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Abstract. An instability of a ferromagnetic state in quantum spin systems induced by a two-particle bound-state condensation is shown to produce, for $S > 2$, a ferrimagnetic ground state with a quantized ($S_z = S - 1$) value of spontaneous magnetization per site. For low S, instead, the ground state below the instability has zero net magnetization, while a spontaneous symmetry breaking reveals itself in quadratic, cubic, or both quadratic and cubic correlations for $S = 1, 2$ or $\frac{3}{2}$, respectively. The role of quantum fluctuations and the reorientation process in a magnetic field are examined.

1. Introduction

In the past few years there has been considerable interest in the study of unusual ground states in quantum isotropic magnetic systems with polynomial exchange interaction between nearest neighbours. For example, it was shown [1–4] that the transition from ferro- to antiferromagnetic ordering in the generalized spin $S = 1$ model may occur via the intermediate so-called spin nematic phase with unbroken time-reversal symmetry, but with spontaneously broken symmetry with respect to quadrupolar correlations $(\langle S_x^2\rangle = \langle S_y^2\rangle \neq \langle S_z^2\rangle, \langle S\rangle = 0)$.

The generic spin S exchange Hamiltonian is a polynomial of order $2S$,

$$H = \sum_{l,\Delta} \sum_{n=1}^{2S} J_n \, (S_l \cdot S_{l+\Delta})^n \tag{1}$$

where all the exchange integrals J_n are generally of the same order of magnitude. The general phase diagram in a $(2S - 1)$-dimensional parameter space involves different phases. One of them is evidently a ferromagnetic phase. For trivial reasons, it definitely realizes a ground state when the energy of a separate pair of spins $E_{\tilde S}$ has a minimum for a total spin $\tilde S$ equal to $2S$. We wish to study what phase replaces a ferromagnetic one when the minimum of $E_{\tilde S}$ shifts from $\tilde S = 2S$.

Strictly speaking, the crossing of the energy levels for a separate pair of spins is a necessary but not a sufficient condition for the replacement to occur. However, in this paper we will restrict ourselves to the case when the crossing first occurs between E_{2S}

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and the energy level E_{2S-2} corresponding to a neighbouring set of symmetrical configurations of a separate pair of spins. The equality $E_{2S} = E_{2S-2}$ is known [5] to be exactly a condition for the two-particle bound states to undergo softening at $k = 2\pi$, and thus the level crossing really produces the change in the ground state.

Moreover, as the replacement occurs via a hysteresis-free first-order phase transition [1–4], the new ground-state wavefunction can be recognized exactly at the critical line in parameter space, where there is no zero-point motion at all. The problem however is to select among a whole family of states minimizing the energy at the first-order transition line a true one which will determine the ground state immediately below this line. Quantum fluctuations can then be included by constructing a perturbation theory with the small parameter indicating the closeness to the transition [3, 4].

We begin with a simple exercise in a spin algebra with the aim of finding the most probable ground-state candidates for various spin values. Let us consider a separate pair of spins S with isotropic polynomial interaction. With the condition $E_{2S} = E_{2S-2}$, all the symmetrical configurations $\Psi_{1,2}$ with $|\bar{S}_z| > 2S - 4$ are ground states. Once the aim is to find a macroscopic wavefunction, we are forced to restrict ourselves to only those $\Psi_{1,2}$ which can be represented as a product of the single-site wavefunctions

$$\Psi_{1,2} = \bar{\Psi}_1 \cdot \bar{\Psi}_2 \tag{2}$$

(the total Ψ is then given by $\Psi = \Pi_l \bar{\Psi}_l$).

For $S > 2$, $\bar{\Psi}_{1,2}$ does not mix the states with positive and negative $\bar{S}_z$ and it is thus possible to restrict ourselves to only one sign of $\bar{S}_z$, say $\bar{S}_z > 0$. The required $\Psi_{1,2}$ can easily be constructed. It involves a one-parameter family of $\bar{\Psi}$:

$$\bar{\Psi} = (|S\rangle + j|S-1\rangle)/(1+j^2)^{1/2} \tag{3}$$

with j arbitrary.

For evident reasons, it is likely that the state furthest from the ferromagnetic one is the ground state immediately below the critical line. For $S > 2$ this state proves to be a quantum *ferrimagnet* with $\bar{\Psi}_i \equiv |S-1\rangle$ (and hence $\langle S_i^z\rangle = S-1$, $\langle S_i^+\rangle = \langle S_i^-\rangle = 0$).

For $S \leqslant 2$ the number of candidates for $\bar{\Psi}$ increases. For $S = 2$ in addition to

$$\bar{\Psi}^{(1)} = (|2\rangle + j|1\rangle)/(1+j^2)^{1/2} \qquad \text{or} \qquad \bar{\Psi}^{(2)} = (|-2\rangle + j|-1\rangle)/(1+j^2)^{1/2} \tag{4}$$

one has

$$\bar{\Psi}^{(3)} = (|2\rangle + \lambda|-1\rangle)/(1+\lambda^2)^{1/2} \qquad \text{and} \qquad \bar{\Psi}^{(4)} = (|-2\rangle + \lambda|1\rangle)/(1+\lambda)^{1/2}. \tag{5}$$

The first two functions describe magnetically ordered states ($\langle S_i^z\rangle \neq 0$) and produce a ferrimagnetic state as a candidate for the ground state below the transition, while with $\bar{\Psi}^{(3),(4)}$ one can also achieve non-magnetic ground states with $\langle S\rangle = 0$.

In the latter case, $\lambda^2 = 2$, quadrupolar symmetry is also unbroken, $\langle S_i S_j\rangle = \delta_{ij} S(S+1)/2$, and the spontaneous symmetry breaking reveals itself in cubic correlators. Thus, for $\bar{\Psi}^{(3)}$ and $\lambda = \sqrt{2}$, in addition to the usual paramagnetic contribution, $\langle S_i S_j S_k\rangle = \mathrm{i}\varepsilon_{ijk}$ (ε is an antisymmetric tensor), one has

$$\begin{aligned} &\langle S_z^3\rangle = 2 \qquad \langle S_x^3\rangle = \sqrt{2} \qquad \langle S_y^3\rangle = 0 \qquad \langle S_x^2 S_z\rangle = \langle S_y^2 S_z\rangle = 1 \\ &\langle S_y^2 S_x\rangle = -\sqrt{2} \qquad \langle S_z^2 S_x\rangle = 0 \qquad \langle S_x^2 S_y\rangle = \langle S_z^2 S_y\rangle = 0. \end{aligned} \tag{6}$$

By rotating the coordinates, the cubic correlation function can be made symmetric with respect to the coordinate frame,

$$\langle S_i S_j S_k\rangle = \mathrm{i}\varepsilon_{ijk} + \sqrt{3}\mu_{ijk} \tag{7}$$

where $\mu_{iij} = 0$ and $\mu_{ijk} = \mu_{jik}$.

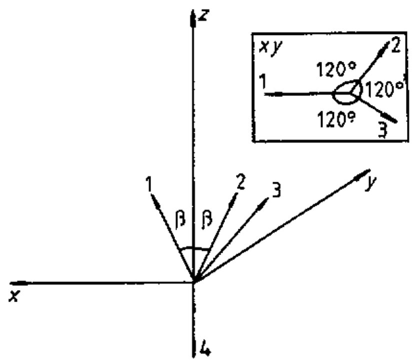

Figure 1. A visual interpretation of a tensor magnet, $\cos\beta = \frac{1}{3}$. Inset: a projection onto the XY plane. In a crude approximation the classical unit vectors can be conceived as $S = \frac{1}{2}$ constituents of $S = 2$. With this arrangement, spin correlations (6) are completely reproduced by (8)

Hereafter we will refer to this structure as a tensor magnet [6]. A visual interpretation of (6) is given by a set of four unit vectors, $\boldsymbol{n}^{(\alpha)}$ ($\alpha = 1, \ldots, 4$), arranged in the form of a tetrahedron (figure 1). With this arrangement, (6) is completely reproduced if we take

$$\langle S_i \rangle = \sum_\alpha n_i^{(\alpha)} \qquad \langle S_i S_j \rangle = A \sum_\alpha n_i^{(\alpha)} n_j^{(\alpha)} \qquad \langle S_i S_j S_k \rangle = B \sum_\alpha n_i^{(\alpha)} n_j^{(\alpha)} n_k^{(\alpha)} \tag{8}$$

with $A = \frac{3}{2}$ and $B = -\frac{4}{9}$.

As is clearly seen from figure 1, the order parameter is a coordinate frame and the low-energy sector thus contains three gapless excitations. Since (6) is an *exact* ground state immediately at the critical line, it thus follows that for $S = 2$ a two-particle instability is accompanied by another instability, which is evidently a three-particle one. The ferrimagnetic state is not contrived in this additional symmetry breaking and therefore seems to be unstable below the transition, thus leaving a tensor magnet as the only probable candidate for the ground state.

For $S = \frac{3}{2}$ the single-site wavefunctions are

$$\begin{aligned}
&\tilde{\Psi}^{(1)} = (|\tfrac{3}{2}\rangle + j|\tfrac{1}{2}\rangle)/(1 + j^2)^{1/2} && \tilde{\Psi}^{(2)} = (|-\tfrac{3}{2}\rangle + j|-\tfrac{1}{2}\rangle)/(1 + j^2)^{1/2} \\
&\tilde{\Psi}^{(3)} = (|\tfrac{3}{2}\rangle + \lambda|-\tfrac{1}{2}\rangle)/(1 + \lambda^2)^{1/2} && \tilde{\Psi}^{(4)} = (|-\tfrac{3}{2}\rangle + \lambda|\tfrac{1}{2}\rangle)/(1 + \lambda^2)^{1/2} \\
&\tilde{\Psi}^{(5)} = (|\tfrac{3}{2}\rangle + \beta|-\tfrac{3}{2}\rangle)/(1 + \beta^2)^{1/2}
\end{aligned} \tag{9}$$

with j, λ and β arbitrary. The first two again favour a ferrimagnetic state $\langle S_z \rangle = \pm\frac{1}{2}$, while the remainder produce one and the same (up to redefinition of the coordinates) non-magnetic state with spontaneously broken symmetry with respect to both quadratic and cubic correlators:

$$\begin{aligned}
&\langle \boldsymbol{S} \rangle = 0 && \langle S_i S_j \rangle = \tfrac{3}{4}\delta_{ij} + \tfrac{3}{2}\delta_{zz} \\
&\langle S_x^3 \rangle = \tfrac{3}{4} && \langle S_y^2 S_x \rangle = -\tfrac{3}{4} && \langle S_z^2 S_x \rangle = 0 \\
&\langle S_y^3 \rangle = 0 && \langle S_x^2 S_y \rangle = 0 && \langle S_i^2 S_z \rangle = 0
\end{aligned} \tag{10}$$

(we choose $\tilde{\Psi}^{(5)}$ and $\beta = 1$).

A visual interpretation of the symmetry properties of (10) is given by a set of three unit vectors $\boldsymbol{n}^{(\alpha)}$ ($\alpha = 1, 2, 3$) arranged in a 120° structure in the XY plane and linked

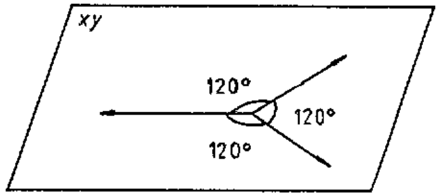

Figure 2. The same as in figure 1 but for $S = \frac{3}{2}$.

with the XY spin correlators in the same way, equations (8), as for $S = 2$, but with $A = \frac{1}{2}$ and $B = 1$ (figure 2).

Note that, while cubic correlators are completely reproduced by this classical picture, for quadrupolar correlators it only points to the existence of a single selected axis.

The low-energy theory of (10) again contains three gapless excitations, indicating that this state is likely to win a competition with the ferrimagnetic state. As was shown in [4], this really happens for $S = \frac{3}{2}$.

For $S = 1$ the required single-site wavefunctions are

$$\bar{\Psi}^{(1)} = (|1\rangle + j|0\rangle)/(1 + j^2)^{1/2} \qquad \bar{\Psi}^{(2)} = (|-1\rangle + j|0\rangle)/(1 + j^2)^{1/2}$$
$$\bar{\Psi}^{(3)} = (|1\rangle + \lambda|-1\rangle)/(1 + \lambda^2)^{1/2}. \tag{11}$$

They produce the only candidate for the ground state below the instability—a non-magnetic configuration with unbroken time-reversal symmetry but with spontaneous symmetry breaking with respect to quadrupolar correlations (spin nematic):

$$\langle S\rangle = 0 \qquad \langle S_i S_j\rangle = \delta_{ij} - \delta_{zz} \tag{12}$$

(we choose $\bar{\Psi}^{(1,2)}$ with $j = \infty$).

In many respects this state resembles an antiferromagnetic one [2]: a visual interpretation of the symmetry properties, in the sense of (8), is given by two antiparallel unit vectors. The only difference is that they both belong to the same site and, hence, the inversion does not produce a new physical state (the order parameter space is isomorphic not to the surface of a unit sphere, S_2, but to a projection plane, P_2).

2. 'Spin-wave' theory

A simple exercise in spin algebra allows us to find, for various S, the most probable configurations replacing the ferromagnetic one after it becomes unstable. For $S = \frac{3}{2}$ and $S = 1$ it has already been proved [1–4] that the unusual long-range order, discussed above, survives the presence of quantum fluctuations. Now we want to prove that this is also the case for higher values of S.

The standard way to do this is to construct the bosonic excitations above the selected configurations. However, all of them presume non-saturation of the site magnetization even in the absence of macroscopic zero-point vibrations, and hence ordinary procedures linking spin operators with a single bosonic field (e.g. Holstein–Primakoff transformation) are invalid. Meanwhile, it is always possible to link spin operators of spin S with $2S$ bosons in that *any* desired single-spin configuration would be a state with no bosons [3, 4, 7, 8]. The corresponding transformations are organized in such a way that commutation relations together with the constraint for $\hat{S}^2$ are satisfied on the physical subspace formed by a vacuum state and $2S$ states with a single excited boson, and the matrix elements between physical and non-physical states are constructed to be zero. With the last condition these transformations are exact at zero temperature for the same reasons as the Holstein–Primakoff one.

Moreover, the presumed mean-field configurations realize exact ground states at the critical lines. Hence, the closeness to the transition indicates a small parameter of the problem (small density of particles in the usual bosonic language).

We begin with $S > 2$, when the mean-field approach favours a ferrimagnetic state, $\langle S_z \rangle = S - 1$.

The general transformation will involve $2S$ bosons. Close to the transition most of them have a finite gap and thus decouple from the low-energy sector discussed below. Only two modes are gapless at the critical line. One of them is the usual spin-wave mode and the second is a two-particle collective excitation with $k = 2\pi$ [5]. Starting from $S_z = S - 1$, these two excitations tend to change S_z by ± 1.

If we restrict ourselves to the exact coefficients for only soft bosons, the general transformation adjusted to the ferrimagnetic state can be written as

$$
\begin{aligned}
S_z &= S - 1 + a^+ a - b^+ b + \dots \\
S_+ &= (2S)^{1/2} a^+ U + [2(2S-1)]^{1/2} U b + \dots \\
S_- &= (S_+^S)^* \qquad U^2 = 1 - a^+ a - b^+ b - \dots
\end{aligned}
\tag{13}
$$

where the dots stand for the contributions from other bosons. For a Heisenberg interaction ($J_1 S_l S_{l+\Delta}$ only), knowledge of (13) is enough to write down the Hamiltonian up to quadratic order in a and b bosons. In the general case of polynomial interaction this is no longer possible. However, the character of the transition implies that exactly at the critical line of ferromagnetic instability, $\delta = J_1 + \Sigma_{n=2}^{2S} \alpha_n J_n = 0$, the low-energy sector contains two *gapless* excitations with no anomalous term present. This justifies a $k = 0$ substitution J_1 directly by δ for $k = 0$ excitations while passing from Heisenberg to polynomial interaction. The part of (1) that is quadratic in bosons thus reads

$$
\begin{aligned}
H = \sum_k a_k^+ a_k [2\delta(2S-1) + \lambda(1-\gamma_k)] + b_k^+ b_k [2\delta S + \lambda(1-\gamma_k)] \\
+ 2\delta[S(2S-1)]^{1/2} \gamma_k (a_k^+ b_{-k}^+ + a_k b_{-k})
\end{aligned}
\tag{14}
$$

where δ is positive below the transition (it is presumed that $\delta \ll 1$), λ (non-universal) stands for the inverse effective mass and $\gamma_k = z^{-1} \Sigma_{\Delta_i} \exp(ik\Delta_i)$, where Δ_i is a vector linking a given spin with one of its z nearest neighbours.

The diagonalization of (14) gives two 'spin-wave' excitations. One of them, $\omega_1(k)$, is gapless and quadratic in k for low momenta, which occurs for any isotropic system with non-zero spontaneous magnetization. The other, $\omega_2(k)$, acquires a finite *positive* gap for $\delta > 0$,

$$
\omega_2(k=0) = 2\delta(S-1). \tag{15}
$$

The spin-wave theory thus confirms the mean-field proposal about the ground state below the transition. Moreover, $\langle a^+ a \rangle = \langle b^+ b \rangle$, ensuring that $\langle S_z \rangle = S - 1$ even in the presence of zero-point fluctuations.

Consider next the phase diagram in a magnetic field. It proves to be practically the same as for classical ferrimagnets. In fact, the application of the field adds $-H$ to $a_k^+ a_k$ and $+H$ to $b_k^+ b_k$.

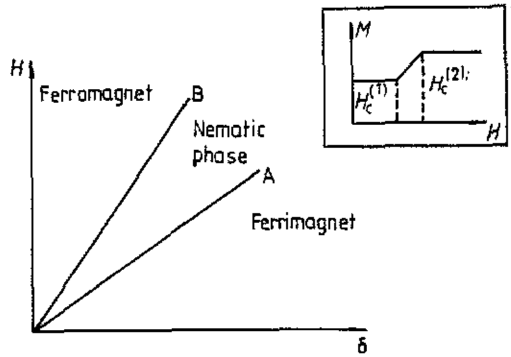

Figure 3. Phase diagram in the δ, H plane for $S > 2$. Inset: field dispersion of the longitudinal magnetization. The lines denote: (A) $H = H_c^{(1)} = 2\delta(S-1)$, spin-flop field; (B) $H = H_c^{(2)} = 2\delta S$, saturation field.

Diagonalization then gives $\omega_1(k=0) \equiv H$, yielding an ordinary precession of the magnetization, while

$$\omega_2(k=0) = 2\delta(S-1) - H. \tag{16}$$

It then follows that a quantum ferrimagnetic state with fixed $\langle S_z \rangle = S - 1$ remains unchanged up to 'spin-flop' field

$$H_c^{(1)} = 2\delta(S-1). \tag{17}$$

In higher fields, the longitudinal magnetization grows with the field and reaches saturation at

$$H_c^{(2)} = 2\delta S. \tag{18}$$

This spin-flip field value can also be obtained by using the same transformation as (16) but adjusted to a ferromagnetic ordering ($S_z = S - a^+a - 2b^+b + \ldots$; $S^+ = (2S)^{1/2}Ua + [2(2S-1)]^{1/2}a^+b + \ldots$). $H_c^{(2)}$ occurs as the point where the gap in the b-boson excitation specrum disappears.

Between $H_c^{(1)}$ and $H_c^{(2)}$ the quadrupolar symmetry with respect to the transverse spin components is spontaneously broken due to the non-zero condensate of ω_2 quasi-particles ($\langle S_i^\perp S_j^\perp \rangle \neq \lambda\delta_{ij}$) and the low-energy spectrum thus contains a gapless branch of excitations.

The phase diagram in the δ, H plane together with the field dispersion of the magnetization are presented in figure 3.

We now turn to the special cases of low S and begin with $S = 2$, where the mean-field and symmetry arguments favour the tensor-like ordering (6) and (7).

The (presumed) additional softening at the transition forces us to write down an exact transformation to bosons. For a tensor-like ground state it can be chosen as follows (the site index is omitted):

$$\begin{aligned} S_z &= \sqrt{2}(a^+d + d^+a) + c^+c - a^+a + \sqrt{2}(c^+U + Uc) \\ S_+ &= (S_-^*) = \sqrt{2}[\sqrt{2}(b^+U + Ua) + (a^+b - b^+c) + \sqrt{2}(c^+d + d^+b)] \end{aligned} \tag{19}$$

where, as usual, $U^2 = 1 - a^+a - b^+b - c^+c - d^+d$.

The unusual form of (19) comes from the fact that the proposed vacuum state, as in (5) with $\lambda^2 = 2$, involves different z projections. One can make sure that all the required conditions are satisfied in a physical subspace and the state with no bosons is specified by (6).

The $S = 2$ polynomial model normally contains four terms

$$H = -\sum_l x_l + \beta x_l^2 + \gamma x_l^3 + \rho x_l^4 \qquad x_l = \sum_\Delta S_l S_{l+\Delta}. \tag{20}$$

A condition for the instability of a ferromagnetic state to occur due to the softening of the two-particle bound states specifies a line

$$1 + \beta + 13\gamma + 25\rho = 0 \tag{21}$$

along which the following inequalities must be fulfilled:

$$\begin{aligned} &1 + 4\beta + 16\gamma + 64\rho > 0 \\ &1 - \beta + 21\gamma - 41\rho > 0 \\ &1 - 2\beta + 28\gamma - 104\rho > 0. \end{aligned} \tag{22}$$

As can be easily checked, (22) is satisfied in some region of parameters fitting (21).

A bosonic spectrum of (20) obtained with the use of (19) contains one decoupled d excitation with a finite positive gap and three massless excitations associated with the broken SO(3) symmetry.

Up to quadratic order in bosons the low-energy part of the Hamiltonian reads

$$\begin{aligned} H_2 = 4\sum_k &(a_k^+ a_k + b_k^+ b_k + c_k^+ c_k)[\delta + \lambda(1 - \gamma_k)] \\ &+ \delta\gamma_k[a_k^+ b_{-k}^+ + a_k b_{-k} + (c_k^+ c_{-k}^+ + c_k c_{-k})/2] \end{aligned} \tag{23}$$

where $\delta = -(1 + \beta + 25\rho)$ and $\lambda = 1 + \beta + 11\rho$ near the critical line (to simplify the calculations, we put $\gamma = 0$).

Diagonalization produces three *identical* gapless branches with low-k dispersion, $w = vk$, where the spin-wave velocity reads

$$v = 4(2\delta\lambda/z)^{1/2} \tag{24}$$

and z is the number of nearest neighbours.

The positiveness of v as well as of the d boson gap ensures the stability (at least, locally) of the tensor-like state below the transition.

The interactions between gapless bosons fit the Adler principle and thus do not destroy linearity in k of the dispersion relation. They do, however, considerably renormalize the spin-wave velocity value since even in the vicinity of the critical line it is only the density of quasiparticles that is small for $\delta \ll 1$; the interaction between bosons is always strong. The exact expression for v can be obtained even without calculations since exactly at the critical line one of the excitations is a conventional ferromagnetic spin wave. Thus the renormalized λ must coincide with $1 + 4\beta + 16\gamma + 64\rho$, and since the relative corrections to δ are small for $\delta \ll 1$, the renormalized spin-wave velocity reads

$$v = 4(2\delta/z)^{1/2}(1 + 4\beta + 16\gamma + 64\rho)^{1/2}. \tag{25}$$

Strictly speaking, exact knowledge of v is less important than the fact that anharmonic terms do not distinguish between three types of bosons and threefold degeneracy of the spectrum thus survives the perturbative corrections. As an obvious consequence, quantum effects do not break the quadrupolar symmetry:

$$\langle S_z^2 \rangle = 2 + \sum_k \langle c_k^+ c_k \rangle + \langle a_k^+ a_k \rangle - 2\langle b_k^+ b_k \rangle \equiv 2. \tag{26}$$

As expected, the second possible candidate for the ground state—a ferrimagnetic configuration—proves to be unstable for $\delta > 0$. To see this, we introduce bosons in the same way as in (13):

$$\begin{aligned} S_z &= 1 + a^+ a - b^+ b - 2d^+ d - 3c^+ c \\ S^+ &= (S^-)^* = 2a^+ U + \sqrt{6} U b + \sqrt{6} b^+ d + 2d^+ c. \end{aligned} \tag{27}$$

The substitution of (27) into (20) results in a bosonic Hamiltonian where one bosonic field has a finite positive mass while the remainder are gapless along the critical line. Below the instability the low-energy part of the Hamiltonian, quadratic in bosons, reads

$$H = \sum_k A_k^{(1)} a_k^+ a_k + A_k^{(2)} b_k^+ b_k + B_k (a_k^+ b_{-k}^+ + a_k b_{-k}) + A_k^{(3)} c_k^+ c_k \tag{28}$$

with

$$\begin{aligned} &A_k^{(1)} = 6\delta + \lambda_1 (1 - \gamma_k) && A_k^{(2)} = 4\delta + \lambda_2 (1 - \gamma_k) \\ &A_k^{(3)} = -6\delta + \lambda_3 (1 - \gamma_k) && B_k = 2\sqrt{6}\delta\gamma_k \qquad \lambda_i > 0. \end{aligned} \tag{29}$$

The negativeness of $A_0^{(3)} = -6\delta$ ensures the instability of the ferrimagnetic state immediately below the transition, which confirms the initial conjecture.

Now we shall discuss the effect of magnetic field. It seems natural to expect that the zero-field susceptibility has a maximum for the most symmetric configuration of four unit vectors on figure 1 with respect to the magnetic field direction. Then the reorientation process would be continuous with unit vectors turning to the field like the petals of flowers. This configuration can easily be found and in the case of a magnetic field directed along z axis is characterized by only two non-zero cubic correlators:

$$\langle S_x^2 S_z \rangle = A \qquad \langle S_y^2 S_z \rangle = -A \qquad A^2 = 3. \tag{30}$$

The application of the field leads to $\langle S_z \rangle = H/2\delta$. The low-energy spectrum constructed above (30) evidently contains a gapless branch reflecting the symmetry breaking in the XY plane and a branch with $\omega(k=0) \equiv H$ yielding a precession of the magnetic moment about the field direction.

Surprisingly, in the 'spin-wave' approximation the third branch also proves to be gapless for $H \neq 0$, though this is not dictated by any kind of broken symmetry.

The situation resembles that in 2D triangular antiferromagnets [9–11] where the 'classical' spectrum in the magnetic field also contains accidental gapless modes. This additional softening is a purely classical phenomenon; quantum fluctuations are known [11] to lift the accidental degeneracy†. However, this peculiarity of the 'classical' system points out that the means of reorientation is governed by quantum fluctuations and may differ significantly from the initially expected most symmetric one.

In 2D triangular antiferromagnets quantum fluctuations favour reorientation via the intermediate collinear ferrimagnetic phase. This may also be the case for tensor magnets.

† This is also the case for some frustrated magnetic systems in zero field [12–14]. The lifting of the accidental degeneracy by quantum fluctuations is known as 'ordering from disorder' [15].

To investigate this possibility, we must calculate the spectrum above the ferrimagnetic state ($\langle S_z \rangle = 1$) in the presence of the field.

Adding the Zeeman terms to (28) as dictated by (27)

$$\delta A^{(1)} = -H \qquad \delta A^{(2)} = H \qquad \delta A^{(3)} = 3H \tag{31}$$

and performing a diagonalization, we obtain in the limit of $k \to 0$:

$$\omega_1 \equiv H \qquad \omega_2 = 2\delta - H \qquad \omega_3 = 3(H - 2\delta). \tag{32}$$

The ferrimagnetic state thus proves to be stable (in the spin-wave approximation) along a line $H \equiv 2\delta$, where ω_2 and ω_3 are simultaneously gapless. Its 'classical' energy at $H = 2S$ is equal to that of the most symmetric configuration and thus, at exactly $H = 2\delta$, the ferrimagnetic configuration belongs to a family of classically degenerate ground states.

Note that, although in the spin-wave approximation ω_2 and ω_3 soften along the same line, $H = 2\delta$, the instabilities governed by ω_2 and ω_3 are of completely different nature. Really, ω_3 is the energy of uncoupled c bosons and it follows from (27) that the condensation of these excitations produces a symmetry breaking with respect to cubic correlators. In contrast, ω_2 excitations are created by mixed a and b bosons and the ω_2 condensation produces a symmetry breaking with respect to *quadrupolar* correlators ($\langle S_x^2 \rangle \neq \langle S_y^2 \rangle$). Hence, in the real system, with quantum fluctuations present, there are absolutely no reasons for both transitions to occur at the same field value.

In 2D Heisenberg triangular antiferromagnets, where the classical scenario is exactly the same, quantum fluctuations were shown [11] to stabilize a collinear ferrimagnetic phase in a *finite* region of fields. Inside, all the excitations acquire finite gaps since no continuous symmetry is broken.

In principle, the same calculations could be performed in the present case. However, for technical reasons it proved to be simpler to investigate what kind of broken symmetry is realized immediately below the ferromagnetic instability in the presence of the field. A symmetry breaking with respect to cubic correlators would evidently lead to a continuous reorientation terminating in the symmetric arrangement (30), while the instability with respect to quadrupolar correlators would produce a reorientation via an intermediate collinear (i.e. ferrimagnetic) phase since different symmetries would be broken at $H = 0$ and near saturation.

An analysis implies use of a transformation adjustable to the $S = 2$ ferromagnetic state. It reads

$$\begin{aligned} S_z &= 2 - a^+a - 2b^+b - 3c^+c - 4d^+d \\ S_- &= (S_+)^* = 2a^+U + \sqrt{6}\,b^+a + \sqrt{6}\,c^+b + 2d^+c \end{aligned} \tag{33}$$

where, as usual, $U^2 = 1 - a^+a - b^+b - c^+c - d^+d$.

To quadratic order in bosons, the Hamiltonian (20) is

$$H = \sum_k A_k^{(1)} a_k^+ a_k + A_k^{(2)} b_k^+ b_k + A_k^{(3)} c_k^+ c_k + A_k^{(4)} d_k^+ d_k \tag{34}$$

where $A_k^{(4)}$ has a finite positive gap for small δ and H, while

$$\begin{aligned} A_k^{(1)} &= H + \lambda(1 - \gamma_k) \\ A_k^{(2)} &= 2(H - 4\delta) + \lambda(1 - \gamma_k) \\ A_k^{(3)} &= 3(H - 4\delta) + \lambda(1 - \gamma_k) \end{aligned} \tag{35}$$

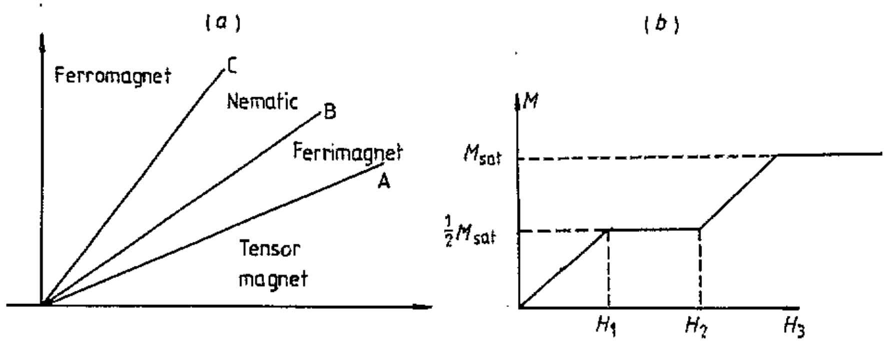

Figure 4. (*a*) Phase diagram on δ, H plane for $S = 2$. The lines A and B denote the boundaries of the intermediate ferrimagnetic phase. In the spin-wave approximation they both coincide with $H = 2\delta$. The finite region of the ferrimagnetic phase is created by quantum fluctuations. The saturation field (line C) is $H = 4\delta$. (*b*) Magnetization versus magnetic field curve. In between H_1 and H_2 the longitudinal magnetization is exactly equal to one-half of the saturation value.

and $\lambda = 4(1 + 4\beta + 16\gamma + 64\rho)$.

As expected, if we restrict ourselves to quadratic order, then the two-particle (b boson) and three-particle (c boson) instabilities occur simultaneously, at $H = 4\delta$, due to accidental degeneracy. We must investigate how this degeneracy is lifted by anharmonic corrections.

In the usual spin-wave theory the possibility to develop a perturbative approach for spin-wave interactions is associated with the smallness of $1/s$. The present approach does not contain such a small parameter and thus, strictly speaking, no exact result can be obtained. However, we believe that even the lowest-order anharmonic corrections will give a qualitatively correct description of the effects due to quantum fluctuations.

The lowest-order anharmonic terms are of cubic origin. Those which renormalize the spin-wave frequencies at $k = 0$ are

$$H_3 = 2\sqrt{6}\delta \sum_k (a_k^+ a_{-k}^+ b_0 + a_k^+ b_{-k}^+ c_0)\gamma_k + \text{HC}. \tag{36}$$

The second-order corrections shift the critical fields of b and c excitations to

$$2(H_b - 4\delta) - (24\delta^2/\lambda)I = 0 \qquad 3(H_c - 4\delta) - (12\delta^2/\lambda)I = 0 \tag{37}$$

where, in three dimensions, $I = W - 1$ and $W = \Sigma_k 1/(1 - \gamma_k)$ is a Watson integral [16].

It follows from (37) that a two-particle instability passes ahead of a three-particle one and the reorientation for $S = 2$ thus follows the scenario stipulating an intermediate ferrimagnetic phase with a magnetization equal to a half of the saturation value (even in the presence of quantum fluctuations).

The phase diagram in the H, δ plane together with the magnetization versus magnetic field curve are presented in figure 4.

Note that the low H stage of the reorientation process can be conceived as a continuous turn of the three unit vectors in figure 1 to the field direction, which for a given arrangement is presumed to coincide with the z axis, while the fourth spin will remain antiparallel to the field. However, in contrast to a 2D triangular antiferromagnet, the high-field phase cannot be visualized as a phase with three parallel spins.

Consider next the case of $S = \frac{3}{2}$. The zero-field structure has already been established in [4]: it is a non-magnetic state (10) with spontaneously broken symmetry with respect to both quadratic and cubic correlators. Here we will focus on the behaviour in non-zero magnetic field $H \equiv H_z$. As is seen from figure 2, the natural way of reorientation is when a triad of unit vectors simultaneously turn towards the field direction. This implies that in a non-zero field a spontaneous symmetry breaking reveals itself only in cubic correlators.

However, a calculation of the excitations above this state in a spin-wave approximation again gives an accidental gapless mode, thus pointing out that the real way of reorientation is governed by quantum fluctuations and may be completely different. By analogy with the $S = 2$ case, one may also propose that the reorientation is accompanied by an intermediate ferrimagnetic phase with $\langle S_z \rangle = \frac{1}{2}$, especially as the spin-wave calculations predict the ferrimagnetic state to remain stable (with two gapless modes) along $H = \tilde{\delta}$ (where $\tilde{\delta}$ stands for a shift from a critical line for the $S = \frac{3}{2}$ version of (20) with $\rho = 0$). However, the situation is different from $S = 2$ in that the symmetric way of reorientation discussed above is the only one where the symmetry with respect to quadrupolar correlators can be preserved. All the other ways immediately imply $\langle S_x^2 \rangle \neq \langle S_y^2 \rangle$, and hence one and the same symmetry is broken at low and high fields. Thus, there are no physical reasons to expect any intermediate phase to exist. The only question to answer is which type of reorientation (symmetric or with symmetry breaking with respect to the quadrupolar correlators) is favoured by quantum fluctuations. To answer this we will again investigate what kind of broken symmetry is realized immediately below the ferromagnetic instability in the presence of the field.

Linking spin operators with bosons by

$$\begin{aligned} S_z &= \tfrac{3}{2} - a^+a - 2b^+b - 3c^+c \\ S_- &= (S_+)^* = \sqrt{3}a^+U + 2b^+a + \sqrt{3}c^+b \end{aligned} \tag{38}$$

and restricting ourselves to quadratic order, we obtain that, as a consequence of accidental degeneracy, the two-particle (b boson) and three-particle (c boson) excitations undergo softening simultaneously at $H = 3\tilde{\delta}$.

The lowest-order renormalization is produced by

$$H_3 = 2\sqrt{3}\tilde{\delta} \sum_k (a_k^+ a_{-k}^{\pm} b_0 + \tfrac{1}{2}\sqrt{3} a_k^+ b_k^+ c_0)\gamma_k + \text{HC}. \tag{39}$$

The second-order corrections shift the critical fields to

$$\begin{aligned} &2(H_b - 3\tilde{\delta}) - (12\delta^2/\lambda)I = 0 \\ &3(H_c - 3\tilde{\delta}) - (9\delta^2/2\lambda)I = 0 \end{aligned} \tag{40}$$

where λ is an inverse effective mass and I is the same as in (37). It follows from (40) that the two-particle instability again passes ahead of the three-particle one, thus producing a continuous reorientation with spontaneous quadrupolar ordering surviving up to saturation.

The case of $S = 1$ has already been solved in [1–4]. Below the instability, the ground state is a spin nematic with a spontaneous symmetry breaking with respect to quadrupolar correlations only. The order parameter space is isomorphic to P_2 and the low-energy sector contains two gapless excitations.

The reorientation proceeds continuously. In the presence of the field one of the excitations acquires a gap $\omega \equiv H$, while the other remains gapless up to $H = 2\delta$, the spin-flip field.

3. Summary and discussion

We have shown that the instability of a ferromagnetic state induced by a two-particle bound-state condensation produces different non-trivial phases. For $S > 2$ a ferromagnetic configuration turns into a ferrimagnetic state with $\langle S_z \rangle = S - 1$; for $S = 2$, 1 and $\frac{3}{2}$ the ground state below the instability has zero net magnetization, while a spontaneous symmetry breaking is revealed in quadratic, cubic, or both quadratic and cubic correlations, respectively.

It should be noted, in the case of $S > 2$, that a further deviation from the instability line of a ferromagnet may produce a set of successive first-order phase transitions with quantum jumps of $\langle S_z \rangle$ by 1 and hence a step-like behaviour of the spontaneous magnetization. A simple calculation shows that the dipolar ordering will survive until $\langle S_z \rangle$ is larger than

$$\langle S_z \rangle_{\min} = \begin{cases} 1 + [S/2] & S = \text{integer} \\ \frac{1}{2} + [(S + \frac{1}{2})/2] & S = \text{half-integer} \end{cases} \tag{41}$$

where [ ] denotes the integer part.

It is also worth noticing that in the 1D version of the problem the low S phases demonstrate a difference in fluctuation effects between integer and half-integer spin values. In fact, up to discrete degrees of freedom, which are inessential near the ferromagnetic instability line [4], the order parameters for $S = 1$ spin nematic and $S = 2$ tensor magnet are in essence one or two mutually perpendicular unit vectors, respectively, and fluctuation effects are exactly the same as in O(3) for $S = 1$ and O(4) for $S = 2$ σ models. Quantum fluctuations *completely* restore the symmetry, producing a singlet ground state with a gap immediately above it†. In contrast, for $S = \frac{3}{2}$ it is obligatory for the order parameter to contain a complex scalar. Though the number of low-energy excitations in the bare theory is the same as for $S = 2$, the order parameter space is now isomorphic to $P_2 \times S_1$, where P_2 comes from a spontaneous symmetry breaking with respect to quadrupolar correlators while S_1 results from that in XY cubic correlators.

As was shown in [4], in the 1D case, the low-energy modes associated with different symmetry breaking decouple at large scales. Those associated with P_2 acquire a dynamically generated gap and thus leave the low-energy sector. The $T = 0$ behaviour thus turns out to be critical, as required [17], and coincides with that in the anisotropic XY model.

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† For $S > 2$, fluctuations above the ferrimagnetic state contain no logarithmic divergences and thus do not modify the 'classical' picture independently of S.

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