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ABSTRACT

The ground state properties of S = 1/2 anisotropic Heisenberg magnets with plaquette frustration are investigated. The models can be classified according to whether a straightforward generalization of the Marshall /1955/ Ansatz can be applied, or the ground state has to be approximated by a linear combination of Ising ground states. In the former case, the quantum ground state may have a zero point entropy. In the latter case, a unique ground state with quantum-liquid-like coherence properties is postulated, generalizing an earlier suggestion by Fazekas and Anderson /1974/.

АННОТАЦИЯ

Предлагаются характеристики основного состояния систем с "крушением надежд", описанных анизотропным гейзенберговским гамильтонианом для спинов S = 1/2. Найдено, что модели принадлежат к одному из двух классов: для первого класса применимо простое обобщение анализа Маршалла, не применимое для второго класса, для которого, однако, первое приближение к основному состоянию дается линейной комбинацией изинговских основных состояний. В первом случае квантовое основное состояние может обладать неисчезающей энтропией у нулевой температуры, а во втором случае мы постулируем единственное основное состояние типа предсказанной ранее Фазекашем и Андерсоном /1974/ спиновой квантовой жидкости.

KIVONAT

Az S=1/2 anizotróp Heisenberg mágnesek alapállapotának tulajdonságait vizsgáljuk, plakett-frusztráció esetében. A modellek két osztályba sorolhatók: az első osztályba tartozók alapállapotát a Marshall-Ansatz kézenfekvő általánositása adja meg, mig a másik osztály elemeire, az alapállapot az Ising-alapállapotok lineáris kombinációjával közelithető. Az előbbi esetben, a kvantum alapállapotnak is lehet zérusponti entrópiája. Az utóbbiban, egyetlen alapállapotot jósolunk, spin-kvantumfolyadék jelleggel, Fazekas és Anderson /1974/ egy korábbi javaslatának általánositásaként.

The frustration effect (Toulouse 1977) manifests itself in ways characteristic of the nature of the spin system. For Ising systems, it is typical to find a zero point entropy proportional to some power of N, where N is the number of spins, while for two- and three-component classical spins, the ground state configuration becomes non-colinear and has to be described in terms of chiral ordering (Villain 1977a, b).

Recently, there has been increasing interest in the question whether the inclusion of quantum effects (allowing for spin-flip terms in the Hamiltonian) would lead to qualitatively new effects. Edwards (1976) pointed out that, for large enough spins, each of the classical ground states should go over into a separate quantum ground state. A mean--field-type calculation by Klemm (1979) indicates that quantum fluctuations may be just strong enough to destroy the spin glass state (Edwards and Anderson 1975) for the lowest spin value $S = 1/2$. On the basis of numerical evidence, Marland and Betts (1979) argue that two-dimensional (2D) Heisenberg or XY spin systems experience no plaquette frustration in the sense that any ground state degeneracy is associated with boundary effects (toroidal frustration). It may appear that the non-degeneracy of the ground state is of the same nature as the proven uniqueness of the ground state of the antiferromagnetic Heisenberg model on bipartite lattices

(Marshall 1955).

The aim of the present note is to point out that the effects of frustration are no less remarkable for (at least, some) $S = 1/2$ quantum spin systems than for their classical counterparts. Even if the ground state turns out to be essentially non-degenerate (meaning that any remaining degeneracy should be associated with global symmetry operations as for the usual kinds of magnetic ordering), this unique ground state possesses coherence properties which are very different from those found in non-frustrated systems.

The present discussion closely follows Marshall's (1955). First we recall how the nature of the quantum ground state of the antiferromagnetic Heisenberg Hamiltonian on bipartite lattices is related to the uniqueness of the corresponding Ising ground state. Later, it is indicated how a generalization of the Marshall Ansatz suitable for the case of competing interactions may be sought, and how the frustration effect manifests itself in its more subtle coherence properties.

Let us consider the anisotropic Heisenberg Hamiltonian

$$H = \sum_{\langle i,j\rangle} J_{ij}[(S_i^z S_j^z - \tfrac{1}{4}) + \tfrac{\alpha}{2}(S_i^+ S_j^- + S_j^+ S_i^-)] \qquad (1),$$

where $\langle i,j\rangle$ refers to summation over nearest neighbour pairs, $J_{ij} = \pm 1$, and $\alpha \geqq 0$ is the anisotropy factor. The dimensionality and the structure of the lattice, and the distribution of ferromagnetic (F) and antiferromagnetic (AF) bonds will be specified later.

Let us seek the lowest energy state in the subspace $\sum_i S_i^z = M$. The ground state is one these but at the level of the present discussion, we cannot decide which. The general form of the wavefunction is

$$\Psi_{min} = \sum_\mu c(\mu)|\mu> \qquad (2),$$

where the summation is over all Ising-eigenstates $|\mu>$ with $(N+2M)/2$ up-spins and $(N-2M)/2$ down-spins. It is easy to verify that

$$H\Psi_{min} = \frac{1}{2}\sum_\mu c(\mu) \sum_{<i,j,\mu>} J_{ij}[\alpha|\mu',ij>-|\mu>] \qquad (3),$$

where the second summation is taken over the antiparalel pairs $<i,j,\mu>$ in the state $|\mu>$, and $|\mu',ij>$ denotes the configuration obtained from $|\mu>$ by interchanging the pair $|i,j,\mu>$. The energy E_{min} is defined as the expectation value of (1) with (2):

$$E_{min} = \frac{\sum_\mu \sum_{\mu'} c^*(\mu')c(\mu)<\mu'|H|\mu>}{\sum_\mu |c(\mu)|^2} \qquad (4)$$

A full solution would require treating all c(μ)s as independent variational parameters. Obviously, this is far too ambitious a task. For the present purpose, however, it is perfectly sufficient to give the moduli $|c(\mu)|$ arbitrary fixed values, and try to find the optimum phase relationships; this enables us to draw conclusions about the uniqueness of the ground state (if present), and about its coherence properties. To this end, it suffices to consider the numerator of (4) which, using (3), turns into

$$\sum_{\mu} \sum_{<i,j,\mu>} J_{ij}[\alpha c^*(\mu',ij)c(\mu)-|c(\mu)|^2] \qquad (5).$$

For bipartite lattices, and for J_{ij} = +1 (all bonds AF), the phase relationships are exactly given by the Marshall Ansatz (1955). The lattice can be divided (in a unique way) into sublattices A and B so that a site on A has all its nearest neighbours on B, and vica versa. Denoting the number of up--spins on sublattice A in the state $|\mu>$ by $p(\mu)$, the lowest energy state belongs to

$$c(\mu) = (-1)^{p(\mu)} a(\mu) \qquad (6),$$

where $a(\mu) \geq 0$, since with this choice all contributions to (5) are negative. Let us note that it is only the expectation value of the spin-flip term that is influenced by the choice of the phases of the c(μ)s. Usually, (6) is meant by the proven uniqueness of the ground state; interchanging the role of sublattices at most changes the sign of Ψ_{min}.

It is worth emphasizing that the choice of signs in (6) turned out to be independent of the values $|c(\mu)|$, as well as of α, and reflects the uniqueness (apart from up-down interchange) of the ground state of the Ising part of the Hamiltonian, which in turn is related to the lack of frustration. In the following, it will be demonstrated that the relationship between the classical and quantum behaviours is generally governed by a suitable generalization of the Marshall Ansatz and thus reflects the structure of the ground states of the Ising part of (1); these will be called Ising ground states (IGS).

It is straightforward to generalize (6) to the quantum versions of the Mattis (1976) models. These are given by (1), where now the distribution of F and AF bonds is such that no plaquette is frustrated. The IGS is essentially unique and the Marshall Ansatz (6) is applicable provided that the sublattice A is redefined to mean the sublattice of up-spins in one of the IGSs. While it is no longer true that all nearest neighbour pairs connect different sublattices, by definition all AF bonds still do, and so the corresponding interchanges give a negative contribution to the first term in (5). Of course, F bonds connect sites within the same sublattice; but for these $J_{ij} < 0$, and the Ansatz does not give a change of sign, so the contribution is once again negative. (for the pure F model, Ψ_{min} is nodeless in every S^z subspace) Hence, it is reasonable to predict that, for small enough α at least, the quantum models show the same kind of ferrimagnetic ordering as the corresponding Ising models.

(6) can be similarly extended to frustrated models with essentially unique IGSs (such as a finite concentration of AF bonds in a F sea).

Let us now turn to models in which frustration leads to a macroscopically large number of IGSs. For these, the question arises whether a separate quantum ground state is defined through the use of (6) for each IGS, or these themselves are mixed by quantum effects into a unique ground state. The outcome seems to depend on the model, and we suggest a simple criterion for models in which a unique ground state has to be expected.

For demonstration, let us consider the best-studied case of the triangular antiferromagnet. It is simple to see that plaquette frustration makes it impossible to simultaneously optimize the contributions of all nearest neighbour interchanges. Let the Ising eigenstates φ_1, φ_2 and φ_3 differ from one another in the position of the down-spin on a single triangle (*Fig. 1*). Since both φ_2 and φ_3 are connected to φ_1 by the spin-flip term, we would want to have a combination like $\varphi_1-\varphi_2-\varphi_3$ in a trial ground state. But this would give identical signs to φ_2 and φ_3, which are also connected by the spin-flip term. In choosing the optimum phases for the coefficients $c(\mu)$, one is faced with exactly the same ambiguity as in determining the IGSs (Wannier 1950).

The generalization of (6) to Mattis models may suggest that it is still the best to pick an arbitrary IGS Φ_ν, define sublattice A as the sublattice of up-spins in Φ_ν, and generate a trial ground state $\Psi_{tr}(\Phi_\nu)$ through (6). Probably, this has to be done for large enough spins (Edwards 1976), but this approach is known to fail for S = 1/2, since it does not take advantage of the fact that (1) has nonvanishing matrix-elements between some IGSs (Fazekas and Anderson 1974). *Fig. 2* shows an antiparalel pair (in the centre of the cluster), the interchange of which takes an IGS into another IGS. Such a pair has been termed an interchangeable pair (IP). Since there exist IGSs with a finite concentration of IPs, for $1>>\alpha>0$, the ground state has to be sought as a linear combination of IGSs:

$$\Psi_{gr} \sim \sum_\nu d^o_\nu \Phi_\nu \qquad (7).$$

Since we are dealing with a problem in degenerate perturbation theory, the leading term in the expansion of the ground state energy is <u>linear</u> in α.

Let us note that (7) is fundamentally different form (6) inasmuch as it contains no reference to a fixed division of the lattice into sublattices. For strong uniaxial anisotropy, the ground state should always be of the form (7), whenever there exist IGSs in which we find a finite concentration of IPs. In *Fig. 3*, we give an IGS for Villain's (1977a) odd model, which seems to have the highest concentration of IPs. Previous numerical experience (Fazekas and Anderson 1974) suggests that about 80 % of the maximum concentration of IPs is effective in lowering the ground state energy: this gives the estimate $E_{min}/NJ = -.5-.13\alpha+\theta(\alpha^2)$.

It must be emphasized that while the statistical mechanics of the Ising Hamiltonian is the same for all models related via gauge transformations, the ground state is susceptible to the actual distribution of F and AF bonds: the IP is <u>not</u> a gauge invariant concept. This is simple to see: the IP is an antiparalel pair surrounded by a zero energy contour. By applying a gauge transformation at one of the sites of the IP, the contour remains the same but now it surrounds a paralel pair.

At this point, the question arises whether the IP is a relevant concept if the distribution of F and AF bonds is random. The answer is certainly yes; the IP is a local phenomenon, depending only on the distribution of bonds in the first shell surrounding a given pair, so if an IP can exist at all, the suitable local configuration has a finite probability, and there is a finite concentration of IPs. For small enough α, these will be the most effective in lowering the energy, and the ground state is of the kind (7).

In the case of the triangular antiferromagnet, evidence has been found (Fazekas and Anderson 1974) that (7) posseses some kind of long-range phase coherence which, not being referred to fixed sublattices, is very different from that shown by (6). It was noticed that if two IGSs are connected by two different sequences of IPs, the parity of the number of interchanges in the two sequences is the same. Hence, if we pick a reference IGS Φ_o, and the number of IP interchanges needed to reach the state Φ_ν from Φ_o is $q(\nu)$, we can seek the ground state in the following form

$$\Psi_{gr} = \sum_\nu (-1)^{q(\nu)} b(\nu) \Phi_\nu \qquad (8),$$

where $b(\nu) > 0$. (8) can be thought of as a non-localized generalization of the Marshall Ansatz, which, because of its distinct phase relationships, was taken to describe a spin--quantum-liquid state. If a state of the kind (7) shows any phase coherence at all, it must be like (8); this should be another general feature of frustrated $S = 1/2$ quantum systems.

When α increases from very small values to 1, higher--lying Ising states are mixed in (7) and (8) which can be described by replacing Φ_ν by $\Psi_{tr}(\Phi_\nu)$. The expansion coefficients d_ν become α-dependent but the coherence properties (8) should remain unchanged up to $\alpha = 1$ (Fazekas and Anderson 1974).

To understand the differences between the two kinds of ground state that we encountered: (6) and (8), it is useful to consider their relation to magnetic ordering. (6), by definition, "feels" the sublattices, so it is not surprising that it is compatible with conventional antiferromagnetic ordering (Thouless 1967, Betts and Oitmaa 1977), though, as shown by the example of the 1D isotropic AF, it does not necessarily entail it. When it does, the simple variational tratment (Marshall 1955) can be used to obtain a quasiclassical ground state which can be approached equally well from the $S \to \infty$ limit (Anderson 1952). Whenever (6) gives the correct ground states of a frustrated model, their multiplicity should reflect that of the IGSs, and each can be reached by continuation from the classical limit (Edwards 1976). It has been repeatedly argued (Anderson 1973, Fazekas and Anderson 1974, Fazekas and Sütő 1976, Sütő and Fazekas 1977) that continuation from the Ising limit to $\alpha = 1$, and continuation from the classical limit to $S = 1/2$ give conflicting results for the triangular lattice. This seems to be generally the case if there is a finite concentration of IPs. However, we cannot be sure that this is also a necessary condition for a unique spin-liquid ground state, or a quantum-mechanical mixture like (7) is obtained whenever a large number of IGSs are connected in a fixed finite order of the spin-flip term of the Hamiltonian. In view of the orthogonality proved by Edwards (1976), the latter may be the case.

Finally, it should be added that since the energetical preference given to (7) relies on the existence of the term linear in α, i.e. a finite concentration of IPs, the present considerations are not directly applicable to 3D models for which the ground state entropy is proportional to $N^{1/3}$, or $N^{2/3}$ (Danielian 1961, 1964). But they may be relevant to models like Chui's (1977), or to the B sublattice of the inverse spinel structure (Anderson 1956) though, in the latter, the spin-flip term connects IGSs only in third order (Cullen 1973).

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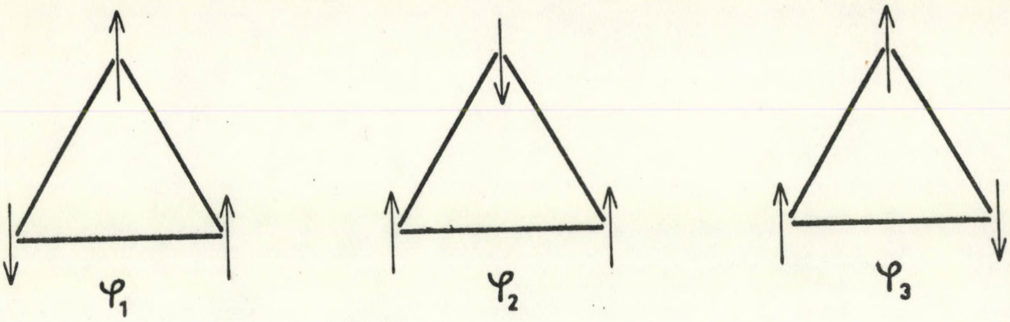

Figure 1.

Three configurations of a plaquette of triangular lattice

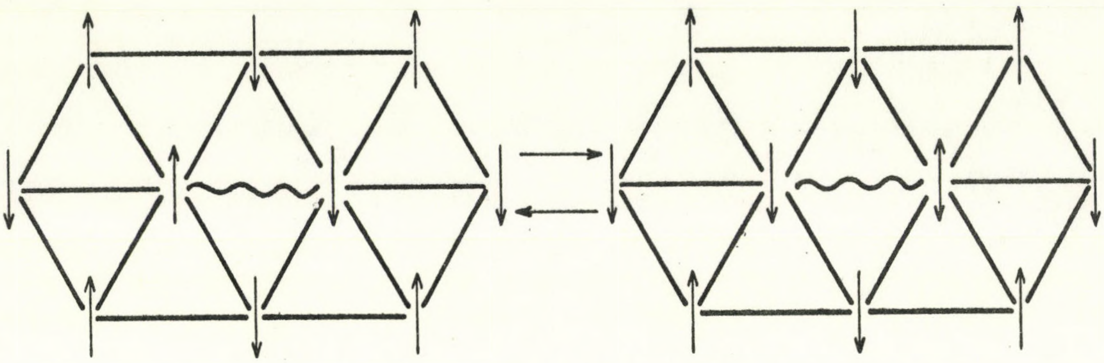

Figure 2.

Interchangeable pair /wavy line/ on the triangular antiferromagnet, in its two possible configurations

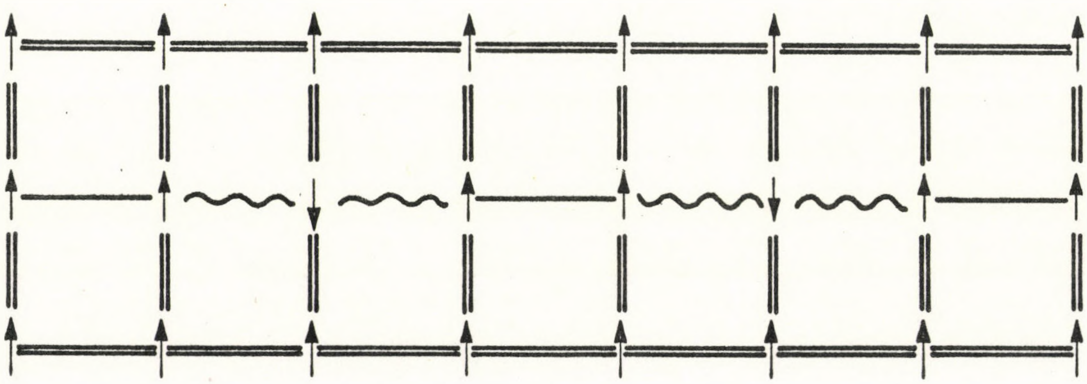

Figure 3.

An IGS for Villain's odd model with a maximum concentration of IPs /wavy lines/. Double lines: F bonds, single lines: AF bonds. The IPs are all on AF bonds.

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