Temperature-Dependent Defect Dynamics in SiO_2

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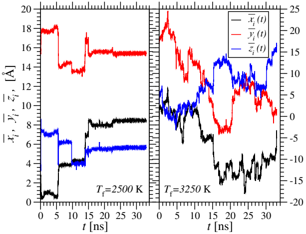
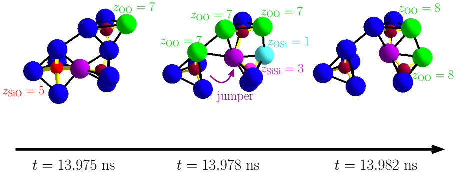
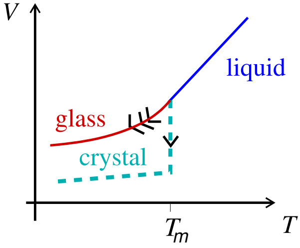
Bucknell University & Göttingen

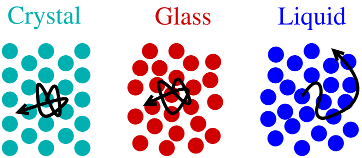


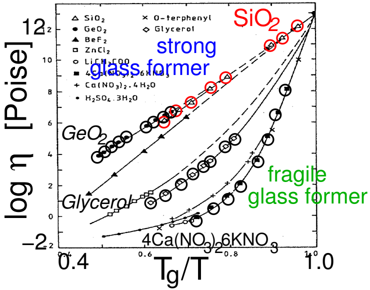


Göttingen, June 12, 2013

Introduction: Glass







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

Dynamics:

Viscocity η as function of inverse temperature T

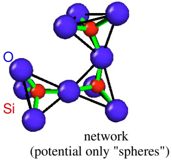
- slowing down of many decades $\longrightarrow$ very interesting dynamics
- strong and fragile glass formers
  Here: SiO_2 (strong glass former)

Model & Simulations

Model: BKS Potential

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

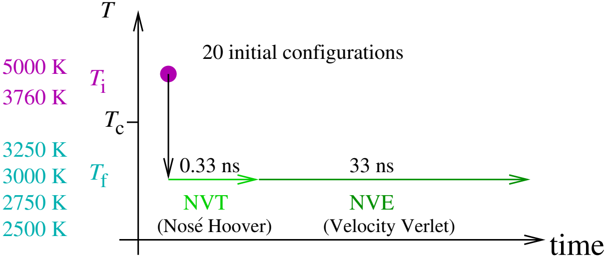
$$\phi(r_{ij}) = \frac{q_i q_j e^2}{r_{ij}} + A_{ij} e^{-B_{ij} r_{ij}} - \frac{C_{ij}}{r_{ij}^6}$$



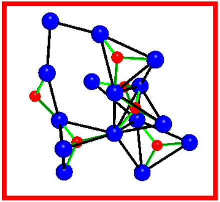
112 Si & 224 O $\rho = 2.32$ g/cm^3

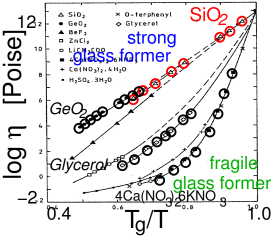
$T_c = 3330$ K

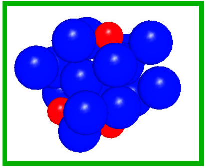
Molecular Dynamics Simulations:



Motivation

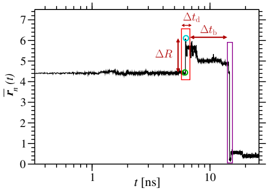






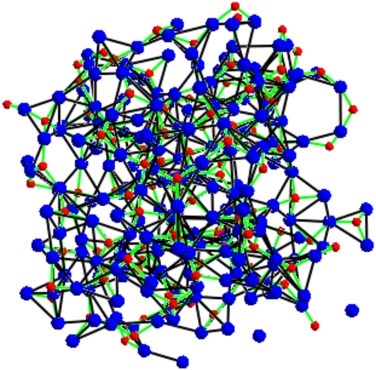
Unexpected Similar Dynamics:

- scaling plots (C_q, χ_4, $P(C_q)$)
- jump statistics ($P(\Delta R)$, $P(\Delta t_\mathrm{b})$)



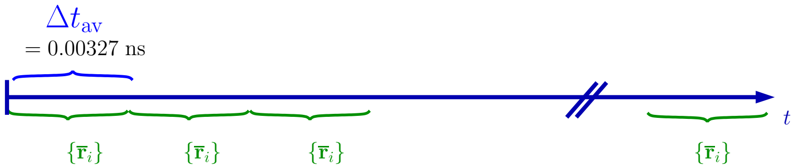
$\longrightarrow$ SiO_2-specific perspective?

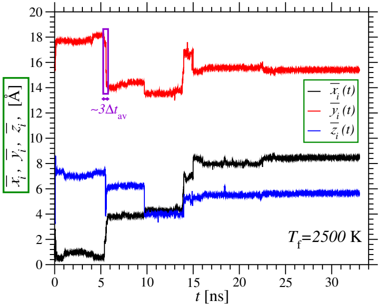
Network Glass SiO_2

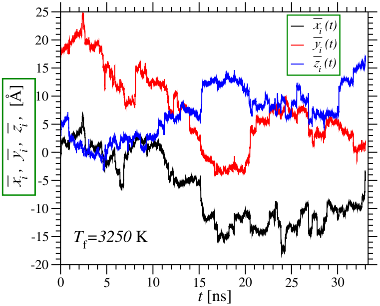


$\longrightarrow$ extract main features

Time Averaged Trajectories



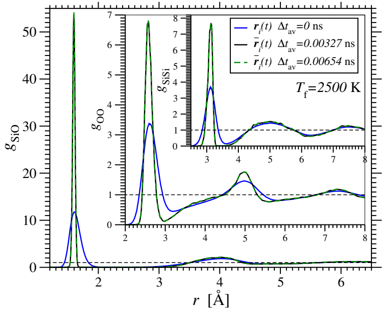


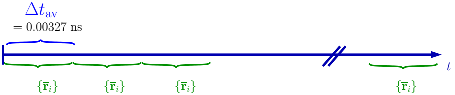


$\longrightarrow$ strong temperature dependence

Structure: Radial Distribution Function

$$g_{\alpha\beta}(r) = \left\langle \frac{V}{N_\alpha N_\beta} \sum_{i=1}^{N_\alpha} \sum_{\substack{j=1 \\ j\neq i}}^{N_\beta} \delta(|\mathbf{r}| - |\bar{\mathbf{r}}_{ij}(t)|) \right\rangle \qquad \alpha, \beta \in \{\text{Si,O}\}$$

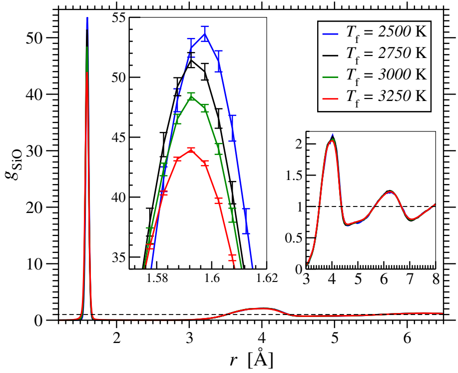




- time average sharpens peaks

Structure: Radial Distribution Function

$$g_{\alpha\beta}(r) = \left\langle \frac{V}{N_\alpha N_\beta} \sum_{i=1}^{N_\alpha} \sum_{\substack{j=1 \\ j\neq i}}^{N_\beta} \delta(|\mathbf{r}| - |\overline{\mathbf{r}}_{ij}(t)|) \right\rangle \qquad \alpha, \beta \in \{\text{Si,O}\}$$

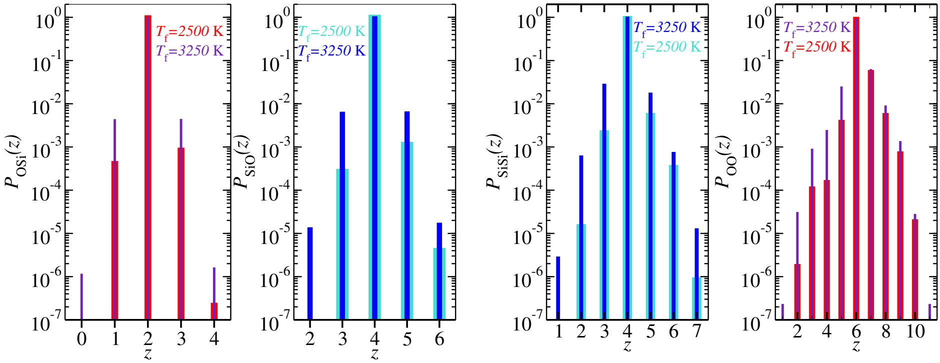


- almost no temperature dependence

Structure: Coordination Number

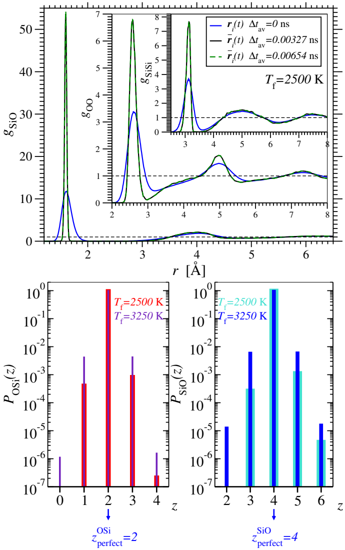
$z_i^{\alpha\beta}$ = number of nearest neighbors

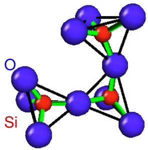
via minimum of $g_{\alpha\beta}$ ($\alpha, \beta \in$ {Si,O})



$\longrightarrow$ sharply peaked

SiO_2-Network



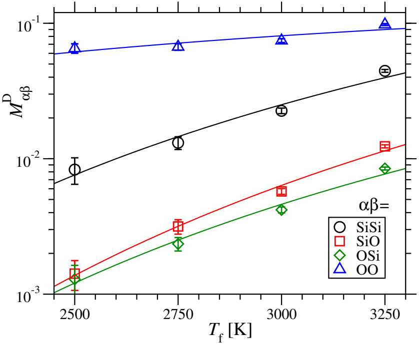


- almost perfect neighborhood
- well defined defects: $z_i^{\alpha\beta} \neq z_{\text{perfect}}^{\alpha\beta}$
- focus on defects

Number of Defects

$$\chi_i^{\mathrm{D}}(t,\beta) = \begin{cases} 1 & \text{if at time } t \quad z_i^{\alpha\beta}(t) \neq z_{\text{perfect}}^{\alpha\beta} \\ 0 & \text{if at time } t \quad z_i^{\alpha\beta}(t) = z_{\text{perfect}}^{\alpha\beta} \end{cases}$$

$$M_{\alpha\beta}^{\mathrm{D}} = \left\langle \frac{1}{N_\alpha} \sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t,\beta) \right\rangle \quad .$$



- few defects (mostly OO)
- strong temperature dependence: $M_{\alpha\beta}^{\mathrm{D}}$ increases with temperature
- Arrhenius fits

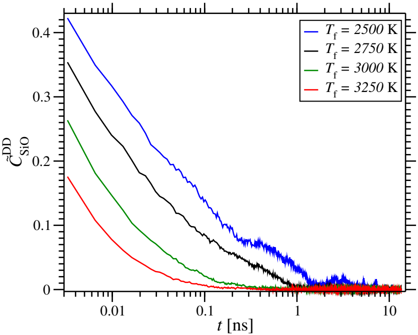
Life Time of Defects?

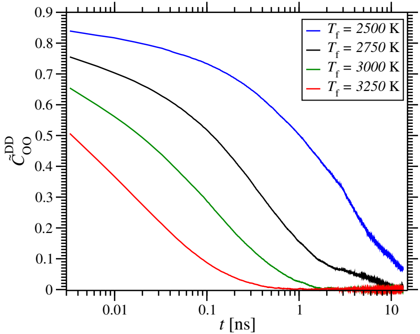
Defect-Defect Correlation

$$\chi_i^{\mathrm{D}}(t,\beta) = \begin{cases} 1 & \text{if at time } t \quad z_i^{\alpha\beta}(t) \neq z_{\mathtt{perfect}}^{\alpha\beta} \\ 0 & \text{if at time } t \quad z_i^{\alpha\beta}(t) = z_{\mathtt{perfect}}^{\alpha\beta} \end{cases}$$

$$C^{\mathrm{DD}}(t,\alpha,\beta) = \left\langle \frac{1}{N_\alpha} \sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t_0,\beta)\chi_i^{\mathrm{D}}(t_0+t,\beta) \right\rangle - \left\langle \frac{1}{N_\alpha} \sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t_0,\beta) \right\rangle \left\langle \frac{1}{N_\alpha} \sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t_0+t,\beta) \right\rangle$$

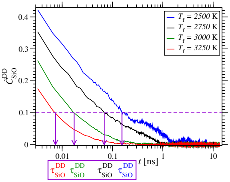
$$\tilde{C}_{\alpha,\beta}^{\mathrm{DD}}(t) = \frac{C^{\mathrm{DD}}(t,\alpha,\beta)}{C^{\mathrm{DD}}(t=0,\alpha,\beta)}$$

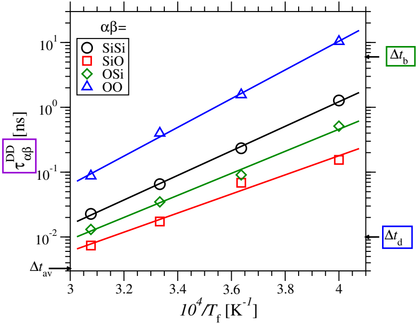


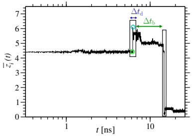


$\longrightarrow$ strong temperature dependence

Life Time of Defects



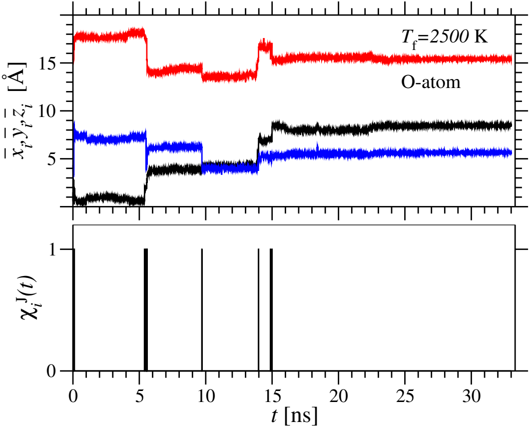




- SiO- & OSi-defects short-lived (flashes)
- OO-defects longer lived

Jumps

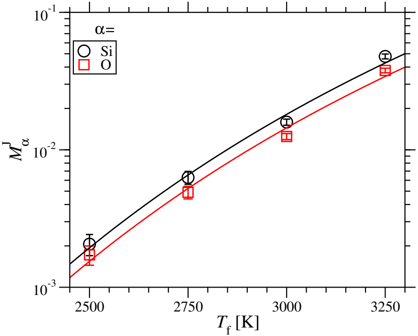
$$\chi_i^{\mathrm{J}}(t) = \begin{cases} 1 & \text{during jump} \\ 0 & \text{otherwise} \end{cases}$$



Number of Jumps

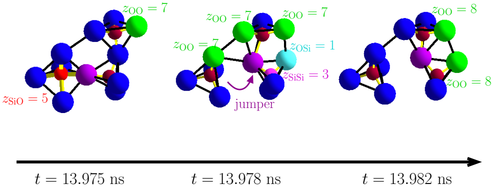
$$\chi_i^{\mathrm{J}}(t) = \begin{cases} 1 & \text{during jump} \\ 0 & \text{otherwise} \end{cases}$$

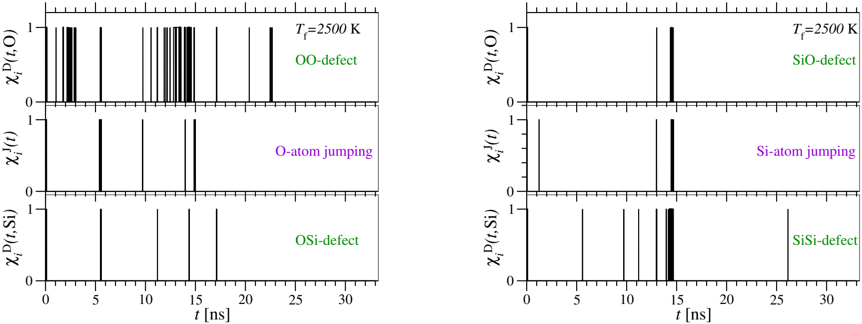
$$M_\alpha^{\mathrm{J}} = \left\langle \frac{1}{N_\alpha} \sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{J}}(t) \right\rangle$$



- very few jumps
- Arrhenius fits
- strong temperature dependence

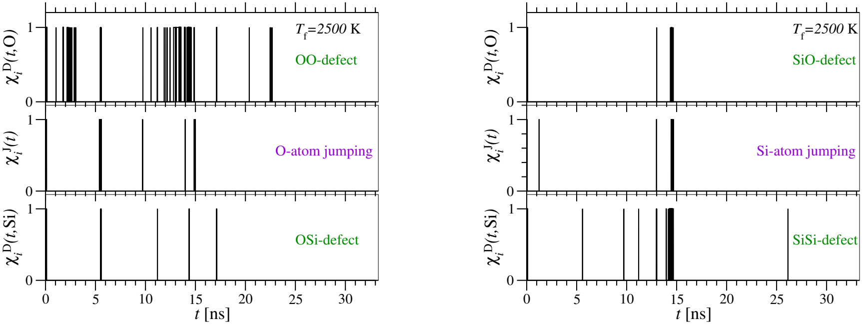
Defect-Jump Correlation





Are Defects and Jumps Correlated?

Defect-Jump Correlation

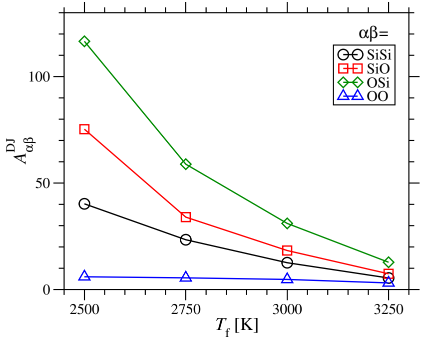


$$A^{\mathrm{DJ}}_{\alpha,\beta} = \frac{\left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t,\beta)\chi_i^{\mathrm{J}}(t)\right\rangle - \left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t,\beta)\right\rangle \left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{J}}(t)\right\rangle}{\left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t,\beta)\right\rangle \left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{J}}(t)\right\rangle}$$

Defect-Jump Correlation

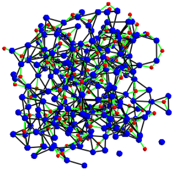
$$A^{\mathrm{DJ}}_{\alpha,\beta} = \frac{\left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t,\beta)\chi_i^{\mathrm{J}}(t) \right\rangle - \left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t,\beta) \right\rangle \left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{J}}(t) \right\rangle}{\left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t,\beta) \right\rangle \left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{J}}(t) \right\rangle}$$

$$= \frac{\left\langle \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha} \chi_i^{\mathrm{D}}(t,\beta)\chi_i^{\mathrm{J}}(t) \right\rangle - M^D_{\alpha\beta} M^J_\alpha}{M^D_{\alpha\beta} M^J_\alpha}$$



- strong correlation between defects and jumps
- correlation is decreasing with increasing temperature

Summary



Simulations of SiO_2-Glass

Extract Information:

- time averaged positions
- $\chi_i^{\mathrm{D}}(t,\beta)$, $\chi_i^{\mathrm{J}}(t)$

Defects:

- well defined $(g_{\alpha\beta}(r)$ & $P_{\alpha\beta}(z))$
- strong temperature dependence of $\tilde{C}^{\mathrm{DD}}_{\alpha,\beta}(t)$
- $\tau^{\mathrm{DD}}_{\alpha,\beta}$:
  - SiO- & OSi-defects short lived and OO-defects long lived
  - $\tau^{\mathrm{DD}}_{\alpha,\beta}$ decreasing with increasing temperature

Jump-Defect Correlation:

- strongly correlated
- $A^{\mathrm{DJ}}_{\alpha,\beta}$ decreases with increasing temperature

Acknowledgments: Supported by DFG via SFB 602 & FOR1394.