Photonic Quantum Technologies – A Revolution in Communication, Sensing, and Metrology

764. WE-Heraeus-Seminar

17 Mar - 19 Mar 2022

Hybrid
at the Physikzentrum Bad Honnef/Germany

WILHELM UND ELSE
HERAEUS-STIFTUNG

Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

Aims and scope of the 762. WE-Heraeus-Seminar:

Accelerator based light sources like synchrotron radiation and Free-Electron-Laser (FEL) facilities have developed into indispensable analytical tools for modern science. They are capable of provide extremely intense and collimated beams of photons extending from the visible to the hardest X-ray regime. These facilities are used by scientist from many fields of science ranging from physics, chemistry, biology, medicine, materials, earth and environmental sciences, and various disciplines of engineering to cultural heritage. Meanwhile, a number ofscience fields depend completely on the availability of the analytical capabilities of these sources and the facilities in Germany alone welcome more than 10000 user visits per year with this number steadily increasing. During the last years accelerator based light sources have gone through a revolutionary phase. The extremely short and intense X-ray pulses of the recently established FELs allow for the investigation of the dynamic properties of matter at atomic length and time scales and thus opening a totally new observation window into matter at the nano and atomic scale. However, also in the field of storage ring-based sources a disruptive revolution is ongoing through the ongoing development of new magnetic storage ring lattices allowing for electron beams of very low emittance and, therefore, extremely bril-liant beam that can efficiently be focused down to the 10 nm scale for the spatial resolved investigation of any kind of matter on this length scale. This seminar will address the scientific opportunities opened by modern accelerator based light sources as well as the technical issues both on the side of the accelerators and of the experiments in order to develop this field even further.

Scientific Organizers:

Prof. Dr. Robert Feidenhans'l	European XFEL GmbH; Schenefeld, Germany E-mail: robert.feidenhansl@xfel.eu
Prof. Dr. Wim Leemans	DESY, Hamburg, Germany E-mail: wim.leemans@desy.de
Prof. Dr. Edgar Weckert	DESY, Hamburg, Germany E-mail: edgar.weckert@desy.de

Introduction

Administrative Organization:

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Registration:

Martina Albert (WE Heraeus Foundation)
at the Physikzentrum, reception office
Monday (10:00 h – 16:00 h) and Tuesday
morning

Program

Program

Monday, 7 March 2022

10:00 – 17:00	Registration	
12:30	*LUNCH*	
13:30	Scientific organizers	**Welcome and opening**
14:00 – 14:45	Sakura Pascarelli (online)	**The European XFEL: experimental capabilities and examples of applications**
14:45 – 15:30	Marius Schmidt (online)	**Time-Resolved Crystallography at XFELs**
15:30 – 16:00	**Student Poster talks I**	
16:00 – 16:30	*COFFEE BREAK*	
16:30 – 17:30	**Poster Session I on site**	
17:30 – 18:15	Abbas Ourmazd (online)	**Advanced algorithms to reconstruct the three-dimensional structure of non-stationary objects from random, ultra-low-signal sightings of unknown orientation**
18:15 – 19:00	Alexander Lichtenstein	**Theory of Magnetism and Electronic Correlations**
19:00 – 19:15	Stefan Jorda	**About the Wilhelm and Else Heraeus Foundation**
19:15	*DINNER*	

Program

Tuesday, 8 March 2022

08:00 – 09:00	*BREAKFAST*	
09:00 – 09:45	Anders Nilsson (online)	**X-rays shine light on the mysterious of water**
09:45 – 10:30	Mathias Kläui	**Ultra-fast manipulation of collinar and chiral magnetic order**
10:30 – 11:00	*COFFEE BREAK*	
11:00 – 11:45	Thomas Pfeifer	**Selecting and driving specific electrons: Atoms and molecules in resonance with intense soft-x-ray FEL light**
11:45 – 12:30	Martin Beye	**Science Case for seeded soft X-ray FELs**
12:30	*LUNCH*	
14:00 – 14:45	Harald Reichert	**First science from the first 4th generation high energy synchrotron radiation source ESRF-EBS**
14:45 – 15:30	Claire Donnelly (online)	**Coherent X-ray imaging of three-dimensional magnetic systems**
15:30 – 16:00	**Student Poster talks II**	
16:00 – 16:30	*COFFEE BREAK*	
16:30 – 17:30	**Poster Session II on site**	
17:30 – 18:15	Sarah Köster (online)	**Using focused X-ray beams to study cellular bio-physics**
18:15 – 19:00	Arwen Pearson (online)	**Time-resolved X-ray crystallographic studies of protein function**
19:00	*HERAEUS DINNER at the Physikzentrum* *(cold & warm buffet, with complimentary drinks)*	

Program

Wednesday, 9 March 2022

08:00 – 09:00	*BREAKFAST*	
09:00 – 09:45	Alexandra Pacureanu (online)	**Hard X-ray bioimaging at the nanoscale with fourth generation synchrotron sources**
09:45 – 10:30	Tim Salditt (online)	**High Resolution Phase Contrast X-ray Tomography of Biological Cells and Tissues**
10:30 – 11:00	*COFFEE BREAK*	
11:00 – 11:45	Jan Lüning	**Science case for a 4th soft X-ray storage ring facility**
11:45 – 12:30	Stefan Eisebitt	**Birth, life and death of nanometer-scale spin textures as seen by coherent x-rays**
12:30	*LUNCH*	
14:00 – 14:45	Lucas Schaper	**FLASH2020+: A fully coherent soft xray light source at MHz repetition rate**
14:45 – 15:30	Gianluca Geloni	**Self seeding at hard X-ray FELs**
15:30 – 16:00	**Student Poster talks III**	
16:00 – 16:30	*COFFEE BREAK*	
16:30 – 17:30	**Poster Session III on site**	
17:30 – 18:15	Riccardo Bartolini (online)	**Design Challenges for 4th generation hard X-ray storage ring source PETRA IV**
18:15 – 19:00	Pantaleo Raimondi (online)	**Toward a diffraction limited storage ring based X-Ray**
19:00	*DINNER*	

Program

Thursday, 10 March 2022

08:00 – 09:00	*BREAKFAST*	
09:00 – 09:45	Claudio Masciovecchio (online)	**Potential of Free Electron Laser based Nonlinear Optics Experiments**
09:45 – 10:30	Gerd Schönhense (online)	**New concepts of ToF electron detection**
10:30 – 11:00	*COFFEE BREAK*	
11:00 – 11:45	Curt Preissner (online)	**Fast sample positioning systems with nm precision**
11:45 – 12:30	Josep Nicolas (online)	**Instrumentation issues on modern synchrotron radiation sources**
12:30	*LUNCH*	
14:00 – 14:45	Ana Diaz	**X-ray ptychographic tomography at the Swiss Light Source: method development and**
14:45 – 15:30	Yong Chu (online)	**Current and near-future x-ray microscopy capabilities at NSLS-II**
15:30 – 16:00	**Student Poster talks IV**	
16:00 – 16:30	*COFFEE BREAK*	
16:30 – 17:30	**Poster Session IV online**	
17:30 – 18:15	James Sethian (online)	**Mathematically-based machine learning for understanding scientific experiments**
18:15 – 19:00	**Panel discussion**	
19:00	*DINNER*	

Program

Friday, 11 March 2022

08:00 – 09:00	*BREAKFAST*	
09:00 – 09:45	Heinz Graafsma	**Detector Developments at DESY-Hamburg**
09:45 – 10:30	Bernd Schmitt	**Detector developments for photon science at PSI**
10:30 – 11:00	*COFFEE BREAK*	
11:00 – 11:45	Saša Bajt (online)	**X-ray imaging with multilayer Laue lenses**
11:45 – 12:30	Raymond Barrett (online)	**Optics for Hard X-ray DLSR sources**
12:30 – 12:45	Scientific organizers	**Closing remarks and poster prize**
12:45	*LUNCH*	

End of Seminar / Departure

Posters

Posters I

Robin Yoel Engel	**Non-Linear X-ray Absorption Spectroscopy of thin Nickel films**
Erik Månsson	**Attosecond and few-femtosecond UV-induced dynamics in complex molecules studied via electron–ion covariance spectrometry**
Mahdi Mohammadi Bidhendi	**Single-shot temporal characterization of XUV SASE FEL Pulses**
Daniele Ronchetti	**X-ray diffraction from population inverted atoms**
Markus Scholz	**The complete interface molecular movie: One-stop imaging of electronic and structural dynamics**
Carola Forster	**In situ microstructure investigation of the laser beam welding process using synchrotron radiation**
Jan-Dierk Grunwaldt	**New Opportunities of Diffraction Limited Synchrotron Light Sources for Catalysis Research**

Posters II

Stephan Klumpp	**PETRA IV - The ultimative 3D-X-ray Microscope - General Project Information**
Thomas Lane	**Statistical crystallography reveals molecular motions of the main protease from SARS-CoV-2**
Patrick Lömker	**Establishing operando catalysis photon electron spectroscopy at PETRA IV with the POLARIS setup**
Leila Noohinejad	**4th Generation- synchrotron X-ray: New opportunities for advanced crystallography with high resolution detectors**
Christian Schroer	**Understanding Complex Materials and Processes in Nature and Technology with the Ultimate X-ray Microscope PETRA IV**
Martin Seyrich	**Multi-electron Wave Propagation Simulations of Nano-Imaging Beamlines at Diffraction Limited Storage Rings: Opportunities and Challenges**
Yoo Jung Sohn	**Synchrotron X-ray microdiffraction on thermal barrier coating revealing multi-phase evolution upon thermal loading**
Jakob Soltau	**Hard x-ray microscopy enhanced by coherent image reconstruction**

Posters III

Marc Widenmeyer	**Quantifying the Role of Defect Dynamics and Dynamic Structural Disorder in Energy Conversion Materials**
Lucas Wolff	**Unraveling Time- and Frequency-Resolved Nuclear Resonant Scattering Spectra**
Wolfgang Diete	**High stability beamline instrumentation for diffraction limited sources**
Miriam Gerharz	**Fast resonant adaptive x-ray optics via mechanically-induced refractive-index control**
Adam Kubec	**XRnanotech - Achromatic X-ray Lens and recent developments in nanostructured X-ray optics**
Steve Ngrebada	**Diffraction Limited Synchrotron Light Sources and Next Generation Free Electron Lasers**
Andrei Trebushinin	**Gaussian random field generator SERVAL: a novel algorithm to simulate partially coherent synchrotron radiation**

Posters IV all online

Nils Brouwer	**Calculating electronic structure and X-ray spectra for XFEL experiments**
Yusuf Bulut	**In-situ investigation during gold HiPIMS deposition onto polymers**
Huaiyu Chen	**Coherent Bragg Imaging of Nanoparticles with Unknown Rotation**
Wolfgang Caliebe	**The Advanced XAFS Beamline P64 at PETRA III**
Seyedmohammadali Hosseini Saber	**The 1Mpix AGIPD system for the HIBEF user consortium**
Rajeshkumar Hyam	**Understanding the synthesis and characterization of TiO2 nanostructures by modified anodization technique using Synchrotron radiations**
Joakim Laksman	**Photoelectron spectrometer for hard X-ray photon diagnostics at the European XFEL**
Hanns-Peter Liermann	**Bridging the gap between static and dynamic compression: New dynamic DAC developments at the Extreme Conditions Beamline P02.2 at PETRA III**
Lourdu Xavier Paulraj	**Sub-3 nm Resolution 3D Diffractive Imaging of Anisotropic Gold Nanoparticles with Millions of Patterns using MHz XFEL Pulses**
Carlos Sato Baraldi Dias	**HIKa – Hierarchical Imaging Karlsruhe**
Nils Oliver Wind	**3-in-1 time-resolved ToF momentum microscopy using FEL and hyperspectral HHG radiation**

Posters IV all online

Mangalika Sinha	**Multimodal spectroscopy with upgraded TRIXS end-station at FLASH**
Martina Müller	**Hard X-ray Photoelectron Spectroscopy (HAXPES) of Tunable Oxide Interfaces**

Abstracts of Lectures

(in alphabetical order)

X-ray imaging with multilayer Laue lenses

S. Bajt

[1] *Center for Free-Electron Laser Science, DESY, Hamburg, Germany*

[2] *The Hamburg Centre for Ultrafast Imaging, Hamburg, Germany*

Diffraction limited synchrotron light sources provide X-rays with very small emittance and X-ray Free Electron Lasers (XFELs) offer intense, coherent and short X-ray pulses. These sources opened up new science fields and applications. However, many applications still require further focusing and shaping of these beams, which can be achieved with high precision X-ray optics. We are developing such X-ray optics based on synthetic multilayer structures. These are fabricated to a precision below 1 Å by magnetron sputtering. Using this technique, we create volume structures that diffract x-rays with very high efficiency. High intensities and nanometer spots can be achieved with multilayer Laue lenses (MLLs) as demonstrated with synchrotron and XFEL beams [1,2]. We made substantial progress in the preparation of these 3D nanostructured optical elements. This is due to better understanding of the material properties and at-wavelength optical metrology, which is used to characterize their performance.

One of the imaging methods that we are developing is projection holography. This method takes advantage of the high numerical aperture (NA) of these lenses. Projection holography is a particularly robust high-resolution method, which is suitable for all kind of x-ray sources including XFELs. Using high NA lenses, such as MLLs, we can create a magnified hologram by placing an object in the highly divergent beam just beyond the focus. Holograms of arbitrarily-large fields of view can be recorded using a new approach we developed (ptychographic X-ray speckle tracking) [3,4,5]. The method can stitch together holograms with high accuracy, preserving the inherent high-resolution set by the focal spot size. In doing so, residual wavefront distortions of the lens are corrected for, providing a sensitive non-interferometric wavefront metrology. Concurrently, the phases of the holograms are retrieved in this process, providing a high-resolution phase-sensitive image of the sample.

References

[1] S. Bajt et al., Light Sci. Appl. 7, 17162 (2018).
[2] M. Prasciolu et al., SPIE Proc. Vol. 11886, 118860M-1 (2021).
[3] A. Morgan et al., J. Appl. Cryst. 53, 760 (2020).
[4] A. Morgan et al., J. Appl. Cryst. 53, 927 (2020).
[5] A. Morgan et al., J. Appl. Cryst. 53, 1603 (2020).

Optics for Hard X-ray DLSR Sources

R. Barrett

European Synchrotron Radiation Facility, CS40220, 38043 Grenoble Cedex 09, France

The ESRF is currently in the final stages of a major facility upgrade: the ESRF-EBS (Extremely Brilliant Source). A major element of this program has been the installation of a new electron storage ring based upon a hybrid multi-bend achromat (HMBA) lattice operating at 6 GeV. The EBS source, which opened to users in 2020, provides high-energy X-ray beams with brilliance and coherence increases of one to two orders of magnitude compared with the previous ESRF storage ring. In parallel, a major program of construction of new beamlines and renewal of existing facilities is in progress to exploit fully the opportunities offered by the new source.

In this talk, I will discuss some of the beam characteristics of such sources and the consequences this can have for the choice of optical systems for the upgraded beamline portfolio. Subsequently I will show some of the strategies which are used for beam-conditioning optics (white-beam mirror systems, multilayer and crystal monochromators…) and for focusing optics.

In the course of the talk we will address some of the major technological challenges for optics instrumentation which are still performance limiting with these new generation sources. Several potential solutions which are being actively investigated to overcome these barriers will be presented as well as some newly developed technologies which promise improved capabilities.

References

[1] J. Susini *et al*, *J. Synchrotron Rad.* **21**, 986–95 (2014) https://doi.org/10.1107/S1600577514015951.

Science case for a 4th soft X-ray storage ring facility

<u>Riccardo Bartolini</u>

DESY, MPY, Hamburg, Germany

Science Case for seeded soft X-ray FELs

M. Beye[1]

[1] *Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany*

Most free-electron lasers (FELs) are operated on the principle of self-amplified spontaneous emission (SASE). In this process of generating the FEL radiation, photons that are emitted from normal undulator radiation are exponentially amplified within the long undulator section. The stochastic nature of the spontaneous undulator radiation results in a shot-to-shot varying spectrum of several spikes with similarly stochastic temporal profile. The generated radiation thus has very high spatial (transversal) coherence, but poor longitudinal coherence. Using external seeding though, the longitudinal profile can be completely controlled as well and fully coherent radiation that has all laser-like properties can be generated [1].
Such photon pulses open a vast field of new science opportunities across all photon science disciplines and some highlight examples will be discussed in the talk [2].

References

[1] O. Gorobtsov et al., Phys. Rev. A **95**, 023843 (2017)

[2] M. Beye et al., FLASH2020+: Making FLASH brighter, faster and more flexible : Conceptual Design Report (2020)

Deep Nanoscale X-ray Imaging at NSLS-II

Yong S. Chu*, Hanfei Yan, Xiaojing Huang, Ajith Pattammattel, Randy Smith, Mingyuan Ge, Xianghui Xiao, and Wah-Keat Lee

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The NSLS-II has outstanding nanoscale x-ray imaging capabilities in the hard x-ray regime, enabling a broad range of scientific research. Full-field imaging through transmission x-ray microscopy (TXM) is an ideal imaging method for visualizing 3D morphological, elemental, and chemical distribution with excellent throughput, field of view, and resolution. In particular, 3D chemical imaging, coupling the spectroscopic sensitivity, finds excellent in-situ application for quantifying 3D chemical transformation in complex materials. Though much slower, scanning microscopy using a small focal spot provides outstanding detection sensitivity for various measurement modalities (morphological, elemental, chemical, crystalline, etc.) with superb spatial resolutions. This so-called multimodal imaging finds broad applications for imaging trace element distribution, chemical speciation, and crystalline defects. At the Hard X-ray Nanoprobe (HXN), significant development efforts are directed toward achieving sub-10 nm 3D resolutions. The presentation will describe the latest technical capabilities, science applications, and near-future directions for deep nanoscale imaging.

Developments and applications of ptychography for hard X-ray imaging

A. Diaz[1]

[1] *Paul Scherrer Institute, Villigen PSI, Switzerland*

X-ray ptychography is a powerful imaging technique achieving high spatial resolution not limited by X-ray optics. At the coherent small-angle X-ray scattering (cSAXS) beamline at the Swiss Light Source we have established X-ray ptychography and its combination with tomography as a valuable tool for X-ray optics characterization [1] and for the investigation of a broad range of complex materials like e. g. hydrated cement [2], solar cells [3], biological tissues [4,5] and strained crystals [6]. In this presentation I will speak on behalf of the beamline staff to introduce our latest developments and applications. Because the performance of X-ray ptychography relates directly to the available coherent flux, it is expected that the upgrade of the Swiss Light Source will have a big impact in our results.

References

[1] A. Kubec *et al.*, Nature Communications, recently accepted.
[2] S. Shirani *et al.*, Cement and Concrete Research **137**, 106201 (2020)
[3] P. S. Jørgensen *et al.*, Physical Review Research **2**, 013379 (2020)
[4] V. Panneels *et al.*, J. Cell Science **134**, jcs258561 (2021)
[5] H. T. Tran *et al.*, Frontiers in Neuroscience **14**, 570019 (2020)
[6] M. Verezhak *et al.*, Physical Review B **103**, 144107 (2021)

Coherent X-ray imaging of three-dimensional magnetic systems

Claire Donnelly[1]

[1]*Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str 40, 01187 Dresden*

Three dimensional magnetic systems promise significant opportunities for applications, for example providing higher density devices [1] and new functionalities associated with complex topology and greater degrees of freedom [2,3]. With the recent development of three-dimensional imaging techniques, it is now possible to map internal three-dimensional magnetic structures, and their response to external excitations. In this way we have observed three-dimensional vortex structures, as well as Bloch point singularities [4,5] and, more recently, nanoscale magnetic vortex rings [6,7].

In addition to the static magnetic structure, the dynamic response of the 3D magnetic configuration to excitations is key to our understanding of both fundamental physics, and applications. With our recent development of X-ray magnetic laminography [5], it is now possible to determine the magnetisation dynamics of a three-dimensional magnetic system [5].

These new experimental capabilities for 3D magnetic systems open the door to complex three-dimensional magnetic structures, and their dynamic behaviour. The next generation of synchrotron radiation represents an exciting opportunity for the field, as current measurements remain flux and noise limited. As a result, the promised increase in coherent flux will improve the achievable spatial resolutions, widen the variety of materials that can be probed, and increase the time efficiency of the measurements.

References

[1] Parkin et al., Science **320**, 190 (2008)
[2] Fernández-Pacheco et al., Nat. Comm. **8**, 15756 (2017)
[3] Donnelly and Scagnoli, J. Phys. D: Cond. Matt. **32**, 213001 (2020).
[4] Donnelly et al., Nature **547**, 328 (2017).
[5] Donnelly et al., Nature Nanotechnology **15**, 356 (2020).
[6] Cooper, PRL. **82**, 1554 (1999).
[7] Donnelly et al., Nat. Phys. **17**, 316 (2020)

Nano- to mesoscale magnetization dynamics: new insight enabled by coherent x-ray pulses

S. Eisebitt

Max Born Institute, Max-Born-Str. 2a, 12489 Berlin, Germany

I will present two examples from our research on magnetic structures to demonstrate the unique potential of coherent x-ray pulses to advance our understanding of dynamics process in materials from the nano- to micrometer scale spanning timescales from femtoseconds to seconds.

Magnetic Skyrmions are spin textures which behave as quasi-particles and are characterized by a specific topology. Some types of skyrmions do exist in suitable thin magnetic multilayer film systems at room temperature, and they can be generated and efficiently moved laterally by spin-polarized current pulses. I will discuss how magnetic skyrmions can be generated via *laser* pulses instead of current pulses at unprecedented speed, with the topology change completed after 300 ps. Insight into the ultrafast laser-induced formation mechanism comes from time-resolved scattering experiments at x-ray lasers and x-ray holography, in combination with atomistic spin simulations. Local topology fluctuations in a transient high temperature phase are identified as the key element for this topological phase transition - a mechanism that may be applicable to phase transitions with a net change of topology in completely different material systems as well. [1]

Rather than following the *ensemble average* dynamics of a system via incoherent scattering, time resolved imaging is a means to observe the *individual* dynamics of a system in real space. Here, triggerable deterministic dynamics is commonly accessible via pump-probe schemes, allowing to collect sufficient statistics for high spatial resolution via repetition of the experiment. The observation of *stochastic* dynamics, however, including equilibrium dynamics, is more problematic: temporal resolution and spatial resolution are in direct competition via the signal to noise achievable during a given exposure time. The situation is identical to the making of a “classic” movie consisting of an untriggered image sequence: it is difficult to shoot a high speed movie at low light. Here, we present *coherent correlation imaging* (CCI) as a novel approach mitigating this dilemma by going away from the blind, continuous averaging of subsequent coherent scattering patterns to a correlation-based classification of coherent scattering patterns. CCI allows to obtain images with high spatial resolution without compromising temporal resolution. As a proof-of-principle, the observation of thermal hopping of magnetic domain walls is presented. [2]

References

[1] F. Büttner, et al., *Observation of Fluctuation-Mediated Picosecond Nucleation of a Topological Phase*, Nature Materials **20**, 30 (2021).

[2] C. Klose, F. Büttner, W. Hu, C. Mazzoli, K. Litzius, R. Battistelli, I. Lemesh, J. M. Bartell, M. Huang, C.M. Günther, M. Schneider, A. Barbour, S.B. Wilkins, G.S.D. Beach, S. Eisebitt and B. Pfau, *Coherent Correlation Imaging: Resolving fluctuating states of matter* (submitted).

The Hard X-ray Self-Seeding System at the European XFEL

G. Geloni[1]

[1] *European XFEL, Schenefeld, Germany*

In this contribution I will describe the Hard X-Ray Self-Seeding (HXRSS) system at the SASE2 FEL line of the European XFEL, on behalf of the HXRSS team. After an introduction about self-seeding and the theory of single-crystal monochromators for self-seeding purposes, I will discuss the peculiarities of our setup, that deals with long undulator and high-repetition rate. I will then move on to discuss the commissioning and current performance of the system, as well as possible future developments.

“Detector Developments at DESY-Hamburg”

Heinz Graafsma, DESY, Hamburg, Germany

This being the first of two presentations on detectors, the second one is given by Bernd Schmitt from PSI, I will start with explaining some very basic concepts about detectors. This will include the principle differences between photon-counting and integrating detectors, the adaptive-gain principle, hybrid pixel array detectors (HPADs), monolithic active pixel sensors (MAPS or CMOS imagers) and back-side illumination (BSI) versus front-side illumination (FSI).

In the second, and main part, of the lecture I will first present the Adaptive Gain Integrating Pixel Detector (AGIPD) developed explicitly for the European XFEL, including its performance parameters and shortcomings. This will be followed by the PERCIVAL system, a monolithic sensor system developed for soft X-rays at Free-Electron Lasers. The last part is devoted to current developments of new systems. First the TimePix4 based system, a versatile chip for event driven readout with time-of-arrival (ToA) and time-over-threshold (ToT) as well as classical frame based photon-counting capabilities. And finally I will present the CoRDIA project, an integrating, adaptive-gain and high frame-rate imager, which is in the early phase of development.

Ultra-fast chiral spin dynamics

M. Kläui[1,2]

[1]*Institute of Physics, Johannes Gutenberg University Mainz, 55099 Mainz, Germany*
[2]*Center for Quantum Spintronics, NTNU Trondheim, 7530 Trondheim, Norway*

Novel spintronic devices can play a role in the quest for GreenIT if they are stable and can transport and manipulate spin with low power. Devices have been proposed, where switching by energy-efficient approaches, such as spin-polarized currents is used to manipulate topological spin structures [1,2].

Firstly, to obtain ultimate stability of states, topological spin structures that emerge due to the Dzyaloshinskii-Moriya interaction (DMI) at structurally asymmetric interfaces, such as chiral domain walls and skyrmions with enhanced topological protection can be used [3-5]. Here we will introduce these spin structures ad we have investigated in detail their dynamics and find that it is governed by the topology of the spin structure [3,5,6]. By designing the materials, we can even obtain a skyrmion lattice phase as the ground state [4]. Beyond 2D structures, we recently developed also systems with chiral interlayer exchange interactions that lend themselves to the formation of chiral 3D structures [7].

We study in particular the chiral order dynamics using time-resolved x-ray scattering at the free electron laser FERMI [8]. By small angle x-ray scattering in a pump-probe experiment after excitation with a infrared laser, we ascertain the timescale of the formation of collinear magnetization in domains and the formation of chiral domain walls. Surprisingly we find that the latter occurs on a faster timescale, showing the robust formation of chiral spin structures [8]. Additionally, we observe oscillations in the recovery of the order, which are novel features that will be discussed in this presentation [9].

References

[1] K. Everschor-Sitte et al., J. Appl. Phys., vol. 124, no. 24, 240901, 2018.
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[9] C. Leveille et al., In preparation

Scanning SAXS imaging of biological cells

S. Köster

University of Göttingen, Institute for X-Ray Physics, Göttingen, Germany

X-rays provide high resolution due to their small wavelength and high penetration power, allowing for imaging of comparatively large, three-dimensional objects. For these reasons, X-rays have been established as complementary probes for bio-imaging, in addition to well-established methods such as visible light fluorescence microscopy and electron microscopy (EM). Scanning small angle X-ray scattering (SAXS), in particular, is well suited for systems with some degree of order, such as bundles of parallel filaments, or high-density aggregates. The method exploits two unique features of X-ray imaging: not only are highly focused beams used to spatially resolve different constituents of biological cells, but each individual scattering pattern contains a wealth of information about the internal structure on molecular length scales. I will present scanning SAXS experiments that were performed at dedicated synchrotron beamlines, which provide a small beam between 100 nm and 2 µm in diameter, high flux, high-end pixel detectors and a sample environment suitable for cell samples. I will summarize the most important results we recently obtained on different biological systems, such as components of the cytoskeleton and the DNA in the nucleus.

Time-dependent XFEL spectroscopy of correlated system

Alexander Lichtenstein

University of Hamburg, Germany

The prospect theoretical description of ultra-fast spectroscopy of correlated electron systems under influence of ultra-strong XFEL pulses will be discussed. Time-dependent dynamical mean field theory in combination with the first-principle DFT-scheme is an optimal starting point to go beyond static density functional approximation and include effects of spin and charge fluctuations in strongly correlated materials [1].

We present a generalization of the recently developed dual fermion approach for correlated lattices to non-equilibrium problems [2]. In its local limit, the approach has been used to devise an efficient impurity solver, the super-perturbation solver for the Anderson impurity model. Here we show that the general dual perturbation theory can be formulated on the Keldysh contour. Starting from a reference Hamiltonian system, in which the time-dependent solution is found by exact diagonalization, we make a dual perturbation expansion in order to account for the time-dependent relaxation effects in different photoemission spectroscopy. Simple results for generic correlated models will be presented.

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Science case for a 4th soft X-ray storage ring facility

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New research opportunities with FELs

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In this paper, we will discuss how recent advances in the performance of the FELs allowed non-linear experiments at sub-optical wavelengths. In particular Second Harmonic Generation (SHG) [1] and Transient Grating (TG) [2] experiments have finally demonstrated the high potential of VUV/soft X-ray wave mixing techniques. SHG is one of the second order non-linear responses of systems that are non-zero only for non-centrosymmetric materials as surfaces or interfaces. The advantages of using electronic resonance would imply a significantly higher surface specificity than existing soft X-ray methods [1]. TG experiments at sub-optical wavelength are relevant for the study of nanoscale dynamics in disordered systems as well as in semiconductors. Exciting phonon modes with nanometer wavelength would allow shedding light on a plethora of scientific open problems ranging from the thermal anomalies in glasses to understanding nanoscale thermal transport. Indeed the study of thermal transport approaching the nanometer is extremely important and motivated by relevant technological needs such as thermal management of electronic devices [3]. Wave mixing in the soft X-ray can be used as well to investigate drug/target intermolecular vibrational dynamics, possessing the potential to understand the marked differences in biological activities of enantiomers, or easily follow the dynamic of metal complexes [4].

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Instrumentation issues on modern synchrotron radiation sources

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Mystery of Water; Oppertunities for X-ray Science

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Water is of extreme importance for our society and the key component of life as we know it but it is also of extraordinary interest due to its remarkable physical properties that differ from almost all other liquids. Deviation of water's properties from a simple liquid exists already in the ambient temperature regime and then becomes strongly enhanced upon supercooling. In particular the finding that the thermodynamic response and correlation functions appear to diverge towards a singular temperature estimated by power-law fits of about 228 K has led to several hypotheses about the origin of waters anomalous properties. One hypothesis to explain the apparent divergence is that there exists a liquid-liquid transition with a liquid-liquid critical point at rather high positive pressures. In this scenario the Widom line, defined as the locus of correlation length maxima in the P-T plane, emanates from the critical point as a continuation of the liquid-liquid transition line into the one-phase region and the divergence in the response functions is towards this line. The challenge is that the temperature T_s lies below the homogeneous ice nucleation temperature 232 K, a region of the phase diagram that has been denoted as "no man's land", since ice crystallization occurs on much faster time scale compared to the experimentally accessible time scale in a typical laboratory setting.

Here I will present how x-ray lasers and synchrotron radiation can be used to probe the liquid in the deep supercooled water regime inside no-man's land. In particular I will discuss if a liquid-liquid transition, Widom line and a critical point exists in deep supercooled water causing fluctuations all the way up to ambient temperature. The existence of the fluctuations in water can have an impact on our understanding of biology and energy technologies.

What can we learn from Machine Learning?

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The advent of high repetition-rate XFELs is generating a torrent of data. Will machine learning conquer the deluge?

Machine learning, a branch of Artificial Intelligence, perform tasks typically reserved for humans. Most machine-learning tasks involve some kind of “recognition”. Examples include recognizing individuals (facial recognition), obstacles (self-driving vehicles), or patterns (stock-market fluctuations).

Recognition tasks are, in essence, labeling exercises. Recognizing a face, for example, involves attaching a name to it. Most machine-learning approaches, such as “Deep Learning”, provide little or no insight into the principles by which the labels are generated. The ability to perform a task does not require understanding the underlying processes. You do not have to understand the workings of the brain to recognize your spouse.

Scientific knowledge, in contrast, entails understanding the underlying processes. A deep understanding of facial recognition, for example, must elucidate the structures and processes by which the brain recognizes faces.

Traditionally, scientific understanding proceeds by assimilating a few experimental clues into a (mathematically sound) theory. This theory is then buttressed by a succession of carefully designed observations. Such discovery processes are designed to make the best use of limited data. The data deluge is undermining this approach.

I will describe how machine learning can help extract scientific understanding from the data deluge.

This work was supported by the US Department of Energy, Office of Science, Basic Energy Sciences under award DE-SC0002164 (underlying dynamical techniques), and by the US National Science Foundation under award STC 1231306 (underlying data analytical techniques).

Hard X-ray bioimaging at the nanoscale with fourth generation synchrotron sources

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Coherent hard X-ray microscopy has seen remarkable progress in recent years and it is increasingly recognized as a disruptive technology for life science research[1-4]. With the advent of the fourth generation synchrotron sources, new opportunities arise to probe the 3D structure and elemental composition of biological specimens with unprecedented precision and speed. The penetration power of high energy X-rays enables imaging of thick biological samples, which can be preserved close to their natural state through vitrification and imaging in cryogenic conditions. The improved coherence of fourth generation sources fosters overcoming frontiers of resolving power.

This talk will cover current capabilities of X-ray phase contrast nano-tomography and X-ray fluorescence at the nano-imaging beamline ID16A of ESRF for exploration of cells, tissues and organisms from our ecosystems. We will discuss as well the challenges to address for improving spatial resolution and scalability, along with opportunities to integrate X-ray microscopy with modalities measuring functional and molecular properties of living organisms in order to advance our understanding of life.

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The European XFEL: new science opportunities and first results

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In the past decade we have seen very important developments in the field of accelerator based X-ray user facilities, with the advent of diffraction limited storage rings and MHz rate free electron lasers. The first hard X-ray free-electron laser, LCLS (US) became operational in 2009 and over the last decade four additional hard X-ray FELs have begun user operation - SACLA (Japan), PAL-FEL (Korea), EuXFEL (Germany) and SwissFEL (Switzerland). Among these, the EuXFEL is the first hard XFEL powered by a superconducting linear accelerator, which enables MHz rate pulse generation. A specificity of XFELs is their very short pulse duration (10-100fs) opening new scientific opportunities to probe matter at the atomic scale, with chemical selectivity and bulk sensitivity, and on the relevant timescales. Ultrashort, high intensity X-ray pulses from FELs are also providing a totally new approach to structural determination with X-rays, where useful structural information from very small, "radiation sensitive" or "dynamic" crystals is acquired before radiation damage sets in. After many years of construction, the EuXFEL has started user operation in 2017 and today operates six instruments. In this lecture I will first briefly introduce the present performance of the facility in terms of electron and photon beam characteristics. I will then report first results from early user experiments, and comment on some important challenges ahead.

Time-resolved X-ray crystallographic studies of protein function

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Structural studies are key to understanding how biological macromolecules are able to carry out diverse functions including binding, signaling and catalysis. A true picture of structure-function relationships requires that we also understand the dynamics of the macromolecule, and these functionally relevant dynamics span many orders in time, from femtosecond chemical steps to slower, larger scale conformational rearrangements that occur over milliseconds and seconds. Time-resolved crystallography provides a window into both structure and dynamics. Recent advances in X-ray sources, automation and sample delivery mean such experiments are no longer only accessible to a small number of highly expert groups. I will review some of these current advances and highlight the remaining bottlenecks in making these experiments fully part of the “standard” tool kit of the structural biologist.

Listening to, and directing, electrons on the atomic level

to understand quantum dynamics from attoseconds to femtoseconds, and beyond

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Electrons are not only the lightest (known) fundamental particles, they are also responsible for virtually anything visible to us from tiny objects under the microscope to the screen or paper we just look at, all the way and far out into the universe. Interactions with electrons also hold atoms together in molecules and electron motion is at the heart of any chemical reaction on earth, our bodies and in outer space. Observing, understanding, and directing electron motion is thus a core topic in physics and chemistry alike, with substantial impact and relevance to other fields of science and technology.

An ongoing world-wide revolution of high-frequency (XUV and x-ray) light-source development keeps unlocking our access to such electron motion, down to their natural time scale of attoseconds. Moreover, the extreme intensity of current free-electron lasers (FELs) allows not only the observation of fundamental quantum dynamics in atoms and molecules, but to actively steer their motion on the fundamental electronic level.

Here in this talk, I will highlight a few examples of our continuing science mission towards building a bottom-up understanding of electron dynamics, starting at the level of isolated atoms featuring only a nucleus and two (correlated) electrons (e.g. in Helium), and working our way up into multi-electron atoms (e.g. neon) and polyatomic molecules (e.g. CH_2I_2 and C_{60}).

Our experimental aim is to obtain the most complete picture of such fundamental light-matter interaction processes from low to high intensity by extracting multidimensional observables. These include the detection of photons, ions and electrons in coincidence.

The results from these experiments not only shed new light on fundamental processes, which are generally applicable also in large-scale systems (also relevant to single-shot imaging of molecules in intense x-ray fields), but also provide new technological tools for pulse characterization, to further optimize the light sources themselves. One exemplary long-term dream of this development is the 3D-printer on the atomic level: The writing of custom molecular structures and supramolecular assemblies by the full spatio-temporal control over the electromagnetic spectrum, coherently provided at high intensity from the infrared into the x-ray domain.

The PtychoProbe and the Advanced Photon Source Upgrade

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The PtychoProbe will be one of the nine feature beamlines constructed as part of the APS Upgrade (APS-U). It will have 5-nm focusing capability, be fast scanning, and combine high resolution structure information with high resolution chemical information. The requirements push the synchrotron instrumentation engineering limits for metrology, control, and data acquisition. In addition to the Ptychoprobe, eight other feature beamlines are being constructed, with the goal of each having a world-class instrument. Upgrades will also be made to 14 existing beamlines so they can take full advantage of the APS-U. Supporting all of this are both new and upgraded conventional facilities. This talk will provide an update on the PtychoProbe and a broad look at developments across the APS-U

This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility at Argonne National Laboratory and is based on research supported by the U.S. DOE Office of Science-Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

Toward a diffraction limited storage ring based X-Ray source

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Multi-bend achromat (MBA) lattices have initiated a fourth generation for storage ring light sources with orders of magnitude increase in brightness and transverse coherence. A few MBA rings have been built, and many others are in design or construction worldwide, including upgrades of APS and ALS in the US. The HMBA (hybrid MBA), developed for the successful ESRF-EBS MBA upgrade has proven to be very effective in addressing the nonlinear dynamics challenges associated with pushing the emittance toward the diffraction limit. The evolution of the HMBA ring designs will be described in this seminar. The new designs are consistent with the breaking of the lattice periodicity found in traditional circular light sources, inserting dedicated sections for efficient injection and additional emittance damping. Techniques developed for high energy physics rings to mitigate nonlinear dynamics challenges associated with breaking periodicity at collision points were applied in the HMBA designs for the injection and damping sections. These techniques were also used to optimize the individual HMBA cell nonlinear dynamics. The resulting HMBA can deliver the long-sought diffraction limited source while maintaining the temporal and transverse stability of third generation light sources due to the long lifetime and traditional off-axis injection enabled by nonlinear dynamics optimization, thus improving upon the performance of rings now under construction.

Science at the first 4th generation high energy synchrotron radiation source

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The ESRF started operation in 1992 as the world's first 3rd generation synchrotron radiation source. Since 2009 the ESRF engaged in an ambitious upgrade programme delivering an accelerator complex and beamlines with orders of magnitude higher performance. The replacement of the old ESRF storage ring employing a standard double-bend achromat (DBA) lattice by the EBS storage ring based on the newly developed HMBA lattice with seven bending magnets per cell started in 2015. In order to match the large increase in source performance, the scientific instrumentation has been upgraded in parallel, including a further evolution of the business model.

During a long shutdown the EBS storage ring was installed in 2019 and went into its commissioning phase in December 2019. Since then the EBS storage ring was successfully commissioned as the first 4th generation high energy synchrotron light source during the first six month in 2020. The ESRF resumed its user operation at full current and nominal emittance as scheduled in August 2020. The user programme was then ramped up quickly with all beamlines taking beam today. The expected improvement of the key beam parameters (brilliance, coherence, flux) were confirmed across the entire beamline portfolio. To date more than 2000 user experiments have already been performed with first exciting scientific results already submitted for publication. The process for the technical upgrade of the facility and the scientific exploitation will be briefly explained together with early scientific results demonstrating the potential of this new generation of X-ray sources for the next big step in X-ray science.

Challenges of multi-scale X-ray imaging: optics, wavefield, reconstruction, and 3d image analysis

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The potential of hard x-rays in view of penetration, spatial resolution, contrast, and compatibility with environmental conditions was for long time locked up by the lack of suitable in x-ray optics. With the advent of highly brilliant radiation, and the development of lens-less diffractive imaging and coherent focusing, the situation has changed. We now have nano-focused coherent x-ray synchrotron beams at hand to probe soft and biological matter as well as materials and nanostructures, both in scanning and in full field imaging and tomography. Source coherence, optics and detection schemes allow to radically rethink our experimental designs. This opens up a plethora of contrast mechanisms, increased resolution, and scalable volume throughput, and automated feature extraction.

In this talk, I will concentrate on the central challenge of inverting the coherent diffraction pattern by suitable reconstruction algorithms in the optical far and near-field [1,2]. I will address optimized experimental design, including illumination with filtered wavefronts based on waveguide optics [1,2], and show how imaging and diffraction can be combined to achieve super-resolution [3]. Finally, different examples of biophysical and biomedical applications will be presented, including 3d virtual histology of human brain tissue [4], as well as Covid-19 [5].

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Science case for a 4th soft X-ray storage ring facility

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Time-Resolved Crystallography at XFELs

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The focus of structural biology is shifting from the determination of static structures to the investigation of dynamical aspects of macromolecular function. With time-resolved serial femtosecond crystallography (TR-SFX) at free electron X-ray lasers (XFELs) bio-macromolecular reactions can be followed on all time scales from femtoseconds to seconds [1-4]. Radiation damage is minimized, and an unperturbed view on the progress on the reaction is provided in real time. Since microcrystals are investigated, pump-probe techniques can be exquisitely well applied since the penetration depth of optical laser light that initiates a reaction in the crystals and the crystal size match [2, 3, 5-7]. Moreover, reactions initiated by mixing substrate with enzyme microcrystals become possible [4, 8-10]. This opens the door to the investigation of all sorts of reactions in bio-medically highly relevant enzymes. This talk covers concepts and newest results obtained at various XFELs worldwide [7, 10, 11].

Research was supported by BioXFEL, a NSF supported Science and Technology Center, grant NSF-1231306.

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Detector Development for Photon Science at PSI

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While hybrid pixel detectors from PSI are used at synchrotrons and XFELs worldwide our detector developments are currently concentrating on the detector needs arising from the new photon science facilities at PSI: Athos, the low energy branch at SWISSFEL, and SLS2, the upgrade of the SLS to a fourth generation diffraction-limited light source.

The two end stations at Athos work in an energy range between 250eV and 2 keV. One focus was therefore the development of sensors for low photon energies. In collaboration with FBK and CIS we optimized in a first step the entrance window for low photon energies achieving an efficiency larger 80% for photon energies above 250eV. In a second step together with FBK we now work on combining the thin entrance window with LGAD technology, i.e. the implementation of a gain layer in the sensor, to overcome the noise in the readout chain and to achieve single photon resolution down to a few hundred eV. These sensors can be combined with charge integrating readout chips for applications at XFELs but also with single photon counting readout chips for low photon energy applications at synchrotrons.

The main challenge at SLS2 is the increased brightness requiring detectors with a count rate capability one order of magnitude higher than what is available today. Therefore, we have recently started the development of Matterhorn our new single photon counting detector optimized for high count rates using a pile up counting architecture which we have already implemented and tested in Mythen3 achieving a count rate capability above 20 MHz per pixel at an efficiency above 80%.

In this presentation I will give an overview of our current and future developments.

New Concepts of ToF Electron Detection

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Momentum microscopy (MM) is a novel way of performing angular-resolved photoelectron spectroscopy (ARPES). Combined with time-of-flight (ToF) energy recording, its high degree of parallelization is advantageous for time-resolved and spin-resolved ARPES, especially in the X-ray range. The performance of ToF-MM is illustrated by selected examples obtained in different spectral ranges. Synchrotron radiation at beamline P04 (PETRA-III) was used for tomographic mapping with soft X-rays [1], revealing the full bulk band structure including Fermi surface, Fermi-velocity distribution $v_F(k,E_F)$, full circular dichroism (CDAD) texture and spin signature of W(110). The large probing depth in the hard-X-ray range enables k-resolved measurements with capped samples. For capped films of the collinear antiferromagnet Mn_2Au, the Néel vector has been aligned *ex-situ* with 60 Tesla pulses, prior to the MM experiment at PETRA-III (beamline P22) [2].

A special ToF-MM (HEXTOF [3]), operated at beamline PG2 at FLASH, merges 3 photoemission spectroscopy techniques into a single setup, namely **time-resolved** *momentum microscopy* (trMM), *core-level spectroscopy* (trXPS), and *X-ray photoelectron diffraction* (trXPD). Its performance is illustrated by the *core-cum-conduction* observation of a Mott transition [4], a sub-ps metamagnetic phase transition in FeRh driven by a NIR laser pulse [5] and first photoelectron diffraction measurements in the ultrafast regime. The recorded 4D data arrays $I(E_B,k_x,k_y,t)$ directly correspond to the natural momentum-energy-time basis, in which theory and calculations are formulated. As an *outlook*, developments towards suppression of the space-charge effect will be addressed [6]. We make use of basic principles of charged-particle optics, such as the large flexibility in beam shaping and the fact that particle lenses can be both accelerating and retarding. First results from FLASH have validated the basic concept. Operating the front lens in a *retarding mode* strips off undesired slow electrons from the photoelectron beam within some 10-100 microns from the sample surface. In turn, one can work at much higher pump fluences in comparison with the conventional MM mode with strong *accelerating* extractor field.

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Mathematically-based machine learning for understanding scientific experiments

J.A. Sethian,

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In this talk, we will discuss some challenges arising from aspects of designing and implementing mathematical algorithms for data analysis from experimental facilities. After a brief introduction of the research undertaken at the Center for Advanced Mathematics for Energy Research Applications (CAMERA), we will focus on methods for tackling challenges in designing machine learning-based algorithms for (1) autonomous self-driving experimentation; (2) inverse problems in single particle imaging, coherent surface scattering imaging, and X-ray photon correlation spectroscopy; and (3) mixed-scale dense convolutional neural networks for applications in the case of limited, scarce training data, with applications to cryoEM.

Abstracts of Posters

(in alphabetical order)

Calculating electronic structure and X-ray spectra for XFEL experiments

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The combination of ultrashort time-scale, short wave length and high intensity provided by XFEL radiations has made possible a large range of new experiments, which can provide novel insights into the electronic structure of matter. XFEL driven X-ray spectroscopy is a promising tool to investigate a broad range of subjects, but in this poster I will highlight two examples: warm dense matter and highly correlated electron systems.

Both, in the design and in the interpretation of XFEL experiments, electronic structure theory can be essential to success. However, new experiments also represent a challenge to theory, where model systems can lack material specificity and commonly used approximations may reach the limit of there applicability. Therefore, model accuracy has to be improved, in accordance with the increasing availability of computational resources.

In this poster, I will show preliminary results for two different kind of electronic structure calculations, that were performed in support of recent and upcoming experiments at European XFEL. First, I will present XANES spectra of transition metals under warm dense matter conditions, which were simulated using ab initio molecular dynamics.

Second, as an example for highly correlated electron systems, I will present electronic structure calculations for LaCoO3 obtained with dynamical mean field theory.

Finally, I will discuss ideas for new models adapted to the needs of XFEL theory support.

In-situ investigation during gold HiPIMS deposition onto polymers

<u>Yusuf Bulut</u>[1,2], Kristian Reck[3], Matthias Schwartzkopf[1], Jonas Drewes[3], Suzhe Liang[2], Tianfu Guan[2], Thomas Strunskus[3], Franz Fraupel[3], Peter Müller-Buschbaum[2,4], Stephan V. Roth[1,5]

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Gold deposition *via* high power impulse magnetron sputtering (HiPIMS) allows to coat thin metal layers on heat sensitive materials such as polymers allowing for increased adhesion and density. HiPIMS allows deposition at a lower total deposited thermal energy in comparision to convetional magnetron sputtering, but this energy is delivered in a very short pulse exhibiting very high power and ionization. The consequences for the nucleation and growth processes during HiPIMS deposition are not sufficiently known. Therefore, we investigate the morphology evolution of thin gold layers on three polymer templates, namely polystyrene (PS), polyvinylalcohol (PVA) and poly-4-vinylpyridin (P4VP). These polymers show different functional moieties and thus are expected to influence the growth of the gold layer. We present first results of our *in-situ* investigations combining grazing-incidence small angle X ray scattering (GISAXS) and grazing incidence wide angle X-ray scattering (GIWAXS).

The Advanced XAFS Beamline P64 at PETRA III

W.A. Caliebe[1]

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X-ray Absorption Spectroscopy (XAS) is a standard analytical method in the application of synchrotron radiation in science. XAS itself provides various important element-specific information on the sample like the oxidation state of the investigated element, its local symmetry and distance to its neighbours. The low absorption by x-rays allow measurements in various sample environments under different conditions, like reactors for homogeneous or heterogeneous catalysis.

Several advanced methods in XAS have been implemented at beamline P64 at PETRA III: A fast-oscillating monochromator reduces the time for one absorption scan from a few minutes to the sub-s time regime, which allows to follow chemical reactions in more detail. The combination of a high-resolution emission spectrometer with XAS reduces the life-time broadening of the absorption edge, which provides more precise information on the local symmetry, and high resolution emission spectroscopy of valence electrons gives additional information on the ligands, which are difficult to get with other methods. Fast, high-resolution cameras can be used for spatially resolved spectra on the micron-length scale with sub-minute time resolution. Photo-excitation of the sample with a high-power laser-pulse is used to study the excited state in light-harvesting metal-organic complexes. All these methods profit from the high flux which is available at PETRA III.

Some heroic experiments like XAS ptychography by the groups of Ch. Schroer [1], and Y. Takahashi [2] made use of the still rather moderate coherent flux in combination with XAS in the hard x-ray regime at 3rd generation SR facilities. This will change at 4th generation SR facilities like PETRA IV, which will reduce the measurement time from days to hours. We envision to continue to develop this method at one of the new XAS-beamlines at PETRA IV in order to provide spatially resolved information on the nm length-scale on the valence and local environment.

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Coherent Bragg Imaging of Nanoparticles with Unknown Rotation

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Bragg coherent diffraction X-ray imaging (BCDI) is a promising technique to measure the three-dimensional strain of crystalline nanoparticles. However, it is difficult to perform BCDI experiments on very small sized nanoparticles due to the unpredictable rotations raised by the local heating or radiation pressure. We are now developing a new method that can assemble a model from such unpredicted Bragg diffraction patterns and use it to reconstruct the shape and the strain of the particle.
Our project is inspired by the paper published in 2020[1]. In this paper. Alex extended the Expansion -Maximization-Compression (EMC) algorithm, which is generally used in tomographic experiment, to the Bragg geometry. He showed that the uncontrolled inadvertent rocking curve at X-ray power of 10eV. We are extending the application of this method to cases where the rotation in each direction is completely unknown.

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High stability beamline instrumentation for diffraction limited sources

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The continuous advances towards diffraction-limited synchrotron light sources and free electron laser facilities (FEL) require beamline components with ever-increasing optical and mechanical performance. Key aspects are the positional stability of the x-ray beam at the experiment and the quality of the installed optical elements.

AXILON designs and builds state-of-the-art beamline instrumentation and in this poster we present recent achievements of critical beamline components:

- Cryo-cooled Double Crystal Monochromators which allow beam stabilities below 50nrad
- Mirror systems with mechanical bending mechanisms maintaining mirror slope errors below 100nrad rms
- X-ray Microscopes with advanced metrology and scan control systems with nanometer accuracies even for fast and complex scan trajectories

Non-Linear X-ray Absorption Spectroscopy of thin Nickel films

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Free-electron lasers in combination with nano-focusing capabilities allow for ready access to X-ray fluences in the range of J/cm² within femtosecond pulses, where more than one photon impinges on the same atom during the core-hole lifetime. This allows for novel experiments exploiting various non-linear phenomena from strong light-matter coupling to wave-mixing techniques. A prerequisite to interpreting non-linear effects however is an understanding of how the well-studied process of absorption of x-rays near a material resonance changes with the transition to extreme fluences. Here we present a study of x-ray absorption near-edge spectra of thin nickel films, measured in transmission with the unattenuated monochromatic fluence of the European XFEL at the spectroscopy & coherent scattering instrument with µm-sized foci from zone-plate focusing optics, resulting in fluences up to 50 J/cm² on the sample. The optics also integrate a beam-splitting normalization scheme that is crucial for studying absorption at fluctuating sources and has since developed to a standard configuration at the instrument. The resulting spectra show interesting changes which we model based on the complex interplay between absorption, saturation and electron system heating during the FEL pulse.

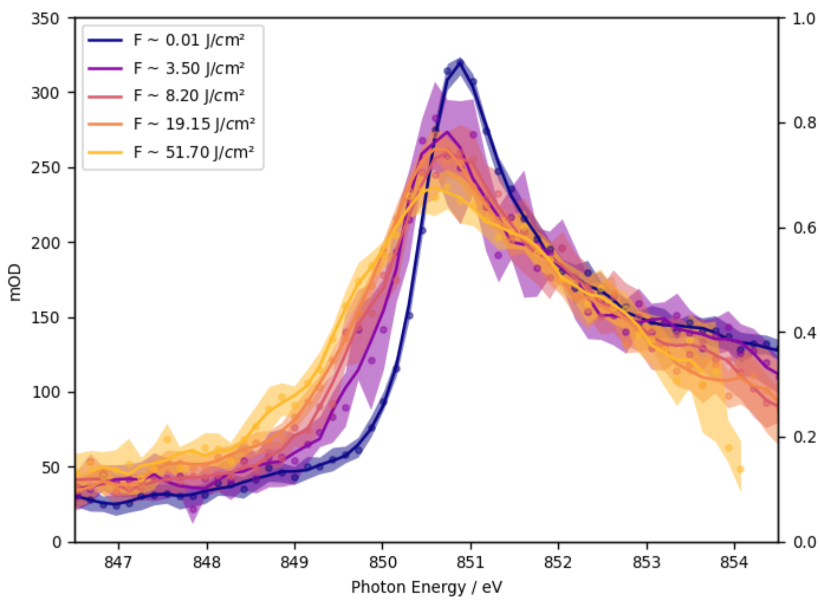

Figure 1: Nickel L3-edge absorption spectra for various fluences.

In situ microstructure investigation of the laser beam welding process using synchrotron radiation

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Laser beam welding has become a key technology in the field of automated industrial manufacturing due to its non-contact machining, short cycle-times and small heat affected zone. However, a major drawback of this processing technology is the possible formation of solidification cracks, reducing the quality and robustness of the weld seam [1]. A new approach to prevent solidification cracks is to influence the shape and flow of the melt pool by changing the process parameters as thermo-fluid dynamic solidification conditions directly influence the properties of the weld seam. However, due to a lack of process knowledge, a time-consuming preliminary process to find the appropriate parameters accompanies this approach [2].

We anticipate that by investigating the laser beam welding process with a synchrotron radiation-based in situ imaging technique with a spatial resolution of 32 - 200 µm, we will be able to resolve the growth and possible breaking off or detaching of individual dendrites of a size of 25 – 250 µm [3]. The gained insights will reveal to what extent and how dendrites influence solidification and crack formation, as in other flow-accompanied solidification situations detached dendrites are serving as condensation nuclei [4]. This will serve as starting point for a more sophisticated process model.

All experiments are going to be executed at DESY as part of a BAG proposal. The planned study is part of a DFG Research Unit, which aims to compile a quantitative process model comprising the relevant phenomena of the laser beam welding process from the macro down to the microscale. The model is going to be implemented in a massively parallel simulation framework to achieve a high performance computing (HPC) simulation of the laser beam welding process. With the help of HPC simulation, it will be possible to identify mechanisms of solidification crack initiation and correlate them with process parameters, to enable the prediction of solidification crack probabilities for various process configurations.

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Fast resonant adaptive x-ray optics via mechanically-induced refractive-index control

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Mössbauer nuclei provide a promising platform for a multitude of applications ranging from fundamental science to applications, due to their extreme properties with transition energies on the order of 10keV and transition widths on the order of a few neV. While having less than one resonant photon per pulse on average at third generation synchrotrons due to the narrow nuclear line width, (next generation) free electron lasers can provide several orders of magnitude higher numbers of resonant photons per pulse. Hence, qualitative new observables and experimental approaches become possible, potentially based on non-linear x-ray-nuclei-interactions or quantum effects. However, better control of the x-rays on nuclear time- and energy scales is necessary to fully exploit the possibilities of the new light sources. In this project we introduce a concept for fast resonant adaptive x- ray optics. Using piezo-control methods, we can displace a solid-state target much faster than the lifetime of its resonances. This creates a mechanically-induced phase shift (see Fig. 1), that can be associated with an additional contribution on resonance to the real part of the refractive index while the imaginary part remains unchanged. Hence, we can achieve polarization control by mechanically-induced birefringence without changes in absorption. We theoretically and experimentally demonstrate the approach with three applications: conversion of linear to circular polarization with optional intensity gain, temporal intensity gating by rotation of linear polarization within a polarimeter and a x-ray polarization interferometer as a sensitive tool for an analysis of mechanical stability on sub-Ångstrom level.

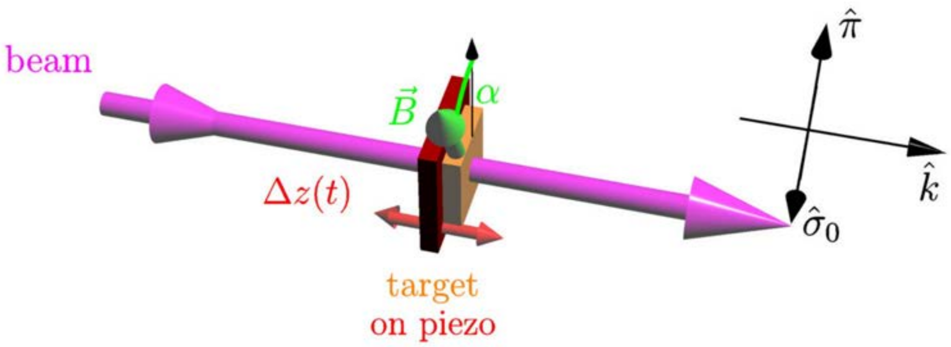

Fig. 1: Schematic setup. The resonant target is mechanically moved via a piezo-electric transducer and thereby imprints a time-dependent phase onto the scattered x-rays. An external magnetic field is used to align the internal magnetic hyperfine field.

New Opportunities of Diffraction Limited Synchrotron Light Sources for Catalysis Research

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Heterogeneous catalysis is the key for the sustainable production of chemicals, clean air and energy conversion technologies. This is also closely related to minimizing CO_2-emissions and the energy transition which is a major challenge and cannot be achieved with the current catalytic processes. New catalyst materials need to be discovered and developed in a knowledge-based way. However, the structure of catalysts steadily changes, especially under working conditions, and usually the active site is part of a complex environment. Synchrotron light is essential for a rational design and with each generation of new synchrotron light sources a new impetus has been found to catalysis.

Examples (see references below) both from chemical energy storage and emission control will be presented demonstrating that the present and upcoming X-ray sources with their complementary portfolio of techniques are required. They allow to cover better and better the various complexity scales in terms of time and length (atomic scale information to mm/cm/m-scale). Hence, both time-resolved, new spectroscopic and new microscopic techniques are required. The understanding of working catalysts can be especially advanced with X-ray techniques due to their high penetration length. The diffraction limited synchrotron light sources and free electron lasers offer new opportunities and at the same time new challenges for this.

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The 1Mpix AGIPD system for the HIBEF user consortium

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The Adaptive Gain Integrating Pixel Detector (AGIPD), a megahertz frame-rate, high-dynamic range integrating pixel detector, was developed for photon science experiments at the European X-Ray Free Electron Laser (European XFEL) and tailored to its unique specifications. Two 1-Megapixel AGIPD detector systems have been installed at the European XFEL and are producing numerous scientific publications. Two completely new AGIPD detector systems are currently being developed for the European XFEL: one 1Mpixel detector for the HED Instrument and one 4Mpixel AGIPD as a second detector for the SPB/SFX instrument. These detectors will be equipped with new readout electronics including all-optical data transmission, a new FPGA, new firmware. A 0.5-Megapixel prototype system, using the new readout electronics, firmware and AGIPD1.2 ASIC has been built, commissioned and operated in user experiments at the HED instrument in 2021 (see figure). This system provided the first Megahertz diffraction capabilities for HED science at the European XFEL. As HED utilizes photon energies in the range of 20-30 keV, at which the quantum efficiency of 500- µm-silicon sensors is small, new high-Z materials such as Gallium-Arsenide (GaAs) and/or Cadmium-Telluride (CdTe) are investigated to be used as sensors. For these sensors, a new ecAGIPD (electron-collecting AGIPD) is developed and a prototype has been tested.

As the presenter joined this project recently, at last, one of his previous projects in the field of optical metrology will be presented. In this work, the Fresnel diffraction of vortex beams from a phase plate is investigated and a novel method to determine the fractional part of the topological charge of vortex beams is proposed. When a vortex beam with a fractional topological charge illuminates the edge region of a transparent plate, the visibility of the diffraction pattern on two sides of the beam will be different. It is because of the summation of the phase difference caused by the fractional topological charge and of the phase plate on one side. The rotation of the phase plate periodically changes the left and right side visibilities of the beam. By measuring three consecutive angles of minimum visibility, the fractional part of the topological charge is obtained. The proposed method is experimentally validated and the result will be obtained independently of the phase plane and vortex beam parameters. The accuracy of the method is better than 0.01 which has been improved at least one order of magnitude.

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Understanding the synthesis and characterization of TiO_2 nanostructures by modified anodization technique using Synchrotron radiations

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[2]

In the present investigation, we have reproduced the conditions for the synthesis of TiO_2 nanopowder (tetragonal crystal structure) from Ti foil (Hexagonal crystal structure) using modified anodization technique. TiO_2 in different phases (anatase, rutile and amorphous) under different washing conditions was prepared using rapid breakdown anodization. The influence of different anodization parameters like, electrode separation, dipped area, applied electric field etc. affect the etch pit formaton on the Ti foil surface, from where the TiO_2 powder emerges out. Using Sychrotron radiation sources, we are planning to study insitu, the mechanism of TiO_2 synthesis from Ti foil surface/edges.

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PETRA IV - The ultimative 3D-X-ray Microscope - General Project Information

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The PETRA IV project at DESY develops an ultra-low emittance storage ring for hard X-ray radiation that will yield three-dimensional insight into matter with unprecedented spatial and temporal resolution. This goal will be achieved by an upgrade of the existing PETRA III facility with recent pioneering developments in accelerator and undulator technologies. The gain in brightness, which is the key parameter to characterise the quality of a light source, will be a factor of 100 for hard X-rays at 10 keV and a factor of almost 1000 at high photon energies up to 150 keV. The high spatial coherence provided by the source, particularly at high photon energies, will enable nearly diffraction-limited focusing capabilities without a severe loss of photon flux. This will result in high sensitivity and high spatial and temporal resolution for *operando* and *in situ* studies of complex systems.
In an international, competitive, landscape with other light sources in USA, China and the rest of the world, PETRA IV plans to restart operation in 2028, keeping Germany and Europe at the forefront of photon science research for the next decades.

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XRnanotech - Achromatic X-ray Lens and recent developments in nanostructured X-ray optics

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At XRnanotech, a spin-off from the Paul Scherrer Institut (PSI) in Switzerland, we develop X-ray optics to enable experiments at many large-scale research facilities. Our goal is to push the limits of diffractive and refractive X-ray optics by continuously, improving the resolution and efficiency enabling new applications in microscopy to make the invisible visible. In X-ray microscopy Fresnel zone plates (FZPs) are used as high-resolution lenses. Their resolution depends mainly on the size of their smallest outermost zones and many years of development were required in order to push this value into ever-smaller regimes.
New procedures have recently also allowed using 3D-printing methods also in the field of X-ray optical applications. For X-ray energies of several keV a good transmission and refractive properties is achieved along with a low surface roughness. Through its versatility 3D-printing allows manufacturing various types of geometries, X-ray optics and (resolution) samples alike. The flexibility of this approach allows achieving previously inaccessible X-ray optical properties.
It has allowed us to achieve achromatic focusing with X-rays using a combination of diffractive and refractive optical elements, which we demonstrated for the first time.
We will also show some of the recent developments in terms of high efficiency, high resolution and structured beam optics.

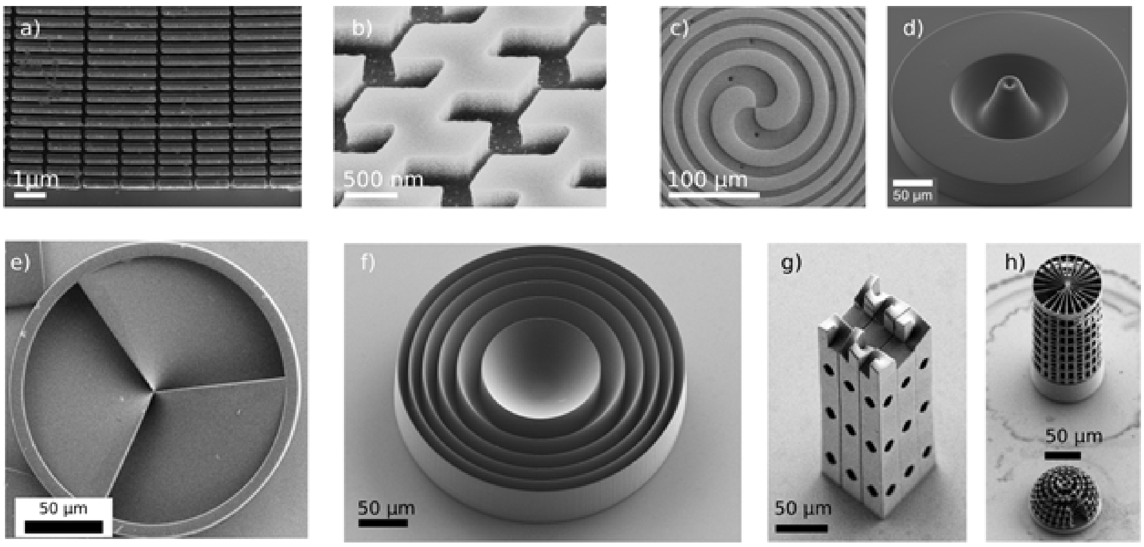

Figure 1: Examples of diffractive and refractive structures as well as resolution test patterns for X-ray applications.

Photoelectron spectrometer for hard X-ray photon diagnostics at the European XFEL

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Development and characterization of an angle-resolved photo-electron spectrometer, based on the electron Time-of-Flight (TOF) concept, designed for hard X-ray photon diagnostics at the European X-ray Free-Electron Laser (EuXFEL) is described. The objective with the instrument is to provide beamline users and operators with pulse resolved, non-invasive spectral distribution diagnostics, which in the hard X-ray regime is a challenge due to the poor cross-section and often very high kinetic energy of photo electrons for the available target gases. In this contribution, we describe tests that were made at beamline P09, PETRA III at DESY.

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Statistical crystallography reveals molecular motions of the main protease from SARS-CoV-2

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The atomic scale motions of proteins make up life at the most fundamental scale and the direct imaging of these motions remains a grand challenge in structural biology. Understanding of protein motion would enable us to understand how proteins fold, how enzymes catalyze chemical reactions, and how signals are communicated via protein matter. Here, the study of an unprecedented number of protein crystals (>10 000), has enabled us to directly measure such information transfer through a protein's structure at the Ångström scale. The crystals, obtained as part of a drug screening campaign at DESY against the main protease (M^{pro}) from SARS-CoV-2, show significant structural variability – even across drug-free datasets obtained under nominally identical conditions. These structural fluctuations from crystal-to-crystal reproduce dynamics similar to what would be seen in solution: they qualitatively reproduce long MD simulations. Further, the "crystal ensemble" allows us to predict which parts of the protein are involved in regulating its function, revealing the inner workings of this nano-scale machine. Our results, based on a dramatic increase in data availability, call into question the notion that a single crystal form results in a single structure, and suggest that it is time to pursue a *statistical* crystallography.

Bridging the gap between static and dynamic compression: New dynamic DAC developments at the Extreme Conditions Beamline P02.2 at PETRA III

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Dynamic compression experiments in the intermediate strain rate regime (10^{-1}-10^{2} s^{-1}) using dynamic Diamond Anvil Cells (dDAC) has attracted much attention over the last decade since it bridges the strain rate gap between static and highly dynamic compression experiments. These experiments have the potential to provide insight into the interpretation of data obtained from high strain rate laser shock or ramp compression experiments, where phase stability fields are often drastically different to those observed in during static compression. Furthermore, it provides the possibility to investigate the kinetics of phase transitions (solid-liquid and solid-solid) as a function of compression rate, which is important to develop a full understanding of crystallization and melting processes at high pressures.
Within this presentation we provide an overview of the dDAC platform that is being developed at the Extreme Conditions Beamline (P02.2) at PETRA III in collaboration with the high-pressure group from LLNL. This set-up combines partially-coherent propagation-based phase contrast imaging and diffraction diagnostics to better understand the processes occurring during a phase transitions driven by varying compression and heating rates.

Establishing operando catalysis photon electron spectroscopy at PETRA IV with the POLARIS setup

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The world's energy landscape needs to be transformed towards a sustainable use of resources. Particularly, since major parts of our society rely on products generated by the chemical industry (plastics, fertilizers etc.). A new foundation for the chemical industry will be based on cheap (solar and wind) electricity. In this hydrogen is produced electrochemically and is reacted into value added products with captured nitrogen and carbon dioxide. The core in these processes are new and still to be developed catalysts working optimally at 1-10 bar and lower than currently employed temperatures. Thus, basic understanding of catalysis under the reaction needs to be established to facilitate these developmental efforts.
As catalytic reactions take place on the surface, developing matching operando characterization methods is imperative. In principle the electronic structure information obtainable from X-ray photoelectron spectroscopy is ideal for this, as it enables an understanding of the (controversially debated) reaction mechanisms at the heart of the reaction. However, transmission of the photoemitted electrons through the reaction gas mixture is challenging because the transmission length is only about 300nm at 10bar.
We propose to utilize the improved foci of PETRA IV to achieve this in the POLARIS experiment and modify the experiment geometry to the required length scale. Additionally, we propose to extend the detection capabilities with simultaneous complementary techniques to address structural properties, i.e., coherent diffraction imaging or small angle X-ray scattering. Furthermore, the new inlet geometry enables simultaneous probing of structured samples. The contribution highlights achievements of POLARIS at PETRA III surmounting the pressure gap, addressing the materials gap and challenging the complexity gap.

Attosecond and few-femtosecond UV-induced dynamics in complex molecules studied via electron–ion covariance spectrometry

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We present a beamline for pump–probe experiments with attosecond or few-femtosecond pulses in three spectral ranges and a spectrometer to distinguish molecular photoreaction paths by utilizing electron–ion covariance data.

"Attochemistry" aims to bridge the gap between electronic processes (e.g. relaxation of inner-valence holes) and photochemistry (e.g. vibration, isomerization and dissociation). Many experiments combine an isolated attosecond pulse in the extreme ultraviolet (XUV) with a near-infrared (NIR) probe-pulse. The main novelty of our beamline is the ability to generate ultraviolet (UV) 4–6 eV pulses with record-breaking few-femtosecond durations[1], through third-harmonic generation in an open gas cell to minimize dispersion. By exciting in the ultraviolet we can study photochemically relevant relaxation dynamics in *neutral* states of many molecules with unprecedented time resolution, probed by either the 5 fs near-infrared (1.7 eV) pulse or the approximately 300 as XUV (15–40 eV) pulse.

High temporal resolution leads to broad spectra, many excited states and dissociation paths. It is therefore useful to record not just the average spectra but single-shot data and analyse the correlations between photoelectrons and fragment ions, conceptually filtering the electron spectrum based on the mass spectrum or vice versa. For this purpose, we have developed a covariance spectrometer[2] combining a 2D velocity map imaging electron spectrometer with a reflectron time-of-flight mass spectrometer. We present results where different parts of the electron spectrum correlate with different fragmentation outcomes, thereby providing additional information to identify the underlying mechanisms and transient states. Combining high mass resolution with the information about energy and angle of the correlated electron opens many possibilities for studying more complex molecules with better-than-vibrational time-resolution.

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Single-shot temporal characterization of XUV SASE FEL Pulses

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The free-electron laser of Hamburg (FLASH) working in the extreme ultraviolet (XUV) and soft X-ray region delivers photon pulses with few-femtosecond (fs) duration and unrivaled intensity [1]. A large fraction of X-ray free-electron lasers (FELs) operate in the self-amplified spontaneous emission (SASE) regime, which means that each pulse has a unique combination of pulse energy, spectrum, arrival time and pulse duration [2]. As it is important to be able to determine the duration and temporal profile of each individual pulse, there have been few methods developed for single-shot temporal analysis [3]. The THz field-driven streaking technique has the potential to deliver single-shot pulse duration information basically wavelength-independent and over a large dynamic range (in pulse duration and FEL energy) [4].

The setup employs a 1030 nm laser and a LiNbO3 crystal to generate single-cycle THz pulses centered at 0.7 THz. The THz pulses are collinearly overlapped with the FEL pulses in a noble gas target. From the interaction of the noble gas atoms and the XUV pulse, photoelectrons are generated and accelerated by the oscillating electric field of the THz pulse. If the XUV pulse is short compared to the oscillation period of the THz field, the XUV temporal profile is mapped onto photoelectron energies. The energy spectrum of the photoelectrons is measured by an electron time-of-flight (TOF) spectrometer. The measured energy spectrum provides the combined temporal and spectral structure of the FEL pulse. The information about the pulse duration can be extracted from the broadening of the peak measured in the photoelectron spectrum due to the presence of the THz field.

Using THz streaking, the single-shot pulse duration has been measured over a wide range from 10 fs to 350 fs (FWHM) [4]. We determined that the XUV pulse arrival time measured with THz streaking was in good agreement with the electron arrival time. The pulse duration determined by THz streaking has been compared with the FEL pulse duration based on spectral analysis. The comparison shows a good agreement for averaged data sets while on a single-shot level both methods are not correlated at all. In addition, to compare the pulse duration measured by THz streaking with another pulse duration diagnostic, a transverse deflecting structure with a variable polarization feature (PolariX TDS) [5] has been used to measure the modulation of the electron bunch [6].

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Hard X-ray Photoelectron Spectroscopy (HAXPES) of Tunable Oxide Interfaces

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The tunability of the oxygen content in complex oxides and heterostructures has emerged as a key to designing their physical functionalities. Controlling the interface reactivity by redox reactions provides a powerful means to deliberately set distinct oxide phases and emerging properties. We present routes on how to control oxygen-driven redox mechanisms in ultrathin ferro(i)magnetic and ferroelectric oxide films and across oxide interfaces [1]. We address the growth and control of metastable EuO oxide phases [2,3], the control of phase transitions of binary Fe oxides by oxygen migration [4,5], the in operando determination of $NiFe_2O_4/SrTiO_3$ interface band alignments, as well as the role of interfacial oxide exchange in ferroelectric HfO_2-based capacitors [6,7] - uncovered by the unique capabilities of photoelectron spectroscopy and, in particular, using hard x-rays.

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ABSTRACT TITLE : Diffraction Limited Synchrotron Light Sources and Next Generation Free Electron Lasers

ABSTRACT

The development of new materials and improvements of existing ones are at the root of the spectacular recent developments of new technologies for synchrotron storage rings and free-electron laser sources. This holds true for all relevant application areas, from electron guns to undulators, x-ray optics, and detectors. As demand grows for more powerful and efficient light sources, efficient optics, and high-speed detectors, an overview of ongoing materials research for these applications is timely. In this article, we focus on the most exciting and demanding areas of materials research and development for synchrotron radiation optics and detectors. Materials issues of components for synchrotron and free-electron laser accelerators are briefly discussed. The articles in this issue expand on these topics.

4th Generation- synchrotron X-ray: New opportunities for advanced crystallography with high resolution detectors.

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I will present the advantages of use of 4th generation synchrotron radiation (SR) source for small- molecule single crystal X-ray diffraction (SCXRD).

Over 100 years of the discovery of the diffraction of X-rays by crystals led some of the great discoveries of modern science, including that of double helix structure of DNA. Having said that, for over 40 years DESY was pioneered in SR SCXRD where the first studies of muscle structure were benefit from its intense light source. Indeed, Beamline P24 at PETRA-III and previously beamlines D3 and F1 at DORIS (now closed) are among the few beamlines across the world devoted full time to the small molecule crystallography and they have great impact in the development of SCXRD in the physical sciences. This technique is still an essential tool for cutting-edge science today, e.g. the study of atomic structure of very complicated structures including incommensurately modulated, quasi crystals, charge density wave compounds, ferroelectric crystal and etc.

Though study of such structures benefit from SR , e.g. tunable energy, narrow beams of highly intense x-rays, the wavelength dependence of the absorption make it difficult to choose shorter wavelength with current PETRA III source and available detectors. For all compounds a short wavelength reduces the absorption. On the other hand, a short wavelength reduces the spatial resolution between individual reflections. The beam properties of the PETRA-IV offer a dramatic increase of possibilities of spatially resolving different Bragg reflections because of the horizontally and vertically well collimated X-ray beam and application of suitable 2D high resolution detectors. The correct interpretation of diffraction pattern will arise to determination of the correct symmetry which will lead to discovery of very excited and interesting materials. Furthermore, the extremely high brilliance of the synchrotron radiation will allow to focus the beam to very small beam sizes without loss of flux. This will allow to obtain diffraction from microcrystals and thus SCXRD will be possible for the most samples with size of 1 to 10 µm for which powder diffraction presently must be chosen.

Atomic-Resolution Bragg Coherent Diffraction Imaging

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Phase retrieval algorithms employed in Bragg coherent diffraction imaging (BCDI) provide a solution to the phase problem of x-ray crystallography. Three dimensional BCDI datasets consist of 3D Bragg peak intensities assembled from two-dimensional diffraction patterns. The inversion of single Bragg peak BCDI datasets yields a volumetric image, which contains a projection of the lattice displacement onto a selected diffraction vector. While the interpretation of the retrieved phases from a single peak is established through a series of assumptions about the physics of the scattering, which are valid under certain conditions, the interpretation of the retrieved phases for atomic resolution datasets is more complex [1, 2]. We outline the existing theory and assumptions for single peak reconstruction, which currently define the applicability of BCDI, as well as current work on combining information from several peaks [3, 4]. Then we interpret the retrieved phases from large regions of reciprocal space covering several Bragg peaks, that will retain the crucial additional information of the relative positions of the Bragg peaks. Measuring such large portions of reciprocal space will be possible at new diffraction limited storage rings such as the upgraded Advanced Photon Source. Here, we derive the differences between single and multi-peak phase retrieval and illustrate the optimal conditions needed to achieve BCDI reconstructions with atomic resolution, and the minimum requirements for realistic datasets from experimental measurements at next generation coherent synchrotron light sources.

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Sub-3 nm Resolution 3D Diffractive Imaging of Anisotropic Gold Nanoparticles with Millions of Patterns using MHz XFEL Pulses

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Single-particle imaging at x-ray free electron lasers (XFELs) has the potential to determine the structure and dynamics of single biomolecules at room temperature. Two major hurdles have prevented this potential from being reached, namely, the collection of sufficient high-quality diffraction patterns and robust computational purification to overcome structural heterogeneity. We report the breaking of both of these barriers using gold nanoparticle test samples, recording around 10 million diffraction patterns at the European XFEL and structurally and orientationally sorting the patterns to obtain better than 3-nm-resolution 3D reconstructions for each of four samples. With these new developments, integrating advancements in x-ray sources, fastframing detectors, efficient sample delivery, and data analysis algorithms, we illuminate the path towards sub-nanometer resolution biomacromolecular XFEL imaging.

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X-ray diffraction from population inverted atoms

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The advent of XFELs opened the path towards ways of controlling x-ray emission and exploring nonlinear and collective emission phenomena. Exploiting the intense x-ray radiation produced by FELs and building up on well consolidated effects such as x-ray lasing and x-ray superfluorescence in the soft [1] and hard [2] x-ray spectral domain we aim at investigate ways of controlling the elastic x-ray scattering response of individual atoms into crystals. Similarly to seeded x-ray stimulated emission, this is achieved by a two-color x-ray pump-probe scheme: the pump pulse prepares atoms in a state of population inversion between inner-shell levels by ionization of an inner electronic state. The probe pulse, tuned on the appropriate inner-shell transition, experiences strong resonant elastic scattering. Under favorable conditions, the atomic scattering factor can reach values few hundred times larger compared to the unpumped atom. In this way, the scattering properties of an element specific sub-ensemble of the crystal can be controlled.

By means of this technique period 4 elements can be converted to scatterers with a large value of imaginary part of the anomalous dispersion correction, thus enabling high-resolution reference-free crystallographic structure solution for the phasing problem without the need for heavy-atom replacement.

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HIKa – Hierarchical Imaging Karlsruhe

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The HIKa (Hierarchical Imaging Karlsruhe) is the new experimental station being constructed in Hamburg from a collaboration between KIT and DESY. This station is being designed as a versatile imaging platform for various purposes including hierarchical imaging, *in vivo* imaging, and high throughput 3D imaging. Also, it is a multipurpose x-ray optical table for experiments on the development of x-ray imaging and microscopy. The station is located at the Ada Yonath Hall on the P23 beamline with a permanent KIT staff coordinating the project. The current construction phase is planned to be completed by the end of 2022, followed by scientific commissioning in 2023.

In the meantime, experiments conducted at P23 already combined the beamline infrastructure at DESY with the expertise from KIT to produce original and new results in the field of x-ray imaging.

With the construction of the HIKa station, DESY may add yet another imaging beamline to its portfolio but with the technical expertise of KIT. Leading to an interesting scenario on future synchrotron source developments.

The complete interface molecular movie: One-stop imaging of electronic and structural dynamics

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Direct imaging of interfacial electronic and structural dynamics in real time requires femtosecond multimodal measurements. Oftentimes separate experiments are required, where keeping experimental conditions identical is challenging. Here, using a dual-messenger combination of time-resolved photoelectron spectroscopy in a single experimental setup, we trace a photoinduced hot charge carrier transfer across a molecule-2D material interface in energy-momentum space and, synchronously, locally at atomic sites. The recorded movie captures atomic rearrangements of the molecules as well as dynamics in the valence and core electrons of both the molecule and the surface. The experiment offers practical all-in-one imaging of charge and energy flow across atomically thin interfaces at their fundamental dimensions.

Understanding Complex Materials and Processes in Nature and Technology with the Ultimate X-ray Microscope PETRA IV

Christian G. Schroer on behalf of the PETRA IV Project Team
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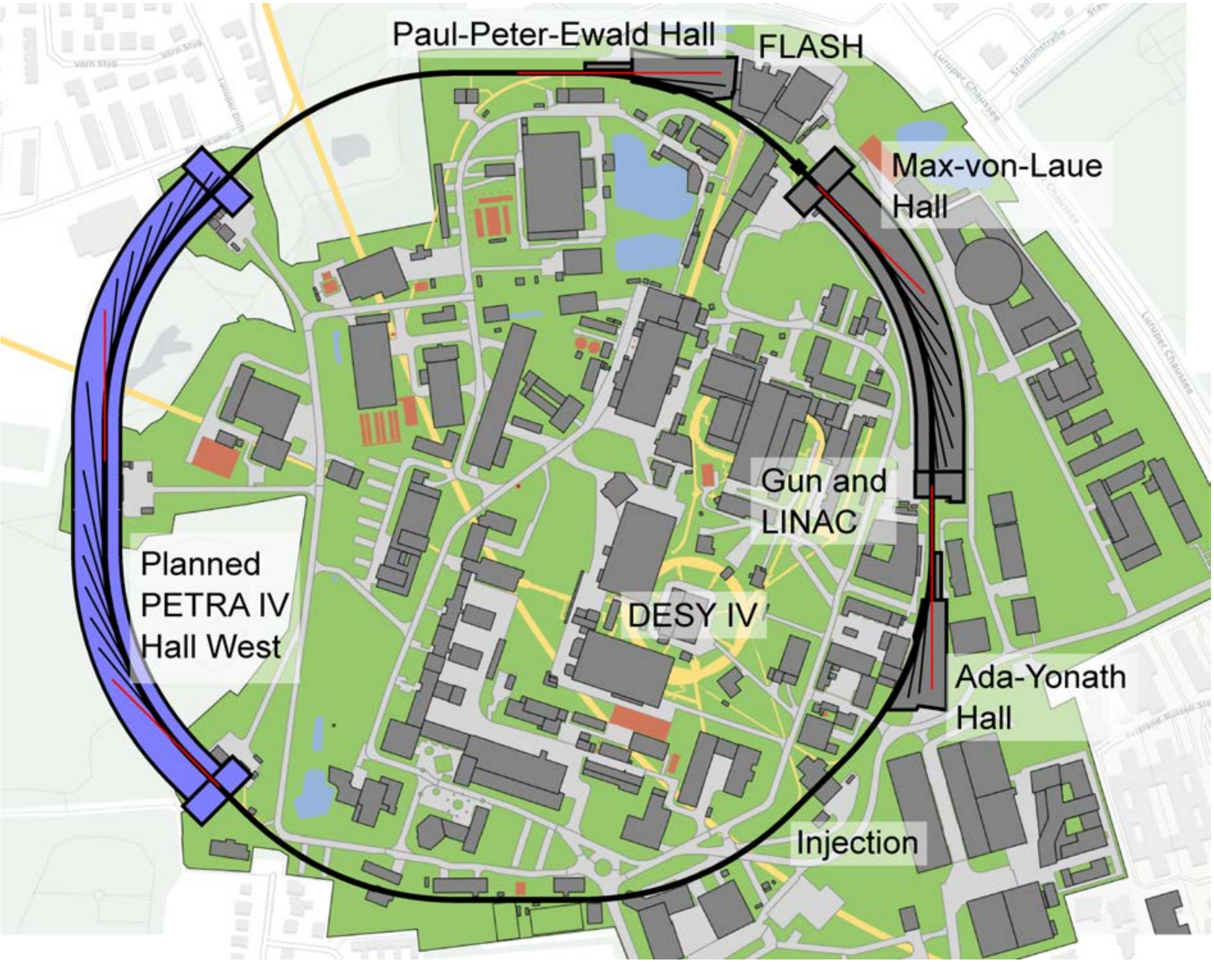
The PETRA IV project aims at upgrading the present synchrotron radiation source PETRA III at DESY into an ultra-low emittance source. Being diffraction limited up to X-rays of about 10 keV, PETRA IV will be ideal for 3D X-ray microscopy of biological, chemical, and physical processes under realistic conditions at length scales from atomic dimensions to millimetres and time scales down to the sub-nanosecond regime. In this way, it will enable groundbreaking studies in many fields of science and industry, such as health, energy, earth & environment, transport, and information technology. The science case is reviewed together with the beamlines and experiments.



With 35 beamlines in four experimental halls, PETRA IV will cater to a large user community in many fields of science.

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Multi-electron Wave Propagation Simulations of Nano-Imaging Beamlines at Diffraction Limited Storage Rings: Opportunities and Challenges

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Diffraction Limited Storage Rings offer unmatched opportunities in X-ray nano-imaging applications as the entire source can be imaged to small spot sizes and the full photon flux can be used in coherent imaging schemes.

However, operating at the diffraction limit also leads to new challenges in beamline designs. Focusing the entire emitted beam into a nanofocus puts much more stringent requirements on the stability of monochromators and other optical components; the effect of imperfections of windows and mirror surfaces are amplified by the high degree of coherence.

Multi-electron wave propagation simulations are the most physically accurate simulation method to address these challenges, while also being the most computationally demanding. The method is suitable for start-to-end simulations, starting from the phase space distribution of the stored electron beam over the insertion device and continuing to all optical components of the beamline, ending in the interaction with the sample and detection. Real-life surface metrology data and stability measurements can be added to the simulation. The influence of individual disturbances to the final image can be isolated in such a digital twin of a beamline.

On this poster, we show the current state of the art and present exemplifying results. We also discuss yet unresolved challenges we are currently facing.

Multimodal spectroscopy with upgraded TRIXS end-station at FLASH

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We report on the capabilities of an upgraded time-resolved resonant inelastic X-ray scattering (TRIXS) end-station at the PG1 monochromator beamline of the soft X-ray/XUV free-electron laser FLASH [1,2]. The new chamber is compatible with the existing TRIXS spectrometer [3,4] for RIXS measurement in the energy range of 35-200 eV, available for users since 2018. The ultrafast time-resolved measurements are performed in conjunction with the new femtosecond optical laser setup (1030 nm + harmonics). The new chamber is equipped with fully motorized laser mirrors for different spectral range (1030-250 nm) which incouples the optical laser with the free electron laser (FEL) in a semi-collinear manner. The chamber comprises necessary diagnostics to establish a temporal overlap between an optical laser and the FEL with 100 fs FWHM accuracy. The presence of variety of detection scheme in the form of photodiodes, microchannel plates, drain current measurements and sample manipulation possibilities will also allow us to perform time-resolved soft X-ray absorption (XAS) and soft X-ray reflectivity (SXR) with 0.06 deg angular resolution on solid samples once the goals of FLASH2020+ project [5] are achieved. Thus, the upgrade of this chamber with cryostat combined, opens up new possibilities to explore dynamics of phase transitions and novel functionalities of different heterostructures, superlattices or multilayer, correlated electron systems with the help of multi-modal techniques. First time-resolved RIXS measurements were already carried out with the new TRIXS chamber and further experiments are envisioned after the restart of FLASH (currently in shutdown), at the end of 2022.

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Synchrotron X-ray microdiffraction on thermal barrier coating revealing multi-phase evolution upon thermal loading

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High-performance thermal barrier coating (TBC) materials at elevated temperatures over 1200°C are intensely searched for enhanced gas turbine engine efficiency. Recently, suspension plasma sprayed $La_2(Al_{1/2}MgTa_{1/2})O_6$ (LAMT) coatings showed significantly improved thermal cycling lifetime results compared to the state-of-the-art yttria partially stabilized zirconia (YSZ) [1]. A detailed crystal structure analysis of this complex rare-earth perovskite and its high-temperature phase transitions were reported using in-situ high-temperature X-ray powder diffraction [2]. Interestingly, the whole TBC coating consisted of a 700 µm-LAMT layer and a 300 µm-YSZ layer underneath remained intact, even though secondary phases were formed after a long-term thermal cycling test [1]. Since a formation of a secondary phase during operation could have introduced an additional stress to the system, leading to an earlier failure, the thermomechanical stability of this double-layered TBC coating was even more surprising. Hence, synchrotron X-ray microdiffraction on the TBC cross-section for both the as-sprayed and the thermally cycled state was conducted. Using a collimated beam of 50 µm (horizontal) x 80 µm (vertical) and an incidence angle of 2°, the cross-section of the TBC coating was rasterized throughout its whole thickness. The degree of crystallinity in the as-sprayed state increases towards the surface and is at the lowest near the substrate, which represents the thermal spraying conditions. Rietveld analysis reveals a coexistence of three different crystalline phases and its continuous evolution in the thermally cycled state as a function of depth. The ratio of the different phases reaches an equilibrium at certain depth in the coating and remains constant down towards the substrate. The tetragonality of YSZ gives a further clue of the operation condition of this double-layered TBC system.

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Hard x-ray microscopy enhanced by coherent image reconstruction

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We present two novel schemes to coherent x-ray microscopy well suited for the current upgrades of synchrotrons to diffraction limited storage rings or even free-electron-lasers.

First, we show a novel approach of full-field holographic x-ray imaging based on cone-beam illumination, but beyond the resolution limit given by the cone-beam numerical aperture. Image information encoded in far-field diffraction and in holographic self-interference is treated in a common reconstruction scheme, without the usual empty beam correction step of in-line holography. An illumination profile tailored by waveguide optics and exactly known by prior probe retrieval is shown to be sufficient for solving the phase problem [1].

Second, we show an approach based on a multilayer zone plate which is positioned behind a sample similar to an objective lens. However, unlike transmission x-ray microscopy, we do not content ourselves with a sharp intensity image, instead we incorporate the lens transfer function directly in an iterative phase retrieval scheme to exploit the large diffraction angles of the small layers. The presence of multiple diffraction orders, which is conventionally a nuisance now comes as advantage for the reconstruction and photon efficiency [2].

In both cases we achieved in first experiments sub-15 nm resolutions and field-of-views of up to 50 micrometers. Altogether, this shows the potential of both approaches for high resolution and dose-effective x-ray imaging well suited for tomography of nanostructures and biological materials.

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Gaussian random field generator SERVAL: a novel algorithm to simulate partially coherent synchrotron radiation

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We propose computationally-efficient algorithm to calculate the field of partially coherent synchrotron radiation pulses from undulators. Wavefront propagation simulations play a pivotal role in designing beamline optics at new synchrotron radiation sources. However, they are complicated by the stochastic behaviour of the initial radiation field, which is due to shot noise in the electron beam. The algorithm we present in this work accounts for shot noise in an electron beam with finite transverse size and divergence, and relies on a method for simulating Gaussian random fields. We initially generate the field as Gaussian white noise, and then we restrict its extent in the direct and in the reciprocal domains by using averaged radiation size and divergence. Strictly speaking, this procedure shapes the correct correlation function of the field but only under the assumption of quasi-homogeneity. However, here we show that the method can be applied with good accuracy also outside of this assumption. Finally, we check consistency of our algorithm with the help of well-established approaches in simulating partially coherent undulator fields.

Quantifying the Role of Defect Dynamics and Dynamic Structural Disorder in Energy Conversion Materials

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Defects and structural disorder are well recognized as crucial for the conversion process in energy materials. However, the precise description of the types and concentrations of defects is an intriguing and complex challenge, requesting the combination of various experimental *in situ* and *post mortem* methods such as laboratory- and synchrotron-based X-ray diffraction, X-ray absorption spectroscopy, X-ray photoelectron spectroscopy, and electrical transport properties with theoretical modeling and simulation techniques. In particular, the changes of the defects and disorder happen in most energy conversion processes during the temperature changes. Three material types are currently in the focus of our research where the defect structure and dynamics are to be understood in detail: i) the change in the occupation of the ideally empty structural site in thermoelectric half-Heusler materials leading to disorder and intrinsic doping effects by the creation of in-gap states in the band structure[1], ii) the transport of oxygen ions and electrons via oxygen vacancies[2,3], and iii) the role of defects on optical properties of perovskite-type halides for photovoltaics.

Besides the effect on the energy conversion, the dynamics of the defects and disorder are expected to play a crucial role for the lifetime of an energy conversion material. Except for photovoltaic and other photo-induced converters, such dynamic effects are studied relatively seldom [4,5]. In addition to the methods mentioned above, time-resolved measurements, e.g., at X-ray free-electron lasers (XFEL) are necessary to get experimental access to the ongoing changes in a material during energy conversion. An extended understanding of the dynamic phenomena is needed in order to enable a conceptional implementation into future material design. This will allow a longer lifetime of the material in a specific energy conversion process helping also to reduce the demand for material resources.

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3-in-1 time-resolved ToF momentum microscopy using FEL and hyperspectral HHG radiation

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Time-resolved photoemission spectroscopy with ultrafast pump and probe pulses is an emerging technique with wide application potential. Combining valence band and core-level spectroscopy with photoelectron diffraction in a single, ultra-efficient photoelectron detection scheme for electronic, chemical and structural analysis requires soft X-ray pulses of few 10 fs duration at some 10 meV spectral resolution. This is feasible at high-repetition-rate FELs. We present an optimized, versatile setup for the use at FLASH as well as a laboratory based high-harmonic generation source that combines short pulsed XUV/soft X-ray capabilities with a multidimensional recording scheme for ultrafast photoemission studies of quantum materials.

Unraveling Time- and Frequency-Resolved Nuclear Resonant Scattering Spectra

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Nuclear Resonant Scattering (NRS) has developed into a versatile tool for probing the local magnetic and charge structure of solids due to the exceptionally narrow line width of Mössbauer nuclei. However, NRS-experiments are in general not sensitive to the phase of the emitted field and require complex model fits to access the spectral and dynamic target structure. Single-line nuclear reference absorbers in collinear geometry (see, e.g., [1,2]) can give direct access to amplitude and phase of the nuclear response and allow one to record time- and frequency-resolved spectra. This has proven crucial in recent experiments on x-ray pulse shaping [3] and coherent control of nuclear dynamics [4], and promises improved insight into the target properties and dynamics via more stringent comparisons between theory and experiment.

Here, we discuss a method to analyze these time- and frequency-resolved spectra utilizing Fourier transforms. Our approach provides intuitive and model-independent access to the spectral structure of nuclear targets and uncovers spectral correlations in the combined scattering system. Further, we propose a phase-cycling scheme that uses existing methods for phase control of collective nuclear excited states [4, 6, 7] to improve the signal-to-noise ratio of these time- and frequency-resolved spectra by decoupling different scattering contributions. We expect this method to be useful not only for spectroscopic applications but also for time-domain problems such as phase-retrieval of the target response (see, e.g. [1,5]). As an outlook, we envision similar phase-cycling schemes to play a comparable role in coherent x-ray multipulse experiments using nuclear targets.

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