Neutron Grating Interferometry for Imaging Magnetic Structures in Bulk Ferromagnetic Materials

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Summary

Magnetic domains form the link between the basic physical properties of a magnetic material and its macroscopic behavior. It is of great interest to learn more about bulk ferromagnetic materials, since such materials are commonly used in daily life, for example, in the yoke of an electromagnet. Today, there is a variety of domain imaging techniques which are based on light, electrons or X-rays. Due to the limited penetration depth of the used radiation, the investigation of domain structures is restricted to the sample surface and to thin film samples. The investigation of magnetic domains in bulk ferromagnetic materials remains a significant challenge. Neutrons can easily penetrate centimeter thick metallic samples and therefore researchers utilize this advantage to investigate domain structures in bulk materials.

In this thesis, a new method for the study of bulk ferromagnetic domain structures is presented. This technique overcomes some of the drawbacks that challenge other neutron imaging methods, in terms of exposure times and image quality. The technique is based on a neutron grating interferometer. It consists of three diffraction gratings: two absorption gratings made of gadolinium and one phase grating made of silicon. The field of view of the setup is 64 × 64 mm² and the spatial resolution is about 100 µm. The image contrast generated within the grating interferometer is based on scattering of unpolarized neutrons at magnetic domain walls in the sample. The measured scattering image is termed “neutron dark-field image” (nDFI).

The technique was used on the one hand to investigate bulk magnetic domain structures and on the other hand to study bulk magnetization processes. These experiments were performed at the ICON beam line at the spallation neutron source SINQ at the Paul Scherrer Institute (PSI). For the bulk magnetic domain structure imaging experiment, we visualized the internal domain wall structure of a single-crystalline (110)−oriented iron silicide (FeSi) disc. This disc had a thickness of 300 µm and a diameter of 10 mm. The obtained experimental results were compared to surface sensitive magneto-optical Kerr microscopy results, where an almost perfect match was found. The study of bulk magnetization processes was obtained by imaging the domain wall density distribution in the sample. The samples under investigation were a single-crystalline (100)−oriented FeSi disc with a thickness of 500 µm and a diameter of 12 mm, and a poly-crystalline sample (steel plate) with a thickness of 750 µm and a square shape with an edge length of 15 mm. Both samples were magnetized by
applying an external magnetic field.

The imaging results of the magnetization processes for both samples were verified by complementary neutron small-angle scattering (SANS) experiments. They were conducted at the SANS-I instrument at PSI. The SANS experiments were performed under the same conditions of sample magnetization and applied field as for the grating interferometry experiments. The obtained SANS magnetization results verified our grating interferometry results and additionally delivered complementary information about the internal domain wall distribution in the samples.

To compare and understand the results of the imaging experiments of the magnetization processes obtained with the grating interferometer, we conducted finite element method (FEM) simulations. The obtained FEM results agreed well with the nDFI results. For the single-crystalline material, the FEM results show an excellent similarity to the experimental results. For the poly-crystalline material, the FEM results show a qualitative agreement with the nDFI results.
Zusammenfassung


In dieser Arbeit wird eine neue Methode für die Untersuchung von massiven ferromagnetischen Domänenstrukturen präsentiert. Diese Technik überwindet einige der Hindernisse, welche die anderen Neutronenmethoden hinsichtlich Bildqualität und Belichtungszeit herausfordert. Die Technik basiert auf einem Neutronengitterinterferometer, welches aus drei Diffraktionsgittern besteht: zwei Absorptionsgitter, hergestellt aus Gadolinium und einem Phasengitter aus Silizium. Das Gesichtsfeld des Aufbaus ist $64 \times 64$ mm$^2$ und die Ortsauflösung ist in etwa 100 µm. Der Bildkontrast, welcher innerhalb des Gitterinterferometers generiert wird, basiert auf der Streuung von unpolarisierten Neutronen an magnetischen Domänenwänden in der Probe. Das gemessene Streubild wird als "Neutronen-Dunkelfeld Bild" bezeichnet (nDFI).

Diese Technik wurde auf der einen Seite verwendet, um massive magnetische Domänenstrukturen zu untersuchen und auf der anderen Seite, um Magnetisierungsprozesse von massiven Materialien zu studieren. Die Experimente wurden an der ICON Strahllinie an der Spallationsquelle SINQ des Paul Scherrer Institutes (PSI) durchgeführt. Für die Abbildungsexperimente von massiven ferromagnetischen Domänenstrukturen haben wir die interne Domänenwandstruktur einer einkristallinen (110)-orientierten FeSi Scheibe sichtbar gemacht. Diese Scheibe hatte eine Dicke von 300 µm und einen Durchmesser von 10 mm. Die erhaltenen experimentellen Ergebnisse wurden mit oberflächensensitiven magneto-optischen Kerr Mikroskopieergebnissen verglichen, wobei eine nahezu perfekte Übereinstimmung gefunden wurde. Die Untersuchung des Magnetisierungsverhal-
tens von massiven ferromagnetischen Materialien wurde durch das Sichtbarmachen der Domänenwanddichteverteilung in der Probe erzielt. Die untersuchten Proben waren: eine einkristalline (100)-orientierte FeSi Scheibe mit einer Dicke von 500 µm und einem Durchmesser von 12 mm, und eine polykristalline Stahlplatte mit einer Dicke von 750 µm und einer quadratischen Form mit einer Kantenlänge von 15 mm. Beide Proben wurden durch ein angelegtes externes Magnetfeld magnetisiert.


## Contents

1 Introduction ........................................... 1
   1.1 Motivation ........................................... 1
   1.2 Existing observation techniques for magnetic structures .......... 3
   1.3 Outline of the thesis ................................ 10

2 Theory ................................................... 11
   2.1 Wave optical description of neutron interaction with matter ....... 11
      2.1.1 Interaction potentials and refractive index ............. 11
      2.1.2 Phase shift of neutron waves passing through matter ...... 14
   2.2 Refraction of neutrons at domain walls ........................ 16

3 Principles of neutron grating interferometry .................. 23
   3.1 Setup and design parameters of the diffraction gratings .......... 23
   3.2 Coherence requirements .................................. 29
   3.3 Fabrication of the diffraction gratings ........................ 32
   3.4 Data acquisition and data processing .......................... 35
   3.5 Characterization of the diffraction gratings ................... 42

4 Interferometry experiments I: Magnetization processes ....... 45
   4.1 Experimental setup ....................................... 45
   4.2 Contrast origin for ferromagnetic materials ................... 49
   4.3 Single-crystalline material .................................. 51
      4.3.1 Sample characterization ................................ 51
      4.3.2 Magnetization process ............................... 51
   4.4 Poly-crystalline material ................................... 54
      4.4.1 Angular dependence of the magnetization processes ...... 54
   4.5 Comparison with FEM simulations: Results and interpretations ... 59
      4.5.1 Single-crystalline material ............................ 61
      4.5.2 Poly-crystalline material ............................. 63

5 Small-angle neutron scattering (SANS) experiments .......... 71
   5.1 Experimental setup ........................................ 71
   5.2 Magnetization process of a single-crystalline sample ........... 73
# Contents

5.2.1 SANS results .......................... 73  
5.2.2 Comparison of SANS results with neutron imaging results  ... 77  
5.3 Magnetization processes of a poly-crystalline sample ............. 80  
5.3.1 SANS results .......................... 80  
5.3.2 Comparison of SANS results with neutron imaging results  ... 86  

6 Interferometry experiments II: Magnetic domain structures  
6.1 Experimental setup .................................. 91  
6.2 Neutron imaging results ............................ 94  
6.3 Comparison with magneto optical Kerr effect measurements ....... 95  
6.4 Sensitivity of the grating interferometer ................... 98  

7 Conclusion and outlook ................................ 101  
Bibliography ....................................... 103  
List of publications .................................. 113  
Acknowledgements ................................... 115  
Curriculum Vitae .................................... 117
Chapter 1

Introduction

1.1 Motivation

Weiss postulated in 1907 that magnetic samples are subdivided into elementary domains, each having an arbitrary magnetization direction. The first observation of these domains was achieved on the surface by Bitter in 1931, with the help of a powder method [Bit1931]. Today, a range of experimental techniques exists for the observation of surface domains like Kerr microscopy or magnetic force microscopy, and for the investigation of domains in thin film samples, provided they are transparent for electrons such as Lorentz microscopy or for X-rays such as spectro-microscopy [Hub1998, Hop2005]. The investigation of internal domains in macroscopic bulk metallic materials, however, still remains a significant challenge. The exceptional case is the Libovicky method [Lib1972]. However, this method is only applicable for alloys with a composition of Fe 12.8 at% Si\(^1\). Moreover this method is destructive, as the specimen has to be cut to access the internal domains. An overview of well-established domain observation techniques is made in the following section.

Despite the multiplicity of domain observation techniques today, it is interesting to observe that little is known about the internal domain structure of bulk ferromagnetic materials. A large part of our understanding of magnetic microstructures in bulk materials relies on theoretical considerations, like those of phase theory in conjunction with the analysis of magnetization and magnetostriction measurements [Hub1998]. For soft magnetic sheets or plates, such as those used in electrical machines or other inductive devices, there is a chance to image the magnetic microstructure, because these samples have large single-crystalline grains, which extend through the thickness of the sheet. Therefore many essential features of the domain structures in such materials can be studied on single crystals. For sample thicknesses larger than the domain width, materials are classified as bulk materials, even if they are only some microns thick. Neutron based techniques have the advantage that neutrons can penetrate centimeter thick samples. Therefore, these techniques have the ability to look into bulk magnetic

\(^1\)Iron with 12.8 atomic percent of Silicon.
domain structures and are not limited to the surface or thin films. Neutron techniques have their own challenge, either in image quality (compare Fig. 1.1(p)-(r)) or in exposure times. These challenges are responsible for the stagnating progress in method development for the investigation of bulk magnetic domain structures.

Researchers have utilized the spin property of neutrons that interacts directly with the local magnetization in ferromagnetic samples [Hal1941]. This stimulated the development of neutron investigation techniques [Sch1988], which are based on single-crystal interferometry [Sch1980, Nak1992, Rau1974, Rau2000], crystal analyzer-based topography [Sch1978, Bar1993] or on neutron depolarization analysis [Sch1973, Rec1973]. Neutron crystal interferometry and analyzer based neutron topography have their greatest practical limitations in the very small beam divergence (a few mdeg) and the small energy spread (a few meV) what is compatible with crystal optics. This typically leads to very sensitive but inefficient setups, that require exposure times of several hours for a single image. The use of polarizers and analyzers in combination with an imaging approach, on the other hand, is severely limited by the practical requirement that mirror polarizers and analyzers are typically used in grazing incidence geometry. This results in large sample-to-detector distances of typically half a meter or more that compromises the spatial resolution.

In this thesis, a neutron imaging approach that delivers insight into bulk magnetic domain structures is presented. The approach relies on a neutron interferometer setup based on diffraction gratings [GruDA, Pfe2006n]. Using this method we are able to overcome the limitations of the other neutron techniques. The relaxed requirements on spatial and temporal coherence of the grating interferometer setup lead to exposure times that are comparable to other “non-neutron” domain observation techniques. The setup is capable of working with a partially chromatic spectrum and beam sizes of several centimeters in diameter. The field of view of the setup extends up to $64 \times 64 \text{ mm}^2$ and offers spatial resolution of about 100 $\mu$m.

The first experiments with this setup were performed in 2005, when we demonstrated its efficiency by recording quantitative projections and even three-dimensional tomographic reconstructions of the complex refractive index [Pfe2006n]. The neutron grating interferometer method is closely related to the X-ray grating interferometer that was also developed at the Paul Scherrer Institut (PSI) in recent years [Dav2002, Wei2005, Pfe2006a].

The experimental results presented in this thesis are based on so called dark-field images\(^1\) (DFI). The image contrast is based on the fact that the spatial coherence of the neutron wave front can be changed through small-angle scattering of unpolarized neutrons at, for example, magnetic domain walls. The grating interferometer technique

---

\(^1\)In general, dark-field microscopy describes methods, well known in both light and electron microscopy, which exclude the unscattered beam from the image. As a result, the field around the specimen (i.e. where there is no specimen to scatter the beam) is generally dark. Dark-field images, under these conditions, allow one to map the diffracted intensity.
is capable of measuring scattering angles of about $10^{-6}$ rad. The interferometer setup therefore provides complementary and otherwise inaccessible structural information about the specimen at the micrometer and submicrometer length scale.

In this thesis it is shown that this technique can be used to visualize internal bulk magnetic domain structures with unprecedented quality. It is also demonstrated that the method is applicable to a wide variety of specimens and is well suited to investigate materials under the influence of external parameters, such as external magnetic field, to visualize bulk magnetization processes.

1.2 Existing observation techniques for magnetic structures

This section gives an overview of the most commonly used techniques to study domains on surfaces, in thin-films and in bulk materials. Further information can be found in [Hub1998, Fre2001, Eim2002].

The interaction of magnetism with light can be observed by the naked eye only under very special circumstances based on Faraday’s discovery of the magnetic influence on optical polarization. Nonetheless, a very impressive number of tools have been developed over the past 150 years that render magnetic phenomena and structure as images, thus making them “visible” to the naked eye. The imaging techniques may be classified into two groups according to the physical mechanism of interaction between the probe and sample. These are magnetic stray field mapping and magnetization mapping. The following sections focus on: (i) the decoration technique, (ii) the Libovicky technique, (iii) magneto-optical methods, (iv) electron microscopy techniques, (v) local probe techniques, (vi) X-ray techniques and (vii) neutron techniques.

(i) Decoration technique:

In the year 1931, Bitter for the first time observed magnetic domains [Bit1931, Bit1932]. He used a surface sensitive method. For a long time refinements of the Bitter method offered the highest spatial resolution. The surface of a magnetic material is dusted with magnetic nano-particles (Fe$_2$O$_3$), which are extracted from a colloidal suspension. The particles agglomerate at positions of maximal magnetic field gradients, typically at domain walls. The final decoration is imaged under an optical microscope. A Bitter image of a cobalt crystal can be seen in Fig. 1.1(a), showing the first domain image ever taken [Bit1931]. Nowadays, high-resolution Bitter scanning microscopy (HRBSM) achieves a resolution of 80 nm by observing ferrofluids or nm-sized particles with a scanning electron microscope (SEM) [Got1977]. A HRBSM image of a section of a hard disc is shown in Fig. 1.1(b) [Kit1996].
(ii) Libovicky technique:
A unique “freezing in” method that permits detailed investigation of domains inside bulk metallic samples was discovered by Libovicky [Lib1972]. A special silicon-iron alloy (Fe 12.8 at% Si) undergoes an irreversible structural transition at about 600°C, namely the formation of ordered submicroscopic precipitates. These platelets orient themselves along the local magnetization direction by elastic interactions. At room temperature, this “texture” gives rise to a birefringence effect after suitable etching treatment, which can be observed with polarized light. By polishing away the surface successively, deeper layers of the domain structure are revealed. A domain image obtained with the Libovicky method is shown in Fig. 1.1(c). The disadvantage of this method is that it is destructive and only permits a one-time investigation of the domains.

(iii) Magneto-optical methods:
Magneto-optical methods rely on detecting small rotations of the polarization plane of linearly polarized light upon reflection (magneto-optical Kerr effect, MOKE) or transmission (Faraday effect) through a magnetic sample [Hub1998]. Different magnetic configurations can be probed in a modified optical polarization microscope (Kerr microscope) by using various geometries (polar-, longitudinal-, transversal-, gradient effect). MOKE measurements are direct and quantitative measurements of the magnetization $M$ of a sample at the surface. A MOKE image of an iron surface is shown in Fig. 1.1(d). MOKE can also be used to study dynamical effects, as magnetic fields can be applied. A spatial resolution close to the diffraction limit (below 200 nm) can be achieved using laser illumination and high numerical aperture objectives. The method is partially surface sensitive, as light penetrates about 20 nm into most metals.

(iv) Electron microscopy techniques:
Lorentz microscopy is a well-established variant of transmission electron microscopy (TEM). In Lorentz microscopy a high-energy (80 to 400 keV) electron beam is incident on a thin (< 150 nm) magnetic sample. The magnetic contrast is derived from the deflection of the electrons due to the Lorentz force upon their passing-through the magnetic induction field in the sample [Cha1999]. A lateral resolution of better than 50 nm can be achieved, bearing in mind that the measurement represents an average over the sample thickness. Different contrast mechanisms such as “Fresnel mode”, “Focault mode” or differential phase contrast allow either domain walls or domains to be imaged. An image obtained with “Fresnel mode” is shown in Fig. 1.1(e).

Electron holography is another variation based on electron transmission microscopy, that enables quantitative magnetic measurements on the nanometer scale [Ton1980, Ton1999]. In electron holography, an electron beam is split by a biprism into two beams, whose relative phase shift is proportional to the enclosed magnetic flux. In the absolute mode one wave packet passes through vacuum and the other
1.2. Existing observation techniques for magnetic structures

(a) Bitter
(b) HRBSM
(c) Libovicky
(d) MOKE
(e) Lorentz
(f) e’ holography
(g) SEM, Type I
(h) SEM, Type II
(i) SEMPA
(j) MFM
(k) SP-STM
(l) Lang
(m) PEEM
(n) STXM
(o) MTXM
(p) pol. topography
(q) unpol. topo
(r) interferometry

Figure 1.1: Domain images obtained by different imaging techniques. Details about the methods can be found in the text. (a) Bitter method, (b) high resolution bitter method, (c) Libovicky method, (d) magneto-optical Kerr image, (e) Lorentz microscopy, (f) electron holography, (g) scanning electron microscopy (contrast type I), (h) scanning electron microscopy (contrast type II), (i) scanning electron microscopy with polarized analysis, (j) magnetic force microscopy, (k) spin-polarized scanning tunneling microscopy, (l) X-ray topography (Lang’s method), (m) X-ray photoemission electron microscopy, (n) scanning transmission X-ray microscopy, (o) magnetic transmission X-ray microscopy, (p) polarized neutron topography, (q) unpolarized neutron topography, and (r) neutron interferometry.
passes through the magnetic sample (thin film), whereas in the differential mode both waves pass through the sample. The hologram obtained by the interference of the two beams needs to be reconstructed in order to obtain laterally resolved magnetic information. Fig. 1.1(f) shows an image of a triangular cobalt particle.

In **SEM**, i.e., scanning electron microscopy, the sample surface is scanned by a focused beam of electrons whose energy ranges from a few eV up to 60 keV. Energy and direction selective detectors collect the re-emitted electrons, which are either back-scattered from the nuclei of the sample atoms or emitted from atoms which are excited by the electron beam. In magnetic microscopy, the **Type I contrast** utilizes the fact that the low-energy secondary electrons are very sensitive to magnetic stray fields outside the sample. An image of a cobalt single crystal is shown in Fig. 1.1(g) [Joy1969]. In contrast, the back scattered high-energy electrons, which mainly interact with magnetic fields inside the sample, are used in **Type II contrast** microscopy. It enables studying rather “thick” samples, with a resolution of about 1 µm and an information depth of about 10 µm for high energy electrons. An image of an iron single crystal is shown in Fig. 1.1(h) [Fat1974].

**SEMPA**, i.e., scanning electron microscopy with polarization analysis, detects the spin polarization of secondary electrons emitted in a scanning electron microscope [Koi1984]. The spin of the secondary electrons is directly related to the magnetization of the sample and therefore magnetic domains can be studied in a quantitative way. The method is extremely surface sensitive (≈ 2 nm) because of the short secondary electron escape depth. The lateral resolution with SEMPA ranges down to 10 nm. A SEMPA image of an iron thin film is shown in Fig. 1.1(h) [All1994].

In conclusion, electron based methods reach extremely high resolutions, however can not be used in conjunction with external magnetic fields as they disturb the contrast. For bulk domain analysis, these methods can not be used due to the low penetration power of electrons. Thus, transmission images can only be obtained for thin film samples.

**(v) Local probe techniques:**
Magnetic force microscopy, **MFM**, records magnetostatic forces or force gradients between a tiny ferromagnetic tip and a magnetic sample [Mar1987]. This technique offers information about magnetic stray fields of the sample. With this method topographic and magnetic contrast images may be simultaneously recorded, with a spatial resolution of down to 10 nm. A magnetic contrast image of a cobalt sample is shown in Fig. 1.1(j) [Mar1987]. Challenges may arise due to the fact that the tip can disturb the sample’s magnetization, thus hindering quantitative imaging.

The spin-polarized scanning tunneling microscope (**SP-STM**) has the highest resolution of all current domain imaging methods, down to the atomic scale (≈ 5 Å) [Wie1990]. SP-STM achieves magnetic contrast through the spin-dependent tunneling probability between a magnetic sample and the tip, which behaves as a source of spin-
polarized electrons. SP-STM delivers quantitative information on in-plane and out-of-plane magnetization and is by definition surface sensitive. The sample topography is measured simultaneously and can be separated from the magnetization. Fig. 1.1(k) is a SP-STM image of a closure domain pattern of an iron island [Wac2002].

In conclusion, local probe techniques reach the highest spatial resolution. However, they are restricted to very special samples or conditions and suffer from the fact that it is difficult to extract quantitative information.

(vi) X-ray techniques:
The Lang method allows X-ray topograms of magnetic domain patterns to be recorded. A monochromatic, parallel X-ray beam is directed onto the sample, which is oriented such that the Bragg’s condition is fulfilled for some set of lattice planes. A perfect crystal would generate a uniform image. Crystal imperfections disturb the process of Bragg reflection, leading to an image of these defects. Lang’s method is sensitive enough to display isolated dislocations. If such structural contrasts are largely absent, the weak magnetostrictive strains and lattice rotations in magnetic crystals can be detected. An example of a Lang topogram of a nearly perfect silicon-iron sample (thickness ≈ 100 µm) is shown in Fig. 1.1(l) [Mil1976].

X-ray spectroscopy imaging techniques rely on synchrotron radiation sources. Effects analogous to the magneto-optical methods also exist using X-rays. The Faraday effect, as known from the magneto-optical methods, is the phenomenon of the magnetization-dependent absorption of circularly polarized light. The analogous effect for X-rays is called X-ray circular dichroism (XMCD). This technique is based on illuminating a magnetic sample with circularly polarized synchrotron radiation and observing the distribution of electrons emitted from the surface with a photoemission electron microscope (PEEM). An image of an iron whisker is shown in Fig. 1.1(m) [Sch1997]. The most powerful feature of this technique is that magnetic domains can be imaged in an element-specific manner.

The XMCD effect is also used in combination with STXM (scanning transmission X-ray microscope) which utilizes zone plates [Kag1996]. The STXM image of the domain pattern of a nickel deposited layer is shown in Fig. 1.1(n).

The use of a full-field transmission X-ray microscope (TXM) to image magnetic domains is called magnetic transmission X-ray microscope MTXM [Fis1998]. A domain image of a gadolinium/iron layer system is shown in Fig. 1.1(o).

(vii) Neutron techniques:
The wavelength of thermal neutrons is in the same range as that of X-rays. Images of lattice deformations obtained with X-rays can, therefore, be reproduced by neutron topography. However, the contrast in neutron topography differs from that of X-rays, because direct magnetic interactions caused by the spin of the neutron are present, in addition to the nuclear interactions. The spin interaction can be exploited
with polarized neutrons. An image of a silicon-iron sample obtained with neutron toponography using polarized neutrons is presented in Fig. 1.1(p), showing the magnetic domains [Bar1993]. The domain walls become visible using unpolarized neutrons as seen in Fig. 1.1(q) [Bar1993].

The **neutron depolarization** method is based on scanning a “fine” (≈ 2 mm ∅) polarized neutron beam over a magnetic sample [Sch1973]. If the beam is polarized along a certain axis (e.g. $z$-axis), then the beam is not depolarized after the passage through a domain where the magnetic induction $B$ is oriented along $±z$. However, in domains where $B$ is oriented along another direction, the neutron will precess around this direction with the Larmor frequency $\omega_L = \gamma_L |B|$, where $\gamma_L$ is the gyromagnetic ratio of the neutron. The polarization of the transmitted beam, when analyzed along the $z$-axis is therefore reduced [Bar1993].

**Neutron interferometry** utilizes the fact that the magnetic induction of the sample directly affects the velocity and hence the phase of a neutron wave function passing through it. This makes it possible to observe ferromagnetic domains via the local changes in intensity caused by the inhomogeneous distribution of the magnetic induction. Using unpolarized neutrons, a contrast is only expected between magnetic domains if the reference beam is passed through a magnetic field. This is due to the fact that unpolarized neutrons cannot distinguish between up and down magnetization, if no reference direction is defined. An image of a 100 $\mu$m thick iron silicon sample obtained with this technique is shown in Fig. 1.1(r) [Sch1980].

In conclusion, the neutron techniques are in principle capable of looking inside the domain structure of bulk metallic samples. However, until now they have not gained widespread use, because the resolution is relatively low, and the exposure times are very long (up to days per image).

The different methods for the observation of magnetic domains can be classified in accordance with the following criteria: spatial resolution, information depth and exposure time. Fig. 1.2 illustrates several observation techniques with respect to these criteria. Note, that the quoted values numbers are only guidelines that depend on the conditions of the experiment. The figure additionally includes the classification of the new nDFI technique. The nDFI expands the information depth to the centimeter scale, whereas the resolution is in the same range as for the neutron topography method. Furthermore the recording time is about several minutes for a single nDFI, which is comparable to X-ray or MFM methods.
1.2. Existing observation techniques for magnetic structures

Figure 1.2: Comparison of magnetic domain observation techniques [Hub1998]. Based on the experimental conditions, the estimated limits are indicated for (a) spatial resolution, (b) information depth, and (c) recording time. The abbreviations used are: MFM (magnetic force microscopy), MO (magneto-optical), SEM (scanning electron microscope) and TEM (transmission electron microscope). The red boxes indicate the classification of the new grating interferometry method presented in this thesis, which provides neutron dark-field images (nDFI’s). The spatial resolution limit of our new technique is presently about 100 µm, and the recording time is in the minutes range. However, the information depth is substantially larger than for all other methods since neutrons can easily penetrate centimeters thick bulk magnetic samples. Images in courtesy of R. Schäfer, IFW Dresden.
1.3 Outline of the thesis

The subsequent chapters are organized as follows:

The second chapter delivers the theoretical background for the wave optical description of the neutron interaction with matter. A closer look at the interaction potentials (nuclear and magnetic) and the refractive index for neutrons is presented. The phase shift that a neutron wave experiences when passing through matter is derived. The scattering of unpolarized neutrons at magnetic domain walls of ferromagnetic samples is explained and a quantitative expression for the scattering angle after the passage of neutrons through a domain wall is given.

The principles of grating-based neutron interferometry are explained in chapter three. Details about the setup and design parameters of the diffraction gratings are given. The grating fabrication processes, which were developed and carried out at the Laboratory for Micro- and Nanotechnology (LMN) of PSI, are explained. The interferometer setup is used in combination with a phase stepping approach. The data acquisition and processing are described in detailed. The characterization of the produced diffraction gratings in terms of spatially evaluating the visibility within the grating interferometer setup is also reported. The visibility map gives a measure of the quality of the fabricated gratings.

In the fourth chapter the origin of the magnetic contrast within the interferometer is explained. Then the experimental imaging results of bulk magnetization processes are shown. Two samples were investigated: a single-crystalline iron silicide disc and a poly-crystalline steel plate. Both samples were magnetized by a variable external magnetic field. The bulk magnetization dependence on the orientation of the plate is investigated. To understand the observed nDFI results, we compare the experimental results for the single-crystalline disc and the poly-crystalline steel plate with finite element method simulations.

Chapter five reports the results of the small-angle neutron scattering (SANS) experiments. They were conducted to compare and to verify the bulk magnetization process results obtained from the neutron grating interferometry experiments. The SANS measurements for the single-crystalline iron silicide disc and the poly-crystalline steel plate were performed under the same magnetization conditions as for the interferometry experiments. The angular dependence of the magnetization process of the steel plate was also investigated by the SANS measurements.

Chapter six shows the grating interferometry imaging results of internal magnetic domain structures of a single-crystalline iron silicide sample. These neutron results are compared with measured surface sensitive MOKE images. The sensitivity of the grating interferometer setup was determined by the use of a FeSi single-crystalline wedge with a superficial domain structure.
Chapter 2

Theory

The theory part in this thesis is geared mainly towards the understanding of our experimental results. In section 2.1, the wave optical description of the neutron interaction with matter is explained. The derivation of the refractive index for neutrons is presented, taking into account the nuclear and magnetic interaction potentials as reported in subsection 2.1.1. In subsection 2.1.2, the phase shift of a neutron wave passing through matter is discussed. The scattering behavior of neutrons at magnetic domain walls is described in section 2.2. A detailed description of the content of this chapter can be found in the text books: “Neutron interferometry” by Rauch and Werner [Rau2000], “Introductory Nuclear Physics” by Krane [Kra1988], and in the habilitation “Verhalten der Neutronen beim Durchgang durch die Blochwand” by Schärpf [Sch1976].

2.1 Wave optical description of neutron interaction with matter

Mathematical analogies between the propagation of light described by Maxwell’s equations and the propagation of low-energy neutrons as described by the Schrödinger equation suggests that most phenomena in light optics have analogies in neutron optics. In subsection 2.1.1 an analogue between neutrons and light is made via the derivation of the refractive index. The phase shift that a neutron wave experiences through the interaction with the nuclear potential is explained in subsection 2.1.2.

2.1.1 Interaction potentials and refractive index

Matter wave fields $\psi(r, t)$ are described by the Schrödinger equation

\[
H\psi(r, t) = \left( -\frac{\hbar^2}{2m} \nabla^2 + V(r, t) \right) \psi(r, t) = i\hbar \frac{\partial \psi(r, t)}{\partial t},
\]

(2.1)

where $H$ is the hamiltonian, $\hbar$ is the reduced Planck constant, $m$ is the mass of the particle (neutron in our case) and $V$ is the interaction potential. Analogously electro-
magnetic wave fields are described by the vacuum wave equation

$$\nabla^2 \psi(r, t) - \frac{1}{c^2} \frac{\partial^2 \psi(r, t)}{\partial t^2} = 0, \quad (2.2)$$

where \(c\) is the speed of light.

These are linear equations which can be solved in free space using the plane wave approach:

$$\psi_k(r, t) = a_k e^{i(k \cdot r - \omega_k t)} = \psi(r) e^{-i\omega t}, \quad (2.3)$$

where \(a_k\) is the amplitude, \(|k| = \frac{2\pi}{\lambda}\) is the modulus of the wave vector, \(\lambda\) is the wave length of the neutron and \(\omega\) is the angular frequency. This leads, in both cases, for stationary situations to the Helmholtz equation

$$\nabla^2 \psi(r) + k^2 \psi(r) = 0, \quad (2.4)$$

for both kinds of waves (electromagnetic and matter) with the following dispersion relations:

$$k^2_m = \frac{2mE}{\hbar^2} \quad \text{(matter waves)}, \quad (2.5)$$

and

$$k^2_{em} = \frac{E^2}{\hbar^2 c^2} \quad \text{(EM waves)}, \quad (2.6)$$

where \(E\) is the energy. The velocity of wave propagation for electromagnetic waves is always equal to the velocity of light, whereas for matter waves it is determined by the de Broglie relation.

Since refraction, reflection, diffraction and interference are consequences of the stationary equations, the complete array of wave optical phenomena known from the wave nature of light occur for neutron waves in matter. Thus the phenomena are analogous, aside from their differing dispersion relations and interactions. For time-dependent phenomena, differences are expected, because the power of the time derivative in the wave equations is different, first in order in Eqn. 2.1 and second order in Eqn. 2.2.

The derivation of the refractive index for neutrons is quantum mechanical in nature. Neutrons which move inside a medium experience a spatially dependent potential \(V(r)\). In free space, the energy eigenstate \((E = \hbar \omega)\) and the wave function

$$\psi(r, t) = \psi(r) e^{-i\omega t} \quad (2.7)$$

satisfies the time-independent Schrödinger equation

$$\nabla^2 \psi(r) + \frac{2m}{\hbar^2} E \psi(r) = 0. \quad (2.8)$$
2.1. Wave optical description of neutron interaction with matter

Interactions within a medium are introduced with a potential term leading to

\[ \nabla^2 \psi(r) + \frac{2m}{\hbar^2} [E - V(r)] \psi(r) = 0. \]  

(2.9)

Both equations are Helmholtz scalar wave equations (Eqn. 2.4), with the wave vector \( k \) outside the medium

\[ k^2 = \frac{2mE}{\hbar^2}, \]  

(2.10)

and the wave vector \( K \) inside the medium (region of the potential)

\[ K^2(r) = \frac{2m}{\hbar^2} [E - V(r)]. \]  

(2.11)

One defines the spatially dependent refractive index as the ratio of this spatial dependent wave vector \( K(r) \) to the free space wave vector \( k \) [Sea1989], such that:

\[ n(r) \equiv \frac{K(r)}{k} = \sqrt{1 - \frac{V(r)}{E}}. \]  

(2.12)

The nuclear potential \( V_{nuc}(r) \) and the magnetic potential \( V_{mag}(r) \) are considered within this thesis.

Since the range of the neutron-nucleus interaction is much shorter than the wavelength of thermal neutrons, the scattering is isotropic. This allows the Fermi pseudo potential to be used to describe the nuclear interactions of neutrons with a sample containing many nuclei [Fer1936]

\[ V_{nuc}(r) = \frac{2\pi}{m} \sum_j b_j \delta(r - R_j), \]  

(2.13)

where \( R_j \) is the position of the j-th nucleus and \( b \) is the neutron scattering length. The mean interaction potential, or neutron optical potential, for a material is defined as:

\[ \langle V_{nuc}(r) \rangle = \frac{2\pi}{m} b_c N. \]  

(2.14)

Here, \( N \) is the mean number of scattering nuclei per unit volume and \( b_c = \langle b \rangle \) is the mean coherent scattering length. The mean scattering length \( b_c \) ranges from -5 to +10 fm, and in most cases it is positive\(^1\).

In magnetic materials, the neutron interacts with the magnetic induction field \( B \) via its magnetic dipole moment \( \mu_N \). The magnetic interaction potential is given by:

\[ V_{mag}(r) = -\mu_N \cdot B. \]  

(2.15)

The strengths of both potentials are comparable in size for ferromagnetic magnetic materials such as iron, nickel or cobalt.

\(^1\)The reason for positive and negative values is explained in detail in [GruDA].
Any absorption or nuclear reaction effects are described by the imaginary part of
the scattering potential in Eqn. 2.14, leading to a complex scattering length $b_c = b' - ib''$ [Bla1952]. This yields, according to Eqn. 2.12, a complex index of refraction.

This complex index of refraction counting for both interaction potentials may also
be written in the form:

$$ n = \sqrt{1 - \frac{V}{E}} = 1 - \delta_{\text{nuc}} - \delta_{\text{mag}} + i\beta . $$

(2.16)

The real part $\delta_{\text{nuc}}$ counts for the nuclear interaction potential and is given by

$$ \delta_{\text{nuc}} = \frac{\lambda^2 Nb_c}{2\pi} . $$

(2.17)

In the presence of a magnetic induction field $B$ a magnetic contribution needs to be
added to the refractive index [Jus1972, Str2007]:

$$ \delta_{\text{mag}} = \pm \frac{\mu_N \cdot B}{2E_0} = \pm \frac{2\mu_N \cdot B m\lambda^2}{\hbar^2} , $$

(2.18)

where $E_0$ is the kinetic energy of the unperturbed neutron. This term accounts for the
interaction of neutrons with magnetic fields, as the magnetic moment of the neutron $\mu_N$
makes the neutrons sensitive to magnetic fields. When neutrons, with their magnetic
moments, are introduced into a region where a magnetic induction field $B$ is present, a
Zeeman splitting of the potential energy of magnitude $\pm \mu_N \cdot B$ occurs, corresponding
to the two quantized spin states of the neutron. This happens at the expense of the
kinetic energy of the neutron, so that a velocity splitting occurs. The different signs
are related to the different momentum changes due to the Zeeman energies $\pm \mu_N \cdot B$
of the eigenstates $|\uparrow\rangle$ and $|\downarrow\rangle$ of the Pauli spin operator. A more detailed derivation
of the refractive index, especially accounting for the magnetic interaction potential is
made in subsection 2.1.3.

The imaginary part $\beta$ accounts for absorption and incoherent scattering processes
and is given by [Gol1974]

$$ \beta = \frac{\sigma_r N \lambda}{4\pi} , $$

(2.19)

where $\sigma_r$ is the total reaction cross-section per atom including the absorption and the
incoherent scattering effects. This imaginary part is typically small in comparison to
the real parts ($\delta/\beta \approx 10^4$).

### 2.1.2 Phase shift of neutron waves passing through matter

To determine one of the several properties of the phase grating later introduced in this
thesis, one must understand the phase shift that a neutron wave-packet experiences
when passing through matter.
2.1. Wave optical description of neutron interaction with matter

As a neutron passes through matter, it experiences a collection of interactions with sample nuclei. The phase along the path of a neutron is given by the Feynman-Dirac path integral in classical mechanics [Fey1948]. In our case, the potential \( V = V(r) \) depends only on the local environment (Eqn. 2.14) and is independent of time and velocity.

The neutron total energy \( E \) is a constant of motion and it is the change in wavelength in the medium that results in the outgoing phase shift. This phase shift is given by [Rau2000]

\[
\Delta \phi = k(1 - n)D .
\]  
(2.20)

Where \( D \) is the effective path length of the neutron in the material. With Eqn. 2.16 for the refractive index \( n \) this simplifies to

\[
\Delta \phi = -Nb_c \lambda D ,
\]  
(2.21)

where the product of the particle density \( N \) and the coherent scattering length \( b_c \) is called the neutron Scattering Length Density (nSLD).

This phase shift is schematically presented in Fig. 2.1. The upper sine wave in Fig. 2.1 illustrates an unperturbed neutron wave (reference wave, \( n = 1 \)) that does not pass through the medium and has a constant wave function described by

\[
\psi_{\text{incident}} = A \cdot e^{ikx} .
\]  
(2.22)
The lower neutron wave in Fig. 2.2 enters the medium which is described by the refractive index \( n = 1 - \delta + i\beta \). The wave is attenuated within the medium \((A \rightarrow A')\). This effect is described by the imaginary part of the refractive index \(i\beta\). The real part of the refractive index leads to the phase shift of the exiting wave. The wave function after the passage through the object is given by

\[
\psi_{exit} = A \cdot e^{inkx} = e^{-i\kappa\delta x} \cdot \frac{A'}{A} \cdot \psi_{incident}.
\]  

(2.23)

The phase shift for neutrons passing through matter can be either positive or negative, depending on the sign of \(b_c\) and according to Eqn. 2.21. There are only a few isotopes which have a negative coherent scattering length, for example manganese \(^{55}\text{Mn}\)(\(-3.73\) fm), hydrogen \(^1\text{H}\)(\(-3.74\) fm) and titanium \(^{48}\text{Ti}\)(\(-6.08\) fm). An experimental proof is shown in [Pfe2006n].

In reality, specimens always exhibit a combination of amplitude and phase effects, producing changes to both the amplitude and phase relationships between the incident and emerging neutron waves. Information about the phase information, however, is only accessible with interferometry experiments.

### 2.2 Refraction of neutrons at domain walls

A ferromagnetic sample has a spontaneous magnetic moment caused by the molecular field (Weiss field), which leads to a parallel adjustment of adjacent magnetic moments of the atoms [Chi1999]. The region where all magnetic moments point in the same directions is called a magnetic domain. Domains are described by a resulting magnetic induction \(B\), as schematically shown in Fig. 2.2. Within a domain, the magnetization \(M\) points along an easy axis of the crystal and has a material specific constant saturation magnetization value, \(B_s\). The easy axis refers to the energetically favorable directions of the spontaneous magnetization in ferromagnetic cubic materials. For iron, these axes are the \((100)\), \((010)\) and \((001)\) directions of the cubic material. The ferromagnetic sample regarded as a whole has a much smaller magnetic moment compared to the saturation magnetization since the individual domains are distributed randomly in the sample when no external field is applied. In the transition regions between the magnetic domains, namely the domain walls, the magnetization direction rotates out of the easy direction of one domain to the other. For a Bloch domain wall, the magnetization rotates through the plane of the wall. For a Néel wall, the magnetization rotates in the plane of the wall. The domain wall thickness depends on the anisotropy of the material and is in the range of 1 nm to 10 nm for iron [Hub1998].

In this section we will discuss the refraction behavior of unpolarized neutrons at domains walls. As the neutrons are interacting with the magnetic induction \(B = \mu_0 H + M\), we will use for the following the magnetic induction \(B\) instead of the magnetization.
Figure 2.2: *Schematic drawing of a plane domain wall separating two adjacent domains with different magnetic induction directions $B$. The neutrons transverse in series the regions I to IV. Region I: Outside the sample (air), with $B_I = 0$. Region II: Magnetic domain with constant magnetic induction $B_{II}$. Region III: Domain wall with magnetic induction $B_{III}$, where the induction turns from $B_{II}$ to $B_{IV}$. Region IV: Magnetic domain with constant magnetic induction $B_{IV}$. Region V: Outside the sample (air), with $B_V = 0$.*

$M$. $\mu_0$ is the permeability constant. This assumption is justified, firstly, because in zero field configuration $B$ equals $M$. Secondly, because the applied external magnetic field $H$ to fully magnetize for instance iron is of the order of several mT, and therefore negligible. The saturation magnetization of iron is 2.2 T.

Neutrons interact with the atomic nucleus and the magnetic moments of the atomic shells as they have a magnetic moment $\mu_N$. The refraction of neutrons in magnetic materials is only described correctly by dynamical theory [Eks1949, Sch1976, Sch1978a, Sch1978b, Pet2000]. This leads, far away from Laue reflections, to a treatment with the Schrödinger equation with a spin-dependent potential, which depends on the average homogeneous magnetic induction. This gives insight into how and why, with refraction, the intensities of the direct and deflected beams depend on the magnetization directions in adjacent domains. In the following, domain walls are considered as plane parallel “plates”. For the problem of the determination of the refraction angle for ferromagnetic materials, one can regard the Bloch wall as infinitely thin. Thus it is therefore not necessary to investigate the behavior of neutrons inside domain walls. This is similar to the Snell’s law in optics, which describes the refraction of a beam at an interface. In this formula the refraction angle is simply given by the two refractive indices of the adjacent media.
We consider the refraction of unpolarized neutrons at domain walls as a boundary value problem of the Schrödinger equation with a spin dependent potential. The Hamilton operator $H$ is given by:

$$H = -\frac{\hbar^2}{2m} \nabla^2 + V(r) - \mu_N \cdot B(r),$$

(2.24)

where $m$ is the neutron mass, $V(r)$ is the nuclear interaction potential and the magnetic interaction potential is the product of $\mu_N$ and $B(r)$. In total, the neutron experiences a constant interaction potential, if it passes through a magnetic domain. Therefore, one can use the time-independent Schrödinger equation to calculate its behavior in the ferromagnetic system

$$H \psi(r) = E \psi(r).$$

(2.25)

Where $\psi(r) = \left( \begin{array}{c} \psi_1 \\ \psi_2 \end{array} \right)$ is the spinor of the neutron. The total energy of the free neutron is $E = \frac{\hbar^2 k^2}{2m}$. The wave equation of the neutron in a magnetic material is then given by:

$$\left( \nabla^2 + k^2 \right) \psi(r) - \frac{2m}{\hbar^2} \left[ V(r) + \mu_N \cdot \sigma \cdot B(r) \right] \psi(r) = 0.$$

(2.26)

The vector $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ is composed of the $2 \times 2$ Pauli spin matrices which couple all components of the spin to the magnetic induction $B$.

Neutrons experience a potential jump if they enter a region of uniform magnetization $B(r) = B$, depending on the spin states parallel or antiparallel with respect to $B$. The boundary condition is the continuity of the wave function at the entrance point on the sample surface, where the neutron enters into the first domain. Due to the constant nuclear potential inside the crystal, only the magnetic potential jump needs to be calculated when the neutron traverses the domain wall.

As mentioned above, the domain wall is supposed to be “thin”, in the sense that for the passage of the neutron through the wall the direction of the spin is fixed in space. Neutrons are refracted as long as the neutron spin experiences a non-adiabatic passage through the domain walls, i.e. the spin of the neutron can not follow the rotation of the magnetic induction inside the wall. As neutrons pass through a domain wall they experience a rotating field with the frequency $\omega$ [Sch1978b]. The Larmor frequency of the neutron is given by $\omega_L = \frac{2|\mu_N| B_s}{\hbar}$. The ratio between $\omega$ and $\omega_L$ determines the refraction probability. It is reported in literature for Nickel [Pet2000] that for $\omega \approx \omega_L$ and an angle of incidence of 0.3 mrad, a small percentage of the neutron spins can adiabatically follow the rotation of the wall. Therefore, behind a domain wall three different neutron beams can be found: (i) an unrefracted beam which is unpolarized, (ii) and (iii) two spin-polarized beams which are displaced by a certain angle against the initial beam direction.

Similarly, for the nuclear potential we also assume for the magnetic induction $B$ an averaged induction $B_{AV}$, with $B_{AV} = \mu_0 H_{AV} + M_{AV}$ in each domain. In this formalism we are considering the sample (crystal) as a continuum and not as a lattice.
The spinor which solves the Schrödinger equation (Eqn. 2.26) has the form

\[ \psi(r) = c \cdot e^{i k_{p,a} \cdot r}. \] (2.27)

Where \( k_p \) is the wave vector for the neutron with the spin parallel to \( B_{AV} \)

\[ k_p^2 = k^2 - \Delta k^2 - k_{B_{AV}}^2, \] (2.28)

and \( k_a \) is the wave vector for the neutron with the spin antiparallel to \( B_{AV} \)

\[ k_a^2 = k^2 - \Delta k^2 + k_{B_{AV}}^2. \] (2.29)

Here \( \Delta k^2 = 2 \pi \cdot N \cdot b_c \) accounts for the nuclear interaction and \( k_{B_{AV}}^2 = 2m/\hbar^2 \cdot \mu \cdot |B_{AV}| \) accounts for the magnetic interaction.

In Eqn. 2.12, the index of refraction is derived from the time-independent Schrödinger equation as the ratio of the spatially dependent wave vectors. The index of refraction for magnetic materials has an additional spin dependent potential contribution,

\[ n^2 = \frac{k_{p,a}^2}{k^2} = \frac{k^2 - \Delta k^2 \pm k_{B_{AV}}^2}{k^2} = 1 - \frac{\Delta k^2 \pm k_{B_{AV}}^2}{k^2}. \] (2.30)

Ferromagnetic samples act as birefringent materials for unpolarized neutrons. Magnetic domains inside the samples are associated with different refractive indices. Neutrons which transverse a ferromagnetic sample, as shown in Fig. 2.3, primarily experience a refraction at the sample surface by passing from air \( B_I \) into the sample \( B_{II} \). A further refraction occurs at the domain wall. Inside the sample, the nuclear potential is constant and may be neglected. Thus the refractive index for a pure magnetic potential jump is given by

\[ n^2 = \frac{k^2 \pm k_{B_{AV}}^2}{k^2 \mp k_{B_{AV}}^2}. \] (2.31)

The refraction behavior of an unpolarized neutron beam at the sample surface and at the domain wall is schematically shown in Fig. 2.3. \( \theta \) is the angle of incidence of the neutrons. The wave vector of the incident neutrons is \( k = (k_x, k_y, k_z) \). The normal vector of the sample surface points into the z-direction and the normal vector of the domain wall into the y-direction respectively. The spin of the incident neutrons is either parallel (denoted by “p”) or antiparallel (denoted by “a”) with respect to the z-direction. Neglecting the homogenous nuclear potential (\( k_{p,a}^2 = k^2 - \Delta k^2 \pm k_{B_{AV}}^2 \approx k^2 \pm k_{B_{AV}}^2 \)), the normal component of the wave vector \( k_z \) at the sample surface makes a jump to \( k_{z(p,a)} = \sqrt{k_z^2 \pm k_{B_{AV}}^2} \). The resulting refraction angles at the sample surface for spin parallel with respect to \( B_{II} \) are given by

\[ \sin \theta_p = \frac{k_y}{\sqrt{k^2 - k_{B_{AV}}^2}}, \] (2.32)
Figure 2.3: Refraction behavior of an unpolarized neutron beam entering a ferromagnetic sample. A domain wall separates two adjacent domains with opposite magnetic induction (compare Fig. 2.2). $\theta$ is the angle of incidence of the neutrons. $k$ is the wave vector of the incident neutrons. The beam is firstly refracted at the sample surface and secondly at the domain wall. At the sample surface the normal component of the wave vector $k_z$ makes a jump to $k_z(p,a) = \sqrt{k_z^2 \pm k_{BAV}^2}$. $\theta_p$ and $\theta_a$ are the refraction angle for spin parallel and antiparallel with respect to the magnetic induction respectively. At the domain wall the normal component $k_y$ changes to $k_y(p,a,p) = \sqrt{k_y^2 \pm 2k_{BAV}^2}$ depending on the spin directions. $\alpha_p$ and $\alpha_a$ are the refraction angles after the passage through the domain wall. The total angle of refraction is given by $\gamma_+ \text{ and } \gamma_-$. The homogeneous nuclear potential inside the material was neglected.

and for spin antiparallel given by

$$\sin \theta_a = \frac{k_y}{\sqrt{k^2 + k_{BAV}^2}}. \quad (2.33)$$

The domain wall is the interface between the two magnetic domains. Depending on the spin configuration of the neutron with respect to the induction direction before and after the domain wall ($B_{II}$ and $B_{IV}$), a potential jump of $\pm 2\mu_N B_S$ occurs, where $B_S = |B_{AV}|$. For the domain wall as a refractive surface, the normal component of
$k_y$ changes from the expression $-k_{BAV}^2$ into the expression $+k_{BAV}^2$. This results in a total change of $2k_{BAV}^2$. The normal component $k_y$, therefore changes to $k_{y(pa, ap)} = \sqrt{k_y^2 \pm 2k_{BAV}^2}$, depending on the spin directions. The other components of the wave vector are unperturbed and remain unchanged. The resulting refraction angles after the passage through the domain wall for spin parallel is given by

$$\sin \alpha_{pa} = \frac{\sqrt{k_y^2 + 2k_{BAV}^2}}{\sqrt{k^2 + k_{BAV}^2}},$$

(2.34)

and for spin antiparallel given by

$$\sin \alpha_{ap} = \frac{\sqrt{k_y^2 - 2k_{BAV}^2}}{\sqrt{k^2 - k_{BAV}^2}}.$$  

(2.35)

The total refraction angles $\gamma_{+,-}$ as shown in Fig. 2.3 behind the wall are given by

$$\gamma_+ = \theta - \alpha_{ap} = \theta - \arcsin \left( \frac{\sqrt{k_y^2 + 2k_{BAV}^2}}{\sqrt{k^2 + k_{BAV}^2}} \right),$$

(2.36)

and

$$\gamma_- = \alpha_{pa} - \theta = \arcsin \left( \frac{\sqrt{k_y^2 - 2k_{BAV}^2}}{\sqrt{k^2 - k_{BAV}^2}} \right) - \theta.$$  

(2.37)

It is seen from Eqn. 2.36 and Eqn. 2.37 that the refraction angles are the same, independent of how the components of $k$ are distributed on $k_x$ and $k_z$. The refraction angle only depends on $k_y$ and $k$ and the component of the spin in z-direction.

The refraction angles are of the order of mrad, which so far can only be detected in a high resolution double crystal diffractometer experiment [Jus1972, Pet2000, Str2007]. With the neutron grating interferometer we are also able to resolve these refraction angles, as shown in the following.
Chapter 3

Principles of neutron grating interferometry

In this chapter, the principles of a neutron grating interferometer, consisting of three diffraction gratings, are explained. In combination with a phase stepping approach, it can be used to simultaneously obtain transmission images (TI), differential phase contrast images (DPCI) and dark-field images (DFI). In section 3.1 the experimental setup is shown. Particular attention is given to the calculation of the setup parameters. These are the inter-grating distances and grating parameters, such as period and grating heights. The coherence requirements are discussed in section 3.2. Section 3.3 describes the fabrication processes for each of the diffraction gratings. The data acquisition and the data processing used to obtain the different images types (TI, DPCI, DFI) are explained in section 3.4. The characterization of the fabricated gratings within the neutron setup is made by locally evaluating the visibility, as in section 3.5.

3.1 Setup and design parameters of the diffraction gratings

The interferometer setup consists of three gratings, as illustrated in Fig. 3.1 [Pfe2006n, Gru2008a]. These are the source grating \( G_0 \), which consists of absorbing lines, a beam splitter or phase grating \( G_1 \), and an analyzer absorption grating \( G_2 \), as shown in Fig. 3.1(a). The field of view provided by the gratings is \( 64 \times 64 \) mm\(^2\). The setup additionally includes a wavelength selector and an imaging detector system.

The source grating \( G_0 \) allows for the use of an incoherent neutron source of a diameter \( c \), since it transforms the source to an array of individual line sources of width \( s \), as shown in Fig. 3.1(b). Each line provides enough spatial coherence for the interferometric contrast. The images created by each line source are superimposed congruently in the detector plane leading to a gain in measured intensity. The moderate temporal coherence requirements of the grating interferometer can be satisfied by using a velocity selector, which delivers a bandwidth of \( \Delta \lambda / \lambda \approx 15\% \) around the design
Figure 3.1: (a) Layout of the grating interferometer setup with its main components, the source grating $G_0$, the phase grating $G_1$, and the analyzer grating $G_2$. (b) Top view of the layout denoting the geometrical setup parameters like inter grating distances ($l$, $d$), the periods of the diffraction gratings ($p_0$, $p_1$, $p_2$), the slit width $s$ of the source grating, and the size of the neutron source $c$.

wavelength. More details about the temporal and spatial coherence requirements are discussed in the following section.

The phase grating $G_1$ is placed at the distance $l$ from $G_0$ and divides the incoming neutron beams mainly into the $\pm 1$ diffraction orders. The corresponding measurements are presented in [Gru2007]. In other words, the phase grating acts as a phase mask, and imprints periodic phase modulations onto the incoming wave field. Through the Talbot effect\(^1\), the phase modulations are transformed into an intensity modulation in the plane of $G_2$, forming a linear periodic fringe pattern perpendicular to the optical axis and parallel to the lines of $G_1$.

\(^1\)The Talbot effect [Tal1836] was first observed by the English scientist Henry Fox Talbot (1800-1877). The phenomenon of Talbot self-imaging is caused by Fresnel diffraction of a grating in the near field. The basic equation used to describe the Talbot effect is the Fresnel diffraction integral ([Bor1980], chapter 8).
The analyzer grating $G_2$, with absorbing lines and the same periodicity and orientation as the fringes created by $G_1$, is placed at distance $d$, directly followed by the imaging detector. In order to make a direct determination of the exact fringe position, micrometer spatial resolution in the detector plane would be needed. However, this is not available for neutron imaging [Gru2006]. Instead, the analyzer grating $G_2$ transforms the fringe positions into measurable intensity modulations on the detector.

For the calculation of the grating and setup parameters as denoted in Fig. 3.1(b), some constraints exist, either from the geometry and wavelength spectrum of the beam line [Kue2005], or by the feasibility of the grating fabrication processes. The constraints given by the beam line are the design wavelength $\lambda = 4.1 \, \text{Å}$ and the distance $l = 5.23 \, \text{m}$ between $G_0$ and $G_1$ as seen in Fig. 3.2. A detailed description about the wavelength selection can be found in [GruDA]. The constraint from the grating fabrication process is the period $p_2$ of the analyzer grating $G_2$; it was set to $p_2 = 4 \, \mu\text{m}$. The other parameters, like the distance $d$, the period $p_0$ of the source grating, the period $p_1$ of the phase grating and the grating heights are calculated to conform with $\lambda$ and $l$.

The contrast of the interference pattern created behind the phase grating $G_1$ changes periodically as a function of the distance $d$ along the optical axis. For an incoming plane wave, a pure phase modulating grating with a period $p_1$, a duty cycle$^1$ of 0.5, introducing a phase-shift of $\pi$, the contrast is strongest when

$$d_n = n \cdot \frac{p_1^2}{8\lambda}, \quad (3.1)$$

for odd $n$ and vanishes for even $n$. Therefore, $d_n$ is known as the fractional Talbot distance [Arr2000].

To observe the interference pattern generated by a spherical wave with radius $l$ behind the beam splitter grating $G_1$, the connection between the Talbot distance $d_n$ for a plane wave and the Talbot distance $d_{sph,n}$ for a spherical wave is given by

$$\frac{1}{d_n} = \frac{1}{l} + \frac{1}{d_{sph,n}}. \quad (3.2)$$

$d_{sph,n}$ originating form a source at distance $l$ is thus given by

$$d_{sph,n} = \frac{ld_n}{l - d_n} = \frac{l \cdot n \cdot \frac{p_1^2}{8\lambda}}{l - n \cdot \frac{p_1^2}{8\lambda}}. \quad (3.3)$$

Notice, for $l \gg d_n$, the difference between $d_n$ and $d_{n,sph}$ is very small.

The triangle intercept theorem holds for the transverse scaling of the diffraction pattern. The image is magnified by a factor of $M$ (Fig. 3.1(b)):

$$M = \frac{l + d_{sph,n}}{l} \quad (3.3) \quad \frac{l}{l - d_n} = \frac{d_{sph,n}}{d_n}. \quad (3.4)$$

$^1$The duty cycle (DC) is defined by the ratio of the trench width to the period of a grating.
Figure 3.2: Layout of the ICON beam line at PSI showing the distance $l$. It is measured from the shutter, where the source grating $G_0$ is installed, to the first experiment station as 5.23 m.
3.1. Setup and design parameters of the diffraction gratings

The spacing of neighboring source lines of the source grating is chosen such that the shift of the interference patterns created by two neighboring virtual line sources along the direction perpendicular to the grating lines exactly matches the periodicity of $G_2$. This condition is fulfilled for

$$p_0 = p_2 \frac{l}{d_{sph,n}}.$$  \hspace{1cm} (3.5)

From Eqn. 3.5 we can express the distance $l$ as

$$l = \frac{p_0}{p_2} \cdot d_{sph,n}.$$  \hspace{1cm} (3.6)

Inserting Eqn. 3.3 for $d_{sph}$, $l$ is given by

$$l = p_0 \frac{ld_n}{p_2 (l - d_n)} \Rightarrow l = d_n \cdot \left(\frac{p_0}{p_2} + 1\right).$$  \hspace{1cm} (3.7)

The magnification of the image can also be expressed in terms of the period $p_2$ of the analyzer grating and the period $p_1$ of the beam splitter grating as

$$M = \frac{2p_2}{p_1} = \frac{d_{sph,n}}{d_n}.$$  \hspace{1cm} (3.8)

Equating Eqn. 3.7 and Eqn. 3.6 leads to

$$\frac{d_{sph,n}}{d_n} = \frac{p_0 + p_2}{p_0},$$  \hspace{1cm} (3.9)

and together with Eqn. 3.8, the period of the phase grating is

$$p_1 = \frac{2p_0p_2}{p_0 + p_2}.$$  \hspace{1cm} (3.10)

The first fractional Talbot distance ($n = 1$) behind the phase grating is then given by

$$d_1 = \frac{p_1^2}{8\lambda} = \left(\frac{p_0p_2}{p_0 + p_2}\right)^2 \cdot \frac{1}{2\lambda}.$$  \hspace{1cm} (3.11)

This can be used in Eqn. 3.7 to obtain the distance $l$ as a function of the two periods $p_0$ and $p_2$:

$$l = \frac{1}{2\lambda} \cdot \left(\frac{p_0p_2}{p_0 + p_2}\right)^2 \cdot \left(1 + \frac{p_0}{p_2}\right) = \frac{1}{2\lambda} \cdot \frac{p_0^2p_2}{p_0 + p_2}.$$  \hspace{1cm} (3.12)

The period $p_0$ is obtained by solving Eqn. 3.12:

$$p_0 = \frac{\lambda l}{p_2} + \sqrt{\left(\frac{\lambda l}{p_2}\right)^2 + 2\lambda l}.$$  \hspace{1cm} (3.13)
With the given parameters $p_2$, $l$ and $\lambda$, it is possible to calculate $p_0$ from Eqn. 3.13. With the value of $p_0$ and Eqn. 3.10 the period $p_1$ can be calculated. With the value of $p_1$, the first fractional Talbot distance $d_1$ can then be calculated using Eqn. 3.11, taking into account the curved wave front. Inserting the given parameters in the equations yields:

\[
\begin{align*}
  p_0 &= 1076 \mu m, \\
  p_1 &= 7.97 \mu m, \\
  d &= 19.4 \text{ mm}.
\end{align*}
\]

The two absorption gratings $G_0$ and $G_2$ should be as opaque as possible for neutrons to guarantee optimum performance of the interferometer. Gadolinium (Gd) is the best absorbing material for neutrons. For $\lambda = 4.1$ Å the neutron (1/e) length for Gd is 3 \mu m [NIST]. For our setup, the source grating $G_0$ is composed of two identical wafers each with a Gd thickness of 10 \mu m. The two wafers are stacked with the Gd layers facing each other, resulting in a total Gd thickness of 20 \mu m for $G_0$, giving an absorption of 99.9 %. Using two wafers instead of one single wafer has the additional advantage that one can adjust the slit width $s$ (see Fig. 3.1(b)) of the source grating, and therefore manipulate the spatial coherence length produced by each individual line source.

For $G_2$ we were able to produce Gd lines of a thickness of 11 \mu m, which provides sufficient absorption of 97 %.

For the phase grating we choose silicon (Si), as it is a well known material for microstructuring processes. The neutron (1/e) length for Si is 0.507 m [NIST]. Therefore, the Si grating lines have negligible absorption ($10^{-5}$) and $G_1$ can be considered as a purely phase modulating grating. The height $h_1$ of the phase grating needs to be such that the neutrons traversing the grating bars undergo a phase shift of $\pi$. Accordingly, $h_1$ is given by Eqn. 2.21

\[
h_1 = \frac{\pi}{N \cdot b_c \cdot \lambda} = \frac{\pi}{n_{SLD} \cdot \lambda}, \quad (3.14)
\]

where $N$ is the particle density and $b_c$ the coherent scattering length. The product of both is known as the neutron scattering length density ($n_{SLD}$) [NIST].

Inserting the corresponding values for Si into Eqn. 3.1 gives the structure height of $h_1 = 37 \mu m$ at 4.1 Å.

An overview of all grating parameters concerning material, calculated periods, calculated heights and required duty cycles is given in Tab. 3.1.

The coherence requirements (temporal coherence and spatial coherence) which are necessary for performing interferometry experiments are discussed in the following section.
3.2. Coherence requirements

Table 3.1: Fabrication parameters of the diffraction gratings.

<table>
<thead>
<tr>
<th>Source grating $G_0$</th>
<th>Phase grating $G_1$</th>
<th>Analyzer grating $G_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Material</td>
<td>Gd</td>
<td>Si</td>
</tr>
<tr>
<td>Period [µm]</td>
<td>1076.00</td>
<td>7.97</td>
</tr>
<tr>
<td>Height [µm]</td>
<td>$2 \times 10.00$</td>
<td>37.00</td>
</tr>
<tr>
<td>Duty Cycle</td>
<td>0.4</td>
<td>0.5</td>
</tr>
</tbody>
</table>

3.2 Coherence requirements

Temporal coherence is related to the monochromaticity of the radiation [Bor1980]. For radiation of a bandwidth of $\Delta \lambda / \lambda$ around a central wavelength $\lambda$, the longitudinal coherence length is $\lambda^2 / \Delta \lambda$. Let us consider two beams originating from the same source point that are superimposed after taking different paths through an optical setup. These beams only have a well-defined phase relation if the difference in optical path lengths is shorter than the longitudinal coherence length. For visible laser light, $\lambda^2 / \Delta \lambda$ can extend over many meters, but it is only of the order of nanometers for neutron beams. Neither the period, nor the lateral position of the interference pattern in our grating interferometer depend on the wavelength of the radiation used. However, three aspects of the interferometer setup are dependent on the neutron wavelength. Firstly, the phase shift of the neutron wave passing through the grating lines of $G_1$ depends on the neutron energy due to the dispersion in the grating material (Eqn. 2.21). The condition to induce a phase shift equal to odd multiples of $\pi$ is not strict and radiation with a certain energy bandwidth of $\Delta \lambda / \lambda \simeq 15\%$ around the design energy can be accepted. A deviation from the phase shift of $\pi$ only affects the contrast and does not change the interference pattern qualitatively. Secondly, the distance $d$ (Eqn. 3.1) is wavelength dependent. Taking into account a certain wavelength distribution, the field distribution behind the phase grating will smear-out and reduce the contrast. Thirdly, the absorption cross-section is velocity dependent [GruDA] and therefore wavelength dependent. The smaller the wavelength, the thicker the absorption materials of the source grating and the analyzer grating need to be.

The requirements on the spatial coherence are now discussed in some more details [Vee2004, Wei2006]. For this consideration we assume that the interferometer is only consisting of the phase grating and analyzer grating and a single source spot as shown in Fig. 3.3. The source spot is a chaotic source with a Gaussian intensity profile. Let $s$ denote the full width at half maximum value (FWHM) of the source intensity profile taken along the direction perpendicular to the lines of the interferometer gratings. For simplicity, it is assumed that the detector is located just behind the analyzer, so the
Chapter 3. Principles of neutron grating interferometry

Figure 3.3: Schematic setup of a two-grating interferometer setup with a single source spot of finite size \( s \). The phase grating \( G_1 \), with period \( p_1 \), is placed at a distance \( l \) from the source. At a distance \( d \) downstream of \( G_1 \), each point in the source creates a set of linear fringes of period \( p_2 \). These fringes are washed out by the non-zero source size, i.e., by the imperfect spatial coherence of the beam illuminating \( G_1 \). The resulting fringe intensity profile is the point-source profile convoluted with the projected source profile of width \( w = sd/l \). The analyzer grating \( G_2 \), with period \( p_2 \), is not shown for simplicity.

Analyzer position coincides with the detector plane. We also restrict our considerations to the case of a quasi-plane wave, so that the scaling factors for a spherical wave (Eqs. 3.3, 3.4) can be neglected. Neglecting higher diffraction orders, the fringe pattern in front of the analyzer grating can be approximated as a sinusoidal intensity profile.

If the inter-grating distance \( d \) is chosen to be one of the Talbot distances at which the contrast takes its maximum, the minima of the intensity profile are zero, considering that the source is infinitesimally small. The fringe profile for completely coherent illumination can then be expressed as

\[
I(x) = I_0(1 + \sin(2\pi x/p_2))
\]

where \( x \) is the transverse coordinate perpendicular to the grating lines, \( I_0 \) the intensity incident on the phase grating, and \( p_2 \) the period of the fringes.

In the case of a source of finite size \( s \), i.e., with only partially coherent illumination, the intensity profile observed is a convolution of the profile from a point source with the projected source profile. With the assumption of a Gaussian-shaped source, this means a convolution with a Gaussian of width

\[
w = \frac{s \cdot d}{l},
\]

where both \( s \) and \( w \) representing the FWHM of the Gaussians. \( w \) is now the projected source size. As a consequence, the visibility \( V \) of the fringes, which is unity in the case
of a point source, is reduced. The visibility is defined as $V = \frac{(I_{\text{max}} - I_{\text{min}})}{(I_{\text{max}} + I_{\text{min}})}$, where $I_{\text{max}}$ and $I_{\text{min}}$ are the maximum and, respectively, minimum values of the intensity of the sinusoidal fringe profile.

The decrease of visibility with increasing projected source size $w$ follows the relation

$$V = e^{-(1.887 \cdot w/p)^2}.$$  \hspace{1cm} (3.17)

This equation follows from an analytical or numerical investigation of the properties of the convolution of a sine with a Gaussian profile [Wei2006]. According to Eqn. 3.17, the visibility decrease as a function of increasing source size and has a Gaussian profile. Thus, for a given required minimum visibility $V_0$, it follows directly from Eqn. 3.17 that the width $w$ of the projected source profile should be

$$w \leq \frac{p}{1.887} \sqrt{\ln V_0}.$$  \hspace{1cm} (3.18)

The projected source size $w$, introduced in Eqn. 3.16, can be expressed in terms of the Talbot order $n$ by substituting Eqn. 3.1 in Eqn. 3.16:

$$w = s \cdot \frac{n p^2}{2 \lambda}.$$  \hspace{1cm} (3.19)

The spatial coherence length $\xi$ is defined as

$$\xi = \frac{\lambda l}{s}.$$  \hspace{1cm} (3.20)

Substituting this for $s$ into Eqn. 3.19 yields to

$$w = \frac{n p^2}{2 \xi}.$$  \hspace{1cm} (3.21)

The visibility as a function of coherence length $\xi$ is then obtained by substituting Eqn. 3.21 into Eqn. 3.17

$$V = e^{-(0.94 \cdot n p^2 \xi)^2}.$$  \hspace{1cm} (3.22)

The coherence length required to obtain a given visibility $V_0$ is finally

$$\xi \geq \left[0.94 \cdot n p^2 (\ln V_0)\right]^{-\frac{1}{2}}.$$  \hspace{1cm} (3.23)

For the setup we choose $V_0 = 0.5$ and $n = 1$. This yields according to Eqn. 3.23 to a spatial coherence length of the order of $\xi \geq 4 \mu m$. Thus, the size of an individual line source $s$ emitting at $\lambda = 4.1$ Å and placed at a distance $l = 5.23$ m, should be smaller than $500 \mu m$, according to Eqn. 3.20. A duty cycle of 0.4 in the source grating $G_0$ fulfills these demands.

Moreover, $G_0$ creates an array of intrinsically coherent, but mutually incoherent, line sources. As there are no limitations in terms of coherence in the direction along the grating lines, a line source can be used. Additionally, the source grating decouples spatial coherence from spatial resolution. The latter is defined by the diameter of the source $c$.

In the following section the fabrication processes for each grating are described.
3.3 Fabrication of the diffraction gratings

As seen from Tab. 3.1, the requirements for each grating concerning the aspect ratio (ratio between structure height and structure width), material and duty cycle are quite different. Both, the source grating $G_0$ and the analyzer grating $G_2$ need to be fabricated from Gd with a sufficient height to absorb the neutrons. The phase grating however needs to be transparent for neutrons. Since the periods of the absorption gratings of $G_0$ and $G_2$ differ by a factor of about 250, two individual fabrication processes have been developed [Gru2008a].

The source grating $G_0$ is an array of absorbing lines with a period of $1076 \, \mu m$, a Gd structure height of $20 \, \mu m$ ($10 \, \mu m$ per wafer $\times$ 2) and a duty cycle of about 0.4. The source width $s$ for each line source is $430 \, \mu m$, which matches the coherence requirements of the calculated value in the last section for $s \leq 500 \, \mu m$. Each grating is fabricated on a 100 mm diameter quartz wafer with a thickness of $500 \, \mu m$. The individual fabrication steps are illustrated in Fig. 3.4(a)-(c). A 20 nm sputtered Cr layer serves as adhesive coating between the quartz substrate and the 10 $\mu m$ thick sputtered Gd absorption layer. The pattern transfer of the grating structure on the Gd layer is achieved by standard photolithography, using a positive photoresist S1813 with a spin coated layer thickness of 1.3 $\mu m$. After a hard bake, the photoresist pattern serves as the etching mask for the Gd layer. The uncovered Gd is etched away in an aqueous solution of sulphuric acid (96 % concentrated) and water with a mass ratio

![Fabrication steps](image)

Figure 3.4: Fabrication of the source grating $G_0$. (a)-(c) Fabrication steps. (d) A photograph of a section of the processed wafer showing the Gd lines (black). (e) Profilometer scan over two grating periods revealing a structure height of 11.5 $\mu m$ consisting of 1.3 $\mu m$ resist layer and the 10.2 $\mu m$ Gd absorption layer with a duty cycle of 0.4.
3.3. Fabrication of the diffraction gratings

Fabrication of the diffraction gratings

(a) Photo lithography
(b) CHF$_3$ + O$_2$ RIE
(c) KOH wet etching

(d)

Figure 3.5: Fabrication of the phase grating $G_1$. (a)-(c) Fabrication steps. (d) Cross section scanning electron microscopy image of the processed grating. The duty cycle of 0.5 is properly achieved for the grating depth of 37 $\mu$m.

of 1:100. The resulting source grating is shown in Figs. 3.4(d) and (e), illustrating a section of a photograph of the processed wafer and a profilometer scan over two grating periods, respectively. The achieved height of about 11.5 $\mu$m, as measured with the profilometer, is composed of the 1.3 $\mu$m resist layer and the sputtered 10.2 $\mu$m Gd absorption layer. The measured DC of the source grating is close to 0.4.

The phase grating $G_1$, with a period of 7.97 $\mu$m, is a Si grating of 37 $\mu$m height and a duty cycle of 0.5, and is fabricated from a 100 mm diameter $<110>$ orientated silicon wafer [Dav2007]. The wafer is 300 $\mu$m thick and polished on both sides. The wafer surface was thermally oxidized to give a 80 nm thin silicon dioxide (SiO$_2$) layer, which later serves as an etching mask for the silicon. The wafer size allows for a square grating of size 64 $\times$ 64 mm$^2$. The fabrication processes are depicted in Fig. 3.5(a)-(c). The grating line pattern is transferred via optical photolithography into the spin coated 500 nm thick positive photoresist layer (S1805). The resist pattern is used as an etching mask to transfer the grid pattern into the SiO$_2$ layer. A dry reactive ion etching (RIE) process containing CHF$_3$ and O$_2$ as etching gases was used. The remaining photoresist was stripped by an oxygen plasma etching step. The phase grating is obtained by etching the uncovered Si in an anisotropic wet etching process in a 20% aqueous potassium hydroxide (KOH) solution. The remaining SiO$_2$ masking layer was removed in buffered oxide etch (BOE) to achieve the final Si grating. A scanning electron
Figure 3.6: Fabrication of the absorption grating $G_2$ by an inclined side wall evaporation process. (a) Schematic view. (b) Cross section of a scanning electron microscope image showing the 11 µm high Gd structures with a duty cycle of 0.25.

Due to the fact that the etch rate of $1.68 \, \mu m/min$ at $76^\circ$ C along the $<110>$ direction is 80 times faster than along the $<111>$ direction, nearly perpendicular side walls of the grating structures are achieved. As seen from the SEM image, the duty cycle of 0.5 and the structure height of 37 $\mu m$ are achieved with high precision.

The manufacturing of the analyzer grating $G_2$ was the most challenging microfabrication task. It has the smallest period of all three gratings and needs to consist of narrow Gd lines with sufficient height to absorb neutrons. Additionally, a large homogeneous grating area of $64 \times 64 \, mm^2$ needs to be fabricated to match the phase grating. The process developed for the analyzer grating includes an inclined side wall evaporation process of Gd on a Si grating, as depicted in Fig. 3.6(a). Therefore, a Si grating, with a period $p_2 = 4 \, \mu m$ and a Si bar width $k$, which needs to be as small as possible, was fabricated on a 100 mm wafer, with the same process steps as for the phase grating. The fabrication limit for the smallest bar width $k$ was found to be 700 nm for a good grid line homogeneity over the whole area. The produced Si grating serves as a basis for the inclined side wall evaporation process. The optimum evaporation angle, $\alpha = 16^\circ$, was found after test runs. The evaporated Gd height $h$ directly affects the thickness $t$ of the Gd at the side walls of the Si grating and, therefore, the duty cycle of the absorption grating. From the geometric relations shown in Fig. 3.6(a), it follows that the duty cycle (DC) is given by

$$DC = \frac{p_2 - k - (h \cdot \sin \alpha)}{p_2}.$$  \hspace{1cm} (3.24)

For an evaporation height $h = 3.7 \, \mu m$, we obtain a DC of 0.35. With the additional Gd layer on the side walls, the complete Gd structure height is $h_2 = 11 \, \mu m$. 
Photographs of the complete diffraction grating set for the neutron interferometer are shown in Fig. 3.7.

The performance of the grating set is experimentally tested by measuring the achieved contrast within the neutron setup in terms of locally evaluating the visibility over the grating area. This is reported in the last section of this chapter. First, the data acquisition, including a phase stepping approach and the data processing to simultaneously obtain the different contrast types, is reported in the following section.

### 3.4 Data acquisition and data processing

The interference pattern created behind $G_1$ with a period of 4 $\mu$m, as already mentioned above, is too small to be individually resolved by the imaging detector. In order to bypass this problem, we use the analyzer grating $G_2$, which is placed in the detection plane. Using a spatial phase stepping approach [Cha1995], the fringe position is transformed into intensity modulations on the detector. The raw data acquisition and the subsequent raw data processing, which simultaneously produce the TI, DPCI and DFI contrast signals, are explained in the following.

The experiment to characterize the gratings was performed at the ICON beam line at the spallation neutron source SINQ at the Paul Scherrer Institut [Kue2005]. The neutron flux density (without velocity selector) at the beam line is $5.8 \times 10^6$ cm$^{-2}$s$^{-1}$ using the 20 mm aperture, which defines the source size $c$. The experimental setup to characterize the gratings is shown in Fig. 3.8. Inset (a) in Fig. 3.8 shows the source grating mount. This is installed as close as possible to the beam exit port. The red plate is the neutron fast shutter. More details of the mount of the source grating and its functions can be found in [GruDA]. Inset (b) shows the velocity selector installed at the beam line. It is used to select neutrons with an average wave length of $\lambda \simeq 4.1$ Å with a FWHM of $\Delta \lambda/\lambda \simeq 15\%$. This provides sufficient temporal coherence for the experiment. The exit window of the velocity selector is 4 cm $\times$ 4 cm. Inset (c)
Figure 3.8: Experimental setup at the ICON beam line. (a) Installation of the source grating close to the beam exit port. (b) Installation of the velocity selector. (c) Installation of the interferometer box at the first experiment place showing the phase grating, the analyzer grating and the detector (CCD). The draft of the beam line is the 3D version of Fig. 3.2.
Figure 3.9: *Data acquisitions after phase stepping of one grating over its grating period along the direction* $x_g$ *(Fig. 3.1(b)). Two stacks with 10 raw images of sample data and flat field data, and one stack with 5 background images are recorded. The exposure time for a single image was 30 seconds.*

shows the interferometer box, which is installed at the first experimental place of the beam line (compare Fig. 3.2). The interferometer box includes the phase grating, the analyzer grating and the imaging detector (CCD). The assembling of these components is described in more detail in [GruDA].

As a test sample to illustrate the data acquisition and data processing, we used two cylindrical metal rods, a copper rod and a brass rod (copper (58 %), zinc (39 %), lead (3 %)), each with a diameter of 5 mm. The data acquisition as shown in Fig. 3.9 is based on recording three different image stacks. The sample data stack and flat field data stack (object removed from of the beam) were recorded by scanning one of the three gratings over a number of equidistant steps over its grating period, along the transverse direction $x_g$ (compare Fig. 3.1(b)). Ten raw images were taken for the sample data stack and for the flat field data stack. The flat field data stack is necessary on the one hand to correct for inhomogeneous illumination by the neutrons and the camera sensitivity, and on the other hand to correct for wave front distortions (phase offset) originating from the gratings themselves. For our setup, we stepped the source grating $G_0$. Since the period is by a factor 250 larger than that of the analyzer grating, this has the advantage that the phase stepping of $G_0$ could be performed with
lower mechanical precision and stability. The stepping of the other gratings would have provided the same result. Additionally, one stack of background images was obtained by exposing the charge coupled device (CCD) camera while its shutter is closed. Five images were taken for the background data stack, using the same exposure time as that for the data and flat field raw images. The background data stack was used to correct for extraneous noise in the CCD, often caused by hot pixels.

Each raw image of the data and of the flat field stack was recorded using a 100 micron thick Li-6/ZnS converter and fluorescence screen with a 1:1 optical lens system and a cooled CCD (Fingerlake Instrumentation, 1024 × 1024 pixels, pixel size: 24 × 24 µm²). The effective spatial resolution was principally determined by the intrinsic blurring of the scintillation screen to 100 µm [Gru2007]. A typical exposure time for a single raw image was 30 seconds.

The first step in the data processing of the recorded sample and flat field raw images is the background correction. The background data stack is first averaged to a single background image. Each sample and flat field raw image is then background corrected by subtracting the averaged background image.

After phase stepping over one grating period, the intensity signal \( I(m, n, x_g) \) at each detector pixel \((m, n)\) oscillates as a function of \(x_g\), as schematically shown in Fig. 3.10(a). As an example, three different pixels are chosen: the blue marker for a pixel behind the Cu rod, a green marker for a pixel behind the brass rod and a red marker for a region with air. This oscillatory behavior can be seen for the three colored detector pixels in both the sample data image stack and flat field image stack, as shown in Fig. 3.10(b),(c), respectively.

To analyze this oscillation quantitatively, we express the intensity oscillation for each detector pixel with a Fourier series

\[
I(m, n, x_g) = \sum_i a_i(m, n) \cos(i k x_g + \phi_i(m, n)) \\
\approx a_0(m, n) + a_1(m, n) \cos(k x_g + \phi_1(m, n)),
\]

where \(a_i\) are the amplitude coefficients, \(\phi_i\) the corresponding phase coefficients, \(k = 2\pi/p_2\), and \(p_2\) is the period of \(G_2\). In practice, the Fourier components and the phase coefficients are calculated by computing the one-dimensional discrete Fourier transform (DFT)\(^1\) for each pixel. Note, a pure sinusoidal oscillation is uniquely described by \(a_0\), \(a_1\) and \(\phi_1\). Therefore, we can now extract from the sample and flat field data the corresponding amplitude coefficients \((a_0, a_1)\) and phase coefficient \((\phi_1)\) for all detector pixels \((m,n)\).

\(^1\)On a standard personal computer (2.0 GHz processor, 2 GByte memory), the processing time needed for a series of eight images with 10⁶ pixels is in the order of a few seconds.
Figure 3.10: (a) Schematic oscillation of one detector pixel after phase stepping. (b) Oscillation for three detector pixels in the sample data stack as shown in Fig. 3.9. (c) Oscillation for three detector pixels in the flat field stack as shown in Fig. 3.9.
The top row of Fig. 3.11 illustrates how processed transmission images (TI) are obtained. An image of the computed zero amplitude coefficients of the sample data, termed $a_s^0$, is shown in Fig. 3.11 in the top left part. The corresponding image of the zero amplitude coefficients of the flat field data, termed $a_f^0$, is shown in the top middle image. The processed transmission image TI, analogous to what would be measured with a conventional neutron radiography setup without interferometer, is obtained by normalizing $a_s^0$ to $a_f^0$ and is given by

$$TI(m, n) = \frac{a_s^0(m, n)}{a_f^0(m, n)}.$$  \hfill (3.26)

The TI is shown in Fig. 3.11 in the top right corner. Due to the similar absorption cross section of copper and brass, the rods are difficult to tell apart.

Analyzing the lateral shift of the intensity modulation, $\phi_1(m, n)$, leads to differential phase contrast images (DPCI) for neutrons, as reported in [GruDA, Pfe2006n] and for X-rays in [Pfe2006x]. DPCI are derived from the component of the gradient of the projected wave-front phase profile $\Phi(m, n)$ of the object and are related to the phase of the intensity oscillation in each pixel, $\phi_1(m, n)$, by [Wei2005]

$$\nabla_x \Phi(m, n) = \frac{p^2}{\lambda d} \phi_1(m, n).$$  \hfill (3.27)

The data processing for the DPCI is illustrated in the middle row in Fig. 3.11. The left image displays the background corrected sample phase coefficients $\phi_s^1$ and the middle image shows the background corrected flat field phase coefficients $\phi_f^1$. To eliminate imperfections from the imaging system, or the incoming wavefront, the effective DPCI is calculated by taking the difference of a measurement with sample $\phi_s^1$ and without sample $\phi_f^1$. Together with the correction for phase wrapping, the DPCI is given by

$$DPCI(m, n) = \phi_s^1(m, n) - \phi_f^1(m, n),$$  \hfill (3.28)

and is shown in the middle right image in Fig. 3.11.

The analysis of the first amplitude coefficients $a_1$ of the sample data and flat field data stacks gives the DFI. The left image in the bottom row of Fig. 3.11 illustrates the first sample amplitude coefficients $a_s^1$ normalized to the zero amplitude coefficients $a_s^0$. The middle image displays the first flat field amplitude coefficients $a_f^1$ normalized to the zero flat field coefficients $a_f^0$. This image, without sample in place, contains information about the maximum contrast produced by the interferometer, namely the visibility, $V$:

$$V(m, n) \equiv \frac{I_{\text{max}}^f - I_{\text{min}}^f}{I_{\text{max}}^f + I_{\text{min}}^f} = \frac{a_f^1(m, n)}{a_f^0(m, n)}.$$  \hfill (3.29)

This evaluation is reported in the next section. The DFI is obtained by normalizing the first sample amplitude coefficients $a_s^1/a_s^0$ to $V$, and is given by

$$DFI(m, n) = \frac{V^*(m, n)}{V^I(m, n)} = \frac{a_s^1(m, n) \cdot a_f^0(m, n)}{a_s^0(m, n) \cdot a_f^1(m, n)}.$$  \hfill (3.30)
3.4. Data acquisition and data processing

Figure 3.11: Data processing to simultaneously obtain the transmission image ($\text{TI} = a^s_0/a^f_0$), differential phase contrast image ($\text{DPCI} = \phi^s_1 - \phi^f_1$) and dark-field image ($\text{DFI} = a^s_1 a^f_0 / a^s_0 a^f_1$). For each detector pixel the amplitude coefficients $a_0, a_1/a_0$ and phase $\phi_i$ coefficients are plotted in one of the images. The coefficients are obtained from the intensity oscillation after phase stepping (see Fig. 3.10(a)). The superscripts $s$ and $f$ denote the values measured with the specimen in place and as a reference without specimen, respectively.

The image at the bottom right corner of Fig. 3.11 shows the corresponding DFI. For plane, homogeneous and non-magnetic specimens, i.e., for samples that only introduce a constant phase shift due to the neutron’s nuclear interaction potential as for instance the copper rod does, the DFI image shows only a contribution at sample edges where the neutrons are refracted. However, specimens that exhibit a strongly varying nuclear interaction potential, such as the brass alloy, with its lead precipitations in the copper-zinc matrix, show a significant contribution to the DFI. Image contrast also arises from the magnetic interaction potential of ferromagnetic materials, namely ferromagnetic domains and domain walls, as reported in the next chapter. More generally, we note that the DFI is a quantity that is inversely proportional to the ability of the sample...
to locally degrade the coherence of a well defined neutron wave front, either through scattering or multiple refraction at nuclear density or magnetic potential fluctuations in the sample.

## 3.5 Characterization of the diffraction gratings

The performance of the gratings was characterized with a pixel wise evaluation of the visibility \cite{Gru2008a}. For the evaluation of the visibility, sixteen raw images with 10 seconds exposure each were taken over two periods of $G_0$. They can be regarded as flat field raw data and were background corrected in the same manner as described before. The duty cycle of the source grating was set to 0.4, i.e. for the largest slit width, thereby maximizing the available flux through the source grating.

The justification that the computed Fourier coefficients $a_{f,0}$, $a_{f,1}$ and $\phi_{f,1}$ can be used to characterize the gratings in terms of the visibility $V$, is illustrated in Fig. 3.12(a). Here the cosine function described by these terms fits well to the recorded data points, following the intensity oscillations.

The result of the evaluated visibility $V$ over the total grating area with the corresponding values of $V(m, n)$ is shown in Fig. 3.12(b). The visibility is homogeneously distributed over the grating area, ranging from 21 % to 25 %. The maximum achievable visibility for a monochromatic three grating interferometer with perfectly opaque $G_0$ and $G_2$, an ideal phase grating $G_1$, and a DC of 0.5 is 50 % for a monochromatic beam. Considering the low temporal coherence conditions of our setup $\Delta \lambda/\lambda \simeq 15\%$, the achieved visibility is remarkably high. However, the largest value of $s$ leads to the smallest spatial coherence length of the setup, resulting in the lowest visibility value concerning the slit width of $G_0$. Reducing $s$ can additionally improve the visibility of the setup. Measurements have shown that for the smallest value of $s$ the averaged visibility improves slightly with an averaged enhancement of about 3 percentage points. However, under these conditions the available neutrons flux reduces dramatically. Consequently, for the following experiments we use the maximum value of $s$.

The experimental results of bulk magnetization processes, which are described in the following chapter, and bulk magnetic domain imaging in chapter 6 are based on the DFI contrast. Therefore, having high visibility of the grating interferometer setup is directly linked to having a large dynamic range for the DFI measurements.
Figure 3.12: Characterization of the diffraction gratings in terms of visibility. (a) Measured neutron data points (red markers) of the intensity modulation for a single detector pixel when one of the gratings is scanned over two grating periods along $x_g$. A discrete Fourier transformation yields the corresponding Fourier series coefficients $a_0$, $a_1$ and $\Phi_1$. The corresponding cosine function (blue line) obtained from the extracted Fourier coefficients nicely matches the oscillation behavior. (b) Visibility map over the whole grating area of $64 \times 64 \text{ mm}^2$ showing the values of $a_1/a_0$ for each detector pixel with an averaged value of 23 %. 
Chapter 4

Interferometry experiments I: Magnetization processes

In this chapter, the experimental results of the magnetization processes of single and poly-crystalline materials with different sample geometries are presented. The experimental setup is reported in section 4.1. In section 4.2 the contrast origin for ferromagnetic samples is explained. In section 4.3, for the single-crystalline material, a (100)-oriented iron silicide disc was primarily characterized by the Laue method to obtain the crystallographic directions, as reported in subsection 4.3.1. In subsection 4.3.2, the magnetization process results of the disc are presented. For a poly-crystalline quadratic steel plate, the angular dependence of the magnetization process was imaged and the results are shown in section 4.4. In the last section 4.5, the magnetization results are compared to finite element simulations, firstly for the single-crystalline sample in subsection 4.5.1 and secondly for the poly-crystalline sample in subsection 4.5.2.

4.1 Experimental setup

The experiments to image the magnetization processes were again carried out at SINQ using the beam port of the cold neutron imaging facility ICON. The schematic layout of the experimental setup is shown in Fig. 4.1(a) [Gru2008b]. It consists of a source grating $G_0$, a phase grating $G_1$ and an analyzer attenuation grating $G_2$. These are the components which were already used in the setup to characterize the gratings. To magnetize the samples, we installed a standard dipole electromagnet (GMW associates, C-frame 3470) with plane cylindrical pole shoes of 40 mm diameter. Photographs of the setup at the experimental station (compare Fig. 3.2) are shown in Fig. 4.2. In Fig. 4.2(a), the electromagnet, the camera optic, including the CCD camera, and the sample holder in the hand of the author can be seen. Fig. 4.2(b) gives a detailed view on the setup showing the sample holder, as well as the phase and the analyzer grating mounted on brackets. The pole gap was 40 mm as seen in Fig. 4.3(a) producing an
Figure 4.1: (a) Setup with the source grating $G_0$, the phase grating $G_1$, the analyzer absorption grating $G_2$, and an electromagnet creating a horizontal variable magnetic field around the sample. The sample can be rotated by an angle $\theta$ around the beam axis. (b) Multiple refraction/scattering of neutrons at magnetic domain walls in the sample causes a local degradation of the coherence of the neutron wave field and results in a decrease of the local fringe visibility of the interference pattern.
4.1. Experimental setup

![Experimental setup diagram](image)

Figure 4.2: Experimental setup at the experiment station (compare Fig. 3.8(c).) (a) Setup showing the electromagnet, the camera optic including the CCD camera and a sample holder. (b) Setup showing the installed phase and analyzer grating mounted on brackets and the mounted sample holder with the sample adjusted between the pole shoes (more detailed in Fig. 4.3(a)).
Chapter 4. Interferometry experiments I: Magnetization processes

Figure 4.3: (a) Photograph of the dipole electromagnet showing the adjusted pole gap of 40 mm, and the plane cylindrical pole shoes of 40 mm diameter, as well as the front end of the sample holder which will be centered in the magnetic field. (b) Photograph of the single-crystalline (100)-oriented FeSi disc with a diameter of 12 mm, mounted with plastic screws on the front of the holder. (c) Photograph of the poly-crystalline steel plate with an edge length of 15 mm. This orientation will be considered as the $\theta = 0^\circ$ case in the following.

Almost uniform horizontal magnetic field\(^1\) with $\Delta H/H < 10^{-3}$ in the sample region. The magnet was driven by a unipolar power supply (Pulse Power & Measurement Ltd., Lambda Genesys GEN 40-19). The maximum input coil current of the magnet was 5 A. For this geometry the maximum field between the poles was measured with a Hall probe to be 250 mT.

The samples used for the experiments are shown in Fig. 4.3(b) and (c). The first is a single-crystalline (100)-oriented iron silicide (FeSi) disc with a diameter of 12 mm. The disc had a thickness of 500 $\mu$m and was cut from a Fe 3 at% Si single crystal alloy. The single crystal was grown using a rf/heated floating zone technique in hydrogen with a rate of 7 mm/h. The sample was produced by Jaromír Kopecek from the group of Pavel Lejček at the Institute of Physics in Prague. The second sample is a poly-crystalline steel plate (steel grade: DC 01 (St 12.03), from the work shop at PSI) as shown in Fig. 4.3(c). The sample has a square form with an edge of 15 mm and a thickness of 750 $\mu$m. Both samples, are mounted with plastic screws on an aluminium capping plate shown in Fig. 4.3(b),(c). The cap itself is fixed with aluminium tape on an aluminium rod, which serves as the sample holder (Fig. 4.2). The sample holder is mounted in the setup on a three axis positioning system to accurately place the sample centered between the pole shoes.

For the setup, we have the option to rotate the holder around the beam axis by an angle $\theta$, as illustrated in Fig. 4.1(a). This is used to image the angular dependence of the magnetization process of the steel plate. For the single-crystalline disc it offers the precise parallel alignment of the crystallographic axis of the sample with the axis of the external magnetic field.

\(^1\)For the remaining pages of the thesis the magnetic field $H$, the magnetic induction $B$, and the magnetization $M$ are associated only with its magnitudes $|H| = H$, $|B| = B$ and $|M| = M$. 

4.2 Contrast origin for ferromagnetic materials

Before the results of the magnetization processes for the single-crystalline and poly-crystalline materials are presented, the underlying contrast origin to image the magnetization processes is explained. The recorded images are based on the dark-field image contrast, DFI, as introduced in the previous chapter. The analysis of the amplitude of the oscillation, $a_1(m,n)$ (compare Fig. 3.10(a)), similar to what has been used to characterize the grating in terms of visibility in section 3.4., yields spatially resolved information on the magnetic domain wall density distribution in the sample. The analysis is based on the fact that the neutron beam undergoes multiple refractions at the magnetic domain walls [Sch1976], which results in a local degradation of the coherence of the neutrons exiting the sample [Pfe2005]. This local degradation decreases the ability of the neutrons to interfere with each other after the phase grating $G_1$ and leads to a local decrease of the interference pattern, as depicted in Fig. 4.1(b). This yields locally smaller values of the fringe visibility detected in the intensity oscillation $I(m,n,x_g)$. For a single DFI, we record eight images for the flat field and sample data stack, over one grating period, with an exposure time of 30 seconds. The effective spatial resolution of 100 $\mu$m was determined by the intrinsic blurring in the scintillation screen [Gru2007].

Figure 4.4 displays experimental results obtained from the (100)-oriented iron silicon single-crystalline disc [Gru2008b]. The conventional TI is shown in Fig. 4.4(a). The corresponding DFI is presented in Fig. 4.4(c). Only the strongly attenuating plastic screws used to mount the disc are visible in the TI. The sample itself is invisible, due to the weak absorption of the “thin” disc. In contrast, the FeSi disc is clearly visible in the DFI. This is seen in Fig. 4.4(b), where the intensity oscillations for two detector pixels is extracted from a series of eight images taken at different values of $x_g$. It is seen that the amplitude coefficient $a_1(m,n)$ of the oscillation extracted behind the disc (blue marker) is very low. This leads to low contrast values in the DFI. The plastic screws are not visible in the DFI, they cause no significant contributions, because plastic is a homogeneous material with essentially no density fluctuations on the relevant length scale.

Fig. 4.4(d) shows the surface domain structure of the specimen recorded by magneto-optical Kerr effect (MOKE) microscopy in the center region of the sample. The MOKE images were obtained with a wide-field Kerr microscope, with image processing for contrast enhancement, performed under real-time conditions [Hub1998, Kro2007]. The zigzag domain walls in Fig. 4.4(d) indicate that the domains are closure domains of internal basic domains, magnetized perpendicularly to the sheet surface (V-line pat-

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1For the MOKE investigation, the rust on the disc was removed with hydrochloric acid, then lapped and carefully polished by using a colloidal suspension of amorphous SiO$_2$. This chemical-mechanical treatment prevents the formation of surface damage by polishing which is essential for MOKE microscopy. We note that the surface polishing was only necessary to acquire the MOKE image; neutron DFI contrast can as well be obtained on unpolished samples.
Figure 4.4: Results obtained for the (100)-oriented single-crystalline FeSi disc. (a) Conventional neutron transmission image (TI). (b) Intensity oscillations for two detector pixels from a series of eight taken images at different values of $x_g$ with 30 seconds exposure time. (c) Neutron dark-field image (DFI) of the same sample. The total exposure time together for the TI and DFI was eight minutes including the data and flat-field image acquisition. (d) Corresponding magneto-optical Kerr effect (MOKE) image of a section of the disc, showing the surface domain structure.

tern [Hub1998]). With a width in the 10 $\mu$m range, these domains are well below the lateral resolution limit of the imaging detector and thus are not resolved individually in Fig.4.4(c). Similar, or even more complex domain structures, are found everywhere on the sample surface, indicating that the specimen is in a mechanically stressed state.

In the following, the magnetization process of single-crystalline and poly-crystalline materials are imaged by spatially resolving the magnetic domain wall density in the samples. Note that in cases where the internal domain structure is larger than the spatial resolution limit of the detection system, single domain walls can be resolved by this technique, as reported in chapter 6.
4.3 Single-crystalline material

The (100)-oriented FeSi disc is primarily characterized by the Laue method to obtain the crystallographic orientation of the disc. The results are shown in subsection 4.3.1. In section 4.3.2 the results of the magnetization process are presented.

4.3.1 Sample characterization

The crystallographic orientations of the single-crystalline (100)-oriented FeSi disc were obtained using the Laue method [Laue], which is often used for accurate crystal orientation. With the Laue analysis, one can determine the orientation of an unspecified crystal using diffraction of x-rays. A polychromatic x-ray beam shines on a crystal and the diffracted beams are photographed by a camera. We used the so-called back-reflection Laue method, where the film is placed between the x-ray source and the crystal, and the backward diffracted beams are recorded.

The diffracted beams form a map of spots as shown in Fig. 4.5(a). By using this map and the known crystal orientation, it is possible using the “Orient Express” program to index each spot, i.e. attribute it to a particular plane. Therefore one can determine the crystallographic directions of the disc as illustrated in Fig. 4.5(b),(c). The directions shown in Fig. 4.5(c) are the magnetic easy axis (soft axis) for the FeSi sample. The magnetic hard axis are those directions which lie on the diagonals. With the knowledge of the crystallographic axis of the disc, the magnetization process experiments are performed and the results are presented in the following.

4.3.2 Magnetization process

The efficiency of our setup, with total exposure times of typically four minutes per DFI, allows us to study the dynamic response of the specimen under the influence of an externally applied magnetic field. For a comparison, an exposure time of 12 h for a single phase projection measured at an equivalent neutron source (National Institute of Standards) is reported in [McM2003]. In [Dub2005], a value of 2 h per image at

![Figure 4.5: (a) Laue image of the (100)-oriented single-crystalline FeSi disc. (b) Indexed Laue image of the FeSi disc to obtain the crystallographic orientation. (c) Determined misalignment angle of 11°.](image)
a 5 to 10 times more intense neutron source (Institute Laue-Langevin) is stated. We conclude that our method, which requires typically 8 raw images of 30 sec exposure time for the simultaneously obtained TI, DPCI and DFI, yields a decrease in exposure times by more than a factor of 100.

The results of the magnetization process of the single-crystalline (100)-oriented FeSi disc are presented in Fig. 4.6 [Gru2008b]. For this experiment, the sample was placed such that a magnetic hard axes, which is the [011] direction of the disc, is aligned parallel to the external horizontal magnetic field axis. As mentioned above, an easy axis refers to the energetically favorable direction of the spontaneous magnetization in a ferromagnetic material. By applying a field along this axis the sample is rather easy to magnetize. In our case the easy axes are the cubic axes forming an angle of 90° as determined by the Laue method (Fig. 4.5). The magnetic hard axes are therefore lying on the diagonals between the easy axis under an angle of 45°. The magnetization along this axis is rather hard, since the internal spontaneous magnetization directions are not parallel to the applied external magnetic field. The field is ramped from $-60 \text{ mT} \leq H \leq 60 \text{ mT}$ in steps of 6 mT. For small magnetic field values of $|H| \leq 20 \text{ mT}$ the disc is clearly visible in the DFIs, indicating the rich multi-domain structure in the specimen. When the magnetic field is increased to values of $|H| \approx 24 \text{ mT}$, the presence of a vertically oriented zone, which starts growing from the center of the disc, becomes visible in the corresponding DFIs. This zone indicates the presence of a monodomain state of a certain, but unknown, magnetization direction. At absolute values of $|H| \geq 30 \text{ mT}$ these regions in the sample are almost fully magnetized along the [011] direction. With increasing field, the zone of saturation expands towards the sample edges that are oriented transverse to the magnetic field axis. This inhomogeneous saturation process is caused by locally varying, internal effective magnetic fields due to inhomogeneous demagnetizing fields caused by the non-ellipsoidal sample shape. The whole sample is almost magnetized when $|H| \approx 60 \text{ mT}$. Reversing the direction of the magnetic field ramp did not reveal any sign of remanence in the magnetization process for this particular sample (see also Fig. 5.6).
Figure 4.6: Neutron dark-field images of the (100)-oriented FeSi single-crystalline disc as a function of a horizontally applied external magnetic field. The presence of a vertically elongated magnetic mono-domain starting to grow from the inside for magnetic field values of $|H| \geq 24$ mT is clearly visible. The sample is almost completely magnetized at $|H| = 60$ mT. 8 phase-steps over one grating period, with an exposure time of 30 seconds per image, were taken to process each DFI.
4.4 Poly-crystalline material

In this section, the results of the magnetization process of the poly-crystalline steel are presented. The magnetization process is imaged for different orientation angles $\theta$ of the plate with respect to the horizontal magnetic field, which was kept the same as for the single-crystalline disc. The magnetic field is ramped from $0 \text{ mT} \leq H \leq 250 \text{ mT}$ in 41 steps. 250 mT is the maximum achievable field for this geometry, limited by the maximum coil current of 5 A. The angular dependence of the magnetization process is shown in the following subsection for three different orientations of the steel plate. These are $\theta = 0^\circ$ (compare Fig. 4.5(c)), $\theta = 22.5^\circ$ and $\theta = 45^\circ$, with respect to the external horizontal magnetic field. 8 phase-steps over one grating period, with an exposure time of 30 seconds per image, were taken to process each DFI.

4.4.1 Angular dependence of the magnetization processes

Magnetization process of the steel plate at $\theta = 0^\circ$: The experimental results are shown in Fig. 4.7. In this case, the external magnetic field axis is parallel to the sample edge. For field values of $H \leq 81 \text{ mT}$ the steel plate is clearly and fully visible in the DFIs, due to the rich multi-domain structure in the specimen. When the magnetic field is increased to values of $H \approx 88 \text{ mT}$, contrast develops from the middle of the plate in the form of a vertical stripe. At field values of $H \geq 94 \text{ mT}$, a vertically elongated region in the middle of the sample with less density of domain walls is visible. However, at each vertical edge of the plate over its whole length, a domain-wall-rich rectangular area remains. For increasing field values between $100 \text{ mT} \leq H \leq 125 \text{ mT}$, the domain-wall-free area in the middle expands toward the sample edges, gaining rapidly in width. At field values between $131 \text{ mT} \leq H \leq 250 \text{ mT}$, the domain-wall-free area only slowly gains in width. For maximum external field value of $H = 250 \text{ mT}$, the larger part of the plate is magnetized beside two small vertical domain-wall-rich strips at the edges.

Magnetization process of the steel plate at $\theta = 22.5^\circ$ orientation: For this case, the steel plate is rotated clockwise by an angle of $22.5^\circ$. The experimental results are shown in Fig. 4.8. For field values of $H \leq 94 \text{ mT}$ the steel plate is visible in the DFIs. When the magnetic field is increased to $H = 100 \text{ mT}$, the contrast recovers, forming a rectangular, vertically inclined stripe in the middle of the plate. The starting position in the middle of the sample is similar to that of the $0^\circ$ case. However, the starting point of the contrast development is 6 mT later. At field values between $106 \text{ mT} \leq H \leq 125 \text{ mT}$, the domain-wall-free rectangular area in the middle expands toward the sample edges and gains rapidly in width. At $H = 131 \text{ mT}$, the rectangular

\footnote{The yellow color in the DFIs indicates domain-wall-rich areas.}
domain wall free area collapses. For field values between $131 \text{ mT} \leq H \leq 175 \text{ mT}$, the domain-wall-free area expands towards the sample corners, which are closer to the pole shoes (top right, bottom left), until these corners are fully magnetized. Further increase of the field to $181 \text{ mT} \leq H \leq 250 \text{ mT}$, leaves two small domain wall rich rectangular areas remaining on the top left and bottom right corner, which shrink slowly with increasing field.

**Magnetization process of the steel plate at $\theta = 45^\circ$ orientation:**
The steel plate is now oriented by an angle of $45^\circ$, i.e. one diagonal is oriented along the magnetic field axis. The experimental results are shown in Fig. 4.9. Up to field values of $H \leq 94 \text{ mT}$, the steel plate is completely visible in the DFIs. For the following magnet field step at $H = 100 \text{ mT}$ the contrast starts to set in again, in the form of two mirror-symmetrical vertically elongated areas. At $H = 106 \text{ mT}$, two oval grainy areas are visible, although the horizontal edges are still not magnetized. For field values between $113 \text{ mT} \leq H \leq 131 \text{ mT}$, these two oval areas expand towards the middle part of the plate as well as to the edges close to the pole shoes. For field values between $138 \text{ mT} \leq H \leq 250 \text{ mT}$, the remaining domain wall rich areas are found at the top and bottom corners which form a rhombus. The area of the rhombus decreases for increasing external field values.
Figure 4.7: Magnetization process of the poly-crystalline steel plate at 0° orientation. At $H = 94$ mT, the formation of an elongated vertical rectangular area with less domain wall density is visible. This area expands towards the vertical sample edges for increasing field values. However, two vertical domain wall rich stripes at the sample edges remain at maximum field of $H = 250$ mT.
Figure 4.8: Magnetization process of the poly-crystalline steel plate at 22.5° orientation. At $H = 100$ mT, the formation of an elongated vertical rectangular inclined area with less domain wall density is visible, which expands towards the sample edges for increasing field values of up to $H = 125$ mT. For further increase of the field, this area collapses and a transformation is made into two remaining domain wall rich areas at the top left and bottom right corners, which shrink slowly, but still are present at maximum field.
Figure 4.9: Magnetization process of the poly-crystalline steel plate at 45° orientation. At $H = 106$ mT, the formation of two mirror-symmetrical oval areas with less domain wall density are visible. Both are growing for $113 \text{ mT} \leq H \leq 131 \text{ mT}$ until two rhombohedral areas at the top and bottom corner are formed. For further increase of the field, these areas decrease, but are still visible at maximum field.
4.5 Comparison with FEM simulations: Results and interpretations

To understand our measured data of the magnetization process of the single-crystalline (100)-oriented FeSi disc and the poly-crystalline steel plate, we compare the experimental results with simulation results. The question arises which method offers the best possibility to simulate and mirror our experimental results. As both samples need to be considered as “large samples”, the contribution of individual domain walls will be completely negligible. For large samples, the detailed geometrical arrangement of domain patterns is ignored, but the spatial periods of the implied domains must be smaller compared to the sample size in all dimensions. This holds in our case as seen in Fig. 4.4(c),(d). Thus, the theory of micro magnetism cannot be applied, especially not for three dimensions [DeS2001, DeS2002].

The only theoretical method to explain magnetization processes of such “large samples”, is an approach with “phase theory” ([Chi1999], [Kne1962], [Hub1998]). Magnetization phases are defined in a similar sense to phases in metallography or in thermodynamics. All domains magnetized in the same direction are gathered into a phase, characterized only by its volume and its magnetization direction. This approximation is valid if the sample is sufficiently large in at least one dimension. Phase theory therefore applies to extended homogeneous and soft magnetic materials, as is our case.

For the interpretation of our measured data, we discuss the magnetization process on the basis of magnetization curves (hysteresis curve, $M-H$ curve) in the context of phase theory. A measured $M-H$ curve (here for the steel plate) is shown in Fig. 4.11. Due to the “small” diameter of the single-crystalline (100)-oriented FeSi disc of 10 mm, we were not able to measure the corresponding $M-H$ curve for a sample geometry with a zero demagnetizing factor [Kne1962]. This is obtained for a sample geometry in the form of a torus as reported in the following. The minimum diameter of the sample for this measurement is 30 mm due to the measurement system. In the magnetization curve, two different modes can be distinguished. Mode I is where the domain wall movement takes place. This movement is associated with a rapid jump in the magnetization curve as seen in Fig. 4.11. The magnetization directions of all participating phases, namely the magnetic domains in this mode, stay constant. The domain wall displacement characterizing Mode I only takes place at small and moderate external field values. Mode II is associated with larger field values, where rotation processes of the magnetization directions of phases dominate. Concerning the magnetization curve, only a slow increase of the magnetization $M$ occurs, as seen in Fig. 4.11. The characteristic point in the magnetization curve is the transition point, called the “knee”, which separates the two modes. Above the “knee”, the external field penetrates the sample and there are no longer domain walls left and only the rotation of the magnetic phases takes place. Below the “knee”, the external field is expelled and only domain wall motion occurs.
For the interpretation of our measured data it is important that, for the transition between Mode I and Mode II, where the external field starts to penetrate the sample, the DFI contrast will reappear, because no domains walls where the neutrons can be refracted remain. In simple terms, in Mode I, the contrast in the DFI is low, whereas in Mode II, the DFI contrast will develop due to the lack of domain walls.

The data points in the hysteresis curve in Fig. 4.11 are obtained by averaging over a sample volume. For the hysteresis curve measurement, a torus of the corresponding material was prepared. Due to this geometry no demagnetization effects will take place and the demagnetization factor of the sample is zero [Kne1962]. With this ring geometry of the material, the intrinsic permeability (geometry independent) can be determined and will be used later as an input parameter in the simulation program. However, if we know the internal magnetic induction values $B$ at each point in the samples for the applied external magnetic field values $H$, then we can interpret our measured results. $B$ is also known as magnetic induction, $B = \mu_0 (H + M)$ and therefore $M = \frac{B}{\mu_0} - H$. In the hysteresis curve in Fig. 4.10 the magnetization $M$ is plotted. Since the values of $H$ [mT] are very small compared to $B$ [T], there is no relevant difference between $M$ and $B$ concerning the hysteresis curve unless the saturation point is reached, where a linear increase in the $B$-$H$ curve will take place. With the knowledge of the local $M$ field values of the samples, and their corresponding hysteresis curve, described by the permeability and the saturation magnetization, we can compare the measured data with the simulation results.

We are able to simulate the local magnetic induction $B$ field values in the sample for each external field value of $H$, for a given sample geometry and orientation. Our simulations are based on the finite element method (FEM) analysis, which is a numerical technique for obtaining approximate solutions of boundary-value problems such as partial differential equations (PDE) as well as integral equations of mathematical physics [Zie2005, Bas2003, Jin1993]. The solution approach is based either on eliminating the differential equation completely (steady state problems), or on rendering the PDE as an approximate system of ordinary differential equations, which are then numerically integrated. In our case, these are the solutions of the Maxwell equations for the electrostatic case. We used the commercially available AMPERES 6.0 software [FEM], which is a 3D magnetic field solver for such applications. Using this FEM software, we are able to simulate, in three dimensions, the internal magnetic induction $B$ for a given sample and external magnetic field geometry. The simulations were made with the help of Yiu Wai Lai from the IFW in Dresden.
4.5.1 Single-crystalline material

As already mentioned, the 12 mm diameter of the single-crystalline (100)-oriented FeSi disc is too small to perform a hysteresis curve $M-H$ measurement to determine the intrinsic permeability. The necessary sample diameter to perform the hysteresis measurement with a ring-shaped sample is 30 mm. Therefore, we resort to literature [Wij1992, Woh1980, Hor2006, Boo1962], where we find the value for the intrinsic permeability, as well as the saturation magnetization, of the single-crystalline FeSi material. The intrinsic permeability is $\sim 7000$, and the saturation magnetization $M_S$ is 0.9 T. The “knee” of the $M-H$ curve for the Fe 3 at% Si material is $M_K = \frac{1}{\sqrt{2}} \cdot M_S \approx 0.63$ T. The value $\frac{1}{\sqrt{2}}$ is given for the magnetization parallel to a [110] direction for cubic crystals [Kne1962], which holds for our situation, where the field is applied along a magnetic hard axis.

The comparison of the DFI results with the FEM simulations is shown in Fig. 4.10. For the simulation, we considered a homogenous cylindrical magnetic field, as created by the pole shoes with the dimensions for the diameter as 40 mm and length as 40 mm. The simulated $B$ field images are scaled to the maximum value $M_K \approx 0.63$ T. Every $B$ field value in the sample larger than $M_K$ appears as white in the simulation results. The white color is associated with areas where Mode II is dominating. In these areas no domain walls are left and correspondingly no scattering can occur.

What we can directly see in the FEM simulations is the same qualitative magnetization behavior as obtained in the DFI measurements. In the FEM simulations, we see an ellipsoidal area, which starts to form from the center of the disc. In both, the DFI and FEM results, a rapid transition between $24 \text{ mT} \leq H \leq 42 \text{ mT}$ is observed. Furthermore, we see that the ellipsoidal area grows along the vertical direction until it reaches the edges of the disc on the top and bottom. The white area in the FEM simulation expands along the horizontal directions, which is also observed in the DFI’s. Finally, the simulations show the same results as the DFI regarding the two curved stripes at the left and right edge, which are slowly shrinking and almost entirely disappearing.
Figure 4.10: Comparison of the DFI results with FEM simulation ones for the FeSi (100)-oriented single-crystalline disc, and external field values of $0 \text{ mT} \leq H \leq 60 \text{ mT}$. White areas in the simulation are associated with Mode II ($B > 0.63 \text{ T}$), where the external field penetrates the sample and no more domain walls are present. These should correspond to the same contrast as for air in the corresponding DFIs. A very good agreement is observed.
4.5.2 Poly-crystalline material

Since the steel plate was cut from a large piece of material, we were able to prepare a torus sample of the same material with a circular cross section, in order to determine the intrinsic permeability. The outer diameter of the ring was 30 mm, and the inner one 15 mm. The $M-H$ curve of the torus is measured with two sets of coil wrapped around it. On passing an electric current through the outer coil, a magnetic field is induced. All loops of the wire make a toroidal contribution to the magnetic field in the same direction inside the torus, namely a ring shaped magnetic field. The magnetic induction in the torus is then picked-up by the inner coil. The relation between the input field $H$ and the output field $B$, with respect to $M$, is obtained and so the intrinsic permeability of the material is deduced. The measured hysteresis curve ($M-H$) of the steel plate material is shown in Fig. 4.11. The “knee” of the curve occurs at $M_K = 0.83 \cdot M_S \approx 1.7$ T for the poly-crystalline steel plate material, where $M_S = 2.1$ T.

![Figure 4.11: Measured magnetization ($M-H$) curve of the steel plate material. The saturation magnetization $M_S$ is 2.1 T. In the region of Mode I domain wall movement takes place, whereas in the region of Mode II magnetization rotation processes occur. The transition point, called the “knee” of the magnetization curve, is given for this material at $M_K = 0.83 \cdot M_S = 1.7$ T.](image)
is the saturation magnetization value. The value 0.83 is a statistical factor giving the ratio between saturation magnetization and remanent magnetization in poly-crystalline materials with cubic anisotropy [Hub1998].

The FEM simulation results are shown together with the measured DFI results for the 0°, 22.5° and 45° orientations of the steel plate in Fig. 4.12, Fig. 4.13 and Fig. 4.14, respectively. The simulations show the local internal magnetic induction values as a function of the externally applied magnetic field values for \( 0 \text{ mT} \leq H \leq 250 \text{ mT} \). The simulated \( B \) field images are scaled to \( M_K \approx 1.7 \text{ T} \). Every \( B \) field value larger than \( M_K \) appears as white in the simulation results. It is seen that for the different sample orientations (0°, 22.5° and 45°) with respect to the external \( H \) field, the simulated \( B \) field distribution in the simulations appear different.

Comparing the FEM simulations with the DFI measurements, the FEM simulations do not qualitatively reflect the DFI results, unlike those obtained for the FeSi single-crystalline disc. However, the trend of the magnetization behavior is reproduced in each of the simulations. For all three orientations, it is imperative that for larger field values the simulation results agree better than for the lower ones. Considering the 0° case in Fig. 4.12, the tendency that the sample starts to magnetize from the center is clearly followed, as well as the increase of the magnetized area in horizontal direction. The curvatures of the white areas, as shown in the simulations, are not obtained in the DFIs, nor is the correct starting point of the magnetization process. In the 22.5° case in Fig. 4.13, the magnetization behavior as simulated in the FEM images can also be seen in the DFIs. Both the inclined domain wall free area and the growing of these areas are seen. In the 45° case in Fig. 4.14, the deviations between the FEM results and the DFI results are largest. At larger \( H \) fields, the diamond shaped areas at the top and bottom edges collapse, which is not observed in the corresponding DFI images. The starting points of magnetization processes for the three different orientations, are not reproduced in the FEM simulations.
Figure 4.12: Comparison of the DFI results with FEM simulation results for the $0^\circ$ orientation of the steel plate and external field values of $0 \text{ mT} \leq H \leq 250 \text{ mT}$. The white areas in the simulation are associated with Mode II, where the external field penetrates the sample and no domain walls are present. These should correspond to red colors in the corresponding DFIs. The tendency of the growth of the domain wall free area can be observed by the FEM simulations.
Figure 4.13: Comparison of the DFI results with the FEM simulation results for the 22.5° orientation of the steel plate and external field values of $0 \text{ mT} \leq H \leq 250 \text{ mT}$. 
Figure 4.14: Comparison of the DFI results with the FEM simulation ones for the 45° orientation of the steel plate and external field values of 0 mT ≤ H ≤ 250 mT.
As pointed out earlier, the steel plate with its microscopic structure is too complex to simulate with a full micro-magnetic model. However, a case that is quite similar to the steel plate in the $\theta = 45^\circ$ orientation can be found in literature [DeS2001, DeS2002]. DeSimone and colleagues calculated the magnetization response of a soft ferromagnetic film to an in-plane applied magnetic field. The results are shown in Fig. 4.15. These simulated results were also verified by MOKE measurements. The sample they used was a square of permalloy ($\text{Ni}_{81}\text{Fe}_{19}$) with an edge length of 60 $\mu$m and a sample thickness of 230 nm. Starting from the Landau domain pattern in Fig. 4.15(a), the sample was magnetized by a horizontal field as shown in (b)-(d). In these images the domain wall movement is visible. For increasing field values, the external field penetrates the sample as shown in (e)-(g) indicated by the black curved lines. The lines show the level curves of the potential of the penetrated field. It is interesting to see that this penetration behavior can also be observed in our case for the steel plate. The development and the propagation of these lines take place in the same geometry as for our steel plate, as seen in Fig. 4.9. The two diamond shaped areas as simulated for the Landau domain pattern can also be observed in our DFI results in Fig. 4.9 at $H > 125$ mT.

It is also interesting to note that our imaging results of the magnetization process of the steel plate in the $45^\circ$ orientation qualitatively show the same behavior as for the calculated magnetization behavior (micro-magnetic simulations) of the permalloy sample. Bear in mind that micro-magnetic simulations can never be used for bulk magnetic samples simulations, due to the complicated and unknown composition of the grain structure of the sample. However, the close analogy of comparing the micro-magnetic simulations and the DFI results allows us to close the gap between bulk magnetic magnetization processes analysis.
4.5. Comparison with FEM simulations: Results and interpretations

Figure 4.15: Simulation results for low energy domain patterns in permalloy ($\text{Ni}_{81}\text{Fe}_{19}$, edge length of 60 $\mu$m, sample thickness of 230 nm) obtained by two-dimensional micromagnetic calculations, which serve to link and verify our experimental observations for the 45° case (see Fig. 4.9). The images are taken from [DeS2001, DeS2002]. (a) Landau domain pattern. (b)-(d) Horizontal applied $H$-field (reduced units and thus dimensionless) as in our case, leading to domain wall displacement in the sample. However, the external field is still expelled. (e)-(g) Penetration of the external field into the sample. The lines show the level curves of the potential of the penetrated field. (h) The annular regions of the field penetration have merged and the regions of field expulsion is no longer connected. It is interesting to see the same quantitative trend as observed in our DFI measurements for the steel plate at 45°, especially the penetration of the field, as well as the two diamond shaped areas at the top and bottom corner.
Chapter 5

Small-angle neutron scattering (SANS) experiments

The experimental SANS measurements reported in this chapter serve as a comparison and verification of the DFI results of the magnetization processes of the single and polycrystalline materials from the previous chapter. In section 5.1, the experimental SANS setup is shown. The SANS data of the magnetization process of the single-crystalline (100)-oriented FeSi disc and the comparison to the DFI magnetization observations are reported in section 5.2. Section 5.3 shows the SANS results of the magnetization process of the polycrystalline sample as well as the comparison with the obtained magnetization process DFI results. In particular, the steel plate was investigated for different orientations with respect to the external magnetic field.

5.1 Experimental setup

SANS is an ideal tool for studying the structure of materials in the size range from few nm to microns. Imaging methods such as transmission electron microscopy (TEM) also have this capability, but SANS is a non-destructive method. It provides structural information averaged over all magnetic domains of different sizes. For instance, in our case, there is a domain wall distribution in a sample, which is described by SANS as an average measure and there is no need, nor even the possibility to visualize each individual domain. In contrast to our DFI results of the magnetization process of the single-crystalline FeSi disc and the polycrystalline steel plate, which deliver information of the scattering signal in real-space, SANS delivers an averaged signal in reciprocal-space. However, due to the probing length scale of the SANS technique, information of the structural arrangement, namely the preferred orientations of the magnetic domains walls, can be obtained.

The SANS experiments were carried out at the SANS I instrument at SINQ at PSI [Koh2000]. The schematic layout and a photograph of the SANS instrument are shown in Fig. 5.1. The instrument is located behind a curved neutron guide, which provides
Figure 5.1: Schematical layout of the SANS I instrument at PSI (above) and a photograph (below). The wavelength was chosen to $\lambda = 18$ Å with the use of a velocity selector which provides a bandwidth of $\Delta \lambda / \lambda \simeq 10\%$. Several apertures are used to collimate the beam onto the sample. This is located at the experimental place surrounded by a variable external horizontal magnetic field, placed in the same way as for the DFI experiments in chapter 4. The SANS detector is located 20 m downstream of the sample.

A cold neutron beam. A wavelength of $\lambda = 18$ Å with a bandwidth of $\Delta \lambda / \lambda \simeq 15\%$ was used.

The sample is mounted in the same electromagnet as that used for the DFI experiments (compare Fig. 4.2(b)). For the SANS measurements, the phase and analyzer gratings were removed, and the imaging detector was replaced by the SANS detector, which is an 2D $^3$He-detector with $128 \times 128$ elements of $7.5 \times 7.5$ mm$^2$, located at the maximum distance of 20 m downstream of the experimental setup. The size of the beam at the sample position was 8 mm in diameter, defined by a 8 mm diameter cadmium aperture glued on the back of the cap of each sample holder (Fig. 4.3(b),(c)),...
limiting the measurements to the central part of the samples.

5.2 Magnetization process of a single-crystalline sample

To validate the interpretation of our DFI results of the magnetization process of the (110)-oriented FeSi disc, we conducted the corresponding SANS measurements, the results of which are shown in subsection 5.2.1. The orientation of the disc and the direction of the external magnetic field were kept identical to the former DFI experiments, to facilitate a direct comparison of the SANS results with the neutron imaging results. The comparison is shown in subsection 5.2.2.

5.2.1 SANS results

We intend to show that the mechanism of multiple refraction of unpolarized neutrons at domain walls in the specimen, which we claimed to be the contrast origin in our DFI observations, is indeed supported by the SANS measurements.

The SANS signal for the (100)-oriented FeSi disc was measured with the same ramping of the external magnetic field as used for the interferometry experiments (0 mT ≤ H ≤ 60 mT). The SANS results for increasing the external magnetic field from $H = 0$ mT to $H = 60$ mT in steps of 6 mT are shown in Fig. 5.2.

Due to the scattering of unpolarized neutrons at the domain walls in the FeSi disc, the diffraction pattern is broadened in the absence of a magnetic field. The typical structure sizes which are investigated by SANS measurements as mentioned are ranging from a few nm to microns. This leads to a SANS scattering pattern with information distributed over the whole detector area, as e.g. observed for the steel plate in Fig.5.7, Fig.5.10. For the FeSi disc the detector image can be regarded as only a broadening of the original beam. Note that each detector image displays only a quarter of the whole detector area. The full detector image (compare Fig.5.7) has an axis scaling in units of the scattering angle $\theta$ ranging from $-20$ mrad ≤ $\theta_{x,y}$ ≤ $20$ mrad. The broadening is related to the fact that the scattering structures (domain walls), which are distributed in dimensions of several microns (compare the domain width in Fig. 4.4(d)), are too large. Note the detector image delivers information in reciprocal space: Small structures → large $\theta$, large structures → small $\theta$. The demagnetized sample is in a complicated multi-domain configuration, as seen in the corresponding MOKE image in Fig.4.4(d). Furthermore, we observe an asymmetry in the SANS pattern at zero field that remains until the external field value reaches $H = 24$ mT, beyond which the asymmetry starts to collapse. A pronounced transition from the quadratic asymmetry to the circular profile is observed at $H = 30$ mT. Further increasing the field maintains the original circular beam profile.
Figure 5.2: SANS results of the (100)–oriented FeSi single-crystalline disc for the \( H \) field ramp (horizontal) from 0 mT to 60 mT. In zero field configuration \( H = 0 \) mT a quadratic asymmetry in the scattering pattern is observed. A broadening of the beam along the vertical and horizontal direction is visible, indicating a dominance of the corresponding domain wall orientations. A collapse of the quadratic SANS pattern asymmetry for \( H = 24 \) mT to \( H = 30 \) mT is observed. With further increase of the magnetic field, the broadening disappears. The original circular beam profile is regained at \( H = 60 \) mT.
Figure 5.3: SANS results of the (100)–oriented FeSi single-crystalline disc. (a), (b) Scattering pattern (logarithmic scale) without and with an external magnetic field, respectively. (c) Profiles through the scattering patterns as a function of the scattering angle $2\theta$.

Figure 5.4: SANS results, same data as shown in Fig. 5.3, but displayed on a linear scale (normalized to maximum intensity). (a), (b) Scattering pattern without and with an external magnetic field, respectively. (c) Quantitatively analyzed profiles with the corresponding $\sigma$-value obtained from a Gauss fit.
The analysis of the SANS measurements is shown in Fig. 5.3 [Gru2008b]. The observed asymmetry in the SANS pattern at zero field is again visible in Fig. 5.3(a). The broadening of the beam along the vertical and horizontal direction indicates the dominance of a perpendicularly oriented domain wall distribution in the sample. This is typical for a (100)-oriented material with its two easy axes, as seen in the Laue image in Fig. 4.5. We rotated the sample by an angle of 45° with respect to these easy axes. The magnetic hard axes are therefore aligned horizontally and vertically with respect to the magnetic field. As the magnetic hard axes coincide with the domain wall directions, the domain walls are predominantly distributed in the horizontal and vertical directions, leading to the scattering pattern seen in Fig. 5.3(a). When the magnetic field is increased to $H = 60 \text{ mT}$, the beam broadening disappears and a circular beam profile, corresponding to the original instrument resolution, is obtained, see Fig. 5.3(b). Fig. 5.3(c) shows profiles through the scattering patterns, with and without the application of an external magnetic field, as a function of the scattering angle $2\theta$.

A quantitative analysis of the beam broadening is shown in Fig. 5.4. Here the same data as that shown in Fig. 5.3 are presented, but on a linear scale. Fig. 5.4(a) shows the scattering pattern without an external magnetic field, normalized to maximum intensity. Fig. 5.4(b) shows the scattering pattern with an external magnetic field of $H = 60 \text{ mT}$, again normalized to maximum intensity. Fig. 5.4(c) shows section profiles along horizontal, vertical and diagonal directions through the small-angle scattering patterns, with and without an externally applied magnetic field. The instrument resolution is $\sigma_0 = 0.69 \pm 0.02 \text{ mrad}$, determined from a Gaussian fit of the data. The analysis of the demagnetized sample leads to a broadening of the width in horizontal and vertical directions to $\sigma_1 = 1.18 \pm 0.02 \text{ mrad}$ (average value). The net increase of vertical and horizontal beam broadening due to the sample amounts to $\Delta \sigma_{HV} = \sqrt{(\sigma_1^2 - \sigma_0^2)} = 0.95 \pm 0.02 \text{ mrad}$. Along the diagonal directions, the net increase in the width of the profiles caused by the demagnetized sample is only $\Delta \sigma_{DIA} = 0.11 \pm 0.02 \text{ mrad}$.

The values of $\Delta \sigma_{HV}$ and $\Delta \sigma_{DIA}$ agree with those reported for similar broadening measurements in the literature [Men1997]. Moreover, these observations indicate that the domain walls are predominantly ordered in horizontal and vertical directions. If this were not true, then we would observe a significant broadening in the diagonal profiles of the SANS pattern. For the fully magnetized sample, the net increase in the width is small, i.e., below the error margin in both vertical and horizontal direction.

A more detailed discussion of the observed values for the beam broadening in the SANS experiments and their link to the DFI results is discussed in the following subsection.
5.2.2 Comparison of SANS results with neutron imaging results

The experimentally observed values of the beam broadening in the SANS measurements can be linked to the angular sensitivity of the grating interferometer and the loss of contrast in the DFI. Based on the simple picture of small-angle scattered neutrons leading to a decrease in the visibility of the interference pattern, we can estimate the range of scattering angles that have the strongest effect on the DFI contrast. Accordingly, the interference pattern produced by $G_1$ in the plane of $G_2$ is obviously most strongly affected by neutrons that are scattered by an angle defined by the ratio of half the period of $G_1$ to the distance between the $G_2$ and $G_2$. In our case, for $p_2 = 4.00 \, \mu m$ and $d = 19.4 \, mm$, this angle amounts to $\alpha = p_2/2d \approx 0.1 \, mrad$. Due to the periodicity of $G_2$, the angular range that affects the DFI also includes all angles which are larger than $\alpha$. This mrad angular range of the DFI measurements is also probed by the SANS measurements. Here the angular range lies between $-20 \, mrad \leq \theta_{x,y} \leq 20 \, mrad$.

The strong link between the SANS and the DFI results can also be qualitatively observed in Fig. 5.5. Both the orientation and external applied field for the SANS and DFI measurements are identical. In both the SANS and DFI measurements, a rapid change on going from $H = 18 \, mT$ to $H = 30 \, mT$ is observed. For the SANS data, the measured asymmetric image collapses into a circular symmetric one. In the DFI image, a vertically aligned mono domain is observed in the middle of the disc. Note that the probing beam size for the SANS measurements was 8 mm in diameter and so for $H > 30 \, mT$ the domain wall rich areas at the edges of the disc are not probed.

A more quantitative analysis is shown in Fig. 5.6, where the measured beam broadening ($\sigma$-value of a Gaussian fit) in the SANS experiments and the averaged DFI signal as a function of the externally applied magnetic field are compared. With this Figure we directly compare and link DFI contrast with SANS results obtained for the (100)-oriented FeSi disc as a function of the external magnetic field. The integrated DFI signal is obtained by averaging each DFI image values over a central $7.5 \times 7.5 \, mm^2$ region of the disc (total diameter: 12 mm), while the SANS values represent the width ($\sigma$-value) of the Gaussian fit to the corresponding SANS patterns. Again, we note that the beam size used for the SANS measurements was 8 mm and thus probes approximately the same region as the integrated DFI signal. For both the DFI signal and the SANS signal, the results for the cases of magnetic field ramped in the positive ($-60 \, mT \rightarrow 60 \, mT$) and the reversed direction were plotted. No measurable remanence in the material was observed, since the corresponding two curves lie on top of each other. When the sample is fully magnetized, at $|H| = 60 \, mT$, no scattering is observed in the SANS patterns and the $\sigma$-value for the width of the SANS pattern reduces to the instrument’s resolution of $\sigma_0 = 0.69 \pm 0.02 \, mrad$. This behavior is also reflected in the DFI signal that is close to unity because no scattering of neutrons at domain walls
Figure 5.5: Comparison of the SANS results with the imaging results of the DFI for a $H$ field ramp from $H = 0 \text{ mT}$ to $H = 60 \text{ mT}$. The change from $H = 18 \text{ mT}$ to $H = 30 \text{ mT}$ is visible for both the SANS results (collapse of the scattering pattern) and the DFI results (growth of a mono domain). The dashed circle in the DFI for $H = 0 \text{ mT}$ represents the probing area for the SANS measurements.
is observed. Between $18 \text{ mT} \leq |H| \leq 30 \text{ mT}$, rapid changes in both the DFI and the SANS signal are observed. These changes are due to the transition of the specimen from a largely mono domain structure to a rich multi domain configuration. In the multi domain configuration, a strong scattering signal is observed for $0 \text{ mT} \leq |H| \leq 24 \text{ mT}$. This is reflected by the DFI value, which decreases to a stable minimum for the small magnetic field values, essentially determined by the measurement statistics.

Based on this comparison, we conclude that dark-field imaging is directly linked to the small-angle neutron scattering images and provides the local scattering cross section of the specimen on a pixel by pixel basis. Therefore, the DFI is real space scattering images. It can be seen from Fig. 5.6 that for field values between $0 \text{ mT} \leq |H| \leq 24 \text{ mT}$ the DFI signal shows no changes and remains constant, whereas the SANS signal still shows an increasing and decreasing signal behavior with its maximum at $H = 0 \text{ mT}$, however, this is not yet fully understood.
5.3 Magnetization processes of a poly-crystalline sample

To validate the interpretation of our DFI results of the magnetization process of the poly-crystalline steel plate, we also conducted the corresponding SANS measurements, which are shown in subsection 5.3.1. The orientations of the steel plate and the direction of the external magnetic field were kept identical to the former DFI experiments\(^1\). The direct comparison of the SANS results with the DFI results for the 0° and 45° case are made in subsection 5.3.2.

5.3.1 SANS results

The SANS signal for the steel plate in 0° orientation was measured with the same ramping of the external magnetic field as for the DFI. The quantitative SANS results for the increasing external magnetic field from \(H = 0\) mT to \(H = 250\) mT in 20 steps are shown in Fig. 5.7. Due to the scattering of neutrons at the domain walls in the steel plate, a wide symmetric circular diffraction pattern is observed when no magnetic field is applied because the sample is in a complex multi-domain configuration state. Here the full detector area is shown in contrast to the SANS measurements for the single-crystalline material. The diffraction pattern starts to slowly collapse at field values of \(13\) mT \(\leq |H| \leq 75\) mT. A dramatic transition of the diffraction pattern is made at field values of \(88\) mT \(\leq |H| \leq 113\) mT, where the wide spread diffraction pattern at \(H = 88\) mT reduces to well defined circular diffraction pattern at \(H = 113\) mT. Further increasing the external field recovers the original circular beam profile.

An analysis of the SANS measurements is shown in Fig. 5.8. The strong scattering behavior of the observed SANS pattern at zero field is again visible in Fig. 5.8(a). The circular scattering pattern indicates a random orientation of domain walls in the sample without preferred directions. This is expected from a poly-crystalline material and other examples can be found in the literature [All1982, Men1997]. When the magnetic field is set \(H = 250\) mT, a beam profile corresponding to the original instrumental resolution is regained in Fig. 5.7(b). Fig. 5.8(c) displays profiles through the scattering patterns with and without the external magnetic field, as a function of the scattering angle \(2\theta\), obtained by radial averaging of each SANS diffraction pattern.

A qualitative analysis of the scattering is shown in Fig. 5.9, where the same data as in Fig. 5.8 are displayed, but on a linear scale. Fig. 5.8(a) shows the scattering pattern without an external magnetic field, normalized to maximum intensity. Fig. 5.8(b) shows the scattering pattern with an external magnetic field of \(H = 250\) mT. For the normalized data, isotropic scattering, as well as large scattering angles compared to the original beam profile, are observed. Fig. 5.8(c) shows section profiles obtained by radial

\(^1\)The SANS results for the 22.5° orientation of the steel plate are missing, since the DFI results have been measured after the SANS experiments.
5.3. Magnetization processes of a poly-crystalline sample

Figure 5.7: SANS results of the magnetization process ($0 \, \text{mT} \leq H \leq 250 \, \text{mT}$) for $0^\circ$ orientation of the steel plate. The isotropic scattering pattern indicating a random domain wall orientation is visible. The collapse of the SANS pattern is seen between $H = 88 \, \text{mT}$ and $H = 113 \, \text{mT}$. For further increase of the magnetic field the scattering disappears and the original circular beam profile is finally obtained.
Figure 5.8: SANS results of the steel plate at 0°. (a) Scattering pattern (logarithmic scale) without external magnetic field. The isotropic scattering, indicating a random domain wall orientation, is clearly visible. (b) Scattering pattern with magnetic field. (c) Profiles through the scattering patterns as a function of the scattering angle 2θ.

Figure 5.9: SANS results, same data as shown in Fig. 5.8 but displayed on a linear scale (normalized to maximum intensity). (a), (b) Scattering pattern without and with an external magnetic field, respectively. (c) Quantitatively analyzed profiles with the corresponding σ-value obtained by a Gauss fit.
5.3. Magnetization processes of a poly-crystalline sample

averaging of the small-angle scattering patterns, with and without magnetic field. The same instrument resolution, of \( \sigma_0 = 0.69 \pm 0.02 \text{ mrad} \), as that obtained in the last section is reproduced. Performing a radial averaging analysis with the demagnetized steel plate data gives \( \sigma_1 = 7.88 \pm 0.02 \text{ mrad} \). Thus the net increase of the scattering due to the sample alone amounts to \( \Delta \sigma = \sqrt{(\sigma_1^2 - \sigma_0^2)} = 7.19 \pm 0.02 \text{ mrad} \). For the magnetized sample at \( H = 250 \text{ mT} \), the net increase in the width is small, i.e., below the error margin and is consequently the same as the instrumental resolution.

The SANS results for the steel plate in 45° orientation are shown in Fig. 5.10. The measured SANS detector pattern for the 45° case resembles that for the 0° orientation (compare Fig. 5.7), so that they could not be distinguished within the SANS measurements. This can be explained due to the random oriented domain walls in the sample. We again observe that the diffraction pattern starts to slowly collapse at the same field values and that the dramatic transition is in the same field interval. Further increasing of the external field again delivers the original beam profile as for the 0° case.

An analysis of the SANS data for the 45° case is shown in Fig. 5.11. The analysis is done in the same way as for the 0° case. The widespread circular scattering pattern at zero field is shown in Fig. 5.11(a) and the scattering pattern for \( H = 250 \text{ mT} \) in Fig. 5.11(b). The profiles through both patterns are shown in Fig. 5.11(c).

The qualitative analysis is shown in Fig. 5.12. It displays the same data as shown in Fig. 5.11 but on a linear scale. The instrumental resolution is determined to \( \sigma_0 = 0.69 \pm 0.02 \text{ mrad} \). The radial averaging analysis for the demagnetized steel plate leads to a scattering of \( \sigma_1 = 8.08 \pm 0.02 \text{ mrad} \). The net increase of the scattering of the beam due to the sample alone thus amounts to \( \Delta \sigma = \sqrt{(\sigma_1^2 - \sigma_0^2)} = 7.39 \pm 0.02 \text{ mrad} \).

A more detailed discussion of the observed values for the scattering in the SANS experiments and their link to the DFI results for both orientations of the steel plate is discussed in the following subsection.
Figure 5.10: SANS results of the magnetization process ($0 \text{ mT} \leq H \leq 250 \text{ mT}$) for $45^\circ$ orientation of the steel plate. The isotropic scattering indicating a random domain wall orientation is visible, as well as the collapse of the SANS pattern between $H = 88 \text{ mT}$ to $H = 113 \text{ mT}$. For further increase of the magnetic field the scattering disappears and the original circular beam profile is finally regained at $H = 250 \text{ mT}$. 
Figure 5.11: SANS results of the steel plate at 45°. (a), Scattering pattern (logarithmic scale) without external magnetic field. The isotropic scattering is visible, indicating a random domain wall orientation. (b) Scattering pattern with magnetic field. (c) Profiles through the scattering patterns as a function of the scattering angle $2\theta$.

Figure 5.12: SANS results, same data as shown in Fig. 5.11 but displayed on a linear scale (normalized to maximum intensity). (a), (b) Scattering pattern without and with an external magnetic field, respectively. (c) Quantitatively analyzed profiles with the corresponding $\sigma$-value, obtained by a Gauss fit.
5.3.2 Comparison of SANS results with neutron imaging results

Similar to what is reported for the FeSi single-crystalline disc in subsection 5.2.2, the strong link between SANS and DFI can also be quantitatively observed for the steel plate.

For the 0° orientation of the steel plate, the direct comparison between the SANS results and the DFI results are shown in Fig. 5.13. The rapid change in the SANS images at the field values between $H = 88$ mT to $H = 113$ mT could be also observed in the DFI image, where a vertical stripe in form of a domain wall free area starts to form. This expansion of this vertical stripe along the horizontal direction in the DFI explains the collapse of the SANS detector pattern.

For the 45° orientation of the steel plate, the direct comparison between the SANS results and the DFI ones are shown in in Fig. 5.14. Here the rapid change in the DFI results is seen at field values of $H = 100$ mT to $H = 113$ mT, where two oval vertical mirror-symmetric domain wall free areas start to form. Regarding the SANS data, the remarkable change is still present at the field values between $H = 88$ mT to $H = 113$ mT, where the diffraction patterns start to collapse, similar to 0° case. There are no remarkable differences to observe between the SANS imaging results for the 0° and 45° orientation.
Figure 5.13: Comparison of the SANS results with the DFI results for H field ramp (0 mT to 250 mT) of the steel plate at $0^\circ$. The change from $H = 88$ mT to $H = 125$ mT is visible for both the SANS results (collapse of the scattering pattern) and DFI results (growth of domain wall free area). Again, the dashed circle illustrates the probing area for the SANS measurements.
Figure 5.14: Comparison of the SANS results with the DFI results for H field ramp of the steel plate at 45°. The change of the SANS pattern at $H = 88$ mT with the collapse is visible and is also seen in the DFI.
A more quantitative analysis is shown in Fig. 5.15, where a comparison of the measured scattering in the SANS experiments and the averaged DFI signal as a function of the externally applied magnetic field is shown. With this figure, we can again directly compare the DFI contrast with the SANS results for both orientations of the steel plate, just as we did for the FeSi single-crystalline disc. The displayed integrated DFI signal is obtained by averaging each DFI image over a central $7.5 \times 7.5 \text{ mm}^2$ region of the steel plate (edge length: 15 mm). The SANS value represents the width of the Gaussian fit, obtained by radial averaging of the corresponding SANS patterns. For both, the DFI signal and the SANS signal, we plotted the results for the $0^\circ$ and $45^\circ$ cases, when the magnetic field was ramped from $H = 0 \text{ mT}$ to $H = 250 \text{ mT}$.

In the multi domain configuration, for $0 \text{ mT} \leq H \leq 75(88) \text{ mT}$ (bracket values are for the $45^\circ$ case), a strong and slowly decreasing scattering signal is observed. This is reflected in the DFI values, which decrease to a stable minimum. Between $75(88) \text{ mT} \leq H \leq 125(138) \text{ mT}$, rapid changes in both the DFI and the SANS signal can be observed. These changes are due to the transition of the sample from a largely demagnetized configuration to a magnetized one. The separation of both curves indicates that the magnetization processes are different for each orientation. For the $0^\circ$ case, the curves of both the SANS and DFI show an earlier and faster transition than that of the $45^\circ$ case. In other words, the magnetization process for the steel plate in $45^\circ$ orientation needs a higher external field to magnetize and is therefore more inert. When the sample is magnetized in the probing area, at $113(125) \text{ mT} \leq H \leq 250 \text{ mT}$, no scattering is observed in the SANS patterns and the $\sigma$-value for the width of the SANS pattern reduces to the instrument resolution. This is also reflected in the DFI signal, which is close to unity, since the probing area is free of domain walls for both cases.

The probing size of the neutron beam for the SANS measurements was 8 mm in diameter defined by the Cd aperture. Consequently, if the area of the domain wall free state is larger than 8 mm in diameter, there is no longer any contributing to the SANS signal. This can be seen for the $0^\circ$ case for $H = 125 \text{ mT}$, where the corresponding $\sigma$-values stay constant on increasing external magnetic field. Due to the fact that the domain wall free vertical stripe already has a width larger than 8 mm, e.g., the outer parts (yellow stripes) no longer contribute to the SANS signal and the $\sigma$-values stay constant for increasing external magnetic field. Regarding the $45^\circ$ case, the stable minimum for the $\sigma$-value is achieved at $H = 138 \text{ mT}$ since it can be seen from the corresponding DFI that a domain wall free area is not formed until this field value.

However, it can be again seen from Fig. 5.15 that for field values between $0 \text{ mT} \leq H \leq 88 \text{ mT}$, the DFI signal remains constant, whereas the SANS signal still shows a decreasing signal behavior with its maximum at $H = 0 \text{ mT}$. Therefore the dynamic range of the DFI for the field values of $0 \text{ mT} \leq H \leq 88 \text{ mT}$ is limited compared to the SANS signal, as already seen for the FeSi disc in Fig. 5.6.
Figure 5.15: Comparison of the integrated DFI signal (blue lines) and the SANS measurement (red line) for the steel plate at $0^\circ$ (squared markers) and $45^\circ$ (diamond markers) as a function of the external magnetic field from $0 \text{ mT} \leq H \leq 250 \text{ mT}$. The averaged DFI values are obtained by averaging over image values in a central region of the steel plate. The SANS values represent the width ($\sigma$-value) of corresponding Gaussian fits.
Chapter 6

Interferometry experiments II: Magnetic domain structures

In this chapter it is reported how the neutron grating interferometer setup yields projection images of the internal domain structure in a bulk ferromagnetic FeSi single-crystalline sample. In section 6.1 the experimental setup is presented. In section 6.2, the DFI results of the basic domains of (110)—oriented sheets in the FeSi sample are presented. The domain structures from the neutron results are compared with surface sensitive magneto-optical Kerr effect micrographs. The MOKE images are shown in section 6.3 and compare favorably with the DFI results.

6.1 Experimental setup

The approach to visualize bulk magnetic domain structures is based on a grating interferometer setup, implemented with polychromatic neutrons [Gru2008c]. In Fig. 6.1(a) a schematic of the experimental setup is shown. It consists of a source grating \( G_0 \), a phase grating \( G_1 \) and an analyzer attenuation grating \( G_2 \). This experimental setup for imaging bulk magnetic domain structures is the same as that used for imaging bulk magnetization processes as described in chapter 4. The only difference is that no magnetic field is needed for the experiment. The removed pole pairs of the electromagnet are seen in Fig. 6.2(b). The sample holder is now seen through the insertion holes of the pole pairs.

The image contrast is again based on neutrons, which are scattered at domain walls of the sample as illustrated in Fig. 6.1(b). The contrast is analogous to that obtained for the bulk magnetization process observations, namely to the DFI contrast. Since the detector resolution is of the order of 100 \( \mu m \) [Gru2006], a sample with larger domain sizes allows us to image bulk magnetic domain structures as reported in the following.

The sample is a (110)-oriented FeSi single-crystalline disc with a thickness of 300 \( \mu m \) as shown in Fig. 6.2(a). It was cut from a Goss-oriented transformer steel with a diameter of 10 mm. The sample is by courtesy of Rudolf Schäfer, IFW Dresden. For
Figure 6.1: Neutron grating interferometer setup to visualize bulk magnetic domain structures. (a) Setup showing the source grating $G_0$, the phase grating $G_1$ at a distance $l = 5.23 \text{ m}$ and the analyzer absorption grating $G_2$. The setup is similar to the one used to visualize bulk magnetization processes in chapter 4, however without an external magnetic field. (b) Neutrons scattered at magnetic domain walls in the specimen locally degrade the interference pattern in the plane of $G_2$. 
6.1. Experimental setup

Figure 6.2: (a) Sample holder made of an aluminium rod with an aluminium plate as a cap. The (110)-oriented FeSi single-crystalline disc with a diameter of 10 mm is mounted on the cap with three plastic screws. (b) Photograph of the experimental setup. The sample holder is mounted horizontally and brought as close as possible in front of the analyzer grating $G_2$. Through the removed pole shoe of the magnet (compare Fig. 4.2) the sample holder could be seen. The holder is additionally mounted on a rotation stage to measure the disc for different rotation angles $\theta$ (see Fig. 6.1(a)).

placement the disc is mounted with three plastic screws on a cap on the aluminium rod, which serves as the sample holder, shown in Fig. 6.2(a). The holder is mounted in the setup as seen in Fig. 6.2(b).

To guarantee maximum possible spatial resolution, the disc is brought as close as possible in front of the analyzer grating $G_2$. The geometrical resolution of the setup is defined by the diameter of the source, the distance from the source to the sample and the sample to detector distance. The source is defined by the used aperture with diameter $c = 10$ mm (Fig. 6.1(b)). The source to sample distance is slightly larger than $l = 5.23$ m. The sample to detector distance is at least $d = 19.4$ mm as seen in Fig. 6.1(b). The geometrical resolution $r > \frac{d \lambda}{l f}$ is then about 50 $\mu$m. The sample holder can be seen in Fig. 6.2(b) through the insertion hole of the cylindrical pole shoe of the magnet. The sample holder is mounted on a rotation stage to allow for DFIs at different rotation angles $\theta$ of the disc, with respect to the axis along the beam, as schematically shown in Fig. 6.1(a).

For the DFI processing we took 32 raw images over two periods with 300 seconds exposure time for each of the data and flat field image stacks. Additionally, five dark field frames with 300 seconds were taken. In the following section we discuss the internal domain structure of the sample in the DFIs.
6.2 Neutron imaging results

The experimental results of bulk magnetic domain wall imaging for the (110)-oriented FeSi disc are shown in Fig. 6.3. The conventional transmission image (TI) is shown in Fig. 6.3(a). Only the strongly attenuating plastic screws used to mount the disc are visible in the TI. The ferromagnetic disc is clearly visible in the DFI images in Figs. 6.3(b)−(d).

In Fig. 6.3(b), for the $\theta = 0^\circ$ orientation, the sample was aligned with the [001] axis parallel to the grating lines. Vertically oriented, elongated domain walls of several millimeters in length and separated by several hundred micron wide domains are clearly visible in the DFI contrast. The observed domain walls are associated with the basic domains of (110)-oriented FeSi sheets, which are magnetized along the surface-parallel

![Figure 6.3](image-url)

Figure 6.3: Neutron imaging results for the (110)-oriented FeSi single-crystalline disc. (a) Conventional neutron transmission image (TI). (b-d) Dark-field images (DFIs) showing a projection of the internal domain wall structure of the sample for several sample orientations. 16 images over two grating periods with 10 minutes exposure time for each raw image were taken to process out one DFI.
easy [001]-axis, separated by 180° domain walls [Hub1998]. This is schematically shown in Fig. 6.5(b). We observe that the width of the domains, as measured by the distance between two domain walls, decreases along the horizontal direction with increasing distance from the center of the disc. Interestingly, the area where these domains are found forms a rhombohedron in the center of the specimen. Outside of this rhombohedral area, only very low contrast in the DFI is observed in Fig. 6.3(b). This picture changes dramatically when the sample is rotated by $\theta = 45^\circ$ as seen in Fig. 6.3(c), or by $\theta = 90^\circ$ as seen in Fig. 6.3(d). The fact that the contrast outside the rhombohedral area increases for increasing values of $\theta$ and reaches its maximum for $\theta = 90^\circ$ indicates that the domains outside the rhombohedral area are oriented perpendicular to those inside the area.

The domains surrounding the rhombohedral area could not be resolved individually. The measured contrast is equal to that obtained for the (100)-oriented FeSi single crystal disc in section 4.2 and can be considered as an image of the domain wall density distribution. In the following section we will compare the neutron results with MOKE results. We will find that the surrounding area contains domains whose sizes are indeed smaller than the detector resolution and could therefore not be individually resolved.

It is interesting to note that the basic domain walls through the FeSi disc, as theoretically predicted, should be oriented along the [010] or [100] direction, as depicted in Fig. 6.5(b). The domain walls are therefore inclined by an angle of 45° with respect to the sample surface. In our case the domain widths that are in the order of several hundred microns are in the same order as the thickness of the disc. Therefore, regarding a projection image, the domain wall will span diagonally through the domain. If this is the case, then the individual domain walls, as imaged in the DFI, should not be seen and complete rhombohedral area should appear with low contrast values as observed for the surrounding area.

### 6.3 Comparison with magneto optical Kerr effect measurements

To interpret the observed DFI results, which are projection images of the internal bulk magnetic domain structure, we recorded surface domain images of the sample using a MOKE microscope. To obtain an equally large field of view, 40 single Kerr micrographs had to be combined into one image. Fig. 6.4(a) shows the resulting MOKE image. Fig. 6.4(b) shows a schematic of the MOKE image. The rhombohedron containing the inner domain structure is readily identified, as well as the decreasing width of the domains as a function of increasing distance from the center of the disc. Note that the DFI shows a frequency doubled image compared to the MOKE image because it shows not the domains but the domain walls. Moreover, the MOKE image
shows fine horizontally aligned domain structures in the periphery of the rhombohedral area. These horizontal domains are closure domains of internal basic domains as shown in Fig. 6.5(c),(d). Such patterns are caused by compressive mechanical stress, which is obviously dominating in the outer sample parts in a symmetric fashion. We note that the fine horizontal domain structure at the outer part of the rhombohedron does not contribute to the DFI shown in Fig. 6.3(b) ($\theta = 0^\circ$) because these domains cause scattering only in a direction parallel to the grating lines. For the same reason, the domains in the inner part of the rhombohedron do not contribute to the DFI in Fig. 6.3(d) ($\theta = 90^\circ$). For the $\theta = 45^\circ$ case, shown in Fig. 6.3(c), both the outer and the inner domain walls contribute to the DFI signal.

It was a fortunate coincidence to obtain such a sample. It delivered nice didactic results, illustrating the resolution limit as well as the angle sensitivity with respect to the grating lines of the method.

Figure 6.4: Magneto-optical Kerr effect image of the (110)-oriented FeSi single-crystalline disc. (a) Magneto optical Kerr image of the surface domain structure. (b) Schematic drawing of the obtained surface domain structure.
Figure 6.5: Domain stress patterns produced by compressive stresses along the preferred axis of the (110)-oriented single-crystalline FeSi disc [Hub1998]. (a) Schematic drawing of the obtained surface domain structure (see Fig. 6.4). (b) Unstressed state (blue box) consisting of slab-like domains magnetized parallel and antiparallel to the easy [001] direction (see Fig. 6.3(b),(c)). (c) Moderate stress leading to the so called stress pattern I (red box), containing superficial $180^\circ$ walls between the basic domains as shown in (d). (d) Cross section through the domain pattern along the green line in (c) depicting surface and basic domain structure. The arrows indicate the magnetization directions, $180^\circ$ for the superficial domains and $90^\circ$ for the basic domains.
6.4 Sensitivity of the grating interferometer

Experimental results from a Fe 12.8 at% Si single crystal wedge with a (111) principal surface illustrate the sensitivity and the limitations of this method in the sense of imaging bulk magnetic domain structures. This sample is courtesy of R. Schäfer, IFW Dresden. A schematic of the wedge is shown in Fig. 6.6(a), where it is illustrated that increasing wedge thickness results in increasing domain width. In reality, the domains in the interior are not oriented perpendicular to the wedge surface as indicated in Fig. 6.6(a), but are oriented along the <100>-axis [Sch2000a].

A fine superficial domain structure is present over the whole wedge surface. A section of the domain structure imaged with the MOKE microscope is shown in Fig. 6.6(b). This MOKE image is recorded at a wedge thickness of 200 \( \mu \text{m} \). The MOKE image is courtesy of R. Schäfer, IFW Dresden. The conventional neutron transmission image of the wedge is shown in Fig. 6.6(c) with the wedge imaged along the z-axis. The corresponding neutron dark-field image is presented in Fig. 6.6(d). The wedge is much more clearly rendered in the DFI than in the transmission image, where it is difficult to detect the wedge to the full extent. It is more clearly seen in the corresponding section profiles through the TI and DFI, as shown in Fig. 6.6(e). It is seen that the contrast in the DFI is homogeneously reduced along the whole wedge. For the TI section the expected linear attenuation behavior is seen. The comparison of the DFI with the TI illustrates the sensitivity of the neutron grating interferometer setup. The left edge of the wedge is fully visible in the DFI, although the wedge thickness is small and so too is the surface domain volume. However, the number of domain walls in this volume is large enough to destroy the contrast in the corresponding DFI. Associated with the high sensitivity of the method is the implied disadvantage that investigations of bulk materials are limited by the presence of a superficial domain structure.
Figure 6.6: Illustration of the grating interferometer sensitivity. The sample is a Fe12.8 at.%Si single crystal wedge. (a) Drawing which illustrates that for increasing wedge thickness the domain width of the volume domains inside the material should increase. (b) MOKE image obtained at the sample surface at a wedge thickness of 200 µm. A fine domain structure at the sample surface is clearly visible. (c) Transmission image (TI) of the wedge along the z-axis. (d) Neutron dark-field image (DFI) of the wedge. The fine surface domain structure completely degrades the coherence of the neutron wave front over the whole wedge. (e) Section profiles through the TI and DFI.
In this thesis it has been shown how a grating based neutron interferometer can provide spatially resolved images of the internal magnetic domain structure. Imaging of individual magnetic domains in bulk materials was investigated in a single-crystalline FeSi disc. Projection images of the internal domain wall structure formed by the basic domains of the FeSi sheets were observed. Magnetic domains 100 $\mu$m wide have been resolved. The DFI results of the internal domain structure are successfully verified by comparing them to surface sensitive magneto-optical Kerr effect images. Additionally, it was demonstrated that this technique has the potential to image magnetization processes in centimeter-sized opaque ferromagnetic specimens. For samples with domain sizes smaller than the resolution of the imaging detector, the DFI of bulk ferromagnetic samples can be interpreted as a projection image of the domain wall density distribution in the sample. We used this information to image bulk magnetization processes of single-crystalline and poly-crystalline materials. Complementary small-angle neutron scattering (SANS) experiments validate the interpretation that the loss of spatial coherence revealed in the dark-field image (DFI) is caused by the refraction of unpolarized neutrons at magnetic domain walls in the specimen. SANS measurements of the magnetization processes underline the strong link between SANS and DFI results. For the SANS measurements the same magnetization behavior was observed as for the DFI results. The DFI magnetization data have been additionally supported by FEM simulations, which show an impressive accordance with the single-crystalline material results and qualitative accordance with the poly-crystalline results. The sensitivity of the grating interferometer was evaluated by the use of a single-crystalline FeSi wedge. The fine superficial domain structure of the wedge already provides a strong contrast signal, which on the other hand disallows the investigation of the underlying volume domains.

Based on the obtained results, one can conclude that bulk magnetic domain imaging is only possible if the sample exhibits no superficial domains in the form of closure domains. A more detailed investigation needs to be done to characterize the minimal domain wall volume for producing signal in the DFI. Tuning the sensitivity of the setup
by adjusting the wavelength and grating periods can also be considered. For the results of the individual resolved domain walls there is a discrepancy between the theoretically predicted angle value under which the basic domains are taking course through the sample and the experimental DFI results. This question could be answered by performing an experiment with a tomography approach, where the sample is investigated for different projection angles.

The greatest potential of the method lies in the possibility to image the penetration of an external magnetic field into arbitrary samples. Since our approach is highly efficient, because our setup is compatible with a broad angular and energy spectrum of the incident neutron beams, it opens the way for systematic tomographic investigations of the magnetic properties of bulk materials. We envisage that the method can be extended to 3D using computed tomography and yield 3D images of the magnetic domain wall distributions of bulk ferromagnetic materials of any geometry. Already the first 3D experimental data have been taken and the results are promising. Finally, implementing our approach with polarized neutrons has the potential to allow for structural characterization of the orientation and the magnitude of the local magnetization of an ensemble of magnetic domains in a bulk material.
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   "Design, fabrication, and characterization of diffraction gratings for neutron phase contrast imaging.

   "Multiple small angle neutron scattering: A new two-dimensional ultrasmall angle neutron scattering technique.

   "Highly absorbing gadolinium test device to characterize the performance of neutron imaging detector systems.
Publications as contributing author

   *Phase-contrast imaging and tomography at 60 keV using a conventional x-ray tube source.*

   *Neutron dark-field tomography.*

   *Hard-X-ray dark-field imaging using a grating interferometer.*

   *Phase contrast X-ray imaging of large samples using an incoherent laboratory source.*

   *Fabrication of diffraction gratings for hard X-ray phase contrast imaging.*

   *Grating interferometer based scanning setup for hard x-ray phase contrast imaging.*

   *Neutron phase imaging and tomography.*
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115

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