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# Random-Field Effects on Field-Induced Transitions in Ising-Type Antiferromagnets

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## Abstract

Random-field effects on 3-dimensional (3-d) and quasi 2-d weakly anisotropic Heisenberg antiferromagnets with easy-axis type anisotropy are discussed. The effective, field-dependent anisotropy model is used. This model yields an accurate and quantitative description of the effects of an applied field on the intrinsic properties of the domainwalls, i.e., the wall tension  $\sigma$  and the wall-thickness  $d_s$ . A comparison with experimental data for  $K_2Mn_{(1-x)}Mg_xF_4$  (quasi 2-d) and  $Mn_{(1-x)}Zn_xF_2$  (3-d) is given. The differences with the strongly anisotropic 3-d antiferromagnetic,  $Fe_{(1-x)}Zn_xF_2$ , are briefly considered. Furthermore it is argued that weakly anisotropic systems offer very promising possibilities for the study of the peculiar relaxation and metastability effects that are so characteristic for random field Ising systems.

## 1. Introduction

Recently, random-field effects in 3-dimensional (3-d) and quasi 2-d Ising-type antiferromagnets have been extensively studied [1–3]. Most efforts have been concentrated on the investigation of the critical behaviour at the phase boundary  $T_c(\mathbf{B})$ . This problem is still not understood although it has already led to basically new ideas about equilibrium critical properties and critical slowing down in random-field Ising systems [4].

The present paper deals with another novel aspect of the topic. For weak Ising-type anisotropy there is a transition in the low-temperature phase which is field-induced, namely the spinflop transition that occurs for a critical value  $B = B_{sf}$  of the applied magnetic field. A very weak rms random field  $B_r$ , already has drastic effects on this transition in the quasi 2-d case. This is evidenced by the experimental observation that even in nominally pure systems the first-order character of the spinflop is destroyed, which yields a strong indication that for  $T < T_c(\mathbf{B})$  a domain structure is always present in the 2-d case. In the 3-d system on the other hand there is no such effect at all. This is in full agreement with the recent theoretical proof of Imbrie [5] that the lower critical dimension  $d_{lc} < 3$ . It should be noted that the occurrence of domains near  $B_{sf}$  in the 2-d case at low temperature can only be understood by the presence of weak random fields [6].

We also argue that investigations of the spinflop effect may contribute to the understanding of the metastability problem which has arisen from the investigations of randomly diluted 3-d systems [2, 3] and which is of high current interest. From our considerations it follows that the weakly anisotropic antiferromagnet may be brought into a region where the relaxation times should be of the order of minutes or even shorter. In that case the metastability behaviour will be accessible to various experimental techniques, a.o. neutron diffraction, NMR, the Mössbauer-effect and frequency dependent susceptibility measurements.

In the present paper we focus mainly on field-induced effects connected with the random field problem. More about field-induced effects in weakly anisotropic Heisenberg antiferromagnets may be found in Ref. [6].

## 2. Domainwalls in the weakly anisotropic antiferromagnet

The central item in our interpretation of the various magnetic phenomena that occur in the weakly anisotropic antiferromagnetic system is the concept of the effective, field-dependent anisotropy [6, 7]. It may be applied to quasi 1-d, quasi 2-d and 3-d cases [6] and is very useful for the description of the domainwalls since it yields an explicit expression for the field-dependence of the surface tension  $\sigma$  and the wall-thickness  $d_s$ . In nearly all theoretical papers one of the basic assumptions is that the wall properties  $\sigma$  and  $d_s$  are irrelevant and can be left out of the discussion. To our opinion this assumption is wrong for weakly anisotropic systems, which is what makes them so interesting.

In general the weakly anisotropic antiferromagnet may be modeled by a classical isotropic interaction hamiltonian with orthorhombic anisotropy

$$H = -2J \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_k (D_z S_{kz}^2 - D_x S_{kx}^2 + g\mu_B \mathbf{B} \cdot \mathbf{S}). \quad (1)$$

Here  $J < 0$  is the isotropic nearest-neighbour exchange interaction and the anisotropies  $D_z$  and  $D_x$  together establish an easy ( $xy$ -)plane for the magnetic moments  $\mathbf{S}$  with the  $x$ -axis as the preferential (Ising-)axis within this plane. The last term in hamiltonian (1) represents the conventional way of including an applied magnetic field  $\mathbf{B}$ . As is explained in detail in [6], for  $\mathbf{B}$  along the easy axis ( $\mathbf{B} = B_x$ ) or along the intermediate axis ( $\mathbf{B} = B_y$ ) one may unite the last two terms in hamiltonian (1) into one effective, field-dependent anisotropy term  $D_{\text{eff}}^x S_{kx}^2$  with

$$D_{\text{eff}}^x = D_x(1 - B_x^2/B_{sf}^2) \quad \text{for } \mathbf{B} = B_x \quad \text{and} \quad (2a)$$

$$D_{\text{eff}}^x = D_x(1 + B_x^2/B_{sf}^2) \quad \text{for } \mathbf{B} = B_y. \quad (2b)$$

This effective, field-dependent anisotropy finds its origin in the difference between the magnetic susceptibility parallel ( $\chi_{\parallel}$ ) and perpendicular ( $\chi_{\perp}$ ) to the easy axis of an antiferromagnet. At low temperatures one has  $\chi_{\parallel} \ll \chi_{\perp}$ . In that case  $D_x$  and  $B_x$  are competing, since the latter tends to polarize the moments perpendicular to itself and thus perpendicular to the easy ( $x$ )axis associated with  $D_x$ . At low temperatures the small  $\chi_{\parallel}$  can be neglected and the net anisotropy  $D_{\text{eff}}^x$  equals the differences between the anisotropy energy  $D_x S^2$  and the Zeeman energy  $\frac{1}{2}\chi_{\perp} B_x^2$ . The spinflop field  $B_{sf}$  corresponds to  $D_{\text{eff}}^x = 0$  and is determined through the equation  $\frac{1}{2}\chi_{\perp} B_{sf}^2 =$

$D_x S^2$ . For  $B_x < B_{sf}$  one has  $D_{\text{eff}}^x > 0$ , and the moments are on the average along the  $x$ -axis. For  $B_x > B_{sf}$  on the other hand one has  $D_{\text{eff}}^x < 0$  so that the moments are mainly perpendicular to the  $x$ -axis and are, in fact, polarized along the  $y$ -axis, due to the additional presence of the positive  $D_z$  term.

We also emphasize that in case of orthorhombic anisotropy the symmetry is of the easy-axis type, both for  $B_x < B_{sf}$  and for  $B_x > B_{sf}$ . Since  $D_{\text{eff}}^x \rightarrow 0$  for  $B_x \rightarrow B_{sf}$  it is the  $D_z$ -term that determines the symmetry around the spinflop transition at the lowest temperatures. Thus only for  $B_x = B_{sf}$  the system becomes of the easy-plane type since then  $D_{\text{eff}}^x = 0$  and  $D_z > 0$ . This will happen even in case of very small  $D_z$ , and since most uniaxial systems will have some orthorhombic component, e.g., due to higher order effects, it is the orthorhombic description which is most suitable for the interpretation of the low-temperature behaviour near to  $B_{sf}$ .

It may be noted that in case of a field parallel to the intermediate axis,  $\mathbf{B} = B_y$ , one may also define an effective anisotropy, cf. eq. (2b). Here the field-induced anisotropy strengthens the easy-axis anisotropy, so that  $D_{\text{eff}}^x$  increases starting from  $D_x$  at  $B_y = 0$ .

The concept of effective, field-dependent anisotropy may be applied to describe quantitatively ( $\sigma$ ,  $d_s$ ) the domainwall in the antiferromagnetic system, for  $B_x < B_{sf}$  [Fig. 1(a)] and for  $B_x > B_{sf}$  [Fig. 1(b)]. An order of magnitude estimate of  $\sigma$  and  $d_s$  for such walls can be obtained using a simple classical mean-field domainwall model [8]. A more sophisticated description can be given [6] on basis of the continuum approximation to Hamiltonian (1), namely the classical 2-d sine-Gordon (SG-)system. Here the domainwalls correspond to the soliton excitations in the SG-system. Taking into account the effective anisotropy one finds

$$\sigma = 4S^2 |D_{\text{eff}}^x(\mathbf{B})J|^{1/2} \quad \text{and} \quad (3)$$

$$d_s = |J/D_{\text{eff}}^x(\mathbf{B})|^{1/2}. \quad (4)$$

It follows immediately that  $\sigma \rightarrow 0$  and  $d_s \rightarrow \infty$  for  $B_x \rightarrow B_{sf}$  and that both quantities vary continuously with  $B_x$ .

It is of interest to connect the above results to the random-field Ising model. The importance of the field-induced anisotropy effects becomes clear if one considers the random-field problem in the original form given by Imry and Ma [9]. In essence it comes down to a competition between the random-field energy and the surface tension of the domainwall. According to the Imry–Ma criterion the energy of a

domain can be estimated by:

$$E_D = \sigma R^{d-1} - B_r R^{d/2}, \quad (5)$$

with  $B_r$  the rms random field and  $R$  the radius of the domain. For  $x < 2$  one has  $E_D < 0$  and the system prefers a domain structure. Recent careful calculations [5, 10] have established that also for  $d = 2$  there is no long-range order.

Consider now a weakly anisotropic antiferromagnet with  $d < d_{lc}$ , subject to a weak random field  $B_r$ . In that case the domains will be large in the absence of an applied field, since  $\sigma$  will be substantial and the domainwalls are fairly narrow (5–10 lattice units). Application of a field  $B_x$  will however decrease  $\sigma$ , and therefore influences the balance between the wall energy and the random field energy, cf. eq. (5), through the *first term*. Consequently the wall-density  $q^{-1}$  should increase, provided that the system stays at equilibrium.

An interesting consequence of this argument is that in order to study random-field effects in weakly anisotropic antiferromagnets there is no strict need to make use of Fishman and Aharony's device [11]. They were the first to remark that an external field  $B_x$ , applied to an antiferromagnet that is randomly diluted with nonmagnetic impurities, will give rise to a random staggered field  $B_r^x \propto B_x$ . This allows the variation of the *second term* in eq. (5). In case of strong anisotropy or low fields and a fairly large amount of nonmagnetic impurities the variation with  $B_x$  of the second term should predominate the first. However, for weak anisotropy and  $B_x \approx B_{sf}$  the variation of the first term may become predominant. Furthermore, for nominally pure systems the random field will hardly depend on the applied field since the very small  $B_r$  that is present should be mainly due to lattice defects and the like. In such cases the first term may still be tuned by the external  $B_x$ . This opens the possibility of investigating the random field effects by means of experiments on formally pure systems, where the additional random-exchange effects arising from nonmagnetic impurities are much less interfering than in heavily diluted systems.

A problem that has attracted much attention recently is the peculiar metastability phenomenon observed in the various experimental cases [2, 3]. It has been found that relaxation of the wall-pattern towards its equilibrium configuration at  $T < T_c(B_x)$  proceeds extremely slow, which has been attributed to pinning of the walls by defects and impurities [12]. This would imply for the present case that, notwithstanding the decrease of  $\sigma$  as  $B_x \rightarrow B_{sf}$  the wall density  $q^{-1}$  would not change, at least not on the timescale of the experiment. As long as  $B_x \ll B_{sf}$  this may happen indeed. However, for  $B_x \approx B_{sf}$  the  $d_s$  will increase and may become of the order of 10–100 lattice units (l.u.). In that case the relaxation times may become quite short. Since  $d_s$  varies continuously with  $B_x$  also the relaxation behaviour may thus be tuned with the field, and probably with the temperature. In particular for temperatures in the vicinity of the phase boundary  $T_c(\mathbf{B})$  one would expect a thermal meandering of the walls.

### 3. Experiments

Below we present some experimental data, supporting the ideas presented in the above. In Fig. 2(a) we show the phase diagram of the quasi 2-d compound  $\text{K}_2\text{MnF}_4$  [13], for fields parallel and perpendicular to the easy axis, whereas Fig. 2(b)

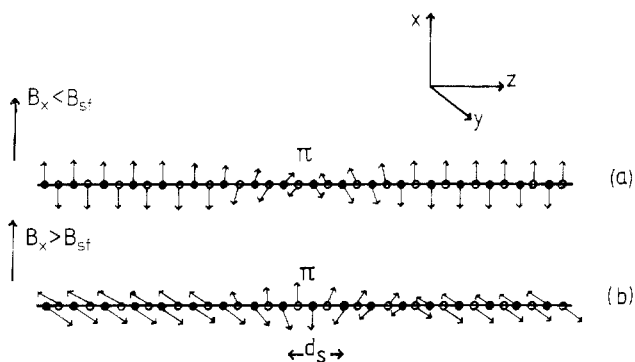


Fig. 1. Domainwalls in weakly anisotropic antiferromagnets (schematic) for  $B_x < B_{sf}$  (a) and for  $B_x > B_{sf}$  (b). The total width is approximately  $\pi d_s$  and the moments on both sublattices rotate through an angle  $\pi$ . The wall corresponds either to a fragment of the flopped phase in the low field phase (a) or to a fragment of the low-field phase in the flopped phase (b).

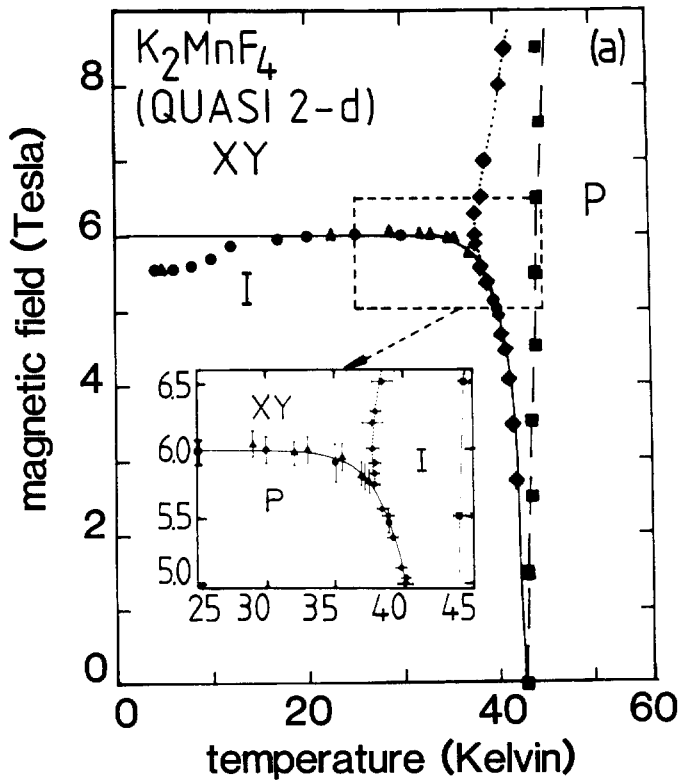


Fig. 2. (a) Phase diagram of chemically pure  $K_2MnF_4$ , which is an example of a quasi 2-d weakly anisotropic antiferromagnet. Data are from Ref. [13] and are for  $B$  parallel ( $\bullet$ ,  $\blacktriangle$ ,  $\blacklozenge$ ) and perpendicular ( $\blacksquare$ ) to the easy  $c$ -axis. The solid and dashed lines represent the theory according to eq. (6). The dotted

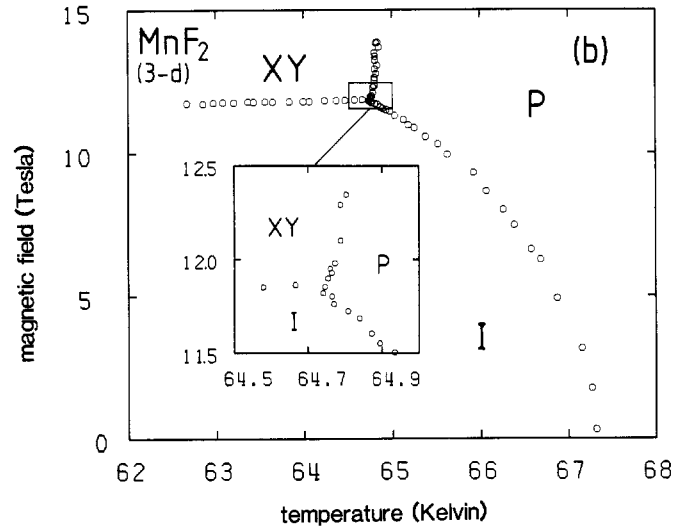
gives the phasediagram of  $MnF_2$  for  $B = B_x$  [14]. Both samples were nominally pure.

For  $K_2MnF_4$  the effective anisotropy is of the uniaxial Ising type for  $B_x < B_{sf}$  and of the XY-type of  $B_x > B_{sf}$  except for the lowest temperatures, where the symmetry should be weakly orthorhombic due to a small distortion of the quadratic layers [13]. In Fig. 2(a) the dotted line is a guide to the eye whereas solid and dashed lines give the predictions for the phase boundaries for  $B = B_y$ , respectively. Here the effective anisotropy model has been applied, yielding the variation of  $D_{eff}^x$  with  $B$  in the expression:

$$T_c(B) \propto [D_{eff}^x(B)]^n. \quad (6)$$

This formula gives the predicted variation of  $T_c$  with anisotropy for a weakly anisotropic 2-d Heisenberg magnet obtained from Monte Carlo calculations [15]. The fit to the experiment gives  $n \approx 0.04$  [13], in good accord with the theoretical value ( $n \approx 0-0.02$ ). The apparent agreement between theory and experiment clearly demonstrates the success of the effective anisotropy model for a specific case. Other excellent examples can be found elsewhere [6], notably for quasi 2-d and quasi 1-d systems.

The spinflop transition corresponds to the horizontal curve in the diagram of Fig. 2(a). It has been obtained from both a.c.-susceptibility and neutron scattering studies. In the neutron experiment the variation of the intensity of a magnetic Bragg reflection with field is followed, yielding the rotation of the staggered magnetization  $m$ . The result of such an experiment, which has been performed at  $T = 4.2$  K, is shown in Fig. 3. It appears that  $m$  rotates gradually as  $B_x$  is increased to a value larger than  $B_{sf}$ , where the total width of the transition is almost one Tesla, i.e., 20% of  $B_{sf}$ ! For subsequent use we define:



curve is a guide to the eye. The bifurcation is shown enlarged in the inset. (b) Phase diagram of the 3-d system  $MnF_2$ , also chemically pure. Data are from Ref. [14] and have been taken in magnetic fields parallel to the easy axis. The bifurcation is shown in detail in the inset.

$$\langle m_p^2 \rangle \equiv \langle m_{\perp}^2 \rangle \quad \text{for } B_x < B_{sf} \quad \text{and}$$

$$\langle m_p^2 \rangle \equiv \langle m_{\parallel}^2 \rangle \quad \text{for } B_x > B_{sf}. \quad (7)$$

Here  $m_{\parallel}$  and  $m_{\perp}$  are the components of  $m$  parallel and perpendicular to the easy axis, respectively. The smeared spinflop detected in the neutron experiment on a 2-d example is widely different from what is observed in the 3-d system. In the latter the total width amounts to  $\approx 10^{-2}$  T and can be fully explained taking into account demagnetizing effects. It is commonly accepted that in the 3-d system the spinflop is first-order. This may be illustrated from Fig. 4, where we show the behaviour of the magnetization in applied fields  $B_x$  for the quasi 2-d system  $K_2MnF_4$  and the 3-d system  $MnF_2$ ,

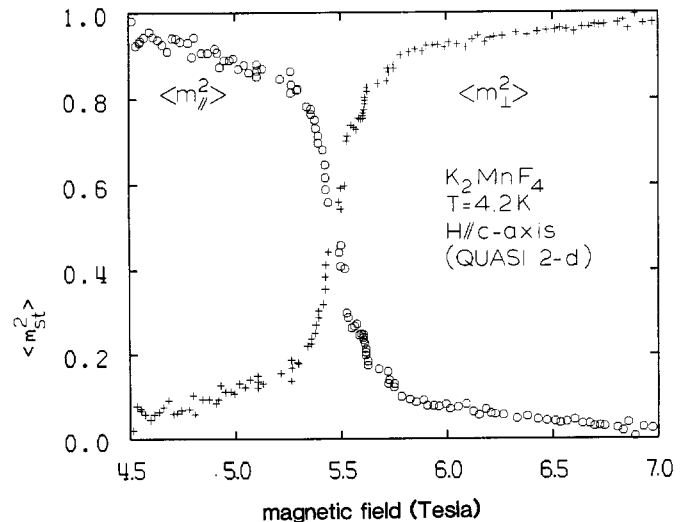


Fig. 3. Rotation of the staggered magnetization in applied fields  $B_x$  for  $K_2MnF_4$  at  $T = 4.2$  K. Circles:  $\langle m_{\parallel}^2 \rangle$ , crosses:  $\langle m_{\perp}^2 \rangle$ .

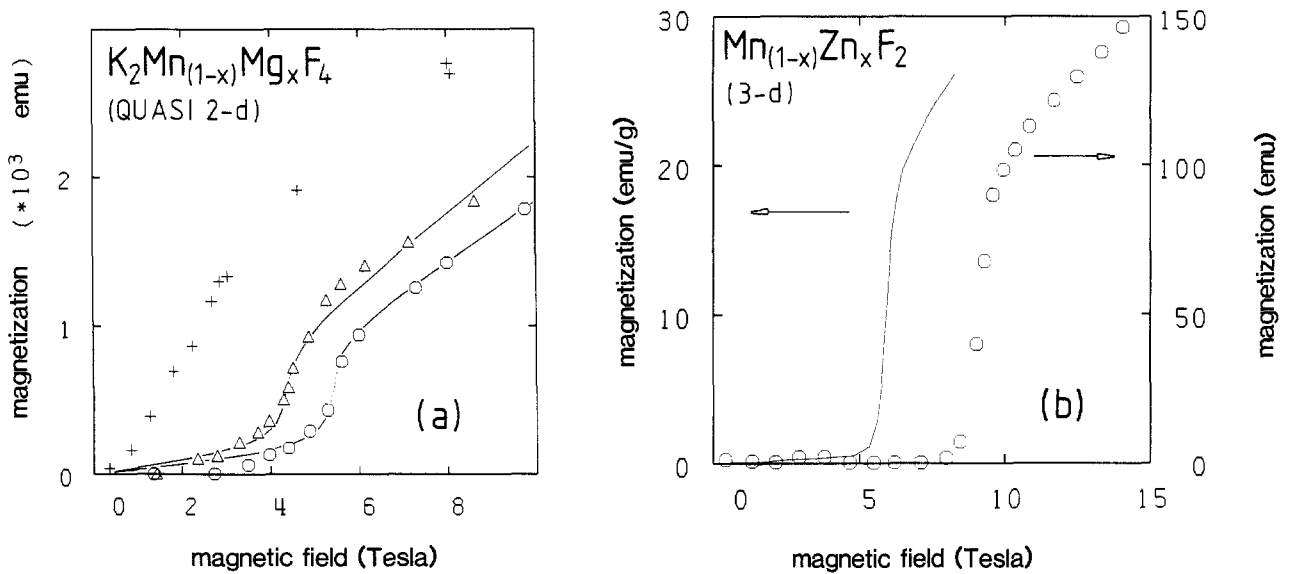


Fig. 4. (a) Spinflop transition in the quasi 2-d-system  $K_2Mn_{(1-x)}Mg_xF_4$ . The magnetization data shown are for the pure compound (O) and for two diluted compounds with  $x = 0.07$  ( $\Delta$ ) and  $x = 0.24$  (+). The experiments have been performed in pulsed fields. Solid curves are fits to eqs. (8) with  $q^{-1} = 10^{-3}$  and with  $\chi_{\perp} = 0.022$  emu/mole and  $\chi_{\perp} = 0.026$  emu/mole for

$x = 0$  and  $x = 0.07$  respectively. (Data taken from Refs. [16]). (b) Spin-flop transitions in the 3-d-system  $Mn_{(1-x)}Zn_xF_2$ , pure (O) and with 25% Zn (solid line). Data are from Refs. [17, 18]. Note the absence of broadening, also for the  $x = 0.25$  compound.

both pure and with nonmagnetic impurities (Mg, Zn). The data are from Refs. [16–18]. The results for the pure compounds are given by the open circles. As regards the width of the transition the difference is quite striking. More examples of sharp spinflops in 3-d systems and broadened spinflops in quasi 2-d systems and also in quasi 1-d systems may be found in the literature [6, 19].

To our opinion the broadening of the spinflop in low-d systems has to be attributed to the presence of domainwalls. In order to explain this we go back to Fig. 1. In the antiferromagnetic domainwall the moments rotate through an angle  $\pi$  on both sublattices. The total width is approximately  $\pi d_s$ . For  $B_x < B_{sf}$  the domainwall may be considered as a region of the flopped phase in the low-field phase [cf. Fig. 1(a)], since inside the wall the average polarization of the individual moments is tilted with respect to the easy ( $x$ -axis) and thus will give rise to perpendicular components  $m_{\perp}$ . Along the same lines one may argue that for  $B_x > B_{sf}$  the domainwall is in fact a region of the low-field phase in the flopped phase [cf. Fig. 1(b)]. As mentioned before there may be two reasons why the walls show up in the experiments for  $B_x \rightarrow B_{sf}$ , namely firstly via the increase of  $d_s$ , and secondly, in case the relaxation of the system is sufficiently fast with respect to the characteristic time of the experiment, due to the increase in the wall density  $q^{-1}$ . In the neutron experiment of Fig. 3 equilibrium could be obtained, although the relaxation took several minutes.

Also in Fig. 4 we show the magnetization curves of some  $K_2Mn_{(1-x)}Mg_xF_4$  [16] and  $Mn_{(1-x)}Zn_xF_2$  [17, 18] compounds. The results for the  $K_2Mn_{(1-x)}Mg_xF_4$  samples have been obtained in pulsed fields where the pulse duration is within  $\approx 0.1$  s. In these diluted compounds the external field will give rise to a random staggered field [11] so that eq. (5) will depend on the applied field through both the first and the second term. However, during such a fast experiment the wall density can not be expected to change since the relaxation times are much longer than the pulse duration. Nevertheless, the effect of the increasing  $d_s$  alone is apparently sufficient to cause a

substantial broadening of the spinflop even in the chemically pure system! The solid curves in Fig. 4(a) are fits to the theoretical prediction [6] for the magnetization as given by

$$\begin{aligned} M &= M_{\text{sol}} & \text{for } B_x < B_{\text{sf}} & \text{ and} \\ M &= \chi_{\perp} B_x - M_{\text{sol}} & \text{for } B_x > B_{\text{sf}}, \end{aligned} \quad (8)$$

with

$$M_{\text{sol}} = 8S^2 \chi_{\perp} B_x \left[ \frac{2Jd_s}{\pi k_B T} \right]^{1/2} q^{-1} \quad (9)$$

the contribution of the static domainwalls to the magnetization [20]. Indeed, from these fits the wall density in the impure system is found to be the same as in the pure one.

In an attempt to analyze the neutron data of Fig. 3 one may compare the experimental results with theoretical predictions for the weakly anisotropic system in a random field. Here we use the close correspondence [6] with the commensurate-incommensurate (CI)-transition in a noble gas monolayer on a solid surface, that has been investigated extensively during the past few years [21]. Both phenomena are described by the same continuum model, the SG-system. In the CI-problem the chemical potential  $\mu$  for walls is variable. For domainwalls in a weakly anisotropic antiferromagnet one has  $\mu = 0$  since a finite  $\mu$  would be a "twist field" that cannot be applied in an experiment.

The influence of random fields on the CI-problem has been investigated a.o. by Villain *et al.* [22]. They considered a 2-d modified Ising-type system with a preferential direction for walls. In such a system the walls will be long and almost straight. Although in this work the isotropic case is considered we shall nevertheless adopt their model since as far as we know it is the best approximation available. Villain *et al.* found that in the low-temperature phase the system has a considerable amount of order but is *not* long-range ordered. The wall-density is low and for  $\mu = 0$  it may be expressed as

$$q^{-1} \propto \exp \{ -\gamma(\sigma/\sigma_0)^{4/3} \}, \quad (10)$$

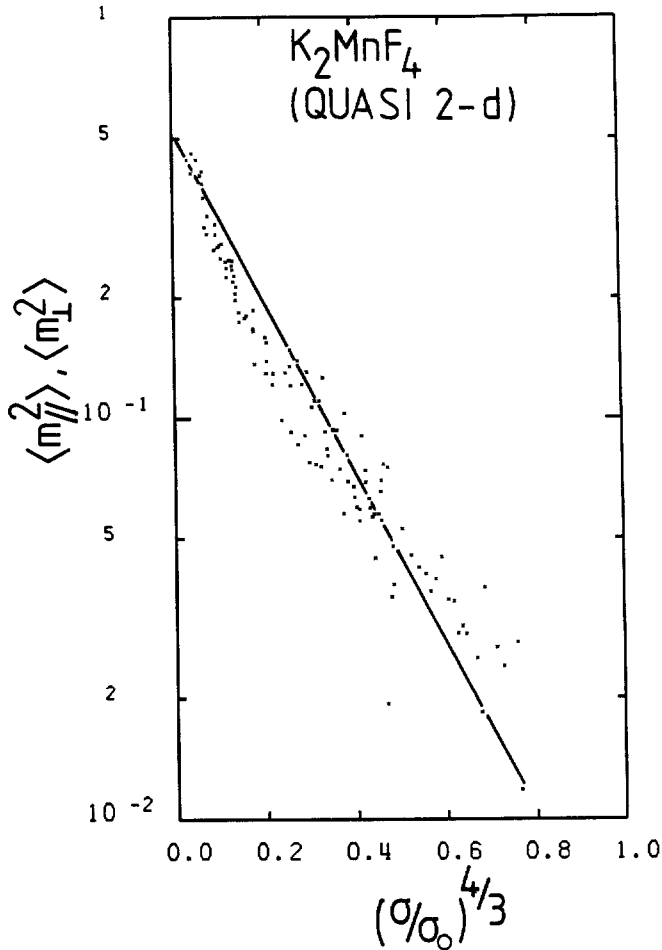


Fig. 5. Semi-logarithmic plot of  $\langle m_p^2 \rangle$  vs.  $(\sigma/\sigma_0)^{4/3}$ . The solid line is the fit to the theory given by eq. (10).

with

$$\sigma_0 = 4S^2\sqrt{|D_x J|} \quad \text{and} \quad (11)$$

$$\gamma = 2(\sigma_0/b_r)^{4/3}. \quad (12)$$

Here  $b_r = g\mu_B B_r/k_B$ . Furthermore one has  $\langle m_p^2 \rangle \propto q^{-1}$ , yielding an exponential law for  $\langle m_p^2 \rangle$ . In Fig. 5 the comparison between the neutron data of Fig. 3 and this theory is shown. From the slope of the curve we obtain  $b_r = 8$  K, which is consistent with a random field strength  $b_0 = b_r/\sqrt{c}$  at the defects of about 500 K, assuming a defect concentration  $c \approx 10^{-3}$ – $10^{-4}$  per l.u. The value  $b_0 \approx 500$  K seems reasonable since it is of the same order as the droplet creation energy [6, 21] in the 2-d SG-approximation,  $4\pi JS^2/k_B \approx 460$  K.

Thus it appears that the intensity variation of the magnetic Bragg peak may be well-explained in terms of domainwalls. Of particular interest here is that the relaxation effects were indeed observed during the neutron measurements.

As far as experiments on 3-d examples is concerned, the attention is focussed mainly on  $Mn_{(1-x)}Zn_xF_2$  [3] and  $Fe_xZn_{(1-x)}F_2$  [2]. In the Mn-compounds the anisotropy is of dipolar origin and is therefore, weak, as in  $K_2MnF_4$ . The  $x = 0.25$  and  $x = 0.35$  Mn-compounds have been extensively studied [23]. Despite the fact that here compounds with  $d > d_{lc}$  are concerned a domain state appears to be thermodynamically stable if the samples are cooled in a field that generates the random field according to the Fishman and Aharony device. One may argue that for these diluted systems the random field approach is too restricted to yield a full

explanation of the experimental observations. Recent mean field computer simulations [24] of diluted 3-d weakly anisotropic antiferromagnets that should provide a better theoretical description than the random field model, indeed yield such a stable domain phase between the paramagnetic and the Ising ordered phase.

In fact it is not so surprising that the random-field model may only partly explain the field-induced phenomena for these diluted systems. The nonmagnetic impurities affect the system in various other ways, e.g., there will be random exchange effects and random anisotropy effects. The former occurs since part of the nearest-neighbour exchange-bonds are removed by the dilution. The latter occurs in case the anisotropy is due to the dipolar interactions between the magnetic moments, which is typically a long-range effect. In diluted systems these dipolar interactions are then non-uniform. Both effects, i.e. the influences on the exchange and on the anisotropy, have important implications for the experimental behaviour. Obviously, for very high impurity concentrations one ultimately reaches the percolation threshold, corresponding to the situation that  $T_c = 0$  and  $D_x = 0$ , but even for moderate concentrations of non-magnetic ions the reduction of  $T_c$  and  $D_x$  are well-observable [25]. Furthermore it is well-known that the phase diagram is quite complex, with many different phases. (This is also found in the computer-simulations of Ref. [24]). Thus it appears that already for moderate impurity concentrations a random-field description, which is essentially a perturbation approach, is too restricted. On the other hand, if one wants to investigate random field effects in magnetic systems, the strategy outlined in the present paper, i.e., variation of the energy-balance through the first term in eq. (5), may be useful, since it should be applicable to chemically pure systems. In that case  $B_r$  is weak and may indeed be considered as a perturbation.

Furthermore, in the experimental literature the influence of the applied field on the intrinsic properties of the domainwalls has not been taken into account. Also in the 3-d case one should consider field-induced anisotropy effects. Considering the amount of dilution one would expect that for low  $B_x$  the effect of the field-dependence of  $B_r$  will be much more important than the effect of the field-induced variation of  $\sigma$ . On the other hand, one has  $\sigma \rightarrow 0$  for  $B_x \rightarrow B_{sf}$ , so that in this limit the field-induced variation of  $\sigma$  is the dominant effect. This might explain why the correlation lengths  $\xi$  observed by Cowley *et al.* [23] in samples of  $Mn_{0.65}Zn_{0.35}F_2$  cooled in fields between 4 T and  $B_{sf}$ , are significantly lower than in zero field. At low temperatures and for  $B_x = 4$  T they find  $\xi^{-1} \approx 0.0003$  and  $\sigma \approx 0.6\sigma_0$ , whereas for  $B_x = 4.8$  T one has  $\xi^{-1} \approx 0.0005$  and  $\sigma \approx 0.3\sigma_0$ , i.e., a decrease of  $\sigma$  implies an increase in the wall-density, which is fully comparable to what is observed for systems with  $d < d_{lc}$ .

Since the walls are broad ( $\pi d_w \approx 11$  l.u. for  $B_x = 0$ ) relaxation is possible, as in  $K_2MnF_4$ , and has been observed indeed in both the real experiments [3, 23] and in the computer simulations [24]. Some differences between the two are still unresolved, namely: in both cases relaxation towards the Ising ordered phase is observed as the field is removed, but in the simulations the domain-state is restored upon subsequent increase of  $B_x$  whereas in the experiments the ordered state is stable since the domain state cannot be obtained starting from  $B_x = 0$ . The latter is in agreement with the spinflop experiment [18] on the  $x = 0.25$  compound shown in

Fig. 3(b). The transition is as narrow as in the pure system, indicating the complete absence of domainwalls.

Also in  $\text{Fe}_{(1-x)}\text{Zn}_x\text{F}_2$  a domain state is observed upon field cooling [26]. Here the anisotropy is due to crystal field effects and is quite strong, which is reflected in the much higher value for  $B_{sf} \approx 40\text{ T}$  [27]. Several diluted compounds have been investigated, for  $B_x$  up to 5 T [2]. Since  $B_x \ll B_{sf}$  the field-induced effects on  $\sigma$  and  $d_s$  are less than 1%, in strong contrast to what happens in the Mn counterpart. Consequently the walls are very narrow,  $\pi d_s \approx 11\text{ u.}$  and for  $T \ll T_c$  they should be strongly pinned by defects and impurities in the field region. This is in agreement with the experimental observations [26]. It has been reported that the domain state does *not* transform into the Ising ordered state if the field is removed at low temperatures,  $T \ll T_c(B_x)$ . On the other hand it has been reported that for  $T \approx T_c(B_x)$  the system behaves similar as  $\text{Mn}_{(1-x)}\text{Zn}_x\text{F}_2$ , i.e., relaxation towards the ordered state upon a field decrease and preservation of the ordered state upon increasing the field. The reason may be that for  $T \approx T_c(B_x)$  there will be an effective decrease of  $\sigma$  due to thermal fluctuations. The latter effect occurs in any Ising-type system around  $T_c$  and is part of the critical behaviour. Within the context of the present work one might state that variation of the temperature in the critical region may be viewed as causing an effective variation of the first term in eq. (5).

Finally we would like to discuss briefly the behaviour observed for  $B_x \approx B_{sf}$  at  $T \approx T_c(B_x)$ . In this region both contributions to the decrease of  $\sigma$  are important, i.e. that due to field-induced effects and that due to thermal fluctuations. Moreover the associated increase of  $d_s$  may have its consequences. Going along the phase line  $T_c(B_x)$  a crossover from metastability in low fields (narrow walls) towards thermal wall meandering for  $B_x \approx B_{sf}$  should take place.

It is of interest to see how these phenomena influence the phase diagrams of the weakly anisotropic systems. Compari-

son of the inserts in Figs. 2(a) and 2(b) shows the difference in behaviour. In the quasi 2-d system the spinflop line and the XY-P and I-P transition bifurcate from the spinflop line. The latter is considered to be the normal situation since it also occurs for quasi 1-d systems and since it has been explained by various theories. However, the anomalous type of bifurcation has also been found in the diluted 3-d  $\text{Mn}_{(1-x)}\text{Zn}_x\text{F}_2$  systems, where this behaviour may be a random-field effect [18].

For the 2-d system this is corroborated by the theoretical work on the CI problem. In Fig. 6(a) we show (schematically) the phase diagram of the pure 2-d weakly anisotropic Ising system [28]. Here  $\mu$  is the "twist-field" or chemical potential for walls and  $\mu_c \propto \sigma$ . There are three phases. The commensurate or ordered phase (C), the disordered phase (D) and the incommensurate (I) phase where there are long meandering domainwalls. At  $T = 0$  and for  $\mu \downarrow \mu_c$  the wall density vanishes so that for  $\mu < \mu_c$  the system is ordered (C-phase). In Fig. 6(b) we give a schematic sketch of what this phase diagram might look like if a random field becomes involved [6]. In that case the wall density decreases but does not completely vanish for  $\mu \downarrow \mu_c$  and the low- $T$ /low- $\mu$  phase should be pseudo-commensurate (PC), that is with only a few walls which are almost static. The IC-D phaseline should join the PC-D line at some point  $(T_2, \mu_2)$ .

In the quasi 2-d magnetic modelsystems discussed here one has  $\mu = 0$  and  $\mu_c \downarrow 0$  for  $B_x \rightarrow B_{sf}$ . However, the  $B_x$  that is present may be considered as an effective chemical potential for walls,  $\mu_r$ , which is almost field-independent for a chemically pure system. If  $B_x \rightarrow B_{sf}$  one has  $\mu_c \downarrow 0$  and  $\mu_r \downarrow 0$ . Hence for  $B_x \approx B_{sf}$  there should be a phase with thermally meandering walls between the ordered phase and the paramagnetic phase. Furthermore the bifurcation may correspond to a Lifshitz-type point.

Note that the IC-D transition is a Kosterlitz-Thouless transition whereas both the IC-PC and the D-PC transitions

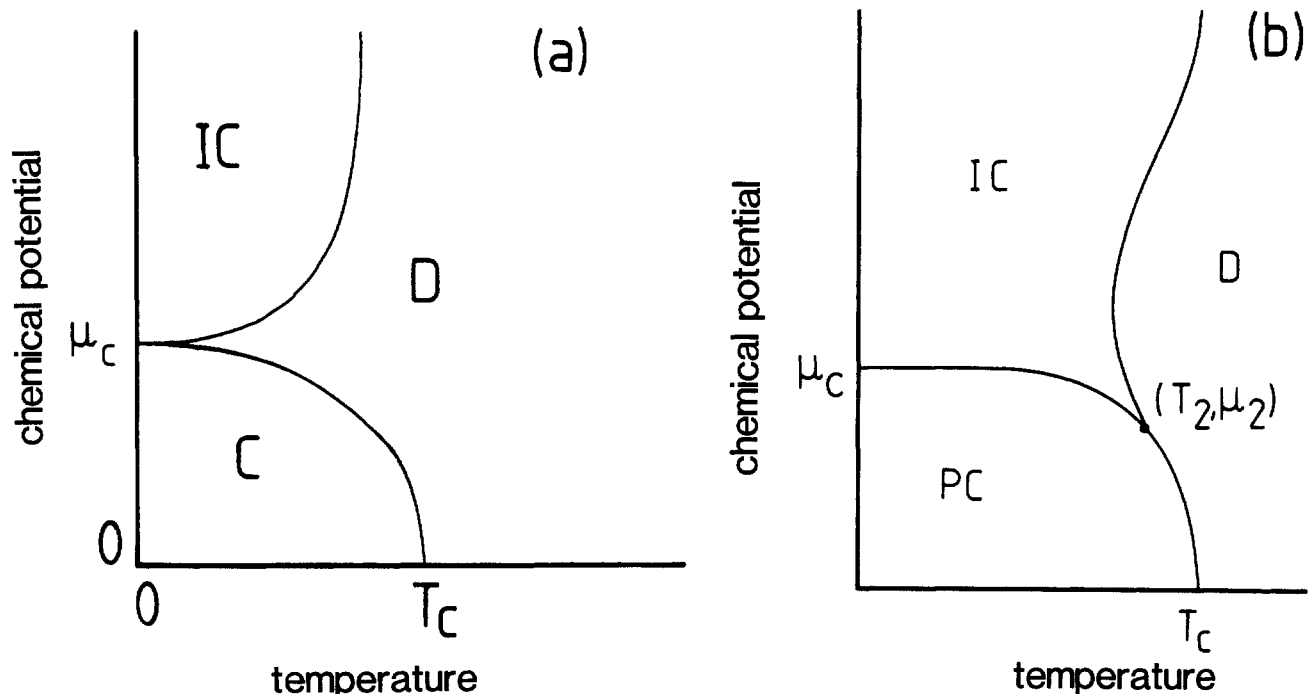


Fig. 6. (a) Phase diagram ( $\mu$ - $T$ ) for the 2-d-SG-system in a "twist-field"  $\mu$  (schematic). The different phases are: disordered (D), ordered commensurate (C) and floating incommensurate (IC). Only in the IC-phase there are

domainwalls in the system. (b) Proposed phase diagram ( $\mu$ - $T$ ) for the 2-d-SG-system in an effective "twist-field"  $\mu_r$  due to a random field  $B_r$ . In the PC-phase the system contains (nearly) frozen walls.



are Ising transitions. This follows from the various renormalization approaches to the CI-problem [21, 22, 28–30], where it appears that Ising-type anisotropies are strongly relevant [31]. This is in agreement with the experimentally found phase diagram of  $K_2MnF_4$  [cf. Fig. 2(a)]. Here the whole boundary of the (pseudo)-ordered phase, including the spinflop line, is described by the Monte-Carlo prediction for systems with weak Ising-type anisotropy (cf. eq. (6)).

Obviously, one of the problems with the above explanation is that it relies heavily on the 2-d nature of the system, whereas experimentally the peculiar bifurcation appears to be also seen in 3-d diluted  $MnF_2$ . It may therefore well be that the explanation should be sought elsewhere, and would apply to the 3-d as well as to the quasi 2-d case. In this respect it is worthwhile to recall that the first-order spinflop line and the bi-critical point in which it ends are extremely sensitive to “hidden” variables in the system. For the spinflop transition both molecular field theory and spin wave-theory [32] predict supercooling and superheating effects, the value of the spinflop field being substantially different for increasing and decreasing applied field. Although these first-order effects have not been observed in pure 3-d antiferromagnets, they might well be triggered by the slightest amount of impurity and give rise to domains of flopped and unflopped phases. As regards the bicritical point, Landau-type free-energy expansions already show that its occurrence should be very sensitive to the presence of other, weak thermodynamic besides the field and the temperature.

In conclusion one may state that within the context of the effective field-dependent anisotropy model, weakly anisotropic systems offer quite promising novel possibilities for the study of random-field and metastability behaviour, possibilities that have so far not been considered. The experimental data discussed above strongly support the various theoretical predictions from the model and yield convincing evidence that the intrinsic properties of the domainwalls,  $\sigma$  and  $d_s$ , are indeed important for a complete description of the various interesting phenomena that are observed in the experiments. We strongly advocate more effort along these lines, theoretically as well as experimentally.

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