NEUTRON DEPOLARIZATION STUDY OF STATIC AND DYNAMIC MAGNETIC PROPERTIES OF FERROMAGNETS

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NEUTRON DEPOLARIZATION STUDY
OF STATIC AND DYNAMIC
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OF FERROMAGNETS

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CHAPTER I

INTRODUCTION

Among the experimental methods used to study the properties of ferromagnets the neutron depolarization experiment belongs to the less commonly applied ones. The need for a neutron source such as a research reactor or a spallation source limits the number of neutron depolarization instruments. The neutron depolarization technique is in the first place a transmission method and hence it can suitably be used at a low power research reactor. Usually this method is applied to study ferromagnetic materials on a micron scale while most other experimental techniques measure magnetic properties either on an atomic or a macroscopic scale.

Neutron depolarization instruments are known to exist at research reactors in Braunschweig, Leningrad, Vienna and Delft and at the neutron spallation source in KENS (Japan). The direction of polarization and analysis of the neutron beam can be calibrated to an orthogonal system of reference usually within a few degrees. Besides time-dependent neutron depolarization analyses have been carried out until now at the instruments in Delft and Vienna with a maximal resolution in time of about 10 μs and 1 ms, respectively. The high time resolution of the instrument used here allows us to study fast magnetization changes.

The large penetration depth of the thermal neutrons in many materials together with the high sensitivity of the neutron magnetic moment for small magnetic inductions gives this method unique possibilities in the investigation of ferromagnets. In a neutron depolarization experiment the change in the state of polarization is measured which is caused by the magnetic interaction between the spin of the neutrons and the matter investigated. The second and third chapter review those aspects of the theory of neutron depolarization and about the experimental set-up as far as they are relevant for the later chapters.

In this thesis the neutron depolarization technique was applied to study static and dynamic magnetic properties in amorphous and crystalline ferromagnets. The subjects studied are concerned with 'domain
structure in magnetically weak uniaxial amorphous ferromagnetic ribbons', 'static critical behaviour at the ferromagnetic-paramagnetic phase transition', 'small magnetic anisotropy in nickel near $T_c$' and 'magnetization reversal in conducting ferromagnets'. All chapters have appeared or are going to appear in scientific journals. This has the advantage that the chapters IV-VII can be read without knowledge of other ones. These different topics will be now introduced briefly.

1) DOMAIN STRUCTURE IN MAGNETICALLY WEAK UNIAXIAL AMORPHOUS FERROMAGNETIC RIBBONS.

During the last two decades special interest was attracted to the investigation of the domain structure in amorphous systems which find industrial application as soft magnetic materials. Due to the absence of long range structural order the easy axis of magnetization is determined by the stress distribution within the specimen. 'Well defined' domain structures can be introduced by the application of external stresses which offers new possibilities in the research on domain structures.

A subject which is in continuous discussion ever since the first domain model calculations in the thirties is the type of domain structure in a ferromagnet with a weak uniaxial anisotropy. The local magnetic induction in the volume is expected to be aligned parallel or antiparallel to the easy axis of magnetization, and the magnetic flux has to be closed near the surface. So far observations of this type of domain structure in conducting materials were limited to the surface using optical techniques. Since the neutron transmits the sample new information can be obtained about the magnetization distribution within the volume of the specimen. The high sensitivity allows one to measure the magnetic induction in thin layers which makes the application of the neutron depolarization experiments attractable to investigate the type of flux closure in a weak uniaxial ferromagnet.

In chapter IV neutron depolarization studies are presented which were performed on metallic amorphous ribbons with a stress induced magnetic anisotropy perpendicular to the ribbon plane. Beside the investigation of the domain structure in zero field we have also studied
the change of the magnetization distribution due to the presence of a magnetic field which was applied in the ribbon plane. A detailed knowledge of the magnetization process is important to understand hysteresis losses in these materials.

2) STATIC CRITICAL BEHAVIOUR AT THE FERROMAGNETIC-PARAMAGNETIC PHASE TRANSITION.

Since the renormalization theory was introduced by Wilson, Kadanoff and Fisher critical exponents at second order phase transitions were calculated very precisely by various authors. These exponents are predicted to depend upon the universality class of the system only. To measure these exponents experimental data should be located very close to the critical point because the calculated exponents are valid only just at the phase transition. Since measured data are always located outside the critical point the analysis should involve correction to the scaling terms. From the knowledge of these correction terms it becomes possible to derive the asymptotic value of the exponent studied.

So far the determination of these small correction terms in ferromagnets was prohibitive due to a lack in precision of the measuring techniques used. The high precision with which the magnetic induction is sensed by the neutron should make it possible to measure these correction terms or at least an upper limit of the size of the correction terms should be determined. The first attempts to investigate the ferromagnetic-paramagnetic phase transition by neutron depolarization experiments were hindered by demagnetizing fields of the elliptically shaped specimens used. In the neutron depolarization experiments which are described here ring shaped iron and nickel specimen were used in order to minimize the demagnetizing fields. The main subject of chapter V will be to discuss the possibilities that the neutron depolarization technique offers to investigate the critical behaviour and in particular to obtain the asymptotic values of the critical exponents.
3) SMALL MAGNETIC ANISOTROPY IN NICKEL NEAR $T_c$.

Various measurements of the crystalline anisotropy in nickel indicate changes of the easy axis of magnetization from the [111]-crystallographic axis at room temperature to other directions at higher temperature. Upon approaching the phase transition at $T_c$ the anisotropy becomes very small and nearly all experimental methods are not sufficiently sensitive to measure the anisotropy within a few degrees close to $T_c$. Chapter VI is devoted to a discussion of neutron depolarization experiments in which the approach of the mean magnetic induction towards its saturation value is measured as a function of an applied field and from which information can be derived about the strength of the anisotropy. The measurements were performed within a range a few degrees Kelvin below $T_c$.

4) MAGNETIZATION REVERSAL IN CONDUCTING FERROMAGNETS.

The physical origin of magnetic losses in conducting ferromagnets is the dissipation of heat by eddy currents. The eddy currents arise from the voltages induced by changes in the magnetic induction. To predict these losses a detailed knowledge is needed of the mechanism of the magnetization reversal.

The time dependent neutron depolarization analysis was demonstrated to be a powerful tool to investigate the spatial and temporal evolution of the reversal process inside the volume of a ferromagnet. The information obtained is on a length scale of about a few microns and a time scale in the order of ten microseconds. In chapter VII measurements are presented of the magnetization reversal in a nickel picture-frame single crystal. These measurements are an extension to former neutron depolarization studies which were performed at FeSi (3.5%) specimen.
CHAPTER II

SOME BASIC CONCEPTS OF NEUTRON DEPOLARIZATION

CONTENTS

1. INTRODUCTION
2. POLARIZATION OF A NEUTRON BEAM
3. DEPOLARIZATION MATRIX
4. EQUATION OF MOTION FOR THE POLARIZATION
5. DEPOLARIZATION BY A MAGNETICALLY SATURATED FERROMAGNET
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7. DEPOLARIZATION BY A FERROMAGNET CLOSE TO MAGNETIC SATURATION

1. Introduction

During the traversal of a polarized neutron beam through a ferromagnetic material a change in the state of polarization $\mathbf{P}$ arises from the Larmor precession of $\mathbf{P}$ around the local magnetic induction $\mathbf{B}$. Any change in $\mathbf{P}$ i.e. a reduction in the degree of $\mathbf{P}$ as well as a change in the orientation of $\mathbf{P}$ will be referred to as depolarization henceforth. While the depolarization is completely determined by the magnetization distribution of the specimen studied the reverse is not true.

Various authors have given an interpretation of the neutron depolarization by ferromagnets in terms of a few domain parameters which give a statistical characterization of the magnetization distribution [1-7]. Since all these calculations are made using specific assumptions about the domain structure the application of these analyses to experimental data has to be considered carefully. In this introductory chapter basic definitions are presented and those calculations of the depolarization by specific domain structures are illustrated which are relevant for the later chapters described in this thesis. The aim is to give some qualitative ideas about this subject to those readers which
are not familiar with the neutron depolarization analysis. Comprehensive treatments are found in ref. [1-7].

2. Polarization of a neutron beam

The neutron is an uncharged particle with a spin \( s = 1/2 \). The polarization vector \( \hat{P} \) of a neutron beam is defined by the excess of one spin state over the other:

\[
\hat{P} = \frac{I_+ - I_-}{I_+ + I_-} \hat{e}_p
\]

with \( I_+ \) and \( I_- \) the intensities of the neutrons with the spin \( \hat{s} \) parallel and antiparallel to \( \hat{e}_p \), respectively. The unit vector \( \hat{e}_p \) points in the direction along which \( I_+ - I_- \) takes on a maximum. The degree of \( \hat{P} \) is identical to \( |\hat{P}| \) which equals one for a fully polarized beam and becomes zero for the completely depolarized neutron beam.

3. Depolarization matrix

In a three-dimensional neutron depolarization analysis the polarization \( \hat{P} \) is adjusted and, after traversal through a specimen, independently analyzed along three orthogonal directions (x,y,z). The depolarization caused by the specimen can then be expressed by a 3x3 depolarization matrix \( D \). If one denotes the intensity of the fully depolarized beam by \( I_S \) and the intensity of the unaffected beam by \( I_0 \) the (i,j) matrix element of \( D \) becomes

\[
D_{i,j} = \frac{I_i - I_{ij}}{I_S - I_0}
\]

with \( I_{i,j} \) the intensity measured with polarizer and analyzer pointing along \( j \) and \( i \) (\( i,j = x,y,z \)). The depolarization is always within the range from -1 to +1.
4. Equation of motion for the polarization

The equation of motion for the polarization vector \( \mathbf{P} \) of a neutron beam in the presence of a magnetic induction \( \mathbf{B} \) is

\[
\frac{d\mathbf{P}}{dt} = \gamma (\mathbf{P} \times \mathbf{B})
\]  

(3)

with the gyromagnetic ratio \( \gamma = 1.8 \times 10^8 \text{ s}^{-1} \text{T}^{-1} \) for the neutron. In a ferromagnet the local magnetic induction \( \mathbf{B} \) is only homogeneous within one domain. Hence the magnetic induction becomes time dependent during the traversal of the neutrons through the specimen. Eq. (3) is solved by

\[
\mathbf{P}_f = \mathbf{P}_0 + \gamma \int_0^t [\mathbf{P}(t') \times \mathbf{B}(t')] dt'
\]

(4)

The vector \( \mathbf{P}_0 \) defines the polarization of the incident beam. The integration can be carried out by successive iteration

\[
\mathbf{P}_f = \sum_{n=0}^{\infty} \gamma^n \int_0^t \ldots \int_0^{t(n-1)} [\mathbf{P}_0 \times \mathbf{B}(t') \times \ldots \times \mathbf{B}(t^{(n)})]
\]

(5)

The final polarization vector is obtained by averaging (5) over the beam cross section. This solution will now be considered for a few magnetization distributions in a ferromagnet which are relevant for later chapters of this thesis.

5. Depolarization by a magnetically saturated ferromagnet

The local magnetic induction has the value \( \mathbf{B} \) throughout the entire volume of the specimen. Hence eq. (5) becomes

\[
\mathbf{P}_f = \sum_{n=0}^{\infty} \frac{(\gamma t)^n}{n!} \ldots [\mathbf{P}_0 \times \mathbf{B}] \times \ldots \times \mathbf{B}
\]

(6)

\( n \times \text{vector product with} \ \mathbf{B} \)
which shows that the component of $\hat{P}_0 \parallel \hat{B}$ is conserved. Choosing $\hat{B}$ and $\hat{P}_0$ to point along $\hat{e}_z$ and $\hat{e}_x$, respectively eq.(6) can be written as

\[
\hat{P}_f = \sum_{n=0}^{\infty} (-1)^n \frac{(\gamma Bt)^{2n}}{(2n)!} \hat{e}_x + \sum_{n=0}^{\infty} (-1)^n \frac{(\gamma Bt)^{2n+1}}{(2n+1)!} \hat{e}_y
\]

which is identical to a rotation of $\hat{P}_0$ around $\hat{B}$. Using the generator of the infinitesimal rotation $\hat{R} = \hat{r} \times \hat{n}$ eq.(6) becomes

\[
\hat{P}_f = e^{\gamma Bt} \hat{P}_0 .
\]

The angle of rotation is

\[
\phi = \gamma Bd/v .
\]

In eq.(9) the traversal time was substituted by the ratio of the thickness $d$ of the specimen along the direction of propagation of the beam and the velocity $v$ of the neutrons. Most experiments which are described in the later chapters were performed with a monochromatic beam of wavelength $\lambda = 0.16\text{nm}$ which corresponds to a neutron velocity of about $v = 2500 \text{m/s}$. The Larmor precession angle amounts then to

\[
\phi = 7.3 \times 10^4 \text{rad T}^{-1} \text{m}^{-1}
\]

which indicates the high sensitivity of this method for small magnetic inductions.

6. Depolarization by an unmagnetized ferromagnet close to the phase transition

The local magnetic induction is randomly oriented in space but has a constant strength $B_0$. Application of eq.(5) and considering only the first three terms yields
The second term vanishes since the specimen is assumed to be unmagnetized. Assuming further that \( \hat{B}(t') \) is only correlated to \( \hat{B}(t'') \) for \(|t' - t''| < \tau\), where \( \tau \) is the time of traversal of neutrons with velocity \( v \) through the mean domain size \( \delta \), eq. (10) can be rewritten as

\[
\hat{P}_f = \hat{P}_0 + \gamma \tau |t'| \left[ [-\hat{P}_0(\hat{B}) + \hat{B}(\hat{P}_0)] \right].
\]  

(11)

Averaging \( \hat{B}(\hat{P}_0, \hat{B}) \) over all orientations of \( \hat{B} \) yields \( \hat{P}_0 B_s^2 / 2 \). Hence one obtains

\[
\hat{P}_f = \hat{P}_0 (1 + (-\gamma B_s^2 \tau / 2)).
\]  

(12)

The last equation is only valid if \( \gamma B_s^2 \tau \) is small compared to one which is expected to be the case for ferromagnets close to \( T_c \) because \( B_s \) approaches zero. It is shown e.g. in [1], that the depolarization by an unmagnetized ferromagnet close to \( T_c \) can be expressed by

\[
D = \left| \frac{\hat{P}_f}{\hat{P}_0} \right| = e^{-\gamma B_s^2 \delta d / 2v^2}.
\]  

(13)

Eq. (12) represents the first two terms in the expansion of the exponential function of eq. (13). In eq. (13) the times \( t \) and \( \tau \) were substituted by \( d/v \) and \( \delta / v \), respectively. Eq. (13) was applied to determine the quantity \( B_s^2 \delta \) from the depolarization close to \( T_c \) at zero magnetic field (see chapter V).

7. Depolarization by a ferromagnet close to magnetic saturation

Close to magnetic saturation the local magnetic induction can be expressed as
\[
\hat{B}(t) = \langle \hat{B} \rangle + i \hat{B}_\parallel(t) + i \hat{B}_\perp(t)
\]  

(14)

with \( \langle \hat{B} \rangle \) the mean magnetic induction of the specimen. The quantities \( \hat{B}_\parallel \) and \( \hat{B}_\perp \) denote the parallel and perpendicular deviations of \( \hat{B}(t) \) from \( \langle \hat{B} \rangle \). Both, \( \hat{B}_\parallel \) and \( \hat{B}_\perp \) are much smaller than \( \langle \hat{B} \rangle \). The first order solution by eq.(5) is obtained by neglecting these small quantities to be the pure Larmor precession around \( \langle \hat{B} \rangle \). Since \( \hat{B}_\parallel \) and \( \hat{B}_\perp \) average to zero they are expected to have no effect on the orientation of \( \hat{B}_F \). It is discussed in \([3,4,6,7]\) that the "fluctuations" of \( \hat{B}(t) \) around \( \langle \hat{B} \rangle \) cause a decrease in the degree of the polarization. Hence the measurement of the Larmor precession angle \( \phi \) determines the mean magnetic induction \( \langle \hat{B} \rangle \). This result was used to measure the approach to magnetic saturation of the mean magnetic induction in chapter VI.

References:

CHAPTER III

EXPERIMENTAL SET-UP

1) Neutron depolarization set-up [1,2]

The neutron depolarization measurements were performed on the crystal polarimeter at the 2 MW swimmingpool reactor HOR of the Interuniversitair Reactor Instituut in Delft. The principal set-up is sketched in fig. 1. The white beam is monochromized ($\lambda = 0.16$ nm) and polarized ($P = 0.91$) by Bragg reflection at a single crystal of Fe$_3$Si (or a Heussler alloy) magnetized near to saturation. The direction of polarization of the beam with its direction of propagation along the x-axis can be adjusted by a polarization turner to one of the three orthogonal x, y or z axes. Inside the polarization turner a homogeneous magnetic field is generated in the y-z plane by currents through two coils so as to rotate $\hat{P}$ into the desired direction. Next the beam passes through a sample, which is positioned in a box of soft iron and $\mu$-metal in order to shield against external magnetic fields. The magnetic induction $B$, measured by a fluxmeter, is less than $2 \times 10^{-5}$ T inside the box. Subsequently the beam passes a second polarization turner (similar to the first one) by means of which each component x, y or z of the polarization vector $\hat{P}$ can be presented parallel to the axis of magnetization of the analyzing crystal (similar to the polarizing crystal). Two guide fields ($\hat{H} \parallel z$), one between the polarizing crystal and the first polarization turner, the other between the second polarization turner and the analyzing crystal, serve to keep the component of $\hat{P}$ parallel to $\hat{H}$. After Bragg reflection at the analyzing crystal the neutrons are detected by a $^6$Li scintillation or a $^3$He-gas counter each with an efficiency of 50% ($\lambda = 0.16$nm). A BF$_3$ proportional counter with a 20% efficiency monitors the intensity at the detector against a preset count of neutrons that passed through the sample.
FIG. 1. Principal set-up. The arrows indicate the state of polarization of the neutron beam. The symbols $D_{1,2}$, $G$ and $M_{p,a}$ denote the polarization turners, guide fields and polarizing (analyzing) crystals. The large arrows indicate the magnetization directions of $M_{p,a}$. The sample holder, monitor counter and detector are denoted by $S$, $M$ and $T$.

The neutron flux at the sample position is about $8 \times 10^4 \text{ n cm}^{-2}\text{s}^{-1}$ and the flux of the depolarized beam at the detector position is about $10^4 \text{ n cm}^{-2}\text{s}^{-1}$. The polarization vector of the neutron beam can be calibrated within one degree to the laboratory system of reference.

Time dependent phenomena can be studied by repeating the process (e.g. a magnetization reversal) periodically in time. In one period the intensity is counted in successive time channels of predefined length $\Delta t$. The number of measuring cycles is determined by the desired counting statistics. The time resolution is about 8 $\mu$s and results from the wavelength spread $\Delta \lambda$ of the beam and the time resolution of the detector.
2) Furnace and temperature control

All experiments above room temperature were performed with the samples positioned in the vacuum furnace shown in Fig. 2. The furnace consists basically of a cylindrical heater from stainless steel, called the inner furnace, which is surrounded by a temperature controlled brass mantle, the outer furnace.

A mechanical fore pump and a turbomolecular pump evacuate the furnace to a pressure below $10^{-5}$ torr measured at the entrance of the furnace. The temperature of the outer furnace is stabilized within 0.1 K to a few ten degrees above room temperature by a temperature controlled water flow circuit. The thermal radiation shield between the inner and the outer furnace consists of three closed cylinders of stainless steel.

FIG. 2. Drawing of the vacuum furnace used for the temperature dependent measurements (see text).
A detailed sketch of the inner furnace is shown in fig. 3. The boron nitride sample holder is positioned in the cylindrical heater and the two cylinder faces are closed by 0.5 mm thick discs of stainless steel. The sample holder is built up from four BN disks, with a diameter $\phi = 32$ mm and a total length $l = 20$ mm. The two inner ones sandwich the ring shaped sample and provide the possibility to wind a toroidal coil (783 turns/meter) around the sample. All BN-disks have a sickle shaped opening just in front of a small part of the sample which defines the diaphragm for the neutrons.

To minimize radiation losses from the sample surface, located at the diaphragm, to the surrounding a few thin radiation shields of molybdenum are built in the diaphragm openings.

FIG. 3. Sketch of the inner furnace (see text).
Two chromel alumel thermocouples are used as temperature sensors. The thermovoltage is measured with a precision of 0.1 μV by a digital voltmeter (Keithley 181). A proportional-integral feedback circuit, controlled by a Fortran program on a PDP-computer [3] stabilizes the temperature within ± 3 mK. The long-term temperature drift was smaller than 25 mK in 24 h. The power needed to heat the inner furnace to a temperature of 1100 K is about 90 Watt.

The field strength that can be applied is limited by the heat dissipation of the current in the field coil. This source of heat introduces some temperature gradient in the sample holder and causes an uncertainty in the temperature measurement of the sample which is derived from the temperature readings of the two thermocouples located at two different positions in the sample holder. The experiments on the iron sample (T = 1000 K) were performed with a field coil consisting of a 0.3 mm thick molybdenum wire and the maximal field strength that could be applied was a few hundred A/m. For the measurements on the nickel sample (T ≤ 700 K) a copper wire was used and fields up to 1500 A/m could be generated.

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CHAPTER IV

DOMAIN STRUCTURE IN MAGNETICALLY WEAK UNIAXIAL AMORPHOUS FERROMAGNETIC RIBBONS

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MAGNETIZATION REVERSAL IN AMORPHOUS RIBBONS STUDIED BY NEUTRON DEPOLARIZATION

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The magnetization process in an amorphous ribbon with an easy axis perpendicular to its plane has been studied with the three-dimensional neutron depolarization technique. A simplified model of three layers is used, describing the magnetization distribution in the bulk and the two closure domain structures at the surface. Our analysis with this model gives direct experimental evidence that the magnetization process due to an applied magnetic field in the ribbon plane is dominantly accounted for by rotation processes of the local magnetization in the bulk. The influence of different types of closure domain structures on the magnetization distribution during the reversal process are discussed and compared with the experimental results.

1. Introduction

Amorphous ferromagnetic ribbons show excellent soft magnetic and high elastic properties [1]. A study of the domain structure in these ribbons during the quasistatic magnetization process is not only valuable because of the widespread technical applications of these materials, but from a fundamental point of view also.

Due to the generally high magnetostriction and the absence of long range structural order, the magnetic anisotropy in the amorphous systems mainly originates from mechanical stresses. These stresses, in addition to the sample shape, predominantly determine the magnetic domain structure. From the domain structure optically observed at the surface [2,3], an inhomogeneous distribution of compressive and tensile stresses in the plane of as-quenched ribbons is indicated, which have been introduced in the rapid quenching during their preparation.

It is simplest and most instructive to study the domain structure during the quasistatic magnetization process in the compressive and tensile stress regions separately. In this paper, we restrict ourselves to the investigation of amorphous ribbons in a compressive stress state. Hence our measurements have been carried out on an annealed amorphous ribbon, with a positive magnetostriction under a biaxial compressive stress in the plane of the ribbon. In view of the "simple" domain structure where the local magnetization in the central part (bulk) of the ribbon is perpendicular to the ribbon plane and closure domains at the surface provide flux closure (Landau-Lifshitz domain structure [4]), the three-dimensional neutron depolarization technique [5] is a versatile tool to study the quasistatic magnetization reversal.

Recently, the unannealed amorphous ribbon Metglass 2826 has been studied using this technique and the results have been analyzed in terms of a magnetic domain model describing a three layer stress distribution across the thickness of the ribbon [6]. Since magneto-optical Kerr observations indicate an inhomogeneous distribution of compressive and tensile stresses at the surface [2,3], there are doubts as to the validity of the last model. The present paper may also provide useful information for the interpretation of neutron depolarization in an unannealed amorphous ribbon.

2. Specimen and experimental technique

Our neutron depolarization experiments have been performed on an annealed amorphous ribbon...
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(60 x 10 x 0.029 mm³) of chemical composition Fe₄₀Ni₁₀Pt₆B₆ supplied by "Vacuumschmelze Hanau". The ribbon was tightly glued with a thin layer of araldite between two aluminium platelets (1 mm thick) at a temperature around 100°C. After cooling down, a biaxial compressive stress is induced in the ribbon plane, due to the different thermal contraction of aluminium (2.5 x 10⁻⁵ K⁻¹) compared with that of the amorphous system (1.2 x 10⁻⁵ K⁻¹). In view of the positive magnetostriction, an easy axis of magnetization perpendicular to the plane of the ribbon is introduced. Here we will investigate the change of the magnetic domain structure in this uniaxial anisotropic system, under the influence of a magnetic field applied parallel to the long side of the ribbon.

Information about the magnetization distribution in the bulk can then be obtained from the three-dimensional neutron depolarization analysis [5]. This experimental technique is based on the Larmor precession of the polarization vector \( \mathbf{P} \) of a polarized neutron beam in a magnetic field:

\[
\frac{d \mathbf{P}}{dt} = \gamma (\mathbf{P} \times \mathbf{B}),
\]

where \( \mathbf{B} \) is the magnetic induction and \( \gamma \) the gyromagnetic ratio of the neutron. In the three-dimensional neutron depolarization technique, the polarization of the monochromatic beam (we use \( \lambda = 0.16 \text{ nm} \)) is adjusted \( (\mathbf{P}_i) \) and, after depolarization by the magnetization distribution in a ferromagnetic sample, analyzed independently \( (\mathbf{P}_j) \) in three orthogonal directions.

\[ \mathbf{P}_j = \mathbf{D} \mathbf{P}_i, \quad (i, j = x, y, z). \]

The depolarization matrix \( \mathbf{D} \) thus obtained is fully determined by the magnetization distribution in the transmitted part of the sample and provides a number of maximum 9 model parameters, by means of which the domain structure may be described.

3. Experimental results

First angular dependent neutron depolarization experiments have been carried out on the unmagnetized sample and an analysis has been performed to investigate the induced domain structure. In fig. 1a and b, \( D_{xx} \) and \( D_{zz} \) of the depolarization matrix are given as a function of the glancing angle \( \theta \), with the sample turned around the \( z' \) and \( y' \) axis, respectively. The value of \( D_{xx} \) near 1 at perpendicular transmission \( (\theta = 0°) \) in fig. 1a and b) is consistent with a magnetic anisotropy perpendicular to the ribbon plane, i.e. a neutron beam polarized in the \( x' \)-direction is practically not affected by the magnetization distribution in the transmitted part of the sample. From the variation in \( D_{zz} \) with \( \theta \) while rotating the ribbon around \( z' \), we determine a mean domain spacing \( D_0 = 5.3 \mu \text{m} \) along the \( y' \)-axis of the ribbon. Here we use a method described in [7]. Similarly, on rotating the ribbon around \( y' \), we find a mean domain spacing of \( = 16 \mu \text{m} \) along \( z' \). Hence the 180° domain walls in the bulk are directed predominantly near the \( z' \)-axis and the local magnetization in the closure domains prefers to lie near \( \pm y' \). The difference in the mean domain spacing of a factor of three along \( y' \) and \( z' \) indicates a magnetic anisotropy in the ribbon plane.

Subsequently, field dependent neutron depolarization experiments have been carried out on the same sample. We applied a periodic triangularly shaped field parallel to \( z' \), with an amplitude of 6500 A/m, choosing a time of 1 s for one period. During the field dependent measurements, the neutron beam transmitted perpendicular to the ribbon plane \( (\theta = 0°) \). The field current warmed up the ribbon to a temperature of 50–60°C, which results in a decrease in the induced magnetoelastic anisotropy of \( = 40\% \), compared with the aniso-

![Fig. 1. xx- and zz-depolarization elements as a function of \( \theta \) as indicated. \( \theta \): glancing angle of the neutrons.](image-url)
entropy present in our angular dependent measurements at room temperature. The value of $D_z$ is still near 1 in a zero field, indicating that the uniaxial magnetization distribution has remained along the $x'$ axis.

The field dependent neutron depolarization matrix $D$ has been analyzed, choosing a simplified four parameter model to describe the magnetization distribution in the ribbon. Our model involves a three layer structure, consisting of two surface layers representing the closure domains and one bulk layer (fig. 2). We confine the mean magnetization to the $z'$-component in all three layers. Hence the two surface layers with their local magnetizations in the $y'$-$z'$ plane are characterized by a mean magnetization $\langle m_{z's} \rangle$ (normalized to the spontaneous magnetization) and a mean direction cosine $\gamma_{z's} = \langle m_{z's}^2 \rangle = 1 - \gamma_{y's}$ (s: surface). In the bulk, the magnetization distribution is assumed to be in the $z'$-$x'$ plane, described in terms of $\langle m_{z'b} \rangle$ (b: bulk). We assume only rotation processes to take place in the bulk, as the applied field is perpendicular to the local magnetization and thus no domains are favoured energetically. Therefore, the magnetization in the bulk is fully determined by $\langle m_{z'b} \rangle$ with the direction cosine $\gamma_{z'b} = 1 - \gamma_{y'b}$.

The three diagonal elements and the $x'$-$y'$ element of the depolarization matrix provide these four parameters in this model of the magnetic domain structure. The other elements in the depolarization matrix serve to check consistency.

The field dependence of the mean magnetization $\langle m_{z'b} \rangle$ in the bulk layer is shown in fig. 3. The linear increase of $\langle m_{z'b} \rangle$ for a field strength $H < 2500$ A/m allows the estimation of the magnetoelastic anisotropy $K_{z'}$ from $\langle m_{z'b} \rangle = HB/2K_{z'}$. This relation expresses the minimum in the magnetic part of the free energy for an uniaxial magnetic system. We get an induced anisotropy $K_{z'} = 1200$ J/m$^3$. Fig. 4 shows the mean thickness $d$ of the closure domains (normalized to the total thickness $\delta$ of the ribbon) as a function of the mean magnetization $\langle m_{z'b} \rangle$ in the bulk. Below $\langle m_{z'b} \rangle = 0.6$, the thickness $d$ increases only slightly with the applied field and the closure domains occupy less than 10% of the ribbon volume. Hence the closure domains contribute only little to the depolarization of the neutron beam and $\langle m_{z'b} \rangle$ and $\gamma_{z'b}$ cannot be determined from the measured depolarization data with significant accuracy.

Above $H = 2000$ A/m, the mean thickness $d$ of the closure domains strongly increases, approaching a value of 1/2 when $\langle m_{z'b} \rangle$ becomes one. However, in this "high field region", most of the ribbon becomes magnetically saturated and therefore, the parameter $d$ loses its meaning. This is reflected in the more pronounced spread of the values $d$ for $\langle m_{z'b} \rangle > 0.7$.

![Fig. 2. Sketch of the 3-layer model; sl = surface layer, bl = bulk layer.](image)

![Fig. 3. Bulk magnetization $\langle m_{z'b} \rangle$ as a function of the applied field $H$ along $x'$.](image)
4. Discussion

The mean domain spacing along the \( y' \) and \( z' \) axis in the ribbon is strikingly different, suggesting that the stress state can be approximated by two principal stresses \( \sigma_y \) and \( \sigma_z \), respectively (\( \sigma_y, \sigma_z < 0 \) for compressive stresses). The smaller domain spacing along the \( y' \) axis indicates that the closure domains are predominantly magnetized near \( \pm y' \) in the original unmagnetized sample. Hence \( |\sigma_y| \) should be lower than \( |\sigma_z| \). Experimental evidence for such a behaviour of the domain structure in an unbalanced biaxial compressive stress state has been given by Kerr observations in an amorphous ribbon with a positive magnetostriction [8]. The mean compressive stress \( \sigma_y \) present during our field dependent measurements is derived to be \( 73 \) MPa, using the above estimated anisotropy \( K_z = 1200 \) J/m\(^3\). Here we used the relation \( K_z = \frac{1}{2} \gamma_y \sigma_y \) with a saturation magnetostriction \( \lambda_y = 11 \times 10^{-6} \) for amorphous \( \text{Fe}_{70}\text{Ni}_{30}\text{P}_{14}\text{B}_2 \) [9]. The mean induced compressive stress \( \sigma_y \) at room temperature can be estimated from the specific wall energy \( \gamma_y \) and the mean spacing \( D_0 \) determined from the angular dependent measurements as

\[
\sigma_y = 4 \gamma_y \delta / 3 \lambda_y D_0 ,
\]

knowing that the closure domains were predominately magnetized near \( \pm y' \). With \( D_0 = 5.3 \) \( \mu m \) and \( \gamma_y = 0.72 \times 10^{-3} \) J/m\(^2\) (from ref. [3] and corrected for the appropriate anisotropy \( K \) present in the \( 180^\circ \) bloch walls orientated along \( z' \)), we get \( \sigma_y = 90 \) MPa at room temperature and hence \( \sigma_z = 54 \) MPa (\( K_z = 900 \) J/m\(^3\)) during our field dependent measurements (\( T \approx 50-60^\circ C \)).

Taking into account this rough estimation of the in-plane stress distribution, we proceed to discuss the field dependence of the parameters characterizing the model of the magnetic domain structure. According to the neutron depolarization analysis, only a slight increase in the mean thickness \( d \) of the closure domains occurs at \( H \leq 2000 \) A/m (fig. 4) and the mean bulk magnetization \( \langle m_z \rangle \) increases approximately linearly with the applied field to a value of \( \approx 0.6 \) at \( H \approx 2000 \) A/m (fig. 3), which will be expected for a pure rotation process in the bulk. Thus, in the “low field region”, the magnetization reversal is predominantly determined by rotation processes in the bulk and no significant extension of the closure domains towards the centre of the ribbon is indicated from the analysis.

Assuming closure domains with their local magnetizations near \( \pm y' \) in zero field (\( y' \) domains), rotations of the magnetization in the bulk can take place without much change in the structure of the closure domains still providing flux closure (fig. 5a). The maximum variation in the mean thickness \( d \) to be expected in the “\( y' \) domains” is a factor of two, due to an extension of the favoured domains at the expense of the unfavoured ones. This behaviour of \( d \) at \( H < 2000 \) A/m is consistent with the results of our depolarization analysis (fig. 4).

On the other hand, if the closure domains are magnetized nearly parallel or antiparallel to the applied field (\( z' \) domains) while the magnetization process is predominantly determined by rotation processes in the bulk, as has been deduced from our depolarization data, movements of the oblique walls are required to keep the flux closed. From Kerr effect observations of a surface structure of “\( z' \) domains”, a linear change of the closure domain width in the “low field region” has been reported [10] and a combination of rotation processes and wall movements has been proposed by these authors. Flux closure during the magneti-
Fig. 5. (a) Sketch of magnetization rotations in the bulk with flux closure provided by "y'-domains"; (b) sketch of magnetization rotations in the bulk with flux closure provided by "z'-domains" [10].

The magnetization process in that model requires (see fig. 5b)

\[
\left(\frac{d_1}{4D_1}\right)^2 = \frac{2}{1 - m_z} - 1,
\]

and

\[
\left(\frac{d_2}{4D_2}\right)^2 = \frac{2}{1 + m_z} - 1,
\]

with the boundary condition \(D_1 + D_2 = 2D_0\), assuming a constant domain spacing \(D_0\) in the bulk. With the relation \((D_1 - D_2)/D_0 = B_1H/2K\), derived from Kerr observations in "low fields" [11], we can evaluate the dependence of the mean thickness \(d\) on \(<m_{y}\rangle\) as well as on the applied field \(H\) from

\[
d = (D_1d_1 + D_2d_2)/(D_1 + D_2).
\]

This relation is drawn in fig. 4 for a value of \(D_0 \approx 6 \mu\text{m}\) (broken line). These calculated values of \(d\) show a more pronounced increase with \(<m_{y}\rangle\) than those analyzed from the neutron depolarization. The deviation suggests that in the "low field region" \((H \leq 2000 \text{ A/m})\) closure domains of the "y'-type" are present, consistent with angular dependent measurements in a zero field. Above \(H = 2300 \text{ A/m}\), the mean thickness \(d\) of the closure domains exceeds the value \(d_0\) in the zero field by more than a factor of two and the more pronounced increase with the applied field suggests that \(d\) approaches the half thickness of the ribbon near its magnetically saturated state. Following our discussion above, an indication is given that part of the closure domains are of the "z'-type", in at least "high fields". A more quantitative description of the magnetization distribution in the closure domains during the reversal process is limited by experimental accuracy. Above, we considered two extreme types of closure domain structure, being aware that a mixing of these two cases is present in real structures.

In conclusion the main results are:
1) From neutron depolarization experiments on an amorphous ribbon within a so called Landau-Lifshitz domain structure [4], the mean magnetization in the bulk and the effective thickness of the closure domains can be determined simultaneously.
2) The analysis shows that the magnetization reversal is dominated by rotation processes in the central part (bulk) of the ribbon.
3) The thickness of the closure domains varies only slightly with the mean magnetization as expected for y' domains.

Acknowledgement

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References

STELLINGEN


2. De precisie, waarmee het kritisch gedrag van ferromagneten m.b.v. de neutronen depolarisatietechniek bepaald kan worden, kan aanleiding geven tot een nieuwe toekenning van de universaliteitsklasse aan een ferromagnetisch systeem. (dit proefschrift hoofdstuk V).

3. De toename van de kritische exponent \( \gamma_{\text{eff}} \) bij nadering van de faseovergang zoals door Kachnowski et al. wordt afgeleid uit de analyse van 'gestoorde hoekkorrelaties', is ondermeer een gevolg van het feit dat de faseovergangstemperatuur in de analyses varieert. (Kachnowski et al., Phys.Rev. B 14, 5022 (1976)).

4. Meetfouten in wetenschappelijke publikaties, welke opgegeven zijn zonder informatie te verstrekken over de wijze hoe deze fouten bepaald zijn, hebben weinig betekenis en kunnen beter weggelaten worden.

5. Het benoemen van natuurwetenschappelijke effecten met de naam van de ontdekker strekt deze weliswaar tot eer, maar is niet bevorderlijk voor de begripsvorming van het effect.

6. Het is te betreuren, dat maar weinig sporten d.m.v. handicap het spelpeil van de tegenstanders op elkaar afstemmen, waardoor het sociale aspect veel meer tot zijn recht komt. Bovendien worden daarmee ook de wedstrijdmogelijkheden tussen gehandicapten en niet-gehandicapten vergroot.

7. Het feit, dat gedurende de promotie de promovendus en opponent slechts door de Rector Magnificus onderbroken mag worden, is niet bevorderlijk voor een stimulerende discussie. (zie promotiereglement 6.5 TU Delft).

Het is zo vanzelfsprekend dat wetenschappelijke verslagen in het engels behoren te zijn, dat daar geen stelling aan gewijd wordt.

N. Stuesser

16.Oktober 1986
Abstract: A neutron depolarization study was carried out of the bulk domain structure in an amorphous Fe$_{40}$Ni$_{40}$P$_{14}$B$_6$ ribbon with a weak magnetic anisotropy perpendicular to the ribbon plane. The depth of the flux closure domains near to the surface could be determined and is compared with a Landau-Lifshitz type closure domain structure. Correlations in the direction of flux closure at the opposite surfaces of the ribbon are discussed. Studies of the domain structure in an external magnetic field show that the bulk domain walls are aligned parallel to the field direction. The last effect is attributed to the magnetostatic energy within the domain walls due to the present field.
1. Introduction

In an uniaxial ferromagnet the magnetization in the bulk is expected to be aligned parallel to the easy direction. Adjacent domains of antiparallel magnetization are separated by 180° domain walls. When the magnetic anisotropy is weak the magnetic flux must be closed in the specimen in order to avoid stray fields by magnetic poles at the surface of the ribbon. Experimental observations of the magnetic domain pattern in specimens with the easy axis perpendicular to the surface studied often show stripe like domain structures at the surface: the magnetization at the surface is oriented perpendicularly to the easy axis and adjacent domains are magnetized antiparallel [1, 2]. These observations suggest a domain structure similar to that proposed by Landau and Lifshitz (LL) [3] for cubic systems (fig. 1).

As pointed out in [4] the LL domain structure is unstable in an uniaxial system and the type of closure domain structures in the case of weak anisotropy remains open to discussion. Nevertheless the LL domain structure will be used to analyse our measurements since the surface domain structures observed [1, 2] indicate that the LL model is perhaps a good approximation of the domain structure being present.

Our neutron depolarization experiments are aimed to investigate the magnetization distribution within the volume of an uniaxial ferromagnet. In particular the mean depth of the closure domain structure will be studied. Furthermore correlations in the direction of the magnetization in the closure domains at opposite faces of the ribbon will be investigated.

The second purpose will be to study the effect of both an applied field and the distribution of stress in the ribbon plane on the orientation of the domain walls in the bulk. So far, the effect of a magnetic field on a LL type domain structure was studied by Kerr effect observations [2] and neutron depolarization [5]. Both experiments were performed on an amorphous ribbon with a stress-induced anisotropy perpendicular to the plane of the ribbon. The field was applied in the ribbon plane. In the case of the Kerr observations the magnetic field was oriented perpendicular to the stripe like domain structure observed at the surface. The orientation of the stripes remained perpendicular to the field until they vanished when the field strength exceeded the saturation value.
In the neutron depolarization experiments the field dependence of a LL type domain structure was studied by applying a periodic triangularly shaped field along the length of the ribbon while measuring the intensity of the transmitted neutrons in time channels synchronized to the periodic field. Angular dependent neutron depolarization experiments revealed that the 180° domain walls in the bulk were mainly parallel to the direction of the applied field. The orientation of the Bloch walls in the bulk was attributed to a possibly present stronger compressive stress along the length compared with the in-plane stress present along the width of the ribbon.

2. Experimental technique and specimen

Detailed information about the magnetization distribution in the specimen studied was obtained from the three-dimensional neutron depolarization analysis [6]. This technique is based on the Larmor precession of the polarization vector $\vec{P}$ of a neutron beam in a magnetic induction $\vec{B}$ described by the differential equation:

$$\frac{d\vec{P}}{dt} = \gamma (\vec{P} \times \vec{B})$$

(1)

where $\gamma$ is the gyromagnetic ratio of the neutron. In a three-dimensional depolarization set-up $\vec{P}$ can be adjusted ($\vec{P}_i$) and analyzed ($\vec{P}_j$) independently along each of the three orthogonal directions x, y, z (fig. 1). The depolarization matrix $D$ is defined by:

$$\vec{P}_j = D_{ij} \vec{P}_i$$

(2)

The measurements on an amorphous ribbon with a LL type domain structure discussed here are a continuation of former studies the difference being the present use of longer wavelength neutrons $\lambda = 0.46$ nm instead of 0.16 nm previously used. The lower neutron velocity increases the time of passage through the domains and hence this enhances the sensitivity to measure the small depolarization caused by the closure domain structure.
FIG. 1. The system of reference $(x,y,z)$ used in the three dimensional neutron polarization analysis and $(x',y',z')$ assigned to the specimen. The broken line indicates that part of the specimen exposed to the neutrons. The angle $\alpha$ denotes the glancing angle of the neutron beam with respect to the ribbon normal. The Landau Lifshitz type domain structure (LL) is sketched.

The annealed amorphous Fe$_{40}$Ni$_{40}$P$_{14}$B$_6$ ribbon investigated has dimensions $60 \times 10 \times 0.016$ mm$^3$. The uniaxial anisotropy was induced by applying a biaxial compressive stress in the ribbon plane as described in [5]. The area exposed to the neutron beam was about $8 \times 5$ mm$^2$. Beside the laboratory system of reference $(x,y,z)$ used in the experimental set-up a second coordinate system $(x', y', z')$ will be chosen that refers to the amorphous ribbon (fig. 1).
3. Experimental Results

First the depolarization elements $D_{xx}$, $D_{yx}$ and $D_{zz}$ were measured as a function of a magnetic field $H$ from $H = 0$ A/m up to the saturation field (fig. 2). The field $H$ was applied along the length of the ribbon ($z'$) which was positioned perpendicularly to the beam i.e. $\alpha = 0$ in fig. 1.

![FIG. 2. Depolarization elements $D_{xx}$, $D_{yx}$, and $D_{zz}$ as a function of the magnetic field measured with $\alpha=0$ (see fig. 1).](image)

A mean thickness of the ribbon of $d = 16.3$ \textmu m is calculated from the Larmor precession $\phi_s = \tan^{-1} (D_{yx} / D_{xx})$ measured with the sample magnetically saturated along $z$ (i.e. $D_{zz} = 1$) and using a spontaneous magnetic induction $B_s = 0.75$ Tesla for amorphous Fe$_{40}$Ni$_{40}$P$_{14}$B$_6$. The depolarization $|D| = \sqrt{D_{xx}^2 + D_{yx}^2}$ was found to be about 0.93. This deviation from $|D| = 1$ is attributed to a spread in the traversal times of the neutrons due to thickness variations known to be present in the part.
of the ribbon exposed to the neutrons. Assuming a continuous distribution of $\phi$ that is symmetric around the mean value $\phi_s$ and assuming furthermore that each value of $\phi$ occurs with equal probability the variation in thickness is estimated to range from 12 $\mu$m to 20 $\mu$m. At $H = 0$ A/m the value of $D_{xx} = 0.92$ is close to 1 which is consistent with a bulk magnetic induction $B$ perpendicular to the ribbon plane. The small depolarization observed is attributed to be dominantly caused by the closure domain structure. A value of $D_{yx}$ close to zero is to be expected with a domain structure in which the domains with $B$ parallel to $+x'$ and $-x'$, respectively each occupy about one half of that volume exposed to the neutrons. The depolarization in the $zz$-element is mainly caused by a rotation of $\delta$ around the $\pm x$ axis due to the magnetic induction in the volume of the ribbon and the value of $D_{zz}$ of about 0.7 is close to the depolarization found for $D_{xx}$ with the specimen magnetically saturated along $z$.

The change of the depolarization elements with applied field strength contains information about the magnetization process in the volume of the ribbon. The last subject was investigated in a former study [5].

In order to estimate the mean value of the anisotropy $K$ it was assumed that mainly rotations of the magnetic induction in the bulk towards the field direction occur during the magnetization process and, hence, the depolarization elements $D_{xx}$ and $D_{zz}$ can be approximated by

$$D_{xx} = 1 - (1 - \cos\phi_s)^{m_z^2}$$

and

$$D_{zz} = 1 - (1 - \cos\phi_s) (1 - m_z^2)$$

From these equations the normalized mean magnetic induction $m_z = \langle B_z \rangle / B_s$ is determined as a function of the field strength $H$. The observed linear increase of $m_z$ versus $H$ allowed to estimate the mean value of the anisotropy $K = 1.7(4) \times 10^{3} J/m^3$ using the relation $m_z = BH/2K$.

Next angular dependent neutron depolarization experiments were carried out. By measuring the angular dependence of $D_{zz}$ with the specimen rotated around $z'$ (see fig. 1) and $y'$ the mean distance $\delta$ between the $180^0$ domain walls in the bulk is determined along $y'$ and $z'$ in the ribbon plane, respectively and from this one obtains the orientation of the walls (see
appendix). Figs. 3a and 3b display the angular dependent depolarization $D_{zz}$ and $D_{xx}$ measured at zero field after the specimen was saturated magnetically along either the $z'$ or the $y'$ direction. These measurements show that the bulk domain walls are dominantly aligned along the direction of the external field that was being applied before.

![Graphs of $D_{xx}$ and $D_{zz}$ vs. angle $\alpha$](image)

**FIG. 3.** Depolarization elements $D_{xx}$ and $D_{zz}$ as a function of glancing angle $\alpha$ measured at $H = 0$ A/m. Symbols + (•) refer to data obtained with the ribbon rotated around $z'$ ($y'$) axis. The ribbon was magnetically saturated along $z'$ (see a)) and along $y'$ (see b)), respectively prior to the measurement (see text).

In order to investigate the effect of the applied field on the bulk domain structure in more detail, we have oriented the domain walls
parallel to the width of the ribbon by using a field along $y'$. Thereafter a magnetic field was applied along the length ($z'$) of the ribbon. The strength was increased in steps of $520 \text{ A/m}$ from zero to $13 \times 10^3 \text{ A/m}$. After each field step the field was reduced to zero and a polarization analysis as a function of angle was made to determine the mean distance $\delta$ of the domain walls in the $y'$-direction (fig. 4.). Under the influence of $H$ the wall distance $\delta$ decreases from $12 \ \mu\text{m}$ for $H = 0 \text{ A/m}$ to slightly above $4 \ \mu\text{m}$ for $H = 60 \times 10^2 \text{ A/m}$. These measurements indicate that the bulk domain structure changes from the initially present arrangement of $180^\circ$ walls parallel to the width to walls parallel to the length of the ribbon. Above $H = 6000 \text{ A/m}$ a slight increase in $\delta$ is observed. In addition a hysteresis in the depolarization as a function of $H$ was observed for the case that the depolarization was measured with increasing and, thereafter, decreasing field strength with $H$ along $z'$ starting with a bulk domain structure with the $180^\circ$ domain walls parallel to $y'$. No hysteresis was found in the same type of measurement if one starts at $H = 0 \text{ A/m}$ with a domain structure with the walls parallel to the applied field.

![FIG. 4. Mean distance $\delta$ of domain walls along the width of the ribbon measured at zero field after applying a field $H$ (horizontal axis) parallel to the length of the ribbon.](image-url)
Furthermore the orientation of the \(180^\circ\) bulk domain walls was measured with the ribbon under an unbalanced stress distribution, i.e. the compressive stress along the ribbon length \(z'\) differed from the compressive stress along the ribbon width \(y'\). The unbalanced stress distribution was achieved by reducing the compressive stress \(\sigma_z\), by the application of an additional tensile stress. Angular dependent measurements were performed with different domain structures with the orientation of the domain walls induced parallel to either the length or the width of the ribbon before applying the additional tensile stress. No change in the domain structure was observed for both cases until the compressive stress \(\sigma_z\), approached zero and the magnetic anisotropy perpendicular to the ribbon plane vanished. The latter was evident from the decrease of \(D_{xx}\) towards zero.

4. Data analysis and discussion

From the depolarization data at zero field the domain structure of the magnetically weak uniaxial ribbon will now be analyzed more quantitatively than was done in the foregoing chapter. The underlying domain model for the analysis will be the Landau-Lifshitz model as pointed out in the introduction.

The mean domain spacing \(d\) which is the closest distance between two adjacent \(180^\circ\) bulk domain walls can be estimated from the measured mean distance of the walls along \(y'\) (\(\delta_{y'} = 4.2(2) \, \mu m\)) and \(z'\) (\(\delta_{z'} = 12.0(5) \, \mu m\)) to be \(d = 3.9(2) \, \mu m\) (see appendix).

For a LL type domain structure in an uniaxial system the domain spacing \(d\) is determined by \(d = \sqrt{8tJ/AK}\) with \(A\) the exchange constant and \(t\) the thickness of the ribbon. Using \(t = 16.3 \, \mu m\) (29 \(\mu m\)), \(K = 1.7(4) \times 10^{-3} \, J/m^3\) (1.2 \(\times 10^3 J/m^3\)) and \(d = 3.9 \, \mu m\) (5 \(\mu m\)) one obtains \(A = 1.9(3) \times 10^{-11} \, J/m\) (1.4 \(\times 10^{-11} \, J/m\)). The first data are related to the ribbon studied here and in parenthesis correspond with a ribbon that was studied earlier [5]. A similar value of \(A = 1.5 \times 10^{-11} \, J/m\) is calculated from data of \(t,d\) and \(K\) reported from Kerr effect observations at a LL type domain structure in an unannealed ribbon [7]. All three values of \(A\) differ significantly from \(A = 2.8 \times 10^{-12} \, J/m\) which was calculated from the spin wave stiffness constant \(D = 4\mu_B^2A/B_s(T,0)\) [8].
In discussing the depolarization $D_{xx} = 0.920 (5)$ measured with perpendicular transmission $a = 0$ two different types of closure domain structures are considered with an identical domain spacing in the bulk as shown in figs. 5a and 5b. In structure a) the depth of the closure domains is only half of that in structure b). In the case that the precession angle in the closure domains is smaller than one radian which is expected to be valid for the specimen investigated, the depolarization $D_{xx}$ caused by either structure is calculated to be

$$D_{xx} = 1 - \frac{\phi_o d^2}{12} [1 - \cos \phi_o t] + \frac{\phi_o d^3}{16} \sin \phi_o t - \frac{\phi_o d^4}{32} \cos \phi_o t \quad (5a)$$

and

$$D_{xx} = 1 - \frac{\phi_o d^2}{3} \left(1 + \frac{\cos \phi_o (t-d)}{2}\right) \quad (5b)$$

for structure a) and b), respectively.

![Diagram of Landau-Lifshitz type domain structure](image)

**FIG. 5.** Landau-Lifshitz type domain structure with closure domains at opposite faces of the specimen magnetized antiparallel (a) or parallel (b). The mean depths of the closure domain structure amounts to $d/4$ in a) and $d/2$ in b). The arrows indicate the local magnetic induction.
With $d = 3.9 \mu m$ and $t = 16.3 \mu m$ the depolarization $D_{xx}$ determined from eqs. (5a) and (5b) amounts to 0.95 and 0.90, respectively while the experimental value of $D_{xx} = 0.920(5)$. At first sight the measured value for $D_{xx}$ indicates that both types of domain structure are present in the ribbon. However, some additional depolarization would be measured in both domain structures if the local magnetic induction in the bulk is not perfectly aligned along the $x'$-axis. This might probably happen because of the thickness variation present in the specimen and due to the fact that the induced easy axis of magnetization is expected to be perpendicular to the surface. The decrease of $D_{xx}$ due to a deviation of the bulk magnetic induction from the direction of propagation $x$ of the neutron beam is evident from the angular dependent depolarization experiments (fig. 3). Hence, the value $D_{xx} = 0.92$ is a lower limit for the depolarization caused by the closure domains and, therefore, at least one half of the ribbon is occupied by a domain structure with the closure domains magnetized antiparallel at opposite faces. The stripe-like domain structures observed by the magneto-optical Kerr-effect [2] favour closure domains with antiparallel magnetization at opposite faces.

From the measurements shown in the figs. 3 and 4 it was observed that the application of a magnetic field induced a preferred orientation of the $180^\circ$ bulk domain walls parallel to the external field direction. Hence, this domain structure must be of lower energy than the structure with the walls perpendicular to $\hat{H}$. Assuming equal domain spacing for both structures the free energy will be calculated in the presence of a magnetic field using the two models of the magnetization process as proposed in [2,5] (see figs. 6a and 6b). Differences between the wall energy density of the two domain structures will be neglected. The free energy will be calculated using the minimum of $E = -BH\sin\theta + K\sin^2\theta$ being $E_m = -(BH)^2/4K$ as the zero of energy. The quantity $\theta$ denotes the angle between $\hat{H}$ and the normal of the ribbon plane.

First the domain structure shown in fig. 6a will be considered. The bulk domains are assumed to have zero energy i.e. $\sin\theta = BH/2K$. The energy density in the closure domains is $\pm BH + K - E_m$ where the $\pm$ sign refers to the two types of domains magnetized either parallel or antiparallel to $H$. Hence, the energy per unit volume of the structure a) becomes (see fig. 6a)
FIG. 6. Model of a magnetization process with the 180° bulk domain walls perpendicular (a) and parallel (b) to the external field $H$. Arrows indicate the magnetic induction.

$$E_a = \frac{1}{2td} \left( [-BH + K - E_m]d_1(d+2x) + [BH + K - E_m]d_2(d-2x) \right)$$  \hspace{1cm} (6)

with $d_1 = \frac{1}{2} \frac{d + 2x}{\tan(\frac{\pi}{4} - \frac{\theta}{2})}$ and $d_2 = \frac{1}{2} \frac{d - 2x}{\tan(\frac{\pi}{4} + \frac{\theta}{2})}$.

For $\theta \ll 1$ the minimization of $E_a$ with respect to $x$ gives $x = \frac{d(BH)}{4K}$. Inserting this in eq. (6) one obtains

$$E_a = \frac{d}{2tc} K \cos^3 \theta$$  \hspace{1cm} (7)

Second the energy density of the domain structure will be calculated with the bulk domain walls parallel to the external field. In the model considered the magnetization process takes place exclusively by rotations of the magnetic induction towards $\hat{n}$ (see fig. 6b). The energy density is given by
The first and second term of eq. (8) are the contribution to $E_b$ by the bulk and the closure domains, respectively. Minimization of $E_b$ with respect to $\sin \theta$ gives $\sin \theta = \frac{\frac{B H}{2}}{t} \frac{t}{2K (t-d/2)}$. Inserting the last result into eq. (8) yields

$$E_b = \left( \frac{d^2}{2t} + \frac{d}{d-2t} \frac{B H}{2K} \right) K .$$

(9)

For small magnetization i.e. $\frac{B H}{2K} \ll 1$ the energy of structure a) is calculated to be lower than that of structure b) (see fig. 7). In contrast the experiment indicated that structure b) has a lower energy than structure a) in the "low field region".

FIG. 7. Energy differences between the domain structures sketched in fig. 6 a), b). $E_b - E_a$ (see eq. (7) and (9)) considers only the magnetostatic part $-\frac{B H}{2K}$ and the anisotropy part of the energy within the closure and bulk domains. The magnetostatic part of the energy within the domain walls $E_{w,a} - E_{w,b}$ is described by eq. (10) .
In the model calculations for structure a) and b) the magnetostatic energy $-BH$ within the domain walls was not considered and this may be relevant to understand the experimental observation. For structure a) the magnetic moments within the domain wall remain perpendicular to the external field. In contrast the moments within the walls of the type b) domain structure are subject to an in-plane field $H$. Hence, the field $H$ has either a parallel or antiparallel component in the direction of the "wall moments" depending on the screw sense of the domain wall. If both screw senses of the bulk domain walls are equally probable there will be no difference in the total magnetostatic wall energy between domain structure a) and b). Grundy et.al. [9] observed by electron microscopy that Bloch lines in thin cobalt crystals which separate right and left hand screwed domain walls move under the influence of an external field $H$ in a way such that the amount of those walls increases which are of lower magnetostatic energy. Assuming that the Bloch lines in the walls of the type b) domain structure have moved under the effect of the external field and only the most favourable screw sense is left within the bulk domain walls, then, a difference will be present in the magnetostatic wall energy between structure a) and b). Using a wall thickness $\delta_w = \pi \sqrt{A/K}$ with $A = 2 \times 10^{-12} \text{J/m}$ [8] and $K = 2 \times 10^3 \text{J/m}^3$ the difference in energy density due to the magnetostatic wall energy between structure a) and b) is estimated by

$$E_{w,a} - E_{w,b} = 0.64 \frac{\delta_B H}{d} (1 - \frac{BH}{2K})$$

(see fig.7). The factor 0.64 results from averaging the cosine between zero and $\pi/2$ due to the rotation of the magnetic moment within the wall. The term $(1 - \frac{BH}{2K})$ describes approximately the alignment of the magnetic moments towards the field direction. For $H = 2K/B$ the specimen will be magnetically saturated and $E_{w,a}$ becomes identical to $E_{w,b}$. It is obvious from fig. 7 that in the low field region the preference for the b)-type domain structure due to the magnetostatic wall energy $E_{w,a} - E_{w,b}$ exceeds the preference for structure a) caused by the energy difference $E_b - E_a$. The latter result is in agreement with the experimental observation.
As was already mentioned, Kerr-effect observations of the magnetization process on a LL type domain structure in an unannealed amorphous Fe\textsubscript{40}Ni\textsubscript{40}P\textsubscript{14}B\textsubscript{6} ribbon with the stripes of the closure domains perpendicular to the external field revealed that the orientation of the stripes remained unchanged until they vanished when the specimen became magnetically saturated [2]. This seems to contradict our results. However, Kerr-effect observations of amorphous ribbons with an unbalanced in-plane stress distribution which induces a perpendicular anisotropy indicated that the closure domains prefer to align along the direction of weakest in-plane stress [10]. Hence, there are two effects which have influence on the orientation of the domain walls, namely the presence of an unbalanced in-plane stress distribution and the magnetostatic energy within the walls due to an applied field. We suppose that a strongly unbalanced in-plane stress was present in the LL domain structure studied in an unannealed ribbon in [2]. The latter is corroborated by surface domain observations on unannealed amorphous Fe\textsubscript{40}Ni\textsubscript{40}P\textsubscript{14}B\textsubscript{6} ribbons where the surface domain structures of 'Landau-Lifshitz islands' vanished at a field H = 14400 A/m when H was applied perpendicularly to the rolling direction, and at H = 1600 A/m when H was parallel to the rolling direction [7]. This difference in stress is by more than a factor of two larger than was studied in our measurements on the effect of an unbalanced stress distribution on the orientation of the 180° bulk domain walls. As pointed out before, our measurements did not show a measurable effect that an unbalanced stress distribution within the investigated stress distribution creates a preferable orientation for the 180°-bulk domain walls.

5. Conclusion

From the analyses of the neutron depolarization in a weak magnetically uniaxial ferromagnetic ribbon valuable information is obtained about the magnetization distribution in the bulk and the flux closure at the surface. It turns out in particular that the flux closure is mutually antiparallel at opposite face of the specimen.

Our measurements indicate that the bulk domain walls align parallel to
an external field. From the above discussion we conclude that the magnetostatic energy within the domain walls is essential to describe the magnetization process in uniaxial ferromagnets.

6. Acknowledgement
We thank Prof J.J. van Loef for critically reading the manuscript.

Appendix

Determination of the mean domain wall distance $\delta$ in the bulk of uniaxial ferromagnets by angular dependent neutron depolarization.

An extensive description about this subject can be found in [11]. The short derivation presented here is limited to small angles of incidence $\alpha$ (see fig. 8), i.e. the number of walls passed by a neutron of the beam is less equal one. The neutron beam can now be divided in a part crossing one wall and a second part crossing no wall. The depolarization element $D_{zz}$ is for $\alpha \neq 0$ is calculated to be (see fig. 8)

$$D_{zz} = \frac{1}{\delta} \left[ 2 \int_{0}^{\delta/2} \cos(\phi_o \alpha \gamma) \, dy + 2 \int_{\alpha/2}^{\delta/2} \cos(\phi_o \gamma) \, dy \right]$$

(A1)

using the approximation $\sin \alpha = \tan \alpha = \alpha$ for $\alpha$ small. The quantity $\phi_o$ denotes the angle of the Larmor precession per unit length. The calculation of the integrals in (A1) yields:

$$D_{zz} = \frac{1}{\delta \phi_o} \left[ (\sin \phi_o \gamma - \phi_o \gamma \cos \phi_o \gamma) \alpha + \cos \phi_o \gamma \right]$$  

(A2)

Using eq. (A2) the mean domain wall distance $\delta_y$, in the plane of incidence can be calculated from the measured depolarization element $D_{zz}$ for $\alpha \neq 0$. The wall distance $\delta_z$, is determined from the angular dependent neutron depolarization by rotating the specimen around the y'-axis. The domain spacing $d$ which is the shortest distance between two adjacent domain walls is obtained from
if one assumes that all walls have an equal orientation in the ribbon plane (see fig. 8).

\[ d = \sqrt{\frac{\delta_y^2 \delta_z^2}{\delta_y^2 + \delta_z^2}} \]  \hspace{1cm} (A3)

FIG. 8. Neutron beam (in \( x'-y' \) plane) passing through an uniaxial ferromagnet under an glancing angle \( \alpha \). The two parts of the beam crossing one and crossing no wall are indicated by I and II. The quantities \( \delta_y \) and \( \delta_z \) denote the mean domain wall distance along \( y' \) and \( z' \), respectively, and \( d \) is the domain spacing which is defined as the shortest distance between two adjacent domain walls.

References:


CHAPTER V

STATIC CRITICAL BEHAVIOUR AT THE FERROMAGNETIC-PARAMAGNETIC PHASE TRANSITION

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Determination of the critical exponents \( \beta \) and \( \gamma \) in iron by neutron depolarization

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By measuring the total angle of the Larmor precession that a polarized neutron beam experiences during the perpendicular transmission of a magnetically saturated ring of polycrystalline iron, the magnetic induction \( B \) can be determined with a precision of \( 1-2 \times 10^{-4} \) T. Measurements have been carried out in a reduced temperature range \( T = 5 \times 10^{-4} \) to \( 5 \times 10^{-3} \) and an effective critical exponent \( \beta_{\text{eff}} = 0.363 \pm 0.004 \) has been derived. The paramagnetic region just above \( T_c \) has been studied at very low fields of about 100 A/m. The critical exponent \( \gamma = 1.33 \pm 0.02 \) is consistent with the results obtained by other techniques in much larger fields.

I. INTRODUCTION

Renormalization-group methods predict very accurately values of the universal critical exponents which relate thermodynamic quantities at phase transitions.\(^1\) To test the predicted values one has to measure the thermodynamic quantities with a high degree of precision in the close vicinity of the phase transition. Measurements carried out in scaling fields (i.e., reduced temperature, the effective magnetic field in a ferromagnet, etc.), which deviate significantly from zero should involve a "correction to scaling" in their analysis.\(^2,3\)

The main purpose of this paper is to demonstrate the possibilities of using the neutron depolarization technique to study the static critical behavior of a ferromagnet. The measurements presented here have been performed on pure iron (0.999975\% pure), whose critical behavior is believed to be described by a three-dimensional Heisenberg model. With neutron depolarization, the magnetic induction can be resolved within 1 to \( 2 \times 10^{-4} \) T, which is considered to be equal to the precision.

A neutron depolarization analysis has been carried out already on a disk-shaped single crystal of iron around \( T_c \).\(^4\) The demagnetizing field of that sample, however, prevented a quantitative analysis of the critical behavior. The measurements presented here were carried out on an an­

II. EXPERIMENTAL DETAILS

A neutron beam with a wavelength of \( \lambda = 1.6 \) Å and a degree of polarization \( P = 0.91 \) can be obtained by Bragg reflection in a magnetized Fe\(_3\)Si single crystal. The polarization \( P \) can be adjusted parallel to any one of the three orthogonal directions \( x, y, z \) (Fig. 1) by means of a polarization turner consisting of two coils wound perpendicularly with respect to each other.\(^5,6\) In the polarization turner a homogeneous field is built up with a well-defined magnitude and direction, thus causing the polarization \( P \) to rotate by Larmor precession to the desired \( x, y, \) or \( z \) direction. During transmission through the ferromagnetic sample, which is positioned inside a soft iron box to exclude external magnetic fields, the neutron beam polariza­tion changes by Larmor precession around the local mag­netization.

The depolarization, i.e., the reduction in the degree of polarization and the change in the direction of \( P \) relative to the incoming beam, is obtained from a successive polariza­tion analysis along the three orthogonal directions.

The analyzer (which consists of a second polarization turner plus a magnetized Fe\(_3\)Si single crystal) mirrors the polarizer. Hence, a three-dimensional depolarization analysis delivers a \((3 \times 3)\) matrix \( D \) defined by

\[
P_{ij} = D_{ij} P_0 ,
\]

where \( i \) and \( j \) refer to the principal axes \( x, y, \) or \( z \) of the laboratory system, \( P_0 \) is the polarization vector of the incident beam along the \( i \) axis, and \( P_{ij} \) defines the direction of analysis. The right-hand side of Eq. (1) has to be summed over \( i \).

Since the experiments were carried out on a polycrys­talline sample in the close vicinity of the phase transition at \( T_c \), the angle of rotation for \( P \) in one domain is small. The total rotation angle of \( P \) in the 2-mm-thick sample may still be appreciable. Thus a small-angle approximation is applicable\(^5-7\) which allows one to determine the mean magnetic induction \( B \) and \( B^2(1 - m^2) \) from a measured depolarization matrix \( D \). Here, \( \delta \) is a measure of the mean domain size in the propagation direction of the neutrons and \( B_0 \) and \( m = B / B_0 \) denote the spontaneous and normalized mean magnetic induction, respectively.

Actually, in our measurements with an applied field, \( B \) is proportional to the total angle of rotation \( \phi \), of the polarization vector \( P \). This angle is directly obtained from the imaginary arguments of the eigenvalues of \( D \).\(^6,7\) This angle is determined modulo \( 2\pi \). The number of multiple rotations of \( 2\pi \) is found by measuring the angle, starting with \( 0 < \phi < 2\pi \) in the paramagnetic region, and successively counting the \( 2\pi \) rotations below \( T_c \) while decreasing the temperature continuously. A rotation angle of \( 2\pi \) corresponds to about \( 1.2 \times 10^{-4} \) T in our experiments. The resolution in \( \phi \) is 1 to \( 3\)°. A more extensive description of the neutron depolarization apparatus and the derivations of the basis equations of the neutron depolarization.

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The experiments were carried out on a ring of polycrystalline pure iron (0.999975% pure). The outer ($q_o$) and inner ($q_i$) diameter of the ring were 16.0 and 10.0 mm, respectively, and the thickness $d=1.97$ mm (Fig. 1). Both surfaces of the ring were mechanically polished so that the maximum variation in the thickness of the sample was within 2 $\mu$m. Hence, the macroscopic demagnetization factor will be very small and the influence of demagnetizing fields can be neglected. The magnetic field was generated by a toroidal coil wound around the ring with 783 turns/m measured at the average circumference of the ring. The part of the sample indicated by the shaded area was transmitted by the neutrons.

The sample was mounted in a vacuum furnace ($p < 10^{-5}$ Torr) whose mantle temperature was controlled within 100 mK by water cooling. Two boron nitride (BN) disks symmetrically sandwiched the ring and they were positioned in the cylindric heater of the furnace. The heater was surrounded by three radiation shields. Both BN disks had a sickle-shaped opening which provided a diaphragm of $6.0 \times 1.5$ mm$^2$ (Fig. 1) for the neutrons. Chromel alumel thermocouples were used as temperature sensors. The thermovoltage was measured with an accuracy of 0.1 $\mu$V and a proportional-integral feedback circuit stabilized the temperature within $\pm 3$ mK. The long-term stability was found to be better than 100 mK during one day. This should be compared with the measuring time of 10 min at one temperature and with the 40 h normally needed to carry out one measuring series with successive increasing and decreasing temperature steps. We could reasonably well correct for small drifts in temperature by comparing the results from the increasing and decreasing temperature runs.

In order to check for temperature gradients in the sample holder which are introduced by heat dissipation of the current in the field coil, one thermocouple was positioned at the center of the BN disks and the other near the outer side of the field coil. A careful analysis revealed that only very small temperature gradients ($\Delta T < 25$ mK) were introduced by currents below 0.2 A ($H < 160$ A/m). Above 0.2 A, the gradient increased almost quadratically with the current. All results presented in this paper are limited to this very-low-field region. In the future we will modify the sample holder such as to minimize the gradient caused by larger currents in the field coil.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Zero-field measurements

To test the sharpness of the phase transition, a neutron depolarization analysis was performed on the unmagnetized sample ($m, H=0$). Figure 2 shows $B_2^2 \delta$ versus temperature very close to $T_c$. From 0.5 K below $T_c$ to the near vicinity of $T_c$, the convex curvature reflects the critical behavior of $B_2$, expressed by a power law with a critical exponent $\beta$. With $\beta=0.37$, a mean domain size $\delta=15$ $\mu$m was determined which appears to be constant in the temperature region $T=T_c-0.5$ K to $T_c-0.1$ K. This indicates that no significant change in the domain structure takes place on approaching $T_c$, consistent with re-
dependence in

B.

Hence it has been concluded that the

\( H = 500 \) A/m revealed no field

observed in the measure-

\( B \)

the mean magnetic induction

been obtained in nickel.

5

Due to this stability in

the domain structure, it should be possible to derive a

value of \( \beta \) in the reduced temperature range

\( t = 1 \times 10^{-4} \)

to \( 5 \times 10^{-4} \). However, to do this with a small error in

the value of \( \beta \), the temperature has to be sensed with better

precision.

Very near to \( T_c \), a small tail in \( B^2 \delta \) versus \( T \) is ob-

served, the origin of which is not quite clear; it might be

caused by a still existing small temperature gradient less

than 50 mK across the sample exposed. The presence of a

tail may also arise from depolarization due to short-range

order fluctuations around \( T_c \). Assuming that the magneti-

zation in one fluctuation cluster is comparable to the

spontaneous magnetization at zero temperature, the clus-

ter size \( \delta \) is estimated to be a few 10th nanometers in a re-

gion \( 30 \) mK around \( T_c \). These values are of the same or-

der of magnitude as found from neutron scattering.4

Similar results in the neutron depolarization have also

been obtained in nickel.5

B. Measurements in low field

These measurements were performed with successive

increasing and decreasing temperature steps. In all the

series the sample was cooled down below \( T_c \), to the start-

ning temperature without applying a field. Thereafter, the

magnetic field was turned on. The mean magnetic induc-

B(7°) and the depolarization matrix element

\( D_{yy} \) [Fig. 3(b)], where

\( D_{yy} \) is related to the mean square of the component of the

domain magnetization \( m_1 \) (normalized to \( B_0 \)) that is per-

pendicular to the applied field direction6 by

\[
\ln(D_{yy}) = -B_0^2 \delta m_1^2 .
\]

It is evident from Fig. 3(b) that \( D_{yy} \) approaches 1, i.e.,

\( m_1 \to 0 \) [Eq. (2)] with increasing field strength at the

same temperature. At a constant field strength, \( D_{yy} \) gra-

dually decreases with decreasing temperature below

\( T = 1044 \) K. Such a behavior can be expected on the basis

of an increase in both the magnetic anisotropy and in \( B_0 \).

On approaching \( T_c \), \( D_{yy} \) decreases with temperature and

shows a minimum about \( T_c \). In addition a pronounced

hysteresis is observed between the runs with increasing
and decreasing temperature steps. The hysteresis in temperature indicates the presence of different domain structures at \( T_c \), dependent upon whether the sample has been warmed up in a field from a temperature well below \( T_c \) or cooled down in the same field from the paramagnetic phase. This feature is probably related to the presence of anisotropy in the critical region and will be discussed elsewhere.\(^{10}\)

C. Critical behavior below \( T_c \)

To determine the spontaneous magnetization \( B_s \) from the measured \( B \), one has to apply a magnetic field sufficiently large to overcome the anisotropy field (see preceding section), and in addition, one has to correct for a field-induced magnetization of paramagnetic origin. This paraeffect becomes significant very near to \( T_c \) and is expected to be a factor of about 4 smaller in the ferromagnetic phase compared with the paramagnetic phase.\(^{11}\) Because the fits are based on data points with \( t \geq 5 \times 10^{-4} \), the contribution of a paraeffect to \( B \) is only relevant at the highest-temperature data considered and then at most 1% at fields \( H \leq 144 \text{ A/m} \). Therefore, this effect is neglected in the analysis. It is evident from the above discussion that the mean magnetic induction \( B \), measured at reduced temperatures \( t \) in the range \( t = 5 \times 10^{-4} \) to \( 5 \times 10^{-3} \) with \( H = 144 \text{ A/m} \), should be closely identical to \( B_s \). Therefore, \( B \) must obey the same critical behavior versus temperature near the phase transition as predicted for \( B_s \), i.e.,

\[
m = \frac{B(T)}{B_s(0)} = \frac{B_s(T)}{B_s(0)} = b t^\beta (1 + at^\gamma), \tag{3}
\]

\( \beta \) and \( b \) are the critical exponent and the critical amplitude, respectively. Equation (3) includes a correction to the scaling term defined by an universal critical exponent \( \Delta \) and a correction to the scaling amplitude \( a \). This last term is expected to be important when data cover a much wider temperature range. In the analysis of our data within the range \( t = 5 \times 10^{-4} \) to \( 5 \times 10^{-3} \), a three-parameter least-squares fit in \( b_{\text{fit}}, T_c \), and \( \beta_{\text{fit}} \) to a simple power law has been made:

\[
m = b_{\text{fit}} t^\beta_{\text{fit}}, \tag{4}
\]

which approximates Eq. (3) for small values of \( t \). Initial values of \( b_{\text{fit}}, T_c \), and \( \beta_{\text{fit}} \) have been obtained from a \( \ln(m) - \ln(t) \) plot, using that value of \( T_c \), which produced the best straight line. Those values in \( b_{\text{fit}}, T_c \), and \( \beta_{\text{fit}} \) that gave the minimum in the sum of the least squares are given in Table I. The fit to the experimental data at \( H = 144 \text{ A/m} \) is shown in Fig. 4. The excellent quality of the fit indicates that the data are well described by the power law (4) and possible deviations due to the correction to scaling are not resolved in the reduced temperature range \( t = 5 \times 10^{-4} \) to \( 5 \times 10^{-3} \). The mean value of \( \beta_{\text{fit}} \) and \( b_{\text{fit}} \) at \( H = 144 \text{ A/m} \) from both runs (decreasing \( T \) steps and increasing \( T \) steps, see Table I) leads to \( \beta_{\text{fit}} = 0.363 \pm 0.004 \) and \( b_{\text{fit}} = 1.525 \pm 0.02 \). The errors are mainly due to the small drift in temperature during the measuring series. The difference between \( b_{\text{fit}} \) and the universal \( b \) can be estimated by evaluating \( d \ln(m)/d \ln(t) \) in Eqs. (3) and (4) which results in

\[
\beta_{\text{fit}} = \beta + a \Delta t^\Delta.
\]

\( \Delta t \) is some mean reduced temperature of the temperature range covered in the fit. With \( \Delta = 0.55 \), predicted theoretically for a three-dimensional Heisenberg model,\(^{12} \) and \( a = -0.5 \) for iron,\(^{11} \) a value of \( \beta = 0.373 \pm 0.004 \) can be estimated using \( \Delta t = 2.5 \times 10^{-3} \). This \( \beta \) is close to \( 0.365 \pm 0.001 \), derived from renormalization theory for a three-dimensional Heisenberg model. The relation between \( \beta_{\text{fit}} \) and \( \beta \) can be estimated from a comparison of Eq. (3) and Eq. (4) with (5) substituted into (4):

\[
b_{\text{fit}} = b \left( \frac{1 + a \Delta t^\Delta}{t^\Delta} \right)_{av}.
\]

\( (\cdot)_{av} \) denotes an averaging in the reduced temperature range under investigation. From Eq. (6) the critical amplitude \( b = 1.57 \pm 0.02 \) is estimated. It is difficult to compare our present results with those obtained from most other techniques, where a value of \( \beta_{\text{fit}} \) is derived from fitting the spontaneous magnetization, measured in a larger temperature range, to the power law (4). Mainly, Mössbauer studies on iron have been performed at temperatures corresponding with reduced temperatures \( t \leq 5 \times 10^{-3} \).\(^{12,13} \) The hyperfine field has been resolved in those experiments within 0.5 to 1 kG, which corresponds to a precision in the magnetic induction \( B \) of \( 30-60 \times 10^{-14} \text{ T} \). The precision in \( B \) is about a factor of 20 worse than that obtained by neutron depolarization.

The lower resolution in \( B \) by Mössbauer experiments becomes evident from the relatively large error \( \Delta \beta_{\text{fit}} = 0.01 \) to 0.02 reported by these authors.

| Table I. Critical exponents of iron. \( \beta_{\text{fit}}, b_{\text{fit}} \) result from fits in a reduced temperature range \( t = 5 \times 10^{-4} \) to \( 5 \times 10^{-3} \). The numbers in parentheses are the uncertainty in the last digit. The uncertainty does not include the small drifts in the temperature (see text). |
|---|---|---|---|---|
| \( \beta_{\text{fit}} \) | \( b_{\text{fit}} \) | \( T_c \) (K) | \( \gamma \) | \( H_{\text{fit}} \) (A/m) |
| 0.360(2) | 1.51 | 1045.84 | 1.330(15) | 144* |
| 0.366(2) | 1.54 | 1045.88 | 1.330(15) | 144* |
| 1.35(3) | 48* |
| 1.34(3) | 48* |

*Measuring run with increasing \( T \) steps.

*Measuring run with decreasing \( T \) steps.
The resolution in the determination of $B$, achieved by the neutron depolarization technique, should be sufficiently high to derive the first-order correction to scaling term in iron and to determine the critical exponent $\Delta$ with significant accuracy. To do this a measurement of $B$ in a wider temperature range from about $T_c - 50$ K up to $T_c$ is needed. Such a measurement has to be performed in larger fields ($H \approx 1500$ A/m) to saturate the polycrystalline iron even at the lowest temperature in this temperature range. Preliminary results show a continuous variation in $B_{\text{eff}}$ with temperature [see Eq. (5)]. However, the temperature gradients, introduced by the field current (see experimental details), prohibit at the moment a reasonably accurate determination of the first-order correction to scaling term in the critical exponent $\Delta$. It should be noted that the correction to scaling in iron has been recently analyzed from Mössbauer experiments. From that analysis a value of the correction-to-scaling amplitude $a = -0.46$ and a universal $\beta = 0.367$ has been derived from a multiparameter fit in four variables ($\beta, \gamma, T_c$, and $g$) of the hyperfine field to Eq. (3). The fit has been performed in a reduced temperature range $t = 10^{-3}$ to $3.4 \times 10^{-1}$, assuming that the theoretically predicted value of $\Delta = 0.55$ is valid in iron. Due to the negative value of $a$, $B_{\text{eff}}$ should always be below the universal $\beta$ [see Eq. (5)]. Values of $B_{\text{eff}}$, derived by this author from the power law (4) for $t \leq 2 \times 10^{-2}$ are between 0.371 and 0.379 and hence located above $\beta = 0.367$. We can estimate from this $\beta$ an expected value of $B_{\text{eff}} = 0.34$ using the correction parameters found by this author and a mean reduced temperature $t = 10^{-2}$ for the temperature range $t = 10^{-3}$ to $2 \times 10^{-2}$, where this author fitted a value of $B_{\text{eff}} = 0.371$ (absorber experiment) or 0.379 (source experiment). From the latter calculations, an inconsistency of about 0.02–0.04 in $B_{\text{eff}}$ or $\beta$ may be concluded.

D. Critical behavior above $T_c$

The static critical behavior just above $T_c$ is described by the relation

$$m = \frac{B(T)}{B(0)} = G^+t^{-\gamma}H.$$  \hspace{1cm} (7)

$m/H$ is assumed to be identical with the initial paramagnetic susceptibility $X_p$ and $t$, $\gamma$, and $G^+$ are the reduced temperature, the critical exponent, and the critical amplitude, respectively. From a theoretical point of view, Eq. (7) is strictly valid in the limiting case $H \rightarrow 0$. A least-squares fit was performed to fit our data to Eq. (7). Initial values of $T_c$, $\gamma$, and $G^+$ were obtained by the same procedure, used in the fit of a similar power law in the ferromagnetic region. Due to the small drift in temperature, the data of the runs with increasing and decreasing temperature have been fitted separately. The values of $\gamma$, which produced the best fits to the individual measuring runs are also summarized in Table I. In Fig. 4 the best fit, using the parameters given in the first column of Table I, to the experimental data is shown for one measurement, which was carried out with increasing temperature steps at $H = 144$ A/m. The fit is excellent. The values of $\gamma$, obtained from the fits, are consistent with those obtained by other methods such as the Faraday balance [$\gamma = 1.33$ with $H \geq 1400$ A/m (Ref. 14)] or induction experiments [$\gamma = 1.33$ with $H \geq 2000$ A/m (Ref. 15)]. It will be noted that the accuracy of $\gamma$ increases when using larger fields. In addition, it should be mentioned that the values of $T_c$, fitted in one run from scaling laws above and below $T_c$, are in all cases consistent within $\Delta T = 20$ mK.

IV. SUMMARY AND CONCLUSION

The results obtained from a neutron depolarization analysis on a ring-shaped iron sample show that the neutron depolarization technique is a promising method to study the critical behavior of a ferromagnet around the phase transition. The magnetic induction $B$, derived from the Larmor precession of a polarized neutron beam in a ring of iron, magnetized to saturation, can be determined with a precision of about 1 to $2 \times 10^{-4}$ T. In the ferromagnetic region this exceeds by more than a factor of 20 the relative precision of the best-known method used up to now, which is the determination of the hyperfine field by means of the Mössbauer effect. 

Fits of the magnetic induction $B$ versus $T$ in a reduced temperature range ($t = 5 \times 10^{-4}$ to $5 \times 10^{-2}$) to a simple power law yield an effective critical exponent $B_{\text{eff}} = 0.363 \pm 0.004$. Taking into account a correction to scaling, this results in an universal $\beta = 0.373 \pm 0.004$, a value which is slightly above the predicted $\beta = 0.365$ for the three-dimensional Heisenberg system. The critical exponent $\gamma = 1.33 \pm 0.02$, derived from a fit of $B$ versus $T$ in the paramagnetic phase with applied fields $H$ below 150 A/m, agrees well with those values obtained from
other techniques, 14, 15 where much larger fields have been used.

The high resolution in \( B \) obtained by the neutron depolarization technique in combination with an elaborate temperature sensing should allow us to analyze data, measured in an extended reduced temperature range \( t \leq 10^{-1} \), to determine the "correction to scaling" and to determine in an experimental way values of both the critical exponents \( \beta \) and \( \Delta \).

The neutron depolarization in zero field indicates that the domain structure is stable within 0.2 K just below \( T_c \), a fact that can, in principle, be used to derive \( \beta_{\text{eff}} \) within \( t \leq 5 \times 10^{-4} \) from the analyzed \( B \).

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Critical exponents, amplitudes, and correction to scaling in nickel measured by neutron depolarization

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The magnetic induction \( B \) was measured with a precision better than \( 2 \times 10^{-4} \) T around the phase transition in a temperature region from \( T_c - 20 \) K to \( T_c + 15 \) K in fields below 1550 A/m. An asymptotic value equal to 0.390(3) was found for the critical exponent \( \beta \). An effective critical exponent \( \gamma_{\text{eff}} = 1.315(15) \) is given in the range \( 5 \times 10^{-4} \leq t \leq 2 \times 10^{-1} \). Four critical amplitudes and one correction to the scaling amplitude are reported.

I. INTRODUCTION

The ferromagnetic-paramagnetic phase transition in nickel has been studied by several authors since the first measurement of the magnetic induction \( B \) around the critical temperature \( T_c \) by Weiss and Forrer in 1926.\(^{1-11}\) Critical exponents derived on the one hand by fitting experimental power laws were assumed to be valid near \( T_c \) along suitable thermodynamic paths. On the other hand, the analysis of \((B, H, T)\) data \((H\) is the magnetic field, \( T \) is the temperature) to "scaled equations of state" have been performed to yield the critical exponents. The reported values for the exponents \( \beta \) and \( \gamma \), defined by

\[
B_c(T) = t^\beta, \quad \chi = t^{-\gamma}
\]

(1)

\( B_c \) is the spontaneous magnetic induction, \( \chi \) the susceptibility, and \( t = | T - T_c | / T_c \) (the reduced temperature), are in most cases located around 0.37–0.39 and 1.28–1.35, respectively. These values deviate slightly from \( \beta = 0.365 \) and \( \gamma = 1.387 \) calculated for a three-dimensional (3D) Heisenberg system.\(^{12}\)

Since the experimental data are outside the asymptotic critical region, correction-to-scaling terms should be considered in the analysis.\(^{13,14}\) The existence of correction terms has been seen experimentally in the phase transitions of fluids.\(^{15}\) The observation of these small correction terms in magnetic systems is hampered by the fact that most experimental methods for measuring \( B \) are not precise enough. Only Mössbauer experiments in iron gave some information about the correction term.\(^{16}\) However, the reported universal \( \beta \) and \( \beta_{\text{eff}} \)'s show some inconsistencies.\(^{17}\)

Recently, it was demonstrated that the neutron depolarization technique, applied to ring-shaped samples with vanishingly small demagnetizing factors, is a very sensitive method for measuring \( B \) in \( \beta \)-romagnets.\(^{17}\) In former neutron depolarization experiments on nickel, a value for the exponent \( \beta \) could be derived from the measured depolarization \( B_c^2 \) \( B_c \) is the spontaneous magnetic induction) within 1 degree close to \( T_c \), where the mean domain size \( 6 \) was shown to be constant in that temperature range.\(^{18,19}\) The analysis of the field-dependent measurements was hampered by the depolarization caused by demagnetizing fields.\(^{20}\) The high resolution in \( B \) below \( 2 \times 10^{-4} \) T, achieved in the experiments presented here, permits the measurement of \( B \) in the close vicinity of \( T_c \) independent of the external field \( H \). The fields applied are on the order of a few hundred A/m.

In this paper we aim to determine the asymptotic value of the exponent \( \beta \) and to give a close estimate of the correction terms to scaling in the asymptotic power law which describes the temperature dependence of the spontaneous magnetic induction \( B_c \) just below \( T_c \).

Secondly, we aim to check the increase of the exponent \( \gamma_{\text{eff}} \) while approaching \( T_c \), observed from analyzed data of a "perturbed angular correlation" experiment within \( t \leq 4 \times 10^{-1} \). The latter experiment gives, so far, the only data available in this small reduced temperature range just above \( T_c \).

Thirdly, the lack of experimentally determined values for critical amplitudes, which are needed to test predicted universal amplitude ratios, has motivated us to derive amplitudes from our data without using any equation-of-state analysis.

II. EXPERIMENT

The neutron depolarization experiments were performed on a pure \((0.99997)\) polycrystalline nickel ring (outer diameter of 16 mm, inner diameter of 10 mm, \( d = 3 \) mm), which had been annealed for 60 h at 1050 K to reduce present internal stresses. The magnetic induction \( B \) was derived from the total angle of Larmor precession \( \varphi \), that a polarized neutron beam experiences during traversal of the sample. Figure 1(a) shows the geometry of the neutron depolarization set up used.\(^{18}\) The magnetic induction \( B \) is given by \( B = \varphi / cd \) with \( c = \gamma / v \), where \( \gamma \) is the gyromagnetic ratio of the neutron, \( v \) is the velocity of neutrons. The angle \( \varphi \) is determined from the directly measured \( x \) and \( z \) components \( D_x \) and \( D_z \), respectively, of an incident beam polarized parallel to the \( x \) axis by

\[
\varphi = \tan^{-1} \left( \frac{D_x}{D_z} \right) + 2\pi n .
\]
FIG. 1. (a) Principal geometry and system of reference in the neutron depolarization setup. The shaded area indicates that part of the ring traversed by the neutrons. P is the polarization vector of the neutron beam. (b) Example of a measured depolarization element $D_a$ versus temperature. The physical meaning of $D_a$ is visible in Fig. 1(a).

The temperature of the sample was measured within 0.01 K. The long-term drifts in temperature were below 0.025 K/24 h. More details are reported elsewhere. The maximal field strength that could be applied was about 1550 A/m. Stray fields at the sample position were below 10 A/m. Measuring series at a given field strength were performed by continuously decreasing and successively increasing the temperature with a constant change of $T$ versus time. Each data point $B(T)$ equals an averaged value over a temperature interval $\Delta T$, determined by the chosen temperature change with time and the required resolution in $B$. The latter is determined by the counting statistics.

The quantity $\Delta T$ amounts from 0.01 to 0.05 K in our measuring series. Typical cooling (heating) rates in a series were about a few 0.01 K/min. All data points of the decreasing and increasing temperature run were consistent within 0.02 K.

III. EXPERIMENTAL RESULTS AND DATA ANALYSIS

A. Critical temperature

The magnetic induction $B$ was measured at different field strengths between $H = 1$ and 1550 A/m in a temperature region from $T_c - 20$ K to $T_c + 15$ K. In the measurements with $H \leq 20$ A/m the point of inflection in $B(T)$ could be determined within 0.01 K and it was assumed to correspond to the value of $T_c$. Figure 2 shows a measurement at $H = 8$ A/m. Adjacent data points in that measurement differ by about 0.01 K in temperature. In larger fields the phase transition smears out and the "definition" of $T_c$ becomes more difficult. One way to obtain $T_c$ is to fit the data both below and above the phase transition to asymptotic power laws (1) with $T_c$ as an adjustable parameter. The comparison of the $T_c$'s obtained from such fits gives some indication of the precision in the value of $T_c$. This standard method was applied by us to neutron depolarization data for iron. However, as is well known, slightly different values of $T_c$ give fits of nearly the same quality with somewhat different values of the critical exponent and amplitude. To avoid this problem measurements of $B(H)$ along isotherms were performed at a few temperatures around $T_c$ (Fig. 3). In these experiments a magnetic field of triangular shape in time with a frequency of one cycle per sec was applied periodically to the sample and the neutron intensity, which determines $D_{ax}$ and $D_{ax}$, was counted in time channels synchronized to the applied field. This sort of measurement will be referred to as "quasistatic". It will be noted that each data point $B(T)$ represents an average value over a temperature interval $\Delta T$, determined by the required resolution in $B$.

The singularity of the initial susceptibility above $T_c$ is characterized by the exponent $\gamma$: $\chi = G^{-\gamma} t^{\gamma n}$, (2)
where $G^+$ is a critical amplitude. The susceptibility is defined by $\chi = \partial m / \partial h$ with $m$ identical to the magnetic induction normalized to the spontaneous magnetic induction $B_s$ at zero temperature [$B_s(T=0)=0.65$ T], and $h$ identical to the magnetic field $H$ normalized to $k_B T_c / n_B = 1.21 \times 10^5$ A/m for nickel ($n_B$ is the magnetic moment per atom).

First, isotherms $B(H)$ were measured at different temperatures close to $T_c$ (Fig. 3). From these measurements a lower bound in the reduced temperature of about $5 \times 10^{-4}$ was found, above which $m$ is linearly dependent on $h$ for $H < 1550$ A/m and $m/h$ is a good measure of the initial susceptibility. Least-squares fits to Eq. (2) were performed on the data in the reduced temperature range $5 \times 10^{-4}$ to $2 \times 10^{-3}$ at a few field strengths $H$ between 390 and 1550 A/m. The upper bound $t = 2 \times 10^{-3}$ in temperature and the minimum field strength of 390 A/m were chosen to keep the relative error in $m$ below a few percent.

All fits within the above $T$ region yielded values for $\gamma_{\text{eff}}$ between 1.30 and 1.34 with a mean value about 1.315. The quantity $G_B$ amounts to 1.40(3).

C. Ferromagnetic phase

Including the first-order correction to the scaling, the temperature dependence of the ordering parameter $m$ near the phase transition is given by

$$m = b \beta^{\Delta_c(T)} + a \Delta_c^{\Delta_c - 1},$$

with $b, a$ and $\Delta_c$ as two critical amplitudes and two critical exponents, respectively. Effective critical exponents obtained by fits of data $B(T)$ in a finite $T$ region near $T_c$ to a simple power law

$$m = b \beta^{\beta_{\text{eff}}},$$

are related to the asymptotic value $\beta$ by

$$\beta_{\text{eff}} = \beta + a \Delta_c^{\beta_{\text{eff}}} + b \Delta_c^{\Delta_c - 1},$$

with $t$ a mean reduced temperature of the covered $T$ region.

To derive the spontaneous magnetic induction $B_s(T)$ from the measured $B(T, H)$ one has to consider two corrections. First, a field induced induction $B_f$ of paramagnetic origin has to be taken into account. Secondly, the presence of magnetic anisotropy causes the measured $B$ to be below $B_s$, with $B$ approaching $B_s$ only in fields much larger than these anisotropy fields.

From the constant slope of $B$ versus the external field plot at $H \geq 150$ A/m, with the data along the $(T_c - 0.44)$ K isotherm (Fig. 3), we estimate a critical amplitude $G_{\text{eff}} = 0.41(4)$. The critical exponent $\gamma$ below $T_c$ was assumed to be identical with the one found above $T_c$. The measured $m$ was corrected for this paraeffect by subtracting $G_{\text{eff}}^{\gamma(\gamma-1)}$. Despite this correction, the data used to fit Eq. (4) were limited to those obtained below $T_c - 0.7$ K ($t = 1.1 \times 10^{-5}$) in order to restrict the correction in $B$ below 2% at the maximum temperature in the highest external field used. Moreover $B(H)$ looses its linearity with $H$ upon approaching $T_c$ so that the applied correction becomes invalid.

The strength of anisotropy fields present was derived from a study of the law of "approach to saturation" and will be discussed in detail elsewhere. In this way we could extrapolate the measured $B$ at the different field strength to obtain a value of $B_s$ with an estimated error below 2% at the lowest temperature. It is evident from Fig. 2 that $B(T, H = 390$ A/m) is already close to saturation ($\Delta B/B_s < 2\%$) above $T_c - 12$ K. Within 1 K below $T_c$, the anisotropy fields are much smaller than 150 A/m, and this justifies attributing the change in $B$ with $H$ at $H > 150$ A/m to paraeffects. On the other hand, paraeffects are negligible below $T_c - 2$ K at fields below 550 A/m, and $B(H)$ is dominated by anisotropy fields.

The data from different measuring series with field strengths between 390 and 1550 A/m were fitted to Eq. (5). The least-squares fit of $\beta_{\text{eff}}(T)$ to Eq. (5) yields an asymptotic $\beta$ of 0.390(3) and a correction to the scaling amplitude $a = -0.42(4)$. The errors in $\beta$ and in the quantity result from the uncertainty in $T_c$, possible errors in the applied corrections to $B$, and the standard deviation obtained in the fit. The value $\Delta$ was kept at 0.55 calculated for the three-dimensional Heisenberg system. The change of $\Delta$ with the dimension of the order parameter is small for three-dimensional systems and the effect on the value of $\beta$ is within the quoted error.

The asymptotic value of $\beta$ was derived from

$$b = b_{\text{eff}} \left( \frac{\gamma_{\text{eff}}}{1 + a \gamma_{\text{eff}}} \right),$$

to be $1.52(2)$ [see Fig. 4(b)]. $\langle \gamma \rangle_{\text{av}}$ denotes an averaging in the reduced temperature range under investigation.
FIG. 4. (a) Effective critical exponent $\beta_{\text{eff}}$ versus reduced temperature $\tilde{T}$ (see text). The errors in the data account for the uncertainty in $T_c$ and possible drifts in temperature. [The same is valid in Fig. 4(b)]. Dashed lines denote the result of least-squares fit to Eq. (5). (b) Effective amplitude $b_{\text{eff}}$ versus $\tilde{T}$. The symbols have the same meaning as in Fig. 4(a). Dashed lines denote the result of least-squares fit to Eq. (6) with fixed parameters $\Delta = 0.55$, $a = -0.415$, and only one fittable parameter $b$.

D. Data near the critical isotherm

On the critical isotherm, $h(m)$ is given by

$$h = Dm |m|^{1-\delta},$$

with $D$ and $\delta$ as a critical amplitude and critical exponent, respectively. The critical exponent can be calculated from the found $\beta$ and $\gamma$ to be $4.38(6)$, using the scaling relation $\delta = 1 + \gamma / \beta$. Data of $m$ and $h$ with $t < 1.5 \times 10^{-3}$ were obtained from the temperature series. From these data a value of $D = 0.32(10)$ was estimated for $H \geq 590$ A/m. The error is caused by the uncertainty in both $\delta$ as well as in $T_c$. To get reliable data of $h(m)$ on the critical isotherm by a "quasistatic" measurement was impossible because of small temperature drifts during the measuring time of a few hours needed to perform such a measurement.

The data above $T_c + 0.06$ K, including those outside the "linear" $h(m)$ region, were transformed to the reduced quantities $\bar{m} = m / t^\beta$ and $\bar{h} = h / t^{\delta+\gamma}$. The nearly linear relation between $\bar{m}^2$ and $\bar{h}/\bar{m}$ (Fig. 5) shows that the scaling function is well approximated by $\bar{h} = f(\bar{m}) = c_1 \bar{m} + c_2 \bar{m}^3$, with $c_1 = 0.72(3)$ and $c_2 = 0.92(10)$. It will be noted that in comparison with data obtained at fields about 1000 times larger, the value of $c_1$ agrees quite well while the value of $c_2$ found here is about a factor of 2 larger. Data were used from measuring series at $H$ above 236 A/m. Data obtained at smaller fields were excluded because $m$ can be measured with a small relative error only very close to $T_c$ at these small field strengths and the reduced quantities $\bar{m}$ and $\bar{h}$ become very sensitive to the choice of $T_c$ in that case.

FIG. 5. Plot of $m^2$ against $h/\bar{m}$ for data obtained in measuring series at three different field strengths. $\bar{m} = m / t^\beta$ and $\bar{h} = h / t^{\delta+\gamma}$ are the reduced order parameter and the reduced field. The error bars of data points indicate the effect of a shift of 5 mK in the value of $T_c$.

IV. SUMMARY AND DISCUSSION

All critical quantities obtained in this study are summarized in Table I and compared with theoretical calculations for the 3D Heisenberg system, which is assumed to be the universality class that describes best the critical behavior in nickel.

The value of 0.390(3) found for $\beta$ in nickel is well above that calculated for the 3D Heisenberg system, 0.365(1). The effective exponents $\beta_{\text{eff}}$'s found in our study are in agreement with most values obtained from the analysis of other data in corresponding temperature ranges. Our analysis shows that correction terms to the scaling are present. These correction terms cause the experimentally found $\beta_{\text{eff}}$'s to be below the asymptotic value $\beta$ in the case of nickel.

Due to the small absolute values of $B$ above $T_c$ for fields $H \leq 1550$ A/m, the exponent $\gamma$ could only be derived with a relatively large uncertainty. The possibility of obtaining data in very small external fields makes an extrapolation of $m(h)$ data to zero field unnecessary for yielding a measure of the initial susceptibility. The value for $\gamma_{\text{eff}} = 1.315(15)$ is based upon data within a reduced temperature range $t \leq 2 \times 10^{-3}$, which was accessible up to now only by a perturbed angular correlation experiment in the small-field region. The data of the latter experiment indicated an increase of $\gamma_{\text{eff}}$ upon approaching $T_c$ at $t \leq 4 \times 10^{-3}$ to perhaps a value of 1.387(1) for the 3D Heisenberg system. Our data do not corroborate this effect, and the value of 1.315 is within the limits of error, in excellent agreement with the analyses of data in a wider
temperature region and at much larger fields \((\gamma) = 1.29 - 1.35\), 1,2,4,10,11

Using zero-order results for universal ratios of correction amplitudes, 14 a correction amplitude of about \(-0.5\) can be estimated which results in a difference of 0.006 between \(\gamma_{expt}\) at \(T = 1 \times 10^{-3}\) and the asymptotic value and, hence, cannot be resolved within the low-field region.

The four derived critical amplitudes \(b, G^+, G^-,\) and \(D\) allow one to calculate two universal ratios \(G^+/G^-\) and \(R_x = G^+ \delta^{1-D}\). The values quoted in Table I agree well with those obtained by the application of a modified Misiori, Levelt Sengers, and Green (MLSG) equation of state to the Weiss-Forrer data. 20,24

The calculated values of \(R_x = 1.23\) (high-temperature series) and 1.33 \((\epsilon\) expansion) do not contradict the experimental value of 1.8(5). The amplitude ratio \(G^+ / G^-\) is theoretically predicted to be zero for systems with \(n \geq 2\) (\(n\) is the dimension of the order parameter), but the ratio found experimentally is significantly different from zero; this may be due to the effect of dipolar interactions. 25

\section*{V. CONCLUSIONS}

The \((B,H,T)\) data presented have a higher precision than those reported by other methods so far and moreover, they are located closer to the asymptotic region. The accessibility of data in the region of low fields allows one to get reliable data of the initial susceptibility without applying nonlinear extrapolations to zero field.

Our experimental data give clear evidence for the existence of a correction term to the simple power law \(m = bt^\alpha\) close to \(T_c\) in nickel. An increase of \(\gamma_{expt}\) to the 3D Heisenberg value with \(T \rightarrow T_c\) is not observed.

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\begin{table}[h]
\centering
\caption{Critical exponents, amplitudes, and correction amplitudes in nickel.}
\begin{tabular}{|c|c|c|c|}
\hline
 & This work & Theory & Results of scaled equation of state analysis on nickel taken from Ref. 21 \\
\hline
 & & & Linear mode & Modified MLSG \\
\hline
\(\beta\) & 0.39(0) & 0.365(1)* & 0.38 & 0.38 \\
\(\gamma\) & 1.31(15) & 1.387(1)* & 1.35 & 1.33 \\
\(b\) & 1.52(2) & & 1.5 & 1.4 \\
\(a\) & \(-0.42(4)\) & & & \\
\(G^+\) & 1.40(3) & & 1.5 & 1.3 \\
\(G^-\) & 0.41(4) & & 0.38 & 1.1 \\
\(D\) & 0.32(10) & & 0.29 & 0.29 \\
\(G^+/G^-\) & 1.85(3) & 1.231,33b & 1.7 & 1.4 \\
\(G^+ \delta^{1-D}\) & 3.4(4) & & 0.365(1)' & \\
\hline
\end{tabular}
\end{table}

SMALL MAGNETIC ANISOTROPY MEASURED BY NEUTRON DEPOLARIZATION IN NICKEL NEAR $T_c$

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The approach to magnetic saturation was studied by neutron depolarization in a ring shaped annealed polycrystalline sample of pure nickel from $T_c - 12$ K to $T_c - 2$ K. The magnetic induction was resolved within $1 \times 10^{-4}$ T and the field dependence of $B$ could be fitted to $B = B_0 (1 - a/H - b/H^2)$. The derived magnetic anisotropy $|K| = 3.6/5 \times B_0$ decreases nearly linearly with temperature from $18 \text{ J/m}^3$ at $T_c - 12$ K to $2 \text{ J/m}^3$ at $T_c - 2$ K.

1. Introduction

Only few experimental data are known on the crystalline anisotropy of simple ferromagnets such as iron and nickel near to the Curie temperature $T_c$. The sensitivity of most used methods is insufficient to measure anisotropy energies of the order of a few $\text{J/m}^3$.

Here we are concerned with the possibilities that the neutron depolarization technique offers to measure these very low anisotropies in ferromagnets. The experiments were carried out on an annealed pure nickel sample in a temperature region around $T_c$.

Recently it has been shown in a study of the critical behaviour in iron that the magnetic induction $B$ could be resolved within $1 \times 2 \times 10^{-4}$ T [1]. This high resolution in $B$ allows us to study the approach of the mean magnetic induction to its saturation value very precisely and to derive the size of the present anisotropy.

2. Experiment

In a three-dimensional neutron depolarization experiment the polarization vector $P$ of the monochromatic neutron beam can be adjusted and, after depolarization by transmitting a ferromagnetic sample, be analyzed in three orthogonal directions ($x, y, z$) independently [2] (fig. 1). The experiments were performed with an incoming neutron beam of wavelength $\lambda = 0.16 \text{ nm}$ and a degree of polarization $P$ of about 0.92.

A ring (OD = 16 mm, ID = 10 mm, with a thickness of 3 mm) of pure polycrystalline nickel (99.997%) was used as the sample. No significant grain orientation was observed at the etched surface of the sample. The ring was carefully polished and the variations in the thickness over the whole circumference of the ring were within 2 $\mu$m. Hence demagnetizing fields could be ne-
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glected. Prior to the measurements the sample was vacuum-annealed at a temperature of about 1050 K for 60 h to reduce present internal stresses. The sample was mounted in a vacuum furnace the temperature of which was controlled within $\pm 5$ mK [1].

The temperature was measured by chromel-alumel thermocouples with an absolute accuracy within 2 K. The drift in temperature over one day was below 25 mK. The magnetic field $H$ was generated by a current through a toroidal coil. A diaphragm confined the neutron beam to a small sickle shaped part ($6 \times 1.5$ mm$^2$) of the ring. Within the diaphragm the direction of the field $H$ was nearly parallel to the $y$-axis of the laboratory system (fig. 1).

3. Measurements and discussion

The mean magnetic induction $B$ was determined from the total angle $\phi$, of Larmor precession that the polarization vector $P$ of the neutron beam experienced during the transmission of the sample. The incoming beam was polarized parallel to the $x$-axis, thus perpendicular to $B$ (|| $y$-axis) (fig. 1). Analyzing the $x$ ($D_x$) and $z$ ($D_z$) component of $P$ after the transmission of the sample the angle $\phi$ was determined by

$$\phi = \arctan \frac{D_x}{D_z} + n 2\pi.$$ (1)

The number of rotations $n$ at a particular temperature $T$ could be determined experimentally by measuring $\phi$, while varying the temperature continuously from above $T_c$ ($\phi = 0$, $n = 0$) down to $T$. The mean magnetic induction $B$ was obtained from

$$B = \frac{1}{cd^2} \phi,$$ (2)

with $d$ the thickness of the sample, $c = \gamma/\nu$, $\gamma$ the gyromagnetic ratio, $\nu$ the velocity of the neutrons. In our neutron set-up $c = 7.3 \times 10^4$ T$^{-1}$ m$^{-1}$. In the experiments the angle $\phi$, was resolved within 0.02 rad which value is primarily determined by counting statistics. Thus, changes in $B$ were resolved better than $1 \times 10^{-4}$ T.

In fig. 2 the experimental data of the mean magnetic induction $B$ as a function of the field $H$ are shown at different temperatures. These data were obtained from quasistatic measurements, i.e. the field $H$ was periodic in time of triangular shape with 1 cycle/s. The field strength between two adjacent data points in fig. 2 differs 17 A/m.

The approach to saturation of $B$ was analyzed in the temperature range from $T_c - 12$ K to $T_c - 2$ K with $H \leq 550$ A/m by fitting the experimental data to [3]

$$B = B_s \left(1 - \frac{a}{H} - \frac{b}{H^2}\right).$$ (3)

with the three parameters $B_s$, $a$ and $b$. The temperature range, where fits of the data to eq. (3) were made, was limited at $T_c - 12$ K by the maximum field strength of 550 A/m that could be generated and at $T - 2$ K by the onset of measurable paraeffects above $2 \times 10^{-4}$ T at the maximum field strength. Hence a term $\chi H$ ($\chi$: susceptibility) can be neglected in eq. (3) for $T \leq T_c - 2$ K and $H$ below 550 A/m. Eq. (3) should be valid only at
The numbers in parenthesis give the uncertainty in the last digit.
fitted excellently to eq. (3) within the temperature range indicated. An example of the best least squares fit with the corresponding experimental data is given in fig. 4. All experimental data of $B$ agreed within $1 \times 10^{-4}$ T with the $B$ generated by the best least squares fit. The validity of eq. (3) to the experimental data was typically confined to values $B \geq 0.95B_c$.

For polycrystalline samples of cubic anisotropy with a random distribution of the crystalline orientations $h$ is related to the crystalline and stress induced anisotropies $h$ is related to the crystalline and stress induced anisotropies $h$.

$$h = \left( \frac{1}{18} K_r^2 + \lambda^2 \sigma_r^2 \right) / B^2.$$  

(4)

as long as $H \ll B_c / \mu_0$.

The quantities $\lambda$ and $\sigma_r^2$ denote the saturation magnetostriction and the mean square of the internal stresses, respectively.

The effective magnetic anisotropy defined as $|K| = \frac{1}{18} h B^2$, is shown versus $T$ in fig. 5. The nearly linear decrease of $|K|$ versus $T$ parallels a similar decrease of $\lambda$, versus $T$ reported for nickel above 500 K [5]. From our measurements it is not possible to separate the crystalline anisotropy from the magnetoelastic one. It is well known from experiments at lower temperatures that the crystal-

ling anisotropy decreases much faster with increasing $T$ than the magnetoelastic anisotropy.

Nevertheless the derived values of $|K|$ give upper limits of the present crystalline anisotropy near to $T_c$. A linear extrapolation of $|K|$ to a temperature $T_f > 30$ K yields $|K| = 45 \text{ J/m}^2$. A comparison of this value with $K_t \approx -45 \text{ J/m}^2$ obtained from torque measurements on a single crystal at 600 K [7] will be only of a speculative nature since the accuracy in the latter determination is very low at that temperature.

The neutron depolarization analyzed in the full temperature region from $T_c$ to $T_f - 200$ K increased monotonously and no local minima were observed. The latter would be expected if the measured anisotropy is dominated by crystalline anisotropy and if the reported changes in sign of $K_1$ are correct [7,8]. However the presence of these minima in $|K_1|$ may be hidden (covered) by a stress induced anisotropy.

An upper limit of present internal stresses about $\sigma_r^2 = 1.6 \times 10^{13} \text{ Pa}^2$ can be deduced from eq. (4) and the measured data.

4. Conclusion

A study of the approach to magnetic saturation by using the neutron depolarization technique on ring shaped samples allows one to obtain quantitative information of low magnetic anisotropies below 20 J/m$^2$. Hence this method is suitable to perform anisotropy studies close to $T_c$ where the magnetic anisotropies become small. Due to both the high resolution in $B$, achieved by the neutron depolarization technique, as well as the possibility to apply arbitrary small magnetic field, there exists, in principle, no lower limit in the anisotropy that can be measured.

Our measurements in nickel show that the approach to saturation can be well described by eq. (3) in the limited temperature range $T_c - 12$ K to $T_c - 2$ K.

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References

CHAPTER VII

MAGNETIZATION REVERSAL IN CONDUCTING FERROMAGNETS
STUDIED BY NEUTRON DEPOLARIZATION IN NICKEL

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Abstract:

The magnetization reversal in a picture-frame single crystal of nickel with the legs parallel to the [111]-crystallographic axes was studied by the time dependent neutron depolarization technique. The observed depolarization is consistent with a magnetic reversal starting at the surface and forming a $180^\circ$ domain wall boundary which moves towards the center of the specimen. From the data analysis the wall velocity was found to be the same along the entire contour of the domain boundary at a time. The wall velocity measured during the magnetization reversal is compared with a calculation based on eddy current limited domain wall motion in a ring shaped specimen.
1. Introduction

The magnetization reversal process in a conducting ferromagnet was studied experimentally and theoretically by various authors in the last decades. Most experimental techniques measure the integral change in the magnetic induction or observe the domain structure at the surface during the reversal process. It was demonstrated that the time dependent neutron depolarization analysis is a unique technique to study both the spatial and temporal evolution of the magnetization reversal inside the volume of a ferromagnet. The last experiments were performed on a FeSi(3.5 wt % Si) picture-frame single crystal with a leg cross section of 2.9 x 0.26 mm$^2$. The studies were confined to a small part of one leg with dimensions 5 x 0.2 mm$^2$. The measurements at high fields corroborated widely accepted models of an eddy current controlled reversal process starting with the nucleation of magnetically reversed domains at the surface which merge and form a 180° domain wall that moves towards the center of the sample.

In this paper the reversal process is studied by neutron depolarization in a picture-frame single crystal of nickel the legs of which had a width of 3.0 mm and a thickness of 1.75 mm. The leg cross section of this crystal is much more quadratic in shape than the FeSi crystal. Hence, the approximation made in the last case to describe the reversal process by the movement of two plane walls is not applicable to a crystal with a nearly square cross section. From induction measurements on an FeSi picture-frame single crystal with a nearly square cross section Williams et al suggest that during the reversal process the boundary between the reversed and unreversed part in the leg curves and finally forms a cylinder collapsing near to the center of the leg. The main purpose of the present study is to measure the spatial and temporal evolution of the whole boundary in one leg during the magnetization reversal process.
2. Experimental

A. Neutron depolari zation technique

In a three dimensional neutron depolari zation experiment the polarization vector $\hat{P}$ of a monochromatic neutron beam can be adjusted and after depolari zation by transmitting a ferromagnetic sample the polarization is analyzed in three orthogonal directions ($x, y, z$) (fig. 1). The depolari zation is caused by the Larmor precession of $\hat{P}$ in a magnetic induction $\hat{B}$ which is described by

$$\frac{d\hat{P}}{dt} = \gamma (\hat{P} \times \hat{B})$$

(1)

with $\gamma$ the gyromagnetic ratio of the neutron. The experiments were performed with an incident beam of wavelength $\lambda = 0.16$ nm and a degree of polarization of about 0.92. In our case the precession angle of $\hat{P}$ is related to the magnetic induction $\hat{B}$ by $\phi = 7.3 \times 10^4 B[\text{Tesla}]d[\text{m}]$ with $d$ the distance along the direction of propagation of the neutrons where the induction $B$ is present.

B. Sample preparation

The picture-frame single crystal was cut by spark erosion out of a Crochalski grown cylindrical single crystal of nickel with purity 0.99995 and a mosaic spread below 20'. The dimensions and orientation of the specimen are given in fig. 1. The variation in thickness of the sample was less than 10 $\mu$m and the depth of the craters introduced by the erosion process at the surface was about 3 $\mu$m. Next to the spark cutting a layer of 6 $\mu$m thickness was removed from the sample surface by chemical polishing for about 15 seconds in a bath of $30\%$ $\text{HNO}_3 + 10\% \text{H}_2\text{SO}_4 + 10\% \text{H}_3\text{PO}_4 + 50\% \text{CH}_3\text{COOH}$ at a temperature of about 90°C. The misorientation of the legs of the picture-frame crystal from the [111]-crystallographic axis was measured by von Laue X-ray diffraction to be less than 30'.
Internal stresses in the crystal were reduced by annealing in an argon atmosphere at 800°C during 24 h and at 1200°C during 3 h. The domain structure in the legs of the crystal was studied by neutron depolarization experiments with the direction of polarization of both incoming and outgoing beam parallel to the leg illuminated by the beam. Our measurements at room temperature in zero field showed strong depolarization which decreased with the application of a magnetic field.
along the circumference of the sample and disappeared at a field strength $H$ above 200 A/m. These last measurements indicate that $H = 200$ A/m is the saturation field and that below the magnetic induction is not completely aligned along the easy [111] crystallographic axis of the legs. The "imperfect domain structure" in this low field region is presumably caused by internal stresses being present which may result from magnetostrictive deformations due to the shape and dimensions of the specimen. A calculation of the size of this effect is consistent with the observed saturation field. Neutron depolarization measurements at LN$_2$ temperature showed that the magnetic induction was aligned parallel to the legs even without field. This is reasonable as with decreasing temperature the crystal anisotropy increases more strongly than the magnetostriction.

3. Measurements and Results

A. General description of the measurement

The magnetization reversal process was studied applying a periodic bipolar magnetic field, block-shaped in time, along the circumference of the sample. The field strengths ranged from 190 to 1500 A/m. The pulse duration was sufficiently long to reverse the magnetic induction completely. The periodic time dependence of the neutron depolarization was measured during the reversal process by synchronizing the first storage channel for neutron counts with the periodic field. The maximal time resolution achieved was better than 10 µs. To confine the neutron beam a vertical slit, 0.4 mm wide and 5 mm high was permanently located in front of one of the legs. The experiments were performed at room temperature.

All measurements were performed using three different polarizer-analyzer adjustments. In the first two ones the incident beam was polarized parallel to the direction of propagation, $x$, of the beam i.e. perpendicular to the applied field. Both the $x$- and $y$- component of $\hat{P}$ of the outgoing beam were analyzed. These two elements, $D_{xx}$ and $D_{yx}$ of the depolarization matrix determine the Larmor precession of the
polarization vector of the beam by the magnetic induction parallel to the z-axis:

\[ \phi = \tan^{-1} \frac{D_{yx}}{D_{xx}} \]  

(2)

The angle \( \phi \) with the sample in saturation was 11.5 \( \times \) \( 2\pi \). The reduction in the degree of polarization is measured by:

\[ |D| = \sqrt{D_{xx}^2 + D_{yx}^2} \]  

(3)
The third polarizer-analyzer adjustment was chosen to be $D_{zz}$. This depolarization element senses all magnetic induction components perpendicular to the applied field direction while $|D|$ senses also variations along the field direction.

In fig. 2 the depolarization spectra $D_{xx}$, $D_{yx}$, $D_{zz}$ and $|D|$ are shown measured at a field strength $H = 195 \text{ A/m}$ with the slit at a central position across the width of the leg. During the reversal process the elements $D_{xx}$ and $D_{yx}$ oscillate in time having a phase shift of $\pi/2$ with respect to each other. The constant values of these elements beyond $t = 31 \text{ ms}$ indicate that then the magnetization reversal process is completed. A small depolarization is observed in $D_{zz}$ while $|D|$ shows a pronounced time dependent depolarization. The remaining depolarization in $|D|$ after completion of the reversal is caused by the neutron wavelength spread $\Delta \lambda / \lambda$ of about 0.03. During reversal the last depolarization which depends on the precession angle $\phi$ is superimposed on the depolarization caused by the magnetization distribution within the specimen.

B. Initial stage of the magnetization reversal process

Fig. 3 displays the precession angle $\phi$ and the depolarization $|D|$ and $D_{zz}$ measured at $H = 195 \text{ A/m}$ during the first 150 $\mu$s after field reversal. The angle $\phi$ was derived from the measured $D_{xx}$ and $D_{yx}$ using eq. (2). The quantity $|D|$ was corrected for the depolarization due to the wavelength spread in first order by dividing the measured $|D|$ by the depolarization observed for the saturated specimen. This is justified by the fact that the mean magnetic induction of the sample is close to its saturation value during the very initial stage of reversal. The time dependence of $\phi$ and $|D|$ is similar to the one observed in earlier experiments on the magnetic reversal in FeSi. The value of $D_{zz}$ close to 1 indicates that the magnetic induction remains parallel to the crystallographic [111]-axis of the leg. The decrease of $|D|$ just after field reversal is attributed to the growth of isolated regions in which magnetization reversal has already taken place which causes a spread in the precession angle of the neutrons. The growth of these regions is limited to the surface since the magnetic field inside the specimen is
FIG. 3. Neutron depolarization $|D|$, $D_{zz}$ and the Larmor precession angle $\phi$ during the first 150 $\mu$s after field reversal at $H = 195$ A/m with the slit at position 4 (see fig. 1).

shielded by eddy currents. With time progressing they merge and form a $180^\circ$ domain wall that moves towards the center of the specimen. A subsequent flattening of the $180^\circ$ domain wall decreases the spread in the precession angle consistent with the gradual increase of $|D|$ observed beyond $t = 50 \mu$s. The experimental results were compared with a simulation of isotropic expansion of reversed regions with half cylindrical shape and a mean distance $\delta \approx 90 \mu$m between the reversed regions was estimated from the minimum value $|D| = 0.25$ at $t = 40 \mu$s. The value of $\phi$ about zero during the first 40 $\mu$s is caused by the strong depolarization and, hence, gives no measure of the mean precession angle.
In order to study the magnetization reversal within the whole volume of one leg the time dependent neutron depolarization was measured with the neutron diaphragm placed at seven different positions across the width of the leg (see fig. 1). The results of $D_{xx}$ at a field strength $H = 910 \, \text{A/m}$ are shown in fig. 4a. At all slit positions an oscillatory behaviour in time is observed for $D_{xx}$. A similar behaviour was measured for $D_{yx}$ the only difference being a phase shift of $\pi/2$. The value of $D_{zz}$ was found to be close to 1 during the entire reversal process.

All measured depolarization elements $D_{xx}$, $D_{yx}$, and $D_{zz}$ are consistent with a magnetic reversal developing by the movement of a $180^\circ$ domain wall from the surface towards the center of the leg. The terms "front wall" and "side wall" will be now used to refer to those parts of the domain boundary which were nucleated at the front and the side surface of the leg, respectively (see fig. 1). The phase $\phi$ (eq. (2)) determines the distance from the position of the wall to the surface along the direction of propagation of the neutrons. From the maximal difference in phase $\phi$ of about $\pi$ measured at the different slit positions a maximal variation of about 20 $\mu$m in the domain wall-surface distance is estimated at any time. Hence, the "front walls" move as nearly flat walls towards the center of the leg.

In the spectra $D_{xx}$ measured with the slit positioned close to the center of the leg the oscillations are present until the magnetization reversal is completed within the entire crystal while the oscillations vanish prior to that moment in the spectra measured closer to the edges of the leg. The onset of constant values in the depolarization measured at the outer leg positions indicate that the "side wall" leaves that part of the leg illuminated by the neutrons. The time that the "side wall" enters the region determined by the slit position was found from the time at which the wall left the adjacent region with the slit position closer to the edge of the leg. Small values of $|D|$ were found at the time that the "side wall" enters the region exposed to the neutrons subsequently followed by a continuous increase of $|D|$ towards a final value during the time interval that the wall passes that region. Fig. 4b displays the depolarization $|D|$ corrected for depolarization by the
neutron wavelength spread. This correction could easily be performed as long as the phase $\phi$ was well determined by the elements $D_{xx}$ and $D_{yx}$. During the final stage of magnetization reversal starting with $|D| = 0$ the measured depolarization $|D|$ was corrected by dividing $|D|$ by the depolarization measured with the specimen magnetically saturated. All spectra in fig. 4b show a reduced depolarization just after the initial stage of magnetization reversal consistent with the picture shown in fig. 3. Thereafter, an increase in the depolarization is observed with a minimum in $|D|$ at an early stage during magnetization reversal for the spectra measured with the slit near to the edges of the leg while the minima in $|D|$ observed at the spectra with the slit near the center are located much closer to the end of magnetization reversal. Since $D_{zz}$ is close to one the depolarization has to be caused by a spread in the precession angles of the neutrons. As already pointed out the minima in $|D|$ occur at the time at which the "side wall" enters the region exposed to the neutrons. Hence this depolarization indicates a curvature in the domain boundary at those places at which the "side walls" and "front walls" meet each other.

From the knowledge of the positions of the "side walls" of the shrinking domain wall obtained from the time dependent depolarization measured at the different slit positions in combination with the positions of the "front walls" derived from the phase $\phi$ it became possible to determine the shape of the domain wall during magnetization reversal. The main result of the analysis is that the distance between the domain wall and the specimen surface is nearly equal along the entire contour of the shrinking domain wall. Hence, in principle, we find the same velocity of the entire domain wall at a time. The wall velocity $\dot{w}$ normalized to the applied field strength $H$ is shown in fig. 5 as a function of the domain wall-surface distance $w$. The data displayed were obtained from the precession angle $\phi$ measured at $H = 195$ A/m. Data obtained at other magnetic fields very well fit the same $\dot{w}/H (w)$ curve.

**FIG. 4.** Neutron depolarization $D_{xx}$ (a) and $|D|$ (b) measured at $H = 910$ A/m during magnetization reversal at the positions indicated in fig. 1.
FIG. 5. Domain wall velocity \( \dot{w} \) normalized to the applied field strength as a function of the wall-surface distance \( w \) (see fig. 6) for the nickel picture-frame crystal. The line indicates the result of the model calculation as explained in the text.

4. Discussion

At first sight the same velocity of the domain wall along its entire contour seems to be surprising because the external magnetic field at the inner edge of the leg is about twice as large as the field at the outer edge. However, the eddy currents induced by the flux change due to the shrinking domain can be assumed to flow around the magnetically un-reversed region generating an induction field opposite to the applied field \( \vec{H} \) (see fig. 6). Since the current is free of sources and the cross
FIG. 6. Sketch of the magnetization reversal as derived from the measured depolarization (fig. 4a, b) within one leg of the picture-frame crystal. The ring shape and the symbols are defined which were used for the model calculations (see eq.(4)).

section for the current near to the outer edge is larger than that near to the inner edge the current density \( j \) is expected to decrease from the inner to the outer edge. This decrease in \( j \) happens to be with the same factor as the decrease in \( H \). The last may give a qualitative explanation for the fact that the wall velocity is the same along the entire domain boundary at a time.

Assuming a uniform wall motion the eddy current limited velocity \( \dot{w} \) of the domain boundary can be calculated as a function of the position \( w \) (see fig. 6) from the magnetostatic energy, supplied by the field due to the change of magnetization, which is transferred in heat by the induced eddy currents. The eddy current density \( j = \sigma E \) is determined by the induction law \( \oint \mathbf{B} \cdot d\mathbf{r} = - \frac{d}{dt} \int \mathbf{B} \cdot d\mathbf{S} \). Due to the lack of symmetry in the geometrical shape of the specimen studied an analytical calculation of the wall velocity \( \dot{w} \) becomes cumbersome. In first order the picture-frame shape can be approximated by a ring with the same cross section as the
leg of the crystal. It is assumed that the domain boundary in the ring moves with a velocity which is the same along its contour. Limiting the eddy currents to the region that is already reversed the wall velocity normalized by the mean applied field strength $H$ is calculated to be

$$\frac{w}{H} = \frac{-R}{4\sigma B_s} \left\{ - (k+1)d + 4w \right\} \left( \int_0^{(kd-2v)/(1/x_1 + 1/x_2) + 2\ln(x_2/x_1)} dv \right)^{-1}$$

with $x_1 = R - (d/2 - v)$ and $x_2 = R + (d/2 - v)$. The quantity $\sigma$ denotes the electrical conductivity. In fig. 6 a cross section through the ring is drawn giving the significance of the other quantities. In the limit $R \gg d$ and $K \ll 1$ the well-known solution for the eddy current limited domain wall motion in a thin tape is obtained:

$$\frac{w}{H} = \frac{1}{2\sigma B_s w}$$

Both solutions are only valid for $w \neq 0$.

The velocity $w$ as a function of $w$, calculated by the exact solution of the model using eq. 4 with an electrical conductivity $\sigma = 1.46 \times 10^7 \Omega^{-1} \text{m}^{-1}$ for pure nickel at 300 K and $R = 6.1 \text{ mm}$, is displayed in fig. 5. The mean radius $R$ was determined by setting $2\pi R$ equal to the mean circumference of the frame.

Moreover van Schaik's data of the wall velocity $\dot{w}$ will be considered which were measured in an iron-silicon picture-frame crystal that had a leg cross section with a thickness to width ratio about of 0.09. With the correct value for the magnetic field $H$ which was taken a factor of 4.2 too large in ref. 6 the normalized wall velocity $\dot{w}/H$ is compared with the result of $\dot{w}/H$ obtained from the model above using $\sigma = 2 \times 10^6 \Omega^{-1} \text{m}^{-1}$ for iron (3.5% Si) and $R = 6.2 \text{ mm}$ (fig. 7). In both, the nickel and the iron specimen, the measured wall velocity is smaller than the calculated one in the initial stage of the magnetization reversal process. In an intermediate region of $w$ an overlap is found between the
FIG. 7. Normalized domain wall velocity \( \dot{w}/H \) as a function of \( w \) measured at a picture-frame iron silicon single crystal. Data are taken from ref. 6. The line indicates the result of the model calculation as explained in the text.

Experimental and calculated data. During the second half of the magnetization reversal the calculations yield higher values of \( \dot{w}/H \) than were obtained from experiment.

The difference in velocity calculated in the model and obtained by the experiment during the initial stage of the reversal process is perhaps caused by the fact that in the last case a flat wall still has to be formed. The existence of a flat wall is a basic assumption of the model. The different behaviour of \( \dot{w}(w) \) between the experimental and calculated data during the magnetization reversal process is not fully understood and may be related to the simplifications of the model used. First, the ring shape may be inappropriate to approximate the actual picture-frame geometry. It is unknown whether the magnetization reversal at the corners of the picture-frame takes place in a way similar to that in the legs. However, if flux closure is accounted for during the entire magnetization reversal process the latter should be expected. Second,
the motion of the domain wall may be uniform neither in space nor in time. The uniform motion of the wall indicated from the analysis of our measurements is based on a time and length scale larger than 10 µs and 10 µm, respectively.

In conclusion, from the time dependent measurement of two depolarization matrix elements which determine the mean magnetic induction \( \langle \hat{B} \rangle \) parallel to the applied field and the variation in B along the field direction it was possible to determine the spatial and temporal evolution of the entire domain boundary during the magnetization reversal process within one leg of a picture-frame crystal. The wall velocity \( \dot{w} \) measured as a function of its position \( w \) is qualitatively consistent with the result of a model calculation for a ring shaped specimen in which the velocity \( \dot{w} \) is determined by eddy current losses caused by the uniform motion of the domain boundary. Systematic quantitative deviations are found in the initial and final stages of the magnetization reversal. To attribute this difference in \( \dot{w} \) exclusively to the non-uniform motion of the wall either in space or time seems to be unjustified because the calculated and the measured wall velocity are in agreement within a small intermediate region of \( w \). Due to the lower symmetry of the picture-frame relative to the ring it is much more difficult to take into account the magnetization reversal process close to the corners of the specimen.

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CHAPTER VIII

SUMMARY

In this thesis neutron depolarization experiments were performed on amorphous and crystalline ferromagnetic materials. The subjects studied are 'domain structure in magnetically weak uniaxial amorphous ferromagnetic ribbons', 'static critical behaviour at the ferromagnetic-paramagnetic phase transition', 'small magnetic anisotropy in nickel near $T_c$', and 'magnetization reversal in conducting ferromagnets'. All subjects will be summarized separately and the main conclusions are added to the experimental results.

The neutron depolarization analysis of the domain structure in a magnetically uniaxial ferromagnet with weak anisotropy was carried out on amorphous ribbons. An easy axis of magnetization perpendicular to the plane of the ribbon was induced by applying external stresses to the ribbon. The data analysis was performed using a Landau Lifshitz type domain structure. The sensitivity of the method used was high enough to measure the flux closure near the surface, in particular, the depth of the closure domains at the faces of the ribbon. Under the influence of an external magnetic field $\mathbf{H}$ in the ribbon plane a direction of preference for the $180^\circ$ Bloch walls parallel to $\mathbf{H}$ was found. This effect is attributed to the magnetostatic energy within the Bloch walls due to the applied field. From the analysis of the magnetization process with the Bloch walls initially parallel to the applied field it turns out that changes in the magnetization distribution are dominated by rotation processes in the bulk. The depth of the flux closure at the faces of the ribbon was found to be unchanged during the magnetization process. A model based on a Landau Lifshitz type domain structure is proposed for the magnetization process which is consistent with the experimental result.

The Larmor precession angle $\phi$ of the polarization vector $\mathbf{P}$ was measured around the ferromagnetic phase transition in polycrystalline
iron and nickel. The specimen were ring-shaped in order to keep demagnetizing fields as small as possible. The magnetic induction $B$ could be determined with a precision of about $1 \times 10^{-4}$ Tesla. From these precise values of $B$ which were measured as a function of temperature $T$ with a resolution in $T$ in the mK range precise values for the critical exponents $\beta$ and $\gamma$ were derived which describe the singularities of the magnetic induction and the magnetic susceptibility, respectively. The uncertainties in $\beta$ and $\gamma$ reported here are about a factor of three smaller than those reported in ferromagnetics so far. The precision in the measurement of $B$ achieved by the neutron depolarization technique exceeds by more than a factor of 20 the precision of the best-known method used up to now, the determination of the hyperfine field by means of the Mössbauer effect. Hence for the first time it is possible to get reliable information of the corrections to scaling in ferromagnets close to $T_c$. The knowledge of these correction terms is essential to derive the asymptotic values of the critical exponents. The asymptotic values for the exponent $\beta$ found for iron and nickel are $0.373(4)$ and $0.390(3)$, respectively. The $\beta$ value for iron is only slightly above $0.365$ which is calculated for a system of the $(3,3)$-universality class, while the value of $\beta$ found for nickel exceeds the last value significantly. This indicates that nickel does not belong to the $(3,3)$-universality class. It will be noted that values of $\beta$ for nickel reported up to now are only effective exponents which are consistent with our data in the same temperature regions. The correction to scaling was determined from our experiments and the asymptotic value of $\beta$ should be larger than the effective exponents found. The exponents $\gamma$ found for both iron ($\gamma=1.33(2)$) and nickel ($\gamma=1.315(15)$) are smaller than $1.387$ calculated for the case of an ideal three-dimensional Heisenberg system. From the neutron depolarization measurements in nickel one correction to the scaling amplitude and four critical amplitudes could be derived. The last four quantities allow to calculate two universal amplitude ratios.

The approach to magnetic saturation was studied by neutron depolarization in a ring specimen of polycrystalline nickel in order to
derive the strength of the magnetic anisotropy. The measurements were carried out in a temperature range from $T_C - 12$ K to $T_C - 2$ K in which the anisotropy becomes too small to be measurable by the techniques usually applied. The field dependence of the mean magnetic induction $B$ close to saturation could be fitted to $B = B_s (1 - a/H - b/H^2)$. It was found that the magnetic anisotropy $K = 3.6B_s^3$ decreases nearly linearly with temperature from $18$ J/m$^3$ at $T_C - 12$ K to $2$ J/m$^3$ at $T_C - 2$ K. This linear decrease of $K$ versus temperature parallels a similar decrease of the magnetostriction $\lambda_s$ versus $T$. From the measurements it is not possible to separate the crystalline anisotropy from the magnetoelastic one. Nevertheless the measured values give an upper limit of the crystalline anisotropy present. Due to the high precision with which $B$ is determined by the neutron depolarization technique and the possibility to apply arbitrarily small external magnetic fields a lower limit in the anisotropy that can be measured does in principle not exist.

The magnetization reversal in a picture frame single crystal of nickel with the legs parallel to the [111] crystallographic axes was studied by the time dependent neutron depolarization technique. It was demonstrated by van Schaik et al that this technique is unique to study both the spatial and temporal evolution of the magnetization inside the volume of a ferromagnet. The experiments presented here were performed on a specimen with a nearly square leg cross section while the former neutron depolarization analyses were applied to study the magnetization reversal within an FeSi(3.5%) picture-frame crystal the leg cross section of which was of a nearly flat geometrical shape. Similar to the results obtained by van Schaik et al the observed depolarization in the nickel specimen is consistent with a magnetization reversal starting at the surface and subsequently a 180° domain wall is formed which moves towards the center of the specimen. By measuring the depolarization at different positions across the width of one leg it became possible to determine the spatial and temporal evolution of the entire domain boundary during the magnetization reversal process. The wall velocity
appears to the same along the entire domain boundary at a time. The last result is based on a time and length scale larger than 10 µs and 10 µm both of which are determined by the resolution of the experimental set-up. The measured wall velocity was found to be qualitatively consistent with a result of a model calculation applied to a ring shaped specimen. The quantitative deviations of the model calculations from the experimental data are not fully understood. It was assumed in the model that the wall motion is uniform and that the magnetostatic energy supplied by the field due to the change of magnetization is completely transferred in heat by the induced eddy currents.
Samenvatting

Neutronen depolarisatie metingen zijn verricht aan amorfe en kristallijn ferromagneten.

De domeinstructuur is onderzocht in een amorf magnetisch folie met de voorkeursas van de magnetisatie loodrecht op het vlak van het folie. Uit de neutronendepolarisatie analyse blijkt dat de 180°-Bloch wanden in het preparaat zich gaan richten langs een aanwezig magnetisch veld in het folievlak. Het statische magnetisatie proces kan worden beschreven door draaiing van de lokale magnetisatie in de richting van het aangelegde veld. De diepte van de afsluitdomeinen dicht bij het oppervlak blijkt niet te veranderen gedurende het magnetiseren van het preparaat.

Door het meten van de Larmor precessiehoek van een gepolariseerde neutronenbundel t.g.v. de magnetisatie in een verzadigd ringvormig preparaat kon de spontane inductie B met een precisie van 10^-4 Tesla bepaald worden. Uit metingen van B als functie van de temperatuur en een magnetisch veld in een gebied rondom de fase-overgang zijn de tot nu toe nauwkeurigste waarden van kritieke exponenten, amplituden en correctietermen van de schaalwetten in de materialen nikkel en ijzer bepaald.

De magnetische anisotropie in nikkel is bepaald in een temperatuurgebied van Tc -12 K tot Tc -2 K door metingen van het benaderen van de magnetisatie tot verzadiging. Door de hoge resolutie in de bepaling van de magnetische inductie B die door de neutronen depolarisatie techniek bereikt wordt, kunnen extreem lage anisotropieën gemeten worden. De gevonden temperatuurafhankelijkheid van K in nikkel wijst op een anisotropie van magnetostrictieve oorsprong.

De magnetisatie-omkeer in een nikkel raamkristal met een bijna quadratische beendoorsnede is bestudeerd met de tijdsafhankelijke neutronen depolarisatie techniek. Het ommagnetiseren gebeurt door het bewegen van een 180°-domeinwand parallel aan het oppervlak in de richting van het centrum van een kristalbeen. Het ruimtelijke en tijdelijke verloop van
de wand gedurende het ommagnetisatieproces kon uit de analyse bepaald worden en wordt vergeleken met op wervelstromen gebaseerde modellen.
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