Rapid Note

Fluctuation-dissipation ratio in lattice-gas models with kinetic constraints

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Abstract. We investigate by Monte-Carlo simulation the linear response function of three dimensional structural glass models defined by short-range kinetic constraints and a trivial equilibrium Boltzmann-Gibbs measure. The breakdown of the fluctuation-dissipation theorem in the glassy phase follows the prediction of mean field low temperature mode-coupling theory.

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1 Introduction

Glassy materials are a paradigmatic example of systems characterized by slow relaxation and physical aging phenomena [1,2]. While thermodynamic observables are rather regular along the glass transformation, dynamical properties undergo drastic changes, and in particular relaxation times increases up to 15 orders of magnitude. The microscopic origin of such a dramatic slowing down is the so called *cage effect*: a tagged particle in a liquid does not perform a simple random walk but rattles in a cage formed by the surrounding particles, and moves away from it only if the particles building up the cage walls are themselves able to move. At low temperature or high density, when the degrees of freedom of the particles become strongly coupled, this cooperative process exceeds observation time and the glass appears like a "solid" [3].

However, structural relaxation is not completely absent in glasses, and molecular rearrangements still occur on longer time scales. Indeed, after a quench in the low temperature or high density phase, particle diffusion does not stop altogether, but rather becomes slower and slower as time goes on. This is the origin of *aging effect*: the response to an external perturbation depends on the "age" of the sample, *i.e.*, on the time elapsed since its preparation, even at very long times. Therefore in this regime glassy systems never reach thermodynamic equilibrium and, consequently, time translational invariance (TTI) and the fluctuation-dissipation theorem (FDT) are violated.

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2 The problem

Despite the remarkable progress realized in the study of out-of-equilibrium dynamics of aging systems [4], the detailed nature of ergodicity breaking in finite-dimensional glasses is not yet fully understood. In mean-field mode-coupling theory the glass transition appears as a purely dynamic effect due to an instability of the equation governing the correlation of density fluctuations [3]. In particular, mean-field disordered models of structural glasses show that the origin of this dynamical transition is the existence of a large number of metastable states which trap the system for an infinite time. On the other hand, the life-time of metastable states in finite-dimensional short range models is finite, since it is always possible to nucleate, by a thermally activated process, a droplet of the stable phase. Therefore the dynamical transition appear as an artifact of the mean-field approximation, and in real glasses this transition would be a *finite-time* effect, at least on time scales much smaller than the life-time of metastable states [5–8].

However, since salient features of glassy behavior are essentially of dynamical nature, and since dynamical universality classes are smaller than the static ones, it is important to establish how glassy effects depend on the details of dynamics. In this paper we explore the limit case of three dimensional lattice gas models defined only by short-range kinetic constraints and by a trivial equilibrium measure, in which the glass transition appears to have a purely dynamical origin unrelated to the existence of metastable states [9,10]. Previous Monte-Carlo simulations have shown that this model reproduces qualitatively the "glassy" phenomenology, including annealing-rate

dependence, irreversibility effects, and waiting-time dependent logarithmic diffusion [10,11]. Here we investigate aging effects in the linear response function in order to characterize the violation of FDT [12,13].

3 The model

Our starting point is the kinetic lattice-gas model introduced by Kob and Andersen in order to mimic the cage effect in supercooled liquids [9]. The system consists of N particles in a cubic lattice of size L^3, with periodic boundary conditions. There can be at most one particle per site. Apart from this hard-core constraint there are no other static interactions among the particles. At each time step a particle and one of its neighbouring sites are chosen at random. The particle moves if the three following conditions are all met:

1. the neighbouring site is empty;
2. the particle has less than 4 nearest neighbours;
3. the particle will have less than 4 nearest neighbours after it has moved.

The rule is symmetric in time, detailed balance is satisfied and the allowed configurations have the same weight in equilibrium. With this simple definition one can proceed to study the dynamical behavior of the model at equilibrium. One observes that the dynamics becomes slower and slower as the particle density ρ increases; in particular, the diffusion coefficient of the particles, D, vanishes as the density ρ approaches the critical value $\rho_c \simeq 0.88$, with a power law

$$D(\rho) \sim (\rho_c - \rho)^{\phi}, \qquad (1)$$

with an exponent $\phi \simeq 3.1$ [9].

Since we are interested in the dynamical approach to the putative equilibrium state we allow the system to exchange particles with a reservoir characterized by a chemical potential μ. Therefore, we alternate the ordinary diffusion sweeps with sweeps of creation/destruction of particles on a single layer with the following Monte-Carlo rule: we randomly choose a site on the layer; if it is empty, we add a new particle; otherwise we remove the old particle with probability $e^{-\beta\mu}$. The number of particles is no longer fixed and the external control parameter is μ, which plays the role of the inverse temperature. In this way we can prepare the system in a non equilibrium state by a process analogous to a quench, which is represented by a jump in $1/\mu$ from above to below $1/\mu_c$ (where μ_c is defined through the state equation $\rho(\mu_c) = \rho_c$). The situation becomes analogous to the *canonic* case in which one controls the temperature, and the energy endeavors to reach its equilibrium value.

4 Linear response

Following a suggestion by Kurchan, we compute the linear response function by applying to our system a small random perturbation at time t_w:

$$\mathcal{H}_\epsilon = \epsilon \sum_{a=1}^{3} \sum_{k=1}^{N} f_k^a r_k^a , \qquad (2)$$

where $f_k^a = \pm 1$ independently for each coordinate a and particle k at position r_k^a. The response function of the system at time $t + t_w$ is defined by

$$R(t + t_w, t_w) = \frac{1}{3N} \sum_{a=1}^{3} \sum_{k=1}^{N} \left\langle \frac{\partial f_k^a r_k^a(t + t_w)}{\partial \epsilon(t_w)} \right\rangle \bigg|_{\epsilon=0} . \qquad (3)$$

The linear regime is probed for small enough values of the perturbation strength ϵ. It is actually more convenient to look at the integrated response function:

$$\chi(t + t_w, t_w) = \int_{t_w}^{t+t_w} R(t + t_w, \tau)\epsilon(\tau)d\tau . \qquad (4)$$

In order to measure this quantity we prepare the system in a non-equilibrium state by quenching to the subcritical value of $1/\mu$ and letting it relax up to time t_w; at t_w, we make a copy of the system and apply the perturbation; we then let evolve the two systems with the *same* succession of random numbers and measure the difference $\Delta r_k^a(t + t_w)$ between the displacements that take place in the two systems at time $t + t_w$. At constant field we obtain

$$\chi(t + t_w, t_w) = \frac{1}{3N} \sum_{a=1}^{3} \sum_{k=1}^{N} \langle f_k^a \Delta r_k^a(t + t_w) \rangle . \qquad (5)$$

We have performed a sudden quench to the subcritical value $1/\mu = 1/2.2$ starting from a configuration with density 0.75 and measured the integrated response function $\chi(t + t_w, t_w)$ *vs.* time t at different waiting times t_w. We have checked that for $0.05 < \epsilon < 0.15$ non-linear effects are absent. In the following we present the results for the case $\epsilon = 0.1$ and for a cubic lattice of size 20^3. Figure 1 shows that for small values of t_w TTI holds, and aging effects set in only for larger value of t_w. To obtain the scaling behavior one has to consider an effective waiting time, $\tau_w = t_w + \tau_0$, that takes into account the relaxation time, τ_0, of the system before the quench. Indeed, Figure 2 shows that, if one plots $\chi(t + t_w, t_w)$ *vs.* t/τ_w, the curves lie roughly on top of each other and follow a *simple* aging logarithmic behavior, $\chi(t + t_w, t_w) \sim \log(1 + t/\tau_w)$. Similar results were previously found also for the mean square displacement [10,11].

5 Fluctuation-dissipation ratio

We are now in a position to investigate the nature of breakdown of thermodynamical equilibrium. We consider the generalized FDT proposed by Cugliandolo and Kurchan in the framework of dynamical mean-field theory of spherical p-spin glass model [12]. In our case the relevant
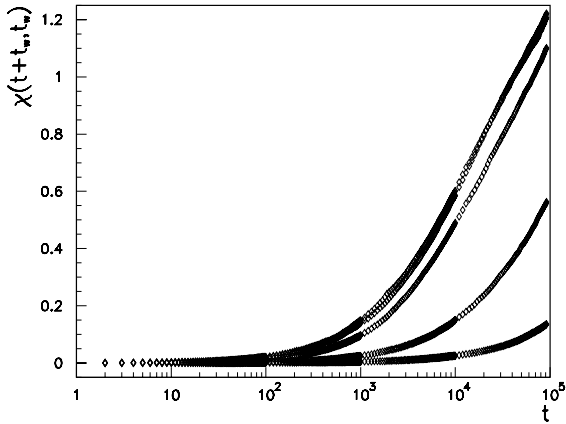
Fig. 1. Integrated response function $\chi(t + t_w, t_w)$ *vs.* time t after a quench to the subcritical values $1/\mu = 1/2.2$. The measurements were carried out after the perturbation was applied at waiting times $t_w = 10, 10^2, 10^3, 10^4, 10^5$. The initial density is 0.75. Average over 20 samples.
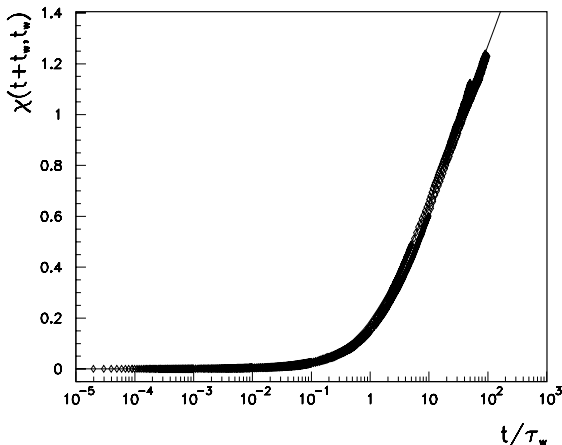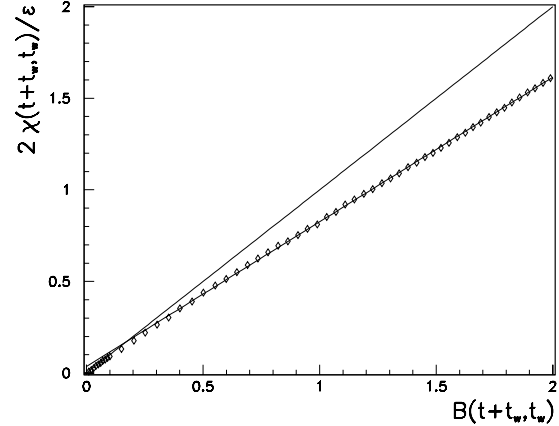

Fig. 3. Parametric plot of $2\chi(t + t_w, t_w)/\epsilon$ *vs.* mean square displacement $B(t + t_w, t_w)$ measured after a waiting time $t_w = 10^5$ from a quench to the subcritical value $1/\mu = 1/2.2$. The density of initial configuration is 0.75. The two straight line have slope 1 (FDT line) and 0.79. Average over 200 samples; error bars comparable to symbol size.

Fig. 2. Scaling behavior of integrated response function $\chi(t + t_w, t_w)$ *vs.* t/τ_w, where $\tau_w = t_w + \tau_0$ with $\tau_0 = 10^3$ and $t_w = 10, 10^2, 10^3, 10^4, 10^5$. The full line corresponds to $\chi(t + t_w, t_w) \sim \log(1 + t/\tau_w)$.

property is the off-equilibrium generalization of Einstein-Stokes relation among diffusion coefficient and viscosity, given by [14]:

$$X(B)\frac{\partial B(t, t_w)}{\partial t_w} = -2TR(t, t_w), \qquad (6)$$

where

$$B(t, t_w) = \frac{1}{3N} \sum_{a=1}^{3} \sum_{k=1}^{N} \left\langle [r_k^a(t) - r_k^a(t_w)]^2 \right\rangle, \qquad (7)$$

is the mean square displacement. The fluctuation-dissipation ratio (FDR), X, depends on both times only

through B and its departure from 1 is a measure of violation of FDT. The integrated version of the generalized FDT allows to write (considering a constant field):

$$\chi(t, t_w) = \frac{\epsilon}{2T} \int_0^{B(t, t_w)} X(B) \, dB . \qquad (8)$$

(We always write explicitly the temperature T, however it does not play any role.) Therefore, if FDT holds, we would have a straight line with slope $\epsilon/2T$ in the parametric plot of $B(t + t_w, t_w)$ *versus* $\chi(t + t_w, t_w)$; while a deviation from this straight line indicates a failure of FDT. The way in which FDT breaks down plays a key role in aging systems since it signals the presence of a time-scale dependent effective temperature [13], and this violation can be bounded by a quantity related to the entropy production [16]. The FDR has been measured by Monte-Carlo simulations in several models such as spin-glasses [17,20], Lennard-Jones binary mixtures [18], domain growth models [19], and directed polymers [21], and so far all the findings are consistent with the prediction of generalized FDT. We have carried out extensive numerical simulation to measure the FDR, $X(B)$, in three dimensional lattice-gas models with kinetic constraints. We observe the two asymptotic regimes also found in the out of equilibrium dynamics of mean field models of structural glasses [12]:

- at times t smaller than t_w we observe a short-time quasi equilibrium regime where FDT holds;
- at larger separation times FDT breaks down with a constant violation factor.

These results appear clearly in the parametric plot [15] of $\chi(t+t_w, t_w)$ *vs.* $B(t+t_w, t_w)$ where, as can be seen in Figure 3, the curve approaches the characteristic broken line. (In order to probe the asymptotic time regime we have
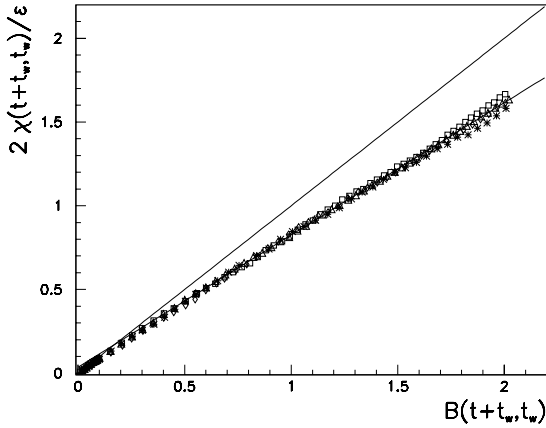
Fig. 4. Parametric plot of $2\chi(t+t_w, t_w)/\epsilon$ *vs.* mean square displacement $B(t+t_w, t_w)$ measured after a waiting time $t_w = 10^5$ from a quench to the subcritical value $1/\mu = 1/2.05$ (stars), $1/2.1$ (triangles), $1/2.2$ (diamonds), $1/2.3$ (squares). The density of initial configuration is 0.75. The two straight line have slope 1 and 0.79. Average over 200 samples; error bars comparable to symbols size.

chosen $t_w = 10^5$.) It is natural in this context to interpret the quasi-equilibrium regime as corresponding to the motion of particle inside a cage, although the size of cage in this model is quite small and of the order of a fraction of lattice spacing. After that the system crosses over a diffusion-cage regime where a departure from equilibrium is observed with a constant FDR.

In order to see if the inverse chemical potential plays the role of a "true" temperature, we have measured the function $X(B)$ by quenching the system at different subcritical values. Figure 4 shows that the slope of the FDT violating line, as well as the location of the breaking point, are independent of the quench value of $1/\mu$, at least in the time window explored in our numerical experiments and in the range of values of $1/\mu$ used. Therefore as suggested in reference [11], the FDR appears locked in at the value the system would exhibit at the critical density, and should be only related with "universal" properties of the model, like for instance the exponent ϕ.

6 Conclusions

To summarize we have shown that three dimensional lattice-gas models defined by short range kinetic constraint and trivial equilibrium Boltzmann-Gibbs measure exhibit a *simple* aging behavior in the integrated response function and violate fluctuation-dissipation theorem in a manner similar to mean field models of structural glasses. The fluctuation-dissipation ratio in the glassy phase appears to be a constant independent on the chemical potential of the reservoir; however, different results are expected if the boundary condition is changed or if creation and destruction of particles within the bulk is allowed.

Given the non-holonomic nature of kinetic constraints and the trivial Hamiltonian of the model, any statistical mechanics approach based on the calculation of some partition function should be unsuitable to grasp the features of the glassy phase. In order to understand if aging is interrupted after a certain characteristic time [22], it would be interesting to analytically investigate the long-time dynamical behavior of the model. A persistent aging scenario would prove that also in finite-dimensional short-range models the glass transition may have a purely dynamic origin unrelated to any underlying thermodynamic singularity.

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