*J*1-*J*2 model: First-order phase transition versus deconfinement of spinons

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We revisit the phase transition from the Néel ordered to a valence bond solid (VBS) state in the two-dimensional J_1-J_2 antiferromagnetic Heisenberg model. In the first part we address the question whether or not this transition could be an example of a second-order phase transition due to a deconfinement of spinons. We give arguments based on series expansion and spin-wave theory that this is not the case and the transition is most likely first order. The method proposed here to detect first-order phase transitions seems to be very sensitive and might be useful in other models as well. In the second part we analyze possible VBS patterns in the magnetically disordered phase based on numerical data for different susceptibilities, obtained in the ordered phase, which test the breaking of lattice symmetries. We conclude that a columnar dimerization pattern is the most likely candidate.

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I. INTRODUCTION

It is well known that microscopically different systems can show similar behavior near a critical point, a phenomena termed universality. The universal behavior is caused by the fact that only a small number of long-wavelength degrees of freedom are relevant for most physical quantities at the critical point. It is indeed often sufficient to consider an effective theory in which all modes other than the order parameter have been eliminated. The concept of the order parameter and effective theories based on this quantity has been developed by Landau and Ginzburg.[1]

Recently, it has been argued that there is a second-order phase transition in the $S=1/2$ square lattice antiferromagnet between the Néel state and a paramagnetic valence bond solid state which is not described by a Ginzburg-Landau (GL) type critical theory.[2,3] Instead the proposed theory involves fractional degrees of freedom (spinons) interacting with an emergent gauge field. One of the best studied models where this scenario could possibly be realized is the spin-$1/2$ J_1-J_2 Heisenberg antiferromagnet in two dimensions

$$H = J_1 \sum_{nn} \mathbf{S}_i \mathbf{S}_j + J_2 \sum_{nnn} \mathbf{S}_i \mathbf{S}_j. \tag{1}$$

Here $J_1>0$ is the nearest-neighbor interaction and $J_2 \geqslant 0$ a frustrating next-nearest-neighbor exchange. There are two well understood limits: For $J_1 \neq 0$, $J_2=0$ the model is just the usual two-dimensional Heisenberg antiferromagnet which is known to possess Néel order although with an order parameter $M \approx 0.3$ which is reduced compared to its classical value $M=1/2$. For $J_1 \to 0$ and $J_2 \neq 0$ and fixed, on the other hand, both sublattices are Néel ordered and J_1 then induces a so-called collinear order. For general couplings J_1, J_2, both limiting ground states become frustrated. Therefore a parameter region might be expected where the magnetic order vanishes and a spin liquid or VBS ground state is formed. Numerical studies including exact diagonalization,[4] variational quantum Monte Carlo[5,6] as well as series expansion[7–11] indeed indicate that for $0.4 \lesssim g \lesssim 0.6$, with $g=J_2/J_1$, no magnetic order exists.

To address the question of whether or not the ground state in the nonmagnetic region breaks a lattice symmetry, the response to a field

$$F_1 = \delta \sum_{i,j} (-1)^i \mathbf{S}_{i,j} \mathbf{S}_{i+1,j} \tag{2}$$

has been calculated.[5,10–12] The series expansion studies[10,11] show that the corresponding susceptibility becomes very large or even diverges when $g \approx 0.4$ is approached from the Néel phase, indicating that translational symmetry by one site is broken and a VBS state is formed. The results obtained in Ref. 5 by a variational quantum Monte Carlo (QMC) technique seem to support this scenario. Furthermore, the QMC data have been shown to be in good agreement with an exact diagonalization of clusters with $N=16,32$ spins. In a later exact diagonalization study, however, it has been shown that the susceptibility decreases when going from the 4×4 to the 6×6 cluster.[12] Further evidence in favor of a breaking of translational invariance has been obtained by a dimer series expansion showing directly that the corresponding order parameter is indeed nonzero in this phase.[9] On the basis of the series expansion data we therefore believe that the existence of a homogeneous spin liquid phase in this parameter region, as proposed in Ref. 6, is highly unlikely.

A very important result of the series calculations in Refs. [9–11] is that the point g_{cv}, where magnetic order vanishes, and the point $g_{c\phi}$, where dimer order becomes established, are very close or even coincident. Furthermore, a crossing of energies between an Ising expansion and a dimer expansion, which would be the indication of a first-order transition, could not be detected. This lead to the assumption that the transition is second order. For a second-order transition it is,

however, difficult to understand within GL theory why g_{cv} and $g_{c\phi}$ should be equal. Each phase has a different broken symmetry (spin-rotational symmetry versus lattice symmetry) so that one would naively expect that each transition is described by its own effective theory containing only the staggered magnetization (order parameter in the Néel phase) or the dimer order parameter, respectively. In this case the transitions should be independent from each other. That this seems to be not the case tells us that the two order parameters must be related. A GL-type theory should therefore at least contain also terms describing the interaction between the two order parameters. Such an effective theory has been proposed in Ref. 11. However, it has been found that within this theory g_{cv} and $g_{c\phi}$ will only be identical if the nonmagnetic phase has massless excitations. This would correspond to a transition to a translationally invariant spin liquid phase which we believe can be ruled out based on the numerical data obtained in Ref. 9.

The new effective theory for the second-order phase transition between the Néel and a VBS state proposed in Refs. 2 and 3 follows an entirely different route. Here the order parameters in the two phases are represented in terms of fractional degrees of freedom (spinons) which become deconfined exactly at the critical point. As the spinons are the constituents of both order parameters this would offer a natural explanation for a direct second-order phase transition between these at first sight very different phases. Among the models proposed to show such a transition is the two-dimensional spin-1/2 model with a four-spin exchange.[13] Quite recently, however, it has been argued that the transition in this model is more likely to be first order.[14] In the present work we will address the same question about the order of the phase transition for the J_1-J_2 model.

Our paper is organized as follows: In Sec. II we give arguments based on series expansion and spin-wave theory why the phase transition is most likely a weak first-order instead of a second-order transition. In Sec. III we will discuss numerical data for three different susceptibilities probing the VBS order in the nonmagnetic phase. These susceptibilities are obtained in the Néel phase where the ground state is known and the series is therefore not biased. We will discuss why these data provide additional evidence against the deconfinement scenario and will conclude that the VBS order is most likely of the columnar dimer type. The last section presents a summary and conclusions.

II. ORDER OF THE PHASE TRANSITION

Usually a first-order phase transition is detected in series calculations by looking for the crossing of energies obtained by expansions starting from different states. For the J_1-J_2 model such an energy crossing has been detected between an Ising expansion in the collinear regime and a dimer expansion in the VBS phase.[9] This shows that the transition at $g \sim 0.62$ from the VBS to the collinear state is first order (see also Fig. 7). For the transition from the Néel to the VBS state at $g \sim 0.4$, on the other hand, no crossing has been found. More precisely, the energies for an Ising expansion and various dimer expansions are so close over a relatively large parameter regime around the transition point that it is not possible to decide within the accuracy of the series if there is a crossing or not (see Ref. 9 and Fig. 7). In all previous series studies it has been implicitly assumed that the transition is second order. Here we propose a more sensitive method to distinguish between first- and second-order transitions and conclude that the transition is most likely weak first order.

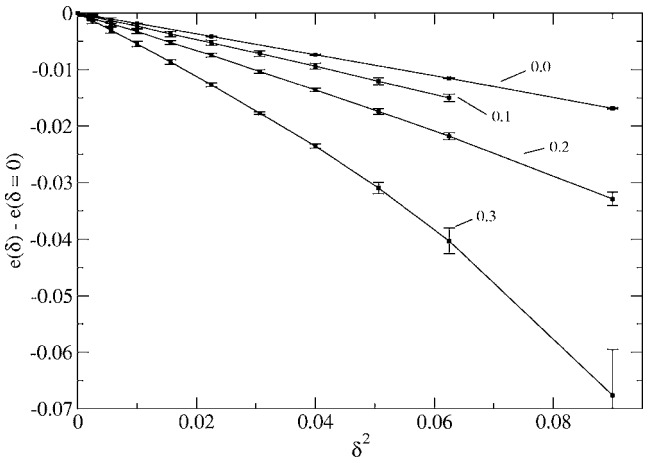

FIG. 1. Ising series data for the ground state energy $e(\delta)$ with the field F_1 included and $g=0.0,0.1,0.2,0.3$. The lines are a guide to the eye.

Let us consider the ground-state energy $e(\delta)$ for the Hamiltonian (1) with the field in Eq. (2) included for $|\delta| \ll 1$ and $g \leqslant 0.4$. We have calculated $e(\delta)$ for different g by Ising series expansion. Using an Ising expansion means that we start with a state which breaks spin-rotational symmetry whereas the lattice symmetries are intact. Obviously, it is then impossible to restore spin-rotational symmetry and break lattice symmetries—as would be required when going from the Néel to the VBS state—in any finite order in the expansion. However, we can expect that an instability of the state we are starting with is signaled by a susceptibility, with respect to the corresponding symmetry breaking field, which is divergent.

In Fig. 1 we present our numerical data. For a fixed g we have fitted $e(\delta)$ by a polynomial of the form

$$e(\delta) - e(0) = \frac{a}{2}\delta^2 + \frac{b}{4}\delta^4 + \frac{c}{6}\delta^6. \tag{3}$$

The susceptibility is then given by

$$\chi_1 = -\left.\frac{\partial^2 e}{\partial \delta^2}\right|_{\delta=0} = -a. \tag{4}$$

As the series for the ground state energy shows better convergence than the series for the susceptibility itself (which was calculated in Ref. 10), we were able to obtain χ_1 with much smaller error bars than before as shown in Fig. 2. The new data are nevertheless consistent with the old data within the given error bars. A strong response to the field F_1 is visible indicating that translational symmetry is broken in the nonmagnetic phase. If the phase transition with respect to the corresponding order parameter would be second order we

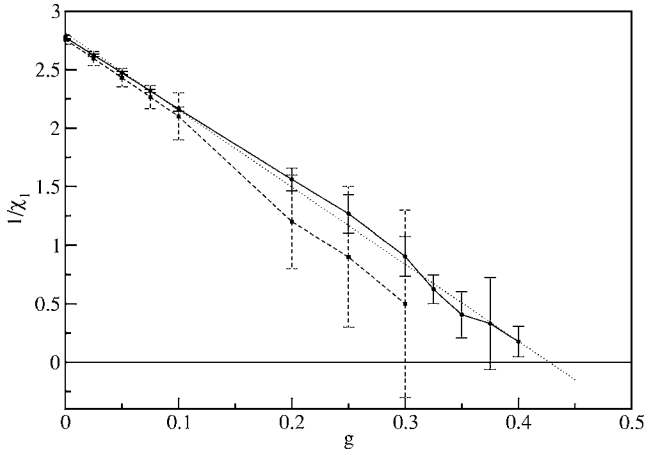

FIG. 2. The susceptibility χ_1 calculated as described in the text (solid line) compared to the old data from Ref. 10 (dashed line). The dotted line represents a linear fit of the new data $1/\chi_1=6.58\times(0.43-g)$.

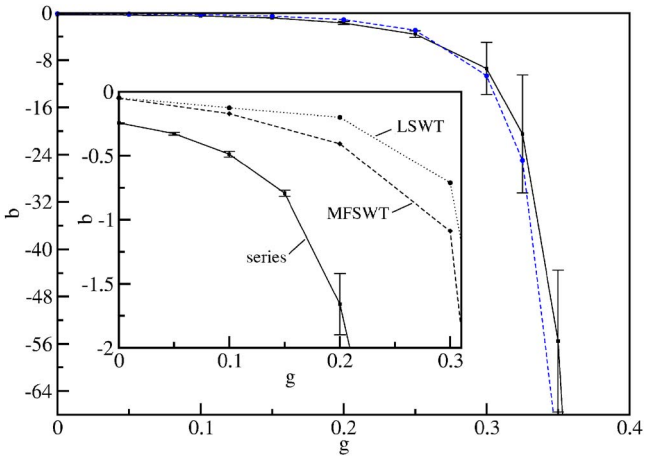

FIG. 3. (Color online) The coefficient b of the quartic term in Eq. (3) obtained from a fit of the data in Fig. 1 (black squares). The dashed line represents a fit $b(g)=-5.68/[6.58(g-0.43)]^4$. Inset, the coefficient b from LSWT and MFSWT in comparison to the series data. The lines are a guide to the eye.

expect $1/\chi_1 \propto (g_{c\phi}-g)^{\gamma_\phi}$ where $g_{c\phi}$ is the critical point and γ_ϕ the critical exponent. In mean-field theory $\gamma_\phi=1$ and we can indeed obtain a nice linear fit of $1/\chi_1$ as shown in Fig. 2. Note, that γ_ϕ is not expected to change dramatically even if fluctuations are taken into account [an $O(1)$-model, for example, would have $\gamma_\phi \approx 1.2$] so that changes to the value of $g_{c\phi}$ would be minor.

To study different possible scenarios for this phase transition we consider an effective field theory for the magnetically ordered phase. In the effective field theory for a two-dimensional antiferromagnet in the ordered phase no topological term (Berry phase) is present.[15] One can therefore describe the system by the following $O(3)$-model:

$$H_{\mathbf{v}} = \frac{1}{2}\{(\partial_t \mathbf{v})^2 + c_v^2(\nabla \mathbf{v})^2 + m_v^2 \mathbf{v}^2\} + \frac{u_v}{4}(\mathbf{v}^2)^2. \tag{5}$$

Assuming a second-order phase transition at a critical point g_{cv} we have $m_v^2 = a_v(g-g_{cv})^{\gamma_v}$ and $u_v>0$. Here γ_v is the critical exponent for the staggered magnetic susceptibility with $\gamma_v=1$ in mean-field theory. At $g<g_{cv}$ the vector field (v) will then show a nonzero ground state expectation value $\langle \boldsymbol{v} \rangle = \sqrt{a_v(g_{cv}-g)/u_v}$.

Consider now the case that we are in the magnetically ordered phase and add the field F_1 as given in Eq. (2) with $|\delta| \ll 1$. The Néel order will then coexist with a small dimerization described by a scalar field

$$H_\phi = \frac{1}{2}\{(\partial_t \phi)^2 + c_\phi^2(\nabla \phi)^2 + m_\phi^2 \phi^2\} + \frac{u_\phi}{4}\phi^4 + \frac{r_\phi}{6}\phi^6 - \delta\phi. \tag{6}$$

If a second-order phase transition with respect to ϕ at a critical point $g_{c\phi}$ would occur, we would have $m_\phi^2 = a_\phi(g_{c\phi}-g)^{\gamma_\phi}$ and $u_\phi>0$. We also want to include an interaction between the vector and the scalar field. The lowest order coupling term allowed by symmetry is

$$H_{\text{int}} = \frac{u_{v\phi}}{2}\mathbf{v}^2\phi^2. \tag{7}$$

The effective field theory in the ordered phase for $\delta \neq 0$ is then given by $H=H_v+H_\phi+H_{\text{int}}$ and we will have a nonzero ground state expectation value

$$\langle \phi \rangle = \frac{\delta}{A} - \frac{u_\phi}{A^4}\delta^3 + \frac{3u_\phi^2 - A r_\phi}{A^7}\delta^5 + \mathcal{O}(\delta^7) \tag{8}$$

with $A=m_\phi+u_{v\phi}\langle \boldsymbol{v} \rangle^2$. This leads to a ground state energy given by

$$e(\delta)-e(\delta=0) = -\frac{1}{2A}\delta^2 + \frac{u_\phi}{4A^4}\delta^4 + \frac{A r_\phi - 3u_\phi^2}{6A^7}\delta^6 + \mathcal{O}(\delta^8). \tag{9}$$

This regular expansion in powers of δ^2 will exist for any $g<g_{c\phi}$ but the radius of convergence will become smaller and smaller when the assumed critical point is approached. Finally, this expansion will break down and directly at the critical point $e(\delta)$ will show a scaling with a critical exponent which is in general noninteger. Nevertheless, in the parameter regime where this expansion is valid we expect the coefficient of the δ^4-term to be *positive* because $u_\phi>0$ for a second-order phase transition.

We have fitted $e(\delta)$ shown in Fig. 1 by the polynomial given in Eq. (3) but studied so far only the coefficient of the quadratic term which gives the susceptibility (see Fig. 2). The coefficient of the quartic term obtained by a fit of these data is shown in Fig. 3. Surprisingly, this coefficient is negative. Before we discuss the consequences we will check if the coefficient follows the form predicted in (9). The coefficient $A=1/\chi_1$ so that we can use the linear fit shown in Fig. 2 for this parameter. With these values for A the data in Fig. 3 are in very good agreement with Eq. (9) where the single parameter fit (shown as dashed line) yields $u_\phi=-5.68$.

A check of our series data for b is provided by calculating this coefficient in spin-wave theory. We know that linear spin-wave theory (LSWT) yields accurate results for quantities like ground-state energy or sublattice magnetization for the two-dimensional Heisenberg antiferromagnet without frustration.[16,17] Surprisingly, LSWT gives also a phase diagram for the J_1-J_2 model[18] which is very similar to the one found by numerical calculations. In particular, a nonmagnetic ground state for $g \in [0.38, 0.51]$ is found. In a spin-wave theory where the quartic terms are treated self-consistently in one-loop approximation (MFSWT), however, the phase diagram changes dramatically and Néel order remains stable up to $g \sim 0.6$.[19] This tells us that we cannot trust spin-wave theory in the strongly frustrated regime. If we consider g close to zero, on the other hand, we might expect that spin-wave theory results are reasonable.

In complete analogy to the series calculations we have calculated the ground-state energy $e(\delta)$ in LSWT and MFSWT for the J_1-J_2 model with the term (2) included. The coefficient of the δ^4-term obtained from these calculations is shown in the inset of Fig. 3 in comparison to the series data. Again we find that b is negative and decreases with increasing g. Quantitatively, b is larger, both in LSWT and MFSWT, as in series and decreases more slowly.

A negative u_ϕ in Eq. (6) means that the phase transition with respect to ϕ will be *first order* and the assumed critical point $g_{c\phi}$ will never be reached. For $\delta=0$ the order parameter $\langle\phi\rangle$ will instead jump from zero to some finite value. To determine the transition point one also needs to know r_ϕ which must be positive for stability reasons. In principle, r_ϕ can be determined from the sixth-order term in (9). In practice, the errors in the numerical data for $e(\delta)$ and in the parameters A, u_ϕ are too large to determine r_ϕ reliably. Due to the interaction term (7) a jump in $\langle\phi\rangle$ will induce a simultaneous jump in the Néel order parameter $\langle\mathbf{v}\rangle$. Depending on the strength of the interaction $u_{v\phi}$ two scenarios are possible: The Néel order parameter could jump to zero yielding a direct first-order transition from the ordered phase to a disordered phase with broken translational invariance. A more exotic scenario, where $\langle\mathbf{v}\rangle$ jumps to a smaller but nonzero value and then decreases further before vanishing at a critical point, is also possible. This would imply that there is a region in the phase diagram where dimerization and Néel order coexist. Based on the currently available numerical data it is impossible to decide which scenario is actually realized. We also want to mention that our mean-field treatment of the order parameters and the use of the expansion (9) are *a posteriori* justified. The finding of a first-order phase transition means that we never get to the assumed critical point $g_{c\phi}$ where the length scale for fluctuations would diverge and a mean-field treatment would therefore be very questionable. It also means that the expansion (9) exists and its radius of convergence will be finite everywhere in the ordered phase.

III. VBS ORDER IN THE NONMAGNETIC PHASE

In this section we provide additional, independent arguments against the deconfinement scenario and in favor of a first-order phase transition. We also address the question which kind of VBS order is actually realized.

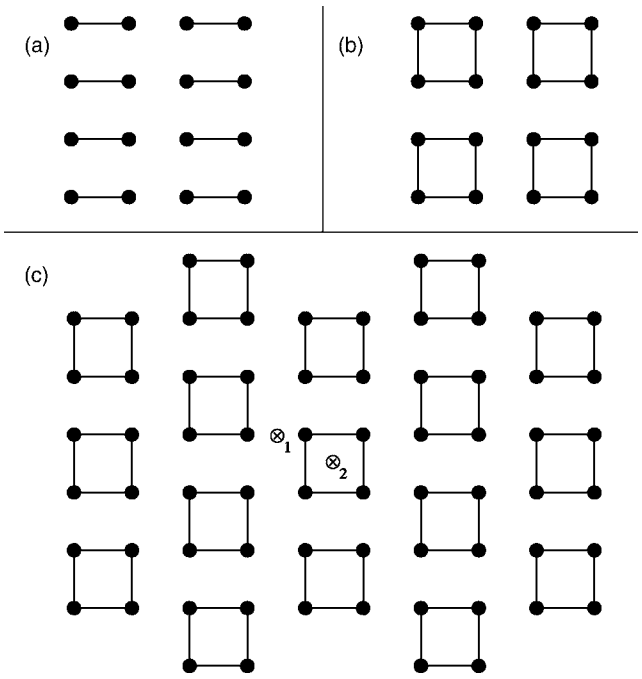

FIG. 4. Possible VBS ordering patterns: (a) columnar dimer order, (b) plaquette order, and (c) plaquette order with every second column of plaquettes shifted by one lattice site.

In Refs. 2 and 3 dealing with a possible deconfined critical point separating a Néel ordered from a VBS phase in a two-dimensional antiferromagnet it has been implicitly assumed that the VBS order is either of columnar dimer type as shown in Fig. 4(a) or of plaquette type as shown in Fig. 4(b). The order parameter for columnar dimerization is given by $O_c=(-1)^i\mathbf{S}_{i,j}\mathbf{S}_{i+1,j}$ whereas the order parameter for the plaquette phase can be represented as $O_p=(-1)^i\mathbf{S}_{i,j}\mathbf{S}_{i+1,j}+(-1)^j\mathbf{S}_{i,j}\mathbf{S}_{i,j+1}$. As discussed in Ref. 3 they can also be interpreted as a single complex order parameter where only the phase is different for the two patterns. At a deconfined critical point, however, this phase will only appear as an irrelevant operator. Therefore *both* order parameters are expected to show power law correlations at such a point and susceptibilities testing the breaking of lattice symmetries with respect to columnar or plaquette order should diverge when the deconfined critical point is approached from the magnetically ordered phase.

First, consider the field

$$F_2 = \delta \sum_{i,j} (\mathbf{S}_{i,j}\mathbf{S}_{i+1,j} - \mathbf{S}_{i,j}\mathbf{S}_{i,j+1}) \tag{10}$$

which tests if rotational symmetry is broken, which would be the case for the columnar dimer state but not for the plaquette state. The corresponding susceptibility χ_2 shows only a very moderate increase with g and certainly no sign of divergence when the possible critical point $g_{c\phi} \approx 0.43$ is approached (see Fig. 5).We mention here that our results for the susceptibilities χ_1 and χ_2 are in qualitative agreement with Ref. 5 where the finite size scaling of these susceptibilities has been investigated by exact diagonalization and varia-

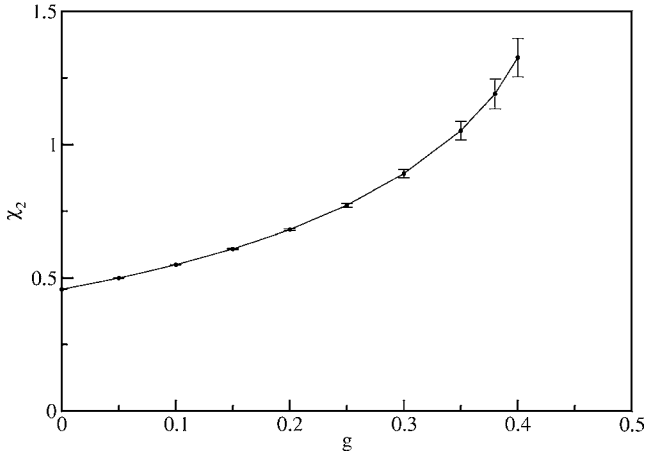

FIG. 5. Susceptibility χ_2 as a function of g calculated by Ising series expansion. The line is a guide to the eye.

tional QMC methods. Note that it does not come as a surprise that there is no quantitative agreement: the variational QMC method explores only a small part of the Hilbert space so that a convergence to the true value for the susceptibility cannot be expected even if arbitrary large clusters could be treated. If a good variational wave function is chosen, however, one might expect that the finite size scaling is at least qualitatively similar to the true finite size scaling.

Next, we consider the susceptibility χ_3 corresponding to the field

$$F_3 = \delta \sum_{i,j} (-1)^{i+j} (S^x_{i,j} S^x_{i+1,j+1} + S^y_{i,j} S^y_{i+1,j+1}) \qquad (11)$$

which has already been calculated in Ref. 11. We reproduce the result in Fig. 6. χ_3 even *decreases* when the nonmagnetic phase is approached which seems to indicate that the corresponding lattice symmetry, which would be broken for the plaquette but not for the columnar dimer state, is intact.

The fact that χ_2 and χ_3 do not diverge when $g \to g_{c\phi} \approx 0.43$ as would be expected for a deconfined critical point provides an argument independent of the considerations in

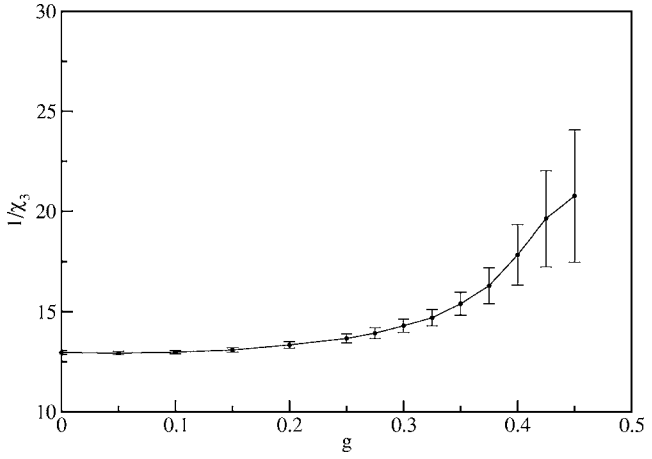

FIG. 6. Susceptibility χ_3 as a function of g calculated by Ising series expansion. The line is a guide to the eye.

Sec. II that the phase transition in the J_1-J_2 model is not an example for the scenario proposed in Refs. 2 and 3. Putting the arguments given in Sec. II aside, this alone, however, does not exclude the possibility of another kind of second-order phase transition from the Néel ordered to a state with VBS order. Nevertheless, in any second-order scenario we should take the fact that χ_2 and χ_3 do not diverge seriously. This means that in this case the VBS order cannot be of columnar dimer type because then χ_2 should diverge, and not of plaquette type because this would lead to a diverging χ_3. So in such a scenario the quest is to find an ordering pattern with lattice symmetries which agree with our findings for all three susceptibilities.

A possible pattern is shown in Fig. 4(c). Translational symmetry by one site along the x axis is broken leading to a divergent χ_1. χ_3 will be finite because of the 180° rotational symmetry around the axis marked by "1" in Fig. 4(c) or identical positions. χ_2 will be finite for this pattern because of the following symmetry: A rotation by 90° around the axis "2" or identical positions, followed by a shift of every second row of plaquettes by one lattice site, followed by a shift of every second column of plaquettes by one lattice site. Although other pattern which have the correct symmetries might be possible, the pattern in Fig. 4(c) seems to be the one with the smallest unit cell.

We have calculated the ground-state energy as well as the singlet and triplet dispersion for different g for this state by series expansion starting from the decoupled plaquettes shown in Fig. 4(c). The expansion parameter x describes the couplings between the plaquettes. For $x=0$ the plaquettes are decoupled whereas $x=1$ corresponds to the case we are finally interested in where the couplings within and between the plaquettes are of equal strength.

In Fig. 7 we show the ground-state energy obtained from this expansion compared to other series data.

The obtained energies for $0.4 \leqslant g \leqslant 0.5$ are similar or even slightly lower than the energies obtained from the columnar dimer expansion. This shows that the state in Fig. 4(c) is indeed a possible candidate for the ground state in this parameter region. In series calculations it is, however, important to make sure that the state which is the starting point for the expansion remains stable when extrapolating to the isotropic limit $x \to 1$. We have therefore calculated also the singlet and triplet dispersions. The plaquettes for this VBS state are arranged on a triangular lattice. With respect to this lattice both singlet and triplet dispersion show a minimum at momentum $\mathbf{k}=(0, 2\pi/\sqrt{3})$. The series coefficients for the singlet and triplet gaps at this point and $g=0.25, 0.45$ are given in Table I. Dlog Padé approximants for these series show that the singlet gap always vanishes before the triplet gap and at a value $x_c < 1$. This indicates that the state in Fig. 4(c) is unstable.

For the columnar dimer state, on the other hand, it is very difficult to obtain the singlet dispersion by series expansion because the state we are starting with consisting of decoupled dimers does not contain a singlet excitation. We have, however, calculated the triplet gap in this case which shows a minimum at $\mathbf{k}=(0, \pi)$ and Dlog Padé approximants suggest that in this case the gap remains nonzero for $x \to 1$. In addition, the instability of the state in Fig. 4(c) which manifests

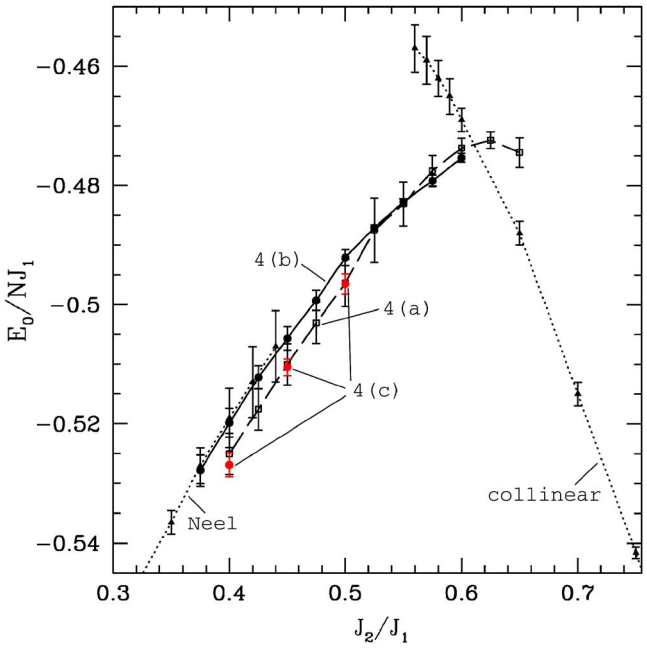

FIG. 7. (Color online) Ground state energies calculated by different series expansions starting from the nonmagnetic states shown in Fig. 4 as well as from the magnetic Néel and collinear states. The curve 4(a) (open squares) corresponds to the columnar dimer state shown in Fig. 4(a), the curve 4(b) (black dots) to the plaquette state shown in Fig. 4(b), and the curve 4(c) (red dots) to the plaquette state shown in Fig. 4(c). The black triangles show results of Ising series expansions starting from the Néel and collinear states.

itself by a vanishing of the singlet gap at $\mathbf{k}=(0,2\pi/\sqrt{3})$ for $x<1$ is an instability towards a columnar dimerization. Although this does not prove that the columnar state is finally stable, it makes the columnar dimerization pattern the most likely candidate for the VBS order in the nonmagnetic region of the phase diagram.

IV. CONCLUSIONS

We have calculated the ground-state energy e for the J_1-J_2 model including a small field F_1 with strength δ, which induces a columnar dimerization in the Néel ordered phase, by Ising series expansion. We have argued that everywhere except directly at a critical point it is possible to expand $e(\delta)$ in a regular series in δ^2 and that this series has a finite radius of convergence which goes to zero when the tentative critical point is approached. The prefactor of the δ^2-term in that series gives the susceptibility χ_1 with respect to F_1. The data for this susceptibility obtained by an Ising series expansion indicate that the Néel state becomes unstable for $g>g_c \approx 0.43$ and that the ground state for $g>g_c$ breaks translational symmetry by one site and therefore seems to be of the VBS type and not a spin liquid. Based on a mean-field treatment of an effective field theory describing the Néel state coexisting with the small dimerization induced by F_1 we have argued that the sign of the δ^4 term in the expansion of $e(\delta)$ determines whether the transition with respect to the VBS order parameter is first or second order. We believe that this is an in general more sensitive and less biased method to distinguish between a first- and a second-order transition than looking for a crossing of energies obtained by different expansions. From the series data we found that the δ^4-term has a negative sign and we showed that the same is true in spin-wave theory. Within the presented GL-type theory this means that the transition is expected to be first order. Our mean-field treatment of the order parameter is *a posteriori* justified because a critical point where such a treatment would break down is never reached.

In the second part we gave arguments in favor of a first-order transition which are independent of any effective field theory by analyzing two additional susceptibilities testing different lattice symmetries. These susceptibilities were calculated based on an Ising expansion in the Néel phase so that the series is not biased by any assumed dimerization pattern. We argued that at a deconfined critical point all three susceptibilities considered here are expected to diverge and that the fact that χ_2 and χ_3 do not diverge excludes this scenario. We further argued that in any second-order scenario the nondivergence of χ_2 would mean that the VBS state is not of the columnar dimer type and the nondivergence of χ_3 would mean that the VBS state is not of plaquette type either. For an assumed second-order transition we have been able to find a VBS pattern which does have the correct lattice symmetries to explain our data for all three susceptibilities. Series expansion data starting from this pattern, however, have proven that this state is unstable with respect to the columnar dimerization pattern.

Taking the arguments given in the two parts together shows that the transition from the Néel state to a VBS state

TABLE I. Series coefficients x^n for the minimum singlet gap Δ_s and triplet gap Δ_t for the plaquette state from Fig. 4(c) and $g=0.25$ ($g=0.45$), respectively.

n	Δ_s/J_1 ($g=0.25$)	Δ_t/J_1 ($g=0.25$)	Δ_s/J_1 ($g=0.45$)	Δ_t/J_1 ($g=0.45$)
0	1.500000000	1.000000000	1.100000000	1.000000000
1	0.000000000	-8.33333334×10^{-1}	0.000000000	-4.33333334×10^{-1}
2	$-4.356195887\times10^{-1}$	$-5.722808442\times10^{-1}$	$-4.151003339\times10^{-1}$	$-5.005966374\times10^{-1}$
3	$-3.880918282\times10^{-1}$	$1.4032850965\times10^{-1}$	$-1.832689336\times10^{-1}$	$8.2113320127\times10^{-2}$
4	$-4.828163654\times10^{-1}$	$-2.265506840\times10^{-1}$	$-1.537390367\times10^{-1}$	$-3.080455443\times10^{-2}$
5	$-6.032418702\times10^{-1}$	$7.5790435073\times10^{-3}$	$-1.592497102\times10^{-1}$	$-1.108096117\times10^{-1}$
6	$-7.988310579\times10^{-1}$	$-4.517676083\times10^{-2}$	$-1.653261118\times10^{-1}$	$1.6084176410\times10^{-2}$

with columnar dimerization is most likely first order. For a first-order phase transition we still expect that the susceptibility χ_1 when calculated by Ising series expansion diverges because the field F_1 directly tests the instability of the Néel state with respect to columnar dimer order irrespective of the order of the phase transition. For the susceptibility χ_2, on the other hand, we can only expect that it diverges if the transition is second order. The fact that χ_2 does increase with g without diverging at $g \approx 0.4$ therefore further supports that the transition is most likely first order and the VBS state most likely of the columnar dimer type.

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