Dimer order with striped correlations in the J_1-J_2 Heisenberg model

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Ground-state energies for plaquette and dimer order in the J_1-J_2 square-lattice spin-half Heisenberg model are compared using series expansion methods. We find that these energies are remarkably close to each other at intermediate values of J_2/J_1, where the model is believed to have a quantum disordered ground state. They join smoothly with those obtained from the Ising expansions for the two-sublattice Néel state at $J_2/J_1 \approx 0.4$, suggesting a second-order transition from a Néel state to a quantum disordered state, whereas they cross the energy for the four-sublattice ordered state at $J_2/J_1 \approx 0.6$ at a large angle, implying a first-order transition to the four-sublattice magnetic state. The strongest evidence that the plaquette phase is *not* realized in this model comes from the analysis of the series for the singlet and triplet excitation spectra, which suggest an instability in the plaquette phase. Thus our study supports the recent work of Kotov *et al.*, which presents a strong picture for columnar dimer order in this model. We also discuss the striped nature of spin correlations in this phase, with substantial resonance all along columns of dimers. [S0163-1829(99)06433-4]

I. INTRODUCTION

There has been considerable study, over the last decade, of the frustrated spin-$\frac{1}{2}$ square lattice Heisenberg antiferromagnet (the ''J_1-J_2 antiferromagnet''). These studies include exact diagonalizations on small systems,[1–4] spin-wave calculations,[5,6] series expansions,[7,8] and a field-theoretic large-N expansion.[9]

These studies, and others, have provided a substantial body of evidence that the ground state of this system, in the region $0.4 \lesssim J_2/J_1 \lesssim 0.6$, has no long-range magnetic order and has a gap to spin excitations. For $J_2/J_1 \lesssim 0.4$ the model has conventional antiferromagnetic Néel order whereas for $J_2/J_1 \gtrsim 0.6$ the system orders in a columnar $(\pi, 0)$ phase. Whether this ''intermediate phase'' is a spatially homogeneous spin liquid, or whether it has some type of spontaneously broken symmetry leading to a more subtle type of long-range order, has not been conclusively established.

Zhitomirsky and Ueda[10] have proposed a plaquette resonating valence bond (RVB) phase, which breaks translational symmetry along both x and y axes, but preserves the symmetry of interchange of the two axes. The horizontal and vertical dimers resonate within a plaquette. An early series study[11] had investigated the relative stability of various spontaneously dimerized states and had concluded that a columnar dimerized phase was the most promising candidate for the intermediate region, in agreement with the large-N expansions. Zhitomirsky and Ueda[10] claim their plaquette phase has a lower energy than this columnar dimer phase, but we find this to be incorrect.

Further support for the columnar dimer scenario comes from recent work of Kotov *et al.*,[12] who combine an analytic many-body theory with extended series and diagonalization results to study the nature and stability of the excitations in the intermediate region. It is argued that where the Néel phase becomes unstable the system will develop not only a gap for triplet excitations but also a gapped low-energy singlet which reflects the spontaneous symmetry breaking. This is clearly seen in the calculations. At $J_2/J_1 \simeq 0.38$ a second-order transition occurs, with the energies of Néel phase and dimerized phase joining smoothly, and the energy gap and dimerization vanishing.

It is the aim of this paper to further investigate, using series methods, the competing possibilities of columnar dimerization versus plaquette order in the intermediate region of the J_1-J_2 antiferromagnet. It is conceivable that both occur, with a transition from one to the other. However, such a transition, reflecting a change of symmetry, is expected to be first order and not well suited to series methods. If both phases are locally stable the most direct way to compare them is by comparison of the ground-state energies. If one is unstable this should show up by the closing of an appropriate gap or by the divergence of an appropriate susceptibility. In this paper we calculate the ground-state energy and the singlet and triplet excitation spectra by series expansions about a disconnected plaquette Hamiltonian. We also calculate the susceptibility for the dimer phase to break translational symmetry in the direction perpendicular to the dimers. This susceptibility will be large if there is substantial resonance in the dimer phase and will diverge if there is an instability to the plaquette RVB phase.

Combining the plaquette expansion results with the dimer expansions of Kotov *et al.*,[12] a very interesting picture emerges for the quantum disordered phase. We find that the plaquette phase is unstable and hence is not the ground state for this model. The dimer phase, on the other hand, is stable. However, there is substantial resonance in the dimer phase. The spin-spin correlations are not simply those of isolated dimers. Instead, the nearest-neighbor correlations are nearly identical along the rungs and chains of dimer columns. In contrast, the correlations from one dimer column to the next are much weaker. The spin-gap phase appears separated

from the Néel phase by a second-order transition, whereas it is separated from the columnar phase by a first-order transition. These results are in remarkable agreement with the large-N theories.[9] The existence of a quantum critical point separating an antiferromagnetic phase and a quantum disordered phase with striped correlations in a microscopic model makes this critical point a particularly interesting one. The role of doping and its implications for high-T_c materials deserves further attention.

II. SERIES EXPANSIONS AND RESULTS

We study the Hamiltonian

$$H = J_1 \sum_{NN} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{NNN} \mathbf{S}_i \cdot \mathbf{S}_j , \tag{1}$$

where the first sum runs over the nearest neighbor and the second over the second-nearest neighbor spin pairs of the square lattice. We denote the ratio of couplings as $y = J_2/J_1$. The linked-cluster expansion method has been previously reviewed in several articles,[13–15] and will not be repeated here. To carry out the series expansion about the disconnected-plaquette state for this system, we take the interactions denoted by the thick solid and dashed bonds in Fig. 1 as the unperturbed Hamiltonian, and the rest of the interactions as a perturbation. That is, we define the following Hamiltonian:

$$H = H_0 + H_1 , \tag{2}$$

where the unperturbed Hamiltonian (H_0) and perturbation (H_1) are

$$H_0 = J_1 \sum_{\langle ij \rangle \in A} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle ij \rangle \in B} \mathbf{S}_i \cdot \mathbf{S}_j ,$$

$$H_1 = \lambda J_1 \sum_{\langle ij \rangle \in C} \mathbf{S}_i \cdot \mathbf{S}_j + \lambda J_2 \sum_{\langle ij \rangle \in D} \mathbf{S}_i \cdot \mathbf{S}_j , \tag{3}$$

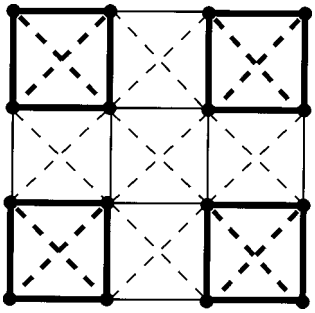

FIG. 1. The generalized J_1-J_2 Heisenberg model with plaquette structure, the couplings J_1, J_2, xJ_1, and xJ_2 indicated by thick solid bonds, thick dashed bonds, thin solid bonds, and thin dashed bonds, respectively.

and the summations are over intraplaquette nearest-neighbor bonds (A), intraplaquette second nearest-neighbor bonds (B), interplaquette nearest-neighbor bonds (C), interplaquette second-nearest-neighbor bonds (D), shown in Fig. 1. With this Hamiltonian, one can carry out an expansion in powers of λ, and at $\lambda = 1$ one recovers the original Hamiltonian in Eq. (1). Thus, although we expand about a particular state, i.e., a plaquette state, our results at $\lambda = 1$ describe the original system without broken symmetries, provided no intervening singularity is present. Such perturbation expansions about an unperturbed plaquette Hamiltonian have been used previously to study Heisenberg models for CaV_4O_9.[16]

It is instructive to consider the states of an isolated plaquette. There are two singlet states, one with energy $(-2 + y/2)J_1$ and the other with energy $(-3y/2)J_1$. The former is the ground state for $y < 1$ and corresponds to pair singlets resonating between the vertical and horizontal bonds of the plaquette. It is even under a $\pi/2$ rotation. The latter is the ground state for $y > 1$ and is odd under a $\pi/2$ rotation. The wave functions for these two singlet states are

$$\psi_1 = \frac{1}{\sqrt{12}}\left[\begin{pmatrix} + & + \\ - & - \end{pmatrix} + \begin{pmatrix} + & - \\ + & - \end{pmatrix} + \begin{pmatrix} - & - \\ + & + \end{pmatrix} + \begin{pmatrix} - & + \\ - & + \end{pmatrix} - 2\begin{pmatrix} + & - \\ - & + \end{pmatrix} - 2\begin{pmatrix} - & + \\ + & - \end{pmatrix}\right]$$

$$= \frac{1}{\sqrt{3}}\left[\left(\begin{array}{c} \circ\!-\!\circ \\ \circ\!-\!\circ \end{array}\right) + \left(\begin{array}{cc} \circ & \circ \\ \circ & \circ \end{array}\right)\right] \tag{4}$$

$$\psi_2 = \frac{1}{2}\left[\begin{pmatrix} + & + \\ - & - \end{pmatrix} - \begin{pmatrix} + & - \\ + & - \end{pmatrix} + \begin{pmatrix} - & - \\ + & + \end{pmatrix} - \begin{pmatrix} - & + \\ - & + \end{pmatrix}\right]$$

$$= \left[\left(\begin{array}{cc} \circ & \circ \\ \circ & \circ \end{array}\right) - \left(\begin{array}{c} \circ\!-\!\circ \\ \circ\!-\!\circ \end{array}\right)\right] \tag{5}$$

where $\circ$–$\circ$ means these two spins form a singlet. There are three triplet states, one with energy $(-1 + y/2)J_1$ and a degenerate pair with energy $(-y/2)J_1$; like the singlets, these have a level crossing at $y = 1$. Under a $\pi/2$ rotation the former is odd, while the latter two are even and odd, respectively. Finally there is a quintuplet state at $(1 + y/2)J_1$, which is even under a $\pi/2$ rotation. For $y < 1/2$ and $y > 2$ the first excited state of the plaquette is a triplet, while for $1/2 < y < 2$ it is the other singlet. These states and corresponding energies are shown in Fig. 2. The eigenstates of H_0, the unperturbed Hamiltonian, are direct products of these plaquette states.

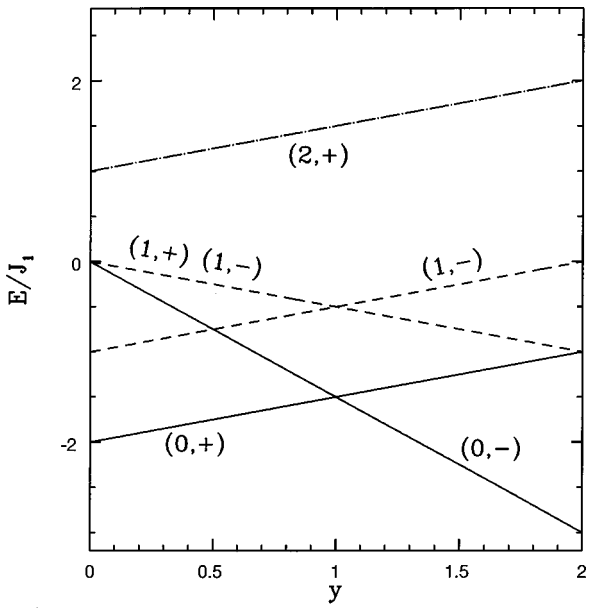

FIG. 2. The energies of an isolated plaquette as function of y. The notation presents the S value and effect of $\pi/2$ rotation ($\pm$) for each state.

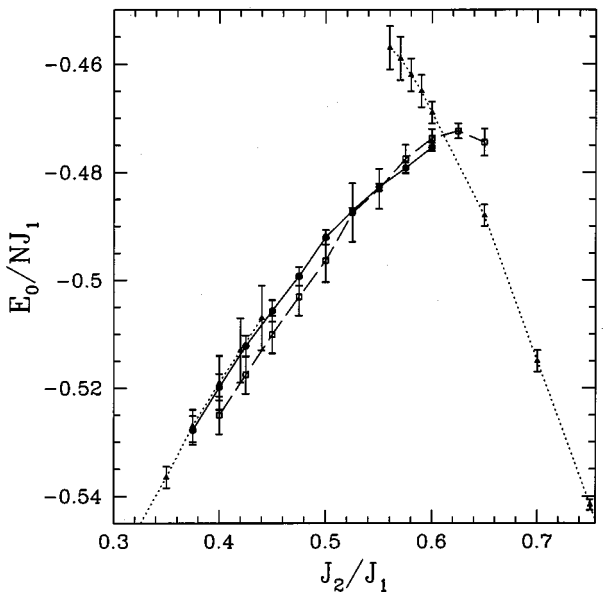

FIG. 3. The ground-state energy per site E_0/NJ_1 as function of J_2/J_1, obtained from the Ising expansions[8] (triangle points connected by dotted lines), the columnar dimer expansions[12] (open square points connected by dashed line), and the plaquette expansion (full circle points connected by solid line).

To derive the plaquette expansions we identify each plaquette as a 16 state quantum object, and these lie at the sites of a square lattice with spacing $2a$, where a is the original lattice spacing. Interactions between plaquettes connect first- and second-neighbor sites on this new lattice. The cluster data is thus identical to that used by us previously[8] to derive Ising expansions for this model. Because there are 16 states at each cluster site, the vector space grows very rapidly with the number of sites and thus limits the maximum attainable order for plaquette expansions to considerably less than can be achieved for dimer or Ising expansions.

We have computed the ground-state energy E_0 to order λ,[7] for fixed values of the coupling ratio y. The series are analyzed using integrated differential approximants,[17] evaluated at $\lambda=1$ to give the ground-state energy of the original Hamiltonian. The estimates with error bars representing confidence limits, are shown in Fig. 3. For comparison we also show previous results obtained from Ising expansions[8] and dimer expansions.[12] We find that, in the intermediate region, the ground-state energy for both plaquette and dimer phases are very close to each other and cannot be used to distinguish between them. The dimer expansion yields slightly lower energies near the transition to the Néel phase. We do not draw any conclusions from this.

Zhitomirsky and Ueda[10] have claimed that the ground-state energy from a second-order plaquette expansion is -0.63 (at $y=0.5$), much lower than the dimer expansion result -0.492. This result appears incorrect. At $J_2/J_1=\frac{1}{2}$ the ground-state energy is given by

$$\begin{aligned}4E_0/NJ_1 = &-7/4-277\lambda^2/1440-0.001\,357\lambda^3\\ &-0.021\,060\,9\lambda^4-0.000\,319\,586\lambda^5\\ &-0.005\,806\,43\lambda^6-0.001\,822\,686\lambda^7+O(\lambda^8).\end{aligned} \tag{6}$$

The second-order result (at $\lambda=1$) is $E_0/NJ_1=-0.485$, rather than -0.63. We note that if the second-order coefficient were four times larger then the resulting energy would be $-0.629\,86$.

We have also derived series, to order λ^6, for the singlet and triplet excitation energies, $\Delta_s(k_x,k_y)$, $\Delta_t(k_x,k_y)$ using the method of Gelfand,[14] and taking as unperturbed eigenfunctions the corresponding plaquette states. The low order terms for $J_2/J_1=0.5$ are given by

$$\begin{aligned}\Delta_s(k_x,k_y)/J_1 = &\,1-301\lambda^2/1440+137\lambda^3/86\,400\\ &+217\lambda^3\cos(k_x)\cos(k_y)/172\,800\\ &+(-5\lambda^2/16-89\lambda^3/9600)[\cos(k_x)\\ &+\cos(k_y)]/2,\end{aligned} \tag{7}$$

$$\begin{aligned}\Delta_t(k_x,k_y)/J_1 = &\,1-3691\lambda^2/30\,240+(-2\lambda/3+11\lambda^2/720)\\ &\times[\cos(k_x)+\cos(k_y)]/2\\ &-\lambda^2[\cos(2k_x)+\cos(2k_y)]/120\\ &+(\lambda/3-5\lambda^2/96)\cos(k_x)\cos(k_y)\\ &-\lambda^2[\cos(2k_x)\cos(k_y)\\ &+\cos(k_x)\cos(2k_y)]/90\\ &+7\lambda^2\cos(2k_x)\cos(2k_y)/360.\end{aligned} \tag{8}$$

The full series are available on request. We first consider the triplet excitations. Figure 4 shows $\Delta_t(k_x,k_y)$ along high-symmetry directions in the Brillouin zone for $\lambda=0.5$ and various coupling ratios y. For $\lambda\lesssim 0.6$ the series are well converged and direct summation and integrated differential approximants give essentially identical results. We find that the minimum gap occurs at (0, 0) for $J_2/J_1\lesssim 0.55$ and moves to

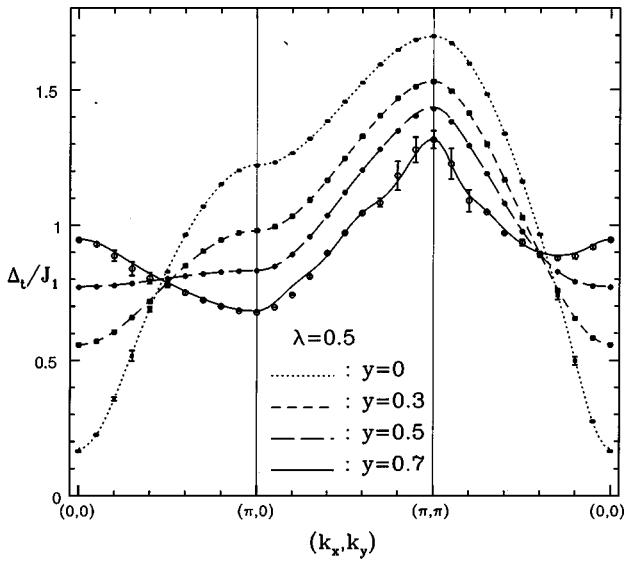

FIG. 4. Plot of the triplet excitation spectrum $\Delta(k_x,k_y)$ along high-symmetry cuts through the Brillouin zone for the system with coupling ratios $y=0$, 0.3, 0.5, 0.7, and $\lambda=0.5$ [shown in the figure from the top to the bottom at (π,π), respectively]; the lines are the estimates by direct sum to the series, and the points with error bar are the estimates of the integrated differential approximants to the series.

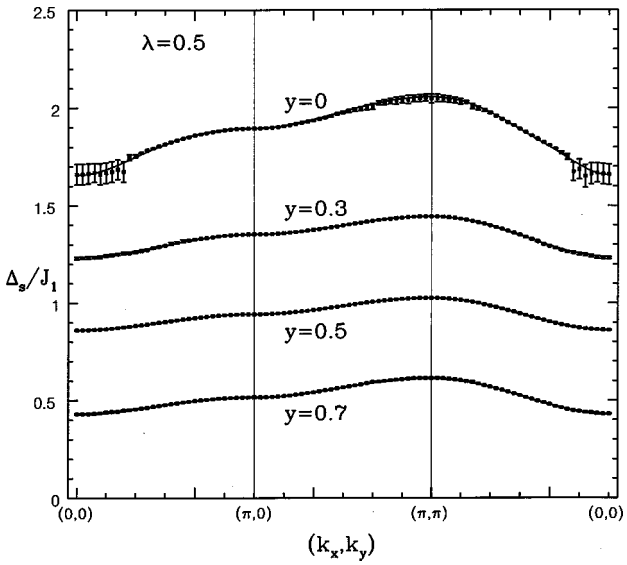

FIG. 5. Plot of the singlet excitation spectrum $\Delta(k_x,k_y)$ along high-symmetry cuts through the Brillouin zone for the system with coupling ratios $y=0$, 0.3, 0.5, 0.7 and $\lambda=0.5$ [shown in the figure from the top to the bottom, respectively]; the lines are the estimates by direct sum to the series, and the points with error bar are the estimates of the integrated differential approximants to the series.

$(\pi, 0)$ for $J_2/J_1 \gtrsim 0.55$. Next we seek to locate the critical point λ_c where the triplet gap vanishes. This is done using Dlog Padé approximants to the gap series at the appropriate (k_x,k_y). In practice this works well when the minimum gap lies at (0, 0). For $J_2=0$ we find a critical point at $\lambda_c = 0.555(10)$. We can compare this result with recent work of Koga *et al.*[18] who obtain $\lambda_c=0.112$ from a modified spin-wave theory and $\lambda_c \simeq 0.54$ from a fourth-order plaquette expansion. The critical point λ_c increases with increasing y. At $y=0.5$, at the approximate center of the intermediate phase, we find $\lambda_c \simeq 0.89(7)$. This result has some uncertainty but, if accurate, means that the plaquette phase becomes unstable before the full Hamiltonian ($\lambda=1$) is reached. The associated critical exponent ν describing the vanishing of the triplet gap is about 0.7 for $J_2/J_1<0.4$, suggesting that the transition lies in the universality class of the classical $d=3$ Heisenberg model. On the other hand, for $J_2/J_1 \gtrsim 0.4$ the exponent ν is about 0.4. This supports the existence of an intermediate phase lying in a different universality class.

Figure 5 shows the singlet excitation energy $\Delta_s(k_x,k_y)$ along high-symmetry directions in the Brillouin zone for $\lambda = 0.5$ and the same coupling ratios y as Fig. 4. Again the series are well converged and direct summation and integrated differential approximants give essentially identical results. We find that the minimum gap occurs at (0, 0) for all J_2/J_1. We have also noted that for $J_2/J_1=0.5$, the triplet excitation and the singlet excitation have the same gap at $\lambda = 0$, but at $\lambda=0.5$, the singlet gap is considerable larger than the triplet gap. This probably means that the triplet gap will close before the singlet gap at $J_2/J_1=0.5$. The critical point obtained by the Dlog Padé approximant to the singlet gap is also generally slightly larger than that obtained from the triplet gap around $J_2/J_1=0.5$ (see Fig. 6).

The full phase diagram in the parameter space of J_2/J_1 and λ could be very interesting from the point of view of quantum phase transitions, but may not be easy to determine by numerical methods. Some possible scenarios are shown in Fig. 7. One possibility is that the plaquette phase, for all J_2/J_1, has an instability to some magnetic phase and the dimerized phase exists only very close to $\lambda=1$ inside the magnetic phases. A second possibility is that the plaquette-Néel critical line meets the Néel-dimer critical line at some multicritical point at a value of J_2/J_1 around 0.5, after which there is a first-order transition between the plaquette and the

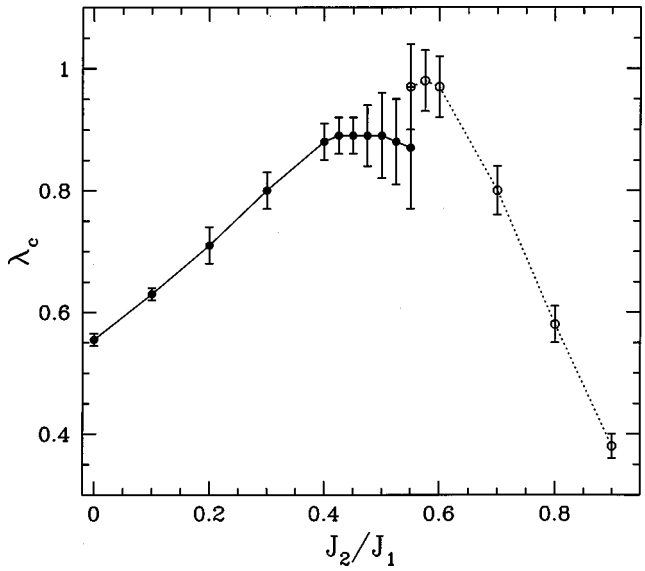

FIG. 6. Phase diagram for generalized J_1-J_2 Heisenberg model with plaquette structure, as determined from the plaquette expansions. The full (open) points with error bars and a solid (dotted) line to guide the eye indicate the line where the (0,0) triplet (singlet) gap vanishes.

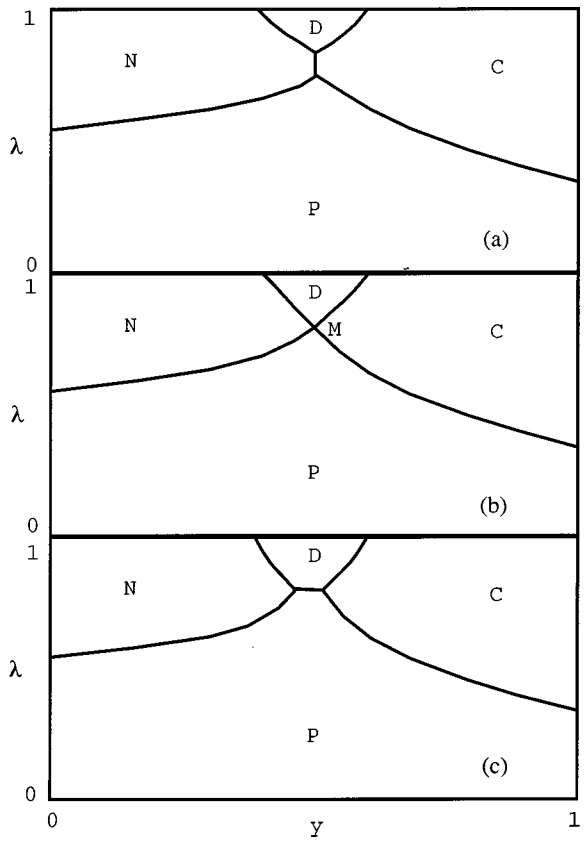

FIG. 7. Some possible topologies for the phase diagram in the λ-y plane. Here, N is the Néel phase, C is the columnar phase, D is the dimer phase, and P is the disordered Plaquette phase with no long-range order. Note that some of the phase boundaries such as those between Plaquette and dimer and between dimer and columnar phases could be first order, whereas the others could be second order. M is a possible multicritical point, where several critical lines meet.

dimer phases. A third possibility is that the plaquette-Néel, Néel-dimer and plaquette-columnar critical lines all meet at some multicritical point. The numerically determined phase diagram is particularly uncertain in the interesting region, $0.5 \lesssim J_2/J_1 \lesssim 0.6$., where incommensurate correlations could also become important.

Lastly we have derived expansions for a number of generalized susceptibilities. These are defined by adding an appropriate field term

$$\Delta H = h \sum_{ij} Q_{ij} \tag{9}$$

to the Hamiltonian and computing the susceptibility from

$$\chi_Q = -\frac{1}{N} \lim_{h \to 0} \frac{\partial^2 E_0(h)}{\partial h^2}. \tag{10}$$

A divergence of any susceptibility signals an instability of that phase with respect to the particular type of order incorporated in χ.

We have computed two different susceptibilities from the plaquette expansion. One is the antiferromagnetic (Néel) susceptibility with the operator Q_{ij}:

$$Q_{i,j} = (-1)^{i+j} S^z_{i,j}. \tag{11}$$

The other is the dimerization susceptibility with the operator Q_{ij}:

$$Q_{i,j} = \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} - \mathbf{S}_{i,j} \cdot \mathbf{S}_{i,j+1} \tag{12}$$

which breaks the symmetry of interchange of x and y axes. We have computed series to order λ^5 for the antiferromagnetic susceptibility and to order λ^4 for the dimerization susceptibility. The series have been analyzed by Dlog Padé approximants. The series for the antiferromagnetic susceptibility shows the same critical points (within error bars) as those obtained from the triplet gap for $J_2/J_1 \lesssim 0.4$. The series for the dimerization susceptibility is very irregular, and does not yield useful results. For example, for $J_2/J_1 = 0.5$, the series is

$$\chi_d = 629/90 + 101\lambda/300 + 2.009\,764\,7\lambda^2 - 0.269\,629\lambda^3 + 0.438\,527\lambda^4 + O(\lambda^5). \tag{13}$$

For completeness, we also compute the susceptibility for the dimer phase to become unstable to the plaquette phase from an expansion about isolated columnar dimers[12] by adding the following field term:

$$\Delta H = h \sum_{i,j} (-1)^j \mathbf{S}_{i,j} \cdot \mathbf{S}_{i,j+1}, \tag{14}$$

which breaks the translational symmetry in the direction perpendicular to the dimers. The series has been computed up to order λ^7 (note that λ here is the parameter of dimerization).

An analysis of the series shows that this susceptibility becomes very large as $\lambda \to 1$, for all J_2/J_1 and the critical λ, where the susceptibility appears to diverge, approaches unity from above as J_2/J_1 is increased to 0.5. This implies that there are staggered bond correlations in the direction perpendicular to the dimers, which extend over a substantial range. An interesting question is, in the absence of the plaquette phase as discussed earlier, what could these correlations represent? At this stage it is useful to recall another calculation by Kotov *et al.*[12] Within the dimer expansion, they calculated two different dimer order parameters:

$$D_x = |\langle \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} \rangle - \langle \mathbf{S}_{i+1,j} \cdot \mathbf{S}_{i+2,j} \rangle| \tag{15}$$

and

$$D_y = |\langle \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} \rangle - \langle \mathbf{S}_{i,j} \cdot \mathbf{S}_{i,j+1} \rangle|, \tag{16}$$

where the elementary dimers connect spins at i,j and $i+1,j$. They found that for $0.4 \lesssim J_2/J_1 \lesssim 0.5$, D_y is nearly zero, whereas D_x only goes to zero at the critical point. These results suggest that the dimer phase consists of strongly correlated two-chain ladders, which are then weakly correlated from one ladder to next. This striped nature of spin correlations in the dimer phase has not been noted before and is clearly a very interesting result. The situation for $J_2/J_1 \gtrsim 0.5$ is again less clear. As discussed before, there are many possibilities for the phase diagram in that region and much longer series are needed to throw more light on the situation. Perhaps there is an interesting multicritical point in that region of the phase diagram.

III. DISCUSSION

We have attempted to further elucidate the nature of the intermediate, magnetically disordered, phase of the spin-$\frac{1}{2}$ J_1-J_2 Heisenberg antiferromagnet on the square lattice. This phase is believed to occur in the range $0.4 \lesssim J_2/J_1 \lesssim 0.6$. Our approach has been to derive perturbation expansions (up to order λ^7) for the ground-state energy, singlet and triplet excitation energies, and various susceptibilities, starting from a system of decoupled plaquettes ($\lambda = 0$) and extrapolating to the homogeneous lattice ($\lambda = 1$). We have also derived expansions about an unperturbed state of isolated columnar dimers (''dimer phase''). Both of these have been proposed as candidates for the intermediate phase.

We find that the ground-state energy for both plaquette and dimer phases are very similar, any difference lying within the error bars. From this result alone we cannot favor one phase over the other.

The analysis of the singlet and triplet excitation spectra suggests an instability in the plaquette phase. In particular, in the disconnected plaquette expansions, Dlog Padé analysis indicates that the gaps would vanish for λ less than unity. The gap appears to close first for the triplets and then for the singlets. This is the strongest evidence that the plaquette phase is *not* realized in this model. However, we should mention here that the critical exponents associated with the vanishing of the gaps are rather small (< 0.4) and the gap closes not too far from λ equal to unity. Thus, with a relatively short series, this should be treated with some caution. One could ask why the energy series appear to converge well despite the instability. However, this is a well known feature of series expansions, that quantities having weak singularities may continue to show reasonable values even if extrapolated past the singularity.

A consistent interpretation of these results is that within the parameter space of our nonuniform Hamiltonian, the plaquette phase is first unstable to a magnetic phase, which then must give way to the columnar dimer phase. Similar results for the instability of the staggered dimer phase were suggested before by Gelfand, Singh, and Huse.[7] However, the full phase diagram in the J_2/J_1 and λ parameter space is difficult to obtain reliably, especially near the transition to the columnar phase. There are possibilities of some novel multicritical points, which deserve further attention.

One of our most interesting results is the finding of striped spin correlations in the dimer phase. In this phase, the nearest-neighbor spin correlations are nearly equal along the rungs and along the chains of a two spin column and there are extended bond correlations along the chains. However, spin correlations from one column to the next are much weaker. In other words, the dimers are strongly resonating along vertical columns. The existence of a quantum critical point separating an antiferromagnetic phase from such a quantum disordered phase with striped correlations is a very interesting feature of this model which deserves further attention in the context of high-T_c materials.

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