Frustrated antiferromagnets at high fields: the Bose-Einstein condensation in degenerate spectra

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Quantum phase transition at the saturation field is studied for a class of frustrated quantum antiferromagnets. The considered models include (i) the $J_1$-$J_2$ frustrated square-lattice antiferromagnet with $J_2 = \frac{1}{2}J_1$ and (ii) the nearest-neighbor Heisenberg antiferromagnet on a face centered cubic lattice. In the fully saturated phase the magnon spectra for the two models have lines of degenerate minima. Transition into partially magnetized state is treated via a mapping to a dilute gas of hard core bosons and by complementary spin-wave calculations. Momentum dependence of the exact four-point boson vertex removes the degeneracy of the single-particle excitation spectra and selects the ordering wave-vectors at $(\pi, \pi)$ and $(\pi, 0, 0)$ for the two models. The asymptotic behavior of the magnetization curve differs significantly from that of conventional antiferromagnet in $d$-spatial dimensions. We predict a unique form for the magnetization curve $\Delta M = S - M \simeq \mu^{(d-1)/2}(\log \mu)^{(d-1)}$, where $\mu$ is a distance from the quantum critical point.

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Heisenberg antiferromagnets exhibit a quantum phase transition between a fully polarized state and a state with partial magnetization at the saturation field $H_c$. A partially magnetized state typically breaks spin-rotational symmetry about the field direction and has a long-range order of transverse spin components. Such a zero temperature transition belongs to the $XY$ universality class. Various properties of quantum antiferromagnets in the vicinity of this quantum critical point are well understood in terms of the Bose condensation of magnons below $H_c$\textsuperscript{1,2}. The antiferromagnetic wave-vector corresponds to the minimum in the magnon spectrum, whereas an arbitrary phase of the condensate describes sublattice orientation in the plane perpendicular to the field.

The above simple picture fails, however, for so-called frustrated antiferromagnets, which have degenerate classical ground states between zero and saturation fields\textsuperscript{3,4}. The excitation spectra of frustrated antiferromagnets above $H_c$ are quite unusual. The magnon dispersion $\epsilon_q$ has a continuous set of energy minima given by a $d'$-dimensional hypersurface embedded in the $d$-dimensional reciprocal space. The problem of the Bose condensation of particles with such degenerate spectra remains unexploited to a large extent. Numerical investigations\textsuperscript{5,6} of a specific model (model A below) show a strong singularity of the magnetization near the saturation field, which has not been interpreted so far. In the present work we investigate two problems: (i) how the quantum fluctuations remove degeneracy of the excitation spectra and select a certain ordering wave-vector and (ii) how the asymptotic behavior of the magnetization curve close to the quantum critical point is modified by a presence of large phase space of soft-mode fluctuations. Specifically, we consider two models with $d' = 1$ in $d = 2$ and $d = 3$ spatial dimensions: (model A) the $J_1$-$J_2$ antiferromagnetic Heisenberg model on a square lattice at its critical point $J_2 = \frac{1}{2}J_1$, which has lines of minima at $(\pi, q)$ and $(q, \pi)$ and (model B) the nearest-neighbor Heisenberg model on a face centered cubic lattice (fcc) with minima at $(\pi, q, 0)$ and equivalent lines.

Convenient approach to deal with quantum antiferromagnets near the saturation field is to employ a hard-core boson representation of spin-1/2 operators: $S_i^z = \frac{1}{2} - b_i^\dagger b_i$, $S_i^+ = b_i$, and $S_i^- = b_i^\dagger$ with $b_i^\dagger b_i = 0$ or 1\textsuperscript{7}. The hard-core constraint is imposed by an on-site repulsion $U \to \infty$. The Heisenberg spin Hamiltonian $\hat{H} = \sum_{\langle i,j \rangle} J_{ij} S_i S_j - H \sum_i S_i^z$ is, then, transformed to

$$\hat{H} = \sum_q \left( \epsilon_q - \mu \right) b_q^\dagger b_q + \frac{1}{2N} \sum_{q,k,k'} V_{pq} b_k^\dagger b_k b_{q-k}^\dagger b_{q-k},$$ (1)

where $\epsilon_q = \frac{1}{2}(\gamma_q - \gamma_{\min})$, $\gamma_q = \sum_{i,j} J_{ij} e^{iqr_{ij}}$ is the Fourier transform of the exchange interaction $J_{ij}$, $\mu = H_c - H$ is a boson chemical potential, and $H_c = \frac{1}{2}(\gamma_0 - \gamma_{\min})$ is the saturation field. A bare four-point boson vertex is given by $V_q = U + \gamma_q$. The exact ground state of the Hamiltonian\textsuperscript{8} is the boson vacuum for $\mu < 0$ ($H > H_c$), it corresponds to a ferromagnetic alignment of spins. In $d \geq 2$ the ground state with finite boson density $\langle b_i^\dagger b_i \rangle \neq 0$ at $\mu > 0$ should be, generally, a superfluid state: $\langle b_q \rangle \neq 0$ for a particular wave-vector $q = Q$, such that $\epsilon_Q - \mu$ vanishes at the saturation field.

In the above two models the bare spectrum $\epsilon_q$ has lines of degenerate minima (see below) and the condensate wave-vector remains undetermined. The degeneracy cannot be lifted by the bare interaction $V_q$ as it depends on momentum transfer only. This is a manifestation of an infinite classical degeneracy of the ground state of the original spin problem below the saturation field. Such an ‘accidental’ degeneracy can be removed by quantum
fluctuations \[8\]. The number of bosons vanishes at the quantum critical point and hence the system becomes dilute in the limit \(\mu \to 0^+\). In this case the leading order corrections to the scattering vertex are generated by the processes in the particle-particle channel. According to the standard procedure the bare interaction is, then, replaced by a solution of the integral equation for the scattering amplitude \(\Gamma\). We show that \(\Gamma\) does depend on the incoming momenta and thus lifts the degeneracy of the original problem. The ordering wave-vector corresponds to the smallest \(\Gamma\), i.e., condensate occurs at the momentum at which bosons less interact with each other in order to minimize their repulsion.

The Bethe-Salpeter equation for the scattering function with zero total frequency reads as

\[
\Gamma_q(k,k') = V_q - \frac{1}{N} \sum_p V_{q-p} \frac{\Gamma_p(k,k')}{\epsilon_{k+p} + \epsilon_{k'-p}} \tag{2}
\]

and is graphically represented in Fig. 1. In the limit \(U \to \infty\), Eq. (2) is reduced to a system

\[
\frac{1}{N} \sum_p \frac{\Gamma_p(k,k')}{\epsilon_{k+p} + \epsilon_{k'-p}} = 1 , \tag{3}
\]

where \(\langle \Gamma \rangle = (1/N) \sum_q \Gamma_q(k,k')\) and the identity \(\langle \gamma \rangle = J_{ii} = 0\) has been used. By expanding \(\Gamma_q(k,k')\) in lattice harmonics of the wave-vector \(q\), the integral equations are transformed to a system of algebraic equations. Since, only a few harmonics appear in such an expansion of \(\Gamma_q(k,k')\), essentially those which are present in \(\gamma_q\), the resulting algebraic system can be easily solved analytically. We shall now describe further details for the two models separately.

\[
\begin{array}{c}
\text{FIG. 1: Graphical representation of the integral equation for four-point vertex.}
\end{array}
\]

Model A: Frustrated antiferromagnet on a square lattice with the nearest-neighbor exchange constant \(J_1 = 1\) and the diagonal coupling of strength \(J_2 = \frac{1}{2} J_1\) has infinitely many classical ground states for \(0 < H < H_c\). The magnon spectrum in the ferromagnetic phase at \(H = H_c\) is \(\epsilon_q = (1 + \cos q_x)(1 + \cos q_y)\). The magnon energy has lines of minima spanned by wave-vectors \(\{q^*\}\): \((\pi, q)\) and \((q, \pi)\). Besides the well-known singularity related to vanishing of a scattering amplitude for two quantum particles in two dimensions \((2d)\), the present 1d type degeneracy leads to an extra infrared divergence in the kernel of the integral equation when the external momenta are fixed to \(k, k' = q^*\). Away from the critical point \(\mu > 0\) the singularity can be cured by introducing an infrared cut-off for single-particle energies defined by \(\epsilon_{k,k'} \geq \mu\). The physics behind this regularization procedure is as follows: at the energy scale of the order of \(\mu\) the interaction effects become important and modify the form of the bare spectrum. As it follows, an infrared behavior of the excitation spectrum is sound-like and, hence, removes in a self-consistent way the divergence of the kernel below \(H_c\).

The condensate wave-vector is chosen by considering the scattering amplitude at zero momentum transfer and external momenta set to \(q^*\). The two main candidate states are the Néel state with \(Q = (\pi, \pi)\), which is stable for a frustrated square lattice antiferromagnet with weaker diagonal bonds \(J_2 < \frac{1}{2} J_1\), and the columnar state ordered on \((\pi, 0) \{0, \pi\}\), which is stable for stronger diagonal bonds \(J_2 > \frac{1}{2} J_1\). Instead of presenting a rather lengthy solution of the algebraic system we obtain an approximate but physically transparent solution by neglecting the momentum-transfer dependence of \(\Gamma_p(q^*, q^*) \propto \Gamma_p(q^*)\) near \(p = 0\) under the integral in equation (3). The result is \(\Gamma_0(q^*) \approx 1/\tau(q^*)\) with \(\tau(q^*) = 1/N \sum_p [\epsilon_p - \epsilon_{q^* + p}] \). It is evident that \(\Gamma_0(q^*)\) is smallest for a \(q^*\) at which the kernel \(\tau(q^*)\) has the strongest divergence. This singles out the Néel wave-vector \(Q = (\pi, \pi)\), which has the softest excitations \(\epsilon_{Q-k^*} \approx k_x^2 k_y^2\). Hence, for \(\mu > 0\) magnons condense at \(q^* = Q\) and a transverse antiferromagnetic order should be formed below the saturation field. Direct estimation of the integral in the kernel \(\tau(q^*)\) yields \(\Gamma_0(Q) \sim \mu^{1/2}/|\log \mu|\), whereas away from ordering wave-vector \(\Gamma_0(q^*) \sim \mu^{1/2}\). The square root behavior of the four-point vertex is intrinsic to 1d systems. This is a consequence of the quasi 1d low-energy part of the bare spectrum of the frustrated model. In addition, there is an extra logarithmic correction to the 1d behavior at the ordering wave-vector \(Q\) related to vanishing stiffness in both directions leading to a Van Hove type singularity in the kernel.

In the ordered state below \(H_c\), the condensate density \(n_0 = \langle b_{q^*}^+ b_{q^*} \rangle\) is found from minimization of the ground state energy density \(e_{g.s.} = -\mu n_0 + \frac{1}{\hbar^2} \Gamma_0(Q) n_0^2\), where we have neglected the noncondensate contribution. The excitation spectrum can be obtained following the standard Bogoliubov scheme and replacing the bare vertex \(V_q\) with the full scattering amplitude:

\[
\begin{align}
\omega_q^2 &= \left(\epsilon_q - \mu + \Sigma_{q}^{11}\right)^2 - \left(\Sigma_{q}^{12}\right)^2, \\
\Sigma_{q}^{11} &= n_0 [\Gamma_0(q, Q) + \Gamma_{q-Q}(q, Q)], \quad \Sigma_{q}^{12} = n_0 \Gamma_{q-Q}(Q). \tag{4}
\end{align}
\]

As it is seen from Eq. (4) the self-energy acquires an additional momentum dependence thanks to the dependence of \(\Gamma\) on the incoming momenta. The magnon spectrum is no longer degenerate and has the unique zero-
energy mode at $q = Q$. For $q \rightarrow Q$ the magnon energy becomes $\omega_q \approx \sqrt{\mu q_0} [1 + \log(q_0)] - \mu$. Expanding the vertex in small $|k| = |q - Q|$, we obtain an acoustic mode $\omega_k \approx sk$ with the velocity $s^2 \approx \mu/|\log \mu|$, which is smaller than the velocity $s^2 \approx \mu$ in nonfrustrated 2d and 3d antiferromagnets near the saturation. Away from the ordering wave-vector, along the degeneracy lines, magnons acquire a dynamically generated gap $\Delta \sim |\mu|/|\log \mu|$. The magnetization of the spin system is related to the total density of particles $M(H) = \frac{1}{2} - (n_0 + n')$, which includes both the condensate $n_0$ and noncondensate $n'$ parts. The noncondensate density of bosons is given by $n' = 1/N \sum_q \sum_{q_1} \{\omega_q - \mu + \sum_{q_1} \omega_{q_1}/[2\omega_q]\}$. The largest contribution to $n'$ is determined by wave-vectors away from $Q$, along the degeneracy lines $q \in \{q^*\}$. The ratio of the two densities is estimated as $n'/n_0 \approx 1/|\log \mu|^{1/2}$ and is logarithmically small, though it exceeds the condensate depletion found for a nondegenerate 2d Bose gas $(n'/n_0 \sim 1/|\log \mu|)$ [12]. Near $H_c$, the magnetization curve exhibits a strong singularity

$$M(H) \sim |\mu|^{1/2} |\log \mu| \sim \sqrt{H - H_c} \log (H_c - H),$$

which fits well to the available numerical data [2] [3].

The above results obtained in the hard-core boson picture are valid in the vicinity of $H_c$ and should be compared with the standard linear spin-wave theory (LSWT), which applies for all fields $0 < H < H_c$. In the LSWT approach one selects a few classical ground states, which applies for all fields $0 < H < H_c$. Another candidate state for $H_c < H < H_c$ is a four-sublattice state, which has identical polarization in three sublattices with the fourth sublattice compensating the net transverse magnetization. Such a partially collinear state is a natural favorite if quantum effects are taken into account via effective biquadratic exchange interaction derived in a second-order real-space perturbation theory [13]. LSWT shows instead that the Néel state has again a lower energy for a frustrated square-lattice antiferromagnet. This discrepancy is explained by a nonanalytic dependence of the ground state energy on applied magnetic field determined by a large number of soft (zero) modes, whereas the real-space perturbation approach captures only analytic contributions. Furthermore, the Néel antiferromagnetic order is stable even at $H = H_c$, where it has a lower energy of zero-point oscillations $E_0 = 0.694S$ than a fully collinear up-up-down-down (uudd) state with $E_0 = 0.703S$. This result corrects the previous spin-wave calculation [11] and is in agreement with the exact diagonalization results [2] [11], which show that the 1/2-magnetization plateau appears in the present model for $0.5 < J_2/J_1 \lesssim 0.65$. In the vicinity of $H_c$, the LSWT magnetization curve for the Néel state shows the same type of singularity with $M = Sh\{1 + \sqrt{2(1 - h)} \ln[128(1 - h)/\pi^4e^2]/\pi^2S\}$, where $h = H/H_c$.

**Model B:** A nearest-neighbor antiferromagnet ($J = 1$) on an fcc lattice is another frustrated model with degenerate ground states at $0 < H < H_c$. The spectrum of magnon excitation at the saturation field is $\epsilon_q = 2[1 + \cos q_x \cos q_y + \cos q_x \cos q_z + \cos q_y \cos q_z]$ and has lines of zeros at $(\pi, q, 0)$ and the cubic symmetry related lines. We then follow the same scheme as for the previous model and examine the kernel for the scattering amplitude with the new spectrum. The lowest value of renormalized four-point boson vertex corresponds to the three wave-vectors $Q_i = (\pi, 0, 0), (0, \pi, 0)$, and $(0, 0, \pi)$. The corresponding vertex is estimated as $\Gamma = \Gamma_0(Q_i, Q_i) \sim 1/|\log \mu|^2$. It has a 2d logarithmic behavior due to a quasi-2d form of the spectrum in the vicinity of $(\pi, q, 0)$ line with an extra logarithmic singularity related again to a vanishing stiffness for $k = 0$: $\epsilon_{Q_x + k} \sim k_x^2 + k_y^2 + k_z^2$. Due to a presence of three equally singular wave-vectors one must check now whether the ground state of the systems is characterized by a Bose condensation at a single $Q_i$ wave-vector or at all three wave-vectors simultaneously [14]. For this we write the ground state energy in the Landau form:

$$E = -\mu \sum_i |\psi_i|^2 + \frac{\Gamma}{2} \sum_{i \neq j} |\psi_i|^4$$

$$+ \sum_{i \neq j} \left[\bar{\Gamma} |\psi_i|^2 |\psi_j|^2 + \frac{1}{2} \bar{\Gamma} (|\psi_i|^2 |\psi_j|^2 + |\psi_j|^2 |\psi_i|^2)\right],$$

where $\psi_i = (b_{Q_i})$ is a complex order parameter, $\bar{\Gamma} = \Gamma_0(Q_i, Q_j) + \Gamma_{Q_i - Q_j}(Q_i, Q_j)$, and $\bar{\Gamma} = \Gamma_{Q_i - Q_j}(Q_i, Q_j)$. A single component phase is stabilized for a sufficiently strong repulsion between components $\Gamma < \Gamma - \bar{\Gamma}$, whereas in the opposite case all three components of the Bose condensate appear with equal weights. Direct calculations show that $\bar{\Gamma} \approx 1/|\log \mu|$ and $\bar{\Gamma} \approx 1/|\log \mu|^2$. Hence, in the vicinity of the saturation field dominated by the logarithmic behavior $\bar{\Gamma} \gg \Gamma$ and the single $k$ state is energetically favorable. The Bose density and asymptotic behavior of the magnetization curve is given by $n = 1/2-M(H) \sim 1/|\log \mu|^2$. We again find a $d-1$ like form for the magnetization with a logarithmic correction. Corrections beyond the leading logarithms can, however, change the energy balance. From explicit estimate of the prefactors, one finds that $\bar{\Gamma} \geq \Gamma$ only in an extremely narrow interval near the saturation field for $\mu \lesssim e^{-20}$. Beyond this interval $\bar{\Gamma} < \Gamma$ and a multi-$k$ state with a real $\bar{\Gamma} < 0$ superposition of all three modes with equal amplitudes is stabilized as the ground state. Such a spin structure corresponds to a partially collinear spin configuration described above in our discussion of the model A. The LSWT calculations confirm the above conclusion showing that for magnetic fields as close to the saturation field as $\Delta H/H_c = 10^{-3}$
the partially collinear state is energetically more favorable than a single-$k$ state. In contrast to the behavior of a frustrated square-lattice antiferromagnet we have also found an $uuud$ configuration for the ground state at $H = \frac{1}{2}H_c$. This state has the zero-point oscillation energy of $E_0 = 1.66S$, while a single-$k$ state has $E_0 = 1.74S$. Thus, a $1/2$-magnetization plateau should appear on the magnetization curve of an fcc antiferromagnet.

Finally, we make a few remarks on finite temperature behavior. The models $A$ and $B$ are magnetic analogs of the weak-crystallization model [15]. In this model the phonon spectrum softens for wave-vectors lying on a sphere. The phase transition to a crystal state appears to be of the first order even if a mean-field theory would predict a continuous transition. Such a fluctuation driven first-order transition is explained by a singular Hartree correction generated by thermal fluctuations, which have a large phase space [15].

For a 3d quantum fcc antiferromagnet a finite temperature transition to the ordered state is expected for all $H < H_c$. In zero magnetic field neglecting quantum fluctuations the Hartree correction is estimated as $\Sigma_H \sim \sum_q T/|q_0 + q| \sim CT |\log \tau|^2$, where $\tau = (T - T_{MF})/T_{MF}$ is a distance from a mean-field critical point $T_{MF}$. The self-consistent equation for the excitation gap becomes $\Delta = \tau + CT |\log \Delta|^2$. The point of absolute instability $\Delta = 0$ cannot be reached at any finite $T$ indicating a first-order transition. For classical Heisenberg and $XY$ model on an fcc lattice the first-order transition at $H = 0$ has been confirmed by the Monte Carlo simulations [16].

The transition between paramagnetic and antiferromagnetic states is, therefore, of the first order at low $H$ and high $T$. The zero temperature phase transition at $H_c$ is instead of the second order. This opens two possible scenarios for the $H-T$ phase diagram (a) a finite temperature tricritical point separates low-field (high-$T$) first-order transition line from the $XY$ transition at high-fields; or (b) fluctuation induced first-order transition survives down to zero temperature and terminates at the quantum critical point. For the two-dimensional model $A$, similar questions can be raised on the interplay between a Berezinski-Kosterlitz-Thoules (BKT) transition and a fluctuation induced first-order transition. At high temperatures (low fields) high degree of degeneracy of the spectra may induce the first-order transition and make the BKT instability inaccessible. Such a scenario is, for example, realized in the theory of weak crystallization of films [17]. While at low temperatures (high fields) a usual BKT transition may take place. These interesting issues require further investigations.

Summary. We have studied a quantum phase transition at high magnetic fields for a class of frustrated antiferromagnets in $d = 2$ and $d = 3$, which have degenerate excitation spectra with lines of minima. Momentum dependence of the exact four-point boson vertex removes the degeneracy and selects the ordering wave-vector $Q$. The new spectra are sound-like near $Q$ and acquire dynamically generated gaps away from it. The asymptotic behavior of the magnetization curve shows the same singularity as a nonfrustrated model in $d - 1$ dimension with an additional logarithmic correction. The developed scheme can be applied to other frustrated antiferromagnets near the saturation and to singlet ground state systems with degenerate gapped triplet excitations, as, e.g., SrCu$_2$(BO$_3$)$_2$ [18] or Cs$_3$Cr$_2$Br$_9$ [19], near the triplet condensation field $H_{c1}$. In addition, recent experimental progress on the Bose condensation of alkali atoms on optical lattices [20] opens a new possibility for experimental study of condensate phenomena on frustrated lattices, the systems to which our results also apply.

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