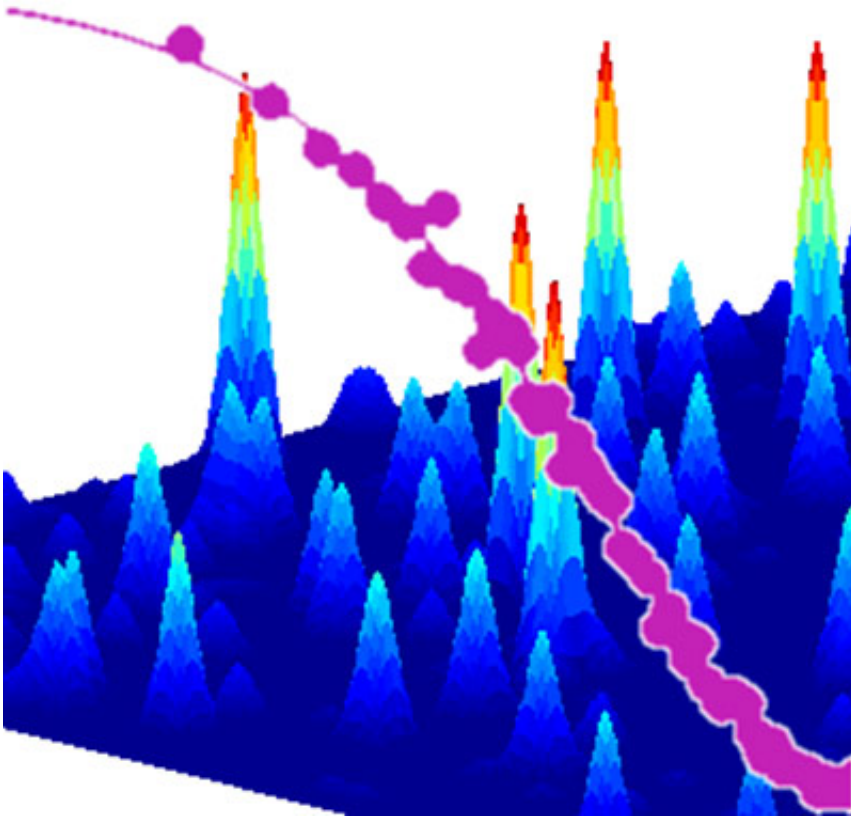


# Revealing the fast atomic motion of network glasses with coherent X-rays

June 11 2014

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3D representation of the weak speckle pattern arising from coherent X-ray illumination of network glasses at the atomic scale. The temporal evolution of the speckle intensity provides information on the atomic motion in the material (curve).

The remarkable properties of glasses are due to their dynamical arrested state in which relaxations occur on time scales too large to be observed,

or so it was believed. Now researchers have discovered the existence of unexpected fast atomic rearrangements occurring within a few minutes for the atomic motion of a silicate glass, even while it was in the deep glassy state. These results modify our conception of the glassy state and call for a new microscopic theory.

Glasses are essential materials in present day science and technology. The optimisation of materials properties for technological applications is often the result of a trial-and-error approach, as a basic understanding of many of their properties is still lacking. In particular, the intrinsic non-equilibrium nature of glasses poses formidable problems both for experiments and theory. Therefore, while important progress has been made in the understanding of the dynamics of their high temperature phases, the so called glass-forming liquid phase, a microscopic theory of glasses is still missing due to the extreme difficulty of probing the structural relaxation of a glass far below the [glass transition temperature](#)  $T_g$ , either with experiments or simulations [1].

Thanks to improved instrumentation for the collection of sparse scattering signals [2] and to an increased flux and coherence of X-ray beams at beamline ID10, X-ray photon correlation spectroscopy (XPCS) has become a very powerful technique able to follow the evolution of the dynamics at the atomic length scale in crystalline and amorphous materials [3].

An international research team used XPCS at beamline ID10 to follow the temporal and temperature evolution of the relaxation dynamics in a prototypical network glass, a sodium silicate, at both the mesoscopic and atomic length scales for the first time. The experiments showed the existence of fast collective atomic motion in the glassy state; this is in contradiction with the common idea of an ultraslow arrested out-of-equilibrium state: upon cooling the system from the melt to the glassy state, the structural relaxation time strongly departs from the supercooled

liquid behaviour and displays a weak temperature dependence characterised by relaxation times of 50-1000 s even in the deep glassy state (Figure 1 and Figure 2). This fast [atomic motion](#) is accompanied by the absence of any detectable physical aging not even in the glass transition region, in disagreement with macroscopic studies and theoretical approaches (Figure 1) [1]. Indeed current phenomenological models for relaxation in glasses, like the TNM model [1], assume that the physical aging takes place on a time scale comparable to the structural relaxation time, and this is usually the case for macroscopic observables. Differently from macroscopic studies, the microscopic dynamics now unveiled remains stationary in the glassy state, even in those cases when the annealing time is much longer than the structural relaxation time expected from a simple extrapolation of the supercooled liquid relaxation times. These results indicate a different physical mechanism responsible for the relaxation dynamics at the microscopic level, something that is novel and not predicted by previous experimental and theoretical work.

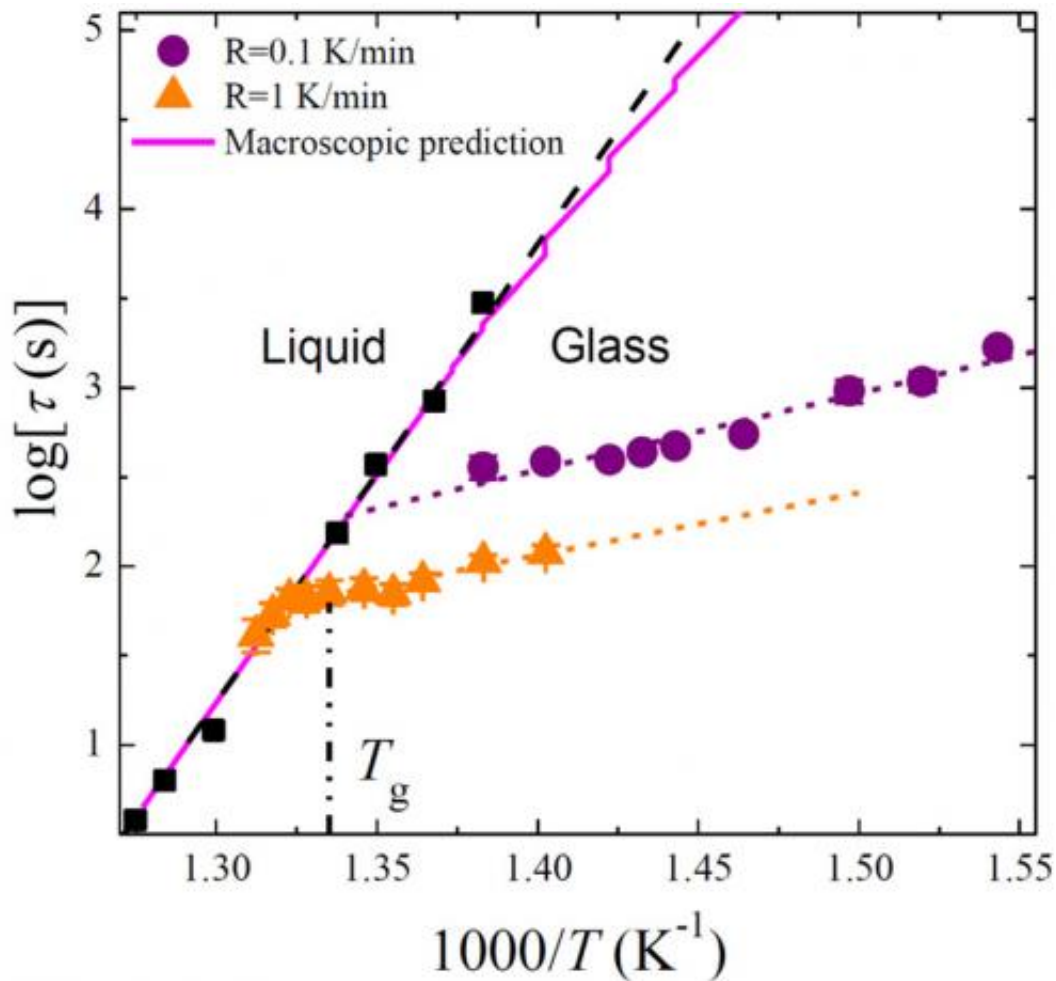


Figure 1. Temperature dependence of the structural relaxation time in the supercooled liquid and glassy state. Arrhenius plot of the relaxation times measured with XPCS at  $Q_{\text{max}}=1.53\text{\AA}^{-1}$  (coloured symbols) by cooling from the ultraviscous liquid with different cooling rates  $R$ . Black symbols are macroscopic data taken from Literature. The dashed lines are Arrhenius fits while the full line is the macroscopic prediction of the TNM model.

Some insight in this novel dynamics can be gained by looking in detail at its dependence on the scattering vector: the [relaxation time](#) shows two different peaks, one close to the position of the maximum of the static structure factor, and a second maximum at the mesoscopic scale, in correspondence to a well-characterised structural peak, known as the

prepeak. While the first one is reminiscent of the de Gennes-like narrowing also observed in coherent neutron scattering experiments of supercooled liquids, the second one is in correspondence to the characteristic distance between the sodium diffusion channels in the network, in agreement with high temperature simulation in molten silicates [4].

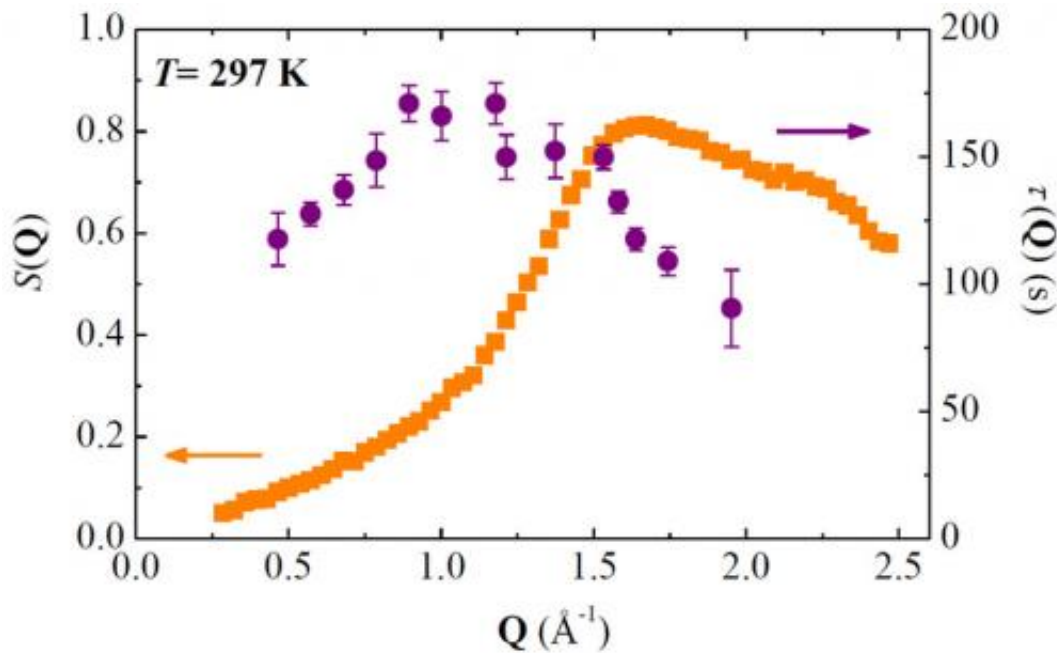


Figure 2. Wave vector dependence of the relaxation times (circles) measured in a silicate glass cooled at ambient temperature with a rate of 100 K/s from the high temperature melt. The data are reported together with the corresponding static structure factor (squares).

Overall this observed complex microscopic dynamics calls for a reexamination of phenomenological models and a need for a new microscopic theory for network glasses.

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