Glassy Dynamics

in Lattice Gas Models

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Work in collaboration with Estelle Pitard and Valery Van Kerrebroeck (`cond-mat/0404214`, and in preparation).

Try and qualify slow dynamics in lattice gas models: is it interesting?

Get information about the spatial structure.

Is the (far away) equilibrium landscape relevant?

A: Probably partially.

ENS, Paris, June 2004

Summary:

- Slow dynamics.
- Lattice gas models.
- Story of the Kob Andersen model.
- The issue of the phase transition, and its solution.
- Length scales in KA. (crucial for qualifying slow dynamics).
- Persistence times.
- Relaxation of correlation functions.
- Spatio-temporal correlation functions.
- Connected clusters of frozen particles.
- The scaling behavior.
- A first analysis of the Biroli-Mézard model.

Slow, *glassy* dynamics:

- very general relevance;
- dramatic features.

But:

- which are the really relevant features?
- what is really happening?

Two principal paradigms:

- (static) landscape is far away but it is important;
- dynamical heterogeneities lead, landscape is not important.

Viscosity increases: what is happening?

Use: lattice gasses.

The Kob Andersen (KA) Model.

(Here 3d).

Purely dynamical.

Lattice gas of N classical particles.

$\vec{R}_i = (x_i, y_i, z_i)$

on a simple cubic lattice with periodic boundary conditions and size L^3.

Each particle occupies one site;
one site can contain at most one particle.

$n_i = (0, 1)$ occupation number of site i.

Simple hard core repulsion (plus dynamical rule, see later: jamming is forced by a dynamical rule).

- $\Longrightarrow$ all configurations have the same probability;
- $\Longrightarrow$ a random configuration is a typical equilibrium configuration.

KA is somehow the mother of all interesting dynamical models.

- no need for equilibration;
- no aging.

We define the dynamics of the model.

1. Select a particle at random.
2. Pick a random nearest neighbor site, and advance the clock of $1/N$.
3. if) the nearest neighbor site is empty AND the particle has m or less neighboring particles AND the nearest neighbor site has $m+1$ or less occupied neighboring sites then swap particles.

   else) do nothing.
4. goto 1.

Small m is blocked, large m is a free gas. We have $m=3$. Relaxation is a diffusive process with additional steric constraints.

Before discussing some (putative and/or correct) features of the KA model, we introduce a similar but very different interesting model.

Biroli-Mezard model.

This is a lattice glass, more similar indeed to hard spheres than KA. The model is defined thermodynamically: configurations that violate a density constraint are forbidden.

Here we have the same lattice structure than for the KA model, but a particle cannot have more than m among its $2d = 6$ nearest neighbor sites occupied.

It is a "coarse-grained" version of an off-lattice hard sphere system.

Bethe-lattice mean field can be analyzed with the cavity method.

We will **not** discuss here about general Facilitated Spin Models (Fredickson Andersen 1984).

$n_1 = 1$, mobile, low density region.

$n_1 = 0$, less mobile, high density region.

(not particles, but coarse-grained densities!)

Low densities regions in the neighborhood *facilitate* rearrangements (i.e. *spin flips*).

A special role is played by versions of the model with directed or asymmetric constraints. For example EAST model, 1991.

Here only nearest neighbor spins in some given lattice direction can act as facilitators.

$d = 1$: flip only if left nearest neighbor is up.

What one finds, at first glance, about the KA model? (Kob Andersen 1993) $d = 3$. Diffusion coefficient D.

$$D \sim (0.881 - \rho)^{+3.2}$$

i.e. there appear to be a critical density $\rho_c \simeq 0.881$ where the system gets blocked. (D is computed from the mean square displacement measured at different densities). SEE FIGURE IN NEXT SLIDE.

A second part of the original study was based on analyzing the density-density correlation function $G(r,t)$, $F(k,t)$.

$$G(r,t) = \frac{1}{3N} \sum_{\alpha} \sum_{i=1}^{N} \langle \delta \left(r_i^{\alpha}(t) - r_i^{\alpha}(0) - r \right) \rangle - \frac{1}{L}$$

and F Fourier transform. That gives a time scale τ. τ scaling is compatible with the same value of ρ_c.

i.e.: dynamical transition from ergodic to non-ergodic behavior? (Answer will be no!)

Sorry for the low level image.

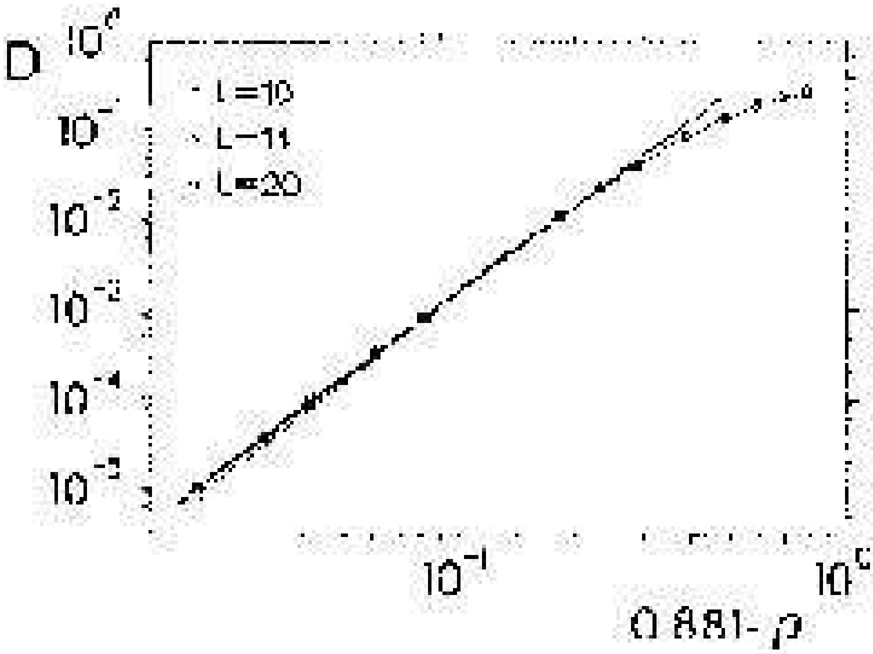


FIG. 2. Diffusion constant D for $L = 10, 14$, and 20 versus $0.891 - \rho$. The straight line is a power-law fit with exponent 4.1.

On many decades...

It would seem a very clean signal.

Franz, Mulet and Parisi, PRE 2002.

KA model: a non-standard mechanism for the glassy transition.

Look at blocked configurations (believe in $\rho_c = 0.881$ in $d = 3$).

(see also Barrat, Kurchan, Loreto and Sellitto, PRL 2000)

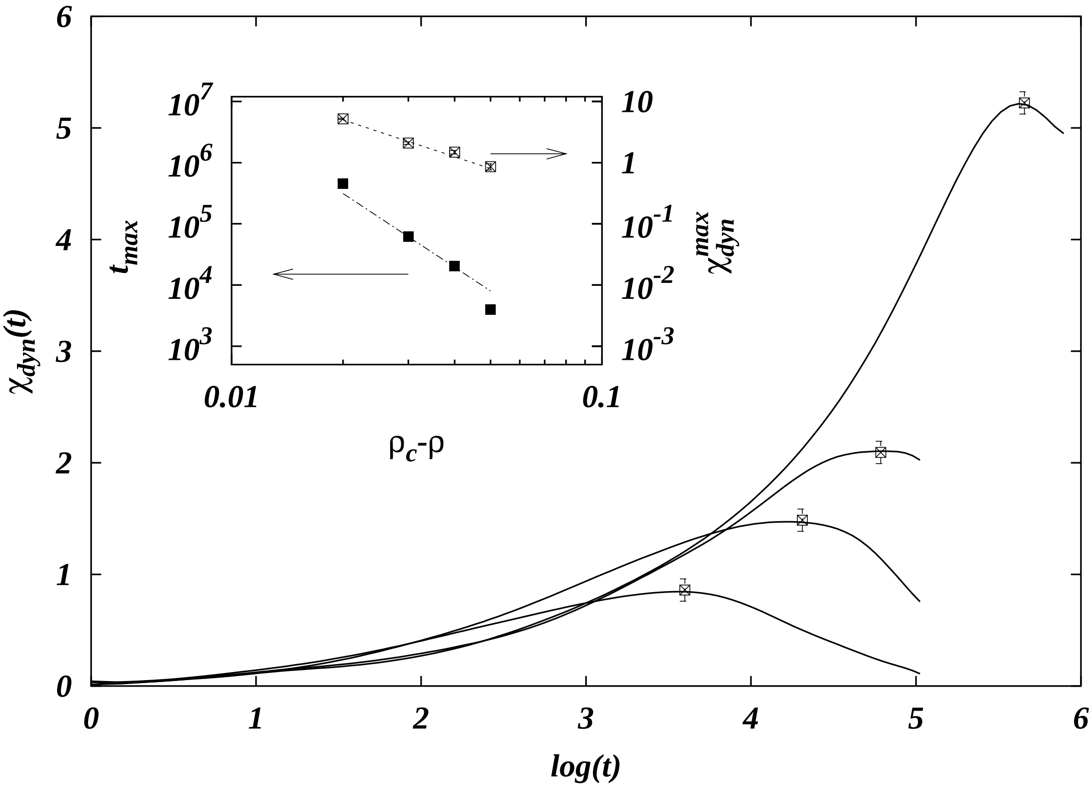
$$q(t) \equiv \frac{1}{N\rho\,(1-\rho)} \sum_i \left(n_i(t) n_i(0) - \rho^2\right)$$

$$\chi_4(t) \equiv N \left(\langle q^2(t) \rangle - \langle q(t) \rangle^2 \right)$$

At $t = t^*$ $\chi_4(t)$ is maximum.

SEE FIGURE IN NEXT SLIDE.

First sign of spatial heterogeneities + relation with p-spin spin glass models and mean field models (Franz Parisi 2000, Donati, Franz, Glotzer and Parisi 1999)



6
5
4
3
2
1
0
$\chi_{dyn}(t)$
0
1
2
3
4
5
6
log(t)
10^7
10^6
10^5
10^4
10^3
t_{max}
10
1
10^{-1}
10^{-2}
10^{-3}
χ_{dyn}^{max}
0.01
0.1
ρ_c-ρ

C. Toninelli, Phd Thesis, Roma La Sapienza, unpublished;

C. Toninelli, M. Biroli, D. Fisher, `cond-mat/0306746`, PRL;

L. Bertini and C. Toninelli, `cond-mat/0304694`;

C. Toninelli and G. Biroli, `cond-mat/0402314`.

Mathematical proof that $\rho_c^{KA,3D} = 1$, no transition can be present. The original hint was not correct. (Here $3d$, $m = 3$).

On the contrary it is proven that there is a dynamical transition on Bethe lattices.

In $3d$ the diffusion coefficient $D \longrightarrow 0$ faster than any power than the vacancy density $(1 - \rho)$.

In $3d$ *ghost of Bethe lattice* is probably connected to the sharp crossover at "ρ_c" ~ 0.881.

$$\xi \sim e^{e^{\frac{c}{1-\rho}}}$$

and Finite Size Effects will be of order $\log\log$.

Our analysis of the KA model.

Can we determine length scales that grow clearly when approaching $\rho = 1$, i.e. criticality?

We will have to start by studying the regime of the "arrest" for $\rho \longrightarrow \rho_c^* \sim 0.881$ and we will have to enquire about the presence of slow stretched exponential relaxations.

Again, we know from Toninelli, Biroli and Fisher that

$$\rho_c(L) \sim 1 - \frac{c}{\log \log L}$$

$$\Xi(\rho) \sim e^{e^{\frac{c}{1-\rho}}}$$

spacing between mobile particles.

We will describe now spatio-temporal correlations and heterogeneities that characterize the dynamics of the KA model.

Persistence times of particles

- Their distribution (a first stretched exponential...).
- Their spatial correlations (our first length scale...).

Start at time $t = 0$ (from a random configuration, that is, also an equilibrium configuration...).

τ_i: time after which particle i moves for the first time to a nearest neighboring site.

$$P_{(\rho)}\left(\tau_i = \tau\right)$$

and we average over all particles and over many initial configurations of the system.

Densities range from 0.5 to 0.8 (see next figure).

Our best fits are for a stretched exponential behavior for the integrated probability distribution

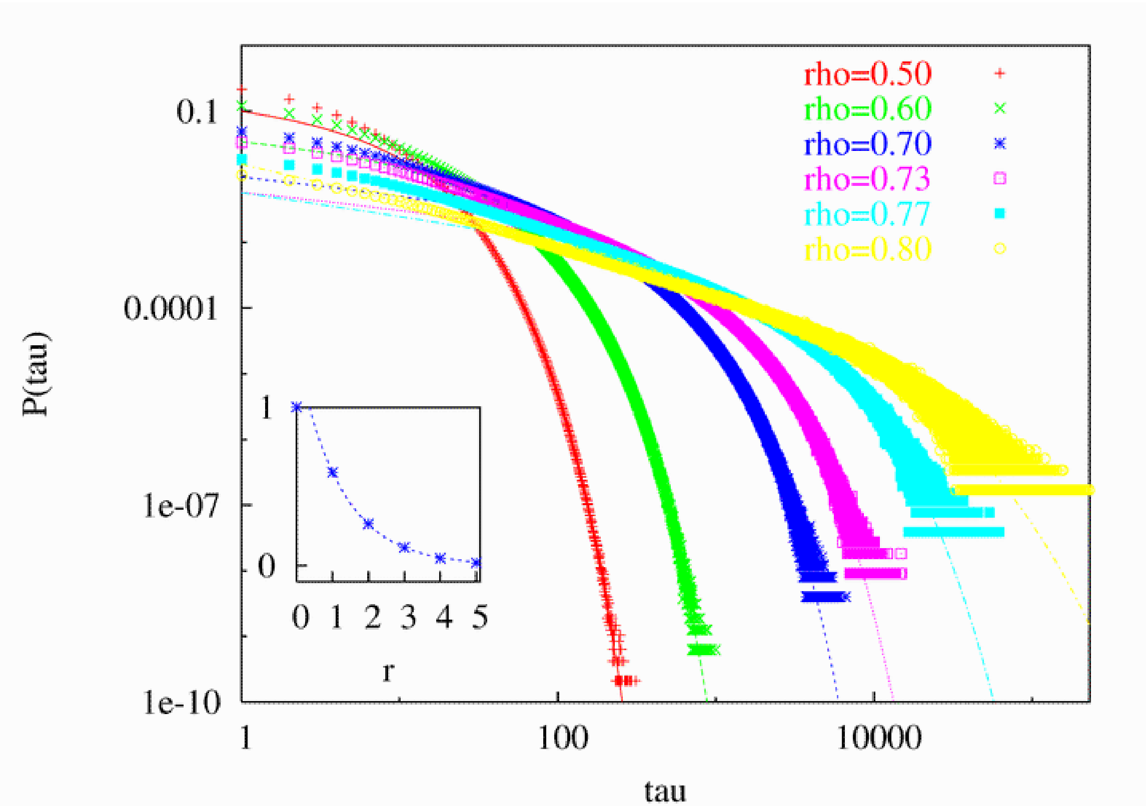
$$\int_{\tau}^{\infty} P(t)\, dt = \exp\left\{ - \left(\frac{\tau}{\tau_{trap}} \right)^{\beta} \right\}$$

We get a very good best fit.

We need $\beta < 1$ even at low densities.

β for best fit decreases as ρ increases. For example $\beta(\rho = 0.5) = 0.80$ and $\beta(\rho = 0.8) = 0.45$.

Analogous behavior is present in Lennard-Jones and in different Kinetic Constrained Models. Similar (but not identical...) values for β: this fact suggests an universal behavior.


rho=0.50
rho=0.60
rho=0.70
rho=0.73
rho=0.77
rho=0.80
0.1
0.0001
1e-07
1e-10
P(tau)
1
100
10000
tau
1
0
0 1 2 3 4 5
r

The site dependent trapping times are not homogeneous in space.

One very relevant question is whether the KA dynamics induces a spatial correlation length among them.

We have computed the spatial correlation

$$C_\rho(r) \equiv \frac{1}{N} \sum_{\vec{a}} (\langle \tau_{\vec{a}} \tau_{\vec{a}+\vec{r}} \rangle - \langle \tau_{\vec{a}} \rangle \langle \tau_{\vec{a}+\vec{r}} \rangle)$$

$\langle \cdots \rangle$ is for an average over initial configurations.

$l_c(\rho)$ is a *dynamical coherence length* such that for distances larger than $l_c(\rho)$ the trapping times are uncorrelated. An exponential fit, where $C_\rho(r) \sim e^{-r/l_c(\rho)}$, works well.

In the density range 0.7-0.8 $l_c(\rho)$ is small and of the order of one or two lattice spacings.

The growth of $l_c(\rho)$ for increasing ρ is well fitted by the correct essential singularity for $\rho \to 1$, $l_c(\rho) \simeq 0.17 \exp(\exp(0.16/(1-\rho)))$.

Relaxation of correlation functions

We have already defined:

$$q(t) \equiv \frac{1}{N\rho\,(1-\rho)} \sum_i \left(n_i(t) n_i(0) - \rho^2\right)$$

with $n_i = 0, 1$. We compute $\langle q(t) \rangle$ as an average over initial configurations.

In the asymptotic regime for a free lattice gas $\langle q(t) \rangle$ should decay as a power law (since dynamics is conserved). We do not see that (but even for a free lattice gas, at high densities the power law decay is far from clear). Asymptotic regime could be at astronomical times (typical in disordered systems) or KA could be different from a free lattice gas (possible but not very probable).

We observe again a very clear stretched exponential behavior, $\langle q(t) \rangle \sim e^{-\left(\frac{t}{t_{relax}}\right)^{\gamma}}$ with for example $\gamma(\rho = 0.5) \sim 0.8$ and $\gamma(\rho = 0.8) \sim 0.35$ (not so different from β).

A simple Griffiths argument would give an incorrect prediction.

Consider a region of size L and the appropriate relaxation time $\tau(L)$. The probability of finding a region of size L at density ρ is

$$P_\rho(L) = \rho^{L^d} = e^{-aL^d}$$

Since asymptotically we have a diffusive motion at all length scales (Toninelli, Biroli and Fisher) we have $\tau(L) \sim \tau_0 L^2$. So we find

$$\langle q(t) \rangle \sim \int dL\ P(l) e^{-\frac{t}{\tau(L)}} \sim e^{-t^\gamma}\ ,$$

with $\gamma = \frac{d}{d+2}$, i.e $\gamma_{d=3} = 0.6$ independent from ρ. This is not so.

Again. We detect a stretched exp behavior and not a power (that is probably only valid at later times: this effect is common in disordered systems). Strictly speaking the theorem implying power decay for $\langle q(t) \rangle$ is not necessarily valid for KA at finite N, but probably the relevant mechanism is not connected to this fact.

Spatio-temporal correlation functions

Franz-Mulet-Parisi used the dynamic susceptibility

$$\chi_4(t) \equiv N \left(\langle q^2(t) \rangle - \langle q(t) \rangle^2 \right)$$

as the main ingredient towards claiming that KA model has non-trivial spatial structures. It has a maximum at t^*, where the sensitivity of the system is maximal.

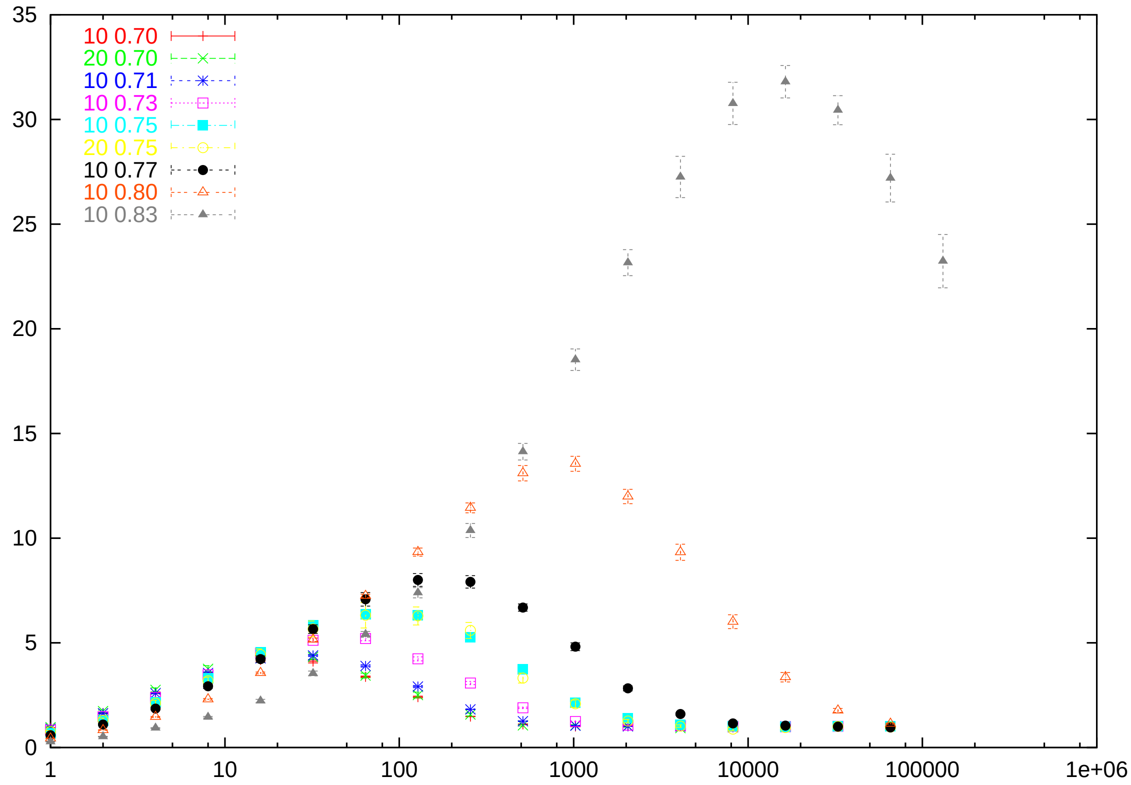
We have repeated (more precisely, in a range of densities and for different lattice sizes) the measurements of FMP. We find a compatible qualitative behavior (our data also have the correct limit $\chi_4(t \longrightarrow \infty) \longrightarrow 1$ established by Ritort and Sollich)

SEE FIGURE IN NEXT SLIDE.

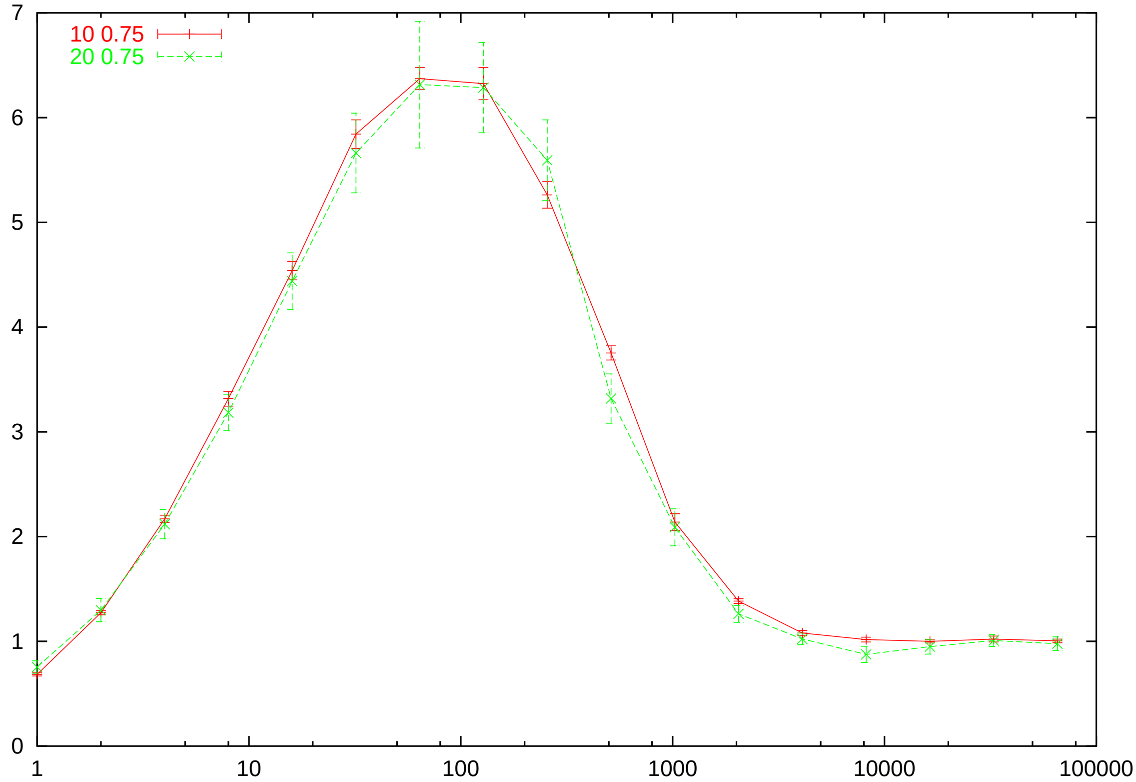
finite size effects are under control

SEE FIGURE IN SECOND NEXT SLIDE.

Both χ_{max} and t^* as a function of ρ are very well fitted by $e^{e^{\frac{c}{1-\rho}}}$ scaling. At $t = t^*$ heterogeneity of the system is maximal.


10 0.70
20 0.70
10 0.71
10 0.73
10 0.75
20 0.75
10 0.77
10 0.80
10 0.83
35
30
25
20
15
10
5
0
1
10
100
1000
10000
100000
1e+06

No Finite Size Effects.



This heterogeneity can be analyzed in better detail by studying

the space dependent susceptibility $g_4(\vec{r}, t)$

which generalizes $\chi_4(t)$.

$$\begin{aligned} g_4(r,t) \equiv (N\rho^2(1-\rho)^2)^{-1} \sum_{|\vec{r}_i - \vec{r}_j| = r} & \\ & (\langle n_i(t) n_i(0) n_j(t) n_j(0) \rangle \\ - & \langle n_i(t) n_i(0) \rangle \langle n_j(t) n_j(0) \rangle) \ . \end{aligned}$$

SEE FIGURE IN NEXT SLIDE.

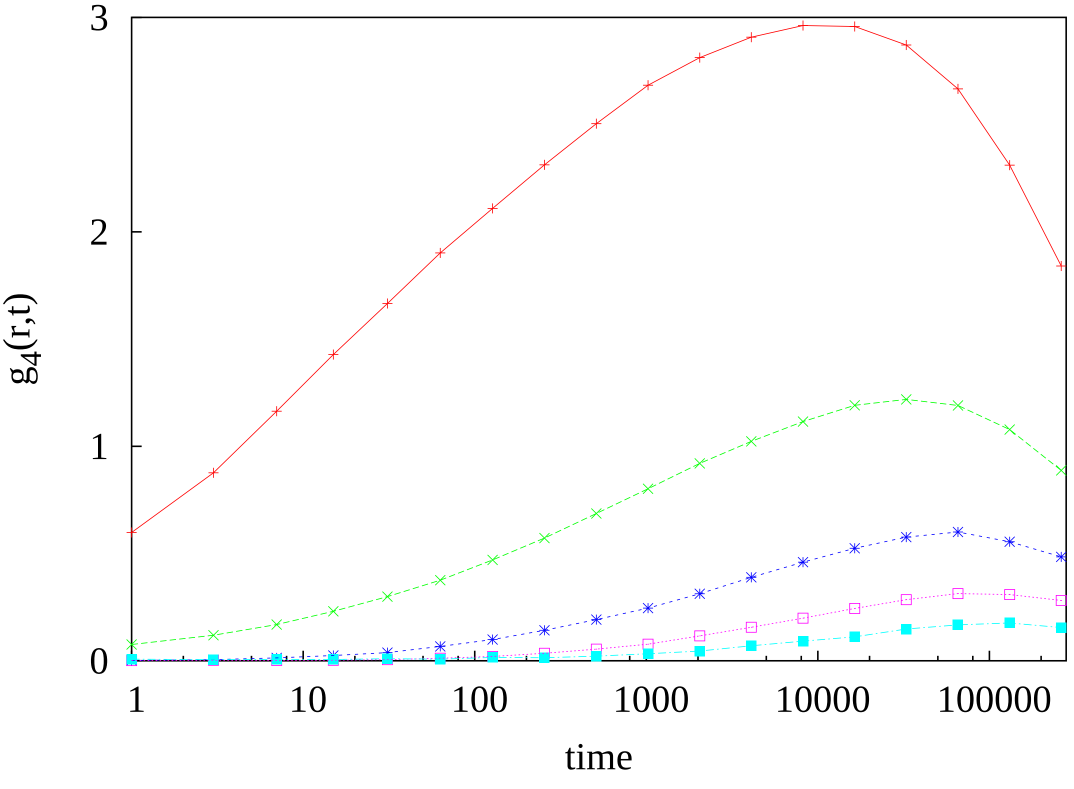
The position of the maximum is of the same order of magnitude than t^*.

It shifts to larger times for increasing r values.

The value of the max decreases for increasing r.

Larger max for smaller r.

Smaller t^* for smaller r.



The decrease of $g_4^{max}(r)$ determines a cooperativity length.

Indeed $g_4^{max}(r)$ shows a good exponential decay.

SEE FIGURE IN NEXT SLIDE.

$$g_4^{max}(r) \sim \frac{c(\rho)}{r^{\alpha(\rho)}} e^{-\frac{r}{\xi(\rho)}}$$

Best fit is good. For densities in the range

$$\rho \in (0.70 - 0.86)$$

we find

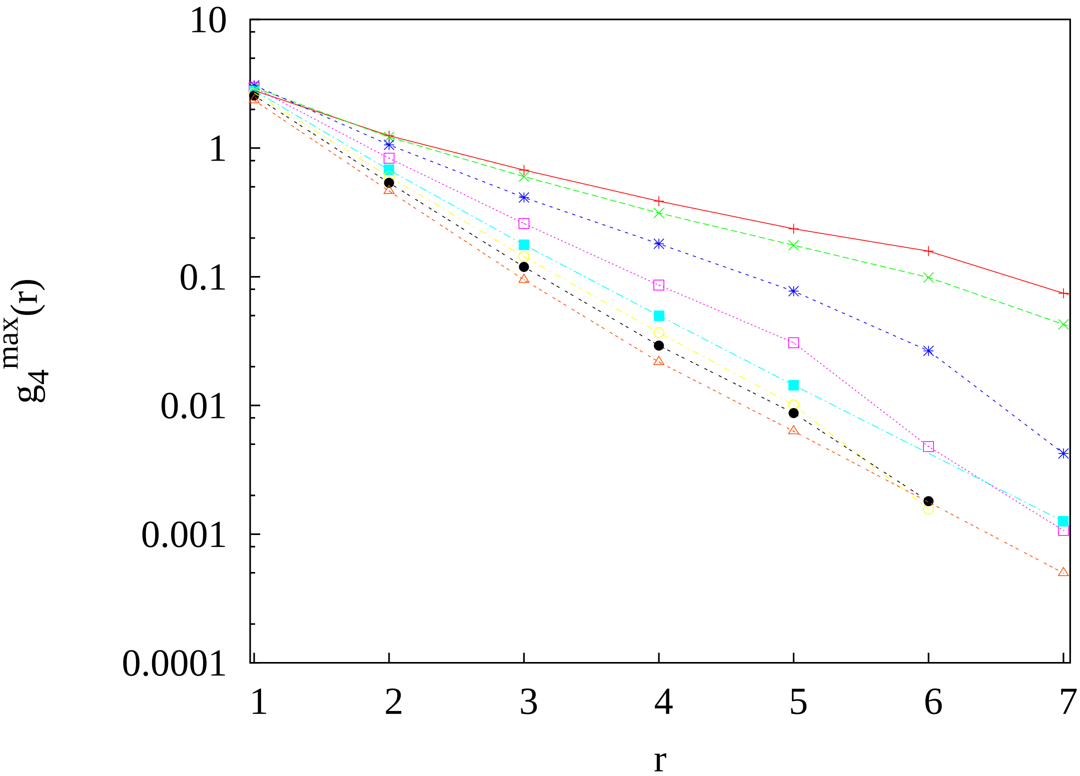
$$\alpha \in (0.22 - 0.64)$$

and

$$\xi \in (0.70 - 2.75)$$

.

When considering ξ as a function of ρ we find again a good fit to a (double) essential singularity as $\rho \to 1$.


10
1
0.1
0.01
0.001
0.0001
1
2
3
4
5
6
7
r
$g_4^{max}(r)$

Connected clusters of frozen particles

An important test of the structure of heterogeneities. Reconstruct connected clusters of particles that never moved at time t.

For example measure the probability of a cluster of size n

$$P(n,t)$$

We define and study different types of clusters:

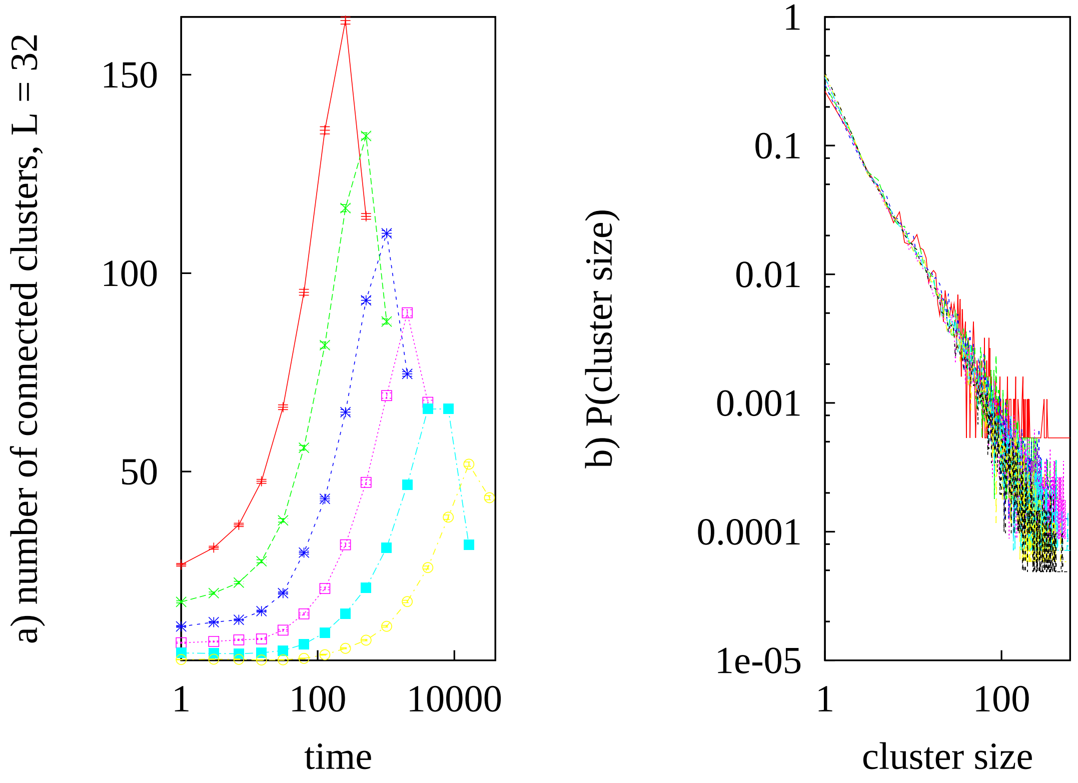
1. particles that never moved;
2. particles that at time t are in the original position even if they moved before.

In these two cases we find very similar results.

The first interesting thing to analyze is the number of blocked clusters. This number is maximal close to $t_{cluster}$.

SEE LEFT FIGURE IN NEXT SLIDE.

This defines a new time scale, again compatible with correct scaling.


a) number of connected clusters, L = 32
150
100
50
1
100
10000
time
b) P(cluster size)
1
0.1
0.01
0.001
0.0001
1e-05
1
100
cluster size

Let us discuss about $P(n,t)$.

At small times there are two different populations:

- few large blocked clusters (they dominate the rate of relaxation)
- small blocked clusters

As time goes particles move: the large clusters break and decrease in size. At intermediate time the two populations merge.
SEE RIGHT FIGURE IN PREVIOUS SLIDE.
Some hint for a power law, but not compelling, probably not very relevant. At the end all particles fly and all clusters disappear. At $t \sim t_{cluster}$ cooperativity is maximal.

We have also analyzed in detail

$$\lambda \equiv \langle n^{\frac{1}{3}} \rangle_{t_{cluster}}$$

and the related fluctuations. Again, λ scales as expected.

So, we can summarize about lengths.

We have determined with different approaches correlation lengths:

$$\xi_{\tau,\tau},\ \xi_{cg_4},\ \xi_{cc}$$

They all scale like

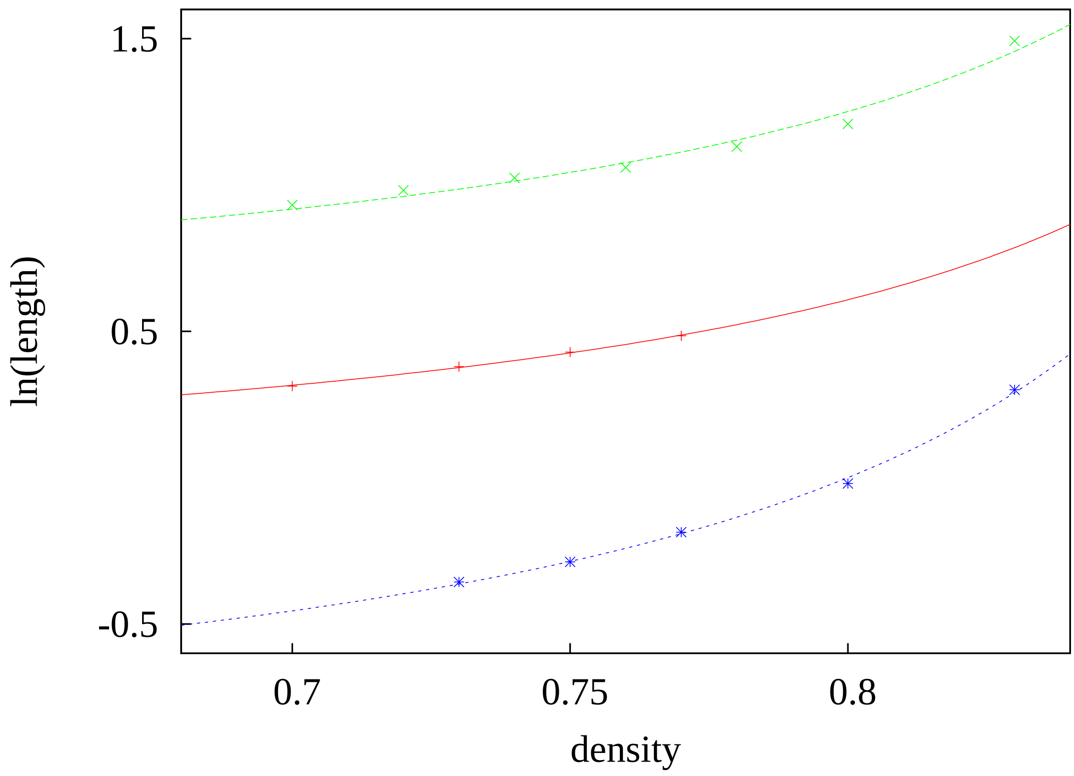
$$\xi \sim e^{e^{\frac{0.15}{1-\rho}}}$$

SEE FIGURE IN NEXT SLIDE.

These lengths are increasing but not very large. We get *blocked* already from *small* spatial regions.

The increasing correlation length is interesting: we detect a typical feature of a glassy dynamics even in absence of a landscape.

But: in the model as it is, without perturbation and with fixed number of particles, you cannot have aging.


1.5
0.5
-0.5
ln(length)
0.7
0.75
0.8
density

Some preliminary results about the Biroli-Mezard model.

Here:

1. simple model;
2. we have a landscape, i.e. you can have aging.

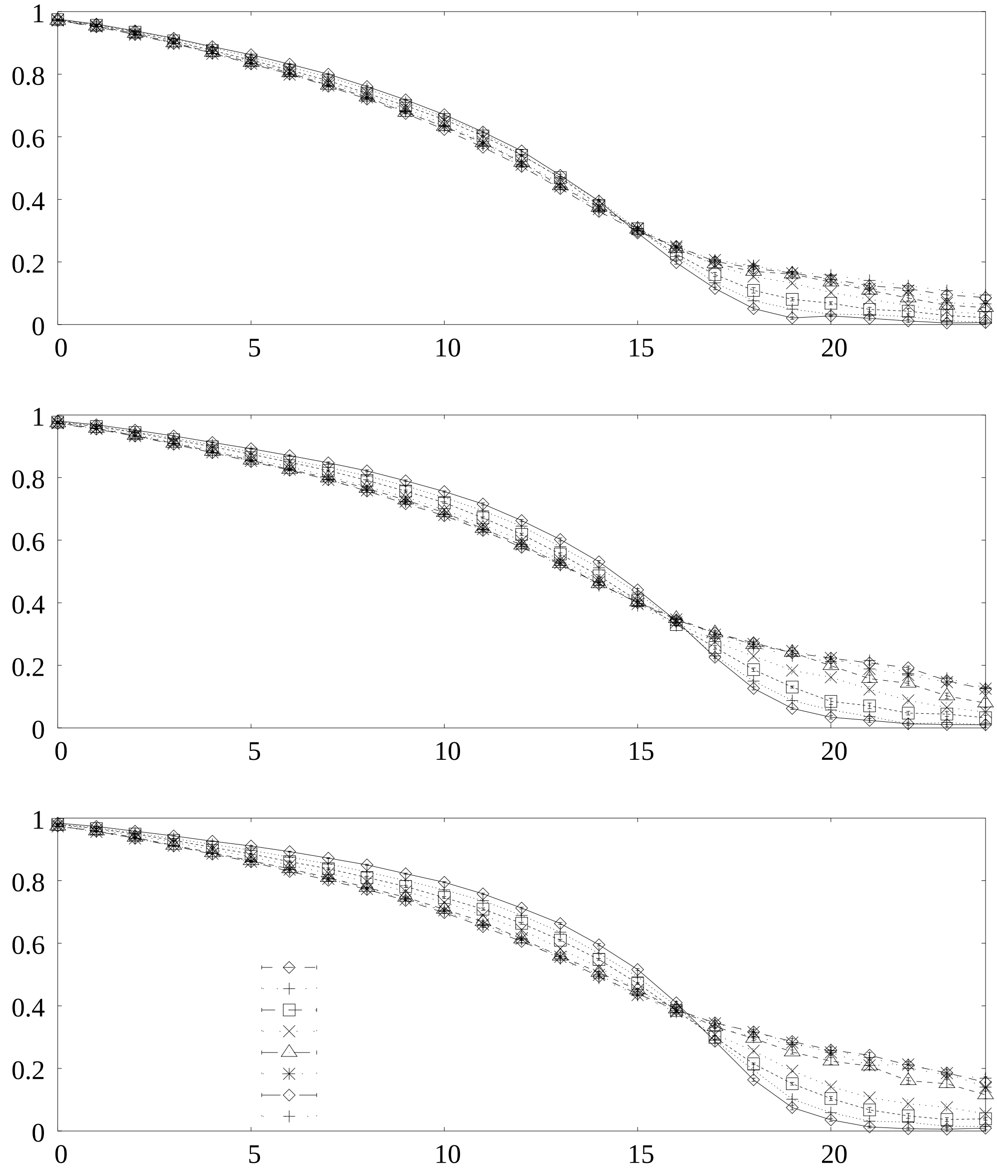
We study this model with the same approach used for KA.

Mixture: 0.7 $m = 3$, 0.3 $m = 1$, to avoid too fast crystallization.

Here preparation is difficult (the particle configuration has to obey all constraints). Annealing.

Results about aging, $\chi_4(t)$ peak, local structure.

Aging. $\rho = 0.535$, 0.540 and 0.542.



χ_4 peak. $\rho = 0.535$, 0.540 and 0.542.