Glassy behaviour due to kinetic constraints

by

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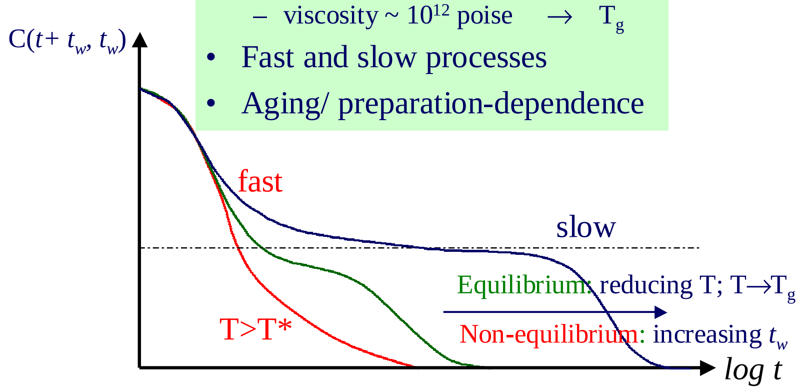
(with T.Aste, A.Buhot, L.Davison, J.Garrahan)

Outline

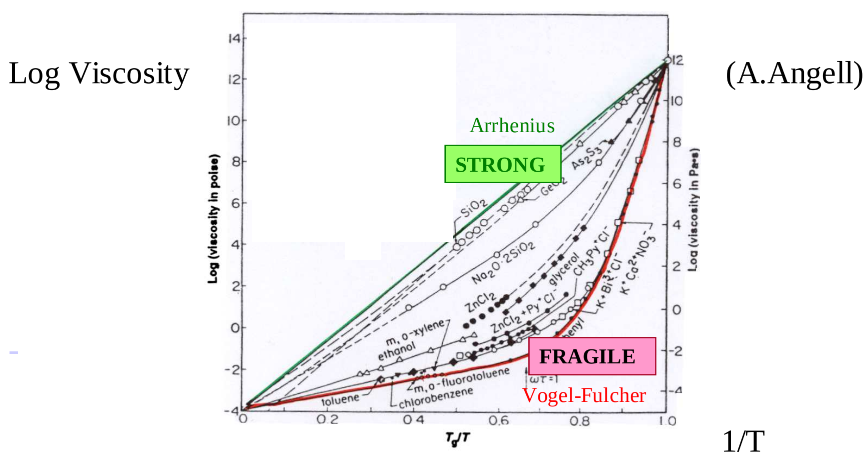
- *Introduction* (to glasses)
- *Minimalist topological model*
  - foams & covalent glasses
  - non-interacting Hamiltonian, constrained dynamics
  - ⇒ glassiness, two-time dynamics
- *Annihilation-diffusion*
- *Lattice analogues*
  - Different absorbing ground states
    - zero degeneracy
    - high degeneracy
- *Ultimate distillation?*
  - Simple strong glass
    - characteristic features
    - mean-field soluble with activation

Glasses

- Non-periodic 'freezing'
  - viscosity ~ 10^{12} poise → T_g
- Fast and slow processes
- Aging/ preparation-dependence



Structural glasses



Structural glasses

Strong: e.g. silica

covalent, strong directional forces

Fragile e.g.argon

weaker, central (non-directional) forces: Lennard-Jones

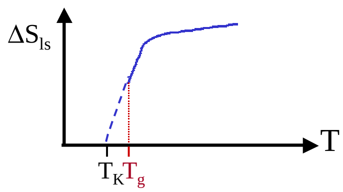
Usual models and systems

- Interacting ‘particles’, simple dynamical moves
  - Spin glasses: quenched disorder
  - Structural glasses: no imposed disorder
    - glassiness self-induced
    - analogies of fragile glasses with D1RSB spin glasses

Fragile glasses/D1RSB

Fragile structural glasses

T_K ~ Kauzmann temperature



T_g ~ Dynamical glass temp.

(viscosity ~ 10^{13} poise)

T* ~ Response plateau

D1RSB spin glass

T_K ~ Thermodynamic trans^n

T_g ~ Dynamical transition

T* ~ Correlation plateau

(& configurational entropy)

Soluble models

Self-consistent theory

Simulations

Correlation functions

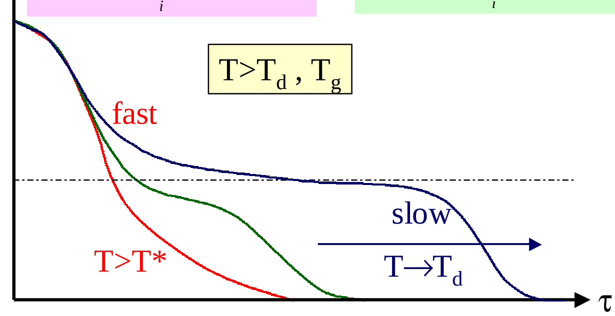
Equilibrium

p-spin glass

$$C(\tau) = N^{-1}\sum_i S_i(t+\tau)S_i(t)$$

Structural glass

$$C_k(\tau) = N^{-1}\sum_i \exp(ik.(R_i(t+\tau) - R_i(t)))$$

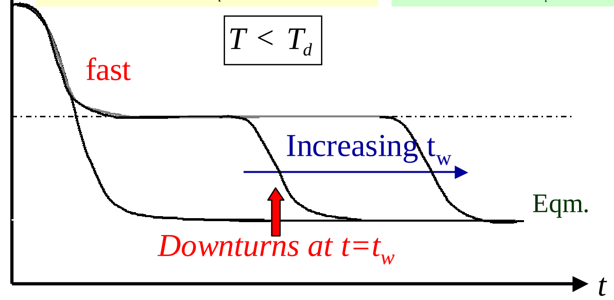


Correlation functions

Non-equilibrium

$C(t+t_w,t_w)=N^{-1}\sum_i S_i(t+t_w)S_i(t_w)$

$C(t+t_w,t_w)=N^{-1}\sum_i \exp(ik(R_i(t+t_w)-R_i(t_w)))$



Models to discuss today

Trivial thermodynamics

but

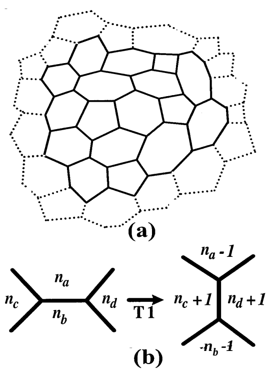
Non-trivial dynamics

due to

kinetic constraints

Topological 'foam'

Minimalist topological model



$$E = \sum_i (n_i - 6)^2$$

Different from usual foam

Glauber-Kawasaki T1 dynamics

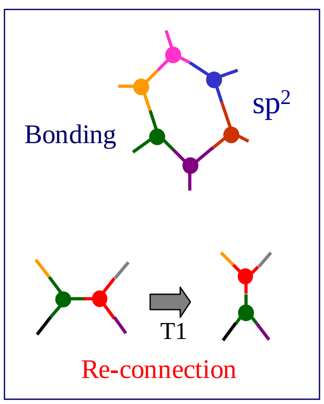
Prob. ~ exp($-\Delta E/T$)

Euler : $\langle n \rangle = 6$

Ground state: hexagonal

Aste, Davison, Sherrington

Covalently bonded glasses



Euler: $\langle n \rangle = 6$

Two dimensions

(for simplicity)

Preferred angle at vertex = 120° = $2p/3$

Preferred crystal: hexagonal

Re-connections?

⇓

Randomly connected network liquid/ glass

Distorted bonds

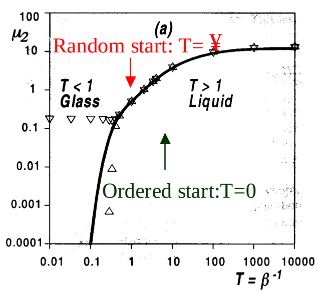
Energy of deviation ~ $(q - 2p/3)^2$

n-sided polygon →

E ~ $(n-6)^2/(6n)^2$

Results for topological model

Energy: different starts

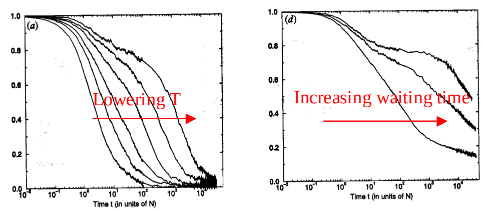


Aste & S

Temporal autocorrelation functions

Equilibrium

Non-equilibrium



Davison & S

Theoretical understanding

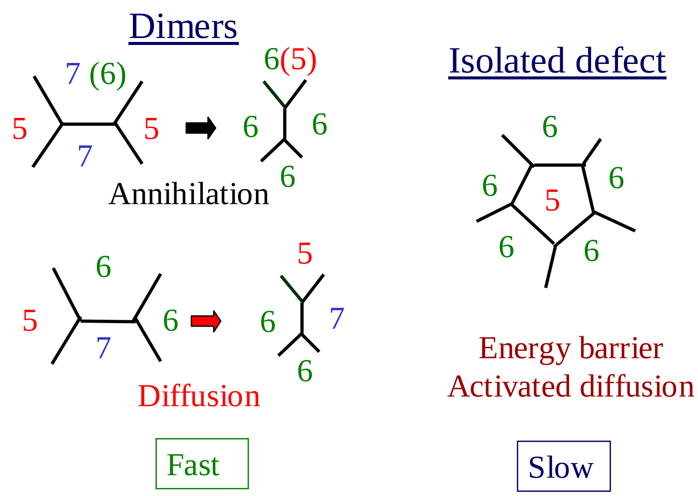
Diffusion & Annihilation

$A+B \rightarrow 0$

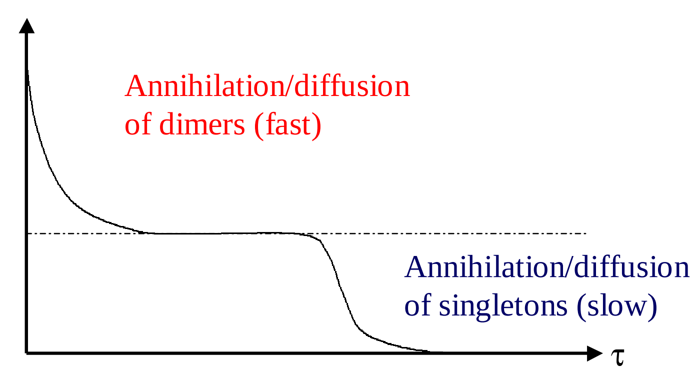
Several types of ‘particle’ (A, B)

Some: Fast T-independent diffusion
Others: Slow T-dependent diffusion

Annihilation-diffusion

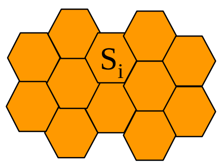


Energy/ correlation function



Lattice-based analogue

Hexagonal lattice

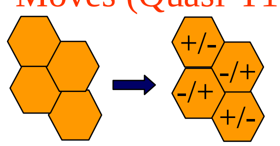


'Spins': $S_i = 1, 0, -1$

Energy: $E = D\sum_i S_i^2$

Conservation: $\sum_i S_i = 0$

Moves (Quasi-T1)



Equal prob.

Dynamics: Metropolis-Kawasaki

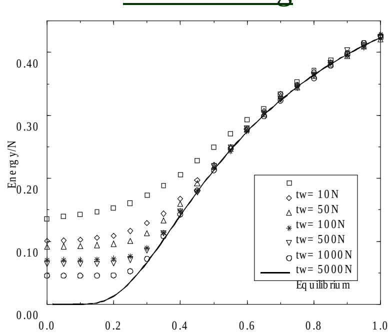
D>0: unique g.s., defects ±1

D<0: degenerate g.s., defects 0

Energy

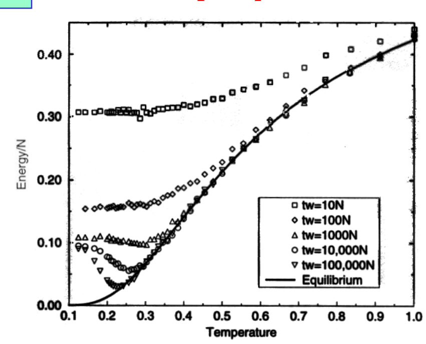
D>0

Slow cooling



(t_W= time at each temperature)

Rapid quench



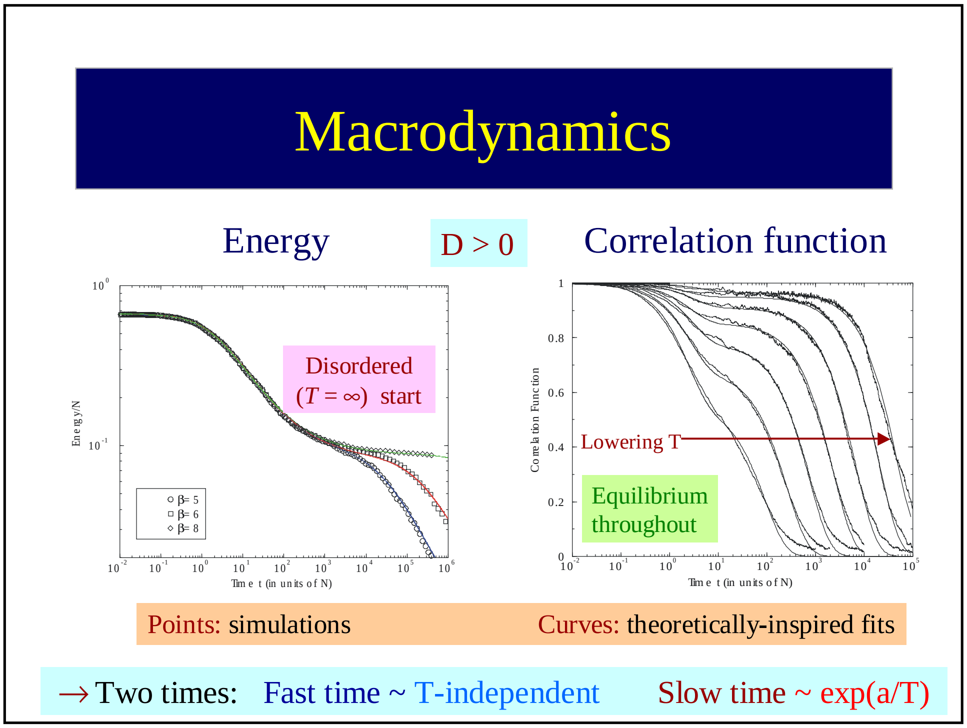
(t_W= time subsequent to quench)

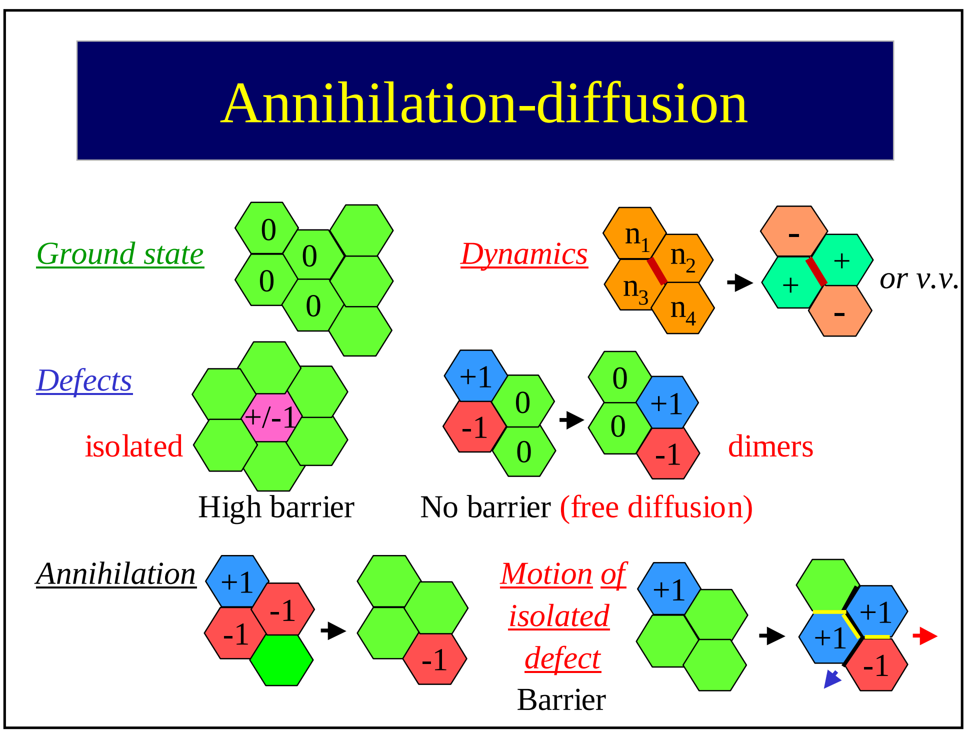
Curves = equilibrium; calculation easy since non-interacting

Falls out of equilibrium

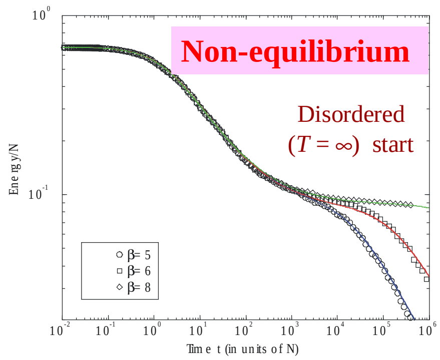
Davison & S

Activation barriers impenetrable at T=0


Macrodynamics
Energy
D > 0
Correlation function
Disordered
(T = ∞) start
Energy/N
Time t (in units of N)
β= 5
β= 6
β= 8
Correlation Function
Lowering T
Equilibrium
throughout
Time t (in units of N)
Points: simulations
Curves: theoretically-inspired fits
→ Two times: Fast time ~ T-independent Slow time ~ exp(a/T)


Annihilation-diffusion
Ground state
Dynamics
or v.v.
Defects
isolated
High barrier
No barrier (free diffusion)
dimers
Annihilation
Motion of
isolated
defect
Barrier

Energy (D>0)



Plateau: fast annihilation complete
slow essentially unstarted

- Ground state unique: {S} = {0}
- Defects: ± 1

- Energy reduces through dimer annihilation
- **Dimers** move freely → **fast** time ~ τ (=2)

$$A+A+B \to A\,;\ \sim t^{-0.5}$$

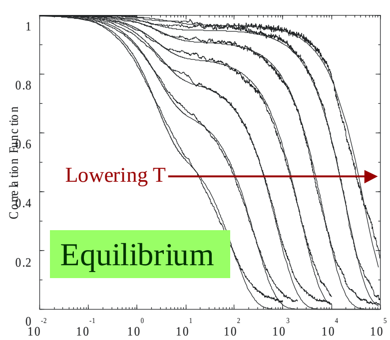
- **Single defects** need to pair to form dimers which can move and annihilate
- **Slow** diffusion
- Energy barriers→τ~exp(Δ/T); Arrhenius

$$A+B \to 0\,;\ \sim (t/\tau)^{-d/4}$$

$$E(t) = (2/3-a)(1+t/2)^{-b} + (a-e_{eq})(1+t/e^{2\beta})^{-c} + e_{eq}; b \sim .5, c \sim .5$$

Auto-correlation function

D > 0



$$C(t) = \sum_i S_i(t+t_w)S_i(t_w) / \sum_i S(t_w)^2$$

One move changes C(t) → exp.

fast dimers: τ~2
slow singletons: τ~2exp(Δ/T)

$$C(t) = \alpha e^{-t/\tau_1} + (1-\alpha)e^{-t/\tau_2}; \tau_1 = 2, \tau_2 \sim 2e^{-2.12\beta}$$

D < O

$$H = D\sum_i S_i^2; \quad S_i = 0, \pm 1; \quad \sum_i S_i = 0$$

Highly degenerate ground state: $\{S_i = \pm 1\}$

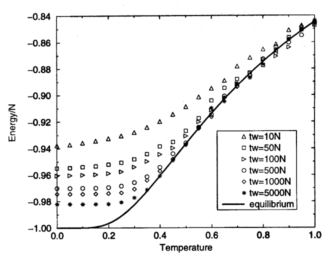
Single defect type: 0

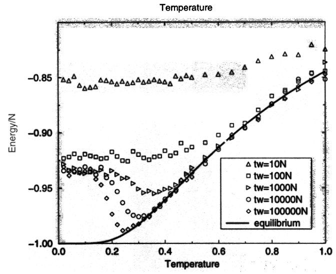
Single dimer type: (0,0):

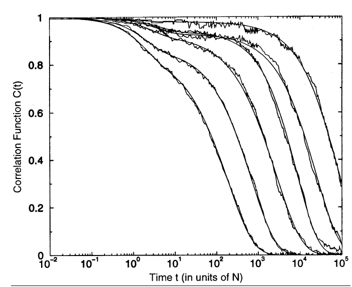
A+A+A→A   A+A→Ø

dimer diffusion can be blocked by disadvantageous environment

D < 0 results







Qualitatively similar to D>0

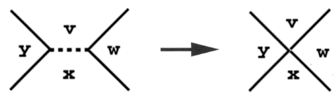
but

different exponents

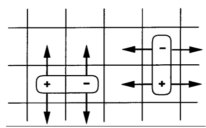
and some

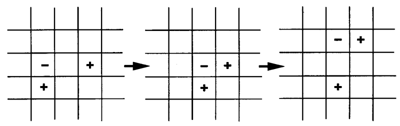
stretched exponential character

Square lattice

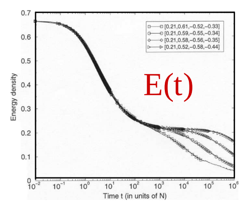


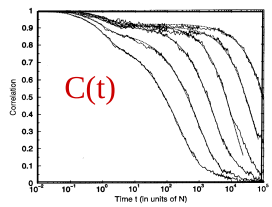
Dimer moves





Singleton move





Summary of processes

- Dimer annihilation:

$$2A+\overline{A}+\phi \rightarrow 3\phi + A$$
$$2\overline{A}+A+\phi \rightarrow 3\phi + \overline{A}$$
$$(2A+2\overline{A} \rightarrow 4\phi)$$

All involve 4 neighbours

- Dimer diffusion: $A+\overline{A}+2\phi \rightarrow 2\phi + A + \overline{A}$
- Defect movement via dimer creation

$$A+3\phi \rightarrow \phi + 2A+\overline{A}$$
$$\overline{A}+3\phi \rightarrow \phi + 2\overline{A}+A$$

A simpler encapsulation?

- Desired features
  - fast annihilation of dimers
  - fast diffusion of dimers
  - hindered motion of isolated defects
  - all only with appropriate environments
    - ‘4-changes’
  - non-degenerate/ absorbing ground states
  - single defect type (A) or two types (A,B)

Constrained ‘backgammon’

- Non-interacting ‘particles’: $H = \sum_{i=1}^{N} n_i$ $n_i \le 3$
  - Trivial equilibrium, unique absorbing g.s.
- Constrained dynamics
  - Annihilation: analogue of dimer annihilation against defect;

    $(n_i, n_j) \to (n_i - 3, n_j + 1)$ *Rate =1*
  - Diffusion: analogue of dimer diffusion

    $(n_i, n_j) \to (n_i - 2, n_j + 2)$ *Rate = D*
  - Creation: analogue of defect motion by dimer creation

    $(n_i, n_j) \to (n_i - 1, n_j + 3)$ *Rate =* $e^{-2\beta}$

Philosophy: follow number of A

- Dimer annihilation:

$$2A+\bar{A}+\phi \to 3\phi + A$$
$$2\bar{A}+A+\phi \to 3\phi + \bar{A}$$
$$\Rightarrow \quad (n_i, n_j) \to (n_i - 3, n_j + 1)$$

- Dimer diffusion:

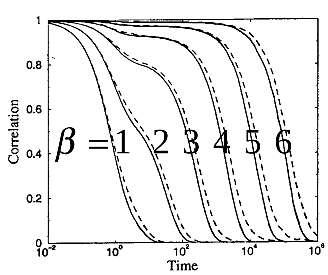
$$A+\bar{A}+2\phi \to 2\phi + A + \bar{A} \quad \Rightarrow \quad (n_i, n_j) \to (n_i - 2, n_j + 2)$$

- Defect movement via dimer creation

$$A+3\phi \to \phi + 2A + \bar{A}$$
$$\bar{A}+3\phi \to \phi + 2\bar{A} + A$$
$$\Rightarrow \quad (n_i, n_j) \to (n_i - 1, n_j + 3)$$

Simulations

Correlation function

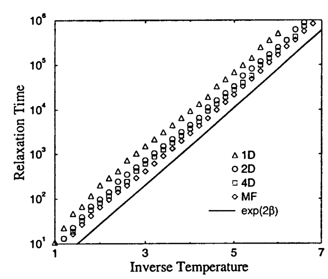


$d = 2$ (dashed), $d = \infty$ (solid)

$C^c_{eq}(t)/C^c_{eq}(0)$; $C(t,t') = \langle n_i(t) n_i(t') \rangle$

$C^c_{eq}(t) = C_{eq}(t) - c^2_{eq}$; $c_{eq} = \langle n_i(\infty) \rangle$

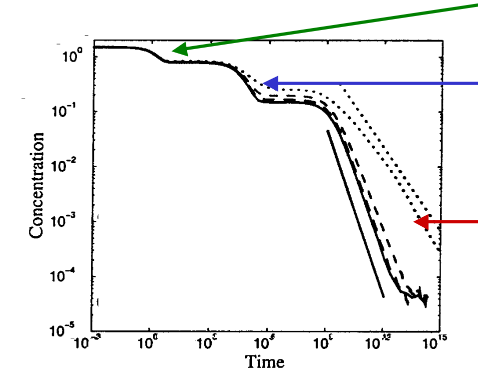
Arrhenius decay



$C^c_{eq}(\tau) = C^c_{eq}(0)/\tau$; $t = 0 \sim T = \infty$

Energy/particle number decay

Single type of particle



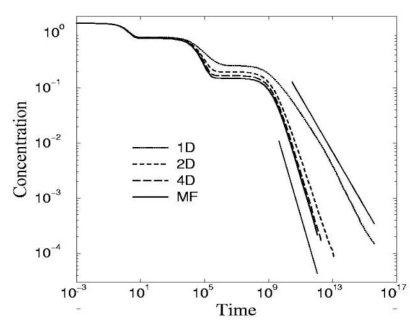
Annihilation without diffusion
T=0

Dimer diffusion and annihilation
T=0

Activated regime
$A + A \to \phi$
$\alpha = d/2;\ d < d_c = 2$

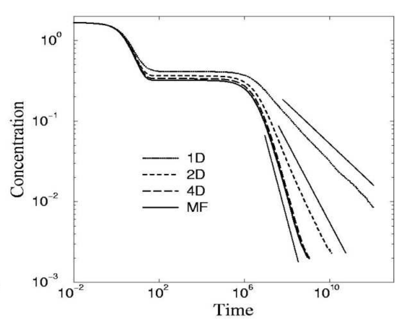
Energy (particle number) decay

Single type of particle



Final decay: $A + A \to \phi$
$\alpha = d/2;\ d_c = 2$

Two types of particle

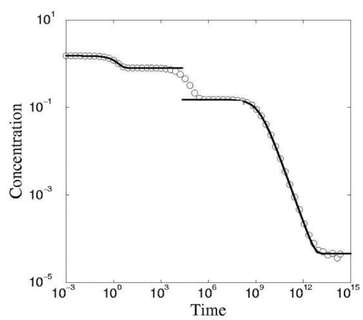


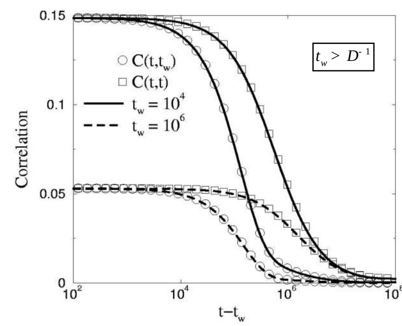
Final decay: $A + B \to \phi$
$\alpha = d/4;\ d_c = 4$

Theory & simulation (infinite d)

Concentration decay after quench

Out of equilibrium correlation & concentration





Circles ~ simulation, lines ~ theory

Circles ~ correlation, squares ~ concentration

Effective temperature: fragile glass

Fluctuation-dissipation ratio

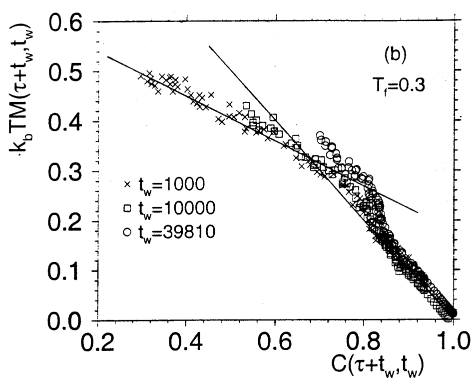
Response/ Correlation= $-T_{eff.}^{-1}$

Equilibrium: $T_{effective} = T$

Away from equilibrium

Short-time: $T_{effective} = T$

Long-time: $T_{effective} > T$

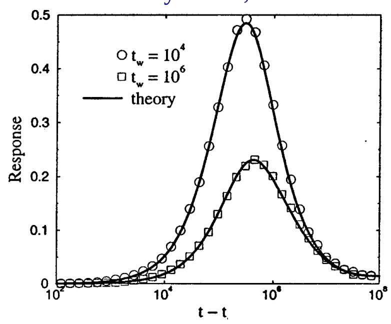


Lennard-Jones glass: Kob & Barrat

Response & FDT

Out of equilibrium response

MM dynamics; d=∞



$t_w > \Delta^{-1}$

$$\delta H = -h \sum_i \varepsilon_i \delta_{n_i,1}$$

$$c_1(t,t_w) = \overline{h^{-1} N^{-1} \sum_i \varepsilon_i \langle \delta_{n_i(t),1} \rangle_h}$$

Non-monotonic

due to

Alignment with perturbation field

&

Annihilation

Good agreement with theory

Also good for M dynamics

Dynamics; M or MM

- Metropolis:

  Prob of acceptance $= \min(1, e^{-\beta \Delta H})$; ΔH = change in energy

- Response perturbation: $\delta H = -h \sum_i \varepsilon_i \delta_{n_i,1}$

- Metropolis (M): $P = \min(1, e^{-\beta \Delta (H + \delta H)})$
- Modified Metropolis (MM):

  $P = \min(1, e^{-\beta \Delta \delta H}) \times \min(1, e^{-\beta \Delta H})$

No difference in results for conventional problems, but MM preferred at low T.

Fluctuation-dissipation

Two different types of dynamics:

Metropolis (M) and modified Metropolis (MM)

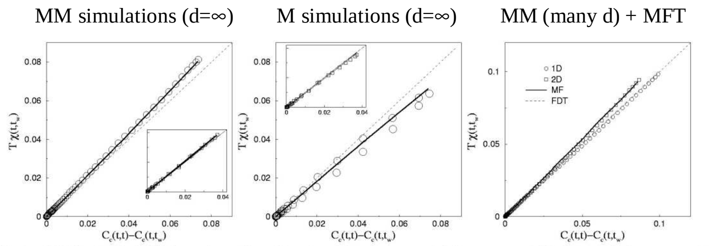


FIG. 9. (a) FD plot for MM dynamics at $T = 1/6$ and waiting time $t_w = 10^4$ (inset: $t_w = 10^6$). The symbols correspond to simulations, the full lines to the analytical result, and the dashed line to FDT. (b) Similar plot for M dynamics. (c) FD plots in various dimensions (for MM dynamics): $d = 1$ (circles), $d = 2$ (squares), and MF (full line); $T = 1/6$, $t_w = 10^4$.

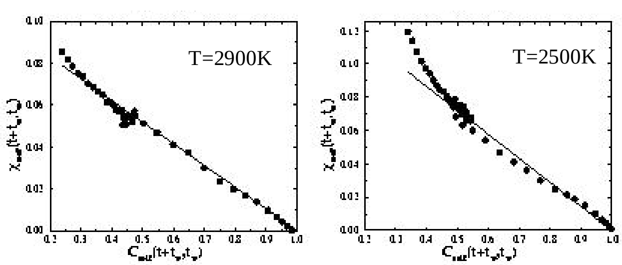
Different from usual fragile glass

No sharp change of T_{eff} at critical correlation, $T_{eff} < T$ for MM dynamics, $T_{eff} > T$ for M dynamics

Simulations of model strong liquid

Scala et. al. cond-mat/0301143

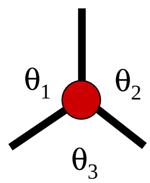
BKS-silica



$T_{eff} < T$ at long times

Static phase transition?

Include correlation energies in Hamiltonian



$l\ f\{(q_1 + q_2 + q_3 - 2p)^2\}$

or add constraints

Speculation

Origin of apparent spin glass behaviour in topologically frustrated magnets beneath effective temperature?

$\chi_{FC} \neq \chi_{ZFC}$

AF interacting spins on frustrated lattice

⇓

Effective simple ‘spins’ with constrained dynamics

Conclusions

- Kinetic constraints can cause glassy dynamics
  - even with non-interacting Hamiltonian
  - and trivial thermodynamics
- Can yield strong glass Arrhenius behaviour
  - several simple models
    - topological foams, idealized covalency
    - constrained spins, multi-spin flips
    - ‘backgammon’ with energetic rather than entropic barriers
      - soluble and significant in mean field limit
- FDT ~ simulations, strong different from fragile.

References

Recent review:

F.Ritort & P.Sollich, cond-mat/0210382

“Glassy dynamics of kinetically constrained models”

(to be published in “Advances in Physics”)

My relevant research papers

T.Aste & D.S., J.Phys.A32, 7049 (1999)

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A.Buhot et. al., J.Phys.A36, 307 (2003)