QUANTUM PHASES IN TWO-DIMENSIONAL FRUSTRATED SPIN-1/2 ANTIFERROMAGNETS

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We describe four phases found in two-dimensional quantum antiferromagnets. Two of them display long range order at $T = 0$: the Néel state and the Valence Bond Crystal. The last two are Spin-Liquids. Properties of these different states are shortly described and likely conditions of their occurrence outlined.

1 INTRODUCTION

Investigating various spin-1/2 systems in two dimensions (2d), by means of exact diagonalization (ED) on small samples of $N$ spins, we have found, up to now, four kinds of antiferromagnetic phases at $T=0$. Two of them display long range order (LRO): Néel or Valence Bond Crystal (VBC) LRO. The others are two different Spin-Liquids. The Néel case may be qualified as semi-classical: it is a ground-state of the (spin-$\infty$) classical model with an order parameter reduced by quantum fluctuations. The others are purely quantum phases.

The hamiltonians considered are SU(2) invariant, like the Heisenberg hamiltonian and its generalizations as the $J_1-J_2$ model or the $J_1-J_2-J_3$ model and the multiple-spin exchange (MSE) model. All these models of magnetism picture exchange (or super-exchange) between fermions. The permutation operator of two spins reads:

$$P_{ij} = 2S_i \cdot S_j + \frac{1}{2}$$

This yields the Heisenberg hamiltonian:

$$H = \sum_{<ij>} S_i \cdot S_j$$

where the sum runs over nearest neighbor pairs of spins on sites $i$ and $j$, the $J_1-J_2$ model:

$$H = J_1 \sum_{<ij>} S_i \cdot S_j + J_2 \sum_{<<ij>>} S_i \cdot S_j$$

where the second sum runs over next-nearest neighbor pairs and the $J_1-J_2-J_3$ model with additional 3th nearest neighbor interrations. We were also motivated to study MSE models involving exchange of more than two spins. Among them we shall focus on the simplest which brings new physics which involves up to 4-spin exchange. Its hamiltonian reads:

$$H = J_2 \sum_{<ij>} P_{ij} + J_4 \sum_{<ijkl>} (P_{ijkl} + P_{ijkl}^{-1})$$
where $P_{ijkl}$ stands for the permutation of 4 spins on sites $i, j, k, l$, $P_{ijkl}^{-1}$ is the inverse permutation (note that $P_{ij} = P_{ij}^{-1}$). The 2d lattices are the square, honeycomb, triangular and kagome lattices.

In order to distinguish between the different phases of such models, from ED calculations on small samples (of up to $N = 36$ spins with present day computers), the analysis of the symmetries and scaling properties of the low-lying eigen-levels of the spectra is a very efficient approach. Typical spectra of systems in the four phases identified are displayed in Fig. 1 as a function of the eigenvalues $S(S + 1)$ of the square $S^2$ of the total spin (a good quantum number as the hamiltonians are $SU(2)$ invariant). A superficial analysis allows an immediate detection of obvious distinctive features:

- **Fig. 1a:** Neél spectrum, the lowest eigen-levels in each $S$ sector form a separate set with an energy increasing as $S(S + 1)$ (the quantum origin of the Landau free energy per spin $f$ increasing with the magnetization $m = S/(N/2)$ as $f(m) = f(0) + m^2/2\chi + \cdots$ for small $m$, where $\chi$ is the spin susceptibility).

- **Fig. 1b and 1c:** VBC or type I Spin-Liquid spectra with a gap $\Delta$ between different sector of spins ($f(m) = f(0) + \Delta m/2 + \cdots$)

- **Fig. 1d:** type II Spin-Liquid spectrum with a spin-gap and a gapless continuum of singlets excitations

We now briefly sketch the spectral properties of the four different phases and outline the groups of systems where they have been found.

## 2 Semi-classical Neél phase

Neél LRO breaks $SU(2)$ and various space symmetries at the thermodynamic limit. This is revealed in the spectra by a basic set of low-lying eigen-levels (states on the dashed line in Fig. 1a), called QDJS for Quasi Degenerate Joint States in Refs., with energies $\sim S(S + 1)/2N\chi$ at least for $S$ values up to $\sqrt{N}$. They collapse together to the absolute ground-state as $1/N$ and separate from the softest magnons (dotted line in Fig. 1a) which decrease as $1/\sqrt{N}$. The QDJS have different $S$ values which enable breaking of $SU(2)$ symmetry. The number of QDJS per $S$ value is characteristic: it is one for collinear LRO and equal to the number of way of adding $p$ spins of size $N/2p$ for noncollinear LRO with $p$ sublattices. The QDJS belong to different irreducible representations of the space group of the lattice which allow and reveal the space symmetry breaking occurring. For instance, in the case of the Heisenberg model on the square lattice of Fig. 1a, where translation symmetry is broken, the QDJS have wave vectors $k = (0, 0)$ for even $S$ and $k = (\pi, \pi)$ for odd $S$ values and are invariant under all operations of the point group (taking the origine on a site), as expected for this collinear Neél LRO. ED and/or Monte-Carlo calculations have shown that the Heisenberg model on the 2d bipartite lattices investigated up to now, square (see Refs. and references therein), honeycomb, 1/5 depleted square, · · ·, displays collinear Neél LRO. In these unfrustrated systems, Monte-Carlo methods can deal now with very large systems which enable
Figure 1. Eigen-energies vs $S(S+1)$ of $N = 36$ samples. (a) Spectrum of the Heisenberg model on the square lattice, characteristic of collinear Néel LRO: the lowest energies in each spin sector $S$ increase as $S(S+1)$ with $S$ (states on the dashed line) and are well separated from the softest magnons (states on the dotted line). (b) Spectrum of the Heisenberg model on the checkerboard lattice, which displays VBC LRO: for $N \to \infty$, the two lowest $S=0$ states (crosses) become degenerate and the gap to the others states remains finite, the finite spin-gap implies that $SU(2)$ remains unbroken. (c) Spectrum of the MSE model (eq. 4 with $J_2 = -2$, $J_4 = 1$) on the triangular lattice, a Type I Spin Liquid: four $S=0$ states (crosses) become degenerate as $N \to \infty$, the excitations above the degenerate ground-state remain gapped. (d) Spectrum of the Heisenberg model on the kagomé lattice, a Type II Spin Liquid: there is a spin-gap and a gapless continuum of singlet states.

the investigation of properties out of range of the ED approach. Monte-Carlo approaches, however, is limited by the well-known sign problem in frustrated
systems like those considered below.

A similar analysis of the ED spectra has shown that the Heisenberg model on the triangular lattice displays noncollinear Néel LRO. It also revealed occurrences of "order by disorder" in the $J_1 - J_2$ model on the triangular lattice and the $J_1 - J_2 - J_3$ model on the honeycomb lattice. In these situations the classical ground-state is a degenerate manifold of different kinds of 4-sublattice Néel order. Evidence of the selection of a collinear LRO by quantum fluctuations appears when increasing $N$, with the separation of the QDJS specific of collinear LRO from the states associated with 4-sublattice order.

Calculations of spin-spin correlations, order parameters, spin susceptibilities, stiffnesses yield results consistent with the spectrum analysis but their finite scaling behavior is less favourable, (i.e. $\sim 1/\sqrt{N}$ in Néel systems) and extrapolation to $N \to \infty$ could be rather inaccurate. Agreement between the scaling behaviors of ED and spin wave results can nevertheless help to circumvent this limitation.

3 VBC LRO

This class includes systems with LRO of singlet objects such as singlet bonds (dimers) or more extented ones (4-site plaquettes...). This kind of LRO usually breaks the space symmetry, so the spectrum must display a corresponding degeneracy in the thermodynamic limit, but it does not break $SU(2)$ which manifest by a finite spin-gap. Such features are visible in Fig 1 which displays the spectrum of the Heisenberg model on the checkerboard lattice: two $S = 0$ states appear in the bottom of the spectrum well separated from all others. A finite size analysis confirm the degeneracy of the two states and that the singlet and the spin gaps remain finite for $N \to \infty$. In the present case, symmetry properties of the two states indicate unambigously LRO of plaquettes on the void squares of the checkerboard lattice, which is confirmed by calculations of correlation functions. Since the excitations are separated from the degenerate ground-state by a gap $\Delta$, the thermal behavior of the spin susceptibility $\chi$ and heat capacity $C_v$ is exponentially activated $\sim \exp(-\Delta/kT)$ at low $T$. Due to the gap $\Delta$, the ground-state energy in the different spin sector $S$ evolves as $E(S) \approx E_0 + \Delta S + \cdots$ at low $S$. This leads, in a continuous Landau description to $f(m) \approx f(0) + \Delta m/2 - mh/2 + \cdots$ in the presence of a magnetic field $h$: a finite field $h_c = \Delta$ is needed to induce a transition to states with a non zero magnetization, the magnetization curve $m(h)$ starts with a plateau at zero $m$.

Much attention has been given to the $J_1 - J_2$ model on the square lattice which for $J_2/J_1 \sim 0.5$ has been suspected to display dimer or plaquette LRO, but the issue is still debated (see references therein). Besides the checkerboard lattice, we have found a VBC phase in the $J_1 - J_2$ model on the honeycomb lattice. In these three systems the VBC phase is located in the phase diagram near a collinear Néel phase. VBC LRO appears as a generic situation to be expected after destabilization by quantum fluctuations of a collinear Néel phase with increased frustration. This is in agreement with various analytical calculations, based on large-$N$ $SU(N)$ or $Sp(N)$ expansions or non linear sigma-model theory taking into account the Berry phases effects. But ED calculations, limited to small systems, does not allow
to investigate the phase transition between Néel and VBC LRO (see Refs. 17, 19 for recently proposed scenarios). Experimental evidence of VBC LRO has been found in the 2d insulating compounds: \( \text{SrCu}_2(BO_3)_2 \) and \( \text{CaV}_4O_9 \). These systems are however somewhat special. The 2d lattices of magnetic ions in these compounds have lower symmetries than the above-mentioned lattices. The Hamiltonian already favors the formation of singlet objects. There is no further symmetry breaking.

4 Type I Spin Liquid

The spectrum of type I Spin Liquid (see Fig. 1) seems at first sight very similar to the spectrum of a VBC. It has a finite gap above a degenerate ground-state. \( \chi \) and \( C_v \) are thermally activated and \( SU(2) \) is not broken. But we are lead to exclude VBC LRO because the few \( S = 0 \) levels collapsing together to the ground-state do not have symmetries that could be explained in the framework a VBC picture and all correlation functions are found short-ranged. Indeed, in the two systems where we think that type I Spin Liquid behavior occurs: for the MSE model on the triangular lattice \( 22, 23 \) and in the \( J_1 - J_2 - J_3 \) model on the honeycomb lattice \( \delta \) VBC LRO would require a larger degeneracy of the ground-state than the one observed. For instance, the simplest VBC LRO on the triangular lattice is 12-fold degenerate whereas the observed degeneracy is 4-fold. This 4-fold degeneracy is not the signature of LRO in a local order parameter but has a topological origin \( 28, 17 \).

Several aspects of the physics of type I Spin Liquids, still deserve further investigations. This fully gapped phase without VBC LRO is likely to be described with short ranged resonating valence bonds (RVB). A possible connection with the RVB phase of quantum dimer model, such as the one found on the triangular lattice \( \delta \) is worth to be clarified. In type I Spin Liquids, the excitations are expected to consist of deconfined spinons \( 13, 18 \). We are presently searching for numerical evidence of deconfined spinons. It also remains to firmly outline the conditions of appearance of type I Spin Liquids. Fully gapped Spin Liquids are predicted to occur after destabilization of non-collinear LRO \( 22, 24 \). The two examples of type I Spin Liquid, found up to now, do not disagree with this. But, as discussed below, type II Spin Liquid behavior with gapless excitations (not described in the approach of Refs. 17, 18), may appear after destabilization of non-collinear LRO. A further clue might be of importance: in the two type I Spin Liquids short range correlations are slightly ferromagnetic. In fact, they appears in the vicinity of a ferromagnetic phase.

Experimentally, this phase might be observed in the low density 2d layers of solid \( ^3\text{He} \) where there is evidence that the MSE model on the triangular lattice with coupling constants that might be in the good range, is realized (see refs. 23, 24 and references therein) and possibly in the Wigner crystal \( \delta \). It might also occur in the \( \text{BaCo}_2(\text{AsO}_4)_2 \) compound \( \delta \) likely to be described by the \( J_1 - J_2 - J_3 \) model on the honeycomb lattice with parameters close to those for which we found indications of this behavior.
Figure 2. Heisenberg model on the kagomé lattice. Logarithm of the number of states in the spin-gap (squares fitted by the full line) vs size $N$ of the samples. The observed behavior differs from the scaling law $\exp(bN^{n/(n+2)})$ deduced from a single mode description of the continuum with a dispersion law $\epsilon(k) = k^n$ for $n = 1$ (short-dashed line) and $n = 2$ (long-dashed line). The linear behavior observed here implies density of states increasing as $(...)^N$ and a $T=0$ residual entropy.

5 Type II Spin Liquid

This phase was first observed in the Heisenberg model on the kagomé lattice. Some possible physical realizations are $SrCrGaO$ or various jarosites which have kagomé planes of magnetic ions. Fig. 4 shows the spectrum of the $N = 36$ sample of the kagomé antiferromagnet. Two features, confirmed by finite size scaling, are visible. First, a finite, although small, spin-gap $\Delta$. So $SU(2)$ spin symmetry remains unbroken and $\chi$ is exponentially activated. Second, a gapless continuum of singlet levels filling the spin-gap. Furthermore, the number of singlet levels in the spin-gap increases exponentially with the size $N$ of the sample as shown in Fig 2. All the correlations (spin-spin, dimer-dimer, chiral-chiral, ...) are very small and short range.

The understanding of the low lying continuum of singlets is still incomplete but some progresses have been done in the last years. Recent ED calculations on a kagomé $N = 24$ sample depleted of two sites suggest that the elementary excitations likely consist of deconfined spinons: as shown in Fig 3, the spin-gap does not depend neither on the presence of the two nonmagnetic holes nor on their separation which suggest that spin-$1/2$ excitations are essentially free (un-
The low lying singlets could be related to a family of short range RVB dimer coverings of the lattice. On the other hand, they cannot be described as Goldstone modes associated to a quasi LRO of dimers. Assuming non-interacting modes with a dispersion law \( \epsilon(k) = k^n \), it is easy to obtain the internal energy and entropy versus temperature and thus the number of states below a given energy. The logarithm of this number of states scales as \( N \ln N \). Fixing the unknown constants from the measured value of the number of singlets in the spin-gap for \( N = 12 \), we see in Fig 2 that the mode description is an order of magnitude off at \( N = 36 \). The exponential increase in the number of eigenlevels in the finite energy interval below the spin-gap suggests a finite entropy per spin at \( T = 0 \). The same exponential increase in the number of eigenlevels in each spin sector can be at the origin of the anomalous glassy behavior "without chemical disorder" seen in several kagomé compounds on which dynamical mean-field theory approaches might shed light. A low lying continuum of singlets is consistent with the observation that the low temperature specific heat of \( \text{SrCrGaO}_3 \) is essentially insensitive to large magnetic fields. This picture is also compatible with the muons experiments of Uemura et al. and the elastic spin diffusion measurements of Lee et al.

Type II Spin-Liquid behavior might also occur in 3d, in pyrochlore compounds and gadolinium gallium garnet. Likewise, 2d systems of coupled 1d chains might exhibit Spin-Liquid like behavior. Lately, neutron scattering evidence for deconfined spinons on an anisotropic triangular lattice has been found.

For a long time, it was widely believed that type II Spin-Liquid behavior was associated to special lattices like the kagomé or pyrochlore lattices, which at the classical level display an infinite local ground-state degeneracy. We have now several counterexamples. The checkerboard lattice which is a 2d analog of the pyrochlore lattice and exhibit a large similar degeneracy at the classical level, was found to display VBC LRO. A kagomé-like spectra is observed on the triangular lattice when the 3-sublattice Néel order is destabilized by a frustrating 4-spin exchange and we have no evidence of an infinite local degeneracy at this point. We now conjecture that the "kagomé-like" phase is one of the possible scenario after destabilization.
a non-collinear LRO. At this point we have not made any difference between the criteria of appearance of the two Spin-Liquids. Attempting, to go further, one could speculate that type I Spin-Liquid behavior might originate from resonating entities including near neighbor triplet pairs assembled in singlet entities of at least 4 spins, whereas type II Spin-Liquid involve essentially near neighbor singlet pairing. A last hint comes from the study of the spin-$S = 1$ kagomé magnet for sizes up to $N = 18$, which point to a rather large spin-gap void of singlets. It is in fact a common general idea that, due to topological reasons, half-integer and integer spin systems could differ. The difference between spin-1/2 and spin-1 kagomé spectra might possibly originate from this topological property.

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