

Coherence 2014 Program
Norris Center, Northwestern University
1999 Campus Drive, Evanston IL 60208

(August 26 version)

Tuesday September 2

2:30 PM Conference Welcome; Introduction to Venue

Organizers: Paul Fuoss, Chris Jacobsen, Brian Stephenson

Opening Session

2:50 PM **Invited: Think Fourier: Structure and Dynamics Revealed with Coherent Beams**

Chair: Gerhard Gruebel
Henry Chapman (CFEL)

3:50 PM Mapping Chemical Composition at Nanometer Resolution by Soft X-ray Ptychography

David Shapiro (ALS)

4:10 PM Reticulated Nanostructures in Calcareous Biocrystals Imaged by 3D Bragg Ptychography

Francesca Mastropietro (Inst. Fresnel, CNRS, Aix Marseille U.)

4:30 PM Break – Louis Room

Imaging I

4:50 PM **Invited: (title TBD)**

Chair: Binhua Lin
Christian Schroer (TU Dresden)
Bastian Pfau (Lund U.)

5:30 PM Time-Resolved X-ray Imaging of a Magnetic Bubble's Gyrotropic Motion

5:50 PM Coherent Bragg Imaging of the Complex Configuration of Inversion Domain Boundaries in GaN Nanowires

Stephane Labat (IM2NP, CNRS, Aix Marseille U.)

6:10 PM **End; free time to explore Evanston**

Wednesday September 3

XPCS I

8:30 AM **Keynote: XPCS: Beyond Simple Time Correlations**

Chair: Anders Madsen
Mark Sutton (McGill U.)
Qingteng Zhang (U. Wisconsin)

9:30 AM Equilibrium Dynamics of Serpentine Striped Domains in a PbTiO₃/SrTiO₃ Superlattice

9:50 AM Charge Induced Dynamics of the Ag (001) Surface

Robert Karl (Rochester Inst. Tech.)
Sylvain Ravy (SOLEIL)

10:10 AM Four-Order Correlation Function as Revealed by Speckle Patterns

10:30 AM Break – Louis Room

Ultrafast Imaging

10:45 AM **Invited: Imaging Dynamics at the Nano- and Meso-scale**

Chair: Oleg Shpyrko
Jesse Clark (Stanford U.)
Rui Xu (UCLA)

11:25 AM Single-Shot Three-Dimensional Structure Determination of Nanocrystals with Femtosecond X-ray Free Electron Laser Pulses

11:45 AM Coherent Imaging of Ultrafast Magnetization Dynamics

Clemens von Korff Schmising (TU Berlin)

12:05 PM Conference photo – then Lunch – Louis Room

Imaging II

1:20 PM **Invited: Recent Progress of Hard X-ray Ptychography at SPring-8**

Chair: Johanna Nelson Weker
Yukio Takahashi (Osaka U.)

2:00 PM Investigation of the Diffusion of Copper into a Gold Nanocrystal by CDI

Ana Katrina Estandarte (UC London)

2:20 PM Strain Imaging of an InP Nanostructured Thin Film by 3D X-ray Bragg Ptychography

Anastasios Pateras (Inst. Fresnel, CNRS, Aix-Marseille U.)

2:40 PM	Break – Louis Room	
	Theory and Computation	Chair: James Fienup
2:55 PM	Keynote: 3D ptychography – Algorithms, challenges, and applications	Manuel Guizar-Sicairos (PSI)
3:55 PM	Signature of Dislocations and Stacking Faults of FCC Nanocrystals in Coherent X-ray Diffraction Patterns	Maxime Dupraz (SIMaP Grenoble-INP)
4:15 PM	Homeopathic Imaging: What Happens When We Look at Really Small Objects	Kartik Ayyer (Cornell U.)
4:35 PM	Single Distance Phase-Contrast Tomography without Need for Additional Constraints	Aike Ruhlandt (Georg-August U.)
4:55 PM	Poster Session I (with refreshments and finger-food) – Louis Room	
6:30 PM	Poster session ends; free time to explore Evanston	

Thursday September 4

	New Sources	Chair: Tetsuya Ishikawa
8:30 AM	Invited: X-ray Free-Electron Laser Sources and Coherent X-ray Science	Massimo Altarelli (European XFEL)
9:10 AM	CDI with High Photon Flux Table-Top XUV and Soft X-ray Sources	Jan Rothhardt (Helmholtz Institute Jena)
9:30 AM	Invited: MAX IV: More Coherence!	Christoph Quitmann (MAX IV)
10:10 AM	Break – Louis Room	
	Imaging III	Chair: Mike Toney
10:25 AM	Invited: Visualizing Lattice Structure in Thin Films with New 2D and 3D Bragg Ptychography Approaches	Stephan Hruszkewycz (Argonne)
11:05 AM	Coherent Diffraction Imaging of Metallic Core-Shell Nanowires	Marie-Ingrid Richard (IM2NP, CNRS, Aix-Marseille U.)
11:25 AM	Topological Defects and Phase Transformations in Operando Battery Nanoparticles	Andrew Ulvestad (UCSD)
11:45 PM	Lunch – Louis Room	
	XPCS II	Chair: Yuya Shinohara
1:10 PM	Invited: Atomic Motion and Physical Aging in Structural Glasses	Beatrice Ruta (ESRF)
1:50 PM	X-ray Correlation Spectroscopy Instrument at the Linac Coherent Light Source	Marcin Sikorski (LCLS)
2:10 PM	Hard X-ray Delay Line for X-ray Photon Correlation Spectroscopy	Wojciech Roseker (DESY)
2:30 PM	Break – Louis Room	
	Advances in Instruments and Methods	Chair: Steve Kevan
2:45 PM	Invited: X-ray Detectors for New Photon Science Facilities: Are They the Remaining Experimental Limitation?	Gabriella Carini (SLAC)
3:25 PM	A Method for Ptychographic X-ray Imaging of Weak-Phase Objects	Akihiro Suzuki (Osaka U.)
3:45 PM	High-Resolution Imaging with Chemical Contrast using Hard X-ray Ptychography	Juliane Reinhardt (DESY)
4:05 PM	Partially Coherent Wavefront Propagation Simulations: from Beamline Performance to Virtual Experiments	Lutz Wiegart (Brookhaven)

4:25 PM Poster Session II (with refreshments) – Louis Room

6:00 PM Transportation to Banquet

6:30 PM Banquet

9:30 PM Transportation to Hotel

Friday September 5

Imaging for Life Sciences

8:30 AM **Invited: Opportunities for Coherent X-ray Imaging of Biological Samples**

Chair: Christian Gutt

Felisa Berenguer (UC London)

9:10 AM Imaging Living Cells in a Beam of Cyanobacteria with an X-ray Laser

Gijs van der Schot (Uppsala U.)

9:30 AM Simultaneous Cryo X-ray Ptychography and Fluorescence Imaging

Junjing Deng (Northwestern U.)

9:50 AM Ptychographic X-ray Nanotomography as a Tool for Biomedical Research

Martin Dierolf (TU Munchen)

10:10 AM Break – Louis Room

XPCS III

10:25 AM **Invited: Jamming and Icy Behavior in Magnetic Systems with Exotic Spin Texture**

Chair: Aymeric Robert

Sujoy Roy (ALS)

11:05 AM X-ray Speckle Visibility and XPCS Study of Diffusion in Concentrated Alpha Crystallin Protein Suspensions

Nuwan Karunaratne (Northern Illinois U.)

11:25 AM Anisotropic Dynamical Heterogeneity in Colloidal Glass

Yuriy Chushkin (ESRF)

11:45 AM **Conference Closing**

Organizers

12:00 PM **Adjourn**

We suggest you leave luggage at the Orrington Hotel, and reserve taxis to airports with the Orrington Hotel concierge. However, we will have a locked storage room available at Norris Center on Friday morning.

Equilibrium Dynamics of Serpentine Striped Domains in a $\text{PbTiO}_3/\text{SrTiO}_3$ Superlattice

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$\text{PbTiO}_3/\text{SrTiO}_3$ superlattices consisting of alternating ferroelectric PbTiO_3 layers and dielectric SrTiO_3 layers at room temperature form disordered striped domains to minimize the electrostatic energy. The domains have a uniform width but random orientation and short in-plane coherence length of only a few domain periods. Coherent x-ray scattering from disordered domains exhibits a speckle pattern that corresponds to the spatial frequency spectrum of the domain configuration within the region illuminated by the focused x-ray beam. Changes in the real-space domain pattern lead to decorrelation of the speckle patterns in reciprocal space. We measured the temporal decorrelation of the coherent scattering patterns from disordered striped domains at room temperature for two $8\text{PbTiO}_3 / 3\text{SrTiO}_3$ superlattice samples. The correlation between the scattering patterns decreases as a function of elapsed time for both samples. For the first sample, the temporal decorrelation can be well described using a compressed exponential function, yielding a characteristic time that ranges from 1340 s to 1890 s among different spatial locations. Such decorrelation is commonly observed in soft material systems where metastable equilibrium states are formed as a result of disordered spatial structures and the system fluctuates among these metastable states under the influence of thermal energy. For the second sample, the decorrelation is slower and the characteristic time is approximated to be around 6000 s using the same compressed exponential function model. We hypothesize that the difference between the fluctuation timescales is likely caused by the structural defects during the fabrication process.

This work is supported by US DOE under Grant No. DE-FG02-10ER46147. Work at ANL is supported by DOE Office of Science and Office of Basic Energy Sciences under Contract No. DE-AC02-06CH11357. Work at Stony Brook University is supported by US NSF DMR1055413.

Single-Shot Three-Dimensional Structure Determination of Nanocrystals with Femtosecond X-ray Free Electron Laser Pulses

Rui Xu¹, Huaidong Jiang², Changyong Song³, Jose A. Rodriguez⁴, Zhifeng Huang¹, Chien-Chun Chen¹, Daewoong Nam^{3,5}, Jaehyun Park³, Marcus Gallagher-Jones^{3,6}, Sangsoo Kim³, Sunam Kim³, Akihiro Suzuki⁷, Yuki Takayama^{3,8}, Tomotaka Oroguchi^{3,8}, Yukio Takahashi⁷, Jiadong Fan², Yunfei Zou¹, Takaki Hatsui³, Yuichi Inubushi³, Takashi Kameshima⁹, Koji Yonekura³, Kensuke Tono⁹, Tadashi Togashi⁹, Takahiro Sato³, Masaki Yamamoto³, Masayoshi Nakasako^{3,8}, Makina Yabashi³, Tetsuya Ishikawa³ and Jianwei Miao¹

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Coherent diffraction imaging (CDI) using synchrotron radiation, X-ray free-electron lasers (XFELs), high harmonic generation, soft X-ray lasers, optical lasers and electrons is a rapidly growing area of research that has found applications across several disciplines. One of the major potential applications of CDI is to determine the three-dimensional (3D) structure of single particles at atomic resolution on few-femtosecond timescales. Recent experiments have confirmed that the two-dimensional (2D) diffraction pattern of a single particle can be recorded and reconstructed from a single XFEL pulse before the onset of radiation damage. However, although a method termed ankylography was recently proposed to obtain 3D structure from a single sample orientation, single-shot 3D imaging has not been experimentally realized as yet. Here we report the experimental demonstration of single-shot 3D structure determination of an object; in this case, individual gold nanocrystals at ~5.5 nm resolution using ~10 fs XFEL pulses. Coherent diffraction patterns are collected from high-index-faceted nanocrystals, each struck by an XFEL pulse. Taking advantage of the symmetry of the nanocrystal and the curvature of the Ewald sphere, we reconstruct the 3D structure of each nanocrystal from a single-shot diffraction pattern. Furthermore, the single-shot diffraction patterns of identical nanocrystals were selected to assemble a 3D diffraction pattern, from which an average 3D reconstruction was achieved with a resolution presently limited by the detector geometry. By averaging a sufficient number of identical nanocrystals, this method may be used to determine the 3D structure of nanocrystals at atomic resolution. As symmetry exists in many virus particles, this method may also be applied to 3D structure studies of such particles at nanometer resolution on femtosecond time scales.

References

1. Miao, J. *et al.* Extending the methodology of X-ray crystallography to allow imaging of micrometre-sized non-crystalline specimens. *Nature* **400**, 342-344 (1999).
2. Raines, K. S. *et al.* Three-dimensional structure determination from a single view. *Nature* **463**, 214-217 (2010).
3. Xu, R. *et al.* Single-shot three-dimensional structure determination of nanocrystals with femtosecond X-ray free-electron laser pulses. *Nature Commun.* **5**, 4061 (2014).

Partially Coherent Wavefront Propagation Simulations: from Beamline Performance to Virtual Experiments

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We have applied fully- and partially-coherent synchrotron radiation wavefront propagation simulations, implemented in the "Synchrotron Radiation Workshop" (SRW) computer code [1], to simulate the performance of the Coherent Hard X-ray (CHX) scattering beamline [2] currently under construction at NSLS-II. The beamline needs to provide the highest possible flux with moderate coherence for measurements of fast dynamics via X-ray Photon Correlation Spectroscopy (XPCS), while Coherent Diffraction Imaging applications require a higher degree of coherence and can cope with a reduced flux. The beamline performance has been simulated, taking real performance data from optical elements, e.g. mirror metrology, into account, and quantified in terms of photon flux and mutual intensities. Electron density maps of colloidal suspensions and biological cells were used to simulate realistic scattering pattern as the CHX beamline could provide upon commissioning in XPCS and full field CDI experiments. We show the potential of partially-coherent wavefront propagation simulations from beamline and experiment optimization to data analysis.

This work has been supported by US DOE, Contract No. DE-AC02-98CH10886.

References

1. O. Chubar, L. Berman, Y.S. Chu, A. Fluerasu, S. Hulbert, M. Idir, K. Kaznatcheev, D. Shapiro, Q. Shen, J. Baltser, *Proc. of SPIE*, **8141**, 814107 (2011).
2. A. Fluerasu, O. Chubar, K. Kaznatcheev, J. Baltser, L. Wiegart, K. Evans-Lutterodt, M. Carlucci-Dayton, L. Berman, *Proc. of SPIE*, **8141** (2011)

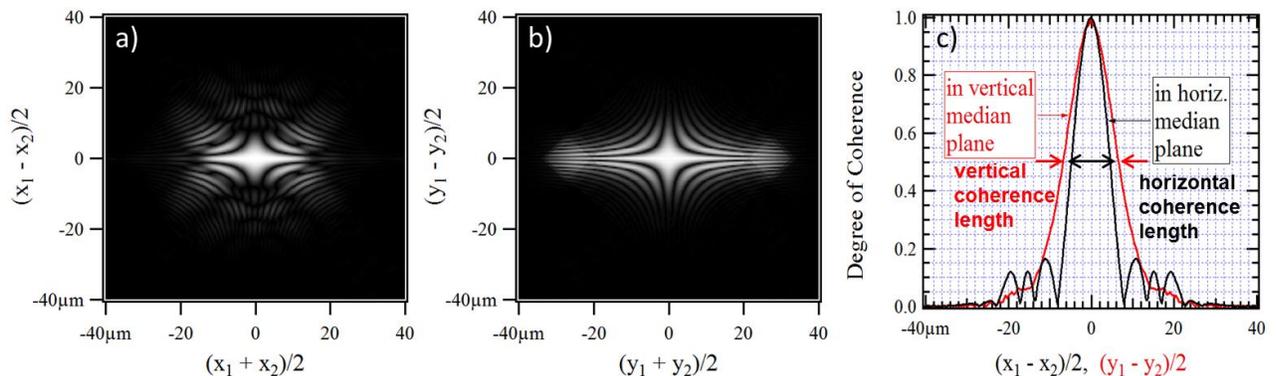


Figure 1: Examples of mutual intensity calculations obtained by partially coherent wavefront propagations at the sample position of the CHX beamline. Degree of coherence in the a) horizontal median plane, b) in the vertical median plane. c) cuts through a) and b) show the horizontal and vertical coherence lengths.

Imaging living cells in a beam of cyanobacteria with an X-ray laser

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Imaging live cells at a resolution higher than achieved using optical microscopy is a challenge. Ultra-fast coherent diffractive imaging [1] with X-ray free-electron lasers (XFELs) has the potential to achieve sub-nanometer resolution on micron-sized living cells [2]. Our container-free injection method can introduce a beam of live cyanobacteria into the micron-sized focus of the Linac Coherent Light Source (LCLS) to record diffraction patterns from individual cells with very low noise at high hit rates. We used variants of the Gerchberg-Saxton-Fienup algorithms for phase retrieval [3-5] to produce two-dimensional projection images directly from the diffraction patterns. Synthetic X-ray Nomarski images [6], calculated from the complex-valued reconstructions, show cells in a similar manner to what one would expect to see using a Nomarski microscope, only at higher resolution than currently available. In a first experiment, we collected diffraction patterns to 33-46 nm full-period resolution, and reconstructed the exit wave front to 82 nm full-period resolution. In a second experiment, we demonstrate that it is indeed possible to record diffraction data to nanometer resolution on live cells with an intense, ultra-short X-ray pulse as predicted earlier [2]. These results are encouraging, and future developments to the XFELs and improvements to the X-ray area-detectors is expected to bring sub-nanometer resolution reconstructions of living cells within reach.

We thank the Swedish Research Council, the Knut and Alice Wallenberg Foundation, the European Research Council, the Röntgen-Ångström Cluster, and Stiftelsen Olle Engkvist Byggmästare for supporting this work.

References

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2. Bergh, M. *et al.* *Q. Rev. Biophys.* **41**, 181-204 (2008).
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4. Fienup, J.R. *Opt. Lett.* **3**, 27-29 (1978).
5. Luke, D.R. *Inverse Problems* **21**, 37-50 (2005).
6. Paganin, D. *et al.* *J. Microsc.* **214**, 315-327 (2004).

Understanding *Operando* Battery Nanomechanics: Lensless Strain Mapping via *In-Situ* Coherent X-ray Diffractive Imaging

Lithium ion batteries are ubiquitous in mobile devices, increasingly used in transportation, and promising candidates for renewable energy integration into the electrical grid. To fulfill their powerful promise, electrodes with increased capacity, faster charge rates, and minimal capacity fade must be developed. Understanding the nanomechanics at the individual particle level is a crucial step in achieving these ambitious goals. Here we reveal three-dimensional strain evolution of a single $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_{4-\delta}$ nanoparticle in a coin cell battery under *operando* conditions during charge/discharge cycles with *in-situ* coherent x-ray diffractive imaging. We report the first direct *operando* observation of both stripe morphologies and coherency strain. Our results suggest the critical size for phase separation is 50 nm. We directly measure the elastic energetics of the phase transformation. Surprisingly, the characteristics of the femtojoule energy barrier depend on charge versus discharge. This approach opens a powerful new avenue for studying battery nanomechanics, phase transformations, and capacity fade under *operando* conditions in a wide range of electrochemical energy storage systems.

A method for ptychographic X-ray imaging of weak-phase objects

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Ptychography, which is a type of coherent diffraction imaging, can visualize both a transmission function of non-isolated samples and an illumination function [1]. At x-ray wavelengths, ptychography provides us high-resolution observation of buried nanostructures. Various experimental factors actually limits the quality of the reconstructed image. A limited dynamic range of a detector is one of prime considerations, especially when we observe weak-phase objects such as soft biological tissues. In this study, to reduce the required dynamic range of a detector, we propose an experimental geometry of ptychography using a reference scatterer (RS) and a beamstop (BS), and then evaluate its feasibility by a computer simulation.

Figure 1 shows a schematic view of the proposed ptychography. The RS is a cylindrical object placed just before a sample. The BS in front of a detector is used to block the strong intensities at low spatial frequencies, which facilitates collecting weakly high- q diffraction pattern within the limited dynamic range of the detector. In addition, the inherent problem of the BS, i.e., missing data at the center leads to low-frequency artefacts on the reconstructed image, is removed by inline holography using the RS. That is, the inline hologram compensates for the low-spatial-frequency diffraction data lost to the BS. The present method can reduce the dynamic range required for the high-resolution reconstruction of weak-phase objects in x-ray ptychography.

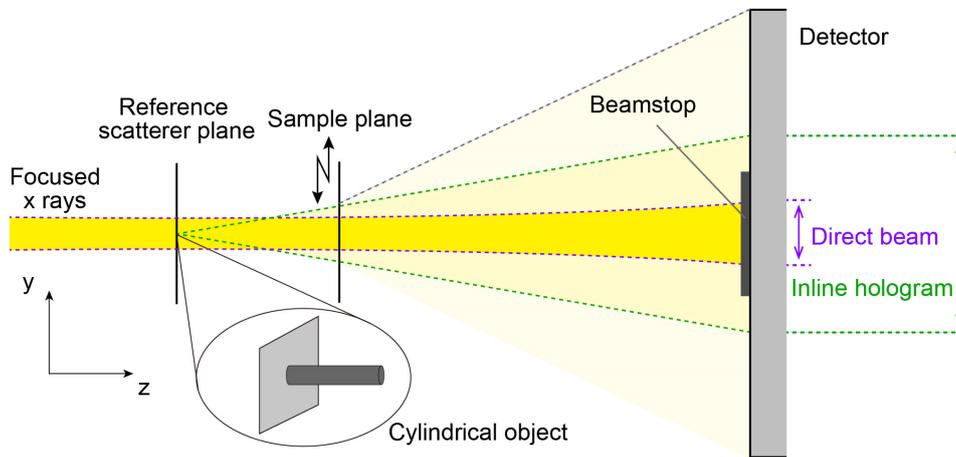


Figure 1: Schematic view of ptychography using a reference scatterer and a beamstop.

Reference

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X-ray Correlation Spectroscopy instrument at the Linac Coherent Light Source

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The X-ray Correlation Spectroscopy Instrument (XCS) is a hard x-ray experimental station operating at the Linac Coherent Light Source (LCLS), the world's first hard x-ray free electron laser. XCS takes full advantage of the unique properties of the LCLS beam (the unprecedented coherence and high flux) to probe dynamical phenomena in condensed matter down to nanometers length scales by means of X-ray Correlation Spectroscopy in various scattering geometries (SAXS, WAXS, Grazing Incidence). The XCS station was designed to enable studies of both slow (i.e. with a characteristic time scales larger than 10ms) in sequential (movie) mode as well as ultrafast dynamics (i.e. ranging between hundreds of femtoseconds up to several nanoseconds) in Split and Delay scheme. An overview of the outcome of the first experiments in the sequential mode will be presented. Impact of LCLS beam pointing jitter and beamline optics configuration on achievable contrast and the longest accessible time scales will be discussed.

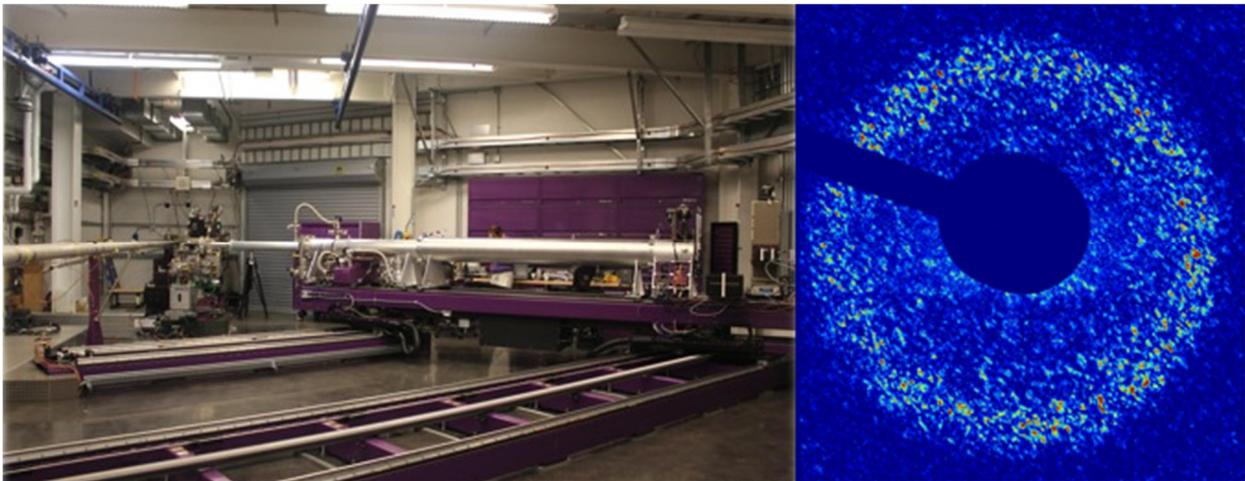


Figure 1: (left) large angle detector mover at the XCS instrument and (right) an example of a single shot speckle pattern collected from colloidal suspension in transmission geometry.

Mapping Chemical Composition at Nanometer Resolution by Soft X-ray Ptychography

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The Advanced Light Source has demonstrated the highest resolution x-ray microscopy ever achieved through the use of soft x-ray ptychography [1]. We demonstrate 3 nm resolution imaging a test object and quantify the resolution enhancement that comes with increased x-ray exposure and improved reconstruction algorithms. We apply our method to the study of lithium intercalation pathways in nano-particles of LiFePO_4 , a material of broad technological and fundamental interest in electrochemical energy storage. Chemical components calculated from the full complex refractive index show enhanced chemical contrast which elucidates a strong correlation between structural defects and chemical phase propagation which is not accessible by other means. Our ptychographic microscope achieves a spatial resolution in chemical component map which is far superior to state-of-the-art conventional microscopes. Soft x-ray ptychography at the new COSMIC beamline is set to revolutionize the study of chemistry in mesoscale systems.

The Advanced Light Source is supported by the Director, Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

References

1. D.A. Shapiro *et al*, *Nature Photonics*, submitted (2014).

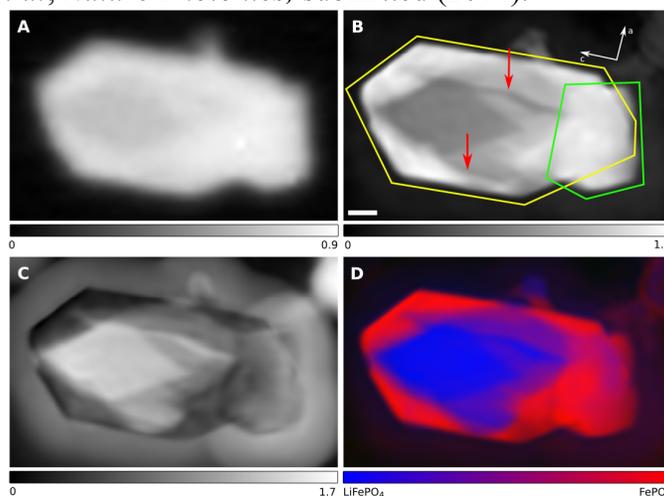


Figure 1: Soft x-ray spectro-ptychography of partially delithiated LiFePO_4 . (a) Standard STXM image with a 25 nm optic at 710 eV showing maximum contrast between FePO_4 and LiFePO_4 . (b) Ptychography absorption image at 710 eV using a 60 nm optic. (c) Ptychography phase image at 709.2 eV (d) Ptychographic chemical mapping using a full energy image stack [1].

Single distance phase-contrast tomography without need for additional constraints

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The main challenge of propagation-based (holographic) x-ray phase-contrast tomography is the phase-retrieval step based on intensity measurements in the detection plane. All common techniques require one or several assumptions about the object or additional measurements, for example at different object to detector distances. For tomography, single distance images for different projection angles of the object are necessarily available. Because of the finite size of the object, these projections are not independent of each other.

We show that this consistency can be used to overcome the need for restrictive additional a priori knowledge about the object by combining the phase-retrieval and tomographic reconstruction as shown in Fig. 1. In particular, no assumptions either on the phase-shifting and absorption properties of the object or on the support are necessary. A first investigation with simulated and noisy data proved the robustness of the algorithm. Subsequently, we have also demonstrated excellent reconstructions for experimental data. With the presented approach, x-ray phase-contrast propagation tomography is now equipped with an algorithm to treat images recorded at a single detector distance, and for the most general case of objects.

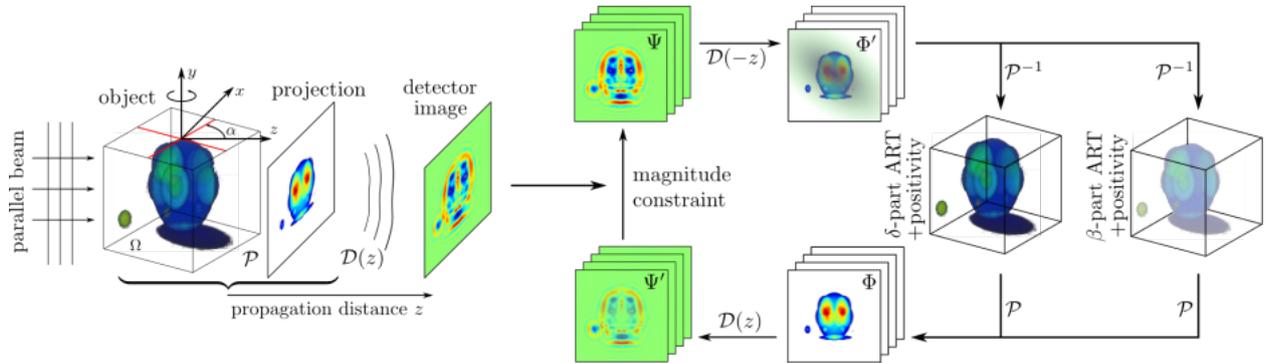


Figure 1: Left: experimental setup. Right: The detector images for all object angles are numerically propagated ($\mathcal{D}(-z)$) to the object plane. There, all images are split into a delta and beta part, as in the complex index of refraction $n=1-\delta + i\beta$. For x-rays, both delta and beta are positive values, thus a positivity constraint can be applied. Both parts are individually reconstructed to three dimensional objects using a modified ART algorithm, guaranteeing the tomographic consistency. The 3d objects are reprojected (\mathcal{P}) in the next step and numerically propagated (\mathcal{D}) to the detector plane, where the magnitude constraint is applied again. The procedure is iterated several times.

CDI with high photon flux table-top XUV and soft X-ray sources

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Laser driven table-top sources of coherent XUV radiation based on high harmonic generation (HHG) have seen enormous progress during recent years. Complementary to large scale facilities (synchrotrons and free-electron lasers) these high harmonic (HH) sources provide high accessibility, good coherence properties but also femtosecond and even attosecond pulse durations for imaging ultrafast processes in life-, material- and fundamental science. Although, table-top coherent diffraction imaging (CDI) demonstrated 22 nm resolution and 3D imaging recently [1] the experiments suffer from the limited photon flux, which is mainly determined by the output power of the driving laser. In this contribution we present first CDI experiments which make use of novel high photon flux HH sources based on femtosecond fiber laser technology which is capable of providing orders of magnitude higher laser output power and consequently higher HH photon flux. In first experiments, we demonstrated sub-100 nm resolution CDI with 33 nm radiation in quasi-real time (1 s integration time). Furthermore, due to the excellent spatial beam quality and narrow bandwidth of the HH radiation, high numerical aperture (NA=0.8) diffraction patterns have been successfully recorded, which allow for reconstructions with a resolution close to one wavelength. Additionally, we present the latest advances in HH source development. More than 10^{13} photons/s per harmonic have been obtained at ~ 40 nm, which corresponds to >100 μW of average power. Furthermore, soft X-rays down to the water window (>283 eV photon energy) have been generated, which hold promise for time-resolved element-specific imaging of magnetic, organic and biological samples with a few nanometer resolution.

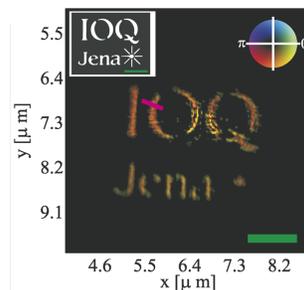


Figure 1: Reconstructed image of an institute logo. The green scale bar is one micron long.

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Hard X-ray delay line for X-ray Photon Correlation Spectroscopy

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Probing matter on time scales ranging from femtoseconds to nanoseconds is a key topic at the X-ray Free Electron Laser sources. The accessible time windows are however compromised by the intrinsic time structure of the FEL source. One way to overcome this limitation is the usage of a delay line [1] in so called Split- Pulse XPCS [2]. The performance of the device has been verified at the XPP and XCS instruments of LCLS [3]. The measured throughput of the device within $1.47e-5$ energy bandwidth of the exit beam at 7.9 keV is 30%. The coherence properties of single FEL pulses passing through the delay line were investigated by analyzing speckle patterns from a static scatterer (see Figure 1). A high speckle contrast of 69% was found, indicating the feasibility of performing coherence based experiments with the delay line. Temporal properties measured with the device indicate single and double temporal modes in the FEL spectrum.

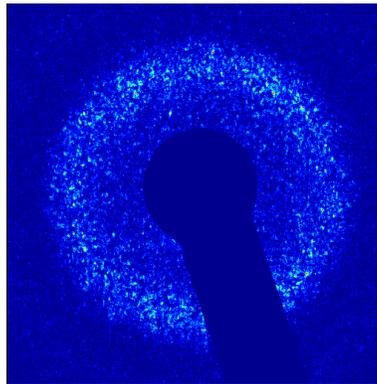


Figure 1: Single shot speckle pattern of dried PMMA nanoparticles recorded with a direct illumination CCD camera.

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Coherent diffraction imaging of metallic core-shell nanowires

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In recent years, low-dimensional materials have attracted a lot of attention because of the expected influence of size on the chemical and physical properties of the nanostructures. Large stress gradients and reduced vacancy concentrations in nano-objects may modify diffusion coefficients. It is thus interesting to investigate interdiffusion in nano- core-shell structures made from fully miscible elements. Characterizing the structural properties (strain gradients, chemical composition, crystal orientation and defects) inside these nanostructures is, however, a grand challenge in materials science. Coherent diffraction imaging (CDI) in Bragg condition is a promising and attractive method to map the shape, concentration and deformation fields simultaneously inside *single* nanostructures (Figure 1) [1]. The method even permits *in situ* experiments owing to its non-destructive nature. Indeed, during an *in situ* annealing experiment, we have demonstrated that core-shell morphology was preserved in Ag/Au nanowire at temperatures that are reported to lead to significant intermixing by volume diffusion in bulk material. Under these conditions, the rate of intermixing in the nanowire was lower than expected from bulk diffusion [2]. This opens new pathways for state-of-the-art investigations on the coupling between chemical intermixing and stresses in nanomaterials.

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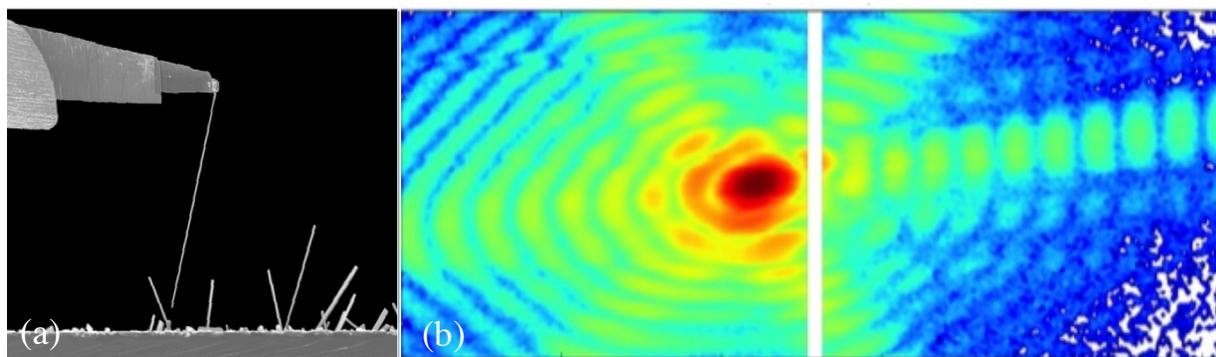


Figure 1: (a) Manipulator tip glued to a *single* Ag/Au core-shell nanowire and (b) CDI pattern of the (1-11) Bragg reflection of the nanowire.

High-Resolution Imaging with Chemical Contrast Using Hard X-Ray Ptychography

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Monitoring the chemical state of catalysts on a large range of length scales down to the nanometer scale is crucial to understand and improve their function under realistic working conditions. Since Ptychography is reaching unprecedented high spatial resolution [1] and can be combined with resonant scattering, it is a suitable tool for nanoanalysis and spectromicroscopy [2,3]. By recording a series of ptychograms at different energies around an absorption edge of an element of interest, the near-edge structure information in terms of absorption and phase shift is recorded with the same high spatial resolution as seen in the ptychographic reconstruction. Within our first experiment [4], we determined the chemical distribution of a weakly scattering model sample, containing metallic gold nanoparticles and platinum, with a spatial resolution of 20 nm in the hard x-ray regime, see Fig 1. Since the main limitation in reconstruction quality is given by the signal-to-noise ratio (SNR) in the diffraction pattern, the x-ray nanoscope at beamline P06 at PETRA III. was optimized, for instance by implementing beam stops. In this way, unwanted background signal due to air and other parasitic scattering effects, e.g. from the detector, have been reduced and the signal-to-noise ratio was increased. We improved the phase shift sensitivity and we pushed the spatial resolution well beyond 10 nm, see Fig. 2. The long-term aim is to investigate morphological and oxidation state variations of single nanoparticles in heterogeneous catalysts in-situ and in-operando with resonant Ptychography.

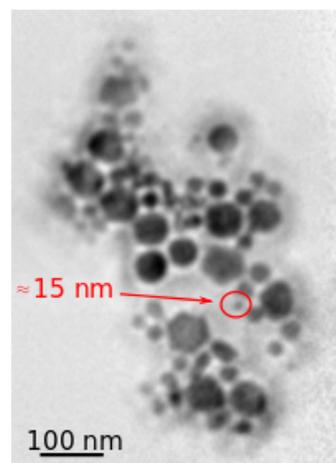
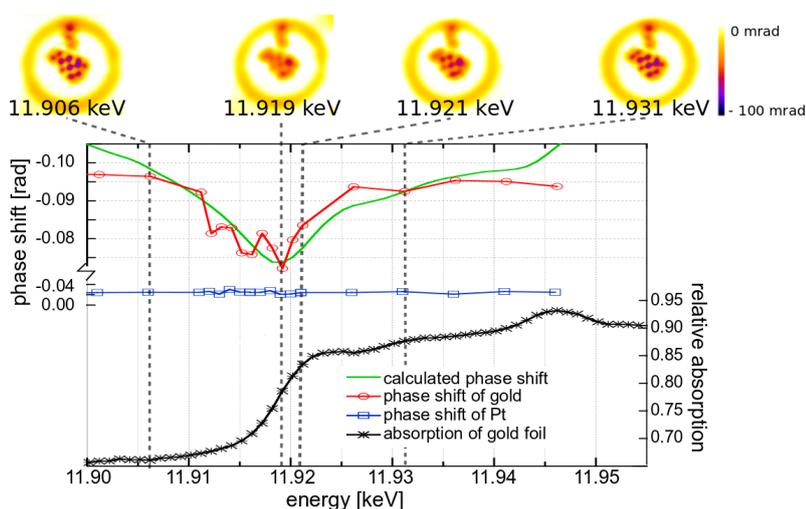


Figure 1: Phase reconstruction at selected energies with 20 nm resolution. The negative phase shift (red circles) of gold nanoparticles ($\varnothing < 100$ nm) decreases at the gold L₃ edge. The phase shift of the platinum ring (blue squares) remains constant.

Figure 2: High-Resolution phase reconstruction of 10 to 90 nm sized particles. (to be published)

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Four-order correlation function as revealed by speckle patterns

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Though phase retrieval methods seems to be the ultimate approach to structure determination, the complete data set it requires is intrinsically tedious and time-consuming to obtain, even by using new sources such as free electron lasers. New research approaches to get direct and quick information beyond the two point correlation functions are thus needed to optimize the use of X-ray coherent beams.

The study of the diffraction patterns of Rudin-Shapiro sequences allows one to give evidence that long-range order high-order correlation functions have a measurable effect on the speckle statistics [1].

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Time-resolved X-ray imaging of a magnetic bubble's gyrotropic motion

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Skyrmions are winding vector fields with a characteristic spherical topology. One example for such a vector field is the winding spin structure of a magnetic bubble in thin-film material with out-of-plane magnetization. Recent theoretical investigations predict a GHz gyrotropic motion of such a topological configuration after excitation in a restoring potential, analogous to vortex gyration. However, in contrast to vortices, bubbles are predicted to exhibit inertial effects, manifesting themselves in a second gyrotropic mode of reverse chirality and in additional degrees of freedom. Here, we present time-resolved images of the gyration of a magnetic bubble recorded using soft-X-ray Fourier-transform holography (FTH) [1]. The motion is triggered by a magnetic field pulse and the pump-probe image sequence is acquired with 50 ps time resolution using the single-bunch mode of the BESSY II storage ring. The microcoil to administer the magnetic field pulses has been lithographically integrated into the mask defining the FTH geometry [2]. From the images, we extracted the relaxation trajectory of the bubble with a position precision of 3 nm. We find that two eigenfrequencies are required to describe the bubble's motion leading to the conclusion that the bubble possesses a quasi-particle inertial mass. We attribute the comparatively large mass to the non-local energy reservoir of the bubble's breathing mode [3].

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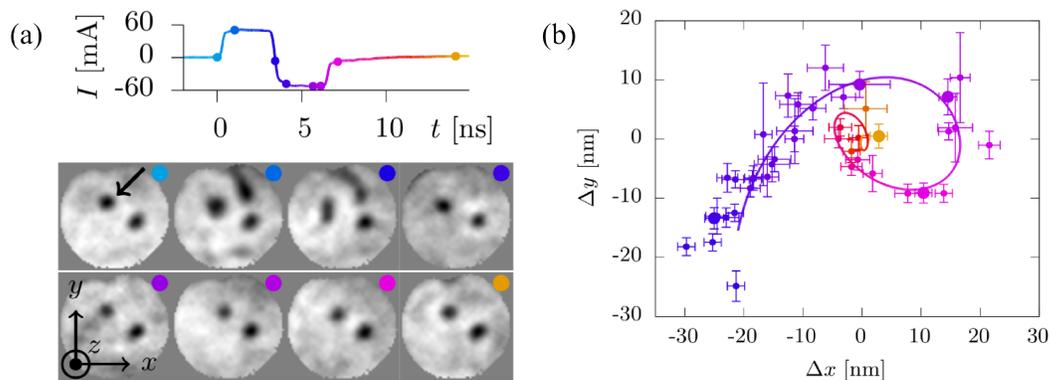


Figure 1: (a) Images of the bubble gyration (marked by the arrow) at time delays as indicated in the time course of the current generating the exciting field. (b) Trajectory of the bubble with a fit according to an analytical model.

Strain imaging of an InP nanostructured thin film by 3D X-Ray Bragg Ptychography

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In modern nanotechnology, strain is a fundamental parameter to manipulate crystalline material properties and performance. As light emission and optical isolation cannot be performed efficiently on Si-based materials, bonding III-V InP-based compounds on Si or Silicon-On-Insulator has been proposed for the designing of integrated devices emitting in the 1.55 μm wavelength domain [1]. Hence, quantification of the strain fields which develop inside the thin films due to the oxide bonding process is of importance for device applications. It requires measurement and visualization of these crystal lattice distortions to fully understand the link between structure and properties.

However, none of the available imaging techniques can provide simultaneously a 3D image of the crystalline properties with nanoscale resolution and a large field of view in a non-invasive way. Among the different available approaches, hard x-rays have the capability to probe strain in a non-destructive way. However, the lack of efficient lenses at these wavelengths is a strong limitation to the development of X-ray strain microscopy. This can be circumvented by the use of coherent X-rays, which allow preserving the phase information encoded in the scattered intensity [2]. In the recently demonstrated 3D Bragg ptychography approach [3, 4], the 3D image of the sample and its internal crystalline properties are retrieved with a resolution of a few tens of nm, without field of view restriction. In addition, it is expected to be robust towards the imaging of non-homogeneous strain field [5]. This approach exploits the redundancy of the data set obtained by measuring the intensity patterns at different but overlapping illuminated positions [6].

In this work, 3D Bragg ptychography is used to investigate the crystalline properties of an InP/InGaAs structured layer bounded onto a silicon wafer [7]. In addition to the infinite film geometry, the layered structure results in the development of non-homogeneous strain field. The experiment was performed at the ID13 beamline at ESRF with a monochromatic beam focused down to 100 nm. The InP (004) Bragg reflection was investigated and a 3D Bragg ptychography data set was acquired. Numerical analysis of the given problem was performed beforehand in order to optimize the inversion strategy and to study the possibility of introducing additional physical constraints through regularization approaches. The inversion of the experimental data was then successfully obtained. The retrieved 3D image of the sample exhibits the expected structural features, further confirmed by laboratory investigation methods (high-resolution X-ray diffraction and transmission electron microscopy). Additionally, it shows strong evidence of crystalline imperfections within the InP layer (e.g. tilted atomic planes). This result opens exciting perspectives for the imaging of complex nano-structured materials.

We thank the Erasmus Mundus Joint Doctorate Programme (EMJD) for financial support.

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Reticulated nanostructures in calcareous biocrystals imaged by 3D Bragg ptychography

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The calcareous structures in living organisms (e.g. shells, corals etc.) are built by crystalline mineralized units that exhibit species-specific morphologies both at the macro and micro-scales. However, studies at the sub-micrometric scale systematically reveal an ensemble of nodular single-crystalline sub-micrometric structures coated by a cortex made of amorphous calcium carbonate and organic residues [1,2]. The understanding of bio-mineralization, which relies on a detailed knowledge of the crystalline structure at the nanoscale, is a fundamental issue with promising perspectives for the development of new materials based on bio-minerals exhibiting superior structural (i. e. mechanical) properties. Lens-less x-ray microscopy approach in Bragg geometry [3] is a non-invasive imaging method allowing for the investigation of crystalline structure of a given crystal orientation and with a spatial resolution of 10-20 nm. The direct space structural information is obtained from coherently diffracted intensity measurements by phase retrieval inversion algorithms. Among these techniques, 3D Bragg ptychography offers the possibility to investigate spatially extended specimen and is therefore perfectly suited to tackle the bio-mineral problem. It is based on the acquisition of several 3D coherent diffraction patterns at different beam-to-sample positions. The large overlapping between two successive transversal positions of the beam onto the sample ensures the redundancy needed to solve the phase problem [4]. Regularization of the solution can be introduced in order to add weak pre-knowledge to the inverse problem [5].

Here we present the successful application of 3D Bragg ptychography for the imaging of a juvenile *Pinctada margaritifera* shell. The Bragg experiment has been performed in transmission geometry at the ID13 beamline (ESRF, Grenoble) profiting of the focused x-ray spot produced by the monochromatic and coherent illumination of a set of refractive lenses. The thickness of the very initial growing edge of the shell is natively in the order of 1 micron, ensuring coherent interferences along the beam propagation direction. The retrieved images show the finite extend of the crystalline coherence within the nodular structure with a resolution of about (40 nm)³.

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Coherent Bragg Imaging of the complex configuration of Inversion Domain Boundaries in GaN nanowires

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Although coherent diffraction imaging is able to determine the displacement field when working in Bragg condition [1], it has been mostly used for perfect crystal with very small strains [2,3]. Nevertheless, this technique has demonstrated recently its ability to evidence the displacement field in crystals containing defects such as dislocations [4]. In this work we report on the study of large displacement fields induced by several Inversion Domain Boundaries (IDBs) in GaN nanowires with a spatial resolution better than 10 nm and a displacement accuracy of a few picometers. It combines 5 Bragg peaks measured on the same nanowire with a 450 nm diameter. The measurements were done on the ID01 beamline at the ESRF (Grenoble-France) using coherent sub-micrometer beam focused with a Fresnel zone plate. The inversion of the 5 diffraction patterns using classical algorithms (Error Reduction, Hybrid Input-Output and Shrink Wrap) allows reconstructing the object with the full displacement vector field. 3 of the 5 reconstructions are shown in Fig.1. The displacement vector field determined in the nanowire is dramatically different from the one theoretically calculated for ideal planar IDBs [5]. This difference may have consequences on the physical properties of the nanowire.

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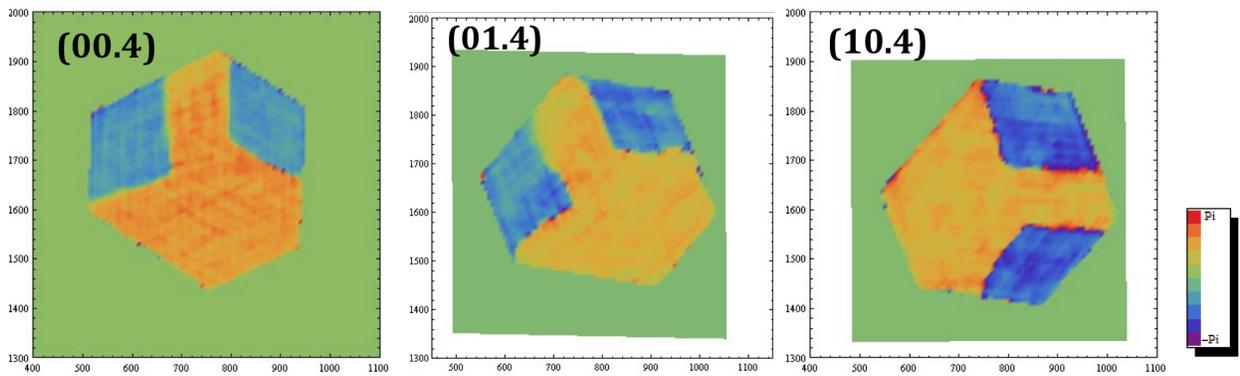


Figure 1: 2D phase map of the complex object reconstructed from 3 Bragg peaks. The in-plane scales are in nm. The phase jumps observed between the orange and the blue areas of the crystal are equal to 2.8 ± 0.1 radians for the 3 reflections.

Coherent Imaging of Ultrafast Magnetization Dynamics

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Ultrashort, coherent and circular polarized x-ray pulses of a free-electron laser are used for the first time to holographically image femtosecond magnetization dynamics within a magnetic domain pattern after creation of a localized optical excitation [1]. Magnetic domain images of a Co/Pd multilayer are recorded via Co M-edge x-ray magnetic circular dichroism at 58.9 eV, demonstrating a diffraction limited spatial resolution of < 70 nm and high quality reconstructions by transformation to in plane Fourier components [2]. Synchronized, near-infrared, femtosecond pump pulses are coupled to generate a spatially localized excitation via interference on the sample leading to a pronounced enhancement of the optical excitation in the upper part of the field of view. The time series of reconstructed images after the excitation shows a corresponding spatially confined bleaching of the magnetic contrast (cf. Figure 1 (a)-(d)) within a couple of hundred femtoseconds followed by a slight recovery on a longer time scale. The summed and normalized magnetic contrast of a 200×200 nm² area (cf. rectangle in Fig. 1b) is shown in Figure 1e) and follows a typical ultrafast magnetization transient. In addition evidence is found for concurrent changes in the spatial magnetization profile, suggesting the presence of ultrafast lateral transport of spin-polarized electrons on a nanometer length scale.

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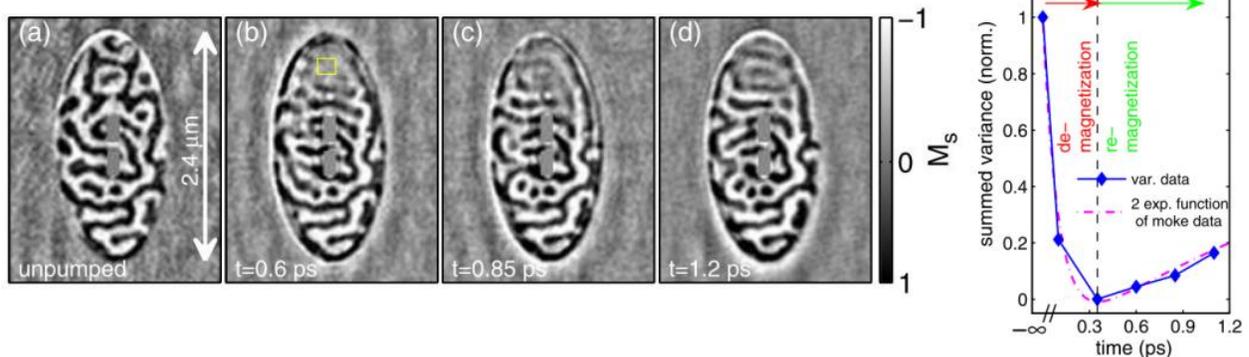


Figure 1: Reconstructions of the magnetic domains for unpumped (a) and pumped samples for selected time delays (b) – (d). The black and white areas within the field of view represent magnetic domains with magnetization M aligned either parallel or antiparallel to the incident x-ray beam direction. In the upper area of the elliptical object hole, the magnetic contrast is reduced. e) Summed and normalized magnetic contrast within yellow rectangle (cf. b) as a function of time showing typical ultrafast magnetization dynamics.

X-ray Speckle Visibility and XPCS Study of Diffusion in Concentrated Alpha Crystallin Protein Suspensions

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The lens of the mammalian eye is primarily composed of a concentrated suspension of proteins called crystallins, which comprise up to 70% of the total tissue mass. Alpha crystallin is the most common of these proteins and plays a crucial role in maintaining lens transparency. Changes in the inter-molecular interactions with aging of the eye are believed to be one of the primary causes of eye-diseases such as presbyopia and cataract. We have used X-ray photon correlation spectroscopy (XPCS) and Speckle Visibility Spectroscopy (SVS) to study the molecular scale dynamics of concentrated suspensions of alpha-crystalline in order to help understand the interactions within these concentrated suspensions.

In the now well established method of XPCS, the time correlation of the frame-to-frame intensity yields the dynamic structure factor of the system. Speckle visibility spectroscopy is similar to XPCS, but rather than correlating intensity between frames it measures the optical contrast within a single frame of fixed exposure time. This variation on XPCS can allow faster dynamics measurements, since it is not limited by the readout speed of the detector, but only by the shutter speed. In addition, we show how SVS can be extended to calculate spatial correlations between frames taken at different times, which yields information similar to XPCS, but with some possible advantages in terms of optimizing signal to noise [1]

XSVS measurements were performed on Alpha Crystallin for a range of volume fractions and at wavevectors corresponding to the peak in the protein-protein static structure factor ($Q=0.2 - 0.5 \text{ nm}^{-1}$). The resulting relaxation times are compared with predictions based on hard-sphere colloid theory. Measurements are also compared with dynamic laser light scattering (DLS) results at longer length scales. We have also carried out studies of the effect of x-ray damage on diffusion in concentrated protein suspensions, which should prove important for future studies.

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Charge Induced Dynamics of the Ag (001) Surface

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Using X-ray Photon Correlation Spectroscopy (XPCS), the rate of surface evolution due to the motion of atomic terraces and islands on Ag (001) in weak NaF electrolyte has been measured. For intermediate applied potentials between hydrogen evolution and oxidation the surface configuration completely changing on timescales of 10-100 seconds up to several 10^4 seconds for regions of largely static surface conditions. These dynamics, directly measured over large areas of the sample surface simultaneously, are precisely related to the amount of surface charge present at a given potential. Furthermore, specular x-ray scattering at different potentials reveals how the electron density at the Ag (001) - electrolyte interface depends upon the rate of surface evolution [1].

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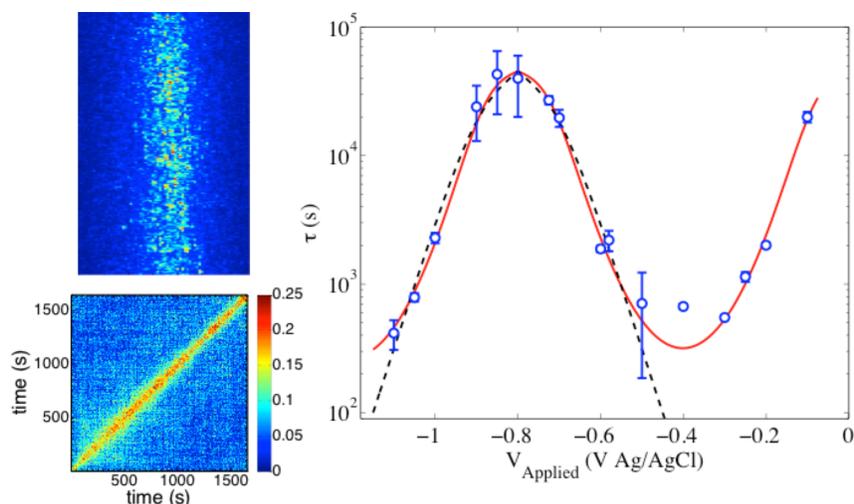


Figure 1: Speckle patterns are collected at an applied potential and used to calculate auto-correlations. The speckle patterns are sensitive to the arrangement of step-edges, terraces, and islands on the surface. The rate of evolution of these features is then determined as a function of applied potential. Interpretation of the potential dependence of the autocorrelation can be extrapolated through double-layer capacitance models.

Ptychographic tomography, advances and applications for high-resolution quantitative 3D imaging using hard X-rays

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Ptychographic tomography combines strengths of ptychography, such as high resolution and sensitivity, with the penetration of hard X-rays needed to study representative volumes in a non-destructive 3D measurement. Furthermore, it offers the possibility to measure the sample close to its native environment. Here we report on advances in instrumentation, acquisition strategy, reconstruction algorithms and image post-processing that allow us to use this technique routinely and to offer it to users, who are not necessarily ptychography experts, to address scientific questions. Examples of applications that exploit the advantages and unique information provided by ptychographic tomography will also be discussed.

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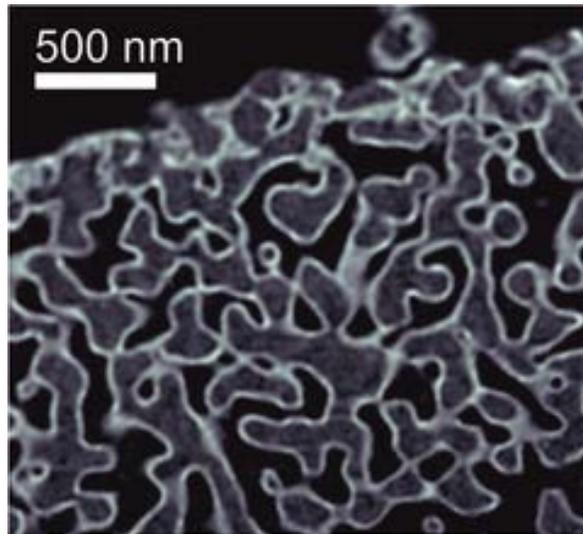


Figure 1: Coronal section of a tomogram on a test sample used to demonstrate 16 nm isotropic 3D resolution for ptychographic tomography [1]. Three distinct gray levels are visible for air (black), glass (gray), and a conformal Ta₂O₅ coating (white).

Investigation of the Diffusion of Copper into a Gold Nanocrystal by CDI

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Understanding how the diffusion process works for nanomaterials is of great importance for their stability and controlled synthesis. Diffusion studies by conventional methods only give rise to macroscopic information [1]; hence, there is a need for new techniques that allow probing of the diffusion process at the atomic scale. In this study, the strain sensitivity [2] of Coherent Diffraction Imaging (CDI) is utilized to investigate the diffusion of copper, as a function of time and temperature, into an individual gold nanocrystal. The CDI experiments were carried out at the I-07 beamline at the Diamond Light Source, UK. The sample was placed inside a UHV chamber and deposition of copper was achieved using a thermal evaporator. The structure of the gold nanocrystal was measured continuously at the (111) Bragg peak after copper deposition at various temperatures for several hours. It was observed that the total amplitude inside the nanocrystal decreased (Figure 1) as the diffusion of copper proceeded and 6 hours after copper deposition, the volume of the nanocrystal decreased to 52% of its original volume. This signifies the removal of the gold crystalline material as a result of alloy formation. Furthermore, the reconstructed phase images show that the gold nanocrystal was relatively unstrained before copper deposition and as the diffusion of copper proceeded, the nanocrystal became more strained signifying the lattice distortions within the nanocrystal induced by the diffusion process (Figure 1). The lattice distortions started at the edges of the nanocrystal where strain is initially present.

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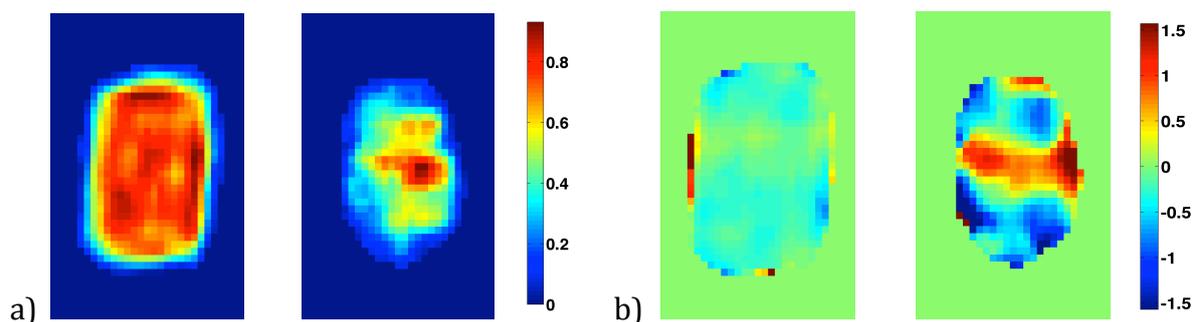


Figure 1: 2D slice at the center of the Au nanocrystal showing the a) reconstructed amplitude before Cu deposition (left) and 6 hours after Cu deposition (right), and b) reconstructed phase before Cu deposition (left) and 6 hours after Cu deposition (right) showing the presence of strain within the gold nanocrystal as a result of the diffusion process.

Signature of dislocations and stacking faults of *fcc* nanocrystals in coherent X-ray diffraction patterns

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Crystal defects induce strong distortions in diffraction patterns. A single defect alone can yield strong and fine features that are observed in high-resolution diffraction experiments such as coherent X-ray diffraction (CXD) [1][2]. The case of face-centred cubic nanocrystals is studied numerically and the signatures of typical defects close to Bragg positions are identified. Crystals of a few tens of nanometres are modelled with realistic atomic potentials [3] and 'relaxed' after introduction of well defined defects such as pure screw or edge dislocations, stacking faults or Frank and prismatic loops.

Diffraction patterns calculated in the kinematic approximation reveal various signatures of the defects depending on the Miller indices. They are strongly modified by the dissociation of the dislocations. We provide selection rules on the Miller indices to observe the maximum effect of given crystal defects, in the initial and relaxed configurations. The effect of several physical and geometrical parameters such as stacking fault energy, crystal shape and defect positions are discussed.

We demonstrate the unique character of the signature induced by a single defect and suggest a procedure based on the measurement of a few reflections to identify all the characteristics of a given single defect (Burgers vector, slip plane, size, position, interstitial or vacancy nature...) when it is alone in its structure.

The method is illustrated on a complex structure resulting from the simulated nanoindentation of a gold nanocrystal. Comparisons with coherent-X-ray diffraction patterns measured on GaN and gold nanowires demonstrate that the method is well suited to the live monitoring of the nucleation of defects and is very complementary to phase retrieval imaging methods [4].

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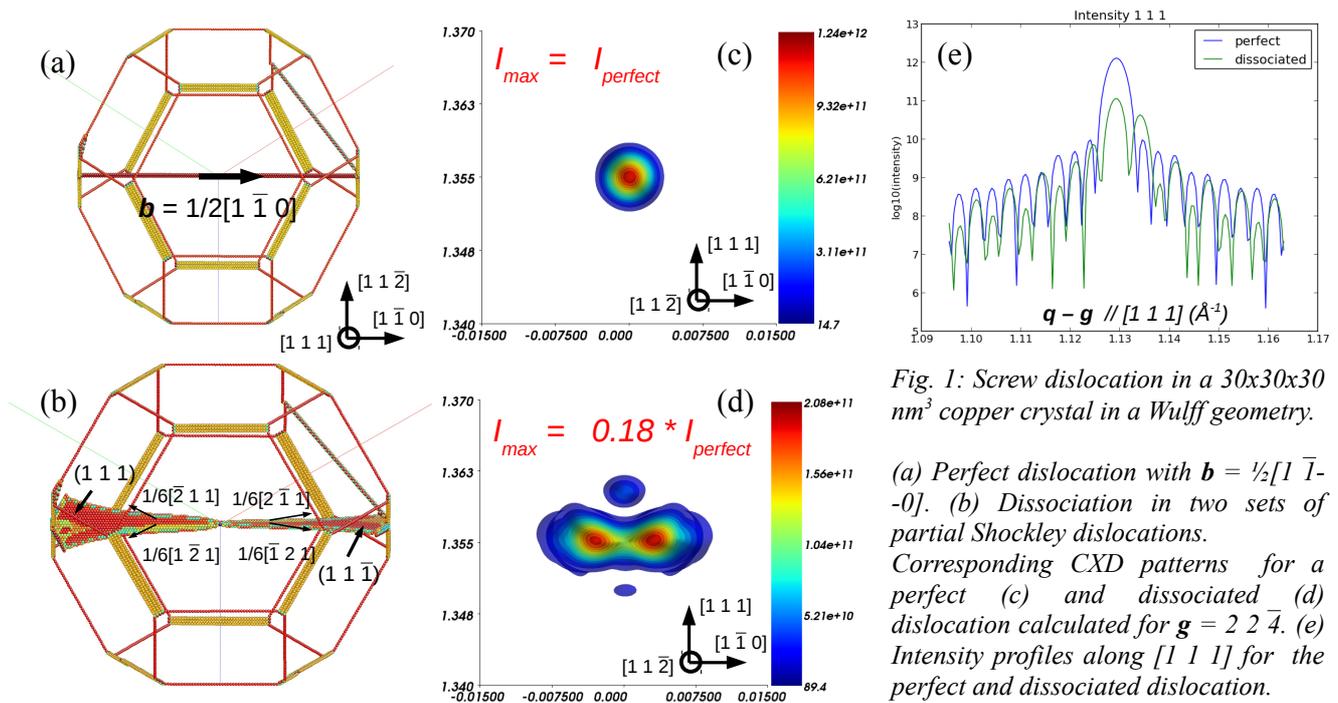


Fig. 1: Screw dislocation in a 30x30x30 nm³ copper crystal in a Wulff geometry.

(a) Perfect dislocation with $\mathbf{b} = \frac{1}{2}[1 \bar{1} 0]$. (b) Dissociation in two sets of partial Shockley dislocations. Corresponding CXD patterns for a perfect (c) and dissociated (d) dislocation calculated for $\mathbf{g} = 2 \bar{2} 4$. (e) Intensity profiles along $[1 1 1]$ for the perfect and dissociated dislocation.

Ptychographic X-ray nanotomography as a tool for biomedical research

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Since its first demonstration in 2010 [1], Ptychographic X-ray Computed Tomography (PXCT) has been successfully applied to a variety of scientific questions, including e.g. the hydration of silk fibers [2], the structure of a barrier marine coating [3], the characterization of carbon fibers [4], and the assessment of the densities of individual phases in hardened cement [5].

All these studies are based on the quantitative volumetric PXCT results with high spatial resolution and high density sensitivity which are obtained through the combination of ptychographically-reconstructed phase-contrast projections with computed tomography. Right from the beginning, these features of the technique have been identified as being invaluable for studies in a biomedical context, e.g. of mineralized tissues on the sub-micron level [1,6].

We will present recent PXCT results obtained on mineralized tissues like bones, teeth or deer antlers. Quantitative mapping of mineralization variations will be demonstrated, which can provide better understanding of the mutual interactions between embedded bone cells (osteocytes, see Fig. 1) and their surrounding matrix [7]. Furthermore, we will show the visualization of collagen fibril orientations inside the bone matrix, which reflect the function of the respective bone [8]. In addition to the results, we will also present advances in the processing of PXCT data, in particular concerning the steps from raw projection images to tomographic volumes.

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Simultaneous Cryo X-ray Ptychography and Fluorescence Imaging

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X-ray fluorescence microscopy (XFM) offers high sensitivity for quantitative mapping of elements in biological samples, while ptychography provides an alternative approach to visualize light-element biological ultrastructure beyond focusing-optic resolution limit [1]. Cryo offers excellent structural preservation and radiation damage resistance [2]. We describe here the combination of fluorescence and ptychography for imaging frozen-hydrated specimens at cryogenic temperature on the Bionanoprobe [3]. By this combination we have obtained a 20 nm resolution structural image and 90 nm resolution elemental images of a frozen-hydrated alga. The challenges in data analysis of this combined method is also discussed.

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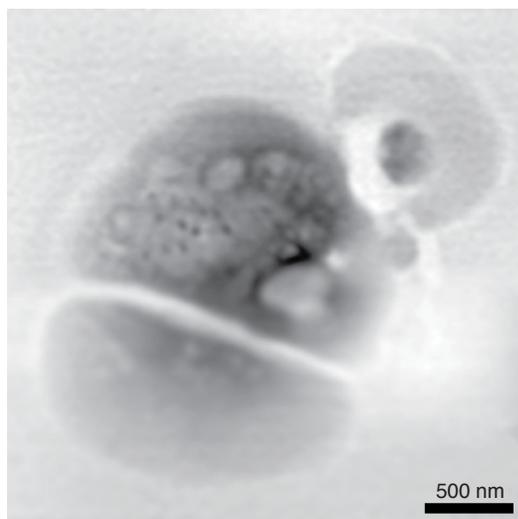


Figure 1: Ptychographic phase contrast image of a frozen-hydrated marine alga (*Ostreococcus* sp.) obtained at cryogenic temperatures.

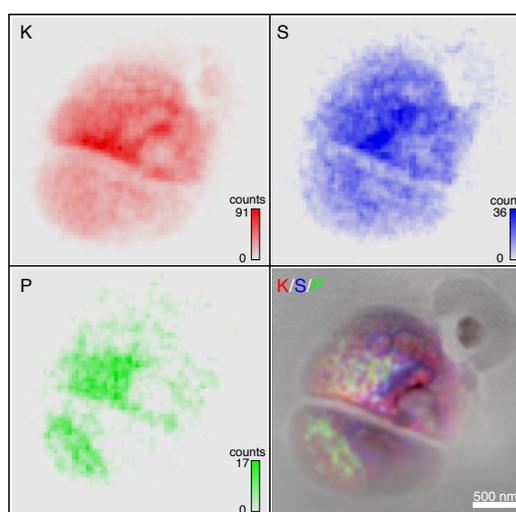


Figure 2: X-ray fluorescence images of the elements K, S, and P, along with their color composite overlay on the image of Fig. 1.

Anisotropic dynamical heterogeneity in colloidal glass

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Glassy solids and liquids are in disordered state but their dynamical behavior is remarkably different. The molecules diffuse freely in liquids but their motions in glasses become increasingly limited. The rapid slow down of the dynamics in liquids upon cooling is called a glass transition phenomenon. The idea that such slowing down is related to the growing cooperative length scale in the system dates back to Adam and Gibbs [1]. The structural relaxation in a deep supercooled state is possible through correlated motions of “blobs” of particles. Such cooperativity results in strong temporal fluctuations of the dynamics. Experimentally temporal heterogeneity is accessible by measuring the variance of the time-resolved correlation function [2]. Recently it was shown that glass transition is accompanied by growing dynamic length scale [3].

In this work we focus on the dynamical heterogeneity in a colloidal glass of magnetic nanoparticles with repulsive interparticle interactions. This dense charge-stabilized dispersion is a model system that replicates the behaviour of a real molecular glass-former. Under an applied magnetic field, the interparticle interactions remains repulsive however becomes anisotropic [4,5]. The investigation of the effect of the magnetic field on the dynamics shows that the field induces a moderate structural but a very strong dynamical anisotropy in the glassy system. It also causes a highly anisotropic cooperativity, almost two orders of magnitude larger in the field direction than in the perpendicular direction or in zero field. The origin of the observed anisotropic dynamics is discussed.

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Homeopathic Imaging: what happens when we look at really small objects

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As we try to image smaller and smaller particles, problems are encountered relating to low scattered intensity, relatively high background and inability to control the orientation of the object. Even for X-ray free electron lasers, things become difficult when we try to image single molecules. Each frame of data has so few photons that it seems impossible to decipher the orientation and hence to solve the structure. The EMC algorithm [1] provides a way out by collectively assigning orientations using Bayesian statistics.

We will discuss three proof-of-principle experiments which show how to analyze experiments that yield sparse data by using a highly attenuated tabletop source to generate very few scattered photons. Each progressively harder experiment encapsulates the key features of sparse data and unknown orientation. First, we will discuss 2D shadowgraphy [2], then 3D real-space tomography [3] and finally crystallography. The third experiment resembles serial crystallography except that the number of scattered photons per frame is too small to allow indexing.

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