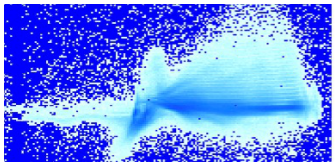

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Spatio-temporal correlations to study timescales of higher-order correlations in liquids and glasses

When a supercooled liquid approaches the glass transition, its relaxation time increases by several orders of magnitude while the liquid-like structure remains [1]. Although there has been extensive research into the nature of the glass transition, its mechanisms remain mostly unclear. Recent works show that long-living locally favored structures (LFS), such as icosahedral structures forming upon supercooling, may play a key role [2].
In this contribution we show results from a combined X-ray Photon Correlation Spectroscopy (XPCS) and X-ray Cross Correlation Analysis (XCCA) experiment on colloidal hard spheres in the vicinity of the glass transition [3]. We defined a new correlation function g_c probing the timescales of higher-order correlations, which combines both XPCS and XCCA by tracking the time evolution of the structural higher-order correlations within the sample. We observed an increase in the ratio of the relaxation times of g_c and the standard individual particle relaxation time obtained by XPCS from about 0.4 to 0.9. While a value of around 0.5 is expected for free diffusion, the increasing values suggest that the local orders within the sample are becoming more long-lived approaching the glass transition. These results indicate that not only the presence but also the lifetime of LFS grows close to the glass transition. This new correlation approach can in principle be extended to detect structure-dynamics correlations on many length scales, allowing studies of phase transitions or lifetimes of transient structure in liquids and benefits exceptionally from the increased brilliance of diffraction-limited storage rings.

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