Phase diagram of the spin-1/2 \( J_1-J_2-J_3 \) Heisenberg model on the square lattice with ferromagnetic \( J_1 \)

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Abstract.

Phase diagram of the \( S = 1/2, J_1-J_2-J_3 \) Heisenberg model on the square lattice with ferromagnetic 1\(^{st}\) neighbor and antiferromagnetic 2\(^{nd}\) and 3\(^{rd}\) neighbor interactions is studied by means of exact diagonalization and spin wave calculations. Quantum fluctuations are shown to induce phases that are not present classically and a shift in the wave vector of the spiral phases. Results are compared with recent experimental data.

1. Introduction

Research in frustrated magnetism has traditionally focused on systems with competing — or geometrically frustrated — antiferromagnetic (AF) interactions. Recently however, a number of systems have come to the fore in which ferromagnetic (FM) and AF interactions compete, giving rise to qualitatively new quantum ground states. Among these are several classes of layered vanadate and cuprate compounds, which are well approximated by a spin-1/2 Heisenberg model on a square lattice, with FM interactions on the nearest-neighbor \( (J_1) \) bonds [1, 2].

In order to better understand these materials, we have performed an extensive numerical investigation of the \( J_1-J_2-J_3 \) Heisenberg model on a square lattice, concentrating on the case where ferromagnetic \( J_1 \) competes with antiferromagnetic second and third neighbor interactions \( J_2 \) and \( J_3 \)

\[
\mathcal{H} = J_1 \sum_{(ij)1} S_i \cdot S_j + J_2 \sum_{(ij)2} S_i \cdot S_j + J_3 \sum_{(ij)3} S_i \cdot S_j - h \sum_i S_i^z, \tag{1}
\]

both with and without magnetic field \( h \). We set \( J_1 = -1 \) throughout this paper.

The principle methods used are exact diagonalization (ED) and classical Monte Carlo (MC) simulations, complemented by spin wave analysis. In this proceedings paper we present preliminary results for the phase diagram of Eq. 1 for \( h = 0 \), as determined by ED calculations. We find a wide variety of ground states, including both collinear and spiral phases, and states with no conventional magnetic order. Connection is made with previous spin wave calculations of Chubukov [3] and Rastelli \textit{et al.} [4, 5] and, where possible, with experiments.

We also briefly discuss some interesting aspects of Eq. 1 in applied magnetic field, notably an \( m = 1/3 \) magnetization plateau occurring for \( J_1 = -1, J_2 \approx 1, J_3 \approx 0.5 \), similar to that recently
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 $J_1 = -1$, $h = 0$. 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; $C(\pi/2, 0)$, $C(\pi/2, \pi/2)$ or $C(\pi, 0)$ — collinear; $d$-BN — quadrupolar (bond-nematic) state; $SG$ — spin-gapped. Dashed lines are the classical ($S = \infty$) phase boundaries.

observed in (CuBr)${}_2$Sr$_2$Nb$_3$O$_{10}$ [7]. Classical MC simulation results for this magnetization plateau will be presented elsewhere in these proceedings [8].

2. Ground-state phases

Our main ED results for the spin-1/2 model are summarized in the phase diagram Fig. 1. Classically, the $J_1$-$J_2$-$J_3$ Heisenberg model on a square lattice has four magnetically ordered phases for FM $J_1$ [4]. These are collinear and spiral states at wave vector $Q_{cl}$ which extend over the regions labelled I-IV in Fig. 1:

I — a FM phase with $Q_{cl} = (0, 0)$,

II — a collinear phase with $Q_{cl} = (\pi, 0)$ or $(0, \pi)$,

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

IV — a spiral phase with $Q_{cl} = (q, q)$ or $(q, -q)$ where $q = \cos^{-1}\left[\frac{-J_1}{2J_2+4J_3}\right]$.

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

At a classical level, all transitions are continuous. However at the special point $J_2 = 0.5$, $J_3 = 0$, and on the boundary $J_3 = J_2/2$, $J_3 > 0.125$ between the $S(q, 0)$ and $S(q, q)$ spiral ground states, the classical ground state becomes degenerate with a family of spirals. These 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 boundaries I-II, II-III and III-IV. Treated more accurately, quantum fluctuations may act to stabilize classical order, modifying the wave vectors of spiral...
Figure 2. (Color online). Incommensurate wave vector $q$ in the spiral phase $S(q,q)$ obtained from exact diagonalization of the $N = 32$ cluster. Left : $q$ vs $J_2$ at constant $J_3$, the lines are $q_{cl} + \delta q(J_2, J_3 = 0.5)$ where $\delta q = q - q_{cl}$ is the deviation from the classical value, inset shows $\delta q(J_2, J_3 = 0.5)$ vs $J_2$. $\delta q$ is seen nearly independent of $J_3$, except at large $J_3$ close to the $C(\pi/2, \pi/2)$ phase and in the vicinity of the spin-gapped SG phase. Right : $q$ vs $J_2$ at constant $J_3$. The lines are $q_{cl} + \delta q(J_2 = 0.35, J_3)$ if $J_3 = 0.35, 0.4$, and $q_{cl} + \delta q(J_2 = 0.7, J_3)$ if $J_3 = 0.5, 0.6$. $\delta q$ varies with $J_2$ and increases sharply in the vicinity of the SG phase.

Ground states [3, 4], or lead to entirely new forms of order. The resulting phase transitions will generally — but not always — be 1st order.

We have investigated the extreme quantum limit spin-1/2, using ED for clusters of $N = 16, 20, 32, 36$ spins. In addition to periodic boundary conditions (PBC), we used twisted boundary conditions (TBC) for the $N = 32$ cluster, in order to investigate possible spiral phases. The ED results (see Fig. 1) suggest that quantum fluctuations do indeed lead to a strong modification of the classical phase diagram. We address some key features of the phase diagram for spin-1/2 below:

(i) The $S(q,q)$ phase shrinks in favor of phases stabilized by quantum fluctuations. The open squares in Fig. 1 mark points where the energy is minimized by a non zero twist in the $\mathbf{e}_x + \mathbf{e}_y$ direction. For a few values of $J_2, J_3$ we computed the spectrum and found that the lowest states in the low spin sectors forms a well defined set of “quasi degenerate joint states” (QDJS), below the others states. These QDJS are the signature of magnetic order. If $q$ is close to a wave vector present in the sample, the QDJS are visible for PBC and then consist of $2(2S + 1)$ energy levels in each spin sector (because this is a non-collinear phase and there are two directions for the spiral) with energies above the ground-state that scale as $\sim S(S + 1)/N$. Otherwise, the QDJS are seen only for TBC, and then include only one energy level in each $S_z$ spin sector that scales as $\sim S^2_z/N$.

(ii) Quantum fluctuations also modify the pitch of these spirals, leading to a shift in $q$, as shown in Fig. 2. The values of these shifts for $N = 32$ are in reasonable agreement with those predicted by spin wave calculations for a finite size system [4], except in the vicinity of the SG phase. Performing finite size scaling for systems with TBC is extremely challenging, and we have not attempted it here. However, from the spin wave calculations we estimate that the true shifts in $q$ will be roughly half as big in the thermodynamic limit.

(iii) As shown in Fig. 2, the shift in the wave vector $q$ in the $S(q,q)$ phase is nearly independent of $J_3$, and decreases with increasing $J_2$. It increases rapidly at the boundary with the SG
phase, where the transition is $2^{nd}$ or weakly $1^{st}$ order. At the border between the $SG$ and $S(q,q)$ phases, the energy is a very flat function of the twist angle in either the $e_x + e_y$ or $e_x$ directions, and may even be minimized for a non-zero twist in the $e_x$ direction, as for a $(q,0)$ spiral. This indicates that the $S(q,q)$ or $S(q,0)$ orders are quasi-degenerate and quite unstable. This is reminiscent of the classical degeneracy on the border between phases III and IV, which was predicted to gain a tiny but finite extension in spin wave calculations including $1/S$ corrections for $0.5 \gtrsim J_2$ [4]. It remains to be established, however, that such a “quantum helix” really survives in the thermodynamic limit. At the border of the $S(q,q)$ phase with the $C(\pi/2,0)$ and $C(\pi/2,\pi/2)$ phases, $q$ jumps discontinuously. The transitions are there clearly $1^{st}$ order.

(iv) Quantum fluctuations also eat into the FM phase for $0.25 \lesssim J_2 \lesssim 0.5$, where the transition out of the FM is $1^{st}$ order. For $J_2 \lesssim 0.25$ the expected $2^{nd}$ order phase transition from FM to $S(q,q)$ occurs with $q \to 0$.

(v) In the range $0.4 \lesssim J_2 \lesssim 0.7$, we find a nematic (i.e. quadrupolar) phase with d-wave symmetry, formed from $S = 1$ triplets on FM $J_1$ bonds, and denoted $d$-$BN$ in this paper. This extends up to quite large $J_3$, at the expense of the FM, $S(q,0)$ and the $C(\pi,0)$ phases. This $d$-$BN$ phase, already identified in the $J_1 - J_2$ model [10], results from the condensation of bound bi-magnons. Bi-magnon bound states first condense out of the saturated state at the critical magnetic field. The binding energy of bi-magnons, which we calculated exactly, is shown in Fig. 3, and takes on its highest values in the regions of high classical ground state degeneracy.

(vi) The transition from the FM and $C(\pi/2,\pi/2)$ phases into the $d$-$BN$ is (empirically) first order. However a continuous transition may still occur between the $C(\pi,0)$ and $SG$ phases and the $d$-$BN$, through the condensation of spin-2 bound states.

(vii) The $d$-$BN$, $C(\pi,0)$ and $SG$ phases have, at first sight, very similar spectra. The $d$-$BN$ phase breaks spin rotation symmetry and has a set of QDJS rather like those of the $C(\pi,0)$ Néel phase. However only the even spin gaps scale as $1/N$ (see Fig. 4 of Ref. [10]), indicating a different form of magnetic order. Finite size scaling also distinguishes the $SG$ phase, where the gaps to the lowest states in spin sectors $S \geq 1$ exhibit a quite different (irregular) dependence on $N$, and appear to extrapolate to a finite value. In addition, the spectra for the $SG$ phase possess additional low-lying singlet states.

(viii) Since the extrapolation of the gaps are subject to some uncertainties with ED calculations...
Figure 4. (Color online). Left: Dimer-dimer correlations at $J_1 = -1, J_2 = 1, J_3 = 0.5$ for $N = 32$, between the reference pair of sites $1 - 10$ and other diagonal pairs of nearest neighbor sites. The width is proportional to the magnitude of the correlations. Positive (negative) values are indicated by full (dashed) lines. Right: A candidate VBS state in the $SG$ phase, formed from two systems (red and blue) of columnar dimers on diagonal bonds.

limited to $N = 36$ sites, the boundaries between the $d$-$BN$, $SG$ and $C(\pi, 0)$ phases are difficult to locate accurately. The point $(J_2, J_3) = (0.7, 0.25)$ or the point $(0.6, 0.2)$ in Fig. 1, denoted as $d$-$BN$ could be already in the $SG$ phase; the point $(0.6, 0)$ shown as $d$-$BN$ is at the border of the $C(\pi, 0)$ phase, and the points $(0.7, 0.125)$ and $(1, 0.2)$ marked as $C(\pi, 0)$ could be in the $SG$ or perhaps the $d$-$BN$ phase.

(ix) We tentatively conclude from ED that the $C(\pi, 0)$ phase extends beyond its classical boundary into the region III. LSW theory predicts a vanishing sublattice magnetization approaching the classical $S(q, 0)$ state. However interacting spin wave results for the the spin-1/2 $J_1$-$J_2$-$J_3$ model with AF $J_1$ suggest that $C(\pi, 0)$ phase is stabilized by fluctuations at the expense of spiral ground state [5, 11]. Further investigation of the boundaries between the $d$-$BN$, $C(\pi, 0)$ and $SG$ phases is underway.

(x) The $S(q, 0)$ phase shrinks to a small pocket on the border of the FM phase where the transition is 2$^{nd}$ order and a pocket adjacent to $S(q, q)$ phase for $0.25 \lesssim J_2 \lesssim 0.7$ where the ED spectra suggest however that such an order may be unstable and another kind of ground-state, possibly spin-gapped, may occurs.

(xi) In part of region III, the $S(q, 0)$ phase is replaced by a collinear phase with wave vector $(\pi/2, 0)$ or $(0, \pi/2)$, noted here $C(\pi/2, 0)$, which also extends into the adjacent regions IV and I. The presence of this phase is clearly identified in the ED spectrum by the presence of a characteristic set of QDJS. This phase occurs for $J_2$ close to 0.5 where the classical value of $q$ is $\pi/2$. $Q_d$ is there 1/4 of a reciprocal wave vector. For classical spins, as noted by Villain [9], such a spiral can be continuously transformed into a collinear state at no cost in energy. The present results show that the collinear state can be stabilized by quantum fluctuations, strongest in a collinear phase, over a significantly large domain. For large $J_3$ the $S(q, q)$ phase is replaced by a collinear $C(\pi/2, \pi/2)$ phase which is stabilized by quantum fluctuations by the same mechanism that favors the $C(\pi/2, 0)$ phase. The extension of these phases may somewhat shrink in the thermodynamic limit, but is likely to remain finite. The transitions out of these phases are all 1$^{st}$ order.

(xii) On both side of the classical border line ($J_3 = J_2/2$) between III-IV, the $S(q, 0)$ and $S(q, q)$
phases are replaced for \( J_2 \gtrsim 0.7 \), by a spin-gapped \( SG \) phase. It is difficult to conclude as to the precise nature of the \( SG \) phase because of the large and irregular finite size effects. However the extrapolated triplet gap is quite large, and the ground state energy is higher for \( N = 20 \) and \( N = 36 \) clusters than those with \( N = 16 \) and \( N = 32 \). This suggests a valence bond solid (VBS) state with \( 8- \) or \( 16- \) site unit cell. For large \( J_3 \), near the line \( J_3 = J_2/2 \), the system could be thought of as two independent AF \( J_1-J_2 \) models, each in a columnar dimer phase, weakly coupled by FM \( J_1 \). Dimer correlation functions within the \( SG \) phase (see Fig. 4-left) are indeed compatible with a VBS state formed of columnar dimers on \( J_2 \) bonds (see Fig. 4-right), and four low-lying singlet states, potentially associated with this order, are seen in associated spectra. However it is difficult to draw any firm conclusion about the presence of this VBS order in the absence of reliable finite size scaling. The precise nature of this \( SG \) phase therefore remains uncertain, and deserves further investigation.

(xiii) In the \( SG \) phase, for \( J_2 = 1 \) and \( J_3 = 0.5 \), ED results for \( N = 36 \) suggest a \( m = 1/3 \) magnetization plateau in an applied magnetic field, where the spins display a ferrimagnetic order at \( Q = (2\pi/3, 0) \). This magnetization plateau is also present in the classical model where it is stabilized by thermal fluctuations [8]. It is also possible \( a \) priori that different forms of plateaux occur for other ratios of \( J_2/J_3 \), for example above the \( S(q, q) \) ground state. This remains a subject for future investigation. We note that, for \( J_2 = 1 \) and \( J_3 = 0.5 \) (and for a wide range of parameter space), the system displays \( d-BN \) order at fields approaching saturation.

3. Comparison with experiments and conclusions

Recent investigations of a number of compounds which realize spin-1/2 on a square lattice have revealed a small or negative Curie-Weiss temperature, suggestive of competing FM and AF interactions. Among them, (CuBr)LaNb\(_2\)O\(_7\) [6] displays collinear AF order with momentum \((\pi, 0)\) and can be presumably described by the \( C(\pi, 0) \) phase in a \( J_1-J_2 \) model or a \( J_1-J_2-J_3 \) model with small \( J_3 \). A simple \( J_1-J_2 \) model is however insufficient for the related compounds (CuCl)LaNb\(_2\)O\(_7\), which exhibits a spin gap [2], and (CuBr)Sr\(_2\)Nb\(_3\)O\(_{10}\), which displays a \( m = 1/3 \) magnetization plateau [7]; a \( J_1-J_2-J_3 \) model might therefore be considered. The presence of a spin gap in (CuCl)LaNb\(_2\)O\(_7\) would then place this compound in the \( SG \) region, but probably away from the point \((J_2 = 1, J_3 = 0.5)\), since this compound does not show a magnetization plateau. The effect of the lattice distortion on the couplings, which appears at low temperature in this compound, needs however to be understood. (CuBr)Sr\(_2\)Nb\(_3\)O\(_{10}\) has no spin-gap and a (small) positive Curie-Weiss temperature. This suggests that values of the AF couplings are not large and places the compound outside the \( SG \) region. Further investigation of the appropriate model for these compounds is underway.

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