# Theory of Magnetization Plateaux in the Shastry-Sutherland Model

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Motivated by the remarkable properties of $SrCu_2(BO_3)_2$ in a magnetic field, we use perturbative continuous unitary transformations to determine the magnetization plateaux of the Shastry-Sutherland model, unveiling an unexpected sequence of plateaux progressively appearing at $2/9$, $1/6$, $1/9$, and $2/15$ upon increasing the interdimer coupling. We predict that a $1/6$ plateau should be present in $SrCu_2(BO_3)_2$, even if residual interactions beyond the Shastry-Sutherland are strong enough to modify the other plateaux below $1/3$. The method is extended to calculate the magnetization profile within the plateaux, leading to a local structure around triplons that agrees with NMR results on $SrCu_2(BO_3)_2$.

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Strongly frustrated quantum magnets in an external field are fascinating systems because the interplay between interactions and kinetics can lead to very rich phase diagrams. The magnetization can be described as a gas of bosonic particles whose density is controlled by the external magnetic field, and since frustration typically reduces the kinetic energy, Mott-insulating (corresponding to magnetization plateaux) [1], superfluid or even supersolid phases have been predicted to occur [2]. The experimental observation of these phases is an on-going challenge. A major player in the field is the layered copper oxide $SrCu_2(BO_3)_2$, in which several magnetization plateaux have been observed [3,4]. However, the definitive sequence of plateaux and the presence of supersolid phases remain open (sometimes controversial) issues that call for further experimental and theoretical investigation.

The magnetization of $SrCu_2(BO_3)_2$ is expected to be described by the 2D spin-$1/2$ Heisenberg model known as the Shastry-Sutherland model [5] in a magnetic field

$$H = J' \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J \sum_{\langle\langle i,j \rangle\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - B \sum_i S_i^z,$$

with $J'/J \simeq 0.63$, where the $\langle\langle i, j\rangle\rangle$ bonds build an array of orthogonal dimers while the $\langle i, j\rangle$ bonds are best seen as interdimer couplings (see Fig. 1). For $J'/J$ smaller than a critical ratio of order 0.7, the ground state of the model is exactly given by the product of dimer singlets, and the magnetization process can be described in terms of hard-core bosons which stand for polarized triplons $|t^1\rangle = |\uparrow\uparrow\rangle$ on the dimers interacting and moving on an effective square lattice [6,7].

All theoretical approaches agree on the presence of magnetization plateaux at $1/3$ and $1/2$ [6–10], in agreement with experiments [3,11]. However, the structure below $1/3$ is rather controversial. On the experimental side, the original pulsed field data have only detected two anomalies interpreted as plateaux at $1/8$ and $1/4$ [3], but the presence of additional phase transitions and of a broken translational symmetry above the $1/8$ plateau has been established by recent torque and NMR measurements up to 31 T [12,13]. The possibility of additional plateaux has been pointed out by Sebastian *et al.* [11], who have interpreted their high-field torque measurements as evidence for plateaux at $1/q$ with $2 \leq q \leq 9$ and at $2/9$. On the theoretical side, the situation is not settled either. The finite clusters available to exact diagonalizations prevent reliable predictions for high-commensurability plateaux, and the accuracy of the Chern-Simons mean-field approach initiated by Misguich *et al.* [9] and recently used by Sebastian *et al.* [11] to explain additional plateaux is hard to assess. The essential difficulty lies in the fact that, since plateaux come from repulsive interactions between triplons, an accurate determination of the low-density, high-commensurability plateaux requires a precise knowledge of the long-range part of the interaction, which could not be determined so far.

In this Letter, we combine perturbative continuous unitary transformations (PCUTs) [14] with an analysis of the effective hard-core boson model reformulated as a spin model to investigate the magnetization process of the Shastry-Sutherland model. We find that below $1/3$, a rich and unexpected plateau structure emerges at small magnetization.

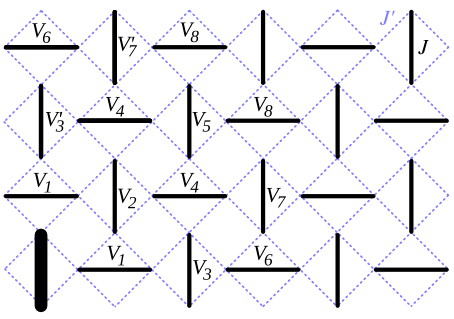


FIG. 1 (color online). Shastry-Sutherland lattice and definition of the two-body interactions. $V_n$ is the coefficient of the two-body interactions between the thick dimer and the dimer labeled $V_n$.

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The PCUT transforms the Shastry-Sutherland model into an effective model conserving the number of elementary triplets (triplons [15]). The relevant processes for the physics in a finite magnetic field have maximum total spin and total $S_z$. Other spin channels relevant for spectroscopic observables have been studied earlier [16]. The general form of the effective Hamiltonian obtained by the PCUT takes the form

$$H_{\text{eff}} = \sum_{n=2,4,6\cdots} \sum_{r_1,\cdots,r_n} C_{r_1,\cdots,r_n} b^\dagger_{r_1} \cdots b^\dagger_{r_{n/2}} b_{r_{n/2+1}} \cdots b_{r_n},$$

where the $r_i$'s are sites of the square lattice formed by the $J$ bonds, while the hard-core boson operator $b^\dagger_r$ creates a polarized triplon $|t^1\rangle$ at site $r$. The coefficients $C_{r_1,r_2,\cdots,r_n}$ are obtained as a series in $J'/J$. We have kept all terms with up to 3 creation and annihilation operators and all four-body interactions $(n_{r_1} n_{r_2} n_{r_3} n_{r_4})$ that first appear at order $\leq 8$. For the two-body interactions $(n_{r_1} n_{r_2})$ we keep more terms, namely, those that first appear at order less or equal to 10. The coefficients $C_{r_1,r_2,\cdots,r_n}$ are evaluated up to order 15 for the two-body interactions and up to order 12 for the other terms, and they are then extrapolated using Padé or $D$ logPadé extrapolants. The resulting Hamiltonian contains more than 15 000 processes.

The dominant terms of the most relevant types are illustrated in Fig. 2. For $J'/J < 0.5$, the pure density-density interaction terms have by far the largest coefficients. Besides, the magnitude of all terms decreases when sites are taken apart, and the physics at low density will be dominated by two-body interactions. The standard two-site hopping is strongly supressed due to the frustration [17], and the kinetic part of the Hamiltonian is dominated by the correlated hopping $(n_{r_1} b^\dagger_{r_2} b_{r_3})$ [6,16], a type of process recently shown to strongly favor supersolid phases [2].

The evolution with $J'/J$ of the two-body interactions defined in Fig. 1 is depicted in Fig. 3. At small $J'/J$, interactions beyond $V_4$ are small and may be neglected, but for larger $J'/J$ the higher order terms $V_3'$, $V_5$, and $V_7$ (appearing at order 6) become important and contribute to the formation of low-density plateaux. For these terms, the bare series and the Padé extrapolations are basically indistinguishable below $J'/J = 0.5$. Beyond that value and up to $J'/J \simeq 0.63$, various Padé extrapolations still give consistent results for these two-body interactions. However, local interactions involving more than two particles become quite strong for $J'/J \geq 0.6$ close to the phase transition and are very hard to extrapolate. We therefore restrict the discussion to $J'/J \leq 0.5$ where the expansion is well controlled.

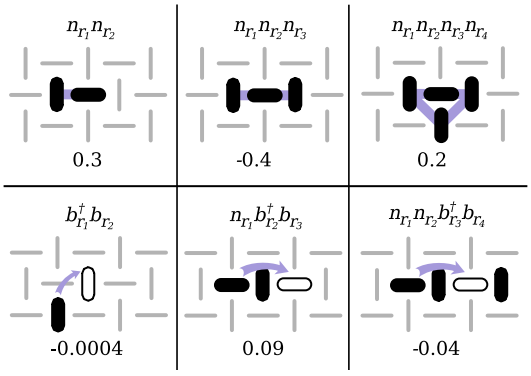


FIG. 2 (color online). Largest process of most relevant types in the Hamiltonian $H_{\text{eff}}$ and its amplitude $C_{r_1,\cdots,r_n}$ in units of $J$ at $J'/J = 0.5$.

The effective Hamiltonian $H_{\text{eff}}$ in a magnetic field is by no means simpler than the original one in general, but it is in the limit of small density and not too large $J'/J$. Indeed, in that limit the kinetic terms are very small, and they can be considered as a perturbation of the interaction part, which is diagonal in the local Fock basis $|n_{r_1}, n_{r_2}, \ldots\rangle$. It is thus appropriate to use a Hartree approximation in which the variational ground state is a product of local boson wave-functions since this approximation becomes exact in the limit of vanishing kinetic energy. In fact, even in models with significant kinetic energy, this approximation has proven to be remarkably good when a comparison to QMC was possible [2,18]. This Hartree approximation is most simply implemented by mapping $H_{\text{eff}}$ onto a spin 1/2 model using the Matsubara-Matsuda representation [19] of hard-core bosons $S^+ = b$, $S^- = b^\dagger$, $S^z = 1/2 - b^\dagger b$ since it just translates into the classical approximation (CA) where the spins are treated as classical vectors of length 1/2. Given the huge number of terms of the model, the energy is minimized numerically on finite size clusters with periodic boundary conditions. To allow for high commensurabilities of various symmetries, all nonequivalent clusters with up to 32 sites for all $J'/J$ (64 for $J'/J = 0.5$) have been tested and compared.

The resulting phase diagram is shown in Fig. 4, and the structure of the plateaux in Fig. 5. In Fig. 4, we only quote

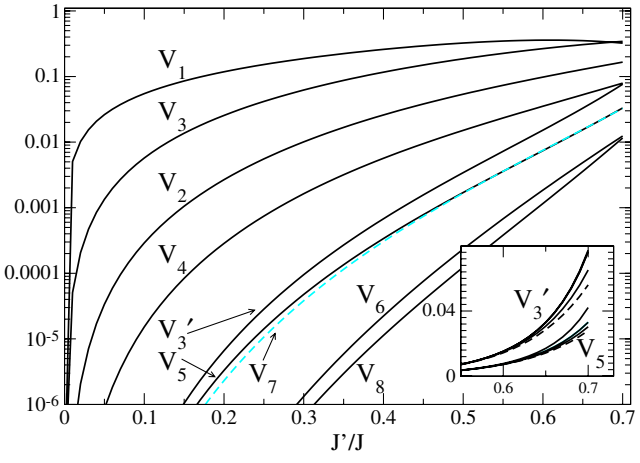


FIG. 3 (color online). Coefficients of the extrapolated two-body interactions as a function of $J'/J$. Inset: Different extrapolants (solid lines) as well as the bare series (dashed lines) for $V_3'$ and $V_5$.

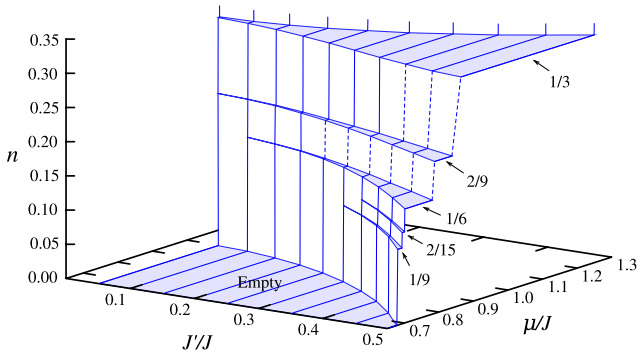


FIG. 4 (color online). Magnetization plateaux as a function of $\mu$ and $J'/J$. The boson density $n$ is equal to the magnetization in units of the saturation value, and the chemical potential $\mu$ is equal to the magnetic field $B$. Solid line denote results that are fully converged with respect to the terms kept in the Hamiltonian (see Fig. 6). Well-converged results are then connected by dashed lines.

results fully converged with respect to the terms kept in the Hamiltonian (see Fig. 6). The phase diagram is dominated by a series of plateaux, at $1/3$ and $1/2$ (not shown) already at very small $J'/J$, then by plateaux at $2/9$, $1/6$, $1/9$ and $2/15$. This plateau structure was to be expected since the kinetic terms are quite small, and since, if they were completely absent, the magnetization curve would simply be a sequence of plateaux. At $J'/J = 0.5$, the $1/6$ plateau is by far the most prominent structure below $1/3$ (see Fig. 6).

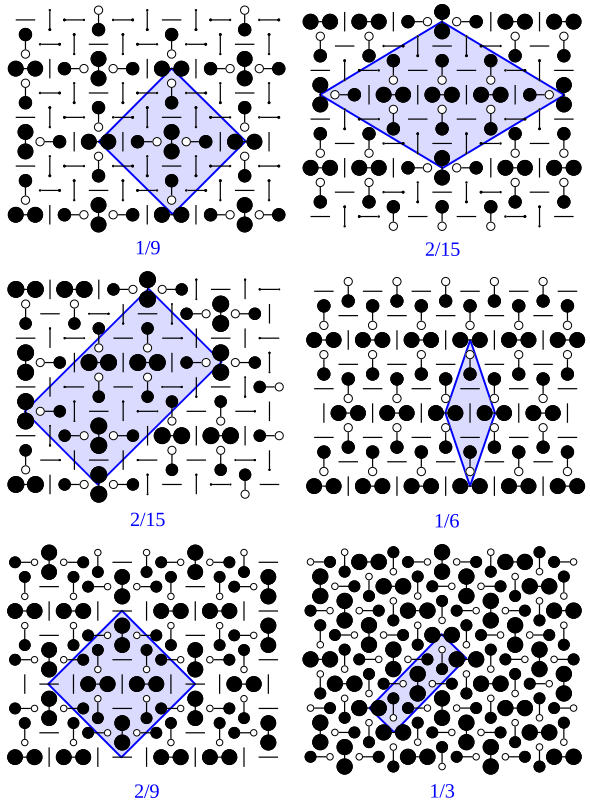


FIG. 5 (color online). Spin density ($S^z$) profile of the main plateaux at $J'/J = 0.5$. Full (empty) circles corresponds to magnetization along (opposite to) the magnetic field. The radius of the circles is proportional to the magnetization amplitude. The blue line shows the unit cell compatible with the periodicity of the state. For the $2/15$ plateau, two structures have the same energy within the error bars of the method.

Despite the various levels of approximation, we believe that this sequence of plateaux is quite robust. Indeed, the essential ingredients to stabilize plateaux are the density-density interaction terms, and the magnitude of the non-negligible ones has been determined essentially exactly for $J'/J \leq 0.5$. The kinetic terms are not expected to change this sequence in any significant way for two reasons. First of all, they are very small in the parameter range of Fig. 4. Besides, they do not tend to stabilize plateaux but superfluid or supersolid phases. So, if treated beyond the Hartree approximation, they can be expected to shift slightly the first-order transitions between plateaux or to replace them by small surperfluid or supersolid phases, but not to stabilize additional plateaux.

A remarkable advantage of using an effective spin model whose ground state is well approximated by a classical configuration is that it gives access to local observables inside the magnetization plateaux since the translational symmetry can already be broken on finite systems [20]. This has allowed us to calculate the magnetization profile inside the various plateaux (Fig. 5). In all cases, the building brick is a triplet and its two up-down neighboring dimers, in agreement with the interpretation of Cu NMR in the first plateau of $SrCu_2(BO_3)_2$ [4]. It might seem surprising that the system can have dimers in an up-down configuration since the effective model is formulated in terms of singlets and polarized triplets. This is due to the fact that, within the PCUT formalism, which is a canonical

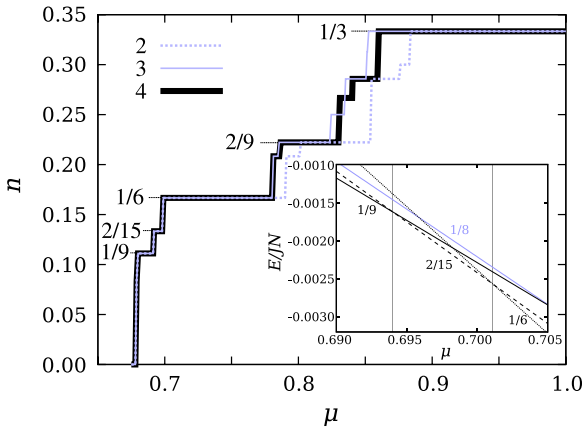


FIG. 6 (color online). Magnetization curve at $J'/J = 0.5$: comparison of the results obtained by keeping terms with up to 2 creators (dotted blue curve), 3 creators (solid blue curve) and 4 creators (black curve). Well-converged plateaux used in Fig. 4 are indicated explicitly. Inset: Energy of the $1/9$, rhomboid $1/8$, $2/15$, and $1/6$ plateaux as a function of $\mu$ at $J'/J = 0.5$. The error bars from the $D$ log Padé extrapolation are smaller than the line width.

transformation, observables have to be transformed as well with the unitary transformation $U$ [14]. So, if $|\psi\rangle$ is the ground state of the original model and $|\tilde{\psi}\rangle = U|\psi\rangle$ the ground state of the effective model, the expectation value of an observable $O$ is given by $\langle\psi|O|\psi\rangle = \langle\tilde{\psi}|UOU^\dagger|\tilde{\psi}\rangle$. In other words, a local observable such as $S^z_{r_i}$ must be calculated in $U^\dagger|\psi\rangle$, which, unlike $|\tilde{\psi}\rangle$, has configurations with dimers not only in the singlet and in the polarized triplet state, but in the other triplet states as well.

Let us now compare the present results with previous works. Momoi and Totsuka used perturbation theory to third order and logically found only plateaux at $1/3$ and $1/2$ [6]. Miyahara and Ueda used a phenomenological form of the long-range two-body interactions in a model without kinetic energy to successfully determine possible structures inside the plateaux at $1/8$, $1/4$ and $1/3$ reported in $SrCu_2(BO_3)_2$ [8]. They found several other plateaux, but the approach was not set up to be predictive regarding the actual plateaux stabilized in the model. Finally, there are obvious similarities between our results and the results of the Chern-Simons theory of Misguich *et al.* [9] recently extended by Sebastian *et al.* [11] to allow for nonuniform mean-field solutions (plateaux at $1/9$, $1/6$ and $2/9$), but some aspects of their results, for instance a well-developed plateau at $1/5$, are definitely ruled out by our analysis and must be considered as artefacts of the method.

Let us now discuss the implications for $SrCu_2(BO_3)_2$. Below $1/3$, we found that the magnetization curve of the Shastry-Sutherland model at $J'/J = 0.5$ is dominated by a large plateau at $1/6$ and three smaller ones at $1/9$, $2/15$ and $2/9$. In contrast with $SrCu_2(BO_3)_2$, we found no evidence of a plateau at $1/8$, as clearly proven by the inset of Fig. 6 where the energy of various low-density plateaux including the $1/8$ plateau with rhomboid unit cell [10] are reported. However, the Shastry-Sutherland model at $J'/J = 0.5$ is not a perfectly accurate model for $SrCu_2(BO_3)_2$ for two reasons. First of all, the ratio $J'/J$ is probably closer to 0.63. This is a parameter range that we cannot access. It is quite unlikely however that the physics changes dramatically. Indeed, upon increasing $J'/J$, a phase transition only occurs around 0.7, and more importantly, it was found to be first order [21]. So we expect the physics to evolve smoothly up to 0.7 [22], and to change abruptly above this value. This is further supported by the very smooth evolution of the magnetization curve up to $J'/J = 0.5$ (see Fig. 4).

Besides, three types of residual couplings have been identified, all of the same order of magnitude (a few times $J/100$) [23]: in-plane Dyzaloshinskii-Moriya interactions (both inter- and intradimer), further neighbor in-plane exchange couplings, and interplane exchange couplings. Being of the same order as the width of the smallest plateaux at $1/9$ and $2/15$, these couplings are clearly strong enough to affect them, and possibly to replace them by other plateaux, for instance at $1/8$. However, since it is very wide, we expect the plateau at $1/6$ to be robust against the introduction of so small residual couplings, and the presence of a $1/6$ plateau in $SrCu_2(BO_3)_2$ must be regarded as a robust prediction of our theory.

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[1] For a review, see A. Honecker, J. Schulenburg, and J. Richter, J. Phys. Condens. Matter **16**, S749 (2004) and references therein.
[2] K. P. Schmidt, J. Dorier, A. Läuchli, and F. Mila, Phys. Rev. Lett. **100**, 090401 (2008).
[3] K. Onizuka *et al.*, J. Phys. Soc. Jpn. **69**, 1016 (2000).
[4] K. Kodama *et al.*, Science **298**, 395 (2002).
[5] B. S. Shastry and B. Sutherland, Physica (Amsterdam) **108B+C**, 1069 (1981).
[6] T. Momoi and K. Totsuka, Phys. Rev. B **62**, 15 067 (2000).
[7] S. Miyahara and K. Ueda, J. Phys. Condens. Matter **15**, R327 (2003).
[8] S. Miyahara and K. Ueda, Phys. Rev. B **61**, 3417 (2000).
[9] G. Misguich, T. Jolicoeur, and S. M. Girvin, Phys. Rev. Lett. **87**, 097203 (2001).
[10] S. Miyahara, F. Becca, and F. Mila, Phys. Rev. B **68**, 024401 (2003).
[11] S. E. Sebastian *et al.*, arXiv:0707.2075v1.
[12] M. Takigawa *et al.*, Phys. Rev. Lett. **101**, 037202 (2008).
[13] F. Levy *et al.*, Europhys. Lett. **81**, 67004 (2008).
[14] C. Knetter and G. S. Uhrig, Eur. Phys. J. B **13**, 209 (2000); C. Knetter, K. P. Schmidt, and G. S. Uhrig, J. Phys. A **36**, 7889 (2003).
[15] K. P. Schmidt and G. S. Uhrig, Phys. Rev. Lett. **90**, 227204 (2003).
[16] C. Knetter, A. Bühler, E. Müller-Hartmann, and G. S. Uhrig, Phys. Rev. Lett. **85**, 3958 (2000); C. Knetter and G. S. Uhrig, Phys. Rev. Lett. **92**, 027204 (2004).
[17] S. Miyahara and K. Ueda, Phys. Rev. Lett. **82**, 3701 (1999).
[18] K. P. Schmidt, J. Dorier, and A. Läuchli, Phys. Rev. Lett. **101**, 150405 (2008).
[19] T. Matsubara and H. Matsuda, Prog. Theor. Phys. **16**, 569 (1956).
[20] In the context of the original model, this requires a spin-Peierls mechanism through a coupling to the lattice, as done in Refs. [4,10].
[21] A. Läuchli, S. Wessel, and M. Sigrist, Phys. Rev. B **66**, 014401 (2002).
[22] Note that working with only one type of triplet might fail beyond this value, but it is certainly valid up to the first level crossing in the ground state.
[23] V. V. Mazurenko *et al.*, Phys. Rev. B **78**, 195110 (2008).