Self-driving synchrotron coherent X-ray scattering on complex fluids

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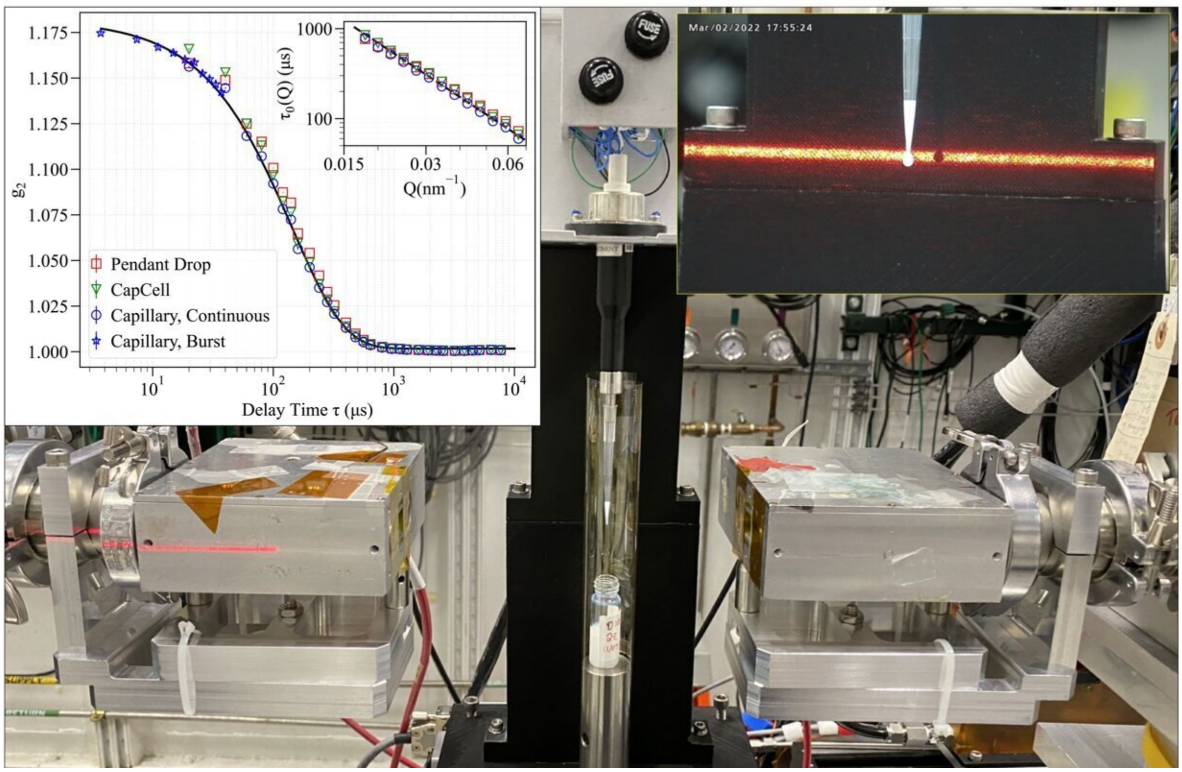
(a)



(a) SA-XPCS measurement on the pendant drop setup at Beamline 8-ID-I, Advanced Photon Source. The inset on the left shows the comparison of the XPCS results from the pendant drop and the reference setups, and the inset on the right shows a zoom-in optical image of the drop hanging from the pipette tip during the measurement. The red laser is used for coarse aligning of the x-ray beam in the vertical direction. (b) 'Digital Twin' of the robotic pendant drop setup in Nvidia Isaac simulation, where the electronic pipette is docked on the mounting plate for x-ray measurements. The red lines and arrows indicate the incoming and scattered x-ray beams. The items labeled in the figure are: 1. Robotic arm; 2. Electronic pipette; 3. Mounting plates; 4. Robotic tool changer; 5. Reflective mirror with 1 mm diameter x-ray passthrough; 6. Optical microscope and camera system; 7. Sample preparation station with PCR plates and pipette tips. (c) The robotic pendant drop setup in the adjacent chemical laboratory of the Beamline 8-ID-I, where the electronic pipette is shown picking up a fresh pipette tip for liquid handling. The inset on the top right shows the suspended pendant drop captured by the inline optical system. Credit: Qingteng Zhang

Soft materials are ubiquitous in our daily lives, from the food we eat to the products we use to the materials that make up our bodies. Some examples of soft materials include cream, toothpaste, and blood. Most soft materials are complex fluids, which means that they contain a macroscopically uniform mixture of two or more phases.

The dynamic competition between the structures of phases in a soft material can have a significant impact on not only its properties, but also the tunability and reversibility of these properties. For example, some liquid becomes more fluidic temporarily after a shear is applied (a.k.a. shear-thinning).

Ketchup is designed this way so that it flows more easily when it is squeezed out of a bottle, and sits still when it is on top of a plate. Understanding the spontaneous dynamics of the spatial structures formed by competing phases under various conditions is therefore essential for the tailored design of soft materials.

The characterization of spontaneous dynamics in soft materials is a challenging task. Consider a vial of silica nanoparticles suspended in water, which is a relatively simple soft material. The dynamics of the nanoparticles (i.e., Brownian motion) occur at the nanometer range and microsecond timescale, which makes it impossible to track the exact location of every particle in the vial at every moment.

Such information may also be unnecessary as the macroscopic properties of the soft material is usually determined by the statistics of the dynamics, i.e., how fast the system evolves at a particular length scale. This is the fundamental quantity reported by photon correlation spectroscopy (PCS, also known as Dynamic Light Scattering).

In PCS, an optical laser is transmitted through the nanoparticle suspension and the variation of the nanoparticle position is evaluated via the temporal decorrelation of the intensity of the scattered light. However, PCS is not suitable for materials that are opaque. In addition, PCS cannot measure diffusivity of systems where the particles are not freely diffusive and the dynamics cannot be described by Einstein-Stokes equation (e.g., colloidal gels).

These limitations were eventually addressed by the development of X-ray Photon Correlation Spectroscopy (XPCS).

XPCS is a powerful technique for characterizing the spontaneous dynamics of soft materials. It uses a spatially-coherent (i.e., "laser-like") X-ray beam to probe the dynamics at all length scales within the micron-nanometer range. This is made possible by the use of a large pixelated area detector, which allows the dynamics to be recorded simultaneously at all length scales since the scattering angle is inversely proportional to the length scale it represents.

The biggest disadvantage of XPCS is that it is much less available than PCS. First of all, there are currently fewer than 10 synchrotrons worldwide capable of performing XPCS experiments. Second of all, the coherent X-ray is obtained by spatially cropping the synchrotron X-ray beam to select the coherent portion, which results in a 10 to 100 times reduction of the X-ray flux. However, these issues are being addressed by the global construction and commissioning of fourth-generation X-ray sources.

These sources will increase the coherent X-ray flux by up to 100 times, therefore reducing the measurement time of flux-limited XPCS characterizations by up to 10,000 times. While this will significantly increase the availability of XPCS, it will also create a new bottleneck: the human bandwidth. The facility users will not be able to make that many samples or process that much information. This challenge, however, is an ideal fit for the rapid-growing field of AI and robotics.

In a new paper published in *Light: Science & Applications*, a team of scientists, led by Dr. Qingteng Zhang from the Advanced Photon Source (APS) at Argonne National Laboratory, has developed an AI-executable, end-to-end-automated XPCS workflow for the study of spontaneous dynamics in complex fluids.

The study is performed in two stages. Stage 1 is conducted at Beamline 8-ID-I of APS. The complex fluid studied in this paper consists of 100 nm-diameter silica nanoparticles suspended in water with a volume fraction of 2.5%. A drop of the sample is dispensed using an electronic pipette and hung from the end of a pipette tip, and the XPCS data are collected by shining the synchrotron X-ray beam through the drop for a few seconds.

At the end of the measurement, the drop is aspirated back into the pipette tip for disposal without human-handling. The XPCS results from the pendant drop are then compared against two reference sample setups to validate the pendant drop setup for the use of XPCS measurements. Reference setup 1 is a 40 mm-length thin-walled quartz capillary from Charles Supper Co. Inc., and reference setup 2 is an aluminum liquid cell sealed with externally-threaded polycarbonate caps.

Both setups are commonly used for Small-Angle X-ray Scattering (SAXS) and XPCS measurements on complex fluids, and the second setup has higher temperature accuracy due to the direct contact of the liquid sample with the aluminum cell body.

The main challenge in stage 1 is resolving the microsecond dynamics of sub-micron-sized nanoparticles in water. This is only possible using a Rigaku XSPA-500k single-photon-counting pixelated array detector with a continuous frame rate of up to 50 kHz. The XSPA-500k detector is also equipped with a burst mode capacity that enables a burst of up to 24 frames, each with an exposure time as short as 1 microsecond, to be externally triggered as frequently as 1 kHz.

In the paper, an exposure time of 3.7 microseconds and a burst of 12 frames are used, resulting in an effective frame rate of 272 kHz and a duty cycle of 4.4%. After data acquisition, the XPCS analysis is performed automatically on the Argonne supercomputing clusters using the APS Data Management workflow, and the results are visualized and re-rendered using open-source software suites, leading to near real-time data interpretation that can help beamline users decide what measurements to perform next.

"The frame rate of the X-ray detector is critical for XPCS as it determines the time resolution of the measurements. By pushing the time resolution of XPCS closer to PCS, the synchrotron X-ray community can benefit from the knowledge base of the light scattering community," said Dr. Qingteng Zhang, the corresponding author of this paper. "Additionally, the substantial volume of data generated by large-area, high-frame rate detectors makes an automatic data management workflow an indispensable component of high-speed XPCS measurements."

Stage 2 is conducted in the adjacent chemistry lab of Beamline 8-ID-I, where the electronic pipette is mounted on a robot arm. This setup enables the preparation of complex fluid samples with precise and repeatable chemical compositions through robotic positioning and electronic pipetting.

All robotic motions are programmed using open-source software (e.g., Python), and orchestrated using the Workflow Execution Interface (WEI) developed at Argonne National Laboratory. WEI allows intricate workflows to be divided into modules, with each module specified in human-readable text format (e.g., YAML). It also utilizes various executors, such as the Python interface to the Experimental Physics and Industrial Control System (pyEPICS) and Robotic Operating System (ROS), to facilitate inter-module communication.

"The modular approach in WEI really simplifies robotic integration because you can reuse the modules you made for other robotic programs with completely different goals," said Mr. Doga Ozgulbas, the lead author of the paper. "I can also import the ROS modules into the Nvidia Isaac simulation to create a 'Digital Twin' of the real world, where I can optimize the positions of objects and check for possible collisions to make sure the robotic program is safe. It is a valuable tool to have."

While the pendant drop setup is not compatible with soft materials that cannot be pipetted, such as gels and soft tissues, these materials can be loaded into the aluminum Cap Cells, one of the reference setups for validating the pendant drop in Stage 1. Loading can be performed either by the beamline user before the experiment or by the robot during the experiment. The automation of both sample handling and XPCS analysis can be combined with AI-assisted result interpretation to achieve a closed-loop, self-driving experiment.

"At APS, we strive to ensure that the limitations of instruments or bandwidth in users' home labs do not hinder their scientific pursuits," added Dr. Qingteng Zhang. "The automation infrastructure we are developing, both in terms of hardware and software, could potentially benefit the entire synchrotron X-ray user community and hopefully contribute to the autonomous design and discovery of functional materials across various disciplines."

More information: Doga Yamac Ozgulbas et al, Robotic pendant drop: containerless liquid for μs-resolved, AI-executable XPCS, *Light: Science & Applications* (2023). DOI: 10.1038/s41377-023-01233-z

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