MULTI-CRITICAL POINTS IN WEAKLY ANISOTROPIC MAGNETIC SYSTEMS

A neutron scattering study of two low dimensional antiferromagnets

BY

J A J BASTEN


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This report is the author's Ph.D. thesis. It starts with an extensive presentation of the basic concepts and ideas of the modern theory of static critical phenomena. Then the predictions of the molecular-field and renormalization-group theories on the phase transitions and critical behaviour in weakly anisotropic antiferromagnets are treated. Especially the behaviour close to the corresponding multi-critical points, as predicted by the extended-scaling theory, is discussed.

In the experimental part of the study a rather complete picture of the critical behaviour of CoBr$_2$.6[0.48D$_2$.0, 0.52H$_2$.0] (CB48) near the Néel point is reported, which has been obtained by quasi-elastic neutron scattering. Subsequently the magnetic phase diagram of CB48 in a parallel magnetic field is presented. Strong evidence has been found for the existence of an intermediate phase between the antiferromagnetic and the spin-flop phase, which implies that the (H,T)-diagram contains a tetra-critical instead of a bicritical point.

Neutron-scattering data on CsMnBr$_2$.2D$_2$.O are used to verify the extended-scaling theory. A striking data-collapsing of the scaled order parameters, determined in the entire (H,T)-phase diagram, on two scaling functions could be achieved. This is a very direct confirmation of the extended-scaling hypothesis. Crossover from critical to bicritical behaviour has been observed. The corresponding critical exponents $\beta$ and $\delta_b$, and the crossover exponent $\delta$ are found to be in good agreement with the theoretical predictions.

KEYWORDS
ANISOTROPY
ANTIFERROMAGNETIC MATERIALS
NEUTRON DIFFRACTION
ANTIFERROMAGNETISM
NEEL TEMPERATURE
CRITICAL FIELD
COBALT BROMIDES
CESIUM COMPOUNDS
MANGANESE COMPOUNDS
PHASE DIAGRAMS
PHASE STUDIES
ORDER-DISORDER TRANSFORMATIONS
EQUATIONS OF STATE
QUASI-ELASTIC SCATTERING
SCALING LAWS
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CHAPTER I

INTRODUCTION

In the description of phase transitions classical theories, such as the molecular-field theory for magnetic systems, may be considered as exact for systems with infinite-range interactions [1]. They usually account fairly well for the occurrence of various types of ordering, also in systems with short-range interactions. However, the validity of this description is drastically reduced near critical points, where the short-range interactions play an important role. So far, no exact solution for three-dimensional systems near criticality exists. Even for the simplified mathematical systems of lower spatial dimensionality d, which have been the subject of widespread interest, only in very few cases an exact solution could be obtained [2]. It appeared that physical realizations of such low-dimensional systems can be found in certain magnetic materials, where the interactions between the magnetic moments are restricted to chains or layers [3]. These so-called low-dimensional magnetic systems offer an interesting possibility to verify calculations based on simplified model Hamiltonians. As an alternative approach several approximative methods have been developed, usually based on series expansions and other extrapolation techniques, which yield fairly good descriptions of ordering processes in d=3 systems even close to a critical point [4].

Both calculations and experiments show that in many systems the same singular behaviour of thermodynamic variables is found near criticality, suggesting that the ordering proceeds in the same way. Apparently the type of the interactions which lead to the long-range order, is of minor importance for the description of the ordering process itself. The critical behaviour appears to depend only on a limited number of parameters, specifically on the spatial dimensionality (d) and the number of degrees of freedom (n) that take part in the ordering. This common feature of ordering systems is known as universality. The origin of long-range order can be found in the divergence of the range of the correlations in a system at the ordering temperature. It appears that
this divergence of the correlation length causes all singularities which are observed in a system near criticality. As we shall discuss in chapter II, the relation to the correlation length imposes strong limitations to the functional form of the singularities and implies close relations between the various critical quantities. This feature of ordering systems is known as scaling [5].

Only recently Wilson et al. [6] integrated these semi-phenomenological concepts of scaling and universality and extended them to a detailed method for the calculations of critical behaviour. This approach, called the Renormalization-Group (RG) approach, had a very important impact on the theoretical understanding of criticality. The success of the RG-theory has encouraged theoreticians to proceed to studies on the phenomena which occur when different kinds of critical behaviour compete. As we shall see in chapter III, the existence of such a competition is usually limited to the neighbourhood of certain points in thermodynamic field space, the so-called multi-critical points [7].

For the description of multi-critical behaviour a straightforward extension of the scaling concept is sufficient. This so-called extended-scaling theory, which is introduced in Chapter III, predicts many characteristics of multi-critical points and gives also a description of the crossover from one kind of critical behaviour to another [8]. In the experimental verification of the extended-scaling theory, which so far has been fragmentary, magnetic systems again play an important role. This is due to the convenient way in which the relevant thermodynamic field parameters can be changed, in combination with the large variety of critical and multi-critical behaviour displayed by magnetic systems. Although the fundamental concepts of the theory on critical phenomena are applicable in a wide range of systems, our discussions will be restricted mainly to magnetic systems and more specifically to antiferromagnets.

In the experimental part of this report results are presented of neutron scattering studies on the critical and multi-critical behaviour of two low-dimensional antiferromagnets. Both materials are so-called spin-flop systems, i.e. antiferromagnets with weakly anisotropic interactions. CoBr$_2$·6(0.48 D$_2$O, 0.52 H$_2$O) is well known as a good approxi-
mation of the $d=2$ XY-model (i.e. $d=2$, $n=2$) \cite{9}. An extensive study has been performed on the critical behaviour of this compound close to the ordering temperature $T_N$. A careful profile analysis of the observed critical-scattering data provides a rather complete picture of the ordering process. We have also investigated the multi-critical behaviour of this material and the spin-flop region. These measurements provide for the first time strong evidence for the existence of a so-called intermediate phase in a spin-flop system, which has been questioned by theoreticians for a long time \cite{10}. The second material investigated is CsMnBr$_3$·2D$_2$O, known as a good approximation of the $d=1$ Heisenberg model (i.e. $d=1$, $n=3$) \cite{11}. The experiments on this compound have been performed to verify various aspects of the extended-scaling theory on multi-critical behaviour in spin-flop systems. The results of this study provide the first verification of the central assumption of this theory, i.e. the extended-scaling hypothesis itself \cite{8}.

The organization of this report is as follows. We start in chapter II with an introduction in the field of critical phenomena. We discuss the concepts of scaling and universality and the integration of both ideas in the calculational approach of the Renormalization-Group (RG) theory. In chapter III we will focus our attention to the magnetic phase diagrams of spin-flop systems and the related critical and multi-critical behaviour. The predictions of both the molecular-field (MF) theory and of scaling theory will be presented, as they are often complementary. Chapter IV contains the results of our neutron-scattering study on the critical behaviour of CoBr$_2$·6(0.48 D$_2$O, 0.52 H$_2$O) near the Néel point. The experiment on the same compound in an applied magnetic field is discussed in chapter V. Finally, in chapter VI the neutron-scattering study on CsMnBr$_3$·2D$_2$O is treated. References have been gathered at the end of each chapter. A list of symbols and abbreviations is given at the end of this report. Throughout this work numerical results of least-squares fits are given with 10-standard deviations, based on statistics only, within parentheses and expressed in units of the last decimal place; for instance $T_N = 3.1175(10)$ K means $T_N = 3.1175 \pm 0.0010$ K.
References to Chapter I


2.1. Introduction

The investigations reported in this work relate to critical phenomena, a field of research which has been the subject of a fastly expanding number of theoretical studies. In the present investigations the choice of the topics, the applied methods in the data analysis and the interpretation of the results are all three closely interwoven with recent theoretical developments. Therefore, we feel that it is necessary to give a review of the concepts and ideas which constitute the basis of the modern theory of critical phenomena.

In this chapter we shall confine ourselves to a presentation of the most important quantities and concepts, with use of a minimum of mathematics. Therefore, we shall often appeal to topological and heuristic arguments. The reader who wants to go into further detail, is referred to the many excellent papers and books which have been published during the last few years and which treat the developments and the results of modern theory on phase transitions and critical phenomena, either on an introductory or on a review level [1-5]. In addition, a large variety of introductions has been published on the Renormalization-Group (RG) approach of calculations related to critical behaviour [6-10]. In this work no such RG-calculations will be performed nor reproduced. On the other hand, we shall repeatedly use predictions and results, which have been obtained by means of RG-calculations. Therefore, it seems convenient to present also an introduction in the terminology of the RG.

In section 2.2. we start with the basic definitions of quantities and concepts used in the description of critical phenomena. The singular behaviour which is observed near critical points, is described in section 2.3. There we shall also show that many aspects of critical behaviour, for example in magnetic systems, can be studied elegantly
by means of neutron scattering. The subsequent two sections treat the concepts of scaling and universality, interpreted as a direct consequence of the divergence of the correlation length $\xi$ at critical points.

The sections 2.6 and 2.7 treat the principles of the RG-approach of the calculation of critical behaviour. This part of the chapter may be skipped without consequences for the understanding of the experimental part of this work. In section 2.6 we shall show how calculations of critical behaviour are simplified in a fundamental way because of the divergence of $\xi$, which permits the use of effective Hamiltonians. In section 2.7 we treat the calculational recipe of the RG-approach and indicate how the results are obtained.

2.2. Basic definitions

We consider a magnetic system in contact with its environment, which consists for instance of a magnetic field $\vec{H}$ and a heat reservoir at temperature $T$. At equilibrium the variable "extensive" quantities of the system, such as the magnetization $\vec{M}$ and the entropy $S$, will take on values which minimize the Gibbs free energy

$$G = U - \vec{H} \cdot \vec{M} - TS ,$$  \hspace{1cm} (2.1)

where $U$ is defined as the internal energy of the sample plus magnetic field [11]. According to Griffiths and Wheeler [12], the thermodynamic variables in (2.1) can be classified as "fields" and "densities". The fields, denoted as $h_i$, have the property that they assume identical values in two or more phases which are in thermodynamic equilibrium. This is not necessarily true for the conjugated densities, which are defined by

$$\rho_i = -\frac{\partial G}{\partial h_i} ,$$  \hspace{1cm} (2.2)

According to this definition, $\vec{H}$ and $T$ in (2.1) are fields, whereas $\vec{M}$ and $S$ are densities. In general the definition of $G$ in (2.1) may be extended with additional pairs of conjugated variables in terms of the
Now, a first-order phase transition can be defined as a discontinuity of at least one of the densities, say $\rho_j$, as a function of at least the conjugated field. At this transition the state of the system is not uniquely defined. One can distinguish between two phases $1$ and $2$, characterized by different density values $\rho_j^{(1)}$ and $\rho_j^{(2)}$, whereas $h_j^{(1)} = h_j^{(2)}$ for all fields $h_j$. In the $z$-dimensional field space this first-order phase transition extends as a $(z-1)$-dimensional hypersurface, the so-called coexistence surface (CXS). The CXS separates the two distinct phases 1 and 2. Such a surface may terminate in various ways \[12\]. Firstly, the CXS may intersect another CXS in a triple boundary, a hypersurface of dimension $z-2$ at which three phases are in equilibrium. Secondly, the CXS may terminate in a critical boundary, a hypersurface of dimension $z-2$, with the property that the discontinuities in the densities vanish continuously upon approaching a point of this critical hypersurface. In this terminology, a multi-critical surface can be defined as a surface in field space, where two or more critical boundaries meet each other. In the rest of this chapter we shall mainly deal with the properties of magnetic systems close to critical boundaries.

The simplest example of a critical boundary in a magnetic system is the Curie point $T_C$ of a ferromagnet. In the two-dimensional field space, spanned by the fields $H$ and $T$, a first-order transition extends as a one-dimensional CXS along the $T$-axis (Fig. 2.1.). At this coexistence line two phases 1 and 2 coexist, which are distinguished by a different value of the density $M$, viz. $M_1$ and $M_2 = -M_1$. As a
function of $T$ the discontinuity in $M$, i.e. $M_1 - M_2$ vanishes continuously upon approaching the critical point $T_c$.

It has been pointed out by Griffiths and Wheeler [12], that the type of critical behaviour which is observed upon approaching a critical boundary, crucially depends on the path of approach in field space. Consequently, for a correct analysis of critical phenomena a further subdivision of the fields and densities is needed. This division has to be determined for each critical point $CP$ at the critical boundary of a CXS, and therefore has only "local" validity. In a point $CP$ in field space, a field for which the corresponding axis is not asymptotically parallel to the CXS at $CP$, is called an ordering field and the conjugated density is called an ordering density or an order parameter. The remaining fields are nonordering fields and the conjugate densities are nonordering densities. The nonordering fields, asymptotically parallel to the CXS at $CP$, are further divided in irrelevant fields, which are asymptotically parallel to the critical boundary at $CP$, and relevant fields, which are not asymptotically parallel to the critical boundary at $CP$. These definitions imply that any relevant field carries the system away from criticality, whereas an irrelevant field carries the system along the critical boundary. *

In this sense also an ordering field is always relevant. Here we emphasize once more, that this subdivision of the various fields not necessarily remains the same along the whole critical boundary. It will be seen in section 2.7., that the above division of fields can be extended in a logical way for the description of multi-critical behaviour.

At the critical point $T_c$ of a ferromagnet in $HT$-space (Fig. 2.1.), the magnetic field $H$ is the ordering field and the magnetization $N$ is the order parameter. The temperature $T$ is a relevant nonordering field and the entropy $S$ is a nonordering density. According to the definitions, also a combination of fields like

$$h = H + (T - T_c)^2$$

(2.3)

*If the critical boundary consists of a single critical point, any nonordering field is relevant, since it carries the system away from criticality.
and
\[ t = H^3 + (T - T_C) \]  

might be used as local definitions of the ordering field and the non-ordering relevant field, respectively. Although this would be not very useful in this simple case, we shall meet situations where it is much less clear which choice of fields is the most convenient one for the description of the encountered critical phenomena.

A phase diagram similar to Fig. 2.1. is shown by an anisotropic antiferromagnet in zero magnetic field (Fig. 2.2.). For \( T < T_N \) the magnetic moments are ordered in two sublattices A and B with sublattice magnetizations \( \vec{M}_A \) parallel and \( \vec{M}_B \) antiparallel to some easy axis.* The order parameter is now the staggered magnetization

\[ \vec{M}_{st} = (\vec{M}_A - \vec{M}_B)/2 \]  

As the conjugated ordering field a fictitious staggered field \( \vec{H}_{st} \) may be defined, which points in opposite directions for the two sublattices, and therefore shows the same spatial symmetry as \( \vec{M}_{st} \). In the two-dimensional field space spanned by \( H_{st} \) and \( T \), again a first-order phase boundary extends as a one-dimensional CXS along the \( T \)-axis (Fig. 2.2.). At this coexistence line two phases 1 and 2 coexist, which are distinguished by a different value of the order parameter \( \vec{M}_{st} \), viz. \( \vec{M}_{st}^{(1)} = (\vec{M}_A - \vec{M}_B)/2 \) and \( \vec{M}_{st}^{(2)} = (\vec{M}_A' - \vec{M}_B')/2 = [(-\vec{M}_A) - (\vec{M}_B)]/2 = -\vec{M}_{st}^{(1)} \). As a function of the relevant nonordering field \( T \) the

* We shall not consider antiferromagnets with more than two sublattices.
order parameter vanishes continuously upon approaching the critical point $T_N$, the Néel point.

If we extend the field space with a third dimension, viz. the external magnetic field $H$, the CXS appears to be a coexistence surface, extending in the $H = 0$ plane (Fig. 2.3.). Therefore, $H$ can be considered as a nonordering field in an antiferromagnet. The two-dimensional CXS can be limited in certain cases entirely by a critical line, as drawn in Fig. 2.3. From the shape of the critical boundary in this particular case, it may be derived that $H$ is an irrelevant field at the Néel point ($H=0$, $T=T_N$), whereas $H$ is relevant elsewhere. $T$ is a relevant field along the entire critical line, except in the point ($H=H_c$, $T=0$).

In the following sections we shall focus on the critical behaviour of a system near a single critical point, which may be part of a more extensive critical boundary. We shall show that many aspects of critical behaviour, among others in ferromagnets and antiferromagnets, can be investigated directly by means of thermal neutron scattering [13]. For the time being we are not concerned with multi-critical points, which will appear again in section 2.7. The possible occurrence of various phase diagrams in real antiferromagnets will be discussed in chapter III.
2.3. Critical behaviour and neutron scattering

Critical fluctuations

A system near a critical point is characterized by large fluctuations in the order parameter, which become slower and extend over larger distances as the critical point is approached. In a magnetic system these fluctuations can be introduced through a space-time spin correlation function $G^{\alpha\beta}(R,t)$ defined by

$$G^{\alpha\beta}(R,t) = <S_{\alpha}^{\alpha}(0)S_{\beta}^{\beta}(t)>. \quad (2.6)$$

Here, the angular brackets denote the thermal average value at a given temperature $T$, so (2.6) is the probability of finding the $\beta$-component $S_{\beta}^{\beta}(t)$ of the spin at position $R$ and at time $t$, given that the $\alpha$-component of the spin at the origin was $S_{\alpha}^{\alpha}(0)$ at time 0. In an antiferromagnet as well as in a ferromagnet the order parameter is proportional to $<S^{\alpha}>$.

If the critical point is approached from the disordered phase along a path in field space corresponding to a relevant nonordering field, the extent $R$ of the region where $G^{\alpha\alpha}(R,t)$ has a finite non-zero value increases, although the value of $<S_{\alpha}^{\alpha}(t)>$ is still zero. This means that the short-range order in the system grows. This growing of the regions with correlated spins continues until at the critical point $<S_{\alpha}^{\alpha}(t)>$ departs from zero, i.e. until long-range order (LRO) is established. Also in the ordered phase fluctuations in the order parameter still exist and wide regions are present where the spins have a "wrong" orientation. Complete ordering will only exist at $T=0$. The fluctuations around the equilibrium value $<S_{\alpha}^{\alpha}>$ are expressed by the net correlation function

$$G^{\alpha\beta}_{n}(R,t) = <S_{\alpha}^{\alpha}(0)S_{\beta}^{\beta}(t)> - <S_{\alpha}^{\alpha}(0)> <S_{\beta}^{\beta}(t)> \quad (2.7)$$

which has the property to be zero far from the critical point, both in the disordered and in the ordered phase. The behaviour of the spin fluctuations sketched above can be studied directly by means of neutron scattering.
Critical neutron scattering

If a beam of thermal neutrons is incident upon a solid, the interaction between the nuclei of the atoms and the neutrons will give rise to nuclear scattering \[14\]. Since the neutron carries a magnetic moment, there will be an additional magnetic scattering, due to the interaction between the magnetic moments of the atoms and the neutron. Here we are interested mainly in the magnetic scattering process. In a scattering experiment part of the neutrons will be scattered, which results in a change in their wave vectors by

\[
\vec{Q} = \vec{k} - \vec{k}_0.
\]  

(2.8)

Here \(\vec{k}_0\) and \(\vec{k}\) are the wave vectors of the incoming and scattered neutrons, respectively, and \(\vec{Q}\) is the so-called scattering vector. The neutron energy loss is equal to

\[
E_0 - E = h\omega = \frac{h^2}{2m}(k_o^2 - k^2),
\]  

(2.9)

where \(h\) is Planck's constant and \(m\) the neutron mass. When unpolarized neutrons are used, the double-differential magnetic scattering cross section per unit solid angle \(\Delta\omega\) and per unit energy \(E\) is given by \[14\]

\[
\frac{d^2\sigma}{d\Omega dE} = \frac{k}{k_0^2} |f(Q)|^2 \sum_{\alpha\beta} \delta_{\alpha\beta} \hat{Q}_\alpha \hat{Q}_\beta \hat{G}_{\alpha\beta}(Q,\omega).
\]  

(2.10)

where

\[
\hat{G}_{\alpha\beta}(Q,\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \int R \exp \{i(Q\cdot R - \omega t)\} G^{\alpha\beta}(R,t).
\]  

(2.11)

In (2.10) \(f(Q)\) is the magnetic formfactor, \(\delta_{\alpha\beta}\) the Kronecker delta and \(\hat{Q}_\alpha\) the \(\alpha\)-component of the unit vector \(\hat{Q}/Q\). From (2.10) it appears that this neutron scattering cross section is proportional to the space-time Fourier transform of the correlation function (2.6).

Close to critical points, the critical fluctuations in the order parameter appear as quasi-static to the scattered neutrons \[15,16\]. This implies that the inelasticity of the scattering is small and
The function $\hat{c}(q, \omega)$ corresponds to a narrow profile centered at $\omega = 0$. As the critical point is approached, the width $\Delta \omega$ of this profile decreases, which is known as the phenomenon of critical slowing down. Here, we shall confine ourselves to a study of this quasi-static behavior of the fluctuations close to criticality. To determine the static correlation function $G^2_b(R) \equiv G^2_b(R, t=0)$, we do not need the complete energy analysis of the scattering cross section (2.10). This can be seen directly from the definition of $G^2_b(R)$. Actually, only the $Q$-dependence of

$$
G^2_b(Q) \sim \int \frac{d\omega}{\omega} G^2_b(Q, \omega)
$$

has to be determined. $G^2_b(Q)$ may be considered as the wave-vector representation of the spin fluctuations. In an ordered solid with reciprocal lattice vectors $Q = 2\pi \mathbf{r}$, corresponding to the magnetic unit cell, $G^2_b(Q)$ has the periodicity of the reciprocal lattice, i.e.,

$$
G^2_b(Q) = G^2_b(q),
$$

where we introduced the deviation vector

$$
q = Q - 2\pi \mathbf{r}.
$$

With the Fourier transform of $S^2_k$ given by

$$
S^2_k = \sum_{\mathbf{R}} \exp(i\mathbf{q} \cdot \mathbf{R}) \frac{S^2_k}{\mathbf{R}},
$$

we can write (2.12) in the commonly used notation

$$
G^2_b(Q) \equiv G^2_b(q) = \sum_{\mathbf{R}} \exp(i\mathbf{q} \cdot \mathbf{R}) \langle S^\alpha_o \mathbf{S}^\beta_k \rangle = \langle S^\alpha_k \mathbf{S}^\beta_k \rangle .
$$

As in (2.7), a net correlation function $c^2_b(q)$ can be defined as

$$
c^2_b(q) = G^2_b(q) - \langle S^\alpha_k \mathbf{S}^\beta_k \rangle .
$$

It will be seen below, that $c^2_b(q)$ is a bell-shaped function, maximum at $q = 0$ and with widths $\kappa_a$, $\kappa_b$ and $\kappa_c$ along the $a$, $b$ and $c$-axes, respectively, which indicate an appropriate orthogonal set of crystal axes.
1.4. \( Q \) and \( Q_x \), corresponding to neutrons which are detected at the same scattering angle but have been scattered elastically and inelastically, respectively.

In a critical-scattering experiment \( G_n(x) \) can be determined to a very good approximation \(|5,16|\) by measuring the differential cross section \( \frac{d\sigma}{d\Omega} \). The essential approximation in this procedure is the following. If the scattering is inelastic, the neutron wave vector will change not only in direction but also in magnitude. It follows directly from (2.8) and (2.9) that for small \( \omega \)

\[
\Delta k = -\frac{m_0}{\hbar k} \omega.
\]  

(2.16)

It is shown in Fig. 2.4. that neutrons, scattered in the same direction with a slightly different energy transfer \( \Delta(\hbar \omega) \), are not scattered with the same momentum transfer \( \Delta \Omega \). Therefore, the \( \Omega \)-dependence of \( G_n(x) \) can be determined directly from \( \frac{d\sigma}{d\Omega} \) only if the condition

\[
\Delta k \ll \kappa
\]  

(2.17)

is fulfilled, i.e., if the spread \( \Delta k \) due to the inelasticity (2.16) is much smaller than the width \( \kappa \) of \( G_n(x) \). The assumption that (2.17) holds in a scattering experiment is known as the quasi-static approximation. The origin of this denomination will be explained below. If (2.17) holds, the expression

\[
\frac{d\sigma}{d\Omega}(\Omega) \propto |f(\Omega)|^2 \sum_{\gamma} \left( \delta_{\alpha\beta} \delta_{\alpha\beta} \right) G_n(x)
\]  

(2.18)
is valid. The cross section can be separated into two terms,

\[ \frac{d\sigma}{d\Omega} (\hat{q}) = |f(Q)|^2 \sum_{\alpha \beta} \left( \tilde{\sigma}_{\alpha \beta} \tilde{\sigma}_{\alpha \beta} \right) . \]  

(2.19)

\[ \left[ \tilde{\sigma}_{\alpha \beta} (\hat{q}) + \sum_{\mathbf{k}} \exp (i\mathbf{q} \cdot \mathbf{R}) <s^\alpha_0> <s^\beta_0> \right] , \]

where the first term represents short-ranged fluctuations and the second term is the Bragg term, corresponding to the LRO.

Fig. 2.5.

\[ \frac{d\sigma}{d\Omega} (\hat{q}) \] through the (111) reciprocal lattice point in RbMnF\(_3\) at T near T\(_c\). The full line is a least-squares fit of \(\tilde{\sigma}_{\alpha \beta} (\hat{q})\) from eq. (2.20) folded with the (high) experimental resolution (taken from 16). 

As an example, Fig. 2.5. shows the neutron scattering cross section \(\frac{d\sigma}{d\Omega} (\hat{q})\), measured with an extremely high resolution around the (111) reciprocal lattice point in cubic RbMnF\(_3\)\(16\). Similar scattering profiles are observed near all reciprocal lattice points \(\hat{q}_0 = 2\pi/\mathbf{a}\), where according to (2.19) magnetic Bragg peaks appear in the ordered phase of RbMnF\(_3\). The shape of \(\tilde{\sigma}_{\alpha \beta} (\hat{q})\) (\(\tilde{\sigma}_{\alpha \beta} (\hat{q})\) in a cubic system) appears to be perfectly described by
The corresponding net static correlation function $G_n^\alpha(R)$ can be found as
\[ G_n^\alpha(R) = \frac{\Lambda^\alpha}{(\kappa^\alpha)^2 + q^2} \exp[-\kappa^\alpha R], \] for $R \to \infty$. (2.21)

In (2.20) and (2.21) the exponent $\kappa^\alpha$ represents the deviation in the shape of $G_n^\alpha$ from the classical Ornstein-Zernike theory. It clearly plays the role of a correlation length, as it is a measure of the range of $G_n^\alpha(R) = \langle S_R^\alpha S_R^\alpha \rangle$. In general a different amplitude $\Lambda^\alpha$ and width $\kappa^\alpha$ are found for each spin component. In a non-cubic, anisotropic system expressions similar to (2.20) and (2.21) hold for each component of $\vec{q}$ and $\vec{R}$, respectively.

With $1/\kappa = \xi$ and $v = \hbar k/m$ as the neutron velocity, the quasi-static approximation (2.17) can be written as
\[ \frac{1}{\omega} = \frac{m}{\hbar k} \frac{1}{|\Delta k|} >> \frac{\xi}{v}, \]
where we used (2.16). The left side represents a characteristic time of the spin fluctuations and the right side is the passage time for a neutron through a correlated region. Therefore, the above requirement implies that the spin fluctuations appear static to the neutrons.

Through the fluctuation-dissipation theorem, relations exist between the net static correlation function $G_n^\alpha(q)$ and many other thermodynamic quantities, such as the generalized susceptibility $\chi^\alpha(q)$. This is the response function of the magnetic system for a static, spatially modulated magnetic field
\[ H^\alpha(R) = H^\alpha(\vec{q}) \exp(i\vec{q} \cdot \vec{R}), \] (2.22)
and is defined as
\( \chi^{\Delta z}(\vec{Q}) = \frac{\partial \mathcal{M}^{z}(\vec{Q})}{\partial H^{3}(\vec{Q})} \).  

(2.23)

For \( \vec{Q} = \pm \pi \vec{T} \), (2.23) corresponds to the familiar susceptibility
\( \chi^{\Delta z} = \frac{\partial \mathcal{M}^{z}}{\partial H^{3}} \) in a ferromagnet and to the staggered susceptibility
\( \chi_{st}^{\Delta z} = \frac{\partial \mathcal{M}^{z}_{st}}{\partial H^{3}_{st}} \) in an antiferromagnet. It can be shown that

\[
\frac{k_{B} T}{g^{2} \mu_{B}^{2}} \chi^{\Delta z}(\vec{Q}) = \int_{-\infty}^{\infty} \frac{1 - \exp(-\hbar \omega/k_{B} T)}{\hbar \omega/k_{B} T} \tilde{G}^{a}_{n}(\vec{Q}, \omega),
\]

(2.24)

where \( k_{B} \) is the Boltzmann constant, \( g \) is the effective g-value of the magnetic ions and \( \mu_{B} \) is the Bohr magneton. Eq. (2.24) reduces to

\[
\frac{k_{B} T}{g^{2} \mu_{B}^{2}} \chi^{\Delta z}(\vec{Q}) = \tilde{G}^{a}_{n}(\vec{Q})
\]

(2.25)

under the condition

\[
\hbar \omega \ll k_{B} T,
\]

(2.26)

which is known as the quasi-elastic approximation.

**Critical singularities**

The double-logarithmic plots in Fig. 2.6 show some experimental results for the inverse correlation range \( \kappa^{d} \) and the staggered susceptibility \( \chi_{st}^{d} \) as a function of temperature, determined in the tetragonal antiferromagnet MnF\(_{2}\) both above and below \( T_{C} \) \( [19,20] \). Here, the parallel signs refer to the fluctuations of the longitudinal spin components, i.e., the components along the easy axis. A part of the data has been determined from a complete analysis of the inelastic scattering cross section (2.10), whereas another part has been obtained from the quasi-elastic scattering cross section (2.19). The absence of systematic deviations indicates that the quasi-static approximation (2.17) holds in the latter analysis.
Fig. 2.6.
Longitudinal staggered susceptibility $\chi_{st}$ and the inverse correlation range $k^\parallel$ vs. $|T-T_c|/T_c$, observed in MnF$_2$, both above and below $T_c$. Solid lines correspond to the optimum fits of eqs. (2.27) and (2.28) to the data, obtained with the parameters indicated. $\chi_{st}^\parallel$ is normalized on the nonordering static susceptibility $\chi^0$ and $k^\parallel$ is normalized on $a_{nn} = \text{the nearest-neighbours distance}$. (Taken from [19,20])

The temperature dependences of $1/\kappa$ and $\chi_{st}$ appear to be well described by so-called single-power laws [1,13] near $T_c$

$$\frac{1}{\kappa} = \xi = \ell_0 |t|^{-\nu}$$

and

$$\chi_{st} = |t|^{-\gamma},$$

where we introduced the reduced temperature

$$t = T/T_c - 1.$$

In (2.27) and (2.28) $\ell_0$ and $\Gamma$ are called critical amplitudes, $\nu$ and $\gamma$ are so-called critical exponents. (2.27) implies a divergence of the
correlation length $\xi = 1/L$ in the net correlation function (2.21) on approaching the critical point, as we anticipated at the beginning of this section. $\chi_{st}$ shows a similar critical singularity. The power laws (2.27) and (2.28) hold both for $T > T_c$ and for $T < T_c$, as is shown by the straight lines in the double-logarithmic plots of Fig. 2.6.

Henceforth we will distinguish quantities for $T < T_c$ by primed symbols.

In addition to $\xi$ and $\chi_{st}$, many other quantities appear to become singular at $T_c$ and to follow a similar power-law behaviour close to $T_c$. The most common singularities are tabulated in Table 2.1., both for a ferromagnet and for an antiferromagnet. From this table it can be seen that the role of $\chi_{st}$ and $\chi$ (and also of $M_{st}$ and $M$) in an antiferromagnet are interchanged in a ferromagnet. We mentioned in section 2.2, that the critical behaviour of a quantity depends on the path of approach to the critical point $|t|$. This can be seen in Table 2.1., comparing the vanishing of the order parameter as a function of $t$ (or any other relevant nonordering field) and as a function of the ordering field. The power laws for the nonordering susceptibility and for the specific heat show the same critical exponent $\alpha$, as both quantities correspond to a derivative of a nonordering density with

<table>
<thead>
<tr>
<th>quantity</th>
<th>singularity + path of approach</th>
<th>ferromagnet</th>
<th>antiferromagnet</th>
</tr>
</thead>
<tbody>
<tr>
<td>correlation length</td>
<td></td>
<td>$\xi(T) = \xi_o</td>
<td>t</td>
</tr>
<tr>
<td>order parameter</td>
<td></td>
<td>$M(T)/M(0) = B</td>
<td>t</td>
</tr>
<tr>
<td>ordering susceptibility</td>
<td></td>
<td>$\chi(T) = \Gamma</td>
<td>t</td>
</tr>
<tr>
<td>specific heat</td>
<td></td>
<td>$C_H(T) = A</td>
<td>t</td>
</tr>
<tr>
<td>nonordering susceptibility</td>
<td></td>
<td>$\chi_{st}(T) = C</td>
<td>t</td>
</tr>
<tr>
<td>order parameter</td>
<td></td>
<td>$</td>
<td>M(H)</td>
</tr>
</tbody>
</table>

Table 2.1. Summary of definitions of critical amplitudes and critical exponents for several singular quantities in magnetic systems $t = T/T_c - 1$. 

section 2.2, that the critical behaviour of a quantity depends on the path of approach to the critical point $|t|$. This can be seen in Table 2.1., comparing the vanishing of the order parameter as a function of $t$ (or any other relevant nonordering field) and as a function of the ordering field. The power laws for the nonordering susceptibility and for the specific heat show the same critical exponent $\alpha$, as both quantities correspond to a derivative of a nonordering density with
respect to the conjugated nonordering field. In the next sections we shall show that all critical singularities are a direct consequence of the divergence of the correlation length $\xi$ at $T_c$.

### 2.4. Universality

In the description of physical phenomena, one often starts from the implicit assumption that the problem contains a minimum length $L$, which is characterized by the following facts:

(a) the length scale of the physical phenomena of interest is much larger than $L$,

(b) the form of the equations and the parameters in the equations describing the physical phenomena are defined with respect to $L$,

(c) these parameters summarize the relevant information concerning motions over a scale smaller than $L$.

One can give many examples of the above statement \[^8\]. For instance, in atomic phenomena the scale of interest is the atomic size, which is much larger than the nuclear size, i.e., $L \sim$ nuclear size $\sim$ a few fermis. In the Schrödinger equation for the electrons parameters are contained which depend on the total nuclear charge and moments. These parameters represent the total effect of the nucleus on the electrons. The motion of each specific nucleon over a scale much less than $L$ and the specific details of the interactions between different nucleons are not of interest. A second example is the sound propagation in a gas of these atoms. The relevant lengths are much larger than the mean free path of the atoms. Thus we have $L \sim$ a few mean free paths $\sim$ microns. In the sound-wave equation parameters appear which contain the compressibility and viscosity. These parameters can be calculated by studying the motion of atoms over scales less than a few mean free paths.

If one wants to give a description of a (magnetic) system reaching a long-range ordered state, one must study the critical fluctuations, described in the previous section. The scale of interest for a description of critical phenomena is of the order of the correlation length $\xi$ and largely exceeds the microscopic distances. Therefore, it
should be possible to define also a minimum length $L$ with the above properties (a) - (c) for the description of critical phenomena. $L$ should be much smaller than $\xi$ and larger than the interatomic distances. Then, it should be possible to give a description of the critical phenomena in equations which are defined with respect to $L$. The parameters in these equations should represent the total effect of all processes and interactions which take place over a scale smaller than $L$. In sections 2.6. and 2.7. we shall show that this is the very approach of the Renormalization-Group technique in describing and calculating critical behaviour. Here we want to emphasize a direct and important consequence of the above statement.

If the exact form of the microscopic interactions between the correlated particles (magnetic moments) is not quite important in the description of critical phenomena, many different systems must behave in the same way close to criticality. This is observed indeed in a variety of experiments and calculations [1,2,4,5]. It appears that only some very general characteristics of the system are important, so that critical systems can be divided in a few so-called universality classes. Systems within the same universality class show identical critical exponents and very similar equations of state. This distinction of universality classes is the content of the universality hypothesis, which states:

The universality class of a critical system with only short-ranged interactions is determined uniquely by:
1. the spatial dimensionality $d$ of the system,
2. the number of independent vector components $n$ of the order parameter.

In antiferromagnetic systems the effect of long-ranged interactions (such as dipole-dipole interactions) is unimportant because of the alternating sign of the magnetic moments. Therefore, all possible antiferromagnetic systems can be collected in an n-d phase diagram. This is shown in Fig. 2.7., where for $1 \leq d \leq 4$ the various physically significant cases are indicated. Systems in which 1, 2 and 3 spin components take part in the ordering process are better known as 'sing, XY and Heisenberg systems, respectively. In addition to certain
magnetic systems, also superfluid helium and liquid $^3\text{He}-^4\text{He}$ mixtures are described by $n=2$. Normal fluids, fluid mixtures and alloys correspond to $n=1$ [1,6]. The case $n=0$ appears to describe the statistics of polymer chains in a solution [21].

![Diagram of the $(n,d)$-plane, showing various physically relevant types of systems. Heavy solid lines correspond to systems of which the critical behaviour has been solved exactly. O are the non-ordering chain systems. The $d=2$ XY and $d=2$ Heisenberg model possibly display a quasi-ordering. The squares indicate the common systems which order in three dimensions. Their critical behaviour is approximately described by methods using expansions in $\epsilon = 4-d$ and/or $1/n$. In Fig. 2.7. the heavy solid lines indicate systems for which exact solutions of the critical behaviour are available. The only realistic system that has been solved exactly at this moment is the $d=2$ Ising model in zero field [22]. The classical or mean-field model appears to apply to all systems with dimensionality $d\geq 4$. This result will be further discussed in section 2.7. and is important as a starting point of approximative RG-calculations (the so-called $\epsilon$-expansion),]
to find solutions for systems with small \( e = 4 - d \). A similar role is played by the exact solution for \( \Phi^4 \), the so-called spherical model, which is the point of departure for the \( 1/n \)-expansion. Thus, approximate solutions in terms of small \( 1/n \) are sought for more realistic problems. The Gaussian model for \( n = -2 \) corresponds to a merely formal solution of the mean-field model, which appears to be also exactly solvable for all \( d \) if one substitutes \( n = -2 \). To our knowledge this model has no physical significance. The one-dimensional lattices (\( d = 1 \)) have been solved for all \( n \) [23] and it is found that they show no LRO for \( T^c \neq 0 \). For the common \( d = 3 \) systems no exact solutions are available, but good approximative descriptions for the critical behaviour of many quantities have been obtained from various series expansions [24].

A summary of the critical-exponent values for several universality classes is given in Table 2.2. The predictions for the various

Table 2.2. Summary of critical-exponent values for several universality classes. The exponents are defined in Table 2.1.

<table>
<thead>
<tr>
<th>( d )</th>
<th>( n )</th>
<th>( v )</th>
<th>( z )</th>
<th>( \nu )</th>
<th>( \gamma )</th>
<th>( \beta )</th>
<th>( \alpha )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>1</td>
<td>0</td>
<td>( 1/8 )</td>
<td>( 1/4 )</td>
<td>15</td>
<td>1</td>
<td>( 1/4 )</td>
</tr>
<tr>
<td>2</td>
<td>-1</td>
<td>( 1/8 )</td>
<td>0.303(8)</td>
<td>0.318(8)</td>
<td>1.230(1)</td>
<td>5.00(5)</td>
<td>0.640(3)</td>
</tr>
<tr>
<td>2</td>
<td>1/2</td>
<td>0.110(8)</td>
<td>0.321(1)</td>
<td>1.260(4)</td>
<td>4.82(8)</td>
<td>0.830(1)</td>
<td>0.031(1)</td>
</tr>
<tr>
<td>2</td>
<td>-1/2</td>
<td>-0.021(7)</td>
<td>0.368(7)</td>
<td>1.318(10)</td>
<td>4.77(6)</td>
<td>0.879(4)</td>
<td>0.041(1)</td>
</tr>
<tr>
<td>2</td>
<td>-1</td>
<td>0.007(9)</td>
<td>0.346(9)</td>
<td>1.318(9)</td>
<td>4.81(8)</td>
<td>0.869(3)</td>
<td>0.030(1)</td>
</tr>
<tr>
<td>2</td>
<td>1/2</td>
<td>-0.14(8)</td>
<td>0.373(14)</td>
<td>1.405(20)</td>
<td>4.94(6)</td>
<td>0.771(7)</td>
<td>0.040(8)</td>
</tr>
<tr>
<td>2</td>
<td>-1/2</td>
<td>-0.15(15)</td>
<td>0.362(12)</td>
<td>1.39(11)</td>
<td>4.82(12)</td>
<td>0.705(5)</td>
<td>0.031(12)</td>
</tr>
<tr>
<td>2+d=4</td>
<td></td>
<td>( (d+2)/(d-2) )</td>
<td>( 1/2 )</td>
<td>( 1/(d-2) )</td>
<td>( 1/(d-2) )</td>
<td>( 1/(d-2) )</td>
<td>( 1/(d-2) )</td>
</tr>
<tr>
<td>2+d=4</td>
<td></td>
<td>( (d+2)/(d-2) )</td>
<td>( 1/2 )</td>
<td>( 1/(d-2) )</td>
<td>( 1/(d-2) )</td>
<td>( 1/(d-2) )</td>
<td>( 1/(d-2) )</td>
</tr>
<tr>
<td>2+d=4</td>
<td>arbitr.</td>
<td>0</td>
<td>( 1/2 )</td>
<td>3</td>
<td>( 1/2 )</td>
<td>0</td>
<td>( 1/2 )</td>
</tr>
</tbody>
</table>

universality classes have been confirmed by experiments on a large number of systems, both magnetic and nonmagnetic. The critical exponents appear to fulfill certain relations such as

\[
\alpha + 2\beta + \gamma = 2
\]

\[
\alpha + \beta(5+1) = 2
\]

\[
dv + \alpha = 2
\]

\[
2(\nu - 2) = \gamma
\]

\( (2-n)v = \gamma \)

\( (2-n)v = \gamma \)

* This relation follows directly from the definition of \( \chi \) in eqs. (2.20) and (2.25).
and many others \cite{1}. As we shall show in the next section, these relations are a direct consequence of the asymptotic scaling invariance of a system near criticality. In addition, several relations between critical amplitudes appear to be universal \cite{30,31}. For three universality classes the values of these ratios are summarized in Table 2.3.

Table 2.3. Summary of critical-amplitude ratios (taken from \cite{50}).

\[ R_{\alpha} = \frac{\alpha}{T} \left( \frac{\delta M(0)}{\delta \epsilon} \right)^{\alpha-1} \quad \text{and} \quad R_{\beta} = AT \left( \frac{\delta M(0)}{\delta \epsilon} \right)^{2} . \]

<table>
<thead>
<tr>
<th>d</th>
<th>n</th>
<th>A/A'</th>
<th>T/T'</th>
<th>R_c</th>
<th>R_x</th>
<th>method</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>1</td>
<td>1</td>
<td>37.69</td>
<td>6.78</td>
<td>0.319</td>
<td>exact</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>0.51</td>
<td>5.07</td>
<td>0.059</td>
<td>1.75</td>
<td>series</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.55</td>
<td>4.80</td>
<td>0.066</td>
<td>1.6</td>
<td>e-exp.</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>1.52</td>
<td>-</td>
<td>0.165</td>
<td>1.23</td>
<td>series</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.36</td>
<td>-</td>
<td>0.17</td>
<td>1.33</td>
<td>e-exp.</td>
</tr>
</tbody>
</table>

2.5. Scaling

So far, our visualization of critical fluctuations is like Fig. 2.8. Droplets, correlated regions of all sizes up to a maximum size $\xi$, appear near the critical point. However, this picture is incomplete. As each fluctuating region of size $\xi$ is also a nearly critical system, fluctuations will appear within these droplets and within these fluctuations yet more appear (see Fig. 2.9.). This clustering of droplets into droplets continues until the scale of microscopic distances is reached. From this picture one may conclude that critical
phenomena are related to fluctuations over all length scales between \( \xi \) and the microscopic distance between particles. (cited from L.P. Kadanoff [32]).

In other words, a system close to the critical point is scaling invariant within the limits set by the distance between particles and \( \xi \).

When observing the fluctuations in such a system through a microscope, one can decrease the resolving power with a factor \( s > 1 \) and the same image will appear. This procedure can be repeated until ultimately the maximum size \( \xi \) of the correlated regions becomes apparent. This observation implies that \( \xi \) must be the only significant length in the description of critical fluctuations. Other lengths, such as interatomic distances are too short to play a role.

A subsequent unavoidable conclusion is that the behaviour of the critical fluctuations in the order parameter on approaching the critical point, may be considered as being due to a simple change of length scale. Since many critical quantities are in turn directly related to these critical fluctuations in the order parameter, a scaling hypothesis may be formulated which states [8]:

"The behaviour of any physical quantity near a critical point can be deduced from the way in which it varies under a change in length scale. The divergence of the only significant length at the critical point accounts for the singular critical behaviour of all other quantities".

The simplest example of this idea is the following. Consider a \( d \)-dimensional system with an energy per unit volume \( \mathcal{E}(L) \). \( L \) is a characteristic minimum length of the system. If the unit length is enlarged
with a factor $s$, the new energy density becomes $s^d$ times as large. Simultaneously the numerical value of the length $L$ decreases with a factor $s$. So, under a change in length scale $G$ varies as

$$\tilde{G}(L/s) = s^d \tilde{G}(L). \quad (2.31)$$

Since $s$ is taken arbitrary, eq. (2.31) defines $\tilde{G}$ as a homogeneous function of $L \mid 1\mid$. This implies that $\tilde{G}$ is known over the whole range of its argument, if it is known in one point. It can be verified that this applies to $\tilde{G}$ in (2.31), by choosing $s=L$. The result is

$$\tilde{G}(L) = L^{-d} \tilde{G}(1), \quad (2.32)$$

which establishes the dependence of $\tilde{G}$ on the minimum length $L$. According to the scaling hypothesis this functional form of $G$ should hold close to a critical point.

With the correlation length $\xi$ as the only relevant length in a critical system, we define our characteristic minimum length $L$ as $L = \xi/M$, with $M$ a large number. Then we can write the variation of $G$ with temperature as

$$G(T) \equiv \tilde{G}[L(T)] = \tilde{G}[\xi(T)/M] = M^d(\xi(T))^{-d} \tilde{G}(1). \quad (2.33)$$

This result shows that the critical behaviour of $G$ is determined by the critical behaviour of $\xi$. For instance, in an antiferromagnet at $H_{st} = 0$, one has $\xi \propto t^{-\nu}$ for $t > 0$ and $\xi \propto |t|^{-\nu'}$ for $t < 0$, and (2.33) becomes

$$G(T) \propto t^{\nu d}, \text{ for } H_{st} = 0 \text{ and } t > 0$$

and

$$G(T) \propto |t|^{\nu' d}, \text{ for } H_{st} = 0 \text{ and } t < 0. \quad (2.34)$$
G as a generalized homogeneous function

In the above introductory example we used an energy density with argument L to elucidate the idea. Now we want to derive the functional form which is imposed by the scaling hypothesis on the more familiar Gibbs free energy of, for instance, an antiferromagnet: \( G(H_{st}, H, T) \). As we shall see, the procedure is straightforward but demands a very careful handling of the definitions of the various fields, which we introduced in section 2.2.

Let us start from a critical point \( CP \), located at \( [(H_{st})_{c}, H_{c}, T_{c}] \). As we have discussed in section 2.3., \( CP \) can be characterized as a point in \( (H_{st}, H, T) \)-space where the correlation length \( \xi \) diverges, irrespective of the path of approach to \( CP \). First we shall choose so-called optimum scaling fields for the description of the critical behaviour near \( CP \). The first optimum scaling field \( h \) is identified with the field which yields the strongest divergence of \( \xi \) close to \( CP \) and is represented by

\[
\xi(h) = \xi_{oh} h^{-\alpha_h}.
\]

(2.35)

Along any path of approach to \( CP \) which has a component along the \( h \)-axis, the divergence of \( \xi \) close enough to \( CP \) will be described by (2.35).

In the plane perpendicular to the \( h \)-axis, we can search for the second optimum scaling axis \( t \), which yields the strongest divergence of \( \xi \), for \( h=0 \). This divergence will be described by

\[
\xi(t) = \xi_{ot} t^{-\alpha_t}.
\]

(2.36)

Again, it can be noted that the divergence of \( \xi \) is correctly described by (2.36) along any path in the \( h=0 \) plane, which has a component along the \( t \)-axis. Similarly, a third optimum scaling axis \( g \) is introduced, along which the divergence of \( \xi \) is given as

\[
\xi(g) = \xi_{og} g^{-\alpha_g}.
\]

(2.37)

Expression (2.37) applies only for \( h=t=0 \).
No new aspects are added when more fields are taken into account. It was pointed out by Griffiths and Wheeler [12], that one needs three and only three types of fields to obtain a complete description of the critical behaviour near CP. In the following we shall confine ourselves to this three-dimensional $(h,g,t)$-space. How the $h$, $g$ and $t$ axes are directed in the $(H,H,T)$-space will not be indicated. We shall treat the general case in which all three types of fields can be distinguished, with corresponding exponents $a_H > a_T > a_G$. After Griffiths and Wheeler, $h, t$ and $g$ can be identified with the optimum choice of the ordering field, relevant nonordering field and irrelevant field, respectively. Now the behaviour of $\xi(h,g,t)$ close to CP may be represented as the sum of three power laws

$$\xi(h,g,t) = \xi_{oh} h^{-a_H} + \xi_{ot} t^{-a_T} + \xi_{og} g^{-a_G}.$$  

(2.38)

When the path of approach contains all three field components, the strongest divergence (2.35) will ultimately win and describe the behaviour of $\xi$ correctly close to CP. For $h=0$ (2.36) takes over and for $h=t=0$ the divergence of $\xi$ is described by (2.37).

According to the scaling hypothesis, the behaviour of any quantity, $G(h,g,t)$ for instance, is determined by the behaviour of $\xi$ only. Therefore we may write

$$G(h,g,t) = \frac{G(\xi(h,g,t)/\xi)}{M} = \xi_{oh} h^{-a_H} + \xi_{ot} t^{-a_T} + \xi_{og} g^{-a_G}. \quad (2.39)$$

Combining (2.31) and (2.39) we can write

$$G(h,g,t) = \frac{G(\xi(h,g,t)/M)}{s} = s^{-d} \frac{G(\xi(h,g,t)/M)}{s^1/a_H} + \xi_{ot} (ts^{1/a_T})^{-a_T} + \xi_{og} (gs^{1/a_G})^{-a_G}. \quad (2.40)$$

According to (2.39) this is identical with

$$G(h,g,t) = s^{-d} G(hs^{1/a_H}, gs^{1/a_G}, ts^{1/a_T}). \quad (2.41)$$
With $s$ an arbitrary number, (2.41) defines $G(h,g,t)$ as a so-called generalized homogeneous function $|1|$. The analogue of (2.32) can also be obtained by choosing $s = t^{\frac{a_T}{a_H}}$ in (2.41):

$$G(h,g,t) = t^{\frac{a_T d}{a_H}} G\left(\frac{h}{a_H}, \frac{g}{a_G}, 1\right) = t^{\frac{a_T d}{a_H}} f\left(\frac{h}{a_H}, \frac{g}{a_G}\right). \quad (2.42)$$

Both notations (2.41) and (2.42) are equivalent $|1|$ and are commonly used as alternative, more mathematical definitions of the scaling hypothesis.

**Exponent relations and scaling functions**

The generalized-homogeneous function formulation of the scaling hypothesis, (2.41) or equivalently (2.42), appears to be a very powerful tool in the description of critical phenomena $[33]$. For instance, it implies that all derivatives of $G$ with respect to the fields, i.e. all densities in the formulation of Eq. (2.2), are generalized homogeneous functions too. This can be verified by repeated differentiation of (2.41). The order parameter $m$, for instance, is obtained as

$$m(h,g,t) = -\frac{\partial u}{\partial h} g_t = s^{(1/a_H - d)} m(h s^{1/a_H}, g s^{1/a_G}, t s^{1/a_T}). \quad (2.43)$$

The ordering susceptibility $\chi$ follows as

$$\chi(h,g,t) = -\frac{\partial^2 G}{\partial h^2} g_t = s^{(2/a_H - d)} \chi(h s^{1/a_H}, g s^{1/a_G}, t s^{1/a_T}). \quad (2.44)$$

The specific heat is found from (2.41), with $t$ identified with $T-T_c$, as

$$C(h,g,t) = \frac{\partial^2 G}{\partial t^2} h_g = s^{(2/a_T - d)} C(h s^{1/a_H}, g s^{1/a_G}, t s^{1/a_T}). \quad (2.45)$$
A second important consequence of the scaling hypothesis (2.41) is that it leads to the experimentally observed exponent relations (2.30). This can be demonstrated as follows. Let us take an antiferromagnet as an example and identify $H_{st}$ as the ordering field $h$, $(T-T_c)/T_c$ as the relevant nonordering field $t$, and $H-H_c$ as an irrelevant nonordering field $g$. From (2.43) we can derive the power law

$$m(h) = s^{(1/a_H-d)}m(h)^{1/a_H^d}, \quad \text{for } g=t=n$$

Using $h = H_{st}$ and $m = M_{st}$, this yields with $s = |H_{st}|^{-a_H}$ substituted:

$$M_{st}(H_{st}) = |H_{st}|^{(da_H-1)}M_{st}(1) = D|H_{st}|^{1/\delta}$$

(2.47)

at $H = H_c$, $T = T_c$. The last equality follows from the usual definition of this power law, as presented in Table 2.1. From (2.47) we find

$$\delta = \frac{1}{da_H-1}$$

(2.48)

From (2.43) with $s = |t|^{-a_T}$ substituted, we obtain at $H = H_c$, $H_{st} = 0$ the power law

$$M_{st}(t) = |t|^{a_T(d-1/a_H)}M_{st}(|t|=1) = B|t|^\gamma$$

(2.49)

This gives for the exponent $\gamma$ the result

$$\gamma = \frac{a_T(d-1/a_H)}{d-2/a_H}$$

(2.50)

Similarly we derive from (2.44) with $s = |t|^{-a_T}$

$$\gamma = \frac{a_T(d-2/a_H)}{d-2/a_H}$$

(2.51)

Finally, for $a$ we find from (2.45) with $s = |t|^{-a_T}$ the relation

$$a = 2-\frac{d}{a_T}$$

(2.52)

From (2.36) we can identify
As all exponents are functions of $a_H$, $a_T$ and $d$, there must exist many relations between the exponents. It can be easily verified by means of (2.48) - (2.53) that among others the first three relations in (2.30) are fulfilled.

Strong limitations are imposed by the scaling hypothesis on, for instance, the functional form of the equation of state. In a ferromagnet (with $h=H$, $g=H_{st}=0$) this can be derived from (2.43), taking $s = (H)^{-a_H}$, viz.

$$M(H,t) = H^{(d-1)/a_H} M(1, t^{-a_T/a_H})$$

$$\equiv H^{1/6} \mathcal{J}(\frac{t}{H^{a_H/a_T}}) .$$

Here, $\mathcal{J}(y)$ is a so-called scaling function, which depends on one single variable

$$y = t \cdot H^{a_H/a_T}$$

(2.54)

(2.55)

and not on $t$ and $H$, separately. An alternative form is obtained by substituting $s = (t)^{-a_T}$ in (2.43), viz.

$$M(H,t) = (t)^{a_T(d-1/a_H)} M(\frac{H}{t^{a_T/a_H}}, 1)$$

$$\equiv t^{1/6} \mathcal{K}(\frac{H}{t^{a_T/a_H}}) .$$

(2.56)

(2.57)

Again, $\mathcal{K}(x)$ is a scaling function, depending on one single variable

$$x = H \cdot t^{1/a_H} .$$

(2.57)

Such relations have been verified in a number of experiments. Some beautiful examples are shown in [1]. It must be noticed that the
scaling hypothesis (2.41) does not predict the values of the exponents, nor the exact shape of the scaling functions.

Concluding remarks

In the sections 2.4. and 2.5. we introduced two important aspects of critical phenomena, viz.

1. Critical fluctuations show an infinitely increasing correlation length on approaching the critical point. Therefore, a length \( L \) can be indicated which is much smaller than \( \xi \) and yet much larger than the interatomic distances \( a_{\text{nn}} \). It should be possible to describe critical phenomena with equations, in which the parameters contain the integrated effects of all processes on length scales smaller than \( L \).

2. A system close to a critical point is scaling invariant between limits which are set by \( a_{\text{nn}} \) on one hand and \( \xi \) on the other hand. The critical behaviour of any quantity may be considered to be due to a change in \( \xi \). Therefore the result of any effect working on a critical system, may be considered as merely a transformation of length scale.

These observations indicate the procedure by which one possibly can perform real calculations of critical phenomena.

In the first place one can try to formulate an effective Hamiltonian \( \mathcal{H} \), defined with respect to a (minimum) length \( L \), in which all irrelevant details (i.e. with a length scale \( \gg L \)) are contained in the parameters. \( \mathcal{H} \) must describe the critical fluctuations on a scale \( \gg L \).

Following the above observation 2., we can study how \( \mathcal{H} \) transforms into an effective Hamiltonian \( \mathcal{H}' \) under a change in length scale. It would be very attractive to find an \( \mathcal{H}' \) with a form similar to the original \( \mathcal{H} \), so that the corresponding parameters can be related to each other. Repeating this transformation process, one can try to obtain recursion relations for these parameters. At least this should be possible for a system at a critical point where, according to the scaling hypothesis, the system is expected to be scaling invariant. Then one may also hope to obtain useful information about a system.
near a critical point, by studying effective Hamiltonians which closely resemble the critical one. The sketched procedure is the approach chosen in the Renormalization-Group (RG) technique for the calculation of critical behaviour.

In the next two sections (2.6. and 2.7.) we shall present a more detailed description of the RG-approach. It must be emphasized that this part may be skipped by readers who are interested mainly in the experimental part of this work.
2.6. The calculation of critical behaviour

In this section we shall start with a more elaborate discussion of the various steps involved in the RG-approach. As a point of departure we use the well known spin-block picture of Kadanoff [1,34], as was done by Wilson in one of his early presentations of the RG-approach [35]. By means of this model the concept of effective Hamiltonians will be elucidated. Next we shall indicate the steps to be performed in the calculation of the RG-transformation. We shall not actually perform these steps, but only show the procedure on basis of the Kadanoff picture. The calculation will be performed for an exactly solvable model in section 2.7. There, the RG-approach in the calculation of critical behaviour will be treated.

The Kadanoff picture

The prescription, how to remove unimportant details from the calculation of large-scale effects, is trivial [8]. Let \( P(y_1, y_2, y_3) \) be the probability distribution function for the random variables \(-\infty < y_1, y_2, y_3 < \infty\). To calculate the average value of any function \( f(y_1, y_2, y_3) \) of these variables we evaluate the integral

\[
I = \int dy_1 dy_2 dy_3 f(y_1, y_2, y_3) P(y_1, y_2, y_3) .
\]

(2.53)

For a function \( f' \) which does not depend on \( y_1 \), we can define an equivalent distribution function \( P'(y_2, y_3) \)

\[
P'(y_2, y_3) = \int_{-\infty}^{\infty} dy_1 P(y_1, y_2, y_3) .
\]

(2.59)

Then we can calculate the average value of \( f' \) as

\[
I' = \int dy_2 dy_3 f'(y_2, y_3) P'(y_2, y_3) .
\]

(2.60)

For the problem of critical fluctuations, this procedure implies that one has to formulate a probability function in which the total effect of all small-scale details are incorporated.
Fig. 2.10. The spin-block construction of Kadanoff, in which a number of Ising spins $S$ are taken together in a block to give a block spin $S'$. 

Consider a ferromagnetic system of $N$ Ising spins on a simple cubic lattice with exchange interactions between nearest neighbours. The Hamiltonian of this system is given by 

$$\mathcal{H} = -2 \sum_{\langle i,j \rangle} J (\vec{R}_i - \vec{R}_j) S_i S_j \tag{2.61}$$

where $S_i$ denotes the spin at site $\vec{R}_i$; $\langle i,j \rangle$ denotes the summation over all nearest-neighbour pairs of spins. We shall use normalized Ising spins with values $\pm 1$. $J$ is clearly defined with respect to a length scale $L = a_{nn}$, the lattice constant. Near a critical point, we are only interested in spin fluctuations over distances much longer than $a_{nn}$. Then we can divide the lattice into $N' = N/s^d$ blocks of side $L = a_{nn}$ (Fig. 2.10) and try to derive a Hamiltonian $\mathcal{H}'$ of the form 

$$\mathcal{H}' = -2 \sum_{\langle i',j' \rangle} J' (\vec{R}'_i - \vec{R}'_j) S'_i S'_j \tag{2.62}$$

which describes the interaction between the block spins $S'$. $\langle i',j' \rangle$ denotes the summation over all pairs of block spins. The details of processes inside the blocks must be contained in $J'$. 

In practice, the transformation of $\mathcal{H}$ into $\mathcal{H}'$ is not straightforward. For instance, the Ising spins in (2.61) have values $\pm 1$, whereas the block spins $S'$ in (2.62) will only have a similar two-fold spin value at $T = 0$, where all spins are perfectly ordered. However, close to $T_c$
Spin fluctuations exist over all length scales up to $\xi$, and we can expect block spins with values varying from $-s^d$ to $s^d$ with an average value $0$. Fortunately the distribution of spin values in blocks with, say, sides 10 $a_{nn}$ or 50 $a_{nn}$ or 350 $a_{nn}$ will be less different. When the spins would have no interactions at all this distribution function would undoubtedly be a Gaussian. Otherwise, it will have a more general bell-shaped form, such as

$$D(S') = \exp(-\nu S'^2 + \omega S'^4 + \ldots).$$

(2.63)

$D(S')$ is expected to be even in $S'$, to reach a maximum at $S' = 0$ and to be integrable.

In the case of the Ising system (2.61) the probability function for the calculation of average values for any quantity is related to the partition function

$$Z = \sum_{S=-1}^{+1} \sum_{S'=-1}^{+1} \exp(-\beta S^2 B_T).$$

(2.64)

$Z$ is obtained by weighting each state by the appropriate Boltzmann factor calculated from (2.61), and the summing over all states of the system. Anticipating the following step, we can rewrite (2.64) as

$$Z = \int ds_1 \ldots \int ds_N \prod_{i=1}^N \{ \prod_{j=1}^{N'} D(S_{ij}) \} \exp(-\beta S^2 B_T)$$

(2.65)

with

$$D(S_{ij}) = \delta(S_{ij}^2 - 1),$$

(2.66)

and $\int ds_1 \ldots \int ds_N$ denoting that the integral has to be performed for all $N$ spins $S_i$. The kernel in (2.65) can be identified with the probability function $P$ in (2.58).

Similarly we can introduce a partition function for the spin-block system as

$$Z' = \int ds_1^1 \ldots \int ds_N^1 \prod_{i=1}^{N'} \{ \prod_{j=1}^{N'} D(S_{ij}') \} \exp(-\beta S^2 B_T),$$

(2.67)
where $\mathcal{H}'$ is the desired new Hamiltonian describing the interactions between block spins. With the spin-distribution function $D(S_i')$ of (2.63) we can write

$$Z' = \int \prod_{i=1}^{N'} dS_i' \exp \left[ -\mathcal{H}' - \sum_{i=1}^{N'} [\nu(S_i')^2 - u(S_i')^4 + \ldots] \right]$$

$$= \int \prod_{i=1}^{N'} dS_i' \exp \left[ -\mathcal{H}' \right]. \quad (2.68)$$

The overlined $\mathcal{H}$ indicates a so-called reduced Hamiltonian. From its definition

$$\mathcal{H} = -\frac{1}{k_B T} + \ln D(S_i)$$

it can be seen that $\mathcal{H}$ is the combination of an energy term and an entropy term. In (2.68) $\exp [\mathcal{H}']$ corresponds to the new probability function $P'$ in (2.59), which we are looking for. $Z'$ is the wanted partition function in terms of variables which are significant for the spin-block system, viz. the block spins $S_i'$. With (2.68) one can calculate, for instance, the two block spin correlation function as

$$\langle S_i' S_j' \rangle = \frac{1}{Z'} \int \prod_{i=1}^{N'} dS_i' (S_i' S_j') \exp [\mathcal{H}'], \quad (2.69)$$

which is the analogue of (2.60). In the following subsection, we shall show how one can arrive from one $\mathcal{H}$ to another $\mathcal{H}'$, i.e. how the step (2.59) can be carried out in the spin-block system.

**The calculation of reduced Hamiltonians**

Here we shall illustrate how the effective reduced Hamiltonian $\mathcal{H}'$ in (2.68) can be calculated from the original reduced Hamiltonian $\mathcal{H}$. This is in fact the first step in the iteration process of a RG-calculation. For such calculations it would be ideal if a set of integration variables can be found in the integral (2.68) which factorize the integrand. For example, if $\mathcal{H}' = 0$ the partition function $Z'$ reduces to the product of $N'$ independent single-variable ($S_i'$) integrals. If $u=0$ and higher-order terms in $S_i'$ are absent, then the integrand factorizes in terms of the
Fourier-transformed variables \( S \), defined in (2.14). For this reason the principle of the transformation from \( \mathcal{F}(m) \) to \( \mathcal{F}(m+1) \), where \( (m) \) replaces a number of \( m \) primes, can be most conveniently explained in momentum space.

We can define \( \mathcal{F} \) in momentum space as

\[
\mathcal{F} = -\frac{1}{2} \int_{\mathbf{q}} \mathcal{J}(\mathbf{q}) S_+ S_-, \tag{2.70}
\]

with

\[
\mathcal{J}(\mathbf{q}) = \sum_{\mathbf{R}} \exp(i\mathbf{q}\cdot\mathbf{R}) J(\mathbf{R}). \tag{2.71}
\]

The integration in (2.70) runs over the first Brillouin zone. \( \mathcal{J} \) has been normalized to give the factor \( \frac{1}{2} \) in front of (2.70). For the present discussion we shall anticipate on a result of the iteration process, namely that the magnitude of the spins appears to be an unimportant quantity in the critical behaviour of a magnetic system. This allows us to use a spin-distribution function of the form (2.63) instead of (2.66) already in the first step of the iteration process.\(^1\)

Then we can write the reduced Hamiltonian \( \mathcal{H} \) as

\[
\mathcal{H} = -\frac{\mathcal{F}}{k_B T} - \int_{\mathbf{R}} \exp(i\mathbf{q}\cdot\mathbf{R}) \left[ \frac{1}{2} S_+^2 - u S_+^4 \right]
= -\frac{1}{2} \int_{\mathbf{q}} \left[ 1 - \mathcal{J}(\mathbf{q})/k_B T \right] S_+ S_-
- \sum_{\mathbf{q}, \mathbf{q}' \mathbf{q}''} \exp\left(-u\mathbf{q}\cdot\mathbf{q}'\right) \epsilon_{\mathbf{q}}^{\mathbf{q}'} \epsilon_{\mathbf{q}'}^{\mathbf{q}''} S_{\mathbf{q}'} S_{\mathbf{q}''} \epsilon_{\mathbf{q}''}^{\mathbf{q}''}. \tag{2.72}
\]

Here \( D(S) \) has been truncated at the quartic term. Now the partition function \( Z \) is written as

\[
Z = \int_{-\infty}^{\infty} dS_+ \exp[\mathcal{H}], \tag{2.73}
\]

where \( \int_{\mathbf{q}} \prod_{\mathbf{q} < \Lambda_0} dS_+ \) indicates that the integration has to be performed over \( \Lambda_0 \).\(^1\)

Moreover, the function \( \exp(-u) \exp\left[u(S_+^2-1)^2\right] \), i.e. (2.63) with \( v = 2u \), closely resembles (2.66), for \( u \to \infty \).
for all $S_q$ with $|\hat{q}|$ smaller than $\Lambda_0$. With $\Lambda_0$ we introduce a cutoff which in (2.73) still coincides with the Brillouin-zone boundary. It will follow from the iteration process that the specific shape of this Brillouin zone is not important at all. Therefore we can simply use a spherical Brillouin zone!

In our spin system the small-scale fluctuations are related to the $S_q$ with large wave vectors $\hat{q}$, i.e. close to $\Lambda$, whereas the large-scale fluctuations are represented by the $S_q$ with small $\hat{q}$ close to the center of the Brillouin zone. This indicates an easy way to eliminate small-scale processes out of our spin-block model and to define an effective reduced Hamiltonian $\tilde{\mathcal{H}}$. Following the guidelines of eqs. (2.59) and (2.60) we define

$$\exp\left[\frac{\mathcal{F}}{\mathcal{Z}}\right] \equiv \int \prod_{\Lambda < \hat{q} < \Lambda_0} dS_q \exp\left[\frac{\mathcal{F}}{\mathcal{Z}}\right]$$ (2.74)

as the new probability function, which contains the total effect of all small-scale processes with wave vectors $\hat{q} > \Lambda$. Here a logical choice for the cutoff is $\Lambda = 2\pi/L$, with $L$ the linear size of the spin block. The new partition function $Z'$ now becomes

$$Z' = \int \prod_{\hat{q} < \Lambda} dS_q \exp\left[\frac{\mathcal{F}}{\mathcal{Z}}\right],$$ (2.75)

which is the equivalent of (2.68). However, in this stage we did not define $\tilde{\mathcal{H}}$.

In the calculation of the expectation value for any quantity which does not depend on the small-scale processes within the spin blocks, we now can use (2.75) instead of (2.73). This means that the number of integrations to be performed is reduced with a factor $(L/a_m)^d = s^d$. This procedure may be repeated: enlarge the linear size of the spin blocks with a factor $s$, reduce the cutoff $\Lambda$ with a factor $s$ and calculate a subsequent reduced Hamiltonian $\tilde{\mathcal{H}}(m)$ and the new partition function $Z(m)$. This process may be identified with the observation of a critical system through a microscope with repeatedly reduced resolving power (compare section 2.5.).

The scaling hypothesis expresses the expectation that in a critical
system the iteration process can be continued without producing essential changes in \( \tilde{H}^{(m)} \), until the block size \( L = 2\pi/\Lambda \) approaches \( \epsilon \).

The variations will take place only in the parameters of \( \tilde{H}^{(m)} \), like \( \tilde{J} \) and \( \tilde{u} \) in (2.72). Finally the situation will be reached, that only a few giant spin blocks remain for which the calculation possibly can be performed. If the variation of the parameters \( \tilde{J}^{(m)} \) and \( \tilde{u}^{(m)} \) can be followed during the iteration process, one can try to reverse the iteration sequence and solve the original many-particle problem. This procedure has been applied recently by Rudnick and Nelson [36], to calculate the equation of state in a critical system.

An alternative interesting possibility occurs if the iteration process ends in an reduced Hamiltonian \( \tilde{H}^{*} \) which remains unchanged under further iterations. This can only occur in a system where the cutoff \( \Lambda \) may become infinitely small, i.e. where the block size may become infinitely large without ever reaching \( \epsilon \). This situation clearly corresponds to a system at a critical point. \( \tilde{H}^{*} \) is called a fixed point and a large amount of work in RG-calculations is spent in the search for fixed points in various systems. It will be shown in the next section that studying the changes due to the iterations in reduced Hamiltonians which slightly differ from \( \tilde{H}^{*} \), yields important numerical predictions about the divergences in a system close to a critical point. For instance, the values of critical exponents can be obtained.

2.7. The Renormalization-Group approach

In section 2.6. we used the Ising model as a starting point for the discussion on effective Hamiltonians. Here we shall proceed with a more general type of Hamiltonian. Many types of physically interesting Hamiltonians can be written as

\[
\tilde{H} = \tilde{H}_X + \tilde{H}_s,i
\]

where \( \tilde{H}_X \) represents "exchange" terms of the form

\[
\tilde{H}_X = -\frac{1}{4} \sum_{\alpha} \sum_{\beta} \sum_{R_i \neq R_j} J^{\alpha\beta}(R_i - R_j) S_\alpha^{R_i} S_\beta^{R_j}
\]

(2.77)
and $s_i$ represents "single-ion" terms of the form

$$
\mathcal{H}_{s_i} = \frac{1}{4} \sum_{m,n} A_i^m s_i^m \cdot s_i^n + \frac{1}{8} B_i^{mn} s_i^m s_i^n s_i^o + \ldots
$$

The most common examples of physically interesting forms of $\mathcal{H}^\infty(q)$ are summarized in Table 2.4. \textsuperscript{37} For $n=3$ the isotropic case corresponds to the Heisenberg model. From the spin-anisotropic type one can arrive at the Ising model ($J_z = J_x = J_y = \delta$) or the XY-model ($J_z = 0, J_x = J_y = \delta$).

<table>
<thead>
<tr>
<th>Type</th>
<th>$J^{AB}(R)$</th>
<th>$V_{i}^{A}(\mathbf{q})$ [to $\sigma(\mathbf{q})$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isotropic</td>
<td>$J(R) \delta_{R}$</td>
<td>$(r+q^2)^{1/2}$</td>
</tr>
<tr>
<td>Spin-anisotropic</td>
<td>$J^3(R) \delta_{R}$</td>
<td>$(r+q^2)^{1/2}$</td>
</tr>
<tr>
<td>Cubic</td>
<td>$J(R^3) \delta_{R}$</td>
<td>$\left[ (r+q^2) + f(q) \right]^{1/2}$</td>
</tr>
<tr>
<td>Isotropic &quot;Dipolar&quot;</td>
<td>$K(R)R^3R^2/R^2$</td>
<td>$-hq_{q}q_{q} + gq_{q}q_{q}$</td>
</tr>
<tr>
<td>Dzyaloshinski-Moriya</td>
<td>$A^{\gamma}(R) \epsilon_{R\gamma}$</td>
<td>$i\gamma q_{q} \epsilon_{R\gamma}$ + constants</td>
</tr>
</tbody>
</table>

With these forms of $\mathcal{H}$ and the commonly used spin-distribution function (2.63) the reduced Hamiltonian $\hat{\mathcal{H}}$ can be expressed in the very general form

$$
\hat{\mathcal{H}} = -\frac{1}{4} \sum_{\alpha \beta} \int_{\mathbf{q}} V^{AB}_{\alpha}(\mathbf{q}) \frac{S_\alpha^A S_\beta^B}{q - q} + \ldots
$$

$$
-\frac{1}{4} \sum_{\alpha \beta \gamma \delta} \int_{\mathbf{q}} V^{AB}_{\alpha}(\mathbf{q}) \frac{S_\alpha^A S_\beta^B S_\gamma^C S_\delta^D}{q - q} + \ldots
$$

where

$$
V_{\gamma}^{AB}(\mathbf{q}) = C\delta_{\alpha \beta} - \left[ J^{10}(\mathbf{q}) - A^{10}\right]/k_B T,
$$
Here we used the generalization of (2.71) for the Fourier transform of 
an anisotropic $J^{AB}(q)$. With the specific choice (2.78) for $S_{\text{S,i}}$ and 
(2.67) for $D$ the quantity $V_{\text{A}}^{AB}\delta$ is a $q$-independent combination of 
$B_{\text{A}}^{AB}\delta$ and $u$. The $q$-dependent terms quartic in $S_{\text{q}}$ are usually unimportant 

In case of short-range exchange interactions, $J^{AB}(q)$ may be expanded 
in powers of $q$,

$$J^{AB}(q) = J^{AB}(0) - jq^2 \delta_{AB} + \sum_{\gamma,\delta} j^{AB}\delta q_{q_{\gamma}}^{\gamma,\delta}(q^\gamma), \quad (2.81)$$

where we introduced $q_a = q_a/A$ and separated the term $q^2 \delta_{AB}$ from the 
sum over $\gamma$ and $\delta$. With (2.81) we find for (2.80) the commonly used 
notation

$$V_{\text{A}}^{AB}(q) = r^{AB} + q^2 \delta_{AB} - \sum_{\gamma,\delta} j^{AB}\delta q_{q_{\gamma}}^{\gamma,\delta}(q^\gamma) \quad (2.82)$$

where

$$r_{AB} = \left[ k_B T \alpha_{AB} - J^{AB}(0) + \Lambda^{AB}\right]/j = k_B (T_0 - T^{AB}_0)/j \quad (2.83)$$

Various examples of the function $V_{\text{A}}^{AB}(q)$ are given in Table 2.4.

In ref. [37] detailed expressions for $V_{\text{A}}^{AB}\delta$ may be found. From (2.83) 
it can be seen how the temperature enters into the calculation. As $T$ 
is lowered, one or more eigenvalues of the matrix $r_{AB}$ may vanish.

It will be seen below, that this leads to criticality at the tempera-
ture $T^{AB}_0$.

The RG-recipe

In the RG-approach one attempts to relate the various constants in the 
$V_{\text{A}}^{AB}$ and $V_{\text{A}}^{AB}\delta$ of two subsequent reduced Hamiltonians $\tilde{\mathcal{H}}(m)$ and $\tilde{\mathcal{H}}(m+1)$, 
calculated from (2.74) in an iteration process where the cutoff $\Lambda$ is 
repeatedly decreased by a factor $s$, i.e. $\Lambda^{(m+1)} = \Lambda^{(m)}/s$. From its 
definition it is obvious that $\tilde{\mathcal{H}}(m)$ is defined with respect to its cut-
toff $\Lambda^{(m)}$. An expression for $\tilde{\mathcal{H}}(m)$ is useful only when the corresponding 
cutoff $\Lambda^{(m)}$ is specified. Therefore, two reduced Hamiltonians can be 
correctly compared only when they have the same cutoff. For a compar-
ison of $\tilde{\mathcal{H}}(m)$ with $\tilde{\mathcal{H}}(m+1)$, a rescaling has to be performed first to
make the numerical value of the cutoff \( \Lambda^{(m+1)} \) equal to \( \Lambda^{(m)} \), by defining a new unit length \( e^{(m+1)}_l = s \cdot e^{(m)}_l \). This has to be done in each iteration cycle. Usually also the spin values are rescaled in order to keep the constant in the \( q^2 \)-term of (2.82) equal to unity.

Summarizing the three steps in a complete cycle of the RG-iteration process, we have:

- \( g^{(m)} \) is transformed in \( g^{(m+1)} \) according to (2.74). The cutoff is reduced as
  \[
  \Lambda^{(m+1)} = \Lambda^{(m)}/s. \tag{2.84}
  \]

The number of degrees of freedom reduces correspondingly as
  \[
  N^{(m+1)} = N^{(m)}/s. \tag{2.85a}
  \]

- As a result of the redefining of the unit length as
  \[
  s e^{(m+1)}_l = s e^{(m)}_l, \tag{2.85b}
  \]

all numerical values of the spatial vectors and momenta are rescaled as

- The spin vectors are rescaled with a factor \( c \) to preserve the numerical constant in the \( q^2 \)-term of (2.82), i.e.
  \[
  q^{(m+1)} = q^{(m)}/c. \tag{2.86}
  \]

With \( g^{(m+1)}_l \) indicating the rescaled \( g^{(m+1)}_l \), the above complete cycle of the iteration process is denoted as

\[
\Lambda^{(m+1)} = \Lambda^{(m)},
\]

where \( \Lambda \) is the symbol of a RG-transformation.

The transformation under \( \Lambda \) of the Gibbs free energy, defined by

\[
G(e^{(m+1)}, g^{(m+1)}) = \lim_{N \to \infty} \frac{1}{N} \ln \{ Z(e^{(m+1)}, g^{(m+1)}) \}
\]
is expressed by the following transformation rule

\[ G^{(m+1)} = s^d G^{(m)} . \tag{2.87} \]

Similarly, one finds for the net spin-correlation function

\[ G_n^{(m+1)} = c^{-2} G_n^{(m)} . \tag{2.88} \]

Now, the iteration process can be started and one can compare the numerical values of the constants in \( f \) and \( X \), and establish recursion relations for the coefficients \( r \), \( j \) and \( V \), as was expressed in Kadanoff's block-spin idea. There is one minor difference between the RG-approach and the Kadanoff picture in section 2.6. In the Kadanoff model one increases the spin blocks in a system of constant magnitude with a given \( \xi \). Here the cutoff is fixed, and \( \xi \) decreases in each cycle as \( \xi^{(m+1)} = \xi^{(m)}/s \) according to (2.85a).

**An exactly solvable model: the classical model**

As an example we consider the reduced Hamiltonian (2.79) containing only the terms bilinear in the spin variables, \( \xi, \gamma \).

\[ \tilde{H} = -\frac{1}{4} \sum_{\alpha, \beta} \int_{q < \Lambda_0} V^{\alpha \beta}(q) S_\alpha^2 S_\beta^2 . \tag{2.89} \]

Here we shall confine ourselves to an isotropic system, where \( \tilde{J}(q) = \tilde{J}_{\alpha \beta} J(q^2) \) and hence we obtain:

\[ \tilde{H} = -\frac{1}{4} \sum_{\alpha} \int_{q < \Lambda_0} (r + q^2) \left( S_\alpha^2 \right)^2 , \tag{2.90} \]

with (cf. (2.83))

\[ r = k_B (T - T_0) / j, \quad k_B T_0 = \tilde{J}(0) - A . \tag{2.91} \]

The corresponding partition function is according to (2.75)

\[ Z = \int_{\alpha} \cdots \frac{d S_\alpha^2}{q < \Lambda_0} \exp \left[ -\frac{1}{4} \int_{q < \Lambda_0} (r + q^2) (S_\alpha^2)^2 \right] . \tag{2.92} \]
This is the product of \( N \times n \) independent single-variable Gaussian integrals (\( n \) = the number of spin components), and can be solved exactly as

\[
Z = (2\pi)^{\frac{nN}{2}} \int_{q < q_o} (r+q^2)^{-\frac{1}{2}}. \tag{2.93}
\]

Also the expectation values of several interesting physical quantities can be calculated exactly. For example, the net two-spin correlation function \( \tilde{G}_n(q) \) is calculated as

\[
\tilde{G}_n(q) \equiv \langle S^z_{\alpha} S^z_{\beta} \rangle = \frac{1}{Z} \sum_{a^z_{\alpha}} \int_{q < q_o} dS^z_{\beta} (S^z_{\alpha} S^z_{\beta}) \exp \left\{ \frac{E}{2} \right\}
\]

\[
= \delta_{\alpha}\beta / (r+q^2). \tag{2.94}
\]

From \( \chi^{\alpha\beta} = T_0^{\alpha\beta}(0) \) we find (cf. (2.91)):

\[
\chi^{\alpha\beta} = \frac{\delta^{\alpha\beta}}{r} = \frac{T_0^{\alpha\beta}}{k_B(T-T_0)}. \tag{2.95}
\]

For \( T = T_0 \) the susceptibility \( \chi^{\alpha\beta} \) diverges, so \( T_0 \) must be identified with the critical temperature. The divergence is described by a critical exponent \( \gamma = 1 \) (cf. Table 2.1). Comparison of (2.94) with (2.20) shows that in the above model

\[
1/\xi^2 = \kappa^2 = r = T - T_0, \tag{2.96}
\]

so \( \eta = 1/2 \), and moreover \( \eta = 0 \). Exactly the same results are obtained from mean-field calculations \(|1| \). Therefore, the above reduced model Hamiltonian (2.89) is known as the classical model. This result is not very surprising, as (2.89) implies a Gaussian spin-distribution function ((2.63) with \( u = 0 \)) in which short-range interactions between spins are not taken into account.

Next, we shall consider the iteration procedure of the RG-approach for this classical model. In the first step (2.84) we reduce the cutoff with a factor \( s \), i.e. \( \Lambda' = \Lambda_o / s \). Then \( 2\gamma' \) is found from (2.74) to be
\[
\exp f = \int_{-\infty}^{\infty} \frac{d S}{q} \, \exp \left[ - \int \frac{(r+q^2)(S^3)}{q^2} \right]
\]

\[
= \int_{-\infty}^{\infty} \frac{d S}{q} \, \exp \left[ - \int \frac{(r+q^2)(S^3)}{q^2} \right]
\]

\[
= \exp \left[ - \int \frac{(r+q^2)(S^3)}{q^2} \right] \times \text{Constant.} \quad (2.97)
\]

So,

\[
\hat{\tau} = \tau + \text{constant.} \quad (2.98)
\]

Rescaling the lengths and spins according to (2.85) and (2.86) yields

\[
\hat{\tau}' = \frac{1}{d+2} \int_{q'/s<s_0} \left[ \frac{(\hat{q}^2)}{s^2} c^2 \right] (S^3)_{q'}^2
\]

\[
= \int_{q^2<s_0} \left[ \frac{(\hat{q}^2)}{s^2} c^2 \right] (S^3)_{q'}^2
\]

\[
= \int_{q^2<s_0} \left[ \frac{(\hat{q}^2)}{s^2} c^2 \right] (S^3)_{q'}^2. \quad (2.99)
\]

In order to keep the constant in the \( \hat{q}^2 \)-term equal to unity, we have substituted

\[
c^2 = \frac{d^2}{d+2} \quad (2.100)
\]

From this procedure it can be concluded firstly, that the exact choice and shape of the cutoff is not significant, since the original choice is restored after each cycle. Secondly, the spin value is rescaled in each cycle, so the value of the original spins is not important for the results of this RG-procedure.

For the recursion relation of \( r \) we find

\[
r' = s \, r \quad (2.101)
\]
When the complete expression for $\mathcal{V}^{\mathcal{A}}(q)$ of (2.82) is taken into account one finds in addition \cite{37}

\begin{equation}
(j_1^{\mathcal{A} \gamma})' = j_1^{\mathcal{A} \gamma} \tag{2.102}
\end{equation}

\begin{equation}
\left[\mathcal{C}(q^4)\right]' = \left[\mathcal{C}(q^4)\right]/s^2 \tag{2.103}
\end{equation}

From the expressions (2.101) to (2.103) it can be seen that under the iterative application of the RG-transformation (with $s > 1$, of course), the $r$-term increases infinitely, whereas the $q^2$-term remains constant and higher-order terms vanish.

In the iteration process, the special case $\mathcal{R}(m+1) = \mathcal{R}(m) = \mathcal{R}^*$ corresponds to a so-called fixed point. This can only occur if the correlation length $\xi$ remains much greater than $1/\Lambda$ in the whole iteration procedure. This means that

\begin{equation}
\lim_{m \to \infty} \xi(m) = \xi(0)/s^m >> 1/\Lambda_0 \tag{2.104}
\end{equation}

which for $s > 1$ holds only if $\xi(0) = \infty$. Apparently a fixed point always corresponds to a system with $\xi = \infty$, i.e. a system at a critical point. In the above example this situation will ultimately be reached under the condition $r = 0$, i.e. for $T = T_0$. This once again identifies $T_0$ as the critical temperature. For $r \neq 0$ (2.101) implies an ever-increasing $r(m)$ due to the iteration process. This corresponds to an increasing $(T-T_0)$, and the series of reduced Hamiltonians $\mathcal{R}(m)$ describes how the system is removed from the critical point. This is in accordance with the content of the scaling hypothesis, viz. the change in any physical quantity near a critical point can be deduced from the way it behaves under a change in length scale.

It is not difficult to derive how the coefficient of the quartic term $\mathcal{V}^3_{\mathcal{A} \gamma} \delta$ in (2.79) changes under the RG-transformation. We find

\begin{equation}
(\mathcal{V}^3_{\mathcal{A} \gamma} \delta)' = \mathcal{A}^{-3d} (\mathcal{V}^3_{\mathcal{A} \gamma} \delta) \tag{2.105}
\end{equation}

(compare (2.99)). With (2.100) one arrives at
From (2.106) it can be concluded that the quartic term vanishes under the iterations for

\[ \varepsilon = 4-d < 0 \]  

(2.107)

This means that the classical model gives a good description for any system with \( d < 4 \). For \( d < 4 \) one can try to treat the quartic term as a small perturbation compared with the quadratic term. From (2.106) it becomes clear that it is far more important that \( \varepsilon = 4-d \) is small, than that \( V_{\text{quad}} \) is small. On basis of this idea a special perturbation method has been developed, the so-called \( \varepsilon \cdot \gamma \)-technique. 

**The RG-iteration process**

In general, the RG-transformation cannot be given in an exact form. If one starts from the reduced Hamiltonian (2.79), which might be extended with odd terms in the spin variables to describe magnetic systems in presence of a magnetic field, the transformation is always approximative. In most cases, the corresponding calculations are complicated numerical procedures, where the use of a large computer is indispensable. The above choice for \( \varepsilon \) as a partial trace over the spin variables in momentum space is certainly not unique. For instance, an \( \varepsilon \) which remains closer to the transformations in the Kadomtsev model and is applied in real space, was developed by Niemier and van Leeuwen to treat Ising systems. Although the calculations involved in RG-transformations for really existing systems are complicated, the principle remains simple, and it is instructive to notice how important interaction on critical behaviour may be derived from the sequence of RG-transformations.

This can be conveniently illustrated by following the variations of \( \varepsilon = \) under the repeated action of the parameter space. The one parameter system is approximated at some intermediate point in the parameter space. This point is approximated by another point in the parameter space which corresponds to \( \varepsilon = \) in the next intermediate parameter space. This process is repeated for each of the parameters.
In the above classical model, the axes would correspond to \( r, j_1, j_2, V_{ij} \), \( A_{ij} \), \( \sigma(q) \) etcetera. The parameter space can be considered as an extension of the field space, which was introduced in section 2.2. All thermodynamic fields are included in one or more parameters, for instance \( r \neq T \) in the classical model. However, not all parameters correspond to thermodynamic fields, as also parameters like the strength of exchange interactions \( J \) or single-ion anisotropies \( A_{ij} \) and \( B_{ij} \) in (2.78) are present.

In parameter space one can indicate iso- \( \tau \) surfaces, \( \ldots \) (hyper) planes of constant correlation length, the plane \( \tau = \) being called the \( \ldots \) \( \ldots \) plane. Fig. 2.11 shows some iso- \( \tau \) surfaces, crossed by the path of a system with smoothly varying temperature. At \( T = T_c \) this path reaches the critical surface. In the classical model the direction of the path would be along the \( r \)-axis. Each point of the path can be taken as \( \ldots \)
starting point for the RG-iteration process. As discussed above, the iterations will carry the system to smaller $\xi$ if one starts at $T \neq T_C$, since

$$\xi^{(m)} = \xi^{(0)} / s^m, \quad s > 1.$$  \hfill (2.108)

However, if the point at the critical surface is the starting point, then $\xi$ remains infinite (see Fig. 2.12.). Starting at $\tilde{\mathcal{F}}$, the path described by $\tilde{\mathcal{F}}^{(m)}$ for increasing $m$ is called the trajectory of $\tilde{\mathcal{F}}$. Fig. 2.12. illustrates that the trajectories of all reduced Hamiltonians, not lying on the critical surface, curve away from $S$. The trajectory of $\tilde{\mathcal{F}} (T=T_C)$ will be a line in the critical surface, which can end in two ways. Either, it can end in a fixed point, where

$$\tilde{\mathcal{F}} = \tilde{\mathcal{F}}^*,$$ \hfill (2.109)

or it can flow to a region in parameter space, in which the partition function has a thermodynamic instability, representing a first-order transition \cite{37}. Here we shall confine ourselves to the fixed points.

In the discussion of the classical model we shortly mentioned that the critical behaviour of a system near a critical point can be derived from the way in which a reduced Hamiltonian $\mathcal{F}$ close to $\mathcal{F}^*$ is transformed under the repeated action of $\mathcal{F}$. The location of $\mathcal{F}$ with respect to $\mathcal{F}^*$ can be indicated as

$$\mathcal{F} = \mathcal{F}^* + \hat{Q},$$ \hfill (2.110)

where $\hat{Q}$ denotes a unit vector in the parameter space and $\epsilon$ corresponds to the "distance" between $\mathcal{F}$ and $\mathcal{F}^*$. To find a description of the critical behaviour, it is assumed in the RG-approach that the RG-transformation $\mathcal{F}$ can be linearized near the fixed point. This is expressed by

$$\mathcal{F}' = \mathcal{F} + \hat{Q} \mathcal{F} + \mathcal{F} (\hat{Q}^2),$$ \hfill (2.111)

where $\mathcal{F}'$ describes a linear transformation of $\mathcal{F}$. 

[Image to be inserted here]
One can define a set of eigenvectors \( \hat{Q}_i \) and eigenvalues \( H_i \) as
\[
\hat{Q}_i = H_i \hat{Q}_i^+.
\]

For a reduced Hamiltonian expressed in terms of these eigenvectors as
\[
\rho^{(m)} = \rho^{(m)} \hat{Q}_i \hat{Q}_i^+ \rho^{(m)} \hat{Q}_i \hat{Q}_i^+ \tag{2.113}
\]
the RG-transformation consists of linear transformations like
\[
\rho_i^{(m+1)} = \rho_i^{(m)} \hat{Q}_i \hat{Q}_i^+ \rho_i^{(m)} = \rho_i^{(m)} H_i \hat{Q}_i \hat{Q}_i^+ \tag{2.114}
\]

The operator \( \rho \) and its eigenvalues depend explicitly on the choice for the scale factor \( s \). The operators \( \rho(s) \) constitute a semi-group, characterized by the property
\[
\rho(s) \rho(s') = \rho(s(s')). \tag{2.115}
\]

Hence we have
\[
H_i(s(s')) = H_i(s)H_i(s'), \tag{2.116}
\]
for all eigenvalues \( H_i \), which is fulfilled by
\[
H_i = s^{\lambda_i}. \tag{2.117}
\]

The change in distance \( \delta_i \) between \( \rho^{(m)} \) and \( \rho^* \) in the direction of \( \hat{Q}_i \) under the action of \( \rho \) is then found as
\[
\delta_i^{(m)} = (s^{\lambda_i})^m \delta_i^0. \tag{2.118}
\]

**Fig. 2.1**:

Fixed point \( \tilde{x}^* \) with the critical surface \( S^* \) spanned by irrelevant eigenvectors \( \hat{Q}_2 \) and \( \hat{Q}_3 \). \( \hat{Q}_1 \) is a relevant eigenvector.
If there is some eigenvector \( \hat{Q}_i \) with eigenvalue \( \lambda_i > 0 \), the distance between \( \hat{q} \) and \( \hat{q}^* \) will increase towards infinity along the \( \hat{Q}_i \)-axis, according to (2.118), unless \( \xi_i = 0 \). As the system is carried away from criticality for \( \xi_i \neq 0 \), the corresponding eigenvector \( \hat{Q}_i \) is called relevant. An eigenvalue \( \lambda_i < 0 \) will decay and the corresponding eigenvector \( \hat{Q}_i \) is called irrelevant. If some \( \lambda_i = 0 \), the corresponding \( \hat{Q}_i \) is called marginal. So, one can imagine that the parameter space near a fixed point on the critical surface is spanned by the complete set of eigenvectors \( \hat{Q}_i \), and that the critical surface is spanned by the irrelevant vectors only (see Fig. 2.13.). The trajectory of a reduced Hamiltonian with the coefficients \( \xi_i \) of all relevant eigenvectors zero, i.e., a point at the critical surface, will end at the fixed point \( \hat{q}^* \). Otherwise, the trajectory will bend away from \( \hat{q}^* \). Therefore, at least some of the coefficients \( \xi_i \) belonging to relevant eigenvectors \( \hat{Q}_i \) may be considered as the relevant fields, which were introduced in section 2.2. A similar conclusion can be drawn about irrelevant fields. For the classical model we already observed that the parameter \( r \) corresponds to a relevant eigenvector \( \hat{Q}_i \) of \( \hat{q} \), as its eigenvalue is \( s^2 \) according to (2.101), i.e., \( \lambda_i = 2 \). The numerical value of \( r \) must be identified with the coefficient \( \xi_i \) of this \( \hat{Q}_i \) which therefore corresponds to the relevant field \( \hat{T} \). By means of eqs. (2.102) and (2.103) one may classify \( j \) as marginal and the terms \( \beta(\hat{q})^4 \) as irrelevant.

With use of the renormalization relations (2.118) for the fields \( \bar{q}_i^{(m)} \), we obtain for the Gibbs free energy (2.87) the expression

\[
G\left[\bar{q}_1^{(m+1)}, \bar{q}_2^{(m+1)}, \ldots \right] = G\left[s^{\lambda_1} \bar{q}_1^{(m)}, s^{\lambda_2} \bar{q}_2^{(m)}, \ldots \right] = s^d G\left[\bar{q}_1^{(m)}, \bar{q}_2^{(m)}, \ldots \right].
\]

So, the RG-approach yields that \( G \) is a generalized homogeneous function, as long as the linearized transformation is a good approximation of \( \hat{q} \), i.e., when the term \( \beta(\hat{q})^4 \) in (2.111) is negligible. With increasing relevant \( \xi_i \) this assumption will ultimately become invalid. We can therefore define the critical region as the region in parameter space...
where the expression (2.119) holds. Also the correlation function (2.88) is a generalized homogeneous function. With \( c = s^a \), where \( a \) is an arbitrary number, we find

\[
G_n = s^{-1} R(m), s^{\lambda_1} \xi_1(m), s^{\lambda_2} \xi_2(m), \ldots
\]

From the exponents \( \lambda_1, \lambda_2 \) etc. all other critical exponents can be derived, as was illustrated extensively in section 2.5. Hence, the critical exponents of a critical system can be calculated in the RG-approach, by determining the eigenvalues of the linearized RG-transformation \( (2.117) \).

**Types of fixed points**

In general, more than one fixed point can be present on the critical surface. To each fixed point a characteristic type of critical behaviour is related. Each fixed point will have its own domain of attraction, which covers all reduced Hamiltonians \( \mathfrak{F} \) on \( S_m \) with trajectories ending in the fixed point under consideration. One can draw an extensive analogy between the trajectory followed by an \( \mathfrak{F}(m) \) under the repeated action of \( \mathfrak{F} \) and the path of a ball on a surface with relief under the action of gravitation \([35]\). A fixed point corresponds in this analogy to a point in the surface relief, where the slope in all directions is zero. Different types of fixed points then can be characterized by their stability. On \( S_m \) there can be one or more stable fixed points (corresponding to potential wells in the analogy), unstable fixed points (mountain tops) or saddle-point-like fixed points. All three types of fixed points are shown in Fig. 2.14.

A **stable fixed point** \( \mathfrak{F} \) (point A in Fig. 2.14) is surrounded by its domain of attraction, which is spanned by the irrelevant vectors \( \mathfrak{Q}_1 \). All reduced Hamiltonians \( \mathfrak{F} \) in this domain have trajectories ending in \( \mathfrak{F} \). This implies that all systems, which are described by some \( \mathfrak{F} \) in this domain, will show the critical behaviour corresponding to \( \mathfrak{F} \).
Different types of fixed points. A is a stable fixed point, B is an unstable fixed point and C is a saddle-point-like fixed point, related to multicritical behaviour. The trajectory of P illustrates the competition between two fixed points, which can give rise to crossover behaviour.

Therefore, one can conclude that these domains of attraction constitute the universality classes, introduced in section 2.4. Each fixed point corresponds to a specific universality class. A saddle-point-like fixed point is unstable with respect to some of the eigenvectors $\hat{Q}_i$ in the critical surface, which apparently are relevant for this specific kind of critical behaviour (point C in Fig. 2.14). This behaviour is interpreted as multi-critical behaviour and this kind of fixed points denoted as multi-critical points. The domain of attraction of such a fixed point (i.e. the multi-critical surface) constitutes only a small subspace of the entire critical surface. Consequently also in the thermodynamic field space, the occurrence of multicritical points will be rare compared with the occurrence of critical points. For an unstable fixed point all eigenvectors are relevant. Therefore, such a point will be seldomly found in nature.

The competition between two fixed points can give rise to crossover phenomena. A trajectory can closely pass by a fixed point and ultimately bend towards another one (see Fig. 2.15). In such a case it is possible to observe two kinds of critical behaviour. During the RG-iteration process one can initially, i.e. on a small length scale, observe a critical behaviour which is characteristic for fixed point C (in Fig. 2.14), whereas finally, i.e. on a large length scale, the characteristic critical behaviour of fixed point A is found. A similar crossover can be observed in thermodynamic field space, when two or more critical points, belonging to different universality classes, are
located at close distance. This behaviour will be described in more
detail in chapter III and constitutes one of the main topics in the
experimental study on $\text{CsMnBr}_3 \cdot 2\text{D}_2\text{O}$, which we describe in chapter VI.

![Diagram](image-url)

*Fig. 1.11. Initial impression of the crossover of a trajectory in*
the initial surface from initial point C to final point B.*
References to Chapter II.

10. Vol. 6 in the series of '2'.
13. See J. Als-Nielsen in vol. 5A of '2'.
'23' H.E. Stanley in Vol. 3 of '12'.
'24' Vol. 3 of '12'.
'25' C. Domb in Vol. 3 of '12', see also Vol. 1 of '2'.
'26' Various authors. Values tabulated by D.J. Wallace in Vol. 6 of '12'.
'32' L.P. Kadanoff in Vol. 5A of '2'.
'37' A. Aharony in Vol. 6 of '12', see also '6'.
'39' Th. Niemeijer and J.M.J. van Leeuwen, in Vol. 6 of '2'.
CHAPTER III

PHASE TRANSITIONS AND CRITICAL BEHAVIOUR IN WEAKLY ANISOTROPIC ANTIFERROMAGNETS

3.1. Introduction

It was pointed out already by Néel [1], that magnetic moments, strongly coupled by an antiferromagnetic exchange interaction, tend to align perpendicularly to an applied magnetic field. If the moments have a preferred orientation in absence of a magnetic field, due to some kind of anisotropic interaction, these two effects will compete if \( \mathbf{H} \) is applied along the preferred direction of the magnetic moments (easy axis). In real antiferromagnets there are many effects which can cause anisotropy, such as ligand field effects, spin-orbit coupling or dipolar interactions. Then the sublattice magnetizations \( \mathbf{M}_A \) and \( \mathbf{M}_B \), which are aligned along the easy axis for small \( H_\parallel \), will jump to a direction more or less perpendicular to the easy axis if \( H_\parallel \) has been increased sufficiently. If we identify the anisotropy with an internal field \( H_A \) and denote \( |\mathbf{M}_A| = |\mathbf{M}_B| = M \), this spin-flop field \( H_{SF} \) can be found from

\[
\frac{1}{2}(\chi_\parallel - \chi_\perp) H_{SF}^2 = M H_A.
\]

Here \( \chi_\parallel \) and \( \chi_\perp \) indicate the parallel and perpendicular susceptibility, respectively. In general

\[
\chi_\perp \gg \chi_\parallel
\]

in antiferromagnets in the ordered state, at least for \( T \ll T_N \). This spin-flop phenomenon was observed for the first time in CuCl\(_2\)·2H\(_2\)O by Poulis and coworkers [2] and appears to be very common in antiferromagnets.

In the calculation of phase diagrams in antiferromagnets, pioneer work has been performed by the group of Gorter in Leiden [3,4]. Their
calculations are based on the molecular-field (MF) approximation, which is exact for systems with \( d \geq 4 \), but displays severe shortcomings close to critical points in realistic systems with \( d \leq 3 \), as we showed in chapter II. Nevertheless, the MF-approximation has proven to be a good guide in the prediction of qualitative features of phase diagrams in antiferromagnets. Therefore, we shall start in section 3.2. with a presentation of the MF-results for antiferromagnets with weak spin anisotropy, often called spin-flop systems. In section 3.2. we shall confine ourselves to the results obtained for spin-flop systems in a parallel magnetic field \( H_{//} \), i.e. with \( H \) applied along the easy axis.

Complementary information, especially on the critical behaviour in spin-flop systems, is obtained from calculations in the RG-approach. The main lines and results of this approach in spin-flop systems are presented in section 3.3. We shall show that in spin-flop systems two different magnetic phase diagrams can occur, which in addition to lines of critical points also display a multi-critical point. So far, only the so-called bicritical point has been observed in spin-flop systems. The so-called tetracritical point is predicted in MF-approximation, but has been questioned for several years on basis of RG-calculations \[5\]. Recently, Bruce and Aharony pointed out that such a point can exist in spin-flop systems, indeed \[6\]. In chapter V of this work we shall present strong evidence for the presence of such a tetracritical point in \( \text{CoBr}_2\cdot6(0.48 \text{D}_2\text{O}, 0.52 \text{H}_2\text{O}) \). Section 3.3. may be skipped by readers who are only interested in the experimental part of this work.

In section 3.4. we present the so-called extended-scaling theory, which describes the critical behaviour close to a multi-critical point. This theory is a straightforward extension of the scaling theory of critical points, treated in section 2.5., and provides also a description of the crossover from critical to multi-critical behaviour. The presentation and discussion of the predictions based on the extended-scaling hypothesis will be rather extensive, as the experimental verification of this hypothesis near the bicritical point in the phase diagram of \( \text{CsMnBr}_3\cdot2\text{D}_2\text{O} \) forms the essential part of chapter VI.
Finally, section 3.5. treats the phase diagrams and the corresponding critical behaviour of spin-flop systems in a skew magnetic field, \( \mathbf{H} \), with \( \mathbf{H} \) not parallel to the easy axis.

3.2. Phase diagrams of spin-flop systems in the MF-approximation

As a starting point of the discussion we consider a microscopic Hamiltonian of the general form

\[
\mathcal{H} = -2 \sum_{\langle i,j \rangle} \sum_{\alpha} J_{ij}^{\alpha} (\mathbf{R}_i - \mathbf{R}_j) \cdot \mathbf{S}_i^\alpha S_j^\alpha - \sum_{i} \sum_{\alpha} H_i^\alpha S_i^\alpha \tag{3.3}
\]

consisting of an anisotropic exchange term and a Zeeman term. \( \langle i,j \rangle \) indicates the sum over all pairs of spins and \( S_i^\alpha \) denotes the \( \alpha \) component of the spin at site \( \mathbf{R}_i \). We shall assume that all tensors \( J_{ij}^{\alpha} \) in (3.3) will have the same principal axes, so that we can write (3.3) as

\[
\mathcal{H} = -2 \sum_{\langle i,j \rangle} \sum_{\alpha} J_{ij}^{\alpha} (\mathbf{R}_i - \mathbf{R}_j) \cdot \mathbf{S}_i^\alpha S_j^\alpha - \sum_{i} \sum_{\alpha} H_i^\alpha S_i^\alpha \tag{3.4}
\]

where \( \sum \) now denotes a summation of the components along the three principal axes of \( J_{ij}^{\alpha} \) and \( J_{ij}^{\alpha} = J_{ij}^{\alpha_1} \). Here we shall confine ourselves to simple antiferromagnets, which can be described by two interpenetrating sublattices \( A \) and \( B \).

In the MF-approximation one can write the Gibbs free energy of such a two-sublattice antiferromagnet as

\[
G = \frac{1}{2} N \sum_{\alpha} \left[ A \mathbf{m}_A^\alpha \mathbf{m}_A^\alpha - \frac{1}{2} D \left( (\mathbf{m}_A^\alpha)^2 + (\mathbf{m}_B^\alpha)^2 \right) - \mu H (\mathbf{m}_A^\alpha + \mathbf{m}_B^\alpha) \right] \tag{3.5}
\]

Here the magnetizations of the sublattices are normalized as

\[
\mathbf{m}_A = \frac{\mathbf{M}_A}{4N\mu} \quad \text{and} \quad \mathbf{m}_B = \frac{\mathbf{M}_B}{4N\mu} \tag{3.6}
\]

where \( \mu = g\mu_B S \).

\[
\mu \quad \text{is the magnitude of the magnetic moment of the N magnetic ions.} \tag{3.7}
\]
In (3.5) $A_\alpha$ and $D_\alpha$ represent the inter-sublattice and intra-sublattice couplings, respectively. These macroscopic coupling constants can be related to the microscopic interaction constants between nearest-neighbours in (3.4) as

$$A_\alpha = -\frac{1}{2}z^2 J_1^\alpha$$
$$D_\alpha = +\frac{1}{2}z^2 J_2^\alpha$$ (3.8)

Here $J_1$ and $J_2$ denote the exchange coupling of a spin with each of its $z_1$ nearest neighbours on the other sublattice and with each of its $z_2$ nearest neighbours on the same sublattice, respectively. Eq. (3.5) is the common starting point of many calculations in the MF-approximation.

Here we shall be concerned with antiferromagnets in which all three components of $\hat{A}$ are positive, i.e.

$$A_\alpha > 0, \text{ for all } \alpha.$$ (3.9)

In that case the sublattice magnetizations will be aligned antiparallel, irrespective of the actual preferred direction. In general one can distinguish between an easy $z$-axis, an intermediate $y$-axis and a hard $x$-axis, the distinction between the axes being determined by $|J|$

$$A_z + D_z > A_y + D_y > A_x + D_x.$$ (3.10)

This is called orthorhombic spin anisotropy, in contrast with uniaxial spin anisotropy, which corresponds to

$$A_y = A_x \text{ and } D_y = D_x.$$ (3.11)

Under the conditions (3.9) and (3.10) and with $H$ along the easy axis, there are four stable phases which minimize $G$ in (3.5). In all phases the magnetic moments are confined to the $yz$-plane:

- the paramagnetic (P) phase, where $\hat{M}_A = \hat{M}_B = 0, \text{ if } H=0$,
- the antiferromagnetic (AF) phase, where $\hat{M}_A$ and $\hat{M}_B$ have opposite directions along the $z$-axis,
- the spin-flop (SF) phase, where $\hat{M}_A$ and $\hat{M}_B$ are symmetrical with respect to $H$, and not aligned along the $z$-axis,
a less familiar, intermediate (I) phase, where \( \hat{M}_A \) and \( \hat{M}_B \) make unequal angles with \( H \).

The behaviour of the sublattice magnetizations as a function of \( H \) can be visualized for \( T=0 \), where \( \hat{M}_A \) and \( \hat{M}_B \) are completely saturated, i.e. \( M_A = M_B = \frac{N}{2} \mu \). Then one can express the homogeneous magnetization \( \hat{M} \) and the staggered magnetization \( \hat{M}_{st} \) as

\[
\hat{M} \equiv \frac{N}{2} (\hat{m}_A + \hat{m}_B) = N \mu \sin \delta (0, -\cos \zeta, \sin \zeta) \tag{3.12}
\]

and

\[
\hat{M}_{st} \equiv \frac{N}{2} (\hat{m}_A - \hat{m}_B) = N \mu \cos \delta (0, \sin \zeta, \cos \zeta), \tag{3.13}
\]

where the angles \( \delta \) and \( \zeta \) are defined in Fig. 3.1.

For the four different phases the variation of \( \delta \) and \( \zeta \) with \( H \) at \( T=0 \) are summarized in Table 3.1., together with the corresponding stability conditions [8].

**Table 3.1.** Character and stability conditions of possible phases in antiferromagnetic systems with two sublattices [3]

<table>
<thead>
<tr>
<th>Phase</th>
<th>Picture</th>
<th>( \sin \zeta )</th>
<th>( \sin \delta )</th>
<th>Stability conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>AF I</td>
<td>[\sin \zeta ]</td>
<td>0 [\sin \delta ] [h_1/\sqrt{Q} ]</td>
<td>0 [h_2/\sqrt{Q} ]</td>
<td>[h_2/\sqrt{Q} ] [\sqrt{Q}(1-R/Q) ]</td>
</tr>
<tr>
<td>SF P</td>
<td>[\sin \zeta ]</td>
<td>1 [\sin \delta ] [h/(1-R) ]</td>
<td>1 [h/(1-R) ]</td>
<td>[h_2/\sqrt{Q}(1-R/Q) ] [\sqrt{Q}(1-R/Q) ]</td>
</tr>
</tbody>
</table>

\( Q=(\Lambda_1^2-\Lambda_1^0)/(\Lambda_2^1+\Lambda_2^0), R=2D/(\Lambda_2^1+\Lambda_2^0), h=\mu H/(\Lambda_2^1+\Lambda_2^0) \)
In an increasing magnetic field, the phase with the lowest free energy will always be realized. For different combinations of molecular-field constants, the sequences of realized solutions are depicted in Fig. 3.2. The same result has been obtained by other authors [9] with

![Diagram](image)

**Fig. 3.2.**

Sequences of thermodynamically stable solutions occurring with increasing \( H \) for different combinations of MF-constants at \( T=0 \). For \( H \) sufficiently high, the \( P \)-phase becomes stable for any combination (taken from [7]).

Reference to the ordered phases in \(^{3}H e\) quantum crystals, a system which is described by a Hamiltonian similar to (3.3). In the present context, we are interested in systems which display two or more ordered phases. Fig. 3.2. shows that this behaviour can be met in systems where

\[
D < A_y < A_z + D. \tag{3.14}
\]

These systems all have an SF-phase and therefore are called spin-flop systems. Materials in which \( D > A_y \) correspond to the so-called metamagnets, which recently have been reviewed by Stryjewski and Giordano [10]. Systems with \( A_y > A_z + D \) do not fulfill condition (3.10), which defines \( z \) as the easy axis. The spin-flop systems can be divided into two classes, which display a qualitatively different behaviour in a magnetic field.

The most familiar class of spin-flop systems consists of materials with \( 0 < D < A_y < A_z + D \). At \( T=0 \) the magnetic moments of these systems are ordered in an AF-phase for \( H < H_{SF} \) and in an SF-phase for \( H > H_{SF} \). The spin-flop transition at \( H = H_{SF} \), introduced in section 3 , is
a first-order transition. At $H_{SF}$ the order parameter of the system in the AF-phase, $M_{st}^\parallel$, changes discontinuously from a finite value to zero. Simultaneously, the order parameter of the system in the SF-phase, $M_{st}^\perp$, changes discontinuously from zero to a finite value. $M_{st}^\perp$ is defined by

$$M_{st}^\perp = M_{st}^\perp < (3 - 16a)$$

in case of a system with orthorhombic anisotropy, and by

$$M_{st}^\perp = (M_{st}^X)^2 + (M_{st}^Z)^2$$

in case of uniaxial anisotropy. At a sufficiently high magnetic field $M_{st}^\perp$ vanishes continuously at the critical point $(H = H_C, T = 0)$. For $H > H_C$ the system is in the P-phase, which in nonzero field corresponds to a field-induced ferromagnetic ordering.

A typical $H - T$ diagram for this class of systems is shown in Fig. 3.3a [4,11]. As indicated in section 2.2., the transition from the AF-phase to the P-phase takes place at a line of critical points $T_C^\parallel (H)$, which is the continuation of the Néel point for $H \neq 0$. At $T_C^\parallel (H)$ the order parameter of the AF-phase, i.e. $M_{st}^\parallel$, vanishes. Similarly, the order parameter of the SF-phase $M_{st}^\perp$ vanishes at a second line of critical points $T_C^\perp (H)$, which is the continuation of $(H = H_C, T = 0)$ for $T \neq 0$. Below, these two critical lines will often be called "the paramagnetic phase boundaries". Both critical lines meet the first-order spin-flop line, being the continuation of the point $(H = H_{SF}, T = 0)$ for $T \neq 0$, at the same point $(H_b, T_b)$. According to the definition in section 2.2., $(H_b, T_b)$ is a multi-critical point. It is commonly called a bicritical point [12], as in the $H - T$ diagram two different critical lines meet in this point. It will be shown below, that this denomination must be considered as an ad-hoc choice, although the terminology suggests a logical division of multi-critical points. In the description of critical behaviour near the bicritical point, which is treated in
section 3.4., the order parameters $M_{st}^{\parallel}$ and $M_{st}^\perp$ both play a role.

![Diagram](image)

*Fig. 5.1. Schematic ($H$, $T$) phase diagrams of a spin-flop system displaying (a) a bicritical and (b) a tetracritical point. Solid lines indicate lines of critical points, the dashed line corresponds to a first-order transition.*

The second class of spin-flop systems is less familiar and consists of materials with $D < 0 < A_{y} < A_{z} + D$. MF-calculations for $T = 0$ [7,11] yield a transition at $H = H_{IA}^*$, where the system passes from the AF-phase into the I-phase. The point $(H = H_{IA}^*, T = 0)$ is a critical point where $M_{st}^\perp$, being zero in the AF-phase, starts to increase. In the I-phase $M_{st}^{\parallel}$ is decreasing but still nonzero, in other words, a gradual rotation of $M_{st}^\perp$ takes place with increasing field $H_{\parallel}$. At $H = H_{IS} > H_{IA}^*, M_{st}^{\parallel}$ vanishes continuously, which indicates that the point $(H = H_{IS}^*, T = 0)$, where the transition to the SF-phase takes place, is a critical point. Apparently both $M_{st}^{\parallel}$ and $M_{st}^\perp$ are important in the description of the magnetic order in the I-phase, as both quantities play the role of order parameters. The characteristic feature of the I-phase is the coupling between both order parameters. This will be extensively discussed in section 3.3. For increasing field values $H_{\parallel} > H_{IS}^*$, $M_{st}^\perp$ gradually decreases and ultimately vanishes at the critical point $(H = H_{c}^*, T = 0)$.

A typical example of the complete $H_{\parallel}$ $T$-diagram displayed by this class
of spin-flop systems |11|, is shown in Fig. 3.3b. In addition to the paramagnetic phase boundaries $T_{c//}(H)$ and $T_{c\perp}(H)$, two other lines of critical points $T_{IA}(T)$ and $H_{IS}(T)$ are present, which separate the I-phase from the AF- and the SF-phase, respectively. All four critical lines meet in a so-called tetracritical point |11|. During several years the existence of such a tetracritical point in common antiferromagnets has been questioned, due to RG-calculations which yield a tetracritical fixed point only for $n>3.1$ as $d>3$. For conclusive arguments one cannot rely on MF-theory, as the validity of the MF-approximation is strongly reduced close to critical points. So, even if MF-theory predicts an I-phase for certain systems, this phase might exist only far from criticality. In section 3.3. we shall discuss the arguments of Bruce and Aharony |6|, which lead to the conclusion that a tetracritical point indeed can exist for $d>3$, but that the corresponding multi-critical behaviour is the same as the behaviour close to a bicritical point.

Finally, it should be noted that the terminology of multi-critical points neither is logical nor uniquely determined. The first failure of this nomenclature becomes clear if the above bicritical point is considered in presence of a (small) field component $H_y$ along the $y$-axis. As is shown in Fig. 3.8. and discussed in section 3.5., at least three different critical lines and surfaces in this three-dimensional space meet in the point $(H = H_y, T = T_b, H_i = 0)$. Moreover, this space of thermodynamic fields is still far from complete. The ambiguity of the terminology becomes clear in the treatment of tetracritical points. In addition to the above tetracritical point with coupled order parameters, an essentially different type of tetracritical point exists in randomly mixed magnetic systems with competing spin anisotropies. In this tetracritical point, which recently has been reported by Bevaart et al. |13,14|, the order parameters are decoupled and a different type of multi-critical behaviour is expected |15|. In section 3.3. it will be shown that the difference between these two tetracritical points is due to the presence or absence of a coupling term of the form $S_x S_y$ in the Hamiltonian.

Section 3.3. treats the RG-approach in the calculation of critical and multi-critical behaviour in spin-flop systems. Especially the debate
about the existence of tetracritical points will be elucidated. Readers, who missed sections 2.6. and 2.7. can also skip section 3.3., without losing the connection with section 3.4., where the predictions of the extended-scaling theory for bicritical and tetracritical points will be presented.

3.3. RG-predictions for critical behaviour in spin-flop systems

In this section we shall consider the reduced Hamiltonian $\hat{H}$ for a spin-flop system, as derived by Kosterlitz et al. [16]. The specific features of this system, differing from the description of the classical model in section 2.7., are:

- the introduction of two sublattices in the calculation,
- the presence of a magnetic field $H$,
- the different role played by two spin components, which changes with the magnitude of $H$,
- the explicit elaboration of the terms quartic in the spin variables.

We shall roughly sketch the treatment of some of these aspects in the derivation in ref. [16], which ultimately yields an expression for $\hat{H}$ which is included in (2.79), viz. the spin-anisotropic type in Table 2.4.

Next, the recursion relations for the relevant parameters, obtained from the RG-iteration, and the role of different relevant fields in the description of the critical and multi-critical behaviour are discussed. Finally the discussion about the existence of tetracritical points in $d=3$ antiferromagnets will be presented extensively.

Derivation of the reduced Hamiltonian

In the RG-calculations of Kosterlitz et al. [16] on the critical behaviour of spin-flop systems, the Hamiltonian (3.4) serves as point of departure. Assuming two sublattices $A$ and $B$, the inter-sublattice and intra-sublattice terms can be separated, which leads to a reduced Hamiltonian $\hat{H}$ of the form:

$$\hat{H} = \frac{1}{k_B T} \sum_{i_A, i_B} \left[ 2 \sum_{j_A, j_B} J^A_{ij} S^A_i S^A_j + J^B_{ij} \left( \sum_{i_A, j_A} S^A_i S^A_j \right) + \sum_{i_A, i_B} H' \left( \sum_{i_A} S^A_i + \sum_{i_B} S^B_i \right) \right] \left( \sum_{i_A} \cos \theta^A_i - \sum_{i_B} \cos \theta^B_i \right) \left( \sum_i \sin \theta^A_i + \sum_i \sin \theta^B_i \right).$$

(3.17)
Here we used the usual truncated spin-distribution function $D(S)$ of (2.63). $S^z_A$ is the $z$-component of the spin at site $\vec{R}_i$ which belongs to sublattice $A$, $\langle i_A, j_B \rangle$ indicates that the summation has to be performed over all pairs of spins which are on different sublattices. Similarly, $\langle i_A, j_A \rangle$ and $\langle i_B, j_B \rangle$ indicate summations over all pairs of spins on sublattice $A$ and $B$, respectively. $J_1$ and $J_2$ represent the inter-sublattice and intra-sublattice exchange interactions, respectively, and depend on the distance between spins.

Like in section 2.6, the quadratic part of (3.17) can be diagonalized in terms of Fourier-transformed spin variables. Here the appropriate Fourier transforms are

$$ (S^q)^z = \frac{1}{2} \left[ \sum_{i_A} \exp(i\vec{q} \cdot \vec{R}_i) S^z_A + \sum_{i_B} \exp(i\vec{q} \cdot \vec{R}_i) S^z_B \right]. \quad (3.18) $$

$S^z_q$ and $S^z_{-q}$ may be considered as the microscopic equivalents of $\vec{M}$ and $\vec{M}_{st}$ in momentum space. Then (3.17) can be written as

$$ \vec{\gamma} = -J \sum_q \left[ \int (1-J_2(q))(S^q)^z (S^-)^z + \int (1-J_2(q))(S^q)^z (S^-)^z \right] $$

$$ + 2 L^2 (S^q)^z \sum_{\alpha \beta} \gamma(S^\alpha_q) \quad (3.19) $$

where

$$ J_2(q) = \sum_{\vec{R}} \exp(i\vec{q} \cdot \vec{R}) \left[ J_1^2(\vec{R}) \pm \right] \quad (3.20) $$

and

$$ L^2 = \frac{\hbar^2}{k_B T}. \quad (3.21) $$

The last term in (3.19) consists of many cross terms of the form

$$ \int \int \int (S^q)^z (S^q)^z (S^-)^z (S^-)^z \gamma(S^q)^z (S^-)^z \quad (3.22) $$

As has been discussed to some extent in section 3.2, it is sufficient to distinguish between the spin component parallel to the easy axis, $\ldots$, $S^z_q$, and the spin components perpendicular to the easy axis, $\ldots$. 

to describe a spin-flop system in a parallel field $H$. $S_q^\perp$ will consist of the $x$ and $y$-components of $S_q^\perp$ in a uniaxial system, whereas only $S_q^\parallel$ is important in an orthorhombic system. In the following we shall confine ourselves to a uniaxial system in a parallel field. The result can be easily adapted to the orthorhombic case. Ref. [16] treats also spin-flop systems in a skew field, which will be discussed in section 3.5. We now introduce the notation

$$S_q^\parallel = \langle S_q^\parallel \rangle, \quad S_q^\perp = \langle S_q^\perp \rangle,$$  \hspace{1cm} (3.23)

where the $q$-subscripts have been omitted and the parallel and perpendicular components of $S_q^\perp$ are distinguished in a convenient way. Then the reduced hamiltonian (3.19) can be decomposed in three parts [16]:

$$\hat{H} = \hat{H}_\parallel + \hat{H}_\perp + \hat{H}_x,$$  \hspace{1cm} (3.24)

where $\hat{H}_\parallel$ and $\hat{H}_\perp$ contain only $\sigma$-terms and $\tau$-terms, respectively, and $\hat{H}_x$ contains the cross terms which are purely quartic in the spin components.

In a similar procedure as in section 2.7, the components of the short-range interactions $J^\parallel(q)$ and $J^\perp(q)$ in (3.19) can be expanded in powers of $\tilde{q} = q/(q_m)$, where the basic temperature variables become

$$\tilde{T}_\parallel = k_B \left( T - T_o^\parallel \right)/J,$$  \hspace{1cm} (3.25a)

and

$$\tilde{T}_\perp = k_B \left( T - T_o^\perp \right)/J.$$  \hspace{1cm} (3.25b)

By definition one has $T_o^\parallel > T_o^\perp$. Next the linear Zeeman term in (3.19) can be eliminated by defining a new spin variable $\sigma_+ \rightarrow \sigma_+^0$ and a corresponding adjustment of all coefficients. For instance, the displaced temperature variables become

$$\tilde{T}_\parallel^0 = k_B \left( T - T_o^\parallel^0 \right)/J + a_0^\parallel \tilde{T}_\parallel^0,$$  \hspace{1cm} (3.26a)

and

$$\tilde{T}_\perp^0 = k_B \left( T - T_o^\perp^0 \right)/J + a_0^\perp \tilde{T}_\perp^0.$$  \hspace{1cm} (3.26b)
where $a_0^\parallel$ and $a_0^{\perp}$ ($c a_0^\parallel$) are positive constants, proportional to $M^2/k_B T$. It appears that all terms in $c_+$ and $s_+$ are either strongly irrelevant or can be integrated out of the partition function $Z_{\parallel 6}$, so that ultimately a reduced Hamiltonian is obtained of the form

$$\hat{H} = -\frac{1}{2} \int \frac{(r_\parallel + \hat{q}^2)q_\parallel^2}{q^2} - \frac{1}{2} \int \frac{(r_\perp + \hat{q}^2)|\hat{s}_\perp|^2}{q^2} +$$

$$+ \int \int \frac{\{wu_\parallel + 2w_\parallel^2|\hat{s}_\parallel|^2 + v|\hat{s}_\parallel|^4\} + \ldots}{q^2}, \quad (3.27)$$

where the usual momentum-conserving subscripts have been deleted for convenience. Terms in $\hat{q}^\mu$ have been omitted as they appear to be irrelevant. The anisotropic terms $\sum_{q_\parallel^\mu} \hat{q}_\parallel^\mu (c_\parallel^\mu, (2.81))$ do not play an important role either and will not be taken into account below.

In the quartic term of (3.27) the coefficients $u$, $v$ and $w$ are positive, vary slowly with $T$ and $H^\parallel$, and satisfy no special relations $|16|$. In absence of the coupling term with coefficient $w$ and higher-order terms, (3.27) would represent two uncoupled critical systems, viz. the system of parallel spin variables $\sigma_\parallel$ and the system of perpendicular spin variables $\hat{s}_\perp$.

**The fixed points of a spin-flop system**

In the discussion of the critical behaviour corresponding to (3.27), we must distinguish between two critical surfaces, corresponding to $\xi_\parallel = 0$ and $\xi_\perp = 0$, where $\xi_\parallel$ and $\xi_\perp$ are the correlation lengths of the spin fluctuations in the parallel and the perpendicular spin components, respectively. Under the iterative action of the RG-transformation $\hat{H}$, the trajectory of any $\hat{H}$ with $r_\parallel$ and $r_\perp$ both nonzero, will bend away from both critical surfaces. The derivation of this result follows the lines of the discussion on the classical model in section 2.7.

Similar to $\chi_{\parallel}$ in section 2.7. ($\chi_{\parallel}$, (2.95)), now the staggered susceptibilities can be expressed with $\sigma_\parallel^{\pm} = (S_\parallel^{\pm})$, $\hat{s}_\perp^{\pm} = (S_\perp^{\pm})$ as

$$\chi_{\parallel}^{\perp} = \frac{1}{q_\parallel^2} \langle \sigma_\parallel^{+} \sigma_\parallel^{-} \rangle_{q_\parallel^2} = \frac{1}{r_\parallel^2} \quad (3.28a)$$
and

\[ \chi_{st}^{\perp} = \frac{s_{\parallel} \cdot \hat{s}_{\perp}}{q=0} = \frac{1}{r_{\perp}}, \]  

(3.28b)

and will diverge for \( r_{\parallel} = 0 \) and for \( r_{\perp} = 0 \), respectively. For \( r_{\parallel} \ll r_{\perp} \) (i.e. for \( H_{\parallel}^{2} \ll (T_{c}^{\perp} - T_{c}^{\parallel})/(a_{0}^{\parallel} - a_{0}^{\perp}) \)), the divergence of \( \chi_{st}^{\parallel} \) takes place at the highest temperature, viz.

\[ T_{c}^{\parallel} (H_{\parallel}^{2}) = T_{c}^{\parallel} - \frac{J_{a_{0}^{\parallel}}}{k_{B}} H_{\parallel}^{2}. \]  

(3.29a)

For \( T < T_{c}^{\parallel} \) a LRO of the parallel spin components is established in an AF-phase. Therefore \( T_{c}^{\parallel} (H_{\parallel}^{2}) \) corresponds to the critical phase boundary \( T_{c}^{\parallel} (H) \) in Fig. 3.3. Since only one spin component is involved in this ordering process, the transition will be Ising-like, i.e. the trajectory of \( \hat{s} \) with \( r_{\parallel} = 0 \) will end in the Ising fixed point \( \hat{s}^{*} \), located at \( r_{\parallel}^{*} = 0, r_{\perp}^{*} = 0, u^{*} = u_{0}, v^{*} = v^{*} = 0 \), with \( u_{0} \) a nonzero constant (to be calculated). For \( r_{\perp} \ll r_{\parallel} \) (i.e. for \( H_{\parallel}^{2} \gg (T_{c}^{\parallel} - T_{c}^{\perp})/(a_{0}^{\parallel} - a_{0}^{\perp}) \)), \( \chi_{st}^{\perp} \) will diverge at the highest \( T \), viz. at the critical points

\[ T_{c}^{\perp} (H_{\parallel}^{2}) = T_{c}^{\perp} - \frac{J_{a_{0}^{\perp}}}{k_{B}} H_{\parallel}^{2}. \]  

(3.29b)

and establish a long-range ordered SF-phase. In case of orthorhombic spin anisotropy this transition will be Ising-like, whereas in a uniaxial system the transition will be XY-like. The corresponding fixed points are located at \( r_{\parallel}^{*} = 0, r_{\perp}^{*} = 0, u^{*} = v^{*} = v_{0} \neq 0 \).

For \( r_{\parallel} \) and \( r_{\perp} \) both close to zero, i.e. close to the intersection of both critical surfaces, multi-critical behaviour can be expected. \( r_{\parallel}^{\perp} \ll 0 \) corresponds to a special region in the \((H_{\parallel}, T)\)-diagram, namely close to the intersection of the critical lines \( T_{c}^{\parallel} (H) \) and \( T_{c}^{\perp} (H) \), i.e. the bicritical (or tetracritical) point of Fig. 3.3. At this point both \( \gamma_{st}^{\parallel} \) and \( \gamma_{st}^{\perp} \) diverge, according to (3.28). This situation, however, deserves a more profound analysis of (3.27) in order to determine the possible fixed points close to the "multi-critical" surface. Considering the Hamiltonian (3.27) with arbitrary numbers \( n_{\parallel} \) and \( n_{\perp} \) of vector components in \( \hat{s} \) and \( \hat{s}_{s} \), respectively, Kosterlitz \( \cdots \) find to order \( \epsilon = 4-d \) in an \( \epsilon \)-expansion, the following recursion relations for the coefficients in (3.27):

\[ \cdots \]
where the functions $f_1$ and $f_2$ are defined as

$$f_1(b) = \lambda^2 \left( 1 - \frac{s^2}{8} \right)^2,$$
$$f_2(s) = \frac{\lambda^2 s}{8},$$

(3.30)

$A$ is the cutoff, as usual fixed at a constant value.

From (3.30) six fixed points have been located at the critical surface near $r_{\parallel} \approx r_{\perp}$, of which four have $w = 0$, corresponding to decoupled Hamiltonians ($\sigma^\prime$, (3.27)). All four are unstable for small perturbations of $w$ from zero, for all $n = n_{\parallel} + n_{\perp} < 11 + \epsilon \langle \epsilon \rangle$. Of the two remaining fixed points, the first one is located at

$$u^* = w^* = v^* = u = -\frac{\epsilon}{4(n+8)},$$
$$r_{\parallel} = r_{\perp} = -\frac{\epsilon}{4(n+8)},$$

(3.31)

with $\epsilon = 8\pi^2 \lambda \epsilon$, and describes bicritical behaviour. It can be seen from (3.27) with $u = v = w$ that this fixed point corresponds to a critical point where $\sigma$ and $\hat{s}$ play an identical role. This fixed point therefore leads to Heisenberg-like critical behaviour in the uniaxial case and XY-like behaviour in the orthorhombic case. It is stable for

$$n < n^X(d) = 4 - 2\epsilon + \epsilon^2,$$

where the value $n^X(d=3) \approx 3.1$. For $n > n^X(d)$ a new fixed point becomes stable, the so-called biconical fixed point $|16|$, which is not
important in this context. From their calculations Kosterlitz et al. concluded that in real antiferromagnets (with $d=3$, $n\leq 3$), the bicritical fixed point (3.30) is the only stable multi-critical fixed point, and therefore no tetracritical behaviour could be expected. The resulting types of critical behaviour in spin-flop systems have been tabulated in Table 3.2.

Table 3.2. Types of critical behaviour in spin-flop systems

<table>
<thead>
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<th>Anisotropy</th>
<th>$r_\parallel \ll r_\perp$</th>
<th>$r_\parallel \gg r_\perp$</th>
<th>$r_\parallel \approx r_\perp$</th>
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<td>XY</td>
<td>Heisenberg</td>
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The existence of tetracritical points for $n \leq 3$

Recently Bruce and Aharony [6] argued that tetracritical and bicritical behaviour are not necessarily connected with different fixed points. They pointed out that an intermediate phase and a tetracritical point can exist in a real antiferromagnet in spite of the fact that the asymptotic multi-critical behaviour is determined by the bicritical fixed point. They attributed the difference between bicritical and tetracritical behaviour to the sign of an irrelevant parameter $\nu_o$ which remains important, however small the parameter $\nu_o$ actually is.

In this subsection we shall discuss the line of arguments of Bruce and Aharony.

First, the reduced Hamiltonian (3.27) is written in a more symmetrical form as

$$\tilde{H} = -\frac{1}{2} \int \frac{(r_o\cdot q^2)(\sigma^2 \cdot |s|^2) - \frac{1}{2} s_o}{q} \int (\sigma^2 - |s|^2)$$

$$- \int \int \frac{u_o (\sigma^2 \cdot |s|^2)^2 + \nu_o \sigma^2 |s|^2}{q\cdot q'q''}.$$  

(3.32)
Now the term with \( g_0 \equiv r_{∥} - r_{⊥} \) represents the relevant quadratic term which breaks the \( n \)-fold spin-rotational symmetry shown by (3.32) for \( g_0 = 0 \). Deviations in \( u, v \) and \( w \) with respect to the bicritical fixed point at \( u^* = v^* = w^* = u_0 \) (\( \mp \); (3.30)), are irrelevant and have been omitted in (3.32). An exception is made for the \( \nu_0 \)-term in (3.32), which must be classified as a dangerous irrelevant parameter \( |6| \), leading to the difference between bicritical and tetracritical points.

From the natural \( \nu \) (corresponding to a minimum in \( \nu \), \( \nu > 0 \); (2.68)), it can be simply verified that for \( g_0 = 0 \) the \( \nu_0 \)-term induces a preferred spin orientation along or perpendicular to the easy axis \( (\hat{s} = 0 \text{ or } \sigma = 0) \) for \( \nu_0 > 0 \). On the other hand, for \( \nu_0 < 0 \) an orientation somewhere along a diagonal direction is preferred \( (\hat{s} \neq 0 \text{ and } \sigma \neq 0) \). This latter possibility corresponds to a skew ordering of the staggered magnetization with respect to the easy axis, with a coupling between \( \sigma \) and \( \hat{s} \). This is exactly what one would expect to occur in the 1-phase. Now, the main question is whether this 1-phase can remain stable even if the multi-critical point is approached. Bruce and Aharony argue that, although the \( \nu_0 \)-term decreases asymptotically, close to the multi-critical point \( (T = T_m) \), as shown by Kosterlitz et al. \( [16] \), it remains the only symmetry breaking term in (3.32) if \( g_0 = 0 \). Therefore, the preferred skew orientation will persist up to \( T_m \), which defines the multi-critical point as a tetracritical point.

In the ordered phases \( (T < T_m) \) of a tetracritical system (with \( \nu_0 < 0 \)) the direction of \( \vec{M}_{st} \) will be determined by the competition between the \( \nu_0 \)-term, which prefers a skew ordering, and the \( g_0 \)-term, which prefers an ordering parallel or perpendicular to the easy axis, for \( \nu_0 < 0 \) and \( g_0 > 0 \), respectively. Only very close to the \( g_0 = 0 \) axis the \( \nu_0 \)-term will be important enough to impose the skew ordering. Therefore the intermediate phase can be expected for \( \nu_0 < 0 \) in a small region close to the \( g_0 = 0 \) axis. With vanishing \( \nu_0 \), i.e., as the multi-critical point is approached, this region narrows asymptotically to zero. This is shown in Fig. 3.5b., which will be discussed extensively in section 3.4. For \( \nu_0 > 0 \), the quartic term has no influence on the ordering process and it consolidates the anisotropy of the quadratic \( g_0 \)-term for \( T = T_m \). This implies that the spins order along the easy axis for \( g_0 < 0 \) and nearly
perpendicular to the easy axis for $g_0 \leq 0$. At $g_0 = 0$, the spins jump from one orientation to the other. This characterizes the line $g_0 = 0$ as the spin-flop line. The corresponding $(g, t)$-diagram is shown in Fig. 3.5a. The direct connections between Figs. 3.3. and 3.5. form a main topic in section 3.4.

Finally, it is convenient to explain in the present context the difference between the above tetracritical point in spin-flop systems and the multi-critical point in mixed magnetic systems with competing spin-anisotropies, which is also called a tetracritical point. From the above discussion it will be clear, that the tetracritical point treated in this section exists owing to a coupling term of the form $\sigma^2 |s|^2$ in the (reduced) Hamiltonian. Therefore it is called a tetracritical point with coupled order-parameters, although this coupling asymptotically vanishes as the multi-critical point is approached. The corresponding multi-critical behaviour is described by the bicritical fixed point (3.30). On the other hand, the tetracritical point in mixed magnets corresponds to one of the above mentioned fixed points at $w^* = 0$, which are unstable for small perturbations of $w$. Therefore, this tetracritical point can exist only in materials where two completely independent spin systems are present, which also will order independently. This tetracritical point with decoupled order parameters is realized when both spin systems accidentally order at the same thermo-dynamic field values.

3.4. The extended-scaling theory of multi-critical behaviour in spin-flop systems

In the $(H, T)$-phase diagrams of spin-flop systems, the ordered AF-phase and SF-phase are separated from the P-phase by the critical lines $T^\perp_c (H)$ and $T^\perp_c (H)$, respectively, which meet in a multi-critical point. $T^\perp_c (H)$ is a line of critical points where the order parameter of the AF-phase, $M^\perp_{st}$, vanishes and where critical fluctuations of the parallel spin components take place. As $M^\perp_{st}$ possesses one relevant component ($n^\perp = 1$), the critical behaviour at $T^\perp_c (H)$ must be classified as Ising-like ( Fig. 2.7.). At the line of critical points $T^\perp_c (H)$ the order parameter of the SF-phase, $M^\perp_{st}$, which consists of two
components \( n_{\perp} = 2 \) and one component \( n_{\perp} = 1 \) in a system with uniaxial and orthorhombic spin anisotropy, respectively, vanishes and critical fluctuations occur of the corresponding perpendicular spin components. This critical behaviour must be classified as XY-like if \( n_{\perp} = 2 \) and as Ising-like if \( n_{\perp} = 1 \).

At the multi-critical point both \( M_{\parallel}^{\text{st}} \) and \( M_{\perp}^{\text{st}} \) vanish and critical fluctuations occur of \( n_{\parallel} + n_{\perp} \) spin components. The corresponding multi-critical behaviour is described as Heisenberg-like (if \( n_{\perp} = 2 \)) or XY-like (if \( n_{\perp} = 1 \)), both near the bicritical point in Fig. 3.3a and near the tetracritical point in Fig. 3.3b. Such a simple addition of relevant components of the order parameters at a multi-critical point is certainly not self-evident. It appears to be a special feature of the multi-critical points in spin-flop systems, as was pointed out in the RG-study of Kosterlitz et al. [16], discussed in section 3.3. They showed that \( n_{\parallel} \) and \( n_{\perp} \) may be simply added at the multi-critical point, as the coupling between \( M_{\parallel}^{\text{st}} \) and \( M_{\perp}^{\text{st}} \) asymptotically vanishes as the multi-critical point is approached, and therefore is irrelevant for the description of the multi-critical behaviour.

Although Kosterlitz et al. [16] showed that only one type of multi-critical behaviour can occur in real spin-flop systems, where \( d \leq 3 \) and \( n \leq 3 \), Bruce and Aharony indicated that in addition to the spin-flop systems displaying a bicritical point, there still may exist a class of spin-flop systems with a tetracritical point in the \((H_{||}, T)\)-diagram [16]. As has been discussed in section 3.3, the difference between both systems must be sought in the sign of a term \( (S_{\parallel})^2 (S_{\perp})^2 \) in the (reduced) Hamiltonian, which term may lead to the occurrence of an I-phase that persists if the multi-critical point is approached, although the corresponding field range narrows asymptotically to zero. This appears to be the only difference between spin-flop systems with a bicritical and a tetracritical point in the \((H_{||}, T)\)-diagram. Therefore, most of the following discussion applies to both kinds of systems.

When a spin-flop system at the multi-critical point \((H_{m}, T_{m})\) is carried away from this point along the critical line \( T_{c}^{||}(H) \), the fluctuations of the parallel spin components persist, whereas the fluctuations of the perpendicular components are gradually suppressed. The reverse
situation will take place, if the system is carried along $T_c(H)$. Starting from the high spin isotropy at the multi-critical point, a gradually increasing anisotropy is induced in this way and ultimately the critical behaviour of the system becomes fully characteristic for an anisotropic (Ising or XY) system. Therefore, one can define an anisotropy parameter $g$, which plays the role of a relevant nonordering field at the multicritical point (cf. section 2.2.), and which is a combination of the applied magnetic field $H/\parallel$ and of the temperature $T$.

This anisotropy $g$ has been first introduced as a field in a scaling analysis of the bicritical point in spin-flop systems by Riedel and Wegner [17]. Subsequently, Pfeuty et al. [18], formulated the idea in the form of the so-called extended-scaling hypothesis for the description of bicritical behaviour:

$$G(H/\parallel, H/\perp, g, t) = s^{-a} G(H/\parallel s^{1/a}, H/\perp s^{1/a}, g s^{1/a}, t s^{1/a})$$

This expression defines the Gibbs free energy at the bicritical point as a generalized homogeneous function (compare (2.41)) of two relevant ordering fields $H/\parallel$ and $H/\perp$, and of two relevant nonordering fields $g$ and $t$. $g$ and $t$ clearly play an identical role as both fields carry the system away from the multi-critical point. With $s = |t|^{-a_T}$ substituted in (3.33) and with the definitions

$$a_T = 2-a_b, \frac{a_T}{a} = \frac{\Delta/\parallel}{\Delta}, \frac{a_T}{a} = \frac{\Delta/\perp}{\Delta} \text{ and } \frac{a_T}{a} = \frac{d}{d_T}$$

one arrives at a commonly used equivalent notation for (3.33):

$$G(H/\parallel, H/\perp, g, t) = |t|^{2-a_b} s^{\frac{H/\parallel}{|t|^{\Delta/\parallel}}} s^{\frac{H/\perp}{|t|^{\Delta/\perp}}} g^{\frac{g}{|t|^d}}$$

(3.35)

Here $a_b$ is the bicritical exponent corresponding to the specific heat (cf. Table 2.1. and eq. (2.52)), $\Delta/\parallel$ and $\Delta/\perp$ are the so-called gap-exponents, which result when (3.33) is repeatedly differentiated with respect to the corresponding ordering fields $H/\parallel$ and $H/\perp$ and $d$ is the so-called crossover exponent for reasons which will become clear shortly. (3.33) or (3.35) are expected to give a good description of the critical behaviour in the vicinity of the multi-critical point, i.e.
for small values of $t$ and $g$.

The extent of the region in which (3.33) or (3.35) holds without severe corrections to the asymptotic scaling behaviour, strongly depends on the choice of the fields $g$ and $t$. Fisher [19] showed that the optimal scaling fields $\tilde{g}$ and $\tilde{t}$ for the description of the bicritical and equivalently of the "tetrcritical" behaviour are given by

\[ \tilde{g} = g - pt \quad \tilde{t} = t + qg \quad (3.36) \]

with

\[ \tilde{g} = H^2 - H^2_m \quad \tilde{t} = (T-T_m)/T_m \quad (3.37) \]

(This form will be familiar for readers who did not omit section 3.3., i.e. (3.26) with $\tilde{g} = r_H - r_H$). As discussed in section 3.3., the optimum choice of the $\tilde{g} = 0$ axis in a bicritical system obviously coincides with the spin-flop line at the bicritical point, so

\[ p = T_m (dH^2_{\text{SF}}/dT)_b \quad (3.38) \]

In a tetracritical system $p$ must be calculated by RG-techniques. The choice of the $\tilde{t} = 0$ axis is less clear. As a zero-order approximation in $\varepsilon = 4-d$, Fisher obtained for $q$ in (3.36) the estimate

\[ q(n=n_\parallel + n_\perp) = \frac{n+2}{3nT_m} \int_{-dH^2_{\parallel}} \frac{dH^2_{\parallel}}{H^2_{\parallel} = 0} \quad (3.39) \]

In Fig. 3.4. the axes corresponding to these optimum scaling fields are shown in the $(H^2_{\parallel}, T)$ phase diagram of a bicritical spin-flop system with uniaxial spin anisotropy (i.e. $n=3$). Henceforth we shall write all scaling results explicitly in terms of $\tilde{g}$ and $\tilde{t}$.

With $G$ as a generalized homogeneous function of $H^\|_{\text{st}}$, $H^\perp_{\text{st}}$, $\tilde{g}$, and $\tilde{t}$, the power laws for all thermodynamic quantities can be obtained in the usual way (i.e. section 2.5.). For instance, single differentiation of (3.33) with respect to $H^\|_{\text{st}}$ gives

\[ M^\|_{\text{st}}(\tilde{g}, \tilde{t}) = s \frac{1}{2dH^\|_{\text{st}}-d} M^\|_{\text{st}}(\tilde{g}^{1/a}, \tilde{t}^{1/\lambda T}), \text{for } H^\|_{\text{st}} = H^\perp_{\text{st}} = 0 \quad (3.40) \]
These expressions show that there are two different exponents connected with the bicritical (or similarly the tetracritical) behaviour of the order parameter in the AF-phase $M_{st}^{\parallel}$ in the $(H_T, T)$-plane, depending on the path of approach to $(H_b^2, T_b)$. $\gamma_b$ describes its $t$-variation along the path $\gamma = 0$, whereas $b/\gamma$ describes its $\gamma$-variation along the path $t = 0$. It appears to be a general property of any critical quantity, that the exponents corresponding to the $t$ and $\gamma$ variation differ by a factor $b$. 

From (3.40) two types of expressions for $M_{st}^{\parallel}$ can be obtained. With $s = |\gamma|^{-a_G}$ one arrives at
which is only interesting for \( g < 0 \), as \( M^\| = 0 \) for \( g > 0 \). With 
\[ s = |t|^{-\Delta_3} \]
substituted in (3.40), one obtains

\[
M^\|_{st}(g, t) = |t|^{-\gamma_b} M^\|_{st}(\frac{g}{|t|^{1/\phi}}, \pm 1) = |\hat{t}|^{-\gamma_b} \chi^\|_{st}(\frac{g}{|t|^{1/\phi}}), \tag{3.47}
\]

This scaling function consists of two branches, \( \chi^\|_{st} \) for \( t > 0 \) and \( \chi^\|^\perp_{st} \) for \( t < 0 \). Similar power laws and scaling functions can be derived for \( \chi^\^\perp_{st} \). By double differentiation of (3.33) with respect to \( H^\^\perp \), one obtains the parallel staggered susceptibility \( \chi^\|_{st} \). Also for this quantity one can derive power laws and scaling functions. Of these we only mention

\[
\chi^\|_{st}(g, t) = |t|^{-\gamma_b} \chi^\|_{st}(\frac{g}{|t|^{1/\phi}}, \pm 1) = |\hat{t}|^{-\gamma_b} \chi^\|_{st}(\frac{g}{|t|^{1/\phi}}), \tag{3.48}
\]

where

\[
\Delta_3 = 2 - a_b - 2\Delta_3^\|, \tag{3.49}
\]

Many similar examples can be given for other quantities. All bicritical exponents \( \tilde{\eta} \) related through exponent relations, similar to (2.30) again, and have bicritical values, which according to Kosterlitz \( \cdots \), \( |b| \) correspond to the \( d=3 \) Heisenberg values if \( n_\perp = 2 \) or the \( d=3 \) XY values if \( n_\perp = 1 \) (see Table 2.2.).

The extended-scaling expression (3.33) for \( G \) is supposed to be valid in the direct surroundings of the bicritical point. Since both the paramagnetic phase boundaries \( \tilde{\chi}^\|_c (\tilde{g}) \) and \( \tilde{\chi}^\|^\perp_c (\tilde{g}) \), and the spin-flop line \( (\tilde{g} = 0) \) join in the bicritical point, (3.33) must contain also information about the corresponding critical and first-order behaviour of all quantities near these phase transitions. For instance, for \( \tilde{g} = 0 \) we expect that \( \chi^\|_{st} \) diverges for \( \tilde{t} \rightarrow \tilde{t}^\|_c (\tilde{g}) \) with an Ising-exponent \( \gamma_b \). . .

\[
\chi^\|_{st}(g, t) = |t|^{-\gamma_b}
\]

where

\[
\hat{t} = (t - \tilde{t}^\|_c (\tilde{g}))/\tilde{t}^\|_c (0). \tag{3.48}
\]
Since this divergence of $\chi^{\parallel}_c$ should be consistent with (3.45) and the factor $t^{-\gamma_b}$ is nonsingular at $t_c(g \neq 0)$, the scaling function $X^\parallel_+(x)$, which depends on one single variable

$$x = \frac{r}{|t|^\phi},$$

must have the form

$$X^\parallel_+(x) = \left| \frac{x-x^*}{x^*} \right|^{-\gamma_b}, \text{ for } x > x^*.$$  (3.50)

Here $x^*$ corresponds to the phase boundary $\tilde{t}^\parallel_c(g)$, which determines the variation of the critical point $t^\parallel_c$ with $g$ as

$$\tilde{t}^\parallel_c(g) = (x^*)^{-1/\phi} \left| g \right|^{1/\phi}.$$  (3.51)

Usually (3.51) is written in the form

$$\tilde{t}^\parallel_c(g) = -\nu^\parallel/|t|^{\phi},$$  (3.52)

where $-\nu^\parallel = x^*$. Similarly the shape of the paramagnetic phase boundary of the SF-phase is written as

$$\tilde{t}^\perp_c(g) = -\nu^\perp/|t|^{\phi}.$$  (3.53)

So, it can be concluded that the extended-scaling hypothesis (3.33) implies a prediction for the shape of the paramagnetic phase boundaries close to the multi-critical point. At a large distance from $(H^2_m, T_m)$, where (3.33) is no longer valid, these phase boundaries will correspond to straight lines in the $(g, t^\parallel)$- and $(H^2_m, T)$-diagrams [16]. This can be seen directly from (3.29) in section 3.3. and is shown in Fig. 3.4.

The values for the crossover exponent $\phi$ in case of uniaxial and orthorhombic spin-antisotropy have been calculated with the RG-technique by Pfeuty [18] as

$$\phi = 1.175(15), \text{ for } n = 2,$$

$$\phi = 1.250(15), \text{ for } n = 3.$$  (3.54)
With $\xi > 1$, the expressions (3.52) and (3.53) show that the critical lines become tangent to the $g = 0$ axis, as $(H_m^c, T_m^c)$ is approached (Fig. 3.5.). Although the amplitudes $w_\|/w_\perp$ are not universal, their ratio is. Fisher [19] pointed out that the result

$$w_\perp/w_\| = 1, \quad \text{for } n = 2$$

(3.55)

is exact, whereas numerical estimates for $d = 3$ yield [18]

$$w_\perp/w_\| = 2.51, \quad \text{for } n = 3.$$  

(3.56)

As discussed above, these results apply to spin-flop systems with a bicritical point as well as with a tetracritical point. In the latter case a similar approach can also be applied to the second-order phase boundaries, separating the $\uparrow$-phase from the SF- and AF-phases. Bruce and Aharony [6] derived

$$g_\perp = |\tilde{\xi}|^{\tilde{\psi}} \quad \text{and} \quad g_\| = -|\tilde{\xi}|^{\tilde{\psi}},$$

(3.57)

where $\tilde{\psi} > \tilde{\psi}$. (3.58)

Therefore, also the lines $g_\perp$ ($\tilde{\xi}$) and $g_\|$ ($\tilde{\xi}$) approach the tetracritical point tangent to the $g = 0$ axis. The resulting geometry of the phase boundaries close to the bicritical and tetracritical point is shown in Fig. 3.5.

In the phase diagram of a bicritical system, to which we shall confine the further discussion, one may expect regions where the behaviour of a thermodynamic quantity, as $M_{st}$ or $\gamma_{st}$, is completely determined by the vicinity of the critical boundaries $t_\\| (g)$ or $t_\\perp (g)$, other regions where the bicritical behaviour strongly dominates (close to the $t = 0$ axis and for $\tilde{\xi} > 0$ close to the $g = 0$ axis) and regions where the first-order characteristics of the spin-flop transition become clear ($g > 0, \tilde{\xi} > 0$). These asymptotic regions will be separated by $\gamma_{st}$ regime, where two kinds of behaviour compete. A schematic picture of such a division of the $(g, \tilde{\xi})$ diagram in asymptotic and crossover regions is shown in Fig. 3.6. The crossover from one type of asymptotic...
behaviour to another can be analyzed very well on the basis of the bicritical scaling functions \[20\]. In principle any scaling function, like (3.43) to (3.45) may be used.

Consider the scaling function \(\mathcal{H}^\prime\) of (3.44) which consists of two branches

\[
\mathcal{H}^\prime(\gamma) \propto \frac{\langle \hat{g}, \hat{c} \rangle}{|\hat{c}|^b}, \quad \text{for } \hat{c} > 0 \quad (3.59a)
\]

and

\[
\mathcal{H}^\prime(\gamma) \propto \frac{\langle \hat{g}, \hat{c} \rangle}{|\hat{c}|^b}, \quad \text{for } \hat{c} < 0 \quad (3.59b)
\]
where $x$ is defined by (3.49) as $x = \frac{t}{\tilde{g}_1/t_1^2}$. In (3.42) we found the $\tilde{g}$-variation of $M_{st}$ along the $t=0$ axis as

$$M_{st}(\tilde{g}, t=0) = \tilde{g}_1^{\frac{3}{2}}b^{\frac{1}{2}}.$$  \hspace{1cm} (3.42)

This implies that both $\tilde{g}$ and $\tilde{g}_s$ must have a bicritical $x$-region for $x \approx x_c$, with the following asymptotic behaviour:

$$\tilde{g}_s(x) \approx \frac{1}{|t|^b} x^{3b/5}, \quad \text{for } x \to x_c.$$  \hspace{1cm} (3.60)

Similar to the scaling function $X_\parallel(x)$ in (3.50), also $\tilde{g}_s(x)$ must display critical behaviour close to the paramagnetic phase boundary (3.52), i.e.,

$$\tilde{g}_s(x) \approx \left[\frac{(x-x_c)/x_c}{\tilde{g}}\right]^{3b/5}, \quad \text{for } x \to x_c + \tilde{g}_s.$$  \hspace{1cm} (3.61)

With (3.41) we find as the asymptotic behaviour of $\tilde{g}_s(x)$ in the first-order region close to the $\tilde{g} = 0$ axis

$$\tilde{g}_s(x) = \frac{|t|^{3b/5}b}{|\tilde{g}|} = \text{constant, for } x \to 0.$$  \hspace{1cm} (3.62)

So, in the scaling function $\tilde{g}_s(x)$ a crossover from a bicritical exponent $3b/5$ (at large $x$) to a critical exponent $8$ (for $x \to \tilde{g}_s$) can be observed. A similar crossover behaviour can be expected in the analogous scaling function $X_\perp(x)$ for $M_{st}$. The scaling function $\tilde{g}_s(x)$ and similarly $\tilde{g}_s(x)$, will show a crossover from the bicritical exponent $3b/5$ (at large $x$) to zero (for $x \to 0$). In the $P$-phase one might use the scaling functions $X_\parallel(x)$ and $X_\perp(x)$ (3.45), to observe crossover from bicritical to critical behaviour. The crossover analysis sketched here will be employed extensively in the analysis of the (multi-) critical behaviour in CsMnBr$_3 \cdot 2$D$_2$O.

3.5. Spin-flop systems in a skew magnetic field

In the previous sections of this chapter only magnetic fields applied along the easy axis were taken into account. Here we shall consider cases in which the external field is skew, i.e., applied at some
 nonzero angle $\psi$ to the easy $z$-axis. In an actual experiment it is hard to avoid some misalignment, which inevitably results in the application of a skew field instead of a parallel field. In the following discussion we shall assume a system with orthorhombic spin anisotropy, also showing the largest variety of effects. A uniaxial system then may be considered as a system with two intermediate axes and without a hard axis.

The most familiar effect of a skew field on a spin-flop system is the disappearance of the first-order spin-flop transition, if the field is rotated from the easy ($z$) axis towards the intermediate ($y$) axis. The extent of the first-order transition at $T = 0$ has been studied in MFA [22,23] and the critical angle between $H$ and the $z$-axis was calculated as

$$\phi_c(T=0) = \arctan \left( \frac{D}{A_z - A_y - D} \right),$$  \hspace{1cm} (3.63)

where the MFA constants $D = D_z - D_y$, $A_z$ and $A_y$ have been defined already in expression (3.5). An equivalent expression in directly measurable quantities is

$$\tan \phi_c(T=0) = \left( \frac{H_{SF}(T=0)}{H_c(T=0)} \right)^2,$$  \hspace{1cm} (3.64)

which is obtained under the assumption $D \approx A_z - A_y - D_y$. When the anisotropies in the inter-sublattice interactions ($A_z - A_y$) and in the intra-sublattice interactions ($D$) are of the same order of magnitude. If $A_y \approx A_z$, $\phi_c$ can also be expressed in the commonly used exchange field ($H_{ex} \equiv A_z$) and anisotropy field ($H_A \equiv D$) as

$$\phi_c(T=0) = \frac{H_A}{2H_{ex}} \text{ rad.} \approx 28.6^\circ \frac{H_A}{H_{ex}},$$  \hspace{1cm} (3.65)

under the assumption $H_A \ll H_{ex}$ as is the case in weakly anisotropic antiferromagnets.

From (3.65) one may derive that $\phi_c$ often is a very small angle ($\approx 1.1^\circ$ in CoBr$_2$, $\approx 6H/0$, $\approx 0.4^\circ$ in MnF$_2$, $\approx 0.08^\circ$ in CsMnBr$_3$, $\approx 2H/0$ and even $\approx 0.015^\circ$ in Cr. $\phi_c$). Therefore, in order to observe the first-order character of the spin-flop transition, the magnetic field must be carefully aligned along the easy axis. For $T > 0$, the situation is
even worse \[ T_{\text{c}}(T) : T_{\text{c}}(0) (T_b - T)/T_b \] since it appears that

The first-order spin-flop "shelf" has been extensively studied in Al [24] and is depicted in Fig. 3.7.

It is bordered by two lines of critical points, indicated as \( T_{\text{c}}^{\text{SF}}(H_i) \).

If the magnetic field is applied under an angle \( \psi > \psi_{\text{c}}(T) \) with respect to the z-axis, no jump will occur in the direction of \( H_{\text{st}} \). Only a rapid reorientation will remain, which for \( \psi > \psi_{\text{c}}(T) \) can be described by the MF-expression [25]

\[
\tan(2\zeta + 2\psi) = \frac{\sin(2\psi)}{\cos(2\psi) - (H/H_{\text{SF}})^2} \tag{3.67}
\]

where \( \zeta \) is the angle between \( H_{\text{st}} \) and the z-axis (see Fig. 3.1.).

Also near the bicritical point, there are pronounced effects caused by a small \( H_i \)-component of the magnetic field. Kosterlitz et al. [16] showed that in a system with orthorhombic spin-anisotropy, the critical behaviour near the entire paramagnetic critical surface \( T_{\text{c}}(H_{\|}, H_i) \) is Ising-like, except for a single point in the \( H_i = 0 \) plane, namely the bicritical point, with XY-exponents (see Fig. 3.7.).

For fixed \( T < T_b \), the phase boundary should be a smooth curve in...
the \((H_{//},H_{\perp})\)-plane. Quite analogous to the variation of \(T_c\) with \(g\), derived in the previous section, the change in \(T_c\) at fixed \(H_{//} = H_0\) when a small field \(H_{\perp}\) is applied should vary as \(|16|\)

\[
T_c(H_{\perp}) - T_c(H_{\perp}=0) \sim |H_{\perp}|^{1/4}, \text{ for } H_{\perp} \rightarrow 0.
\] (3.68)

Since \(\alpha > 1\) (as before), the phase boundary in the \(H_{//} = H_0\) plane is tangent to the line \(H_{\perp} = 0\) as shown in Fig. 3.8. The shape of the boundaries \(T^{SF}_{c}(H_{\perp})\) and \(T_{c}(H_{\perp})\) has been verified in GaAlO\(_3\) by Rohrer and Gerber [26]. In the projection in Fig. 3.8., the bicritical point of the \((H_{//},T)\)-diagram here appears as a tetracritical rather than a bicritical point. This indicates the completely "ad hoc" terminology for multi-critical points.

If a magnetic field component \(H_h\) exists along the hard axis of an orthorhombic antiferromagnet, a completely different behaviour can be observed. In this case the first-order spin-flop phase boundary, which will have a hyperbolic shape [3], is connected to the paramagnetic phase boundary for all temperatures. This gives rise to a line of bicritical points \(T_b(H_h)\), as shown in Fig. 3.9. [27].

For a uniaxial system the \((H_{//},H_{\perp},T)\)-diagram is slightly different from Fig. 3.7. Here the perpendicular field component destroys not only the Heisenberg behaviour at the bicritical point, but also the XY-characteristics at the SF-P phase boundary. This results in the \((H_{//},H_{\perp},T)\)-phase diagram of Fig. 3.10.
Fig. 4.9. (a) $(H_{//}, H_{\perp}, T)$-phase diagram of an antiferromagnet with orthorhombic spin anisotropy.

(b) Projection of the spin-flop surface, bracketing a line of bicritical points $T_b(H_{//})$. 

Fig. 4.10.

$(H_{//}, H_{\perp}, T)$-Diagram of a uniaxial antiferromagnet.
Far too little experimental or theoretical evidence exists to provide an analogous summary for antiferromagnets that display a tetracritical point in the $H_p-T$ diagram. MF-studies [24a] at $T = 0$ show that the $I$-phase only extends in the easy-hard plane and possibly coexists with the paramagnetic phase at a tetracritical point. The $(H_p,H_r,T)$-diagram might strongly resemble Fig. 3.9(a) with the first-order surface "split up" into two second-order surfaces, which both connect to the Ising surface in one tetracritical line. However, the $I$-phase does not necessarily coexist with the paramagnetic phase at all temperatures and the picture might be much more complex.

References to Chapter III


CHAPTER IV

CRITICAL BEHAVIOUR OF CoBr$_2•6$(0.48 D$_2$O, 0.52 H$_2$O) NEAR THE NÉEL POINT

4.1. Introduction

As was described in chapter II, the present knowledge of the (quasi-) static behaviour of systems near criticality is concisely contained in the concepts of scaling and universality. In these concepts critical systems are classified by only two quantities, viz. the lattice dimensionality $d$ and the number $n$ of relevant vector components of the order parameter. In $n$-d space the case $n=2$, $d=2$ forms a curious point. It has been pointed out by Stanley and Kaplan [1] that such a system possibly shows a transition which is characterized by a divergence of the ordering susceptibility, whereas the order parameter remains zero below the transition. This so-called Stanley-Kaplan transition to a quasi-ordered state has been questioned by many authors. In the calculations of a number of critical quantities considerable difficulties are encountered in the $n=2$, $d=2$ limit and large differences are found in the results from different calculational methods. Therefore, Betts et al. [2] conclude that $d=2$ XY-systems near criticality appear to form a particular class, in which universality possibly is violated.

In this respect it appeared worthwhile to start an investigation of the critical behaviour in the antiferromagnet CoBr$_2•6$H$_2$O, which is known as a good magnetic approximation of the $d=2$ XY-model [3-5]. The transition to $d=3$ long-range order (LRO) observed at $T_N \approx 3.15$ K [4] must be due to small deviations from the $d=2$ XY-model, as the ideal model cannot show LRO [6]. In order to search for $d=2$ XY-features and to investigate the mechanism that triggers the transition to LRO, quasi-elastic neutron scattering experiments have been performed. For several reasons, elucidated in the next two sections, a partly deuterated sample was selected for the present work. Some preliminary results have been presented at the International Conference on Magnetism 1976 [7]. The content of this chapter has been published in a somewhat different form in the proceedings of the International Symposium on Neutron Inelastic Scattering in Vienna, 1977 [8].
4.2. Crystallography and Magnetic Interactions

The structure of CoBr$_2$·6H$_2$O is described by the monoclinic, face-centered space group C 2/m, with unit-cell dimensions: a = 11.00 Å, b = 7.16 Å, c = 6.90 Å and $\beta = 124^\circ$. The unit cell contains two formula units. Cobalt ions are situated on inversion centers and are octahedrally coordinated by four oxygen and two bromine ions (Fig. 4.1a).

The isolated [CoBr$_2$O$_4$] clusters show approximately fourfold symmetry around the Br-Co-Br axis, which is close to the $a^\circ$-axis (Fig. 4.1b).

The perfect cleavage of the crystals

**Fig. 4.1.** Details of the crystal structure of CoBr$_2$·6H$_2$O. Cobalt atoms are black, bromine atoms shaded and oxygen atoms are represented by open circles.

a) The face-centered arrangement of the [CoBr$_2$O$_4$]-octahedra in the ab-plane. $d_1$, $d_2$ and $d_3$ indicate exchange paths in the ab-plane. Both the chemical and magnetic coupling between adjacent ab-planes appear to be relatively weak.

b) Sketch of the ac-mirror plane containing the hard (a) and easy (c) axes of the susceptibility tensor |$\chi$|. The intermediate (b) axis coincides with the unique b-axis ($\chi^B_{13}$, $\chi^B_{11}$).
parallel to the (001) plane indicates that the chemical bonding between adjacent ab-planes is relatively weak. The completely deuterated material, on the other hand, undergoes a crystallographic phase transition from monoclinic C 2/m to a triclinic structure with space group P 1 between 77 ° and 4.2 K [10]. The triclinic deformation of the unit cell and the changes in the ionic positions within the cell are rather small. Previous NMR and AFMR experiments on CoBr₂·6(xD₂O(1-x)H₂O) with varying x, denoted as CBx, indicate that at T=1.2 K the monoclinic-triclinic transition occurs at x ≈ 0.55 [11]. The anticipated monoclinic-triclinic phase boundary in xT-space is shown in Fig. 4.2.

Fig. 4.2.
Anticipated crystallographic phase diagram of the system CoBr₂·6(xD₂O(1-x)H₂O) with varying x. The arrow indicates the composition of the sample chosen for the present neutron scattering study.

At this phase boundary a spontaneous and reversible twinning of the crystal occurs, so that in the triclinic phase four kinds of crystallographic domains exist which in pairs show mirror symmetry with respect to the original monoclinic ac-mirror plane [10].

The magnetic moments of the Co²⁺-ions in CBx are ordered antiferromagnetically below the Néel temperature T_N ≈ 3.1 K. As the Co-ions are located at a twofold axis, in the C 2/m structure the direction of the moments is either perpendicular or parallel to the b-axis.

In the purely hydrated material the preferred direction (γ-axis) of the sublattice magnetization is in the ac-plane at 80° from the c-axis towards the c°-axis (Fig. 4.1b) [3]. The array of magnetic moments is shown in Fig. 4.3a and can be described by the magnetic space group C₂c 2'/m' [12]. In the systems with a triclinic structure (x > 0.55) the magnetic moments are rotated away from the ac-plane towards the b-axis over an angle φ [11]. The division in two sublattices is
essentially the same as in the monoclinic structure (Fig. 4.3) and the corresponding magnetic space group is $P_{2_1}$ [10]. The variation of $\phi$ as a function of $x$ is shown in Fig. 4.4.

**Fig. 4.3.** Arrays of magnetic moments in the system CBx.

a) In the monoclinic structure the easy axis is in the ac-mirror plane at approximately $8^\circ$ from the c-axis towards the c-axis [3].

b) In the triclinic structure the easy axis is rotated over an angle $\phi$ out of the ac-plane towards the b-axis [10,11].

**Fig. 4.4.** Rotation angle $\phi$ of the easy axis out of the ac-plane as a function of the deuterium fraction $x$. From Hijmans et al. [11].

The large variation in the angle $\phi$ as a result of only slight changes in the surrounding octahedron of the Co-ion indicates that the xy-plane in CBx is a plane with a weak spin anisotropy. The existence of this easy plane and the corresponding XY-character of CBx has mainly been detected by susceptibility measurements [3,13]. In terms of an effective spin $S' = |\rangle$ with anisotropic g-value and exchange parameter $J$, Hijmans et al. [3] obtained $g_\gamma = 4.70$, $g_\beta = 4.82$ and $g_\alpha = 2.02$ (subscripts $\alpha$, $\beta$ and $\gamma$ refer to the principal axes of the susceptibility tensor,
which are indicated in Fig. 4.1b). Taking into account the orbital contributions to the \( g \)-values, they arrive at \( (J_\alpha/J_\gamma) = 0.29 \) and \( (J_\gamma/J_\beta) = 0.98 \). These values clearly indicate that the \( \beta \)-plane is an easy plane in which the anisotropy is only small.

The spatial dimensionality of the magnetic system has been discussed by Haseda [14]. There are two features which are of importance, i.e., the interactions in the centered ab-plane and the interactions between the planes. As is shown in Fig. 4.1a, a number of possible exchange interactions within the ab-planes may be distinguished. Analyzing the nature of the exchange paths, Haseda concluded that \( J_2 \) and \( J_3 \) probably would be small compared with \( J_1 \). From specific heat measurements Kopringa et al. [14] obtained \( 2(J_2+J_3) \approx 1.0 \text{ K} \) compared with \( 4J_1 = 19.4 \text{ K} \). The exchange interaction \( J' \) between the ab-planes was estimated by Haseda to range between 0.01 \( \text{ K} \) and 0.1 \( \text{ K} \) [14]. The ratio \( J'/J_1 \approx 10^{-2} \) indicates the \( d^2 \) character of \( \text{CB}x \).

4.3. Experimental

Because of the large cross section of hydrogen for incoherent scattering, in neutron diffraction experiments a purely deuterated sample is preferred. However, in the present experiment a partly deuterated sample has been used to avoid difficulties due to the presence of domains in the triclinic structure for \( x > 0.55 \). Single crystals were grown by slow evaporation at room temperature from a saturated solution of \( \text{CoBr}_2 \) in a mixture of 50\% \( \text{D}_2\text{O} \) and 50\% \( \text{H}_2\text{O} \). Typical dimensions of the samples were \( 2 \times 1.5 \times 0.5 \text{ cm}^3 \). To check on the crystallographic structure, part of the product was used to record a neutron diffraction powder diagram at 4.2 K. The anticipated \( C \ 2/m \) structure was readily confirmed, with unit-cell dimensions \( a = 10.973(3) \text{ Å} \), \( b = 7.120(2) \text{ Å} \), \( c = 6.838(1) \text{ Å} \) and \( \beta = 124.92(1)^\circ \). From refinement of the effective scattering length at the deuterium positions, a value \( x = 0.483(8) \) was obtained. This sample will further be denoted as \( \text{CB}48 \).

The experiments have been performed on a two-axis diffractometer at the Petten HFR reactor. Neutron wavelengths of 2.570 Å and 1.163 Å, corresponding to neutron energies of 12.4 meV and 60.5 meV respectively, were obtained from a Cu (111) monochromator. In the first case a
pyrolytic graphite filter was used to diminish higher order wavelength contaminations. Soller slits with a horizontal divergence of 30° were placed before the monochromator and in front of the BF3-detector. Vertical collimation was defined by 60° soller slits between monochromator and sample and in front of the detector. Temperature control was achieved by regulating the vapour pressure of the He-bath in which the sample was immersed. Pressure could be kept constant for half a day (typical time required for a complete scan of the critical scattering) within 0.5–1.0 mm Hg, corresponding to an uncertainty of 1.0 mK in the temperature region near $T_N$.

4.4. Temperature dependence of the staggered magnetization

From the general expression for the (quasi-) static magnetic scattering cross section, presented in eq. (2.19), it can be derived that the LRO in CB48, described by the magnetic space group $C_{2v}^{2}$ and shown in Fig. 4.3a, leads to magnetic Bragg reflections $(h,k,l)$ with $(h+k)=2n+1$ and $l=(2n+1)/2$, indexed with respect to the crystallographic unit cell. The intensities of these reflections can be derived from eq. (2.19) as

$$I(Q,T) = |f(Q)|^2 \sum_{\alpha} (1-Q_{\alpha}^2)[M_{st}^{\alpha}(T)]^2,$$

where $\alpha$ now refers to the principal axes $\alpha$, $\beta$ and $\gamma$ of the susceptibility tensor (cf. Fig. 4.1b) and $M_{st}^{\alpha}$ is the $\alpha$-component of the staggered magnetization. Because of the transformation properties of the space group C 2/m nuclear reflections are confined to reciprocal lattice points with $h+k=2n$.

Our measurements were restricted to the $a*c*$-reciprocal lattice plane, which is shown in Fig. 4.5. The temperature dependence of $M_{st}$ has been determined from the intensity variation of a few intense magnetic Bragg reflections with $T$. In order to check on extinction effects, measurements were performed on both a large (1.5 cm$^3$) and a small (0.1 cm$^3$) single crystal. From this it was concluded that even in the large sample extinction is negligible. Most data of $M_{st}(T)$ were obtained from the top intensities of magnetic Bragg peaks, corrected for
Fig. 4.5. The \(a^*c^*\)-reciprocal lattice plane of CB48. Dots and squares correspond to magnetic and nuclear reflections, respectively. Also the easy \(\gamma\)-axis is indicated. \(q = \frac{q}{2\pi}\) represents the deviation of \(q\) from the nearest magnetic Bragg reflection and is used in section 4.6.

The background and second-order nuclear contributions, which were determined above \(T_N^*\). Correction of the magnetic intensities for critical-scattering contributions appeared to be significant only very close to \(T_N^*\). In this temperature region a sufficient number of critical scattering profiles was recorded to determine the correction at the \((10\bar{1})\) reflection by interpolation. Typical values are 50\% for \(t = \left|1-T/T_N^*\right| = 10^{-6}\), 30\% for \(t = 10^{-3}\) and only 3\% for \(t = 10^{-2}\). From the values at the \((10\bar{1})\) reflection the correction for other reflections was calculated.

To compare the observed temperature variation of \(M_{st}\) with the predicted single-power law behaviour (cf. section 2.3) and to obtain estimates for the parameters \(T_{N^*}\), \(\beta\) and \(B\), least-squares fits of

\[
I(T) = \left[\frac{M_{st}(T)}{M_{st}(0)}\right]^2 = B^2 t^{2\beta}
\]

(4.2)

to the data were performed. First the \(t\)-range in which (4.2) holds, i.e. the extent of the critical region has been determined. Our approach to this problem is based on the requirement that different fits, in which data points within the interval \(0 < t \leq t_{\text{max}}\) are taken into account, must yield the same estimates for \(\beta\) and \(B\), independent of the specific choice of \(t_{\text{max}}\) up to an upper value of \(t_{\text{max}}\), which then
corresponds to the edge of the critical region. This requirement will be fulfilled only if the correct value of the parameter $T_N$ is used.

The procedure is illustrated in Fig. 4.6, where for several choices of $T_N$ the resulting estimate for $\beta$ is shown as a function of $t_{\text{max}}$. From these results one may conclude that (4.2) gives an adequate description of the data for $t < 0.015(5)$ and $T_N = 3.1175(10) K$.

![Fig. 4.6.](image)

Values for the exponent $\beta$, obtained from least-squares fits of (4.2) to the observed $M_2^s(T)$, where $T_N$ was fixed and only data with $t < t_{\text{max}}$ were taken into account. Eq. (4.2) gives an adequate description of the data for $t < 0.015(5)$ and $T_N = 3.1175(10) K$.

The experimental data for $M_2^s(T)$ are shown in the double-logarithmic plot in Fig. 4.7, together with the best fit of (4.2) to the data with $t < 0.015$ and $T_N$ fixed at 3.1175 K. For this best fit the estimates $\beta = 0.326(5)$ and $B = 1.7(2)$ are obtained. The uncertainty in $B$ is mainly caused by the extrapolation of $M_2^s(T)$ to $T=0$ over a temperature region of 0.45 $T_N$. For $t > 0.02$ the data clearly show a deviation from the power law. However, the significance of attributing an additional $\beta$-value to the data in the range $0.02 < t < 0.2$ is doubtful. A further discussion of these results will be given in section 4.6., in combination with the results of the critical scattering analyses.
4.5. Critical scattering near $T_\mu$

Theory

In section 2.3. the phenomenon of critical scattering near critical points has been introduced. We stated that the scattering of neutrons by fluctuating spins is quasi-elastic \cite{15} and quasi-static \cite{16}, the (quasi-elastic) scattering cross section and the static susceptibility tensor $\chi$ are related by (cf. (2.19) and (2.25))

$$\frac{d\sigma}{d\Omega} = T |\ell(Q)|^2 \sum_{\alpha} (1 - Q_{\alpha}^2) \chi^{\alpha\alpha}(Q)$$

(4.3)

For the $Q$-dependence of $\chi$ several approximants have been proposed. Here a slightly modified version of the isotropic first approximant, proposed by Fisher and Burford \cite{17} and introduced earlier as (2.20),
will be used, namely

\[ \chi^{\alpha\alpha}(q) = \chi^{\alpha\alpha}(\vec{q}) = \left\{ \chi_{st} \cdot \left[ 1 + (q_a/\kappa_a)^2 + (q_b/\kappa_b)^2 + (q_c/\kappa_c)^2 \right]^{-1+\eta/2} \right\} \]

which applies to a low-dimensional system. In (4.4) \( \vec{q} = \vec{Q} - 2\pi \hat{r} \), the deviation vector (see Fig. 4.5.). The staggered susceptibility is defined by \( \chi_{st} = \chi(\vec{Q} = 2\pi \hat{r}) \) and \( \kappa \) is the anisotropic inverse correlation length of the fluctuations. In general a different \( \chi_{st}^{\alpha\alpha} \) and inverse correlation lengths \( \kappa^{\alpha\alpha} \) are found for each spin component. Again \( \eta \) describes the deviation from the simple Lorentzian shape of \( \chi(q) \) which is predicted by Ornstein-Zernike theory.

**Spin anisotropy**

In order to select the most suitable magnetic reflections for the observations on spin components parallel and perpendicular to the easy \( \gamma \)-axis, the direction of the latter in the ac-plane of CB48 had to be determined. From the variation of the critical field \( H_{IA} \) at which the spin system enters the I-phase, as a function of the direction of the external magnetic field applied in the ac-plane, the \( \gamma \)-axis has been determined at 7.6(5)° from the c-axis towards the c-axis (see Fig. 5.5.). From Fig. 4.5, it can be seen that at the \( (10^3/2) \) reflection the angle between \( \vec{Q} \) and the easy axis is only 3°. Therefore, only perpendicular spin components should be observable in that case.

Fig. 4.8. shows some observed intensities in scans parallel to the \( \alpha \)-axis through the \( (10^3/2) \) reflection for \( T > T_N \). Even for \( T = 3.122 \) K.

![Observed intensities in scans parallel to the \( \alpha \)-axis through the \( (10^3/2) \) reflection](image)
very close to $T_N = 3.1175 \text{ K}$, no critical scattering could be observed. At most a slight elevation of the background with respect to the $4.24 \text{ K}$ level is visible. From this it may be concluded that all perpendicular spin fluctuations are absent or very small. In contrast, the longitudinal spin fluctuations, observed near the $(10^{1/2})$ reflection, are very pronounced for $T > T_N$, as we shall see below. Apparently, the small spin anisotropy in the easy $c\bar{c}$-plane is sufficient to suppress fluctuations in $S_{\alpha \alpha}$ for $T < T < 4.24 \text{ K}$. Therefore, the transition at $T_N$ must be characterized as Ising-like ($n=1$) rather than $XY$-like ($n=2$). A further discussion of this result will follow in section 4.6.

Profile analysis of $\chi_{\gamma\gamma}$

The $q$-dependence of the longitudinal fluctuations as a function of temperature has been determined near the $(10^{1/2})$ reflection, both above and below $T_N$. In reciprocal space $q$-scans were made parallel to the $a$-axis and the $c\bar{c}$-axis, to determine the correlation lengths $1/\kappa_a$ within the $ab$-layers and $1/\kappa_{cc}$ between the layers. Fig. 4.9. shows some typical observed critical scattering profiles for $T > T_N$.

These experimentally observed intensity distributions in $q$-space are a convolution of the scattering cross section (4.3) and the instrumental resolution function

$$I(q) = T|f(q)|^2 \sum_\alpha \int dq \chi^{\alpha \alpha}(q) R(q-q_0) \quad (4.5)$$

The resolution function $R(q-q_0)$ for a two-crystal spectrometer may be expressed by

$$R(q-q_0) = R_0 \exp \left\{-\frac{3}{2} \sum_{i=1}^{3} M_{ij}^*(q_i-q_{0i})(q_j-q_{0j}) \right\} \quad (4.6)$$

For each $q_0$ the matrix elements $M_{ij}(q_0)$, which describe the shape and orientation of the corresponding resolution ellipsoid, can be calculated $|18|$. As a check on this calculation, several actual resolution ellipses in the $ac$-plane were mapped out, using Bragg reflections as a probe. A few typical results are shown in Fig. 4.10.
Fig. 4.9. Typical critical scattering profiles for $T > T_N$, observed near the $(10\frac{1}{2})$ reflection in scans parallel to the $a$- and $c^*$-axis ($\lambda = 2.570 \ \text{Å}$). Solid lines are best fits of (4.5) to the data with $\chi^{aa} = \chi^{BB} = 0$. For each $T$ both scans were fitted simultaneously using the same value of $\chi_{st} = \chi^{YY}(q=0)$.

The observed intensity profiles near the $(10\frac{1}{2})$ reflection were fitted with the theoretical expression (4.5), where $\chi^{aa}$ and $\chi^{BB}$ were taken zero in agreement with the conclusion from the measurements near the $(10\frac{3}{2})$ reflection. The three-dimensional integral of (4.5) cannot be solved analytically, but Yelon et al. [19] pointed out that, with expression (4.6) inserted for $\chi^{YY}$, it can be reduced to a one-dimensional integration. In the deconvolution program, which has been employed, the final integration has been performed in a Gaussian quadratures calculation [20]. For each point $\mathbf{Q}_0$ of a scan the magnetic form factor $f(Q_0)$, the factor $(1-\mathbf{Q}_0^2)$ and the resolution ellipsoid $R(\mathbf{Q}_0)$ were calculated. In the least-squares fits scans parallel and perpendicular to the $a$-axis were treated simultaneously with the
Fig. 4.10.
Comparison between two observed and calculated resolution ellipses in the ac-plane. Circles denote data with an intensity half the peak intensity. Solid lines are the calculated 50% ellipses.

quantities \( \chi_{st} \equiv \chi^{YY}(q=0), \kappa_a, \kappa_c \), and the intensity of the second-order Bragg peak as free parameters. Since the same path elements of the dominant exchange interaction \( J_1 \) are involved in the spin correlations along the \( a \)- and \( b \)-axis (Fig. 4.1a.), the numbers of correlated spins along these axes were assumed to be equal. Therefore, we have inserted \( \kappa_a/\kappa_b = b/a \) in eq. (4.4), where \( 1/\kappa \) is expressed in \( \AA \) and not in numbers of spins. In the fits the background, which is very high due to the large fraction of hydrogen in the sample, was approximated by a second-order polynomial in \( q_a \) and \( q_c \). In the final determination of the critical scattering parameters the background and (for \( T > T_N \)) the second-order nuclear Bragg peak have been fixed at their average values. As expected, the obtained estimates for \( \chi_{st} \) and \( \kappa \) are strongly correlated. No serious attempt could be undertaken to determine a deviation of the profiles from Ornstein-Zernike theory (\( r=0 \) in (4.4)), as for this purpose much better counting statistics and higher resolution are required.
Results

From the profile analysis the temperature dependence of both the intralayer correlation length ($1/\kappa_a$) and the interlayer correlation length $1/\kappa_c^*$ could be determined in the entire range $2.9 \, K < T < 4.25 \, K$, in which measurements were performed. Both correlations show the same temperature dependence as is reflected in the constant ratios $\kappa_c^*/\kappa_a \approx 3.7$ for $T > T_N$ and $\kappa_c^*/\kappa_a \approx 2.8$ for $T < T_N$. (If only one scan was available for a fit, the ratio $\kappa_c^*/\kappa_a$ was fixed to this value).

This behaviour means that the transition to LRO in CB48 is essentially three dimensional in character over this temperature range. The only remainder of the $d=2$ characteristics of CBx at higher temperature [5] is that the numbers of correlated spins along the a-axis ($N_a$) and along the b-axis ($N_b$, assumed to be equal to $N_a$) for $T > T_N$ are $3.6(3)$ times as large as the number of correlated spins along the c*-axis, perpendicular to the ab-plane ($N_c^*$). This can be easily calculated from

$$\frac{N_a}{N_c^*} = \frac{c \sin \beta}{\frac{1}{a}} \frac{\kappa_c^*}{\kappa_a}.$$ 

The temperature dependences of the inverse correlation lengths and of the staggered susceptibility have been compared with the single-power laws

$$\chi_{st}(T) = \Gamma T^{-\gamma}$$

and

$$\kappa_{\chi}(T) = K_x T^{\nu} \quad x = a, c^*$$

both for $T > T_N$ and for $T < T_N$. Primed symbols will be used to indicate the quantities obtained for $T < T_N$. Least-squares fits of (4.7) and (4.8) to the data with $t < 0.02$ yield an average value for the ordering temperature $T_N = 3.11(1) \, K$, which is in perfect agreement with the value determined from the staggered-magnetization data.

Figs. 4.11. and 4.12. present the temperature dependence of the inverse intralayer ($\kappa_a$) and interlayer ($\kappa_c^*$) correlation lengths for $T > T_N$ and $T < T_N$, respectively. The identical behaviour of $\kappa_a$ en $\kappa_c^*$...
Fig. 4.11. Dependence of the inverse intralayer ($\kappa_a$) and interlayer ($\kappa_c^*$) correlation lengths on $t$ for $T > T_N$. Circles and squares are obtained with $\lambda = 2.570 \, \AA$ and $\lambda = 1.163 \, \AA$, respectively. The solid lines are least-squares fits of (4.8) to the data, with $\gamma_d$ fixed at $2.1175 \, K$.

in this respect is obvious. Solid lines are least-squares fits of (4.8) to the data, with again $T_N$ fixed at $3.1175 \, K$ and $t_{\text{max}} = 0.02$. The resulting $\gamma$-values are 0.66(8) for $\kappa_a$ and 0.67(4) for $\kappa_c^*$. With $\gamma$ fixed at the value $\gamma = 0.67$, the amplitudes $\kappa_a = 0.28(1)$ and $\kappa_c^* = 1.01(3)$ are obtained. For $T > T_N$ the exponents $\gamma' = 0.65(11)$ and $\gamma'' = 0.69(6)$ result for $\kappa'_a$ and $\kappa''_c$, respectively. With $\gamma'$ fixed at 0.67 one obtains $\kappa'_a = 0.65(3)$ and $\kappa''_c = 1.81(6)$.

The staggered susceptibility data for $T > T_N$ and $T < T_N$ are shown in Fig. 4.13. Data from different samples were brought on the same scale by a proportionality factor. The drawn lines correspond to (4.7) fitted with $T_N = 3.1175 \, K$ and $t_{\text{max}} = 0.02$. The corresponding exponents are $\gamma = 1.18(7)$ for $T > T_N$ and $\gamma' = 1.16(13)$ for $T < T_N$. From a fit
Fig. 4.12. Dependence of $\chi'_a$ and $\chi'_o$ on $t$ ($T < T_\nu$). Compare Fig. 4.11.

For the sake of clearness the $\chi'_a$ data have been shifted low along the vertical axis over one decade. Solid lines are fits of (4.7) to the data, with $T_\nu$ fixed at 4.1175 K. See further Fig. 4.11.
with $\gamma = \gamma' = 1.17$ the ratio of the amplitudes was obtained to be $
abla/\nabla' = 4.0(3)$.

For $t > 0.02$ both the correlation lengths and the staggered susceptibility data appear to deviate markedly from the single-power law behaviour. The region in which these quantities follow a power-law behaviour is roughly equal to the observed critical region of the staggered magnetization (cf. section 4.4.), in agreement with the theoretical predictions [25]. However, there may be severe systematic errors in the values obtained for $k$ and $\chi_{st}$ in the region $t > 0.02$, as they strongly rely on a correct determination of the high background.

4.6. Conclusions

The critical region of CB48 has been investigated by detailed quasi-elastic neutron scattering experiments. The critical scattering data were analysed within the quasi-static approximation [16]. As our results do not show systematic differences between $k$-values derived from data taken with neutrons of different energies (Figs. 4.11. and 4.12.), this approximation appears to hold for the experiments that were described.

Although the system CBx is a good approximation of the $d=2$ XY-model at high temperature $T \gg T_N$ [3–5], the present experiment shows that the critical behaviour in the temperature range $0 < t < 0.02$ may be described as $d=3$ Ising-like. The $d=3$ character can be derived from the identical temperature dependence of the interlayer and intralayer correlation lengths, which indicates that both correlations play an identical role in the ordering process. Also the critical exponents that were obtained, certainly exclude a $d=2$ ordering (cf. Table 4.1.). In the critical region $t < 0.02$ the only remainder of the low-dimensional characteristics of CBx is, that the number of correlated spins along a direction in the ab-plane is a factor 3 to 4 larger than the number perpendicular to the layers. A possible crossover to $d=2$ behaviour at $t > 0.02$ could not be observed. The experimental data for $\chi_{st}$ and $k$ show even an opposite tendency. However, from this observation no clear conclusions may be drawn, since the results for $\chi_{st}$ and $k$ at $t > 0.02$ might be affected by severe systematic errors, as we noted.
already in the previous section.

### Table 4.1. Critical exponents in CB48, compared with some model systems

<table>
<thead>
<tr>
<th>System/Region</th>
<th>$\beta$</th>
<th>$\gamma$</th>
<th>$\gamma'$</th>
<th>$\nu$</th>
<th>$\nu'$</th>
<th>$\eta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CB48</td>
<td>0.326(5)</td>
<td>1.18(7)</td>
<td>1.16(13)</td>
<td>0.67(4)</td>
<td>0.67(6)</td>
<td>$\gamma$</td>
</tr>
<tr>
<td>$d=3\ n=1$</td>
<td>0.325(1)</td>
<td>1.240(1)</td>
<td>1.240(1)</td>
<td>0.630(1)</td>
<td>0.630(1)</td>
<td>0.0315(25)</td>
</tr>
<tr>
<td>$d=3\ n=2$</td>
<td>0.346(5)</td>
<td>1.316(9)</td>
<td>1.316(9)</td>
<td>0.669(3)</td>
<td>0.669(3)</td>
<td>0.032(15)</td>
</tr>
<tr>
<td>$d=2\ n=1$</td>
<td>0.348(7)</td>
<td>1.318(10)</td>
<td>1.318(10)</td>
<td>0.670(6)</td>
<td>0.670(6)</td>
<td>0.04(1)</td>
</tr>
</tbody>
</table>

From the critical exponents one cannot distinguish if the ordering in CB48 is Ising-like or XY-like, as the differences between the theoretical values are only small. However, the Ising-character of the ordering may be deduced from the non-divergence of the spin fluctuations perpendicular to the easy axis. Apparently the small exchange anisotropy in the $xy$-plane is sufficient to suppress spin fluctuations along the $z$-axis. A similar behaviour was reported for MnF$_2$, where also a small (unaxial) anisotropy is present [16]. The perpendicular spin fluctuations do not diverge in MnF$_2$ either, but they could be observed. To estimate the magnitude of the perpendicular spin fluctuations in CB48, a crude comparison can be made.

In MnF$_2$ $\chi_{\perp}^{\perp}$ and $\chi_{\parallel}$, related to the perpendicular spin fluctuations, are almost temperature independent for $t < 0.01$ and roughly equal to the longitudinal values for $t \approx 5 \times 10^{-2}$. If the same properties are assumed in CB48, at most a broad profile ($\chi_{\perp}^{\perp} \approx 0.03 \AA^{-1}$) of low intensity, comparable with the upper profiles in Fig. 4.9., would result even at $T_N$. Moreover, only spin fluctuations along the $z$-axis probably would contribute to $\chi_{\perp}^{\perp}$ in CB48. Therefore, it is not surprising that near the $(10^3/2)$ reflection only a slight elevation of the background was observed.
Within the experimental errors several exponent relations are ful-
filled in CB48, such as $\gamma = \gamma', \nu = \nu'$ and $\gamma = (2-\eta)\nu$. A good estimate of $\eta$ can be derived from the values of $\beta$ and $\nu$ that were obtained. From the exponent relation $\eta = 28/\nu$, a value $\eta = -0.03(6)$ results. This value and the obvious $d=3$ characteristics of the critical scattering in CB48, justify the profile analysis with $\eta=0$ a posteriori.

Besides the critical exponents, also the obtained critical amplitudes are in good agreement with the theoretical values for the $d=3$ Ising model. The amplitude $B = 1.7(2)$ may be compared with the calculated value $B = 1.569(3)$ for a simple cubic lattice with $S = \frac{1}{2} |22|$. The experimental value $4.0(3)$ for the ratio $\Gamma/\Gamma'$ may be compared with the theoretical values $5.07$ (from series expansions) and $4.80$ (from the $\epsilon$-expansion, cf. Table 2.3.).

In conclusion it may be stated that in CB48 below 4.2 K both the aniso-
tropy in the XY-plane and the interlayer interactions already exert their influences on the ordering, and no $d=2$ XY-features can be recovered for $t < 0.5$.

4.7. Appendix

Weighing procedure in single-power law fits

In least-squares fits of a single-power law

$$I = I_o t^a$$  \hspace{1cm} (A4.1)

to a number of data points $(I_i, t_i)$, it is important that also the uncertainty in $t_i$ is taken into account in a correct way. Whereas the data usually have a comparable uncertainty in $T$, the relative error in $t$ increases rapidly as $T_i$ approaches $T_c$. In a double-logarithmic plot around each datapoint an "error ellipse" may be drawn with $\sigma_y$ and $\sigma_x$ as main axes. Here $\sigma_y$ and $\sigma_x$ are the uncertainties in $y_i = \ln I_i$ and in $x_i = \ln t_i$, respectively. All ellipses, which are congruent with the error ellipse at $(y_i, x_i)$ can be denoted as

$$\frac{(y-y_i)^2}{\sigma_y^2} + \frac{(x-x_i)^2}{\sigma_x^2} = g^2.$$  \hspace{1cm} (A4.2)
One of these ellipses, indicated with \( g = g^*_1 \), will touch the straight line, which represents the power law (A4.1). The here applied weighing procedure now minimizes the sum \( \sum_i g_i^2 \).

A direct consequence of this method is that for each choice of \( T_c \) the weights must be recalculated as the \( \sigma_x \) depend on \( T_c \). Although the present procedure results in larger uncertainties in the estimates of \( I_0, T_c \) and \( \sigma \) in (A4.1), it is certainly more realistic than a method in which a constant \( \sigma_x \) or no \( \sigma_x \) at all is used.

References to chapter IV


D.J. Wallace in "Phase Transitions and Critical Phenomena" Vol.6 (C. Domb and M.S. Green, Eds.). (Academic Press, New York, 1976), Table VI.


CHAPTER V

TETRACRITICAL BEHAVIOUR OF CoBr$_2$ $\cdot$ 6 $\{0.48$ $\text{D}_2\text{O}, 0.52$ $\text{H}_2\text{O}\}$

5.1. Introduction

CoBr$_2$ $\cdot$ 6H$_2$O has been reported to show a spin-flop transition in a parallel field at $H_{SF} \approx 7.5$ kOe (for $T=0)$ $[1,2]$. This rather low value is due to the weak spin anisotropy in the $b\gamma$-plane, which we discussed to some extent in section 4.2. In view of the convenient location of the bicritical point at ($T_b \approx 2.90$ K, $H_b \approx 9.31$ kOe) $[1,2]$, it appeared attractive to perform a neutron scattering study on the bicritical behaviour in this material.

Purpose of the present experiment was to study the intensity variations for several magnetic Bragg reflections as a function of a parallel magnetic field and the temperature. From these results we wanted to determine the magnetic phase diagram and to test the smoothness hypothesis $[3]$ along the paramagnetic phase boundaries $T_C^\uparrow(H)$ and $T_C^\downarrow(H)$.

As a well-defined easy axis is crucial to perform such a study, we had to avoid the domain structure, which is present in the triclinic phase for deuterium fractions $x > 0.55$ (Fig. 4.2.). On the other hand, a diminishing of the incoherent neutron scattering due to hydrogen atoms would be preferable. Therefore, a sample with $x = 0.48$ (CB48) was used, like in the zero-field experiment described in chapter IV.

To our surprise, the magnetic phase diagram for CB48 appeared to be essentially different from the reported diagram for CoBr$_2$ $\cdot$ 6H$_2$O $[2]$. The present neutron scattering experiment provides strong evidence that in CB48 the AF-phase and the SF-phase are separated by an intermediate phase and not by the usual spin-flop transition. To our knowledge this is the first time that an I-phase with coupled order parameters is reported for an antiferromagnetic system.
5.2. Experimental

As mentioned in section 4.5., the \( \gamma \)-axis of CB48 is situated in the \( a^\ast c^\ast \)-mirror plane, but its direction within this plane does not coincide with one of the crystallographic axes (Fig. 4.5.). This is a substantial handicap for the performance of neutron scattering experiments in a parallel magnetic field. Obviously, the possibility to use a vertical magnetic field along the easy \( \gamma \)-axis will be excluded, since in general no magnetic Bragg reflections will be observable in the horizontal \( a^\beta \)-plane. Therefore, we had to use a geometry with a horizontal magnetic field and the \( a^\ast c^\ast \) reciprocal-lattice plane, which - as was shown in section 4.4. - contains many magnetic reflections, as scattering plane. This, however, has the disadvantage that many reflections cannot be measured because of the interception of the incoming or scattered neutron beam by the poles of the magnet (Fig. 5.1a.). As can be seen from Fig. 5.1b. only a small part of the \( a^\ast c^\ast \)-plane can be investigated, the extent depending on the neutron wavelength used.

The present experiment has been performed on the two-axis diffractometer HB5 at the Petten HFR reactor. A neutron wavelength of 1.163 Å has been used, obtained from a Cu(111) monochromator. The sample was a large single crystal of \( 2 \times 1.5 \times 0.5 \text{ cm}^3 \) which was shaped roughly ellipsoidal to reduce inhomogeneities in the demagnetizing field. The sample was mounted in a liquid-\( \text{He} \) bath cryostat, which was mounted on a conventional magnet with \( \vec{H} \) horizontal. The cryostat + sample could be rotated around a vertical axis with respect to the magnet. The inhomogeneity of the magnetic field over the sample was better than 0.5%. Temperature control within 1-2 mK was achieved by regulating the vapour pressure of the He-bath.

Special care has been taken to obtain an accurate horizontal orientation of the magnetic field \( \vec{H} \) and the \( a^\ast c^\ast \) reciprocal-lattice plane. The deviation of \( \vec{H} \) from horizontal due to imperfections in the construction of the diffractometer has been determined to be smaller than 4'. The \( a^\ast c^\ast \)-plane has been adjusted horizontally with 10' vertical collimators between monochromator and sample and in front of the BF3 detector. For this purpose the intensities of several sets of Bragg
Fig. 5.1.

(a) Schematic drawing of the scattering geometry with the horizontal magnetic field \( \mathbf{H} \) along the easy \( \gamma \)-axis and the scattering vector \( \mathbf{k} \) in the \( (30^\circ/2) \) direction. \( \mathbf{k}_0 \) and \( \mathbf{k} \) indicate the incoming and scattered neutron beam with \( \lambda = 1.163 \AA \).

(b) Reciprocal lattice of CB48. Dots and squares indicate magnetic and nuclear reflections, respectively. The shaded and doubly shaded areas correspond to the attainable part of the reciprocal space for \( \lambda = 1.163 \AA \) and \( \lambda = 2.670 \AA \), respectively, when \( \mathbf{H} \) is directed along the easy axis.

reflections have been optimized. In this way a horizontal orientation of the \( a^*c^* \)-plane has been achieved within 2'-3'. Therefore the maximum angle between \( \mathbf{H} \) and the easy-hard \( a^*c^* \)-plane has been estimated as 7'. Optimum orientation of \( \mathbf{H} \) along the easy axis within the \( a^*c^* \)-plane has been obtained by rotating \( \mathbf{H} \) with respect to the sample (see section 5.4.).

With \( \mathbf{H} \) along the easy axis, all attainable magnetic Bragg reflections (Fig. 5.1b.) have weak intensities in the AF-phase, since the corres-
ponding factors \((1-\tilde{Q}^2)\) are small \((c.\text{ eq.}(4.1))\). To determine the temperature and field dependence of the order parameter in the AF-phase, \(i.e. M_{\parallel}^{\text{st}} = M_{\parallel}^{\text{AF}}\), the intensity variation of the weak \((10\overline{2})\) reflection has been studied. The \((H,T)\) behaviour of the order parameter in the SF-phase, \(i.e. M_{\perp}^{\text{st}} = M_{\perp}^{\text{SF}}\), can be determined directly from the intensity variation of the \((10\overline{3})\)-reflection. With \((1-\tilde{Q}^2) = 1\) and \((1-\tilde{Q}^2) \gg 0\), only \(M_{\perp}^{\text{st}}\) will contribute to the magnetic intensity of this Bragg reflection. Finally, for the observation of the homogeneous magnetization \(M(H,T)\), which gives a small magnetic contribution at the same reciprocal lattice points where nuclear Bragg peaks occur, the weak nuclear \((00\overline{1})\) reflection has been used.

5.3. Determination of the paramagnetic phase boundaries

The locations of the AF-P phase boundary \(T_{c}(H)\) and the SF-P phase boundary \(T_{c}(H)\) in the \(H,T\)-diagram have been obtained from the disappearance of the order parameters \(M_{\parallel}^{\text{st}}\) and \(M_{\perp}^{\text{st}}\) respectively. For the determination of \(M_{\perp}^{\text{st}}(H,T)\) the temperature variation of the \((10\overline{3})\) intensity has been observed at several \(H\)-values. After correction of the data for the background and second-order nuclear scattering, which were determined far above \(T_{N}\), for each \(H\)-value least-squares fits of

\[
I(T) = \left[\frac{M_{\perp}^{\text{st}}(T)}{M_{\perp}^{\text{st}}(0)}\right]^2 = B^2(t)^{2\beta}\]

(5.1)

to the data have been performed. Here \(t = \frac{(1-T/T_{c}^\perp)}{T_{c}^\perp}\) and \(B, \beta\) and \(T_{c}^\perp\) were treated as free parameters. The fitting procedure described in chapter IV has been used.

The results for fits in which all data have been taken into account, are shown in Fig. 5.2. The corresponding \(\beta\)-values and critical temperatures \(T_{c}^\perp(H)\) are collected in Table 5.1. If only data with \(t < 0.035\) are taken into account, the resulting \(\beta\)-values are slightly larger (average value \(\overline{\beta} = 0.32(2)\)) but the results for \(T_{c}^\perp\) are not significantly different from the tabulated values. In accordance with the smoothness hypothesis \(|3|\), no systematic change in \(\beta\) is observed along the phase-boundary \(T_{c}^\perp(H)\). The weighted average value \(\overline{\beta} = 0.31(1)\) is in good agreement with predicted values for the \(d=3\) Ising model.
Fig. 5.2. Double-logarithmic plot of the observed $(\frac{H}{H_c})^2$ data at several $H$-values as a function of $t = 1-T/T_c^1(H)$. The solid line corresponds to the average value from least-squares fits $28\pm 0.62(2)$.

(cf. Table 4.1.). The slight increase of $T_c^1$ with $H$ has been observed also in the purely hydrated compound [2].

Table 5.1. Estimates of $T_c^1(H)$ and $\delta_\perp$ from least-squares fits of (5.1) to the $(10\bar{3}/2)$ intensities at constant $H$.

<table>
<thead>
<tr>
<th>$H$(kOe)</th>
<th>$T_c^1$(K)</th>
<th>$\delta_\perp$</th>
</tr>
</thead>
<tbody>
<tr>
<td>15.00</td>
<td>2.870(2)</td>
<td>0.306(12)</td>
</tr>
<tr>
<td>14.27</td>
<td>2.871(1)</td>
<td>0.311(7)</td>
</tr>
<tr>
<td>13.29</td>
<td>2.871(2)</td>
<td>0.313(11)</td>
</tr>
<tr>
<td>11.99</td>
<td>2.867(2)</td>
<td>0.306(13)</td>
</tr>
<tr>
<td>11.00</td>
<td>2.861(2)</td>
<td>0.308(14)</td>
</tr>
<tr>
<td>9.91</td>
<td>2.855(2)</td>
<td>0.331(19)</td>
</tr>
</tbody>
</table>

For the determination of $T_c^\parallel(H)$ the temperature dependence of the weak magnetic intensity of the $(10\bar{3}/2)$ reflection has been determined at $H = 2.50$ kOe and $H = 8.00$ kOe. These measurements were very time-
consuming and the data appeared not accurate. A least-squares fit of

\[ I(T) = \left| \frac{M''(T)}{M''(0)} \right|^2 = B^2 \left| 1 - T/T_c^\parallel \right|^2 \]  

(5.2)

with \( B, \beta_\parallel \) and \( T_c^\parallel (H) \) simultaneously variable. Therefore, \( \beta_\parallel \) was fixed at the theoretical value 0.325, in agreement with the observed value at the Néel point (Table 4.1). Fig. 5.3 shows the fits of (5.2) to the observed intensities at both \( H \)-values, which obviously are well described by this value of \( \beta \). The corresponding values of \( T_c^\parallel \) are tabulated in Table 5.2.

![Fig. 5.3. Double-logarithmic plot of the observed \((M''^\parallel)^2\) data at \( H = 8.00 \) kOe and \( H = 2.50 \) kOe. The solid lines represent best fits of eq. (5.2) to the data with \( \beta_\parallel \) fixed at 0.325.](image)

At five constant temperatures \( T < T_N \) the variation of the top intensity of the \((10\bar{5}/2)\) reflection with \( H \) has been recorded. Although the data of these field scans are too inaccurate to extract the critical exponent \( \beta_\parallel \) from them, they do yield rough estimates for the corresponding \( H_c^\parallel(\bar{T}) \). These values are also collected in Table 5.2.

5.4. The transition to the SF-phase

For the observation of the AF-SF transition the variation in the peak intensity of the \((10\bar{5}/2)\) reflection, proportional to \((M_{st}^\parallel)^2\), has been recorded in field scans at constant temperature. A typical result for \( T << T_N \) is shown in Fig. 5.4. In the AF-phase \((H < H_{1A})\) the
6.2. Estimates of $T_c(h)$ and $\beta_H$ from the $(10^5/2)$ intensity.

(see main text)

<table>
<thead>
<tr>
<th>$H$(kOe)</th>
<th>$T_c(K)$</th>
<th>$\beta_H$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>3.1175(10)</td>
<td>0.326(5)</td>
</tr>
<tr>
<td>2.50</td>
<td>3.101(4)</td>
<td>0.325 fixed</td>
</tr>
<tr>
<td>4.75(25)</td>
<td>3.048(2)</td>
<td>-</td>
</tr>
<tr>
<td>6.25(25)</td>
<td>2.997(2)</td>
<td>-</td>
</tr>
<tr>
<td>7.25(25)</td>
<td>2.946(2)</td>
<td>-</td>
</tr>
<tr>
<td>8.00</td>
<td>2.915(5)</td>
<td>0.325 fixed</td>
</tr>
<tr>
<td>8.25(25)</td>
<td>2.898(2)</td>
<td>-</td>
</tr>
<tr>
<td>9.00(15)</td>
<td>2.848(2)</td>
<td>-</td>
</tr>
</tbody>
</table>

observed intensity hardly exceeds the background, as was expected from $1-Q^2 \approx 0$. Between $H_{IA}$ and $H_{IS}$ a linear increase in intensity is observed and in the SF-phase ($H > H_{IS}$) the intensity stays almost constant up to the maximum applied field of 15 kOe.

Fig. 5.4.

Top intensity of the $(10^3/2)$ reflection, observed in a field scan at $T = 1.743$ K. $\omega$ indicates the angle between $H$ and the $c^*$-axis.

Determination of the easy axis

The precise direction of the easy axis in the $a^*c^*$-plane can be determined from the variation of $H_{IA}$ and $H_{IS}$ when the applied magnetic field is rotated in this plane. $H_{IA}(\omega)$ is shown in Fig. 5.5., where $\omega$
represents the angle between $\vec{H}$ and the $c^*$-axis. The observed dependence is symmetrical around $\omega_o = 27.3(5)^\circ$ and closely resembles a cosecant function. The $\omega$-dependence of $H_{IS}$ is completely similar over the entire scanned $\omega$-range. From symmetry considerations the direction of the easy axis must be identified with $\omega_o$, i.e. at $7.6(5)^\circ$ from the $c$-axis towards the $c^*$-axis (cf. Fig. 5.1b.). This value hardly differs from the direction reported for the hydrated compound $|4|$. The approximate cosecant behaviour of $H_{IA}(\omega)$ and $H_{IS}(\omega)$ indicates that the component of $H$ along the hard $\alpha$-axis has little influence on these critical fields.

Fig. 5.5.
Dependence of $H_{IA}$ on $\omega$, symmetrical around $\omega_o = 27.3(5)^\circ$. The dash-dotted line corresponds to $H_{IA}(\omega) = 7.85 \text{ kOe}/\cos(\omega-\omega_o)$.

Character of the AF-SF transition

At a first-order spin flop transition a linear increase in intensity is expected over a small range of external fields due to demagnetization effects $|5|$. However, the field range $H_{IS} - H_{IA} \approx 1500 \text{ Oe}$ where linearity is observed at $T = 1.743 \text{ K}$, amply exceeds the maximum value of $\Delta H$ which can be expected from demagnetization. In general this
field range $\Delta H$ is well predicted by MF-calculations $|6|$. Taking the $\chi_\perp$ value reported for the hydrated material $|4|$ and applying the maximum demagnetization factor $N(4\pi)$, one arrives in MF-approximation at

$$\Delta H_{\text{max}}(T=0) = 4\pi \chi_\perp H_{\text{SF}}(T=0) \approx 160 \text{ Oe}.$$  

(5.3)

Frequently, the origin of a discrepancy between the observed and calculated $\Delta H$ is due to a deviation of $\hat{H}$ from the easy-hard plane. If the mismatch angle $\psi$ between $\hat{H}$ and the easy-hard plane exceeds the critical angle $\psi_c$ $|7|$, no first-order transition takes place and a broadening of the field range in which the spin rotation takes place, will be observed (cf. section 3.5.). This appeared to be the case in our experiment on the spin-flop system CsMnBr$_3$-2D$_2$O (CMB) $|8|$, which we shall report in chapter VI. Here we shall show that this explanation is certainly not appropriate for the discrepancy in CB48.

As we discussed in section 3.5., the rotation of $\hat{M}_{\text{st}}$ can be described by the MF-expression

$$\tan(2\zeta + 2\psi) = \frac{\sin 2\psi}{\cos 2\psi - (H/H_{\text{SF}})^2},$$  

(5.4)

if the angle between $\hat{H}$ and the $\alpha\gamma$-plane, which corresponds to $\psi = \arctan(H_y/H_y)$, is much greater than the critical angle ($\psi \gg \psi_c$). Here $\zeta$ is the angle between $\hat{M}_{\text{st}}$ and the easy $\gamma$-axis. The corresponding field dependence of the $(10^3/2)$ intensity is then given by

$$I = (M_{\text{st}}^2)^2 = M_{\text{st}}^2 \sin^2 \zeta.$$  

(5.5)

In Fig. 5.6. the observed intensity variation at 1.743 K is compared with the MF-predictions for two values of $\psi$. Here we assumed $M_{\text{st}}^2$ = constant over the shown field range, which will not induce gross errors in a weakly anisotropic system with $H_{\text{SF}}(T=0) \ll H_c(T=0)$. It can be seen in Fig. 3.1. that the sublattice magnetizations will remain almost antiparallel during the rotation, as can be calculated in MF-approximation from
Fig. 5.6.

Accurately determined shape of the increase in top intensity of the $(10 \frac{1}{2})$ reflection compared with the molecular-field predictions for two values of the misalignment angle $\psi$ between $\mathbf{H}$ and the easy axis.

\[ H_{st}^2 = H_o^2 \cos^2 \delta \gtrsim H_{o}^2 \left(1 - \frac{H_{2}^2}{H_{c}^2}\right) \]  

(5.6)

for $T=0$ (compare Table 3.1.). The failure of the description of the observed intensity variation by (5.5) is concluded from the following facts.

- The agreement in Fig. 5.6. is very poor indeed, whereas in CHB the rotation of $H_{st}$ is perfectly described by (5.5) (compare Fig. 6.10.). Even on a highly enlarged field scale the intensity increase in CB48 appears to be really linear, in contrast to the calculated curves.

- To account for a field range $\Delta H$ of 1.5 kOe for the rotation of $H_{st}$ from an AF- to an SF-ordering, a mismatch angle $\psi$ of approximately 50° must be assumed. Such a severe misalignment can be absolutely discarded, as we discussed at length in section 5.2., where the mismatch in this experiment was estimated as $\psi \approx 7°$. Moreover, $\psi = \arctan(H_{2}/H_{c})$ would vary clearly in case of a severe misalignment if $H$ is rotated with respect to the sample. Therefore one would expect then clear changes in the ratio $(H_{S} - H_{A})/H_{A}$ as a function of $\omega$ which, however,
have not been observed in the entire scanned range $-5^\circ < \omega < 50^\circ$.

![Graph showing field dependence of intensities](image)

**Fig. 5.7.**

*Observed field dependence of the top intensities of the $(10^3/2)$ and the $(00\bar{1})$ reflection at $T = 2.096$ K. The two spikes in the $(00\bar{1})$ intensity near $H_{IA}$ and $H_{IS}$ are of nuclear origin and indicate the presence of crystallographic changes.*

The anomalous behaviour of this transition in CB48 is also indicated by the observed field dependence of the peak intensity of the $(00\bar{1})$ reflection, shown in Fig. 5.7. for $T = 2.096$ K. At a normal first-order spin-flop transition the nuclear and magnetic intensities are additive and an increase $\Delta I$ in the magnetic contribution to the reflection is expected. An expression similar to (4.1) shows that this magnetic contribution is quadratic in $M$, so $\Delta I$ is proportional to $\Delta M^2$, where $\Delta M$ is the discontinuous change in the homogeneous magnetization at the transition. Here, however, two spikes are present in $I(00\bar{1})$, approximately at $H_{IA}$ and $H_{IS}$. These cannot be from purely magnetic origin since the negative derivatives $dI/dH$ would correspond to a
negative susceptibility $\frac{dM}{dH}$. Apparently by the application of an external magnetic field, crystallographic changes are induced in CB48 which are large enough to be observable.

**Evidence for the presence of an intermediate state**

Referring to our discussion in chapter III, an alternative explanation of the broad H-range, in which the rotation of $\vec{M}$ takes place, automatically emerges, namely the presence of an intermediate I-phase.

In this case $H_{IA}$ and $H_{IS}$ must be considered as the critical fields separating the I-phase from the AF- and the SF-phase respectively.

In addition to the above-mentioned arguments, there are several other indications which support the suggestion of an intermediate phase in CB48.

1. MF-theory predicts a linear increase of the $(10^3/2)$ intensity (see Fig. 5.6.) with H in an intermediate phase. In MF-approximation $|9|$ (see Table 3.1.), the rotation of the sublattice magnetizations in the I-phase is described by

$$\sin^2 \zeta = \frac{(h - \sqrt{Q})}{(-R\sqrt{Q})}$$

$$\sin^2 \delta = \frac{Q(h - \sqrt{Q})}{(-R\sqrt{Q})},$$

in the stability range

$$\sqrt{Q} < h < \sqrt{Q(1-l)}.$$

Here $h = \mu H/(A_z + A_y + D)$, $Q = (A_z - A_y + D)/(A_z + A_y + D)$, $R = +2D/(A_z + A_y + D)$ and $D = D_z - D_y$. The inter-sublattice coupling $\hat{A}$ and the intra-sublattice coupling $\hat{B}$ have been defined in chapter III, eq. (3.8).

In a weakly anisotropic system $Q \ll 1$, so that

$$\sin^2 \delta \ll \sin^2 \zeta.$$

This means that during the rotation the sublattice magnetizations remain almost antiparallel. Therefore the magnitude of $M_{st}$ may be considered as a constant and
which expresses the linearity of $I(10^{3/2})$ as a function of $H$.

2. The extent of the field range $H_{IS} - H_{IA} \approx 1500$ Oe is quite acceptable in the case of an I-phase. Accepting the idea of an I-phase for a moment, we can calculate the anisotropy in $A$ and $D$ from the observed fields $H_{IA}$ and $H_{IS}$, in combination with the reported value of $H_c$ in the hydrated compound [2]. From Table 3.1. we take the MF-expressions

$$\mu_{H_{IA}} = \left[ (A_x - A_y + D)(A_x - A_y + D) \right]^{1/3}$$  \hspace{1cm} (5.11)

$$\mu_{H_{IS}} = (A_x + A_y - D) \left[ (A_x - A_y + D)/(A_x + A_y + D) \right]^{1/3}$$  \hspace{1cm} (5.12)

and

$$\mu_{H_c} = (A_x + A_y - D).$$  \hspace{1cm} (5.13)

For $T = 1.743$ K we observed $H_{IA} = 7.35$ kOe and $H_{IS} = 8.95$ kOe. From [2] we take $H_c(1.743$ K) = 47.5 kOe. With these values inserted in (5.11) - (5.13) we obtain

$$A_y/A_x = 0.77 \hspace{1cm} (D_z - D_y)/A_z = -0.17$$  \hspace{1cm} (5.14)

and

$$A_z = 4.14 K.$$  \hspace{1cm} (5.15)

If we use extrapolated values for $T = 0$, viz. $H_{IA} \approx 6.1$ kOe, $H_{IS} \approx 7.7$ kOe and $H_c \approx 54$ kOe, the ratios (5.14) hardly change, whereas $A_z$ is increased to $A_z = 4.60$ K. These values can be compared with the reported results in CoBr$_2$·6H$_2$O [10]

$$A_y/A_x = 0.95 \hspace{1cm} D_z/A_x \approx -0.10 \hspace{1cm} and \hspace{1cm} A_z = 4.80 K.$$  \hspace{1cm} (5.16)

The anisotropy in $D$ is not known. Although the anisotropy in $A$ for the hydrated material is clearly smaller than the calculated value (5.14), the results (5.14) and (5.15) appear to be quite acceptable, indeed.

3. In case of an I-phase, $H_{IA}$ and $H_{IS}$ are second-order phase boundaries. Then the slightly increased intensity in the AF-phase at $H \approx H_{IA}$ is

\[ I(10^{3/2}) = \left( H_{IA} \right)^2 = H_{IA}^2 \sin^2 \theta = hV, \]  \hspace{1cm} (5.10)
(cf. Fig. 5.6.) can be interpreted as critical scattering from fluctuations of spin components along the intermediate axis.

(Near a first-order transition no critical scattering would occur.) Similarly the slightly decreased intensity for $H > H_{IS}$ can be considered as an effect of fluctuations in $M^{\parallel}$, reducing the component $M_{st}^{\perp}$ which is visible.

4. As we have seen in section 4.2., a crystallographic phase transition takes place in the system CBx near $x = 0.55$ for low $T$. Therefore it is not astonishing, that the monoclinic crystal structure of CB48 is quite unstable. Apparently, the magnetic and electrostatic interactions in CB48 correspond to comparable energies, since we are able to induce crystallographic changes by the application of a magnetic field. The presence of magneto-elastic coupling in CB48, makes the occurrence of an intermediate phase very conceivable. As is derived in the Appendix this coupling gives rise to terms of the form $\frac{1}{2} S_{b}^{2} S_{\perp}^{2}$ in the free energy, which couples the order parameters $M_{st}^{\parallel}$ and $M_{st}^{\perp}$. As was pointed out by Bruce and Aharony [11], the presence of a (strong) term of this form in the Hamiltonian is a necessary condition for the appearance of an I-phase, where $M_{st}^{\parallel}$ and $M_{st}^{\perp}$ are ordered simultaneously (cf. section 3.3.).

From these arguments at least a strong suspicion emerges that in CB48 no simple spin-flop transition occurs but that an intermediate phase exists between the AF- and SF-phase. Only the verification whether the intensity increase at $H < H_{IA}$ and the decrease at $H > H_{IS}$ are due to critical scattering indeed, would be a completely conclusive argument. Unfortunately, such an experiment has not been performed yet.

5.5. The magnetic phase diagram

Fig. 5.8. shows the intensity variation of the magnetic $\langle 103/2 \rangle$ reflection ($= (M_{st}^{\perp})^{2}$), as a function of $H$ and $T$. From this picture a good impression is obtained of the location of the I-phase in the HT phase diagram. The corresponding critical lines $H_{IA}(T)$ and $H_{IS}(T)$ are also shown in the $H^2$ vs. $T$ diagram of Fig. 5.9., together with the paramagnetic phase boundaries $T_{c}^{\parallel}(H)$ and $T_{c}^{\perp}(H)$. From these data the
tetracritical point may be determined at $H_t = 9.2(2) \, \text{kOe}$, $T_t = 2.82(2) \, \text{K}$. Although the data are not numerous and accurate enough to permit serious numerical tests of the predictions of RG-theory for tetracritical behaviour, a rough comparison between the theory and the present experimental results can be made.

As we discussed extensively in section 3.4., the shape of the paramagnetic phase boundaries close to the multi-critical point in a spin-flop system with orthorhombic spin anisotropy ($n=2$), like CB48, is given by

$$
\tilde{g}_1 = -\nu_\parallel \tilde{\tau}^\parallel \quad \text{and} \quad \tilde{g}_1 = +\nu_\perp \tilde{\tau}^\perp \quad (5.16)
$$
Fig. 5.9. Observed $H_{zz}^2$ vs. $T$ diagram of CB48. Estimated scaling axes $\tilde{g}=0$ and $\tilde{t}=0$ are indicated, together with Fisher's estimate of the $\tilde{g}=0$ axis [13]. The tetracritical point is located at $H_c = 9.2(2)$ kOe, $T_c = 2.82(2)$ K.

with $\psi_\parallel = +\psi_\perp$. The boundaries of the I-phase are [11]

$$\tilde{g}_2 = -\psi_\parallel \tilde{\phi}_2 \quad \text{and} \quad \tilde{g}_2 = +\psi_\perp \tilde{\psi}_2$$

(5.17)

with $\psi_\parallel = +\psi_\perp$. Bruce and Aharony [11] pointed out that

$$\psi_2 > \phi.$$  

(5.18)

For a rough comparison between theory and experiment, one can determine the directions of the optimum $\tilde{g}=0$ and $\tilde{t}=0$ scaling axes using the predicted symmetry for both pairs of phase boundaries (5.16) and (5.17). The estimates of both axes are shown in Fig. 5.9. Also Fisher's
estimate \(|12|\) for the direction of the \(\mathbf{\hat{c}}=0\) axis is shown, according to (3.39) taken with a slope \((n+2)/3n = 2/3\) as large as the slope \([dT_c/d(H^2)]_{H=0}\) of the paramagnetic phase boundary \(T_c(H^2)\) at \(H=0\).

The difference between the two estimates is possibly due to the \(d=2\) character of CB48. In a low-dimensional system the MF-type of approach which leads to Fisher's estimate \(|12|\) is not entirely appropriate.

Along the major part of \(T_c(H^2)\), the MF-prediction of a straight line appears to hold. As expected, deviations from MF-theory become apparent only very close to the tetracritical point. As in many spin-flop systems \(|13|\), it is hardly visible that \(T_c(H^2)\) comes in tangent to the \(\mathbf{\hat{c}}=0\) axis. Finally, it can be concluded that the theoretical prediction (5.18) appears to hold in CB48, as the \(I\)-phase boundaries start to approach the \(\mathbf{\hat{c}}=0\) axis at a much larger distance from the tetracritical point than the paramagnetic phase boundaries do.

5.6. Discussion

In this neutron-scattering experiment on CB48 it was found that the transition from the AF-phase to the SF-phase extends over an anomalously broad range for the applied magnetic field. The existence of this broad field range has been confirmed recently by preliminary magnetization measurements on a number of small single crystals grown from the same solution as the sample used in the neutron-scattering study \(|14|\). As we have shown in section 5.4., the observations cannot be explained if one assumes a first-order spin-flop transition. Moreover, in the same field range anomalous intensity changes for a nuclear reflection were observed, which indicate a variation in the crystallographic structure. We showed that these observations are strong indications for the presence of an intermediate phase in CB48, although the evidence is certainly not exhaustive. Many questions still remain unanswered and deserve further investigation.

We have not enough information to our disposal to decide which kind of crystallographic changes occur in CB48 at \(H_{IA}\) and \(H_{IS}\). Group-theoretical arguments indicate that the crystallographic unit cell must undergo a triclinic distortion (whatever small) in case of an \(I\)-phase. This would imply a (small) shift in the peak positions of
all Bragg reflections and possibly a variation of the nuclear intensities. Unfortunately we have no data to verify whether a shift in the peak positions actually occurs. It is possible indeed, to explain the spike in the (001) intensity (Fig. 5.7.) near $H_{IA}$ as the effect of a small ($=2\gamma$) increase of the nuclear intensity combined with a slight shift in peak position. If the system is assumed to return to a monoclinic symmetry in the SF-phase again, the reverse process should occur near $H_{IS}$. If the shifts are only small, the effect on the recorded (10$^\parallel/2$) intensities can remain hidden in the experimental scatter, as much shorter counting times were used than for the study of the (001) reflection. This explanation is speculative and the line of reasoning is heavily based on the assumption that the magnetic moment rotation and the crystallographic distortion are directly related.

It appears to be very probable that in CB48 a subtle equilibrium exists between a weak magnetic anisotropy in the $B_\parallel$-plane and an easily distorted crystallographic structure. The crystallographic instability of CB48 might be expected from the proximity of the monoclinic-triclinic phase transition at $x \approx 0.55$ for low $T$ (Fig. 4.2.), which we discussed to some extent in section 4.2. Apparently the intermediate phase in CB48 and the triclinic phase of CBx for $x > 0.55$ are closely related. Whereas in CB48 a rotation of $\hat{M}_{st}$ appears to induce crystallographic changes, the reverse process occurs in CBx for $x > 0.55$, where in zero-field $\hat{M}_{st}$ gradually rotates as a function of $x$ (see Fig. 4.4.), due to an increasing triclinic distortion of the crystallographic unit cell [15,16]. Recently, calculations were reported on a cubic antiferromagnet with externally applied stress [17], which yield results completely comparable with our observations in CB48.

From the similarity of the magnetic ordering in the 1-phase of CB48 and in the triclinic phase of CBx in zero-field, one might imagine that these systems are part of the same phase in a (three-dimensional) $xHT$-diagram. To verify this assertion and to study how in CBx the behaviour changes from bicritical (at $x=0$) into tetracritical (at $x=0.48$), it would be interesting to determine the HT-phase diagrams.
of a series of samples with varying $x$. Such an experimental determination of the entire $x$HT-diagram of CB$x$ can provide deeper insight in the relations between different kinds of critical and multi-critical behaviour [18].

Summarizing the results of the present experiment one may conclude that at least strong evidence is found for the existence of an I-phase in CB48. To our knowledge, this would be the first time that an intermediate phase with coupled order parameters is observed in an antiferromagnetic system. This coupling is probably due to magneto-elastic coupling. A further investigation of criticality near the intermediate phase boundaries $H_{IA}(T)$ and $H_{IS}(T)$, near the paramagnetic phase boundaries $T_c^\parallel(H)$ and $T_c^\perp(H)$ and close to the corresponding tetracritical point, would be straightforward. However, the determination of $H_{St}^\parallel(H,T)$ would be time consuming if the same instrumental geometry is used as in the present experiment. In conclusion it can be stated that this neutron-scattering experiment on CB48 in a parallel magnetic field indicates, that in the series of spin-flop systems CB$x$ with varying $x$ a large variety of magnetic phase diagrams may be expected.
5.7. Appendix

Coupling of \( H_{st}^{||} \) and \( H_{st}^\perp \) via magneto-elastic coupling

Magneto-elastic coupling exists to some extent in any magnetic system, as a result of the fact that various magnetic interactions, for instance exchange interactions, spin-orbit coupling and dipole-dipole interactions depend on the interionic distances. In addition to shifts in the ionic coordinates within each unit cell, so-called "internal" magnetostrictive coupling, also a change in the macroscopic crystal dimensions, i.e., "external" magnetostriction and a modified crystal symmetry may result. This phenomenon probably takes place near the critical fields \( H_{IA} \) and \( H_{IS} \), observed in CB48 (Fig. 5.7.). In this appendix we shall not discuss this observation in CB48. Here, we merely show how a coupling term of the form \( S^z_1 S^z_2 \) in the Hamiltonian can originate from magneto-elastic coupling.

The treatment of magnetostriction and internal strain in an antiferromagnet is taken to follow similar lines as the corresponding treatment in ferromagnetic materials, but with the contribution from each sublattice to the magneto-elastic energy being additively combined. For each sublattice we can write down the usual expression for the energy density

\[
E = E_{el} + E_{m.e}
\]  

(A5.1)

where \( E_{el} \) is the energy density due to internal strain and \( E_{m.e} \) is the magneto-elastic coupling term. In the classical static theory of magnetostriction, the sublattice magnetization is coupled to the uniform macroscopic strain by terms in \( E \), involving polynomials in the strains multiplied by polynomials in the direction cosines of the (sublattice) magnetization. We shall follow a similar, but quantum-mechanical procedure, taken from Callen and Callen [19, 20], in which the spin and elastic modes are coupled in the Hamiltonian rather than in the free energy.

After [19] the purely elastic term in the Hamiltonian can be written as
where the standard strain components $\varepsilon$ are presented in Table 5.3. $c_{\mu\nu}$ are elastic constants. The lowest-order contributions to the magneto-elastic coupling term $\mathcal{H}_{\text{m.e}}$ are linear in the strain components and of zero and second degree in the spin components. Terms linear in the spin components can be excluded, since they are not symmetric under time reversal. For our purpose it is sufficient to consider only one sublattice and to confine ourselves to the one-ion magneto-elastic coupling. Then $\mathcal{H}_{\text{m.e}}$ takes the form

$$\mathcal{H}_{\text{m.e}} = \sum_{\mu, \nu=1}^{6} \varepsilon_\mu c_{\mu\nu} \xi_\nu$$

Table 5.3. Elastic and magnetic basis functions in the elastic and magneto-elastic energy terms, taken from [19]

<table>
<thead>
<tr>
<th>$\mu$</th>
<th>$\varepsilon_\mu$</th>
<th>$\sigma_\mu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\varepsilon_{11}$</td>
<td>$S_x^2$</td>
</tr>
<tr>
<td>2</td>
<td>$\varepsilon_{22}$</td>
<td>$S_y^2$</td>
</tr>
<tr>
<td>3</td>
<td>$\varepsilon_{33}$</td>
<td>$S_z^2$</td>
</tr>
<tr>
<td>4</td>
<td>$\varepsilon_{23}$</td>
<td>$1(S_y S_z + S_z S_y)$</td>
</tr>
<tr>
<td>5</td>
<td>$\varepsilon_{13}$</td>
<td>$1(S_x S_z + S_z S_x)$</td>
</tr>
<tr>
<td>6</td>
<td>$\varepsilon_{12}$</td>
<td>$1(S_x S_y + S_y S_x)$</td>
</tr>
</tbody>
</table>

Here $\sigma_\nu$ represent the six basis functions for the spin components, collected in Table 5.3., and $B_{\mu\nu}$ are the one-ion magneto-elastic coupling constants. In the spin basis functions products like $[S_x S_y - S_y S_x] = iS_z$ are absent, since they are of first rather than second degree.

Both terms (A5.2) and (A5.3) can be added to the familiar magnetic Hamiltonian, consisting of an anisotropic exchange coupling term and a Zeeman term, for instance. In a MF-type of approach we can find
the ground-state error through an initial minimization of the expectation value \( \langle \mathcal{O} \rangle \) with respect to the strain components \( \varepsilon_{ij} \). This corresponds to the more familiar procedure of a minimization of (A5.1) with respect to the macroscopic strains \( |\sigma| \). Without going into details it can be seen that the expressions for the equilibrium strains, resulting from

\[
\frac{\partial \langle \mathcal{O} \rangle}{\partial \varepsilon_{ij}} = \sum_{ij} C_{ij} \varepsilon_{ij} + \sum_{ij} B_{ij} \langle \sigma_{ij} \rangle = 0 \quad (A5.4)
\]

will be linear combinations of the various \( \langle \sigma_{ij} \rangle \). With these expressions substituted, \( \mathcal{E}_{el} \) and \( \mathcal{E}_{me} \) will consist of terms of the form \( \langle \sigma_{ij} \rangle \langle \sigma_{kl} \rangle \), which represent a coupling between the order parameters of the AF-phase and the SF-phase, indeed.

References to chapter V

[18] Such a study has been performed on the system K$_2$Mn$_{1-x}$Fe$_x$F$_4$.
CHAPTER VI

EXPERIMENTAL STUDY OF BICRITICAL BEHAVIOUR IN CsMnBr$_3$·2D$_2$O

6.1. Introduction

CsMnBr$_3$·2D$_2$O (CMB) belongs to a series of compounds AMB$_3$·2aq with A = Cs, Rb; M = Fe, Co, Mn; B = Cl, Br and aq = H$_2$O, D$_2$O, which are interesting on account of their pseudo one-dimensional (d=1) characteristics and the variety of their magnetic phase diagrams. The strongly anisotropic, metamagnetic compound CsCoCl$_3$·2D$_2$O (CCC) has been the subject of detailed studies in order to gather evidence about the scaling properties and the universality of tricritical and critical behaviour [1]. The nature of the interactions in RbFeCl$_3$·2H$_2$O and CsFeCl$_3$·2H$_2$O seems to be even more pronounced Ising-like than in CCC. In these Fe-compounds resonances of large spin clusters can be observed [2] and their magnetic phase diagrams contain a ferrimagnetic phase [3]. Like CCC, these Fe-compounds are isomorphous [4] with the well known CsMnCl$_3$·2H$_2$O (CMC).

CMC has been the subject of a large number of experimental investigations to observe the properties of the pseudo d=1 Heisenberg system [5]. Neutron scattering experiments on CMC gave for the first time conclusive evidence for the existence of an antiferromagnetic chain-structure [6]. As a consequence of the nearly perfect Heisenberg character, CMC displays a spin-flop transition at a rather low parallel magnetic field $H_{SP}(T=0) \approx 17$ kOe [7]. The bicritical point has been localized at $T_b = 4.36$ K, $H_b = 20.55$ kOe [8]. From X-ray and NMR measurements it was concluded that the crystallographic structures of CMB and CMC are isomorphous [9] and are described by the orthorhombic spacegroup Pcca [10]. Although both compounds have very similar magnetic phase diagrams [11], the symmetry in the ordered state appears to be different. The magnetic spacegroup of CMB is P$c'a'$, which implies that the magnetic and crystallographic unit cell are the same [9]. In CMC the magnetic unit cell has a doubled b-axis, the spacegroup is $P_{2}_1c'a'$ [12]. In both systems the easy axis coincides with
the crystallographic b-axis. Here we shall give a report of the results of a neutron scattering study on the magnetic phase transitions in deuterated CMB in a parallel magnetic field. Specifically we shall focus on the behaviour close to the bicritical point.

For this experiment CMB, rather than the better known CMC was chosen, on account of experimental restrictions. Because of the value of the bicritical field $H_b$, the experiment had to be performed within a cryomagnetic system, in which a vertical magnetic field is combined with a horizontal scattering plane geometry. In contradistinction to CMB, CMC does not have magnetic reflections in the ac-plane, which is horizontal when $H$ is applied along the easy axis.

Section 2 contains details about the crystallographic and magnetic structure of deuterated CMB, verified in a neutron powder diffraction experiment. Section 3 contains experimental details. In section 4 the neutron scattering measurements on a single crystal of CMB in a (nearly) parallel magnetic field are presented. A detailed analysis of the phase diagram is given in sections 5 and 6. In the next two sections we compare the experimental data with the predictions of the extended-scaling theory on bicritical behaviour. Finally, the results are discussed in section 9.

6.2. Crystallography and magnetic interactions

In order to verify the conclusions of Swüste et al. [9] on the crystallographic and magnetic structure of CMB, a neutron powder diffraction experiment was performed. The sample consisted of approximately 30 g powdered CsMnBr$_3$·2D$_2$O from the same batch as the sample that we used in the single-crystal experiment.

Neutron diffraction patterns in the range $0.02 \lesssim \sin \theta / \lambda \lesssim 0.36 \, \text{Å}^{-1}$ were recorded at 300 K and 1.2 K. The observed and calculated profiles were compared in Rietveld's profile refinement program [13]. In this analysis the scattering lengths were taken as 0.541 (Cs), -0.387 (Mn), 0.679 (Br), 0.667 (O), -0.374 (H) and 0.580 (O) in units of $10^{-12}$ cm. The Watson and Freeman [14] formfactor of Mn$^{2+}$ was used and the structural parameters of CsCoCl$_3$·2D$_2$O served as starting values of
the atomic positions. The deuterium fraction $x$ of the sample could be obtained from a refinement of the effective scattering length at the deuterium positions.

In the analysis of the diffraction pattern at 300 K the expected Pcca structure was readily confirmed. The final parameter values at room temperature did not differ much from the values at 1.2 K, which are given in Table 6.1. The unit cell contains four formula units. Part

Table 6.1. Structural and magnetic parameters of deuterated CMB at 1.2 K. The crystallographic and magnetic spacegroups are Pcca and P'c'a', respectively. In standard deviations, based on statistics only, are given within parentheses in units of the last decimal.

<table>
<thead>
<tr>
<th>atom</th>
<th>$x$</th>
<th>$y$</th>
<th>$z$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs</td>
<td>0</td>
<td>0</td>
<td>0.1441(21)</td>
</tr>
<tr>
<td>Mn</td>
<td>0</td>
<td>0.4644(29)</td>
<td>0.1491(18)</td>
</tr>
<tr>
<td>Br_I</td>
<td>0.0931(8)</td>
<td>0.2122(8)</td>
<td>0.3922(9)</td>
</tr>
<tr>
<td>Br_{II}</td>
<td>0.0652(18)</td>
<td>0.6809(18)</td>
<td>0.3632(10)</td>
</tr>
<tr>
<td>O</td>
<td>0.0285(15)</td>
<td>0.6912(20)</td>
<td>0.4411(13)</td>
</tr>
<tr>
<td>D_I</td>
<td>0.1633(15)</td>
<td>0.6918(19)</td>
<td>0.3790(16)</td>
</tr>
<tr>
<td>D_{II}</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
</tbody>
</table>

$a = 9.4527(9)$ \( \AA \) \quad $\mu_a = 0$

$b = 7.4267(6)$ \( \AA \) \quad $\mu_b = 3.99(6)$ \( \mu_B \)

$c = 11.8642(13)$ \( \AA \) \quad $\mu_c = 0$

$\mu = 0.824(11)$

of the structure is shown in Fig. 6.1. Like in the isomorphs, the structure of CMB consists of cis-octahedra which are coupled along the $a$-axis by a shared bromine ion. The resulting chains are separated from each other by layers of Cs-ions in the $b$-direction and by hydrogen (deuterium) bonds along the $c$-direction.
Fig. 6.1. Schematic representation of the crystallographic chain-structure of \( \text{CsMnBr}_3 \cdot 2\text{D}_2\text{O} \) (CMB). Only one set of deuteriums and deuterium bonds is shown.

In the diagram at 1.2 K additional intensity, attributed to magnetic order, is observed to be concentrated mainly at the same scattering angles where the nuclear Bragg reflections occur. This is what one would expect if the magnetic unit cell is not doubled with respect to the crystallographic cell. The magnetic contributions to the reflections with \( h = i = \text{odd} \) are the most obvious. No special extinction conditions exist in the various magnetic subgroups of the Pcca-structure. However, due to the particular positions of the \( \text{Mn}^{2+} \)-ions in the unit cell at \( \vec{R}_j = (0, \frac{1}{2}-\delta, \frac{1}{2}), (0, \frac{1}{2}+\delta, \frac{1}{2}), (\frac{1}{2}, \frac{1}{2}+\delta, \frac{1}{2}) \) and \( (\frac{1}{2}, \frac{1}{2}-\delta, \frac{1}{2}) \) with a small \( \delta \approx 0.036 \), there are approximative extinction conditions for the magnetic reflections. This readily can be derived from the magnetic structure factor in the magnetic Bragg-scattering cross section

\[
\frac{d\sigma}{d\Omega}(\vec{Q}) = |f(Q)|^2 \sum \alpha (1-\delta^2) \left[ \sum_{j} \nu_j^\alpha \exp(i\vec{Q} \cdot \vec{R}_j) \right]^2.
\]
(6.1) in a slightly different notation for eq. (2.19). \( \mu_j \) is the magnetic moment at site \( R_j \) and \( a \) refers to the principal axes of the \( \chi \)-tensor, which in an orthorhombic system coincide with the crystallographic axes. From the nearly-extinction conditions it can be concluded that the presence of strong \( h = \ell \) odd reflections indicates an antiferromagnetic coupling of the magnetic moments, both along the \( a \)-axis and along the \( c \)-axis, which corresponds to the predicted magnetic spacegroup \( Pc'c'a' \) indeed \[9\]. The resulting array of magnetic moments, which in the AF-phase are directed along the \( b \)-axis, is shown in Fig. 6.2.

![Fig. 6.2. The array of ordered magnetic moments in CMB in the AF-phase (spacegroup \( Pc'c'a' \)].

In a magnetic field along the \( b \)-axis CMB undergoes a spin-flop transition \( (H_{SF} \approx 21 \text{kOe at } T = 0) \), where the magnetic moments jump approximately to the intermediate \( c \)-axis \[15\]. The weakly anisotropic character of CMB can also be derived from susceptibility and magnetization measurements, which yield for \( T = 0 \) an anisotropy field \( H_A \approx H_b - H_c = 770 \text{ Oe} \) and an exchange field \( H_{ex} \approx 273 \text{kOe} \), i.e. \( H_A/H_{ex} \approx 2.8 \times 10^{-3} \). Dipole-field calculations give \( H_b - H_c = 756 \text{ Oe} \) and \( H_b - H_a = 2166 \text{ Oe} \), which indicates that the anisotropy is mainly due to dipole-dipole interaction \[15\]. The low-dimensional behaviour of CMB can, for instance, be derived from specific-heat measurements \[16\]. The temperature dependence of the magnetic heat capacity is very well
approximated by the theoretical estimate for an $S = \frac{5}{2}$ Heisenberg linear chain system with an intrachain exchange interaction $J/k_B = -2.6\ K$ (Fig. 6.3.). An estimate of the interchain interaction $J'$ can be obtained, when this $J$-value and the observed Néel temperature of hydrated CMB, $T_N = 5.75\ K$, are inserted in a relation proposed by Oguchi [17]. This yields a ratio $|J'/J| = 1.4 \times 10^{-2}$, which defines CMB as a fairly good pseudo one-dimensional system.

6.3. Sample and apparatus

Large single crystals of CsMnBr$_3$·2D$_2$O were grown by evaporation of a saturated solution of MnBr$_2$ and CsBr in a molar ratio of approximately 6:1 in D$_2$O at room temperature. The sample was shaped to an approximately disk-like ellipsoid with dimensions $15 \times 15 \times 4\ mm^3$, in order
to minimize inhomogeneities in the internal field due to demagnetization effects. Since the sample had to be mounted in the inner vacuum chamber (IVC) of the cryostat, it was placed in a sealed gas-filled copper capsule to prevent dehydration.

The experiments were performed on a conventional double-axis diffractometer at the Petten HFR reactor. The neutron wavelength of 1.473 Å was obtained from a Zn (002) monochromator. Soller slits with a horizontal divergence of 30° were placed before the monochromator and in front of the 3He-detector. Vertical collimation was defined by 60° soller slits between monochromator and sample and in front of the detector. We used an Oxford Instruments cryostat with a superconducting magnet, which can produce a vertical magnetic field up to 50 kOe. The homogeneity of the field is better than 0.1% over a 15 mm diameter spherical volume. In the field scans of the present experiment the magnetic field was increased in steps of about 60 Oe. Like the diffractometer settings also the adjustment of the field was controlled by a P 9205 on-line computer.

The sample was pre-oriented in the sample holder in order to achieve an accurate alignment of the easy b-axis along the vertical field direction after mounting. To this purpose the intensities of several nuclear reflections in the reciprocal a*c*-plane were optimized in presence of a 30° vertical collimation. After mounting of the sample holder on the extension of the 1 K chamber of the cryostat, no final adjustments could be made. From the required canting angle of the cryomagnetic system (+ sample) out of the vertical position in order to realize a horizontal orientation of the a*c*-plane, the mismatch between H and the easy axis was estimated as 0.3 ± 0.3° both in the bc- and in the ab-plane.

Temperature control was achieved by balancing the cooling effect of the 4.2 K bath and a constant current supply to a heating resistor. To obtain temperatures below 4.2 K the pressure in the 1 K chamber was reduced. The heat contact between the sample holder and the 4.2 K bath was improved by admitting a few cm³ He contact gas in the IVC, which separates both systems. In this way a short-term temperature stability better than 1 mK was obtained near the bicritical temperature
$T_b \approx 5.25$ K, with a maximum drift of 5 mK over a day. To obtain temperatures above 10 K the IVC was evacuated and a commercial temperature controller could be used.

A calibrated Ge-resistor was used for the temperature registration in zero field. This thermometer was placed on the same block to which the sample holder was attached. In order to realize a reliable temperature registration also in presence of a magnetic field, the magneto-resistance of the Ge-resistor was measured in steps of 1 kOe at several (better than 1 mK) constant temperature values. By interpolation with a 5th degree polynomial a complete ($H, T$) calibration network was obtained.

6.4. Short-range and long-range order

In the $a^*c^*$-reciprocal lattice plane, to which our experiments were confined, the magnetic and nuclear Bragg reflections do not coincide. Using the transformation properties of $P_c'c'a'$ it can be derived from (6.1) that the only non-zero magnetic Bragg reflections in the $a^*c^*$-plane occur for $h = \ell = 2n+1$ with intensities

$$I_{h0\ell} = |f(Q)|^2 \sum_a (1 - \frac{Q_a^2}{\delta_a^2}) (N_{st}^a)^2, a = a, b, c.$$  

(6.2)

According to the crystallographic spacegroup $P_{cca}$, nuclear reflections occur at the reciprocal lattice points with $\ell = 2n$ (see Fig. 6.4.).

![Fig. 6.4.](image)

The $a^*c^*$-plane of the reciprocal lattice. Magnetic and nuclear reflections are indicated by filled and open circles, respectively. $\phi$ is the angle between $\hat{a}$ and the intermediate ($c$) axis.

In an applied magnetic field the induced homogeneous magnetization $M$ will also contribute slightly to these nuclear reflections.
Short-range order

The magnetic scattering will be concentrated in magnetic Bragg reflections, only if complete d=3 magnetic long-range order is established in the system. For $T \gg T_N$ (2.63 K) the magnetic correlations along the chain direction will be dominant, which leads to magnetic critical scattering concentrated in planes perpendicular to the $a$-axis. This effect was observed indeed and is illustrated in Fig. 6.5.

For a few temperature values the intensity profile is shown, observed along a path perpendicular to the $h=3$ plane at $t = 0.85$ (Fig. 6.5a) and along a path within the $h=3$ plane (Fig. 6.5b). At the reciprocal lattice point $(301)$ a second order nuclear peak is present. At $12.9$ K
a uniform increase of the intensity with respect to the background at
28 K is observed along the entire (30t) line, whereas the intensity
in the perpendicular direction is obviously concentrated around h=3.
This indicates that the fluctuations are strongly correlated along the
a-axis, whereas the chains are very weakly coupled along the c-axis.
It has not been verified explicitly that also the coupling along the
b-direction is negligible.

The data for T = 8.6 K (Fig. 6.5b) show an apparent concentration of
the intensity in the h=3 plane into a scattering peak around the
lattice point l = 1. So, the interchain coupling J' exerts its in-
fluence already at T/T_N \approx 2. This same value of 2 T_N has been reported
by Skalyo et al. for CMC [6], where |J'/J| = 8 \times 10^{-3} \quad [16], which is
not very different from the CMB-value. The influence of J' increases
as the number of correlated spins in the chains increases and T_N is
approached. In Fig. 6.5, it is clear that both the interlayer and the
intralayer correlation lengths grow below 13 K. In the present ex-
periment rather complete sets of critical scattering data near the
(103) and (301) reflections have been recorded as a function of both
temperature and magnetic field.

**Long-range order**

In the remainder of this chapter we shall focus on the magnetic Bragg
scattering, which occurs if the system is ordered in all three di-
mensions. Both in the AF-phase and in the SF-phase, the variations in
peak intensity of three magnetic Bragg reflections, viz. (103), (101)
and (301), have been recorded mainly in field scans at constant
temperature. From these intensities the (H,T) variation of the order
parameters in both phases can be obtained. As can be seen from eq.
(6.2), the intensity of a magnetic Bragg-reflection (h0l) in the
a* c*-plane may be written as

\[ I_{h0l}(H,T) = C_{h0l} \left[ \frac{\hbar}{2} (H,T) \right]^2 + \sin^2 \theta_{h0l} \left| \mathbf{M}_{sf}^\perp (H,T) \right|^2. \]

(6.3)

Here \( \mathbf{M}_{sf}^\perp \) is the component of \( \mathbf{M}_{sf} \) along the easy b-axis, i.e. the
order parameter of the AF-phase, and \( \mathbf{M}_{sf}^\perp \) is the component of \( \mathbf{M}_{sf} \)
along the intermediate c-axis, i.e. the order parameter of the SF-phase. We assumed $M_{st}^\parallel (H,T)$ along the hard a-axis to be zero. In (6.3) $\Theta_{hi}$ is the angle between the scattering vector $\vec{Q}$ and the c-axis (see Fig. 6.4.). The scale factor $C_{hi}$ depends on many quantities, such as the magnetic formfactor, the instrumental resolution and the illumination factor of the sample in a specific scattering geometry. In addition to the magnetic intensity (6.3) the observed peak intensities will contain also a non-magnetic contribution $B_{hi}$, which consists of background and second-order nuclear scattering.

It is not very problematic to obtain $M_{st}^\parallel$ or $M_{st}^\perp$ from any reflection with a sufficiently high magnetic intensity, if the whole sample is either in the AF-phase (with $M_{st}^\perp = 0$) or in the SF-phase (with $M_{st}^\parallel = 0$). However, if the sample is divided in domains, so that AF- and SF-order coexist, or if $\vec{H}_{st}$ has a direction somewhere between the b-axis and the c-axis, the separation of $M_{st}^\parallel$ and $M_{st}^\perp$ is not completely straightforward. This situation is encountered for instance near the spin-flop transition. Then we have to determine $M_{st}^\parallel$ and $M_{st}^\perp$ separately by comparing the intensities of two or more magnetic reflections. Eq. (6.3) indicates that this determination is possible, if for each reflection $C_{hi}$, $\Theta_{hi}$ and the non-magnetic contribution $B_{hi}$ are known. With the $\Theta_{hi}$ calculated from the crystal structure data, the constants $C_{hi}$ and $B_{hi}$, which in the ideal case are independent of H and T, can be obtained from the intensities observed within the AF- and SF-phases. The employed procedures to determine these constants and to obtain statistically justified estimates for $M_{st}^\parallel (H,T)$ and $M_{st}^\perp (H,T)$ is described in Appendix 6.10.

Some typical peak-intensity variations, observed in field scans close to the spin-flop transition at various T-values, are shown in Fig. 6.6a. The data have been corrected for the non-magnetic contributions $B_{hi}$ and the scale factors $C_{hi}$ have been eliminated to achieve that all three curves coincide in the AF-phase. Details of the data collapsing are shown in Fig. 6.7. and discussed in Appendix 6.10. The fact that the same set of constants $(B_{hi}, C_{hi})$ could be used for all temperatures, indicates that extinction can be neglected. The scaled intensities in the SF-phase are found to be proportional to the corresponding
The field dependence of the scaled peak intensities of three magnetic Bragg reflections for different T. Triangles (100), squares (101) and circles (101). 

Fig. 6.6.

(a) The field dependence of the scaled peak intensities of three magnetic Bragg reflections for different T. Triangles (100), squares (101) and circles (101). 

(b) The resulting field dependences of \((M_{st}^{\|})^2\) and \((M_{st}^\perp)^2\), obtained from the three curves on the left, are indicated by open and closed circles, respectively.

\[\sin^2 \theta_{hk}, \text{ which shows that the c-axis is the intermediate axis indeed.}\]

The results for the corresponding field variations of \((M_{st}^{\|})^2\) and \((M_{st}^\perp)^2\) separately are shown in Fig. 6.6b. In many calculational programs in the further analyses, such separated \(M_{st}^{\|}\) and \(M_{st}^\perp\) results have been used as input data.

6.5. The spin-flop transition \((T < T_h)\)

Although the observed change in the orientation of \(\mathbf{M}_{st}\) at \(T = 3.736\, K\) seems to be a rather rapid one in Fig. 6.6b., it is not a first-order transition. This is better visible on the enlarged field scale in Fig. 6.8., where the rounding in \((M_{st}^{\|}(H))\) and \((M_{st}^\perp(H))\) is obvious,
Fig. 6.7. The achieved data collapsing in the AF-phase for the boxed $(H,T)$ regions of Fig. 6.6. For (a) a region was selected where the magnetic intensity is much higher than the background. In (b) a region with very low magnetic intensities is shown.

and can be concluded also from a simple calculation of the maximum field range in which the first-order spin-flop transition in CMB might occur. Taking the $\chi^{1\perp}$ value reported for hydrated CMB [15] and applying the maximum demagnetization factor $N (=4\pi)$ one arrives in MF-approximation at

$$\Delta H_{\text{max}} = 4\pi \chi^{1\perp} H_{\text{SF}} \approx 34 \text{ Oe.} \quad (6.4)$$
however, the reorientation of $\hat{M}_{st}$ clearly extends over more than 500 Oe. The 0.1% field inhomogeneity can account for only $\lesssim 25$ Oe. Indeed, as we have seen in section 3.5., the first-order character of the spin-flop transition can only be observed if the mismatch angle $\psi$ between $\hat{H}$ and the easy axis does not exceed the critical angle $|\beta|$ (eq. (3.65)).

$$\psi_c(T=0) \approx 28.6^\circ \frac{H_A}{H_{ex}} \approx 0.08^\circ \quad (6.5)$$

Moreover, at $T = 3.7$ K $\gtrsim 0.7 T_b$ the critical angle will be reduced already to about $0.02^\circ$ (eq. (3.66)), whereas the mismatch in the present experiment has been estimated as $\psi \gtrsim 0.3^\circ \pm 0.3^\circ$.

Since $\psi > \psi_c(T)$ for all our data, the MF-expression $|19|

$$\tan (2\zeta + 2\psi) = \frac{\sin 2\psi}{\cos 2\psi - (H/H_{SF})^2} \quad (6.6)$$

which was introduced in chapter III as eq. (3.67), should give a good description of the rotation of $\hat{M}_{st}$ with increasing $H$. For a few values of $\psi$ the field dependence of

$$\cos^2 \zeta = \frac{(M'')^2}{M_{st}^2} \quad (6.7)$$

is shown in Fig. 6.9. In order to verify the MF-prediction for our data, first the field dependence of $M_{st}^2(H)$, which is not predicted by (6.6), has to be constructed from the observed data

$$[M_{st}(H)]^2 = [M''_{st}(H)]^2_{obs} + [M'_{st}(H)]^2_{obs} \quad (6.8)$$

Subsequently, the theoretical predictions for the separate components $M''_{st}(H)$ and $M'_{st}(H)$ can be calculated as

$$M''_{st}(H) = M_{st}(H) \cos \zeta \quad (6.9)$$

$$M'_{st}(H) = M_{st}(H) \sin \zeta \quad (6.10)$$

with $\zeta$ determined by (6.6). Some typical results are shown in Fig. 6.10.
Fig. 6.9.

MF-prediction for the rotation of $\tilde{H}_{et}$ with increasing $H$ for several values of the misalignment angle $\psi$. The angle $\iota$ is determined by eq. (6.6).

$(M_{et}^H)^2$ and $(M_{et}^\perp)^2$ are proportional to $\cos^2 \iota$ and $1-\cos^2 \iota$, respectively. from which one may conclude that over the whole investigated temperature range the observed rotation of $\tilde{H}_{et}$ is excellently described by the MF-prediction with $\psi = 0.5^\circ$.

Fig. 6.10. Comparison of the observed and calculated rotation of $\tilde{H}_{et}$ at several temperature values. The solid lines represent the MF-prediction of $(M_{et}^H)^2$ and $(M_{et}^\perp)^2$ for $\psi = 0.5^\circ$. The variation in $H_{et}^2$ is shown by the dash-dotted line.
The intersection of the curves for \((\hat{H}_{st}^\parallel)^2\) and \((\hat{M}_{st}^\perp)^2\) occurs at

\[
H_{i.s.} = H_{SF} / \cos 2\psi .
\]  

(6.11)

Since the difference between \(H_{i.s.}\) and \(H_{SF}\) is negligible for \(\psi = 0.5^\circ\), these points of intersection provide an accurate determination of the spin-flop field \(H_{SF}(T)\). The spin-flop line, obtained in this way, is shown in Fig. 6.16., together with the results for the other phase boundaries.

The theoretical curves in Fig. 6.9. also provide an easy way to correct the data for the rounding effects due to the misalignment. Since the curve for \((\hat{H}_{st}^\parallel)^2\) approaches the stepfunction closer as \(\psi\) decreases, one can take

\[
(\hat{M}_{st}^\parallel)^2 = M_{st}^2 , \quad (\hat{M}_{st}^\perp)^2 = 0 \quad \text{for } H < H_{SF}
\]

and

\[
(\hat{M}_{st}^\parallel)^2 = 0 , \quad (\hat{M}_{st}^\perp)^2 = M_{st}^2 \quad \text{for } H > H_{SF}
\]  

(6.12)

as the result for \(\psi = 0\). If the data would be gathered with an exact alignment of \(\hat{H}\) along the easy axis, such a discontinuous behaviour of \(\hat{H}_{st}^\parallel\) and \(\hat{M}_{st}^\perp\) would have been observed. Fig. 6.11. shows the temperature dependence of \(M_{st}^2(\hat{H}=H_{SF})\), which can be described fairly well by a single-power law with \(T_c = 5.275(9)\) K and an exponent \(\beta = 0.58(2)\).

We shall return to this result in section 6.7.
For a few temperature values also the field variation of the nuclear (200) reflection has been recorded, mainly close to the "spin-flop" transition in order to observe the intensity increase caused by the induced homogeneous magnetization \( M \). A calculation, similar to the one in (6.2) shows that this magnetic contribution is quadratic in \( M \). Although the (200) nuclear reflection is very weak, the magnetic contribution is still a very small fraction of the total intensity (see Fig. 6.12.). The expected jump in the magnetization at the spin-flop transition is well resolved at \( T = 3.75 \) K, but for each point in Fig. 6.12. a counting time of 100 minutes was required to get sufficient statistical accuracy. Therefore, it would have been too time-consuming to undertake extensive measurements of the temperature variation of this jump. Finally we show in Fig. 6.13. the square root of the magnetic contribution to the (200) intensity at \( T = 3.75 \) K, as a function of \( H \). According to MF-theory \( M \) should be proportional to \( H \) in the SF-phase, which appears to be the case indeed.

Fig. 6.12.
Jump in the intensity of the nuclear (200) reflection at the spin-flop transition for \( T = 3.75 \) K, due to the contribution of the homogeneous magnetization \( M \).

Fig. 6.13.
Field dependence of the homogeneous magnetization at \( T = 3.75 \) K.
6.6. The paramagnetic phase boundaries (T > T_b)

For field scans at a temperature sufficiently far above the bicritical temperature T_b, one does not need the involved separation procedure of M^∥ and M^⊥. As can be seen in Fig. 6.6b., even at T = 5.299 K, which appears to be fairly close to T_b ≈ 5.25 K, the AF- and SF-ordering occurs in well-separated (H,T) regions, with a disordered paramagnetic phase in between. The intensities of the magnetic reflections then are simply proportional to either (M^∥)^2 or (M^⊥)^2 and can be used directly to determine the phase boundaries of the ordered phases. To determine the location of these lines of critical points T^∥_c(H) and T^⊥_c(H), we performed least-squares fits of the power laws

\[ I^∥/I^⊥ = (M^∥/M^⊥)^2 = B^2 \cdot t^{2B,⊥} \]  

(6.13)

to the data. Here t = |T/T^∥_c(H)-1| and t = |H/H^∥_c(H)-1| in temperature and field scans, respectively. In the fits B, α and H_c or T_c were variable. As equal counting times were used for the three reflections, the intensity of the (103) reflection appeared too weak in the SF-phase, to yield reliable results. No corrections have been applied for possible critical scattering contributions. Instead, in the analysis of the data with t min ≤ t ≤ t max, the lower cutoff t min has been varied between 10^-3 and 5*10^-3, which appeared to have little effect on the resulting parameters. This indicates that corrections due to critical scattering may be neglected in the present rough fits. Since the inhomogeneity of the magnetic field is ≈ 0.1%, data with t < 10^-3 have been omitted.

Fig. 6.14. shows some typical log(I) - log(t) results for peak intensities in both the AF-phase and the SF-phase, recorded in field scans at constant temperature. It appears that the intensities approximately follow a single-power law over a wide field range. Because of the large number of field scans which were taken, also the number of resulting β-values is rather large. Fig. 6.15. gives the results for β from fits of (6.13) to the data with 10^-3 < t < 10^-1 and presented as a function of the resulting T^⊥_c = T^⊥_c(H)/T_b-1. A statistical analysis of the data, which show no systematic variation along the phase
boundaries, yields the average values $\bar{\beta}_{\parallel} = 0.321(6)$ and $\bar{\beta}_{\perp} = 0.326(7)$. Both values are in good agreement with the theoretical predictions for the d=3 Ising model, viz., $\beta = 0.325(1)$ from $t$-expansions [20] and $\beta = 0.312(5)$ from series expansions [21], and show no systematic differences. From this it may be concluded that in spite of the Heisenberg-like character of CMB at high temperature, the critical behaviour close to $T^{\parallel}_{c}$ and $T^{\perp}_{c}$ is Ising-like. Apparently the weak anisotropy is sufficient to break the $n=3$ symmetry.

An analysis of the influence of a variation in the upper cutoff $t_{\text{max}}$ indicates a slight but systematic decrease in $\beta$ with increasing $t_{\text{max}}$ (see Table 6.2.). A possible explanation of this effect is that the data with high $t$-values are not within the critical region, and a crossover region to bicritical behaviour is entered at high $t$. Since
the exact location of these crossover regions is unknown in this stage of the analysis, the determination of \( \beta \)-values from single field scans or temperature scans must be presented with some reservation. In previous reports of the present investigation [22,23] we have quoted the \( \beta \)-values for \( t_{\text{max}} = 0.1 \) since for \( t < 0.1 \) the changes remain within the statistical uncertainties, whereas this is not true for \( t > 0.1 \).

As we shall see in section 6.8, the extended-scaling analysis provides a more reliable determination of \( \beta_H \) and \( \beta_\perp \).

For the present discussion it is much more important that the resulting values of \( H_c(T) \) appear to change hardly with varying \( t_{\text{max}} \). The AF-P and SF-P phase boundaries obtained are shown in Fig. 6.16. The striking increase of \( T_{\text{c}}^H(H) \) for \( H > H_b \) is a characteristic feature of pseudo d=1 Heisenberg systems [11,24] and has nothing to do with the predicted bicritical "umbilicus" [25], which was discussed in sections 3.4. and 3.5.
In order to determine the shape of the phase boundaries close to the triple point \((T_m, H_m)\) and in particular to obtain an accurate estimate of the location of this point, a closer analysis of the data in the region around \((T_m, H_m)\) has been performed. For this purpose the separated \(\langle M^\parallel \rangle^2\) and \(\langle M^\perp \rangle^2\) data were used, obtained from field scans at constant temperature both for \(T < T_m\) and for \(T > T_m\). These \(\langle M^\parallel \rangle^2\) and \(\langle M^\perp \rangle^2\) data have been compared with the power-law behaviour (6.13), with \(\beta\) fixed at 0.32. The resulting values for \(H_c^\parallel(T)\) and \(H_c^\perp(T)\) are shown in Fig. 6.17a. In the fits only data with \(0.015 < |H/H_c(T)-1| < 0.1\) were taken into account. Apparently the phase boundaries intersect each other at \(T_m \approx 5.275(10)\) K. Of course the results for \(T < T_m\) have no physical meaning, but the results for \(T > T_m\) have! To check this statement, the same data were also fitted
to (6.13) with a variable \( \beta \). The resulting \( \beta \)-values are shown in Fig. 6.17b. It is obvious that the results deviate from \( \beta = 0.32 \) for

![Graph](image)

Fig. 6.17.

(a) Critical field values \( H_{c2}(T) \) (triangles) and \( H(0) \) (dots), determined from fits of (6.13) with \( \beta \) fixed at 0.32. Separated data of \([\gamma_{eq}(H)]^2 \) and \([H_{c2}(H)]^2 \) for \( T \) both above and below \( T_m \) were used. Spin-flop field data are indicated by open circles.

(b) Resulting \( \beta_{\parallel} \) (dots) and \( \beta_{\perp} \) (triangles) values, from similar fits of (6.13) to the same data as in (a), now with \( \beta \) variable. From these results the triple point was located at \( T_m = 5.275(10) \) K, \( H_m = 26.55(5) \) kOe.

\( T > T_m \) but are well described by this value for \( T > T_m \). In this stage it must also be noted that \( M_{st}^2 (H-H_{SF}) \) disappears at the same temperature \( T_m \) (see Fig. 6.11.). From this analysis the common point of the three phase boundaries can be located rather accurately at \( T_m = 5.275(10) \) K, \( H_m = 26.55(5) \) kOe. From Fig. 6.17a, or similarly from the insert in Fig. 6.16., it is obvious that the phase boundaries \( T_{c2}(H) \) and \( T_{c2}(H) \) do not come in tangent to the "spin-flop line" at \( T = T_m \). This is indeed what should be expected if the magnetic field is not aligned perfectly along the easy axis.
6.7. Analysis of the magnetic phase diagram

Although the overall features of the magnetic phase diagram of CMB deviate markedly from those for $d=3$ systems, the critical behaviour near the phase boundaries must be considered as essentially three-dimensional. As we mentioned in the previous section, the observed $\beta$-values are in good agreement with the $d=3$ Ising value, which is theoretically predicted for $\beta_{\parallel}$ and $\beta_{\perp}$ in an $n=2$ spin-flop system [26]. Consequently, we expect that also the behaviour close to the bicritical point in CMB will show the regular characteristics of a $d=3$ $n=2$ spin-flop system. This implies for instance, that the theoretical predictions for the shape of the paramagnetic phase boundaries $T_c(\parallel H)$ and $T_c(\perp H)$, must also hold in CMB. As these predictions have been discussed extensively in chapter III, section 3.4, we shall confine ourselves here to a recapitulation of the expressions which are relevant in the present analysis.

According to the extended-scaling theory [25, 26], the shape of the AF-P and SF-P phase boundaries is described by

$$\frac{\gamma}{\eta} = -\eta_{\parallel} \quad \text{and} \quad \frac{\gamma}{\eta_{\perp}} = +\eta_{\perp}. \quad (6.14)$$

The optimum scaling axes are given by [27]

$$\bar{g} = \bar{g} - pt \quad \text{and} \quad \bar{\tau} = \bar{\tau} + qg, \quad (6.15)$$

where

$$\bar{g} = H^2 - H_b^2 \quad \text{and} \quad \bar{\tau} = T/T_b - 1. \quad (6.16)$$

The $\bar{g}=0$ axis must be tangent to the spin-flop line at $T_b$, that is

$$p = T_b \left( \frac{dH_{SF}^2}{dT} \right)_b. \quad (6.17)$$

The value of $q$ is not universal. Starting from mean-field theory Fisher [27] obtained the estimate

$$q(n) = -\frac{n+2}{3nT_b} \left( \frac{d\eta_{\parallel}}{dT} \right)_{H=0}. \quad (6.18)$$
which is expected to be valid for small values of \((4-d) = \epsilon\). So it is doubtful whether (6.18) also applies to a pseudo \(d=1\) system like CMB. Eq. (6.15) - (6.18) are identical with the expressions (3.36) - (3.39) in chapter III. Although the amplitudes \(w_\parallel\) and \(w_\perp\) in (6.14) are not universal, their ratio is. Fisher [27] showed that

\[
Q(n) = \frac{w_\perp}{w_\parallel} = n^{-1} + \epsilon(n) .
\]  

(6.19)

Numerical estimates yield \(|26| Q(3) \approx 2.51\) for \(d=3\). Again it is very questionable whether this estimate would apply to pseudo \(d=1\) systems. For \(n=2\) the ratio is exactly determined by symmetry as \(Q(2) = 1\).

In the previous section we noted that the phase boundaries \(T_{c\parallel}\) and \(T_{c\perp}\) do not approach to the spin-flop line tangentially, in contradistinction to the theoretical prediction (6.14). This effect must be expected when the magnetic field is not perfectly aligned along the easy axis, as has been emphasized by Rohrer [28]. This is illustrated in Fig. 6.18., where the theoretically predicted shape of the critical surface for an orthorhombic spin-flop system in the three-dimensional \((T, H_\parallel, H_i\parallel)\) space is shown. Here \(H_\parallel\) and \(H_i\parallel\) represent the components of the applied magnetic field along the easy and intermediate axis, respectively. The component along the hard axis \((H_i\perp)\) does not play a role in the following discussion. Close to the \((n=2)\) bicritical point \((T_b, H_\parallel, 0)\) the \(n=1\) critical surface has an "umbilical" shape [25]. Any section of this surface which contains the bicritical point, has a shape similar to the one expressed by (6.14) for \(T_{c\parallel}(H_\parallel)\) and \(T_{c\perp}(H_\parallel)\) in the \(H_i\parallel=0\) plane.

If the applied field is slightly misaligned, and therefore a small nonzero \(H_i\) is present, the observed phase diagram will not be represented by the section of the critical surface at \(H_i=0\), but by a section at a small value of \(H_i\). Both sections are indicated in Fig. 6.18. Apparently the section at finite \(H_i\) is rounded at a temperature \(T = T_m\). This is indeed the shape of the phase diagram observed in CMB. However, one always can define a pseudo-bicritical point \((T_b', H_b, H_i')\) in such a way that the shape of the section at finite \(H_i\) is well described by (6.14) with respect to this pseudo-bicritical point at temperatures...
Critical surface of an n=2 spin-flop system in $(T, H^\parallel, H^\perp)$-space. $H^\parallel$ and $H^\perp$ are the components of $\vec{H}$ along the easy and intermediate axes, respectively. (Compare Figs. 3.4. and 3.8.). Sections of this surface at $H^\perp = 0$ and $H^\perp$ nonzero are indicated. For high $T$ the shape of the section at nonzero $H^\perp$ is described by (6.14) with respect to the pseudo-bicritical point $(T'_b, H^\parallel_b, H^\perp)$. The section is rounded at $T = T_m$.

sufficiently far above $T_m$, $T'_b$ will be somewhat higher than $T_b$. However, when $H^\parallel$ is only small, the differences $T'_b - T_b$ will not be large and the deviation of the observed phase boundaries from the ideal shape (6.14) will be localized in a small region close to $T_m$.

In this spirit we may compare our data for $T^\parallel_c(H)$ and $T^\perp_c(H)$, obtained in a slightly misaligned field, with the theoretical expressions (6.14). It must be emphasized here, that the above arguments only apply to the shape of the phase boundaries. From the analysis in this section we
shall obtain estimates for the crossover exponent $\phi$ and the location of the pseudo-bicritical point. We shall not find bicritical exponents on approaching the critical point $T_m$, because it is a point of the Ising surface and therefore will show $d=3$ Ising characteristics.

In order to obtain $T'_b, H_b$ and $\phi$, we have compared the expressions (6.14) with our data for $T > 5.285$ K in least-squares fits. In the following we shall distinguish no longer explicitly between $T'_b$ and $T_b$. In all fits the value $(dK^2_{SF}/dT)_b = 75.4$ kOe$^2$/K, obtained from the slope of the spin-flop transition line, was used. The ratio $\omega_\perp/\omega_\parallel$ was fixed to unity, because of the very strong correlation with the choice of the $\tau=0$ axis, i.e. with the value of $q$.

![Graph showing fitted values of $\phi$ and $\omega_\parallel = \omega_\perp$ as a function of $\tilde{T}_{\text{max}}$ for several choices of $T_b$. From these results $T_b = 5.285(6)$ K has been estimated. For $\tilde{T} > 0.15$ the data deviate significantly from (6.14).](image)

Fig. 6.19. Fitted values of $\phi$ and $\omega_\parallel = \omega_\perp$ as a function of $\tilde{T}_{\text{max}}$ for several choices of $T_b$. From these results $T_b = 5.285(6)$ K has been estimated. For $\tilde{T} > 0.15$ the data deviate significantly from (6.14).

As the range of $\tilde{T}$ in which (6.14) holds is not universal, we have tried to determine this range in an objective way. The approach is based on the requirement that all fits in which data with $\tilde{T} \leq \tilde{T}_{\text{max}}$ are taken into account, must yield the same estimate for any variable, independent of $\tilde{T}_{\text{max}}$ up to an upper value of $\tilde{T}_{\text{max}}$. This requirement can only be fulfilled if the correct value of $T_b$ is used. The procedure is illustrated in Fig. 6.19., where for several choices of $T_b$ the fitted values of $\phi$ and $\omega_\parallel = \omega_\perp$ are shown as a function of $\tilde{T}_{\text{max}}$. The estimates
for $q$ and $H_b$ are not shown, as these hardly vary with $T_b$ and $t_{\text{max}}$.

From the results in Fig. 6.19, we can determine

$$T_b = 5.255(5) \text{ K},$$

which estimate gives constant parameter values up to $t_{\text{max}} \lesssim 0.15$. For $t_{\text{max}} > 0.15$ all curves show a systematic increase with $t_{\text{max}}^*$, which indicates that systematic deviations of the data from (6.14) become significant for $t > 0.15$.

With $T_b$ fixed at 5.255 K, a least-squares fit to the data with $t \leq 0.15$ gives

$$H_b = 26.541(17) \text{ kOe}, \quad \phi = 1.226(9),$$

$$q = 5.38(10) \times 10^{-5} \text{ kOe}^{-2} \quad \text{and} \quad \varpi_{\parallel} = \varpi_{\perp} = 7033(52) \text{ kOe}^2.$$

If, in addition to $T_b$, $\phi$ is fixed at the theoretical prediction $\phi = 1.175$, the values

$$H_b = 26.555(21) \text{ kOe}, \quad q = 5.57(12) \times 10^{-5} \text{ kOe}^{-2}$$

and

$$\varpi_{\parallel} = \varpi_{\perp} = 6226(38) \text{ kOe}^2$$

result. Both fitted curves are compared with the data in Fig. 6.20. Also Fisher's estimate for the $\varpi=0$ axis (6.18) has been included.

Indeed, the observed phase boundaries appear to be well described by the theoretical expressions (6.14) with respect to a pseudo-bicritical point at 5.255 K. Only a slight rounding effect due to the misalignment of the field is observed for $5.255 < T < 5.285$ K. The curve fitted with fixed $\phi = 1.175$ deviates rapidly from the data for $T > 5.5$ K, whereas the best fitted curve ($\phi = 1.226$) starts to deviate at $T \gtrsim 5.8$ K.

With the uncertainty in $T_b$ taken into account, we would quote as final parameter estimates

$$T_b = 5.255(5) \text{ K}, \quad H_b = 26.55(2) \text{ kOe}$$

$$\phi = 1.22(6) \quad \text{and} \quad q = 5.5(2) \times 10^{-5} \text{ kOe}^{-2}.$$
Fig. 6.20. $H^2$ vs $T$-diagram of the observed phase boundaries compared with best fits of (6.14) in which $\phi$ was variable (solid line) and $\psi = 1.755$ (dash-dotted). In these fits to the data with $\varepsilon < 0.15$, $T_b$ was fixed at 5.255 K. The fitted $\varepsilon = 0$ axis (solid) and the theoretically predicted (dashed) $\varepsilon = 0$ axis (6.18) are also shown. The insert shows details near $T_b$.

where it should be emphasized that $T_b$ in fact refers to the pseudo-bicritical point.

From this analysis of the phase diagram it may be concluded that also in the pseudo $d=1$ system CMB the shape of the second-order phase boundaries near the bicritical point is predicted correctly by extended scaling theory. The observed $\phi$-value is in fair agreement with the theoretical prediction for an $n=2$ system. However, it should be emphasized that the fitted slope $q$ of the $\varepsilon=0$ axis strongly deviates from Fisher's estimate (6.18) (See Fig. 6.20.). Apparently this estimate is not appropriate for CMB, as might be expected, because the mean-field type of approach used in the determination is probably not applicable to pseudo $d=1$ systems.
6.8. Experimental test of the extended-scaling hypothesis

The heart of the theory on bicritical behaviour is the extended-scaling hypothesis [25,26]. As we discussed extensively in chapter III, this hypothesis can be formulated as a generalized homogeneous-function postulate for the free energy

\[ G(H^{\parallel}, H^{\perp}, \tilde{\xi}, \tilde{\zeta}) = |\tilde{\zeta}|^{2-\eta_b} \left( \frac{H^{\parallel}}{|\tilde{\zeta}|^{\beta_{\parallel}}} + \frac{H^{\perp}}{|\tilde{\zeta}|^{\beta_{\perp}}} + \frac{\tilde{\xi}}{|\tilde{\zeta}|^{\eta}} \right) \] (6.21)

Eq. (6.21) is a recapitulation of (3.35), where the various quantities have been defined. In the last few years experimental studies on a number of spin-flop systems have been undertaken to verify the extended-scaling theory for bicritical points [28-35]. In most cases the tests were restricted to a comparison of the experimentally determined shape of the paramagnetic phase boundaries with the theoretical predictions (6.14), similar to the analysis performed in the previous section. Hitherto a more direct experimental verification of the scaling functions was lacking. In this section, however, we shall present an extensive scaling analysis of our data on \( M^{\parallel}_{\text{st}}(H,T) \) and \( M^{\perp}_{\text{st}}(H,T) \) in CMB. The results of this analysis represent the first direct test of the central assumption in the theory on bicritical behaviour, namely of the extended-scaling hypothesis (6.21) itself.

A direct implication of (6.21) is that also the derivatives of \( G \) must be generalized homogeneous functions. For instance, the order parameter in the SF-phase should vary as

\[ M^{\perp}_{\text{st}}(\tilde{\xi}, \tilde{\zeta}) = B|\tilde{\zeta}|^{\beta_{b}} \cdot \frac{H^{\perp}}{|\tilde{\zeta}|^{\phi}} \] (6.22)

Similarly, a scaling function \( \mathcal{A} \) is obtained for the order parameter in the AF-phase (cf. section 3.4., eq. (3.44)). \( \mathcal{A} \) is nonzero only for \( \tilde{\xi} < 0 \), whereas \( \mathcal{A}^{\perp} \) is of interest for \( \tilde{\xi} > 0 \). It must be noticed that the scaling function \( \mathcal{A}^{\perp} \) (and similarly \( \mathcal{A}^{\parallel} \)) consists of two branches, \( \mathcal{A}^{\perp} \) for \( \tilde{\xi} > 0 \) and \( \mathcal{A}^{\perp} \) for \( \tilde{\xi} < 0 \). The exponent \( \beta_{b} \) corresponds to a purely bicritical behaviour of the order parameters (cf. section 3.4.) and is predicted to have the d=3 XY value [25]. Alternative scaling functions for \( H^{\parallel}_{\text{st}} \) and \( H^{\perp}_{\text{st}} \) have been defined in (3.43) as
and the analogue for $M_{st}$. Since scaling functions are expected to be universal \[36,37]\, the scaling functions for $M_{st}$ and $M_{st}^\perp$ should be identical in an $n=2$ spin-flop system. The scaling relations (6.22) and (6.23), and the analogues for $M_{st}$, should hold within the entire neighbourhood of the bicritical point. This can be verified directly in the present experiment on CMB. Since $x$ depends on

\[ x = \left| \frac{\tilde{z}}{\partial \tilde{z} / \partial \tilde{c}} \right|^{1/\phi} \quad (6.24) \]

and not on $\tilde{g}$ and $\tilde{c}$ separately, (6.22) predicts that our data $M_{st}(\tilde{g},\tilde{c})$ and $M_{st}^\perp(\tilde{g},\tilde{c})$, properly scaled with a factor $b|\tilde{c}|^{1/\phi}$, must coincide with the curve $\mathcal{G}_+(x)$ for $\tilde{c} > 0$ and with $\mathcal{G}_-(x)$ for $\tilde{c} < 0$. A similar data collapsing should occur according to (6.23), when the data scaled with a factor $b|\tilde{g}|^{1/\phi}$, are plotted as a function of

\[ y = \tilde{c}/\left| \partial \tilde{c} / \partial \tilde{g} \right|^{1/\phi}. \quad (6.25) \]

Both scaling functions $\mathcal{G}$ and $\mathcal{H}$ can be used equally well in order to test the data collapsing in CMB. However, the way in which information on different kinds of critical behaviour is contained in $\mathcal{G}$ and $\mathcal{H}$, is utterly dissimilar. In the scaling function $\mathcal{G}$, data gathered in a $(\tilde{g},\tilde{c})$-region close to the spin-flop line $\tilde{g}=0$ are fully displayed in a wide $y$-range ($y \rightarrow -\infty$), whereas the information from the $(\tilde{g},\tilde{c})$ region close to the $\tilde{c}=0$ axis (see Fig. 6.20) is compressed in a small $y$-interval around $y=0$. On the other hand, the scaling function $\mathcal{H}$ gives hardly information about the $(\tilde{g},\tilde{c})$ region close to the spin-flop line, which is compressed around $x=0$ in $\mathcal{G}$. Now, the data originating from the $(\tilde{g},\tilde{c})$ region close to the $\tilde{c}=0$ axis, are highly emphasized and cover the entire large-$x$ range of both branches of $\mathcal{G}(x)$. It is clear that the scaling procedures are complementary. Therefore, both analyses will be performed in order to get a complete image of the ordered phases in CMB.
The scaling functions $f(y)$

From $f(y)$ information can be extracted in particular on the behaviour of $M_{st}$ close to the spin-flop line. As we discussed in section 3.4., the $t$-variation of $M_{st}^\parallel$ and $M_{st}^\perp$ along the $g=0$ axis is given by

$$M_{st}^\parallel, M_{st}^\perp (g=0) \sim |\xi|^{\delta_b}.$$  \hspace{1cm} (6.26)

This implies that the scaling function $f(y)$ for $y \to -\infty$ must show the asymptotic behaviour given by

$$f(y) = \frac{M_{st}(g,\xi)}{|g|^{\delta_b/\phi}} \sim \frac{|\xi|^{\delta_b}}{|g|^{\delta_b/\phi}} = y^{\delta_b}, \text{ for } y \to -\infty.$$  \hspace{1cm} (6.27)

Close to the paramagnetic phase boundaries, given by $g = \pm \xi (i.e. \, y=1)$, $f(y)$ must display critical behaviour:

$$f(y) \sim (1-y)^{\delta}, \text{ for } y \to 1.$$  \hspace{1cm} (6.28)

In (6.27) and (6.28) the exponents $\delta_b$ and $\delta$ are predicted to have the $d=3$ XY and Ising values, respectively \cite{25}. Therefore, it should be possible to observe the crossover from bicritical to critical behaviour as a change in the slope of $f(y)$ drawn in a double-logarithmic plot as a function of $(1-y)$.

To verify the predicted data collapsing and to observe the crossover behaviour in $f(y)$ we have compared the scaled data of CMB in a plot of $\ln(M_{st}(g,\xi)/|g|^{\delta_b/\phi})$ vs. $\ln(1-y)$. In this scaling analysis the directions of the $g=0$ and $\xi=0$ axes were taken from the analysis of the phase boundaries in the previous section. The values of $\phi$, $\delta_b$ and the position of the bicritical point $(H_b^2, T_b)$ have been varied, in order to study the effects of these variations on the scaling results. It appears that good estimates for these parameters can be obtained from the quality of the data collapsing. The value of $\omega = \omega^\parallel = \omega^\perp$, which is used in the normalization of $y$, was adapted simultaneously with a change in $\phi$ or a shift in $(H_b^2, T_b)$, in order to retain the best description of the paramagnetic phase boundaries $T^\parallel$ and $T^\perp$. 

Fig. 6.21.
Optimum data collapsing of $M^r_{st}$ and $M^\perp_{st}$ data, scaled according to (6.23) to yield the scaling functions $W$ and $F$. In the scaling process the parameter values (6.33) were used. In this double-logarithmic plot of $W^r \times y$ (defined by (6.26)) the predicted critical behaviour close to the paramagnetic phase boundaries (6.28) is represented by the solid lines. The dashed curves correspond to the theoretical prediction (6.31) for $W$ with $\epsilon=1$. 

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Fig. 6.21. shows the optimum result for the scaled data which was obtained for $T_b = 5.255 \text{ K}$, $\phi = 1.20$ and $\beta_b = 0.34$. Approximately 5500 data points from 35 field scans ($20 \text{ kOe} < H < 32 \text{ kOe}$) in the temperature range $3.5 \text{ K} < T < 5.7 \text{ K}$ are contained in these scaling plots.

Some data very close to the paramagnetic phase boundaries, which clearly contained a contribution from critical scattering, have been omitted. Close to the spin-flop line the data for $\mathcal{M}_{\parallel}^{\text{st}} = (\mathcal{M}_{\parallel}^{\text{st}})^2 + (\mathcal{M}_{\perp}^{\text{st}})^2$ have been used instead of the observed $\mathcal{M}_{\parallel}^{\text{st}}$ or $\mathcal{M}_{\perp}^{\text{st}}$-data, in order to correct for the rounding effect due to the small misalignment of the field (cf. eq.(6.12)).

In addition to the striking data collapsing, the symmetry between the $\mathcal{M}_{\parallel}^{\text{st}}$ and $\mathcal{M}_{\perp}^{\text{st}}$-data, which is expected for an $n=2$ spin-flop system, is obvious. Both $\mathcal{M}_{\parallel}^{\text{st}}$ and $\mathcal{M}_{\perp}^{\text{st}}$ show the expected asymptotic behaviour close the paramagnetic phase boundaries (6.28), as is shown by the straight lines for $1-y \rightarrow 0$, which have a slope $\beta = 0.325$, i.e. the theoretical value for a $d=3$ Ising system [20]. However, no crossover to the bicritical behaviour (6.27) is observed. This could not be expected either, because we are not observing the behaviour (6.26) of $\mathcal{M}_{\parallel}^{\text{st}}$ along the spin-flop line, as $y \rightarrow 0$, but rather the behaviour along a "pseudo-spin-flop line" at finite nonzero $H_z$ (compare Fig. 6.18.). In fact we are scanning in a slightly different way the temperature dependence of $\mathcal{M}_{\parallel}^{\text{st}}$ along this pseudo-spin-flop line, i.e. Fig. 6.11. We shall discuss this behaviour in more detail in the next section, together with the other results of the scaling analysis.

In Fig. 6.21. also a theoretical prediction for $\mathcal{M}^{\cdot}(y)$ is shown, derived from RG-calculations to first order in $\epsilon = 4-d$ [38] as

$$
\mathcal{M}^{\cdot}(y) = \left[ (n+8)(n-1)/8\pi^2 n c \right]^{1/3} (1-y)^{\beta} R^{1/3} (1-y) \cdot 
\left[ 1 - \frac{3\epsilon}{4} \ln \left( \frac{2(1-n^{-1})}{2(n+8)} R(1-y) \right) \right]. 
$$

The function $R(1-y)$ is given by

$$
R(1-y) = \frac{n-1}{n+8} (1-y)^{\epsilon/2} + \frac{9}{n+8}.
$$
For an \( n=2 \) spin-flop system (6.29) simplifies to

\[
\mathcal{W}(y) = \frac{1}{4\pi} \sqrt{\frac{10}{e}} (1-y)^{\beta} \left\{ \frac{1}{10} (1-y)^{5/2} + \frac{9}{10} \right\}^{1/2}.
\]  

(6.31)

In the critical region, i.e. \( y \rightarrow 1 \), the \((1-y)^{\beta}\) term ensures that (6.31) vanishes with the correct Ising exponent \( \beta \). Close to the spin-flop transition, i.e. \( y \rightarrow 0 \), expression (6.31) predicts a power-law behaviour with an exponent

\[
\beta + \epsilon/4 \gg \beta_b
\]

(6.32)

for all \( \epsilon \geq 1 \). Indeed (6.31) is supposed to hold only in the range \(-1 \leq y < 1\), i.e. not too close to the first-order spin-flop transition.

In Fig. 6.21, the expression (6.31) for \( \epsilon=1 \) is shown, with \( \mathcal{W}(y) \) scaled to coincide with the data for \( 1-y = 1 \). It appears that within the range of validity, the deviation of (6.31) from the single-power law (6.28) is much smaller than the scatter in our data. Therefore, no firm conclusions can be drawn about the correctness of (6.31).

The quality of the data collapsing becomes distinctly worse if one of the parameters \( T_b \), \( \beta_b \) or \( \phi \) in the scaling process, deviates from the optimum values used in Fig. 6.21. The most obvious is the effect of a change in \( T_b \). Both for \( T_b < 5.24 \) K and for \( T_b > 5.27 \) K absolutely none satisfactory data collapsing can be obtained for any value of \( \beta_b \) and \( \phi \). Within these boundaries for \( T_b \), the optimum values of \( \beta_b \) and \( \phi \) were estimated as \( \beta_b = 0.34(2) \) and \( \phi = 1.20(3) \). For parameter values \( T_b \), \( \beta_b \) and \( \phi \) outside these ranges, the data gathered in field scans at different temperature start to deviate from the single-power law behaviour at different \( y \)-values in the range \( 1 > 1-y > 0 \). In the scaling plots this looks like a fan which is unfurled. Although it is difficult to quantify the criteria used in judging the plots, we feel confident to give the following estimates for the parameters in CMB:

\[
T_b = 5.255(10) \text{ K}, \quad \phi = 1.20(3) \quad \text{and} \quad \beta_b = 0.34(2).
\]  

(6.33)
and \( \Phi \) (6.22). The values for \( \beta_0 \) and \( \phi \) are in good agreement with the theoretical predictions \( \beta_0 = 0.346(9) \) \( 39 \) and \( \phi = 1.175(15) \) \( 26 \), respectively.

**The scaling functions \( \Phi(x) \)**

The scaling functions \( \Phi(x) \) with \( x \) defined in (6.24), are especially suitable to analyze the behaviour of \( M_{st} \) in the \((\hat{g},\tilde{t})\) region close to the \( \tilde{t}=0 \) axis. As we discussed in section 3.4, the \( \hat{g} \)-variation of \( M_{st} \) along the \( \tilde{t}=0 \) axis is given by

\[
M_{st}(\tilde{t}=0) = |\hat{g}|^{\beta_0/b}/\phi.
\]

This implies that both branches \( \Phi^+ \) and \( \Phi^- \) must have the following asymptotic behaviour for \( x \rightarrow \infty \):

\[
\Phi^\pm(x) \sim x^{\beta_0/b}/\phi, \text{ for } x \rightarrow \infty.
\]

Close to the paramagnetic phase boundaries \( \tilde{g} = \pm \sqrt{\tilde{t}} \), \( \Phi(x) \) must display critical behaviour, i.e.

\[
\Phi^+(x) = (x-1)^\beta, \text{ for } x \rightarrow 1.
\]

With (6.26) the asymptotic behaviour of \( \Phi^+(x) \) close to the spin-flop transition \( x \rightarrow 0 \) is found as

\[
\Phi^+(x) = \frac{M^+(\tilde{g},\tilde{t})}{|\tilde{t}|^{\beta_0}} = \text{constant, for } x \rightarrow 0.
\]

In a plot of \( \ln \Phi^\pm(x) \) vs. \( \ln(x-1) \) the crossover from critical to bicritical behaviour, represented by (6.36) and (6.35) respectively, should be observable as a change in slope from \( \beta \) to \( \beta_0/\phi \). Similarly, a plot of \( \ln \Phi^\mp(x) \) vs. \( \ln(x) \) should show a change in slope from zero to \( \beta_0/\phi \) with increasing \( x \). The discussion of \( M_{st}^\perp(\tilde{g},\tilde{t}) \) proceeds in a completely similar way and therefore eqs. (6.35) to (6.37) also apply to \( \Phi^\perp \).
Optimum data collapsing of $M_{\parallel}$ and $M_{\perp}$ data, scaled according to (6.22), to yield the scaling functions $M_{\parallel}$ and $M_{\perp}$. Both functions consist of two branches $M_{+}$ (squares) and $M_{-}$ (circles). The parameter values (6.23) have been used. In this plot of $\ln M_{\parallel} =: \ln(x-x_0)$, $x$ is defined by (6.24), $x_0 = 0$ for $M_{+}$ and $x_0 = 1$ for $M_{-}$. The solid lines correspond to the asymptotic bicritical (cf. (6.35)) and critical (cf. (6.36)) behaviour. The shaded lines enclose the 25% crossover regions defined in the main text.
Fig. 6.22. shows the optimum scaling results for the data on CMB in a plot of $\ln\left[\frac{\tilde{\mu}(\tilde{t},\tilde{\xi})}{\tilde{\xi}^{\tilde{b}_{||}}(\tilde{t})}\right]$ vs. $\ln(x-x_{C})$, obtained with the parameter values (6.33). Here $x_{C} = 1$ for $\mu_{+}$ and $x_{C} = 0$ for $\mu_{-}$. Again the data collapsing is excellent and the symmetry between $\mu_{+}$ and $\mu_{-}$ is striking. The predicted asymptotic critical behaviour (6.36) of $\mu_{+}$ and $\mu_{-}$ for $x-x_{C} \to 0$ appears to be fulfilled. Least-squares fits of (6.36) to the data with $(x-x_{C}) < 1.4$ yield the slopes $\beta_{||} = 0.321(4)$ and $\beta_{\perp} = 0.326(4)$. These values may be compared with the estimates from the separate field scans in the previous section (Table 6.2.). Both are in good agreement with the theoretical predictions $\beta = 0.325(1)$ (c-expansion |20|) and 0.312(5) (series |21|). The predicted bicritical behaviour of $\mu_{+}$ for $x-x_{C} \to +0$ (cf. (6.35)) is clearly confirmed, too. Although the difference between the theoretical values for $\beta$ and $\beta_{b}/\phi$ is only small, the crossover from critical to bicritical behaviour is obvious. Least-squares fits of (6.35) to the data with $(x-x_{C}) > 30$ yield the slopes $(\beta_{b}/\phi)_{||} = 0.288(2)$ and $(\beta_{b}/\phi)_{\perp} = 0.280(3)$. Of course, these results may not be considered as independent estimates for $\beta_{b}/\phi$, since the exponents $\beta_{b}$ and $\phi$ separately have been used in the scaling procedure. However, these values demonstrate that the disappearance of the order parameter in the bicritical region along the $t=0$ axis is described correctly by the exponent $\beta_{b}/\phi = 0.34/1.2 = 0.283$, in agreement with (6.34). Also $\mu_{-}$ shows the predicted asymptotic behaviour (6.35) for $x-x_{C} \to 0$. For $\mu_{+}$ we find $(\beta_{b}/\phi)_{||} = 0.280(2)$ and for $\mu_{-}$ the slope $(\beta_{b}/\phi)_{\perp} = 0.282(4)$ is obtained. For $x-x_{C} \to 0$ the slope of $\mu_{-}$ strongly decreases, but apparently a small slope of approximately 0.03 remains present for small $x-x_{C}$. This can be explained as an effect of the small misalignment of the magnetic field.

In order to obtain a more quantitative picture of the crossover from critical to bicritical behaviour in $\mu_{+}$ and from bicritical to first-order behaviour in $\mu_{-}$, we have determined in least-squares fits for each half-decade in $x-x_{C}$ the effective exponent $\beta_{eff}$, which corresponds to the observed local slope in Fig. 6.22. |40|. The results for $\mu_{+}$ and $\mu_{-}$ are shown in Fig. 6.23., where $x_{r}$ indicates the center of the related $(x-x_{C})$ range. In the same figure, the exponents are shown, which result from least-squares fits of a single power law to all data with $(x-x_{C}) < x_{r}$ as well as the results for all data with $(x-x_{C}) > x_{r}$.
The effective exponents $\beta_{\parallel}$ and $\beta_{\perp}$, determined from $\alpha_{\parallel}$ and $\alpha_{\perp}$ for each half decade in $(x-x_c)$ centred around $x_c$ (open circles). Triangles and dots correspond to the exponents $\beta$ resulting from least-squares fits of a power law to all data with $x-x_c < x_p$ and $x-x_c > x_p$, respectively. The dashed line approximately represents the crossover from $\beta_{\parallel}$ to $\beta_{\perp}$.

Fig. 6.22.

These combined data give a good impression of the crossover of $\beta_{\text{eff}}$ in spite of the small difference between the asymptotic values $\beta$ and $\beta_{b/\phi}$. The arrows indicate the $(x-x_c)$ values where $\beta_{\text{eff}}$ is estimated to deviate 25% of $|\beta - \beta_{b/\phi}|$ from either $\beta$ or $\beta_{b/\phi}$. The 25% crossover regions both for $\alpha_{\parallel}$ and $\alpha_{\perp}$ are also indicated in Fig. 6.22, and their location in the $h^2$ vs. $T$ phase diagram is shown in Fig. 6.24. The widths of the 25% crossover regions in the scaling plot Fig. 6.22, determined as

$$W_{\text{CR}}^{+}(0.25) \approx 10^{0.8}$$

for $\alpha_{\parallel}$, and as

$$W_{\text{CR}}^{-}(0.25) \approx 10^{1.5}$$

for $\alpha_{\perp}$, appear to be in good agreement with the predictions of Riedel.
and Wegner |40|. A further discussion of these results is presented in the next section.

![Diagam](image)

Fig. 6.14.
Part of the $H^2$ vs $T$ diagram of CMB (cf. Fig. 6.13.), with the experimentally determined cross-over regions in the ordered phases enclosed by shaded lines.

6.9. Discussion and conclusions

Relevance of the present investigation

Up to now most experiments on bicritical behaviour have been confined to an accurate determination of the shape of the magnetic phase boundaries |29-34|. Comparison with the extended-scaling prediction (6.14) leads to an estimate of the crossover exponent $\phi$, the ratio of amplitudes $Q(n) = \nu_{\parallel}/\nu_{\perp}$ and the direction $q$ of the $\tilde{t}=0$ axis. In most cases either $Q$ or $q$ has been determined with the other quantity fixed, because of their strong mutual correlation. This prevents an unambiguous determination of the $(n=2)$ or $(n=3)$ character of the system, since Fisher's estimate (6.18) for $q$ appears to be not very reliable. In all materials investigated up to now, it has been possible to achieve a good description of the observed phase boundaries both with the $n=2$ values $Q(2) = 1$, $\phi = 1.175(15)$ and with the $n=3$ values $Q(3) \approx 2.51$, $\phi \approx 1.25$, if $q$ is treated as an adjustable parameter. Also in the present analysis of the phase diagram of CMB, the $q$-estimate appears
to be completely unreliable because of the pseudo one-dimensional characteristics of CMB. So, also in this study the n=2 character of CMB could not be extracted from the shape of the phase boundaries, but was ultimately confirmed by the complete similarity of the scaling plots for $\delta M$ and $\delta M'$. More interesting than the determination of the phase boundaries are studies of the behaviour of a critical quantity in the vicinity of the bicritical point. To our knowledge such studies thusfar were only performed on GdAlO$_3$, for which compound the jump in the homogeneous magnetization $\Delta M(\vec{c}<0, \vec{g}=0)$ along the spin-flop line and the homogeneous susceptibility $\chi$ along the $\vec{g}=0$ axis ($t>0$) have been reported [28]. Also the behaviour of $\Delta M$ at small nonzero $H_1$ and in particular the crossover in $\Delta M$ from bicritical behaviour to the critical behaviour corresponding to the critical edge of the spin-flop shelf $T_{cSF}$ (Fig.3.3) has been studied [35].

The present investigation yields the first experimental test of the central hypothesis in the theory of bicritical behaviour, i.e. of the extended-scaling hypothesis (6.21). The neutron scattering data on CMB provide the possibility of an extensive scaling analysis, in which all theoretical predictions are fully confirmed. Crossover of critical to bicritical behaviour is observed in $M_{st}(H,T)$ and the corresponding critical exponents have been determined.

The importance of an exact field alignment

In the papers of Rohrer et al. [34,35], the importance of an exact alignment of $\vec{H}$ along the easy axis for a correct determination of bicritical exponents is emphasized. A mismatch angle of $10^{-3} - 10^{-2}$ degrees would change the behaviour in a crucial way. On the contrary we prove in the present experiment on CMB that it is possible to determine bicritical exponents in a magnetic field which is misaligned by 0.5°, which is much greater than the critical angle $\psi_c$. In this discussion we shall show that both findings are not contradictory but more or less complementary. To understand this, one must consider within which region in $(H, H_1, T)$-space the data are collected which are used in the analysis of the bicritical behaviour.
Rohrer et al. [28,35] state correctly that a very precise alignment of the easy axis is essential in order to obtain bicritical exponents from measurements in the \((H_{\parallel},T)\)-region close to the \(g=0\) axis. Indeed, we have found a rounding of the paramagnetic phase boundaries close to the (pseudo-) bicritical point (Figs. 6.18., 6.20.). We have detected also that the jump in \(M\) behaves more or less Ising-like rather than XY-like along the \(g=0\) axis (Fig. 6.11.). We did not observe a crossover from critical \((\eta=1)\) to bicritical \((\eta=2)\) behaviour in the scaling functions \(\phi^\eta(y)\) and \(\phi^1(y)\) for \(y=\infty\) (Fig. 6.21.). Neither the scaling functions \(\phi(x)\) and \(\phi^1(x)\) completely constant for \(x\to 0\), but retained a small slope corresponding roughly to the difference between \(B\) and \(B\) (Fig. 6.22.).

However, in the present experiment the most interesting information is not extracted from the regions close to \(g=0\), but from the entire \((H_{\parallel},T)\)-space and especially from the bicritical region around the \(g=0\) axis (Fig. 6.24.). The advantage of this bicritical region in \((H_{\parallel},H_1,T)\)-space is that it extends far from the bicritical point, like a wheel with the \(g=0\) axis in the \((H_{\parallel},T)\) plane as a spindle, and that it is unaffected by critical lines close by. Therefore data from a wide region, also far from the \(H_1=0\) plane, can be used to determine bicritical behaviour. Apparently the relatively small difference between the positions of the bicritical point \((T_b,H_1=0)\) and the pseudo-bicritical point \((T'_b,H_1,small)\) appears to be negligible for the behaviour in the \((H_{\parallel},H_1,T)\)-plane at constant \(H_1/H_{\parallel}\). As shown in Fig. 6.22., even for large \(x\)-values no deviation from a single power law with bicritical exponent \(8/\delta\) is observed.

When the location of the \(g=0\) axis is known, it should be possible to attain a very precise and independent determination of the exponent \(8/\delta\) from direct measurements of the intensity variation of a magnetic Bragg reflection along \(g=0\). This would be a much simpler experiment than the investigations along the \(g=0\) axis. A disadvantage of the region close to \(g=0\) is that in that region only the staggered magnetization and the staggered susceptibility are really relevant quantities, which are only measurable by neutron scattering experiments.
Conclusions

The results of the present neutron scattering experiment on CMB may be summarized by stating that the predictions of the extended-scaling hypothesis, which is underlying renormalization-group calculations and the theory on bicritical behaviour, are fully confirmed. Even in this pseudo d=1 system the shape of the phase boundaries near the bicritical point appear to be predicted correctly. Fisher's estimate of the non-universal value q is not appropriate, as might be expected from the MF-type of approach used in the determination, which clearly is not applicable in pseudo d=1 systems.

A much stronger verification of the extended-scaling hypothesis itself is the striking collapsing of scaled data from the entire (H, T)-diagram on two scaling functions \(f^I\) and \(f^\perp\) (or alternatively on \(f^I\) and \(f^\perp\)). Crossover from critical to bicritical behaviour has been observed in \(f^I\) and \(f^\perp\). The corresponding critical exponents \(\beta\) and \(\beta^b\) appear to be in excellent agreement with the d=3 Ising and XY-values, respectively, as predicted by RG-calculations [20,39]. Also the theoretical prediction for the crossover exponent \(\phi\) has been confirmed. It is the first time that such a penetrating experimental verification of the extended-scaling hypothesis in spin-flop systems has been performed.
Appendix

Determination of \( M^\parallel (H) \) and \( M^\perp (H) \) from the peak intensities \( I_{ij}(H) \)

In order to determine the best estimates for the scale factor \( C_j \) and for the non-magnetic background \( B_j = B_{0j} \) in each separate scan \( i_j(H,T) = I_{hOj}(H,T) \), the three available scans have been compared mutually in a least-squares fitting program. With the observed intensities in a scan denoted by

\[
I_j(H,T) = C_j \left[ M^\parallel_{st}(H_j,T) \right]^2 + B_j \quad ; \quad j = 1, 2, 3 \tag{A 6.1}
\]

in the AF-phase and by

\[
I_j(H,T) = C_j \sin^2 \theta_j \left[ M^\perp_{st}(H_j,T) \right]^2 + B_j \quad ; \quad j = 1, 2, 3 \tag{A 6.2}
\]

in the SF-phase, the fitting procedure implies that for each \( T \)-value the quantities

\[
\sum_{H_i \in AF \text{ cycl.}} \left\{ I_j(H_i,T) - a_j^\parallel I_{j+1}(H_i,T) - b_j^\parallel \right\}^2 ; \quad j = 1, 2, 3 \tag{A 6.3}
\]

and

\[
\sum_{H_i \in SF \text{ cycl.}} \left\{ I_j(H_i,T) - a_j^\perp I_{j+1}(H_i,T) - b_j^\perp \right\}^2 ; \quad j = 1, 2, 3 \tag{A 6.4}
\]

are minimized. Here

\[
a_j^\parallel = C_j/C_{j+1} \quad ; \quad a_j^\perp = a_j^\parallel \sin^2 \theta_j / \sin^2 \theta_{j+1} \tag{A 6.5}
\]

\[
b_j^\parallel = B_j - a_j^\parallel B_{j+1} \quad ; \quad b_j^\perp = B_j - a_j^\perp B_{j+1} \tag{A 6.6}
\]

Since the angles \( \theta_j \) are known, estimates for \( C_j \) and \( B_j \) can be obtained from the fitted parameters \( a_j^\parallel, a_j^\perp \) and \( b_j^\parallel, b_j^\perp \). The symbols \( H_i \in AF, SF \) indicate that for the fits only those field ranges were used, which undoubtedly belong to the AF-phase and SF-phase, respectively. Although the estimates obtained in this way were satisfactory, the \( a_j^\parallel, a_j^\perp \) and \( b_j^\parallel, b_j^\perp \) values appeared to be strongly correlated.
In order to improve the accuracy of the estimates, the values of the background scattering \( L \) were derived first from fits in which only \((H,T)\)-regions were taken into account where the magnetic contributions to the peak intensities are very small. In these fits first-order approximations for the \( C_j \)-values were used. Subsequently, better \( C_j \)-values were derived from fits which used only \((H,T)\)-regions where the magnetic intensities are much higher than the background. From this procedure a set of \( C_j \) and \( B_j \)-values resulted, which leads to scaled field scans \( I_{j}^{SC}(H,T) = (I_{j} - B_{j})/C_{j} \) for all three reflections, which coincide perfectly at all \((H,T)\)-values in the AF-phase. A good example of the quality of this data collapsing in two completely different \((h,T)\)-regions is shown in Fig. 6.7. From this result it may be concluded in the first place that extinction effects can be neglected. Secondly, that the \( c \)-axis is proven to be the intermediate direction, indeed, since the scaled intensities are found to be proportional to the corresponding \( \sin^2 \theta \) in the SF-phase.

With the obtained parameter set, the contributions of \( M^\parallel \) and \( M^\perp \) to the intensity of a reflection can be separated close to the AF-SF phase boundary. For the scaled intensities

\[
I_{j}^{SC} = (I_{j} - B_{j})/C_{j} = (M^\parallel_{st})^2 + \sin^2 \theta_{j} (M^\perp_{st})^2 \quad j=1,2,3 \quad \text{(A 6.7)}
\]

standard deviations \( \sigma_{I_{j}} \) have been derived from the counting statistics of \( I_{j} \) and from the estimated uncertainties in \( B_{j} \) and \( C_{j} \). Then \( (M^\perp_{st})^2 \) has been determined as

\[
(M^\perp_{st})^2 = \sum_{j} \left\{ C_{j}^\perp (I_{j}^{SC} - I_{j+1}^{SC})/(\sin^2 \theta_{j} - \sin^2 \theta_{j+1}) \right\} \sum_{j} C_{j}^\perp , \quad j=1,2,3 \quad \text{cyc1.} \quad \text{(A 6.8)}
\]

where

\[
C_{j}^\perp = \left[ (\sigma_{I_{j}})^2 + (\sigma_{I_{j+1}})^2 \right]^{-1} . \quad \text{(A 6.9)}
\]

\( (M^\parallel_{st})^2 \) has been derived as
\[(a^{st}_{\perp})^2 = \frac{1}{j} \left\{ \left[\sum_{j}^{\infty} \frac{S_{0,j}^{2} - 1}{S_{0,j}^{2}} \right] \left[\frac{\sin^2 \theta_j - \sin^2 \theta_{j+1}}{\sin^2 \theta_j} \right] \right\} \frac{1}{J_{0,j}^2}, \]

\[j = 1, 2, 3 \text{ cycl.} \quad \text{(A 6.10)}\]

with \[G_j = \left[ \left( \delta_{j+1} \right)^2 \sin^2 \theta_j + \left( \delta_j \right)^2 \sin^2 \theta_{j+1} \right]^{-1}. \quad \text{(A 6.11)}\]

**References to Chapter VI**

[34] R.A. Butera and D.R. Rutter, J. Appl. Phys. 49, 1344 (1978) on MnCl₂·4H₂O.
Summary

This report starts with a rather extensive presentation of the concepts and ideas which constitute the basis of the modern theory of static critical phenomena. It is shown how at a critical point the semi-phenomenological concepts of universality and scaling are directly related to the divergence of the correlation length and how they are extended to a calculational method for critical behaviour in Wilson's Renormalization-Group (RG) approach. Subsequently the predictions of the molecular-field and RG-theories on the phase transitions and critical behaviour in weakly anisotropic antiferromagnets are treated. In a magnetic field applied along the easy axis, these materials can display an (H,T) phase diagram which contains either a bicritical point or a tetracritical point. Especially the behaviour close to these multi-critical points, as predicted by the extended-scaling theory, is discussed.

The experimental part of this study is contained in the chapters IV-VI. Results are presented of elastic and quasi-elastic neutron-scattering studies on the critical and multi-critical behaviour in two low-dimensional, weakly anisotropic antiferromagnets, viz. \( \text{CoBr}_2.6\{0.48\text{D}_2\text{O},0.52\text{H}_2\text{O}\} \) (CB48) and \( \text{CsMnBr}_3.2\text{D}_2\text{O} \) (CMB). In chapter IV we report the rather complete picture which has been obtained of the critical behaviour of CB48 near the Neel point. Although CB48 at high temperature is a good approximation of the two-dimensional \((d=2)\) XY-model, the present experiment shows that the critical behaviour may be described as \(d=3\) Ising-like. Both the critical exponents and the critical amplitudes that were obtained are in good agreement with the theoretical values for the \(d=3\) Ising model.

In chapter V the magnetic phase diagram of CB48 in a parallel magnetic field is presented. Strong evidence has been found for the existence of an intermediate (I) phase between the antiferromagnetic and the spin-flop phase, which implies that the \((H,T)\)-diagram contains a tetracritical instead of a bicritical point. This would be the first time that an I-phase with coupled order parameters has been observed in an antiferromagnetic system.

The neutron-scattering data on CMB (chapter VI) are used to verify
the extended-scaling hypothesis, which is the central assumption in the theory and (RG-) calculations of multi-critical behaviour. Even in this pseudo-d=1 system the shape of the phase boundaries near the bicritical point is found to be correctly predicted by the extended-scaling theory. Moreover, a striking data-collapsing of the scaled order parameters $M_{\parallel}^{\mathrm{st}}(H,T)$ and $M_{\perp}^{\mathrm{st}}(H,T)$, determined in the entire $(H_{\parallel},T)$-phase diagram, on two scaling functions could be achieved. This is a very direct confirmation of the extended-scaling hypothesis. Crossover from critical to bicritical behaviour has been observed. The corresponding critical exponents $\beta$ and $\beta_{h}$, and the crossover exponent $\phi$ are found to be in good agreement with the theoretical values.
LIST OF ABBREVIATIONS AND SYMBOLS

AF     antiferromagnetic (phase)
AFMR   antiferromagnetic resonance
CBX    CoBr$_2$.6[xD$_2$O,(1−x)H$_2$O]
CBx    CBx with x=0.48
CMB    CaMnBr$_3$.2D$_2$O
CXS    coexistence surface
I      intermediate (phase)
LRO    long-range order
MF     molecular field
NMR    nuclear magnetic resonance
P      paramagnetic (phase)
RG     renormalization group
SF     spin-flop (phase)

\hat{A}   inter-sublattice interaction     63
\hat{D}   intra-sublattice interaction     63
D        \equiv D_x - D_y                    65
d        spin distribution function         38
G        Gibbs free energy                   8
G^{G8}(\vec{R},t)  space-time correlation function 13
G^{G8}(\vec{R},t)  net space-time correlation function 13
\tilde{G}^{G8}(\vec{R})  \equiv G^{G8}_n(\vec{R},t=0), net static correlation function 15
\tilde{G}^{G8}(\vec{q},\omega)  doubly Fourier-transformed \tilde{G}^{G8}(\vec{R},t) 14
\tilde{G}^{G8}(\vec{q})  \equiv G^{G8}_n(\vec{q}), Fourier transform of G^{G8}_n(\vec{R}) 15
H        magnetic field along the easy axis   61
H_A      anisotropy field                     61
H_ex     exchange field                       88
H_{IA}   AF → I transition field             68
H_{IS}   SF → I transition field             68
H_{SF}   AF → SF transition field, spin-flop field 61
\hat{H}_{st}  staggered magnetic field      11
H  Hamiltonian expressed in \vec{Q}-dependent quantities 40
\hat{H}  effective Hamiltonian               34
\tilde{H}  reduced Hamiltonian              39
| \( p \) | \( m \)-times renormalized reduced \( p \) | 40 |
| \( J(q) \) | Fourier-transformed exchange interaction | 40 |
| \( J(\vec{R}_1-\vec{R}_j) \) | linearized RG-transformation | 51 |
| \( m_{st} \) | staggered magnetization | 11 |
| \( \mathcal{F}(x) \) | scaling function of the order parameter | 33 |
| \( \mathcal{Q} \) | \( \equiv k-k_0 \), scattering vector | 13 |
| \( \mathcal{Q} \) | \( \equiv \hat{Q}/Q \) | 14 |
| \( \mathcal{Q} \) | RG-transformation | 44 |
| \( \tau_c^{1,1}(H) \) | paramagnetic phase boundaries | 67 |
| \( \tau_c^{1,1}(y) \) | scaling function of the order parameter | 33 |
| \( \tau_c^{1,1}(c) \) | scaling factor for spin values | 28 |
| \( d \) | spatial dimensionality | 6 |
| \( g_{ph} \) | anisotropy parameter | 76 |
| \( \xi \) | optimum scaling field | 81 |
| \( n \) | spin dimensionality | 6 |
| \( t \) | \( \equiv \hat{Q}-2\pi \hat{r}, \) deviation vector | 15 |
| \( s \) | scaling factor for lengths and momenta | 28 |
| \( \xi \) | optimum scaling field | 71 |
| \( \xi \) | \( S^q \) term in \( \xi \) | 42 |
| \( \xi_0 \) | dangerous irrelevant variable | 76 |
| \( \xi_1,\xi_2 \) | factors in the shape of \( \tau_c^{1,1}(H^2) \) | 84 |
| \( \chi_{x,y} \) | scaling variables | 33 |
| \( \alpha,\beta,\gamma \) | critical exponents | 21 |
| \( \alpha_3,\beta_3,\gamma_3 \) | principal axes of the \( \chi \)-tensor | 96 |
| \( \alpha,\beta,\gamma \) | bicritical exponents | 80 |
| \( \epsilon \) | \( \equiv 4-d \) | 25 |
| \( \kappa \) | inverse correlation length of a spin components | 15 |
| \( \nu \) | critical exponent of \( \xi \) | 20 |
| \( \xi \) | correlation length | 6 |
| \( \tau \) | reciprocal lattice vector | 15 |
| \( \phi \) | crossover exponent | 80 |
| \( \chi_{x,y}(Q) \) | generalized susceptibility tensor | 18 |
| \( \chi_{st}(Q) \) | staggered susceptibility tensor | 19 |
| \( \psi_c(T) \) | critical angle | 86 |