Aging to In-Equilibrium Dynamics of SiO_2

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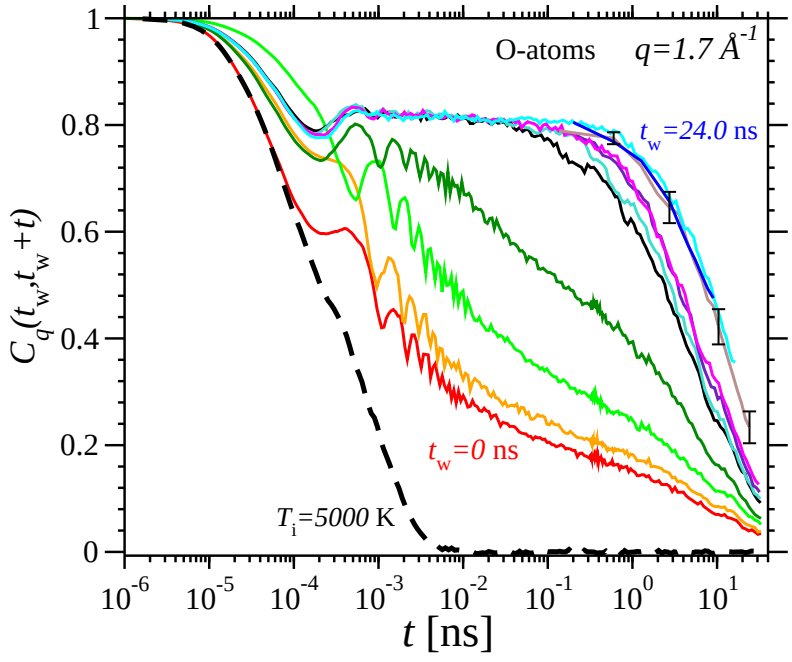


Acknowledgments: A. Zippelius & Institute of Theoretical Physics, University Göttingen, Germany

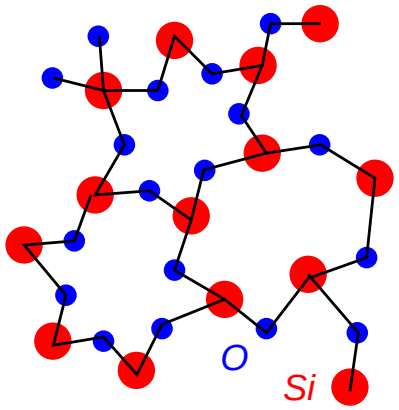
Model & Simulation

SiO_2: BKS Potential

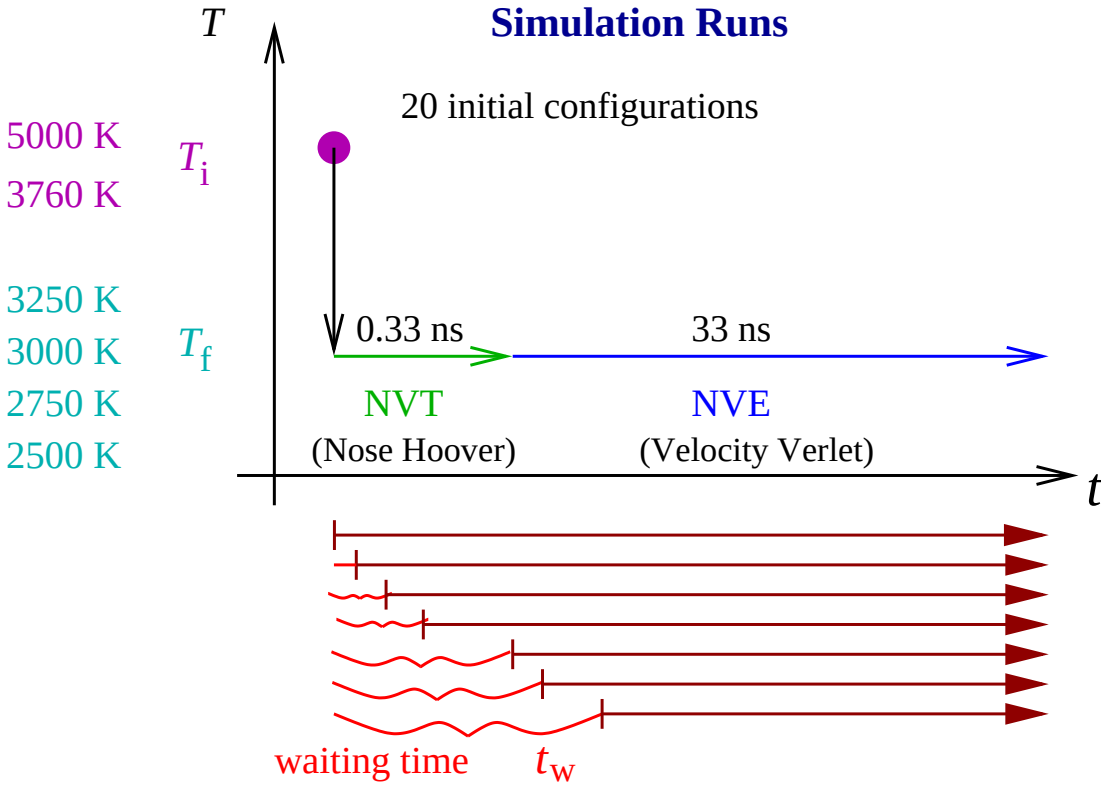
[B.W.H. van Beest *et al.*, PRL 64, 1955 (1990)]

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

$T_c = 3330$ K

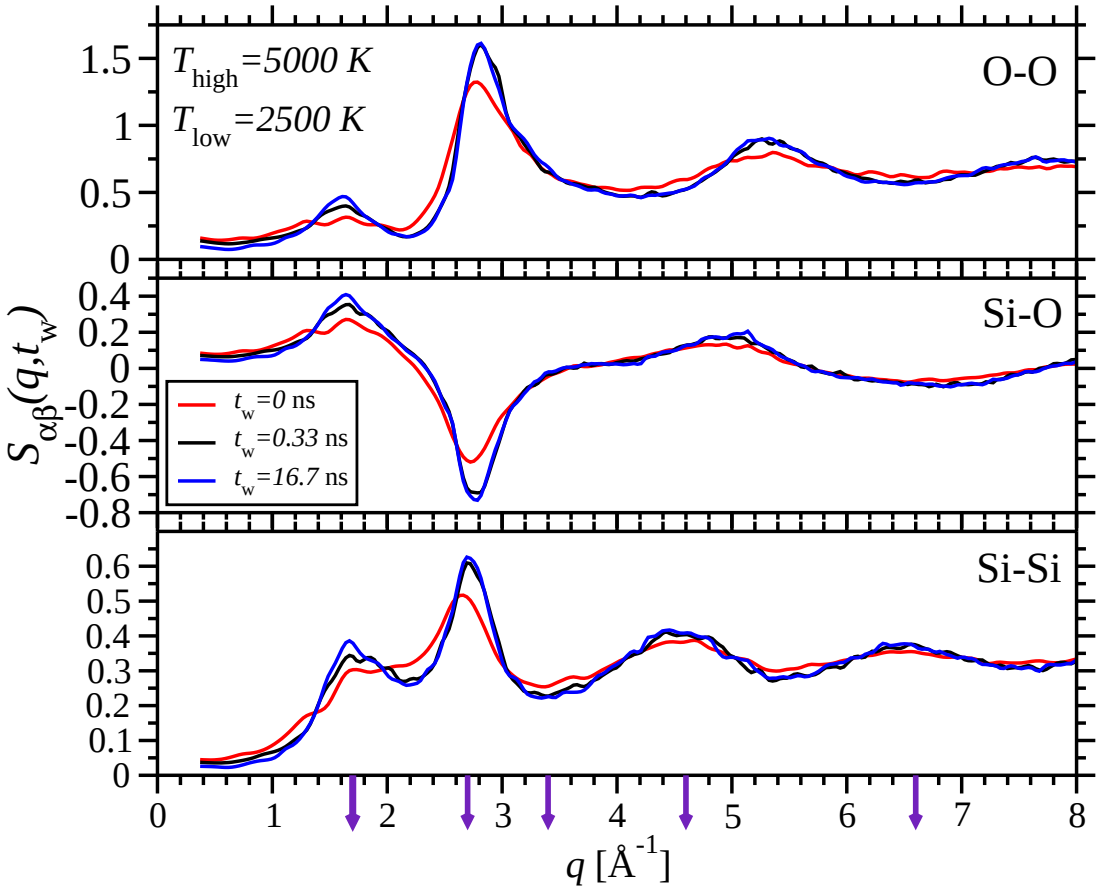


Dynamics: Out-of-Equilibrium to In-Equilibrium



Partial Structure Factors

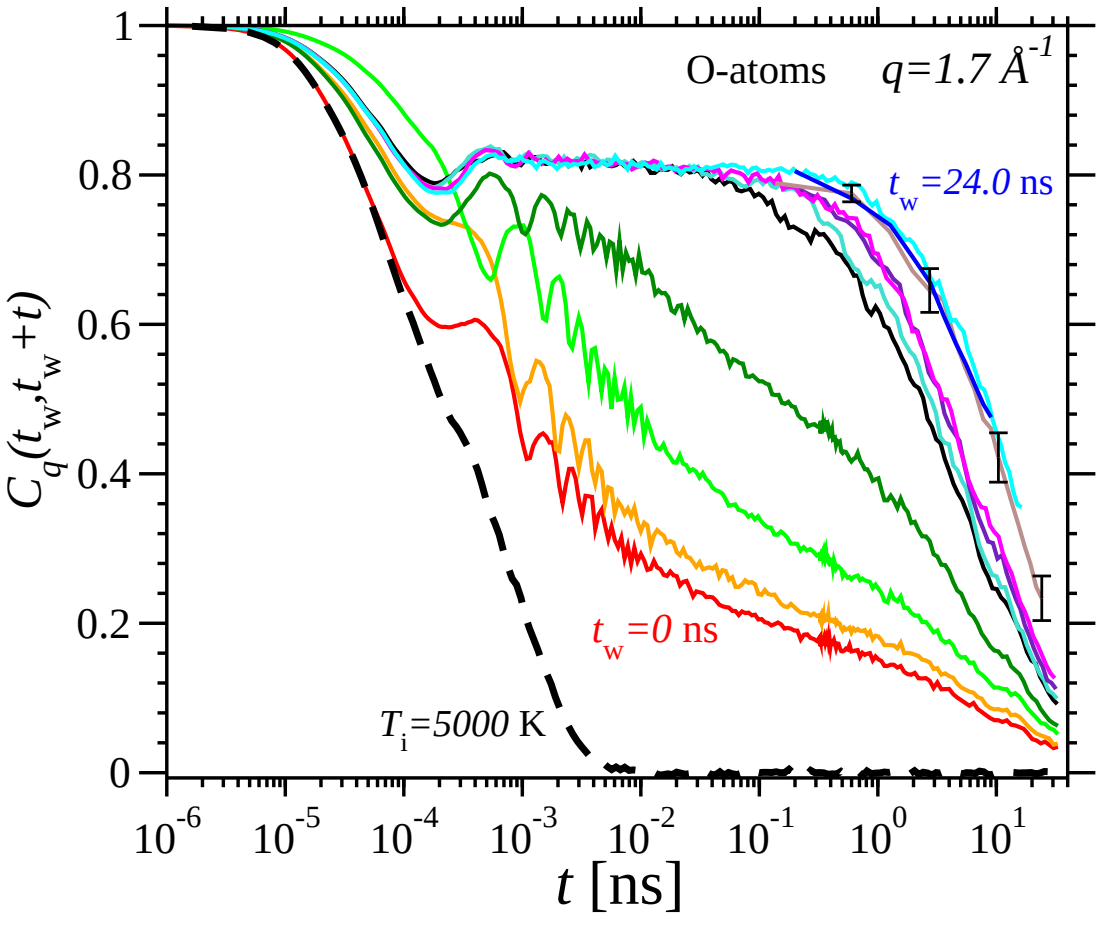
$$S_{\alpha\beta}(q, t_\mathrm{w}) = \frac{1}{N} \sum_{i=1}^{N_\alpha} \sum_{j=1}^{N_\beta} \mathrm{e}^{i\vec{q}\cdot(\vec{r}_i(t_\mathrm{w}) - \vec{r}_j(t_\mathrm{w}))}$$



- t_w dependence weak
- in following:
  - $C_q(t_\mathrm{w}, t_\mathrm{w} + t)$
    (mostly q of FSDP)
  - $\Delta r^2(t_\mathrm{w}, t_\mathrm{w} + t)$

Generalized Intermediate Incoherent Scattering Function

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

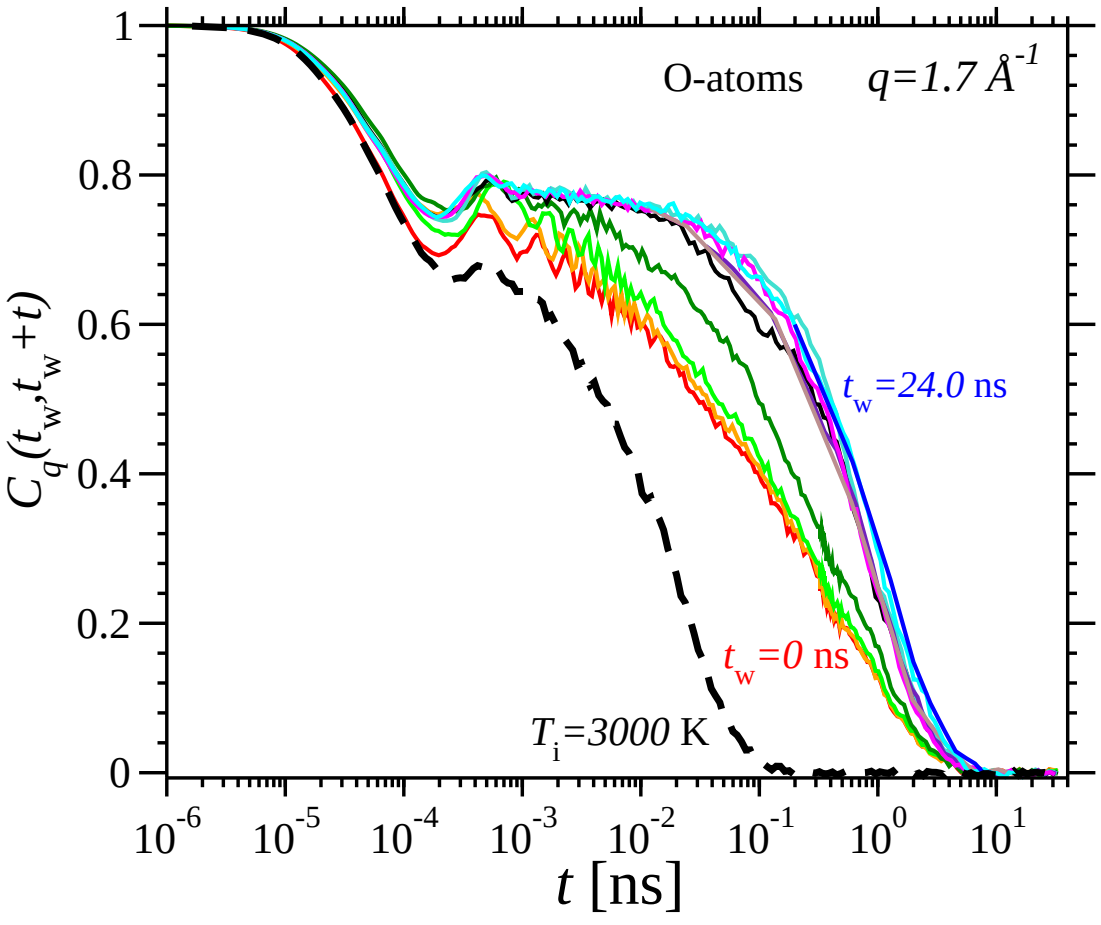


$T_\mathrm{i} = 5000$ K $T_\mathrm{f} = 2500$ K

- t_w **small:**
  - $t_\mathrm{w} = 0 \,\&\, t \lesssim 5 \cdot 10^{-5}$ ns: T_i good approx.
  - no plateau
  - decay t_w-dependent
- t_w **intermediate:**
  - plateau t_w-indep.
  - decay t_w-dependent
- t_w **large:** t_w-indep.
  $\longrightarrow$ equilibrium

Generalized Intermediate Incoherent Scattering Function

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

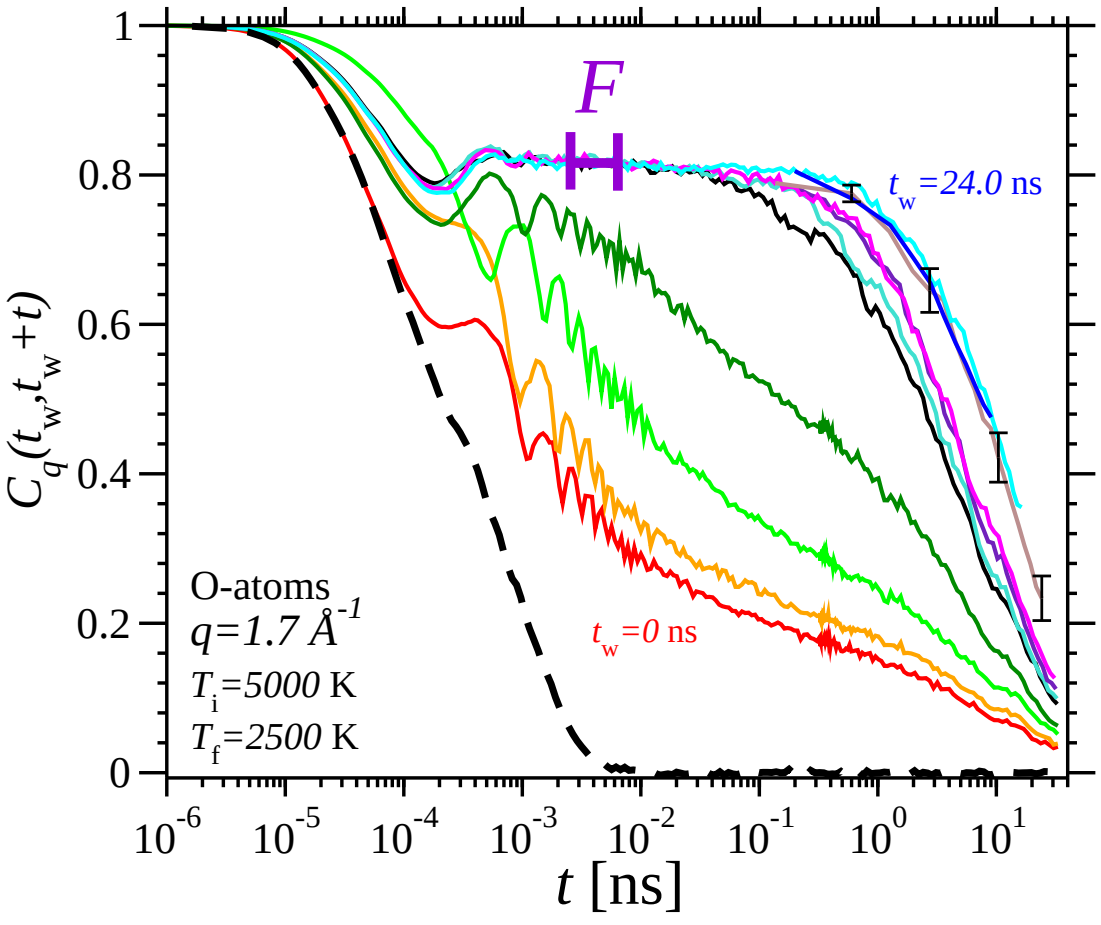


$T_\mathrm{i} = 3760$ K $T_\mathrm{f} = 3000$ K

- t_w **small:**
  - $t_\mathrm{w} = 0 \,\&\, t \lesssim 5 \cdot 10^{-5}$ ns: T_i good approx.
  - no plateau
  - decay t_w-dependent
- t_w **intermediate:**
  - plateau t_w-indep.
  - decay t_w-dependent
- t_w **large:** t_w-indep.
  $\longrightarrow$ equilibrium

Generalized Intermediate Incoherent Scattering Function

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



- t_w **small:**
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  - decay t_w-dependent
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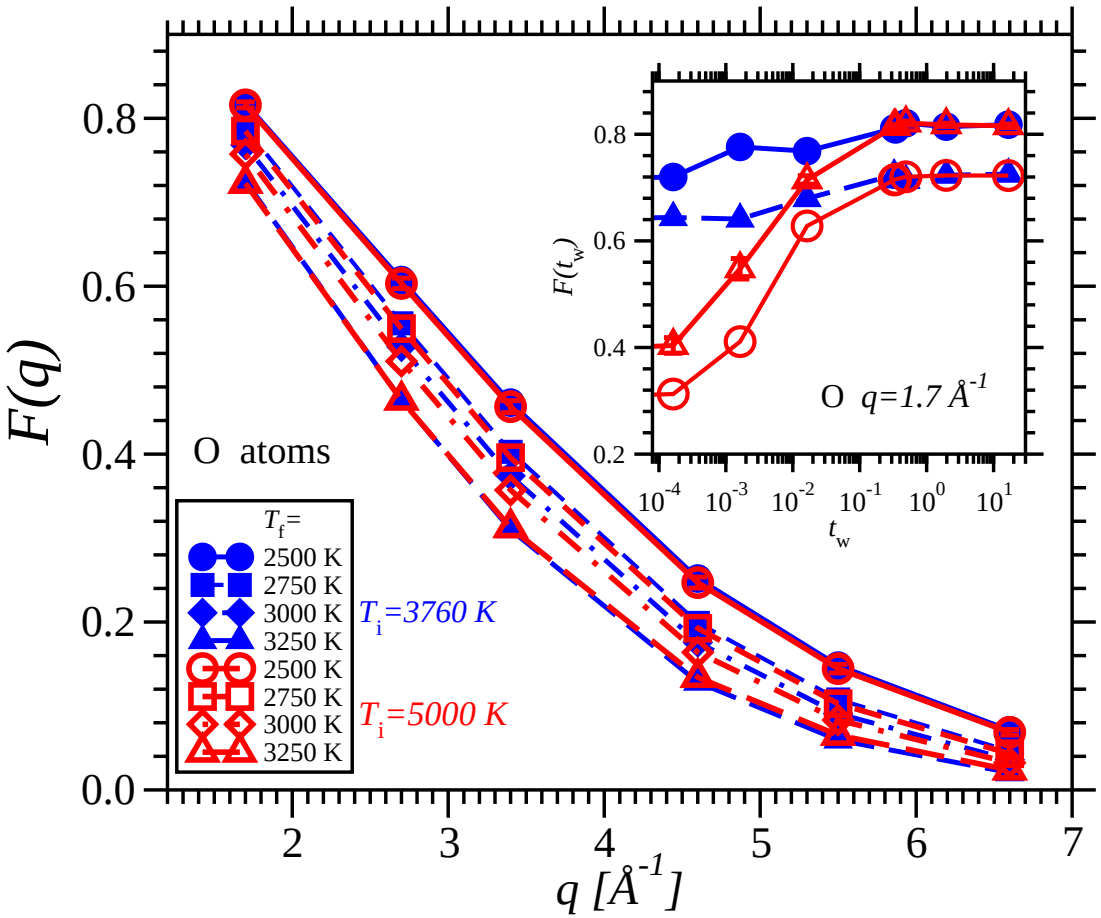
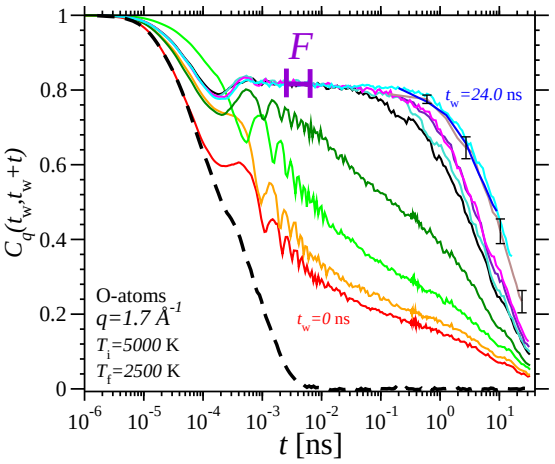
  - decay t_w-dependent
- t_w **large:** t_w-indep. $\longrightarrow$ equilibrium

Plateau Height



Definition:

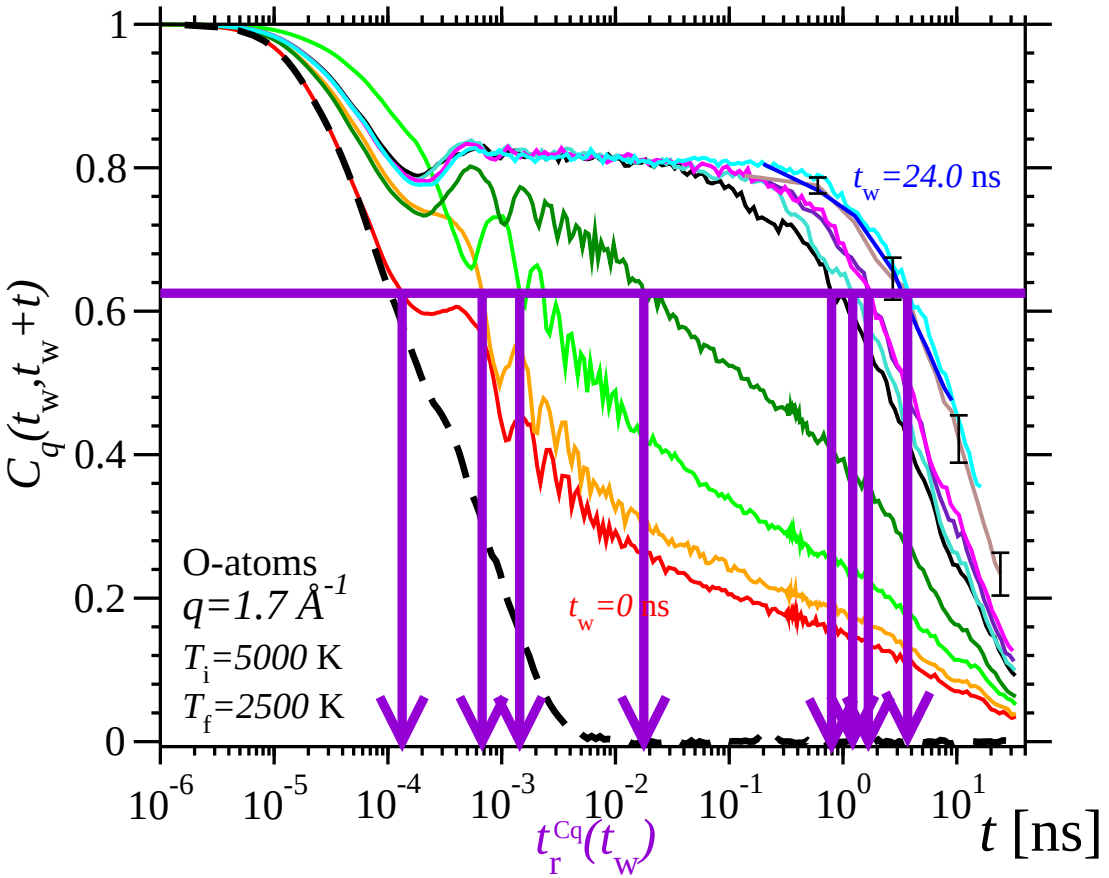


intermediate and large t_{w}:

- $F(t_{\mathrm{w}})$ indep. of t_{w}
- $F(q)$ independent of T_{i}

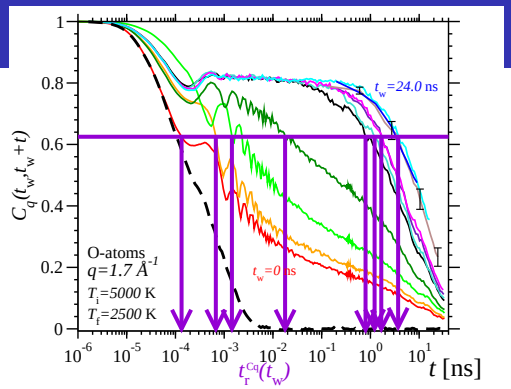
Generalized Intermediate Incoherent Scattering Function

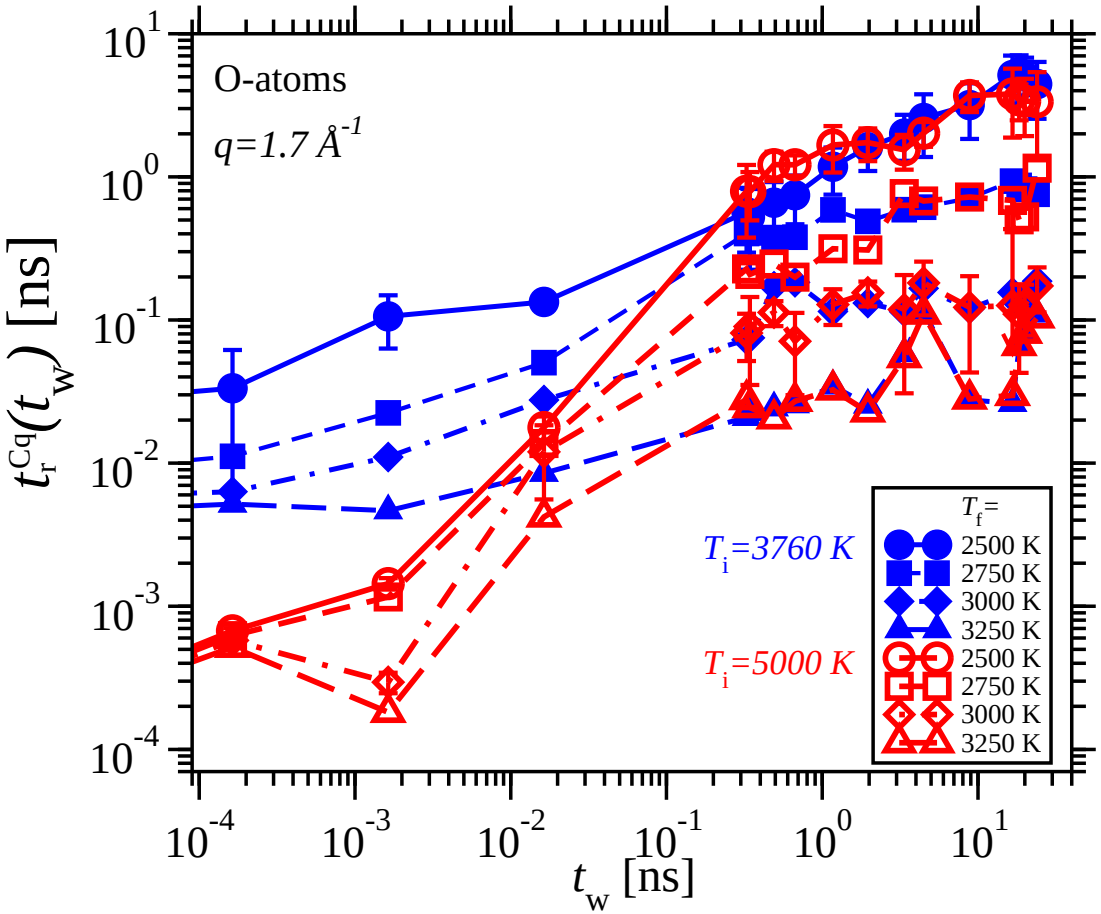
$$C_q(t_\mathrm{w}, t_\mathrm{w}+t) = \frac{1}{N_\alpha} \sum_{j=1}^{N_\alpha} \mathrm{e}^{i\vec{q}\cdot(\vec{r}_j(t_\mathrm{w}+t)-\vec{r}_j(t_\mathrm{w}))}$$



- t_w **small:**
  - $t_\mathrm{w} = 0 \,\&\, t \lesssim 5 \cdot 10^{-5}$ ns: T_i good approx.
  - no plateau
  - decay t_w-dependent
- t_w **intermediate:**
  - plateau t_w-indep.
  - decay t_w-dependent
- t_w **large:** t_w-indep.
  $\longrightarrow$ equilibrium

Decay Time





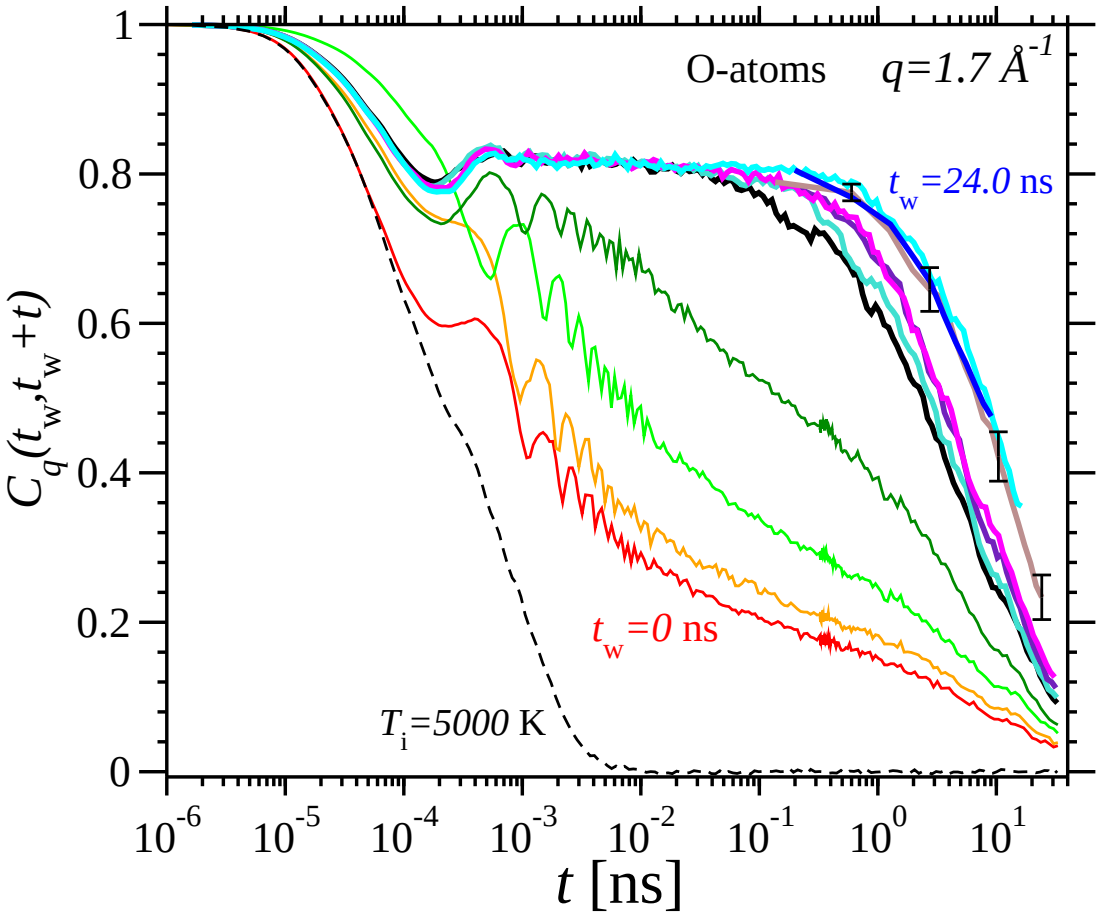
Three t_{w} Ranges:

- t_{w} **small:**
  - $t_{\mathrm{r}}^{\mathrm{Cq}}$ incr. with incr. t_{w}
  - slope T_{i} & T_{f} dep.
- t_{w} **intermediate:**
  $t_{\mathrm{r}}^{\mathrm{Cq}}$ incr. with incr. t_{w}
- t_{w} **large:**
  $t_{\mathrm{r}}^{\mathrm{Cq}}$ indep. of t_{w} & T_{i}
  $\Rightarrow$ equilibrium reached

t_{w} Ranges dependent on T_{i}

Generalized Intermediate Incoherent Scattering Function

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

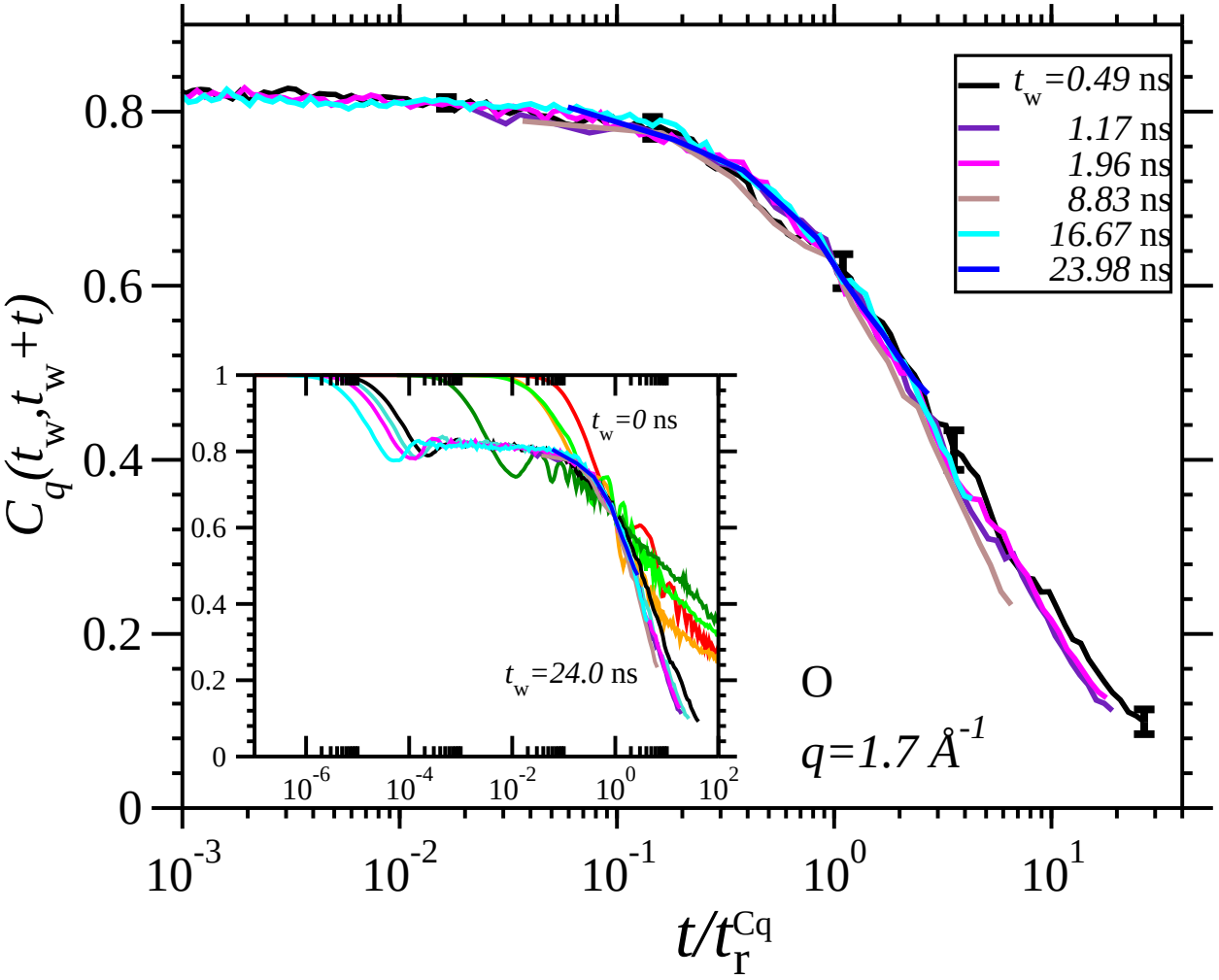


- t_w **small:**
  - $t_\mathrm{w} = 0 \,\&\, t \lesssim 5 \cdot 10^{-5}$ ns: T_i good approx.
  - no plateau
  - decay t_w-dependent
- t_w **intermediate:**
  - plateau t_w-indep.
  - decay t_w-dependent
  - time superposition ?
- t_w **large:** t_w-indep.
  $\longrightarrow$ equilibrium

Generalized Intermediate Incoherent Scattering Function

MF: $C_q(t_\mathrm{w}, t_\mathrm{w}+t) = C_q^\mathrm{ST}(t) + C_q^\mathrm{AG}\left(\frac{h(t_\mathrm{w}+t)}{h(t_\mathrm{w})}\right)$

Superposition: $C_q(t_\mathrm{w}, t_\mathrm{w}+t) = C_q^\mathrm{ST}(t) + C_q^\mathrm{AG}\left(\frac{t}{t_\mathrm{r}^\mathrm{Cq}(t_\mathrm{w})}\right)$



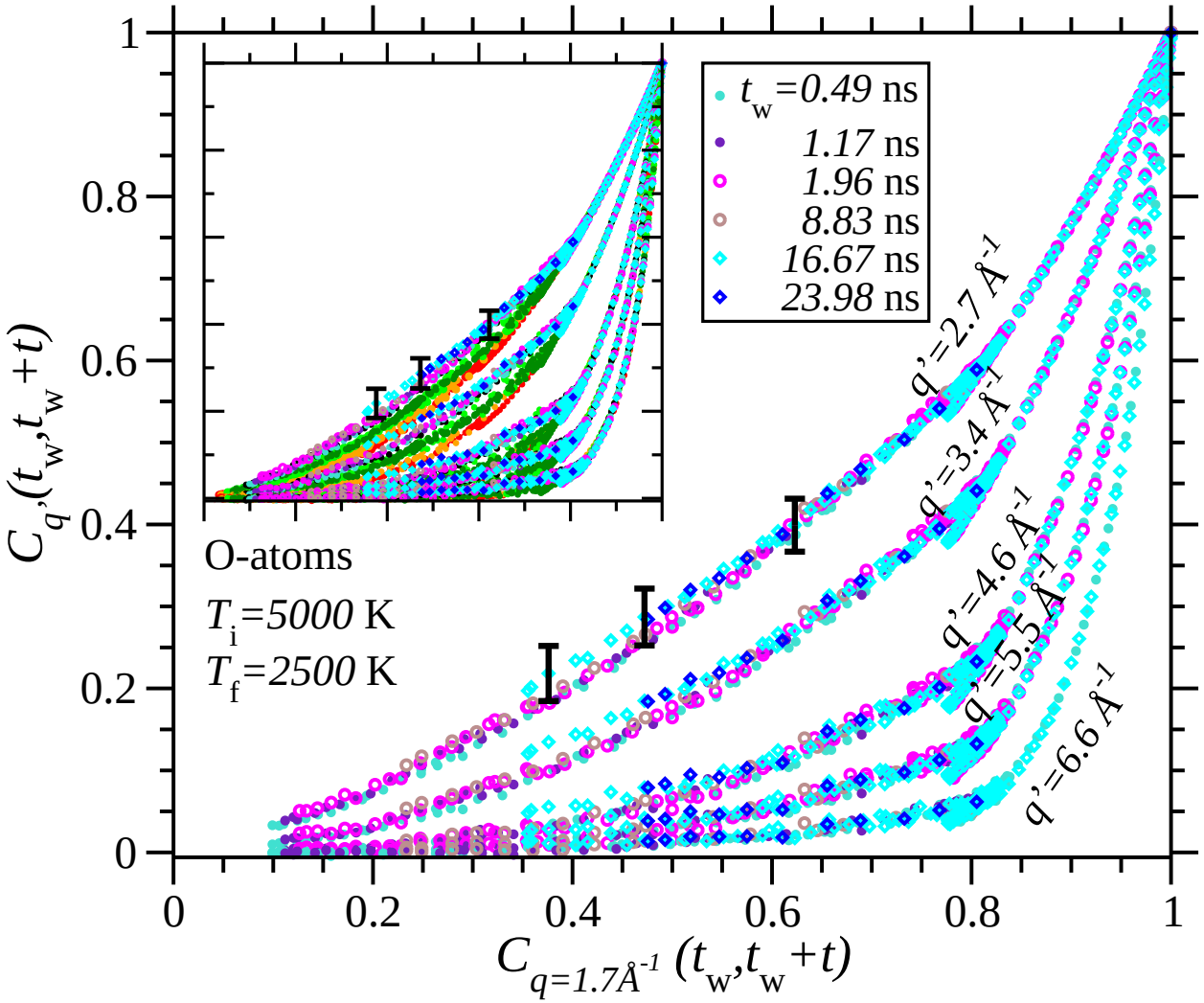
- t_w **small:** no time superposition
- t_w **intermediate:** time superposition
- t_w **large:** superposition includes equilibrium curve

LJ: [Kob & Barrat, PRL 78, 24 (1997)]

Generalized Intermediate Incoherent Scattering Function

$$C_q(t_\mathrm{w}, t_\mathrm{w}+t) = C_q^\mathrm{ST}(t) + C_q^\mathrm{AG}\left(\frac{h(t_\mathrm{w}+t)}{h(t_\mathrm{w})}\right)$$

Is h dependent on C_q?

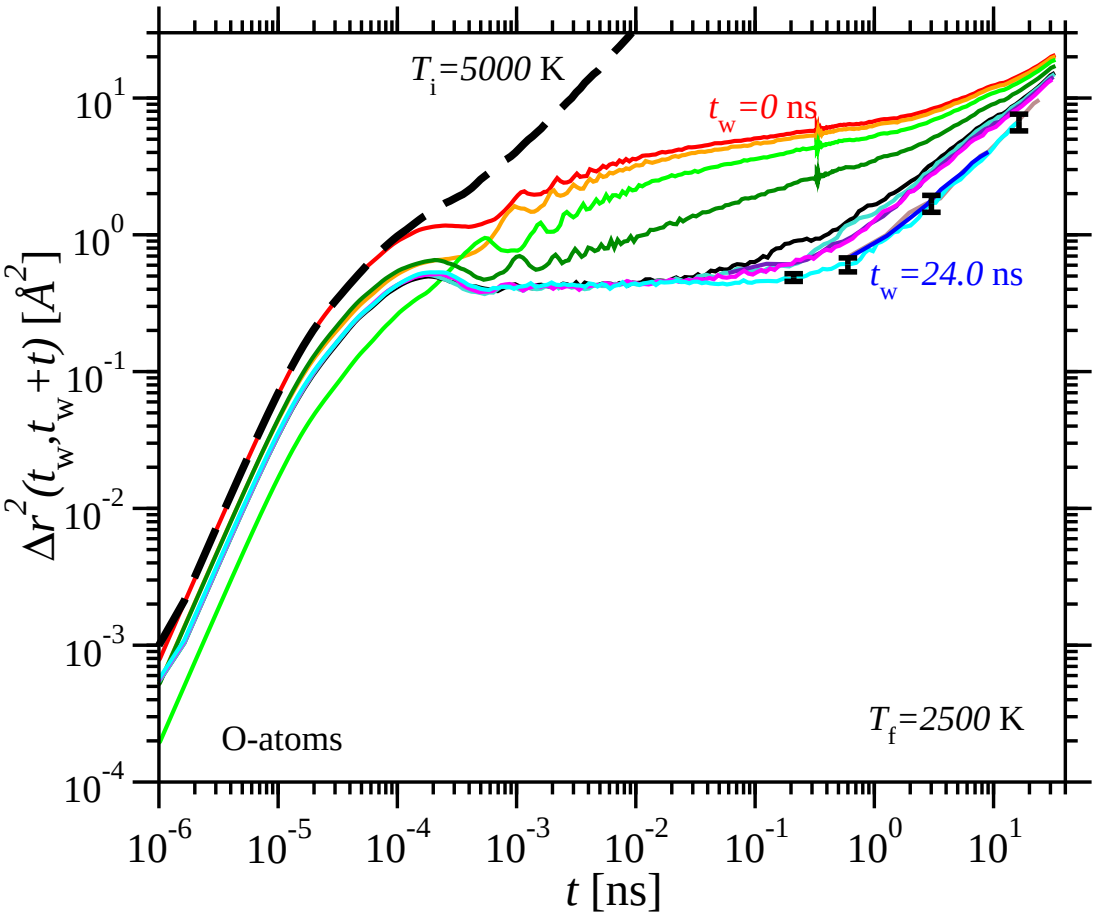


- t_w **small:** no superposition
- t_w **intermediate:** superposition of $C_{q'}(C_q)$ $\Rightarrow$ h indep.of C_q
- t_w **large:** superposition includes equilibrium curve

LJ: [Kob & Barrat, EPJ B 13, 319 (2000)]

Mean Square Displacement

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

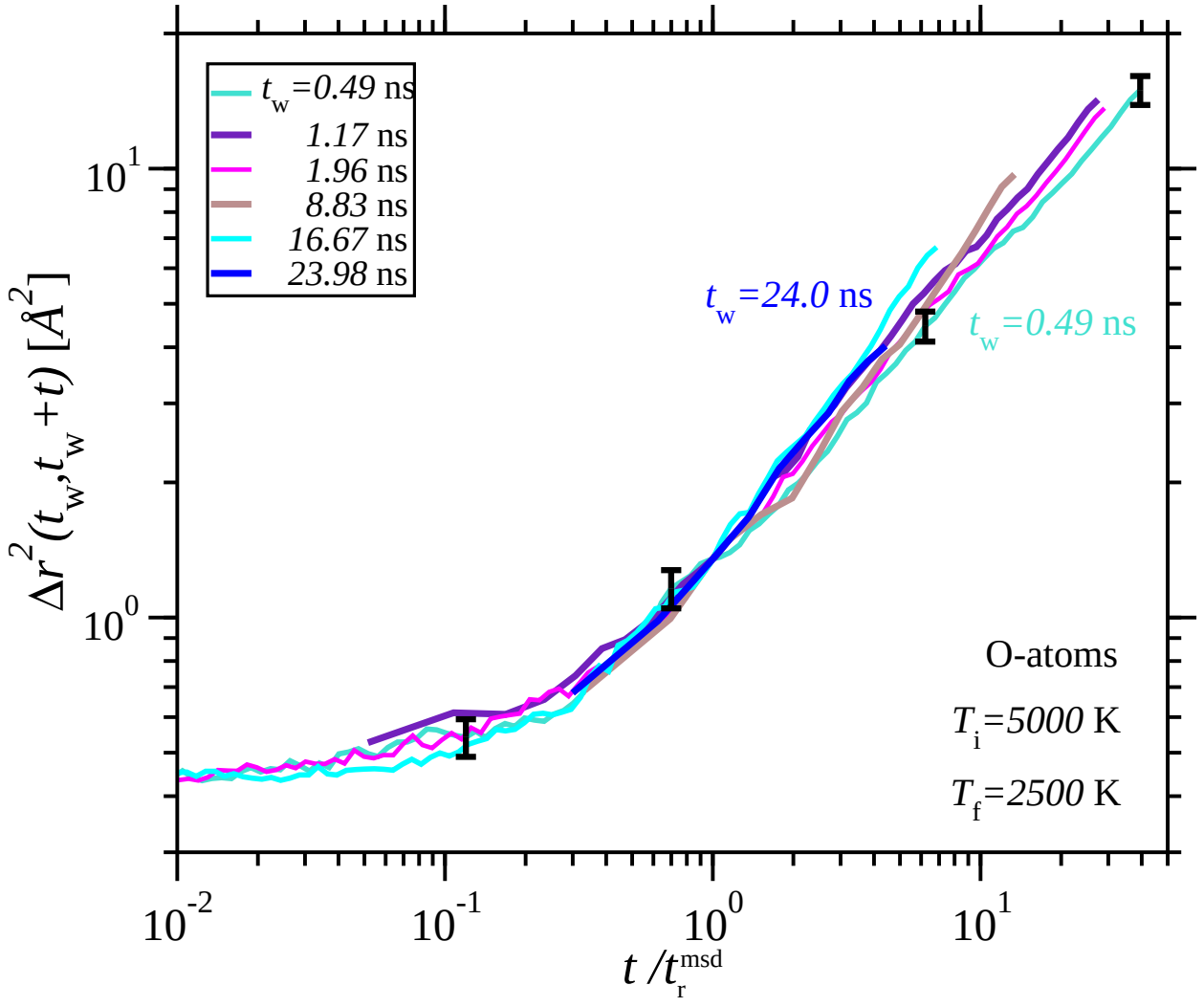


Three t_w Ranges:

- t_w **small:**
  - $t_\mathrm{w} = 0 \,\&\, t \lesssim 5 \cdot 10^{-5}$ ns: T_i good approx.
  - no plateau
  - increase t_w-dependent
- t_w **intermediate:**
  - plateau t_w-indep.
  - increase t_w-dependent
- t_w **large:** t_w-indep. $\longrightarrow$ equilibrium

Mean Square Displacement

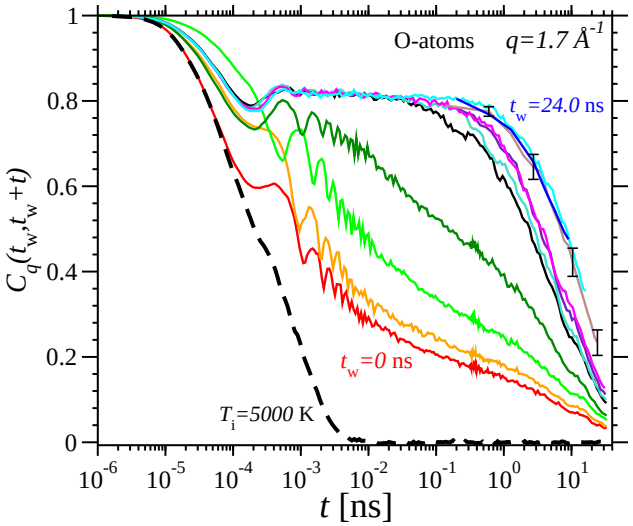
$$\Delta r^2(t_{\mathrm{w}}, t_{\mathrm{w}}+t) = \left(\Delta r^2\right)^{\mathrm{ST}}(t) + \left(\Delta r^2\right)^{\mathrm{AG}}\left(\frac{t}{t_{\mathrm{r}}^{\mathrm{msd}}(t_{\mathrm{w}})}\right)$$



- t_w **small:** no time superposition
- t_w **intermediate:** **no** time superposition
- t_w **large:** **no** time superposition

Summary

$C_q(t_\mathrm{w}, t_\mathrm{w}+t)$ and $\Delta r^2(t_\mathrm{w}, t_\mathrm{w}+t)$:



Three t_w Ranges:

- t_w **small:**
  - $t_\mathrm{w} = 0$ and t small: T_i good approx.
  - dependent on t_w, T_i, T_f
- t_w **intermediate:**
  - plateau indep. of t_w and T_i
  - C_q time superposition (not Δr^2)
  - $C_q^{\mathrm{AG}}\left(\frac{h(t_\mathrm{w}+t)}{h(t_\mathrm{w})}\right)$: h is C_q indep.
- t_w **large:**
  - indep. of t_w and $T_i \longrightarrow$ equilibrium
  - for C_q equilibrium included in superposition

Past & Future:

Binary Lennard Jones:

- jumps [KVL, JCP 121, 4781 (2004)]
- self-organized criticality (correlated jumps)
  [KVL,E.A. Baker, EPL 76, 1130 (2006)]

SiO_2:

- aging to equilibrium [to be submitted to PRE]
- local C_q [A. Parsaeian, H.E. Castillo, KVL, to be published]
- jumps (R. Bjorkquist, L. Chambers)

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A. Zippelius & Institute of Theoretical Physics, University Göttingen, Germany