FELs OF EUROPE NEWS 1|22

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HED instrument, © European XFEL / Jan Hosan

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EDITORIAL

We proudly present you the first 2022 newsletter of the FELs OF EUROPE cooperation. Last year was another challenging, but overall, very successful year for us. Despite the difficulties caused by the pandemic we managed to perform a good range of important and exciting experiments at all European FEL-facilities, including a large number of experiments contributing to the fight against COVID-19. With this, we have demonstrated our abilities to react quickly, being able to switch focus of part of our user programs to emerging scientific and societal questions. Several highlights of the 2021 research are presented in this newsletter.

Looking back on 2021 and everything that we have achieved together, we are convinced that 2022 will be an even more successful year – despite the continued impact of COVID-19 on many aspects of our facilities operation and the daily lives of all of us. The new year started with another wave of the pandemic, but we are well-prepared now to deal with the related challenges and are confident to successfully carry on with the state-of-the-art research involving broad user community, remotely or in-person.

Upcoming events

We hope very much that beginning from spring 2022 we will be able to organize events in-person again. The first one is the FELs OF EUROPE Topical Workshop on Selected Problems in FEL Physics: from soft X-rays to THz, planned to be held on April 25-27 in Dresden. It is organized by the Helmholtz Zentrum Dresden-Rossendorf (HZDR) and addresses important topics of advanced schemes of FELs operation.

Preparation of the SCIENCE@FELs conference is in full swing. It will be held in-person from 20 - 23 September in Hamburg, organized jointly by DESY and European XFEL. The program committee is formed now and the speakers will be invited soon. During this conference we will celebrate the 10th anniversary of the FELs OF EUROPE cooperation in form of a dedicated session with a special festive agenda. The SCIENCE@FELs conference will be followed by the PhotonDiag 2022 workshop, to be held from 4 to 7 October, organized by the Helmholtz Zentrum Berlin.

We hope very much that all our efforts in 2022 will be successful. We will push ourselves to achieve our goals!

FELs OF EUROPE

Serguei Molodtsov
Chair FELs OF EUROPE

FERMI

An EUV X-ray polarimeter for single-shot FEL experiments

The analysis of the polarisation status of electromagnetic waves is fundamental for a large number of fields of research and technological applications. The majority of the polarization sensitive techniques work in the visible range, thanks to the abundance of optical elements like polarisers and wave plates. In the X-ray regime, the response of matter strongly depends on the energy of the incoming radiation, and any polarisation sensitive device needs to be specifically optimized. It is of fundamental relevance to extend polarimetry to the X-ray regime, as it would add element sensitivity. An example is to perform MOKE experiments in resonance with atomic core transitions. Such experiments, referred to as resonant MOKE (RMOKE), had been demonstrated in the EUV range. The advantages of RMOKE are M-edge feasibility, measurement of polarization rotation and ellipticity, use of linear polarization, giant Kerr effect, element sensitivity, and sub-picosecond time-resolution. In order to explore and establish detection schemes for resonant magneto-dynamical studies, we developed a polarimeter designed for the EUV photon energy range (10-125 eV), with single-shot sensitivity. The Wollaston polarimeter for X-ray FEL sources (TONIX) is coupled to a cryomagnetic chamber with 20 K and 1.6 T capabilities, and it is currently installed at the MagneDyn beamline at the FERMI FEL in Trieste, Italy.

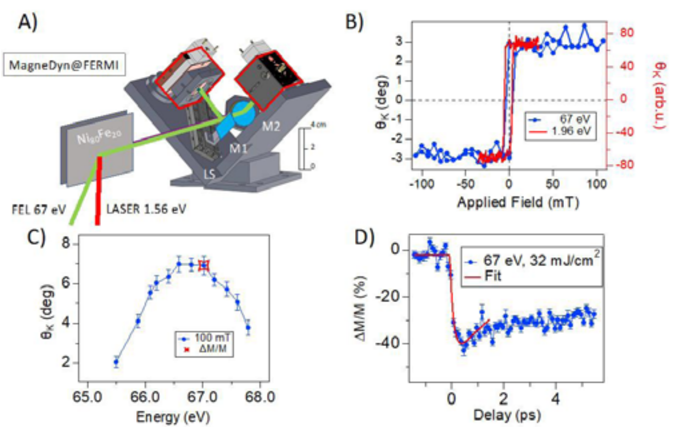

Fig. 1: A) Sketch of the experiment. The incoming FEL radiation (green beam) is reflected by the magnetic sample Ni80Fe20 at the angle of incidence of 45 deg. The reflected beam is collected by the TONIX polarimeter, where the Kerr rotation is measured. A pulse laser beam at 1.56 eV excites the system at a controlled time-delay with respect to the arrival of the FEL pulse.
B) Kerr rotation as a function of the applied field measured in the visible (red) and in the EUV range (blue).
C) Kerr rotation angle at maximum magnetization as a function of the incoming photon energy.
D) Time-resolved magnetization dynamics measured at 67 eV with 32mJ/cm^2 pump laser fluence.

In optics, polarization sensitive balanced photodetection is performed by: A) decomposition of the incoming beam into two components orthogonal in polarization and equal in intensity with the use of high-quality polarizers; B) simultaneous detection of the intensity of the two components; C) polarization angle reconstruction by the difference of the two signals. In the TONIX, the decomposition and the polarization selection are performed by two mirrors placed at the Brewster angle, mounted on two orthogonal planes of incidence. The mirrors are made of 100 nm Nb film grown on Si substrate by MBE technique. A 3D drawing of the assembly is shown in panel A. The first mirror M1, besides reflecting the beam to the first multichannel plate MCP-reflection (MCP-R), splits the beam in half. The second mirror M2 reflects the transmitted beam to the second MCP-transmission (MCP-T). M1 is mounted on a linear translation stage (LS) in order to fine tune the beam splitting in the exact half. Such geometry is chosen to maximise the sensitivity for incoming linear vertical or linear horizontal X-ray beam polarization states.

We tested the TONIX operation through RMOKE measurements on a $Ni_{80}Fe_{20}$ permalloy with the FERMI FEL pulses tuned at the Ni $M_{2,3}$ edge (Ni 3p, 67 eV). The experimental scheme is illustrated in panel A. First, we recorded a static Kerr hysteresis at 67 eV. As it is shown in panel B, the measurement is in agreement with the visible MOKE collected at 1.96 eV (632 nm). In panel C we show the sample Kerr rotation as a function of the FEL photon energy. We carried out time-resolved demagnetization experiments with 1.56 eV (794 nm) laser pump excitation, probing the Kerr rotation at 67 eV. In panel D we show the reconstructed demagnetization curve and the typical fitting function, consistent with the literature.

Interesting is the potential application of X-ray polarimetry in several research fields, considering in particular the extension to the soft X-ray regime. Technically, by replacing M1 and M2 with flat multilayer mirrors, it will be possible to extend the working range of the TONIX from the EUV to 1 keV, reaching the L-edge of all 3d transition metals. Accordingly, it will be possible to study dichroic phenomena like XMCD in ferromagnetic metals like Fe, Co, and Ni; charge and orbital order in cuprate superconductors or metal-insulator transitions in manganites.

Claudio Masciovecchio

Original publication:
A. Caretta, S. Laterza, V. Bonanni, R. Sergo, C. Dri, G. Cautero, F. Galassi, M. Zamolo, A. Simoncig, M. Zangrando, A. Gessini, S. Dal Zilio, R. Flammini, P. Moras, A. Demidovich, M. Danailov, F. Parmigiani, and M. Malvestuto, A Novel free-electron laser single-pulse Wollaston polarimeter for magneto dynamical studies, Structural Dynamics 8, 034304 (2021)
https://doi.org/10.1063/4.0000104

Superradiant operation of FERMI breaks the 10 fs barrier

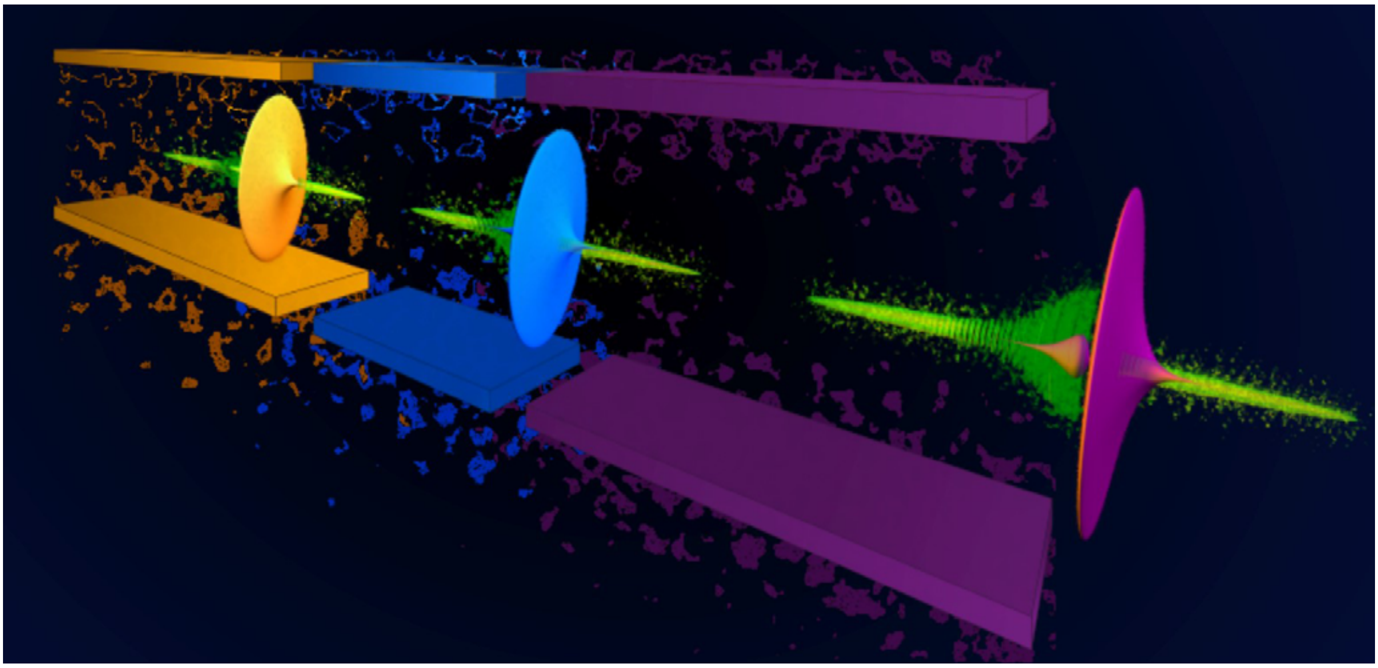

Fig. 2 Artistic visualization of the process of harmonic conversion and emission of light in the different stages of the superradiant cascade.

Generation of few-femtosecond pulses in the EUV-soft X-ray range is a primary goal of FELs. Such short pulses enable experiments to ‘freeze’ atomic motion and observe matter at the smallest scale, controlling chemical reactions, following the dynamics of chemical bonds or energy transfer processes in a variety of systems. The high intensities are crucial for exploring non-linear optical behavior. The 1s core holes of low-Z elements, and the 2p core holes of many light transition metals have lifetimes in this range; likewise fast nuclear motion in condensed matter and in molecules occurs on this timescale.

The pulse duration of a seeded FEL configured in the high-gain harmonic generation scheme (HGHG), the standard operation mode in FERMI FEL-1 and FEL-2, is proportional to the seed laser duration, and the scaling factor decreases with the increase of the harmonic up-conversion order. FERMI produces photon pulses in the wavelength range 4—100 nm, and durations in the range 20-70 fs (full width at half maximum). In order to reduce the pulse duration for a given harmonic order, one can (ignoring all considerations on intensity loss) decrease the seed duration, but a lower limit is imposed by the slippage of the electrons, that are inevitably slower than the light pulse: an infinitesimally short seed would indeed affect the (finite) portion of the electron beam overtaken by the light. Interestingly, slippage itself can be exploited to beat slippage: at the onset of saturation, a light pulse can propagate over fresh electrons, in the form of a self-similar solitary wave, undergoing longitudinal compression. The strong high-order components of the modulation at the leading edge of the pulse make for efficient frequency up-conversion: the idealized superradiant condition is attained in a cascade of undulators resonant at progressively higher harmonics of the initial seed. The team at FERMI realized a practical approximation of this condition by modifying the layout of FEL-2 as shown in Fig. 3.

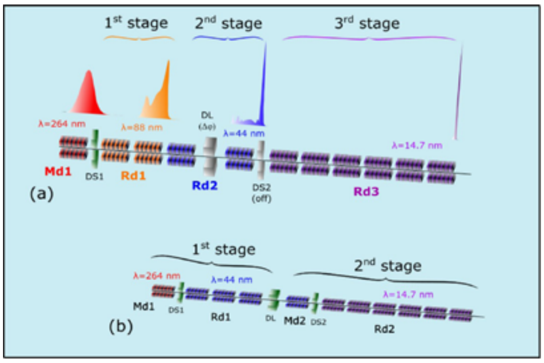

Fig. 3 FEL-2 in SR mode. Each of the radiators Rd1, Rd2 and Rd3 resonates at a low-order harmonic of the preceding one. The modulator Md1, resonant with the UV seed laser and separated from the first radiator Rd1 by the dispersive element (DS1), initiate the sequence. Seed intensity and dispersion are tuned to reach saturation at the exit of Rd1. The delay line (DL), set at low current (2.6 A), plays the role of a phase shifter ($\Delta\varphi$) between the two modules of Rd2.

The available undulator modules were apportioned into three amplifier sections tuned to harmonic 3, 6 and 18 of the seed, corresponding to a final wavelength of 14.7 nm. The spectra of the pulses produced by each section have been measured and compared with theoretical calculations that support the superradiant description; in particular the spectral bandwidth of the pulse is quantitatively consistent, in the Fourier transform limit, with the expected progressive reduction of the pulse duration. A direct autocorrelation measurement was performed at the Low Density Matter beamline (LDM) to further confirm this;

the measurement was based on the energy-resolved non-linear photoelectron signal from the two-photon above-threshold ionization (ATI) process in gaseous Argon. The full width at half maximum of the autocorrelation curves are 6.6 ± 0.9 fs and 31 ± 6 fs respectively, corresponding to a reconstructed pulse duration (FWHM) of 4.7 ± 0.6 fs in superradiant mode, versus 22 ± 4 fs in HGHG mode. Let us note that the former value is much shorter than the slippage in the modulator,≈23 fs, and that the pulse peak power in superradiant mode can be significantly higher (only marginally, in the present case) than in HGHG mode. Further work (not reported here) has been done, testing the tunability of the final wavelength; as well, a pilot spectroscopy experiment has been performed.

Carlo Callegari and Luca Giannessi

Original publication:
Mirian N.S., Di Fraia M., Spampinati S., Sottocorona F., Allaria E., Badano L., Danailov M.B., Demidovich A., De Ninno G., Di Mitri S., Penco G., Rebernik Ribič P., Spezzani C., Gaio G., Trovó M., Mahne N., Manfredda M., Raimondi L., Zangrando M., Plekan O., Prince K.C., Mazza T., Squibb R.J., Callegari C., Yang X., Giannessi L., Generation and measurement of intense few-femtosecond superradiant extreme-ultraviolet free-electron laser pulses, Nature Photonics,15, 523-529 (2021)
Doi: 10.1038/s41566-021-00815-w

EUXFEL

Completion of four successful operational years at European XFEL

2021 marks the fourth successful year of user operation at European XFEL located near Hamburg, Germany. Over the year more than 1000 users performed 44 experiments with more than 4600 hours of beamtime. 220 users were able to join on site. As of 2021, the users now have the possibility to stay in the new guesthouse directly on campus. The guesthouse, which was inaugurated in June, has 58 beds in 55 rooms, and enables visiting scientists make efficient use of their time at the experiment stations and be more flexible with their free time.

The Helmholtz International Beamline for Extreme Fields (HIBEF) was inaugurated on 31 August. The beamline will be part of the High Energy Density experiment station which enables deep insights into the structure of materials and very fast natural processes in plasma physics, thus driving innovations in materials and accelerator research.

Detail view inside the TWIN amplifier of the HIBEF HI Laser at the HED instrument showing Ti:Sapphire gain medium encased in a cryogenic cooling chamber.
Copyright: European XFEL / Jan Hosan

In total, six instruments, are currently available in the experiment hall for a wide range of experiments in various fields such as biochemistry, medicine, nanotechnology, material sciences, and energy technology. The following highlights provide an insight into some of the experiments performed over the last year.

An interdisciplinary team of scientists led by Johannes Hagemann from DESY investigated extremely fast exploding jets of water using ultrashort X-rays pulses.[1] In their experiment, they used a jet of water with a diameter of just 0.04 millimetres. The infrared light of the laser caused the water to heat abruptly and evaporate within 15 nanoseconds. The exploding water jet was then analysed using the received images from the MID instrument. The results are of practical importance, as rapid evaporation by short laser pulses can be used for medical surgery, for example. In addition, this experiment has shown that such fine water jets are also suitable for carrying larger objects, such as intact, living cells, into the X-ray beam to study their structure. The use of a water jet has the advantage that the cells remain in an aqueous environment as in the body. They do not have to be immobilised or dried.

In the field of coronavirus research, experiments have shown new insights in the quest to fight COVID. Research teams from European XFEL, EMBL-Hamburg, CFEL, DESY and the Max Planck Institute for the Structure and Dynamics of Matter used Small Angle X-ray Scattering (SAXS) to study samples in their natural form under physiological conditions where biological reactions occur. Using the SPB/SFX instrument, they derived corresponding form factors, which are the observed scattering patterns with all noise and background removed. Leaving only the signal from the sample, a form factor provides information about a protein's shape

EUXFEL

and size and how it changes when, for example, antibodies bind. The hope is that this will provide researchers with a new tool that can improve understanding of our bodies' immune response to coronavirus and help develop medical strategies.

An international research team led by Maria Novella Piancastelli and Renaud Guillemin from the Sorbonne in Paris, Ludger Inhester from DESY and Till Jahnke from European XFEL presented their findings on how radiation damage occurs in biological tissue in the journal Physical Review X[2]. They studied in detail how water molecules are broken apart by high-energy radiation, creating potentially dangerous electrically charged ions that can then trigger harmful reactions in the organism. Using soft X-ray pulses at the SQS instrument, they bombarded single water molecules, some of which absorbed more than one photon due to the intense pulses. Using a reaction microscope, the team measured the flight directions and velocities of the fragments. The movement of the molecule's atoms that occurred between the two absorptions was carefully analysed and allowed the scientists to record the disintegration of the water molecule, which lasted only a few femtoseconds, in the form of a film. The newly gained insights address elementary questions about reaction dynamics in water. In addition, these results have shown that it will also be possible to study other solvents and molecules with more complex structures, such as ethanol or cyclic compounds, which are of great interest in chemistry and other disciplines.

Interaction chamber 2 was pushed into the beam. From left to right: Karin Prien (Minister for Education, Science and Culture, Schleswig-Holstein), Prof. Robert Feidenhans'l (Chairman of the European XFEL Management Board), Christian Luft (State Secretary BMBF), Dr Eva Gümbel (State Secretary, Hamburg Ministry of Science, Research, Equalities and District Authority), Prof. Sebastian M. Schmidt (Scientific Director of the HZDR). Copyright: European XFEL / Axel Heimken

In another research highlight, an international group of researchers led by Yongjae Lee from Yonsei University in South Korea pioneered a new way for conducting static high-pressure and high-temperature experiments using diamond-anvil cells.[3] They discovered a new, faster method to produce iron nitrides by using an iron foil with nitrogen under X-ray bombardment at 50,000 times atmospheric pressure in diamond-anvil cells (DACs). A series of ultra-bright X-ray pulses both initiated and probed the chemical reaction in the samples, which were as hot as 5400 degrees Celsius. Under the right pulse conditions, they observed the formation of the iron nitride known as ϵ-$Fe_3N_{1.33}$ in just hundreds of nanoseconds – about a billion times faster than with other techniques used previously. Iron nitrides are an important group of industrially relevant materials and promising candidates for high-density data storage and other applications.

In summary, 2021 was a successful year for users and visiting scientists at European XFEL despite the pandemic. Although the COVID-19 situation continues to be very challenging, European XFEL is cautiously optimistic for 2022, and expects to be able to further increase the beamtime available for the users as well as the number of users on site.

Sarah Aretz

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[1] Johannes Hagemann, Malte Vassholz, Hannes Hoeppe, Markus Osterhoff, Juan M. Rosselló, Robert Mettin, Frank Seiboth, Andreas Schropp, Johannes Möller, Jörg Hallmann, Chan Kim, Markus Scholz, Ulrike Boesenberg, Robert Schaffer, Alexey Zozulya, Wei Lu, Roman Shayduk, Anders Madsen, Christian G. Schroer and Tim Salditt, Single-Pulse Phase-Contrast Imaging at Free-Electron Lasers in the Hard X-ray Regime, Journal of Synchrotron Radiation 28, 52-63 (2021) DOI: 10.1107/S160057752001557X

[2]. T. Jahnke, R. Guillemin, L. Inhester, et al.; Inner-Shell-Ionization-Induced Femtosecond Structural Dynamics of Water Molecules Imaged at an X-Ray Free-Electron Laser, Physical Review X 11, 041044 (2021) DOI: https://doi.org/10.1103/PhysRevX.11.041044

[3] Huijeong Hwang et al, X-ray Free Electron Laser-Induced Synthesis of ϵ-Iron Nitride at High Pressures, Journal of Physical Chemistry Letters12, 12, 3246–3252 (2021) doi.org/10.1021/acs.jpclett.1c00150

SwissFEL

Two-color X-ray FEL by photocathode laser emittance spoiler

X-ray FELs are well-suited to interrogate a wide range of processes initiated using conventional lasers. In these pump-and-probe schemes, the FEL and the laser are typically synchronized at best within tens of femtoseconds. The challenge of understanding the physics of transient processes in depth would be facilitated by higher temporal synchronization. Two-color hard X-ray emission is an emerging mode of operation for FELs, which has the potential not only to reduce the timing jitter between the pump and the probe by generating the two pulses from the same electron bunch, but also to enable new X-ray multispectral spectroscopy schemes. Until now, two-color FELs have been realized with less efficient use of the undulators or through a time-consuming machine setup which is not always compatible with user facilities operation.

At SwissFEL, we recently demonstrated a straightforward and noninvasive method to produce two-color X-ray pulses. This new approach relies on electron beam emittance spoiling based on the use of two photocathode drive lasers. A sub-picosecond laser pulse (emittance spoiler) is overlapped with the photocathode laser. The excess of charge leads to a localized increase of the emittance, which prevents FEL amplification. The unspoiled slices of the bunch still produce FEL emission which is centered at two different wavelengths due to the linear energy chirp accumulated along the linear accelerator. The temporal and spectral separation between the two colors can be tuned by changing the compression and the energy chirp of the electron beam or by adjusting the intensity of the laser emittance spoiler. The time delay can be varied between a few and hundreds of femtoseconds. The maximum relative color separation expected at SwissFEL is approximately 2%.

The laser emittance spoiler surpasses other two-color FEL approaches in many relevant figures of merit. This method allows, in fact, fast and efficient setup of the two-color FEL using the optimal machine configuration. As a result, high FEL pulse energy and remarkable intensity and spectral stability can be achieved. In the experiment at SwissFEL, we produced two-color pulses around 12 keV with energy of 160 microjoules corresponding to the 56% of pulse energy of standard SASE FEL mode. In contrast to other two-color schemes, the laser emittance spoiler does not require solid obstacles placed on the electron bunch or complex beam manipulation. Therefore, it does not cause beam radiation losses and it is readily applicable to high-average power/high repetition rate FELs. In addition, this method allows FEL pulse shaping as it introduces an additional means of control over the duration of the individual (single-color) pulses and their temporal and spectral separation. Finally, with the laser emittance spoiler, sophisticated pump and probe schemes with shot-to-shot mode switch between one and two color FEL become feasible.

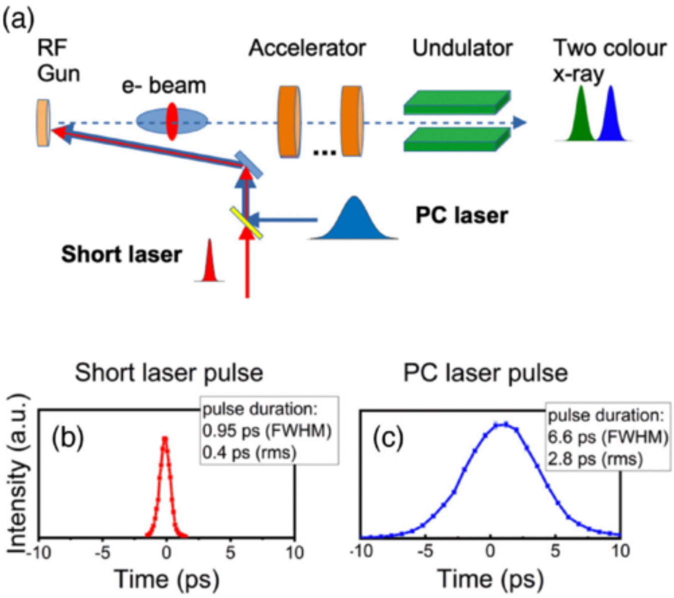

Fig. 4 Experimental setup and two color FEL spectra for different delays of the laser emittance spoiler. Copyright: PSI

Ernest Courant Outstanding Paper Recognition

The original article has been selected as the winner of the first Ernest Courant Outstanding Paper Recognition, a honor sponsored by the Journal Physical Review Accelerators and Beams (PRAB) and the APS Division of Physics of Beams (DPB). This honor recognizes the most outstanding paper published in PRAB annually.

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Original publication:
Carlo Vicario, Simona Bettoni, Alberto Lutman, Andreas Dax, Martin Huppert and Alexandre Trisorio,
Two-color x-ray free-electron laser by photocathode laser emittance spoiler, Phys. Rev. Accel. Beams 24, 060703 – (2021).
https://doi.org/10.1103/PhysRevAccelBeams.24.060703

Single-atom catalysis: how a C_{60} molecule can help a V^+ ion to split water

Water splitting is an important source of hydrogen, a promising future carrier for clean and renew-able energy. Hydrogen is currently formed through electrolysis, at an energetic price. This can be reduced by the use of a suitable catalyst, but remains unfavorable. To design future, more efficient catalysts a detailed understanding of the water splitting reaction mechanism over catalysts is essen-tial.

Catalyst material is often made up of metal particles, with nanometer dimension to ensure the most efficient use of the surface of the particles, frequently consisting of precious metals. The ultimate size limit of such a particle is that of a single atom. In that so-called single-atom catalysis regime, the atomic chemical properties are very different from those of the bulk, offering exciting prospects in terms of both activity and efficiency.

To get a fundamental understanding of the chemical activity of single atoms, a vast body of research has addressed reactions of single atoms with water by isolating them in the gas phase. However, in real-life electrolysis, the catalyst material must be attached to the surface of either anode or cathode, often done through embedding the active metal material on a host or support of for instance amorphous carbon or alumina. This embedding can have a profound influence on the reaction of the catalyst with the water molecule, and is often very hard to study in detail.

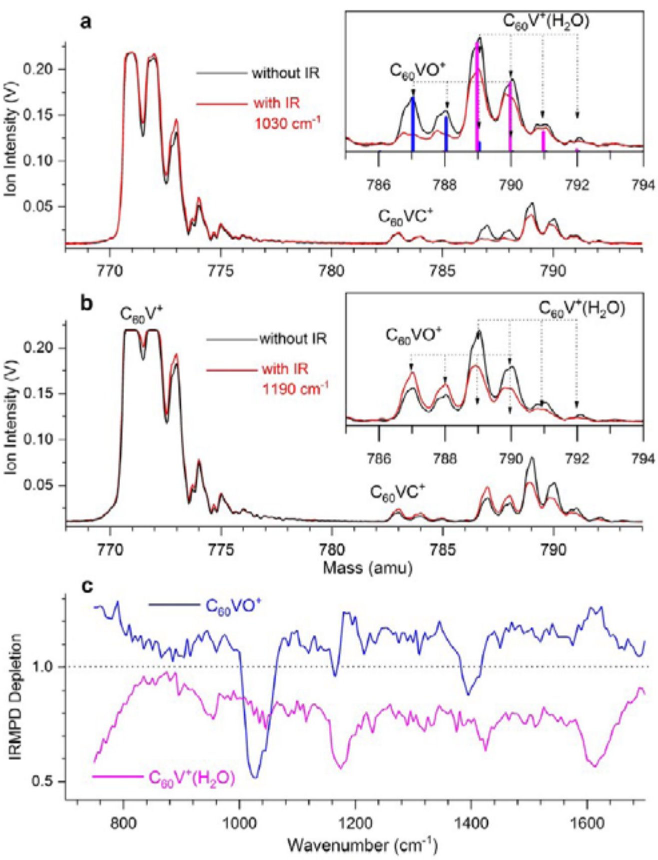

Fig. 5 : (a+b) Mass distributions of $C_{60}V^+$, $C_{60}VO^+$, and $C_{60}V^+(H_2O)$ with (red) and without (black) IR irradiation at 1030 cm-1 (a) and 1190 cm-1 (b). The simulated natural isotopic distributions of $C_{60}VO^+$ and $C_{60}V^+(H_2O)$ are presented separately in the inset of panel a by blue and pink sticks, respectively. (c): IRMPD depletion spectra of $C_{60}VO^+$ (blue trace) and $C_{60}V^+(H_2O)$ (pink) show the concurrent $C_{60}VO^+$ signal growth and $C_{60}V^+(H_2O)$ signal loss indicative of H_2 loss via $C_{60}V^+(H_2O) \rightarrow C_{60}$ + VO+. The $C_{60}VO^+$ spectrum exhibits a combination of growth due to H_2 loss from $C_{60}V^+(H_2O)$, and loss via $C_{60}VO^+ \rightarrow C_{60} + VO^+$ (most pronounced around 1030 and 1400 cm-1).

A team of researchers of HFML-FELIX and KU Leuven used a trick to be able to use the highly sensitive instrumentation that has been developed to study gas-phase reactions to explicitly study the influence of the support on the chemical activity of a single vanadium atom. They synthesized and ionized a complex of fullerene C_{60}, the football-shaped molecule, with a single vanadium atom bound to one of the C_{60} surface hexagons. The C_{60} thereby adopted the role of support for the vanadium single-atom catalyst. They then evaluated how a water molecule adsorbs on the vanadium atom using IR spectroscopy facilitated by the FELICE intracavity free-electron laser. Through the IR spectral signature, they found that the water adsorbs intact onto the $C_{60}V^+$ complex. However, once IR photons were absorbed, they found that a H_2 molecule was formed and ejected from the $C_{60}V^+(H_2O)$ complex. This implies that the injection of a minute amount of vibrational energy is sufficient to drive the water splitting reaction. Because this effect was not observed in a similar study of $V^+(H_2O)$, the C_{60} support's influence must be decisive in this process.

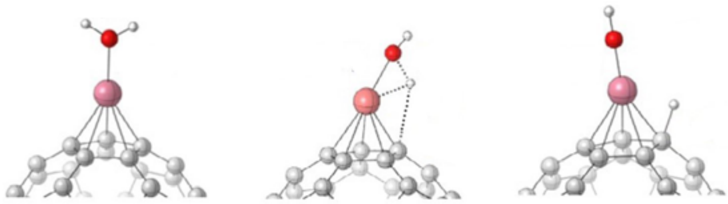

Fig. 6 : Snapshots from the calculated reaction path for IR-induced water splitting on a C_{60} sup-ported V^+.

Accompanying theoretical calculations explore the detailed reaction pathway of water adsorption, and the subsequent water splitting reaction. By comparing this reaction for V^+ alone with that for $C_{60}V^+$, it could be demonstrated that the C_{60} support has an important role in lowering the reaction barrier due to a large orbital overlap of one water hydrogen atom with one of C_{60}'s carbon atoms. In a way, the C_{60} offers the hydrogen atom a convenient temporary waiting room to be pulled off the water molecules, which is not offered when only the V+ is there. This fundamental insight in the water splitting reaction showcases the importance of supports in single atom catalysis, and how they modify the reaction potential energy surface.

Joost Bakker

Original publication:
Gao-Lei Hou, Tao Yang, Mengyang Li, Jan Vanbuel, Olga V. Lushchikova, Piero Ferra-ri, Joost M. Bakker, Ewald Janssens, Water Splitting by C_{60}-Supported Vanadium Sin-gle Atoms, Angewandte Chemie International Edition 60 52,27095-27101 (2021)
https://doi.org/10.1002/anie.202112398

FLASH

FEL-based time-of-flight momentum microscopy: 3 time-resolved photoemission modalities in 1 experiment

In the quest to gain predictive understanding of the functionality of condensed matter systems, the energy and momentum analysis of photoemitted electrons provide the most direct insights. When photoemission spectroscopy (PES) is used in combination with high-repetition rate free-electron lasers in the XUV and soft X-ray regime such as FLASH (DESY, Hamburg) or with table-top laser systems based on high harmonic generation this has a high potential for time-resolved PES. The possibility to use both X-ray and laser pulses with time duration of few tens of femtoseconds allows one to access ultrafast electronic phenomena, lattice dynamics and chemical reactions. The photo-emitted electrons carry all the information regarding the electronic states of the system in the photo-generated non-equilibrium state. To fully exploit this information, it is necessary to use very efficient detection schemes for the photoelectrons, such as a time-of-flight momentum microscope – e.g. HEXTOF setup[1].

HEXTOF merges three photoemission spectroscopy techniques into a single setup, namely time-resolved momentum microscopy (trMM), time-resolved X-ray photoelectron spectroscopy (trXPS), and time-resolved X-ray photoelectron diffraction (trXPD). Traditionally, these experiments are carried out using separate instruments. However, we managed to combine all three techniques into the same instrument. The combination with pump-probe schemes of FLASH extends the available range for measurements into the sub-picosecond time domain. With this instrument the two momenta of photoelectrons transversal to the sample surface can be measured simultaneously (trMM) typically from the valence and conduction bands of materials. Furthermore, it is possible to investigate the dynamics of electronic core levels (trXPS) or momentum-resolved changes in the coherent photoelectron scattering with bulk and surface (trXPD).

HEXTOF is routinely used at the PG2 monochromator beamline at FLASH for pump probe photoemission studies, as well as - in between FLASH beamtimes - with a table-top high harmonic laser source in the Center for Free-Electron Laser Science (CFEL) to perform either sample pre-characterization before FLASH beamtimes or independent research activities in collaboration with users. It features an experimental energy resolution of 130 meV, a momentum resolution of 0.06 Å^{-1}, and a system response function of 150 fs could be achieved (all values are full width at half maximum). It includes also a sample preparation chamber and corresponding characterization tools. The overall performance was demonstrated by investigating the carrier dynamics in a model system of the layered semiconductor – tungsten diselenide (WSe_2)[1,2].

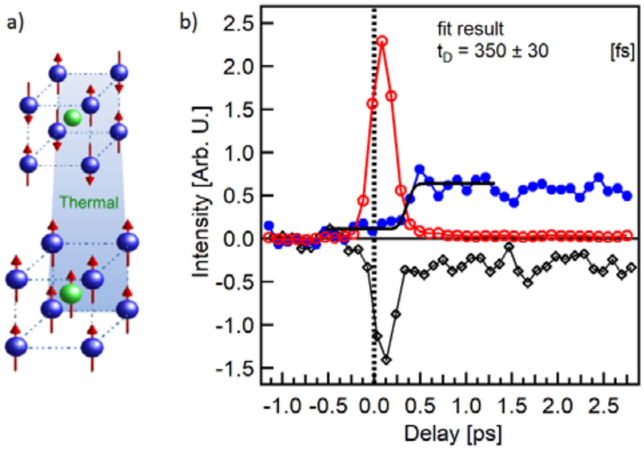

Fig. 7 (a) Sketch of the isostructural metamagnetic phase transition in FeRh. At room temperature (top) the system is anti-ferromagnetic showing atomic magnetic moments only at the Fe atom sites ($m_{Fe}=\pm 3.3\mu_B$). Above 360 K (bottom) the system has ferromagnetically coupled magnetic moments at the Fe ($m_{Fe}=3.1\mu B$) and Rh ($m_{Rh}=0.9\mu_B$) sites. The whole unit cell expands isotropically by about 1% in volume. (b) Electronic structure evolution during the laser induced phase transition. The blue circles show the appearance of the ferromagnetic state within the first 500 fs after laser excitation.

Recently, HEXTOF was used at FLASH to investigate the role of electronic structure and charge transfer in the first order phase transition in FeRh, by combining time-resolved soft X-ray photoemission results with time-dependent Density Functional Theory (DFT) calculations. The studies showed that an ultrashort laser pulse can indeed induce a first order phase transition. Consequently, the magnetic moments switch from an anti-parallel to a parallel configuration. The evolution of the electronic state of FeRh during this transition were measured with femtosecond temporal resolution. For the interpretation of the experimental results, the collaboration with two teams of the European Theoretical Spectroscopy Facility (ETSF), based in Rome and Paris, was essential. The interpretation of the experimental results revealed that the Optical Inter-Site Spin Transfer (OISTR), namely the ultra-fast "electron charge and spin transfer" from Rh atoms to Fe atoms is the most suitable candidate as a trigger for the phase transition. Furthermore, it could be shown (see Fig.7) that the time-scale of the induced phase transition is on the order of few hundreds of femtoseconds[3].

These results will foster the understanding of the fundamental triggers of electronic and magnetic phase transitions also in other system with similar behaviors such as topological insulators or cobaltites. Spin and charge transfer, where electronic and magnetic degrees of freedom are strongly correlated, are probably playing a more important role in ultrafast dynamics than previously expected. Future technologies will be based on the controlled manipulation of the electron spin, which may open the pathway towards higher storage density memories, faster switching speed and more efficient power use.

Dmytro Kutnyakhov

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FERMI

FERMI 2.0 upgrade plan

In December 2010 the world's first externally seeded FEL facility, FERMI at the Elettra Sincrotrone Trieste laboratory, produced the first VUV pulses. FERMI began user operation in 2012 with a scientific impact that has been one of the most successful of the recent years, with an intense effect on different scientific fields, ranging from nonlinear optics to atomic and molecular science and from extreme thermodynamic conditions to ultrafast magnetism.

FERMI can provide ultrashort pulses with unprecedented stability in terms of energy and time jitter. The seeding scheme offers the possibility to carry out experiments based on multi-field, phase-controlled interactions in atoms and molecules, as well as in solid state samples. The achievements obtained at FERMI are influencing the future development of other FEL facilities around the world, including FLASH (Germany), the Swiss-FEL (Switzerland) as well as the Linac Coherent Light Source at Stanford (US).

Based on experiences and feedback coming from the user community, FERMI is undergoing an upgrade plan, as described in the FERMI 2.0 Conceptual Design Report.

The ultimate goals are extending the emission spectral range to fully cover the water window and to reduce the pulse duration below the characteristic lifetime of core holes in this photon energy range. One of the main pre-requisites of this upgrade is the preservation of the uniqueness of FERMI, i.e. the possibility to control the properties of the radiation by seeding the FEL with an external laser system. The seed allows control of the microbunching formation in the electron beam leading to FEL amplification of almost Fourier transform-limited pulses.

The panorama of the possible technical solutions is evolving, with new schemes and ideas coming into play. One of the most promising approaches is echo-enabled harmonic generation (EEHG), where two external seed lasers are used to precisely control the spectro-temporal properties of the FEL pulse[2]. The scheme was demonstrated at FERMI in 2018[3]. The experiment consisted of a modification of the FEL-2 layout to accommodate the installation of dedicated systems necessary for the EEHG. To extend the FEL-2 spectral range to include the oxygen K-edge, two options were considered, either by using EEHG directly or with a cascade employing both EEHG and HGHG techniques in the present "fresh-bunch" injection technique (see Fig. 8).

The first solution requires a first large dispersion chicane of up to 15 mm for optimized EEHG operation. This makes the scheme prone to a number of effects which may result in a degradation of the FEL spectral purity and of the FEL gain in the final radiator. The large chicane is indeed an amplifier of microbunching instability (MBI). The resulting modulations are further amplified and converted into density modulations by the dispersion in the second modulator and the second chicane. These modulations are the source of sideband structures in the final FEL spectrum, degrading the FEL spectral purity. A second issue is the emission of incoherent synchrotron radiation (ISR) and the intra-beam scattering (IBS) along the chicane. These two effects are the source of mixing of the filamented phase space that produces the

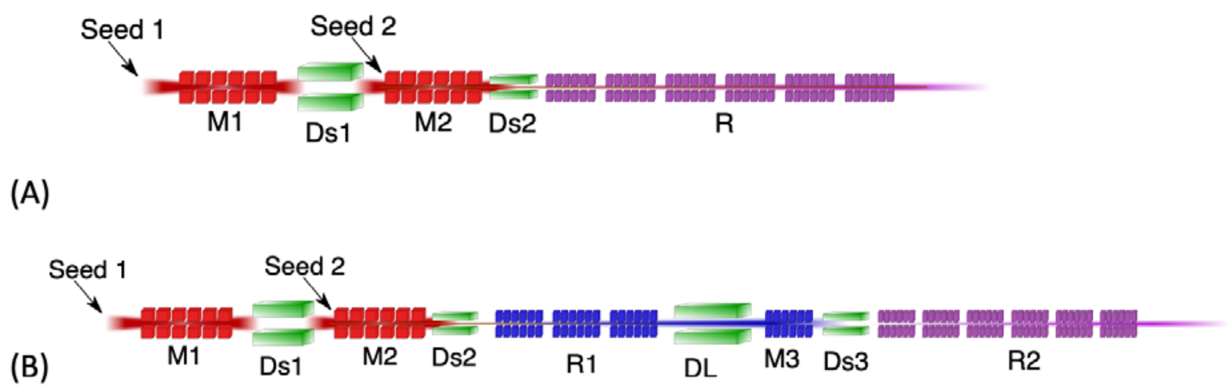

Fig 8: (A) Single stage EEHG FEL Configuration (B) Double stage cascade where the first stage is an EEHG FEL and the second stage a HGHG FEL. The second stage is seeded by the first stage in fresh-bunch injection configuration.

high harmonic bunching in EEHG after the second chicane, a factor reducing the bunching at the entrance of the amplifier. The result is a longer saturation length, and a larger SASE background. All these effects are mitigated in a scheme where the chicanes have a lower dispersion. This is the reason why we considered a second option, where the EEHG generates a seed that is then used in fresh-bunch to seed a second HGHG stage, similarly to what is done in the present FEL-2 configuration. The present double-stage HGHG with fresh-bunch scheme, can be upgraded by converting the first stage to an EEHG configuration aimed at reaching harmonics of the order of 30. The second stage would then up-convert the output of the first stage to harmonics of the order 120-130 as required.

In view of the upgrade, the photon energy distribution between the two FERMI FELs has to be adapted to the upgraded scenario, with FEL-1 still covering the low photon energy range, but extended to reach a photon energy of 100 eV, and FEL-2 dedicated to the high energy range, from 100 eV to about 550 eV. [1]

L. Giannessi, C. Masciovecchio

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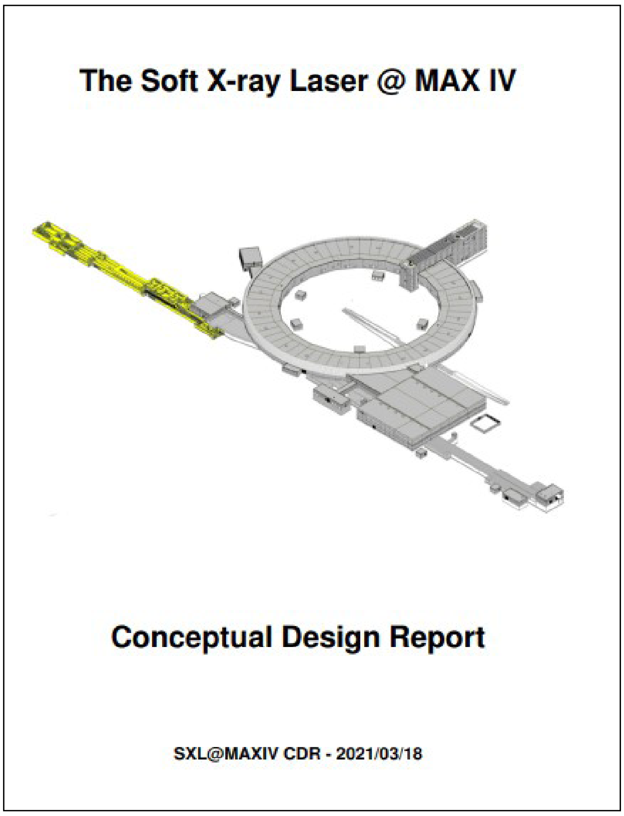
MAX IV

MAX IV: continued development of the science case for the Soft X-ray Laser

As reported in the previous newsletter, we finalized the Conceptual Design Report (CDR) for a soft X-ray FEL (SXL) at the MAX IV Laboratory in spring 2021.

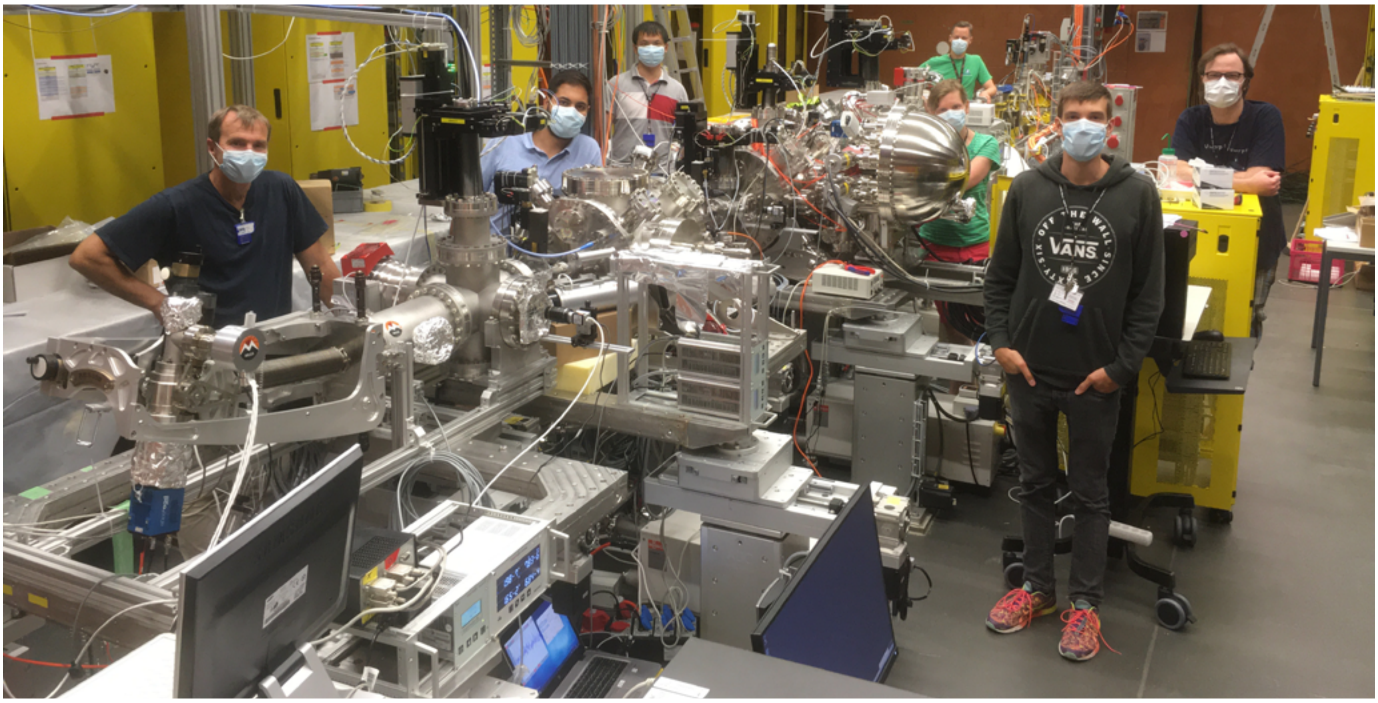
Following the publication of the CDR, the Swedish community of existing and potential FEL users formed a science case core group. Members are from major universities in Sweden and cover all the science fields from the original Science Case. During fall 2021, the core group initiated the update of the Science Case based on the CDR, arranging a number of events involving the Swedish as well as the international user community. Following a digital presentation of the CDR in September, we arranged an online meeting on Ultrafast X-ray science using SXL at MAX IV in October, during the MAX IV User Meeting. It gathered about 100 participants. Based on the CDR, the original Science Case and the input of speakers from the international FEL user community, the meeting highlighted the future possible science at SXL. It resulted in a user-driven Expression of Interest for SXL that was submitted as input to the future strategy and roadmap of the MAX IV Laboratory. In addition, it initiated the update of the Science Case, a process that we will finalize at a coming larger workshop in **Stockholm on June 13-14, 2022** in preparation for the start of the work with the Technical Design.

Per Eng-Johnsson
Lund University & Sverker Werin, MAX IV Laboratory



PSI

SwissFEL Athos/Maloja into user operation



A great success, bringing the Athos/Maloja endstation in times of corona to the user operation phase. The happy Maloja endstation team! From front to rear: Gregor Knopp, Jonas Knurr, Andre Al Haddad, Sven Augustin, Kirsten Schnorr (head of the Maloja team), Zhibin Sun, and Christoph Bostedt (Head of Laboratory for Synchrotron Radiation and Femtochemistry). Missing on the photo are Florian Amrein and Simon Tiefenbacher. Copyright photo: PSI

At the SwissFEL X-ray free-electron laser of the Paul Scherrer Institute (PSI), the second beamline is going into user operation. Parallel operation of the already working hard X-ray beamline Aramis and the new soft X-ray beamline Athos is successfully realized. The energy range of the Athos branch includes the absorption K-edges of the important light elements such as C, O and N or the L-edges of 3d transition metals such as Ti, Mn, Fe, and Cu. The Aramis and Athos electron bunches rushing down the accelerator are separated by a time interval of just 28 nanoseconds and then separated by a fast switching element developed at PSI, that 'kicks' the two electron packets apart with high stability. In Athos the electrons continue their way directly to a novel undulator line, very different from the Aramis undulators.

Between every two undulators there is a "chicane", that allows the electron packets to be compressed or shifted. The combination of CHIC ('Chicanes for High Power and Improved Coherence') and the Athos APPLE-X undulator line is unique in the world and the elaborate design allows controlling the properties of the light and a range of different operating modes. It is possible to generate extremely short light pulses, or flashes with a narrow energy distribution, or even double pulses for pump/probe experiments. The installations in the beamline and the experiment station went hand in hand. The last Athos undulators were set up in 2021, which was directly followed by the first successful pilot experiments at the Maloja AMO endstation involving teams of 'friendly' users from different countries.

The Maloja endstation is specialized in studying ultrafast processes in atomic, molecular, nonlinear and chemical sciences. The overall design of the instrument is flexible with the goal to support a wide range The first attempts pilot experiments at the Maloja endstation employed time resolved optical-X-Ray and X-Ray spectroscopy to unravel correlated electron and nuclear dynamics. Soft X-ray pulse parameters achieved during the pilot phase include a two color mode with a variable time delay between -20 fs and 600 fs, pulse durations below < 20 fs (FWHM) and flexible energy separation between 350eV and 1000eV.

The highly intense Athos branch X-ray pulses also enabled the observation of nonlinear X-ray phenomena in the form of an observed stimulated emission process in Neon gas. The experiment verified the applicability of Athos/Maloja for time-resolved spectroscopy as well as future nonlinear X-ray investigation. Concurrent to the pilot experiments the first call for proposals was published. The user beamtimes from the first call in September 2021 will commence this spring. The Athos branch is now in user operation and part of the SwissFEL call with submission deadlines in mid March and mid September.

Gregor Knopp
Paul Scherrer Institute

FLASH

Updates from the FLASH2020+ project at DESY

Lucas Schaper (left) takes over the lead of the FLASH2020+-project from Enrico Allaria (right). Copyright photo: DESY/M. Mayer

In the current 9 month shutdown of the FLASH facility, the upgrades within the FLASH2020+ project focus on the accelerator section to broaden the available parameter range for the user community already in late 2022.

Starting mid-November 2021, the superconducting accelerator modules warmed up from their 2K operation temperature. They reached ambient temperature some weeks later, allowing for the replacement of two out of the seven accelerating modules. The year 2022 started thereby with the uncommon view of the FLASH accelerator uncovered, where the roof has partially been opened to allow for the removal of the old modules. They will be exchanged by two new state-of-the-art modules which are following the XFEL design and will bring up the maximum electron energy of FLASH by 100MeV, resulting in a total of 1.35GeV. This will enable FLASH photons to reach further into the water window and reduce the shortest wavelength by about 20%.

In the wake also the surrounding sections will be upgraded with new components vital for future operation, e.g. the new laser heater, the new injector lasers and the afterburner, installed in the FLASH2 beamline. The new injector lasers, which provide the ultraviolet photon pulse to release electrons at the photocathode, are an upgrade to more easily maintainable laser technology while at the same time allowing for even better disentanglement of the two FEL beamlines. The lasers will be housed in the same new laboratory as the laser heater laser. Just upstream of the first bunch compressor (BC1) this laser interacts with the electron bunch and allows to modify its energy spread and thus has direct impact on the gain of the microbunching instability. Downstream BC1 the electron beam traverses the new accelerating modules, followed by a renewed second bunch compressor section designed to correct intra bunch transverse orbit correlations and increased diagnostic capabilities. To even more effectively use the higher electron beam energy, a so called called afterburner will be installed in the FLASH2 beamline downstream of the main undulators allowing to increase the pulse energy at the third harmonic of the main undulators and additionally allow for variable polarization (including circular) at wavelengths down to 1.33 nm. This radiation will also be transported through the new beamline FL23 that is currently being set up, which implements a time-compensating monochromator for the first time at an XUV FEL and thus allows for keeping shortest pulse durations at much smaller energy bandwidth, right towards the Fourier-limit.

Mechanical and general installation work in the tunnel is well on track for this first upgrade phase. For the second upgrade in 2024, where the complete 110 m FLASH1 beamline will be replaced by a dedicated external seeding beamline, the preparations are progressing. For example, simulations for the optimal implementation of the external seeding at the full repetition rate of FLASH (MHz bursts repeated at 10 Hz) are ongoing. New prototypes for the new variable gap and variable polarization undulators for FLASH1 have been fabricated and the design is being finalized. A substantial upgrade of the photon diagnostics section is foreseen to be ready for seeded operation. At the same time, the pump-probe lasers will be completely overhauled and will offer many new opportunities for users.

In parallel to the numerous modifications in the experimental facilities the project management also exhibited a change: The former project leader Enrico Allaria has moved back to Italy for private reasons, and Lucas Schaper, the former deputy of Enrico, has taken over the project leadership and is excited to bring new opportunities for users in 2025 with a transformed FLASH facility.

Lucas Schaper, Martin Beye, Karolin Baev

Save the date: Science@FELs 2022: 19-22 September

The international Science@FELs 2022 conference will take place from 19-22 September 2022 in the Hamburg Metropolitan Area. Following the first fully online Science@FELs 2020, also organized by DESY and European XFEL, we hope to be able to finally see you again in person in Hamburg.

Science@FELs 2022 will focus on recent scientific highlights and related research activities from FEL facilities world-wide. The meeting will bring together scientists from all areas active in research with FELs, featuring invited and contributed talks, poster sessions, tours of the European XFEL and FLASH and tutorials for PhD students and young postdocs. The 'FELs of Europe award on FEL Science and Applications 2022' will be awarded during the conference in recognition of recent work in the area of free electron laser science and applications in its broadest sense by a young scientist.

This will be the 6th conference of the Science@FELs series which his 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), in collaboration with LaserLab Europe. FELs OF EUROPE has been founded in 2012 and will celebrate its 10th anniversary with a special event during the conference.

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 22-23 September 2022

For further information, please check the conference website: www.desy.de/scienceatfels2022

Elke Plönjes- Palm

Save the date: 5-7 October PhotonDiag 2022

The 6th FELs OF EUROPE workshop on FEL Photon Diagnostics, Instrumentation and Beamline Design, **PhotonDiag 2022**, will take place at the Helmholtz-Zentrum Berlin from 5-7 October 2022, following a virtual format in 2020 organized by the Paul Scherrer Institute.

The PhotonDiag workshop – hopefully in person this year - concentrates on new developments and advances in the field of photon diagnostics and FEL beamlines design, X-Ray-Optics, ranging from new devices for online measurements, to data processing and machine learning, new experimental challenges and future developments for next generation X-ray light sources. We would like to extend the invitation to, and enrich the audience with, representatives of the community at diffraction limited storage rings (DLSR), or multi-bend achromat (MBA) storage rings, who have similar tasks to fulfil and have comparable needs as the FEL community is facing now. Like at previous events we intend to publish a special issue in a peer-reviewed journal.

In a dedicated ceremony, FELs OF EUROPE will award a special prize on Photon Transport and Diagnostics.

The PhotonDiag 2022 will focus on the following topics:

- Temporal diagnostics
- Spectral measurements
- Coherence, profile, and position measurements
- Polarization measurements
- Scientific computing, machine learning and large data management
- Software and simulation
- New developments in photon diagnostics
- Developments in optics and optical systems
- Challenges in optics for high rep rate operations, damage and wavefront preservation
- New detectors for photon diagnostics
- Photon Diagnostics at synchrotrons
- Photon diagnostics requirements for new experimental methods

Frank Siewert

2022 FEL Conference

The International's Free-Electron Laser conference (FEL 2022) returns to the city of Trieste for its 40th edition, from 22 to 26 August 2022.

The conference is organized by Elettra-Sincrotrone Trieste S.c.p.A., an international, multidisciplinary research center of excellence, specialized in the generation of synchrotron and free-electron laser radiation together with their applications in material and life sciences.

FEL 2022 will focus on recent advances in free-electron laser theory and experiments; electron beam, photon beam and undulator technologies; and applications of free- electron lasers. The conference Scientific Program Committee, coordinated by Giovanni De Ninno and Simone Di Mitri is at work for the definition of a rich conference program, including sessions dedicated to FEL theory, SASE, seeded and oscillator FELs, electron sources, diagnostics timing and synchronization, photon beamlines instrumentation, undulator technology, and experiments using the light from FELs.

Trieste is a multi-cultural city, well renowned for its writers, artists and food. A city well-worth discovering, as did famously James Joyce and several other distinguished writers a century ago. Trieste is a maritime city that benefits from a nice Mediterranean climate, particularly pleasant at the end of August. Trieste is also an extremely safe city, where hotels, numerous excellent restaurants and cafes, monuments such as the Roman Amphitheatre, the Art Museum, the main railway station and shopping areas are within walking distance of one another.

This 40th FEL conference is following the 2019 FEL conference in Hamburg. It was originally planned in 2021 and postponed by one year, with the hope we could leave behind COVID-19 and organize an in-person event again. The health of participants will be our first priority. The conference will take place in the new Trieste Convention Center in the old harbor of the city, where 5,000 m2 exhibition area, open spaces and a conference room with 1800 seats will provide a safe workplace for the meeting. Further information concerning the access to the conference site, like vaccination rules, certification and the medical services provided will be communicated on the conference website.

Luca Giannessi and Michele Svandrlik
FERMI FEL

About FELs of Europe

FELs OF EUROPE is an initiative of the ESFRI projects EuroFEL and European XFEL. It is a collaboration of all free electron laser (FEL) facilities in Europe, with the goal to meet technological and scientific challenges of these novel and rapidly developing technologies, and to provide a worldwide unique, pan-European research infrastructure that enables exploiting the full scientific potential of these unique accelerator based short-pulse light sources. The collaboration includes 14 facilities in 10 countries.

All members are either operating or developing free electron laser (FEL) facilities and/or advanced short-pulse light sources (SPS), based on accelerator technologies. Due to their unique properties, these light sources provide a step change in the ability to address research needs across the disciplines of physics, chemistry, materials, and life sciences. FELs will improve our understanding of processes on a molecular level, leading to development of new materials and methods for tomorrow's technological advancement, clean environment, sustainable energy, and health care.

FELs OF EUROPE will facilitate the enhancement and exploitation of the full scientific potential of FELs in an efficient way by promoting joint technical development and collaborating closely with users and related communities. It will promote efficient open access to the research infrastructure and optimal conditions for users.

More info at: www.fels-of-europe.eu

CURRENT AND UPCOMING CALLS FOR PROPOSALS

www.fels-of-europe.eu/user_area/call_for_proposals

For experiments at FERMI
now open
For experiments at FELBE
Deadline: 12 April 2022
For experiments at FELIX
Deadline: 15 May 2022
For experiments at SwissFEL
Deadline: mid May 2022
For experiments at FLASH
Deadline: in summer

UPCOMING EVENTS

Webinar Series in 2022:
Follow the announcements on
https://www.fels-of-europe.eu/

FELs OF EUROPE Topical Workshop on Selected Problems in FEL Physics: from soft X-rays to THz at HZDR
25-27 April 2022

2022 FEL Conference, Trieste
22 - 26 August 2022

Science@FELs Conference at DESY
20-23 September 2022

PhotonDiag Conference at HZB
4-7 October 2022

Celebration of the 10th Anniversary FELs OF EUROPE in 2022

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