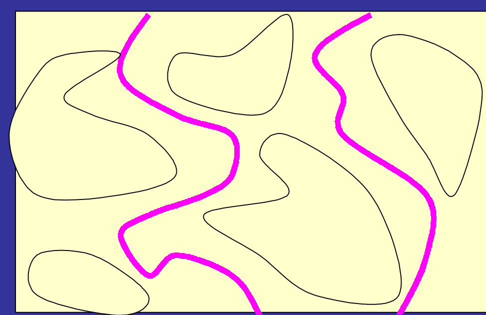
Activated dynamics at a non-disordered critical point: Constraints and the Vogel-Fulcher Law



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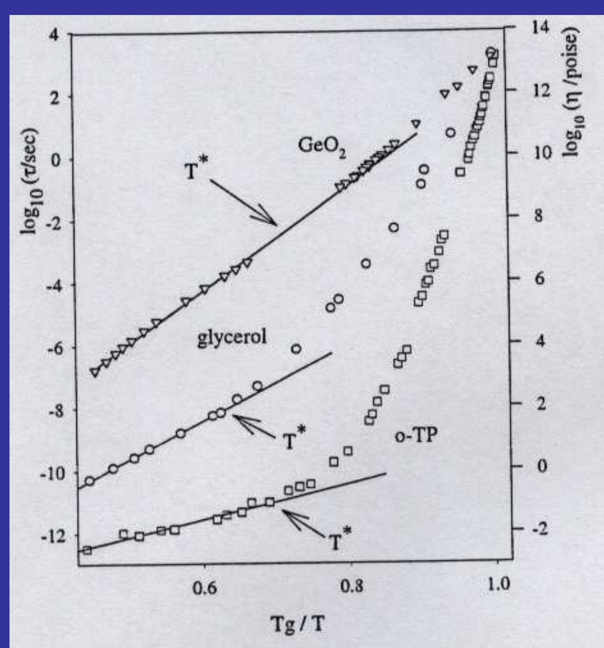
supported by NSF

Viscosity and relaxation time as a function of temperature

Figure taken from

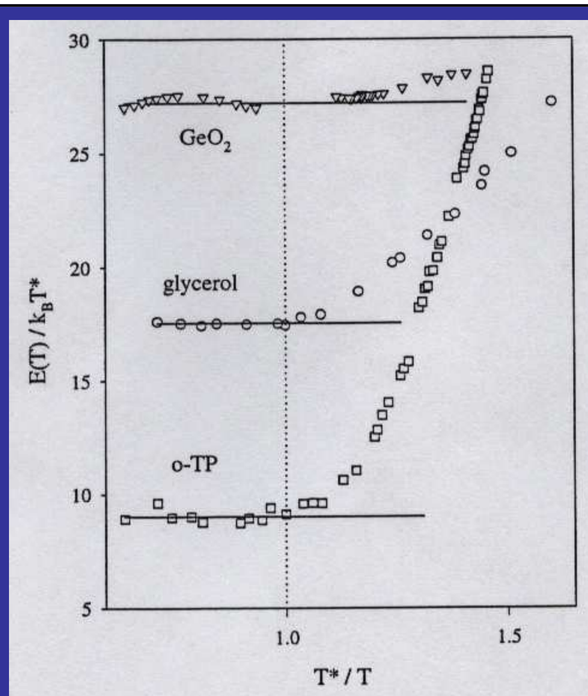
G Tarjus et al J.Phys. Cond Mat (2000)



Barrier heights as a function of temperature

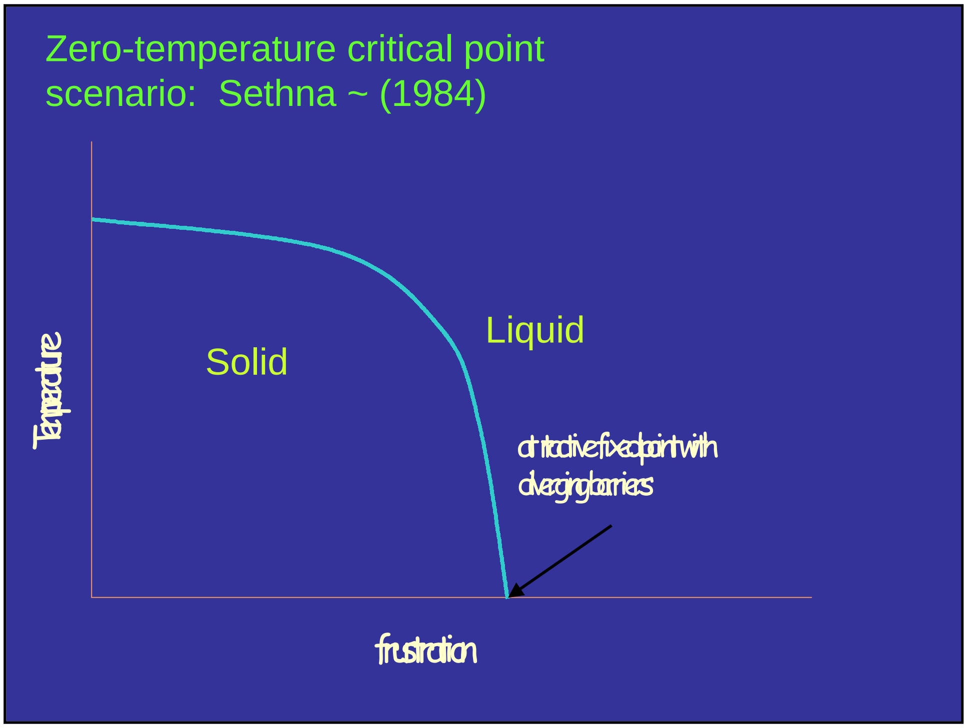
Figure taken from

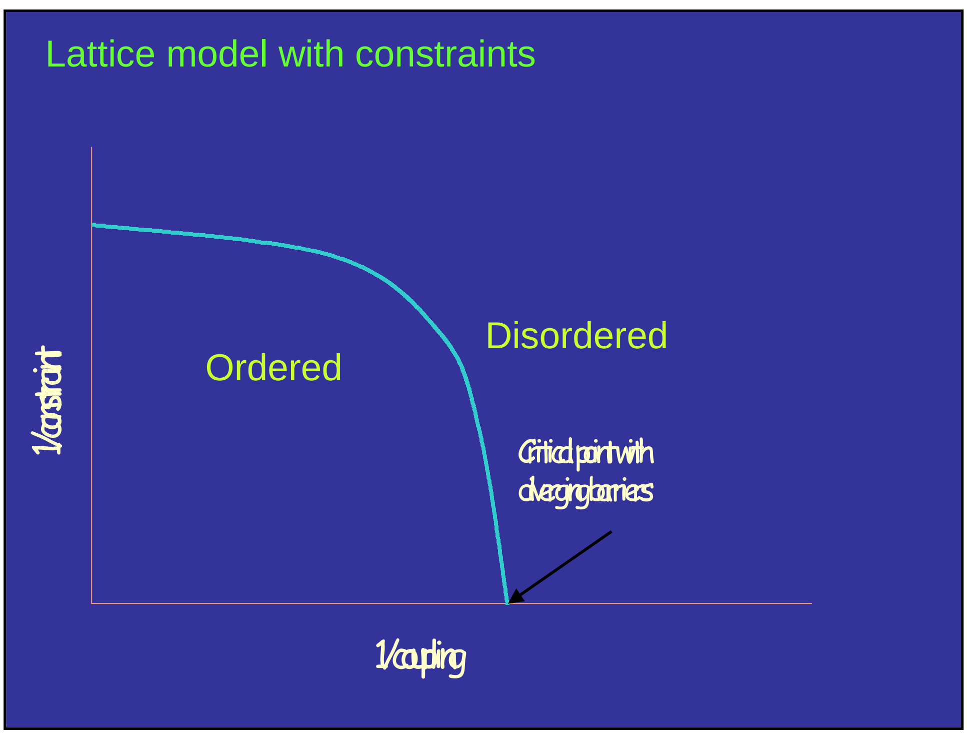
G Tarjus et al J.Phys. Cond Mat (2000)



Critical Point with activated dynamics

- Correlation length diverges as
  - $\xi \sim |T-T_C|^{-\nu}$
- Relaxation time diverges as
  - $\tau \sim \exp[\xi^{\theta}]$ ➔ Diverging barriers

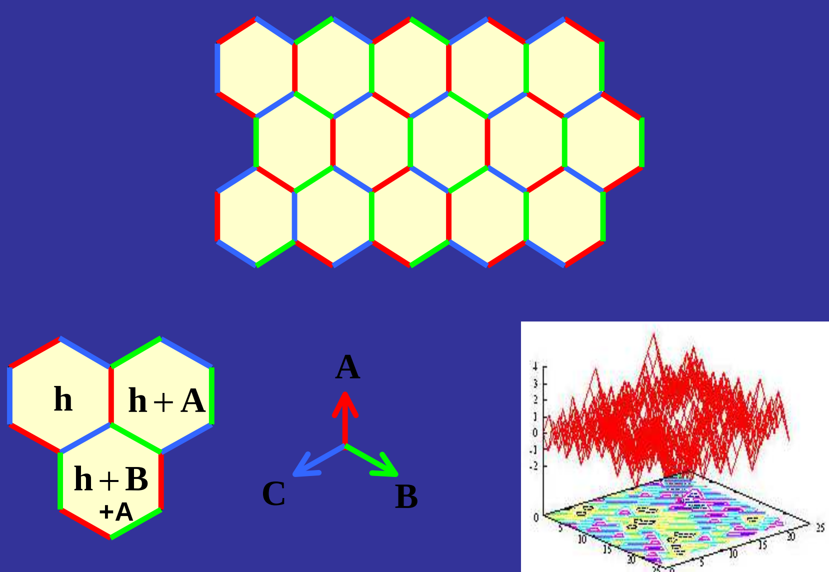

Zero-temperature critical point
scenario: Sethna ~ (1984)
Liquid
Solid


Lattice model with constraints
Disordered
Ordered

Outline

- Outline of Lattice Model
  - ❖Simulation Results
- Construction of 'coarse-grained' dynamics
  - ❖Saddle point analysis of correlation function
  - ❖Argument for Vogel-Fulcher Divergence
- Continuum Model
  - ❖Exact Results ➔ Vogel-Fulcher
  - ❖Connection to Bouchaud's trap model
- Predictions
- Future Work

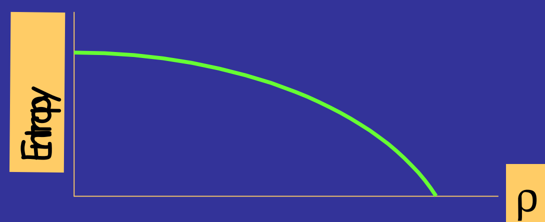
Three-coloring model



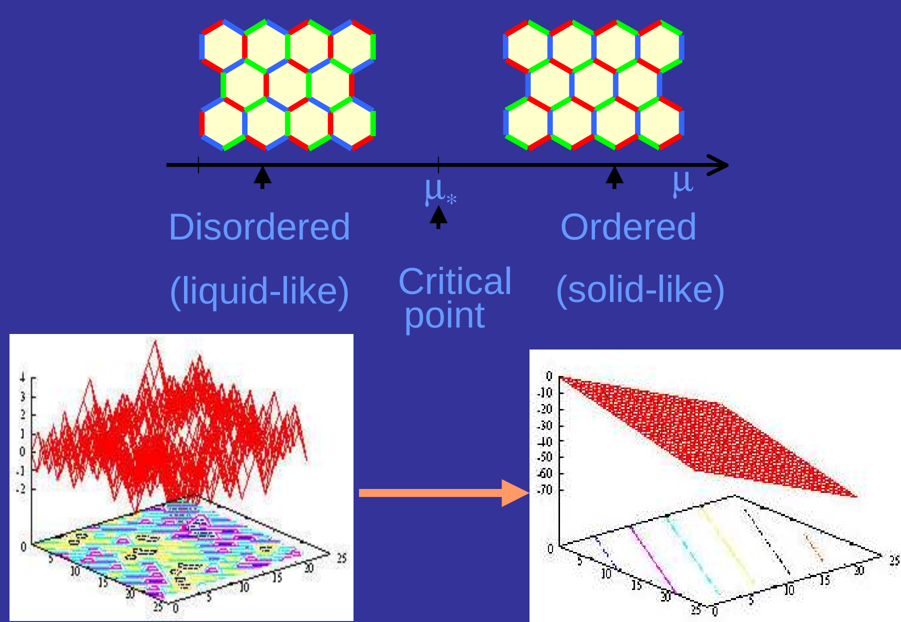
- Energy depends on the global tilt (ρ) of one height component

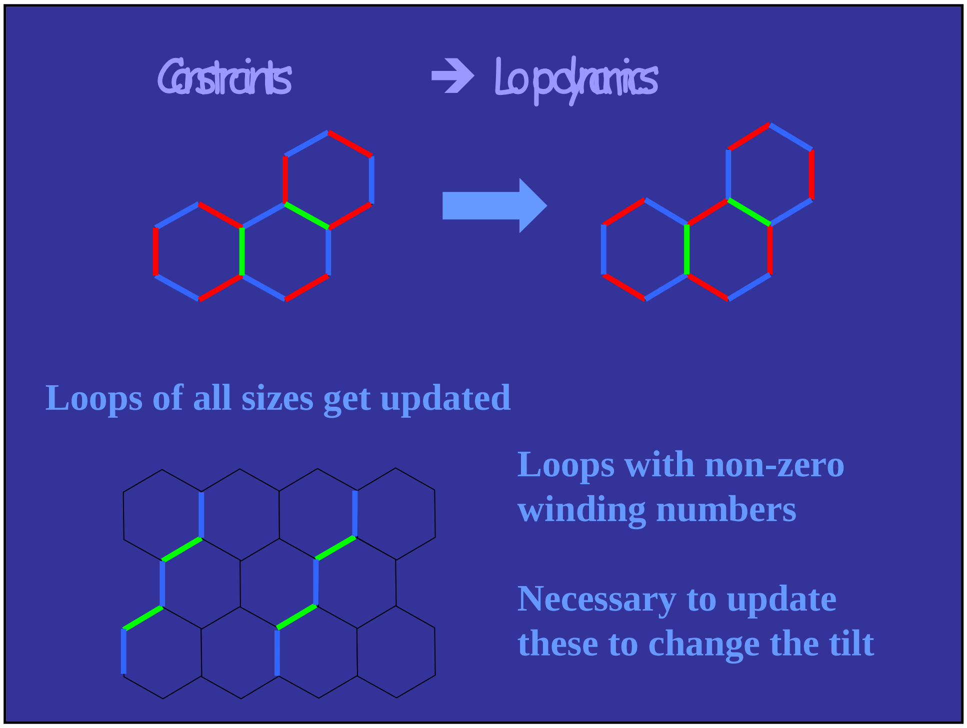
$$E(\rho) = -(\mu L^2/3) \quad (1 + 8\rho^2);$$

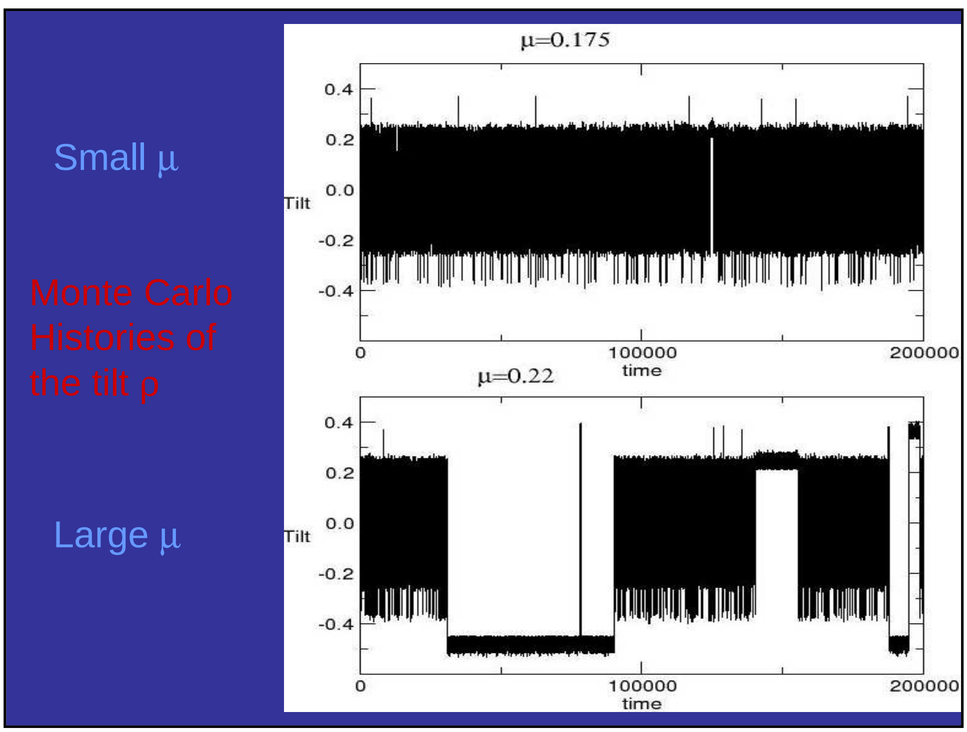
- Entropy of different tilt sectors is "known"

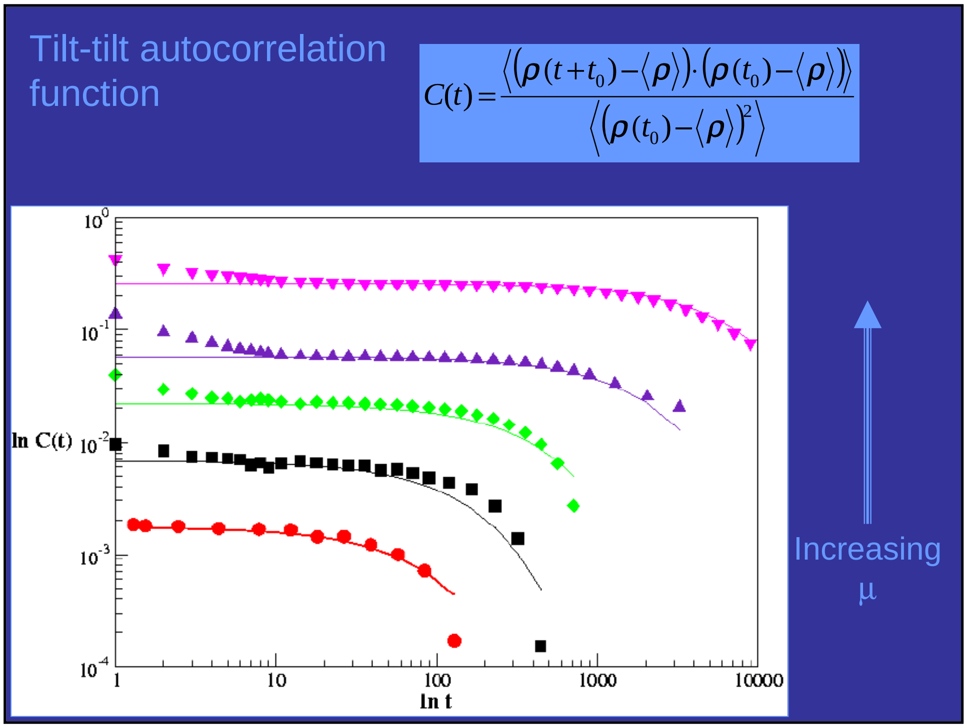


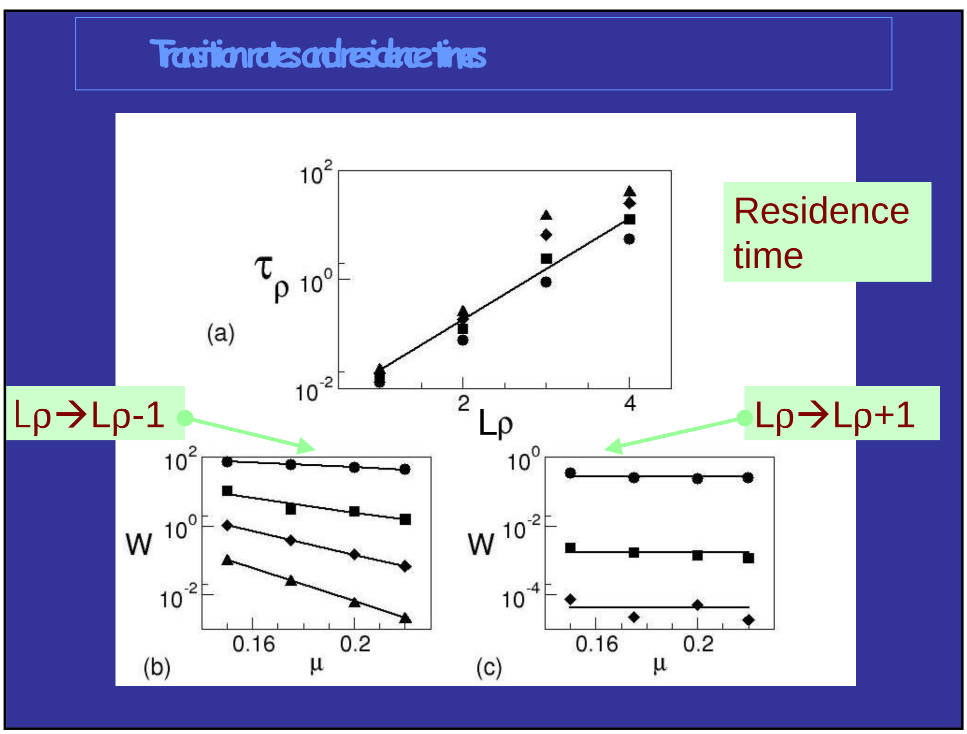
Equilibrium transition




Constraints
Loop dynamics
Loops of all sizes get updated
Loops with non-zero winding numbers
Necessary to update these to change the tilt


Small μ
Monte Carlo Histories of the tilt ρ
Large μ
μ=0.175
μ=0.22
Tilt
time
0.4
0.2
0.0
-0.2
-0.4
0
100000
200000

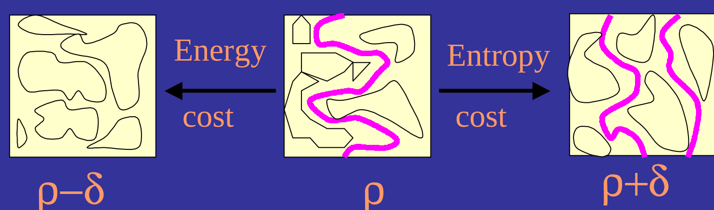

Tilt-tilt autocorrelation function
$C(t)=\frac{\langle(\rho(t+t_0)-\langle\rho\rangle)\cdot(\rho(t_0)-\langle\rho\rangle)\rangle}{\langle(\rho(t_0)-\langle\rho\rangle)^2\rangle}$
ln C(t)
ln t
Increasing μ


Residence time
τ_ρ
(a)
Lρ
Lρ→Lρ-1
Lρ→Lρ+1
W
μ
(b)
(c)

Transition Rates

$W_{L\rho \to L\rho-1} \sim \exp[-\Delta E]$

$W_{L\rho-1 \to L\rho} \sim \exp[-\Delta S]$

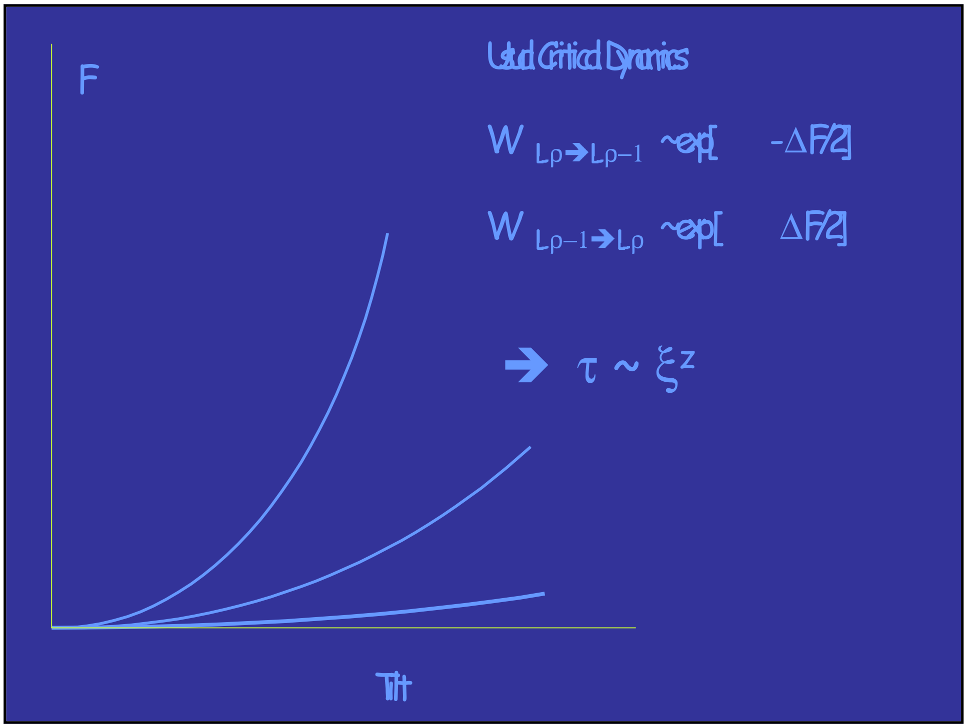


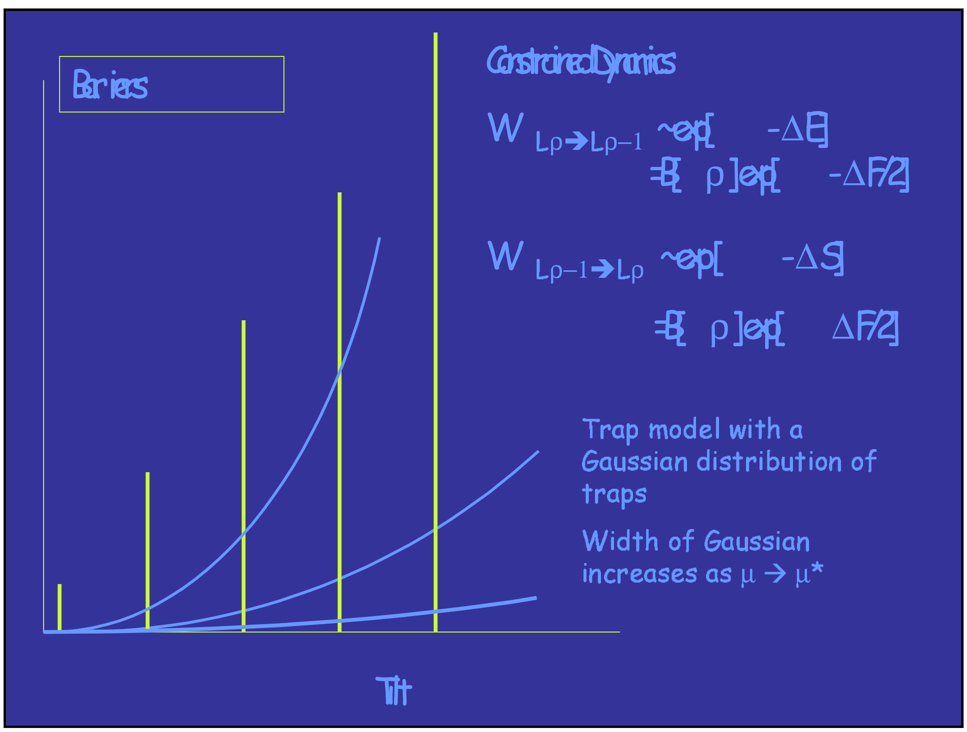
Vogel-Fulcher Divergence

- Saddle Point result for C(t)
  - $C(t) \sim e^{-(\mu^*-\mu)[\ln(t)]^2}$

$C(\tau)=e^{-c_0}$

$\Rightarrow \tau \sim e^{[c_0/(\mu^*-\mu)]^{1/2}}$


F
W Lρ→Lρ−1 ~exp[-ΔF/2]
W Lρ−1→Lρ ~exp[ΔF/2]
→ τ ~ ξz


W Lρ→Lρ−1 ~exp[-ΔE]
W Lρ−1→Lρ ~exp[-ΔS]
Trap model with a Gaussian distribution of traps
Width of Gaussian increases as μ → μ*

Continuum Model

- Purely quadratic free energy:

  $f(\rho) = (\mu^* - \mu)\,\rho^2$

- Limit of $\mu \rightarrow \mu^*, L \rightarrow \infty$,

  $x = L\rho$, $\lambda = \mu^* - \mu$,

  Equation for P(x,t)

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial x}\left[\lambda x e^{-\mu^* x} P + 2D e^{-\mu^* x} \frac{\partial P}{\partial x}\right]$$

Without these factors, this would lead to usual Langevin equation for x

Continuum Model

- Solution P(x,t)

$$P(x,t) \simeq e^{-\left(\sqrt{\frac{2}{tD\mu^{*2}}}\right)e^{\mu^* x/2}} e^{-\frac{\lambda}{2D}x^2}$$

Two length scales

$$l_1 \simeq \log t/\mu^*$$

$$l_2 = \sqrt{\frac{2D}{\lambda}}$$

Crossover time:

$$\tau_c = e^{2\mu^*}\sqrt{D/\lambda}$$

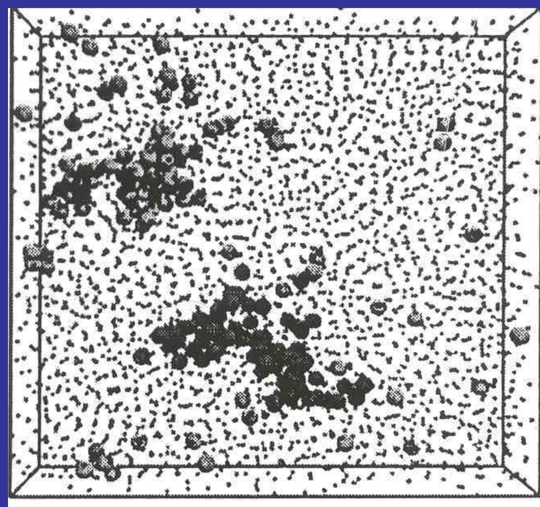
Connection to trap models

- $y = e^{\mu^* x/2}$ ~ the barriers
- Equilibrium distribution of barriers
  - ❖ $yP(y) = e^{(-\lambda \log(y)^2)}$
- Observed trapping time dependence in binary Lennard-Jones!

Connection to "real" liquids

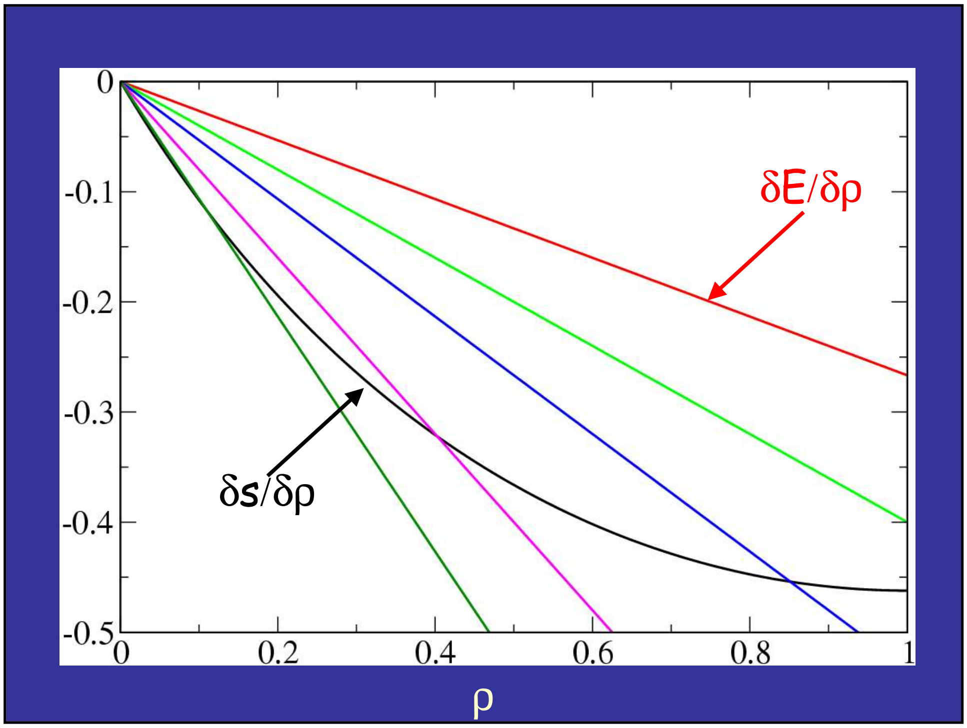
- Extended structures generate constraints
- Trap-like dynamics
- Evidence from simulations of Lennard-Jones fluid
- Prediction ➔ Diffusion constant decays exponentially with "length"

Extended Structures



Future work

- Aging
- Other types of barriers
- Parameter controlling fragility
- Connection to atomistic models


0
-0.1
-0.2
-0.3
-0.4
-0.5
0
0.2
0.4
0.6
0.8
1
δE/δρ
δs/δρ
ρ