Critical Phenomena and Exchange Coupling in Magnetic Heterostructures

MARTINA AHLBERG
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Abstract


The continuous phase transition in thin magnetic films and superlattices has been studied using the magneto-optical Kerr effect (MOKE) and polarized neutron scattering (PNS). It has been shown that the critical behavior of amorphous thin films belonging to the 2D XY universality class can be described within the same theory as crystalline sample. This means that quenched disorder only serves as a marginal perturbation in systems with this symmetry.

The connection between interlayer exchange coupling and the observed critical behavior in Fe/V superlattices was explored. The results prove that the origin of unusually high values of the exponent β can be traced to a position dependence of the magnetization at elevated temperatures. The magnetization of the outermost layers within the superlattice shows a more pronounced decrease at lower temperatures, compared to the inner layers, which in turn have a more abrupt decrease in the vicinity of the critical temperature. This translates to a high exponent, especially when the layers are probed by a technique where more weight is given to the layers close to the surface, e.g., MOKE. The interlayer exchange coupling as a function of spacer thickness and temperature was also studied in its own right. The data was compared to the literature, and a dependence on the thickness of the magnetic layers was concluded.

The phase transition in amorphous FeZr/CoZr multilayers, where the magnetization emanates from ferromagnetic proximity effects, was investigated. Even though the determined exponents of the zero-field magnetization, the susceptibility and the critical isotherm did not correspond to any universality class, scaling plots displayed an excellent data collapse.

Samples consisting of Fe δ-layers (0.3-1.4 monolayers) embedded in Pd were studied using element-specific resonant x-ray magnetic scattering. The magnetization of the two constituents showed distinctly different temperature dependences.

Keywords: phase transitions, critical behavior, 2D XY model, MOKE, magnetic thin films and superlattices, amorphous multilayers, magnetic proximity effects

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Inspiration is for amateurs.
-Jan Guillou

Till mamma och pappa
List of papers

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

I  **Influence of boundaries on magnetic ordering in Fe/V superlattices**

II  **Two-dimensional XY-like amorphous Co$_{68}$Fe$_{24}$Zr$_8$/Al$_{70}$Zr$_{30}$ multilayers**

III  **Experimental realization of amorphous two-dimensional XY magnets**

IV  **Temperature dependence of magnetic properties in weakly exchange coupled Fe/V superlattices**
    Martina Ahlberg, Evangelos Th Papaioannou, Gregor Nowak and Björgvin Hjörvarsson, *submitted to Phys. Rev. B*

V  **The effect of ferromagnetic proximity on critical behavior**
    Martina Ahlberg, Panagiotis Korelis, Gabriella Andersson, and Björgvin Hjörvarsson, *submitted to Phys. Rev. B*

VI  **Element Specific Magnetization in Fe/Pd Quantum Well Structures**
    Thomas P. A. Hase, Matthew S. Brewer, Unnar Arnalds, Martina Ahlberg, Vassilios Kapaklis, Evangelos Th. Papaioannou, Matts Björck, Laurence Bouchenoire, Paul Thompson, Daniel Haskel, Jonathan Lang, Yongseong Choi, Cecilia Sanchez-Hanke, and Björgvin Hjörvarsson, *In manuscript*

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My contributions to the papers in this thesis

I  Responsible for the MOKE measurements and participated in the data acquisition at ILL. Performed most of the analysis. Shared responsibility of writing the manuscript.

II  Made the samples. Carried out all measurements and analysis. Main responsible of writing the manuscript.

III  Participated in the analysis and contributed to the manuscript.

IV  Performed the low field MOKE measurements and made all analysis. Main responsible of writing the manuscript.

V  Initiated the paper and performed all analysis. Main responsible of writing the manuscript.

VI  Made some of the samples. Participated in the XRMS measurements. Contributed to the manuscript.
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1. Introduction

We have all come across phase transitions when we boil water to cook, melt metals for making tin soldiers or dry our laundry. These are all examples of so called first-order phase transitions, which are associated with the release or uptake of latent heat and often with a change in the structure of the material.

The research presented in this thesis is about second-order or continuous phase transitions and they are scarce in our everyday life. It is nevertheless possible to give a down-to-earth illustration of this type. A common bar magnet attracts needles, but if it is heated the attractive force will be reduced and some of the needles will fall off the magnet. The higher the temperature, the more needles will fall off and eventually the magnet has lost all ability to hold on to the needles. This will happen at a very specific temperature that is called the critical temperature ($T_c$), or alternatively, in the case of ferromagnets, the Curie temperature.

![Magnetization vs Temperature Graph](image)

*Figure 1.1.* When a magnet is heated to temperatures above the critical temperature ($T_c$) it loses its net magnetization and therefore the ability to attract drawing pins. This is an example of a continuous phase transition. Close to $T_c$ the decrease in the magnetization follows a simple equation: $m \propto (1 - T/T_c)^\beta$.

Nothing happens with the atomic structure of the material, but the magnetic properties are clearly different. Those changes can be described with simple equations where the magnetic quantity is proportional to the distance from $T_c$ to the power of some exponent: e.g. $m \propto (1 - T/T_c)^\beta$, where $m$ is the magnetization and $T$ is the temperature.
In a simple picture, where the atoms only interact with their nearest neighbors, all materials can be divided into six universality classes where the categorization only depends on the spatial dimensionality and the symmetry of the order parameter. In a magnetic context the latter means how many directions the spin can point in, but the classes are by no means restricted to magnetic phase transitions and that is why they are called universal. For example the same picture applies to the gas-liquid coexistent curve in fluids. There are three symmetries and they are known by the name within the parentheses: the spins can only point up or down (Ising), the spins are free to rotate within a plane (XY) and the spins can point in any direction (Heisenberg). The bar magnet mentioned above is classified as a 3D Heisenberg system. But we are interested in what happens when the physical extension is confined and the system can be described as two-dimensional. To explore this, samples are manufactured in the form of thin films with thicknesses of only a few atomic layers. Another intriguing topic is the critical behavior in amorphous materials, which differ from the theoretical models and crystalline materials in the sense that the atoms are not ordered in a well defined lattice. Work presented in this thesis shows that the magnetic phase transition in amorphous thin films can be described within the same framework as crystalline films. In addition, the ambiguous limit between 2D and 3D is explored.

Figure 1.2. A schematic picture of a multilayer consisting of two materials. Note that the proportion between the thickness and extension of the sample is misleading. If the figure were to scale, a 1 mm film thickness would correspond to a sample side of roughly 10 km.
Thin films of different materials can be combined to form a multilayer or a superlattice. This is illustrated in Fig. 1.2. When magnetic and non-magnetic materials are alternated in this fashion it can give rise to a mechanism called interlayer exchange coupling (IEC). Via this coupling two magnetic layers can interact through the non-magnetic spacer and the relative direction of their magnetizations depends on the thickness of the spacer layer. Typically the alignment of the magnetizations will be parallel or anti-parallel, but it might also be perpendicular. The impact of IEC on the observed critical behavior is a long-time unanswered (and to certain extent unasked) question that is addressed in this thesis.

So why do we care about continuous phase transitions when there are so many "real" problems troubling the world? The defensive answer is that they are coupled to many other topics such as the distributions of earthquakes, turbulence in fluids and the financial market. In addition the research on two-dimensional magnets gives significant contributions to the vast area of thin films which covers everything from tool making to data handling. But in fact the answer is probably more connected to the inherent motto of humanity: Do not leave any stone unturned! This is the common factor of mountain climbing and basic research. If the mountain is there, climb it. If the phenomenon is there, understand it.

\[^{1}\text{A multilayer is any layered structure consisting of at least two materials. The notation superlattice implies that the atomic structure is preserved throughout the sample, which means the sample can be described as a single crystal even though it is built up of different materials.}\]
2. Magnetic phase transitions - single layer

2.1 Say hello to infinity - Phase transitions in the thermodynamic limit

A magnetic phase diagram is easy to draw, which is exemplified in Fig. 2.1, but the basic mechanisms behind the behavior were mysterious for a long time. The critical temperature \( T_c \) can be defined in many ways, but the easiest phrasing is that it corresponds to the temperature where the zero-field magnetization vanishes. The phase transition is also accompanied by a diverging magnetic susceptibility \( \chi \), which is infinitely large at \( T_c \). Both theoretical and experimental work has proven that, close to the critical temperature, the magnetic quantities follow simple power laws of the form \( a = |t|^b \), where \( t \equiv T/T_c - 1 \) is the reduced temperature and \( b \) represents any critical exponent. A number of critical exponents, together with their definitions are found in Table 2.1. The physical quantities are described in later sections.

An expression often encountered in the phase transition community is "the thermodynamic limit", the meaning of which might be hard to comprehend. The essence is however not complicated, it means that the system size is infinitely large. The term comes from the fact that the energy of a system fluctuates, but as the system size goes to infinity these instabilities becomes negligible (except at the critical temperature) [1].

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Definition</th>
<th>Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific heat</td>
<td>( C_H \propto</td>
<td>t</td>
</tr>
<tr>
<td>Magnetization</td>
<td>( m \propto (-t)^\beta )</td>
<td>( h = 0, T &lt; T_c, T \to T_c )</td>
</tr>
<tr>
<td>Susceptibility</td>
<td>( \chi \propto</td>
<td>t</td>
</tr>
<tr>
<td>Critical isotherm</td>
<td>( m \propto</td>
<td>h</td>
</tr>
<tr>
<td>Connected two-point</td>
<td>( G_c^2(r) \propto 1/r^{d-2+\eta} )</td>
<td>( h = 0, r \text{ large}, T = T_c )</td>
</tr>
<tr>
<td>correlation function</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Correlation length</td>
<td>( \xi \propto</td>
<td>r</td>
</tr>
</tbody>
</table>

Table 2.1. Definition of the critical exponents, from Binney et. al [1]. The reduced temperature is defined as \( t \equiv T/T_c - 1 \). The physical quantities are described in later sections.

An interesting hypothesis states that the exponents only depend on the spatial and spin dimensionalities of the material. This is called universality and gives a correct picture in many cases, but fails to capture what happens when interactions are long-ranged and when the spatial dimension is ambiguous. A
Figure 2.1. (a) Magnetic phase diagram. Following the dashed arrows gives the response showed in (b) and (c), respectively. (b) The solid line shows the zero-field magnetization as function of temperature. The dash dotted line is a sketch of the magnetization when \( h \neq 0 \). (c) Magnetic isotherms at three different temperatures, above, below and exactly at \( T_c \).

single layer, i.e. a single thin film of a magnetic material, can be considered as a three-dimensional (3D) or a two-dimensional (2D) system, depending on its thickness. The threshold between 3D and 2D depends on the material and the atomic structure, but films typically have to be thinner than \( \approx 10-15 \, \text{Å} \) to display two-dimensional critical behavior [2, 3]. The transition from bulk-like to 2D is not sharp, but for a certain thickness range thin films exhibit exponents that can not be attributed to any universality class.

2.1.1 The contributions of Dr Landau - The Mean Field (MF) model

The earliest theory of magnetism was formulated by Weiss in 1907 [4] and is based on the assumption that the problem of many interacting spins can be simplified by only considering one spin sitting in an average or mean field produced by all the other spins. Within this theory it is (usually) also assumed that the spins only interact with their nearest neighbors. One of the main results of this approach is that the critical temperature only depends on the interaction between the spins \( (J) \) and the coordination number \( (z) \) of the lattice, i.e. the number of nearest neighbors to one spin. This gives the equation:
\[ T_c = \frac{zJ}{k_B} \]  

(2.1)

where \( k_B \) is the Boltzmann constant. This result underestimates the critical temperature, but gives a useful rule of thumb suggesting that \( T_c \) scales with the strength of the average interactions in a system.

Using Landau theory \([5, 6]\) it is possible to calculate the critical exponents. It is based on the assumption that the free energy \( (F) \) of a system is small and analytical close to the critical temperature. This allows a Taylor expansion of \( F \), giving

\[
F(m, T) = \alpha_0(T) + \alpha_2(T)m^2 + \alpha_4(T)m^4 + hm
\]

(2.2)

where \( \alpha_i \) are undetermined coefficients which vary with temperature and \( h \) is an external magnetic field. All odd coefficients must vanish since \( F \) does not depend on the sign of \( m \) in zero field. \( \alpha_0 \) merely sets the zero energy and is of little importance. In Fig. 2.2 the free energy is plotted as a function of \( m \) for different values of \( \alpha_2 \) and \( \alpha_4 \). It is clear that \( \alpha_4 \) must be greater than zero, otherwise \( F \) can be minimized by letting \( m \) go to infinity which would represent an unnatural scenario. When \( \alpha_2 < 0 \) two minima emerge, corresponding to the two possible configurations of \( m \) in the ordered phase, and thus \( \alpha_2 = 0 \) marks the critical temperature.

![Fig. 2.2. The free energy plotted as a function of temperature for different \( \alpha_2 \) and \( \alpha_4 \) in Eq (2.2).](image)

In the vicinity of the critical temperature, \( \alpha_4 \) is considered to be constant and the temperature dependence of \( \alpha_2 \) is captured by the relation:

\[
\alpha_2 = t \alpha_2^0
\]

(2.3)
where $t = T/T_c - 1$ and $\alpha_2^0$ is a constant. This makes it possible to evaluate the temperature dependence of $m$ in zero field by minimizing the free energy,

$$\frac{\partial F}{\partial m} = 2t\alpha_2^0 m + 4\alpha_4 m^3 = 0 \quad (2.4)$$

and finding the two solutions:

$$m = \begin{cases} 
\pm \sqrt{\frac{2|t|\alpha_2^0}{4\alpha_4}} & t < 0 \\
0 & t > 0 
\end{cases} \quad (2.5)$$

which allows us to identify $m \propto |t|^{1/2}$ and therefore $\beta = 1/2$.

It is also easy to calculate $\delta$ of the critical isotherm at $t = 0$, since it follows that $h + 4\alpha_4 m^3 = 0$, giving $m \propto h^{1/3}$ and $\delta = 3$. The other exponents are also possible to evaluate with this approach. However, the results of the calculations agree very badly with experimental findings. The reason is that the Landau theory fails to acknowledge fluctuations and correlations in the zero-field magnetization close to the critical temperature.

### 2.1.2 Fluctuations gone wild - Statistical mechanics

The continuous phase transition in magnetic materials is often referred to as the order-disorder transition. A common misunderstanding is that all magnetic ordering disappears above the critical temperature, but spins are correlated in blocks also at higher temperatures. The difference is that below $T_c$ one can follow a path of ordered spins throughout the whole system, while above $T_c$ the ordered blocks are much smaller. Statistical mechanics provides the tools needed to understand the concepts of fluctuations and correlations that play a crucial role in the vicinity of the critical point and are the reason why mean-field theories fail to quantitatively describe phase transitions. The fluctuations are not only something turning up in equations and simulations, but can be observed by the naked eye in some binary fluid mixtures, like methanol and n-hexane [1]. In these fluids the solubility is a function of temperature and at high temperatures the fluid forms a single phase, but at low temperatures the two solutions are immiscible and separated into two phases. In-between there is a critical temperature separating the two conditions. When this temperature is approached, density fluctuations scatter the light and the initially transparent liquid becomes opaque. This phenomenon is called critical opalescence and is also observed e.g. in carbon dioxide around the critical point where gas and liquid become indistinguishable [7]. Similar fluctuations arise in the magnetization in magnetic materials and with the right tools they are observable. Neutron scattering allows probing of the so called two-point correlation function ($G^2$) [1]. It is a measure of the degree of correlation between different parts of the system and is defined by:
\[ G^2(r) \equiv \langle \vec{s}_i \cdot \vec{s}_j \rangle \]  

where the brackets denote the thermal average and \( r = |\vec{r}_i - \vec{r}_j| \). This definition is valid when the system is isotropic in all directions and applies to discrete lattices only when \( r \) is large. Below \( T_c \) the spins show long-range order and \( G^2 \) will be large for all \( r \). Above \( T_c \) only spins at relatively short distances are correlated and \( G^2(r) \) goes to zero for large site separations. But if an external field is applied to the magnetic material the spins tend to align and the correlation function increases. In addition, \( G^2 \) does not capture the interesting fluctuations. This lead to the introduction of the connected two-point correlation function:

\[ G^2_c(r) \equiv \langle \vec{s}_i \cdot \vec{s}_j \rangle - \langle \vec{s}_i \rangle \langle \vec{s}_j \rangle \]  

Above the critical temperature, in zero field, \( G^2 \) and \( G^2_c \) are equal since the thermal average of the spins is zero. The major motivation of introducing \( G^2_c \) is that it only counts the deviation from the general alignment, \( i.e. \) the fluctuations in the magnetization. Thus it goes to zero as the temperature approaches zero and the alignment of the spins increases.

It has been shown that \( G^2_c \) follows a power law exactly at the critical temperature:

\[ G^2_c(r) \propto 1/r^{d-2+\eta} \]  

where \( d \) is the spatial dimensionality of the system, and \( \eta \) is the associated critical exponent. Eq. 2.8 is not valid if the temperature is close to, but not equal to, \( T_c \). Instead \( G^2_c \) is exponentially decaying according to:

\[ G^2_c(r) \propto \exp(-r/\xi) \]  

where \( \xi \) is the correlation length. This is an important parameter that gives a measure of the threshold size of fluctuating blocks with correlated spins. Correlated fluctuations larger than \( \xi \) are exponentially rare. The correlation length also follows a power law, see Table 2.1, and is infinitely large at the critical temperature. This implies that the range of interactions is infinite at \( T_c \) and a single spin flip can influence all the other spins, which is the reason why the susceptibility diverges; the smallest perturbation of the system will cause a huge response.

Computer simulations are a valuable tool to get insight about what happens with the magnetic ordering at different temperatures. The simplest model with a finite \( T_c \) is a 2D Ising system and since the spins only can point in two directions they are easily visualized in black and white. Fig. 2.3 shows how the size of spin blocks evolves from temperatures close to zero (top left), passing through the critical temperature (middle right), up to high temperatures (lower right). Thermal fluctuations are strong enough to flip some spins even at low
temperatures and as the temperature increases the white regions grow. At $T_c$ the black and white regions are equally large. In addition, one can find correlated blocks of all sizes, i.e. there are fluctuations on all length scales. At slightly higher temperatures there are still blocks with aligned spins of significant radii, but as the temperature rises a salt-and-pepper effect emerges. The correlation length can be viewed as the size of a typical white region.
Figure 2.3. Illustration of typical spin block sizes at different temperatures in a 2D Ising model; from Ref. [1]. The top left figure represents a temperature close to zero and the middle right corresponds to the critical temperature. See text for details. Reprint made with permission from the publisher.
2.1.3 Same same, but different - Universality and scaling

Universality

The observation of fluctuations on all length scales, mentioned in the last section, forms the foundation of the concept of universality. The idea is that the correlated behavior of largely separated spins washes out the details of the local interactions and leaves the critical behavior to be governed only by the spin and spatial dimensionalities of the system. In this simple scheme there are six universality classes and they are illustrated in Fig. 2.4. While all systems follow the same basic relations (see Table 2.1), the set of critical exponents differs from one class to another and serves as a characteristic for the model.

It can be shown that there is no phase transition when the spatial dimensionality is restricted to 1D and that the only model which in fact shows magnetic long-range order in two dimensions is the Ising symmetry. However, there is a loophole in this argument, which will be discussed in Section 2.2.2. The 2D Ising model is also the only model that has been solved exactly. This famous accomplishment by Lars Onsager [8] is one of the cornerstones in modern theory of phase transitions. The other models have to be addressed using approximate methods such as Monte Carlo simulations or renormalization group theory. Table 2.2 lists the values of different critical exponents for the most important universality classes. A closer look reveals an important aspect for experimentalists; while the exponents differ considerably from 2D to 3D, most of them are quite similar for three-dimensional Ising and Heisenberg systems. This might make it hard to distinguish different spin symmetries and sometimes one has to settle with the fact that the sample can be classified as 3D.

The general applicability of the different models is supported by the fact that two such widely different systems as a monolayer of iron [11] and the structural ordering on a gold surface [12] show the same critical exponents as expected in the 2D Ising framework. Moreover, a vast variety of substances show the same behavior along the liquid-gas coexistence curve as the magnetic transition of MnF$_2$, and they all belong to the same universality class as the three-dimensional Ising model. Examples of 3D Heisenberg systems are the ferromagnets La$_{0.67}$Sr$_{0.33}$CoO$_3$ [13] and bulk amorphous FeCoZr [14]. The
Table 2.2. Values of the critical exponents for different models.

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>α</td>
<td>0</td>
<td>0</td>
<td>0.1106</td>
<td>-0.1336</td>
</tr>
<tr>
<td>β</td>
<td>1/2</td>
<td>1/8</td>
<td>0.3264</td>
<td>0.3689</td>
</tr>
<tr>
<td>γ</td>
<td>1</td>
<td>7/4</td>
<td>1.2365</td>
<td>1.3960</td>
</tr>
<tr>
<td>δ</td>
<td>3</td>
<td>15</td>
<td>4.7883</td>
<td>4.783</td>
</tr>
<tr>
<td>η</td>
<td>0</td>
<td>1/4</td>
<td>0.0366</td>
<td>0.0375</td>
</tr>
<tr>
<td>ν</td>
<td>1/2</td>
<td>1</td>
<td>0.6298</td>
<td>0.7112</td>
</tr>
</tbody>
</table>

latter is a striking example of how the critical behavior is unaffected by local properties; here the symmetry of the lattice.

However, there are grey shades to the rigid six class categorization of different materials. The models are limited to nearest neighbor (nn) interactions. Fisher et al. [15] showed already in 1972, using a renormalization group approach, that the exponents vary with the range of the interactions. The decay of the interactions is described as $r^{-(d+\sigma)}$, where $d$ is the spatial dimension of the system ($\sigma > 0$). There are three different regimes: if $\sigma > 2$ the exponents take the same value as in a nearest neighbor model and if $2\sigma - d < 0$ the exponents will be the same as in a classical mean-field model. But when $d/2 < \sigma < 2$ the exponents vary continuously with the value of $\sigma$, which has been observed in for example Fe-Pt alloys [16] and CdCr$_2$Se$_4$ [17]. Also the magnetic archetypes Fe and Ni seem to fit into this picture [18], even though they show "pure" Ising behavior as thin films [11, 19, 20].

Furthermore, it has also been demonstrated that the temperature dependence of the magnetic profile within a magnetic layer is heavily affected by the range of interactions [21]. This has implications on the measured value of $\beta$ and will lead to a higher value than expected from nn models.

Even when it is acknowledged that the simple universality table has to be restricted to materials/models with nearest neighbor (or at least short ranged) interactions there is another hazy shade to consider: the crossover from 2D to 3D is not discontinuous. Measurements of the critical exponent $\beta$ in thin Ni films, as a function of film thickness, show that it increases continuously from the expected 2D value to a 3D-like value [2]. However, the dimensionality crossover is not only governed by the thickness, but depends on excitations of spin waves. A study of $\delta$-doped Pd(Fe) layers [22] demonstrated a clear difference in the effective dimensionality at low and high temperatures. Both a sample with relatively low Fe content (0.5 ML) and another sample with more Fe (1.4 ML) could be classified as 2D at low temperatures. Nevertheless, analysis of the critical behavior revealed that $\beta$ of the Fe rich sample had a significantly higher value compared to the other sample, implying a change of dimensionality. The reason for the discrepancy lies in the critical temperature; a thin film will show 3D behavior if $T_c$ is high enough to allow thermal exci-
tations of spin waves in the out-of-plane direction. A similar line of argument is found in Ref. [3].

The importance of spin wave excitations is also addressed in Paper V, where the phase transition in amorphous FeZr(x)/CoZr(1 Å) multilayers was studied. The conclusions state that the magnetization in the sample with x=25 Å must be continuous with a magnetic thickness corresponding to the physical extension of 250 Å. However, the magnetization is expected to be modulated along the z-direction (out-of-plane), peaking at the CoZr layers and this give rise to a magnetic dispersion that depends on the angle from the in-plane direction. The resulting critical behavior is not 3D, as would be expected if the sample only consisted of one material, but the exponents reveal a non-universal behavior in-between 2D and 3D.

### Scaling

While the concept of universality predicts a set of exponents determined solely from the spatial and spin dimensionalities, the scaling hypothesis predicts a number of equalities between the exponents. A straightforward derivation of those scaling laws is found in Ref. [23]. The relationships are listed in Table 2.3, and an important implication is that one only needs to determine two exponents to be able to calculate all the others, if the dimensionality is known. This simplifies comparisons between experimental results on similar materials, but focused on different magnetic properties and thus different exponents.

| \( \gamma = \beta (\delta - 1) \) |
| \( 2\beta + \gamma = 2 - \alpha \) |
| \( \nu d = 2 - \alpha \) |
| \( \nu (2 - \eta) = \gamma \) |

**Table 2.3.** Equalities between the critical exponents [23, 24].

Another statement within the scaling hypothesis is that if the magnetization \( (M(t,H)) \) and the applied field \( (H) \) are scaled properly, all data close to the critical temperature will collapse on a single curve. This technique and the relations above are widely used to confirm the validity of experimentally determined exponents [25, 26, 27, 28, 29, 30]. In contrast to the universality classes which rely on \( nn \) interactions, good data collapses have been observed for materials that can not be classified within the classical models [30, 31].
The scaled magnetization is defined as \( m' \equiv M/|t|^{\beta} \) and the scaled field as \( h' \equiv H/|t|^{\beta+\gamma} \) \[7\]. There is not one unique way of graphically presenting the properties \[23, 30, 32\] and both the temperature range and the chosen axes will affect the perceived quality of the collapse \[33\]. High field data is emphasized if \((m')^2\) is plotted versus \(h'/m'\) and the low field region is highlighted if \(\ln(m')\) is displayed as a function of \(\ln(h')\). In Paper V we explore the critical behavior of amorphous FeZr/CoZr multilayers and even though the exponent \(\beta\) takes values expected in a dimensional crossover, the data show an excellent collapse, see Fig. 2.5 and 2.6. Also \(\delta\) and \(\gamma\) were determined in the paper and the results are found in Table 2.4. Clearly the exponents do not match the expected value for any universality class.

<table>
<thead>
<tr>
<th>Sample</th>
<th>(\beta)</th>
<th>(\delta)</th>
<th>(\gamma)</th>
<th>(\gamma = \beta(\delta - 1))</th>
</tr>
</thead>
<tbody>
<tr>
<td>25-1</td>
<td>0.35(1)</td>
<td>6.10(31)</td>
<td>1.74(2)</td>
<td>1.79(12)</td>
</tr>
<tr>
<td>50-1</td>
<td>0.31(1)</td>
<td>6.34(29)</td>
<td>1.71(2)</td>
<td>1.63(10)</td>
</tr>
<tr>
<td>75-1</td>
<td>0.26(1)</td>
<td>8.54(13)</td>
<td>2.02(1)</td>
<td>1.99(10)</td>
</tr>
</tbody>
</table>

Table 2.4. Critical exponents of FeZr(x Å)/CoZr(1 Å) multilayers, where \(x=25, 50\), and 75 Å. \(\gamma\) was determined from both the susceptibility and the Widom equality; from Paper V.
2.1.4 Size matters! - Finite size effects

The discussion above has mostly covered the condition where samples can be seen as infinitely large. This applies to many experimental situations, especially when bulk materials are studied. However, in the area of thin films this restriction is often not fulfilled and finite size effects emerge. The most striking consequence of limiting the physical extensions is seen in the critical temperature; merely decreasing the thickness can shift $T_c$ from several hundred °C to tens of Kelvin or suppress the magnetic order completely. Also the reduction of the critical temperature can be parameterized with a power law:

$$T_c(l) = T_c^\infty (1 - bl^{-\lambda})$$  \hspace{1cm} (2.10)

where $T_c^\infty$ is the bulk critical temperature, $b$ is a constant and $l$ is the film thickness [34]. The suppression of $T_c$ is connected with a cut-off in the correlation length and $\lambda$ is equal to $1/\nu$. The power law is only valid for relatively thick films, with a lower limit of about 5 monolayers. In the thin thickness range the decrease in $T_c$ is linear [35, 36, 37]. Zhang and Willis [38] used a mean-field model to link the reduction of $T_c$ in the linear region to the average spin-spin interaction range ($N_0$) and showed that the model gives a good approximation that fits experimental data. In Paper II a similar approach was used giving an estimate of $N_0 \approx 8.1$ Å in amorphous CoFeZr.
The magnetic observables will also change as a result of finite size effects, since the phase transition is smeared over some temperature range. The susceptibility of bulk materials is assumed to be infinite at the critical temperature. This will naturally never be observed in an experiment, although the signal can be huge. A limited size not only reduces the maximum, but also shifts it to a lower temperature and this situation is schematically illustrated in Fig. 2.7. The magnetization is also influenced by finite sizes and it shows a rounding instead of an abrupt decrease at $T_c$, which makes it hard to determine the true critical temperature in the analysis of data.

![Figure 2.7](image_url)

**Figure 2.7.** Schematic illustration of (a) the shift of the maximum in the susceptibility and (b) the rounding of the magnetization, caused by finite size effects. (c) The tail in the magnetization above $T_c$ in FeZr($x$)/CoZr(1 Å) multilayers, with $x=25$, 50 and 75 Å. None of the samples belong to any universality class, instead their exponents reveal a dimensionality crossover were 25-1 verges on the 3D limit and 75-1 approaches 2D behavior. The larger system size of 25-1 is reflected in the smaller rounding of the magnetization.

Although samples that can be described as infinitely large are relatively easy to manufacture experimentally, at least in the 3D limit, theoretical methods using computer simulations have to handle the fact that the samples always will be finite. Hence there is a need for ways of extrapolating theoretical
results on finite lattices to the thermodynamic limit. The technique to do this is called finite size scaling and is similar to the scaling approach discussed in the last section. Using the magnetization as an example leads to the relation:

\[ L^{\beta/\nu}M(|\tau|, L) = \left( L^{1/\nu} |\tau| \right)^{\beta} f \left( L^{1/\nu} |\tau| \right) \]  

(2.11)

where \( L \) is the total lattice length, \( f \) is some function and \( \tau = (T - T^\infty_c) / T^\infty_c \) [34]. For the appropriate values of the exponent and the critical temperature, the data of different \( L \) should collapse on two branches, one for \( T < T^\infty_c \) and one for \( T > T^\infty_c \) in a graphic presentation. Even though this method is most common in theoretical studies, where \( L \) is exactly known, it has also been used in experimental work [18].

2.2 On a visit to the world of stick figures - The 2D XY model

It is well known in the scientific community that no magnetic phase transition can occur in two-dimensional materials with XY symmetry, since any thermal excitation will cause such heavy perturbations on the system that all ordering is destroyed. This was rigorously proven by Mermin and Wagner in 1966 [39]. However, tell me the truth that lasts, seven years later Kosterlitz and Thouless did calculations on the subject and showed that even though no net magnetization will occur, ordering develops in the form of spin vortices and antivortices [40]. This did not, however, explain the numerous findings of the exponent \( \beta \) taking values of \( \approx 0.23 \) in layered magnetic structures. Bramwell and Holdsworth finally explained this, and attributed it to a 2D XY phase transition in finite sized samples [41, 42, 43]. In this class the thermodynamic limit is also pushed to extreme scales; while an iron cube with a side of one centimeter can be classified as being infinite (after all it contains about \( 10^{24} \) atoms), the lateral extension of a magnetic 2D XY film has to be greater than the size of Texas to be large enough to suppress the net magnetization.

2.2.1 Back to infinity - The Kosterlitz-Thouless model

The divergence of the correlation length (\( \xi \)) and the susceptibility (\( \chi \)) in the Kosterlitz-Thouless (KT) model does not follow a power law behavior, unlike in the conventional phase transitions. Instead the divergence is faster and shows an exponential increase [44],

\[ \chi(T) \propto \xi^{2-\eta} \propto \exp \left[ \frac{(2-\eta)b}{(T/T_{KT}-1)^{1/2}} \right] = \exp \left( \frac{B}{\epsilon^{1/2}} \right) \]  

(2.12)
where $T_{KT}$ is the critical temperature, denoted the Kosterlitz-Thouless temperature. Above this temperature the vortex and antivortex pairs unbind. The exponent $b$ is often cited as equal to 1.5, due to an error in the original paper. The correct number is 2.2, but the value is non-universal and depends on the system [45]. The absence of a power law also directly implies that the exponents $\gamma$ and $\nu$ are not defined and thus the scaling relations discussed in Section 2.1.3 are not applicable. It is important to note that the susceptibility does not go to zero below $T_{KT}$, but stays infinite and KT systems are characterized by being critical at all temperatures below the phase transition.

There are two critical exponents which take universal values also in this model. At $T_{KT}$, $\delta = 15$ and $\eta = 1/4$, the same values as in the 2D Ising model. The KT model has been successful in describing the superfluid transition in He films [46, 47], but the model needs some modifications, incorporating finite-size effects, to fit the behavior of 2D XY magnets.

![Figure 2.8. The type of topological order found below $T_{KT}$, a vortex-antivortex pair. Above the Kosterlitz-Thouless temperature the pairs unbind and single vortices are found; from Ref. [48].](image)

### 2.2.2 Infinity is an illusion - The finite size 2D XY model

The observation of a net magnetization in thin films with continuous spin symmetry originates in the fact that the system size is far from the thermodynamic limit. Therefore magnetic phase transitions in 2D XY systems always display a significant smearing of the transition, which complicates the experimental determination of the critical temperature. In addition, the critical behavior of finite sized 2D XY systems is characterized by not one, but three significant temperatures: the temperature $T_c$ where $\xi$ is equal to the system size, the temperature $T_{KT}$ where vortex pairs unbind, and the shifted KT temperature, $T^*$. They are related through the expression

$$T_c - T_{KT} = 4(T^* - T_{KT})$$

(2.13)
The temperature dependence of the correlation length and susceptibility follows Eq. (2.12) as for infinitely large samples, whereas the point where $\delta$ and $\eta$ take their universal values of 15 and $1/4$, respectively, is shifted to the slightly higher temperature $T^*$. Furthermore, the exponent associated with the magnetization, $\beta$, also takes a universal value at this temperature and equals 0.23. When averaged over a larger temperature range the value of $\beta$ depends on the system size, but in practice the experimental findings typically yield values in the close neighborhood of the universal 0.23 [42, 48].

The 2D XY model can also be broadened to take the presence of anisotropy into account. It has been shown that the value of $\beta$ depends on the strength of the four-fold crystal field ($h_4$) compared to the interactions between the spins ($J$) [49]. The exponent demonstrates a bimodal distribution bounded by 0.23 for weak fields and an Ising value of 0.125 in the strong field limit, see Fig. 2.9.

![Figure 2.9. Distribution of the critical exponent $\beta$ in two-dimensional materials. The histogram is the result of an extensive literature search by Taroni [49].](image)

Two-dimensional magnetic systems are not limited to thin films. Materials like Rb$_2$CrCl$_4$ and K$_2$CuF$_4$ have a strong coupling between lattice sites within a plane and a very weak coupling between those layers giving a quasi 2D behavior. Rb$_2$CrCl$_4$ was used to explore the predictions of the finite sized 2D XY model in a comprehensive paper by Als-Nielsen et al. [50]. In the vicinity of the critical temperature the material shows three-dimensional behavior, but at both higher and lower temperatures the experimental results are consistent with the theoretical expectations of a two-dimensional phase transition.

The temperature dependence of the magnetization has been investigated in thin films made of a diverse range of materials [49]. An example of a system
showing 2D XY behavior is Fe embedded in V [51]. This might be surprising since Fe on W was the first observation of a magnetic phase transition belonging to the 2D Ising class [11], which further illustrates the importance of the interplay between the magnetic element and the surrounding material in the two-dimensional limit. An alternative and significant impact on the magnetic anisotropy, and thus the critical behavior, is that of the crystallographic directions. Elmers et al. observed that 1 monolayer (ML) of iron on W(110) can be described as 2D Ising [52], while 2 ML of the same material on W(100) show the characteristics of the 2D XY model [53]. Many studies focus solely on the exponent $\beta$ and overlook the critical behavior of other quantities, such as the field dependence at $T_c$ or the susceptibility. This is in particular true for 2D XY systems although there are exceptions, for example Ref. [53] where it was found that the temperature dependence of $\chi$ followed the expected KT behavior.

In Paper II we studied the critical behavior in amorphous CoFeZr/AlZr multilayers. The magneto-optical Kerr effect (MOKE) was utilized to measure the magnetization and the ac susceptibility of samples with five different CoFeZr thicknesses: $d=11, 12, 13, 14$ and $15$ Å. The critical temperature of the thinnest sample was below 15 K and the thickest sample was still ferromagnetic at 300 K, therefore the phase transition in those samples could not be studied with the experimental setup at hand, but the results still serve as a guide to the changes in $T_c$ as a function of $d$ in this material. All the other samples displayed a temperature dependence of the magnetization corresponding to a 2D XY system. The somewhat lower $\beta$ of two samples, compared to the expected 0.23, can be attributed to either an influence of random anisotropy in the samples, shifting the value in accordance with the 2D $h_4$ model, or to the temperature range used in the fitting.

To further explore the critical behavior, both the exponents $\delta$ of the critical isotherm and $\gamma$ of the susceptibility were extracted. Neither of the exponents matched any theoretical model in the thermodynamic limit. Instead the temperature where $\delta = 15$ was located, identifying $T^*$, and via Eq. (2.13) the KT-temperature was determined. The value of $T_{KT}$ from this analysis matched the one estimated from a fit of Eq. (2.12) to the susceptibility, giving compelling evidence that the divergence follows the theory of the 2D XY model rather than a power law, albeit the two descriptions seemingly fit the data equally well, see Fig. 2.10. In addition, a new approach to give a measure of the correlation length was used to determine $b$ in Eq. (2.12), which will be explained in detail in the Appendix. If the same assumption as in Ref. [50] is used, i.e. $\eta = 0$ above $T_c$, the expected relation $B = 2b$ is fulfilled for two of the samples. The discrepancy in the values of $B$ and $b$ for the 12 Å sample can be attributed to noisy data, even though it can not be disregarded that some, for the author unknown, physics may be concealed in this result.

Paper III presents a similar investigation as Paper II, but with amorphous FeZr as the magnetic material. In this study the MOKE measurements were
supplemented with the use of a SQUID magnetometer allowing higher magnetic fields. A series of multilayers with different thicknesses of the spacer layer AlZr was manufactured and the observation of a constant critical temperature for all samples made it possible to exclude the existence of any coupling between the magnetic layers. The significance of this conclusion is made clear in the Section 3.1.1, but in short the presence of interactions would modify the phase transition.

A close analysis of the critical behavior was restricted to one sample with 15 Å thick FeZr layers and the exponent $\beta$ yielded a value of 0.21(1) in good agreement with theory. $T^*$ was identified as above, making it possible to calculate $T_{KT} = 134.6(1)$ K and a fit of Eq. (2.12) ($i.e. \xi \propto b/\epsilon^{1/2}$) to the correlation length gave the same value of the KT-temperature within error bars. Fig. 2.11 displays $\xi$ versus temperature with a fit where $T_{KT}$, deduced from $T_c$ and $T^*$, was used as a fixed parameter. The correlation length was determined using the assumptions that large correlated spin blocks exist above the critical temperature and that the size of the total magnetic moment in those dynamic regions is reflected in the magnetic response to an applied field. The lateral size of the correlated areas was estimated using SQUID data to extract the moment per atom below $T_c$. The calculation shows that regions with diameters as large as $\approx 1000$ Å are present 20 K above $T_c$.
Figure 2.11. Number of correlated spins in the unit of Bohr magnetons. The right axis shows the corresponding diameter. The solid line display a fit of Eq. (2.12) in this thesis to the data, using $T_{KT}$ as a fixed parameter, giving $b = 1.22(5)$; from Paper III.

The influences of structural disorder and random anisotropy on phase transitions have been addressed in many theoretical and experimental studies [54, 55, 56, 57, 58, 59, 60]. The results in Paper II and Paper III reveal that disorder in the form of an amorphous structure is a marginal perturbation of the system, which in essence does not alter the critical behavior in the 2D XY model. In addition, the amorphous CoFeZr and FeZr show a 2D signature for larger film thicknesses, compared to crystalline Fe.
3. Magnetic phase transitions - coupled layers

3.1 What happens on the edge of the world? - Boundary effects

"God made the bulk; the surface was invented by the devil." W. Pauli

Pauli made a harsh statement about surfaces, suggesting that they are nothing but a problem. Still, challenging is just another word for interesting, and surfaces and boundaries always play an important role in objects on the nanoscale. Even the phase transition in the common Heisenberg model shows the impact of boundaries: if the interactions are the same in the bulk and at the surface their critical exponents are $\beta_b = 0.32$ and $\beta_s = 0.84$, respectively [61]. This has been experimentally confirmed using Gd where $\beta_b = 0.376(15)$ and $\beta_s = 0.83(4)$ [62]. The influence of boundaries is also seen within a thin layer, where measurements demonstrate that the magnetization varies as a function of atomic index and the temperature dependence of the magnetization is different for the interior of the layer compared to the vicinity of the interface [63]. The diverse effects of symmetry breaking make it essential to explore what happens with magnetic and critical properties as boundary conditions change. One mechanism that can be the origin of an altered behavior is the interlayer exchange coupling.

3.1.1 No layer is an island - Interlayer exchange coupling

Multilayers can be used merely to increase the signal in measurements, which often is beneficial and sometimes necessary when dealing with thin films consisting of only a few atomic layers. Still, there is more to it. Magnetic layers which are separated by non-magnetic spacers can interact with each other through the interlayer exchange coupling (IEC), usually denoted $J'$ in mathematical descriptions. It should not be confused with the intralayer exchange coupling ($J$) which governs the magnetic interactions within a material.

The interlayer exchange coupling has been described using an RKKY framework in a theory developed by Bruno [64, 65]. In this picture, the origin of the coupling is a spin-dependent reflection of the spacer layer electrons at the interfaces. The IEC has an oscillatory nature, consisting of long- and short-period contributions, alternating between ferromagnetic and antiferromagnetic ordering of the magnetic layers, as schematically illustrated in Fig. 3.1. In the
Figure 3.1. Schematic illustration of FM (left) and AFM (right) interlayer exchange coupling between adjacent magnetic layers. The non-magnetic spacer is represented by the empty space between the layers.

thick spacer limit the dependence on the spacer layer thickness is described by the following equation:

\[
J' = \frac{A}{L_V^2} \sin \left( \frac{2\pi L_V - \phi}{\lambda} \right)
\]  

(3.1)

where \(A\) is the coupling strength, \(L_V\) is the spacer layer thickness, \(\phi\) is the phase and \(\lambda\) is the period, all expressed in monolayers. Eq. (3.1) only takes the long-period oscillation into account, since the interfacial roughness which is expected in sputtered samples is known to quench the short-period part [66]. The contribution of the short period is easily captured by adding a similar expression and sum over the two components.

IEC can also be explained within a quantum well model [67]. This description gives the same dependence on the thickness of the spacer layer, but more length scales turn out to be important and the interlayer coupling can not only be seen as an interface effect. Experimental observations support this view and oscillations in \(J'\) are found as functions of both magnetic layer thickness [68, 69] and cap layer thickness [70].

Fe/V superlattices have been used as a model system in numerous papers exploring the interlayer exchange coupling [71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81]. A small piece has been missing in the experimental mapping of the IEC in this materials combination: the behavior in the large spacer layer limit complemented with the temperature dependence of the magnetic properties. This is addressed in Paper IV, where a series of Fe(3)/V(x) superlattices, with \(x=16-27\) monolayers, is examined.

In a mean field approximation the critical temperature is directly proportional to the average exchange coupling in a superlattice and if the intralayer coupling is kept constant (i.e. fixed Fe layer thickness) all variations in \(T_c\) are due to the interlayer coupling. Fig. 3.2 shows the oscillations in the critical temperature together with a fit of Eq. (3.1) to the data, bearing in mind that \(T_c\) will increase irrespective of the sign of \(J'\) and therefore the absolute value of
the equation is used. X-ray analysis, together with an inspection of the shutter timing during deposition, revealed that the three last samples grown (V17, V20 and V27) had slightly thinner Fe layers than the others and this is taken into account in the figure. The first maximum in $T_c$ corresponds to results on Fe(2)/V(x) [75] and is in agreement with an earlier study on Fe(3)/V(x) [82]. The magnitude of the critical temperature is different though, and it is possible to conclude that it scales with the Fe thickness, while the oscillations are governed by the spacer thickness and thereby the IEC.

![Graph showing critical temperature versus vanadium thickness in Fe/V superlattices. The dashed line corresponds to a fit of the data to the absolute value of Eq. (3.1). The notations V16 etc. refer to the nominal thickness of the V layers in monolayers. The open squares show $T_c$ adjusted for a lower Fe thickness in these samples. (bottom) The effective exponent $\beta_{\text{eff}}$. The solid line serves as a guide to the eye. See text for details. The plot is supplemented with data from the literature and the corresponding references in this thesis are; red circles: [82] and green triangels: [75]. The figure is taken from Paper IV.](image)

Figure 3.2. (top) The critical temperature versus vanadium thickness in Fe/V superlattices. The dashed line corresponds to a fit of the data to the absolute value of Eq. (3.1). The notations V16 etc. refer to the nominal thickness of the V layers in monolayers. The open squares show $T_c$ adjusted for a lower Fe thickness in these samples. (bottom) The effective exponent $\beta_{\text{eff}}$. The solid line serves as a guide to the eye. See text for details. The plot is supplemented with data from the literature and the corresponding references in this thesis are; red circles: [82] and green triangles: [75]. The figure is taken from Paper IV.

The interlayer coupling was further investigated by analyzing the ratio between the remanence and the saturation magnetization as well as the saturation field, which can be used as an indicator of the strength of $J'$ in AFM coupled superlattices [80, 83]. The results are found in Fig. 3.3 and the observed oscillations follow closely the variation in $T_c$. Comparing our data to the measurements of Schwickert et al. [80] leads to the conclusion that not only the spacer thickness, but also the thickness of the magnetic layers govern the pe-
period and phase of the IEC. This means that the picture where only reflections of electrons at the interfaces play a role has to be abandoned for a quantum well description.

![Graph showing the ratio between remanence and saturation magnetization](image)

Figure 3.3. The ratio between the remanence and saturation magnetization in Fe(3)/V(x) superlattices (top), together with the saturation field (bottom). The black solid line shows an oscillation with the same period and phase as determined from the variations in $T_c$ (see Fig. 3.2). The black dash-dotted lines and the blue dotted lines mark the AFM minima in our data and the data from Ref. [80], respectively. The plot is supplemented with data from the literature and the corresponding references in this thesis are; green triangels: [75], red circles: [78] and blue dashed line: [80]. The figure is taken from Paper IV.

The temperature dependence of $J'$ has been experimentally determined to follow a power law [84]:

$$J' = J'_0 \left[ 1 - \left( \frac{T}{T_c} \right)^{3/2} \right]$$  \hspace{1cm} (3.2)

but theoretical results imply that the exponent might deviate from $3/2$ and its value is inferred to depend on both the magnetic and the spacer materials as well as the interface [85]. Nevertheless, Eq. (3.2) has been confirmed [86] and is supported by the results in Paper IV. The close resemblance between Eq. (3.2) and Bloch’s law [87] highlights the importance of spin wave excitations on the temperature dependence of the interlayer coupling.
Implications for the critical behavior

The notation $\beta$ is usually restricted to exponents determined for systems that can be compared to the theory of critical phenomena. If the magnetization in the system is influenced by interactions which are not captured within this theory, or if the exponent is calculated in a temperature region that can not be considered as the critical region, one usually speaks about an effective $\beta$ ($\beta_{\text{eff}}$). Fig. 3.2 displays $\beta_{\text{eff}}$ as a function of the vanadium thickness in Fe/V superlattices. It is evident that the values decrease with decreasing coupling strength, approaching the theoretical 0.23 of the 2D XY model. An explanation of this behavior is found in the results of Paper I. The idea behind the paper is justified with the following gedankenexperiment: imagine a superlattice with exchange coupled magnetic layers and let us restrict the discussion to nearest neighbor interactions. The two outermost layers will only have one neighbor each, while all others have two, and thus the magnetic ordering in the outer layers will be less affected by the stabilizing IEC. There is reason to believe that this would give rise to different temperature dependences in the layers due to their placement in the superlattice, and thus a gradient in the magnetic profile at elevated temperatures.

The most straightforward method to investigate the influence of boundaries, in the sense of missing neighbors, on the critical behavior is to investigate the temperature dependence of the magnetization in superlattices with different number of repetitions ($n$). The results showed that the exponent $\beta_{\text{eff}}$ increased from a 2D-like value of 0.19-0.26 for $n = 2$ to 0.32-0.41 for $n = 4 - 8$. This is in line with the predicted effect of an inhomogenous distribution of the magnetization between the layers in the superlattices where $n > 2$. A less abrupt decrease with temperature in the outer layers gives a higher exponent compared to the interior, and the outcome is effectively given by $\beta_{\text{eff}} = \frac{1}{n} \sum_{i}^{n} \beta_{i}$, summed over the monitored layers.

To further explore the magnetic profile of exchange coupled superlattices a sample was manufactured with a somewhat complicated structure, designed to allow layer resolved measurements. We entitled it a super-superlattice, since the basic construction consists of two superlattices, and the overall structure is described by: substrate/V(9)/[SL$_1$/SL$_2$]$_{15}$. SL$_1$ stands for the sequence [Fe(3)/V(9)]$_4$/Fe(3) and SL$_2$ represents V(12)/Fe(1)/V(10)/Fe(1)/V(12), where the numbers within the brackets denote the number of monolayers. The only reason for the two iron layers in SL$_2$ is growth aspects and they are not magnetic at any temperature [88].

The sample was studied using polarized neutron reflectivity. In this technique the magnetic contrast is given by the spin asymmetry,

$$S = \frac{I^\uparrow - I^\downarrow}{I^\uparrow + I^\downarrow}$$

(3.3)

where $I$ is the integrated intensity of the spin-up and spin-down reflectivities. $S$ is only sensitive to the projection of the magnetization ($M_y$) along the po-
larization axis of the neutrons and is proportional to $\Delta b M_y$, where $\Delta b$ is the difference in the nuclear scattering lengths of Fe and V [89, 90, 91]. Scattering techniques probe periodic variations in a structure, such as lattice constants or the bilayer thickness in a multilayer. The super-superlattice has an additional repeat distance, consisting of $[\text{Fe/}\text{V}]_5$ embedded in a non-magnetic slab, which is mirrored in the neutron reflectivity curves as six Bragg peaks. It is possible to reconstruct the magnetic profile of a single $[\text{Fe/}\text{V}]_5$ superlattice by monitoring these peaks at different temperatures. The result is found in Fig. 3.4 and more details about the approach is found in Paper I. The figure shows a profound gradient in the magnetic profile, which evolves with temperature. This observation confirms the view where the enhanced values of $\beta$, in exchange coupled superlattices, has its origin in distinct temperature dependences of the magnetizations in the individual layers, caused by their position within the sample. The conclusion is also warranted in the light of other observations. Hydrogen loading of Fe/V superlattices is a route to tune the interlayer exchange coupling [78], but modifications of the IEC does not change the exponent in a sample with only two repetitions and the value is consistent with the 2D XY model [51]. On the other hand, exchange coupled superlattices with $n > 2$ are continually reported to demonstrate higher values of $\beta \approx 0.3 - 0.8$ [51, 82, 88, 92].

![Figure 3.4](image.png)

*Figure 3.4.* The magnetic profile of an Fe(3)/V(9) superlattice with five repetitions. The image is constructed from data on a super-superlattice consisting of fifteen repetitions of the structure that is illustrated in the lower part of the figure. It should be noted that the apparent magnetizations in the Fe(1) layers are an artifact, due to the limited number of Fourier components available in the analysis; from Paper I.
3.1.2 You are who you associate with - Proximity effects

Experimental studies of two-dimensional systems are often made on thin films. However, such samples can never be ideal in the sense that they have a well defined thickness over the whole sample area. Atomic steps will always be present and this complicates the interpretation of the data. For example, if a film is found to be 2.5 monolayers (ML) thick, it means that some parts consist of two ML and others of three, since one can not speak about half an atom (this applies to crystalline materials). Moreover, most likely the thickness is even more distributed. Combining this fact with an increase in the critical temperature of $\approx 170 \text{ K/ML}$ (Fe in V [88]), implies that the value of $T_c$ will be unclear. There exist attempts to capture those effects, e.g. to assume a Gaussian distribution of critical temperatures [52].

One way to avoid the problems of thickness distributions and other atomic defects, while still being able to investigate properties of the 2D models, is to make Pd/Fe/Pd trilayers. Palladium is on the verge of being ferromagnetic, has a high susceptibility [93] and alloys of this material with a small amount of iron have been studied for a long time. A single ferromagnetic atom induces a polarization in the paramagnetic matrix with a radius of roughly 10 Å, giving a magnetic moment of 9-12 $\mu_B$ per Fe atom [94, 95, 96]. Therefore, thin films of Fe embedded in Pd represent a system where the magnetic thickness is much larger than any structural imperfections and they can be considered to be magnetically "perfect" [22]. In the ultrathin limit where the iron coverage is about 1 ML or less, the Fe atoms merely represent a magnetic doping of the Pd, and we adopt the term $\delta$-layer to stress the minute amount.

The critical behavior in samples with $\delta$-layers of Fe in Pd shows a dimensional crossover from 2D to 3D, when the amount of Fe is increased from 0.4 to 1.0 ML [97]. In Ref. [97] the magneto-optical Kerr effect was utilized to measure the magnetization. This method is to a large extent only sensitive to the palladium moment, as seen in the opposite sign of the Kerr rotation compared to Fe/V samples. In Paper VI we used element specific x-ray resonant magnetic scattering (XRMS) to further explore the magnetization in this system. The magnetic information in XRMS is contained in the flipping ratio of positively ($I^+$) and negatively ($I^-$) circularly polarized photons: $F.R. = (I^+ - I^-)/(I^+ + I^-)$. The results reveal a distinct difference in the temperature dependence of the element specific magnetizations, where $M(T)$ of Fe is described by a lower effective exponent compared to the Pd magnetization. Furthermore, in contrast to the palladium matrix which is ferromagnetic, $\delta$-layers with a thickness smaller than 0.7 ML show no spontaneous ferromagnetism. This is illustrated in Fig. 3.5, which shows the magnetic signal of the two samples with lowest Fe coverage in a field of 8 mT. We attribute the observations to a non-collinear magnetic ordering between the iron atoms.

Another system exhibiting magnetic proximity effects is amorphous FeZr/CoZr multilayers, which were studied in Paper V, and the results on the criti-
Figure 3.5. Flipping ratio versus energy of palladium layers with 0.7 ML and 0.5 ML thick $\delta$-layers of Fe. The scan was made in a field of 8 mT. Inset: The field dependence of the 0.7 ML sample; from Paper VI.

cal behavior have been addressed in Section 2.1.3. The CoZr coverage is only 1 Å, and thus not enough to form a complete layer. However, the proximity to the Co containing regions induces a magnetic order in the FeZr layers above their inherent critical temperature, which is approximately 150 K [98]. The range of interactions between the CoZr layers can be inferred from changes in $T_c$ as the spacing is decreased. Only a minor variation in the critical temperature can be observed between two samples with 75 and 50 Å thick FeZr layers. When the thickness is reduced to 25 Å, $T_c$ rises from 230 K to 270 K and this leads to the conclusion that the FeZr layers are polarized over a range which is smaller or equal to 50 Å, but larger than 25 Å in the vicinity of $T_c$. In addition, the difference in critical behavior between the samples makes it possible to deduce that the interactions actually extend roughly 50 Å from each CoZr layer.
4. Conclusions

The research in this thesis is centered around critical phenomena in magnetic thin films. It has been established that the strength of the interlayer exchange coupling ($J'$) in superlattices influences the measured effective exponents $\beta_{\text{eff}}$ when the sample includes more than two magnetic layers. Typically $\beta_{\text{eff}} > \beta_{\text{theory}}$, where the difference decreases as $J'$ goes to zero. Layer resolved polarized neutron reflectivity measurements revealed that the source of the modified exponents is different temperature dependences of the magnetization in outer and inner layers of the superlattices. In addition, the interlayer coupling in Fe/V superlattices was mapped out in the thick spacer layer limit and the results were supplemented with data from the literature. The results show that the phase changes and the period decreases with about 0.8 monolayers (ML) when the Fe layer thickness is increased from 3 ML to 7 ML and this yields a significant shift in the position of the antiferromagnetic maxima (about two monolayers).

The phase transition in thin films of amorphous materials are found to be described by the same theory as crystalline films and follow the expected behavior of the 2D XY model. The classification is not limited to the exponent associated with the zero-field magnetization, but also includes the critical behavior of the susceptibility, magnetic isotherms and the correlation length. Such broad approach is relatively rare in the literature where many studies on thin films barely focus on one magnetic quantity.

The critical behavior in FeZr(x)/CoZr(1Å) multilayers was investigated, where x=25, 50 and 75 Å. The small amount of Co is not enough to form a complete layer, but induces magnetic ordering in the FeZr layers above their inherent ordering temperature. The range of interactions between the Co containing regions was estimated to be about 50 Å, as inferred from the critical temperature and exponents. Scaling plots of the magnetization as a function of applied field and temperature display an excellent data collapse, although the samples display a non-universal critical behavior. This demonstrates that the scaling hypothesis is valid also within systems outside the universality classes and when the origin of the magnetization is ferromagnetic proximity effects.

Element specific magnetic measurements were conducted on Pd films with a $\delta$-layer of Fe, using synchrotron radiation. The temperature dependence of the palladium mirrors the overall behavior of the magnetization as measured by MOKE, while large discrepancies were found in the iron signal. The most intriguing observation is the absence of a magnetic signal for Fe with a coverage less than 0.7 monolayers.
5. Outlook

Paper I and Paper IV serve in essence as a closing point of the story on interlayer exchange coupling in Fe/V superlattices. But, there is one thing left to examine: the compelling evidence of long-range interactions between the magnetic layers. A dedicated experimental study on the relationship between the critical temperature and the number of repetitions, together with theoretical support, has the prospect of shining more light on the impact of different length scales on the collective magnetic behavior in a superlattice.

In Paper II and Paper III a methodology is employed which utilizes the magneto-optical Kerr effect to get a measure of the correlation length. It would be desirable to compare this approach with measurements using a more mature technique, such as neutron scattering.

Paper V explores the crossover from two-dimensional to three-dimensional behavior including not only \( \beta \), but also \( \delta \) and \( \gamma \). A natural continuation of this work is to study a model material, where the limiting cases of samples which can be classified as genuinely 2D XY and 3D Heisenberg are included. One question is how 2D XY-like a system can be before scaling fails, and another task is to complete the picture of the development within the crossover region. A promising candidate for this kind of study is amorphous FeZr, which is already proven to display 2D behavior and has a low critical temperature \([98]\). The situation is complicated by preliminary results show that thin films of \( \text{Fe}_{90}\text{Zr}_{10} \) display a decreasing critical temperature when the thickness is increased from 30 Å to 70 Å, the difference in \( T_c \) being 20 K. More measurements are needed to explore and explain this behavior. However, this system might be an excellent playground for further studies on the connection between the excitation of spin waves and the critical behavior, since a larger spatial extension is accompanied by a reduction in the thermal energy.
6. Populärvetenskaplig sammanfattning

Vi har alla erfarenhet av fasövergångar i vardagen då vi kokar vatten i köket, gjuter tennsoldater eller torkar tvätt. Detta är exempel på så kallade diskontinuerliga (första ordningens) fasövergångar och de är förknippade med en viss mängd energi som antingen frigörs eller måste tillföras, samt vanligen en omvandling av materialets struktur. Forskningen som beskrivs i denna avhandling handlar om en annan sorts fasövergångar som kallas kontinuerliga (andra ordningens) och som man inte stöter på i sina dagliga göromål. De går dock att förklara med ett jordnära exempel. En vanlig kylskåpsmagnet drar till sig häftstift, men om den upphettas kommer den inte kunna hålla kvar lika många häftstift och ju högre temperatur, desto fler stift ramlar av. Magneten är inte längre magnetisk \(^1\) vid och över en tydligt definerad temperatur som benämns den \textit{kritiska} temperaturen, \(T_c\) (ett alternativt namn är Curietemperaturen som syftar på Maries berömde make Pierre). Ingenting hänger med materialets atomära struktur, men ändå är de magnetiska egenskaperna helt annorlunda. Vad som har hänt är att magnetiseringen på atomär nivå, som huvudsakligen är en egenskap hos elektronen som kallas spinn, har gått från ett ordnat till ett oordnat tillstånd. Man kan ge en invecklad förklaring av vad spinn är, men ändå inte nära sig sanningen, så låt oss bara konstatera att det oftast avbildas som en pil, eller en kompassnål, och alltså har en rikting. Förändringarna i magnetiseringen kan beskrivas med enkla matematiska samband och kännetecknas av att de bara beror på skillnaden mellan den aktuella temperaturen och \(T_c\) samt en exponent. Även om magneters temperaturberoende är ett vanligt modellsystem för fasövergångar av den här typen, så återfinns de i många andra fysikaliska situationer. Över ett visst tryck och temperatur går det inte att särskilja flytande vatten och vattenånga, men under denna kritiska punkt är deras densiteter olika. Om man följer hur skillnaden i densitet minskar med ökande temperatur, så följer den exakt samma mönster som minskningen av magnetiseringen vid uppvärmning av kylskåpsmagneten.

I en enkel modell kan alla material delas in i sex universella klasser som bara beror på objektets fysiska utsträckning, dess dimension, och i vilka riktningar spinnet kan peka. 2D XY kallas klassen där objektet är tvådimensionellt (världen som en streckgubbe målad på ett papper skulle leva i, om den nu levde, är 2D) och spinnen kan rotera fritt i planet. Den största delen av forskningen i denna avhandling fokuserar på just denna klass. Tidigare har undersökningar av fasövergångar i tvådimensionella system nästan enbart

\(^1\)Strikt talat är den \textit{paramagnetisk}, men inte \textit{ferromagnetisk}.
rört kristallina material, där atomerna är regelbundet ordnade i ett gitter, och undantagen som finns gäller en annan symmetri där spinnen bara kan peka upp eller ner, den så kallade Isingmodellen. Vi har studerat vad som händer i amorfa, det vill säga glasliknande, material och har kunnat visa att den magnetiska fasövergången kan beskrivas med samma teori som i kristallina fallen trots att atomernas placering inte följer något regelbundet mönster.


Figure 6.1. Illustration av ett multilager. Notera att proportionerna är missledande. I en skalenlig figur skulle ett lager med en tjocklek på 1 mm motsvara en bredd på 1 mil.

Om ett antal järnatomer placeras mellan två lager av palladium kommer järnet att inducera ett magnetiskt moment i palladiumet. Detta system har tidigare studerats med metoder som bara mäter ett genomsnitt av vad som händer i båda materialen. Här har en annan teknik använts som bara är känslig

2Ett multilager är en skiktad struktur som består av minst två material. Beteckningen supergitter innebär att atomerna har samma regelbundna struktur genom hela provet och därfor kan ses som en enda kristall.
för ett ämne i taget, och mätningarna visar att järnets magnetisering har ett temperaturberoende som är väsentligt annorlunda jämfört med det som ob-
serveras i palladiumet, trots att de är djupt sammankopplade. Vi tillverkade
även amorfa multilager som växlande består av 25-75 Å tjocka FeZr-skikt
samt ofullständiga skikt av CoZr och de uppvisar en liknande inducerad mag-
netisering som Fe-Pd. Amorft FeZr har en mycket lägre kritisk temperatur än
kristallint järn och fasövergången sker redan vid cirka -120°C. Närvaron av
Co gör dock att magnetiseringen förblir ordnad vid temperaturer som är upp
till 100°C högre. En undersökning av provernas kritiska beteende visade att de
varken kan bli klassificerade som två- eller tredimensionella, utan uppträder
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Appendix: Data acquisition and analysis

The laboratory workhorse - The magneto-optical Kerr effect (MOKE)

Figure 6.2. The possible geometries in MOKE measurements. The red reflected lines represent the light and the thicker black arrows show the direction of the probed magnetization in each geometry, labeled accordingly.

When linearly polarized light is reflected on a magnetic surface, the polarization axis will rotate and the rotation angle is to a first approximation directly proportional to the magnetization. An ellipticity in the polarization will also be introduced. The mechanism is called the magneto-optical Kerr effect and has been utilized as a standard tool throughout the studies included in this thesis. There are three different geometries of MOKE that are sensitive to different directions of the magnetization and they are illustrated in Fig. 6.2. The only configuration used here is the longitudinal, but the others are presented for completeness. The signal in the longitudinal geometry is proportional to \( \cos \theta \), where \( \theta \) is the incident angle, while the penetration depth is proportional to \( \sin \theta \). In measurements on multilayers, the direction of the light therefore represents a balance between these two parameters. In general the beam probes about 100-200 Å of a metallic sample. The magnetization as a function of the temperature is obtained by measuring hysteresis loops, either continuously while changing \( T \) slowly or at certain temperatures.
Data analysis

A common assumption is that the spontaneous magnetization addressed in the theory of critical phenomena corresponds to the remanence in hysteresis loops measured by MOKE [11, 53, 99, 100]. However, it is important that the easy axis of the magnetization and the applied field are along the probed direction and that the field is high enough to saturate the sample. If the noise levels are high, it is beneficial to smooth the data. A method that phenomenological has been found to work well is to fit the following expression to the data:

\[ M(H) = B_0 \arctan \left( \frac{2 C_0 (\mu_0 H \pm a_0)}{\mu_0 H} \right) + A_0 \mu_0 H \]  

(6.1)

where \( B_0 \) is the height, \( C_0 \) is the squareness, \( A_0 \) is the slope and \( a_0 \) is the coercivity of the hysteresis loop. \( M \) is the magnetization and \( H \) is the applied field. Data from antiferromagnetically coupled layers has to be limited to fields below the saturation field, since a change of slope is not captured with Eq. (6.1). Three examples of measured loops together with fits are found in Fig. 6.3.

\[ M_0 \left[ \frac{dM_0}{dT} \right]^{-1} = \frac{(T - T_c)}{\beta} \]  

(6.2)

where \( M_0 \) is the zero field magnetization. It implies that if \( M_0 \left[ \frac{dM_0}{dT} \right]^{-1} \) is plotted versus temperature, the data should follow a linear dependence with

Figure 6.3. Hysteresis loops of three Fe/V superlattices, together with fits of Eq. (6.1) to the data. The notations refer to the nominal vanadium thickness in monolayers. Note the two different x-axes. The figure is taken from Paper IV.
a slope of $1/\beta$ and the intersection at $y = 0$ marks the critical temperature. This routine has a severe drawback when dealing with MOKE data. Typically the $M$ versus $T$ curve is not perfectly smooth, but there is always some noise that is amplified when differentiating the data. Fig. 6.4 shows the magnetization in one of the samples characterized in Paper V and the corresponding Kouvel-Fisher plots. The scattered data will inevitably make it hard, not to say impossible, to perform a reliable analysis.

![Figure 6.4.](image)

**Figure 6.4.** (a) Zero-field magnetization of the 50-1 FeZr/CoZr sample in Paper V. (b) The corresponding Kouvel-Fisher plot. (c) Plot of the same data as in (b), but zoomed around the critical temperature. The black line shows the expected behavior of the data, with a slope of $1/\beta$ and passing through $y = 0$ at $T_c$. The critical temperature is marked by the red cross.

Another method of finding $\beta$ and $\gamma$, together with the critical temperature, is the use of modified Arrot plots [104]. With a suitable choice of the exponents, $M^{1/\beta}$ versus $[H/M]^{1/\gamma}$ plots are parallel to each other at various temperatures and the line corresponding to $T = T_c$ goes through the origin. This technique is sometimes cited as only valid for high fields, but the reason why the linearization fails at low fields is demagnetizing effects, and they are usually negligible in thin films with in-plane magnetization. Therefore it might be well worth trying to employ this method in later studies, even though it has not been done in any of the papers in this thesis. Instead the main idea has been to find the best linear fit in a double logarithmic plot of $M$ versus $t$, where $t$ is the reduced temperature $1 - T/T_c$. (Note that the definition here differs from the conventional one used in the other chapters, by defining temperatures below $T_c$ as positive.) The simple approach of using a fixed fitting range, typically
$10^{-2} \leq t \leq 10^{-1}$, was subsequently replaced with a more rigid method where \( \chi^2 \) in the following expression was minimized:

\[
\chi^2 = \frac{1}{n-2} \sum_{t_{\text{min}}}^{t_{\text{max}}} \left[ \log M_i - (\log k + \beta \log (1 - T_i/T_c)) \right]^2
\]  \hspace{1cm} (6.3)

where \( n \) is the number of data points included in the fit, \( t_{\text{max}} \) is the maximum reduced temperature, \( t_{\text{min}} \) is the minimum reduced temperature and \( k \) is a constant. The values of \( t_{\text{max}} \) or \( t_{\text{min}} \) and \( T_c \) are changed in small steps and the best fit, identified as the minimum in \( \chi^2 \), determines the values of \( \beta \) and \( T_c \). In addition, direct fits of the relation \( M \propto t^\beta \) to the data together with a Gaussian distribution in \( T_c \) [52, 105] has also been used in some of the studies.

In Paper II and Paper III a measure of the correlation length (\( \xi \)) is obtained from the magnetic response above the critical temperature. The idea is that the strong response of the magnetization to an applied field above the ordering temperature is inconsistent with a paramagnetic state, but reflects the presence of dynamic macrospins. The \( M \) curves are \( s \)-shaped and low fields serve to line up otherwise uncorrelated macrospins. The slope at higher fields is much lower, indicating that the vast majority of the macrospins are aligned. We defined a so called cross-over field (COF) as the crossing of two lines representing the magnetic response at low and high fields, respectively, and this is illustrated in Fig. 6.5(a). Larger regions of correlated spins are expected to align at lower fields and thus the COF is inversely proportional to the size of the macrospins.

![Hysteresis loop of an amorphous multilayer with 12 Å thick CoFeZr layers, measured at 114 K, i.e. \( 1 - T/T_c = -0.06 \). The cross-over field is defined as the intersection of two linear fits to the high and low field regions, respectively. The figure is taken from Paper II.](image)

This approach only gives an approximation of the relative change of the size with temperature. Using a Langevin description [106] of the magnetization allows us to estimate the actual diameter of the correlated regions:
\[ M(H) \propto n \mu L \left( \frac{\mu_0 H \mu}{k_B T} \right). \quad (6.4) \]

where \( n \) is the density of macrospins, \( \mu_0 \) is the permeability of free space, \( k_B \) is Boltzmann’s constant and \( L(x) = 1/\tanh(x) - 1/x \) is the Langevin function. The fit parameter \( \mu \) gives the magnetic moment of the macrospins, which is proportional to their volume (\( V \)). Assuming that they have a cylindrical shape makes it possible to calculate the correlation length

\[ \xi \propto \left( \frac{V}{d} \right)^{1/2} \propto \left( \frac{\mu}{md} \right)^{1/2} \quad (6.5) \]

where \( m \) is moment per magnetic atom and \( d \) is the thickness of the layer. The result from \textbf{Paper II} is plotted in Fig. 6.6 and illustrates that the COF and Langevin approaches complement each other by being applicable in different temperature ranges.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure66.png}
\caption{The magnetic moment of the macrospins and the corresponding correlation length, determined from Langevin fits and the inverse cross-over field, using measurements on a amorphous multilayer with 14 Å thick CoFeZr layers. The COF data is rescaled with respect to the Langevin data at 237 K; from \textbf{Paper II}.}
\end{figure}
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