Phase diagram of the spin-1/2 $J_1$-$J_2$-$J_3$ Heisenberg model on the square lattice

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Abstract. We presents the results of an extensive numerical study of the phase diagram of the spin-1/2, $J_1$-$J_2$-$J_3$ Heisenberg model on a square lattice, for both ferromagnetic and antiferromagnetic nearest-neighbor interactions $J_1$, using exact diagonalization with periodic and twisted boundary conditions. Comparison is made with published spin wave calculations. We show that quantum fluctuations play a very important role, changing both the extent and the wave vector of classical spiral phases, and leading to new quantum phases where the classical spiral states have a high degeneracy. These include a new phase with small or vanishing spin-stiffness, in addition to known valence-bond-solid and bond-nematic phases.

1. Introduction
The effects of quantum fluctuations on the ground-state is a major question in frustrated magnetism. One of the most studied examples is the $J_1$-$J_2$ Heisenberg model with spin $S = 1/2$ on a square lattice with competing antiferromagnetic (AF) first and second neighbor exchange, where the nature of the non-magnetic ground-state that occurs around the maximally frustrated point $J_2/J_1 \sim 0.5$ has been much debated. Suggestions include valence bond crystals (VBC’s) with columnar, plaquette, or mixed columnar-plaquette order, and spin liquid (SL) states. One approach to this problem is to include a third-neighbor exchange $J_3$, and a recent study of the $J_1$-$J_2$-$J_3$ model by Mambrini et al. using exact diagonalization (ED) in full and reduced basis sets found plaquette VBC order extending down to the $J_3 = 0$ limit [1].

In past years, several classes of layered vanadate and cuprate compounds, which are well approximated by a spin-1/2 Heisenberg model on a square lattice have come to the fore [2, 3, 4]. Some of these seem to be well described by a $J_1$-$J_2$ model with AF $J_2$, and either FM, or AF $J_1$. However others have more complex properties which hint at longer range interactions. This motivates us to consider $J_1$-$J_2$-$J_3$ models with both FM and AF $J_1$ [5, 6].

In this proceedings paper, we report results of further studies of the $J_1$-$J_2$-$J_3$ model:

$$\mathcal{H} = J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle ij \rangle_2} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle ij \rangle_3} \mathbf{S}_i \cdot \mathbf{S}_j$$

(1)

that include now both the case of FM and AF $J_1$. We set $J_1 = -1$ ($J_1 = 1$) when $J_1$ is FM (AF) throughout. As before we principaly used exact diagonalizations (ED) on clusters...
Figure 1. (Color online). Groundstate phase diagram for the spin-1/2, $J_1$-$J_2$-$J_3$ Heisenberg model on a square lattice for $J_3 = -1$. Symbols indicate the ground state found in exact diagonalization of a 32-site cluster, allowing for twisted boundary conditions: FM — ferromagnetic; $S(q,q)$ or $S(q,0)$ — spiral; $SR$ $S(q,0)$ — sort-range order (see text); $C(\pi/2,0)$, $C(\pi/2,\pi/2)$ or $C(\pi,0)$ — collinear; $d$-$BN$ — quadrupolar (bond-nematic) state; $SG$ — spin-gapped. Dashed lines are the classical ($S = \infty$) phase boundaries.

of $N = 16, 20, 32, 36$ spins with periodic boundary conditions (PBC), and $N = 16, 20, 32$ spins with twisted boundary conditions (TBC), complemented by spin wave analysis following previous calculations of Chubukov [7] and Rastelli et al. [8, 9].

We find a wide variety of ground states, including both collinear and spiral phases, and states with no conventional magnetic order. For the case of FM $J_1$ our present phase diagram is a refinement of that proposed in [6]. Further support has been found for the suggestion that, bordering the $S(q,q)$ phase for $0.25 \lesssim J_2 \lesssim 0.7$, $S(q,0)$, spiral order is unstable against a new form of order. For the case of AF $J_1$ we find a picture for the nature of disordered phase next to the $Q = (\pi,\pi)$ slightly different from the one proposed in [1] (and recent series expansion calculations [10]): mixed columnar-plaquette VBC order as proposed in [11], with possibly pure columnar VBC order for $J_2 \gtrsim 0.5J_1$ (as suggested by recent variational calculations based on entangled pair states [12]) and provide evidence for additional new phases at larger values of $J_3$. In this paper we limit ourselves to the ground-state in zero magnetic field. The study of magnetization plateaux will be reported in a further publication.

2. Ground-state phases

Our main ED results for the spin-1/2 model are summarized in the phase diagrams Fig. 1 (FM $J_1$) and Fig. 2 (AF $J_1$) in which the boundaries of the spiral phases have been deduced from ED with TBC on $N = 32$ clusters, and our conclusions on the nature of the ground-state include the results of an analysis of the spectra obtained by ED of up to $N = 36$ spins. We note that these boundaries may differ from those of an infinite system.

Classically, the $J_1$-$J_2$-$J_3$ Heisenberg model on a square lattice has four magnetically ordered phases for $J_1$ FM or AF [8, 9]. These are collinear and spiral states at wave vector $Q_{cl}$ which extend over the regions labelled I-IV in Fig. 1 (FM $J_1$) and Fig. 2 (AF $J_1$):

I – a FM phase with $Q_{cl} = (0,0)$ for FM $J_1$, a collinear phase with $Q_{cl} = (\pi,\pi)$ for AF $J_1$,

II – a collinear phase with $Q_{cl} = (\pi,0)$ or $(0,\pi)$,
Figure 2. (Color online). Groundstate phase diagram for $J_1 = 1$. Symbols indicate the ground state found in exact diagonalization of a 32-site cluster, allowing for twisted boundary conditions: $S(q, q)$ — spiral; $SR S(q, \pi)$ — sort-range order (see text); $C(\pi, \pi)$, $C(\pi/2, \pi)$, $C(\pi/2, \pi/2)$ or $C(\pi, 0)$ — collinear; $MP-VBC$ — mixed columnar-plaquette VBC state; $C-VBC$ — columnar VBC state; $SG$ — spin-gapped. Dashed lines are the classical ($S = \infty$) phase boundaries.

III — a spiral phase with $Q_{cl} = (q, 0)$ or $(0, q)$ where $q = \cos^{-1}\left[-\frac{2J_2 + J_1}{4J_3}\right]$ for FM $J_1$, and $Q_{cl} = (q, \pi)$ or $(\pi, q)$ with $q = \cos^{-1}\left[\frac{2J_2 - J_1}{4J_3}\right]$ for AF $J_1$,

IV — a spiral phase with $Q_{cl} = (q, q)$ or $(q, -q)$ where $q = \cos^{-1}\left[-\frac{J_1}{2J_2 + 4J_3}\right]$ (so that $q \in [0, \pi/2]$ for FM $J_1$ and $q \in [\pi/2, \pi]$ for AF $J_1$).

These magnetic orders will be referred as FM, $C(\pi, \pi)$, $C(\pi/2, \pi)$, $S(q, 0)$ $S(q, \pi)$, and $S(q, q)$ in this paper.

At a classical level, all transitions are continuous since at the special point $J_2 = 0.5$, $J_3 = 0$, and on the boundary between III and IV, the classical ground state becomes degenerate with a family of spirals which interpolate continuously between the competing values of $Q_{cl}$, and appear as additional zero modes in linear spin wave theory (LSW) calculations. As a result of the high density of low-lying excitations, LSW predicts that the sublattice magnetization of the classically ordered states vanishes in the vicinity of all boundaries (except the boundary of the FM phase). Treated more accurately, quantum fluctuations may act to modify the wave vectors of spiral ground states [7, 8, 9], enhance the extension of AF collinear phases, or lead to entirely new forms of order. The resulting phase transitions will generally be 1st order.

2.1. Phase diagram for FM $J_1$

The phase diagram of Fig. 1 only differs from the one presented in [6] in the region $0.25 \lesssim J_2 \lesssim 0.7$ below the $S(q, q)$ phase which we now label as $SR S(q, 0)$ instead of $S(q, 0)$. There, as already noted in [6], the spectra of the $N = 32$ cluster at the twist angle that minimize the energy do not exhibit a well defined tower of state characteristic of spiral order. Moreover, in this region, the energy is a very flat function of the twist angles over a large region of twists which bridge between twists that allow to accommodate a $S(q, 0)$ and a $S(q, q)$ spiral. This region is indeed close the line boundary between classical regions III and IV which is a line of degenerate spirals interpolating between $S(q, 0)$ and $S(q, q)$ spirals. Spiral order appears thus
quite unstable and another kind of ground-state occurs with short-range spin-spin correlations. In view of the spectra, this ground-state does not exhibit a p-nematic order that correspond to the selection of a preferential plane for the spins as found in [13] and is probably spin-gapped.

2.2. Phase diagram for AF $J_1$

The phase diagram in the case of AF $J_1$ is shown in Fig. 2. The extension of the spirals is there also much reduced, and the $S(q, \pi)$ order is completely suppressed. One has in addition a region, labelled $SR S(q, q)$, possibly spin-gapped, where the energy is minimized for TBC but the spectra does not show spiral order. This region is located between the VBC and $S(q, q)$ where it has been proposed that a $Z_2$ spin-liquid occurs [14]. The wave vector of the $S(q, q)$ spiral is also shifted by quantum fluctuations [6]. The SG state is similar to the one found for FM $J_1$. The collinear $C(\pi, 0)$ and $C(\pi, \pi)$ phases clearly extends beyond their classical boundaries.

The main difference is the appearence of a spin-gapped phase with likely VBC order. In region adjacent to the $C(\pi, \pi)$ phase, this VBC phase exhibit, in addition to columnar dimer-dimer correlations, strong plaquette correlations, largest around the line $(J_2 + J_3)/J_1 = 1/2$ and with decreasing $J_2$, as previously pointed in [1]. However an analysis of the symmetry properties of the low lying singlets present in the spectra and their evolution with the size of the clusters indicates that an eight-fold degenerate mixed plaquette-columnar order, as proposed in [15] and recently in [11], or even possibly a more complex pattern, is much more likely than a purely plaquette VBC order. Moreover both the spectra and correlations seems to favor a (quasi) pure columnar (four-fold degenerate) VBC order for $J_2 \geq 0.5J_1$.

For $0.5 \lesssim J_2/J_1 \lesssim 0.75$ and $J_3/J_1 \lesssim 0.35$, we found a region where the spectrum that suggests (four-fold degenerate) $C(\pi/2, \pi)$ collinear order. This state is one of the classical ground-states at $J_2/J_1 = 0.5$ for $0 < J_3/J_1 < 0.25$, degenerate with the $S(\pi/2, \pi)$ spiral. But quantum fluctuations may not be sufficiently strong to stabilize this collinear state by an order by disorder mechanism, at variance with the FM $J_1$ case, where one nicely sees the signature of the similar $C(\pi/2, 0)$ order. In the AF case, such a collinear state seems in competion with VBC order which could gain more from quantum fluctuations. The ground-state may be spin-gapped with possibly columnar VBC order, as suggested the correlations and the symmetry of the lowest singlets (which are the same as those of the singlets of the Anderson tower of the $C(\pi/2, \pi)$) or even could be a spin-liquid. The nature of the ground state in this region and the regions labelled $SR S(q, 0)$, $SR S(q, \pi)$ or $SR S(q, q)$ deserves further investigations.

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