

**Experiment title:**

Atomic dynamics in superionic conducting glasses studied with XPCS

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Report:

Fast ionic conductors are of interest especially due to their potential application as solid electrolytes in high energy density solid state batteries. Borate glasses have the ability to incorporate a wide range of different elements, thus they are an ideal candidate to study the atomic motion mechanisms of ions in glasses. Most of the experimental methods applied to measure diffusion in these materials are only sensitive to macroscopic scales corresponding to thousands of atomic jumps. In contrast to these approaches atomic X-ray Photon Correlation Spectroscopy (aXPCS), a new method for studying dynamics in solid state materials, can reveal information about single atomic motion in time and in space. It is based on the fact that a particular arrangement of atoms in a sample produces a characteristic intensity pattern when a coherent beam of X-rays is scattered. Collecting a series of intensity patterns at a scattering angle of up to 30° yields details of reciprocal space at certain times. The associated reciprocal space vector corresponds to a real space vector on the order of atomic distances. Correlating the scattering patterns at a multitude of angles, we can establish a model of the atomistic diffusion mechanisms in the material.

For amorphous materials, absence of translational symmetry allows for a wide range of possible configurations, thereby posing a challenge for developing a picture of the solid on the fundamental scale. The question of dynamics in amorphous media is far from being solved.

We have succeeded in demonstrating the feasibility of studies of atomic diffusion by aXPCS [1–4]. In a recent experiment, we proved that measuring dynamics on the atomic scale in oxide glasses is feasible at temperatures of 643–713 K. We found clear evidence for ion hopping on specific, well defined distances, directly observing the atomic network migration in the binary glass former PbO-SiO₂ [5]. In the beam time at hand, we measured for the first time alkali ion diffusion in a borate glass, studying 2(Rb₂O)98(B₂O₃) at 150 K (see Fig. 1).

The additional complexity of borate glasses requires more advanced techniques and models to achieve the goal of investigating their dynamics. We have determined the wave vector dependence of the autocorrelation function at scattering vectors up to $\approx 1 \text{ \AA}^{-1}$. Our first results indicate that the diffusion takes place on two different length and time scales as the data are best described by an extended Chudley-Elliott model (see Fig. 1). It has been shown [6] that micro-inhomogeneities of a size similar to these jump lengths occur in rubidium borate glass. This supports the idea of jump diffusion linked to the size of the inhomogeneous regions, i.e., to the medium-range order scale of this material. Further investigations on alkali borate glasses promise insight into the diffusion processes. Managing the measurement of fast diffusion in rubidium borate glasses opens new possible fields of even faster diffusion processes of the lighter alkali ions, which yield higher ionic mobilities and superior conductivity properties.

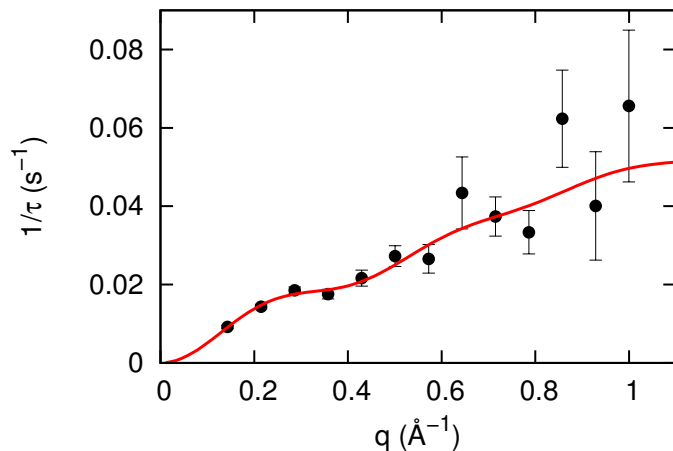


Fig. 1: Preliminary evaluation of an aXPCS measurement of 2(Rb₂O)98(B₂O₃) glass: Inverse correlation times as a function of the wave vector transfer at 150 K, fitted with the Chudley-Elliott model.

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