SRI2024

Contribution ID: **426** Type: **Contributed talk**

Real-Time Kinetics of Nanogels Driven by XFEL Pulses with MHz Repetition Rate

Wednesday 28 August 2024 17:15 (15 minutes)

Stimuli-responsive polymers are an important class of materials with many applications in nanotechnology and drug delivery. The most prominent one is poly-N-isopropylacrylamide (PNIPAm) which has a lower critical solution temperature (LCST) around 32 ℃. Below the LCST the polymer is swollen with water, while at higher temperatures the water is expelled from the polymer. The characterization of the kinetics of this configurational change after a temperature jump is still a lively research topic, especially at nm-length scales where it is not possible to rely on conventional microscopic techniques. In this contribution we show results from a real-time experiment of the collapse of a PNIPAm shell on silica nanoparticles with MHz X-ray photon correlation spectroscopy (XPCS) at the European XFEL [1]. Here, the X-ray pulses act both as pump for heating the sample and probe to determine the sample's structure and dynamics [2], see figure. We characterize the changes of the particles'diffusion constant as a function of time and consequently local temperature on sub-μs time scales. We developed a phenomenological model to describe the observed data and extract the characteristic times associated to the swelling and collapse processes. In contrast to previous studies tracking the turbidity of PNIPAm dispersions and using laser heating, we find collapse times below μs time scales and two to three orders of magnitude slower swelling times. Finally, we discuss consequences for μs dynamics studies on soft and biological matter at MHz XFEL facilities.

[1] F. Dallari, I. Lokteva, J. Möller, W. Roseker, C. Goy, F. Westermeier, U. Boesenberg, J. Hallmann, A. Rodriguez-Fernandez, M. Scholz, R. Shayduk, A. Madsen, G. Grübel, and F. Lehmkühler. Science Advances 10, eadm7876 (2024).
[2] F. Lehmkühler, F. Dallari et al. PNAS 117, 24110 (2020).

I plan to submit also conference proceedings

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Session Classification: Mikrosymposium 6/2: FELs: New facilities and Opportunitites

Track Classification: 6. FELs: New facilities and opportunities