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PHASE TRANSITIONS IN UNIAXIAL ANTIFERROMAGNETS

by

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ABSTRACT

We study an instability of the antiferromagnetic phase when subjected to an external field along the easy axis of the uniaxial antiferromagnet. This instability leads to a formation of a magnetic order with more than two sublattices. This instability occurs when the intra-sublattice exchange is also antiferromagnetic, and it may happen for fields smaller than the critical fields for the transition to the spin-flop or intermediate phases.

1. Introduction

The magnetic phases of uniaxial antiferromagnets have been studied by Yamashita[1]. That work was preceded by several papers, among which the one Rohrer and Thomas[2] and the one by Fairall and Cowen[3]. More recently, Morrison[4] extended Yamashita's work by the inclusion of the single-ion second anisotropy interaction and the Dzyaloshinsky-Moriya exchange. All these papers dealt with antiferromagnets with two sublattices in an external field along the easy axis, and at T=0K.

The Hamiltonian for the problem, restricting ourselves to the interactions studied by Yamashita, and using that author's notation, can be written as

$$E = \frac{1}{2}\Gamma\,(M^{+}.M^{+} + M^{-}.M^{-}) + A\,M^{+}.M^{-}$$

$$+ \frac{1}{2}\Phi_x\,(M_x^{+}M_x^{+} + M_x^{-}M_x^{-}) + \Phi_x'\,M_x^{+}M_x^{-} - H(M_x^{+} + M_x^{-}) \quad (1)$$

where x is the direction of anisotropy along which we apply the field H. In(1) the second and fourth terms are the inter-sublattice isotropic and anisotropic exchange interactions. The first and third terms refer to an interaction happening within a sublattice. These terms can be variously interpreted either as an exchange interaction in the sublattice, or as a crystalline field acting on each spin. If one uses the first interpretation, and within the framework of the molecular field theory (statistical independence of spins), the average value of these terms is treated as

$$\frac{1}{2}\,\Phi_x < M_x^+ M_x^+ > = \frac{1}{2}\,\Phi_x < M_x^+ >^2 \qquad (3)$$

since the two spin operators refer to different units in the sublattice. On the other hand, the crystalline field interpretation does not permit one to equal the average of the product of the operators to the product of the averages. In this case, the two operators refer to the same spin. In what follows, the results obtained with the two interpretations are very different.

The preceding works mentione above have the common feature of assumeng that at T=OK the average of a spin operator always attains its maximum value M_o. Barring one special circumstance to be discussed below, this assumption is amply justified within the molecular field framework. Indeed, a molecular field is a sum of the external field and of effective fields due to the exchange interaction with neighbouring spins. At T=OK, if the molecular field acting on a spin is not zero, even if it is very small, the spin will minimize its energy by choosing a state with maximum projection along the molecular field. Thus one has only to consider the direction of this axis and minimize the energy of (1) with respect to the angles defining this axis. This procedure was fully explored by Yamashita[(1)].

A special case may happen when the molecular field for one of the sublattices is exactly zero. In this case, the average value of the sublattice magnetization becomes undetermined and may assume any value between zero and M_o. In this circumstance, the magnitude of the spin becomes a variational parameter for the minimization of (1). One may see that this case is only possible when the two sublattices have magnetizations aligned with the external field. Indeed, for canted magnetizations, the vector sum of the exchange and external fields is never null.

In what follows we are going to study the situtions when the two sublattice magnetizations are aligned with the field but have arbitrary values between $-M_0$ and M_0. The study neglects quantum effects and is made for the intra-sublattice exchange interpretation of the 1st and 3rd terms in (1)(section 2), as well as for the crystalline field interpretation (section 4).

2. Intra-sublattice exchange interaction

In this case Eq.(2) is valid, and we let σ and μ be the average magnetizations of the two sublattices, assuming that both are aligned with the field. Then, the average value of the energy (at T=OK) becomes

$$\langle E\rangle = \frac{1}{2}(\Gamma + \Phi_x)(\sigma^2+\mu^2) + (A+\Phi_x')\,\sigma\mu - H(\sigma+\mu) \qquad (3)$$

This ecpression has an extreme at

$$\sigma = \mu = \frac{H}{\Gamma + A + \Phi_x + \Phi_x'} \qquad (4)$$

and this extreme is a minimum if

$$\Gamma + \Phi_x > A + \Phi_x' > 0 \qquad (5)$$

If (5) is satisfied, the solution may exist for fields wich make (4) to be in the range of zero to M_o, and whenever its energy is lower than the energy of the antiferromagnetic (AF) phase, or

$$0 < H < M_o(\Gamma + A + \Phi_x + \Phi_x')$$

The inequality (5) means that the intra-sublattice exchange is greater than the inter-sublattice interaction. This is an unlikely situation is an antiferromagnet with two sublattices because it favours other types of magnetic arrangements. For this reason we are not taking the discussion of this solution any further.

If (5) is not satisfied, namely

$$A + \Phi_x' > \Gamma + \Phi_x \qquad (6)$$

then one of the sublattices has a magnetization equal to M_o. Thus setting

$$\mu = M_o$$

in (3) and minimizing the energy with respect to σ we obtain

$$\sigma = \frac{H - M_o\ (A + \Phi'_x)}{\Gamma + \Phi_x} \tag{7}$$

if $-M_o < \sigma < M_o$

and the stability condition

$$\Gamma + \Phi_x > 0 \tag{8}$$

This solution represents an antiferromagnetic phase in which the sublattice anti-paralel to the external field as a magnetization smaller than M_o. From here on we refer to this solution as the Reduced Spin (RS) phase.

The RS phase has a critical field of transition

$$H_{c1} = M_o(A+\Phi'_x - \Gamma - \Phi_x) \tag{9}$$

to the normal AF phase, and a critical field

$$H_{c2} = M_o\ (A+\Phi'_x + \Gamma + \Phi_x) \tag{10}$$

to the ferromagnetic phase. Depending on the value of Γ, the RS phase may be stable or not with respect to the other phases studied by Yamashita. It is worth noting that the parameter Γ does not come in any theory which, at T=OK, begins by making the sublattice magnetization equal to M_o. In that case, tje corresponding term in Eq.(1) is a constant and may be discarded. However,Γ becomes an important parameter when we study a situation, such as the present, in which the sublattice magnetization is less than M_o.

When varying Γ between the limits set by (6) and (8), one may obtain many situations in which the RS phase is the most stable. For instance, defining, as Yamashita, the following symbols

$$Q = \frac{\Phi'_x - \Phi_x}{2A + \Phi'_x - \Phi_x} \tag{11}$$

$$R = -\frac{2\,\Phi_x}{2A + \Phi'_x - \Phi_x} \tag{12}$$

and letting

$$\gamma = \frac{\Gamma}{2A + \Phi'_x - \Phi_x}$$

it is simple to establish that in the region of the Q vs R plane where

$$R > 0 \;, \qquad Q > 0 \quad \text{and} \quad Q + R < 1$$

when, according to Yamashita, we could have only the AF, the SF (spin flop) and the F phases, the RS phase sets in for a lower field than the SF. In order for this to happen it is enough to satisfy the inequality

$$H_{AF \to SF} > H_{AF \to SR}$$

or

$$\sqrt{Q(1 - R)} > \frac{1+Q}{2} - \gamma \tag{13}$$

Certainly, a value of γ may be chosen to satisfy (13) and the inequalities (6) and (8), here rewritten as

$$\frac{1+Q}{2} > \gamma > R/2 \qquad (14)$$

3 - Instability of the RS phase

The RS phase is characterized by the onset of a new order parameter: the value of the magnetization of one of the sublattices. The RS is not truly a phase since at $T \neq 0K$ the parameter of order does not appear suddenly, as the field increases, but already exists at $H = 0$. This means that though there is a discontinuity of the differential susceptibility at $T = 0K$, the discontinuity ceases to exist athigher temperatures.

On the other hand, the RS phase is unstable against the formation of new sublattices. For instance, assume that, in the reduced spin sublattice, the exchange parameters Γ and Φ_x refer to an interaction between neighbours. The reduced spin sublattice may be decomposed into two new sublattices, one having an average spin $\sigma + \xi$ and the other $\sigma - \xi$. Then, the first and third terms in (1), corresponding to the reduced spin sublattice, add up to

$$\frac{1}{2}\,(\Gamma + \Phi_x)(\sigma^2 - \xi^2)$$

If $\Gamma + \Phi_x$ is greater than zero, a necessary condition for the existence of the RS phase (see (8)), the energy is lowered by making ξ as large as possible. Thus we arrive at the conclusion that the onset of the RS phase is truly the onset of a much more complicate magnetic ordering, the nature of which is difficult to guess.

Thus our calculation with the two sublattice model sets the limits of its own validity. These limits are established by the occurence of the RS phase, which is unstable against the formation of a more complicate magnetic structure.

4 - Crystal Field Interaction

When the first and third terms in (1) are interpreted as the effect of the crystalline field, we cannot set the average of the product of two magnetizations as the product of the averages. In this case both magnetizations refer to the same spin.

Since

$$M_x^2 + M_y^2 + M_z^2 = M_o^2 \tag{15}$$

we can subtract from (1) a constant, or equivalently set

$$\Gamma = 0 \tag{16}$$

The variance of a magnetization component is always greater than zero, or

$$< M_x^2 > - < M_x >^2 \geq 0$$

Thus, confining ourselves to the case when the magnetizations are aligned with the field (direction x), and letting

$$\sigma = < M_x^- >$$

$$\mu = < M_x^+ >$$

we have the following cases:

1) $\Phi_x > 0$

In this case, for given values of σ and μ, the energy is minimized when

$$< M_x^{-2} > = < M_x^{-} >^2 = \sigma^2$$

$$< M_x^{+2} > = < M_x^{+} >^2 = \mu^2$$

Thus the present case is reduced to the case of intra sublattice exchange interaction with $\Gamma = 0$.

Because of the signal of Φ_x, the parameter R given by (12) is negative. According to Yamashita, in this region of the Q vs R plane the first phase to appear after the AF is the Intermediate phase. It can be verified that the critical field for the transition AF → RS is greater than the field for the transition AF → Intermediate. Thus the RS phase is never realized. In other words, in this case there seems to be no field for which there is instability against the formation of complicate magnetic structures.

2) $\Phi_x < 0$

In this case the energy is minimized if one sets

$$< M_x^{-2} > = < M_x^{+2} > = M_o^2$$

By subtracting a constant from (1) we can set

$$\Phi_x = 0$$

and again the RS phase does not exist.

Thus, while the intra-sublattice exchange can build a RS phase, the crystalline field is unable to do so. In this respect, the two interpretations of the first and third terms in (1) lead to very different results.

5. Extension for $T \neq 0K$

We consider the case when the first and third terms in (1) are interpreted as an intra-sublattice exchange. As we have explained before, for $T \neq 0K$, the sublattice magnetization is different from M_0 even for a zero external field. Thus the sublattice magnetization is not a parameter of order that varies after a certain critical field is reached. Then, in order to determine the beginning of the RS phase, we must look for the point of instability when one of the sublattices breaks down into other sublattices.

Let μ be the average magnetization of the "+" sublattice

$$< M_x^{+} > = \mu \tag{17}$$

and let us decompose the "-" sublattice into two, the "-+" and the "--" sublattices, each with half as many spins as the "+" sublattice. Their average magnetizations are

$$< M_x^{-+} > = \sigma + \xi \tag{18}$$

$$< M_x^{--} > = \sigma - \xi \tag{19}$$

For given values of σ, μ and ξ, the energy is determined from (1) as

$$E = \frac{N}{2}(\Gamma + \Phi_x)(\sigma^2+\mu^2-\xi^2) + N(A+\Phi_x')\,\sigma\mu - NH(\sigma+\mu) \tag{20}$$

where N is the number of spin pairs in the crystal. Thus one should maximize the entropy keeping constant the average magnetizations given by(17), (18) and (19). Introducing the Lagrange multipliers ρ ,λ and η, one has to minimize the following expression

$$F_o = -\frac{N}{2}(\rho+\eta)\langle M_x^{-+}\rangle - \frac{N}{2}(\rho-\eta)\langle M_x^{--}\rangle - N\lambda\langle M_x^{+}\rangle - TS$$

$$= -N(\rho\sigma + \lambda\mu + \rho\xi) - TS \qquad (21)$$

This minimization is attained when the entropy S and the averages are calculated in an ensemble with the following Hamiltonian[5]

$$H_o = -(\rho+\eta)\sum M_x^{-+} - (\rho-\eta)\sum M_x^{--} - \lambda\sum M_x^{+} \qquad (22)$$

where the sums extend over the spins of each sublattice. Thus

$$F_o = -\frac{N}{2} kT\, \ell n\, \mathrm{Tr}\exp\left[(\rho+\eta)M_x^{-+}/kT\right] - \qquad (23)$$

$$-\frac{N}{2} kT\, \ell n\, \mathrm{Tr}\exp\left[(\rho-\eta)M_x^{--}/kT\right] - N\, kT\, \ell n\, \mathrm{Tr}\exp\left[\lambda M_x^{+}/kT\right]$$

The Lagrange multipliers are determined from the fixed values of σ, μ and ξ according to the equations

$$\frac{1}{N}\frac{\partial F_o}{\partial \rho} = -\sigma \qquad (24a)$$

$$\frac{1}{N}\frac{\partial F_o}{\partial \lambda} = -\mu \qquad (24b)$$

$$\frac{1}{N}\frac{\partial F_o}{\partial \eta} = -\xi \qquad (24c)$$

Finally, the average values σ , μ and ξ of the magnetizations should be determined from the minimization of the free energy, which is given by the following expression

$$F = E - TS = E + N\,(\rho\sigma + \lambda\mu + \rho\xi) + F_0 \qquad (25)$$

where E is given by (20). Considering σ, μ and ξ as indipendent variables, we arrive at the equilibrium equations

$$\frac{1}{N}\,\frac{\partial E}{\partial \sigma} + \rho = 0 \qquad (26a)$$

$$\frac{1}{N}\,\frac{\partial E}{\partial \mu} + \lambda = 0 \qquad (26b)$$

$$\frac{1}{N}\,\frac{\partial E}{\partial \xi} + \rho = 0 \qquad (26c)$$

which, together with (24a-c) solve the problem.

The equations (26a-c) and the effective Hamiltonian(22) give a simple interpretation to the Lagrange multipliers ρ,λ, η. These multipliers are the molecular fields acting on the various sublattices.

Equation (26c) can be written as

$$\eta - (\Gamma + \Phi_x)\ \xi = 0 \qquad (27)$$

while (24c) can be rewritten as

$$\xi = \frac{1}{2}\,\mathrm{Tr}\left\{ M_x^{-+}\ \exp\left[(\rho + \eta)\ M_x^{-+}/kT \right]\right\} /\mathrm{Tr}\left\{ \exp\left[(\rho + \eta)\ M_x^{-+}/kT\right] \right\}$$
$$- \frac{1}{2}\,\mathrm{Tr}\left\{ M_x^{--}\ \exp\left[(\rho - \eta)\ M_x^{--}/kT \right]\right\} /\mathrm{Tr}\left\{ \exp\left[(\rho - \eta)\ M_x^{--}/kT\right] \right\} \qquad (28)$$

One solution to these equations is

$$\xi = \eta = 0$$

In this solution there is no difference between the sublattices "--" and "-+". Depending on the value of ρ, one may have a second solution with non zero ξ and η. The beginning of this second solution is determined by expanding (28) as a power series of η, and keeping only the linear terms in the small parameter η. We arrive at

$$\xi = \eta \frac{\partial}{\partial \rho} < M_x^- > \qquad (29)$$

where $< M_x^- >$, the net average magnetization of the "--" and "-+" sublattices, is defined by

$$< M_x^- > = Tr\{ M_x^- \exp (\rho M_x^-/kT)\} /Tr\{ \exp(\rho M_x^-/kT) \}$$

A plot of $\partial < M_x^- >/\partial\rho$ as function of the molecular field ρ is given in Fig.1. Eqs.(29) and (27) establish the beginning of the RS phase as the external field is increased from zero. For the critical field one has

$$\frac{1}{\Gamma + \Phi_x} = \frac{\partial < M_x^- >}{\partial \rho} \qquad (30)$$

As T is lowered, the curve becomes taller and thiner, because its area does not depend on T. Thus, the critical point moves towards $\rho = 0$, namely it approaches the point where the molecular field ρ is null. Then (26a) tends to the energy minimization made at T = OK. With increasing T, the RS phase disappears when the whole curve moves under the dashed line, or when

$$\frac{1}{\Gamma + \Phi_x} > \frac{M_o^2}{3kT}$$

Insert Figure 1

6 - Concluding Remarks

In this paper we have completed the work by Yamashita on uniaxial antiferromagnets, by studying an instability that has been overlooked by that author. This instability happens when the intrasublattice exchange is also antiferromagnetic and leads, for certain values of the external field, to the breaking down of the two sublattice model. In this case, a much richer magnetic order may result.

Note added in proof - It has been called to our attention the papers by:

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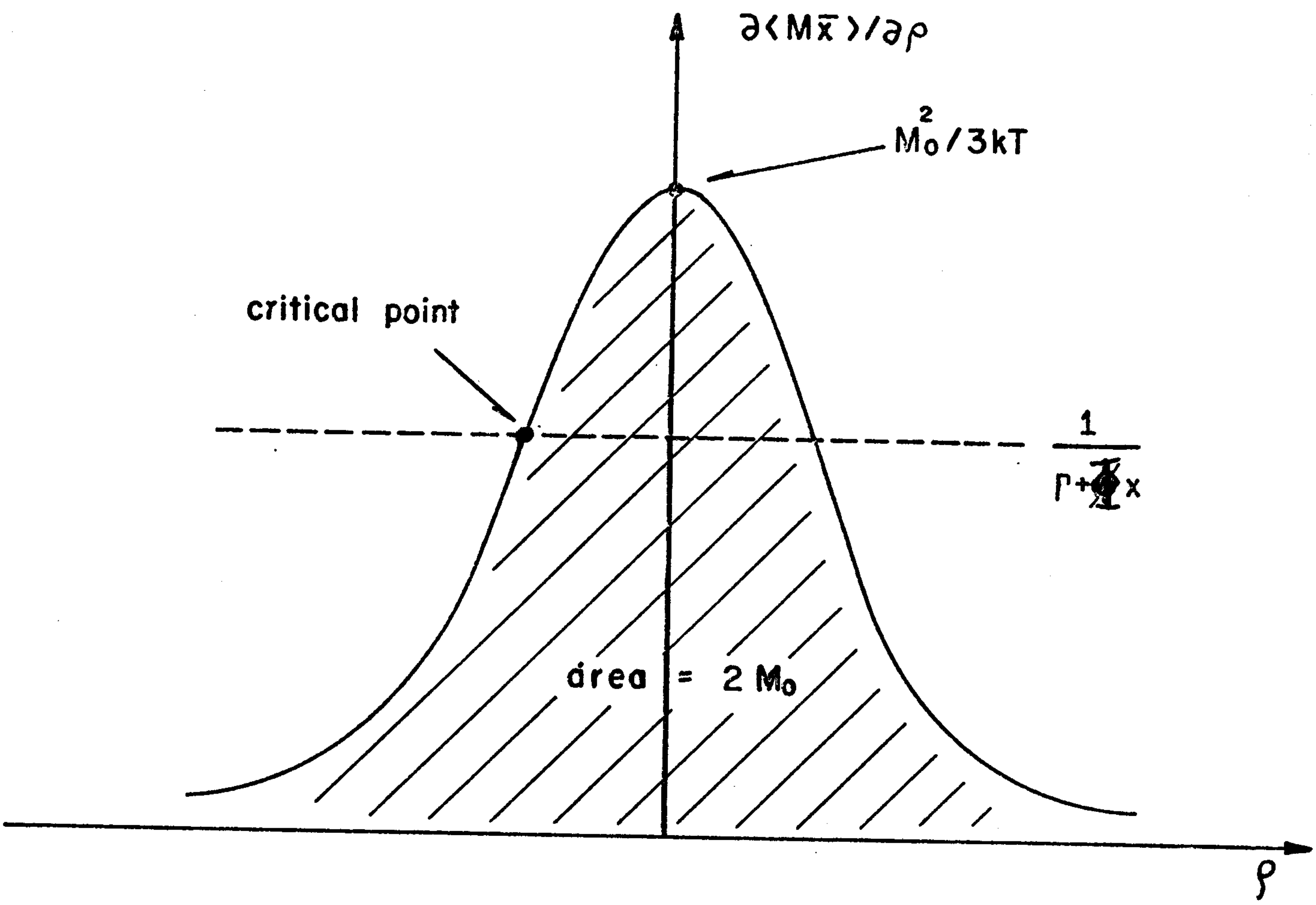

∂⟨Mx̄⟩/∂ρ
$M_o^2/3kT$
critical point
$\frac{1}{\Gamma+\Phi x}$
area = 2 M_o
ρ

FIGURE CAPTIONS

Fig. 1 - $\partial < M_x^- > / \partial \rho$ as function of ρ. The area under the curve does not depend on the temperature and is equal to $2M_o$.