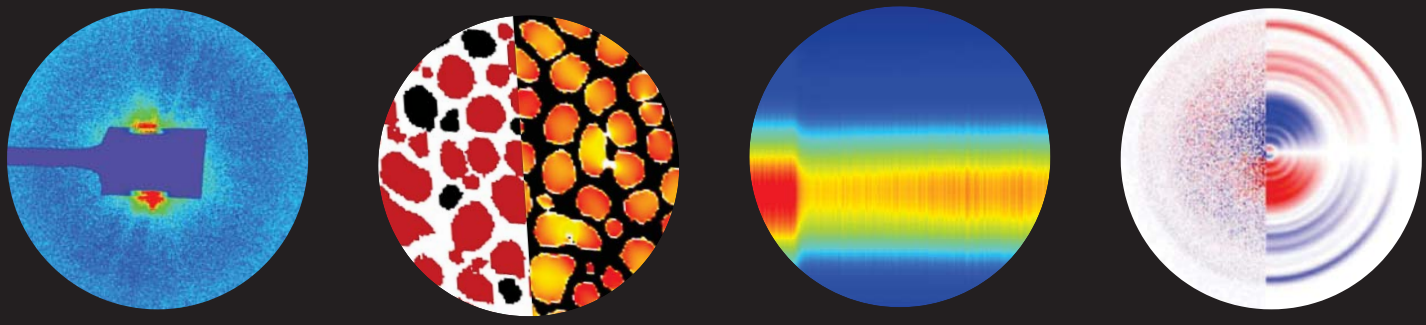
Science@FELs 2024



Venue: Synchrotron SOLEIL & Sorbonne Université - Pierre et Marie CURIE Campus, Paris, France

Dates: 17-21 June 2024

The international Science@FELs 2024 conference will take place from 17-20 June 2024 at Paris Sorbonne and Synchrotron SOLEIL in Paris.

It will focus on recent scientific highlights, related research activities from FEL facilities world wide, features tutorials and the "FELs OF EUROPE award on FEL Science and Applications 2024".

This will be the 7th conference of the Science@FELs series, following the event in 2022. It is organised as a biannual meeting by FELs OF EUROPE, the collaboration of European free electron laser (FEL) facilities and advanced short-pulse light sources (SPS).

Again, the one and a half day "Forum on Advanced FEL Techniques", aiming to bring together FEL experts and FEL users, will take place right after the main conference, from 20-21 June 2024.

Deadlines

FELs of Europe Award application: March 15th, 2024
Oral communication and poster: March 25th, 2024
Invited talk: May 13th, 2024
Registration: April 8th, 2024
Late Registration: May 13th, 2024

Information and registration

Scientific committee

Félicie ALBERT (LLNL)
Majed CHERGUI (EPFL)
Stefan EISEBITT (MBI)
Intae EOM (PAL-FEL)
David LE BOLLOC'H (LPS)
Sebastien LEPAPE (LULI)
Filipe MAIA (Upssala Univseristy)
Claudio MASCIOVECCHIO (Elettra)
Serguei MOLODTSOV (EuXFEL)
Elke PLOENJES PALM (DESY)
Britta REDLICH (Radboud University)
Nina ROHRINGER (MPIK)
Bob SCHOENLEIN (LCLS)
Debora SCUDERI (ICP / CLIO)
Martin WEIK (IBS)
Makina YABASHI (RIKEN)
Junko YANO (LBNL)

Local organising committee

John Bozek (SOLEIL)
Marine Capodano (SOLEIL)
Marie-Emmanuelle Couprie (SOLEIL)
Camille Enjommet (SOLEIL)
Frédérique Fraissard (SOLEIL)
Emmanuelle Jal (LCPMR)
Nicolas Jaouen (SOLEIL)
Jean-Marc Lucacchioni (SOLEIL)
David Massot (LCPMR)
François Mathieu (LULI)
Laurent Nahon (SOLEIL)
Rui Prazeres (ICP/CLIO)
Thierry Ruchon (LIDyL)
Marc Simon (LCPMR)
Cédric Thaury (LOA)
Franck Vidal (INSP)

Contact
conf-scienceatfels2024@synchrotron-soleil.fr

SOLEIL SYNCHROTRON LCPMR FELS OF EUROPE

Science@FELs 2024

17-21 June 2024 | Paris, France

*****

ABSTRACTS BOOKLET

*****

Summary

Science@FELs 2024

17-21 June 2024 I Paris, France

Programme

Monday, June 17th, 2024 – SOLEIL Synchrotron

08:30 – 09:50	Registration, welcome coffee
09:50 – 10:00	WELCOME, ***Jean DAILLANT, SOLEIL General Director***
	Chair: ***Franck VIDAL***
10:00 – 10:50	**Plenary - *Steven Johnson*** *- ETH Zurich / Paul Scherrer Institute, Villigen, Swizerland* X-ray probes of coherence: Tools for understanding coupling and phase transitions in materials
10:50 – 11:10	*Coffee break*
11:10 – 11:30	**Contributed - *Masoud Lazemi*** *- Utrecht University, Netherlands* Real-time and element-specific observation of ultrafast carrier dynamics in $LaFeO_3$ epitaxial thin films by femtosecond X-ray absorption spectroscopy
11:30 – 12:00	**Invited - *Justine Schlappa*** *- European XFEL GmbH, Schenefeld, Germany* Relaxation dynamics in photodoped cuprates from time-resolved Resonant Inelastic X-ray Scattering at European XFEL
12:00 – 12:30	**Invited – *Jan-Christoph Deinert*** *- Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany* Probing complex material properties by THz harmonic generation
12:30 – 13:40	*Lunch at SOLEIL*
13:40 – 14:10	**Invited - *Simon Wall*** *- Department of Physics and Astronomy, Aarhus University, Denmark* Controlling phase transitions with correlated disorder
14:10 – 14:30	***Walking distance / Bus transfer for visits outside SOLEIL***
14:30 – 16:00	**Laboratory Visits** **Walking distance** – SOLEIL, ATTOLAB, APPOLON **Bus transfer** – CLIO
16:00 – 16:30	***Walking distance / Bus transfer to SOLEIL***
16:30 – 19:30	**Posters and appetizers**
19:30 - 20:30	***Optional bus transfer to Paris***

Science@FELs 2024

17-21 June 2024 I Paris, France

Tuesday, June 18th, 2024 – Sorbonne Université

Chair: ***Debora SCUDERI***

09:00 – 09:50	**Plenary - *Jos Oomens* -** *Radboud University, Nijmegen, The Netherland* FEL-based infrared ion spectroscopy: Methods and applications
09:50 – 10:20	**Invited - *Cristian Svetina* -** *Instituto Madrileño de Estudios Avanzados en Nanociencia, Madrid, Spain* Advances of X-ray transient grating at European X-FEL
10:20 – 10:40	***Chair: Manfred HELM*** *FELs of Europe Award*
10:40 – 11:10	*Coffee break*

Chair: ***Jacques-Philippe COLLETIER***

11:10 – 11:40	**Invited - *Jean Yves Salpin* -** *Laboratoire Analyse, Modélisation, Matériaux pour la Biologie et l'Environnement - LAMBE, Université d'Evry Val d'Essonne, France* Gas-phase interactions of platinum drugs towards nucleic acid building blocks
11:40 – 12:00	**Contributed - *Laura Foglia*** *- Elettra Sincrotrone Trieste, Italy* Core-resonant self-diffraction of femtosecond extreme ultraviolet pulses
12:00 – 12:30	**Invited - *Valérie Panneels*** *- Paul Scherrer Institute, Villigen, Swizerland* Ultrafast dynamics of visual rhodopsin using an X-ray free-electron laser
12:30 – 13:40	*Lunch at Sorbonne Université*

Chair: ***Marc SIMON***

13:40 – 14:10	**Invited - *Fabiano Lever*** *- Deutsches Elektronen - Synchrotron DESY, Hamburg, Germany* Electronic molecular movies with FELs: The ultrafast dynamics of 2-thiouracil
14:10 – 14:30	**Contributed - *Christian Bressler*** *- European XFEL, Schenefeld, Germany* Femtosecond solvation dynamics around nascent aqueous halogen atoms Br^0 and I^0
14:30 – 14:50	**Contributed - *Timo Dededrichs*** *- Uppsala University, Sweden* Time-resolved XAS and RIXS of C-H bond activating transition metal complexes in solution
14:50 – 15:20	**Invited - *Michael Meyer*** *- European XFEL, Schenefeld, Germany* Recent results and new developments at the small quantum systems (SQS) instrument at European XFEL
15:20 – 15:40	**Contributed - *Rebecca Boll*** *- European XFEL, Schenefeld, Germany* Imaging a molecular elimination reaction with an X-ray free-electron laser

15:40 – 16:00 *Coffee break*

Chair: ***John BOZEK***

16:00 – 16:30
Invited - *Ulrike Frühling* - *Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany*
Ultrafast AMO experiments at FLASH

16:30 – 16:50
Contributed - *Saikat Nandi* *- CNRS, Institut Lumiere Matiere, France*
Seeded free-electron laser generates quantum entanglement between two massive particles

16:50 – 17:10
Contributed - *Abhishek Verma* *- Synchrotron SOLEIL, France*
Experimental study of post-collision interaction (PCI) in neon after double-core-ionization using the FEL radiation

17:10 – 17:40
Invited - *Ulrich Eichmann* *– Max Born Institute, Berlin, Germany*
Two-color stimulated Raman transitions in atomic systems from the XUV to the soft X-ray regime

17:40 – 18:00
Contributed – *Wieland Schöllkopf* *- Fritz-Haber-*Institut *der Max-Planck-Gesellschaft, Germany*
The 2-color infrared FEL at the Fritz Haber Institute in Berlin

18:00 – 18:20
Contributed - *Oliviero Cannelli* *- CFEL-ATTO, DESY, Switzerland*
On the element-selectivity of core-level transient grating spectroscopies

19:30 *Conference dinner in Paris at* ***La Coupole*** *- 102, boulevard de Montparnasse, 75014 PARIS*
For those who have registered for dinner.

Science@FELs 2024

17-21 June 2024 I Paris, France

Wednesday, June 19th, 2024 – Sorbonne Université

Chair: ***Serguei MOLODTSOV***

09:00 – 09:50	**Plenary -** ***Jacques-Philippe Colletier*** *- Institut de Biologie Structurale, Grenoble, France* Iluminating protein structural dynamics by use of static and time-resolved crystallography at XFELs
09:50 – 10:20	**Invited -** ***Jasper van Thor*** *- Imperial College London, United Kingdom* Control and analysis of coherence in ultrafast X-ray crystallography
10:20 – 10:40	**Contributed -** ***Abhishek Mall*** *- Max Planck Institute for the Structure and Dynamics of Matter - Germany* High-throughput X-ray single particle imaging reveals structure variability and polymorphism in viral capsids
10:40 – 11:10	*Coffee break*

Chair: ***Laurent NAHON***

11:10 – 11:40	**Invited -** ***Minna Patanen*** *- University of Oulu, Finland* Coherent diffractive imaging of salt nanoparticles and sea spray aerosols
11:40 – 12:00	**Contributed –** ***Ltaief Ben Ltaief*** *- Department of Physics and Astronomy, Aarhus University, Denmark* Activation of nanoplasma in pure and doped He nanodroplets by XUV and soft X-ray light pulses
12:00 – 12:30	**Invited -** ***Thierry Ruchon*** **-** *LIDYL, CEA - Saclay, Gif-sur-Yvette, France* Dynamics of magnetic structures probed by magnetic helicoidal dichroism
12:30 – 13:40	*Lunch at Sorbonne Université*

Chair: ***Nicolas JAOUEN***

13:40 – 14:10	**Invited -** ***Emmanuelle Jal*** **-** *Laboratoire de Chimie Physique - Matière et Rayonnement, Paris, France* XFELs to unravel ultrafast magnetic dynamics
14:10 – 14:30	***Contributed - Elisa Collet*** *- IMDEA Nanoscience, Spain* Ultrafast X-ray and optical studies of charge carrier dynamics in colloidal quantum dots
14:30 – 15:00	**Invited -** ***Carl S. Davies*** **-** *HFML-FELIX, Radboud University, Nijmegen, The Netherlands* Ultrafast switching of magnetization at the frequency of optical phonons
15:00 – 15:20	***Contributed - Matthias Riepp*** *- Sorbonne Université, CNRS, Laboratoire de Chimie Physique – Matière et Rayonnement, LCPMR, France* THz-driven coherent magnetization dynamics in a labyrinth domain state

15:20 – 15:40 ***Contributed - Martin Mittendorff*** *- Universität Duisburg-Essen, Germany*
Strong magnetic fields from plasmonic ring currents in graphene disks

15:40 – 16:00 *Coffee break*

Chair: ***Minna PATANEN***

16:00 – 16:30 **Invited -** ***Dominik Kraus*** *- University of Rostock, Germany*
Light elements at Mbar to Gbar pressure

16:30 – 16:50 ***Contributed – Flavio Capotondi*** *- Elettra-Sincrotrone Trieste, Italy*
Time-domain EUV diffuse scattering phonon spectroscopy

16:50 – 17:10 ***Contributed - Vladimir Lipp*** *- Center for Free-Electron Science CFEL, DESY, Germany*
Modeling software SURFwiX to guide high-precision processing of materials of industrial relevance

17:10 – 17:40 **Invited -** ***June Wicks*** *- The Johns Hopkins University, Baltimore, USA*
X-ray illumination of phase transitions through shock compression and release

17:40 – 18:00 ***Contributed - Samuele Pelatti*** *- Università di Modena and Reggio Emilia, Italy*
XFEL-based pump-probe XAS and XES characterization of photoexcited states in CeO_2

18:00 – 18:20 ***Contributed - Zeinab Ebrahimpour*** *- Elettra-Sincrotrone Trieste S.C.p., Italy*
Pioneering plasma accelerators and novel sample delivery methods for future FEL experiments

18:20 End of conference

Science@FELs 2024

17-21 June 2024 I Paris, France

SATELLITE WORKSHOP
Forum on Advanced FEL Techniques
Program

Thursday, June 20th, 2024 – Sorbonne Université

09:00 - 09:10 **Welcome and Introduction** - *Marie-Emmanuelle Couprie, John Bozek, Sverker Werin*

09:10 - 10:20 **Introduction and Overview of high repetition rate FELs -I**

Chair: ***Sverker Werin***

09:10 - 09:40 ***Marc Guetg*** - *DESY / EuXFEL*

Outlook on current high repetition rate FELs

09:40 - 10:00 ***Sandra Mous*** – *SLAC*

Science opportunities at LCLS-II and LCLS-II-HE

10:00 - 10:20 ***Ralf Röhlsberger*** – *EuXFEL*

Perspective for nuclear resonances at high repetition rate FELS

10:20 - 10:50 *Coffee Break*

10:50 - 12:15 **Overview of high repetition rate FELs -II**

Chair: ***Gianluca Geloni***

10:50 - 11:10 ***Immo Bahns*** – *EuXfel*

Status of the cavity based XFEL at European XFEL

11:10 - 11:30 ***David Dunning*** - *ASTEC/UKFEL project*

The UK XFEL Conceptual Design and Options Analysis

11:30 - 12:15 Discussion

Moderators : ***G. Geloni, S. Werin***

12:15 - 14:00 *Lunch at Sorbonne Université*

14:00 - 15:20 **Tailoring pulses I. (attosecond, polarization, seeding..)**

Chair: ***Luca Giannessi***

14:00 - 14:20 ***Carlo Spezzani*** – *FERMI*

Fast polarisation switching, multicolour operation and EEHG seeding at FERMI

14:20 - 14:40 ***Eugenio Ferrari*** – *FLASH*

First EEHG lasing at FLASH and future prospectives

14:40 - 15:00 ***Flavio Capotondi*** - *ELLETRA*

Seeded pulses at FERMI - User perspective

15:00 - 15:20	***Evgeniy Schneidmiller***– *DESY* Using reverse undulator taper for polarization control and short pulse generation in XFELs
15:20 - 15:50	*Coffee Break*
15:50 - 17:15	**Combining multi-colour pulses in FELs** Chair: ***John Bozek***
15:50 - 16:10	***Ziang Li*** – *PITZ* THz FEL at PITZ: Development on high-power accelerator based THz source for pump-probe experiments at the European XFEL
16:10 - 16:30	***Rebecca Boll*** – *EuXFEL* Two-color X-ray pump X-ray probe at FELs: opportunities and challenges
16:30 - 17:15	Discussion Moderators: ***John Bozek, Luca Giannessi***
19:00 - 22:00	*Workshop dinner in Paris at **Au Port Du Salut** - 163 Rue Saint-Jacques, 75005 Paris* *For those who have registered for dinner.*

Friday, June 21st, 2024 – Sorbonne Université

09:00 - 10:40	Chair: ***Marc Guetg*** Tailoring pulses II. Attoseconds and seeding
09:00 - 09:30	***Hugo Marroux*** – *CEA* HHG vs XFEL, strengths and weaknesses
09:30 - 09:50	***Svitozar Serkez*** – *EuXFEL* Towards attosecond pulses at the European XFEL
09:50 - 10:10	***Stephan Kuschel*** - *TU-Darmstadt/LCLS* First imaging experiments with hard and soft sub-fs FEL pulses at LCLS
10:10 - 10:40	Discussion Moderators: ***John Bozek, Marc Guetg***
10:40 - 11:10	*Coffee Break*
11:10 – 13:00	**Compact sources, and an outlook to the future** Chair: ***Marie-Emmanuelle Couprie***
11:10 - 11:40	***Bruce Dunham*** - *Xlight Inc.* Requirements for FEL in industrial applications in lithography

Science@FELs 2024

17-21 June 2024 I Paris, France

11:40 - 12:00	***Marie Labat*** - *Synchrotron Soleil* First results and prospects on Free Laser Electrons driven by Laser Plasma Accelerators
12:00 - 12:20	***Enrica Chiadroni*** – *INFN* EuPRAXIA & SPARC-LAB, the path to FELs with plasma wakefield e-beam acceleration
12:20 - 12:50	Discussion - User perspective compact Moderators: ***Marie-Emmanuelle Couprie, Luca Giannessi***
12:50 - 13:00	***Marie-Emmanuelle Couprie, Sverker Werin*** Closeout summary
13:00	*End*

ABSTRACTS

Monday, June 17th, 2024

POSTERS

Science@FELs 2024

17-21 June 2024 I Paris, France

Monday, June 17th

Chair: ***Franck Vidal***

PT-01	X-ray probes of coherence: Tools for understanding coupling and phase transitions in materials ***Steven Johnson***
OC-01	Real-time and element-specific observation of ultrafast carrier dynamics in $LaFeO_3$ epitaxial thin films by femtosecond X-ray absorption spectroscopy ***Masoud Lazemi***
IT-01	Relaxation dynamics in photodoped cuprates from time-resolved Resonant Inelastic X-ray Scattering at European XFEL ***Justine Schlappa***
IT-02	Probing complex material properties by THz harmonic generation ***Jan-Christoph Deinert***
IT-03	Controlling phase transitions with correlated disorder ***Simon Wall***

X-ray Probes of Coherence: Tools for Understanding Coupling and Phase Transitions in Materials

S.L. Johnson

Institute for Quantum Electronics, Department of Physics, ETH Zurich, Auguste-Piccard-Hof 1, 8093 Zurich, Switzerland
SwissFEL, Paul Scherrer Institut, Forschungstrasse 111, 5232 Villigen PSI, Switzerland

ABSTRACT

The ultrashort x-rays produced by XFELs offer many unique opportunities to study coherent dynamics in materials, offering insights into the coupling between excitations and the dynamics of phase transitions. In this talk I will give an tutorial-level overview of how both elastic and inelastic scattering of x-rays from materials can be influenced by coherent dynamics, and how this can be used in some cases to infer quantitative measures of nonlinearities and coherence associated with important problems in materials. I will highlight these capabilities via recent illustrative examples. These include the decomposition of electromagnons into spin and lattice components[1], the coherent control of 4f orbital wavefunctions[2], the discovery of finite-wavevector polarization fluctuations in $SrTiO_3$[3], and the strong modulation of carrier effective mass in WTe_2[4]. I will also discuss some perspectives for future applications of these methods.

REFERENCES

1. H. Ueda et al., *Nature Commun.* 14, 7778 (2023).
2. R. Mankowsky et al., arXiv:2309.12751 (2023).
3. G. Orenstein et al., arXiv:2403.17203 (2023).
4. D. Soranzio et al., *npj 2D Mater. and Appl.* 6, 71 (2022).

Real-time and Element-specific Observation of Ultrafast Carrier Dynamics in $LaFeO_3$ Epitaxial Thin Films by Femtosecond X-ray Absorption Spectroscopy

M. Lazemi[1], U. Bergmann[2], and F.M.F. de Groot[1]

[1]Materials Chemistry and Catalysis, Debye Institute for Nanomaterials Science, Utrecht University, Universiteitsweg 99, 3584 CG Utrecht, The Netherlands
[2]Department of Physics, University of Wisconsin Madison, 1150 University Ave., Madison, Wisconsin 53706, USA

ABSTRACT

Deciphering the ultrafast carrier dynamics in transition metal oxides (TMOs) is a steppingstone to comprehending their catalytic activity. The emergence of pump-probe techniques at X-ray free electron lasers (XFELs) and high-harmonic generation (HHG) sources has paved the way for the real-time and element-specific measurement of photo-excited carrier dynamics. Femtosecond X-ray absorption spectroscopy (fs-XAS) is emerging as a promising technique that simultaneously reveals electronic and geometric aspects, shedding light on their nonequilibrium dynamic interplay[1–5].
During this presentation, I will highlight the epitaxial (textured) growth of $LaFeO_3$ thin films on single-crystal substrates (SiNx Membranes) by pulsed laser deposition (PLD). Then, I will discuss our recent results on the fs-XAS study of $LaFeO_3$ thin films. This involves employing PAL-XFEL for O 1s and Fe 2p along with the X-FAST[6] for Fe 3p investigations at the University of Wisconsin-Madison.

REFERENCES

1. Park, S. H. *et al.* Direct and real-time observation of hole transport dynamics in anatase TiO2 using X-ray free-electron laser. *Nat. Commun.* **13**, 2531 (2022).
2. Uemura, Y. *et al.* Hole Dynamics in Photoexcited Hematite Studied with Femtosecond Oxygen K-edge X-ray Absorption Spectroscopy. *J. Phys. Chem. Lett.* **13**, 4207–4214 (2022).
3. Uemura, Y. *et al.* Femtosecond Charge Density Modulations in Photoexcited CuWO4. *J. Phys. Chem. C* **125**, 7329–7336 (2021).
4. Ismail, A. S. M. *et al.* Direct observation of the electronic states of photoexcited hematite with ultrafast 2p3d X-ray absorption spectroscopy and resonant inelastic X-ray scattering. *Phys. Chem. Chem. Phys.* **22**, 2685–2692 (2020).
5. Vura-Weis, J. *et al.* Femtosecond M 2,3 -Edge Spectroscopy of Transition-Metal Oxides: Photoinduced Oxidation State Change in α-Fe2O3. *J. Phys. Chem. Lett.* **4**, 3667–3671 (2013).
6. Ash, R. *et al.* X-FAST: A versatile, high-throughput, and user-friendly XUV femtosecond absorption spectroscopy tabletop instrument. *Rev. Sci. Instrum.* **94**, (2023

Relaxation Dynamics in Photodoped Cuprates from Time-resolved Resonant Inelastic X-ray Scattering at European XFEL

J. Schlappa

SCS Instrument, European X-Ray Free-Electron Laser Facility GmbH, Holzkoppel 4, 22869 Schenefeld, Germany

ABSTRACT

Resonant Inelasic X-ray Scattering is powerfull tool for microscopic studies of condensed matter, because can reveal details on charge, spin, orbital and nuclear degrees of freedom. At Free-Electron Laser (FEL) facilities photoexcitation dynamics and also novel transient states can be explored. The Heisenberg RIXS (hRIXS) spectrometer was built in order to enable time-resolved RIXS close to the transfer limit of a monochromatic FEL source [1]. The high-repetition rate of the European XFEL, together with the unique properties of the SCS instrument, provide the ideal conditions to host such an instrument [1-3]. First results demonstrate high stability and feasibility of time-resolved RIXS down to 100 fs in time resolution and up to 10,000 in energy resolving power.

Data on first high-resolution time-resolved RIXS studies from photodoped cuprate systems will be presented. Photodoping offers the option to modify material properties through an optical pump, which is complementary to e.g chemical doping. Photoexcitation above the band gap leads to redistribution of charges in the cupper-oxide planes. The renormalization of low-energy excitations is visible in the tr-RIXS spectra and their dynamics as function of time-delay between the pump and probe reveals details on the transient photodoped state. Potential for tr-RIXS at SCS from complex materials will be discussed and an outlook given.

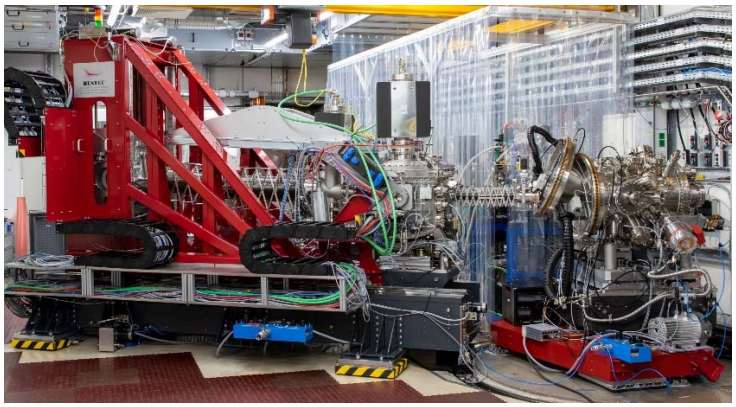


Fig. 1: hRIXS spectrometer and XRD chamber at SCS Instrument

REFERENCES

1. J. Schlappa at al., arXiv:2403,08461 (2024).
2. N. Gerasimova at al., J. Synchrotron Radiat. 29, 1299 (2022).
3. https://www.xfel.eu/facility/instruments/scs/index_eng.html

Probing Complex Material Properties by THz Harmonic Generation

J-C. Deinert[1], I. Ilyakov[1], G. L. Prajapati[1], A. Arshad[1], S. Kovalev[1,2]

[1]*Institute of Radiation Physics, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany*
[2]*Faculty of Physics, Techische Universität Dortmund, 44227 Dortmund, Germany*

ABSTRACT

Intense terahertz (THz) light pulses are a formidable tool to examine many classes of matter. In this low-energy frequency range, the energy from the THz photons leads to strong interactions with, e.g., lattice vibrations, molecular rotations, spin precession and the motion of (quasi-)free electrons. A particularly interesting outcome of this light-matter interaction is the emission of THz harmonics, i.e. the multiplication of the frequency of the incoming THz lightwave. This nonlinear process relies on high THz field strengths, in excess of hundreds of kV/cm, which are routinely generated at the TELBE accelerator-based THz light source at HZDR. The harmonics are not only a very interesting route for making novel THz sources, but they can also give a deep insight into complex material properties and exotic phenomena in matter. In this contribution, I will describe the strategy to use our growing understanding of carrier dynamics in Dirac materials, e.g. graphene[1-3] and topological insulators[4-6], to find the ultimate THz frequency multiplier. Further, I will explain, how the harmonic response in superconducting materials enables a direct view onto oscillations of the superconducting order parameter via the excitation of Higgs modes[7,8]. Finally, I show how complex phase transitions in correlated oxide materials can be traced and examined via the THz harmonic response.

REFERENCES

1. H. A. Hafez, S. Kovalev, J.-C. Deinert et al., *Nature* **561**, 507-511 (2018).
2. J.-C. Deinert, D. Alcaraz Iranzo et al., *ACS Nano* **15**, 1145-1154 (2021).
3. A. Arshad, H. Nur Koyun et al., *Adv. Photonics Res*. **4**, 2300088 (2023).
4. S. Kovalev K.-J. Tielrooij et al., *npj Quantum Mater*. **6**, 84 (2021).
5. K.-J. Tielrooij, A. Principi et al., *Light Sci Appl* 11, 315 (2022).
6. I Ilyakov, A. Brataas et al.,*Nano Lett.* **23**, 3872 (2023).
7. H. Chu, M.-J. Kim et al., *Nat. Commun.* **11**, 1793 (2020).
8. H. Chu, S. Kovalev et al., *Nat. Commun.* **14**, 1343 (2023).

Controlling Phase Transitions with Correlated Disorder

S. Wall

Department of Physics and Astronomy, Aarhus University, Aarhus, Denmark

ABSTRACT

Using vibrational coherence to control the structure of solids has recently been used to drive phase transitions on the ultrafast timescale through the manipulation of long range order[1,2]. However it is unclear if phase transitions that require changes in the local order, such as glass or disorder transition, can benefit from ultrafast control strategies. Here we demonstrate that ultrafast lasers can generate incoherent structural fluctuations with picosecond lifetimes that lower the energy barrier for phase transformation[3]. Seeding inhomogeneous structural fluctuations presents an alternative, more energy-efficient, route for controlling materials that may be applicable to much broader class of phase transitions.

REFERENCES

1. Horstmann et al. Nature 583, 232 (2020)
2. Maklar et al. *Science Advances.* **9**, eadi4661 (2023)
3. Johnson et al. *Nature Physics 10.1038/s41567-024-02474-4* (2024)

POSTERS SESSION

Monday, June 17th, 2024

List of Posters

PO-01	Single-shot temporal characterization of XUV FEL pulses ***M.M. Bidhendi***
PO-02	Small Bandwidth and Short FEL pulses: The new pulse-length preserving double monochromator beamline at FLASH ***G. Brenner***
PO-03	Single-shot double-blind holography of self-amplified spontaneous emission pulses of free electron lasers ***O. Cannelli***
PO-04	Excitation and detection of coherent spin waves at nanoscale wavelength via EUV transient gratings ***P. Carrara***
PO-05	Recent advancements and science capabilities at the FemtoMAX beamline ***J. C. Ekström***
PO-06	Liquid jet development at FERMI ***M. Krstulović***
PO-07	Thermal-emf of $Na_{0.03}Cu_{1.92}S$ nanocomposite alloy ***M.M. Kubenova***
PO-08	MHz gas attenuator design for Shenzhen superconducting soft FEL ***X. Liu***
PO-09	The MagneDyn beamline at the FERMI Free Electron Laser ***M. Malvestuto***
PO-10	Double-core-hole states in neon: Sequential and distinct pathways via multi-photons ***D. Peng***
PO-11	Resonant double core hole spectroscopy of ultrafast decay dynamics in Fe complexes ***J. Schwarz***
PO-12	Mapping the interplay between rotational dynamics and charge transfer ability by velocity map imaging ***B. Senfftleben***
PO-13	Alpha-synuclein fiber diffraction at the EuXFEL ***F. Stellato***
PO-14	Developments of X-ray absorption spectroscopies and related techniques at FXE instrument ***Y. Uemura***

PO-15 Magneto-structural phase transitions of MnAs studied by time-resolved X-ray diffraction

F. Vidal

PO-16 Enhancing thermal management in FEL mirrors through interface optimization: An integrated simulation and experimental approach

X. Wang

Single-shot Temporal Characterization of XUV FEL Pulses

M. Bidhendi[1], G. Goetzke[1], R. Ivanov[2], I. Bermúdez[2], E. Schneidmiller[1], M. V. Yurkov[1] and S. Düsterer[1]

[1]Deutsches Elektronen-Synchrotron (DESY), Notkestr. 85, 22607 Hamburg, Germany
[2]European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany

ABSTRACT

FLASH, the free-electron laser in Hamburg, operates in the self-amplified spontaneous emission (SASE) regime, leading to a unique combination of energy, spectrum, arrival time and pulse duration. Thus, it is critical to be able to determine the pulse duration and arrival time of each pulse. THz field-driven streaking has the potential to deliver single-shot pulse duration information basically wavelength-independent and over a large dynamic range (in pulse duration and FEL pulse energy) [1, 2].
Using THz streaking, the single-shot pulse duration has been measured over a wide range from 10 fs to 350 fs (FWHM) [2] and correlations with other photon beam parameters (e.g. pulse energy, spectral distribution) have been investigated [3]. Furthermore, the study included an examination of the impact of the number of undulators on the pulse duration contributing to lasing, which was compared to results from 1D and 3D FEL simulations [4].
In SASE FELs, non-linear energy modulation of the electron bunch gives rise to natural harmonics of the fundamental wavelength. In this regard, we have conducted measurements and simulations to determine the pulse duration of the third harmonic and to observe its evolution along the undulators.
Using two opposing time-of-flight spectrometers in the THz streaking setup, a potential temporal wavelength chirp within the SASE pulse was quantified and compared to measurements of the electron longitudinal phase space.

REFERENCES

1. I. Grguraš et al., Nat. Photonics 6, 852 (2012)
2. R. Ivanov et al., J. Phys. B 53, 184004 (2020)
3. I. Bermúdez et al., Opt. Express 29, 10491 (2021)
4. M. Bidhendi et al., Appl. Sci. 12, 7048 (2022)

Small Bandwidth and Short FEL Pulses: The New Pulse-length Preserving Double Monochromator Beamline at FLASH

G. Brenner[1], L. Poletto[2], S. Dziarzhytski[1], M. Ruiz-López[1], B. Keitel[1], M. Brachmanski[1], H. Weigelt[1], M. Sinha[1], P. Chopra[1], S. Grunewald[1], M. Degenhardt[1], H. Cankaya[1], S. Alisauskas[1], A. Tajalli[1], V. Hariton[1], C. Mohr[1], U. Grosse-Wortmann[1], S. H. Salman[1], A. Swiderski[1], I. Hartl[1], H. Bienert[1], T.Hans[1], B. Steffen[1], M. Hesse[1], R. Treusch[1], K. Tiedtke[1], S. Düsterer[1], S. Toleikis[1], H. Schulte-Schrepping[1], and E. Plönjes[1]

1 Deutsches Elektronen Synchrotron – DESY, Notkestraße 85, Hamburg 22603, Germany
2 National Research Council – Institute of Photonics and Nanotechnologies, Via Trasea 7, Padova 35136, Italy

ABSTRACT

FLASH, the soft X-ray free-electron laser (FEL) in Hamburg provides high-brilliance ultrashort femtosecond pulses at MHz repetition rate for user experiments. For high resolution spectroscopic and dynamical studies in various research fields a narrow FEL energy bandwidth and ultrashort pulses are a prerequisite. While single grating monochromators provide high-energy resolution they introduce a pulse-front tilt which effectively elongates the longitudinal pulse profile, thus decreasing the time resolution. In order to preserve a short pulse duration and still monochromatize the FEL radiation, the new pulse-length preserving monochromator beamline FL23 at FLASH2 uses a double-grating design. A first grating disperses the radiation and an intermediate slit reduces the spectral bandwidth, a second grating operating in compensating configuration turns back the pulse front tilt, thereby preserving the ultrashort photon pulses.

The open port beamline covers the spectral range between 1.3 nm and 20 nm with a spectral resolving power of approximately 1500 [1]. A bendable Kirkpatrick-Baez mirror system– similar to the one used at the FL24 beamline at FLASH – provides flexible microfocusing at the experiment. A femtosecond optical laser synchronized to the FEL is provided for pumpprobe experiments. The beamline concept and design has been developed using ray tracing simulations and confirmed by wavefront propagation simulations [2]. The commissioning phase was successfully completed and since 2024 the beamline is in FLASH user operation.

Here, results from the technical commissioning of the beamline and its components as well as of first user experiments will be presented.

REFERENCES

1. [1] L. Poletto, F. Frassetto, G. Brenner, M. Kuhlmann and E. Plönjes,, J. Synchrotron Rad. 25, 131-137 (2018); https://doi.org/10.1107/S1600577517013777
2. M. Ruiz-Lopez, L. Samoylova, G. Brenner, M. Mehrjoo, B. Faatz, M. Kuhlmann, L. Poletto and E. Plönjes,, J. Synchrotron Rad. 26, 899-905 (2019); https://doi.org/10.1107/S160057751900345X

Single-shot Double-blind Holography of Self-amplified Spontaneous Emission Pulses of Free Electron Lasers

A. Azzolin[1,2,3], O. Cannelli[2], E. P. Månsson[2], J. Hahne[1,2,3], M. Seitz[1,2], A. bin Wahid[1,2], K.F. Wong[1,2,3], C. Papadopoulou[4], E. Appi[4], U. Fruehling[4], V. J. Yallapraganda[5], P. Biesterfeld[6], P. Mosel[6], S. Fröhlich[6], M. Braune[4], S. Duesterer[4], M. Kovacev[6], U. Morgner[6], R. Moshammer[7], T. Lang[4], C. Heyl[4], D. Oron[8], N. Dudovich[8], C. Ott[7], T. Pfeifer[7], E. Schneidmiller[4], V. Wanie[2], A. Trabattoni[2,6], F. Calegari[1,2,3]

1. Physics Department, University of Hamburg, Hamburg, Germany
2. Centre for Free-Electron Laser Science, DESY, Hamburg, Germany
3. The Hamburg Centre for Ultrafast Imaging, Universität Hamburg, Hamburg, Germany
4. Deutsches-Elektronen Synchrotron, DESY, Hamburg, Germany
5. Indian Institute of Technology Kanpur, IIT Kanpur, Kanpur, India
6. Institute of Quantum Optics, Leibniz Universität Hannover, Hannover, Germany
7. Max-Planck-Institut für Kernphysik, Max-Planck-Gesellschaft, Heidelberg, Germany
8. Weizmann Institute of Science, Rehovot, Israel

ABSTRACT

Ultrafast spectroscopy requires a precise temporal characterization of the laser pulses, which is not feasible directly sampling the waveform in the time domain. Conventional schemes map pulse durations and spectral phase into electron spectra as function of the delay of a probing laser pulse via the interaction with a gas target[1–3]. These strategies require repeating the same experiment at time delays controlled in steps smaller than the pulse duration, which is impractical in the case of self-amplified spontaneous emission (SASE) free electron laser (FEL) pulses due to the high delay jitter, and variability of both spectral content and time duration. The attosecond angular streaking[4] method is more suitable for the reconstruction of FEL pulses, however it still requires the interaction with a gas target and an angularly resolving electron spectrometer, which makes the measurement technically challenging and hard to use routinely.

An all-optical alternative which avoids using light-matter interaction is the so-called double-blind holography (DBH) technique. In the past, we implemented this technique to fully characterize attosecond pulses obtained via High-order Harmonic Generation (HHG)[5]. Here, we extend this method to SASE FEL pulses, demonstrating the possibility to retrieve spectral phase and temporal properties on a single-shot basis. We performed the experiment at the FL26 beamline of FLASH2. The two unknown pulses used for the DBH reconstruction correspond to the SASE FEL pulse and the output of a tabletop HHG setup synchronized with the timing structure of the FEL[6]. The HHG and FEL beams were recombined in a partial spatial overlap geometry to perform a simultaneous measurement of the two separate pulses and of their interference. The reconstructed temporal pulse of the FEL is partially asymmetric and has a time duration (9.48 fs) comparable to the transform limit duration (7.02 fs), showing the high spectral and temporal quality of the delivered FEL pulses.

REFERENCES

1. Paul, P. M. *et al. Science* **292**, 1689–1692 (2001).
2. Tzallas, P., Charalambidis, D., Papadogiannis, N. A., Witte, K. & Tsakiris, G. D. *Nature* **426**, 267–271 (2003).
3. Mairesse, Y. & Quéré, F. *Phys. Rev. A* **71**, 011401 (2005).
4. Hartmann, N. *et al. Nat. Photonics* **12**, 215–220 (2018).
5. Pedatzur, O. *et al. Nat. Photonics* **13**, 91–95 (2019).
6. Appi, E. *et al. Rev. Sci. Instrum.* **92**, 123004 (2021)

Excitation And Detection of Coherent Spin Waves at Nanoscale Wavelength via EUV Transient Gratings

P.R. Miedaner,[1] N. Berndt,[1] J. Deschamps,[1] S. Urazhdin,[2] N. Khatu,[3,4] D. Fainozzi,[3] M. Brioschi,[5,6] P. Carrara,[5,6] R. Cucini,[6] G. Rossi,[5,6] S. Wittrock,[7] D. Ksenzov,[8] R. Mincigrucci,[3] F. Bencivenga,[3] L. Foglia,[3] E. Paltanin,[3] S. Bonetti,[4,9] D. Engel,[7] D. Schick,[7] C. Gutt,[8] R. Comin,[10] K.A. Nelson,[1] A.A. Maznev[1]

[1]Department of Chemistry, MIT, 77 Massachussets Avenue, 02139 Cambridge, MA
[2]Department of Physics, Emory University, 400 Downman Drive, 30322 Atlanta, GA
[3]Elettra Sincrotrone Trieste, S.S. 14 km 163.5, 34149 Basovizza (TS), IT
[4]Department of Molecular Sciences and Nanosystems, Ca' Foscari, 30123 Venice, IT
[5]Dipartimento di Fisica, Università degli Studi di Milano, Via Celoria 16, 20133 Milano, IT
[6]CNR - Istituto Officina dei Materiali (IOM), Unità di Trieste, 34149 Basovizza (TS), IT
[7]Max Born Institute, Max-Born-Strasse 2A, 12439 Berlin, DE
[8]Department of Physics, University of Siegen, Walter-Flex-Str. 3, 57068 Siegen, DE
[9]Department of Physics, Stockholm University, Rogslagstullsbacken 21, 11421 Stockholm, SE
[10]Department of Physics, MIT, 182 Memorial Drive, 02139 Cambridge, MA

ABSTRACT

Low-energy excitations in the solid state (magnons, phonons…) naturally arise as a consequence of spontaneous symmetry breaking and the onset of ordered phases; addressing such excitations is particularly relevant for the investigation of the rich phase diagram of correlated solids. Beyond the optical range, free electron lasers (FELs) allow to explore interactions of extreme ultraviolet (EUV) photons with collective excitations in solids;[1] a suitable experimental approach is transient grating (TG) spectroscopy, a non-collinear four-wave mixing technique that allows to impulsively trigger finite-wavevector excitations,[2] as extensively demonstrated for acoustic phonons.[3]

At FERMI FEL we performed EUV-TG experiments to address coherent magnons in FeGd ferrimagnetic multilayers. Magnons with tens of nanometers wavelengths were excited by a pair of femtosecond EUV pulses and detected via diffraction of a probe pulse tuned to a Gd absorption edge. We measured the magnon dispersion at wavevectors up to 0.12 rad/nm and observed a trend consistent with the prediction of diverging spin-wave stiffness near the ferrimagnetic compensation point.[4] The results unlock the potential of EUV radiation for studying magnons and provide a spectroscopy tool for the wavevector range not covered by existing inelastic scattering techniques.

REFERENCES

1. M. Chergui et al., *Nature Reviews Physics* **5**, 578 (2023).
2. L. Foglia et al., *Photoacoustics* **29**, 100453 (2023).
3. A. A. Maznev et al., *Applied Physics Letters* **119**, 044102 (2021).
4. J. Van Kranendonk & J. H. Van Vleck, *Reviews of Modern Physics* **30**, 1 (1958).

Recent Advancements and Science Capabilities at the FemtoMAX Beamline

J. C. Ekström, B. Ahn, A. Jurgilaitis, D. Kroon, V-T. Pham, J. Larsson

MAX IV Laboratory, Lund University, P.O. Box 118, SE-221 00 Lund, Sweden

ABSTRACT

The FemtoMAX beamline is a unique LINAC-driven time-resolved laser pump/x-ray probe beamline dedicated to study solids and liquids. The beamline is designed to explore dynamics in condensed matter materials at time scales ranging from femtoseconds to microseconds[1]. The sub-50 fs x-ray pulses are generated in two in-vacuum undulators, with a photon energy tunable between 1.8 – 15 keV at a repetition rate of 10 Hz. The femtosecond laser excitation source spans wavelengths of 400 nm-1.6 μm and THz frequencies. A Pilatus time-over-threshold single photon counting detector and a collection of sCMOS detectors are employed to capture SAXS/WAXS and diffraction signals. In addition, ultrashort x-ray pulses in combination with fast detectors can be used to study x-ray time resolved fluorescence from fast scintillators, nanofilms and organic materials.

Here, we present recent advances in providing methods and capabilities to the user community, this includes solution scattering and transient x-ray spectroscopy, which is under development and will be ready for general user experiments year 2024. We also present a novel approach to do timestamping and sorting pump/probe data, based on optical cross-correlation, leading to a significant increase in achievable time resolution.

With these efforts, we are widening the scientific scope of the FemtoMAX beamline and open up for building a large user community.

REFERENCES

1. Enquist Henrik, Jurgilaitis Andrius, Jarnac Amelie, Bengtsson Asa U. J., Burza Matthias, Curbis Francesca, Disch Christian, Ekstrom J. Carl, Harb Maher, Isaksson Lennart, Kotur Marija, Kroon David, Lindau Filip, Mansten Erik, Nygaard Jesper, Persson Anna I. H., Van Thai Pham, Rissi Michael, Thorin Sara, Tu Chien-Ming, Wallen Erik, Wang Xiaocui, Werin Sverker, Larsson Jorgen, FemtoMAX - an X-ray beamline for structural dynamics at the short-pulse facility of MAX IV, JOURNAL OF SYNCHROTRON RADIATION 25 570-579 (2018)

Liquid Jet Development at FERMI

M. Krstulović [1], M. Zamolo[1], G. Bonano[2], A. Gessini[1], C. Soncini[1], D. De Angelis[3], M. Coreno[4], E. Principi[1], C. Masciovecchio[1] and R. Mincigrucci[1]

[1]Elettra - Sincrotrone Trieste S.C.p.A. Strada Statale 14 - km 163,5 in AREA Science Park 34149 Basovizza, Trieste, Italia.
[2]Università degli studi di Modena e Reggio Emilia, Via Università, 4, 41121 Modena MO
[3]IOM CNR, Edificio Q2 Area Science Park Strada Statale 14, km 163,5, 34149 Basovizza TS
[4]CNR-ISM, Basovizza, 320, 34149 Trieste TS

ABSTRACT

FERMI is a high-gain harmonic gerneration (HGHG) seeded free electron laser (FEL) that operates at extreme ultraviolet (EUV) and soft x-ray energy range (SXR) with extremely high brilliance (10^{13} photons/pulse). It produces ultra-short pulses (10 – 100 fs) with high transversal and longitudinal coherence. Its six end-stations are optimized for time resolved research on a broad range of physical and chemical phenomena in various states of matter using spectroscopic, diffraction and scattering techniques.

Most natural liquid systems are composed of light elements with absorption edges at low photon energis. However, probing liquid samples using SXR or EUV radiation requires sub-micrometric sample thickness, which is why fundamental properties of many physical, chemical and biological systems remain unexplored. Probing thin liquid sample without radiation induced sample destruction became possible only recently when sub-micrometric fast- and free-flowing laminar liquid sheets have been created and tested using IR and SXR spectroscopy [1].

At FERMI we are developing two liquid jet setups for experiments in vacuum. One liquid jet setup has already been successfully tested at FERMI and Elettra beamlines [2]. As an independent end-station, it is suitable for experiments at open-ended beamlines. To overcome this limitation, currently a new, portable setup is under development. It has a dedicated miniature chamber with independent vacuum system and it can be positioned inside various experimental chambers, allowing liquid jet experiments also in non open-port end-stations. Current design of the setup will allow the probing of liquid samples by using FERMI (and Elettra) radiation in transmission geometry.

The setup will extend the research capability for FERMI and Elettra user community interested in liquid systems.

REFERENCES

1. J. D. Koralek, J. B. Kim, P. Brůža, C. B. Curry, C. Zhijiang, H. A. Bechtel, A. A. Cordones, P. Sperling, S. Toleikis, J. F. Kern, S. P. Moeller, S. H. Glenzer, D. P. DePonte, *Nat Commun* 9, 1353 (2018).
2. D. De Angelis, L. Longetti, G. Bonano, J. S. Pelli Cresi, L. Foglia, M. Pancaldi, F. Capotondi, E. Pedersoli, F. Bencivenga, M. Krstulović, R. Menk, S. D'Addato, S. Orlando, M. de Simone, R. Ingle, D. Bleiner, M. Coreno, E. Principi, M. Chergui, R. Mincigrucci, and C. Masciovecchio, *accepted*.

Thermal-emf of $Na_{0.03}Cu_{1.92}S$ Nanocomposite Alloy

M.M. Kubenova[1], A.M. Kabyshev[1], R. Alina[1], A.K. Kulanov[1]

[1]*L.N. Gumilyov Eurasian National University, 2 Satpayev str., 010008 Astana, Kazakhstan*

ABSTRACT

The work presents experimental results of the temperature dependence of the electron thermo-emf coefficient of nanocrystalline copper sulfide, the cationic sublattice of which was doped with sodium, with the general chemical formula $Na_{0.03}Cu_{1.92}S$ in the temperature range from 20 ºC to 500 ºC. The sign of the thermo-emf coefficient e was positive, which, taking into account the rule for choosing the sign for all samples, corresponds to the movement of electron holes from the hot end of the sample to the cold one.

The development of semiconductor microelectronics places high demands on the structural perfection and uniformity of properties of semiconductor materials [1]. The main emphasis of the current stage of development is on the decisive role of changing the properties of solids, both through control of structure and as a result of new possibilities for doping elements, regardless of their chemical nature and atomic size. In this regard, improving local methods for monitoring electrophysical parameters in semiconductor materials, knowledge of which is necessary to create devices for thermoelectric purposes, is an equally important task. One of these important parameters is the control of the distribution of charge carrier concentration in the volume of the material. A nanocomposite sample of superionic copper chalcogenide $Na_{0.03}Cu_{1.92}S$ with inclusions of Cu_2O and Na_2S nanoparticles was synthesized.

The electrical conductivity and thermo-EMF of the nanostructured $Na_{0.03}Cu_{1.92}S$ sample were studied using an experimental setup for studying the Seebeck coefficient and electrical resistance (Ulvac ZEM-3, Japan). Electrical resistance is measured by the four-terminal constant current method, in which a constant current (I) is applied to both ends of the sample to measure and determine the voltage drop dV between the same thermocouple leads by subtracting the thermoelectromotive force between the leads.

The Seebeck coefficient and electrical resistance are found using the following formula:

$$\alpha = \Delta V/(T_1 - T_2) \quad (1)$$

where S is the Seebeck coefficient; ΔV – voltage; T_1 – temperature of the bottom sample; T_2 – temperature of the upper sample.

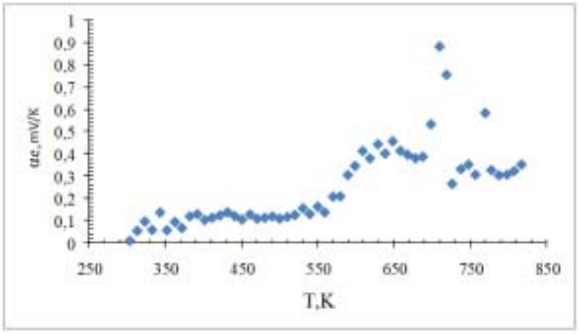


Figure 1 – Temperature dependence of the electron thermo-emf coefficient of $Na_{0.03}Cu_{1.92}S$

In general, the thermo-emf coefficient increases with increasing temperature from room temperature to approximately 650 - 660 K. The highest values of the electron thermo-emf coefficient of 0.9 mV/K are achieved at 700 K. In the vicinity of a temperature of 660 K there is a maximum, apparently, the nature of which is apparently associated with the ongoing phase transition [2,3].

REFERENCES

1 Iokota I. On the Theory of Mixed Conduction with Special Refer-ence to Conduction in Silver Sulfide Group Semiconductors // J. Phys. Soc. Jpn. 1961. Vol. 16. Pp. 2213–2223.

2 Zhang Y.X., Ma Zh., Ge Zh.-H. et al. Highly enhanced thermoelectric properties of $Cu_{1.8}S$ by introducing PbS // Journal of Alloys and Compounds. – 2018. – Vol. 764. – P. 738-744.

3 Kubenova M.M., Kuterbekov K.A., Balapanov M.K., Ishembetov R.K., Kabyshev A.M., Bekmyrza K.Z. Some Thermoelectric Phenomena in Copper Chalcogenides Replaced by Lithium and Sodium Alkaline Metals Review// Nanomaterials.– 2021.– Vol.11.– P.2238.

MHz Gas Attenuator Design for Shenzhen Superconducting Soft FEL

X. Liu [1], Y. Liu[1], Y. Gong[1] and W. Zhang[1]

[1]*institute of advanced science facilities, Shenzhen*

ABSTRACT

The gas attenuator is one of the key devices for free electron laser beamline, which is the capacity of FEL intensity manipulation by adjusting the gas cell pressure. Recently, due to the fast development of FEL, more and more attention has focused on the CW high repetition FEL. In this sense, the stability from the gas attenuator becomes a challenge as the filamentation effect[1] dominates transmission efficiency above 1MHz with mJ pulse energy. To obtain better performance for intensity control for future FEL experiments in high repetition rate, it must redesign a novel gas attenuator.

However, the structure of the gas attenuator has been less mentioned before, the optimization of the structure is promising to decrease the influence of filamentation effect. For example, decreasing the gas cell diameter of the first several meters can effectively reduce the filamentation effect, as the simulation of the filamentation effect reveals the fact that the main heat load is always located at the very beginning of gas cell [2].

Although, the simulation gives the positive result, we will carry out the specific experiment measure for support it soon.

REFERENCES

1. Feng, Y. *et al.* Direct experimental observation of the gas density depression effect using a two-bunch X-ray FEL beam. *J Synchrotron Rad* **25**, 145–150 (2018).
2. Feng, Y. & Raubenheimer, T. O. Duty-cycle dependence of the filamentation effect in gas devices for high repetition rate pulsed x-ray FELs. in (eds. Tschentscher, T. & Patthey, L.) 1023714 (Prague, Czech Republic, 2017). doi:10.1117/12.2261226.

The MagneDyn Beamline at the FERMI Free Electron Laser

M. Malvestuto[a, b,c, *] A. Caretta,[a] R. Bhardwaj,[a] S. Laterza,[a, c] F. Parmigiani,[a, c] A. Gessini,[a] M. Zamolo,[a] F. Galassi,[a] R. Sergo,[a] G. Cautero,[a] M.B. Danailov,[a] A. Demidovic,[a] P. Sigalotti,[a] M. Lonza,[a] R. Borghes,[a] A. Contillo,[a] A. Simoncig,[a] M. Manfredda,[a] L. Raimondi,[a] and M. Zangrando[a, b]

[a]Elettra-Sincrotrone Trieste S.C.p.A. Strada Statale 14 - km 163.5 in AREA Science Park 34149 Basovizza, Trieste, Italy
[b]CNR-IOM Istituto Officina Dei Materiali, Consiglio Nazionale Delle Ricerche, Area Science Park, Strada Statale 14, km 163.5 Trieste, Basovizza, 34149, Italy
[c]Department of Physics, University of Trieste, Via A. Valerio 2, 34127 Trieste, Italy
**marco.malvestuto@elettra.eu*

ABSTRACT

The scope of this communication is to outline the main marks and performances of the MagneDyn beamline [1] at the Free Electron Laser FERMI which was designed and built to perform ultrafast magnetodynamic studies in solids. Open to users since 2019, MagneDyn operates with variable circular and linear polarized femtosecond pulses delivered by the externally laser-seeded FERMI free-electron laser (FEL). The very high degree of polarization, the high pulse-to-pulse stability, and the photon energy tunability in the 50-300 eV range allow to perform advanced time-resolved magnetic dichroic experiments at the K-edge of light elements, e.g. carbon and at the M- and N-edge of the 3d-transition-metals and rare earth elements, respectively. To this end two experimental end-stations are available. The first is equipped with an in-situ dedicated electromagnet, a cryostat, and an extreme ultraviolet (EUV) Wollaston-like polarimeter[2,3]. The second, designed for carry-in users instruments, hosts also a spectrometer for pump-probe resonant X-ray emission and inelastic spectroscopy experiments with a sub-eV energy resolution. A Kirkpatrick–Baez active optics system provides a minimum focus of ~20x20 μm^2 FWHM at the sample. A pump laser setup, synchronized with the FEL-laser seeding system, delivers sub-picosecond pulses with photon energies ranging from the mid-IR to near-UV for optical pump-FEL probe experiments with a minimal pump-probe jitter of few femtoseconds. The overall combination of these features renders MagneDyn a unique state-of-the-art tool for studying ultrafast magnetic phenomena in solids.

REFERENCES

[1] Malvestuto, M. *et al.* The MagneDyn beamline at the FERMI free electron laser. *Rev Sci Instrum* **93**, 115109 (2022).
[2] Caretta, A. *et al.* A novel free-electron laser single-pulse Wollaston polarimeter for magneto-dynamical studies. *Struct Dynam-us* **8**, 034304 (2021).
[3] Laterza, S. *et al.* All-optical spin injection in silicon investigated by element-specific time-resolved Kerr effect. *Optica* **9**, 1333 (2022).

Double-core-hole States in Neon: Sequential and Distinct Pathways via Multi-photons

D. Peng[1]*, I. Ismail[1], A. Verma[1], F. Penent[1], R. Guillemin[1], T. Marchenko[1], O. Travnikova[1], N. Velasquez[1], E. Kukk[6], F. Trinter[7], D. Koulentianos[8], T. Mazza[2], T. Baumann[2], D.E. Rivas[2], Y. Ovcharenko[2], R. Boll[2], S. Dold[2], A. De Fanis[2], M. Ilchen[2], M. Meyer[2], M.N. Piancastelli[1,5], S. Carniato[1], J. Bozek[4], J-P. Rueff[4] R. Püttner[3], and M. Simon[1]

[1] *Sorbonne Université, CNRS, Laboratoire de Chimie Physique Matière et Rayonnement, F-75005, Paris, France*
[2] *European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany*
[3] *Fachbereich Physik, Freie Universit¨at Berlin, Arnimallee 14, D-14195 Berlin, Germany*
[4] *Synchrotron SOLEIL, l'Orme des Merisiers,Saint-Aubin, F-91192 Gif-sur-Yvette, Cedex, France*
[5] *Department of Physics and Astronomy, Uppsala University, SE-75120 Uppsala, Sweden*
[6] *Department of Physics and Astronomy, University of Turku, FI-20014 Turku, Finland*
[7] *Institut f¨ur Kernphysik, Goethe-Universit¨at, Max-von-Laue-Strase 1, 60438 Frankfurt am Main, Germany*
[8] *Center for Free-Electron Laser Science CFEL, Deutsches*
**Contact:* *dawei.peng@sorbonne-universite.fr*

ABSTRACT

Keywords: Double-core-hole, XFEL, distinct pathways, multi-photon process

In this research, our objective is to study DCH states, which are generated by two-photon absorption, through the detection of subsequent Hyper-satellite Auger electrons. The $K^{-2}V$ DCH state can be created in two ways, depending on the photon energy. In the 'intuitive' way (denoted as Pathway A), which was documented in Reference [1], an initial photon ionizes one 1s electron, and a subsequent photon excites the remaining 1s electron into an unoccupied orbital. For neon, a photon energy of around 980 eV is required to facilitate the $Ne1s^1 \rightarrow Ne1s^23p$ transition in the core-ionized Ne^+ ion. Interestingly, a less expected pathway (denoted as Pathway B) is also observed, in which the ionization induced by the first photon is coupled with a shake-up electron excited to an unoccupied orbital. This process was also observed in water molecules[2]. In this pathway,the second step's excitation is predominantly due to the promotion of the second 1s electron into the vacancy orbital left by the shake-up process. These states are detected at around 40 eV lower photon energy compared to pathway A, with the same kinetic energy. This allows the $K^{-2}V$ states in the second step to occur at lower photon energy.

Within the region corresponding to pathway A, we noted the presence of various resonance states, all exhibiting identical photon energy but manifesting differing kinetic energy. We posit that this discrepancy may be attributed to the shake-up/off effect initiated by the Auger electron. For instance, the resonance states at approximately 983 eV photon energy share the same intermediate state, yet they possess distinct kinetic energies, suggesting disparate final states. This phenomenon can be depicted as an initial photon ionizing a 1s electron, followed by a subsequent photon stimulating the remaining 1s electron into an unoccupied orbital (in this case, 3p). An Auger decay process ensues, which is coupled with a shake-up/off transition from 3p to 4p/ε p. In this scenario, the final Auger decay coincides with a shake-up of a valence electron, yielding different final states (3p/4p/ε p) that are differentiated by their respective kinetic energies.

Additionally, we replicated the same measurements under Self-Amplified Spontaneous Emission (SASE) mode, which bypasses the use of a monochromator, and consequently, yields considerably higher photon intensities. We observed the appearance of novel structures within the lower kinetic energy region in the absence of a monochromator. Considering that increased photon intensity typically leads to an elevated cross-section for multi-photon processes, we suppose that these structures may correspond to Ne^+ ion DCH states, generated through a three-photon process.

REFERENCES

[1] Mazza, T. et al., Phys. Rev. X 10, 041056 (2020).
[2] Ismail, I. et al. Phys. Rev. L submitted (2023)

Resonant Double Core Hole Spectroscopy of Ultrafast Decay Dynamics in Fe Complexes

J. Schwarz[1], M. Nissen[1], A. De Fanis[2], A. Rörig[2], K. Baev[5], F. Trinter[4], T. Laarmann[1,6], A. Przystawik[1,6], N. Huse[1], P. Wernet[3], H. Wikmark[3] M. Meyer[2], Y. Ovcharenko[2], Thomas Baumann[2], T. Mazza[2], S. Usenko[2] S. Dold[2], M. Ilchen[1] and M. Martins[1]

[1]Universität Hamburg, Germany [2]European XFEL, Hamburg, Germany [3]Uppsala University,Sweden [4]Fritz-Haber-Institut, Berlin, Germany [5]DESY, Hamburg, Germany [6] The Hamburg Centre for Ultrafast Imaging, Hamburg, Germany

ABSTRACT

Resonant double core hole (DCH) spectroscopy was used to observe ultrafast dynamic processes in small 3d-metal compounds in the gas phase.

Using the small quantum systems (SQS) chamber of European XFEL, 3d metal complexes with iron centers have been investigated with double core hole spectroscopy at the *L*-edge. This represents a next step in the evolution of DCH spectroscopy, as previous results of *L*-edge spectroscopy are rare and stem from synchrotron sources[1]. To achieve DCH spectroscopy, a second core hole electron must be excited within the lifetime of the Auger-Meitner process. Observing DCH excitation products of these systems thus allows access to ultrafast electron dynamics occurring on this short time scale[2]. XFEL transmission was regulated to compare high intensity and low intensity measurements. Ion- and electron-spectroscopy was carried out to detect the product fragments of such photon-matter interactions. Separation of processes due to DCH excitation from those caused by SCH relaxation responses is enabled by kinetic energy shifts in product fragments, which are caused by the presence of the second core hole.

The Auger-Meitner signature of Fe $2p^2$ resonant DCH excitation in iron pentacarbonyl and ferrocene was resolved. Comparing the experimental results to theoretical calculations based on a Hartree-Fock approximation[3], reconstructs single core hole (SCH) and DCH photon-matter interactions in the two targets. The DCH Auger-Meitner electron signals offer insight to the electron dynamics during the core hole lifetime and their dependence on the chemical environment. Evidence for a relaxation bias towards channels involving the resonantly excited electron were found. The product ions show evidence for DCH processes in previously multiply charged iron cations.

REFERENCES

1.Linusson, P., Takahashi, O., Ueda, K., Eland, J.H.D., & Feifel, R. 2011, Physical Review A, 83, 022506, doi:10.1103/PhysRevA.83.022506

2. Rörig, A., Son, S.-K., Mazza, T., et al. 2023, Nature Communications, 14, 5738, doi:10.1038/s41467-023-41505

3. McGuinness, C., Martins, M., Wernet, P., et al. 1999, Journal of Physics B: Atomic, Molecular and Optical Physics, 32, L583, doi: 10.1088/0953-4075/32/20/1

Mapping the Interplay between Rotational Dynamics and Charge Transfer Ability by Velocity Map Imaging

B. Senfftleben[1], F. Allum[2], L. Banares[3], M. Burt[4], I. Casasús[3], T. Chou[4], M. E. Corrales[5], A. de Fanis[1], R. de Nalda[6], A. Duley[7], B. Erk[8], A. Green[1,2], H. Lam[7], X. Li[2], B. Maingot[9], T. Mullins[1], Y. Ovcharenko[1], S. Marggi Poullain[3], D. Raiser[1], P. Recio[3], D. Rolles[7], A. Rouzee[9], A. Rudenko[7], K. Shaw[1,3], S. Usenko[1], E. Warne[4], T. Walmsley[4], E. Wang[7], T. Witting[9], M. Meyer[1] and R. Boll[1]

[1]European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany
[2]SLAC National Accelerator Laboratory, Stanford, CA 94305, United States
[3]Universidad Complutense Madrid, Av. Complutense s/n Ciudad Universitaria 28040 Madrid, Spain
[4]University of Oxford, 12 Mansfield Road Oxford OX1 3TA, United Kingdom
[5]Universidad Autónoma de Madrid, 7. Ciudad Universitaria de Cantoblanco, 28049 Madrid, Spain
[6]Consejo Superior de Investigaciones Científicas, Serrano 119, 28006, Madrid, Spain
[7]Kansas State University, 116 Cardwell Hall Manhattan, KS 66506-2601, United States
[8]DESY Deutsches Elektronen-Synchrotron, Notkestrasse 85 - 22607 Hamburg, Germany
[9]Max-Born-Institut, Max-Born-Strasse 2A 12489 Berlin, Germany

ABSTRACT

Charge transfer is fundamental to many complex chemical processes. Precise modeling requires a detailed understanding of the process at the molecular level. One crucial aspect concerns the interplay between the position of nuclei and the probability of transferring charges. Past experiments studied the dependence of the charge transfer probability on the distance of two molecular fragments [1-4]. Those found that a classical over-the-barrier model [5] is applicable – implying a critical distance beyond which no further charge transfer is possible. Here, we present the results of a recent experiment that took the next step: For dihalomethanes, it is well-known that UV-excitation triggers a dissociation, which subsequently causes the CH_2 group to rotate around the remaining halogen [6,7]. For those molecules, the critical distance may be crossed several times for certain charge states. For this, the classical over-the-barrier model implies a temporary recovery of the ability to transfer charges. The presented data tests for such behavior.

The (in)ability to transfer charges can be measured by site-selective ionization. When no charge transfer is possible, no recoil due to Coulomb forces between the fragments occurs because the other one stays neutral. Consequently, only the ionized fragment is measured with low kinetic energy. We used a velocity map imaging spectrometer with an MCP-phosphor-stack detector and a Timepix3-based optical ns-timestamping camera to isolate the kinetic energy-, delay-, mass-, and charge-dependent signals. With this detection setup, capturing an average of ca. 100 ions per pump-probe-pulse-pair at an intratrain repetition rate of 94 kHz was feasible.

REFERENCES

1. Erk et al., Science 345, 288-291 (2014)
2. Boll et al., Structural Dynamics 3, 043207 (2016)
3. Amini et al., Structural Dynamics 5, 014301 (2018)
4. Allum et al., Faraday Discuss., 2021, 228, 571-596
5. Niehaus, J. Phys. At. Mol. Opt. Phys. 19, 2925–2937 (1986)
6. Burt et al., Phys. Rev. A 96, 043415 (2017)
7. Murillo-Sánchez et al., Phys. Chem. Chem. Phys., 2018, 20, 20766

Alpha-synuclein Fiber Diffraction at the EuXFEL

G. Bais[1], R. Bean[2], J. Bielecki[2], L. Bubacco[3], K. Doerner[2], P. Mariani[4], V. Minicozzi[5], S. Morante[5], M.G. Ortore[4], N. Plotegher[3], M. Polentarutti[1], R. Schubert[2], F. Spinozzi[4] and F. Stellato[5]

[1]Elettra Sincrotrone Trieste, Italy;
[2]European XFEL GmbH;
[3]Università di Padova, Padova, Italy;
[4]Università Politecnica delle Marche, Italy;
[5]Università di Roma ''Tor Vergata'' & INFN, Roma, Italy

ABSTRACT

Parkinson's disease (PD) is the second most common neurodegenerative disease after Alzheimer's disease and the most common movement disorder, affecting nearly 3% of the population over age 70. The current hypotheses of PD pathogenesis invoke multiple mechanisms, but the α-synuclein protein (α-syn), in its oligomeric, aggregated, and fibrillar forms, has taken center stage as potentially toxic. The aggregates are known to be involved in various cellular processes leading to neurodegeneration. Although cell-to-cell transmission of aggregated α-syn has been demonstrated in both cell culture and animal models, the exact molecular mechanisms and evolution of PD's neuropathology remain elusive [Walsh 2016]. Unraveling the structure of α-syn fibers is therefore of the utmost importance from the biomedical point of view.

Here we describe the preliminary results of α-syn fiber diffraction measurements performed at the SPB beamline of the EuXFEL. The fibrils were delivered by injecting the solutions by means of a gas dynamic virtual nozzle, as installed at the EuXFEL, thus leaving them in a condition as close as possible to their functional microenvironment. We acquired millions of diffraction patterns, which are currently being analyzed in order to identify potential hits to be further processed. These screening experiments allowed us to identify stable sample injection conditions and paved the way for further investigations of protein fibers in native conditions.

Developments of X-ray Absorption Spectroscopies and Related Techniques at FXE Instrument

Y. Uemura, P. Zalden, X. Huang, F. Alves Lima, F. Ardana-Lamas, M. Knoll, P. Frankenberger, S. Heder, H. Wang, H. Xu, D. Vinci, Y. Jiang, M. Biednov, S.P. Dutta, H. Yousef, D. Jakobsen, D. Khakhulin, C. Milne

FXE instrument, European XFEL, Holzkoppel 4, 22869 Schenefeld,

ABSTRACT

X-ray Absorption Spectroscopy (XAS) is one of the essential techniques for materials characterisation. Since XAS can be applied to various kinds of systems, such as solid state materials or molecules in solution, XAS has been employed to study photoexcited states of materials at X-ray Free Electron Lasers. The Femtosecond X-ray Exeriments (FXE) instrument is dedicated to the study of photoexcited states of materials using a pump-probe methodology. At FXE non-resonant x-ray emission spectroscopy (nr-XES) has been employed to track photoexcited states of molecules in solution since nr-XES does not require monochromatic x-rays nor energy scanning, measuring non-resonant XES was straightforward. On the other hand, due to the intrinsically stochastic x-ray spectral features of the self-amplified spontaneous emission (SASE) process, using monochromatic x-rays at XFELs has been historically challenging.

Recently we have been focusing on x-ray absorption fine structure (XAFS) measurements. More x-ray photons are available at FXE owing to the burst mode which should translate into XAFS spectra with a good signal-to-noise (S/N) ratio. The XAFS capability is important for our user community. In addition, advanced x-ray spectroscopic measurements such as high energy resolution fluorescence detected x-ray absorption spectroscopy (HERFD-XAS) are also available if the XAFS measurement at FXE is established. In Fig. 1, pump-probe XAFS of $Cu(dmp)_2$ molecules measured at FXE is displayed. X-ray is monochromatized using a Si (111) monochromator and the incident and fluorescence x-ray intensities are measured using photodiodes. The S/N of pump-probe XAS (i.e. $\Delta XAS_{positive\_delay}$/ $\Delta XAS_{negative\_delay}$) is ~100. We will also present our recent achievements at the FXE instrument.

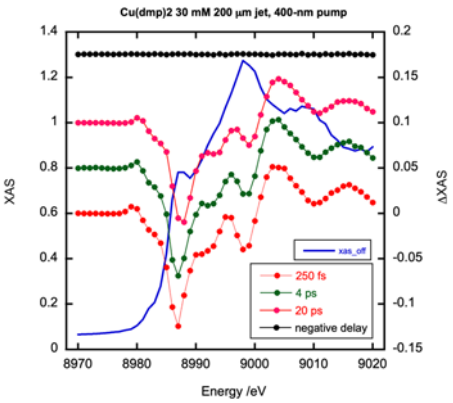


Fig. 1 pump-probe XAFS of $Cu(dmp)_2$ molecules using a liquid jet (*diameter*: 200 µm)

Magneto-structural Phase Transitions of MnAs Studied by Time-resolved X-ray Diffraction

F. Vidal,[1] Y. Zheng,[1] E. Ferrari,[2] E. Allaria,[3] C. Spezzani,[3] C. Laulhé,[4,5] H. Popescu,[5] P. Atkinson,[1] N. Casaretto,[1] M. Eddrief,[1] H. Wang,[6] J. Zhao,[6] M. Seaberg,[7] R. Alonso-Mori,[7] J. M. Glownia,[7] M. Chollet,[7] H. Xu,[8] X. Huang,[8] F. Lima,[8] D. Khakhulin,[8] Y. J. Bermudez Macias,[8] D. Vinci,[8] M. Biednov,[8] Y. Jiang,[8] Y. Uemura,[8] F. Ardana-Lamas,[8] P. Zalden,[8] C. Milne,[8] M. Sacchi [1,5]

1 INSP, UMR7588 CNRS – Sorbonne Université, 4 place Jussieu, 75005 Paris
2 FLASH – Deutsches Elektronen-Synchrotron, D-22607 Hamburg, Germany.
3 FERMI – ELETTRA – Sincrotrone Trieste, S.S.14, Km 163.5, I-34149 Trieste, Italy
4 Université Paris-Saclay, CNRS UMR8502, LPS, 91405, Orsay, France.
5 Synchrotron SOLEIL, L'Orme des Merisiers, Départementale 128, 91190 Saint-Aubin
6 State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, P.O. Box 912, Beijing 100083, People's Republic of China
7 LCLS, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA
8 European XFEL GmbH, Holzkoppel 4, 22869 Schenefeld, Germany

ABSTRACT

MnAs is a magnetic semi-metal with interesting applications in, e.g., magnetic refrigeration and spin-injection. Its properties are strongly linked to an unusual sequence of magneto-structural phase transitions between the α (hexagonal, ferromagnetic), β (orthorhombic) and γ (hexagonal, paramagnetic) phases. DFT calculations suggest that strong spin-phonon coupling is a key ingredient in the mechanism leading to this sequence of phase transitions [1]. The role of a THz soft-phonon mode in the β-γ transition has been shown experimentally [2]. Concerning the α-β transition, recent calculations indicate that it results from the competition between exchange and bonding energies in Mn-Mn orbitals lying in hexagonal planes of MnAs [3].

In this contribution, we will present the results of time-resolved x-ray diffraction measurements performed at FEL sources and discuss how these measurements, together with time-resolved magneto-optical data, can shed light on the mechanisms driving the magneto-structural phase transitions in MnAs.

REFERENCES

1. J. Łażewski et al, Phys.Rev.Lett. 104, 147205 (2010); Phys.Rev. B83, 054108 (2011).
2. F. Vidal et al., Phys.Rev.Lett. 122, 145702 (2019).
3. J. D. Bocarsly et al, Phys. Rev. Res. 2, 042048(R) (2020).

Enhancing Thermal Management in FEL Mirrors through Interface Optimization: An Integrated Simulation and Experimental Approach

WANG Xudong and XU Zhongmin

IASF, Institute of Advanced Science Facilities, Guangming, Shenzhen, P.R.China

ABSTRACT

This study tackles the thermal control challenges in Free-Electron Lasers (FELs), concentrating on refining the interfaces within large-size grazing-incidence mirrors, which are vital for producing high-quality light emissions. Against the background of significant advances in both experimental and computational methodologies [1-4], our work proposes a refined approach to interface optimization, designed to elevate mirror performance further. By integrating molecular dynamics with finite element analysis in a multi-scale simulation framework, we simulate the thermal behaviors at interfaces, aiming to foresee and reduce thermal deformations. This innovative approach provides an in-depth insight into how interface engineering can enhance thermal management, ultimately leading to superior FEL functionality. Moreover, we have innovated a sub-nanometer precision system for detecting thermal deformations, enabling real-time surface monitoring, and offering empirical support for our simulation outcomes and strategies for interface optimization. Our research lays out a streamlined and effective strategy for overcoming thermal challenges in FELs, heralding advancements in light emission quality and system dependability through sophisticated interface management.

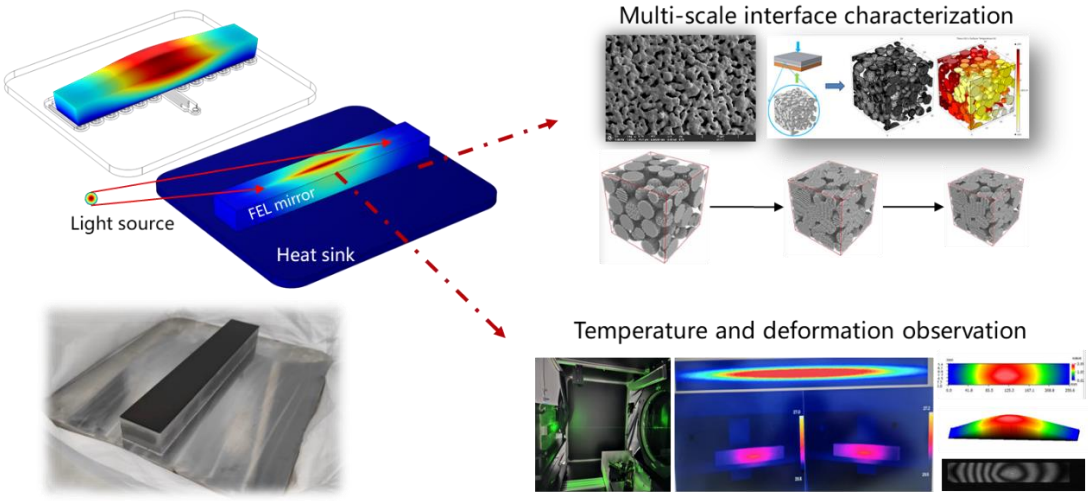


Fig. The combined simulation and experimental approach

REFERENCES

1. Y. Wang, Z. Fan, P. Qian, M. Caro, & T. Ala-Nissila, Phys. Rev. B 107, 054303 (2022).
2. T. Wei, L. Zhang, M. Asheghi, & K. Goodson, J. Phys.: Conf. Ser. 2380 (2022).
3. A. Haber, A. Polo, I. Maj, S. F. Pereira, H. Urbach, & M. Verhaegen, Opt. Express 21(18), 21530-21541 (2013).
4. D. Cocco, C. Hardin, D. Morton, L. Lee, M. L. Ng, Lin Zhang, L. Assoufid, W. Grizolli, Xianbo Shi, D. Walko, G. Cutler, K. Goldberg, A. Wojdyla, M. Idir, Lei Huang, G. Dovillaire, Opt. Express 28, 19242-19254 (2020).

ABSTRACTS

Tuesday, June 18th, 2024

Science@FELs 2024

17-21 June 2024 I Paris, France

Tuesday, June 18th

Chair: ***Debora SCUDERI***

PT-02 FEL-based infrared ion spectroscopy: Methods and applications
Jos Oomens

IT-04 Advances of X-ray transient grating at European X-FEL
Cristian Svetina

Chair: ***Jacques-Philippe COLLETIER***

IT-05 Gas-phase interactions of platinum drugs towards nucleic acid building blocks
Jean Yves Salpin

OC-02 Core-resonant self-diffraction of femtosecond extreme ultraviolet pulses
Laura Foglia

IT-06 Ultrafast dynamics of visual rhodopsin using an X-ray free-electron laser
Valérie Panneels

Chair: ***Marc SIMON***

IT-07 Electronic molecular movies with FELs: The ultrafast dynamics of 2-thiouracil
Fabiano Lever

OC-03 Femtosecond solvation dynamics around nascent aqueous halogen atoms Br^0 and I^0
Christian Bressler

OC-04 Time-resolved XAS and RIXS of C-H bond activating transition metal complexes in solution
Timo Dededrichs

IT-08 Recent results and new developments at the Small Quantum Systems (SQS) instrument at European XFEL
Michael Meyer

OC-05 Imaging a molecular elimination reaction with an X-ray free-electron laser
Rebecca Boll

Science@FELs 2024

17-21 June 2024 I Paris, France

Tuesday, June 18th

Chair: ***John BOZEK***

IT-09	Ultrafast AMO experiments at FLASH ***Ulrike Frühling***
OC-06	Seeded free-electron laser generates quantum entanglement between two massive particles ***Saikat Nandi***
OC-07	Experimental study of post-collision interaction (PCI) in neon after double-core-ionization using the FEL radiation ***Abhishek Verma***
IT-10	Two-color stimulated Raman transitions in atomic systems from the XUV to the soft X-ray regime ***Ulrich Eichmann***
OC-08	The 2-color infrared FEL at the Fritz Haber Institute in Berlin ***Wieland Schöllkopf***
OC-09	On the element-selectivity of core-level transient grating spectroscopies ***Oliviero Cannelli***

FEL-based Infrared Ion Spectroscopy: Methods and Applications

J. Oomens

FELIX Laboratory, Radboud University, Nijmegen, The Netherlands

ABSTRACT

Infrared ion spectroscopy (IRIS) constitutes the powerful myriad of mass spectrometry with IR spectroscopy and is driven by the development of widely tunable IR laser sources. Free-electron lasers have been particularly instrumental in the renaissance of IRIS over the past two decades. I will outline the main aspects of FEL-based IRIS experiments and show how IRIS provides unique opportunities in studies that range from fundamental ion chemistry[1] to astrophysics[2] and bio-analytical chemistry[3].

REFERENCES

1. van Tetering et al. *Comm. Chem.* **7**, 30 (2024)
2. Palotás et al. *Nature Astron.* **4**, 240 (2020)
3. Martens et al. *Anal. Chim. Acta* **1093**, 1 (2020)

Advances of X-ray Transient Grating at European X-FEL

C. Svetina

[1] *IMDEA Nanociencia, Calle Faraday 9, Ciudad Universitaria de Cantoblanco, Madrid, 28049, Spain.*
[2] *European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany*
*cristian.svetina@imdea.org

ABSTRACT

Table-top laser based four wave-mixing techniques, a special class of nonlinear wave-mixing, have provided a major advance in experimental and theoretical physics, chemistry and biology [1]. Transient Grating (TG) spectroscopy is a special case of four wave-mixing employing two crossed laser beams interacting at the sample and generating an interference pattern. A third time-delayed probe beam monitors the time evolution of the induced dynamics that transiently changes the sample's index of refraction. TG is routinely used to gain information on transport and diffusion processes and charge-magnetic-vibrational and quasi-particle dynamics [2]. However optical wavelengths limit the reachable spatial and temporal resolutions while extension to higher photon energies allows to reach nanometer and atto-femtosecond scales adding element and chemical specificity being able to access to K, L and M shells [3]. In the last years a large international community has formed to study extension of TG in the X-rays [4, 5] and provide this method to the scientific community. In the talk I will describe and status of the Long Term Proposal "Dynamics of nanoscale phenomena in solids and liquids studied with X-ray Transient Gratings at European XFEL" [6] sharing the protocols we have developed, showing the latest results and look at the future activities. I will conclude introducing the COST Action NEXT for the development and application of nonlinear wave-mixing methods in the extreme ultraviolet and X-rays [7].

REFERENCES

[1] R.L. Sutherland, *Handbook of Nonlinear Optics*, 2nd Edition, CRC Press (2003)
[2] H.J. Eichler, P. Gunter and D.W. Pohl, *Laser-Induced Dynamic Gratings*, Springer, Berlin (1986)
[3] M. Chergui, M. Beye, S. Mukamel, C. Svetina and C. Masciovecchio, *Nat Rev Phys* **5**, 578–596 (2023)
[4] C. Svetina *et al. Opt. Lett.* **44**, 574–577 (2019)
[5] J.R. Rouxel *et al. Nat. Photon.* **15**, 499–503 (2021)
[6] https://www.nanociencia.imdea.org/home-en/news/item/mixing-x-rays-the-future-of-ultrafast-science
[7] https://www.cost.eu/actions/CA22148/

Gas-phase Interactions of Platinum Drugs Towards Nucleic Acid Building Blocks

J-Y. Salpin

LAMBE – Laboratory Analyse, Modélisation, Matériaux pour la biologie et l'Environnement – UMR 8587 – Université d'Evry Paris-Saclay – Bd F. Mitterrand – 91025 Evry-Courcouronnes, France

ABSTRACT

Cisplatin, [cis-Pt(NH_3)$_2$(Cl_2)]), is the first platinum-based antitumor agent, and it is still widely used in chemotherapy. In spite of its clinical success, cisplatin displays severe toxic side effects and therapy resistance. This has motivated searches for structurally and/or functionally analogous alternatives. Many platinum-based agents have been synthetized during the last 30 years, leading to carboplatin and oxaliplatin as second and third-generation platinum drug, respectively. The aim of our work is to elucidate the intrinsic interactions occurring between the platinum agents and the two nucleobases adenine (A) and guanine (G) and their corresponding nucleotides, known to be the target of this kind of drugs. In this context, gas-phase studies may provide useful insights about the mechanisms occurring at the molecular level, and to determine whether the intrinsic interaction properties of these three drugs are similar in the gas phase.

Aqueous solutions of Pt-drugs and analytes (5'-dGMP) or (5'-dAMP) were mixed together and stored at room temperature for 24 h, to allow the formation of the different complexes. Complexes were generated in the gas phase by electrospray ionization. Their structure was probed by combining mass spectrometry and InfraRed Multiple Photon Dissociation (IRMPD) spectroscopy to DFT calculations. IRMPD spectra were recorded in the 700-2000 cm^{-1} energy range with the CLIO (Centre Laser Infrarouge d'Orsay) free electron laser (FEL), on a FT-ICR instrument (APEX-Qe, Bruker). The XH stretches frequency range (the 3100−3800 cm^{-1}) was recorded by means of a OPO/OPA laser system coupled to an ion trap (Esquire 6000+, Bruker). DFT calculations were carried out using the hybrid B3LYP density functional. Geometry optimization was achieved using the Los Alamos effective core potential (ECP) with the LACV3P** basis set for Pt, and the 6-311G** basis set for the remaining atoms.

This combination of approaches allows operating in the gas phase, thereby allowing overcome any solvent or counter-ion effect and the description of the intrinsic reactivity of the Pt-agents. Excellent agreements were generally observed between the experimental IRMPD spectra and the global minima determined by DFT calculations for the different complexes observed. In the particular case of adenine, differences have been observed concerning the interaction process of carboplatin and oxaliplatin as compared to that of cisplatin.

REFERENCES

1. B. Chiavarino, M. E. Crestoni, S. Fornarini, D. Scuderi, J. Y. Salpin, *J. Am. Chem. Soc.* **135**, 1445-1455 (2013)
2. B. Chiavarino, M. E. Crestoni, S. Fornarini, D. Scuderi, J. Y. Salpin, *Inorg. Chem.* **54**, 3513-3522 (2015)
3. B. Chiavarino, M. E. Crestoni, S. Fornarini, D. Scuderi, J. Y. Salpin, *Inorg. Chem.* **56**, 8793-8801 (2017)
4. B. Chiavarino, L. Rotari, M-E. Crestoni, D. Corinti, S. Fornarini, D. Scuderi, J. Y. Salpin, *Inorg. Chem.* **62**, 14548-14558 (2023)
5. B. Chiavarino, L. Rotari, M-E. Crestoni, D. Corinti, D. Scuderi, J. Y. Salpin, *in preparation*

Core-resonant Self-diffraction of Femtosecond Extreme Ultraviolet Pulses

L. Foglia[1], M. Altarelli[3], F. Bencivenga[1], S. Cushing[5], D. Fainozzi[1], D. Ksenzov[4], W. Lee[5], C. Masciovecchio[1], A. Maznev[2], R. Mincigrucci[1], E. Paltanin[1], A. Rubio[3], N. Tancogne-Dejean[3].

[1] Elettra Sincrotrone Trieste S.C.p.A, Strada Statale 14 km 163.5, 34149 Trieste, Italia
[2] Department of Chemistry, Massachusetts Institute of Technology, Cambridge, MA 02139, USA
[3]Center For Free-Electron Laser Science, Luruper Chaussee 149, 22761 Hamburg, Germany
[4] Department Physik, Universität Siegen, Walter-Flex-Str. 3, 57072, Siegen, Germany
[5] Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, CA 91125, USA

ABSTRACT

The advent of free electron lasers (FELs) enabled the expansion of nonlinear optical spectroscopy methods into extreme ultraviolet (EUV) and x-ray ranges. In particular, four-wave mixing techniques with EUV and EUV/optical fields are being actively developed [1-4]. Self-diffraction is the simplest non-collinear four-wave mixing process, in which the interference of two beams crossed in the sample results in a spatially periodic excitation acting as a diffraction grating for the same beams. Self-diffraction is being widely used in optics [5] has not hitherto been observed in the EUV or x-ray ranges. Generally, the self-diffraction signal from femtosecond EUV FEL pulses is expected to be low, as the refractive index modulation in the EUV range is mainly determined by the average electron density, which does not have time to change significantly within the femtosecond pulse duration. Here we show how the EUV self-diffraction efficiency is greatly enhanced when the EUV photon energy is resonantly tuned to a core absorption edge. We crossed two EUV pulses on a Co thin film and scanned the photon energy across the Co $M_{2,3}$-edge (52 – 74 eV), observing a sharp peak in the self-diffraction efficiency. DFT-based simulations involving a model assumption that the excitation is equivalent to an increase of the electronic temperature are found to reproduce the main peak, but not the fine structure observed above the edge. A similar trend was observed in a subsequent experiment performed at the Si L-edge (95 – 125 eV) on thin membranes of Si, SiC, Si_3N_4 and SiO_2, where the main peak is found to qualitatively shift according to the expected chemical shift. More importantly, in a finer spectrum acquired on Si, clear spectral structures were observed above the edge, with a sharp intensity drop for energies larger than 102.5 eV. This energy corresponds to the onset of the absorption into the continuum, suggesting that the measured spectrum of the self-diffraction intensity can be related to excitonic features.
We believe that the results open the prospect for a new element-specific nonlinear EUV spectroscopy technique.

REFERENCES

1. F. Bencivenga et al., *Advances in Physics: X*, **8** (2023)
2. L. Foglia et al., *Phys. Rev. Lett.* **120**, 263901 (2018)
3. H. Rottke et al., *Sci Adv.* **8**, eabn5127 (2022).
4. R. Mincigrucci et al., *Optica* **10**, 1383-1388 (2023)
5. T. Rappen et al., *Phys. Rev. B* **49**,10774 (1994)

Ultrafast Dynamics of Visual Rhodopsin using an X-ray Free-electron Laser

M. Rodrigues[1], T. Gruhl[1], T. Weinert[1],
Schertler group[1,2], Nango group[3], Neutze group[4],
Nogly group[2], SwissFEL group[5], SACLA group[6], Standfuss group[1],
G. Schertler[1,2], V. Panneels[1]

[1] Division of Biology and Chemistry, Laboratory for Biomolecular Research, Paul Scherrer Institute, Villigen-PSI, Switzerland
[2] Department of Biology, ETH Zurich, Villigen PSI, Switzerland.
[3] RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan.
[4] Department of Chemistry and Molecular Biology, University of Gothenburg, Sweden.
[5] SwissFEL, Paul Scherrer Institute, Villigen-PSI, Switzerlan
[6] Japan Synchrotron Radiation Research Institute (JASRI), Kouto, Sayo-gun, Hyogo, 679-5198, Japan

ABSTRACT

Mammalian rhodopsin is our receptor for vision. It belongs to the highly druggable G protein-coupled receptor family. Upon light illumination, the photoreceptor binds and activates the intracellular G protein transducin, triggering in milliseconds a cascade of signalling events to the brain via the optic nerve. However, the intramolecular initial events transforming the rhodopsin resting state[1-2] (dark state) into the transducin-binding activated state[3-5] (Meta II state) are not completely understood.

Recently, we snapshotted the native bovine rhodopsin at room temperature using time-resolved ultrafast serial femtosecond crystallography, already successfully used for the proton pump bacteriorhodopsin[6-7], at the SACLA and SwissFEL X-ray free electron lasers (XFELs). Thousands of rhodopsin microcrystals grown in the dark are successively injected in the light of a pump laser and probed after various time-delays from femtoseconds to milliseconds using an XFEL. Upon photon absorption, the 11-cis retinal chromophore of rhodopsin undergoes one of the fastest events in biology, which happens in the femtosecond range: its isomerisation into the all-trans conformation. After 1 picosecond, we observe a distorted all-trans retinal that has induced a few changes in its binding pocket while the excess of 480nm-photon energy dissipates anisotropically inside rhodopsin as a protein breathing motion towards the extracellular domain. Complementary analyses like QM/MM and molecular dynamics helped to further characterize our new models. Interestingly, some amino acids known to be key elements in the transduction of the signal are involved in the protein breathing motion.

REFERENCES

[1] Okada et al. J Mol Biol. (2004) 342, 571–583.
[2] Li et al. J Mol Biol. (2004) 343, 1409–1438.
[3] Standfuss et al. Nature (2011) 471(7340):656-60.
[4] Choe et al. Nature (2011) 471(7340):651-5.
[5] Deupi et al. Proc Natl Acad Sci U S A. (2012) 109(1):119-24.
[6] Nango et al. Science. (2016) 354(6319):1552-1557.
[7] Nogly et al. Science. (2018) 13;361(6398).

Electronic Molecular Movies with FELs: The Ultrafast Dynamics of 2-thiouracil

F. Lever

Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany

ABSTRACT

Light-induced ultrafast reactions play a fundamental role in the photophysics of DNA, as they can lead to the formation of lesions in the genetic code. Thionucleobases are sulfur-substituted nucleobases that show an increased rate of UV-induced lesion formation. In a previous work by this group, we have investigated the photoinduced dynamics of 2-thiouracil (2-tUra) via time resolved x-ray photoemission [1,2].
In this work, we study the ultrafast dynamic 2-thiouracil in a UV-pump, x-ray probe experiment at the Free Electron Laser FLASH. The molecule is excited by the pump pulse in the S2 state (ππ* character), and its dynamic is probed by a delayed x-ray pulse that is resonant with 2p and 2s excitations of the sulfur atom. The localized probe on the sulfur allows us to image the excitation of the dark S1 (nπ) state within a 100fs time scale, and to follow oscillations in its population fraction with a period of 200fs.

REFERENCES

1. Mayer et al. Nat Comm 13, 198 (2022)
2. Lever et al. J. Phys. B 54 014002 (2021)

Femtosecond Solvation Dynamics around Nascent Aqueous Halogen Atoms Br^0 and I^0

Z. Nurekeyev[1,2], H. Hwang[1,2], M. Thorwart[3,4], C. Herrmann[4,5], K. Kubicek[1,2], T. Katayama[6], S. Nozawa[7], and C. Bressler[1,2,4]

[1]European XFEL; [2]Dept. of Experimental Physics, Universität Hamburg; [3]Dept. of Theoretical Physics, Universität Hamburg,[4]The Hamburg Centre for Ultrafast Imaging; [5]Dept of Theoretical Chemistry, Universität Hamburg; [6]SACLA, [7]KEK

ABSTRACT

We present sub-picosecond resolved investigations of the structural solvent reorganization and geminate recombination dynamics following photodetachment of a valence p electron from the aqueous ionic solutes I^-(aq) and Br^-(aq). Femtosecond X-ray Absorption Near Edge Structure (TR-XANES) spectroscopy and X-ray Solution Scattering (TR-XSS) complemented picosecond studies at ESRF next to laser-only studies.

For I^-(aq) [1] the XANES measurements at the L1-edge yield an average electron ejection distance of 7.4 ± 1.5 Å. The kinetic traces are in agreement with a purely diffusion-driven geminate recombination model without the need for a long-lived (I^0:e^-) contact pair. This will be compared to the case of geminate recombination for bromine atoms at SACLA.

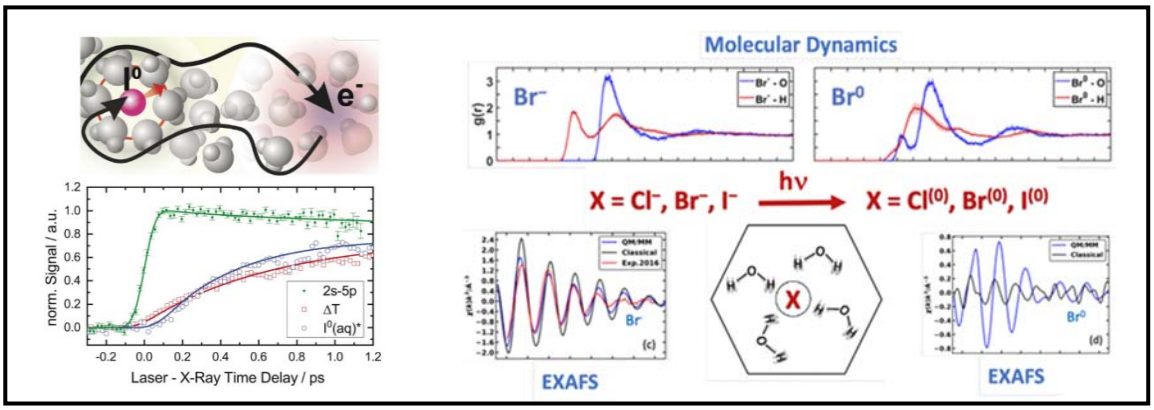


Fig. 1 Left: Halide detachment experiment (top) and femtosecond results (bottom) showing the delayed cage response (blue) against the prompt detachment (green). Right: MD simulations for the analogue bromide detachment experiment showing the nearest neighbor distances (top) and the simulated EXAFS results (bottom).

Non-equilibrium classical molecular dynamics simulations [2] indicate a delayed response of the caging H_2O solvent shell [3] and this is supported by the XSS data [1]: We identify a two-step process exhibiting a 0.1 ps delayed solvent shell reorganization time within the tight H-bond network and a 0.3 ps time constant for the mean iodine-oxygen distance changes. Theoretical and experimental details of this solvation process will be given.

REFERENCES

1. P. Vester, K. Kubicek, R. Alonso-Mori *et al.*, *J. Chem. Phys.* **157**, 224201 (2022).
2. M. Reidelbach, M. Bai, M. Schneeberger *et al.*, *J. Phys. Chem.* B 127, 1399 (2023).
3. M. Reidelbach *et al.*, preprint (2023).

Time-resolved XAS and RIXS of C-H Bond Activating Transition Metal Complexes in Solution

T. Dederichs

Department of Physics and Astronomy, Uppsala University,751 20 Uppsala, Sweden

ABSTRACT

The direct and selective activation and functionalization of carbon (C-H) bonds has been a desire of chemists for a long time. Facilitating C-H activation reactions in a catalytic fashion possesses the transformative power of highly efficient synthesis and sustainable chemical manufacturing. The main challenge of these reactions is the chemical inertness of the C-H bond due to its low polarity [1]. Until now, C-H activation reactions rely on precious metal catalysts in high loadings (e.g. Rh, Ir, Pd) and they must often be conducted at high temperatures [2]. One goal of current C-H activation research is to move towards earthabundant metal catalysts and mild conditions. One approach to achieving the latter is to photoinduced the reactions, where the reactive species is generated upon irradiation with light.Developing strategies for improving photoinduced C-H activation reactions requires understanding of the underlying mechanism and electronic structure changes during such reactions. This allows for optimization of catalyst design and their reactivity.

Motivated by this need, we conducted a series of time-resolved X-ray spectroscopy experiments on C-H bond activating metal complexes in solution at three FELs. Most importantly, we succeeded in measuring time-resolved Rhodium L-edge RIXS spectra of $Cp^*Rh(CO)_2$ in octane solution at SwissFEL [3]. Time-resolved L-edge RIXS of a 4d metal, to the best of our knowledge, was achieved for the first time and allows for an insightful view of the electronic structure of the valence electrons of the metal. This is of particular interest as these are involved in chemical bonding and C-H bond activation. Upon irradiation with UV light, the Rh complex dissociates one carbon monoxide ligand (CO) generating the highly reactive16-valence electron Cp*RhCO species (within 250 fs). This allows for the coordination of an octane molecule with one of its C-H bonds, forming a so-called σ-complex (Cp*RhCO-octane, on a 10 ps time scale). On a time scale of 100 ns, the metal can insert into the C-H bond via an oxidative addition and split the C-H bond to form the final product $Cp^*Rh(CO)\text{-}H\text{-}C_8H_{17}$. Both, the 16-valence electron species and the σ-complex, have been characterized with timeresolved Rh L-edge RIXS. Doing so provides a direct way to the electronic structure of the intermediates and reveals their reactivity.

The lighter and heavier 3d and 5d homologs of the Rhodium complex, $CpCo(CO)_2$ and $Cp^*Ir(CO)_2$, were then investigated at the European XFEL [4] and the LCLS [5], respectively. In contrast to the Rhodium and Iridium complexes, for the cobalt analog, the reaction stops after the formation of the σ-complex. This was ascertained with Co L-edge RIXS signatures showing the formation of an unreactive triplet which inhibits a further C-H bond activation reaction. Comparing our results of $Cp^*Rh(CO)_2$, $CpCo(CO)_2$ and $Cp^*Ir(CO)_2$ enables us formulating explanations for how 3d, 4d, and 5d metals modulate reactivity of C-H bond activation based on electronic-structure descriptors as measured by time-resolved X-ray spectroscopy.

REFERENCES

1. Altus, Kristof M., and Jennifer A. Love. "The continuum of carbon–hydrogen (C–H) activation mechanisms and terminology." Communications Chemistry 4.1 (2021): 173.
2. Dalton, Toryn, Teresa Faber, and Frank Glorius. "C–H activation: Toward sustainability and applications." ACS Central Science 7.2 (2021): 245-261.
3. Raphael Jay, Robert Stefanuik, Victoria Kabanova, Timo Dederichs, Philippe Wernet (Uppsala University), Claudio Cirelli, Camila Bacellar, Philip Johnson, Emma Beale, Florian Dworkowski, Rebeca Castillo (SwissFEL), Antonia Freibert, Nils Huse (University Hamburg).
4. Raphael Jay, Ambar Banerjee, Timo Dederichs, Victoria Kabanova, Philippe Wernet (Uppsala University), Ben van Kuiken, Nahid Ghodrati, Manuel Harder, Zhong Yin, Robert Carley, Laurent Mercadier, Martin Teichmann, Sergii Parchenko, Natalia Gerasimova, Luigi Adriano, Andreas Scherz (EuXFEL), Jessica Harich, Ru-Pan Wang, Moritz Dybowski, Nils Huse (University Hamburg), Erik Nibbering (Max-Born-Institut), Michael Coates, Michael Odelius (Stockholm University).
5. Raphael Jay, Timo Dederichs, Victoria Kabanova, Ambar Banerjee, Philippe Wernet (Uppsala University), Tim van Driel, Pamela Schleissner, Cali Antolini, Mathieu Cholet, Sumana Raj, Shuri Francis, Adi Natan, Marco Reinhard, Thomas Kroll, Amy Cordones, Kelly Gaffney, Roberto Alonso-Mori, Abdullah Kahraman (LCLS/SLAC), Michael Sachs, Elisa Biasin (PNNL).

Recent Results and New Developments at the Small Quantum Systems (SQS) instrument at the European XFEL

M. Meyer

European XFEL, Holzkoppel 4, 20869 Schenefeld, Germany

ABSTRACT

The Small Quantum Systems (SQS) scientific instrument at the European X-ray Free Electron Laser in Germany is dedicated to investigations on atoms, molecules, clusters and nanoparticles. The instrument is installed behind the SASE3 soft X-ray undulator, which produces intense X-ray pulses in the photon energy range between 250 eV and 3000 eV. The accelerator is typically delivering up to 3500 pulses per second to the SQS instrument with pulse energies of up to 10 mJ. These characteristics in combination with pulse durations in the range of 10 to 30 fs provide an ideal basis for numerous investigations involving site-selective core excitations with a special focus on non-linear phenomena and the dynamics of ultrafast processes.

After a short introduction to the SQS instrument and its experimental capabilities, the talk will focus on results of recent investigations providing new insight into non-linear processes and multi-photon excitations, especially on the formation of double-core holes, on the complex pathways and dynamics of atomic multiple ionization as well as on resonant excitation processes in highly charged ions. In addition, examples of time-resolved experiments will be discussed giving access to dynamical information on the processes under study by the combined use of X-ray FEL and optical lasers pulses or of two X-ray FEL pulses of different photon energies. Finally, some new opportunities and applications will be presented related to recent developments for accessing the attosecond temporal domain for time-resolved experiments and generating X-ray pulses with variable (linear and circular) polarization for experiments on oriented and chiral systems.

Imaging a Molecular Elimination Reaction with an X-ray Free-electron Laser

R. Boll on behalf of the EuXFEL beamtime 2676 collaboration

European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany

ABSTRACT

The short and intense X-ray pulses from free-electron lasers are an exquisite tool for Coulomb explosion imaging (CEI) [1–3]. Snapshot images of the complete structure of complex molecules, including all hydrogens, can be captured. The very rapid charge-up leads to a violent Coulomb explosion that preserves the information about the molecular structure at the instant of ionization. This allows studying processes such as the influence of transient molecular resonances [4], intramolecular charge rearrangement [1] and molecular fragmentation dynamics [3].

The multidimensionality of CEI can allow to specifically investigate certain aspects of molecular structural dynamics. We recently demonstrated how X-ray induced CEI can be used to trace a molecular elimination reaction, a minority reaction channel that involves the breaking of two molecular bonds and the formation of a new one [5]. Simultaneously, we mapped light-induced bending vibrations of the bound molecular wave packet, disentangled different dissociation pathways, and directly imaged the correlated dynamics leading to ejection of a newly formed molecular fragment.

REFERENCES

[1] R. Boll et al. Nat. Phys. 18, 423 (2022)
[2] X. Li et al. Phys. Rev. Res. 4, 013029 (2022)
[3] T. Jahnke et al. Phys. Rev. X 11, 041044 (2021)
[4] X. Li et al. Phys. Rev. A 105, 053102 (2022)
[5] X. Li et al., in preparation (2024)

Ultrafast AMO Experiments at FLASH

U. Frühling

Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85,22607 Hamburg, Germany

ABSTRACT

FLASH is an XUV and soft X-ray FEL offering femtosecond light pulses with high repetition rates[1]. It is equipped with XUV split-and-delay units, synchronized pump-probe lasers and in 2020 a synchronized laser based (HHG) VUV source[2,3] has been added to the FL26 endstation offering unique multi-color options for pump-probe experiments.

I will give an overview of the facility and present recent results from the AMO endstations at FLASH including coincidence experiments in atoms and molecules, transient absorption spectroscopy and XUV/VUV pump-probe experiments.

REFERENCES

1. K. Tiedke et al., *New Journal of Physics*, **11**, 023029 (2009).
2. E. Appi et al., *Scientific Reports*, **10**, 6867 (2020).
3. E. Appi et al., *Review of Scientific Instruments*, **92**, 1230004 (2021).

Seeded Free-electron Laser Generates Quantum Entanglement between Two Massive Particles

S. Nandi

Univ Claude Bernard Lyon 1, CNRS, Institut Lumière Matière, F-69622, Villeurbanne, France

ABSTRACT

Generating entanglement between the degrees of freedom typically encountered in the classical world is a challenging task because particles can easily interact with the surrounding environment [1]. In this regard, femtosecond pulses at extreme-ultraviolet (XUV) wavelengths from a seeded free-electron laser provide a unique opportunity to study coherent light-matter interaction across ultrafast timescales [2]. Here, using intense XUV pulses from FERMI [3], we were able to generate entanglement in a novel quantum bipartite system: specifically, a photoelectron – a free-moving particle, and a light-dressed photo-ion – a hybrid state of light and matter [4]. In the absence of quantum entanglement between these two particles, the measured photoelectron spectra increases monotonically as dictated by Einstein's photoelectric equation (Fig. 1A). However, if they are entangled, the photoelectron spectra develop a clear avoided crossing originating from the dressed-state dynamics in the atomic ion (Fig. 1B). Following emission into the continuum, the photoelectron wave-packet expands up to a distance of almost 200 nm, allowing us to reveal the existence of mesoscopic entanglement in XUV-induced photoionization processes. We show that by adjusting the intensity of the XUV pulse, one can control the manifestation of the entanglement. Finally, we demonstrate that entanglement between unconfined degrees of freedom is not immediately generated; instead, it requires a particular amount of time – in our case, one Rabi period, for the entanglement to develop fully. By calculating the von Neumann entropy of entanglement, we could show that once developed, the entanglement persists [4]. Our findings can be valuable for the community, as improving light sources allows not only for observation but also for the characterization and potential exploitation of entanglement in these ultrafast processes at short wavelengths.

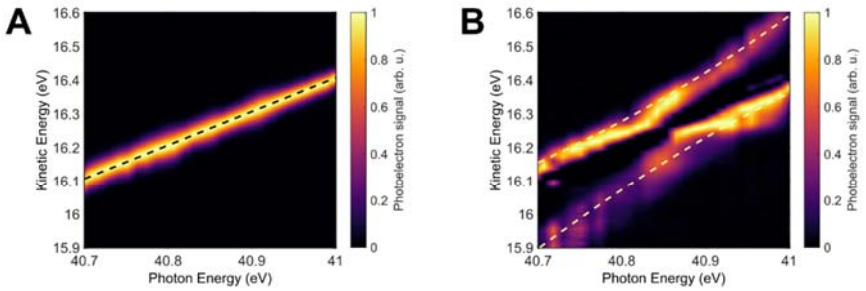


Figure 1. (A) Photoelectron spectra when the XUV-dressed ion and the photoelectron are not entangled. (B) The same spectra when these two particles are entangled [4].

REFERENCES

1. J. D. Jost, *et al*., Entangled mechanical oscillators. *Nature* **459**, 683-685 (2009).
2. S. Nandi, *et al*., Observation of Rabi dynamics with a short-wavelength free-electron laser. *Nature* **608**, 488-493 (2022).
3. E. Allaria, *et al*., Highly coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet. *Nature Photonics* **6**, 699-704 (2012).
4. S. Nandi, *et al*., Generation of entanglement using a short-wavelength seeded free-electron. *Science Advances* (**in press**, 2024). Preprint: arXiv:2312.04442 [quant-ph]

Experimental Study of Post-collision Interaction (PCI) in Neon after Double-core-ionization using the FEL Radiation

A. Verma[1,2*], D. Peng[1], R. Guillemin[1], I. Ismail[1] R. Püttner[3], D. Koulentianos[4], T. Marchenko[1], O. Travnikova[1], M. N. Piancastelli[1], S. Carniato[1], F. Trinter[7], G. Doumy[5], L. Young[6], S.H. Southworth[6], A. de Fanis[5], T. Baumann[5], R. Boll[5], D. Rivas[5], E. Kukk[7], T. Mazza[5], M. Meyer[5], J. Bozek[2] and M. Simon[1]

[1] *Sorbonne Université, CNRS, Laboratoire de Chimie Physique Matière et Rayonnement, F-75005, Paris, France*
[2] *Synchrotron SOLEIL, l'Orme des Merisiers,Saint-Aubin, F-91192 Gif-sur-Yvette, Cedex, France*
[3] *Fachbereich Physik, Freie Universitat Berlin, Arnimallee 14, D-14195 Berlin, Germany*
[4] *Center for Free-Electron Laser Science CFEL,Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany*
[5] *European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany*
[6] *Advanced Photon Source, Argonne National Laboratory, Lemont, Illinois 60439, USA*
[7] *Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany*
[8] *Department of Physics and Astronomy, University of Turku, FI-20014 Turku, Finland.*
Contact: abhishek.verma@sorbonne-universite.fr

ABSTRACT

Keywords: Post-collision interaction, core photoionization, relaxation, XFEL radiation, double corehole.

Photoionization of an atom at an energy close to the ionization threshold of an inner shell produces a slow photoelectron and the subsequent relaxation process emits a high kinetic energy Auger electron [1], leaving the atom positively charged. The photoelectron and the Auger electron strongly interact with the ionic field of the ion and three-body interaction leads to the exchange of energy and angular momentum [2]. This process is called post-collision interaction (PCI). PCI has been studied for more than three decades [3] but is still not completely understood. To better describe the effect, we have studied the PCI effect in Auger spectra of Neon double core hole (DCH) 1s−2(n, ε)p using two-photon sequential core ionization and excitation with XFEL radiation, in which two electrons are removed from the core level sequentially with two photons of the same energy within the lifetime of first core hole. To experiment, we used the SQS (Small Quantum Systems) beamline at EuXFEL which gives access to the required energy range

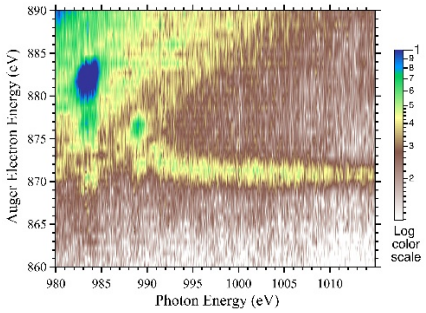


Figure 1: 2D map of Ne KL $_{2,3}$L$_{2,3}$ hyper satellite Auger spectra after the creation of the double-core-hole in Ne. The horizontal axis represents the photon energy (eV) and the vertical axis represents the kinetic energy (eV) of Auger electrons kinetic energy. The color code encodes the intensity of KL $_{2,3}$L$_{2,3}$ Auger spectra.

The process of creation of a double core hole using X-FEL radiation of an isolated Neon atom, followed by KLL hyper-satellite Auger decay, can be written as follows:

$$2\gamma + Ne \rightarrow Ne2+(1s-2)+eph1+eph2(E0+i\Gamma DHC/2) \rightarrow Ne3+(1s-12p-2)+eph1+eph2+eAuger \quad (1)$$

The first incident photon γ ionizes the 1s shell electron of the Neon atom, and before decay of the first vacancy can take place, another photon γ ionizes the second electron from the 1s shell of the same atom and leaves the residual ion Ne(2+∗) in a metastable state. Consequently, electronic relaxation occurs through the emission of a hyper-satellite Auger. The sequential removal of both electrons from the Ne 1s core-shell results in an increase of DCH core lifetime width to 902.5 meV [4], which is around 3.4 times the width of the single core hole of neon. This increased lifetime width enhances the occurrence of post-collision interaction in neon DCH. Furthermore, the observed spectra exhibit strong shake-up and shake-off transitions in the resonance region of the 1s−23p and 1s−24p states. The dispersion of the resonant Auger spectra with photon energy around the 1s−2 ionization is not attributed to the recapturing of photoelectrons in higher Rydberg states. Instead, it is due to the emission of photoelectrons from the 2p state, leading to the same final states 1s−2np(n=3,4,5, . . .) through photoelectron shake-up. This phenomenon contributes to the understanding of the complex processes involved in the Auger decay of neon DCH states.

REFERENCES

[1] M.Y. Kuchiev et al., Sov. Phys. Usp. 32, 569 (1989).
[2] R. Guillemin et al., Phys. Rev. A 99, 063409 (2019).
[3] A. Russek et al., J. Phys. B: At. Mol. Phys. 19, 911 (1986).
[4] G. Goldsztejn et al., Phys. Rev. Lett. 117, 133001 (2016).

Two-color Stimulated Raman Transitions in Atomic Systems from the XUV to the Soft X-ray Regime

U. Eichmann

Max Born Institute, Max-Born Strasse 2A, 12489 Berlin, Germany

ABSTRACT

Using the two-color operation modes available at the FERMI FEL facility and the European XFEL we studied in detail nonlinear stimulated two-color Raman scattering in atoms and molecules in the XUV and soft-x ray regimes on a single atom level. Stimulated Raman scattering is considered a fundamental building block of nonlinear optics, which, particularly in the x-ray regime, is expected to enable studies on ultrafast coherent evolution of electronic wave functions. The experiments are based on the photon recoil imaging technique [1].

Using He as target we experimentally demonstrated two-color stimulated Raman transitions with coherent XUV pulses from the seeded FEL at Fermi through the continuum to the He(2s) metastable singlet state [2]. The two XUV colors of the FEL are in essence two different orders of harmonics derived from a single seed laser. We used pairs of harmonics of orders n and n-4, with $5<n<12$, at a suitable photon energy of the seed laser (~5.14eV) such that the difference of the two harmonics match the excitation energy of He(2s). Upon scanning the seed laser photon energy we observed Raman spectra, which we successfully analyzed using time dependent Schrödinger equation (TDSE) and density matrix calculations. Moreover, due to the coherence of the XUV radiation and the possibility to vary the phase between the two harmonics, we were able to demonstrate interference of two different Raman pathways.

At European XFEL we investigated nonlinear two-color Raman scattering far-off-resonance and in the vicinity of inner shell transitions in Ne 1s-3p (867eV) and in CO, $1s\text{-}2\pi$ (532eV) [3]. By tuning the two independently tunable photon energies to the Raman resonance condition near the inner shell resonance we could dramatically enhance the stimulated Raman process. Intensity dependent scans clearly show the two-photon nature of the process. By tuning the photon energies far-off-resonance but keeping the Raman resonance condition, we observed a pure signal from the stimulated process conserving in principle the coherence in the system. This investigation provides the experimental conditions for achieving stimulated x-ray Raman scattering and clearly shows the feasibility to transfer energy via inner shell transitions.

REFERENCES

1. U. Eichmann et al., Science **369**, 1630 (2020)
2. U. Eichmann, S. Patchkovskii, S. Eisebitt, J.-E. Rubensson, J. Söderström, M Genevriez, J. Mikosch, C. Callegari, M. DiFraia, O Plekan, N. Pal, beamtime FERMI
3. L. Germeroth, M. Agåker, T. Baumann, R. Boll, A. De Fanis, S. Eisebitt, M. Génévriez, V. Kimberg, H. Lee, E. Marin-Bujedo, T. Mazza, M. Meyer, J.Mikosch, Y. Ovcharenko, S. Patchkovskii, D. Reiser, J.-E.Rubensson, J. Söderström, P. Schmidt, B. Senfftleben, A. Senftleben, S. Usenko, and U. Eichmann, beamtime European XFEL

The 2-Color Infrared FEL at the Fritz Haber Institute in Berlin

W. Schöllkopf, W.B. Colson,[1] M. De Pas, D.H. Dowell,[1] S. Gewinner, S.C. Gottschalk,[1] H. Junkes, J.W. Rathke,[1] T.J. Schultheiss,[1] A.M.M. Todd,[1] G. von Helden, L.M. Young,[1] and G. Meijer

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Since 2013 the infrared FEL at the Fritz Haber Institute (FHI FEL) has been providing intense, pulsed mid-infrared (MIR) radiation continuously tunable from <3 μm to >50 μm for in-house user experiments [1]. This has resulted in about 100 peer-reviewed publications in diverse fields ranging from spectroscopy of clusters and (bio-)molecules in the gas-phase to nonlinear solid-state spectroscopy and surface science. Over the past years the FHI FEL has been upgraded to include a second short-Rayleigh-range undulator FEL beamline, as outlined in Fig. 1, that permits lasing in the far-infrared (FIR) regime from <5 μm to >170 μm [2,3]. In addition, a 500 MHz kicker cavity has been installed downstream of the electron accelerator. It operates in a dipole mode using the strong electric field between two vanes to deflect electron bunches up to 50 MeV energy alternatingly left and right by an angle of ±2°. It thus splits the high-repetition-rate (1 GHz) electron bunch train into two bunch trains of half the repetition rate each; one is steered to the MIR FEL and the other one to the FIR FEL. The wavelengths in both FELs can be tuned independently over wide ranges of up to a factor of four by undulator gap variation. In addition, 2-color operation is also available at reduced repetition rates (e.g. 55.5 MHz of both MIR and FIR pulses), as needed for some applications. Furthermore, two additional small dipole magnets upfront and behind the kicker cavity permit conventional single-color operation of either the MIR or the FIR FEL when the 500 MHz kicker field is off. The upgrade was commissioned in 2023. Regular user operation in 2-color mode will start in 2024.

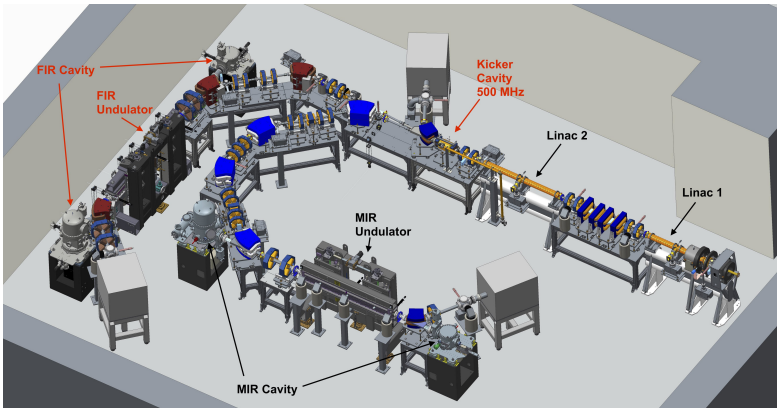


Figure 1: Layout of the FHI FEL 2-color upgrade. A kicker cavity as well as the undulator, optical cavity, and electron beamline of the new FIR branch have been installed and commissioned in 2023.

REFERENCES

1. W. Schöllkopf et al., *The new IR and THz FEL Facility at the Fritz Haber Institute in Berlin*, Proc. of SPIE **9512**, 95121L (2015).
2. W. Schöllkopf et al., *The FHI FEL Upgrade Design*, Proc. of FEL 2019, Hamburg, Germany, August 2019, paper TUP006. doi:10.18429/JACoW-FEL2019-TUP006
3. A.M.M. Todd et al., *Analyses Supporting the Two-color Upgrade to the IR FEL at FHI, Berlin*, Proc. of FEL 2022, Trieste, Italy, August 2022, paper MOP31. doi:10.18429/JACoW-FEL2022-MOP31

[1] Consultants to the Fritz Haber Institute

On the Element-selectivity of Core-level Transient Grating Spectroscopies

O. Cannelli,[1,2] R.A. Ingle,[1,3] G.F. Mancini,[1,4] C. Bacellar,[1,5] R. Geneaux,[6] G. Knopp,[5] L. Foglia,[7] E. Pedersoli,[7] F. Capotondi,[7] F. Bencivenga,[7] R. Mincigrucci,[7] C. Masciovecchio,[7] H.J.B. Marroux,[1,6*] M. Chergui[1,7*]

1. Laboratory of Ultrafast Spectroscopy (LSU) and Lausanne Centre for Ultrafast Science (LACUS), École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland
2. Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron (DESY), Notkestraße 85, 22607 Hamburg, Germany
3. Department of Chemistry, University College London, 20 Gordon Street, London WC1H 0AJ, United Kingdom
4. Laboratory for Ultrafast X-ray and Electron Microscopy (LUXEM), Department of Physics, University of Pavia, I-27100 Pavia, Italy
5. SwissFEL, Paul Scherrer Institute (PSI), Villigen, CH-5232, Switzerland
6. Université Paris-Saclay, CEA, CNRS, LIDYL,91191 Gif-sur-Yvette, France
7. Elettra Sincrotrone Trieste S.C.p.A., Strada Statale 14 - km 163,5, 34149 Basovizza, Trieste, Italy

ABSTRACT

Optical domain transient grating (TG) geometry is one of the most popular four-wave-mixing (FWM) methods due to its numerous applications[1]. Going to the extreme-ultraviolet and X-ray range should enable creating element-selective TG, i.e. generating the dynamics via excitation of a specific atomic centre. Demonstrating this capability would pave the way to multidimensional core-level spectroscopies[2].

Here we report on an extreme ultraviolet (EUV) TG study of spinel Co_3O_4, which hints towards element-selectivity. Co_3O_4 consists of Co^{2+} and Co^{3+} ions in tetrahedral and octahedral sites with a 1:2 stoichiometry. We generated the EUV-TG at different photon energies across the Co $M_{2,3}$-edges, probing it with the first order diffraction of a 400 nm pulse. The traces exhibit a pure electronic response at t=0 and a decay component that varies with the EUV energy, changing from 250 fs up to the threshold of the $M_{2,3}$-edge to >500 fs above the edge of the Co^{3+} centre. These results suggest the presence of excitation wavelength-specific relaxation channels created by the TG excitation.

REFERENCES

1. F. Bencivenga, F. Capotondi, L. Foglia, R. Mincigrucci, and C. Masciovecchio, *Adv. Phys. X*, vol. 8, no. 1, p. 2220363, Dec. 2023, doi: 10.1080/23746149.2023.2220363.
2. M. Chergui, M. Beye, S. Mukamel, C. Svetina, and C. Masciovecchio, *Nat. Rev. Phys.*, vol. 5, no. 10, pp. 578–596, Oct. 2023, doi: 10.1038/s42254-023-00643-7.

ABSTRACTS

Wednesday, June 19th, 2024

Science@FELs 2024

17-21 June 2024 I Paris, France

Wednesday, June 19th

Chair: ***Serguei MOLODTSOV***

PT-03	Illuminating protein structural dynamics by use of static and time-resolved crystallography at XFELs ***Jacques-Philippe Colletier***
IT-11	Control and analysis of coherence in ultrafast X-ray crystallography ***Jasper van Thor***
OC-10	High-throughput X-ray single particle imaging reveals structure variability and polymorphism in viral capsids ***Abhishek Mall***

Chair: ***Laurent NAHON***

IT-12	Coherent diffractive imaging of salt nanoparticles and sea spray aerosols ***Minna Patanen***
OC-11	Activation of nanoplasma in pure and doped He nanodroplets by XUV and soft X-ray light pulses ***Ltaief Ben Ltaief***
IT-13	Dynamics of magnetic structures probed by magnetic helicoidal dichroism ***Thierry Ruchon***

Chair: ***Nicolas JAOUEN***

IT-14	XFELs to unravel ultrafast magnetic dynamics ***Emmanuelle Jal***
OC-12	Ultrafast X-ray and optical studies of charge carrier dynamics in colloidal quantum dots ***Elisa Collet***
IT-15	Ultrafast switching of magnetization at the frequency of optical phonons ***Carl S. Davies***
OC-13	THz-driven coherent magnetization dynamics in a labyrinth domain state ***Matthias Riepp***
OC-14	Strong magnetic fields from plasmonic ring currents in graphene disks ***Martin Mittendorff***

Science@FELs 2024

17-21 June 2024 I Paris, France

Wednesday, June 19th

Chair: ***Minna PATANEN***

IT-16	Light elements at Mbar to Gbar pressure ***Dominik Kraus***
OC-15	Time-domain EUV diffuse scattering phonon spectroscopy ***Flavio Capotondi***
OC-16	Modeling software SURFwiX to guide high-precision processing of materials of industrial relevance ***Vladimir Lipp***
IT-17	X-ray illumination of phase transitions through shock compression and release ***June Wicks***
OC-17	XFEL-based pump-probe XAS and XES characterization of photoexcited states in CeO_2 ***Samuele Pelatti***
OC-18	Pioneering plasma accelerators and novel sample delivery methods for future FEL experiments ***Zeinab Ebrahimpour***

Illuminating Protein Structural Dynamics by use of Static and Time-resolved Crystallography at XFELs

J-P. Colletier

Institut de Biologie Structurale, UMR 5075, CNRS-CEA-UGA, Grenoble

ABSTRACT

XFELs deliver highly-intense, highly-focused, ultra-short (5-40 fs) X-ray pulses that can be used to collect radiation-damage-free diffraction data from sub-micron sized macromolecular crystals[1,2]. The shortness of XFEL pulses also enables time-resolved (TR) structural studies on the fs-ms[3] timescale. The intense X-ray beam eventually destroys the crystals, necessitating the constant replenishment of sample such that data is collected from thousands to millions of crystals, in an approach termed Serial Femtosecond Crystallography (SFX)[2]. We will review the differences between conventional macromolecular crystallography and (TR)-SFX, before illustrating the leap forward enabled by the latter using examples from the literature.

REFERENCES

[1] Chapman *et al.* Nature (2011) 470, 73–77.
[2] Barends *et al.* Nat. Rev. Methods Primer (2022) 2, 1–24.
[3] Brändén & Neutze. Science (2021) 373, eaba0954.

Control and Analysis of Coherence in Ultrafast X-ray Crystallography

J.J. van Thor

Imperial College London, UK

ABSTRACT

We have demonstrated a coherent control experiment of vibrational coherence with ultrafast X-ray crystallographic observation [1]. The photoisomerisation reaction of a fluorescent protein chromophore includes ultrafast barrier crossings and the avoidance of internal conversion processes. The relationship between experimentally observed ultrafast nuclear motions and the biological reaction coordinate is not known. High resolution pump-probe X-ray crystallography measurements reveal complex sub-Ångström ultrafast motional dynamics and hydrogen bonding rearrangements in the active site. However, we demonstrate that the measured motions are not part of the reaction coordinates but instead arise from impulsively driven coherent vibrational processes of the electronic ground state. We apply the analysis of impulsive stimulated Raman spectroscopy and coherent control methodology to the real-space observation of low frequency vibrational dynamics and displacements, that we have measured by ultrafast X-ray crystallography [1,2]. Using the 'Tannor-Rice' coherent control method it is shown that ultrafast motions can be strongly amplified. This demonstrates that the conventional pump-probe measurements, without application of optical control, are dominated by electronic ground state displacements that are unrelated to the reactive photoisomerisation coordinate. We present the first application of coherent control using X-ray crystallography in combination with coherence theory that has application to the real space wavepacket observation [2].

REFERNCES

[1]. Christopher D.M. Hutchison, et al. Optical control of ultrafast structural dynamics in a fluorescent protein. (2023) ***Nature Chemistry.*** **15**, 1607–1615

[2] Samuel Perrett et al. Application of Density Matrix Wigner Transforms for Ultrafast Macromolecular and Chemical X-ray Crystallography. (2024) ***J. Chem. Phys.*** *In Press*

High-throughput X-ray Single Particle Imaging Reveals Structure Variability and Polymorphism in Viral Capsids

A. Mall[1], A. Munke[2,3], S. Zhou[1], P. Mazumder[1] and K.Ayyer[1,4]

[1] Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany
[2] Center for Free Electron Laser Science, Deutsches Elektronen Synchrotron (DESY), 22607 Hamburg, Germany
[3] Laboratory of Molecular Biophysics, Department of Cell and Molecular Biology, Uppsala University, Uppsala, SE-75124, Sweden
[4] The Hamburg Center for Ultrafast Imaging, Universität Hamburg, 22761 Hamburg, Germany

ABSTRACT

Single-stranded RNA viruses co-assemble their capsid with the genome, and the variability in capsid structure affects the ordered packing of the genome, influencing their interactions with the host [1,2]. Here, aerosolized MS2 bacteriophage particles were exposed to ultrashort X-ray free electron laser (XFEL) pulses one at a time to produce a large number of coherent diffraction patterns. Each pattern resulted from a particle in not only a random orientation but also a different level of hydration. Using a combination of Bayesian machine learning and deep learning tools [3,4], this dataset was classified to yield a 3D structure variation of the capsids as a function of time spent in the aerosol phase. The analysis also uncovers a capsids structure with an octahedral shape — a known form of polymorphism in MS2 phages [5]. From around 300,000 diffraction patterns, we observe continuous changes in the virus morphology, associated with hydration dependent capsid protein interaction. In the process, we demonstrate how such an aerosol-phase structure probe with an XFEL can be invaluable in studying the structural dynamics in ensembles of nanosystems.

REFERENCES

1. Klug, Aaron. "The tobacco mosaic virus particle: structure and assembly." *Philosophical Transactions of the Royal Society of London. Series B: Biological Sciences* 354.1383 (1999): 531-535.
2. Sun, Siyang, Venigalla B. Rao, and Michael G. Rossmann. "Genome packaging in viruses." *Current opinion in structural biology* 20.1 (2010): 114-120.
3. Ayyer, Kartik, et al. "Dragonfly: an implementation of the expand–maximize–compress algorithm for single-particle imaging." *Journal of applied crystallography* 49.4 (2016): 1320-1335..
4. Zhuang, Yulong, et al. "Unsupervised learning approaches to characterizing heterogeneous samples using X-ray single-particle imaging." *IUCrJ* 9.2 (2022): 204-214.
5. de Martín Garrido, Natàlia, et al. "Bacteriophage MS2 displays unreported capsid variability assembling T= 4 and mixed capsids." *Molecular Microbiology* 113.1 (2020): 143-152.

Coherent Diffractive Imaging of Salt Nanoparticles and Sea Spray Aerosols

M. Patanen

Nano and molecular systems research unit, Faculty of Science, P.O. Box 3000, 90014 University of Oulu, Finland

ABSTRACT

Sea spray aerosols play a key role in the climate system, but their impact on global climate remains highly uncertain. Key knowledge gaps related to sea spray aerosol particles are e.g. size-dependent chemical composition and differences in surface vs. bulk composition. Several studies have reported that certain species are enriched in sea spray aerosol particles compared to bulk seawater (see e.g. [1] and references therein). Previously, we have used synchrotron radiation excited X-ray photoelectron spectroscopy (XPS) to study surface enrichment of *in situ* generated model salt particles and sea salt aerosols [2,3,4]. While XPS can give extremely valuable information on the surface chemistry, the signal is averaged over a large particle ensemble from which we additionally know only their aerodynamic diameter distribution, without any morphological insights. Thus, single particle Coherent Diffraction Imaging (CDI) experiments which give information on the morphology of individual particles were carried out at SQS instrument at European X-ray Free Electron Laser (EuXFEL). The high brightness of FEL pulses allows to get images of isolated particles while they are freeflying in vacuum, avoiding any interaction with the surroundings (e.g. deposition on a substrate) that may affect their structure and behavior. We have analysed simple salt systems as well as Arctic sea water sampled during the ARTofMELT 2023 expedition (www.su.se/artofmelt). We compared different ways to generate the aerosols and the effect of relative humidity (RH) on their morphology. We found that the particle morphology depends strongly on their composition and the RH of the particle stream before the particles enter the vacuum system. Machine learning (ML) algorithms are under development to analyse the large number of CDI data this kind of experiment produces. CDI experiments and ML guided data analysis are promising tools to obtain even 3D information on *in situ* produced particles, and to follow e.g. their sizedependent properties. Techniques used here are applicable to different types of aerosol particles (organic, inorganic, metals, etc.) and aerosol generation methods (flames, air-blast nebulisers, bubblers, plasma generation, etc.)

Acknowledgement: I want to thank SQS team at EuXFEL, especially Y. Ovrachenko for their help during the experiments, and my collaborators in the project: J. Asplund, K. Ayyer, M. Babayan, O. Björneholm, A. Colombo, R. Dagar, M. Huttula, S. Ingram, F. Maia, J. Malila, A. Mall, P. Mazumder, B. Reischl, D. Rupp, M. Sauppe, A. Sehati, J. Sellberg, Z. Shen, P. Svensson, O. Veteläinen, N. Walsh, T. You, and P. Zieger. I also want to thank A. Summers and A. Rudenko for letting us use their aerodynamic lens setup.

REFERENCES

1. M. E. Salter, E. Hamacher-Barth, C. Leck, J. Werner, C. M. Johnson, I. Riipinen, E. D. Nilsson, and P. Zieger, Geophys. Res. Lett. **43**, 8277–8285 (2016).
2. M. Patanen, I. Unger, C.-M. Saak, G. Gopakumar, R. Lexelius, O. Björneholm, M. Salter, and P. Zieger, Env. Sci. Atmosph. **2**, 1032 - 1040 (2022).
3. E. Pelimanni, C.-M. Saak, G. Michailoudi, N. Prisle, M. Huttula, and M. Patanen, Phys. Chem. Chem. Phys. **24**, 2934 - 2943 (2022).
4. I. Unger, C.-M. Saak, M. Salter, P. Zieger, M. Patanen, and O. Björneholm, J. Phys. Chem. A **124**, 422-429 (2020).

Activation of Nanoplasma in Pure and Doped He Nanodroplets by XUV and Soft X-ray Light Pulses

L. Ben Ltaief[1], C. Medina[2], D. Schomas[2], A. Ø. Lægdsmand[1], Z. Hoque[3], A. H. Roos[3], E. K. Klimesová[3], M. Krikunova[3], J. Andreasson[3], C. Ott[4], R. Moshammer[4], S. R. Krishnan[5], A. Heidenreich[6] and M. Mudrich[1]

[1] *Department of Physics and Astronomy, Aarhus University, 8000 Aarhus C, Denmark*
[2] *Institute of Physics, University of Freiburg, 79104 Freiburg, Germany*
[3] *ELI Beamlines Centre, ELI ERIC, Za Radnicí 835, 252 41 Dolní Břežany, Czech Republic*
[4] *Max-Plank-Institut für Kernphysik, 69117 Heidelberg, Germany*
[5] *Indian Institute of Technology Madras, Chennai 600036, India*
[6] *IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain*

ABSTRACT

Nanoplasma induced from nanometer-sized clusters and nanodroplets in interaction with intense light are intriguing transient states of matter featuring extraordinary properties. They are capable to efficiently absorb intense light and to convert it into energetic highly charged ions, fast electrons and radiation covering the entire electromagnetic spectrum up to x-rays [1]. Here, we report on the dynamics of He nanoplasma formed in pure and heavier rare gas atoms doped He nanodroplets by employing pump-probe electron/ions spectroscopy.

For doped He nanodroplets, the dynamics of He nanoplasma is studied at the XUV-FEL facility FLASH at DESY in Hamburg. First, a soft x-ray pump pulse selectively inner-shell ionizes the core cluster formed of heavier rare-gas atoms in the He nanodroplet, causing electron migration from the He shell to the highly charged core. This ignites a He nanoplasma which is then driven by an intense near-infrared probe pulse. The ultrafast charge redistribution, evidenced by the rise of He^+ and He^{2+} ion yields from the nanoplasma within $\lesssim 70$ fs, leads to strong damping of the core cluster expansion [2]. Thus, He droplets act as efficient tampers that reduce the radiation damage of embedded nanostructures, a property that could be exploited for improving coherent diffraction images [3].

For pure He nanodroplets, the dynamics of He nanoplasma is studied at ELI beamlines facility in Dolni Brezaney near Prague. Here, we mainly studied the dynamics of avalanche ionization of pure helium nanodroplets activated by a weak extreme-ultraviolet (XUV) pulse generated from high harmonics (HH 14_{th} or HH 19_{th}) and driven by an intense near-infrared (NIR) pulse. In addition to a transient enhancement of ignition of a nanoplasma caused by the injection of seed electrons into the droplets by XUV photoemission at short time delays ~200 fs, long-term activation of the nanodroplets lasting up to a few nanoseconds is observed as due to electrons remaining loosely bound to photoions which form stable 'snowball' structures in the droplets. This long-term activation of He nanoplasma indicates that XUV irradiation can induce long-lasting changes of the strong-field optical properties of nanoparticles, potentially opening new routes to controlling avalanche-ionization phenomena in nanostructures and condensed-phase systems [4].

REFERENCES

1. T. Fennel, K-H. Meiwes-Broer, J. Tiggesbäumker *et al.*, *Rev. Mod. Phys.* **82** 1793–842 (2010)
2. D. Schomas, C. Medina, L. Ben Ltaief *et al.*, 2020 (*arXiv:*2005.02944 [physics])
3. B. Ziaja, H. Chapman, R. Santra *et al.*, *Phys. Rev. A* **84**, 033201 (2011); C. Gnodtke *et al.*, *Phys. Rev. A* **79**, 041201 (2009)
4. C. Medina, A.Ø. Lægdsmand, L. Ben Ltaief *et al.*, *New J. Phys.* **25**, 053030 (2023)

Dynamics of Magnetic Structures Probed by Magnetic Helicoidal Dichroism

M. Fanciulli[1,2,3], M. Pancaldi,[4, 5], A-E. Stanciu,[6] M. Guer,[2] E. Pedersoli,[4] D. De Angelis,[4] P.R. Ribic,[4] D. Bresteau,[2] M. Luttmann,[2] P. Carrara,[7] B. Rösner,[8] C. David,[8] C. Spezzani,[4] M. Manfredda,[4] R. Sousa,[6] L. Vila,[6] I.L. Prejbeanu,[6] L.D. Buda-Prejbeanu,[6] B. Dieny,[6] G. De Ninno,[4, 9] F. Capotondi,[4] T. Ruchon,[2] and M. Sacchi[7, 10]

[1] CY Cergy Paris Université, CEA, LIDYL, 91191 Gif-sur-Yvette, France; [2] Université Paris-Saclay, CEA, LIDYL, 91191 Gif-sur-Yvette, France; [3] New Technologies Research Center, University of West Bohemia, 30100 Pilzen, Czech Republic; [4] Elettra-Sincrotrone Trieste S.C.p.A., 34149 Basovizza, Trieste, Italy; [5] Department of Molecular Sciences and Nanosystems, Ca' Foscari University of Venice, 30172 Venezia, Italy; [6] Universit´e Grenoble Alpes, CNRS, CEA, Grenoble INP, IRIG-SPINTEC, 38000 Grenoble, France; [7] Sorbonne Universit´e, CNRS, Institut des NanoSciences de Paris, INSP, 75005 Paris, France; [8] Paul Scherrer Institut, 5232 Villigen-PSI, Switzerland; [9] Laboratory of Quantum Optics, University of Nova Gorica, 5001 Nova Gorica, Slovenia; [10] Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin, B. P. 48, 91192 Gif-sur-Yvette, France

ABSTRACT

The interaction of polarized light beams with magnetic materials defines the rich set of analytical tools used in magneto-optics and is widely employed for studying magnetization dynamics. Light polarization is associated with a spin angular momentum, whose projection on the propagation axis, takes values $\sigma=\pm\hbar$ for circular polarization, imparting a well-defined handedness to the photon beam. It is less common to exploit the orbital angular momentum (OAM) carried by light beams characterized by an helicoidal wavefront. The quantified projection of the OAM ($\ell\hbar$) can take any integer multiple of $\hbar$, with a sign determined by the spiraling sense of the phase. After finding many applications in the visible range, OAM beams with ultra-short pulse duration and XUV wavelengths became available recently at high-harmonic generation (HHG) and free-electron laser (FEL) sources [1,2], widening considerably their application range.

We explored theoretically and experimentally the interaction of OAM light beams with magnetic structures featuring non-uniform magnetization, in particular of XUV beams with magnetic vortices. We predicted and observed, at the DiProI station of the FERMI free-electron laser source, that the far field intensity profile encodes the magnetic vortex symmetry in a way that depends on the sign and value of ℓ, introducing a Magnetic Helicoidal Dichroism (MHD) [3]. It further allowed to follow the evolution of the magnetization topology at the sub-ps timescale after an optical excitation. The obtained experimental results [4] match very well our theoretical predictions [3], confirming the potential of the new toolset provided by MHD for studying complex magnetic structures dynamics.

REFERENCES

[1] R. Géneaux, et al., Nat. Comm., 7, 12583 (2016).
[2] P. R. Ribič, et al, Phys. Rev. X 7, 031036 (2017).
[3] M. Fanciulli et al., Phys. Rev. A, 103, 013501 (2021)
[4] M. Fanciulli et al.,accepted in Phys. Rev. Lett., 128, 077401 (2022)

X-FELs to Unravel Ultrafast Magnetic Dynamics

E. Jal

1 Sorbonne Université, CNRS, Laboratoire de Chimie Physique-Matière et Rayonnement, LCPMR, 75005 Paris, France

ABSTRACT

The discovery of laser induced ultrafast magnetization dynamics by Beaurepaire et al. [1] has stimulated an intense research activity and opened up the field of femtomagnetism in condensed matter physics. Even after thirty years of studies and debates, the mechanisms of ultrafast demagnetization remain disputed. In order to bring new experimental informations into this field, and with the advent of femtosecond X-rays sources, we are using time resolved extreme ultraviolet (XUV) and soft X-rays based techniques on ultrathin ferromagnetic thin films. During my talk I will give an overview of what can be done with XUV and soft x-ray short pulses provided by X-FELs to probe electronic, magnetic and structural dynamics of thin ferromagnet. A special emphasis will be given on (i) simultaneous electronic and magnetism dynamics [2,3,4] (ii) magnetic domain wall dynamics [5,6,7] and (iii) simultaneous structural and magnetism dynamics [8,9].

REFERENCES

[1] Beaurepaire et al. Phys. Rev. Lett. 76, 4250 (1996) https://doi.org/10.1103/PhysRevLett.76.4250
[2] Rösner et al. Struct. Dyn. 7, 054302 (2020) https://doi.org/10.1063/4.0000033
[3] Yao et al. Phys. Rev. B 102, 100405(R) (2020) https://doi.org/10.1103/PhysRevB.102.100405
[4] Hennes et al. Appl. Sci. 11m 325 (2021) https://doi.org/10.3390/app11010325
[5] Hennes et al. Phys. Rev. B 102, 174437 (2020) https://doi.org/10.1103/PhysRevB.102.174437
[6] Zhou Hagström et al. Phys. Rev. B 106, 224424 (2023) https://doi.org/10.1103/PhysRevB.106.224424
[7] Suturin et al. Phys. Rev. B 108 174444 (2023) https://doi.org/10.1103/PhysRevB.108.174444
[8] Jal et al. Phys. Rev. B 95, 184422 (2017) https://doi.org/10.1103/PhysRevB.95.184422
[9] Chardonnet et al. Struct. Dyn. 8, 034305 (2021) https://doi.org/10.1063/4.0000109

Ultrafast X-ray and Optical Studies of Charge Carrier Dynamics in Colloidal Quantum Dots

W. Gawelda[1,2], A. Burgos[2], J. Hidalgo[2], E. Collet[1], M.E. Corrales[1], B.R. de Carvalho Vale[3], A.F. Vale da Fonseca[3], L. Padilha[3], I.M. Casasús[4], K. Shaw[2], David Sorribes[1,4], L. Bañares[4], T-K. Choi[5], T. Katayama[6], M.A. Schiavon[7], M. Biednov[8], F. Ardana[8], F.A. Lima[8], C. Milne[8], S. Nozawa[9], S-I. Adachi[9]

[1]Universidad Autónoma de Madrid, 28049 Madrid, Spain
[2]IMDEA Nanoscience, 28049 Madrid, Spain
[3]Universidade Estadual de Campinas, 13083-859 Campinas-SP, Brazil
[4]Universidad Complutense de Madrid, 28040 Madrid, Spain
[5]PAL XFEL Pohang Accelerator Laboratory, Pohang, Gyeongbuk 37673, South Korea
[6]SACLA XFEL, Japan Synchrotron Radiation Research Institute (JASRI), Hyogo 679-5198, Japan
[7]Universidade Federal de São João Del Rei, 36307-352 Minas Gerais, Brazil
[8]European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany
[9]High Energy Accelerator Research Organization, Tsukuba, Ibaraki 305-0801, Japan

ABSTRACT

The interest in $CuInS_2$ (CIS) quantum dots (QDs) has increased significantly in the past few years due to broad range of their potential applications, such as photodynamic therapy or solar cells[1]. Recently, CIS QDs have been shown to perform efficiently as a light harvester in combination with a molecular catalyst in hydrogen production[2]. This owes to extraordinary optoelectronic properties of this material, such as broad photoluminescence with a large Stokes shift and long charge carrier lifetimes. To elucidate the origin of these unique features, several mechanisms for the radiative recombination in CIS QDs have been proposed theoretically and experimentally.

In this work we aim to understand and confirm the possibility of observing the radiative recombination resulting from an electron in the conduction band and a hole in the so-called confined hole state (CHS)[3-4], next to the effects of stoichiometry and Zn doping on the passivation of the QDs and the formation process of the CHS[5].

We approached these questions through a combination of ultrafast laser and XFEL pump-probe techniques complemented with steady-state synchrotron measurements. The element and oxidation state specificity of X-ray techniques will serve as a direct probe to track recombination and localization dynamics of photogenerated holes, while optical probes will follow the electron dynamics. We probed the oxidation state of Cu via fs-resolved XANES (SACLA XFEL) and non-resonant XES (European XFEL) and comparing with reference data obtained from static XANES and EXAFS at Cu, Zn and S K-edges (ALBA). This allowed us to gain insights and correlate the observed charge carrier dynamics with the underlying CIS QD structures, the degree of their surface passivation and the different Zn doping levels. This is then complemented by our time-resolved optical studies monitoring transient absorption and photoluminescence.

REFERENCES

[1] J. Kolny-Olesiak, H. Weller; *ACS Appl. Mater. Interfaces* **5**, 12221 (2013)
[2] A. J. Bagnall, *et al.*, *ACS Catal.* **14**, 4186−4201 (2024)
[3] W. van der Stam, *et al.*; *ACS Nano* **12**, 11244 (2018)
[4] A. Fuhr, et al.; *ACS Nano* **14**, 2212 (2020)
[5] B. R. C. Vale, *et al.*; *J. Phys. Chem. C* 1**24**, 27858 (2020)

Ultrafast Switching of Magnetization at the Frequency of Optical Phonons

C.S. Davies

HFML-FELIX, Radboud University, Toernooiveld 7, 6525 ED Nijmegen, The Netherlands

INTRODUCTION

The demonstration of magnetic switching on ultrafast timescales and with minimal dissipations is mandatory for the future development of data-storage technologies. All-optical switching (AOS) of magnetization – whereby ultrashort femtosecond-long optical pulses switch magnetization without the need for any bias magnetic fields – offers a promising route for magnetic data-recording in terms of its speed, efficiency and versatility [1]. Here, we explore how tailored light-wave excitations matching the frequency of optical phonons can switch magnetization. Specifically, using narrow-band infrared (IR) optical pulses delivered by the free-electron laser facility FELIX with frequency ranging between 6 THz and 30 THz (wavelength 10-50 μm) [2], we target the characteristic frequency of longitudinal or transverse optical phonons in different systems. We probe the resulting magnetization dynamics and switching using static and time-resolved magneto-optical microscopy.

RESULTS

We find that an ultrafast IR excitation at the frequency of longitudinal optical phonon modes in magnetic iron-garnet films switches magnetization with a peculiar quadrupolar domain pattern. This unusual fingerprint of switching unambiguously reveals its rooting in the magneto-elastic mechanism [3]. In contrast, the excitation of strongly absorbing transverse phonon modes results only in thermal demagnetization. The demonstrated displacive mechanism of switching is also found to operate in other materials such as weak ferromagnets, antiferromagnets [4] and even ferroelectrics, which have very different crystallographic and symmetry properties. We argue that this switching is empowered by the strong light-matter interaction characteristic of the epsilon-near-zero regime [5].

We have also discovered that optical phonons in paramagnetic glass and sapphire substrates can, when driven at resonance by circularly-polarized pulses, can generate a transient magnetization that switches magnetization. In these experiments, we sweep circularly-polarized IR pulses across thin GdFeCo films mounted on SiO_2 or Al_2O_3. The resulting helicity-dependent magnetic switching scales in efficiency with the phonon spectrum characteristic not of the metal but rather the substrate. Moreover, we prove that this is distinct from exchange-driven switching by stretching the width of the optical pulses, and observing that the helicity-dependent switching persists while the helicity-independent mechanism fails. Thus, we demonstrate that the ultrafast Barnett effect (excited within the substrate) can remotely drive magnetic switching in the mounted overlayer [6].

REFERENCES

1. A. Kirilyuk, A.V. Kimel and T. Rasing, *Rev. Mod. Phys.* **82**, 2731 (2010).
2. G. M. H. Knippels et al, *Phys. Rev. Lett.* **83**, 1578 (1999).
3. A. Stupakiewicz et al, *Nature Physics* 17, 489 (2021).
4. P. Stremoukhov et al, *New J. Physics* 24, 023009 (2022).
5. M. Kwaaitaal et al, *Nature Photonics* (in press, 2024).
6. C. S. Davies et al, *Nature* (in press, 2024).

THz-driven Coherent Magnetization Dynamics in a Labyrinth Domain State

M. Riepp[1,2], A. Philippi-Kobs[2], L. Müller[2], W. Roseker[2], R. Rysov[2], R. Frömter[3], K. Bagschik[2], M. Hennes[4], D. Gupta[1], S. Marotzke[2,5], M. Walther[2], S. Bajt[6,7], R. Pan[2], T. Golz[2], N. Stojanovic[8], C. Boeglin[1], and G. Grübel[2,9]

[1] *Institut de Physique et Chimie des Matériaux de Strasbourg, UMR7504, CNRS, Université de Strasbourg, Strasbourg, 67000, France.*
[2] *Deutsches Elektronen-Synchrotron DESY, Hamburg, 22607, Germany.*
[3] *Institute of Physics, Johannes Gutenberg-University Mainz, Mainz, 55099, Germany.*
[4] *Institut des NanoSciences de Paris, UMR7588, CNRS, Sorbonne Université, Paris, 75005, France.*
[5] *Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Kiel, 24098, Germany.*
[6] *Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg, 22607, Germany.*
[7] *The Hamburg Centre for Ultrafast Imaging, Hamburg, 22761, Germany.*
[8] *Institute for Optical Sensor Systems, Deutsches Zentrum für Luft- und Raumfahrt, Berlin, 12489, Germany.*
[9] *Present address: European X-Ray Free-Electron Laser Facility GmbH, Schenefeld, 22869, Germany.*

ABSTRACT

Terahertz (THz) light pulses can be used for an ultrafast coherent manipulation of the magnetization [1,2]. Driving the magnetization at THz frequencies is currently the fastest way of writing magnetic information in ferromagnets [3]. Using time-resolved resonant magnetic scattering at the free-electron laser FLASH beamline BL3 [4], we gain new insights to the THz-driven coherent magnetization dynamics on nanometer length scales. We observe ultrafast demagnetization and coherent magnetization oscillations that are governed by a time-dependent damping. This damping is determined by the interplay of lattice heating and magnetic anisotropy reduction revealing an upper speed limit for THz-induced magnetization switching. We show that in the presence of nanometer-sized magnetic domains, the ultrafast magnetization oscillations are associated with a correlated beating of the domain walls. The overall domain structure thereby remains largely unaffected which highlights the applicability of THz-induced switching on the nanoscale.

REFERENCES

[1] Kampfrath, T., Sell, A., Klatt, G., Pashkin, A., Mährlein, S., Dekorsy, T., Wolf, M., Fiebig, M., Leitenstorfer, A., Huber, R.: *Coherent terahertz control of antiferromagnetic spin waves.* Nature Photonics **5**(1), 31–34 (2011).
[2] Bonetti, S., Hoffmann, M.C., Sher, M.-J., Chen, Z., Yang, S.-H., Samant, M.G., Parkin, S.S.P., Dürr, H.A.: *THz-Driven Ultrafast Spin-Lattice Scattering in Amorphous Metallic Ferromagnets.* Physical Review Letters **117**(8), 087205 (2016).
[3] Tudosa, I., Stamm, C., Kashuba, A.B., King, F., Siegmann, H.C., Stöhr, J., Ju, G., Lu, B., Weller, D.: *The ultimate speed of magnetic switching in granular recording media*. Nature **428**(6985), 831–833 (2004).
[4] Pan, R., Zapolnova, E., Golz, T., Krmpot, A.J., Rabasovic, M.D., Petrovic, J., Asgekar, V., Faatz, B., Tavella, F., Perucchi, A., et al.: *Photon diagnostics at the FLASH THz beamline*. Journal of Synchrotron Radiation **26**(3), 700–707 (2019).

Strong Magnetic Fields from Plasmonic Ring Currents in Graphene Disks

J.W. Han[1], P. Sai[2], S. Winnerl[3], T.E. Murphy[4], and M. Mittendorff[1]

[1] *Universität Duisburg-Essen, Fakultät für Physik, Lotharstr. 1, 47057 Duisburg, Germany*
[2] *CENTERA Laboratories, Institute of High Pressure Physics PAS, 01-142 Warsaw, Poland*
[3] *Helmholtz-Zentrum Dresden-Rossendorf, Dresden 01328, Germany*
[4] *University of Maryland, College Park MD 20740 MD, USA*

ABSTRACT

Graphene plasmonic elements can strongly enhance the light matter interaction and therewith also the nonlinear optical properties. This nonlinearity is dominated by thermal effects, though also strong electric fields can induce a nonlinear effect [1]. Furthermore, the plasmonic motion can hybridize with the cyclotron motion in magnetic fields [2], allowing efficient tuning of the nonlinear effects [3]. Here we show that illuminating graphene disks with circularly polarized radiation induces circulating plasmonic currents that induce strong out-of plane quasi-static magnetic fields [4].

Graphene disks with a diameter of 1.2 µm and a plasmon frequency of 3.5 THz where illuminated resonantly with circularly polarized THz pulses from the free-electron laser FELBE at Helmholtz-Zentrum Dresden-Rossendorf. To probe the magnetic field in the vicinity of the disks, the Faraday rotation was probed with a linearly polarized pulse. The pump-induced Faraday rotation as a function of the time delay is shown in Fig. 1 (a). As a calibration experiment, we measured the Faraday rotation in static magnetic fields, the corresponding field strength is shown on the right side of Fig. 1 (a). To analyze the data and to characterize the spatial distribution of the magnetic field in the vicinity of the disks, we performed finite-element simulations as shown in Fig. 1 (b).

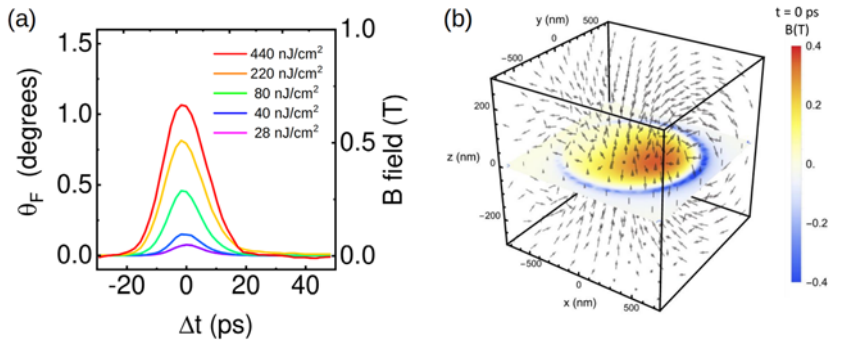


Figure 1: (a) Pump-induced Faraday rotation as a function of the time delay between pump and probe pulse. (b) Magnetic field distribution around the graphene disk, the field strength at the position of the graphene disk is color coded (taken from [4]).

REFERENCES

1. J. W. Han et al., Adv. Photonics Res. **3**, 2100218 (2022).
2. H. Yan et al., Nano Lett. **12**, 3766 (2012).
3. M. L. Chin et al., J. Phys. Photonics **3**, 01LT01 (2021).
4 J. W. Han et al., Nat. Commun. **14**, 7493 (2023).

Light Elements at Mbar to Gbar Pressures

D. Kraus[1,2]

University of Rostock, 18051 Rostock, Germany
Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany

ABSTRACT

I will discuss recent experiments at high energy laser systems to mimic planetary and stellar interiors in the lab. Via nanosecond dynamic compression in combination with XFEL probing, we can access exotic chemistry and the formation of unusual structures. With shorter pulses giving higher intensities and using spectrally resolved X-ray scattering, we can create and probe matter at conditions similar to the envelopes of white dwarfs.

Time-domain EUV Diffuse Scattering Phonon Spectroscopy

F. Capotondi[1], A. Maznev[2], F. Bencivenga[1], D. Fausti[1], L. Foglia[1], C. Gutt[3], N. Jaouen[4], D. Ksenzov[3], C. Masciovecchio[1], R. Mincigrucci[1], K.A. Nelson[2], I. Nikolov[1], M. Pancaldi[1], E. Pedersoli[1], B. Pfau[5], L. Raimondi[1], R. Totani[1], M. Trigo[6]

1 Elettra Sincrotrone Trieste, Strada Statale 14, km 163.5, 34149 Basovizza, TS, Italy;
2Massachusetts Institute of Technology Cambridge, Massachusetts 02139, USA
3 Universität Siegen, Walter-Flex-Strasse 3, 57072 Siegen, Germany.
4 Synchrotron SOLEIL, Saint-Aubin, Boite Postale 48, 91192, Gif-sur-Yvette Cedex, France
5 Max Born Institute, Max-Born-Straße 2A, 12489 Berlin, Germany
6 SLAC National Accelerator Laboratory, Menlo Park, CA, USA

ABSTRACT

Thin film technology assumes a pivotal role in precisely defining electronic, mechanical, or magnetic characteristics through the functionalization of material properties. As the significance and complexity of advanced coatings and films grow, the characterization of their physical properties becomes increasingly crucial. A challenge in providing a comprehensive account of acoustic response at nanometer length scale stemmed from the limited number of experimental techniques capable of addressing this specific range. Indeed at large spatial frequency excitation (i.e, wave vector $Q \leq 0.1$ nm^{-1}), photoacoustic technique based on visible laser pulses can be used to stimulate coherent thermoelastic perturbation that can be probed either by Brillouin scattering or by acoustic echoes [1]. At the opposite extreme, inelastic hard X-ray scattering and thermal neutron scattering can provide information for collective lattice vibration sensitive to interatomic distances, i.e. with wave vectors $Q > 1$ nm^{-1} [2,3] Only recently, the emergence of free electron lasers and the subsequent advancement of extreme ultraviolet (EUV) and hard X-ray transient grating (TG) spectroscopy have enabled the exploration of the intermediate mesoscopic regime [4,5]. Here we present findings on an alternative approach based on the sensitivity of EUV diffuse scattering in reflection geometry for tracking the temporal evolution of surface phonon modes. Our observations reveal that across a diverse range of samples, including single metallic films, metallic multilayer structures, and bulk semiconductors, the optical stimulus induces a discernible perturbation in the diffuse scattering intensity at specific exchanged momentum. This approach allows for the direct measurement of Rayleigh modes dispersion across a broad range of wave-numbers, limited only by the spatial dimension of the pixeled detector (ranging from 0.02 to 0.12 nm^{-1}). This coverage spans the spatial dimensionality typical of optical probe EUV transient grating experiments (~300 nm) [4] down to EUV probed experiments (~50 nm) [5].

REFERENCES

1. Pascal Ruello and Vitalyi E. Gusev, Ultrasonics 56 (2015) 21–35
2. Masciovecchio C, et al. Nucl Instrum Methods Phys Res Sect B Beam Interact Mater At. 1996;111:181.
3. Marshall W, Lovesey SW, Nelkin M. Am J Phys. 1972;40:1715.
4. F. Bencivenga, et al. Nature, 520 (2015), pp. 205-208, 10.1038/nature14341
5. F. Bencivenga, et al. Sci. Adv., 5 (2019), p. eaaw5805, 10.1126/sciadv.aaw5805

Modeling Software SURFwiX to Guide High-precision Processing of Materials of Industrial Relevance

V. Lipp [1,2], I. Inoue [3], S. Egawa [4], G. Yamaguchi [3], D. Breus [5,6], H. Noei [6], S. Toleikis [6], B. Ziaja [1,2]

[1] *Center for Free-Electron-Laser Science CFEL, Deutsches Elektronen Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany*
[2] *Institute of Nuclear Physics, Polish Academy of Sciences, Radzikowskiego 152, 31-342 Kraków, Poland*
[3] *RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo, Hyogo 679-5148, Japan*
[4] *Research Center for Advanced Science and Technology, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8904 Japan*
[5] *Taras Shevchenko National University of Kyiv, 64/13, Volodymyrska Street, City of Kyiv, 01601 Ukraine*
[6] *Deutsches Elektronen Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany*

ABSTRACT

We report on our computational tool called SURFwiX [1], able to predict the response of technologically relevant materials (here, silicon and diamond) to a focused XUV/soft X-ray pulse [1]. The tool could guide high-precision material processing in industrial applications, such as 2D & 3D patterning and sculpturing [2-3] through direct writing with an X-ray laser. The mechanism explored for material processing in this application is ultrafast material ablation (in semiconductors preceded by sub-picosecond non-thermal melting [4]), triggered by a tightly focused XUV/soft X-ray pulse. Its effect on the material, i.e., the dimensions and shapes of the created craters, can be controlled through tuning X-ray beam parameters. Some of those control options were explored in the preliminary experimental study [5] for micrometer focusing. Our recent experimental study performed at SACLA [6] enables us to check the validity of the SURFwiX predictions in a broad XUV-ray parameter range. It is expected that by scaling down the beam focal spot from micro- to nanosize, a direct laser machining of materials with nanometer precision can be achieved [3].

REFERENCES

[1] Lipp, Ziaja, Classical Molecular Dynamics Simulations of Surface Modifications Triggered by a Femtosecond Laser Pulse, *Modelling 3*, 333 (2022); https://doi.org/10.3390/modelling3030021.
[2] M. Terakawa, “Femtosecond Laser Direct Writing,” in *Micro and Nano Fabrication Technology, Micro/Nano Technologies*, J. Yan, ed. (Springer, 2018), ISBN 978-981-13-0097-4.
[3] G. Vaschenko, A. Garcia Etxarri, C. S. Menoni, J. J. Rocca, O. Hemberg, S. Bloom, W. Chao, E. H. Anderson, D. T. Attwood, Y. Lu, and B. Parkinson, "Nanometer-scale ablation with a table-top soft x-ray laser," Opt. Lett. **31**, 3615-3617 (2006) .
[4] T. Zier et al.,"Signatures of nonthermal melting", Str. Dyn. 2, 054101 (2015).
[5] T.-H. Dinh et al., "Controlling the Response of Silicon to Strong Excitation – a Path towards Processing Materials at Sub-nanometer Precision", Commun. Phys. 2, 150 (2019) .
[6] I. Inoue, G. Yamaguchi et al., in preparation (2023).

X-ray Illumination of Phase Transitions through Shock Compression and Release

J. Wicks

The Johns Hopkins University, Baltimore, USA

XFEL-based Pump-probe XAS and XES Characterization of Photoexcited States in CeO_2

S. Pelatti[1,2], E. Spurio[1,2], J. S. Pelli Cresi[3], S. Benedetti[2], A. di Bona[2], S. D'Addato[1,2], P. Zalden[4], Y. Jiang[4], Y. Uemura[4], H. Wang[4], D. Vinci[4], X. Huang[4], F. A. Lima[4], M. Biednov[4], D. Khakhulin[4], C. Milne[4], S. Turchini[5], G. Ammirati[5], P.O'Keeffe[6], D. Catone[5], F. Boscherini[7], P. Luches[2]

[1]Dipartimento FIM, Università degli Studi di Modena e Reggio Emilia, 41125 Modena, Italy
[2]Istituto Nanoscienze, CNR (NANO-CNR) ,41125 Modena, Italy
[3]Elettra-Sincrotrone Trieste, 34012 Basovizza, Trieste, Italy
[4]European XFEL, 22869 Schenefeld, Germany
[5]Istituto di Struttura della Materia – CNR (ISM-CNR), EFSL, Rome, Italy
[6]Istituto di Struttura della Materia – CNR (ISM-CNR), EFSL, Monterotondo Scalo, Italy
[7]Dipartimento di Fisica e Astronomia, Alma Mater Studiorum − Università di Bologna, Bologna, Italy

ABSTRACT

Transition metal oxides have interesting properties that make them promising candidates as efficient photocatalysts. It is then essential to investigate the dynamics of the electronic and structural modifications induced by photoexcitation, to understand and possibly tune the functionality of the material. For some materials like CeO_2, the formation of photoinduced polarons can enhance reducibility and photocatalytic activity, while the structural deformations correlated with the charge localization, may also play a role in further decreasing the oxygen vacancy formation energy and enhance the redox functionality. In a recent study performed with pump-probe optical spectroscopy on CeO2, an ultrafast blue shift of photoinduced absorption by 0.4 eV was identified and tentatively ascribed to the formation of photoinduced small-polaron states within ultrashort times (330 fs) from photoexcitation [1].

Here we present an experiment that exploits ultrafast pump-probe XFEL-based XAS and XES to correlate the dynamics of the element-specific electronic modifications with the structural changes, to investigate the out-of-equilibrium processes caused by photoexcitation in CeO_2. The experiment has been conducted at the FXE instrument of the European XFEL on a 50 nm polycrystalline CeO_2 film. We performed pump-probe Ce L_3 edge X-ray absorption spectroscopy in the near (XANES) and extended (EXAFS) energy range and Ce L_α X-ray emission spectroscopy (XES) experiments, exploiting the ultra-short and ultra-intense XFEL pulses to probe the dynamics of photoinduced modifications of the local electronic and atomic structure around cerium ions with a time resolution below 100 fs. The film was excited using a laser pump with energy above the CeO_2 band gap (4.6 eV) and the dynamics of the excited states was probed within a delay time range extending up to 250 ps. The pump-probe XANES spectra indicate a Ce3+-like excited phase at short Δt (≤ 300 fs), which decays in ~300-400 fs into a metastable excited state, tentatively ascribed to the formation of a photoinduced small polaron. This result is confirmed by the pump-probe Ce L_α XES spectra measurements, that show compatible dynamics. The analysis of EXAFS spectra demonstrates the presence of structural distortions in the photoexcited state, compatible with small polaron formation.

REFERENCES

1. J. S. Pelli Cresi et al., J. Phys. Chem. Lett. 11, 5686 (2020

Pioneering Plasma Accelerators and Novel Sample Delivery Methods for Future FEL Experiments

Z. Ebrahimpour[1,2], F. Stellato[3], R. Mincigrucci[1], E. Principi[1]

[1]*Elettra-Sincrotrone Trieste S.C.p.A., Strada Statale 14 - km 163, 5 in AREA Science Park, 34149 Basovizza, Trieste, Italy*
[2]*INFN-Laboratori Nazionali di Frascati, Via E. Fermi 54, 00044 Frascati, Italy*
[3]*Physics Department, University of Rome Tor Vergata, and INFN-Sezione di Roma Tor Vergata, Via della Ricerca Scientifica, 00133 Roma, Italy*

ABSTRACT

The European Plasma Research Accelerator with eXcellence In Applications (EuPRAXIA) stands as an innovative facility at the forefront of scientific exploration, employing plasma-based technology. Recognized in the 2021 ESFRI Roadmap, EuPRAXIA constructs electron and laser-driven plasma accelerators to revolutionize ultra-fast science. EuPRAXIA signifies a transformative leap in ultra-compact accelerator development and applications, supported by the Preparatory Phase project (EuPRAXIA-PP) and the Italian Next Generation EU program (PNRR).

The EuPRAXIA@SPARC_LAB project, located at Laboratori Nazionali di Frascati of the INFN, endeavors to establish a cost-effective, high-gradient plasma-accelerated fifth-generation FEL facility [1]. Complemented by already initiated construction efforts, this facility aims for its first pilot user operation by 2030. EuPRAXIA@SPARC_LAB, the core beam-driven pillar, envisages FEL electron beams up to 1-5 GeV delivering photons at short wavelengths, marking Europe's first plasma accelerator-based research infrastructure.

An X-band normal conducting Linac will be used to accelerate the electron beam, followed by a Plasma WakeField Acceleration (PWFA) stage. Two beamlines are planned for the facility that produce ultra-bright photon pulses. The first beamline, AQUA, will utilize a SASE configuration, operating in the XUV to soft X-ray range spanning from 4 to 10 nm [2]. The second beamline, ARIA, will feature a seeded FEL setup, functioning within the wavelength range of 50 to 180 nm. [3]. These beamlines facilitate varied imaging and spectroscopy experiments, enabling the exploration of structural properties and dynamic behaviors across biological, soft matter, and inorganic samples [4].

Innovative sample delivery methods relevant for future large-scale user facilities, including EuPRAXIA sources as well as more conventional synchrotrons and FELs, are under study. In particular, the LEAPS- INNOV European project is fostering the development of the “optical tweezers” technique for manipulation, positioning and alignment of micron and submicron droplets of liquid samples in a vacuum environment.

An overview of the EuPRAXIA project, including the recent user survey initiative [5], as well as the description of the “optical tweezers” technique are presented in this contribution.

REFERENCES

1. A. Balerna, et al. EuPRAXIA@ SPARC_LAB for radiation-based techniques. *Cond. Matter*., 2019, 4: 30.
2. F. Villa, et al. EuPRAXIA@ SPARC_LAB status update *Proceedings 12581, X-Ray Free-Electron Lasers: Advances in Source Development and Instrumentation*, 2023, VI, 125810H.
3. F. Villa, et al. ARIA—A VUV Beamline for EuPRAXIA@ SPARC_LAB. *Cond. Matter*, 2022, 7, 11.
4. Z. Ebrahimpour, et al. *Cond. Matter*., 2022, 7, 72.
5. Conference attendees are invited to participate in the EuPRAXIA-PP survey: https://surveys.infn.it/index.php/718177