Ising Transition Driven by Frustration in a 2D Classical Model with Continuous Symmetry

Cédric Weber,[1,2] Luca Capriotti,[3] Grégoire Misguich,[4] Federico Becca,[5] Maged Elhajal,[1] and Frédéric Mila[1]

[1]*Institut de Physique Théorique, Université de Lausanne, CH-1015 Lausanne, Switzerland*
[2]*Institut Romand de Recherche numérique sur les Matériaux, Ecole Polytechnique Fédérale de Lausanne,
CH-1015 Lausanne, Switzerland*
[3]*Kavli Institute for Theoretical Physics, University of California, Santa Barbara California 93106-4030, USA*
[4]*Service de Physique Théorique, CEA Saclay, 91191 Gif-sur-Yvette Cedex, France*
[5]*INFM-Democritos, National Simulation Centre, and SISSA I-34014 Trieste, Italy*

We study the thermal properties of the classical antiferromagnetic Heisenberg model with both nearest (J_1) and next-nearest (J_2) exchange couplings on the square lattice by extensive Monte Carlo simulations. We show that, for $J_2/J_1 > 1/2$, thermal fluctuations give rise to an effective Z_2 symmetry leading to a *finite-temperature* phase transition. We provide strong numerical evidence that this transition is in the 2D Ising universality class, and that $T_c \to 0$ with an infinite slope when $J_2/J_1 \to 1/2$.

Since the milestone papers by Hohenberg and by Mermin and Wagner [1] it is known that in two-dimensional systems a continuous symmetry cannot be broken at any finite temperature, and only systems with a discrete symmetry can show a finite-temperature phase transition. In this regard, a proposal by Chandra, Coleman, and Larkin (CCL) [2] opened a new route to finite-temperature phase transitions in systems with a continuous spin-rotational invariance: CCL argued that the presence of frustrating interactions can induce nontrivial *discrete* degrees of freedom, that may undergo a phase transition at low temperatures. In particular, in Ref. [2], the authors considered the antiferromagnetic Heisenberg model with both nearest (J_1) and next-nearest neighbor (J_2) couplings:

$$\hat{\mathcal{H}} = J_1 \sum_{nn} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + J_2 \sum_{nnn} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j, \tag{1}$$

where $\hat{\mathbf{S}}_i$ are spin S operators on a periodic square lattice with $N = L \times L$ sites. For $J_2/J_1 < 1/2$, the classical ground state has antiferromagnetic Néel order with pitch vector $\mathbf{Q} = (\pi, \pi)$, while for $J_2/J_1 > 1/2$, the classical ground state consists of two independent sublattices with antiferromagnetic order [3]. The ground-state energy does not depend on the relative orientations between the magnetizations of the two sublattices, and the ground state has an O(3) × O(3) symmetry. Following Henley's analysis of the *XY* case [4], CCL showed that both quantum and thermal fluctuations are expected to lift this degeneracy by an *order by disorder* mechanism [5] and to select two collinear states which are the helical states with pitch vectors $\mathbf{Q} = (0, \pi)$ and $(\pi, 0)$, respectively, reducing the symmetry to O(3) × Z_2. CCL further argued that the Z_2 symmetry should give rise to an Ising phase transition at finite temperature and provided an estimate of the transition temperature.

The interest in this model raised recently with the discovery of two vanadates which can be considered as prototypes of the $J_1 - J_2$ model in the collinear region: Li_2VOSiO_4 and Li_2VOGeO_4 [6,7]. Indeed, although the value of J_2/J_1 is not exactly known [7–9], all estimates indicate that $J_2 \gtrsim J_1$. In particular, NMR and muon spin rotation magnetization in Li_2VOSiO_4 provide clear evidence for the presence of a phase transition to a collinear order at $T_c \sim 2.8$ K. While several additional ingredients, such as interlayer coupling and lattice distortion [10], are probably involved in the transition, the basic explanation relies on the presence of the Ising transition predicted by CCL.

However, CCL's predictions have been challenged by a number of numerical studies. Using Monte Carlo simulations, Loison and Simon [11] reported the presence of two phase transitions for the *XY* version of the classical model for $J_2/J_1 > 0.5$: A Kosterliz-Thouless transition, as expected for *XY* models, followed by a transition which is continuous but does not seem to be in the Ising universality class since their estimates of the critical exponents depend on the ratio J_2/J_1. More recently, the $S = 1/2$ Heisenberg case has been investigated by Singh and co-workers [12] using a combination of series expansion methods and linear spin-wave theory. They show that if there is a phase transition it can occur only at temperatures much lower than that predicted by CCL for $S = 1/2$, and they argue that T_c is actually equal to zero.

In this Letter, we show, on the basis of extensive Monte Carlo simulations, that the classical limit of the model of Eq. (1), where spins are classical vectors of length 1, indeed undergoes a continuous phase transition at a finite temperature, that the critical exponents agree with the Ising universality class, and that, modulo minor adjustments of CCL's estimate, T_c is in good *quantitative* agreement with CCL's prediction in the range $J_1 < J_2$ where their approximation is expected to be valid. However,

contrary to CCL's prediction, we show that T_c goes continuously to zero when $J_2/J_1 \rightarrow 1/2$, and we argue that this is due to a competition between Néel and collinear order at finite temperature in this parameter range.

Starting from the original spin variables $\hat{\mathbf{S}}_i$, we construct the Ising-like variable of the dual lattice:

$$\sigma_\alpha = \frac{(\hat{\mathbf{S}}_i - \hat{\mathbf{S}}_k) \cdot (\hat{\mathbf{S}}_j - \hat{\mathbf{S}}_l)}{|(\hat{\mathbf{S}}_i - \hat{\mathbf{S}}_k) \cdot (\hat{\mathbf{S}}_j - \hat{\mathbf{S}}_l)|}, \tag{2}$$

where (i, j, k, l) are the corners with diagonal (i, k) and (j, l) of the plaquette centered at the site α of the dual lattice. The two collinear states with $\mathbf{Q} = (\pi, 0)$ and $\mathbf{Q} = (0, \pi)$ have $\sigma_\alpha = \pm 1$. It is important to stress that the normalization term does not affect the critical properties of the model. It is only introduced to have a normalized variable. The Ising-like order parameter is defined as $M_\sigma = (1/N)\sum_\alpha \sigma_\alpha$.

We have performed classical Monte Carlo simulations using both local and global algorithms as well as more recent broad histogram methods [13] (details will be given elsewhere [14]) to calculate the temperature and size dependence of several quantities including the Binder cumulant, the susceptibility, and the correlation length associated to M_σ, as well as the specific heat, for sizes up to 200×200 and for several values of J_2/J_1 between $1/2$ and 2. For reasons discussed below, the critical behavior is easier to detect for small values of J_2/J_1, and we first concentrate on $J_2/J_1 = 0.55$.

As a first hint of a phase transition, we report the temperature dependence of the susceptibility defined by $\chi = (N/T)(\langle M_\sigma^2 \rangle - \langle |M_\sigma| \rangle^2)$ for different sizes [see Fig. 1(a)]. If there is a phase transition, this susceptibility is expected to diverge at T_c in the thermodynamic limit, and, indeed, the development of a peak around $T/J_1 = 0.2$ upon increasing the system size is clearly visible. To get a precise estimate of T_c, we have calculated Binder's fourth cumulant of the order parameter defined by $U_4(T) = 1 - \langle M_\sigma^4 \rangle/3\langle M_\sigma^2 \rangle^2$. This cumulant should go to $2/3$ below T_c and to zero above T_c when the size increases, and the finite-size estimates are expected to cross around T_c. Binder cumulants for different sizes are reported in Fig. 1(b), and they indeed cross around $T/J_1 \simeq 0.197$. In Fig. 1(c), we report $U_4(T)$ as a function of $1/L$ for several temperatures around 0.197. Excluding temperatures for which U_4 clearly increases or decreases with $1/L$ leads to the remarkably precise estimate $T_c/J_1 = 0.1970(2)$.

To identify the universality class of the phase transition, we have looked at the finite-size scaling of several quantities. The critical exponents ν and γ can be extracted from the dependence of the peak position of the susceptibility $T_c(L) = T_c + aL^{-1/\nu}$ and from its value $\chi(L, T_c) \sim L^{\gamma/\nu}$ as a function of L. Using the value of T_c deduced from Binder's cumulant, the fits lead to $\nu = 1.0(1)$ and $\gamma/\nu = 1.76(2)$. A more precise estimate of the exponent ν can be obtained from the temperature depen-
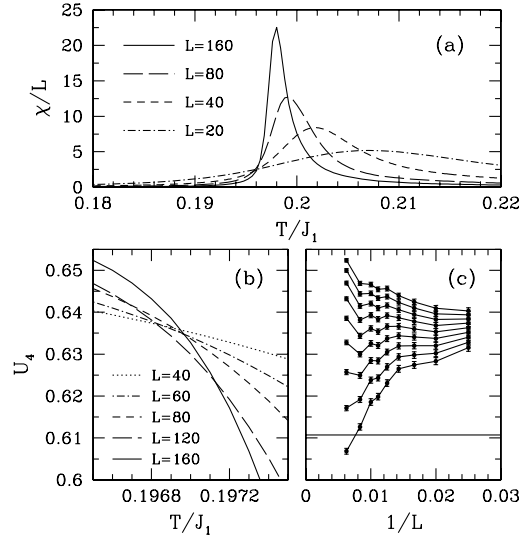

FIG. 1.   (a) Finite-size susceptibility $\chi(L)/L$ and (b) Binder cumulant U_4 as a function of the temperature for different sizes; (c) Binder cumulant U_4 as a function of $1/L$ for different temperatures $(0.1965, 0.1966, \ldots, 0.1973,$ from top to bottom). The lines are guides to the eye and the horizontal line marks the value for U_4 at the critical point for the Ising universality class [15]. All data are for $J_2/J_1 = 0.55$.

dence of the second-moment correlation length ξ [16], extracted from the Fourier components of the correlation functions of the Ising-like variable (2). By considering only values such that $\xi \lesssim L/6$, where the finite-size effects are found to be negligible, it is possible to have a very accurate value of the critical exponent from the fact that, for $T \gtrsim T_c$, $\xi^{-1} = A(T - T_c)^\nu$. In Fig. 2, we report the behavior of the correlation length ξ as a function of the temperature. By performing a three-parameter fit (for A, T_c, and ν) we obtain $T_c/J_1 = 0.1965(5)$ and $\nu = 1.00(3)$. This value of T_c is compatible with the estimation given by Binder's cumulant.

These exponents agree with those of the Ising universality class in 2D ($\nu = 1$ and $\gamma = 7/4$). A cross-check for this universality class comes from the measure of the critical exponent α, related to the divergence of the
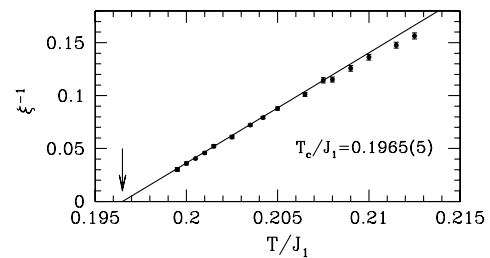

FIG. 2.   Inverse of the correlation length ξ^{-1} as a function of the temperature for different sizes of the lattice and $J_2/J_1 = 0.55$. The critical exponent ν and T_c can be extracted from the behavior of ξ^{-1} as a function of the temperature. The arrow marks the resulting T_c.

specific heat per site, $C_{max}(L) \sim L^{\alpha/\nu}$. Indeed, the value of α is the fingerprint for the 2D Ising universality class, for which we have $\alpha = 0$ and a logarithmic divergence of the specific heat as a function of L. In Fig. 3, we show the results for the specific heat per site. We have obtained a very accurate fit of the maximum of the specific heat per site as a function of L with the known expression for the leading finite-size corrections of the 2D Ising model [17], $C_{max}(L) = a_0 + a_1 \ln(L) + a_2/L$, with a_0, a_1, and a_2 fitting parameters, while a power law is clearly inadequate. These results are consistent with $\alpha = 0$.

Finally, if the phase transition is indeed Ising, Binder's cumulant at T_c should reach the universal value $U_4(T_c) \sim 0.6107$ in the thermodynamic limit [15]. The nonmonotonic behavior of Binder's cumulant with the size prevents a precise extrapolation, but this value is not incompatible with our numerical data [see Fig. 1(c)].

Altogether, these results show unambiguously that a phase transition occurs for $J_2/J_1 = 0.55$ at $T_c = 0.1970(2)$ and give strong evidence in favor of 2D Ising universality class, in agreement with CCL's prediction. The same analysis can be repeated for different values of the frustrating ratio J_2/J_1, and the complete phase diagram is shown in Fig. 4, where we report T_c as a function of J_2/J_1. While the critical behavior is everywhere consistent with Ising, it turns out that finite-size effects become more and more severe upon increasing J_2/J_1, preventing a meaningful determination of T_c with available cluster sizes beyond $J_2/J_1 \simeq 2$. Indeed, for large ratios J_2/J_1, physical quantities such as the susceptibility and the specific heat only exhibit broad peaks while the mean value of the order parameter goes very smoothly to zero, a behavior typical of strong finite-size effects. This
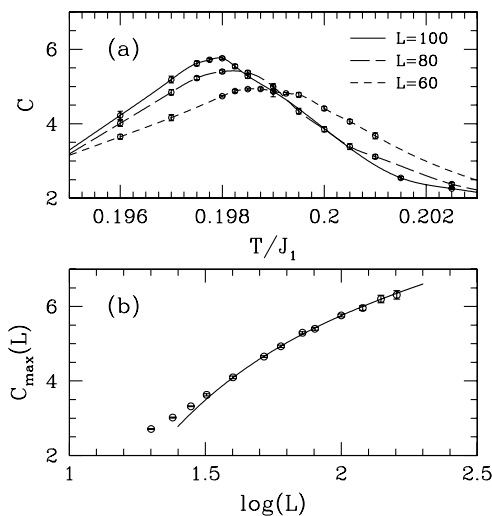
we believe can be traced back to the width of the domain walls between domains with $\mathbf{Q} = (\pi, 0)$ and $\mathbf{Q} = (0, \pi)$, which we have estimated by studying systems with fixed boundary conditions. Details will be presented elsewhere [14], but the width increases very fast with J_2/J_1, starting around ten lattice spacings for $J_2/J_1 \gtrsim 1/2$, and already reaching values of the order of 40 lattice spacings for $J_2/J_1 \simeq 1.5$. Since cluster sizes should be significantly larger than the width of the domain walls to observe the critical behavior, we are not able to go beyond $J_2/J_1 \simeq 2$.

Let us now discuss the dependence of T_c upon J_2/J_1 (see Fig. 4). Two regimes can clearly be identified: (i) A large J_2 regime ($J_2/J_1 > 1$) where T_c/J_1 scales more or less linearly with J_2/J_1; (ii) a smaller J_2 regime defined by $(J_2/J_1 - 1/2) \ll 1$ where T_c vanishes with an infinite slope as $J_2/J_1 \to 1/2$. For $J_2/J_1 \sim 1/2$, this disagrees with CCL's prediction that T_c/J_2 reaches its maximum when $J_2/J_1 \to 1/2$. However, since their approach is based on an expansion in J_1/J_2, this is not a final blow, and CCL's predictions should be tested in the large J_2 regime, where the approximations are better controlled. CCL's central criterion for estimating T_c is the equation $T_c \simeq E(T_c)[\xi_N(T_c)/a]^2$, where $E(T_c)$ is the energy barrier to go from one domain to the other through the intermediate canted state where sublattice staggered magnetizations are perpendicular, and $\xi_N(T)$ is the Néel correlation length of each sublattice. To get a quantitative estimate of T_c based on this approach, we have solved this equation using the exactly known temperature dependence of $\xi_N(T)$ for the classical antiferromagnet [18] and a corrected expression for $E(T)$ [19]. This leads to $T_c = 0.768 J_2/[1 + 0.135 \ln(J_2/J_1)]$. The best way to check this approach would be to detect the logarithmic correction, but unfortunately this would require one to go to temperatures much larger than what we can reach, and in the temperature range available to our simulations, CCL's prediction reduces to $T_c \simeq 0.77 J_2$. This prediction is in good agreement with our results: In the large J_2 regime, T_c indeed scales linearly with J_2, within error bars our high J_2 data extrapolate to 0 at $J_2 = 0$, and the slope is equal to 0.55. Note that the slight difference in slopes is not significant since including a constant factor
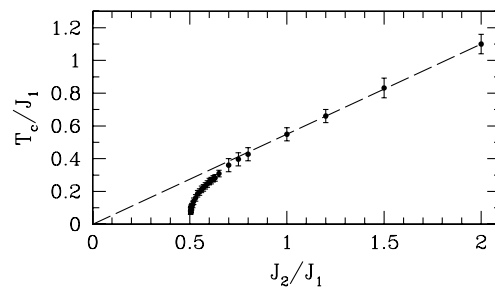

FIG. 3. (a) Specific heat per site C as a function of the temperature for different sizes of the lattice and $J_2/J_1 = 0.55$. The lines are guides to the eye. (b) Maximum of the specific heat per site $C_{max}(L)$ as a function of L. The line is a three-parameter fit (see text).

FIG. 4. Monte Carlo results for the critical temperature T_c as a function of the frustrating ratio J_2/J_1. The line is an extrapolation of the large J_2 data down to $J_2 = 0$ (see text).

in front of $E(T_c)$ in the self-consistent equation for T_c would change the slope. So altogether we believe that the present results support CCL's prediction when J_2/J_1 is not too close to $1/2$. In that respect, we note that a similar estimate of T_c can be performed on the basis of CCL's criterion for $S = 1/2$ using recent estimates of $\xi_N(T)$ for the $S = 1/2$ Heisenberg antiferromagnet on the square lattice [20], which leads to $T_c = 0.496 J_2/[1 + 0.78 \ln(J_2/J_1)]$. These estimates are considerably lower than those used in Ref. [12], and with these new estimates we believe that a phase transition cannot be excluded on the basis of the numerical results of Ref. [12]. In fact, although the precise values of J_1 and J_2 in LiVOSiO$_4$ are still a matter of discussion [7–9], we note that the order of magnitude of these new estimates is consistent with the experimental results of Ref. [7].

Finally, let us discuss the behavior of T_c close to $J_2/J_1 = 1/2$. The disagreement with CCL suggests that one ingredient is missing in that range. In fact, we found that the fluctuations have a clear Néel character when $J_2/J_1 \sim 1/2$, implying that, although the collinear phase is energetically favored at zero temperature when $J_2/J_1 > 1/2$, thermal fluctuations favor the Néel state, as quantum fluctuations indeed do [21]. We then expect a crossover to take place between a high-temperature Néel phase and a low-temperature collinear phase. To estimate the crossover temperature, we start from the low-temperature expansion of the free energy per site for classical spins which reads, when only quadratic modes are thermodynamically relevant,

$$ f = e_0 - T \ln T - \left(\frac{1}{N} \sum_q \ln \omega_q \right) T + a_2 T^2 + \dots, $$

where e_0 is the ground-state energy per site and ω_q are the frequencies of the quadratic modes. For the present purpose, the coefficients of this expansion should be determined in the limit $J_2/J_1 \to 1/2$ from below and above for the Néel and collinear states, respectively, and the crossover temperature T_0 is determined by $f_{\text{Neel}} = f_{\text{coll}}$. It turns out that, in the limit $J_2/J_1 \to 1/2$, $\sum_q \ln \omega_q$ has the same value for the Néel and collinear phases, so that the linear term drops from this equation, leading to $T_0 \propto (J_2/J_1 - 1/2)^{1/2}$ since $e_0^{\text{Neel}} = -2J_1 + 2J_2$ and $e_0^{\text{coll}} = -2J_2$ [22]. Now, the Ising transition can only occur below T_0 since the system should already have short-range collinear fluctuations. Then, when the intrinsic temperature scale of the Ising transition as determined by CCL is larger than T_0, we expect the transition temperature to be of the order of T_0. This argument thus predicts that, as $J_2/J_1 \to 1/2$, T_c should vanish with an infinite slope and with an exponent equal to $1/2$ [22]. This is in qualitative agreement with our numerical data, which clearly indicate that the slope is infinite, and are consistent with an exponent in the range 0.3–0.5. Large

scale numerical simulations are in progress to try to refine this estimate.

In conclusion, we have established the presence of a finite-temperature phase transition in the classical $J_1 - J_2$ Heisenberg model on a square lattice when $J_2/J_1 > 1/2$, we have provided strong arguments in favor of Ising universality class, and we have determined T_c as a function of J_2/J_1 with high accuracy, showing, in particular, that it vanishes with an infinite slope when $J_2/J_1 \to 1/2$. These results, together with revised estimates of T_c in the $S = 1/2$ case following CCL, are expected to set the stage for further theoretical and experimental investigations.

[1] P. C. Hohenberg, Phys. Rev. **158**, 383 (1967); N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).
[2] P. Chandra *et al.*, Phys. Rev. Lett. **64**, 88 (1989).
[3] P. Chandra and B. Doucot, Phys. Rev. B **38**, 9335 (1988).
[4] C. L. Henley, Phys. Rev. Lett. **62**, 2056 (1989).
[5] J. Villain *et al.*, J. Phys. (Paris) **41**, 1263 (1980).
[6] P. Millet and C. Satto, Mater. Res. Bull. **33**, 1339 (1998).
[7] R. Melzi *et al.*, Phys. Rev. Lett. **85**, 1318 (2000); Phys. Rev. B **64**, 024409 (2001).
[8] H. Rosner *et al.*, Phys. Rev. Lett. **88**, 186405 (2002).
[9] G. Misguich *et al.*, Phys. Rev. B **68**, 113409 (2003).
[10] F. Becca and F. Mila, Phys. Rev. Lett. **89**, 037204 (2002).
[11] D. Loison and P. Simon, Phys. Rev. B **61**, 6114 (2000).
[12] R. R. P. Singh *et al.*, Phys. Rev. Lett. **91**, 017201 (2003).
[13] P. M. C. de Olivera and T. J. P. Penna, Braz. J. Phys. **26**, 677 (1996); P. M. C. de Olivera, Braz. J. Phys. **30**, 195 (2000).
[14] C. Weber *et al.* (unpublished).
[15] G. Kamieniarz and H.W. J. Blöte, J. Phys. A **26**, 201 (1993).
[16] P. Olsson, Phys. Rev. B **55**, 3585 (1997); L. Capriotti *et al.*, Phys. Rev. B **58**, 273 (1998); F. Cooper *et al.*, Nucl. Phys. **B210**, 210 (1982).
[17] A. Ferdinand and M. E. Fisher, Phys. Rev. **185**, 832 (1969).
[18] See S. Caracciolo and A. Pelissetto, Nucl. Phys. **B420**, 141 (1994), and references therein.
[19] According to our derivation, the values given by CCL for their coefficients γ_Q and γ_T should be divided by 4.
[20] J.-K. Kim and M. Troyer, Phys. Rev. Lett. **80**, 2705 (1998).
[21] F. Mila *et al.*, Phys. Rev. B **43**, 7891 (1991).
[22] This argument is based on the assumption that $a_2^{\text{coll}} \neq a_2^{\text{Neel}}$. If they are equal, the exponent will be smaller.