

## Magnetic ordering in the three-dimensional frustrated Heisenberg model

J. R. Thomson, Hong Guo, D. H. Ryan, M. J. Zuckermann, and Martin Grant

*Physics Department, Rutherford Building, McGill University, 3600 rue University, Montréal, Québec, Canada H3A 2T8*

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Monte Carlo simulations of moderately frustrated three-dimensional Heisenberg spin systems show evidence for two distinct ordering events. The upper one, at  $T_c$ , marks the onset of long-range ferromagnetic order and exhibits fluctuations which scale as the system size. The lower event, at  $T_{xy}$ , marks the freezing of transverse degrees of freedom and does not affect the long-range order in the system. Critical levels of frustration for the onset of ferromagnetism have been determined on several lattices and a phase diagram is presented.

Experimental studies of frustrated magnetic systems show that the presence of competing ferromagnetic and antiferromagnetic interactions leads to a loss of collinearity in the low-temperature state and to the formation of a spin glass. However, if the concentration of antiferromagnetic interactions is small enough, it is possible to form a system which, despite the presence of mixed exchange, possesses many of the characteristics of a Heisenberg ferromagnet.<sup>1</sup> Such materials exhibit two distinct ordering events as the temperature is lowered. The upper one (at  $T_c$ ) is a ferromagnetic transition, marking the onset of long-range collinear (ferromagnetic) order. Below  $T_{xy}$ , the temperature of the second event, the system ceases to be collinear as the spins acquire a significant transverse component.<sup>2</sup> This event is characterized by a local ordering in the system, which we will call transverse spin freezing.

In this paper we present results of an extensive numerical study of this phenomena. We use the Monte Carlo method to investigate the simplest model which could be expected to show transverse spin freezing, namely, the moderately frustrated Heisenberg model in three dimensions. We find that the model is capable of reproducing all the experimentally observed features of transverse spin freezing. By analysis of the fluctuations occurring at the point of freezing, we conclude that it involves only a change in short-range order, and therefore is not a phase transition.

Previous work on low levels of frustration in Heisenberg spin systems has not led to a conclusive description of the magnetic ordering. The mean-field solution of the random-exchange Heisenberg model with a Gaussian distribution of exchange strengths predicts magnetic transitions, first from the paramagnetic state to a collinear ferromagnet, then at a lower temperature, to a mixed state where the transverse degrees of freedom order,<sup>3</sup> and finally to a state where replica symmetry is spontaneously broken at a still lower temperature. Recent local mean-field simulations of the two-dimensional  $XY$  model by Saslow and Parker<sup>4</sup> were aimed at obtaining a physical picture of reentrant magnetic transitions. Below the ferromagnetic transition, they reported two transitions, at  $T_K$  and a lower temperature  $T_r$ : As one decreased the

temperature below  $T_K$ , the frustrated bonds made themselves felt, and the system began to order in the transverse direction, while at  $T_r$ , their results were interpreted in terms of spin canting, mediated by the melting of frustrated spins. However, despite the appeal of a canting transition, on repeating these calculations, and doing Monte Carlo work in three dimensions, we have found that the system is easily trapped in metastable states, and that the apparent canting transition is an artifact of insufficient system equilibration.

The spin canting picture differs qualitatively from that of transverse spin freezing. On cooling through the onset of spin canting, the average magnetization,  $M$ , will decrease with respect to the average moment,  $S_{rms}$  (since the spins tilt away from their initially collinear configuration), while  $S_{rms}$  itself will only exhibit the normal increase due to reduced thermal fluctuations. The opposite occurs during transverse spin freezing: here transverse components, ordering with random directions in the  $x$ - $y$  plane, have no influence on  $M$ , but cause  $S_{rms}$  to increase. Experimentally, there is a marked change in the behavior of  $S_{rms}$  at  $T_{xy}$ , often displaying an apparent discontinuity in the slope, but no such changes are seen in  $M$ .<sup>1,5,6</sup>

We consider the classical Heisenberg spins on a simple-cubic lattice governed by the nearest-neighbor Hamiltonian:

$$\mathcal{H} = - \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \mathbf{H} \cdot \left[ \sum_j \mathbf{S}_j \right] \quad (1)$$

with exchange strength  $J_{ij}$  and unit spin. The second term includes the effect of an external magnetic field. The Heisenberg ferromagnetic (all  $J_{ij} = +J$ ) ordering temperature is  $T_H = 1.44J$ .<sup>7</sup> Exchange frustration is introduced by randomly replacing a fraction,  $f$ , of the interactions by antiferromagnetic bonds. Numerical simulations were performed by the Monte Carlo method. In order to investigate the behavior of the transverse spin components, we specified the  $z$  axis using two methods. The first method defines the  $z$  axis externally by applying a small magnetic field (in our case  $H_z = 0.03J$ ), as done in most experimental observations of transverse spin freez-

ing. In the second method, the  $z$  axis is defined by the instantaneous direction of the spontaneous magnetization. The data presented below were obtained using the second method. In order to study fluctuations we calculated the variation in  $S_{\text{rms}}$  from configuration to configuration,  $\langle S_{\text{rms}}^2 \rangle - \langle S_{\text{rms}} \rangle^2$ , where the brackets indicate an average over bond configurations.

The scaling results presented here are averages over at least 100 distinct bond configurations on various system sizes, with  $f=0.15$ , while the data used to determine points on the phase diagram are averages over at least 20 distinct bond configurations for each value of  $f$  on  $8^3$  lattices. In most cases our procedure was as follows: for each bond configuration the system was prepared well above  $T_c$ , then cooled in 50 temperature steps using a Monte Carlo algorithm with Glauber dynamics.<sup>8</sup> At each temperature, the system was allowed to equilibrate for 2000 Monte Carlo steps before averages were taken over 4000 Monte Carlo steps. The exception to this procedure was that used to determine the bond fraction above which the system is no longer ferromagnetic. In this case we prepared the system with  $f=0.5$  at a particular temperature and then progressively replaced antiferromagnetic bonds with ferromagnetic bonds. For these systems, we examined the susceptibility as a function of  $f$ .

We calculated two local time-averaged quantities. The first, a time average of the spin components at each site:

$$\mathbf{m}_i = \frac{1}{\tau} \sum_{\tau'=0}^{\tau} \mathbf{S}_i(\tau'), \quad (2)$$

where time  $\tau=4000$  Monte Carlo steps and  $i$  is the site index. The second was the average of the square of the transverse spin components at each site:

$$m_{\perp i}^2 = \frac{1}{\tau} \sum_{\tau'=0}^{\tau} [\mathbf{S}_i - (\mathbf{S}_i \cdot \hat{\mathbf{z}})\hat{\mathbf{z}}]^2. \quad (3)$$

These were then used to obtain the following bulk averages: the root-mean-square spin length  $S_{\text{rms}} = [N^{-1} \sum_i (\mathbf{m}_i \cdot \mathbf{m}_i)]^{1/2}$ ; the bulk magnetization  $M = N^{-1} |\sum_i \mathbf{m}_i|$ ; the mean-square transverse spin, which is a measure of the average component of the spins which lie perpendicular to the  $z$  axis independent of angular motion in the  $x$ - $y$  plane,  $Q_{\perp} = N^{-1} \sum_i m_{\perp i}^2$ ; and finally, the mean transverse spin length, which averages over rotational motion of the spins in the  $x$ - $y$  plane and measures the ordering of transverse spin components,  $Q_{xy} = N^{-1} \sum_i [\mathbf{m}_i - (\mathbf{m}_i \cdot \hat{\mathbf{z}})\hat{\mathbf{z}}]^2$ .

Figure 1 shows the temperature evolution of the macroscopic quantities defined above for a number of frustration levels. For  $f=0$ , all bonds positive, the system is a Heisenberg ferromagnet below  $T_H$ . Since  $S_{\text{rms}}$  is identical to  $M$  at all  $T < T_H$ , the system must be ordered collinearly.  $Q_{\perp}$  decreases smoothly from  $\frac{2}{3}$  above  $T_H$ , corresponding to the isotropic spin distribution of a paramagnet, to zero at  $T=0$ , as expected for a collinear ferromagnet.  $Q_{xy}=0$  at all temperatures, indicating that there are no ordered transverse components.

New features appear for  $f > 0$ . First, since  $Q_{\perp}$  does not reach zero for any  $f > 0$ , some degree of spin noncol-

linearity must persist even at  $T=0$ . More significantly, although  $Q_{xy}$  is initially zero below  $T_c$ , as in the ferromagnetic case, it becomes finite at a temperature  $T_{xy}$ , well below  $T_c$ , marking the onset of transverse spin freezing. On cooling from  $T_c$  to  $T_{xy}$ , the root-mean-square spin length  $S_{\text{rms}}$  is essentially equal<sup>9</sup> to the bulk magnetization  $M$ , but at  $T_{xy}$ ,  $S_{\text{rms}}$  begins to exceed  $M$  and remains larger down to  $T=0$ . We attribute the excess growth in  $S_{\text{rms}}$  to transverse spin freezing. Above  $T_{xy}$ , the configurations show that a particular spin possesses a transverse component, but it is in motion and the time average of that component vanishes. Thus,  $\mathbf{m}_i$  only has a  $z$  component for  $T_{xy} < T < T_c$ , making  $S_{\text{rms}}$  and  $M$  equivalent and  $Q_{xy}=0$ . Below  $T_{xy}$ , the transverse spin components freeze in random directions so the sum of their site average cancels, leaving  $M$  unaffected. The transverse components, however, do contribute to the sum so that  $S_{\text{rms}}$  increases. The persistence of large  $Q_{\perp}$  above  $T_{xy}$  and the existence of finite  $Q_{xy}$  only at temperatures less than  $T_{xy}$  reinforce the experimental and theoretical observations of the increase of  $S_{\text{rms}}$  over  $M$  as direct evidence of transverse spin freezing. These data are incompatible with a spin canting model of this behavior.

As  $f$  is increased,  $T_c$  and  $T_{xy}$  converge:  $T_c$  falls reflecting a decline in the average exchange strength, and

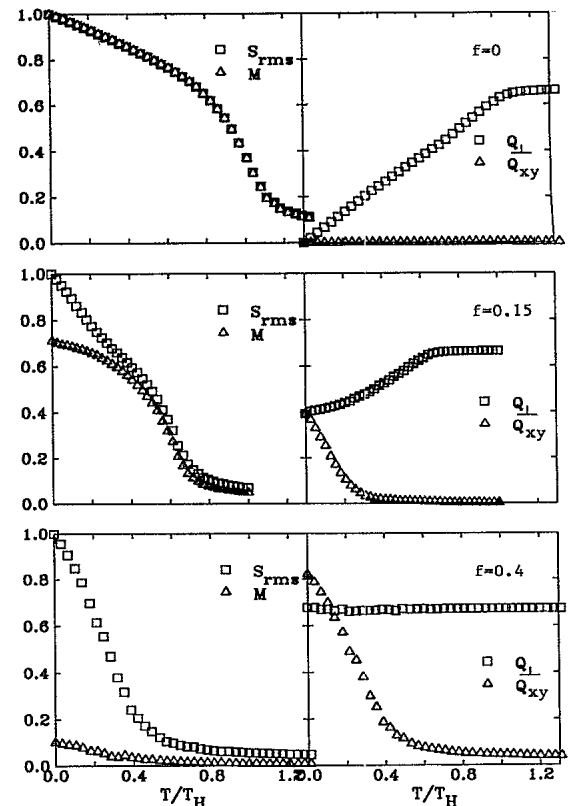


FIG. 1. Temperature dependence of longitudinal and transverse spin lengths for different levels of exchange frustration. The temperature scale is normalized to the Heisenberg ferromagnetic transition temperature,  $T_H$ .

$T_{xy}$  rises due to the increase in frustration. Finally, at  $f_c \approx 0.25$ ,  $T_c$  and  $T_{xy}$  merge and  $Q_{\perp}$  remains  $\frac{2}{3}$  for all temperatures. This indicates that the spin directions are isotropic, a spin-glass-like configuration. On three-dimensional lattices with higher coordination numbers (bcc  $q=8$  and fcc  $q=12$ ) further simulations have indicated that the ferromagnetic behavior persists to higher levels of frustration (bcc  $f \approx 0.30$ , fcc  $f \approx 0.32$ ). The ordering behavior is very similar to that deduced from experiments,<sup>1,2,10</sup> however, we caution that the low-temperature behavior of the fully frustrated three-dimensional Heisenberg model is still an unresolved issue.<sup>10-12</sup> In this work we address the ordering of systems in a regime of moderate frustration distinct from that of the spin-glass model.

The phase diagram given in Fig. 2 summarizes the frustration dependence of the ordering events. The system is paramagnetic at high temperatures. Notwithstanding the reservations noted above, for  $f > 0.25$  the system enters a spin-glass-like phase on cooling through  $T_{sg}$ . The vertical line at  $f_c = 0.25$  separating that phase from the ferromagnet was located by the peak of the fluctuations in  $M$ , as a function of frustration. For  $0 < f < 0.25$ , the system goes through a transition at  $T_c$  to a ferromagnetic phase. For a given  $f$ , this line marks the temperature where  $S_{rms}$  and  $M$  deviate from zero. Immediately below  $T_c$  the order is ferromagnetic with a significant transverse spin component which time averages to zero. The dotted line,  $T_{xy}$ , marks the temperature at which  $Q_{xy}$  ceases to be zero, which coincides with the temperature at which  $S_{rms}$  and  $M$  separate. The system exhibits ferromagnetic order on both sides of the line, but below  $T_{xy}$  the transverse spin components are frozen; above it they average over time to zero.

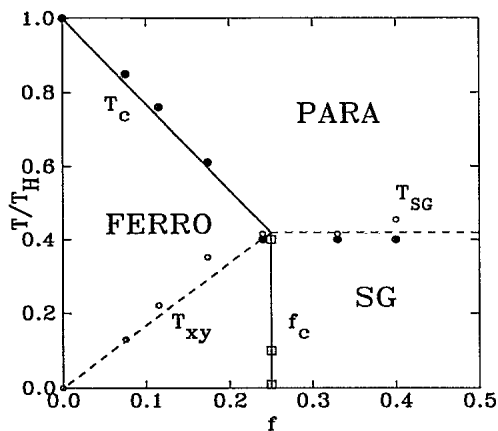


FIG. 2. The phase diagram for the model showing two magnetic transitions in the concentration region between  $f=0$  and the spin-glass-like phase at  $f > 0.25$ . At  $T_c$ , collinear order sets in, followed by the freezing of transverse spin components below  $T_{xy}$ .  $f_c$  marks the boundary between the ferromagnet and the spin-glass-like phase. The temperature scale is normalized to the Heisenberg ferromagnetic transition temperature,  $T_H$ . Open circles indicate the temperature at which  $Q_{xy}$  becomes finite, while the solid circles represent the temperature of the maximum of the susceptibility.

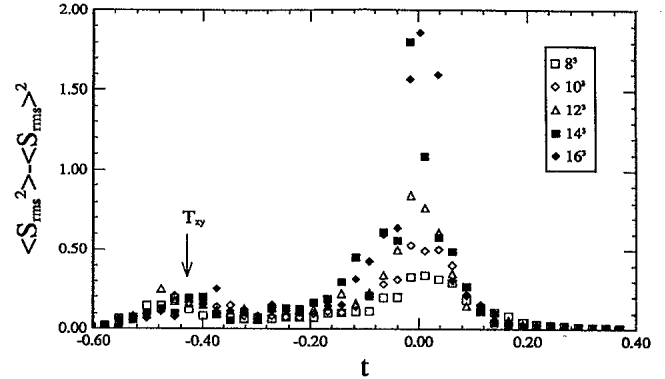


FIG. 3. The configuration to configuration fluctuations in  $S_{rms}$  as a function of  $t = (T - T_c)/T_H$ . Note that the peak at  $T_{xy}$  does not grow with system size, while that at  $T_c$  grows as the system size is increased.

The configuration to configuration fluctuations in  $S_{rms}$  shows a peak at both  $T_c$  and  $T_{xy}$ . We examined the effect of finite system size on the amplitude of these peaks. Figure 3 shows these fluctuations, averaged over at least 100 different bond configurations, as a function of temperature for system sizes with edge length  $L = 6, 8, 10, 12, 14$ , and  $16$  ( $\sim 25$  bond configurations for the  $L = 14$  and  $16$ ). It is clear that, while the peak at  $T_c$  grows as system size is increased, the peak at  $T_{xy}$  saturates and does not increase. We therefore interpret the results at  $T_{xy}$  as characteristic of local ordering events which do not induce fluctuations on the order of correlation length. In finite-size scaling analysis, where we expect the amplitude of critical point fluctuations to scale as  $L^{\gamma/\nu}$ ,<sup>13</sup> we obtained a value for  $\gamma/\nu = 1.91 \pm 0.01$  corresponding accurately to the accepted values of the critical exponents for the Heisenberg ferromagnet in three dimensions.<sup>7</sup> Furthermore, when we use the scaling form,  $\chi \propto |t|^{-\gamma} \bar{\chi}(Lt^\nu)$ , where  $t = (T - T_c)/T_H$ , with  $L$ 's varying by a factor of 2, a very good collapse of the curves is obtained with the three-dimensional Heisenberg ferromagnet exponents, as shown in Fig. 4, although there are some systematic deviations at high temperatures.

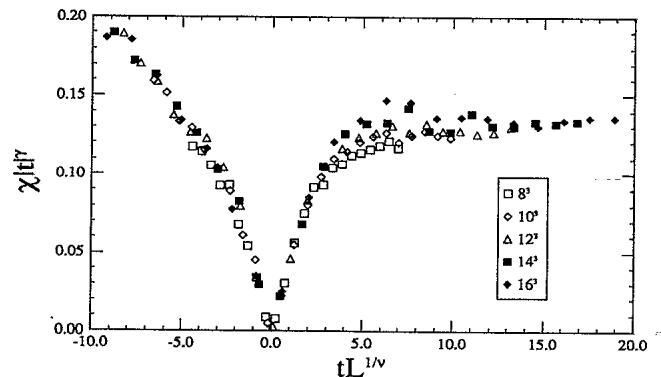


FIG. 4. The static susceptibility,  $\chi$ , rescaled by  $\chi \propto |t|^{-\gamma} \bar{\chi}(Lt^\nu)$ , where  $t = (T - T_c)/T_H$ . ( $T_c$  is taken at the position of the maximum of  $\chi$  for  $L = 12$ , and  $f = 0.15$ .) The values for the exponents were  $\gamma = 1.33$  and  $\nu = 0.692$  (Ref. 7).

This reinforces the interpretation that below the critical fraction of antiferromagnetic bonds ( $f=0.25$ ) the system is a Heisenberg ferromagnet.

To conclude, our Monte Carlo simulations are in good qualitative agreement with experimental observations. In particular, we found two magnetic ordering events for the low levels of frustration: ferromagnetic ordering at  $T_c$  followed by transverse spin freezing at  $T_{xy}$ . The critical behavior at  $T_c$  is consistent with a Heisenberg fer-

romagnetic transition in three dimensions, while we interpret the behavior at  $T_{xy}$  in terms of a change in short-range order rather than a phase transition.

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- <sup>1</sup>R. A. Brand, V. Manns, and W. Keune, *Heidelberg Colloquium on Spin Glasses*, Lecture Notes in Physics, Vol. 192 (Springer-Verlag, Berlin, 1983), p. 79.
- <sup>2</sup>D. H. Ryan, J. O. Ström-Olsen, R. Provencher, and M. Townsend, *J. Appl. Phys.* **64**, 5787 (1988).
- <sup>3</sup>M. Gabay and G. Toulouse, *Phys. Rev. Lett.* **47**, 201 (1981).
- <sup>4</sup>W. M. Saslow and G. Parker, *Phys. Rev. Lett.* **56**, 1074 (1986).
- <sup>5</sup>B. Huck and J. Hesse, *J. Magn. Magn. Mater.* **78**, 247 (1989).
- <sup>6</sup>Some related theoretical work includes the following: D. M. Cragg, D. Sherrington, and M. Gabay, *Phys. Rev. Lett.* **49**, 158 (1982); D. Elderfield and D. Sherrington, *J. Phys. C* **16**, 4865 (1983); D. Sherrington, in *Heidelberg Colloquium on Spin Glasses*, edited by J. L. Van Hemmen and I. Morgenstern (Springer, Berlin, 1983); M. W. Dunlop and D. Sherrington, *J. Phys. C* **18**, 1465 (1985); S. Jain and A. P. Young,

*ibid.* **19**, 3913 (1986); J. A. Olive, A. P. Young, and D. Sherrington, *Phys. Rev. B* **34**, 6341 (1986).

- <sup>7</sup>C. Domb and M. S. Green, *Phase Transitions and Critical Phenomena* (Academic, London, 1974), Vol. 3.
- <sup>8</sup>O. G. Mouritsen, *Computer Studies of Phase Transitions and Critical Phenomena* (Springer-Verlag, Heidelberg, 1984).
- <sup>9</sup>The difference between  $M$  and  $S_{rms}$  above  $T_{xy}$  is a result of sites that are completely antiferromagnetic.
- <sup>10</sup>J. Souletie, *Ann. Phys. (Paris)* **10**, 69 (1985).
- <sup>11</sup>K. Binder and A. P. Young, *Rev. Mod. Phys.* **58**, 801 (1986).
- <sup>12</sup>A. Maugher, J. Villain, Y. Zhou, C. Rigaux, N. Bontemps, and J. Férré, *Phys. Rev. B* **41**, 4587 (1990).
- <sup>13</sup>V. Privman, *Finite Size Scaling and Numerical Simulation of Statistical Systems* (World-Scientific, London, 1986).