Magnetization Plateaus of $SrCu_2(BO_3)_2$ from a Chern-Simons Theory

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The antiferromagnetic Heisenberg model on the frustrated Shastry-Sutherland lattice is studied by a mapping onto spinless fermions carrying one quantum of statistical flux. Using a mean-field approximation these fermions populate the bands of a generalized Hofstadter problem. Their filling leads to the magnetization curve. For $SrCu_2(BO_3)_2$ we reproduce plateaus at $1/3$ and $1/4$ of the saturation moment and predict a new one at $1/2$. Gaussian fluctuations of the gauge field are shown to be massive at these plateau values.

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Two-dimensional (2D) quantum spin systems that do not order magnetically at zero temperature are currently a subject of great theoretical and experimental interest. The recently discovered [1] compound $SrCu_2(BO_3)_2$ is an antiferromagnet (AF) with localized spins $S = \frac{1}{2}$, a gap above the ground state [2], and the unique property that its magnetization curve has plateaus at $1/3$, $1/4$, and $1/8$ of the full saturation moment [3]. The spin system may be described by a 2D Heisenberg model on a square lattice with exchange constant J' and additional diagonal bonds J on half of the square plaquettes (see inset of Fig. 1).

This lattice was studied many years ago by Shastry and Sutherland [4] who noted that there is an exact eigenstate which is obtained by putting singlets on all diagonal J bonds. This eigenstate is the ground state for a wide interval of J'/J. For J'/J smaller than ~ 0.7 [5], the system has dimer long-range order, and for larger J' it has conventional Néel long-range AF order. There may be additional phases in between such as a plaquette singlet phase [6–8] but they are apparently not realized in $SrCu_2(BO_3)_2$ where J'/J is estimated to be smaller than 0.65 [9,10]. This dimer ground state explains the spin gap as seen in experiments. However, the existence of plateaus has no immediate explanation since the simplicity of the ground state does not extend to the excited states of this peculiar lattice. In this Letter we use a mean-field approximation with a Chern-Simons (CS) field-theoretic approach to quantum magnets suggested some time ago [11,12], and we obtain an excellent quantitative fit of the magnetization curve [see Fig. 1] for realistic values of the exchange constants. As we discuss, this may be evidence for unconventional character of the plateau ground states.

Starting from the pure dimer state, the first excited state can be constructed by first breaking a singlet bond into a triplet state. Because of the exchange J', such a state will move and we expect a dispersive band of triplets as low-lying states at least for strong J'.

However, due to the peculiar triangular coupling between the diagonal bonds, the hopping of the triplet is forbidden at low orders in perturbation theory [2]. As a consequence, the triplet band is very flat, a striking fact observed by neutron scattering experiments [13]. Since the triplets are very massive particles, it is natural to expect that they can crystallize at finite density, and it has been proposed that the plateaus are Wigner crystals of triplets [14–16]. There exists a spin model which is derived from the Shastry-Sutherland Hamiltonian [17] for which the plateaus are demonstrated to originate from such ordered states. However, in this model there are plateaus at $1/4$, $1/2$, and $3/4$, and the overall shape of the magnetization curve is not in agreement with experiments. A closely related physical picture is obtained by describing the magnetized triplets by hard-core bosons [15]. Then the repulsion may favor charge-density wave states that are among the known insulating phases of the lattice Bose gas.

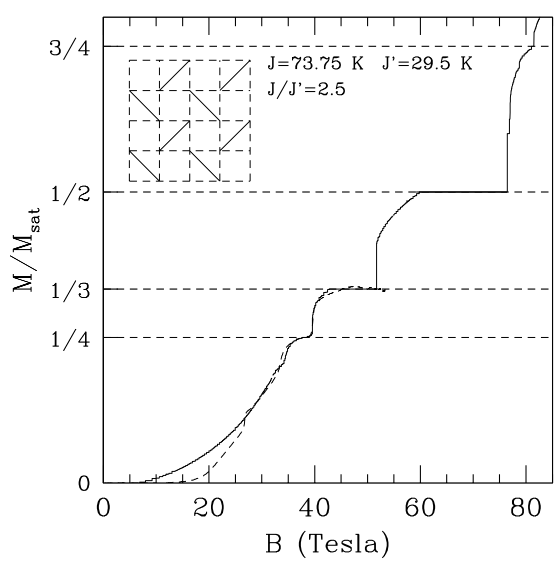

FIG. 1. Comparison between the magnetization curve of $SrCu_2(BO_3)_2$ measured by Onizuka *et al.* (dashed line) and the mean-field result (solid line). Inset: Shastry-Sutherland lattice. The exchange interaction is J on black links and J' on the dotted ones.

We take a different approach here, mapping the spin problem onto a hard-core boson problem and then solving the hard-core constraint exactly by a further mapping onto spinless fermions coupled to a CS gauge field [12,18,19]. Within a mean-field approximation, the spin excitation gap that produces the observed magnetization plateaus arises from some of the Landau level gaps in the integer quantum Hall effect for the fermions on a lattice.

The Hamiltonian for the AF Heisenberg model on the Shastry-Sutherland lattice is

$$H = \sum_{\langle i,j \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j - B \sum_i S_i^z , \tag{1}$$

where $\vec{S}_i$ are spin-$\frac{1}{2}$ operators, the exchange couplings J_{ij} are equal to J' when i, j are nearest neighbors on the square lattice and equal to J when i, j are related by a diagonal bond ($J, J' > 0$), and the external magnetic field B is applied along the z axis. We then map the spin operators to hard-core boson operators:

$$H = H_{xy} + H_z , \tag{2}$$

$$H_{xy} = \frac{1}{2} \sum_{\langle i,j \rangle} J_{ij} (b_i^\dagger b_j + b_j^\dagger b_i) , \tag{3}$$

$$H_z = \sum_{\langle i,j \rangle} J_{ij}^z (n_i - 1/2)(n_j - 1/2) - B \sum_i (n_i - 1/2) , \tag{4}$$

where $n_i \equiv S_i^z + 1/2$ is the occupation number of site i. The bosons are then treated as fermions with an attached flux tube carrying one flux quantum of fictitious magnetic field. This can be formulated as an *exact* mapping between the spin problem and spinless fermions interacting with a statistical CS gauge field. In a mean-field treatment, the flux tubes are smeared out into a uniform background magnetic field. The flux per square plaquette ϕ is then tied to the density of fermions and thus to the magnetization of the spin system:

$$\frac{\phi}{2\pi} = \langle n \rangle = \left(\langle S^z \rangle + \frac{1}{2} \right) = M + \frac{1}{2} . \tag{5}$$

It is important to note that the *real* magnetic field B applied to the spins acts as a chemical potential for the fermions as seen from Eq. (4) but does not contribute to the *statistical* flux. We treat the Ising term H_z in Eq. (4) by a simple mean-field decoupling so it becomes a simple function of the magnetization. The kinetic energy term H_{xy} leads to a Hofstadter [20] problem for fermions hopping on the Shastry-Sutherland lattice. We use a flux attachment choice leading to flux $\phi/2$ on each triangular plaquette. The one-body problem from H_{xy} can be straightforwardly analyzed for rational values $\phi = 2\pi(p/q)$.

Fixing the magnetization M gives us the flux and the number of fermions through Eq. (5). For this value of ϕ, we compute the band spectrum of H_{xy} and fill the bands with the available fermions. The energy of the filled bands leads to a first contribution $E_{xy}(M)$. We then add the contribution from the Ising interaction H_z to obtain the total energy $E(M)$. The magnetization is obtained as a function of B by minimizing $E(M) - BM$. The Hofstadter diagram for $J = J'$ is given in Fig. 2 where the lowest curve marks the Fermi level (highest occupied state) and the upper one marks the lowest unoccupied level. Jumps of the Fermi energy as a function of M lead to discontinuity of the slope of the function $E(M)$. These jumps corresponds to plateaus in the magnetization curve. The structure of the Hofstadter butterfly thus reflects itself in the appearance of plateaus. The effects of geometry are encoded in the Hofstadter spectrum, whose complexity arises from the diffraction of the cyclotron orbits by the lattice.

When J is set to zero, the model reduces to the square lattice which has Néel long-range order. The wave functions obtained in the spatially uniform CS mean-field approximation certainly do not encompass this physics. However, the magnetization curve obtained from this approach is qualitatively similar to that of the ordered system: It is featureless all the way to full saturation (see the $J = 0$ curve in Fig. 3). If we consider the case of the triangular lattice, the uniform mean field leads to curve 4(a). In addition to the zero-field gap found by Yang, Warman, and Girvin [18], there are many plateaus in 4(a). This is unrealistic: The triangular lattice spin system is known to be long-range ordered and, hence, gapless, and its magnetization process shows only a single plateau [21] at $M/M_{\mathrm{sat}} = 1/3$. There is a way

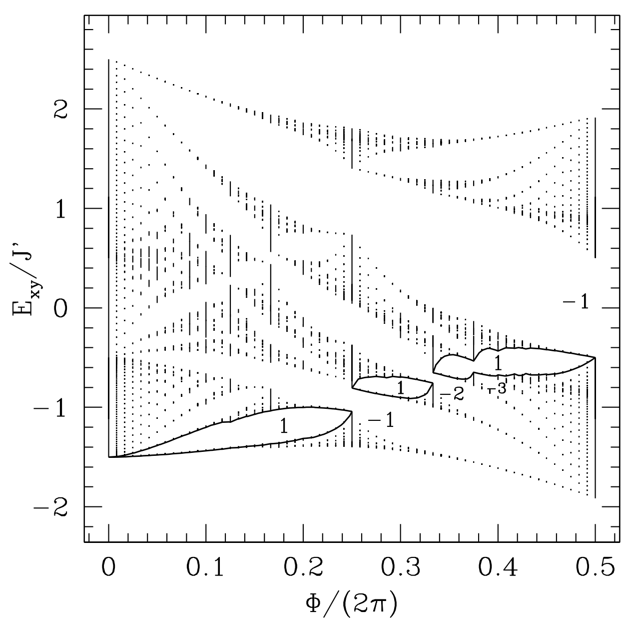

FIG. 2. Hofstadter spectrum for the Shastry-Sutherland lattice at $J = J' = 1$. Vertical lines mark the energy bands as a function of the statistical flux ϕ per square plaquette. Hall conductances σ_{xy} [Thouless–Kohmoto–Nightingale–den-Nijs (TKNN) integers] are indicated for the regions of the spectrum which are explored in the magnetization process from $M/M_{\mathrm{sat}} = -1$ ($\phi = 0$) to $M/M_{\mathrm{sat}} = 0$ ($\phi = \pi$).

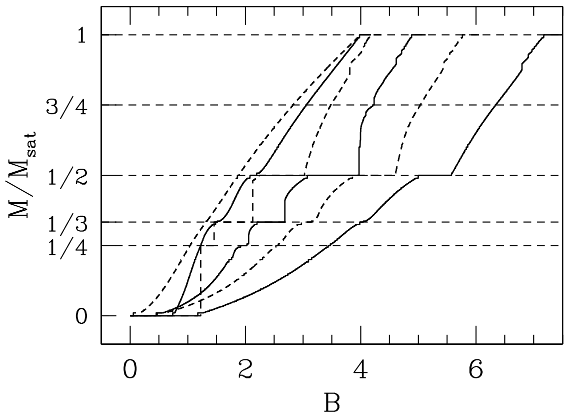

FIG. 3. Magnetization curves for the Shastry-Sutherland model (uniform mean field). From left to right $J = 0$ (dashed line), 0.75 (full line), 1.5 (dashed line), 2.5 (full line), 3.5 (dashed line), and 5 (full line), with $J' = 1$.

to improve these results by allowing the mean field to have a three-sublattice structure: One introduces three fermion densities n_A, n_B, n_C and numerically searches for a self-consistent solution where the flux ϕ_α matches the density n_α on each sublattice. For $M = 0$, we find that the self-consistent solution remains uniform. However, for nonzero magnetization the translation symmetry is broken. This nonuniform mean-field solution leads to a magnetization curve 4(b) which is much closer to the truth albeit the zero-field ground state remains unrealistic (Néel long-ranged order is absent). The $1/3$ plateau has a semiclassical origin: It is easily understood in the Ising limit $J^z \gg J$ and survives up to the isotropic point $J^z = J$ [21]. Our calculation reproduces these features and we find the three-sublattice magnetizations $n_A = n_B = 0.922$ and $n_C = 0.155$.

We have explored the magnetization process of the Shastry-Sutherland lattice as a function of the ratio J/J'. The curves $M(B)$ are drawn in Fig. 3. For small J we are close to the square lattice result, i.e., a smooth curve. Increasing J we observe plateaus developing, the more complex structure being in the regime $J/J' \approx 2$–3.

For very large J the curves are again simple. There remain only two plateaus at 0 and $1/2$. The plateau at zero

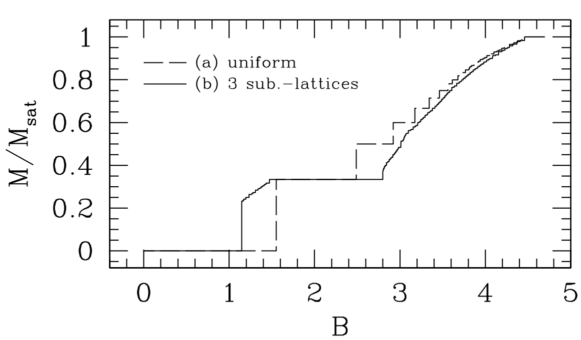

FIG. 4. Magnetization curve of the triangular antiferromagnet: (a) uniform mean field; (b) mean-field with three sublattices.

field is due to the fact that for large J the tight-binding bands separate into two groups with very small dispersion, and there is essentially a huge gap in the Hofstadter spectrum that does not depend much upon flux. For $J' = 0$ and an appropriate choice of gauge, our wave function exactly reproduces the dimer limit. In the intermediate regime, the curve depends on the details of the spectrum. The plateaus have a finite domain of stability which is given in Fig. 5.

To reproduce the qualitative shape of the experimental magnetization curve for $SrCu_2(BO_3)_2$, we find that $J' = 29.5$ K and $J = 74$ K; the resulting fit is shown in Fig. 1. While the zero-field gap is not well reproduced, we find 13 instead of 34 K, the field strengths at which the plateaus occur and the roundings close to the plateaus are in very good agreement with the experiment. The values of J, J' are reasonably close to the recent estimates [10] from neutron scattering data $J' = 43$ K and $J = 71$ K. These values are not precisely known since there is some amount of three-dimensional dispersion of the low-lying triplet that should be taken into account. We predict prominent plateaus at $1/3$ and $1/2$ and nothing else until full saturation. Note also that at $1/4$ there is in fact an avoided plateau: The value of J'/J is just outside the range of stability of the $1/4$ plateau on Fig. 5. The result of CS mean-field theory is quite different from the semiclassical analysis of the effective Bose gas [16] and from the exactly soluble spin model [17].

As can be seen in Fig. 5, the spin gap (plateau at $M/M_{sat} = 0$) opens at $J/J' = 0$ in this approach instead of opening at the correct critical coupling $J/J' \sim 1.5$. This feature comes from the fact that the Néel state of the square-lattice antiferromagnet is not correctly described in the *uniform* mean-field approximation, as mentioned above in the case of the triangular-lattice antiferromagnet. This is corrected by computing nonuniform solutions of the mean-field CS approach with two sublattices [19].

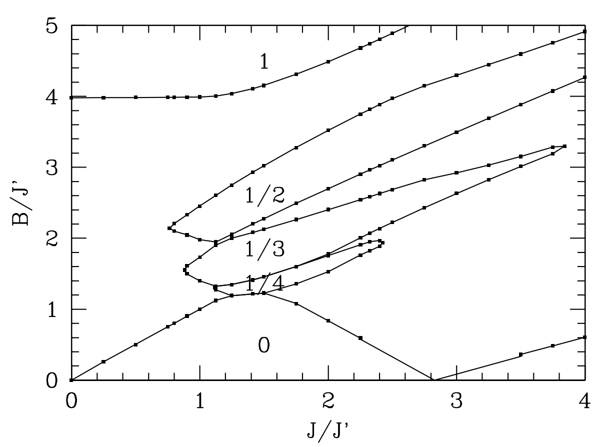

FIG. 5. Width of the magnetization plateaus vs J/J' for $M/M_{sat} = 0$, $\frac{1}{2}$, $\frac{1}{3}$, and $\frac{1}{4}$. Additional plateaus at fractions $\frac{1}{n}$ for $n \geq 5$ exist near $J/J' \simeq 1.5$.

From the good fit in Fig. 1, we deduce that the CS mean-field theory works better for magnetized than unmagnetized states. This is because the initial mapping to hard-core bosons selects out a preferred spin quantization axis which, together with the mean-field treatment of the Ising term, obscures the SU(2) symmetry in the underlying spin problem. The Zeeman energy reduces the SU(2) to the U(1) symmetry associated with conservation of S^z, which in the language of the fermions is simply particle number conservation. We note that the physical magnetic field breaks T symmetry in the orbital part of a Hubbard model description of the system but has no effect in a pure spin model description other than to introduce a Zeeman term. However, our mean-field solution does break T symmetry. The increased size of the "magnetic" unit cell (due to the CS flux) gives an integer number of particles per unit cell as required for the existence of a gap [22] even though there is no translation symmetry breaking in the spin density.

It is important to check whether the plateau states are robust to fluctuations beyond mean field. Following an idea of Fradkin [23], Yang *et al.* [18] showed that Gaussian fluctuations of the CS gauge field are massive provided that the Thouless–Kohmoto–Nightingale–den-Nijs [24] integer describing the quantized Hall coefficient of the fermions on the frustrated lattice differs from the continuum value of *unity*. We have computed the TKNN integers by following the evolution of gaps from the square lattice case where the TKNNs are given by a Diophantine equation. None of the plateaus are suppressed by fluctuations since we find $\sigma = -3$, -2, and -1, respectively, for the plateaus at $\frac{1}{4}$, $\frac{1}{3}$, and $\frac{1}{2}$ (see Fig. 2). At $M/M_{\rm sat} = 0$, we have $\sigma = -1$ (respectively, 0) for $J/J' < \sqrt{(2)}$ [respectively, $J/J' > \sqrt{(2)}$].

A consequence of these nontrivial quantized Hall coefficients for the fermions is that the spin state is chiral and exhibits a "spin quantum Hall effect" [25]. Whether this new physics is actually occurring in $SrCu_2(BO_3)_2$ or is an unrealistic feature of our mean-field approximation remains to be seen. One test of our model is the prediction of a strong plateau in the magnetization beginning at 60 T, a field which is within reach of modern pulsed magnets.

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Note added.—Totsuka *et al.* [26] have recently investigated the nature of various excited states in the Shastry-Sutherland model.

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