"Melting" of Frustrated Spins: Mechanism for Reentrant Ferromagnetic–Spin-Glass Behavior

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The local mean-field equations are solved for XY spins on a square lattice with competing nearest-neighbor interactions. For a model with randomly chosen frustrated *sites*, on lowering the temperature one finds the following sequence of phases: paramagnet; nonuniform ferromagnet; noncollinear, nonuniform ferromagnet; reentrant spin-glass. The reentrant transition occurs via the mechanism of frustrated-spin "melting." For randomly chosen negative *bonds* it is more difficult to obtain reentrance and to provide a physical explanation.

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There is a well established yet rapidly growing experimental literature on the phenomenon known as "reentrance" (of the magnetization), which is associated with disordered magnets. Briefly, one finds that at high temperatures the system is a paramagnet; below a "Curie" temperature T_C the system is a nonuniform ferromagnet; and below a somewhat ill-defined reentrant temperature T_r the system begins to show irreversibility and to lose its magnetization, as it enters a spin-glass (SG) state.[1] For random exchange systems, the infinite-range model with both ferromagnetic and random exchange[2]—in the replica-symmetry–breaking solution of Gabay and Toulouse (GT)[3]—yields a sequence of phase transitions in agreement with experiments sensitive to the transverse spin components, but does not include the reentrant phenomenon. It is the purpose of the present paper to discuss a simple model which, within local mean-field theory, yields (and provides an explanation for) reentrance.

The sequence found by GT is as follows: on lowering the temperature, one goes from the paramagnetic phase (P) to a ferromagneticlike phase (F); then to a mixed phase ($M1$) with both a magnetization and nonzero transverse spin components (equivalently, the $M1$ phase is noncollinear, or canted); and on further lowering of the temperature, the solution develops replica-symmetry breaking (interpreted as the onset of irreversibility), as one enters a phase ($M2$) which, like $M1$ has a magnetization and is noncollinear.[4] This sequence is observed experimentally if, in addition to magnetization measurements, one also employs techniques which are sensitive to the transverse spin components, such as small-angle neutron scattering, Mössbauer effect, or nuclear orientation studies.[5] In particular, a transition to a noncollinear state occurs at a temperature $T_K < T_C$, and at an even lower temperature (T_r) one sees the onset of irreversibility. Nevertheless, the infinite-range model does not yield the observed decrease in magnetization at the onset of irreversibility.

We have undertaken local mean-field calculations on a simple model in order to shed some light on the reentrant phenomenon. We find that, as in the experiments and in the GT theory for the infinite-range model, there are what appear to be three phase transitions, but in the present model the lowest-temperature transition appears to be associated both with the onset of multiple solutions (irreversibility) *and* with reentrance. It is possible to present a clear physical picture of what is happening at this lowest transition temperature, utilizing the concept of "melting" of frustrated spins.

For simplicity, we consider a system of classical spins on a square lattice with nearest-neighbor interactions, and we restrict the spins to lie in a plane (XY model).[6] This has the advantages that it can yield noncollinear states (as opposed to the Ising model), and that a system of only moderate size can have multiple equilibrium configurations (as opposed to the Heisenberg model[7]). An essential aspect of the present model is that it introduces randomly placed frustrated spin *sites* rather than introducing *plaquette* frustration[8] with randomly placed negative bonds. The latter sort of frustration has produced less satisfactory results, and has a less clear physical interpretation.

To appreciate the significance of site frustration, consider the case of a square lattice with only nearest-neighbor ferromagnetic bonds, at zero temperature, within mean-field theory. Clearly, all of the spins will be aligned.[9] Now, at one site let the signs of two of the four bonds be changed, thereby producing a situation wherein the spin at this site has no preferred orientation. We call this *site* frustration. The system lowers its energy with the following sequence of responses: First, the frustrated spin aligns perpendicular to the others; second, its ferromagnetically coupled neighbors tip slightly toward it, and its antiferromagnetically coupled neighbors tip slightly away from it. (Further neighbors also make adjustments, but these are smaller.) Thus, the system is stabilized, but only weakly (through a second-order response). See Fig 1(a) for the case of negative bonds which are opposite one another. If one now raises the temperature, the

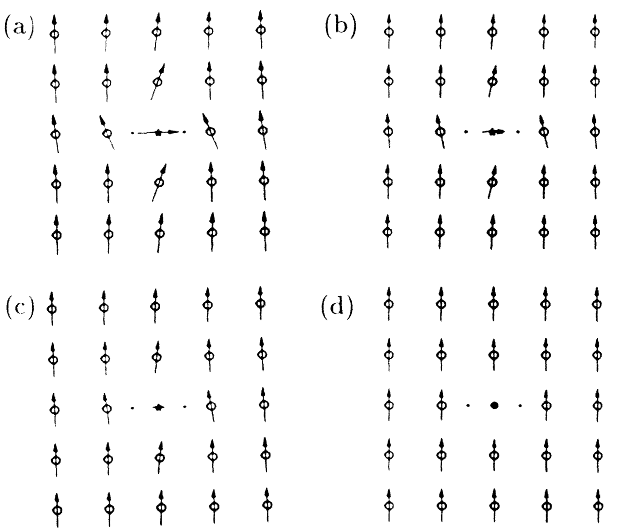

FIG. 1. Solutions to the local mean-field equations for a single frustrated spin, whose negative bonds are given by the dots. The reduced temperatures $T/T_C^{(0)}$ (where $T_C^{(0)}$ is for the host ferromagnetic system) are (a) 0.00, (b) 0.40, (c) 0.45, and (d) 0.50.

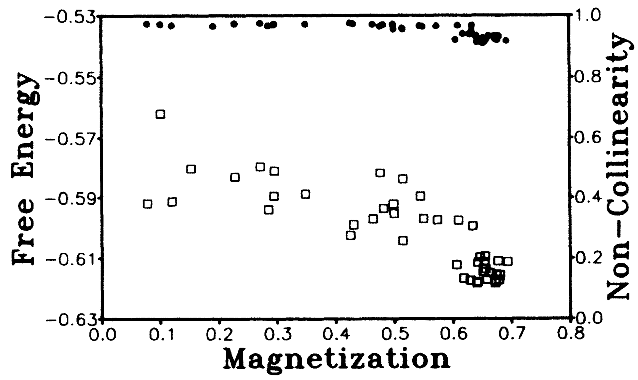

FIG. 2. Scatter plot at $T = 0.05$ of fifty solutions to the mean-field equations. Squares denote free energy and dots denote noncollinearity. The concentration of frustrated spins is $c = 0.20$.

mean-field equations yield the result that *the frustrated spin begins to melt before the others, because it is not held in place by as strong a mean field.* As it melts, it distorts the rest of the spins less. At a temperature which is below $T_C^{(0)}$, the melting (Curie) temperature of the host, the frustrated spin melts completely, leaving the rest of the lattice collinear. This may be seen in Fig. 1, which shows the central 25 spins of a periodically repeated 11×11 lattice, for four temperatures. (The same qualitative considerations hold if the negative bonds are adjacent, but the distortion extends much further in space because the system is somewhat like a dipole, whereas for opposite negative bonds the system is somewhat like a quadrupole.)

One can now consider what should occur as one goes from one to many (but not too many—one does not want to suppress the ferromagnetism completely) frustrated spins. At low temperatures they can be expected to produce a spin arrangement which is so distorted that there is no significant magnetization (i.e., an SG), but at progressively higher temperatures the various frustrated spins can be expected to melt, thereby permitting the rest of the spins to become more collinear. *This melting of the frustrated spins, and the increased alignment of the remaining spins, provides a mechanism for a net magnetic moment to develop at somewhat elevated temperatures, despite the fact that the individual rms spin lengths are shortened because of thermal disorder.*

In what follows we describe a study to corroborate the above mechanism, in the context of relatively small (10×10) periodically repeated lattices. Our procedure is first to determine a lattice by randomly choosing sites, at a variable concentration c, where two negative bonds are placed. This can be done so that they are opposite, adjacent, or randomly chosen (in the present case they were taken to be opposite). This procedure permits overlap to occur, with some sites receiving more than two negative bonds, if a nearest-neighbor site is also chosen. For this lattice, we then iterate the local mean-field equations[10] to a solution at a low temperature, with (typically) fifty randomly chosen sets of initial conditions. (In terms of Ref. 10, our convergence criterion is one part in 10^{12}.) The energy and noncollinearity (see below) are then plotted versus magnetization in an initial-condition scatter plot (see Fig. 2, for $c = 0.20$), or scatter plot, for short. For a given lattice and concentration, a solution is chosen, for which we determine the temperature dependence of the magnetization $m = |N^{-1}\sum_i \langle \mathbf{S}_i \rangle|$, rms spin length $S_{\mathrm{rms}} = [N^{-1}\sum_i |\langle \mathbf{S}_i \rangle|^2]^{1/2}$, and noncollinearity order parameter

$$Q_{\mathrm{nc}} = [(2/N^2)\sum_{ij} |\langle \mathbf{S}_i \rangle \times \langle \mathbf{S}_j \rangle|^2]^{1/4}$$

(where N is the number of spins, and $S_{i\alpha}$ is the αth component of the spin on the ith site). Note that m, S_{rms}, and Q_{nc} all scale linearly with the zero-temperature length of the spins (for convenience, taken to be unity). The factor of 2 ensures that $Q_{\mathrm{nc}} \to 1$ for random spin orientations at $T = 0$, and the cross product ensures that $Q_{\mathrm{nc}} \to 0$ for collinear spins.

Figure 3 presents (from the point of view both of reentrance and of physical interpretation) a particularly good example of what is found. It was obtained by beginning with a low-magnetization solution at low temperature, and then raising the temperature in small steps, using each previous solution as the starting point for our iteration. Similar curves are found on use of the same low-temperature initial condition at all temperatures. On the other hand, for a cooling procedure where the previous solution is the starting point for the iteration at the next lowest temperature,

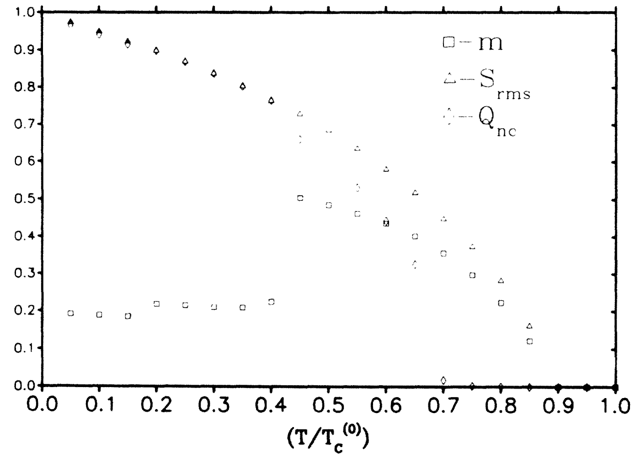

FIG. 3. Temperature dependence of the magnetization m, rms spin length $S_{\rm rms}$, and noncollinearity order parameter $Q_{\rm nc}$, for a solution taken from in Fig. 2.

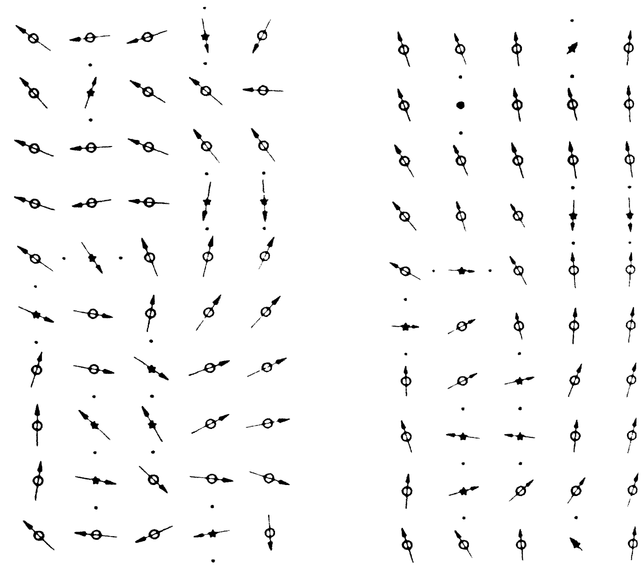

FIG. 4. Spin configurations for half of the lattice employed in Figs. 2 and 3. On the left, $T/T_C^{(0)} = 0.30$; on the right, $T/T_C^{(0)} = 0.50$. Frustrated sites are starred; other sites are circled; negative bonds are indicated by small dots. One spin, whose length is less than 0.15, is indicated as a large dot.

there is a tendency towards collinearity (rather than towards the noncollinear SG phase), and the magnetization saturates (so that the reentrant effect is not observed). This indicates that, at the same temperature at which the magnetization jumps, irreversibility enters the problem, in the form of multiple solutions. We have verified this by performing scatter-plot calculations of free energy[10] and magnetization as a function of temperature, finding a very narrow distribution for the highest temperatures. (For $T/T_C^{(0)} = 0.9$, 0.8, 0.7, the fifty solutions would overlap completely on the scale of Fig. 2; not until $T/T_C^{(0)} = 0.6$ would more than one point appear on the scatter plot; and not until $T/T_C^{(0)} = 0.3$ would there be any obvious spread in the free energy.[11]) Because this standard iteration procedure does not provide a good sampling of phase space, it is not possible to mimic the temperature and field cycling that is observed experimentally. Indeed, even Monte Carlo calculations can be hard pressed to find good regions of phase space in problems involving disorder. For that reason, we believe that "better" low-magnetization states (in the sense of lower free energy) exist. It is expected that the "better" states will show the same sequence of phases.

Examination of Fig. 3 shows that at very low temperatures the system has a low magnetization, and is rather noncollinear, with $Q_{\rm nc}$ near unity (as is characteristic of random spin orientations). As the temperature is increased, all three of m, $S_{\rm rms}$, and $Q_{\rm nc}$ decrease slightly, in approximately the same fashion (indicating that the internal structure is not changing very much), until one reaches a temperature T_r (here, near $T = 0.4T_C^{(0)}$) at which the magnetization undergoes an obvious jump upward, and $Q_{\rm nc}$ undergoes a somewhat less noticeable jump downward. This corresponds to the melting of one of the spins, as can be seen in Fig. 4, which shows "snapshots" of half of the system both before and after the frustrated spin melts. (This is an especially clear example of the melting phenomenon; we have found other cases where reentrance occurs but the snapshots have no such clear physical interpretation.) Note that the frustrated spin does not literally melt; rather, its rms length becomes so short that it can no longer impose itself significantly on its neighbors. When this occurs, the system becomes unstable, and undergoes what amounts to a local first-order phase transition. (This appears to be quite sharp.) In a macroscopic system, the effect of a large number of such local "phase transitions," with a spread in their temperatures, would yield a smooth curve displaying the reentrant phenomenon. (Note that the present system also shows a minor instability near $0.2T_C^{(0)}$.) Observe the greater degree of alignment of the other spins after the frustrated spin has melted. Above the jump, $Q_{\rm nc}$ and $S_{\rm rms}$ no longer track together. Indeed, $Q_{\rm nc}$ goes to zero at a lower temperature (T_K) than do m and $S_{\rm rms}$ (which both go to zero at T_C).

Although we do not display the results, we have varied the strength of the negative bonds somewhat, finding qualitatively similar effects. (Not surprisingly, if the negative bonds are weakened, a larger concentration of them is needed to produce reentrance.) Furthermore, it is found that an applied field extends the ferromagnetic regime to both higher and lower temperatures, as found experimentally.

We have also studied a few 10×10 lattices with randomly chosen *bonds,* finding that it is more difficult to obtain the reentrant phenomenon, and when found, it is less pronounced. However, our studies of this case were less thorough than in the random-sites case.

We now present a physical picture for the sequence of transitions which occurs in the present model, utilizing the concept of melting of frustrated spins, but now working from higher temperatures downward, and treating the curve of Fig. 3 as if it had been obtained on cooling, rather than on heating. As one decreases the temperature below T_C, the ferromagnetic bonds begin to assert themselves, producing a state which is collinear but nonuniform; in this regime, about half of the frustrated spins are antiparallel to the magnetization. As one decreases the temperature below T_K, the frustrated bonds make themselves felt, and the system begins to order in the transverse direction. The rms spin length is so short at T_K that the spins can develop a transverse component at no noticeable cost to the longitudinal one, so that the magnetization continues to rise (cf. Fig. 3.) Eventually, at T_r, the most frustrated spins finally freeze (typically, transverse to the net magnetization), and at this point the rms spin length of the other spins is sufficiently well developed that they can only adjust to the newly frozen frustrated spins by tipping, thus leading to a decrease in the magnetization (cf. Fig. 4). Note that the magnetization need not go to zero for this mechanism to be appropriate: In the present case $m \approx 0.2$, whereas a random sum of 100 unit spins would give $m \approx 0.1$.

Let us now consider what the calculation tells us about the nature of each of the transitions. First, the magnetization displays the square-root onset (cf. Fig. 3) which is characteristic of what happens in mean-field theory at second-order phase transitions, so that T_C indeed indicates a second-order phase transition (associated with a change in the order of the system, from paramagnetic to ferromagnetic). Second, the noncollinearity order parameter also displays what appears to be a square-root onset, so that T_K, too, indicates a second-order phase transition (associated with a change in order of the system, from collinear to noncollinear). On the other hand, at T_r the magnetization appears to undergo a discontinuous drop, unlike what occurs experimentally. As discussed above, this is due to a local phase transition, and when averaged over a macroscopic system, the spread in local melting temperatures will yield a smooth curve of no obvious characteristic behavior, other than a decrease in the magnetization: the reentrant phenomenon.

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[1]Over twenty experimental papers on such topics have been reported at both the Thirtieth Conference on Magnetism and Magnetic Materials [see the spin-glass sections in J. Appl. Phys. **57**, 3386–3491 (1985)], and the 1985 International Conference on Magnetism [J. Appl. Phys. (to be published)]. The phenomenon is so ubiquitous that it would not be possible to list only a few papers without offending legions of unreferenced authors.

[2]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. **35**, 1792 (1975).

[3]M. Gabay and G. Toulouse, Phys. Rev. Lett. **47**, 201 (1981).

[4]More recent work indicates that replica-symmetry breaking also occurs as one goes from the F to the $M1$ phase. However, it is generally believed that the most important irreversibility is that associated with longitudinal ordering, as described in Ref. 3. See D. M. Cragg, D. Sherrington, and M. Gabay, Phys. Rev. Lett. **49**, 158 (1982).

[5]A particularly comprehensive study has been made by I. Mirabeau *et al.*, in Proceedings of the 1985 International Conference on Magnetism [J. Appl. Phys. (to be published)], paper 4Pj5.

[6]Other short-ranged models producing noncollinearity have also been studied: J. Villain, J. Phys. C **10**, 4793 (1977), and **11**, 745 (1978); W. Kinzel and K. Binder, Phys. Rev. B **24**, 2701 (1981); M. W. Dunlop and D. Sherrington, J. Phys. C **18**, 1465 (1985). Also see M. Cieplak and M. Z. Cieplak, J. Phys. C **18**, 1481 (1985), for zero-temperature simulation studies of reentrance in the Edwards-Anderson model.

[7]See R. E. Walstedt, in *Heidelberg Colloquium on Spin Glasses,* edited by J. L. van Hemmen and I. Morgenstern, Lecture Notes in Physics Vol. 192 (Springer-Verlag, Berlin, 1983), p. 177.

[8]G. Toulouse, Commun. Phys. **2**, 115 (1977).

[9]We neglect the possibility that an infinitely large version of the present model may need anisotropy to stabilize it. Moreover, mean-field theory should be a reliable guide to its solution, so long as critical phenomena are not discussed.

[10]C. M. Soukoulis, K. Levin, and G. S. Grest, Phys. Rev. B **29**, 1495 (1983).

[11]The small spread at $T=0.6T_C^{(0)}$, which corresponds to the $M1$ phase, provides support for weak transverse irreversibility. See Ref. 5.