Glassiness due to constrained dynamics: from topological foam to backgammon

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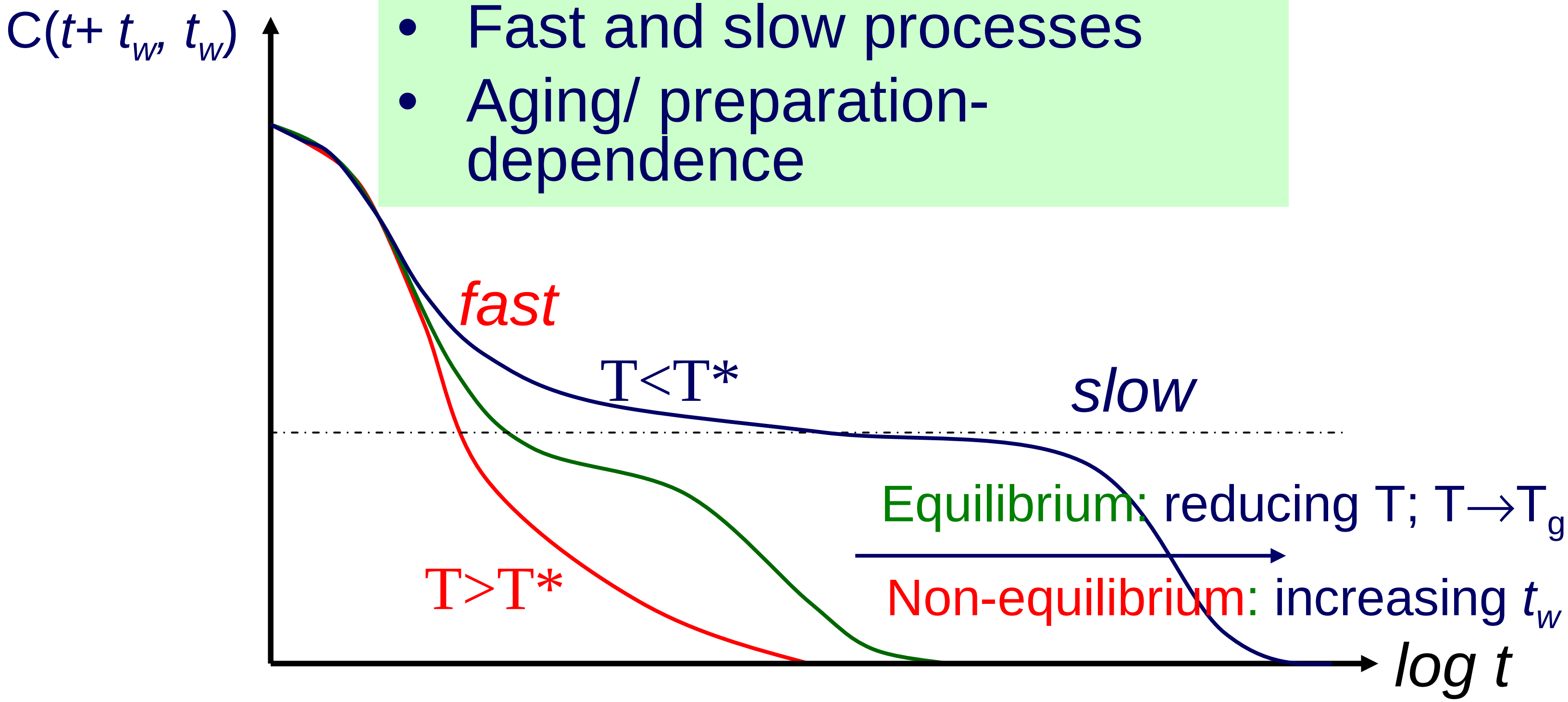
Peking University, September 2007

Outline

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

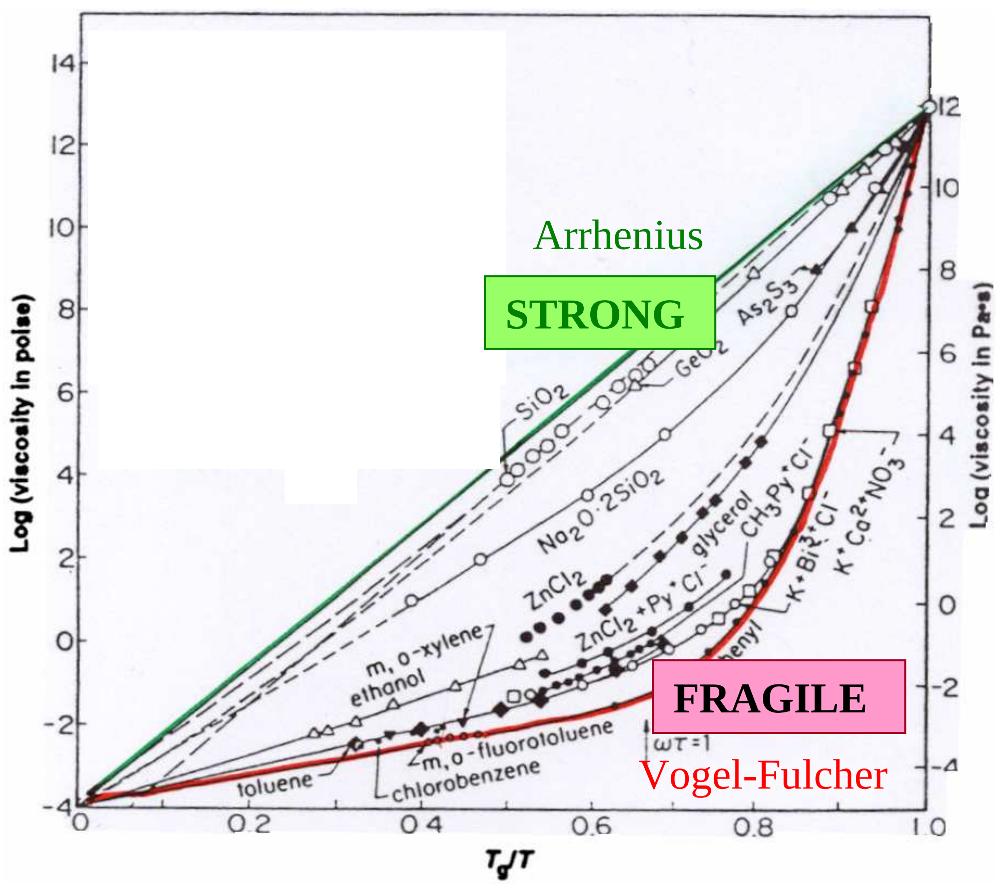
Glasses

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



Structural glasses

Log Viscosity



(A.Angell)

1/T

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

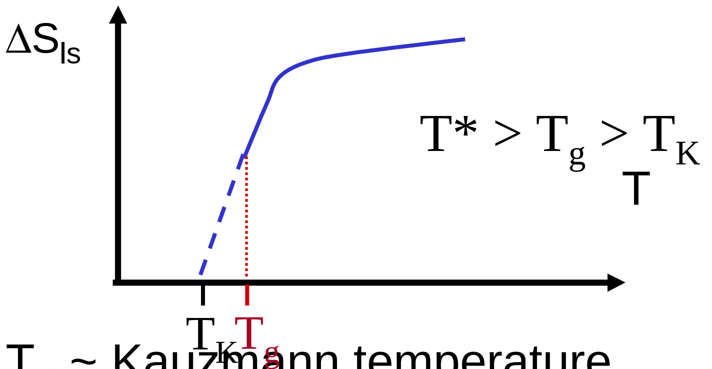
- Interacting ‘particles’, simple dynamical moves

  – *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 transⁿ
T_g ~ Dynamical transition
T^* ~ Correlation plateau
(onset of extensive config. entropy)

Soluble models (range-free)
Self-consistent theory

Simulations

Main models to discuss today

Trivial thermodynamics

but

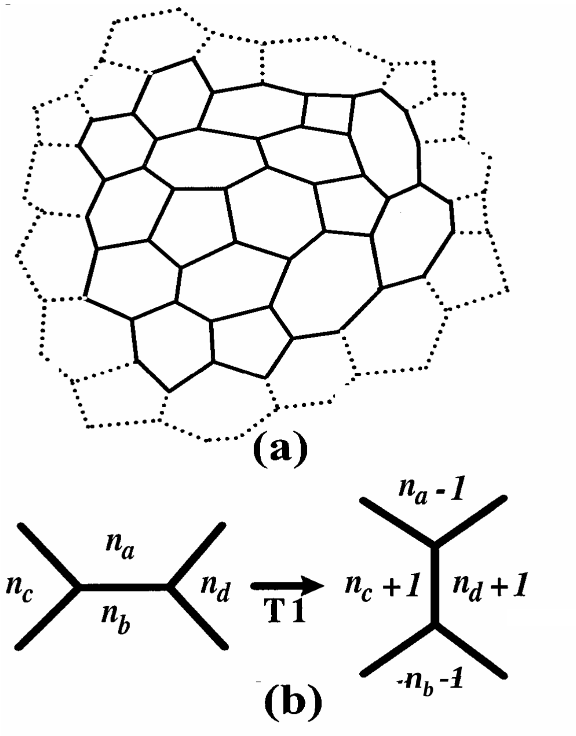
Non-trivial dynamics

due to

kinetic constraints

Topological 'foam'

Minimalist topological model



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

Different from usual foam

'Glauber-Kawasaki' T1 dynamics

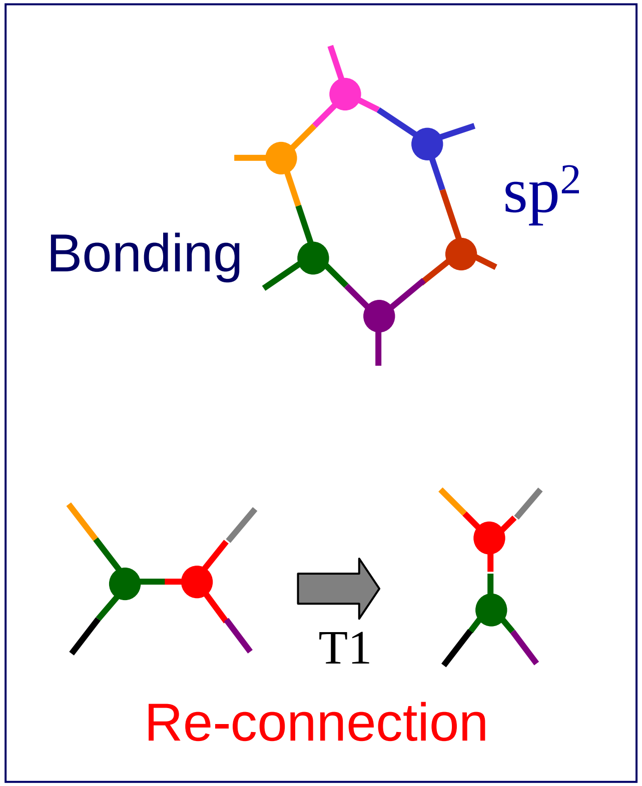
Prob. ~ exp(-$\Delta E/T$)

Euler : $<n> = 6$

Ground state: hexagonal

Aste & Sherrington

Covalently bonded glasses



Euler: <n> = 6

Two dimensions

(for simplicity)

Preferred angle at vertex = 120° = $2\pi/3$

Preferred crystal: hexagonal

Re-connections?

⇓

Randomly connected network liquid/ glass

Distorted bonds

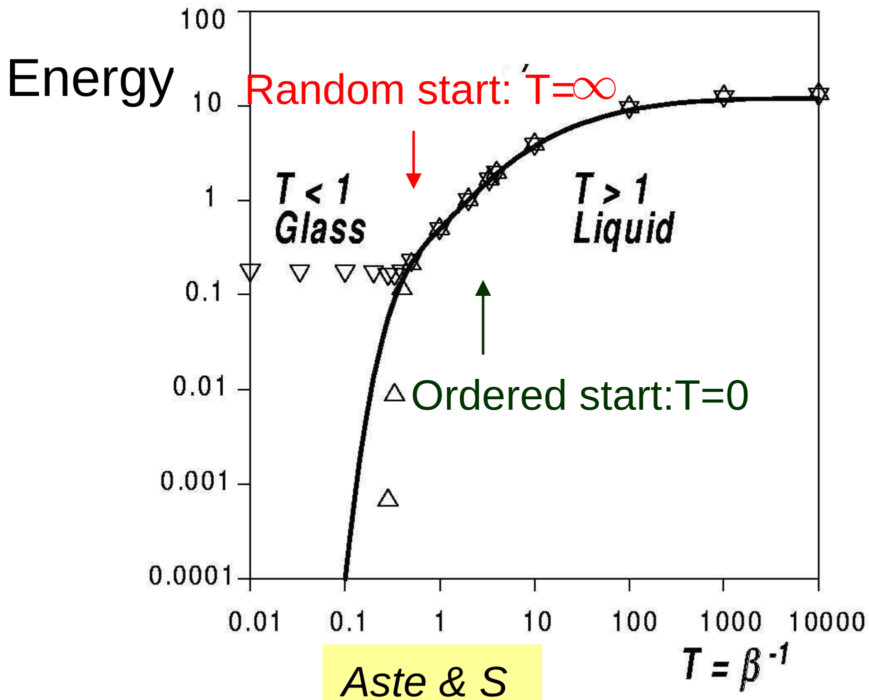
Energy of deviation ~ $(\theta - 2\pi/3)^2$

n-sided polygon

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

Results for topological model

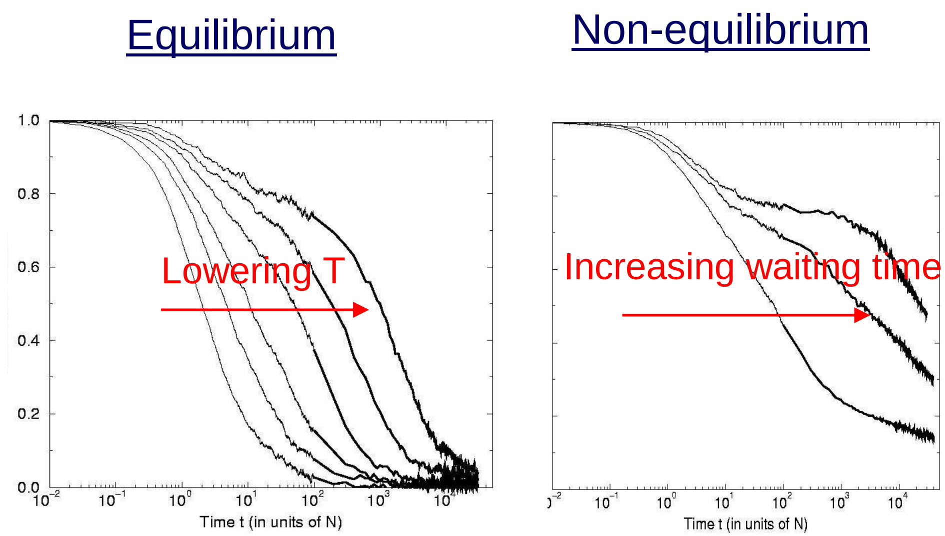
Energy: different starts



Aste & S

1100*N* timesteps

Temporal autocorrelation fns



Davison & S

$\beta = 1, 2, 2.5, 3, 3.5, 4$

$\beta = 6; \tau = 10^2 N, 10^3 N, 10^4 N$

Theoretical understanding

Diffusion & Annihilation

$$A+B\rightarrow 0$$

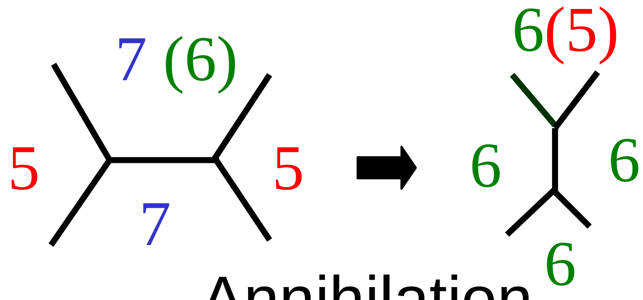
Several types of 'particle' (A, B)

Some: Fast T-independent diffusion

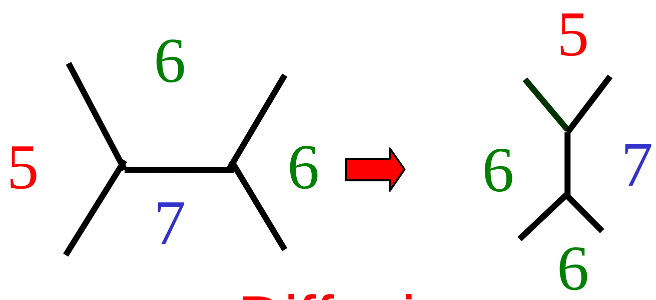
Others: Slow T-dependent diffusion

Annihilation-diffusion

5-7 Dimers



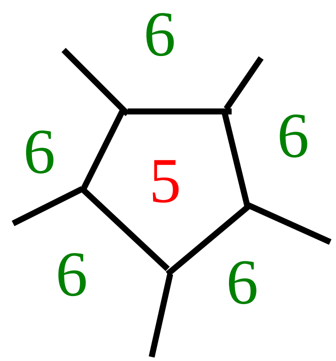
Annihilation



Diffusion

Fast

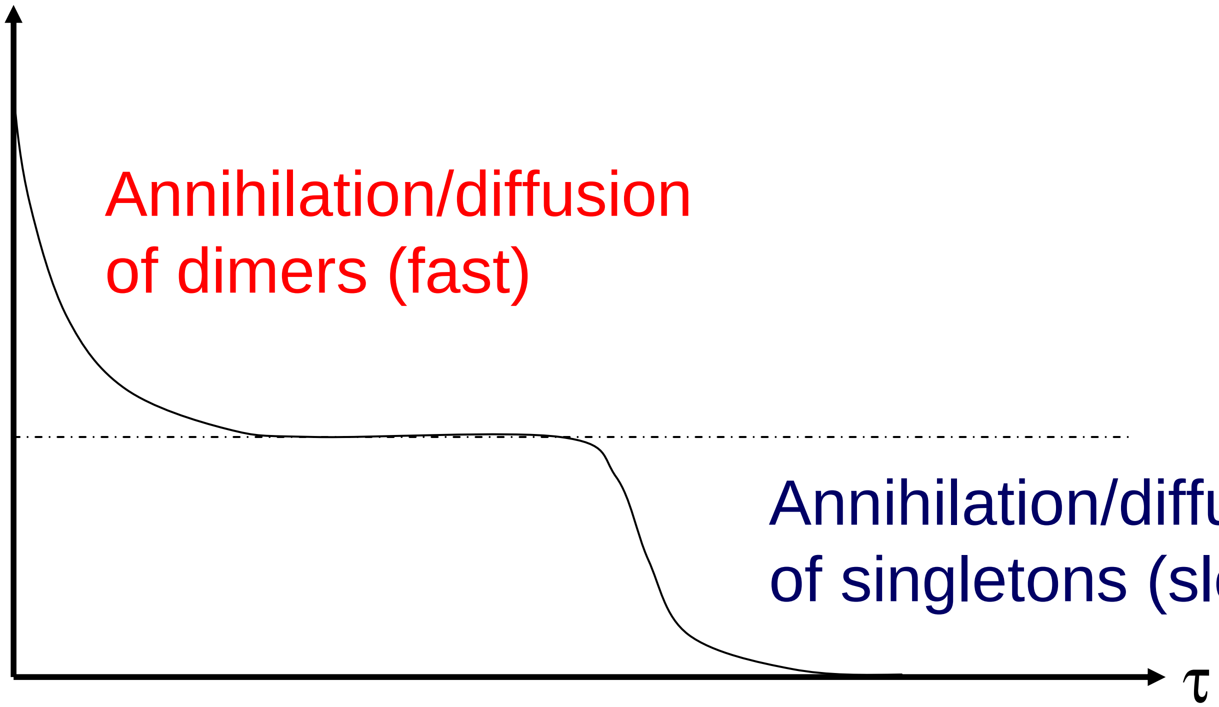
Isolated defect



Energy barrier
Activated diffusion

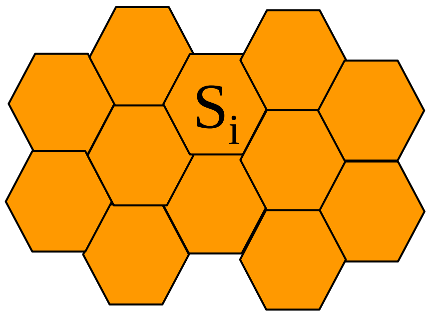
Slow

Energy or correlation function



Lattice-based analogue

Hexagonal lattice

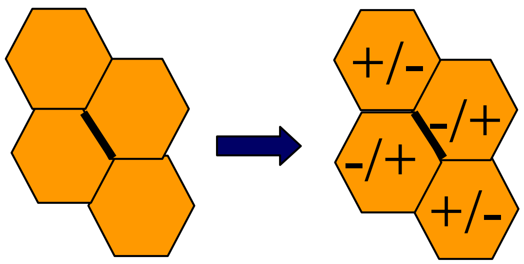


‘Spins’: S_i = 1, 0, -1

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

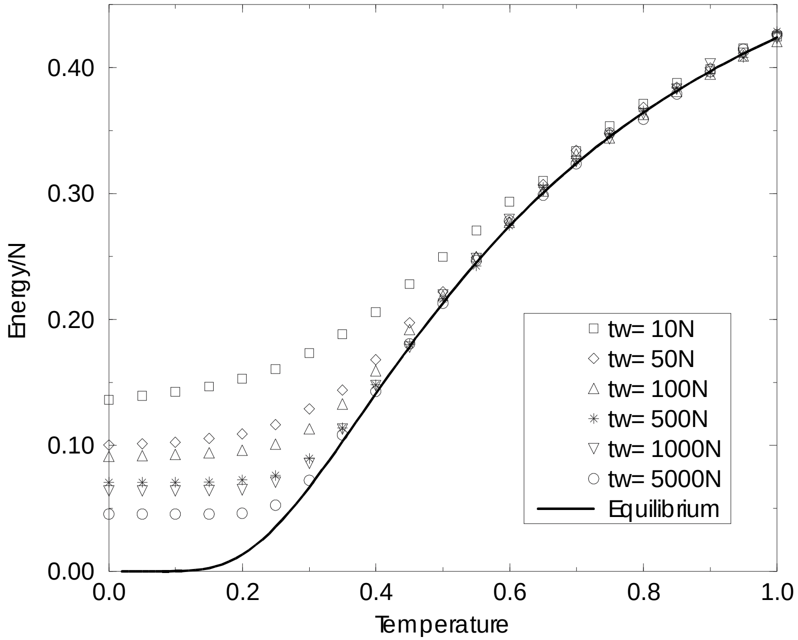
D<0: degenerate g.s., defects 0

Energy

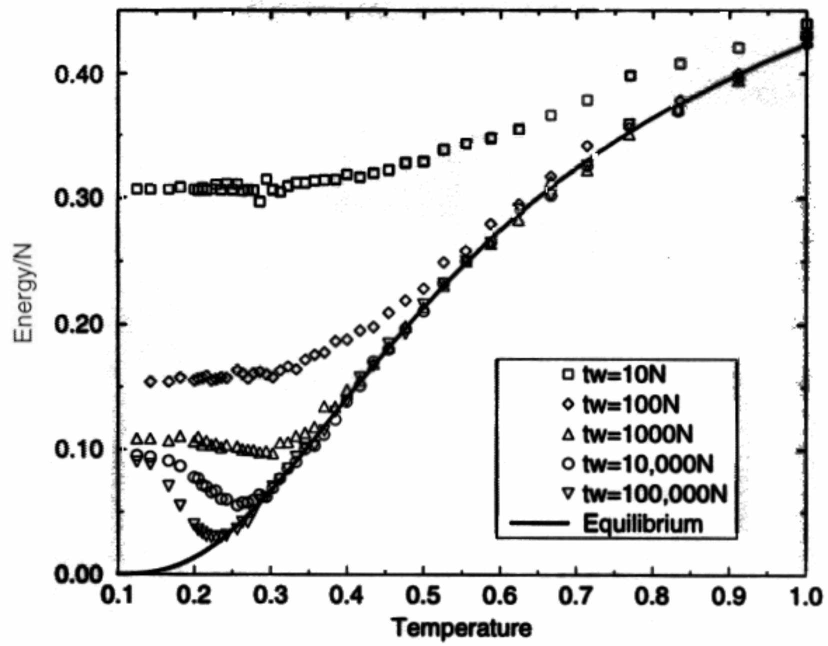
Slow cooling

D>0

Rapid quench



(t_W= time at each temperature)



(t_W= time following quench)

Curves = equilibrium; calculation easy since non-interacting

Falls out of equilibrium

Davison & S

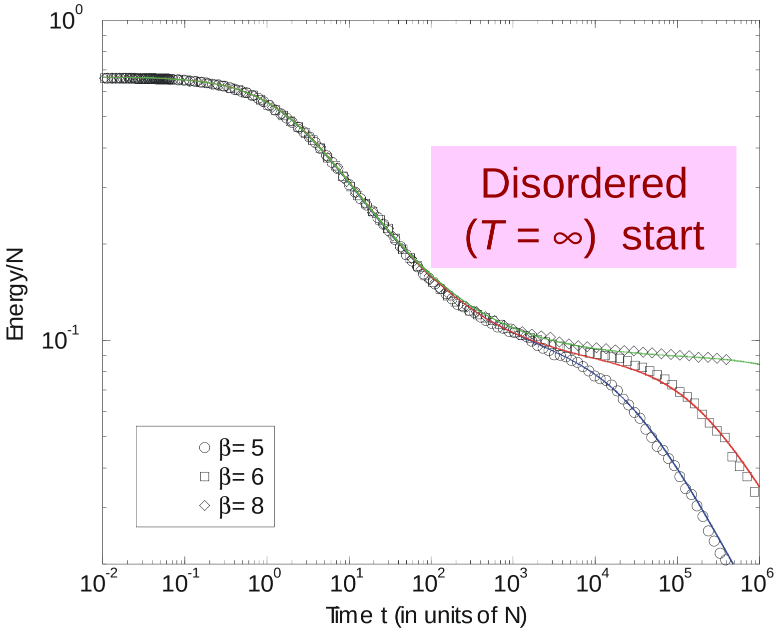
Activation barriers impenetrable at T=0

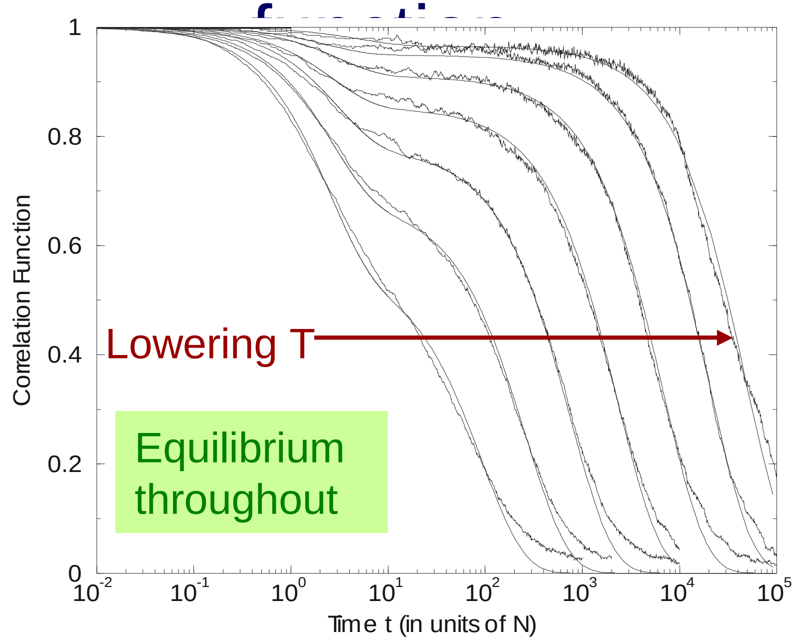
Macrodynamics

Energy

D > 0

Correlation function

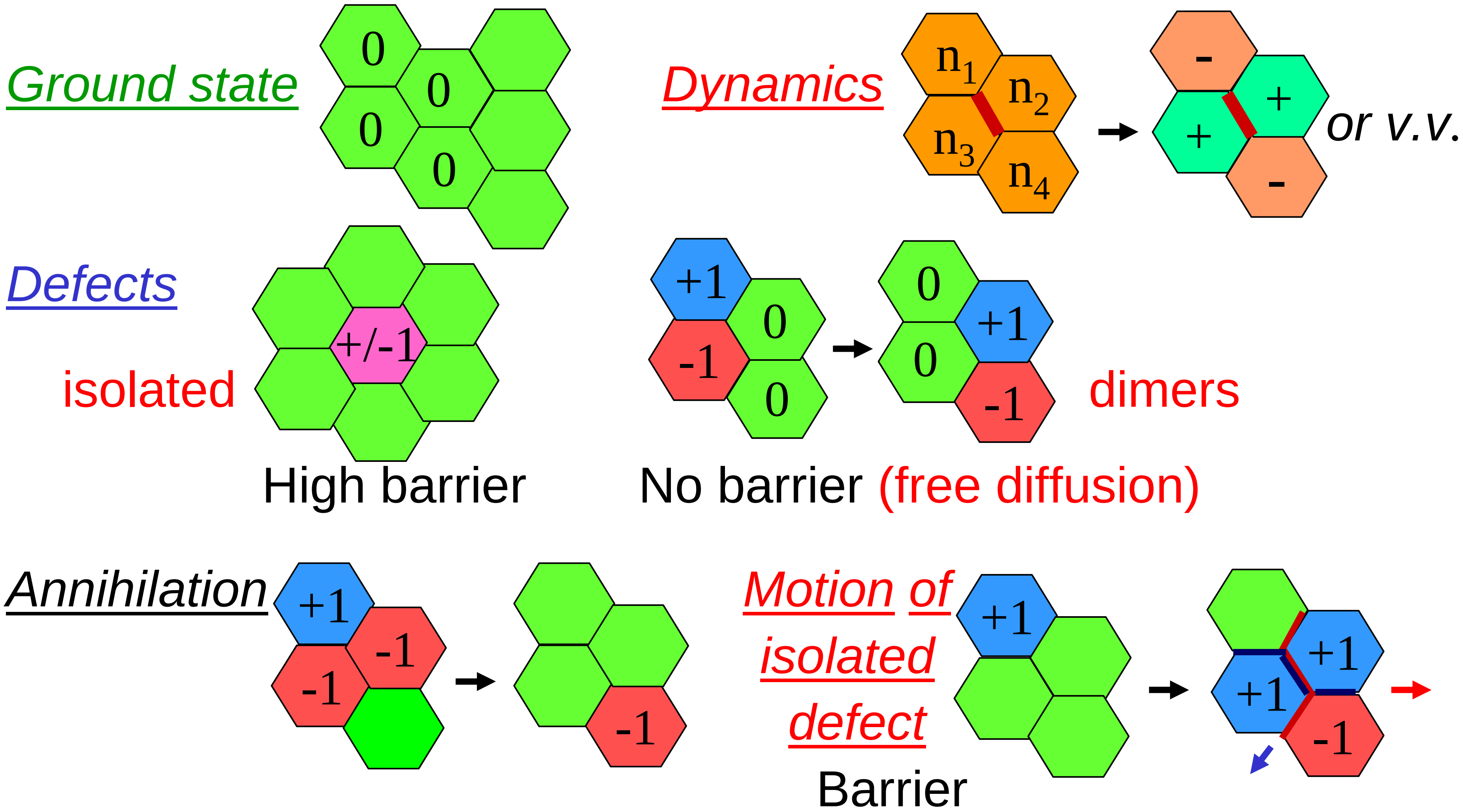




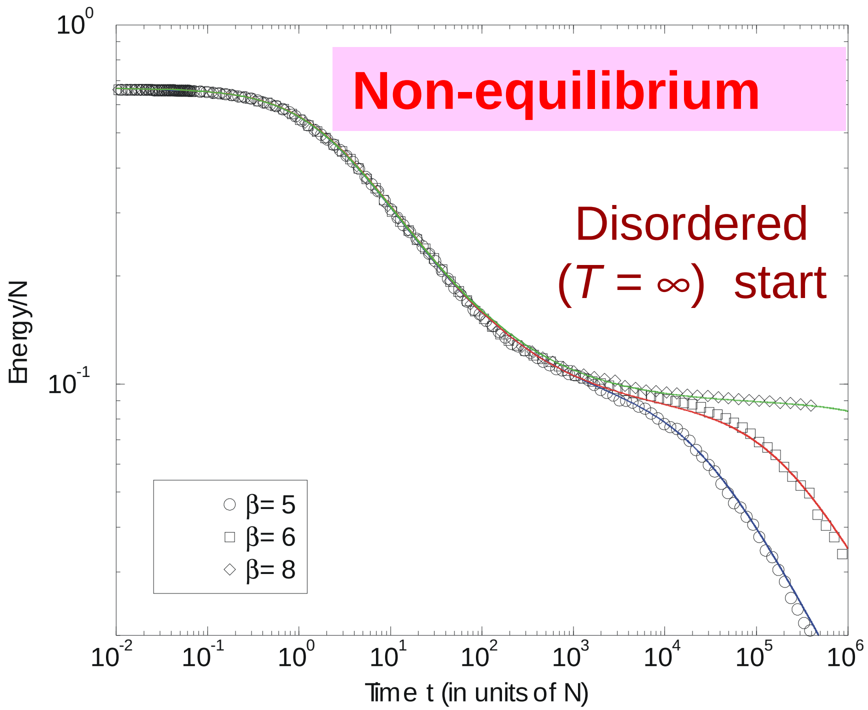
Points: simulations Curves: theoretically-inspired fits

Two times: Fast time ~ T-independent Slow time ~ exp(a/T)

Annihilation-diffusion



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)

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

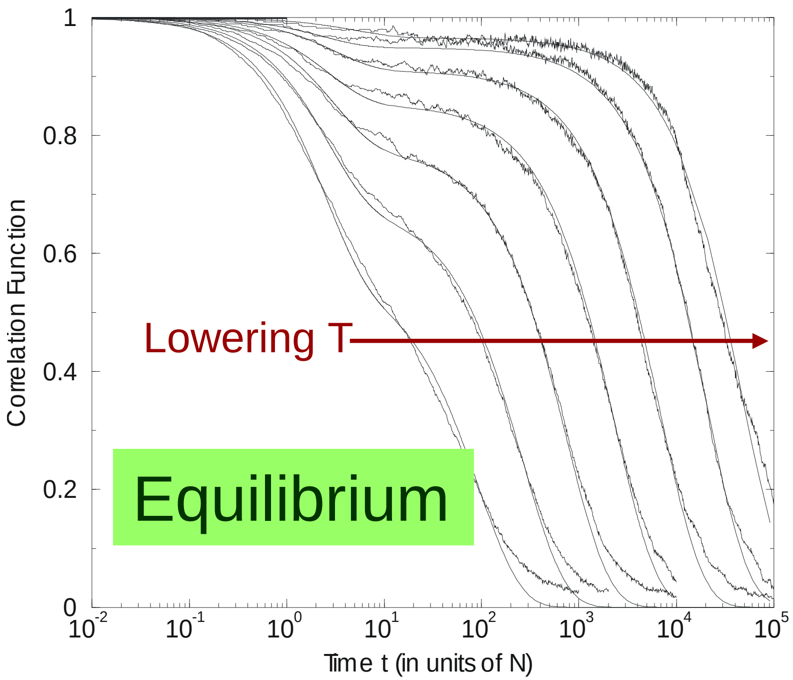
- **Single defects** need to pair to form dimers which can move and annihilate
- **Slow** diffusion
- Energy barriers→τ~exp(Δ/T);Arrhenius

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

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

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

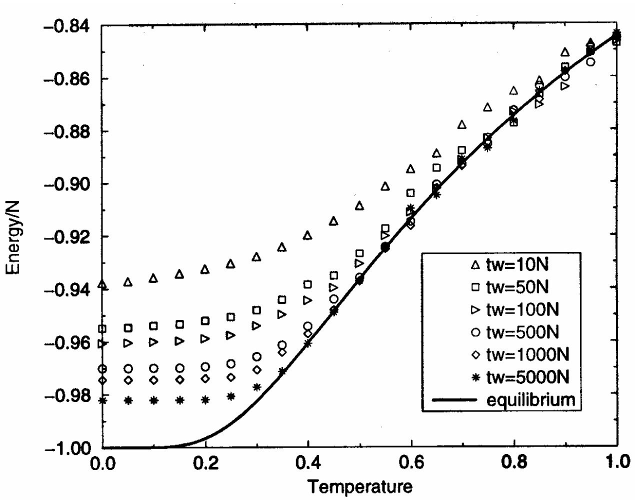
Single dimer type: (0,0):

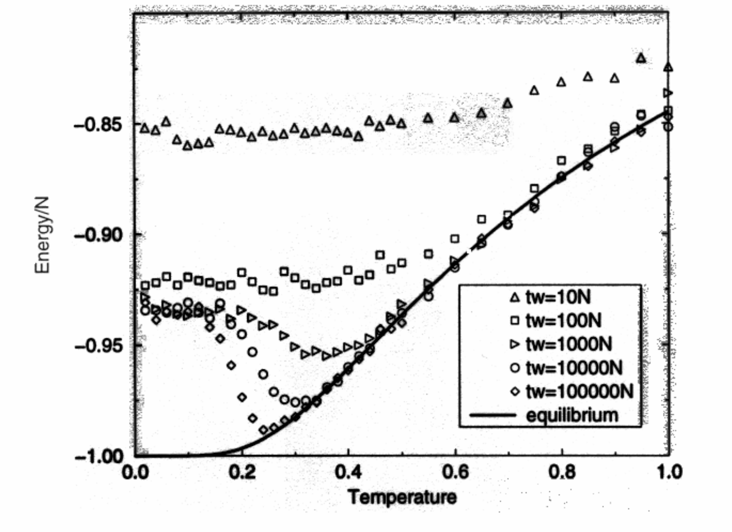
A+A+A→A A+A→Ø

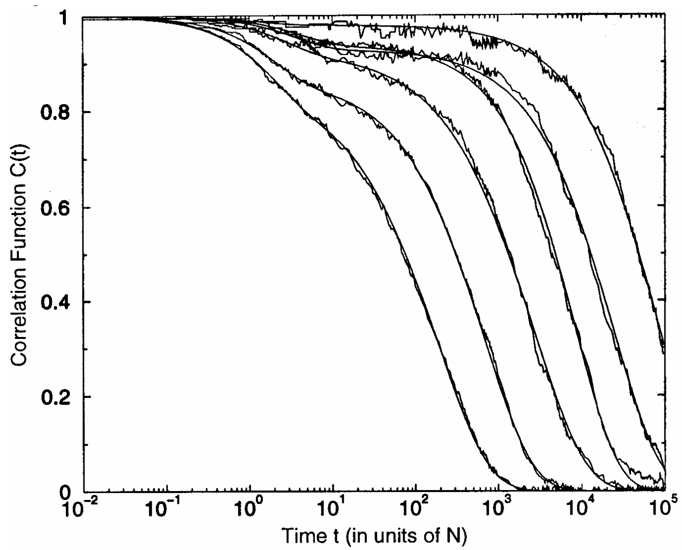
Different asymptotic decay exponent

Dimer diffusion can be blocked by disadvantageous environment

D < 0 results







Qualitatively similar to D>0

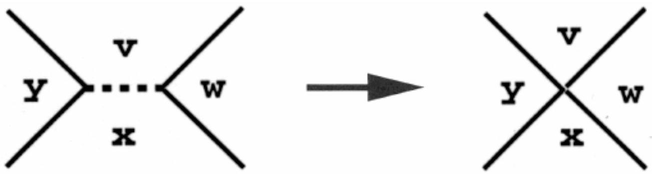
but

different exponents

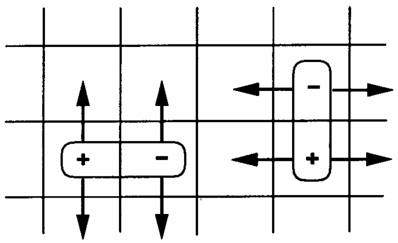
and some

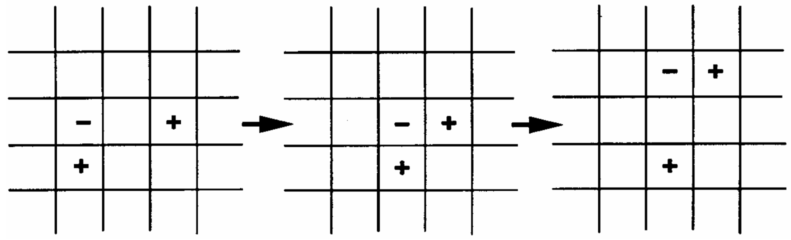
stretched exponential character

Square lattice

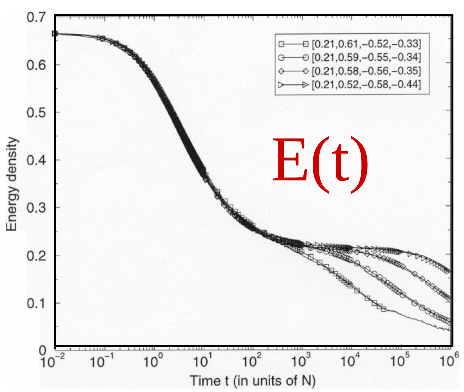


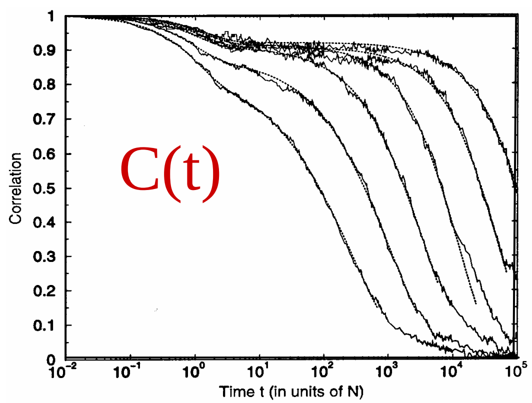
Dimer moves





Singleton move





Summary of processes

- Dimer annihilation:

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

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

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

All involve 4 neighbours

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

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

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

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*
  - *Either single defect type (A) or two types (A,B)*

Constrained 'backgammon'

- Non-interacting 'particles':

$$H = \sum_{i=1}^{N} n_i \quad n_i \leq 3$$

  - Trivial equilibrium, unique absorbing g.s.
- Constrained dynamics
  - Annihilation: analogue of dimer annihilation against defect;

$$(n_i, n_j) \rightarrow (n_i - 3, n_j + 1)$$

Rate =1

  - Diffusion: analogue of dimer diffusion

$$(n_i, n_j) \rightarrow (n_i - 2, n_j + 2)$$

Rate = D

  - Creation: analogue of defect motion by dimer creation

$$(n_i, n_j) \rightarrow (n_i - 1, n_j + 3)$$

Rate = $e^{-2\beta}$

Philosophy: follow number of A

- Dimer annihilation:

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

$\Rightarrow$ $(n_i, n_j) \rightarrow (n_i - 3, n_j + 1)$

- Dimer diffusion:

$$A+\bar{A}+2\phi \rightarrow 2\phi + A + \bar{A}$$

$\Rightarrow$ $(n_i, n_j) \rightarrow (n_i - 2, n_j + 2)$

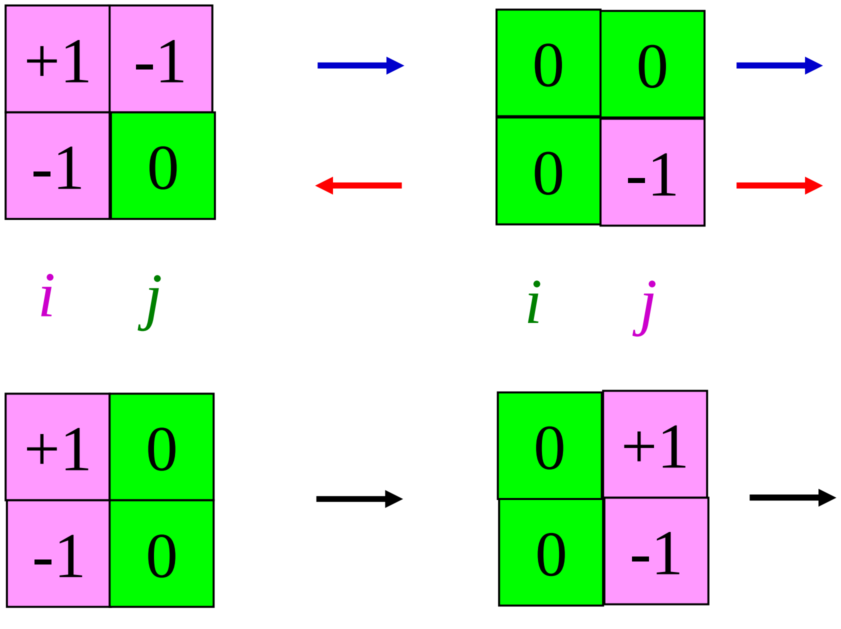
- Defect movement via dimer creation

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

$\Rightarrow$ $(n_i, n_j) \rightarrow (n_i - 1, n_j + 3)$

Dictionary: $A, \bar{A} \equiv$ defects, $\phi \equiv$ ground state

Translation between ‘languages’



$$(n_i, n_j) \rightarrow (n_i - 3, n_j + 1)$$

$$(n_j, n_i) \rightarrow (n_j - 1, n_i + 3)$$

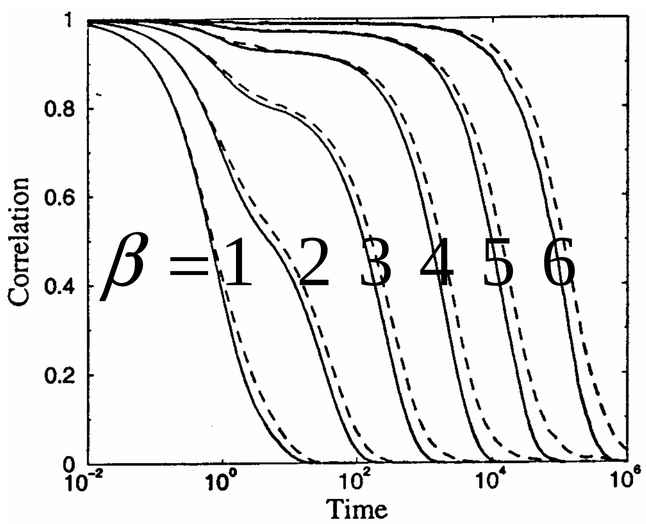
$$(n_i, n_j) \rightarrow (n_i - 2, n_j + 2)$$

Gains or losses of defects

Simulations

Equilibrium

Correlation function

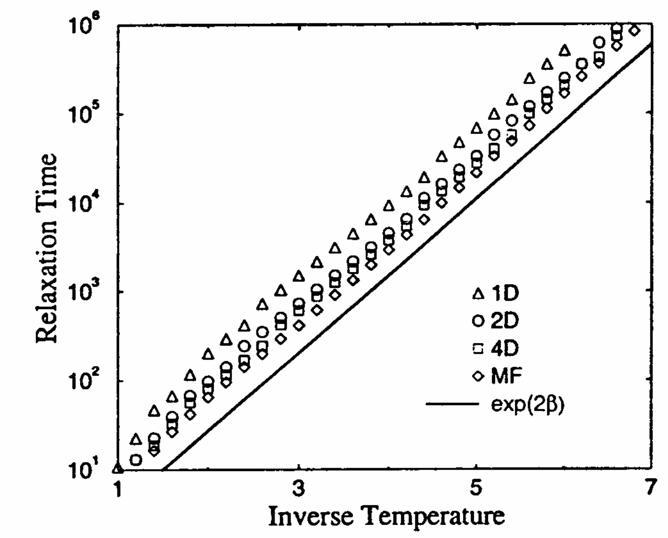


$d = 2$ (dashed), $d = \infty$ (solid)

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

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

Arrhenius decay



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

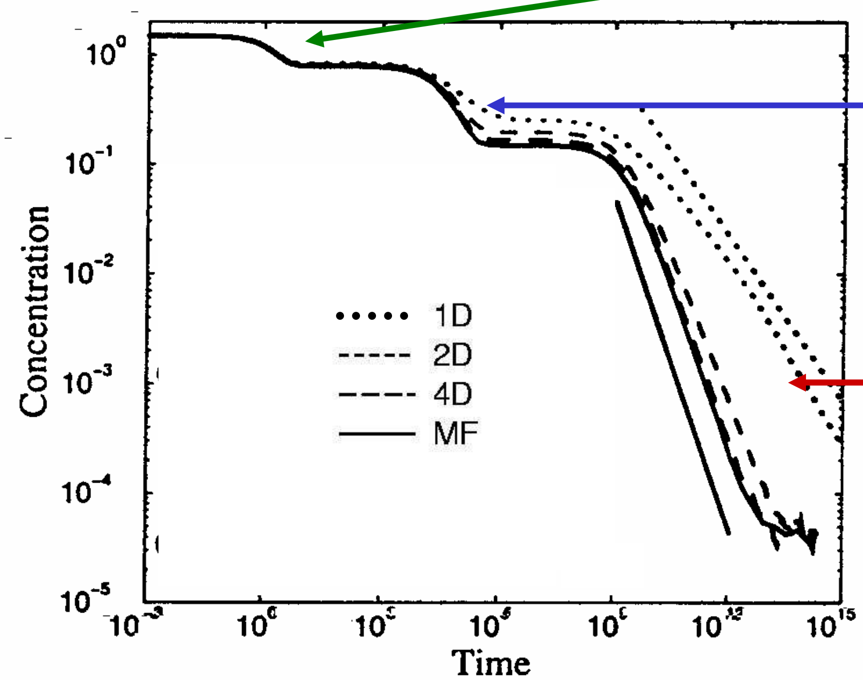
Energy/particle number decay

Single type of particle

Annihilation without diffusion (occurs even at T=0)

Dimer diffusion and annihilation (occurs even at T=0)

T=0.1



after quench

$$A + A \rightarrow \phi$$

$$\downarrow$$

field theory prediction

$$\alpha = d/2; \; d < d_c = 2$$

2 types of particle

$$H = \sum_{i=1}^{N} (n_i^A + n_i^B); \quad (n_i^A + n_i^B) \le 3$$

•

- Annihilation: analogue of dimer annihilation against defect;

  $$[(AAB)_{i,} X_j] \rightarrow [\phi_i, (AX)_j]$$ *Rate =1*

- Diffusion: analogue of dimer diffusion

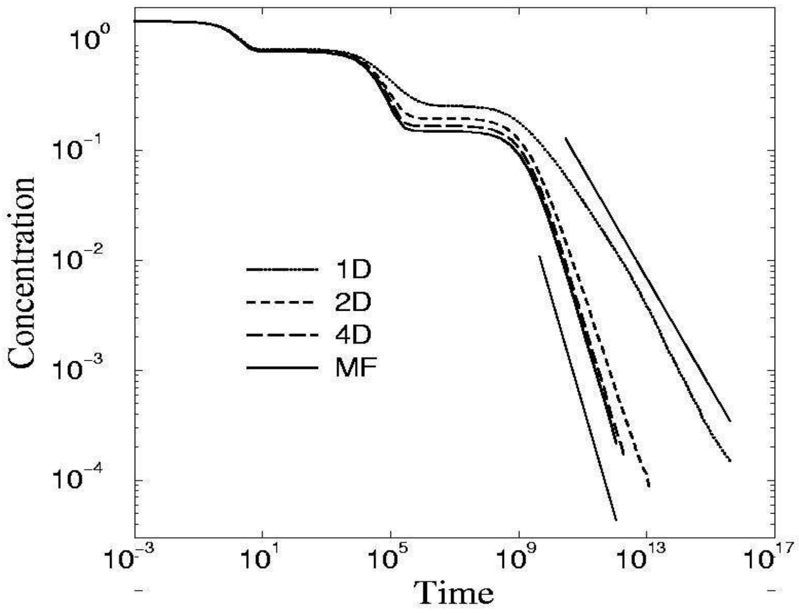
  $$[(ABX)_i, Y_j] \rightarrow [X_i, (ABY)_j]$$ *Rate = D*

- Creation: analogue of defect motion by dimer creation

  $$[(AX)_i, \phi_j] \rightarrow [X_i, (AAB)_j]$$ *Rate =* $e^{-2\beta}$

Energy (particle number) decay

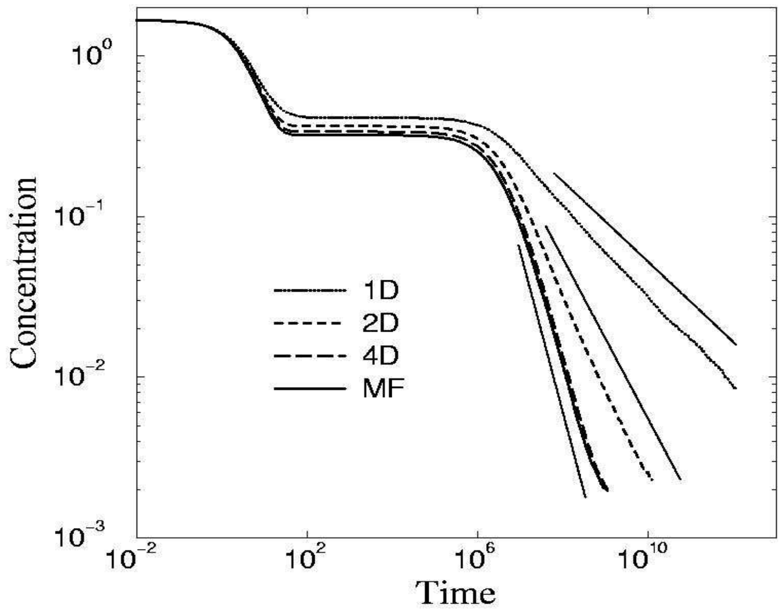
Single type of particle



Final decay: $A + A \rightarrow \phi$

$\alpha = d/2;\ d_c = 2$

Two types of particle



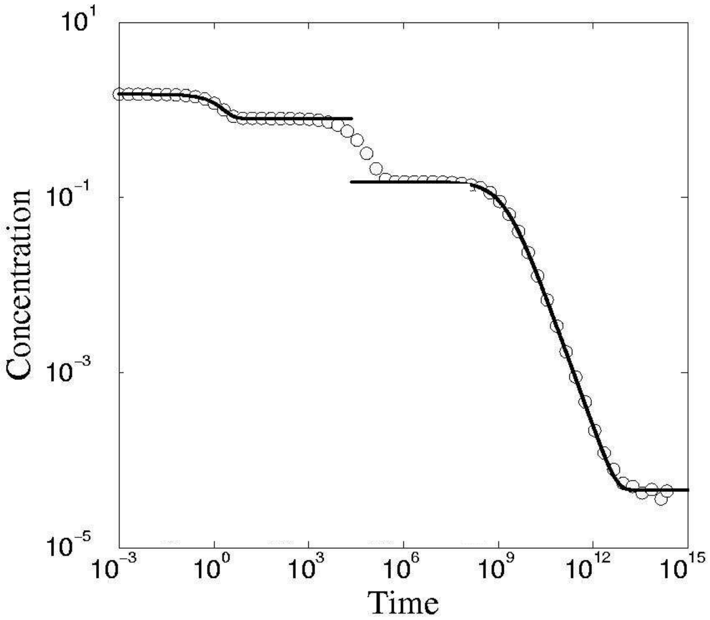
Final decay: $A + B \rightarrow \phi$

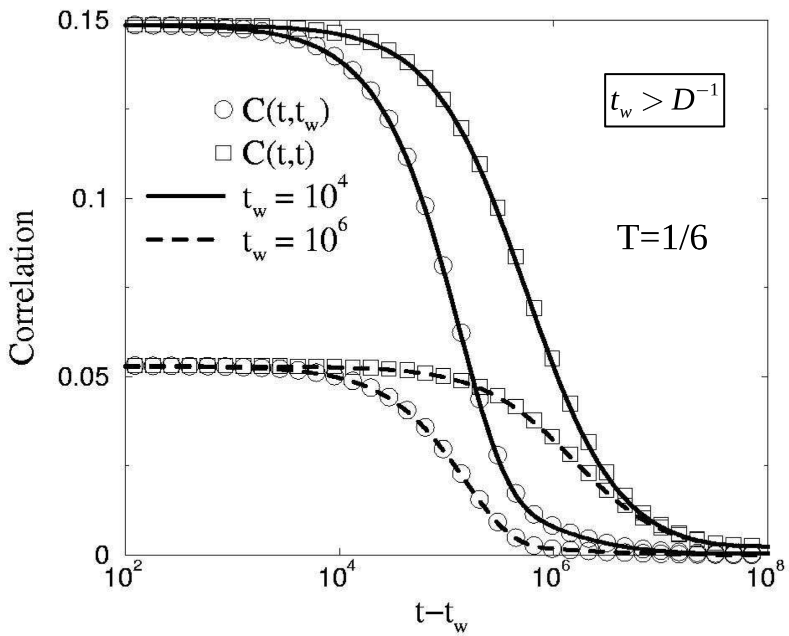
$\alpha = d/4;\ d_c = 4$

Theory & simulation (infinite d)

Concentration decay after quench

Out of equilibrium correlation & concentration





Circles ~ simulation, lines ~ theory

Circles ~ correlation, squares ~ concentration

Other systems/models

- Background
- Other common models
- Extensions

Return to

Current philosophy

Glassiness through kinetic constraints

Replace

Real interacting systems with simple constraints

by

Effective systems with no or weaker Hamiltonian interactions but more constrained dynamics

usually heuristic

Example

Spin-facilitated Ising models

Frederickson-Andersen

Idea: dense liquid

- Many regions of high density, few regions of low density.
- Atomic motion only possible if enough nearby mobile low-density regions to facilitate

Model: SFIM

$$H = \sum_i s_i - J \sum_{(ij)} s_i s_j$$

- Spins: $\downarrow \equiv$ dense, $\uparrow \equiv$ dilute,
- Heat bath/Glauber/Metropolis dynamics
  - but constrained
  - spin-flip only if f ≥ 1 of neighbours are up (nearby dilute/ mobile region).
- Gives glassy dynamics

Usually ignore *J:* $$H = \sum_i s_i$$

Field theory

Instantaneous distribution: $P(\{n_i(t)\})$

Dynamics: Master equation: $\partial P(\{n_i(t)\}) / \partial t = f(P(\{n_i(t)\})$

State functions: $\Psi(t) = \sum_{\{i\}} P(\{n_i(t)\})(a_1^+)^{n_1} ...(a_p^+)^{n_p} .....|0\rangle$

involving creation operators

$a^+\{n...,n_i,n......\} = \{n....,n_i+1,n...\}$

Dynamics: $\partial\ \Psi(t) / \partial t = H\Psi(t)$

H: Non-Hermitian Hamiltonian,

involving creation and annihilation operators

$a_i\{n...,n_i,n......\} = \{n....,n_i-1,n...\}$

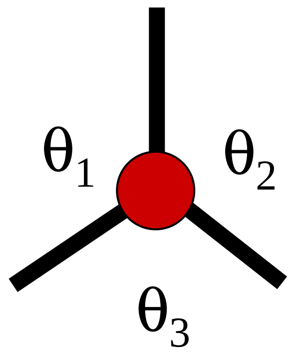
Coherent state representation: c-number fields

Generating functional integral: $Z = \int D\varphi D\varphi^*\ \exp(-S(\{\varphi(t),\varphi^*(t)\}))$

Renormalization group.

Static phase transition to crystalline order in present problem?

Include correlation energies in Hamiltonian



$$\lambda f\{(\theta_1+\theta_2+\theta_3-2\pi)^2\}$$

but not yet done

Instead

Model with ‘crystalline’ phase

Baxter’s 8-vertex model

2-d lattice of spins (i,j): exactly soluble thermodynamics

$$H = \sum_{ij} \{ -D\sigma_{ij}\sigma_{i+1,j}\sigma_{i,j+1}\sigma_{i+1,j+1} - J(\sigma_{ij}\sigma_{i+1,j+1} + \sigma_{i+1,j}\sigma_{i,j+1}) \}$$

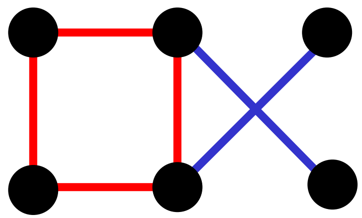
plaquette

Alone gives no thermodynamic transition

ferromagnetic on diagonals

Alone gives 2 separable systems with ferromagnretic transition at $T_c^0 = 2J/\sinh^{-1}1$

Glassy slow-down at $T_0 \sim D$



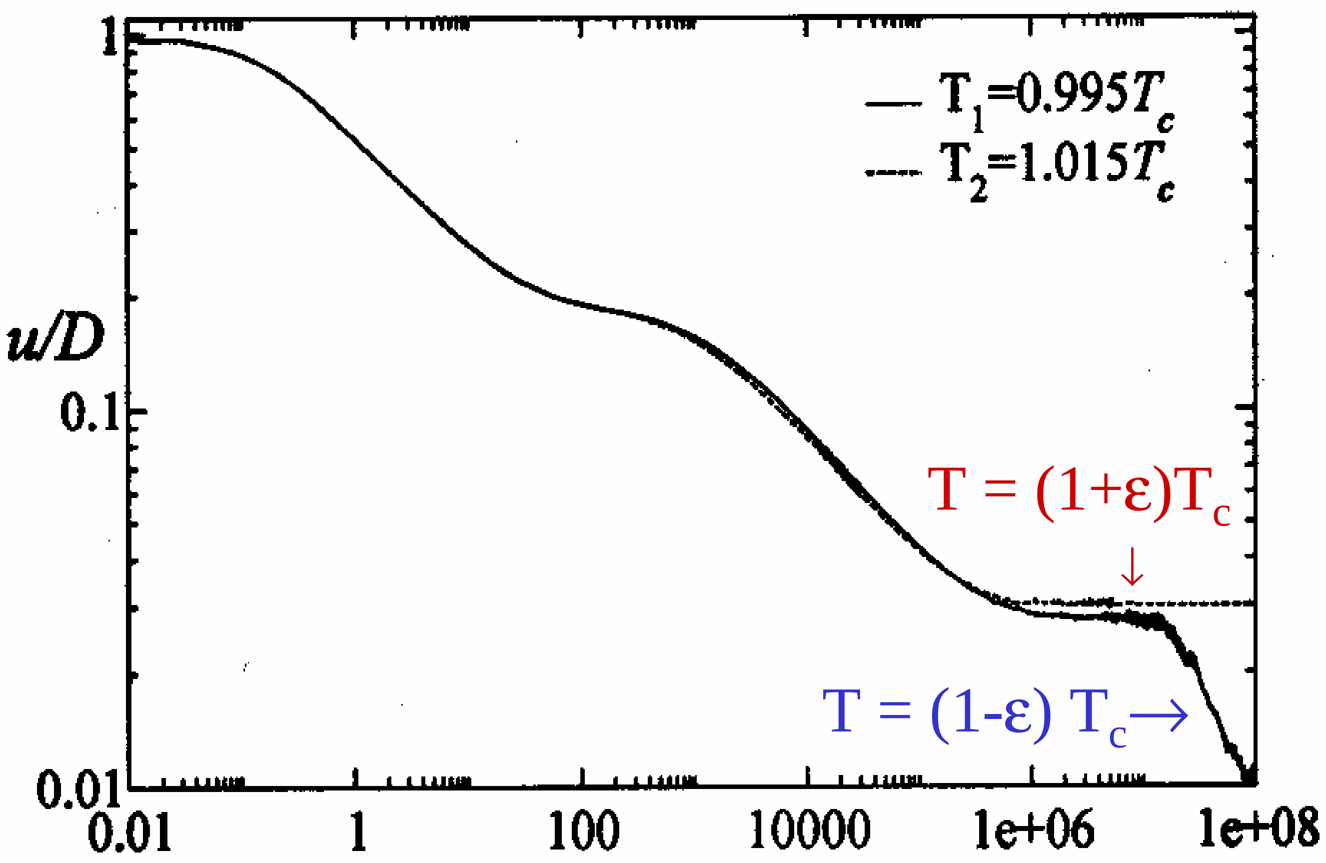
(Jack, Garrahan, S)

Ferromagnetic transition at T_c

$$\sinh(2J/T_c) = \exp(-D/T_c)$$

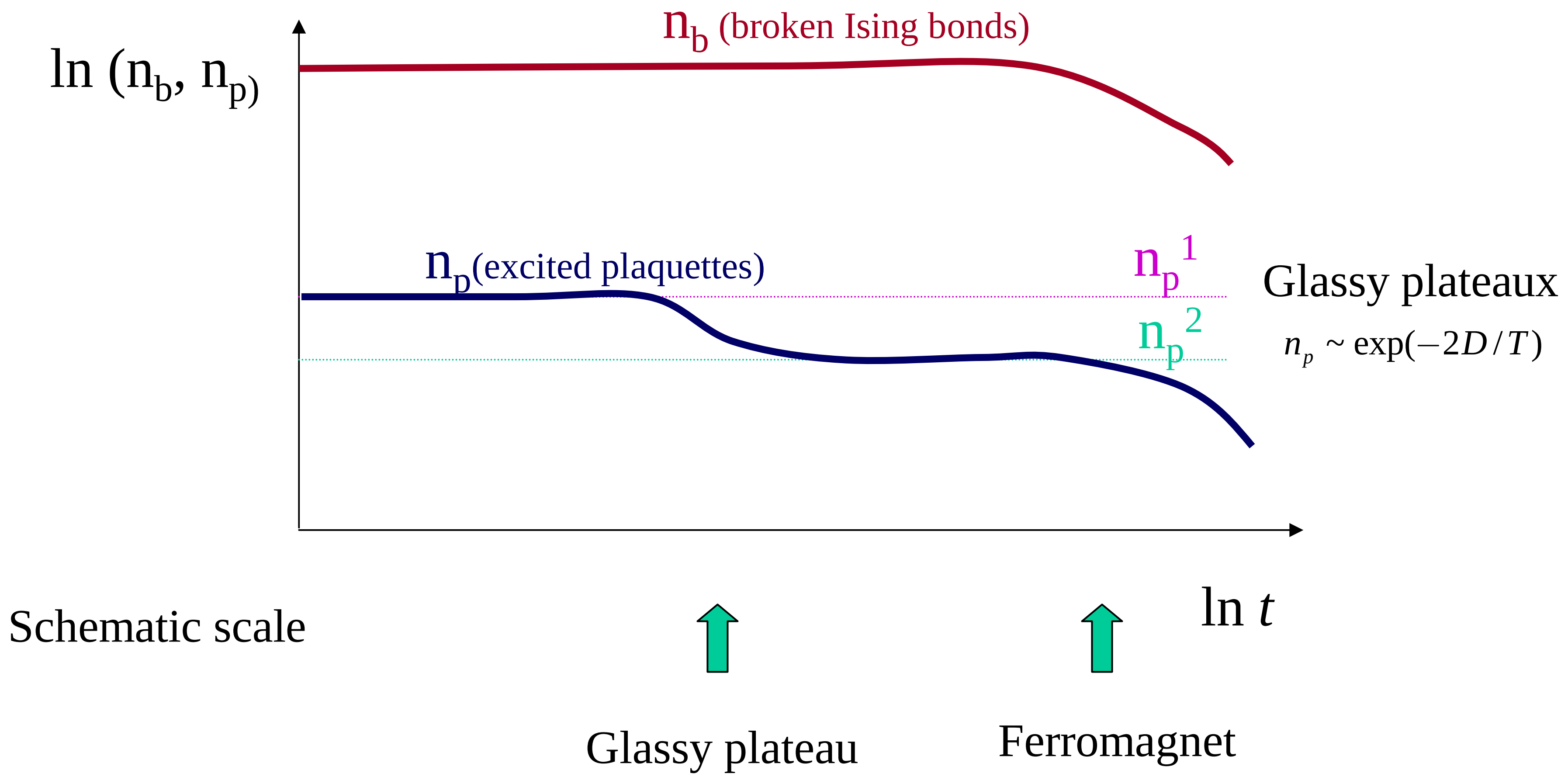
E(t); quenches from T(*t*=0) = ∞

$T_g \sim D > T_c$



Broken Ising bonds, excited plaquettes

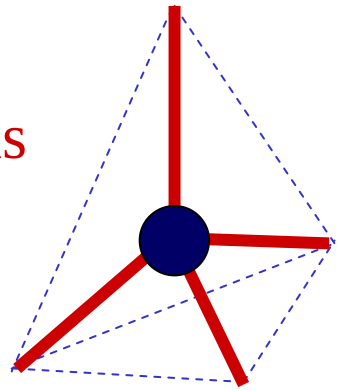
Quench: $T^1 \sim (1+\varepsilon)T_c \;\rightarrow\; T^2 \sim (1-\varepsilon)T_c$



3-d? sp^3-bonds etc.?

- 3-d networks with sp^3 bonds: *cf. α-silicon*

Random connections between bonds
(no dangling bonds)



Random pairwise re-connection dynamics:
e.g. Glauber-Kawasaki-WWW

(Wejchert, Weaire, Wooten)

? Effective constrained dynamics?

?What are analogues of the cells?

3-d volumes?

2-d areas?

Does it matter?

Other rules?

Strong / fragile?

Above strong Lennard-Jones fragile

Both have foam-like structure

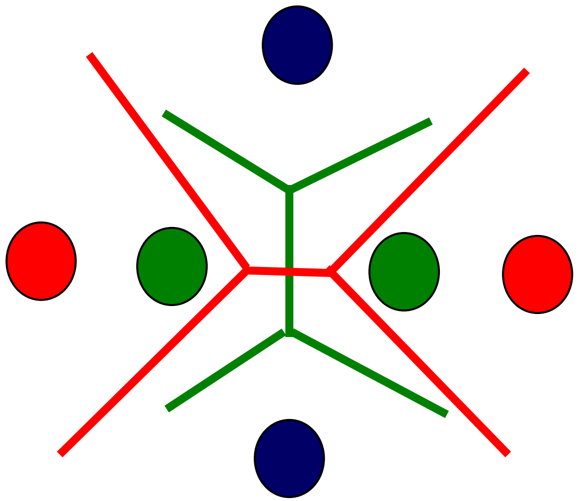
Covalent bonds Dual Wigner-Seitz cells

But different energetics for changes

Mainly topology Softer

Spherical atoms: Voronoid cells

Motion of the spheres



Continuous range of positions and energies from green to red

Strong to fragile?

Binary glasses

- 2 sizes of atom
  - ? Topological analogue
  - Eckmann:
    - Two “colours” of plaquette, “red” & “blue”
    - “red” want 5 sides, “blue” want 7 sides

$$H = \sum_{red} (n_i - 5)^2 + \sum_{blue} (n_j - 7)^2$$

- But actually more subtle: packing “reds” together or “blues” together they want to be 6-sided
  - Also Euler’s theorem always true (independent of $\#_{red} / \#_{blue}$

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
- Potentially interesting extensions