The Influence of Interlayer Exchange Coupling on Magnetic Ordering in Fe-based Heterostructures

MARTIN PÄRNASTE
Dissertation presented at Uppsala University to be publicly examined in Häggsalen, Ångström laboratorium, Lägerhyddsvägen 1, Uppsala, Thursday, May 31, 2007 at 13:15 for the degree of Doctor of Philosophy. The examination will be conducted in English.

Abstract

Temperature dependent magnetization measurements were conducted on Fe-based heterostructures. A linear increase of the magnetic critical temperature with increasing Fe thickness was found for Fe/V superlattices with strong interlayer exchange coupling. For weakly coupled Fe/V superlattices anomalously large values of the critical exponent $\beta$ were attributed to differences in the effective interlayer exchange coupling in the surface region and in the interior of the superlattice stack.

Hydrogen loading of a sample containing a thin Fe film, up to a maximum pressure of 4 mbar gave an increase of the magnetic critical temperature of $\approx$21 K. A sample with a double layer of Fe, exchange coupled over V, showed oscillations in the critical temperature when loaded to increasing pressure of hydrogen. The oscillations in the critical temperature indicate the presence of quasi-2D phases.

Superlattices of Fe and V were investigated by x-ray magnetic circular dichroism. It was found that the orbital magnetic moment shows the same trend as the magnetic anisotropy energy with thickness of the Fe layers. A model which takes into account a varying strain and interface density successfully described the changes in the orbital magnetic moment.

The magnetization was measured as a function of temperature for a series of magnetically $\delta$-doped Pd samples. A thin film of Fe induced a magnetic moment in surrounding Pd layers, leading to a magnetic thickness one order of magnitude larger than the thickness of the Fe film. A crossover in the magnetic spatial dimensionality was found as the thickness of the Fe film increased from $\approx0.4$ monolayers to $\approx1$ monolayer. First principle calculations of the magnetization profile together with a spin wave quantum well model were used to explain the dimensionality crossover by an increase in the available thermal energy for population of perpendicular spin wave modes.

Keywords: magnetism, magnetic dimensionality, MOKE, magneto-optics, hydrogen loading, phase transitions

Martin Pärnaste, Department of Physics, Materials Physics, Box 530, Uppsala University, SE-75121 Uppsala, Sweden

© Martin Pärnaste 2007

ISSN 1651-6214
urn:nbn:se:uu:diva-7885 (http://urn.kb.se/resolve?urn=urn:nbn:se:uu:diva-7885)
Science! Curse thee, thou vain toy
- Capt. Ahab, from 'Moby Dick' by H. Melville

M♡ I och Å
List of Papers

This dissertation is based on the following papers, which are referred to in the text by their Roman numerals.

I  Temperature dependent magnetization and susceptibility of Fe$_n$/V$_7$ superlattices
   M. Pärnaste, M. van Kampen, R. Brucas, and B. Hjörvarsson (2005)
   *Phys. Rev. B*, 71:104426

II  The influence of interlayer exchange coupling on $T_c$ in Fe/V superlattices
    M. Marcellini, M. Pärnaste, B. Hjörvarsson, G. Nowak, and H. Zabel (2007)
    *submitted to Phys. Rev. B*

III Oscillatory exchange coupling in the two dimensional limit
    M. Pärnaste, M. Marcellini, and B. Hjörvarsson (2005)
    *J. Phys.: Condens. Matter*, 17:L477

IV  The effect of strain and interfaces on the orbital moment in Fe/V superlattices

V   Dimensionality crossover in the induced magnetization of Pd layers
    *submitted to J. Phys.: Condens. Matter*

VI  Magnetic ordering in Fe $\delta$-doped Pd
    *in manuscript*
Reprints were made with permission from the publishers.

I coauthored the following articles that are not included in this dissertation.

**VII** Stability of the induced magnetic V moment in Fe/V superlattices upon hydrogen loading  
*Superlatt. Microstruct.*, doi:10.1016/j.spmi.2006.08.008

**VIII** Remote control of the exchange splitting in magnetic heterostructures  
*submitted to Europhys. Lett.*

**Comments to my contribution**

The level of my participation is reflected by the position of my name in the author list. In all publications I actively participated in the planning of the experiments and in the manuscript preparation. Beyond that, my participation can be summarized as

I Responsible for the MOKE magnetic characterization and data analysis.

II Participated in the sample preparation, the MOKE magnetic characterization, and data analysis.

III Participated in the MOKE and XMCD measurements, data analysis, anisotropy calculations, and modeling.

IV Responsible for the hydrogen loading, MOKE magnetic characterization and data analysis, participated in the sample preparation.

V Responsible for MOKE magnetic characterization and data analysis, participated in the sample preparation.

VI Responsible for MOKE magnetic characterization and data analysis, participated in the sample preparation.
Contents

Introduction ................................................................. 1
1 Phase transitions, critical phenomena, and magnetic measurements 3
  1.1 Magnetization and magnetic order ................................. 4
     1.1.1 Critical exponents ..................................... 5
     1.1.2 Correlation function .................................. 6
     1.1.3 Scaling laws .......................................... 8
     1.1.4 Universality classes .................................. 8
  1.2 Magnetic measurement techniques ............................ 10
     1.2.1 Magneto-optical methods ................................. 10
     1.2.2 Using the Kerr effect ................................ 16
     1.2.3 Other methods ......................................... 19
2 Magnetism and magnetic dimensionality ............................. 21
  2.1 Magnetism of 3d transition metals .............................. 21
  2.2 Magnetic anisotropy ............................................ 23
  2.3 Exchange couplings .............................................. 24
     2.3.1 Interlayer exchange coupling .......................... 24
  2.4 Magnetic dimensionality ......................................... 27
     2.4.1 The 2D-XY dimensionality class ...................... 28
     2.4.2 Dimensionality crossovers ............................... 30
3 Analysis and interpretation of magnetic measurements ............... 33
  3.1 Magnetic measurements .......................................... 33
     3.1.1 Magnetic susceptibility ................................ 38
  3.2 Temporary loading with hydrogen .................................. 40
     3.2.1 Effects of hydrogen loading on magnetization ............ 40
     3.2.2 Practical hydrogen loading ............................. 41
     3.2.3 Hydrogen loading of Fe/V heterostructures ............. 42
4 Summary of papers .................................................... 43
  4.1 Magnetization and magnetic susceptibility of Fe\textsubscript{n}/V\textsubscript{7} superlattices (Paper I) ......................... 43
  4.2 Magnetic dimensionality of Fe/V (Papers II and III) .......... 44
  4.3 Strain and interface effects in Fe/V superlattices (Paper IV) ... 45
  4.4 Dimensionality of induced Pd magnetization (Papers V and VI) 46
Summary in Swedish ....................................................... 49
Acknowledgments ........................................................... 53
A "Homer" laboratory measurement system .............................. 55
  A.1 Overview .......................................................... 55
Introduction

The dimensionality of a magnet, i.e., the magnet’s external dimensions and internal directional restrictions, are largely responsible for the behavior of the magnetization. This is especially true as the magnetic order in a magnet is undergoing a phase transition. By reducing physical extensions, e.g., thickness or width of a magnetic specimen it is possible to realize a two-dimensional or one-dimensional magnet, c.f. Fig. 1. Furthermore, the spin dimensionality, i.e., the degrees of freedom for atomic magnetic spins are affected by the reduced external dimensions. A thin iron film favors a magnetization direction in the plane of the film whereas in a larger, or bulk, piece of iron the magnetization is free to rotate more or less isotropically. Figure 2 shows an artistic illustration of possible spin dimensionalities.

Various observables, e.g., the magnetization and the magnetic susceptibility, display critical behavior as a magnet undergoes a ferromagnetic-paramagnetic phase transition. The magnetization vanishes, while the magnetic susceptibility reaches a maximum at the critical temperature $T_c$, which marks the transition. The decay of the magnetization and the peak of the susceptibility can be approximated with power law behavior and parametrized with (critical) exponents. The values of these exponents reflects the dimensionality of magnetic fluctuations.

Magnetic layers separated by layers of non-magnetic metals interact through interlayer exchange coupling (IEC). Magnetic multilayers, i.e., structures with magnetic and non-magnetic layers alternately stacked, are very useful tools in the investigation of the the effect of magnetic IEC on the dimensionality. The IEC between the magnetic layers is dependent on

![Figure 1: Artistic illustration of different spatial dimensionalities.](image)
Figure 2: Illustration of possible spin dimensionalities. These are often referred to as, from left to right, Heisenberg, XY, and Ising spins.

the thickness of the non-magnetic layers. By varying the thickness of the non-magnetic layers it is possible to get three-dimensional behavior even though the magnetic layers individually show two-dimensional behavior. For some materials combinations, e.g., iron and vanadium, antiferromagnetic alignment between neighboring magnetic layers is also possible. The incorporation of hydrogen in magnetic multilayers offers a route to modify the IEC in a single sample. By exposing a magnetic multilayer to hydrogen gas of various pressure it is possible to continuously tune the strength of the interlayer exchange coupling.

The work presented in this dissertation is aimed toward an understanding of the connection between extension and order for magnetic systems. An increasing extension of a low-dimensional magnetic film, e.g., created by increasing the thickness of a magnetic film or by enhancing the interlayer exchange coupling between magnetic layers, will lead to an increasing dimensionality and a new behavior for the magnetic fluctuations.

This dissertation is based on a number of research articles which are presented in the end. The text found in the chapters before the articles is intended to serve as an introduction to the field of experimental investigations of the effect of extension on order.
1. Phase transitions, critical phenomena, and magnetic measurements

A phase transition brings a system (an ice cube, a piece of magnetic material, or water in a pot on the stove) from one phase to another. After the transition the properties of the system are altered significantly in one or more ways. A phase can be defined, following Goldenfeld [1], as: If the sum of the coupling constants $K_n$ define the Hamiltonian for the system and $f[K]$ is the free energy per volume unit, then a region where $f[K]$ is analytic is called a phase. In this work solely magnetic phase transitions are considered where the coupling constants $K_n$ represent different magnetic couplings.

For a ferromagnet, the transition from an ordered ferromagnetic phase to the disordered paramagnetic phase is an example of a continuous second-order phase transition. The transition is classified as continuous since the magnetization decreases continuously to zero as the ordering temperature is approached. At the ordering temperature, second order derivatives of the free energy (e.g., the magnetic susceptibility) diverge, hence the transition is classified as being of second order. A second order phase transition does not involve the absorption or release of latent heat, the phase of such a system is therefore uniquely determined by the temperature, given that all other external parameters are unchanged. The phase change in the melting or freezing of ice is a discontinuous transition, either the water is in its solid phase or not, there is nothing in-between and the transition between the solid and the liquid phases takes place at a fixed temperature. At this temperature latent heat is absorbed or released, i.e., the ice is melting or freezing. Such transitions are classified as being of first order.

Mostly, phase transitions are thermally driven, i.e., they are accomplished by a change of temperatures. This is, however, not the case for all phase transitions. For example the zero-temperature Ising model has a magnetic-field induced phase transition at $H = 0$, where $H$ is an external magnetic field. Liquid/gas transitions and percolation transitions can occur through changes in pressure and concentration, respectively. The discussion in this work will mainly consider thermally driven phase transitions.

A more detailed description of critical phenomena can be found in introductory textbooks, for example [1–4].

This chapter also gives an introduction to the magnetic measurement techniques that I have used in my work. The magneto-optical effects discovered

---

1This temperature is determined by other external parameters, e.g., pressure.
by Faraday and Voigt are briefly described and the Kerr effect, which has been my main investigative tool, is described in more detail together with a discussion on different experimental setups utilizing the Kerr effect.

1.1 Magnetization and magnetic order

To be able to describe the transition from an ordered low temperature phase to a disordered high temperature phase it is necessary to first define which parameter represents order. This is the order parameter and it should be chosen such that it has a finite value in the ordered phase, reflecting the degree of order, and that it should be zero in the disordered phase representing the total absence of (long range) order. The temperature at which the phase transition occurs is defined as the critical temperature \( T_c \). For ferro- and antiferromagnetic systems the critical temperature is referred to as the Curie temperature and the Néel temperature, respectively. Henceforth, the more general notation of a critical temperature will be used.

The order parameter may fluctuate in time and space, whereas the thermal average of the order parameter is a well defined function of, e.g., temperature. The definition of the order parameter as a fluctuating quantity is required for describing such phenomena as critical opalescence [2]. The critical exponents introduced in the next section, \( \beta \), \( \gamma \), etc., however, describe the variation of the thermal average of the order parameter.

For magnetic systems the commonly used order parameter is the local zero field mean magnetization \( \Phi(x) \), which is the mean magnetization in a small volume around \( x \) with no external magnetic fields present. \( \Phi(x) \) is a fluctuating quantity given by the sum of magnetic spins \( s_i \) enclosed within some volume \( V \) centered on \( x \)

\[
\Phi(x) = \frac{a}{V} \sum_{s_i \in V} s_i, \tag{1.1}
\]

where \( a \) is a constant. The thermal average of \( \Phi(x) \) is the zero field magnetic moment per unit volume, the magnetization \( m \), which is the order parameter for the ferromagnetic critical behavior discussed in this work.\(^2\)

When \( T_c \) is approached from below, the magnetization decreases and vanishes completely at the ordering temperature, the order parameter then remains zero as the temperature is increased further. Critical phenomena are not limited to the order parameter. Various response functions also show critical behavior, e.g., the magnetic susceptibility and the specific heat both diverge at the critical temperature.

\(^2\)The physical dimensions of ferromagnets are, to some extent, a function of the magnetization. This phenomenon is known as magnetostriction [5]. Thus, a more consistent definition of magnetization is the magnetic moment per atom.
1.1.1 Critical exponents

In the vicinity of the critical temperature the temperature dependence of various quantities such as the magnetization and the magnetic susceptibility can be described by critical exponents. The temperature dependence can be approximated by a power law expansion in the reduced temperature, $t = \frac{T - T_c}{T_c}$, where the power of $t$ is the critical exponent. The reduced temperature is a dimensionless linear variable used to measure the relative temperature deviation from the critical temperature.

The critical exponent of a general function $f(t)$ is for positive $t$ defined as

$$\lambda = \lim_{t \to 0} \frac{\ln(f(t))}{\ln(t)},$$

(1.2)

given that this limit exists. The function $f(t)$ can be generally expressed as:

$$f(t) = At^x(1 + C_1t^{y_1} + C_2t^{y_2} + \ldots) \quad y_i > 0.$$  

(1.3)

From which, as $t \to 0$, the behavior in the vicinity of $T_c$ is:

$$f(t) \sim t^\lambda,$$  

(1.4)

where $\lambda$ is the critical exponent of $f$. Hence, the phase transition can be described by the values of $\lambda$ for various observables. As will be discussed in Section 1.1.4, phase transitions can generally be classified in a finite number of universality classes based on the numerical values of the critical exponents.

No exact rule exists for defining the region around $T_c$ where the power law approximation is valid. This region varies with system type and, as will be described later, on the universality class.

At the critical temperature several temperature independent quantities can also be described by power law behavior and critical exponents. For example, the critical isotherm, i.e., the relation between magnetization and applied field at $T_c$ ($t = 0$), is described by a power law with the critical exponent $\delta$

$$H \propto |m|^{\delta} \text{sgn}(m).$$

(1.5)

The functional form of Eq. (1.4) for $t$ close to zero is such that a negative $\lambda$ corresponds to a function that diverges as $T_c$ is approached from below ($t \to 0^-$) while a positive $\lambda$ corresponds to a function which approaches zero as $t \to 0^-$. Furthermore, a smaller value of $|\lambda|$ results in a more abrupt change of $f(t)$ as $t \to 0^-$. Figure 1.1 shows hypothetical magnetization curves following power law behavior with different values of the critical exponent $\beta$ of the magnetization. There is an obvious difference in the behavior close to $t = 0$ between $\beta = 0.25$, represented by the full curve, and $\beta = 0.5$, represented by the dashed curve. $\beta = 1$ corresponds to a linear behavior of the (hypothetical) magnetization (dash dotted line). In Fig. 1.1 also the diverging behavior of a negative $\beta = -0.05$ is shown, represented by the dotted line.
Figure 1.1: Comparison of hypothetical magnetization curves following the theoretical power law. The sharpness of the transition is reflected in the critical exponent \( \beta \), where a smaller \( \beta = 0.25 \), represented by the full line, has a sharper transition than a higher \( \beta = 0.5 \), represented by the dashed line. \( \beta = 1 \) represents a linear decay of the magnetization, as indicated by the dash dotted line. The dotted line represents a negative \( \beta = -0.05 \), consequently the function diverges.

magnetization is, at least within the framework of transition metal heterostructures, unphysical and is used here merely as an illustration. Table 1.1 lists some of the critical exponents together with their respective functional definitions.

1.1.2 Correlation function

An important quantity linking the microscopic behavior of individual magnetic spins to the macroscopic view described by quantities such as the magnetization, the susceptibility, or the specific heat is the correlation function. The correlation function between two spins is the thermal average of the scalar product of spins \( s_i \) and \( s_j \):

\[
G^{(2)}(r) \equiv \langle s_i s_j \rangle.
\] (1.6)

The two-spin correlation function describes the thermal average of the alignment of the two spins \( s_i \) and \( s_j \) which are separated by \( r_{ij} \). The spin thermal average \( \langle s_i \rangle \) is zero above \( T_c \) whereas \( G^{(2)}(r) \) will vary with the distance between spins since the interaction between spins in the system favors parallel (or antiparallel) alignment of neighboring spins. In zero field above \( T_c \), \( G^{(2)}(r) \) will approach zero as \( r_{ij} \rightarrow \infty \) due to the decreased interaction between two largely separated spins.
Table 1.1: Definitions of various critical exponents, from Stanley [3].

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Exponent</th>
<th>Definition</th>
<th>Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific heat, zero field</td>
<td>$\alpha$</td>
<td>$C_H \propto t^{-\alpha}$</td>
<td>$t &gt; 0; H = 0; m = 0$</td>
</tr>
<tr>
<td>Magnetization, zero field</td>
<td>$\beta$</td>
<td>$m \propto (-t)^\beta$</td>
<td>$t &lt; 0; H = 0; m \neq 0$</td>
</tr>
<tr>
<td>Isothermal susceptibility, zero field</td>
<td>$\gamma$</td>
<td>$\chi \propto t^{-\gamma}$</td>
<td>$t &gt; 0; H = 0; m = 0$</td>
</tr>
<tr>
<td>Critical isotherm</td>
<td>$\delta$</td>
<td>$H \propto</td>
<td>m</td>
</tr>
<tr>
<td>Correlation length</td>
<td>$\nu$</td>
<td>$\xi \propto t^{-\nu}$</td>
<td>$t &gt; 0; H = 0; m = 0$</td>
</tr>
<tr>
<td>Connected correlation function</td>
<td>$\eta$</td>
<td>$G_c^{(2)} \propto \frac{1}{r^{d-2-\eta}}$</td>
<td>$t = 0; H = 0; m = 0$</td>
</tr>
</tbody>
</table>

If a magnetic field is applied to the system for $T > T_c$, $G^{(2)}$ will no longer tend to zero at large distances because each spin will align with the field even when the interaction between spins is lost. One way to circumvent this, i.e., to only measure the interaction between spins, is through the connected correlation function:

$$G_c^{(2)} \equiv \langle s_is_j \rangle - \langle s_i \rangle \langle s_j \rangle = \left( \langle s_i \rangle - \langle \langle s_i \rangle \rangle \right) \times \left( \langle s_j \rangle - \langle \langle s_j \rangle \rangle \right).$$  \hspace{1cm} (1.7)

Using the connected correlation function it is possible to consider only the collective fluctuations between spins $s_i$ and $s_j$ as the thermal average of each spin is subtracted, as indicated by the last term in Eq. (1.7). The connected correlation function is zero at $T = 0$ although the correlation function $G$ at $T = 0$ is not zero, the former function, however, gives a more relevant description of the system close to $T_c$ as $G_c^{(2)}$ will always be zero when there are no spin interactions present.

At temperatures different from $T_c$ the distance dependence of $G_c^{(2)}$ can be expressed as [4]

$$G_c^{(2)}(r) \sim r^{-\tau} e^{-r/\xi},$$  \hspace{1cm} (1.8)

with $\xi$ being the correlation length. Although $\langle s_i \rangle \neq 0$ below $T_c$, Eq. (1.7) measures only the correlation between spin fluctuations which means that Eq. (1.8) is valid also below $T_c$.

At $T_c$ the distance dependence of the connected correlation function can be described by a power law [4]

$$G_c^{(2)}(r) \sim \frac{1}{r^{d-2-\eta}},$$  \hspace{1cm} (1.9)

where $\eta$ is the critical exponent of the connected correlation function and $d$ is the spatial dimensionality of the system. For $T > T_c$ the critical behavior of the correlation length is described by the critical exponent $\nu$, c.f. Table 1.1.

The correlation- and connected correlation functions can be defined for any number of spins ($G^{(n)}$ and $G_c^{(n)}$ respectively). In most cases, however, adequate information is given by the two-spin correlation functions and higher order corrections are not necessary.
1.1.3 Scaling laws

The different critical exponents are not independent, but are related to each other through so-called *scaling laws* [6]. In 1965 Benjamin Widom [7, 8] suggested that the specific free energy close to $T_c$ could be approximated by a function $\psi$ of only one variable:

$$f(T, B) = t^{1/y} \psi \left( \frac{B}{t^{1/y}} \right).$$

(1.10)

Equation (1.10), the *Widom scaling hypothesis*, is the basis for the concept of scaling laws. By use of the Widom scaling hypothesis it is possible to calculate the zero-field magnetization through the relation:

$$m = -\left( \frac{\partial f}{\partial B} \right)_T = - (t^{(1-x)/y}) \psi'(0).$$

(1.11)

Identification with the definition of $\beta$ (from Table 1.1) yields:

$$\beta = \frac{1-x}{y}.$$  

(1.12)

The same procedure can be used to calculate the zero-field susceptibility, the zero-field specific heat, and the critical isotherm at $t = 0$:

$$\gamma = \frac{2x - 1}{y}$$

(1.13a)

$$\alpha = 2 - 1/y$$

(1.13b)

$$\delta = \frac{x}{1-x}.$$  

(1.13c)

The combination of the above expressions finally leads to the possibility of eliminating the variables $x$ and $y$, thereby obtaining relations between sets of critical exponents. These are the scaling laws, sometimes referred to as the exponent equalities. The relations obtained from these equations are known as Rushbrooke’s law [9] and Griffith’s law [10] and are presented in Table 1.2. Table 1.2 also presents Fisher’s law [11] and Josephson’s law [12, 13]. More scaling laws are found in, e.g., the book by Stanley [3].

Closely connected to the scaling laws is a relation between the correlation length and the magnetic susceptibility. This relationship, derived by Nicoll and Chang [14], has the form:

$$\chi \sim \xi^{2-\eta}.$$  

(1.14)

1.1.4 Universality classes

The critical behavior of different magnetic materials will, if properly scaled, collapse onto the same curve. Also, such seemingly different events as the
transition between liquid and gas phases and the separation of mixed chemicals can be described by similar or equal critical exponents.

In 1970 Leo Kadanoff introduced the concept of universality classes. The critical behavior of various systems is divided into universality classes depending on the spatial dimensionality \( d = 1, 2, 3 \) of the system and of the spin dimensionality \( n = 1, 2, 3 \). The cases of \( n = 1, 2 \) and 3 are commonly referred to as respectively Ising-, XY- and Heisenberg spin dimensionality. Systems belonging to the same universality class show identical (within errors) critical behavior under proper scaling.

Not all universality classes represent systems that are macroscopically ordered over a long range at non-zero temperature. Table 1.3 specifies whether or not a universality class represents a system that can sustain long-range order (LRO) for \( T > 0 \). As indicated in Table 1.3 the two-dimensional XY model requires some additional explanation. It was shown theoretically \([15–17]\) that 2D-XY systems can not have an ordered ferromagnetic (or antiferromagnetic) state above 0 K. It was, however, later shown that 2D-XY systems do have a state of topological order and theoretical predictions by Bramwell and Holdsworth \([18]\) give \( \beta = \frac{3\pi^2}{128} \approx 0.23 \) for the 2D-XY model. The 2D-XY model is described in some more detail in Section 2.4.1.

A summary of theoretical values for some of the critical exponents is presented in Table 1.4, it is straightforward to verify that the exponents for different models satisfy the scaling laws in Table 1.2. For example, the values in Table 1.4 pertaining to the 2D Ising-model \((d = 2, n = 1)\) satisfies both Rushbrooke’s law and Griffith’s law.

### Table 1.2: Some of the exponent equalities or scaling laws, from Binney et al. \([2]\).

<table>
<thead>
<tr>
<th>Relation</th>
<th>Derived by</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \alpha + 2\beta + \gamma = 2 )</td>
<td>Rushbrooke</td>
</tr>
<tr>
<td>( \alpha + \beta(\delta + 1) = 2 )</td>
<td>Griffiths</td>
</tr>
<tr>
<td>( (2 - \eta)\nu = \gamma )</td>
<td>Fisher</td>
</tr>
<tr>
<td>( \alpha + \nu d = 2 )</td>
<td>Josephsson</td>
</tr>
</tbody>
</table>

### Table 1.3: The table indicates which universality classes correspond to systems with long-range order (LRO).

<table>
<thead>
<tr>
<th>Spatial dimensionality</th>
<th>n = 1 (Ising)</th>
<th>n = 2 (XY)</th>
<th>n = 3 (Heisenberg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>LRO</td>
<td>(-)</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>LRO</td>
<td>LRO</td>
<td>LRO</td>
</tr>
</tbody>
</table>
Table 1.4: Values of critical exponents derived from soluble models or numerical calculations.

<table>
<thead>
<tr>
<th>Exponent</th>
<th>$d = 3$</th>
<th>$d = 2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$</td>
<td>$\approx 0.125$</td>
<td>$\approx 1/8$</td>
</tr>
<tr>
<td>$\beta$</td>
<td>$\approx 0.31$</td>
<td>$\approx 0.33$</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>$1.25$</td>
<td>$\approx 1.33$</td>
</tr>
<tr>
<td>$\delta$</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

1.2 Magnetic measurement techniques

This section is intended to give a brief review of the magnetic measurement techniques used in this work. The main part of the work is based on the magneto-optical Kerr effect where magnetization is measured through polarization changes in light reflected from a sample surface. The Kerr effect is sensitive enough to measure the magnetization of thin films in the monolayer (ML) range.

SQUID magnetometry has been used for characterization of some samples presented in this work. A SQUID magnetometer measures the total magnetic moment of a sample. For temperature dependent zero-field measurements of magnetization the specially designed "Homer" setup (see Appendix A) is more suitable than a SQUID magnetometer due to the effective triple mumetal shield, and the lack of remanent magnetic fields in the Helmholtz coils. SQUID magnetometry gives a quantitative measure of the magnetic moment which can be used for calibration of MOKE measurements.

The element specific measurements of the magnetization in Paper IV were done by x-ray magnetic circular dichroism. From these measurements it is also possible to separate contributions from spin and orbital magnetic moments.

1.2.1 Magneto-optical methods

Magneto-optics is a common term describing interactions between light and a magnetized material. Essentially, all magneto-optical phenomena are consequences of the Zeeman effect, i.e., the splitting of energy levels by the application of a magnetic field. Nevertheless, it is convenient to separately describe the magneto-optical effects arising from different field and light configurations.

Common for the magneto-optical effects is that they are manifested as changes of the polarization state of light incident on a magnetic medium. The magneto-optical effects are combinations of birefringence (different refractive indices for different polarization states) and dichroism (different absorp-
tion for different polarization states). The resulting effect on light after the magneto-optical interaction is, in the case of linearly polarized light, a rotation of the polarization axis and an introduced ellipticity.

The Voigt effect, also known as the magnetic linear birefringence, and the Faraday effect, also known as magnetic circular birefringence, describe the magneto-optical effect of linearly polarized light propagating in a medium magnetized perpendicular and parallel to the propagation direction, respectively. The Kerr effect describes polarization changes for light reflected from the surface of a magnetized material. The Kerr effect has three basic configurations depending on the relative orientation between the magnetization of the material and the incoming light, c.f. Fig. 1.2. The magneto-optical effects are phenomenologically described through the dielectric permittivity tensor which describes the interaction between matter and the electric field of the incident light. The effect of the material’s magnetic permeability on optical phenomena is small and is therefore normally neglected.

A classical explanation of magneto-optical effects is through the effect of the Lorentz force on the motion of electrons in a medium. For a non-magnetic medium, the electric field of right circularly polarized (RCP) and left circularly polarized (LCP) electromagnetic waves will drive the electrons in the medium into right and left circular motion with equal radii. When the medium is magnetized, the Lorentz force acting on the electrons will expand or shrink the radius of the electron orbit depending on the direction of the magnetization, thereby creating a difference in the dielectric constants for RCP and LCP light.

Below, a brief description of the different magneto-optical effects will be given. More detailed descriptions can be found in, e.g., Refs. [19–25].

The Faraday effect
In 1845 Michael Faraday discovered the effect of magnetic circular birefringence that eventually came to bear his name [23]. As light passes through a magnetic material (either inherently ferromagnetic or magnetized by an external field) the polarization state of the light is altered.

Plane polarized light transmitted through a piece of glass subjected to an externally applied magnetic field parallel to the propagation direction has its plane of polarization rotated. This is the magnetic circular birefringence. Figure 1.3 shows a schematic illustration of the Faraday effect. In an absorbing medium the applied magnetic field splits the absorption coefficients for right and left hand circular polarized light (RCP and LCP) which introduce an ellipticity to the transmitted light. This is the magnetic circular dichroism. The angle of rotation induced by the magnetic circular birefringence is to first approximation given by the rather simple expression

\[ \theta = VHL, \]  

(1.15)
Figure 1.2: The main Kerr configurations together with definitions of incident and reflected s and p polarizations.
where $H$ is the applied magnetic field and $L$ is the optical length in the medium. $V$, the Verdet constant, is a frequency- and temperature dependent materials parameter specifying the rotation angle per unit length and unit field strength.

One important feature of the Faraday effect is that the sign of $\theta$, as it appears in Eq. (1.15), is not dependent on the direction of the transmitted light, only on the sign of the applied magnetic field, $H$. A beam of light propagating in the field direction and subsequently reflected by a mirror back in the opposite direction will thus have its plane of polarization rotated by $2\theta$. This phenomenon makes the Faraday effect different from naturally occurring optical activity and the effect is used in, for example, optical isolators where reflected light is blocked by two subsequent polarization-plane rotations of 45°.

The Zeeman splitting of energy levels which gives rise to the Faraday effect results in two different refractive indexes, $N_L$ and $N_R$, for LCP and RCP light respectively. The difference in refractive index means that LCP and RCP light of equal angular frequency will travel through the material with velocities $c/N_L$ and $c/N_R$ respectively.

To describe the Faraday effect in terms of rotation and ellipticity one must consider the phase difference and attenuation of the light after passing through the medium. The Faraday rotation angle per unit length is given by the real part of the refractive indexes:

$$\theta_F(\omega) = \frac{\omega}{2c} \mathcal{R}(N_L - N_R).$$  \hspace{1cm} (1.16)

The Faraday ellipticity is given by the imaginary part of the refractive indexes;

$$\eta_F(\omega) = -\frac{\omega}{2c} \mathcal{I}(N_L - N_R).$$ \hspace{1cm} (1.17)
The ellipticity is defined as the ratio of the minor to the major axis of the ellipse. The complex indexes of refraction, $N_{\pm} = \varepsilon_{xx} \pm i\varepsilon_{xy}$, are given by the elements of the dielectric tensor.

The Faraday effect can be utilized to modulate the signal in Kerr effect measurement setups, as described below.

**The Voigt effect**

Woldemar Voigt discovered that light passing through a vapor subjected to a magnetic field introduces birefringence in the light. Subsequently Aimé Cotton and Henri Mouton discovered a similar effect in a liquid. Hence, the Voigt effect is sometimes referred to as the Cotton-Mouton effect, especially when considering liquids.

While the Faraday effect describes changes of the state of polarization of light propagation parallel to the magnetization direction (either from an external magnetic field or from spontaneous (ferro)magnetism), the Voigt effect describes changes for light propagation in a material magnetized perpendicular to the propagation direction. In this case magnetic linear birefringence introduces an ellipticity to the light polarization and linear dichroism describes the attenuation. The Voigt effect is an even function of the magnetization, typically proportional to $m^2$.

**The Kerr Effect**

The Faraday and Voigt effects are manifest as an optical anisotropy in the transmission of polarized light through a magnetized material. A similar effect, but occurring in reflection of light from the surface of a magnetized material is the Kerr effect, named after John Kerr who discovered the effect in 1877 [26, 27].

Three basic configurations for the Kerr effect can be distinguished from the relative orientation between the $\mathbf{k}$-vector of the incident light and the magnetization vector $\mathbf{m}$ in the material. These are shown in Fig. 1.2 and are referred to as polar (Fig. 1.2(a)), longitudinal (Fig. 1.2(b)), and transverse (Fig. 1.2(c)).

The influence from the magnetization on an incident beam of linearly polarized light in the polar and longitudinal geometries is a rotation of the polarization plane and an induced ellipticity. For the polar and longitudinal geometries, a non-zero projection of the incident wave vector on the magnetization direction is required, c.f. the angle $\theta$ in Fig. 1.2. The magnetic response in the longitudinal geometry is thus proportional to $\cos(\theta)$ whereas the penetration depth is proportional to $\sin(\theta)$. A proper choice of the incidence angle $\theta$, depending on the preferred property, must therefore be done. In the polar geometry, where both the magnetic signal and penetration depth scales with $\sin(\theta)$, incident light perpendicular to the the sample plane is preferred. For the transverse Kerr geometry the effect of the magnetization is a change (decrease) of the light intensity and a phase shift with respect to the incident light.
Figure 1.4: Illustration of the geometry for $s$ and $p$ polarization.

The Jones matrices [28] for reflection and transmission of light at the interface between a non-magnetic and a magnetic medium are:

\[
\begin{pmatrix}
E_s^r \\
E_p^r
\end{pmatrix} = \begin{pmatrix}
rs \\
rp
\end{pmatrix} \begin{pmatrix}
E_s^i \\
E_p^i
\end{pmatrix} \quad (1.18a)
\]

\[
\begin{pmatrix}
E_s^t \\
E_p^t
\end{pmatrix} = \begin{pmatrix}
ts \\
tp
\end{pmatrix} \begin{pmatrix}
E_s^i \\
E_p^i
\end{pmatrix} \quad (1.18b)
\]

From Eqs. 1.18 it is possible to derive general formulas for the Kerr effect. These are for the polar, longitudinal, and transverse Kerr geometries respectively [19]:

\[
\Phi_{s,p} = \Re \frac{r_{sp}}{r_{ss}(r_{pp})} = 3 \frac{\eta^2[\sqrt{\eta^2 - \sin^2 \phi} \mp \sin \phi \tan \phi]}{(\eta^2 - 1)(\eta^2 - \tan^2 \phi)} Q, \quad (1.19a)
\]

\[
\Phi_{s,p} = \Re \frac{\pm r_{sp}}{r_{ss}(r_{pp})} = 3 \frac{\sin \phi \eta^2 (\sin \phi \tan \phi \pm \sqrt{\eta^2 - \sin^2 \phi})}{(\eta^2 - 1)(\eta^2 - \tan^2 \phi) \sqrt{\eta^2 - \sin^2 \phi}} Q \quad (1.19b)
\]

\[
\frac{\Delta I}{I} = -3 \frac{4 \tan \phi \eta^2}{(\eta^2 - 1)(\eta^2 - \tan^2 \phi)} Q \quad (1.19c)
\]

Where $\eta = n_2/n_1$ is the (complex) relative refractive index of the two materials, $Q$ is the magneto-optical parameter [19] and $\phi$ is the angle toward the surface normal of the incident (and reflected) light. The upper and lower signs correspond to $s$ and $p$ polarization respectively, c.f. Fig. 1.4.
1.2.2 Using the Kerr effect

The relative simplicity in employing the magneto-optical Kerr effect (MOKE) for magnetization measurements has made it a basic tool in many labs around the world. Advantages of MOKE are, e.g., that there is no need for vacuum- or *in-situ* conditions, as is the case for magnetic measurements by, e.g., electron techniques [29]. There is no principle limit to, or hindrance by, applied magnetic fields, which is also a limiting factor for electron techniques.

MOKE measurements are not limited to surface layers, but can be considered surface sensitive if magnetic layers are positioned close to the surface of a sample. There is no hindrance in probing magnetic layers covered by non-magnetic layers. Nevertheless, since the attenuation depth of the light is in the order of 100 Å, the signal-to-noise ratio will suffer when measuring the magnetic signal from layers deep inside a sample. The magnetic signal from magnetic multilayers will be a summation of the signals from each layer weighted by the depth attenuation of each layer.

The penetration depth is dependent on the angle of incident light as stated above. The incidence angle can therefore be used to vary the depth sensitivity in thin films [30]. Also, the measured signal is a summation of the magnetization in the region covered by the incident light. This can give rise to finite-size effects (see Chapter 3) if the probed region is composed of domains with different magnetization.

Measuring hysteresis loops or susceptibility signals from magnetic layers of only a few atomic layers is possible, and by separating the magnetic layer from, the possibly optically active, substrate material, contributions to the signal from the substrate are avoided. This is an advantage of MOKE as compared to techniques sensitive to the total sample magnetization, e.g., SQUID.

The incident light in a MOKE setup is generally polarized laser light, giving a narrow bandwidth, high intensity beam. Linear polarization is achieved by passive polarizers. Preferably, an intensity-stabilized laser should be used in order to minimize the effect of intensity variations. Various He-Ne lasers are commonly used, these are reliable and relatively low priced. Operating with visible light simplifies the optical alignment. However, since the magneto-optical response is wavelength-dependent [19, 31] there may be situations where wavelengths of invisible light are preferred.

Other light sources, such as mercury or xenon lamps are used in applications which require high light intensity but where one wishes to avoid the coherence of a laser. One example of such applications is Kerr microscopy [32], where a large portion of the sample is illuminated by polarized light in order to visualize domain formation. In this case a coherent beam could cause unwanted interference. In spectroscopic magneto-optics [33] the spectrum of ordinary lasers is too narrow, meaning that tunable wavelength lasers or bright lamps are required.
A common usage of MOKE is in the crossed-polarizer configuration, where two polarizing crystals or sheets are used. The first one, henceforth the “polarizer”, is used to obtain a linear polarization of the incident light. Consider the case with \( p \)-polarized light incident on a ferromagnetic sample. The interaction between the electric field of the incident light and the sample will give rise to a Kerr rotation of the incident light, introducing an \( s \)-component in the reflected light proportional to the magnetization. By measuring this induced \( s \)-component it is thus possible to measure the magnetization of the sample.

By setting the second polarizer, the ”analyzer”, at 90° angle with respect to the polarizer it is possible to detect the \( s \)-component, \( E_s \), in the reflected light. The light intensity reaching a detector placed after the analyzer will be proportional to \( |E_s|^2 \). Since the rotation of the polarization is proportional to the magnetization in the sample, the intensity reaching the detector is proportional to the square of the sample magnetization.

A more useful configuration is the nearly-crossed polarizer configuration where the analyzer is set at an angle \( \delta \) away from total light extinction, i.e., at an angle \( (90 + \delta)° \) with respect to the polarizer. The light after reflection from the magnetic sample will be composed of a dominating \( p \)-component and the induced \( s \)-component. The ratio \( E_s/E_p \) is the complex Kerr rotation, i.e. \( E_s/E_p = \phi' + i\phi'' \), with \( \phi' \) being the Kerr rotation and \( \phi'' \) the Kerr ellipticity [23].

The light after passing the analyzer can then be written

\[
E_s \cos(\delta) + E_p \sin(\delta),
\]

(1.20)

with intensity

\[
I \propto |E_s \cos \delta + E_p \sin \delta|^2 \approx |E_s + E_p \delta|^2 = E_p^2 |E_s/E_p + \delta|^2.
\]

(1.21)

Using \( E_s/E_p = \phi' + i\phi'' \), Eq. (1.21) can be expressed as

\[
I \propto |E_p|^2 \delta^2 (1 + 2\phi') = I_0(1 + 2\phi'),
\]

(1.22)

where \( I_0 = |E_p|^2 \delta^2 \) is the intensity for a non-magnetic sample. The intensity described by Eq. (1.22) is proportional to both the incident intensity and to the rotation of the polarization, i.e., the magnetization in the sample.

By simultaneously measuring the intensities before and after the analyzer it is possible to compensate for variations in the incident light intensity, caused by, e.g., instabilities in the laser output. In order to obtain quantitative measures of magnetic moments, the MOKE signal must be calibrated to a quantitative measure of the magnetization, obtained from, e.g., SQUID measurements [34, 35].

In the nearly-crossed polarizer configuration one often uses a modulation of the light in order to improve the signal-to-noise ratio. One way to accomplish this modulation is through a Faraday modulator, which consists of a coil.
wound around a glass rod. An ac-current in the coil generates an ac-magnetic field which in turn introduces a modulated Faraday rotation to the light polarization. The rotation of the polarization plane is dependent on the Verdet constant of the glass, the length of the rod and on the current passing through the coil. Typically, the modulation signal is sinusoidal with a frequency, $f \approx 1$ kHz. After the light passes the analyzer the modulated polarization corresponds to a modulation of the light amplitude

$$E = E_0 \sin(\omega(t) + \delta + \theta(H)) \approx E_0(\omega(t) + \delta + \theta(H)),$$  \label{eq1.23}

where $\omega(t) = A \sin(2\pi ft)$ corresponds to the Faraday modulation, $\delta$ is the analyzer offset angle, and $\theta(H)$ is the rotation of the polarization induced by the magnetization in the sample. The signal picked up by the detector is proportional to the light intensity

$$I = E^2 \approx E_0(\omega(t)^2 + 2\omega(t)\delta + 2\omega(t)\theta(H) + 2\delta\theta(H)),$$  \label{eq1.24}

where the term $\omega(t)^2 = A^2 \sin^2(2\pi ft) = \frac{A^2}{2} \cos(4\pi ft)$ describes an oscillation of frequency $2f$. Thus, the detected signal consists of the ground frequency ($f$) and of the first harmonic ($2f$). The signal intensity at the first harmonic $I_{2f}$ is proportional only to the light intensity, whereas the intensity $I_f$ at the modulation frequency is proportional to both the light intensity and to the magnetization of the sample. Using a lock-in technique to measure both components allows one to normalize the two intensities. $I_f/I_{2f}$ is therefore proportional to only the sample magnetization. Figure 1.5 illustrates the light amplitude and intensity for two values of $\theta(H)$, i.e., two different magnetization states.

The modulation of the light can also be accomplished using an optical chopper or a photoelastic modulator. A photoelastic modulator allows modulation at significantly higher frequencies than with the Faraday modulator.
One method to obtain quantitative measures of the Kerr rotation is to use the nearly-crossed polarizer configuration together with a Faraday modulator magnetized by a feedback dc-circuit. The feedback loop minimizes the detector signal at all times by regulating the dc-current fed through the coil surrounding the Faraday modulator. Knowledge of the Verdet constant of the glass and the current through the coil gives the rotation of the polarization plane induced by the magnetization of the measured sample. In order to quantify the magnetization it is, however, necessary to also know the Kerr response for the investigated material at the wavelength used.

The simplest (but also the most rugged) measurement method of the Kerr effect is to use the nearly-crossed polarizer configuration without modulation to the signal. The signal in this setup is sensitive to changes of the light intensity but avoids any interference coming from, e.g., unwanted Faraday effects in the Faraday modulator from sources other than the surrounding coil.

In the above description only linear Kerr effects are considered. Higher order terms are, however, always present, which can give non-linear contributions to $M(H)$. Non-linear magneto-optical hysteresis loops have previously been reported in, e.g., Ni [36] and Fe [36, 37].

1.2.3 Other methods

Despite the versatility and ease-of-use of the Kerr effect, supplementary techniques are often necessary for a complete characterization of a magnetic sample. The Kerr effect is limited by, e.g., the light penetration depth and by the need to use reflecting samples. Also, since MOKE, on its own, yields only relative measures of magnetization/magnetic moments, calibrations using other techniques are necessary for quantitative measures.

**SQUID magnetometry**

Superconducting quantum interference devices (SQUIDs) are the most sensitive devices for measuring magnetic fields available [38]. The SQUID itself is a construction consisting of one or two Josephson junctions [39], integrated in a superconducting loop.

The most used application is the dc-SQUID configuration which uses two Josephson junctions in parallel. A varying magnetic field, e.g., that generated by moving a magnetic sample, generates a current in the detection coil. The current in the detection coil generates a magnetic field which is picked up by the superconducting loop that contains the SQUID. The output from the SQUID is an oscillating voltage with the period of one flux quantum, $\Phi_0$. The SQUID thus acts as a flux to voltage transducer, capable of detecting flux changes much smaller than $\Phi_0$.

A SQUID magnetometer gives a quantitative measure of the magnetic moment in a sample. However, due to the extreme sensitivity of the SQUID also “unwanted” magnetic moments from, e.g., sample substrates or dirt on a sam-
ple surface are detected. Therefore, if one uses SQUID measurements for cal-
ibrating the measurement of other, qualitative, methods such as MOKE, care
must be taken in order to not over- or underestimate the magnetic moment.

The SQUID technique and applications are described in several reviews and
books, see, e.g., Ref. [40].

**XMCD**

X-ray magnetic circular dichroism (XMCD) enables quantitative element
specific measurements of spin and orbital magnetic moments. The magnetic
dichroism is manifested through different excitation probabilities of low
lying electrons (L-edge) to the valence band for left- and right-hand circularly
polarized photons [41]. A magnetic sample shows a spin asymmetry in
the valence band and by using knowledge of the excitation probabilities
for spin-up and spin-down electrons and left- and right-hand circularly
polarized photons at the different absorption edges it is possible to map out
this asymmetry and thereby obtain information of the magnetic moment in
the sample. Since different elements require different excitation energies for
low level states it is possible to achieve element specificity by tuning the
excitation energy to the corresponding element. Six sum rules relate the
ground state properties with the X-ray absorption for different orbitals [41].

XMCD can be measured in transmission, where the absorption is measured
versus the incident photon energy [42]. It is also possible to measure the pho-
ton absorption indirectly via fluorescence or electron yield.

One drawback for the use of XMCD is the need for large synchrotron fa-
cilities for production of high intensity polarized light. For more detailed de-
scriptions of the XMCD technique see Refs. [41–43].
2. Magnetism and magnetic dimensionality

This chapter is intended to give a brief introduction to the concept of magnetism and magnetic dimensionality. A short summary of the Stoner theory of ferromagnetism is given, followed by a discussion of magnetic dimensionality and dimensionality crossovers.

Generally, dimensionality concerns the size and shape of an object and the limitations put on internal mechanisms by the size and shape of the object. A magnetic thin film can be considered spatially two-dimensional if the thickness does not permit thermal fluctuations in a direction perpendicular to the film plane. Similarly, a magnetic wire where magnetic fluctuations are possible in only one dimension would give a one-dimensional magnet. The internal mechanisms in the context of magnetism are the magnetic spins. Limitations to the spin dimensionality are determined by magnetic anisotropies.

2.1 Magnetism of 3d transition metals

The carriers of magnetism in the 3d transition metals are the 3d electrons. These electrons are very loosely bound to the atomic nuclei and can therefore be considered delocalized or itinerant.\(^1\) The electronic wave functions of the 3d conduction electrons overlap and form a common band structure which is split into two subbands containing, respectively, spin-up and spin-down electrons.

The conduction band energy can be expressed as \([44]\)

\[
E_B = \int_{E_1}^{E_2} E D_s(E) dE + \int_{E_1}^{E_2} E D_s(E) dE,
\]

where \(D_s(E)\) is the density of states (DOS), and \(E_1\) and \(E_2\) are the Fermi levels of the spin-up and spin-down subbands, respectively. A difference in the filling of the subbands will create a spin imbalance, resulting in a net magnetic moment.

The spin imbalance in the conduction band is related to the exchange interaction and Edmund C. Stoner showed that the exchange interaction energy

\(^1\)itinerant - Journeying; travelling from place to place: not fixed or stationary. (From the Oxford English Dictionary)
can be expressed as

\[ E_{\text{ex}} = -\frac{I}{4}(N_- - N_+)^2, \tag{2.2} \]

where both Coulomb and exchange interactions are contained in the Stoner parameter \( I \). Ferromagnetism in transition metals becomes favorable if the Stoner criterion

\[ D_s(E_F)I \geq 1, \tag{2.3} \]

is fulfilled. As seen in Eq. (2.3), a high DOS at the Fermi level is a requirement for ferromagnetism and only Fe, Co, and Ni fulfill the Stoner criterion due the high DOS at the Fermi level. For Fe \( D_s(E_F) \approx 1.5 \text{ eV}^{-1} \), and \( I \approx 1.1 \text{ eV} \) [44].

The Stoner criterion is nearly fulfilled for some other transition metals such as Sc and Pd. The DOS at the Fermi level for, e.g., Pd is lowered due to the large overlap of the 4d wave functions. In Paper V and Paper VI the high polarizability of Pd, which is a result of the strong Pauli susceptibility, is employed to allow a study of the magnetic dimensionality of induced magnetization.

Although the Stoner criterion correctly predicts the existence of itinerant ferromagnetism it is not useful in determining the magnitude of magnetic moments. To be able to predict the magnetic moments it is necessary to consider the population of the spin-split electron bands more carefully. Co and Ni are considered strong ferromagnets, as their respective majority carrier subbands are completely filled, whereas Fe, which has an incompletely filled majority carrier subband, is a weak ferromagnet. Figure 2.1 illustrates the two cases of strong and weak ferromagnets.
2.2 Magnetic anisotropy

Magnetic anisotropy describes the energies associated with different directions of the magnetization vector in a magnetic material. More specifically, it is energetically favorable for the magnetization to be oriented along certain directions determined by, e.g., the crystal symmetry or the shape of a magnetic specimen. The magnetic anisotropy determines, to large part, the shape of the magnetic hysteresis loop. Hence, the shapes of hysteresis loops in different directions in the magnetic material allows for a mapping of the magnetic anisotropy.

The magnetization vector can be expressed in terms of the angles $\theta$ and $\phi$ as

$$
m = m_s (\sin \theta \sin \phi \hat{x} + \sin \theta \cos \phi \hat{y} + \cos \theta \hat{z}),$$

where $m_s$ is the magnitude of the spontaneous magnetization and where the unit vectors $\hat{x}$, $\hat{y}$, and $\hat{z}$ are parallel to the principal crystal axes.

The magnetic anisotropy is normally treated as a combination of two parts, namely the shape anisotropy and the magnetocrystalline anisotropy. The shape anisotropy governs magnetization directions depending on the shape of magnetic particles. The spontaneous magnetization in the particle creates the demagnetizing field

$$
H_d = -Dm,
$$

where the demagnetizing factor, $0 \leq D \leq 1$, is determined by the shape. For a sphere $D = 1/3$ in all directions while $D \approx 1$ in the out-of-plane direction of a thin film. Thus the shape anisotropy favors in-plane magnetization in thin films. Shape anisotropy originates in a minimization of the magnetostatic energy. Although the shape anisotropy favors in-plane magnetization for thin films, other effects, e.g., crystal distortions in combination with the magnetocrystalline anisotropy can overcome the shape anisotropy and lead to, e.g., out-of-plane magnetization.

Like for the magnetic shape anisotropy, the magnetocrystalline anisotropy originates partly in magnetostatic dipolar interactions. However, the main contributions to the magnetocrystalline anisotropy comes from magnetoelastic effects. The magnetoelastic anisotropy involves electrostatic interactions from the crystal field and spin-orbit coupling.

For a crystal with cubic symmetry the magnetocrystalline anisotropy energy can be expressed using the angles $\theta$ and $\phi$ as:

$$
\frac{E_a}{V} = K_1 (\cos^2 \theta \sin^2 \theta \cos^2 \phi + \sin^4 \theta \cos^2 \phi \sin^2 \phi + 
+ \cos^2 \theta \sin^2 \theta \sin^2 \phi) + 
+ K_2 \cos^2 \theta \sin^4 \theta \cos^2 \phi \sin^2 \phi + \ldots ,
$$

where $V$ is the volume of the sample. It can be shown that if $|K_1| > |K_2|$, $K_1 > 0$ corresponds to an anisotropy which favors magnetization along the [001] principal axes, whereas $K_1 < 0$ corresponds to a favored alignment along the [111] direction.
2.3 Exchange couplings

The tendency of magnetic moments to align parallel (or antiparallel) to each other is electrostatic in origin and can be derived from a quantum mechanical theory of magnetism [5, 44]. This effect is determined by the overlap of electronic wave functions belonging to different (neighboring) atoms. When calculating the total energy of many electron systems this overlap is referred to as the exchange integral or exchange energy. When discussing not the energy but rather the interplay between two magnetic moments the term exchange coupling is often used. The exchange coupling is often denoted by \( J \) which enables one to express the exchange Hamiltonian (i.e., ignoring external fields etc.) as

\[
H = -\frac{1}{2} \sum_{i,j} J_{ij} S_i S_j, \tag{2.7}
\]

where \( J_{ij} \) is the exchange coupling between the spins \( S_i \) and \( S_j \). The sum is normally taken over all nearest neighbor pairs. The sign of \( J \) in Eq. (2.7) then specifies the type of order, a positive \( J \) favors parallel alignment of neighboring spins.

2.3.1 Interlayer exchange coupling

In artificially layered structures, composed of magnetic and non-magnetic metals, the magnetic/non-magnetic interfaces act as spin-polarized disturbances that polarize the conduction electrons of the non-magnetic layers according to the spin asymmetry at the Fermi level [45, 46]. This results in a coupling that is oscillating, both in sign and strength, as a function of the thickness of the spacer layer, but which is also sensitive to other changes of the Fermi surface. This coupling is the interlayer exchange coupling (IEC) which is normally denoted by \( J' \). The exchange interaction \( J \) is, in this context, normally referred to as the intralayer exchange coupling. For the transition metal heterostructures described in this work the ratio \( J'/J \) is typically \( 10^{-4} - 10^{-2} \). A schematic illustration of the different couplings are shown in Fig. 2.2.

Many theoretical approaches to explain the IEC have been investigated. The Ruderman-Kittel-Kasuya-Yosida (RKKY) [47–49] type coupling was derived for the coupling between magnetic impurities in a nonmagnetic metallic host and has been used to explain also the IEC.

Bruno has formulated a universal explanation in terms of reflection coefficients for the conduction electrons in the nonmagnetic (paramagnetic) spacer layer as these are reflected at the magnetic/non-magnetic interface [50]. The dependence of the IEC with the thickness of the spacer layer is for the transition metals given by a sum of terms

\[
J'(d) = \sum_i \frac{J'_i}{d^2} \sin(q_i d + \phi_i), \tag{2.8}
\]
Figure 2.2: Schematic illustration of the exchange couplings $J$ and $J'$ in a magnetic multilayer. Note that the non-magnetic spacer layers are not drawn but are represented by the spacing between the magnetic layers.
where $d$ is the thickness of the spacer layer, $q_i^\perp$ are critical spanning vectors which connect parallel surfaces of the Fermi levels of the magnetic layers, and $\phi_i$ describes phase differences.

Similar to the intralayer exchange coupling, the interlayer exchange coupling can lead to both parallel and antiparallel alignments depending on the sign of $J'$. A Hamiltonian similar to that of Eq. (2.7), but with the layer magnetization replacing the individual spins, can be expressed. Parallel alignment between neighboring magnetic layers is normally denoted ferromagnetic and vice versa. Note that the order in the individual layers is normally ferromagnetic irregardless of the alignment between the layers. Although $J$ and $J'$ are of different origin the essential mechanisms are the same and the ratio $J'/J$ largely governs the magnetic dimensionality of exchange coupled multilayers.

Figure 2.3 shows experimental values of the IEC between Fe layers in Fe/V heterostructures. The ferromagnetic couplings (positive $J'$) for V thickness $\sim 1 - 11$ ML (squares) are taken from Brillouin light scattering experiments by Vohl et al. [51]. The antiferromagnetic $J'$ between 11 and 15 ML of V (circles) are determined by the antiferromagnetic saturation field and are taken from Broddefalk et al. [52]. The last ferromagnetic points for V thicknesses 15–20 ML (triangles) are derived from first principle calculations by Skubic et al. [53]. In extrapolating the behavior of the IEC to 0 the value reached is significantly lower than the value for the intralayer exchange coupling for Fe, $J \approx 150 \text{ mJ/m}^2$. This shows the difference in magnitude between the interlayer- and intralayer couplings and also that the IEC should not simply be regarded as a weakened direct exchange coupling.
Also biquadratic interlayer couplings are possible which can lead to perpendicular alignments of layer magnetization [54]. Combinations of bilinear and biquadratic couplings can lead to magnetization rotations between layers which are different from 0 or 180°, depending on the relative ratio of the interlayer couplings involved.

2.4 Magnetic dimensionality

The concept of universality classes was introduced in Sec. 1.1.4. It is possible to classify the critical behavior of diverse systems depending on the spatial dimensionality (d) and on the spin dimensionality (n) for the system in question. The critical behaviors of systems belonging to the same universality class are identical and from a characterization of the critical behavior it is possible to determine both the spatial dimensionality and the spin dimensionality.

For magnetic systems, only spatial two- and three-dimensional (2D and 3D) systems are long-range ordered at temperatures above 0 K. For spatial 2D systems, two spin dimensionalities have long range ordered phases, namely the Ising and the XY. For spatial 3D systems, all three possible spin dimensionalities have magnetically ordered states above 0 K. Discrimination between the universality classes can be done by analysis of such observables as the specific heat, the magnetic susceptibility, or the spontaneous magnetization. In the experimental work presented here, conclusions about dimensionality are mainly based on the critical behavior of the spontaneous magnetization. As described in Sec. 1.1.1 the magnetization can, in the vicinity of the ordering temperature, be described by a power law;

\[ m \propto (-t)^\beta, \]

where \( t \) is the reduced temperature and \( \beta \) is the (critical) exponent of the magnetization. The theoretical values for \( \beta \) for the five dimensionality classes that show long range magnetic order are specified in Tab. 2.1.

The difference in \( \beta \) between 2D and 3D classes makes a determination of the spatial dimensionality possible and also makes it possible to determine the spin dimensionality (Ising or XY) for 2D classes. For 3D classes however, determining the spin dimensionality is generally more difficult since the differences between the expected values for \( \beta \) are relatively small.

In magnetic/non-magnetic heterostructures, the ratio of the exchange couplings \( (J'/J) \) governs the dimensionality to a large extent. In layered magnets, the length scale of 2D fluctuations is \( L_{2D} \leq \sqrt{J/J'} \), expressed in units of the lattice constant [18]. Fluctuations on length scales larger than \( L_{2D} \) are 3D. If the correlation length is smaller than \( L_{2D} \), all fluctuations are 2D and the magnetic behavior is spatially 2D.

\(^2\)The grouping is general but I will restrict the discussion to magnetic critical behavior and magnetic systems.
<table>
<thead>
<tr>
<th>Dimensionality class</th>
<th>$\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2D Ising</td>
<td>0.125</td>
</tr>
<tr>
<td>2D XY</td>
<td>$\approx 0.23$</td>
</tr>
<tr>
<td>3D Ising</td>
<td>$\approx 0.31$</td>
</tr>
<tr>
<td>3D XY</td>
<td>$\approx 0.33$</td>
</tr>
<tr>
<td>3D Heisenberg</td>
<td>$\approx 0.36$</td>
</tr>
</tbody>
</table>

Table 2.1: Values of the critical exponent $\beta$.

The upper limit on the correlation length imposed by the exchange coupling for 2D spatial ordering is a general condition in that there is no upper limit to the size of the system. It is of course possible to limit the correlation length in one or more directions by physically limiting the system. If one extension (e.g., thickness) is less than $L_{2D}$ then all correlations will be less than or equal to $L_{2D}$ and the magnetic behavior will therefore be spatially 2D.

For a thin film, also the spin dimensionality is affected by the film thickness. The shape anisotropy favors in-plane magnetization for thin films, which means that with decreasing film thickness a Heisenberg spin dimensionality will be reduced to an XY dimensionality.

2.4.1 The 2D-XY dimensionality class

The Mermin-Wagner theorem\(^3\) rigorously proves the absence of long range magnetic order in the 2D-XY dimensionality class for $T > 0$ K\([15–17]\). However, high temperature expansions of 2D models indicated critical behavior in the magnetic susceptibility\([55–57]\).

In the 2D-XY model the zero-field Hamiltonian is often written as

$$H = -J \sum_{i,j} S_i \cdot S_j = -J \sum_{i,j} \cos(\phi_i - \phi_j),$$

(2.10)

where $S_i$ and $S_j$ are normalized unit vectors, and $\phi_i - \phi_j$ is the angular difference between spins $i$ and $j$, summed over all nearest neighbors. For low temperatures, a spin wave analysis of Eq. (2.10) is adequate whilst with increasing temperature other excitations need to be considered. The small angular difference between neighboring spins can lead to the formation of so-called vortices, i.e., circular spin configurations about points in-between points in the lattice. Figure 2.4 displays several vortices with both clockwise and anticlockwise directions from Monte Carlo simulations at different temperatures, taken from Taroni\([58]\). Kosterlitz and Thouless\([59, 60]\) showed that it is possible to separate the angular distribution $\phi(r)$ of spins in the system in one part that

\(^3\)Sometimes also referred to as the Mermin-Wagner-Hohenberg theorem.
Figure 2.4: Spin configurations on a two-dimensional square lattice from Monte Carlo simulations at $T = 0.4, 0.5$ and $0.65$ in units of $J/k_B$. The figure is taken from Taroni [58].
Thermal population of spin wave modes confined within a quantum well of thickness $d_{Fe} = 11$ ML.

Figure 2.5: Thermal population of spin wave modes confined within a quantum well of thickness $d_{Fe} = 11$ ML.

2.4.2 Dimensionality crossovers

A bulk specimen of a magnetic material has a spatial 3D dimensionality where all three dimensions are equal with regards to magnetic fluctuations. If the thickness $d$ of the specimen is reduced in one dimension, excitations along this dimension will be hindered and as $d$ is below some critical thickness the spatial dimensionality will change from 3D to 2D [61, 62]. A magnetic film is thus spatially 2D if spin wave excitations out-of-plane are hindered [62, 63]. If two such 2D films are fabricated with a separating spacer layer in-between, the spatial dimensionality will be a function of the IEC through the spacer layer. The IEC is a function of the thickness of the spacer layer (see Eq. (2.8)), but it is also possible to modify the IEC through hydrogen loading (see Sec. 3.2). At low temperatures, the available thermal energy $k_B T$ is lower than the IEC and 3D fluctuations are hindered. With increasing temperature the increased thermal energy can overcome the coupling energy along the weakly coupled direction and give rise to 3D fluctuations. In Paper V and Paper VI the excitation of spin waves in the out-of-plane direction is proposed as an explanation for a crossover in the spatial magnetic dimensionality of Fe $\delta$-doped Pd with increasing thickness $d_{Fe}$ of the Fe $\delta$-layer. The population of
Figure 2.6: (From Paper V) Dimensionality crossover in Fe/Pd as reflected in the value of the critical exponent $\beta$. For Fe thicknesses $d_{\text{Fe}} \leq 0.4$ ML $\beta \approx 0.23$ and for $d_{\text{Fe}} \geq 1.0$ ML $\beta \approx 0.33$. For intermediate $d_{\text{Fe}}$ $\beta$ increases with increasing $d_{\text{Fe}}$.

Spin wave modes is described by a Planck distribution

\[
N_n = \left[ \exp \left( \frac{D \pi^2 n^2}{W^2 (a/2)^2 c k_B d_{\text{Fe}}} \right) - 1 \right]^{-1} \approx \left[ \exp \left( \frac{0.188n^2}{d_{\text{Fe}}} \right) - 1 \right]^{-1},
\]

where $D = 150 \text{ meV} \text{Å}^2$ is the calculated spin-stiffness, and $a = 3.89$ Å is the lattice constant of Pd. Figure 2.5 displays the population of some of the lowest magnon modes where the increase in available thermal energy with increasing temperature leads to the population of the modes. Figure 2.6 displays the values of the exponent $\beta$ versus the thickness of the $\delta$-doped samples from Paper V and Paper VI. $T_c$ increases linearly with $d_{\text{Fe}}$ as $T_c = 200d_{\text{Fe}}$ and the spatial dimensionality crossover, indicated by the change of $\beta$, from $\sim 0.23$ for $d_{\text{Fe}} \lesssim 0.4$ ML to $\sim 0.35$ for $d_{\text{Fe}} \gtrsim 1.0$ ML, is explained by the increased population of out-of-plane spin waves with increasing $d_{\text{Fe}}$. $T_c$ is an upper limit of the available thermal energy for spin waves. Above $T_c$ magnetic order is lost due to in-plane spin wave fluctuations which means that the population of out-of-plane modes for the thinner samples is not sufficient to yield spatial 3D magnetic behavior. With increasing $d_{\text{Fe}}$, i.e., increasing amount of Fe, $T_c$ is increased, hence the upper limit of available thermal energy is increased. As sufficient population of out-of-plane spin wave modes is accomplished, spatially 3D magnetic behavior is observed and the 2D to 3D dimensionality crossover is completed.
3. Analysis and interpretation of magnetic measurements

This chapter gives an overview of analysis of magnetization measurements and how it is possible to draw conclusions on dimensionality from such measurements.

The behavior of, e.g., the magnetization close to a phase transition can be approximated with power law behavior, as described in chapter 1. Real measurements, however, show finite-size effects that differ from the theoretical power law. In order to correctly interpret measurements it is therefore important to consider finite-size effects in the analysis.

Exchange coupled magnetic multilayers can show different magnetic critical behavior depending on the total number of magnetic layers. Also, thickness variations in both the magnetic layers and the spacer layers will add to the complexity in the analysis.

Defining the magnetic critical temperature is a non-trivial task due to finite-size broadening of the critical region. Often one is restricted to determining a mean value of $T_c$ together with a broadening (standard deviation) which is linked to the extent of the finite-size effects.

3.1 Magnetic measurements

All temperature dependent MOKE magnetization measurements presented in this work are based on hysteresis loops, i.e., magnetization versus applied field or $m(H)$. From the hysteresis loop it is possible to extract, e.g., the saturation magnetization, the coercive field strength, and the remanent spontaneous magnetization. By recording several hysteresis loops using different angles of the applied field it is possible to characterize the magnetic anisotropy, e.g., easy and hard axes, out-of-plane magnetization etc. Figure 3.1 shows two typical hysteresis loops, one ferromagnetic and one paramagnetic. It is worth pointing out that the two loops displayed in Fig. 3.1 represent only a subset of a large variation of hysteresis loops. Antiferromagnetic samples, e.g., multilayers with antiferromagnetic alignment between adjacent ferromagnetic layers display a linear dependence of the magnetization with the applied field. Out-of-plane easy axes display a reduction of magnetization proportional to $\cos \theta$, where $\theta$ is the angle between the easy axis and the sample surface normal. More complex anisotropies are described in detail in e.g., [64]. Magnetization
versus temperature can be constructed from hysteresis loops recorded at different temperatures by extracting the remanent magnetization from each loop. In a similar fashion magnetization in an applied field can be constructed by extracting the magnetization at the corresponding field from the hysteresis curves.

MOKE measures the magnetization in an area the size of the incident laser spot. This is typically of the order of 1 mm$^2$. Laser light penetration in metals decays exponentially with depth like $I = I_0 e^{-d/\kappa}$, where the penetration depth $\kappa$ for visible light is of the order of 100 Å. The response from magnetization in the interior of a sample is thus weighted with this decay. The magnetic signal is a summation of the magnetization within the probed volume. Therefore, any regions with a magnetization differing from the "mean" value will give rise to finite-size broadening of the critical region.

**Finite-size effects**
In the thermodynamic limit, i.e., in a system where the linear size $L \to \infty$, the order parameter vanishes completely at $T_c$ and response functions such as the magnetic susceptibility diverge. In any real system however, the susceptibility can not diverge due to the finiteness of the system. Likewise, the magnetization will not decay with infinite slope toward zero but will show a rounding that scales inversely with system size. The rounding of the magnetization versus temperature curve can be explained by regions of the sample having slightly different size, thickness, exchange coupling etc. which will give each small part a different $T_c$. This type of finite-size effects can be modeled by a distribution of $T_c$, as described in the next section.

Pronounced finite size effects are often encountered in numerical simulations, e.g., Monte Carlo simulations [65]. Figure 3.2 shows Monte Carlo
Monte Carlo simulations of a two-dimensional lattice with Ising-type spins for two different system sizes $N = L^2$. The difference in the finite size rounding is apparent. Note also that the asymptotic value of the magnetization scales inversely with $L$.

Finite-size effects in data from numerical simulations are typically more pronounced than those seen in measurements on real systems due to the difficulties involved with simulations of systems with a large linear size $L$. For a finite system there is always a non-zero probability for a jump between degenerate system configurations, e.g., from $+|M_s|$ to $-|M_s|$, especially in the transition region.

**Power law fitting**

The Mermin-Wagner theorem states that the 2D XY universality class does not have a magnetically ordered phase above 0 K. The Mermin-Wagner theorem holds for infinite systems. Any realizable 2D XY system, however, has ferromagnetic order above 0 K, as shown by, e.g., Bramwell and Holdsworth [18, 66]. They found that magnetization and $T_c$ are dependent on the size of the system. A large magnetic system displays a lower $T_c$ than a small system and consequently domains in a magnetic sample will have slightly varying $T_c$'s depending on their size [18]. Summation of the magnetic contribution from domains of different size will thus broaden the magnetic critical region and the, ideally sharp, decay of the magnetization at $T_c$ will be smeared out, resulting in a tail in magnetization versus temperature, c.f. Fig. 3.3. A similar effect, manifested in the same way, originates in thickness variations of magnetic layers and, in exchange coupled superlattices, also in thickness variation in non-magnetic spacer layers. These finite-size effects make determinations of such quantities as $T_c$ and critical exponents difficult.
A power law of the type

\[ m = m_0 (-t)^\beta, \]

will not fit with a broadened critical region (c.f. Fig. 3.3). One possibility is to exclude a part of the magnetization versus temperature data in order to have a better fit with the power law. It is, however, not trivial to determine where the tailing ends and which region to include in a fit. It has been argued by Kuz’mín and Tishin [67] that, due to impurities and inhomogeneities, a single critical point is not realizable in any real sample. The critical point in any real sample is rather a critical region of width \( \Delta T \). Kuz’mín and Tishin also propose that power law fitting within a reduced temperature region \( |\Delta T/T_c| \) of the critical temperature, either direct or through linearization, will increase the value of the extracted critical exponents.

The effects described by Kuz’mín and Tishin have been observed in the experiments reported in this work. Large finite-size tailing, corresponding to a large \( \Delta T \), obscures the interpretation of the exponent \( \beta \). A smaller \( \beta \) corresponds to a sharper decrease to zero at \( T_c \). A broadening of the region close to \( T_c \) will then appear as a larger \( \beta \) since the transition is smeared out. A correct determination of \( T_c \) is crucial for the construction of the reduced temperature \( t = (T - T_c)/T_c \) which is needed when comparing the critical behavior of samples with different \( T_c \). A correctly determined reduced temperature is also needed when extracting the exponent \( \beta \) from double logarithmic plotting (see below).

According to the central limit theorem, the sum of many random variables is approximately normally distributed. This can be used in the treatment of the finite-size tailing of magnetization versus temperature by assuming a Gaussian distribution of the critical temperature with standard deviation \( \sigma \) and expec-
tation value $T_c$ convoluted with the power law decay (Eq. (3.1)). Figure 3.3 displays magnetization versus temperature together with fitted curves with (dashed line) and without (full line) convolution with the Gaussian distribution. In Paper I magnetization versus temperature for several samples was fitted with both methods. Using the Gaussian distribution method a systematic shift of the extracted critical exponent $\beta$ toward the expected values for spatial 3D dimensionality was found.

The Gaussian distribution of $T_c$ is symmetric, i.e., finite-size tailing appears also below $T_c$. Thus, if the real distribution of domain sizes is Gaussian and the high temperature tail is correctly fitted the Gaussian distribution correctly describes the magnetization also for temperatures well below $T_c$. The Gaussian distribution is applied to the entire temperature range considered in the fitting, the impact is however largest close to $T_c$ where the rate of the magnetization decay is maximized.

Not all finite-size effects can be described by a Gaussian distribution of $T_c$. For example, in a system close to a percolation limit (2D or 3D percolation) there can be regions of different area or volume where some regions are magnetic and some are not. In this case a symmetric distribution such as the Gaussian is not describing the physics correctly.

For measurements showing very large finite-size effects one must ensure that the temperature range available for the fitting is sufficiently large since the convolution of the power law function and the Gaussian distribution will otherwise interfere with the lower temperature limit.

**Linear fitting methods**

Taking the logarithm of Eq. (3.1) gives a linear equation for the exponent $\beta$

$$\log(m) = \log(m_0) + \beta \log(-t),$$  \hspace{1cm} (3.2)

from which $\beta$ can be extracted as the linear slope. The reduced temperature $t$ is constructed using a value of $T_c$ determined independently from, e.g., direct fitting with a Gaussian distribution of $T_c$ as described above. Figure 3.4 displays the measurements of two samples belonging to different dimensionality classes, together with fits of the linear slope of the respective magnetizations. The data was fitted in the range $0.02 \leq -t \leq 0.2$, thereby not including the near-$T_c$ region. Close to $T_c$ ($-t \to 0$) the slope increases due to the finite-size tailing as the condition $|t| \geq |\Delta T/T_c|$ is not fulfilled.

Double logarithmic plotting of $m$ versus $t$ using $t(T_c)$ as a parameter has been used to extract both $\beta$ and $T_c$ by choosing the value of $T_c$ that maximizes the linear region [68]. Similar methods are using Arrott plots [69] of $m^2$ versus $H/m$, modified Arrott plots [70] of $m^{1/\beta}$ versus $(H/m)^{1/\gamma}$ or Kouvel-Fisher plots [71] of $m/(\partial m/\partial T)$ versus $T$. The Arrott- and modified Arrott plots yield $T_c$ as the value that linearize the respective curve whereas the Kouvel-Fisher plot yields $T_c$ as the intercept of the extrapolation of the linear region.
Figure 3.4: Double logarithmic plot of magnetization vs reduced temperature for two samples with different magnetic dimensionalities. The dashed vertical lines represent the fitting region, \(0.02 \leq (-t) \leq 0.2\).

with the line of \(T = 0\). In the latter case it is again important to consider the proximity to \(T_c\).

**Region of critical behavior**

The temperature region used in the fitting can influence the value of extracted parameters. The magnetization of 3D dimensionality, for example, will display Bloch \(T^{3/2}\) behavior [72] at low temperatures, whereas systems with 2D XY dimensionality are critical for all temperatures below \(T_c\) [73].

When comparing \(T_c\) and critical exponents from different samples it is therefore important to consider the region of temperatures used in the fit. For direct power law fitting as well as for the linear fitting methods, both a lower limit and an upper limit of the temperature must be chosen. For the direct fitting with the Gaussian distribution of \(T_c\) only a lower temperature limit is needed as the finite-size broadening is included in the model. This simplifies the fitting process somewhat.

3.1.1 Magnetic susceptibility

The magnetic susceptibility, defined as \(\chi \equiv (\frac{\partial m}{\partial H})_T\) (for practical use the definition; \(\chi = (\frac{\Delta m}{\Delta H}), H \rightarrow 0\) is better) can, in the vicinity of \(T_c\), be approximated with the diverging power law

\[
\chi \propto t^{-\gamma}.
\]  

(3.3)

Finite-size effects, however, do not permit infinite divergences in real measurements but the susceptibility shows a finite peak in the critical region. It is therefore not possible to fit real data to Eq. (3.3) at \(T_c\) \((t = 0)\). However,
the power law behavior of the susceptibility is valid in a large enough region around $T_c$ to enable a determination of the critical exponent $\gamma$.

The condition $H \to 0$ in the definition of the susceptibility, i.e., the requirement of a zero-field measurement, makes it, in a strict sense, impossible to measure the true susceptibility at $T_c$.

Several difficulties in connection with measuring and analyzing the magnetic susceptibility are discussed in \textit{Paper I}. The dependence on the applied magnetic field is closely related to the magnetic coercivity. Figure 3.5 shows the magnetic susceptibility of an Fe/V superlattice measured with different excitation field amplitudes. The left and right panels show the real (in-phase), and the imaginary (out-of-phase) components, respectively.

In Fig. 3.6 the peak positions for the real (circles) and imaginary (squares) parts of the susceptibilities of Fig. 3.5 are shown. Due to the finite field excitation there will be a contribution to the susceptibility signal from ferromagnetic switching. As the temperature is increased from below $T_c$ the coercive magnetic field will decrease, c.f. Fig 3.6. As the coercive field is comparable to the excitation field amplitude, the contributions from ferromagnetic switching will peak, which will give rise to the peak of the magnetic susceptibility. As a consequence, the susceptibility peak is shifted toward lower temperatures with increasing excitation field amplitude. Similar behavior has been observed by Elmers et al. in thin Fe films [74]. The straightforward method to minimize the effect on the susceptibility is to use as small applied magnetic field as possible. This will, however, reduce the signal-to-noise ratio.
3.2 Temporary loading with hydrogen

The magnetization and the magnetic order can be varied by the incorporation of hydrogen in the system. Temporary hydrogen loading of artificial magnetic heterostructures, e.g., superlattices and trilayers, leads to changes of the structural and the electronic properties of the host material. For artificial heterostructures deposited on rigid substrates with sufficient film-to-substrate adhesion the incorporation of hydrogen atoms in interstitial crystal sites leads to a one-dimensional expansion out of the plane of the substrate. The structural changes can be substantial, e.g., a $\sim 10\%$ volume increase of $V$ in an Fe/V superlattice was measured by Andersson et al. [75]. However, the changes to the magnetic behavior of artificial Fe/V heterostructures can not be sufficiently explained by this thickness increase but changes of the electronic properties, e.g., the hybridization between Fe and V, need to be considered.

3.2.1 Effects of hydrogen loading on magnetization

The interlayer exchange coupling (IEC) (see Sec. 2.3.1) in transition metal superlattices can be explained by RKKY interactions [46–49]. Temporary hydrogen loading provides a relatively simple way of altering the Fermi surface through the inherent crystal expansion caused by the introduction of hydrogen atoms into the structure. The largest effects on the IEC are, however, due to a modified electronic structure through hybridization of the host material and the hydrogen [76].

Following the discovery of oscillatory interlayer exchange coupling (OEC) in metallic multilayers [77, 78], considerable effort was focused on the explo-
ration and understanding of the OEC. Temporary hydrogen loading was found to switch the IEC from antiferromagnetic (AFM) to ferromagnetic (FM) in Fe/Nb multilayers [79–82], thereby providing a way of altering the oscillatory behavior of the IEC in these structures.

For Fe/V multilayers, switching in both directions, i.e., AFM to FM and vice versa has been demonstrated [83]. Changes of the IEC with hydrogen loading have also been seen in heterostructures of, e.g., Co/Nb [84] and Ho/Y [85–87].

Watts and Rodmacq found that while the bilinear IEC was highly sensitive to the hydrogen concentration, the biquadratic IEC was not significantly different between hydrogenated and non-hydrogenated samples of Co/Nb multilayers [84].

3.2.2 Practical hydrogen loading

Reversible hydrogen loading can only be accomplished within a limited concentration range, above which irreversible effects appear. If the adhesion to the sample substrate is strong enough to prevent cracking or bending [76, 88] the one-dimensional reduction of symmetry can for some thin film or thin film multilayers give a tetragonal crystal distortion that allows reversible hydrogen loading to higher concentrations than for bulk specimens.

A number of the non-magnetic transition metals absorb hydrogen exothermically, whereas the magnetic\textsuperscript{1} transition metals (Cr, Mn, Fe, Co, and Ni) only absorb hydrogen endothermically.

A surface layer of Pd increases the uptake rate of hydrogen, primarily by acting as a catalyst for hydrogen dissociation [89–91]. Furthermore, a Pd capping layer protects the underlying structure from oxidation [90, 91]. Samples intended for hydrogen loading experiments are typically capped by a 20–50 Å Pd layer. The addition of an Pd overlayer can, however, influence the sample magnetization due its strong paramagnetic susceptibility and high polarizability.

The hydrogen concentration can be monitored by measuring the sample resistivity during loading. Pressure-concentration and pressure-resistivity isotherms have been recorded for different combinations of Fe and V [92, 93]. Concentration-pressure relationships have been measured in Nb thin films [94, 95] and for W/Nb and Fe/Nb multilayers [96]. Changes of the magneto-optical properties of Fe/Pd bilayers upon hydrogen loading have also been detected [97] which, at least in principle, enables another method of monitoring the hydrogen concentration.

Typically, hydrogen loading is accomplished by exposing a sample to hydrogen gas at room temperature or higher. While an elevated temperature will give a higher absorption rate [81], it is usually not necessary as the timescale

\textsuperscript{1}Ferromagnetic and antiferromagnetic.
of hydrogen loading at room temperature is in the order of minutes for samples with a Pd capping layer. Below $\sim 250$ K, absorption-desorption is effectively hindered due to the formation of water on the sample surface. The hydrogen concentration is thereby effectively fixed in the sample. Desorption is a much slower process than absorption. This allows the use of measurement systems that do not facilitate hydrogen loading, e.g., the SQUID measurements at different hydrogen concentrations in Ref. [83] were conducted in this way.

In order to facilitate hydrogen loading for tuning the magnetic properties of M/NM heterostructures gas loading equipment was attached to the low temperature magnetic measurement system Homer (see Appendix A).

### 3.2.3 Hydrogen loading of Fe/V heterostructures

Reversible switching from AFM alignment to FM alignment of the magnetization of Fe layers in an Fe/V superlattice was demonstrated ten years ago by Hjörvarsson et al. [83]. The OEC of Fe/V multilayers has been thoroughly mapped out as a function of the thickness of the V spacer layer [83, 98–100] (see also Fig. 2.3). The IEC in Fe/V multilayers is FM for V thicknesses below $\sim 11$ ML as well as for V thicknesses above $\sim 15$ ML. For V layers with a thickness between 12 and 14 ML the IEC is negative, hence, the Fe layers are AFM coupled. Also, it is known that as an Fe/V sample is loaded with hydrogen, the hydrogen resides only in the V layers [92].

Thus, by only considering the effect of an expansion of the V layers, a FM coupled multilayer with a V thickness $\geq 15$ ML should stay FM upon hydrogen loading, while a FM coupled multilayer with a V thickness of 11 ML should enter into the AFM coupled region. It is found, however, that hydrogen loading ”shifts” the IEC versus V thickness curve (Fig. 2.3) to the right. This means that in FM coupled layers with V thicknesses $\lesssim 11$ ML the IEC will increase while for AFM coupled layers the IEC will be AFM for low concentrations of hydrogen and switch to FM for higher concentrations.

**Paper III** describes the effects on the IEC of hydrogen loading of a single Fe layer embedded between two V layers and on a double layer of Fe. In the single layer, loading of hydrogen to a maximum pressure of 4 mbar increases $T_c$ by $\sim 21$ K. In the double layer, the Fe layers are separated by a 14 ML V spacer which means that the IEC for the pristine/unloaded sample is FM. Loading the sample to increasing concentrations of hydrogen leads to an oscillating behavior of $T_c$. The oscillations in $T_c$ are linked to the changes of the IEC. For small hydrogen concentrations the IEC is FM and decreasing with increasing concentration. For intermediate concentrations the IEC will pass through the AFM region with increasing hydrogen concentration which results in decreasing ordering temperatures. For higher concentrations the IEC is increasingly FM with increasing concentration of hydrogen, leading to increasing values of $T_c$. 

42
4. Summary of papers

4.1 Magnetization and magnetic susceptibility of Fe\textsubscript{n}/V\textsubscript{7} superlattices (Paper I)

The temperature dependence of the magnetization and the magnetic susceptibility for a series of \([\text{Fe}\textsubscript{n}/\text{V}\textsubscript{7}]_{50}\) superlattices was investigated. \(n\), the thickness of the Fe layer, was increased in steps of 0.1 ML, from 1.7 to 2.6 ML. The idea behind the use of fractional thicknesses was to investigate the impact of deliberate fractional roughness on the magnetic dimensionality.

For the investigated Fe/V superlattices \(J'/J \approx 1/400\), as determined from the zero crossing of \(J'\) at \(\approx 10\) ML, and \(J'/J \approx 1/250\) for a V thickness of 5 ML. Previous studies of Fe\textsubscript{2}/V\textsubscript{5} superlattices had shown 2D spatial magnetic dimensionality in a region of reduced temperature from \(1 \times 10^{-4}\) to \(1 \times 10^{-1}\) [101].

Magnetization and magnetic susceptibility was measured in the "Homer" low temperature MOKE setup (see Appendix A). A close-to linear increase of \(T_c\) with increasing Fe layer thickness, was found, c.f. Fig. 4.1. The slope of \(T_c\) versus Fe layer thickness was found to be \(\approx 200\) K/ML. No correlation between Fe layer thickness and the critical exponent \(\beta\) was found. \(\beta\) was in the range 0.34–0.48 when extracted from a direct fit of the approximate power law to the data. Using a Gaussian distribution of the critical temperature yielded, overall, more consistent values of \(\beta\). However, the determined values of \(\beta\) indicated spatial 3D dimensionality for all investigated superlattices.

The analysis of the magnetic susceptibility indicated a relationship between the finite-size tailing of the magnetization and the full width at half maximum (FWHM) of the susceptibility peak. Furthermore, a close relation between the finite-size tailing of the magnetization and the onset of the imaginary (out-of-phase) part of the susceptibility was established. The onset of the imaginary part of the susceptibility has previously been argued to mark \(T_c\) as it indicates the onset of ferromagnetic switching in the susceptibility signal. However, the found relation between finite-size and the onset of the imaginary part of the susceptibility indicates that the susceptibility signal contains no good marker for \(T_c\).
4.2 Magnetic dimensionality of Fe/V (Papers II and III)

The effect on the magnetic dimensionality with changes of the interlayer exchange coupling (IEC) was studied in the Fe/V system. In Paper II the IEC was varied by the thickness of the V spacer layer in Fe/V superlattices. The investigated thickness range, 7–11 ML of V, covers a range from intermediate to very weak IEC. In Paper III temporary hydrogen loading of a sample consisting of a single Fe layer sandwiched between two V layers yielded an increase of \( T_c \). Hydrogen loading of a similar sample with two Fe layers, initially ferromagnetically coupled, showed an oscillating IEC with increasing hydrogen content.

The magnetic dimensionality of the superlattices studied in Paper II all displayed anomalous values of the exponent \( \beta \). For all samples \( \beta > 0.5 \), the value expected from a mean-field approach. The anomalous \( \beta \) values can be explained by surface effects, e.g., Binder et al. [102–105] have found high exponents for surface magnetization. For the Fe/V superlattices the effective IEC:s for a surface layer and for an interior layer are different.

\( T_c \) decreases with increasing V layer thickness and the change in \( T_c \) with varying IEC can be described by

\[
T_c = T_{c,0} + \frac{2\pi J}{\ln(\frac{J' D^2}{A_0 (D - X_0)})},
\]

where \( D \) is the thickness of the V spacer layer, \( X_0 \) is the thickness that gives \( J' = 0 \), \( T_{c,0} \) is the \( T_c \) of a single, non-interacting layer, and \( A_0 \) is a constant. The IEC \( (J') \) versus V spacer layer thickness is described by \( J' \approx A_0 \frac{D - X_0}{D} \) where \( X_0 = 11.4 \pm 0.6 \) ML.

Paper III describes an oscillatory IEC with varying hydrogen content. A single sample consisting of two 3 ML thick Fe layers, separated by 14.4 ML of
Figure 4.2: (From Paper III) $T_c$ vs the square root of the hydrogen loading pressure for two 3ML Fe layers separated by 14.4 ML of V. The inset shows the critical exponent $\beta$ vs the square root of the hydrogen loading pressure.

$V$ was exposed to an increasing amount of $H_2$ gas. The initial state of the sample was ferromagnetic and upon increasing hydrogen content the IEC initially decreases and becomes antiferromagnetic. A further increase of the hydrogen content yields a maximum in the antiferromagnetic IEC, followed by another decrease as the IEC moves into the ferromagnetic region again. Figure 4.2 shows $T_c$ and the critical exponent $\beta$ versus the square root of the hydrogen loading pressure. For small and intermediate hydrogen pressures the hydrogen concentration scales with the square root of the loading pressure. A comparison with measurements on a hydrogenated single layer of Fe reveals that the double layer has its strongest coupling in the pristine (unloaded) state. The oscillations in the IEC indicates the presence of quasi-2D regions.

4.3 Strain and interface effects in Fe/V superlattices (Paper IV)

A study of the effect of strain and interfaces on the magnetic orbital moment in Fe/V superlattices is described in Paper IV. Two series of Fe/V superlattices were fabricated, one with a varying Fe thickness, and one with varying V thickness. The idea behind the combination of these two series is that it is possible to vary the interface effects (interface density) through the thickness of the Fe layers and vary the strain by varying the V layer thickness.

The magnetic spin and orbital moments were extracted from XMCD measurements at the $L_2$ and $L_3$ absorption edges. The magnetic anisotropy energy difference between the [100] and [110] directions were extracted from MOKE measurements at a temperature of 80 K. The trends for the magnetic orbital
moment and the anisotropy energy with increasing Fe layer thickness were similar.

An increased interface density is expected to increase the orbital moment as the broken symmetry at an interface acts to reduce the quenching of the orbital moment. Similarly, an increased strain which gives a bcc to bct transformation is expected to increase the orbital magnetic moment due to a reduction of the symmetry. However, a reduction of the orbital magnetic moment with decreasing Fe layer thickness, i.e., increasing interface density, is found for the investigated samples. Hybridization between Fe and V at the interfaces lowers the Fe spin magnetic moment. Through the correlation between the spin and orbital moments, the hybridization dominates over the reduction of the symmetry, causing a decrease of the orbital magnetic moment with increasing interface density.

A model containing the effects of strain (in terms of the ratio $c/a$) and the Fe/V interface density (in terms of $1/t_{Fe}$, where $t_{Fe}$ is the Fe layer thickness) on the magnetic orbital moment was proposed.

$$M_{orb} = M_B + \frac{\Delta M_c}{a} + \frac{2\Delta M_{int}}{t_{Fe}}, \quad (4.2)$$

where $M_B$ is the bulk contribution to the orbital moment, $\Delta M_c/a$ is the contribution from strain, and $\Delta M_{int}$ is the contribution from the interfaces. Fitting the data to Eq. (4.2) gave $M_{orb} \approx 0.14(2) \mu_B/\text{atom}$, in reasonable agreement with the bulk value $0.09 \mu_B/\text{atom}$. The proposed model captures the effects seen in the experiment, a negative contribution from the interfaces and a positive contribution from the strain.

4.4 Dimensionality of induced Pd magnetization (Papers V and VI)

Paper V and Paper VI describe investigations on the effect of confinement on magnetic order. Both papers are based on studies of Fe $\delta$-doped Pd layers, where a very thin ($\approx 1$ ML) Fe layer is sandwiched between two much thicker Pd layers. The high polarizability of Pd leads to a total magnetic thickness of $\approx 10$ ML. The lengthscale of the magnetic polarization in Pd is larger than that of atomic defects, which therefore are effectively masked at some distance from the magnetizing Fe layer. Figure 4.3 displays an artistic view of this phenomenon for three different Fe/V configurations. In Paper V the thickness of the Fe $\delta$-layer is varied in the range $0.2 \leq t_{Fe} \leq 1.6$ and the magnetic spatial dimensionality displays a 2D–3D crossover in the range $0.4 \leq t_{Fe} \leq 1.0$ ML. The dimensionality crossover is marked by an increase in the exponent $\beta$, as deduced from direct fitting and double logarithmic plotting. The 2D–3D dimensionality crossover is accompanied by a reduction of the critical region.

46
Figure 4.3: (From Paper VI) Illustration of three different Fe/V configurations and the respective polarization patterns. a) A surface layer, b) an alloy, and c) a $\delta$-layer. Filled and empty circles represent Fe and Pd atoms, respectively. The polarizations are calculated using periodic boundary conditions and an exponential decay.
This can be seen in Fig. 4.4 as a deviation from linearity as $t_{Fe}$ increases. Also, $T_c$ increases close to linearly with the thickness of the Fe layer.

In Paper V the dimensionality crossover is explained by a quantum well model for the perpendicular spin wave excitations. The increasing $T_c$ with increasing $t_{Fe}$ leads to a larger population of out-of-plane spin waves. Eventually, the larger population leads to 3D dimensionality.

In Paper VI the findings of Paper V are compared with measurements on Fe/V alloys, containing the same amount of Fe as some of the $\delta$-doped samples but homogeneously distributed in a 20 ML thick alloy. Two alloy samples were fabricated, containing the equivalent of 0.45 and 0.75 ML Fe, respectively. While the 0.75 ML alloy displayed 3D magnetic behavior ($\beta \approx 0.31$) and a lower $T_c$ than its $\delta$-doped equivalent, the 0.45 ML sample was not magnetic. This shows the importance of the induced magnetization in the Pd in defining the magnetic properties. Moreover it confirms the strong relation between confinement of the magnetic "species" and the magnetic dimensionality.

Comparing $T_c$ versus the amount of Fe in the $\delta$-doped samples and Fe/V alloys gives yet another confirmation of the importance of extension on order. Also, it enables an estimate of the range of Fe-Fe interaction to $\approx 10$ ML, consistent with previous results.
Inverkan av lagerväxelverkan på magnetisk ordning i Fe-baserade heterostrukturer

De magnetiska material som undersökts i denna avhandling består av metaller arrangerade i skikt vars tjocklek endast är några atomära monolager. Enkla tunnfilmer, bestående av exempelvis ett ferromagnetiskt skikt med skikt av en icke-magnetiska metall på vardera sida har undersökts. Även supergitter, en struktur där två eller fler skikt alterneras ett flertal gånger, har studerats. Genom att variera tjockleken på de magnetiska skikten kan man påverka magnetisering och magnetisk anisotropi. Vidare är de magnetiska skikten magnetiskt kopplade till varandra via utbytesväxelverkan. Denna växelverkan kan ändras genom att variera tjockleken på de icke-magnetiska skikten.


Ett magnetiskt materials dimensionalitet bestäms av dess rumsliga utsträckning samt av den interna dimensionaliteten, ofta benämnd spinndimensionalitet. Spinndimensionaliten bestämmer tillåtna riktningar för materialets magnetisering. En mycket tunn film av ett magnetiskt material, exempelvis järn,
har andra magnetiska egenskaper än vad järn har i bulkform\textsuperscript{1}. En tunn film är en tvådimensionell struktur då dess utsträckning är begränsad i en riktning. Genom att på samma sätt begränsa utsträckningen i ytterligare en riktning kan man skapa en endimensionell tråd. Även spinndimensionaliteten påverkas av begränsningar av den rumsliga utsträckningen. Exempelvis gör den magnetiska formanisotropin hos en mycket tunn film att magnetiseringen begränsas till riktningar parallella med ytan, istället för att som tidigare kunna ha en i princip godtycklig riktning. De tre möjliga rumsliga dimensionerna (noll-

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig5_1.png}
\caption{Illustration av de möjliga rumsliga dimensionaliteterna.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig5_2.png}
\caption{Illustration av de möjliga spinndimensionaliteterna.}
\end{figure}

dimensionella punkter undantagna), se Fig. 5.1, tillsammans med tre möjliga spinndimensionaliteter, se Fig. 5.2, bildar tillsammans nio möjliga dimensionalitetsklasser. Fem av dessa klasser representerar kombinationer som är ferromagnetiska vid temperaturer över absoluta nollpunkten. Beteendet hos magnetiska observabler, exempelvis temperaturberoendet hos magnetisering och magnetisk susceptibility, kan beskrivas med så kallade kritiska exponenter vilka är unika för varje dimensionalitetsklass. Genom att experimentellt

\textsuperscript{1}bulkegenskaper - de egenskaper hos materien i en kropp som är relaterade till kroppens hela volym. De är i motsats till ytegenskaper inte beroende av ytans tillstånd. - Från Nationalencyklopedin
bestämma dessa exponenter kan man därför föra varje magnetiskt prov till en dimensionalitetsklasse.

De resultat som redovisas i denna avhandling är primärt baserade på mätningar av den magnetooptiska Kerr-effekten (MOKE). MOKE beskriver en polarisationsförändring hos ljus som reflekteras från en magnetisk yta. Polarisationsförändringen är proportionell mot magnetiseringen vilket medför att denna kan bestämmas som en funktion av exempelvis externt magnetfält och temperatur. Mätningar av det magnetiska temperaturberoendet har genomförts där det studerade materialet placerats i en kryostat2 med optiska öppningar för att kunna mäta MOKE. Temperaturberoendet hos magnetiseringen har sedan använts för att bestämma den magnetiska dimensionaliteten hos materialet.

Genom att variera utbytesväxelverkan via tjockleken hos ickemagnetiska skikt och via vätgaskoncentrationen samt genom att variera tjockleken hos magnetiska skikt kan dimensionaliteten varieras. Genom att studera hur dimensionaliteten förändras när man varierar en av dessa parametrar kan man dra slutsatser om utsträckningen av exempelvis magnetiska fluktuationer och styrkan av utbytesväxelverkan mellan magnetiska skikt.

---

2kryostat - köldaggregat konstruerat för att hålla mycket låga temperaturer konstanta. - Från Nationalencyklopedin
Acknowledgments

I want to thank the people at Fysik III in Uppsala for all help I have received during my years as a Ph. D. student. So thanks to Gabriella, Moreno, Panos, Andreas, Gunnar, Hossein, Wang, Petra, Lennart, Bengt, Tore, Ola, Roger, and Erik. Special thanks to Matts, good luck in Switzerland. I have also to thank some of the former members of the group Maarten, Emil, Stefan, Anna-Maria, Rimas, and Inna. Extra very special thanks to Till, Cyril and Claudia.

My supervisor Björgvin, thanks for this time. You have taught me so many things, not just about physics.

Thanks to the Ångström workshop and especially Pierre for all help. Also, Erland thanks for all help with cryogenics and gases.

Some other people I have worked with and wish to thank are Andrea, Gregor, Arndt, Erik, Jonas, and Nick.
A. "Homer" laboratory measurement system

A.1 Overview

The laboratory MOKE-measurement system "Homer" is shown schematically in Fig. A.1. The "Homer" system is equipped with a pair of Helmholtz coils with a radius of 100 mm. The absence of remanence in Helmholtz coils is a requirement for measurements of the magnetic susceptibility where the applied field amplitude is typically below 10 $\mu$T. Also, when measuring the magnetization, the absence of stray fields can greatly simplify both the measurement and the subsequent analysis. For this reason, the entire system is shielded by a triple $\mu$-metal shield to eliminate the influence of the earth’s magnetic field as well as any other external fields. The axial stray field inside the shielding is below 1 $\mu$T and by applying a compensating dc-offset in the driving current to the coils the effective stray field can be reduced even further.

The low inductance of Helmholtz coils, compared to that of ordinary electromagnets makes ac-measurements of magnetization (hysteresis) and susceptibility possible, even for frequencies above 200 Hz. Ac-fields of up to 10 mT amplitude can be generated. The setup is thus best suited for samples with low coercivity or measurements very close to $T_c$ where the coercivity normally is very small.

For susceptibility measurements, the sample response to the ac-driving field is phase-sensitively recorded at the frequency of the driving field which then acts as signal modulation. Sinusoidal fields with amplitudes ranging from 2 $\mu$T can be applied at an arbitrary frequency. Typically, fields of 5–10 $\mu$T amplitude at frequencies $\approx 200$ Hz are used. The in-phase and the out-of-phase components are recorded to obtain both the real and the imaginary part of the susceptibility. The imaginary part of the susceptibility is a measure of hysteretic losses in the sample and thus acts as an indication of ferromagnetism.

Temperature dependent measurements of the magnetization are possible by placing the sample in an optical cryostat. The cryostat is cooled by the evaporation of liquid helium or nitrogen for temperatures down to $\sim 5$ and $\sim 77$ K respectively. Elevated temperatures up to 450 K are possible using a resistive heater. The system is fitted with two temperature sensors. One RhFe-sensor positioned close to the heater where feedback from this sensor is used to control the temperature through the flow of cryogenics and through the heater.
Figure A.1: Overview of the MOKE-measurement system "Homer”. Details are given in the text.

current. The actual sample temperature is measured by a Cernox resistive temperature sensor placed very close to the sample.

In order to reach liquid helium temperatures, adequate heat insulation is necessary. This is achieved by maintaining a vacuum in the space surrounding the cryostat sample holder and the cryogenics transfer tube. Figure A.2 is a schematic overview of the cryogenic- and vacuum parts of the "Homer” system. The system is pumped by a Pfeiffer TMU260 turbo-molecular pump backed by an Edwards roughing pump. The base pressure of the system is \( \lesssim 10^{-7} \) mbar which is sufficient for thermal insulation and also permits hydrogen loading.

A.1.1 Water deposition

The deposition of an optically transparent medium on a surface will give rise to interference of light with a well-defined wavelength as it is reflected from the surface. For a measurement technique sensitive to the reflection of light from a surface this is a potential problem. It is, however, possible to continuously monitor the reflected light intensity so as to maintain a normalization reference for, e.g., reflectivity measurements. In MOKE this is, unfortunately, not a useful approach as the rotation of polarization, i.e., the measure of magnetization, occurs in reflection. Multiple reflections will thus corrupt the response in a way which is not directly linked to the intensity variations of the interference.
Figure A.2: Overview of the cryogenic and vacuum parts of "Homer". The system is pumped by a Pfeiffer TMU 260 turbo-molecular pump (TMP), backed by an Edwards fore-vacuum pump. The valves are: (1) Valve for bypassing the TMP for rough pumping high pressures (2) Valve to the TMP, closed when rough pumping or hydrogen loading (3) Valve for letting hydrogen gas into the sample space (4) Valve for letting hydrogen gas into the system (5) Valve for venting the system with nitrogen gas.

Removal of water from a vacuum system is effectively achieved through cryopumping, i.e., condensing water onto a cold surface. This is in fact one of the most efficient ways of pumping water vapor. Figure A.3 shows the partial pressures of some gases present in the "Homer" system. In cooling down the system, mainly water condenses on the cold interior. The condensing of water in this way is a relatively slow process. As seen in Fig. A.3 the partial pressure of water is virtually constant as the temperature is lowered. The anomalies seen in the data on decreasing temperature are artefacts coming from turning on (marked by the right arrow) and turning off (marked by the left arrow) of a cold cathode pressure gauge attached to the system. Note that the partial pressures of water and $N_2$ are almost identical as the cryostat is cooled. Maintaining the cryostat at liquid nitrogen temperature for several hours reduces the water pressure in the system to a level lower than that of $N_2$, as can be seen in the right panel of Fig. A.3. Upon heating, the partial pressure of water is constant up to a temperature of $\sim 170$ K where it starts to rise. This marks the release of water due to the locally increased temperature in the immediate surrounding of the resistive heater. As the temperature is further increased it is clear that the total pressure inside the vacuum compartment is almost completely due to water.

It is possible to overcome the problem of water condensation. Firstly measurements should, as far as possible, be conducted during cooling instead of
Figure A.3: Partial pressures of various gases inside the cryostat sample space vs temperature in cooling (left panel) and heating (right panel). The anomalies seen in the cooling data are related to the switching on (right arrow) and switching off (left arrow) of a cold cathode pressure gauge.

during heating as the partial pressure of water is practically constant during cooling. Secondly, the cryostat should be kept at high temperature (room temperature) for some time between measurements to allow the cryo-trapped water to evaporate. Venting the system with N₂ gas instead of air also helps to keep water from entering the sample space. Proper vacuum baking [106] will also reduce the water content inside the system.

A.1.2 Hydrogen loading facility

"Homer" is constructed to allow hydrogen loading of samples. Hydrogen loading is usually done at 300 K by letting hydrogen gas into the space surrounding the sample. For hydrogen loading at low pressures it is possible to first fill a smaller volume with a higher pressure of hydrogen and then let the gas fill the total space in order to have better control of the pressure. The uptake of hydrogen is monitored by the four-point resistance of the sample, measured by contacts on the sample surface. When the hydrogen content in the sample is equilibrated the sample is cooled and the hydrogen atmosphere is removed by evacuating the sample space. This is done when the temperature is in the range 240–270 K. The hydrogen concentration in the sample is then constant as long as the temperature does not exceed ∼ 240 K. It is also possible to measure a hydrogen loaded sample at higher temperatures by keeping a hydrogen atmosphere inside the sample space. In this case, however, the temperature should not be lowered below ∼ 270 K with hydrogen (or any gas) inside the sample space as ice will form on the outside surface of the cryostat and block the optical access.
A.2 Data collection and treatment

The main advantage of ac-measurements of the magnetization is that a large number of \( m(H) \) curves can be collected in a relatively short time. In the normal measurement mode sinusoidal fields with typical amplitudes between 1 and 5 mT are applied at a frequency in the range 5–10 Hz, resulting in 50–100 datasets within 10 seconds. By averaging these datasets, low-noise hysteresis loops are obtained, which in turn can be used to illustrate the behavior of the magnetization as a function of the sample temperature. The measure of the magnetization can be in remanence or at any field value in the range of the applied field amplitude. Figure A.4 shows an overview of the signals and electronics of the "Homer" system. Communication between the measurement computer and external units, such as the temperature control unit and multimeters for measuring the four-point resistance and temperature, is going either through the IEEE GPIB-bus or through a National Instruments PCI-6052E I/O card. The applied magnetic field is generated by sending a
current through the Helmholtz coils. This current is generated by amplifying a sine-wave signal generated in the I/O card. As the signal is generated in software it is easy to include a dc-bias to further remove stray fields, as mentioned above. The magnetic field is measured by a hall sensor placed close to the sample position, measuring the field along the center axis of the Helmholtz coils. The hall sensor output is amplified and fed to the I/O card. The magnetic signal of the sample is measured in the nearly-crossed polarizer configuration, the current generated in the detecting photo diode is converted to a voltage, amplified, and fed to the I/O card. The signals from the hall sensor and the detector are sampled simultaneously using a sampling frequency of $\sim 50$ kHz during a time specified by the user, normally 10–20 seconds.

In software, the zero-crossings of the magnetic field signal are detected and the signal is divided into several parts, each containing one full period. The number of parts is thus the frequency times the measurement time. Likewise, the detector signal is split into an equal number of parts at the zero-crossings of the applied field. The parts of each signal are then averaged to reduce the noise, and plotting the detector signal versus the applied field gives $m(H)$ of the sample. Since no filtering is applied to either of the two detected signals, there is a negligible phase difference between them. Subsequent filtering of the remaining high-frequency noise can be accomplished in software.

The beam reflected from the sample is split in two beams. One is led to the detector via the analyzer and the other is detected directly and fed to the I/O card as a measure of the reflected intensity. In the subsequent data treatment this measure can be used to normalize the magnetic response to the intensity of each curve to eliminate any variations in, e.g., the laser output intensity. In software, also other types of signal treatment can be accomplished, such as normalization of both field and magnetic response or fitting to give, e.g., the coercivity as a function of temperature.

The Oxford Instruments ITC 503 temperature controller can be programmed to sweep a temperature interval at a given rate. This function is used to measure the temperature dependence of the magnetization. By measuring $m(H)$ during a fixed time interval, typically 20 seconds, and relating the result to the mean temperature during the time interval, the temperature dependence of the magnetization is mapped out.
Bibliography


62


[53] B. Skubic. Private communication.


Acta Universitatis Upsaliensis

Digital Comprehensive Summaries of Uppsala Dissertations from the Faculty of Science and Technology 307

Editor: The Dean of the Faculty of Science and Technology

A doctoral dissertation from the Faculty of Science and Technology, Uppsala University, is usually a summary of a number of papers. A few copies of the complete dissertation are kept at major Swedish research libraries, while the summary alone is distributed internationally through the series Digital Comprehensive Summaries of Uppsala Dissertations from the Faculty of Science and Technology. (Prior to January, 2005, the series was published under the title “Comprehensive Summaries of Uppsala Dissertations from the Faculty of Science and Technology”.)