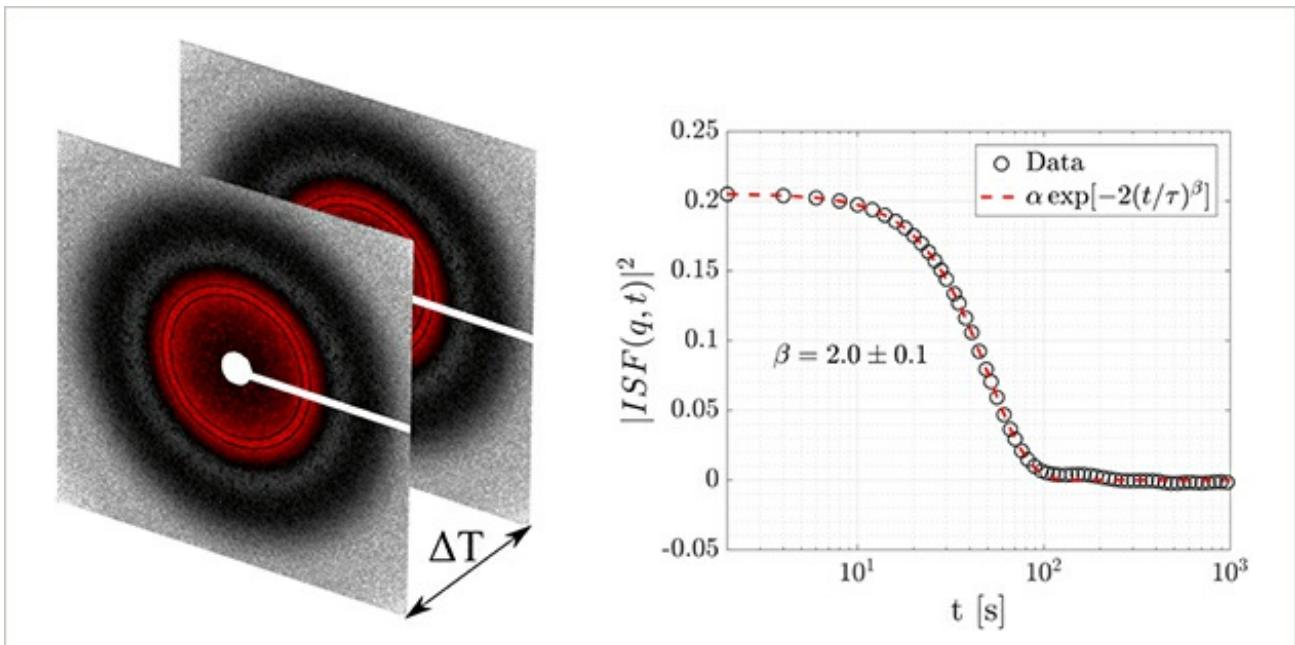


Photon Correlation Spectroscopy applied to colloidal glasses

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Small angle X-ray scattering pattern (left) for a colloidal glass of 100 nm silica nanoparticles in a water-lutidine solution. Images are collected at delays ΔT and the time autocorrelation is performed averaging over pixels corresponding to the same exchange wave-vector (black rings). The Intermediate Scattering Function (right) shows very clearly a super-diffusive behaviour ($\beta > 1$).

Suspensions of nanoparticles, also known as colloids, can be found in everyday life. Depending on the volume fraction of nanoparticles, colloids can be prepared either in liquid form or – for concentrations close to or larger than a critical one – in an arrested state. If crystallization is avoided, one obtains a so called colloidal glass. These systems are commonly used in industry (paints, dye, clays) and medicine (drug delivery, sentinel lymph-node radiotracers). They are well known for the possibility to tune their mechanical properties, and in fact they are very versatile; at the same time, however, they also show a very pronounced aging behaviour which strongly limits their use in several potential applications. A large effort is therefore devoted to investigate the physics beyond the aging process and in general the glassy dynamics. Several theories and experiments suggest that the dynamics near the structural arrest becomes cooperative, leading to the development of highly correlated regions (called dynamical heterogeneities) with dimensions that increase on approaching the arrested state.

One of the most successful techniques to investigate slow dynamical properties typical of glassy

systems is Photon Correlation Spectroscopy (PCS). PCS is a scattering technique based on the evaluation of the time-autocorrelation function of the scattered intensity, and provides information on the dynamics at length scales of the order of the inverse of the scattering vector \mathbf{q} , with \mathbf{q} is the scattering angle. This technique requires the coherence of the incident radiation and for this reason it was first developed with visible lasers. In the last decade, thanks to the advent of third generation synchrotron radiation facilities, its X-ray counterpart (XPCS) has been developed, opening the way to experiments on a wider range of exchanged wave-vectors q . The main quantity extracted from these experiments is the so called Intermediate Scattering Function (ISF). The latter is a decaying function that gives information on the typical time (τ) that a particle needs to move over a distance $\sim 1/q$. Moreover, from the ISF it is possible to extract the stretching parameter (β) which is strictly related to the type of dynamics underlying the particle motion. For example, for a Brownian diffusive motion one expects $\beta=1$.

In recent years, it has become clear that colloidal glasses age differently than common glasses, and that the dynamics underlying the relaxation process is very peculiar. In atomic glasses, one usually observes a stretching parameter $\beta < 1$, symptom of a heterogeneous distribution of relaxation processes. On the other hand, it is well established that in soft matter the dynamics often evolves with a super-diffusive ($\beta > 1$) dependence. This feature is attributed to the presence of relevant stress fields in these materials, and both theoretical simulations and experiments qualitatively confirm this picture. Nevertheless, there is an intense research aiming at a full understanding of these stress-induced dynamics and of its properties.

In our work, we use XPCS in Small Angle geometry (SAXS) to investigate both dynamic and structural properties of a colloidal glass of 100 nm silica nanoparticles in a water-lutidine binary mixture, and learn about the relevant length scale of the involved processes. The system presents a compressed ISF ($\beta > 1$), and an accurate analysis of the data shows that aging proceeds through an intermittent and heterogeneous cooperative dynamics, characterized by a sequence of micron-scale rearrangements very fast on the typical aging timescale.