Study of atomic motion in rubidium borate glasses

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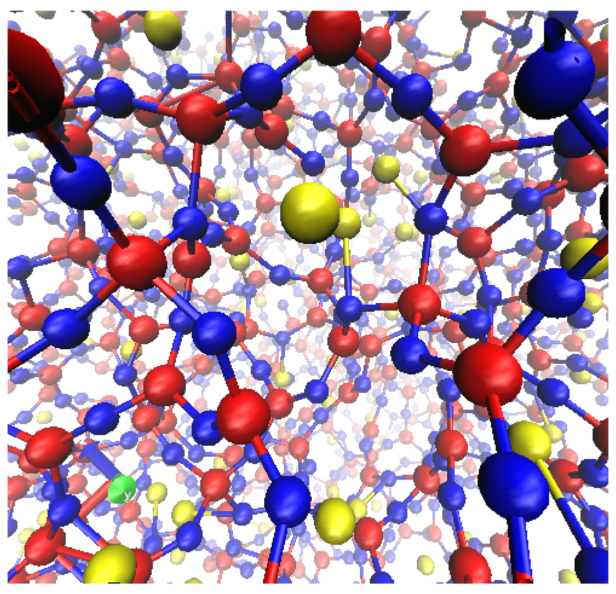
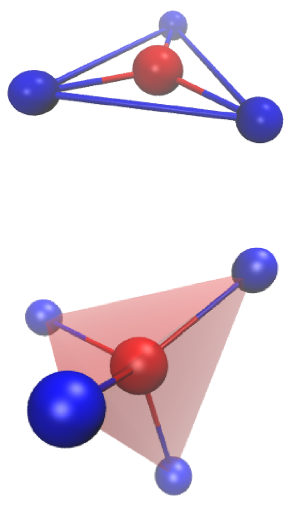
Introduction

Atomic Scale X-ray Photon Correlation Spectroscopy (**a**XPCS) uses coherent X-rays to probe the dynamics of materials on an atomic scale. It was applied to study atomic diffusion in crystals [1], but its application was recently extended to glasses as well [2]. Results of dynamics and structural studies on high- and low-alkali content rubidium borate glasses are presented here. Structural studies include total scattering experiments yielding the pair distribution function alongside Small-Angle X-ray Scattering (SAXS) studies giving information about inhomogeneities in the samples.

Theory

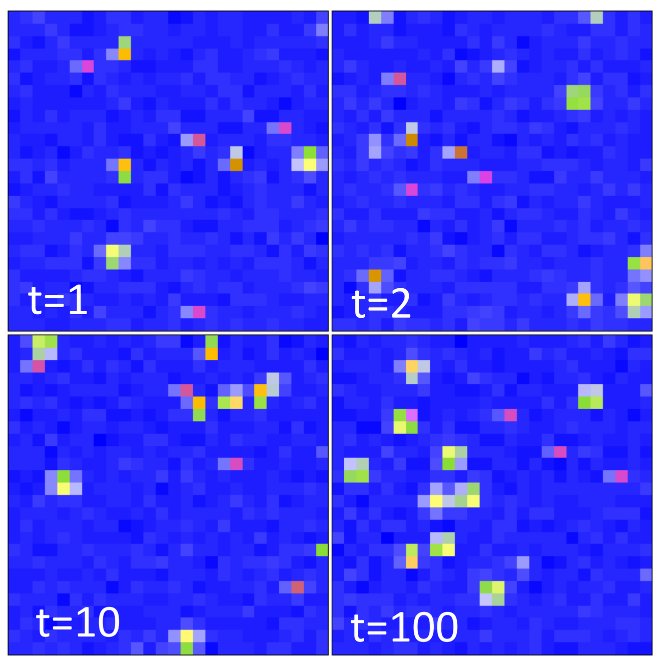
Real space structure: changes in time results in different scattering patterns





Space and time dependence described by the Van Hove Pair Correlation Function

Sequence of scattering patterns ⟶ Time correlation

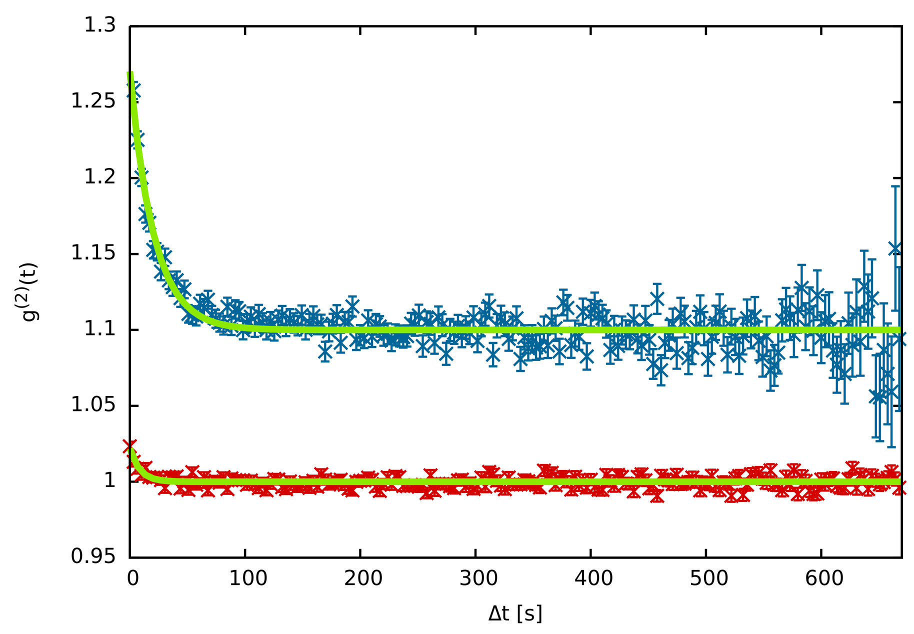


Intensity Auto Correlation Function (ACF)

$$g^{(2)}(\vec{q},\Delta t) := \frac{\langle I(\vec{q},t)I(\vec{q},t+\Delta t)\rangle}{\langle I(\vec{q},t)\rangle^2}$$

Functional form of the ACF:

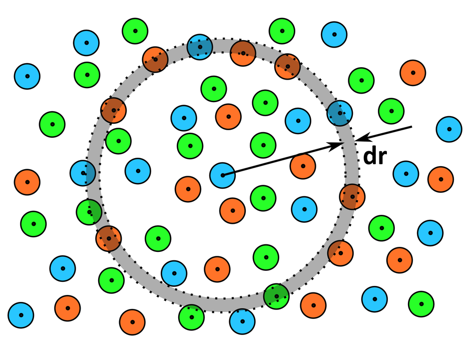
$$g^{(2)}(q) = 1+\beta e^{-\left(\frac{2\Delta t}{\tau}\right)^{\alpha}}$$



Normalization by the total structure factor

$$\tau_{coh}(\vec{q})^{-1} = \frac{\tau_{inc}(\vec{q})^{-1}}{S_{total}(q)}$$

Structure



Scattering function in general includes both Bragg and diffuse scattering → for amorphous substances only diffuse scattering occurs.

Pair distribution function (PDF) is the probability of finding two atoms separated by a distance r.

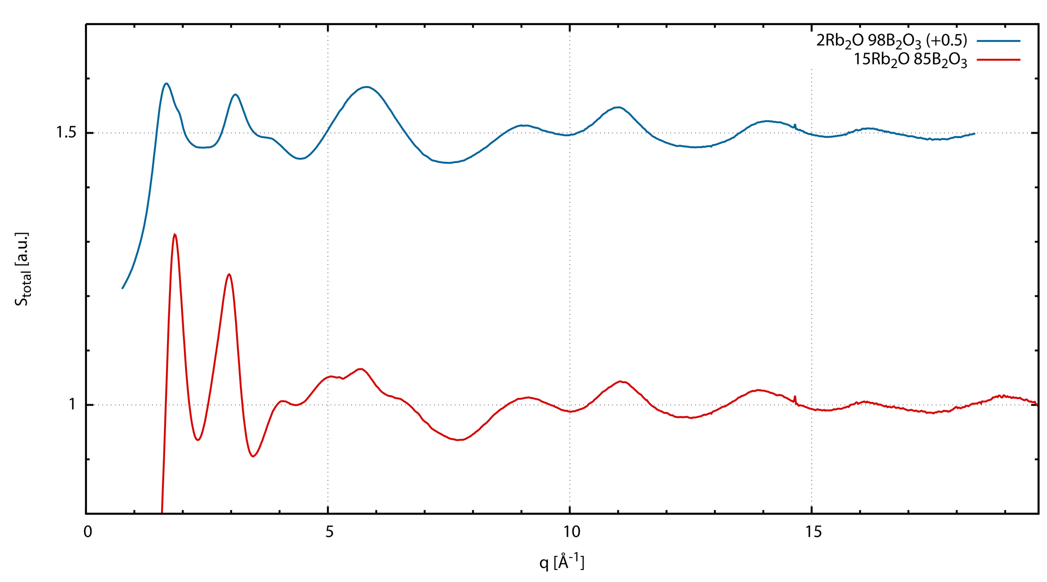
Connection of the PDF $G(r)$ to the measured scattered intensity $S(Q)$ via Fourier sine transform

$$G(r) = \frac{2}{\pi}\int_0^{\infty} \mathrm{d}Q\, Q\,(S(Q)-1)\,sin(Qr)$$

The scattered intensity is related to the total (static) structure factor by

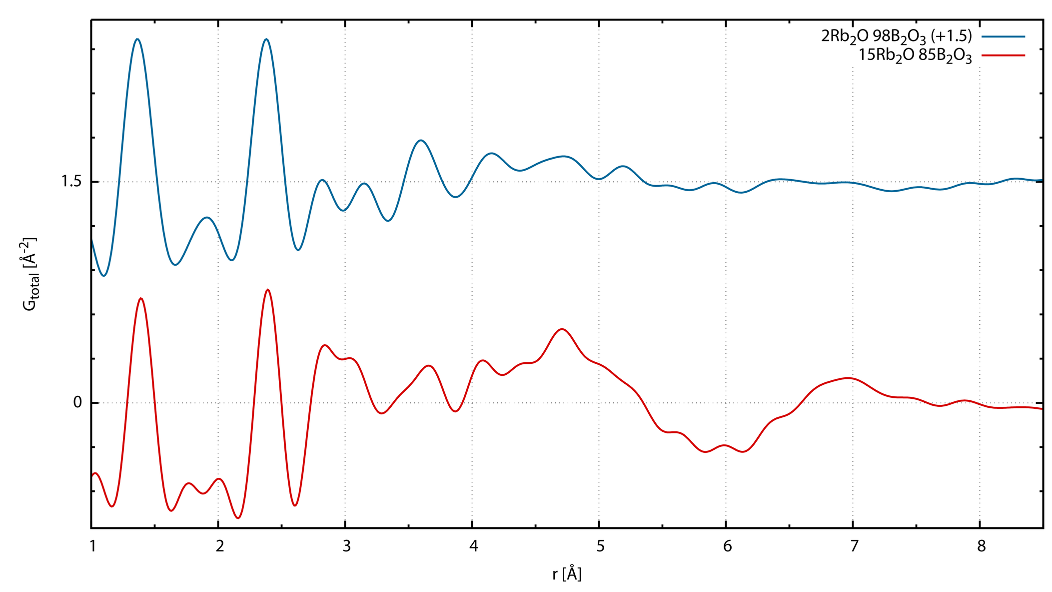
$$I(q) = N\left[\mathrm{E}\langle f^2\rangle - \mathrm{E}\langle f\rangle^2 + \mathrm{E}\langle f\rangle^2 S_{total}(q)\right]$$

Total structure factor and pair distribution function



Total structure factor as a function of momentum transfer

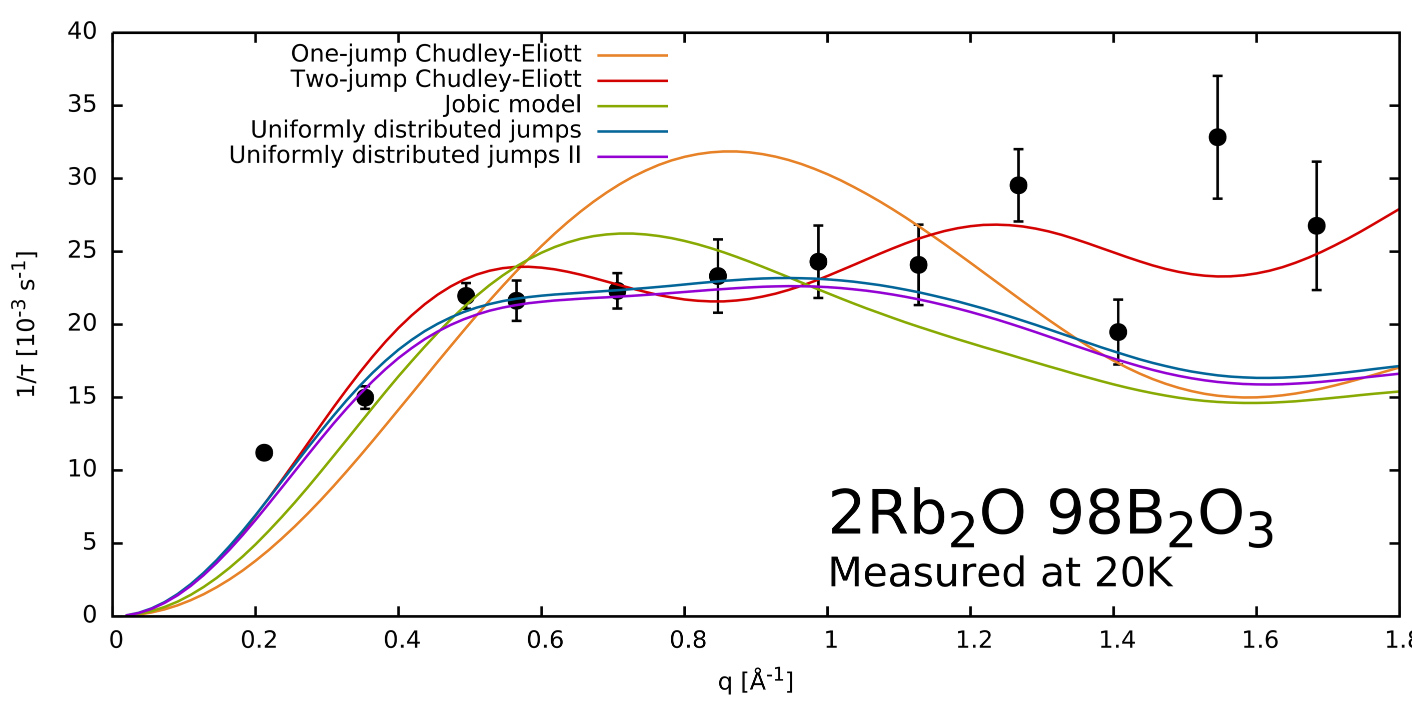
- Measured as platelets in transmission geometry → only weak contribution from surface → smooth curves
- Shows features up to high momentum transfers q → distinguished from most glasses with liquid-like structure factors



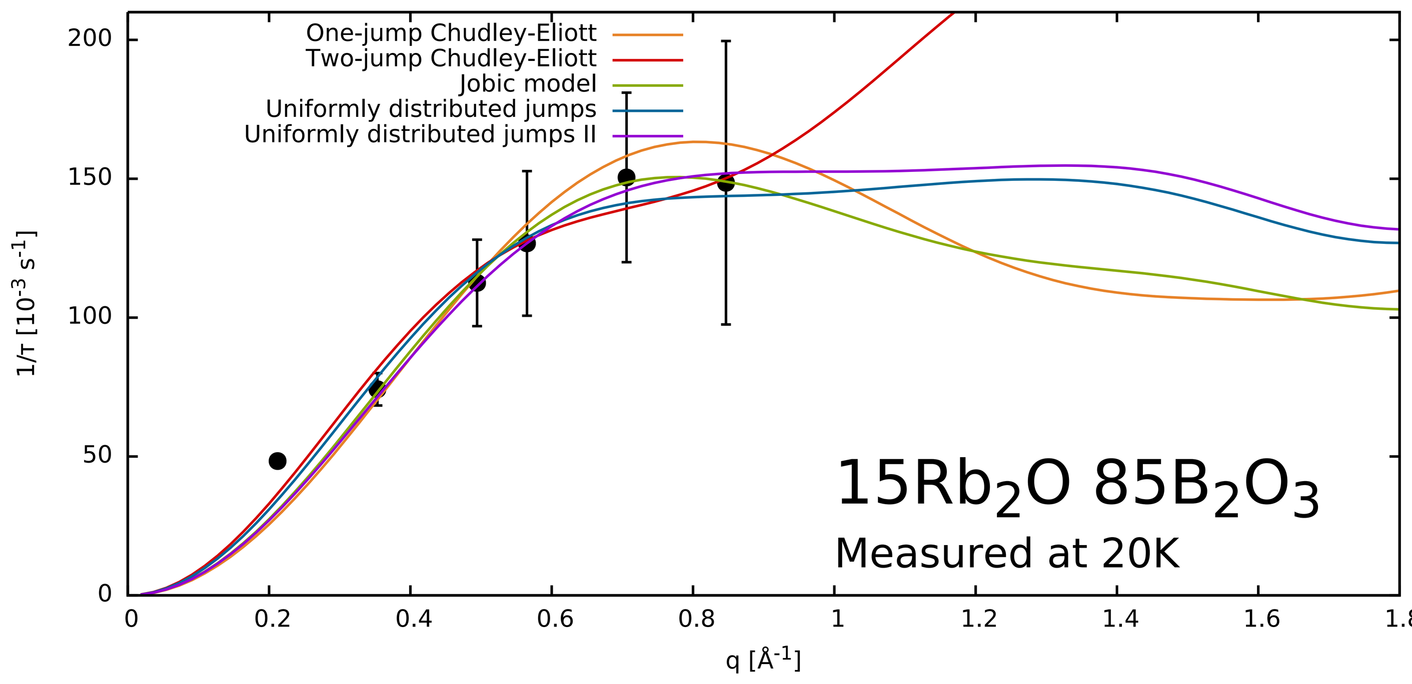
Pair distribution function as a function of radial distance

- Very sharp peaks at small distances 1.4 and 2.4 Å→ corresponding to rigid borate network → alkali ions are the diffusing species
- Well-defined short-range order resulting from strong covalent bonding

Results: fit to jump models

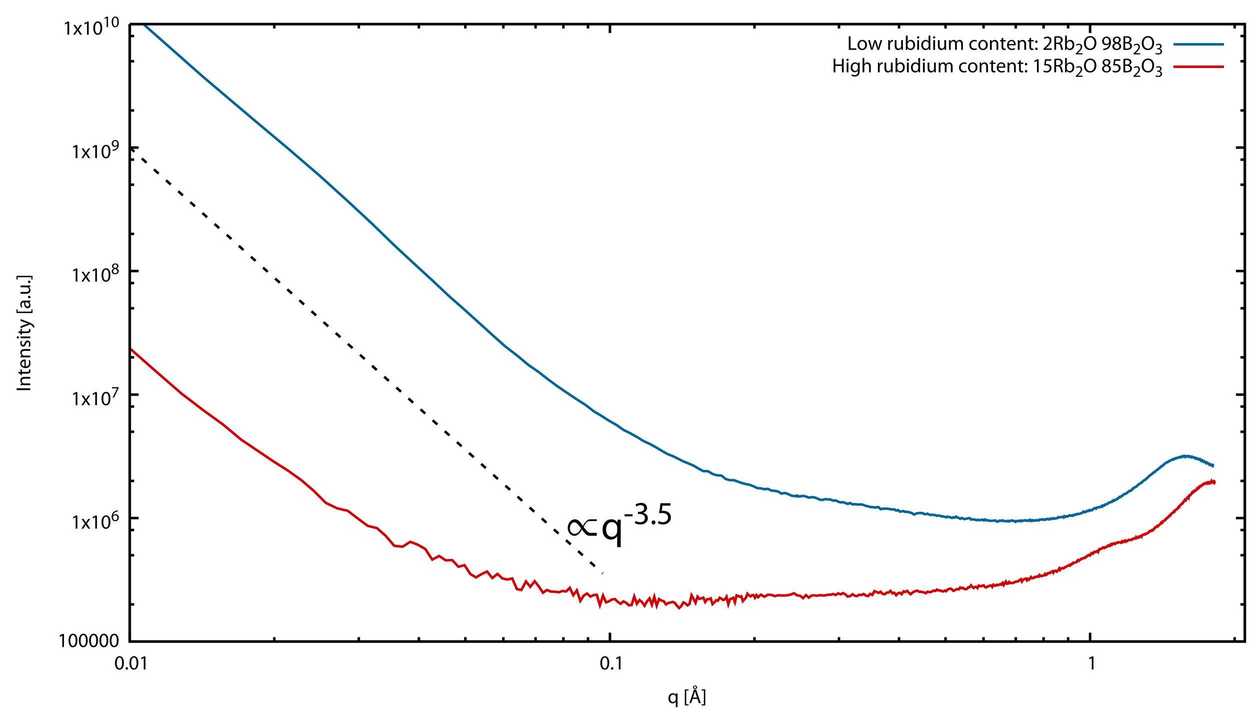


Best fit: Two-jump distances Chudley-Eliott Model with jump lengths $d_1 = 1.0$ Å and $d_2 = 8.5$ Å, with 3/4 of short-distance jumps



top: low alkali content glass, **bottom**: High alkali content glass. Several models can be fitted at small q-range. Fits are equally good in the hydrodynamic limit.

SAXS



SAXS intensities as a function of momentum transfer

- Inhomogeneities on a length scale > 60 nm are visible
- Only Porod region visible → Inhomogeneities too large for SAXS
- Slope of -3.5 indicating 3D inhomogeneities with smooth interfaces

Conclusion

- Diffusion in low rubidium content borate glasses best described by two-jump model
- Large difference in diffusivity between low and high alkali glass
- Diffusion at 20K still too fast to be measured in high alkali glass
- Inhomogenities are much larger in high alkali sample

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Der Wissenschaftsfonds.