A case study of spin-1 Heisenberg model in a triangular lattice

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(Dated: February 18, 2014)

We study the spin-1 model in a triangular lattice in presence of a uniaxial anisotropy field using a Cluster Mean Field approach (CMF). The interplay between antiferromagnetic exchange, lattice geometry and anisotropy forces Gutzwiller mean-field approaches to fail in certain region of the phase diagram. There, the CMF yields two supersolid (SS) phases compatible with those present in the spin-1/2 XXZ model onto which the spin-1 system maps. Between these two SS phases, the three-sublattice order is broken and the results of the CMF depend heavily on the geometry and size of the cluster, strongly suggesting the presence of a spin liquid.

PACS numbers: 75.10.Jm, 75.10.Kt

The quest for realistic magnetic quantum models that support topological and spin liquid ground states is at the frontier of current research in condensed matter and in quantum information science. These exotic states of matter do not display local order (i.e. do not break any local symmetry), but possess long-range entanglement encoding a global structure that cannot be detected by any local measurement. This absence of local order makes them robust against local perturbations, and thus, promising candidates for technological applications [1].

The triangular lattice is the archetype for classical geometric frustration. This phenomenon is believed to play a crucial role in the stabilization of certain spin liquid quantum phases, such as the resonating valence bond states proposed by Anderson [2]. Such states are not only interesting because of exhibiting this exotic global order, but also they are expected to be related, when doped, to non-conventional superconductivity.

The study of the spin-1 bilinear-biquadratic model in the triangular lattice in presence of single-ion anisotropy is at least two-fold motivated. On the one hand, the competition between dipolar and quadrupolar local magnetic ordering leads to a much richer physics than in spin-1/2 systems. Furthermore, when the uniaxial anisotropy is strong enough it changes the physics of the spin-1 system bringing it to an effective spin-1/2 frustrated model. On the other hand, recent experimental results on some complex crystals where this model might be present show an anomalous behavior which could be related to a spin liquid state [3]. Moreover, quantum systems other than real crystals could efficiently simulate this model [4]. For instance, ultracold atoms trapped in a triangular optical lattice [5].

In contrast to the $S = 1/2$ model in a triangular lattice, which has been thoroughly investigated [6], exact results for the $S = 1$ in this geometry are scarce, since powerful numerical techniques (e.g. DMRG, QMC) cannot be easily applied here. First attempts to describe the full phase diagram of the model have been based on a mean-field (Gutzwiller) approach, which explicitly assumes a three-sublattice ordering [7]. While this approach properly sketches locally ordered phases, it cannot describe states dominated by inter-site quantum correlations like spin liquids. In the absence of the anisotropy field, exact diagonalization (of up to 27-sites plaquettes) shows that three-sublattice order is always present, ruling out the presence of a spin liquid [8]. However, for a finite value of the anisotropy field, controversial results regarding the stability of spin liquid phases have appeared using variational MC [9] and mean-field analysis in the triplon fermionic spin representation [10, 11].

Aiming at investigating the above scenario, we study the $S = 1$ bilinear-biquadratic model in the presence of a uniaxial field in the triangular lattice using a Cluster Mean-Field (CMF) approach [13]. Within this approach, the spins in the lattice are grouped into small clusters which are exactly diagonalized, while each cluster is mean-field coupled to the other ones (see Fig. 1). This ansatz clearly constitutes a step forward as compared to the Gutzwiller mean-field approach since it explicitly treats quantum correlations between all sites belonging to the same cluster. Undoubtedly, it goes also beyond the results obtained from the exact diagonalization of an isolated plaquette of the same size.

Before proceeding further with the details of our study, we briefly outline here our major findings: (i) the CMF provides corrections to some continuous quantum phase boundaries obtained with the Gutzwiller approach, (ii) the CMF converges where the Gutzwiller ansatz fails, i.e. in the regions where only quantum correlations can break the non-trivial degeneracy of the classical model onto which it maps, and shows two distinct supersolid (SS) phases that might continuously connect to those for the spin-1/2 XXZ model in the triangular lattice, and (iii)
between the two SS phases we find a region where our results strongly depend on the cluster geometry and size, indicating the crucial role of quantum correlations there. The instability of our method in this region strongly suggests, very much in the spirit of the instabilities shown by generalized spin wave theory in anisotropic triangular lattices in spin-1/2 systems [13], the possible presence of a spin liquid phase and bounds quite precisely this region as compared to previous results [11].

\[ H_{\text{eff}} = H_c + \sum_{c' \neq c} H_{c'c}(|\psi_{c'}\rangle) \]  

FIG. 1: (Color Online). The CMF method works by coupling via mean-field distinct clusters that are exactly diagonalized. Thick lines: quantum bonds between clusters. Colors indicate the 3-sublattice order.

The model: The most general rotationally invariant Hamiltonian for spin-1 particles reads:

\[ H_{\text{eff}}(\theta) = \cos \theta \sum_{\langle i,j \rangle} S_i \cdot S_j + \sin \theta \sum_{\langle i,j \rangle} (S_i \cdot S_j)^2 \]  

where \( S = (S^x, S^y, S^z) \) are the usual spin \( S = 1 \) operators and \( \langle i,j \rangle \) runs over all possible pairs of first neighboring sites of the triangular lattice. The presence of a uniaxial field breaks the \( SU(2) \) symmetry

\[ H(\theta, D) = H_{\text{eff}}(\theta) + D \sum_i (S_i^z)^2. \]  

In the thermodynamic limit the system can display both, dipolar and/or quadrupolar magnetic order, associated to the expectation values of \( \langle S \rangle \) and \( \langle Q \rangle = \langle (S_x^2 - S_y^2, 2S_z^2 - S_x^2 - S_y^2)/\sqrt{3}, \{S_x, S_y\}, \{S_y, S_z\}, \{S_z, S_x\} \rangle \). As it can be easily shown \( (S_i \cdot S_j)^2 = (Q_i \cdot Q_j - S_i \cdot S_j)/2 \) and thus, the quadrupolar vector appears as a meaningful order parameter. In the CMF, the effective Hamiltonian acting on a cluster \( c \) is given by:

Here the expectation values are evaluated over the self-consistent ground state \( |\psi_c\rangle \). We proceed as follows: starting with a random initial state for each cluster we iteratively solve \( H_{c'c} \) until convergence is achieved. We use clusters of different geometries and sizes (\( \sim 3-10 \) sites). In order to guarantee the presence of three-sublattice order when the system displays it, the geometry and size of the clusters have to be chosen appropriately. For example, for a 9-sites cluster, a single configuration for all the plaquettes is sufficient but for the 7-sites cluster we must use 3 different initial plaquettes to ensure that three-sublattice order can be recovered (see Fig. 1). Also a major effect of coupling the plaquettes with the others (which act as an environment) is that certain symmetries (e.g., rotational invariance) are spontaneously broken, similarly to what happens in the thermodynamic limit. As a consequence, the CMF allows for a direct evaluation of the local order parameters \( \langle S \rangle \) or \( \langle Q \rangle \), in sharp contrast to the exact diagonalization of a single cluster. For the latter one evaluates instead the dipolar and quadrupolar structure factors which take into account account spin-spin and quadrupolar-quadrupolar correlations respectively, as done in [3] for the \( D = 0 \) case. On the other hand, the Gutzwiller ansatz in the triangular lattice is constructed by three independent fields \( (|\psi_A\rangle, |\psi_B\rangle \) and \( |\psi_C\rangle \) at each of the sublattices \( \lambda \). Within this approach, and because any quantum correlation between sites is neglected, the local parameters always fulfill \( \langle S \rangle^2 + \langle Q \rangle^2 = 4/3 \), and therefore, phases are always locally ordered in one way or another. This ansatz fails to describe phases without three-sublattice order and/or dominated by quantum correlations.

To test the performance of our CMF approach we start by revisiting the \( SU(2) \) symmetric Hamiltonian [1], whose ground state structure has been studied by means of the Gutzwiller ansatz [7] as well as by exact diagonalization [3]. Within the CMF approach three-sublattice order is also found at any value of the parameter \( \theta \). In particular, four phases with local magnetic order emerge: Ferro-Magnetic order (FM) for \( \theta \in (\pi/2, 3\pi/4) \), Ferro-Quadrupolar order (FQ) for \( \theta \in (5\pi/4, \pi) \), Antiferro-Magnetic order (AFM) for \( \theta \in (\pi/4, \pi/4) \), and Antiferro-Quadrupolar order (AFQ) for \( \theta \in (\pi/4, \pi) \). The Gutzwiller ansatz already yields the correct phases and for all but the FQ-AFM transition the phase boundaries coincide with those obtained by exact diagonalization. For the latter, the exact diagonalization and the CMF size scaling yield respectively \( \Theta_{\text{ED}} \approx -0.11\pi \) and \( \Theta_{\text{CMF}} \approx -0.17\pi \), while the transition with the Gutzwiller ansatz occurs at \( \Theta_{\text{Gutz}} = -0.35\pi \).

The presence of a uniaxial anisotropy field along an arbitrary axis, e.g., \( \hat{z} \)-axis, drastically changes the nature of the ground state. When the anisotropy does not dominate over the exchange coupling the system is far from being well understood. Let us first examine the already known limiting regimes.
Limit \( D \gg 1 \). For very large and positive uniaxial field the ground state is the product state \(|0,\ldots,0\rangle\), which has only quadrupolar order (FQ) at each site (large-\(D \) phase). The corresponding vector directors are oriented along \( \hat{z} \) and spin fluctuations restricted to the normal plane to \( \hat{z} \) as depicted in Fig. 2.

Limit \( D \ll -1 \). In this opposite limit, the local \(|0\rangle\) component is adiabatically suppressed and the two left spin components per site can be regarded as pseudospin-1/2, when mapping \( \{|\uparrow\rangle,|\downarrow\rangle\} \rightarrow \{|\uparrow\rangle,|\downarrow\rangle\} \). Strictly in the limit \( D = -\infty \), any configuration in this subspace has the same energy and there is a macroscopic degeneracy. For large but finite value of \(|D|\) the degeneracy is broken by the spin exchange terms. Taking into account that \( \mathcal{P}(\mathbf{S}_i \cdot \mathbf{S}_j)^2 \mathcal{P} = 2(\sigma_i^x \sigma_j^x + \sigma_i^y \sigma_j^y) \) and \( \mathcal{P} \mathbf{S}_i \cdot \mathbf{S}_j \mathcal{P} = \sigma_i \cdot \sigma_j \) (where \( \mathcal{P} \) is a projector to the subspace with no local component \(|0\rangle\)), the effective Hamiltonian in this limit corresponds to the spin-1/2 XXZ model:

\[
H_{XXZ} = \sum_{<i,j>} \left[ J_{\perp} (\sigma_i^x \sigma_j^x + \sigma_i^y \sigma_j^y) + J_z \sigma_i^z \sigma_j^z \right]
\]  

with parameters depending on \( \theta \) as \( 2J_{\perp} = \sin \theta \) and \( 2J_z = (2 \cos \theta - \sin \theta) \). It is easy to see that magnetization along the \( \hat{z} \)-direction in the \( S = 1/2 \) model directly maps into dipolar order along the same axis for \( S = 1 \), whereas the transverse magnetization maps into quadrupolar order for the latter. More specifically, the magnetization along \( \hat{x} \) and \( \hat{y} \)-axis in the spin-1/2 case map respectively to the components \( Q_1 \) and \( Q_3 \) of the quadrupolar vector. The spin-1/2 model on the triangular lattice has been addressed by several approaches (like QMC or variational methods) \(^6\) and predicts the presence of two distinct supersolid phases (which we denote by SS1 and SS2). These two phases are characterized by long range \( \sqrt{3} \times \sqrt{3} \) crystal order coexisting with superfluidity, which in spin language dubs to the same axis \( \sqrt{3} \times \sqrt{3} \) order in the spin \( z \)-component and non-vanishing magnetization in the perpendicular plane. This transverse magnetization differs in the two phases: for SS1 it takes the same vale at each sublattice \((m_{\perp}, m_{\perp}, m_{\perp})\), whereas for SS2 it corresponds to a configuration \((0, m_{\perp}, -m_{\perp})\). Intermediate values of \( D \). For \( D > 0 \), the phase diagram can be qualitatively well characterized by the Gutzwiller ansatz approach as shown in Fig. 2 where phase boundaries from Gutzwiller ansatz (dashed lines) and CMF approach (solid) are drawn. Such characterization notably fails in a region of \( D < 0 \), where the interplay between the different Hamiltonian terms becomes quite complex. The phases depicted in Fig. 2 can be summarized as follows:

**Region a:** Ferromagnetic (FM) order. — The FM phase persists for any \( D \leq 0 \), showing magnetization along the field direction, and a double degeneracy due to \( Z_2 \) symmetry. When crossing to the \( D > 0 \) side, the spins point instead along any perpendicular direction to the field. As \( D \) increases the magnetization continuously decreases until it vanishes and only FQ order remains. Close to the

**Region b:** Ferroquadrupolar (FQ) order. — As in region \( b \), there is no dipolar but quadrupolar local order. Now the vector directors have different orientation at each of the three sublattices, and their angle continuously varies with \( D \). In particular, for \( D < D_{c1} \), the directors are no longer parallel and start to form an umbrella configuration (region \( c1 \) in Fig. 2), until they are mutually orthogonal for \( D = 0 \). By further decreasing \( D \) the angle still increases until they all lie within the \( \hat{x} \hat{y} \)-plane at \( D = D_{c2} \). For \( D < D_{c2} \) (region \( c2 \)) they remain in this configuration, subtending a \( 120^\circ \) angle. Both critical fields \( (D_{c1} \) and \( D_{c2} \) depend on \( \theta \).

**Region c:** Antiferroquadrupolar (AFQ) order. — As in region \( b \), there is no dipolar but quadrupolar local order. Now the vector directors have different orientation at each of the three sublattices, and their angle continuously varies with \( D \). In particular, for \( D < D_{c1} \), the directors are no longer parallel and start to form an umbrella configuration (region \( c1 \) in Fig. 2), until they are mutually orthogonal for \( D = 0 \). By further decreasing \( D \) the angle still increases until they all lie within the \( \hat{x} \hat{y} \)-plane at \( D = D_{c2} \). For \( D < D_{c2} \) (region \( c2 \)) they remain in this configuration, subtending a \( 120^\circ \) angle. Both critical fields \( (D_{c1} \) and \( D_{c2} \) depend on \( \theta \).

**Region d:** Antiferromagnetic (AFM) order. — This region sustains different quantum phases. They are characterized by local dipolar order which is not ferromagnetic, but still show a three-sublattice structure, and a non-vanishing magnetization \( \langle S_+ \rangle \neq 0 \) at least at one of the
three sublattices. For $D > 0$ (region $d1$), the ground state is in a 120°-Néel ordered phase (similarly to the $D = 0$ case, but now with the spins contained in the $x\hat{y}$-plane). For $D < 0$, there are at least four different phases that are already observed within the mean-field approach. They are found consecutively when increasing $\theta$ from negative to positive values at fixed $D$ and characterized through the net magnetization along the parallel and perpendicular directions to the field axis ($\langle S^z_{\text{tot}} \rangle$ and $\langle S^\perp_{\text{tot}} \rangle$ respectively) as follows: $d2$) $\langle S^z_{\text{tot}} \rangle \neq 0$, $\langle S^\perp_{\text{tot}} \rangle = 0$; $d3$) $\langle S^z_{\text{tot}} \rangle \neq 0$, $\langle S^\perp_{\text{tot}} \rangle \neq 0$; $d4$) $\langle S^z_{\text{tot}} \rangle = 0$, $\langle S^\perp_{\text{tot}} \rangle \neq 0$; $d5$) the spins representing each sublattice are not contained in the same plane, leading to a non-zero value of the spin-chirality scalar defined as $\kappa = \langle S_1 \cdot (S_2 \times S_3) \rangle$.

Regions SS1 and SS2: Supersolid Phases. — By decreasing $D$ ($D \lesssim -3.5$) in region $d$, a new order emerges characterized by $\langle S^\perp \rangle = 0$ at any lattice site. In this region the Gutzwiller ansatz fails. Within this ansatz the local $|0\rangle$ component is completely suppressed, and thus, the model reduces to the spin-1/2 XXZ model, exactly as in the $D \to -\infty$ limit. The mean-field XXZ spin-1/2 model further maps into the classical XXZ model, which exhibits phases with qualitatively different local order and even non-trivial macroscopic degeneracies that are broken in the quantum model. In contrast, the CMF yields two phases with spin and quadrupolar order that might continuously connect with the mapped supersolid phases (SS1 and SS2) expected for the effective spin-1/2 XXZ model in the large easy-axis anisotropy limit ($D \ll -1$).

Despite the fact that in this region there is no transverse magnetization, we find that due to quantum correlations the local $|0\rangle$ component has not been totally suppressed, in stark contrast to the Gutzwiller case. Since we deal with non-bipartite lattices, the two SS phases cannot be mapped into each other ($|J_1 \rangle \leftrightarrow -|J_1 \rangle$). Thus, it is not surprising that the prolongation of these phases to finite values of $D$ is different. In particular, we find that for the SS1 phase the suppression of the $|0\rangle$ component is much more gradual than for the SS2 phase. Finally, in the region comprised between the two supersolid phases, the exact results obtained by CMF strongly depend on the geometry and size of the chosen clusters, indicating the importance of the role of quantum correlations in the stabilization of these phases. Indeed, we have recovered the SS2 phase only by using triangular clusters, while the convergence of the method when approaching the Ising point (the transition between SS1 and SS2 at $\theta = 0$) is strongly hindered for small clusters, for which three-sublattice order is broken. In Fig. 3 we depict the scaling of the phase boundary as a function of the connecting parameter $x = N_6/(L \cdot z/2)$ for different cluster sizes. Here $N_6$ are the number of quantum-mechanical bonds and sites of the cluster, while $z$ is the lattice coordination number. While transitions $FQ - AFM$ for $D = 0$ and $FQ - SS1$ at $D = -7$ accept a linear fit in $x$, the transition SS1–? clearly does not support such a fit.

FIG. 3: Scaling of different phase boundaries for different values of $x$ (see text) within CMF: (a) The $FQ - AFM$ boundary at $D = 0$, (first point corresponds to the Gutzwiller value for comparison), (b) $FQ - SS1$ phase at $D = -7$ (first point corresponds to the Gutzwiller value for comparison), (c) boundary between SS1–? phase at $D = -3.5$ (squares) and $D = -7$ (circles) showing lack of scaling.

Summarizing, we obtain the complete phase diagram of the spin-1 model with uniaxial anisotropy in a triangular lattice. Using the CMF we recover all the quantum phases already predicted by mean-field ansatz with corrected boundaries, plus phases in the region where the mean-field ansatz clearly fails. There, the CMF approach predicts two supersolid phases compatible with the effective spin-1/2 model expected in the limiting regime of large easy-axis anisotropy. Between them there is a phase whose nature we cannot determine —maybe due to the limited size of our clusters. Although we have not found any signature of a disordered phase or direct evidence of a spin liquid, the instability we observe in this region might be directly linked to an enhancement of quantum correlations. Thus, this approach bounds quite precisely (compared to previous results) the region more favorable for the stabilization of a spin liquid phase.

We acknowledge financial support from: MINECO FIS2008-01236 (Spain), Grant ID SGR2009-1289 (Catalonia), Grant ID 43467 (Templeton Foundation), Grant ID EP/L005026/1 (EPSRC), and Grant ID 618074 (EU Project TherMIQ). Fruitful discussions with A. Läuchli and M. Lewenstein are appreciated.

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