Antiferromagnetic stacked triangular lattices with Heisenberg spins: Phase transition and effect of next-nearest-neighbor interaction

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We study by extensive histogram Monte Carlo simulations the phase transition in the antiferromagnetic stacked triangular lattices with *classical* Heisenberg spins. It is shown that in a range of the antiferromagnetic next-nearest-neighbor interaction J_2, the transition is clearly of first order. We also reconsider the controversial question concerning the nature of the phase transition when $J_2=0$: we show that the critical exponents obtained, in agreement with previous simulations, exclude the possibility of the O(4) class predicted by a nonlinear σ model in a $2+\varepsilon$ renormalization-group calculation. The phase diagram in the (J_2,T) space (T: temperature) is shown and discussed. For comparison, the phase diagram obtained by a Green-function method in the case of *quantum* Heisenberg spins is also shown.

I. INTRODUCTION

Recently, there has been a growing interest in the nature of the phase transition in the stacked triangular lattices with classical Heisenberg spins interacting with each other via nearest-neighbor (NN) antiferromagnetic interaction J_1.[1-4] This is one of the simplest frustrated systems where the frustration is due to the interaction geometry. Despite extensive studies done in the last few years, the nature of the transition in that system remains an open question at present: using Monte Carlo (MC) simulations and a renormalization-group analysis with $4-\varepsilon$ expansion, Kawamura[1,2] has conjectured a new class of universality in contradiction to Azaria, Delamotte, and Jolicoeur[3,4] who predicted, by a nonlinear σ model in a $2+\varepsilon$ calculation, that if the phase transition is not of first order, then it is of second order with either mean-field tricritical exponents ($\nu=0.5$, $\beta=0.25$, $\alpha=0.5$, $\gamma=1$) or the known O(4) class ($\nu=0.74$, $\beta=0.39$, $\alpha=-0.22$, $\gamma=1.47$). Moreover, another recent theoretical calculation based on the so-called local potential approximation[5] gives the ν exponent equal to 0.63 and a recent Monte Carlo (MC) work[6] supports the MC results of Kawamura.[1,2]

Generally speaking, the stacked triangular antiferromagnets (STAF) with classical Heisenberg spins belong to the family of helimagnetic structures. Theoretical investigations[7-9] on the helimagnetic transition include early works by Garel and Pfeuty,[7] and by Bailin, Love, and Moore.[8] An important number of experimental investigations has also been done to study the nature of the helimagnetic-paramagnetic transition.[10-15] Though most of the experimental data seem to give the critical exponents in agreement with those obtained by MC simulations,[1,2] some of them do not exclude the possibility of a weakly first-order transition,[14] and in the Holmium case, there is no such agreement.[15] Note that MC simulations of a helical structure on a body-centered-tetragonal lattice[16] show a continuous transition with critical exponent $\nu=0.57$.

The controversy between $2+\varepsilon$ and $4-\varepsilon$ calculations on the nature of the phase transition in the Heisenberg STAF with NN interaction has motivated the present work. Our purpose is double fold. First, we recalculate the value of the ν exponent obtained by various authors,[1,2,5,6,16] and secondly, we study the nature of the phase transition in the STAF taking into account the NN and next-nearest-neighbor (NNN) interactions, J_1 and J_2, respectively. As it turns out, we find in this work that the transition at the $J_2=0$ point is definitely of second order with critical exponents clearly different from those of O(4) class and mean-field tricritical point, in agreement with earlier MC results.[1,2,6,16] The scenarios predicted by Azaria, Delamotte, and Jolicoeur[3,4] are thus not verified. When antiferromagnetic NNN interaction is taken into account, we find the existence of a first-order transition at a finite temperature in the vicinity of the critical value $J_2=0.125J_1$, where collinear spin configurations coexist with the 120° structure in the classical ground state (GS).

In Sec. II, we briefly recall the model and the MC method. Results are shown and discussed in Sec. III where the phase diagram in the phase space (J_2/J_1, temperature) is displayed. In Sec. IV, we briefly show, for comparison, the phase diagram in the same space obtained for *quantum* Heisenberg spins by a Green-function method. Concluding remarks are given in Sec. V.

II. MODEL AND METHOD

We consider a system composed of the triangular lattices stacked along the z axis. The Hamiltonian is given by

$$H=\sum_{ij}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j \ , \qquad (1)$$

where $\mathbf{S}_i$ ($\mathbf{S}_j$) is a vector spin of unit length occupying the lattice site i (j) and the sum runs over NN and NNN in the xy planes (perpendicular to z, the stacking direction) and NN in the z direction. We call J_1 the antiferromag-

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netic NN interaction (>0) and J_2 the antiferromagnetic NNN interaction (>0) in the xy planes, and we take the NN interaction along the z direction J_3 equal to J_1, for simplicity. Hereafter, the energy and temperature will be measured in the unit of J_1.

For vector spins, the classical GS can be determined by minimizing the energy after a Fourier transform.[17] It is given as follows: for $0<J_2<0.125J_1$, the classical GS is the 120° structure, for $0.125J_1<J_2<J_1$ the classical GS degeneracy is continuous, the collinear (antiparallel) configurations are particular GS configurations in this range of parameters. For $J_2>J_1$ the GS is incommensurate with threefold degeneracy: spins on each line are parallel and two neighboring spin lines form an angle α given by $\cos\alpha=-(1+a)/2a$ where $a=J_2/J_1$ (see Ref. 17).

We have performed extensive histogram MC simulations. The method has been described elsewhere.[18,19] The sample sizes used in our simulations are $L\times L\times L$ with $L=12$, 18, 24, 30, and 36. Periodic boundary conditions have been used (in the incommensurate phase, $J_2>J_1$, care has been taken to choose only compatible angles). In each run, from 300 000 to 500 000 MC steps per spin have been discarded for equilibrating and 2 000 000–4 000 000 MC steps per spin have been used for averaging. During each run, we followed the intermediate results and stopped the simulation only when the results do not depend significantly on the time.

III. RESULTS

A. $J_2=0$

We show first the results for the case where there is only NN interaction ($J_2=0$). The temperature at which the simulations are made is $T_0=0.9576J_1$, which is the critical temperature estimated for the infinite system.[6]

Figure 1(a) shows the internal energy per spin E as function of the inverse of the number of MC steps per spin. As seen, only after 10^6 MC steps per spin that E is stabilized. Other quantities are also very well stabilized with little fluctuations around the respective stabilized mean values, except the specific heat with strong fluctuations around its stabilized mean value [about 2%, see Fig. 1(b)].

The transition is clearly of second order since the energy cumulant $C_u=1-\langle E^4\rangle/(3\langle E^2\rangle^2)$ tends to $\frac{2}{3}$ as L becomes large (not shown here). In order to calculate the exponent ν, we calculated the V_1 cumulant defined as

$$V_1=\langle ME\rangle/\langle M\rangle-\langle E\rangle\ , \tag{2}$$

where M and E are magnetization and energy per spin, and $\langle\ \cdots\ \rangle$ means thermal average. For a second-order transition, V_1 behaves as $V_1(L)L^{1/\nu}$. We show in Fig. 2 the log-log plot of V_1 versus L. The points lie remarkably on a straight line whose slope gives the value of $1/\nu$. We obtain

$$\nu=0.59+/-0.01\ , \tag{3}$$

where the error was estimated by taking into account the

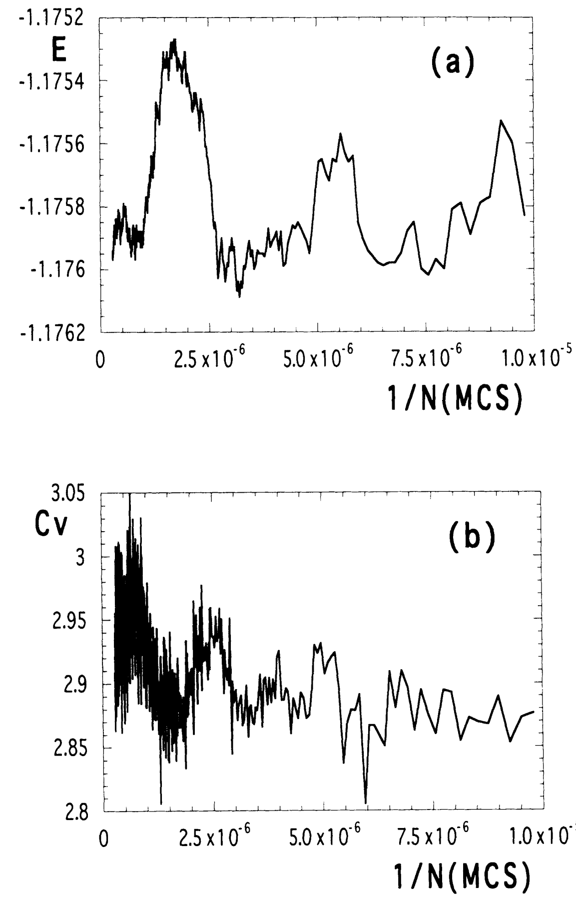

FIG. 1. Internal energy E (a) and specific heat C_v (b) as functions of inverse Monte Carlo steps per spin N (MCS), at the infinite-size transition temperature, for $L=24$.

different fits between different couples of sizes. This value is in agreement with that recently obtained by Kawamura,[20] and by the Ref. 6 which is $0.585+/-0.009$.

At this stage, it is worth to emphasize that the above value does not correspond to any known universality class as noted earlier by Kawamura.[1,2] If one rejects the conjecture of Kawamura for a new universality class, the reason is certainly not from the precision on the value of ν, but should come from another deeper argument.

The exponents β and γ are obtained from the log-log

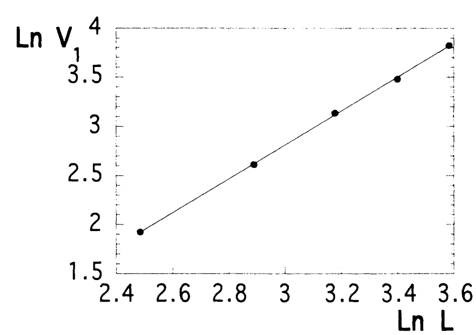

FIG. 2. The cumulant V_1 versus L in the ln-ln scale. The slope gives the value of $1/\nu$.

plots M and X (susceptibility) versus L shown in Figs. 3 and 4, respectively. One obtains from the slopes of the straight lines of values of $-\beta/\nu$ and γ/ν:

$$\beta/\nu = 0.50 +/- 0.02 ,$$

$$\gamma/\nu = 2.12 +/- 0.03 .$$

From these, one has $\beta = 0.28 +/- 0.015$ and $\gamma = 1.25 +/- 0.03$. These values are in agreement with those given by Kawamura ($\beta = 0.30 +/- 0.015$ and $\nu = 1.17 +/- 0.07$). The errors were estimated from the fits and errors on T_0.[6]

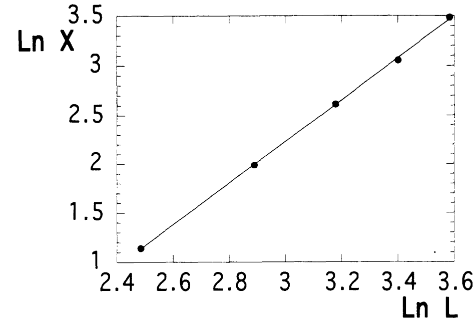

FIG. 4. Susceptibility X versus L in the ln-ln scale. The slope gives γ/ν.

B. $J_2 > 0$

Figure 5 displays the phase diagram in the phase space $(J_2/J_1, T)$. Let us describe briefly this diagram. For J_2 small and not close to $0.115J_1$, there is a second-order transition from the 120° structure to the paramagnetic state at a finite temperature. However, when J_2 larger than about $0.115J_1$, there are two transitions: the low-temperature one is between the 120° structure and a collinear spin configuration, and the high-temperature one between the latter and the paramagnetic phase. For $0.125J_1 < J_2 < J_1$, there is only on transition between the collinear state and the paramagnetic state. For $J_1 < J_2 < 1.12J_1$, one has two transitions. Finally, for J_2 larger than $1.12J_1$, there is a transition between the incommensurate state and the paramagnetic state.

We have checked by histogram method that the transitions denoted by black circles in Fig. 5 are of first order. If the system is at the transition temperature, then the energy distribution $P(E)$ is bimodal in a first-order transition. We show in Fig. 6, $P(E)$ and $J_2 = 0.12J_1$ at *almost* the transition temperature for each lattice size. As seen, the bimodal energy distribution is observed. The heights of the two peaks in each size are not equal because the temperature T at which the simulation was performed is not *exactly* the transition temperature. Of course, by varying T, one can obtain the exact transition temperature when the peak heights are equal. Furthermore, because of the finite-size effect, there is no real gap between the two peaks. In histogram method, when a bimodal distribution of $P(E)$ is observed, it is not easy to calculate from $P(E)$ physical quantities in the neighborhood of the simulation temperature. We therefore show in Fig. 7 the temperature dependence of the internal energy per spin E for $J_2 = 0.120J_1$ obtained by standard MC runs. The high-temperature transition has a first-order aspect with an almost "discontinuity" in E, while the low-temperature one shows only a slight, but sudden, change in the slope of E (not easy to see in the scale of Fig. 7). The low-temperature transition has a second-order feature though the symmetries of the two phases (120° and collinear) are not compatible. We think that the spin-wave excitations play an important role in making that transition a continuous one.

Other physical quantities confirm the first-order character of the high-temperature transition. The inset of Fig. 7 shows the finite-size scaling of the maximum of the specific heat, $C_{v\max}$, as a function of L, in an ln-ln scale. The slope for largest sizes studied in standard MC runs is 2.6 which shows that for larger sizes, $C_{v\max}$ is likely proportional the system volume, suggesting that the transition is of first order.[21]

Another interesting point in Fig. 5 is the fact that the collinear configurations are preferred at finite temperatures over the infinite number of GS in the range $0.125J_1 < J_2 < J_1$. This verifies the conjecture by Henley,[22] in analogy with what was called order by disorder predicted for Ising spin systems by Villain *et al.*[23] The collinear phase space is, in addition, extended over the noncollinear ones at finite T as seen in Fig. 5, where the

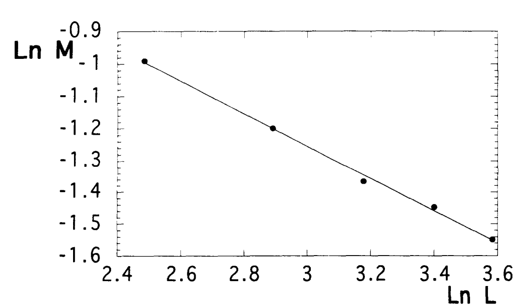

FIG. 3. Magnetization M versus L in the ln-ln scale. The slope gives $-\beta/\nu$.

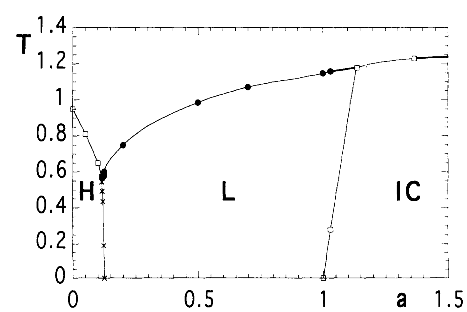

FIG. 5. Phase diagram in the $(a = J_2/J_1, T)$ space. First-order transitions are shown by black circles. Squares and crosses indicate the second-order transitions. L, H, and IC means collinear, helimagnetic (120° structure), and incommensurate phases, respectively. See text for comments.

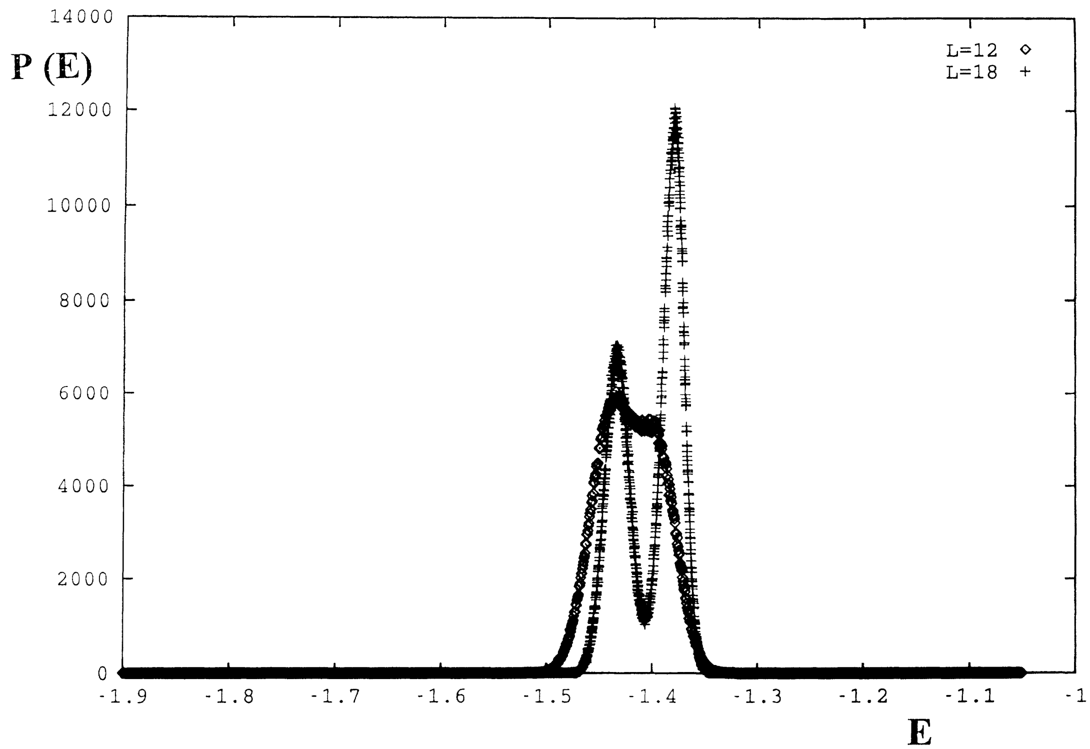

FIG. 6. Bimodal distribution of $P(E)$ is observed for $a=0.12$ with $L=12$ (diamonds) and $L=18$ (crosses), at a temperature very close to the transition temperature of each size. See text for comments.

curve issued from $J_2=0.125J_1$ is canted to the left, and that from $J_2=J_1$ to the right.

Let us give a simple explanation for the first-order transition between the colinear phase and the paramagnetic phase: in the collinear phase selected by entropy effect,[22] the degeneracy, apart from the infinite degeneracy due to global rotation, is three since there are three ways to choose the antiparallel spin pairs in a triangle. This threefold degeneracy is reminiscent of the three-state Potts model in three dimensions which is known to undergo a first-order transition at finite temperature. Note however that unlike the Potts model, there is a possible influence of the continuous degrees of freedom of vector spins (excitations of spin waves) that can change the nature of the phase transition. This may explain why, away from the critical values of J_2 ($0.125J_1$ and J_1), the first-order character of the transition is weakened, and in the range $J_2>J_1$, the transition is of second order.

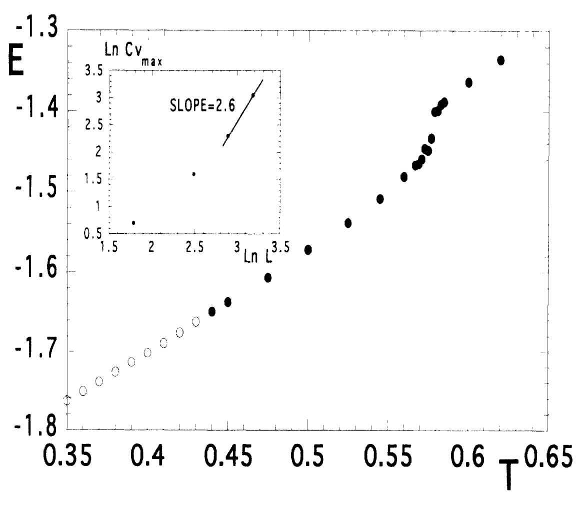

FIG. 7. Internal energy per spin E vs temperature T for $a=0.120$. The low-T phase is indicated by void circles and the high-T one up to paramagnetic phase by solid circles. The low-T transition is characterized by a slight (but sudden) change in the slope of U (not easily seen in this scale). The high-T transition is of first order. The inset shows $\ln(C_{v\max})$ versus $\ln(L)$.

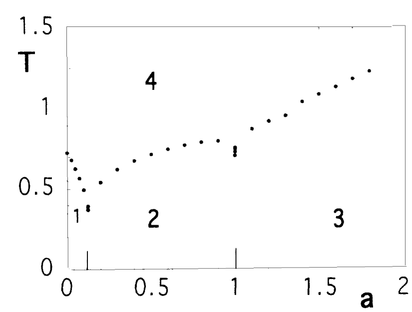

FIG. 8. Phase diagram in the space (a,T) in the case of quantum Heisenberg spins, obtained by the Green-function method. 1, 2, 3, and 4 mean helimagnetic (120° structure), collinear, incommensurate, and paramagnetic phases, respectively. See text for comments.

IV. PHASE DIAGRAM IN THE QUANTUM CASE

In order to compare qualitatively to the case of classical spins, we have studied the quantum spins on the STAF. We briefly show the results in the following.

We define the following double-time Green functions[24] $G_{ij}(t,t')$ and $F_{ij}(t,t')$ by

$$G_{ij}(t,t')=\langle\langle S_i^+(t);S_j^-(t')\rangle\rangle ,$$

$$F_{ij}(t,t')=\langle\langle S_i^-(t);S_j^-(t')\rangle\rangle ,$$

where $S_i^+(t)$ and $S_j^-(T)$ are the usual notations defined from the x and z spin components. For each spin, the local z axis is chosen along the direction of the spin. We write the equations of motion for these functions, and use the Tyablikov decoupling scheme. Furthermore, we introduce the two-dimensional and time Fourier transforms in the xz plane. The resulting equations of motion can be rewritten under a matrix, we diagonalize it and use spectral theorem[24] to calculate the local magnetization at arbitrary temperature T for spin $\frac{1}{2}$. We note that in the infinite degeneracy region $(0.125<a<1)$, the energy calculated with collinear configuration is always lower than that with noncollinear configuration even at $T=0$. The same is true at the degenerate point $a=0.125$: the GS energy calculated by using the collinear classical GS is lower than that using 120° structure. Thus, quantum and thermal fluctuations lift the degeneracy and select collinear configuration as conjectured.[22] The critical temperature T_c is determined when the local magnetization becomes zero. Details of the calculations have been shown elsewhere.[25] We only show in Fig. 8 the resulting phase diagram in the space $(a=J_2/J_1,T_c)$. As seen, there is no inclination of the critical lines issue from $a=0.125$ and $a=1$. Such behavior is thus a consequence of the spin quantum nature. Furthermore, due to approximations used in the decoupling of correlation functions to reduce higher-order Green functions, all lines in Fig. 8 are of second order. The nature of the transition, therefore, cannot be clarified by such a method.

V. CONCLUDING REMARKS

We have reconsidered the controversy of the problem of the nature of the phase transition in antiferromagnetic stacked triangular lattices with Heisenberg spins predicted by $2+\varepsilon$ and $4-\varepsilon$ calculations. The critical exponents have been carefully calculated. Our results are in agreement with those recently obtained,[20,6] but in contradiction with the scenarios predicted by nonlinear σ model with $2+\varepsilon$ expansion.[3,4] At this stage, we note that while there is a consensus about the value of the critical exponent ν, one can neither confirm nor reject the conjecture about the new universality class proposed by Kawamura. It is interesting to emphasize that for frustrated systems studied here, the $2+\varepsilon$ and $4-\varepsilon$ expansions do not agree with each other in three dimensions as they do for nonfrustrated systems. More investigation should be done to explain this contradiction. As the first step in this direction, we have checked the validity of the rigidity condition used in the nonlinear σ model.[3,4] The rigidity condition consists in neglecting the local-spin fluctuations in each triangle, i.e., the three spins on each triangle form rigidly a 120° structure even at finite T. We have found that when the rigidity is imposed on the system as it has been done in the nonlinear σ model,[3,4] the nature of the phase transition changes, namely the values of the critical exponents are significantly different.[26]

The effect of NNN interaction J_2 on the phase transition has also been studied here. A nontrivial phase diagram was obtained. The existence of a region of first-order transitions is seen. Furthermore, in the range of $0.125J_1<J_2<J_1$ where the GS is infinitely degenerate, it was found that only the collinear spin configurations are selected by thermal fluctuations. This is in agreement with a theoretical conjecture.[22] Finally, we hope that the results discussed in this paper may help to stimulate further theoretical effort on the problem of phase transition in frustrated spin systems.

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