First order transition between magnetic order and valence-bond order in a 2D frustrated Heisenberg model

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Abstract

We study the competition between magnetic order and valence bond order in a two dimensional (2D) frustrated Heisenberg model introduced some time ago by Shastry and Sutherland (B. Sriram Shastry and Bill Sutherland, Physica 108B,1069 (1981) ) for which a configuration of dimers is known to be the ground state in a certain range of parameters. Using exact diagonalisation of small clusters, linear spin wave theory and Schwinger boson mean field theory, we show that the transition between the two types of order is first-order, and that it takes place inside the domain where magnetic long-range order is stable with respect to quantum fluctuations.

PACS 75.10.Jm Quantized spin models
PACS 75.30.Ds Spin waves
PACS 75.30.Kz Magnetic phase boundaries
Frustrating two-dimensional quantum magnets can yield new and exciting physics. The possible effects include: i) an increase of quantum fluctuations that can lead to the disappearance of long-range magnetic order; ii) the appearance of new types of ground-states. While these effects are somehow compatible - they both lead to the destruction of the usual type of order - the interplay between them is not well understood. Let us be more specific and consider for a moment a model that has been extensively studied in the past few years, namely the $J_1-J_2$ model \([1]\). Linear spin-wave theory (LSWT) predicts that magnetic order is destroyed around $J_2/J_1 = 1/2$ due to large quantum fluctuations \([2]\). Consistent with this prediction, series expansions for $S = 1/2$ suggest that a kind of valence-bond order appears in that parameter range \([3]\). Finally, numerical simulations \([4]\) on this model suggest that the magnetization vanishes continuously with increasing frustration, leading to the conclusion that the phase transition between Néel order and valence-bond order is second order. While the overall picture is appealing, this conclusion is very surprising because the degeneracy of the ground state is not the same in these phases (no degeneracy for Néel order, four-fold degeneracy for valence bond order), and one would have expected a transition due to a level crossing, in which case the smooth disappearance of Néel order at the transition is a coincidence. Besides, according to the Schwinger boson mean-field theory (SBMFT), which is in principle an improvement over LSWT, quantum fluctuations are never strong enough to destroy long-range order \([5]\). So, whether magnetic long-range order has to disappear before another type of order can be stabilized is still an open question. As far as the $J_1-J_2$ model is concerned, it seems difficult at the moment to go further because each technique is limited to a certain type of order. For instance, the presence of valence-bond order has not been identified by numerical diagonalization of small clusters.

In this paper, we propose to address that question by turning to another model introduced some time ago by Shastry and Sutherland \([6]\). This model is a 2D frustrated Heisenberg model defined by the Hamiltonian:

$$H = J_1 \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{<lm>} \vec{S}_l \cdot \vec{S}_m$$

(1)

where $<ij>$ means pairs of nearest neighbors and $<lm>$ means pairs of next-nearest neighbors linked by a dashed line in Figure \(\text{a}\). The main interest of this model is that a valence bond solid has been shown to be the ground state for certain values of the parameters. For such a model, the problem reduces to one question: do quantum fluctuations destroy LRO before (or possibly when) the parameters are such that the products of singlets becomes the ground state or do we still have LRO at the transition?

Let us start with a review of the exact results obtained by Shastry and Sutherland. In the classical limit, when $S \to \infty$, the ground state is Néel ordered if $J_1 < J_2$ and is helical otherwise. One can easily show, by minimizing the energy of a single triangle, that the twist between one spin and its nearest neighbor is given by $\theta = \pi \pm \arccos(J_1/J_2)$ and that the three spins must be coplanar. Once two neighbour triangles are in this ground state, one can construct in a unique way such a pattern on all the other triangles of the lattice, thus building a helix. One can show that this helix can be directed in four different directions ( (0,±1) and (±1,0) ) depending on the choice of the values of $\theta$ for the first two triangles. This adds a discrete four-fold degeneracy to the ground state which is also obviously continuously degenerate because of the isotropy of the Hamiltonian. A helix pointing towards $O_x$ is represented on Figure \(\text{a}\).
In the quantum case, the state defined by $|\varphi> = \prod_{lm}[l, m]$, where $[l, m]$ is the singlet state of the diagonal pair $(l, m)$, is an eigenstate of $H$ with an energy per site $E_0 = -S(S + 1)J_2/2J_1$. Using a variational principle, one can prove that this state is the ground state if $J_2/J_1 > 2$ for $S = \frac{1}{2}$ and $J_2/J_1 > 2(1 + S)$ for $S \geq 1$. This state has been called a quantum spin liquid by Shastry and Sutherland because of the lack of long range order. This exact result is the main interest of this model. All these properties are summarized on the phase diagram of Figure 3.

To study quantum fluctuations, it is natural to start with linear spin wave theory (LSWT) \[10\]. The Hamiltonian being quadratic at this level of approximation, one can easily determine the spectrum, and, from it, calculate the magnetization from $<S^z> = S - <a_ia_i^\dagger>$. The curve of stability of the Néel ordered phase has the same shape as for the $J_1-J_2$ and the $J_1-J_3$ models: It drops at the critical value of the frustration for which Néel order disappears \[11\]. However, the results for the helical phase are quite surprising: LSWT predicts that this phase is unstable under quantum fluctuation for arbitrary large values of $S$. This is due to a line of modes of zero frequency defined by $:\cos^2 k_x + \cos^2 k_y = 1 + \frac{b^\dagger_i b_i}{J_2}$. This makes the corrections to the staggered magnetization divergent in 2D. However, this result must be viewed as an artifact of the method and not as a meaningful result: We know that the ground state is four-fold degenerate, so the exact spectrum cannot vanish on a continuous line of wave-vectors. This paradox can be lifted as follows: The dispersion given by LSWT corresponds to modes of vanishingly small amplitudes in the classical limit. But for a quantum system, the amplitudes of the local fluctuations cannot be arbitrarily small, and one should recover a finite frequency for these modes. A similar effect has actually been observed in the collinear phase of the $J_1-J_2$ model by Chandra et al. \[11\].

As shown by these authors, a good method to take this effect into account is to use the Schwinger boson mean field theory (SBMFT) \[10\]. This method starts from a representation of the spin algebra in terms of bosonic operators: $\vec{S} = \frac{1}{2}b^\dagger_\sigma \vec{\sigma}_\sigma' b_{\sigma'}$, the size of the spin being fixed by a constraint on the number of particles on each site: $b^\dagger_i b_i + b^\dagger_\downarrow b_\downarrow_i = 2S$. Defining operators that are quadratic in terms of the bosonic operators by $2B^\dagger_{ij} = b^\dagger_i b_{j\uparrow} + b^\dagger_\downarrow b_{j\downarrow}$ and $2A^\dagger_{ij} = b^\dagger_i b^\dagger_{j\uparrow} - b_i^\dagger b_{j\downarrow}$, the Hamiltonian can be written

$$H = \sum_{(i,j)} J_{ij} (B^\dagger_{ij}B_{ij} - A^\dagger_{ij}A_{ij}) \quad (2)$$

At the mean-field level, one introduces the following order paramaters: $<A^\dagger_{ij}> = 2\alpha_{ij}$ and $<B^\dagger_{ij}> = 2\beta_{ij}$ and the Hamiltonian is replaced with:

$$H_{MF} = \sum_{(i,j)} \left( \beta_{ij} (B_{ij} + B^\dagger_{ij}) - \alpha_{ij} (A_{ij} + A^\dagger_{ij}) - \beta_{ij}^2 + \alpha_{ij}^2 \right) \quad (3)$$

Finally, the local constraint is replaced by a global one and is enforced only on the average through the addition to the Hamiltonian of a term $\mu \sum_i \left(b^\dagger_i b_i + b^\dagger_\downarrow b_\downarrow_i - 2S \right)$, where the chemical potential $\mu$ plays the role of a Lagrange parameter. In order to describe long range helical order in this formalism, one has to multiply each Bose operator by a phase factor \[11\]: $b_i \rightarrow b_i \exp(iQ\vec{r}_i/2)$, where $Q$ is the pitch of the helix. This is equivalent to
perform a local rotation of angle $\vec{Q}_i\vec{r}_i$ on each site around a uniform quantization axis $z$. The order parameters are then given by:

$$\alpha_{ij} = \exp(-i\vec{Q}_i\vec{r}_{ij}/2) <b_{i\uparrow}^\dagger b_{j\uparrow}> + \exp(i\vec{Q}_i\vec{r}_{ij}/2) <b_{i\downarrow}^\dagger b_{j\downarrow}>$$
$$\beta_{ij} = \exp(i\vec{Q}_i\vec{r}_{ij}/2) <b_{i\uparrow}^\dagger b_{j\downarrow}> - \exp(-i\vec{Q}_i\vec{r}_{ij}/2) <b_{i\downarrow}^\dagger b_{j\uparrow}>$$

(4)

while the constraints reads:

$$2S = <b_{i\uparrow}^\dagger b_{i\uparrow}> + <b_{i\downarrow}^\dagger b_{i\downarrow}>$$

(5)

The expectation values $<b_{i\sigma}^\dagger b_{j\sigma}>$ and $<b_{i\sigma} b_{j\sigma'}>$ can be calculated from $H_{MF}$ in terms of $\alpha_{ij}$ and $\beta_{ij}$, so equations (4,5) are a system of non-linear equations from which $\alpha_{ij}$ and $\beta_{ij}$ can be calculated. A LRO state is described by a Bose condensate of $S^*$ particles [5] which gives the long range part of the correlation functions. Solving equations (4,5) for an arbitrary value of $\vec{Q}$ and a given value of $S$ leads to an excitation spectrum that vanishes at three points of the Brillouin zone located at $k = \frac{Q_0}{2}$ and $k = \frac{Q_0}{2} \pm Q_0$, where $Q_0$ depends on $S$ and $J_2/J_1$, but not on $\vec{Q}$. So the physical solution, i.e. the solution that has a Goldstone mode at $k = 0$, is obtained by choosing $\vec{Q} = \vec{Q}_0$. The spin correlation functions $<\vec{S}_i \cdot \vec{S}_j>$ have then the following long range behaviour:

$$<\vec{S}_i \cdot \vec{S}_j> \sim S^* \cos(Q_0 \vec{r}_{ij})$$

(6)

Note that $\vec{Q}_0$ is not equal to the classical value. This theory thus allows one to calculate in an easy way the renormalization of the pitch of the helix due to quantum fluctuations. Note also that the rotation we perform is quite different from the one done in [9] which led to $(\sin \theta_{ij}) \vec{S}_i \wedge \vec{S}_j$ terms in the Hamiltonian. They are difficult to treat because they cannot be written in terms of $A_{ij}$ and $B_{ij}$ and neglecting them gives a spectrum that does not have the appropriate Goldstone modes.

Looking for a solution of the mean-field equations such that both the Bose condensate and the spectrum vanishes, we find the critical value of the spin. The phase diagram is depicted in figure 2. Within SBMFT, the Néel phase is found to be stable against quantum fluctuations in a larger part of the phase diagram than within LSWT. For every physical value of the spin, we find a domain of frustration in which the system sustains helical LRO. This is the main advantage of the SBMFT over LSWT which is unable to describe helical LRO in the particular case of the Shastry-Sutherland model. The transition between Néel and helical order is always second order and takes place at a frustration slowly increasing from $J_2/J_1 = 1$ in the classical case to $J_2/J_1 \approx 1.1$ for $S = 1/2$.

For a given value of the spin, the Shastry-Sutherland model has generally two solutions at the mean-field level: In addition to the LRO one which exists for small enough frustration, there is a spin liquid solution defined by: $|\phi> = \Pi_{<i,j>} A_{ij}^\dagger |0>$. This solution has a gap in the excitation spectrum and the correlation functions vanish at long distance. It is the equivalent, within SBMFT, of the dimerized state, which is always an eigenstate, and it has exactly the same energy. That the agreement between the energies is perfect is to a certain extent fortuitous because the wave-functions are not the same: $|\phi>$ does not represent a spin configuration because it mixes wave-functions with different numbers of particles, which is allowed by our approximation because the constraint is satisfied only on the average.
The physical picture that emerges from these results is that for all the physical values of $S$, a first order transition takes place between long range helical order and the valence bond ground state, a transition at which the magnetization drops abruptly to zero, and a finite gap in the excitation spectrum opens. For spin 1/2, this transition occurs at $J_2/J_1 \approx 1.65$. An other indication that the transition we observed is first order comes from the coexistence of the two solutions of the mean field equations on the both sides of the transition.

For spin one-half, we have performed exact diagonalisation on small clusters using Lanczos algorithm. Our results are summarized on figure 3. We have performed finite size scaling from the 8,16 and 20 sites clusters for the staggered magnetization and the ground state energy following the formula given in \[4\]. These results agree very well SBMFT for $J_2/J_1 < 1.1$ and $J_2/J_1 > 1.65$. This confirms the reliability of the approximation we used and our conclusions on the behaviour of the Shastry-Sutherland model. We want to point out that exact diagonalisation on small cluster gives no valuable results in the strongly frustrated regime $1.1 < J_2/J_1 < 1.65$ because small clusters cannot sustain helical LRO. As a consequence, it is impossible to extract information about the transition between LRO and valence bond order from this technique.

In conclusion, we predict that for any physical value of the spin, the model undergoes a first order phase transition between magnetic order and valence-bond order. This is quite different from the scenario outlined in the introduction for the $J_1 - J_2$ model and according to which the system loses its LRO continuously at the transition to a valence-bond type of order. More qualitatively, our results for spin one-half can be summarized as follows: For $J_2/J_1 < 1.1$, the system exhibits Néel order. This type of order is replaced by helical order around $J_2/J_1 \approx 1.1$. Finally, for $J_2/J_1 \approx 1.65$, there is a first order phase transition between the helical ordered phase and the spin liquid.

Another lesson to be learnt from this work concerns the limitation of exact diagonalisation to localize a transition between an ordered state and a spin liquid. From our study of the Shastry-Sutherland model for $S = 1/2$, one would have been tempted, on the basis of exact diagonalisation alone, to conclude that the transition is second order and takes place at $J_2/J_1 \approx 1.5$. However, the SBMFT provides convincing arguments that this is an artifact of this method because helical order cannot be realized on small cluster. In the case of the regular $J_1 - J_2$ model mentioned in the introduction, helical order is also a good candidate around $J_2/J_1 = 1/2$. At the classical level a whole class of helical states are actually degenerate. Whether the second order transition inferred from exact diagonalisation is a real one can then be questioned on the basis of our results. Work along these lines is in progress.

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FIGURES

FIG. 1. Shastry-Sutherland model. Full lines: links of magnitude $J_1$; Long dashed lines: links of magnitude $J_2$. Short dashed lines: unit cell. Arrows: typical helical configuration.

FIG. 2. Phase diagram of the Shastry-Sutherland model.

FIG. 3. Variation of several quantities as a function of $J_2/J_1$ for $S = 1/2$. (a) Pitch of the helix; (b) staggered magnetisation; (c) ground state energy.
FIG. 2

Dimerized state:
SBMFT and variational principle:

0.5 1 1.5 2 2.5 3 3.5 4 4.5 5 5.5 6
0 0.5 1 1.5 2 2.5 3 3.5 4 4.5 5

Néel
Helix

LSWT
SBMFT
2nd order transition

J_2/J_1
1/S
S

1/2
3/2

0 1 1.5 2 2.5 3 3.5 4 4.5 5 5.5 6
