Two-dimensional (2D) quantum spin systems with a spin gap have been the subject of considerable interest. In particular, the effect of the frustration in these systems plays an important role in stabilizing the disordered ground state. The 2D spin-gap compound SrCu$_2$(BO$_3$)$_2$ found recently$^1$ has strong frustration due to its orthogonal-dimer structure, which has provided a variety of interesting topics.$^2$–$^7$ The system can be described by the 2D Heisenberg model on a square lattice with some diagonal bonds, which is referred to as the Shastry-Sutherland model.$^8$ as reported by Miyahara and Ueda.$^9$ In this 2D model, most of the fundamental properties including the ground state as well as the excited states have been clarified by various theoretical approaches.$^8$–$^{21}$ On the other hand, some recent reports claim that the compound SrCu$_2$(BO$_3$)$_2$ has a large interlayer coupling, which may not be negligible in explaining the experimental findings.$^{10,11}$ Therefore it is necessary to study the phase diagram for the three-dimensional (3D) orthogonal-dimer system.

Motivated by the above findings, we investigate here the quantum phase transitions for the 3D orthogonal-dimer system which was introduced by Ueda and Miyahara.$^{12}$ The Hamiltonian is described by

$$H = J \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j + J' \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j + J'' \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j$$

where $\mathbf{S}_i$ is the $S = 1/2$ spin operator at the $i$-th site, and $J$, $J'$ and $J''$ are antiferromagnetic couplings. The structure of this model is shown schematically in Fig. 1. In some limiting cases, the 3D model is reduced to interesting orthogonal-dimer systems which have been studied intensively thus far. When $J'' = 0$, the system is the well-known Shastry-Sutherland model$^8$ (see Fig. 2), for which many groups have theoretically discussed various properties such as plateaus in the magnetization curve,$^9$–$^{17}$ two-magnon excitations,$^{11,17,18}$ and quantum phase transitions.$^9$–$^{11,16,19,21}$ In particular, the nontrivial disordered state adiabatically connected to isolated plaquettes$^{21}$ may be the most probable singlet state, which competes with the dimer state as well as the antiferromagnetically ordered state. In this paper, this phase is referred to as a "plaquette phase", for simplicity. On the other hand, in the case $J' = 0$, the system is reduced to the two-leg ladders with the diagonal bonds (see Fig. 4), which may be regarded as chains of the tetrahedra with shared edges, as pointed out by Gelfand.$^{22}$ In the ladder system, the dimer phase undergoes a first-order quantum phase transition to another disordered phase, which is exactly formed by the triplet ($S = 1$) on each rung. Note that the singlet on each rung is completely decoupled from the triplet for any value of $J''$, due to the orthogonal-dimer structure.$^{22}$–$^{26}$ Therefore, this disordered phase is equivalent to the Haldane phase realized in the $S = 1$ quantum Heisenberg chain$^{27}$–$^{33}$ Concerning the first-order transition, it is known that the cusp and jump singularities appear in the excitations$^{22,25}$ and the magnetization curve.$^{26}$

In this study, we discuss the phase diagram for the 3D system by means of the series expansion method.$^{34}$ We observe how the frustration affects the quantum phase transitions. It is also clarified that the spin-gap compound SrCu$_2$(BO$_3$)$_2$ is located in the vicinity of the phase boundary which separates the dimer and the magnetically ordered phases.

In the series expansion approach,$^{34,35}$ we start with
isolated clusters of a proper spin-singlet configuration. For this purpose, we divide the original Hamiltonian into two parts as $H = H_0 + \lambda H_1$, where $H_0(H_1)$ represents the unperturbed (perturbed) Hamiltonian and $\lambda$ is an auxiliary parameter. Details of the initial configurations for various phases are given in the following. By turning on the couplings among the clusters perturbatively ($H_1$), we carry out the series expansion in $\lambda$ for the ground state energy and the spin gap. Then the critical point for the first- (second-)order transition is determined by applying the first-order inhomogeneous differential method (Padé approximants) to the obtained series. In the following, starting with the Shastry-Sutherland model and the two-leg ladder model with diagonal bonds, we deal with quantum phase transitions for the 3D orthogonal-dimer model.

Note that the direct product of the dimer-singlet states formed by the couplings $J$ is always an eigenstate of the Hamiltonian (0.1) with the energy $E = -3/8 NJ$, even in the 3D model where $N$ is the total number of spins. We then discuss how the interlayer coupling on the 2D Shastry-Sutherland model ($J'' = 0$) affects the quantum phase transitions. We recall here the scenario in our previous paper where the first- (second-)order quantum phase transition in 2D was discussed by calculating the ground state energy (the spin gap) for the plaquette phase. Following this concept, we compute the quantities for the plaquette phase by means of the series expansion technique. We choose the arrangement of plaquettes shown in Fig. 2 as a starting configuration. In this figure, the shaded squares represent the plaquettes formed by the exchange coupling $J'$. The other bold and solid lines indicate the exchange couplings between the plaquettes, $\lambda J$ and $\lambda J''$, respectively. We also set the interlayer coupling as $\lambda J''$, which is not shown in this figure, for simplicity. To discuss the first-order quantum phase transition to the dimer phase, we perform the plaquette expansion for the ground state energy up to the fifth order in $\lambda$ for several values of $J'$ and $J''$. We estimate the ground state energy in the case $\lambda = 1$, where the model is reduced to the original orthogonal-dimer model by means of the first-order inhomogeneous differential method. Comparing the energies for the dimer and the plaquette phases, we determine the phase boundary shown as the bold line in Fig. 3, which separates two disordered phases. When $J'' = 0$ (Shastry-Sutherland model), the plaquette phase undergoes the first-order quantum phase transition at $(J'/J)_c = 0.68$. Since this critical value obtained by the fifth-order expansion is consistent with the result $(J'/J)_c = 0.677$ obtained by the seventh-order expansion, it is expected that this lower-order expansion with the first-order inhomogeneous differential method may give the phase boundary quantitatively well even in the 3D model. By introducing the interlayer coupling $J''$, the energy for the plaquette phase seems to decrease quadratically, while the energy for the dimer phase is not changed due to its orthogonal-dimer structure. This implies that the phase boundary between the two spin-gap phases is little affected as long as the interlayer couplings are small.

In order to completely discuss the interlayer effects, the antiferromagnetically ordered phase stabilized by the interlayer coupling must be properly taken into account. Performing the plaquette expansion for the spin gap up to the fourth order in $\lambda$, we determine the second-order quantum phase transition point by means of the biased Padé approximants with the critical exponent $\nu \sim 0.6$. The introduction of the interlayer couplings induces the antiferromagnetic correlation, while it suppresses the frustration which stabilizes the plaquette phase. The multi-critical point can be estimated as $(J'/J, J''/J) \sim (0.67, 0.08)$, and it is seen that the plaquette phase induced by the frustration is not stable against the interlayer coupling.

We next consider the present system as the coupled two-leg ladders $(J' \sim 0)$, as shown in Fig. 4. We discuss how the dimer and the Haldane phases compete with the magnetically ordered phase induced by

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Fig. 2. The initial configuration of the plaquettes used to treat the frustration-induced disordered phase by means of the series expansion technique.

Fig. 3. The phase diagram for the 3D orthogonal-dimer system. The bold and solid lines represent the phase boundaries where the first- and the second-order quantum phase transitions occur, respectively. The open circles indicate the locations of the orthogonal-dimer compound SrCu_2(BO_3)_2 obtained by Miyahara and Ueda and Knetter et al.

Fig. 4. The two-leg ladder with diagonal bonds, which may be considered to be a chain of tetrahedra with shared edges.
the inter-ladder coupling $J'$. Since the Haldane phase is formed by the triplet state ($S = 1$) on each rung, the dimer expansion starting from the singlet state ($S = 0$) is not appropriate to discuss the critical phenomena. Nevertheless, following the concept of the valence bond solid, we can choose a suitable initial configuration for the Haldane phase in the framework of the series expansion. Namely, we introduce the unperturbed Hamiltonian $H_0$, by choosing the initial configuration of isolated plaquettes indicated by the shaded rectangles in Fig. 4. Considering the other couplings as perturbations, we then discuss the stability of the Haldane phase.

To study the first- (second-)order transition, we perform the plaquette expansion for the ground state energy (the spin gap) up to the sixth (fifth) order in $\lambda$. Using the approximation discussed above, we then determine the phase boundaries shown in Fig. 3. When $J' = 0$, the first-order quantum phase transition from the Haldane phase to the dimer phase occurs at the critical value $(J''/J)_{c} \sim 0.71$, which is in good agreement with the accurate value $(J''/J)_{c} = 0.7135(3)$ obtained by Gelfand. Introducing the interladder coupling $J''$ in the Haldane phase $(J''/J > 0.71)$, the antiferromagnetic correlation is enhanced and the spin gap decreases rapidly. Finally, the second-order quantum phase transition occurs to the magnetically ordered phase. We estimate the multicritical point $(J'/J, J''/J) \sim (0.015, 0.7)$ where the dimer, the Haldane and the magnetically ordered phases compete with each other.

Finally, to discuss the competition between the dimer and the antiferromagnetically ordered phases, we use the Ising expansion technique. To this end, we divide the original Heisenberg Hamiltonian into two parts as $H = \sum J_{ij} S_{i}^{x} S_{j}^{x} + \lambda \sum J_{ij}(S_{i}^{x} S_{j}^{y} + S_{i}^{y} S_{j}^{x})$, where $J_{ij}$ is the exchange coupling for each bond. The ground state energy $E_{AF}$ for the antiferromagnetically ordered phase is given by

$$E_{AF} = \frac{1}{2} NJS^2 - 2NJ'S^2 - 2NJ''S^2 + \epsilon(\lambda), \quad (0.2)$$

where $\epsilon(\lambda)$ is the quantum correction due to the $XY$ components. Performing the Ising expansion for $\epsilon(\lambda)$ up to the fourth order in $\lambda$ and applying the Padé approximants to the obtained series, we then deduce the energy $E_{AF}$. By comparing it with the energy for the dimer phase, we can determine the phase boundary, which is shown as the bold line in Fig. 3. Near the multicritical points, the energy $E_{AF}$ has not been deduced accurately, and thereby the phase boundary shown as the broken line is to guide the eyes. When $J'' = 0$, the 3D orthogonal-dimer system belongs to the dimer phase with the exact dimer-singlet state. Note that in the dimer phase the interladder coupling $J''$ has little effect on the ground state properties due to its orthogonal-dimer structure, which is quite different from the case of the coupling $J'$. Therefore the introduction of the interdimer couplings $J'$ and $J''$ enhances the 2D spin-spin correlation, and finally induces the first-order quantum phase transition to the antiferromagnetically ordered phase with the 3D structure, as seen in Fig. 3.

Recently it was clarified that the compound SrCu$_2$(BO$_3$)$_2$ has a large interlayer coupling. In the reports by Miyahara and Ueda and Knetter et al., the ratios of the interdimer and interlayer exchange couplings are estimated as $(J'/J, J''/J) = (0.635, 0.09)$ and $(0.603, 0.21)$, which are indicated by open circles in Fig. 3. It is found that the compound SrCu$_2$(BO$_3$)$_2$ may be located in the dimer phase close to the phase boundary.

Some comments are in order for the possibility of another phase induced by the frustration in the ground state. Recently Knetter et al. have found that the instability of the two-magnon excitation in the Shastry-Sutherland model $(J'' = 0)$ exists in the dimer phase. It is also claimed that the first-order quantum phase transition occurs to the classical helical ordered phase at the critical point $(J'/J)_{c} = 0.630(5)$. This result appears to contrast with the other results except for those obtained by the Schwinger boson approach.

Although their conclusion of the first-order transition in terms of the analysis of elementary excitations is speculative, it is an interesting open problem to clarify what this instability really implies, which is now under consideration.

In summary, we have investigated the quantum phase transitions in the 3D orthogonal-dimer system for the compound SrCu$_2$(BO$_3$)$_2$ by means of the plaquette expansion technique. We have thus determined a phase diagram with a rich structure, in which it is clarified that the frustration-induced disordered phase is unstable against the interlayer couplings. Also it is found that the compound SrCu$_2$(BO$_3$)$_2$ is located in the dimer phase close to the phase boundary.

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