Comment on "Quantum phase transition in the four-spin exchange antiferromagnet"

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In a recent paper [Phys. Rev. **B80**, 174403 (2009)] Kotov *et al.* studied the paramagnetic-to-antiferromagnetic transition in the J - Q model. Their findings were claimed to be in "fairly good agreement" with previous quantum Monte-Carlo (QMC) results. In this Comment we show that the above claim is misleading and in reality their phase transition point is not only far from the corresponding QMC value but also lies in a region of parameter space not yet explored in the literature. We also show that their reference dimer state is unstable against formation of a plaquette condensate, which could in part explain the large fluctuations they found.

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The J - Q model, proposed by Sandvik¹, is defined by the Hamiltonian:

$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \mathbf{S}_j - Q \sum_{\langle ijkl \rangle} \left(\mathbf{S}_i \mathbf{S}_j - \frac{1}{4} \right) \left(\mathbf{S}_k \mathbf{S}_l - \frac{1}{4} \right) \quad (1)$$

$$= -\frac{NQ}{8} + \left(J + \frac{Q}{2} \right) \sum_{\langle ij \rangle} \mathbf{S}_i \mathbf{S}_j - Q \sum_{\langle ijkl \rangle} (\mathbf{S}_i \mathbf{S}_j)(\mathbf{S}_k \mathbf{S}_l),$$

where $i, j \dots$ denote sites in a 2D square lattice, N is the number of spins, and $\mathbf{S}_i$ are spin-1/2 operators. The four-spin interaction Q and next-nearest neighbor exchange J were originally assumed to be positive¹. For reasons that will become clear later, we will also consider the sector with $J < 0$ (but keeping $Q > 0$). The model of Eq. (1) with $J > 0$ exhibits a quantum phase transition (QPT) between the antiferromagnetic (AF) and paramagnetic singlet phases. However, the location and nature of this transition as well as the structure of the singlet phase are still debated. In particular, in Ref. 1 it was concluded, using QMC simulations, that the QPT is 2nd order, occurs at $Q_c/J \approx 25$ and is consistent with the deconfined quantum criticality scenario². In a later work³ by the present authors, using a hierarchical mean-field (HMF) approach, the location of this QPT was found at $Q_c/J \approx 2 - 3$, and the transition itself most likely describable within the Ginzburg-Landau paradigm.

The controversy arising from this difference in values of Q_c/J was recently addressed⁴ by Kotov *et al.*. By considering effects of fluctuations around their trial (paramagnetic) columnar dimer state (CDS), they came to the conclusion that a mean-field theory is incapable of correctly describing the QPT in the J - Q model. The nature and numerical value of the phase transition point, found in their paper, was claimed to agree with the work of Sandvik¹. This circumstance was also used to speculate that the small, compared to QMC, value of Q_c/J resulting from the analysis of Ref. 3, is due to limitations of the HMF approach. We would like to stress that the HMF method is not a *standard* mean-field approach, as the one used in Ref. 4, and it becomes an exact method in the thermodynamic limit. Moreover, for a finite system HMF can be implemented as a variational theory in

Figure 1. Q_c/J ($Q_c > 0$), obtained in Refs. 1, 3 and 4.

terms of the energy, and finite-size scaling needs to be performed to extrapolate to the thermodynamic limit.

In the present note we show that the claims of Ref. 4 are misleading. We strongly oppose the statement "Near the QCP [quantum critical point], whose location [$K_c/J_K \approx 2.16$] we find in fairly good agreement with recent QMC studies...", made in Ref. 4, by demonstrating that a direct comparison of the two results is inappropriate, because the phase transition point, claimed by Kotov *et al.*, in reality lies in the yet unexplored region $J < 0$ of the J - Q phase diagram. This circumstance may raise doubts regarding the nature of the magnetic state found by Kotov *et al.* We use a simple *variational* argument to show that the large fluctuations found in their work could be attributed to an instability of their reference dimer state against formation of a plaquette condensate.

Up to an irrelevant constant, the Hamiltonian (1) can be rewritten⁴ in the form:

$$H_K = J_K \sum_{\langle ij \rangle} \mathbf{S}_i \mathbf{S}_j - K \sum_{\langle ijkl \rangle} (\mathbf{S}_i \mathbf{S}_j)(\mathbf{S}_k \mathbf{S}_l) \quad (2)$$

with $J_K, K > 0$. The new coupling constant K/J_K is related to the old one Q/J by the formula:

$$Q/J = (K/J_K) / [1 - K/2J_K] \quad (3)$$

Clearly, for positive Q and J the parameter range that can be explored with the Hamiltonian (2) is $0 \leq K/J_K \leq 2$. The values of K/J_K , larger than 2 correspond to the region $Q/J < 0$, which implies either (i) $Q < 0, J > 0$, or (ii) $Q > 0, J < 0$. We will assume, as Ref. 4, that K and Q have the same sign and disregard the case (i). The case (ii) defines the ferromagnetic (FM) part of the phase diagram of the Hamiltonian (1). In the new representation (3) the results of Refs. 1 and 3 are $K_c/J_K \approx 1.85$ and $K_c/J_K \approx 1 - 1.2$, respectively. The critical value,

Figure 2. Phase diagram of the J - Q model for $Q > 0$. The part $J > 0$ was computed in Ref. 3. The point $Q_0/J \approx -1.3$ indicates a 1st order QPT between the FM state and singlet plaquette crystal (PC) phase³. The arrow shows $Q_c/J \approx -27$, obtained in Ref. 4. The PC regions at $J < 0$ and $J > 0$ are adiabatically connected by changing J (Q/J) through zero (infinity), i.e. there is no direct FM-to-AF transition.

obtained in Ref. 4, is $K_c/J_K \approx 2.16$ which, after going back to the original units, corresponds to $Q_c/J \approx -27$. This value should be compared with the result of QMC simulations¹ $Q_c/J \approx +25$. Results of Refs. 1, 3 and 4 are summarized in Fig. 1. Thus, the claim⁴ that the transition point $K_c/J_K \approx 2.16$ is in “good agreement” with the QMC result, is unjustified. A fair comparison involves more than *numerology*, as we show below.

Since the ratio Q_c/J obtained by Kotov *et al.* is negative, one might think that in reality Ref. 4 studies the FM-to-singlet phase QPT. However, this is not necessarily the case. In fact, negative values of Q/J ($Q > 0$) do not imply a FM phase. In order to address this issue, we used the HMF approach³ and exact diagonalization (ED), both in clusters of 2×2 and 4×4 spins, to determine the J - Q phase diagram in the AF ($J > 0$) and FM ($J < 0$) regimes. The results are shown in Fig. 2. The QPT separating FM and singlet phases is 1st order: it manifests itself as a level crossing both in ED and HMF. In Table I we present numerical values for Q_0/J obtained from these methods. We see that they are in excellent agreement with each other, which is not surprising, given the fact that the FM state is semiclassical and the singlet phase is gapped. Thus, the paramagnetic phase displays *two*, FM and AF instabilities. The region $K/J_K > 2$ and the point $K_c/J_K \approx 2.16$ reside in the singlet phase of Fig. 2, at negative values of Q/J . On the other hand, the CDS⁴ can accommodate a Néel phase only if the magnetic unit cell includes *two* dimers. Ref. 4 seems to consider only homogeneous phases with one dimer per unit cell. This might raise doubts regarding the AF nature of the CDS instability discussed by Kotov *et al.*

Finally, we shall comment on the huge fluctuation cor-

Table I. Numerical values for the 1st order transition point Q_0/J , obtained using ED and HMF for different cluster sizes.

	2×2	4×4
ED	-1.0	-1.19
HMF	-1.32	-1.26

Figure 3. Energy (5) as a function of θ . The plaquette state corresponds to a local minimum $\varepsilon_0(5\pi/6) = -73/96$. The columnar dimer configuration has a higher energy $\varepsilon_0(0) = -66/96$ and does not correspond to an extremal point of $\varepsilon_0(\theta)$.

rections to the value of K_c , found in Ref. 4. These corrections were argued to be responsible for shifting the QPT towards negative values of Q/J and seem to be intimately related to the use by Kotov *et al.* of the CDS as a physical vacuum for their analysis. This dimer state is unstable, when compared to the plaquette structure³. Let us consider a trial paramagnetic state of the form:

$$|\Psi_0\rangle = \prod_{\square} [\cos \theta |\psi_d\rangle + \sin \theta (|\psi_{\square}\rangle - \langle \psi_d | \psi_{\square} \rangle |\psi_d\rangle)], \quad (4)$$

where the product runs over $N/4$ 2×2 plaquettes. The wavefunction of Eq. (4) interpolates between the dimer condensate⁴ $\prod |\psi_d\rangle$ ($|\psi_d\rangle$ is the direct product of spin singlets on two parallel links of a plaquette), for $\theta = 0$, and the plaquette state³ $\prod |\psi_{\square}\rangle$, which corresponds to $\theta = 5\pi/6$. The expectation value of Hamiltonian (1), $\varepsilon_0(\theta) \equiv \langle \Psi_0(\theta) | H | \Psi_0(\theta) \rangle / NQ$, is given by:

$$\varepsilon_0(\theta) = -\left(\frac{J}{8Q}\right) (\sin \theta - \sqrt{3} \cos \theta)^2 - \frac{1}{96} \times \quad (5) \\ \times (51 + 13 \cos 2\theta + 2 \cos 4\theta - 13\sqrt{3} \sin 2\theta + 2\sqrt{3} \sin 4\theta).$$

For $Q \gg J$, this function is shown in Fig. 3. The plaquette state corresponds to a local energy minimum. On the contrary, the CDS does not describe any extremal point. Moreover, it has a higher energy compared to the plaquette configuration. The energy difference between the CDS and a correlated plaquette structure will be even larger if 4×4 spin clusters³ are used as a basis for the HMF analysis.

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