Field Induced Supersolid Phase in Spin-One Heisenberg Models

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We use quantum Monte Carlo methods to demonstrate that the quantum phase diagram of the S=1 Heisenberg model with uniaxial anisotropy contains an extended supersolid phase. We also show that this Hamiltonian is a particular case of a more general and ubiquitous model that describes the low energy spectrum of a class of isotropic and frustrated spin systems. This crucial result provides the required guidance for finding experimental realizations of a spin supersolid state.

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Theoretical proposals [1] for studying the Bose-Einstein condensation (BEC) with magnetic systems were followed by a vast number of experimental works [2]. These studies were done on spin dimer compounds for which the relevant $U(1)$ symmetry is “protected” by the intra-dimer singlet-triplet spin gap [3]. Magnetic systems have the advantage that the magnetic field, which plays role of the chemical potential, can be varied continuously over a large range of values. A natural question that arises is whether other phases that have been proposed for bosonic gases of atoms can be realized in quantum magnets. The supersolid (SS) state is a prominent and interesting example because the experimental evidence for this novel phase is still inconclusive [4].

The search for the SS phase has motivated the study of different models for hard-core bosons on frustrated lattices [5]. These models are relevant for gases of atoms in a periodic potential (substrate or optical lattice). However, the spin $S=1/2$ Hamiltonians that are obtained from these models by applying a Matsubara–Matsuda transformation [6] are not relevant for real magnetic systems. What makes these models unrealistic for magnetic systems is the large uniaxial exchange anisotropy. Moreover, the longitudinal and the transverse components of the exchange interaction have opposite signs: while the Ising interaction is antiferromagnetic (AFM), the transverse exchange coupling is ferromagnetic. It is then natural and relevant to ask if a SS spin phase can exist in a magnetic system with isotropic (Heisenberg) interactions. In this letter, we provide an affirmative answer to this question by calculating the quantum phase diagram of an $S=1$ spin–dimer Heisenberg model. The spin SS phase is induced by the application of a magnetic field whose Zeeman splitting is comparable to the magnitude of the exchange interactions.

To understand the physical origin of the spin SS, we shall start by considering the simplest (although non-realistic) $S=1$ Hamiltonian that contains this phase in its phase diagram. This is an $S=1$–Heisenberg model with uniaxial single–ion and exchange anisotropies on a square lattice:

$$H_H = J \sum_{\langle i,j \rangle} (S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z) + \sum_i (D S_i^{\perp 2} - B S_i^z )$$

where $\langle i,j \rangle$ indicates that $i$ and $j$ are nearest neighbor sites, $D$ is the amplitude of the single ion-anisotropy and $\Delta$ determines the magnitude of the exchange uniaxial anisotropy. Note that although the exchange interaction is anisotropic, the longitudinal ($J$) and transverse ($\Delta$) couplings are both AFM (positive). Henceforth, $J$ is set to unity and all the parameters are expressed in units of $J$.

The quantum phase diagrams for the spin models considered in this paper were obtained by using the Stochastic Series expansion (SSE) quantum Monte Carlo (QMC) method. The simulations were carried out on a square lattice of size $N = L \times L$, with $8 \leq L \leq 16$. We find rapid convergence with $N$ for the system sizes studied (see Fig. 1). Since the SSE is formulated in the grand canonical ensemble, the simulations are performed at fixed magnetic field, instead of fixed magnetization.

As the external field, $B$, is varied, the ground state of $H_H$ goes through a number of phases, including spin-gapped (IS) phases with Ising-like ordering and gapless (XY) phases with dominant XY-ordering. The IS phases are characterized by long-range (staggered) diagonal order measured by the longitudinal component of the static structure factor (SSF),

$$S^{zz}(q) = \frac{1}{N} \sum_{j,k} e^{-i q \cdot (r_j - r_k)} \langle S_{j}^{z} S_{k}^{z} \rangle. \quad (2)$$

The XY–phase has long-range off-diagonal ordering measured by the transverse component of the SSF,

$$S^{+-}(q) = \frac{1}{N} \sum_{j,k} e^{-i q \cdot (r_j - r_k)} \langle S_{j}^{+} S_{k}^{-} \rangle. \quad (3)$$

The XY–ordering is equivalent to a Bose–Einstein condensation (BEC) whose condensate fraction is equal to $S^{+-}(Q)$ with $Q$ being the ordering wave–vector ($Q = (\pi, \pi)$ for the case under consideration). The superfluid density corresponds to the spin stiffness, $\rho_s$, defined as the response of the system to a twist in the boundary conditions. The stiffness is obtained from the winding numbers of the world lines ($W_x$ and $W_y$) in the $x$– and $y$– directions: $\rho_s = (W_x^2 + W_y^2)/2\beta$.

The IS (XY) phase is marked by a diverging value of $S^{+-}(Q) \propto N$ ($S^{+-}(Q) \propto N$) in the thermodynamic limit $N \to \infty$. In addition, $\rho_s$ vanishes in the gapped IS phase while it is finite in the gapless XY phase. A spin SS phase...
is characterized by a finite value of both \( S^z(Q)/N \) and \( \rho_s \). Both quantities are always finite for finite size systems and estimates for \( N \to \infty \) are obtained from finite-size scaling.

Fig. 1 shows the quantum phase diagram as a function of magnetic field, \( B \), for \( D = 1.5 \) and \( \Delta = 1.8 \). For clarity of presentation, \( S^z(Q) \) and \( \rho_s \) are plotted as a function of the resulting magnetization \( m_z \). The \( m_z(B) \) curve features two prominent plateaus corresponding to different IS phases. For small \( B \), the ground state is a gapped AFM solid (IS1) with no net magnetization. The stiffness, \( \rho_s \), and \( S^z(Q) \) vanish in the thermodynamic limit, while \( S^z(Q)/N \) is slightly smaller than 1 because the spins are mainly in the \( S_z = \pm 1 \) states depending on which sublattice they belong to. The magnetization stays zero up to the critical field, \( B_{c1} \), that marks a second order BEC quantum phase transition (QPT) to a state with a finite fraction of spins in the \( S_z = 0 \) state. This state has a finite \( S^z(Q)/N \) as well as finite \( \rho_s \) and \( S^z(Q)/N \), i.e., SS order. The diagonal order results from the \( S_z^2 \) sublattices while the off-diagonal order arises out of a BEC of the flipped spins (\( S^z = 0 \) “particles”). The magnetization increases continuously up to \( B \approx 6.4 \), where there is a second BEC–QPT to a second Ising-like state (IS2) where all the \( S_z^2 = -1 \) have been flipped to the \( S_z^2 = 0 \) state. \( S^z(Q)/N \) remains divergent for \( N \to \infty \), but the stiffness, \( \rho_s \), and condensate fraction, \( S^z(Q) \), drop to zero. The ground state remains in the IS2 phase for \( 6.4 < B < 7.2 \). Upon further increasing the field, there is a first order transition to a pure XY–AFM phase (\( m_z \) changes discontinuously from \( m_z = 0.5 \) to \( m_z \approx 0.59 \)). In the grand canonical ensemble, no ground state with any intermediate value of the magnetization is realized. For a canonical ensemble with a fixed magnetization \(-0.6 < m_z < -0.5\), the ground state will phase separate into IS2 and XY regions with \( m_z = 0.5 \) and \( m_z = 0.59 \). In the pure XY phase, the diagonal order vanishes while \( \rho_s \) and \( S^z(Q)/N \) remain finite. This situation persists until all the spins have flipped to the \( S_z^2 = 1 \) (fully polarized) state.

Further insight into the SS phase is obtained from the momentum dependence of \( S^z(q) \) (Fig. 1c). The peaks at \( q = (0, 0) \) and \( q = Q \) indicate that the off-diagonal long range order is modulated by the presence of solid order. This confirms that the SF component of the SS phase results from a BEC of \( S_z^2 = 0 \) spin states that occupy the \( S^z = -1 \) or down–sublattice with higher probability. This feature distinguishes the SS phase from a uniform canted AFM phase. In experiments with magnetic materials, the components of the structure factor are selectively measured using standard polarized neutron scattering experiments.

For smaller values of \( \Delta (< D) \), the second magnetization plateau disappears completely (Fig. 2) leaving a second order transition from the SS to the XY–phase. Consequently, there is no phase separation regime. The extent of the SS phase decreases with decreasing \( \Delta \) and vanishes for \( \Delta \approx 1 \).

We shall now discuss the relevance of these results for finding a SS phase in real magnets. We note that although a \( U(1) \) invariant model provides a good description of spin compounds whose anisotropy terms are very small compared to the Heisenberg interactions, this invariance is never perfect. The transition metal magnetic ions belong to this class because the spin-orbit interaction is much smaller than the crystal field splitting. These spin systems have small exchange anisotropies for the same reason. Therefore, models that assume opposite signs for \( J_z \) and \( J_\perp \) or large values of \( J_\perp/J_z \) are not applicable to these spin compounds. We will show below that it is not necessary to assume a strong uniaxial exchange anisotropy for obtaining a SS phase.

The system to be considered is a square lattice of \( S = 1 \) dimers (Fig. 3) which only includes isotropic (Heisenberg) AFM interactions, an intra-dimer exchange \( J_0 \) and inter-dimer interactions...
frustrated couplings $J_1$ and $J_2$:}

$$H_D = J_0 \sum_i S_{i+} \cdot S_{i-} + J_1 \sum_{\langle i,j \rangle, \alpha} S_{i\alpha} \cdot S_{j\alpha}$$

$$+ J_2 \sum_{\langle i,j \rangle, \alpha} S_{i\alpha} \cdot S_{j\bar{\alpha}} - B \sum_{i\alpha} S_{i\alpha}^z. \quad (4)$$

The index $\alpha = \pm$ denotes the two spins on each dimer. The single dimer spectrum consists of a singlet, a triplet and a quintuplet (see Fig. 3). The energy difference between the singlet and the triplet is $J_0$, while the difference between the quintuplet and the triplet is $2J_0$.

For $J_1, J_2 \ll J_0$, the low energy subspace of $H_D$ consists of the singlet, the $S^z = 1$ triplet and the $S^z = 2$ quintuplet (see Fig. 3). The low energy effective model, $H$, that results from restricting $H_D$ to this subspace is conveniently expressed in terms of semi-hard-core bosonic operators, $g_i^+\text{ and } g_i\text{ that satisfy the exclusion condition } g_i^{\dagger 3} = 0$ (no more than two per site) $[\bar{8},[10]]$ and obey the commutation relations of canonical bosons except for the commutator $[g_i^+, g_j] = \delta_{ij}(1-n_i)$ ($n_i = g_i^+ g_i\text{ is the number operator}$). The expression of $H$ in terms of these operators is:

$$H = \frac{1}{2} \sum_{\langle i,j \rangle} (g_i^+ g_j g_i + g_j^+ g_i g_j) (h_1 + h_2 + h_3) - \mu \sum_i n_i$$

$$+ \frac{U}{2} \sum_i n_i(n_i - 1) + V \sum_{\langle i,j \rangle} (n_i - 1)(n_j - 1). \quad (5)$$

with $h_1 = t_1(n_{ij} - 2)(n_{ij} - 3)$, $h_2 = 2t_2(n_{ij} - 1)(3 - n_{ij})$, $h_3 = t_3(n_{ij} - 1)(n_{ij} - 2)$, and $n_{ij} = n_i + n_j$. The amplitudes $t_1, t_2, t_3$ correspond to single-particle hopping terms when there are one, two or three particles respectively on the corresponding bond $\langle i,j \rangle$. The case $t_1 = t_2 = t_3 = t$ corresponds to the bosonic Hubbard model with n.n. repulsion $[7]$ in a truncated Hilbert space. Our $S = 1$ Heisenberg Hamiltonian with uniaxial anisotropy, $H_H$, is obtained for $U = D$, $V = \Delta J$, $\mu = D + B$ and $t_j = \sqrt{2}J/2^{j/2}$ with $j = 1, 2, 3$ after we map on each site the eigenstates of $S_i^z$ onto the eigenstates of $n_i$: $S_i^z = n_i - 1$ and $S_i^+ = g_i^{\dagger} [\sqrt{2} + (1 - \sqrt{2})n_i]$. The second magnetization plateau disappears completely. Instead, there is a direct (continuous) SS-XY transition at high fields, there is a transition to a fully polarized state (PL).

As we mentioned before, $H$ also describes the low energy spectrum of $H_D$. In this case, we have $U = J_0$, $V = (J_1 + J_2)/2$, $\mu = B - J_0 - z(J_1 + J_2)/2$ and $t_j = 8a^2(J_1 - J_2)/3\sqrt{3}$ with $a = \sqrt{3}/2$, after mapping the eigenstates of $S_i^z$ into the eigenstates of $n_i$ by the simple relations: $n_i = S_i^z + S_i^z$ (see Fig 3) and $g_i^{\dagger} = (S_i^+ - S_i^-)[\sqrt{2}/2 + (1 - \sqrt{2})S_i^z]$. Fig. 3 shows the quantum phase diagram as a function of $\mu$ (or $B$) for $U = 30.0$ and $V = 7.0$ (in this case we take $(J_1 - J_2)/2$ as the unit of energy). This set of parameters corresponds to $J_0 = 30J_1 = 8$ and $J_2 = 6$ that satisfies the conditions $J_0 > z(J_1 + J_2)/2$ and $J_0 > z(J_1 - J_2)/2$ necessary for the validity of $H$ as a low energy effective model for $H_D$.

At small $\mu$ or $B$, the empty state (all the dimers in a singlet state) has the lowest energy. For $\mu > \mu_c3 (B > B_{c3})$ a finite density of bosons (triplets) is stabilized in the ground state giving rise to a BEC (XY–AFM ordering) at $T = 0$ with a finite the stiffness $\rho_s$. The absence of solid (Ising) order is indicated by $S^z(Q)/N \to 0$. The density (magnetization) increases monotonically as a function of $\mu$ or $B$ until $\mu = \mu_c2 \approx 2.9$ where there is a discontinuous transition to a charge-density wave (CDW) or Ising-like phase with $n = m_z = 0.5$ (the dimers of one sublattice are in a triplet state while the other dimers remain in the singlet state). For $\mu > \mu_c3 \approx 23.4$, some of the dimers of the singlet sublattice are turned into triplets that propagate primarily on the singlet sublattice ($U \gtrsim zV$ where $z = 4$ is the coordination number). Consequently, there is a BEC–QPT (broken $U(1)$ symmetry under rotations around the $z$-axis) in $D = d + 2$ dimensions to a SS phase, where $d$ is the spatial dimensionality. The diagonal or solid order disappears at an Ising–like quantum critical point in $D = d + 1$ dimensions for $\mu = \mu_c2 \approx 25.4$ (broken $Z_2$ symmetry of translation by one lattice parameter followed by a $\pi$–rotation around the $z$-axis). Upon further increase in $\mu$, the filling increases monotonically in the resulting SF phase till the ground state enters a Mott insulating (MI) phase with
The energy cost to place a boson at an empty (occupied) site is $\mu_0 = zV - \mu$ ($E_1 = U - \mu$). Respectively, for $U \gg zV$, the additional bosons fill empty sites and mask the checkerboard modulation; for $U - zV \gg |t|$, the situation is precisely particle-hole conjugate to hole doping. In particular, in the hard-core limit $U \to \infty$, the crystalline order is always unstable for $n \neq 1/2$. However, for $zV \sim U$, the bosons can be placed on either an occupied or unoccupied site. The kinetic energy gain of the added boson is now linear in $t$ because the potential barrier, $|zV - U|$, for moving the bosons to nearest neighbors is not much bigger than $t$. As a result, the added bosons form a SF phase on top of the density wave background and hence the ground state has simultaneous solid and SF orders. This SS phase is stable for a sufficiently small concentration of added bosons. This is confirmed by the quantum phase diagram shown in Fig. 4 where the SS phase appears right next to the $n = 1/2$ CDW. We emphasize that this phase requires to have two bosons on the same site, which is not possible for hard-core bosons (or, equivalently, for $S = \frac{1}{2}$ spins).

In summary, we have shown that simple two-dimensional $S = 1$ Heisenberg models have a spin SS ground state induced by magnetic field. The physical mechanism that leads to this phase does not depend on the dimensionality and similar results are expected for three- and one-dimensional lattices \[11 \quad 12\]. In particular, by finding a SS phase in an isotropic $S=1$ Heisenberg model we are providing the required guidance for finding this novel phase in real spin systems. The crucial ingredients for the described mechanism are: $S = 1$ spins, a dimerized structure and a frustrated inter–dimer coupling.

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