Zero-temperature ordering in two-dimensional frustrated quantum Heisenberg antiferromagnets

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We present series-expansion investigations of the square-lattice spin-$\frac{1}{2}$ Heisenberg antiferromagnet with nearest-neighbor (J_1), second-neighbor (J_2), and third-neighbor (J_3) exchanges. Expansions for ground-state properties around dimerized Hamiltonians in conjunction with finite-size studies and cluster mean-field theories allow us to map out the magnetically ordered phase boundaries. The magnetically disordered phase of the square-lattice Heisenberg model, which is accessible to the dimer expansions, appears to be spontaneously dimerized in the "columnar" pattern predicted by Read and Sachdev. Estimates for the dimerization order parameter in this phase are a substantial fraction of that of the fully dimerized state.

I. INTRODUCTION

The possibility[1] of spin fluctuations playing an essential role in the mechanism for superconductivity in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-x}$ has generated interest in understanding the low-temperature properties of two-dimensional (2D), $S=\frac{1}{2}$ Heisenberg antiferromagnets.[2] The generic phase diagram of these materials shows that by varying the carrier concentration by doping, the materials can be driven from insulating to metallic behavior. At the same time the antiferromagnetic correlation length is reduced, and the system instead of entering a Néel phase at low temperatures becomes a superconductor. Short-wavelength spin fluctuations survive even in the superconducting materials.[3] This has led to the suggestion that certain properties of magnetically disordered ground states may be central to the superconductivity. In fact, it has been suggested that the addition of carriers to certain magnetically disordered ground states immediately makes them superconductors.[4]

While the actual role of a finite density of mobile charge carriers in a magnetic system is hard to understand, it seems sensible to first study the nature of magnetically disordered ground states by introducing frustrating further neighbor interactions to a nearest-neighbor Heisenberg model. By now there is overwhelming evidence[5] that the antiferromagnetic nearest-neighbor square-lattice Heisenberg model has a magnetically ordered ground state. The insulating phase of La_2CuO_4 also seems to be well described by such a nearest-neighbor model.[6] In the presence of a small density of carriers, one might be able to formally integrate out the charge degrees of freedom,[7] to obtain an effective spin Hamiltonian with further neighbor interactions. Here we shall not be interested in such a connection with doped systems but rather study the frustrated spin Hamiltonians for their own interest. In the real doped system there is also the possibility that the charge and spin degrees of freedom are so intimately connected that the properties of the ground state cannot be accurately modeled by a purely spin system.

The specific model we study here is

$$H=\sum_{\mathrm{NN}}\mathbf{S}_i\cdot\mathbf{S}_j+J_2\sum_{\mathrm{2NN}}\mathbf{S}_i\cdot\mathbf{S}_j+J_3\sum_{\mathrm{3NN}}\mathbf{S}_i\cdot\mathbf{S}_j\ , \tag{1.1}$$

where the sums run over all first, second, and third nearest-neighbor pairs on a square lattice. There have been many studies of this and related models, analytical as well as numerical.[8–13] While all the studies are consistent with the idea that the Néel order of the ground state disappears at some value of the frustrating interactions, they disagree sharply regarding the nature of the disordered phase. Chandra and Doucot,[9] on the basis of a $1/S$ expansion for the sublattice magnetization, predicted that the Néel order vanishes for $J_2/J_1\approx 0.38$ for $S=\frac{1}{2}$ and $J_3=0$. They suggest that for a range of J_2/J_1 beyond 0.38 the system should be a spin liquid. This idea has been reinforced in the numerical studies of Figueirido *et al.*,[10] who looked at the spectra of 4×4 systems with different boundary conditions. By comparing the overlap of exact ground states with certain trial states they argued for the existence of a resonating valence bond (RVB), or spin-liquid, state with no broken symmetries. On the other hand, based on general arguments[11] and certain large-N expansions (spin models being $N=2$), Read and Sachdev[12] have argued that when the Néel state is disordered by changing parameters, the new ground state should be spontaneously dimerized. This latter state does not have the full symmetries of the square-lattice Heisenberg model. Note, however, that Read and Sachdev[12] have not explicitly treated models with frustration due to further neighbor interactions; instead the Néel state is destabilized in their study by the stronger quantum fluctuations at large N.

Here, we present systematic series-expansion studies for frustrated spin-$\frac{1}{2}$ Heisenberg models. The starting points for these perturbation expansions are given dimer coverings of the lattice; see Fig. 1. All the exchanges for bonds not present in the dimer covering are set to zero to form the unperturbed Hamiltonian, which then consists only of the intradimer couplings. The ground state of this unperturbed Hamiltonian is a product of singlets over all the dimers. The interaction Hamiltonian consists of all couplings between dimers, with the second- or third-neighbor couplings kept proportional to the previously omitted nearest-neighbor couplings. When the strength of these couplings λ reaches unity ($\lambda=1$), the full symmetry of the square lattice is restored. Thus the models at $\lambda=1$ are the ones we are primarily interested in here and they are accessed by perturbing away from the trivial models at $\lambda=0$. For lack of a better name we shall call these models at $\lambda=1$ "translationally invariant models." The models we shall study here always have a translational symmetry. It is the symmetry under translation by one site that will be broken for $\lambda\neq 1$. (Note $\lambda\neq 1$ also fully breaks the symmetry of the lattice under rotation around a lattice point.) If the ground state at $\lambda=1$ is magnetically ordered, the appropriate structure factor, $\hat{S}(\mathbf{q})$ diverges at some $\lambda_c<1$. Thus by studying the wave-vector-dependent structure factors, one can hope to find the boundaries separating all magnetically ordered phases from the disordered phase. For certain parameters and some starting dimer configurations the series expansions in powers of λ appear to be convergent all the way to $\lambda=1$. This is then evidence that for those parameters the ground state of the system at $\lambda=1$ is not magnetically ordered. In these cases we explicitly investigate the possibility of spontaneous dimerization (see Sec. IV).

In addition, we also carry out some finite-size studies. We use a cluster mean-field theory, where the boundary spins of a finite cluster are acted upon by an external field. This external field represents the interaction between the magnetizations on the neighboring clusters. The self-consistency requirement, namely, that the cluster have the same magnetization as that needed to produce the field, leads to mean-field equations. Its solution gives us a value for the sublattice magnetization, for a given set of parameters. By going to larger clusters one can, in principle, obtain the magnetic phase boundaries by this method. Here we are limited to the size 4×4 sites or less. Mata and Arnold[14] have used a similar method to study the nearest-neighbor Heisenberg model. Hence, the method is discussed only briefly. We have also obtained the symmetries of various low-lying excited states for 4×4 systems with periodic boundary conditions in different parameter regions, to infer the nature of possible ordering in an infinite system. Again, since our results overlap with those of Figueirido *et al.*[10] and Dagotto and Moreo[13] we shall present them only very briefly.

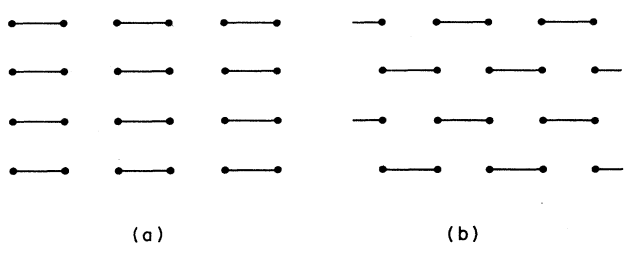

FIG. 1. Columnar (a) and staggered (b) dimer coverings of the square lattice used to set up the series expansions.

The plan of the paper is as follows: In Sec. II we outline the Hamiltonian, its classical (large-S) behavior, and the various parameter regions studied. In Sec. III we present the series expansions for $\hat{S}(\mathbf{q})$ and obtain the phase boundaries for the magnetically ordered regions. In Sec. IV we discuss the nature of the magnetically disordered phase, which is accessible to the series study and appears to be spontaneously dimerized. In Sec. V the results of cluster mean-field theory and finite-size studies are presented. Finally, in Sec. VI we present our conclusions.

II. MODELS

The models of interest consist of $S=\frac{1}{2}$ spins sitting on a square lattice, and coupled by Heisenberg exchange terms. The *translationally invariant* J_1-J_2-J_3 model is given by (1.1). To be more explicit, the second nearest-neighbor pairs are those connected by the diagonal vectors $(\pm 1,\pm 1)$ on the square lattice, while the third nearest-neighbor pairs are those connected by the vectors $(0,\pm 2)$ or $(\pm 2,0)$. Note that we have chosen the nearest-neighbor coupling to be antiferromagnetic and of unit strength. (We will typically take J_2 and J_3 to be antiferromagnetic and thus greater than 0 in our calculations, but that it is not always essential.)

All previous work on frustrated antiferromagnets has been concerned with translationally invariant systems, such as those described by (1.1). However, we are motivated by physical and technical considerations (namely, the prediction of spontaneous dimerization and the need for a trivial starting Hamiltonian for series expansions) to consider a broader class of models; which can be characterized as *dimerized* J_1-J_2-J_3 models. Let $\mathcal{D}$ represent a nearest-neighbor *dimer covering* of the square lattice, i.e., a partitioning of the points of the lattice into a collection of disjoint, nearest-neighbor pairs. Each dimer covering is associated with a three-parameter family of *dimerized* J_1-J_2-J_3 models of the form

$$H=\sum_{(i,j)\in D}\mathbf{S}_i\cdot\mathbf{S}_j+\lambda\left[\sum_{\substack{\mathrm{NN}\\(i,j)\notin D}}\mathbf{S}_i\cdot\mathbf{S}_j+J_2\sum_{\mathrm{2NN}}\mathbf{S}_i\cdot\mathbf{S}_j+J_3\sum_{\mathrm{3NN}}\mathbf{S}_i\cdot\mathbf{S}_j\right]. \tag{2.1}$$

If $\lambda=1$, one recovers the translationally invariant model (1.1), but if $\lambda=0$, one has a trivially solvable model consisting of independent pairs of antiferromagnetically coupled spins; its unique ground state is the product of the singlet states associated with each pair.

Because there are an infinite number of nearest-neighbor dimer coverings of the square lattice, it will not

be possible to consider every model of the form (2.1). Indeed, we shall only consider two choices for $\mathcal{D}$, namely, the "columnar" and "staggered" dimer coverings displayed in Fig. 1. These are exactly the ones considered in Ref. 15 (where models with nearest-neighbor coupling were studied); they were chosen because the graph enumeration required for the series expansions is easiest in those cases. In the present context, these two dimer configurations are also interesting physically: The columnar dimer covering reflects the pattern of *spontaneous dimerization* for the translationally invariant models predicted by Read and Sachdev, while the staggered dimers allow for a comparison between the results for the expected pattern and some other one.[16]

To recapitulate, for each dimer covering $\mathcal{D}$ there is a three-dimensional phase diagram, describing the ground state of (2.1), which we are interested in probing; the case $\lambda=1$ (which is the same for all $\mathcal{D}$) is of special interest. Most of our calculations are concerned with two *slices* through these phase diagrams, namely, the J_1-J_2 ($J_3=0$), and J_1-J_3 ($J_2=0$) models. (The only other case we examine is $J_2=2J_3$, and this is treated in less detail.)

We conclude this section with a review of the $T=0$ phase diagram for the *classical* ($S\to\infty$), translationally invariant J_1-J_2-J_3 model, since this guides some of our investigations. The phase diagram is displayed in Fig. 2, with the phases denoted by the wave vectors characterizing their ordering.

There are four classical phases present for the antiferromagnetic case $J_1=1$. They are (i) the usual Néel phase ordered at wave vector (π,π) ($k_x=\pi,k_y=\pi$), (ii) a four-sublattice antiferromagnetic phase ordered at wave vectors $(0,\pi)$ or $(\pi,0)$; (iii) a spiral antiferromagnet phase with the spiral along the diagonal, ordered at $(\pm q,\pm q)$ with $\cos q=-1/(2J_2+4J_3)$ and (iv) a spiral phase with the spiral parallel to a lattice axis, ordered at $(\pm q,\pi)$ or $(\pi,\pm q)$ with $\cos q=(2J_2-1)/4J_3$. There are special points of classical degeneracy: At the point $J_3=0$ and $J_2=\frac{1}{2}$ all spirals ordered at $(\pm q,\pi)$ or $(\pi,\pm q)$ with any q become degenerate in energy and similarly on the half-line $J_2=2J_3\geq\frac{1}{4}$ all spirals with $(\cos k_x+\cos k_y)=-1/2J_2$ are degenerate in energy.

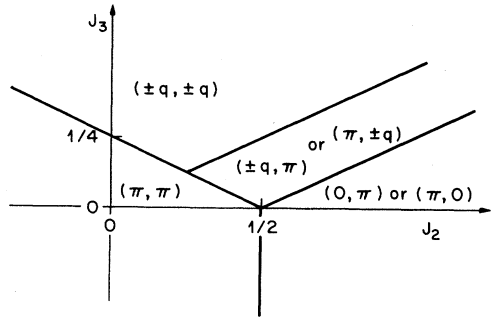

FIG. 2. Phase diagram for the classical J_1-J_2-J_3 model with $J_1=1$ at zero temperature. The wave vectors where the various phases order are indicated.

III. SERIES EXPANSIONS: PHASE BOUNDARIES

A. Preliminaries

The series expansions in λ for the models described by Eq. (2.1) were calculated by the cluster method of Singh, Gelfand, and Huse[15] to order λ^5. Most of the calculations were confined to the cases $J_2\neq 0, J_3=0$ and $J_3\neq 0, J_2=0$ because (for the columnar dimerization) there are less than half the number of connected graphs at $O(\lambda^5)$ for these cases than for the general case $J_2,J_3\neq 0$. All calculations, except those for $J_2,J_3\neq 0$ with the columnar dimerization, took roughly one day of Sun 3/50 time for each choice of J_2,J_3, and dimerization; the exceptions would have taken about three days but they were run instead on an IBM 3090.

The quantities for which the series were calculated were (i) the ground-state energy per spin E_g, (ii) the full set of spin-spin correlations $S(\mathbf{r})=\langle \mathbf{S}_{(0,0)}\cdot\mathbf{S}_{\mathbf{r}}\rangle$, and (iii) the "dimerization," defined by

$$D=4(\langle \mathbf{S}_{(2,0)}\cdot\mathbf{S}_{(1,0)}\rangle-\langle \mathbf{S}_{(0,0)}\cdot\mathbf{S}_{(1,0)}\rangle)\ , \tag{3.1}$$

where the nearest-neighbor spins (0,0) and (1,0) are assumed to be in the same dimer. The factor of 4 is for convenience, so that $D(\lambda=0)=3$.

It should be noted that even at fifth order in λ, the range of correlations which are accounted for in the series calculations is quite large. $S(\mathbf{r})$ is nonzero at fifth order for $|x|$ as large as 11, even without second or third-neighbor couplings; when the further neighbor interactions are included, the entire zone $|x|\leq 11, |y|\leq 10$ has nonzero correlations at fifth order. Thus, the series can effectively probe systems much larger than those which can be exactly diagonalized. In addition, one does not have to worry that the boundary conditions are favoring one type of possible ordering over some other, since the series are exact for the infinite system.

In the rest of this section, we will focus on the analysis of the spin-spin correlation series. The "ideal" way to handle the series would be to calculate the wave-vector-dependent structure factor

$$\hat{S}(\mathbf{q})=\sum_{r}\exp(i\mathbf{q}\cdot\mathbf{r})S(\mathbf{r}) \tag{3.2}$$

and its second moment

$$\hat{M}(\mathbf{q})=\sum_{r}\exp(i\mathbf{q}\cdot\mathbf{r})S(\mathbf{r})\mathbf{r}^2 \tag{3.3}$$

for each $\mathbf{q}$ and locate the values of λ where they diverge. The smallest such $\lambda_c(\mathbf{q})$ would then determine the limit of stability of the magnetically disordered phase, and the nature of the instability for each given dimer covering J_2, and J_3.

In practice, this is not what was done. In many cases (especially for $J_3=0$), analysis of the classical model indicates the wave vector at which ordering should occur; under those circumstances, there was little reason to thoroughly explore $\mathbf{q}$ space for every choice of coupling constants. In the cases in which the nature of the preferred orderings was *not* obvious *a priori,* the favored wave vector $\mathbf{q}_0$ was instead determined by looking for

maxima in the *coefficients* of the $\hat{S}$ and $\hat{M}$ series (at each order in λ) as a function of $\mathbf{q}$, following Redner and Stanley.[17] This method is preferable to studying $\lambda_c(\mathbf{q})$, because, for the short series available, the λ_c estimates are subject to large uncertainties, whereas the coefficients are known exactly. Once $\mathbf{q}_0$ has been obtained, $\lambda_c(\mathbf{q}_0)$ is estimated by the standard ratio method. The uncertainty in $\lambda_c(\mathbf{q}_0)$ is typically due only in small part to the lack of exact knowledge of $\mathbf{q}_0$; rather, it is dominated by nonlinearity of the ratios (i.e., corrections to asymptotic behavior) and by discrepancies between the extrapolations for the $\hat{S}(\mathbf{q}_0)$ and $\hat{M}(\mathbf{q}_0)$ series. The uncertainties in λ_c are largest for λ_c close to 1—and they do become quite large, on the level of 15%—even when $\mathbf{q}_0$ is believed to be known exactly.

In the following subsections, our results for the phase diagrams will be presented for the cases $J_2 \neq 0, J_3 = 0$; $J_2 = 0, J_3 \neq 0$; and $J_2 = 2J_3$; within each subsection the columnar and staggered dimer configurations will be compared.

Finally, it should be remembered that the analysis of the $S(\mathbf{r})$ series will only reveal critical points. First-order transitions cannot be seen directly—but evidence for the existence of first-order transitions in the phase diagrams will be presented later. Thus, some of the phase diagrams displayed are incomplete. Of course, it is also impossible for the series to reveal all critical points—only the ones closest to $\lambda = 0$ are accessible in this study.

B. $J_1 - J_2$ model

When $J_3 = 0$ and J_2 is not near $\frac{1}{2}$, the preferred ordering is unambiguous. For "small" $J_2 (< \frac{1}{2}$, classically) one has "two-sublattice Néel" order, that is, the usual (π,π) antiferromagnetic order. For "large" J_2 ($> \frac{1}{2}$ classically) the ordering is "four-sublattice Néel"; for the translationally invariant models, $(\pi,0)$ and $(0,\pi)$ are degenerate. However, any transitions at $\lambda < 1$ to a four-sublattice phase should be via an instability at $(\pi,0)$ *only*, since at the transition some of the horizontal nearest-neighbor bonds are stronger than the vertical ones; this is confirmed by inspection of the relevant series. For J_2 near $\frac{1}{2}$ one must check for order at all momenta of the form (π,q).

For the columnar dimerization, the phase diagram is displayed in Fig. 3. Series were calculated for J_2 at intervals of 0.1 between 0 and 1, and also for some $J_2 > 1$, which are not displayed. Notice that for $J_2 = 0.4$, 0.5, and 0.6 there was no indication that the system underwent a magnetic phase transition at any momentum $(\mathbf{q})$ and $\lambda \leq 1$. With only terms to $O(\lambda^5)$, transitions at $\lambda > 1$ are difficult to ascertain; in any event, as will be discussed in Sec. IV it appears that a first-order transition exists on this phase diagram at $\lambda = 1$ when there is no magnetic ordering at $\lambda < 1$.

In contrast, for the staggered dimerization there is *always* a transition to an ordered phase at $\lambda \lesssim 1$: see Fig. 4. For $0.5 \leq J_2 \leq 0.6$, series were calculated for J_2 at an interval of 0.01 in order to estimate $J_2^{(b)}$, where the (π,π) instability is replaced by the $(\pi,0)$ instability. We find[18] $J_2^{(b)} = 0.53 \pm 0.01$; at $J_2^{(b)}$ stability of the disordered phase all the way to $\lambda = 1$ cannot be ruled out. If, as appears to be the case, we assume $\lambda_c(J_2^{(b)}) < 1$, then this is a *bicritical* point, with a first-order transition between (π,π) and $(\pi,0)$ ordering as J_2 varies for λ just above $\lambda_c(J_2^{(b)})$. Although we have not made a detailed analysis of the problem, the most natural surmise is that it is a classical $d = 3$ O(6) bicritical point, since the transitions on either side are expected to be in the classical $d = 3$ Heisenberg universality class.[15]

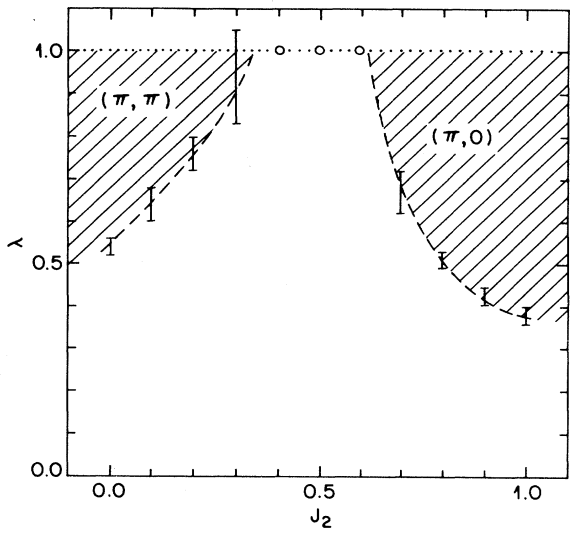

FIG. 3. Phase diagram obtained by series expansion for the columnar dimerized J_1-J_2 model. The open circles on the line $\lambda = 1$ indicate values of J_2 where there is no sign of magnetic ordering for $\lambda \leq 1$.

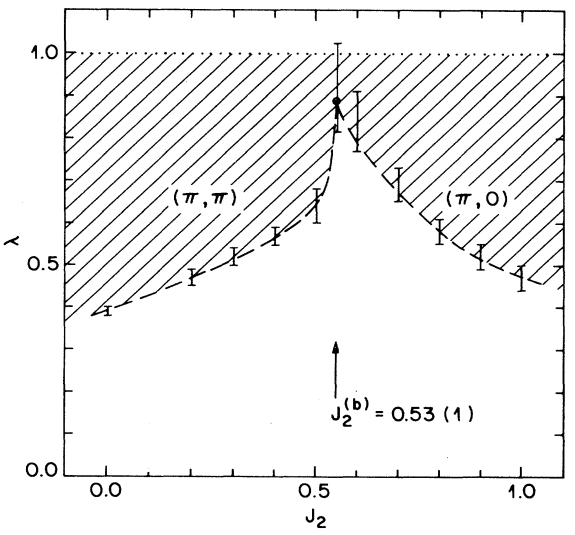

FIG. 4. Phase diagram for the staggered dimerized J_1-J_2 model.

C. J_1-J_3 model

For the J_1-J_3 model ($J_2=0$) the classical phase diagram displayed in Fig. 2 suggests that for sufficiently large J_3, the ordering wave vector at the transition from the disordered phase varies continuously with J_3. For the *columnar* dimerization, a continuously varying $\mathbf{q}_0$ seems to hold; see Fig. 5. The evidence for a $\mathbf{q}_0$ which is not strictly locked in to $(\pi/2,\pi/2)$ (to which $\mathbf{q}_0$ must approach, as $J_3\to\infty$) is fairly strong, even though the series are only to fifth order. The maxima in the coefficients are reasonably far from $(\pi/2,\pi/2)$, and do not move strongly towards it with increasing order. There is also an interval in J_3 for which apparently no critical point is encountered between $\lambda=0$ and $\lambda=1$. In addition, but not so germane to the matter of interest here, for $J_3>0.6$ we find a transition to a $(\pi,0)$ phase at *negative* λ, which is consistent with what one would expect classically.

In contrast with the J_1-J_2 model, for the J_1-J_3 case the phase diagram for the *staggered* dimerization is qualitatively similar to that for the columnar dimerization. Again, there is a $(\pi,0)$ phase at negative λ and an interval in J_3 for which the translationally invariant limit $\lambda=1$ can be reached from $\lambda=0$ *without* an intervening critical point. The transitions at $\lambda>0$ for $J_3\geq 0.8$ appear to have a continuously varying $\mathbf{q}_0$, though $\mathbf{q}_0$ varies less strongly with J_3 than in the case of a columnar dimerization, and a lockin at $\mathbf{q}_0=(\pi/2,\pi/2)$ cannot be ruled out.

Unfortunately, the series method cannot give any information regarding the details of the ordered phase of the J_1-J_3 model at $J_3\gtrsim 0.7$ and $\lambda>\lambda_c$. One possible scenario, based on a naive correspondence with the three-dimensional, finite-temperature biaxial next-nearest-neighbor Ising (Bi-ANNNI) model (which itself is not fully understood) is that the ordering wave vector does lock in to commensurate values. It is difficult to conceive of any practical numerical scheme to test this hypothesis, and determine which lockins occur, since the largest finite systems of $S=\frac{1}{2}$ spins which can be exactly diagonalized are smaller than 6^2, and present quantum Monte Carlo methods are not well suited to frustrated models.

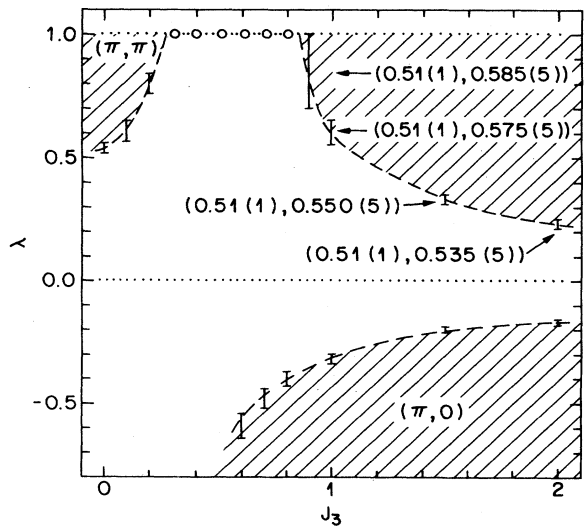

FIG. 5. Phase diagram for the columnar dimerized J_1-J_3 model. The estimated ordering wave vectors (in multiples of π) are indicated by arrows.

D. J_1-J_2-J_3 model

Our exploration of the full J_1-J_2-J_3 model has been limited in scope: We have studied only one slice through the phase diagram, with $J_2=2J_3$. We have found that λ_c for the disordered and/or (π,π) phase boundary increases with J_2, reaching $\lambda_c=1$ at $J_2^*\simeq 0.2$ for columnar dimers and $J_2^*\simeq 0.4$ for staggered dimers. No ordering is evident for $J_2\gtrsim J_2^*$ and $0<\lambda<1$, but we have not attempted to determine the character of ordering which may set in at large J_2. In short, the J_1-J_2-J_3 model looks very much like the J_1-J_3 model, at least for $J_2\lesssim 0.7$.

IV. SERIES EXPANSIONS: NATURE OF THE QUANTUM DISORDERED PHASE

It is now clear, based on the phase diagrams in Figs. 3–5 and also on other work (including that in Sec. V), that the two-dimensional, translationally invariant, frustrated $S=\frac{1}{2}$ Heisenberg antiferromagnets lack long-range spin-spin correlations over a range of coupling strengths J_2,J_3. The question remains, however, as to the nature of this "magnetically disordered" phase. In particular, is the ground state *nondegenerate,* as proposed by Chandra and Doucot[9] or is it fourfold degenerate, with long-range *energy-energy* correlations in the same pattern as the columnar dimerization, as proposed by Read and Sachdev.[12]

The series expansions about the columnar dimerization are analogous, in the present context, to expansions about infinite magnetic field at fixed temperature in a classical Ising ferromagnet. The issue is whether or not any order remains, spontaneously, even after the imposed ordering field is removed. Of course, since we also have expansions about the staggered dimerization, one can ask the same question in that case. However, it is clear, that if the ground state *is* spontaneously dimerized at $\lambda=1$, it can only be dimerized in one pattern (and equivalent patterns obtained by uniform translation and rotation in space) except at special values of the coupling constants. One such case is the "Klein model[19]" (which also contains 4-spin couplings) for which any state composed of singlets on a nearest-neighbor dimer covering is an exact ground state.

Our observations relevant to the nature of the quantum disordered phase begin with the phase diagrams. The fact that, for the J_1-J_2 model, there is a range of J_2 for which no phase transition is evident between $\lambda=0$ and $\lambda=1$ for the columnar dimerization (Fig. 3), but not for the staggered dimerization (Fig. 4), is surely a *hint* that the quantum disordered phase in this model is columnar dimerized.[16] However, that qualitative difference between the phase diagrams for columnar and staggered di-

merizations appears to be restricted to $J_3=0$. Indeed, the *lack* of critical points between $\lambda=0$ and $\lambda=1$ for the columnar *and* staggered dimerizations over a range of J_3 (in both J_1-J_3 and J_1-J_2-J_3 models) might be taken as evidence in favor of a nondegenerate, nondimerized quantum disordered phase.

The conflicting evidence provided by the phase diagrams is resolved by examining the ground-state *energies* at $\lambda=1$ for those values of (J_2,J_3) in the quantum disordered regime. In Fig. 6 various estimates of the energy for the J_1-J_3 model at $\lambda=1$ are displayed for the columnar and staggered dimerizations, respectively. (Ordinary Padé approximations are used in the expectation that the disordered phase is noncritical.) It is apparent that the energies one finds in each case are different, whereas they should be the same if the quantum disordered phase were nondegenerate. Furthermore, the columnar dimerization yields lower energies than the staggered dimerization. Finally, and most significantly, the estimated energies for the staggered dimerization lie above rigorous (variational) upper bounds, while the columnar energies lie below these bounds, which are also shown in Fig. 6. (For the J_1-J_2-J_3 model with $J_2=2J_3$, the staggered dimerization violates the variational bound for $0.4\lesssim J_2\lesssim 0.64$, and exceeds the energy for columnar dimerization over a wider range, at least 0.3 to 0.7.)

These variational bounds are obtained by numerically determining the ground-state energy for the 4×4 cluster with free boundaries. The energy per spin for that cluster is necessarily greater than the energy per spin on the infinite lattice, since one can partition the infinite lattice into nonoverlapping 4×4 blocks (which are, however, connected by interactions) and use, as a variational wave function, a product of the ground state for the 4×4 clusters. The energy per spin of this trial state is the same as for each individual 4×4 block, since spins in different blocks are uncorrelated, and so terms in the Hamiltonian which couple such clusters give no contribution to the energy of the trial state. Although such a calculation gives poor bounds for the ground-state energy of an unfrustrated system we expect the bounds to be close to the true answer for a highly frustrated system, due to cancellations in the boundary interactions.

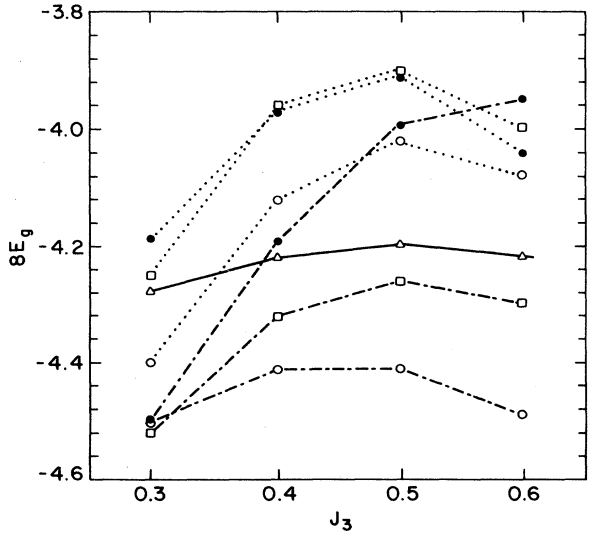

FIG. 6. Estimated ground-state energies at $\lambda=1$, for the J_1-J_3 model. The solid line shows the variational upper bound (see text). The dotted lines are for the case of staggered dimerization, while the dashed-dotted lines are for columnar dimerization. The dark and the open circles stand for partial sums of the energy series to order λ^4 and λ^5, respectively, whereas the squares represent the [2/2] Padé estimate.

Since the energies for the staggered dimerization violate the variational bounds, there is presumably a line of phase transitions somewhere, which makes it impossible to reach $\lambda=1$ from $\lambda=0$ without crossing the phase boundary. If we believe that the magnetically disordered phase is columnar dimerized, the phase diagram of Fig. 3 is also incomplete; there must be a first-order transition at $\lambda=1$, where the degenerate ground states exchange stability. A similar situation arises in the 1D frustrated antiferromagnets.[20] It is not obvious, *a priori*, whether the $\lambda=1^+$ ground state is horizontally dimerized (but shifted by one lattice spacing compared to that of $\lambda=1^-$), or, vertically dimerized and hence twofold degenerate, but a comparison of the estimated correlations at $\lambda=1$ indicates that the horizontal dimerization is favored for $\lambda>1$.

The ground-state energy series show that columnar and staggered dimerizations are not on an equal footing in regard to the disordered regime, and that a columnar spontaneous dimerization is preferred. Further evidence in support of column dimerization comes from examining the dimerization, or order parameter, series [recall Eq. (3.1)]. In Fig. 7 various estimates of D at $\lambda=1$ are shown for the J_1-J_2 model starting from columnar dimerization. The Padé approximants give physically sensible results, in the range $0\le D\le 3$, for J_2 in the magnetically disordered regime, but outside of that regime the approximants tend to have poles in the interval $0\le\lambda\le1$ (reflecting the critical points there) which make D vary widely. The value of D in the quantum disordered regime appears to be roughly 1.5–2.0, which is easily distinguished from zero.

The Read-Sachdev theory[12] suggests that $D(\lambda=1)$ should be identically zero for $J_2<J_2^*$, where J_2^* is the boundary between the (π,π) ordered phase and the magnetically disordered phase, and increases from zero with a singularity as J_2 increases past J_2^*. Such subtleties clearly cannot be captured by the series expansions presented here; the extrapolations of D to $\lambda=1$ will be unreliable for $J_2<J_2^*$ because of the intervening singularities; even for J_2 greater than but close to J_2^*, the estimates from a short series will not be trustworthy.

The analysis of dimerization series for the J_1-J_3 model with columnar dimerization again indicate a substantial spontaneous dimerization, in the range 1 to 2, in the quantum disordered phase. A positive D is also consistent with an ordered phase with $q_x=\frac{1}{2}\pi$ and collinear spin ordering, which could account for the reasonably good behavior of the Padé approximants for J_3 on the high side of the magnetically disordered regime, in con-

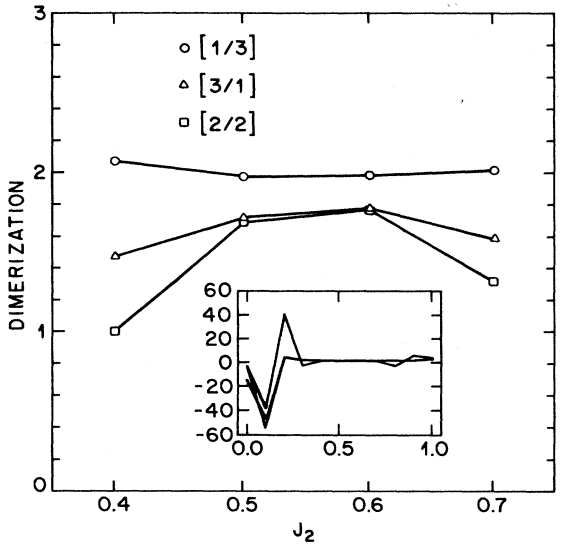

FIG. 7. Padé estimates for the columnar dimerization order parameter (3.1) at $\lambda=1$, for the J_1-J_2 model. The inset shows the behavior of the Padés in the extended range $0<J_2<1$. Outside the dimerized phase this estimate is clearly meaningless as it falls outside the physical bounds of [0,3].

trast with the situation for the J_1-J_2 model. As one might expect, the J_1-J_2-J_3 model also seems to be spontaneously dimerized to a large degree in the quantum disordered regime, with $D=2.0$–2.5. For comparison with other work, in Table I we present the series for the ground-state energy and dimerization for selected values of J_2 and J_3, starting from the columnar dimerization.

It is clear from the evidence presented that the quantum disordered phase is *not* spontaneously dimerized in the staggered pattern. The variational bounds on the energy provide evidence that staggered dimerized states, if they exist, have higher energy than the ground state (in the J_1-J_3 and J_1-J_2-J_3 models). But, the fact that the variational bounds are satisfied in the columnar case does not prove that the adiabatically continued state, which appears column dimerized, is the ground state for $\lambda=1$. In principle, some other pattern of dimerization might give yet a lower ground-state energy. Also the possibility of a first-order transition to a state with totally different symmetries cannot be ruled out by our calculations. However, as we shall see in Sec. V, finite-size studies reinforce the evidence that the ground states are column dimerized.

Apart from the question of the ground state itself, the evidence presented here is certainly suggestive that there are fourfold degenerate sets of eigenstates in the quantum disordered phase, in agreement with Haldane's prediction.[11] If one *accepts* that there are eigenstates which are columnar dimerized, then the estimates of D presented here should be reliable at least in the middle of the dimerized phases. Furthermore, this throws strong doubt on a nearest-neighbor RVB ground state, as the latter is not orthogonal to these dimerized states.

V. FINITE-SIZE STUDIES

In this section we shall discuss the results of cluster mean-field theory and finite-size studies for the J_1-J_2-J_3 models. Both of these methods rely on exact diagonalization of finite-size systems, to extract information about the ordering in the infinite system.

The cluster mean-field theory is a straightforward generalization of Kikuchi's cluster method to quantum spin systems.[14] In the simplest approximation, the cluster consists of a single site, and the mean-field equations correspond to Weiss molecular field theory. One can systematically improve the mean-field treatment by going to larger clusters. In that case the molecular fields are applied to the boundary spins of the cluster, the cluster is then treated exactly and self-consistent equations are obtained for the resulting magnetization in the interior of the cluster.

The method is illustrated by considering the 2-site cluster of a nearest-neighbor pair. If this cluster is embedded in a lattice when the outside spins have a fixed magnitude of the magnetization m, then the cluster Hamiltonian is given by

$$H=\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2+h(\sigma_1^z-\sigma_2^z)\ . \tag{5.1}$$

Here the $\boldsymbol{\sigma}$ are Pauli spin matrices, and the molecular field h depends on the pattern of ordering. For the two-sublattice Néel ordering [$\mathbf{q}=(\pi,\pi)$],

$$h=3\lambda m-4\lambda J_2 m-4\lambda J_3 m\ . \tag{5.2}$$

We can easily diagonalize the Hamiltonian in (5.1) and evaluate the magnetization on the site 1, in the ground

TABLE I. Ground-state energy and dimerization series (coefficients of λ^n to five significant digits), for the columnar dimerization, for selected values of J_2 and J_3 in the magnetically disordered regime. The λ^0 terms for $8E_g$ and D are -3 and 3, respectively, in every case.

	$J_2=0.5, J_3=0$		$J_2=0.6, J_3=0$		$J_2=0.4, J_3=0.2$		$J_2=0.5, J_3=0.25$	
n	$8E_g$	D	$8E_g$	D	$8E_g$	D	$8E_g$	D
1	0	−0.75	0	−0.75	0	−0.45	0	−0.375
2	−0.9375	−0.656 25	−0.885	−0.626 25	−0.855	−0.506 25	−0.75	−0.351 56
3	0.164 06	0.261 72	0.213 75	0.297 56	0.078 75	−0.071 44	0.140 63	0.053 71
4	−0.159 91	−0.315 67	−0.149 77	−0.288 81	−0.214 44	−0.393 68	−0.222 66	−0.309 58
5	0.097 20	0.134 66	0.130 85	0.268 60	0.158 71	0.184 25	0.174 25	0.232 05

state, by the formula

$$m_1 = |\langle \psi_0 | \sigma_1^z | \psi_0 \rangle / \langle \psi_0 | \psi_0 \rangle| \ . \tag{5.3}$$

The self-consistency requirement is, then,

$$m_1 = m \ . \tag{5.4}$$

Diagonalizing the Hamiltonian, we find that for $J_3 = 0$ a nonzero solution for m exists provided

$$\lambda > 1/(3-4J_2) \ . \tag{5.5}$$

Similarly for the ordering at $q = (0,\pi)$ a nonzero solution for m exists for $J_3 = 0$ when

$$\lambda > 1/(4J_2 - 1) \ . \tag{5.6}$$

The resulting phase boundaries are shown in Figs. 8(a) and 8(b).

One can now consider the mean-field theory for $4, \ldots, 16, 18, \ldots$ site clusters. Since the phase boundaries cannot be extrapolated in a systematic manner from the sizes studied, and are qualitatively similar for 16- and 18-site clusters, we shall restrict attention to the $4\times4 = 16$-site cluster. For this cluster the Hilbert space is too large to be treated analytically. However, the ground state is readily obtained by the Lanczos method. We have investigated both the J_1-J_2 model and the J_1-J_3 model with both columnar and staggered dimerizations.

The phase diagrams obtained for the J_1-J_2 model are shown in Figs. 8(a) and 8(b) with the dashed lines separating the disordered and magnetically ordered phases. The solid line in the columnar dimer case [Fig. 8(a)] represents a first-order phase boundary, where a jump in the spontaneous magnetization occurs. The dashed-dotted line represents a "spinodal" beyond which the zero magnetization state becomes unstable to small perturbations. The region in between shrinks as one goes from 2×2 to 4×4 clusters and hence is consistent with the possibility that the actual transition is second order.

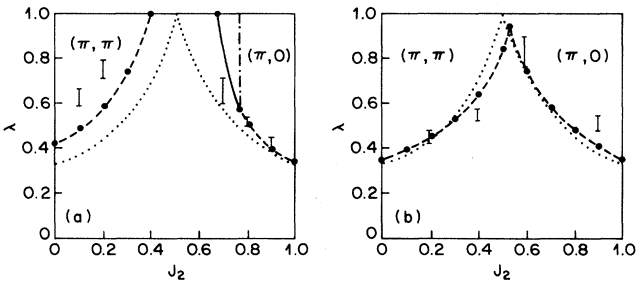

FIG. 8. Phase diagram obtained by cluster mean-field theory for the J_1-J_2 model with (a) columnar and (b) staggered dimerizations. The dotted lines represent the phase boundary obtained by 2-site cluster mean-field theory. The other phase boundaries are from the 4×4 clusters. The solid line represents a first-order phase transition, while the dashed-dotted line is the corresponding spinodal (see text). Some of the series estimates for the phase boundaries from Figs. 3 and 4 are shown for comparison.

These results are quite similar to those obtained by series extrapolations. For the columnar dimers there is a region $0.4 \lesssim J_2 \lesssim 0.6$, where the translationally invariant Heisenberg model appears to be magnetically disordered and connected to the dimerized phase. In contrast, for the staggered case no such phase seems to appear. The phase diagrams obtained for the J_1-J_3 model are also similar to those from series expansions, except that incommensurate ordering is not captured.

Unfortunately this mean-field theory does not give us any information on the nature of the magnetically disordered phase, which is the subject of prime interest here. There does not appear to be any straightforward way of constructing a mean-field theory that shows spontaneous dimerization for this model.[21] However, we will now consider the symmetries of the low-lying states, in finite systems, which are informative of the nature of these phases.

For a 4×4 system with periodic boundary conditions, we have found the energies and quantum numbers of the ground and low-lying excited states of (1.1). Similar work has been done independently in Refs. 10 and 13. The ground state is always of the highest symmetry: It is a singlet with total spin zero, no momentum, and is even under all reflections and rotations of the lattice. If the ground state in the thermodynamic limit spontaneously breaks a symmetry, for the finite system it should be thought of as a sum of all the different symmetry-broken states with equal weights. Low-lying excited states of a large but finite system can be constructed by adding the symmetry-broken states with unequal weights in a way with lower symmetry. However, the only symmetries that can be broken in such low-lying excited states are those that the system spontaneously breaks in the thermodynamic limit. Thus the quantum numbers of the lowest-lying excited states in a finite-size system suggest the possible nature of the broken symmetries in the thermodynamic limit. Of course, even if the system does not spontaneously break any symmetries in the thermodynamic limit the finite-size systems will have lowest-lying excited states with certain quantum numbers. Thus the evidence presented here is merely suggestive and is always also consistent with a fully disordered phase.

In the usual two-sublattice Néel phase there is magnetic order at momentum (π,π) and this is indicated by the lowest-lying excited states being a triplet with total spin unity and momentum (π,π). This occurs for $J_3 = 0$ and $J_2 \lesssim 0.41$; $J_2 = 2J_3$ and $J_2 \lesssim 0.33$; and $J_2 = 0$ and $J_3 \lesssim 0.38$. In the four-sublattice Néel phase there is magnetic order at momentum $(0,\pi)$ or $(\pi,0)$. Since the system has two different momenta at which it may order, it also breaks the symmetry under rotation of the lattice by angle $\pi/2$. The lowest-lying excited states are a pair of triplets with total spin unity and momenta $(\pi,0)$ and $(0,\pi)$ for $J_3 = 0$ and $J_2 \gtrsim 1.27$, indicating this phase. For $J_3 = 0$ and $0.41 \lesssim J_2 \lesssim 1.27$ the lowest-lying excited state is a singlet with all the same quantum numbers as the ground state *except* it is odd under rotation of the lattice by $\pi/2$ (see also Ref. 13).

A spontaneously columnar dimerized state has total spin zero and breaks translational and rotational sym-

metries. The four postulated ordered states can be added four ways to produce (i) the fully symmetric ground state, (ii) a state that is odd under rotation by $\pi/2$ of the lattice but otherwise fully symmetric, and (iii) two states with momenta $(0,\pi)$ and $(\pi,0)$. Note that the lowest-lying excited state for $J_3=0$ and $0.41 \lesssim J_2 \lesssim 1.27$ is consistent with both the four-sublattice Néel phase and the columnar dimer phase. Thus to get an indication of which of the two ordered phases is more plausible we look for the lowest-lying excited state that discriminates between the two. For $J_3=0$ and $J_2 \gtrsim 0.64$ this is the pair of triplets with total spin unity and momenta $(0,\pi)$ and $(\pi,0)$, suggesting that four-sublattice Néel order prevails in that region. Over the remainder of the interval $0.41 \lesssim J_2 \lesssim 0.64$ there is a pair of singlets with total spin zero and moments $(0,\pi)$ and $(\pi,0)$, as expected for the columnar dimer phase (see also Ref. 13).

For $J_2=2J_3$ and $0.33 \lesssim J_2 \lesssim 0.8$, as well as for $J_2=0$ and $0.38 \lesssim J_3 \lesssim 0.7$, the three lowest-lying excited states with symmetry different from the ground state are as expected for the columnar dimer phase. We did not look at larger values of J_2 or J_3 for either of these cases.

Thus the excited-state quantum numbers of this small system are consistent with the occurrence of a columnar dimer phase over a substantial range of J_2 and J_3, in general agreement with our series and mean-field results.

VI. CONCLUSIONS

We have presented here a detailed study of the ground states of frustrated quantum $S=\frac{1}{2}$ Heisenberg models on the square lattice. The approaches used include series expansions around dimerized Hamiltonians, cluster mean-field theory, and an investigation of the low-lying excited states of 4×4 systems. All these studies indicate that there are regions in the parameter space where the ground state has no long-range magnetic order in the thermodynamic limit. Furthermore, there is strong evidence that in this magnetically disordered phase there occurs a different type of spontaneous ordering, namely, spontaneous dimerization in the columnar pattern.

To the best of our knowledge, there is no evidence for such a spontaneous dimerization in doped La_2CuO_4. This suggests, not surprisingly, that the effects of doping are not accurately modeled by simply adding further neighbor interactions to a Heisenberg model. Precisely what type of ground state such a doping actually leads to remains to be seen.

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