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LETTER TO THE EDITOR

Spin reorientation in hexagonal antiferromagnets

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Abstract. A reorientation process in quasi-one-dimensional weakly anisotropic easy-plane antiferromagnets on a stacked triangular lattice is shown to occur via two intermediate phases. The first is of spin-flop character, while the second, with two pairs of flipped sublattices, normally exists only in a narrow interval near a saturation field.

An attempt to find an experimental verification of the Haldane conjecture for 1D antiferromagnets [1] renewed interest in the investigation, both experimental and theoretical, of quasi-one-dimensional antiferromagnets on a stacked triangular lattice [2–10].

In a previous letter [11] (hereafter referred to as I) one of us reported on the results of quasiclassical calculations for reorientation processes in XY-like antiferromagnets with an external field applied in the basal plane. This case proved to be rather interesting since the anisotropy and the exchange in the basal plane, of the same order of magnitude due to quasi-one-dimensionality, compete with each other: the anisotropy, D, tends to confine the spins to the basal plane while the exchange interaction, J', tends to align them perpendicularly to the field.

As was shown in I, for $D > 3J'$ (this is believed to be the case for $CsMnBr_3$ [3, 4] and $KNiCl_3$ [6]) the planar arrangement survives the application of the field completely. The reorientation was proved to be accompanied by a second-order transition when two pairs of sublattices flip. AFMR experiments [5, 8] confirm this scenario.

In the present letter we report on calculations for the reorientation process in the opposite case of relatively small anisotropy, when $q = D/3J' < 1$.

We start with two points already mentioned in I:

(i) For $q < 1$ the planar arrangement becomes unstable prior to the flip, at $H = H_c^{(1)} = 4S\sqrt{JD}$, which is a typical field for a spin-flop transition.

(ii) A saturation field $H_c^{(3)} = 8JS + 18J'S$ does not depend on D, so immediately after a ferromagnetic instability the spins are confined to the basal plane.

It is thus natural to propose that the reorientation process for $q < 1$ involves an additional intermediate phase with a non-planar spin arrangement. The width of this phase tends to zero when $q \rightarrow 1$.

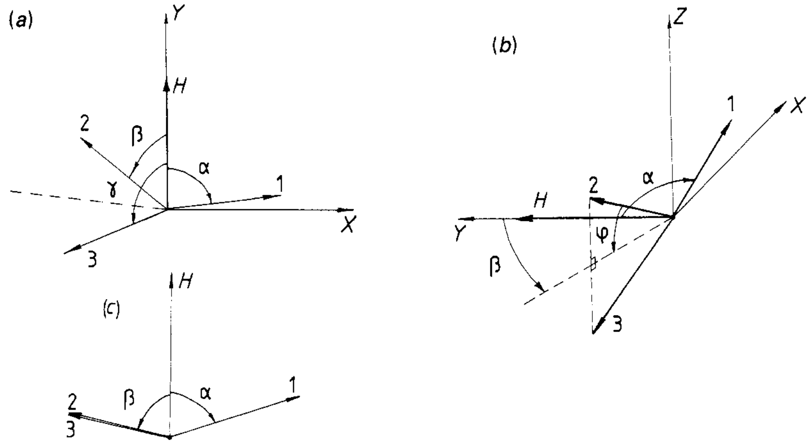

Figure 1. Successive spin configurations: (*a*) Planar phase, $H < H_c^{(1)}$; a broken line denotes $(\beta+\gamma)/2$; (*b*) spin-flop phase, $H^{(1)} < H < H_c^{(2)}$; (*c*) after a spin flop of two pairs of sublattices. The numbers denote sublattice spins in a given plane. An arrangement in the subsequent plane differs as follows: $\alpha \to -\alpha$, $\varphi \to -\varphi$, $\beta \to -\gamma$, $\gamma \to -\beta$.

We start with the microscopical spin Hamiltonian of an easy-plane antiferromagnet on a stacked triangular lattice:

$$\mathcal{H} = 2J\sum S_i S_{i+\Delta_z} + 2J'\sum S_i S_{i+\Delta_\perp} + D\sum (S_i^z)^2 - H\sum S_i^y. \tag{1}$$

The quasi-one-dimensionality implies $J \gg J'$, D, while $q = D/3J'$ (or, to be more exact, $\tilde{D}/3\tilde{J}'$ where $\tilde{D}$ and $\tilde{J}'$ differ from D and J' due to a short-range quantum renormalization; see I) is presumed to be less than unity.

A classical equilibrium spin configuration results from a minimization of (1) with sublattice spins substituted for by classical vectors. We have found three different successive phases (see figure 1). Independently of q, the reorientation always starts from the planar arrangement. In the leading order in D/J and J'/J the angles in figure 1(*a*) are given by

$$\cos\alpha = p \qquad \cos\left(\frac{\beta+\gamma}{2}\right) = \frac{p}{2-z} \qquad \cos\left(\frac{\beta-\gamma}{2}\right) = \frac{1}{2-z} \tag{2}$$

where $p = H/8JS$, $z = H^2/48JJ'S^2$.

At low fields, α and $(\beta+\gamma)/2$ are close to $\pi/2$ while $\beta-\gamma$ undergoes a rapid change and, in the absence of preliminary instabilities, two pairs of sublattices flip at $z = 1$, i.e. at $H = H_c = \sqrt{48JJ'S^2}$. This is exactly the situation for $q > 1$. However, for small anisotropy, when $q < 1$, the planar arrangement becomes unstable with respect to small deviations along the Z axis at lower fields, when H approaches $H_c^{(1)} = 4S\sqrt{JD}$. In the classical approach this instability results in a hysteresis-free first-order transition to the configuration shown in figure 1(*b*) with

$$\begin{gathered}\cos\alpha = p\left(1-\delta\frac{a+1}{a}\right) \qquad \cos\beta = p\left(1-\delta\frac{a+1}{2}\right)\Big/\cos\varphi \\ \cos\varphi = \left(p^2+\frac{1-p^2}{a^2}\right)^{1/2}\left(1-\frac{\delta p^2}{2a}\frac{(a+1)(a^3-2)}{(1+p^2(a^2-1))}\right)\end{gathered} \tag{3}$$

where $a = 2-q$, $\delta = 3J'/2J$.

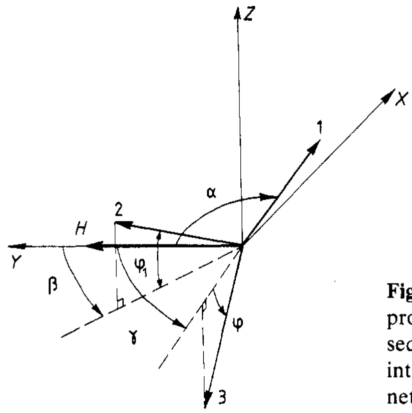

Figure 2. A displacement of the sublattice spins producing an instability at $H = H_c^{(1)}$. For the subsequent plane $\varphi_1 \Rightarrow -\varphi_2$, $\varphi_2 \Rightarrow -\varphi_1$, and the intermediate configurations thus have non-zero net Z-magnetization.

Note that although the final result of the transition can be represented as a rotation by $\pi/2$ about the broken axis in figure 1(a), the instability itself is induced by the displacements shown in figure 2, with

$$\varphi_2/\varphi_1 = (1+f)/(1-f) \qquad \text{where } f = (H_c^{(1)}/8JS)\,[(3-q)(1-q)]^{1/2}/(2-q). \tag{4}$$

Hence, the first-order transition involves a set of intermediate states with non-zero Z-magnetization. Since in any real situation a transition always has some finite width, this result can be verified experimentally.

The intermediate phase for $H > H_c^{(1)}$ is a typical spin–flop phase with non-zero Z-projections for two pairs of sublattice spins (figure 1(b)). When the field increases, φ diminishes and goes to zero at $H = H_c^{(2)}$, where

$$H_c^{(2)} = 8JSp_c \qquad p_c = [1 + (\delta/a)(2-a^3)/(a-1)]^{-1/2}. \tag{5}$$

The subsequent reorientation occurs in exactly the same way as in I (figure 1(c)) with

$$\cos\alpha = p[1 - \delta(t+1)/t] \qquad \cos\beta = p[1 - \delta(t+1)/2] \tag{6}$$

where $t = \sin\alpha/\sin\beta$ is a real root of the equation

$$t(t-1)/(2-t^3) = \delta p^2/(1-p^2). \tag{6$'$}$$

At $H = H_c^{(2)}$, $t = a$, as follows from (6′), ensuring the coincidence between (3) with $\varphi = 0$ and (6), while for $\alpha, \beta \to 0$ one has $t = 2$ and, hence, $H = H_c^{(3)} = 8JS + 18J'S$ coinciding with the saturation field, as expected.

Note that for D not very close to $3J'$, $H_c^{(2)} \simeq 8JS$ and differs from $H_c^{(3)}$ only in J'-corrections. The second intermediate phase thus normally exists only in a narrow region close to the saturation field. When D increases, the width of the non-planar phase diminishes slightly, but the essential drop in $H_c^{(2)}$ occurs only in a very narrow region of D close to the critical value, D_c, when the non-planar phase disappears. Up to the second order in δ,

$$D_c = 3J'(1 - \delta^2/2). \tag{7}$$

When $(1 - D/D_c) \simeq \delta$, it follows from (5) that $H_c^{(2)} \simeq H_c^{(1)}$.

The longitudinal magnetization is mainly sensitive to the first-order transition, where it undergoes a finite jump. The exact classical expression at relatively low fields $H \ll JS$ is

$$M(H) = \frac{H}{8JS}\begin{cases}[1 + 2/(2-z)^2]/3 & H < H_c^{(1)} \\ 1 & H > H_c^{(2)}.\end{cases}$$

The peculiarities of the reorientation process also affect the AFMR frequencies. In particular, one can evidently expect softening at $H_c^{(2)}$ and $H_c^{(3)}$ as well as at the hysteresis-free first-order transition point, $H_c^{(1)}$. The calculations were performed in the same manner as in I, in the framework of a six-sublattice model. In the interests of brevity, we will simply give the results.

For $H < H_c^{(1)}$ the configuration is the same as in I, so six AFMR frequencies are in essence the solutions of (9)–(10) in I. For $q \ll 1$ the 'relativistic' (i.e. going to zero for $H = D = 0$) branches are disposed significantly below the exchange ones. In this limit one can simplify the equations for the low-energy modes and obtain $\omega_3 \simeq 4S\sqrt{JD}$, and ω_1 and ω_2 as the solutions of

$$(\omega^2 - \tilde{\omega}_1^2)(\omega^2 - \tilde{\omega}_2^2) = 2H^2\omega^2 \tag{8}$$

where $\tilde{\omega}_1^2 = 3H^6/4H_c^4$ and $\tilde{\omega}_2^2 = 16JDS^2 - H^2$.

For $H = H_c^{(1)}$, $\omega_1 = 0$ while $\omega_2^2 = 16JDS^2(2 + \frac{3}{4}q^2)$.

As usual, the first-order transition is accompanied by a discontinuity in the spectrum. For $H \geqslant H_c^{(1)}$ the equations resemble those for $H < H_c^{(1)}$ (equation (9) in I), but without the right-hand side. The result is

$$\begin{aligned}
\omega_1 &= H_c\left[\!\left[\frac{3}{2(2-q)}\left\{1 + \frac{2q}{3(2-q)} - \left[\left(1 + \frac{2q}{3(2-q)}\right)^2 - \frac{4}{9}q^3\frac{(3-q)^2}{2-q}\right]^{1/2}\right\}\right]\!\right]^{1/2} \\
\omega_2 &= (H^2 - 16JDS^2)^{1/2} \\
\omega_3 &= H \\
\omega_4 &= H_c\left[\!\left[\frac{3}{2(2-q)}\left\{1 + \frac{2q}{3(2-q)} + \left[\left(1 + \frac{2q}{3(2-q)}\right)^2 - \frac{4}{9}q^3\frac{(3-q)^2}{2-q}\right]^{1/2}\right\}\right]\!\right]^{1/2} \\
\omega_5 &= H_c[(1-q)(3-q)/(2-q)]^{1/2} \\
\omega_6 &= [H^2 + H_c^2(6 - 4q + q^2)/(2-q)]^{1/2}.
\end{aligned} \tag{9}$$

For low enough D, in passing through $H_c^{(1)}$ ω_1 and ω_2 jump to $\Delta\omega_1 \simeq (12JDS^2q^2)^{1/2}$ and $\Delta\omega_2 \simeq -(32JDS^2)^{1/2}$, while ω_3 remains practically unchanged.

The low-H field dispersion of AFMR frequencies for $q = 0.5$ and $H_c = 64$ kOe is shown in figure 3.

The case of high fields is rather methodological since we cannot really hope for experimental verification. However, for the sake of completeness we report the results for the AFMR modes in this region. Three of them—the continuations of two relativistic, (ω_2 and ω_3) and one exchange (ω_6) mode grow linearly with the field, while the others

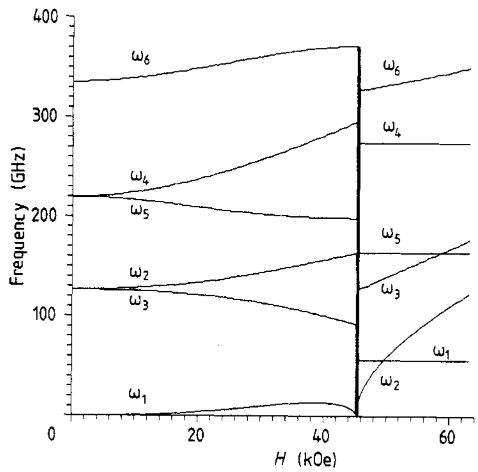

Figure 3. Field dispersion of AFMR frequencies for $q = 0.5$ and $H_c = 64$ kOe.

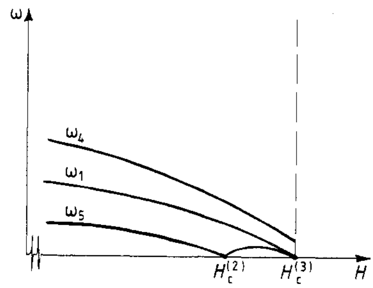

Figure 4. Field dispersion of the low-energy AFMR modes near saturation.

diminish in comparison with their values at $H = H_c^{(1)}$ and prove to be of the order of $J'S$ for $H \simeq 8JS$. The instability of the non-planar phase for $H = H_c^{(2)}$ is governed by ω_5:

$$\omega_5^2 = 4S^2\{[R + 3J'(a-2)\cos^2\varphi](3J'a\sin^2\varphi + Q) - QR\} \tag{10}$$

where φ is given by (3), while

$$R = 4J\{1 - p^2[1 - \delta(1+a)]\}/\cos^2\varphi \qquad Q = 4Jp^2[1 - \delta(1+a)]\sin^2\varphi. \tag{10$'$}$$

For $H \ll JS$ we return to (9), while for $H \gg H_c^{(2)}$ it follows from (10, 10′) that $\omega_5 \to 0$. Calculations were also made for the other two low-energy modes, ω_1 and ω_4, but the expressions are too cumbersome to be presented here.

At least for $H_c^{(2)} < H < H_c^{(3)}$ we again deal with the planar configuration. In this phase

$$\omega_5^2 = 4S^2[R + 3J'(t-2)][3J'(t-a)] \tag{11}$$

where t was introduced in (6′) and R is the same as in (10′) with $\varphi = 0$ and t substituted for a. Evidently, ω_5 softens on both phase boundaries, at $H_c^{(2)}$ as well as at $H_c^{(3)}$. At the saturation field one also has

$$\omega_1 = 0 \qquad \omega_4 = 2S[9J'(D + 9J')]^{1/2} \tag{12}$$

in agreement with the calculations within the one-sublattice model in a paramagnetic phase. The field dispersions of the low-energy AFMR frequencies near the saturation are shown in figure 4.

In conclusion, we have investigated the spin reorientation in weakly anisotropic quasi-one-dimensional antiferromagnets. The process was shown to be accompanied by two phase transitions of first and second order with a non-planar spin arrangement in the intermediate fields.

The condition $D < 3J'$ is apparently fulfilled in the vanadium compounds $CsVX_3$ (X = Cl, Br, I) [12]. Although $H_c^{(1)}$ is expected to be rather high ($H_c^{(1)} \simeq 165$ kOe for $CsVBr_3$ and 180 kOe for $CsVCl_3$), we still believe it to be possible to verify the scenario proposed by the investigation of the low-H dispersions of the AFMR frequencies.

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