Exact Demonstration of Magnetization Plateaus and First-Order Dimer-Néel Phase Transitions in a Modified Shastry-Sutherland Model for $SrCu_2(BO_3)_2$

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We study a generalized Shastry-Sutherland model for the material $SrCu_2(BO_3)_2$. Along a line in the parameter space, we 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.

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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_4O_9 [1], $Na_2Ti_2Sb_2O$ [2], and $SrCu_2(BO_3)_2$ [3]. The latter material is particularly interesting in that, by virtue of the crystal geometry, it is an experimental realization of the Shastry-Sutherland model [4], 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

$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle i,k \rangle} \vec{S}_i \cdot \vec{S}_k + J_3 \sum_{\langle i,l \rangle} \vec{S}_i \cdot \vec{S}_l \,, \tag{1}$$

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. It was noted by Weihong *et al.* [6] that J_3 keeps the dimer state an exact eigenstate. We find that 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 a (in general 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 [7], and whose numerical properties are very well established [8]. 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% 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

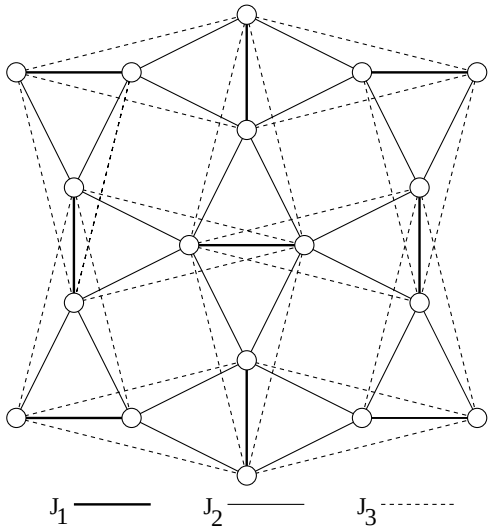

FIG. 1. The geometry of spins and couplings in the material $SrCu_2(BO_3)_2$.

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$J_2 \geq J_3$. First, let us compare energies of various classically ordered phases, obtained by a static mean-field calculation, with the energy of the dimer phase to get a first view of various phases and their boundaries.

The different phases are shown in Fig. 2, with the dimer phase denoted by D. The magnetically ordered phases are best described with respect to a rotated lattice where individual spins have four nearest neighbors (connected by J_2 bonds) [4]. This lattice is topologically equivalent to a square lattice. On this lattice, in addition to a ferromagnetic phase (F) and an antiferromagnetic Néel phase (N), there are columnar (C_N and C_F) and helical (H) phases. The two columnar phases are collinear and equivalent to each other. In the C_N phase the spins order antiferromagnetically along one of the axes and have period four ($\uparrow\uparrow\downarrow\downarrow$) in the perpendicular direction. In the C_F phase, the spins are ferromagnetic in one direction and have period four in the other direction. Classically, the four helical phases can be mapped onto each other. In the helical phase for $J_2 > -J_3 > 0$, the helix runs along one of the axes, with successive spins rotating by an amount $+\theta$ as one moves along that direction [9]. Along the perpendicular direction the change in spin directions alternates between $+\theta$ and $-\theta$. The angle θ is nonunique and is one of the solutions to the equation

$$\cos(\pi - \theta) = \frac{2(J_2 - J_3)}{J_1 + \sqrt{J_1^2 - 24J_3(J_2 - J_3)}}. \quad (2)$$

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 $x_{\rm tr} = (45 - 4\sqrt{6})/36 \approx 0.9778$, $y_{\rm tr} = -(9 + 4\sqrt{6})/36 \approx -0.5222$. 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 $y = \frac{1}{19}[-8x + 1 + O(1/x)]$.

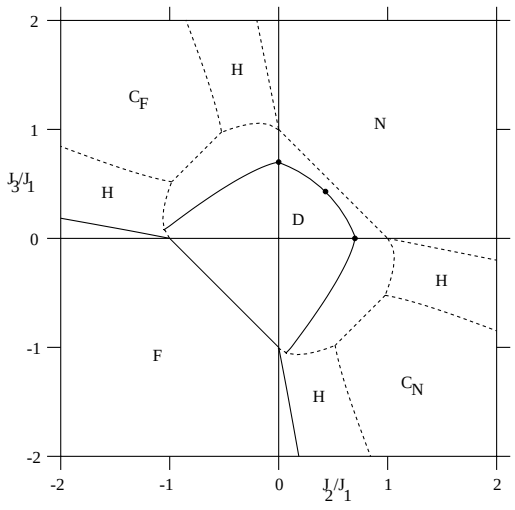

FIG. 2. Phase diagram for the model showing ferromagnetic (F), Néel (N), columnar (C_F and C_N), helical (H), and dimer (D) phases. See text for discussion of phase boundaries.

In the ferromagnetic and the dimer phases there are no quantum fluctuations, as the ground states are exact eigenstates. Hence the phase boundary shown between them is exact. The ferromagnetic-helical boundary shown by solid lines in Fig. 2 is also exact, as we have included the quantum corrections to the helical energy. Second-order perturbation theory is sufficient for the quantum corrections, since the pitch angle θ vanishes at the boundary. Note that the dimer-ferromagnetic boundary is first order, whereas the ferromagnetic-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 $x = 0.429\,57(2)$, and has been given in previous numerical studies along $J_3 = 0$ (and equivalently $J_2 = 0$) [6]. 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 [9] 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.* [6] 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 [5,9] also do not suggest any helical phases. However, 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 collinear 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 [10]. On general grounds, Ferrer [11] 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 Padé 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 [6]. The primary reason for believing that the transition is first order is that the energies for the

Néel and dimer phases appear to cross at a nonzero angle. However, if the transition is continuous, then very close to the transition, the Néel energy curves should change slope [6]. 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.

Note that $J_3 < 0$ favors triplet hopping thus reducing the gap while it disfavors the Néel phase. So negative J_3 pushes the system toward a continuous transition and it is natural to expect a continuous transition below the J_2 axis in Fig. 2. 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 [12]. 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 Padé 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 for $J_3 \leq 0$ but close to $J_3 = 0$, 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 nonlinear 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 ν of 0.45(2). Different d-log Padé approximants show remarkable consistency with each other. Figure 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

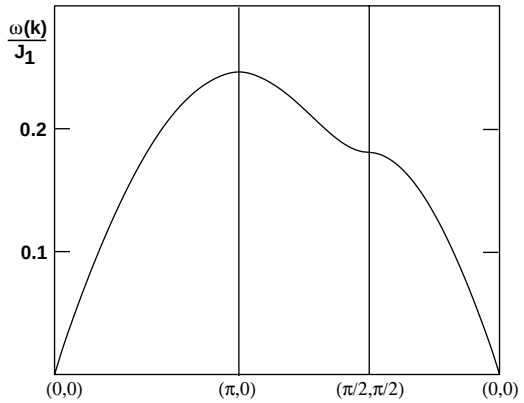

FIG. 3. The triplet spectrum along the $J_3 = 0$ line at $x = 0.697$, where the gap vanishes.

spectrum. These results suggest that this transition belongs to a new universality class [16].

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 [17].

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 antiferromagnetic coupling between neighboring horizontal J_1 bonds (and similarly neighboring vertical J_1 bonds) of the form $J_4(\vec{S}_1 + \vec{S}_2) \cdot (\vec{S}_3 + \vec{S}_4)$, the triplets remain

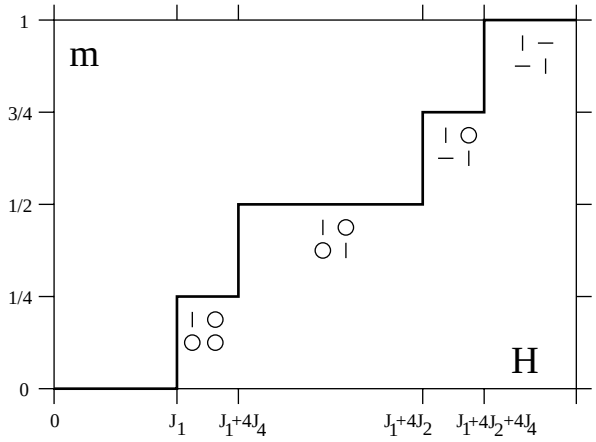

FIG. 4. Magnetization as a function of magnetic field. Note that the plateaus are valid for $J_4 < J_2 < 0.43J_1$.

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 [18], 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 $J_3 = 0$ [5]. 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 [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 [5,6]. The ratio of J_2 to J_1 has been placed in the literature [5] 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.

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