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LETTER TO THE EDITOR

The glassy transition of the frustrated Ising lattice gas

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Abstract. A three-dimensional (3D) spin-glass model diluted with lattice gas variables is investigated and compared with glass-forming liquids. The spin variables which play the role of internal degrees of freedom exhibit strong fluctuations. While a critical length associated to these fluctuations diverges at the glass transition, where the diffusion coefficient is found to vanish, the density fluctuations do not show any critical behaviour.

Recently there has been renewed interest in the study of glass-forming liquids [1, 2] and spin glasses [3, 4]. Both systems are characterized by an infinitely large relaxation time as the glass transition is approached, and exhibit a complex dynamical behaviour at low temperature (and also high density in the case of liquids) typically well above the glass transition. However, there are some basic differences. The spin-glass (SG) transition is characterized by a diverging length, associated with the spin pair correlation function and consequently exhibits a strong divergence in nonlinear susceptibility. In glass forming liquids there is no apparent diverging length and no critical density fluctuations and therefore no divergence in the compressibility. Nevertheless the theory of Adam and Gibbs [5] gives an intuitive physical explanation of the glass transition by assuming a diverging cooperative length, although the precise definition of such a length has always been lacking. Whether or not there exists a diverging length associated to the glass transition still remains an open problem.

In order to answer some of these questions and find a relation between spin glasses and glass forming liquids we introduce here a frustrated spin lattice gas model, with the spin playing the role of internal degrees of freedom, to describe the behaviour of a glass forming liquid.

In spin-glasses, frustration originates from the presence of *quenched* disorder in the distribution of spin interactions which cannot all be satisfied simultaneously [3, 4]. The origin of frustration in glass-forming liquids is generally different: when they have no 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 the molecules from reaching the crystalline state. So 'frustration' in glass-forming liquids *evolves in time*, but at a low enough temperature, or high enough density, the dynamics is so slow as to allow us to treat the variables from which frustration originates as quenched.

The model we introduce is a SG model diluted with lattice gas variables:

$$\beta\mathcal{H} = -J\sum_{\langle ij\rangle}(\epsilon_{ij}S_iS_j - 1)n_in_j - \mu\sum_i n_i. \tag{1}$$

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

This model reproduces the $\pm J$ Ising spin glass in the limit $\mu \to \infty$, because all sites are occupied ($n_i = 1$). We will consider here the other limit $J \to \infty$. 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 (1) implies that two nearest-neighbour sites can be occupied only if their spin variables satisfy the interaction, i.e. if $\epsilon_{ij} S_i S_j = 1$, else they feel an infinite repulsion ($J = \infty$). Therefore, the particles must either move apart or change the relative orientation of the spins. Since in a frustrated loop the spins cannot satisfy all the interactions, in this model particle configurations in which a frustrated loop is fully occupied are not allowed (a frustrated loop is a closed polygon where the product of all the signs ϵ_{ij} is -1 [3, 4]).

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 [6]. The frustrated loops in the model are the same as for the SG model and correspond in 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 [7].

In the limit $J \to \infty$, the partition function of model Hamiltonian (1) can be written as

$$Z = \sum_C \mathrm{e}^{\mu n(C)} 2^{N_c} \tag{2}$$

where the sum is over all particle configurations which do not contain a frustrated loop, $n(C)$ is the number of particles and $N_c(C)$ is the number of clusters of nearest-neighbours particles in the configuration C. Apart from the factor $2^{N_c(C)}$, the partition function (2) coincides with the site frustrated percolation which has been introduced to model glass-forming liquids [8, 9].

The difference between the site frustrated percolation model and the frustrated spin lattice gas model considered here is that in this case we can also study quantities related to the internal degrees of freedom. This is physically more satisfactory and also computationally simpler than the case where the spin variables do not appear.

In analogy with results on site and bond percolation [8] we expect for the Hamiltonian model (1) (for $J \to \infty$) that in the high-density region there is a SG transition at μ_g, in the same universality class as the Ising SG transition, namely the correlation length associated to the spin pair correlation function diverges with a critical exponent ν identical to the exponent of the Ising SG model. We also expect well before the glass transition, at $\mu_p < \mu_g$, a percolation transition associated to a percolating cluster of particles in the same universality class of random percolation. This percolation transition corresponds in the Ising SG model to the percolation of the Kasteleyn–Fortuin Coniglio–Klein clusters and it has been suggested that this percolation transition may be accompanied by stretched exponential relaxation, or other anomalies preceding the glass transition. This transition should not be confused with the dynamical transition found in mean-field calculations for some SG models.

In what follows we present the results from Monte Carlo (MC) simulations in 3D of the Hamiltonian model (1) (for $J \to \infty$) for static and dynamic behaviour.

We find that the particle diffusion coefficient vanishes at a dynamical critical point numerically consistent with the static SG critical point μ_g. We have also located the percolation transition μ_p and found that it signals the onset of stretched exponentials in the long-time behaviour of time correlation functions.

The model shows, moreover, that in spite of the smooth behaviour of the compressibility and specific heat, fluctuations associated to the 'internal degree of freedom' become critical at the glass transition. While in spin glasses this diverging length can be detected by measuring the nonlinear susceptibility, in glass-forming liquids the internal degrees of freedom are not easily probed in experiments and this length is not usually seen, unless probes, which couple to the internal degrees of freedom, are used. In fact there are some experiments where a critical length associated to orientational correlations has been detected [10], and recently evidence for a divergent dielectric susceptibility at the glass transition has been reported [12].

The picture which emerges from this model is that for low density the behaviour is the same as that of a normal fluid, with simple exponential relaxation. As the density increases a percolation threshold is reached above which the dynamics starts to deviate from simple exponential relaxation. In an intermediate region between μ_p and μ_g we find that strong 'non-equilibrium' phenomena set in, as a precursor to the true glass transition. In this region we observe that the particle mean-square displacement shows apparent anomalies. Moreover, in this region, the long-time asymptote of the density correlation functions, the Debye–Waller factors, become different from zero on our observation time scales and system sizes.

Most of the data presented below concern a system on a cubic lattice (analogous results are found in two dimensions) of linear size $L = 8, 16$ with periodic boundary conditions, fixing $J = 10$ and varying μ (i.e. the density). The data do not change for $J = 10\,000$, showing that we are in the limit of $J = \infty$. We used a standard MC *dynamic*, in which particle diffusion and spin are updated according to a spin-flip Metropolis algorithm. Our MC simulations were performed after successive thermalization of the systems at higher and higher μ for about 2×10^4 MC sweeps at each value of external parameters, and then by obtaining measurements for about 2×10^7 MC sweeps, for a given configuration of ϵ_{ij}. We have adopted the usual MC techniques (see [4]) to extract our results concerning thermodynamic properties, and our results are well established up to the deep glassy region ($\mu \sim 5.0$) above which they may only be indicative (due to extremely long CPU times).

The properties related to the *spin variables* are the same as found in standard spin glasses. We have located the glass transition by studying Binder's cumulant. The quantities

$$g_Q = \frac{1}{2}\left(3 - \frac{\langle Q^4\rangle}{\langle Q^2\rangle}\right)$$

with

$$Q = \left\langle \frac{1}{N}\sum_i s_i^a n_i^a s_i^b n_i^b \right\rangle$$

(a and b are two replicas, i.e. two systems of same size, interactions configuration and Hamiltonian parameters, which evolve in parallel with different number generators, and N is the total number of particles), for different sizes $L = 8, 16$, show an intersection zone in the region $\mu_g \geqslant 5.5$ (corresponding to a density $\rho_g \geqslant 0.67$). Slightly below μ_g, the SG susceptivity $\chi_{SG} = N(\langle Q^2\rangle - \langle Q\rangle^2)$ seems to diverge as shown in figure 1 (for $L = 8$). As in standard spin glasses the linear susceptivity and the specific heat do not present apparent anomalies in the glass transition region and the same happens to *'density' variables*. The density $\rho = \langle (1/L^3)\sum_i n_i\rangle$, with increasing μ, approaches a plateau limited by the value $\rho \sim 0.69$ (see figure 3, later). The compressibility $K = L^3(\langle \rho^2\rangle - \langle \rho\rangle^2)$, which is in strict correspondence with the specific heat C_s, is smooth at the SG transition (see figure 1).

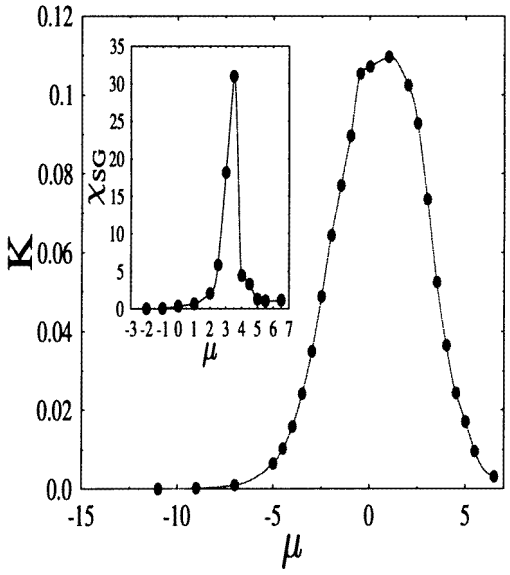

Figure 1. Compressibility K as a function of μ in a 3D system of size $L = 8$ at $J = 10$. The inset depicts the SG susceptibility $\chi_{\rm SG}$ for the same system. While $\chi_{\rm SG}$ presents a well defined peak, K is a smooth function of μ.

It is possible to define percolation clusters of particles in our model following Kasteleyn–Fortuin and Coniglio–Klein [13], with a bond probability given by [14]

$$p_{\rm KF} = (1 - {\rm e}^{-2Jn_in_j})\delta_{\epsilon_{ij}S_iS_j,1}.$$

These clusters in our context describe the physics of unfrustrated sets of sites in the systems, and in the $J \to \infty$ limit are simply the groups of neighbouring particles. They percolate at $\mu_{\rm p} = 0.75$, a value obtained by finite-size scaling with data from sizes $L = 8, 16$. This $\mu_{\rm p}$ corresponds to a density $\rho_{\rm p} = 0.38$, which is a little higher than the value of the standard 3D random percolation, $\rho_{\rm rp} = 0.31$. This percolation transition is in the 3D *random percolation* universality class.

The dramatic effect of frustration on particle motion can be figured considering the particles which diffuse, exchanging their positions with a nearest-neighbour hole, in such a way that no unsatisfied spin interactions are introduced. At low particle densities, motion is not inhibited by quenched frustration because of the abundance of holes. However, at high densities, a given particle can diffuse through the system only by a large-scale, cooperative rearrangement of many particles.

This effect becomes apparent by studying quantities strictly related to the atomic motion of sites (in this section we present data for a system with $L = 16$, and analogous results are found for $L = 8$). The particles mean-square displacement,

$$R^2(t) = \left\langle \frac{1}{N}\sum_i (r_i(t) - r_i(0))^2 \right\rangle,$$

shows deviation from simple Brownian linear time dependence and exhibits at intermediate times with an inflection region, which becomes evident in a region above $\mu_{\rm p}$ and increases

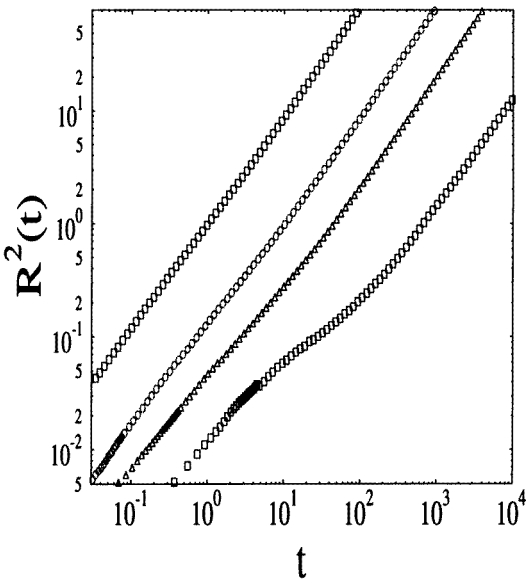

Figure 2. Mean-square displacement $R(t)^2$ in a 3D system of size $L = 16$ at $J = 10$ for $\rho = 0.271, 0.440, 0.581, 0.674$ (higher curves correspond to lower densities).

approaching μ_g (see figure 2). The long-time asymptotic linear time behaviour of $R^2(t)$ defines the diffusivity D, which shows an apparent shoulder at about $\mu^* \sim 2$ ($\rho^* \sim 0.5$) as shown in figure 3. Below this value, it is possible to fit $D(\mu)$ with a power law

$$D(\mu) = A_0\ (\mu_0 - \mu)^\gamma \tag{3}$$

with $A_0 = 1.2 \times 10^5$, $\mu_0 = 5.3$ and $\gamma = 7.0$, and above this value with a Voghel–Tamman–Fulcher law

$$D^{-1}(\mu) = A_1\ \exp(B/(\mu^{-1} - \mu_1^{-1})) \tag{4}$$

with $A_1 = 17$, $\mu_1 = 11.4$ and $B = 0.3$. An Arrhenius fit works too. The value μ_0 from the power-law fit corresponds to the characteristic temperature T_c of mode coupling theory, or to the 'dynamic transition' of mean-field theory of p-spin glasses. A crossover from power law to Arrhenius (or Voghel–Tamman–Fulcher) behaviour is also observed in real experiments [1, 2].

We have studied other time-dependent quantities in the system [15], such as the Fourier transform density–density and square magnetization autocorrelation functions:

$$C_k(t) = \langle \rho_k(t)\rho_{-k}(0)\rangle / \langle \rho_k \rho_{-k}\rangle \qquad \text{and} \qquad C_m(t) = \langle m^2(t)m^2(0)\rangle / \langle m^4\rangle$$

(here ρ_k is the Fourier transform of density and m the magnetization). They show a distinct two time decay above μ_p and have a definite long-time plateau value higher than zero in the region above μ_g (see figure 4). The shorter of these two times, τ_{exp}, is linked to the short-time exponential decay (time is measured in such a way that a single lattice update corresponds to unity). The longer time is associated to a stretched exponential decay on the long-time scale $C(t) = A\ \exp(-(t/\tau)^\beta)$ [16], with an exponent β which becomes less than 1 just around μ_p, approaching a value $\beta(\mu_g) \sim 0.2$ at the glass transition. The time τ, so defined, has a behaviour analogous to D^{-1} with a less apparent change at μ^*.

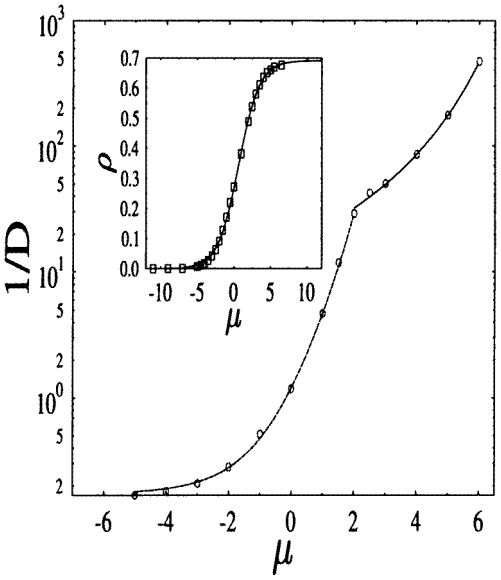

Figure 3. Inverse of diffusivity $D(\mu)$ as a function of μ in a 3D system of size $L = 16$, at $J = 10$. Superimposed lines are the power-law (broken curve) and Voghel–Tamman–Fulcher (full curve) fits quoted in the text. The inset reports the density ρ as a function of μ. The full line is a Fermi–Dirac fit: $\rho(\mu) = \rho_\infty/[1 + \exp(-(\mu - \mu_0)/\Delta\mu_0)]$, with $\rho_\infty = 0.69$, $\mu_0 = 0.66$ and $\Delta\mu_0 = 1.4$.

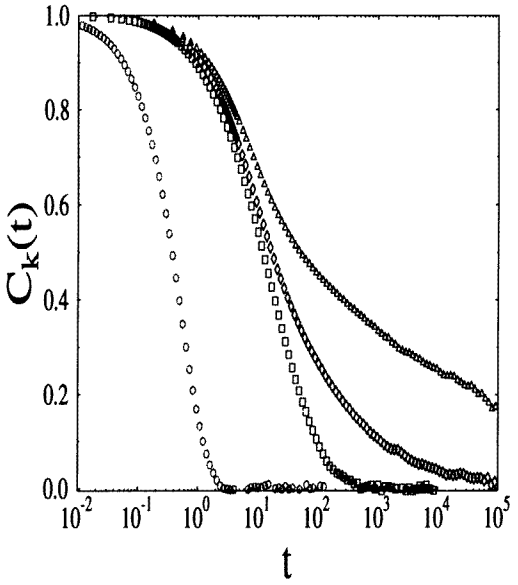

Figure 4. The Fourier transformed density time correlation function, $C_k(t)$, for an intermediate value of k, as a function of time t for several values of the chemical potential μ ($\mu = -2.0, 3.0, 4.0, 6.0$), in a 3D system of size $L = 16$, at $J = 10$.

In this paper we have studied the properties of a 3D frustrated lattice gas. This model, which bridges spin glasses and site frustrated percolation, exhibits a dynamical behaviour similar to that found in glass-forming systems.

This model, similar to frustrated percolation, exhibits a percolation transition at μ_p, before the glass transition μ_g, where unfrustrated clusters of neighbouring sites begin to feel non-local effects of frustration. This transition corresponds to a crossover from normal behaviour to anomalous behaviour where precursors of the glass transition set in.

This may also suggest that in glass-forming liquids there may be a relevant temperature well above the glass transition corresponding to a crossover from normal to anomalous behaviour which may be responsible for various precursor phenomena, such as the onset of stretched exponentials, the breakdown of the Stokes–Einstein relation [15] and the presence of temporal heterogeneity [11]. A percolation transition well before the glass transition has recently been discovered by Tomida and Egami [17] in a molecular-dynamics simulation of monoatomic liquids. It is also interesting to note that Kivelson [18] showed that the viscosity of 15 glass-forming liquids could be collapsed on one single curve, by assuming only one characteristic temperature well above the glass transition.

In this model, as in frustrated percolation, we observe at μ_g the divergence of a cooperative length ξ, as suggested in the original theory of Adam and Gibbs [5]. This point, which seems to be located where the particle macroscopic diffusion motion is arrested, does not show any divergence in the density fluctuations.

We have observed that, at an intermediate region between μ_p and μ_g (around μ^*), the diffusivity constant changes its behaviour with μ. This region corresponds to the non-equilibrium 'freezing temperature' found in spin glasses [4] or the manifestation in finite-dimensional systems of the mean-field 'dynamical transition' known in the SG literature [19]. In this region, where the diffusivity $D(\rho)$ shows an apparent discontinuity in its derivative, many features commonly observed in experiments on glass-forming liquids appear [1, 2].

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