

## Magnetic ordering in the three-dimensional site-disordered Heisenberg model

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Monte Carlo simulations have been carried out on a simple cubic ferromagnet with nearest-neighbor interactions. In order to model the effects of site frustration, a fraction  $f$  of the sites are occupied at random by moments that couple antiferromagnetically (AF) to their neighbors. When the concentration of AF sites is less than  $\sim \frac{1}{6}$ , the system has one magnetic transition from paramagnet to ferromagnet at a critical temperature  $T_c$ . For  $f > \frac{1}{6}$  the system exhibits a second distinct ordering event at a lower temperature  $T_{xy}$ , where the transverse spin components freeze out leading to an increase in total spin length. Below  $T_{xy}$  the system is in a mixed state, in that the  $z$  components of the spins are ferromagnetically ordered while the transverse components exhibit AF correlations. The approximate magnetic phase diagram for our model is consistent with experimental results on site-disordered systems such as  $\text{Eu}_{1-x}\text{Gd}_x\text{S}$  and  $\text{Fe}_{3-x}\text{Mn}_x\text{Si}$ .

### I. INTRODUCTION

Experimental studies<sup>1-4</sup> of partially frustrated magnetic systems have demonstrated the existence of transverse spin freezing at a temperature  $T_{xy}$  below the ferromagnetic transition. This phenomenon is found in systems such as  $a\text{-Fe}_x\text{Zr}_{1-x}$  (Ref. 1) and  $\text{Au}_{1-x}\text{Fe}_x$ .<sup>5</sup> Mössbauer spectroscopy<sup>1</sup> shows that above  $T_{xy}$ , the local spin length as a function of temperature coincides with the magnetization  $M$ . However at  $T_{xy}$ , while  $M$  changes smoothly as a function of temperature without any abnormal behavior, the local spin length abruptly increases. It is believed that this behavior reflects the freezing out of transverse components of the spins, as follows. The magnetic hyperfine field  $B_{\text{hf}}$  at the nucleus is approximately proportional to the ordered moment on the atom, independent of the direction in which the moment points. Thus when ferromagnetic (FM) order appears below  $T_c$ , and the transverse components precess rapidly so as to time average to zero, the effective spin length measured by Mössbauer spectroscopy follows the same temperature curve as the order parameter  $M$  measured by magnetization. Below  $T_{xy}$ , if the transverse components freeze out in random directions, their sum remains zero and thus the order parameter does not exhibit any abnormal behavior. However, the local spin length does increase since the frozen transverse components now contribute. This leads to an abrupt increase in the measured hyperfine field at  $T_{xy}$ .  $T_{xy}$  can be determined<sup>1</sup> as the point where  $B_{\text{hf}}$  and  $M$  start to be different and the order becomes noncollinear.

In a recent paper,<sup>6</sup> we examined transverse spin freezing and the critical behavior of a bond frustrated, three-dimensional Heisenberg model designed to replicate the physical situation in the experimental systems discussed above. In this model the frustration was introduced by replacing a fraction ( $f$ ) of the FM exchange bonds by antiferromagnetic (AF) exchange interactions. The system contains some degree of frustration for all  $f \neq 0$ . The model was investigated using standard Metropolis Monte Carlo simulations and the results were in semiquantitative agreement with experimental findings.<sup>1</sup> Transverse spin freezing was found

for  $0 < f < 0.25$ . For all  $f \neq 0$  the critical temperature  $T_c$  is less than  $T_H$  (the critical temperature for the pure Heisenberg model). For  $f > 0.25$  no FM phase can exist and the low temperature phase is a disordered state which possess some spin-glass-like properties.<sup>6,7</sup> Below  $T_{xy}$  the average spin length and the magnetization were found to separate, as in the experimental systems. From finite size scaling arguments it was shown that the transition at  $T_c$  belongs to the Heisenberg ferromagnet universality class for the range of  $f$  studied. The ordering event at  $T_{xy}$  was short range, and no true phase transition occurred. Furthermore, there was no loss of FM order when the transverse spin components froze. For this model the Monte Carlo results of the spin behavior at the  $T_{xy}$  line were qualitatively similar to that on the FM- $M1$  phase boundary in the mean-field model,<sup>8</sup> although in this latter case the line marked a true thermodynamic phase transition.

The bond-frustrated model clearly describes transverse spin freezing in systems such as  $a\text{-Fe}_x\text{Zr}_{1-x}$ , where there is only one magnetic species, and the magnetic interactions are determined by nearest-neighbor distances. However, there are other magnetic systems for which this model is not appropriate. Two examples are  $\text{Eu}_{1-x}\text{Gd}_x\text{S}$  (Refs. 9,10) and  $\text{Fe}_{3-x}\text{Mn}_x\text{Si}$ ,<sup>11</sup> in which a spin-glass-like phase is induced by site disorder. For small values of  $x$  the systems remain ferromagnetic with some reduction in  $T_c$ ; however, further substitution leads to the development of a second transition (which we denote  $T_{xy}$ ), below  $T_c$ , to a state with irreversibilities reminiscent of a spin glass. The two transitions merge with increasing  $x$  to yield a spin glass, while the fully substituted materials ( $\text{GdS}$  and  $\text{Mn}_3\text{Si}$ ) are both antiferromagnetic. As has been the case with the bond-frustrated systems described above, the behavior at  $T_{xy}$  in these site-frustrated materials has often been described as being either "reentrant" with the FM order transforming to a spin glass, or, in AF-dominated compositions, due to the freezing of FM clusters in an AF matrix. Since the phenomenology appears similar to the bond-frustrated case, it is of interest to investigate whether there are any differences in the details of the ordering in site-frustrated systems, particularly in the behav-

ior at  $T_{xy}$  which is driven entirely by the presence of the frustration and might be expected to be sensitive to the manner in which the frustration is introduced.

The purpose of this work is therefore to investigate the behavior of the site-disordered Heisenberg model using Monte Carlo simulations, looking especially at the transverse spin freezing phenomenon. Theoretically, site disorder can be introduced by replacing a fraction  $f$  of FM sites by AF sites. In contrast with the bond-disordered model, no frustration is expected if  $f$  is small. However, at larger values of  $f$  when there are nearest-neighbor AF sites in the system, the competing interactions will lead to frustration and change the magnetic behavior. We have computed the quantities related to transverse spin freezing, and our conclusion is that for values of  $f$  up to  $\sim 0.17$ , no transverse spin freezing is found; beyond this value we observed freezing. At large  $f$  a “mixed phase” is observed. This state is characterized by coexisting FM and AF order. We have traced the  $T_{xy}$  line and mapped out the global phase diagram of this model in the  $f$ - $T$  plane. Comparison with experimental systems shows excellent agreement with the predictions of the site-frustrated model.

## II. THE MODEL

We consider a system of classical Heisenberg spins,  $\vec{S}_i$ , on a simple cubic lattice in zero magnetic field. The Hamiltonian is given by

$$H = - \sum_{\text{NN}} J_{ij} \vec{S}_i \vec{S}_j. \quad (1)$$

Here  $J_{ij}$  is the exchange interaction between nearest-neighbor spins on lattice sites  $i$  and  $j$ , respectively. We introduce site disorder into the system by replacing a fraction  $f$  of FM sites by AF sites. This is achieved using three possible values for  $J_{ij}$ :  $J_{\text{FF}}$  representing the interaction between two FM sites,  $J_{\text{AF}}$  representing the interaction between a FM and an AF site, and  $J_{\text{AA}}$  representing the interaction between two AF sites. Thus a FM site is replaced by an AF site if the nearest-neighbor bonds are changed appropriately. In the calculations we used  $J_{\text{FF}} = +J$ ,  $J_{\text{FA}} = -J$ , and  $J_{\text{AA}} = -J$ . This choice was made for simplicity, and is partially justified by the experimental observation of reentrance in site-frustrated systems with  $J_{\text{FF}} \approx -J_{\text{FA}}$ .<sup>10</sup> The values for the Curie temperature of the FM Heisenberg model ( $f=0$ ) and the Néel temperature of the AF Heisenberg model ( $f=1$ ) are identical,  $T_H = 1.44J$ .

With this choice for the values for  $J_{ij}$  we expect the following basic magnetic phase behavior for the model. When a FM site is replaced by an isolated AF site, the average FM coupling of the system is reduced. Therefore the FM transition temperature is expected to be lower than  $T_H$ . This is true for all values of  $f \neq 0$ . On the other hand, replacing an AF site with an isolated FM site does not change the average AF coupling of the system since the two coupling constants are equal:  $J_{\text{AA}} = J_{\text{FA}} = -J$ . We expect this to be true until FM-FM pairs start to appear, which will be at  $f = f_t \equiv 1 - q^{-1}$ , where  $q$  is the coordination number of the lattice. Beyond this point ( $f < f_t$ ) FM-FM pairs will reduce the average AF exchange strength and  $T_N$  will fall. For

$f > q^{-1}$  the presence of AF-AF pairs in the FM matrix will lead to frustration and transverse spin freezing. Furthermore, concerning the transverse spin freezing phenomenon, the model is symmetric in  $f$ : exchanging one site in either of the pure systems does not generate any frustration until we have exchanged two neighboring sites. We therefore expect the system to exhibit similar behavior at  $f$  and  $1-f$ . On average, frustration should set in at  $f = q^{-1}$  on the FM side and at  $f = 1 - q^{-1}$  on the AF side.

The model with site disorder was investigated using the standard Metropolis Monte Carlo method. A three-dimensional system of  $8^3$  spins on a simple cubic lattice was initialized with high temperature configurations.<sup>12</sup> The system was then annealed via 25 temperature intervals to a low temperature of  $0.005T_H$ . At each temperature, statistical averages of relevant physical quantities were taken. A calculation of the relaxation time for temperatures away from  $T_c$  revealed that about 2000 Monte Carlo steps per spin (MCSs) were needed to reach equilibrium at such temperatures, while a larger number of MCSs were needed close to  $T_c$  where critical fluctuations become important. After the system reached equilibrium, statistical averages of physical quantities were collected over 4000 MCSs. We found that fluctuations in the energy were indeed Gaussian distributed for 4000 MCSs indicating that our choice of the number of steps was reasonable. Finally, results for 10 different realizations of the site randomness for a given  $f$  were averaged. The values of  $f$  used in the simulations ranged from 0 to 1 and we were thus able to calculate the entire magnetic phase diagram for the system.

In order to investigate the behavior of the transverse spin components, we need to keep track of the transverse spin plane in the system. This can be done in the following two ways. (1) Applying a small external field removes the rotational invariance of the Hamiltonian and one may then take the direction of the external field as a fixed  $z$  axis in the system. This is commonly used in experiments. For numerical calculations this external field needs to be of the order  $0.05J$  in order to keep the magnetic order fixed in space (and thus to define a meaningful  $z$  direction). However, the coupling constant  $J$  in a system such as  $\text{Eu}_{1-x}\text{Gd}_x\text{S}$  is of the order 150 T (Ref. 13) and thus even at  $0.05J$  the external field is actually quite high. (2) The  $z$  axis may be defined as the direction of the bulk magnetization (i.e., either the FM order parameter or the AF order parameter depending on the value of  $f$ ).<sup>14</sup> With this definition of the  $z$ -axis, the transverse plane (the  $xy$  plane) is also specified. We have used method (2) here to avoid any effects of externally applied field, and to eliminate the complication of having to apply a staggered field in the AF-dominated half of the phase diagram.

We compute a time average of the spin components and of the square of the transverse spin components at each site, i.e.,

$$\vec{m}_i = \frac{1}{\tau} \sum_{\tau'=1}^{\tau} \vec{S}_i(\tau')$$

and

$$m_{\perp i}^2 = \frac{1}{\tau} \sum_{\tau'=1}^{\tau} [S_{ix}(\tau')^2 + S_{iy}(\tau')^2],$$

where, as mentioned above,  $\tau=4000$  Monte Carlo (MC) steps. From these local quantities the following spatially averaged quantities are extracted.

- (1) The root-mean-square spin length

$$S_{\text{rms}} = \frac{1}{N} \sum_{i=1}^N (\vec{m}_i \cdot \vec{m}_i)^{1/2}.$$

- (2) The bulk magnetization

$$M_f = \frac{1}{N} \left| \sum_{i=1}^N \vec{m}_i \right|;$$

this is the order parameter for the FM phase.

- (3) The staggered magnetization

$$M_{\text{st}} = \frac{1}{N} \left| \sum_{i=1}^N \mathcal{S} \vec{m}_i \right|,$$

where  $\mathcal{S}$  is a matrix with the symmetry of the AF structure of the lattice. This is the order parameter for the AF phase.

- (4) The mean square of the spin projection on the  $xy$  plane

$$Q_{\perp} = \frac{1}{N} \sum_{i=1}^N m_{\perp i}^2.$$

This quantity measures the mean transverse spin length.

- (5) The  $xy$  projection of the time averaged spin components

$$Q_{xy} = \frac{1}{N} \sum_{i=1}^N (m_{ix}^2 + m_{iy}^2).$$

The last two quantities take into account ordering in the  $xy$  plane and may therefore exhibit the onset of noncollinearity and transverse spin freezing in the system.

### III. RESULTS

Figures 1(a)–1(d) show  $M_f$ ,  $S_{\text{rms}}$ ,  $M_{\text{st}}$ ,  $Q_{\perp}$ , and  $Q_{xy}$  as functions of temperature, for several values of concentration  $f$ . Figure 1(a) shows the pure FM case ( $f=0$ ). As expected, the FM order parameter  $M_f$  and the local spin length  $S_{\text{rms}}$  are identical below the critical temperature  $T_c$  ( $=T_H$  in this case), as the system orders in a collinear configuration. Above  $T_c$ ,  $Q_{\perp}$  takes the value  $2/3$  since the spins are randomly rotating in three dimensions thus spending  $2/3$  of the time in the transverse plane where  $Q_{\perp}$  is computed. Below  $T_c$ ,  $Q_{\perp}$  decreases, reaching zero at  $T=0$  as expected for  $f=0$ . Naturally the quantity  $Q_{xy}$  is zero for all  $T$  when  $f=0$  since there is no frustration and thus no transverse spin freezing.

Figure 1(b) shows the data for  $f=0.12$ . As mentioned above, this concentration is below the concentration needed to form AF clusters and the AF sites are, on average, isolated in the FM matrix and thus do not generate any frustration. Indeed, the system behavior is essentially the same as the  $f=0$  case except that the bulk magnetization,  $M_f$ , is now lower than the root-mean-square spin length  $S_{\text{rms}}$  below  $T_c$ . However, this difference has a trivial origin: the AF sites in the system simply order antiparallel to the FM sites, hence

the FM order parameter  $M_f$ , calculated as the sum of all spins, is reduced by 24% ( $= 2 \times 12\%$ ) at  $T=0$ , as observed in our data. We note also that the FM transition temperature  $T_c$  has shifted to a lower value.

Up to  $f \sim 0.17$ , the behavior is similar to the  $f=0$  case, but as we increase the site disorder further, features start to appear. Figure 1(c) shows the data for  $f=0.18$ . Here we can see that  $Q_{xy}$  becomes nonzero at a low temperature  $T_{xy}$ . Since  $Q_{xy}$  measures the time-averaged length of the spin components in the transverse plane, a nonzero value of  $Q_{xy}$  signals the freezing out of that component. In this situation  $Q_{\perp}$ , the mean square of the spin projection on the  $xy$  plane, remains nonzero as  $T$  approaches 0. Hence as we expected, transverse spin freezing is observed around the threshold value for the appearance of AF pairs at  $f \sim 0.17$ . This behavior is quite different from that of the bond-disordered system, where any finite  $f$  gave rise to transverse spin freezing.<sup>6</sup> In the bond-disordered system, frustration may be induced by replacing just one FM bond with an AF bond, whereas in the site-disordered model, replacement of a single FM site by an isolated AF site only decreases the bulk magnetization and has no other effect. Frustration can only occur for the site-disordered model when there are at least two neighboring AF sites. This occurs on average at a value given by  $f = q^{-1}$ , as mentioned above. For the simple cubic lattice studied here,  $q=6$ , and thus we expect frustration to appear for  $f > 0.167$  as observed here. Finally, we note that the difference between  $M_f$  and  $S_{\text{rms}}$  below  $T_c$  is now nontrivial: at  $T=0$  for  $f=0.18$  it is more than the 36% which would be expected simply from reversed spins on AF sites. There is now a contribution from the noncollinearity associated with transverse spin freezing, induced by the presence of frustration at this concentration of impurities. As might be expected, the frustration at  $f=0.18$  is weak and the transverse spin freezing does not dominate the ordering, and the difference between  $S_{\text{rms}}$  and  $M_f$  remains quite close to 36%.

For higher values of  $f$ , the features present in Fig. 1(c) become more pronounced. The increased frustration in systems with  $f > q^{-1}$  makes it easier to freeze out the transverse spin components, and  $T_{xy}$  increases. At the same time,  $T_c$  shifts to lower values reflecting the decline in the average exchange strength as the level of disorder is increased. Increased frustration also leads to larger transverse components and thus a greater difference between  $M_f$  and  $S_{\text{rms}}$ . Figure 1(d) shows the behavior at  $f=0.36$ .

Our results on the bond-frustrated model showed that the phase transition at  $T_c$  was not destroyed by small amounts of impurities,<sup>6</sup> and we expect the same to be true of the site-frustrated system. This was checked using finite size scaling analysis. We computed the susceptibility  $\chi(T, L)$  for systems of various linear sizes  $L$  at temperatures  $T$  near  $T_c$  for fixed  $f$ . From the finite size scaling ansatz,<sup>15</sup> near  $T_c$   $\chi$  should scale as  $\chi \sim |t|^{-\gamma} \bar{\chi}(Lt^{\nu})$ , where  $t$  is the reduced temperature and  $\gamma$  and  $\nu$  are critical exponents for  $\chi$  and the correlation length. For smaller system sizes the data were averaged over at least 15 realizations of the site disorder  $f=0.36$ . For larger system sizes  $L=16$  and  $20$  we used five realizations. The critical exponents for the pure FM Heisenberg model in three dimensions<sup>16</sup> were used. The data collapse shown in Fig. 2 is quite reasonable very close to the transition, indicating that the long range FM order below  $T_c$  is not destroyed at least

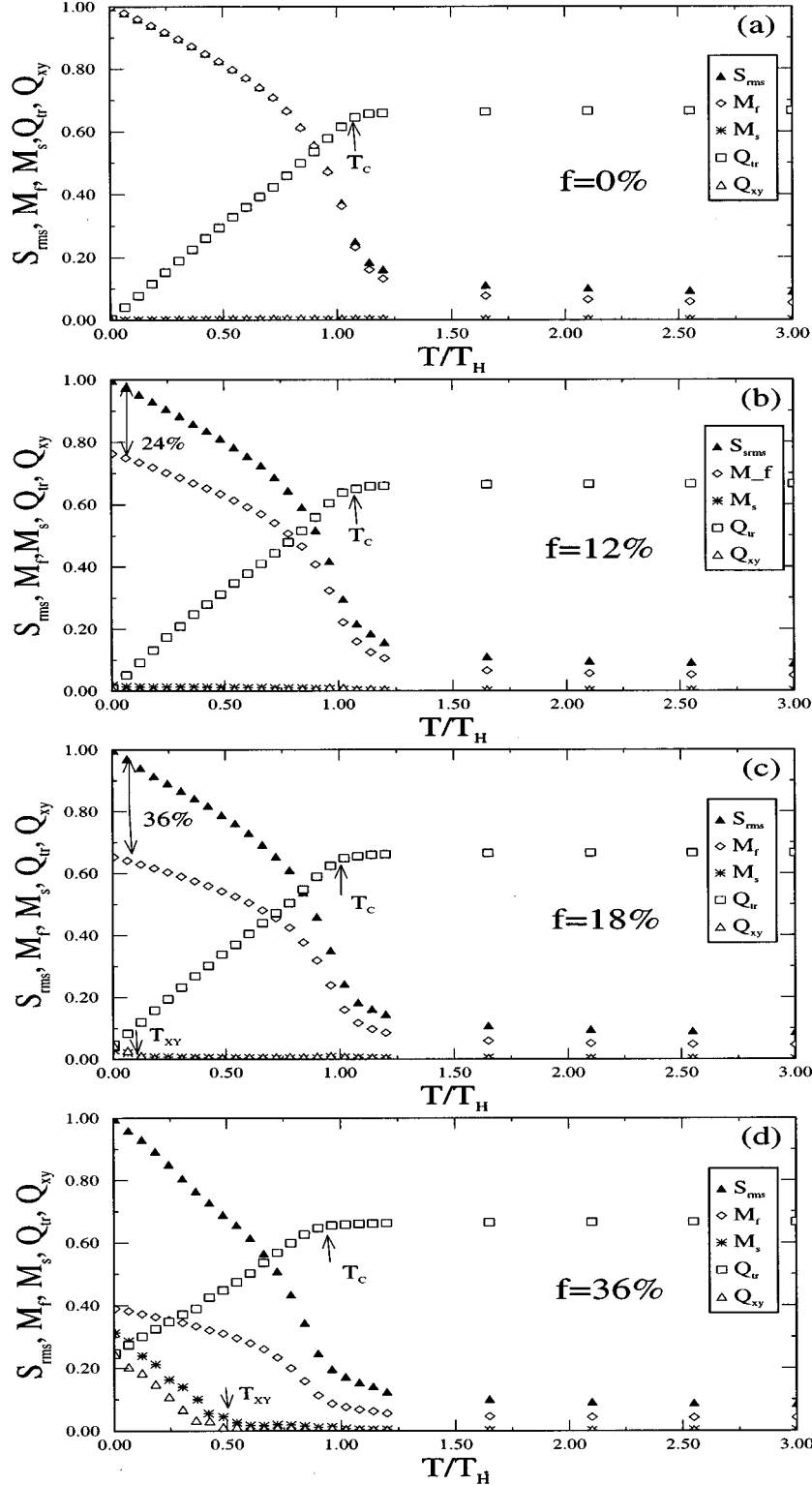


FIG. 1. Temperature dependence of the different spin averages. Solid  $\triangle$   $S_{rms}$ ,  $\diamond$   $M_f$ ,  $*$   $M_{st}$ ,  $\square$   $Q_{\perp}$ , and open  $\triangle$   $Q_{xy}$ .  $T_c$  is the temperature where  $Q_{\perp}$  starts to decrease from  $2/3$ . (a)  $f=0\%$ ; (b)  $f=0.12$ ; (c)  $f=0.18$ ; and (d)  $f=0.36$ .  $T_{xy}$  in (c) and (d) is the temperature where  $Q_{xy}$  becomes nonzero.

up to  $f=0.36$ . Furthermore, since the critical exponents used for the scaling analysis were those of the pure Heisenberg FM, the site disorder does not seem to change the universality class of the model.

Apart from the percolation related threshold for the onset of transverse spin freezing, the results obtained for the bond- and site-frustrated models appear very similar. However, the strong correlations between the locations of the antiferro-

magnetic bonds (all links to disorder sites are AF) coupled with the requirement that AF-AF first neighbor pairs exist for frustration to be present, leads to behavior which appears in the antiferromagnetic order parameter,  $M_{st}$ , the staggered magnetization. On increasing  $f$  from zero,  $M_{st}$  remains zero at all temperatures until the threshold concentration for transverse spin freezing is reached. Beyond  $f=0.17$ ,  $M_{st}$  becomes nonzero at  $T_{xy}$ , i.e., the same temperature at which the trans-

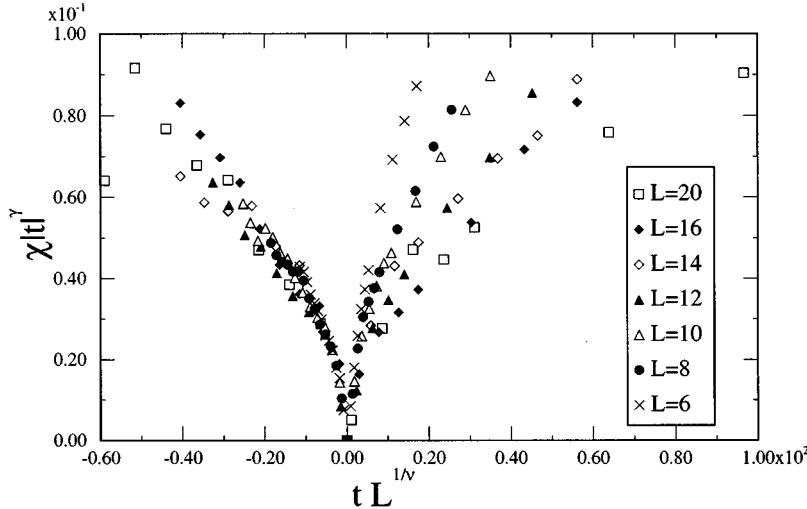


FIG. 2. Finite size scaling for  $\chi$  at  $f=0.36$ . The critical exponents  $\gamma$  and  $\nu$  are those of the Heisenberg FM. The data are averaged over at least 15 realization of disorder, except for  $L=16$  and 20 where only five realizations were used.

verse spin components freeze. Furthermore, the direction along which  $M_{st}$  is a maximum always lies in the  $xy$  plane, perpendicular to the magnetization. These observations suggest the existence of AF correlations in the transverse spin components. Indeed, small AF domains are clearly visible in Fig. 3, which shows a typical configuration of the transverse spin components for a  $20^3$  system for  $f=0.36$  and  $T=0.1T_H$ . We found that the correlation length in the transverse plane increased with the system size and the average size of the domains became somewhat larger. Unfortunately we were not able to study much larger systems as the computational effort became prohibitively demanding. We expect that the domain size will remain finite in the thermodynamic limit since no phase transition occurs at  $T_{xy}$ .

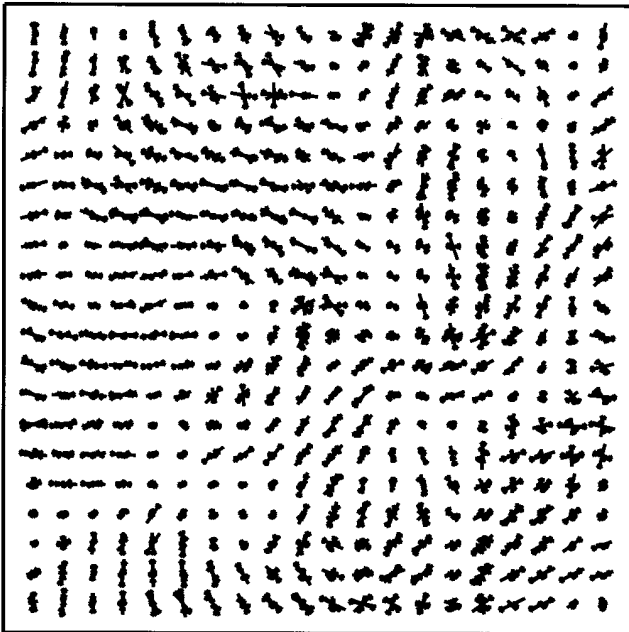


FIG. 3. Time averaged spin configuration for  $f=0.36$  and  $T=0.1T_H$  of a  $20^3$  system projected onto the  $xy$  plane. Each point in the plot is the projection of the first six planes in the lattice.

Figure 4 shows an estimate of the phase diagram for this model. The phase transition lines from the paramagnetic (PM) to the ferromagnetic (FM) and antiferromagnetic (AF) states were determined by calculating the susceptibility of the system. We took the position of the maximum in the susceptibility as the transition point.<sup>17</sup> The  $T_{xy}$  lines were determined from the temperature where  $Q_{\perp}$  just became non-zero. We emphasize that there is no phase transition at  $T_{xy}$  and it represents only the short range freezing of transverse spin components. We have previously shown in the bond-frustrated model that although the susceptibility has a peak at  $T_{xy}$ , this peak does not grow as the system size is increased.<sup>6</sup> Thus the  $T_{xy}$  line is only a disorder line signaling the buildup of short range order due to the transverse spin freezing. The nature of the  $T_{xy}$  line in the site-frustrated model studied here is the same as that of the bond-frustrated model. The onset of transverse spin freezing occurs at  $f \sim 0.17$  in the FM state, and  $f \sim 0.83$  in the AF state. The AF phase transition temperature stays constant at  $T_N = T_H$  for  $1 \geq f \sim 0.83$  then drops slightly as the effects of frustration become significant, while for all  $f \neq 0$  the FM phase transition temperature  $T_c$  is lower than  $T_H$ . On the FM side,  $T_c$  and  $T_{xy}$  merge in the region between  $f=0.45$  and  $f=0.50$ . However, we cannot determine from our numerical data the exact point at which  $T_c$  and  $T_{xy}$  merge. In the case of the bond-frustrated model, this point is  $f \approx 0.25$  (Ref. 6) and there is a region in the middle of the phase diagram where the PM phase crosses over to a spin-glass-like behavior as  $T$  is reduced. We expect that the same is true here for the site-frustrated model near  $f=1/2$ , namely that the PM phase should cross over to the mixed behavior (see below and Fig. 4) in that region.

Apart from the states previously observed in the bond-frustrated model, there is a region in the lower middle of the phase diagram shown in Fig. 4 which we have labeled “mixed.” To locate this region, we obtained the susceptibility at fixed temperature while sweeping  $f$ . A peak occurred in the susceptibility at the dashed line in Fig. 4, which separates the “mixed” region from the FM and AF regions on either side. Both the FM and the AF order parameters are significant in the “mixed” region as a result of the AF domains in the  $xy$  plane coexisting with the FM  $z$  components

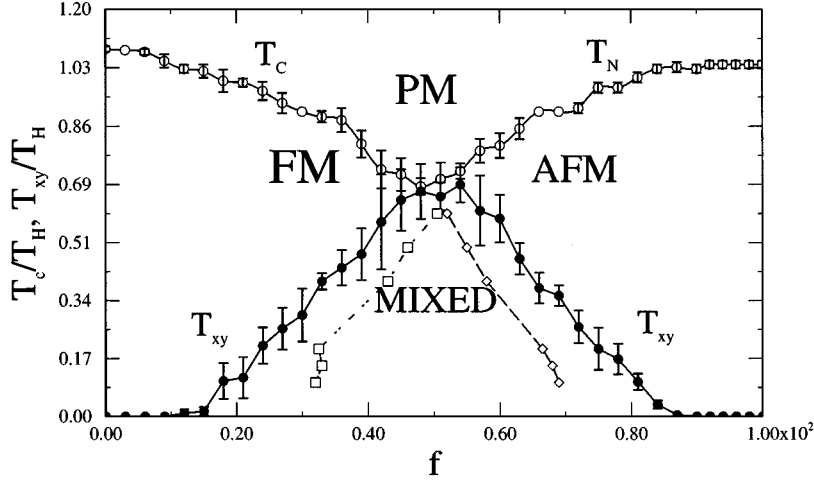


FIG. 4. An approximate phase diagram of the model. Open circles are temperatures  $T_c$  and  $T_N$ . Solid circles are temperatures  $T_{xy}$ . The dashed line with open boxes and diamonds roughly separates the mixed phase from the FM and AF regions. The line is determined from the point of maximum susceptibility as we sweep  $f$  at fixed temperature. The error bars are the standard deviation of 10 realizations of site randomness for each value of  $f$ .

of the spins. The spins are thus quite noncollinear in this region of the phase diagram. This conclusion was checked directly by examining the spin configurations, such as that shown in Fig. 3. In the “mixed” region many spins are tilted away from the  $z$  direction along which the magnetization is a maximum, and the  $xy$ -spin components are frozen into AF configurations. The “mixed” region, for  $f < 0.45$ , is thus characterized by a substantial FM order parameter defining the  $z$  direction, combined with short range AF order in the transverse plane. For  $f > 0.55$ , on the other hand, it is characterized by a substantial AF order parameter in the  $z$  direction, mixed with short ranged FM domains in the transverse plane. Since the spins are not separated into two distinct phases by an interface, the “mixed” state is not a true coexistence region. Again, we do not anticipate a true phase transition entering the “mixed” region, as the path from FM or AF to the “mixed” region merely involves a gradual slowing down of the motion of the spin components in the  $xy$  plane until it becomes so slow that these components are dynamically frozen. Since transverse spin freezing starts to occur at  $T_{xy}$  where  $Q_{xy}$  becomes nonzero, and AF domains with a substantial  $M_{st}$  in the transverse plane appear inside the mixed region, it is reasonable to speculate that upon increasing the system size and the Monte Carlo sampling times, the  $T_{xy}$  and the dashed line in Fig. 4 will actually merge. Unfortunately a numerical check on this possibility is beyond our current computational ability. In any event, an important conclusion is that the phase behavior here is quite different from that of the bond-frustrated model.<sup>6</sup>

Our calculated phase diagram exhibits all of the features observed for  $\text{Eu}_{1-x}\text{Gd}_x\text{S}$ , with two transitions being observed in both the AF and FM dominated compositions.<sup>10</sup> Even the more rapid decline in  $T_c$  with composition compared with  $T_N$  is reproduced. The requirement for first-neighbor impurity pairs to occur before frustration appears, coupled with the need for substantial frustration to be present before the FM or AF order can be destroyed, explains the rather narrow composition range over which the spin glass is observed.<sup>9</sup> The observation of substantial FM and AF order parameters in the mixed region in the lower center of our phase diagram is fully consistent with both  $\chi_{ac}$  and heat capacity measurements, which were interpreted as showing evidence of ferromagnetic clusters for  $x > 0.5$  where AF in-

teractions were expected to be dominant.<sup>9</sup> However, examination of the ordering in our model shows that no segregation into FM clusters within an AF matrix occurs. The FM order does not appear as clusters of FM spins, but rather as short-ranged FM correlations in the transverse spin components perpendicular to the AF ordering direction.

The rather high capture cross sections of both Eu and Gd make detailed neutron diffraction studies of the magnetic structure in  $\text{Eu}_{1-x}\text{Gd}_x\text{S}$  extremely difficult, and no data exist. However  $\text{Fe}_{3-x}\text{Mn}_x\text{Si}$  has been studied in some detail, revealing features in remarkable agreement with the model predictions. Initially the Mn atoms substitute on only one of the three possible Fe sites in the structure and no Mn-Mn contacts occur.  $T_c$  and  $M$  both fall with increasing  $x$ , but the effects of frustration (the second transition and magnetic irreversibility) only appear for  $x > 0.75$ , when Mn starts to occupy the other two sites and Mn-Mn pairs are formed.<sup>11</sup> The two transitions merge at  $x \sim 1.75$  to yield a spin glass and complete the expected phase diagram. Neutron diffraction studies of powder samples have shown that the second transition is associated with the onset of an antiferromagnetic component in the magnetic structure.<sup>11</sup> While more recent polarized neutron scattering on a single crystal of  $\text{Fe}_2\text{MnSi}$  confirmed the presence of the AF ordering at  $T_{xy}$  and, more significantly, showed that the AF correlations which appear at  $T_{xy}$  develop only in the plane perpendicular to the FM order.<sup>18</sup>

#### IV. CONCLUSIONS

Our Monte Carlo simulations on the site-disordered Heisenberg model give qualitatively similar results for certain phenomena as those of the bond-frustrated model. In particular transverse spin freezing is observed which leads to an increase of the local spin length without affecting the ferromagnetic order formed at  $T_c$ . However, frustration does not set in until the site disorder reaches  $\sim 0.17$  where AF-AF pairs become likely. Thus the transverse spin freezing temperature  $T_{xy}$  remains zero until  $f$  reaches that value. For the FM system we observed a decrease in  $T_c$  for all  $f \neq 0$ , while  $T_N$  for the AF system remains unchanged until the onset of frustration. We have observed a “mixed” configuration for large values of the site disorder at low temperatures. The

magnetic order in this region is characterized by substantial values of both the FM and the AF order parameters. This is possible since FM order involves only the  $z$  components of the spins, while the AF order parameter comes from the transverse components. The “mixed” state is not a state of coexistence since the spins are not separated by a physical interface. Finite size scaling near  $T_c$  showed that the moderately disordered Heisenberg model has a long range ordered phase below  $T_c$  and that the transition at  $T_c$  belongs to the Heisenberg FM universality class. At the  $T_{xy}$  line or on entering the “mixed” region of the phase diagram, we believe there is no true phase transition and the events represent the

dynamical freezing of the transverse spin components. Finally, the estimated phase diagram of this model is consistent with experimental findings, and the predicted mutually perpendicular FM and AF order below  $T_{xy}$  has been observed in  $\text{Fe}_2\text{MnSi}$ .<sup>18</sup>

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<sup>12</sup>Some simulations were performed using  $20^3$  spins to check for finite size effects and qualitatively the same results were obtained.

<sup>13</sup>In a mean-field solution to the pure Heisenberg ferromagnet the critical temperature is given by  $T_c = qJ/k_B 3$ , where  $q$  is the coordination number of the lattice. For  $\text{Eu}_{1-x}\text{Gd}_x\text{S}$   $T_c \sim 100$  K and hence  $J \sim 150$  T.

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<sup>17</sup>Proper finite size scaling analysis should be performed to accurately determine the transition points, as was done in Ref. 6. However, here we are primarily interested in the general phase behavior, thus we simply took the peak position of the susceptibility as the transition temperature.

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