Magnetization plateaus as insulator-superfluid transitions in quantum spin systems

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We study the magnetization process in two-dimensional $S = 1/2$ spin systems, to discuss the appearance of a plateau structure. The following three cases are considered: 1) the Heisenberg antiferromagnet and multiple-spin exchange model on the triangular lattice, 2) Shastry-Sutherland type lattice, (which is a possible model for SrCu$_2$(BO$_3$)$_2$) 3) 1/5-depleted lattice (for CaV$_3$O$_9$). We find in these systems that magnetization plateaus can appear owing to a transition from superfluid to a Mott insulator of magnetic excitations. The plateau states have CDW order of the excitations. The magnetizations of the plateaus depend on components of the magnetic excitations, range of the repulsive interaction, and the geometry of the lattice.

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In some one-dimensional spin systems, spin-density-wave states with finite spin gap appear under a finite magnetic field accompanying plateau structures in the magnetization process. Magnetization plateaus were observed in some quasi one-dimensional materials [1]. Theoretical arguments clarify that the appearance of the plateau is explained by an insulator-conductor transition of magnetic excitations [2]. In two- or higher-dimensional systems, magnetization plateaus have been also found in both theoretical [3,4] and experimental studies [5]. In this paper, we propose a rather general picture that these two-dimensional plateaus are formed owing to field-induced insulator-superfluid transitions of magnetic excitations. To demonstrate how it works, we discuss three examples in details.

The first example is a family of antiferromagnets on a triangular lattice. For the $S = 1/2$ antiferromagnet on a triangular lattice (AFT), Nishimori and Miyashita [6] found a magnetization plateau at $m/m_{\text{sat}} = 1/3$, which comes from the appearance of a collinear state with three sublattices, i.e., the so-called "unit" state. This plateau was actually observed in AFT materials like CsCuCl$_3$ (Ref. 7). Recently in a multiple-spin exchange (MSE) model, which is a possible model [8] for solid $^3$He films, a magnetization plateau was predicted at $m/m_{\text{sat}} = 1/2$. In this case, the plateau is attributed to the formation of a similar collinear state but with four sublattices. The magnetization processes of these systems have been studied extensively and here we just attempt interpreting the known results to test the new picture.

We take as the second example the $S = 1/2$ Heisenberg antiferromagnet (HAF) on the Shastry-Sutherland lattice (Shastry-Sutherland model, hereafter. See Fig. 1), which is known to have an exact dimer ground state. Recently Kageyama et al. [9] found that SrCu$_2$(BO$_3$)$_2$ realizes a lattice structure equivalent to that discussed in Ref. 10 and that it has a gapful ground state. The magnetization measurements show plateaus at $m/m_{\text{sat}} = 1/8$ and 1/4. The last is the $S = 1/2$ HAF on the 1/5-depleted square lattice (Fig. 2), which includes a model Hamiltonian for CaV$_3$O$_9$. In this system, the plaquette singlet state is realized in the ground state [11].

In our picture, the plateau states can be regarded as Mott insulators of effective magnetic particles; repulsive interactions induce various kinds of charge-density-wave (CDW) long-range order leading to a finite energy gap in particle-hole excitations. Except for the plateau phases, magnetic particles are conducting to form supersolid, in which superfluidity and CDW coexist, and magnetization increases smoothly. Of course, the charge density is translated into the spin ($S^z$) density and superfluidity here means long-range order in the direction perpendicular to the field. Although this essential picture is common to the three examples, the concrete forms of the magnetic particles are different.

In the first example, i.e. AFT and an MSE model, a single flipped spin itself works as the magnetic particle, while the triplets on the dimer ($J$) bonds are the relevant particles of the Shastry-Sutherland model. We find plateaus at $m/m_{\text{sat}} = 1/2$ and 1/3. In the third one, i.e. the 1/5-depleted square lattice, plaquette triplets behave as particles. We predict plateaus at $m/m_{\text{sat}} = 1/8, 1/4, 1/2$.

The magnetizations where a plateau appears depend on (i) the form of the magnetic excitations, (ii) range of the repulsion between them, and (iii) the geometry of the lattice. Finally we summarize common features on properties of the phase transition.

Spin = magnetic particle: The magnetization plateau for the AFT system was found in spin 1/2 anisotropic and isotropic Heisenberg models. The Hamiltonian is

$$H = J \sum_{(i,j)} (S^x_i S^x_j + S^y_i S^y_j + \eta S^z_i S^z_j) - B \sum_i S^z_i, \quad (1)$$

where the summation runs over all nearest-neighbor pairs and $B$ denotes the magnetic field. For $\eta \geq 1$, the magnetization curve has a plateau at $m/m_{\text{sat}} = 1/3$. The
the ground state in the plateau phase is of a collinear structure with three sublattices, where two of three spins direct upward and the other downward. In the other phases, magnetic states have non-collinear structures with off-diagonal long-range order (ODLRO). If we introduce a particle picture, i.e., recognize spin dynamics as induced by the motion of a certain kind of particles, the appearance of plateau is easily understood from simple consideration about compressibility of the particles. Regarding an up spin as a hard-core boson and a down one as a vacancy, we can rewrite the Hamiltonian as

\[ H = \frac{J}{2} \sum_{i,j} \left( \langle b_i^+ b_j + \text{h.c.} \rangle + 2\eta n_i n_j \right) - (B + 3J\eta) \sum_i n_i, \]

where \( b_i^+ \) denotes the creation operator of the hard-core boson on site \( i \), and \( n_i \) the number operator. Since particles carry magnetic moment unity, the chemical potential \( \mu = B + 3J\eta \) is controlled by the magnetic field and the \( \mu \)-dependence of the particle density \( n \) corresponds to the magnetization curve of the original spin system.

The hopping term comes from the spin exchange (XY) term and the repulsive interaction from the diagonal (Ising) part. The anisotropic case, \( \eta > 1 \), is mapped to the strong coupling (i.e., strong repulsion) region of the corresponding boson system. The particle-hole transformation converts the system into that of holes with repulsion of the same strength; in the strong coupling limit, the ground state at the filling \( n = 2/3 \) (\( m/m_{\text{sat}} = 1/3 \)) has the density wave long-range order, with the three-sublattice structure shown in Fig. 3(a). Due to the repulsive interaction, this state is incompressible, i.e. \( dn/d\mu = 0 \), and density-fluctuation energy has a finite gap above the ground state. Except for the filling(s) \( n = 2/3 \) (and \( 1/3 \)), there are vacancies and hence particles are mobile (conducting). Since the particles obey boson statistics and the system is uniform, the system presumably shows superfluidity. There is perfect correspondence between the above consideration and the previous results in the insulating CDW state with \( dn/d\mu = 0 \), is consistent with the spin collinear state, where susceptibility is vanishing. On the other hand, superfluidity of bosons corresponds to non-collinear ODLRO of spins.

The particle density where CDW stabilizes depends on the range of repulsive interactions. To see this, we next discuss MSE model with four-spin exchange on the triangular lattice, where repulsion acts further than in the Heisenberg model. The Hamiltonian is given by

\[ \mathcal{H} = J \sum_{\langle i,j \rangle} \sigma_i \cdot \sigma_j + K \sum_p h_p - B \sum_i \sigma_i^z, \]

where \( \sigma_i \) denote Pauli matrices. The second summation runs over all minimum diamond clusters and \( h_p \) is the four-spin exchange \( h_p = 4(P_3 + P_4^{-1}) - 1 \) with the ring permutation of four spins \( P_4 \). It was shown that three- and four-spin exchange interactions are very strong in two-dimensional solid \( ^3 \)He due to strong quantum fluctuations. Theoretically a magnetization plateau was found at \( m/m_{\text{sat}} = 1/2 \) instead of \( m/m_{\text{sat}} = 1/3 \). In the particle picture, bosons feel the following two-body repulsion

\[ V = 4(J + 5K) \sum_{\langle i,j \rangle} n_i n_j + 4K \sum_{\langle i,j \rangle} \epsilon^{\langle i,j \rangle} n_i n_j. \]

The repulsive interaction acts in both nearest- and next-nearest-neighbor sites. Figure 3(b) shows the region where the interaction works. Because of the range of repulsion, the particles can solidify at the density \( n = 1/4 \). (Note that the solidification occurs only if the repulsion overcomes the effect of the hopping term.) This insulating phase at \( n = 1/4 \) corresponds to the magnetization plateau at \( m/m_{\text{sat}} = 1/2 \) in the original spin system. The previous numerical result in Ref. on the ground state of the plateau phase is consistent with the CDW order shown in Fig. 3(b).
and 1/4. Because of the special structure of the lattice, the lattice realizes for $J'/J < 0.69$. (Ref. 13) Recently Kageyama et al. found this lattice structure in SrCu$_2$(BO$_3$)$_2$ and observed the magnetization plateaus at $m/m_{sat} = 1/8$ and 1/4. Of the special structure of the lattice, a triplet excitation is almost localized. Considering the dimer triplet state with $S^z$ = 1 as a particle (a hard-core boson by definition) and the dimer singlet as a vacancy, we derive an effective Hamiltonian for it using the perturbational expansion from the $J' = 0$ limit. The expansion is performed up to the 3rd order in $J'/J$ from degenerate states with a constant number of dimer triplets. The effective Hamiltonian up to the 2nd order is

$$H = \left( J - B - \frac{J'^2}{J} \right) \sum_i n_i + \left( \frac{J'}{2} + \frac{J'^2}{2J} \right) \sum_{(i,j)} n_i n_j$$

$$+ \frac{J'^2}{4J} \sum_{i \in A} \left\{ b_i^\dagger (b_i e_1 - b_{i-1} e_2) h.c.n_i e_2 - n_i e_2 \right\}$$

$$+ 2n_{i+e_2} (1 - n_i) n_{i-2} + (b^\dagger_{i+e_1} b_{i-e_1} + h.c.n_i)$$

$$+ \frac{J'^2}{4J} \sum_{i \in B} \{ e_1 \leftrightarrow e_2 \},$$

where $i(j)$ runs over an effective square lattice consisting of dimer bonds (both horizontal and vertical) and horizontal (vertical) ones belong to $A$ ($B$) sublattice. The full form of the effective Hamiltonian up to the 3rd order will be published elsewhere. The derived Hamiltonian does not have the one-particle hopping term (as was already reported in ref. 13), but contains many correlated-hopping processes, where an effective hopping of a particle is mediated by another one. This is one of our main observations. Most 3rd-order terms concern the correlated hopping. Longer-range repulsions between particles appear from higher-order perturbations. Diagonal repulsive interactions up to the 3rd order in $J'/J$ are shown graphically in Fig. 4. The resulting Hamiltonian does not have 90° rotational invariance, since the lattice structure has low symmetry, and this may lead to highly anisotropic CDW states.

We study the effective Hamiltonian in the classical limit. To this end, we map the hard-core boson system to the $S = 1/2$ quantum spin system and then approximate the spin-1/2 by a classical unit vector. We search for the ground state with large sublattice structures (e.g. a stripe-like one with 6-sublattice) both with the mean-field approximation and a Monte Carlo method by decreasing temperatures gradually. The evaluated magnetization process is shown in Fig. 5. Note a clear difference between the high- and low-field region. There appear plateau structures at $m/m_{sat} = 1/2$ and 1/3. The plateau states have CDW long-range orders shown in Fig. 6. Configurations realized for $m/m_{sat} = 1/2$ and 1/3 correspond to perfect closed packings provided that particles avoid repulsion from 1st- and 2nd-order perturbation, respectively. The plateau at $m/m_{sat} = 1/2$ appears only in the region $0 < J'/J < 0.50$ and, for large $J'/J$ the CDW is destroyed by the correlated hoppings, which are dominant in the higher order terms. The correlated hoppings are so efficient also at large particle density that any plateau does not appear for $1/2 < m/m_{sat} < 1$. Below $m/m_{sat} = 1/3$, the correlated hoppings occur rarely, because of a low particle density. The observed 1/4-plateau (and 1/8-plateau) of SrCu$_2$(BO$_3$)$_2$ may be formed by weak longer-range repulsions which are not taken into account in the present study. Recently Miyahara and Ueda discussed semi-phenomenologically that the 1/4-plateau might come from a CDW state with a stripe structure. In our approach, the repulsive interaction relevant to the stripe CDW may come from the higher-order terms in perturbation, otherwise from other spin interactions that are not considered in the Shastry-Sutherland model. This remains to be a future problem.
Plaque triplet: When the interactions or a special geometry of the lattice allows four-spin plaquettes, in each of which four spins are coupled more strongly than to the others, individual plaquettes form singlets in the ground state. The triplet states with \( S^z = 1 \) on plaquettes are dominant excitations in a weak magnetic field. The insulator-conductor transition of these excitations can take place thereby producing magnetization plateaus.

An example of the plaquette singlet ground states is seen in the \( S = 1/2 \) HAF on the 1/5-depleted square lattice, which includes a possible model for \( \text{CaV}_2\text{O}_5 \) as a special case. The lattice is shown in Fig. 6. In the isolated plaquette limit \( J_1 = J_2 = 0 \), a trivial plateau already appears at \( m/m_{\text{sat}} = 1/2 \) for \( J < B < 2J \) (see Fig. 6), where every plaquette is in the triplet excited state with \( S^z = 1 \). When \( m/m_{\text{sat}} < 1/2 \), the triplet excitations (particle) tend to hop if \( J_1 \) and \( J_2 \) are turned on, and at specific (commensurate) values of \( m/m_{\text{sat}} \) they can show insulator-conductor transitions as a consequence of the competition between the hopping and the repulsive interaction. Above \( m/m_{\text{sat}} = 1/2 \), a plaquette quintuplet \( (S = 2) \) with \( S^z = 2 \) behaves as a particle and can show magnetization plateaus between \( 1/2 < m/m_{\text{sat}} < 1 \). In the following, we focus on a weak magnetic-field region, which corresponds to the magnetization \( 0 < m/m_{\text{sat}} < 1/2 \). Regarding the plaquette triplet excitation with \( S^z = 1 \) as a particle and the singlet state as a vacancy, we derive the effective Hamiltonian of the particle by the 2nd-order perturbation around the limit \( J_1 = J_2 = 0 \). (The explicit form will be shown elsewhere\(^{[1]}\)). The repulsive interactions range from a plaquette to its nearest- and next-nearest neighbors. If parameters satisfy \( J_1 \approx 2J_2 \), the hopping term is weak and hence the system is in the strong coupling regime. Then we may expect that the triplets crystallizes and the magnetization plateaus appear at \( m/m_{\text{sat}} = 1/8 \) and \( 1/4 \). The mean-field approximation of the effective Hamiltonian indeed shows magnetization plateaus at \( m/m_{\text{sat}} = 1/8 \) and \( 1/4 \) (see Fig. 6). They come from the insulating phases with CDW long-range order of a square structure. A remark is in order here about the relevance of our results to \( \text{CaV}_2\text{O}_5 \). Quite recently, it was shown\(^{[1]}\) that plaquettes of another type (metaplaquettes) consisting of \( J_2 \)-bonds play the main role in \( \text{CaV}_2\text{O}_5 \) contrary to earlier studies\(^{[2]}\). The physics is, however, almost the same also in this case; the metaplaquette excitations behave like particles and \( J_2 \)- and \( J_1 \)-bonds induce hopping, and so on. The detailed results will be reported in a longer paper\(^{[1]}\).

Common features: To conclude this paper, we discuss a few features shared by the three examples. According to an analogy to many-particle theories, a plateau state corresponds to a CDW insulating state and gapless ones to supersolids. As the plateau state collapses by increasing the applied field, superfluidity appears, whereas CDW exists in both phases. Let us consider the case of 2nd order transition, where the magnetization changes continuously. Assuming that the on-set of superfluidity is well described by the effective Hamiltonian of the ordinary bosons with a short-range repulsion for low energies, we conclude this transition is of the dynamical exponent \( z = 2 \), magnetization increases linearly like \( |H - H_c| \) apart from possible logarithmic corrections. (Note that the form is quite different from that in 1D.)

Note added in proof: K. Onizuka et al. recently observed a clear 1/3 plateau in \( \text{SrCu}_2(\text{BO}_3)_2 \), which we had predicted in this paper and had argued to be of a stripe structure.

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