Local physics of magnetization plateaux in the Shastry-Sutherland model

L. Isaev¹, G. Ortiz¹, and J. Dukelsky²

¹Department of Physics, Indiana University, Bloomington IN 47405, USA
²Instituto de Estructura de la Materia - CSIC, Serrano 123, 28006 Madrid, Spain

We address the physical mechanism responsible for the emergence of magnetization plateaux in the Shastry-Sutherland model. By using a hierarchical mean-field approach we demonstrate that a plateau is stabilized in a certain spin pattern, satisfying local commensurability conditions derived from our formalism. Our results provide evidence in favor of a robust local physics nature of the plateaux states, and are in agreement with recent NMR experiments on SrCu₂(BO₃)₂.

PACS numbers: 75.10.Jm, 75.60.Ej

Introduction.– The interplay between quantum mechanics and the atomic lattice topology often leads to a complex mosaic of physical phenomena in low-dimensional frustrated magnets. A prominent representative of this class of materials is the layered compound SrCu₂(BO₃)₂, which recently received a lot of attention because of its fascinating properties in an external magnetic field, namely the emergence of magnetic plateaux at certain fractions of the saturated magnetization $M_{sat}$. The first experimental observations of the plateaux were reported in $m$ for $m = M/M_{sat} = 1/8$ and 1/4, and somewhat later for $m = 1/3$. Subsequent nuclear magnetic resonance (NMR) experiments revealed spontaneous breaking of the lattice translational symmetry within the 1/8 plateau, and also indicated that the spin superlattice persists right above this fraction. The field was reignited by the work of Sebastian et al., where additional plateaux at exotic values $m = 1/9, 1/7, 1/5$ and 2/9 were reported. However, direct observation of the emerging spin superstructures remains an experimental challenge, primarily due to the high magnetic fields ($\sim 30 – 50$ Tesla) involved in measurements.

The nature of the magnetic states and physical mechanism leading to the plateaux are also yet to be understood. It is believed that the Heisenberg antiferromagnetic model on a frustrated Shastry-Sutherland (SS) lattice with $N$ sites captures the essential magnetic properties of SrCu₂(BO₃)₂ in relatively high fields. In Eq. 1 $S_i$ denotes a spin-1/2 operator at site $i$; the first sum is the usual nearest-neighbor (NN) Heisenberg term, while the second one runs over dimers; $J$ and $\alpha \geq 0$. This model is quasi-exactly solvable for $\alpha \geq 1 + h/2$: the ground state (GS) is a direct product of singlet dimer states, and was shown to be stable up to $\alpha \sim 0.71$-0.75 in zero field. In general, it is an intractable quantum many-body problem where approximation schemes are needed to deal with large-$N$ systems.

All theories proposed to address this unusual magnetization phenomenon start from the SS model. However, the physical mechanism stabilizing the plateau states, their nature, and the structure of the magnetization curve are still actively debated. Current ideas can be broadly divided into two groups. The first one advocates subtle non-local (in the spins) correlations leading to an underlying spin structure which preserves lattice symmetries. Technically, it employs a mapping of the original spins to fermions coupled to a Chern-Simons gauge field, and then performs a Hartree-Fock decoupling. In this way, the qualitative shape of the SrCu₂(BO₃)₂ magnetization curve was reproduced in high fields, but the lowest plateau at 1/8 was missing. Later, this non-local mean-field approach was extended to include inhomogeneous phases, and it was argued that the plateaux correspond to stripe states with broken lattice symmetries. Remarkably, the length scale $\xi$ associated with the emerging spin superlattice was found to be $\xi \sim 100$ lattice spacings. The second group contends that the magnetization process can be described in terms of polarized dimers (triplons), which propagate in the background of singlet dimers. They developed effective core boson models (truncating the original dimer Hilbert space), solved by perturbative or CORE techniques, and found that the plateaux states correspond to crystal phases with $\xi \sim 10$ lattice constants.

\[ H = J \sum_{\langle ij \rangle} S_i \cdot S_j + 2 \alpha \sum_{\langle ij \rangle} S_i \cdot S_j - h \sum_i S_i^z, \]
Such diversity of theoretical predictions demands further investigation. In this paper we use the hierarchical mean-field (HMF) method \[14\], in an attempt to clarify the nature and physical mechanism, responsible for the emergence of magnetic plateaux in the SS model. Unlike previous calculations, we deal directly with the SS Hamiltonian \[1\], not with effective Hamiltonians as in Refs. \[12\, 13\], and combine exact diagonalization data with a simple and controlled approximation for the GS wavefunction. For instance, we do not discard the \( M = 0 \) dimer triplet states, necessary for the propagation of a triplon. We focus on higher-lying fractions, whose existence has been confirmed experimentally. Our results support the local physics nature of the plateau states. In particular, it is explicitly demonstrated how to construct those robust states based on a set of commensurability rules that we derived. Our conclusions are also in agreement with the interpretation of NMR measurements \[3\].

Method. – The HMF approach is based on the assumption that the physics of the problem is local in a particular representation. Since the SS model is formulated in terms of localized spins, it is natural to work in real space. The main idea of our method revolves around the concept of a relevant degree of freedom – a spin cluster – which is used to build up the system. Essential quantum correlations, which drive the physics of the problem, are captured by this local representation. The SS Hamiltonian is then rewritten in terms of these coarse-grained variables and a mean-field decoupling is eventually applied to determine properties of the system. The method is only limited by finite-size effects and becomes asymptotically exact in the thermodynamic limit. Thus, the (generally) exponentially hard problem of determining the GS of the model is reduced to a polynomially complex one. By identifying each state of a cluster with a Schwinger boson (SB) and computing matrix elements of spin operators between these states, one can rewrite exactly the Hamiltonian of Eq. (1) in terms of new (cluster) variables

\[
H = \sum_i \epsilon_a(\alpha, h) \gamma_i^\dagger \gamma_i + \sum_{\langle ij \rangle_\sigma} (H_{\text{int}}^\sigma)_{ab} \gamma_i^{\alpha'} \gamma_j^{\beta'} \gamma_i^\dagger \gamma_j^\dagger \gamma_i^\dagger \gamma_j^\dagger.
\]

Here the repeated indices \( a, b \), etc., which label states of an \( N_q \)-spin lattice, are summed over, and \( i \) denotes sites in the coarse-grained lattice. The operators \( \gamma_i^\dagger \) that create a particular state of a cluster are \( SU(2^{N_q}) \) SBs subject to the constraint \( \sum_a \gamma_i^{\alpha a} \gamma_i^a = 1 \) on each site; \( \epsilon_a \) are exact cluster eigenenergies. Since the original SS Hamiltonian involves only two-spin interactions, in the new representation there will be only two-boson scattering processes: the corresponding matrix elements are denoted by \( (H_{\text{int}}^\sigma)^{ab}_{\gamma \gamma'}. \) The second term in Eq. (2) describes the renormalization of the cluster energy due to its interaction with environment. Thus, our method deals with an infinite system and finite-size effects enter only through a particular choice of the cluster. The symbol \( \langle ij \rangle_\sigma \), with \( \sigma = 1, 2, \ldots \), indicates pairs of neighboring blocks, coupled by the same number of \( J \)-links.

Application of the HMF method to the SS model starts by recalling that the phases within plateaux break the lattice translational invariance. Therefore, the best solution will be obtained, if the degree of freedom matches the unit cell of the spin superstructure. For each cluster size, \( N_q \), and magnetization

\[
m = \frac{1}{M_{\text{sat}}} \sum_i \langle S_i^z \rangle = \frac{2}{N} \sum_i \langle S_i^z \rangle = \frac{2}{N_q} \sum_{j=1}^{N_q} \langle S_j^z \rangle,
\]

we determine the lowest-energy configuration (i.e., the cluster shape and corresponding tiling of the lattice). By virtue of previous argument, this solution will have the “right” symmetry. Performing this operation for successive values of \( N_q \) up to the largest one that can be handled, we obtain a set of magnetization plateaux together with their corresponding spin profiles. It follows that the particular choice of coarse graining is critical for the success of this program. One should recall that the experimental value for \( \alpha \) is 0.74-0.84, i.e. the intradimer coupling seems to be “more relevant” than the interdimer one. Therefore, it is natural to consider only those clusters, which contain an integer number of \( \alpha \)-links. This constraint turns out to be quite severe. It follows that the degree of freedom must also contain an integer number of “minimal” blocks, shown in Fig. 1 in black: otherwise the tiling of the lattice will not be complete. These requirements comprise a set of local commensurability conditions, necessary to stabilize a plateau.

Another crucial issue is the way the interaction terms in Eq. (2) are handled. In an attempt to simplify matters, we use the straightforward Hartree approximation, i.e., we consider the trial GS wavefunction

\[
|\psi_0\rangle = \prod_i (R_a \gamma_i^\dagger |0\rangle); \quad R_a^* R_a = 1.
\]

Here \( |0\rangle \) is the SB vacuum and \( R_a \) are variational parameters, which constitute the cluster wavefunction. Since \( H \) is real-valued, we can choose \( R_a \) to be also real. Clearly, this state is cluster translationally invariant and has exactly one boson per coarse-grained lattice site (so the constraint is exactly satisfied). Next, we compute the expectation value of \( H \) in the state \( |\psi_0\rangle \), subject to periodic boundary conditions, and minimize it with respect to \( R_a \). In this manner one obtains the approximate GS energy \( E_0 \) as a function of the magnetic field \( h \).

It is important to emphasize the simplicity of our approach. By using a more sophisticated ansatz (e.g. a Jastrow-type correlated wavefunction \[14\]), we could improve energies but the physical mechanism and robust structure of the plateaux will remain intact. Despite its simplicity, the ansatz of Eq. (3) was accurate enough to
yield the quantitatively correct phase diagram of the $J_1$-$J_2$ model \cite{14}, which involves gapless phases. In the SS model states within the plateaux are gapped, therefore, our method should be tailored for this problem.

Results.– To guarantee that our results reduce to the exact solution in the limit $h \rightarrow 0$, we mainly consider the region $\alpha \gtrsim 1$. The simplest degree of freedom, consisting of 4 spins, is shown in Fig. 1. Using this cluster in our HMF scheme one obtains stable plateaux only at $m = 1/3$ and $m = 1$. Clearly, larger blocks are necessary to stabilize plateaux at lower magnetization fractions. Here, we consider cluster sizes $N_q = 4k$ with $k = 2, \ldots, 6$ and discuss only plateaux at $1/3$, $1/4$, $1/5$, $1/6$ and $1/8$, supported in minimal clusters of $N_q = 12, 8, 20, 12$ and 16 spins, respectively ($M_{\text{sat}} = N_q/2$). In Fig. 2 we present local spin profiles, corresponding to the lowest energy configurations, for each of these fractions. Comparison of patterns for different plateaux, shows that states $1/n$ with $n$ even are characterized by one polarized dimer per unit cell, while cells of odd-$n$ states have two triplons. For a given plateau, there typically exist several possible coarse-graining scenarios, characterized by different clusters and tessellations of the SS lattice, but identical unit cells. Although these configurations have slightly different energies, their existence provides an important check for robustness of local correlations, stabilizing the plateau states. The patterns for all fractions except $1/5$, are similar to those obtained in Refs. \cite{1,12}. Strictly speaking, the profiles in Fig. 2 are well defined only for large values of $\alpha > 1$ and quickly smear out with decreasing $\alpha$. This effect is difficult to capture within the effective model calculations, like \cite{12}.

A clear advantage of our approach, compared to previous works, is its ability to compute GS energies of the original SS model. Within each plateau we have: $E_0(h)/N = -\varepsilon_0 - mh/2$. The parameter $\varepsilon_0$ is presented in Table I for some values of $\alpha$ and different cluster sizes. In order to address finite-size effects, in Fig. 3 we present the high magnetic field phase diagram of the SS model for $\alpha \gtrsim 1$. All fractions were calculated using the largest possible cluster. Due to the insulating nature of the plateau states, the finite-size corrections are not expected to significantly affect their stability. For instance, for $m = 1/6$ the energy difference between 12- and 24-spin clusters is only $\sim 5\%$ of its width for the values of $\alpha$ shown in Table I. This observation serves as additional evidence in favor of a universal physical mechanism leading to the plateaux.

As it was already mentioned, the HMF method does not involve truncation of the dimer Hilbert space. In order to understand consequences of this approximation, we computed $\varepsilon_0$ for different plateaux, ignoring the dimer state $|\uparrow\uparrow\rangle$. The resulting absolute error is of the same order of magnitude as finite size effects and plateau widths (cf. Table I), which leads to a sizable change in the relative stability of the plateaux. For example, at $\alpha = 1.1$ the average error is $3 \cdot 10^{-3} J$, and the lower boundary of the $1/8$ state shifts by $0.04 J$. Therefore, conclusions of the effective boson model calculations, which employ similar truncation, should generally be taken with caution.
Discussion. – Although the effective model approach does yield a sequence of plateaux, their understanding remains incomplete. Our work addresses this issue by focusing on the nature and correlations of the magnetic plate states. In particular, the analysis presented focusing on the nature and correlations of the magnetic plate states. First, the experimental observed plateaux at 1/4 and 1/8, which we found to be quite robust, were claimed to be unstable in [12]. However, the magnetization profile, presented in Fig. 2 for m = 1/8, which persists at α = 0.787, adequate for SrCu$_2$(BO$_3$)$_2$, is consistent with the interpretation of available NMR data [4, 5] for this material. We believe that the origin of these states is purely magnetic and no additional interactions beyond the SS model are required, in contradiction with the claim of Ref. [12]. Our results also yield a stable 1/5 plateau, contrary to the conclusions of Refs. [12] and [13]. We note that this fraction was observed in torque measurements of [7], however, their proposed spin superlattice differs dramatically from the one predicted in our Fig. 2. Another distinction concerns the robustness of the 1/6 plateau advocated in [12], which, although present in our calculation, has a significantly smaller relative width (see the discussion above). Other fractions at 1/9, 2/9 and 2/15, observed in Refs. [12] and [13], can also be obtained within our approach, but this requires significantly larger clusters than the ones used here. By virtue of our commensurability arguments, we expect the plateaux at 1/9 and 2/9 to emerge in degrees of freedom containing at least 36 spins, while the 2/15 fraction will be stabilized in a 60-spin cluster.

Finally we note that the precise shape of the magnetization curve (the relative energy stability of different plateaux) is quite sensitive to the value of α (i.e., the particular compound) and, most importantly, since there is no exact solution of the SS model at these high fields, it depends on the particular approximation scheme. Experimentally, other physical interactions not included in the SS model may also add to this uncertainty.

Acknowledgements. – We thank C. D. Batista for stimulating discussions. Calculations were performed on Quarry and NTC clusters at IUB. JD acknowledges support from the Spanish DGI grant FIS2006-12783-C03-01.

[1] J. Richter, J. Schulenburg and A. Honecker in Quantum Magnetism, U. Shollwöck, J. Richter, F. J. J. Farnel and R. F. Bishop eds., Springer-Verlag, Berlin 2004.
[2] H. Kageyama et al., Phys. Rev. Lett. 82, 3168 (1999).
[3] K. Onizuka et al., J. Phys. Soc. Jpn. 69, 1016 (2000).
[4] K. Kodama et al., Science 298, 395 (2002).
[5] M. Takigawa at al., J. Phys. Conf. Ser. 51, 23 (2006).
[6] M. Takigawa et al., Phys. Rev. Lett. 101, 037202 (2008).
[7] S. E. Sebastian et al., PNAS 105, 20157 (2008).
[8] B. S. Shastry and B. Sutherland, Physica 108B, 1069 (1981).
[9] S. Miyahara and K. Ueda, J. Phys.: Cond. Mat. 15, R327 (2003).
[10] G. Misguich, T. Jolicoeur, and S. M. Girvin, Phys. Rev. Lett. 87, 097203 (2001).
[11] S. Miyahara and K. Ueda, Phys. Rev. Lett. 82, 3701 (1999); Phys. Rev. B61, 3417 (2000). T. Momoi and K. Totsuka, Phys. Rev. B62, 15067 (2000).
[12] J. Dorier, K. P. Schmidt, and F. Mila, Phys. Rev. Lett. 101, 250402 (2008).
[13] A. Abendschein and S. Capponi, Phys. Rev. Lett. 101, 227201 (2008).
[14] L. Isaev, G. Ortiz, and J. Dukelsky, Phys. Rev. B79, 024409 (2009).