Nanosecond Two-Pulse X-ray Speckle Visibility Spectroscopy at LCLS

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The Linac Coherent Light Source (LCLS) has recently demonstrated the capability to produce two X-ray pulses separated in time by multiples of the accelerator radiofrequency period (i.e. 350ps) [1]. This opens up new opportunities to extend X-ray Photon Correlation Spectroscopy (XPCS) studies far beyond the milli- and micro-second timescales currently reachable with 3rd generation synchrotron sources. This is accomplished with Two-Pulse X-ray Speckle Visibility Spectroscopy (2P-XSVS) by which the contrast of summed speckle patterns $\beta(Q,\Delta T)$ from two independent FEL pulses is evaluated as function of their time separation $\Delta T$ and the wavevector transfer $Q$ [2]. This approach allows measurement of Q-dependent relaxation times far beyond the intrinsic repetition rate of the FEL down to the nanosecond timescales (i.e. limited by $\Delta T$).

Following the recent demonstration of this technique with soft X-rays [3], we present here the first demonstration of 2P-XSVS measurements using this unique LCLS operation mode in the hard X-ray regime on a model system. A transmissive diagnostic was developed to provide the intensity ratio $\rho$ between the two incident FEL pulses [4]. This allows classifying the contrast of summed speckle patterns as a function of $\rho$, which is critical in extracting characteristic relaxation times from the analysis.

We first used a static reference sample to optimize and characterize the optical properties of the setup (i.e. beam size and beam profile of each pulse, spatial overlap of the two pulses, etc.). We then investigated the diffusion behavior of a model “Brownian” system. It consists of 30nm diameter spherical gold colloidal nanoparticles dispersed in water with a volume fraction $\Phi\approx0.03\%$. We specifically characterized the contrast of the summed speckle patterns $\beta$ for two time separations $\Delta T=49$ and 210ns. The wavevector dependence of $\beta$ was also obtained, from which the diffusion constant of the colloidal particles could be extracted. We show that this result qualitatively matches the prediction of established theoretical model for free Brownian particles.

References

References