Universal Aging Dynamics of Silica

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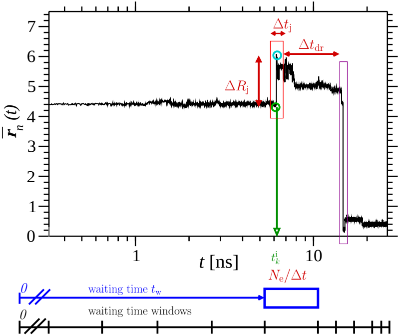
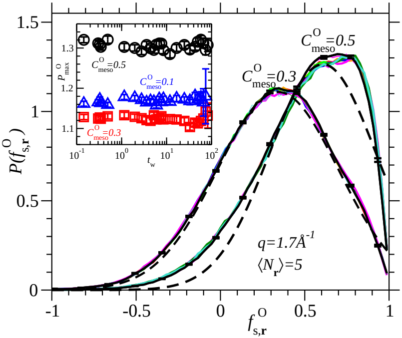
Ohio University, April 6, 2017

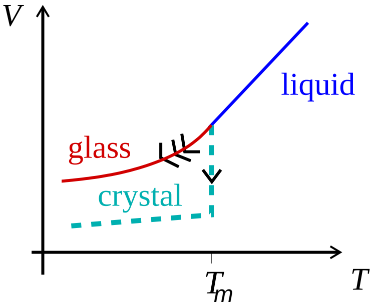




Acknowledgments:

H. E. Castillo, C. H. Gorman, R. Bjorkquist, L. M. Chambers

Introduction: Glass

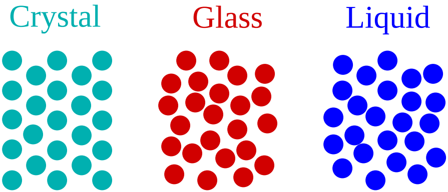


Examples:

- drinking glass
- golf club
- lenses, fiber optics
- vulcanic glass

Glass:

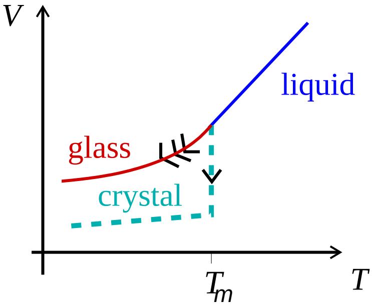
Out of Equilibrium



Structure: disordered

Dynamics: frozen in

Introduction: Glass



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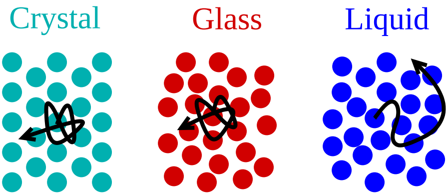

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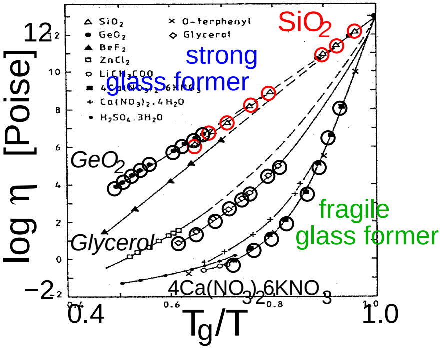


[C.A. Angell and W. Sichina, Ann. NY Acad. Sci. 279, 53 (1976)]

Dynamics:

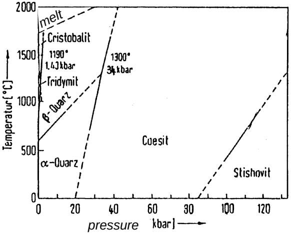
Viscocity η as function of inverse temperature T

- slowing down of many decades $\longrightarrow$ very interesting dynamics
- large variety of glass formers
- strong and fragile glass formers
  Here: SiO$_2$ (strong glass former)
  Below: comparison with fragile glass former

System: Silica (SiO_2)

Special Properties:

- sand and window glass
- similar to water (H_2O):
    - rich phase diagram
    - density maximum

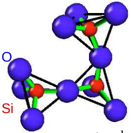


[S. Stoeffler and J. Arndt, Naturwissenschaften 56, 100 (1969)

Model: BKS Potential

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

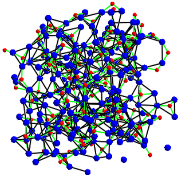
$$\phi(r_{ij}) = \frac{q_i q_j e^2}{r_{ij}} + A_{ij} e^{-B_{ij} r_{ij}} - \frac{C_{ij}}{r_{ij}^6}$$



network
(potential only "spheres")

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

$T_c = 3330$ K



Molecular Dynamics Simulation

Newton’s Equations:

$$\frac{d^2}{dt^2}\vec{r}_i(t) = \vec{a}_i(t) = \vec{F}_i(t)/m_i = -\vec{\nabla}_i\, U(t)/m_i = -\vec{\nabla}_i \sum_{j,k} \phi\,(r_{jk})/m_i$$

Initialize:

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

particles $i=1,\dots,N$

three dimensions

$$\downarrow$$

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

$$\downarrow$$

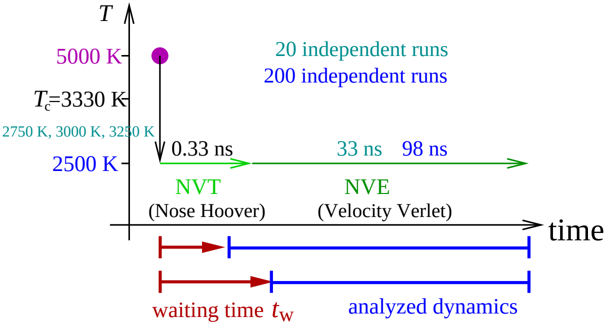
$\vec{r}_i(t_0+2\,\Delta t),\ \vec{v}_i(t_0+2\,\Delta t),\ \vec{a}_i(t_0+2\,\Delta t)$

$\downarrow$ etc.

$\downarrow$ = Iteration Step: (Velocity Verlet)

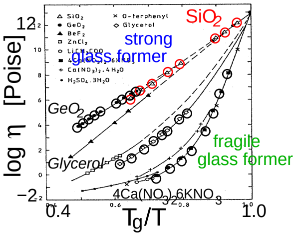
$$\vec{r}_i(t+\Delta t)=\vec{r}_i(t)+\vec{v}_i(t)\Delta t+\vec{a}_i(t)(\Delta t)^2/2$$
$$\vec{v}_i(t+\Delta t)=\vec{v}_i(t)+(\vec{a}_i(t)+\vec{a}_i(t+\Delta t))\Delta t/2$$
$$\vec{a}_i(t)=\vec{F}_i(t)/m_i = -\vec{\nabla}_i\, U(t)/m_i$$

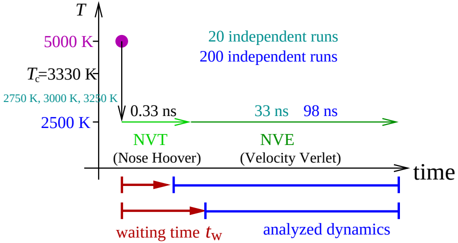
Simulation Runs



Out of Equilibrium ⟶ Dynamics depends on Waiting Time t_{w} (Aging)

Outline: Universal Aging Dynamics of Silica





Aging Dynamics ($\boxed{t_{\mathrm{w}}}$ -Dependence) :

- Microscopic Single Particle Jumps [PRL 2013,EPL 2015]
- Scaling of Dynamic Heterogeneities [JCP 2016]

Universality:

- Here SiO_2, compare with fragile glass formers

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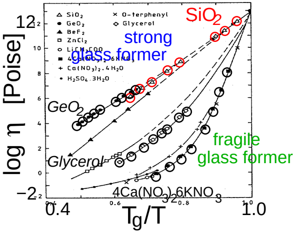

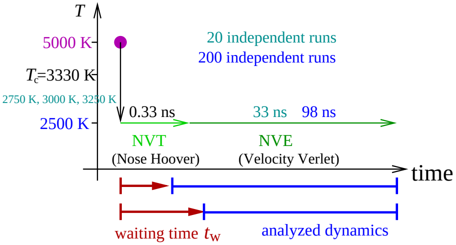

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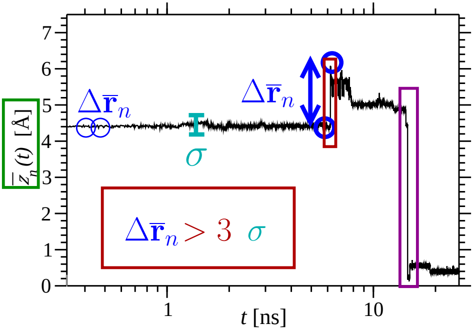
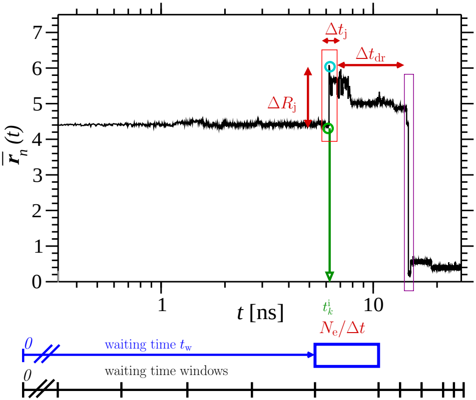
1. Microscopic: Jump Definition & Jump Statistics





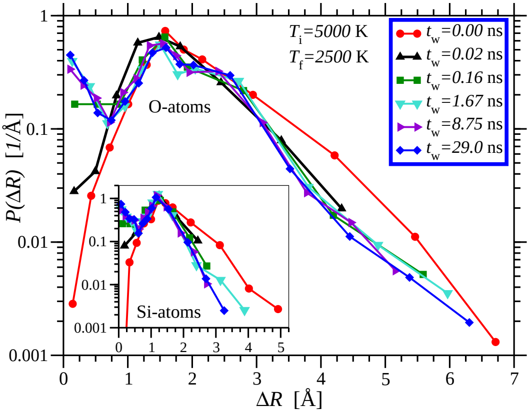
[KVL, R. Bjorkquist, L.M. Chambers, PRL 110, 017801 (2013)]

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

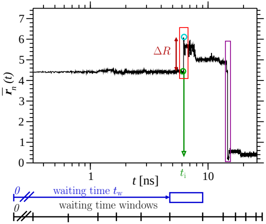
jumps ⟷ defects [KVL & A. Zippelius, PRE 88, 052145 (2013)]

Jump Length Distribution

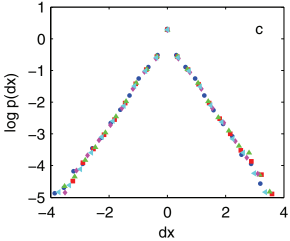
strong glass former SiO_2:



- $P(\Delta R)$ independent of t_{w}

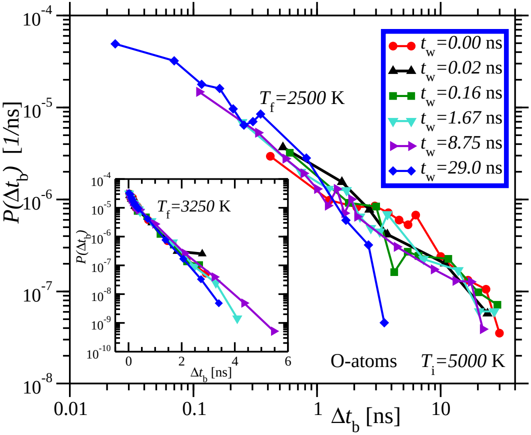


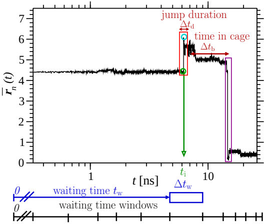
- compare fragile glassformer binary LJ (& polymer) [Warren & Rottler,EPL(2009)]



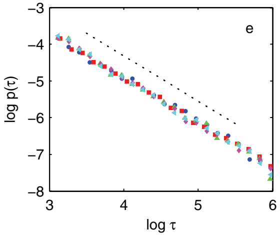
Distribution of Time in Cage $P(\Delta t_{\mathrm{b}})$: t_{w} varied

strong glass former SiO_2:



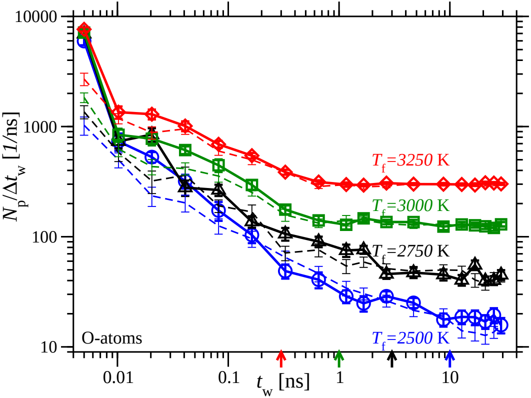


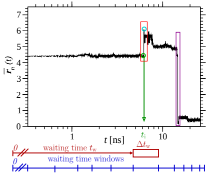
- compare fragile glassformer (binary LJ &) polymer [Warren & Rottler,EPL(2009)]



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

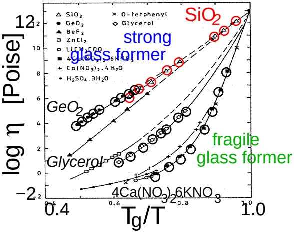
Number of Jumping Particles per Time

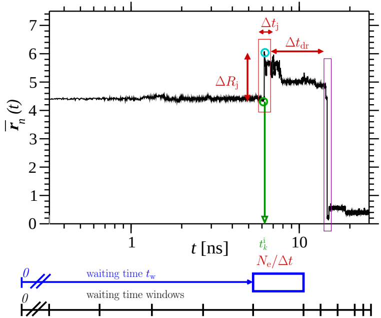




- $N_p/\Delta t_w$ depends strongly on waiting time t_w
  - $N_p/\Delta t_{tw}$ decreasing with increasing t_w
  - compare: soft colloids [Yunker et al.,PRL (2009)]
  - compare fragile glassformer polymer; CTRW [Helfferich et al., EPL 2015]

Summary for Single Particle Jumps





Aging Dynamics (t_w - Dependence):

- Microscopic Single Particle Jumps [PRL 2013,EPL 2015]

  $\longrightarrow$ Only t_w-dependence: $N_p/\Delta t_w$

  (not $P(\Delta R)$ and not $P(\Delta t_b)$)

Universality:

- Similar Single Particle Jump Dynamics for strong and fragile glass formers

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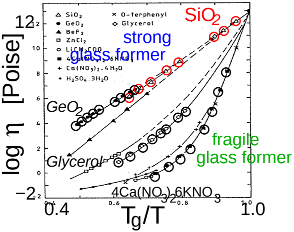

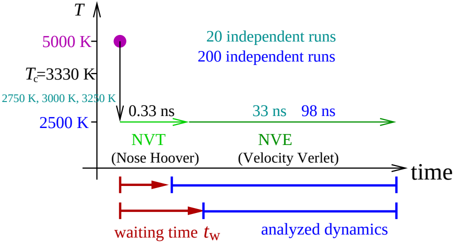

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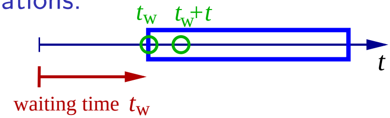
Scaling of Dynamic Heterogeneities

Two-Time Correlations:



Previous Work:

- quantities χ and $P(f_{s,\mathbf{r}})$ depend on two times (t, t_{w})
- fragile glass formers and spin glasses
- (t, t_{w})-dependence is governed by $C(t, t_{\mathrm{w}})$

[Castillo et al. 2002 —]

Universality:

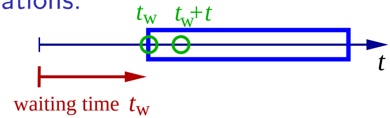
Test here for strong glass former SiO_2

[KVL, Gorman, Castillo, JCP 144, 234510 (2016)]

(JCP Editor's Choice: Paper & Talk)

Scaling of Dynamic Heterogeneities

Two-Time Correlations:



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[Castillo et al. 2002 —]

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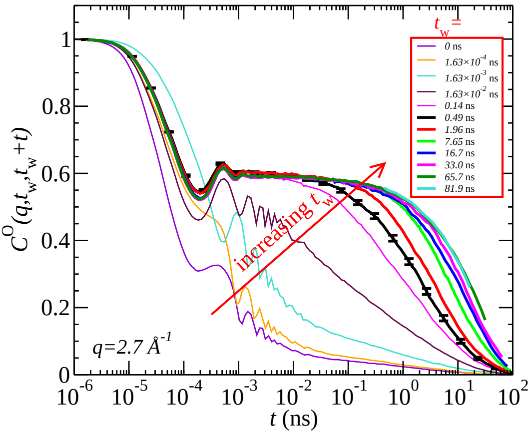
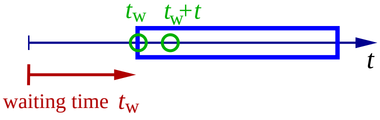
Global Incoherent Intermediate Scattering Function C

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

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





- $C_q(t_\mathrm{w}, t_\mathrm{w}+t)$ depends on waiting time t_w (colors)

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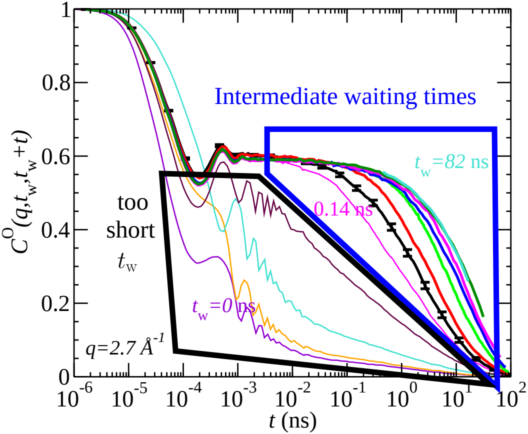
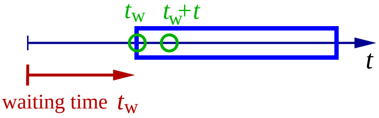
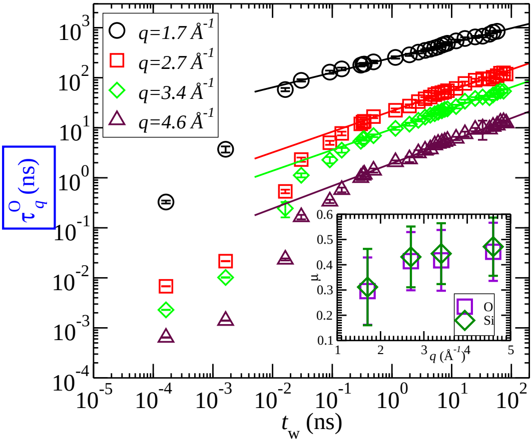
$$C^{\alpha} = \langle f_s^{\alpha}(t_{\mathrm{w}}, t_{\mathrm{w}}+t, \mathbf{q})\rangle$$

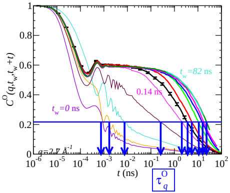




- $C_q(t_{\mathrm{w}}, t_{\mathrm{w}}+t)$ depends on waiting time t_{w} (colors)
- t_{w} time windows:
  - too short
  - intermediate (scaling)
  - equilibrium [KVL et al., PRE (2010)]

Relaxation Time τ_q



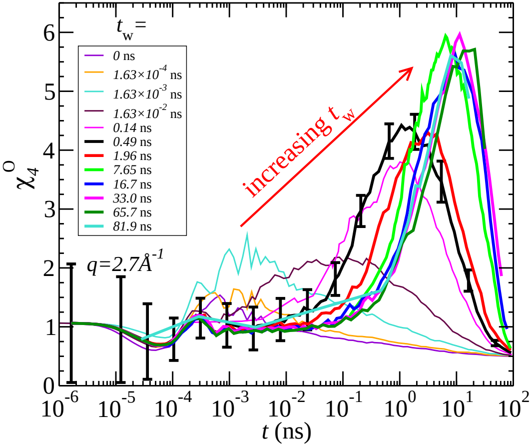


- two t_w time windows → t_w ranges
- power law → same exponent for Si & O ⟶ common aging clock? (see below)

Dynamic Susceptibility χ

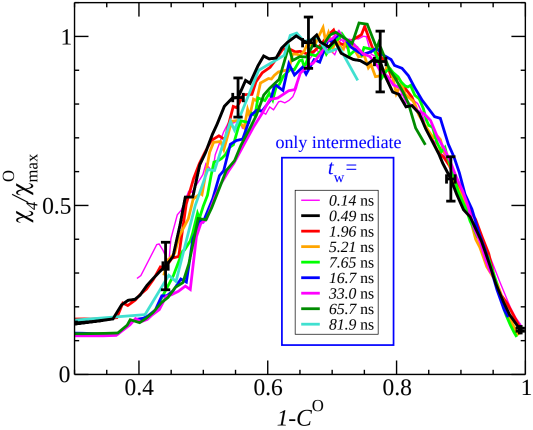
$$f_s^\alpha(t_\mathrm{w}, t_\mathrm{w}+t, \mathbf{q}) = \frac{1}{N_\alpha}\sum_{j=1}^{N_\alpha}\cos\left\{\mathbf{q}\cdot\left(\mathbf{r}_j(t_\mathrm{w}+t)-\mathbf{r}_j(t_\mathrm{w})\right)\right\}$$

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

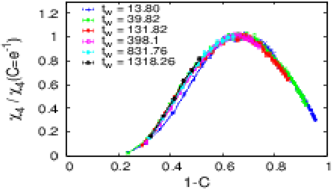


Scaling of Dynamic Susceptibility χ

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



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



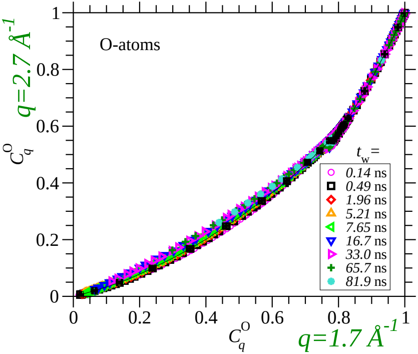
$\longrightarrow$ for SiO$_2$ scaling: $\chi_4^\alpha/\chi_\mathrm{max}^\alpha = \phi(\mathbf{C}(\mathbf{t}, \mathbf{t}_\mathrm{w}), q, \alpha)$ $\longrightarrow$ universal

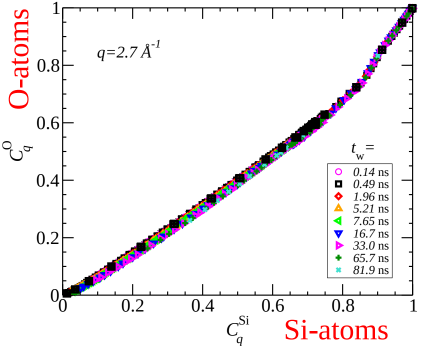
Scaling of Global Incoh. Intermediate Scattering Fct. C

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

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

$\alpha=$ particle type Si,O



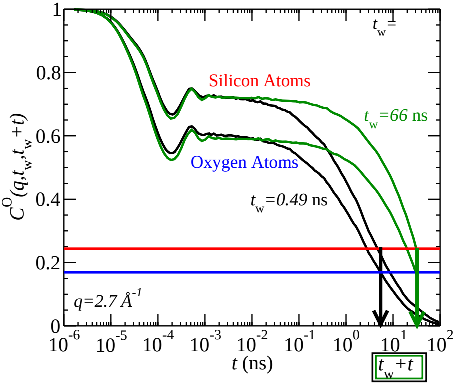


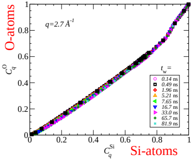
- [Kob,Barrat, EPJB (2000)] for fragile glass former(intermediate t_w)
- for strong glass former SiO$_2$: $C^\alpha = C^\alpha(z(t_\mathrm{w}, t), q)$

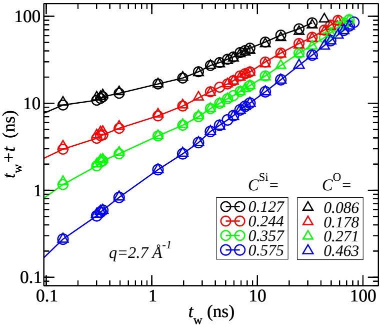
- **directly** particle dependence
- common aging clock
- $C_q = C_q(z(t_\mathrm{w}, t), \alpha)$

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

Particle Type: Common Aging Clock





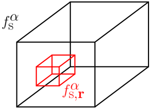


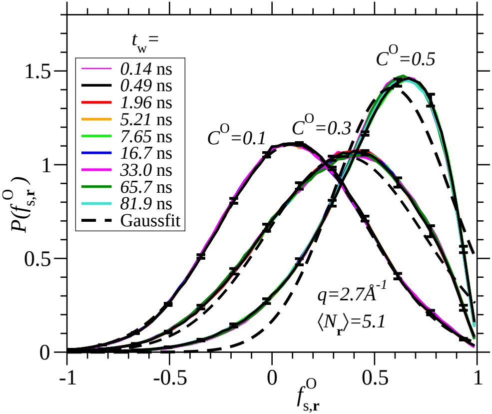
⟶ faster O-atoms and slower Si-atoms have

common aging clock

Local Incoherent Intermediate Scattering Function

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

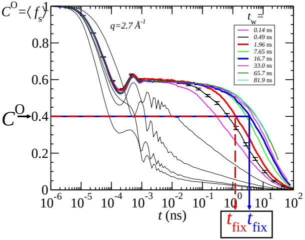




Theory:

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

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

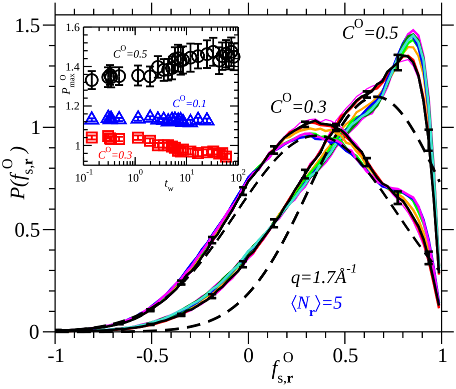


compare fragile & spin glasses: ⟶ universal scaling

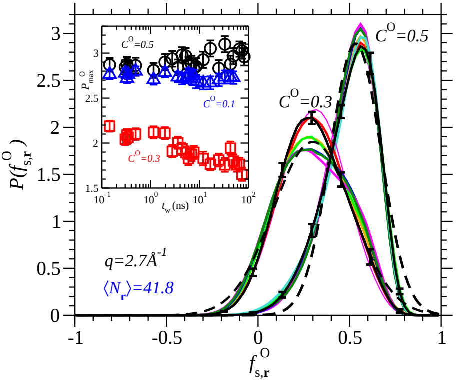
[Castillo & Parsaeian, '07, '09] & [Castillo & Chamon et al., '02 – '04]

NOT Scaling of Local Incoh. Intermediate Scattering Fct.

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



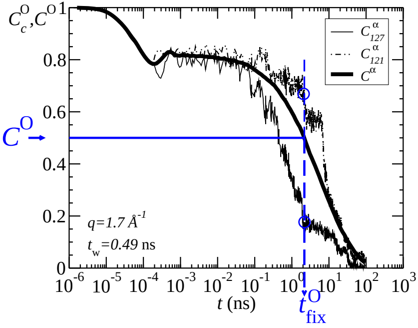
wave vector q too small



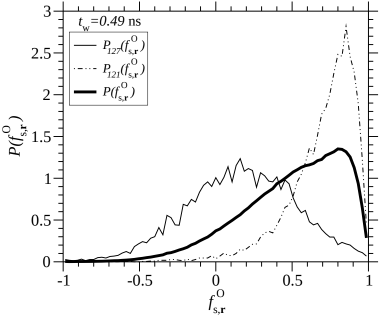
subbox $B_{\mathbf{r}}$ too large

NOT Scaling: Reason

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

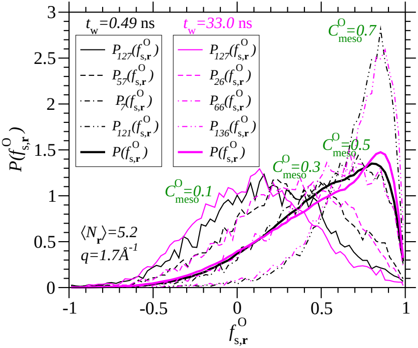


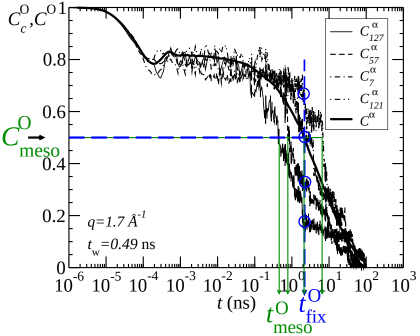
to determine for $c = 1, 2, \ldots, 200$ independent simulation runs $P_c(f_{s,\mathbf{r}})$



NOT Scaling: Reason

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

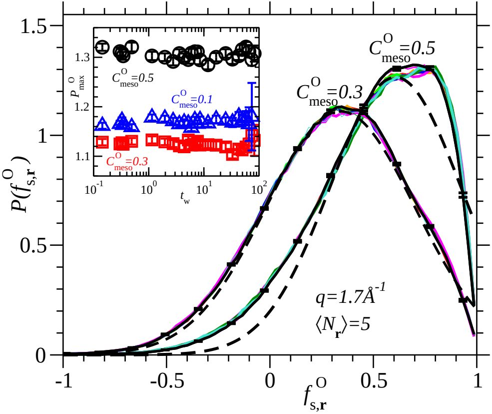


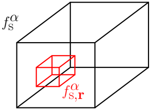


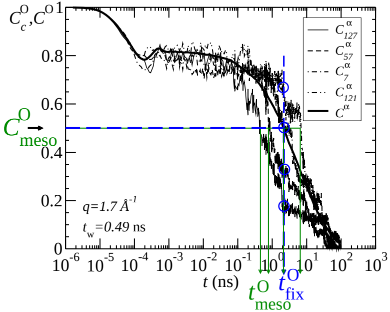
⟶ distribution of distributions

Improved Scaling of Local Incoh. Intermed. 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:

- **Microscopic: Single Particle Jump Dynamics:**
  - Only t_w-dependence: $N_\mathrm{p}/\Delta t_\mathrm{w}$ (not $P(\Delta R)$ and not $P(\Delta t_\mathrm{b})$) [PRL 2013,EPL 2015]
- **Scaling of Dynamic Heterogeneities:**
  - (t, t_w)-dependence of χ and $P(f_{s,\mathbf{r}})$ governed by $C(t, t_\mathrm{w})$
  - $P(f_{s,\mathbf{r}}) \longrightarrow$ intermediate length scale?
  - common aging clock for Si and O

[KVL, CH Gorman, HE Castillo, JCP 144, 234510 (2016)]

Universality:

- Similar aging dynamics of strong & fragile glass formers (microscopic & scaling)

Acknowledgments:

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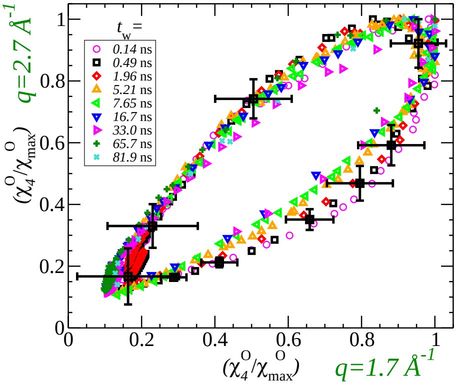
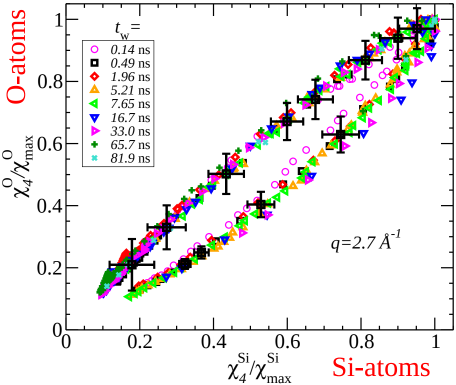
Extra Slides:

Scaling of Dynamic Susceptibility

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





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