Low-Temperature Broken-Symmetry Phases of Spiral Antiferromagnets

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(Received 23 September 2004; published 14 December 2004)

We study Heisenberg antiferromagnets with nearest- (J_1) and third- (J_3) neighbor exchange on the square lattice. In the limit of spin $S \to \infty$, there is a zero temperature (T) Lifshitz point at $J_3 = \frac{1}{4}J_1$, with long-range spiral spin order at $T = 0$ for $J_3 > \frac{1}{4}J_1$. We present classical Monte Carlo simulations and a theory for $T > 0$ crossovers near the Lifshitz point: spin rotation symmetry is restored at any $T > 0$, but there is a broken lattice reflection symmetry for $0 \leq T < T_c \sim (J_3 - \frac{1}{4}J_1)S^2$. The transition at $T = T_c$ is consistent with Ising universality. We also discuss the quantum phase diagram for finite S.

DOI: 10.1103/PhysRevLett.93.257206 PACS numbers: 75.10.Hk, 75.10.Jm

Frustrated antiferromagnets have recently attracted much interest in connection with the possibility of stabilizing unconventional low-temperature (T) phases, with novel types of "quantum order" [1]. A very promising candidate for a *spin-liquid* phase is the $J_1 - J_3$ model

$$\hat{H} = J_1 \sum_{\langle i,j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + J_3 \sum_{\langle\langle i,j \rangle\rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j, \qquad (1)$$

where $\hat{\mathbf{S}}_i$ are spin-S operators on a square lattice and $J_1, J_3 \geq 0$ are the nearest- and third-neighbor antiferromagnetic couplings along the two coordinate axes. For this model, early large N computations, [2] and recent large scale density matrix renormalization group (DMRG) calculations for $S = 1/2$ [3] have suggested the existence of a gapped spin-liquid state with exponentially decaying spin correlations and no broken translation symmetry in the regime of strong frustration ($J_3/J_1 \simeq 0.5$).

This Letter will describe properties of the above model for large S and discuss consequences for general S. Our results, obtained by classical Monte Carlo simulations and a theory described below, are summarized in Fig. 1 for the limit $S \to \infty$. There is a $T = 0$ state with long-range spiral spin order for $J_3 > \frac{1}{4}J_1$. We establish that at $0 < T < T_c \sim (J_3 - \frac{1}{4}J_1)S^2$, above this state there is a phase with broken discrete symmetry of lattice reflections about the x and y axes, while spin rotation invariance is preserved. This phase has "Ising nematic" order. We present strong numerical evidence that the transition at T_c is indeed in the Ising universality class. Such Ising nematic order[4] was originally proposed in Ref. [2] for $S = 1/2$ in a $T = 0$ spin-liquid phase described by a Z_2 gauge theory [5]. Thus the same Ising nematic order can appear when spiral spin order is destroyed either by thermal fluctuations (as in the present Letter; see Fig. 1) or by quantum fluctuations (as in Ref. [2]). Our large S results are therefore consonant with the possibility of a spin-liquid phase at $S = 1/2$ as described in Refs. [2,3]; we will discuss the quantum finite S phase diagram further towards the end of the Letter. We also suggest that discrete lattice symmetries may play a role near other quantum critical points with spiral order [6].

Broken discrete symmetries have also been discussed [7,8] in the context of the $J_1 - J_2$ model, with first- and second-neighbor couplings on the square lattice. However, this model has only collinear, commensurate spin correlations, and this makes both the classical and quantum theory quite different from that considered here. As will become clear below, the spiral order and associated Lifshitz point play a central role in the structure of our theory and in the T dependence of observables.

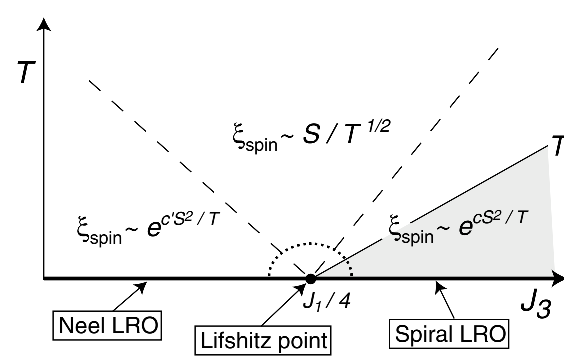

FIG. 1. Phase diagram of $\hat{H}$ in the limit $S \to \infty$. The shaded region has a broken symmetry of lattice reflections about the x and y axes, leading to Ising nematic order. The Ising transition is at the temperature $T_c \sim (J_3 - \frac{1}{4}J_1)S^2$. The spin correlation length, ξ_{spin}, is finite for all $T > 0$, with the T dependencies as shown, with $c/2 = c' = 8\pi|J_3 - \frac{1}{4}J_1|$; the crossovers between the different behaviors of ξ_{spin} are at the dashed lines at $T \sim |J_3 - \frac{1}{4}J_1|S^2$. Spin rotation symmetry is broken only at $T = 0$ where $\xi_{\text{spin}} = \infty$. There is no Lifshitz point at finite S because it is preempted [13] by quantum effects within the dotted semicircle: here there is a $T = 0$ spin gap $\Delta \sim S \exp(-\tilde{c}S)$ and spin rotation symmetry is preserved. This semicircular region extends over $T \sim |J_3 - \frac{1}{4}J_1|S \sim \Delta$. Further details on the physics within this region appear at the end of the Letter.

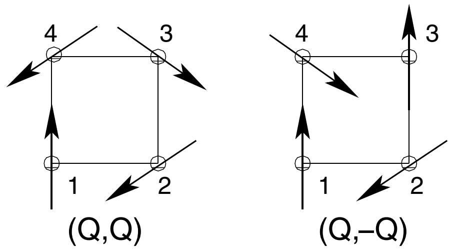

FIG. 2. The two different minimum energy configurations with magnetic wave vectors $\vec{Q} = (Q, Q)$ and $\vec{Q}^\star = (Q, -Q)$ with $Q = 2\pi/3$, corresponding to $J_3/J_1 = 0.5$.

We begin by recalling [9] the ground states of $\mathcal{H}$ at $S = \infty$. There is conventional Néel order with magnetic wave vector $\vec{Q} = (\pi, \pi)$ for $J_3/J_1 \leq \frac{1}{4}$. For $J_3/J_1 > \frac{1}{4}$, the ground state has planar incommensurate antiferromagnetic order at a wave vector $\vec{Q} = (Q, Q)$, with Q decreasing from π as $J_3/J_1 > \frac{1}{4}$ and approaching $Q = \pi/2$ monotonically for $J_3/J_1 \to \infty$. The spiral order is incommensurate for $\frac{1}{4} < J_3/J_1 < \infty$, except at $J_3/J_1 = 0.5$ where $Q = 2\pi/3$, corresponding to an angle of 120 ° between spins (see Fig. 2). Interestingly, for each spiral state with $\vec{Q} = (Q, Q)$ there is a distinct but equivalent configuration at $\vec{Q}^\star = (-Q, Q)$ (for $Q \neq \pi$). This state cannot be obtained from the one with wave vector $\vec{Q}$ by a global spin rotation. Instead, the two configurations are connected by a global rotation combined with a reflection about the x or y axes. The global symmetry of the classical ground state is O(3) $\times$ Z_2, with an additional twofold degeneracy beyond that of the Néel case.

One of the main claims in Fig. 1 is that the broken Z_2 symmetry survives for a finite range of $T > 0$, while continuous O(3) symmetry is immediately restored at any nonzero T. We established this by extensive Monte Carlo simulation using a combination of Metropolis and overrelaxed algorithm for periodic clusters of size up to $M = 120 \times 120$, and for several values of J_3/J_1 between 0.25 and 4. Indeed, the presence of a finite T phase transition is clearly indicated by a sharp peak of the specific heat which is illustrated in Fig. 3 [10]. This sharp feature is to be contrasted to the broad maximum displayed by the same quantity for $J_3/J_1 < \frac{1}{4}$, i.e., when the classical ground state displays ordinary Néel order. In particular, the maximum of the specific heat is consistent with a logarithmic dependence on system size (see the inset of Fig. 3) corresponding to a critical exponent $\alpha = 0$, in agreement with Ising universality.

This critical behavior can be directly related to the broken lattice reflection symmetry by studying an appropriate Ising nematic order parameter. From the symmetries of Fig. 2, we deduce that the order parameter is $\sigma = 1/M(\sum_a \sigma_a)$ with

$$\sigma_a = (\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_3 - \hat{\mathbf{S}}_2 \cdot \hat{\mathbf{S}}_4)_a, \tag{2}$$

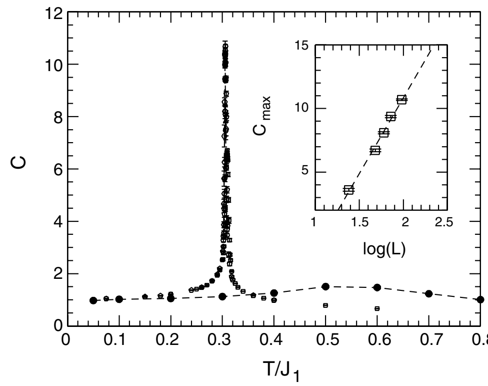

FIG. 3. T dependence of the specific heat for $J_3/J_1 = 0.5$. Different symbols refer to different clusters with liner size between $L = 24$ and $L = 120$. Data for $J_3/J_1 = 0.1$ are shown for comparison (full dots and dashed line). Inset: size scaling of the maximum of the specific heat.

where a labels each plaquette of the square lattice and $(1, 2, 3, 4)$ are its corners. The variables σ_a are zero for a Néel antiferromagnet, while they assume opposite signs on the two degenerate ground states in the spiral phase. Consequently, a phase with Ising nematic order is signaled by a $\langle \sigma_a \rangle \neq 0$.

Our numerical results contain strong evidence for a continuous Ising phase transition between a low T phase with $\langle \sigma \rangle \neq 0$, and a homogeneous high T phase with $\langle \sigma \rangle = 0$. The divergence in the specific heat (Fig. 3) is accompanied by a divergence in the susceptibility of the

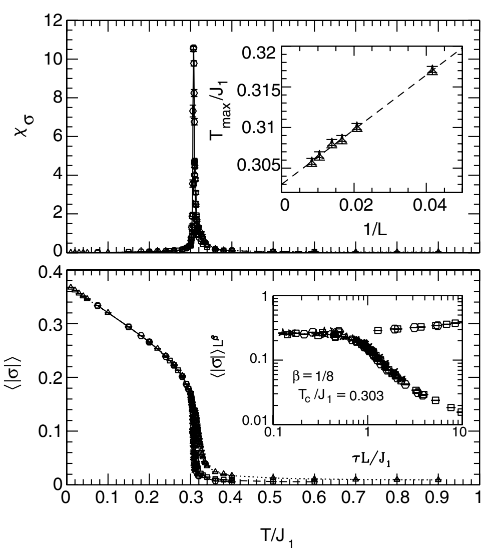

FIG. 4. Bottom: T dependence of the order parameter, σ, [see Eq. (2)] for different cluster sizes and $J_3/J_1 = 0.5$. The inset shows the data collapse according to the scaling hypothesis with Ising exponents $\beta = 1/8$ and $\nu = 1$, and $T_c = 0.303$. Top: temperature dependence of the susceptibility of σ for $J_3/J_1 = 0.5$. The inset shows the size scaling of the temperature corresponding to the maximum of the susceptibility.

Ising nematic order parameter $\chi_\sigma = (M/T) \times (\langle\sigma^2\rangle - \langle|\sigma|\rangle^2)$ (Fig. 4, upper panel), and by universal T dependence in Binder's fourth cumulant $U_4 = 1 - \langle\sigma^4\rangle/3\langle\sigma^2\rangle^2$ (not shown). The critical exponent ν can be estimated from the size dependence of the T corresponding to the maximum of the susceptibility, which is expected to scale as $T_{\max}(L) = T_c + aL^{-1/\nu}$, where T_c is the thermodynamic critical temperature; Fig. 4 (upper inset) shows $\nu = 1$, as expected. The exponent β is also in agreement with Ising universality. This can be extracted from the scaling law $|\sigma| = L^{-\beta/\nu} f(x)$, where $f(x)$ is the scaling function and $x = \tau L^{1/\nu}$ with $\tau = |T - T_c|$; this scaling is shown in Fig. 4, upper inset, with $T_c = 0.303(1)$ estimated from the position of the maximum of the susceptibility and the behavior of the Binder's cumulant. Excellent data collapse is obtained for $\beta/\nu = 1/8$.

We have repeated a similar analysis for several values of J_3/J_1 and the complete phase diagram is shown in Fig. 5, where we have plotted T_c versus J_3/J_1. We find that T_c vanishes linearly for $J_3/J_1 \to 1/4$; a theory for this behavior will now be presented.

Near the classical Lifshitz point, we can model quantum and thermal fluctuations by a continuum unit vector field $\mathbf{n}(r, \tau)$, where $r = (x, y)$ is spatial coordinate, τ is imaginary time, and $\mathbf{n}^2 = 1$ at all r, τ. This field is proportional to the Néel order parameter with $\hat{\mathbf{S}}_j \propto (-1)^{x_j + y_j}\mathbf{n}(r_j, \tau)$. Spiral order will therefore appear as sinusoidal dependence of $\mathbf{n}$ on r. The action for $\mathbf{n}$ is the conventional O(3) nonlinear sigma model, expanded to include quartic gradient terms ($\hbar = k_B =$ lattice spacing $= 1$): $S_\mathbf{n} = \int_0^{1/T} d\tau \int d^2 r \mathcal{L}_\mathbf{n}$ with

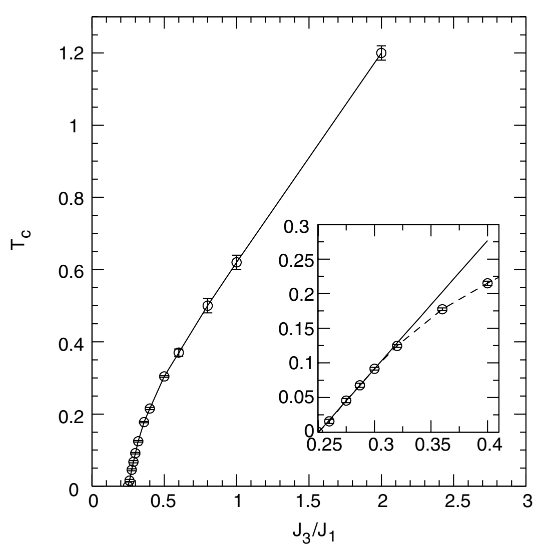

FIG. 5. Critical temperature as a function of the frustration ratio J_3/J_1.

$$\mathcal{L}_\mathbf{n} = \frac{\chi_\perp}{2}(\partial_\tau \mathbf{n})^2 + \frac{\rho}{2}[(\partial_x \mathbf{n})^2 + (\partial_y \mathbf{n})^2] + \frac{\zeta_1}{2}[(\partial_x^2 \mathbf{n})^2 + (\partial_y^2 \mathbf{n})^2] + \zeta_2 \partial_x^2 \mathbf{n} \cdot \partial_y^2 \mathbf{n} + \lambda_1[(\partial_x \mathbf{n} \cdot \partial_y \mathbf{n})^2 + (\partial_y \mathbf{n} \cdot \partial_y \mathbf{n})^2] + \lambda_2(\partial_x \mathbf{n} \cdot \partial_y \mathbf{n})^2 \cdots \quad (3)$$

where the ellipses denote a finite number of additional λ_i couplings involving four powers of $\mathbf{n}$ and four spatial derivatives invariant under spin rotations and lattice symmetries. In the limit $S \to \infty$, we have $\chi_\perp = 1/(8J_1)$, $\rho = (J_1 - 4J_3)S^2$, $\zeta_1 = (16J_3 - J_1)S^2/12$, $\zeta_2 = 0$, and all $\lambda_i = 0$. Notice that ρ crosses zero at the Lifshitz point and so can be regarded as the tuning parameter; $\rho = 0$ generally locates the Lifshitz point for when $\rho < 0$ it is energetically advantageous to have a r-dependent spiral in $\mathbf{n}$.

A convenient analysis of the properties of $S_\mathbf{n}$ is provided by a direct generalization of the $1/N$ expansion of Ref. [11]. The results quoted in Fig. 1 and its caption were obtained from the $N = \infty$ saddle point equation, and (apart from certain preexponential factors) all functional forms are exact. The saddle point implements the constraint $\mathbf{n}^2 = 1$ and takes the form

$$3T \sum_{\omega_n} \int \frac{d^2 k}{4\pi^2} \chi_\mathbf{n}(k, \omega_n) = 1, \quad (4)$$

where k is a wave vector, ω_n is a Matsubara frequency, and $\chi_\mathbf{n}$ is the dynamic staggered spin susceptibility with

$$\chi_\mathbf{n}(k, \omega_n) = [m^2 + \chi_\perp \omega_n^2 + \rho(k_x^2 + k_y^2) + \zeta_1(k_x^4 + k_y^4) + 2\zeta_2 k_x^2 k_y^2]^{-1}. \quad (5)$$

The parameter m is determined by solving Eq. (4).

In the classical limit, $S \to \infty$, we need only retain the $\omega_n = 0$ term in Eq. (4) [12]. A solution for m exists for all $T > 0$, and leads to the crossovers in the spin correlation length ξ_{spin} shown in Fig. 1. The value of ξ_{spin}, and the pitch of the spiral order $\sim\sqrt{-\rho}$, as $T \to 0$ are obtained from the spatial Fourier transform of $\chi_\mathbf{n}(k, 0)$.

To investigate the Ising nematic order, we need to study correlations of the order parameter $\sigma(r, \tau)$ which we define by a gradient expansion of Eq. (2)

$$\sigma = \mathbf{n} \cdot \partial_x \partial_y \mathbf{n} - \partial_x \mathbf{n} \cdot \partial_y \mathbf{n}. \quad (6)$$

The Ising susceptibility, χ_σ, is then $\chi_\sigma = \int_0^{1/T} d\tau \int d^2 r \langle\sigma(r, \tau)\sigma(0, 0)\rangle$.

In the classical limit, $S \to \infty$, important exact properties of χ_σ follow from the ultraviolet finiteness of the two-dimensional field theory with Boltzmann weight $\exp[-(1/T) \int d^2 r \mathcal{L}_\mathbf{n}]$ and $\mathbf{n}$ independent of τ. Under a length rescaling analysis of this theory in which the ζ_i and λ_i are fixed, we see that both T and ρ scale as inverse length squared. These scaling dimensions establish that in the classical limit

$$\xi_{\rm spin} = \sqrt{\zeta_1/T}\Phi_1(\rho/T); \qquad \chi_\sigma = \zeta_1^{-1}\Phi_2(\rho/T), \quad (7)$$

where Φ_i is the cutoff independent scaling function which depends only on ratios of the ζ_i and λ_i. The Ising phase transition is associated with a divergence of Φ_2 at some negative argument of order unity, and Eq. (7) then implies the $T_c \sim -\rho$ dependence shown in Fig. 1 and verified numerically in Fig. 5. We can also compute χ_σ (including the quantum $\omega_n \neq 0$ modes) in the large N limit:

$$\chi_\sigma = 24T\sum_{\omega_n}\int\frac{d^2k}{4\pi^2}k_x^2k_y^2\chi_{\mathbf{n}}^2(k, \omega_n). \quad (8)$$

Using the results in Eqs. (5) and (8) predicts an exponential divergence in $1/T$ as $T \rightarrow 0$ for $\rho < 0$. This is, of course, an artifact of the large N limit, as our Monte Carlo studies clearly show that χ_σ diverges with a power-law Ising exponent at a $T_c > 0$.

We turn now to a discussion of the quantum physics at finite S. A key feature again emerges from an analysis of Eqs. (4) and (5) while retaining the full frequency summation: the soft spin spectrum ($\omega \sim k^2$) at the Lifshitz point implies that there cannot be long-range magnetic order over a finite regime of parameters for all finite S [13]. After evaluating the frequency integral at $T = 0$, a solution with m real exists for a range of values of $|J_3 - \frac{1}{4}J_1|$ smaller than $\sim e^{-\tilde{c}S}$, implying there is a spin gap in this regime. We can reasonably expect that the Ising nematic order survives into at least a portion this spin gap phase, as it does at $T > 0$.

A more careful analysis of the spin gap phase requires consideration of Berry phases [2,14], which are absent in $\mathcal{L}_{\mathbf{n}}$. Assuming second order quantum critical points, with increasing J_3, we first expect a spin gap state with valence bond solid (VBS) order and confined $S = 1/2$ spinon excitations after leaving the collinear Néel state. Conversely, decreasing J_3 from the spiral spin ordered phase, we expect a Z_2 spin liquid with Ising nematic order and deconfined bosonic spinons [2,5]. So quite remarkably, we expect the following sequence of four phases to appear *for all half-odd-integer spin S* with increasing J_3: Néel long-range-order (LRO), VBS, Z_2 spin-liquid, spiral LRO. The two intermediate phases have a spin gap, and they appear in a window which is exponentially small in S for large S; the latter two phases have Ising nematic order. Theories for the three quantum critical points between these four phases appear in Refs. [14,15]. We cannot rule out the possibility that the some of these critical points and intermediate phases are preempted by a first order transition.

It is interesting to note that other Z_2 spin liquids with fermionic $S = 1/2$ spinons have been proposed [16], in which the ground state does *not* have Ising nematic order. Our present results naturally suggest a spin gap state with Ising nematic order, and mean field theories for such states have only been obtained with bosonic spinons [2]. Further studies of Ising nematic order in quantum spin models will therefore be valuable in resolving the nature of the spin-liquid state.

We acknowledge useful discussions with F. Becca, D. Scalapino, S. Sorella, and S. White. This work was supported by the NCSA under Grant No. DMR020027, the NSF under Grants No. DMR-0098226 (S. S.) and No. DMR-0210790, No. PHY-9907949 at the Kavli Institute for Theoretical Physics, and by the John Simon Guggenheim Memorial Foundation (S. S.). Kind hospitality provided by INFM-Democritos and SISSA is gratefully acknowledged (L. C.).

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