Classical Spin-Glass Model

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A simple model of a classical spin-glass with weakly correlated disorder is presented. It includes both randomness and frustration, and is exactly soluble, but its solution can be obtained without replicas. Among the several phases of the model a mixed phase is found where spin-glass and ferromagnetism coexist. In addition the characteristic S shape of the spin-glass magnetization is reproduced.

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A spin-glass is a disordered, magnetic system with a well-defined freezing temperature T_f such that for $T<T_f$ the magnetic moments are frozen in random orientations without a conventional long-range order.[1] At the moment there is some, at least experimental, consensus[2] that one has a *phase transition* at $T=T_f$. In this Letter I aim to provide a soluble model to describe the spin-glass transition and the spin-glass phase.

To model a typical spin-glass like *Au*Fe the following facts must be considered. First, the concentration of the magnetic moments (Fe) in the metallic host (*Au*) is rather low, so that the distance between them is fairly large, and their locations are only weakly correlated. Second, the spins interact mainly indirectly, via the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction whose characteristics are (a) a long range, and (b) a strongly oscillating sign (as a function of the distance). Finally, there may also be a direct, ferromagnetic coupling.

One may assume that the spins are on a regular lattice and take the interaction as random.[3] The simplest assumption, a $\pm J$ or Gaussian nearest-neighbor interaction, is unlikely to produce a phase transition at positive temperatures.[4] An *infinite*-range Gaussian interaction,[5] though intuitively reasonable, does not yet allow a simple analytical treatment—in spite of considerable efforts[6] which, however, did show a spin-glass phase transition and pointed out the possibility of a "mixed" phase. I therefore propose a new *Ansatz*, which is consistent with the randomly oscillating, long-range, RKKY interaction.

We start with the Hamiltonian

$$\mathcal{H}_N = -\frac{J_0}{N}\sum_{(i,j)} S(i)S(j) - \sum_{(i,j)} J_{ij}S(i)S(j) - h\sum_i S(i). \tag{1}$$

This describes N Ising spins interacting with an external magnetic field h, and with each other in pairs (i,j). A direct, ferromagnetic coupling has been incorporated via J_0. The J_{ij} contain the randomness,

$$J_{ij} = (J/N)(\xi_i\eta_j + \xi_j\eta_i), \tag{2}$$

where the ξ_i's and η_j's are independent, identically distributed random variables with mean zero and, say, variance one. It can be shown that the J_{ij} are weakly correlated. Here I will outline a new method to calculate the free energy, discuss the phase diagram, and show that about half of the spins belong to a fully frustrated[7] configuration.

The free energy $f(\beta)$ is given by

$$-\beta f(\beta) = \lim_{N\to\infty} N^{-1}\ln \mathrm{Tr}\exp(-\beta\mathcal{H}_N). \tag{3}$$

The trace is a finite sum over all spin configurations, and the ξ_i's and η_j's have fixed values, randomly chosen according to their distribution. We put

$$m_N = \frac{1}{N}\sum_{i=1}^N S(i), \quad q_{1N} = \frac{1}{N}\sum_{i=1}^N \xi_i S(i), \quad q_{2N} = \frac{1}{N}\sum_{j=1}^N \eta_j S(j), \tag{4}$$

and rewrite the Hamiltonian $\mathcal{H}_N$,

$$-\beta\mathcal{H}_N = N(\tfrac{1}{2}K_0 m_N^2 + Kq_{1N}q_{2N} + Hm_N) \equiv NQ(\vec{m}) \tag{5}$$

with $\beta J_0 = K_0$, $\beta J = K$, and $\beta h = H$. Instead of the N spins we will use the three components of the vector $\vec{m} = (m_N, q_{1N}, q_{2N})$ as summation variables in the trace and, hence, need something like a Jacobian. To see how this may be accomplished we must make a small detour.

Suppose we have a sequence of independent, identically distributed stochastic variables σ_i

with mean zero and finite variance. As $N \to \infty$,

$$\underline{S}_N \equiv \frac{1}{N}\sum_{i=1}^{N} \sigma_i \to 0 \tag{6}$$

with probability one.[8] The event $\{\underline{S}_N \geq \epsilon\}$ with $\epsilon > 0$ is called a *large deviation* since it becomes more and more improbable as $N \to \infty$. We wish to estimate its probability $\text{Prob}\{\underline{S}_N \geq \epsilon\}$. To this end we introduce two functions,

$$c(t) = \ln\langle \exp(t\sigma)\rangle , \tag{7}$$

and

$$c^*(m) = \sup_{-\infty<t<+\infty} \{mt - c(t)\}. \tag{8}$$

The function $c(t)$ is convex and so is its *Legendre transform* $c^*(m)$.[9] We have[10]

$$\lim_{N\to\infty} N^{-1} \ln \text{Prob}\{\underline{S}_N \geq \epsilon\} = -c^*(\epsilon), \tag{9}$$

where $c^*(m) \geq 0$, with equality if and only if $m = 0$. For the events $\{\underline{S}_N \leq \epsilon\}$ with $\epsilon < 0$ an analogous formula holds. The limit $N \to \infty$ is essential in obtaining Eq. (9), which suggests that as $N \to \infty$

$$\text{Prob}\{m \leq \underline{S}_N \leq m + dm\} \sim \exp\{-Nc^*(m)\}\, dm. \tag{10}$$

We now return to the original problem.

Consider the Ising spins as independent stochastic variables which assume the values ± 1 with equal probability. Given N we divide the trace by 2^N in order to get a *normalized* trace $E_S\{X\} = 2^{-N}\,\text{Tr}\{X\}$, a spin-configurational average of X with $E_S\{1\} = 1$. Let $\vec{W}_N = (Nm_N, Nq_{1N}, Nq_{2N})$ and define

$$c(\vec{t}) = \lim_{N\to\infty} N^{-1} \ln E_S\{\exp(\vec{t}\cdot\vec{W}_N)\}. \tag{11}$$

For Ising spins (there is an analog for n-vector models) we easily obtain[8]

$$c(\vec{t}) = \langle \ln[\cosh(t_1 + t_2\xi + t_3\eta)]\rangle , \tag{12}$$

for almost every random configuration of ξ_i's and η_j's. The average in Eq. (12) is taken with respect to one ξ and one η. The function $c(\vec{t})$ is convex and so is its Legendre transform[9] $c^*(\vec{m})$. Using Eq. (5) and a slight generalization of Eq. (10) we then find, as $N \to \infty$,

$$E_S\{\exp(-\beta\mathcal{H}_N)\} \sim \int_{R_3} d^3m \exp N\{Q(\vec{m}) - c^*(\vec{m})\}, \tag{13}$$

and thus

$$-\beta f(\beta) = \max_{\vec{m}}\{Q(\vec{m}) - c^*(\vec{m})\}, \tag{14}$$

apart from a trivial ln2, which comes from the normalization of the trace.

The maxiumum in (14) is realized for a certain $\vec{m} = (m, q_1, q_2)$ and the negative of $c^*(\vec{m})$ is the mean entropy. Using the convexity of $c^*(\vec{m})$ one shows that $q_1 = q_2 \equiv q$ maximizes the free energy functional in (14). The remaining order parameters m and q satisfy the equations

$$m = \langle \tanh\{K_0 m + H + Kq(\xi+\eta)\}\rangle , \tag{15a}$$

$$q = \langle \tanh\{K_0 m + H + Kq(\xi+\eta)\}(\xi+\eta)/2\rangle . \tag{15b}$$

Putting $H = 0$ we quickly recognize three phases as special solutions of (15). The trivial solution $m = q = 0$ represents a paramagnet (P). If $q = 0$ and $m \neq 0$, we have a ferromagnet (F), and when $m = 0$ and $q \neq 0$, a spin-glass phase (SG) appears. But what about a *mixed* phase? If there were a mixed phase (II), both m and q would have to be nonzero. To see how this might occur we take ξ and η as ± 1 with equal probability; cf. Fig. 1.

The N lattice points can be divided into two disjoint subsets according to the sign of $\xi_i\eta_i$. We call the points with $\xi_i\eta_i = +1$ *blue* and the remaining ones, where $\xi_i\eta_i = -1$, *red*. Since

$$\{\xi_i\eta_j + \xi_j\eta_i\} = \xi_i\eta_j\{1 + \xi_i\eta_i\xi_j\eta_j\} = 0,$$

whenever i and j have a different color, the random interaction only connects points of the same color. The ferromagnetic interaction is "color blind." Which one wins depends on $\alpha = J_0/J$ and $K^{-1} = T/J$.

If the ferromagnetic interaction is absent ($J_0 = 0$), the system breaks up into two decoupled subsystems, blue and red, which both contain about $\frac{1}{2}N$ points. The Mattis[12] transformation $S(i) \to \xi_i S(i)$ (the model is classical!) transforms

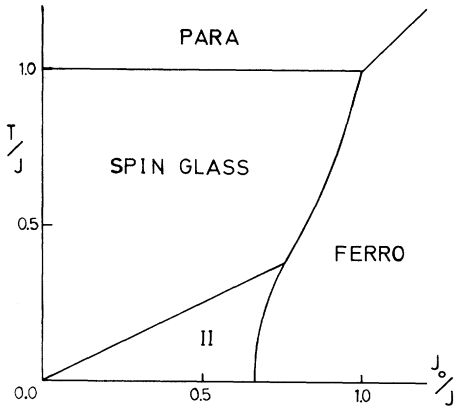

FIG. 1. Phase diagram for ξ and $\eta = \pm 1$ with equal probability. II is the mixed phase. Note that (1, 1) is a triple point (Ref. 11). There is no external field.

blue into ferromagnetic, and red into antiferromagnetic. At sufficiently low temperatures ($K^{-1}<1$) blue will thus[13] be ordered, whereas red remains disordered (uncorrelated) down to $T=0$ because of the large amount of frustration—all the simple closed loops (i.e., all triangles) are frustrated. We now undo the Mattis transformation, and note in passing that we have also shown that about half of the original lattice is fully frustrated as a Mattis transformation is frustration preserving.[14] Blue has spin-glass order with order parameter q_{1N}, whereas red remains uncorrelated.

If $0<\alpha\ll 1$ and $K^{-1}<1$, the random interaction dominates and, as before, we find blue in a spin-glass phase. Whatever its strength, the random interaction can never order red. Nevertheless ferromagnetic order will appear on red *provided* $\beta J_0=K_0>2$, because in this way red gains extra energy via the J_0 term; i.e., we obtain a mixed phase. Note that the entropy does not yet play a significant role. At $\beta J_0\approx 2$ red has a ferromagnetic phase transition and becomes uncorrelated above this line. We enter the pure SG phase, which will persist up to $K^{-1}=1$.

On the α axis of Fig. 1 we find a II-F transition at $\alpha=\frac{2}{3}$. Here $T=0$, and so an energy argument suffices: With $S(i)=\xi_i$ on blue and $S(i)=+1$ on red the ground-state energy is $-\frac{1}{8}J_0-\frac{1}{4}J$, whereas it is $-\frac{1}{2}J_0$ when all the points are ferromagnetic. If $\alpha>1$, the J_0 term should dominate, whatever the temperature. This is indeed the case.

Analytically one has to solve the fixed-point equations (15) and choose the solution that maximizes (14). The mixed phase appears, through a secondary bifurcation, as a twig on the main SG branch. It is acceptable if $\alpha\lesssim\frac{2}{3}$.

The spin-glass magnetization $m(h)$ exhibits a distinct S-shape character (see Fig. 2). It has a field-induced transition[15] to a state of higher magnetization above a certain threshold field, whose value h_t increases as the temperature is lower. The initial susceptibility χ_0 has a cusp at the P-SG boundary and a divergence at the SG-II boundary; both lines are critical. The specific heat has its main singularity (a jump) at the P-SG boundary if $0<\alpha<1$. Because of the ferromagnetic feedback, the model has an essentially unique ground state with zero entropy. The last three observations follow easily from the blue and red picture.

In summary, I have obtained a simple spin-glass model with frustration. It is easy to handle, and reproduces many experimental features of a spin-glass[2,11,15] quite well. Probability distributions more general than ξ and η equal to ± 1 will be considered elsewhere.[16]

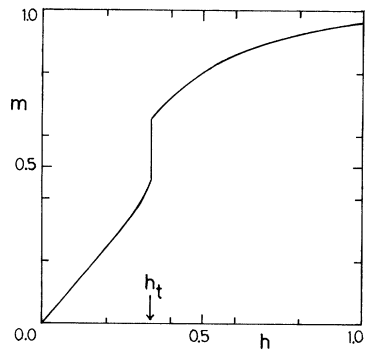

FIG. 2. The spin-glass magnetization m as a function of the external field h, with $\alpha=0.2$ and $K^{-1}=0.6$ where the units are such that $J=1$. At h_t there is a field-induced transition to a state of higher magnetization. The magnetization is convex on the left of h_t and concave on the right; i.e., h_t acts as a point of inflection.

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New Universality Class for Kinetic Gelation

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Percolation is often used as a model for critical behavior of the sol-gel transition. A three-dimensional kinetic model for gelation, similar to that of Manneville and de Seze, is investigated with a Monte Carlo method. By looking at the ratios of the critical amplitudes of the "susceptibility" (weight-average degree of polymerization) below and above the percolation threshold, clear evidence is found that this model is neither in the universality class of standard percolation nor in that of classical (Flory-Stockmayer) theory.

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The sol-gel transition[1] is apparently more complicated than predicted by the current theories. This was first shown in 1976 when Stauffer[2] and de Gennes[3] suggested that percolation on a three-dimensional (3D) lattice is a good model for critical phenomena in gelation, since the classical theory of Flory[4] and Stockmayer[5] is identical to percolation on a Cayley tree. As in the case of coagulation[6] or some other kinetic percolation processes[7] there has been mounting criticism[1] that standard percolation does not consider the growth process of the gelation, i.e., the kinetics. It has been suggested that a kinetic gelation model might not even be in the universality class of standard percolation.[8]

In this Letter we present Monte Carlo evidence that indeed a rather realistic growth model for the sol-gel transition, similar to that of Manneville and de Seze,[8] is *not* in the same universality class as percolation.

We want to describe a model of gelation for the irreversible free-radical copolymerization process. Here the sol consists of small monomers and the gelation is initiated by radicals. The radicals saturate, opening up a double bond of a monomer and leaving one bond in the monomer unsaturated. This creates a new radical that continues the growth process. We use $L\times L\times L$ simple cubic lattices with periodic boundary conditions containing a concentration c_2 of bifunctional sites and a concentration $1-c_2$ of tetrafunctional sites. (A bifunctional or tetrafunctional site can have at most two or four occupied bonds incident, respectively.) The initialization is performed by randomly occupying a fraction c_1 of bonds. For simplicity, no adjacent bonds are allowed to be occupied. Chemically, an occupied bond means a broken double bond between carbon atoms. The two free ends of an occupied bond are the radicals or "active centers." Now, the growth process is performed as follows: We randomly choose an active center and an adjacent bond of this center. If the other end of this bond is not forbidden, i.e., is not a bifunctional or tetrafunctional site al-