

Pressure dependence of the magnetization plateaus of $\text{SrCu}_2(\text{BO}_3)_2$

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We show that the critical fields of the magnetization plateaus of the Shastry-Sutherland model decrease significantly upon increasing the ratio of inter- to intradimer coupling and accordingly that the magnetization plateaus of $\text{SrCu}_2(\text{BO}_3)_2$ shift to lower field under pressure, making the first two plateaus at $1/8$ and $2/15$ potentially accessible to neutron scattering experiments. These conclusions are based on the derivation of an effective classical model of interacting pinwheel-shaped spin-2 bound states using a combination of perturbative and graph-based continuous unitary transformations, showing that pinwheel crystals are indeed the lowest-energy plateau structures at low magnetization and that a simple model of intermediate-range two-body repulsion between pinwheels is able to account quantitatively for the plateau sequence.

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Introduction. The frustrated quantum magnet $\text{SrCu}_2(\text{BO}_3)_2$ is one of the most important players in the field of highly frustrated quantum magnetism [1] due to its very rich and complex magnetization curve [2–11]. Indeed, experiments in ultrastrong magnetic fields unveil a multitude of intriguing behaviors such as a series of magnetization plateaus archetypical of frustrated quantum magnetism. Despite a huge body of literature over the last 15 years [2–23], a consistent theoretical understanding of the full sequence of plateaus seemingly emerged only recently [23]. The low part of the magnetization curve is most exciting since the magnetization plateaus are predicted to be built from exotic pinwheel-shaped spin-2 bound states [23] and not from individual triplon excitations [24].

Triplons are indeed the natural building block for the magnetization plateaus in $\text{SrCu}_2(\text{BO}_3)_2$, since the underlying microscopic description corresponds remarkably well to the quantum $S = 1/2$ Heisenberg antiferromagnet on the two-dimensional Shastry-Sutherland lattice. The Shastry-Sutherland model [25] can be seen as a set of mutually orthogonal dimers, which are coupled by an interdimer coupling J' . Its beauty arises from the exact ground state in terms of a product state of singlets at zero magnetic field. One natural approach is then to view the magnetization process as populating the dimers by triplets. Even though this approach gives important insights for the plateaus at $1/3$ and $1/2$ [11,19,21], the prediction in terms of crystals of triplons for the plateau sequence at low magnetization disagrees with the experimental one, whereas pinwheels naturally give rise to the experimental sequence $1/8$, $2/15$, and $1/6$ [23].

Neutron scattering of the low-magnetization plateaus $1/8$, $2/15$, and $1/6$ represents a powerful tool to clarify the magnetization structure of these plateaus, but experiments are challenging due to the rather large external magnetic fields needed (27 T for the first plateau at $1/8$). However, external pressure on $\text{SrCu}_2(\text{BO}_3)_2$ is known to reduce the exchange couplings J and J' and to increase the ratio J'/J [26,27], which implies that the critical fields of the plateaus will change with pressure. Consequently, it is of major importance to predict the sequence of plateaus as a function of field and pressure and to calculate the experimental signatures of pinwheel crystals in neutron scattering experiments.

This is exactly the objective of this Rapid Communication. First, we establish an effective low-energy model in terms of interacting pinwheels, which captures the low-magnetization plateaus of the frustrated quantum magnet $\text{SrCu}_2(\text{BO}_3)_2$ quantitatively. The flexibility of our approach allows us to predict the evolution of pinwheel crystals when varying microscopic coupling constants and to show that the critical magnetic fields of the low-magnetization plateaus decrease with pressure, making the pinwheel crystals at $1/8$ and $2/15$ accessible to neutron scattering with experimentally realistic values of field and pressure. In addition, we calculate the local magnetization of these plateaus, which is of direct importance for elastic neutron scattering experiments on $\text{SrCu}_2(\text{BO}_3)_2$.

Model. We study the spin- $1/2$ Shastry-Sutherland model in an external magnetic field h , which reads

$$\mathcal{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J' \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j - h \sum_i S_i^z, \quad (1)$$

with the bonds $\langle i,j \rangle$ building an array of orthogonal dimers and the bonds $\langle\langle i,j \rangle\rangle$ representing interdimer couplings. The geometry of the Shastry-Sutherland model and its two-dimer unit cell, made of a vertical and a horizontal dimer, is illustrated in Fig. 1.

Here we focus on the phase diagram of Eq. (1) for parameter ratios $J'/J \leq 0.675$ for which the pure Shastry-Sutherland model is in the exact product state of singlets for $h = 0$ [22,28], since this is the relevant coupling regime for $\text{SrCu}_2(\text{BO}_3)_2$ [11].

Approach. We apply a continuous unitary transformation (CUT) to map the Shastry-Sutherland model onto an effective low-energy model describing triplons on a square lattice along the lines of Ref. [19]. The essential feature of the derived effective Hamiltonian is the conservation of the total number of triplons. In a finite magnetic field, the relevant triplon states have maximal S_z . Other channels are only important if bound states of triplons with different quantum numbers become relevant at low energies [29]. Furthermore, we concentrate on the plateaus at low densities where pinwheels are expected. Pinwheels correspond to two-triplon bound states with $S = 2$ stabilized by two-triplon correlated hopping processes [14,30]. We therefore restrict the terms in the effective model to

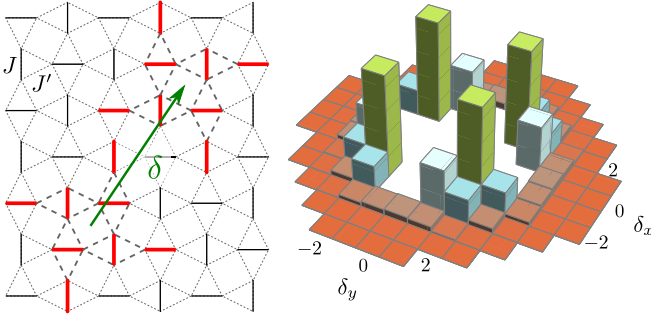


FIG. 1. Left: Illustration of the Shastry-Sutherland model. Solid (dashed) bonds denote intradimer (interdimer) couplings J (J'). Additionally, two pinwheels at distance $\delta = (\delta_x, \delta_y)$ are shown as thicker red bonds. Right: The effective interaction $V_{pw}(\delta)$ between two pinwheels located at sites $(0,0)$ and δ of the effective square lattice formed by the centers of the pinwheels for $J'/J = 0.63$. Note that the inner hardcore potential for small δ is not given (white area).

one- and two-particle operators

$$\begin{aligned} \mathcal{H}_{\text{eff}} = & \mu \sum_i \hat{n}_i + \sum_{i,j} V_{ij} \hat{n}_i \hat{n}_j + \sum'_{i,j,k} t_c^{ijk} (b_i^\dagger b_j \hat{n}_k + \text{H.c.}) \\ & + \sum'_{i,j,k,l} P_{ijkl} b_i^\dagger b_j^\dagger b_k b_l, \end{aligned} \quad (2)$$

where the sums run over the effective square lattice built by the J dimers (see also Fig. 1). The primed sums indicate that all involved sites have to be pairwise different. The hardcore boson operator b_i^\dagger (b_i) creates (annihilates) a triplet $|t^1\rangle$ at site i , and $\hat{n}_i = b_i^\dagger b_i$ is the local density operator with eigenvalues 0 and 1.

In Ref. [19], the coefficients of \mathcal{H}_{eff} have been determined perturbatively up to high orders in J'/J applying perturbative CUTs (pCUTs) [31,32]. This expansion gives a satisfactory effective model up to $J'/J \approx 0.5$. For larger values of J'/J , while extrapolations of the chemical potential μ and of the two-particle interactions V_{ij} still work satisfactorily, no consistent extrapolation of correlated hopping t_c^{ijk} and pair hopping P_{ijkl} processes exists.

As a consequence, we use the following strategy, which is well adapted to the physics of pinwheel crystals: (i) We apply nonperturbative graph-based CUTs (gCUTs) [33,34] to treat the rather local quantum fluctuations like correlated hopping. (ii) We take the longer-range part of the (extrapolated) two-triplon repulsive density-density interactions V_{ij} obtained by pCUTs to quantify the interaction between pinwheels. This is reasonable, since the entanglement between different pinwheels is expected to be low. Let us stress that the *same* quasiparticle generator for the CUT is used in pCUTs and gCUTs, so it is indeed the same CUT which is performed for all couplings of the effective model. The effective Hamiltonian takes then the form $\mathcal{H}_{\text{eff}} = \mathcal{H}_{\text{eff}}^{\text{gCUT}} + \mathcal{H}_{\text{eff}}^{\text{pCUT}}$. The chemical potential μ and (most of) the repulsive interactions V_{ij} are taken from Ref. [19]. All other two-particle processes are deduced by gCUTs and are contained in $\mathcal{H}_{\text{eff}}^{\text{gCUT}}$.

The general idea of gCUTs [33,34] is to perform an exact CUT on topologically distinct graphs. Here we do not

perform a full graph expansion, but we consider the single C_4 -symmetric cluster of 8 dimers as shown in Fig. 1, since this is the smallest cluster on which a pinwheel fits. From the cluster-dependent amplitudes we extract the closest to the thermodynamic limit approximations of the two-triplon amplitudes [35]. In contrast to pCUT calculations where all quantum fluctuations up to a certain order in J'/J are included, the nonperturbative amplitudes contain all processes to infinite order in J'/J fitting on the graphs under consideration. This renders the nonperturbative analysis more powerful in many cases.

The Hamiltonian \mathcal{H}_{eff} represents an interacting hardcore boson model with exotic kinetic terms. It can describe conventional crystals made of single triplons as well as pinwheel crystals. The conventional plateaus of triplons are well treated in the classical limit along the lines of Ref. [19], which amounts to looking for product wave functions of localized triplons. A quantitative description of pinwheels is obtained by the following procedure. We assume that different pinwheels only interact via the diagonal interactions V_{ij} ; i.e., pinwheel crystals are well described by product wave functions of single pinwheels living on the 8-dimer cluster shown in Fig. 1. The centers of pinwheels build again an effective square lattice. We therefore solve the two-particle problem on this cluster using \mathcal{H}_{eff} which gives access to the chemical potential μ_{pw} of a pinwheel as well as its density profile $n_i = \langle \hat{n}_i \rangle$ for all dimers i of a given pinwheel. The latter allows us to determine the effective interaction $V_{pw}(\delta)$ between two pinwheels at distance δ (see Fig. 1 right) by summing up the various two-particle interactions $V_{ij} n_i n_j$ so that i and j correspond to dimers of different pinwheels. One then obtains the following effective pinwheel Hamiltonian

$$\mathcal{H}_{\text{pw}} = \mu_{\text{pw}} \sum_{\mathbf{i}} \hat{n}_{\mathbf{i}}^{\text{pw}} + \sum_{\mathbf{i}, \delta} V_{\text{pw}}(\delta) \hat{n}_{\mathbf{i}}^{\text{pw}} \hat{n}_{\mathbf{i}+\delta}^{\text{pw}}, \quad (3)$$

where \mathbf{i} runs over the sites of the effective square lattice built by the centers of pinwheels and $\hat{n}_{\mathbf{i}}^{\text{pw}}$ is the local density operator of pinwheels with eigenvalues 0 and 1. This procedure can be further optimized by determining the local particle densities n_i inside the pinwheels self-consistently (see Ref. [35]). However, all essential properties discussed below are already present without this self-consistency loop.

The effective model \mathcal{H}_{pw} is essentially classical since it only contains pinwheel density operators. This implies that the treatment of pinwheel crystals becomes extremely simple and transparent. In particular, the energy can be extracted by summing up the relevant pinwheel interactions $V_{pw}(\delta)$. One can also calculate the local magnetizations $m_i = \langle S_i^z \rangle$ of any *spin* site of the Shastry-Sutherland model using the same kind of approximation. To this end we transform the observables S_i^z for each of the 16 spin sites on a single pinwheel cluster by the same CUT and calculate the expectation values with respect to the ground-state solution of the effective model \mathcal{H}_{eff} .

Phase diagram. We have calculated the energy of both types of crystal structures, along the lines of Ref. [19] for the conventional triplon crystals, and by solving the effective pinwheel model of Eq. (3) for the pinwheel crystals. Remarkably, the energy of conventional plateaus is considerably larger than that of the competing pinwheel crystals for $M \leq 1/4$ and

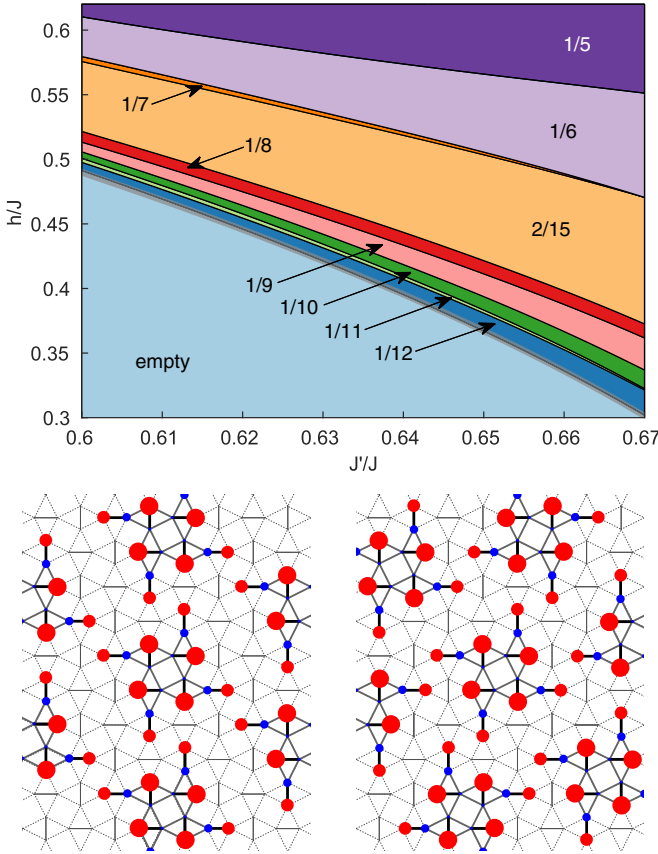


FIG. 2. Upper panel: Phase diagram of the Shastry-Sutherland model as a function of J'/J and magnetic field h/J obtained from the effective pinwheel model \mathcal{H}_{pw} in a self-consistent calculation. Lower panel: Local magnetization of the pinwheel crystals at densities $1/8$ (left) and $2/15$ (right). The area of the red (blue) circles is proportional to the positive (negative) magnetization along the z direction orthogonal to the displayed plane.

all values of J'/J , resulting in the phase diagram displayed in Fig. 2 that contains only pinwheel crystals. The results are in excellent agreement with recent numerical calculations [23,36]. In particular, it contains the experimental sequence $1/8$, $2/15$, and $1/6$, and the arrangements of pinwheels are identical. The same is even true for the tiny domain-wall crystal at density $1/7$, which is located between $2/15$ and $1/6$ [37]. Furthermore, our energies of the various plateaus are in quantitative agreement with those of Ref. [23] for $J'/J = 0.63$, the largest deviation between both calculations being below $10^{-3}J$ and only $\approx 10^{-4}J$ for $1/8$ and $2/15$. Note that our phase diagram also contains lower magnetization plateaus which are not present in experiments, presumably because the pinwheels can delocalize at very low density, a possibility not included in our model, which is aimed at comparing plateau structures among themselves but not with compressible phases.

The effective model \mathcal{H}_{pw} therefore represents a quantitative, light, and physically intuitive description of pinwheel crystals in the Shastry-Sutherland model in a broad range of ratios J'/J including the relevant regime for the frustrated quantum magnet $\text{SrCu}_2(\text{BO}_3)_2$. These properties of \mathcal{H}_{pw} are exploited

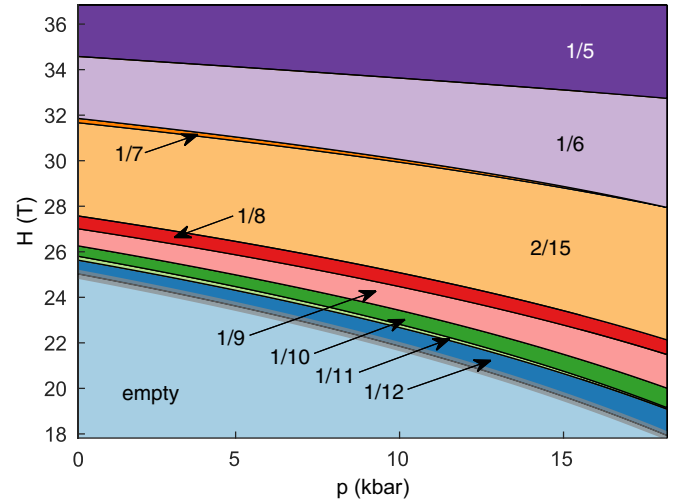


FIG. 3. Phase diagram of $\text{SrCu}_2(\text{BO}_3)_2$ as a function of pressure p and magnetic field H obtained from the effective pinwheel model \mathcal{H}_{pw} .

in the following to give precise predictions in order to identify pinwheel crystals experimentally.

$\text{SrCu}_2(\text{BO}_3)_2$ *under pressure*. One may wonder how to unambiguously identify pinwheel crystals experimentally keeping in mind the rather large critical fields of the compound $\text{SrCu}_2(\text{BO}_3)_2$, e.g., 27 T for the lowest plateau at $1/8$. The best option to pinpoint pinwheel crystals is certainly neutron scattering, which is, however, challenging at these large field values. Here we propose to apply external pressure, turning neutron scattering experiments on the pinwheel crystals of $\text{SrCu}_2(\text{BO}_3)_2$ into a realistic and valuable option.

The effect of pressure p is known from zero-field measurements of the magnetic susceptibility to be twofold in $\text{SrCu}_2(\text{BO}_3)_2$ [26,27]. First, the absolute values of J and J' are reduced. Second, the ratio J'/J is further increased, but one stays in the same zero-field phase as long as $p \leq 17$ kbar, which defines the relevant pressure window. To a good approximation [26,27], the coupling constants J (J') decrease linearly with a slope -0.63 T/kbar (-0.29 T/kbar), enabling us to convert our theoretical phase diagram Fig. 2 as a function of J'/J into one where the critical magnetic fields of the various plateaus is shown as a function of pressure p . We fix the zero-pressure values to $J = 59.4$ T and $J' = 37.4$ T ($J'/J \approx 0.63$) so that we recover the critical field $H_c = 27$ T for the lowest $1/8$ plateau. The resulting phase diagram is displayed in Fig. 3.

As expected, all critical fields for the various plateaus decrease with increasing pressure. Experimentally, neutron scattering can be done up to fields of 26 T with present technology [38]; i.e., one almost reaches the lowest $1/8$ plateau ($H_c = 27$ T at ambient pressure). Interestingly, already a moderate pressure between 5 and 10 kbar is sufficient to push the critical field of the $1/8$ plateau below 26 T. Furthermore, one will also access the $2/15$ plateau when applying a pressure of 10 kbar. These results therefore clearly demonstrate that neutron scattering at realistic values of the magnetic field and at moderate pressure is a promising option to determine the structure of the lowest two magnetization plateaus of

the frustrated quantum magnet $\text{SrCu}_2(\text{BO}_3)_2$, and to identify pinwheels as the elementary building blocks of these exotic states of quantum matter. Indeed, the Bragg peaks will give access to the unit cells (which are specific to pinwheel crystals as opposed to triplon crystals), and the static structure factor to important information on the local magnetization inside the unit cell (see Supplementary Materials [35] for specific data).

Finally, it is known from NMR measurements that the widths of the $1/8$ and $2/15$ plateaus are similar and of the order 1 T and that an incommensurate phase exists between the plateaus at $2/15$ and $1/6$ in which the translational symmetry is still broken while the magnetization raises monotonically with field [10]. If one assumes that the width of the $2/15$ plateau remains small when applying moderate pressure, our results also predict that this phase should be accessible by future neutron scattering experiments.

Discussion. We have derived an effective low-energy model directly in terms of pinwheels. The quantitative agreement in the experimentally relevant coupling regime $J'/J = 0.63$ between our results and the numerical ones using tensor network calculations [23] implies that pinwheel crystals can be very well understood as product-wave functions where the elementary building blocks are individual pinwheels living on eight dimers of the Shastry-Sutherland lattice. Each pinwheel corresponds microscopically to a two-triplon bound state stabilized by correlated hopping processes.

The effective pinwheel model \mathcal{H}_{pw} is purely classical since only the location of pinwheels matters. In fact, one can rewrite this effective Hamiltonian exactly as an antiferromagnetic Ising model on the square lattice with peculiar types of geometrically frustrated Ising interactions quantified by $V_{\text{pw}}(\delta)$. In this picture, it is the competition between these frustrated interactions that leads to a sequence of pinwheel crystals that includes the experimentally relevant ones at $1/8$, $2/15$, and

$1/6$. Let us remark that the $1/5$ plateau is expected to be unstable under the inclusion of DM interactions [23].

In addition, this effective pinwheel model potentially offers a natural explanation for the incommensurate regime between the plateaus at $2/15$ and $1/6$ reported in NMR experiments in terms of a devil's staircase and the associated proliferation of domain walls, a phenomenon well identified in the phase diagram of frustrated Ising models [39,40]. Unfortunately, we have found no devil's staircase in the effective model \mathcal{H}_{pw} . The stabilization of such a devil's staircase might require additional ingredients such as DM interactions, known to be present in $\text{SrCu}_2(\text{BO}_3)_2$, or longer-range pinwheel interactions. This is an interesting subject for future investigation. Note that the understanding of this incommensurate regime is also essential in order to determine the correct width of the $2/15$ plateau, which is too large in all theoretical treatments.

To summarize, we have found a physically intuitive and quantitative description of pinwheel crystals in the frustrated quantum magnet $\text{SrCu}_2(\text{BO}_3)_2$. This has allowed us to come up with detailed predictions for future experiments like the evolution of pinwheel crystals under pressure. In future investigations it would be interesting to see whether one can also calculate the dynamical structure factors of pinwheel crystals and whether superfluid or supersolid phases can also be addressed in our framework by melting the observed pinwheel crystals.

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