Quantum Phase Transitions in the Shastry-Sutherland Model for $SrCu_2(BO_3)_2$

Akihisa Koga and Norio Kawakami
Department of Applied Physics, Osaka University, Suita, Osaka 565-0871, Japan
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We investigate 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, adiabatically connected to the plaquette-singlet phase, exists between the dimer and the magnetically ordered phases known thus far. When the ratio of the competing exchange couplings $\alpha(= J'/J)$ is varied, this spin-gap phase exhibits a first- (second-) order quantum phase transition to the dimer (the magnetically ordered) phase at the critical point $\alpha_{c1} = 0.677(2)$ [$\alpha_{c2} = 0.86(1)$]. Our results shed light on some controversial arguments about the nature of quantum phase transitions in this model.

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Two-dimensional (2D) antiferromagnetic quantum spin systems with a spin gap have been the subject of considerable interest. A typical compound found recently is $SrCu_2(BO_3)_2$ [1], 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 plateaus in the magnetization curve observed at $1/3, 1/4$, and $1/8$ of the full moment [1,2]. 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 [3], as pointed out by Miyahara and Ueda [4]. 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 nontrivial quantum phase transitions when the nearest-neighbor coupling J and the next-nearest-neighbor coupling J' are varied. Albrecht and Mila [5] 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 [4,6]. Furthermore, a more recent study [7] claims that the phase transition should be of the second order with the nontrivial critical exponent $\nu = 0.45(2)$. These controversial conclusions may come from the fact that quantum phase transition in the Shastry-Sutherland model suffers from strong frustration due to the competing exchange interactions J and J', and therefore a careful treatment should be necessary to figure out 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 first- (second-) order quantum phase transition to the dimer (the ordered) phase, when the exchange couplings J and J' are varied. The existence of the new phase can resolve controversial conclusions [4–7] deduced for 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 [3,4]), which is described by the following Hamiltonian:

$$H = J \sum_{\mathrm{nn}} \mathbf{S}_i \cdot \mathbf{S}_j + J' \sum_{\mathrm{nnn}} \mathbf{S}_i \cdot \mathbf{S}_j , \qquad (1)$$

where $\mathbf{S}_i$ is the $s = 1/2$ spin operator at the ith site and J (J') represents the nearest-neighbor (next-nearest-neighbor) antiferromagnetic exchange coupling. For later convenience, we introduce the ratio $\alpha = J'/J$. In Fig. 1, we have drawn the 2D Heisenberg model schematically. We note that the system with only the next-nearest-neighbor coupling J' is equivalent to the Heisenberg model on the square lattice which has a spontaneous staggered

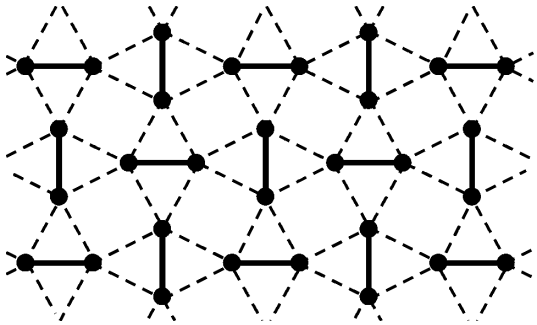

FIG. 1. Lattice structure of the Cu^{2+} spins of $SrCu_2(BO_3)_2$. The nearest-neighbor bonds (J) are expressed by the solid lines and the next-nearest-neighbor bonds (J') by the dashed lines.

magnetization at $T = 0$ [8,9]. 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' [3,4].

In order to study quantum phase transitions in this spin system, we employ the series expansion method developed by Singh, Gelfand, and Huse [10]. We recall here that quantum phase transitions in the Shastry-Sutherland model have been discussed by Weihong *et al.* [6] and Müller-Hartmann *et al.* [7], 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 $\alpha_c = 0.691(6)$ and $0.697(2)$, respectively. As mentioned above, however, there is a controversy to be resolved about the nature of the phase transitions. Also, in order to determine the complete phase diagram, it is crucial to figure out whether there may exist another spin-gap phase besides the above two phases. We will address this problem by using the series expansion method.

To see our strategy clearly, we start with the 2D quantum spin model schematically shown in Fig. 2 [3,4], which is topologically equivalent to the original model in Fig. 1.

In this figure, we have introduced an auxiliary parameter λ, which parametrizes the antiferromagnetic couplings labeled by the bold solid, the thin solid, and the dashed lines, respectively, as J', $\lambda J'$, and $\lambda J'/\alpha (= \lambda J)$. Note that the system is reduced to the original Shastry-Sutherland model in the case of $\lambda = 1$. An important point is that the introduction of λ enables us to perform the cluster expansion starting from the isolated *plaquette singlets* ($\lambda = 0$), which naturally describes the most likely spin-gap phase distinct from the dimer phase.

To proceed with the analysis based on the series expansion, we divide the original Hamiltonian Eq. (1) into two parts as $H = J'[\sum \mathbf{S}_i \cdot \mathbf{S}_j + \lambda \sum \Gamma_{ij} \mathbf{S}_i \cdot \mathbf{S}_j]$, where $\Gamma_{ij} = 1$ or α^{-1} for each bond on the square lattice (see Fig. 2). The first term is the unperturbed Hamiltonian which stabilizes the isolated plaquette singlets with the spin excitation gap. The perturbed Hamiltonian labeled by λ connects these isolated plaquette singlets, from which a 2D network develops. We expand the staggered susceptibility χ_{AF}, the spin-triplet excitation energy $E(\mathbf{k})$, and the ground state energy E_g as a power series in λ. Here, to estimate the susceptibility, we introduce the Zeeman term $H' = h[\sum_{i \in A} S_i^z - \sum_{i \in B} S_i^z]$, where h is the staggered magnetic field and $A(B)$ 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 χ_{AF} diverges and the spin gap $\Delta = E(\mathbf{k} = \mathbf{0})$ vanishes. For this purpose, we make use of the Padé approximants [11] for both quantities obtained up to the finite order in λ. Besides ordinary Dlog Padé approximants, we also employ *biased* Padé approximants [11], 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 λ_c is determined by the formula $\chi_{\mathrm{AF}} \sim (\lambda_c - \lambda)^{-\gamma}$ and $\Delta \sim (\lambda_c - \lambda)^{\nu}$ with the known exponents $\gamma = 1.4$ and $\nu = 0.71$ [12].

We first calculate the staggered susceptibility χ_{AF} and the spin gap Δ by means of the plaquette expansion up to the fourth and the fifth order in λ, respectively, for various values of α. Using the Dlog and the biased Padé approximants, we end up with the phase diagram shown in Fig. 3.

In this figure, the solid (dashed) line represents the phase boundary obtained by the biased Padé approximants for the spin gap (the staggered susceptibility). When $\alpha \to \infty$ and $\lambda = 0$, the system is reduced to an assembly of the isolated plaquettes with the spin gap. As λ is increased, the correlation between these plaquettes grows up and second-order quantum phase transition [8] from the spin-gap phase to the magnetically ordered phase occurs at the critical point $\lambda_c = 0.56$ for $\alpha \to \infty$, which has already been studied by several groups [13–15]. On the other hand, decreasing α enhances the frustration, which in turn suppresses the antiferromagnetic correlation, thus shifting the phase boundary upward for smaller α in the phase diagram. It is seen that two lines obtained from the distinct quantities are in good agreement with each other, which implies

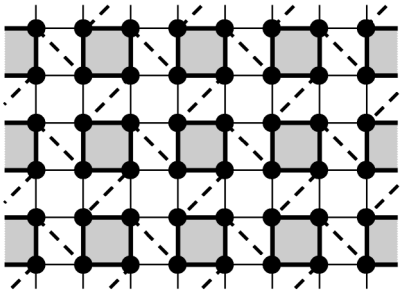

FIG. 2. 2D spin system with the plaquette structure. The solid circles represent the $s = 1/2$ spin. The bold solid, the thin solid, and the dashed lines represent the coupling constants J', $\lambda J'$ and $\lambda J'/\alpha$. When $\lambda = 1$, this system is reduced to the Shastry-Sutherland model for $SrCu_2(BO_3)_2$.

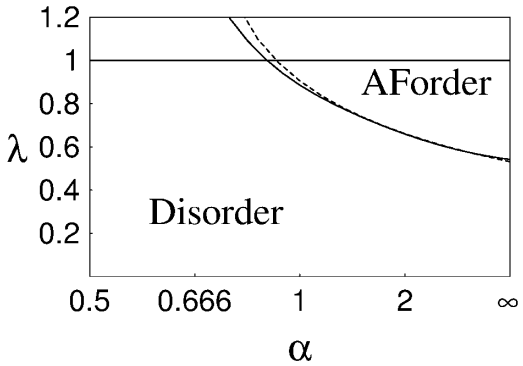

FIG. 3. Phase diagram for the 2D spin system with the plaquette structure in Fig. 2. The solid (dashed) line indicates the phase boundary obtained by biased Padé approximants for the spin gap (the staggered susceptibility).

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 $\alpha_{c2} = 0.86(1)$ for $\lambda = 1$. Recall that the system is reduced to the original model only for $\lambda = 1$. We thus find that the Shastry-Sutherland model has the disordered ground state in the region $(0 < \alpha < \alpha_{c2})$ on the $\lambda = 1$ line.

The above result does not necessarily imply that in the region $0 < \alpha < \alpha_{c2}$ 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 $\alpha = 0$ [3,4]. Therefore, it is necessary to clarify how these two spin-gap phases compete 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 λ with α being fixed, we estimate the ground state energy E_g for the Shastry-Sutherland model ($\lambda = 1$) by means of the first-order inhomogeneous differential method [11]. The results are shown in Fig. 4.

As mentioned above [3,4], the system stabilizes the orthogonal dimer ground state for smaller α. It is found, however, that the first-order transition to the novel spin-gap phase introduced here occurs at the critical point $\alpha_{c1} = 0.677(2)$. It is also seen from this figure that further increase of α induces the antiferromagnetic order, whose transition point is determined by the crossing point of the ground state energy obtained, respectively, by the Ising [6] and plaquette expansions. The result confirms the second-order phase transition deduced above, and the transition point estimated from the figure is consistent with $\alpha_{c2} = 0.86(1)$ obtained by the analysis of the susceptibility and the spin gap. Consequently, we end up with the phase diagram for the Shastry-Sutherland model as shown in Fig. 5.

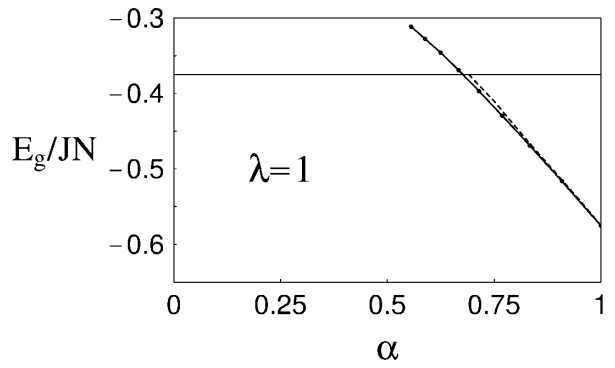

FIG. 4. Ground state energy per site as a function of $\alpha = J'/J$ ($\lambda = 1$, Shastry-Sutherland model). The flat line ($E_g/JN = -3/8$) is the energy of the exact dimer state, while the solid line with dots (error bars are smaller than the linewidth) is obtained by the plaquette expansion. For comparison, we also show the ground state energy obtained by Ising expansion [6] as the dashed line.

The present results shed light on the controversial arguments whether quantum phase transition in this model is of the first or second order [4,6,7]. 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 confusion. Our phase diagram clearly resolves this problem by explicitly showing the existence of the new spin-gap phase (II) which undergoes the first- (I↔II) as well as the second-order transitions (II↔III).

To check the validity of the above phase diagram, we also show the results for the spin gap as a function of $\alpha = J'/J$ in Fig. 6.

In this figure, the results obtained by Weihong *et al.* [6] are shown for the orthogonal dimer phase (I: $0 < \alpha < \alpha_{c1}$). In the new phase (II: $\alpha_{c1} < \alpha < \alpha_{c2}$), we determine the values of the spin gap at $\mathbf{k} = \mathbf{0}$ by means of the plaquette expansion up to the fifth order in λ with the first-order inhomogeneous differential method. The results are shown as dots with error bars. As seen in this figure, with the decrease of α from the second-order transition point α_{c2}, the spin gap continuously grows up to stabilize the disordered ground state. As α is further decreased, first-order phase transition occurs at α_{c1}.

In order to further confirm the present results, we have performed a different series expansion by choosing 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 $\alpha_{c2} = 0.87(3)$, being consistent with the above results. Furthermore, to confirm 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 [16] evolves with the increase of the interchain couplings. By performing the exact diagonalization studies for the 4×4 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 ($\alpha_{c1} \sim 0.66$), providing further support to our conclusion on the phase diagram. Although our results still seem to be partly contradicted by the staggered magnetization obtained by Weihong *et al.* [6], 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 formation in the magnetization curve.

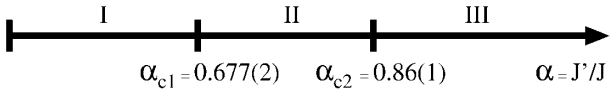

FIG. 5. Phase diagram for the Shastry-Sutherland model. Phase I represents the orthogonal dimer phase. Phase II newly obtained is adiabatically connected to the plaquette singlet phase. Phase III is the magnetically ordered phase.

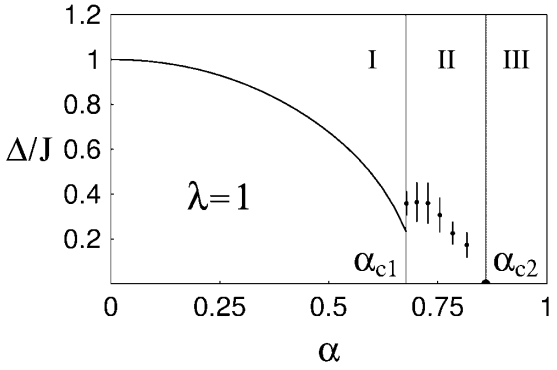

FIG. 6. The spin gap as a function of $\alpha = J'/J$ for the Shastry-Sutherland model. The solid line for $\alpha < \alpha_{c1}$ is the result obtained by Weihong *et al.* [6]. The dots with error bars for $\alpha_{c1} < \alpha < \alpha_{c2}$ represent the spin gap at $\mathbf{k} = \mathbf{0}$ obtained by the plaquette expansion.

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 full moment [1,2]. In theoretical studies [17–19] on the dimer phase, it has been clarified that the stripe order of the isolated dimer triplets is important 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 [17,18]. We think that the formation of the $1/8$ plateau may reflect 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 implicitly. We note here that the new spin-gap phase belongs to the same phase as the Heisenberg model on the $1/5$-depleted square lattice proposed for CaV_4O_9. Therefore it is likely that the $1/8$ plateau could occur in the same origin discussed by Momoi and Totsuka [18] 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 thus far. When the exchange coupling ratio $\alpha = J'/J$ is varied, first-order phase transition occurs from the dimer state to the new spin-gap state, while second-order phase transition occurs from this spin-gap state to the magnetically ordered state. This sheds light on the nature of quantum phase transitions in this model, and resolves apparently controversial conclusions on this issue.

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