Highlight #6: DISORDERED AND MULTISCALE SYSTEMS and METHODOLOGIES
Glasses by X-ray Photon Correlation Spectroscopy

Glasses are liquid frozen in an amorphous state, far from thermodynamic equilibrium: they flow on timescales larger than device life, human life or even thousands of years depending on compositions and temperatures. As a consequence, all properties of glasses have a memory of the thermo-mechanical treatments applied to the material, making difficult their control during processing and applications. By coupling extreme pressures to coherent X-ray scattering in one of the world most brilliant X-ray source, we made possible density dependent observations of the microscopic "atomic rearrangements" within glasses. Our results reveal a counter-intuitive behavior in a model metallic glass: under pressure, atoms actually move faster in compact environment, even though the available space is reduced.

Despite being the oldest man-made materials, and being ubiquitous in our daily lives, glasses remain puzzling to researchers. In particular, we still lack a comprehensive microscopic theory to understand how they form from their liquid parents and their spontaneous evolution with time. Everything starts during cooling a liquid below its melting temperature fast enough to avoid crystallization. In this case, the system enters a metastable state called supercooled state where the liquid viscosity increases by up to 16 orders of magnitude until the liquid flow is not measurable in typical observation times and a glass is formed.

As such, glasses are out-of-equilibrium materials (one would see a normal liquid behavior if the observation time is long enough, in the order of 104 years for silicate glasses at room temperature!) and are defined dynamically (their structure is virtually indistinguishable from that of the liquid). This implies that we need a dynamical description of glasses, a quest far more complex than getting an averaged structural description. Coherent X-rays can help: when they are scattered by the sample, they form a scattering pattern which is a footprint of the exact, non-averaged structure. Following this scattering pattern over time provides information on the temporal evolution, and thus the dynamics, of the constituents (atoms, molecules…). Yet, so far, only light sample environments could be used with coherent X-rays, enabling mainly temperature dependent studies. Here, we manage to investigate the dynamical properties of a model metallic glass at extreme pressures (comparable to that of the earth upper mantle) at the atomic scale, by placing our sample in a specific high-pressure apparatus: a Diamond Anvil Cell, which allows for the pressure generation, the preservation of the X-ray's coherence, and the required sample stability below the micron scale. This enables the study of the density dependence of the microscopic relaxation processes which control the macroscopic properties of the glass. Our results highlight a complex behavior under pressure: while the density increases monotonously as expected, the microscopic dynamics shows a two-stages evolution, including an acceleration of the atomic motion in the first part. This acceleration is really counter-intuitive: it's like people moving faster in a crowded environment. Our analysis suggests that this strange dynamical behavior is stress driven, and characterized by sudden relaxation events involving a large ensemble of atoms, a feature visible in simulations and called cascade relaxations. This eventually disappears at large pressures, when pressure dramatically slow down the particle motion, and is in agreement with a recently developed theoretical model.

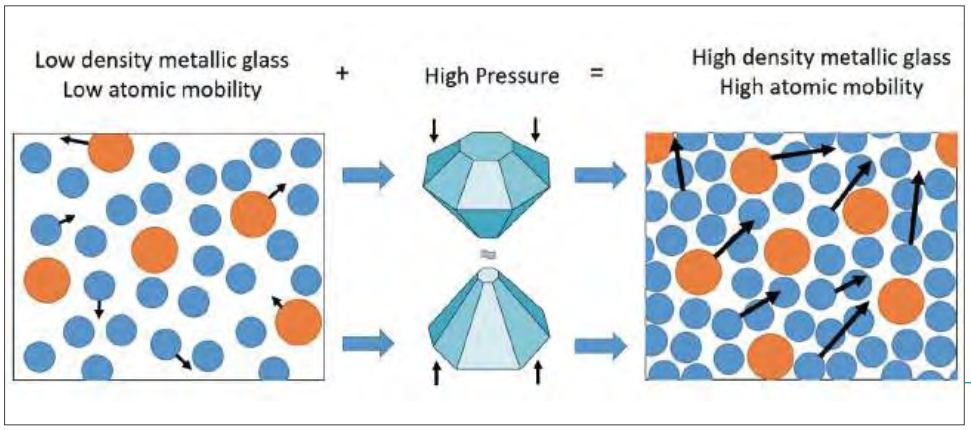

Fig.: *Sketch of the pressure induced acceleration of the atomic motion. At high pressure, the density increases and the dynamics is hyper-diffusive with a collective ballistic-like atomic motion.*

References

Antoine Cornet *et al.*, *Acta Materialia* **255**, 119065 (2023).

"High-pressure X-ray photon correlation spectroscopy at fourth-generation synchrotron sources", *J Synchrotron Rad* **31** (2024).