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LETTER TO THE EDITOR

Ginzburg–Landau theory of the cluster glass phase

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Abstract. On the basis of a recent field theory for site-disordered spin glasses, a Ginzburg–Landau free energy is proposed to describe the low-temperature glassy phase(s) of site-disordered magnets. The prefactors of the cubic and dominant quartic terms change gradually along the transition line in the concentration–temperature phase diagram. Either of them may vanish at certain points (c_*, T_*), where new transition lines originate. The new phases are classified.

A simple mixture of a metallic host with a magnetic atom, such as $Au_{1-c}Fe_c$, is known to have a rather complicated phase diagram. According to Mydosh [1] the following phases occur at zero temperature. At very low c there is the Kondo regime of *independently* compensated spins in a metallic host. At somewhat larger concentrations there is a spin glass phase of *interacting single spins* with $T_g \propto c$. For $0.5\% < c < 10\%$ the spin glass experiences gradual *cluster formation*, while for $10\% < c < 16\%$ one has the *cluster glass phase*. For $c > 16\%$ one enters the percolated *ferromagnetic phase*, which also partly behaves as a cluster glass.

The Kondo regime and the low-concentration spin glass phase are relatively well understood. The latter is described by an Edwards–Anderson model with RKKY interactions. Its properties are obtained from a mean-field approach [2] and from numerical analysis, see e.g. [3]. Whether or not a thermodynamic phase transition occurs in zero field or even in non-zero field remains a topic of much controversy.

Although ferromagnetism by itself is well known, clustering properties of inhomogeneous ferromagnets are far from well understood. It is known that replica symmetry breaking may occur before the onset of ferromagnetism¶, possibly describing Griffiths singularities.

The situation for the clustering spin glass (with clusters containing up to five atoms) and the cluster glass (where as many as 2000 atoms may build a cluster; these clusters order in a glassy way) is less satisfactory. Little is known about these phases. There seems to be no experimental evidence that the given names correspond to thermodynamic phases that are significantly different from the spin glass phase. Nevertheless, the existence of new glassy phases is the main question we wish to investigate theoretically in this work.

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¶ In the approach of [2] this follows immediately from the onset of SG ($T_g \sim \sqrt{c}$) and ferromagnetic phases ($T_F \sim c$) at low c. For a detailed analysis in a related model, see [4(a)]. For RSB in renormalization group flows, see [4(b)].

Recently, one of us [2] formulated a field theory for site-disordered Ising systems. With the exception of the Kondo regime, this applies to the whole phase diagram of systems, such as those mentioned above. We thus consider a system with translationally invariant pair couplings $J(r-r')$ with a fraction c ($0 < c < 1$) of the lattice sites occupied at random. We restrict ourselves to the second-order cumulant expansion. This description is Gaussian in the magnetization fields, and equivalent to a variational ('Hartree') approximation. It is quite close to that of the Sherrington–Kirkpatrick (SK) model since it involves only the order parameters $q_{\alpha\beta}$ ($= \langle s_\alpha s_\beta \rangle$ for small c) and their conjugates $p_{\alpha\beta}$. The replicated free energy per spin reads

$$\beta F_n = \frac{1}{2c}\int \frac{\mathrm{d}^3k}{(2\pi)^3}\sum_\alpha \{\ln(1-c\beta\hat{J}(k)q)\}_{\alpha\alpha} + \frac{1}{2}\sum_{\alpha\beta} q_{\alpha\beta}p_{\alpha\beta} + \sum_{l=1}^{\infty}\gamma_l(1-\mathrm{tr}_s^{(l)}\exp X^{(l)}) \tag{1}$$

with $\gamma_l = (-c)^{l-1}/l(1-c)^l$ and $X^{(l)} = \beta H\sum_\alpha \sigma_\alpha + \frac{1}{2}\sum_{\alpha\beta} p_{\alpha\beta}\sigma_\alpha\sigma_\beta$, where $\sigma_\alpha = s_\alpha^{(1)} + \cdots + s_\alpha^{(l)}$ denote nl replicated spins, and $\mathrm{tr}_s^{(l)}$ denotes the sum over $s_\alpha^{(j)} = \pm 1$.

This expression is quite rich and embodies the effect of clustering. Indeed, by expanding the logarithm in powers of $q_{\alpha\neq\beta}$ one observes an effective coupling $\hat{J}_{\mathrm{eff}}(k) = \hat{J}(k)/(1-c\beta\hat{J}(k)q_d)$, due to the presence of the diagonal elements $q_{\alpha\alpha} \equiv q_d(c,T) < 1$. If $\hat{J}$ is peaked at some k, $\hat{J}_{\mathrm{eff}}(k)$ will be peaked much stronger, thus exhibiting clustering effects. When $\hat{J}(k) = J_0$ for $k_0 < k < k_1$, while vanishing elsewhere, one considers the long-range oscillating interaction $J_{\mathrm{eff}}(r) \sim (k_0\cos k_0 r - k_1\cos k_1 r)/r^2$ at large r. In the scaling limit $k_1 - k_0 \sim c \to 0$, the mean field becomes exact. Equation (1) then has as limit the Hopfield model and the SK model [2].

From equation (1) a Ginzburg–Landau (GL) free energy can be derived. Omitting the paramagnetic background, eliminating the q's and fluctuations of $p_d \equiv p_{\alpha\alpha}$, and denoting $p_{\alpha\beta}$ again by $q_{\alpha\beta}$, we end up with

$$\beta F_n = -\frac{h^2}{2}\sum_{\alpha\beta} q_{\alpha\beta} - \frac{\tau}{2}\sum_\alpha (q^2)_{\alpha\alpha} - \frac{w}{6}\sum_\alpha (q^3)_{\alpha\alpha} - \frac{y_1}{8}\sum_{\alpha\beta} q_{\alpha\beta}^4 - \frac{y_2}{8}\sum_{\alpha\beta\gamma} q_{\alpha\beta}^2 q_{\alpha\gamma}^2 - \frac{y_3}{8}\sum_\alpha (q^4)_{\alpha\alpha} \tag{2}$$

where now $q_{\alpha\alpha} = 0$ and $h^2 = \beta^2 H^2 \mu_2$. The prefactor of the quadratic term, $\tau = (\mu_{22} - T^2/cJ_2)/2$, vanishes at the spin glass temperature $T_{\mathrm{g}}(c) \equiv \sqrt{cJ_2\mu_{22}}$. Furthermore, $w = \mu_{222} + T^3 J_3/(cJ_2^3)$, $y_1 = 3\mu_{2222}/2 + \mu_{44}/6 - \mu_{422}$, $y_3 = \mu_{2222} + T^4(J_2J_4 - 2J_3^2)/(cJ_2^5)$, and we have a similar expression for y_2. We introduced the moments of the effective coupling

$$J_l = \int \frac{\mathrm{d}^3k}{(2\pi)^3}[\hat{J}_{\mathrm{eff}}(k)]^l$$

and the spin moments

$$\mu_{k_1\ldots k_j} \equiv \sum_{l=1}^{\infty}\gamma_l \frac{m_{k_1}^{(l)}}{m_0^{(l)}}\cdots\frac{m_{k_j}^{(l)}}{m_0^{(l)}} \tag{3}$$

where $m_k^{(l)} = \mathrm{tr}_\sigma \sigma^k \exp(p_d\sigma^2/2)$ with $\sigma = s^{(1)} + \cdots + s^{(l)}$.

The paramagnetic behaviour is coded in the parameters p_d and q_d, that satisfy the coupled mean-field equations $p_d = \beta J_1$ and $q_d = \mu_2$. All information on clustering is contained in τ, w, and the y, and therefore in μ and J_l. In the limit $c \to 0$ μ goes to unity and for $T \sim \sqrt{c}$ the J_3 and J_4 terms vanish, so that one recovers the GL free energy of the SK model. The important factors then are $w = 1$, $y_1 = 2/3$, while the values of y_2 $(= -2)$ and y_3 $(= 1)$ are irrelevant. When following the transition line $T = T_{\mathrm{g}}(c)$ in the c–T phase diagram as a function of c, it is seen that the higher μ's are rapidly oscillating functions. For instance, if $J_3/J_2^{3/2} \approx J_4/J_2^2 \approx 0$, then y_1 changes sign at $c = 2.7\%$ and at $c = 4.3\%$, while w becomes negative at 6.7%.

Based on these observations, we are led to assume that the relevant physics near the phase transition(s) is still contained in the GL free energy (2). However, there is no reason to assume that w and y_1 will always be positive. (A sign change of y_1 also occurs in a Potts glass [5].) Given the type of the lattice and the values of the spin–spin couplings, the c–T phase diagram may exhibit a limited number of special points (c_*, T_*) where either w or y_1 vanishes, and new phase boundaries originate.

When the $q_{\alpha\beta}$ are expressed in the Parisi order parameter function $q(x)$, one obtains the following free energy:

$$\beta F = \int_0^1 \mathrm{d}x \left\{ \frac{h^2}{2} q(x) + \frac{\tau}{2} q^2(x) - \frac{w}{3} q(x) T(x) + \frac{y_1}{8} q^4(x) - \frac{y_2 + y_3}{8} q^2(x) \int_0^1 \mathrm{d}y\, q^2(y) + \frac{y_3}{2} T^2(x) \right\} \tag{4}$$

with $T(x) = xq^2(x)/2 + q(x) \int_x^1 \mathrm{d}y\, q(y) + \int_0^x \mathrm{d}y\, q^2(y)/2$.

We first investigate the region where w goes through zero $(-1 \ll w \ll 1)$ while $y_1 > 0$ is fixed. In figure 1 we depict a fictitious phase diagram with such a situation. On the side where $w > 0$ one has the well known spin glass solution of Parisi type, as depicted in figure 2(a). The interesting domain is $w < 0$ and $\tau \sim w^2$, since y_2 and y_3 become relevant. In order to find an acceptable solution we assume that $y_3 < -y_1$ so that $\alpha \equiv \sqrt{-y_1/y_3} < 1$. At $h = 0$ the spin glass order parameter function

$$q(x) = \frac{w\sqrt{y_1 + y_3 x_1^2}}{3(y_1 + y_3 x_1)} \frac{x}{\sqrt{y_1 + y_3 x^2}} \tag{5}$$

has plateau value $q_1 = q(x_1)$, determined by

$$\tau = wq_1 - \tfrac{3}{2}(y_1 + y_3) q_1^2 + \tfrac{1}{2} y_2 (1 - x_1) q_1^2 + \tfrac{1}{2} y_2 I_2 \tag{6}$$

where $I_2 = \int_0^{x_1} \mathrm{d}y\, q^2(y)$. The solution is physically acceptable as soon as y_2 exceeds a certain bound and exists for parameters such that x_1 ranges from $x_1 = 0$ up to $x_1 = \alpha$. For (c, T) such that $x_1 \to \alpha$ the solution squeezes and becomes a one-step replica symmetry breaking (1RSB) solution with lower plateau at $q_0 = 0$ (in zero field), see figure 2(b).

For $w < 0$, 1RSB solutions are present in a whole domain. In general, a 1RSB occurs in two shapes, static and dynamic. The static case describes physics on exponentially large time scales where the system can overcome the free energy barriers between pure states. Here one maximizes the free energy with respect to x_1, which yields the plateau value

$$q_1^{\mathrm{g}} = \frac{wx_1}{\frac{3}{2} y_1 + 3y_3 x_1 (1 - \frac{1}{2} x_1)}. \tag{7}$$

It sets in from $x_1 = 1$ as a first-order phase transition without latent heat at temperature

$$T_{\mathrm{g}}^{\mathrm{1RSB}} = T_{\mathrm{g}}(c) - \tau_{\mathrm{g}} \equiv T_{\mathrm{g}}(c) + \frac{w^2}{9|y_1 + y_3|}. \tag{8}$$

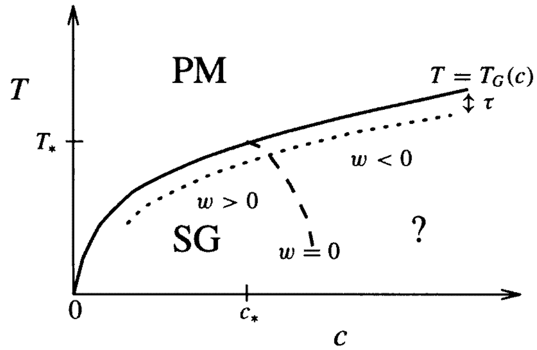

Figure 1. c–T phase diagram for a fictitious system with a line $w(c, T) = 0$. PM = paramagnet; SG + spin glass.

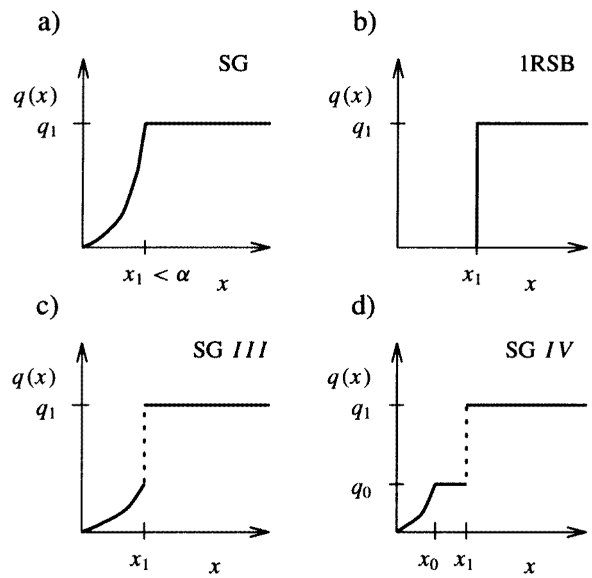

Figure 2. Shapes of the spin glass order parameter funcion. (a) standard form for infinite-order replica symmetry breaking; (b) one-step replica symmetry breaking solution; (c) the discontinuous SG III function; (d) the SG IV function.

Whereas the transition from paramagnet to spin glass has a continuous specific heat, the analogy to real glasses makes us expect that (also beyond the mean field) the specific heat jumps downwards at the transition PM $\rightarrow$ 1RSB. Both the SG and 1RSB phases will exhibit a difference between field-cooled and zero-field-cooled susceptibilities.

In the mean field the metastable states have infinite lifetime. Therefore, the dynamical 1RSB equations lead to a sharp phase transition at temperature $T_c > T_g$ [6–8]. The thermodynamics of this dynamical transition is uncommon [9]. The entropy of the frozen state is much below the paramagnetic one. A crucial role is played by the complexity (configurational entropy), which is extensive. This scenario explains thermodynamically why the dynamical glass transition takes place: the system just goes to the available state with lowest free energy [10]. Beyond the mean field the dynamical aspects are reflected in the dependence on the cooling rate.

For a dynamical 1RSB phase the q_1-plateau is marginally stable and equal to

$$q_1^{\mathrm{c}} = \frac{w x_1}{2y_1 + y_3 x_1 (3 - x_1)}. \tag{9}$$

This dynamical solution sets in at a larger temperature

$$T_{\mathrm{c}}^{\mathrm{1RSB}} = T_{\mathrm{g}}(c) - \tau_{\mathrm{c}} \equiv T_{\mathrm{g}}(c) + \frac{w^2}{8|y_1 + y_3|}. \tag{10}$$

Both the static and dynamical solutions exist down to

$$T_{\mathrm{sg}}(w) = T_{\mathrm{g}}(c) - \tau_{\mathrm{sg}} \equiv T_{\mathrm{g}}(c) - \frac{w^2}{6y_3}\left(1 + \frac{y_2}{3y_3(1-\alpha)}\right). \tag{11}$$

This is exactly the line where, coming from positive w, the SG solution gets squeezed into a 1RSB solution. The full phase diagram is depicted in figure 3.

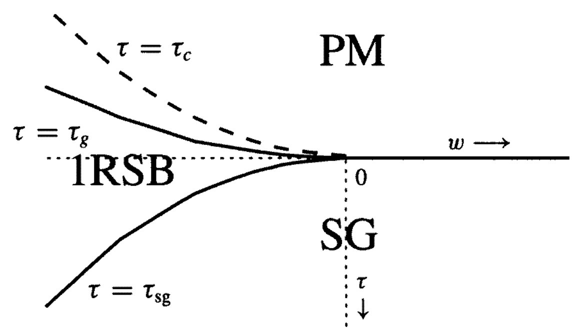

Figure 3. τ–w phase diagram for a system with $y_1 > 0$, $y_s < -y_1$, and y_2 sufficiently positive; with w increasing from right to left it may appear in figure 1 around the point (c_*, T_*). The full (dashed) curves are static (dynamical) transition lines.

Next we consider the situation where y_1 goes through zero, while $w > 0$ is fixed. (In the case when $w < 0$ the system will already have undergone a non-perturbative first-order transition at some negative τ.) One now expects a transition from a spin glass phase ($y_1 > 0$) to a replica symmetric or Edwards–Anderson (EA) phase ($y_1 < 0$). In the EA phase there is no difference between field-cooled and zero-field-cooled susceptibility.

As it was the case for Parisi's solution of the SK model, the values of y_2 and y_3 are now irrelevant. However, higher-order replica symmetry breaking terms will become relevant. All fifth-order terms have been considered for the above model. The most dangerous one is $-(y_5/8)\sum_{\alpha\beta} q_{\alpha\beta}^3 (q^2)_{\alpha\beta}$ with $y_5 = 6\mu_{22222} - 4\mu_{4222} + \frac{2}{3}\mu_{442}$. (For SK, $y_5 = 8/3$). We can absorb this term in our previous free energy using the saddle-point equation $(q^2)_{\alpha\beta} \approx -2\tau q_{\alpha\beta}/w$, which amounts to replacing y_1 by $\tilde{y}_1 = y_1 - 2\tau y_5/w$. The most dangerous sixth-order term is $-(y_6/6)\sum_{\alpha\beta} q_{\alpha\beta}^6$, where $y_6 = \frac{15}{4}\mu_{222222} - \frac{15}{4}\mu_{42222} + \frac{15}{16}\mu_{4422} + \frac{1}{4}\mu_{6222} - \frac{1}{8}\mu_{642} + \frac{1}{240}\mu_{66}$ ($y_6 = 16/15$ for SK).

The interesting region is where the $q_{\alpha\beta}^4$ term is of same order of magnitude as the $q_{\alpha\beta}^6$ term. This occurs when $y_4 \equiv \tilde{y}_1 w^2/2\tau^2$ is of order unity. At fixed small positive τ we now follow the system by changing y_4. We thus vary c and T over a line at fixed distance τ to the critical line. This is indicated by the dotted curve in figure (1), where w should now read y_1. For $y_4 \gg 1$ we will have a standard SG, while for $y_4 \ll -1$ there is the EA phase.

When $y_6 > 0$ is fixed, we find that in between the SG phase and the EA phase there is a SG phase with $q_0 > 0$, although there is no external field. Coming from the SG phase, q_0 starts to become non-zero at $y_4 = 0^-$. For $y_4 \to -2y_6$ replica symmetry is restored since q_0 approaches q_1. The y_1–τ phase diagram for the case $y_6 > 0$ is shown in figure 4. As it is the case with the AT line in a field, the transition EA $\to$ SG ($q_0 \neq 0$) may very well be smeared beyond the mean field.

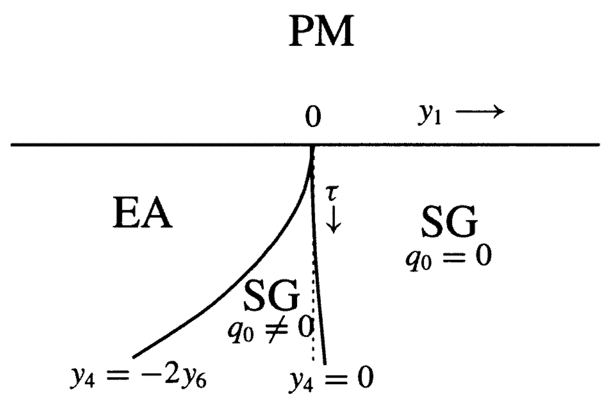

Figure 4. y_1–τ phase diagram for $w > 0$, $y_6 > 0$. The function $q(x)$ in the SG phase is drawn in figure 2(a) for the case $q_0 = 0$. In the EA phase $q(x)$ is constant (no RSB).

When $y_6 < 0$ we find a new, discontinuous order parameter function, that we call SG III: $q(x) = q_c(x)$ for $x \leqslant x_1$, while $q(x) = q_1 > q_c(x_1)$ for $x > x_1$, see figure 2(c). As for static 1RSB solutions, the plateau has stable fluctuations. Coming from the EA phase, SG III sets in with $x_1 = 0$, leading to irreversibility. With respect to the EA phase, the SG III phase has a smaller replica free energy with a discontinuous slope. There occurs a static first-order transition without latent heat but with a discontinuity in the specific heat, as usual for glasses.

At $y_4 = 10|y_6|$ the discontinuity of $q(x)$ disappears and the standard SG solution takes over, see figure 5.

There are also other solutions with free energy between the ones of the EA and the SG III states. At $y_4 = |y_6|$ a 1RSB solution with marginal lower plateau occurs, as in a Potts glass [9]. Now the breakpoint sets in from $x_1 = 0$. This 1RSB solution becomes unstable at $y_4 = 3|y_6|$, where the q_0 plateau is lifted and a foot grows near $x = 0$. We call this the SG IV solution, see figure 2(d). Like the SG III, it exists up to $y_4 = 10|y_6|$, where the SG IV discontinuity disappears and it matches the standard SG solution (see figure 2(a)). In analogy with the marginal 1RSB solution, we anticipate that this 1RSB-SG IV traject is the one that occurs in dynamics.

Also in the standard region where w and y_1 are still positive some clustering effects occur. Consider the slope of the field-cooled susceptibility $\chi_{\mathrm{FC}} = \beta(1 - \int_0^1 \mathrm{d}x\, q(x))$. At T_{g}^- one has $\mathrm{d}\chi_{\mathrm{FC}}/\mathrm{d}T = -T_{\mathrm{g}}^{-2} + (wT_{\mathrm{g}})^{-1}\,\mathrm{d}\tau/\mathrm{d}T$. In mean-field models with ∞RSB χ_{FC} is usually constant below T_{g}, so these two terms cancel. There does not seem to be a general reason for this. Experimentally, the values in the SG phase are usually lower than at T_{g}.

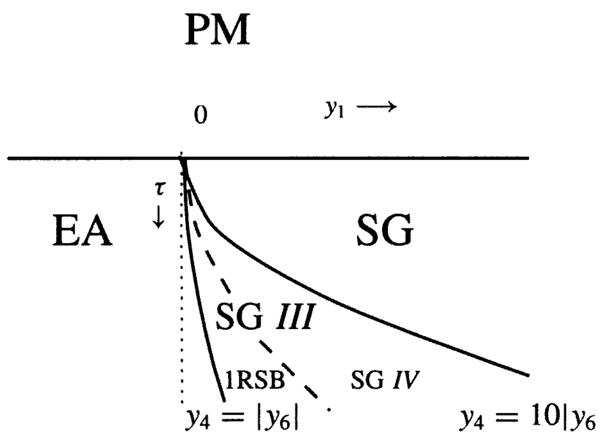

Figure 5. y_1–τ phase diagram for $w > 0$, $y_6 > 0$. In the SG III phase $q(x)$ is as in figure 2(c). Dynamicallly this phase splits up into a 1RSB phase and a SG IV phase. see figure 2(b) and (d).

However, in the mechanically milled amorphous Co_2Ge spin glass of Zhou and Bakker, that has about 67% of magnetic atoms, one expects large clustering effects. Indeed, χ_{FC} is monotonically decreasing with T [11]. Both these phenomena can be explained by our formula.

So far our results mainly concern the mean field. Whether or not fluctuations change them qualitatively is unknown.

In conclusion, we have proposed a Ginzburg–Landau free energy for site-disordered spin glasses. It is motivated that the prefactors of the cubic and quartic terms can have zeros. From these points new transition lines originate. We find spin glass phases of the Parisi type (∞RSB), with 1RSB, without RSB (EA phases), and of new types, the SG III and SG IV phases. For the latter phases the dynamics will be of a new nature.

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