# Ising Transition in the Two-Dimensional Quantum $J_1 - J_2$ Heisenberg Model

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(Received 2 December 2003; published 15 April 2004)

We study the thermodynamics of the spin-$S$ two-dimensional quantum Heisenberg antiferromagnet on the square lattice with nearest ($J_1$) and next-nearest ($J_2$) neighbor couplings in its collinear phase ($J_2/J_1 > 0.5$), using the pure-quantum self-consistent harmonic approximation. Our results show the persistence of a finite-temperature Ising phase transition for every value of the spin, provided that the ratio $J_2/J_1$ is greater than a critical value corresponding to the onset of collinear long-range order at zero temperature. We also calculate the spin and temperature dependence of the collinear susceptibility and correlation length, and we discuss our results in light of the experiments on $Li_2VOSiO_4$ and related compounds.

DOI: 10.1103/PhysRevLett.92.157202 PACS numbers: 75.10.Jm, 05.30.–d, 75.40.–s, 75.40.Cx

The study of frustrated quantum spin systems is one of the most challenging and exciting topics in theoretical magnetism. A very extensively investigated, yet largely debated model is the so-called $J_1 - J_2$ Heisenberg model with competing antiferromagnetic couplings ($J_1, J_2 > 0$) between nearest-neighbors (NN) and next-nearest-neighbors (NNN)

$$\hat{\mathcal{H}} = J_1 \sum_{\mathrm{NN}} \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j + J_2 \sum_{\mathrm{NNN}} \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j, \qquad (1)$$

where $\hat{\boldsymbol{S}}_i$ are spin-$S$ operators on a periodic lattice with $N = L \times L$ sites; hereafter $\alpha = J_2/J_1$ defines the frustration ratio.

In the classical limit ($S \to \infty$), the minimum energy configuration has conventional Néel order with magnetic wave vector $\boldsymbol{Q} = (\pi, \pi)$ for $\alpha < 0.5$. Instead, for $\alpha > 0.5$, the antiferromagnetic order is established independently on the two sublattices, with the two (staggered) magnetizations free to rotate with respect to each other. Among this degenerate manifold, two families of *collinear states,* with pitch vectors $\boldsymbol{Q} = (\pi, 0)$ or $(0, \pi)$, are selected by an *order-by-disorder* mechanism as soon as thermal or quantum fluctuations are taken into account. As a result, for $\alpha > 0.5$ the classical ground state breaks not only the spin rotational and translational invariance of the Hamiltonian — as the conventional Néel phase — but also its invariance under $\pi/2$ lattice rotations, the resulting degeneracy corresponding to the group $O(3) \times Z_2$. Remarkably, the additional *discrete* $Z_2$ symmetry can, in principle, be broken at finite temperatures without violating the Mermin-Wagner theorem [1], which applies in two dimensions only to continuous ones. On this basis, in a seminal paper [2], Chandra, Coleman, and Larkin (CCL) proposed that the two-dimensional $J_1 - J_2$ model could sustain an Ising phase transition at finite temperature, with an order parameter directly related to the $Z_2$ degree of freedom induced by frustration. They also provided quantitative estimates of the critical temperatures in the large-$\alpha$ limit for both the classical and the quantum case.

While the CCL transition in the classical model has been recently established by an extensive Monte Carlo (MC) study [3], the occurrence of a low-temperature phase with a discrete broken symmetry in the quantum case is still a subject of debate. Besides its own theoretical interest, this issue has become particularly important in connection with the discovery of three vanadate compounds ($Li_2VOSiO_4$, $Li_2VOGeO_4$, and $VOMoO_4$) whose relevant magnetic interactions involve nearest and next-nearest spin-1/2 $V^{4+}$ ions on weakly coupled stacked planes [4,5]. In particular, NMR and $\mu$SR(muon spin resonance) measurements on $Li_2VOSiO_4$ [4] indicate the occurrence of a transition to a low-temperature phase with collinear order at $T_N \simeq 2.8$ K. The selection of the collinear-ground state suggests the CCL mechanism as a possible underlying explanation. Unfortunately, a clear experimental and theoretical picture is still elusive: in the experiments with vanadate compounds, structural distortions, interlayer coupling, and anisotropy effects are likely to come into play [4], and on the other hand the theoretical investigation cannot rely on the insight provided by quantum Monte Carlo methods as their reliability in the presence of frustration is strongly limited by the infamous *sign problem* [6].

The necessary condition for the CCL transition to take place is the presence of collinear order at zero temperature. To this respect, the existent theoretical results [6] point towards a collinear-ground state for frustration ratios $\alpha > \alpha_c$, with the critical value, $\alpha_c$, increasing as the value of the spin decreases (for $S = 1/2$, $\alpha_c \simeq 0.6$). Below this value quantum fluctuations seem to be strong enough to stabilize a low-temperature phase with short-range magnetic correlations [7], but above it a CCL transition is, in principle, possible. However, this possibility has

  

been recently challenged by various high-temperature expansion studies [8–10] which were not able to detect any evidence of a finite-temperature transition for $S = 1/2$. On this basis, Singh *et al.* [9] have recently proposed a scenario where, due to quantum fluctuations, the broken lattice symmetry of the collinear-ground state would be restored at any nonzero temperature.

In this Letter, we present a complete study of the thermodynamic properties of the quantum $J_1 - J_2$ model in its collinear phase, obtained within an effective Hamiltonian approach: the pure-quantum self-consistent harmonic approximation (PQSCHA) [11]. This approach, based on the path-integral formalism, allows one to separate the classical from the pure-quantum contribution to the thermodynamics of the system. Both the classical physics and the purely quantum linear effects are exactly described within the PQSCHA at *any* temperature, while the purely quantum nonlinear contributions are treated within a self-consistent harmonic approximation. This feature makes the PQSCHA a valid tool to investigate the effects of quantum fluctuations on phase transitions—like the CCL one—whose character is essentially classical. In particular, this approach has been successfully applied to a variety of spin systems displaying Kosterlitz-Thouless and/or Ising critical behaviors, providing reliable estimates of the transition temperatures even for $S = 1/2$ [12].

Within the PQSCHA framework, the thermodynamics of a quantum system is rephrased in terms of a classical effective Hamiltonian with renormalized parameters depending on the spin value, temperature, and frustration. The derivation of the effective Hamiltonian for the $J_1 - J_2$ model closely follows the steps shown in Ref. [11]. The only detail which is worth mentioning here is that, in this case, the calculation of quantum renormalizations, involving a harmonic expansion around one of the two families of collinear states, gives rise in general to solutions with an explicitly broken symmetry under $\pi/2$ lattice rotations. However, it is possible to show that, to $O(1/S)$, the effective Hamiltonian can be recast in a form preserving all the symmetries of the original model, and that reads (except for uniform terms)

$$\mathcal{H}^{\text{eff}} = J_1^{\text{eff}} \tilde{S}^2 \sum_{\text{NN}} \boldsymbol{s}_i \cdot \boldsymbol{s}_j + J_2^{\text{eff}} \tilde{S}^2 \sum_{\text{NNN}} \boldsymbol{s}_i \cdot \boldsymbol{s}_j, \qquad (2)$$

where $\boldsymbol{s}_i$ are classical vectors of length 1, $\tilde{S} = S + \frac{1}{2}$ is the effective spin length [11], and $J_1^{\text{eff}} = (\theta_x^2 + \theta_y^2)\theta_2^2 J_1/2$, $J_2^{\text{eff}} = \theta_2^4 J_2$ are the quantum-renormalized exchange integrals, with spin-, temperature-, and frustration-dependent renormalization parameters $\theta_x$, $\theta_y$, and $\theta_2$. These, e.g., referred to the ground state with $\boldsymbol{Q} = (\pi, 0)$, are given by $\theta_\alpha^2 = 1 - \mathcal{D}_\alpha/2$ ($\alpha = x, y, 2$) where the coefficients

$$\mathcal{D}_\alpha = \frac{1}{N\tilde{S}} \sum_{\boldsymbol{k}} \frac{a_{\boldsymbol{k}}^+}{a_{\boldsymbol{k}}^-} (1 - \gamma_{\boldsymbol{k}}^{(\alpha)}) \mathcal{L}_{\boldsymbol{k}} \qquad (3)$$

are self-consistently determined with

$$\frac{a_{\boldsymbol{k}}^\pm}{2\tilde{S}\sqrt{J_1}} = \sqrt{\alpha\theta_2^2(1 \pm \gamma_{\boldsymbol{k}}^{(2)}) + \frac{\theta_{xy}^2}{2} \pm \frac{\theta_x^2}{2}\gamma_{\boldsymbol{k}}^{(x)} + \frac{\theta_y^2}{2}\gamma_{\boldsymbol{k}}^{(y)}},$$

$$\mathcal{L}_{\boldsymbol{k}} = \coth f_{\boldsymbol{k}} - \frac{1}{f_{\boldsymbol{k}}}, \qquad f_{\boldsymbol{k}} = \frac{\hbar\omega_{\boldsymbol{k}}}{2\tilde{S}k_{\text{B}}T}, \qquad (4)$$

where $\theta_{xy}^2 = \theta_x^2 - \theta_y^2$, $\gamma_{\boldsymbol{k}}^{(x,y)} = \cos k_{x,y}$, $\gamma_{\boldsymbol{k}}^{(2)} = \cos k_x \cos k_y$, $T$ is the temperature, and the renormalized dispersion relation is $\omega_{\boldsymbol{k}} = a_{\boldsymbol{k}}^+ a_{\boldsymbol{k}}^-$. Interestingly, at $T = 0$ the PQSCHA turns out to be equivalent to the modified spin-wave theory [9,13] which is expected to be a faithful representation of the low-energy properties in the collinear phase. In particular, the spin-wave dispersion relation $\omega_{\boldsymbol{k}}$ has been recently shown to be in remarkable quantitative agreement with series expansion results in the entire range of momenta for $\alpha > \alpha_c$, and it is therefore expected to provide an accurate description of the low-energy excitations of the model [9].

The occurrence of the CCL transition in the quantum case can be directly addressed within our approach by calculating the critical temperatures as functions of the spin and of the frustration ratio: a zero value of the critical temperature, or the breakdown of the self-consistent harmonic treatment of quantum fluctuations, would signal a possible absence of the phase transition. In particular, using a simple scaling argument the critical temperatures in the quantum case $T_c(S, \alpha)$ can be related to those of the classical model $T_c^{(\text{cl})}(\alpha)$ through the following self-consistent relation [12]:

$$T_c(S, \alpha) = j_1^{\text{eff}}(T_c, S, \alpha) T_c^{(\text{cl})}[\alpha^{\text{eff}}(T_c, S, \alpha)], \qquad (5)$$

where $j_1^{\text{eff}} = J_1^{\text{eff}} \tilde{S}^2/J_1$ and $\alpha^{\text{eff}} = J_2^{\text{eff}}/J_1^{\text{eff}}$. The classical transition temperature $T_c^{(\text{cl})}(\alpha)$ is accurately known through extensive MC simulations for $\alpha \leq 2$; it vanishes for $\alpha \to 1/2$ and grows more or less linearly for $\alpha > 1$. Beyond $\alpha \simeq 2$ the determination of the critical temperatures becomes troublesome due to severe finite-size effects related to the width of the domain walls between domains with $\boldsymbol{Q} = (\pi, 0)$ and $\boldsymbol{Q} = (0, \pi)$ [3]. However, for large values of the frustration ratio the classical CCL estimate of the critical temperature, $T_c/J_2 = 0.768/[1 + 0.135\ln(J_2/J_1)]$, is expected to be reliable [2,3]. This vanishes logarithmically in the limit $J_2/J_1 \to \infty$, corresponding to two decoupled unfrustrated Heisenberg systems. In order to represent the whole interval of $\alpha \in [1/2, \infty)$ in Fig. 1 we have plotted both the MC and the CCL estimates of the classical critical temperatures as a function of $\alpha/(1 + \alpha)$. The mismatch between the MC and CCL predictions is a minor flaw that can be easily accounted for and corrected by slightly modifying CCL's criterion for the determination of the transition temperature as explained in Ref. [3]. This gives rise to the curve marked in Fig. 1 as CCL*. Using the latter estimate, and the classical MC results of Ref. [3], the transition temperatures in the quantum case can be determined by

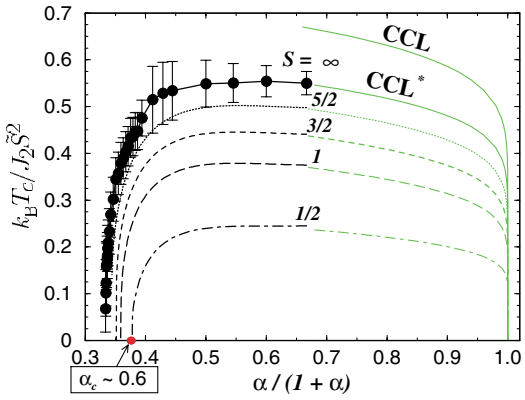


FIG. 1 (color online). Renormalized critical temperature of the CCL transition for various values of the spin evaluated using Eq. (5) (nonsolid lines). Classical data (●) are taken from Ref. [3]. The solid lines on the right are the CCL and the CCL* predictions for the classical case (see text). The arrow marks the boundary of the nonmagnetic phase for $S = 1/2$ [14].

numerically solving Eq. (5). As shown in Fig. 1, the critical temperatures decrease as the spin decreases, due to the enhancement of quantum fluctuations. Remarkably, while for large $\alpha$ the transition temperature in units of $J_2$ vanishes for $\alpha \to \infty$ for any value of the spin, in the opposite limit the critical temperatures vanish approaching a critical value $\alpha_c > 0.5$ that increases as $S$ decreases, thus confirming the existence of a nonmagnetic phase in the regime of high frustration. In particular, for $S = 1/2$, $\alpha_c \simeq 0.6$ in agreement with the previous estimates of the zero-temperature quantum critical point [6]. For large $\alpha$, the PQSCHA results turn out to be consistent with the CCL estimates for the quantum $S = 1/2$ system [2,3]. For intermediate values of $\alpha$, the transition temperatures remain finite for any spin value. However for $S = 1/2$, though sizable ($T_c/J_2 \simeq 0.2$), the critical temperatures are more than an order of magnitude smaller than those considered in Ref. [9] in the discussion of the high-temperature expansion results leading to the proposal of a $T = 0$ critical scenario. On the contrary, a finite-temperature phase transition at the critical temperatures we estimate is in fact consistent with the numerical results of Ref. [9].

The study of the thermodynamics of the effective classical model (2) provides a deeper analysis of the CCL transition. To this end, we have performed classical MC simulations on the effective Hamiltonian on $L \times L$ lattices, with $L$ up to 300. The PQSCHA expression for the order parameter associated to the CCL transition [2] is $\sigma = \theta_2^2(\theta_x^2 + \theta_y^2)\langle|(\boldsymbol{s}_1 - \boldsymbol{s}_3) \cdot (\boldsymbol{s}_2 - \boldsymbol{s}_4)|\rangle_{\text{eff}}/8$, where (1, 2, 3, 4) are the sites of an elementary plaquette of the lattice with counterclockwise numbering, and $\langle \cdots \rangle_{\text{eff}}$ is the statistical average associated to the effective classical Hamiltonian (2). Another quantity bearing clear signatures of the transition is the specific heat, calculated here as the numeric derivative of the internal energy per spin, $c = \partial u/\partial T$, where $u = \langle \mathcal{H}_{\text{eff}} \rangle / N - J_1 \tilde{S}^2(\theta_x^4 - \theta_y^4)$.

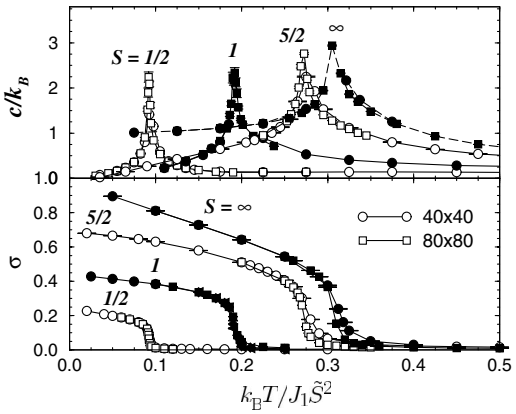


FIG. 2. PQSCHA results for the specific heat (upper panel) and the order parameter $\sigma$ (lower panel) as functions of temperature for various spins, in the case $\alpha = 0.65$.

These quantities are shown for $\alpha = 0.65$ in Fig. 2. The Ising transition is clearly marked by a (finite-size) peak in the specific heat in correspondence to the temperature where the Ising order parameter vanishes. By decreasing the spin, the saturated values of $\sigma$ decrease as well, due to the increased quantum fluctuations, and so does the peak of the specific heat, making the transition in the quantum case generally weaker than in the classical one. When $\alpha$ is increased, the signatures of the transition are even more dramatically suppressed, due to the mentioned effects related to the width of the domain walls [3]. The Heisenberg features become more and more important with respect to the frustration effects; in fact, as shown in Fig. 3, already for $\alpha = 1$, the Ising peak of the specific heat tends to be masked by the broad maximum reminiscent of the unfrustrated limit $\alpha \to \infty$, and very large values of $L$ are required in order to resolve the logarithmic divergence (inset). Similarly, the absence of critical features in the recent numerical calculations of the specific heat [10] can be traced back to the

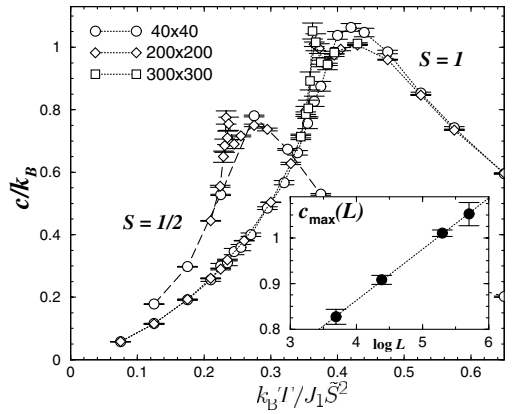


FIG. 3. PQSCHA results for the specific heat in the cases $\alpha = 1$, $S = 1, 1/2$. Inset: scaling of the specific heat at the peak temperature $k_B T/J_1\tilde{S}^2 = 0.36$ for $S = 1$.

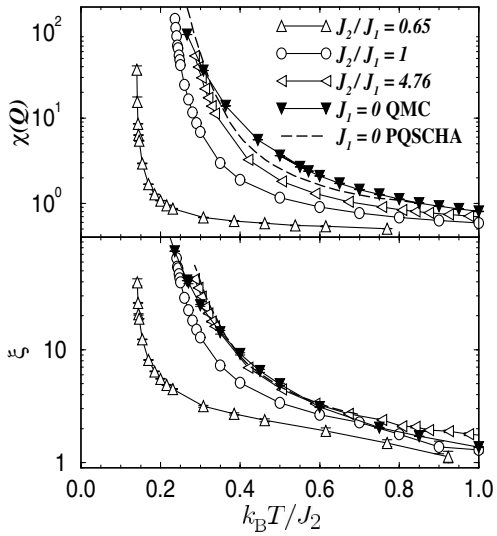


FIG. 4. Upper panel: $S = 1/2$ staggered susceptibility, with $\boldsymbol{Q} = (\pi, 0)$ and $(\pi, \pi)$ in the frustrated (open symbols) and unfrustrated cases, respectively. Lower panel: $S = 1/2$ correlation length. The data for the unfrustrated cases are taken from Ref. [11] and partly from Ref. [17].

limited range of the correlators generated by the high-temperature expansion [15].

In order to investigate the effect of frustration on the antiferromagnetic correlations, we have also calculated the collinear susceptibility

$$\chi(\mathbf{Q}) = \frac{S(S+1)}{3} + \frac{\theta_0^4 \tilde{S}^2}{3} \sum_{\mathbf{r} \neq \mathbf{0}} e^{i\boldsymbol{Q}\cdot\mathbf{r}} \langle \boldsymbol{s}_0 \cdot \boldsymbol{s}_r \rangle_{\text{eff}}, \qquad (7)$$

with $\mathbf{Q} = (\pi, 0)$ or $(0, \pi)$, and the corresponding correlation length (through the second moment estimator [16]). The results for the case $S = 1/2$ for different values of $\alpha$ are shown in Fig. 4. We observe that both quantities are strongly suppressed by frustration for $\alpha \lesssim 1$, but for larger $\alpha$ the results are extremely close to those in the unfrustrated Heisenberg model, corresponding to $\alpha \to \infty$. Therefore neutron scattering experiments on $Li_2VOSiO_4$ and related compounds with a large $\alpha$ are not likely to show any strong frustration effects in the magnetic correlations, as also indicated by preliminary experimental results [18].

Finally, we compare the estimated critical temperature for $S = 1/2$ with the transition temperature, $T_N = 2.8$ K, observed in $Li_2VOSiO_4$. For this compound, various ratios $J_2/J_1$ have been estimated, ranging from $J_2/J_1 = 1.1$ with $J_1 = 3.9$ K [4] to $J_2/J_1 = 4.76$ with $J_1 = 1.25$ K [10]. Using these estimates, we get $T_c \approx 1.01$ and 1.46 K, respectively, which are well below the transition to three-dimensional collinear order observed at $T_N \simeq 2.8$ K. The CCL critical behavior is therefore not detectable in this compound [4]. In general, due to the weak nature of the transition for $J_2/J_1 \gtrsim 1$, previously discussed, the observation of the CCL transition would require realizations of a $J_1 - J_2$ model with a smaller value of the ratio $J_2/J_1$.

In conclusion, using the pure-quantum self-consistent harmonic approximation, we have provided a complete and consistent picture of the collinear phase of the 2D quantum $J_1 - J_2$ antiferromagnet. Our results indicate that the finite-temperature transition predicted by Chandra, Coleman, and Larkin [2] persists down to $S = 1/2$ provided that the ratio $J_2/J_1$ is greater than a critical value corresponding to the stabilization of collinear long-range order in the ground state. We believe that our findings will be a valid reference point for future experimental investigations on $Li_2VOSiO_4$ and related compounds.

We thank F. Becca, P. Carretta, A. Cuccoli, G. Misguich, and F. Mila for fruitful discussions and correspondence. This work was supported by NSF under Grant No. DMR02-11166 (L. C.), by DOE under Grant No. DE-FG03-01ER45908 (T. R.), and by INFN, INFM, and MIUR-COFIN2002 (A. F. and V. T.).

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