The S = 1 Bilinear Biquadratic Spin Model on the Square Lattice: A Series Expansion Study

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We use extensive series expansions at T = 0 to investigate the phase diagram of a spin-1 Hamiltonian on the square lattice. The model includes bilinear and biquadratic interactions (the 'J-K' model) and has been studied recently using a variety of other methods. We find a clear indication of the three-sublattice order conjectured recently via indirect arguments. We also compute the energy and order parameter in the quadrupolar phases.

1. Introduction

There has been considerable interest recently in spin-1 Heisenberg models with both bilinear and biquadratic nearest-neighbour exchange terms. The generic Hamiltonian, which we refer to hereafter as the 'J-K ' model, is

$$H = J \sum_{<i,j>} \mathbf{S}_i \cdot \mathbf{S}_j + K \sum_{<I,j>} (\mathbf{S}_i \cdot \mathbf{S}_i)^2$$

The exchange parameters J, K may have either sign, and are often parametrized by $J = \cos \theta$, $K = \sin \theta$.

Studies of this Hamiltonian go back at least to the 1960s [1,2]. The mean-field phase diagram was discussed by Chen and Levy [2], where phases of both dipolar and quadrupolar nature were identified, and a more detailed discussion was given by Papanicolaou on the basis of semiclassical 'flavour-wave' theory [3]. More recently, the model on the triangular lattice was invoked to explain unusual properties of the material $NiGe_2S$ [4-7], and the presence of significant biquadratic exchange (with K < 0) in the iron pnictides has been suggested [8].

In the present paper we consider the model defined above on the two-dimensional square lattice. For K = 0, and by continuity for |K| << |J|, we expect a normal collinear ferromagnetic or antiferromagnetic (Néel) phase for J < 0, J > 0 respectively. In other regions of the phase diagram, phases with quadrupolar order, ferroquadrupolar (FQ) or antiferroquadrupolar (AFQ), may occur. Another feature of this Hamiltonian is the existence of special points ($\theta = \pi/4$, $-3\pi/4$, $\pm \pi$), where the spin symmetry group is enlarged from O(3) to U(3) [3].

In Figure 1 we show a schematic phase diagram, based on the flavour-wave theory of Papanicolaou [3], confirmed by Quantum Monte Carlo (QMC) studies [9,10] for K < 0, and a more recent study by Toth *et al.* [11] combining flavour-wave theory with numerical exact diagonalization. The phase structure is firmly established in the lower half of the phase diagram, where QMC calculations are valid, but not in the upper half, since QMC calculations suffer from a minus sign problem for K > 0. The most interesting quadrant is the region $0 < \theta < \pi/2$. Toth *et al.* [11] find a 3-sublattice AFQ phase (termed 'AFQ3') to be stable throughout the region $\pi/4 < \theta < \pi/2$. Their approach becomes less certain below $\theta = \pi/4$, but they argue, by continuity, that a three-sublattice antiferromagnetic phase will be the preferred ground state in a finite window below $\theta = \pi/4$. This is the conjectured AFM3 phase illustrated in Fig. 1, in which the spins
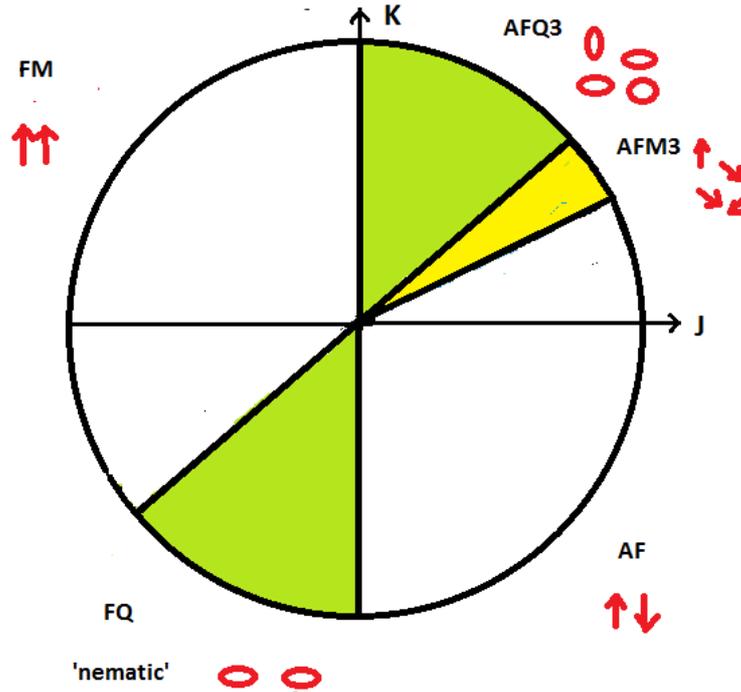
Fig. 1 Phase diagram as a function of angle θ. Key: FM: ferromagnetic; AF: antiferromagnetic; FQ: ferroquadrupolar; AFQ3: antiferroquadrupolar with 3-sublattice order; AFM3: antiferromagnetic, with 3-sublattice order.

are coplanar with a 120^0 angle between spins on successive diagonal sublattices.

The initial motivation for the present work was to find more direct evidence for the AFM3 phase, and to locate the position of the boundary between the AFM3 and Néel phases. At the same time we also obtain independent estimates of both the ground state energy and quadrupole moment in the quadrupolar phases. Given the inherent uncertainty in analytic approaches to S = 1 systems, it is important to test such results by other methods.

2. Method

In our calculations, we employ the method of linked cluster perturbation expansions [12], in which the Hamiltonian is written as

$$H = H_0 + \lambda V$$

and series are obtained for various quantities, such as ground state energy and order parameters, in powers of λ up to some order (typically 10-15). A basis is chosen in each phase, usually consisting of simple product states, where the ground state possesses the expected order in that phase. The unperturbed Hamiltonian H_0 then contains all the diagonal terms in that basis, and the perturbation V all the off-diagonal ones, so that the physical Hamiltonian corresponds to $\lambda = 1$. The series are analysed by standard methods (usually direct or Dlog Pade approximants), and evaluated at $\lambda = 1$. The unperturbed ground state is expected to be adiabatically connected to the true many-body ground state of the full Hamiltonian H. It is possible to check this by testing for a singularity with $\lambda < 1$ in the series. The method has been used successfully in many previous studies, including our previous work [13] on a spin-1 model with single-ion anisotropy.

The calculation then proceeds along standard lines [12], with series for a sequence of finite clusters of increasing size combined to give series for the bulk system in the thermodynamic limit. We have used a total of 23937 clusters with 14 or fewer sites, which yields series for the ground state energy per site $E_0(\lambda)$, the magnetization $M(\lambda) = <S^z_i>$ and
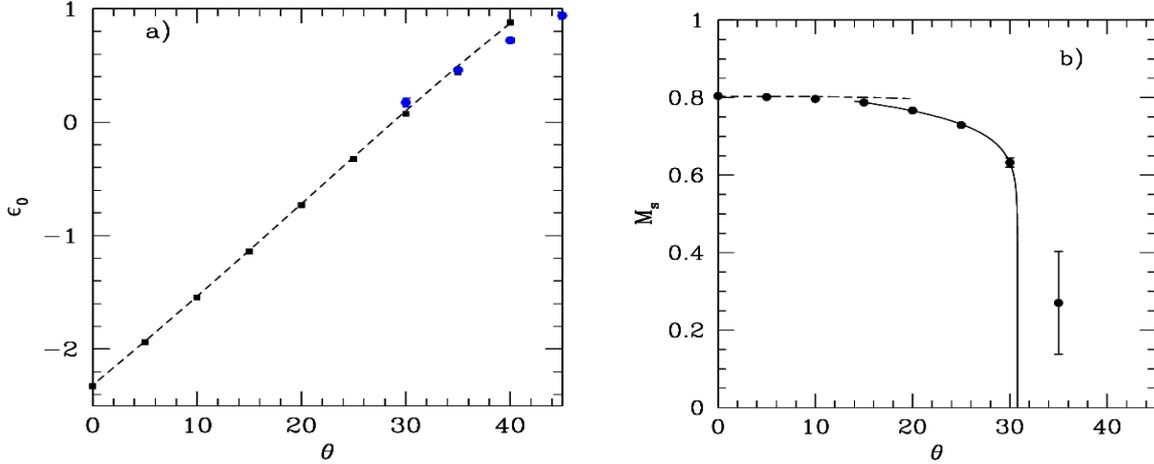
Fig.2. (a) Ground state energy per site from expansions in the Néel phase (black squares) and the AFM3 phase (blue circles). Dashed line: flavour-wave prediction; (b) Estimates of the magnetization in the Néel phase. Dashed line: flavour-wave prediction; solid line: a fit to the data.

the quantity $Q = <(S^z_i)^2>$, which is related to the quadrupole moment, to order λ^{15}. The series data are too extensive to reproduce here, but can be supplied on request. The results are discussed below.

3. Results

3.1 The Néel and AFM3 phases

We now turn to the results. In Fig. 2(a) we show the ground state energy per site, as a function of θ, from series in both the Néel and AFM3 phases. It can be seen that the Néel energy is the lower at small θ, until the AFM3 energy crosses over to become the lower at a transition point which we estimate around $\theta = 36^0$. This is in good agreement with the estimates of Toth *et al* [11]. The flavour-wave estimate of the Néel energy is in extremely good agreement with the series results.

Figure 2(b) shows our estimates of the magnetization as a function of θ. It can be seen that the flavour-wave estimate is again very accurate at small θ. A fit to the simple form $M^z \sim a(\theta_c - \theta)^\beta$ to the data over the range $15 < \theta < 30$ degrees predicts that the magnetization vanishes at about $\theta = 31^0$, with a very small exponent $\beta \sim 0.1$. Even including the uncertain point at 35^0 in the fit gives a transition point near but below $\theta = 35^0$.

The transition from one ordered state to a state with different order is generally expected to be 1st order. From the data above, the transition between the Néel and AFM3 phases looks more like a 2nd order transition. If it is a 1st order transition, it must be a very weak one. Another possibility might be the existence of intermediate (spiral?) phases. We do not explore that possibility further here.

3.2 The AFQ3 Quadrupolar Phase

In the region $\pi/4 < \theta < \pi/2$, Toth *et al* [11] propose a 3-sublattice antiferroquadrupolar (AFQ3) phase. Our series results for the ground state energy per site and quadrupolar moment Q in this region are shown in Figs. 3(a) and 3(b), respectively. As can be seen from Fig. 3(a), our series results for the ground state energy are in excellent agreement with those of Toth *et al* in this region, and join on smoothly to those for the AFM3 phase. This argues for a 2nd order transition between the AFM3 and AFQ3 phases, presumably fixed at $\theta = 45^0$ by the higher symmetry obtaining there, where the dipolar AFM3 order parameter vanishes. Our
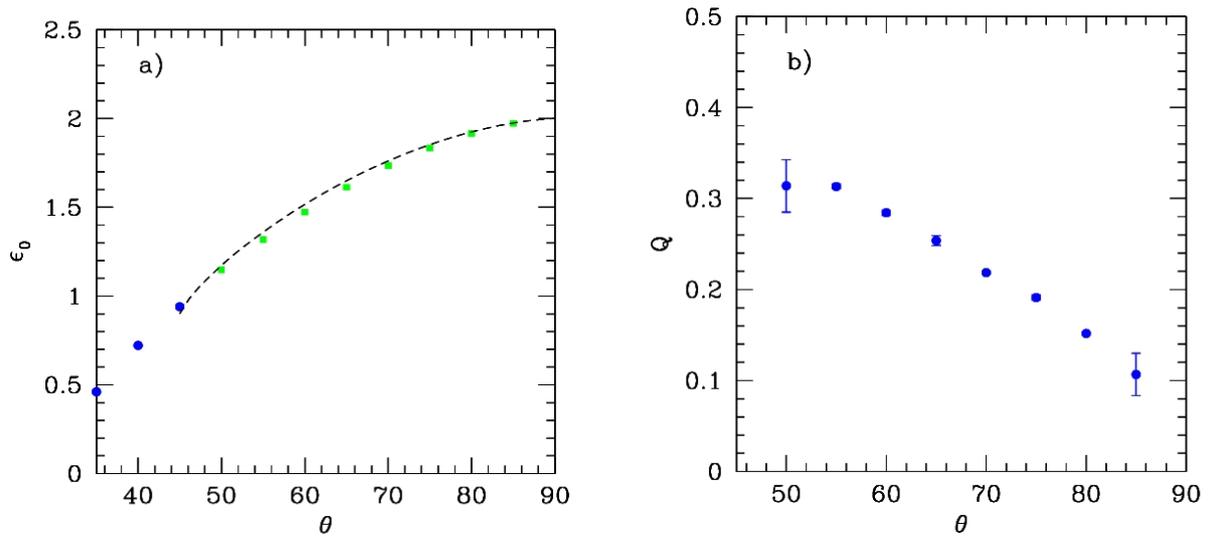
Fig.3. (a) Ground state energy per site from expansions in the AFQ3 phase (green squares) and the AFM3 phase (blue circles). Dashed line: flavour-wave prediction (Toth et al [11]; (b) Estimates of the quadrupole order parameter Q in the AFQ3 phase.

results for this order parameter (not shown) are not sufficiently accurate to confirm this, but are consistent with the magnetization vanishing with exponent ½ at $\theta = 45^0$., as would be predicted by spin-wave/flavour-wave theory.

4. Summary and Conclusions

Our series results are in excellent agreement with the phase diagram suggested by Toth *et al* [11] for this model. The nature of the phase transitions between the different phases requires further exploration.

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