Microscopic Picture of Aging in Silica Glass: A Computer Simulation

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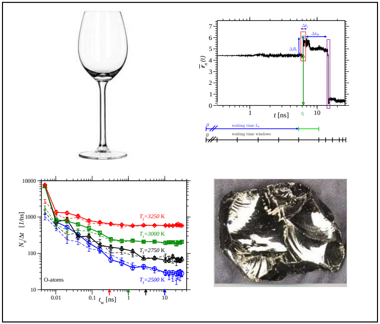
Bucknell University



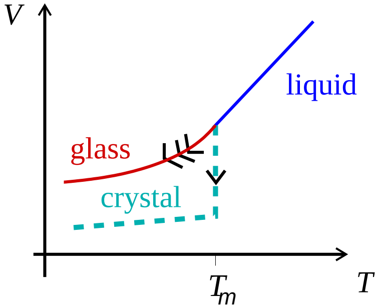
Acknowledgments: J. Horbach & A. Zippelius

What is a Glass?

Examples:

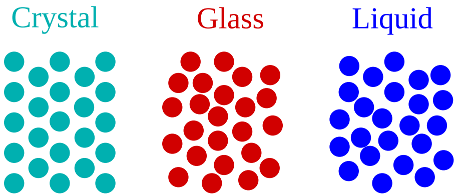
- Drinking Glass, Pyrex, Schott Ceran Cooktop
- Fiber-Optics, Telescope Lense, Reading Glasses
- Solar Panel Glass
- Vulcanic Glass
- Golf-Club

What is a Glass?



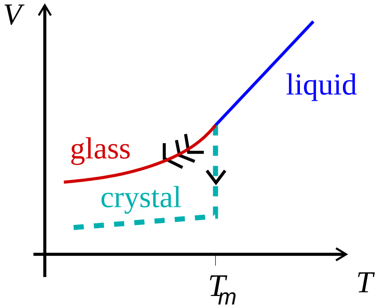
Glass:

⟶ system falls out of equilibrium



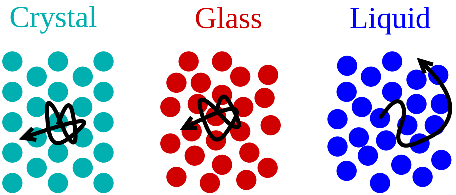
Structure: discordered

What is a Glass?



Glass:

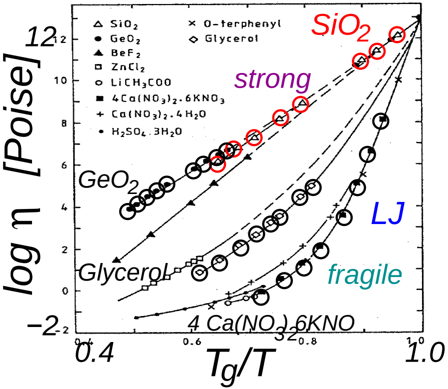
→ system falls out of equilibrium



Structure: discordered

Dynamics: frozen in

What is a Glass?



[C.A. Angell and W. Sichina, Ann. NY Acad. Sci. 279, 53 (1976)]

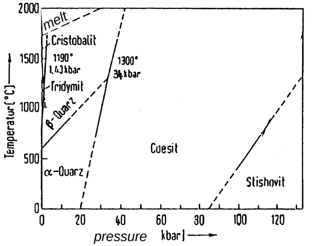
Dynamics:

Viscocity η as function of inverse temperature T

- slowing down of many decades
- strong and fragile glass formers
- SiO_2 strong glass former

$\longrightarrow$ very interesting dynamics

System: SiO_2



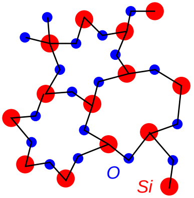
[S. Stoeffler and J. Arndt, Naturwissenschaften 56, 100 (1969)]

- rich phase diagram
- similar to water (H_2O)

Model: BKS Potential

[B.W.H. van Beest *et al.*, PRL 64, 1955 (1990)]

$$\phi(r_{ij}) = \frac{q_i q_j e^2}{r_{ij}} + A_{ij} e^{-B_{ij} r_{ij}} - \frac{C_{ij}}{r_{ij}^6}$$



112 Si & 224 O $\rho = 2.32$ g/cm^3

$T_c = 3330$ K

Numerical Solution: Euler Step

Initialize:

$x(t_0),\ v(t_0),\ a(t_0)$

↓

$x(t_0+\Delta t),\ v(t_0+\Delta t),\ a(t_0+\Delta t)$

↓

$x(t_0+2\Delta t),\ v(t_0+2\Delta t),\ a(t_0+2\Delta t)$

↓ etc.

↓ = Iteration Step:

$$x(t+\Delta t)=x(t)+v(t)\Delta t$$
$$v(t+\Delta t)=v(t)+a(t)\Delta t$$
$$a(t)=F(t)/m=-(dU/dx)(t)$$

Molecular Dynamics Simulation

Initialize:

$\vec{x}_i(t_0),\ \vec{v}_i(t_0),\ \vec{a}_i(t_0)$

particles $i=1,\ldots,N$

three dimensions

$\vec{x}_i(t_0+\Delta t),\ \vec{v}_i(t_0+\Delta t),\ \vec{a}_i(t_0+\Delta t)$

$\vec{x}_i(t_0+2\Delta t),\ \vec{v}_i(t_0+2\Delta t),\ \vec{a}_i(t_0+2\Delta t)$

etc.

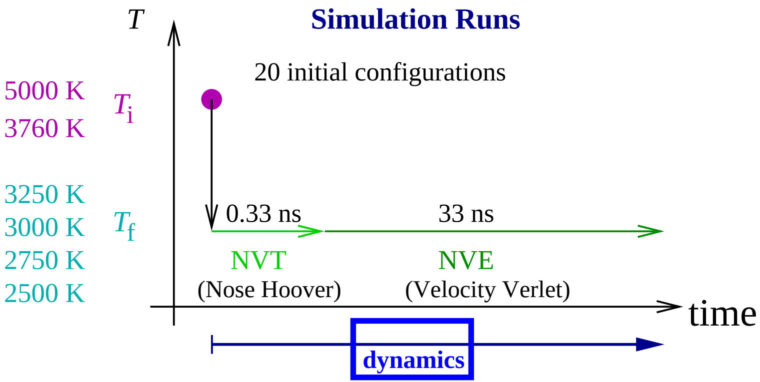
↓ = Iteration Step: (Velocity Verlet)

$$\vec{x}_i(t+\Delta t)=\vec{x}_i(t)+\vec{v}_i(t)\Delta t+\vec{a}_i(t)(\Delta t)^2/2$$

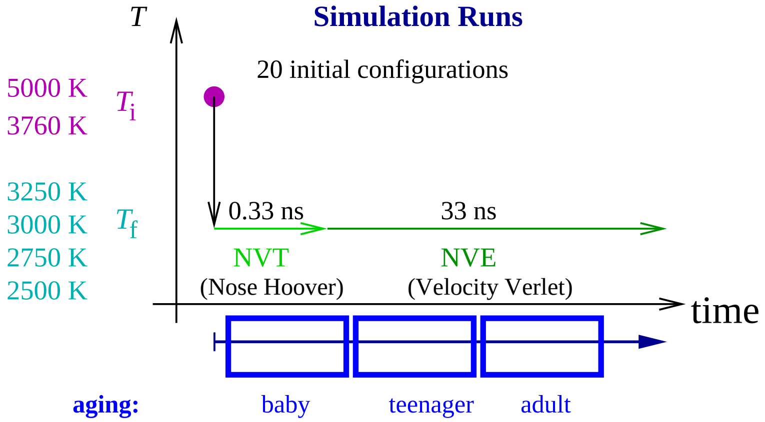
$$\vec{v}_i(t+\Delta t)=\vec{v}_i(t)+(\vec{a}_i(t)+\vec{a}_i(t+\Delta t))\,\Delta t/2$$

$$\vec{a}_i(t)=\vec{F}_i(t)/m_i = -\vec{\nabla}_i\, U(t)/m_i$$

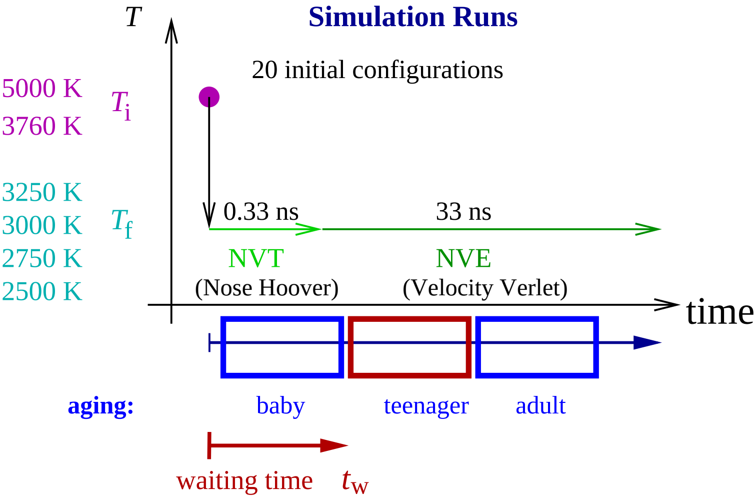
Simulations



Aging


Simulation Runs
T
20 initial configurations
5000 K
3760 K
T_i
3250 K
3000 K
2750 K
2500 K
T_f
0.33 ns
33 ns
NVT
(Nose Hoover)
NVE
(Velocity Verlet)
time
aging:
baby
teenager
adult

Aging



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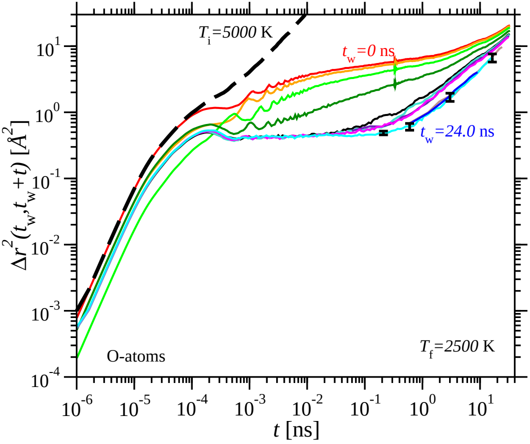
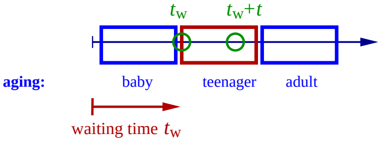
time
aging:
baby
teenager
adult
waiting time
t_w

Aging Example: Mean Square Displacement

$$\Delta r^2(t_w, t_w + t) = \frac{1}{N} \sum_{i=1}^{N} (\mathbf{r}_i(t_w + t) - \mathbf{r}_i(t_w))^2$$





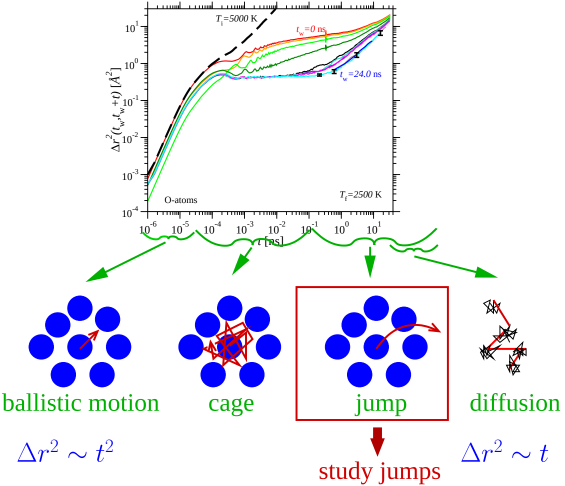
- mean square displacement Δr^2 depends on waiting time t_w (colors) [KVL et al., 2010]
- similarly for $C_q(t_w, t_w + t)$ and $\chi_4(t_w, t_w + t)$ (see talk of Chris Gorman)

Goal: Single Particle Picture (not $\frac{1}{N} \sum_{i=1}^{N}$)

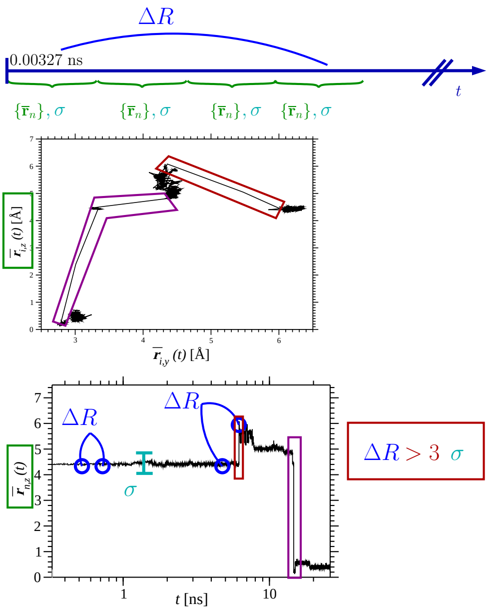
Goal: Single Particle Picture

Cage Picture

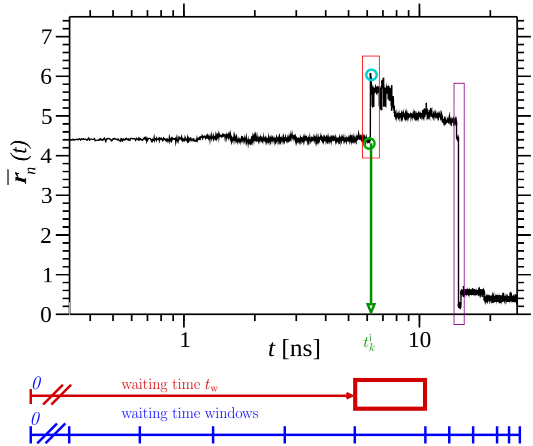
$$\Delta r^2(t_\mathrm{w}, t_\mathrm{w}+t) = \frac{1}{N}\sum_{i=1}^{N}(\mathbf{r}_i(t_\mathrm{w}+t) - \mathbf{r}_i(t_\mathrm{w}))^2$$



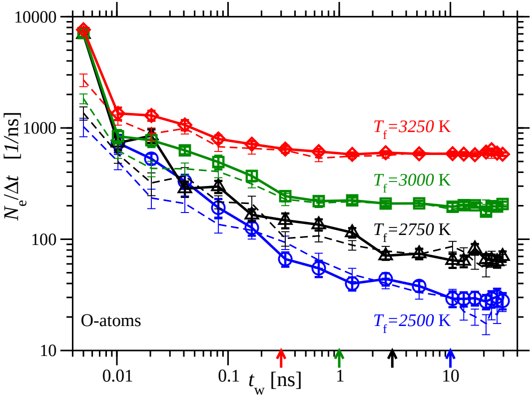
Jump Definition

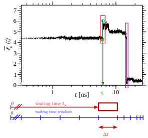


Jump Definition: Aging Dependence



Number of Jumping Particles per Time



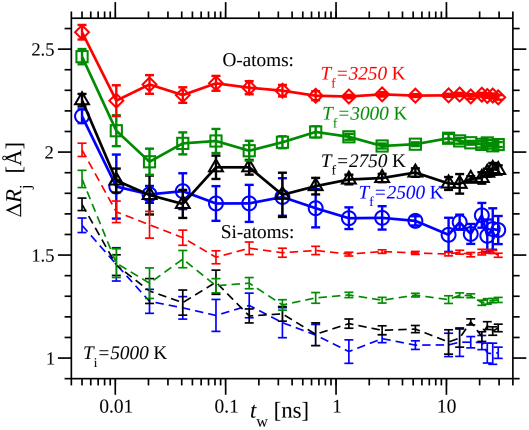


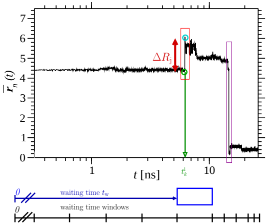
$N_e/\Delta t$ = Number of jump events occuring in time interval Δt

- equilibration time consistent with C_q, Δr^2, and χ_4 (see arrows)
- $N_e/\Delta t$ decreasing with increasing t_w

$\longrightarrow$ $N_e/\Delta t$ depends strongly on waiting time t_w

Average Jump Length

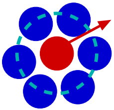




- O-atoms jump farther than Si-atoms
- compare:

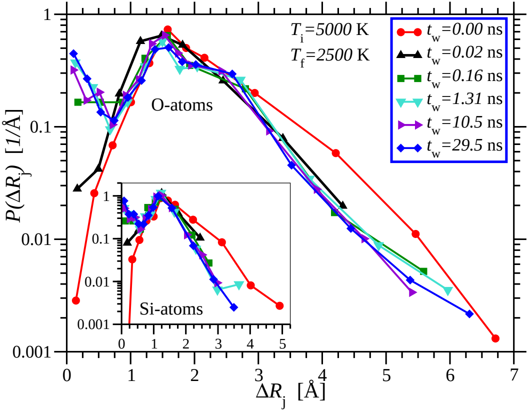
  $d_{\mathrm{OSi}} = 1.608\text{Å}$, $d_{\mathrm{OO}} = 2.626\text{Å}$,

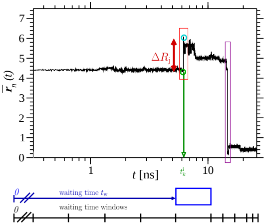
  $d_{\mathrm{SiSi}} = 3.077\text{Å}$



$\longrightarrow$ ΔR_{j} is mostly independent of t_{w}

Jump Length Distribution

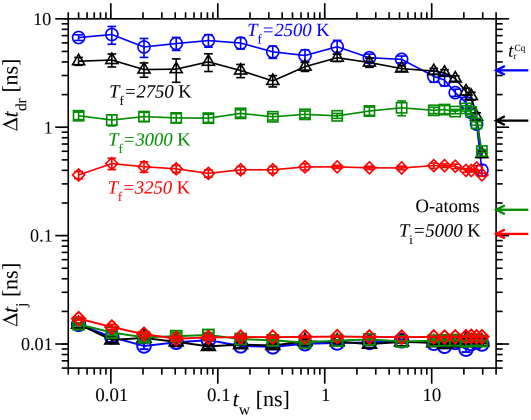


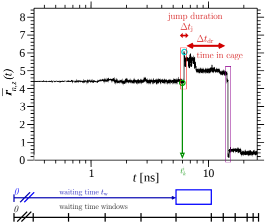


- peak at $\Delta R_\mathrm{j} = 0$: reversible jumps
- exponential decay
- compare LJ & Polymer [Warren & Rottler, EPL 2009]

$\longrightarrow P(\Delta R_\mathrm{j})$ is independent of t_w (colors)

Time Averages: Jump Duration Δt_{j} & Time in Cage Δt_{dr}

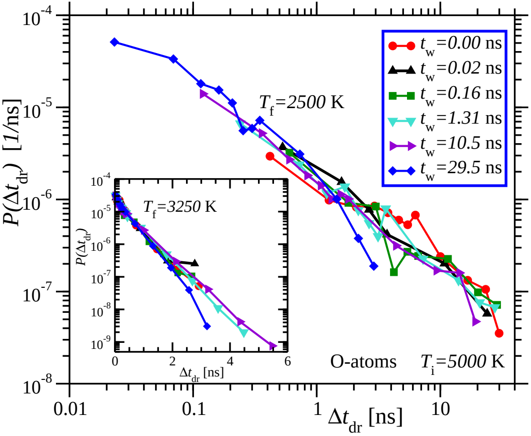


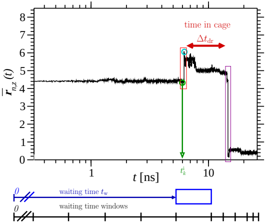


- $\Delta t_{\mathrm{dr}} \gg \Delta t_{\mathrm{j}}$
- no artifact due to finite time-window
- artifact due to finite simulation run

$\longrightarrow$ Δt_{dr} is independent of t_{w}!

Distribution of Time in Cage $P(\Delta t_{dr})$

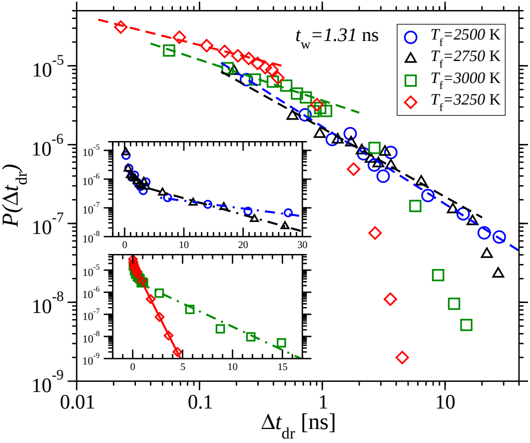


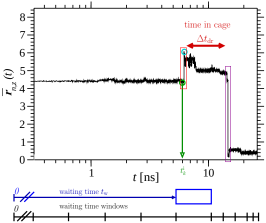


- 2500 K: powerlaw
  compare LJ & Polymer
  [Warren & Rottler, EPL 2009]
- 3250 K: exponential

$\longrightarrow$ $P(\Delta t_{dr})$ is independent of t_w!

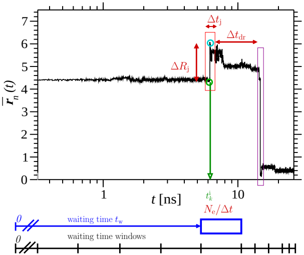
Distribution of Time in Cage $P(\Delta t_{dr})$





- power law to exponential
- compare:
  1 d FA-& East-model
  (dynamic fascilitation)
  [Jung et. al., JCP 2005]

Summary



Aging:

Main waiting time t_w-dependence due to number of jump events per time $N_e/\Delta t$. (not ΔR_j, Δt_j, Δt_{dr})

$\longrightarrow$ Cooperative Processes:

- space-time clusters introduce t_w-dependence of length & time
- dynamic heterogeneity [Donati et al.,PRL 1998]
- avalanches [KVL,Baker, EPL 2006]
- dynamic fascilitation [Jung et al.,JCP 2005]

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