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2018

Wierschem, K., Zhang, Z., Wibawa, A., & Sengupta, P. (2018). Magnetization plateaus and supersolid phases in an extended Shastry–Sutherland model. *European Physical Journal B*, 91(9), 201-. doi:10.1140/epjb/e2018-90356-5

<https://hdl.handle.net/10356/137393>

<https://doi.org/10.1140/epjb/e2018-90356-5>

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Magnetization plateaus and supersolid phases in an extended Shastry-Sutherland model

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Obtaining strong magnetoelectric couplings in bulk materials and heterostructures is an ongoing challenge. We demonstrate that manganite heterostructures of the form (Insulator)/(LaMnO₃)_n/Interface/(CaMnO₃)_n/(Insulator) show strong multiferroicity in magnetic manganites where ferroelectric polarization is realized by charges leaking from LaMnO₃ to CaMnO₃ due to repulsion. Here, an effective nearest-neighbor electron-electron (electron-hole) repulsion (attraction) is generated by cooperative electron-phonon interaction. Double exchange, when a particle virtually hops to its unoccupied neighboring site and back, produces magnetic polarons that polarize antiferromagnetic regions. Thus a striking giant magnetoelectric effect ensues when an external electrical field enhances the electron leakage across the interface.

I. INTRODUCTION

The search for complex quantum many body states with coexisting long range orders that break different symmetries in strongly correlated systems have been at the center of Condensed Matter research for the past several decades. Frustrated quantum magnets have long played a major role in the discovery of novel quantum states as well as for realising magnetic analogues of many body bosonic states in a more controllable manner. The interplay between competing interactions, geometric frustration, and external magnetic fields result in the emergence of a rich variety of magnetic states. Magnetic systems have the advantage that the magnetic field, which plays role of the chemical potential, can be varied continuously over a large range of values.

The supersolid (SS) is a unique state of matter that possesses simultaneous diagonal and off-diagonal long range order. Ever since Penrose and Onsager's speculation on the possibility of such co-existence,¹ the supersolid has been the subject of intense theoretical and experimental investigation.²⁻⁴ However, a conclusive experimental realization of this novel state of matter remains elusive to this date. While the supersolid phase of helium remains controversial, Quantum magnets provide an alternate route via the spin analog of the supersolid phase. Identification of realistic systems with supersolid order is a valuable step towards the ultimate realization of this elusive phase.

In this work, we demonstrate the emergence of the spin supersolid phase and magnetization plateaus in an anisotropic $S = 1/2$ XXZ model with longer range interactions on the geometrically frustrated Shastry-Sutherland lattice (SSL).⁵ The SSL provides a versatile framework to study the emergence of frustration driven complex magnetic phases.⁶⁻¹² The existence of several quasi-2D quantum magnets – including the extensively studied $SrCu_2(BO_3)_2$ as well as a completely family of rare earth tetraborides, RB_4 (R=Tm, Er, Dy, Ho, Eb)¹³⁻¹⁵ and 221 compounds such as Ce_2Pt_2Pb , Yb_2Pt_2Pb and Ce_2Mg_2Pb – where the magnetic ions are arranged in a SSL configuration ensures that a wide

range of parameter regimes (such as varying degrees of frustration, strength and nature of interactions) can be explored experimentally, complementing any theoretical studies. Magnetization plateaus are ubiquitous in many frustrated quantum magnets. Several studies have demonstrated that a spin SS ground state is stabilized in the vicinity of magnetization plateaus, including our own work on the SSL. In this work, we explore a wide range of parameter regimes with varying degrees of frustration as well as the nature and strength of longer range interactions.

II. MODEL AND BACKGROUND

We consider a spin-1/2 XXZ model defined by the Hamiltonian

$$\mathcal{H} = \sum_{\langle ij \rangle} -|J_{ij}\Delta| (S_i^x S_j^x + S_i^y S_j^y) + J_{ij} S_i^z S_j^z - h_z \sum_i S_i^z, \quad (1)$$

with interacting pairs $\langle ij \rangle$ defined by the Shastry-Sutherland lattice with extended long range interactions as depicted in Fig. 1(a). The Hamiltonian is parameterized by the bond strengths $J_{ij} \in \{J_1, J_2, J_3, J_4\}$, the exchange anisotropy Δ , and the applied magnetic field h_z . Note that the transverse coupling is taken to be strictly ferromagnetic and the applied magnetic field is longitudinal (i.e. parallel to the axis of exchange anisotropy). These are important features for the quantum Monte Carlo methods we will employ, as discussed in the next section.

Several versions of the above model have been studied previously¹⁶⁻²³. These began with initial studies of the Ising model on the bare Shastry-Sutherland lattice. It was quickly recognized that the ground state magnetization process in this case includes a single plateau at $m/m_s = 1/3$ only¹⁶⁻¹⁸. Incorporating quantum fluctuations via the XXZ model, a narrow plateau at $m/m_s = 1/2$ can also be stabilized^{16,17}. However, to fully stabilize the $m/m_s = 1/2$ plateau, extended interactions beyond the bare Shastry-Sutherland lattice are required, both for the Ising model^{21,22} and for the XXZ model^{19,20,23,24}.

FIG. 1. (Color online) (a) Unit cell of the Shastry-Sutherland lattice with extended long range interactions. (b) Schematic depiction of J_3 - J_4 parameter space with strongly frustrated regime (quadrant I) in red and weakly frustrated regime (quadrants II, III, and IV) in blue.

It is worth mentioning that long range interactions can also lead to novel plateau structures. For example, Huang *et al.*²⁵ have considered the Ising model on the Shastry-Sutherland lattice with added dipole-dipole interactions and find the appearance of a $m/m_s = 1/2$ plateau. Another way to incorporate long range interactions is through the Kondo coupling between localized moments and itinerant electrons. Such an interaction is known to lead to long range oscillating interactions i.e. the RKKY interactions. Farkašovský *et al.*²⁶ considered a simplified Kondo lattice model on the Shastry-Sutherland lattice with local Ising moments to find plateaus at $m = m_s/2$ and $m = m_s/3$ as well as narrow plateaus at $m/m_s = 1/5$ and $m/m_s = 1/7$. Finally, direct consideration of the RKKY interactions can lead to stabilization of the $m/m_s = 1/2$ plateau for suitably chosen values of the Fermi momentum²⁷.

III. METHODS AND OBSERVABLES

A. SSE Quantum Monte Carlo

Our primary method is the stochastic series expansion (SSE) formulation of quantum Monte Carlo²⁸. We employ directed loop updates²⁹ as well as the plaquette based vertex expansion^{30,31}. Ground state properties in the thermodynamic limit are accessed via finite size scaling of the inverse temperature β and system size L . Within SSE correlations such as $\langle S_i^x S_j^x + S_i^y S_j^y \rangle$ and $\langle S_i^z S_j^z \rangle$ are easily obtained^{28,32}, and their Fourier transforms define corresponding static structure factors $S^{xy}()$ and $S^{zz}(k)$ that measure transverse and longitudinal order, respectively. In the equivalent system of hard-core bosons, these map onto off-diagonal and diagonal ordering, respectively, and the simultaneous presence of both types of order defines a supersolid phase.

The spin stiffness ρ_s is defined as the response to

a twist in the boundary conditions³³. In simulations that sample multiple winding number sectors, as in the present implementation of the directed loop algorithm, ρ_s is simply related to the winding number of the world lines. For square simulation cells in two dimensions, $\rho_s = (w_x^2 + w_y^2)/2\beta$, where w_x and w_y are the winding numbers in the x and y directions³⁴.

B. Explicit construction of ground states in the Ising limit

We also employ direct calculation of ground state energies for spin configurations in the Ising limit. While in principle these calculations are exact, in practice it can be difficult to construct all of the appropriate ground state spin configurations — especially in the presence of multiple competing interactions. For this reason, we have carefully compared our results for the Ising limit phase diagram to results for the XXZ model. These are expected to be qualitatively alike in the vicinity of the Ising limit. The construction of explicit ground states in the Ising limit provides a helpful insight into the nature of the spin configurations in the complex ground state phases arising in the Hamiltonian (1) for Ising-like anisotropy.

To include the effects of weak quantum fluctuations ($|\Delta| \ll 1$), we have applied second order perturbation theory to find the leading order quantum corrections to the Ising ground states. In some cases, this can lead to exact results. For example, the saturation field h_{sat} can often be found as follows. Starting from the fully polarized ground state at large magnetic field, calculate the energy cost to flip a single spin. This energy cost will be zero at $h_z = h_{sat}$ for the case of antiferromagnetic longitudinal spin exchanges. This leads to the expression

$$h_{sat} = (2J_1 + J_2/2 + J_3 + 2J_4)(1 + \Delta), \quad (2)$$

which is exact in the antiferromagnetic parameter regimes of $\mathcal{H}$. In fact, if one or two parameters are weakly ferromagnetic, this formula often holds true by simply replacing $J_\alpha(1 + \Delta)$ by $J_\alpha + |J_\alpha\Delta|$ for the FM interactions. Note that the uniform magnetization per site $m = \sum_j \langle S_j^z \rangle / N$ saturates at $m_s = 1/2$.

C. Spiral Plaquette Representation

To further gain insight into the structure of the magnetization plateaus and the mechanism behind their emergence, we have developed a spiral plaquette representation to describe these states. Generally, plateaus are assumed to have crystal structures with various unit cells. Each unit cell could consist of a certain number of translationally invariant smaller cells of the lattice. One of such translationally invariant cell is the spiral plaquette as shown in Figure 2. The spiral plaquette (splqt) con-

FIG. 2. The spiral plaquette contains one central square and four spirally surrounding dimers. The position of the plaquette is denoted at the center of the square.

sists of four dimers surrounding a central square. Each dimer could be represented by the following four states

$$|u\rangle = |\uparrow\uparrow\rangle, |d\rangle = |\downarrow\downarrow\rangle, |l\rangle = |\uparrow\downarrow\rangle \text{ and } |r\rangle = |\downarrow\uparrow\rangle, \quad (3)$$

where the first spin is located at the corner of the central square and we define their dual states:

$$|\bar{u}\rangle = |u\rangle, |\bar{d}\rangle = |d\rangle, |\bar{l}\rangle = |r\rangle \text{ and } |\bar{r}\rangle = |l\rangle. \quad (4)$$

The splqt is represented by the direct product of the four dimer states:

$$|sp_{\mathbf{r}}\rangle = |a_1, a_2, a_3, a_4\rangle_{\mathbf{r}} \quad (5)$$

where $|a_i\rangle \in \{|u\rangle, |d\rangle, |l\rangle, |r\rangle\}$ and $\mathbf{r}$ denotes the position of the splqt. Because of the over-completeness, the splqt states obey the following constraints:

$$|a_3\rangle_{\mathbf{r}} = |\bar{a}_1\rangle_{\mathbf{r}+\mathbf{x}} \quad \text{and} \quad |a_4\rangle_{\mathbf{r}} = |\bar{a}_2\rangle_{\mathbf{r}+\mathbf{y}}. \quad (6)$$

We introduce Schwinger bosons to represent the four dimers in the splqt:

$$\hat{u}^\dagger|\emptyset\rangle = |u\rangle, \hat{d}^\dagger|\emptyset\rangle = |d\rangle, \hat{l}^\dagger|\emptyset\rangle = |l\rangle \text{ and } \hat{r}^\dagger|\emptyset\rangle = |r\rangle, \quad (7)$$

where $|\emptyset\rangle$ is the vacuum state. The spin operators at the n^{th} dimers can be represented as:

$$\begin{aligned} \hat{S}_{n,1}^z &= \frac{1}{2}(\hat{L}_n - \hat{R}_n + \hat{U}_n - \hat{D}_n), \\ \hat{S}_{n,2}^z &= \frac{1}{2}(-\hat{L}_n + \hat{R}_n + \hat{U}_n - \hat{D}_n). \end{aligned} \quad (8)$$

The subscript 1 (2) denotes the site at the corner of the central square (at the edge). $\hat{L}_n = \hat{l}_n^\dagger \hat{l}_n$ is the number operator and similar for the other three. After some algebra, the Ising Hamiltonian can be expressed in a diagonal form as:

$$\begin{aligned} \hat{H}_{Ising} &= \sum_{i,n} \left(\frac{1}{8}J_2 - \frac{1}{2}h \right) \hat{U}_{n,i} + \left(\frac{1}{8}J_2 + \frac{1}{2}h \right) \hat{D}_{n,i} - \frac{1}{8}J_2(\hat{L}_{n,i} + \hat{R}_{n,i}) \\ &+ \frac{1}{2}J_1 \sum_{i,n} (\hat{U}_{n,i} - \hat{D}_{n,i})(\hat{U}_{n+1,i} + \hat{L}_{n+1,i} - \hat{D}_{n+1,i} - \hat{R}_{n+1,i}) \\ &+ \frac{1}{4}(2J_4 + J_3) \sum_{\langle i,j \rangle} (\hat{U}_{1,i} - \hat{D}_{1,i})(\hat{U}_{1,j} - \hat{D}_{1,j}) + (\hat{U}_{2,i} - \hat{D}_{2,i})(\hat{U}_{2,j} - \hat{D}_{2,j}) \\ &+ \frac{1}{4}(2J_4 - J_3) \sum_{\langle i,j \rangle} (\hat{L}_{1,i} - \hat{R}_{1,i})(\hat{L}_{1,j} - \hat{R}_{1,j}) + (\hat{L}_{2,i} - \hat{R}_{2,i})(\hat{L}_{2,j} - \hat{R}_{2,j}) \\ &+ \frac{1}{4}J_3 \sum_{\langle i,j \rangle} (\hat{L}_{1,i} - \hat{R}_{1,i})(\hat{U}_{1,j} - \hat{D}_{1,j}) - (\hat{U}_{1,i} - \hat{D}_{1,i})(\hat{L}_{1,j} - \hat{R}_{1,j}) \\ &+ \frac{1}{4}J_3 \sum_{\langle i,j \rangle} \eta_{ij}(\hat{U}_{2,i} - \hat{D}_{2,i})(\hat{L}_{2,j} - \hat{R}_{2,j}) - \eta_{ij}(\hat{L}_{2,i} - \hat{R}_{2,i})(\hat{U}_{2,j} - \hat{D}_{2,j}) \end{aligned} \quad (9)$$

where $\langle i, j \rangle$ means nearest splqt and $\eta_{ij} = 1$ if the bond is along $\mathbf{x}$ direction and -1 if along $\mathbf{y}$ direction.

The number of splqt in a unit cell N is closely related to the magnetization q/p :

$$\frac{q}{p} = \frac{1}{4N} \sum_{i=1}^N S_i^z \quad (10)$$

where $|S_i^z| \leq 4$ is the magnetization of the i^{th} splqt. q and p are coprime integers. This relation restricts the possible number of splqt in a unit cell as $4N/p$ has to be an integer. For example, for the $1/3$ plateau, $(p, q) = (1, 3)$; therefore N should be a multiple of 3. The existence of possible magnetization plateaus in different parameter ranges could be inferred from the Ising Hamiltonian (9) with the constraints (6).

We only consider antiferromagnetic (AFM) J_1 and J_2 . In the strong ferromagnetic (FM) J_3 and J_4 range, the third quadrant, the third line of the Ising Hamiltonian (9) dominates. Identical adjacent splqt formed by u or d type dimers are preferred, which suggests singlet splqt unit cell. And in the strong FM J_4 and AFM J_3 range, the fourth quadrant, the fourth line of the Ising Hamiltonian dominates and it also implies identical adjacent splqt formed by l or r type dimers. On the other hand, in the strong AFM J_4 and FM J_3 range, the second quadrant, the fourth line of the Ising Hamiltonian dominates and prefers unit cell with two splqt of configuration $\begin{smallmatrix} A & B \\ B & A \end{smallmatrix}$ formed by l and r types dimers. In the first quadrant, the extremely frustrated regime, complicated large unit cells are possible.

IV. ZERO-FIELD PHASE DIAGRAM IN ISING LIMIT

There are four possible zero-field magnetically ordered ground states of $\mathcal{H}$ in the Ising limit as shown in Fig. 3. In addition to these states there is a non-magnetic dimer product state. This exponentially degenerate state is composed of the direct products of dimers on the J_2 bonds. The limits of stability for these five Ising ground states are obtained as follows.

For $J_3 = J_4 = 0$ we have an Ising antiferromagnetic ground state for $J_1 > J_2/2$, otherwise the ground state is the exponentially degenerate Ising dimer state. For non-zero J_3 and / or J_4 , this degeneracy is generically lifted (except along portions of the line $J_3 = 2J_4$). New ground states are stabilized as follows. For large AFM J_3 and FM (or weak AFM) J_4 the columnar state with $(\pi, 0)$ ordering is stabilized. Conversely, the “ J_3 Chessboard” state, with all four spins up / down on alternate J_3 plaquettes, is stabilized for large AFM J_4 and FM (or weak AFM) J_3 . When both J_3 and J_4 are strongly AFM, the “ J_2 Chessboard” state of alternately polarized J_2 plaquettes commences. Finally, the Néel state dominates the region where both J_3 and J_4 are FM.

The constraint for the Ising dimer state in the absence of longer range interactions, $J_1 \leq J_2/2$, is modified in the presence of non-zero J_3 and J_4 . The requirements instead become (i) $J_3 = 2J_4$ (ii) $J_1 \leq J_2/2 + J_3$ and (iii) $J_3 \leq J_2/2$. When weak quantum fluctuations are taken into account $0 < \Delta \ll 1$ the stability of the quantum Ising dimer expands to a finite region of the J_3 – J_4 plane due to the formation of spin-zero triplets on each J_2 bond.

In Fig. 4 we show the ground state Ising phase diagram

FIG. 3. Magnetically ordered zero-field Ising ground states: (a) Néel antiferromagnet, (b) Columnar antiferromagnet, (c) J_2 Chessboard antiferromagnet, and (d) J_3 Chessboard antiferromagnet. Not shown is the exponentially degenerate non-magnetic dimer product state in which each J_2 bond consists of one up and one down spin.

in the J_3 – J_4 plane for $J_1 = J_2$ and $5J_1 = 2J_2$. The qualitative features of these two phase diagrams are preserved for any $J_1 \geq J_2/2$ and $J_1 < J_2/2$, respectively. The main effect of increasing/decreasing J_1 relative to J_2 is to enhance/diminish the extent of the Néel state, respectively. Note, however, that for $J_1 > J_2$ a phase boundary develops between the Néel state and the J_2 Chessboard state, similar to the Columnar to J_3 Chessboard phase boundary for $J_1 < J_2$.

V. WEAKLY FRUSTRATED REGIME

When J_3 or J_4 (or both) are ferromagnetic, the system is in the weakly frustrated regime. In the limit that the ferromagnetic terms dominate the system approaches a simple ferromagnet. We first consider this weakly frustrated regime, corresponding to quadrants II, III, and IV of Fig. 1(b). We will demonstrate how $J_3 < 0$ and $J_4 < 0$ stabilize $1/2$ plateaus in this regime.

By the construction of exact Ising ground states and the use of the splqt representation the spin configurations for the ground state phase are identified for the plateaus at $m/m_s = 1/3$, $1/2$ and $5/9$. By comparison of their energies at a fixed magnetization, including phase separated mixtures of states, we construct the phase diagram at $m/m_s = 1/3$ and $m/m_s = 1/2$ in the J_3 – J_4 plane for $J_1 = J_2$ (see Fig. 5). At $m/m_s = 1/3$ it is clear that either FM J_3 or J_4 will act to destabilize the third plateau states. In contrast, the half plateau states at $m/m_s = 1/2$ are only destabilized by J_3 and J_4 both FM or AFM. Thus there are significant regions of the J_3 – J_4 plane that display a 0 – $\frac{1}{2}$ – 1 magnetization sequence, all of which contain at least one FM interaction (i.e. they

FIG. 4. Ground state Ising phase diagram in the J_3 - J_4 plane for $J_1 = J_2$ (upper panel) and $5J_1 = 2J_2$ (lower panel). In both cases we set $\Delta = 0$ and $J_2 = 1$. In the lower panel, we sketch the extent of the quantum dimer state (dashed blue line boundary) for $\Delta = 0.1$ up to first order in Δ .

lie within the weakly frustrated regime considered in the present section). The plateauless 0-1 magnetization sequence occurs for J_3 and J_4 both strongly FM. At small values of J_3 there is a $0-\frac{1}{3}-\frac{1}{2}-1$ magnetization sequence, as long as $J_4 > -1/4$. In the strongly frustrated regime considered in the next section, we shall see two additional Ising limit magnetization sequences that include a plateau at $m/m_s = 5/9$ which appears whenever all four interactions are AFM. They are the $0-\frac{1}{3}-\frac{1}{2}-\frac{5}{9}-1$ and $0-\frac{1}{3}-\frac{5}{9}-1$ magnetization sequences.

The Ising structures of the third plateau are shown in Fig. 6 and those of the one-half and five-ninths plateaus are shown in Fig. 7.

Finally, we show the possible magnetization sequences for $J_1 = J_2$ in the J_3 - J_4 plane in Fig. 8.

A. Supersolids near 1/2 plateau

In Figs. 9 and 10 we show the appearance of supersolid phases just below the 1/2 plateau in the Columnar (AFM J_3) and J3 Chessboard (AFM J_4) regimes, re-

FIG. 5. Ground state Ising phase diagram in the J_3 - J_4 plane for $J_1 = J_2 = 1$ at $m/m_s = 1/3$ (upper panel) and $m/m_s = 1/2$ (lower panel). In both cases $\Delta = 0$ and the phase separated regions indicate plateau instability.

FIG. 6. The four allowed Third Plateaus (TP) at $m/m_s = 1/3$ when $J_1 = J_2$.

FIG. 7. (a), (b) and (c) The three allowed Half Plateaus (HP) at $m/m_s = 1/2$ when $J_1 = J_2$. (d) Five-ninths Plateau at $m/m_s = 5/9$.

FIG. 8. Ising ground state magnetization sequences in the J_3 - J_4 plane for $J_1 = J_2$. The parameter sets that capture the magnetic properties of ErB_4 and TmB_4 are marked, as well as the location of two emergent supersolid phases (for non-zero Δ) considered later in the text.

spectively. We refer to these as SS1 and SS2, and their locations on the J_3 - J_4 plane are shown in Fig. 8. The zero field ground states exhibit long range magnetic ordering – columnar AFM order for $J_3 > 0, J_4 < 0$ and chessboard AFM for $J_3 < 0, J_4 > 0$ – with a finite gap to lowest magnetic excitations. This is indicated by finite m_c^2 (m_b^2) for the former (latter) situation and vanishing spin stiffness for both. The ground state remains unchanged at weak applied fields, but at a critical field of $h_z \sim 1.2$, the magnetic ordering is quenched via a discontinuous transition and the system enters a gapless spin superfluid phase. The AFM order parameters vanish indicating the suppression of AFM ordering, while the spin

FIG. 9. (Color online) Evidence for the supersolid phase SS1 with columnar magnetic order below the half plateau in the Columnar regime of the model when $J_1 = J_2$.

stiffness acquires a finite value. The uniform magnetization jumps to a finite value ($m_z \approx 0.2$) and increases monotonically with increasing field. At a field strength of $h_z \sim 2$, there is a second transition whereby long range AFM magnetic sets in. Unlike the low field AFM order, the ground state in this field range develops a staggered (π, π) AFM order. This is true for both the parameter regimes under consideration. However, the spin stiffness does not vanish for $2 \lesssim h_z \lesssim 2.1$, indicating a simultaneous spin superfluid ordering. Thus, for this range of applied field, the ground state has co-existing long range diagonal (staggered AFM) and off-diagonal (spin superfluid) ordering – in other words, the ground state is a spin supersolid. In both cases, the corresponding FM parameter acts to stabilize the solid ordering, while finite exchange Δ enhances superfluid ordering. One intriguing difference between SS1 and SS2 is that while they both emerge from superfluid phases as a half plateau is approached from below, in SS1 this occurs via a discontinuous phase transition while in SS2 this occurs as a continuous phase transition. It is tempting to speculate that this difference is related to the fact that SS1 is adiabatically connected to the supersolid phase of the ErB_4 parameter set, which has been shown to occur within an unfrustrated sublattice in the Ising limit²³.

VI. STRONGLY FRUSTRATED REGIME

In this section we focus on the complex magnetic phase diagram that emerges in the strongly frustrated regime, where $J_i > 0 \forall i$. Although we have tried to capture all of the relevant plateaus using ground state energy comparison of diverse Ising structures, this method may miss some complicated plateau structures with large unit cells. Additionally, new plateaus may emerge when quantum

FIG. 10. (Color online) Evidence for the supersolid phase SS2 with chessboard magnetic order below the half plateau in the J3 Chessboard regime of the model when $J_1 = J_2$.

fluctuations $\Delta > 0$ are added to this highly frustrated regime via the quantum order by disorder mechanism. The situation at finite temperature becomes even more complicated. The competition between AFM nearest and next-nearest neighbor interactions (in particular, J_1 and J_4) is similar to that found in the axial next nearest neighbor Ising (ANNNI) model³⁵. As such, a continuum of modulated phases can be expected to exist at intermediate temperatures.

For the above reasons, we have mapped out the ground state magnetic phase diagram in this regime via extensive quantum Monte Carlo simulations, in the two regimes of the bare Shastry-Sutherland interactions, namely the Neel regime $J_1 > 2J_2$ and the dimer regime $J_1 < 2J_2$. The plateau sequence is found to be much more complex when $J_1 < 2J_2$. The magnetic phase diagram in the $h_z - J_3$ parameter space for $\Delta = 0.1$ and $J_4 = 0.2$ is shown in Fig. 11 for a representative values of J_1 and J_2 . However, some of these putative plateaus may be metastable states and more in-depth investigation is needed to confirm their existence in the thermodynamic limit. In this study, we focus on a few stable and prominent plateaus.

5/9 plateau: One interesting feature to be found in Fig. 11 is the broad stability of the 5/9 plateau for $J_3 > 0$, replacing the 1/2 plateau found when $J_3 < 0$.

This new plateau is ubiquitous throughout the strongly frustrated region. The appearance of this plateau can be explained by the following argument. The largest magnetic plateau should simultaneously maximize the uniform magnetization (in order to gain energy from the coupling to magnetic field) and minimize the number of frustrated bonds. Starting from the fully polarized phase, all bonds are frustrated, yet the uniform magnetization is globally maximized. From here, if we start to flip spins in an effort to minimize frustrated bonds, the best we

FIG. 11. (Color online) Magnetic phase diagram in the $h_z - J_3$ plane for $5J_1 = 2J_2$. We set $\Delta = 0.1$ and $J_4 = 0.2$ at $\beta = 32$ using a 18×18 simulation cell.

can do turns out to be forming the 5/9 plateau. In this case, each flipped spin now lies on unfrustrated bonds, and any additional spin flip will actually create more frustrated bonds by forcing at least two flipped spins to correlate ferromagnetically (remember that we are in the regime where all interactions are antiferromagnetic, and therefore ferromagnetic correlations increase the energy). Hence we arrive at the conclusion that starting at the fully polarized state and lowering the magnetic field, we must always cross a finite regime of 5/9 plateau.

In constructing our model Hamiltonian $\mathcal{H}$ in Eq. 1, we have neglected the second cross diagonal term on J_2 plaquettes. Labeling this term as J'_2 , it is clear that the 5/9 plateau is only possible when $J'_2 \leq 0$. However, for AFM values $J'_2 > 0$ we can still apply the same argument as above, only now it will lead to a 3/5 plateau. Such a distinction is important when applying $\mathcal{H}$ as an effective model of rare earth tetraboride magnetism.

5/9 supersolid: Looking at the 5/9 plateau in Fig. 11, there appears to be a related supersolid phase for magnetizations just below this plateau. This putative supersolid phase exists in the intervening field range between the 1/3 and 5/9 plateaus. The 1/3 and 5/9 plateaus do not share the same ordering wave vectors. It is likely that the mechanism is similar to that discussed in the previous section. The magnetic order at the 1/3 plateau melts at a critical field, to be replaced by a spin superfluid phase. At a higher field, the magnetic ordering corresponding to the 5/9 plateau sets in, without quenching the superfluid order completely. The two orders co-exist for a finite range of fields beyond which the offdiagonal order is suppressed and the 5/9 plateau is stabilised. We are currently investigating this putative supersolid phase.

2/9 plateau: This plateau was previously observed in $\text{SrCu}_2(\text{B})_3$ and found theoretically in the canonical

Shastry-Sutherland model¹². Later it was found in the extended Shastry-Sutherland model by Suzuki *et al.*²⁰ in the field range $0.6 < h_z < 0.74$ for parameters $\Delta = 0.2$, $2J_1 = J_2$, $J_3 = 0$ and $J_1 = \sqrt{2}^3 J_4$. They found a magnetic structure similar to that of the 5/9 plateau, but with additional blocks of spins that form plaquette singlet states. In the parameter ranges that we have considered in this study, a weak 2/9 plateau emerges for $J_1 < 2J_2$, whereas it is absent in the dimer regime ($J_1 < 2J_2$).

1/9 plateau: This plateau appears for a narrow window of parameter values, as seen in the upper panel of Fig. 11. Due to the finite size of our simulation cell, however, this phase may or may not be metastable. This relatively small plateau is of importance in the understanding of similar small plateaus in the rare earth tetraboride TmB_4 ¹⁵.

VII. CONCLUSION

We have investigated the emergence of field induced magnetization plateaus and spin supersolid phase in an extended Shastry Sutherland model with longer range interactions and Ising-like exchange anisotropy. The model is motivated by its relevance to understanding the magnetic behavior in rare earth tetraborides. Our results demonstrate that the sequence of field induced plateaus depends strongly on the nature and strength of the longer range interactions, and that multiple spin supersolid phases are stabilized in different parameter

regimes. By explicitly constructing the spin configurations at the plateaus in the Ising limit, we are able to gain insight into the nature of the different supersolid phases and the underlying mechanism behind their emergence. One interesting aspect of highly frustrated XXZ models in the Ising limit that we did not touch on in this work is that in some cases spin liquid phases can become stabilized. This is the case, for example, in the $J_1 - J_2 - J_3$ kagome lattice XXZ model in the Ising limit³⁶. Similar spin liquid phases have also been proposed for Ising limit XXZ models on the $J_1 - J_2$ square lattice and the $J_1 - J_2 - J_3$ honeycomb lattice^{37,38}.

VIII. ACKNOWLEDGMENTS

It is a pleasure to thank Ernest Siang for helpful discussions and help in preparing the figures. Financial support for the work was provided by the Ministry of Education, Singapore, through grant MOE2014-T2-2-112. This research used resources of the National Energy Research Scientific Computing Center, which is supported by the Office of Science of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

IX. AUTHORS CONTRIBUTIONS

All the authors were involved in the preparation of the manuscript. All the authors have read and approved the final manuscript.

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