Phase diagrams of weakly anisotropic Heisenberg antiferromagnets: III. Nonlinear excitations and random fields in quasi 2-dimensional systems

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PHASE DIAGRAMS OF WEAKLY ANISOTROPIC HEISENBERG ANTIFERROMAGNETS:III. NONLINEAR EXCITATIONS AND RANDOM FIELDS IN QUASI 2-DIMENSIONAL SYSTEMS.

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The concept of effective field-dependent anisotropy is applied to the "spinflop" transition in the quasi 2-d Heisenberg antiferromagnet with weak orthorhombic anisotropy. From the correspondence between the "spinflop" problem and the commensurate-incommensurate transition it follows that the "spinflop" is not first order and that random fields may cause domain-wall formation. This would explain the observed broadening of the "spinflop" in K_2MnF_4. In 3-d antiferromagnets such anomalous broadening is not observed, which would agree with the critical dimensionality $d_c = 2$ for the random-field problem.

In two previous papers [1,2] (I and II) the field-induced phase transitions occurring in respectively the quasi one-dimensional (1-d) and quasi two-dimensional (2-d) Heisenberg antiferromagnets with weak anisotropies have been discussed using the concept of effective field-dependent anisotropy. For both cases theoretical predictions for the magnetic phase diagram were deduced and compared with experimental data available in the literature. Furthermore it was shown in paper I that the physical mechanisms underlying the field-induced behaviour in the quasi 1-d system are quite different from those in the 3-d case. In particular the "spinflop" phenomenon obtains a different meaning and does not correspond with a first order transition. It was argued that at finite temperatures it may even be a continuous (perhaps infinite-order) transition, in a lattice of antiferromagnetic soliton-pair-states.

Encouraged by these results we proceeded to consider in more detail the "spinflop" transition in the quasi 2-d system as well. In the present work we apply the same field-dependent anisotropy concept to this problem and we point out the close relationships with other topics in 2-d physics, as there are the commensurate-incommensurate (C-I) transition [3], the Kosterlitz-Thouless (K-T) transition [4] and the random field or random anisotropy problem [5].

When applied to the 2-d Heisenberg antiferromagnet with weak orthorhombic anisotropy, the effective anisotropy model yields the hamiltonian:

$$\mathcal{H} = -2J \sum_{\langle i,j \rangle} \vec{S}_i \vec{S}_j - \sum_k \left(D_{eff} S_{kx}^2 - D_z S_{kz}^2 \right) \qquad (1)$$

with $J<0$ and $D_z>0$. Here D_{eff} is the effective anisotropy as given by the equation:

$$D_{eff} = D_x \left(1 - H_x^2 / H_{sf}^2 \right) . \qquad (2)$$

As in papers I and II, D_x is the anisotropy in zero field favouring the X-axis ($D_x>0$), and $H_{sf} = 4S\,|2zD_xJ|^{1/2}/g\mu_B$ is the "spinflop" field. D_{eff} arises from the competition between the anisotropy energy $D_xS(S+1)$ and the Zeeman energy $\frac{1}{2}\chi_\perp H_x^2$ associated with a field parallel to the easy axis. Application of the effective anisotropy model to a hamiltonian which contains an easy-axis term $D_xS_x^2$ together with a field term $g\mu_B H_x S_x$ amounts to replacing D_x by D_{eff} and skipping the field term, which results in hamiltonian (1). The first term represents the exchange interaction, which is taken to be isotropic, and the last term establishes an easy (XY) plane. The symmetry in this plane is broken by the anisotropy D_{eff}, that favours either the X-axis ($D_{eff}>0$) or the Y-axis ($D_{eff}<0$).

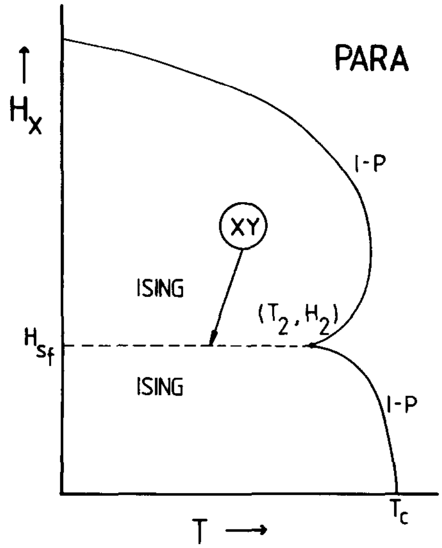

Fig. 1 Phase diagram (schematic) of the 2-d antiferromagnet with orthorhombic anisotropy for fields H_x along the easy axis.

The phase diagram for $H = H_x$ for a such a system was discussed in paper II and is sketched in fig. 1. For $H_x < H_{sf}$ the anisotropy D_{eff} is of the Ising type and the phase line (I-P) corresponds to transitions from the 2-d long-range uniaxially ordered Ising state towards the paramagnetic phase. For $H_x = H_{sf}$ one has $D_{eff} = 0$ and the anisotropy is of the XY-type due to the presence of the D_z term in eq.(1). Hence the (dashed) "spinflop" line should be a line of continuous phase transitions ending in the bicritical point (T_2,H_2). The latter should correspond to a Kosterlitz-Thouless transition, i.e. a transition from a state of topological order towards the paramagnetic state. For $H_x > H_{sf}$ the system is also Ising, as for $H_x < H_{sf}$, but with the Y-axis as the easy axis since the sign of D_{eff} is negative in this case and the X-axis becomes the intermediate axis. When H_x is further increased, one passes into a situation where $D_{eff} > D_z$, and a switching [6] of the hard axis occurs. For the present work, however, this will not be considered since we shall concentrate on the phenomena observed for fields H_x close to H_{sf}.

In paper II the field-dependent anisotropy concept was applied to the phase diagrams of the compounds $Mn(HCOO)_2.2H_2O$ and K_2MnF_4. From the good agreement between theory and the experiments it appeared that the effective anisotropy concept is a powerful tool for understanding the field-induced transitions in the weakly anisotropic 2-d antiferromagnet. Therefore it is of interest to investigate the extent to which it can be used to explain the physical phenomena leading to the "spinflop" transition. The latter again turns out to be quite different from the conventional spinflop in the 3-d antiferromagnet. In fig. 2 we reanalyse neutron scattering data [7] for K_2MnF_4 that demonstrate this. In the experiment in ref.7 the intensity I of the magnetic Bragg-peak at the reciprocal lattice vector $(h,k,l) = (3/2,1/2,0)$ was followed at $T = 4.2$ K for magnetic fields H_x between 45 kOe and 70 kOe. It is clear from fig. 3 of ref. 7 that the average orientation of the magnetic moment changes from mainly perpendicular to (3/2,1/2,0) at $H_x = 45$ kOe to mainly parallel to (3/2,1/2,0) at $H_x = 70$ kOe, corresponding to a spinflop-type transition at $H_{sf} \approx 55$ kOe. At low temperatures and for $H_x > H_{sf}$ the symmetry of this compound is effectively orthorhombic [7] since an antiferromagnetic spin component within the basal plane will induce a distortion of the tetragonal lattice, as can be understood from magnetic-grouptheoretical arguments. Then the system is decribed by the effective spinhamiltonian (1) with small D_z. From the relation:

$$I(3/2,1/2,0) \propto \langle m_\parallel^2\rangle\langle\sin^2\theta_\parallel\rangle + \langle m_\perp^2\rangle\langle\sin^2\theta_\perp\rangle \quad (3)$$

where $m_\perp$ and $m_\parallel$ are the components of the normalized staggered magnetization parallel and perpendicular to the c-axis, and $\theta_\perp$, $\theta_\parallel$ the angles they make with the (3/2,1/2,0) direction, the field dependence of $\langle m_\perp^2\rangle$ and $\langle m_\parallel^2\rangle$ can be deduced. The neutron beam in the experiment was perpendicular to the c-axis, so $\theta_\parallel = 90°$. Now for $H_x \ll H_{sf}$ one may safely state $\langle m_\perp^2\rangle = 0$ and $\langle m_\parallel^2\rangle = 1$ with $I(3/2,1/2,0) = I_0$. For $H_x \gg H_{sf}$ on the other hand, $\langle m_\parallel^2\rangle = 0$ and $\langle m_\perp^2\rangle = 1$ so $I(3/2,1/2,0) = I_0\langle\sin^2\theta_\perp\rangle$. If the symmetry of the high-field phase were of the planar type there would be no preferential value for $\theta_\perp$ so one would have $\langle\sin^2\theta_\perp\rangle = \frac{1}{2}$. This would yield $I(3/2,1/2,0) \simeq \frac{1}{2}I_0$ for $H_x \gg H_{sf}$, which is half the intensity in low field. This clearly disagrees with fig. 3 of ref.7, where one has $I(H_x \gg H_{sf}) \approx 0.2\ I_0$, so it follows that the quadratic symmetry is indeed not retained for $H_x > H_{sf}$. When the lattice transforms into a lozenge, the moments can be either along the (-1,1,0) direction, with $\langle\sin^2\theta_\perp\rangle = 0.8$, or along the (1,1,0) direction with $\langle\sin^2\theta_\perp\rangle = 0.2$. The latter corresponds to the experimental observation and thus we will assume below that in the "flopped" phase the moments are along the (1,1,0) direction. The reason why the crystal becomes a single domain can be e.g. a small misorientation of the sample. This is of course inevitable and will result in a small perpendicular component of the applied field that will favour one of the two possible transformations. On basis of the above discussion we show in fig. 2 the field-dependence of $\langle m_\perp^2\rangle$ and $\langle m_\parallel^2\rangle$, according to eq.(3) with $\langle\sin^2\theta_\perp\rangle = 0.2$. It is seen that the transition is 10 kOe broad, much broader indeed than could possibly be explained by demagnetizing effects or sample misorientation, since those would cause a broadening of at most 1 kOe. Similar widely smeared "spinflop" transitions have been observed [8] in other 2-d antiferromagnets, as in K_2NiF_4 and Rb_2NiF_4.

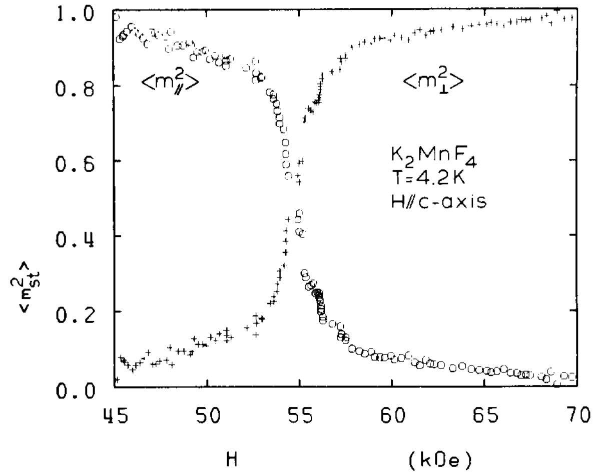

Fig. 2 Rotation of the staggered magnetization m^2 versus an applied field H∥c-axis at $T = 4.2$ K in K_2MnF_4. Circles: $\langle m_\parallel^2\rangle$, crosses $\langle m_\perp^2\rangle$.

Since the broadening is much too large to be due to demagnetizing effects or misorientation, another process must be involved. It was argued in paper I that in the quasi 1-d antiferromagnet, where comparable broadening effects are observed, the thermal excitation of small 1-d droplets accounts well for the observed broadening. These may be considered as bound states of a soliton and an antisoliton (domain-walls) in the 1-d magnet. The idea behind it is that inside a wall the spins have components perpendicular to the preferential axis and therefore a wall corresponds to a fragment of the flopped phase in the low-field phase for $H_x < H_{sf}$ and to a fragment of the low-field phase in the flopped phase for $H_x > H_{sf}$. The question now arises whether walls may also exist in the quasi 2-d antiferromagnet, either in the form of small droplets or as large meandering domain-walls, and yield similar effects as in the 1-d case.

First we shall consider the model of excitation of small droplets, in view of its success in the chainlike systems. It has been pointed out that weakly localized magnon bound states or vortices may exist in the 2-d antiferromagnet with weak Ising anisotropy [9]. They will possess a certain stability provided they contain sufficient magnons, due to the fact that the radius of the droplet must be larger than the wall thickness $d = |J/D_{eff}|^{\frac{1}{2}}$ of an isolated domain-wall. The creation energy of a droplet equals $E_d = E_v + E_w$ with $E_v = 8\pi|J|S^2Q$ for the vortex part and $E_w = 4S^2|D_{eff}J|^{\frac{1}{2}}.\ell$ for the wall part. Here Q is the topological charge of the vortex (i.e. the amount of vorticity) and ℓ is the perimeter of the droplet. For K_2MnF_4 one then calculates $E_d \approx 650$ K as the minimum value for E_d, that is for $Q = 0$ and $\ell = 2\pi d$. This value does not depend on D_{eff} since $d = |J/D_{eff}|^{\frac{1}{2}}$. We may conclude that the probability of such a vortex excitation will be very low at T = 4.2 K, which is the temperature at which the neutron diffraction experiment of fig.2 was performed. Apparently, the analogue of the 1-d soliton pair-state invoked in paper I to explain the spinflop in chain systems can not be responsible for the observed broadening in the 2-d case.

As regards the second possibility of large meandering domain-walls, the situation is more complex. In the 2-d magnetic layer the walls will form line-patterns and, besides the wall-energy which depends on the length ℓ of the wall as in the above, also the entropy arising from the wall pattern may give an important contribution to the free energy of the system. If one considers a large, closed domain-wall (i.e. a droplet with $\ell >> 1$), this can be seen as a self-avoiding closed loop. For large ℓ the number of realisations is given by $n(\ell) = \mu^{\ell}\ell^{-h}$, with $\mu = 2.6395$ and $h = 3/2$ for the square lattice [10]. The contribution of such a vortex to the free energy is estimated as
$\Delta F = (4S^2|D_{eff}J|^{\frac{1}{2}} - T\ell n(\mu))\ell + Th\ell n(\ell)$. Since for sufficiently large ℓ one has $\ell >> \ell n(\ell)$ the last term can be neglected. Dissociation of the vortex occurs for $4S^2|D_{eff}J|^{\frac{1}{2}} < \ell n(\mu)$, because then ΔF will become negative [4]. Although this would be an interesting type of Kosterlitz-Thouless transition, it is not of help for the present problem. A calculation for K_2MnF_4 shows that it would happen for fields $0.97 < H_x/H_{sf} < 1$ whereas the observed broadening already starts at $H_x/H_{sf} \approx 0.8$.

Recently, it has been shown that notwithstanding the above considerations 2-d systems may become unstable with respect to domain-wall formation [5,11] in the presence of (infinitesimal) random fields or random anisotropies. Also it was suggested by Imry and Wortis [12] that first order transitions are smeared in the presence of random fields for systems below the lower critical dimensionality. Now there are two possible sources of random fields in the present case. First, in the solid state there will always be a small amount of impurities or lattice defects present (typical concentration $c \lesssim 10^{-3}$) which may give rise to random field effects. Second, in case of partial flopping of regions of spins, the interplane interactions may act as a random field too. Also random anisotropy effects will be present since the anisotropy is of dipolar origin so that the presence of excitations will cause variations in the anisotropy at the same time. Although we cannot calculate the strength of the random field, it is possible to estimate its order of magnitude and we shall show that it may indeed explain the observed broadening of the "spinflop".

The "spinflop" phenomenon in the quasi 2-d antiferromagnet is in fact closely related to the C-I transition in a 2-d Ising-type system (see fig. 3), which has been extensively studied in the last few years [3]. This can be seen as follows. In the C-I problem one considers a chemical potential δ for the domain-wall. This δ can be varied in the experiment e.g. by changing the pressure. For δ smaller than a critical value δ_c the system is in the commensurate (ordered) state. For $\delta > \delta_c$ the system enters the

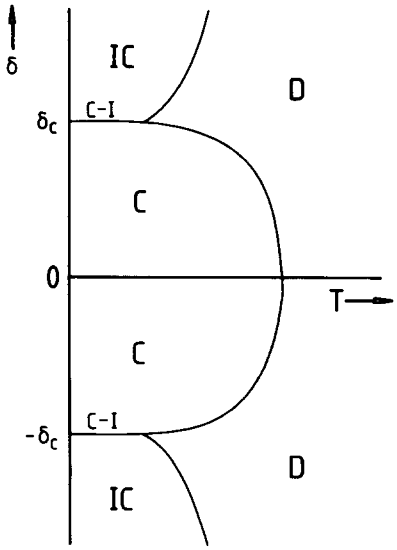

Fig. 3 Phase diagram (schematic) of the 2-d system with a twist field δ. The layer can be in the commensurate (C), the incommensurate (IC) or the disordered (D) state. The lines C-I indicate the commensurate-incommensurate transitions.

incommensurate state, which consists of large ordered regions, separated by incommensurate domain-walls. In the 2-d antiferromagnet δ would correspond to a "winding field", which cannot be physically realized and therefore one has $\delta = 0$ always. Usually a description [13,14] of the C-I transition is given in terms of 1-d quantum-field models, such as the Quantum Sine-Gordon and the Massive Thirring model via the transfer matrix. When applied to the spinflop problem it follows that $\delta_c \approx 4S^2|D_{eff}J|^{\frac{1}{2}}$. So, in some sense the "spinflop" in the 2-d antiferromagnet is the magnetic analogue of the C-I transition. Whereas in the C-I problem the quantity δ is variable and is increased towards δ_c, the "spinflop" problem corresponds to a situation where δ_c approaches zero, since $D_{eff} \to 0$ for $H_x \to H_{sf}$. From this correspondence it follows that the "spinflop" cannot be first order [15]. Lately, it has been argued that random fields may entirely suppress the commensurate (ordered) phase [16], although for $\delta \ll \delta_c$ only few walls are present and the system is almost completely ordered. In the model of Villain et al. the free energy was found to be given by the expression [16]:

$$\frac{F}{N} = q^{-1}\{\delta_c + 4\delta_c \exp(-q/d) - \tfrac{1}{2}H_r(H_r/g)^{1/3}\ln(q)\} \quad (4)$$

Here q is the average distance between long, almost straight domain-walls (which cross the system), and H_r is the rms random field $H_r = H_0\sqrt{c}$, with H_0 the average strength of the impurities. The parameter g equals δ_c for the present case. When only few domain-walls are present ($q \gg 1$) one may omit the exponential term in eq.(4), which is the contribution arising from the interaction between the walls. Minimalization of F with respect to q then yields:

$$q^{-1} \propto \exp\{-\gamma\,|1-H^2/H_{sf}^2|^{2/3}\} \quad (5)$$

(with $\gamma = 2(4S^2/H_r)^{4/3}(D_x J)^{2/3}$). For a Sine-Gordon domain-wall one may calculate (in an analogous way as was done in paper I to obtain eq.(5)) the average spin-components perpendicular to the preferential direction:

$$\left.\begin{matrix}\langle m_\perp^2\rangle(H_x < H_{sf}) \\ \langle m_\parallel^2\rangle(H_x > H_{sf})\end{matrix}\right\} = \frac{1}{q}\int_{-\frac{1}{2}q}^{\frac{1}{2}q} \sin^2\{\tan^{-1}\exp(\tfrac{z}{d})\}dz \approx \frac{2d}{q} \quad (6)$$

for $d \ll q$. From the two equations (5) and (6) it follows that $\langle m_\perp^2\rangle(H < H_{sf})$ and $\langle m_\parallel^2\rangle(H > H_{sf})$ should be proportional to $\exp(-\gamma|1-H^2/H_{sf}^2|^{2/3})$. This is of considerable interest since it provides a direct comparison of the theory with an experiment. Thus we show in fig. 4 a log-plot of the experimental values for $\langle m_\perp^2\rangle$ and $\langle m_\parallel^2\rangle$ versus $|1-H^2/H_{sf}^2|^{2/3}$. Clearly an exponential law is followed, with $\gamma = 5$. If one assumes that the system contains a small amount ($c \approx 10^{-3}$) of impurities, it follows that they nevertheless provide a strong random field with $H_0 \approx 350$ K. This is a reasonable value since H_0 should be of the order of a vortex creation energy. Evidently, the quantitative results from this comparison should be regarded with some caution. Since $\langle m_{\perp,\parallel}^2\rangle \propto d/q$ and $d \propto D_{eff}^{-\frac{1}{2}}$ the field-dependence of d (which follows from eq.(2) should be taken into account. Thus for $H \to H_{sf}$ the field-dependence of d would even become the leading term. In that limit, however, the behaviour will become quite complex. The walls will start to overlap as soon as $d \approx q$, and the present simple model does not cover that situation.

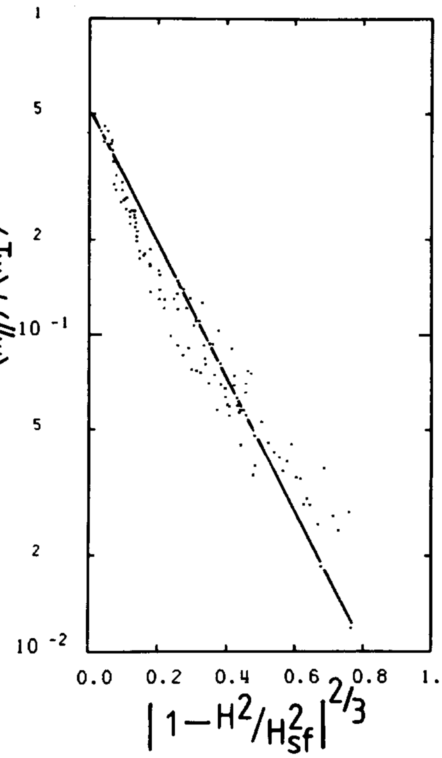

Fig. 4 Semi-logarithmic-plot of $\langle m_\perp^2\rangle$ for $H_x < H_{sf}$ and $\langle m_\parallel^2\rangle$ for $H_x > H_{sf}$ versus $|1 - H_x^2/H_{sf}^2|^{2/3}$. The solid line is the prediction from the random field model of Villain et al. (eq.(5)) with $\gamma = 5.0$.

Concluding remarks.

The strong broadening of the "spinflop" observed in quasi 2-d antiferromagnets with weak Ising anisotropy cannot be explained by demagnetizing effects as in the 3-d case. Also a droplet analogue of the soliton pair-states used in paper I to explain similar broadening in the chainlike system, does not seem to apply. Instead, we have argued that random fields may be responsible for domain-wall formation near H_{sf} in the quasi 2-d antiferromagnet, since their presence can explain the observed exponential behaviour of the broadening for reasonable values of the random field parameters. Thus it appears that the spinflop problem in the weakly anisotropic antiferromagnet offers a new and exciting possibility for experimental studies of the random field problem. A final remark to be made is the following. A crucial problem in the theories of random fields is the calculation of the lower critical dimension d_c, below which random fields may lead to a domain pattern [17]. There is a debate on whether d_c equals three or two [18]. Indeed the latter value would agree with the present analysis of the "spinflop" phenomenon. Although impurities will certainly also be present in the 3-d antiferromagnets, they apparently do not lead to domain-wall formation, since the observed spinflop transitions the in 3-d case are quite narrow [19]. The observed widths can be fully accounted for by demagnetizing effects so that the transition is apparently first-order in 3-d antiferromagnets. This is in agreement with recent theoretical developments [20] that favour $d_c = 2$.

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