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Nanosecond dynamics measured with Split-Pulse X-ray Photon Correlation Spectroscopy At Free Electron Laser Sources

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One of the important challenges in condensed matter science is to understand ultrafast, atomic-scale fluctuations that dictate dynamic processes in equilibrium and non-equilibrium materials. These fluctuations can be measured on the relevant time scale by time-correlating coherent scattering speckle patterns taken with ultrashort X-ray pulses from a free electron laser (FEL) in an X-ray Photon Correlation Spectroscopy (XPCS) experiment if the time separation of the X-ray pulses can be controlled on that very timescale. Here, we report an important step towards reaching that goal by using a prototype perfect crystal-based split-and-delay system, capable of splitting individual X-ray pulses and introducing femto- to nanosecond time delays. We show the results of the first ultrafast XPCS experiment[1] at LCLS where split X-ray pulses were used to measure the dynamics of gold nanoparticles suspended in a liquid and verify the validity of the Stokes-Einstein relation on ns-ps timescales. We show how reliable speckle contrast values can be extracted even from very low intensity FEL speckle patterns by applying maximum likelihood fitting, thus demonstrating the potential of a split-and-delay approach for dynamics measurements at FEL sources. This capability promises to elucidate the underlying dynamics of a wide variety of systems and will enable the discovery of new physical processes therein.

References:
[1] Roseker, W. et al. Nature Communications 9,1704 (2018).

Primary authors: Dr ROSEKER, Wojciech (Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany); Dr HRUSZKEWYCZ, Stephan (Argonne National Laboratory, Argonne, Illinois 60439 USA); Dr LEHMKÜHLER, Felix (Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany); Dr WALTHER, Michael (Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany); Dr SCHULTE-SCHREPPING, Horst (Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany); Dr LEE, Sooheyong (Korea Research Institute of Standards and Science, Daejeon 305-340, Rep.of Korea); Dr OSAKA, Taito (RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan); Dr STRÜDER, Lothar (PNSensor GmbH, Otto-Hahn-Ring 6, 81739 München, Germany); Dr HARTMANN, Robert (PNSensor GmbH, Otto-Hahn-Ring 6, 81739 München, Germany); Dr SIKORSKI, Marcin (European X-Ray Free-Electron Laser Facility, Holzkoppel 4, 22869 Schenefeld, Germany); Dr SONG, Sanghoon (LAC National Accelerator Laboratory, Menlo Park, CA 94025, USA); ROBERT, Aymeric (LAC National Accelerator Laboratory, Menlo Park, CA 94025, USA); Dr FUOSS, Paul (LAC National Accelerator Laboratory, Menlo Park, CA 94025, USA); Prof. SUTTON, Mark (McGill University, Montreal, Quebec H3A2T8, Canada); Dr STEPHENSON, Brian (Argonne National Laboratory, Argonne, Illinois 60439 USA); Prof. GRÜBEL, Gerhard (Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany)

Presenter: Dr ROSEKER, Wojciech (Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany)

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