Activated aging dynamics and negative fluctuation-dissipation ratios

L Berthier[1], J-P Garrahan[2], S Leonard[1], P Mayer[3,4], P Sollich[4]; R Jack[5]

[1]Montpellier, [2]Nottingham, [3]Columbia, [4]King's College London, [5]Oxford/Berkeley

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Outline

1. Motivation
   - Glasses, aging, mean-field effective temperatures
   - Non-mean-field effects: heterogeneity, **activation**
2. Fredrickson-Andersen model
   - Motivation, definition & intuition
   - Field theory for $d > 2$
   - Exact results for $d = 1$
   - Why is $d_c = 2$?
3. Summary
4. Outlook: Cooperative/fragile glasses – East model

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Glasses and aging

- Glasses: Paradigmatic non-equilibrium systems
- Relaxation times τ grow as T is lowered
- Or as density is increased: 'jamming' in colloids, granulars
- Glass transition: relaxation time $\sim$ experimental time scale
- Properties depend on time since preparation: aging
- Probe with two-time correlation & response functions
- E.g. for observable O, conjugate field h:
  - Correlation (t_w = 'age', $t > t_\mathrm{w}$ measurement time) $C(t, t_\mathrm{w}) = \langle O(t)O(t_\mathrm{w})\rangle - \langle O(t)\rangle \langle O(t_\mathrm{w})\rangle$
  - Impulse response: $R(t, t_\mathrm{w}) = \delta \langle O(t)\rangle / \delta h(t_\mathrm{w})$
  - Susceptibility (step response): $\chi(t, t_\mathrm{w}) = \int_{t_\mathrm{w}}^{t} dt' \, R(t, t')$

Fluctuation-dissipation theorem and FD ratio

- In equilibrium, have FDT

$$-\frac{\partial}{\partial t_{\rm w}}\chi(t-t_{\rm w}) = R(t-t_{\rm w}) = \frac{1}{T}\frac{\partial}{\partial t_{\rm w}}C(t-t_{\rm w})$$

- Out of equil$^{\rm m}$: Define fluctuation-dissipation ratio (FDR) X

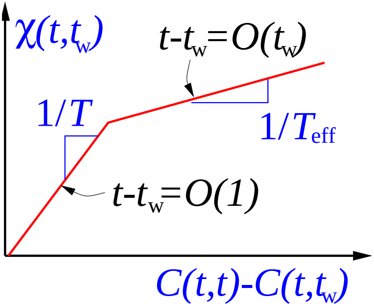
$$-\frac{\partial}{\partial t_{\rm w}}\chi(t,t_{\rm w}) = \frac{X(t,t_{\rm w})}{T}\frac{\partial}{\partial t_{\rm w}}C(t,t_{\rm w})$$

- FD plot: $\chi(t,t_{\rm w})$ vs $C(t,t)-C(t,t_{\rm w})$, with fixed t and curve parameter $t_{\rm w}$
- Equilibrium: straight line, slope $1/T$
- Out of equilibrium: slope $X/T = 1/T_{\rm eff}$?

Mean-field scenario: Effective temperature

- In mean-field spin glasses, $X(t, t_\mathrm{w}) \rightarrow$ f$^\mathrm{n}$ $X(C)$ of $C(t, t_\mathrm{w})$ for large times
- If also $C(t, t) \rightarrow$ const $= C_0$, then have limit FD plot

$$\chi(t, t_\mathrm{w}) = \frac{1}{T} \int_{C(t,t_\mathrm{w})}^{C_0} dC \, X(C)$$



- Often two straight lines; T_eff-interpretation works beautifully:
- T_eff constant within each 'time sector'
- T_eff same for many observables O
- Link to statics (configurational entropy), ...

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Non-mean-field systems

- Mean-field dynamics: no spatial information; never descend far beyond 'plateau' into minima of (free) energy landscape
- Real (short-range) glasses: dynamics are spatially heterogeneous & involve activated crossing of energy barriers
- For some systems (e.g. binary LJ & lattice glasses), FD plots nevertheless look mean-field like
- In others:
  - non-monotonic response
  - no limit FD relation for long times
  - dependence on observable ($\Rightarrow$ lengthscale), ...
- Want to understand for which systems and to what extent a mean-field T_{eff}-interpretation works

Independent motivation: Dynamical universality

- In non-equilibrium critical dynamics, FDRs have been recognized as universal amplitude ratios
- Most important is the asymptotic FDR for $t \gg t_\mathrm{w} \gg 1$

$$X^\infty = \lim_{t_\mathrm{w}\to\infty} \lim_{t\to\infty} X(t, t_\mathrm{w})$$

- Different values distinguish different dynamical universality classes
- Typically $0 < X^\infty < 1$
- Much studied recently e.g. for critical coarsening

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Why study the Fredrickson-Andersen (FA) model?

- Simple coarse-grained lattice model for ('strong') glasses
- Dynamics is activated and heterogeneous
- Good laboratory for non-mean-field effects
- At low temperatures, dynamics becomes critical: length and time scales diverge
- So we also learn something about FDRs in non-equilibrium critical dynamics
- Key result: FDRs remain well-defined, but are negative
- Should be generic for observables that couple to activated processes

FA model definition

- Binary mobility field $n_i \in \{0, 1\}$ on cubic lattice
- $n_i = 1$ models low-density, mobile regions
- High-density/immobile regions are preferred: energy $E = \sum_i n_i$
- So can think of sites with $n_i = 1$ as 'excited'
- Single-site dynamics; rates obey detailed balance w.r.t. E:

$$0 \xrightarrow{c} 1, \qquad 1 \xrightarrow{1-c} 0, \qquad c = \langle n_i \rangle = (1 + e^{1/T})^{-1}$$

- Jamming modelled by kinetic constraint: change at site i possible only if at least one excited neighbour
- Relaxation times and lengthscales diverge as Arrhenius laws ($\sim e^{A/T}$) for low T

What we want to calculate

- Correlations $C_{ij}(t, t_w) = \langle n_i(t) n_j(t_w) \rangle - \langle n_i(t) \rangle \langle n_j(t_w) \rangle$ and conjugate responses
- Fourier transform $C_{\boldsymbol{q}}$ is correlator of Fourier mode $n_{\boldsymbol{q}} = \sum_i n_i \exp(i\boldsymbol{q} \cdot \boldsymbol{r}_i)$
- Wavevector $\boldsymbol{q}$ tunes length scale probed by the observable
- Extreme cases:
  - Global ('coherent'): $C_{\boldsymbol{0}}(t, t_w)$, energy auto-correlation
  - Local ('incoherent'): $C_{ii}(t, t_w)$
- Expect to get same X^∞, though FD plots will differ (cf. critical coarsening)
- Normalize all FD plots by $C(t, t)$ to fix scale

Low-T dynamics: Intuition

- At low T, states with all excitations isolated are metastable
- Transitions between metastable states are activated processes:
- Diffusion with effective rate $D \sim c$

$$\begin{matrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{matrix} \rightarrow \begin{matrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 1 & 0 \\ 0 & 0 & 0 & 0 \end{matrix} \rightarrow \begin{matrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 \end{matrix}$$

- Coalescence, effective rate $\lambda \sim c$

$$\begin{matrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 1 \\ 0 & 0 & 0 & 0 \end{matrix} \rightarrow \begin{matrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 1 & 1 \\ 0 & 0 & 0 & 0 \end{matrix} \rightarrow \begin{matrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 1 & 0 \\ 0 & 0 & 0 & 0 \end{matrix} \rightarrow \begin{matrix} 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{matrix}$$

- Branching: reverse of annihilation, effective rate $\gamma \sim c^2$

Fast vs activated dynamics after quench

- Consider quench from $T = \infty$ to low T (low c)
- For times $t = \mathcal{O}(1)$, fast energy decrease until all excitations isolated
- Roughly similar to mean-field spin-glass dynamics: non-activated descent to energy plateau
- Further energy decrease by diffusion and coalescence requires activation: $D, \lambda, \gamma \to 0$ for $c \to 0$
- We focus on this activated long-time regime ($t, t_w \gg 1/c$)
- Branching negligible to leading order ($\gamma \sim c^2 \ll D, \lambda \sim c$)

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Field theory in $d > 2$ dimensions

- Write master operator in terms of creation and annihilation operators
- Map to (bosonic) field theory with $n_i \to (1 + \bar{\phi}(\boldsymbol{r}_i))\phi(\boldsymbol{r}_i)$
- In continuum limit $\boldsymbol{r}_i \to \boldsymbol{r}$, leading terms in action are

$$S = \int_{\boldsymbol{r},t} \bar{\phi}(\partial_t - D\nabla^2 - \gamma)\phi - \gamma\bar{\phi}^2\phi + \lambda\bar{\phi}(1 + \bar{\phi})\phi^2$$

- Use diagrammatic expansion to calculate correlation $C_{\boldsymbol{q}}$ and response $R_{\boldsymbol{q}}$ at tree level
- Should be reliable above upper critical dimension $d_c = 2$

Fourier mode correlations

- We get

$$C_{\boldsymbol{q}}(t, t_w) \approx t_w (\lambda t^2)^{-1} e^{-Dq^2(t-t_w)} f(Dq^2 t_w)$$

with $z = Dq^2 t_w$ and

$$f(z) = \begin{cases} (1+z)/3 & \text{for} \quad z \ll 1 \\ 1 - 1/z & \text{for} \quad z \gg 1 \end{cases}$$

- Characteristic length scale $q^{-1} \sim \xi(t_w) = (Dt_w)^{1/2}$: controlled by diffusion
- Energy auto-correlation ($\boldsymbol{q} = \mathbf{0}$): $C_{\mathbf{0}}(t, t_w) \approx n(t) t_w / 3t$
- $n(t) = \langle n_i(t) \rangle \approx (\lambda t)^{-1}$ is mean excitation density ($= E/N$); compare 'classical' rate equation $\partial_t n = -\lambda n^2$

Fourier mode response

- Needs some care: field at site i affects rate of diffusion to rather than from site i
- We get

$$R_{\boldsymbol{q}}(t, t_w) \approx (\lambda t^2)^{-1} e^{-Dq^2(t-t_w)}(z-1)$$

- Energy response: $R_{\mathbf{0}}(t, t_w) \approx -n(t)/t$;
  susceptibility $\chi_{\mathbf{0}}(t, t_w) \approx -n(t)(1 - t_w/t)$
- Why are these negative?
- Increase in T accelerates dynamics because of activation $\Rightarrow$ faster decay of energy $\Rightarrow$ negative response. Generic (!)
- Small initial 'bump' ($\chi > 0$) becomes negligible for small c

Fourier mode FDR

- Combine results for correlation and response:

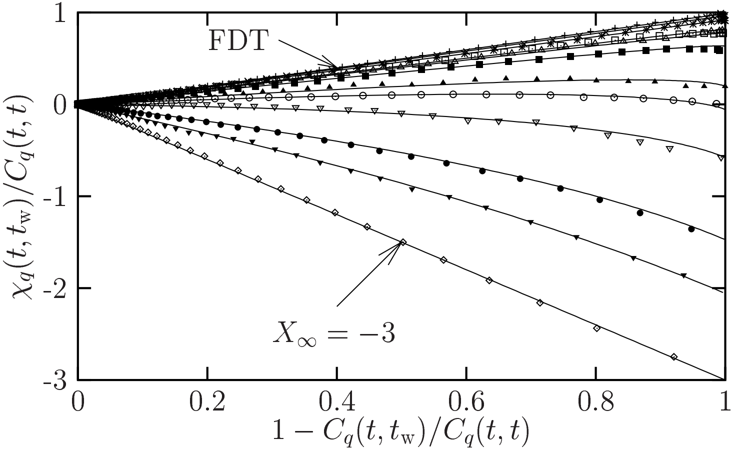
$$X_{\boldsymbol{q}}(t, t_{\rm w}) = \frac{T R_{\boldsymbol{q}}(t, t_{\rm w})}{\partial_{t_{\rm w}} C_{\boldsymbol{q}}(t, t_{\rm w})} \approx \frac{z - 1}{(1 + \partial_z) z f(z)} \approx \begin{cases} 1 - 1/z & (z \gg 1) \\ -3 + 12z & (z \ll 1) \end{cases}$$

- Short length scales $q^{-1} \ll \xi(t_{\rm w})$ are equilibrated ($z = Dq^2 t_{\rm w} \gg 1$)
- Longer length scales remain out of equilibrium ($z = \mathcal{O}(1)$)
- For the energy ($z = 0$) get

$$X^\infty = X_{\boldsymbol{0}}(t, t_{\rm w}) = -3$$

- Constant $\Rightarrow$ straight-line FD plot, as for mean-field systems
- But X^∞ is negative (!)

FD plots: Theory vs simulation in $d = 3$



q decreasing top to bottom, $T = 0.1$, $t = 2 \times 10^5$ fixed

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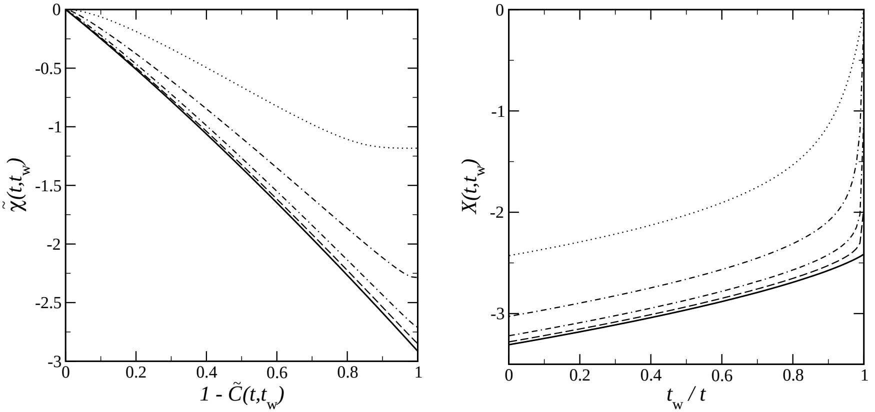
How do we deal with fluctuation effects in $d = 1$?

Mapping to Glauber-Ising chain

- In $d = 1$, classical/tree level calculation no longer works
- Need to account for fluctuations in local excitation density
- Fortunately, we can find an exact mapping of the master operator to a solvable model: the Glauber-Ising spin chain
- Empty intervals in the FA model map to intervals with an even number of domain walls in the spin chain
- Correlation and response functions don't map directly, but can still be worked out for local/global case
- Get exact results in the long-time, far-from-equilibrium regime

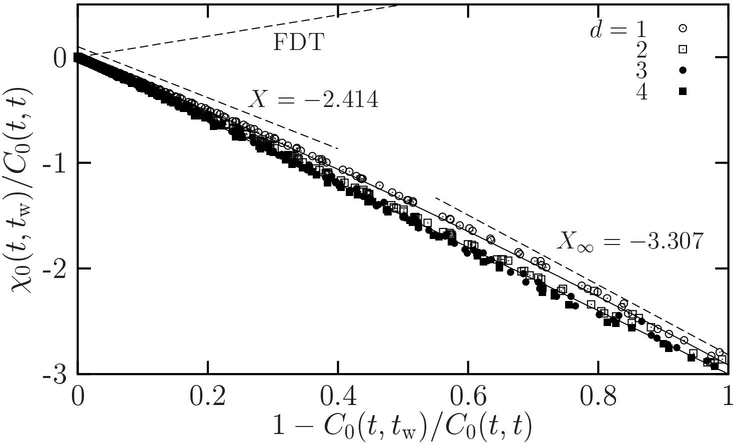
Energy FD relation



- FD plot slightly curved but χ is negative as before
- $X^\infty = -3\pi/(16-6\pi) = -3.307\ldots$

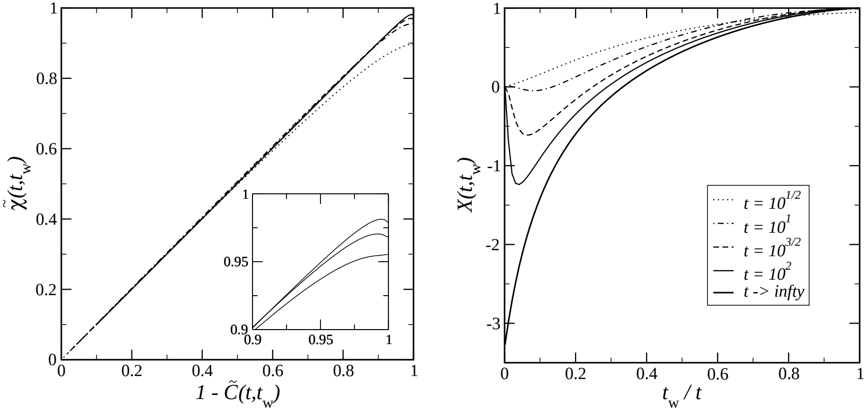
Energy FD relation

Comparison with simulations



- Good agreement for all d
- χ measured without field from special correlation function

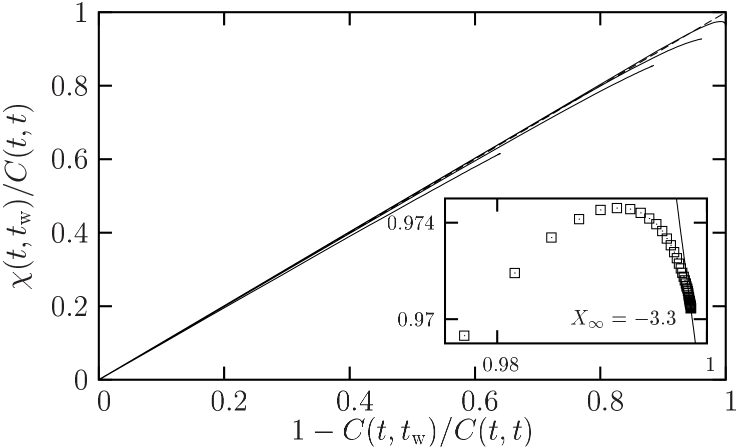
Local FD relation



- FDR crosses over from 1 to same X^∞, but this is hidden in long-time FD plot in a small region $\sim 1/\sqrt{t}$
- X^∞ much easier to measure from global quantity (E)

Local FD relation

Comparison with simulations



- Good agreement of asymptotic slope with predicted X^∞

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Why is $d_c = 2$?

- Power-counting on field-theoretic action S suggests $d_c = 4$
- Renormalization group equations look like directed percolation
- But our results for X^∞ suggest $d_c = 2$. Why?
- Can extend our mapping to general d
- Exact relation between 'reaction-diffusion models' $A + A \leftrightarrow A$ (FA) and $A + A \leftrightarrow \emptyset$ (pair annihilat^n & creation)
- But $A + A \leftrightarrow \emptyset$ has a parity symmetry (number of particles either odd or even)
- FA model inherits this as a hidden symmetry
- When action is written to reveal this, see that indeed $d_c = 2$

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Summary

- Some mean-field concepts carry over to activated dynamics: Time sectors, well-defined FDRs
- But response functions and FDRs are negative
- Effective temperature interpretation unlikely
- First negative asymptotic FDRs in critical dynamics
- Conclusion should apply generically to dynamics of observables controlled by activated processes
- Activation doesn't have to be thermal: e.g. tapping in granulars (Depken & Stinchcombe)

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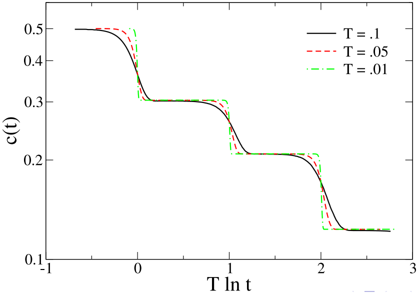
Cooperative/fragile glasses

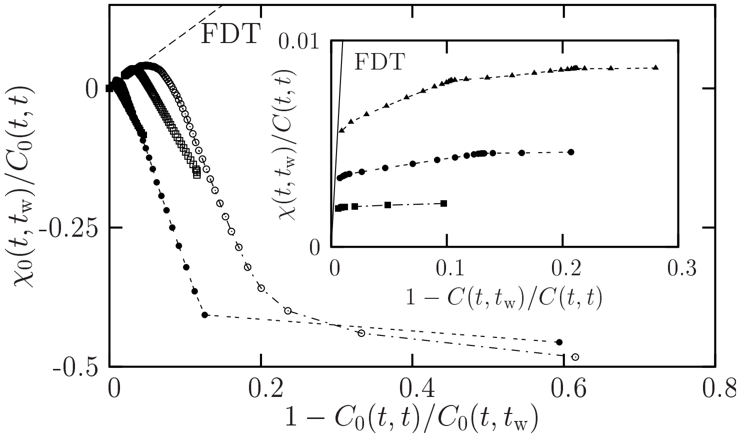
East model

- FA model is for strong glasses
- Contrast with fragile glasses: more cooperative dynamics, much stronger time scale divergence at low T
- Simplest test case: East model ($\tau \sim e^{A/T^2}$)
- Like FA in $d = 1$, but need an excited neighbour on left
- Produces hierarchy of timescales, dynamics in stages



FD relations

Simulations for C, exact relations for χ



Global (main graph): χ again negative after initial bump

Local: looks like well-defined FDRs for different stages ($X^\infty = 0$)