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1991 J. Phys.: Condens. Matter 3 69

(http://iopscience.iop.org/0953-8984/3/1/005)

J. Phys.: Condens. Matter 3 (1991) 69–82. Printed in the UK

Quantum theory of an antiferromagnet on a triangular lattice in a magnetic field

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Received 9 February 1990

Abstract. The reorientation process in a magnetic field in two-dimensional isotropic and *XY* quantum Heisenberg antiferromagnets is shown to occur through the intermediate phase with unbroken continuous symmetry and constant magnetization equal to one third of the saturation value. The same reorientation process is also found in the more complicated classical models.

1. Introduction

In the last few years there has been renewed interest in the study of the possibility for the disordered ground state to occur in $S = 1/2$ two dimensional (2D) antiferromagnets on a triangular lattice (AFMT) due to strong zero-point vibrations [1–3]. Strictly speaking, the exact answer whether this is possible or not is not yet known, but most investigators now believe that long-range order, though strongly suppressed by fluctuations, does exist in AFMT even for sufficiently small values of the site spin [4, 5].

In this paper we also study 2D AFMT but we carry out our investigations in the presence of an external magnetic field. Interest in this problem stems from the fact that at $T = 0$ switching on the magnetic field does not change the degree of continuous degeneracy in the classical Heisenberg model [6]. To see how this happens let us consider first the situation in three dimensions (3D) (i.e. in the so-called antiferromagnet on a stacked triangular lattice). Here the spin arrangement in the presence of the field is well established: it is of the 'umbrella' type and the order parameter space is reduced from SO(3) in the zero-field case to $SO(2) \times Z_2$ for $H \neq 0$ (SO(2) corresponds to a rotational symmetry around the magnetic-field axis and Z_2 distinguishes left- and right-twisted 120° helicoids formed by the components perpendicular to the field). Due to the non-collinearity of the spin structure the excitation spectra contains three low-energy modes [7]: a Goldstone mode with $\omega_1 \sim |k|$ associated with the breaking of SO(2), a mode with $\omega_2(k = 0) \equiv 2\mu H$, representing the precession of the total magnetic moment around the field direction, and the third mode with energy $\omega_3 = \eta 2\mu H$, where $\eta = (\chi_\perp - \chi_\parallel)/\chi_\parallel$ is the anisotropy of susceptibilities. In the classical limit ($S \to \infty$) η can be expressed in terms of the microscopic parameters as follows [8]:

$$\eta = (1 + \tfrac{9}{4}J/J'')^{-1} \tag{1}$$

where J and J'' are in-plane and interplane exchange integrals. One can immediately

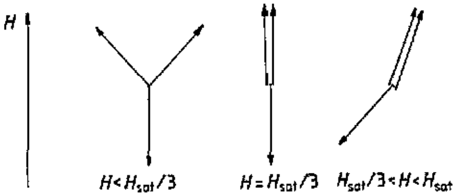

Figure 1. Reorientation process in the magnetic field in 2D Heisenberg AFM on a triangular lattice. Zero-point fluctuations stabilize the collinear phase in the finite region $H_1 < H < H_2$ in the vicinity of $H_{sat}/3$.

see that in the purely 2D case ($J'' = 0$), $\eta = 0$, that is, in addition to the Goldstone mode associated with the SO(2) breaking there exists an 'accidental' gapless branch of excitations. This is of course a reflection of the fact that the Heisenberg interaction for the triad of classical spins can be expressed only in terms of magnetization vector

$$S_1 \cdot S_2 + S_2 \cdot S_3 + S_3 \cdot S_1 = \tfrac{1}{2}M^2 - \text{constant} \qquad M = S_1 + S_2 + S_3$$

without drawing any of the antiferromagnetic vectors. According to simple calculations, ω_3 remains gapless in all fields up to the saturation value ($H_{sat} = 18JS$). Moreover, in non-zero fields this mode turns out to be quadratic in k (the state of a triad of classical unit vectors is specified to an accuracy of the rotation around the field axis by three equations for five angles). As a result, many exotic configurations have classically the same ground state energy as the umbrella-like one and, hence, the type of reorientation at $T = 0$ in a real quantum Heisenberg model must be selected by quantum fluctuations.

The same is also true for easy-plane systems, and the *XY* model serves here as a good example [9–12]. The order parameter space must normally be reduced from SO(2) $\times$ Z_2 in the zero-field case to $Z_2 \times Z_3$ in the presence of a magnetic field [9]. However, for classical spins the Goldstone mode, associated with SO(2) breaking in a zero field, does not acquire a gap in all fields up to the saturation value (again, the ground state of a triad of two-component unit vectors is completely specified by two equations for three angles). One way to lift the 'accidental' degeneracy in the *XY* model was proposed in [9, 12]. It was shown that at non-zero temperature the 'lacking at $T = 0$' condition for the angles arises thus fixing the mode of reorientation. For $T \to 0$ it occurs in conformity with figure 1 and is accompanied by a phase transition at $H = H_{sat}/3$ when the spins of the two sublattices align parallel to each other. Moreover, numerical experiments [9] indicate that at non-zero temperatures the collinear phase survives in the finite range of magnetic fields. This is very natural since in spite of the fact that the order parameter space is the same ($Z_2 \times Z_3$) in low and high-field phases, it is easy to determine the parameter distinguishing between them. This is a chirality vector $\mathcal{K}$, which for each elementary spin triangle is a measure of proximity to a 120° structure:

$$\mathcal{K} = (2/3\sqrt{3})(S_1 \times S_2 + S_2 \times S_3 + S_3 \times S_1). \tag{2}$$

Evidently, $\mathcal{K} \equiv 0$ in the high-field phase when the spins of two sublattices are parallel.

Less information is known about the isotropic system, where the degeneracy of the ground state is much stronger. Kawamura and Miyashita proposed [6] that at $T \neq 0$ the process of reorientation for classical spins occurs in the same way as in the *XY* system, that is, firstly, in the presence of the field all the spins remain in the same plane and, secondly, the reorientation occurs via the intermediate collinear phase with unbroken continuous symmetry. Numerical calculations [6] seem to confirm this scheme.

The aim of the present paper is firstly to show that quantum fluctuations also remove the 'accidental' degeneracy both in Heisenberg and *XY* models and select the same type of reorientation as do the temperature fluctuations. The isotropic Heisenberg model will be considered in section 2 and the results for the XY model will be reported in section 3. Note that in contrast to the case of non-zero temperature, the analysis of the zero-point motion effects is performed analytically on the basis of $1/S$ expansion. We also note that this is not the only case when quantum fluctuations lift the accidental degeneracy existing on the classical level. The same holds in a number of systems with competition between nearest and next-nearest neighbours exchange interactions [13–15]. Finally, we would like to mention that here one meets the phenomenon which Villain *et al* [16] termed 'order from disorder', since if there were no gap generation then the ordering in the ground state would be destroyed by quantum fluctuations due to the existence of the quadratic in k mode in the bare spectrum.

Evidently, the 'accidental' degeneracy is to a great extent a peculiarity of the Heisenberg model. The complication of the model removes the degeneracy even for classical systems. In the exchange approximation this is achieved (for $S > \frac{1}{2}$) by adding the biquadratic coupling [17]. An analogous effect occurs in the four-sublattice antiferromagnet UO_2 [18]. We discuss this in more detail in section 4. The important point is that though the accidental degeneracy is removed by biquadratic coupling, the umbrella-like configuration peculiar to 3D systems occurs only for one sign of this coupling, while for the other sign the non-trivial mode of reorientation as in figure 1, i.e. via the intermediate collinear phase, becomes energetically favourable. Hence, the adding of biquadratic coupling may lead to the same consequences as the account of quantum fluctuations.

The other way to remove the degeneracy (also available only for $S > \frac{1}{2}$) is to add a single-ion anisotropy of the easy-axis type [19]. It is remarkable that this additional interaction also favours a planar arrangement and reorientation via an intermediate collinear phase; but now, due to the fact that, in the presence of the easy-axis anisotropy, reorientation always (even in three dimensions) starts from the planar arrangement (antiferromagnetic phase [20]), the peculiarity of the 2D case reveals in the formal divergency of the spin-flop field value [8]

$$H_{cr}^2 = 16J''DS^2(1 + \tfrac{9}{4}J/J'')(1 + \tfrac{9}{8}J/J'') \tag{3}$$

that is, the antiferromagnetic (planar) phase survives in all fields. The role of easy-axis anisotropy is also considered in more detail in section 4.

A brief comparison with experiment is given in section 5.

A short version of the paper was published in [21].

2. Isotropic case

We start with an isotropic Heisenberg antiferromagnetic on a triangular lattice:

$$\mathcal{H} = J\sum_{l,\Delta} S_l S_{l+\Delta} - 2\mu H \sum_l S_l^z. \tag{4}$$

We presume that the classical 120° structure in zero field is not destroyed by quantum fluctuations and we will explore a traditional spin wave approach, based on the $1/S$ expansion†. The type of lattice requires us to introduce three bosonic fields. Doing this

† Numerically, the reduction of the sublattice magnetization in the leading order in $1/S$ is $\langle S\rangle/S = 1 - 0.26/S$.

with the help of the Holstein–Primakoff transformation we obtain the bosonic version of the spin Hamiltonian.

We shall not present the calculations step by step since they are absolutely standard though very cumbersome. In all cases the problem actually was to find the canonical transformation diagonalizing the quadratic form for three bosonic fields. The corresponding transformation at zero field is presented as an example in appendix 1. In the following we shall instead list only the results.

Our first aim is to establish the sign of the anisotropy of susceptibilities η since it determines the type of arrangement (umbrella-like or planar) in low fields. Direct calculations lead to the following result in the leading order in $1/S$:

$$\eta = \frac{\chi_\perp - \chi_\parallel}{\chi_\parallel} \simeq \frac{2}{S}\frac{1}{N}\sum_k{}' \left(\frac{(1+f_k-\varepsilon_k)f_k}{\varepsilon_k} - \frac{3f_k^2}{\varepsilon_k^+ + \varepsilon_k^-}\frac{(1+4f_k^+)(1+4f_k^-)}{\varepsilon_k^+\varepsilon_k^-}\right) \tag{5}$$

where

$$\varepsilon_k^\pm = \varepsilon_{k\pm 2\pi/3} \qquad f_k^\pm = f_{k\pm 2\pi/3} \qquad \varepsilon_k = ((1+4f_k)(1-2f_k))^{1/2}$$

$$f_k = \tfrac{1}{4}(\nu_k + \nu_{-k}) \qquad \nu_k = \tfrac{1}{3}\{\exp(\mathrm{i}k_x) + \exp(\mathrm{i}k_y) + \exp[-\mathrm{i}(k_x+k_y)]\}.$$

Here and below the 'prime' symbol at Σ indicates that the summation is performed over the whole Brillouin zone ($\Sigma_k' 1 = 3\Sigma_k 1 = N$, where N is the total number of spins), and the X and Y axes are directed towards the nearest neighbours ($\boldsymbol{n}_x \cdot \boldsymbol{n}_y = -\frac{1}{2}$). Transformation to the extended zone scheme is possible since at $H = 0$ the spectrum has no gaps at the zone boundaries.

Numerical calculations give

$$\eta \approx -0.08/S. \tag{6}$$

The minus sign means that at least at low fields quantum fluctuations select the planar arrangement (though the configuration inside the plane is not yet specified). In order to exclude definitely the possibility of an umbrella-like ground state we calculated and compared the first quantum corrections to the ground state energies for umbrella-like and planar (as in figure 1) configurations at $H = H_{\mathrm{sat}}/3 = 6JS$ (for trivial reasons quantum fluctuations do not renormalize the saturation field value). The results is

$$E_{\mathrm{planar}} - E_{\mathrm{umbrella}} = -NJS^2Q \tag{7}$$

where

$$Q \approx 2\frac{3}{NS}\sum_k \frac{\lambda_k}{(\varepsilon_1+\varepsilon_3)(\varepsilon_2+\varepsilon_3)} - 3\frac{1}{NS}\sum_k{}' [1 + \tfrac{2}{3}f_k - (1 + \tfrac{4}{3}f_k - \tfrac{20}{3}f_k^2)^{1/2}] \tag{7$'$}$$

$$\lambda_k = -4 + 10|\nu_k|^2 + 2\varepsilon_3|\nu_k|^2 + 4\varepsilon_3^2 - 3[\nu_k^3 + (\nu_k^*)^3]$$

and ε_i are the (positive) classical frequencies of three branches of excitations above the collinear state in figure 1. The values ε_1, ε_2 and $-\varepsilon_3$ are the roots of the following equation:

$$\varepsilon^3 - \varepsilon^2 - \varepsilon(1 - |\nu_k|^2) + 1 - 3|\nu_k|^2 + \nu_k^3 + (\nu_k^*)^3 = 0. \tag{8}$$

The result of numerical calculation is

$$Q \approx 0.13/S$$

that is, a planar arrangement is energetically favourable also at $H = H_{\mathrm{sat}}/3$ and, hence most likely at all other fields.

The next step is to find the real in-plane arrangement. At low fields, the degree of freedom associated with the rotation of a triad of spins inside the plane remains massless if we restrict ourselves to the leading order in H, i.e. with energies, ΔE, of the order of H^2. Thus we have to compare the energies in the next order in a magnetic field. This was done for two configurations: those as in figure 1 and a second where one of the spins of a triad is parallel to the field direction. The result is that the first configuration is energetically favourable, the difference in energies being

$$\Delta E \propto (J/S)(H/J)^3. \tag{9}$$

Thus we tentatively conclude that quantum fluctuations select the same mode of reorientation as do the temperature fluctuations. Below we present another argument confirming this conclusion.

We anticipate that with ΔE as in equation (9) the gap in the low-energy mode, ω_3, will be proportional to $(|\eta|\Delta E/\chi_\parallel)^{1/2} \propto (J/S)(H/J)^{3/2}$ for a purely isotropic system.

Now we examine the situation near $H_{sat}/3$. Classically, the collinear configuration exists only at a single field value. Meanwhile, the transitions at $H \to H_{sat}/3$ from above and from below are of completely different nature and there are no reasons to expect both of them to occur at the same field value. The calculation of the lability points of low- and high-field phases as those where the renormalized values of the angles between the two spins of the triad and the magnetic field tend to zero confirms this suspicion: the low-field phase becomes unstable at

$$h = h_1 \simeq 1 - \frac{3}{NS}\sum_k \frac{(3-\varepsilon_3)|\nu_k|^2 - \frac{3}{2}(\nu_k^3 + \nu_{-k}^3)}{(\varepsilon_1+\varepsilon_3)(\varepsilon_2+\varepsilon_3)} \simeq 1 - 0.084/S \tag{10}$$

while the high-field phase loses stability at

$$h = h_2 \simeq 1 + \frac{3}{NS}\sum_k \frac{(1+\varepsilon_3)|\nu_k|^2 - \frac{1}{2}(\nu_k^3 + \nu_{-k}^3)}{(\varepsilon_1+\varepsilon_3)(\varepsilon_2+\varepsilon_3)} \simeq 1 + 0.215/S \tag{11}$$

(here and below $h \equiv 2\mu H/(6SJ) = 3H/H_{sat}$). As expected, $h_2 > h_1$. In the intermediate region, $h_1 < h < h_2$, the collinear configuration is stable. The absence of broken continuous symmetry then implies that all the excitations have a finite gap. Direct calculation of the spectrum with quantum corrections involved leads to the following result:

$$\omega_1/(6JS) \approx h - h_1 \qquad \omega_2/(6JS) \approx h \qquad \omega_3/(6JS) \approx h_2 - h. \tag{12}$$

Note that the mode associated with the precession of the magnetic moment is identically equal to $2\mu H$ also for a quantum system [22]. As found in equation (12), the collinear phase is really stable for $h_1 < h < h_2$, and the gaps obtained for two would-be Goldstone modes are proportional to $1/S$. The AFMR frequencies versus magnetic field are shown schematically in figure 2.

The absence of continuous degeneracy of the ground state probably implies that the magnetization, M_z, remains constant inside the collinear phase since the exchange Hamiltonian commutes with the z-projection of the total spin*. Classically, M_z equals one third of the saturation value at $h = 1$. We calculated the first quantum correction to M_z and ascertained that it equals zero, that is the magnetization retains its classical value,

* This is not an exact statement since the discrete R_3 degeneracy of the ground state remains and thus the renormalization of magnetization of the order of $\exp(-S)$ is not excluded in principle, but not expected in reality [7].

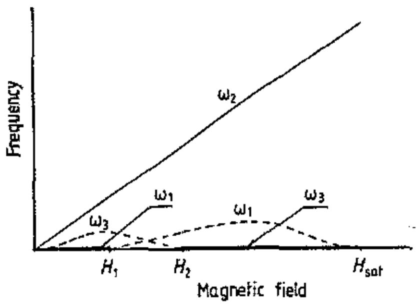

Figure 2. The anticipated behaviour of AFMR frequencies versus magnetic field in 2D Heisenberg AFM on a triangular lattice. The broken lines denote the branches which would be gapless in the classical treatment.

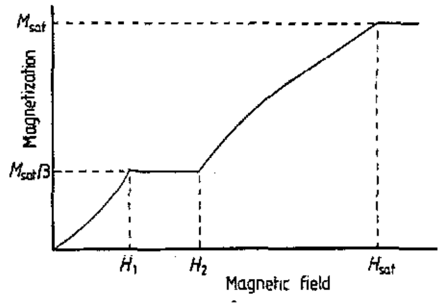

Figure 3. The anticipated behaviour of longitudinal magnetization in 2D Heisenberg AFM on a triangular lattice. The plateau on the magnetization curve results from the stabilization of the collinear phase in the finite region of magnetic fields due to zero-point motion.

$\mu S/3$, also in the quantum case. We believe this result to be true in all orders of perturbation in $1/S$. The anticipated behaviour of magnetization is shown in figure 3.

We conclude this section with a brief discussion of the more realistic quasi-2D system. For ferromagnetic interplane interaction (this is the case for a metamagnet) the mode of reorientation will evidently remain unchanged independently of the strength of interplane exchange. The case of antiferromagnetic interplane interaction, J'', is less trivial. Firstly, since all the excitations in the intermediate phase with constant magnetization do have finite gaps, this phase cannot be destroyed by small perturbations. In contrast, the situation in low fields is very sensitive to the switching of the new interactions and whatever small J'' is the reorientation will always start with a different planar configuration which is antiferromagnetic in the direction perpendicular to the plane [23, 24]. The transition to a 2D planar arrangement (which is ferromagnetic in the direction perpendicular to the plane) will occur when the gain in energy $\Delta E \sim (J/S)(H/J)^3$ favouring a metamagnetic configuration reaches the energy difference associated with the interplane exchange, $\Delta E \sim J''$, i.e. when $H \sim J((J''/J)S)^{1/3}$. Secondly, the increase in J'' will evidently change the sign of η and, hence, for not very small J'' the reorientation will start from the umbrella-like configuration. We cannot definitely answer the question what will happen in higher fields since it depends on the correlations between unknown numerical parameters†.

3. Easy-plane systems

As was pointed out in the introduction, the accidental degeneracy in the classical AFMT does not disappear when we switch on the anisotropy favouring the spin arrangement in the basal plane (now singled out initially and not as a result of spontaneous breaking of symmetry) and direct the field along the plane.

† The discussion of quasi-2D properties in section 3 in [21] was based on the proposal that η may be identically equal to zero in 2D AFMT. The direct calculation of η (see equation (6)) does not confirm this proposal.

For simplicity, we shall mainly restrict ourselves to the case of the XY model:

$$\mathcal{H} = J \sum_{l,\Delta} (S_l^+ S_{l+\Delta}^- + S_l^- S_{l+\Delta}^+) - 2\mu H \sum_l S_l^z. \tag{13}$$

We shall first assume that the reorientation again occurs in direct compliance with figure 1 and then verify this by calculating the spectrum.

The classical picture of excitations above the state in figure 1 is rather simple: in addition to the 'accidental' massless branch there are two excitations with finite gaps, one of them softens at the transition point $H = H_{sat}/3$. Trivial calculations give ($h \equiv 2\mu H/6JS = 3H/H_{sat}$)

$$\begin{aligned} &\omega_{1,2} = 3\sqrt{2}JS[3 \mp h(h+2)]^{1/2} \qquad \omega_3 = 0 \qquad 0 < h < 1 \\ &\omega_{2,3} = 3JS[3 \pm (\sqrt{2}/2)(h^4 - 10h^2 + 27)^{1/2}]^{1/2} \qquad \omega_1 = 0 \qquad 1 < h < 3. \end{aligned} \tag{14}$$

The analogous expressions for the case of AFMT with small single-ion anisotropy, closer to the experiment, are presented in appendix 2.

Our aim is to investigate the effect of zero-point vibrations. To do this we calculated in the first order in $1/S$ the lability points for the low- and high-field phases. The critical fields, h_1 and h_2 respectively, are again different, favouring the collinear ground state to occur in the finite region of the magnetic field:

$$\begin{aligned} h_1 &\approx 1 - \frac{1}{2NS}\sum_k{}' \left(\frac{2}{\varepsilon_k} - 1 - \varepsilon_k\right) \\ h_2 &\approx 1 + \frac{1}{2NS}\sum_k{}' (1 - \varepsilon_k) \\ h_2 - h_1 &\approx \frac{1}{NS}\sum_k{}' \left(\frac{1}{\varepsilon_k} - \varepsilon_k\right) \equiv \frac{1}{NS}\sum_k{}' \frac{(\varepsilon_k - 1)^2(\varepsilon_k + 1)}{\varepsilon_k} > 0. \end{aligned} \tag{15}$$

Here

$$\varepsilon_k = (1 + \nu_k + \nu_{-k})^{1/2} \qquad \left(\text{obviously } \frac{1}{NS}\sum_k{}' \varepsilon_k^2 = 1\right).$$

Numerically, $h_1 \approx 1 - 0.59/S$; $h_2 \approx 1 + 0.05/S$. Note, that in the XY case quantum fluctuations also renormalize h_{sat}: $h_{sat}/3 \approx 1 - 0.14/S$.

The same stability boundaries were determined by studing the AFMR modes above the collinear state. They happened to be positive, and soften separately at $h = h_1$ and h_2:

$$\omega_1 \approx 6JS(h - h_1)^{1/2} \qquad \omega_3 \approx (6JS/\sqrt{3})(h_2 - h)^{1/2}. \tag{16}$$

Correspondingly the lowest gaps in the collinear phase are proportional to $S^{-1/2}$. Note that at low fields the gap, picking out the configuration as in figure 1, will be for obvious reasons proportional to $(J/\sqrt{S})(H/J)^{3/2}$. The field dependencies of the AFMR frequencies are presented schematically in figure 4.

Classically, the magnetization in the collinear phase is constant and equals one third of the saturation value. In contrast to the Heisenberg system this is no longer true for the quantum XY case, since the Hamiltonian of the XY model, equation (13), does not commute with the X projection of the total spin.

Concluding this section we note that the planar case allows a simple explanation of the difference between phase transitions at $h = h_1$ and $h = h_2$. In fact, in both low- and high-field phases the order parameter space is $Z_2 \times Z_3$ (Z_2 disappears within the collinear phase), but the nature of the Z_2 element is quite different: in the low-field phase Z_2 reflects the breakdown of chiral symmetry while in high fields Z_2 arises as a result of the breakdown of the usual Ising symmetry. The difference in the realizations of the order parameter space evidently means that there are no physical reasons for both transitions to occur at the same field value.

4. Other spin models

In this section we will show that the 'accidental' degeneracy in a magnetic field is to a great extent a peculiarity of the Heisenberg model and that the unusual mode of reorientation may become energetically favourable at $T = 0$ even on the classical level, but for more general Hamiltonians. In order to demonstrate this we will at first restrict ourselves to a purely exchange system (see equation (4)), but will add the biquadratic nearest-neighbour interaction:

$$\delta\mathcal{H} = \frac{BJ}{S^2}\sum_{n,\Delta}(S_n S_{n+\Delta})^2. \tag{17}$$

Being of exchange origin, this term will not moderate the 120° structure in a zero field. Nevertheless, for a triad of spins (classical unit vectors) this term will give rise to the coupling between the vectors of ferro- and antiferromagnetism and, hence, lead to non-zero η:

$$\eta = 3B. \tag{18}$$

As seen from equation (18), the umbrella-like configuration is to be chosen for $B > 0$ while for negative B the planar arrangement becomes energetically favourable.

Determination of the arrangement within the plane demands comparison of the energies in the next to leading order in H. Doing this we ascertain that the (expected) configuration shown in figure 1 has a minimal energy for $B < 0$.

We therefore conclude that biquadratic coupling with negative B should lead to the same reorientation process as do quantum (or temperature [6, 9–12]) fluctuations, the role of $1/S$ (or $T/(JS)$) being played by $|B|$. For completeness, we present below the expressions for the critical fields, AFMR frequencies and longitudinal magnetization under the assumption that biquadratic coupling is small compared to the Heisenberg one ($B \ll 1$). Qualitatively, these expressions repeat those from section 2.

(i) The lability fields for low- and high-field phases are, correspondingly,

$$h_1 = 1 - 6|B| \qquad h_2 = 1 + 2|B| \tag{19}$$

while $h_{sat} = 3(1 - 2|B|)$. Note that $(h_1 + h_2)/2 = h_{sat}/3$.

(ii) The exchange nature of the interaction forces one of the AFMR frequencies to be identically equal to $2\mu H$, while the other two are given by the following expressions:

$$\left.\begin{aligned}\omega_1 &= 0\\ \omega_3 &\approx 6JS|B|h^{3/2}(h+3)^{3/2}\end{aligned}\right\}\qquad 0 < h < h_1 \tag{20a}$$

$$\left.\begin{aligned}\omega_1 &= 6JS(h - 1 + 6|B|)\\ \omega_3 &= 6JS(1 + 2|B| - h)\end{aligned}\right\}\qquad h_1 < h < h_2 \tag{20b}$$

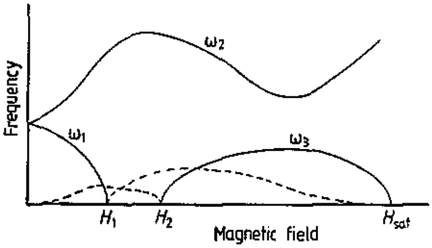

Figure 4. The same as figure 2 but for the *XY* model. The broken lines denote the branches which would be gapless in the classical treatment.

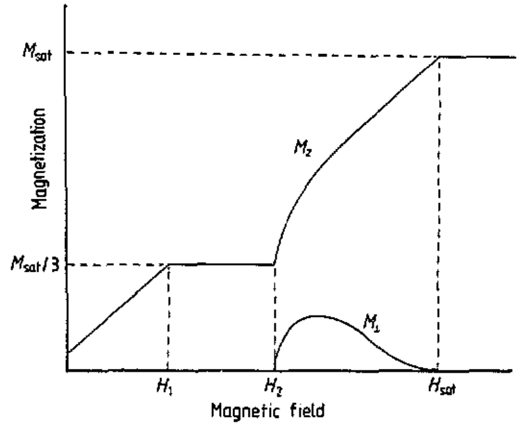

Figure 5. The field dependence of the longitudinal and transverse magnetizations in 2D classical Heisenberg AFM with easy-axis anisotropy on the triangular lattice (see equations (27) and (28)).

$$\left.\begin{aligned}\omega_1 &\approx 6JS|B|[(h_{\text{sat}}^2 - h^2)/2]^{3/2} \\ \omega_3 &= 0\end{aligned}\right\} \qquad h_2 < h < h_{\text{sat}}. \tag{20c}$$

The field dependencies given by equation (20) are similar to those presented in figure 2.

(iii) The longitudinal magnetization is evidently constant and equal to $2\mu S/3$ within the collinear phase. Outside this region it behaves as follows:

$$M_{\parallel} \simeq 2\mu \frac{Sh}{3} \begin{cases} 1 + |B|(h+1)(h+2) & 0 < h < h_1 \\ 1 + (|B|/2)(h^2 - 5) & h_2 < h < h_{\text{sat}} \\ 3 & h > h_{\text{sat}}. \end{cases} \tag{21}$$

Note, that complementary to the analysis of quantum effects the engaging of the however small easy-plane anisotropy immediately changes the dependence of AFMR frequencies on $|B|$: at extremely low fields ω_3 becomes proportional to $|B|^{1/2}h^{3/2}$ instead of $|B|h^{3/2}$ as in equation (20).

The other way to lift a degeneracy is to engage the single-ion anisotropy of easy-axis type

$$\delta\mathcal{H} = -D\sum_l (S_l^z)^2 \qquad D > 0. \tag{22}$$

This term breaks the 120° structure in a zero field and normally must produce a spin-flop field $H_c \sim (DJ)^{1/2}$, that is, the reorientation starts from a planar configuration and then there would be a transition into the umbrella-like structure [20]. The peculiarity of the 2D case, already mentioned in the introduction, is that since η reduces to zero, the calculated value of this field formally tends to infinity (see equation (3)) and as a result the antiferromagnetic (planar) phase with the spin arrangement as in figure 1 survives in all fields up to the saturation value [19]. The different nature of the transitions when approaching the collinear configuration from above and from below (though in both cases the order parameter space is isomorphic to $SO(2) \times Z_3$) is again apparent in a finite stability region of the intermediate phase with only discrete symmetry Z_3 broken [19]:

$$h_1 = 1 - 2\tilde{D} \qquad h_2 \approx 1 + 6\tilde{D}, \tag{23}$$

while $h_{sat} \simeq 3(1 - \frac{4}{3}\tilde{D})$. Here the role of $1/S$ is played by the dimensionless ratio $\tilde{D} = D[1 - 1/(2S)]/(6J)$. We propose that $\tilde{D} \ll 1$.

As usual, the AFMR frequencies are adapted to the non-zero $h_2 - h_1$. For $h < h_1$, $\omega_1 = 0$ reflecting the trivial invariance with respect to the rotation about the magnetic field axis, while

$$\omega_2 \simeq 6JS(h^2 + 3\tilde{D}(1 + h)^2)^{1/2} \tag{24a}$$

and the third non-zero frequency, the analogue of quantum gap, is given by the following expression:

$$\omega_3 \simeq 6JS\left(\frac{h\tilde{D}^2(h+3)^3 + 54\tilde{D}^4}{h^2 + 3\tilde{D}(h+1)^2}\right)^{1/2}. \tag{24b}$$

In the limiting cases equation (24b) becomes:

$$\omega_3 \simeq 6JS\begin{cases} 3\sqrt{2}\tilde{D}^{3/2}(1 + \frac{1}{4}h/\tilde{D}^2) & h \ll \tilde{D}^2 \\ 3(h\tilde{D})^{1/2} & \tilde{D}^2 \ll h \ll \tilde{D}^{1/2} \\ \tilde{D}(h+3)^{3/2}/h^{1/2} & h \gg \tilde{D}^{1/2}. \end{cases} \tag{24c}$$

At zero field $\omega_3 \propto \tilde{D}^{3/2}$. An analogous result has been obtained in the opposite case of quasi-1D AFMT [8, 19, 25]. In the 2D case the $\tilde{D}^{3/2}$ dependence was found in [26] though with different coefficient. The source of the discrepancy is not known to us.

Note that though the spin-flop field formally tends to infinity, the region $\mu H \sim S(DJ)^{1/2}$ is singled out as the ω_3 value passes through a maximum, $\omega_3 \sim 6SJ\tilde{D}^{3/4}$, at these fields.

For $h_1 < h < h_2$ the AFMR frequencies are practically the same as in the isotropic quantum case:

$$\omega_1 \simeq 6JS(h - h_1) \qquad \omega_2 \simeq 6JSh \qquad \omega_3 \simeq 6JS(h_2 - h). \tag{25}$$

They all have finite gaps since the order parameter has no transverse components.

At least for $h_{sat} > h > h_2$, the analogue of the quantum gap is maintained in the ω_1 mode, while the ω_3 excitation turns out to be massless:

$$\omega_1 \approx 6JS\tilde{D}(h_{sat}^2 - h^2)^{3/2}/(2\sqrt{2}h^2) \qquad \omega_2 \approx 6JSh \qquad \omega_3 = 0. \tag{26}$$

Evidently, the longitudinal magnetization remains constant and equal to one third of the saturation value in the intermediate region between h_1 and h_2. Outside this region it behaves as follows:

$$M_\parallel \simeq 2\mu\frac{S}{3}\begin{cases} h + \tilde{D}(1+h) & 0 < h < h_1 \\ h + (3\tilde{D}/4h^3)(h^4 - 9) & h_2 < h < h_{sat}. \end{cases} \tag{27}$$

The peculiarity of the easy-axis case is that in addition to the external field there is a non-zero internal anisotropy field. This field violates the condition $\sin\alpha = 2\sin\beta$ for the high-field phase in figure 1 and leads to a non-zero transverse magnetization for $h > h_2$:

$$M_\perp \approx 2\mu S(9\tilde{D}/4)[(h/h_2)^2 - 1]^{1/2}[1 - (h/h_{sat})^2]^{3/2}(h_{sat}/(3h)^3. \qquad (28)$$

Note that $M_\perp \propto (h - h_2)^{1/2}$ for $h \gtrsim h_2$ and $\propto (h_{sat} - h)^{3/2}$ for $h \lesssim h_{sat}$. The field dependence of magnetization is presented in figure 5.

We mention that the difference of 'susceptibilities' with respect to the easy-axis anisotropy may be regarded as a measure of the difference between low- and high-field non-collinear phases.

5. Comparison with experiment

In this section we wish to discuss briefly the possibilities of observing the unusual reorientation process discussed in this paper. A relatively large number of substances are known which with high accuracy can be regarded as quasi-2D AFMT [27, 28]. Among them two vanadium compounds, VCl_2 ($T_N = 36$ K) and VBr_2 ($T_N = 28.5$ K), are the most well known [29–33]. Susceptibility measurements [30] have shown that the transverse and longitudinal susceptibilities are practically equal to each other, that is, η is actually very small. By experimental estimations, in both substances $J'' = 0.2$ K [31] while the in-plane exchange is 23 K in VCl_2 and 16 K in VBr_2 [32]. The anisotropy constant (measured through the anisotropy of the g-factor in the paramagnetic state) was predicted to be negative (i.e. of easy-axis type) and of the order of 0.1 K [31].

Unfortunately, large in-plane exchange (peculiar for vanadium compounds) forces the saturation fields to be of the order of 10^2 T, which makes observation of the plateau on the field dependence of longitudinal magnetization very problematical.

Nevertheless, useful information can be obtained from AFMR measurements in relatively low fields. In both substances the low-energy mode at zero field was observed at $\omega = 9.1$ GHz for $T = 1.7$ K (VBr_2) and $T = 2.6$ K (VCl_2) [30, 33]. Meanwhile, according to calculations with the given values of $\tilde{D}$ and J (the latter is usually known to very high accuracy from neutron measurements), the lowest mode had to be placed at $\omega =$ 0.5 GHz. An attempt to resolve this discrepancy was undertaken in [26]: in addition to the increase of $\tilde{D}$ value it was proposed that a superfine interaction also plays an essential role. We propose to verify whether the low-lying mode is really ω_3 and also to determine the anisotropy constant by studying the field dependence of this mode since, as found in equation (24*b*), this mode increases with the field at low fields and passes through a maximum at $\mu H \sim 6JS\tilde{D}^{1/2}$.

Fortunately, a quasi-2D substance with a rather low saturation field is known. This is the intercalation compound C_6Eu [34, 35]. This substance is a metamagnet (the interplane exchange is ferromagnetic) and thus for our aims the smallness of the inter- and in-plane exchange integral, though it exists in practice due to the difference in the interatomic spacings, is not of great importance. Suematsu *et al* [35] have investigated the magnetization versus magnetic field in fields up to 25 T for two mutually orthogonal field orientations. For one of the orientations the magnetization increased more or less monotonically with the magnetic field while for the other they found a plateau on the magnetization curve which tended to be increasingly noticeable with decreasing temperatures. For the convenience of readers the experimental curves of [35] are

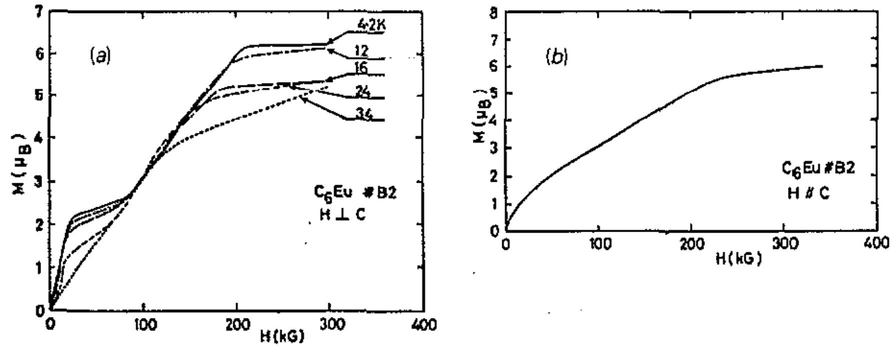

Figure 6. Magnetization curves of C_6Eu: (*a*) $H \perp c$ (for various temperatures); (*b*) $H \parallel c$ (T = 4.2 K). The figure is taken from Suematsu *et al* [35].

presented in figure 6. This is exactly the situation in the easy-plane AFMT (section 3) with the easy-plane anisotropy strong enough to destroy the plateau for the field directed perpendicularly to the plane (strictly speaking, traces of the plateau can be seen on the experimental curves even in this case). We therefore conclude that C_6Eu serves as an experimental example of 2D AFMT with the unusual mode of reorientation discussed in this article. The intermediate region of constant magnetization (which with good accuracy equals one third of the saturation value 7μ) was estimated in [35] as an interval 22 kG $< H <$ 82 kG, while the saturation fields were 240 kG and 205 kG for H perpendicular and parallel to the plane, respectively [35]. We cannot definitely answer what mechanism (quantum or classical) is responsible for the plateau, but since the spin value is rather high, $S = \frac{7}{2}$, the mode of reorientation through the intermediate phase seems to be mainly singled out by classical effects: biquadratic (or possibly even more exotic permitted for $S = \frac{7}{2}$) exchange interaction. We hope that AFMR experiments may clarify the situation.

6. Summary

We have found that spin reorientations at $T = 0$ in the external magnetic field in quasi-2D antiferromagnets on a triangular lattice may occur in an unusual way via an intermediate collinear phase with the constant longitudinal magnetization equal to one third of the saturation value. For quasi-2D Heisenberg and XY antiferromagnets this mode of reorientation is singled out by quantum fluctuations, while for more complicated Hamiltonians this may already be the case for classical spins. The reorientation via intermediate phase with a plateau on the $M_{\parallel}(H)$ curve was detected experimentally.

Acknowledgments

The authors would like to thank M I Kaganov, R Z Levitin, V I Marchenko and S E Korshunov for helpful discussions.

Appendix 1

At zero field the bosonic version of the spin Hamiltonian for isotropic 2D AFM on the triangular lattice is as follows:

$$\mathcal{H}/6SJ = E_0 + \sum_k \{a_k^+ a_k + b_k^+ b_k + c_k^+ c_k + \tfrac{1}{4}[(a_k^+ b_k + c_k^+ a_k + b_k^+ c_k)\nu_{-k} + (a_k^+ c_k + c_k^+ b_k + b_k^+ a_k)\nu_k] - \tfrac{3}{4}[(a_k^+ b_{-k}^+ + b_k^+ c_{-k}^+ + c_k^+ a_{-k}^+)\nu_{-k} + (a_k b_{-k} + b_k c_{-k} + c_k a_{-k})\nu_k]\} + \ldots \quad \text{(A1)}$$

where the dots stand for the anharmonic terms. The canonical transformation diagonalizing the quadratic form of equation (A1) is:

$$a_k = (1/\sqrt{3})(p_k - q_k - \mathrm{i}y_k r_k)$$
$$b_k = (1/\sqrt{3})\{p_k - \exp[-(2\pi/3)\mathrm{i}y_k]q_k - \mathrm{i}y_k \exp[(2\pi/3)\mathrm{i}y_k]r_k\} \quad \text{(A2)}$$
$$c_k = (1/\sqrt{3})(p_k - \exp[(2\pi/3)\mathrm{i}y_k]q_k - \mathrm{i}y_k \exp[-(2\pi/3)\mathrm{i}y_k]r_k)$$

where p_k, q_k and r_k are new bosonic fields and $y_k = \mathrm{sign}(\mathrm{Im}\,\nu_k)$. Knowledge of the transformation allows calculation of the first quantum corrections to the ground state energies for umbrella-like and planar configurations, that is, determination of η (see equation (5)).

Appendix 2

The classical AFMR frequencies for the 2D AFMT with single-ion easy-plane anisotropy, $D(S_i^z)^2$ $(D > 0)$, are the following $[\tilde{D} = D(1 - 1/(2S))/(6J),\ h = 2\mu H/(6JS)]$: for $0 < h < 1$,

$$\omega_1 = 6JS[\tilde{D}(3 - 2h - h^2)]^{1/2}$$
$$\omega_2 = 6JS[\tilde{D}(3 + 2h + h^2) + h^2]^{1/2} \qquad \omega_3 = 0$$

while for $1 < h < 3$ ($h = 3$ is the saturation value) and $\tilde{D} \ll 1$,

$$\omega_1 = 0$$
$$\omega_2 \approx 6JS[h^2 + (\tilde{D}/16h^2)(h^6 - 3h^4 + 35h^2 + 63)]^{1/2}$$
$$\omega_3 \approx 6JS[\tilde{D}(9 - h^2)(h^2 - 1)(h^2 + 7)/(16h^2)]^{1/2}.$$

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