Universal Aging Dynamics in SiO_2

Katharina Vollmayr-Lee (Bucknell University),
Robin Bjorkquist (Cornell University), Landon M. Chambers (Texas A&M University), Christopher H. Gorman (UCSB), and Horacio E. Castillo (Ohio University)

UCGP VI, Aspen, February 6, 2015

Outline:

1. Microscropic: Single Particle Jump Dynamics
2. Scaling:
   - global incoherent intermediate scattering function,
   - dynamic susceptibility
   - local incoherent intermediate scattering function

Model: BKS Potential

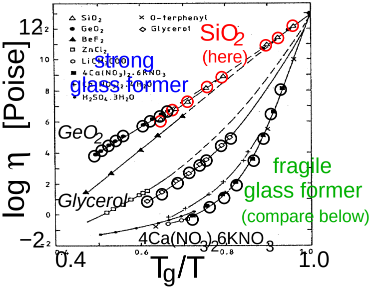
[B.W.H. van Beest *et al.*, PRL 64, 1955 (1990)]

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

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

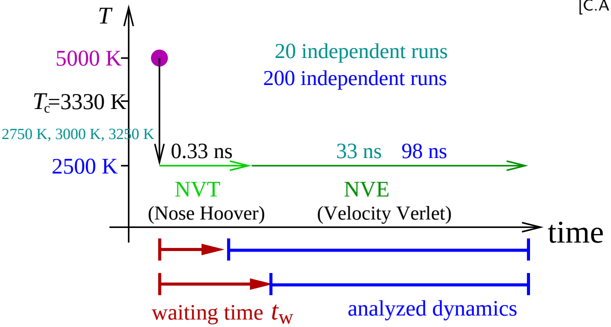
$T_c = 3330$ K

Dynamics:

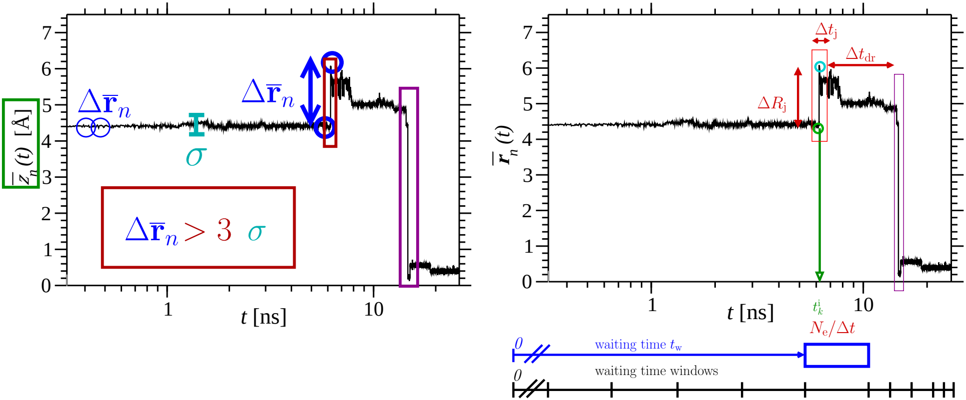


[C.A. Angell et al. 1976]

Simulation Runs:



1. Microscopic: Jump Definition & Jump Statistics



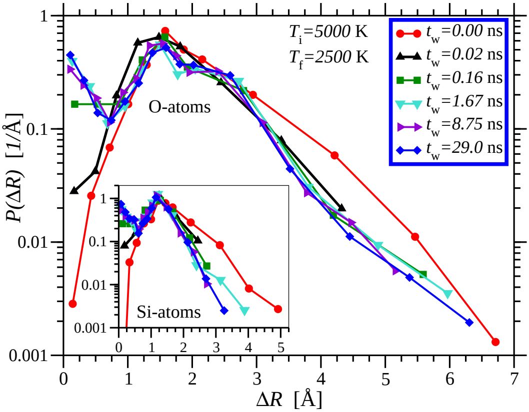
[KVL, R. Bjorkquist, L.M. Chambers, PRL 110, 017801 (2013)]

see also [KVL, J. Chem. Phys. 121, 4781 (2004)]

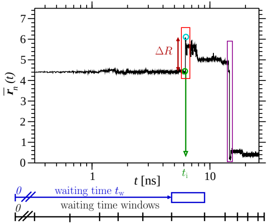
jumps ⟷ defects [KVL & A. Zippelius, PRE 88, 052145 (2013)]

Jump Length Distribution

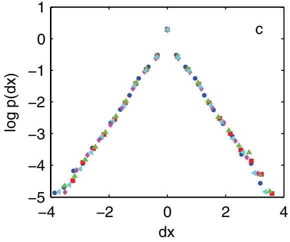
strong glass former SiO_2:



- $P(\Delta R)$ independent of t_w

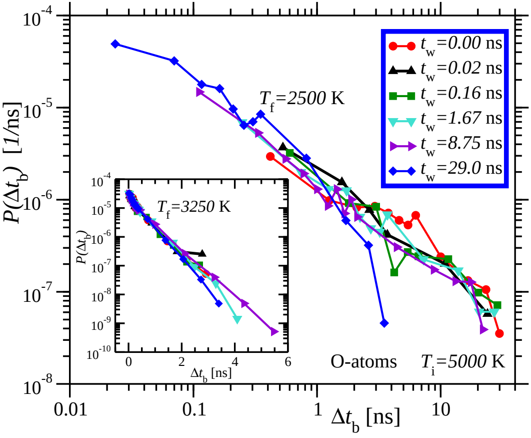


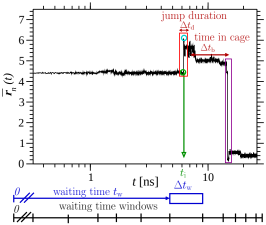
- compare fragile glassformer binary LJ (& polymer) [Warren & Rottler,EPL(2009)]



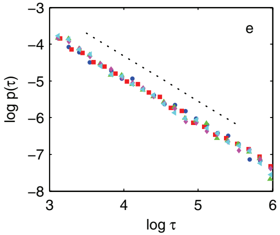
Distribution of Time in Cage $P(\Delta t_\mathrm{b})$: t_w varied

strong glass former SiO_2:





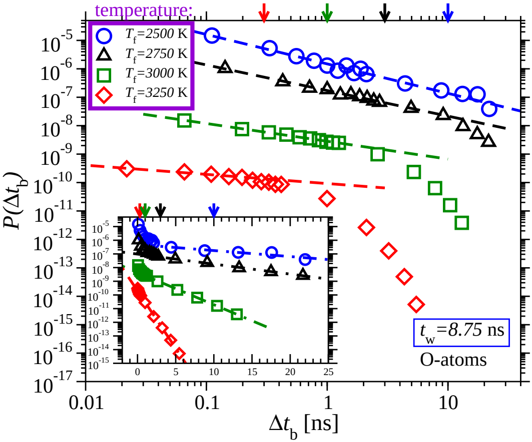
- compare fragile glassformer (binary LJ &) polymer [Warren & Rottler,EPL(2009)]



- $P(\Delta t_\mathrm{b})$ independent of t_w!

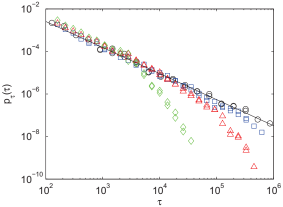
Distribution of Time in Cage $P(\Delta t_b)$: T_f varied

strong glass former SiO_2:



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

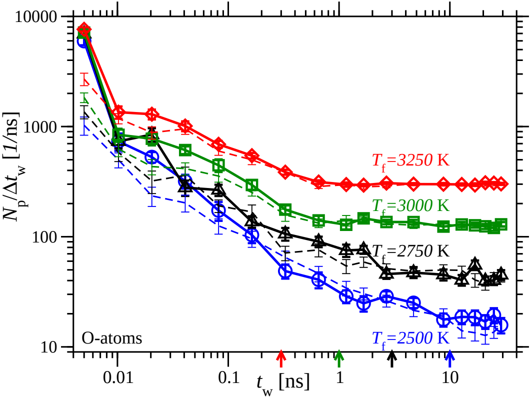
binary LJ [Warren & Rottler,'13]

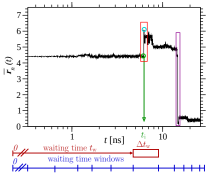


polymer; CTRW

[Helfferich et al. '15]

Number of Jumping Particles per Time





$\longrightarrow$ Only t_w-dependence: $N_p/\Delta t_w$
(not $P(\Delta R)$ and $P(\Delta t_b)$)

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

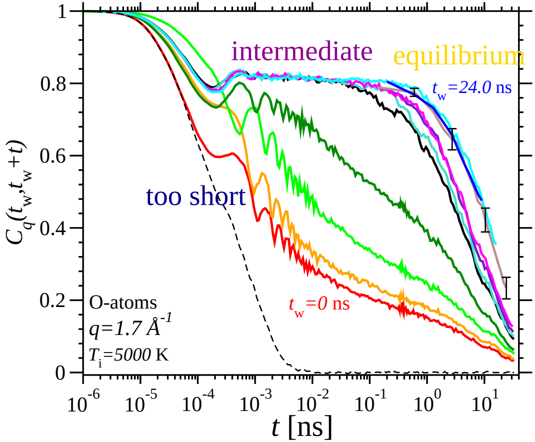
- $N_p/\Delta t_w$ depends strongly on waiting time t_w
  - $N_p/\Delta t_{tw}$ decreasing with increasing t_w
  - compare: soft colloids [Yunker et al.,PRL (2009)] compare polymer; CTRW [Helfferich et al., EPL 2015]
- equilibration at t_{eq}^{j}

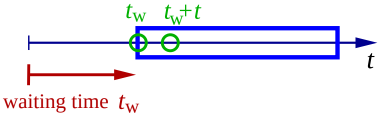
  $t_{eq}^{j} \approx t_{eq}^{C}$ (arrows)

2. Scaling: Global Incoherent Intermediate Scattering Fct.

$$f_s^\alpha(t_w, t_w + t, \mathbf{q}) = \frac{1}{N_\alpha} \sum_{j=1}^{N_\alpha} \cos\left\{\mathbf{q} \cdot \left(\mathbf{r}_j(t_w + t) - \mathbf{r}_j(t_w)\right)\right\}$$

$$C^\alpha = \langle f_s^\alpha(t_w, t_w + t, \mathbf{q}) \rangle$$





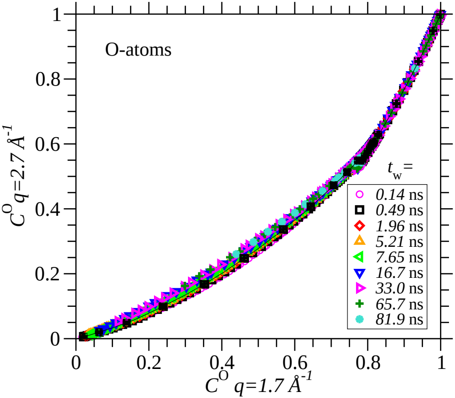
- $C_q(t_w, t_w + t)$ depends on waiting time t_w (colors)
- three time windows:
  - too short
  - intermediate (scaling)
  - equilibrium (t_{eq}^C)
- equilibrium curve included in scaling

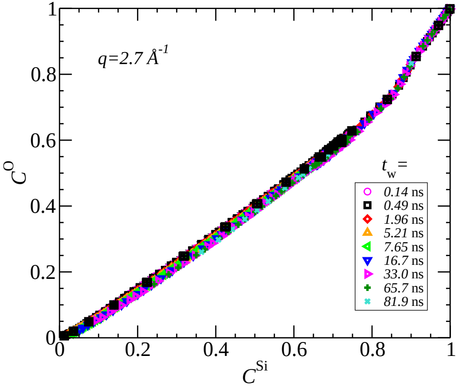
[KVL, J. Roman, J.Horbach, PRE 81, 061203 (2010)]

2. Scaling: Global Incoherent Intermediate Scattering Fct.

$$f_s^\alpha(t_\mathrm{w}, t_\mathrm{w}+t, \mathbf{q}) = \frac{1}{N_\alpha}\sum_{j=1}^{N_\alpha} \cos\left\{\mathbf{q}\cdot\left(\mathbf{r}_j(t_\mathrm{w}+t) - \mathbf{r}_j(t_\mathrm{w})\right)\right\}$$

$C^\alpha = \langle f_s^\alpha(t_\mathrm{w}, t_\mathrm{w}+t, \mathbf{q})\rangle$ α= particle type Si,O





$C^\alpha = C^\alpha(z(t_\mathrm{w}, t), q)$

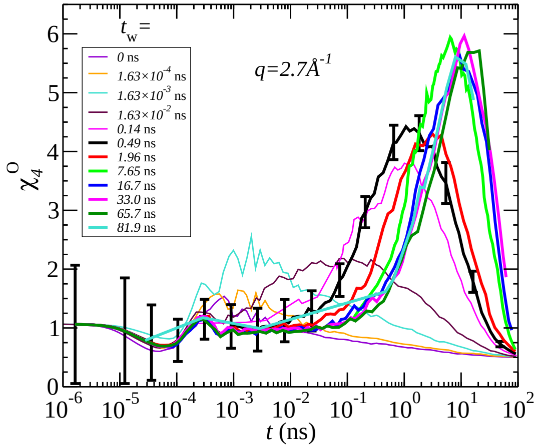
$C_q = C_q(z(t_\mathrm{w}, t), \alpha)$

$$C = C(z(t_\mathrm{w}, t), q, \alpha)$$

Dynamic Susceptibility

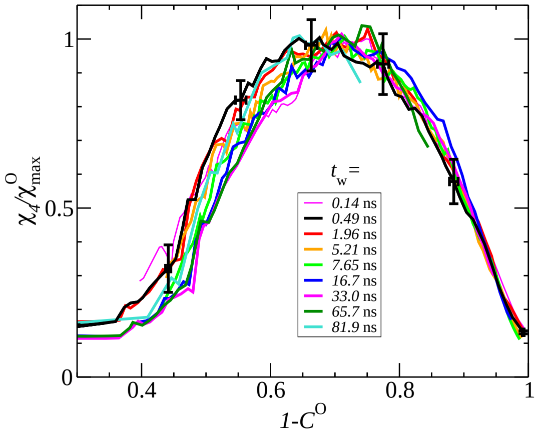
$$f_s^{\alpha}(t_{\rm w}, t_{\rm w}+t, \mathbf{q}) = \frac{1}{N_\alpha}\sum_{j=1}^{N_\alpha} \cos\left\{\mathbf{q}\cdot\left(\mathbf{r}_j(t_{\rm w}+t) - \mathbf{r}_j(t_{\rm w})\right)\right\}$$

$$\chi_4^{\alpha}(t_{\rm w}, t_{\rm w}+t, q) = N_\alpha\left[\left\langle (f_s^\alpha)^2\right\rangle - (\langle f_s^\alpha\rangle)^2\right]$$



Scaling of Dynamic Susceptibility

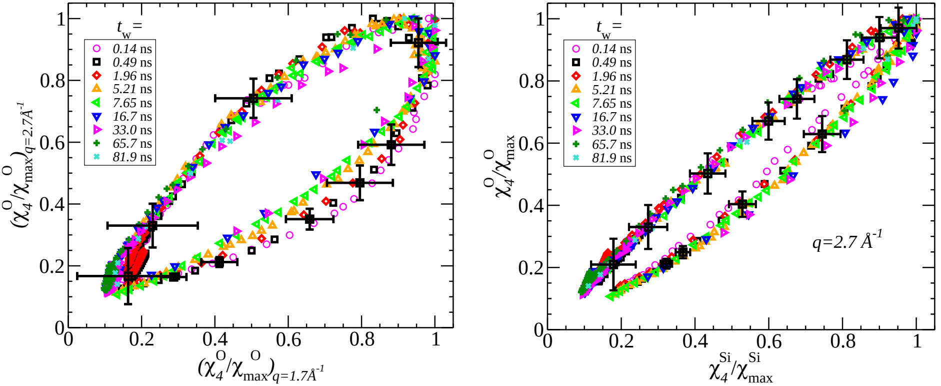
$$\chi_4^\alpha(t_w, t_w+t, q) = N_\alpha \left[\left\langle (f_s^\alpha)^2 \right\rangle - (\langle f_s^\alpha \rangle)^2 \right]$$



- theory:
  $\chi_4/\chi_4^{\max} = \chi_4/\chi_4^{\max}(C)$
  [Castillo et.al '02 – '04,'08]
- compare fragile glassformers:
  [Parsaeian & Castillo, '08,'09]

Scaling of Dynamic Susceptibility

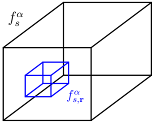
$$\chi_4^\alpha(t_\mathrm{w}, t_\mathrm{w}+t, q) = N_\alpha \left[\left\langle (f_s^\alpha)^2 \right\rangle - (\langle f_s^\alpha \rangle)^2 \right]$$

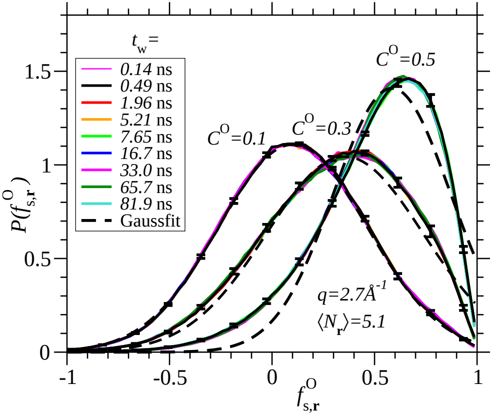


$$\longrightarrow \chi_4/\chi_4^{\max} = \chi_4/\chi_4^{\max}\left(C(z(t_\mathrm{w}, t), q, \alpha)\right)$$

Local Incoherent Intermediate Scattering Function

$$f^{\alpha}_{s,\mathbf{r}}(t_{\mathrm{w}}, t_{\mathrm{w}} + t, \mathbf{q})) = \frac{1}{N^{\alpha}_{\mathbf{r}}} \sum_{\mathbf{r}_j(t_{\mathrm{w}}) \in B_{\mathbf{r}}} \cos\left(\mathbf{q} \cdot [\mathbf{r}_j(t_{\mathrm{w}} + t) - \mathbf{r}_j(t_{\mathrm{w}})]\right)$$

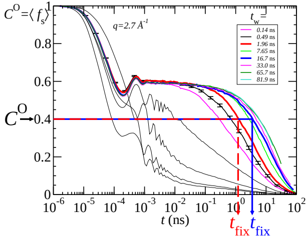




Theory:

$P(f_{s,\mathbf{r}})$ scales with C^{α}

[Castillo et al.'02,'03; Chamon et al. '04]

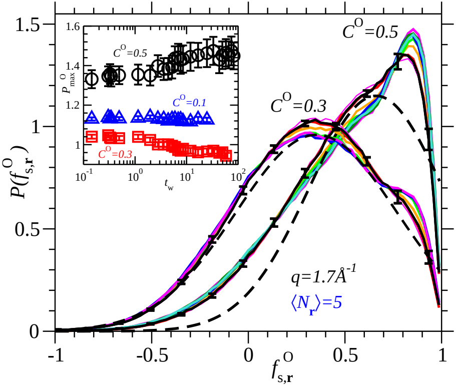


compare fragile & spin glasses:

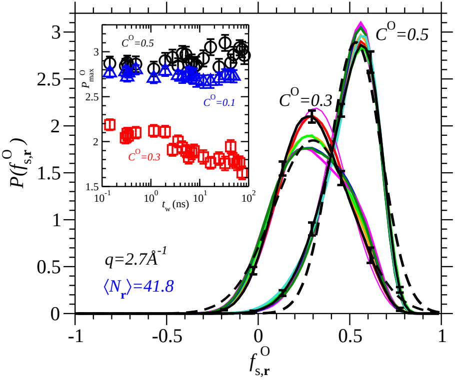
[Castillo & Parsaeian, '07, '09] & [Castillo & Chamon et al., '02 – '04]

NOT Scaling of Local Incoh. Intermediate Scattering Fct.

$$f^{\alpha}_{s,\mathbf{r}}(t_{\mathrm{w}}, t_{\mathrm{w}} + t, \mathbf{q})) = \frac{1}{N^{\alpha}_{\mathbf{r}}} \sum_{\mathbf{r}_j(t_{\mathrm{w}}) \in B_{\mathbf{r}}} \cos\left(\mathbf{q} \cdot [\mathbf{r}_j(t_{\mathrm{w}} + t) - \mathbf{r}_j(t_{\mathrm{w}})]\right)$$



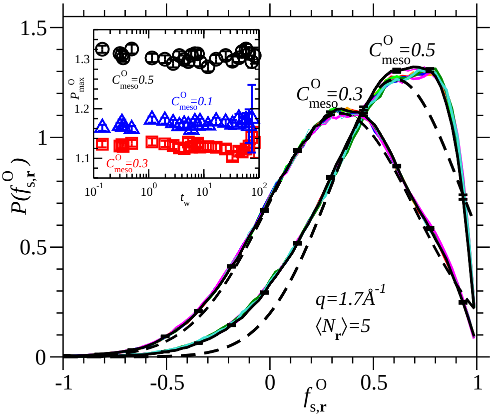
wave vector q too large

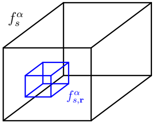


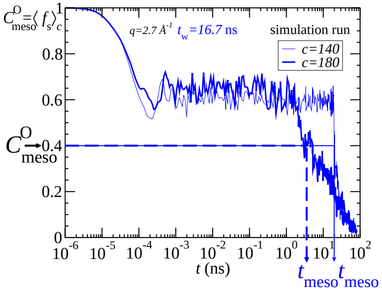
subbox $B_{\mathbf{r}}$ too large

Scaling of Local Incoh. Intermediate Scattering Fct.

$$f^{\alpha}_{s,\mathbf{r}}(t_{\rm w}, t_{\rm w}+t, \mathbf{q})) = \frac{1}{N^{\alpha}_{\mathbf{r}}} \sum_{\mathbf{r}_j(t_{\rm w}) \in B_{\mathbf{r}}} \cos\left(\mathbf{q}\cdot[\mathbf{r}_j(t_{\rm w}+t) - \mathbf{r}_j(t_{\rm w})]\right)$$







$\longrightarrow$ $P(f^{\alpha}_{s,\mathbf{r}}(t_{\rm w}, t))$ scales with $C^{\alpha}_{\rm meso}(t_{\rm w}, t)$

Summary:

Aging Dynamics of Strong Glass Former SiO_2:

1. Microscopic: Single Particle Jump Dynamics
   - Only t_w-dependence: $N_\mathrm{p}/\Delta t_\mathrm{w}$ (not $P(\Delta R)$ and $P(\Delta t_\mathrm{b})$)
2. Scaling:
   - global incoh. interm. scattering function $C = C(z(t_\mathrm{w}, t), q, \alpha)$ α=Si,O
   - dynamic susceptibility $\chi_4/\chi_4^{\max} = \chi_4/\chi_4^{\max}(C(z(t_\mathrm{w}, t), q, \alpha))$
   - local incoh. interm. sc. fct. distribution $P(f_{s,\mathbf{r}})$ scales with C
3. similar aging dynamics of strong & fragile glasses

Acknowledgments:

Supported by SFB 602, FOR1394, NSF REU grants PHY-0552790, PHY-0997424, PHY-1156964, and DOE Grant No. DE-FG02-06ER46300. KVL thanks A. Zipplelius & University Göttingen.