# Monte Carlo Study of Spin-Glass Ordering on Dilute Frustrated Lattices

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Results of computer simulations for a dilute Ising system on the triangle and fcc lattices are reported. Though we include only a nearest-neighbor antiferromagnetic interaction $J < 0$, these systems freeze into a spin-glass for $0.50 < x < 1.0$ for the triangle lattice and $0.18 \lesssim x \lesssim 0.40 \pm 0.10$ for the fcc lattice, where $x$ is the concentration of magnetic spins. The spin-glass freezing is a result of the frustration inherent in the lattice and not from competition between ferromagnetic and antiferromagnetic bonds.

Spin-glasses are random dilute alloys which have interesting low-temperature properties.[1] The most extensively studied systems have been the metallic alloys, such as AuFe and CuMn. In these alloys, the long-range oscillatory Ruderman-Kittel-Kasuya-Yosida interaction via the conduction electrons results in a competition between ferromagnetic and antiferromagnetic interactions which gives rise to the spin-glass state. At present, however, there is no consensus on whether this state really exists or not. For this reason, it is important to study other, possibly simpler systems which also show spin-glass ordering. Possible systems include dilute magnetic insulating and semiconducting materials, in which the interaction is predominantly short ranged and not mediated by the conduction electrons. The insulating system[2] $(Eu_xSr_{1-x})S$, with $0.13 \leq x \leq 0.5$ is one example. The low-temperature spin-glass properties have been modeled by including ferromagnetic nearest-neighbor (nn) and antiferromagnetic next-nearest-neighbor (nnn) exchange. However, this competition between ferromagnetic and antiferromagnetic interaction is not common in insulators[3] and probably cannot apply to the magnetic semiconductors $Cd_{1-x}Mn_xTe$ and $Hg_{1-x}Mn_xTe$. In these systems measurements of the specific heat and susceptibility indicate that they are probably spin-glasses in which only antiferromagnetic nn interactions are present.[4] The spin-glass freezing in this system is a result of the frustration in the underlying lattice. This ordering was first suggested by De Seze[5] and recently discussed by Villain.[3] It is the purpose of this Letter to present the first Monte Carlo simulations that show spin-glass freezing can occur in a system with only antiferromagnetic nn interactions.

We studied a dilute Ising model with nn antiferromagnetic (AF) interactions on the two-dimensional (2D) triangle and 3D fcc lattices. Both lattices have a high ground-state degeneracy. With no dilution, the triangle lattice is fully frustrated and remains paramagnetic down to $T = 0$. The fcc lattice is only partially frustrated and has a first-order transition to an infinitely degenerated AF ground state in which one third of the bonds are unsatisfied.[6] When some of the spins are randomly removed, it is possible to obtain a spin-glass state. Let $x$ be the concentration of spins present; then for $0.50 < x < 1.0$ in the triangle and $0.18 \lesssim x \lesssim 0.40 \pm 0.10$ in the fcc lattices, we find that the system orders into a spin-glass state at a finite temperature. The lower limit in each case is the theoretical percolation threshold, below which only finite clusters are present and the system must be paramagnetic down to $T = 0$. We also find that the first-order transition for $x = 1.0$ in the fcc lattice becomes second order for $x < x_t$, with a tricritical point $x_t = 0.93 \pm 0.01$. Since this is a finite-time simulation, we cannot say whether these are real spin-glasses in the limit of infinite time or whether it is only a finite-time effect. Relaxation studies for long times give results similar to those found for the random Gaussian spin-glass model. We will consider these states to be spin-glasses, in the same sense that previous workers have done so.

We find that these systems differ in one remarkable way from models which have competing ferromagnetic and antiferromagnetic interactions. The system is less sensitive to a magnetic field and has negligible thermal remanent effects. The susceptibility does not depend crucially on whether we cool in either a zero or nonzero field. The system relaxes immediately to a state of zero magnetization even for $T < T_{SG}$ from an initial ferromagnetic state. This differs from the random Gaussian model[7,8] and from the experiments for the metallic spin-glasses. This is because all the bonds are AF and strongly oppose the magnetic field. This is compared with models in which half the bonds are ferromagnetic, and support the magnetic field. However, from an initial staggered state, we find that the staggered magnetization does decay very slowly for $T < T_{SG}$.

We studied a model system of Ising spins ($\sigma_i = 2S_i^z = \pm 1$) described by the Hamiltonian

$$\mathcal{H} = -J\sum\nolimits_{nn}\sigma_i\sigma_j - \mu_B H\sum\nolimits_i\sigma_i, \tag{1}$$

where the sum is over nn pairs. The exchange constant is antiferromagnetic $J < 0$, $H$ is the external magnetic field, and we set $k_B = 1$. The spins are situated at random, with a concentration $x$, on either a triangle or fcc lattice with periodic boundary conditions. The simulations were carried out by the Monte Carlo method[7,8] for various size lattices $N$ (here $N$ is the total number of sites, not the number of spins). We applied single-spin-flip (Glauber) dynamics, over time intervals between 700–2200 Monte Carlo steps (MCS) per spin, at various temperatures and initial configurations. Besides the energy and magnetization, the time-averaged Edwards-Anderson[9] order parameter $q(t)$ was calculated to determine the spin-glass ordering. The order parameter $q(t) = \overline{\langle\sigma_i\rangle^2}$ is defined as the configuration average over the square of the magnetization. Though $q(t)$ relaxes towards its equilibrium value very slowly and cannot be considered in equilibrium for the runs made here, it does indicate when spin-glass ordering has occurred. This is in the experimental sense, not in the sense of rigorous equilibrium in the thermodynamic limit.

*Triangle lattice.*—Results for the triangle lattice with nn AF interactions with $x = 1.0$ are well known. The system is completely frustrated and does not order. When some of the spins are removed at random, the large ground-state degeneracy is reduced and the system can order. Because the sites are removed at random, the only possible ordering is into a spin-glass state. In this state, the ground-state energy is usually lower than its value $E(0) = J$ for $x = 1.0$, $T = 0$. In Fig. 1(a), we show results for $q(t)$ after 2.000 MCS/spin for four values of $x$. As $x \to 1.0$, $q(t)$ for $T = 0$ goes to zero, which proves that the simulation is capable of detecting the lack of order for this case. In Fig. 1(b), we show results for $q(t)$ versus time for several temperatures at $x = 0.75$. These are very similar to those obtained previously by Binder and Schröder[8] for the 2D square lattice with Gaussian random interactions. Studies of smaller systems for time up to $10^4$ MCS/spin show nonequilibrium phenomena similar to that observed in Ref. 7. From these results, we conclude that the system will have a finite spin-glass freezing temperature for $0.50 < x < 1.0$, in the same sense that the Gaussian model does. In Fig. 2, we show the results for the heat capacity $C$ obtained from $\partial E/\partial T$. Again, this shows a characteristic spin-glass behavior.

*fcc lattice.*—The fcc differs from the triangle lattice, in that it orders via a first-order phase transition to an AF state for $x = 1.0$. This state has an infinite ground-state degeneracy. The random removal of spins reduces this degeneracy. Phani *et al.*[6] found that the introduction of a ferromagnetic nnn interaction favored an AF ground state in which two of the four simple-cubic sublattice were spin up and two were spin down. They also found a tricritical point at a value of $J_{nnn}/J_{nn} \approx -0.25$. In our case, the removal of random sites splits the ground-state degeneracy, but does not favor the simple ground state found by Phani *et al.*[6] Plots of the average energy $E(T)/J$ vs $T/|J|$ are shown in Fig. 3 for values of $x$ in

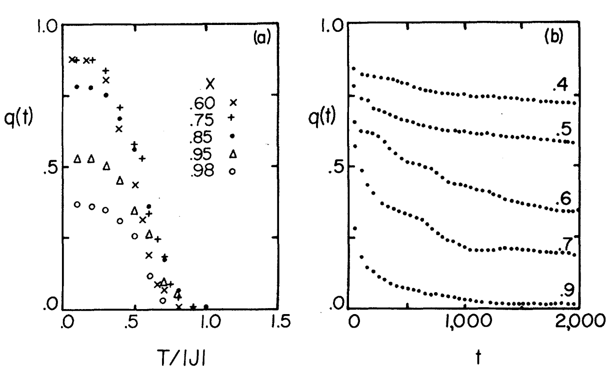


FIG. 1. (a) Order parameter $q(t)$ plotted vs temperature for a 2D triangle lattice, after $t = 2000$ MCS/spin for various concentrations $x$. For $x \geq 0.75$, $N = 2601$ sites while for $x = 0.60$, $N = 3249$ sites. (b) $q(t)$ plotted vs time for $x = 0.75$. The parameter of the curves is $T/|J|$.

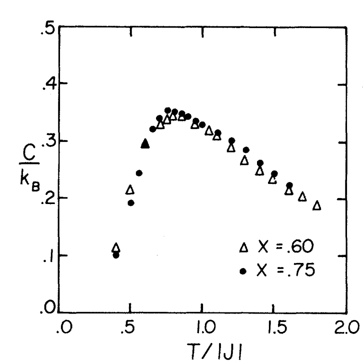


FIG. 2. Specific heat vs $T/|J|$ for $x = 0.75$ and 0.60 for a 2D triangle lattice. Data obtained via $C = \partial E/\partial T$.

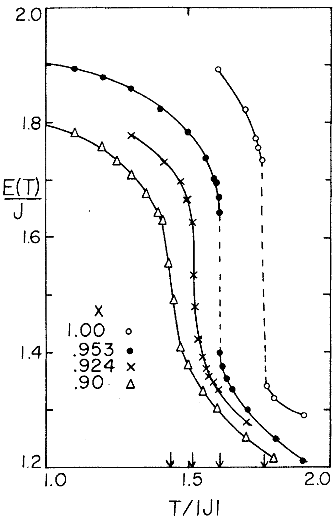


FIG. 3. Average energy $E(T)/J$ vs $T/|J|$ for an fcc lattice for values of $x$ shown and $N=2048$ sites. The transition is discontinuous for $x=1.00$ and 0.953, while it is continuous for $x=0.924$ and 0.90. The transition points are indicated by arrows. Results for $x=1.00$ are consistent with those in Ref. 6.

the range $0.90 \leq x \leq 1.00$. From these results and studies of the two-step relaxation processes on quenching to slightly below the transition temperature from high temperatures, we find that the system has a tricritical point for $x_t \sim 0.93 \pm 0.01$. For $x < x_t$, the transition is second order. As the dilution is increased, our system of 2048 spins breaks up into multiple domains. Finally, for $x \lesssim 0.40 \pm 0.10$, the system cannot have any true AF ordering. Because the AF ordering even for $x=0.90$ is not a perfectly ordered four-sublattice structure, it is difficult to detect where the crossover from AF to spin-glass ordering occurred. That such a crossover has indeed occurred can best be seen from studying samples with $x=0.20$ and 0.30 as shown below. However, as $x$ is lowered, the sharp peak in the heat capacity is reduced and finally becomes rounded, with a maximum at a temperature which is greater than the temperature at which $q(t) \rightarrow 0$.

The dependence of $q(t)$ is similar to that obtained for the triangle lattice. We find that $q(t) \neq 1$ even at low temperatures for $x \lesssim 0.40$, since there are many spins which are in small clusters and do not freeze at $T_{SG}$. In Fig. 4, we show results for $C$ for $x=0.30$ with $H=0$ and $|J|$. Results for $H=0.2|J|$ were indistinguishable from those for $H=0$. This differs from results for the Gaussian model, in which a relatively small magnetic field shifts the maximum in $C$ to higher temperatures. Results for $x=0.20$ (not shown) show a peak maximum at a temperature roughly 1.5 larger than that where $q(t) \rightarrow 0$. In Fig. 5, we show results for $M/H$ vs $T$ for values of $x$ and $H$ shown, obtained by cooling in a field. We found that the system depended little on the field history, as noted above. One sees that $M/H$ for $H=0.2|J|$ has a large paramagnetic contribution at low temperature, which overshadows the contributions from spin-glass freezing. This additional contribution is from the samll, finite clusters which are present at these values of $x$, which are just above the percolation threshold, $x_c \approx 0.18$. The freezing into a spin-glass state is only by those spins which are in the infinitely connected network. The others are free or quasifree and contribute an additional paramagneticlike term to $\chi$ or $M/H$. For the larger field, these free spins

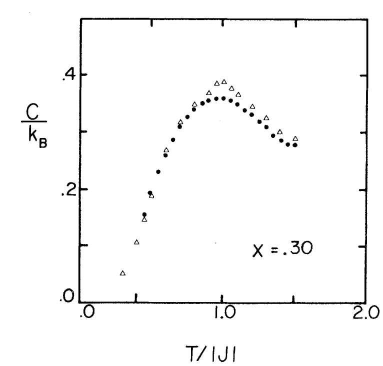


FIG. 4. Specific heat vs temperature for $x=0.30$ with $H=0$ (closed circles) and $|J|$ (open triangles) for an fcc lattice with $N=6912$ sites. Data obtained via $C=\partial E/\partial T$.

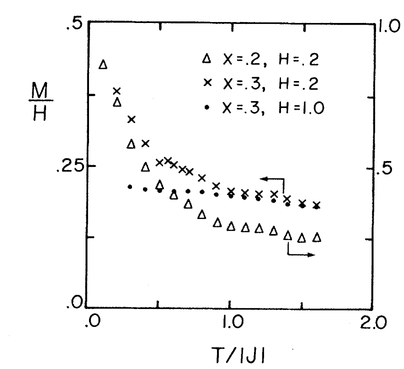


FIG. 5. $M/H$ vs $T/|J|$ for an fcc lattice for the value of $x$ and $H/|J|$ shown. Results are obtained by cooling in the field for a sample with $N=6912$ sites. Note the vertical scale for $x=0.20$ on right, while that for $x=0.30$ is on the left.

are frozen and $M/H$ is essentially constant. We are unable to say whether the contribution to $M/H$ from the infinite cluster is constant for $T < T_{SG}$ as observed both experimentally in AuFe and CuMn and in simulations of the random Gaussian model when the system is cooled in a field.[10] This extra contribution from the paramagnetic spins coupled with the lack of a strong thermal-history effect could cause some problems interpreting experimental results for the susceptibility.

In conclusion, we have observed spin-glass freezing in dilute Ising models which arise from the frustration in the underlying lattice, not as a competition between ferromagnetic and antiferromagnetic interactions. We should reiterate that the spin freezing occurs on a time scale of our "experiment" which was studied up to $10^4$ MCS/spin for some small systems. While this does not prove that real spin-glass ordering exists (no Monte Carlo simulation can ever hope to do this), it does indicate new interesting behavior in a system which is, in our mind, simpler than the canonical model for a spin-glass.

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# Appearance-Potential and Characteristic–Electron-Energy-Loss Spectroscopies of Lanthanum: New Interpretation of Resonant X-Ray Emission

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Using characteristic–electron-energy-loss spectroscopy and different appearance-potential techniques, we have obtained results which suggest a new interpretation of the resonant x-ray emission in lanthanum. Identification of structures in energy-loss and x-ray–emission spectra shows that the resonant x-ray emission is mainly due to characteristic transitions and not bremsstrahlung as believed earlier.

The short-wavelength limit (SWL) parts of the x-ray spectra emitted from lanthanum and cesium have been found to undergo resonantlike intensity variations as the energy of the bombarding electrons is scanned through the excitation potentials of the $3d$ subshells.[1,2] These variations were interpreted as resonances in the cross section for scattering the incident electrons into vacant $4f$ states. This so-called "bremsstrahlung" process results in a final-state configuration $[\mathrm{La}]4f^1$. Theoretical treatment of the bremsstrahlung from La explained such resonances in terms of virtual core-state excitations[3] and thus the phenomenon seemed to be established. There are, however, a couple of features in the SWL spectra which were rather neglected in the above-mentioned analysis, but which in the light of the present results are quite significant. First, in the case of Ce, the SWL spectra shown in Ref. 2 cover an energy range which includes the characteristic emission line $3d^95p^6 \rightarrow 3d^{10}5p^5$. Although the intensity of this line is relatively low so that it cannot

 