Simulation of Aging in SiO_2: Single Particle Jump Analysis

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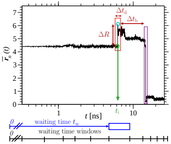
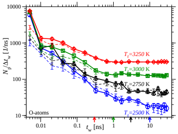




DPG-Tagung Regensburg, March 12, 2013

Model: BKS Potential

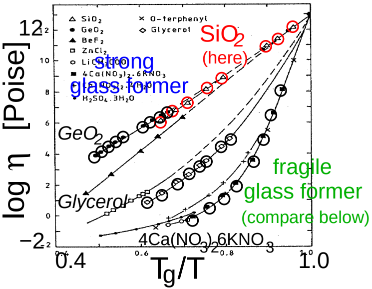
[B.W.H. van Beest *et al.*, PRL 64, 1955 (1990)]

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

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

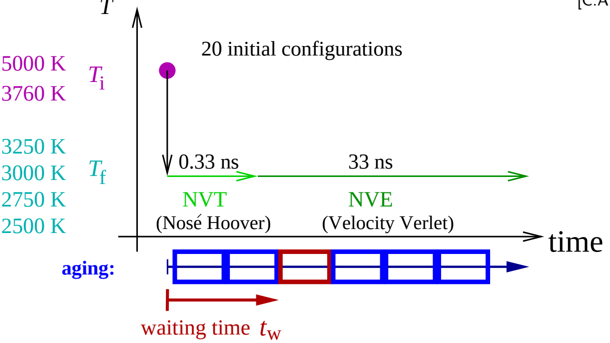
$T_c = 3330$ K

Dynamics:



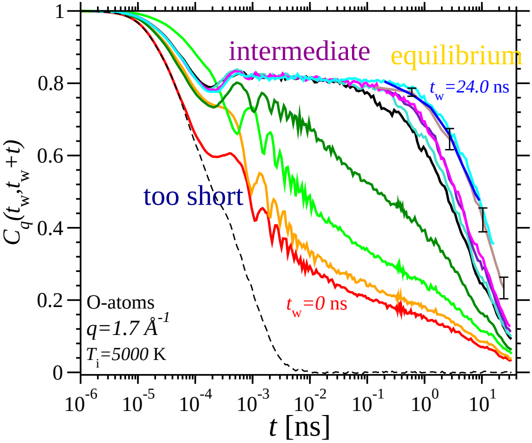
[C.A. Angell et al. 1976]

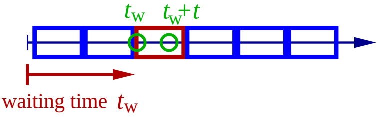
Simulation Runs:



Generalized Intermediate Incoherent Scattering Function

$$C_q(t_\mathrm{w}, t_\mathrm{w}+t) = \frac{1}{N_\alpha}\sum_{j=1}^{N_\alpha} \mathrm{e}^{i\vec{q}\cdot(\vec{r}_j(t_\mathrm{w}+t)-\vec{r}_j(t_\mathrm{w}))}$$



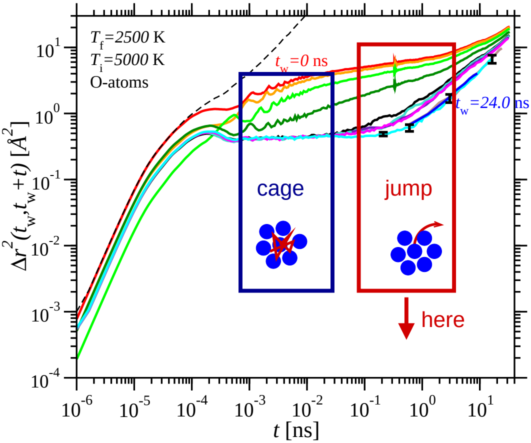


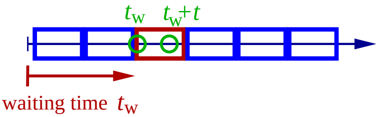
- $C_q(t_\mathrm{w}, t_\mathrm{w}+t)$ depends on waiting time t_w (colors)
- three time windows:
  - too short
  - intermediate (scaling)
  - equilibrium (t_eq^C)
- equilibrium curve included in scaling

[KVL, J. Roman, J.Horbach, PRE 81, 061203 (2010)]

Mean Square Displacement

$$\Delta r^2(t_\mathrm{w}, t_\mathrm{w}+t) \;=\; \frac{1}{N}\sum_{j=1}^{N}\left(\mathbf{r}_j(t_\mathrm{w}+t)-\mathbf{r}_j(t_\mathrm{w})\right)^2$$





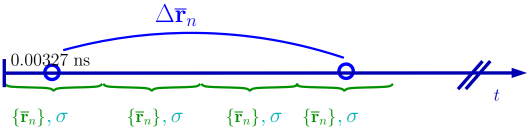
- $\Delta r^2(t_\mathrm{w}, t_\mathrm{w}+t)$ depends on waiting time t_w (colors)
- three time windows

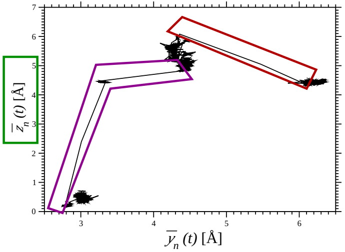
Goal:

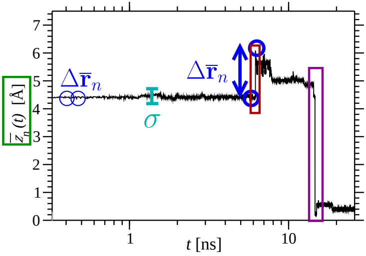
Single Particle Picture

(not $\frac{1}{N}\sum_{j=1}^{N}$)

Jump Definition



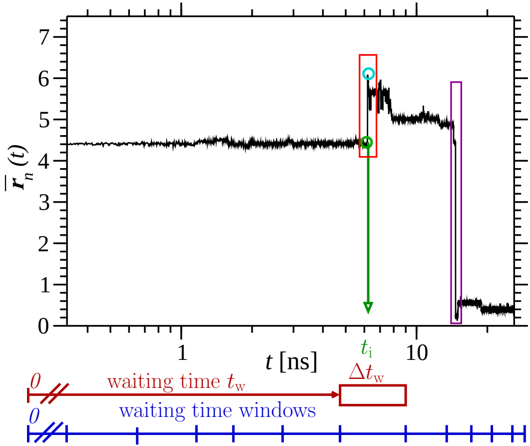




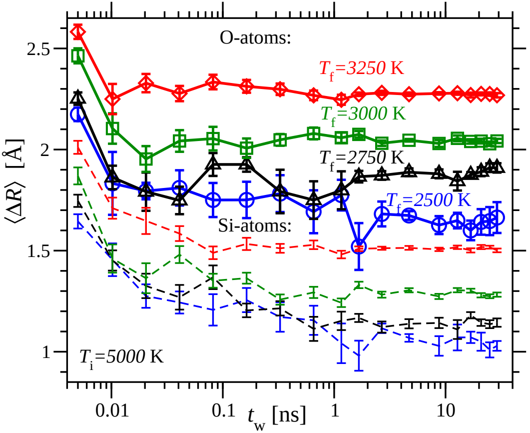
$$\Delta\overline{\mathbf{r}}_n > 3\ \sigma$$

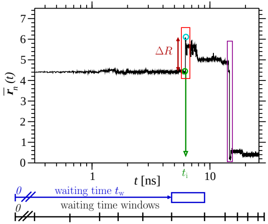
[KVL, R. Bjorkquist, L.M. Chambers, PRL 110, 017801 (2013)]

Jump Definition: Aging Dependence



Average Jump Length



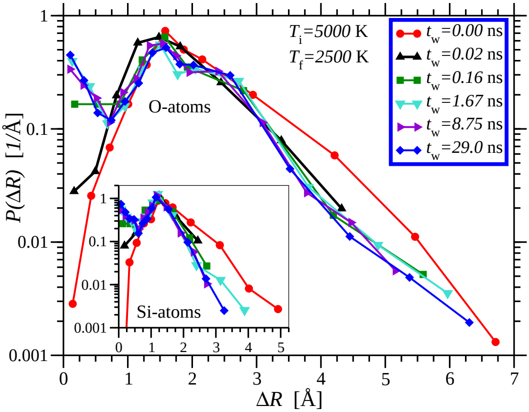


- O-atoms jump farther than Si-atoms
- compare:

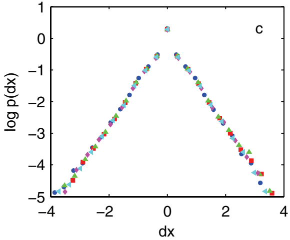
  $d_{\mathrm{SiO}} = 1.6 \mathring{A}$, $d_{\mathrm{OO}} = 2.6 \mathring{A}$, $d_{\mathrm{SiSi}} = 3.1 \mathring{A}$
- ΔR mostly independent of t_{w}

Jump Length Distribution

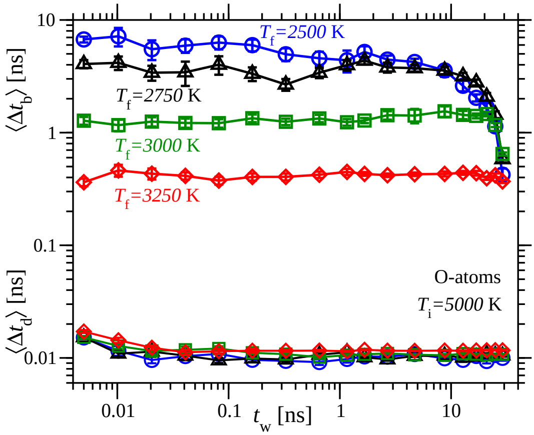
strong glass former SiO_2:

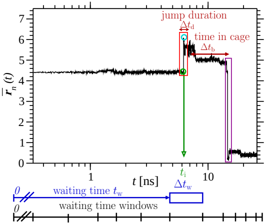


- $P(\Delta R)$ independent of t_w
- exponential decay
- compare fragile glassformer binary LJ (& polymer) [Warren & Rottler,EPL(2009)]



Time Averages: Jump Duration Δt_{d} & Time in Cage Δt_{b}

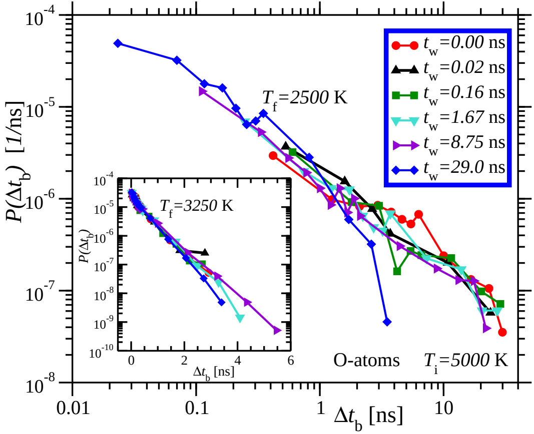




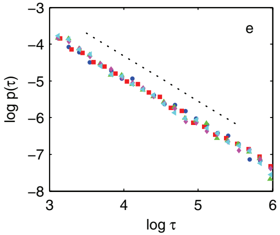
- $\Delta t_{\mathrm{b}} \gg \Delta t_{\mathrm{d}}$
- $t_{\mathrm{w}} \gtrapprox 10$ artifact due to finite simulation run
- Δt_{b} independent of t_{w}!

Distribution of Time in Cage $P(\Delta t_b)$: t_w varied

strong glass former SiO_2:

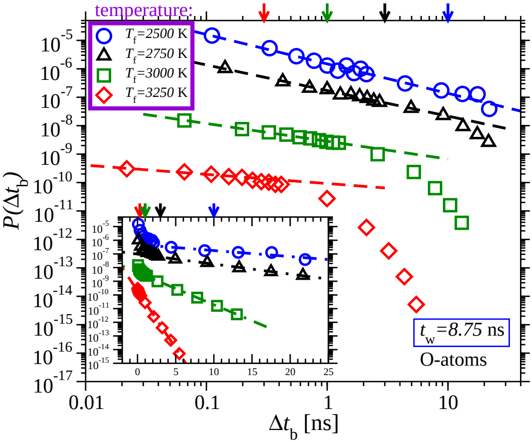


- $P(\Delta t_b)$ independent of t_w!
- compare fragile glassformer (binary LJ &) polymer [Warren & Rottler,EPL(2009)]



Distribution of Time in Cage $P(\Delta t_\mathrm{b})$: T_f varied

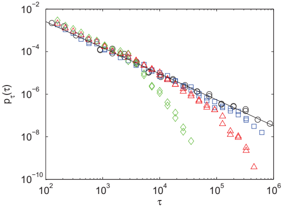
strong glass former SiO_2:



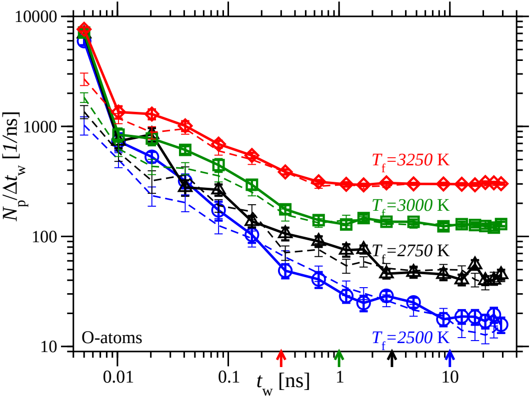
- crossover
  - power law to exponential
  - at $t_\mathrm{cross} \approx t_\mathrm{eq}^C$
- compare fragile glassformer

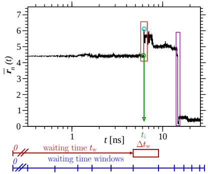
  binary LJ

  [Warren & Rottler, PRL 2013]



Number of Jumping Particles per Time

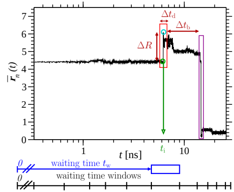


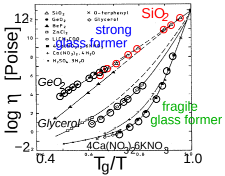


- $N_p/\Delta t_w$ depends strongly on waiting time t_w
  - $N_p/\Delta t_{tw}$ decreasing with increasing t_w
  - compare: soft colloids [Yunker et al.,PRL (2009)]
- equilibration at t_{eq}^{j}

  $t_{eq}^{j} \approx t_{eq}^{C}$ (arrows)

Summary: Microscopic Picture of Aging





Aging of SiO_2:

- Only t_w-dependence: $N_p/\Delta t_w$ (not $P(\Delta R)$ and $P(\Delta t_b)$)
- $P(\Delta t_b)$ crossover power law to exponential
  - at $t_{cross} \approx t_{eq}^{j} \approx t_{eq}^{C}$

[KVL, R. Bjorkquist, L.M. Chambers, PRL 110, 017801 (2013)]

Compare with Fragile Glassformer:

- Surprising similar jump dynamics of strong and fragile glass formers
  - $P(\Delta R)$ and $P(\Delta t_b)$ t_w-independent
  - $P(\Delta t_b)$ crossover

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