Exact Demonstration of Magnetization Plateaus and First Order Dimer-Néel Phase Transitions in a Modified Shastry-Sutherland Model for SrCu$_2$(BO$_3$)$_2$

Erwin Müller-Hartmann and Rajiv R. P. Singh

Institute for Theoretical Physics, University of California, Santa Barbara, CA 93106

Christian Knetter and Götz S. Uhrig

Institut für Theoretische Physik, Universität zu Köln, Zülpicher Str. 77, D-50937 Köln, Germany

We study a generalized Shastry-Sutherland model for the material SrCu$_2$(BO$_3$)$_2$. Along a line in the parameter space, we can show rigorously that the model has a first order phase transition between Dimerized and Néel-ordered ground states. Furthermore, when a magnetic field is applied in the Dimerized phase, magnetization plateaus develop at commensurate values of the magnetization. We also discuss various aspects of the phase diagram and properties of this model away from this exactly soluble line, which include gap-closing continuous transitions between Dimerized and magnetically ordered phases.

In recent years, many novel magnetic materials have been synthesized which exhibit spin-gap behavior. In these materials the ground state is a spin-singlet and there is a gap to all spin excitations. Such phenomena have long been studied in quasi-one dimensional systems, but much recent interest has arisen from the discovery of quasi-two dimensional spin-gap materials CaV$_4$O$_9$ [1], Na$_2$Tl$_2$Sb$_2$O and SrCu$_2$(BO$_3$)$_2$ [2]. The latter material is particularly interesting in that, by virtue of the crystal geometry, it is an experimental realization of the Shastry-Sutherland model [3], for which an exact dimerized singlet eigenstate can be written down, which for a range of parameters is the ground state of the model. Among the interesting experimental findings for SrCu$_2$(BO$_3$)$_2$ are that the system appears very close to a transition to a Néel phase and it also shows magnetization plateaus as a function of magnetic field [3,5].

Here we consider a generalized Shastry-Sutherland model, with Hamiltonian

$$\mathcal{H} = J_1 \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{<i,k>} \vec{S}_i \cdot \vec{S}_k + J_3 \sum_{<i,l>} \vec{S}_i \cdot \vec{S}_l,$$

(1)

where the bonds corresponding to interactions $J_1$, $J_2$ and $J_3$ are shown in Fig. 1. We assume $J_1$ is antiferromagnetic and can henceforth set $J_1$ to unity. For $J_3 = 0$ the model reduces to the original Shastry-Sutherland model. For $J_2 = J_3$, the model has infinitely many conserved quantities. The total spin on each $J_1$ bond commutes with $\mathcal{H}$ and thus each eigenstate of the Hamiltonian can be characterized by the number and position of the triplets present. These triplets then form (in general a diluted) spin-one Heisenberg model, with nearest-neighbor interactions on the square-lattice. It is easy to show that this model has three phases at $T = 0$ with first order transitions between them. For large negative $J_2$ the ground state is a fully polarized ferromagnet, for large positive $J_2$ the ground state is equivalent to the Néel ground state of a spin-one Heisenberg model on the square-lattice, which is rigorously known to be ordered [4], and whose numerical properties are very well established [5]. In between the ground state is the Dimerized Shastry-Sutherland singlet phase. At the Dimer to Néel transition the sublattice magnetization jumps from 0 to about 80 percent of the classical value.

Away from the $J_2 = J_3$ line, we use symmetry arguments, Dimer series expansions [6] together with considerations of the classical limit to discuss the ground-state phase diagram and properties of this model. The model with $J_2$ and $J_3$ interchanged can be mapped into the original one by interchanging the spins on all $J_1$ bonds. Thus the phase diagram is symmetric with respect to the $J_2 = J_3$ line. We will concentrate our discussion on...
the region $J_2 \geq J_3$. First, let us compare energies of various classically ordered phases with the energy of the Dimer phase to get a first view of various phases and their boundaries.

\[
\cos (\pi - \theta) = \frac{2(J_2 - J_3)}{J_1 + \sqrt{J_1 - 24J_3(J_2 - J_3)}}
\]

Defining $x = J_2/J_1$, $y = J_3/J_1$, the columnar, helical, Dimer triple point in the classical phase diagram is located at $x_{tr} = (45 - 4\sqrt{6})/36 \approx 0.9778$, $y_{tr} = -(9 + 4\sqrt{6})/36 \approx -0.5222$. The Néel-helical phase boundary is given by $x + 5y = 1$, whereas the asymptotic ($x \to \infty$) phase boundary between the helical and columnar phases is given by the equation $y = \frac{1}{5}(-8x + 1 + O(1/x))$.

Since there are no quantum fluctuations in the ferromagnetic and Dimer phases and on the boundary between ferromagnetic and helical phases, these remain true phase boundaries in the model and are shown by solid lines in Fig. 2. Note that the Dimer-ferromagnetic boundary is first order, whereas the Dimer-helical boundary is second order. The other classical phase boundaries are shown by dashed lines. They leave an oval-like Dimer phase in the middle. The first-order Dimer-Néel phase boundary can be determined quite accurately along $J_2 = J_3$ to be at $x = 0.42957(2)$, and has been given in previous numerical studies along $J_3 = 0$ (and equivalently $J_2 = 0$). These points are shown by solid dots and we connect them smoothly to indicate a first order Dimer-Néel phase boundary.

Actually, it is not evident whether the Dimer-Néel phase boundary along the line $J_3 = 0$ is first or second order, or even whether there is an intermediate phase between the two. Albrecht and Mila have argued that there is an intermediate helical phase between the Néel and Dimer phases. Their Schwinger Boson mean-field treatment leads to the estimate that the Néel phase extends only down to $x \approx 0.91$, whereas the helical phase exists between $0.61 < x < 0.91$. On the other hand, using series expansions, Weihong et al. have argued that the Néel phase extends down to $x = 0.691$ at which point there is a first order transition to the Dimer phase. The finite-size calculations also do not suggest any helical phases. Though, Albrecht and Mila have argued that this is because the helical phases are not properly accommodated in finite geometries.

The quantitative validity of Schwinger Boson calculations is difficult to judge. One generally expects quantum fluctuations to stabilize colinear phases. And this could considerably reduce the extent of the helical phases in the phase diagram. In several spin models, where numerical calculations have been done, the sublattice magnetization of the Néel phase goes continuously to zero and it is separated from incommensurate phases by a singlet phase. On general grounds, Ferrer has argued that the Néel phase must be separated from helicoidal phases by an intermediate spin-liquid phase. Thus, it is reasonable to assume that along $J_3 = 0$ there is a direct transition between Dimer and Néel phases.

Along $J_3 = 0$, Weihong et al. estimate that the Dimer to Néel transition happens at $J_2/J_1 = 0.691(6)$. Using d-log Pade approximants to analyze the gap series, we estimate that it vanishes at $J_2/J_1 \approx 0.697(2)$. Thus, within the uncertainties of the series analysis, this transition could be continuous. Around this value of the couplings, the sublattice magnetization series from the Néel side is also consistent with zero. The primary reason for believing that the transition is first order is that

\[\text{FIG. 2. Phase diagram for the model showing ferromagnetic (F), Néel (N), columnar (C}_F\text{ and C}_N\text{), helical (H) and dimer (D) phases. See text for discussion of phase boundaries.}\]
the energies for the Néel and Dimer phases appear to cross at a non-zero angle. However, if the transition is continuous, then very close to the transition, the Néel energy curves should change slope [8]. Thus, given all the numerical evidence, a plausible conclusion is that the transition is very weakly first order, though it could also be second order.

Given the above, it is natural to expect some continuous transitions when \( J_3 < 0 \). To explore this possibility, we have developed series expansions for the triplet excitations in the dimer phase to 15th order for arbitrary \( J_3/J_2 \) using the flow equation method [10]. For \( J_3 = 0 \), the expansion coefficients agree with those calculated by Weihong et al. The unit cell of the lattice contains two dimers, giving rise to two triplet modes. These modes are almost degenerate throughout the Brillouin zone and become exactly degenerate as \( q \to 0 \) due to symmetry. We find that the gap minimum is always at \( q \) equal to zero even as one moves from the Néel towards the ferromagnetic phase. We use d-log Pade approximants to calculate the locus of points, where the triplet gap closes. This contour is also depicted by a solid line in Fig. 2. It marks a boundary at which the Dimer phase becomes locally unstable, and hence the dimer phase cannot exist beyond that line. Without studying all eigenstates of our system, it is not possible to say if some other level crossing transition leads to a different ground state before we get to this line. It is plausible to think that at least parts of this line represent continuous phase transitions between the Dimer and magnetically ordered phases. As seen in Fig. 2, a possible consistent scenario is that very near the \( J_3 = 0 \) line, we have a multicritical point where a second order transition line meets a first order phase boundary.

These continuous phase transitions between the Dimer phase and various magnetic phases are rather unusual. They are not in the conventional 2 + 1-dimensional \( O(3) \) universality class as expected for the non-linear sigma models [13]. This is evident from the fact that in the Dimer phase, the ground state remains unchanged and hence the correlation length remains of order unity. In contrast, for a generic dimerized spin system, the correlation length gradually grows and diverges as the gap goes to zero [14]. The continuous phase transition, here, is somewhat analogous to the density driven generic phase transitions in the Bosonic Mott insulators [15].

However, there are some important differences. Unlike the case of Bosonic Mott insulators, the spectrum appears to be linear at the transition. Along the \( J_3 = 0 \) line, we estimate that the gap vanishes at \( x = 0.697(2) \), with an apparent exponent \( \nu \) of 0.45(2). Different d-log Pade approximants show remarkable consistency with each other. Fig. 3 shows the spectrum, in the reduced Brillouin zone, at the transition. Different ways of analyzing the series all point to a finite spin-wave velocity and a linear spectrum. These results suggest that this transition belongs to a new universality class [17].

With our present calculations we cannot study the transitions from the ordered side and thus cannot establish the full nature of these phase transitions nor can we say anything about the stability of columnar and helical phases in the overall phase diagram. Quantum fluctuations can lead to additional singlet (spin-gap) phases between the Néel and the helical phases and possibly eliminate helical phases altogether from the Néel side of the phase diagram. These Néel to singlet phase transitions should be similar to those found in the \( J_1 - J_2 \) square-lattice Heisenberg model [18].

When a magnetic field is applied to the Dimer ground state along the special line \( J_2 = J_3 \), the resulting magnetization is shown in Fig. 4. The triplet excitations, aligned by the field, have no dispersion, but a nearest neighbor repulsion. Thus they form a simple Wigner crystal (or a Bosonic Mott insulator) at one half the saturation magnetization. If we add an additional weak antiferromag-
netic coupling between neighboring horizontal $J_1$ bonds (and similarly neighboring vertical $J_1$ bonds) of the form $J_4(S^1_1 + S^2_2) \cdot (S^3_1 + S^4_2)$, the triplets remain dispersionless but they now have an additional second neighbor repulsion. This leads to additional Wigner crystal phases at one and three quarter fillings and hence plateaus in the magnetization at one and three quarters of the saturation value. In Fig. 4, we also show the ordering pattern of the Wigner crystal on different plateaus by showing four $J_1$ bonds. A line denotes a $(S^z = 1)$ triplet on the bond whereas a circle denotes a singlet.

As we move away from the $J_2 = J_3$ line, the triplet develops dispersion. It is useful to think of the problem in terms of the Bose Hubbard model \cite{16}, with the $(S^z = 1)$ triplet representing hard core Bosons. These Bosons have a strong nearest-neighbor repulsion and a weak diagonal (second-neighbor) and further-range hopping. At half-filling, the strong repulsion will clearly lead to a Wigner crystal and hence the magnetization plateaus will remain. However, the transition between the magnetization plateaus may now be second order. Indications of such plateaus also exist in the finite size calculations of Miyahara and Ueda for $J_3 = 0$ \cite{18}. However, due to the finite size, all plateaus appear discontinuously in their study. The question of whether there will be additional magnetization plateaus at other rational fillings perhaps including valence bond states, as in one dimension \cite{19}, deserves further attention.

We now comment on the materials: One would naively expect the SrCu$_2$(BO$_3$)$_2$ system to be close to $J_3 = 0$, a limit that has been considered by other authors \cite{14}. The ratio of $J_2$ to $J_1$ has been placed in the literature \cite{14} close to the Dimer to Néel transition. Even though this transition may be first order, it would be very weakly so, due to the vicinity of the multicritical point. In this sense, this material may allow us to study a special quantum critical point, not the generic Néel to singlet quantum phase transition. However, this transition may be unstable to the generic $O(3)$ transition, when the special eigenstate of Shastry and Sutherland is not a true eigenstate due to some small perturbations. It would be interesting to study this crossover theoretically. An interesting problem could be the instability of these special transitions due to spin-lattice couplings.

Magnetization plateaus have been observed in the material at one eighth and one quarter of the saturation magnetization. The exactly soluble model suggests that these phases may be regarded as simple Wigner crystals of local triplets. The question of whether the models away from $J_2 = J_3$ will have magnetization plateaus at other rational fractions, or whether other couplings such as $J_4$ are needed for this, deserves further attention.

We thank Ian Affleck, Leon Balents and Matthew Fisher for discussions. This work is supported in part by the National Science Foundation grants PHY94-07194 and DMR96-16574 and by the Deutsche Forschungsgemeinschaft, SFB 341 and SPP 1073.

---

\* Permanent address: Institut für Theoretische Physik, Universität zu Köln, Zülpicher Str. 77, D-50937 Köln, Germany.
\dagger Permanent address: Department of Physics, University of California, Davis CA 95616.

[1] S. Taniguchi et al., J. Phys. Soc. Jpn., 64, 2758 (1995).
[2] E. A. Axtell et al., J. Solid State Chem., 134, 423 (1997).
[3] H. Kageyama et al., Phys. Rev. Lett. 82, 3168 (1999).
[4] B. S. Shastry and B. Sutherland, Physica B 108, 1069 (1981).
[5] S. Miyahara and K. Ueda, Phys. Rev. Lett. 82, 3701 (1999).
[6] E. Jordao Neves and J. Fernando Perez, Phys. Lett. 114A, 331 (1986).
[7] Z. Weihong, J. Oitmaa and C. Hamer, Phys. Rev. B 43, 8321 (1991).
[8] Z. Weihong, C. J. Hamer and J. Oitmaa, Phys. Rev. B 60, 6608 (1999).
[9] M. Albrecht and F. Mila, Europhys. Lett. 34, 145 (1996).
[10] C. Knetter and G. S. Uhrig, cond-mat/9906243.
[11] M. P. Gelfand, R. R. P. Singh and D. A. Huse, Phys. Rev. B 40, 10801 (1989). W. H. Zheng, R. H. McKenzie and R. R. P. Singh, Phys. Rev. B 59, 14367 (1999).
[12] J. Ferrer, Phys. Rev. B 47, 8769 (1993).
[13] S. Chakravarty, B. I. Halperin and D. R. Nelson, Phys. Rev. Lett. 60, 1057 (1988); A. V. Chubukov, S. Sachdev and J. Ye, Phys. Rev. B 49, 11919 (1994).
[14] R. R. P. Singh, M. P. Gelfand and D. A. Huse, Phys. Rev. Lett. 61, 2484 (1988).
[15] M. P. A. Fisher, P. B. Weichman, G. Grinstein and D. S. Fisher, Phys. Rev. B 40, 546 (1989).
[16] G. G. Batrouni, R. T. Scalettar, G. T. Zimanyi and A. P. Kampf, Phys. Rev. Lett. 74, 2527 (1995); N. Elstner and H. Monien, Phys Rev B 59, 12184 (1999).
[17] The estimated value of $\nu$ increases slowly as one moves towards the ferromagnetic phase and is about 0.73 along the negative $J_4$ axis.
[18] N. Read and S. Sachdev, Phys. Rev. Lett. 66, 1773 (1991).
[19] M. Oshikawa, M. Yamanaka and I. Affleck, Phys. Rev. Lett 78, 1984 (1997).