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Quantum phase transitions for the Haldane system in higher dimensions: A mixed-spin cluster expansion approach

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We present a mixed-spin cluster expansion method to discuss the quantum phase transitions for the Haldane system in two and three dimensions. By mapping the $s=1$ antiferromagnetic spin model on square and cubic lattices to the equivalent mixed-spin model, we study the competition among the Haldane, the dimer, and the magnetically ordered phases. The mixed-spin cluster expansion proposed here realizes the notion of the valence bond solid in a perturbation theory. This method allows us to directly deal with the Haldane phase, which may not be reached by standard series expansion methods. The zero-temperature phase diagram is thus determined rather precisely for the two- and three-dimensional Haldane systems.

I. INTRODUCTION

Low-dimensional spin systems with the spin gap for the excitation spectrum have been extensively studied since the Haldane conjecture,[1] which clarified that the gap formation in the integer-spin Heisenberg chain reflects the topological nature of spins. Recent extensive experimental and theoretical investigations on the stability of the Haldane system against various perturbations have been providing a variety of interesting topics. The instability of the spin-gap phase in the $s=1$ spin models has been studied in detail so far for one-dimensional (1D) systems. For instance, the effect of the bond alternation is understood qualitatively well by the nonlinear sigma model,[2] as well as the valence bond solid (VBS) approach.[3] The accurate critical point between the dimer and the Haldane phases has been further obtained by the series expansion,[4] the exact diagonalization,[5,6] the quantum Monte Carlo simulations,[7] and the density matrix renormalization group (DMRG).[5] On the other hand, the $s=1$ spin systems with the 2D or 3D structures have not been studied so well, although the effects of the antiferromagnetic correlations due to the interchain couplings should be important for real materials. So far, Sakai and Takahashi[8] investigated a quasi-1D $s=1$ spin system by combining the mean field theory with the exact diagonalization results for the spin chain, and gave a rough estimate for the phase-transition point to the antiferromagnetic phase.

In this paper, we systematically study how the Haldane and the dimer phases for the $s=1$ antiferromagnetic chain are driven to the magnetically ordered phase in 2D and 3D systems by exploiting the series expansion techniques. In particular, we propose a *mixed-spin cluster expansion* by mapping the $s=1$ spin model to the equivalent mixed-spin model, which allows us to deal with the Haldane phase. This approach is a realization of the notion of the VBS in a perturbation theory. We determine the phase diagram rather precisely both for the 2D and 3D cases by computing the spin excitation gap and the staggered susceptibility.

II. MODEL HAMILTONIAN

Let us first consider the $s=1$ antiferromagnetic quantum spin system on a 2D square lattice, which is described by the Hamiltonian

$$H=\sum_{i,j}\,[\Gamma_i \mathbf{S}_{i,j}\cdot\mathbf{S}_{i+1,j}+J\mathbf{S}_{i,j}\cdot\mathbf{S}_{i,j+1}], \tag{1}$$

where J is the interchain coupling and $\mathbf{S}_{i,j}$ is the $s=1$ operator at the (i,j)th site in the $(x-y)$ plane. Here we have introduced the bond-alternation parameter $\alpha(0\leqslant\alpha\leqslant1)$ along the x direction, $\Gamma_i=1(\alpha)$ for even (odd) i. All the exchange couplings are assumed to be antiferromagnetic.

We employ the series expansion method developed by Singh, Gelfand, and Huse.[9,10] Since this method combines the conventional perturbation theory with the cluster expansion, it has an advantage to deal with the spin system in higher dimensions even for the cases for which the reliable results are difficult to be obtained by the exact diagonalization, the DMRG, etc. In fact, the series expansion method has been successfully applied to the 2D spin systems with various structures,[9,11] Kondo lattice,[12] bilayer systems,[13,14] etc. However, to apply the series expansion technique to the present system including the Haldane phase, a nontrivial generalization is needed, since a naive cluster expansion may not describe the Haldane state. For instance, the dimer state is adiabatically connected to the isolated $s=1$ dimers, but the Haldane state does not have its analogue in the isolated local singlets composed of several $s=1$ spins. To overcome this problem, we wish to recall the notion of the VBS,[3] which captures the essence of the Haldane-gap formation. To realize this idea in the series expansion, we first divide half of the $s=1$ spins into two $s=1/2$ spins as schematically shown in Fig. 1,[15] and map the system to the mixed-spin system which is equivalent to the original model except for a trivial isolated excited mode. As a starting configuration in the perturbative expansion, we can then consider two types of the mixed-spin cluster singlets formed by the solid lines in Figs. 1(b) and 1(c). It is seen that by starting from the configuration (b) we can directly deal with the Haldane phase since it has the structure of the Haldane state in the VBS picture, whereas if the configuration (c) is chosen, we naturally end up with the standard dimer expansion. The above mapping thus gives us an important message that *the Haldane phase is adiabatically connected to the isolated mixed-spin singlet states in Fig. 1(b), and thereby can be*

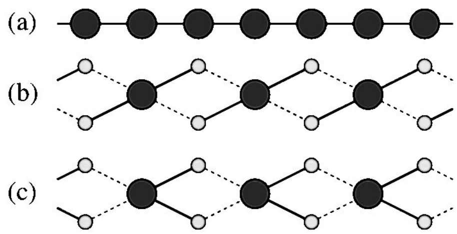

FIG. 1. (a) The $s=1$ spin chain, which is decomposed into (b) the mixed-spin chain in the Haldane phase and into (c) that in the dimer phase. Large and small solid circles represent the $s=1$ spin and $s=1/2$ spins, respectively. The solid lines in (b) and (c) indicate the strong bonds which make local singlets whereas the dashed lines the weak bonds which are treated perturbatively.

treated by the mixed-spin cluster expansion method. The resulting cluster expansion around the isolated mixed-spin singlets should provide a quite powerful method, which enables us to deal with the competition among the Haldane phase, the dimer phase and the magnetically ordered phase in 2D and 3D systems.

III. QUANTUM PHASE TRANSITIONS

We first discuss the quantum phase transitions among the Haldane phase, the dimer phase and the antiferromagnetic phase for the 2D $s=1$ spin system with bond alternation. In the end of this section, we further apply the cluster expansion method to the 3D $s=1$ Haldane system.

A. Haldane phase

In order to apply the mixed-spin cluster expansion to the Haldane phase in 2D, we first convert the $s=1$ spin model into the effective mixed-spin model shown in Fig. 2. In this figure, the large (small) circle represents the $s=1(s=1/2)$ spin. The bold solid, the thin solid and the dashed lines indicate the coupling constant 1,λ and $J\lambda$, respectively. Here an auxiliary parameter λ, which changes from 0 to 1, is introduced to perform the cluster expansion. In this figure, the model without bond alternation is drawn for simplicity. We note that the mixed-spin system reproduces the original 2D spin system at $\lambda=1$. To perform the cluster expansion, the Hamiltonian is first divided into two parts as $H=H_0+\lambda H_1$, where

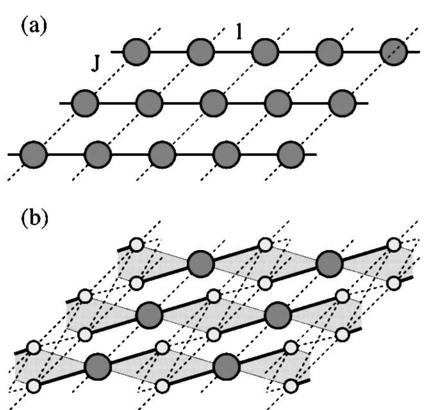

FIG. 2. (a) The 2D $s=1$ spin model and (b) the corresponding mixed-spin system.

$$H_0=\sum_{i,j}\left[\mathbf{s}^{(1)}_{2i,j}\cdot\mathbf{S}_{2i+1,j}+\alpha\mathbf{S}_{2i+1,j}\cdot\mathbf{s}^{(2)}_{2i+2,j}\right], \tag{2}$$

$$H_1=\sum_{i,j}\left[\mathbf{s}^{(2)}_{2i,j}\cdot\mathbf{S}_{2i+1,j}+\alpha\mathbf{S}_{2i+1,j}\cdot\mathbf{s}^{(1)}_{2i+2,j}+J\left(\mathbf{S}_{2i+1,j}\cdot\mathbf{S}_{2i+1,j+1}+\sum_{m,n}\mathbf{s}^{(m)}_{2i,j}\cdot\mathbf{s}^{(n)}_{2i,j+1}\right)\right], \tag{3}$$

where $\mathbf{s}^{(1)}(\mathbf{s}^{(2)})$ is the $s=1/2$ operator which represents one of the decomposed spins, and $m(n)$ runs from 1 to 2. The first term H_0 is the unperturbed Hamiltonian which stabilizes the isolated mixed-spin cluster singlets. The corresponding mixed-spin cluster has the configuration, 1/2∘1∘1/2, which is formed by the antiferromagnetic couplings 1 and α. These isolated clusters have the singlet ground state with the spin gap $\Delta=(3\alpha+3-\sqrt{9-14\alpha+9\alpha^2})/4$. The perturbed part H_1 of the Hamiltonian labeled by λ connects these isolated mixed-spin singlets to form a 2D network and thus enhances the antiferromagnetic correlation. We compute the staggered susceptibility $\chi_{\rm AF}$, and the singlet-triplet excitation gap Δ at the ordering wave vector. These quantities are then expanded as a power series in λ. We finally determine the phase boundary by the divergent staggered susceptibility and the vanishing spin gap, which are estimated by applying the Padé approximants[16] to the quantities obtained up to the finite order in λ.

To confirm how well our mixed-spin cluster approach works, we first investigate the $s=1$ spin chain without bond alternation. Performing the mixed-spin cluster expansion, we calculate the ground state energy E_g, the staggered susceptibility $\chi_{\rm AF}$ and the singlet-triplet excitation gap Δ up to the eleventh, the fifth and the seventh order, respectively. At first sight, the order in the series for the staggered susceptibility and the excitation gap might not be high enough to produce the accurate values at $\lambda=1$ (the Haldane point) by means of the ordinary differential methods.[16] It is remarkable, however, that there exists an additional symmetry property like $Q(\lambda)=\lambda Q(1/\lambda)$ for each physical quantity Q in our effective mixed-spin chain. This symmetry relation follows from the invariance under interchanging the solid line and the broken line in Fig. 1(b). This enables us to expand the quantity Q as a power series even around $\lambda=1$ as $Q(\lambda)=\Sigma Q_n(\lambda-1)^n$, where the symmetry property described above yields the following relations:

$$Q_1=\frac{1}{2}Q_0,\quad Q_3=-\frac{1}{2}Q_2,\quad Q_5=\frac{1}{4}Q_2-\frac{3}{2}Q_4,\ \dots\ . \tag{4}$$

Fitting this power series with that obtained by the cluster expansion, we calculate Q_0 and thus obtain the rather accurate values, $E_g=-1.4022$, $\chi_{\rm AF}=19.6$ and $\Delta=0.404$, which are compared with those of the Monte Carlo simulations: $E_g=-1.4015\pm0.0005$, $\Delta=0.41$ in Ref. 17, and also the exact diagonalization $\Delta=0.411\pm0.001$, $\chi_{\rm AF}=18.4\pm1.3$ in Ref. 18.

To observe the quantum phase transition on a 2D lattice by increasing the interchain couplings, we evaluate the singlet-triplet excitation gap Δ by means of the mixed-spin cluster expansion up to the fifth order in λ for various choices of α and J.

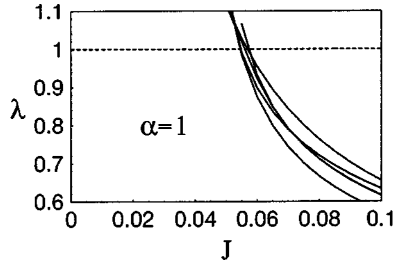

FIG. 3. Phase boundaries obtained for 2D mixed-spin system without bond alternation ($\alpha=1$). The solid lines indicate the boundaries obtained by different kinds of the Dlog Padé approximants ([1/1], [1/2], [1/3], [2/1], [2/2] approximants), where the left (right) side of the boundary is in the Haldane (magnetically ordered) phase. Note that only when $\lambda=1$ (dashed line), this mixed-spin model is reduced to the original Haldane system.

Taking the isotropic case ($\alpha=1$) as an example, we briefly explain how to determine the phase boundary between the Haldane and the magnetically ordered phases from the power series obtained. By applying several different kinds of Dlog Padé approximants we first deduce the phase boundaries in ($J-\lambda$) plane for the mixed-spin system, as shown in Fig. 3. Recall that this mixed-spin system reproduces the original Haldane system only in the case $\lambda=1$ (dashed line). By exploiting the values of J for which the phase boundary intersects the $\lambda=1$ line in Fig. 3, we obtain the critical value $J_c=0.056\pm0.001$ for the phase transition point in the original Haldane system. Our results for $\alpha=1$ are much more accurate than those of the mean field theory combined with the exact diagonalization,[8] which claimed the critical value to be $J_c>0.025$.

We note here that the obtained critical exponent $\nu=1.86\pm0.08$ is different from the value $\nu=0.71$[19] expected for the 3D classical Heisenberg model.[20] This does not imply that the universality class of the quantum phase transition for the original Haldane system is different from that of the 3D classical Heisenberg model. In our approach, by dividing the $s=1$ spin into two $s=1/2$ spins and further introducing an *auxiliary parameter* λ, we convert the original 2D $s=1$ model into the effective mixed-spin model on a complicated lattice structure. This model reproduces the original $s=1$ Haldane model only for the specific value of $\lambda=1$, but for general couplings of λ, it does not have any correspondence to the $s=1$ spin model. It should be noted that an unusual exponent $\nu=1.86\pm0.08$ shows up for the phase transition when we change this *auxiliary parameter* λ rather than the original couplings J and α. Therefore, an apparently unusual exponent we have encountered comes from the nontrivial decomposition into the mixed-spin system, which does not contradict the well-known result that the quantum phase transition for the 2D $s=1$ system belongs to the 3D classical Heisenberg model.[20]

By the similar procedure, we determine the phase boundary shown by the dots with error bars in Fig. 4. The error bars come from the different values obtained by different biased Padé approximants employed: [1/2], [2/1], [2/2], [2/3], [3/2] approximants. Since the error bars increase with the decrease of α away from unity, it seems difficult to determine the phase boundary in the region close to the dimer phase. However, it is to be noted that this phase diagram

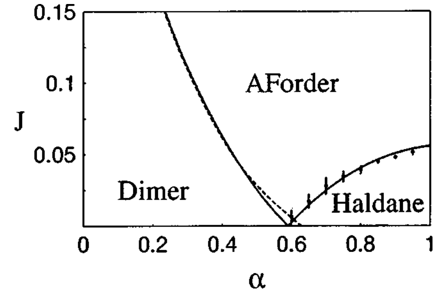

FIG. 4. Phase diagram for the 2D $s=1$ quantum spin system with bond alternation α. The phase boundary between the Haldane phase and the ordered phase is determined by the mixed-spin cluster expansion. The left solid (the left dashed) phase boundary around the dimer phase is determined by the dimer expansion with the biased [2/3] Padé approximants for the excitation gap (the staggered susceptibility).

should have the symmetry property as $J(\alpha)=\alpha J(1/\alpha)$. Taking this into account, we can thus determine rather precisely the phase boundary between the Haldane phase and the antiferromagnetic phase, which is drawn by the solid line in Fig. 4. We shall see momentarily that the critical point between the dimer and the Haldane phases determined in this procedure is consistent with that obtained by the dimer expansion.

B. Dimer phase

Let us now turn to the dimer phase. In this case, our mixed-spin cluster expansion is equivalent to the standard dimer expansion.[10] We perform the dimer expansion of the staggered susceptibility and the spin gap up to the fifth and the sixth order in λ for various J, respectively. To estimate the phase boundary which separates the dimer phase and the antiferromagnetic phase, we use the ordinary Padé approximants[16] as well as the biased Padé approximants, for which the phase transition is assumed to belong to the universality class of the 3D classical Heisenberg model.[20] Using these Padé approximants, we arrive at the phase diagram shown in Fig. 4. When $J=0$ with small α, the system is reduced to the isolated $s=1$ bond-alternating chain, which is known to have disordered ground state with the spin gap due to the dimer singlet. Increasing the parameter J and α, the antiferromagnetic correlation grows up, and the quantum phase transition to the magnetically ordered state occurs. We wish to note that the critical point $(\alpha,J)=(0.59,0)$, which is determined from the series expansion of the spin gap, separates the Haldane phase, the dimer phase and the antiferromagnetically ordered phase in Fig. 4. Since the system in this case is reduced to the independent $s=1$ spin chains with bond alternation, our numerical results reproduce the well-known fact[4–7] that the ground state of the reduced chain with $\alpha_c=0.59$ is in a critical phase with neither the spin gap nor the long-range order. To confirm how accurate our results for 2D cases are, we have directly analyzed the spin chain ($J=0$) by applying the Dlog Padé approximants to the spin gap computed up to the eighth order. This gives $\alpha_c=0.612\pm0.004$, which is close to the value 0.59 obtained above, and also to 0.60 ± 0.01 obtained by DMRG.[5] Judging from these results, we can say that our phase boundary determined by

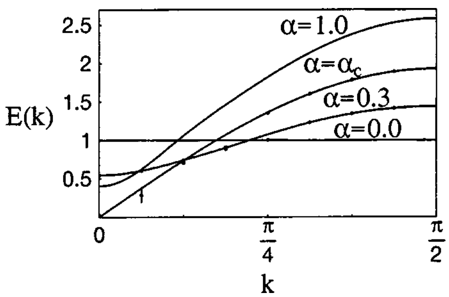

FIG. 5. Dispersion relations of the spin-triplet excited states for the $s=1$ chain with bond alternation α. The results for $\alpha=0{,}0.3$ and $\alpha_c(=0.59)$ are obtained by the dimer expansion, while the other for the Haldane phase is obtained by the mixed-spin cluster expansion with the help of a symmetry property (see the text).

the excitation gap in Fig. 4 is quite accurate, while that by the staggered susceptibility has a slight deviation only around the critical point.

C. Dispersion relation

In order to demonstrate that our approach is also powerful to compute the elementary excitation with finite momenta, we show the calculated dispersion relation in Fig. 5 along the specific line of $J=0$. Reflecting the isolated spin-singlet structure, the Brillouin zone becomes half of the original one. In the dimer phase $0<\alpha<\alpha_c$, using the first order inhomogeneous differential method,[16] we can obtain the dispersion relation. Here, to obtain the dispersion for the Haldane phase, we have again made use of the additional symmetry property inherent in the effective mixed-spin model mentioned above. It is to be emphasized that such a precise dispersion is obtained within the lower-order perturbations, which is indeed due to the additional symmetry we have used.

D. Phase diagram for the 3D case

We now move to the 3D system. The advantage of our approach is particularly remarkable for the 3D problem because other numerical methods may often meet some difficulties to treat a large system in the 3D case. We consider here a cubic lattice system by adding the interchain couplings J in the z direction to the spin model discussed above. By extending our treatment to the 3D system, we thus study the competition between the two kind of gapped states and the antiferromagnetic state. Applying the dimer expansion to calculate the spin gap and the staggered susceptibility up to the fifth order and using the Dlog [2/2] Padé approximants, we first determine the phase boundary which separates the dimer and the antiferromagnetic ordered phase in Fig. 6. When $\alpha=0$, our system reproduces the $s=1$ bilayer Heisenberg model. Increasing the interdimer coupling J from zero, the antiferromagnetic correlation grows up and the quantum phase transition occurs at $J_c=0.143\pm0.006$. We note that the quantum phase transitions in the bilayer model have already been studied by Gelfand *et al.*[14] with the series expansion method. On the other hand, to observe the phase transition from the Haldane phase to the ordered phase, we further perform the mixed-spin cluster expansion up to the fourth order for both of the above quantities. In the homogeneous case ($\alpha=1$), by analyzing the data in terms of various Dlog Padé approximants we end up with the critical point $J_c=0.026\pm0.001$, which is consistent with those of the nonlinear σ model approach[21] and the mean field theory combined with the numerical method.[8] The phase diagram thus determined is shown in Fig. 6.

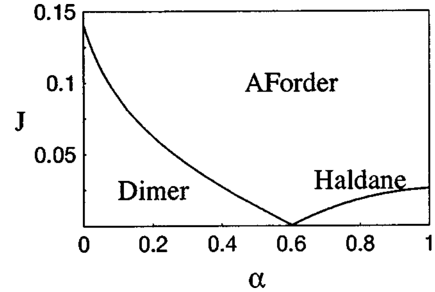

FIG. 6. Phase diagram for the 3D $s=1$ spin system with bond alternation α.

IV. SUMMARY

We have investigated the quantum phase transitions for the $s=1$ quantum systems with the 2D and 3D structures. Using the series expansion, we have discussed how the dimer phase and the Haldane phase realized in 1D compete with the magnetically ordered phase in higher dimensions. In particular, we have proposed an approach based on the mixed-spin cluster expansion which realizes the idea of the VBS in the perturbation theory. This approach has made it possible to treat the Haldane phase in the series expansion framework, which was not dealt with so far by ordinary series expansion methods. For the spin chain case, we have obtained fairly good results comparable to other numerical methods.[17,18] For the 2D and 3D cases, the phase diagram has been determined rather precisely by making use of an additional symmetry property in the effective mixed-spin model. It is quite interesting to further apply the mixed-spin cluster approach to the frustrated case, the anisotropic case, etc., in quasi-1D Haldane systems, which is now under consideration.

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