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PHASE DIAGRAMS OF WEAKLY ANISOTROPIC HEISENBERG ANTIFERROMAGNETS: I. QUASI 1-DIMENSIONAL SYSTEMS

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By utilizing the concept of effective field-dependent anisotropy, it is argued that the field-induced transitions in a weakly anisotropic quasi 1-d Heisenberg antiferromagnet are "classical examples" of soliton-mediated phase transitions. For any $T > 0$ the "spinflip transition" is probably absent; if it still occurs, it is no longer first order as in 3-d. Excellent agreement with data on $(\text{CH}_3)_4\text{NMnCl}_3$ and K_2FeF_5 is found.

1. Introduction

Domainwalls in weakly anisotropic antiferromagnetic Heisenberg chains are of interest since they provide excellent examples of Sine-Gordon solitons¹⁻⁴. In this note we show that the antiferromagnetic phasediagrams (T_c versus H) of such quasi 1-d antiferromagnets can be fully explained on basis of the field-dependence of the soliton density and the soliton width. The field-induced transitions in these systems turn out to be examples of soliton-mediated phase transitions. The physics underlying the behaviour of the phase boundaries in quasi 1-d antiferromagnets is thus considerably different from the conventional 3-d counterparts.

2. The soliton-model

Consider an antiferromagnetic Heisenberg chain with weak orthorhombic (bi-axial) anisotropy and classical spins $\vec{S}_i$. The interaction Hamiltonian is:

$$H = \sum_i \{ -2J \vec{S}_i \cdot \vec{S}_{i+1} - D_x S_x^2 + D_z S_z^2 - g\mu_B \vec{H} \cdot \vec{S}_i \} \quad (1)$$

where $J < 0$, $D_x > 0$, $D_z > 0$, $D_z > D_x$ and $|J| \gg D_{x,z}$. The term $D_z S_z^2$ introduces a planar anisotropy, with the z axis as the hard axis. We will assume, without loss of generality, this axis to coincide with the chain direction (cf. fig. 1). In the easy (XY) plane the term $D_x S_x^2$ singles out the X axis as the preferential axis.

Applying a magnetic field $\vec{H} = H_x$ along the easy axis the well-known spinflop phenomenon will occur. As soon as the difference $\frac{1}{2}(\chi_y - \chi_x)H_x^2$ in the Zeeman energy between the perpendicular and the parallel orientation of the moments with respect to H_x exceeds the anisotropy energy $D_x S^2$, the moments will flop towards the Y -axis. Strictly speaking this spinflop transition applies to the long-range ordered phase of a 3-d antiferromagnet, i.e. below T_c . However, in a quasi 1-d antiferromagnet an analogous phenomenon also occurs in the paramagnetic phase, as

soon as the temperature is low enough that the 1-d correlation length along the chains is sufficiently well developed, so that the difference $\chi_y - \chi_x$ between the perpendicular and parallel magnetic susceptibility is appreciable. Then the moments within a correlated chain segment will be on the average perpendicular to H_x as soon as this exceeds the spinflop-value H_{sf} given by $H_{sf}^2 = 2D_x S^2 / (\chi_y - \chi_x)$.

Consider now the antiferromagnetic domain-wall (soliton) for $H_x < H_{sf}$ as sketched in fig. 1a. On both sides of the wall the moments only have X components, whereas over the wall width d_s they rotate in the XY plane, and have both X and Y components. The wall energy E_s (i.e. the soliton creation energy) can be easily estimated^{2,5} by an argument similar to that used in

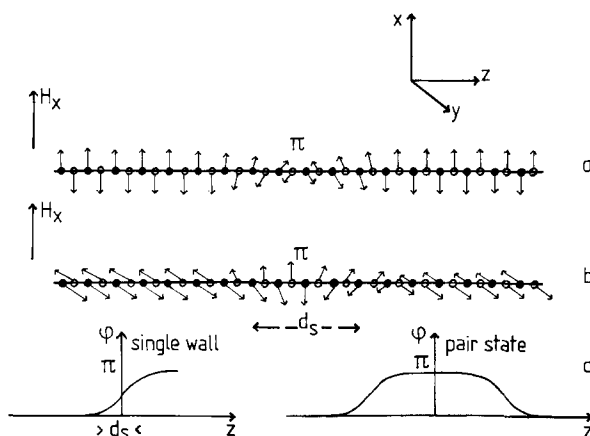


Fig. 1. Sine-Gordon solitons in antiferromagnetic chains.

(a) antiferromagnetic π soliton for $H < H_{sf}$, (b) antiferromagnetic π soliton for $H > H_{sf}$ and (c) schematic representation of a single wall and a pair state.

ferromagnetic domain theory⁶. If we measure the extension of the wall by means of an effective thickness over which interval the spin rotation angle amounts to $\Delta\phi$ and the number of spins to N , the wall-energy will be given approximately by:

$$E_s = (\Delta\phi)^2 JS^2/N + ND_x S^2 - \frac{1}{2}N(\chi_y - \chi_x)H_x^2,$$

since it is the sum of exchange-, anisotropy-, and Zeeman-energy terms. After minimization with respect to N one finds that, apart from a factor of order unity, the wall energy equals:

$$E_s = \frac{\Delta\phi}{2} g\mu_B S(H_{sf}^2 - H_x^2)^{\frac{1}{2}} (1 - \chi_x/\chi_y)^{\frac{1}{2}}, (H_x < H_{sf}).$$

For $H_x > H_{sf}$, on the other hand, the moments on both sides of the wall will be on the average along the Y direction (cf. fig. 1b), and one finds similarly for the wall energy:

$$E_s = \frac{\Delta\phi}{2} g\mu_B S(H_x^2 - H_{sf}^2)^{\frac{1}{2}} (1 - \chi_x/\chi_y)^{\frac{1}{2}}, (H_x > H_{sf}).$$

In both cases the wall width N (in units of the lattice constant a_0) is given by $N = 2(\Delta\phi)^2 |J| S^2 / E_s$. At low temperatures $\chi_x \ll \chi_y$, so that the expression for E_s may then be summarized by:

$$E_s = \frac{\Delta\phi}{2} g\mu_B S H_{sf} \left| 1 - H_x^2/H_{sf}^2 \right|^{\frac{1}{2}} \quad (2)$$

Relation (2) has been derived previously in a more rigorous way by Leung et al.⁴ for the Hamiltonian (1), transformed to its Sine-Gordon form. In order to conform with their formalism, we take $\Delta\phi = 0.5$ and $d_s = 8N$, where d_s now represents the Sine-Gordon wall-width $d_s = m_s^{-1}$. For fields $H_x \gg H_{sf}$ eq. (2) reduces to $E_s \approx g\mu_B S H_x$, as used by Boucher et al.¹ in the analyses of NMR and neutron scattering data on the solitons in $[(CH_3)_4MnCl_3]$ (alias TMMC). We note that the derivation of the above formulae is only valid under the conditions $g\mu_B H_x \ll D_x S^2$ and $g\mu_B H_x \ll |J| S(S+1)$.

Furthermore, for $H_x \rightarrow 0$ eq. (2) reduces to $E_s = g\mu_B S H_{sf} = 4S^2(D_x J)^{\frac{1}{2}}$, which is identical to the energy of a ferro-magnetic domainwall⁶ (and to the soliton energy in an antiferromagnetic⁷ or ferromagnetic⁸ Ising-like chain). For the antiferromagnetic case with $H_x \neq 0$ we may thus write:

$$E_s = 4S^2(D_x^H |J|)^{\frac{1}{2}}, \text{ with } D_x^H = D_x \left| 1 - H_x^2/H_{sf}^2 \right|$$

The quantity D_x^H may be interpreted as an effective, field-dependent anisotropy⁹. The crucial difference with the ferromagnet is therefore that in the antiferromagnet the anisotropy and thus E_s may be varied and even reduced to zero for $H_x \rightarrow H_{sf}$. The "spinflop" phenomenon may be seen as a softening of the soliton excitation mode. As E_s is lowered for $H \rightarrow H_{sf}$, the wall density n_s and the wall width d_s increases. For Sine-Gordon solitons the number of kinks and antikinks is reported to be given in the low-density limit by⁴:

$$n_s = 2(2/\pi)^{\frac{1}{2}} d_s^{-1} (E_s/k_B T)^{\frac{1}{2}} \exp(-E_s/k_B T) \quad (3)$$

At H_{sf} there will be an equipartition of parallel (X) and perpendicular (Y) spin components,

and one passes from one description (fig. 1a) into the other (fig. 1b).

On the other hand, if $\vec{H}$ is applied along the next-preferred axis, i.e. $\vec{H} = H_y$ (with $g\mu_B S H_y \ll D_x S^2$), the Zeeman energy adds up to the anisotropy energy $D_x S^2$ instead of competing with it, so that the effective field-dependent anisotropy⁹ is given by: $D_x^H = D_x(1 + H_y^2/H_{sf}^2)$. For increasing H_y there is only an increase of E_s (lowering of n_s):

$$E_s = g\mu_B S H_{sf} (1 + H_y^2/H_{sf}^2)^{\frac{1}{2}} \quad (4)$$

3. Antiferromagnetic phase diagrams

As a first application we consider the field dependence of the 3-d ordering temperature $T_c(H)$ of a system of weakly coupled antiferromagnetic chains induced by the weak interchain coupling J' (typically $J'/J \approx 10^{-2} - 10^{-4}$). The value of T_c can be reasonably estimated by a molecular field type argument, in which the thermal energy at the transition, $k_B T_c$, is equated to the interaction energy between two neighbouring correlated chain segments¹⁰, the length of which is given by the intrachain correlation length $\xi(T)$: $k_B T_c \approx |J'| S(S+1) \xi(T_c)$. Although this equation only yields approximate quantitative values for T_c , the proportionality $T_c \propto \xi$ should have a validity beyond the mean field approximation. We shall use it therefore to predict the relative variation of T_c with H on basis of the soliton model. Since the number of (thermally) excited solitons will determine ξ , we may put $\xi(H, T) \propto 1/n_s(H, T)$, so that the relative variation of T_c with H will be given by: $T_c(H)/T_c(0) = n_s(0)/n_s(H)$. Inserting formula (2) for $E_s(H_x)$ in expression (3) for $n_s(E_s, T)$, the variation of n_s and thus of T_c with H_x can be easily evaluated numerically. As an example we show in fig. 2 results obtained for the well-known quasi 1-d antiferromagnet TMMC. The calculations (dashed lines) are seen to account quite

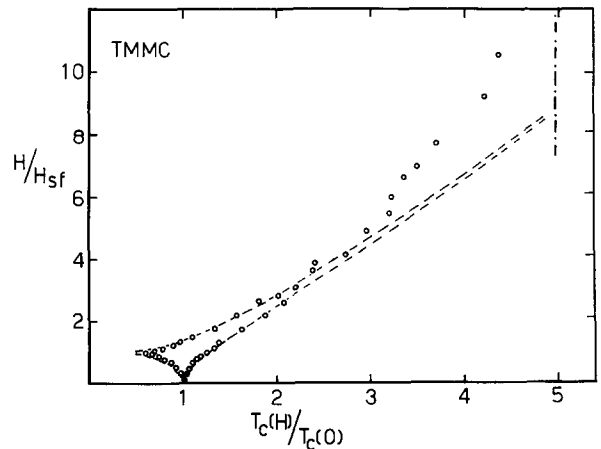


Fig. 2 Phase diagram of TMMC. Data obtained from refs 1,9,12. The dashed lines give the prediction from the XY soliton model; the dot-dashed line gives the limiting behavior from the YZ soliton model.

well for the observed behaviour^{9,11,12} up to a field $H_x \approx 6H_{sf}$. Similar agreement is found for $H = H_y$, where now eq. (4) is used for E_s . We stress that, except for $T_c(0) = 0.85$ K and $H_{sf} = 10$ kOe, there are no adjustable parameters in our simple model.

In fact the underlying reason why the model works so well up to fields relatively high compared to H_{sf} is the large value for the ratio D_z/D_x which is about 90 for TMMC^{1,12}. For the model to apply it is essential that the field energy remains small compared to the planar anisotropy term $D_z S^2$ that keeps the moments within the XY plane^{7,13}. We may associate a critical field $H_c = (2D_z S^2/\chi_\perp)^{1/2}$ with this anisotropy term by equating it to the Zeeman energy $\frac{1}{2}\chi_\perp H^2$. Now, if $D_x \ll D_z$ then $H_{sf} \ll H_c$, and the moments will remain essentially within the XY plane in a field range extending from $H = 0$ up to values much larger than H_{sf} . Only the direction of the easy axis is changed from X to Y at H_{sf} ; on both sides of H_{sf} the symmetry of the system is planar with a weaker inplane Ising-type component, as required by the model.

On the other hand, as the field H_x becomes of the order of H_c , the planar XY anisotropy due to the $D_z S^2$ term is more and more reduced by the field energy term, which competes with it since the field H_x forces the moments to lie within the YZ plane. As pointed out by Harada et al.¹³ both planar anisotropies balance at the critical field H_c , where the symmetry of the system becomes purely uniaxial with the Y-axis as the Ising axis. For $H_x > H_c$ the system is once more planar, with an in-plane Ising component, but now with the YZ plane as the easy plane. Also in this field range solitons occur⁷, however the rotation over the wall width is now essentially confined to the YZ plane, whereas the soliton creation energy becomes $E_s \approx 4S^2(D_z|J|)^{1/2}$. With the field along the Y axis the same phenomenon occurs, but now the easy plane is changed at $H_y = H_c$ from the XY to the XZ plane. The dot-dashed curve in fig. 2 shows the prediction for this high-field range (for TMMC the field $H_c = 110$ kOe) with $n_s(H) = n_s(H_c)$.

Recently, NMR experiments have been performed¹⁴ in the high field region, which also show the existence of the high-field soliton regime.

The symmetry changes brought about by the applied field are therefore basically different for orthorhombic anisotropy as for an uniaxially symmetric hamiltonian ($D_z = 0$ in eq. 1). Then the change at $H = H_{sf}$ is from purely uniaxial (Ising) to purely planar (XY), with complete Heisenberg isotropy at H_{sf} . This is the situation studied in most theoretical papers, which mainly concentrate on the bi-critical behavior expected at this Heisenberg point in 3-d antiferromagnets. For orthorhombic anisotropy, however, the symmetry of the system remains Ising-type with a superimposed planar anisotropy at all field values except at $H = H_{sf}$, where the symmetry becomes purely planar (XY), and at H_c , where it becomes purely uniaxial (Ising). We stress these differences since they appear to be not always realised. Also, it is clear that the strong variation of T_c with H for TMMC cannot possibly be attributed to a field-induced symmetry change from Ising-type for $H < H_{sf}$ to XY type for $H > H_{sf}$, as has been postulated previously^{9,10,11}.

In concluding this section we mention that previously the phase boundaries of quasi 1-d antiferromagnets have been successfully explained in terms of transfer matrix theory^{12,15}. In our opinion our present results do not contradict this earlier work, since any correct description should (implicitly) take into account the presence of the nonlinear excitations. The advantage of our simple soliton model seems to be that it demonstrates quite clearly the physics responsible for the observed huge variations, which is not so transparent from the transfer matrix calculations.

4. "Spinfloptransition" in the 3-d ordered phase

Another consequence of the model is that the excitation of a soliton in the low-field phase ($H_x < H_{sf}$) may be viewed as the admixture of a small segment of the high-field phase ($H_x > H_{sf}$) in this low-field phase (cf. fig. 1a), because over the wall width the spins have perpendicular components. Similarly the excitation of a soliton in the high-field phase (fig. 1b) corresponds to the admixture of a small low-field segment. Since for $T \neq 0$ and any finite field (smaller than H_c) there is a finite probability for the excitation of these walls, they will change the character of the spinflop transition, which would be first order in a 3-d antiferromagnet. In the conventional spinflop the magnetization changes discontinuously from $M_x = \chi_x H_x$, which is very small at low temperatures, to $M_y = \chi_y H_x$, which is nearly independent of temperature. However, if solitons are excited as in fig. 1a,b the contributions from the spins over the wall width correspond with an increase and a decrease of the magnetization for $H_x < H_{sf}$ and $H_x > H_{sf}$ respectively. For $H_x \rightarrow H_{sf}$ the wall width and the number of walls increase, and the first order character of the transition is destroyed (no discontinuity in the magnetization), except at $T = 0$. Indeed, this would explain the absence of discontinuities in M at the "spinfloptransitions", as reported in several experimental studies of the magnetization curves in quasi 1-d antiferromagnets.

An important point is that in the 3-d ordered state the static (topological) solitons should occur in pairs due to the interchain interactions. The excitation of a single π -wall in this phase would imply that about half of the spins of a chain would be overturned against the interchain coupling J' . Although J' is small, it then has to be multiplied by a large number of spins, so that the cost in interchain interaction energy becomes much larger than the wall energy itself. In case of a wall-pair creation, on the other hand, only the spins inside and in between the two walls are contributing to the interchain interaction energy. If ϕ is the angle the spins make with the X axis, the variation of ϕ along the chain (Z) axis in case of a wall-pair is given by¹⁶:

$$\frac{1}{2}\phi = \tan^{-1}[(s/v) \sinh vt / \cosh z]$$

The energy of such a pair is equal to $2E_s$. In the limit $vt \gg 1$ (soliton and antisoliton wide

apart) this equation can be approximated by¹⁶:

$$\frac{1}{2}\phi = \tan^{-1} \exp(-\gamma(z+z_0+vt)) - \tan^{-1} \exp(-\gamma(z-z_0-vt))$$

which is just the combination of a noninteracting soliton and an antisoliton (cf. fig. 1c). Assuming thus the variation of $\phi(z)$ over the wall-width to be governed by the Sine-Gordon equation, it is easy to calculate the wall contribution to the magnetization. Since this contribution is proportional to the density, the comparison with the experiment allows in principle a direct experimental test of the prediction for n_s . As an example we show in fig. 3 data for K_2FeF_5 , obtained by Gupta et al.¹⁷, who followed the field variation of the average angle $\langle\phi\rangle$ by means of the Mössbauer effect. This Fe^{3+} compound is another example of the weakly anisotropic Heisenberg antiferromagnetic chain with $T_C(0) \approx 7.0$ K, $H_{sf} = 36$ kOe, and $|J'/J| \approx 10^{-3}$. From this experiment one would conclude that the moments rotate continuously from the X to the Y axis, as already remarked in ref. 17. The width of the "spinflop transition" is 20 kOe! This is nearly two orders of magnitude larger than expected from demagnetizing effects that accompany a first-order magnetic transition.

The theoretical curves in fig. 3 have been obtained as follows. In the antiferromagnet, the Mössbauer effect cannot distinguish between an angle ϕ and an angle $\pi - \phi$, so in order to obtain $\langle\phi\rangle$ from Sine-Gordon theory we have to average over the interval $\langle 0, \pi/2 \rangle$, which is in fact half the soliton. Since for a given density n_s each soliton has an average "free space" n_s^{-1} we obtain:

$$\langle\phi\rangle = 2n_s \int_0^\infty \phi(z) dz = 2n_s d_s \int_0^{\pi/2} \frac{\phi}{\sin\phi} d\phi \approx 3.6 n_s d_s$$

If we apply (3) we get:

$$\langle\phi\rangle \approx 18.0 (2E_s/k_B T)^{\frac{1}{2}} \exp(-2E_s/k_B T), \quad (5)$$

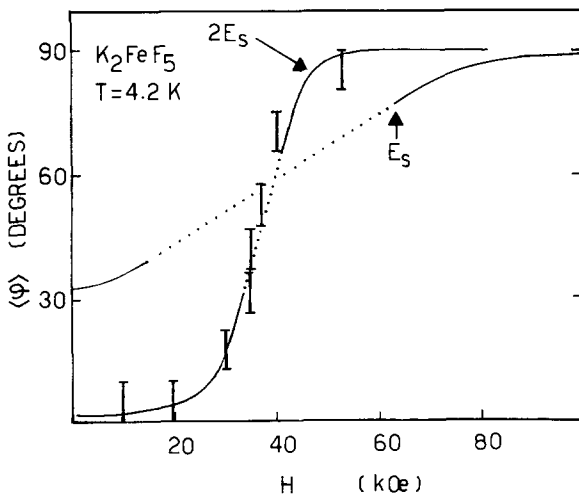


Fig. 3 Spin-flop in K_2FeF_5 at $T = 4.2$ K. Data obtained from ref. 17. Solid curves are predictions from the soliton model with E_s and $2E_s$. Dotted curves are interpolations.

where we have used an excitation energy of $2E_s$. Inserting eq. (2) for E_s the field-dependence of $\langle\phi\rangle$ can be estimated, except in the immediate neighbourhood of H_{sf} , where the divergence of the exponential leads to unphysical behavior. Clearly, for $H \rightarrow H_{sf}$ the density n_s as well as the wall-width d_s increase, so that the walls will start to overlap and the low-density, non-interacting approximation breaks down. Thus the calculated curves in fig. 3 extend to about 10 % around H_{sf} , the high- and low-field portion having been connected by linear interpolation. (dotted curves). There appears to be good agreement with experiment. For comparison we also show in fig. 3 the result obtained on basis of (5), but now with E_s instead of $2E_s$ as the excitation energy. This would apply if the solitons and antisolitons would be excited independently, each with excitation energy E_s . One would conclude that only the wall-pair approach may account for the experimental findings.

We also remark that in a recent study¹⁸ of the Mössbauer spectra of K_2FeF_5 as a function of field the Liverpool group has observed considerable line-broadening as the field approaches the value for H_{sf} . This may be explained in terms of a distribution of ϕ values for $H \approx H_{sf}$, in agreement with our model.

5. Concluding remarks

In the above we have argued that the excitation of solitons can account for the field-dependent thermodynamic behavior of quasi 1-d antiferromagnets. In the paramagnetic phase the walls are dynamic; they determine the intrachain correlation length, and thus the value of the 3-d ordering temperature $T_C(H)$. In the 3-d ordered phase the solitons will be static and will be excited in pairs. As explained, this may account for the absence of a first-order spinflop transition in such materials. In case the transition would still persist as a second order line, the phase diagram would look as in fig. 4a where only the point at $T = 0$, $H = H_{sf}$ would be first order. On the other hand there seems to be no reason why the transition would not be completely absent except at $T = 0$. One could well imagine a continuous evolution at low temperature as a function of field from a situation where $\langle S_x \rangle \gg \langle S_y \rangle$ to the situation where $\langle S_y \rangle \gg \langle S_x \rangle$. There would then be just a single antiferromagnetic phase, characterized by a soliton lattice, separated from the paramagnetic phase by a boundary as sketched schematically in fig. 4b. The umbilicus at $H = H_{sf}$ in this curve would correspond to $\langle S_x \rangle = \langle S_y \rangle$, in accord with the expected planar symmetry at this point. In the soliton model this would occur as soon as the density and width of the walls are so large that neighbouring walls touch one another, i.e. as soon as $1/n_s = 2d_s$, which leads to the condition $x^2 e^{-x} = (\pi/32)^{\frac{1}{2}}$, or $x \approx 0.13$, where $x = E_s/k_B T$. This would correspond to $H_x/H_{sf} = 0.999$, for TMMC at $T = 0.8$ K as well as for K_2FeF_5 at $T = 4.2$ K. This implies that the precise behavior for $H = H_{sf}$ will be difficult to observe experimentally; the high- and low-field part of the phase boundary would at first sight appear to bifurcate with a common tangent

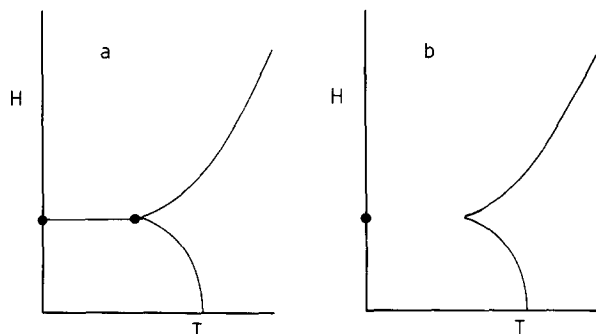


Fig. 4 Possible phasediagrams proposed for quasi 1-d antiferromagnetic with orthorhombic anisotropy (see text).

from a single (bicritical) point, as they do in the conventional 3-d case. Obviously, additional accurate experimental work would be welcome, as well as more sophisticated theoretical treatments of the points raised by our simple model. We also stress the analogies between the above treated problem and the field-induced transitions in Spin-Peierls system, as already pointed out elsewhere⁵.

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