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# Universality in glassy systems

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**Abstract.** The properties of a diluted spin-glass model are reviewed. This model interpolates between a spin glass and a frustrated lattice gas which displays glassy behaviour at high density.

Using the effective potential introduced by Franz and Parisi, we show in the mean-field formalism that its static properties are closely related to those of the p-spin-glass models. These models are strongly related to glass-forming liquids. In the mean-field formalism they exhibit in fact a dynamical transition described by the same equations as in the schematic mode-coupling theory.

Compared to the p-spin model, the frustrated lattice gas model is more appropriate for describing a supercooled liquid, being constituted essentially of diffusing particles. Numerically in three dimensions we in fact show that the frustrated lattice gas model exhibits qualitatively the properties of glass-forming liquids. Interestingly, the diffusion coefficient exhibits a sharp crossover from a power-law behaviour at low density to a Vogel–Fulcher or Arrhenius behaviour at high density, suggesting a crossover from mode-coupling behaviour to a hopping regime.

## 1. Introduction

There are many physical systems and models which exhibit a glass transition, and it is therefore extremely important to understand the problem of universality in glassy systems. It is known that a generalization of spin glasses, namely the p-spin-glass models, in the mean-field formalism exhibit general properties closely related to those of glass-forming liquids. Nevertheless they are microscopically quite different from liquids. In fact they are spin models interacting via p-body interactions with quenched disorder, and thus bear no resemblance to a liquid. The aim of this article is to introduce a lattice model which, beside having properties closely related to those of p-spin glasses in the mean-field formalism, is constituted from diffusing particles, and is therefore well suited for studying quantities like the diffusion coefficient, and the density–density correlation functions, that are usually important in the study of liquids.

## 2. Spin glasses and p-spin models

Spin glasses are well described by the Edwards–Anderson model, defined by the Hamiltonian

$$H = \sum_{ij} J_{ij} S_i S_j \quad (1)$$

where  $S_i = \pm 1$ , and the interactions  $J_{ij}$  are random quenched interactions distributed over positive and negative values, such that the average  $\langle J_{ij} \rangle = 0$ . Spin glasses exhibit a second-order transition at a temperature  $T = T_{SG}$ , where the Edwards–Anderson order parameter

vanishes as the temperature approaches  $T_{SG}$  from below. At this same temperature there is also a dynamical transition characterized by a diverging relaxation time.

A generalization of the Edwards–Anderson spin-glass model is the p-spin model [1]. In these models the spins interact via p-spin interactions. For example, for  $p = 3$ , the Hamiltonian is given by

$$H = \sum_{ijk} J_{ijk} S_i S_j S_k \quad (2)$$

where the interactions  $J_{ijk}$  are random quenched interactions distributed over positive and negative values, such that the average  $\langle J_{ijk} \rangle = 0$ .

This model exhibits, in the mean-field formalism, a thermodynamic first-order transition at a temperature  $T_s$ , and a dynamical transition at a higher temperature  $T_D > T_s$ , characterized by a diverging relaxation time. This dynamical transition is described by the same dynamical equations of the mode-coupling theory for supercooled liquids [2, 3]. Recently the interplay between statics and dynamics has been elucidated by Franz and Parisi [4], introducing the method of coupling two replicas of the system.

### 3. Coupling replicas

Consider two identical replicas  $A$  and  $B$  of the spin model (2), coupled by a ferromagnetic interaction:

$$\mathcal{H} = H^A + H^B - \epsilon \sum_i S_i^A S_i^B. \quad (3)$$

The overlap between the two replicas is given by

$$q = -\frac{\partial F}{\partial \epsilon} = \frac{1}{N} \sum_i S_i^A S_i^B \quad (4)$$

where  $F(\epsilon)$  is the free energy per site of the coupled replicas (3). The quantity  $q$  plays here the role of an order parameter. For high temperature  $q = 0$ , while for low temperature in the glassy phase  $q \neq 0$ .

The need for coupling two replicas comes from the fact that we must explicitly break the symmetry in order to find the order parameter. For an Ising ferromagnet, for example, the symmetry that is broken in the ordered phase is the simple up–down symmetry, so in that case we can couple the system to a magnetic field, and then let the field go to zero. Here we do not know *a priori* the symmetries which are broken in the glassy phase, so we couple the system to a copy of itself. In this way each of the two copies, say  $A$ , plays the role of a staggered field  $\epsilon S_i^A$  acting on the corresponding sites of the other replica  $B$ . It is clear that, for  $\epsilon > 0$ , the two replicas are constrained to be in the same ‘free-energy valley’, and this will be true also if we let  $\epsilon \rightarrow 0^+$ . The symmetry will then be explicitly broken and we will find an ‘order parameter’  $q$  different from zero.

The effective potential  $V(q)$  is the Legendre transform of  $F(\epsilon)$ :

$$V(q) = \max_{\epsilon} [F(\epsilon) + \epsilon q] \quad (5)$$

which has the property  $\partial V / \partial q = \epsilon$ . The equilibrium values of  $q$  corresponding to  $\epsilon = 0$  are therefore the minima of the effective potential  $V(q)$ .

Franz and Parisi [4] have calculated the effective potential for the p-spin model with  $p = 4$ . For high temperature they found one minimum corresponding to  $q = 0$ . As the temperature is lowered below  $T_D$ , a new secondary minimum at a value  $q > 0$  appears. At the temperature

$T_s$  the secondary minimum reaches the level of the minimum at  $q = 0$ , and a thermodynamic first-order transition appears.

Interestingly, the temperature  $T_D$  coincides with that of the dynamical transition, and the dynamic equations are the same as those predicted by the schematic MCT theory for supercooled liquids.

The dynamical interpretation of the static picture is that, in the mean-field formalism,  $T_D$  marks the onset of metastable states with infinitely long lifetime. When the two replicas are in the same metastable state, the value of  $q$  is higher than zero, while if they are in different states,  $q = 0$ . As different metastable states are characterized by the same internal energy and entropy, the difference between the values of  $V(q)$  for the two minima,  $\Delta V$ , can be interpreted as the configurational entropy  $\Sigma$  (also called the complexity, which is the logarithm of the number of metastable states) multiplied by  $T$ . As  $T$  approaches the static transition  $T_s$ ,  $\Delta V$  goes to zero and consequently the configurational entropy vanishes. This picture has also been verified for hard spheres using the hypernetted-chain approximation [5]. In 3D one expects this picture to hold also for supercooled liquids, except that the metastable state now has a finite lifetime, and therefore the relaxation time is expected to show an apparent power-law divergence at  $T_D$  corresponding to the prediction of the mode-coupling theory, with a crossover towards a Vogel–Fulcher behaviour as  $T \rightarrow T_s$ .

The p-spin models have elucidated very nicely the approximations involved in MCT theory, and the mechanism of the arrested dynamics which occurs at  $T_D$  based on the onset of metastable states. However, it is difficult to get microscopic insight into the molecular motion of a supercooled liquid as it approaches the glass transition. In fact the model is microscopically very different, and the interaction is rather unphysical since the spins interact via interactions involving three or more bodies. Moreover the generalization of the model in finite dimensions is not straightforward.

#### 4. The frustrated lattice gas model

To gain a better insight into the motion of the particles we want to discuss a simple model which combines features of spin glasses and lattice gas.

The concept of frustration in spin glasses can be quantitatively defined [6]. For example, in the nearest-neighbour  $\pm J$  Ising spin-glass model, a loop is frustrated when the spins cannot satisfy all of the interactions along it. It is easy to verify that a loop is frustrated if and only if the product of the signs of the interactions equals  $-1$ .

In glass-forming liquids frustration arises as a packing problem, and is formally less clearly defined. In systems without underlying crystalline order, frustration is typically generated by the geometrical shape of the molecules which prevents the formation of close-packed configurations at low temperature or high density. For systems with underlying crystalline order, frustration arises when the local arrangement of molecules kinetically prevents all of the molecules from reaching the crystalline state.

To model systems without underlying crystalline order, we consider the following spin-glass model diluted with lattice gas variables [7, 8]:

$$H = -J \sum_{\langle ij \rangle} (\epsilon_{ij} S_i S_j - 1) n_i n_j - \mu \sum_i n_i. \quad (6)$$

Here the occupancy variables  $n_i = 0, 1$  have the internal degrees of freedom  $S_i = \pm 1$ ,  $\epsilon_{ij} = \pm 1$  are quenched random interactions, and  $\mu$  is the chemical potential for the particles.

This model reproduces the  $\pm J$  Ising spin glass in the limit  $\mu \rightarrow \infty$  (all sites occupied,  $n_i \equiv 1$ ). We will consider here the other limit of large  $J$ . In this case the model describes

a frustrated lattice gas with properties recalling those of a ‘frustrated’ liquid. In fact the first term of Hamiltonian (6) implies that two nearest-neighbour sites can be freely occupied only if their spin variables satisfy the interaction—that is, if  $\epsilon_{ij}S_iS_j = 1$ —otherwise they feel a strong repulsion.

To make the connection with a liquid, we note that the internal degree of freedom  $S_i$  may represent for example internal orientation of a particle with non-symmetric shape. Two particles can be nearest neighbours only if the relative orientation is appropriate; otherwise they have to move apart. Since in a frustrated loop the spins cannot satisfy all of the interactions, in this model particle configurations in which a frustrated loop is fully occupied are not allowed. The frustrated loops in the model are the same as in the spin-glass model and correspond for the liquid to those loops which, due to geometrical hindrance, cannot be fully occupied by the particles. A connection between frustrated loops in spin glasses and defect lines in ordinary glasses has been discussed by Rivier [9].

The model (6) has been solved in the mean-field formalism [10] for any value of  $J$  and  $\mu$ . The phase diagram in the  $J$ - $\mu$  plane shows a line of critical points starting on the axis  $\mu = \infty$ , where the transition coincides with the Ising spin-glass transition. As  $\mu$  decreases the critical value  $J_c$  increases, and the transition is still in the same universality class of the Ising spin-glass transition, up to a value  $J_c^*$  above which the transition becomes first-order-like in the p-spin model [1]. And, just like in p-spin models, there appears another critical value  $\mu_D$  signalling the onset of a metastable phase.

In conclusion, for fixed high values of  $\mu$ , on varying  $J$  the model behaves as the Ising spin-glass model, while for large values of  $J$ , on varying  $\mu$  the model behaves as a p-spin model, characterized by a static first-order thermodynamic transition at low temperature, and a characteristic higher temperature  $T_D$  signalling the onset of metastable states.

We have calculated [11] the effective potential for large values of  $J$ , and we find the same behaviour as was found in the p-spin model and in the hard-spheres hypernetted approximation. In this case the two replicas have been coupled through a term  $\epsilon \sum_i n_i^A n_i^B$ , where  $n_i^A$  and  $n_i^B$  are the lattice gas variables of replicas  $A$  and  $B$ , respectively. Given the free energy per site  $F(\epsilon)$  of the coupled replicas, we consider the order parameter

$$q_d = -\frac{\partial F}{\partial \epsilon} = \frac{1}{N} \sum_i \langle n_i^A n_i^B \rangle. \quad (7)$$

The equilibrium values of  $q_d$  will be given by the minima of the effective potential

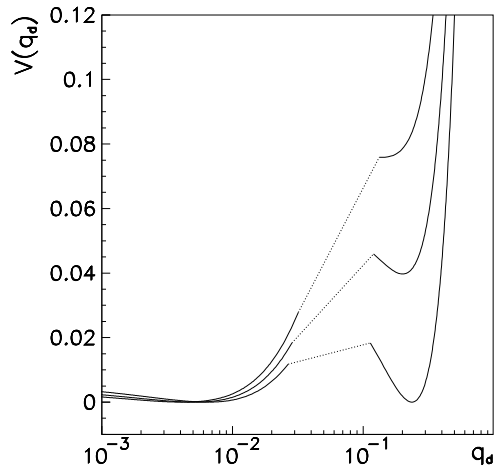
$$V(q_d) = \max_{\epsilon} [F(\epsilon) + \epsilon q_d].$$

In figure 1 we show  $V(q_d)$  for three values of  $\mu$ , which plays here the role of an inverse temperature. There is a value  $\mu_D$  at which a high- $q_d$  secondary minimum appears, together with an exponential number (given by  $e^{N\Sigma}$ , where  $\Sigma = \Delta V$  is the complexity) of metastable states. At a higher value  $\mu_s$  (at lower temperature) the secondary minimum reaches the level of the low- $q_d$  minimum, and there is a first-order transition to a glassy phase.

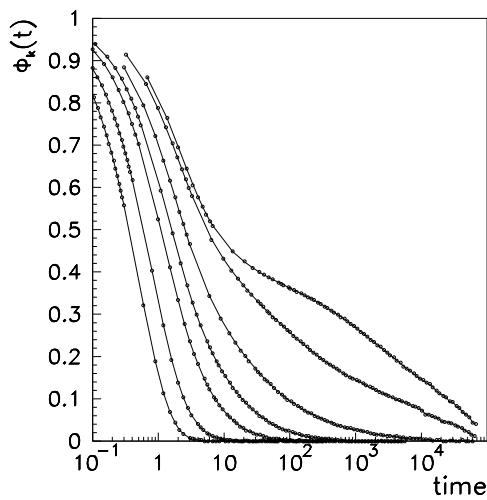
Since in the mean-field formalism the appearance of the metastable states is the mechanism for the dynamical transition, we expect in the frustrated lattice gas model, for large values of  $J$ , the same dynamical properties as those of the p-spin model, and therefore a dynamical transition described by the same equations as are found in the mode-coupling theory for simple liquids.

To find the behaviour of the model for large  $J$  in finite dimensions, Monte Carlo calculations have been performed on a cubic lattice [8, 12].

We thermalize the system (6) with some chemical potential  $\mu$ , and obtain some equilibrium density of particles (sites with  $n_i = 1$ ). Then we perform a diffusive dynamics, where particles can only move from a site to a nearest-neighbour site, but their total number is held fixed. In



**Figure 1.** The constrained potential  $V(q_d)$  in the mean-field model, for  $J = 4$  and (from top to bottom)  $\mu = -3.192, -3.135, -3.079$ .

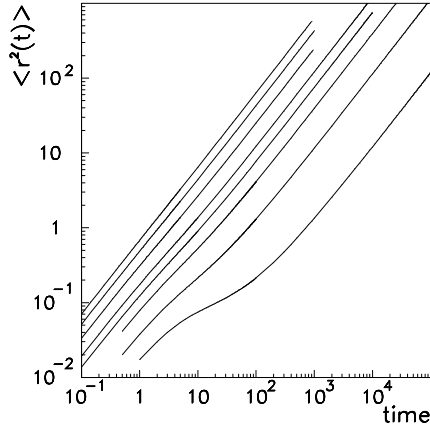


**Figure 2.** Relaxation functions of the density fluctuations, in the 3D model with  $L = 16$ , for densities  $\rho = 0.3, 0.4, 0.5, 0.55, 0.6, 0.65, 0.675$ . The wave vector is  $k = (\pi/a, 0, 0)$ , where  $a$  is the lattice spacing.

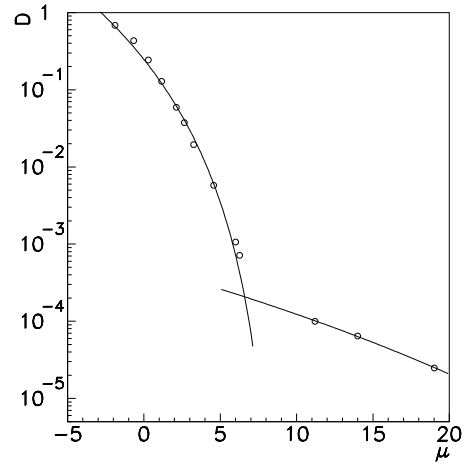
figure 2 we show the density–density autocorrelation functions for different values of the density. At very high density it is evident that there are different time regimes, as predicted by the *mode-coupling* theory of supercooled liquids, and observed both in some molecular dynamics simulations of glass-forming liquids [13, 14] and in some experimental measurements for glass-forming liquids. There is first a short-time relaxation, corresponding to relaxation inside non-frozen domains, surrounded by a frozen ‘cage’; finally there is a long-time regime ( $\alpha$ -relaxation) corresponding to structural relaxation and decay to equilibrium.

In figure 3 we show the mean square displacement of a tagged particle. For low density the curves show a linear behaviour, which corresponds to normal diffusion. For high densities, the mean square displacement shows a plateau followed by linear behaviour in the long-time regime. This feature can also be observed in molecular dynamics simulations [13, 14].

The plateaus observed, both in the relaxation functions and in the mean square displacements, are the sign of the glassy nature of the dynamic properties of the model, which compare well with those of glass-forming liquids.



**Figure 3.** The mean square displacement of a tagged particle as a function of time, in the 3D model with  $L = 16$ , for densities (from top to bottom)  $\rho = 0.2, 0.3, 0.4, 0.5, 0.55, 0.6, 0.65, 0.675$ .



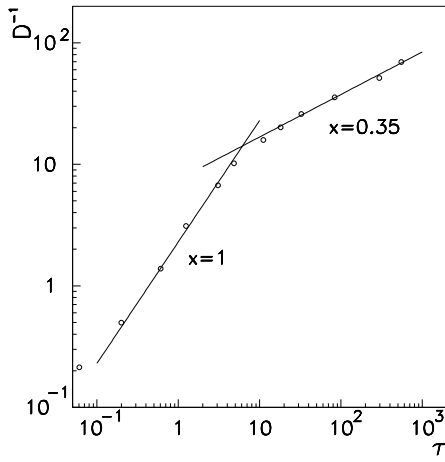
**Figure 4.** The diffusivity  $D$  as a function of the chemical potential  $\mu$ . While for  $\mu < \mu_c$  the diffusivity seems to follow a power law as a function of  $\mu$ , for  $\mu > \mu_c$  there is a sharp crossover to another regime.

We then plotted the diffusion coefficient  $D$  as a function of the density or chemical potential. The coefficient  $D$  is found as the slope of the linear long-time tail of the mean square displacement as a function of the time. From the data it is found that, in the case where  $J = \infty$ , the diffusion coefficient  $D$  vanishes at a value of the density  $\rho_c$ , with a power law as predicted by the MCT theory [12]. We have also simulated the model in the case where  $J = 5$ . The diffusivity behaves in the same way as in the case  $J = \infty$ , up to a density close to  $\rho_c$ , but it changes behaviour when  $\rho \simeq \rho_c$ —that is, when the chemical potential  $\mu$  is of the order of  $2J$ . In particular it does not vanish, but exhibits a neat crossover to another regime which can be fitted by a Vogel–Fulcher form (but also an Arrhenius behaviour cannot be excluded). In figure 4 we show the diffusivity  $D$  as a function of the chemical potential  $\mu$  for  $J = 5$ . The crossover in the behaviour of  $D$  may be due to activated processes that happen at a rate proportional to  $e^{-2J}$ . Interestingly, this change of behaviour is also found in experimental data [15]. The hopping process which is due to the softening of the potential seems to favour the crossover from mode-coupling or mean-field behaviour to a Vogel–Fulcher or Arrhenius behaviour.

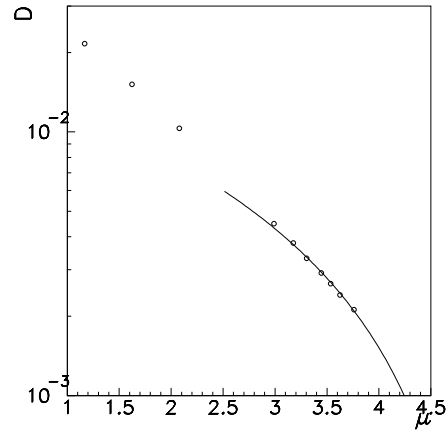
We have also calculated the relaxation time  $\tau$  from the integral of the density–density autocorrelation functions, and plotted  $D$  as a function of  $\tau$  (figure 5). For low density the data show that the SE relation  $D^{-1} \propto \tau$  holds, while for higher densities there is a sharp crossover to the fractional SE relation  $D^{-1} \propto \tau^x$ , with  $x \simeq 0.35$ , not much different from the value  $x \simeq 0.28$  found for supercooled orthoterphenyl [15]. We have to point out, anyway, that the experimental data refer to rotational and not to translational diffusion.

How do we explain the breakdown of the SE relation microscopically?

We have looked at the distribution of quasi-frozen clusters of particles in the system [12, 16], and we found that they grow as the temperature is lowered, until at some temperature  $T_g$  a spanning quasi-frozen cluster appears. The presence of a wide size distribution of quasi-frozen clusters suggests that the distribution of waiting times before the particles make a jump should also be wide, and reasonably is expected to become a power law with a cut-off at large time [17]. This has been verified in the 3D frustrated lattice gas model, where at



**Figure 5.** The breakdown of the Stokes–Einstein relation in the 3D model. The diffusivity  $D$  is plotted versus the integral autocorrelation times of the density fluctuations. For low densities (or high temperature) the data follow the SE relation  $D^{-1} \propto \tau$ , while for densities greater than  $\rho \simeq 0.5$  they follow the fractional SE relation  $D^{-1} \propto \tau^{0.35}$ .



**Figure 6.** The diffusion coefficient  $D$  as a function of the chemical potential  $\mu$  for the 3D model with annealed interactions. The solid curve is a power law.

low temperature we find a wide distribution of waiting times [12]. When there is only one characteristic time, both the diffusion coefficient  $D$  and the relaxation time  $\tau$  are related to it, and therefore they are linked in a simple way via the SE relation. When there is a wide distribution of waiting times, the diffusion coefficient depends on the form of the distribution at short time, while the relaxation time depends on the largest time which appears as the cut-off of the distribution, and as a consequence the Stokes–Einstein relation is broken [18].

In the model, as mentioned before, the frustration has been treated as quenched. On the other hand in glass-forming liquids frustration evolves over time. So a more realistic model should be obtained treating the variables from which frustration originates as annealed, and making  $\epsilon_{ij}$  evolve in the Hamiltonian (6) as a function of the local environment.

We have performed a diffusive dynamics, where at each step a variable  $\epsilon_{ij}$ , randomly selected, can change sign if both sites  $i$  and  $j$  are vacant. In figure 6 we have plotted the diffusion coefficient  $D$  as a function of the chemical potential. We find that the diffusion coefficient vanishes, at a value of the chemical potential  $\mu_c$ , following a power law.

These results suggest that, as the temperature lowers, the local environment changes so slowly that the variables  $\epsilon_{ij}$  can be treated more and more as frozen, playing the role of self-induced quenched variables.

## 5. Conclusions

We have reviewed the properties of the diluted spin-glass model, which interpolates between the spin glass, for low values of the ratio  $J/\mu$ , and the frustrated lattice gas model, for high  $J/\mu$ . We have studied the model in this latter limit, both in the mean-field formalism, and in finite dimensions by Monte Carlo simulations. Its properties in the mean-field formalism are closely related to those of p-spin-glass models, with a first-order transition at a temperature  $T_s$  in the parameter  $q_d$ , which is the ‘density overlap’  $\langle n_i^A n_i^B \rangle$  between two replicas, and the onset



of metastable states at a temperature  $T_D > T_s$ .

In finite dimensions the model has a behaviour very similar to that of structural glass-forming liquids, with two-step relaxations, an apparent power-law singularity in the diffusion coefficient, and a crossover at low temperatures to a ‘hopping regime’, and the breakdown of the Stokes–Einstein relation.

Work is in progress to clarify whether the mean-field picture of a discontinuous transition in the order parameter is still present in finite dimensions.

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