Stellingen bij het proefschrift

**Neutronen Depolarisatie in Materialen voor Magnetische Registratie**

1. De neutronen spin-echo techniek met een gedraaide precessie spoel geometrie\(^1\) maakt het met relatief goedkopere eenvoudige middelen mogelijk om bij een medium-flux reactor een kleine-hoek neutronenverstrooiingsopstelling te bouwen, waarvan de prestaties gelijkwaardig zijn aan de opstellingen bij hoge-flux reactoren.


2. Het gebruik van neutronenspinflippers opgebouwd uit twee adiabatische polarisatiedraaiers\(^1\) verdient de voorkeur boven Drabkin spinflippers\(^2\), omdat voor de eerstgenoemde flipper de correcties voor de spinflip-efficiency beter gedefinieerd en eenvoudiger uitvoerbaar zijn, en vanwege zijn geringe gevoeligheid voor stoorvelden.


\(^2\) T.J.L. Jones et al., Nucl. Instr. and Meth. 152 (1978) 463.

3. De neutronendepolarisatie techniek kan gebruikt worden om de geomagnetische geschiedenis "te lezen" aan de hand van de richting van de remanente magnetisatie van magnetietdeeltjes bevattend stollingsgesteente en aan de hand van de grootte en het kwadraat van de gemiddelde magnetisatie-oriëntatie van deze deeltjes.

4. Alle miljoenen guldens ten spijt die jaarlijks wereldwijd worden besteed aan het onderzoek aan hoge-dichtheid informatieopslag is de natuur reeds miljoenen jaren lang in staat om in het DNA miljoenen "bits" aan informatie voor zeer lange tijd te leggen in een volume waarin nu slechts een enkele bit opgeslagen kan worden.

5. Het neutronenbundel-onderzoek van magnetische materialen hoeft de concurrentie van krachtige synchrotronbronnen nog niet te vrezen, dit in tegenstelling tot de niet magnetische onderzoeksgebieden waarin het neutronenbundel-onderzoek traditioneel sterk vertegenwoordigd was.

6. Wanneer bij nieuwbouw van Nederlandse neutronenbronnen, versnellerinstituten en faciliteiten voor behandeling van tumoren, naar het voorbeeld van het Paul Scherrer Instituut in Zwitserland, de faciliteiten worden samengebracht op één locatie, zal een hoger rendement uit het totale onderzoeksbudget gehaald kunnen worden.

7. De huidige rooi premie-regeling voor fruitteelers, die geen strikte beperkingen voor nieuwe aanplant oplegt, leidt niet tot de gewenste structurele verlaging van de fruitproductie in Nederland. Velen zullen deze regeling dan ook zien en gebruiken als een welkome voortzetting van de afgeschafte WIR.
8. Het spreekwoord "beter een goede buur dan een verre vriend" spiegelt ons een verkeerd beeld van de werkelijkheid voor in tijden van watersnood wanneer het water letterlijk tot de lippen dreigt te komen.

P.T. Por
Oktober 1995
Theses going with the thesis

Neutron Depolarisation in Magnetic Recording Materials

1. The neutron spin-echo technique using a tilted precession coil geometry\(^1\) allows building at a medium flux reactor a small angle neutron scattering (SANS) set-up at relatively low cost and with simple means, and with a performance comparable with conventional SANS set-ups at high flux reactors.

\(^\dagger\) T. Keller et al., Neutron News Vol. 6 No. 3 (1995) 16.

2. A neutron spinflipper comprising two adiabatic polarisation rotators\(^\dagger\) is to be preferred rather than a Drabkin spinflipper\(^\dagger\), because for the former flipper the corrections for spinflip efficiency are better defined and easier realisable, and because of its negligible sensitivity to magnetic stray fields.


\(^\dagger\) T.J.L. Jones et al., Nucl. Instr. and Meth. 152 (1978) 463.

3. The neutron depolarisation technique can be used for "reading" the geomagnetic history recorded in igneous rocks containing magnetite particles from their size and mean-square magnetisation orientation, and from the remanent magnetisation of the rock.

4. Despite the millions of guilders spent annually on worldwide research on high-density recording, nature has been using for millions of years DNA to record millions of "bits" for many years in a volume that human technology needs for a few bits.

5. For the time being, neutron beam research of magnetic materials is not yet threatened by the competition of high brilliance synchrotron sources, contrary to the non-magnetic research areas in which neutron beam research traditionally had a great share.

6. When new Dutch neutron sources, accelerators and facilities for treatment of cancerous tumours, in analogy to the Paul Scherrer Institute in Switzerland, are located at one site, a higher efficiency of the combined research budgets will be realised.

7. The present allowance for Dutch fruit-growers to pull up fruit trees, which fails to dictate strict limitations to planting new trees, will not lead to the desired structural decrease of the fruit yield in the Netherlands. Many growers, therefore, will consider and use this allowance as a welcome prolongation of the abolished WIR (law of investment contribution).
8. The proverb "a near neighbour is better than a distant cousin" gives a false picture of reality in times of floods when water literally is about to rise to your lips.

P.T. Por

October 1995
Neutron Depolarisation in Magnetic Recording Materials

Neutronen Depolarisatie in Materialen voor Magnetische Registratie

P.T. Por

Interfacultair Reactor Instituut
Technische Universiteit Delft / Delft University of Technology
Oktober 1995
Cover: Drawing from Poulsen’s first patent for the telegraphone (Denmark No. 2653, year 1898). An iron piano string $l$, the medium, was used to store the information. The information was recorded and played using the recording head $m$.

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Por, P.T.

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Subject headings: neutron depolarisation/magnetic neutron scattering/ magnetic materials.

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Neutron Depolarisation in Magnetic Recording Materials

Proefschrift

ter verkrijging van de graad van doctor
aan de Technische Universiteit Delft,
op het gezag van de Rector Magnificus Prof. ir. K.F. Wakker,
in het openbaar te verdedigen ten overstaan van een commissie,
door het College van Dekanen aangewezen,
op maandag 23 oktober te 13:30 uur
door

Pieter Tjeerd POR

technisch natuurkundig ingenieur (HBO)
geboren te Zuid-Beijerland
Dit proefschrift is goedgekeurd door de promotoren:
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Prof. dr. R.W. Chantrell.

Dr. M.Th. Rekveldt heeft als begeleider in belangrijke mate aan het totstandkomen van het proefschrift bijgedragen.

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The research described in this thesis was performed within the Polarised Neutron Group of the department of Radiation Physics of the Interfacultair Reactor Instituut, Delft University of Technology, Mekelweg 15, 2629 JB Delft, The Netherlands. Financial support has been obtained from the Concerted Action on Magnetic Storage Technology (CAMST) a science program of the European Union.
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List of Symbols

\(a\)  
\(a, b\)  
\(b_d\)  
\(\mathbf{B}\)  
\(c\)  
\(2d\)  
\(D\)  
\(d\sigma/d\Omega\)  
\(d^2\sigma/d\Omega dE'\)  
\(\hat{D}\)  
\(E\)  
\(f_n\)  
\(F_N(\kappa)\)  
\(F_M(\kappa)\)  
\(F_p(s)\)  
\(h\)  
\(2h\)  
\(H_a\)  
\(H_c\)  
\(H\)  
\(\hbar\)  
\(i_x, i_y, i_z\)  
\(I_c, I_c^0\)  
\(I_1, I_0, I_{ij}\)  
\(I_s\)  
\(v\)  

distance between two exchanged coupled spins \([m]\); chapter 7  
coefficients of neutron spin-wave function; chapter 8  
nuclear scattering length \([m]\)  
magnetic induction \([T]\)  
depolarisation constant \(= 2.18 \times 10^{29} \ T^{-2} m^{-4}\)  
diameter of an elongated particle \([m]\)  
grain size \([m]\); chapter 7  
differential cross-section \([m^2 sr^{-1}]\)  
partial or double differential cross-section \([m^2 J^{-1} sr^{-1}]\)  
depolarisation matrix  
energy of neutron \([J], [eV]\)  
magnetic form factor  
unit-cell structure factor \([m]\)  
magnetic unit-cell structure factor \([m]\)  
particle form-factor  
thickness of magnetic layer \([m]\)  
length of an elongated particle \([m]\)  
magnetic field pulse amplitude \([A/m]\)  
coercive field \([A/m]\)  
magnetic field \([A/m]\)  
Planck's constant/\(2\pi\) \([Js]\)  
unit-vector along the x, y or z-direction  
DC-current through a magnetic field coil \([A]\)  
intensity \([s^{-1}]\)  
shim intensity i.e. intensity of a fully depolarised beam \([s^{-1}]\)  
unit-imaginary number
**List of Symbols**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Im$</td>
<td>imaginary part</td>
</tr>
<tr>
<td>$J_1(x)$</td>
<td>Bessel function of order one</td>
</tr>
<tr>
<td>$k$</td>
<td>Boltzmann’s constant [JK$^{-1}$]</td>
</tr>
<tr>
<td>$k$</td>
<td>wave vector of neutron ($= 2\pi/\lambda$) [nm$^{-1}$]</td>
</tr>
<tr>
<td>$K$, $K'$</td>
<td>coil constant of a precession coil [m$^{-1}$A$^{-1}$] and [TmA$^{-1}$], respectively</td>
</tr>
<tr>
<td>$K$</td>
<td>magnetocrystalline anisotropy constant [J/m$^3$]; chapter 7</td>
</tr>
<tr>
<td>$L$</td>
<td>thickness of sample [m] and length of precession coil [m]</td>
</tr>
<tr>
<td>$m$</td>
<td>neutron mass [kg]</td>
</tr>
<tr>
<td>$m$</td>
<td>reduced magnetisation ($= M/M_s$)</td>
</tr>
<tr>
<td>$M$</td>
<td>magnetisation [A/m] and magnetic interaction vector; chapter 8</td>
</tr>
<tr>
<td>$M_d$</td>
<td>DC-demagnetisation remanence [A/m]; section 4.3</td>
</tr>
<tr>
<td>$M_r$</td>
<td>remanent magnetisation [A/m] and isothermal remanent magnetisation [A/m]; section 4.3</td>
</tr>
<tr>
<td>$M_s$</td>
<td>saturation magnetisation [A/m]</td>
</tr>
<tr>
<td>$n_i$</td>
<td>the ‘$i$-component’ of the unit magnetic induction</td>
</tr>
<tr>
<td>$\vec{n}$</td>
<td>neutron propagation direction</td>
</tr>
<tr>
<td>$N$</td>
<td>nuclear interaction vector; chapter 8 and number of particles in the computation cell; chapter 5</td>
</tr>
<tr>
<td>$p_A$</td>
<td>probability distribution for initial target states</td>
</tr>
<tr>
<td>$P$</td>
<td>polarisation</td>
</tr>
<tr>
<td>$P$</td>
<td>polarisation vector and polarising power of the polariser; chapter 8</td>
</tr>
<tr>
<td>$\hat{P}$</td>
<td>P-matrix, matrix describing the quality of the polarising system</td>
</tr>
<tr>
<td>$Q$</td>
<td>polarising power of the analyser; chapter 8</td>
</tr>
<tr>
<td>$\hat{Q}$</td>
<td>Q-matrix, matrix describing the quality of the analysing system</td>
</tr>
<tr>
<td>$r$</td>
<td>position vector [m]; chapter 6</td>
</tr>
<tr>
<td>$r_n$</td>
<td>position vector of an atom in unit-cell [nm]; chapter 8</td>
</tr>
<tr>
<td>$r_0$</td>
<td>classical electron radius [nm]</td>
</tr>
<tr>
<td>$R$</td>
<td>polarising power of a polarising sample; chapter 8</td>
</tr>
<tr>
<td>$\hat{R}$</td>
<td>rotation matrix</td>
</tr>
<tr>
<td>$R^{+-}$, $R^{-+}$</td>
<td>spinflip reflectivity</td>
</tr>
<tr>
<td>$R^{--}$, $R^{++}$</td>
<td>non-spinflip reflectivity</td>
</tr>
</tbody>
</table>
### List of Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\mathbb{R}$</td>
<td>real part</td>
</tr>
<tr>
<td>$s$</td>
<td>reciprocal vector [nm$^{-1}$]</td>
</tr>
<tr>
<td>$\bar{s}$</td>
<td>unit reciprocal vector</td>
</tr>
<tr>
<td>$S$</td>
<td>reciprocal plane, i.e. plane perpendicular to $\bar{n}$</td>
</tr>
<tr>
<td>$S_p(s)$</td>
<td>structure factor</td>
</tr>
<tr>
<td>$t$</td>
<td>time variable [s] and particle thickness [m]; section 4.3</td>
</tr>
<tr>
<td>$t_p$, $t_Q$, $t_R$</td>
<td>transmission of a polariser, analyser and sample, respectively</td>
</tr>
<tr>
<td>$T$, $T_1$, $T_2$</td>
<td>depolarisation factors; chapter 8</td>
</tr>
<tr>
<td>$T_c$</td>
<td>Curie temperature [K]</td>
</tr>
<tr>
<td>$Tr[\hat{z}]$</td>
<td>trace of matrix $\hat{z}$</td>
</tr>
<tr>
<td>$V$</td>
<td>particle volume [m$^3$]</td>
</tr>
<tr>
<td>$\hat{V}_R$</td>
<td>scattering operator [m$^2$]</td>
</tr>
<tr>
<td>$\hat{V}_R(\kappa)$</td>
<td>Fourier transform of neutron-sample interaction potential multiplied by $(m/2\pi\hbar^2)$</td>
</tr>
<tr>
<td>$w$</td>
<td>normalised neutron wavelength spectrum [nm$^{-1}$] ($\int_0^\infty d\lambda w(\lambda) = 1$)</td>
</tr>
<tr>
<td>$W$</td>
<td>sample volume [m$^3$]</td>
</tr>
<tr>
<td>$x$, $y$ and $z$</td>
<td>Cartesian coordinate</td>
</tr>
<tr>
<td>$\hat{\alpha}$, $\hat{\beta}$</td>
<td>depolarisation matrix of polarisation rotator or spinflipper; chapter 8</td>
</tr>
<tr>
<td>$\hat{\alpha}$</td>
<td>correlation matrix [T$^2$m]</td>
</tr>
<tr>
<td>$\beta$</td>
<td>bond-angle [$^\circ$, [rad]]</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>gyromagnetic ratio ($= 1.83 \times 10^8$ s$^{-1}$T$^{-1}$)</td>
</tr>
<tr>
<td>$\gamma_i$</td>
<td>mean-square direction cosine</td>
</tr>
<tr>
<td>$\delta^L$, $\delta^T$</td>
<td>longitudinal and transverse domain size [m]</td>
</tr>
<tr>
<td>$\delta(x)$</td>
<td>Dirac delta function</td>
</tr>
<tr>
<td>$\delta_{ij}$</td>
<td>Kronecker delta function</td>
</tr>
<tr>
<td>$\Delta$</td>
<td>magnetic domain size [m]; chapter 7</td>
</tr>
<tr>
<td>$\Delta B$</td>
<td>deviation of the local magnetic induction from the average [T]</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>magnetic volume fraction of sample</td>
</tr>
<tr>
<td>$\zeta$</td>
<td>correlation length [m]</td>
</tr>
</tbody>
</table>
List of Symbols

$\eta$ mean direction of magnetic moment in sample
direction of magnetic moment

$\tilde{\eta}$

$\theta$ transmission angle [$^\circ$, [rad]]

$\kappa = k^0 - k'$ neutron wave vector change [nm$^{-1}$]

$\tilde{\kappa}$ unit neutron wave vector change

$\lambda$ wavelength of neutrons [nm]

$\lambda$ states of neutron in interaction with sample; chapter 8

$\mu_n$ atomic moment [JT$^{-1}$]

$\mu_0$ permeability of free space ($= 1.2566 \times 10^6$ mkG$^{-2}$)

$\mu$ permeability of matter [mkG$^{-2}$]

$\xi$ correlation parameter ($=\sum_{i} \alpha_{ii}$) [T$^2$m]

$\hat{\rho}$ spin density operator

$\hat{\sigma}$ Pauli matrix or spin operator

$\phi, \varphi$ rotation angle [rad], [$^\circ$]

$\chi$ spin-wave function

$\psi$ orientation of domain magnetisation [$^\circ$, [rad]]

$\omega$ neutron energy change divided by $\hbar$ [rad s$^{-1}$]

$\omega_L$ angular Larmor frequency [rad s$^{-1}$]

$\omega_G$ rate of change of the direction of $\mathbf{B}$ [rad s$^{-1}$]

$\Omega$ solid angle [sr]

$\hat{1}$ (2×2) unity matrix

$\langle x \rangle$ average of $x$

$\lvert \mathbf{z} \rvert$ length of vector $\mathbf{z}$

super- and subscripts:

$i, j$ and $k$ Cartesian component index

0 initial

$\prime$ final

$\perp$ perpendicular

$\parallel$ parallel
Chapter 1

General Introduction

In 1888 Oberlin Smith [Smi88] described how a thread of cotton or silk covered with small magnetic particles (the medium) could record and play sound signals. His idea was worked out by the Danish scientist Valdemar Poulsen in 1898 [Pou00a, Pou00b]. He demonstrated an apparatus for recording human voices at the World Fair in Paris in 1900; it consists of an iron piano string, the medium, at which the information is stored. A recording head consisting of a coil wound around a magnetic core was used to record and play the information. This apparatus, the so-called telegraphophone (see cover), was used to record telephone calls and, if necessary, to reproduce the call later on. Most parts of the telegraphophone can still be found in recorders present in our daily life. The only difference with the current generation recorders is the recording density; this is nowadays much larger. A comprehensive description of the development of magnetic recording from the turn of the century to present times can be found in ref. [Cam85].

The current applications of magnetic recording in the field of audio, video and data storage in the form of tapes, floppy disks and rigid disks in products such as computers and video-recorders, and for High Definition TeleVision (HDTV) applications, require high recording densities to ensure long playing times of the media. The current state-of-the-art is 0.6 gigabit/in², it is expected that it will increase to more than 5 gigabit/in² [Sim95] at the turn of the century. To achieve these high densities the size of the particles, i.e. the medium, has to become much smaller, i.e. down to 1000 nm³, and the recording head comes in direct contact with the media. In doing so, one meets various technical problems, e.g. the size of the particles becomes close to the superparamagnetic limit at which the magnetisation of the particle becomes thermally unstable. Furthermore, direct head-to-media contact causes serious wear problems of both media and head.

To overcome these difficulties, to push the current recording technology to its limits and to understand most of the processes involved in magnetic recording requires thorough study using different disciplines in science and technology, such as in magnetism, tribology, signal/data processing, materials science etc. In the late 80's, scientists working in these fields realised that high recording densities can only be achieved when the expertise in these fields is brought together. For that reason and to stimulate the re-
search activities in magnetic recording technology in both university and industry, the European research and development program entitled 'Concerted Action on Magnetic Storage Technology' (CAMST) was founded. In the early 90's, the 'Polarised Neutron Group' at the Interfacultair Reactor Instituut (IRI) in Delft (NL) joined CAMST. Using their expertise in the field of neutron depolarisation (ND), information on the magnetic correlations and interactions within the medium can be obtained from the change in polarisation of a polarised beam after transmission through the sample [RR91b]. It should be noted that the ND-technique measures the bulk magnetic properties rather than the surface properties of the media. Together with the results of magnetisation measurements, susceptibility measurements, magnetic force microscopy (MFM), transmission electron microscopy (TEM) and micromagnetic calculations it provides detailed insight in the magnetic properties of recording media. The aim of the research described in this thesis is to study the magnetic properties of both the recording medium and the recording head by means of the ND-technique. A part of the research described in this thesis was performed in the framework of the CAMST program.

Chapter 2 gives a brief summary of the ND-theory, a description of the ND set-ups located at IRI and the latest developments in upgrading the instruments. When the sample is in a (remanent) magnetised state the wavelength distribution of the beam results in additional depolarisation [RR92]. To avoid this additional depolarisation, the complicated numerical correction procedure, and in order to use the complete thermal neutron spectrum, a special method has been developed. This method, the so-called rotation compensation method, is based on the neutron spin-echo technique developed by Mezei [Mes72]. The rotation compensation method will be described in section 2.4. Up to now, three-dimensional ND-experiments have been carried out only in media which are in a remanent magnetised state or in media which are magnetised by low magnetic fields (up to several kA/m). To extend the range of applied fields up to 400 kA/m, so that ND-experiments can be performed along the magnetisation loop of magnetic recording media, a special magnetic coils system has been developed, built and tested. This system is also based on the principle of neutron spin-echo and hence it is baptised Spin-Echo Coil System (SpECS). A detailed description of this system is given in chapter 3.

Small $\text{CrO}_2$, $\gamma$-$\text{Fe}_2\text{O}_3$- and barium hexaferrite particles are some of the key ingredients in the manufacturing process of the recording medium. The intrinsic magnetic properties such as the saturation magnetisation, anisotropy and switching field distribution of these particles, together with the interaction between the particles determine to a large extent the recording characteristics and properties of the final media. Mostly, these properties are studied with the help of remanence curves and initial susceptibility measurements [SBCW88]. Information on the magnetic correlations and interactions between magnetic particles can be obtained from ND-experiments. In section 4.2 the ND-technique is applied to study magnetic correlations in media ($\gamma$-$\text{Fe}_2\text{O}_3$ particles) that are magnetised by high magnetic fields. These experiments were performed for the first time ever; it has been published in a slightly modified form as ref. [PKMR]. Section 4.3 describes the ND-experiments in media (barium hexaferrite particles) that
are in a remanent magnetised state after magnetisation by a short magnetic field pulse. These experiments were carried out in co-operation with the Institut für Physikalische Hochtechnologie in Jena (BRD). The results have been published in ref. [PGM].

An important step in the manufacturing process of (flexible) recording media is the coating of a suspension of magnetic particles in a binder solution, i.e. a dispersion, onto a polymer substrate, followed by drying and calendering [OGH91]. The microstructure of the dispersion and that of the final layer have also a great influence on the recording characteristics of the media. In section 5.2, the depolarisation of the beam is simulated from the calculated microstructure of the dispersion obtained from Monte-Carlo simulations. These simulations were carried out at the 'Magnetic Materials Group' at Keele University (UK). The results of these computations can be correlated with the experimental results in order to check the correctness of the model and to obtain insight in the dominant interactions that are responsible for the microstructure. This part of the chapter has been published as ref. [PCC]. In the framework of the CAMST Mobile Mill project the effect of the dispersion (milling) process of the particles in the dispersion is studied by various techniques at various laboratories around Europe; one of the techniques used was ND. The aim of the ND-experiments was to study magnetic correlations between particles as a function of the milling time. The results of these experiments are described in section 5.3 and published as ref. [PKM].

Most flexible recording media are manufactured by coating a solution of particles onto a substrate. Rigid disks, such as hard disks for computers, are usually thin magnetic films prepared by a sputtering or evaporation of a Co-alloy. In the mid 80's, the latter preparation process became also available for manufacturing of flexible media used in Hi-8 VCR applications. For this type of tape, a CoNi-alloy is evaporated in an oxygen atmosphere onto a substrate [FM84]; this tape is referred to as Hi8 Metal-Evaporated (ME) tape. In chapter 6, the ND-technique and Small Angle Neutron Scattering (SANS) have been used to determine the magnetic structure, i.e. the thickness of the magnetic layer, size of the magnetic domains and the orientation of the domain magnetisation in such a magnetic tape. Parts of this chapter have been published as refs. [PRC94, PRK].

Thus far in this thesis, the ND-technique has been used to study magnetic recording media. In the next chapter (chapter 7), based on ref. [ZJJN91], the ND-technique has been applied to study the relation between the magnetic domain size and grain size in MnZn-ferrites. MnZn ferrites are amongst others used in recording heads, where the characteristics of the head are at least partly determined by the magnetic structure of the head. Although this research has been initiated by the Philips Research Laboratory in Eindhoven (NL) and most of the work has been carried out there, it is a part of this thesis because it nicely illustrates the applicability of the ND-technique in the study of magnetic domain structures. Moreover, it confirms the theory that above a critical grain size the grains become multi-domain.

The experiments described in this thesis would have been impossible to perform without the existence of neutron polarisers. Mostly, magnetised single crystals or magnetised mirrors are used to polarise the beam and to analyse the polarisation of the
beam [Wil88, chap. 3]. Important characteristics of a neutron polariser that have to be optimised when developing and designing a new polariser are the transmission and/or reflection, and the net polarising power of the polariser. In a recent paper [PKR94], a method to determine the net polarising power of neutron polarisers, the so-called Sp-2s-method, has been described. This method has been used to measure the polarising power of the SpiegelPolarimeter (SP) polarisers at IRI, see ref. [SPP94]. In chapter 8 this method has been generalised in order to describe the measured intensity in a set-up composed of two polarisers, two spin-flippers and a polarising sample. In this generalised form, it gives useful hints to deduce the magnetic and crystallographic properties of the sample from the measured intensities. This chapter is a copy of ref. [PRF].

In the final chapter some concluding remarks will be given. Furthermore, suggestions will be given for the application of the neutron depolarisation technique and small angle neutron scattering in the study of magnetic recording media.
Chapter 2

Neutron Depolarisation Technique

2.1 Introduction

Three-dimensional neutron depolarisation (ND) is a powerful technique to obtain information on the micro and sub-micro magnetic properties of e.g. hard and soft magnetic materials [Rek89, ZJN91, DR91, JSRB94], spin-glasses [MIM91, MYE92] and superconductors [ZRM92, RR93]. Recently, the ND-technique has also been used to study austenitic to ferritic phase transformations in steel [KRBZ94].

In a 3-dimensional ND-experiment a polarised neutron beam, with its polarisation vector successively parallel to one of the laboratory axes, is sent through a magnetic material. After transmission, the polarisation is analysed along these axes. A net magnetisation of the sample results in a net rotation of the polarisation vector and inhomogeneities in the magnetic induction within the sample result in a reduction of the length of the polarisation vector, called depolarisation henceforth. From the change the mean magnetic domain size or magnetic correlation length along the propagation direction of the neutron beam, the mean-square direction cosines of the local magnetisation and the mean magnetisation of the sample can be determined. The characteristic length of the magnetic correlations or the size of the magnetic domains that can be measured in an ND-experiment ranges from 10 nm up to the millimeter region. This length scale is complementary to the sizes of magnetic inhomogeneities that can be measured in a Small Angle Neutron Scattering (SANS) experiment.

Rosman [Ros91] derived a relation between the micromagnetic state of the sample and the measured depolarisation. The relevant parts of the ND-theory will be summarised in this chapter. A more detailed and comprehensive description of the ND-theory is given by Rosman [Ros91].

The ND-experiments described in this thesis are carried out on the polarised neutron set-ups SpiegelPolarimeter (SP) and KristalPolarimeter (KP) located at IRI-Delft. Both set-ups are briefly described in section 2.3. The SP depolarisation module and the SP adiabatic polarisation rotators will be discussed in more detail in appendices 2.A and 2.B, respectively. Furthermore, the relation between the measured intensities
and the change in polarisation will be given. Roest and Rekveldt [RR92] realised that in magnetised media the beam will also depolarise due to the wavelength distribution of the beam. The numerical correction procedure for the additional depolarisation will also be discussed. To avoid this correction procedure and in order to perform ND-experiments using the complete thermal neutron spectrum, the neutron spin-echo method [Mes72] has been applied to compensate the rotation of the polarisation vector in the sample. This method, the so-called rotation compensation method, is described in section 2.4. It has been used in the ND-experiments described in chapter 4. First a summary of the ND-theory will be given in the next section.

2.2 Summary of the Neutron Depolarisation Theory

In an ND-experiment the change in polarisation of a polarised neutron beam is determined. This change can be expressed in the so-called $(3 \times 3)$ depolarisation matrix $\hat{D}$ defined by

$$ P' = \hat{D} P^0, \quad (2.1) $$

with $P^0$ and $P'$ the polarisation vector before and after transmission through a magnetic material, respectively. The polarisation vector is nothing else than the expectation value of the spin-operator $\hat{\sigma}$, i.e $P \equiv \langle \hat{\sigma} \rangle$.

In the Larmor approach to describe neutron depolarisation, the change in polarisation is a result of Larmor precession of the polarisation vector around the magnetic induction, averaged over the cross-section of the neutron beam. The time dependence of the polarisation vector $P(t)$ of a polarised neutron beam subjected to a magnetic induction $B(t)$ is given by the Larmor equation

$$ \frac{dP(t)}{dt} = \gamma P(t) \times B(t), \quad (2.2) $$

with $\gamma$ the gyromagnetic ratio $\gamma = 1.83 \times 10^8$ s$^{-1}$ T$^{-1}$. Equation (2.2) can be solved by successive iteration for a monochromatic neutron beam with wavelength $\lambda$. If the mean magnetic induction $\langle B \rangle$ of the sample is unequal to zero, it is convenient to write the local induction as $B(r) = \langle B \rangle + \Delta B(r)$. The component $\langle B \rangle$ results in a net rotation of the polarisation vector around $\langle B \rangle$ whilst $\Delta B(r)$, i.e. the deviation of the local induction from the average induction, results in depolarisation of the beam. Using eq. (2.1), the solution of eq. (2.2) averaged over the beam cross-section, for a sample consisting of $N$ uncorrelated thin slices of equal thickness $L_w$ and with a volume $W$, can be written as $^1$

$$ \hat{D} = \left( \hat{R} \left( \frac{L_w}{2} \right) \hat{D}(L_w, \hat{\alpha}) \hat{R} \left( \frac{L_w}{2} \right) \right)^N. \quad (2.3) $$

$^1$provided $\phi \ll 1$ in eq. (2.4).
2.2. Summary of the Neutron Depolarisation Theory

The expression between the outer braces describes the change in polarisation after transmission through one thin slice with mean magnetic induction \( \langle B \rangle \). The rotation matrix \( \tilde{R}(\mathbf{B}) \) describes the rotation of the polarisation vector due to transmission through such a slice over a distance \( L_w/2 \); its elements are given by

\[
\begin{align*}
R_{ii} & = 1 - (1 - \cos \phi) (1 - n_i^2), \\
R_{ij} & = (1 - \cos \phi) n_i n_j - \epsilon^{ijk} n_k \sin \phi, \quad i \neq j.
\end{align*}
\]  

(2.4)

Here, \( \phi = \sqrt{\varepsilon \lambda(B) L_w/2} \) is the rotation angle of the polarisation vector, the constant \( c = 2.18 \times 10^{29} \, \text{T}^{-2} \text{m}^{-4} \), \( n_i = \langle B_i \rangle / \langle B \rangle \), the quantity \( \epsilon^{ijk} \) is equal to 1 or -1 if \( i, j, k \) are (not) in cyclic order and the indices \( i, j, k \) are equal to \( x, y \) or \( z \). The exponent \( N = L/L_w \) is chosen such that the change in polarisation upon transmission over a distance \( L_w \) is small, i.e. \( |\Delta P| \ll |P| \), and \( L_w \) should be larger than, or at least equal to, the characteristic size of the magnetic correlations.

The relation between the elements of \( \tilde{D} \) and the micromagnetic state of the sample is given by [Ros91]

\[
\tilde{D}_{ij} = \delta_{ij} (1 - c \lambda^2 L_w \tilde{\xi}) + c \lambda^2 L_w \alpha_{ij},
\]  

(2.5)

with \( \xi = \sum_i \alpha_{ii} \) and \( \delta_{ij} \) the Kronecker delta. The subscripts \( j \) and \( i \) refer to the directions (i.e. \( x, y \) or \( z \)-direction) along which the polarisation is offered onto the sample and is analysed after transmission, respectively. In eq. (2.5)

\[
\alpha_{ij} = \frac{1}{W} \int d^3r \int_0^x dx' \Delta B_i(x') \Delta B_j(x') = \frac{8\pi^4}{W} \int_S d^2s \frac{B_i(s)B_j(-s) - L_w}{2 \langle B_i \rangle \langle B_j \rangle},
\]  

(2.6)

and

\[
\xi = \frac{1}{W} \int d^3r \int_0^x dx' \Delta \mathbf{B}(x') \cdot \Delta \mathbf{B}(x') = \frac{8\pi^4}{W} \int_S d^2s \mathbf{B}(s) \cdot \mathbf{B}(-s) - \frac{L_w}{2 \langle B \rangle^2},
\]  

(2.7)

with \( S \) the reciprocal plane perpendicular to the propagation direction \( \mathbf{n} \) of the neutron beam. In the experiments described in this thesis, \( \mathbf{n} \) is along the \( x \)-direction, hence \( S \) is the \( yz \)-plane. The last terms of eqs. (2.6) and (2.7) correct for the change in polarisation within \( W \) due to \( \langle B \rangle \). In other words they correct for the contribution of \( \langle B \rangle \) to the integral in the terms of eqs. (2.6) and (2.7) at \( s = 0 \). This contribution does not yield depolarisation. \( \mathbf{B}(s) \) is the Fourier transform of the magnetic induction distribution \( \mathbf{B}(r) \) in a volume \( W \) of the sample with a thickness \( L_w \); it is given by

\[
\mathbf{B}(s) = \frac{1}{(2\pi)^3} \int_W d^3r \mathbf{B}(r) e^{is \cdot r} = \frac{\mu_0}{(2\pi)^3} \int_W d^3r (\mathbf{s} \times \mathbf{M}(r) \times \mathbf{s}) e^{is \cdot r}.
\]  

(2.8)

The vector \( s \) is a reciprocal vector in plane \( S \) and \( \mathbf{s} = s/|s| \). The expression between brackets in eq. (2.8) is nothing else than the component of \( \mathbf{M}(r) \) perpendicular to \( s \), that causes the depolarisation. The vector product is a consequence of applying Maxwell laws \( \nabla \cdot \mathbf{B}(r) = 0 \) and \( \nabla \times \mathbf{H}(r) = 0 \). The final result shows that only the local magnetisation \( \mathbf{M}(r) \) is involved in the final expression for \( \mathbf{B}(s) \) (see eq. (2.8)).
Note, that one does not have to bother about the demagnetising fields in a particulate media, because they have been effectively accounted for using this formalism.

The element $\alpha_{ij}$ of the correlation matrix $\hat{\alpha}$ describes the mean correlation between $\Delta B_i$ and $\Delta B_j$ along the neutron path $\hat{n}$. The correlation parameter $\xi$ is proportional to the correlation length of $(\Delta B)^2$ along $\hat{n}$. For particulate media the magnetic correlation length $\zeta$ is defined as

$$\zeta = \frac{2\xi}{\varepsilon (\Delta B)^2},$$

with $(\Delta B)^2 = (\mu_0 M_r)^2$, $M_r$ is the saturation magnetisation of the particles and $\varepsilon$ is the volume fraction of the magnetic material in the sample. The correlation length $\zeta$ may be seen as the mean length of a neutron trajectory in which the $B(r)$ is aligned more or less parallel. A factor two is added to the definition of $\zeta$ in order to relate it directly to the mean magnetic domain size along $\hat{n}$. It should be noted that a mean magnetic induction of the sample unequal to zero results in a trivial decrease of $\Delta B(r)$ and therefore of $\zeta$. This is important when analysing the dependence of $\xi$ (and also $\gamma_i$) on the magnetic state of the sample. However, Rosman [Ros91, pp. 36] showed that this correction is small for most particulate magnetic media dealt with in this thesis. Hence, it will be omitted in the analysis of these ND-experiments. The mean-square direction cosines of $\Delta B(r)$, i.e. the magnetic texture, $\gamma_i$ are given by

$$\gamma_i = \frac{\alpha_{ii}}{\xi}.$$  

(2.10)

For an isotropically distributed, magnetically uncorrelated ensemble of single domain particles with $\hat{n}$ along the x-direction, the theory derived by Rosman [RR91b] predicts

$$\gamma_x = 1/2,$$
$$\gamma_y = 1/4$$
$$\gamma_z = 1/4$$

independent of the shape of the particles. For a non-isotropic distribution of particles or magnetic inductions within the sample, the values for $\gamma_i$ depend on the actual shape and orientation of the particles.

If the sample has no net magnetisation $\hat{R}$ is equal to the unity matrix, and if $\alpha_{ij} = 0$ for $i \neq j$ the depolarisation matrix is diagonal

$$D_{ii} = e^{-c\lambda^2 L (\xi - \alpha_{ii})},$$
$$D_{ij} = 0, \quad i \neq j.$$  

(2.11)

The diagonal element $\alpha_{ii}$ and correlation parameter $\xi$ follow from

$$\alpha_{ii} = \frac{2\ln(D_{ii}) - \ln(\det(\hat{D}))}{2c\lambda^2 L},$$

(2.12)

and

$$\xi = -\frac{\ln(\det(\hat{D}))}{2c\lambda^2 L}.$$  

(2.13)
2.3. Description of a Neutron Depolarisation Experiment

In the case that the sample has a net magnetisation along the z-axis and under the approximation that \( \alpha_{xx} = \alpha_{yy} = \frac{(\xi - \alpha_{zz})}{2} \) the depolarisation matrix has finite non-diagonal elements. Then the elements of \( \tilde{D} \) are given by

\[
\begin{align*}
D_{xx} &= D_{yy} = \cos \phi \ e^{-c \lambda \mathcal{L} (\xi - \alpha_{zz})}, \\
D_{xy} &= -D_{yx} = \sin \phi \ e^{-c \lambda \mathcal{L} (\xi - \alpha_{zz})}, \\
D_{zz} &= e^{-c \lambda \mathcal{L} (\xi - \alpha_{zz})} \quad \text{and} \\
D_{zz} &= D_{xx} = D_{yx} = D_{xy} = 0,
\end{align*}
\]

with \( \phi = \sqrt{c \lambda (B \mathcal{L})} \). It should be noted that the ND-technique yields only the rotation angle \( \phi \) modulo \( 2\pi \). The correlation parameter \( \xi \) and the element \( \alpha_{zz} \) follow from eqs. (2.13) and (2.12), respectively. In the next section the ND set-ups at IRI-Delft and the relation between the measured intensity and the elements of the depolarisation matrix will be described.

2.3 Description of a Neutron Depolarisation Experiment

2.3.1 Neutron Depolarisation Set-ups SP and KP

The experiments described in this thesis are carried out on the SpiegelPolarimeter (chapter 4 - 6) and KristalPolarimeter (chapter 7), denoted 'SP' and 'KP', henceforth. The set-ups are operated at the 2MW swimming pool reactor Hoger Onderwijs Reactor HOR of IRI-Delft.

SP

A polychromatic thermal neutron beam with \( \lambda \) between \( \approx 0.1 - 0.7 \) nm, emerging from the Stacked Neutron Guide [WHR91], located at beam tube L2 of the HOR, is polarised by a polariser (\( P \)). Figure 2.1 gives a schematic outline of the SP. Polariser \( P \)

![Diagram of SP set-up](image)

Figure 2.1: Schematic drawing of the SP. The propagation direction of the neutrons in the set-up is given by \( \hat{n} \). See the text for explanation of the other symbols.

is of the so-called curved polarising Soller guide type. It consists of a large number of polarising Co\(_{64}\)Fe\(_{34}\)V\(_2\)/Ti\(_{60}\)Zr\(_{40}\) multilayer mirrors, which are stacked together to form
a series of curved polarising and conducting channels. The mirrors are magnetised to saturation by a magnetic field applied in the z-direction. Neutrons with their spin parallel to the direction of magnetisation are (totally) reflected. Neutrons with their spin in the opposite direction are (almost) not reflected; they are absorbed in the mirrors and shielding around the polariser. The polychromatic beam beyond $P$, as seen along the neutron path, is polarised in the z-direction.

The polarisation of the beam can be rotated towards the x or y-direction (or their opposite direction) or preserved in the z-direction using the first adiabatic polarisation rotator (R1). Details about the polarisation rotator are given in appendix 2.B. The polarisation of the beam is successively analysed along these three directions using a second adiabatic polarisation rotator (R2) and an analyser ($Q$). The analyser is of the same type as the polariser but the mirrors are magnetised in the opposite direction. The polarising power and transmission of $Q$ as a function of the neutron wavelength are shown in figure 2.2. Details about the construction and characteristics of the polariser

![Figure 2.2: Polarising power (x) and transmission (o) of analyser Q as a function of the neutron wavelength (from [SPP94]). The polarising power has been determined using the '3p-2s' method (see chapter 8).](image)

and analyser are given by Schebetov et al. [SPP94]. The sample is positioned between R1 and R2 inside a samplebox (S). This iron samplebox screens the sample and neutron beam from magnetic stray fields from outside. The two polarisation rotators and the samplebox are assembled in one rigid module, the so-called SP depolarisation module. The depolarisation module will be described in appendix 2.A.

If desirable, the neutron beam can be monochromatised by Bragg-reflection at the (002)-planes of a pyrolytic graphite monochromator crystal (MC). The reflected neutrons are detected in the multidetector (MD), comprising an array of thirty-two $^3$He-detectors. The non-reflected neutrons are detected in the BF$_3$-detector (D). The
shim intensity, i.e. the intensity of a fully depolarised beam, is typically $10^5$ ncm$^{-2}$s$^{-1}$ for the polychromatic beam.

KP

The KP is located at beam tube R2 of the HOR. In the KP the neutron beam is polarised and at the same time monochromatised by Bragg-reflection at the (111)-planes of a Cu$_2$MnAl single-crystal. The crystal is magnetised to saturation in the z-direction. Then the nuclear and magnetic structure factor will add for neutrons with their spin anti-parallel to the direction of magnetisation of the crystal. The total structure factor is zero for neutrons with their spin in the opposite direction. Hence, they are not reflected at the reflecting planes of the crystal. The reflected monochromatic beam with $\lambda = (0.16 \pm 0.01)$ nm is polarised along the z-direction.

Using polarisation rotators, based on the principle of Larmor precession [Rek71, RS79], the polarisation vector can be offered to the sample along the three laboratory axes $x$, $y$ or $z$. The optimal value for the DC-current through the rotators is found in a calibration experiment with a calibration coil at the position of the sample (see for a description [RR92]). After transmission through the sample the polarisation is successively analysed along one of the laboratory axes using a second polarisation rotator, an analyser and a $^3$He-detector. The analyser is of the same type as the polariser; it is magnetised in the opposite direction. The shim intensity was typically 2000 ncm$^{-2}$s$^{-1}$ and the polarisation of an empty beam is $\approx 0.83$.

Recently, the polariser and analyser were replaced by curved polarising Soller guides. The beam is monochromatised by a pyrolytic graphite crystal. The shim intensity is increased by approximately a factor of 20. Furthermore, the polarisation of the empty beam is increased to $\approx 0.95$.

2.3.2 Relation Between the Measured Intensity and the Elements of the Depolarisation Matrix

The relation between the elements of the depolarisation matrix and the intensity measured when the set-up operates in the 'ij-mode' and when the polariser and analyser are magnetised in opposite directions is given by [RR92]

$$1 - \frac{I_{ij}}{I_s} = \int d\lambda \ w(\lambda) \left( \hat{Q}(\lambda) \hat{D}(\lambda, \hat{\alpha}, \langle B_i \rangle) \hat{P}(\lambda) \right)_{ij}.$$  

(2.15)

The matrices $\hat{P}$ and $\hat{Q}$ describe the quality of the polarising ($P$ and R1, see figure 2.1) and analysing system ($Q$ and R2, see figure 2.1), respectively. The elements of these matrices are determined in a separate calibration experiment. This procedure is described by Roest and Rekveldt in ref. [RR92]. The integral on the right-hand side of eq. (2.15) accounts for the wavelength distribution $w(\lambda)$ of the neutrons that enter the detector(s); $w(\lambda)$ obeys $\int_0^\infty d\lambda \ w(\lambda) = 1$. The intensity $I_s$ is the shim-intensity. The shim intensity can be determined using an iron-sheet, the 'shim'. When
a polarised neutron beam is sent through the shim it fully depolarises due to the large magnetic domains, with high saturation magnetisation, within the shim. It can also be determined from the measured intensities \( I_i \) and \( I_{ii} \), with \( i \) is x, y or z and \( i \) the negative i-direction. Then \( I_s \) is defined as

\[
I_s = \frac{I_{ii} + I_{ii}}{2}.
\]

This method of determining \( I_s \) has the advantage that one does not need to correct for the wavelength dependent transmission of the shim.

In general, the element \( \alpha_{ii} \) and the component of the mean magnetic induction \( \langle B_i \rangle \) can only be determined numerically from eq. (2.15). However, in the specific case that \( \tilde{D}, \tilde{\hat{P}} \) and \( \tilde{Q} \) are diagonal matrices, then \( D_{ij} \) is equal to zero for \( i \neq j \) and eq. (2.15) can be rewritten into

\[
D_{ii} = e^{-c\lambda^2(t-\alpha_{ii})} = \frac{1 - I_{ii}/I_s}{P^n_{ii}},
\]

with \( P^n_{ii} = (Q \tilde{P})_{ii} \) the polarisation of a so-called empty beam. \( P^n_{ii} \) is found in an experiment without a (magnetic) sample in the beam, by a formula analogous to the numerator of eq. (2.17). In the case that the neutron beam is monochromatic, the elements of \( \hat{\alpha} \) and hence \( \zeta \) and \( \gamma_i \) follow directly from the measured intensities using the equations given in section 2.2. If the beam is not monochromatic, but \( \hat{\tilde{D}}, \hat{\tilde{Q}} \) and \( \hat{\tilde{D}} \) are still diagonal matrices, \( \zeta \) and \( \gamma_i \) can be calculated using the same formulae as in the monochromatic case. Then one simply has to substitute for the neutron wavelength \( \lambda \) the effective or mean wavelength of the neutron spectrum \( w(\lambda) \) used.

In all other cases one has to deduce \( \alpha_{ii} \) and \( \langle B_i \rangle \) numerically from eq. (2.15). When doing so one encounters two major numerical and experimental difficulties:

- the rotation angle \( \phi \) can only be determined within \( 2\pi \), hence the mean magnetic induction becomes ambiguous when \( |\phi| > 2\pi \),

- if the mean magnetic induction \( \langle B \rangle \) is finite the rotation angle \( \phi \) is unequal to zero.

If the beam is not monochromatic, i.e. \( \Delta \lambda / \lambda \neq 0 \), the rotation angle \( \phi \) smears out due to the wavelength spread \( \Delta \lambda \) of the beam (see fig. 2.3). As a result, the component of the polarisation of the neutron beam that is perpendicular to the direction of the mean magnetic induction depolarises. After transmission through the sample the neutron beam is not only depolarised due to \( \Delta \mathbf{B}(r) \) but also due to \( \Delta \lambda \). To distinguish between the two causes of depolarisation the first will be referred to as ordinary depolarisation and the second will be referred to as additional depolarisation. The amount of additional depolarisation increases with increasing rotation angle and increasing wavelength spread. Eventually, the neutron beam will fully depolarise. The expression at the right-hand side of eq. (2.15) approaches zero. Then retrieval of the elements of the correlation matrix \( \hat{\alpha} \) from the measured intensities, by solving eq. (2.15), is not possible any more. So, the information on the magnetic correlations in the sample, contained in the ordinary depolarisation, is lost.
2.4. Rotation Compensation Method

The additional depolarisation may dominate the measured depolarisation in magnetised media, especially in media where the ordinary depolarisation is small. In order to measure these small changes in polarisation of the beam with sufficient statistics one has to collect a large number of neutrons in the detector. Usually, the complete thermal neutron spectrum is used. For three-dimensional ND-experiments in magnetised media utilisation of the complete spectrum is not possible in the normal way, because of the additional depolarisation.

In order to use the complete neutron spectrum for the ND-experiments and to avoid the numerical correction procedure [RR92] for the additional depolarisation, a method was developed to rotate the polarisation vector, after transmission through the sample, back to its original orientation. So, no net rotation of the polarisation vector occurs between the first and second polarisation rotator and hence no additional depolarisation occurs, only the ordinary depolarisation is left. This method will be referred to as rotation compensation method, henceforth.

2.4.1 Description of the Method

When applying the rotation compensation method, which is based on the principle of neutron spin-echo [Mea72], the polarisation vector of the neutron beam after transmission through the sample is rotated back to its original direction. For this purpose, a precession coil is positioned behind the sample (see fig. 2.3). This coil, referred to as the compensation coil henceforth, allows one to apply a magnetic field along the direction of the mean magnetic induction within the sample.

When the neutron beam successively traverses the sample and the compensation coil, the change in polarisation can be expressed in a resultant depolarisation matrix \( \hat{D}' = \hat{R} \hat{D} \). The matrix \( \hat{R} \) is the rotation matrix that describes the rotation of the

![Figure 2.3: Principle of the rotation compensation method.](image)

\[ P' = \hat{R} \hat{D} P^0 = \hat{D}' P^0 \]

\[ I_c = I_c^0 \quad \text{comp. coil} \]

\[ \lambda_0 + \Delta \lambda \quad \lambda_0 - \Delta \lambda \]

\[ \langle B \rangle L \quad \text{sample} \]

\[ \hat{D} \]

\[ \phi \]

\[ \hat{R} \]

\[ \phi_c \]

\[ P' \]

\[ P^0 \]

\[ \hat{n} \]

---

2This section will be published in a modified form as: 'Neutron Spin-echo Technique Applied to 3-Dimensional Polarisation Analysis in Magnetised Media', P.T. Por, W.H. Kraan and M.Th. Rekveldt, Nucl. Instr. Meth. in Phys. Res. (submitted).
polarisation vector due to transmission through the compensation coil. The elements of this matrix are given by eq. (2.4). The matrix $\bar{D}$ is the depolarisation matrix of the sample; its elements are given by eq. (2.14). If the mean magnetic induction of the sample and the applied field in the compensation coil are both along the z-direction, the elements of $\bar{D}^\prime$ are given by

\begin{align}
D^\prime_{xx} &= D^\prime_{yy} = \cos(\phi + \phi_c) e^{-c\lambda^2 L(\xi - \alpha_{xx})}, \\
D^\prime_{xy} &= -D^\prime_{yx} = \sin(\phi + \phi_c) e^{-c\lambda^2 L(\xi - \alpha_{zz})}, \\
D^\prime_{zz} &= e^{-c\lambda^2 L(\xi - \alpha_{zz})} \text{ and} \\
D^\prime_{xx} &= D^\prime_{yy} = D^\prime_{yz} = D^\prime_{xy} = 0.
\end{align}

(2.18)

The angle $\phi_c (= \sqrt{c\lambda B_c L_c} = K \lambda^2 I_c)$ is the rotation angle of the polarisation vector due to transmission through the compensation coil. The constant $K$ is a so-called coil constant and $I_c$ is the DC-current through the compensation coil. For reasons of simplicity, the depolarisation of the beam due to inhomogeneities in the magnetic field applied in the compensation coil is omitted. It can easily be taken into account by including a depolarisation coefficient in the argument of the exponent in eq. (2.18).

The polarisation vector is rotated back to its original orientation when $\phi_c + \phi = 0 \mod 2\pi$ rad. However, this is only the case for neutrons with a specific wavelength $\lambda_0$. For neutrons with a wavelength slightly different from $\lambda_0$, the net rotation angle, i.e. $\phi_c + \phi$, is not equal to $(0 \mod 2\pi)$ rad. The polarisation vector is rotated back to its original orientation, independent of the wavelength of the neutrons used, only if $\phi_c = -\phi$. Then it is said that the rotation of the polarisation vector is compensated. Hence, $\bar{D}^\prime$ is a diagonal matrix.

The measured depolarisation matrix only contains information on the magnetic correlations in the sample. The magnetic correlation length $\xi$ and the mean-square direction cosines of the local magnetisation $\gamma_i$ can easily be calculated from the measured depolarisation using the procedure described in the previous section. The information on the mean magnetic induction $\langle B \rangle$ of the sample is not contained in the depolarisation matrix itself. It can be calculated from the DC-current $I_c^0$ through the compensation coil at which the rotation of the polarisation vector is compensated

\[ \langle B \rangle = \frac{-K I_c^0}{\sqrt{c} L} = \frac{-K' I_c^0}{L}. \]

(2.19)

The constant $K'$ is a constant depending on the coil geometry, number of turns and the length of the compensation coil; it can be determined in a separate calibration experiment in which the sample is replaced by a coil or a calibration sample, e.g. nickel, both with a known $\langle B \rangle L$.

### 2.4.2 Implementation of the Method

The DC-current $I_c^0$ is found in a separate experiment prior to the measurement of the elements of the depolarisation matrix. For this purpose, a so-called Larmor-spectrum is measured (see figure 2.4). In Larmor-spectroscopy experiments [KTR89],...
the intensity of an initially polarised neutron beam is measured as a function of the magnetic field applied in, or DC-current through, a precession coil (for application of the rotation compensation method the compensation coil is used as the precession coil). This coil is positioned between the polariser and analyser. The field in the coil is applied perpendicular to the initial direction of the polarisation vector. After transmission, the polarisation is analysed along the initial direction of the polarisation. Kraan et al. [KTR89] showed that, when the field is applied along the z-direction and the set-up operates in the 'zx-mode', the intensity is given by

\[ I_{zx}(I_c) = I_{\text{ shim}} \left( 1 - \int d\lambda \, w(\lambda) P_{zx}^0(\lambda) D_{xx}(\lambda) \cos \varphi \right), \tag{2.20} \]

with \( \varphi = K\lambda I_c \) the rotation angle of the polarisation vector due to transmission through the coil.

When applying the compensation method, \( \varphi \) is the sum of the rotation angle \( \phi \) and \( \phi_c \):

\[ \varphi = K\lambda I_c + \phi, \tag{2.21} \]

with \( \phi = \sqrt{\langle B \rangle} L \). One can easily show that eq. (2.20) has a minimum, independent of the neutron spectrum \( w(\lambda) \) used, when the rotation angle \( \varphi \) of the polarisation vector is equal to zero, i.e. \( \phi = -K\lambda I_c \). So, the current \( I_c \) at which the intensity \( I_{zx} \) has a minimum, yields the compensation current \( I_c^0 \) and hence the mean magnetic induction \( \langle B \rangle \) follows from eq. (2.19). Figure 2.4 gives an example of a measurement of \( I_{zx} \) as a function of the current through the compensation coil when \( \langle B \rangle L \) is equal and unequal to zero. It should be noted that the top curve is shifted over a distance equivalent to 10000 units.

The procedure of measuring the intensity \( I_{zx} \) as a function of the DC-current \( I_c \) has been implemented in the software of the SP. After measuring the required intensities, the program searches for the current at which the measured intensity has a minimum. Since \( I_{zx} \) is measured at a step-by-step increasing current \( I_c \), the minimum thus found does not necessarily coincide with the real position of \( I_c^0 \). In order to find the exact value for \( I_c^0 \), the measured intensity in the vicinity, i.e. left and right nearest neighbour, of the minimum intensity is approximated by a parabola. The current at which this parabola has a minimum yields the compensation current \( I_c^0 \).

### 2.4.3 Final Remarks

Before finishing this section dealing with the rotation compensation method two final remarks have to be made.

- First, the procedure of finding the compensation current uses only three data-points out of a set of \( N \) data-points (in the experiments described in this thesis \( N \) is equal to 200). The other data points are ignored. If the ordinary depolarisation is large and/or the wavelength spread \( \Delta \lambda \) of the neutron beam is small, the difference in intensity of the 'real' minimum and a neighbouring minimum, denoted
0 and ±1 respectively in figure 2.4, is small. Then it may occur, due to the bad counting statistics in each data-point, that the procedure finds a 'wrong' compensation current, i.e. \( \phi_c + \phi \neq 0 \). Hence, \( \mathbf{D}' \) has finite non-diagonal elements and additional depolarisation occurs. The number of 'wrong' compensations can be decreased by improving the counting statistics in the Larmor-spectroscopy experiment. Note that the time needed to measure a Larmor-spectrum then increases. However, if the neutron beam is monochromatic and the depolarisation is large, it is still possible that the program finds a 'wrong' compensation current. Instead of only improving the counting statistics, the performance of the method can be improved by using the other data-points that are ignored thus far.

As stated before, equation (2.20) has a minimum independent of \( w(\lambda) \) when \( \varphi \) is equal to zero. Besides this characteristic property, equation (2.20) and hence the measured intensity is symmetric with respect to \( \varphi = 0 \), i.e. \( I_{xx}(-\varphi) = I_{xx}(\varphi) \). Instead of only searching for the minimum intensity, the procedure searches for the current at which the measured intensity is symmetric. This can be realised in the software by choosing an arbitrary current as 'symmetry' current. Next, the difference in intensity of corresponding data-points, with respect to the symmetry current is calculated. The squares of these differences are summed up for all, e.g. \( N/2-1 \), data-points. This procedure is repeated, for different posi-
2.4. **Rotation Compensation Method**

...tions of the symmetry current. The symmetry current at which the sum of the square differences has a minimum yields the compensation current \( J^0_c \). Using this procedure, the statistics in every measured intensity is averaged out. This will improve the performance of the compensation method, i.e. the number of wrong compensation is decreased. Furthermore, the compensation method can still be used if the depolarisation is large and probably also when the neutron beam is monochromatic.

- Secondly, using the rotation compensation method the direction of the mean magnetic induction of the sample has to be known. Thus the direction along which the field in the compensation method has to be applied is known prior to the measurement of the Larmor-spectrum. This is a direct consequence of the non-commutativity of rotation matrices when the rotation angles are large, which is usually the case when applying the rotation compensation method.
2. A SP Depolarisation Module

The adiabatic polarisation rotators and the samplebox are assembled in one rigid box, the so-called depolarisation module (see figure 2.A.1). The module is used to screen translation stage rotation stage diaphragm

iron shielding

polarisation rotator permanent magnets

μ-metal shielding

Figure 2.A.1: Schematic drawing of the SP depolarisation module. In the figure the lid of the depolarisation module has been removed. Note that in this figure the propagation direction \( \hat{n} \) of the neutron beam is from right to left.

the sample and also the neutron beam from magnetic stray fields, e.g. magnetic fields emerging from the polariser and analyser and the earth magnetic field.

Magnetic Stray Field Shielding

A magnetic field applied to the polarised neutron beam results in a rotation of the polarisation around the magnetic field. The rotation angle is proportional to the wavelength of the neutrons, the magnitude of the field, and to the length over which the field is present. Furthermore, the neutron beam depolarises also due to transmission
through a region with an inhomogeneous magnetic field. In order to avoid depolarisation and to keep the magnetic field as low as possible, i.e. $< 1 \text{ A/m}$, so that the net rotation angle of the polarisation vector is less than 1 rad, the shielding is made of iron and $\mu$-metal. The idea of using a magnetic shielding using several layers of shielding material is described by Boll and Borek [BB80] and has been successfully used by Bitter et al. [BEE91]. The iron shielding, which has a thickness of 10 mm, is used to screen the high magnetic fields. The remaining field is screened by $\mu$-metal with a thickness of 1.5 mm. The iron and $\mu$-metal shielding are separated by 40 mm non-magnetic material, which is in this case air.

Stray Field Compensation Coils

From measurements of the magnetic field inside the module it appeared that the fields are in the order of 1 A/m, independent of the strength of the field outside the module. However, a magnetic field in the order of 1 A/m present over a length of 0.3 m still results in a net rotation angle of approximately 1 rad, for neutrons with a wavelength of 0.4 nm. To reduce the rotation angle, three small Helmholtz-coils are mounted inside the module (not shown in the figure). They allow to apply a magnetic field, independent from each other, in the x, y and z-direction. The DC-current at which the net rotation angle is equal to zero is found in an experiment in which the non-diagonal elements of the depolarisation matrix are measured. In this procedure the complete spectrum of the SP is used. A computer program, implemented in the software of the SP, changes iteratively the current through the coils until the element $D_{ij}$ of the depolarisation matrix is equal, within the accuracy of the experiment, to the element $D_{ij}$ (with $i \neq j = x, y$ or $z$). Then the net rotation angle is zero. In an ideal case all the non-diagonal elements are equal to zero, but due to misadjustment of the polarisation vector, with respect to the laboratory axes $x$, $y$ and $z$, a small deviation from zero occurs. But in first approximation the depolarisation matrix is a rotation matrix. As can easily be verified using eq. (2.4) the net rotation angle is zero if $D_{ij} = 0$ for $i \neq j$. It should be noted that the field applied by the Helmholtz-coils only compensates the net rotation of the polarisation vector. This will not necessarily mean that in every position inside the depolarisation module the magnetic field is reduced to zero.

Rotation and Translation Stages

In order to rotate and/or to translate the sample in the beam, a rotation stage and two translation stages are mounted in the module. One translation stage allows to translate the sample over a distance of 20 mm in the $z$-direction, the so-called 'up-down' movement. The other stage is used to translate the sample over a distance of 40 mm in the $y$-direction, the so-called 'forward-backward' movement. These two stages are driven by small DC-motors. With the rotation stage, driven by a stepping motor, the sample can be rotated over $\pm 180^\circ$. All the movements of the sample can be controlled
either by the electronics and software of the SP or manually.

Diaphragms

Two diaphragms are mounted onto the left and right side of the depolarisation module. The distance between the two diaphragms is one meter. With the diaphragms, made of boron nitride, the cross-section of the beam can be confined to a rectangle with dimensions \((y \times z)\) mm\(^2\). The width \(y\) can be adjusted from 0 to 40 mm and the height \(z\) from 0 to 60 mm. The accuracy in adjusting the diaphragms is approximately 0.05 mm.

Adiabatic Polarisation Rotators

The depolarisation module also contains the two adiabatic polarisation rotators and a set of permanent magnets used to improve the performance of these rotators. These components are discussed in appendix 2.B.
2.B SP Adiabatic Polarisation Rotators

In 1989, the SP polarisation rotators, at that time equivalent to the ones currently installed in the KP, were replaced by rotators which are based on the principle of adiabatic rotation. In contrast to the former ones, they realise an orthogonal adjustment of the polarisation vector for the complete thermal neutron spectrum. The so-called adiabatic polarisation rotators allow to measure the elements of the depolarisation matrix simultaneously at different wavelengths. Furthermore, the complete neutron spectrum can be used.

Principle of Operation

The operation of the rotators is based on the equation of motion for the polarisation vector $\mathbf{P}$ in a magnetic field $\mathbf{B}$ (see eq. (2.2)). In a homogeneous field its solution is a rotation of $\mathbf{P}$ around $\mathbf{B}$ with the angle between $\mathbf{P}$ and $\mathbf{B}$ remaining constant. This is the well-known Larmor precession which occurs at an angular frequency $\omega_L = \gamma |\mathbf{B}|$.

If the field is inhomogeneous, one should consider the rate of change $\dot{\omega}_G$ of the direction of $\mathbf{B}$ as seen in a coordinate system moving with the neutron. If

$$\dot{\omega}_G \ll \omega_L,$$  \hspace{1cm} (2.B.1)

and if the initial angle between $\mathbf{P}$ and $\mathbf{B}$ is zero, the vector $\mathbf{P}$ will precess around the local $\mathbf{B}$ and the angle between $\mathbf{P}$ and $\mathbf{B}$ remains less than $\omega_G/\omega_L$. The vector $\mathbf{P}$ follows $\mathbf{B}$ adiabatically. Considering this, an adiabatic rotation of the vector $\mathbf{P}$ from its initial direction ($z$) towards a desired direction $x$ and $y$ can be realised by providing a magnetic field along the neutron path that gradually rotates from the $z$-direction to the $x$ or $y$-direction. If the strength of the fields is chosen such that at each position along the neutron path the condition (2.B.1) is fulfilled for the minimum wavelength in the neutron spectrum, it will be fulfilled over the entire spectrum.

The realisation and testing of such rotators is described by Kraan et al. [KRP91]. They found that the misadjustment of the polarisation vector along the axes of the laboratory coordinate system is less than $5^\circ$ for neutron wavelengths ranging between 0.2 and 0.6 nm.

Realisation

The polariser and analyser of the SP which were recently installed have, in contrast to the former ones, a much better transmission for neutrons with small wavelength. Hence, the spectrum of the SP is shifted towards smaller neutron wavelengths, i.e. $\lambda \approx 0.1 - 0.7$ nm. The neutrons with such a small wavelength do not fulfill the condition for adiabatic rotation sufficiently well. As a consequence, the design of the rotators had to be reconsidered. To fulfill the condition for adiabatic rotation (a) the strength of the field has to be increased or (b) the length over which the gradual change in direction of $\mathbf{B}$ takes place has to be increased, or it can be fulfilled by combining
Figure 2.B.1: Top: schematic cross-section view of the rotator. For reasons of clarity the coils 3 are omitted. These coils are used to maintain the polarisation in the z-direction. Centre: the magnetic field generated by coils 1 (o) and 2 (∆) and the stray field and field emerging from the permanent magnets (dashed line) versus position x along the neutron path. Bottom: result of a simulation of the rotation of the polarisation vector starting (0,0,1) at $x = -0.3$ to (0,1,0) at $x = 0.0$ for neutrons with $\lambda = 0.25$ nm.
options (a) and (b). For the present rotators, the length over which the gradual change in direction takes place has been increased from 0.15 to 0.3 m. The strength of the field in the both the x, y and z-direction has been increased by 10%. The size of the rotator perpendicular to the direction of the beam, i.e. size in the yz-plane, remains unchanged.

Figure 2.8.1 (top) shows a schematic outline of the adiabatic polarisation rotator currently used in the SP. It consists of a set of three orthogonal coils denoted 1, 2 and 3. For reasons of clarity the coils 3 (analogous to the coils used for rotation towards the y-direction, but rotated over π/2 around x) are omitted.

The stray field of the polariser (or analyser) and the field emerging from the permanent magnets are combined with the fields of the coils for the realisation of the rotation of the polarisation vector. For a rotation from the z-direction towards the x or y-direction, coils 1 and 2 are used, respectively. Coils 3 are used for preserving the polarisation vector in the z-direction.

Figure 2.8.1 (center) gives the fields generated by the coils 1 and 2 for a current of 1 A. Coils 2 and 3 are shaped such that the field gradually increases along the neutron path and suddenly drops to zero at x = 0.0, a so-called Majorana field flip [Maj32]. This is realised by shaping these coils like wire screens at position x = 0. For x > 0.0 the vector P is no longer affected by the magnetic field in the rotator. For coil 2 this is not possible because of Maxwell’s laws; it can only be shaped such that the field field gradually decreases without causing a too inhomogeneous magnetic field. In this way the depolarisation of the beam is small.

In figure 2.8.1 (bottom) the behaviour of the polarisation vector, of a neutron beam with λ = 0.25 nm and initially polarised in the z-direction, is given when coils 2 are energised. The small wiggles in the behaviour of the vector P are caused by the ratio \( \omega_G/\omega_L \) that is unequal to zero. The period of these wiggles depends on the wavelength of the neutron beam. When calculating the behaviour of P for a neutron beam, with \( \Delta \lambda \neq 0 \), these wiggles will average out (see bottom of fig. 2.8.1); upon transmission through the rotator the vector P of a neutron beam has been rotated almost perfectly from the z to the y-direction.

**Final Remark**

When two polarisation rotators, as described above, are put with their wire screens together, they can be used as a neutron spin flipper. Using this type of spin flipper the spin of the neutron can be flipped simultaneously for every wavelength present in the thermal neutron spectrum of the SP. This type of flipper was used in the experiments to determine the polarising power of neutron polariser using the 3p-2s method (see Por et al. [PKR94] or see chapter 8).
Chapter 3

Spin-Echo Coil System (SpECS) for 3d Neutron Depolarisation in High Magnetic Fields

3.1 Introduction

Up to the work described in this chapter, only three-dimensional (3d) neutron depolarisation (ND) experiments have been carried out in media which are in a non-magnetised or in a remanent magnetised state. Furthermore, 3d ND-experiments were also possible in media which are subjected to small magnetic fields, i.e. up to a few kA/m. If the applied field exceeds several kA/m, the net rotation of the polarisation vector, averaged over the wavelength spectrum of the beam results in additional depolarisation (see section 2.3.2). The component of the polarisation vector parallel to the direction of the applied field does not rotate. Hence, additional depolarisation of this component of the polarisation vector does not take place. The perpendicular component of the polarisation vector, however, rotates in the magnetic field and additional depolarisation of this component takes place. Eventually, if the magnetic field exceeds several kA/m, this component will fully depolarise. So, only 1d ND-experiments, i.e. with the polarisation vector parallel to the direction of the applied magnetic field, were possible in high magnetic fields. An example of 1d ND-experiments in high magnetic fields can be found in ref. [KMR95], where the domain size in CoCr-thin films has been measured as a function of the applied field.

In order to perform 3d ND-experiments in media which are subjected to high magnetic fields up to 400 kA/m (1 T = 796 kA/m), a special magnetic coil system was developed. This coil system, referred to as Spin-Echo Coil System (SpECS) henceforth, is based on the principle of neutron spin-echo [Mez72]. The SpECS will be described

in detail in section 3.2.

A computational study of the behaviour of the polarisation vector of a neutron beam while it passes through the system was carried out. The results of the computations are given in section 3.3. In section 3.4 these results will be compared with the results of the 3d ND-experiments performed in an empty SpECS using the complete thermal wavelength spectrum of the neutron beam of the SP.

3.2 Description of the SpECS

The SpECS consists of two identical coil systems, which are successively traversed by the neutron beam (see figure 3.1). One section, in the figure the right section, of the system is used to magnetise the sample. Note that in this figure the propagation direction of the beam is from left to right. Upon transmission through this section, the perpendicular component of the polarisation vector, with respect to the direction of the applied field, has been smeared out due to the wavelength distribution of the beam and the applied field in the system (see fig. 2.3). The second section is used to rotate the polarisation vector, for every neutron wavelength in the beam back to its original orientation, after transmission through the first section and sample. This is done by anti-parallel orientation of the field in the second section with respect to the first section. After transmission through the SpECS, the smearing effect has vanished and no additional depolarisation takes place (see fig. 2.3). In the next sections the various parts of the SpECS will be discussed.

Cu and Al Guide-field Coils

With the guide-field coil a relatively small magnetic field in the order of 3 kA/m is applied along the path of the polarised neutron beam. This field prevents depolarisation of the beam due to the magnetic stray fields in the system. Furthermore, at the entrance and exit of the SpECS, and at the boundary between the left and right sections this field provides a sudden change in the magnetic field. Note that this sudden change is also present when the field applied in the main-field coil is small. The polarisation of the neutron beam, with wavelengths ranging from 0.1 to 1 nm, which is either parallel or perpendicular to the direction of the applied field can not follow this sudden change adiabatically\(^2\). Hence the polarisation vector does not change its orientation, only the orientation with respect to the direction of the field might be reversed. This process occurs upon entering or leaving the SpECS. Here the direction of polarisation has not changed; the polarisation is no longer affected by the fields generated by the SpECS. At the boundary between the two sections, however, the orientation of the polarisation with respect to the direction of the applied field has been reversed. This transition is known as a so-called Majorana field flip [Maj32].

\(^{2}\)The polarisation vector is said to follow a gradual change in the orientation of B adiabatically, if the angle between the polarisation vector and direction of B remains the same (see appendix 2.B).
Figure 3.1: Schematic drawing of the Spin-Echo Coil System for 3d ND-experiments in the presence of high magnetic fields. The bottom-graph gives a side view of the system. The top-graph gives the cross-section view of the system according to the dashed line A-A. Note that the left and right section of the system are identical.
The guide-field coil is predominantly wound using copper wires. Where the neutron beam traverses the wires of the coil, aluminium is used instead of copper. The reason for this is to simultaneously minimise the absorption of neutrons in passing through the windings and to minimise the electrical resistance.

The top and bottom sides of the Al guide-field coil have a peculiar shape: the guide-field coil is wound such that a sample can be easily positioned in the system without removing the complete system from the SP depolarisation module.

Main-field Coils

To generate the desired magnetic field to magnetise the sample and to provide the compensating field, so-called split-pair coils are used, the main-field coils. The reason for using a split-pair coil is two fold: the sample has to be positioned in the coil from above and the neutron beam has to traverse the system without too much loss of intensity. This coil generates a magnetic field, at the position of the sample and in the absence of a yoke, of approximately 8 kA/m for a DC-current of 1 A. The main-field coil is cooled by water cooling. The system is protected against overheating by a bi-metal temperature sensor. This sensor switches the power supplies off if the temperature in the system exceeds 90°C.

Iron Yoke and Iron and Ferrite Cores

The guide-field coils and the main-field coils are positioned in an iron yoke, that short-circuits the magnetic flux so that the magnetic field outside the system remains zero. Furthermore, due to the presence of the yoke and the iron and ferrite core the magnetic field generated by the coils, at the position of the sample, is increased with respect to the case without yoke and core. The increase in field strength at the position of the sample, which can be represented by an 'amplification factor', can be calculated using Maxwell's first law

\[ \oint H \, dl = nI \]  

(3.1)

Assuming that the magnetic field \( H \) inside the yoke and inside the cores is zero, i.e. the relative permeability \( \mu_r = \mu/\mu_0 \) is much larger that unity, the amplification factor is simply given by the ratio of the sum of the total length of the core and the gap-width, and the width of the gap itself. In the SpECS the amplification factor is (20 cm/2 cm) = 10. It should be noted that if the magnetic field generated by the coil increases, the permeability of the core decreases. Eventually, if the core is magnetically saturated, i.e. \( B = B_s, \mu_r \) is equal to unity. Then, the amplification factor is equal to one, i.e. no amplification occurs.

The amplification factor decreases when the magnetic field generated by the coil increases. The effect of saturating the core is illustrated in figure 3.2. It shows the measured magnetic field \( H_4 \) at the position of the sample as a function of the current \( I_c \) through the main-field coils. The straight line represents \( H_a(I_c) \) as it would be when
no saturation occurs. It appears that it is almost impossible to increase the magnetic field $H$ above $\approx 450$ kA/m.

The core to 'amplify' the field and to short-circuit any flux emerging from the sample is made of soft iron and ferrite. The part of the core which is close to the sample is made of ferrite because the SpECS will also be used for time-dependent ND-experiments. In a time-dependent experiment the sample is magnetised with a short magnetic field pulse. In general this pulse generates in the core so-called eddy-currents. These currents will magnetise the sample inhomogeneously. Moreover, the magnetic energy of the field pulse is only partly used to magnetise the sample while the remaining energy is dissipated by the eddy-currents as thermal energy in the core itself. However, since ferrite is an electric insulator, the eddy-currents are suppressed. A disadvantage of using ferrite instead of soft iron is that the saturation induction of ferrite is much smaller than that of iron. As a result, the maximum applicable field is approximately 450 kA/m, nearly independent of the gap-width. To increase the maximum applicable field up to 1 T, the ferrite core has to be replaced by e.g a core made of a sandwich of 1 mm thick iron plates and non-conducting layers. This layered structure will suppress the eddy-currents in the time-dependent application of the SpECS.

At the time of writing this thesis, the core has been replaced and also the width of the gap has been decreased to 1 cm. In this configuration the maximum applicable field is in the order 1 T for a DC-current of 5 A. It appears that the polarisation of the beam is still larger than 0.4 for $\mu_0 H = 1$ T. To achieve this degree of polarisation, the width of the beam had to be decreased to 1.5 mm.
Compensation Coil

In reality, the two sections of the SpECS are not identical. To compensate for the differences between the two sections and to compensate the rotation of the polarisation vector due to transmission through the sample, the system is equipped with a compensation coil. This coil allows one to use the rotation compensation method (see section 2.4). It is wound around the main-field coil in one of the sections (in figure 3.1 the left section of the system). The sample is positioned between the ferrite cores in the other section.

Wire Screen

As mentioned before, the main-field coil is split in two parts. The magnetic field in the gap, i.e. position (c) in fig. 3.3, between the two parts is in the ±y-direction. However, at a position that is neither on the symmetry axis of the SpECS (dashed line in fig. 3.1) nor right within the gap, the field has also a component in the x-direction. This component increases as the distance to the symmetry axis increases. In a region with high magnetic fields, position (a) in fig. 3.3, the influence of the 'unwanted' x-component on the polarisation vector is negligible, because the polarisation vector can follow the change in orientation of the field adiabatically. In a region with low fields, position (b) in fig. 3.3, however, the polarisation vector can not follow this change adiabatically. On the other hand this change in direction takes place over a relative large distance, which means that it is too 'slow' for a so-called Majorana field flip polarisation change. Because the polarisation vector can neither follow the change in orientation adiabatically nor a Majorana field flip takes place, the change in orientation is not homogeneous over the cross-section of the beam. Hence, the beam will depolarise due to transmission through such a region. Such a region with low magnetic fields and relatively large x-components exists between the two sections of the SpECS. In order to compensate or suppress the x-component of the field a wire screen has been positioned between the two sections. This aluminium wire screen allows one to apply a magnetic field in the x-direction, such that the resultant field in the x-direction at every position in the neutron beam, which is in the vicinity of the wire screen, is (nearly) zero (see fig. 3.4). The construction of the wire screen is such that not only the 'unwanted' x-component of the field is suppressed but that at the same time the 'wanted' y-component of the field is increased (see fig. 3.3). The width of the wire screen is 6 mm. The total current through the windings is 80 A, i.e. the number of windings times the current per winding; then the x-components are suppressed and the field is predominantly in the y-direction over a distance equal to the width of the wire screen. Hence, the polarisation of the beam does not change upon travelling from the left to the right section of the system and the maximum beam width is 6 mm.
Figure 3.3: Schematic drawing of the magnetic field lines in the SpECS. For reasons of clarity the field lines due to the guide-field coils are omitted.

Figure 3.4: The solid lines give the calculated 'unwanted' x and 'wanted' y-component of the applied field along line A-A in fig. 3.3 as a function of the distance to the symmetry axis when the current through the wire screen is zero. The dashed lines give the magnetic field from the wire screen itself for a current at which the x-component of the field is almost perfectly suppressed. Note that the y-component of the field of the wire screen is not in the opposite direction.
Iron Screen Plate

The SpECS has to be positioned inside the SP depolarisation module. The space between the SpECS and the polarisation rotators (see fig. 2.A.1) is only a few centimeters. Magnetic stray fields emerging from the SpECS, exceeding \( \approx 0.1 \) kA/m at the position of the rotator, will disturb the action of the rotator. The polarisation vector does not rotate adiabatically from the \( z \)-direction towards one of the laboratory axes but to a direction determined by the field applied in the rotators and the stray fields of the SpECS.

It appears, both from numerical calculations and magnetic field measurements, that the magnitude of the stray field can be as large as 20 kA/m when the magnetic field in the gap, position (c) in fig. 3.3, is 400 kA/m. On the other hand, the magnetic field applied in the polarisation rotators is in the order of 2 kA/m. To decrease the magnitude of stray fields, iron screen plates are mounted onto the SpECS. These plates, with a thickness of 1.5 mm, short-circuit the magnetic stray fields, so that the fields are reduced to acceptable levels, i.e. less than 10 A/m. However, near the edges of the plates the magnetic field becomes inhomogeneous. If the region with an inhomogeneous magnetic field falls within the neutron beam, the beam will depolarise. To avoid depolarisation of the beam the distance between the plates is chosen such that no significant depolarisation occurs and that at the same time the stray fields are sufficiently suppressed. In the SpECS the optimal distance is found to be 20 mm. This is by chance similar to the width of the gap and to the distance between the polarisation rotator and the SpECS. Furthermore, it is much larger than the maximum applicable width of the beam as determined by the width of the wire screen, which equals 6 mm.

3.3 Numerical Calculations

The behaviour of the polarisation vector while the neutron beam passes through the SpECS is simulated. First, a computer program calculates, using Biot-Savart's law, the magnetic field at a position along the neutron path. For this purpose, the windings of the rectangular coils are approximated by four straight lines. The iron yoke, meant to short-circuit the flux, is described by magnetic field coils. These coils are positioned such that they increase the length, and at the same time decrease the 'demagnetising' factor, of the main-field and guide-field coils. The ferrite and iron cores are also described by magnetic field coils. The current through these 'coils' is chosen such that the magnetic field within the gap, position (c) in fig. 3.3, is increased by a factor of 10 with respect to the case when no cores are present. The field generated by the compensation coil is zero because in the computation the two sections are really identical. Figure 3.5 gives the calculated magnetic field along a neutron path that coincides with the symmetry axis of the SpECS.

Secondly, the behaviour of the polarisation vector of a neutron beam, having an infinitesimal small cross-section and a wavelength \( \lambda \), traveling through the system is
3.3. Numerical Calculations

![Graph showing magnetic field and number of precessions against position](image)

Figure 3.5: The measured (●) and calculated (solid line) magnetic field in the SpECS as functions of the position along the symmetry axis of the system. The DC-current through the main-field coils is 5 A, the current through the guide-field coils and wire screen is zero. The accumulated number of precessions of the polarisation vector, for $\lambda = 0.2$ nm, as a function of the position is given by the open diamonds (○). Note the zero of the horizontal scale is at the entrance of the left section of the SpECS.

Calculated by successive multiplication of rotation matrices. The elements of the rotation matrix are given by eq. (2.4) and by the magnitude and orientation of the magnetic field along the neutron path. Finally, to obtain the depolarisation matrix of a neutron beam with a realistic cross-section, i.e. a cross-section as in real experiments, the resultant rotation matrix for an infinitesimal small neutron beam is averaged over all such neutron beams that are within the cross-section of the real beam. In the computations and in the real experiments the cross-section ($y \times z$) is chosen to be equal to $(3 \times 8)$ mm$^2$.

In order to compare the results of the numerical calculations with the results of the experiments performed with the complete thermal neutron spectrum, the depolarisation matrix is averaged over the wavelength spectrum of the neutron beam (see inset of figure 2.4). Furthermore, the polarising power of the polariser and analyser has been taken into account. The polarising power of the polariser and analyser are assumed to be identical. The data has been taken from figure 2.2, the depolarisation of the beam while it passes through the polarisation rotators is assumed to be negligibly small. Hence, it will not have a significant effect on the calculated depolarisation and therefore it has been neglected.

To investigate the influence of the magnetic field generated by the wire screen on the polarisation of the beam, the elements of the depolarisation matrix were calculated for different applied fields $H_a$ within the gap for different currents through the wire
screen. The current through the wire screen has been increased from zero to maximum
current in steps of $1/4$ of the maximum current. The maximum current is defined as
the current at which the $x$-component of the field at the position of the wire screen
is almost perfectly suppressed. Hence, it will be proportional to the magnitude of the
applied field $H_a$ within the gap.

From the results of the computation it appears that the wire screen has a significant
influence on the polarisation of a neutron beam with a long wavelength, i.e. $\lambda > 0.5$ nm.
When the current through the wire screen is zero, the polarisation of the beam is
dramatically decreased and the orientation of the polarisation is even reversed. With
increasing current this effect vanishes. At perfect suppression of the $x$-component no
significant depolarisation is observed. This effect can easily be understood; a neutron
with a long wavelength spends a relatively long time in the region with a relatively
small $y$-component and a large $x$-component. The polarisation vector has changed
significantly upon traversing this region; the influence of the $x$-component is large.
When increasing the field of the wire screen itself, the $x$-component of the field decreases
and at the same time the $y$-component of the field increases. Then the neutron beam
experiences predominantly a field in the $y$-direction and no depolarisation or change
in orientation of the polarisation vector occurs. In contrast to neutrons with a long
wavelength, neutrons with a short wavelength $\lambda < 0.3$ nm spend a relatively short time
in this region. So, they do not have time to change their orientation of polarisation
significantly when traversing this region. Then applying a field in the wire screen does
not have a significant effect.

The experiments are performed using the complete thermal neutron spectrum of the
SP. In this spectrum (see figure 2.4) the mean wavelength is approximately 0.25 nm and
neutrons with a long wavelength are hardly present. So, the polarisation of the beam
is mostly determined by the neutrons with a short wavelength and the influence of the
current through the wire screen is not as dramatic as just described for neutrons with
long wavelengths. The results of the computations of the elements of the depolarisation
matrix are shown in figure 3.6. The dashed line represents the polarisation of the beam
when no $\text{SpECS}$ is present in the beam.

From the results it can be concluded that for the complete thermal neutron spec-
trum the polarisation of the beam can be increased when applying a field in the wire
screen. In the computation the wire screen is perfectly positioned, i.e. the position
at which the $x$-component of the field generated by wire screen itself coincides with
the 'zero', i.e. symmetry axis of the $\text{SpECS}$. In reality, it is possible that this con-
figuration is not met. To investigate whether such a displacement of the wire screen
has an influence on the polarisation of the beam, some additional computations were
carried. It appears that a displacement in the order of 1 mm results in a less effective
improvement of the polarisation of the beam when applying a field in the wire screen.
In the next section the results of the computations will be compared with the results of
experiments performed with an empty $\text{SpECS}$, i.e. no sample is present in the $\text{SpECS}$. 
3.3. **Numerical Calculations**

Figure 3.6: Calculated diagonal elements of the depolarization matrix ($D_{ij} = 0$ for $i \neq j$) as functions of the applied field in the SpECS when the current through the wire screen is equal to zero ($\circ$) and $1/4$ ($\triangle$), $1/2$ ($\bullet$), $3/4$ ($\circ$) and $1/1$ ($\square$) of the maximum current, i.e. the current at which the $x$-fields are almost perfectly suppressed. The dashed lines represent the polarisation without the SpECS in the beam. The depolarisation was calculated for the complete thermal spectrum of the SP and the cross-section of the beam was $(3 \times 8)$ mm$^2$. 
3.4 Comparison with the Experimental Results

To compare the calculated magnetic field as a function of the position x along the symmetry axis of the system with the real magnetic field applied, the field was measured along this axis at a DC-current of 5 A through the main-field coils. The current through the guide-field coils and the wire screen is zero. The results are given by the solid circles in figure 3.5. In the figure one can clearly see the saturation effect of the core; the field within the gap is significantly smaller than as calculated. This effect can partly be attributed to a smaller number of windings in the real system in comparison with the system simulated. Furthermore, the field is of equal strength within the gap between the cores; samples positioned in the gap are homogeneously magnetised. In figure 3.5, the number of precessions (\( \propto \int_0^x H(x')dx' \)) is given as a function of the position along the neutron path, for a neutron with a wavelength of 0.2 nm. For a field in the gap \( H_a \) in the order of 400 kA/m, the total number of precessions in one section is approximately 500. If the number of precessions in two sections differ only by a little as 0.1%, it results in a net precession angle of one precession. Then the beam will significantly depolarise due to additional depolarisation. Moreover, utilisation of the complete thermal neutron spectrum becomes impossible.

Figure 3.7 gives the three diagonal elements of the depolarisation matrix as a function of the applied field \( H_a \), when the rotation of the polarisation vector is compensated. To compensate the net rotation of the polarisation vector the rotation compensation method (see section 2.4) has been used prior to the measurement of the elements of each depolarisation matrix. The elements have been measured using the complete neutron spectrum of the SP (see inset fig. 2.4). The neutron beam was confined to a cross-section of \((3 \times 8)\) mm\(^2\). The non-diagonal elements are within \( \pm 5\% \) equal to zero. The deviation from zero is caused by the misadjustment of the polarisation with respect to the three orthogonal axes x, y and z. At high magnetic fields the polarisation of the beam is still larger than 0.65 but is significantly smaller than the polarisation at low magnetic fields. The decrease in polarisation, upon increasing the magnetic field, is probably caused by inhomogeneities in the magnetic field and/or a non-perfect suppression of the x-component of the magnetic field at the position of the wire screen. The latter may be a result of a small displacement of the wire screen or of the field generated by the wire screen being too small.

The measurement of the element \( D_{xy} \), with y the direction of the gap field, as a function of the current through the wire screen at different values for the applied field, shows that the polarisation of the beam increases, as one might expect, when increasing the current through the wire screen. However, this effect is not as significant as calculated. This is probably a result of a small displacement of the wire screen. Furthermore, it appears that at high magnetic fields the x-components of the field are not as perfectly suppressed as observed at low magnetic fields. To improve the performance of the wire screen at high magnetic fields, the current per unit width of the screen has to be increased. Note that an increase in the width of the screen results in a decrease of the derivative of x-component of the field with respect to the y-position.
Figure 3.7: The measured diagonal elements of the depolarisation matrix, for increasing (o) and decreasing (□) field, as a function of the field applied in the SpeCS. The cross-section of the poly-chromatic beam, with an average wavelength of 0.23 nm (see inset fig. 2.4), was confined to an area of (3 x 8) mm². The non-diagonal elements (not shown) are close to zero.
Unfortunately, an increase in the current through the wire screen is impossible, because this will lead to a break down of the screen caused by overheating. This will make the screen unreliable for using this while experimenting. On the other hand, increasing the thickness of the screen, i.e. increasing the number of windings in the propagation direction of the beam, may be disadvantageous because the path length with small magnetic fields increases also. In the search for an improved wire screen, a numerical calculation was carried out for candidate wire screens. The simulated wire screens consisted of wires and/or iron plates positioned outside the neutron beam to increase the x-component of the field generated by the wire screen itself. For this screen the y-component of the field of the wire screen is opposite to that of the main-field coils, and hence the magnitude of the field drop is decreased. It appears that, although the x-components are almost perfectly suppressed, the depolarisation of the beam upon traversing the wire screen has not vanished. From these simulations it became clear that an optimally designed wire screen should both decrease the x-components and increase the y-component of the field in order to avoid depolarisation of the beam. Nevertheless the SpECS, with the not optimally designed wire screen, has been used for the ND-experiments described in chapter 4, because the beam is still reasonably polarised for gap fields up to 0.5 T.
Chapter 4

Small Magnetic Particles

4.1 Introduction

In the past decades, powders of small magnetic particles have found many practical applications, e.g. for magnetic recording applications (CrO₂, γ-Fe₂O₃ and barium ferrite particles) and for the production of permanent magnets (barium ferrite particles). Therefore, understanding the relevant properties of those particle systems is very important. In particular, two of the most essential characteristics are the anisotropy field or switching field distribution and the particle-particle interaction. These properties can be investigated with the help of initial susceptibility and remanence curves [SBCW88].

More insight into the particle-particle interactions can be obtained by measuring the magnetic correlation length, i.e. the mean size of a magnetically correlated region, and the mean-square direction cosines of the local magnetisation as functions of the applied magnetic field and the (remanent) magnetisation of the particle system. In this chapter, the neutron depolarisation (ND) technique has been applied to study the magnetic interactions and correlations in barium hexaferrite particles, mainly. These particles, prepared by the glass-crystallisation method [MSG94, Mul94], are promising candidates for application in high-density magnetic recording media. The magnetic correlation length has been measured as a function of the remanent state of the particle system (see section 4.3).

The ND-experiments on the barium hexaferrite particles were performed before the completion and realisation of the SpECS (see chapter 3.1). Therefore, experiments in the presence of a magnetic field were not possible at that time. Meanwhile, the SpECS has been completed and tested (see chapter 3). This system has been used to perform ND-experiments in a Co-modified γ-Fe₂O₃ particle system. In the next sections the results of the magnetic field-dependent ND-experiments in γ-Fe₂O₃ particles and of the ND-experiments in barium hexaferrite particles in a remanent state after a pulsed magnetic field will be successively discussed.
4.2 Magnetic States Induced by Fields up to 400 kA/m\(^1\)

In this section the first three-dimensional neutron depolarisation (ND) experiment in the presence of magnetic fields up to 400 kA/m will be described. This new type of experiment was made possible by the realisation of the spin-echo coil system SpECS. At first the sample characteristics and some details of the experiment will be described. Thereafter, the results of the experiment are discussed. In the following, fields will be denoted by kA/m or by their corresponding magnetic induction \(\mu_0 H\) in T (1 T \(\equiv\) 796 kA/m).

4.2.1 Sample Characteristics and Experiment

The sample consists of elongated Co-modified \(\gamma\)-Fe\(_2\)O\(_3\) particles embedded in a solid matrix. This sample has been obtained by drying a well dispersed aged particulate dispersion. This dispersion was previously used in the second round of the CAMST Mobile Mill project (see chapter 5). The particles exhibit a spontaneous induction of 0.44 T [Bat91], have a length of 0.25 \(\mu\)m and an aspect-ratio of 10:1. The volume fraction \(\varepsilon\) of the randomly oriented and distributed particles is 28\%. The sample, with a total thickness \(L\) of 5.5 mm, is positioned between the ferrite cores of the first section of the SpECS. A detailed description of this system is given in chapter 3.1. The field was applied perpendicular to the propagation direction of the neutron beam. The maximum field is 400 kA/m \((\mu_0 H \approx 0.5\ T)\).

The rotation compensation method (see section 2.4) was used to adjust the current of the second coil in the SpECS, such that the polarisation is exactly rotated back to its initial orientation. Applying this method with and without a sample in the SpECS gives two compensation currents for the second coil. Their difference is proportional to the mean magnetisation \(\mu_0 M\) of the sample. The constant of proportionality was determined using a Ni calibration sample with known thickness and saturation magnetisation. By using a calibration sample with the same size and shape as the sample, one effectively corrects the measured magnetisation for the demagnetising effects. For the determination of the elements of the depolarisation matrix and the compensation current, the complete thermal neutron spectrum of the SP (see for a description section 2.3) was used.

When the rotation of the polarisation vector is compensated, the measured depolarisation matrix \(\hat{D}\) is diagonal. The magnetic correlation length \(\zeta\) and the mean-square direction cosines \(\gamma_i\) (\(i = x, y \text{ or } z\)) were calculated from

\[
\zeta = \frac{-\ln(\det(\hat{D}))}{c\lambda^2 \varepsilon L(\Delta B)^2}, \tag{4.1}
\]

\(^1\)This section is published in a modified form as: 'Magnetic Correlations between Co-modified \(\gamma\)-Fe\(_2\)O\(_3\) Particles in a Solid Matrix Studied by Neutron Depolarisation in Fields up to 0.5 T', P.T. Por, W.H. Kraan, J. Mes and M.Th. Rekveldt, J. Magn. Magn. Mater. (in press).
\[ \gamma_i = 1 - \frac{2 \ln(D_{ii})}{\ln(\det(D))} \] (4.2)

Here, \( c \) is the depolarisation constant equal to \( 2.18 \times 10^{29} \text{ m}^{-4} \text{T}^{-2} \), \( \lambda = 0.235 \text{ nm} \) the mean wavelength of the beam, \( \epsilon \) the particle filling fraction, \( L \) the thickness of the sample. \( \Delta B \) has been taken equal to the spontaneous induction \( \mu_0 M_s \) of the particles within the sample. \( \det(D) \) is the determinant of the depolarisation matrix; here it is simply the product of the diagonal elements. The diagonal element \( D_{ii} \) follows from the measured intensity using eq. (2.17). The neutron beam was confined to a size of \( (3 \times 8) \text{ mm}^2 \) and the total number of neutrons in a fully depolarised beam, i.e. the shim intensity, was typically 44000.

4.2.2 Results and Discussion

Figure 4.1 gives the reduced mean magnetisation \( m = M/M_s \) (top) and the magnetic correlation length \( \zeta \) (centre) as a function of the applied magnetic field. The bottom figure gives the mean-square direction cosine \( \gamma_\parallel \), as measured along the direction of the applied field, as a function of the applied field. In the current SpECS, magnetic fields \( |\mu_0 H| \) below 0.04 T were not accessible, because a field of this strength was necessary to guide the polarisation through the SpECS in order to avoid depolarisation. Furthermore, in this field region the actual position of the measuring points may have been shifted by a small systematical error in the determination of the magnitude of the applied field.

Prior to the measurements, the sample had been magnetised in a field of \(-0.55 \text{ T}\). The reduced saturation remanent magnetisation \( (M_r/M_s) \) is \(-0.55\). While the field is increased up to \(+0.55 \text{ T}\), \( \zeta \) initially decreases and reaches a minimum at the coercive field \( \mu_0 H_c = -0.07 \text{ T} \) (see the section of the curves that is marked with \( \Delta \)). Along this part of the hysteresis loop the correlations between particles within clusters of particles, as probed by the neutrons, are broken. At the coercive field \( \zeta \) equals \( \approx 50 \text{ nm} \), which is \( 2 - 3 \) times the mean particle size (for a randomly oriented uncorrelated particle system, consisting of elongated particles the mean particle size equals the diameter of the particle see ref. [Ros91, fig. 3.1]). This means that at \( m = 0 \) still positive correlations exist between particles. The increased value, with respect to the virgin state, for \( \zeta \) at \( m = 0 \) after magnetisation was also observed in previous experiments in remanent state (see e.g. ref. [RR91a]).

When further increasing the applied field, the magnetisation of each particle rotates towards the field direction and correlations between particles are induced. This results in an increase in \( m \) and \( \gamma_\parallel \). Furthermore, \( \zeta \) increases to a maximum of 300 nm (i.e. 12 - 15 particle diameters) for \( \mu_0 H = 0.15 \text{ T} \). At this value the sample starts to saturate and the \( \gamma_\parallel \) is close to one. Such an increased value for \( \zeta \) was also observed in ND-experiments in which the sample is in a remanent magnetised state. It is probably caused by fluctuations in the density of particles in the sample [RR91a]. The deviation of \( \gamma_\parallel \) from unity, for high magnetic fields, gives an estimate of the inaccuracy and/or
Figure 4.1: From top to bottom: the reduced mean magnetisation $m$, the magnetic correlation length $\zeta$ and the mean-square direction cosine $\gamma_\parallel$ as functions of the applied magnetic field. The curves marked by symbols have actually been measured. The solid lines without markers are obtained by an appropriate mirror operation.
systematical-error in the measurement of $\gamma_\parallel$.

At $\mu_0H = 0.40$ T, $\zeta$ starts to decrease significantly whilst $\gamma_\parallel$ and $m$ are little affected by the increased field. This effect has never been observed before and can be explained as follows. Assume that the sample consists of clusters of particles, which originate from variations in the particle density within the sample. The depolarisation is assumed to be caused by the clusters of particles. The depolarisation due to individual particles is negligibly small. Then each cluster can be considered as a 'quasi-particle' with a mean size equal to the cluster size. Its magnetisation is determined by the local density and orientation of magnetisation of the particles. The mean magnetisation of each 'quasi particle' is aligned along the direction of the mean magnetisation and hence $\gamma_\parallel$ is close to unity.

If the applied field exceeds the demagnetising field inside a cluster experienced by a specific particle, its magnetisation is forced into the field direction. So, the mean magnetisation of that cluster increases, and the mean magnetisation of that cluster remains along the direction of the mean magnetisation of the sample. Its size is hardly affected and $\gamma_\parallel$ remains close to one, because the mean magnetisation remains along the field direction. However, the fluctuation of the magnetisation $\Delta M$ around the average $\langle M \rangle$ decreases. The measured depolarisation, expressed in the correlation function $\xi$ (see eq. (2.7)), is proportional to magnetic induction fluctuations around the average. In general, the larger the fluctuations the larger the depolarisation. So, the decrease of $\Delta M$ means that the depolarisation $\langle \xi \rangle$ decreases. In order to calculate $\zeta$ one has to divide $\xi$ by $\langle B \rangle^2$; which has been taken equal to $(\mu_0M_s)^2$. The latter is for a system consisting of 'quasi-particles' larger than the real (unknown) value for $\langle B \rangle^2$. In this way one can qualitatively understand that $\zeta$ (see eq. (4.1)) decreases.

At high magnetic fields, all particles are aligned along the field direction and $\zeta$ should reach a plateau. In the present experiments no plateau at high fields is observed. Apparently a field of 0.55 T is not strong enough to saturate the sample. At the time of writing this thesis the SpECS is being modified in order to allow ND-experiments up to 1 T. This will enable one to check whether such a plateau exists and to verify the validity of the just described argument for the origin of the decrease in $\zeta$.

When relaxing the field from +0.55 T to zero, the opposite of the process described above occurs and $\zeta$ increases, remains constant and starts to decrease. At the same time $m$ decreases to +0.55. In this part of the hysteresis loop the correlations between particles, that were induced in the magnetisation process, are broken.

In conclusion, from the field dependence of the magnetic correlation length it appeared that the magnetic correlations originate predominantly from variations in the particle density within the sample. These variations can be considered as 'quasi particles' which magnetise differently as a function of the applied field.
4.3 Magnetic States Induced by a Pulsed Magnetic Field

In this section the results of the ND-experiments in barium hexaferrite particles are described. In contrast to the previous section, the particle system is in a remanent magnetised state after a pulsed magnetic field, i.e. no magnetic field is applied during the experiment.

Barium hexaferrite powders are used in manufacturing permanent magnets and will be used as high-density magnetic recording media. Both applications can be realised by employing the $\text{BaFe}_{12-2z}\text{Co}_z\text{Ti}_z\text{O}_{19}$ system. The maximum value for the coercivity $H_c$ is about 430 kA/m for a particle system with $z = 0$. Such a high value is convenient for permanent magnets, however it is too high for recording applications. By substitution of Fe by Ti and Co (or Sn) the coercivity can be decreased to about 80 kA/m which is required for magnetic recording applications [GSS90]. In the present study the magnetic correlations between the particles are studied for two different types of barium hexaferrites; the nano-crystalline ferrites (section 4.3.2) and the micro-crystalline ferrites (section 4.3.3). Both types of ferrite have a platelet hexagonal shape, characterised by a diameter $\delta$ and thickness $t$, and were prepared by the glass-crystallisation method [MSG94, Mul94]. The size of the platelets can be manipulated by adjusting the annealing time and temperature of the particles. The anisotropy direction of the particle magnetisation, i.e. the preferred orientation of the magnetisation, is perpendicular to the basal plane of the hexaferrite. At first the ND-experiment will be described in the next section. Thereafter, the results of the ND-experiments will be discussed separately for the two types of ferrites.

4.3.1 Experimental

A glass sample holder, filled with 'as-prepared' barium ferrite powder, was positioned in a magnetic yoke, in order to short-circuit any flux emerging from the sample. The sample holder and magnetic yoke were positioned in the sample box of the SP. Behind the sample holder, a magnetic coil was positioned in the beam in order to use the rotation compensation method (see section 2.4). In contrast to the previous section the neutron beam is monochromatised by Bragg-reflection at the (002)-planes of a pyrolytic monochromator crystal. The wavelength of the monochromatic beam was $(0.29 \pm 0.03)$ nm. The spectrum of the non-reflected neutrons, i.e. the remaining neutrons in the direct beam, was used to find the compensation current in the rotation compensation method.

To obtain the remanent states, the samples were magnetised with a short magnetic

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4.3. Magnetic States Induced by a Pulsed Magnetic Field

Field pulse applied in a magnetic coil wound around the sample. The power supply [Haa90] and the coil allowed to apply magnetic field pulses with an amplitude $H_a$ ranging between 13 kA/m and 790 kA/m, and with a pulse width at half maximum $t_{FWHM}$ of $\approx 200 \mu s$. The direction of the applied field was the $z$-direction, this is perpendicular to the propagation direction of the beam. The depolarisation was measured along two different static remanence curves:

- the *Isothermal Remanence Magnetisation* (IRM), $M_r(H_a)$ or $m_r(H_a)$; where $m_r$ is defined as the ratio between $M_r$ and the saturation magnetisation of the sample $M_s$. A magnetic field pulse is applied to an initially demagnetised sample, then the remanent magnetisation of the sample is determined using the rotation compensation method. Next, the elements of the depolarisation matrix are measured. This procedure is repeated, with successively increasing pulse amplitude $H_a$ until the maximum applicable field is reached.

- the *DC-Demagnetisation* (DCD) remanence, $M_d(H_a)$ or $m_d(H_a) = M_d(H_a)/M_s$. The DCD-remanence is obtained starting from the saturated remanent state, then the sample is magnetised with a short negative magnetic pulse field. After determination of the DCD-remanence and the elements of the depolarisation matrix, this procedure is repeated with successively increasing negative pulse amplitudes until the sample reaches its negative saturated remanent state.

From the two remanence curves and the so-called Henkel-plot [Hen64], information can be obtained on the nature and strength of the predominant magnetic interactions in the sample. Furthermore, they give a measure of the energy barrier distribution (i.e. switching field distribution) in the sample. In the absence of interactions the IRM and the DCD curves are related via the relation of Wohlfarth [Woh58]:

$$M_d(H_a)/M_d(\infty) = 1 - 2M_r(H_a)/M_r(\infty) . \quad (4.3)$$

For a particle system with particle interactions, Kelly et al. [KOMC89] developed a method in which a comparison is made between the left- and right-hand side of eq. (4.3). This method is known as the 'ΔM($H_a$)-curve'

$$\Delta M(H_a) \equiv M_d(H_a)/M_d(\infty) - [1 - 2M_r(H_a)/M_r(\infty)] . \quad (4.4)$$

In general, if $\Delta M > 0$ the interactions are *positive* and act in such a way that they promote the magnetised state, e.g. stacking of the barium ferrite platelets. On the other hand when $\Delta M < 0$ the interactions promote the demagnetised state or magnetisation reversal, e.g. the particles form clusters with anti-parallel particle magnetisation orientation. In this case it is said that the interaction between the particles is *negative*. In a real system both type of interactions are present but one of them is dominant.

The magnetic correlation length $\xi$ and the mean-square direction cosines $\gamma_i$ were calculated from the measured depolarisation using eqs. (4.1) and (4.2), respectively.
The thickness $L$ of the sample was between 3 and 8 mm in order to avoid complete depolarisation of the beam. The particle filling fraction $\varepsilon$ is approximately 25% for all samples. It should be noted that one has to substitute for $\varepsilon$ in eq. (4.1) the filling fraction of the 'magnetic' barium ferrite particles in the sample. This is not necessarily equal to the filling fraction of the particles, because the particles may consist of a non-magnetic or dead-magnetic layer [RGS91] which does not contribute to depolarisation. This layer with a thickness in the order of 1 nm has a great influence on the measured magnetic properties in particular for small particles, i.e. for the nano-crystalline barium ferrite samples. Such a non-magnetic layer not only strongly influences the particle interaction, but it also occupies a relatively large volume of the particle. In order to correct effectively for this layer, the following assumptions were made:

- the spontaneous induction $\mu_0 M_s$ of the barium ferrite particles is equal to the spontaneous induction of the bulk barium ferrite with the same composition.

- the volume fraction $\varepsilon$ of the magnetic ferrite barium particles, is equal to the particle filling fraction multiplied by the ratio between the saturation magnetisation of the particles $M_s$ and the bulk saturation magnetisation $M_{s\text{bulk}}$.

In the next section the results of the ND-experiments on the nano-crystalline barium-ferrite particles will be described.

### 4.3.2 Nano-crystalline Barium-ferrite Particles

#### Sample Preparation and Sample Characteristics

The barium hexaferrite powders $\text{BaFe}_{12-2z}\text{Co}_z\text{Ti}_2\text{O}_{19}$ with $z = 0$, 0.8 and 1.5 were prepared by the glass-crystallisation method at the Institut für Physikalische Hochtechnologie (IPHT), Jena (BRD). A $40\text{BaO-33B}_2\text{O}_3-27(\text{Fe}_2\text{O}_3+\text{CoO+TiO}_2)$ mol % glass was melted in air at $\approx 1400^\circ\text{C}$ in a Pt-crucible and was quenched between two rotating steel rollers to get amorphous flakes. These flakes were heat treated at temperatures between 580 and 780$^\circ\text{C}$ for different times (see table 4.1) which leads to crystallisation of the hexaferrite particles within a boron matrix. The ferrite particles were isolated by dissolving the remaining matrix with acetic acid. The slurry was resinized and dried. The glass-crystallised powders consist of single-crystalline particles and have a narrow particle size distribution. The size of the particles can be controlled by adjusting the annealing time and temperature. These parameters where such that the mean particle diameter ranges between 10 and 34 nm. More detailed information about the preparation is given in [MSG94].

The magnetic and geometric properties of the barium hexaferrites are given in table 4.1. The magnetic properties, such as the spontaneous magnetic induction $\mu_0 M_s$, saturation remanence $M_r/M_s$ and the coercivity $H_c$, were determined using a PAR vibrating sample magnetometer. In contrast to the unsubstituted ferrites and the ones with $z = 0.8$, the particles with $z = 1.5$ have a planar anisotropy direction, i.e the
4.3. Magnetic States Induced by a Pulsed Magnetic Field

Table 4.1: Magnetic and geometric properties of the nano-crystalline Barium ferrite sample (From R. Müller IPHT). The chemical composition of the particles is BaFe$_{12-2x}$Co$_x$Ti$_x$O$_{19}$.

<table>
<thead>
<tr>
<th>Batch number</th>
<th>$z = 0$</th>
<th>$z = 0.8$</th>
<th>$z = 1.5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t_{an.}$ [d]</td>
<td>10</td>
<td>10</td>
<td>7</td>
</tr>
<tr>
<td>$T_{an.}$ [°C]</td>
<td>580 600</td>
<td>580 600</td>
<td>640 640</td>
</tr>
<tr>
<td>$\mu_0 M$ for 1.5 T [T]</td>
<td>0.19 0.32</td>
<td>0.12 0.17</td>
<td>0.31 0.30</td>
</tr>
<tr>
<td>$\mu_0 M_s$ [T]</td>
<td>0.20 0.34</td>
<td>0.14 0.20</td>
<td>0.32 0.31</td>
</tr>
<tr>
<td>$M_r/M_s$</td>
<td>0.32 0.45</td>
<td>0.007 0.024</td>
<td>0.256 0.266</td>
</tr>
<tr>
<td>$H_c$ [kA/m]</td>
<td>101 213</td>
<td>0.95 1.83</td>
<td>31.1 38.8</td>
</tr>
<tr>
<td>$S_{BET}$ [m$^2$/g]</td>
<td>62 56</td>
<td>126 96</td>
<td>74 76</td>
</tr>
<tr>
<td>$\delta (t/\delta = 1)$ [nm]</td>
<td>21 23</td>
<td>10 14</td>
<td>16 16</td>
</tr>
</tbody>
</table>

Anisotropy is in the basal plane of the particle. The specific surface measurements (BET-method) were carried out on a Ströhlein Areamat. The geometric and magnetic properties were determined at IPHT. The mean geometric diameter of the ferrites was calculated from the specific surface $S_{BET}$, using the realistic assumption (from SEM-images) that the ratio between the thickness $t$ and the diameter $\delta$ of the hexaferrite particles is unity. The spontaneous magnetic induction $\mu_0 M_s^{bulk}$ of the bulk barium ferrite is equal to 0.47 T, 0.37 T and 0.31 T for $z = 0$, 0.8 and 1.5, respectively.

Results and Discussion

Figure 4.2 shows for the sample with batch number 2678 the quantities $\zeta$ and $\gamma_{II}$ as a function of $m_r$ and $m_d$, and $m_r$ and $m_d$ as a function of the applied field $H_a$. The statistical accuracy in the determination of these parameters can be estimated from the 'band-width' of the scattering in the data-points for the high field pulse region. The modified Henkel-plot or $\Delta M$-plot (see eq. (4.4)) is given in figure 4.3. Characteristic properties for all the samples studied are given in table 4.2. The modified Henkel-plot is, over the entire field range, predominately negative for all samples. In the discussion of the results the focus will first be on all samples excluding samples with batch number 2727 and 2880. Thereafter, the latter two samples will be discussed.

At $m_r = 0$, i.e. the virginal state, $\zeta$ exceeds the mean particle diameter as calculated from the specific surface measurements; i.e. $\zeta$ is one to seven particle diameters. Apparently, in the virginal state stacks of particles are present in the sample. The strong attractive force between the particles together with the perpendicular anisotropy causes the particles to form stacks of particles. The actual size of these stacks and the configuration of the particles within a stack is to a large extent determined by the size and shape (i.e. $\delta/t$) of the individual particles and the friction that they experience in order to form stacks. These stacks are likely to be formed in the manufacturing process of the
Figure 4.2: Results of the ND-experiments on batch number 2678: the reduced remanent magnetisation $m_r$ (a) and $m_d$ (b) as a function of the applied field $H_a$. The magnetic correlation length $\zeta$ and the mean squared direction cosine $\gamma_\parallel$ are given in (c and e) and (d and f) as a function of $m_r$ and $m_d$, respectively.
Figure 4.3: $\Delta M$-plot of batch number 2678 as a function of the applied magnetic field $H_a$.

Table 4.2: Characteristics of the nano-crystalline BaFe-samples determined by the ND-experiments at IRI.

<table>
<thead>
<tr>
<th>Batch number</th>
<th>$\delta_{BET} (t/\delta = 1)$ [nm]</th>
<th>$\zeta (m_r = 0)$ [nm]</th>
<th>$\zeta (\mu_0 H_a = 1 \text{ T})$ [nm]</th>
<th>$m_r (\mu_0 H_a = 1 \text{ T})$</th>
<th>$H_a (m_d = 0)$ [kA/m]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3015 2879</td>
<td>70 35</td>
<td>190 385</td>
<td>0.38 0.48</td>
<td>253 272</td>
</tr>
<tr>
<td>$x = 0$</td>
<td></td>
<td></td>
<td>$&lt; 20 &lt; 20$</td>
<td>$0.24 0.33$</td>
<td></td>
</tr>
<tr>
<td>$x = 0.8$</td>
<td></td>
<td></td>
<td>500 200</td>
<td>0.13</td>
<td></td>
</tr>
<tr>
<td>$x = 1.5$</td>
<td></td>
<td></td>
<td>115</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2727 2880 2678 2881 3025</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
particles, i.e. during the dissolving of the boron matrix, in which the particles have the opportunity to move. However, for the particles with planar anisotropy (batch number 3025) the magnetic correlation length approaches the mean diameter.

When the remanent magnetisation \( m_r \) increases, \( \zeta \) gradually increases to a correlation length which is much larger than the mean particle diameter. In the magnetisation reversal process \( \zeta \) decreases and subsequently increases. The correlation length is close to its minimum at the demagnetised state, i.e. \( m_d = 0 \). In the demagnetised state the correlation length is still a few times larger than in the virginal state (except for batch number 2879 c.f. section 4.3.3). Moreover, in the magnetisation reversal process the correlation length as a function of \( m_d \) is not symmetric around \( m_d = 0 \). The mean-square direction cosine \( \gamma_\parallel \) for the virginal state approaches a value of 1/4 as expected for a randomly oriented particle system. As the remanent magnetisation increases, \( \gamma_\parallel \) increases to \( \approx 0.75 \) at the saturation remanance. In the magnetisation reversal process, it gradually decreases and subsequently increases to \( \approx 0.75 \). The value for \( \gamma_\parallel \) at \( m_d = 0 \) is larger than in the virginal state. In the high field pulse region both \( \zeta \) and \( \gamma_\parallel \) are large, this indicates that the depolarisation is caused by fluctuations in the component of the magnetisation parallel to the mean magnetisation of the sample.

The observed magnetisation and magnetisation reversal process can be described as follows. Clusters of particles and/or clusters of stacks, which have no net magnetisation at \( m_r = 0 \), are magnetised when the remanent magnetisation increases. This results in an increased correlation length at the saturation remanent magnetisation. In the magnetisation reversal process the correlations between particles within a cluster or between stacks of particles are broken and the correlation decreases. The particles within a stack tend to have their magnetisation directions arranged anti-parallel (negative interaction). Hence, the correlation length is decreased. With increasing \( m_d \) the correlations are built up and the correlation length increases again. This process is similar to that in the magnetisation process.

For batch number 2727 and 2880 the depolarisation of the beam was too small to actually obtain \( \zeta \) and the \( \gamma \)'s from the depolarisation. From the measured depolarisation one can only derive an upper limit for \( \zeta \); \( \zeta \) remains smaller than 20 nm independent of the magnetised state of the samples. Furthermore, the remanent magnetisation is much smaller than in the other samples. Apparently, these particles are much smaller than the other samples, and are superparamagnetic at room temperature, which is the cause of the low remanent magnetisation and the small correlation length. This has been verified by Mössbauer spectroscopy experiments [Boe] on samples with batch numbers 2727 and 2879. The Mössbauer spectrum for batch number 2727 at room temperature shows a line-spectrum which is characteristic for a superparamagnetic particle. When reducing the temperature to 77 K, the spectra for both samples look similar and are characteristic for magnetically ordered particles, i.e. the superparamagnetic behaviour at a time scale of typically \( 10^{-9} \)s has disappeared. Note that for a thermal neutron the magnetisation of a particle is not flipped while it passes through the particle. Hence, the measured depolarisation is not influenced by the superparamagnetic behaviour of the particles as long as the particle is stable over a time scale of the order
of $10^{-11}$ s. Only the mean magnetisation is influenced by the superparamagnetic particles, because a measurement of the magnetisation of an ensemble of particles takes (milli)seconds rather than picoseconds. Surprisingly, the specific surface (or the mean particle diameter) differs not much from the other sample. Probably, a relatively larger part of the particle volume is non-magnetic due to the presence of the non-magnetic layer.

### 4.3.3 Micro-crystalline Barium-ferrite Particles

#### Sample Preparation and Sample Characteristics

The micro-crystalline barium hexaferrite powders $\text{BaFe}_{12-2x}\text{Co}_x\text{Ti}_{3-y}\text{Sn}_y\text{O}_{19}$ were prepared by the glass-crystallisation method [MSG94] at IPHT in Jena (BRD). In contrast to the nano-crystalline particles, some powders were prepared using an admixture of $\text{SiO}_2$. Electron microscopy observations show that the $\text{SiO}_2$ particles separate the ferrite particles. Furthermore, $\text{SiO}_2$ can change the particle diameter. The annealing time and temperature were such that the mean particle diameter ranges between 44 and 420 nm for the different samples.

The magnetic and geometric properties of the barium hexaferrite particles are given in table 4.3. The magnetic properties, such as the spontaneous magnetic induction $\mu_0M_s$, the coercivity $H_c$, and the saturation remanence $M_r/M_s$ were determined at IPHT. The mean diameter $\delta$ and the mean thickness $t$ have been obtained from transmission electron microscopy using a Jeol JSM 35 electron microscope.

<table>
<thead>
<tr>
<th>Batch number</th>
<th>mixed with SiO$_2$</th>
<th>without SiO$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>J45</td>
<td>D65</td>
</tr>
<tr>
<td>$x$</td>
<td>0.00</td>
<td>0.60</td>
</tr>
<tr>
<td>$y$</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>$\mu_0M_s$ for 1.5 T</td>
<td>[T]</td>
<td>0.43</td>
</tr>
<tr>
<td>$H_c$</td>
<td>[kA/m]</td>
<td>380</td>
</tr>
<tr>
<td>$S_{RET}$</td>
<td>[m$^2$/g]</td>
<td>11</td>
</tr>
<tr>
<td>$\delta_{TEM}$</td>
<td>[nm]</td>
<td>420</td>
</tr>
<tr>
<td>$t_{TEM}$</td>
<td>[nm]</td>
<td>147</td>
</tr>
</tbody>
</table>
Results and Discussion

Table 4.4 gives some characteristic properties of the micro-crystalline barium hexaferrite samples determined by the ND-experiments. For all samples the modified Henkel-plots, i.e. $\Delta M(H_a)$, are negative for the entire field range. This indicates that the interaction between the particles is such that it assists the demagnetised state.

The magnetic correlation length in the virginal state exceeds a few particle thicknesses. This means that also for the micro-crystalline ferrite particles the particles form stacks. During the magnetisation process $\zeta$ increases with increasing remanent magnetisation $m_r$, in a similar way as shown in fig. 4.2. At the saturation remanent magnetisation $\zeta$ is 3 to 10 times of that in the virginal state. In the magnetisation reversal process $\zeta$ initially increases and subsequently decreases. In the demagnetised state, i.e. for $m_d = 0$, $\zeta$ is much larger than in the virginal state. Moreover, it is of the order of the correlation length measured in the saturated remanent state. This is in contrast with the observations for the nano-crystalline samples. In these samples the correlation length is always smaller than the value observed in the saturated remanent state. Moreover, at $m_d = 0$ the correlation length for these samples is few times larger than the correlation length in the virginal state. Only for batch number 2879, the sample with the highest saturation remanence, is the behaviour of $\zeta$ as a function of $m_r$ similar to that of the micro-crystalline ferrites. It should be noted that in the magnetisation reversal process $\zeta$ is not symmetric around $m_d = 0$, which is also the case for batch number 2879.

The mean-square direction cosine $\gamma_\parallel$ approaches $1/4$ expected for a randomly oriented particles system, and increases as the remanent magnetisation $m_r$ increases. At the saturation remanence $\gamma_r$ is $\approx 0.6$. In the magnetisation reversal process $\gamma_\parallel$ decreases and subsequently increases to 0.6. In the demagnetised state, i.e. $m_d = 0$, $\gamma_\parallel$ is larger than in the virginal state.

<table>
<thead>
<tr>
<th>Batch number</th>
<th>mixed with SiO$_2$</th>
<th>without SiO$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>J45</td>
<td>D65</td>
</tr>
<tr>
<td>$d_{TEM}$</td>
<td>[nm]</td>
<td></td>
</tr>
<tr>
<td>420</td>
<td>62</td>
<td>44</td>
</tr>
<tr>
<td>$t_{TEM}$</td>
<td>[nm]</td>
<td></td>
</tr>
<tr>
<td>147</td>
<td>11</td>
<td>12</td>
</tr>
<tr>
<td>$\zeta(m_r = 0)$</td>
<td>[nm]</td>
<td></td>
</tr>
<tr>
<td>380</td>
<td>80</td>
<td>70</td>
</tr>
<tr>
<td>$\zeta(\mu_0H_a = 1 \text{ T})$</td>
<td>[nm]</td>
<td></td>
</tr>
<tr>
<td>1000</td>
<td>600</td>
<td>475</td>
</tr>
<tr>
<td>0.50</td>
<td>0.47</td>
<td>0.45</td>
</tr>
<tr>
<td>$\zeta(m_d = 0)$</td>
<td>[nm]</td>
<td></td>
</tr>
<tr>
<td>3000</td>
<td>1500</td>
<td>600</td>
</tr>
<tr>
<td>$H_a(m_d = 0)$</td>
<td>[kA/m]</td>
<td></td>
</tr>
<tr>
<td>250</td>
<td>150</td>
<td>90</td>
</tr>
</tbody>
</table>
4.4. Conclusion

The magnetisation and magnetisation reversal process can be described as follows. Clusters of particles and clusters of stacks are magnetised and form larger clusters or correlated regions when the remanent magnetisation \( m_r \) increases. This results in an increase in the measured correlation length which exceeds the correlation length measured in the virginal state. The \( \gamma \parallel \) is the mean-square direction cosine of the mean magnetisation of these clusters which are on average more aligned along the field direction. When the field pulse is reversed, i.e. in the magnetisation reversal process, the correlations within the clusters are not broken due to the strong interaction. Hence, the cluster reverses its magnetisation as a whole, whereby the correlation length does not differ much from the correlation length measured in the saturated remanent state. This is in contrast to the observations for the nano-crystalline ferrites. Apparently, the interactions between the nano-crystalline particles are smaller than in the micro-crystalline samples. This might be due to by the presence of a relatively thick non-magnetic layer for the nano-crystalline ferrites, which decreases the strength of the interactions. The interaction might also be smaller between the nano-crystalline ferrite particles because these particles are smaller than the macro-crystalline ferrite particles.

4.4 Conclusion

In this chapter the neutron depolarisation technique has been applied to study the magnetic correlations in small magnetic particle systems. For a sample consisting of Co-modified \( \gamma \)-Fe\(_2\)O\(_3\) particles embedded in a solid matrix the depolarisation was measured in the presence of magnetic fields up to 400 kA/m. The results indicate that the correlations originate from variation in the particle density within the sample. These variations can be considered as 'quasi-particles' that magnetise differently in magnetic fields.

For two types of barium hexaferrite particle systems, i.e. nano- and micro-crystalline particles, the depolarisation has been measured as a function of the remanent magnetisation after a pulsed field. For both the nano-crystalline and the micro-crystalline particle system the magnetic correlation length, in the virginal state, exceeds the mean particle size. This indicates the presence of stacks of particles. With increasing remanent magnetisation, correlations between clusters of stacks and/or clusters of particles are induced. For the nano-crystalline ferrites these correlations are broken in the magnetisation reversal process, whilst for the micro-crystalline ferrites the correlations are maintained. This difference is likely to be caused by the presence of a relatively thick non-magnetic layer at the basal planes of the nano-crystalline particles, which reduces the interaction between the particles. The interaction between the nano-crystalline ferrite particles may also be reduced because these particles are smaller than the macro-crystalline particles.
Chapter 5

Magnetic Particle Dispersions

5.1 Introduction

Particulate magnetic tapes are manufactured via a process that involves coating a suspension of magnetic particles in a binder solution onto a polymer substrate followed by drying and calendering [OGH91]. The suspension of particles, with a surfactant layer to prevent physical contact between particles, and binder solution is often referred to as a magnetic particles dispersion. It is known that, besides the magnetic properties of the particles itself, the microstructure of the dispersion has a large influence on the magnetic and recording characteristics of the final medium. Detailed and quantitative knowledge, and understanding the microstructure of the dispersion will help to understand the recording performance of the recording media.

In the framework of the CAMST (Concerted Action on Magnetic Storage Technology) Mobile Mill project, an experimental study of the magnetic properties of a dispersion of CrO$_2$ particles was carried out. In this 'large-scale' study, a large number of diverse techniques such as static and time-dependent magnetisation measurements, magnetisation measurements under shear, and neutron depolarisation (ND) were used. Because the techniques were based in different laboratories and because of the long-term instability of the dispersion of strongly interacting particles it is not possible to exchange samples between laboratories. This problem was solved by transporting a mill between laboratories in order to ensure that the measurements were made on fresh samples. The results of this experimental study are described in ref. [MOS93].

In the second round of the mobile mill project, Co-modified $\gamma$-Fe$_2$O$_3$ particles were used instead of CrO$_2$ particles. Distinct from the experiments performed in the first round of the project, the dispersion was magnetised in order to induce correlations in the dispersion so that ND can probe clusters of particles in the dispersion. A brief description of the dispersion preparation and characterisation is given in section 5.3. In section 5.4, the results of the ND-experiments performed in the second round of the project are given and discussed.

Recently, Coverdale et al. [CCHP93, CCHP94] reported on a computational study
of the three-dimensional microstructure of strongly interacting particulate dispersions using a force-bias Monte-Carlo scheme. In order to correlate the predicted microstructure by the Monte-Carlo scheme with the results of the ND-experiments, the ND-theory of Rosman [RR91b] has to be applied to the predicted microstructure in order to simulate the neutron depolarisation. In the next section the application of the ND-theory to the predicted microstructure in zero- and saturating field and in remanence will be described. First, a brief description of the Monte-Carlo scheme will be given.

5.2 Numerical Calculations

Description of the Monte-Carlo Scheme

In the computational study to investigate the three-dimensional microstructure of a particulate dispersion of 1000 elongated iron particles, the particles have a length of 0.25 μm and an aspect-ratio 10:1, and exhibit a bulk magnetisation of 1700 kA/m. Moreover, the particles are modelled as spherocylinders. Note that in the computational study iron particles are used rather than Co-modified γ-Fe₃O₃ particles which were used in the experimental study. The magnetostatic properties are approximated by a magnetic pole sited at the centre of each hemisphere. The computation cell, with a dimension of (1.05 × 1.05 × 1.05) μm³, is extended using periodic boundary conditions. The volume fraction ε of the particles in the cell is approximately 10%.

A combination of the magnetostatic potential with a "surface" potential to simulate the repulsive surface (surfactant) layer determines the behaviour of the particle in the cell. The interaction between the particles is effectively magnetostatic and attractive until the distance between the particles decreases to ≈ 6 nm. Then the surface potential begins to take effect, providing an effective barrier to prevent particle overlap. At equilibrium, a particle will tend to fluctuate at about 5 nm from its neighbours.

The particles are positioned in the computation cell by an algorithm that ensures a random spatial and angular distribution. A force-bias Monte-Carlo scheme [PRB78] is used to obtain an equilibrium particle configuration. This scheme employs the forces and torques derived from the magnetostatic and surface potential to influence the path of the particle in the cell.

During the computations, the particle ensemble is subjected to a cluster analysis which allocates particles with low mutual magnetostatic energy to a cluster. A cluster is then treated as a rigid body which may undergo small displacements and rotations in the cell under the Monte-Carlo scheme. A more detailed and comprehensive description of the Monte-Carlo scheme can be found in refs. [CCHP93, CCHP94].

5.2. Numerical Calculations

Description of the Depolarisation Simulation

The magnetic correlation length $\zeta$ and the mean-square direction cosines $\gamma_i$ (with $i = x, y$ or $z$) are calculated from the equilibrium position and orientation of the particles in the computation cell. In order to do so, it is convenient for particulate media to define a correlation matrix $\hat{\omega}$ such that it does not depend on the magnetic filling fraction $\epsilon$ of the particles in the computation cell. Moreover, it is also convenient that the correlation matrix has the dimension of length. The matrix $\hat{\omega}$ is defined as

$$\hat{\omega} \equiv \frac{2\hat{\alpha}}{\epsilon(\mu_0 M_s)^2} ,$$

(5.1)

where elements of $\hat{\alpha}$ are exactly equal to the matrix defined in eq. (2.6). The quantity $M_s$ is the saturation magnetisation of a particle. Then $\zeta$ and $\gamma_i$ can be written as (see section 2.2)

$$\zeta = \sum_i \omega_{ii} \quad \text{and}$$

$$\gamma_i = \frac{\omega_{ii}}{\zeta} .$$

(5.2) (5.3)

Using the definition of the matrix $\hat{\alpha}$ (see section 2.2) the expression for the element $\omega_{ij}$ of the correlation matrix $\hat{\omega}$ can be written as

$$\omega_{ij} = \frac{1}{\epsilon W 4\pi^2} \sum_{l=1}^{N} \sum_{k=1}^{N} \int_S V_i V_j m_i^l(\mathbf{s}) m_j^k(\mathbf{s}) F^l(\mathbf{s}) F^k(-\mathbf{s}) e^{i\mathbf{s} \cdot (\mathbf{r}^l - \mathbf{r}^k)} d^2 s - \frac{L_w}{2} \epsilon m_i m_j ,$$

(5.4)

with $L_w$ the thickness of the sample in the propagation direction of the neutron beam and $m_i$ the $i$-component of the reduced magnetisation $m$ ($\langle M \rangle = \epsilon M_s m$). $N$ is the number of particles in the computation cell, and $\mathbf{r}^l$ and $\mathbf{m}^l$ are the position and orientation of the magnetisation of a particle with volume $V_i$, respectively. $m_i^l(\mathbf{s})$ is a short-hand notation for the $i$-component of the vector $(\mathbf{s} \times \mathbf{m}^l \times \mathbf{s})$, and $F^l(\mathbf{s})$ is the particle form-factor of particle 'i', which is defined as

$$F^l(\mathbf{s}) = \frac{1}{V_i} \int_{V_i} e^{i\mathbf{s} \cdot \mathbf{r}'} d^3 r' .$$

(5.5)

In the ND-computations, the sphero-cylinders are approximated by homogeneously magnetised cylinders with length $2\mathbf{h}$ and diameter $2d$. The particle form-factor $F(\mathbf{s})$ for such a particle is given by [Kos79, pp. 233]

$$F(\mathbf{s}) = 2 \frac{\sin(s_{\parallel}\mathbf{h})}{s_{\parallel}\mathbf{h}} \frac{J_1(s_{\perp}d)}{s_{\perp}d} ,$$

(5.6)

with $s_{\parallel}$ and $s_{\perp}$ the components of the vector $\mathbf{s}$ parallel and perpendicular to the axis of the cylinder, respectively. The function $J_1$ is the Bessel-function of order one.
The last term in the right-hand side of eq. (5.4) corrects for the trivial change in polarisation due to the mean magnetisation of the sample (see section 2.2). The extension of the computation cell in the Monte-Carlo scheme using periodic boundary conditions, transforms the integral in plane $S$ into a summation over a set of discrete $s_y$ and $s_z$ values with intervals of $2\pi/L_y$ and $2\pi/L_z$, respectively. Here, $L_y$ and $L_z$ are the dimensions of the cell in the $y$ and $z$-directions, respectively.

Results and Discussion

Using the force-bias Monte-Carlo scheme and the above described ND-formulae, the magnetic correlation length $\zeta$ and the mean-square direction cosines $\gamma$ were calculated for three different magnetic states of the dispersion, i.e. for a dispersion

- without a magnetic field, referred to as virginal state henceforth;
- with a saturating applied field, referred to as saturated state henceforth;
- without a magnetic field after applying a saturating field, referred to as remanent state henceforth.

In order to obtain information on the 3D-micromagnetic state of the dispersion, the computation cell is rotated in the neutron beam in such a way that the mean magnetisation is successively parallel and perpendicular to the propagation direction of the neutron beam, denoted $n\parallel$ and $n\perp$, respectively. In the computations and in the ND-experiments the propagation direction is the $x$-direction. The results of the conducted simulations are given in table 5.1. In zero field, i.e. in the virginal state, no

<table>
<thead>
<tr>
<th></th>
<th>virginal</th>
<th>saturated</th>
<th>remanent</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\zeta$ [nm]</td>
<td>28</td>
<td>213</td>
<td>150</td>
</tr>
<tr>
<td>$\gamma_x$</td>
<td>0.48</td>
<td>1.00</td>
<td>0.99</td>
</tr>
<tr>
<td>$\gamma_y$</td>
<td>0.28</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>$\gamma_z$</td>
<td>0.24</td>
<td>0.96</td>
<td>0.93</td>
</tr>
</tbody>
</table>

dependence of $\zeta$ and $\gamma$ on the orientation of the computation cell was observed. The mean-square direction cosines $\gamma$ approach the theoretical values of $1/2$, $1/4$ and $1/4$, expected for an isotropically randomly oriented ensemble of particles (see section 2.2). Furthermore, the magnetic correlation length approaches the mean particle size, which is in agreement with the experimental results (see section 5.4). The mean particle size is defined as the length of a neutron trajectory within a particle averaged over all possible transmission angles of a neutron beam passing through that particle. For a
5.2. Numerical Calculations

randomly oriented elongated particle the mean particle size equals the diameter of the particle [Ros91, fig. 3.1]. In zero field the particles are randomly oriented as one would expect for a system that is not subjected to an orienting magnetic field. The apparent inconsistency between the random structure predicted by ND and the strong clustering evident from the analysis of the microstructure [CCHP93, CCHP94] can be resolved by considerations of the microstructure shown in figure 5.1. The ND-experiment is sensitive to correlations along the neutron path. The form of the aggregates is somewhat 'open', giving few magnetic correlations on average for any neutron path. So, this type of ordered microstructure is, for the neutron beam, indistinguishable from a real randomly oriented particle system.

In the saturated state of the dispersion, the particles form chainlike structures (see fig. 5.2). The mean-square direction cosine measured along the direction of mean magnetisation, is approximately equal to unity. The correlation length along the direction of magnetisation is slightly smaller than the length of a single particle. Apparently, the particle axes, i.e. the direction of magnetisation of the particles, are almost perfectly aligned along the direction of mean magnetisation of the dispersion. Moreover, the ‘bond-angle’ (for a definition see figure 5.3) approaches 180°. Furthermore, the particles form a zig-zag or spaghetti-type of chain (see fig. 5.3.b), in which the poles of two adjacent particles are connected side-by-side, rather than a linear-type of chain (see fig. 5.3.a). This spaghetti-type of chains may be due to the form of the potential used or it is a real effect. A distinction between these two cases (see fig. 5.3) can be made from ND-experiments in such particles systems. The correlation length measured perpendicular to the mean magnetisation equals the radius of the particles.

Figure 5.1: The zero field microstructure as obtained from the Monte-Carlo scheme.
Figure 5.2: The calculated microstructure in the saturated state as obtained from the Monte-Carlo scheme. The left figure gives a side-view of the computation cell and the right figure a top-view. The chains are aligned along the direction of the applied field.

Figure 5.3: When the propagation direction of the beam is along the direction of the chain, i.e. along the direction of the applied field $H_a$, the magnetic correlation length exceeds the particle length for a linear-type of chain (a). For a zig-zag or spaghetti-type of chain (b) the correlation length approaches the particle length. Equivalent magnetic poles are indicated by the black dot in the particles. The definition of the bond-angle $\beta$ is given in (c).
5.3. Experimental

When reducing the applied field to zero, i.e. the dispersion is in the remanent state, the particles are still aligned in chains. The correlation length along, and perpendicular, to the direction of magnetisation are smaller and larger respectively, than in the saturated state, suggesting that the bond-angle is some what smaller than that in the saturated state. The high degree of orientation, i.e. the alignment of particles in chains, is maintained in the remanent state.

5.3.1 Sample Preparation and Characterisation

In the second round of the mobil mill project the magnetic correlations in a dispersion consisting of Co-modified \( \gamma \)-Fe\(_2\)O\(_3\) particles were studied. The particles have a length of 0.25 \( \mu \text{m} \) and an aspect-ratio 10:1. The mill to produce the dispersion was set up apart from the ND set-up. The milling procedure can be summarised as follows: the first stage of the milling process involves thorough wetting of the iron particle surface. This was achieved by a solution of the wetting agent Lecithin in a 60/40 ratio of the solvent toluene and methyl-ethyl-ketone (MEK). The powder was then slowly stirred into the wetting agent solution. The pigment was wetted overnight. Then a solution of MEK and a hard vinyl binder resin was added to the wetted pigment. The solution was mixed using a sawtooth-blade mixer. At this stage of the milling process the first sample was extracted from the dispersion. The sample had a considerable high solid-ratio of \( \approx 50\% \) (the ratio of the dry to the wet dispersion weight). This high ratio is advantageous during the milling process; it optimises the efficiency of the dispersing and milling procedure.

During the milling process, using a bead mill, the second and third sample were extracted after 15 minutes and after one hour of milling, respectively. Following the one hour of milling, an MEK solution of urethane (soft binder) was added to the solution and mixed and milled for 30 minutes. As a consequence, of the addition of the MEK solution, the solid-ratio dropped to 35\%. After these 30 minutes, the fourth sample was taken from the dispersion.

The final stages of the dispersion process involved lowering the dispersion viscosity by the addition of MEK solvent. This has been done in three successive steps, in order to minimise the effects of possible aggregation of previously dispersed magnetic particles. After each addition of MEK, the dispersion was mixed and milled for 30 minutes, followed by extraction of the samples for the ND-experiments. In this stage of the milling process the fifth to the seventh sample were extracted. The solid-ratio for the final dispersion was approximately 28\%. Three additional samples were taken from the final dispersion in order to measure the effect of aging of the dispersion.

samples were characterised by ND one day, two days and two weeks after they were extracted from the mill.

The fresh samples (with a volume of $\approx 2$ cc) were measured immediately, after they were taken from the dispersion mill, in order to minimise the effects of possible aggregation and aging of the dispersion. Distinct from earlier ND-measurements on a CrO$_2$ particle dispersion [MOS93], the dispersion was subjected to magnetic treatments. In a non-magnetised sample consisting of separated randomly oriented and distributed particles the magnetic correlation length equals the mean size of a particle (see fig. 5.4). If the particles form aggregates, i.e. clusters of particles which exhibit negative

\[ \zeta \sim d_p \]

\[ \zeta \sim d_p \]

\[ \zeta \sim d_c \]

Figure 5.4: Left: for an ensemble of magnetically uncorrelated particles the measured correlation length $\zeta$ equals the mean diameter $d_p$ of a single particle. In a sample consisting of non-magnetised clusters of particles, which exhibit negative interactions, $\zeta$ also equals also $d_p$. Right: when the clusters are magnetised, and the particles exhibit positive interactions, the correlation length equals the mean cluster size $d_c$.

interactions, and the mean magnetisation of a cluster is zero, the correlation length for such a particle configuration also equals the mean size of a single particle. Hence, no distinction can be made between these two particle configurations.

In order to make ND sensitive to clusters of magnetically correlated particles, one has to induce magnetic correlations between the particles within a cluster. These correlations were induced by magnetising the dispersion. After magnetisation, the dispersion is demagnetised by shaking (called demagnetising henceforth). In doing so, the correlations between clusters, and between clusters and single particles will average out, because the clusters are not physically connected since they are surrounded by a repulsive surfactant layer. On the other hand, the correlations within a cluster are maintained, because the magnetic interaction between the particles is too strong to break by shaking. In the demagnetised state after shaking, the correlation length equals the mean size of the clusters in the dispersion.

It should be noted that during the milling procedure the solid-ratio changes significantly, and hence the viscosity of the dispersion. The ease of demagnetising the
sample by shaking may depend on the viscosity. In order to break or average out the correlations between clusters, the clusters have to migrate and/or to rotate. This process is more difficult in a viscous than in a diluted dispersion. So, to average out the correlations between clusters effectively, the necessary shaking time may depend on the viscosity of the dispersion.

5.3.2 Neutron Depolarisation

The ND-experiments on the magnetic particle dispersions were carried out on the SP (see for a description section 2.3). The wavelength of the monochromatic neutron beam was \((0.42 \pm 0.02)\) nm. For this purpose a pyrolytic graphite monochromator crystal was positioned in the beam.

The sample holders, with a thickness of 5 mm and freshly filled with dispersion, were placed in the neutron beam. In this state of the sample, referred to as virginal, the diagonal elements of the depolarisation matrix were measured. Next, the dispersion was magnetised with a short magnetic field pulse in order to induce correlations in the dispersion. The field pulse had a duration of \(\approx 150\) \(\mu\)s and an amplitude of 240 kA/m, which is much larger than the coercivity of the dispersion. Following, the demagnetisation of the sample by shaking, the polarisation of the beam was measured. Finally, the sample was magnetised for 40 seconds in a DC magnetic field of 32 kA/m, which is well below the coercivity. Again the sample was demagnetised by shaking and the diagonal elements were measured. The chosen magnetic treatments allows to distinguish between two mechanisms that induce correlations within the dispersion: correlations which are induced by switching of the magnetisation of a particle whilst the particle orientation remains unaltered, and correlations which are induced by physical rotation and migration of particles, so-called swinging.

Prior to the measurement of the diagonal elements, the non-diagonal elements were measured in order to check whether the samples were in a demagnetised state after shaking. In this case the mean magnetisation is equal to zero and the measured depolarisation matrix is diagonal. The non-diagonal elements were, within the systematic error in the adjustment of the polarisation vector, equal to zero. Confirming that the dispersion was indeed in a non-magnetised state during the ND-experiment.

The magnetic correlation length \(\zeta\) was calculated from the measured depolarisation using

\[
\zeta = \frac{-\ln(\text{det}(\hat{D}))}{c\lambda^2 (\mu_0 M_s)^2 \epsilon L},
\]

(5.7)

with the depolarisation constant \(c\lambda^2\) equal to \(38.5 \times 10^9\) T\(^{-2}\)m\(^{-2}\). The quantity \(\epsilon\) is the magnetic filling fraction of the particles in the sample; it can be calculated from the solid-ratio and the dispersion formulation/recipe [PKRM92]. \(\mu_0 M_s\) is the spontaneous magnetic induction of the \(\text{Fe}_2\text{O}_3\) particles; its numerical value is 0.44 T (from ref. [Bat91]). Equation (5.7) follows from eqs. (2.9), (2.10) and (2.13). \text{det}(\hat{D})
is the determinant of the depolarisation matrix, which is in this case the product of the diagonal elements \( D_{ii} \) (with \( i = x, y \) or \( z \)). The diagonal element \( D_{ii} \) follows from the measured intensity using eq. (2.17). The accuracy in the determination of the magnetic correlation length depends on the total number of neutrons counted in a fully depolarised beam and when the set-up operates in the '\( ii \)-mode'. The total number of neutrons counted in a fully depolarised beam was typically in the order of \( 7 \times 10^8 \). The time needed to measure the three diagonal elements in one magnetised state of the dispersion was approximately 30 minutes.

5.4 Results and Discussion

The magnetic correlation length \( \zeta \), measured after different magnetic treatments, as a function of the milling time, is given in figure 5.5.

![Figure 5.5: The measured correlation length \( \zeta \) as a function of the milling time for the virginal dispersion (○), after a pulsed magnetic field (◇) and after DC-magnetisation (△). The mean particle size is approximately 25 nm.](image)

In the virginal state, the correlation length is slightly above the particle size, and decreases as the milling process proceeds. Apparently, large clusters or networks of magnetically correlated particles are breaking during the milling process.

After the short field pulse, the magnetic correlation length has increased for samples drawn in the first hour of the milling process. The amplitude of the magnetic field pulse is high enough to switch the magnetisation of all particles into the field direction. However, the pulse is too short to rotate the particles in the clusters physically (swinging). When the pulse is over and the dispersion is demagnetised, the correlations
between the particles within the clusters are maintained. Hence, an increased magnetic correlation length is observed.

After applying a DC-field and demagnetising the dispersion by shaking, a dramatically increased correlation length is observed for samples drawn in the first hour of the milling process. As a result of the DC-field, particles with a switching field smaller than the applied field will switch their magnetisation into the field direction. Distinct from the pulsed field case, the particles have time to swing into the field direction; they will align along the field direction. Relaxing the magnetic field and shaking the dispersion, only the correlations within the clusters are maintained. Hence, an increased correlation length is observed. This increased correlation length is caused by the presence of clusters of particles which are not destroyed by demagnetisation. However, swinging and switching can not alone explain the dramatically increased correlation length observed in the first stages of the milling process. Apparently, the clusters and/or separated particles have time to migrate due to the long range interactions between these magnetic entities. The migrated clusters and particles form large chains oriented in the field direction. The size of such chains is determined by the extension and strength of the interaction. This depends on the size of the clusters; the larger the size the larger the range of interaction and, moreover, the less effective the repulsive potential of the surfactant becomes. The interaction between clusters in the chains is strong, because the extension of the magnetic interaction is larger than the range of the repulsive force of the surfactant layer around the clusters. Hence, after demagnetising, the chains are maintained and the correlation length increases dramatically. After applying a DC-field of a strength and duration different from the values mentioned above, the results of the preliminary measurements showed that the correlation length depends strongly on the field strength and duration of the field. As the milling process proceeds before diluting the dispersion, the effect of both the pulsed, and DC-field on the magnetic correlation length becomes less dramatic. The interaction between the clusters is not strong enough to form large chains and to maintain these chains after demagnetisation. This means that the clusters or networks of particles are breaking, as a result of the milling procedure.

When the dispersion is well dispersed, i.e. consisting of separated particles surrounded by surfactant, the magnetisation of the particle may change either by switching or swinging into the field direction in response to the pulsed- or DC-field treatment. The magnetic treatments create large networks or chains of particles aligned in the field direction. Relaxing the applied field and demagnetising the dispersion, the networks and chains are broken into individual particles surrounded by surfactant. This occurs because in the final stage of the milling process the particles are not physically connected and in addition the magnetic interaction between particles is lowered by the surfactant around each particle. The extension of the magnetic interaction is more or less equal to the extension of the repulsive interaction. So, it is not strong enough to maintain these networks and chains. In this demagnetised state the magnetic correlation length equals the mean particle size, independent of the magnetic treatment followed by demagnetisation of the dispersion.
After 90 minutes of milling no significant changes in the correlation length after different magnetic treatments are seen; the correlation length is in the order of the mean diameter of the particles. In other words, the dispersion remains well dispersed after dilution in the final stages of the milling process.

More samples were drawn from the final dispersion, one and two days, and two weeks after preparation. This state of the dispersion is referred to as aged. The magnetic correlation length was measured after different magnetic treatments, as functions of the age of the dispersion. The results of these experiments are shown in table 5.2. From table 5.2, no significant change in the magnetic correlation length, after different magnetic treatments, is observed. Apparently, the dispersion remains stable and well dispersed, i.e. no significant re-aggregation of the particles occurs, over a period of two weeks.

5.5 Conclusion

For a magnetic particle dispersion, the magnetic correlation length was determined by neutron depolarisation. In order to make ND sensitive to clusters of magnetically correlated particles, the dispersion was magnetised by a magnetic field pulse and a DC magnetic field. From the measured correlation length, after demagnetisation by shaking, it appeared that clusters or networks of particles are breaking in the first 90 minutes of the milling process.

In the final stage of the milling process, i.e. the addition of MEK in order to obtain an optimal viscosity for coating, the correlation length remains constant. Furthermore, no dependence of the correlation length on the magnetic treatment is observed. This means that diluting the dispersion causes no re-aggregation of previously dispersed particles.

The results of the ND-experiments in the 'aged' dispersion indicate that the dispersion remains stable and well dispersed over a period of two weeks.

The results of the numerical study of the three-dimensional microstructure of a particulate dispersion in zero field, using a force-bias Monte-Carlo scheme, are in agreement with the experiments; the correlation length approaches the mean diameter of a particle. The results of the numerical study of dispersions that are in a magnetised state,
can not be compared with the experimental data. In the experiments the dispersion was magnetised followed by demagnetisation by shaking. This demagnetisation procedure is not simulated in the computations. To verify also the correctness of the model in magnetised media, ND-experiments have to be carried out in such media. Using carefully chosen samples and due to the recent developments in the application of ND in magnetised media, using the SpECS and the rotation compensation method and the gain in intensity in the last year, these experiments are easy to perform in a limited amount of time.
Chapter 6

Metal-Evaporated Tape

6.1 Introduction

Metal-Evaporated (ME) tape, manufactured by oblique evaporation [FM84], attracted major interest as high-density recording tape, and is now being used in high-band 8mm video tape. In the near future, excellent recording characteristics over a wide frequency will be necessary for high-performance recording tapes in order to use them for new recording systems, for instance High Definition TeleVision (HDTV) systems and/or fast information/data-transfer systems. Co$_{80}$Ni$_{20}$ tapes, obliquely evaporated in the presence of an oxygen atmosphere, comply to these requirements and are therefore nowadays used in 8mm video systems.

Usually, thin-film recording media are investigated by means of for instance TEM- and SEM-micrographs, magnetisation measurements and torque magnetometry to analyse the crystallographic and magnetic properties of these materials. Using the 3D Neutron Depolarisation (ND) technique, information can be obtained on the magnetic structure, i.e. magnetic domain width and height, and mean domain magnetisation direction in the bulk of the tape. This method has previously been applied in the study of the magnetic domains in CoCr thin films with perpendicular anisotropy [HLRK84, KMRL88, KRHL93].

In this chapter the ND-technique is applied to determine the magnetic structure in a commercially available two-layer metal-evaporated tape. The results of these experiments are given in section 6.4. Furthermore, they will be discussed using the results of the numerical computations performed in section 6.3. First the sample preparation and characterisation and a brief description of the ND-experiment are discussed in the next section.

6.2 Experimental

6.2.1 Sample Preparation and Characterisation

The sample studied is a commercially available metal-evaporated TDK Hi8 videotape, manufactured by oblique evaporation of Co$_{80}$Ni$_{20}$ in the presence of an oxygen atmosphere. The tape consists of two sub-layers, which are evaporated in two consecutive runs. Its total film thickness $h$, determined by Transmission Electron Microscopy (TEM) at the Philips Research Laboratory (PRL) in Eindhoven (NL), is 180 nm. As a result of the oblique evaporation process, each sub-layer has a tilted columnar structure and therefore the anisotropy direction is inclined with respect to the film plane. The two layers have a column orientation according to figs. 6.1 and 6.3. The morphology of the tape has been determined by cross-section TEM (see fig. 6.3). The chemical composition of the tape has been determined as a function of the depth by means of Auger (AES) depth profiling at PRL; its average composition is found to be Co$_{72}$Ni$_{16}$O$_{12}$.

The magnetic properties of the tape were determined by a biaxial vibrating-sample magnetometer (VSM) at PRL [BC91]. The product $M_s h$ (after correction for demagnetising fields) is equal to (66.6 ± 3) mA, the in-plane coercive field $H_c$ equals 114 kA/m and the remanence after in-plane magnetisation $M_r / M_s$ is 0.85.

The depolarisation of the beam, measured in a single tape, can be estimated from the $M_s h$ determined by VSM-measurements. Assuming that the neutron beam, with $\lambda = 0.29$ nm, traverses the tape parallel to the orientation of the columns in one layer and the depolarisation is only caused by this layer, the polarisation will only differ from unity by $\approx 3 \times 10^{-5}$. To enhance the depolarisation effect, a stack of 448 tapes was used instead of a single tape.

Two ND-experiments were performed on this tape; one on an 'as-received' tape, i.e. without a magnetic treatment prior to the ND-experiment. The second experiment was performed on a tape that is in a remanent state after magnetisation in a magnetic field of 800 kA/m applied perpendicular to the plane of the tape. To avoid a net rotation of the polarisation vector, the stack was split, prior to the magnetisation treatment, into two stacks of 224 tapes. The stacks were magnetised in opposite directions, and joined after the magnetic treatment. Then the sample exhibits no net magnetisation, and the depolarisation matrix will be diagonal. Furthermore, a possible asymmetry in the depolarisation as a function of $\theta$ is retained. The asymmetry, if present, may be a result of a difference in orientation of the domain magnetisation, thickness of each sub-layer, and magnetisation of each sub-layer.

6.2.2 Neutron Depolarisation

The ND-experiments in the ME-tapes were carried out on the SP (see for a description section 2.3). The wavelength of the monochromatic neutron beam was equal to (0.29 ± 0.03) nm.

The correlation function $2\xi h$ is used as a measure for the depolarisation since it can
easily be compared with the results of the numerical calculations. Moreover, no a-priori knowledge of the magnetic properties is needed to calculate \( 2\xi h \) from the measured depolarisation. The correlation function \( 2\xi h \), normalised to a single tape, follows from the measured depolarisation using

\[
2\xi h = \frac{-\ln(\det(\hat{D})) \times \cos(\theta)}{448 \times c \lambda^2}, 
\]

with the depolarisation constant \( c \lambda^2 \) equal to \( 18.33 \times 10^9 \text{T}^{-2}\text{m}^{-2} \). Equation (6.1) follows from eq. (2.13) by substituting \( L = 448h / \cos(\theta) \). The \( \cos(\theta) \) in the numerator of eq. (6.1) corrects for a trivial change in the transmission length upon changing the transmission angle \( \theta \). The factor 448 in the denominator normalises the measured depolarisation to a single tape; \( \det(\hat{D}) \) is the determinant of the depolarisation matrix.

In the ND-experiments performed on the ME-tapes, no net rotation of the polarisation vector occurs and the depolarisation matrix is diagonal; then \( \det(\hat{D}) \) is simply the product of the diagonal elements \( D_{ii} \) (with \( i = x, y \) or \( z \)). The diagonal element \( D_{ii} \) follows from the measured intensity using eq. (2.17).

In order to determine the magnetic domain size and domain magnetisation orientation in the ME-tape, the sample is rotated over an angle \( \theta \) (with \(-60^\circ \leq \theta \leq 60^\circ\)) around the z-axis. This axis lies in the plane of the film and is perpendicular to the running direction, i.e. the longitudinal direction, of the film. The magnetic layers of the ME-tape studied are evaporated onto a substrate, in this case an approximately 10 \( \mu \text{m} \) thick PET-film. The shim intensity of the beam after inserting the sample has decreased by a factor of five at perpendicular transmission \( (\theta = 0^\circ) \) and by a factor of twenty-five at \( \theta = \pm 60^\circ \). Due to the large change in the beam intensity and a wavelength-dependent transmission, the spectrum of the beam might have changed with respect to the case without sample. In general, the polarisation of an empty beam \( P_{ii}^0 \) depends on the wavelength (see fig. 2.2). Hence, a change in the neutron spectrum, or a shift of the mean wavelength, results in a change in polarisation of an empty beam. To compensate effectively for this effect, the polarisation of an empty beam is determined using a non-magnetic dummy sample. The thickness of this perspex dummy sample is chosen such that its transmission is equal to the transmission of the stack of 448 tapes. The cross-section of the beam was \((8 \times 6) \text{ mm}^2\); it has been confined to this size by means of the two diaphragms of the SP depolarisation module. The divergence of the beam was 8 \( \text{mrad} \) and 6 \( \text{mrad} \) in the y and z-direction, respectively. At perpendicular transmission the number of neutrons counted in the shim was typically \( 5 \times 10^5 \); then the statistical accuracy in the determination of the diagonal elements was 0.05%.

In order to determine the magnetic domain size and domain magnetisation orientation in the tape from the measured \( 2\xi h \), the correlation function \( 2\xi h \) has to be calculated as a function of \( \theta \) using the formulae given in section 2.2 and a model for the magnetisation distribution \( M(r) \) in the tape. The calculation of \( 2\xi h \) is repeated, for different models for the magnetisation in the tape, until the calculated \( 2\xi h \) is in good agreement with its measured counterpart. Before discussing the results of the ND-experiments,
the computation method will be described and the results of the computation are given in the next section.

6.3 Numerical Calculations

Description of the Computation

The computation of the correlation function $2\xi h$ is based on the eqs. (2.6) and (2.15) which relate the magnetisation distribution, via eq. (2.8), to the measured depolarisation. In order to perform the computations, one has to model the magnetisation distribution in the tape. First, the model for the magnetisation distribution and the computation method will be described for a transmission angle $\theta$ equal to zero, i.e. the coordinate system $x'y'z'$ of the tape itself coincides with that of the laboratory ($xyz$); $M(r) = M(r')$. Thereafter, the computation method will be generalised to the case $\theta \neq 0$.

In the model for $M(r)$, the magnetic domain structure in each sub-layer is described by a checker-board like structure of magnetic domains (see fig. 6.1). The slightly curved columns (see fig. 6.3) are approximated by straight columns. The magnetisation in each sub-layer is oriented at an angle $\psi_1$ and $\psi_2$; the domain height and lateral domain widths (in Longitudinal and Transverse direction) are $h_1$, $\delta^L_1$, $\delta^T_1$ and $h_2$, $\delta^L_2$, $\delta^T_2$, respectively. Furthermore, the magnetic domains are assumed to be homogeneously magnetised, i.e. no regions with a magnetisation which are higher or lower

Figure 6.1: Left: model of the magnetisation distribution in the tape. Right: top view of the model of the magnetisation distribution and the coordinate system used in the computations.
6.3. Numerical Calculations

than the average magnetisation are present; the domain magnetisation is equal to \( M_s \). The magnetisation \( M_s \) is found by magnetisation measurements performed in a high magnetic field.

The magnetisation distribution \( M(r) \) is periodic in the yz-plane of the film; the domain period in the y (longitudinal)-direction and in the z (transverse)-direction is \( 2\delta^L \) and \( 2\delta^T \), respectively. The periodicity of the modeled domain structure can be used in the calculation of the Fourier transform \( B(s) \) of \( M(r) \) (see eq. (2.8)). For this purpose, the position vector \( r \) can be rewritten as

\[
r = r'' + 2g\delta^L i_y + 2l\delta^T i_z,
\]

with \( r'' \) the position vector in a representative sample volume \( W'' \) \((= 2\delta^L \times 2\delta^T \times h)\). \( g \) and \( l \) are integers and \( i_y \) and \( i_z \) are the unit vectors for the y and z-coordinate axes, respectively. Substituting eq. (6.2) in (2.8) and the resulting expression for \( B(s) \) in eq. (2.7), the expression for the correlation function \( 2\xi h \) can be written as

\[
2\xi h = 2h \frac{8\pi^4}{W''} \int_S d^2 s |F_p(s)|^2 S_p(s), \tag{6.3}
\]

with

\[
|F_p(s)|^2 = B''(s) \cdot B''(-s), \tag{6.4}
\]

\[
S_p(s) = \frac{\pi^2}{\delta^L \delta^T} \sum_{n=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \delta(s_y - n\frac{\pi}{\delta^L})\delta(s_z - m\frac{\pi}{\delta^T}), \tag{6.5}
\]

and

\[
B''(s) = \frac{\mu_0}{(2\pi)^3} \int_{W''} d^3 r'' (\mathbf{\hat{s}} \times \mathbf{M}(r'') \times \mathbf{\hat{s}}) e^{i\mathbf{s} \cdot \mathbf{r}''}. \tag{6.6}
\]

In the argument of the integral over the plane \( S \), the factor \( S_p(s) \) may be seen as the structure factor of the domains; it describes the arrangement of the domains in the yz-plane of the film. \( |F_p(s)|^2 \) is the form-factor squared of the representative sample volume itself. The product of \( |F_p(s)|^2 \) and \( S_p(s) \) is proportional to the intensity that one would measure in a SANS-experiment. Then the arbitrary vector \( s \) is the neutron wave vector change \( \kappa \). Substitution of eq. (6.5) into eq. (6.3) transforms the integration in the \( S \)-plane to a summation over a set of discrete \( s_y \) and \( s_z \) values with intervals of \( \pi/\delta^L \) and \( \pi/\delta^T \), respectively.

When rotating the sample around the z-axis over an angle \( \theta \), the coordinate system \( x'y'z' \), and hence \( M(r') \), is rotated in the laboratory system xyz. The x, y and z-coordinates can be expressed in the \( x' \), \( y' \) and \( z' \) coordinates using

\[
x = x' \cos(\theta) - y' \sin(\theta),
\]

\[
y = y' \cos(\theta) + x' \sin(\theta)
\]

and

\[
z = z'. \tag{6.7}
\]
The magnetisation distribution \( M(x') \) within the sample itself can be expressed in the \( M(x) \), as seen in the laboratory system, using

\[
\begin{align*}
M_x(x) &= M_x(x') \cos(\theta) - M_y(x') \sin(\theta), \\
M_y(x) &= M_y(x') \cos(\theta) + M_x(x') \sin(\theta) \quad \text{and} \\
M_z(x) &= M_z(x').
\end{align*}
\]

Furthermore, one should bear in mind that the integration plane \( S \) remains perpendicular to the propagation direction of the neutron beam. As a result of this coordinate system transformation, \( \delta^L \cos(\theta) \) in eq. (6.5) has to be replaced by \( \delta^L \cos(\theta) \). In the next section the results of the computation for a single-layer and a two-layer tape will be discussed.

Results of the Computation

To gain insight in the dependence of the measured depolarisation on the domain structure, \( 2\xi h \) is calculated and discussed for three different magnetic domain configurations. In these computations the domain magnetisation orientation \( \psi_1 = \psi_2 = 44^\circ \) and the domain magnetisation \( M \) is equal to 380 kA/m. The longitudinal and transverse domain size equals the thickness of the magnetic layer; i.e. 90 nm. These parameters are chosen such that they describe approximately the magnetic domains of the tape studied.

The top figure in fig. 6.2 gives the calculated correlation function \( 2\xi h \) as a function of \( \theta \) for a single-layer tape with the domain sizes as mentioned above. It appears that the angle at which \( 2\xi h \) has a maximum coincides with the orientation of the domain magnetisation, i.e. \( \theta_0 = \psi_1 \). Moreover, the absolute value of this maximum is equal to \( (\mu_0 M_s)^2 h^2 / \cos \theta_0 \). The full width at half maximum (FWHM) of \( 2\xi h \) is proportional to the ratio of the domain width \( \delta^L \) and the domain height \( h_1 \); the smaller the domains are, the narrower \( 2\xi h \) around \( \theta = \theta_0 \), and the smaller \( 2\xi h \) at \( \theta = 0 \). It should be noted that only the size of the magnetic domains, perpendicular to the axis around which the tape is rotated, can be determined. Hence, the results of the computation do not depend on the size of the magnetic domains in the transverse direction, i.e. \( \delta^T \). If the domain size in the transverse direction has to be determined, the sample has to be rotated around the y-axis.

In the next computation, \( 2\xi h \) is calculated for a two-layer tape with equal domain sizes in both sub-layers but with opposite domain magnetisation orientation (see inset figure 6.2). The domain sizes are equal to those in the previous calculation. It has been assumed that no correlation exists between the domain structures in the first and second layer; the tape can be considered as a tape consisting of two independent sub-layers. Then the correlation function \( 2\xi h \) is simply the sum of the correlation functions calculated for the two separate layers. The results are given in the centre figure of fig. 6.2. By analogy to the single-layer tape, the position at which \( 2\xi h \) has a maximum yields the orientation of the domain magnetisation, the thickness of each sub-layer follows from the absolute value of the corresponding maximum. The latter is only valid
Figure 6.2: The calculated correlation function $2\xi h$ as a function of the transmission angle $\theta$ for a single-layer tape (top), for a tape consisting of two sub-layers which are magnetically uncorrelated (centre) and magnetically correlated (bottom). The inset of each figure gives a schematic drawing of the domain magnetisation distribution within the tape.
if the contribution of the other sub-layer to the correlation function is negligible for this angle \( \theta \) (see figure 6.2). FWHM for \( \theta = \psi_{1,2} \) is proportional to the ratio of the width and the height of the domains in each sub-layer.

The calculation of \( 2 \xi h \) has been repeated for a two-layer tape as described above, but in this case the magnetic domain structure in the first and second layer are correlated; the tape can no longer be considered as a tape consisting of two independent layers. The results are given in the bottom figure of fig. 6.2. For this specific domain configuration, the behaviour of \( 2 \xi h \) at transmission angles \( \theta \) in the vicinity of \( \pm \psi_{1,2} \) is identical to that for a tape consisting of two uncorrelated sub-layers. For domain magnetisation orientations unequal to \( \pm 45^\circ \), the height of the peaks at \( \theta = \pm \psi_{1,2} \) may differ even when the layer thicknesses \( h_1 \) and \( h_2 \) are equal. At small transmission angles the calculated \( 2 \xi h \) is no longer identical to that calculated for the uncorrelated two-layer tape. Moreover, the \( 2 \xi h \) for the 'uncorrelated' tape is symmetric around \( \theta = 0 \) whilst the \( 2 \xi h \) for the 'correlated' tape shows a clear asymmetry. In order to investigate the influence of the domain parameters \( \delta \), \( \psi \) and \( h \) on the behaviour of \( 2 \xi h \), the calculation of the correlation function has been repeated for different domain configurations. No asymmetry is observed if the domain width is such that the correlation function consists of two well separated peaks, i.e. \( 2 \xi h \) for \( \theta = 0 \) is equal to zero. As a result no distinction can be made in this case between a tape consisting of two layers with or without a correlation between the domain structure in the two layers.

### 6.4 Results and Discussion

As a result of the evaporation of Co_{80}Ni_{20} in an oxygen atmosphere, ME-tape consists of small crystalline CoO and CoNi [Ric93] grains which form obliquely oriented columns. Clusters of small crystal grains may form magnetic grains that may exceed the size of the crystal grains. These magnetic grains form an obliquely oriented magnetic domain structure, i.e. a domain structure with a tilted anisotropy direction. Figure 6.3 gives a cross-section view of the ME-tape. The longitudinal direction of the tape is indicated by the horizontal arrow. In the figure one can clearly see the slightly curved columnar structure of the two sub-layers in the tape. The average orientation of the columns in each sub-layer is approximately \( 45^\circ \); the sub-layers have opposite column orientation.

#### 6.4.1 'As-received' Tape

The diagonal elements of the depolarisation matrix \( D_{ii} \), after correction for the polarisation of the empty beam, as a function of the transmission angle \( \theta \) are shown in figure 6.4. The statistical accuracy in the determination of the elements is in the order of the size of the symbols. Note that, although a stack of 448-tapes was used, the depolarisation of the beam is only 2%. The correlation function \( 2 \xi h \) for a single tape is calculated using eq. (6.1); the results are given in figure 6.5. Comparing the measured
2\xi h with the results of the computations shown in fig. 6.2, one can qualitatively draw the following conclusions: the orientation of the domain magnetisation in both layers is \( \approx 45^\circ \). The values for \( 2\xi h \) at \( \theta = \pm 45^\circ \) are not identical. This may be a result of a difference in the height of the domains in the two layers when no correlation exists between the domain structure in the sub-layers. Or, it is a result of a correlation between the domain structure in the two sub-layers. The latter should be accompanied with a clear asymmetry in \( 2\xi h \) for small \( \theta \) (see fig. 6.2). However, this feature is not observed in the measurement. From the absence of this asymmetry, one can conclude that the tape consists of two magnetically uncorrelated or independent sub-layers with different \( M_s h \). The magnetic interactions between the sub-layers, if present, are too small to influence the measured correlation function. The observation, that the tape can be considered as a tape consisting of two independent sub-layers, is supported by the observation of Bernard et al. [BSC94]. They studied the angular dependence of the coercivity and squareness of the magnetic hysteresis loop for a single- and a two-layer tape. It appeared that the simulated magnetic properties of a two-layer tape, assuming two independent layers, are in good agreement with the experimentally observed properties. Moreover, recent polarised-neutron reflectometry (PNR) experiments performed on the same tape revealed the existence of an interface between the two sub-layers with a strongly reduced magnetisation [GPRS]. Auger-experiments [BSC94] show a broad interface between the sub-layers with a high oxygen content. It is well known that the oxygen content has a significant influence on the saturation magnetisation; the saturation magnetisation at room temperature decreases when the oxygen content increases. This oxygen-enriched interface may reduce the magnetic coupling between the two sub-layers.
Figure 6.4: The measured diagonal elements $D_{ii}$ ($i = x, y$ or $z$) as a function of the transmission angle $\theta$, measured in a stack of 448 'as-received' tapes. Note the small deviations from unity.
6.4. **Results and Discussion**

Figure 6.5: The measured (o) and calculated (solid line) correlation function $2\xi h$ as a function of the transmission angle $\theta$ for the 'as-received' tape. The correlation function has been calculated for model A. The contribution of the first and second layer to the calculated $2\xi h$ is given by the dashed lines.

**Homogeneously Magnetised Domains (model A)**

Using the above-mentioned observations, the data has been analysed qualitatively using the computation method described in section 6.3. The simulated tape consists of two independent or magnetically uncorrelated sub-layers with domain sizes, domain magnetisation orientations and domain magnetisation as given in table 6.1. The actual values are obtained by changing these parameters until the calculated $2\xi h$ is in good agreement with the measured $2\xi h$. The calculated correlation function $2\xi h$ for this tape is given by the solid line in figure 6.5. The contribution of each sub-layer to the measured depolarisation is given by the dashed lines. The magnetic layer 'thickness', i.e. $(M_r h)_{1,2}$,

<table>
<thead>
<tr>
<th>model</th>
<th>calculated parameters</th>
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<tbody>
<tr>
<td></td>
<td>$M_r h$</td>
</tr>
<tr>
<td>A</td>
<td>72</td>
</tr>
<tr>
<td>B</td>
<td>66.6</td>
</tr>
</tbody>
</table>

is different for each sub-layer. The results of the PNR-experiments [GFRS] also show a difference in the magnetic film thickness. Furthermore, magnetisation measurements
performed by Bernards et al. [BSC94], and Stupp and Bernards [SB] showed a difference in the magnetic properties of the two sub-layers. This difference might be caused by a difference in switching field, a different magnetic layer thickness, or a different domain magnetisation orientation. From the results of the ND-experiments it appears that the domain magnetisation orientation (= 44°) is identical for the two sub-layers. Based on this, a difference in domain magnetisation orientation can be excluded as the cause of the different magnetic properties as observed by Bernards et al.; apparently it is caused by a difference in the magnetic film thickness 'M,h'. According to Stupp and Bernards [SB], the ratio between (M,h)1 and (M,h)2 is 0.87 which is in good agreement with the results obtained by ND. The domain period 2δL in the longitudinal direction of the tape is approximately 2.5 times the thickness of a magnetic sub-layer; 2δL = 225 nm.

Small Magnetic Grains (model B)

If the domains in the tape are homogeneously magnetised, the value for M,h obtained by ND and the model fit (72 mA) should be equal to that found in a VSM-measurement ((66.6 ± 3) mA). However, a difference of almost 10% is observed whilst the accuracy in the determination of M,h by a VSM-measurement is approximately 5%. The depolarisation of the beam is larger than calculated using a model based on homogeneously magnetised domains. This extra depolarisation may be caused by inhomogeneously magnetised domains. Consider a tape consisting of small magnetic grains (see fig. 6.6) with high saturation magnetisation Mbulk.

![Figure 6.6: Schematic drawing of a cross-section of one magnetic super domain consisting of small magnetic grains (model B).](image)

in the tape consists of voids or non-magnetic grains [Ric93]. The arrangement of the magnetisation of the magnetic grains is such that they form a so-called super domain structure. This structure has an alternating direction of magnetisation; the period of these domains is 2δL and 2δT (cf. figure 6.1). The mean magnetisation ⟨M⟩ of a super domain is equal to the saturation magnetisation M,s found in a magnetisation measurement.

For such a tape, the measured correlation function can be written as the sum of two correlation functions

\[
2ξh = (2ξh)_d + (2ξh)_g ,
\]

\[
\propto ⟨M⟩^2ξ_dh + (ΔM^2)ξ_gh ,
\]

(6.9)
with \((2\xi h)_s\) the contribution of the super domain structure, with correlation length \(\zeta_d\), to the measured correlation function; it is given by an expression analogous to the homogeneously magnetised domain model (model A). The correlation function \((2\xi h)_g\) arises from the contribution of \(\Delta M_1\), i.e., the deviation of the local magnetisation from the mean domain magnetisation. This is the contribution of the magnetic grains to the measured correlation function, its correlation length is \(\zeta_g\). Note that this contribution is always larger or equal to zero.

The correlation function \((2\xi h)_g\) at \(\theta = \pm \psi_{1,2}\), i.e., the orientation of the domain magnetisation, can be calculated from the difference between the measured correlation function and the calculated correlation function for a homogeneously magnetised domain \((M_s h)_1 + (M_s h)_2 = 66.6\) mA. At this angle the measured correlation function

![Graph](image)

\(\theta \text{ [°]}\)

Figure 6.7: The measured (○) and calculated (solid line) correlation function \(2\xi h\) as a function of the transmission angle \(\theta\) for the 'as-received' tape. The correlation function has been calculated for model B. The contributions of the first (a) and second (b) layer and the grains (c) to the calculated \(2\xi h\) are given by the dashed lines.

hardly depends on the width of the super domains but only on the \(M_s h\) (which is equal to 66.6 mA) of the super domains (see figure 6.7). The average measured and calculated value for \(2\xi h\) at \(\theta = \pm 44^o\) are equal to 2.85 and 2.45 \(10^3\) T\(^2\)nm\(^2\), respectively. Hence, the correlation function for the grains at this angle is 400 T\(^2\)nm\(^2\). The accuracy in the determination of \((2\xi h)_g\) is mainly caused by the accuracy in the determination of \(M_s h\). The 5\% inaccuracy in \(M_s h\) leads to an inaccuracy in the determination of \((2\xi h)_g\) of approximately 50%.

Assuming that the ratio of the mean grain size and the correlation length \(\zeta_g\) of the grains is in the order of unity, \(\zeta_g\) follows from (see [RR91b, section 3.2.2])

\[
\zeta_g e h (\mu_0 M_s^{bulk})^2 \left(1 - (4\pi e^2 / 81)^{1/3} m_2^3\right) = (0.4 \pm 0.2)10^3\, \text{T}^2\text{nm}^2.
\] (6.10)
Here $\epsilon$ is the magnetic filling fraction of the magnetic grains; it is equal to the ratio of $M_s (\approx 370 \text{ kA/m (from VSM)})$ and $M_{s,bulk} (\approx 1170 \text{ kA/m [Ric93]})$. The quantity $m_\theta$ is the reduced mean magnetisation of one single domain; it is assumed to be equal to unity. Assuming spherical grains, one finds that $\zeta_\theta = (5 \pm 3) \text{ nm}$ corresponding to the mean radius of the grains. In this case the depolarisation due to the grains is assumed to be independent of $\theta$. However, when the grains are rod-shaped ellipsoids, oriented in the growth direction of the columns, the depolarisation depends on $\theta$. In the case of strongly elongated ellipsoids, $(2\xi h)_\theta$ will have a sharp maximum when the neutrons are transmitted parallel to the orientation of the elongated ellipsoids. This feature, however, has not been observed. When the ellipsoids are less elongated, this maximum is less pronounced. The dependence of $(2\xi h)_\theta$ on the angle of incidence $\theta$ is determined by the actual shape or aspect-ratio of the ellipsoids. From the correlation function it is not possible to determine the aspect-ratio of the ellipsoids. Hence, the magnetic domain period $\delta_{1,2}^\ell$ and domain height $h_{1,2}$ of the super domain structure is calculated assuming spherical grains and the mean magnetisation of the super domains $\langle M_h \rangle = M_h h = 66.6 \text{ mA}$. Moreover, it is assumed that the tape consists of two uncorrelated or independent sub-layers. Figure 6.7 gives the result of this computation using the parameters listed in table 6.1. The domain period $2\delta^\ell_1$ is approximately 210 nm. It should be noted that the actual value depends slightly on the actual shape of the magnetic grains.

The domain period found by ND can be compared with the results of other domain imaging-techniques, e.g. using Magnetic Force Microscope (MFM) images taken from the magnetic domain structure at the interface of the tape. Unfortunately, at the time of writing this thesis these images were not yet available for this tape. In order to have some idea of the correctness of the order of magnitude of the domain period determined by ND, the results of a domain-imaging study of the domain period of ME-tape performed by Abelmann et al. [ABLP94] will be used. They studied, using Kerr-, MFM- and TEM-images, and using the Bitter-technique the relation between the domain period and the tape thickness for a series of single-layer ME-tape with tape thickness ranging between 40 nm and 1.7 $\mu$m. However, the ME-tape studied by ND consists of two layers. So, on first-sight Abelmann’s results can not directly be used for this comparison. However, when the observation is used that the tape can be considered as two independent single-layer tapes, with a thickness of 90 nm, a comparison can still be made. Then, the second layer is ignored and from figure 5 of ref. [ABLP94] one finds for a ME-tape with a film thickness of 90 nm a domain period of 250 nm, which is in reasonable agreement with the value observed by ND. The reader should be reminded that in MFM-imaging the stray fields emerging from the domains at the interface are imaged rather than the bulk domain structure as determined in an ND-experiment.

To verify the existence of the small magnetic grains in the tape, 1-dimensional ND-experiments were carried out on the SP. In these experiments the stack of 448 tapes was magnetised in fields between 0 and 600 kA/m, applied in the transverse direction of the tape. The neutron beam was sent through the stack of tapes parallel to the orientation of the mean magnetisation in one of the sub-layers; i.e. the direction of the applied field
was perpendicular to the propagation direction of the beam. The initial direction of the polarisation of the beam was parallel to the applied field. After transmission through the sample the component of the polarisation parallel to the direction of the field was analysed. The accuracy in the determination of the depolarisation $D_\parallel$ is in the order of 0.4%; this value has been determined from the spread in measuring points. If the sample is magnetised to saturation then the depolarisation due to the super domain structure has vanished. The depolarisation due to the magnetic grains, however, does not disappear. Assuming spherical grains and that the tape is magnetised at saturation, the remaining depolarisation i.e. the deviation from one, appears to be smaller than the accuracy of the experimental data. The remaining depolarisation has been calculated using [RR91b, eq. (3.16) pp. 34]. Unfortunately, this ND-experiment cannot confirm the existence of magnetic grains in the tape. Small-Angle Neutron Scattering (SANS) experiments, with or without polarised neutrons, can provide information on the crystallographic and magnetic structure of the tape. This can be done without a-priori knowledge of the saturation magnetisation of the tape itself and of the small magnetic grains. Recently, SANS experiments were carried out on a stack of 200 ME-tapes at the SANS-instrument LOQ located at ISIS, Rutherford Appleton Laboratory Chilton UK. In order to distinguish the nuclear scattering from the magnetic scattering the tapes were magnetised in a field of 0.95 T. The scattering in the direction parallel to the applied field is caused by scattering due to the crystallographic inhomogeneities in the tape, whilst the scattering perpendicular to the applied field is caused by both magnetic as well as by crystallographic inhomogeneities. The magnetic scattering in the direction perpendicular to the applied field does not disappear. This means that there is some magnetic contrast present, which is caused by regions with high magnetisation embedded in a non-magnetic matrix. The regions with high magnetisation can be associated with the small magnetic grains. From the dependence of the scattered intensity on the wave vector transfer, a characteristic diameter of the grains of approximately 16 nm is found. This value is in reasonable agreement with the value found by ND. More information on the SANS-experiments can be found in ref. [PRK]

6.4.2 Perpendicular Remanent State

In the remanent state after magnetisation in a perpendicular field, i.e. perpendicular to the plane of the tape, the measured correlation function has decreased with respect to the 'as-received' tape (see figure 6.8). Note that prior to the magnetisation treatment the stack was split into two stacks of 224 tapes. The two stacks were magnetised in opposite directions and joined after the magnetic treatment. In doing so, the stack of tapes exhibits no net remanent magnetisation. In doing so, the stack of tapes exhibits no net remanent magnetisation. The decreased value for the correlation function is a result of the remanent magnetisation $m_r$ of a single tape. If the tape consists of homogeneously magnetised domains, i.e. $(2\zeta h)_p$ (see eq. (6.9)) is zero, the correlation function is proportional to $(1 - m_r^2)(2\zeta h)_d$, with $(2\zeta h)_d$ the correlation function for $m_r$ is zero. For the tape studied the correlation function has two contributions; a contribution of the super domain structure and a contribution of the
small magnetic grains. The first is proportional to \((1 - m_r^2)\). If one assumes that the
correlation due to the grains is hardly affected by the mean remanent magnetisation
of one tape, the remanent magnetisation can be calculated. The value thus observed
for \(m_r\) is approximately 0.82. The effective domain orientation is 38°. This orientation
would not necessarily be the orientation of the domain magnetisation, but may be a
result of the way the two stacks are joined after magnetisation. The difference, at
\(\theta = 0°\), between the measured correlation function and the contribution of the grains,
after correction for \(m_r\), did not change with respect to the as-received tape. Since the
correlation function at \(\theta = 0°\) is a measure of the domain width, one can conclude that
the average domain width did not change.

## 6.5 Conclusion

For a commercially available CoNi metal-evaporated two-layer TDK Hi8 video
tape, the mean domain magnetisation orientation and the magnetic structure of each
sub-layer has been determined. It appears that the magnetic domains in the tape are
not homogeneously magnetised, but consist of regions with high magnetisation, so-
called small magnetic grains. These grains are embedded in a non- or weakly magnetic
matrix. The correlation length of the grains is found to be \((5 \pm 3)\) nm. When assuming
spherical grains, this correlation length corresponds to the mean radius of the grains.
The two sub-layers have opposite domain magnetisation orientation; for both layers
this is 44° with respect to the normal to the plane of the film. The two sub-layers can
be considered as two independent, or magnetically uncorrelated layers. The magnetic interactions between the layers, if present, are too weak, to influence the measured depolarisation. The two sub-layers are magnetically non identical; the magnetic thicknesses, i.e $M_s h$, differ by 12%. The domain period in the longitudinal direction of the tape is 210 nm.

For the tape in a remanent state after perpendicular magnetisation the measured depolarisation has decreased as a result of the mean remanent magnetisation $m_r$ of a single tape. From this decrease a remanence of 0.82 is found. The mean domain size did not change in the remanent state.
Chapter 7

Poly-crystalline MnZn-ferrites

7.1 Introduction

The microstructure of spinel ferrites, e.g. poly-crystalline MnZn-ferrite, has a great influence on their magnetic properties such as the coercivity and the magnetic permeability. The knowledge of the critical grain size, below which the grains become mono-domain, is of interest from a theoretical point of view. From an experimental determination of this critical grain size, ideas about domain-wall formation, interaction between domains, demagnetising fields and the origin of the initial magnetic permeability can be tested. The general interest in this type of study is triggered by numerous practical applications of these ferrites. Understanding the influence of the microstructure on the magnetic properties offers opportunity to design materials, whose magnetic properties are optimally adapted to the applications of such materials.

Due to recent developments in ceramic processing of ferrites [NV88, NS91] it became possible to prepare sintered MnZn-ferrites with grain sizes smaller than 1 μm. As the MnZn-ferrites, subject of this study, exhibit a low magnetocrystalline anisotropy and therefore relatively broad domain walls, a situation might occur that for sufficiently small grain sizes the domain wall thickness exceeds the grain size. Since it is difficult to imagine that a domain wall exists in such small grains, these grains become mono-domain. Furthermore, below a critical grain size the reduction of the magnetostatic energy, achieved by subdividing the grains into multiple-domains, becomes insufficient to compensate for the additional domain wall energy required to form a multi-domain structure in the grains.

Thus far, only Goto et al. [GIS80] reported on a study of the relation between domain size and grain size for Ba-ferrite. In this study a colloid SEM-technique was used. Although the possible observation of mono-domain particles was reported, no clear relation between grain size and domain size was observed. This may be due to the

---

fact that individual particles on the surface of sintered ferrites were studied. Neutron depolarisation (ND), however, offers the advantage of studying the average domain size within the bulk of a ferrite. Moreover, magnetic domain studies of surfaces give not a measure of the domain size within the bulk, because these results are influence by demagnetsing effects which are present at the surface. Furthermore, most magnetic properties such as the initial magnetic permeability are determined by the bulk of the material rather than by the surface. The results of the ND-study of the domain size in MnZn-ferrites are given and discussed in section 7.3. Furthermore, a theoretically expected critical grain size, below which the grains become mono-domain, is given. First, the preparation and characterisation of the samples and a brief description of the ND-experiment will be given in the next section.

7.2 Experimental

7.2.1 Sample Preparation and Characterisation

The MnZn-ferrites used in this study were prepared at the Philips Research Laboratories (PRL) in Eindhoven (NL) by wet chemical methods in the first steps of their preparation, namely in the powder preparation and in the consolidation. This method realises small initial particles with a narrow particle size distribution. Further details of the preparation method can be found in ref. [NV88, NS91]. By adjusting the sinter temperature, between 750 and 1250°C, MnZn-ferrites were prepared with different grain sizes varying between 0.3 and 30 μm without altering their chemical composition. The standard deviation of the grain size distribution $\sigma/D$ was approximately 35% for all samples, indicating the narrow size distribution.

The composition of the ferrites was determined by Electron Probe MicroAnalysis (EPMA). The composition was found to be Mn$_{0.60}$Zn$_{0.35}$Fe$_{2.05}$O$_4$, independent of the grain size. The main impurity in these ferrites was Na. Its content is lower than 100 weight ppm. The Si content is less that 40 weight ppm.

After the samples were polished and annealed at 500°C, microstructure photographs were taken at PRL using a Scanning Electron Microscope (Philips SEM 515). The mean intercept D is taken both as a measure and as a definition of the grain size. D corresponds to the mean size of the domains along an arbitrary line crossing the microstructure image. The mean intercept thus determined is a number average and has the advantage of representing a unique definition of the grain size, which does not depend on the actual shape of the grains. Furthermore, it is also a natural parameter to compare with the magnetic domain sizes as determined by ND. This is because neutrons, as they traverse the sample, cross the grains on random trajectories, and therefore 'detect' the domain size in a mean intercept manner. The grain sizes were determined at different parts of the sample to determine the average mean intercept. The error bars given correspond to the standard deviation in D, as determined from the different SEM-images of each ferrite. Typically, 100 - 150 grains were counted for
7.2. Experimental

Each ferrite.

The saturation magnetisation $M_s$ and the Curie temperature $T_c$ were determined on a temperature controlled magnetometer. For the MnZn-ferrites studied here $\mu_0 M_s = 0.52$ T at 18°C and $T_c = 430$ K. In the depolarisation technique, the neutron polarisation experiences a Larmor precession due to the local magnetic induction within each grain. Consequently, in the determination of the magnetic induction within each grain, the theoretical density of 5.16 g/cm$^3$ was used rather than the real density of the sample which lies between 4.9 and 5.1 g/cm$^3$. In this way the porosity of the samples, which is between 1% and 5%, has been compensated for.

7.2.2 Neutron Depolarisation

The ND-experiments in these poly-crystalline MnZn-ferrites were carried out on the KP (see for a description section 2.3). The wavelength of the monochromatic neutron beam was equal to $(0.16 \pm 0.01)$ nm. Prior to the ND-measurements the samples were demagnetised by heating them to 300°C, i.e. well above their $T_c$. To check whether the samples are demagnetised, all nine elements of the depolarisation matrix were measured. The non-diagonal elements were equal to zero, within the systematic error in the adjustment of the polarisation vector, confirming that the samples were in a non-magnetised state during the ND-experiment.

The magnetic domain size $\Delta$ and the mean-square direction cosines of the magnetic induction within the ferrites (magnetic texture) $\gamma_i$ were calculated from the measured depolarisation using

$$\Delta = -\frac{\ln(\det(\hat{D}))}{c\lambda^2(\mu_0 M_s)^2 L} \quad \text{and}$$

$$\gamma_i = 1 - \frac{2\ln(D_{ii})}{\ln(\det(\hat{D}))},$$

with the depolarisation constant $c\lambda^2$ equal to $5.581 \times 10^9$ T$^{-2}$m$^{-2}$. Equations (7.1) and (7.2) are derived from eqs. (2.9), (2.10) and (2.13) with $\Delta = \zeta$ and using $\epsilon = 1$. $\det \hat{D}$ is the determinant of the depolarisation matrix, which is in this case the product of the diagonal elements $D_{ii}$ (with $i = x$, $y$ or $z$). The diagonal element $D_{ii}$ follows from the measured intensity using eq. (2.17). The accuracy in the determination of the domain size $\Delta$ is determined by the statistics in the number of counts measured in a fully depolarised beam and in a beam when the set-up operates in the 'ii-mode'. The accuracy in $\Delta$ is derived to be in the order of 10%. For the ND-experiments, the thickness of the samples ranges between 250 \(\mu\)m and 1.3 mm, depending on the domain size, thus avoiding complete depolarisation of the beam.
7.3 Results and Discussion

Figure 7.1 shows from top to bottom the SEM-images of the microstructure of three representative samples of this study, namely for a sample with \( D = (0.35 \pm 0.05) \ \mu m \), \( D = (2.48 \pm 0.22) \ \mu m \) and \( D = (10.1 \pm 1.7) \ \mu m \).

The measured magnetic domain size \( \Delta \) as a function of the grain size \( D \) is shown in figure 7.2. For the grain size the mean intercept is used as explained before in the experimental section. The results indicate that, for the MnZn-ferrites studied here, the magnetic domain size and the grain size are equal for samples with a grain size between 0.3 and \( \approx 4 \ \mu m \). Hence, for these samples the grains are single-domain. The solid line in figure 7.2 corresponds to \( \Delta / D = 1 \), the ratio as one would expect for grains consisting of one magnetic domain. For grain sizes larger than \( 4 \ \mu m \) the magnetic domain size differs significantly from the solid line, indicating the existence of domain walls within the grains.

The dashed line in figure 7.2 corresponds to ratios of \( \Delta / D \) calculated for a grain consisting of two domains. This ratio, which is equal to 0.63, is obtained by assuming spherical grains, in which a domain wall exists that is confined to the equatorial plane of the sphere [Rek91]. The domain size of such a grain, averaged over all transmissions directions is equal to 0.63 times the size of the grains consisting of one domain. It should be borne in mind that the spread in grain sizes, whose standard deviation is \( \approx 35\% \), is larger than the error in \( D \) obtained from different SEM-images. Hence, it is possible that in samples with \( D \geq 4 \ \mu m \) grains exist without a domain wall and vice versa for \( D \leq 4 \ \mu m \). The same argument applies for more complex domain structures in even larger grain sizes. A sharp transition from a two- to a more complex domain structure is therefore not expected. The magnetic domain size was also measured for a sample with \( D = 27 \ \mu m \) (not shown in fig. 7.2). In this case \( \Delta \) was found to be equal to \( 8.1 \ \mu m \).

A first approximation for the expression for the critical mean intercept \( D_{C} \), at which the domain structure changes from mono-domain to two domains, is given by van der Zaag et al. [ZRN93, eq. (7)]. This expression has been derived starting from the simple basic-equation for the case of magnetically isolated grains given by Chikazumi and Charap [SC78, eq. (11.46)]. However, the soft magnetic environment in which the grains are embedded reduces the magnetostatic energy. This has been taken into account by van der Zaag et al. by the effective relative permeability \( \mu_{r+} \). The critical mean intercept is proportional to the domain wall energy and inversely proportional to the magnetostatic energy. For spherical grains \( D_{C} \) is given by

\[
D_{C} \approx \frac{4\mu_{r+}}{\mu_{0}M_{s}^2} \sqrt{\frac{2kT_{C}|K|}{a}},
\]

(7.3)

with \( k \) the Boltzmann constant, \( K \) the magnetocrystalline anisotropy constant and \( a \) the distance between the exchange-coupled spins. Substituting for \( \mu_{0}M_{s} = 0.52 \ \text{T}, T_{C} = 430 \ \text{K}, a \approx 0.4 \ \text{nm}, K = 32 \ \text{J/m}^3 \) and \( \mu_{r+} \approx 1200 \) [ZRN93], \( D_{C} \) is approximately 0.7 \( \mu m \).
7.3. Results and Discussion

Figure 7.1: Scanning electron microscope images of the microstructure of three representative samples: (top) mean intercept $D = (0.35 \pm 0.05) \, \mu m$, (centre) $D = (2.48 \pm 0.22) \, \mu m$ and (bottom) $D = (10.1 \pm 1.7) \, \mu m$. 
Figure 7.2: The magnetic domain size $\Delta$ as a function of the grain size $D$ for Mn$_{0.60}$Zn$_{0.35}$Fe$_{2.04}$O$_4$ ferrite. The domain sizes were determined by neutron depolarisation. The solid line corresponds to $\Delta/D = 1$, the ratio expected for single magnetic domain grains. The dashed line corresponds to $\Delta/D = 0.63$, the ratio calculated for grains consisting of two domains.

Despite the fact that the experimentally observed $D_C$ is considerably larger than the domain wall thickness ($\approx 1 \mu m$), the agreement between the experimentally observed and calculated $D_C$ is modest. A discrepancy of almost an order of magnitude is found. Nevertheless, taking into account the soft-magnetic environment in the basic-equation for $D_C$ given by Chikazumi for magnetically isolated grains has improved the correspondence between observed and calculated value for $D_C$ by orders of magnitude. However, starting from the exact expressions of Brown [Bro68] for a lower and upper bound for the critical radius of a magnetically isolated grain instead of the expression given by Chikazumi and Charap gives a better correspondence between the calculated and observed $D_C$. The observed value for $D_C = 3.8 \mu m$ falls well between the upper and lower bound, provided that the equations of Brown are transformed to those applying in a soft-magnetic environment.

The presence of a magnetic texture in the ferrites can also be measured in an ND-experiment. The magnetic texture is expressed by the parameter $\gamma$; it is related to the mean-square direction cosines of the local magnetic induction within the domains. Figure 7.3 gives the parameter $\gamma_x$, as measured along the neutron path. The vertical dashed line at $D = 3.8 \mu m$ marks the cross-over from the mono- to the multi-domain state. The horizontal dashed line gives the expected theoretical value for $\gamma_x$; it is equal to 1/2 for an isotropic distribution of domain magnetisations (see section 2.2). For the mono-domain state an average value of $\gamma_x = 0.49 \pm 0.04$ is found, whereas for the multi-
domain state the average value is equal to $0.48 \pm 0.06$. Since the expected accuracy in the determination of $\gamma_z$ is approximately $5\%$, the value found for $\gamma_z$ for the mono-domain state agrees well with the expected values for an isotropic distribution of domain magnetisations in these thermally demagnetised ferrites. The values for $\gamma_z$, and also for $\gamma_y$ and $\gamma_x$, for the multi-domain state are, within the experimental accuracy, equal to those for the mono-domain state. The values for $\gamma_y = 0.26 \pm 0.04$ and $\gamma_z = 0.25 \pm 0.04$ for the mono-domain state also agree with the expected theoretical value of $0.25$ for an isotropic distribution of domain magnetisations.

7.4 Conclusion

In a series of thermally demagnetised poly-crystalline MnZn ferrites with grain sizes varying between $0.3$ and $30 \mu m$ and unaltered composition $\text{Mn}_{0.60}\text{Zn}_{0.35}\text{Fe}_{2.05}\text{O}_4$, the magnetic domain size has been determined. A region is found for $D$ between $0.3$ and $3.8 \mu m$ where the domain size is equal to the grain size; no domain walls are present. These grains are mono-domain. For $D \geq 3.8 \mu m$ the domain size is significantly smaller than the grain size; a domain wall begins to appear within the grains. Initially the grains consist of two domains and with increasing grain sizes the domain structure changes to a more complicated structure. A sharp transition from a two- to a more complicated domain structure is not observed. Calculations of the transition from a mono- to a
two-domain structure show some agreement with the experimentally observed value, if the soft-magnetic environment is taken into account with respect to the expressions derived in standard textbooks.

The results of the ND-experiments show that no magnetic texture is present in the thermally demagnetised ferrites, i.e. the magnetic induction of the grains within the ferrites is oriented at random.
Chapter 8

Three Polariser - Two Shim Method\(^1\)

8.1 Introduction

Polarised neutron scattering, polarised neutron reflectometry and neutron depolarisation are nowadays widely used to study the magnetic properties of magnetic materials and superconductors. Although the principles and applications of these techniques may differ, the set-up for application of polarised neutrons always consists of a polarising device, a polarisation rotator or spinflipper, the sample and a neutron detector. When the polarisation of the neutron beam after interaction with the sample is analysed, the set-up also consists of a second spinflipper and a polarisation analysing device positioned between the sample and the neutron detector. Although most of the interactions of neutrons with matter are treated in the literature (see e.g. ref. [Blu63, MRK69]), little attention has been paid to the corrections that have to be carried out in order to obtain information on the nuclear and magnetic properties from the measured data. One of the corrections that has to be performed is the correction of the measured data for the polarising power of the polariser (and analyser) and the efficiency of the spinflippers.

A method of determining the polarising power of a neutron polariser is to measure the so-called flipping-ratio or shim-ratio of a set-up composed of two polarisers, one with unknown polarising power and an auxiliary polariser (see ref. [BF64, KWA69, NSB91]). When using this method, the polarising power of the auxiliary polariser has to be known, or both polarisers are chosen to be identical, i.e. one assumes that the polarising power of the two polarisers is equal.

Por et al. [PKR94] described a method of separating the net polarising power of

neutron polarisers from the depolarisation in a set-up composed of three polarisers. The main characteristic of this method, the so-called 'three polariser-two shim' (3p-2s) method, is the natural separation of the net polarising power of the second neutron polariser from the depolarisation in the set-up. The depolarisation and the efficiencies of the spinflippers are included in the polarising power obtained for the first and last polariser. Recently, the 3p2s-method has been applied to determine the polarising power of neutron polarisers for the neutron depolarisation set-ups at IRI-Delft [SPP94]. Figure 2.2 shows the measured polarising power of the SP-analyser as a function of the neutron wavelength.

When applying this method to magnetic scattering experiments, with the sample at the position of the second polariser, the ratio between the magnetic and the nuclear structure factor can be determined, without any corrections for depolarisation of the beam occurring in the set-up and the sample. However, the interaction between the neutrons and the sample has to be an entirely non-spinflip process.

In this chapter, a general expression for the measured intensity in a set-up composed according to the 3p-2s method, referred to as 3p-2s set-up henceforth, is derived using the spin density formalism. The set-up consists of two polarisers, two spinflippers or polarisation rotators and a (polarising) sample. In contrast with reference [PKR94] the neutrons with the sample may be such that it involves both a non-spinflip as well as a spinflip process. Using the general formulation of the 3p-2s method, the applications and limitations of this method are extensively discussed.

In section 8.2, the spin density formalism will be briefly described and a general equation for the measured intensity in a 3p-2s set-up is derived.

Section 8.3 discusses the applications and the limitations of the 3p-2s method in the fields of polarised neutron scattering, neutron reflectometry and neutron depolarisation.

The principles and applications of the 3p-2s method are summarised in section 8.4.

8.2 Theory

8.2.1 Introduction to the Spin Density Formalism

Before deriving an expression for the measured intensity in a 3p-2s set-up [PKR94] the spin density formalism [Ste63] will be briefly introduced.

The general form of a spin-wave function \( \chi \) for a neutron is

\[
\chi = a\chi_1 + b\chi_\uparrow ,
\]

where \( \chi_1 \) and \( \chi_\uparrow \) are the eigenfunctions of \( \hat{\sigma}_z \). The quantities \( aa^* \) and \( bb^* \) are proportional to the probabilities that a measurement of the \( z \)-component of the neutron spin will find that the spin is parallel (\( \uparrow \)) or anti-parallel (\( \downarrow \)) to the \( z \)-direction, respectively.

The spin-state of a neutron can be expressed by a spin density operator \( \hat{\rho} \) defined as

\[
\hat{\rho} = \chi\chi^* = \begin{pmatrix} a \\ b \end{pmatrix} (a^*b^*) = \begin{pmatrix} aa^* & ab^* \\ ba^* & bb^* \end{pmatrix} .
\]
8.2. Theory

In a real experiment we are dealing with a neutron beam. The spin density operator \( \hat{\rho} \) of a neutron beam, i.e. an ensemble of neutrons, can be found by averaging the wave functions in the beam. This gives an expression for \( \hat{\rho} \) analogous to eq. (8.2)

\[
\hat{\rho} = \begin{pmatrix} \langle aa^* \rangle & \langle ab^* \rangle \\ \langle ba^* \rangle & \langle bb^* \rangle \end{pmatrix}.
\]  
(8.3)

It is convenient to define \( a \) and \( b \) such that

\[
Tr[\hat{\rho}] = \langle aa^* \rangle + \langle bb^* \rangle = I,
\]  
(8.4)

where \( I \) is the intensity of the neutron beam.

Because \( \hat{\rho} \) is a (2 \( \times \) 2) Hermitian matrix, it can be expanded in terms of 4 linearly independent matrices, the unity matrix \( \hat{1} \) and the three Pauli matrices \( \hat{\sigma}_i \) (\( i = x, y, z \)), and the intensity \( I \)

\[
\hat{\rho} = \frac{I}{2} (\hat{1} + \mathbf{P} \cdot \hat{\sigma}),
\]  
(8.5)

where \( \mathbf{P} \) (\( ||\mathbf{P}|| \leq 1 \)) is the polarisation of the neutron beam, defined as

\[
\mathbf{P} \equiv \langle \hat{\sigma} \rangle = \frac{Tr[\hat{\rho} \hat{\sigma}]}{Tr[\hat{\rho}]},
\]  
(8.6)

Substitution of eq. (8.6) into eq. (8.5), gives an alternative expression for the spin density operator \( \hat{\rho} \)

\[
\hat{\rho} = \frac{1}{2} (Tr[\hat{\rho}] \hat{1} + Tr[\hat{\rho} \hat{\sigma}] \cdot \hat{\sigma}).
\]  
(8.7)

8.2.2 Generalised Formulation of the 3p-2s Method

Set-up

A 3p-2s set-up is composed of two neutron polarisers \( \mathcal{P} \) and \( \mathcal{Q} \) and a third polarising device or sample, referred to as sample \( \mathcal{R} \) henceforth, positioned between \( \mathcal{P} \) and \( \mathcal{Q} \). A schematic outline of the set-up is given in figure 8.1. To rotate the direction of polarisation from its original direction at the exit of \( \mathcal{P} \), towards the \( x \), \( y \) or \( z \)-direction (or their opposite directions) in front of the sample, a polarisation rotator \( \alpha_j \) is installed between \( \mathcal{P} \) and \( \mathcal{R} \). This rotator can be operated in the 'j-mode' (with \( j = x, y, z \) or their opposites denoting the direction to which the polarisation is rotated). Its action on the polarisation of the beam can be expressed by a (3 \( \times \) 3) matrix \( \hat{\alpha}_j \). The matrix \( \hat{\alpha}_j \) obeys

\[
\mathbf{P}_j^0 = \hat{\alpha}_j \mathbf{P},
\]  
(8.8)

with \( \mathbf{P}_j^0 \) the polarisation of the beam just before it enters the sample, and \( \mathbf{P} \) the polarisation of the beam beyond \( \mathcal{P} \) as seen along the neutron trajectory. If the beam does not depolarise in the rotator, the matrix \( \hat{\alpha}_j \) is a pure rotation matrix; its determinant is equal to one. However, if the beam depolarises upon transmission through the rotator
the determinant is smaller than one. Since the depolarisation of the beam occurring between $\mathcal{P}$ and $\mathcal{R}$ and the depolarisation in the rotator and the polarising power of $\mathcal{P}$ itself are indistinguishable, they can be included in the matrix $\hat{\alpha}_j$. Note that in general a set of six $\hat{\alpha}_j$-matrices describes the rotation towards the $x$, $y$ or $z$-direction or their opposite directions. For instance, let the beam beyond $\mathcal{P}$ be polarised in the $z$-direction, then $P_j^0$ can be written as

$$P_j^0 = \begin{pmatrix} \alpha_{zx} \\ \alpha_{zy} \\ \alpha_{zz} \end{pmatrix}, \quad (8.9)$$

with $|P_j^0| \leq 1$. Although, locally in the polariser $\mathcal{P}$ the polarisation may deviate from the $z$-direction, due to the strong magnetic field present in the polariser and in the rotator, it will average out to a vector along the $z$-direction. Its length, i.e. the degree of polarisation, may be reduced. This effect can be described by a depolarisation factor; it has been taken into account in the definition of the vector $P_j^0$. It appears that a set of eighteen scalars defines the six different rotations of the polarisation vector in the first polarisation rotator.

Upon interaction of the neutron beam with sample $\mathcal{R}$, characterised by its interaction potential $V_R$, the spin density operator, i.e. the polarisation and the intensity, of the beam have been changed. To analyse the polarisation of the beam beyond $\mathcal{R}$, a second polarisation rotator $\hat{\beta}_i$ is installed between $\mathcal{R}$ and $\mathcal{Q}$. This rotator $\hat{\beta}_i$ can be operated in six different 'i-modes', with $i = x$, $y$, $z$ or their opposites. In each mode the $i$-component of the polarisation can be rotated towards the direction of $\mathcal{Q}$. By analogy to the action of the first rotator, the action of the second rotator can be expressed by a $(3 \times 3)$ matrix $\hat{\beta}_i$. The matrix $\hat{\beta}_i$ obeys the relation

$$P_{ij}^{''} = \hat{\beta}_i P_j^0,$$  

with $P_j'$ and $P_{ij}^{''}$ the polarisation of the beam beyond $\mathcal{R}$ and just in front of $\mathcal{Q}$, respectively.
8.2. Theory

The intensity \( I_{ij} \) of the neutron beam beyond \( Q \) is given by eq. (8.A.6) (see appendix 8.A). It is convenient to rewrite the expression for \( I_{ij} \) such that it contains the polarisation of the beam beyond \( R \) separately, and the matrix \( \hat{\beta}_i \) and the analysing direction of \( Q \). Then eq. (8.A.6) can be rewritten as

\[
I_{ij} \propto (1 + P_j' \cdot (\hat{\beta}_i^T Q)) ,
\]

(8.11)

Let the analysing direction of the polariser \( Q \) be the \( z \)-direction, one can then define a vector \( Q_i' \), obeying

\[
Q_i' = \beta_i^T Q = \begin{pmatrix} \beta_{xx} \\ \beta_{xy} \\ \beta_{xz} \end{pmatrix} .
\]

(8.12)

Using the same arguments as used to define the vector \( P_j' \), the vector \( Q_i' \) contains also the depolarisation of the beam occurring between \( R \) and \( Q \), in the rotator, and also the polarising power of \( Q \). Once again a set of eighteen scalars defines the six different rotations of the polarisation in the second rotator. In the next section an expression for the polarisation beyond \( R \) and thereafter an expression for the intensity \( I_{ij} \) will be derived.

Expression for the Measured Intensity

When the first polarisation rotator operates in the \( j \)-mode, the spin density operator \( \hat{\rho}_j^0 \) of the neutron beam impinging onto the sample (see fig. 8.1) can, according to the definition of the spin density operator, be expressed in the polarisation of the beam \( P_j^0 \) and in the intensity of the beam \( t_P I_0 \)

\[
\hat{\rho}_j^0 = \frac{t_P I_0}{2} (\hat{1} + P_j^0 \cdot \hat{\sigma}) .
\]

(8.13)

Here \( t_P \) is the transmission of \( P \), when illuminated by a fully unpolarised beam, and \( I_0 \) the intensity of the beam at the entrance of the set-up. Upon interaction with \( R \) the spin density operator of the neutron beam has been changed into

\[
\hat{\rho}_j^I = \hat{\nu}_R \hat{\rho}_j^0 \hat{\nu}_R^\dagger ,
\]

(8.14)

with \( \hat{\nu}_R \) the interaction operator between the sample \( R \) and the neutrons given by [Lov84]

\[
\hat{\nu}_R = \langle \lambda | \hat{V}_R(\kappa) | \lambda' \rangle .
\]

(8.15)

In eq. (8.15) \( \lambda \) and \( \lambda' \) are the initial and final states of sample \( R \) in interaction with the neutron beam and \( \hat{V}_R(\kappa) \) is the Fourier transform of the interaction potential between \( R \) and the incident neutrons multiplied by \((m/2\pi\hbar^2)\).

Between \( R \) and \( Q \) the intensity of the beam does not change, hence \( Tr[\hat{\rho}_{ij}^I] = Tr[\hat{\rho}_{ij}^I] \). The matrix \( \hat{\rho}_{ij}'' \) is the density matrix of the beam just before it enters \( Q \). However, the
polarisation of the beam \( P'_j \equiv Tr[\hat{\rho}'_j \hat{\sigma}] / Tr[\hat{\rho}'_j] \) has changed, due to the action of the second polarisation rotator \( \beta \), operating in the 'i-mode', into

\[
P''_{ij} = \frac{Tr[\hat{\rho}''_{ij} \hat{\sigma}]}{Tr[\hat{\rho}''_{ij}]} = \beta_i P'_j .
\]

(8.16)

Hence, the spin density operator of the beam just before it enters \( Q \) is given by

\[
\hat{\rho}''_{ij} = \frac{Tr[\hat{\rho}''_{ij} \hat{\sigma}]}{2}(\mathbb{1} + P''_{ij} \cdot \hat{\sigma}) .
\]

(8.17)

According to eq. (8.A.6) and eq. (8.12) the measured intensity, when the set-up operates in the 'ij-mode, is given by:

\[
I_{ij} = \frac{tQ}{2}(Tr[\hat{\rho}'_j] + Tr[\hat{\rho}'_j \cdot Q'_i]) = \frac{tQ}{2}Tr[\hat{\rho}'_j](1 + P'_j \cdot Q'_i) .
\]

(8.18)

In a scattering experiment, with a scatterer at the position of \( R \), the intensity is measured as a function of the wave vector transfer \( \mathbf{k} \), and energy transfer \( \hbar \omega \) in a solid angle \( d\Omega \) and energy interval \( dE' \). To obtain the measured intensity \( I_{ij}(\mathbf{k}, \omega) \) we have to average eq. (8.18) over all scattering processes within \( d\Omega \) and \( dE' \)

\[
I_{ij}(\mathbf{k}, \omega) \propto t_P t_Q I_0 \frac{k'}{k} \sum_{\lambda, \lambda'} p_{\lambda}(Tr[\hat{\rho}'_j] + Tr[\hat{\rho}'_j \cdot Q'_i]) \delta(\hbar \omega + E_\lambda - E_{\lambda'}) ,
\]

(8.19)

with \( k, k', \mathbf{k} \) and \( \hbar \omega \) the wave vector of the neutron before and after interaction with \( R \) and the wave vector and energy transfer of the neutrons, respectively. In eq. (8.19) \( \hat{\rho}'_j \) is defined as

\[
\hat{\rho}'_j = \frac{1}{2} \hat{\theta}_R(\hat{\mathbb{1}} + P'_j \cdot \hat{\sigma}) \hat{\theta}_R^\dagger ,
\]

(8.20)

By definition

\[
\left( \frac{d^2 \sigma}{d\Omega dE'} \right)_j = \frac{k'}{k} \sum_{\lambda, \lambda'} p_{\lambda} Tr[\hat{\rho}'_j] \delta(\hbar \omega + E_\lambda - E_{\lambda'}) \quad \text{and}
\]

\[
\left( P' \frac{d^2 \sigma}{d\Omega dE'} \right)_j = \frac{k'}{k} \sum_{\lambda, \lambda'} p_{\lambda} Tr[\hat{\rho}'_j \cdot Q'_i] \delta(\hbar \omega + E_\lambda - E_{\lambda'}) .
\]

(8.21)

(8.22)

Hence, we obtain for the measured intensity

\[
I_{ij}(\mathbf{k}, \omega) \propto t_P t_Q I_0 \left[ \left( \frac{d^2 \sigma}{d\Omega dE'} \right)_j + \left( P' \frac{d^2 \sigma}{d\Omega dE'} \right)_j \cdot Q'_i \right] ,
\]

(8.23)

with expressions for the partial differential cross-section \( (d^2 \sigma / d\Omega dE')_j \) and \( (P' d^2 \sigma / d\Omega dE')_j \) given by Blume [Blu63].
In general, the partial differential cross-sections \( (d^2 \sigma / d \Omega d E') \) and \( (P' d^2 \sigma / d \Omega d E') \) can be written as a linear combination of two cross-sections: a polarisation independent and a polarisation dependent cross-section, denoted by the subscript 0 and \( p \), respectively. The latter is an odd function of the polarisation and is zero when the incident polarisation is equal to zero. Then

\[
I_{ij}(\kappa, \omega) \propto t_p t_q I_0 \times \left[ \left( \frac{d^2 \sigma}{d \Omega d E'} \right)_0 + \left( \frac{P' d^2 \sigma}{d \Omega d E'} \right)_0 \cdot Q'_i + \left( \frac{d^2 \sigma}{d \Omega d E'} \right)_p + \left( \frac{P' d^2 \sigma}{d \Omega d E'} \right)_p \cdot Q'_i \right].
\]

Equation (8.24) gives a general expression for the intensity measured in a set-up composed of two polarisers, two spin-flipper or polarisation rotators, and a sample positioned between the two rotators.

**Formulation of the 3p-2s Method.**

When applying the 3p-2s method, the intensities \( I_{i,j}, I_{i,\bar{j}}, I_{i,-j} \) and \( I_{i,-\bar{j}} \) are measured \((i,j = x, y, z)\). When polarisation rotators or spinflippers as described by Por et al. [PKR94] and Kraan et al. [KRP91] are installed in the set-up it is by symmetry allowed to assume that \( P_{2,j}^0 = -P_{j}^0 \) and \( Q'_{-i} = -Q'_i \). Then the following quantities can be deduced from the measured intensities:

\[
\left( \frac{P' \frac{d^2 \sigma}{d \Omega d E'}}{\frac{d^2 \sigma}{d \Omega d E'}} \right)_p \cdot Q'_i = \frac{I_{i,j} - I_{i,-j} + I_{i,-j} - I_{i,-j}}{I_{i,j} + I_{i,-j} + I_{i,-j} + I_{i,-j}}.
\]

\[
\left( \frac{P' \frac{d^2 \sigma}{d \Omega d E'}}{\frac{d^2 \sigma}{d \Omega d E'}} \right)_0 \cdot Q'_i = \frac{I_{i,j} - I_{i,-j} + I_{i,-j} - I_{i,-j}}{I_{i,j} + I_{i,-j} + I_{i,-j} + I_{i,-j}}.
\]

\[
\left( \frac{P' \frac{d^2 \sigma}{d \Omega d E'}}{\frac{d^2 \sigma}{d \Omega d E'}} \right)_p = \frac{I_{i,j} + I_{i,-j} - I_{i,-j} - I_{i,-j}}{I_{i,j} + I_{i,-j} + I_{i,-j} + I_{i,-j}}.
\]

The two rotators \( \alpha \) and \( \beta \) allow one to measure 36 intensities. Together with the equations (8.25), (8.26) and (8.27) they define three times nine expressions. These 27 equations are the basic equations for the 3p-2s method, they relate the measured intensities via the partial differential cross-section to the nuclear or crystallographic and magnetic properties of the sample. In general, 18 parameters are needed to describe the action of the two rotators. The number of parameters describing the nuclear and magnetic properties of the sample depends on the sample studied; these parameters will be described in the 'application section'. Note that mostly four intensities are measured, because \( i \) and \( j \) are restricted to the direction of magnetisation in \( \mathcal{R} \). Hence, the 27 equations are reduced to a set of three equations.
8.3 Applications

In this section the application of the $3p-2s$ method in the fields of polarised neutron scattering experiments, polarised neutron reflectometry and neutron depolarisation will be discussed. In the just mentioned techniques the set-up is composed of two polarisers $\mathcal{P}$ and $\mathcal{Q}$, two polarisation rotators or spinflippers $\alpha_i$ and $\beta_i$, and a sample $\mathcal{R}$ positioned between the two spinflippers or polarisation rotators (see fig 8.1).

In the first two techniques, the interaction between the sample and the neutron beam is such that it might polarise the beam as well as that it might change the direction of polarisation. In a neutron depolarisation experiment, however, only the degree and direction of polarisation changes upon transmission through the sample. Hence, the sample itself does not create polarisation.

8.3.1 Magnetic Elastic Neutron Scattering

In order to apply the $3p-2s$ method to magnetic elastic neutron scattering with polarisation analysis, which is known under the name spherical neutron polarimetry [NBFT91], one has to derive the appropriate expressions for the partial differential cross-sections. These cross-sections relate the nuclear and magnetic properties of the sample, via the magnetic and nuclear structure factor, to the measured intensity and polarisation. A general expression for the scattered intensity and the polarisation of the scattered polarisation at a given Bragg-reflection, as a function of the incident polarisation $\mathbf{P}_j^0$ has been derived by Blume [Blu63]; it can be written as

\[
\left( \frac{d\sigma}{d\Omega} \right)_0 = |N|^2 + \mathbf{M} \cdot \mathbf{M}^* \tag{8.28}
\]

\[
\left( \mathbf{P} \cdot \frac{d\sigma}{d\Omega} \right)_0 = NM^* + N^*M - \mathbf{P} \cdot (M^* \times M) \tag{8.29}
\]

\[
\left( \frac{d\sigma}{d\Omega} \right)_p = N^* \mathbf{P}_j^0 \cdot \mathbf{M} + N \mathbf{P}_j^0 \cdot \mathbf{M}^* + \mathbf{P}_j^0 \cdot (M^* \times M) \tag{8.30}
\]

\[
\left( \mathbf{P} \cdot \frac{d\sigma}{d\Omega} \right)_p = (|N|^2 - \mathbf{M} \cdot \mathbf{M}^*) \mathbf{P}_j^0 - M(\mathbf{P}_j^0 \cdot \mathbf{M}^*) + M^*(\mathbf{P}_j^0 \cdot \mathbf{M}) - i\mathbf{P}_j^0 \times (MN^* - M^*N) \tag{8.31}
\]

$N$ and $M$ are defined as

\[
N = F_N(\kappa) = \sum_{n=1}^{\text{unit-cell}} b_n e^{-i\kappa \cdot r_n}, \tag{8.32}
\]

\[
M = r_0 F_M(\kappa) \eta^\perp = (-r_0/2) \sum_{n=1}^{\text{unit-cell}} f_n(\kappa) \mu_n e^{-i\kappa \cdot r_n} (\kappa \times \eta_n \times \overline{\kappa}), \tag{8.33}
\]

with $b_n$, $f_n(\kappa)$ and $\mu_n$ the nuclear scattering length, magnetic form factor and atomic moment of an atom at position $r_n$ in the unit cell, respectively and $r_0$ is the classical
8.3. Applications

electron radius. The vector $\eta^\perp$ is a shorthand notation for $\mathbf{r} \times \mathbf{\eta} \times \mathbf{r}$, the vector $\mathbf{r}$ is unit vector of the wave vector transfer of the neutron to the sample and $\mathbf{\eta}$ is the direction of magnetisation. The cross-sections that describe the scattering of the neutron beam in the sample contain seven unknown parameters; the real and imaginary part of $F_N$ and $F_M$ and the three components of the vector $\eta^\perp$. Together with the 18 parameters that describe the rotation of the polarisation vector in the two rotators one ends up with 25 unknown parameters. In principle these parameters can be determined from the measured intensities using the equations (8.25)–(8.27), provided that these equations are all independent.

Consider a sample $\mathcal{R}$, positioned between the two polarisation rotators (see fig. 8.1), in which the spins are oriented either parallel or anti-parallel to the direction $\mathbf{\eta}$. Then the terms with $M^* \times M$ in eqs. (8.28)–(8.31) vanish. If the sample, e.g. a ferromagnet, is not magnetised to saturation, it consists of magnetic domains. The spins within a domain are pointing in the same direction, denoted by $\mathbf{\eta}_l$. The orientation of the spins $\mathbf{\eta}_l$ in different magnetic domains, labelled with $l$, may differ.

The partial differential cross-sections, for magnetic elastic neutron scattering in such a sample, are found by averaging the cross-sections for a given orientation of the spins $\mathbf{\eta}_l$ over the different orientations of the domains in the sample. Defining $\mathbf{\eta}_l = \langle \mathbf{\eta}_l \rangle + \Delta \mathbf{\eta}_l$, we can also define $\eta^\perp_l = \langle \eta^\perp_l \rangle + \Delta \eta^\perp_l$ and rewrite the magnetic structure factor $M$ for a domain labeled with $l$ as $M_l = \langle M \rangle + \Delta M_l$. The partial differential cross-sections at a given Bragg-reflection can then be written as

\[
\langle \frac{d\sigma}{d\Omega} \rangle_0^l = |N|^2 + \langle M \rangle^2 + \langle \Delta M^2 \rangle . \tag{8.34}
\]

\[
\langle P \frac{d\sigma}{d\Omega} \rangle_0^l = N^* \langle M \rangle + N \langle M^* \rangle . \tag{8.35}
\]

\[
\langle \frac{d\sigma}{d\Omega} \rangle_p^l = N^* P^0_j \cdot \langle M \rangle + N P^0_j \cdot \langle M^* \rangle \tag{8.36}
\]

\[
\langle P \frac{d\sigma}{d\Omega} \rangle_p^l = \left( |N|^2 + \langle M \rangle^2 + \langle \Delta M^2 \rangle \right) \frac{P^0_j}{\Delta^2}
- 2 \left( \langle M^* \rangle \times P^0_j \times \langle M \rangle + \langle \Delta M^*_l \times P^0_j \times \Delta M_l \rangle \right)
- i P^0_j \cdot (\langle M \rangle N^* - \langle M^* \rangle N) . \tag{8.37}
\]

In eqs. (8.34)–(8.37) $\langle \rangle_0$ is used to indicate that the cross-sections are averaged over the magnetic domains, which have different domain magnetisation orientations. $\langle M \rangle^2$ and $\langle \Delta M^2 \rangle$ are shorthand notations for $\langle M \rangle \cdot \langle M^* \rangle$ and $\langle \Delta M^*_l \cdot \Delta M_l \rangle$, respectively. Note that $\langle M \rangle$ and $\langle M^* \rangle$ are zero if the sample has no net magnetisation; the sample does not polarise the beam as one would expect for such a system.

The incident polarisation dependent ‘polarisation’ $(P' \frac{d\sigma}{d\Omega})_p$ of the scattered neutrons (eq. (8.37)) consists of three terms. The first is the contribution of scattered neutrons that do not change their polarisation upon interaction with $\mathcal{R}$; this scattering
process will be referred to as non-spinflip scattering.

The second term gives the contribution of neutrons that change their direction of polarisation, due to the presence of components of the local magnetisation both perpendicular to the direction of the initial polarisation and to the wave vector transfer $\kappa$. This process will be referred to as spinflip scattering.

The last term gives the contribution of neutrons that change their polarisation towards a direction perpendicular to the direction of the initial polarisation and perpendicular to $\langle \eta f^+ \rangle$. This process only occurs if the incident polarisation is not parallel to the $\langle \eta f^+ \rangle$ and the imaginary part of $N(M^*)$ is unequal to zero. Note that the polarisation $(P^d\sigma/d\Omega)_0$ of the scattered beam (eq. (8.35)), which does not depend on the incident polarisation and responsible for polarisation of an initially unpolarised beam, is proportional to the real part of $N(M^*)$. A more detailed identification and discussion of the terms that are responsible change in polarisation of the incident beam after scattering in the sample is given by Nunez et al. [NBFT91].

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In the particular case that the mean magnetisation $\langle \tilde{\eta}_f \rangle$ and the incident polarisation $P^d_0$ are parallel, and both perpendicular to $\kappa$, both the last term in eq. (8.37) and $(M^*) \times P^d_0 \times (M)$ are equal to zero. When the polarisation of the scattered beam is analysed along $\eta_f$, the polarising power of the first and second polariser, including the depolarisation occurring in the set-up, and the sample can be expressed in the measured intensities. Choosing the $z$-direction as the direction of the mean magnetisation $\langle \tilde{\eta}_f \rangle$, and bearing in mind that $R \equiv (P^d\sigma/d\Omega)_0 / (\sigma d\Omega)_0$ we find

$$T (\alpha_{zz}) = \sqrt{\frac{(I_{z,zz} - I_{z,-zz})^2 - (I_{z,-zz} - I_{z,zz})^2}{(I_{z,zz} + I_{z,-zz})^2 - (I_{z,-zz} + I_{z,zz})^2}},$$  \hspace{1cm} (8.38)

$$R_T = \sqrt{\frac{(I_{z,zz} - I_{z,-zz})^2 - (I_{z,-zz} - I_{z,zz})^2}{(I_{z,zz} + I_{z,-zz})^2 - (I_{z,-zz} + I_{z,zz})^2}},$$  \hspace{1cm} (8.39)

$$T (\beta_{zz}) = \sqrt{\frac{(I_{z,zz} - I_{z,-zz})^2 - (I_{z,-zz} - I_{z,zz})^2}{(I_{z,zz} + I_{z,-zz})^2 - (I_{z,-zz} + I_{z,zz})^2}},$$  \hspace{1cm} (8.40)

with

$$T^2 = 1 - \frac{\langle \Delta M^2 \rangle \langle (\Delta \tilde{\eta}_f^+ \cdot (\hat{z} \times \hat{r})^2) \rangle}{|N|^2 + \langle M \rangle^2 + \langle \Delta M^2 \rangle},$$

with $\hat{z}$ the unit vector along the $z$-direction and $\Delta \tilde{\eta}_f^+ = \frac{\Delta \eta_f^+}{\Delta \eta_f^+}$. It should be noted that in the expressions above we assumed that $(\alpha_{zz})_z = (\alpha_{yz})_z = (\beta_{zz})_z = (\beta_{yz})_z = 0$. In a real scattering experiment this is usually true, because the components of the polarisation perpendicular to $\langle \tilde{\eta}_f \rangle$ are averaged out due to the (high) magnetic field at the position of the sample, in the guide field and due to the finite wavelength resolution of the beam (see also set-up section).
8.3. Applications

$T$, the so-called depolarisation factor, accounts for the spin flip contribution to the scattering process, and provides information on the mean deviation of the local magnetisation from the mean magnetisation in the sample.

When the local magnetisation has components neither perpendicular to $\vec{n}$ nor perpendicular to the direction $\langle \vec{\eta} \rangle$, that is parallel to the incident polarisation, $T$ is equal to one. Hence, the scattering is an entirely non-spinflip process. In this case, the net polarising power of the sample can be determined without any corrections for depolarisation effects occurring in the set-up. These effects are included in the polarising power obtained for the first and last polariser. From the net polarising power of the sample, the ratio between the nuclear and the magnetic structure factor can be determined (see ref. [PKR94]).

In the case that the scattering involves both a spin flip and a non-spinflip process, $T$ is unequal to one. The measured net polarising power of the sample is affected by the depolarisation factor $T$. The 'intrinsic' depolarisation in the set-up and polarising power of the polarisers $(\alpha_{zz})_z$ and $(\beta_{zz})_z$, and the depolarisation due to spinflip scattering, can not be distinguished in that case. Once $(\alpha_{zz})_z$ and $(\beta_{zz})_z$ are known, $T$ can be determined from the measured intensities, see eqs. (8.38) and (8.40). $(\alpha_{zz})_z$ and $(\beta_{zz})_z$ are found in an experiment in which the sample is magnetically saturated, i.e. $T \equiv 1$, or in an experiment with a 'non-spinflip' polariser at the position of $\mathcal{R}$. It should be noted that it is assumed that the intrinsic depolarisation in the set-up does not change upon applying the magnetic field onto the sample or exchanging the sample by a non-spinflip polariser. This can easily be verified by calculating the quotient of eqs. (8.38) and (8.40), this quotient should be constant when applying a magnetic field. When the depolarisation factor $T$ is known, the net polarising power of the sample can be calculated using eq. (8.39), and hence the ratio between the nuclear and the magnetic structure factor can be determined.

8.3.2 Neutron Reflectometry

In order to derive an expression for the measured intensities, when the $3p-2s$ method is applied to polarised neutron reflectometry, one has to start with the general equation for the measured intensity. Substituting

$$Tr[\rho_j] = Tr[\rho_j^0] + Tr[\rho_j^p],$$

and

$$Tr[\rho_j^\sigma] = Tr[\rho_j^\sigma^0] + Tr[\rho_j^\sigma^p],$$

in equation (8.18), it reads

$$I_{ij} = \frac{t_Q}{2} \left[ Tr[\rho_j^0] + Tr[\rho_j^\sigma^0] \cdot Q_i^0 + Tr[\rho_j^p] + Tr[\rho_j^\sigma^p] \cdot Q_i^p \right],$$

(8.41)

wherein the polarisation independent and polarisation dependent part of the spin density operators, are denoted by the subscript 0 and $p$, respectively. Note the similarities
between eq. (8.41) and (8.24), the intensity of the incident beam and the transmission of the first polariser are included in the traces of the spin density operator. The more general equation for the measured intensity (eq. (8.41)) is used rather than eq. (8.24), because in a neutron reflectivity experiment the measured intensity is expressed in terms of reflectivity. In a neutron scattering experiment, however, it is convenient to write the measured intensity in terms of the partial differential cross-sections.

The spin density operator \( \hat{\rho}_j' \) of the reflected neutron is given by

\[
\hat{\rho}_j' = \hat{v}_R \hat{\rho}_j' \hat{v}_R^\dagger, \tag{8.42}
\]

with

\[
\hat{v}_R = \begin{pmatrix}
R_{++} & R_{-+} \\
R_{+-} & R_{--}
\end{pmatrix},
\tag{8.43}
\]

where the products \( R_{++} R_{-+} \), etc. are the probabilities that the spin-state of a neutron changes from + to - , etc. These probabilities can be calculated by a scheme described by Felcher et al. [FCH87] or by Pleshanov [Ple94]. In general, the reflectivities in eq. (8.43) are a function of the wave vector transfer \( \kappa = 4\pi \sin(\theta)/\lambda \), but for reasons of readability they are omitted.

Using the definition of the spin density operator for the incident neutron beam \( \hat{\rho}_j^0 \) (eq. (8.13)) one obtains the following expressions for \( Tr[\hat{\rho}_j']_0 \), \( Tr[\hat{\rho}_j']_p \), \( Tr[\hat{\rho}_j' \hat{\sigma}]_0 \) and \( Tr[\hat{\rho}_j' \hat{\sigma}]_p \) in eq. (8.41):

\[
Tr[\hat{\rho}_j']_0 = \frac{t_p I_0}{2} \left( |R_{++}|^2 + |R_{+-}|^2 + |R_{-+}|^2 + |R_{--}|^2 \right), \tag{8.44}
\]

\[
Tr[\hat{\rho}_j']_p = \frac{t_p I_0}{2} \left( \frac{2 \Re \left( R_{++} R_{++}^* + R_{+-} R_{-+}^* \right)}{(|R_{++}|^2 + |R_{+-}|^2 - |R_{++}|^2 - |R_{-+}|^2)}, \tag{8.45}
\right)

\[
Tr[\hat{\rho}_j' \hat{\sigma}]_0 = \frac{t_p I_0}{2} \left( \frac{2 \Re \left( R_{++} R_{-+}^* + R_{-+} R_{++}^* \right)}{2 \Im \left( R_{++} R_{+-}^* + R_{-+} R_{-+}^* \right) + |R_{++}|^2 + |R_{+-}|^2 - |R_{++}|^2 - |R_{-+}|^2}), \tag{8.46}
\right)

\[
Tr[\hat{\rho}_j' \hat{\sigma}]_p = \frac{t_p I_0}{2} \left[ \left( \alpha_{xz} \right)_j \left( \frac{2 \Re \left( R_{++} R_{-+}^* + R_{-+} R_{++}^* \right)}{2 \Im \left( R_{++} R_{+-}^* + R_{-+} R_{-+}^* \right) + 2 \Im \left( R_{++} R_{-+}^* - R_{-+} R_{++}^* \right) + |R_{++}|^2 - |R_{+-}|^2 - |R_{++}|^2 - |R_{-+}|^2 \right) \right)
\right.

\[
\left. \left( \alpha_{yz} \right)_j \left( \frac{-2 \Re \left( R_{++} R_{-+}^* - R_{-+} R_{++}^* \right)}{2 \Im \left( R_{++} R_{+-}^* - R_{-+} R_{-+}^* \right) + 2 \Im \left( R_{++} R_{-+}^* - R_{-+} R_{++}^* \right) + |R_{++}|^2 - |R_{+-}|^2 - |R_{++}|^2 - |R_{-+}|^2 \right) \right) \right], \tag{8.47}
\]

with e.g. \( (\alpha_{xz}) \); the \( l \) (\( = x, y \) or \( z \))-component of the incident polarisation when the first polarisation rotator operates in the 'j-mode' and polariser \( \mathcal{P} \) is magnetised in the
z-direction. $\mathcal{R}$ and $\Im$ denote the real and imaginary part of the expression between parentheses, respectively.

In the next section it is assumed that $R_{+-}$ and $R_{-+}$ are not equal. In a magnetic sample (see figure 8.2) with $B_\perp$ equal to zero, the spinflip reflectivities $R_{+-}$ and $R_{-+}$ vanish.

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Figure 8.2 gives a schematic outline of the sample geometry and the magnetisation of the sample. Suppose the polarisation of the incident neutron beam is either parallel or anti-parallel to $B_\parallel$, in our case the $z$-direction, $(\alpha_{xz})_x = (\alpha_{yz})_x = 0$. When the polarisation of the reflected beam is analysed along the $z$-direction and $(\beta_{xz})_x = (\beta_{yz})_x = 0$, one can derive expressions, that relate the measured intensities to the polarising power of the first and last polariser and to the polarising power of the sample, similar to the ones derived in section 8.3.1.

\[
T_1 T_2 \left( \alpha_{xz} \right)_x = \frac{(I_{z,x} - I_{z,-x})^2 - (I_{z,-x} - I_{z,x})^2}{(I_{z,x} + I_{z,-x})^2 - (I_{z,-x} + I_{z,x})^2}, \quad (8.48)
\]

\[
\frac{T_2}{T_1} \left( \mathcal{R} \right)_x = \frac{(I_{z,x} - I_{z,-x})^2 - (I_{z,-x} - I_{z,x})^2}{(I_{z,x} + I_{z,-x})^2 - (I_{z,-x} + I_{z,x})^2}, \quad (8.49)
\]

\[
\frac{T_1}{T_2} \left( \beta_{xz} \right)_x = \frac{(I_{z,x} - I_{z,-x})^2 - (I_{z,-x} - I_{z,x})^2}{(I_{z,x} + I_{z,-x})^2 - (I_{z,-x} + I_{z,x})^2}, \quad (8.50)
\]
with \( T_1 \) and \( T_2 \) the so-called depolarisation factors, that account for the spinflip reflectivities and are given by

\[
T_1^2 = 1 - 2 \frac{|R_{++}|^2 + |R_{+-}|^2}{|R_{++}|^2 + |R_{+-}|^2 + |R_{-+}|^2 + |R_{--}|^2},
\]

(8.51)

\[
T_2^2 = 1 - 2 \frac{|R_{++}|^2 - |R_{+-}|^2}{|R_{++}|^2 + |R_{+-}|^2 - |R_{-+}|^2 - |R_{--}|^2},
\]

(8.52)

and the polarising power of the sample is defined by

\[
(R)_z \equiv \left( \frac{\text{Tr}[\hat{\rho}_j \tilde{\sigma}]_0}{\text{Tr}[\hat{\rho}_j^0]_0} \right)_z = \frac{|R_{++}|^2 + |R_{--}|^2 - |R_{+-}|^2 - |R_{-+}|^2}{|R_{++}|^2 + |R_{+-}|^2 + |R_{-+}|^2 + |R_{--}|^2}.
\]

(8.53)

When the spinflip reflectivities \( |R_{++}|^2 \) and \( |R_{--}|^2 \) are equal to zero, the two depolarisation factors are equal to one. Hence, the polarising power of the sample can directly be determined from the measured intensities. The depolarisation occurring in the set-up is included in the definition of \( P_j^0 \) and \( Q_i^j \).

In the case that the spinflip reflectivities are non-zero, the obtained polarising power of the sample is affected by the depolarisation factors \( T_1 \) and \( T_2 \). Moreover, the intrinsic depolarisation in the set-up and the depolarisation, due to spinflip reflectivity, cannot be distinguished in that case. These factors can be determined using the method which is described in section 8.3.1. It should be noted that in contrast with section 8.3.1, the obtained polarising power of the first and last polarisers are not equally influenced by the depolarisation factors. This makes a distinction between a trivial change in \((\alpha_z)_z\) and \((\beta_{zz})_z\) and in \( T_1 \) and \( T_2 \) upon applying a magnetic field more difficult than in the case when \( T_2 \) is equal to one.

When the spinflip reflectivities \( R_{+-} \) and \( R_{-+} \) are equal and non-zero, the depolarisation factor \( T_1 \) is equal to one, and we obtain expressions for the polarising power analogous to the expressions discussed in section 8.3.1.

### 8.3.3 Neutron Depolarisation

In a neutron depolarisation (ND) experiment the change in polarisation of a polarised neutron beam is analysed after transmission through the sample [RR91b]. This change is expressed in the so-called \((3 \times 3)\) depolarisation matrix \( \hat{D} \) :

\[
P_j' = \frac{\text{Tr}[\hat{\rho}_j \tilde{\sigma}]}{\text{Tr}[\hat{\rho}_j^0]} = \hat{D} \frac{\text{Tr}[\hat{\rho}_j^0 \tilde{\sigma}]}{\text{Tr}[\hat{\rho}_j^0]} = \hat{D} P_j^0,
\]

(8.54)

with \( \text{Tr}[\hat{\rho}_j^0] = \text{Tr}[\hat{\rho}_j^0] t_R \) and \( t_R \) the transmission of the sample. A schematic drawing of the set-up is given in figure 8.1.

The measured intensity, when the set-up operates in the 'ij-mode' follows from eq. (8.18) and (8.54):

\[
I_{ij} = \frac{t_p t_R t_Q t_0}{4} (1 + \hat{D} P_j^0 \cdot Q_i).
\]

(8.55)
8.4 Summary and Final Remarks

It is obvious (see eq. (8.41)) that eq. (8.55) does not contain the polarisation dependent 'intensity' $Tr[p\hat{\sigma}]_p$ and the polarisation independent 'polarisation' $Tr[p\hat{\sigma}]_0$. The latter implies that the sample does not create polarisation, i.e. $R \equiv 0$.

When applying the $3p-2s$ method, one will find that $I_{i,j} = I_{-i,-j}$ and $I_{-i,j} = I_{i,-j}$. As a consequence, the elements of the depolarisation matrix $\hat{D}$, $\mathbf{P}_j^0$ and $\mathbf{Q}_i^0$ are not determinable from the measured intensities. Moreover, $\mathbf{P}_j^0$ and $\mathbf{Q}_i^0$ are not separable, hence only the quantity $\hat{D}\hat{P}_j^0,\mathbf{Q}_i^0$ can be determined. However, when the elements of the depolarisation matrix $\hat{D}$ are known, $\mathbf{P}_j^0$ and $\mathbf{Q}_i^0$ can be determined from the measured intensities using the calibration procedure described by Roest and Rekveldt [RR92].

In the section above, the change in polarisation in an ND-experiment is described in the so-called Larmor-approach (see section 2.2). Using this approach the depolarisation is a result of the precession of the polarisation vector $\mathbf{P}_j^0$ around the local magnetic induction in the sample. The neutron beam does not change its direction while it passes through the sample, i.e. the neutrons do not scatter in the sample. Moreover, all incident neutrons, except those that are absorbed in the sample, enter the detector. The absorption of the neutrons in the sample is accounted for by $t_R$, which does not depend on the polarisation of the incident beam.

Topperve and Weniger [TW89] described ND in the scattering approach. In this approach depolarisation is a result of magnetic scattering of neutrons, while they pass through the sample, within the aperture of the detector. Besides the neutrons that are absorbed in the sample, the intensity of the beam is decreased by neutrons that do not enter the detector after scattering, as the neutrons may scatter at angles larger than the acceptance angle of the analysing system, i.e. analyser and detector. This additional decrease in intensity, however, may depend on the polarisation of the incident beam.

In this case the $3p-2s$ method (see section 8.3.1) can be used to separate the elements of the depolarisation matrix $\hat{D}$ from the incident polarisation dependent transmission. In doing so, and measuring the polarisation as a function of the wavelength of the neutrons and the aperture of the detector, detailed information can be obtained on the nuclear and magnetic properties of the sample.

8.4 Summary and Final Remarks

This chapter showed that the application of the $3p-2s$ method to polarised neutron experiments, where the sample being investigated is positioned between the first and last polariser, allows one to determine from the measured intensities, the net polarising power or the ratio between the nuclear and magnetic structure factor of the sample. This can be done without any corrections for the depolarisation of the beam occurring in the set-up and sample. These effects are included in the polarising power obtained for the first and last polariser. The only restriction is, however, that the interaction between the neutrons and the sample is such that the interaction process is entirely non-spinflip and polarisation rotators or spinflippers, as mentioned in section 8.2.2, are installed in the set-up.
When the interaction process involves both a spinflip and a non-spinflip process, the obtained polarising power for the two polarisers and the sample are affected by depolarisation factors, which account for the spinflip contribution to the interaction process. From this factor, information can be obtained on the components of the local magnetisation both perpendicular to the wave vector transfer and to the direction of the initial polarisation. The depolarisation factor can be determined from the change in the obtained polarising power of the first and last polariser, upon applying a magnetic field that saturates magnetically the sample. In this specific case, the polarising powers obtained for the first and last polariser are equally influenced by the depolarisation factor.

In a polarised neutron reflectometry experiment, information on the spinflip reflectivities can be obtained from the depolarisation factors. When the spinflip reflectivities are not equal, the obtained polarising power for the first and last polariser are not equally influenced by these factors.

In general, the polarising power of the first and last polariser, with the possible influence of the depolarisation factor and the depolarisation occurring in the set-up, can be determined from the measured intensities, provided that the polarising power of the second polariser, i.e. the sample, is unequal to zero. When the polarising power of the sample is equal to zero, only the product of the polarising power of the first and last polariser can be determined. This is the case when applying the $3p-2s$ method to neutron depolarisation experiments.
8.A Intensity Beyond a Polariser

In order to derive an expression for the measured intensity in the detector placed beyond a polariser, the following interaction potential $\tilde{v}_Q$ of a neutron polariser is used

$$\tilde{v}_Q = \frac{t_Q^\frac{1}{2}}{\sqrt{1 + |q|^2}} (\hat{1} + q \cdot \hat{\sigma}) ,$$  \hspace{1cm} (8.A.1)

where $t_Q$ is the transmission of the polarising device. Eq. (8.A.1) is a slightly modified form of an interaction potential given by Steinsvoll [Ste63, pp. 16]. The direction of the vector $q$, $\hat{q}$, gives the direction of the polarisation of the beam, when $Q$ is illuminated by an unpolarised beam, and the length of $q$ accounts for the polarising power of $Q$.

After transmission through the polariser, the spin density operator of the incident beam $\hat{\rho}'_{ij}$ changes into $\hat{\rho}''_{ij}$ (see fig. (8.1))

$$\hat{\rho}''_{ij} = \hat{v}_Q \hat{\rho}''_{ij} \hat{v}_Q^\dagger ,$$  \hspace{1cm} (8.A.2)

with

$$\hat{\rho}''_{ij} = \frac{Tr[\hat{\rho}''_{ij}]}{2} (\hat{1} + P_{ij}'' \cdot \hat{\sigma}) = \frac{1}{2} (Tr[\hat{\rho}''_{ij}] \hat{1} + Tr[\hat{\rho}''_{ij} \hat{\sigma}] \cdot \hat{\sigma}) .$$  \hspace{1cm} (8.A.3)

Bearing in mind that, when the polariser is illuminated by an unpolarised beam, i.e. $P_{ij}'' = 0$ or $\hat{\rho}''_{ij} = \frac{1}{2} I_0 \hat{1}$, the polarising of the beam beyond $Q$ equals the polarising power of $Q$. $Q$ is defined as:

$$Q \equiv \left| \frac{Tr[\hat{v}_Q \hat{\rho}''_{ij} \hat{v}_Q^\dagger \hat{\sigma}]}{Tr[\hat{v}_Q \hat{\rho}''_{ij} \hat{v}_Q^\dagger]} \right|_{\hat{\rho}''_{ij} = \frac{1}{2} I_0 \hat{1}} = \frac{2|q|}{1 + |q|^2} \hat{q} .$$  \hspace{1cm} (8.A.4)

Using eqs. (8.A.2)–(8.A.4) the intensity measured in a detector positioned beyond the last polariser $Q$ when the set-up operates in the 'ij-mode' can be written as:

$$I_{ij} = Tr[\hat{\rho}''_{ij}] = \frac{t_Q}{2} (Tr[\hat{\rho}''_{ij}] + Tr[\hat{\rho}''_{ij} \hat{\sigma}] \cdot Q) ,$$  \hspace{1cm} (8.A.5)

and its equivalence

$$I_{ij} = Tr[\hat{\rho}''_{ij}] = \frac{t_Q Tr[\hat{\rho}''_{ij}]}{2} (1 + P_{ij}'' \cdot Q) .$$  \hspace{1cm} (8.A.6)
Chapter 9

Conclusions and Final Remarks

Three-dimensional neutron depolarisation (ND) is a powerful technique to obtain information on the micro and sub-micro magnetic properties of e.g. hard- and soft magnetic materials. In order to apply ND to the still expanding range of applications, the ND set-ups at IRI had to be upgraded and new measuring techniques had to be developed. For the SpiegelPolarimeter SP and the KristalPolarimeter KP at IRI new polarisers and analysers were installed. This resulted in a gain in neutron intensity by at least an order of magnitude. Furthermore, the polarisation of the beam increased significantly. A new depolarisation module was designed, constructed and installed in the SP. In this module two translation stages and one rotation stage were installed allowing ND-experiments as a function of the position and transmission angle for samples and sample holders weighing up to 50 kg. Such experiments can be performed fully automated and computer controlled. To shield the neutron beam and sample from outside magnetic stray fields, a sandwich structure of iron and μ-metal plates was used. This construction reduces the stray fields to 10 A/m. Moreover, the remaining field inside the module is insensitive to changes in the magnetic field outside the module.

The rotation compensation method, based on the principle of neutron spin-echo, was developed. This method prevents the beam from additional depolarisation in magnetised media due to the wavelength spread of the beam. Hence, the complicated numerical corrections for this depolarisation are no longer necessary. Moreover, the complete thermal neutron spectrum can be used for ND-experiments in magnetised media. This has the advantage that the intensity is approximately a factor 10 – 20 larger than that of a monochromatic beam.

The principle of neutron spin-echo was also used to design and construct a special magnetic coil system, the so-called Spin-Echo Coil System (SpECS). This system allows three-dimensional ND-experiments in fields up to 0.5 T. An important part of this system is a specially designed and constructed wire screen positioned between the two sections of the SpECS. The magnetic field of this screen suppresses the unwanted components of the magnetic field and so depolarisation due to these components is prevented. At the time of writing this thesis the maximum applicable field in the SpECS has been increased up to 1 T. This allows measurements of the depolarisation along
the entire magnetic hysteresis-loop for most recording materials such as magnetic small particles, thin CoCr-films and metal-evaporated tape, and magnetic particle dispersions and much more.

Using the SpECS, the first three-dimensional ND-experiment in high magnetic fields ever was performed. In this experiment, the magnetic correlations between Co-modified $\gamma$-Fe$_2$O$_3$ particles embedded in a solid matrix were studied as a function of the applied magnetic field. From the magnetic field dependence of the magnetic correlation length one can conclude that the magnetic correlations originate predominantly from variations in the particle density within the sample. These variations can be considered as 'quasi-particles' which magnetise differently as a function of the applied magnetic field. In the demagnetised state, the magnetic correlation length exceeds the mean particle size by a factor of 2 – 3. This means that in this magnetic state positive correlations still exist between the particles.

In the virginial state of both the nano- and the micro-crystalline barium hexaferrite samples the particles form stacks. These stacks are formed during the preparation process of the particles. With increasing remanent magnetisation, after a pulsed magnetic field, correlations between clusters of stacks and/or clusters of particles are induced. For the nano-crystalline ferrites these correlations are broken in the magnetisation reversal process, whilst for the micro-crystalline ferrites the correlations are maintained. This difference is likely to be caused by the presence of a relatively thick non-magnetic layer at the basal planes of the nano-crystalline ferrites, which reduces the interaction between the particles. It might also be a trivial effect because of the difference in particle size of the nano- and micro-crystalline ferrites.

The effect of milling an Fe$_2$O$_3$ particle dispersion was studied by means of ND. The results of the experiments after different magnetic treatments and demagnetisation by shaking showed that as the milling process proceeds clusters of particles are broken, and the dispersion is well dispersed after 90 minutes of milling. Furthermore, the dispersion remains stable and well dispersed over a period of two weeks after preparation. Monte-Carlo (MC) computations were performed to get insight in the mechanism involved in the formation of the 3D-microstructure of the dispersion. To check the correctness of the model, the neutron depolarisation is simulated using the results of MC-computations and the ND-theory adapted to particulate dispersions. The zero-field particle configuration is in agreement with the experiments; the correlation length equals the mean particle size which is expected for a randomly oriented particle system. However, the results of the computation for a dispersion that is magnetised in a magnetic field or in a remanent magnetised state could not be correlated with the experimental data. These measurements were not taken using the same experimental conditions as in the numerical simulations. Correlation of the simulations with experimental data can be realised by carefully choosing well defined samples and magnetic treatments. Using the SpECS and the rotation compensation method these experiments can be performed quite easily. The results of previous ND-experiments in powder samples in a magnetised state showed that the magnetic correlations are predominantly caused by particle density fluctuations. These fluctuations are likely to be present in real dispersions. Allowing
these fluctuations also in the MC-computations will probably improve the correctness of the model.

For a commercially available CoNi metal-evaporated two-layer TDK Hi8 video tape, the mean domain magnetisation orientation and the magnetic structure of each sub-layer were determined from the angle dependence of the depolarisation. It appeared that the magnetic domains are not homogeneously magnetised, but that they consist of regions with high magnetisation, so-called small magnetic grains. Assuming spherical grains the grain diameter is estimated to be 10±6 nm. The existence of small magnetic grains was also observed in recently performed small angle neutron scattering (SANS) experiments. From the measured depolarisation it appeared that the two sub-layers can be considered as two independent, or magnetically uncorrelated layers. The magnetic interaction between the layers, if present, is too weak to have an influence on the measured depolarisation. The two layers are magnetically not identical; the magnetic thicknesses i.e. $M_n h$ differ by 12%. The domain period in the longitudinal direction of the tape is 210 nm, i.e. $\approx 2.25$ times the thickness of one single layer. ND and SANS and also polarised neutron reflectometry (PNR) provide unique and detailed information on the magnetic and nuclear structure of thin film magnetic media such as ME-tape and CoCr-films. When this information is combined with the results of magnetisation measurements, Auger-spectroscopy, magnetic force microscopy and torque-measurements etc. it will lead to a better understanding of the magnetic properties of the tape.

In a series of thermally demagnetised poly-crystalline MnZn-ferrites with grain sizes varying between 0.3 and 30 $\mu$m and unaltered composition the magnetic domain size was measured. A region is found for grain sizes between 0.3 and 3.8 $\mu$m where the domain size equals the grain size; no domain walls are present. These grains are monodomains. For grain sizes larger than 3.8 $\mu$m the domain size is significantly smaller than the grain size; a domain wall begins to appear within the grains. Initially the grains consist of two domains and with increasing grain size the domain structure changes to a more complicated multi-domain structure. The results of the ND-experiments show that no magnetic texture is present in the thermally demagnetised ferrites, i.e. the magnetic induction within the ferrites is oriented at random.

In a set-up composed according to the $3p-2s$ method, i.e. any set-up (ND-instrument, reflectometer and diffractometer etc.) composed of two polarisers, two spin-flippers and a polarising sample/device, the net polarising power of the sample can be directly determined from the measured intensities. The depolarisation of the beam occurring in the set-up is included in the polarising power obtained for the two polarisers. The only restriction is, however, that the interaction is such that it is a non spin-flip process. In the case of magnetic neutron scattering the ratio between the nuclear and magnetic structure factor can be determined from the polarising power of the sample. If the interaction is such that it involves both a spin-flip as well as a non-spinflip process, information can be obtained on the components of the local magnetic induction that are both perpendicular to the neutron momentum transfer and to the direction of the initial polarisation from the measured intensities.
In conclusion, concerted research activities using the three available (polarised) neutron techniques at IRI-Delft (ND, SANS and PNR) and with other characterisation techniques will lead to a better understanding of magnetic recording. Moreover, it will help to push the recording technology to recording densities over $\approx 5$ gigabit/in$^2$ at the time we celebrate the 100$^{th}$ anniversary of the first demonstration of magnetic recording, the telegraphone, by the Danish scientist Valdemar Poulsen.
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Neutron Depolarisation in Magnetic Recording Materials

by Pieter Tjeerd POR

Summary

In this thesis the three-dimensional neutron depolarisation (ND) technique was applied to study the magnetic properties of magnetic recording materials. These materials are used nowadays, or will be used in future, as materials at which information/data is stored, i.e. the medium, or as materials used to write and read information, i.e. the recording head. The magnetic correlations within a sample and the mean magnetisation of a sample determine the change in the polarisation of a polarised neutron beam after transmission through the sample. In order to apply ND to this study and also to the study of the magnetic properties of other materials, the ND set-ups SP and KP were upgraded by installing new polarisers and analysers. For the SP a new depolarisation module was designed and installed. This module allows one to perform fully automated and computer controlled measurements of the depolarisation as a function of the position and transmission angle of the neutron beam in the sample. The rotation compensation method and a spin-echo coil system (SpECS) were developed, tested and installed in the SP. Both the spin-echo method and the SpECS are based on the principle of neutron spin-echo and allow three-dimensional ND-experiments in magnetised media using the complete thermal neutron spectrum.

The experimental sections of this thesis can be subdivided into two parts. In the second part the ND-technique was applied to study materials used for recording head applications. In the first part materials were studied which are used for the recording media. Using the SpECS, three-dimensional ND-experiments in high magnetic fields were performed for the first time ever. In this experiment the magnetic correlations between Co-modified γ-Fe₂O₃ particles embedded in a solid matrix were studied in fields up to 0.5 T. It appeared that, for this sample, the correlations originate from
variations in the particle density within the matrix. These variations can be considered as 'quasi particles' that magnetise differently as a function of the applied field. In the demagnetised state after magnetisation, the correlation length exceeds 2 – 3 times the mean particle size. This indicates the presence of positive correlations between the particles which give rise to the increased correlation length.

For a sample consisting of micro- or nano-crystalline barium hexaferrite particles, prepared by the glass-crystallisation method at Institut für Physikalische Hochtechnologie (IPHT) in Jena (BRD), the magnetic correlations were studied as function of the remanent magnetisation after a pulsed magnetic field. With increasing remanent magnetisation correlations between particles are induced. These correlations are broken in the magnetisation reversal process for the nano-crystalline particles, whilst the correlations are maintained for the micro-crystalline particles. This difference is likely to be caused by the non-magnetic layer on top of the nano-crystalline particles which screens the interaction between the particles. Moreover, the interactions are stronger between the micro-crystalline particles than between the nano-crystalline particles because micro-crystalline particles are in general larger than the nano-crystalline particles.

After milling a particulate dispersion of Co-modified \(\gamma\)-Fe\(_2\)O\(_3\) particles in a bead mill for 90 min no significant change in the magnetic correlation length is observed after different magnetic treatments. However, a significant increase in the magnetic correlation length was observed in the first stages of the milling process. This indicates that the initial clusters of particles are broken upon milling and that the dispersion is well dispersed after 90 min of milling. Moreover, it was found that the dispersion remains stable and well dispersed over a period of two weeks after preparation. Monte-Carlo computations and ND-simulations were carried out to study the three-dimensional micro-structure of a particulate dispersion. The results of the model for the zero field configuration, i.e. virginal state, agrees well with the experimental results; the correlation length approaches the mean particle size which is expected for a randomly oriented particle system that is not subjected to an orienting magnetic field. The results of the computations on the magnetised state could not be correlated with the experiments, because the simulated magnetic treatments were not identical to the magnetic treatments applied in the experiment. This comparison can now easily be made using carefully chosen samples, i.e. similar particle configurations, magnetic treatments, and using the SpECS and the rotation compensation method.

From the angular dependence of the depolarisation in a commercially available two-layer Hi8 TDK Metal Evaporated VCR-tape it appeared that the two sub-layers are not magnetically identical. The domain size in the longitudinal direction of the tape approaches the thickness of a single layer. The interaction between the two layers, if present, is too weak to have an influence on the depolarisation. For the ND-experiments the tape can be described by two independent or uncorrelated single layers. The results of the ND-experiments and the Small-Angle Neutron Scattering (SANS) experiments, performed at LOQ located at ISIS Didcot (UK), revealed the existence of small magnetic grains within the tape. The mean diameter of these grains is estimated to be
(10±6) nm.

In the next part of the thesis, the ND-technique was used to study, in co-operation with the Philips Research Laboratory Eindhoven (NL), the relation between the grain size and domain size in MnZn-ferrites. MnZn-ferrites are amongst others used as materials for recording heads. The results showed that in a series of thermally demagnetised poly-crystalline MnZn-ferrites with grain sizes ranging between 0.3 and 30 μm and unaltered composition a region exists for grain sizes below 3.8 μm where the grains are mono-domain. Above this critical grain size the grains become multi-domain. In both the mono- as well as the multi-domain ferrites no magnetic texture was found.

Thus far, the applications of the ND-technique to study the magnetic properties of magnetic materials were described. In chapter 8 of this thesis, a description is given of the principles and applications of the '3p-2s' method in the fields of magnetic elastic neutron scattering, polarised neutron reflectometry and neutron depolarisation. The most eye-striking characteristic of this method is the natural separation of the depolarisation and the polarising power of a polarising sample/device positioned between two polarisers and two spin flippers. This allows, for instance in a magnetic elastic scattering experiment, for the determination of the ratio of the nuclear and magnetic structure factor without any corrections for depolarisation of the beam occurring in the set-up.

In the final chapter of this thesis some conclusions and final remarks are given.
Neutronen Depolarisatie in Materialen voor Magnetische Registratie

door Pieter Tjeerd POR

Samenvatting (Summary in Dutch)

In dit proefschrift is de drie-dimensionale neutronen-depolarisatie (ND) techniek toegepast voor het bestuderen van de magnetische eigenschappen van materialen voor magnetische registratie. Deze materialen worden nu, of in de toekomst, toegepast als materialen waarin de informatie/data wordt opgeslagen, het *medium*, of als materialen waarmee informatie weggeschreven of gelezen kan worden, de zogenaamde *lees/schrijfkop*. Magnetische correlaties in het preparaat en de gemiddelde magnetisatie van het preparaat veroorzaken een verandering in de polarisatie van een gepolariseerde neutronenbundel na transmissie door het preparaat. Om de ND-techniek toe te passen voor deze studie en voor het bestuderen van de magnetische eigenschappen van andere materialen zijn de ND-opstellingen SP en KP verbeterd door de installatie van nieuwe polarisatoren en analysatoren. Voor de SP is een nieuwe depolarisatiemodule ontworpen, getest en geïnstalleerd. Deze module maakt volledig geautomatiseerde metingen van de depolarisatie mogelijk als een functie van de positie en/of transmissiehoek van de neutronenbundel door het preparaat. De rotatie compensatie methode en een spin-echo spoelensysteem (SpECS) zijn ontwikkeld, getest en geïnstalleerd in de SP. Zowel de rotatie compensatie methode als het SpECS zijn gebaseerd op het principe van neutronen spin-echo en maken het mogelijk om drie-dimensionale ND-experimenten uit te voeren in gemagnetiseerde materialen met het gebruik van het complete thermische neutronenspectrum.

De hoofdstukken van dit proefschrift waarin de experimentele resultaten worden beschreven kunnen opgedeeld worden in twee delen. In het tweede gedeelte is de
ND-techniek toegepast op materialen die hun toepassing vinden in lees/schrijf-koppen. In het eerste gedeelte is de techniek toegepast op materialen voor het opslagmedium zelf. Met behulp van de SpECS zijn voor de allererste keer drie-dimensionale ND-experimenten uitgevoerd in de aanwezigheid van sterke magnetische velden. In dit experiment zijn de magnetische correlaties tussen kobalt-gemodificeerde $\gamma$-Fe$_2$O$_3$ deeltjes ingebouwd in een vaste matrix bestudeerd in velden tot 0.5 T. Het blijkt dat de magnetische correlaties voornamelijk worden veroorzaakt door variaties in de dichtheid van de deeltjes in de matrix. Deze variaties kunnen opgevat worden als 'quasi-deeltjes' die verschillend magnetiseren in het aangelegde magnetisch veld. In de gedemagnetiseerde toestand na magnetisatie is de gemeten correlatielengte twee tot drie maal zo groot als de gemiddelde deeltjesafmetingen. Dit betekent dat tussen de deeltjes een positieve interactie bestaat die de vergrote correlatielengte veroorzaakt.

In een preparaat bestaande uit micro- of macro-kristallijn barium hexa-ferriet deeltjes, welke vervaardigd zijn met behulp van de glas-kristallisatie methode in het Instituut für Physikalische Hochtechnologie (IPHT) in Jena (BRD), zijn de magnetische correlaties bestudeerd als een functie van de remanente magnetisatie na magnetisatie met een korte magnetische puls. Met toenemende remanente magnetisatie worden er tussen de deeltjes correlaties geinduceerd. Deze correlaties worden afgebroken in het magnetisatie omkoers proces van de nano-kristallijne deeltjes, terwijl voor de macro-kristallijne deeltjes de correlaties behouden blijven. Dit verschil wordt in alle waarschijnlijkheid veroorzaakt door de aanwezigheid van een niet-magnetische laag om de nano-kristallijne deeltjes; deze laag veroorzaakt een afname in sterkte van de interacties tussen de deeltjes. Ook zijn in het algemeen de interacties sterker tussen de micro-kristallijne deeltjes dan tussen de nano-kristallijne deeltjes omdat de micro-kristallijne deeltjes groter zijn dan de nano-kristallijne deeltjes.

Na anderhalf uur malen van een dispersie bestaande uit kobalt-gemodificeerde $\gamma$-Fe$_2$O$_3$ deeltjes is geen significante verandering van de correlatielengte waargenomen na diverse magnetische behandelingen van de preparaat. Deze verandering was wel aanwezig in de eerste fase van het maalproces. Uit deze observatie volgt dat de clusters van deeltjes, die aanvankelijk aanwezig waren in de dispersie, worden afgebroken tijdens het malen. Na 90 minuten zijn bijna alle clusters afgebroken en zijn de deeltjes goed gedispergeerd in de dispersie. Verder blijkt ook dat de dispersie stabiel blijft en de deeltjes goed gedispergeerd blijven tot zeker twee weken na vervaardiging. Monte-Carlo berekeningen en ND-simulaties zijn uitgevoerd met als doel het bestuderen van de drie-dimensionale micro-structuur van een magnetische deeltjes dispersie. Het gebruikte model in de MC-berekeningen voor de maagdelijke dispersie komt goed overeen met de experimentele waarnemingen. De correlatielengte is in de orde grootte van de gemiddelde deeltjes afmetingen wat ook te verwachten is voor een dispersie bestaande uit isotroop georiënteerde deeltjes. Geen experimenten, met vergelijkbare magnetische behandelingen, waren beschikbaar om de resultaten van het model te testen in het geval de dispersie gemagnetiseerd is of zich in een remanente toestand bevindt. Een vergelijking tussen experiment en model is nu wel mogelijk door een zorgvuldige keuze van het preparaat en het gebruik maken van de rotatie compensatie methode en het
SpECS.

Uit de hoekafhankelijkheid van de depolarisatie in een commerciële twee-laags Hi8 TDK Metal-Evaporated (ME) video-tape blijkt dat de twee lagen magnetische gezien niet identiek zijn. De domeinafmetingen in de longitudinale richting van de tape zijn ongeveer gelijk aan de dikte van een enkele laag. De interactie tussen de twee lagen is, indien aanwezig, te zwak om een invloed te hebben op de gemeten depolarisatie. Voor de ND-experimenten kan de twee-laags tape opgevat worden als twee onafhankelijke op elkaar gestapelde enkel-laags tapes. Uit de ND-experimenten en de kleine-hoek neutronen verstrooingsexperimenten (SANS), welke uitgevoerd werden op het SANS-instrument LOQ bij ISIS, Didcot (UK), volgt dat de tape bestaat uit kleine magnetische deeltjes met een afmeting van ongeveer \((10 \pm 6) \text{ nm}\).

In het tweede gedeelte van het proefschrift is de ND-techniek toegepast voor de bestudering van de relatie tussen de domeingrootte en korrelgrootte in MnZn-ferreten. Deze experimenten zijn uitgevoerd in samenwerking met het Philips Natuurkundig Laboratorium in Eindhoven (NL). MnZn-ferreten vinden onder andere hun toepassing als materialen voor de fabricage van lees/schrijf-koppen. In een serie van thermisch gede-magnetiseerde poly-kristallijn MnZn-ferreten met korrelgrootte tussen 0.3 en 30 \(\mu\text{m}\) en ongewijzigde chemische samenstelling bestaan ferreten met korrelgrootte tot 3.8 \(\mu\text{m}\) uit slechts één magnetisch domein. Boven deze kritische korrelgrootte wordt een meer gecompliceerde multi-domein structuur gevormd. Zowel in de één- als de multi-domein ferreten is geen magnetische textuur waargenomen.

Tot dusver zijn de toepassingen van de ND-techniek in de bestudering van magnetische eigenschappen besproken. In hoofdstuk 8 wordt een beschrijving gegeven van de principes en toepassingen van de 'Sp-2s' methode op het gebied van magnetische elastische neutronenverstrooing, gepolariseerde neutronenreflectometrie en neutronen-depolarisatie. De meest in het oog springende eigenschap van de methode is de scheiding van de depolarisatie in de opstelling van het netto-polariserend vermogen van een polariserend preparaat/apparaat dat gepositioneerd is tussen twee andere polarisatoren en twee spinflippers. In magnetische elastische neutronenverstrooings experimenten maakt deze methode het bijvoorbeeld mogelijk om de verhouding tussen de nucleairen magnetische structuurfactor te bepalen zonder correcties voor de depolarisatie van de op het preparaat invalende bundel.

In het laatste hoofdstuk van dit proefschrift worden enkele conclusies getrokken en enkele slotopmerkingen geplaatst.
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Curriculum Vitae


List of Publications


