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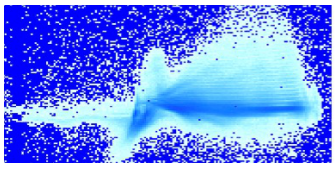

COHERENCE 2024
11th International Conference
on Phase Retrieval
and Coherent Scattering

MAX IV

Book of Abstracts

Contents

X-ray Photon Correlation Spectroscopy / **134**

Chair

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X-ray Photon Correlation Spectroscopy / **62**

Reaching the Yield Point of a Glass During X-Ray Irradiation

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A solid loaded beyond the yield stress loses its elastic properties and becomes plastic. From a microscopic point of view, this limit corresponds to the condition where plastic regions become so densely packed that they give rise to system-spanning structures. This limit for glasses is abrupt, which makes experimental in-vestigations challenging. Here, the yield point is reached by the alternative ap-proach of increasing the density of plastic regions by generation of point defects during x-ray irradiation. For the case of a LiBO2 glass, we show that at low doses, i.e., for a low density of defects, the defects behave as isolated stress sources that induce atomic displacements typical of an elastic solid. As the density of defects in-creases, the mechanical response of the glass at the local scale changes from elastic to more and more plastic, until reaching the limit where it becomes charac-teristic of a flowing system, which signals that the yield point is reached.

X-ray Photon Correlation Spectroscopy / **81**

Probing protein dynamics using XPCS despite radiation damage.

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X-ray Photon Correlation Spectroscopy (XPCS) is a well-established technique to study slow dynamics in disordered materials at nanometer down to angstrom length scales [1]. The method exploits the coherent fraction of the synchrotron radiation and benefits enormously from the recent upgrade of the ESRF source (EBS) [2]. The high degree of coherence opens new avenues for application of XPCS. One of the promising avenues is the study of dynamics in concentrated protein solutions at nearest neighbor distances. Diffusion of proteins on length scales of their own diameter in highly concentrated solutions is essential for understanding biological systems such as a living cell, but its experimental characterization remains a challenge. Our work addresses this problem and discusses the use of X-ray Photon Correlation Spectroscopy at a recently upgraded 4th generation synchrotron source for this purpose. While X-ray radiation damage was generally believed to seriously threaten the application of XPCS to biological systems, we now present a dedicated experimental

and analysis strategy [3] to overcome this obstacle. We report a successful test of this approach to highly concentrated solutions of the eye lens protein alpha crystallin [4], which has previously been established as a model protein exhibiting the classic behavior of hard sphere colloids under these conditions [5]. The thus obtained intrinsic relaxation times for so-called long-time cage diffusion indeed agree with macroscopic measurements of the zero shear viscosity [6]. Our experiments also reveal a complex dependence of the key structural and dynamic properties of the protein solutions on both the total absorbed radiation dose as well as the dose rate. We discuss possible mechanisms responsible for the observed radiation effects and their consequences for future applications of XPCS.

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X-ray Photon Correlation Spectroscopy / 31

Spatio-temporal scaling laws in a heated egg yolk

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Denaturation, aggregation and gelation of proteins and lipids are biologically relevant out-of equilibrium processes which are coupled by a hierarchy of length, time and energy scales. Finding the characteristic scaling laws governing these processes on the relevant time and length scales is necessary to predict the changes of biomolecules to future time scales.

Here, we use heated egg yolk as a model system to reveal the spatio-temporal relationships underlying these intricate processes for a wide range of time and temperature combinations [1]. Using low-dose X-ray photon correlation spectroscopy (beamline P10 at PETRA III, Hamburg, Germany) in ultra-small angle X-ray scattering geometry, we follow the time-resolved structural and dynamical evolution of multiple non-equilibrium processes occurring in a heated hen egg yolk. Following key structural and dynamical features, we identify non-equilibrium processes such as denaturation and aggregation of proteins, protein gelation, gel ageing, two-step aggregation of yolk low-density lipoproteins (LDLs), and gelation of yolk granules. We find that the overall kinetics and dynamics governing protein denaturation, aggregation, and gelation follow Arrhenius-type time-temperature superposition (TTS). This implies an identical mechanism underlying these consecutive processes, with a temperature-dependent reaction rate. At high temperatures, TTS breaks down during gelation and temperature-independent gelation dynamics is observed. This indeed reflects the complex association of protein aggregates that results in a gel network. In a broader sense, our research [1] provides an illustration of how to comprehend the fascinating non-equilibrium events in inherently complex, multi-component, thermally driven biological systems on length scales ranging from nanometers to micrometers in a time spectrum of milli-seconds to hours.

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X-ray Photon Correlation Spectroscopy / **88**

Anomalous Protein Diffusion and Solvent-mediated interactions in Crowded Solutions with Coherent X-Ray Scattering Using XFELs

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Proteins play essential roles in life, for instance serving as carriers, participants in the immune response or for their structural role. In vivo, they exist within crowded environments with protein volume fractions typically ranging as high as 30%. When the environment becomes highly concentrated, the dynamics of proteins deviate significantly from those observed in a dilute system. However, the precise mechanisms influencing these dynamics across different time scales are not yet fully understood. Here we present our recent results [1], where we investigated the effect of self-crowding on protein diffusion in a ferritin solution with varying concentrations using X-ray Photon Correlation Spectroscopy. This technique allows simultaneous monitoring of both the structure, through small angle scattering, and the diffusion, through intensity-autocorrelation functions, of the protein solution, as demonstrated in our previous study [2]. By analyzing the scattering intensity, we observed that the ferritin particles become more densely packed with increasing protein concentration, indicated by a pronounced peak in the structure factor that shifts towards lower momentum transfer values. The protein diffusion, measured at all concentrations, follows a Brownian type of motion, but exhibits deviations at the peak position. This deviation can be attributed to the crowding effect caused by neighboring proteins, which act through hydrodynamic interactions. The hydrodynamic functions, which reflects these interactions, exhibit a peak which coincides with that of the structure factor indicating the connection of the crowding and the hydrodynamic interactions. To elucidate the underlying mechanism, we compare the hydrodynamic functions with estimations based on the δγ-theory, which considers the non-trivial interactions between particles. The model indicates that the protein diffusion is slower than that of non-interacting hard spheres due to the presence of solvent-mediated interactions and effective local friction between the particles.

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Ptychography at different wavelengths - S1 / **135**

Chair

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Ptychography at different wavelengths - S1 / **127**

Recent advances of Bragg ptychography and perspectives at 4th generation synchrotron

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Bragg ptychography is a phase retrieval method combining crystalline sensitivity and the possibility to image extended 3D samples [God2011, Mas2017]. Recent progresses in Bragg ptychography formalism, namely, the introduction of the back projection operator [Hru2017] and the advent of 4th generation synchrotron sources have enabled the use of 3D Bragg ptychography with unprecedented accuracy, sensitivity and spatial resolution. Specifically, the probe retrieval [Li2021] and the beam-position refinement [Li2022] are now made possible and provide improved image quality.
In this talk, I will first detail recent advance of Bragg ptychography. Using a series of preliminary results, I will further highlight the perspectives offered by 4th generation synchrotron sources, in terms of temporal resolution and strain analysis. I will finally present a project of integration of Bragg ptychography at synchrotron beamlines, performed with collaborators from ESRF, MAXIV and Diamond. It aims at enabling the use of this 3D crystalline microscopy approach to a larger x-ray community.

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Ptychography at different wavelengths - S1 / 97

Single-shot ptychography at free-electron laser

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Ptychography is a scanning coherent diffraction imaging technique capable of simultaneous imaging of extended samples and beam characterization with diffraction-limited resolution. It is well developed at synchrotrons, but its scanning nature prevents its use for single-shot imaging at wide applications on FEL facilities.

Single-shot ptychography can be performed by collecting the diffraction patterns of multiple overlapping beams in one shot, thus measuring the whole dataset at once and removing the need for scanning. A setup realizing this principle was proposed for visible light[1]; however, it cannot be straightforwardly applied to X-ray due to the use of refractive optics.

We solved this problem by using a single-shot ptychography setup based on a combination of X-ray focusing optics and beam-splitting grating and a corresponding forward model that facilitates single-shot imaging of extended samples at soft X-ray wavelengths [2]. The setup was tested during the proof of concept experiment at the free-electron laser FLASH at DESY and allowed us to obtain a reconstruction of a test sample and probe wavefield from the data measured with a single pulse of FLASH. However, the fidelity and resolution of the reconstruction were limited by the low performance of the diffraction grating and the inability of the forward model to fit the inter-beamlet interference.

Here, we present further progress in the single-shot psychography at FELs. We used an improved experimental setup based on the Damman grating [3] with higher diffraction efficiency and diffraction order uniformity and an improved forward model. These improvements allowed us to perform single-shot ptychographical imaging and beam characterization during the beamtime at FLASH. This technique further improved and adapted for harder X-rays, will allow the high-resolution single-shot imaging of extended dynamical samples as well as the single-shot beam characterization at X-ray free-electron lasers.

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Ptychography at different wavelengths - S1 / 34

High resolution spin texture imaging in spin caloritronics prototype device structures: New opportunities for coherent hard X-rays with resonant scattering at 4th generation light sources.

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Spin caloritronics are currently a science focus due to their potential exploitation in the next generation of spintronics applications. This class of materials combine both spintronic and thermoelectric functionalities by interconversion of charge, spin and heat currents [1]. Revealing how atomic strain and magnetic structure are intertwined at the nanoscale is of central importance to the development of emerging spin caloritronic nanotechnologies [2]. The recent investment in high brilliance 4th generation synchrotron sources hold promise for the development of new microscopic tools to reveal simultaneously atomic and magnetic nano-structure.
Here, we present preliminary results from ID01 of the ESRF-EBS and NanoMAX of MAX IV Laboratory. The first combine Bragg ptychography with X-ray resonant scattering at low temperatures to investigate prototype spin caloritronic devices structures of Gd3Fe5O12 epitaxial films capped with a Platinum layer. The second are focussed on the analysis of structure and strain of diverse prototype structures at room temperature. From our analysis exploiting inverse microscopy approaches, we demonstrate the potential to correlate atomic strain and magnetic structures down to 16 nm spatial resolution.
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XPCS, polarised and chiral beams / 136

Chair

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XPCS, polarised and chiral beams / **100**

Magnetic spectroscopy and imaging with x-rays carrying orbital angular momentum

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The interaction of light beams with magnetic materials defines the rich set of analytical tools in magneto-optics, covering photon energies from infra-red to hard x-rays. In addition to the spin angular momentum (SAM) associated to the light polarization, Laguerre-Gaussian (LG) beams carry also an orbital angular momentum (OAM) of $\ell\hbar$/photon [1] associated to an azimuthal dependence $\exp(i\ell\phi)$ of the electric field phase. Over the last thirty years, OAM beams at vis-IR wavelengths found applications in biology, telecommunication, imaging and quantum technologies [2]. Their capability to exert a mechanical torque was exploited to create optical spanners for manipulating small particles. The azimuthal phase dependence introduces a singularity on the propagation axis and a radial modulation of the intensity (ring-shaped), properties that have been used to modify magnetic ordering, to improve the spatial resolution in microscopy, and to enhance the edge sharpness in phase-contrast imaging.
Over the last decade, the generation of OAM beams at shorter wavelengths, from XUV to hard x-rays, is also finding an increasing number of applications, often based on extrapolations of previous work carried out in the visible range. For instance, as it happened for the SAM, the handedness imposed by the OAM has been exploited to perform x-ray spectroscopic studies of magnetic materials [3] and of chiral molecules [4], and a recent ptychography study [5] showed that the attainable spatial resolution in the reconstructed XUV images increases with ℓ.
We will review recent extensions in the use of OAM beams from visible to short wavelengths, with focus on applications of 10-100 fs XUV-OAM pulses for element-selective spectroscopy and imaging of magnetic structures. We will show how time-resolved resonant scattering experiments offer new perspectives for tracking the dynamics of complex magnetic topologies.

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XPCS, polarised and chiral beams / **9**

Coherent Correlation Imaging for resolving fluctuating states of matter

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Fluctuations and stochastic processes are ubiquitous in nanometer-scale systems, especially in the presence of disorder. Real-space access to fluctuating states is impeded by a fundamental dilemma between spatial and temporal resolution. Averaging over an extended period of time (or repetitions) is key for the majority of high-resolution imaging experiments, especially in weak contrast systems. If, by lack of better knowledge, averaging is indiscriminate, it leads to a loss of temporal resolution and to motion-blurred images.

We present coherent correlation imaging (CCI) [1] – a high-resolution, full-field imaging technique that realizes multi-shot, time-resolved imaging of stochastic processes. The key idea of CCI is the classification of Fourier-space coherent scattering images based on the speckle fingerprints of the real-space state – even at a low photon count where imaging is not possible. Contrast and spatial resolution emerge by averaging selectively over same-state frames. Temporal resolution down to the acquisition time of single frames arises independently from an exceptionally low misclassification rate, which we achieve by combining correlation-based similarity metric with powerful classification algorithm.

We apply CCI to study previously inaccessible magnetic fluctuations in a highly degenerate magnetic stripe domain state with nanometer-scale resolution. Our material is a Co-based chiral ferromagnetic multilayer with magnetic pinning low enough to exhibit stochastically recurring dynamics that resemble thermally-induced Barkhausen jumps near room temperature. CCI reconstructs high-resolution real-space images of all domain states by holographically aided phase retrieval [2, 3] and, unlike previous approaches, also tracks the time when these states occur. The spatiotemporal imaging reveals an intrinsic transition network between the states and unprecedented details of the magnetic pinning landscape allowing us to explain the dynamics on a microscopic level.

CCI massively expands the potential of emerging high-coherence X-ray sources and paves the way for addressing large fundamental questions such as the contribution of pinning and topology in phase transitions and the role of spin and charge order fluctuations in high-temperature superconductivity.

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XPCS, polarised and chiral beams / **42**

Probing thermally activated dynamics in Artificial Spin Ices with coherent X-rays

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Arrays of nanoscaled magnetic elements, each acting as a single mesospin, are the building blocks of artificial systems of varying complexity in which the mesospin and lattice geometry can be used to design emergent mesoscale magnetic order. The geometry of the mesospin lattice determines the magnetic dimensionality and the interactions between the elements affect the global ordering and thermally driven dynamics [1]. Here, we focus on two types mesospin arrays arranged as Ising chains and square artificial spin ice (SASI) structures [2,3]. Different mesospin gaps generate varying interaction energies which compete with the thermally active Fe/Pd base material to drive the collective behaviour . As a function of increasing temperature, individual mesospins start to reverse, introducing defects into the arrays and reducing the correlations over characteristic timescales. Direct imaging using PEEM is limited for fluctuating systems due to long acquisition times and a limited field of view, so here we use a different approach and combine coherent magnetic scattering with x-ray photon correlation spectroscopy (XPCS). We measure one-time and two-time correlation functions as a function of applied field and temperature. We particularly concentrate on the temperature window between the mesospin blocking temperature TB (fixed by the Zeeman and shape anisotropy) and the Curie temperature of the Fe/Pd base material. This study yields new insights into the dynamics of magnetic excitations in these arrays, with both high spatial and temporal resolution.

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XPCS, polarised and chiral beams / **59**

Piezoelectric response in PMN relaxor ferroelectric probed by in-situ wide-angle X-ray Photon Correlation Spectroscopy

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The development of advanced functional materials relies on understanding interactions and heterogeneity at nanometer-to-micrometer length scales. The extraordinary electromechanical properties of relaxor ferroelectrics are widely attributed to the crucial role of spatial structural heterogeneity. Recent developments in coherent x-ray sources and methods significantly advance the possibilities of nanoscale measurements, offering superb spatial and temporal resolution, and support also in-situ type experimental techniques. Wide-angle X-ray photon correlation spectroscopy (XPCS) is a powerful tool to probe dynamics of heterogeneity in condensed matter, both in equilibrium and under applied stimuli [1,2]. Here, we present an in-situ XPCS study of the relaxor ferroelectric PbMg1/3Nb2/3O3 (PMN) under applied AC electric field [3]. We observed strong periodic response in two-time correlation function (TTCF) calculated from the diffuse scattering speckle pattern, even for relatively weak applied AC fields. This is surprising since PMN is electrostrictive, with no linear piezoelectric response at zero field. The periodic behavior in the TTCF was shown to arise from local tilting of the illuminated sample volume due to the combined AC field and a static field caused by the incident X-ray beam. To qualitatively describe the results (tilt amplitude and direction) we developed a model that combines the electrostrictive response of the PMN material and the non-uniform charging due to the incident micrometre-scale X-ray beam. The X-ray-induced piezoresponse may play a crucial role in interpreting XPCS and nanodiffraction studies on other insulating materials subjected to applied AC fields or varying X-ray illumination.

Work supported by the US Department of Energy (DOE), Office of Science, Basic Energy Sciences, Materials Science and Engineering Division. The experiments were performed at beamline 8-ID-E, and also 12-ID-D and 33-BM of the Advanced Photon Source, a DOE Office of Science User Facility operated by Argonne National Laboratory under Contract No. DE-AC02-06CH11357. Work used resources at the Center for Nanoscale Materials, a DOE Office of Science User Facility, undersame contract. This research was also supported by the Natural Sciences and Engineering Research Council of Canada (NSERC, Discovery Grant No. RGPIN-2023-04416).

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Instrumentation and method development - S1 / **137**

Chair

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Instrumentation and method development - S1 / **6**

Photon correlation measurements using a MHz repetition rate X-ray free-electron laser

Author: Anders Madsen[None]

I will present recent advances in X-Ray Photon Correlation Spectroscopy (XPCS) at the MID instrument of the European XFEL. Access to sub-ms timescales is important for studying diffusion-type dynamics in aqueous solutions, for instance of colloids and bio-molecules. The European XFEL with its MHz repetition rate, provides this opportunity. XFEL experiments often require specialized sample replenishing and sample environments, for instance liquid jet delivery or fast scanning methods. The short-pulse duration and high peak brilliance of the source also gives unique possibilities for

investigating the initial states of crystallization, for instance in a super-cooled liquid. X-ray Cross-Correlation Analysis (XCCA) yields direct access to studying the crystalline order and emerging defects, beyond classical crystallography experiments.

Instrumentation and method development - S1 / 49

Rapid scanning ptychography with 3D real-time position registration at HXN beamline

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In the past decades, X-ray ptychography has been demonstrated as a powerful technique of coherent diffractive imaging because of its capability to achieve quantitative phase contrast and nanoscale resolution not limited by the performance of X-ray lenses. It has seen applications in a broad range of research fields from microelectronics to biology [1,2]. However, because ptychography is a scanning-based imaging method and requires high positioning precision of the instruments, the long measurement time and noticeable efforts needed in developing and operating the hardware have been two of the major factors limiting more general utilization of the technique. At the Hard X-ray Nanoprobe (HXN) beamline [3] of NSLS-II in the US, we have developed a system for ptychography and tomography imaging, which enables rapid scanning up to 1kHz speed and simplifies sample preparation and alignment process for more versatile applications.

The Rapid Scanning Microscopy Instrument-II (RASMI-II), being commissioned at the HXN beamline, is the main component of the imaging system which consists of motorized stages for sample and focusing optics. The sample stage uses a precision metrology disc and linear laser interferometers for position referencing, which enables continuous tracking of the sample position in 3D during scanning and sample rotation. This design largely simplifies the sample alignment procedure before measurement and tackles sample drifts during measurement to reduce scan overhead time. By combining the RASMI-II with a fast-response detector and high-speed data collection devices, we were able to push the scanning speed of ptychography measurements to 1kHz in fly-scan mode, which is the fastest frame rate of the detector, thereby achieving tomography measurement time down to 1 hour per sample. We have also developed specialized software and algorithms which incorporates fly-scan trajectories in the iterative phase retrieval process for faster convergence and noise reduction in rapid fly-scan ptychography reconstruction, as well as utilizes 3D position registration data for tomographic alignment and reconstruction.

In summary, the new ptychography and tomography imaging system at HXN beamline based on RASMI-II will provide a platform for high-throughput and versatile nanotomography imaging. Its instrument approach finds a broader potential application for other microscopy beamlines, particularly at the diffraction-limited light sources, where the scanning speed should be commensurate with significantly higher coherent flux.

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Instrumentation and method development - S1 / 77

Exploring memory effects in Quantum Materials

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Soft X-ray coherent techniques have proved to be a powerful tool in the investigation of static and dynamic phenomena in solid-state quantum materials, such as ultrafast light-induced phase transitions [1], fluctuating domains [2] or memory effects [3]. One particular aspect of phase transitions is their possibility of being non-reversible and/or stochastic, which ties into mostly unexplored underlying physical mechanisms and is critical when devising functional devices that exploit the nanoscale behaviour of a given system. Coherent Diffractive Imaging (CDI) and Fourier Transform Holography (FTH) can provide the spatial resolution, contrast mechanisms and sample environments required to tackle the nanoscale repeatability of a transition under a variety of conditions. Here we show a study on the repeatability of the light induced insulator-to-metal phase transition in the prototypical material VO_2, showing stochastic behaviour, unstable domains with different lifetimes and permanent metallic domains stable well below the critical temperature. In addition, we will present recent advances in the coherent imaging program of the BOREAS beamline at ALBA Synchrotron, highlighting imaging on magnetic Van der Waals materials.

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Instrumentation and method development - S1 / 61

Unlocking the potential of ptychographic imaging with upgraded coherent sources

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Recent breakthroughs in electron storage rings have enabled a notable increase in coherent X-ray flux, promising a potential acceleration in coherent imaging speed by several orders of magnitude. However, realizing this speed enhancement requires concurrent advancements in instrumentation and computation algorithms for X-ray ptychography. At the Advanced Photon Source, we have

developed high-speed ptychographic scanning that offers distinct advantages in handling decoherence effects, mitigating radiation damage, and providing capability of imaging large 3D volumes. With a prototype pixel area detector operating continuously at 20 kHz, a fast ptychographic scan was showcased at a remarkable scanning speed of 4 mm/s. Furthermore, we demonstrated high-speed ptychographic tomography on lithium nickel manganese cobalt oxide (NMC) samples with dimensions of approximately 10 microns, achieving completion within just a few minutes. These developments will unlock new possibilities for utilizing X-ray ptychography to expolre samples into the fourth dimension. This includes enabling the observation of chemical states across relevant energy edges and monitoring sample evolution in in-situ environments.

This work was performed at the Advanced Photon Source, a U.S. Department of Energy Office of Science User Facility under Contract No. DE-AC02-06CH11357.

Instrumentation and method development - S1 / 118

Next Generation Detectors for High-Resolution Experiments with High-Energy Coherent Synchrotron Radiation

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High-resolution synchrotron experiments increasingly require advanced detection capabilities to address the challenges of high-energy coherent synchrotron radiation. Hybrid Photon Counting (HPC) X-ray detectors have emerged as pivotal tools in this context [1].The EIGER2 detector advances HPC technology with its 75 µm × 75 µm pixel size, kilohertz frame rates, negligible dead time (100 ns), and count rates up to 107 photons per pixel. Particularly for experiments aiming to exploit coherent radiation, such as Ptychography, Bragg Coherent Diffraction Imaging (BCDI) or X-Ray Photon Correlation Spectroscopy (XPCS) these advantages are crucial.

Upgrades to 4th generation synchrotron sources will significantly enhance the brilliance and coherence of X-ray beams. This increase will be most dramatic in the high-energy regime, resulting in an effective increase of coherent radiation of 2-3 orders of magnitude at X-Ray energies >20 keV or higher – depending on the synchrotron source [2,3]. Coherent scattering techniques that are so far established for X Ray energies below 12 keV will also start to become relevant at energies >20 keV which will likely enable new and groundbreaking science.

HPC detectors with high-Z sensor materials are essential for enabling these experiments. They combine (a) noise-free detection with no incoherent noise contribution, (b) fast, digital readout for fast correlation times and scanning speeds, (c) high-dynamic range to resolve both weak and strong signals simultaneously with (d) high detection quantum efficiency (DQE). Particularly for experiments carried out at >12 keV where Silicon sensors become increasingly transparent, the high DQE of high-Z sensors not only reduces the needed acquisition time, but also drastically reduces the effective X-Ray dose on the sample.

We report on the EIGER2 detectors paired with the high-Z sensor CdTe in coherent experiments, demonstrating their performance, stability and reliability. We present experimental results for both XPCS and Ptychography at >20 keV, giving a glimpse on the potential of next generation HPC detectors for future experiments utilizing high-energy coherent radiation. The presented measurements were performed at several synchrotron sources: ESRF (Grenoble, France), APS (Chicago, USA), and PETRA III (Hamburg, Germany).

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Coherent Diffraction Imaging with X-rays / 138

Chair

Coherent Diffraction Imaging with X-rays / 123

Ultrafast Polaronic Lattice Distortions using Time-resolved Coherent Diffraction Imaging

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The recent advance of X-ray free electron laser (XFEL) opens the area of ultrafast structural dynamics with a few tens of femtoseconds time resolution due to its unique characteristics of X-rays. XFEL makes it possible to obtain critical information on the intermediate states or pathways during the phase transformation, which only measures the initial and final states with many existing techniques. In my talk, I show the results of ultrafast lattice distortions with photoexcitation of perovskite oxides by time-resolved Bragg coherent X-ray diffraction imaging by taking advantage of almost 100 % of transverse coherence available from XFEL. I will present a direct observation of initial polaron generation and relevant strain evolution in perovskite-oxide nanocrystals.
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Coherent Diffraction Imaging with X-rays / 5

Structured probes for BCDI: Toward the imaging of dynamic and distorted crystals

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This work proposes speckle Bragg coherent diffraction imaging (spBCDI), a new approach to improve the time-resolution in BCDI microscopy. The method uses structured (or speckled) illumination to induce a convolution of the 3D frequency content associated with the nano-sized crystal with a broad

kernel, acting partially along the rocking-curve (RC) direction. Such a convolution encodes 3D structural information about the sample in each 2D image, and hence significantly reduces the required oversampling ratio along the RC direction (i.e., with respect to the Nyquist frequency).

Extensive numerical simulations demonstrate data spBCDI obtained with low oversampling ratios (i.e., < 2) along the RC yield reliable reconstruction of the crystal phase. Such reductions in the oversampling ratio reduce the measurement times which are between 1.5 – 14 times (depending on the incidence angle and the speckle size) shorter than for standard (plane wave) BCDI [1], enabling spBCDI for imaging time-evolving systems in the 0.5 - 1s time scale at 4th generation synchrotrons. The simulations also explore the limits of spBCDI by evaluating the minimum oversampling ratio along the RC which still yields an invertible data set [1]. This new method is also remarkably well suited for robust imaging of strongly distorted crystals, which remains challenging in plane wave BCDI [1].

A proof-of-concept experiment has been performed at the ID13 beamline (ESRF) [2]. The structured illumination was obtained by introducing a tailored phase-plate, that we designed to match the required speckle and beam envelope sizes (manufactured by XRnanotech GmbH, Switzerland). Combined with the powerful focusing optics of ID13 (Multi-Laue lenses), this new optical device produces a structured illumination at the sample position, with speckle grains of 70 nm laterally distributed within a beam of ~ 1 µm size. This illumination was used to image a 3D crystalline nanostructured Si thin-film with a set of oversampling ratios along the RC direction ranging from 4 down to 0.4.

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Coherent Diffraction Imaging with X-rays / 85

Time-resolved Bragg coherent diffraction imaging of Pd nanoparticles during in situ acetylene hydrogenation

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The study of nanoparticle (NP) assisted chemical reactions is important for the development of efficient catalyst materials for a wide range of environmental and energy applications [1]. Such studies are primarily focused on the role of surface sites - whether on particle facets or vertices. The rate and efficiency of heterogeneous catalysis is dependent on these distinct adsorption energies and turnover rates [2]. In addition to facet-dependent catalytic activity, lattice strain is known to influence the reactivity of metal surfaces [3]. One technique that can be used to probe lattice displacement with pm precision is Bragg Coherent Diffraction Imaging (BCDI) [4]. From the lattice displacement, the local strain of metal NPs can be examined in situ during catalyst-enhanced reactions. In this study, we probe the hydrogenation of acetylene on Pd nanoparticles (NP) [5], where the partial hydrogenation to ethylene is desired. This relies on fine control of the sample temperature, gas pressure, and the reactant ratios [6].

In order to probe the kinetics of heterogeneous catalysis a dynamic probe of strain evolution is found using real-time, and stroboscopic BCDI at the ID01-EBS beamline of ESRF [7]. This is applied to Pd NP in situ during acetylene adsorption, hydrogen absorption, and under acetylene hydrogenation. With this study it is possible to track the evolution of the average lattice expansion of the particle as a function of time. By distinguishing the changes in local lattice parameter under each of these conditions, we can understand the role of various Pd facets in the partial hydrogenation of alkynes [5]. With this knowledge, we performed stroboscopic BCDI on Pd NP under reversible reaction conditions to observe the mechanism with a time resolution of 0.3 s. The evolution of the lattice parameter on the sub second timescale shows a clear facet dependence which we believe is linked to the preferential adsorption of acetylene. We observe that the top (111) facet exhibits a lattice contraction relative to the bulk of the Pd NP within a few seconds upon introduction of acetylene gas. Through DFT simulations we will unravel whether the rate-limiting step is pressure stabilisation or the surface reaction with absorbed hydrogen as has been previously proposed [6].

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Coherent Diffraction Imaging with X-rays / 79

Single-Shot Imaging and Phase Retrieval of Void-Shockwave Dynamics in Extreme Conditions

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The recent accomplishment of a 3.88 MJ yield in 2023 along with subsequent successes at the National Ignition Facility (NIF), indicates a transformative era in Inertial Fusion Energy (IFE) research. This milestone showcases nuclear fusion's potential as a sustainable, safe, and virtually inexhaustive source of energy, positioning it as a promising solution to meet the world's growing energy demands. Further improvements to IFE research are dependent on assessing material response during dynamic compression. To address this, our research team is pioneering the imaging of instabilities as fusion capsules collapse. This effort aims to quantify the impact of imperfections in ablator materials during dynamic compression and how it could influence the overall fusion yield. In an experiment conducted at the Matter in Extreme Conditions (MEC) instrument at the Linac Coherent Light Source (LCLS), we employed an x-ray phase-contrast imaging (XPCI) method to accurately measure the capsule's dynamic areal density [1, 2] during this process. To mitigate the effects of random fluctuations of the x-ray free electron laser (XFEL), we implemented a flat-field correction scheme that normalizes the dynamic images, compensating for both XFEL beam inhomogeneities and imperfections accumulated along the x-ray path [3]. For quantitative evaluation, particularly

for calculating the phase to determine areal density, we propose two distinct phase retrieval strategies tailored for extracting the phase from a complex, single-shot, multi-material, dynamic image. Our advanced techniques circumvent the typical limitations encountered with conventional phase retrieval methods. These traditional methods often necessitate a single-material composition, require the object to be isolated within the field of view, and are constrained to specific propagation regimes. In contrast, our approaches are not limited to these requirements and can determine the absolute phase, obviating the need for phase unwrapping. This feature is particularly beneficial since large phase excursions are induced by the compressive force of the shock wave in relation to the surrounding material. The first method involves using a single, flat-field corrected dynamic image for phase reconstruction. This method incorporates automatic differentiation (AD) [4, 5] with the transport of intensity equation (TIE), enabling the fine-tuning of specific parameters to best match experimental conditions and optimize the phase result. Further refinement of the reconstructed image is achieved by adopting the method of reconstructing the projected refractive index, a technique validated by Wittwer et al. [6] The second phase retrieval strategy leverages the inherent speckle pattern and phase contrast that naturally emerges during light propagation in the raw dynamic image. This approach avoids the conventional reliance on flat-field correction, but instead utilizes the unaltered data to uncover the phase information [7]. By using the speckle information, we reconstruct the large-scale structures, while propagation allows for the resolution of finer, small-scale features. These techniques represent a significant advancement in dynamic imaging, offering unprecedented clarity and detail in the visualization of material density. Acquiring areal density information is vital, providing insights into the complexities of achieve self-sustaining fusion reactions in current ICF experiments, has the potential to revolutionize experimental compression approaches, and can introduce additional physics to the computational models used in ICF research.

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Instrumentation and method development - S1 / 139

Chair

Instrumentation and method development - S1 / 22

Harnessing coherence in femtosecond fluorescence for photon diagnostics and future X-ray sources

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This talk will focus on coherence properties of femtosecond fluorescence produced by pumping the transition metal elements with X-ray free-electron laser (XFEL) pulses.

When the intensity of the incident beam is sufficiently low, the fluorescence will be incoherent light. The higher-order coherence of such incoherent light, especially intensity correlation between separated positions, provides rich information about the incident pulses, such as pulse duration and shape [1,2], as well as beam size on the target [3]. In the first half of my talk, I will discuss spatiotemporal diagnostics of XFEL pulses using fluorescence and their advantages over existing techniques while presenting the results obtained at SPring-8 Angstrom Compact free-electron LAser (SACLA) [4-6].

When the pump intensity is sufficiently strong to induce population inversion between inner-shell states (e.g., 1s and 2p states), the fluorescence photons become collective and exhibit directionality [7]. This coherent radiation, known as amplified spontaneous emission, can serve as the basis for an X-ray laser oscillator that generates fully coherent pulses by combining with cavity optics [8]. In the second half of my talk, I will talk about the coherence properties of amplified spontaneous emission. Based on the experimental results at SACLA and theoretical simulation, I will discuss the feasibility of an X-ray laser oscillator and requirement for X-ray optics.
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Instrumentation and method development - S1 / 83

Coupling X-ray photon correlation spectroscopy and dynamic coherent X-ray diffraction imaging using triangular aperture

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Coherent X-ray diffraction imaging (CXDI) is a powerful method for visualizing the structure of an object with a high spatial resolution that exceeds the performance of the lens. CXDI is of several types, based on the optical systems and reconstruction methods. Plane-wave CXDI, in which a coherent planar beam is incident on a sample, can be used to observe isolated objects. Nonetheless,

scanning CXDI, commonly known as ptychographic CXDI, is superior because it enables the observation of extended samples; however, its disadvantage is that improving its temporal resolution is challenging as it is based on multiframe data collection. Therefore, a method for reconstructing the image of an extended object using a single-frame diffraction intensity pattern must be established.

Previously, we proposed and demonstrated a practical method for single-frame CXDI in the hard X-ray regime [1, 2], in which a triangular aperture is used as a critical element in the optical system. The phase image of a selected field of view of an extended object was reconstructed from the single-frame diffraction intensity pattern based on a phase retrieval calculation. Furthermore, recently, we also proposed and demonstrated an approach to analyze particle motion in heterogeneous solutions over a wide spatiotemporal scale by combining XPCS and single-frame CXDI using triangular aperture [3]. By applying this approach to analyze of the dynamics of colloidal gold nanoparticles dispersed in aqueous polyvinyl alcohol solutions, we found that Brownian motion exists in the range of a few hundred nanometers, and two modes of motion exist in the micrometer range. In this presentation, the details of the CXDI method and the experimental results are presented.

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Instrumentation and method development - S1 / **74**

Sub-hour 3D ptychography at I13-1: Coherence, Diamond Light Source

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X-ray ptychographic tomography is now routinely used at synchrotron facilities around the world, producing nanoscale resolutions in 3D. However, the acquisition times still lag significantly behind other imaging methods. As many synchrotrons upgrade to diffraction limited rings, the community is being presented with a dramatic increase in the coherent flux available. However, translating that increase in flux into increased scientific throughput remains a challenge.

The latest advances in high-speed ptychography at I13-1: Coherence of the Diamond Light Source have combined a novel acquisition scheme with the latest detector technologies to achieve a ptychography collection rate of over 100 kHz. This is providing sub-second projections and sub-hour 3D ptychographic tomography. We present the latest technical developments as well as their applications in the fields of battery materials and brain imaging.

Instrumentation and method development - S1 / **43**

Development of 5.04 Mpixel CITIUS detector for high-resolution ptychography

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We have developed and deployed CITIUS detectors for the XFEL facility SACLA and the synchrotron radiation facility SPring-8. At SACLA, we are building a 20.2 Mpixel detector system that can run at a maximum frame rate of 5 kHz. The CITIUS detectors for synchrotron radiation applications were demonstrated to operate at the maximum frame rate of 26.1 kHz at the full-image readout condition and have a detection capability of 1 Gphoton/s/pixel at 10 keV while keeping the single photon sensitivity. These high performances are achieved by the new integration-type pixels without charge amplifiers; it eliminates the major analog power dissipation source, thus enabling camera heads to be compact even at the high frame rate operation. Feasibility demonstration to coherent diffraction experiments was reported previously for Bragg CDI at ESRF [1] and ptychography at SPring-8 [2] with 280k and 840k pixels, respectively, and reviewed in a commentary [3].
In this talk, we report the development status of a new 5.04 Mpixel CITIUS detector for high-resolution ptychography. The system will have a clock trigger distributor to suppress any false beat signal arising from the frame rate comparable to the revolving frequency [3]. The stream data rate from the sensors reaches 30.6 petabytes/day. We report our development on the data acquisition system with FPGA cards in the edge servers and the SPring-8 data center for on-the-fly data reduction. We also briefly address the development status of a high-speed analog digitizer, which synchronously operates with CITIUS, for recording incident X-ray intensities/positions and stage positions.

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XPCS and Polarised beams / **130**

Chair

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XPCS and Polarised beams / **124**

Exploiting the coherence of synchrotron soft x-rays to study the dynamics of quantum matter

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A challenging aspect of designing functional materials is to understand the impact of a wide range of characteristic spatial and temporal scales on stabilizing novel phases. Commonly used mean field approaches often provide reasonable estimates for static properties at macroscopic length scales. However, it does not provide the requisite fundamental insight into the important processes governing deviations from these averages where fluctuations play a critical role. Soft x-rays are a powerful element-specific probe to study such mesoscopic charge and spin textures. The coherence available

at current and newly upgraded light sources can enhance our established tools to give significantly more detailed information of otherwise difficult to probe quantum states. We select the coherent part of the x-ray beam to produce an interference pattern known as speckle. Here I will discuss how we use that speckle at the Advanced Light Source to look inside, and better understand the transitions in collective electronic systems such as those of orbital ordering, metal-to-insulator materials and amorphous helical magnets1,2.

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XPCS and Polarised beams / **115**

Insights to Collective Skyrmion Lattice Dynamics from Coherent Soft X-ray REXS

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Ordering of large length-scale magnetic structures are commonly studied using techniques like electron microscopy (EM), small angle neutron scattering (SANS), and resonant elastic x-ray scattering (REXS). Using coherent X-rays to investigate these systems, we can probe magnetic textures in ways that were once exclusive to either EM or SANS: domain dynamics, inter-mixed phases, long and short range ordering – all with a flexible probe size (<10um). As such, coherent REXS is an important tool to extend our fundamental understanding of magnetic skyrmions, which is essential if they are to be a platform for quantum information systems. Particularly, Cu2OSeO3 (CSO) is an interesting skyrmion host material because it is an insulator – a requirement for low energy consumption devices. We will present new findings associated with skyrmion lattice (SkL) dynamics in CSO from experiments that push the collective behavior into different excited states. The insights gained from coherent REXS help to better understand domain motion, which aids in completing a microscopic explanation for the SkL dynamics.

XPCS and Polarised beams / **50**

High-Speed XPCS Studies of Critical Fluctuations in Liquids

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High-speed XPCS using new coherent X-ray sources and fast detectors opens new avenues to explore fluctuation dynamics in fluids. We have been studying systems relevant to liquid-liquid extraction

processes, where an organic solution of extractant molecules is used to separate ions from an aqueous solution by formation of nanoscale molecular complexes. The organic phase exhibits incipient phase separation, and critical fluctuations play a key role in the structure of the molecular complexes [1-5]. Here we present XPCS studies of microsecond timescale fluctuations within 5 K of the critical temperature Tc, carried out at APS beamline 8ID. With the 500 times higher coherent X-ray flux that will become available from the APS Upgrade, it should be possible to observe fluctuation dynamics much further away from Tc. This will enable exploration of the crossover from Ising to mean-field behavior, as well as the changes in dynamics expected at the Widom line (the locus of fluctuation maxima extending from the critical point into the single-phase region).

Work supported by U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division, Separation Science Program, under Contract DE-AC02-06CH11357. This research used resources of beamlines 12-ID-C and 8-ID-I at the Advanced Photon Source, a DOE Office of Science User Facility operated by Argonne National Laboratory.

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XPCS and Polarised beams / **27**

Role of nanoscale heterogeneties on the metal to insulator transition in rare earth nickelates

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Rare earth nickelates RNiO3 display an insulator to metal transition (MIT) which is accompanied by a magnetic transition, charge ordering, and a crystal structure change from orthorhombic low temperature to monoclinic high temperature state. While this system has been widely studied, the nature of the fluctuation across the transition, and the associated time- and length-scales is not known. Spontaneous fluctuations are important components for stabilizing topological magnetic structures such as skyrmions in quantum materials. However, the dynamic susceptibility of the nickelates remains relatively unexplored, and the role played by nanoscale phenomena such as domain-wall formation and motion, local strain fields and phase separation in underlying pathways of MIT is not well- understood. In our work, we focus on understanding the role of nanoscale heterogeneities and their fluctuations in rare earth nickelates by employing x-ray photon correlation spectroscopy (XPCS), a technique requiring highly coherent x-rays. Our XPCS measurements on NdNiO3 thin films, show complex evolution of fluctuations dependent upon temperature and wavevector q. We also observe unexpected non-equilibrium dynamics and suggests a new approach to understanding these materials.

Ptychography at different wavelengths - S2 / **131**

Chair

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Ptychography at different wavelengths - S2 / 125

Electron Ptychography: An Emerging Imaging Technology for Physical and Biological Sciences

Coherent diffraction imaging (CDI) and ptychography [1] have been widely used in X-ray synchrotron sources. The advantage of ptychography over traditional CDI is that it does not need prior information about the probe function and overcomes some of the other issues of CDI, such as non-unique solutions and a limited field of view.
In electron microscopy, ptychography has also attracted considerable interest due to its potential to achieve super-resolution [1] without using aberration correctors. Unlike conventional imaging modes, the image-forming optics of ptychography replaced by computational methods (like a 'Digital Lens') using an array of electron diffractions collected by fast detectors (Left). New generation of direct detection cameras are particularly suited to ptychographic 4D data acquisition with new modes of operation, such as electron counting and fast acquisition. In this talk, I will review the current development and capabilities of this emerging imaging technology (electron ptychography) in my group for light atomic detecting [2], low dose imaging [3], 3D reconstruction [4-5], coupling to spectroscopy [6], EM field mapping, and cryogenic EM [7,8], which can then tackle characterization challenges across the physical and life sciences, ranging from ferroic and battery materials to biological macromolecules.
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Ptychography at different wavelengths - S2 / 101

Vibration-Corrected Electron Ptychography

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Far-field electron ptychography is considered to be one of the most powerful phase retrieval techniques currently available for electron microscopists. Recently it was shown that ptychography is capable to surpass the Abbe resolution limit [1] and resolve specimens features as fine as the blurring due to the vibrations of the atoms [2]. A rather simple mathematical model of coherent diffraction pattern formation standing behind conventional algorithms [1,2,3,4] does not account for any motion or entangled states [3,4]. In order to overcome the resolution limit caused by the lattice vibrations we utilize the mixed-object formalism initially proposed in 2013 for x-Rays [4], but never applied to electron microscopy data. In contrast to conventional ptychography [1,2] that treats a single transmission function of a specimen, mixed-object ptychographic reconstruction considers a sequence of entangled transmission function states, each producing a coherent diffraction pattern via a multislice simulation [2,3]. The total diffraction pattern corresponding to the entangled system is formed as an incoherent sum over the states [3,4].

For the tests of mixed-object ptychography we used a 4D-STEM dataset [6] of a monolayer MoS_2 acquired in the Nion Hermes microscope at an accelerating voltage, convergence semi-angle, scan step and electron dose of 60 kV, 33 mrad, 0.2 Å and 5.3e6 $e^-/^2$, respectively.

To evaluate the resolution of the ptychographic reconstructions we propose a novel approach, akin to the Young fringe resolution test widely applied in TEM imaging [7]. We perform two ptychographic reconstructions with different initial guess for the object, e.g. uniform prior. Then the two independent results are shifted with respect to each other and the Fourier intensity of the difference is computed. The arising interference fringes indicate the range of spatial frequencies identical in the two results, allowing to conclude how deterministic a particular reconstruction is. Further, in contrast to conventional resolution tests, e.g. visibility of the Fourier peaks [1,2], the proposed approach allows to include aperiodic features that are crucial for a moving specimen.

Assuming a single illumination mode [2,4] we conducted pairs of ptychographic reconstructions in two scenarios: one involving a single-state and the other involving 10 entangled states of the transmission function. After 500 iterations of the gradient-descent [3] the achieved resolution was estimated. As a result, two independent pure-state reconstructions appear to produce identical information up to 1.9 $^{-1}$, while the mixed-object reconstructions contain deterministic information up to 2.3 $^{-1}$.

Even with a monolayer specimen that is not supposed to produce a noticeable amount of incoherent scattering, we show that considering multiple entangled states of the transmission function makes the underlying model more realistic and improves the quality of the ptychographic fit. Thus, we liberate ptychography from atomic vibrations, its last known resolution limit [2,3].

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Ptychography at different wavelengths - S2 / 109

Using Tilted Multislice Electron Ptychography to Retrieve 3D Atomic Coordinates in 2D Materials

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Multislice ptychography has been used to characterize thicker samples in 3D dimensions with spatial resolution beyond the diffraction limit. When applied to the diffraction patterns collected by scanning transmission electron microscopes, multislice ptychography reconstructs the object as a series of slices with a lateral resolution of 10s of pm and a depth resolution of 2-3 nm. Further improvements to this technique could allow atomic resolution imaging in three dimensions. Compared to electron tomography, multislice electron ptychography views the sample from only a single direction, avoiding the complexities of tilt series and image alignment and allowing for larger samples to be studied. However, the depth resolution is limiting. When viewing bulk crystalline samples from a major zone axis, most of the atoms overlap in projection and are difficult to distinguish individually. Therefore, in this work, we apply multislice electron ptychography to twisted bilayer 2D materials, tilted to minimize overlap, allowing us to extract 3D atomic coordinates for most of the atoms in the field of view. Compared against the known structure of the samples, these coordinates have sub-angstrom precision, though there are systematic issues with the coordinates along the beam direction. We also perform simulations to explore the range of experimental parameters that enable this level of depth resolution and precision. These findings underline the potential of

multislice electron ptychography for determining atomic structure in 3D, even though challenges remain for extending this approach to thicker samples.

Ptychography at different wavelengths - S2 / 84

Near-field X-ray ptychography using a laser driven X-ray source

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X-ray ptychography (NFP) is a coherent imaging technique widely used at synchrotron facilities, due to the ability to retrieve quantitative phase information of extended objects at a micrometric image resolution [1]. The advent of novel bright sources, an alternative to large-scale facilities, is paving the way for the translation of coherent X-ray imaging techniques outside synchrotrons and free electron lasers [2].
The ELI Beamlines facility of the Extreme Light Infrastructure ERIC near Prague has recently commissioned a laser-driven plasma X-ray source (PXS), based on a 20 mJ, sub-20 femtosecond, 1 kHz laser interacting with a copper tape to generate copper K-alpha emission at 8 keV with sub-ps pulses. We present here a proof-of-concept for translating x-ray near field ptychography to laser-driven x-ray sources. In particular, we report the ELI PXS source characterization toward coherent diffraction imaging and the results of the first near field ptychographic imaging performed at a laser source. We discuss the results, limitations, perspectives and future developments.

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Novel microscopies / 132

Chair

Novel microscopies / 19

Intrinsic speckle-tracking for rapid retrieval of a sample's attenuation, phase shift, and diffusive dark-field images

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Speckle-based X-ray imaging (SBXI) [1, 2] is a technique that utilises random speckle modulations imprinted into an X-ray wavefront to retrieve multimodal sample information. Here, the term "multimodal" is used in the sense that SBXI can recover information regarding a sample's X-ray attenuation, refraction, and diffusion information – three complementary signals. Requiring only a piece of sandpaper in the experimental set-up as a mask, SBXI is an appealing technique for use in a broad range of applications. Furthermore, the signal-retrieving algorithm we have developed—Multimodal Intrinsic Speckle-Tracking (MIST) [3]—makes SXBI even more appealing as it is computationally rapid yet still capable of retrieving high-quality images of the sample. Transverse speckle shifts and speckle blurring are associated with the recovered phase-shift and diffuse dark-field (DDF) signals, respectively. MIST analyses these speckle changes by considering local energy conservation for each speckle in the SBXI regime, wherein, the Fokker-Planck equation for paraxial X-ray imaging [4, 5] is combined with the geometric flow formalism for SBXI [6]. There are various iterations of the MIST algorithm [3, 7, 8, 9], with each increasing in generality by reducing the number of sample requirements. In all the published MIST approaches, the multimodal inverse problem is solved by linearising the associated Fokker-Planck equation and deriving analytical or least-square solutions for the multimodal signals. Within this presentation, a general overview of the currently published MIST approaches will be provided. This will cover the mathematical techniques utilised in solving the Fokker-Planck inverse problem, as well as the underlying assumptions. Retrieved signals from various samples imaged using a synchrotron SBXI technique will be shown. The closing section of the presentation will discuss ongoing research avenues.

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Novel microscopies / **32**

Multibeam Ptychography up to 20keV: Opportunities and Challenges

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X-ray ptychography stands out as a robust phase-retrieval coherent imaging technique, well-suited for investigating samples with diverse scale structures. However, its scanning nature necessitates a delicate balance between achieving high resolution and accommodating a large field-of-view (FOV), considering factors such as scanning time, stage travel range, etc. Typically, the FOV and resolution ratio is between 100 and 1,000. For instance, the current high-resolution ptychographic scanning using high-speed multi-pass scanning mode can attain resolution in the tens of nanometers within a $3\mu m$ x $10\mu m$ region in 38 seconds [1]. Nevertheless, scanning over a hundred-μm FOV at such resolutions would extend the time required by a factor of 300. Within this timeframe, conducting ptychographic tomography on large-scale samples at high resolutions (<$50nm$) becomes impractical. A breakthrough addressing this compromise arises with the development of multibeam ptychography (MBP) [2], promising to transcend the current limitations of conventional ptychography and maximize photon utilization. MBP achieves this by employing multiple coded probes simultaneously using a 3D-printed lens array. The speed enhancement in MBP scales directly with the number of probes utilized. By overcoming the speed limitation, MBP offers the tantalizing prospect of conducting 3D nano-imaging on a large scale at high energy.
In our discussion, we will delve into the advancements in MBP development, including the maximum number of probes achieved and demonstrations with various sample systems. While the MBP method presents several opportunities, it also raises concerns regarding the comparisons with conventional ptychography and poses challenges that affect its performance in real-world experiments. Therefore, we will further discuss the quality of reconstruction comparison between single-beam and multi-beam ptychography, as well as how factors such as vibrations and photon statistics impact the data reconstruction quality.
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Novel microscopies / **45**

Incoherent diffractive imaging with spectral dispersion for oxidation-state sensitive structure determination

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Incoherent diffractive imaging (IDI) is a novel imaging technique which uses the transient coherence of X-ray fluorescence to image the structure of the emitting atoms to nanometer resolution. By employing second-order spatial intensity correlations akin to Hanbury Brown and Twiss's stellar intensity interferometry, IDI retrieves the spatial distribution of the underlying emitters, facilitating high-resolution characterization of heterogeneous nanoparticles with element specificity. We present recent results from our single-particle IDI experiments at European XFEL, showing for the first time the feasibility of 3D imaging of nanoparticles using IDI.
Building upon the concept of IDI, we introduce Spectral Incoherent Diffractive Imaging (SIDI), a novel method for achieving dark-field imaging of nanostructures with heterogeneous oxidation states. With SIDI, shifts in photoemission profiles can be spatially resolved, enabling the independent imaging of the underlying emitter distributions contributing to each spectral line. In the X-ray domain, this approach offers unique insights beyond the conventional combination of diffraction and X-ray Emission Spectroscopy (XES). Our proposed method opens avenues for time-resolved, element

specific and oxidation state-specific imaging of electron transfer in 3d-transition metal compounds or to study heterogeneous catalysts and battery materials where the nanoscale spatial distribution of elemental oxidation states are crucial for understanding function.

Novel microscopies / **73**

High-throughput hard X-ray projection imaging with a sub-5 nm resolution

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Knowledge of the structure of materials and biological samples at nanometer scales and over large volumes is essential to understand the mechanics behind their function. Coherent x-ray imaging is being developed to address this need. Far-field diffraction methods suffer from noise sensitivity and high dynamic range requirement of detector. Recently, with the rapid development of x-ray focusing elements, projection imaging provides an alternative solution with unique advantages over these limitations. Here, with our own manufactured multilayer Laue lenses, we demonstrated such a projection imaging modality at Petra III P11 beamline of DESY. An object placed just out of the focus forms a magnified hologram or projection image of the sample on a pixel-array detector. Magnifications of 30,000 or more can be obtained, meaning a 75 μm detector pixel maps to an image pixel of 2.5 nm. On the other hand, at an illumination numerical aperture of 0.014, a defocus distance of 100 um would give a field of view of 2.8 um, in single shot. This mode is particularly fast and efficient for phase-contrast imaging over a large field of view that can be easily "zoomed". Signals contained in the holograms are boosted thousands of times with an improved signal-to-noise ratio, which makes it robust and easier to retrieve the object information from the measurements. For robust phase retrieval of the holograms, we stepped the sample relative to the probe, which was the method of near-field ptychography. A 4 nm half-period resolution imaging of hierarchical nanoporous gold has been achieved at the energy of 17.4 keV. Furthermore, a systematical numerical study was carried out to quantitatively illustrate the advantages of projection imaging modality over far-field diffraction method in terms of noise robustness and dose efficiency.

Numerical methods / **133**

Chair

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Numerical methods / **63**

Deep learning 3D dynamical (4D) reconstructions from sparse projections

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The development of high-brilliance X-ray sources, such as the fourth-generation diffraction-limited storage rings and X-ray free-electron lasers, have opened up new possibilities for X-ray imaging. X-ray Multi-Projection Imaging (XMPI)1 is a novel technique that exploits the unique capabilities of the high-brilliance X-ray sources and enables volumetric information using single pulses. Unlike tomographic experiments which record projections by sample rotation, XMPI is a rotation-free technique that split the beam into beamlets and record simultaneously multiple projections from different angles. As a result, XMPI can acquire 3D movies (4D) at least three orders of magnitude faster than tomographic methods. However, it is extremely challenging to reconstruct 4D from highly sparse projections acquired by XMPI. Deep learning's advancement offers a potential solution to this problem. Current deep-learning implementations for X-ray imaging, on the other hand, face two major challenges. First, they usually work in a supervised manner, which requires paired training datasets. Second, the robustness and reliance of such methods is not guaranteed.

This presentation will discuss how deep-learning approaches can potentially address three-dimensional (3D) and four-dimensional (4D) reconstructions for XMPI experiments by exploiting the large amounts of data provided by high-brilliance X-ray sources. We will focus on ONIX2 and 4D-ONIX3, two self-supervised deep-learning methods that reconstruct 3D/4D information from sparse XMPI projections and enhance their reliance by including the physics of the image formation. ONIX is a novel 3D reconstruction method that learns the self-consistency of sparsely recorded radiographs using physics-based neural networks. It can retrieve volumetric information from less than ten projections at previously impossible quality levels without requiring any prior knowledge. 4D-ONIX is based on ONIX, it extends the capability of ONIX by including time as a 4th dimension and applying adversarial training to enforce consistency between the measurement and the reconstructions.

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Numerical methods / 111

Deep Learning and innovative experimental setup accelerate Ptychographic X-ray Computed Tomography for characterizing heterogeneous materials

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Ptychographic X-ray computed tomography (PXCT) is a high-resolution imaging technique that is widely used for characterizing various materials. It has two variants, namely far-field and near-field, which, when combined with spectroscopic techniques, provide new possibilities for material characterization. PXCT can be used to analyze mass density, localize chemical elements, inspect microstructure, and even map the magnetic field of a sample. Although the acquisition of data has become faster and the size of possible scanned volumes has increased, the dream of time-resolved analysis or hyperspectral nanoimaging still requires further acceleration of data acquisition. This challenge can be addressed in two ways: either by improving the instrumentation and the geometry of the experimental setup or by modifying the data processing strategy to work with less data while still maintaining high-resolution and quantitative contrast.
Some beamlines, such as cSAXS, PSI, CH, and SWING at SOLEIL Synchrotron in France, have already adopted the first method. They offer experimental setups allowing very fast acquisition and

with large beam sizes at the sample due to the long sample-to-detector distance. In this regard, we will introduce here the new French beamline, FAME-PIX, which has been in construction at the ESRF. This beamline will be dedicated to PXCT and spectro-ptycho, and it will employ an innovative scanning technology that enables quick sample scanning. We will showcase the applications of this technology by sharing the results we obtained at the SWING beamlines at SOLEIL.
Modifying the data analysis process is the second way to speed up the PXCT acquisition. We will be discussing an approach that uses Deep Learning networks based on MSDNET and TomoGAN to reduce the amount of data required by a factor of 4 or more without compromising the quality of the images. We ensure the accuracy of our results by implementing a robust refinement process that corrects any artifacts that may have been introduced by the Deep Learning networks. This approach can be applied to various tomographic techniques, and to facilitate the sharing of our neural networks with the community, we have created an AI tomographic hub called AIAX at the University of Grenoble Alpes in collaboration with other laboratories at Grenoble. The primary objective of AIAX is to assist the community in processing their data using our neural networks.

Numerical methods / **48**

Algorithmic and computational advances in holotomography

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Holotomography is a coherent imaging technique that provides three-dimensional reconstruction of a sample's complex refractive index by integrating holography principles with tomographic methods. This approach is optimal for the micro- and nano-tomography instruments at the newest generation of synchrotron sources. With the aim of facilitating the use of the optimal phase retrieval approaches, we present a family of novel algorithms wrapped in an efficient software implementation of X-ray holotomography reconstruction [1]. This development incorporates advanced iterative schemes for simultaneous object phase and beam illumination retrieval, facilitating high-fidelity reconstructions with enhanced accuracy. Notably, these schemes minimize the number of measurements, such as holography planes (distances) and tomography angles, required for accurate object reconstruction, thereby optimizing data acquisition efficiency. Basic holography and tomography operators are implemented using CUDA C-language functions, leveraging the computational power of GPUs for rapid processing. Iterative reconstruction schemes are written in CuPy, ensuring seamless integration into existing workflows. For interactive data exploration and analysis, the package is accessible through Jupyter Notebooks, well-suited for fast prototyping to advance the field of 3D phase contrast imaging. The efficacy of the software is demonstrated with experimental data from the Projection X-ray Microscope Instrument at the ID16A beamline of ESRF.

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Coherent Diffraction Imaging with electrons / **128**

Chair

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Coherent Diffraction Imaging with electrons / **60**

Self-coherence and the wave/particle duality of electrons

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Apart from being an indispensable tool in material sciences of outstanding spatiotemporal resolution that reaches towards 1 Å at 1 ps [1,2], electron microscopy is uniquely suited to study basic quantum mechanical aspects because the wave/particle duality of electrons can be seen as a window into quantum physics [3]. This contribution experimentally validates a model of electron self-interferences. They originate from coherent-inelastic scattering processes with energy loss $\Delta E = \hbar/2/\Delta t$ at the Heisenberg limit, which marks a boundary between the classical mechanics of particle physics and the quantum mechanics of wave functions. Thereby, wave packages of finite widths are created with 0.5 radian decoherence phase that are characterized by a self-coherence length ls. A method is described that allows measuring ls and thereby self-coherence by the extension of an evanescent field at sample/vacuum interfaces. It is caused by the localization of wave functions upon pulse-like interaction of single electron waves with matter. Further, self-interferences of single matter waves rather than ensemble interferences of many electrons determine the occurrence of lattice fringes in high resolution images if energy losses do not exceed ≈ 100 eV, where the ability is lost to coherently illuminate crystal unit cells. At larger energy losses the wave packages are comparable to the size of atoms and even their constituents and are perceived as particles. The description of electron scattering in a wave picture is mandatory because decoherence determines the measured self-coherence length but does not exist in particle views, which is why a collapse of wave functions upon detection is often postulated.
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Coherent Diffraction Imaging with electrons / **117**

Organic Crystals – TEM Imaging and Techniques Developed for Elucidating Polymorphism and Crystallization

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Crystallization is a well-established topic. It covers many life science fields, and its importance varies from understanding nanoparticle arrays to protein formations and drug engineering through molecular design. One of the most popular methods for direct imaging of crystal formation, defects, etc., is transmission electron microscopy (TEM), mainly used for inorganic materials since the electron dose is not as limiting as it is for the imaging of organic materials. While challenging, high-resolution electron microscopy of organic material is possible due to the recent revolutions of stable and sensitive detectors and microscopes. Real-space images of organic crystals and their aperiodic features obtained by a modern cryo-electron microscope potentially show the detailed structure. However, high-resolution details remain hidden because strong defocus conditions must be applied to produce contrast at a low electron dose. We used phase retrieval by focal series reconstruction (FSR) developed originally for inorganic samples. We have adapted the method for fast series recording of hundreds of frames within a few seconds on a direct-electron detector, all at low dose conditions

(at the order of 101-102 e-/Å2 per acquisition) in cryogenic temperatures. After the reconstruction of the exit-plane wavefunction, we could eventually retrieve focus and two-fold astigmatism aberration-fixed phase images of small organic crystals with a resolution up to the information limit of the microscope. Implying further techniques, such as density functional theory (DFT), ultimately allows for solving a crystal structure with high confidence based on the image in real-space.

Coherent Diffraction Imaging with electrons / 114

Electron Holography: A Technique for Phase Plate Optimization

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Electron microscopy is a powerful tool for imaging soft biological samples, providing superior resolution compared to both light and x-ray imaging techniques. Unfortunately, biological samples are weak-phase objects in the electron microscope, providing minimal inherent contrast [1, 2]. This is in part because our detectors are only sensitive to variations in electron intensity and not phase. There are different approaches to overcome these limitations, including heavy metal staining (mass-thickness contrast), defocus phase contrast, and phase plate induced phase contrast.

In this work we are developing thin film based phase plate devices, operating in Zernike and Hilbert mode respectively, to enhance phase contrast at minimal defocus. These phase plates introduce a phase shift of pi/2, or pi respectively, to the scattered beam. The phase shift produced by a thin film is governed by the mean inner potential of the film, the film thickness and the accelerating voltage of the electron microscope [3].

We utilize off-axis and in-line electron holography to characterize the phase shift induced by the thin film material of our devices [4, 5]. Employing the accelerating voltage, the known film thickness, and the measured phase shift, we can determine the mean inner potential for the material [6]. With the mean inner potential in hand we can now produce phase plate devices with the correct thickness to induce the target phase shift. With these devices we will be able to optimize the contrast from a variety of weak-phase objects yielding TEM images with both higher resolution and higher contrast.

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Ptychography at different wavelengths - S3 / 129

Chair

Ptychography at different wavelengths - S3 / 126

Vectorial ptychography: when polarization comes into play

Traditional ptychography usually assumes that the objects being imaged scatter light without changing its polarization state. However, when this assumption does not hold, we have shown the need for a more comprehensive vectorial approach to ptychography to accurately characterize such objects.

In this presentation, I will cover the fundamental concepts of light polarization and introduce the framework of vectorial ptychography. I will also delve into the experimental and numerical implementations of this technique. Several recent applications of this method will be presented for various challenging materials, including biominerals, engineered metasurfaces, and cholesteric liquid crystal films. Additionally, I will discuss the imaging of vectorial light, where polarization varies across the beam's cross-section.

Ptychography at different wavelengths - S3 / 95

Towards ptycho(tomo)graphy with a hyperspectral detector

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X-ray ptychography currently relies on monochromatic sample illumination, typically produced by selecting an x-ray beam energy with a monochromator to achieve the required temporal coherence. However, for broadband sources, this means that most of the flux is unused. Finding an alternative to monochromators to use the flux more efficiently would be transformative, in particular for laboratory based x-ray ptychography where every photon counts, and spectral filtering is therefore too costly in terms of flux.

A possibility is to obtain the required temporal coherence at the detection stage by using a detector sensitive to spectral information. The SLcam hyperspectral detector has such a capability and its high energy resolution (140 eV FWHM at 6.5 keV), makes it suitable for coherent diffractive imaging. Moreover, using the full broadband spectrum in hyperspectral ptychography enables elemental identification at high spatial resolution [1].

However, hyperspectral detectors are a relatively new technology, currently with an extremely low count rate (~5 photons/pixel/second for the SLcam. To overcome this limitation and maximize the efficiency of hyperspectral ptychography, we have developed two approaches:

1. Reconstruction method: By using "energy smart" ptychographic reconstruction algorithms, spatial information within one energy bin aids the reconstruction of spatial information in another energy bin.
2. Experimental method: The low count rate also increases the importance of "fairly" distributing diffraction intensities on the detector to make efficient use of all its pixels, minimizing acquisition

time. Zone plates are typically used for this purpose, but when used with a broader spectrum their energy dependent spot size in the sample plane introduces issues. One obvious way to spread detector illumination while still respecting the sampling condition is to use Kirkpatrick-Baez mirrors, but they are relatively inaccessible due to their cost and low ease of use. Here we explore an alternative solution that provides a trade-off between detector illumination spreading and sample spot size variation: A small diameter zone plate (a few tens of microns), where the allowed spectral bandwidth of the illumination is limited by the diameter of the zone plate. In our case we used a 20um zone plate (10-20 times smaller than usual) for a bandwidth of interest between 7.5 keV and 8.5 keV.

We present the optical design choices and latest results in applying these methods to battery materials and pushing towards hyperspectral ptychographic tomography.

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Ptychography at different wavelengths - S3 / 116

Imaging extended single crystal lattice distortion fields with multi-peak Bragg ptychography

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Recent advances in phase-retrieval-based x-ray imaging methods have demonstrated the ability to reconstruct 3D distortion vector fields within a nanocrystal by using coherent diffraction information from multiple crystal Bragg reflections [1-6]. However, these works do not provide a solution to the challenges encountered in imaging lattice distortions in crystals with significant defect content that result in phase wrapping. Moreover, these methods only apply to isolated crystals smaller than the x-ray illumination, and therefore cannot be used for imaging of distortions in extended crystals. In this work, we introduce Multi-Peak Bragg Ptychography which addresses both challenges via an optimization framework that combines stochastic gradient descent and phase unwrapping methods for robust image reconstruction of lattice distortions and defects in extended crystals. Our work uses modern automatic differentiation [7] toolsets so that the method is easy to extend to other settings and easy to implement in high-performance computers. This work is particularly timely given the broad interest in using the increased coherent flux in fourth-generation synchrotrons for innovative material research, and we comment on experimental prospects at these sources.

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Ptychography at different wavelengths - S3 / 25

Quantitative Imaging of Nanophotonic Structures in Butterfly Scales using X-ray Ptychography

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Butterfly scales are microscopic bio-composites made of chitin embedded in a protein matrix, often along with pigments. A common structure consists of a laminar thin film (180-200 nm thick) connected to a sculpted upper lamina via pillar-like trabeculae. The upper lamina is made of longitudinal ridges connected by cross-ribs [1]. The cuticle of these scales produces colour by incorporating pigments and by reflecting light off its nano-structured surfaces with different refractive indices. It remains unknown, however, how the distribution of pigments within a scale correlates with cuticle mass density variations to impact the refractive index and resultant colour.

X-ray ptychography [2,3] is a lens-less quantitative imaging technique with high spatial resolution that can be combined with tomography [4] and/or spectral mapping [5] to study the three-dimensional structure and chemical composition of amorphous materials.

With data collected at the I13-1 instrument at the Diamond Light Source, we successfully used ptychographic X-ray computed tomography (PXCT) to determine the three-dimensional mass density and morphological variations of two pairs of scales with pigmentation differences in two species of nymphalid butterflies, *Junonia orithya* and *Bicyclus anynana*. By comparing densities with colour profiles, we determined that the lower lamina in all scales has the highest mass density, with density being inversely correlated with pigmentation within each species [6]. Furthermore, with recent spectro-ptychographic data collected at the I08-1 soft X-ray ptychography instrument at the Diamond Light Source across the carbon, oxygen, and nitrogen L3 edges, we explored the relative amount of chitin ($C_8H_{13}O_5N$) and melanin ($C_{18}H_{10}O_2N_2$) making up the photonic nanostructure of *Junonia orithya* and *Bicyclus anynana* butterfly scales.

Butterfly scales are extensions of single cells and how individual cells control the development of these precisely nano-structured materials is a growing field of study, with applications in future bio-engineered systems. Though the presence of pigments and chitin in butterfly scales has been known for decades, quantification of the spatial distribution of these materials has been a challenging task due to the nanometre length scales of these structures and lack of the technology to investigate it. With the help of quantitative coherent imaging techniques such as ptychography, we hope to make headway in answering these questions.

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Ptychography at different wavelengths - S3 / 37

Coherent Surface Scattering Imaging with Nanometer Resolution for 3D Mesoscale Structures at Surfaces and Interfaces

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Many nano and quantum devices have intricate, low-dimensional, non-uniform, or hierarchical structures, spanning from millimeters down to sub-nanometer sizes, on surfaces and interfaces, making high-resolution surface-sensitive characterizations imperative for understanding their function-structure relationship. This often entails visualizing surface-supported and buried planar mesoscale structures, which can be done non-destructively by high-resolution X-ray imaging and scattering techniques. Newly developed Coherent Surface Scattering Imaging (CSSI), which operates in grazing-incidence reflection geometry [1], effectively tackles the aforementioned challenges. We will present several recent advancements to illustrate the capabilities of different imaging techniques within the framework of CSSI. For example, hard X-ray ptychographic reflectometry imaging merges the two-dimensional imaging capabilities of hard X-ray ptychography with the depth profiling capabilities of X-ray reflectivity, for surface and interfacial structures [2]. The amplitude and phase information obtained from ptychography reconstructions at various reflected angles not only reveals surface topography and localized structures such as shapes and electron densities but also provides statistical details such as interfacial roughness. Moreover, employing advanced 3D finite-element-based multibeam-scattering analysis enables the extraction of heterogeneous electric-field distributions and high-resolution 3D mesoscopic surface structures [3]. This holography imaging method holds great potential for single-shot structural metrology, allowing visualization of irreversible and morphology-transforming physical and chemical processes in situ or operando. Additionally, we will introduce the unique CSSI beamline currently under construction for APS Upgrade [4]. Achieving spatial resolution in the order of a few nanometers in all three dimensions is made possible by state-of-the-art wavefront-preserving X-ray optics for 2-D focusing, high-precision motion-decoupled nano-positioning stages for surface alignment and scanning, and a cutting-edge vacuum detector system ensuring sufficient imaging oversampling in a reflection geometry. We will also discuss the potential for further development of CSSI.

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122

Direct Phase Retrieval with Bragg Peak Gradients in Macromolecular Crystallography

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Abstract:
We present a novel algorithmic approach to solve the phase problem in macromolecular crystallography directly. The information captured by Bragg peaks - as used by conventional methods - is not sufficient to reconstruct the original image unambiguously. In fact, Sayre showed that at least twice as many data points would be needed to reconstruct a crystal from intensity alone [1]. To overcome this limitation, we present an approach that uses off peak information.
Our approach involves computing the effective intensity gradients on every Bragg peak giving us one additional data point per dimension d which potentially increases the constraint ratio by a factor of 1+d. These extra datapoints are then introduced as an additional Fourier/data constraint based on work by Elser [2].
We have successfully validated this methodology through simulations in both 2 and 3 dimensions and we are currently working on its application to real data.

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121

Coherent X-ray Imaging data analysis using PyNX

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4th generation synchrotron sources provide two orders of magnitude more coherent photons, and thus the ability to collect coherent X-ray imaging datasets faster and/or with a higher resolution. Consequently, the increased volume of data requires dedicated tools to fully take advantage of the improved coherent flux.

PyNX[1,2,3] is developed at ESRF - it has been written from the ground up to provide state-of-the-art performance using optimised GPU programming, both in terms of speed and memory requirements (to handle larger datasets). It is used on multiple beamlines notably at ESRF, Soleil, Petra-III, TPS, with scripts for data analysis easily expandable for new instruments (only the data input functions need to be updated). Input/output using the standard CXI format is also supported.

We will present the applications to various experimental techniques: Coherent Diffraction Imaging (CDI) and Ptychography (far field and near field) for two and three-dimensional imaging, also in the Bragg geometry to provide strain information in nano-crystals, and finally holo-tomography (currently in development).

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119

Cdiutils: A Python Package for Bragg Coherent X-ray Diffraction Imaging Processing, Analysis and Visualisatio

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Bragg Coherent Diffraction X-ray Imaging (BCDI) is a non-invasive X-ray characterisation technique for probing in three-dimensions (3D) structures of single nano-objects. While traditional BCDI analysis yields 3D maps of electron density, displacement, and heterogeneous strain, this work introduces a methodology and a user-friendly open-source tool for resolving 3D d-spacing maps within the examined object.

In BCDI, the primary output of phase retrieval algorithms is a 3D map of phase with a spatial resolution of approximately 10 nm. However, analysing BCDI data goes beyond this, and essentially revolves around deriving this phase into more physics related aspects such as atomic displacement and strain. To address potential ambiguities and differences in strain definitions across different scientific communities, we introduce the concepts of heterogeneous and homogeneous strains. Heterogeneous strain aligns with traditional BCDI strain analysis, while homogeneous strain corresponds to the shift of the Bragg peak in reciprocal space, a fundamental quantity in X-ray analysis under Bragg conditions. To combine both types of strain information, we present a methodology for computing 3D maps of local d-spacing, an absolute and more fundamental quantity than traditional heterogeneous strain maps. Expanding on this concept allows for the generation of 3D maps of global strain, a quantity analogous to d-spacing.

Cdiutils [1] is a python package designed to facilitate pre- and post-processing stages of BCDI data. It meticulously manages the cropping of the Fourier window prior to phasing, with a particular focus on handling the centre of mass of the 3D Bragg peak intensity relative to the cropped Fourier window. This feature enables the subsequent post-processing of the phase ramp, strain shift, and particularly, d-spacing maps. When coupled with PyNX software [2] for the phasing stage, cdiutils integrates all the steps of the entire data analysis process, including pre processing, phasing and post-processing, all within a single user-friendly notebook. Multiple methods for heterogeneous strain computation are routinely employed and compared to ensure robust data analysis. Cdiutils runs on ESRF machine environment, transitions between CPUs and GPUs when required, can handle several data formats and beamline geometries and is easily adaptable to any other machine environments and beamline geometries.

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113

A new soft x-ray coherent imaging branch line at MAX IV's SoftiMAX

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The SoftiMAX beamline at MAX IV is currently installing and commissioning a new brancline dedicated to coherent methods. The new CXI branch is is equipped with a KB focusing system to create a 20-micrometer focus, making it suitable for methods such as XPCS, FTH, and CDI. The branch can be used as an open port for user end-stations, but an internal end-station is currently under assembly.

The main feature of the CXI end-station is the sample and detector can rotate in the horizontal plane, and a large detector distance of up to 2 meters from the sample. In this contribution, we discuss the design of the branch line and end-station, provide a status update, share commissioning results, and outline the scientific program ahead. The CXI branch promises exciting capabilities for coherence method development and complex materials research.

90

High-energy X-ray Photon Correlation Spectroscopy to probe the dynamics slowing down in compressed metallic glasses

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Modern 4th generation synchrotron-radiation sources like the ESRF Extremely Brilliant Source are characterized by large transverse coherence lengths and high-flux coherent X-ray beams thanks to the jump in source brilliance [1]. X-ray Photon Correlation Spectroscopy (XPCS) retrieves the dynamics in condensed system via the intensity-intensity temporal correlation of the speckle fluctuations and largely benefits from the brilliance gain as the coherent flux scales with the brilliance and the smallest accessible sampling time has a quadratic dependence on the brilliance gain for the same signal-to-noise ratio (SNR). One major new opportunity is the availability of intense X-ray beams with a sufficient degree of coherence at high energies above 20 keV. We show that when combined with modern two-dimensional detectors this makes possible to apply XPCS to probe atomic dynamics in compressed matter down to the ms timescale with high SNR. In particular, we demonstrate that XPCS is feasible at high pressures in diamond anvil cells [2,3]. Applying pressure on a metallic glasses (MG) leads to dynamical effects as peculiar aging behaviors differing from common wisdom. We also show that when combined with temperature protocols it is possible to tune the ergodicity level of the glass out-of-equilibrium state and drive the system into different polyamorphic states persisting in the re-heated equilibrium (super-cooled) liquid [4].

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20

Coherent Imaging of Incoherent Dynamics

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Coherent X-ray imaging is often used to image nanoscale systems with high spatial resolution and is increasingly being applied to studying dynamical processes [1,2]. However, many materials show stochastic dynamics at the nanoscale, effects like fluctuating domain structures or the motion of charge carriers [3]. While single-shot imaging could in principle directly return images of these effects, damage thresholds from intense X-rays usually limit us to acquiring many exposures in order to construct an image, and so the stochastic components are blurred out. Recent advances in tracking correlations between partial frames relax these limitations [4], but there remain many cases in which tracking individual fluctuations will remain infeasible.
In this work, we theoretically and numerically demonstrate a method to separate the contribution of stochastic processes from the static part in a coherent diffraction pattern by using the principles of Fourier transform holography. This allows to directly recover the statistical properties of the fluctuations, independent of other deterministic dynamics. For a few special cases, we can also retrieve both the amplitude and phase information by applying iterative phase reconstruction algorithms on the retrieved momentum spectrum of the fluctuations. Our versatile approach will enable the direct recovery of the spatial, spectral, and temporal properties of stochastic material dynamics in a wide variety of systems currently unobtainable with existing methods.

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33

Single shot imaging with randomized structured illumination at a free electron laser

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Single-shot full-field imaging is a major application of Free Electron Lasers (FELs), able to study the spatial structure of non-repeatable dynamics on the femtosecond timescale. However, the methods currently in use at FELs either require a mask to be deposited on the sample (coherent diffractive imaging and off-axis holography) or require a relationship between the amplitude and phase contrast of the sample to be known a priori (single-frame in-line holography). We report on our implementation of a new diffractive imaging method called Randomized Probe Imaging (RPI) at the FERMI FEL. RPI enables reliable, scalable, and mask-free quantitative imaging using a simple experimental design. This implementation achieved single-shot reconstructions with a full-pitch resolution of 400 nm across a space-bandwidth product in excess of 20,000. We will outline the prospects for improvement in both resolution and space-bandwidth product, and discuss possible applications in fields including nanomagnetism, the physics of shock wave propagation, and the dynamics of collective electronic phases.

The results we will present are based upon work supported by the Department of Energy, Office of Science, Office of Basic Energy Sciences, under Award Number DE-SC0021939. K.K. was supported by the German Research Foundation (DFG) under project number 428809035.

18

NSLS-II's up-coming coherent diffractive imaging (CDI) beamline

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Coherent diffractive imaging continues to produce unique insights into the structure and dynamics of materials. At NSLS-II, we are nearing completion of a new beamline designed to provide novel controlled x-ray illumination, sample environments, and experimental geometries. The design considerations, beamline optical system simulations, endstation provisions, and progress toward its completion in 2025 will be presented here.

The CDI beamline's source will be provided by an 18-mm-period in-vacuum undulator and the 3 GeV electrons from NSLS-II. The undulator will feature a variable taper, delivering an increased x-ray bandwidth of at least 5% RMS at 10 keV. The optical system will use two bendable x-ray mirrors in conjunction with two fixed-figure mirrors to provide a sample illumination that allows variable coherence properties in a "zoomable" x-ray focal spot of about 1 to 10 microns in lateral size. Thus, the optical design provides a unique opportunity to tailor beam properties to the needs of any particular coherent imaging experiment. The final optics will provide a very long working distance of approx. 1.5 m and the sample-to-detector distance will be variable from 0.5 m to 10 m. Two area detectors will be independently positionable, allowing for simultaneous measurements in either or both of the forward-scattering and a Bragg-reflection geometry. The angular coverage of the detector system varies from approx. 70(V) x 120(H) to 11(V) x 120(H) degrees as a function of sample-to-detector distance.

The CDI beamline will present an exciting capability for routine, high-stability coherent imaging measurements and a uniquely-capable test-bed for the development and refinement of future imaging methods. We are currently on-track to commission this beamline in Summer 2025.

58

Intelligent Parameter Tuning for Ptychography using Physics-informed Bayesian optimization and Large Language Models

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Ptychography has become a standard technique in X-ray and electron microscopy and enabled numerous applications from integrated circuits to biological molecules. Despite of its tremendous success, obtaining accurate reconstructions remain nontrivial as it requires optimization of numerous types of experimental and algorithmic parameters that are highly data-dependent and often selected by trial-and-error in practice. To address this issue, we have developed an automatic parameter tuning workflow for ptychography using Bayesian optimization with Gaussian processes (BO-GP). With minimal assumptions about sample and data, our workflow consistently produces high-resolution reconstructions similar or even superior to those processed by human experts. The technique also enables researchers to efficiently search for the most ideal experimental conditions and probe structures at low dose levels. In this talk, I will discuss our ongoing research that further improves the workflow's efficiency with additional prior knowledge. We first carried out data analysis on various simulated datasets to obtain comprehensive relationships between key parameters and reconstruction quality. The trends were modeled by custom decision boundaries that can be readily integrated

with BO-GP as constraints to reduce the search space. What's more, we developed customized large language models that can interact with beamline users and suggest tailored parameters based on physical principles or instrument-specific knowledge.

70

Phase retrieval for Nuclear Resonance Scattering

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Mössbauer spectroscopy delves into atomic-level properties of an object by examining how atomic nuclei interact with their magnetic and chemical surroundings. With the advent of advanced synchrotron sources, a time-domain version of this technique, called nuclear resonant scattering (NRS), has emerged - which relies on the recoil-less resonant scattering of synchrotron radiation by Mössbauer nuclei. NRS detects the nuclear excitations as beat patterns in time, serving as a tool for material science and studying light-matter interactions.

Time-domain NRS offers advantages over traditional energy-domain Mössbauer spectroscopy, including better resolution, sensitivity, and shorter measurement times - thanks to the superior focus, coherence, and brilliance of synchrotron radiation compared to lab-based sources. However, analyzing time-domain NRS data is complex, prompting a focus on developing synchrotron Mössbauer sources for energy-domain measurements. However, these sources are difficult to fabricate, stabilize and replicate. Instead, we propose a computational approach inspired by Ptychography, a scanning imaging technique, to extract the scattering magnitude and phase in the energy domain from time-domain measurements using their Fourier relationship. This is a one-dimensional phase retrieval problem. Numerical tests and proof-of-concept experiments on samples containing $^{57}\mathbf{Fe}$ nuclei demonstrate the robustness of our approach, highlighting its potential for future research with modern X-ray sources and other Mössbauer isotopes.

44

Multi-beam ptychography with stacked Fresnel zone plates

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Multi-beam X-ray ptychography enables faster imaging of larger areas than standard ptychography and has been demonstrated with refractive, reflective, and diffractive optics. Here we present a novel implementation with stacked Fresnel zone plates. The optics are not fixed and moving them apart generates separate probes. The resulting probes are close in space (just a few microns apart) and are

coherent with themselves and (partially) with each other. This gives us a framework for studying interference effects on probe and object reconstructions in multi-beam ptychography. By treating the separate probes as incoherent modes that are separate in space, we are able to obtain successful reconstructions. This regime of operation is new to the application of multi-beam X-ray ptychography, and the flexibility of our method holds promises for boosting synchrotron experiments and saving beamtime.

41

Kinetic viscoelasticity during early polymer-polymer spinodal dewetting

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The dewetting kinetics of a supported polymer bilayer were measured in-situ using coherent grazing incidence x-ray scattering. X-ray photon correlation spectroscopy (XPCS) provides both the two-time correlation functions and the cross-correlation function which measures the average spatial shift of the speckles produced by the coherent x-rays. The stress in the ultra-thin dewetting top film can be directly observed due to exquisite sensitivity to sample curvature changes provided by the x-ray speckle correlation functions. The hole opening events in the film are found to be associated with significant changes to the stress. These results are interpreted through an analogy between viscoelastic spinodal dewetting and early-stage bulk viscoelastic phase separation. The frequency of hole initiation events during dewetting decreases with time as a power law, the power-law exponent can be linked to non-linear viscoelastic effects; showing similarity in their stress relief dynamics to aftershock decays.

We will briefly present data on dewetting velocity from holes and late stage coarsening as well.

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80

Understanding the dynamical heterogeneity near the order-disorder transition of spin-helices in amorphous Fe-Ge thin-films

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Speckle patterns manifesting from the interaction of coherent X-rays with matter provides glimpse into the dynamics of nanoscale domains that underpin many emergent phenomena in quantum materials and 2D systems. While the average structure in these materials can be followed with time-resolved X-ray diffraction, the dynamical heterogeneity as a function of time and length-scale near phase transitions have thus far eluded detection due to several experimental limitations and insufficient coherent flux. In this talk, I will present about the work done using coherent resonant X-ray scattering technique at the Advanced Light Source (ALS) where we observed that nanoscale spin fluctuations deep inside the ordered states of amorphous Fe-Ge thin-films can characterize phase transitions, independent of the underlying magnetic interactions. The results showed that the origin of the helical-to-paramagnetic phase transition lies in the appearance of enhanced fluctuation, or "hot spots," deep within the ordered regions. Our analysis method simultaneously extracts the time and ensemble averaged fluctuations of spin-helices which in turn predicts the nature of dynamics, whether ergodic or not in this system. Controlling and tailoring the fluctuating fraction can provide a new direction to predicting and understanding phase transitions in materials relevant to high-density, energy-efficient microelectronics.

35

Visualizing catalysts at the atomic-scale

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Electron microscopy has become a powerful tool for visualizing matter at high spatial and temporal resolution [1-5]. While the ultimate imaging resolution is no longer limited by instrumental performances, the main challenge is nowadays beam-sample interactions that intricately modulate atomic structures by dynamic processes induced by the electron beam [6-9]. Hence, it is mandatory to exercise control over the electron illumination to extract chemically relevant observations. In this contribution, we outline atomic-resolution imaging concepts for suppressing electron beam-induced sample alterations and illustrate their applications for visualizing heterogeneous catalysts in their working state.

Specifically, we focus on bright-field transmission electron microscopy to detect atoms with the fewest scattered electrons as well as restoration of the electron exit-wave function to achieve the most informative structural fingerprint [10]. Imaging is conducted under low-dose-rate illumination to ensure only induce weak object excitations [6-9] and include, importantly, coherent-inelastic scattering events in the single-electron-limit [11]. By analyzing the electron exit-wave function within channeling theory, we take advantage of the full exit-wave shape of the atomic columns to account for the three-dimensional atomic structure at single-atom sensitivity as well as for its vibrational excitation [10]. These new imaging concepts are generally applicable to in situ and operando electron microscopy experimentation and open up for unprecedented new views on the dynamic behavoir and functionality of catalysts under chemical relevant environments [1-4].

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86

A computational framework for BCDI experiment

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Synchrotron radiation (SR) have developed from the 3rd to 4th generation, and the electron emittance was reduced to enhance the brilliance and transverse coherence of the emitted X-ray beam. Benefiting from this, coherent scattering techniques could have new possibilities, such as better spatial resolution for Bragg coherent diffraction imaging (BCDI). To help design and perform BCDI experiment, we will present here a complete framework of BCDI experiment, which includes three modules covering from the pre-experiment analysis to the final data treatment. Firstly, module 1 aims to provide help for performing the BCDI experiment design and pre-experiment analysis of the sample. With the updated CAT (coherence analysis toolbox) software, source-to-end (S2E) simulation from coherence beamline to BCDI experiment could be performed taking advantage of the highly efficient beamline model construction and coherent modes propagation. Secondly, module 2 contains functions helping to perform the BCDI experiment, including nanoparticle locating (scanning X-ray diffraction map), and center-of-rotation alignment. Thirdly, module 3 is pyCXIM (python-scripted for coherence X-ray imaging methods) software developed for the BCDI data treatment. It includes functions to generate three-dimensional reciprocal space maps, GPU-based 3D phase retrieval, and post-process data treatment. Based on these three modules, we developed a novel experimental method aiming to assist the convergence of phase-retrieval process for BCDI experiment. This method will be applied in hard X-ray coherent scattering beamline of High Energy Photon Source (HEPS), which is a new 4th generation SR source under construction.

108

Direct probing relaxation dynamics in glass forming liquid by XFEL-based X-ray photon correlation spectroscopy

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Structural relaxation processes in glass forming liquids are important for many materials properties. However, atomic rearrangements underlying the relaxations have not been well understood due to the experimental difficulties in characterizing the disordered systems. Here we demonstrate an experimental concept of probing the relaxation dynamics in a model system $Ge_{15}Te_{85}$ by employing coherent X-ray pulses produced by X-ray free electron lasers (XFEL). By collecting the summed speckle patterns from two rapidly successive, nearly identical X-ray pulses generated using a split-delay system, we can extract the contrast decay of speckle patterns originating from sample dynamics and observe the full decorrelation of local order on the sub-picosecond timescale. This provides the direct atomic-level evidence of the highly fragile liquid behavior of $Ge_{15}Te_{85}$ above its dynamic crossover. Our methodology can be applied to a variety of glass forming liquids, opening a new avenue of direct experimental studies of relaxation dynamics in the ultrafast regimes.

96

Using single-shot phase retrieval neural network for the prediction of ptychography probe positions directly from diffraction patterns

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Using single-shot phase retrieval neural network for the prediction of ptychography probe positions directly from diffraction patterns
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Introduction

Probe position errors are a major source of image distortions, artifacts or reconstruction failures in ptychographic imaging. Many existing ptychography reconstruction frameworks correct probe position errors using optimization-based methods, where probe positions are updated with either the gradient of the main reconstruction loss function [1], or an update vector solved through linearization [2]. While having demonstrated their success in practice, optimization-based methods can either fail or take many iterations to converge when the error accumulates along the scan path, or has very large magnitudes.

We describe the development of a deep learning (DL) based method that targets the problem of correcting large and accumulating probe position errors using a single-shot phase retrieval neural network. The network predicts the phase in the local region of each scan point which are then used to calculate the positional offsets at all scan points. We show that our method brings accumulating

position errors on the order of 10^2 pixels down to a few pixels, which can then be easily corrected by optimization-based algorithms.

Methodology

The core of our method is built on a previously published neural network named PtychoNN [3] which, once properly trained, can predict the phase of a local area given just one diffraction pattern. Our approach uses PtychoNN to make predictions on every diffraction pattern collected. We then use common image registration methods to find out the pairwise positional offsets among the predicted images associated with all scan points. After that, we solve a series of equations in the form of $r_j - r_i = d_{i/j}$, where r_i and r_j are the positions of scan point i and j, and $d_{i/j}$ is the offset between them found by image registration. These equations are collectively solved as a linear problem $Ax = b$, through which we can obtain a least-square solution of all scan points' positions simultaneously.

Results

For the most extreme test case used in our experiment, where data were collected with a nominal step size of 100 nm and the deviation distance from nominal positions is 800 nm (or 100 pixels with a prediction pixel size of 8 nm) on average and 1800 nm at maximum, our method was able to predict the positions with a mean error distance of 24 nm or 8 pixels. If the positions found by our method are used as the initial guess for optimization-based position correction algorithms, they can precisely correct the remaining errors; otherwise, these algorithms would fail completely. For smaller or non-accumulating errors, we also show that our predicted positions can accelerate the convergence of both position errors and reconstruction loss. Additionally, our method is robust to small training set size: the averaged position prediction error over all the test cases remained stable when the training set was shrunk by half.

Conclusion

We demonstrated a novel ptychography probe position prediction method based on single-shot phase retrieval neural network, which can bring down the probe position error to a few pixels from over 10^2 pixels' large accumulating errors. Our method can work synergistically with optimization-based position correction methods used in common ptychography phase retrieval frameworks, enabling the accurate reconstruction of ptychography data collected without advanced stage positioning instruments.

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12

Transport Coefficient Approach for Characterizing Non-equilibrium Dynamics in Soft Matter

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With a substantial increase in beam coherence from the upgrade of the 4th generation synchrotron, X-ray Photon Correlation Spectroscopy (XPCS) now processes the capabilities to explore previously inaccessible aspects of structure and dynamics, including examining cooperativity and heterogeneity under external stimuli. However, the existing theoretical framework for analysis has not kept pace with the improved temporal and spatial resolution achievable through the upgrade, impeding scientists from fully comprehending subtle dynamical changes over time, consequently limiting the scientific productivity and beamline performance. In response to this challenge, we have developed an innovative method to integrate the collective influence of internal and external forces acting on the system within the framework of Markov chain and introduce a universal parameter, transport coefficient, to characterize dynamics over time. This method is verified using molecular dynamics (MD) simulated colloidal system subjected to temperature changes and a complex fluid under experimental conditions known for their non-equilibrium characteristics. The results reveal detailed dynamical information in non-equilibrium states and align with previous observation while providing enhanced vision of the dynamical processes. This approach represents an advancement in XPCS analysis, addressing the growing demand to extract intricate non-equilibrium dynamics. Further, the methods presented are agnostic to the nature of the material system and can be potentially expanded to other condensed matter systems.

104

Real-time ptychography

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Ptychography is an X-ray scanning microscopy imaging technique which obtains an image containing both structural and chemical information of the sample. Samples can be imaged down to approximately 10 nm resolution using Ptychography when using light produced at a synchrotron. This entails a complex data analysis and requires high computational power, as the phase is being retrieved by iterative algorithms.

Traditionally the ptychographic reconstruction would be started after the scan has finished, and any feedback of the experimental settings and how well the scan would reconstruct would be further delayed by the long processing times.

To overcome these barriers, we have developed algorithms to perform the reconstruction in real-time, as the data is acquired by the detectors. By starting the reconstruction process before the scan is complete, real-time ptychography facilitates faster optimisation of experimental parameters in response to the observed sample features. This real-time feedback can also help identify sample drift and damage, allowing researchers to adjust experimental conditions as needed to maintain sample integrity and data quality. Real-time ptychography thereby help researchers making more efficient use of the experimental resources. Continuing forward, our primary focus lies in uncovering the potential differences between real-time and traditional ptychographic reconstructions.

105

Evolution of Light-induced Transitions in Dielectric-metallic Nanostructures with SwissFEL

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Ultrafast and intense laser pulses can induce dynamic phase transitions out of thermal equilibrium, such as non-thermal melting [1], bond hardening [2], and the formation of dense electron-hole plasma [3]. When highly intense femtosecond pulses are used to excite semiconductors, a sizeable portion of valence electrons are excited to the conduction band and the interatomic forces are altered. Consequently, the electrons as well as crystal lattice are pushed out of equilibrium. On the nanoscale, it is still difficult to observe phase transitions in situ with femtosecond and nanometer spatiotemporal resolution directly [4]. Furthermore, melting speeds are affected by the deposited energy and morphology of nanoparticles, which can trigger homogeneous or heterogeneous melting. These further complicate our understanding of phase transitions in metallic and dielectric-metallic core-shell nanostructures.

We employed time-resolved single-shot single-particle imaging of free-flying silica-coated gold nanoparticles at the SwissFEL Maloja endstation [5]. The nanoparticles (50 nm diameter core size and 20 nm shell thickness) were pumped with an intense femtosecond laser pulse (800 nm, 35 fs FWHM, focal size of 120 µm FWHM). The polarization and pulse energy of the pump laser were tuneable during the measurement. A minimally absorbed energy density of 23.3cm-3 corresponding to 250 µJ pulse energy, was used to elevate the lattice temperature above the melting point of gold. The typical X-ray pulse energy is around 2 mJ, and the X-ray photon energy was set at roughly 1000 eV.

Sequential single-shot single-particle diffraction patterns with femto- to nanosecond delay times yield insight into the ultrafast laser-induced dynamics. The coherent images of the nanoparticles show a consistent pathway depending on the pump lasers pulse energy and polarization. For lower pump energies (250 µJ), clear gold diffraction speckles can still be seen after 200 ps. At greater pump energies (1400 µJ), the particles are nearly annihilated after 10 ps. For medium pump energies (500 and 750 µJ), a fast structural change occurs at around 10 ps after that the speckle oscillation smear out along with the delay time. Streaking tail speckles were also found from the medium pump energies due to the near-field enhancement. The data also show that during the first few picoseconds, the silica is fully disintegrated. In particular, for the medium and high pulse energies, the silica contributes with significant ionization potential to the electron trapping and melting process. Bare gold nanoparticles without the silica shell melt more quickly than core-shell nanoparticles. Our research suggests an intriguing connection between metallic gold core and dielectric silica shell, which strongly alters the melting regime and mechanism on the nanoscale and ultrafast timescale.

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94

Design Considerations for Exploring Sub-Microsecond Dynamics at XFELs using X-Ray Coherent Scattering

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At the dawn of x-ray free electron laser (XFEL) science, conceptual studies predicted that x-ray photon correlation spectroscopy (XPCS), particularly a double pulse variant, would be a powerful tool to study the structural dynamics of materials over time scales ranging down to tens of femtoseconds and atomic lengths [1,2]. Two decades later, that promise is beginning to be fulfilled. However, the application of XPCS and other speckle based techniques at an XFEL is still challenging due to limited instrument optical performance and radiation effects (e.g. damage) on the sample.
We will discuss design considerations for a dedicated coherent diffraction instrument from the point of view of minimizing x-ray radiation effects through a series of optimizations including 1) by increasing the sample thickness, 2) defining a temporal coherence that matches the sample thickness, 3) choosing an optimum photon energy and 4) maximizing the illuminated area consistent with maximizing speckle contrast and signal rate.
Based on current detector parameters, these optimized parameters require a very long sample-to-detector distance (SDD) to maintain a high contrast. We will present an instrument design that can efficiently provide long SDD while maintaining the ability to rapidly set momentum transfer by inserting crystals with different instead of moving the detector.
We will present the results of a modeling study of a concept instrument including wave front propagation of FEL pulses through the instrument, analysis of speckle size and contrast as well as the signal-to-noise ratio for several classes of samples using the optimized beam and sample parameters.
This work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Contract No. DE-AC02-76SF00515.
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66

A high-performance reconstruction method for partially coherent ptychography

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Ptychography is now integrated into mainstream microscopy allowing quantitative and high-resolution imaging capabilities over a wide field of view. However, its ultimate performance is inevitably limited by the available coherent flux when implemented using electrons or laboratory X-ray sources. We present a universal reconstruction algorithm with good tolerance to low coherence for both far-field and near-field ptychography. The approach is practical for partially temporal and spatial coherence and requires no prior knowledge of the source properties. Our initial visible-light and electron data show that the method can dramatically improve the reconstruction quality and accelerate the convergence rate of the reconstruction. The approach also integrates well with existing ptychographic engines. It can also improve mixed-state and numerical monochromatisation methods, requiring a smaller number of coherent modes or lower dimensionality of Krylov subspace while providing more stable and faster convergence. Hence this approach could have a far-reaching impact on studies of weakly scattering samples.

64

Forward modeling of dynamical effect in Bragg coherent diffractive imaging

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Bragg coherent diffractive imaging (BCDI) has proven to be a powerful tool for mapping the three-dimensional morphology and lattice deformation in crystalline materials. However, most BCDI phase retrieval algorithms rely on the kinematic approximation, which assumes that the exit complex wave of the diffracted beam is simply a projection of the complex crystal function. This approximation limits the application of BCDI to crystals with dimensions smaller than the corresponding x-ray extinction lengths.

Before developing a phase retrieval algorithm in the dynamical diffraction regime, it is necessary to accurately simulate the Bragg coherent diffraction from a finite crystal that incorporates the dynamical diffraction effect. Here, we present a forward simulation model and corresponding experimental validation of dynamical Bragg coherent diffraction from an arbitrarily shaped crystal. We demonstrate that our model accurately reproduces the subtle changes in interference fringes of the diffraction pattern due to the dynamical diffraction.

The proposed forward simulation method can serve as a cross-validation tool to assess the accuracy of Fourier transform-based models and guide the development of new algorithms capable of accommodating dynamical diffraction effects in BCDI phase retrieval.

7

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

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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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98

More is simple: Multiple Beamspot Coherent Scattering

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Speckle pattern arising from coherent scattering carries fingerprints of mesoscale heterogeneity. We previously showed how the dilute distribution of antiferromagnetic domains produces clear modulation of intensity as a speckle pattern on the antiferromagnetic Bragg peak. The simplicity and symmetry of the speckle pattern enabled us to reconstruct the real space distribution of the domains without the use of any algorithm from a single shot [1]. Here we will expand on our work showing how to utilize the dilute limit in a densely packed domain distribution.
Our approach involves the creation of new designs of Fresnel zone plates which restructures the beam footprint on the sample. A finite number of beam spots makes the scattered intensity distribution follow the same argument of simplicity in speckle pattern as observed above. First, we will discuss these designs of zone plates. Then through simulation, we will explore utilizing structured beam spots concerning correlation experiments and imaging experiments in condensed matter systems such as nickelates. Finally, we will show how a structured beam illumination leads to studying transport phenomena in such heterogeneous systems.

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75

Row-action methods for the general TIE equation in the Fresnel regime

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In this work we present a comparison of row-action methods for the original transport of intensity equation (TIE) in the Fresnel regime $\frac{\partial I(x,y)}{\partial z} = -\frac{\lambda}{2\pi}\text{div}[I(x,y)\nabla_\perp \phi(x,y)]$. For pure phase objects, it is already known that the original partial differential equation can be approximately solved by a convolution kernel (widely known as the Paganin solution). By assuming the original TIE without approximations, we use the fact that this is a linear problem on the phase, say $T(\phi; I) = \frac{\partial I(x,y)}{\partial z}$, with T describing the linear TIE-PDE transformation. It is easy to demonstrate that T is a self-adjoint operator allowing us to numerically compose an efficient row-action algorithm for computing the solution via orthogonal projections. Row-action methods are well know in the literature of inverse problems and can be used in different contexts, e.g., Cimmino, Hildreth and Bregman

strategies. Since the discretized matrix of T is sparse, we compare the obtained iterative strategies with a conventional numerical solution obtained through a sparse solver via finite differences of the TIE. Several simulated examples are presented, and a comparison with real data is presented. The proposed algorithms are an effort to numerically solve the phase-retrieval problem of some imaging beamlines of Sirius, the 4th generation Brazilian Synchrotron.

36

Phase retrieval beyond the homogeneous object assumption for X-ray phase contrast imaging

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From insights of pancreatic tumor tissue in medical research to nondestructive analysis of catalytic particles, highly resolved three-dimensional imaging provides a fundamental tool. Propagation based X-ray phase contrast imaging (XPCI) enables imaging of samples lacking conventional absorption-based contrast. Combined with coherent synchrotron radiation and computed tomography resolution in the sub-micrometer regime is achieved.

Differences in the samples' electron density imprints phase shifts in the probing X-ray beam that manifest in measurable intensity variations on the detector after sufficient propagation. Since the measurements are intensity-only, phase retrieval has to be performed. For XPCI this poses a severely ill-posed nonlinear inverse problem. Thus, commonly applied phase retrieval methods incorporate various approximations and regularization. One ubiquitous for the hard X-ray regime is that samples are *homogeneous*, *i.e.* solely made out of a single (refractive) material. Albeit this approximation is adequate for a broad range of samples, it poses an inexactness for multi-material compositions, *e.g.* stained tissue, that cause quality diminishing artifacts.

To this end, we extend phase retrieval based on the widely used contrast transfer function (CTF) for samples beyond the homogeneous object. Using the linearized CTF allows fast phase retrieval for large tomographic datasets. To improve reconstruction quality and robustness, our method allows to incorporate a priori knowledge in the reconstruction. Moreover, a nonlinear phase reconstruction algorithm is presented for samples beyond the validity of the CTF. We demonstrate our methods on experimental data taken at the GINIX instrument located at the P10 beamline of the PETRA III storage ring at DESY, Hamburg.

69

Multi-beam ptychography limits: a simulation study

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Ptychography has become an essential tool for nanometer-resolution X-ray imaging with the development of synchrotron radiation facilities. Based on the working properties of ptychography, experiment time increases when seeking large fields of view at very high resolution. Employing multiple mutually incoherent beams in parallel helps to mitigate this problem, which also makes

use of a greater portion of the available X-ray radiation. Multi-beam ptychography has the potential to collect information faster than single-beam ptychography, as larger areas are studied in the same amount of time. Furthermore, prospective requirements for data storage are reduced because the same information is collected in fewer diffraction patterns.

It has been shown that multibeam ptychography works successfully with up to six beams [1, 2, 3]. Knowing the limitations is a fundamental step in gaining insights into the potential of multibeam ptychography and driving the refinement. This study is discovering the limit by doing a series of simulations, and also exploring what factors are limiting the maximum number of beams. Identifying the maximum number of probes that can be utilized without compromising successful reconstruction.

Using simulated datasets, the reconstructability of multi-beam ptychography and the dependency on experimental parameters has been demonstrated. The primary goal is to discern patterns in the relationship between the number of probes and the feasibility of reconstruction with defined sampling parameters.

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51

Alternative Methods for Achieving Atomic Resolution in BCDI

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Many synchrotrons around the world are undergoing multi-bend achromat upgrades to become diffraction limited storage rings, such as the Advanced Photon Source – Upgrade. These upgrades will increase the incident photon flux by around 200 times. Theoretically, these upgrades will allow Bragg coherent diffraction imaging (BCDI) reconstructions to resolve atomic positions in materials. However, current BCDI phase retrieval techniques would require arrays of immense sizes and correspondingly expensive computational cost when based on fast Fourier transforms alone. In addition, it is not clear how effective algorithms such as shrinkwrap would be in resolving individual atoms. To address these challenges, we propose a new algorithm, termed Phase Retrieval with Atomic Modeling and Molecular dynamics (PRAMMol), which attempts to directly solve for atomic positions using maximum likelihood estimation rather than enforcing a Cartesian grid across the reconstruction. Solving for atomic positions directly allows PRAMMol to utilize molecular dynamics (LAMMPS) to restrict the search space to physically realistic objects. We will show that PRAMMol can effectively reconstruct small grains (5000-10000 atoms) with a screw dislocation or void from their simulated diffraction patterns.

68

3D Bragg Coherent Diffraction Imaging of Extended Nanowire Heterostructures

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The nanowire (NW) geometry allows for growth of complex device heterostructures that can accommodate much larger strains than thin films. Beyond just axial NWs, heterostructures formed along the NW length, more complex radial heterostructures can be designed to enable unique device capabilities. For example, InGaAs radial quantum wells (QWs) can be grown on a GaAs core to form compact near-IR lasers, with the QW acting as the emitter and the NW core acting as a lasing cavity. Beyond III-V's, NWs are of interest for light harvesting and sensing, using materials such as $CsPbBr_3$ or WO_3, due to their large surface area compared to thin films. However, the complex structures that can be designed in the NW geometry are difficult to study due to their 3D nature. Bragg coherent diffraction imaging (BCDI) is a popular choice for 3D imaging of strain in nanoparticles, fitting with the constraint that the crystal must be fully isolated and fit within the coherent beam. However, the extended length of NWs typically preclude them from investigation using BCDI. In this work, we demonstrate multiple cases of BCDI investigation of extended NW structures by using unintentional and intentional structural variations along the wire length. First, we investigated III-V radial QW heterostructures with BCDI, looking at NWs larger Zincblende twinning, which does not effect the overall strain or performance of the NW device allows for an isolated (twinned) segments to be reconstructed using BCDI. In these reconstructed segments we observe strain associated with the radial QW and a line dislocation propagating down the length of the wire, giving insights into the optical performance of the device. Second, we recently investigate III-V heterostructures of GaInP with embedded InP axial segments. Preliminary reconstructions show features associated with out-of-plane strain due to the mismatch between InP and GaInP. Both of these examples demonstrate the feasibility of BCDI as a tool for investigating unaltered extended NWs to access high resolution strain information in 3D. In this way, we can better understand the relationship between strain and performance in complex nanowire device heterostructures.

76

Nanoscale Diffusion of Supercooled Proteins Investigated by X-ray Photon Correlation Spectroscopy

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Understanding protein dynamics is crucial for unraveling cellular processes in complex biological environments. Here, we present our investigations into the nanoscale diffusion of both hydrated proteins [1] and concentrated protein solutions [2], extending into the supercooled regime. We employed X-ray Photon Correlation Spectroscopy (XPCS), a technique that utilizes coherent X-rays and can resolve collective nanoscale dynamics from microseconds to hours in a broad range of soft condensed matter systems. Firstly, our analysis of hydrated proteins reveals an increase in dynamical heterogeneity upon cooling, along with enhanced fluctuations observed in the two-time correlation functions and a maximum in the dynamic susceptibility, quantified by the normalized variance of the two-time correlation functions. These findings align with previous studies on hydrated proteins [3], suggesting the significant role of density and enthalpy fluctuations in hydration water. Our study is

consistent with the observed two-state structure of liquid water and the crossing of the Widom line around 230 K at ambient pressure. Secondly, the concentrated ferritin solutions in a glycerol/water mixture indicates Brownian-like motion down to cryogenic temperatures (T = 210 K) throughout the probed q-range ($q \approx$ 0.1-0.5 nm-1), as evidenced by the intensity correlation functions. The extracted diffusion coefficients exhibit Arrhenius temperature dependence for both dilute (7 mg/ml) and concentrated (200 – 450 mg/ml) ferritin solutions, with significantly different activation energy barriers (≈30 and ≈50 kJ/mol). We hypothesize that this variation is attributable to differences in protein concentration, which can indicate the presence of protein transient clusters due to the increased strength of protein-protein interactions. Our studies showcase the applicability of XPCS utilizing synchrotron radiation to probe collective protein dynamics at the length scale of proteins in supercooled conditions. These insights contribute to advancing our understanding of protein stability in cryogenic environments, with direct relevance to biotechnical cryo-storage technologies.

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103

High-resolution ptychographic imaging at a seeded free-electron laser source using OAM beams

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The applications of electromagnetic waves with orbital angular momentum (OAM) span from optical communications and quantum technologies, to optical tweezers and microscopy. OAM fs-pulses at extreme-ultra-violet (XUV) wavelengths became recently available at High Harmonic Generation and Free Electron Laser (FEL) sources, allowing for cutting-edge applications, such as the detection of helical dichroic effects in chiral molecules [2] and magnetic nanostructures [3]. In microscopy applications, OAM beams were proposed for enhancing edge detection in phase-contrast microscopy [4], and for increasing spatial resolution [5] in optical microscopy.

In this study, we performed a single-shot-per-position ptychography on a nanostructured object; the experiment carried out at the DiProI station [6] exploited XUV beams (produced by a seeded FEL - FERMI) of different topological charge order ℓ, generated by different spiral zone plates [7].

We show that the ℓ-dependent beam-profile features can lead to an improved spatial resolution with respect to conventional Gaussian profile illumination (ℓ=0) [8].

This result enhances the capabilities of coherent diffraction imaging techniques and paves the way for time-resolved and high-resolution microscopy of extended specimens, in turn boosting both imaging and dichroism future experiments.

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10

Grazing Incidence X-ray Ptychography

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The morphology and distribution of nanoscale structures, such as catalytic active nanoparticles and quantum dots on surfaces, have a significant impact on their function. Thus, the capability of monitoring these properties during manufacturing and operation is crucial for the development of devices that rely on such materials. We demonstrate a technique that allows highly surface-sensitive imaging of nanostructures on planar surfaces over large areas. The capabilities of hard x-ray grazing-incidence ptychography combine aspects from imaging, reflectometry, and grazing-incidence small angle scattering in providing images that cover a large field of view along the beam direction while providing high surface sensitivity. For homogeneous samples, it yields a surface profile sensitivity better than 1 nm normal to the surface, with a poorer resolution in the sample surface plane, (i.e., along the beam and transverse to the beam). Like other surface scattering methods, this technique facilitates the characterization of nanostructures across statistically significant surface areas or volumes but with additional spatial information.

106

Ptycho-tomography: recent advancements for robust 3D imaging

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Ptychography, a coherent diffraction imaging method, surpasses the limitations of traditional lens-based microscopy. It involves scanning an object to gather diffraction images from overlapping probe regions, enabling high-resolution imaging of microscopic specimens. Recent interest lies in extending ptychography to three dimensions for reconstructing an object's refractive properties while maintaining reasonable data acquisition times. Traditionally, this involves sequential steps of ptychography and tomographic reconstruction, keeping the necessary illumination overlap in ptychography, which limits efficiency. A more efficient approach involves jointly solving ptychography and tomography problems, leveraging the inherent information coupling among projections. We use a distributed optimization approach by employing alternating direction method of multipliers (ADMM) to split the joint problem into ptychography, tomography, and regularizer components. In this talk, I will summarize recent advancements aimed at enhancing the robustness of 3D imaging.

26

Characterising Nanoparticle Dynamics by Coherent Diffractive Imaging and X-ray Photon Correlation Spectroscopy

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Understanding the behaviour and function of nanomaterials necessitates characterisation *in situ* and *operando*. Achieving high spatial resolution at the required timescales, however, is inherently difficult. With the emergence of diffraction limited light sources [1] and the development of fast-framing detectors [2], coherent X-ray techniques, such as X-ray photon correlation spectroscopy (XPCS) and coherent diffractive imaging (CDI), are changing this situation. XPCS has routinely been used to characterise dynamics systems with high temporal resolution [3], although it lacks spatially resolved information. Meanwhile, CDI can provide nanoscale spatial resolution [4], but requirements of a high signal-to-noise in the diffraction patterns limits the achievable temporal resolution, and therefore, CDI has typically been used for static systems. Consequently, although both techniques require the measurement and analysis of coherent diffraction patterns, the combination of both techniques has been out of reach until recently [5].
Here, we demonstrate a proof-of-principle experiment to explore the spatio-temporal space which can be accessed by the combination of XPCS and CDI. We performed an experiment at the P10 coherence applications beamline at PETRA III combining both techniques to study the Brownian motion of gold nanoparticles with different diameters and at different temperatures. We show that different motion modes can be determined by XPCS, and that the dynamic behaviour of individual nanoparticles can be tracked by CDI. The current limits of this combination for the different dynamic behaviours will also be discussed.

53

What does BCDI bring to the study of heterogeneous catalysis ?

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The main objective of this work is to study heterogeneous catalysts *in situ* and *operando* during the oxidation of ammonia by approaching industrial temperature and pressure values.

Currently, this catalytic process and the associated structural changes are poorly understood, we propose to use different samples in platinum, nanoparticles and single crystals in order to reduce the gap between scientific studies on model samples and catalysts used in industry. The catalytic activity of the different samples is measured to link structure and selectivity during the reaction, which can be focused towards the production of nitrogen (N2), nitric oxide (NO), or nitrous oxide (N2O). Three techniques were mainly used, Bragg coherent diffraction imaging (BCDI), surface X-ray diffraction (SXRD) and X-ray photoelectron spectroscopy (XPS), combined with mass spectrometry measurements. These techniques are compatible with ambient pressure, making it possible to reduce the difference in pressure between surface science studies and industrial conditions for heterogeneous catalysis.

Measuring the structure of nanoparticles at the nanoscale with BCDI makes it possible to reveal the effects of volume, surface and interface tension and compression, as well as the existence of different types of defects. In addition to imaging studies by BCDI on individual nanoparticles, the study of a set of nanoparticles is carried out via grazing incidence X-ray diffraction, revealing critical complementary information such as changes of particle morphology in the group of particles. Moreover, different behaviours were revealed during the oxidation reaction on two nanoparticles measured with BCDI at 400℃, which present a different size, shape, facet coverage and initial deformation state, while no significant changes were measured below 600℃ by SXRD. In addition, two types of surfaces present on the nanoparticles ({111} and {100}) were also studied using single crystals with SXRD and XPS, revealing phenomena associated to heterogeneous catalysis invisible with BCDI, such as the growth of surface oxides, the presence of surface reconstructions, and surface relaxation. Therefore, the surface structure as well as the presence of adsorbed species can be linked to the measured catalytic activity, allowing a better understanding of the reaction mechanism.

110

Dynamics of Bentonite Water Interaction

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Bentonite is used in a wide range of applications due to its intrinsic density, swelling capacity and isolation capabilities, including in nuclear waste facility where bentonite swelling is employed to

isolate the waste from surrounding hydrogeological systems [1]. Sol-gel transitions and colloidal transport properties are central to bentonite applications [2]. Bentonite materials swelling capacity has been the focus of many studies, primarily looking at the swelling pressure and extent. Different bentonites have varying swelling volumes and pressures, those with sodium interstitial ions have been reported to swell up to 15x their original volume. We investigated, in recent X-ray photon correlation spectroscopy (XPCS) experiments conducted on ID10 at ESRF, the dynamics of calcium and sodium bentonite swelling as water is added slowly. The momentum transfer region investigated was between 0.00004 and 0.49 $^{-1}$, including the (001) Montmorillonite reflection (primary component of bentonites) which moves as the bentonite swells. Early analysis indicate that bentonites undergo rapid a transition initially as water is added to the sample. This can be roughly interpreted as the water being drawn into the course grain bentonite, a sort of wetting mechanism. The relaxation rate slow slightly with time as water is drawn into the bentonite layers. The onset of gel formation brings about a stabilization of the bentonite movements, similar to the results reported by Whittaker et al. [3,4]. As the gel ages the relaxation slows, becoming slower as the sample begins to dry. Dynamics continue to slow until the relaxation becomes too slow to observe with in the experiment time. Our experiment has potential impact on the design of containment structures as understanding the swelling and drying of bentonites is inherent in such isolation systems.

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92

Structural characterization of calcium carbonate microparticles using combined coherent diffraction imaging and x-ray diffraction

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Calcium carbonate is a ubiquitous substance in nature that appears in three anhydrous crystalline phases: vaterite, aragonite, and calcite [1]. These different phases can be synthesized by precipitation from solution by controlling temperature, reactant and additives concentration, reaction time, and solution stirring rate. The crystals of CaCO3 spanning a few nanometers in size can self-assemble into microparticles with various shapes and morphologies [2]. Thus CaCO3 is studied as a model system to decipher the mechanisms of nucleation pathways and crystal growth, biomineralization, and self-assembly. In this work, we demonstrate the combined use of coherent diffraction imaging (CDI) and x-ray diffraction (XRD) to understand the morphology and structure of CaCO3 microparticles. CDI is a lensless imaging technique that allows us to visualize the three-dimensional organization of nanocrystals at nanometer resolution [2,3] whereas XRD provides information on crystalline phases and the preferred orientation of the crystallites in a single particle. In this presentation we will focus on the following examples – i) the self-transformation of CaCO3 vaterite microparticles into core-shell and hollow spheres [2], ii) the formation of calcite microparticles in the presence of protein Fetuin-A, and finally iii) investigate the effect of temperature and reaction time on the formation of vaterite and aragonite phases. These case studies highlight the unique capabilities of the combined CDI- XRD technique for the investigation of crystal growth and self-assembly.

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14

Patching-Based Deep Learning model for the Inpainting of Bragg Coherent Diffraction patterns affected by detectors' gaps

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In hard X-ray detectors gaps are common due to manufacturing limitations. The information lost during detection can lead to artefacts in the subsequent data analysis. Here we demonstrate a novel, efficient and versatile deep learning algorithm to inpaint these gaps, applied in the field of Bragg Coherent Diffraction Imaging (BCDI) where artefacts arise when the gaps are left empty.
The key aspect of our method lies in the choice of training the neural network with cropped sections of both experimental diffraction data and simulated data and subsequently patching the predictions generated by the model along the gap, thus completing the full diffraction peak. [1]
With respect to other similar works [2,3], this strategy enhances the accuracy of inpainting and by augmenting the training dataset with experimental data it facilitates faster model training due to its limited size. The method is image size independent and can be applied to arbitrarily larger BCDI datasets.
Beyond broadening the scope of application, our approach ensures the preservation of data integrity and reliability, even in the face of challenging experimental conditions. This novel deep learning-based inpainting technique represents a significant step towards more robust and accurate reconstructions.

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71

XPCS in bunch mode: XPCS-echo and wide timescale measurements

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With the emergence of 4th generation synchrotrons, X-ray Photon Correlation Spectroscopy (XPCS) has improved both in terms of the available coherent photon flux and speckle contrast [Chevremont2024]. Together with fast photon counting pixel array detectors, this technique has become more attractive for the investigation of a broad range of systems [Narayanan2023]. However, one of the main limitations of XPCS measurements is the sample degradation by the X-ray beam and resulting beam-induced dynamics [Chushkin2022]. Yet another bottleneck when performing XPCS at a high frame rate for a long time is the time and memory needed to process the data, which scale quadratically with the number of frames.

To address these issues, a novel acquisition scheme has been developed at the TRUSAXS Instrument (beamline ID02), ESRF. This new scheme has been termed as “time resolved” or “bunch mode” XPCS. It involves acquiring frames at the highest frame rate according to the lowest lag time to be measured only when needed and thus exposure of the sample to the beam is minimized. Bunches of frames are acquired, spaced by variable dead time where the fast beam shutter remains closed and the sample unexposed to X-ray beam. The autocorrelation functions are still calculated by performing the correlation between all the frames, and reconstructing the two-time correlation function (TTCF) before averaging.

In this work, the application of the bunch mode XPCS is demonstrated on samples (colloidal gels formed by short-range attraction between particles) whose correlation functions span over several orders of magnitude in lag time. Another interesting application is for XPCS-echo, where the acquisition is synchronized with an oscillatory motion of the sample imposed by a rheometer [Pham2004]. In this experiment, the detector frames are acquired on each oscillation period and echoes in the autocorrelation function [g2(q,t)] appear each time the sample returns to the initial position. The envelope of echoes measured corresponds to the decay of the autocorrelation function due to the intrinsic dynamics in the sample. For fully reversible motion, the envelope strictly corresponds to the autocorrelation function of the sample at rest. On the other hand, an acceleration of the decorrelation occurs when the sample yields with increasing amplitude of deformation. The XPCS-echo then provides an elegant way to measure the intrinsic dynamics within the sample, discriminating the Doppler shifts caused by the shear.

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13

Real-time XPCS study of an InN thin film growth by Plasma-Enhanced Atomic Layer Deposition

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III-V nitrides have unique electronic and optical properties and a wide range of applications across multiple industries. In this work, Plasma-Enhanced Atomic Layer Deposition (PE-ALD) of an epitaxial indium nitride film was studied real-time using X-ray Photon Correlation Spectroscopy (XPCS) to understand the nanoscale surface dynamics during growth. Ultrathin films were synthesized from

repeated cycles of alternating self-limited surface reactions induced by temporally separated pulses of the material precursor and plasma reactant, allowing the influence of each on the evolving morphology to be examined. Our results document the surface morphology evolution at each growth stage. Numerous overlapping stress-relief events are observed during the initial film growth as sudden correlation changes in the calculated Two-Time Correlation Function (TTCF). Subsequently, as the film achieves continuity after the initial growth stage, the nanoscale surface morphology undergoes an abrupt transition to a long-lived state, with correlation times spanning the experiment's duration. Throughout the later part of the growth experiment where average scattering intensities remained stable, there is a consistent repeating pattern of correlations in TTCF associated with the cyclic growth process, which is modeled as transitions between different surface states. The measured relaxation time information for each modeled surface state could be useful for studying and fine-tuning other complicated growth processes that include a greater number of disparate stages per cycle.

11

Theoretical Analysis of Coherence Properties of the Gratings

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In this work we aim to clarify theoretically the spatial coherence properties of the X-ray beam passing the variable line space (VLS) grating in the focal plane of this grating. Assuming that the VLS grating is located in horizontal direction we are interested in coherence properties of the beam in dispersion or in the vertical direction. We first consider the fully spatial coherent beam illuminating the grating. We show that spatial coherence properties depend on the bandwidth of the incoming radiation. Being fully spatially coherent up to the VLS grating, the spatial coherence properties degrade in the focal plane of the VLS grating. We attribute this to coupling of spatial and frequency components at each diffraction order in the focal plane of the grating. Next, we examine partially coherent beams incoming on a VLS grating. We assume that the incoming on a grating radiation is of Gaussian Schell-model type and obtain analytical expression for the spatial coherence properties of the beam in the focal plane of the VLS grating for such a field. Next, we consider a monochromator setting that is provided by installing slits in the focal plane of the VLS grating and examine degree of coherence in this case. We finally evaluate the degree of coherence at different openings of the exit slits assuming coherent illumination of the grating and the bandwidth on the order of 2·10^{-4}.

16

Advancements in Sample Delivery Techniques for FXS: Harnessing Sheet Jets at XFEL Facilities

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Fluctuation XRAY scattering (FXS) presents a powerful avenue for 3D imaging of hydrated, native-state biomolecular configurations without the need for crystallization, cryogenic freezing or aerosolization. It requires the extremely bright, femtosecond-duration exposures produced at X-ray Free Electron Laser (XFEL) facilities. The challenge lies in effectively extracting three-dimensional electron density maps with sub nanometer resolution from ostensibly rotationally isotropic scattering data, which is achieved by extracting two-point intensity correlations. Our research has focused on enhancing the Signal-to-Noise Ratio (SNR) in FXS experiments by developing ultra-thin liquid sample injectors that allow us to reach the shot-noise limit of statistical errors. We investigated various methods, including nano-droplets, nano-jets, and nano-sheets, for delivering protein samples during XFEL measurements. Our exploration revealed that nano-sheets are a promising approach because they produce a stable film thickness of under 100 nanometers with a high hit rate. Through our experiment conducted at CXI end station at LCLS (LY59), we successfully obtained a SAXS profile of Hemoglobin using a nanosheet, marking a significant milestone in solution scattering and FXS research. To facilitate sheet-jet injection, we developed a 3D printed nozzle with micron-level resolution, which utilizes helium gas jet to break a Rayleigh jet into a nanosheet in vacuum. Despite challenges such as sample freezing in vacuum and high sample consumption rates, our ongoing engineering efforts have significantly mitigated these issues. Subsequent improvements have not only addressed freezing concerns but have also reduced sample consumption by a factor of three and potentially higher, paving the way for more reliable and efficient FXS measurements.

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4

Coexisting Multi-Timescale Fluctuations at the Chiral Nematic Phase Boundary in Amorphous FeGe

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Spatiotemporal correlations stabilize exotic phases of quantum materials. Here I present recent results from resonant soft coherent scattering experiments on the Smectic-like to Nematic-like phase transition in amorphous $Fe_{51}Ge_{49}$. By combining synchrotron measurements from the Advanced Light Source and free electron laser measurements from the Linearly Coherent Light Source, we have found a region of instability as a precursor to the Nematic-like phase where fluctuations coexist on the timescale of both minutes and sub-nanoseconds. These results emphasize the general

nature of multi-step and multi-time scale topological phase transitions and are likely applicable to a broad class of materials and phenomena.

54

4D Characterization of Dynamic Samples with X-Ray Multi-Projection Coherent Imaging

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Coherent Diffraction Imaging (CDI) has emerged as a seminal technique for investigating nanoscale structures, enabling non-intrusive insights into the internal composition of specimens. Implementation of a 3D CDI experiment necessitates a rotation stage, capturing several tens of projections from diverse angles to reconstruct a 3D object. The characterization of dynamic objects presents challenges due to the rotational stage limiting maximum speed.
In response to this limitation, our research group has introduced an X-ray multi-projection imaging (XMPI) method, simultaneously capturing three specimen perspectives. The integration of XMPI with CDI is introduced to characterize dynamic objects. We propose a novel deep learning-based reconstruction algorithm to mitigate sparse view challenges inherent to XMPI and the requirement for reproducible samples in the 3D imaging experiments in XFELs. This method requires fewer than three projections captured at each time point to reconstruct 4D movies of dynamic samples. Meanwhile, our method overcomes the requirement of reproducible samples through transfer learning. Samples with similar structures are enough for the neural network to capture the features of these objects, resulting in a more efficient way to conduct 3D and 4D imaging in XFELs.
By leveraging neural networks and advanced learning strategies, our algorithm can learn and comprehend the intricate variations in dynamic samples, effectively preserving details and motion information during reconstruction and mitigating the requirement for reproducible samples in XFEL experiments. This innovative approach enables the acquisition of ultra-high spatiotemporal resolution reconstructions of dynamic samples and opens up new possibilities in various fields, such as scientific research and medical imaging.

67

The (small) influence of Compton scattering in coherent diffraction imaging experiments

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Compton scattering is generally neglected in diffraction experiments because the incoherent radiation it generates does not give rise to interference effects and therefore is negligible at Bragg peaks. However, as the scattering volume is reduced, the difference between the Rayleigh (coherent) and Compton (incoherent) contributions at Bragg peaks diminishes and the incoherent part may become substantial. The consequences can be significant for coherent diffraction imaging at high scattering angles: the incoherent radiation produces background that smears out the secondary interference fringes, affecting thus the achievable resolution of the technique. We introduce a criterion that relates the object shape and the resolution. The Compton contribution for several object shapes is quantified, and it is shown that the maximum achievable resolution along different directions has a strong dependence on the crystal shape and size. These results can readily be extended to other types of noise or background.

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40

Perfecting the gas-focused liquid microjet

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Gas-Dynamic Virtual Nozzles (GDVNs) have been vital for XFEL (X-Ray Free-Electron Laser) experiments since the Linac Coherent Light Source (LCLS) began operating in 2009. They produce high-speed liquid microjets that are particularly important for serial crystallography and solution scattering experiments at MHz and for rapid mix-and-inject studies. Despite continuous improvements to GDVN fabrication methods, nozzle failures still cause substantial losses in beamtime productivity. In order to minimize such losses, and to expand the operational range of GDVNs, we have developed a semi-automated platform for detailed characterizations of liquid microjets. We have studied how microjet properties vary according to nozzle geometry, operating conditions, and liquid properties.

In this poster, we will outline the automated platform that we developed for this purpose, which includes an open-source python-based software package with a graphical interface. We will delve into quantitative results that show how variations in liquid properties such as viscosity and microparticle size influence the transition from jetting to dripping phases. We will also show how some nozzle geometries can dramatically affect the jet-to-drip transition and enable ultra-low flows down to 0.1 μL/min, which results in jet diameters below 300 nm. We discuss how nanojet protein solution scattering of ultra-fine nanojets can help enable XFEL-based biomolecular imaging methods such as Fluctuation X-ray Scattering.

This analysis correlates closely with experimental observations that were seen in the LY59 beamtime at the LCLS through diffraction of a nanojet and fringes that allowed us to measure a it in real time. We are offering a detailed characterization of the transitions between jetting and dripping, and the dynamics of nanojet formation. Our investigation, delves into the impact of sample concentration on jet performance, providing valuable insights for optimizing microjet and nanojet configurations in XFEL applications.

This research is laying the groundwork for future studies to improve XFEL crystallography and solution scattering experiments. By tackling the challenges of varying sample concentrations, our findings enhance experimental design, opening avenues for groundbreaking discoveries in XFEL research.

87

X-ray Photon Correlation Spectroscopy of Supercooled Ferritin Protein Solutions

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Studying protein dynamics is fundamental to understanding cellular processes, such as transport, assembly and disassembly occurring, for instance, in crowded biological environments. Here, I will present our measurements of the diffusion of concentrated solutions of the protein ferritin in glycerol/water mixture, extending into the supercooled regime 1. We employed X-ray Photon Correlation Spectroscopy (XPCS) to resolve collective nanoscale dynamics ranging from microseconds to

hours [2]. The intensity correlation functions indicate Brownian-like motion down to cryogenic temperatures (T = 210 K) throughout q-range probed ($q \approx$ 0.1-0.5 nm-1). The extracted diffusion coefficients exhibit an Arrhenius temperature dependence for both dilute (7 mg/ml) and concentrated (200 – 450 mg/ml) ferritin solutions, although with significantly different activation energy barriers (≈30 and ≈50 kJ/mol). We hypothesize that this variation is due to differences in protein concentration, which can indicate the presence protein transient clusters due to the increased strength of protein-protein interactions. Our study demonstrates the applicability of XPCS utilizing synchrotron radiation, for probing collective protein dynamics at the length scale of proteins in supercooled conditions. These insights can advance our understanding of protein stability under cryogenic environments, with relevance to biotechnical cryo-storage technologies.

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8

Microsecond Dynamics in Complex Liquids with MHz XPCS

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Complex liquids are a broad family of materials that cover key roles in several aspects of everyday life, from biological processes that can take place only in such environments to industrial applications that sees complex liquids either as a final product or as a fundamental manufacturing step. From a physicist perspective most of the interesting phenomena take place at inter-particle distances, which for proteins and nanoparticles correspond to few nanometers, accessible only via X-ray based techniques. Moreover, such processes are often connected to diffusion mechanisms, which for water-based systems implies timescales of the order of few microseconds. These time and spatial constraints pose a real challenge to current 3rd generation synchrotron sources, limiting experiments only to a handful of complex experiments on prototypical systems 1.
The MHz repetition rate of the European XFEL matches perfectly with these timescales making it an ideal choice for this kind of experiments. Here we report the results from MHz X-ray Photon Correlation Spectroscopy (XPCS) experiments performed at the instruments MID and SPB/SFX [2,3], showing how it is possible to execute measurements both on prototypical charge-stabilized silica in water and on radiation-sensitive core (silica) – shell (PNIPAm) nanoparticles. Tuning the pulse intensity and repetition rate it is possible not only to measure the original dynamics of the systems but also to control the radiation-induced heating of the system without necessarily damaging it even for more delicate PNIPAm - based samples. This possibility, combined with the capabilities of the XPCS techniques, opens the way to the study of out of equilibrium dynamics in the microsecond time-scale for a large variety of complex systems.

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21

Time-resolved Ultrafast Strain Evolution in SrTiO3 Nanocrystals by Bragg Coherent X-ray Diffraction Imaging

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The formation of polarons and relevant deformations in atomic displacement in perovskite-oxides is due to the localization of charged carriers within photo-excited crystal lattices. It is important to understand these phenomena and processes in an ultrafast time scale for practical applications and the underlying science. Such a polaron affects structural distortions and carrier transport in perovskite-oxides.
Polarons are expected to form in less than one picosecond (1 ps) by non-thermal effect. So far, polaron formation and propagation have been explained by computational outcomes or changes in observable peaks. There is no clear experimental observation or explanation for polaron formation and propagation behavior. It is crucial to obtain precise insights into the generation and initial propagation of polarons before their development as phonons to explore the potential of materials.
In this presentation, I will present the ultrafast lattice distortions by photoexcitation in perovskite oxides to understand initial polaron generation and evolution by time-resolved Bragg coherent X-ray diffraction imaging (tr-BCDI) with an optical laser pump and an X-ray pulse probe. By doing that, we can observe the lattice distortions caused by excited carriers at the nanocrystal level. By direct observation of atomic-scale motions linked to momentary internal structural distortions in nanoscale perovskite-oxides, I will review the polaronic distortion in ultrafast time scale. This work was supported by the National Research Foundation of Korea grant NRF- 2021R1A3B1077076.

56

Construction of Ptychography at Taiwan Photon Source

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Ptychography, a scanning coherent diffraction imaging, has been rapidly and successful development over the past decade. This technique allows for the retrieval of complete information with high spatial resolutions, including the wavefield of incident beams and the transmission function of samples. In Taiwan Photon Source (TPS), a third-generation synchrotron facility, preliminary tests of hard X-ray ptychography were conducted at beamline TPS 25A. Using a standard gold Siemens star specimen, the spatial resolution of the images obtained was determined to be sub-10 nm, as estimated by half-bit threshold of Fourier shell correlation (FSC).
The system consists of focusing optics (zone plate), piezo-stages and Eiger detectors. The phase retrieval program is a custom-built software utilizing the iterative cores of extended ptychographical iterative engine (ePIE), maximum-likelihood (ML) and difference map (DM) algorithms. Optimized

Fermat spiral trajectories are employed during the scanning process in ptychography. Position correction and mixture-state method are included to extend the tolerance for errors casued by beam-pointing instabilities and non-ideal sources.

28

Merging Xray Diffraction Data with Monte Carlo Expectation Maximization

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The crucial pre-phasing step of merging two-dimensional XFEL diffraction data into consistent, three-dimensional diffraction volumes is greatly complicated by the presence of latent, shot-by-shot variations. For example, in the case of single-particle diffraction experiments, the orientation of the target is unknown. There are many other types of these unmeasured variations that can happen between shots in a diversity of experiments, such as beam fluence, beam temporal profile, relative background strength, pump-probe timing, etc. Presented here is an algorithmic framework for handling these types of merging challenges given a reasonable probabilistic forward model for the unmeasured variations which leverages Monte Carlo integration to perform the high dimensional integrations involved.

24

3D CT reconstruction with sparse X-ray projections using Generative Neural Radiance Fields

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Medical imaging plays a crucial role in the diagnosis and treatment of many diseases. Computed tomography (CT) is a widely used imaging technique that uses X-rays to produce cross-sectional images of the body. However, the availability of projection data in clinical settings and in the nanoscale often remains limited. Additionally, the acquisition is expensive, and patients and samples frequently endure high doses of radiation. Excessive ionizing radiation will have decisive and harmful effects. This motivates the exploration of generating enough 3D-aware CT projections to improve the reconstruction. Thus, we propose a model that combines generative adversarial networks (GAN) and neural radiance fields in biomedical applications to render CT projections and improve reconstruction. We conduct a comprehensive performance evaluation comparing our approach against existing state-of-the-art techniques.

72

Rapid coupled phase retrieval for multipeak Bragg coherent diffraction imaging

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Multipeak (or multi-reflection) Bragg coherent diffraction imaging (BCDI) allows for highly consistent reconstruction of the three-dimensional strain field within nano- and microcrystals. However, the high computational cost of such reconstructions combined with the increasing brilliance of coherent x-ray sources presents a problem: data production is increasingly outpacing data processing and will soon be orders of magnitude faster. To help prevent a bottleneck, we present a multipeak phase retrieval technique for BCDI, based on serial-constrained optimization (SCO), designed to minimize both the time and memory required to perform reconstructions. We show that SCO is able to quickly and consistently reconstruct experimentally obtained diffraction patterns from crystals with varying levels of strain.

29

Chrono coherent diffractive imaging with crystallographic data

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It is known that crystallographic data under-sample the modulus of the Fourier transform of the unit cell, resulting in the crystallographic phase problem [1-2].

Here we apply the method of chrono coherent diffractive imaging [3], which utilises multiple datasets of the same sample collected at different time points to augment the information content of the phase retrieval problem. The datasets are tied together by continuity constraints, and we explore the different conditions required for successful reconstructions through simulations.

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30

The BRIGHT Nanoprobe beamline at the Australian Synchrotron

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The Australian Nuclear Science and Technology Organisation (ANSTO) has commenced the construction of a 100 m-long Nanoprobe beamline at the Australian synchrotron, as part of the BRIGHT project delivering 8 new beamlines. A cryogenic permanent magnet undulator source provides hard

X-rays for fluorescence mapping and ptychography imaging. The optical layout of the beamline includes a secondary source aperture, used to define the beam coherence and render the endstation insensitive to subtle variations in angle and position of the source. A double-multilayer monochromator will maximize the intensity at the sample. To achieve the spatial resolution target of 60 nm for fluorescence mapping, with a reasonable working distance of 80 mm between the focusing mirror vessel and the sample, the endstation is located far from the source in a dedicated satellite building. The building was designed to minimize the effect of thermal variations, drift, and vibration at the sample location, as well as providing convenient laboratory space for the users. The sample scanner provides 3 mm scan range in all directions with <10 nm positioning precision, with a rotation stage enabling tomographic extension of all imaging modalities. The status of the ANSTO Nanoprobe construction project will be presented.

78

Correcting angular distortions in Bragg Coherent X-ray Diffraction Imaging

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Bragg Coherent Diffraction Imaging (BCDI) is a powerful X-ray imaging technique to reveal 3D strain distribution of crystalline nanoparticles. The method records the 3D diffraction intensity of a nanoparticle slice by slice by incrementally rotating the sample within a very small angular range. The iterative phase retrieval method will then be employed to phase the sampled 3D diffraction intensity and provides the detailed three-dimensional (3D) distribution of strain. Thanks to the coherence produced by the latest 4th generation of highly brilliant X-ray beams, BCDI can achieve a very high spatial resolution. However, any angular distortions from nominal rocking angles due to factors like the radiation heating, pressure or the imprecise rotation stage in the data acquisition process can introduce the artifacts in the following phase retrieval, which limits the applicability of BCDI. This prevents us from exploring more in material science, especially for the case of small nanoparticles.
In this study, we introduce a pre-processing algorithm designed to mitigate the impact of unexpected orientations. Inspired by the Extension-Maximize-Compress algorithm commonly employed in single particle x-ray imaging, our approach generates and refines a 3D diffraction intensity volume from measured 2D diffraction patterns. It achieves this by maximizing a likelihood function informed by Poisson statistics. This function includes cross-correlation between photon counts in each measurement and pixels in each slice of the generated volume, facilitating the determination of the relative orientation trajectory. Additionally, we further impose spatial constraint (envelope) on the 3D diffraction volume update, effectively limiting the field of view and enforcing the particle's maximum physical dimensions.
Our method demonstrates significant resilience to angular distortions, accurately correcting for distortions up to 16.4 times (1640%) of the angular step size dθ=0.004^◦, which is comparable to the fringe spacing in our simulated dataset. The corrected result remarkably improves the quality of subsequent phase retrieval reconstruction, even in presence of Poisson noise.
The validation test underscores the potential of our pre-processing method to recover highly distorted experimental data that would otherwise be unusable. This advantage not only salvages data previously considered lost but also enhances the robustness of BCDI under less-than-ideal conditions. For example, our method can handle the data from the continuous scanning BCDI experiment.
In conclusion, the implications of this work extend to enabling BCDI in more demanding and challenging environments, fully leveraging the intensity of beam from 4th generation synchrotrons, pushing the frontiers of material science research.

91

Investigating pressure-induced modifications in CoSb3 nanoparticles by Bragg coherent diffractive imaging

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Bragg coherent diffractive imaging (BCDI) is a powerful tool in materials science, enables investigations of a diverse phenomenon at nano-scale. It has proven effective in examining changes in the 3D shape, strain, and defects of single crystals [1, 2]. BCDI technology has been applied under various complex conditions, including at different temperatures and atmospheres to investigate catalysts [3], at varying voltages to monitor particles changes during current flow [4], and at extreme high pressures to explore strain in particles [5]. However, applying BCDI under high pressure presents significant challenges, with previous applications primarily limited to simple systems, such as gold nanoparticles [5]. In this research, we address this limitation by selecting CoSb3 as the sample to investigate the boundaries of BCDI under high pressure. CoSb3, is a prominent skutterudite compound known for its exceptional thermoelectric properties, it has garnered significant interest from researchers. Numerous studies have focused on enhancing its Seebeck coefficient and thermoelectric efficiency through pressure-induced modifications [6]. Pressure-induced structural modifications in CoSb3 have revealed a 'self-insertion' process, characterized by complex atomic redistribution, particularly above 25 GPa [7].
In this study, we employed phase retrieval and shrinkwrap algorithms to reconstruct 3D images of CoSb3 single crystal particles. Our reconstruction results demonstrated expected changes in particle compression, thereby expanding the limitations of BCDI under high-pressure conditions. Through an analysis of diffraction patterns in 3D reciprocal space, we observed pressure-correlated changes, with a notable shift in the diffraction pattern occurring above 25 GPa, indicative of a critical threshold in CoSb3's response to pressure. Additionally, the Bragg peaks intensities as the function of d-spacing values resulting from the compression revealed discontinuous changes at 25 GPa. This study illuminated the complicate interplay between pressure and particle evolution, providing valuable insights into the underlying mechanisms of CoSb3's structural transformations elucidated by BCDI.

65

"Studying Domain Dynamics of Charge-Density Wave Materials using X-ray Photon Correlation Spectroscopy at XFELs"

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Mesoscale structural features and dynamics, particularly at domain and grain boundaries, significantly influence the macroscopic properties of materials across various classes and length scales, impacting, for example, electronic behavior, mechanical response, and dielectric properties. Correlating macroscopic properties of a system driven by an external stimulus to the fluctuations originating from mesoscale order provides an opportunity for new insights into structure-property relationships. In particular, for strongly correlated materials systems, these connections remain less explored.

Transition metal dichalcogenides (TMDCs), exemplified by tantalum disulfide (1T-TaS2), are known for their unique electronic characteristics, making them promising candidates for applications in charge-configurational memory devices and beyond. In 1T-TaS2 specifically, understanding the fluctuations or reconfiguration of charge-density wave (CDW) domains can illuminate mechanisms central to transport properties. The CDW order is a long-range electron density modulation that is also strongly coupled to a distortion of the 1T-TaS2 lattice, making it possible to use techniques based on x-ray diffraction.

In this study, we focus on fluctuations and rearrangement of these CDW domains under an applied electric field. Employing a combination of DC and AC electric field modulation, we probe the CDW order using X-ray Photon Correlation Spectroscopy (XPCS) at MHz frame rates at the European X-ray Free Electron Laser (Eu-XFEL), to study the characteristic timescale and determine the nature of their fluctuations (continuous or stochastic). Extracting accurate dynamical parameters from MHz frame rate detectors via frame-to-frame XPCS of weakly scattering peaks, such as those from a CDW system in a wide-angle scattering geometry is possible with analysis techniques that are adapted to extremely low per-shot signal rates. The analysis pipeline includes filtering of incident pulses based on x-ray fluence, accounting for frame-to-frame incident mean photon count variations, weighting matrix to reduce signal to noise. Time and voltage dependent correlation coefficients derived using this approach span time ranges from hundreds of ns to tens of μs at a wide range of applied electric fields. Future experiments at XFELs and 4th generation synchrotrons will be used to further investigate the nature of these CDW domain reconfigurations in different regimes of CDW domain dynamics.

Our study advances our understanding of mesoscale phenomena in 1T-TaS2 and paves the way for using XPCS to investigate domain dynamics in diverse materials systems in wide-angle scattering geometries at high repetition rate XFELs.

39

Extending BCDI Into the High X-ray Energy Domain: Enabling Bulk Polycrystalline Characterization with Nanoscale Resolution

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Polycrystals are an essential class of materials. Their use in everyday applications is due to their unique grain structure and grain boundary phenomena that can be tailored for a broad range applications and conditions. The grain boundary phenomena can degrade or enhance the macroscopic properties and device performance, for example due to local ion and charge transport and dislocation motion1. It is therefore crucial to determine the interplay between grain boundaries and grains within a bulk system to enable the fine control of the grain boundary for tailor made material designs. Existing characterization techniques for bulk polycrystals, such as High Energy Diffraction Microscopy (HEDM)2, provide micron-scale maps of strain distributions of large grains, typically those larger than tens of microns. However, HEDM does not capture sub-micron details due to its spatial resolution (5 microns)2. This results in obscuring sub-micron features critical for understanding the interplay between small and large grains and their grain boundaries. Bragg Coherent Diffraction Imaging (BCDI) offers high spatial (30-50 nm) and strain resolutions (20^-4) critical for elucidating size-dependent and grain boundary properties and typically maps strain within individual nanocrystals (0.05-1 microns)3. Extending BCDI into the high energy regime needed to penetrate a bulk polycrystal system promises to reveal the sub-micron features outside of HEDM's resolution limit4. However, fulfilling the Nyquist sampling criterion, or the requirement that the diffraction pattern's mean speckle size be twice the detector's pixel size5, is challenging with HE-BCDI due to the compression of reciprocal space at high X-ray energies4. Moreover, large crystal volumes result in smaller fringe spacing on the detector, further exacerbating the challenge of sufficient sampling in these measurements5. Overall, HE-BCDI presents an undersampled regime with overbinned diffraction fringes that are unsuitable for conventional BCDI phase retrieval4.

Fourth generation synchrotron sources (ESRF & APS-U) have sufficient coherence capabilities in the high energy X-ray regime to enable HE-BCDI4 and present a ripe opportunity to develop alternative phase retrieval algorithms and data collection methodologies that complete the picture of the microstructure from bulk polycrystals. In this presentation we address the undersampling problem and demonstrate a successful recovery of the fringe structure obscured by coarse pixels in a high-energy diffraction experiment at a fourth-generation synchrotron source. Diffracting from a well-behaved particle in the overbinned fringe regime at fixed energy, we implement a series of sub-pixel translations in the detector plane that can reveal the overbinned fringe sub-pixel detail. We then evaluate the fidelity of this fringe recovery strategy by comparing the recovered fringe pattern to a fine fringe measurement in which fringes were fully resolved via a large sample-to-detector distance. This approach represents development towards enabling HE-BCDI that can help elucidate how sub-micron sized features influence large scale domains and their mechanical properties in a bulk polycrystal system.

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112

Unraveling the Three-Dimensional Structure of Intermuscular Bone in Atlantic herring Using X-ray Coherent Diffraction Imaging

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The stiffness and toughness of bone are determined by the organization and relationships between its main components (minerals and collagen). Intermuscular bone found in herring species presents a fascinating subject of study due to its absence of bone cells and simplified internal organization. Utilizing Small Angle X-ray scattering techniques, previous research has revealed that collagen fibrils in intermuscular bone assemble directly into bone, presenting a departure from the complex plywood-like structure observed in traditional bone. However, questions persist regarding the arrangement and orientation of collagen fibers within this seemingly simplistic structure.

In this study, we employed plane wave X-ray coherent diffraction imaging to delve into the three-dimensional architecture of low-mineralized intramuscular bone. Through the direct assembly of 27 diffraction patterns into a three-dimensional Fourier grid for phase recovery and tomography reconstruction, we achieved a remarkable resolution of 32.7nm based on the criterion 1/e of PRTF. Our findings unveil a layered composition of low-mineralized intramuscular bone, characterized by planar layers housing primarily aligned collagen fibrils intersecting at various angles. Notably, these distinct orientations of collagen fibers, evident in both diffraction patterns and three-dimensional reconstructions, are absent in high-mineralized fishbone samples.

We propose a method to analyze the data from two aspects of diffraction pattern and real space structure.The result show that the unique layered structure and diverse orientations of collagen bundles in low-mineralized intramuscular bone underlie its exceptional mechanical properties, offering insights into the biomechanics of fish bone. This study not only enriches our understanding of intramuscular bone structure but also holds implications for biomimetic material design and tissue engineering applications.

57

Soft X-ray Transmission Holography at ESRF

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The soft X-ray beamline at ESRF (ID32) provides X-rays in the energy range 400 eV – 1800 eV to perform polarization-dependent spectroscopic studies of magnetic and electronic properties of matter, with the main end-stations specialized in X-ray Resonant Inelastic Scattering (RIXS) and X-ray

Magnetic Circular Dichroism (XMCD) measurements. In addition, a compact side-station is dedicated to coherent small angle scattering experiments, in particular Fourier-transform holography on magnetic samples, with time-resolved and 3D magnetic holo-tomography capabilities.
The end-station is equipped with a fast scientific-grade fully in-vacuum sCMOS camera from Axis Photonique, provides a 0.4 T magnetic field along the beam direction and a combination of sample rotations and translations enabling a variety of holographic measurements to be performed, with or without beamstop.
Examples on studies of magnetic vortices, different domain wall structures and 3D magnetic holo-tomography together with a detailed description of the experimental setup will be given.
The Extremely Brilliant Source (ESRF upgrade phase 2) has boosted the partial degree of coherence of the ID32 source, theoretically approaching 50%, satisfactorily preserved throughout the numerous beamline optical elements. The examples listed above will illustrate how challenging coherent scattering experiments on magnetic materials can be easily performed on a beamline initially optimized for magnetism rather than coherence.

15

High resolution strain measurements in highly disordered materials

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The ability to measure small deformations or strains is useful for understanding many aspects of materials especially in soft condensed matter systems. Systematic shifts of speckles arising from small angle x-ray coherent diffraction when analyzed enable flow patterns of particle in the elastomers to be inferred. This information is obtained from cross-correlations of speckle patterns. This speckle tracking technique measures strain patterns with a accuracy similar to X-ray single crystal measurements but in amorphous or highly disordered materials.

23

Three-dimensional identification of dislocation dynamics in perovskite oxide during exsolution

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Identifying the three-dimensional (3D) evolution of dislocations in nanostructures is increasingly crucial for various fields such as catalysis, energy, and electronics applications. Despite its significance, in-situ imaging of 3D dislocation dynamics has been limited to date due to their buried nature. In this study, we unveiled the dynamics of dislocations within perovskite host oxide nanoparticles during the exsolution process, wherein doped transition metals migrate and form catalytic

nanoparticles on the surface. By utilizing the retrieved phase information from Bragg coherent X-ray diffraction imaging (BCDI), we revealed the nucleation of dislocations starting within the bulk and propagating towards the surface. Further analysis of multiple dislocations demonstrated that these dynamics occur differently depending on the dislocation types. Combined with elementally resolved electron microscopy, a correlation between structural deformation and chemistry changes along dislocations was also identified. These results suggest that dislocations can act as energetically favored pathways for segregations, migration, and exsolution.
This work was supported by the National Research Foundation of Korea grant NRF-2021R1A3B1077076.

55

Hard X-ray split-delay system at Linac Coherent Light Source

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Over the last decade, hard x-ray split delay (HXRSD) optics have been designed and implemented at several X-ray Free Electron Laser (XFEL) facilities due to the high demand for multi x-ray pulse capabilities. At Linac Coherent Light Source (LCLS), we have designed and commissioned HXRSD systems, which are wavefront splitting and amplitude-splitting. Here we present a operation and performance of each HXRSD. We present examples of the experiment schemes that combine x-ray probe-probe, x-ray pump-probe with x-ray diffraction and absorption spectroscopy.

52

Effect of crowder molecules on the diffusive behavior of the protein ferritin studied via MHz XPCS at the European XFEL

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Understanding molecular processes in living cells is of great interest, not only to model and predict the dynamic properties such as intracellular transport, but also to develop future drug formulations for the pharmaceutical industry. In vitro experiments often focus on model systems, such as proteins in diluted solutions. However, molecular crowding takes place in living cells. In such highly concentrated environments, the protein dynamics deviate significantly from those in diluted systems due to increased molecular and hydrodynamic interactions modifying the collective diffusion constants D(q). Here, we study these collective dynamics in crowded protein systems on molecular length and microsecond time scales employing Megahertz X-ray photon correlation spectroscopy (MHz XPCS) at beamline MID of the European XFEL 1. We investigate model systems for crowded solutions consisting of the globular protein ferritin and different types and concentrations of crowder molecules. Our experiments reveal a strong dependence of the crowding agents on the q-dependence of the collective diffusion D(q). For small crowding agents we observe a pronounced decrease in D(q) for

increasing q-values which is linked to the predominantly repulsive interaction between the proteins and small crowder molecules such as sucrose and polyethylene glycol 400. However, a different picture emerges for larger crowder molecules such as the polysaccharides Ficoll and Dextran. Here we observe an almost constant behavior of D(q) at high concentrations which we attribute to increasing attractive interactions and increasing mean-field behavior of hydrodynamic interactions. In addition, modulations in the D(q) of dextran related to the molecular weight and concentration of the crowder molecules became apparent. This indicates a (transient) structure formation of the crowder molecules within the solution upon exceeding the overlap concentration threshold.
In summary, our experiments shed new light on the diffusive dynamics of proteins in crowded environments on molecular length scale. They reveal a strong dependence of the protein dynamics on both type and concentration of the crowding agents.

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82

Unveiling the Structure and Thermodynamics of Deeply Supercooled Glycerol-Water Microdroplets with Ultrafast X-ray Scattering

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The water liquid-liquid critical point hypothesis (LLCP) suggests the existence of two distinct liquid states in water, namely high- and low-density liquid (HDL, LDL), with an LLCP concealed in the deeply supercooled range at heightened pressures. The effects of liquid polymorphism on solvation and structural dynamics in organic aqueous solutions still remains unclear. In this study, a dilute solution of water and glycerol (3.2 mol%) was employed, serving as a prototypical system to explore the presence of the HDL/LDL states. By utilizing evaporative cooling of microdroplets along with ultrafast X-ray scattering, crystallization was successfully avoided down to a temperature of approximately 229 K. In addition to the experimental findings, molecular dynamics (MD) simulations were conducted across an extended temperature range, revealing increased fluctuations and the first structure factor peak of the mixture, as temperature decreased. The methodology introduced in this study opens up new possibilities for experimentally identifying an LLCP in aqueous solutions by adjusting the solute concentration.

References
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