. If the transition from mobile stripes to stationary stripes observed here is to be interpreted as an ergodicity-breaking transition of the type investigated by Grousson et al., the ergodicity-breaking transition has to occur below the temperature of the fluctuation-induced first-order transition from the paramagnetic to the modulated phase. Such an inversion is indeed observed by Grousson et al. for small values of the frustration parameter $Q$. Thus, it seems that the experimentally found dynamic transition can be associated with the ergodicity-breaking transition of Grousson et al. [18]. However, the results obtained by Schmalian and Wolynes [17] and Grousson et al. [18] have been challenged by Geissler and Reichman [170]. In their dynamical simulations of a minimal model of frustration-limited domains, they observe that the relaxation times increase in a non-Arrhenius manner when the microphase separation transition is approached, but no transition to a nonergodic regime.

3.12.4 Mechanisms Mediating the Transition between the High-Temperature Stripes and the Liquid Phase

The transition from the high-temperature stripes to the liquid phase is depicted in Fig. 3.60. It is characterized by a gradual loss of both orientational and translational order. The domains that form in the liquid phase are highly irregular. The transition thus cannot be described in terms of well-defined topological defects as it was the case with the other transitions considered in Sec. 3.12. While the domain boundaries remain sharp, the equally spaced domains of the high-temperature stripe pattern seem to split and merge as the liquid phase is approached. Such processes where domains are split into unconnected parts or unconnected domains merge occur rarely in the other transitions. The bridge instability which is found during the transition from the labyrinthine phase to the high-temperature phase involves a similar rearrangement. Note, however, that in that transition the domains are still

\[27\] The corresponding transition on a stepped substrate is displayed in Fig. 3.23.
Figure 3.60: Transition to the liquid phase. \(a\)–\(c\), Transition from the high-temperature stripes to the liquid phase as observed on a wedge. The three images were recorded on the same wedge and can be matched to produce a single continuous image showing the gradual decay of order on approaching the liquid phase.

\(w_{a-c} = 75 \, \mu m, \; D_a = 1.79 \, ML \) (left) – 1.71 ML (right), \(D_b = 1.71 \, ML \) (left) – 1.64 ML (right), \(D_c = 1.64 \, ML \) (left) – 1.56 ML (right), \(T_{a-c} = 154 \, K\)

well defined and the bridge instability can be understood in terms of topological defects.
The relative ease with which merging and splitting processes occur as the liquid phase is approached may be due to the high effective temperature.
Chapter 4

Discussion

The system of ultrathin Fe films on Cu(001) displays a number of different domain patterns. These patterns have been imaged on a microscopic level using Scanning Electron Microscopy with Polarization Analysis (SEMPA). In the thickness range investigated in this study, the sequence of phases can be interpreted as a function of an effective temperature. At low effective temperatures, the domain pattern is characterized by stripe domains. At higher effective temperatures, this stripe-domain pattern transforms into a labyrinthine pattern. The orientational order of the low-temperature stripe phase is lost. At even higher effective temperatures, however, the labyrinthine pattern is replaced by a high-temperature stripe phase. While the sequence of these phases can be described as a function of the effective temperature, the route taken by the system on its way to the paramagnetic phase depends on the absolute value of the temperature. At low absolute temperatures, the well-ordered high-temperature stripe phase is separated from the paramagnetic phase by a phase with highly irregular domains, the liquid phase. When the transition to the paramagnetic phase is observed at sufficiently high absolute temperatures, however, no such intervening liquid state is found. Instead, the high-temperature stripe phase undergoes a dynamic transition. At temperatures below the temperature of the dynamic transition, the stripe domains form a stationary pattern. At higher temperatures, the stripe domains become mobile. The dynamic transition occurs only slightly below the transition to the paramagnetic phase.

An effort has been made to understand the various phases from a theoretical point of view. An important property of domain patterns is the mean domain width. Its dependence on temperature near the Curie temperature is obtained by minimizing the Fourier transform of a mean-field free energy containing both exchange and dipolar interactions with respect to the wave vector. It is found that the domain width $L$ near the Curie temperature $T_c$ varies as $(T_c - T)^2$, a temperature dependence that is confirmed experimentally. In order to compare the free energies of domain patterns with different symmetries, selection rules for Fourier components compatible with a
given symmetry are elaborated. The free energy of domain patterns with an overall \( C_2 \), \( C_4 \), or \( C_6 \) rotational symmetry is calculated. With the approximations made, it is found that higher-symmetry phases have a higher free energy. A transition between phases with different symmetries is thus not expected from the mean-field free energy.

The mean-field free energy, however, forms the basis for a continuum model that describes the free-energy in terms of elastic constants and deviations from a perfect stripe structure. The model obtained is the model used for dealing with two-dimensional smectics. In this model, the energy of a single dislocation is finite. Hence Toner and Nelson predict that dislocation pairs would dissociate at any finite temperature \[79\]. At sufficiently large distances, the order would be that of a nematic. The continuum model of a smectic has also been analyzed by Abanov et al. \[43\]. In addition to the terms already considered by Toner and Nelson, they include an anisotropy for the stripe direction due to higher-order gradient terms of the exchange interaction. The type of order is then sensitive to the strength of this anisotropy as it decides on whether the energy of a single dislocation is finite or infinite. For sufficiently small values of the anisotropy, a single dislocation has a finite energy and the scenario already discussed by Toner and Nelson is expected. For higher values of the anisotropy energy, however, the energy of a single dislocation becomes infinite so that dislocations remain paired at low temperatures. The disordering of the stripe pattern is then found to proceed in two-steps. In the first step, the positional order of the low-temperature phase with the characteristics of a smectic is predicted to be lost as dislocation pairs unbind. Above this transition the system is in a nematic-like state. The orientational order of the nematic state is then predicted to give way to an isotropic phase via the unbinding of disclination pairs. Experimentally, we find that the orientational correlations of both low-temperature and high-temperature stripes are long-ranged. This corresponds to the latter case where the anisotropy due to the higher-order gradient terms is sufficiently large to prevent a dissociation of dislocation pairs at low temperatures. Indeed, the stripes in both low-temperature and high-temperature stripes are found to be aligned along the \( \langle 100 \rangle \) direction throughout films extending over several square millimeters. This is the direction expected by Abanov et al. after analyzing the higher-order gradient terms. Nonetheless, both positional and orientational correlations of the stripe phases can be described by the functional forms of the correlation functions calculated for two-dimensional smectics without anisotropy. Note, however, that the parameters obtained in the fits along different directions are not compatible with each other. While the positional correlation functions of two-dimensional smectics have been calculated before, approximations for the orientational correlation are presented in this study. The correlation functions including the anisotropy energy can be expected to describe the behavior at larger distances. On the range accessible by analyzing a single image, the correlation functions calculated for smectics
without anisotropy are sufficient. But beyond the initial decay, the correlations can equally well be fitted with the correlation functions including the anisotropy energy. When the stripe phases are considered as the smectic state predicted by Abanov et al. for a sufficiently large anisotropy, the liquid phase found to separate the high-temperature stripes and the paramagnetic state at low temperatures could be interpreted as the tetragonal liquid expected by Abanov et al. after the unbinding of disclination pairs. However, the experimentally observed transition from the high-temperature stripe phase to the liquid phase cannot be described in terms of topological defects. The liquid phase is rather the result of a general instability of the domain pattern close to the paramagnetic phase. In the liquid phase, both positional and orientational order are short-ranged. The orientational correlation is thereby found to be well described by a function exp[−(r/R)μ] where μ is on the order of 1.3–1.4. The same dependence on the distance r is observed for the orientational correlation in the labyrinthine phase. The transition from the liquid phase to the paramagnetic phase remains to be explored in more details in future experiments.

At lower effective temperatures, however, the domains are robust. The transitions between the low-temperature stripe phase and the labyrinthine phase as well as the ensuing transition to the high-temperature stripe phase can thus be analyzed in topological terms. The transition from the low-temperature stripes to the labyrinthine pattern proceeds via transversal instabilities which lead to new domain segments perpendicular to the stripe direction. The ensuing growth of the new segments can be interpreted as the unbinding of disclination pairs. The labyrinthine phase is thus not the result of unbinding dislocation pairs as predicted for the disordering of the stripe phase by Toner and Nelson or Abanov et al. [43, 79]. An analysis of the director field shows that the labyrinthine pattern decays into patches with a uniform director field inside. The patches are separated by phase and amplitude grain boundaries. A role for grain boundaries in the disordering of striped systems is predicted by Chui [83, 84]. The corresponding phase transition is expected to be first order. This may explain the long time required for the low-temperature stripe phase to evolve from the labyrinthine phase. However, the exact way by which the grain boundaries are expected to form is not clear from [83, 84]. The knee-bend instability leading from the labyrinthine phase to the high-temperature stripe phase can be interpreted as the decay of higher-order dislocations with a high energy into simple dislocations with a lower total energy. If the focus is on the evolution of the newly created domain segments at the knee-bend, the mechanism mediating the transition can also be identified as an unbinding of disclination pairs.

In both the transition from the low-temperature stripe phase to the labyrinthine pattern and the transition from the labyrinthine pattern to the high-temperature stripe phase, new domain segments are created, in a direction perpendicular to the low-temperature stripes and at knee-bends respectively. The creation of new domain
segments allows adjusting the mean domain width to the value required by the free energy. Within the stripe phases, the required mean domain width is obtained by gliding dislocations.

Both positional and orientational correlations in the labyrinthine phase are short-ranged. The orientational correlation is very similar to the orientational correlation in the liquid phase and again described by a function \( \exp \left( -r/R \mu \right) \) where \( \mu \) is on the order of 1.3–1.4. The applicability of this function to the orientational correlation of both the labyrinthine and the liquid phase calls for a common theoretical model. The positional correlation in the labyrinthine phase is not as short-ranged as in the liquid phase. It only vanishes after a small number of oscillations.

The experimentally established phases and the mechanisms found to mediate the transitions between them can serve as a testing ground for future theoretical models. At least as far as the mechanisms are concerned, the temperature dependence of the domain width has to be taken into account. Its knowledge is pivotal to understanding the mechanisms involved in the transitions. None of the models so far has explicitly included this variation with temperature. A particularly striking feature of the sequence of phases is the reentrance of the lower-symmetry high-temperature stripe phase. No explanation for this behavior has been found so far and the reentrance remains an intriguing puzzle. Due to the ubiquity of modulated phases, insight gained in this system of ultrathin Fe films on Cu(001) magnetized perpendicular to the film plane can be relevant for a variety of physically diverse systems. Interestingly, at sufficiently high temperatures, the high-temperature stripe domains become mobile shortly before the paramagnetic state is reached. The transition to this mobile phase is announced by a series of abrupt displacements of the stripe domains. In the mobile phase, some areas remain stationary and exhibit the same stripe domain pattern as in the high-temperature stripe phase. Hence, it can be assumed that the domain pattern in the mobile phase remains characterized by stripe domains. However, faster imaging techniques are required to obtain a more comprehensive and more definite idea of the mobile phase. Especially desirable are sequences of images taken at a well-defined fixed temperature and at regular intervals. Such sequences will allow calculating the time correlation and hence shed more light on the temperature dependence of the relaxation time. This will be useful to compare the properties of the system of ultrathin Fe films on Cu(001) to results obtained in simulations and theoretical models. Insight gained in this system may be relevant for understanding the dynamics of stripes in similar systems such as high-temperature superconductors \([171–173]\). An interesting question is how the assumed patches of mobile stripe domains are connected to the stationary patches, and whether the stripe domain structure is preserved until the paramagnetic phase is reached. A comparison with the model of a Coulomb-frustrated ferromagnet suggests that the dynamic transition observed here might be a glass transition. In this case, the ultrathin magnetic films investigated here would be a system where such
a glass transition can be followed in the real space and in great detail. Another field for future work is the interplay of film structure and magnetic properties on the level of magnetic domains. The labyrinthine phase only occurs in films grown at elevated temperatures. In films grown at lower temperatures, the stripe phase covers the whole temperature range from low temperatures to close to the Curie temperature. In order to find out whether the labyrinthine phase is coupled to a specific structure of the Fe film, LEED $I(E)$ curves have been measured for films of different thicknesses and grown under different conditions. While different growth conditions do result in slightly different film structures, no structures peculiar to the labyrinthine phase can be identified. The structure at the thickness where the labyrinthine pattern is observed does not differ from the structure in films slightly thinner or thicker. Film growth at elevated temperatures is found to lead to an increased interlayer distance perpendicular to the film plane. While the LEED $I(E)$ curves do deliver some information on the structure, more information on this complex epitaxial system can be expected from scanning tunneling microscopy (STM). Such an investigation would also be desirable from another point of view. On a stepped substrate, high-temperature stripes are very sensitive to the growth conditions. They are either perpendicular or parallel to the steps of the substrate. It would be highly interesting to identify the features of the film structure or morphology that have such a profound impact on the domain pattern.
Part II

Ultrathin Iron Dots on Cu(100)
Chapter 1

Introduction

As has been shown in Part I, ultrathin Fe films on Cu(001) are magnetized along a direction perpendicular to the film plane and show a variety of magnetic domain patterns. The order of the domain pattern is thereby a function of the effective temperature. Except for the contrastless paramagnetic state, both spin-up and spin-down domains are present in each of these phases. The domain width is largest at low temperatures and decreases as the effective temperature is increased.

In this Part, the influence of a lateral confinement is investigated. The question of whether the multi-domain state found in large samples will eventually transform into a single-domain state for sufficiently small samples has long been the subject of both theoretical and experimental work. Calculating the energy of different plausible magnetic-domain configurations for simple geometries such as extended films, needles, or cubes, Kittel has shown that the flux-closure multi-domain state of large samples should give way to a uniform magnetization distribution as the dimensions of a sample are reduced [33]. Kittel thereby considered the cases of increasingly thin films, increasingly thin needles, and cubes shrinking in all three dimensions. The transition to the single-domain state is predicted to occur as the shrinking dimensions become of the order of the domain width in the case of films\(^1\) or an order of magnitude smaller in the other cases.

The case relevant for this study is that where the lateral dimensions of an ultrathin dot are varied. This case has been investigated theoretically by Kashuba and Pokrovsky [3]. They showed that the difference in magnetostatic energy between a two-domain and a uniform state scales as \(-L \ln(L/L_0)\), where \(L\) is the lateral dimension of the dot and \(L_0\) is the width of the domain wall. The competing domain wall energy scales as \(L\), hence a multi-domain state can be expected in large samples but a single-domain state should emerge as the lateral dimensions are reduced below

\(^1\) The case of alternating spin-up and spin-down stripe domains without flux-closure domains near the film surfaces is predicted to occur only for sufficiently strong uniaxial anisotropies with the easy axis perpendicular to the film plane.
a critical value. Not neglecting the finite width of domain walls, Skomski et al. find that the critical value is an order of magnitude larger than the width of the stripe domains [174].

Experimental findings, however, suggest that the transition to the single-domain state occurs when the lateral dimensions are of the order of the stripe-domain width [175, 176]. These experiments were carried out with ultrathin dots of Fe on Cu(001). Here, the tunability of the stripe width by adjusting the temperature or the film thickness is used to investigate the transition somewhat more extensively. Part of the results obtained here has been published [177]. Note that the case of perpendicularly magnetized dots differs from the case of in-plane magnetized dots. There, it has been shown that the various energy contributions have the same scaling behavior so that no transition to a multi-domain state should occur [176]. Such a behavior has been confirmed experimentally in ultrathin Co dots on Cu(001) [175]. However, Gu et al. do find a transition from a single-domain to a multi-domain state in 150 Å thick Fe(100) elements as the lateral dimensions are reduced [178]. This inverse transition is explained as due to the increasing importance of the demagnetizing fields originating at the boundaries as the elements become smaller and smaller.
Chapter 2

Experimental Procedures

The laterally confined dots are produced using the procedures described in Sec. 2.1 of Part I. As in the case of wedges, a mask is placed between the evaporation source and the substrate. However, in this case, the mask is not moved during the evaporation process. The substrate is placed right behind the mask. Its orientation is carefully controlled to ensure a parallel alignment of the mask and the substrate. This is necessary to minimize the distance between the mask and the substrate so that shadow effects can be reduced. The mask consists of a 1 \( \mu \text{m} \) thick foil with holes created by focused-ion-beam irradiation. These holes define the two-dimensional Fe structures to be investigated. SEM topography images suggest that the sharpness of the structures created by this procedure is on the order of 0.3 \( \mu \text{m} \).

The control of the film thickness is crucial when the domain width at room temperature is to be adjusted by the thickness of the film. Unlike in extended films, Auger electron spectroscopy cannot be used to determine the thickness of isolated dots with dimensions on the order of microns or fractions of microns. Instead, the film thickness has to be calculated as the product of evaporation time and rate. The evaporation rate is determined in extended films both before and after the experiments with the laterally confined dots.

The magnetic domain structure of the dots is imaged using SEMPA, cf. Subsec. 3.4.1 of Part I. In order not to influence the domain pattern with the impinging electrons of the scanning electron microscope, the acceleration voltage of the primary electron beam is kept at 2 kV. Care has to be taken to minimize the exposure time of the dots. This is especially true when small dots are to be imaged at a high lateral resolution.
Chapter 3

Variation of Thickness

It has been shown in Part I that the domain width depends on the effective temperature. The effective temperature of an area is defined as the ratio between the actual temperature and the Curie temperature corresponding to films of this thickness. If the domain structure of the dots is to be monitored at room temperature, the domain width can be adjusted by the film thickness. The Curie temperature as a function of film thickness is discussed in Sec. 3.6 of Part I. After a steep initial increase, the Curie temperature decreases again as the film thickness is further increased. Hence the domain width in films with a thickness up to about 2 ML is highly sensitive to the film thickness. As the film thickness of dots can only be determined in an indirect way, it is more suitable to use thicker films where the Curie temperature varies more slowly with the film thickness. Thus, the experiments reported in Part II have a thickness in the range beyond the maximum of the Curie temperature.

Fig. 3.1 shows the magnetic domain pattern of a group of square-shaped dots at various film thicknesses measured at room temperature together with the corresponding topography images. In extended films in this thickness range, the domain width decreases as the thickness is increased. The same behavior is observed in these laterally confined structures. The thickness of the dots increases from Fig. 3.1a to i, and so does the domain density. The relatively thin dots of Fig. 3.1a are in a single-domain state. The lateral dimensions of the dots are smaller than the domain width of extended films with the same thickness. It is found that the single-domain state of a dot gives way to a multi-domain state as soon as the domain width is on the order of the lateral dimension of the dot. The domain width is uniform across dots with the same thickness. An exception is the 2 μm dot in Fig. 3.1g, which consists of two domains substantially larger than expected from the domain width in the largest dot of the same series. This may be due to a reduced film thickness in the smaller dots. The topography images reveal that the mask may have moved vertically during the evaporation process.

Thus, in general, the domain pattern of ultrathin films remains unaltered as the
Figure 3.1: Number of domains in ultrathin Fe dots on Cu(001) as a function of thickness. The left-hand column shows the magnetic domain patterns as obtained from SEMPA. The corresponding topography images are displayed in the right-hand column. The thickness of the dots increases from top to bottom, and so does the effective temperature. Consequently, the density of domains increases from top to bottom. The approximate side lengths of the squares are 0.2 μm, 0.5 μm, 1 μm, 2 μm, and 5 μm respectively.

\( w_{a-j} = 23 \mu m, \ D_{a,b} \approx 3.02 \ ML, \ D_{c,d} \approx 3.05 \ ML, \ D_{e,f} \approx 3.09 \ ML, \ D_{g,h} \approx 3.17 \ ML, \ D_{i,j} \approx 3.23 \ ML, \ T_{a-j} = 293 \ K \)

lateral dimensions are reduced. There seems, however, to be a small anisotropy for the stripe orientation due to the orientation of the dots. While the domain walls on a flat substrate tend to be aligned along a \( \langle 100 \rangle \) direction, the domain walls in the dots run mostly parallel to the sides of the dots.
Chapter 4

Variation of Temperature

The variation of the domain width with temperature in extended films has been investigated in Subsec. 3.7.2 of Part I. The domain density increases with temperature. Fig. 4.1 shows how the domain pattern in square-shaped dots of various sizes evolves as the temperature increases. The side lengths of the squares are 1 µm, 2 µm, and 5 µm respectively. As soon as the domain width is reduced to the dimension of a given dot, the single-domain state of that dot will be transformed into a multi-domain state. Typically, the newly formed areas with a reversed magnetization are created at the edges of the dots, notably at the edges perpendicular to the stripe direction. The newly created domains then proceed into the interior of the dots and eventually reach the opposite edge. Typically, the first reversed areas follow the edges parallel to the stripe direction, cf. Fig. 4.1 b and d. New domains are thus not created by a sudden reversal of the magnetization in large areas, and the initial multi-domain state is not the symmetric two-domain state considered in model calculations [3, 174].

Note that at the location of the dots the substrate is characterized by a high density of steps running along the vertical of the images. The orientation of the stripes along the vertical line can thus be understood as due to the anisotropy introduced by the steps, in addition to a possible anisotropy induced by the edges of the dots.
Figure 4.1: Number of domains in ultrathin Fe dots on Cu(001) as a function of temperature. The images a to j show the domain structure of a group of square-shaped dots as the temperature is increased. The domain density increases with temperature. A multidomain structure enters the dots when the dimensions of the dots become of the order of the domain width. The approximate side lengths of the squares are 1 µm, 2 µm, and 5 µm respectively. ($w_{a-j} = 18.4 \mu m$, $D_{a-j} = 2.86$ ML, $T_a \approx 296$ K, $T_b \approx 300$ K, $T_c \approx 303$ K, $T_d \approx 305$ K, $T_e \approx 308$ K, $T_f \approx 310$ K, $T_g \approx 312$ K, $T_h \approx 314$ K, $T_i \approx 317$ K, $T_j \approx 318$ K)
Part III

Domain Walls in Ultrathin Iron Films on $W(110)$
Chapter 1

Introduction

The experiment reported in Part III has been inspired by a recent research paper by Baek et al. [179]. The paper focuses on an in-plane–to–in-plane reorientation transition in thin Fe films on W(110). In contrast to ultrathin Fe films on Cu(001) considered in Part I and Part II, thin Fe films grown on W(110) grow as bcc Fe(110) and are characterized by a strong in-plane anisotropy. At a critical thickness of 85 Å, the magnetization direction switches from the [1 10] in-plane direction to the perpendicular in-plane direction of [001].

While many aspects investigated in the paper by Baek et al. have already been the subject of previous publications (e.g. [180–182]), the spin-resolved photoemission spectra presented in the paper also provide some data on the spin polarization as a function of film thickness. They show that the sum of the spin polarizations along the [1 10] and [001] directions remains constant across the narrow transition region of only about one monolayer. This suggests that both domains oriented along the [1 10] direction and domains oriented along the [001] direction should be present in the transition region. However, the magnetic configuration at this in-plane–to–in-plane spin reorientation transition has so far neither been calculated nor imaged.

The spatial resolution of SEMP A makes it an ideal tool to tackle this still unresolved issue. In order to image the transition region in detail, Fe films on W(110) are grown as extended wedges. The results of this study are presented in Chapter 3 and reveal that the transition region is indeed marked by the simultaneous presence of domains oriented along the [1 10] direction and domains oriented along the [001] direction. The results obtained by Baek et al. using spin-polarized photoemission can thus be confirmed. It is found that the domain wall separating the domains oriented along the two mutually perpendicular in-plane directions of [1 10] and [001] form an asymmetric zig-zag line. Such a configuration can be understood from the point of view of domain-wall energy. Unlike Part I and Part II, the focus of Part III will be on the domain walls rather than on the domains themselves.

Chapter 4 then deals with domain walls in thicker Fe films on W(110). There, the
system is characterized by a uniaxial anisotropy with the [001] direction as the easy axis. Domain walls separating head-to-head or tail-to-tail domains are found to assume the form of characteristic zig-zag lines. It is a fortunate coincidence that the width of these domain walls is on a scale large enough to allow a thorough, spatially resolved analysis of how the magnetization direction changes across the wall.

Almost forty years ago, head-to-head domain walls were suggested as basic ingredients for developing magnetic logic networks driven by a magnetic field [183–186]. In these so-called domain-tip propagation logic (DTPL) devices, the magnetic field moves head-to-head domain walls along pre-defined channel-like structures with low coercivity. More recently, with the advent of activities in the field of spintronics, head-to-head domain walls have again become a subject of considerable interest. Berger has calculated a number of different ways a domain wall can be displaced by letting an electric current flow across a domain wall [187–190]. Berger’s ideas have been invoked to explain experimental results in systems with head-to-head domain walls [191–197]. A detailed experimental characterization of such head-to-head domain walls is desirable in order to truly understand the physical mechanisms involved in domain wall motions induced by an electric current.

In the head-to-head domain walls investigated here, the magnetization vector remains within the plane of the film even inside the domain walls. These domain walls are Néel domain walls, which tend to occur in thinner films [198]. Néel walls are charged. They carry a magnetic charge density per unit wall area of \( \sigma_m = 2M \) where \( M \) is the magnitude of the magnetization vector. Charged domain walls in three-dimensional bodies are very rare as the stray field produced by a charged domain wall does not fall off with distance. The stray field generated by a line charge in an essentially two-dimensional magnetic body, however, falls off like \( 1/r \) where \( r \) is the distance from the domain-wall line. The magnetostatic energy associated with charged domain walls in two-dimensional films is thus not prohibitively large. It plays, however, an essential role in determining both the overall symmetric zig-zag shape of the domain walls and the microscopic structure of the magnetization vector within the domain walls. Despite the overall symmetric zig-zag shape of the domain walls, an asymmetric configuration of the magnetization vector is found on a truly microscopic scale, and this is due to the magnetic charge.
Chapter 2

Experimental Procedures

The methods employed for producing and characterizing thin Fe films on W(110) are similar to those used for ultrathin Fe films on Cu(001) and discussed in Sec. 3.1 of Part I. However, there are some differences. A different procedure has to be adopted to prepare the substrate. Moreover, the films studied here are substantially thicker than the ultrathin Fe films used in Part I and Part II. This requires another procedure for determining the film thickness. Finally, the focus here is on the in-plane components of the magnetization. It is shown that the setup originally conceived for measuring the out-of-plane and one in-plane component of the magnetization vector can be used to determine two mutually perpendicular in-plane components simultaneously.

2.1 Sample Preparation

The W(110) substrate was prepared according to the procedure outlined in [199]. The most important contaminant of tungsten is carbon, which originates in the bulk and segregates at the surface when tungsten is heated. The carbon contamination is reduced when tungsten is heated and simultaneously exposed to oxygen. Here, the W(110) substrate was typically held at 1770 K during 20 min while the oxygen pressure in the preparation chamber was set to $4 \cdot 10^{-8}$ mbar. Subsequent flashing at temperatures around 2270 K was used to remove the residual oxygen contamination on the tungsten surface. The cleanliness of the W(110) substrate was verified with Auger Electron Spectroscopy.

The Fe films were then deposited using Molecular Beam Epitaxy. Although the settings were very similar to the ones used for ultrathin Fe films on Cu(100), the evaporation rate turned out to be somewhat higher. The deposition rate as determined from Auger electron spectra was roughly 1.8 ML/min.

All experiments were performed on wedge-shaped samples (cf. Part I, Subsec. 3.2.1). The reorientation transition was observed on the slope; head-to-head domain walls were investigated on the flat area following the slope.
2.2 Determination of Film Thickness

The thickness of the investigated Fe films on W(110) greatly exceeded the mean free path of Auger electrons so that no sizable contribution from the substrate could be detected in Auger electron spectra taken on the Fe films. The film thickness had thus to be calculated as the product of deposition time and deposition rate. The deposition rate was determined separately on films thin enough to be susceptible to the method of film thickness determination using Auger electron spectra described in Part I, Subsec. 3.4.3.

2.3 Determination of In-Plane Magnetization

The setup of the SEMPA chamber is such that the out-of-plane and one in-plane component of the magnetization (or rather the polarization) vector can be measured simultaneously (cf. Part I, Subsec. 3.4.1). When no out-of-plane component is present, the other in-plane component can be measured as well by tilting the sample slightly along the horizontal axis connecting the sample and the opening leading to the Mott detector, cf. Fig. 2.1. While the originally measured in-plane component is unaltered, the other in-plane component shows up as a signal in the original out-of-plane image.

As has been shown by Baek et al. [179], the polarization is independent of the orientation along one of the two in-plane directions [110] and [001]. In order to calculate the magnetization direction at each pixel, the two in-plane magnetization components have to be normalized. For this purpose, the center of the Gaussian distribution of the magnetization components in extended areas uniformly magnetized along one of the two in-plane directions is computed. Once this is done for a particular image, the calculation of the magnetization directions in this image is straightforward.

![Figure 2.1: Measuring two in-plane components. The sample is mounted on the slope of the wedge. The original setup allows measuring the magnetization component perpendicular to the film plane as well as the magnetization component along the gradient. When the sample is slightly rotated around the axis indicated by the black arrow, the in-plane magnetization indicated by the dashed black arrows provides a contribution along the original perpendicular direction.](image-url)
The switching of the easy magnetization axis from parallel to the [110] direction to parallel to the [001] direction for thin films of Fe(110) on W(110) was first reported by Gradmann et al. [180]. A similar switching had, however, already been observed in the similar system of Fe(110) on GaAs(110) [200]. A competition of surface and bulk anisotropies has been identified as the cause for the switching [180–182].

The influence of bulk anisotropy increases with film thickness. Using the directional cosines

\begin{align}
\alpha_1 &= \sin \theta \cdot \cos \varphi, \\
\alpha_2 &= \sin \theta \cdot \sin \varphi, \\
\alpha_3 &= \cos \theta
\end{align}

with respect to a Cartesian system with the z axis parallel to the surface normal and the x axis along the [001] direction, the bulk anisotropy energy per unit volume can be expressed as

\begin{equation}
f = K_1 \left( \alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2 \right).
\end{equation}

If the magnetization vector is restricted to lie within the plane of the film, this expression simplifies to [180]:

\begin{equation}
f_\parallel = \frac{K_1}{4} \left( \sin^2 \varphi + \sin^4 \varphi \right).
\end{equation}

Due to the relatively large lattice mismatch for bcc Fe(110) on W(110) of −9.4 %, elastic strain energy cannot be neglected. This adds an extra term to the bulk anisotropy so that the bulk anisotropy energy per unit volume contains two independent constants \( K_{v,p} \) and \( K_{v,pp} \) [181]:

\begin{equation}
f_\parallel = K_{v,p} \sin^2 \varphi + K_{v,pp} \sin^4 \varphi.
\end{equation}
Part III: Chapter 4. 90° Domain Walls

The positive signs of the bulk anisotropy constants \( K_1 = 4.8 \times 10^4 \text{ J/m}^3, \) \( K_{v,p} = (10.5 \pm 1.0) \times 10^4 \text{ J/m}^3, \) \([182]\) indicate that an orientation of the magnetization vector along the [001] direction is favored by bulk anisotropy.

The surface anisotropy energy of the Fe(110) surface per unit area due to interatomic interactions can be written as

\[
\sigma = K_s \cdot \alpha_3^2 + K_{s,p} \cdot \alpha_1^2 = K_s \cdot \cos^2 \theta + K_{s,p} \cdot \sin^2 \theta \cdot \cos^2 \varphi ,
\]

which simplifies to

\[
\sigma_\parallel = K_{s,p} \cdot \cos^2 \varphi
\]

if only in-plane magnetizations are considered \([180]\). The positive sign of \( K_s \) \( (K_s = 1.0(2) \times 10^{-3} \text{ J/m}^2, K_{s,p} = 0.61(3) \times 10^{-3} \text{ J/m}^2, \) \([182]\) means that the [110] direction is favored by this magnetocrystalline contribution to the surface anisotropy.\(^1\) Another contribution to the surface anisotropy arises from the shape anisotropy. The magnetostatic energy is reduced when the magnetization vector lies within the plane of the film. At a certain thickness \( D_c \) where the bulk anisotropy energy balances the surface anisotropy energy, a reorientation transition of the magnetization vector from along the [110] direction to the [001] direction is to be expected.

The reorientation transition as observed on a wedge is shown in Fig. 3.1. While the magnetization vector is found to be parallel to the [110] direction favored by the surface anisotropy in areas thinner than \( D_c \), the magnetization vector is parallel to the [001] direction favored by the bulk anisotropy in areas thicker than \( D_c \). In the narrow region where the transition occurs, both areas magnetized along [110] and areas magnetized along [001] are present. These areas are separated by a thin zig-zag domain wall. There, the magnetization vector changes on a very short scale. However, the modulus of the magnetization within each domain remains constant. This is consistent with the result obtained by Baek et al. that the sum of the magnetization components remains constant across the reorientation transition \([179]\).

Fig. 3.2 depicts the magnetic configuration at the reorientation transition for a

\(^1\) Based on Néel’s model of surface anisotropy, which relies on a pairwise anisotropic atomic interaction energy of

\[ w = l \left( \cos^2 \varphi - \frac{1}{3} \right) , \]

Elmers et al. calculated the surface anisotropy constants as:

\[
K_s = -\frac{2\sqrt{2}}{3a^2} l , \quad (3.10)
\]

\[
K_{s,p} = -\frac{\sqrt{2}}{3a^2} l , \quad (3.11)
\]

where \( a \) is the lattice parameter \([181, 182]\). The ratio of \( K_s \) and \( K_{s,p} \) was confirmed experimentally.
a, Magnetization component along the [1¯10] direction favored by the surface anisotropy. At small thicknesses the magnetization is oriented along the [1¯10] direction, after the reorientation transition, this magnetization component vanishes.

b, Magnetization component along the [001] direction favored by the bulk anisotropy. At large thicknesses the magnetization is oriented along the [001] direction, before the reorientation transition, this magnetization component vanishes.

\( w_{a,b} = 4.6 \, \mu\text{m}, D_{a,b} = 39 \, \text{ML}, T_{a,b} = 293 \, \text{K} \)
Figure 3.2: Reorientation transition in iron films on W(110). When the film thickness is increased beyond about 39 ML, the uniaxial easy magnetization axis changes from parallel to the [110] in-plane direction to parallel to the perpendicular in-plane direction of [001]. Figs. a, c, and d show the asymmetric zig-zag domain walls that form at the transition. The film thickness rises by 0.1 ML from left to right within each image. The local magnetization direction is color-coded. The mapping between direction and color is given in b together with the mutually perpendicular in-plane directions [110] and [001]. ($w_{a,c,d} = 4.6 \mu m$, $D_{a,c,d} = 39 ML$, $T_{a,c,d} = 293 K$)

magnetization is equal on both sides of the domain wall, $\sigma_m$ is given by

$$\sigma_m = M(\cos \vartheta_1 - \cos \vartheta_2) ,$$

(3.13)
where $\vartheta_i$ is the angle between the surface normal $\vec{n}$ and the magnetization direction on side $i$. If we impose the restriction that $\vartheta_1 - \vartheta_2 = \pi/2$—as is the case at the reorientation transition—we obtain

$$\sigma_m = M(\cos \vartheta_1 - \sin \vartheta_1) .$$

(3.14)

The magnetic charge density per unit wall area $\sigma_m$ vanishes for $\vartheta_1 = \pi/4$. As the reorientation transition is observed on a wedge, a global angle of $\vartheta_1 = \pi/4$ is made impossible by the shape of the wedge. The overall situation is that given in Fig. 3.3a. This configuration is associated with a high charge density $\sigma_m$. The magnetostatic energy of a distribution of magnetic charges can be calculated like the electrostatic energy of an equivalent electric charge distribution [23]:

$$E_d = \frac{\mu_0}{8\pi} \int_V \int_V \frac{\rho_m(\vec{r})\rho_m(\vec{r}')}{|\vec{r} - \vec{r}'|} d^3r d^3r' .$$

(3.15)

The quantity $\rho_m$ is the magnetic charge density per unit volume $-\text{div}\vec{M}$. As the domain wall considered here has a net magnetic charge, it will interact with other magnetic charges. However, other magnetic charges on the wedge are far apart and minor modifications of the domain wall geometry will not entail significant changes of the interaction energy with these other magnetic charges. The following considerations will therefore be focused on the magnetostatic energy of magnetic charges directly involved in the formation of the wall. The magnetostatic energy of the configuration shown in Fig. 3.3a could be reduced by simply spreading the negative magnetic charge over a larger band. This would mean that the magnetization direction switches more slowly and increase the exchange energy. The exchange energy limits the domain wall width. The domain wall width found at the reorientation transition is rather small. In Chapter 4, wider domain walls will be found and the details of the magnetization switching within the domain wall will be analyzed. The magnetostatic energy of the wall in Fig. 3.3a, however, can also be reduced when the wall assumes a zig-zag line as in Fig. 3.3b. The magnetic charge on each segment of the zig-zag domain wall is reduced with respect to the magnetic charge in the configuration of Fig. 3.3a. The angles between the bisectors of the teeth and the magnetization vectors on either side of the wall are very close to $\pi/4$. If the domain walls were along this direction, there would be no magnetic charge. But the domain walls have to be off this favorable direction in order to enable the overall domain wall orientation shown in Fig. 3.3a. A larger angle at the teeth tip reduces the domain wall length and thus the exchange energy while increasing the magnetostatic energy. The angle at the teeth tips is therefore determined by the competition of the exchange energy with the magnetostatic energy.

Based on calculations by Aharoni [201], Jakubovics obtained approximate expressions for the exchange energy, the anisotropy energy, and the magnetostatic energy.
Figure 3.3: Model for the charge at an asymmetric zig-zag wall. a, A 90° domain wall that is parallel to the magnetization within one of the two domains is associated with a high magnetic charge per unit domain wall length, which is energetically unfavorable. b, The density of the magnetic charge per domain wall length is reduced when the straight domain wall is transformed into a zig-zag wall. The magnetic charge density can now be thought of as distributed over a larger area, which reduces the magnetostatic energy. A longer domain wall length, however, increases the domain wall energy. Positive magnetic charges are indicated by a plus sign (+), negative magnetic charges by a minus sign (−).

of an infinitely extended straight charged domain wall [202]. He found that the magnetostatic energy of the wall was proportional to \( \sin^2\left(\frac{1}{2}\psi - \vartheta_0\right) \), where \( \psi \) is the angle by which the magnetization vector rotates across the wall and \( \vartheta_0 \) is the angle between the magnetization vector in one of the domains and a vector normal to the wall. If we assume the bisector of a tooth to enclose an angle of exactly \( \pi/4 \) with the magnetization vectors on either side of the domain wall and denote the angular deviation of a domain wall segment from the direction of the bisector as \( \alpha \), the change of the magnetostatic energy with \( \alpha \) is accordingly found to be proportional to \( \sin^2(\alpha) \). This is not surprising since the magnetic charge density per unit wall area is given by

\[
\sigma_m = M \left\{ \cos \left( \frac{\pi}{4} - \alpha \right) - \sin \left( \frac{\pi}{4} - \alpha \right) \right\} = \sqrt{2} M \sin(\alpha) .
\]  

(3.16)

For a visualization of the quantities used in this discussion, cf. Fig. 3.4 a. The ratio of the domain-wall length of a tooth to the length covered along the overall domain
Figure 3.4: Model calculation for the asymmetric zig-zag domain wall. a, Definition of the angle $\alpha$, the length of the long tooth side $k$, the length of the short tooth side $l$, and the distance along the overall domain-wall direction $x$. The dotted line is the bisector. b, The normalized domain-wall energy $E/B$ as a function of the angle $\alpha$. The ratio $A/B$ is set to 0.07 such as to obtain the experimentally observed angle of $\alpha = 15^\circ$, which corresponds to 0.26 rad.

Wall direction is given by:

$$\frac{k + l}{x} = \frac{1}{\sin\left(\frac{\pi}{4} + \alpha\right) - \cos\left(\frac{\pi}{4} + \alpha\right)} = \frac{1}{\sqrt{2} \sin(\alpha)},$$

(3.17)

where $k$ is the length of the longer side of the tooth, $l$ the length of the shorter side of the tooth, and $x$ is the distance of the tooth basis along the overall domain-wall direction. The total energy of the wall per unit length along the overall domain wall direction then reads:

$$E = \left\{ A + B \sin^2(\alpha) \right\} \cdot \frac{k + l}{x} = \frac{1}{\sqrt{2}} \left\{ \frac{A}{\sin(\alpha)} + B \sin(\alpha) \right\},$$

(3.18)

where $A$ comprises the exchange energy, the anisotropy energy and part of the magnetostatic energy; $B$ denotes that part of the magnetostatic energy that varies with $\alpha$. For not too large values of $A$, the angle $\alpha$ will assume a finite value. The minimum energy is obtained for $\sin^2(\alpha) = A/B$. Using the experimentally determined
value for $\alpha$ of around 15°, the ratio $A/B$ can be estimated to be 0.07. The normalized energy $E/B$ for this ratio is plotted in Fig. 3.4b. Note that in these considerations, the magnetostatic energy between the individual domain-wall segments has been neglected. This interaction favors smaller angles $\alpha$.

Dressler and Judy calculated the magnetostatic energy of a single straight homogeneously charged domain-wall section of finite length and finite thickness as well as the magnetostatic interaction energy of a symmetric zig-zag wall [203]. They found the interaction energy per unit length along the overall domain-wall direction to amount to only a few percents of the magnetostatic self-energy of a single straight domain-wall section, again calculated per unit length along the overall domain-wall direction. A rough estimate of the domain-wall energy due to the exchange interaction and anisotropy yielded an energy on the order of a few percents of the magnetostatic self-energy in the case of 100 nm thick films of cobalt and cobalt-chromium. This led to a value of $A/B$ similar to the value determined here.

The thickness range over which the asymmetric zig-zag domain wall is found is given by the anisotropy. Areas inside a tooth but close to the tip are magnetized in a direction perpendicular to the local easy direction for the magnetization. This adds a significant contribution to the wall energy and limits the extent of the asymmetric zig-zag domain wall. The reorientation transition occurs at a film thickness of around 80 Å (39 ML), which is in fair agreement with the transition thickness of 85 Å found by Baek et al. [179]. The reorientation transition is completed within less than one monolayer. This is again consistent with the observations by Baek et al. [179].
Chapter 4

180° Domain Walls in Thicker Films

The asymmetric zig-zag walls in thin Fe films on W(110) discussed in Chapter 3 occur due to the reorientation of the easy axis for the magnetization as the film thickness is increased. The subject of this chapter are domain walls that show up in thicker Fe films on W(110), i.e. in a thickness range where the easy axis for the magnetization is along the [001] direction. Here, the magnetostatic energy favors a single-domain state. Experimentally, however, multi-domain configurations do occur and the overall geometry of the domain walls is irregular and given by the history and imperfections of the films. Locally, the domain walls consist of straight sections that form a zig-zag line. The angle enclosed by the straight sections and the easy axis for the magnetization reflects the balance between the various energy contributions. The local structure of the domain walls is independent of the overall domain configuration.

The domain walls in thicker films considered here separate domains that meet head to head or tail to tail. There is thus a symmetry between walls enclosing an angle of $\alpha$ with the easy axis for the magnetization and walls enclosing an angle of $-\alpha$. Locally, the zig-zag line is thus symmetric with respect to the easy axis. Experimentally observed symmetric zig-zag domain walls are shown in Fig. 4.1.

The thickness of the films investigated here is in a range where the magnetization vector remains in the plane of the film throughout the domain walls. No out-of-plane component of the magnetization can be detected. The magnetization within the domain walls is visible in Figs. 4.1a and b. Calculations by Jakubovics [202] and Hubert [204] show that the width of charged Néel walls increases with the charge density of the wall. Indeed, the internal structure of the 90° walls discussed in Chapter 3 could not be resolved, but the internal structure of the 180° walls in thicker films is revealed in high-resolution images. On crossing the domain wall, the magnetization rotates either clockwise or counterclockwise. This breaks the
Figure 4.1: Domain walls in thicker iron films on W(110). a, In thicker iron films on W(110), the uniaxial easy axis for the magnetization is along the [001] direction. When domains meet head to head, the domain wall separating them assumes the form of a zig-zag line. b, The magnetization within the domain wall remains in the plane of the film. The symmetry between a clockwise and a counterclockwise rotation of the magnetization from left to right is broken by external factors, such as a weak magnetic field. This image shows a short section of the domain wall where the magnetization within the domain wall is reversed (blue). ($w_a = 11.5 \mu m$, $w_b = 2.3 \mu m$, $D_a = 74$ ML, $D_b = 86$ ML, $T_{a,b} = 293$ K)

symmetry between the two in-plane directions perpendicular to the easy axis for the magnetization.\footnote{The magnetization configuration across the domain wall corresponds to a wall structure considered by Finzi and Hartmann in their Fig. 5(b) \cite{205}. In their analysis of zig-zag domain walls in permalloy films with a thickness of 200 nm, Finzi and Hartman finally discarded this wall structure in favor of a Bloch spine and curling regions.} In the experiment, the direction of the magnetization within the domain walls seems to be influenced by the residual magnetic field within the SEMPA chamber. However, sections with a reversed magnetization direction can be found. Such a case is shown in Fig. 4.1b.

As in the case of the asymmetric zig-zag walls in Chapter 3, the formation of the symmetric zig-zag walls in thicker films can be understood as the result of the competition between the magnetostatic energy of the magnetic charges and the
Figure 4.2: Model for the charge at a symmetric zig-zag wall.  

a, A straight domain wall that separates two oppositely magnetized domains and is perpendicular to the magnetization axis is characterized by a large magnetic charge density per domain wall length.  
b, The negative magnetic charge is distributed over a larger area when the domain wall forms a zig-zag line. This reduces the magnetostatic energy but increases the domain wall energy.  
c, A detailed look at the domain wall structure reveals that the magnetization rotates over a finite width. A prompt change of the magnetization direction would entail a large energy due to the exchange interaction. On a microscopic level, a separation of positive and negative charges occurs. The modulus of the magnetic charge on the two sides is not identical. In the situation depicted above, the modulus of the negative charge density per wall length distinctly exceeds the modulus of the positive magnetic charge density on the other side of the wall.  

Positive magnetic charges are indicated by a plus sign (+), negative magnetic charges by a minus sign (−).
Figure 4.3: Model calculation for the symmetric zig-zag domain wall. a, Definition of the angle $\alpha$, the length of the tooth side $k$, and the distance along the overall domain-wall direction $x$. The dotted line is the bisector. b, The normalized domain-wall energy $E/B$ as a function of the angle $\alpha$. The ratio $A/B$ is set to 0.18 such as to obtain the experimentally observed angle of $\alpha = 25^\circ$, which corresponds to 0.26 rad.

Exchange and anisotropy energies per unit length along the direction of a section. The magnetostatic energy favors a small angle $\alpha$ because the magnetic charge is spread over a larger area in such a case. The charge per unit length along the direction of a domain-wall section is reduced. The magnetic charges associated with a straight domain wall and a symmetric zig-zag domain wall are depicted in Figs. 4.2a and b respectively. Exchange and anisotropy energies, however, scale with the length of the domain wall and thus increase as the angle $\alpha$ is reduced. Using the quantities given in Fig. 4.3a, we obtain the following expression for the length of the straight domain wall sections per unit length along the direction perpendicular to the easy direction for the magnetization:

$$\frac{2k}{x} = \frac{1}{\sin \alpha} .$$  \hspace{1cm} (4.1)

The magnetostatic energy is proportional to the square of the charge density, which scales as $\sin \alpha$. Thus, the total energy per unit length along the direction perpendicular to the easy axis is given by

$$E = \{A + B \sin^2 \alpha\} \cdot \frac{2k}{x} = \frac{A}{\sin \alpha} + B \sin \alpha ,$$  \hspace{1cm} (4.2)
where exchange and anisotropy energies are represented by $A$, and the magnetostatic energy is taken into account by the constant $B$. The dependence of the total energy $E$ on the angle $\alpha$ is shown in Fig. 4.3b. The ratio $A/B$ is chosen such as to obtain the minimum total energy for the experimentally observed angle on the order of $25^\circ$. The enhanced value of the ratio $A/B$ with respect to the case of the asymmetric zig-zag domain walls at the reorientation transition may be due to the increased anisotropy energy in thicker films.

Riedel and Seeger analyzed uncharged Néel walls analytically and showed that such walls can be decomposed into a narrow core and extended tails [206], thereby confirming previous numerical calculations. In the tails, the direction cosine was found to decrease logarithmically with distance. The dilute charge in the tails caused an increase of the anisotropy energy. Freiser explained the formation of zig-zag domain walls as the result of the competition between the magnetostatic energy and the anisotropy energy caused by extended tails [207]. Freiser obtained expressions for the angle of the vertex and the length of the straight sections that reproduced the experimentally observed values in amorphous Gd-Co films. In their experimental investigation of cross-tie walls forming a zig-zag line, Dressler and Judy found a relation between the coercivity of their films and the amplitude of the zig-zag structure [203]. In the present system of thin Fe films on W(110), the length of the straight wall sections varies substantially. It is not clear whether there is an intrinsic scale for the length of the straight sections. It will be shown below that most of the magnetic charge density is concentrated in a relatively narrow core so that a description in terms of a competition between magnetostatic energy and anisotropy energy due to extended tails seems inappropriate.

The direction of the magnetization in the interior of the straight domain sections affects the distribution of the magnetic charges. Fig. 4.2c shows a detailed sketch of a straight section separating two domains magnetized in opposite directions. The in-plane rotation of the magnetization vector across the domain wall causes a separation of positive and negative magnetic charges. On the side of the domain wall where the magnetizations within the domain and within the domain wall meet tail to tail, the magnetic charge density is larger than the total magnetic charge density. On the other side of the wall, the magnetic charge is of the opposite sign but smaller in magnitude than where the magnetizations meet tail to tail. The unequal distribution of the magnetic charges has an impact on the magnetization profile across the domain wall. The asymmetry of the charge distribution entails an asymmetry of the magnetization profile.

On the side of the wall with the higher charge density, the density is spread over a wide band so as to reduce the magnetostatic energy. Consequently, the direction of the magnetization changes more slowly than on the side with the lower charge density. This is especially conspicuous at the vertices. The magnetization configuration at vertices is shown in Fig. 4.4 at different magnifications.
Due to the measuring procedure where one of the in-plane components is measured as a small contribution to the out-of-plane signal of the original setup, one of the in-plane components is marked by a significant noise. In order to obtain the magnetization profile across a straight section of the domain wall, the magnetization components are therefore averaged over a large number of parallel lines running along a direction perpendicular to the domain wall. Figs. 4.5a and c show the locations where magnetization profiles have been determined across domain walls. The magnetization components both parallel and perpendicular to the easy axis and parallel and perpendicular to the domain wall have been measured. The profiles are presented in Figs. 4.6–4.9. The symbols used to identify the various magnetization components are explained in Figs. 4.5b and d. The various profiles of Figs. 4.6–4.9 are given in order to show that the magnetization profiles can be determined quite reproducibly with the method adopted here. In the following discussion, we will concentrate on the profile presented in Fig. 4.6. The same discussion can be applied to the profiles presented in Figs. 4.7–4.9.

Note that the width of the domain walls is an order of magnitude larger than the film thickness. Using Lorentz microscopy, Chapman et al. investigated 180° walls in thin foils of a Ni-Fe-Co-Ti alloy and found that their measurements were compatible with the assumption of a vortex-like magnetization distribution in the cross section of the wall [208]. In their experiments, the film thickness was of the same order as the domain-wall width. Here, a vortex-like magnetization distribution can be ruled out. The small thickness of the film as compared to the domain-wall width suggests that the measured in-plane magnetization distribution reflects the magnetization distribution throughout the film thickness.

Fig. 4.6a shows the magnetization components parallel and perpendicular to the easy axis and the wall for wall I indicated in Fig. 4.5a. The magnetization component parallel to the easy axis changes to the opposite polarity within a distance on the order of 200 nm. The component perpendicular to the easy axis, however, varies on a considerably larger distance on the order of 500 nm. It increases even beyond the point where the parallel component vanishes. Its maximum is shifted towards the side of the domain wall that has the higher charge density. In all the profiles of Figs. 4.6–4.9, the side with the higher charge density is on the right-hand side of the plots. Note that the perpendicular component does not show any plateau within the domain wall. Therefore, the area with the magnetization perpendicular to the easy axis cannot be interpreted as belonging to a separate domain lying between the two domains magnetized along the easy axis. The parallel component also varies continuously over the domain wall. Fig. 4.6b displays the variation of the angle of the magnetization across the domain wall. The angle changes rather abruptly on the side with the low magnetic charge density but varies slowly on the side with the large charge density. The rotation of the magnetization is completed after a distance of the order of 500 nm, which is the range over which the magnetization component
Figure 4.4: Magnetization tails at symmetric zig-zag walls in thicker iron films on W(110). a, b, In symmetric zig-zag domain walls, the two sides of a straight domain wall segment are not symmetric. While the magnetization direction switches quickly on the weakly charged side, an extended tail is visible on the strongly charged side. There, the magnetization direction changes only over larger distances. c, d, Detailed images of the magnetization direction distribution at the tips of symmetric zig-zag walls reveal an asymmetric configuration. The symmetry is broken by the magnetization direction within the domain wall. ($w_{a,b} = 4.6 \, \mu m$, $w_{c,d} = 0.9 \, \mu m$, $D_{a,c} = 74$ ML, $D_b = 67$ ML, $D_d = 86$ ML, $T_{a,b} = 293$ K)
Figure 4.5: Location and orientation of cross sections. Figs. 4.6, 4.7, 4.8, and 4.9 show how the magnetization components change across a straight segment of the symmetric zig-zag wall. The magnetization components are measured along lines perpendicular to the domain wall. Location and orientation of these cross sections are indicated by white arrows in Figs. a and c. These figures are identical to Figs. 4.4 a and c. Figs. b and d show the magnetization components with their respective labels. I: Fig. 4.6, II: Fig. 4.7, III: Fig. 4.8, IV: Fig. 4.9. (w_a = 4.6 µm, w_c = 0.9 µm, D_a,c = 74 ML, T_{a,b} = 293 K)
perpendicular to the easy axis deviates from zero.

It has been argued that the domain walls in thin films should have similar properties as the surface region of domain walls in bulk materials [27]. Using SEMPA, Oepen and Kirschner investigated the surface region of 180° domains in bulk iron [209]. Indeed, they find that the Bloch-type walls of the bulk terminate as Néel-type structures on the Fe(100) surface with a width of (210 ± 40) nm. This is of the same order of magnitude as the width of the Bloch wall in the bulk obtained using the strengths of anisotropy and exchange. This view has been challenged by Scheinfein et al. [210, 211]. While confirming experimentally the width of the Néel-type structure at the Fe(100) surface to be around 225 nm, they use micromagnetic calculations to relate this width to the width of the Bloch wall in the bulk and find that the width of the Néel-type structure at the surface is at least the double of the Bloch-wall width. They find the transition from the Bloch-type wall to the Néel-type wall to occur over a distance on the order of the Bloch-wall width. Here, in thin films where the wall can be expected to be of the Néel type throughout the interior of the film, we find that the width of the Néel-type wall is rather larger than for the Néel-type structure at the surface of bulk iron. It is on the order of 200–500 nm, depending on the quantity used to define the wall width.

Profiles of charged walls in thin magnetic films have been calculated by Hubert for a range of configurations [204]. For 180° domain walls, Hubert has determined the wall profile as a function of the angle between the wall and the easy direction. The two symmetric limiting cases where the wall is either parallel or perpendicular to the easy direction are characterized by (point) symmetric wall profiles. In the intermediate cases, however, Hubert finds that the wall profile is asymmetric, i.e. the variation of the magnetization across the domain wall is asymmetric. According to Hubert’s calculations, the magnetization angle varies quickly on the lightly charged and slowly on the heavily charged side of the wall. The experimental findings presented in this chapter are in good qualitative agreement with these calculations.

The detailed knowledge of the magnetization vector across the domain wall can be exploited to calculate the distribution of the magnetic charge density. Although the wall profiles have been obtained by averaging over a large number of parallel lines perpendicular to the walls, the noise is still considerable. The magnetic charges can only be retrieved from the wall profiles after a number of smoothing operations. Fig. 4.10 shows the distribution of the magnetic charges across wall I of Fig. 4.5 extracted from the wall profile of Fig. 4.6b. The magnitude of the magnetic charge density reaches its maxima near the center of the domain wall. There the magnetic charge density changes sign abruptly. The magnitudes of the maxima of the positive and negative charges are of the same order, but the modulus of the total magnetic charge on the right-hand side is twice as large as on the left-hand side. The additional magnetic charge on the right-hand side is located in a tail of considerable length. This imbalance of the magnetic charges has to be expected from
Figure 4.6: Magnetization components across a straight segment of the symmetric zig-zag wall I. a. Magnetization components along [001] (solid circles), along [110] (open circles), parallel to the wall (solid triangles), and perpendicular to the wall (open triangles). b. The angle of the magnetization switches quickly on the weakly charged side of the wall (left) but relaxes only slowly on the strongly charged side of the wall (right). Both location and orientation of the wall cross section are indicated in Fig. 4.5a.
Figure 4.7: Magnetization components across a straight segment of the symmetric zig-zag wall II. a, Magnetization components along [001] (solid circles), along [110] (open circles), parallel to the wall (solid triangles), and perpendicular to the wall (open triangles). b, The angle of the magnetization switches quickly on the weakly charged side of the wall (left) but relaxes only slowly on the strongly charged side of the wall (right). Both location and orientation of the wall cross section are indicated in Fig. 4.5 a.
Figure 4.8: Magnetization components across a straight segment of the symmetric zig-zag wall III. a, Magnetization components along [001] (solid circles), along [110] (open circles), parallel to the wall (solid triangles), and perpendicular to the wall (open triangles). b, The angle of the magnetization switches quickly on the weakly charged side of the wall (left) but relaxes only slowly on the strongly charged side of the wall (right). Both location and orientation of the wall cross section are indicated in Fig. 4.5b.
Figure 4.9: Magnetization components across a straight segment of the symmetric zig-zag wall IV. 

a. Magnetization components along [001] (solid circles), along [110] (open circles), parallel to the wall (solid triangles), and perpendicular to the wall (open triangles).

b. The angle of the magnetization switches quickly on the weakly charged side of the wall (left) but relaxes only slowly on the strongly charged side of the wall (right).

Both location and orientation of the wall cross section are indicated in Fig. 4.5b.
Figure 4.10: Magnetic charge density across a straight segment of the symmetric zig-zag wall I. The total charge on the left-hand, positively charged side is about half the total charge on the right-hand, negatively charged side. The extra charge on the right-hand side is distributed in the tail. Both location and orientation of the wall cross section are indicated in Fig. 4.5a.

The overall geometry of the wall, cf. Fig. 4.2c, and confirms that the slow variation of the magnetization vector on the heavily charged side of the domain wall is due to a smearing out of the excess charge, which reduces the magnetostatic energy. Counteracting this tendency of the magnetic charge is the anisotropy energy. It limits the width of the domain wall. Based on the experimentally determined wall angle $\alpha$, cf. Fig. 4.3a, the ratio of the total magnetic charges on the two sides can be calculated as

$$\frac{\rho_+}{\rho_-} = \frac{\sin \alpha + \cos \alpha}{\sin \alpha - \cos \alpha} \approx -3.$$  \hspace{1cm} (4.3)

This deviates from the ratio of approximately $-2$ determined from Fig. 4.10. The representation of the charge distribution of Fig. 4.10 is only obtained after several steps of data processing and should thus be considered as rather qualitative than quantitative. Nevertheless, the analysis yields reasonable information about the distribution of the magnetic charge density across a domain wall.
Bibliography


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ACKNOWLEDGMENT

In the course of this dissertation, several people contributed in one way or another. But first of all, I would like to express my gratitude to my dissertation supervisor Danilo Pescia for the opportunity to carry out this thesis in his group. I enjoyed the great trust and freedom he gave me in my work. His continuing interest in my work and his enthusiasm about new results encouraged me in my work both with the experiments and the ensuing analysis. Besides that, he also made many very important contributions and was my main collaborator.

The subject of this thesis touches on a range of fields that, in the beginning, we were not familiar with. It was therefore a hard work to place the results into an appropriate context. I benefited from his broad overview, his help, his intuition, and his strong capability of making out the key points. As the findings in this thesis extend into a number of areas previously unknown to us, Danilo Pescia suggested writing a somewhat more extensive dissertation. I would like to thank him for giving me the time to do this.

In the beginning of my doctoral studies, I followed the lecture on magnetism given jointly by Christian Back and Rolf Allenspach. This lecture deepened my understanding of magnetism in general and proved beneficial in several stages of my research. I felt very honored when they readily agreed to act as co-examiners of this dissertation despite their many other obligations. I would also like to thank them for carefully reading this dissertation despite its length and their helpful comments. I enjoyed the company of Christian Back while he was still in our group as well as assisting Rolf Allenspach in his lectures on magnetism.

Research on the main topic of this thesis, domain patterns in ultrathin iron films on Cu(001), was initiated here by Andreas Vaterlaus and Christian Stamm. Christian Stamm was my predecessor as a Ph.D. student at the SEMPA. I was lucky that I could still share some time with him. I thank him for helping me get acquainted with the experimental apparatus in the laboratory, his good sense of humor and our many stimulating discussions. Andreas Vaterlaus also introduced me to the work with the SEMPA. I was fortunate to be able to profit from his thorough knowledge of the experimental apparatus and his readiness to help whenever problems occurred. I would also like to thank him for the assistance he lent me in projects not reported in this thesis. It was a great pleasure to join him for experiments at the Swiss Light Source in Villigen.
I would like to thank Urs Maier for the excellent experimental equipment with the easy-to-use wobble sticks. In the beginning, when my hands were tired and shaky after a long day of wobble-stick manipulations, however, I was grateful when he helped me put the samples to a safe place so that I could continue the day after. I also thank him and Andreas Vaterlaus for the pleasant Ferrovac dinners.

At a time when the various magnetic domain patterns and their systematics were still largely mysterious or even unknown, I was happy to have the company of Giovanni Isella. We had a good time together in the laboratory and many stimulating and controversial discussions about possible systematics. I thank him for this memorable time. I like recalling the days when I was his guest in Como.

The strong dependence of the magnetic domain pattern on preparation conditions has always fascinated me. Therefore, I appreciated it when Thomas Flückiger provided a CCD camera and offered to help me in performing the LEED measurements to gain at least some insight into the relation between film structure and magnetic domain patterns.

Tuomas Knowles was a student we raised almost from the beginning of his studies at ETH, having been a student in my supervision class in his first two years. Our efforts were greatly rewarded when he joined our group for doing his degree thesis. I enjoyed the close and intensive collaboration during that time. The result of this work is a program that uses a binarized version of the domain patterns to produce an accurate representation of the director field. It is a great pleasure for me to thank him for his highly valuable contribution that made possible the analysis of orientational order in theses patterns and his never-ending commitment to strive for the best possible results.

Several aspects of the complex behavior of the magnetic domain pattern in ultrathin iron films on Cu(001) are still not understood thoroughly. With his background in applied theory, Alessandro Vindigni brought up new ideas and different approaches, which, hopefully, will lead to a better understanding of the experimental observations. It was a delight to work with him. We had many discussions. At times we had diverging concepts, but that made the discussions all the more interesting. I am very grateful to him for reading several sections of this dissertation with great care, his readiness to help, and the good time we had together in the office.
The magnetic characterization of the samples would not have been possible without the Mott detector. The detector had been provided by Vladimir Petrov and Mikhail Galaktionov from Saint Petersburg. Whenever a power failure at ETH damaged the detectors or the electronics, they would soon be here for an analysis and repair the damaged parts. I highly appreciated their prompt response and the patience with which they explained the inner workings of the electronics to me. Over the years, they improved the Mott detector step by step so that it was a pleasure to work with it.

After he joined our group, Kurt Brunner was a frequent companion in the laboratory. It is through him that I was informed about many things. It was indeed an enjoyable time. I thank him for keeping me company and sharing many ideas and thoughts.

In the initial stages of my thesis work, I shared my time with the fellow Ph.D. students Yves Acremann and Matthias Buess. I am thankful to Yves Acremann for administrating our computer network. With Matthias Buess I enjoyed several extended stays at conferences. It was a time with intense scientific debates and many good memories. In the later stages of my thesis work, Thomas Michlmayr and Niculín Saratz became fellow Ph.D. students. I wish them luck with their projects.

After he became a member of our group, I enjoyed the many entertaining discussions Thomas Bähler stimulated with his unconventional thoughts.

Even though not naming them individually, I would also like to thank numerous people outside our research group, both inside and outside the department of physics. It has been a great pleasure to know them. They helped make the stay at ETH a very pleasant time.

Last but not least, I would like to express my deep gratitude to my parents for supporting me through all the years. I feel very fortunate that I have had the chance to pursue my educational interests.
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