Quantum Phase Transitions in the Shastry-Sutherland Model for SrCu$_2$(BO$_3$)$_2$

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We investigate the quantum phase transitions in the frustrated antiferromagnetic Heisenberg model for SrCu$_2$(BO$_3$)$_2$, by using the series expansion method. It is found that a novel spin-gap phase, which is adiabatically connected to the plaquette-singlet phase, exists between the dimer and the magnetically ordered phases known so far. We show that the ratio of the competing exchange couplings

\[ \frac{J}{J_0} \]

is varied, this spin-gap phase exhibits the first-order quantum phase transition to the dimer (the magnetically ordered) phase at the critical point

\[ \kappa_1 = 0.677(2) \]  \[ \kappa_2 = 0.86(1) \]

Our results shed light on some controversial arguments about the nature of the quantum phase transitions in this model.

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Two-dimensional (2D) antiferromagnetic quantum spin systems with the spin gap have been the subject of considerable interest. A typical compound found recently is SrCu$_2$(BO$_3$)$_2$, in which the characteristic lattice structure of the Cu$^{2+}$ spins (see Fig. 1) stabilizes the singlet ground state. This system has been providing a variety of interesting phenomena such as the plateaux in the magnetization curve observed at $1/3$, $1/4$, and $1/8$ of the full moment. The spin system may be described by the 2D Heisenberg model on the square lattice with some diagonal bonds which is referred to as the Shastry-Sutherland model. The key structure with the orthogonal dimers shown in Fig. 1 makes the system unique and particularly interesting among 2D spin-gap compounds. In this frustrated system, there may occur non-trivial quantum phase transitions when the nearest-neighbor coupling $J$ and the next-nearest-neighbor coupling $J_0$ are varied. Albrect and Mila discussed the possibility of a helical phase between the dimer and the magnetically ordered phases by means of the Schwinger boson mean-field theory. Recent theoretical studies, however, have suggested that there may not be such a helical phase, but the first-order phase transition occurs from the dimer to the ordered phases. Furthemore, recent study claims that the phase transition should be of the second order with a non-trivial critical exponent $\nu = 0.45(2)$. These controversial conclusions may come from the fact that the quantum phase transition in the Shastry-Sutherland model occurs from the strong frustration due to the competing exchange interactions $J$ and $J_0$, and therefore a careful treatment should be necessary to guess the correct nature of the phase transition. In particular, we have to keep in mind that such a strong frustration may possibly stabilize another spin-gap phase distinct from the dimer phase.

In this paper, by calculating the ground state energy, the staggered susceptibility and the spin gap by means of the series expansion method, we find that there should exist a novel spin-gap phase with the disordered ground state, which is stabilized by the strong frustration, between the dimer and the magnetically ordered phases. The spin-gap phase found in this paper undergoes the first-order quantum phase transition to the dimer (the ordered) phase, when the exchange couplings $J$ and $J_0$ are varied. The existence of the new phase can resolve controversial conclusions deduced for the quantum phase transitions in this frustrated model. We also point out that the material SrCu$_2$(BO$_3$)$_2$ lies around the phase boundary between these two spin-gap phases, which may give a natural interpretation for the $1/8$-plateau formation in the magnetization curve.

To investigate the frustrated spin system for the compound SrCu$_2$(BO$_3$)$_2$, we consider the 2D quantum Heisenberg model (Shastry-Sutherland model), which is described by the following Hamiltonian

\[ H = J \sum_{\langle \langle \langle} S_i \cdot S_j + J_0 \sum_{\langle \langle} S_i \cdot S_j \]

where $S_i$ is the $s = 1/2$ spin operator at the $i$th site and $J(J_0)$ represents the nearest-neighbor (next-nearest-neighbor) antiferromagnetic exchange coupling. For later convenience, we introduce the ratio $\kappa = J_0/J$. In Fig.
we have drawn the 2D Heisenberg model schematically. We note that the system with only the next-nearest-neighbor coupling \( J^0 \) is equivalent to the Heisenberg model on the square lattice which has a spontaneous staggered magnetization at \( T = 0 \). From this point of view, the nearest-neighbor coupling \( J \) is regarded as the coupling for a diagonal bond (see Fig. 2), which gives rise to the frustration together with \( J^0 \).

In order to study the quantum phase transitions in this spin system, we employ the series expansion method developed by Singh, Gelfand and Huse [3]. We recall here that the quantum phase transitions in the Shastry-Sutherland model have been discussed by Wehling et al. [4] and Muller-Hartmann et al. [5], by means of the dimer and the Ising expansions, from which the critical point between the dimer phase and the magnetically ordered phase has been estimated as \( c = 0.691(6) \) and \( 0.697(2) \), respectively. As mentioned above, however, there is a controversy to be resolved about the nature of the phase transitions. AISO, in order to determine the complete phase diagram, it is crucial to rule out whether there may exist another spin-gap phase besides the above two phases. We will address this problem in the following by using the series expansion method.

To see our strategy clearly, we start with the 2D quantum spin model schematically shown in Fig. 3, which is topologically equivalent to the original model in Fig. 1. In this gure, we have introduced an auxiliary parameter \( \lambda \), which parametrizes the antiferromagnetic couplings labeled by the bold solid, the thin solid and the dashed lines, respectively, as \( J^0 \), \( J^0 \) and \( J^\text{spin} = \lambda J \). When \( \lambda = 1 \), this system is reduced to the Shastry-Sutherland model for \( \text{SrCu}_2(\text{BO}_3)_2 \).

To proceed the analysis based on the series expansion, we divide the original Hamiltonian eq. (1) into two parts as \( H = J^0 \sum_i S_i \otimes S_j + J^\text{spin} \sum_i S_i \otimes S_j \), where \( i \neq j \). For each bond on the square lattice (see Fig. 3), the first term is the unperturbed Hamiltonian which stabilizes the isolated plaquette singlets with the spin excitation gap. The perturbed Hamiltonian labeled by connects these isolated plaquette singlets, by which a 2D network develops. We expand the staggered susceptibility \( \chi^\text{spin} \), the spin-triplet excitation energy \( E \) and the ground state energy \( E_g \) as a power series in \( \lambda \). Here, to estimate the susceptibility, we introduce the Zeev ansatz \( H^0 = \sum_A S_i^A \otimes S_j^B \), where \( A \) is the staggered magnetic field and \( \sum_B \) denotes one of the two sublattices. Note that an asymptotic analysis of the series expansion is necessary to deduce the accurate phase boundary on which the susceptibility \( \chi^\text{spin} \) diverges and the spin gap \( E_\text{gap} = E \) vanishes. For this purpose, we make use of the Padé approximants [6] for both quantities obtained up to the ninth order in \( \lambda \). Besides ordinary DlogPadé approximants, we also employ biased Padé approximants [7], for which we assume that the phase transition in our 2D quantum spin models should belong to the universality class of the 3D classical Heisenberg model [8]. Then the critical value of \( c \) is determined by the formulas \( A_F(c) \) and \( c \), respectively, for various values of \( \lambda \). Using the Dlog and the biased Padé approximants, we end up with the phase diagram shown in Fig. 3. In this gure, the solid (dashed) line repre-
quentes grows up and the second-order quantum phase transition from the spin-gap phase to the magnetically ordered phase occurs at the critical point $c = 0.56$ for $\lambda = 1$, which has already been studied by several groups [3, 13, 14]. On the other hand, decreasing $\lambda$ enhances the frustration, which in turn suppresses the antiferro magnetic correlation, thus shifting the phase boundary upward for smaller $\lambda$ in the phase diagram. It is seen that two lines obtained from the distinct quantities are in good agreement with each other, which implies that the obtained phase boundary is rather accurate in spite of the lower-order perturbative calculation. By exploiting the phase boundary determined by means of biased Padé approximants for the spin gap, the critical value is given by $c_2 = 0.86(1)$ for $\lambda = 1$. Recall that the system is reduced to the original model only for $\lambda = 1$. We thus find that the Shroffy-Sutherland model has the disordered ground state in the region $(0 < c < c_2)$ on the $c=1$ line.

The above result does not necessarily imply that in the same region $0 < c < c_2$ the system always belongs to the disordered phase which is continuously connected to isolated plaquettes. In fact, it is known that the orthogonal dimer phase appears in the vicinity of $c = 0$. Therefore, it is necessary to clarify how these two spin-gap phases connect with each other by carefully comparing the ground state energy $E_g$. To this end, performing the plaquette expansion up to the seventh order in $c$ with being fixed, we estimate the ground state energy $E_g$ for the Shroffy-Sutherland model ($\lambda = 1$) by means of the first-order inhomogeneous dimer method [3]. The results are shown in Fig. 3. As mentioned above [3, 13], from this figure that further increase of $\lambda$ induces the antiferromagnetic order, whose transition point is determined by the crossing point of the ground-state energy obtained respectively by the Ising and plaquette expansions. The result concerns the second-order phase transition deduced above, and the transition point estimated from the figure is consistent with $c_2 = 0.86(1)$ obtained by the analysis of the susceptibility and the spin gap. Consequently, we end up with the phase diagram for the Shroffy-Sutherland model as shown in Fig. 3. The present results shed light on the controversy among others whether the quantum phase transition in this model exists or not in second order [3, 13, 14]. In those previous studies, it was believed that the phase transition occurs only once between the dimer phase (I) and the ordered phase (III), giving rise to some confusions. Our phase diagram clearly resolves this problem by explicitly showing the existence of the new spin-gap phase (II) which undergoes the first- (II) as well as the second-order transitions (IIS III).

![Figure 3](image3.png)

FIG. 3. Phase diagram for the Shroffy-Sutherland model. The phase I represents the orthogonal dimer phase. The phase II newly obtained is adiabatically connected to the plaquette singlet phase. III is the magnetically ordered phase.

To check the validity of the above phase diagram, we also show the results for the spin gap as a function of $\lambda = J^g / J$ in Fig. 4. In this figure, the results obtained by W. Eshong et al. [3] are shown for the orthogonal dimer phase (I: $c = 0 < c_1$). In the new phase (II: $c_1 < c_2$), we determine the values of the spin gap at $k = 0$ by means of the plaquette expansion up to the fourth order in $c$ with the first-order inhomogeneous dimer method. The results are shown as the dots with the
error bars. As seen in this figure, with the decrease of the second-order transition point $c_2$, the spin-gap continuously grows up to stabilize the disordered ground state. As $c_2$ is further decreased, the first-order phase transition occurs at $c_1$.

In order to further confirm the present results, we have performed a different series expansion by choosing the isolated plaquettes with diagonal bonds as an initial configuration, which is different from the one shown in Fig. [2]. The calculation of the susceptibility up to the fourth order yields second-order transition with $c_2 = 0.87(3)$, being consistent with the above results. Furthermore, to confirm the first-order phase transition between the two spin-gap states, we have checked how the first-order phase transition point known for the 1D orthogonal-dimer chain [4] evolves with the increase of the inter-chain couplings. By performing the exact diagonalization studies for the $4 \times 4$ system, we have found that the first-order transition point for 1D is continuously changed, and in the Shastry-Sutherland case, it coincides with the one found above within reasonable accuracy ($c_1 \approx 0.56$), providing further support to our conclusion on the phase diagram. Although our results still seem to be partly contradicted to the staggered magnetization obtained by W. Eichhorn et al. [2], we believe that this could be resolved by further analysis of the results of the Ising expansion.

Before concluding the paper, a brief comment is in order for the plateau-spin formation in the magnetization curve. Experimentally, the plateaus in the magnetization curve have been observed for the compound SrCu$_2$(BO$_3$)$_2$ at 1/3, 1/4 and 1-8 of the film neutrons [6]. In the theoretical studies [7-11], on the spin chain, it has been clarified that the stripe order of the isolated dimers is important in order to understand the 1/3 and 1/4 plateaus. On the other hand, it is not so trivial why the 1/8 plateau occurs in this compound, although a possible mechanism has been proposed in [10]. We think that the formation of the 1/8 plateau may be the fact that this compound is located around the first-order phase transition point between the two spin-gap phases and thereby possesses the dual properties inherent in two distinct phases in this system. We note here that the new spin-gap phase belongs to the same type as the Heisenberg model on the 1/5-depleted square lattice proposed for CaV$_4$O$_9$. Therefore, it is likely that the 1/8 plateau could occur in the same region discussed by M. Ono and Totsuka [10] for the plaquette system related to the 1/5-depleted Heisenberg model. It is interesting to further clarify the mechanism of the 1/8 plateau by taking into account the above dual properties explicitly, which is now under consideration.

In conclusion, we have discussed the phase diagram for the Shastry-Sutherland model for the compound SrCu$_2$(BO$_3$)$_2$ by means of the series expansion method. Our analysis has shown that there exists a novel spin-gap phase with the disordered ground state, which is adiabatically connected to the plaquette-singlet phase, between the dimer and the magnetically ordered phases known so far. When the exchange coupling ratio $J^{x}_{0}/J$ is varied, the first-order phase transition occurs from the dimer state to the new spin-gap state, while the second-order phase transition occurs from this spin-gap state to the magnetically ordered state. This sheds light on the nature of the quantum phase transitions in this model, and resolves apparently controversial conclusions on this issue.

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References

1. H. Kageyama, K. Yoshidu, R. Stem, N. V. M. Ustinikov, K. Onizuka, M. Kato, K. Kouge, C. S. Richter, T. Goto and Y. Ueda, Phys. Rev. Lett. 82, 3168 (1999).
2. K. Onizuka, H. Kageyama, Y. Ninomiya, K. Indio, Y. Ueda and T. Goto, preprint.
3. B. S. Shastry and B. Sutherland, Physica 10B, 1069 (1981).
4. S. Miyahara and K. Ueda, Phys. Rev. Lett. 82, 3701 (1999).
5. M. A. Imreth and F. M. Liu, Europhys. Lett. 34, 145 (1996).
6. W. Eichhorn, C. J. Hamer and J. O. Stination, Phys. Rev. B 60, 6608 (1999).
7. E. Muller-Hartmann, R. R. Singh, C. Knetter and G. S. Uhrig, cond-mat / 9910165.
8. S. Chakravarty, B. I. Halperin and D. R. Nelson, Phys. Rev. B 39, 2344 (1989).
9. J. D. Recher and A. P. Young, Phys. Rev. B 37, 5978 (1988).
10. R. R. Singh, M. P. Gold and D. A. Huse, Phys. Rev. Lett. 61, 2484 (1988).
11. A. J. Guttmann, in Phase Transitions and Critical Phenomena, edited by C. Domb and J. L. Lebowitz (Aca demic Press, New York, 1989), Vol. 13.
12. M. Feshier and A. Hamada and E. Hines, J. Phys. Rev. B 34, 6481 (1986).
13. Y. Fukumoto and A. O. Guchi, J. Phys. Soc. Jpn. 67, 697 (1998); J. Phys. Soc. Jpn. 67, 2205 (1998).
14. W. Eichhorn, J. O. Stination and C. J. Hamer, Phys. Rev. B 58, 14147 (1998); R. R. Singh, W. Eichhorn, C. J. Hamer and J. O. Stination, cond-mat / 9904064.
15. A. Koga, S. Kuma and N. K. Kawashima, J. Phys. Soc. Jpn. 68, 2373 (1999); cond-mat / 9908458.
16. N. B. Ivanov and J. Richter, Phys. Lett. 232A, 308 (1997).
17. S. Miyahara and K. Ueda, preprint.
18. T. M. Ono and K. Totsuka, cond-mat / 9910057.
19. Y. Fukumoto and A. O. Guchi, preprint.