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Planned research activities

The research activities of ELI-ALPS have been selected using a variety of sources including the White Book of ELI-PP, the demand observed during several workshops, bilateral meetings and discussions with emphasis to the two users' workshops that ELI-ALPS has organized in November 2013 and September 2014 in Szeged, Hungary. ELI-HU Nkft. is especially indebted to the colleagues who contributed to the research program of ELI-ALPS:

Martin Aeschlimann, H. van Amerongen, Alexander Andreev, Patrizio Antici, Kunio Awazu, M. Barberio, Peter Baum, Mihály Benedict, Jens Biegert, Michael Bussmann, Francesca Calegari, Lorenz Cederbaum, Jean-Paul Chambaret, Dimitrios Charalambidis, John Collier, Eric Cormier, N. Cox, R. Croce, Aladár Czitrovszky, K. Gibasiewicz , Zsolt Divéki, Gagik Djotyan, Péter Dombi, Markus Drescher, Ralph Ernstorfer, Gyula Faigel, Alexei Fedorov, L. Fiedor, István Földes, József András Fülöp, G. Garab, Eleftherios Goulielmakis, G. Groma, János Hajdu, János Hebling, Katalin Hideghéty, Javier Honrubia, S. L. Johnson, M. Jones, Mikhail Kalashnikov, I Kézsmárki, Stefan Karsch, Ulf Kleineberg, Matthias Kling, J. Kern, Ferenc Krausz, Georg Korn, Gibasiewicz , P. H. Lambrev, Peter Lambropoulos, Raphael Levine, Victor Malka, Rodrigo López-Martens, Fernando Martin, Mattias Marklund, Stefan Mathias, Jürgen Meyer-ter-Vehn, A. Morabito, Gerard Mourou, David Neely, Mauro Nisoli, Zoltán Oláh, P. Ormos, Károly Osvay, Gerhard Paulus, Franz Pfeiffer, Luca Poletto, Fabien Quere, Predrag Ranitovic, Françoise Remacle, Th. Renger , B. Robert , Artem Rudenko, Giuseppe Sansone, M. Scisciò, A. Sharma, Emma Springate, Mark Stockman, Gábor Szabó, Sándor Szatmári, Toshi Tajima, Hideaki Takabe, Franz Tavella, John Tisch, J. Tobias, Martin Tolley, Csaba Tóth, George Tsakiris, Paris Tzallas, Mousumi Upadhyay Kahaly, L. Valkunas , Zoltan Varallyay, Katalin Varjú, Sándor Varró, László Veisz, S. Veltri, Marc Vrakking, Tibor Wittmann, Martin Wolf.

1. Research activities structure

1.1 Introduction

The frontiers of contemporary photonics are advancing in parallel with the characteristics of available photon sources. Today, synchrotrons and X-ray free-electron lasers offer angstrom wavelengths combined with high flux and brilliance, providing unique opportunities to explore the structure of matter with sub-atomic resolution all the way from crystalline solids, through nanoparticles to individual molecules. Laser-driven high-order harmonic sources, on the other hand, deliver flashes of extreme ultraviolet and soft-x-ray light with durations below hundred attoseconds, allowing direct time-domain insight into both structural and electronic motions, i.e. any dynamics taking place outside the atomic core.

The Attosecond Light Pulse Source – ALPS – of the Extreme Light Infrastructure – ELI – will combine these cutting-edge characteristics of modern photon sources, namely the short-wavelength and high flux of third-generation synchrotron sources with the incomparable pulse duration of laser-driven harmonic sources. Thanks to this combination of parameters never achieved before, ALPS' energetic attosecond X-ray pulses will have the dream of atomic, molecular and condensed-matter scientists come true: recording freeze-frame images of the dynamical electronic-structural behavior of complex systems, with attosecond-picometer resolution. This sub-atomic-resolution 4-dimensional (space-time) imaging affords a discovery potential that justifies the establishment of ELI-ALPS in its own right. At the same time the high XUV/x-ray pulse energies combined with the high repetition rate of the sources will allow for the first time XUV-pump-XUV-probe studies of attosecond dynamics in conjunction with product coincidence detection techniques i.e. the ideal tools for complete or close to complete experiments in all states of matter. However, ALPS will

have much more to offer. Its attosecond XUV/X-ray pulses will come synchronized with the controlled few-cycle to sub-cycle electromagnetic fields of intense laser light all the way from terahertz (far infrared) to petahertz (ultraviolet) frequencies. These ultrashort-pulsed, ultrastrong fields – locked to attosecond probes – open the prospect for unprecedented control of microscopic processes and provide a real-time look into an unsurpassed range of non-equilibrium states of matter. It is impossible to assess the impact and implications of the new insight into and control over microscopic processes ALPS will bring about. This summary profiting from the previous ELI documents[1] does not attempt to forecast what scientists are going to discover or the technologies that will be advanced (or newly created) with ALPS, rather, it aims at identifying its unique features and the resultant research opportunities for (a) exploring and controlling dynamics of the microcosm and (b) advancing/creating information, biological, and medical technologies.

1.2 The research structure

Based on these fundamental objectives, the concrete research activities can be broken down into eight distinct but complementary research areas- three addressing fundamental and five applied research- each of them representing completely novel research directions on their own. They have in common that the availability of unprecedented primary (laser) and secondary (attosecond, terahertz, particle etc.) source parameters will enable the community to pursue these research directions thereby creating a substantial demand for the facility beamtime in the potential user community worldwide.

The first two research activities encompassing a broad range of research topics relate to the development of primary and secondary photon and particle sources. The major research goal with respect to the primary sources is to reach the few-to-single cycle regime for all major laser systems, thus assigning a distinctive character to the facility under implementation. Research towards the development of the secondary sources includes the generation of ultrashort electromagnetic pulses in various spectral bands with the generation of energetic and high repetition rate attosecond pulses in the XUV to the X-ray domain being at the forefront of the development. Additionally to the photon sources the secondary sources research and development activities will target the development and installation of laser plasma based electron and ion acceleration sources. Even though the high pulse energy and high repetition rate laser sources support unprecedented secondary source parameters, several aspects of the underlying novel nonlinear light conversion processes have to be investigated, challenging an optimal performance of the sources. These investigations define a separate research direction on its own.

Equipped with such primary and secondary sources, ELI-ALPS will be able to tackle grand challenges in atomic, molecular and optical (AMO) physics by providing attosecond-duration excitation (pump) and probe pulses with unmatched fluxes. It is not only atomic and molecular systems that can be investigated with these tools but collective excitation dynamics in solids and surfaces, strongly correlated systems and plasma physics.

Having ultrahigh temporal resolution offers another significant potential, namely the combination of the attosecond "toolkit" with standard structural imaging techniques. The route towards and the project objectives in attosecond 4D imaging are described in the later Sections.

[1] Amiranoff et al., *Proposal for a European Extreme Light Infrastructure*, 2006; G. A. Mourou, G. Korn, W. Sandner, J. L. Collier (Eds.*), ELI – Extreme Light Infrastructure Science and Technology with ultra-intense lasers - Whitebook,* CNRS, Paris 2011.

In addition, the parallel existence of attosecond pulses and PW-class lasers within the same facility offer the time-resolved investigation possibility of relativistic light-matter interaction processes. The resulting novel plasma diagnostic tools will be of particular use for understanding laser-driven particle acceleration for medical applications or to better understand the approach towards inertial fusion energy, just to name a few important applications.

New research paradigms are also offered by other sources of the facility, as the interaction of intense Mid IR pulses (primary source) and THz fields (as unique secondary sources) with matter can be investigated. These additional activities complete the facility profile with synergic relationship to the main attoscience research lines.

The high repetition rate of the sources together with the specialized UHV end-stations provide an ideal environment for dynamical and structural studies in surface and condensed matter science.

The broad spectral coverage and ultrashort-pulse nature of the secondary sources will represent a set of compact, high-brilliance sources for more direct biological, medical, materials science and industrial applications. In summary, the most important links between these principal research directions are shown in the table below.

Table 1. Overview of the main research directions and their synergies to be pursued at the ELI-ALPS facility (GHHG: gas-target high harmonic generation, SHHG: solid-target high harmonic generation).

RA #	Research activity (RA)	Synergies with RAs	Secondary sources	Driving lasers
1	**Laser R&D**	2-4		10 HR, SYLOS, HF, Mid IR
2	**Secondary sources R&D**	1, 3, 4	GHHG, SHHG, THz, electron, ion	11 HR, SYLOS, HF, Mid IR
3	**Fundamental Research** ▪ ***AMO science*** ▪ ***Laser Plasma science*** ▪ ***Surface & condensed matter science***	1,2,4	GHHG, SHHG, THz, electron, ion	12 HR, SYLOS, Mid IR 13 SYLOS, HF HR, SYLOS, Mid IR
4	**Applied Research** ▪ ***Biomedical applications*** ▪ ***Medical applications*** ▪ ***Radiobiology*** ▪ ***Material science*** ▪ ***Manipulation of matter by intense THz fields*** ▪ ***Industrial applications***	1,2,3	GHHG, SHHG, THz, electron, ion	HR, SYLOS, Mid IR HR, SYLOS, Mid IR, HR, SYLOS, Mid IR, HFHR, SYLOS, Mid IR HR, SYLOS HR, SYLOS, Mid IR, HF HR, SYLOS, Mid IR, HF

Table 2: Overview of the major primary and secondary sources for each stages of implementation as well as for the foreseeable developments in the operation phase (to be up-dated)

	Primary sources						Secondary sources (Chapter 7.2.2 and 9.3)				
	Laser system	Peak /average power	Rep.rate	Pulse energy	Pulse duration	Spectral range	UV/XUV	X-ray	Ions	Electrons	THz
Stage 1 (commissioned by end 2016)	ALPS-HR1	> 0.16 TW / 100 W	100 kHz	1 mJ	< 6.2 fs (CEP stable)	0.7 - 1.4 µm	14-73 nm 17-90 eV/ 0.07-0.3 nJ GHHG		-	-	
	SYLOS1	4.5 TW / 45 W	1 kHz	45 mJ	< 10 fs (CEP stable)	0.5 - 1.3 µm	13-73 nm 17-100 eV/ 1 nJ - 1 µJ GHHG 31-154 nm 8-40 eV/ 1-10 µJ SHHG		-	-	FIR/THz: 0.3-3 THz, 100 µm-1 mm, 1.24-12.4 meV / 10 µJ, 3 MV/cm
Stage 2 (commissioned by end 2017)	ALPS-HR2	> 1 TW / 500 W	100 kHz	5 mJ	5 fs (CEP stable)	0.6 - 1.4 µm	14-73 nm 17-90 eV/ 0.35-1.5 nJ GHHG		-	-	
	SYLOS2	> 20 TW / 100 W	1 kHz	100 mJ	< 5 fs (CEP stable)	0.5 - 1.3 µm	13-73 nm 17-100 eV/ 5 nJ - 5 µJ GHHG 21-154 nm 8-60 eV/ 100-10 µJ SHHG	1-10keV, 1.2-12Å / 25µJ	-	50-100 MeV, 5-10 pC	FIR/THz: 0.3-3 THz, 100 µm-1 mm, 1.24-12.4 meV / >1 mJ, up to 100 MV/cm
	ALPS-MIR	> 25 GW / 15 W	100 kHz	0.15 mJ	< 4 cycles	2.2 - 3.9 µm					-
	ALPS-HF 100	> 50 TW / 50 W	100 Hz	0.5 J	< 10 fs	0.5 - 1 µm	(10 - 1000 eV, 120 - 1.2 nm, 50 µJ - 0.2 µJ)	1-10 keV, 1.2-12 Å / <0.05 µJ			
	ALPS-HF PW	> 2 PW / 340 W	10 Hz	34 J	17 fs	0.7 - 0.9 µm	(10 - 1000 eV, 120 - 1.2 nm, 1 mJ - 5 µJ)	1-10 keV, 1.2-12 Å / < 50 µJ	Protons: up to 160 MeV 1 nC	2 GeV, 0.2 nC	
Dev	ALPS-HR	> 2.5 TW / 1000 W	100 kHz	10 mJ	<4 fs (CEP stable)	0.5 - 1.5 µm	12-300 nm 4-100 eV / 100 - 10 nJ	3-12 nm, 100-400 eV >2nJ	-	-	MIR: 3-30 THz, 10-100 µm, 12.4-124 meV / 20-
	SYLOS	> 200 TW / 1000 W	1 kHz	1 J	< 5 fs (CEP stable)	0.5 - 1.3 µm	10 - 1000 eV, 120 - 1.2 nm, 100 µJ - 100 nJ	1-10 keV, 1.2-12 Å / < 500 µJ	-	100-200 MeV, 10-20 pC	T0 FIR/THz: 0.3-3 THz, 100 µm-1 mm, 1.24-12.4 meV / 10 mJ, up to 300 MV/cm
	ALPS-HF 100	> 50 TW / 50 W	100 Hz	0.5 J	< 10 fs	0.5 - 1 µm	10 - 1000 eV, 120 - 1.2 nm, 50 µJ - 0.2 µJ	1-10 keV, 1.2-12 Å / <0.05 µJ	Protons: up to 250 MeV 1 nC	2 GeV, 0.2 nC	-
	ALPS-HF PW	> 3 PW / 300 W	10 Hz	30 J	< 10 fs	0.5 - 1 µm	10 - 1000 eV, 120 - 1.2 nm, 100 µJ - 0.5 µJ	1-10 keV, 1.2-12 Å / <0.1 µJ			
	ALPS-MIR HE	> 1 TW / 100 W	10 kHz	10 mJ	< 3 cycles	3 - 8 µm					

2. Laser research and development

Research and development of the primary sources of ELI-ALPS will take place right from the beginning of the implementation phase. The necessary developments have been briefly listed in the technology chapters (Technological option analysis and the Description of technology). As it might become clear, the specifications of the major laser systems will represent optimum exploitation of material and immaterial resources adapting also to the user demand.

It is important to recognize that one of the main issues of keeping a future leading role of ELI-ALPS in attosecond science is to ensure the continuous upgrade of the driving lasers themselves. In this chapter we are dealing with laser research and developments, which are foreseeably necessary to be accomplished from in the operational phase. The results would be then gradually implemented on the laser systems, keeping in mind that the lasers should remain

- stable in specifications;
- reliable in operation;
- the time necessary for implementation of an upgrade should be minimized.

The planned and the envisaged parameters of the three major laser systems are shown in the below table. As it might be obvious from the research field of ELI-ALPS, the major goal is to shorten the pulse duration as much as possible, down to even less than one cycle, while the energy of the pulse is to be kept or even to be increased.

Table 3. Overview of the parameters of the main laser systems.

Laser	Specs to be delivered by 2018	Specs by Dec 2023
ALPS-HR	100kHz, 5 mJ, <6fs @ 1030nm (CEP)	100kHz, 10 mJ, <4fs @1030nm (CEP)
ALPS-SYLOS	1 kHz, 100 mJ, < 5 fs (CEP)	1 kHz, 1J, <3 fs (CEP)
ALPS MIR	100 kHz, 150 μJ, <60 fs @3.1μm (CEP)	10 kHz, 10 mJ, <60 fs @ 6-8 μm (CEP)
ALPS-HF PW	10 Hz, 34 J, 17 fs	10 Hz, 30J, <10fs
ALPS HF 100		100 Hz, 0.5J, <10 fs

For the first look the planned measure of pulse shortening might sound not that challenging. However, regarding that the bandwidth of the pulses is to be doubled, this would pose severe implications and issues in almost all elements in the laser chain, some of them are detailed below.

2.1 Development of broadband optical components

The ultimate issue of increasing the bandwidth of the laser systems is the availability of optical components offering appropriately large spectral bandwidth for transmission or reflection.

Mirrors

The complexity and hence the sensitivity of a multilayer coating system increases with increasing the reflection bandwidth. Moreover, such mirrors shall exhibit low dispersion, which further complicates the design. Mirrors designed for having special spectral phase shift (the so called chirped mirrors) with a bandwidth over an octave are the most sensitive to precision and control of various manufacturing steps and conditions, and usually have considerably lower laser damage threshold than low bandwidth components.

The mirror sets to be developed by the industry, partially with an initiative from ELI-ALPS, are expected to withstand the fluence of the ALPS-HR laser line while support the necessary bandwidth of an octave. Such mirror sets are to be further developed especially for having larger bandwidth to be used at the ALPS-Sylos beamline. Similarly, the coating to be used of ALPS-SYLOS available in 2016 can be used as a basis for upgrading the ALPS-HF system, under the condition of further R&D work on increase the damage threshold further. One of the major challenges is to scale the production of such, special broadband and high damage threshold mirrors for apertures of 30-40cm required by the HF laser.

A further challenge of broadband mirror developments is the high average power of the laser systems, especially for the HR and ALPS lasers. Since the mirrors are in vacuum, a minor absorption in the coating system may result in thermal origin deformation of the substrate, and might lead unwanted effects in further parts of the beamlines. Moreover, the planned operation condition of the facility will likely cause aging effects in the coatings and in the substrate.

MidIR beam propagation is an issue and requires specific optical material and coatings. We will launch dedicated R&D programs dedicated to the study and developments of specific materials and coatings with high efficiency and reliability at these wavelength.

Gratings

It is expected that broadband (~200nm), high damage threshold (>200mJ/cm^2) and very efficient (>95%) gratings will be available for the ALPS-HF system by 2015. To shorten the pulse duration on the target around 10 fs, the bandwidth of the gratings and hence the stretcher compressor system of the ALPS-HF system shall be increased to 300 nm at least. To overcome bandwidth limitation of diffraction gratings, which may happen once the bandwidth is increased substantially, efforts are worth devoting for development further stretching techniques based on combinations of prism /transmission grating-based stretchers with bulk material and chirped mirror recompression, as used for mJ level systems (shorter stretched pulse).

2.2 Development of pump lasers and amplification schemes

ALPS-HR

It is foreseeable that the level of pump laser energy required for this system will not be an issue by 2017 due to the high efficiency fiber amplifier technology which will be employed in the HR laser system. The available laser diode (LD) technology is advanced enough to pump the photonic crystal fiber (PCF) based large mode area (LMA) amplifier system which will be the core technology of the high energy pulse amplification scheme with a center wavelength of 1030nm.

In order to increase the wall-plug efficiency of the system, we intend to launch joint R&D programs to increase the efficiency of the main amplifier stages, e.g. on novel fiber design as well as on the optimal core composition of the LMA PCF to reduce the quantum defect heating. The double-clad pumping scheme can be further developed by designing such cladding structures, which make the pump absorption across the fiber core more homogeneous or may improve the delocalization of higher order modes (HOMs). This kind of effort may imply the development of high power LDs to make them more efficient, while their mode is also expected to improve via e.g. photonic crystal cladding in the distributed Bragg reflectors of LDs.

Recent fiber technology with nearly single mode operation and with a mode field area (MFA) >4000μm^2 allows us to amplify laser pulses well above 100 W average power. To increase the MFA further, it may introduce mode instability which limits the power scalability of chirped pulse, LMA

fiber amplifiers. Hence, further boost of power can be achieved only with an R&D program based on a mitigation strategy of the mode instabilities in the very large mode area fiber.

ALPS-Sylos

The direction of successful pump laser development will influence the development of future amplification schemes. Namely, the so called short pulse pumped (or ps pulse pumped) OPCPA design can be maintained under the condition that pump lasers are available having (near)infrared laser pulses with a few ps pulse duration and 10 J energy at a repetition rate of 1 kHz. Such 10 kW average power lasers emitting pulses of 10 TW peak power is a challenge itself for a decade. Judging from current pump laser developments (e.g. by Trumpf Scientific Lasers), it seems likely that Joule-class few-ps kHz pump lasers could be readily available in the next few years.

In order to support large bandwidth amplification, however, the performance of multiple colour / multiple beam pumping schemes has to be improved and / or other broadband amplification schemes has to be invented. The foreseeable increase of ps pulse energy makes it possible to test the feasibility of high-energy multi-colour OPCPA schemes so that one can expect to achieve single cycle operation without loss of energy with respect to current performance.

The current poor spectral quality of few-cycle pulses produced by OPCPA calls for the implementation of intermediate nonlinear temporal filtering stages inside the laser chain and thus multiple-CPA architectures. Despite an increase in the complexity of the system, this should facilitate dispersion management throughout the system and increase the efficiency of beam combining in the case of high-intensity multi-colour waveform synthesis. Indeed, achromatic temporal filtering techniques, such as XPW, seem perfectly adapted for achieving spectrally clean broadband pulses at different central wavelengths with smooth spectral phase over most of the bandwidth. The addition of nonlinear temporal filtering should also significantly enhance the temporal contrast of short-pulse OPCPA systems, rendering ultra-high intensity laser-solid interactions highly controllable and maybe even obliterating the need for high repetition rate plasma mirrors.

ALPS-HF

The primary aim of the PW laser development is to shorten the pulse duration below 10 fs. One of the most obvious ways is the replacement of the Ti:S duty end amplifier for a broadband OPCPA stage. An eventual upgrade of the system with further OPCPA stages requires the solution of the problem of temporal contrast degradation occurring in OPA stages pumped by nanosecond laser pulses. This needs to be done both theoretically and experimentally. To support the bandwidth required for shortening the pulse duration below 10 fs, already existing methods are to be explored at this high intensity regime, as two-color pumping, angularly dispersed or multi-beam pumped optical parametric amplification.

Despite that the pump lasers will use flash lamps, a possibility to substitute them in future by diode pumped modules at a lower level of the output pump energy, keeping the repetition rate at 10 Hz is feasible. Moreover, this change of technology may open the route to the increase of repetition rate further to 20 or even 50Hz. Such an increase of the repetition rate may require development of specific pump lasers based either on the technology of active mirrors, which will be implemented by the start of the facility at 10Hz, or by other technologies as multiple slabs for instance (see Mercury, DiPOLE and HiLASE).

A backup solution for the the pulse shortening, in case of the mentioned above problems with pump lasers and OPCPA cannot not be solved or afforded, is to accomplish further pulse shaping methods in the TiS amplifier chain. This may require the development of technologies towards efficient heat

removal (similar to those used in pump lasers) from Ti:S active medium along with new technologies for a broad band amplification in Ti:S.

ALPS-MIR

We foresee a rapidly increasing demand for a versatile but still robust MidIR laser. Hence MidIR laser development will be of high interest and we will dedicate it with high effort. One of the primary aims is to increase the energy of the MidIR laser pulses, so that keV photons can be generated with sufficient number to drive attosecond experiments in and beyond the water window. There are two major possibilities. The one is to add additional stages to increase the energy up to the ten mJ level with additional high average power ps pump lasers (400 W) able to deliver 40 mJ @ 10 kHz or 4 mJ at 100 kHz. The repetition rate tunability would be kept as an important issue for coincidence experiments. The second is that optical parametric amplifiers will be added pumped by existing lasers around 1 μm (the fiber laser system of HR delivers 3 mJ, 300 fs pulses per channel prior to compression at 100kHz, the SYLOS laser itself provides 100mJ pulses at 1kHz). This solution seems cost effective, but the trade in is the use of at least part of other laser systems of ELI-ALPS.

Parallel to the increase of the pulse energy, we will develop and implement technologies to reduce the pulse duration down to single optical cycle. In this way not only highly CEP sensitive experiments can be accomplished, but it would allow to generate true isolated attosecond pulses (IAP) in the water window. A simple way is the use of pulse compression, similar is already used in the HR laser, where the beam (signal or idler) is steered into a gas filled hollow core fiber or a Kagome fiber (preferred in our case). The pulse with increased spectrum is further compressed with dispersive devices. We also propose to extend such post compression techniques with other non-linear medium.

Finally, few cycle light pulses at longer wavelengths (4 to 10 μm) would attract high scientific interest and a wide range of experiments. To generate and amplify such pulses different types of non-linear crystals are needed, such as ZGP or AGS for instance. Unfortunately these crystals are opaque to 1 micron pump lasers. We may therefore contribute to the development of the technology of high average power, high energy pump lasers operating at 2 microns. This technology is based on CPA architecture with Ho-doped laser materials and will match phase II MIR requirements (high energy lower repetition rate).

THz-pump laser

To increase the efficiency of the THz pulse generation, we may explore the use of sub-ps, Joule-level pump pulses at longer wavelength (around 1.5 μm or even 2.1 μm). The latter could be accomplished with dedicated developments the aforementioned holmium lasers, while the former can be approached with the use of existing lasers of ELI-ALPS via OPA stages.

To increase the average power of the generated energetic THz pulses the repetition rate of the 1 J laser at 1 μm has to be increased by a factor of ten. Joule level kHz repetition rate lasers will be likely around in the next few years (see the SYLOS related developments) with a few ps pulse duration. Dedicated R&D would be necessary either to decrease the pulse duration of such lasers, or develop new lasers with sub-ps duration9.2.3 High-intensity waveform synthesis.

To generate single-cycle and even sub-cycle laser pulses, the already proven concept of waveform synthesis shall be further developed. At this moment the scheme works satisfactory for sub-mJ pulses and needs R&D to make it running reliable enough at facility level by 2015. Due to the sensitivity of the scheme and the difficulties arising from the increased fluence of the laser pulses, synthesis of the wave form of tens of mJ laser pulses may need further technological upgrade.

2.3 High-intensity waveform synthesis

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In the first phase of the HR system, 8 amplifier channels of 150 fs pulses while in the second phase, likely 16 amplifier channels will be combined coherently before the two stage HCF pulse compressor. Although it is theoretically possible to combine many more channels, but the damage of the HCF may prevent one from further scaling this architecture. A possible route would be to explore the pulse stacking methods together with coherent combination. This latter step could be also a form of wave form synthesis, as the generated pulses are likely close to the single cycle regime.

The pulse duration of the MidIR system can be also shortened with the synthesis of the electrical fields of the signal and idler waves. By upgrading the femtosecond system oscillator with a CEP stabilization, we will be able to mix the OPA signal and idler waves by coherent combination. This approach has the great advantage to recycle the idler energy (150 μJ as well) and double the output energy with an even shorter pulse, increasing the peak power by up to a factor of 5

2.4 Diagnostics

All the diagnostics equipments and devices has to be continuously upgraded and developed, in order to make them suitable to support the increasing bandwidth. Judging from current developments in the industry, it seems very likely that companies will bring to market devices capable of measuring near-single-cycle pulses on a single shot basis with several orders of magnitude dynamic range (e.g. d-scan or Wizzler-USP devices). The priority of ELI-ALPS is to drive the development of integrated diagnostic devices capable of fully optimizing the temporal pulse waveform through active feedback onto the laser operation itself.

A different class of challenges is set by the MidIR laser, as very few devices exist todays that are adapted or adaptable to the characterization of midIR beams. We plan to launch extensive studies aiming at providing us with the necessary tools for diagnosing our lasers in the midIR.

3 Secondary sources

The secondary sources of the facility will be driven by state-of-the-art ultrashort-pulse laser sources described previously. The parameter combination of each driver laser will facilitate novel parameter regimes in terms of the corresponding secondary sources. Each laser system will be connected to at least one secondary source development beamline and at least one research-oriented user beamline.

ALPS-HR will drive – via low-order (perturbative) nonlinear interactions – infrared beamlines, ALPS-HR-T0, ALPS-HR-T1 and ALPS-HR-T2, producing few-cycle to sub-cycle waveforms all the way from the near (~1.μm) to the far infrared (~1 mm). Together with their driver, ALPS-HR, they will allow to create any waveform over the terahertz-petahertz frequency band. Synchronized attosecond extreme ultraviolet (XUV) and soft-x-ray (SXR) pulses from ALPS-HR-P1 and ALPS-HR-P2 (via high-order (non-perturbative) processes such as atomic high-order harmonic generation), respectively, allow full characterization of these waveforms (via attosecond streaking).

Coming at multi-kHz repetition rates and in combination with synchronized attosecond XUV/SXR pulses ALPS-HR will open up new prospects in controlling electronic and structural dynamics in the microcosm.

ALPS-SYLOS will be used for creating ALPS' first-of-its-kind attosecond extreme-ultraviolet/soft-X-ray/hard-X-ray/diagnostic-X-ray (briefly X-ray) sources of never-before-achieved pulse energy and photon flux: ALPS-SYLOS-S1, ALPS-SYLOS-S2, and the incomparable attosecond hard-X-ray sources ALPS-SYLOS-H1. ALPS-SYLOS will open up three entirely new areas of applications. The "S-band" (S1 for XUV and S2 soft-X-ray) will allow for the first time attosecond-soft-X-ray-pump/attosecond-soft-X-ray-probe spectroscopy, which, will be extended to the hard-X-ray regime with ALPS-SYLOS-H1, bringing nonlinear attosecond-X-ray spectroscopy to fruition. The H-band source will also permit time-resolved X-ray diffraction with true attosecond time resolution for the first time. The pulse energies will be increased by two-three orders of magnitude, once atomic harmonic generators can be replaced by surface harmonic generators driven by relativistic fields. ALPS-SYLOS-H2 will be based on Thomson scattering of the ps-scale laser pulses, from the Yb:YAG pump source of ALPS-SYLOS, off few-femtosecond relativistic electron bunches accelerated by the > 100 TW pulses. ALPS-SYLOS-H2 will provide the first probe ever for time-resolving inner shell (including K-shell) dynamics of medium and high-Z elements. The kHz rate of these unprecedented attosecond pulses will permit the utilization of mature techniques such as attosecond streaking or non-linear autocorrelation for their characterization.

By the end of 2015 a downscaled secondary source performance is expected corresponding to the laser pulse parameters available at that time detailed in the primary sources chapter.

ALPS-HF is the most powerful laser system of ELI-ALPS. This feature will be exploited on 3 beamlines. Since SHHG scaling expectations promises ALPS-HF to be an efficient driver for such a source, it will serve as the basis of intense and high-energy X-ray emission on low repetition rate allowing X-ray spectroscopy and/or diffraction. Moreover, ALPS-HF will serve as the driver for high-energy electron and proton/ion acceleration. For the particle acceleration community this laser offers unique parameters, concerning its pulse duration and intensity. This These sources are practical tools for applications where compact, high repetition rate particle sources are needed in a specific energy band due to their small size, low cost and easy access. Furthermore, these beamlines with the high-energy charged particles will lead to faster development of radiobiology and tumor therapies, materials science etc.

3.1 Attosecond pulse generation

The duration of available ultrashort pulses limits the temporal resolution in the exploration of the dynamic behaviour of matter. Femtosecond laser pulses have been successfully used to study the dynamics of atoms and molecules involved in chemical reactions. Femtosecond laser pulses are also the key tools to produce attosecond pulses in a nonlinear interaction between the high intensity laser pulse and matter providing an ideal tool for studying electronic processes with short wavelength (i.e. high photon energy) and sub-femtosecond pulse duration. For a straightforward interpretation of spectroscopic data isolated (single) attosecond pulses are required. Furthermore, attosecond light pulses can be used to produce electron wave packets via one-photon ionization, with properties closely related to those of the optical wave packet of high kinetic energy and attosecond duration.

High-energy attosecond sources will extend the capability of attosecond science to probing valence and core electron dynamics and to four-dimensional imaging of the electronic structure of matter

with attosecond temporal and picometre spatial resolution. Brilliant laboratory-scale X-ray and electron sources hold promise for advancing biological, medical, and industrial applications.

We describe here briefly the three main methods for attosecond pulse production. There are further novel techniques proposed in the literature like parametric amplification of soft-X-ray radiation seeded with high-order-harmonic radiation generated in the same medium[2] or high-order harmonic generation in bulk crystals[3].

3.2 Attosecond pulse production via gas harmonics generation (GHHG)

When an intense femtosecond light pulse is focused into a gas medium, the electronic response is highly nonlinear and extremely high harmonics of the driving laser frequency can be generated[4]. High-order harmonic generation (HHG) in gas is a widely used technique for the production of coherent extreme ultraviolet (XUV) radiation[5]. The physical processes leading to the generation of XUV or soft-x-ray radiation by HHG can be understood using the so-called three step model[6]. In the framework of this semi-classical model, an electron exposed to an intense, linearly polarized electromagnetic field is emitted from the atom by tunnel ionization. The liberated electron can be driven back towards its parent ion by the external field and, with small probability, it recombines to the ground state thus emitting a photon with an energy equal to the sum of the ionization potential and the electron kinetic energy gained in the laser field. Such three-step process is periodically repeated every half optical cycle of the fundamental radiation, thus leading to a periodic emission of very short radiation bursts, with duration in the attosecond range, for each half cycle of the driving radiation. The periodicity in the temporal domain leads to a periodicity in the spectral domain, at twice the fundamental frequency.

Motivation, state of the art

In the last decade several methods have been experimentally implemented for the generation of high-energy attosecond pulses, based on the use of high-energy driving pulses focused in gas cells[7,8] or in hollow-core fibers[9,10]. In particular, in the case of trains of attosecond pulses, a very powerful scheme for the generation of microjoule level XUV radiation is the loose-focusing geometry, based on the use of high-energy driving pulses focused into long gas cells by long-focal-length lenses. Good phase-matching conditions can be obtained by compensating the Gouy geometric shift with the positive dispersion of the neutral medium at low pressure. The maximum value of the driving intensity is roughly determined by the saturation intensity of the gas used for HHG, defined as the peak intensity leading to an ionization probability of 1. Since the product of medium length and gas pressure is limited by self-absorption of the harmonic radiation, the XUV photon flux can be increased by increasing the spot area of the driving radiation at the interaction region, thus increasing the interaction volume. In order to maintain good phase-matching condition, gas pressure must be decreased in order to properly balance the reduced Gouy geometrical dispersion. In this way interaction length can be increased to maintain a constant pressure-length product. Loose-focusing geometry allows one to fulfil the optimizing conditions discussed above. XUV energies above 10 μJ have been achieved at wavelengths between 73.6 nm and 42.6 nm by using 35-fs

[2] J. Seres et al., Nat. Phys. 6, 455 (2010)
[3] S. Ghimire et al., Nat. Phys. 7, 138 (2010)
[4] A. L'Huillier and Ph. Balcou, Phys Rev Lett 70, 774 (1993).
[5] P. B. Corkum and F. Krausz, Nat. Phys. 3, 381 (2007), P. Agostini and L. F. DiMauro, Rep. Prog. Phys. 67, 813 (2004) and references therein.
[6] K. J. Schafer and K. C. Kulander, Phys. Rev. Lett. 70, 1599 (1993), P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).
[7] J.-F. Hergott et al., Phys. Rev. A 66, 021801(R) (2002).
[8] E. Takahashi et al., Phys. Rev. A 66, 021802 (2002).
[9] A. Rundquist et al., Science 280, 1412 (1998).
[10] R. Bartels et al., Nature 406, 164 (2000).

driving pulses, with an energy of 14 mJ, focused into a long cell (xenon) by a 5-m-focal length lens[11].

Research directions and activities

Quasi-phase-matching (QPM) schemes have been proposed and implemented in order to optimize the harmonic conversion efficiency by preventing back-conversion of the harmonic generation process by a periodic correction of the phase mismatch every coherence length. Hollow-core waveguides with modulated internal diameter have been employed to achieve a periodic modulation of the driving peak intensity, thus leading to a periodic phase modulation of the XUV field[12]. All-optical schemes have been also developed, which employ weak counter-propagating pulses[13]. Another QPM approach is based on the use of a multi gas cell target[14]: it has been demonstrated that it is possible to increase the conversion efficiency by using n consecutive gas cells placed in such a way that the generation process in consecutive cells sums up coherently. This can be obtained if the overall phase of the atomic dipole oscillations at the input of a particular cell is shifted by π with respect to the output of the preceding section. In the ideal case the atomic density can be increased by a factor n, thus resulting in a conversion efficiency increase by a factor n2. Such a multi gas cell target scheme might be particularly useful in the case of loose focusing geometry.

The generation of high-energy isolated attosecond pulses can be achieved by using various techniques. There are three main classes of generation schemes. The first method, called amplitude gating, is based on spectral selection of the cutoff region of the HHG radiation[15]. Such a generation scheme requires the use of intense sub-5-fs driving pulses, with controlled electric field: only in this case it is possible to generate a broadband and spectrally continuous radiation in the cutoff region. The second method, called temporal gating, is based on the generation of a short temporal window, where harmonic generation is confined. The very recent third scheme can be named as spatial gating, which is based on the attosecond lighthouse effect[16]. It produces a manifold of perfectly synchronized, tuneable attosecond pulses for performing true "attosecond + attosecond" pump-probe experiments via angular dispersion of the incident attosecond pulse train.

Three temporal gating schemes have been proposed and implemented: (i) polarization gating, based on the modulation of the polarization state of the driving field; sub-nanojoule isolated attosecond pulses have been produced. (ii) Ionization gating, based on the sub-cycle ionization dynamics in a gas cell excited by high-intensity few-optical-cycle pulses. Sub-160-as isolated pulses with energy up to 2 nJ were reported[17]. (iii) Two-color gating, employing an excitation field produced by the combination of on infrared pulse (with central wavelength in the range 1.3-1.8 μm) and a visible (typically 0.8-μm central wavelength) pulse[18,19]; in such a case, by implementing the loose-focusing geometry, isolated attosecond pulses with energy in the microjoule range might be generated, using TW laser systems.

The extension of temporal gating techniques to high-energy driving pulses with peak power up to the petawatt level is a very active research field. One of the main limiting factors is related to the depletion of the neutral atom population on the leading edge of the driving field. The generalized double-optical gating (GDOG) technique was implemented for the generation of isolated attosecond

[11] E. J. Takahashi et al., Opt. Lett. 27, 1920 (2002).
[12] E.A. Gibson et al., Science 302, 95 (2003).
[13] T. Popmintchev et al., Nature Phot. 4, 822 (2010).
[14] J. Seres et al., Nature Phys. 3, 878 (2007).
[15] M. Hentschel et al., Nature 414, 509 (2001).
[16] Vincenti et al., Phys. Rev. Lett. 2012
[17] F. Ferrari et al., Nature Phot. 4, 875 (2010).
[18] F. Calegari et al., Opt. Lett. 34, 3125 (2009).
[19] E.J. Takahashi et al., Phys. Rev. Lett. 104, 233901 (2010).

pulses by using sub-30-fs driving pulses[20]. By using the interferometric polarization gating technique[21] a continuous XUV emission was demonstrated by loosely focusing 55-fs driving pulse, with peak power of 2 TW, in a pulsed gas jet: the energy of the continuum, at photon energies in the range 30-70 eV was ~20 nJ[22].

One of the main scientific goals of the facility is to make widely accessible for external users a XUV-pump-XUV-probe approach for the investigation of time-resolved electronic dynamics in atoms and molecules. This approach should allow for a more straightforward interpretation of the experimental results with respect to the already implemented XUV-pump-IR-probe approach due to the strong field distortions imposed by the IR field on the atomic and molecular energy structure. The feasibility of this calls for the generation of attosecond pulses (trains and isolated pulses) characterized by a high energy and for the focalization of the XUV light down to a few micrometers in order to access a high intensity regime ($I>10^{14}$ W/cm^2) . Motivated by this main goal different research activities are currently ongoing within ELI-ALPS mostly focused on:a) the simulations of the characteristics of the gas high harmonics driven by high energy driving laser fields and b) the complete characterization of the focal spot of XUV pulses with sub-micrometer resolution.

a) Since the laser pulse characteristics that ELI ALPS' lasers will possess are not available for testing the proposed HHG arrangements, we rely on model calculations to forecast future XUV pulse parameters. We implemented some of the proposed configurations in a 3D non-adiabatic numerical model [1] V. Tosa et al., Phys. Rev. A 71, 063807 (2005);[2] V. Tosa et al., Phys. Rev. A 71, 063808 (2005)]. Due to the fact that for ELI-ALPS unprecedented laser parameters and focusing conditions are proposed, the code needed modifications to be able to handle the extreme conditions.

In particular the simulations were focused on the two different Operation Modes outlined in the Conceptual Design Report (CDR) for the SYLOS developmental beamline, with the first one optimized for the generation of isolated attosecond pulses by ionization gating, and the second for the achievement of trains of attosecond pulses with high photon flux in the 17-100 eV spectral range. In the following we present the main results obtained with the HHG conditions discussed in the CDR. A 20 mJ, 10 fs, 800 nm laser pulse, Gaussian in space and time with 60 mm diameter is focused with a 36 m focal length mirror. The gas cell is 1.4 m long filled with Ar (Ne, Xe) at 0.2 mbar pressure, and placed in the geometrical focus. According to the CDR depending on the target gas the goal is to obtain high flux XUV pulse in the spectral domains: 17-30 eV (Xe), 20-45 eV (Ar), 60-100 eV or 30-70 eV (Ne).

The three types of gases are characterized by different ionization potentials and third order susceptibilities, leading to very different levels of ionization at a given laser pulse intensity. First we illustrate the IR laser pulse distortion by plotting the IR electric fields and ionization rates at the entrance and exit of the interaction region.

[20] X. Feng, et al., Phys. Rev. Lett. 103, 183901 (2009).
[21] P. Tzallas et al., Nature Phys. 3, 846 (2007).
[22] E. Skantzakis et al., Opt. Lett. 34, 1732 (2009).

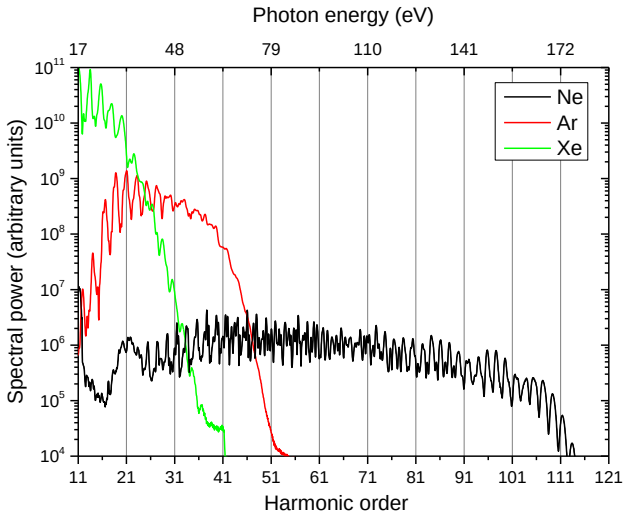


Fig. 1. Total harmonic spectra obtained in the three gas media: Ne, Ar, Xe at the end of the cells.

Simulations indicate that the proposed arrangements will provide radiation in the expected spectral ranges. The spectral power is expressed in arbitrary units. Although in the simulations SFA calculations are used and consequently the spectral power is obtained in arbitrary units, the relative harmonic yields obtained in the different generation media are comparable. Integrating the area below the spectra confirms the ratio of the yields proposed in the CDR. The calibration of the code to provide absolute units is under progress.

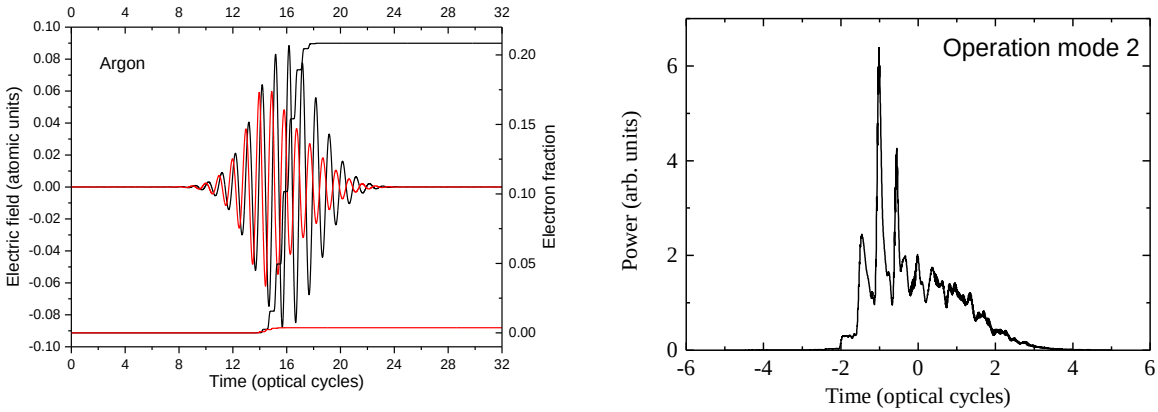


Fig. 2. (a) Initial (black) and final (at the exit of the HHG region, red) temporal shape of the IR laser pulse which interacts with Ar gas. (b) Temporal structure of the radiation in the far field, radially integrated.

Due to the very long focusing geometry, ionization level and pulse distortion is kept moderately low. (Fig. 2a) This is planned to assist phase-matching in the extremely long generation cell.
The simulations allow us to follow the formation of the harmonic field as propagation evolves, carrying information also about the macroscopic phase-matching conditions. Below we show the build-up of three selected harmonics for each generation medium in the targeted spectral range.

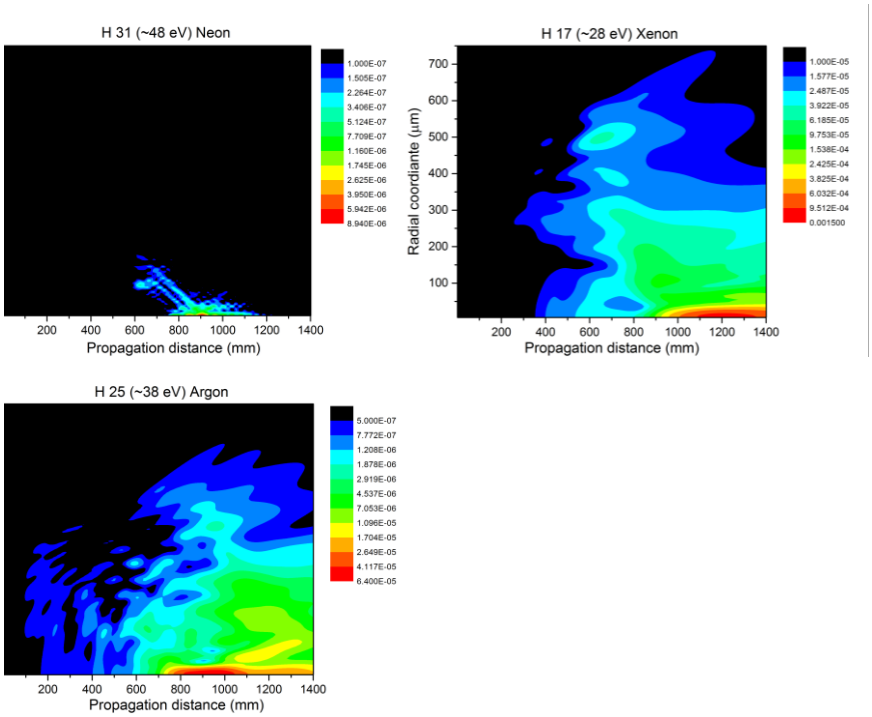


Fig. 3. (r,z) maps of harmonics as they build up in the three gas media. The color scale is logarithmic and spans 2.5 orders of magnitude.

As it is shown in Fig. 3, the macroscopic conditions support phase-matching on-axis, the maximum yield is achieved towards the end of the cell. In addition for some of the cases we also find favorable conditions off-axis; this latter circumstance contributes to the high yield in the radially integrated spectrum.

Modelling Operation Mode 1 in the CDR, is not a straightforward task because of the high intensities required for the proposed ionization gating method. This method requires the complete ionization of the gas medium at the rising edge of the generating laser pulse, allowing the generation only of a single attosecond pulse even by multi-cycle pulses. Compared to Operation mode 2, the generation of an isolated attosecond pulse in Operation mode 1 necessitates tighter focusing, a shorter gas cell and higher pressure medium. We used 10 mJ of the available energy in a 21 m focusing geometry, and a 3.2 cm long gas cell that is put in the focus, filled with argon at 2.6 mbar pressure. The milder focusing used in the simulation (compared to the one proposed in the CDR) is only required because of the limits of the simulation model, which was not designed for the high intensities these laser will be able to achieve. Nonetheless, as seen in Fig 4, already with this focusing geometry the signature of the ionization gating is seen in the field propagation, showing complete ionization of the medium at the rising edge of the pulse. We verify that stronger focusing and higher gas pressure should contribute to a reduced temporal profile of the emission.

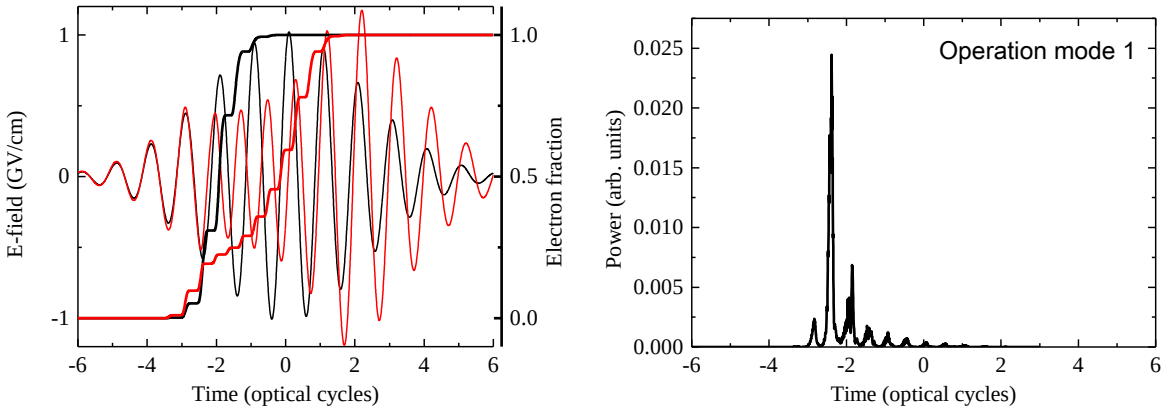


Fig 4: (a) Initial (black) and final (red) temporal shape of the laser electric fields in Argon and ionization fraction. (b) Temporal structure of the radiation in the far field, radially integrated.

b) The accurate measurement of the XUV diameter in the interaction point is a crucial information to determine the actual XUV intensity on target.

An approach with which the above information can be directly retrieved, is based on mapping the spatial XUV-intensity distribution into a spatial ion distribution, produced in the XUV focal area through 1-XUV-photon ionization of atoms. The image of the spatial ion distribution can monitored by means of an Time-Of-Flight Ion-Imaging-Detector (TOF I-ID)[23] [24]. A schematic of the TOF I-ID is shown in Fig. 1a. This device can image the XUV focus with spatial resolution in the sub-μm scale. Images of the spatial ion distribution produced in the XUV focal area through a linear ionization processes in atoms (Fig. 1b) have been recently recorded by means of an ≈ 1 μm spatial resolution in Attosecond Science and Technology (ATS) laboratory in FORTH-IESL. Images as this which is shown in Fig. 2 can be obtain in real time and can be used for the accurate measurement of the XUV intensity at any point along the XUV focus.

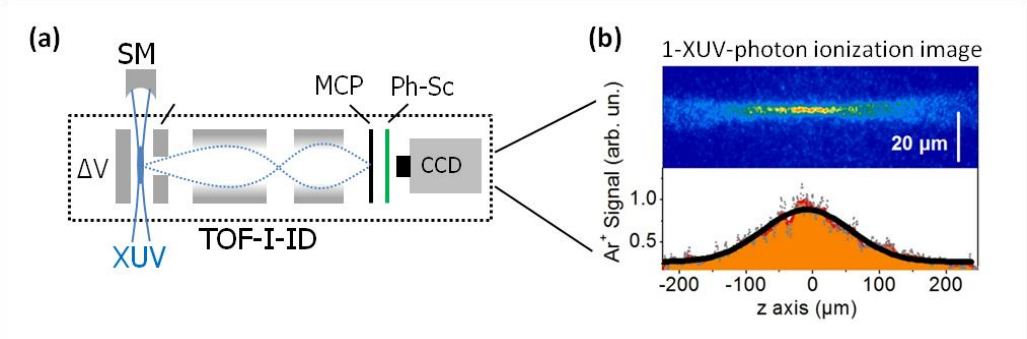


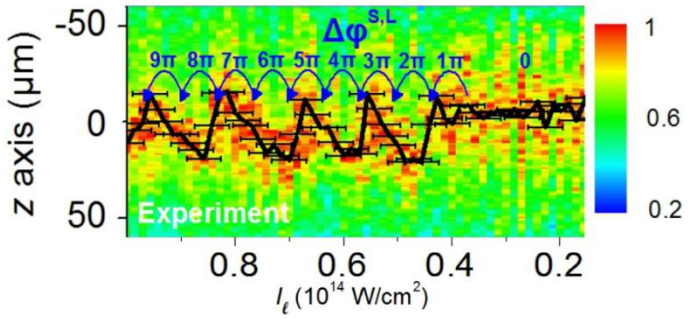
Figure 1. a) The time-of-flight imaging detector (TOF-I-ID) which is consisted by 1) the TOF acceleration region (ΔV) at which the XUV pulse is focused producing an ion distribution along the XUV propagation axis (z) and 2) a configuration of electrostatic lenses which is used for imaging the ion distribution created in the TOF acceleration region onto an MCP. The MCP is coupled with a phosphor screen (Ph-Sc) and the resulted image is recorded by a CCD camera. b) ion distribution produced in the XUV focal area through 1-XUV-photon ionization processes in Argon .

[23] Kaesdorf S Geräte für Forschung und Industrie, Gabelsbergerstr. 59 München, Germany

[24] 2 M. Schultze et al., New, J. Phys. 13, 033001 (2011)

The detailed knowledge of the XUV intensity distribution in the focus can provide significant information about the high order harmonic generation process and thus the attosecond pulse generation. In case that the XUV radiation is generated in gas phase media the properties of the electron wave packets which participate in the recollision process strongly correlate with those of the emitted XUV radiation. Thus, by spatially resolving the interference pattern generated by overlapping the harmonic radiation emitted by different interfering electron quantum paths, the intricacies associated with the recollision process can be unraveled. An example is given in Fig. 2 which shows the on-axis dependence of the XUV intensity distribution in the focus on the intensity of the driving IR pulse. This contour plot has been used in order to reveal the quantum path details of the recollision process[25] [26].

Figure 2. Imaging the intensity-dependent spatial XUV (harmonic 11th-15th) interference pattern. Contour plot



showing the dependence of the interference pattern along the *z* axis on intensity of the driving field. The black line depicts the mean value of the ion distribution.

Furthermore, XUV intensity distribution in the focus can be used towards the development of an single-shot 2nd order XUV autocorrelation [5 G. Kolliopoulos et al., J. Opt. Soc. Am. B 31, 926 (2014)]. This can significantly contribute in studies of ultrafast dynamics by providing in real time information about the instabilities (energy, CEP, pulse duration) and the spatiotemporal overlapping of the pump and probe pulses in a single or multicolor pump-probe experiment. Recently, has proposed a novel single-shot 2nd-order autocorrelation scheme for temporal characterization of the XUV radiation. In this scheme an TOF-I-ID is used in order to spatially resolve the pattern of the products of the second order process induced by two crossed beams of the radiation to be characterized. A schematic drawing of the scheme is shown in Fig. 3, while a detailed description of the approach can be found in ref. [5 G. Kolliopoulos et al., J. Opt. Soc. Am. B 31, 926 (2014)]. An important step towards this goal has been recently achieved in ATS laboratory by recording the image of the XUV focus resulting from the 2-EUV-photon-ionization of He.

[25] G. Kolliopoulos, et al., Phys.Rev. A 90, 013822 (2014)
[26] I. K. Kominis, et al., Phys. Rev. A 89, 063827 (2014)

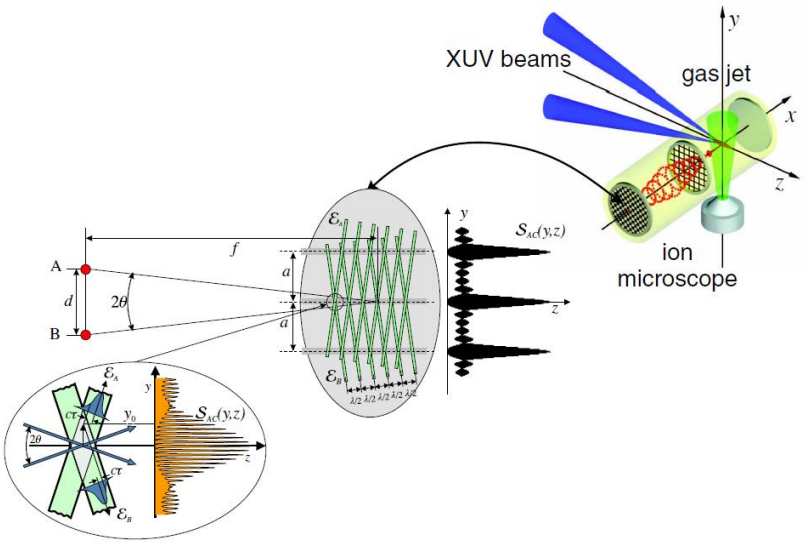


Figure 3. Concept of the proposed single-shot 2nd order autocorrelation scheme taken from ref. [5]. In the upper-right inset, a schematic of the proposed setup is shown. The imaging ion-microscope provides an image of the ionization products around the common focal spots of the intersecting XUV beams. In the lower-left inset, the analogy to single-shot AC using fs-laser pulses and crystals is indicated. Provided that a two-photon ionization occurs, the fringe pattern shown schematically is the second-order AC trace to be analyzed.

Concluding, the TOF I-ID is a device which can be used for the visualization in real time of the XUV spatial intensity distribution in the focus. This is done by mapping the ion distribution induced by the interaction of the XUV radiation with gas phase media into an image plane. The spatial resolution of an TOF I-ID can be in the sub-μm level. The on-line measurement of the XUV spatial intensity distribution in the focus can provide significant information about the spatiotemporal properties of the generated XUV radiation and at the same time can significantly contribute to the pump-probe studies and attosecond pulse characterization by means of an single-shot XUV autocorrelator.

Outlook, future implications

The main limitation of upscaling gas HHG is the ionization of the medium responsible for depletion and phase-mismatch. For noble gases this condition limits the applicable laser intensity to the range of 1014– 1015 W/cm2 (depending on the specific atom and the pulse duration of the driving field). Increasing the spot area will increase the photon yield. To maximize the harmonic photon yield, the objective is to use as high power laser as possible and the need to keep the intensity below a certain value force us to work in the loose focusing geometry.

Gas HHG source will be driven at ELI-ALPS by both the ALPS-HR and the ALPS-SYLOS lasers. In Stage 1, the pulse duration of both lasers will be longer than 5 fs, therefore we foresee the installation of one (or more) of the temporal gating techniques in order to produce isolated attosecond pulses.

3.3 Attosecond pulse production from plasma mirrors

When an intense ultrashort laser pulse strikes a solid target, it generates a dense plasma at the surface, whose expansion into vacuum remains very small compared to the laser wavelength. This sharp plasma-vacuum interface behaves as a high quality mirror for the incident laser field. At

ultrahigh laser intensities, the non-linear response of this mirror leads to a periodic temporal distortion of the laser field, resulting in the generation of high order harmonics of the incident frequency in the reflected field, associated in the time domain to attosecond pulses of light. One of the main strengths of attosecond sources based on plasma mirrors is that there is no known limit on the laser intensities at which they can be driven. They are thus ideal systems to exploit the highest light intensities that can be achieved by laser technology.

Motivation, state of the art

In the past few years, the different mechanisms leading to harmonic generation on plasma mirrors have been clearly identified, both in experiments and simulations[27]. Surface plasma harmonic emission has led so far to the highest attosecond pulse train energy of 40 mJ, which allowed the verification of sub-fs confinement through 2nd order XUV autocorrelation measurements[28]. Two main processes dominate: Coherent Wake Emission (CWE), which corresponds to light emission by collective electron oscillations in the dense part of the plasma mirror, and the Relativistic Oscillating Mirror (ROM) process, which corresponds to the Doppler effect induced on the reflected field by the laser-driven relativistic motion of the mirror surface.

ROM is the most promising mechanism for future attosecond sources based on plasma mirrors, since the maximum generated frequency grows with laser intensity, and has been predicted to eventually reach multi-keV photon energies and pulse durations down to the zeptosecond range[29] for intensities beyond $\approx 10^{21}$ W/cm2. Extremely efficient single attosecond pulse generation is predicted to occur when ultra-high intensity few-cycle pulses are confined to focal volumes on the order of the wavelength cubed[30].

At present, such multi-keV harmonic photon energies have been observed in one experiment with 500 fs laser pulses with the VULCAN laser in UK[31], at intensities of a few 1020 W/cm2. Such long driving pulses are clearly not adequate for attosecond science. With laser pulses from 20 to 60 fs from 10 TW scale lasers, the maximum observed harmonic order have so far remained below 40[32], for intensities slightly above 1019 W/cm2. The first major challenge for the coming years will be to demonstrate much higher orders with ultrashort lasers at high repetition rates. Recently, harmonic generation up to order ≈20 from plasma mirrors driven by few-cycle pulses has been demonstrated with repetition rates reaching up to 1 kHz[33]. Moreover, sub-cycle steering of overdense plasma dynamics has been very recently demonstrated with the use of mJ-level, CEP-controlled 5fs pulses at 1kHz[34], which offers exciting prospects for driving SHHG with the ALPS-SYLOS laser.

Research directions and activities

Plasma mirrors attosecond sources based on ROM are estimated to become competitive for laser intensities exceeding $\approx 10^{21}$ W/cm^2. There are two main challenges to drive such a source:

1- The first one is to reach such light intensities with few-cycle pulses. This is especially true for the production of single attosecond pulses, which will require almost single cycle laser pulses, unless clever temporal gating techniques suited to plasma mirrors are developed[35]. The "wavelength

[27] C. Thaury et al., Nature Phys. 3, 424 (2007).

[28] Y. Nomura *et al., Nature Phys.* **5,** 124 (2009)

[29] S. Gordienko et al., Phys. Rev. Lett. 93 115002 (2004).

[30] N.M. Naumova et al., Phys. Rev. Lett. 92 063902 (2004).

[31] B. Dromey et al., Nature Phys. 2, 456 (2006)., Phys. Rev. Lett. 99, 085001 (2007).

[32] B. Dromey et al., *Nature Phys.* **5**, 146 (2009).

[33] A. Borot et al., Optics Letters 36, 1461 (2011).

[34] A. Borot et al., Nature Phys. 12, (2012)

[35] T. Baeva, Phys. Rev. E, **74**, 046404 (2006)

cubed" approach is one possible method for obtaining isolated attosecond pulses with unprecedented peak-power. This is foreseen for Stage 2 of the project.

2- The second challenge is to keep the light pedestal and parasite pulses preceding the main laser pulse low enough as to not create a plasma on the target well before the arrival of the main pulse. This generally requires temporally "cleaning" the laser pulse before it interacts with the target by means of optical switching with plasma mirrors used at lower intensities. However there are already promising improvements to extend the harmonic orders up to 150 using low contrast lasers[36]. As a result of Stage 1 investment, these investigations can be carried out at the facility.

Harmonic generation from plasma mirrors can be driven up to 1kHz repetition rate using a high-speed rotational solid target holder to refresh the target between each laser shot while preserving identical interaction conditions. Such a device has been successfully used to drive harmonic generation from a plasma mirror at 1kHz repetition with mJ energy driving pulses. Scaling to J level pulse energies does not pose any major technical challenge since the target holding device can be made to accommodate larger target dimensions moving at higher speeds and optics close to the impact area can be shielded from debris using thin pellicles available in large sizes. These simple developments should make it possible to operate the plasma mirrors continuously, either as switches or as attosecond sources, at high repetition rates over extended periods of time.

Outlook, future implications

Future source characteristics depend on the generation conditions, and much work is still needed to determine the potential of this source, be it in terms of pulse energy or of quality (divergence, coherence) of the produced beams. In terms of conversion efficiency, the predictions of 1D PIC simulations[37] are summarized in Figure 17. Single attosecond pulses at longer wavelengths (20-70 eV, the most promising range for obtaining high-peak power attosecond pulses) can be generated with efficiency up to a few percent.

With respect to the figure below $aL < 10$ can be reached in Stage 1 for attosecond pulse trains. $aL > 10$ and isolated pulses concern Stage 2. Surface harmonic generation will be mainly driven by the ALPS-SYLOS system on the S1, S2 and H1 beamlines. The laser parameters given in Stage 1 are already promising to achieve new results in SHHG and to develop techniques that can pave the way for the research with the upgraded SYLOS system in Stage 2.

[36] invited talk of Hee Nam at LEI conference; Szeged, (2011)
[37] G.D. Tsakiris et al., New J. Phys. 8, 19 (2006).

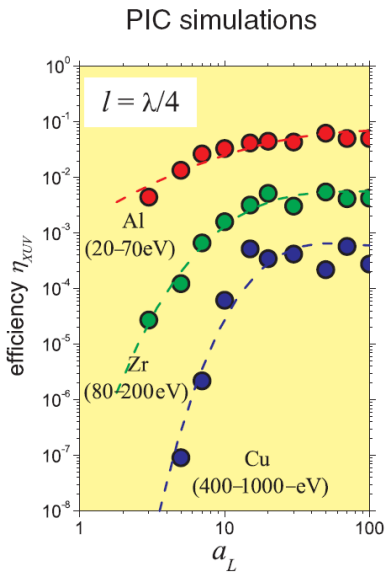


Figure 1. Variation of theXUV pulse efficiency ηXUV with the normalized vector potential aL for three different spectral ranges determined by the indicated thin filter used. From ref. [38]

3.4 Attosecond pulse production via Thomson scattering

Motivation, principle of operation

Dense electron sheets moving with high γ_x -factor ($\gamma_x = \left(1-\beta_x^2\right)^{-1/2}$, $\beta_x = v_x/c$ normal velocity) can be used as relativistic mirrors, compressing light pulses and up-shifting frequency by the relativistic Doppler factor $4\gamma_x^{\,2}$. With $\gamma_x = 16$, achievable at the ALPS-SYLOS beam line, one can compress optical few-femtosecond pulses to keV photon pulses of few attosecond duration. The required electron sheets can be generated from ultra-thin (nanometer) target foils (e.g. carbon foils) when irradiated by 10^{19} – 10^{20} W/cm^2 drive pulses with high contrast ratio (>10^{12}). The leading edge of such pulses ionizes the foil and pushes electrons out, completely separating electrons from ions. For this to happen, the driving laser field has to exceed the electrostatic field caused by charge separation. Simulations show that the electron sheet is surfing on the drive pulse keeping the initial solid density for a few light cycles before it decays due to expansion. This is long enough for reflecting a counter-propagating probe pulse. The goal is to achieve coherent Thomson backscattering. This project is at the border line between Stage 1 and 2. Only preparatory work can be carried out as a result of Stage 1 completion, This work is to be concluded in Stage 2.

Research directions and activities

A shortcoming of this scheme is that each electron acquires some transverse momentum $p_\perp = a \cdot mc$ in addition to the longitudinal momentum $p_x \propto a^2$; this degrades the electron's total $\gamma \propto a^2$ according to $\gamma_x = \gamma \Big/ \left(1+\left(p_\perp/mc\right)^2\right)^{1/2} \propto a$; here a is the normalized vector potential of the drive pulse. A way to suppress the transverse momentum is to place a reflector foil behind the source foil sufficiently thick to fully reflect the drive pulse, but to let the relativistic electrons of the sheet pass unperturbed. Simulations confirm that all electrons of the sheet emerge from the reflector with

[38] G.D. Tsakiris et al., New J. Phys. 8, 19 (2006).

$p_\perp = 0$, cruising with constant $\gamma_x = \gamma$ exactly in normal direction and ready for coherent backscattering with full Doppler factor $4\gamma^2$ [39].

The major challenge is to develop drive laser pulses with extremely sharp front edge and contrast ratio. This is to avoid premature destruction of the target foils which have to be much thinner than the skin depth and are transparent to the drive pulse, even though over-dense. Foil electrons are all taken along with the leading edge of the pulse so that the shortest laser pulses possible are just adequate for this purpose. Few-fs drive pulses with a = 3 – 5 are already available. Also carbon foils with thickness down to 1 nm have been fabricated recently and even mono-atomic carbon layers (graphene) which are very attractive. Of course, it needs to be studied how target systems for repetitive use can be made, in particular, the double layer version.

Outlook, future implications

The promise is an attosecond (and shorter) source of coherent (laser-like) keV-photons. The feasibility still needs to be demonstrated. It depends on the quality of the relativistic electron layer. Its reflectivity is expected to decay exponentially for scattered wavelengths shorter than the layer thickness. This limits photon energies to 1 – 10 keV. Higher photon energies can be obtained in the incoherent regime. For single layer targets, accelerating electrons have time-dependent $\gamma(t)$ producing scattered spectra chirped over several octaves. The duration of the scattered pulses will be set by the mirror decay time rather than the probe pulse duration in most cases. Anyhow, the observed spectra will provide detailed information about the mirror evolution. If successful, this method will become a superior tool for attosecond science.

ALPS-SYLOS-H2 beamline is dedicated to achieve these extra-energetic ultrashort pulses. Due to the strong requirements on laser parameters this technique is planned to be applied after the upgrade is carried out in Stage 2.

3.5 THz sources

Motivation, state of the art

The rapid development of THz sciences in the last two decades is mainly due to the availability of table-top sources based on compact solid-state femtosecond lasers. The other important aspect is that the time-dependent electric field of THz pulses can be directly measured by using electro-optic sampling.

Up to 1 μJ energy was demonstrated from large-area photoconductive switches illuminated by femtosecond laser pulses[40]. Single-cycle THz pulses with ~100 THz bandwidth and high field strengths were demonstrated by focusing a fundamental and second harmonic laser pulse together into ambient air[41]. Up to 5 μJ energy was demonstrated with 1×10-4 efficiency[42]. However, the further scalability of this technique is limited. Air plasmas can also be used for detection of THz pulses2. Optical rectification (OR) of intense femtosecond laser pulses offers a route to the generation of single-cycle THz pulses with extremely high intensity and field strength. Ultrashort THz pulses with 1.5 μJ energy have been generated by using ZnTe wafers with 75 mm diameter pumped at 800 nm[43]. THz pulses with 20 μJ energy were generated by OR of infrared pulses in the

[39] H.C. Wu et al., Phys. Rev. Lett. 104, 234801 (2010).
[40] R.R. Jones et al., Phys. Rev. Lett. 70, 1236 (1993).
[41] N. Karpowicz et al., Mod. Opt. 56, 1137 (2009).
[42] K.Y. Kim et al., Nature Photon. 2, 605 (2008).
[43] F. Blanchard et al., Opt. Express 15, 13213 (2007).

organic crystal DAST with conversion efficiency as high as 2%[44]. By using LiNbO3 (LN) pumped with fs pulses with tilted pulse fronts[45] (near) single-cycle THz pulses in the 1-THz frequency range with energies up to 50 μJ[46,47] and focused electric field strengths exceeding 1 MV/cm were demonstrated[48]. Applications requiring higher-frequency, mid-infrared (MIR) radiation can make use of DFG[49]- or OPA-based[50] sources delivering up to 100 MV/cm peak electric field strengths.

Applications requiring extremely strong THz fields are nonlinear THz spectroscopy, THz-assisted attosecond pulse generation, attosecond streaking with large time window, investigation of material properties and processes under the influence of extremely high quasi-static (THz) fields, multispectral single-shot imaging, manipulation and characterization of accelerated ultrashort electron bunches. Some of these applications require femtosecond synchronization of THz pulses to ultrashort pulses in other spectral ranges.

Research directions and activities

The most promising technique for generating ultrashort THz pulses with the required several tens to 100 MV/cm field strength in the few-THz range is OR with tilted-pulse-front pumping, a technique which is scalable to higher THz energies by increasing the pumped area and the pump energy. Calculations predict that this level can be achieved by using LN as the nonlinear crystal, optimal pump pulse duration of about 500 fs (rather than the commonly used ~100 fs pulses), cryogenic crystal temperatures (to suppress THz absorption) in combination with a large-area (~5 cm diameter) contact grating on the surface of LN.[51,52] Even by using longer than optimal pump pulses of 1.3 ps duration, 2.5× higher THz pulse energy (125 μJ) was measured with 5× higher pump-to-THz energy conversion efficiency (0.25%) than reported previously with shorter pulses[53] (Fig. 2). These results verify the advantage of longer pump pulses and support the expectation that mJ-level THz pulses will be available by cooling the crystal and using large pumped area. THz pulses with energies on the 10-mJ scale with pump-to-THz conversion efficiencies on the order of 10% are feasible when pumped with sub-Joule-class diode-pumped solid-state lasers.

[44] C. P. Hauri et al., Appl. Phys. Lett. 99, 161116 (2011).
[45] J. Hebling et al., Opt. Express 10, 1161 (2002).
[46] K.L. Yeh et al., Appl. Phys. Lett. 90, 171121 (2007).
[47] A.G. Stepanov et al.,, Appl. Phys. B 101, 11 (2010).
[48] H. Hirori et al., Appl. Phys. Lett. 98, 091106 (2011).
[49] A. Sell et al, Opt. Lett. 33, 2767 (2008).
[50] F. Junginger et al., Opt. Lett. 35, 2645 (2010).
[51] L. Pálfalvi et al., Appl. Phys. Lett. 92, 171107 (2008).
[52] J. A. Fülöp et al., Opt. Express 19, 15090 (2011).
[53] J. A. Fülöp et al., Opt. Lett. 37, 557 (2012).

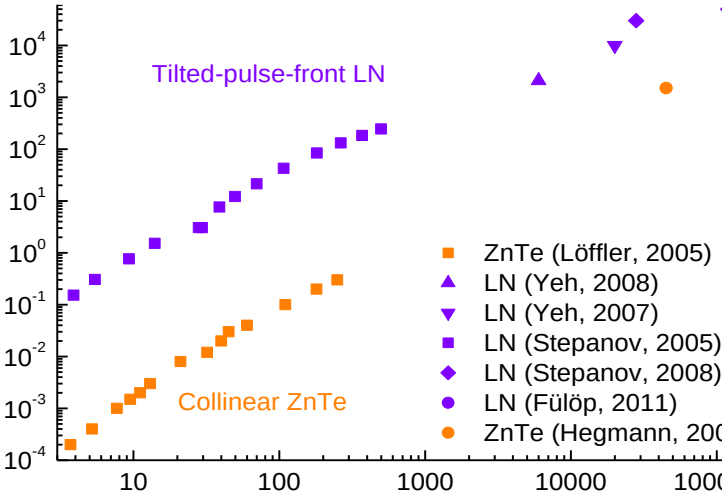


Figure 2. Experimental values of THz pulse energy vs. pump pulse energy for optical rectification.

The THz sources of ELI-ALPS are going to be driven by a dedicated driving laser, synchronized to the other primary sources. Such an optimized source can be, e.g., a diode-pumped Yb-based amplifier chain with ~500 fs pulse duration. Shorter pump pulses can be used with THz generation from air plasma. A MIR source based on difference-frequency generation (DFG) or OPA can be driven with a few-mJ, ~100 fs laser synchronized to the HR or the SYLOS primary sources.

Outlook, future implications

One key parameter determining the characteristics of the THz source is the repetition rate, which determines the available pump pulse energy. By driving OR with 1 J, 500 fs pump pulses THz pulses in the 1-THz frequency range with over 10 mJ energy and up to 100 MV/cm focused peak electric field strength can be expected. Assuming 1 kW pump power from an Yb-based amplifier, the available pump energy is 1 J, 100 mJ, and 10 mJ for 1 kHz, 10 kHz, and 100 kHz repetition rates, respectively. In the few-THz frequency range THz pulse energies (focused peak electric field strengths) on the 10 mJ (up to 100 MV/cm), 1 mJ (up to 30 MV/cm), and 0.1 mJ (up to 10 MV/cm) scale can be expected, respectively. For applications, where the frequency can be higher, DFG in GaSe[54] or THz generation from air plasma are possible alternatives. Former can provide 100 MV/cm peak electric field strengths around 30 THz. Especially the latter is compatible with very high repetition rates; also shorter pump pulses (20–30 fs) can be used.

Phasing

Stage 1: The main goal is to realize a dedicated pump laser for driving optical rectification. The target parameters of the diode-pumped solid-state pump laser are: 500 fs Fourier-limited pulse duration, 1 J pulse energy, and 100 Hz repetition rate. A THz pulse source in the 1-THz frequency range will be developed, which is scalable to the mJ energy level with good focusability. The targeted THz pulse parameters in Phase 1 are multi-100-μJ energy and 10-MV/cm-scale focused peak electric field strength in the 1-THz frequency range.

Stage 2: The driver laser will be upgraded to 1 kHz repetition rate allowing full-repetition-rate synchronization of the high-energy THz source to the SYLOS and the PW sources. The THz pulse energy will be boosted to the mJ-level, with focused peak values of the electric field up to 100 MV/cm in the 1-THz frequency range. Additional ultraintense THz sources are planned to cover neighbouring higher-frequency THz spectral ranges.

[54] A. Sell et al., Opt. Lett. 33, 2767 (2008).

4 Fundamental research activities

In this chapter the scientific areas in which ELI-ALPS envisages to perform outstanding fundamental research are detailed. The activities range from AMO physics to relativistic laser plasma interactions and surface/condensed matter science.

4.1 AMO science

The ELI-ALPS XUV/x-ray light sources allow the development of novel strategies for studying atomic and molecular dynamics with the potential for significant advancements in valence and core electron science. These novel strategies benefit from several of the unique characteristics of the ELI-ALPS light sources, namely the unique, attosecond and few-femtosecond, time structures of the pulses and the unique wavelength tunability from the far-infrared to the X-ray regime, which are both available combined with a high repetition rate.

Valence electron science

Motivation, state of the art

The attosecond and few-femtosecond time-structure of the ELI-ALPS light sources allows unprecedented access to the ultrafast electronic dynamics that constitutes the primary response of atomic and molecular systems to incident light. Previously, in atoms, time-resolved experiments have provided insight into the orbital motion of Rydberg electrons, while in molecules time-resolved "femtochemistry" experiments have revealed the evolution of nuclear motion, and the adiabatic/non-adiabatic adaptation of valence electronic states to the evolving structure of a molecule. With the ELI-ALPS facility the response of valence electrons to incident light can be measured and a purely electronic response can be induced in molecules, which can lead to ultrafast electron transfer processes in extended molecules on the attosecond or few-femtosecond timescale[55,56], which can be used to control chemical reactivity. In addition to these purely electronic responses, ELI-ALPS will allow to study the coupling of electronic and nuclear degrees of freedom that occurs during and after photo-absorption, as well as a range of intra- and inter-molecular electronic responses, the latter including the ubiquitous ICD (interatomic Coulombic decay) process that has recently been observed[57]. In doing so, the ELI-ALPS facility will allow attosecond science to venture into novel domains with a complexity that vastly exceeds that of any systems that have been studied so far, and that include attosecond plasmonics and attosecond surface chemistry. The end result of these efforts could be a mastery of the control over electronic processes to such a degree of sophistication that ultrafast electronic processes can be used to process information, thereby bringing to life the concept of "lightwave electronics".

The wavelength structure of the ELI-ALPS light source allows to develop completely novel strategies for probing valence dynamics that significantly go beyond the methods of femtochemistry, where photo-absorption by a pump laser pulse induces nuclear motion that is typically probed by monitoring the evolution of photo-absorption spectra (either directly, or indirectly as part of a high-order non-linear optical scheme). Although proven to be powerful in smaller systems, these methods depend in an undesirable way on ones' ability to know, calculate or infer the dependence of the absorption spectrum on the instantaneous molecular structure. The ELI-ALPS light sources allow to probe molecular structure without a prior knowledge of the electronic structure, using the ejection of photoelectrons that carry a de Broglie wavelength comparable to the internuclear distances that exist in a molecule, and inducing diffractive structures in the

[55] A.I. Kuleff et al., Journal of Chemical Physics 123, 044111 (2005).
[56] F. Remacle et al., Proceedings of the National Academy of Sciences of the United States of America 103, 6793 (2006).
[57] R. Santra et al., Phys Rev Lett 85, 4490 (Nov 20, 2000).

photoelectron angular distributions that are reminiscent of the structures in the photoionization efficiency measured in EXAFS experiments[58].

Research directions and activities

Valence electron science occupies itself – by definition – with electrons that are only bound by a moderate amount of energy (typ. 0-15 eV). From this it follows that the applications of ELI-ALPS to valence electron science will likely occur through one and two-color pump-probe experiments where an attosecond pump or probe laser pulse is combined with a probe or pump laser that can span the entire range from the vacuum-ultraviolet (VUV) to the far-infrared that is to be offered by the facility. In these experiments the achievable signal levels scale linearly with the number of available attosecond photons, and will often only be limited by this number, given that the accompanying transition in the (VUV→far-IR) range can often easily be saturated. It follows that the strength of the ELI-ALPS in studies of valence electron science is both the unprecedented pulse energy and the unprecedented pulse duration and tunability of the primary and secondary sources, and the fact that it will be possible to perform experiments at a very high repetition rate (100 kHz). The latter allows for the use of sophisticated coincidence detectors that provide a much deeper insight into the outcome of the experiment than lower-dimensional detectors that integrate a large number of events in a single measurement. In addition, the use of high repetition rates is important when space charge effects come into play, such as in surface experiments. Therefore, experiments on valence electron science and/or surface science will largely be carried out using the ALPS-HR sources.

Many of the experiments on valence electron science that will be performed at ELI-ALPS will require the use of isolated attosecond laser pulses. During Stage 1 of the facility development, it will be possible to generate these pulses using combinations of polarization gating and/or two-color harmonic generation methods. During Stage 2 of the facility development the enhanced pulse energy and reduced pulse duration of the high repetition rate lasers will greatly facilitate and increase the intensity of the isolated attosecond laser pulses that can be generated, thereby significantly extending the range of scientific problems that can be accessed.

In order to illustrate the possibilities that will thereby be offered, selected examples of recent and future work will now be presented.

Atomic valence electron science

To date, most of the successful attosecond pump-probe experiments making use of isolated attosecond laser pulses have been carried out on atoms. The main experimental technique, which was also used in the first-ever attosecond pump-probe experiment that determined Auger lifetimes of Kr, has been photoelectron streaking[59] In a photoelectron streaking experiment a velocity change of an ejected electron in a co-propagating IR laser field allows to determine the instance when the electron enters the laser field with a time-resolution that is a small fraction of the duration of the optical cycle of the IR field (2.7 fs). Using this technique, a very small (21 attosecond) delay between the ejection of Ne 2s and 2p electrons upon ionization by a 100 eV attosecond laser pulses was recently revealed[60] (see figure below). Alternative experimental schemes have been the use of a sub-cycle time dependence of strong-field ionization rates to measure electron dynamics in bound states of Xe and Ne atoms[61], the introduction of attosecond transient absorption in recent

[58] F. Krasniqi et al., Phys Rev A 81, 033411 (Mar, 2010).
[59] J. Itatani et al., Phys Rev Lett 88, 173903 (2002).
[60] M. Schultze et al., Science 328, 1658 (Jun 25, 2010).
[61] M. Uiberacker et al., Nature 446, 627 (2007).

experiments on strong-field ionization of Kr[62], and the recent use of wavepacket interferometry in two-color XUV+IR ionization of He[63].

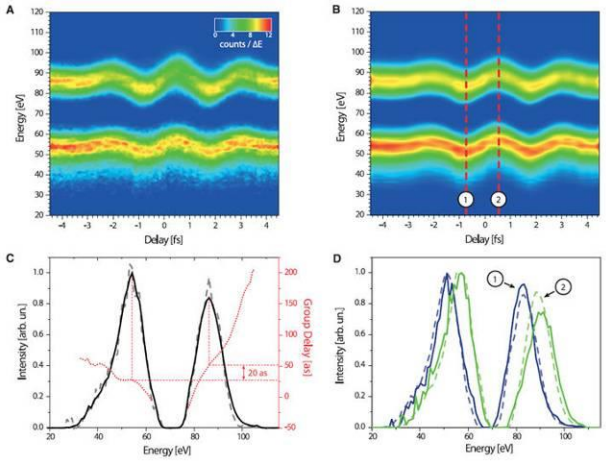


Figure 3. Delay between the emission of 2s and 2p electron in XUV photo-emission of Ne, measured by means of attosecond streaking

A message that has been learned from these early atomic attosecond pump-probe experiments, is that the information content of the experiments depends to a significant extent on the signal-to-noise that can be achieved. This is where the ELI-ALPS facility allows enormous progress to be made. Using ELI-ALPS-HF it will become possible to carry out these experiments at a hundred-fold increased repetition rate of 100 kHz, providing a key advantage that will allow the extraction of information on multi-electron correlations that has simply not been accessible in the experiments that have been performed up to now.

A major advancement in the study of valence electron dynamics to be offered by the high intensity 1kHz repetition rate attosecond beam-line that will be driven by the ELI-ALPS SYLOS laser chain is the access to attosecond resolution XUV-pump-XUV-probe, close to complete, experiments utilizing full momentum mapping techniques such as COLTRIMS. The way to this type of experiments was paved by the first prove of principle XUV-pump-XUV-probe study of

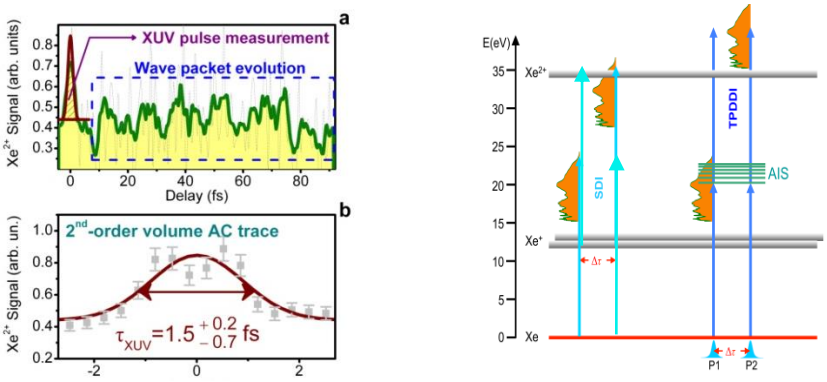


Figure 24. XUV-pump-XUV-probe of 1fs scale electron dynamics and the observation of 2-photon direct double ionization in Xenon.

[62] E. Goulielmakis et al., Nature 466, 739 (Aug 5, 2010).
[63] J. Mauritsson et al., Phys Rev Lett 105, 053001 (Jul 27, 2010).

1fs dynamics in atomic Xenon has been demonstrated in recent years[64]. In this experiment the dynamics of an atomic coherence excited above the ionization threshold (coherent superposition of a manifold of autoionizing states) through an XUV pulse has been probed by a second, time delayed, XUV pulse. At the same time, in this experiment a two-photon direct double ionization has been for the time demonstrated. Key parameters to this demonstration are the available high XUV intensity that allows the observation of two photon ionization and the short duration of the XUV pulse, a prerequisite for the direct process to become dominant against the sequential one [65].

Two-photon direct double ionization is a process of fundamental interest in atomic physics as it may involve electron-electron correlation effects. The problem has been extensively studied theoretically for the He atom. Here all time scales of different correlation effects are sub-fs to attosecond scales.Thus in He two-photon double ionization with 70eV photon energy the initial, i.e. the ground state, correlation decay time is calculated to be of the order of 20 attoseconds; the intermediate correlation, i.e. the induced polarization on He^+ by the outgoing first electron, decay time is of the order of 120 attoseconds, while the final correlation time, i.e. the electron-electron interaction time in the continuum is sub-fs.

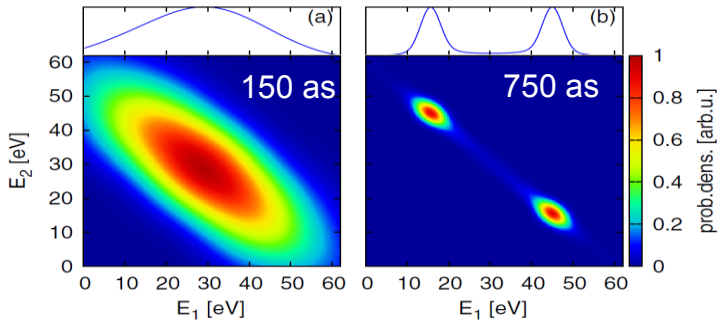


Figure 25. Electron-electron energy correlation contours in He double ionization at two different times of the ionization process[66]

Time resolved (with attosecond temporal resolution) correlated electron momenta mapping (e.g. with a reaction microscope) is the tool for the experimental investigation of such effects. The high intensity of the 1KHz repetition rate ELI-ALPS SYLOS driven attosecond beam line provides the required source parameters, which in combination with the planned reaction microscope end-station, opens up the venue towards the measurement of electron-electron correlation times in atomic valence electron processes.

Molecular valence electron science

In molecular science, the ELI-ALPS facility allows to provide original insights into the coupling of valence electrons to incident light as well as the coupling of valence electron excitation to nuclear dynamics. Molecules are largely unexplored in attosecond science. The first molecular application of attosecond pump-probe spectroscopy was only recently reported[67]. In the experiment, H_2 resp. D_2 molecules were ionized by an isolated attosecond laser pulse. The dissociation of the molecule was induced/influenced by a time-delayed few-cycle infrared laser pulse, that induced an electron localization in the dissociating H^{2+} resp D^{2+} that was measured through an asymmetric ejection of the H^+ resp. D^+ formed. The measured asymmetry, shown in below figure, depended with

[64] P. Tzallas et al. *Nature Physics* **7**, 781 (2011).

[65] P. Lambropoulos et al. *PR*A 78, 055402 (2008).

[66] Feist et al, PRL **103**, 063002 (2009).

[67] G. Sansone et al., Nature 465, 763 (Jun 10, 2010).

attosecond time-resolution on the delay between the isolated attosecond pulse and the IR pulse. Analysis revealed the presence of two electron localization mechanisms, namely one relying on the coupling of electronic and nuclear degrees of freedom and one relying on the coupling of multiple electronic degrees of freedom in the course of auto-ionization.

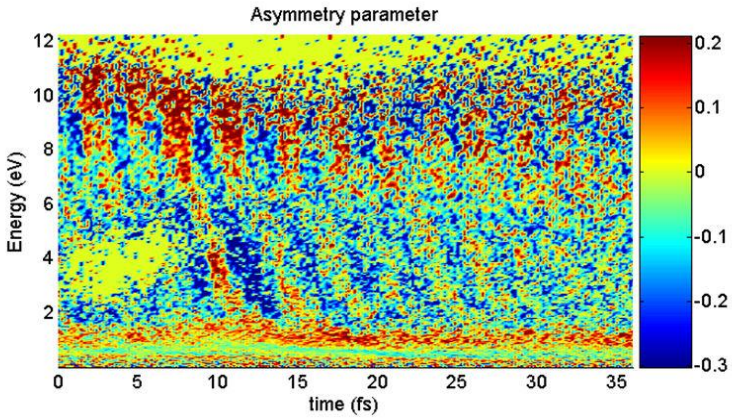


Figure 26. Asymmetry in the ejection of D^+ in two-color dissociative ionization of D_2, revealing the attosecond time dependence of two electron localization mechanisms that rely on coupling of electronic and nuclear degrees of freedom

Although capable of revealing first insights into attosecond time-scale electron dynamics, the experiment introduced above leaves a lot to be desired. The contour plot shown above is a testament to the only marginally sufficient attosecond photon flux that was available for inducing the ionization process. In addition, the use of 5-6 fs long IR pulse for probing the electron dynamics is highly undesirable, as it introduces an ambiguity in the time at which the observation is made, similar to the ambiguity that the use of attosecond pulse trains induces in pump-probe experiments. Using ELI-ALPS, major progress could be made on this problem. Using the ALPS-HR sources the IR interaction could be restricted to a single optical cycle and at 100 KHz multi-electron correlations could be investigated in a much more informative way, by recording coincidence between the primary electron that is formed upon photo-ionization and the ion recoil momentum (which indicates the localization of the second electron). Furthermore, using the wavelength tunability of the ALPS-HR source, the location along the reaction coordinate where the coupling between the electronic and nuclear degrees of freedom is laser-engineered can be chosen. Finally, using the ALPS-SYLOS source, attosecond pump-attosecond probe experiments come within reach, increasing the time-resolution by at least an order of magnitude.

A recent publication[68] highlighted the significance of XUV-pump-XUV-probe experiments in the study of molecular dynamics. In this work it is stressed using IR pulses that usually strongly perturb the molecular potential, modifying the inherent to the system dynamics. Thus access to the intrinsic dynamics of a molecular system requires pumping and probing of the system with gentle XUV pulses. The first proof of principle XUV-pump-XUV-probe experiment for the study of 1fs scaleelectronic, vibrational and ionization dynamics in molecules has been realized in H_2 [69] (see fig. 25).

[68] A. Palacios et al. PNAS 111, 3973 (2014).

[69] P. A. Carpeggiani, et al. *Phys. Rev.* **A89**, 023420 (2014).

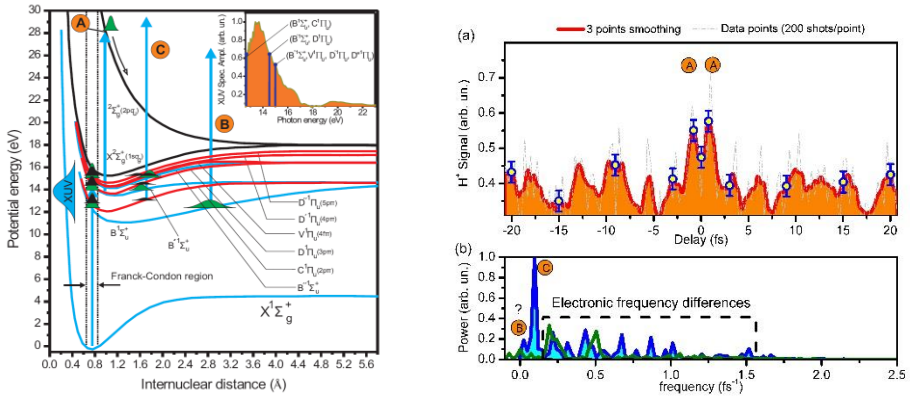


Figure27. XUV-pump-XUV-probe ionization-dissociation scheme in molecular Hydrogen and a measured XUV-pump-XUV-probe trace together with its Fourier Transform frequency spectrum.

More sophisticated experimental studies of this type including investigations of the interplay between electronic and nuclear motion, as well as the role of autoionization, as theoretically investigated by F. Martin et al. [70], [71] require ion-electron coincidence approaches and improved stability and statistics.

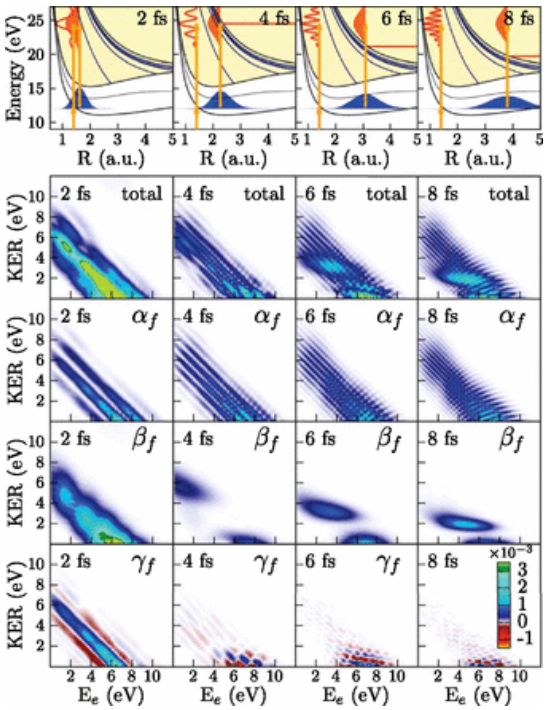


Figure 28. XUV-pump-XUV-probe dissociative ionization of H_2. Upper row: relevant potential energy curves, an illustration of the direct and sequential ionization processes, and the evolution of the different WPs at different time delays. Lower row: dissociative ionization probabilities as a function of electron energy (x axis) and nuclear kinetic energy (y axis) in eV. Each column corresponds to a given time delay between the pulses. [66])

The ELI-ALPS SYLOS driven attosecond beam-line will provide the necessary for such experiments XUV intensities at a repetition rate and stability that allows this type of approaches.

[70] A. Palacios et al. PNAS 111, 3973 (2014).

[71] Gonzalez-Castrillo, A., *et al. Phys. Rev. Lett.* **108**, 063009 (2012).

At the same type the foreseen reaction microscope user end-stations at ELI-ALPS is the most appropriate instrumentation for such coincidence studies.

Traditionally, chemical processes are described using the Born-Oppenheimer approximation, which holds that electronic states (orbitals, wavefunctions) adiabatically follow the time-evolving structure of a molecule.. In such a picture, a breakdown of the Born-Oppenheimer approximation occurs at curve crossings, where electronic and nuclear timescales become comparable to each other. The experiment in 10 can be viewed as a first example where electron dynamics has been observed in what some have called a "post Born-Oppenheimer regime"[72]. Upon instantaneous (attosecond) photo-excitation, a regime can be reached where time-dependent electronic motion is enabled, without any participation of the nuclei or preceding this participation. Under these conditions the electron motion can control the nuclear motion, setting the state for so-called "charge-directed reactivity", a completely novel paradigm in chemistry. The ELI-ALPS facility is ideally suited for studies of charge-directed reactivity. The huge flexibility in the choice of the parameters of the attosecond and primary HR-sources allows the design of experiments that can explore the existing predictions for attosecond to few-femtosecond time-scale electron transfer in extended systems upon attosecond photo-excitation/ionization. Furthermore, the ability to synthesize pulse shapes of almost arbitrary complexity allows to explore the control of electron transfer and thereby the outcome of a chemical reaction realizing a long-standing goal in molecular photo-chemistry.

Attosecond spectroscopy of complex molecules

Dynamical processes in molecules occur on an ultrafast temporal scale, ranging from picoseconds to femtoseconds when concerned with a structural change, down to attoseconds when dealing with electrons. As the charge density plays a very important role in bond-formation and bond-breakage, one can assert that the final chemical reactivity of a molecule is strongly influenced by the electron dynamics. At the most basic level, we can think at photo-induced chemical reactions as the result of interactions between electrons that ultimately drive the atomic rearrangement.

When the electronic motion is mediated by the nuclear motion, a process named *charge transfer* can take place. This process is at the basis of a number of important physical and chemical processes such as photosynthesis, cellular respiration and electron transport along large peptides and proteins[73,74]. The study of this dynamics belongs to the realm of femtochemistry, a well-established field that for more than twenty years has been able to capture and even control the nuclear motion in chemical reactions and intramolecular processes[75]. On the other hand, the very initial step of a chemical reaction, i.e. the electronic rearrangement, remains to be fully understood. In this contest, an important question still needs to be addressed *"is there an electron time scale in chemistry?"*. This question has triggered a new class of experiments that can be envisioned for the ELI-ALPS facility, taking the advantage of the extremely high temporal resolution that will be provided by the ALPS secondary sources.

Recently, it has been theoretically predicted that the sudden removal of an electron from a molecular orbital can give rise to charge oscillations from one end of the molecule to the other and back. This process has been named *charge migration* (CM) to distinguish it from charge transfer

[72] B.H. Muskatel et al., Physica Scripta 80, 048101 (Oct, 2009).

[73] H. Gray, J. Winkler, "Electron tunneling through proteins," Q. Rev. Biophys., vol. 36, pp. 341-372, 2003.

[74] J. R. Winkler, H. B. Gray, T. R. Prytkova, I. V. Kurnikov, D. N. Beratan, "Electron transfer through proteins," in Bioelectronics, I. Willner, E. Katz, Eds. Weinheim: Wiley-VCH Verlag, 2005, pp. 15-33.

[75] A. H. Zewail, "Femtochemistry: Atomic-Scale Dynamics of the Chemical Bond," J. Phys. Chem. A, vol. 104, no. 24, pp. 5660-5694, 2000.

mediated by nuclear motion[76,77]. Since this is a purely electronic process, it is expected to occur on a time scale ranging from a few femtoseconds down to hundreds attoseconds. For example, the positive charge created by prompt inner-valence ionization of the C-terminally methylamidated dipeptide Gly–Gly–NH–CH_3, initially localized on the methylamine end, is expected to migrate to the remote glycine in about 5-6 fs (Fig. 1)[78].

Attosecond technology provides an ideal tool for studying this ultrafast process[79]. Pump-probe experiments on molecules, exploiting the combination of XUV attosecond pulses to promote the electronic wave packet in an excited state of the molecular cation and IR femtosecond pulses to probe the subsequent dynamics, have been mostly performed on simple diatomic systems such as H_2, D_2[80,81], N_2 [82] and O_2[83], while the investigation of more complex molecules by attosecond pulses is only in its infancy.

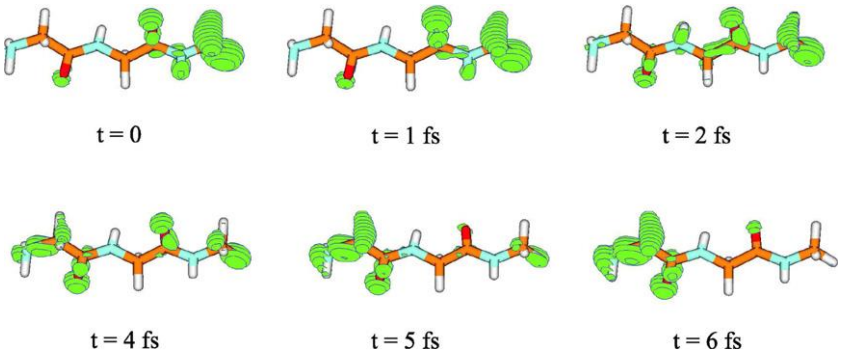


Figure29 Charge migration occurring in the C-terminally methylamidated dipeptide Gly–Gly–NH–CH_3 following inner-valence ionization. Reproduced from Ref. [74].

Recently, the first extension of attosecond spectroscopy to a biologically relevant molecule, i.e. the amino acid phenylalanine, has been reported[84]. In this experiment, charge dynamics was initiated in phenylalanine by isolated sub-300-as pulses, with photon energy in the spectral range between 15 eV and 35 eV and it was probed by 4-fs, waveform-controlled IR pulses, with central wavelength of 720 nm (see left panel of Fig. 2).A clean plume of isolated and neutral molecules of phenylalanine was generated by evaporation of the amino acid from a thin metallic foil heated by a CW diode laser. The parent and fragment ions produced by the interaction of the molecules with the pump and probe pulses were then collected by a linear time-of-flight device for mass analysis, where the metallic foil was integrated into one of the end electrodes. The temporal evolution of the doubly-charged immonium ion yield evidenced the presence of ultrafast oscillations (right panel of Fig. 2).

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[76] L. S. Cederbaum, J. Zobeley, ,Chem. Phys. Lett., vol. 307, no. 3-4, pp. 205-210, 1999.

[77] F. Remacle, R. D. Levine, PNAS, vol. 103, no. 18, pp. 6793-6798, 2006.

[78] A. I. Kuleff, S. Lünnemann, L. S. Cederbaum, Chem. Phys., vol. 414, pp. 100-105, 2013.

[79] F. Krausz, M. Ivanov, Rev. Mod. Phys., vol. 81, no. 1, pp. 163-234, 2009.

[80] G. Sansone et alNature, vol. 465, no. 7299, pp. 763-766, 2010.

[81] F. Kelkensberg et al., Phys. Rev. Lett., vol. 107, no. 4, p. 043002, 2011.

[82] W. Siu et al., Phys. Rev. A, vol. 84, no. 6, pp. 063412, 2011.

[83] M. Lucchini et al. Phys. Rev. A, vol. 86, pp. 043404, 2012.

[84] F. Calegari et al., Science, vol. 346, no. 6207, pp. 336-339, Oct. 2014.

The main periodicity of 4.5-fs has been assigned to charge migration from the amino to the carboxyl group of the molecule.

From this pioneering experiment, it has been learned that these measurements require an extremely high signal-to-noise ratio to extract significant information. Due to the low density of the biomolecular target (typically a few 10^9 cm^{-3}), in order to achieve sufficient statistics, a high-repetition rate source is desired. This is exactly what will be provided by the ALPS-HR secondary source; this beam line operating at 100 kHz will allow for a tremendous progress, providing the possibility to perform ion/electron coincidence measurements that have not been accessible until now. Another significant advance will be achieved by using isolated attosecond pulses also in the probing step. Indeed, due to the limited photon flux provided by the conventional XUV sources, the typical configuration adopted in an attosecond experiment is based on the combination of an XUV attosecond pump pulse and an IR femtosecond probe pulse. By using the ALPS-SYLOS source, it will be possible to perform attosecond pump - attosecond probe experiments, allowing to follow the pure electron dynamics with unprecedented temporal resolution.

The ELI-ALPS facility will open new frontiers in attosecond science with the possibility to investigate complex systems, far beyond the systems studied so far. New horizons can be foreseen for photochemistry and photobiology, that in which few-femtosecond and even sub-femtosecond electron processes determine the fate of biomolecules. The investigation of the pure electron dynamics in complex molecules will also open new perspectives for the development of new technologies, for example in molecular electronics, where electron processes on an ultrafast temporal scale are essential to trigger and control the electron current on the scale of the molecule.

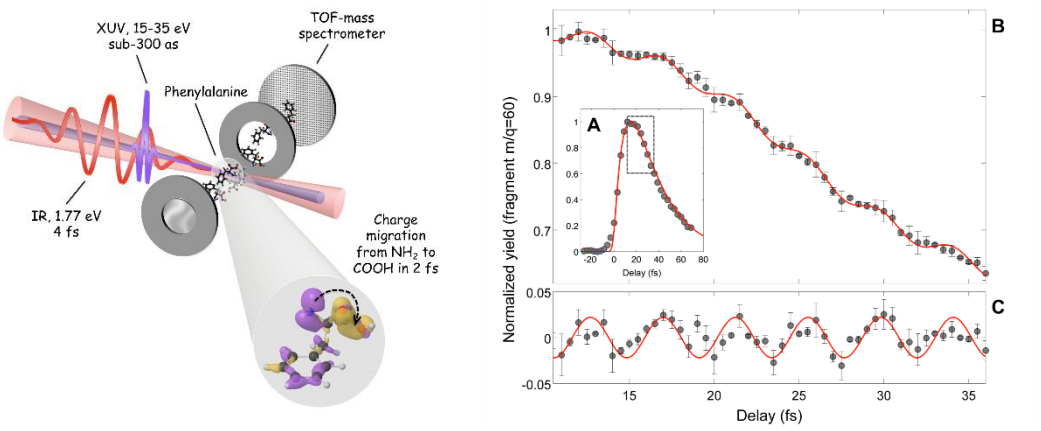


Figure 30 Left panel: scheme of the XUV pump IR probe experiment performed on phenylalanine molecules. Right panels: time dependent doubly charged immonium yield (a) and zoom over the 10-35 fs time delay range (b). The oscillatory pattern left after subtraction of the slow dynamics is reported in panel (c). Reproduced from Ref. [12].

THz radiation driven valence and free electrons

ELI-ALPS will offer unique sources of highest-intensity pulsed THz radiation, precisely synchronized to the main laser sources. Relying to world-leading in-house expertise, high-intensity ultrashort THz sources with unprecedented peak electric field strength up to 10 MV/cm and pulse energy exceeding 1 mJ will be available in the low-frequency (0.1–2 THz) part of the THz spectrum.. Advanced pump laser technology will enable later developments and the implementation of THz sources in neighbouring higher-frequency spectral ranges possibly extending up to 100 THz.

This infrastructure will enable new types of (multi-colour pump-probe) spectroscopic studies and the control of matter (charged particles, molecules, nanostructures, and condensed matter) by intense THz fields. (see chapter 9.5), Combined with the various other sources available at ELI-ALPS from x-ray to infrared wavelengths and their ultrashort pulse durations will enable an unprecedented variety of structural and dynamical studies. The envisioned main application areas of intense THz pulses at ELI-ALPS are summarized in the table below.

*Table 4. Here the highly promising applications of THz radiation in THz-assisted attosecond pulse generation, manipulation and characterization of relativistic electron beams and post-acceleration of ion beams using THz radiation for hadron therapy are elaborated.**Hiba! A hivatkozási forrás nem található.*The remaining part of this Section gives a brief description of each application area.

- THz-assisted attosecond pulse generation

A recently proposed scheme for attosecond pulse generation relies on HHG in presence of a strong dc or quasi-static electric field[85,86]. The applied field increases the asymmetry between subsequent half optical cycles of a few-cycle laser pulse thereby enables the generation of higher-order harmonics, and as a consequence, attosecond pulse shortening. With this method only one attosecond pulse is generated within a single oscillation cycle of the laser. For a pronounced effect the electric field of the applied quasi-static field has to be on the order of several tens of MV/cm. Quasi-phase-matching[87] is also achievable with the help of THz radiation, where the intensity of HHG in the cut-off region[88] is predicted to be increased by several orders of magnitude. A few MV/cm peak electric field strength is predicted to be sufficient in this case. Experimental realization of THz-assisted attosecond pulse generation will require a dedicated attosecond source with a synchronized high-intensity THz source, possibly with controllable chirp.

- Manipulation and characterization of relativistic electron beams using THz pulses

Manipulation of electron beams is important for many exciting applications, including ultrafast electron diffraction and the x-ray free-electron laser (FEL)[89]. There are implementations and proposals to use microwave or optical radiation for electron acceleration, undulation, deflection, and spatial as well as temporal focusing[90,91]. Using high-energy laser radiation with micrometer-scale wavelength for undulation[92,93] could result in FEL radiation on the Ångström range using electrons with only a few tens of MeV energy. A further advantage of the laser undulator is a significantly shorter gain length. Laser acceleration of electrons in a dielectric microstructure was demonstrated with acceleration gradients exceeding 250 MeV/m[94].

Using terahertz (THz) radiation in such applications can be superior to microwave or optical radiation since THz pulses can be generated with significantly smaller temporal jitter to the electron bunch than microwave pulses, and contrary to the optical pulses, the much longer wavelength of THz pulses fits well to typical sizes of electron bunches[95]. Focused THz field strengths on the order

[85] W. Hong et al., Opt. Express 17, 5139 (2009).

[86] E. Balogh et al., Phys. Rev. A 84, 023806 (2011).

[87] C. Serrat, and J. Biegert, Phys. Rev. Lett. 104, 073901 (2010).

[88] K. Kovács et al., Phys. Rev. Lett. 108, 193903 (2012).

[89] P. Emma et al., Nature Photonics 4, 641 (2010).

[90] T. Plettner et al., Phys. Rev. ST Accel. Beams 9, 111301 (2006).

[91] T. Plettner at al., Phys. Rev. ST Accel. Beams 12, 101302 (2009).

[92] P. Dobiasch et al., IEEE J. Quantum Electron. QE-19, 1812 (1983).

[93] T. Plettner and R. L. Byer, Phys. Rev. ST Accel. Beams 11, 030704 (2008).

[94] E. A. Peralta et al., Nature 503, 91 (2013).

[95] J. Hebling et al., arXiv:1109.6852 (2011).

of 10 MV/cm are feasible from sources operating in the 0.1–1 THz frequency range. At higher (1–5 THz) frequencies 40 MV/cm was demonstrated from organic crystals[96]. These field strengths are high enough for the manipulation of relativistic electron beams by polarised THz pulses in various geometries. It is possible, e.g., to construct a 30 cm long THz undulator for saturated FEL working at ~10 nm wavelength using electron bunches with 100 MeV energy and 0.4 nC charge. This allows an order of magnitude reduction of the electron energy, and the size (and cost) of FELs.

Another exciting application possibility is temporal focusing of electron bunches[97] to sub-fs duration by using, e.g., a geometry consisting of a THz standing wave with polarisation parallel to the propagation direction of the particles, and the interaction length restricted to a fraction of the wavelength. In combination with a single- or few cycle (THz) undulation such ultrashort electron bunches can offer a route to single- or few cycle sub-fs (or even attosecond) pulse generation. Recent theoretical studies show the promise of energetic low-frequency THz pulses in realizing compact electron acceleration and bunch compression schemes[98].

THz pulses can be used not only for manipulation but also for measuring ultrashort electron bunches. For example, the streaking technique was used recently for characterizing electron bunches in FELs[99,100].

Acceleration of protons to high energies by laser-plasma interaction is possible due to the production of hot electrons and the generation of very strong ambipolar fields. Hot electrons sheath production depends on large value of $I_{laser} \times \lambda^2$ and on steep plasma density gradients. Furthermore, the ion energy scales approximately with irradiance $(I_{laser} \times \lambda^2)^{1/2}$. High-field and low-frequency THz pulses may offer an alternative route to particle acceleration since the ponderomotive potential has a quadratic dependence both on electric field and on the inverse of the laser frequency, thereby offering very high values of the ponderomotive potential.

Core electron science

Motivation, state of the art

The foreseen unique attosecond beam parameters at wavelengths ranging from the XUV to the x-ray spectral region open up excellent perspectives for novel core electron experiments. Obviously inner-shells are accessible due to the short wavelengths available. In the soft x-ray domain, ionization occurs mainly removing electrons from inside an atom in contrast to the outer-shell peeling caused at wavelengths in the IR-VUV spectral region. This is conventional physics that has been extensively studied e.g. at synchrotron installations. The exceptional combination of different short pulse radiation sources, emitting from THz to x-rays, that ALPS will offer, allows time resolved studies of core electron dynamics at highest possible temporal resolution. Furthermore, the ultra short pulse durations and XUV/x-ray intensity levels of ALPS open for the first time the route to non-linear inner-shell processes. The two-color and non-linear core electron interactions perspective of ALPS is discussed and established in the following.

[96] C. Vicario et al., Opt. Lett. 39, 6632 (2014).

[97] A. E. Kaplan and A. L. Pokrovsky, Opt. Express 17, 6194 (2009).

[98] L. J. Wong et al., Opt. Express 21, 9792 (2013).

[99] U. Frühling, J. Phys. B 44, 243001 (2011).

[100] I. Grguras et al., Nature Photon. 6, 851 (2012).

Research directions and activities

While high photon energy photon fluxes at unprecedented pulse durations is expected to allow the study of non-linear soft x-ray inner-shell processes, two-color applications take advantage of the multi-synchronized-beam possibility offered by ALPS.

Electron streaking with mid-infrared light fields

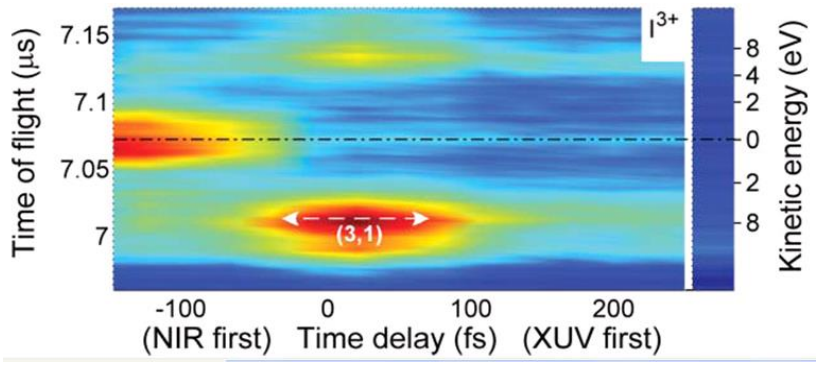
Long wavelength radiation, if accurately synchronized with the attosecond pulses can be used for the streaking of electrons released from inner-shells by the attosecond pulses

To give an example the 3 nm -12 nm radiation of the HR-P2 beamline photo-ionizes atomic core shells. The synchronized MIR radiation of the beamline HR-T1, with wavelengths 10 μm – 100 μm, can be used in order to streak photo/ Auger electrons. The long wavelengths of the MIR radiation provide substantial streaking even at moderate MIR intensities, at not highest the possible but at sufficiently high temporal resolution. The combination of these two sources serves perfectly the study of electron relaxation dynamics in the few fs range. Stage 1 completion provides ideal conditions for performing these experiments.

Ion-Charge-State Chronoscopy with XUV-pump /IR, vis, UV-probe schemes

The interaction of molecules with electromagnetic radiation leads in several cases to complex intertwined electronic and nuclear dynamics. XUV/X-ray radiation induces core dynamics in molecules such as, interplay between Auger decay and nuclear motion, ultrafast charge migration in molecules upon inner-shell absorption and structural rearrangement, izomerization & elimination reactions. Introduction of two color interaction (e.g. XUV/X-ray pump by SYLOS or HR – IR/visible probe by SYLOS or HR) allows the study of such dynamics in a path selective way when many reaction channels are present.

A recent example is the study of photofragmentation dynamics of molecular iodine interacting with femtosecond 800 nm near-infrared and 13 nm extreme ultraviolet (XUV) pulses at FLASH[101]. The delayed XUV pulse provided a way of following molecular photodissociation of I2 after the laser-induced formation of antibonding states. A preceding XUV pulse, on the other hand, preferably creates a 4d−1 inner-shell vacancy followed by the fast Auger cascade. Some fraction of molecular cationic states undergoes subsequent Coulomb explosion, see figure below, and the evolution of the launched molecular wave packet on the repulsive Coulomb potential was accessed by the laser-induced postionization. A further unexpected photofragmentation channel, which relies on the collective action of XUV and laser fields, was attributed to a laser-promoted charge transfer transition in the exploding molecule. The two-color measurements revealed characteristic times of these dynamics.



[101] M. Krikunova et al., J. Chem. Phys. 134, 024313 (2011).

Figure 31. Momentum resolved transient I^{3+} ion yields as a function of the time delay between XUV and NIR pulses. The axis at the right indicates the fragment kinetic energy for ions

One of the problems in this experiment was the limited time resolution due to XUV pulse duration and FEL-Laser jitter. The perfect synchronization of pulses of different sources at ALPS and the short duration of the XUV pulses can overcome this type of limitations. To give an example, the HR-P2 beamline XUV radiation at 3 nm -12 nm can create highly excited transient electronic states, while the HR-P1 UV pulses at 12 nm – 300 nm can select ionization / fragmentation channels. Thus, these experiments are foreseen for after Stage 2 completion.

Another example with similar timing is given by the time-resolved imaging of izomerisation reactions, e.g., similar to the recent XUV-pump / XUV probe experiment on C_2H_2 at FLASH[102], presented in the below figure. This experiment requires ≥1011 photons/pulse and thus could be performed at the SYLOS-S1beamline of ALPS with higher temporal resolution and avoiding the stochastic pulse shape of FLASH.

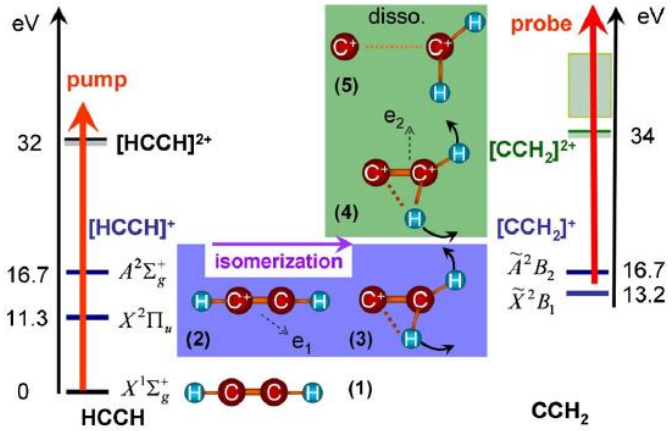


Figure 32. Time-resolved imaging of izomerisation dynamics in C_2H_2 cation studied with the XUV-XUV pumpe-probe scheme

Performing this kind of experiments with soft X-rays above carbon K-edge allows one to study isomerization dynamics upon inner-shell ionization (e.g., in the dication states) and visualize the interplay between the Auger decay and structural rearrangement of the molecule[103]. Furthermore, since ALPS will provide the two color option with few-fs time resolution, perfect X-ray/IR synchronization, precisely known pulse shapes and high repetition rate, this study can utilize XUV/soft X-ray pump / IR probe scheme, which could not be used at FLASH because of the FEL/laser jitter. This will not only allow one to avoid background signal originating from the same pump and probe wavelength, but also considerably relax requirements for the number of the X-ray photons, enabling these experiments for the SYLOS-S2 beamline of ALPS. Thus, these experiments can be performed after Stage 1 completion.

Interatomic Coulombic Decay: coulomb explosion

A novel electron-electron correlation effect that has recently attracted the interest of the scientific community is what is called an Interatomic Coulombic Decay (ICD) [104]. The effect is observable in Hydrogen bonded clusters suh as $(H_20)_n$, $(HF)_n$ as well as inVan der Waals clusters like (Ne)n; Ne-

[102] Y. Jiang et al., Phys. Rev. Lett. 105 263002 (2010).
[103] T. Osipov et al., Phys. Rev. Lett. 90 233002 (2003).
[104] L.S. Cederbaum et al. Phys. Rev. Lett. **79**, 4778 (1997); T. Jahnke et al. Phys. Rev. Lett. **93**, 163401 (2004)

Ar; He-He and can be used as a source of low energy electrons. In ICD an inner shell electron of one of the atoms of the cluster is ejected through absorbtion of an XUV-photon. The created hole is filled by an outer shell electron of the same atom, while through electron-electron interaction a second electron, belonging to another atom of the cluster is ejected. The thus doubly charged cluster undergoes Coulomb explosion. The time scale of the process is of the order of 100fs. Its dynamics can be studied trough momentuim resolved coincidence measurements betwenn the different fragments and electrons. The temporal resolution and the intensities of the ELI-ALPS sources is by far suficient for such studies. At the same time the HR laser driven XUV source provides excellent repetition rates that are required for good statistics in the coincidence measurements, while the availability of reaction microscope end stations provide experimental conditions at their best.

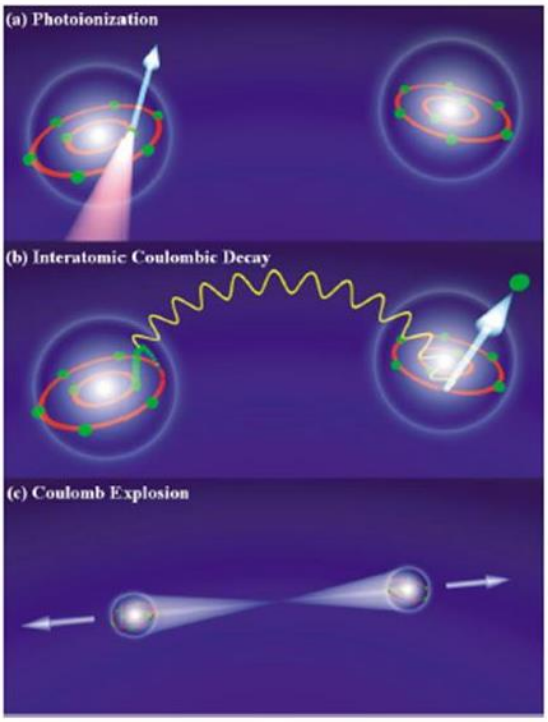


Figure33. Interatomic Coulombic Decay in Neon dimmer . From Ref [105]

Time-resolved photoelectron diffraction and holography

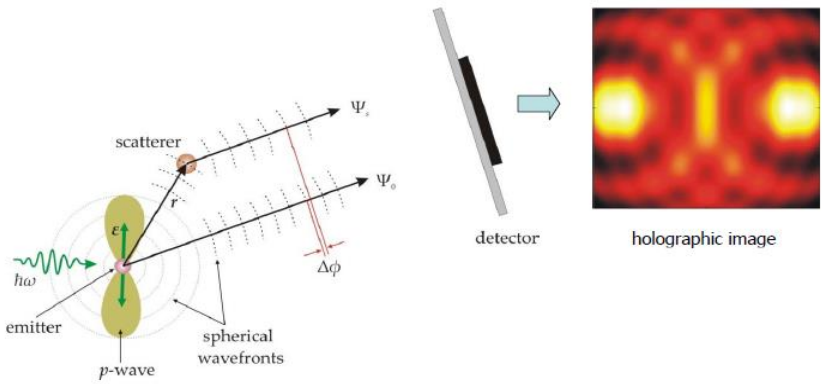


Figure 34.The sketch of the photoelectron holography approach

This is a new class of experiments that can be performed at ALPS utilizing uniqe combination of wavelengths and timing properties of its two beam facilities. Here, a visible/UV pump pulse of the HR/SYLOS beam-lines induces some molecular dynamics that could be photo-dissociation, bond rearrangement, photolysis via Norish I,II type reactions, dynamics at conical intersections etc. An X-ray probe pulse of the SYLOS-S2/H1 beam-lines ejects a core electron that leaves the molecule with ~0.01-1keV kinetic energy interacting with the neighbor atoms and producing a diffraction pattern on the imaging detector. Depending on the photon energy, and, thus, on the De Broglie wavelength of the photoelectron, these data can provide information on the molecular structure and electronic potentials, and in certain cases allow for a holographic reconstruction of the positions of the nuclei. The latter principle is illustrated in the above figure[105].

The emitted photoelectron can reach the detector either directly or after being scattered on one of the neighbor atoms, thus producing a holographic image of the molecule. The variable delay between the two pulses allows one to acquire a series of time-resolved snap shots of the molecule. The ejection of the core electron provides selectivity of the atom it originates from and thus Å scale spatial localization of the source. The spatial resolution is defined by the excess kinetic energy of the electron.

This type of experiments requires aligned (ideally, oriented) molecular ensemble. Currently, at the low repetition rate FEL sources, it is realized via laser-induced alignment. The high repetition rate of the ALPS will enable a posteriori determination of the molecular orientation from the emission direction of the fragment ions in photoion-photoelectron coincidence experiments. This approach, which is free of the alignment laser field and its influence on the molecular dynamics, is well-established at synchrotron sources, where, however, time-resolved experiments are not feasible because of the pulse duration. The few-fs time resolution and perfect laser/X-ray synchronization of the ALPS will provide optimal conditions for such investigations. The more involving nature of these experiments render them ideal for after Stage 2 completion.

Non-linear XUV/x-ray inner-shell interactions

Due to the short wavelengths of the radiation and consequently to the restricted ponderomotive energy interactions are mainly of multiphoton character. The figure below shows an example of the adiabaticity or Keldysh parameter dependence on the pulse wavelength and intensity. The value of this parameter with respect to unity determines the multiphoton or tunneling character of the ionization process. At the two-photon inner-shell ionization of this example with intensities $<10^{18}$W/cm^2 the interaction is of multiphoton character for all wavelengths. At higher intensities though, becoming available at ALPS-SYLOS, XUV wavelengths can lead to strong field interactions, leading to tunneling-type ionization of inner shells. While rates and efficiency of two-photon inner-shell ionization, competing with single-photon outer-shell ionization are discussed below, strong field effects and tunnel ionization of inner shells is an untouched research area that could be investigated at ALPS.

[105] F. Krasniqi et al., Phys. Rev. A 81 033411 (2010).

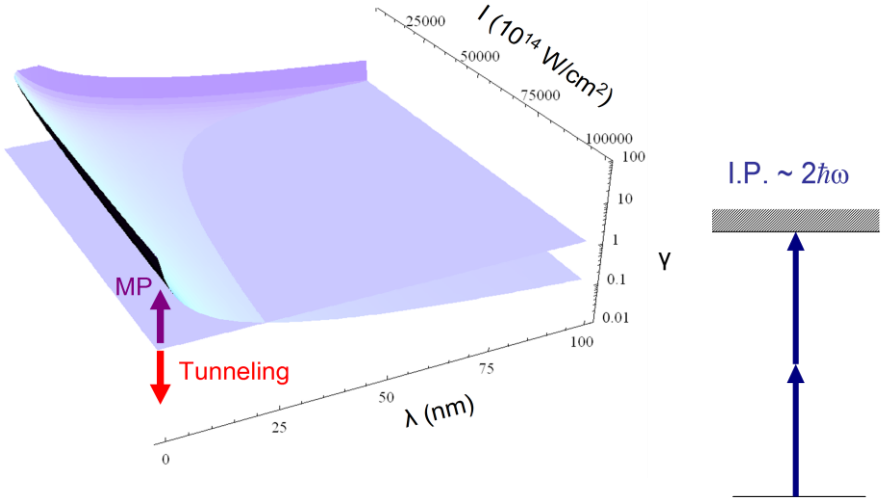


Figure 35. Adiabaticity (Keldysh) parameter as a function of laser intensity and wavelength for a two-photon ionization scheme

Multi-photon inner-shell processes

Multi-photon processes of core electrons have not been studied or observed so far because of two until today missing parameters. The required high radiation intensity in order for process to be observable and the short pulse duration in order for the atom to "see" the high intensity before it is completely ionized through single-photon outer-shell ionization. The figure below shows two-photon K-shell ionization generalized cross sections (blue curve) of H-like ions and required photon energies as a function of the atomic number Z. The red curve shows the K-shell ionization energy. H-like ion cross sections is a good estimate for the corresponding atom having the same K-shell electron binding energy as inner-shell electrons are weakly perturbed by outer-shell electrons. The colored band indicates the photon energy region to be covered by ALPS-SYLOS.

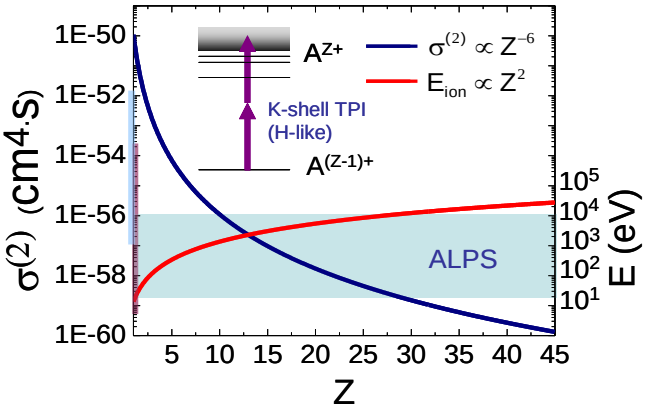


Figure36. Generalized two-photon K-shell ionization cross-sections and ionization energies of H-like ions

For the foreseen pulse energies at ALPS a two-photon K-shell ionization signal is estimated to be observable through energy-resolved photoelectron spectroscopy for elements with Z up to 30. However, the two-photon ionization peaks will appear on top of significantly more pronounced single photon outer shell ionization spectra and thus not obviously observable. What is markedly interesting, though, is that for low Z numbers (up to Z=10), due to the very short pulse duration the two-photon K-shell ionization can dominate the single-photon L or shells ionization prior to the depletion of the atom. This allows the convenient study of the two-photon process from ion mass

spectra. At attosecond pulse durations saturation of ionization shifts to higher intensities above the intensity threshold at which two-photon inner shell ionization becomes the dominant ionization process. This is a unique property of ultra-intense attosecond radiation sources and thus of the ALPS installation not available in any other XUV/x-ray source, such as synchrotron or XFEL sources.

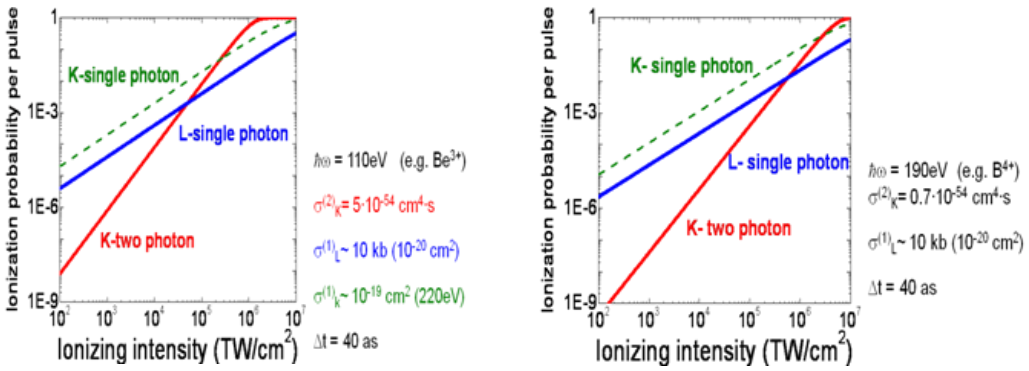


Figure 37. Two-photon K-shell, single-photon L-shell and single photon K-shell ionization probability of H-like Be and B

The above figure illustrates this unique property in example estimations. The figure compares the two-photon K-shell with the one photon L-shell ionization in Be and B. For the given pulse duration of 40 fs the two-photon process becomes stronger at intensities above $5x10^{16}$ W/cm^2 and $5x10^{17}$ W/cm^2 respectively and prior to ionization saturation. For Z > 5 the saturation of ionization sets above the intensity threshold for relativistic interactions. This opens up a further entirely new research topic that can be at the center of the ALPS research, namely relativistic/ultra-relativistic XUV/x-ray processes. Here again the key parameters are high intensities and short pulse duration so that the highest values of the spatiotemporal intensity distribution are "seen" by the atom before it falls apart.

Multi-photon inner-shell processes are important tools for a number of applications. Time-resolved spectroscopy, such as pump-probe spectroscopy relies on few (commonly two) photon processes. Accessibility to such processes will open up the path to time domain studies of inner-shell dynamics and in particular of inner-shell electron-electron correlation dynamics. In the area of diagnostics, multi-photon processes allow analytical applications in bulk materials at highest spatiotemporal resolution. The key point is that the intensity-dependent process is observable only at the focus of the radiation, thus providing spatial selectivity, keeping at the same time the option of pump-probe measurements open. To give an example, a K-shell hole can be created by a multi-photon process at a very specific 10-16 m^3 scale volume, elemental analysis being possible observing L-shell fluorescence. Furthermore, inner-shell multi-photon processes provide a valuable tool for the temporal characterization of the attosecond pulses. These experiments are foreseen for Stage 2 of the project.

Direct multiple inner-shell ionization

Under the existing and presently developing FEL and XFEL radiation sources, it has been possible to observe multiple ionization, including stripping from inner shells, as well as coupling between Auger resonances, albeit to a rather limited extent and results still needing refinement. Double core hole has also been observed in at least one occasion. As far as inner-electron dynamics is concerned, in atomic and molecular systems, what would usher in a novel area of studies is non-linear and/or strongly driven transitions involving inner electrons. Although transitions from inner shells and

even removal of all 10 neon electrons, as well as high stages of ionization in all rare gases, have been observed, the underlying mechanism has been a chain of sequential, multiphoton stripping, beginning in, for example, cases like xenon from lower subshells like the 4d. The basic limitation has been the relatively long pulse duration, say 10 fs or longer, and the low repetition rate. At the 1fs pulse duration level inner sub-shell two-XUV-photon direct double ionization has very recently made its debut (ref. P. Tzallas et all. Nature Physics 7, 781 (2011). Considerably shorter pulses are needed so as to explore in real time the dynamics and strong coupling of Auger resonances, while higher repetition rate is essential in the study of photoelectron energy and angular analysis of novel processes, such as, for example, direct double and eventually multiple ejection. For example, already at the SYLOS-S1 beamline of ALPS, XUV radiation of wavelength 12 nm -120 nm can multiply ionize atoms from core levels. In addition, the FEL pulses exhibit intensity fluctuations, essentially similar, although technically speaking not exactly identical to a chaotic state of the radiation field. These and new under development sources will steadily improve in the future, however, it is not fully clear to what extent and when. Reproducible XUV pulses with predictable temporal shape at ALPS will enable conclusive data interpretation in these types of experiments.

The ELI-ALPS project appears promising in circumventing those limitations.If we take as an example and reference point energy of 10μJ, pulse duration 100 as and photon energy 500 eV, it gives 1011 W. For spot area 10^{-8} cm^2, it gives $10^{19}W/cm^2$. The corresponding photon flux is then be about 1035photons/cm2sec. Taking, to be conservative, a cross section of 10-20 cm^2, it gives a lifetime against single-photon ejection of 1 fs. Clearly, changing the intensity by one order of magnitude, or the pulse duration by a factor of 5, or reducing the focal area by a factor of 10 or even 100 etc., it appears that one can reach the transition rates of sub fs, which means shorter than many lower shell hole states. If at the same time, the temporal shape of the pulse is smooth (no fluctuations) a number of possibilities appear within reach.

Some examples are:

1) Strong driving of an Auger resonance; interplay between Rabi time and lifetime.

2) Coupling, strong or weak, of Auger resonances. This would be a unique way of testing atomic structure and dynamics of highly excited states. An example of this in the VUV-XUV range has been discussed some time ago in Themelis et al.[106] and has served as the motivation for a recent experimental attempt in Argon. Here the possibility of synchronization of an X- ray pulse (say, soft X-ray) with an optical or UV source offers great possibilities. Such synchronization has been a difficult task for experiments of the pump- probe type at FLASH,

3) Although double core hole creation under FEL radiation was observed last summer at LCLS, one could now realistically ponder the creation and study in time of several holes. The high repetition rate can make feasible the study of photoelectron energy spectra of such highly excited hollow atoms.

4) From on-going theoretical work, it can be said that applying radiation of about 200 eV photon energy, intensity of the order of 10^{19} W/cm^2 and pulse duration of about 100 as one can realistically contemplate lifting all 10 electrons of the 4d shell of xenon to the continuum. Since this is a d→f transition with the double well, quite novel behavior in unknown territory can be expected. The

[106]S. Themelis, J Phys B 38, 2119 (2005)

above is an extreme case of more modest direct multielectron ejection processes that have been introduced and discussed recently in Lambropoulos et al.[107].

5) The above multi-core-electron ejection is a generalization of the much discussed Helium 2-photon double ionization, for which to this day there are no data on photoelectron energy spectra because the existing sources (around 40-60 eV) are either not sufficiently intense (HHG) or the repetition rate is too low (FLASH). The experiments in this section are foreseen for Stage 2 of the project.

Attosecond imaging in 4D

Motivation, state of the art

Atoms, molecules, crystals, nanostructures or biomolecules consist of two essential ingredients: nuclei and electrons. The 3D-arrangement of atoms and charge densities determines the structure and the material's static properties, but functionality and reaction to changes involve the motion of atoms and electrons from initial to final configurations. A 4D-recording in space and time is therefore required.

Today, 4D imaging is possible with femtosecond time resolution and atomic-scale spatial resolution, for example by electron diffraction/microscopy[108], by high-resolution 'flash' imaging in free electron lasers [109], or by table-top experiments with laser-excited plasmas[110]. Recent findings include the visualization of atomic pathways during phase transformations and melting processes[111,112], nanostructural transformations[113], or the dynamics during explosion of particles/foils[114,115]. Common to these achievements is, however, the limitation to structural and atomic motions, while the dynamics of electron density could not be accessed. Questions regarding the fundamental nature of light-matter interaction, the ultimate speed limits of electronics, the role of molecular orbitals for the making and breaking of bonds, the pathways of charge transfer in biological systems, or the function of photonic materials and nanostructures are therefore still largely unanswered.

By virtue of ELI-ALPS, with its unique combination of synthesized light fields with x-ray pulses of attosecond duration, we will advance 4D imaging into the regime of electronic motion. By time-resolved diffraction and scattering (see figures below), the most fundamental primary processes of light-matter interaction can be visualized with combined attosecond and Angstrom resolutions. This will reveal how light-matter interaction works on the fundamental scale of field-electron interaction, and how to impose control on an electronic time and length scale.

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[107] P. Lambropoulos, Phys. Rev. A 83, R021407 (2011)
[108] A.H. Zewail, Science 328, 187 (2010).
[109] H.N. Chapman et al., Nature 470, 73 (2011).
[110] M. Wörner et al., J. Chem. Phys. 133, 064509 (2010).
[111] P. Baum et al., Science 318, 788 (2007).
[112] R.J.D. Miller et al., Acta Crystal. A 66, 137 (2010).
[113] O.H. Kwon et al., Science 328, 1668 (2011).
[114] H.N. Chapman et al., Nature 448, 676 (2007).
[115] C.M. Günther et al., Nature Photonics 5, 99 (2011).

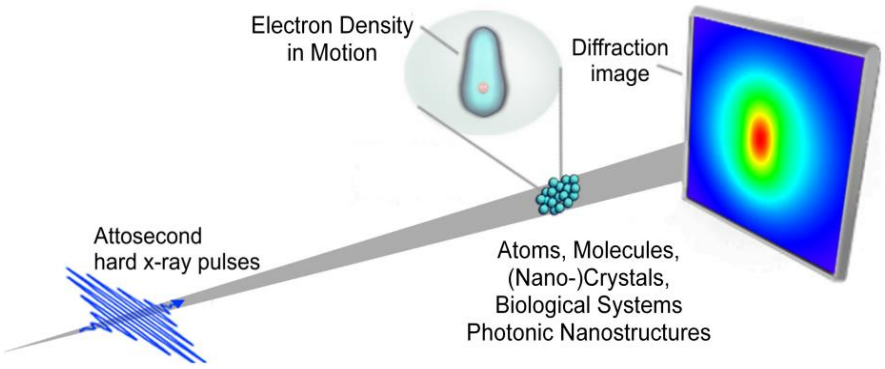


Figure 38. General concept for visualizing the attosecond dynamics of charge density motions (see text)

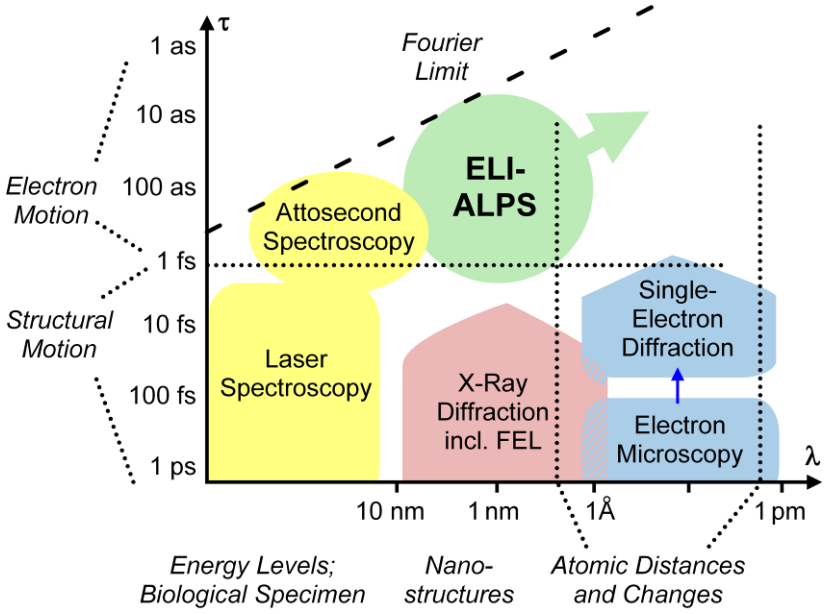


Figure 39. Current approaches for visualizing structural dynamics

The above figure shows an overview regarding current approaches for visualizing the ultrafast dynamics within matter. Spectroscopy (yellow) is currently the only technique capable of reaching attosecond resolutions; the novel concepts that ELI-ALPS offers here are described in the (2) Secondary Sources chapter. Ultrafast electron diffraction and microscopy (blue) offer picometer wavelength and superior resolution of atomic distances, but Coulomb repulsion and the fermionic nature of the electron prevent short pulses to be generated with high density and flux. Regarding the possibility to image electron densities in motion, a central disadvantage of using electrons as probe beam is the inherent sensitivity of the scattering and diffraction processes to nuclear potentials, which are only partially shielded by the inner electrons, while x-ray diffraction measures electron densities only. X-ray plasma sources and free electron lasers (red) offer wavelengths short enough for structural imaging, but lack of sufficient time resolution and reproducibility for reaching the attosecond time scale of electron dynamics. In free electron lasers, attempts are made to shorten the pulse length; the most promising development is seeding. However, the temporal characteristics will not exceed those of ELI-ALPS, where precise control and synchronization of electromagnetic fields, including absolute phase, is an inherent characteristics of the fully coherent x-ray generation mechanism. This ‘phase controlled attosecond advantage’ is therefore unique and motivates most of this section’s proposals. As the proposed experimental techniques and methodology is still in their infancy, the realization of the proposed research directions and activities are foreseen for the

period from 2016 on, thus Stage 2 of the ELI-ALPS project will provide ideal conditions for these studies.

Research directions and activities

4D imaging is based on diffraction or direct imaging with probing pulses at a wavelength comparable to the structures of interest, and a short enough duration for snapshot imaging. Our envisioned experiments are all feasible in a pump-probe arrangement and do not a priori require single-shot or single-particle imaging, however attosecond timing is essential. There is good hope that ELI-ALPS' attosecond pulses will be intense enough, especially on the SYLOS system, for single-shot imaging at a later stage, but this is not essential here.

Imaging charge dynamics with x-ray diffraction requires a significant influence of moving charge density to diffraction, in order to be instructive. Estimations show that the structure factor of one delocalized electron (sphere of 0.1 nm diameter) has the same magnitude as a neutral hydrogen atom[116]. Motion of charge densities with femtosecond resolution has already been experimentally resolved with plasma-generated x-ray pulses3; this will not be different on attosecond time scales. Therefore, charge density in motion is detectable as a change in diffraction during the dynamical process.

Attosecond pulses have non-monochromatic spectra; for example 100-as corresponds to a bandwidth of several tens of eV. This is significant, but still small as compared to the XUV and x-ray wavelengths required for diffraction (see below). Imaging and diffraction with attosecond resolution requires small enough samples, such that the scattered attosecond pulses can interfere with themselves. At the speed of light, 100 attoseconds correspond to a distance of 30 nm, which poses a limitation to the complexity of samples to be studied. 30-nm is, however, well above the size of most (bio-)chemically interesting substances; in larger systems, an electron moving that far within attosecond times is hardly conceivable, since it would approach the speed of light. If the dynamics is slower, however, longer probe pulses can be used. The connection of timing to the achievable resolution is therefore no significant limit for our studies.

In the following we describe several examples of possible studies; this is intended to provide a general overview of what fields could be covered and shall not be understood as an exclusive list. Theoretical physics is currently starting intensely to consider the possibilities of 4D-electronic imaging[117,118]. From such studies, we are sure that many additional and feasible ideas will be generated until the commissioning of ELI-ALPS and thereafter. All examples are doable in a pump-probe arrangement and do not require single-shot or single-particle imaging, however attosecond timing is essential.

Origin of the refractive index and nonlinear optics

A material's refractive index originates from coherent and phase-delayed oscillations of charge density in the light field, as described in physics textbooks. It is, however, largely unknown what types of charges (inner or bound electrons, localized or delocalized bands, holes or quasiparticles) provide what type of phase delay and re-radiation strength. This becomes even more significant in nonlinear optical processes, where nonlinear responses, usually into different spatial directions, lead to the generation of novel frequencies and polarizations. Diffraction of ELI-ALPS' attosecond x-ray pulses from a dielectric crystal, such as for example BBO (beta-barium-borate), during the presence of a light field, will lead to a diffraction pattern of the crystal, but with Bragg spot intensities changed by the displaced charge density. Since the crystal will not be destroyed,

[116] P. Baum et al., Chem. Phys. 366, 2 (2009).
[117] P. Baum et al., Science China G 53, 987 (2010).
[118] H.C. Shao et al., Phys. Rev. Lett. 105, 263201 (2010).

extended pump-probe measurements are possible and will map the three-dimensional motion of charge density in the material with optically phase-resolved 4D-attosecond resolution. Such studies have the potential to let us understand, control, and advance nonlinear optics from a fundamental perspective, and will open up a route towards directed engineering of novel functional materials for optics and lasers.

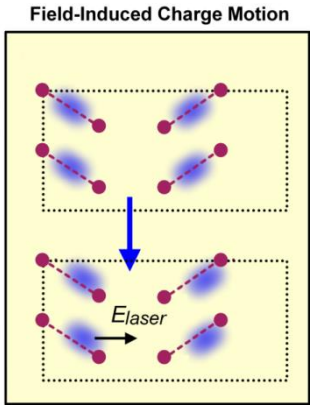


Figure 40. Illustration of field-induced charge motion

Charge transfer in light-sensitive molecules

Molecules hold promise for going beyond nanoscience in the construction of ultra small machinery, efficient energy converters, ultrafast switches, or biological agents. Photo-induced charge transfer is an essential mechanism for such applications, for example in dye-based solar cells, the processes of vision and photosynthesis, or in some ideas for molecular machinery. For example, charge transfer in organic solar cells is ultrafast (down to 6 fs) and is believed to involve a combination of structural and electronic dynamics[119]. Or, the light-energy conversions process in Bacteriorhodopsin involves electron and proton transfer on ultrashort time scales[120,121122], but the spatial pathway and the primary channel of motion on femtosecond scales are largely unknown. ELI-ALPS' XUV pulses can provide a 4D-movie of charge in motion within such molecules. Typically, one or less than an electron is transferred from one to the other side of the molecule, over nm distances. Probe pulses with nm wavelength are therefore sufficient for the here envisioned imaging, because the atomic structure does not need to be resolved. Diffraction experiments will be done at ALPS-SYLOS-S2, and also at -H1 if necessary. We conceive first studies to be made with single crystals, which are available from many photosensitive and biological molecules; this will highly facilitate diffraction and repetitive pump-probe experiments. Later, single particles can be imaged as well, with more intense pulses.

Electromagnetic field flow in nanostructures, metamaterials

Photonic materials are currently being developed, in order to engineer optical behaviour that can not be found in nature. Examples include materials with negative refractive index or left-handed electromagnetic susceptibility. Metamaterials usually gain their properties from nanoscale structures rather than composition. Their function is therefore determined by the dynamics of the interaction of light with the material's sub-wavelength structure. ELI-ALPS will provide the possibility to image the flow and oscillation of charge within such structures. The nanostructure can

[119] B. O'Regan et al., Nature 353, 6346 (1991).
[120] G.I. Groma et al., PNAS 105, 6888-6893 (2008).
[121] K. Edman et al., Nature 401, 822-826 (1999).
[122] L.Fabian et al., Opt.Express 19, 18861-18870 (2011)

be excited with visible, infrared or THz fields, and XUV pulses can be used in an imaging/microscopy arrangement to record images of the structure; thereby, the transient charge density should be observable by phase contrast mechanisms[123]. No particular short probe wavelength is required and these experiments will be feasible with the ALPS-SYLOS-S1/S2 source.

Causes of chemical bond breakage

In chemistry, the making and breaking of bonds is defined by a sufficient separation or sufficient converging of the two atoms. The primary cause for bond breakage is often a change in the molecular orbital, for example by light that changes the gluing forces between the atoms to become repulsive. By virtue of ELI-ALPS' attosecond/few-femtosecond x-ray pulses, the electronic structure of a photo-excited molecule could be imaged before the atoms have time to move apart, thus before the photo-induced structural dynamics starts. This will reveal the role of orbital changes to the subsequent chemical pathways and will clarify what synthesized light pulses could steer chemical reactions by controlling transient electronic motions and the intermediate shapes of molecular charge densities. Also, conical intersections can be studied; they play a key mechanistic role in non-adiabatic molecular processes, because the energy exchange between the electrons and nuclei becomes significant and ultrafast. In many important cases like dissociation, proton transfer, isomerization, and radiationless deactivation of the excited state, for example in DNA, conical intersections provide very efficient channels for ultrafast interstate crossing on the femtosecond time scale. Technologically, this research requires the short wavelengths of the ALPS-SYLOS-H2 source.

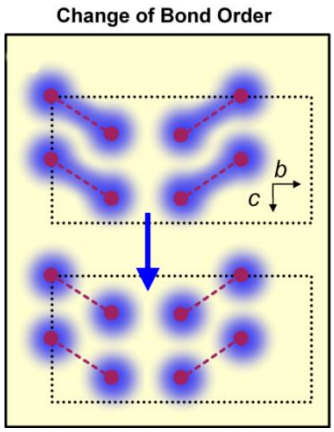


Figure 41. Illustration of change of bond order

Spatio-temporal inner-shell dynamics

A central question of atomic physics is the role and time scale of multi-electron interaction during ultrafast changes such as ionization. Spectroscopic approaches can reveal transient, non-equilibrium electron configurations by time-resolved energy measurements (streaking); the Secondary Sources chapter described how ELI-ALPS will make many of such studies possible for the first time. A central question, however, is whether the suspected sequences of electronic configurations indeed involve enough changes in energy to be resolved by spectroscopy. Here we outline one of the extreme perspectives of ELI-ALPS in this context: the 4D-imaging of inner-shell dynamics. The envisioned experiments consist of a sample of atoms, either as gas or in the condensed phase. We

[123] M. Dierolf et al., Nature 467, 437 (2010).

conceive a pump-probe arrangement with an attosecond x-ray pulse for impulsive, direct ionization of an inner-shell electron, and another attosecond x-ray probe pulse used for diffraction/scattering. Provided that the probing x-ray wavelength can resolve the inner shell, the diffraction rings will reveal transient changes in the atom's inner electron density by an increase/decrease of certain radial contributions, dependent on the attosecond time delay after ionization. Important is that this measurement does not rely on any significant changes in energy during the sequences of inner-shell rearrangements, but directly measures the transient charge density, including any spatial anisotropy or symmetry breaking. This experiment requires probe pulses with a wavelength considerably smaller than the atom's size, ~0.1 Angstrom. We are aware that this is the very shortest wavelength ELI-ALPS is believed to be able to provide (ALPS-SYLOS-H2). If so, however, this experiment represents a totally novel way of searching for the ultrafast dynamics of electron-electron interaction, independently of energy levels.

Multiple coherent projections for single-particle imaging

Simultaneous illumination from multiple directions and with synchronized/coherent pulses could provide a full 3D view of an object. Methods have been proposed for obtaining three simultaneous projections of a target from a single radiation pulse[124,125]. ELI-ALPS offers new possibilities here, because timing precision is perfect (on the attosecond scale) and all three pulses are naturally fully coherent to each other. The principal axes of a compact charge-density distribution can be derived from projections of its autocorrelation function, which is directly accessible from the individual diffraction patterns in this geometry. Combined with a synthesized light field for the initiation of dynamics, 4D-structural movies of single particles seem feasible to be obtained with ELI-ALPS. These experiments require the increased pulse intensities from ALPS-SYLOS-S1/S2.

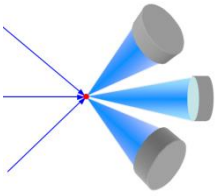


Figure 42. Illustration of multiple coherent projections for single-particle imaging

4.2 Laser plasma science – Relativistic interactions

Motivation, state of the art

The interaction of high power (TW to PW) laser pulses with any form of matter is usually accompanied by a host of processes that their dynamical evolution unfolds at atomic time scales i.e. at femto- or attoseconds. The study of these processes is possible only if the appropriate laser system is available to initiate them and simultaneously a short probe pulse can interrogate the state of the process in well defined temporal intervals. This capability can be realized only if a high power laser system is equipped with a synchronized femto- or attosecond pulse. This is exactly the combination envisioned for the ELI-ALPS facility in Szeged-Hungary. More specifically the two secondary sources ALPS-HF and the various versions of the ALPS-SYLOS are ideally matched for this purpose. This will be a unique capability carrying no similarity to any of those in other facilities worldwide. Some fundamental processes of outmost importance ranging from laser electron acceleration to inertial fusion energy to non-linear effects in quantum electrodynamics (QED) can

[124] M. Bergh, Quar. Rev. Biophys. 41, 181 (2008).
[125] K.E. Schmidt et al., Phys. Rev. Lett. 101, 115507 (2008).

now be put under real time scrutiny. As is described later the ELI-ALPS facility has the potential to provide the basic tools to attempt a fascinating observation that of tracing the vacuum breakdown.

Research directions and activities

Some of the envisioned applications of the unique capabilities to be offered by the ELI-ALPS facility are sketched in what follows.

Overdense plasma

As it was mentioned above in plasma of high (solid) density ROM is the most promising mechanism for future attosecond sources based on plasma mirrors, since the maximum generated frequency grows with laser intensity, and has been predicted to eventually reach multi-keV photon energies and pulse durations down to the zeptosecond range[126] for ultra-relativistic laser intensities. Efficient single attosecond pulse generation was predicted to occur when such intense few-cycle pulse is focus to a small volume.

Fast dynamical processes in overdense plasma

The ordinary laser plasma theory (model) is considering processes with duration comparable or bigger than few femtoseconds. Recent experimental technique gives us an opportunity to operate with even smaller (attosecond) time scales this demanding to revise some assumptions of standard models.

One important example is the investigation of fast dynamical processes in overdense plasma because in the field of dense plasma physics, in particular plasmas close to solid densities originating from the sudden heating of solid matter, the time scale of all plasma oscillations is set by the plasma frequency $\omega_p = 2\pi/T_p$. The period of these oscillations relative to the laser period is given by $T_p/T_L=(n_c/n_e)^{0.5}$ where the critical density n_c is about $10^{21}cm^{-3}$ for laser light of period about 3 fs and n_e - the actual electron density near 10^{24} cm^{-3}. This implies oscillation periods of 100 – 300 as, and thus time-resolved measurements will require pulses in attosecond range. Detailed investigations of plasma dynamics on these time scales will be crucial for a number of applications, such as laser-driven ion acceleration from thin solid targets and high harmonics generation from solid surfaces. Here Rayleigh-Taylor-type instability of laser-driven electron fronts can play a major role and need to be controlled (see, for example ref. [127])

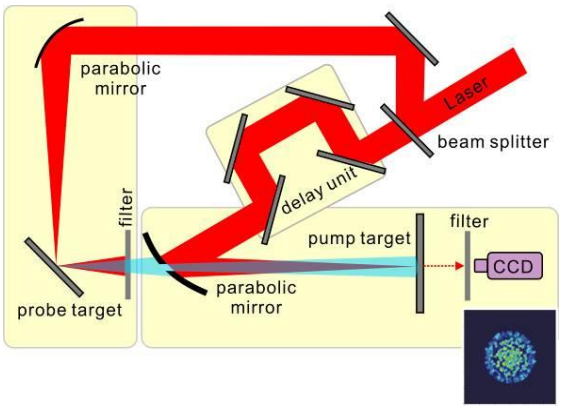


Figure 43. Scheme of a proposed setup for the study of the filamentation instability in dense plasmas

An example of outmost importance for the successful operation of the fast ignition concept in Inertial Fusion Energy (IFE) is the deposition of enough energy in the highly compressed fuel core

[126] S. Gordienko et al., Phys. Rev. Lett. **93**, 115002 (2004); A. Andreev et al., Quant. Electron. **41**, 729 (2011)

[127] S.Steinke et al., PHYSICAL REVIEW ST - ACCELERATORS AND BEAMS **16**, 011303 (2013)

by means of electron beams carrying currents of up to 1 GA. However, the beam transport is possible only if there is a counter current of cold electrons to counteract the generated strong magnetic fields[128].

The sub-cycle relativistic ionization dynamics can then be probed by an attosecond pulse, which when absorbed will lead to the production of a higher charge state. Similarly to [129], variation of the delay between the multiply ionizing pulse and the attosecond pulse will reveal the relativistic ionization dynamics. Besides multiple ionization, relativistic interaction (vxB- term) introduces a modified dependence of the partial sequential ionization cross-section on the electron ejection angle on a plane containing the propagation axis of the ionizing field. Measurement of the electron angular distribution will reveal the effect. Again the process is dynamic and depends on the phase of the ionizing field at which the electron is ejected. For a few cycle laser field, relativistic interactions can be restricted to the central cycle having the highest field amplitude. Probing of the ionizing phase can be through an attosecond pulse of appropriate central wavelength that will promote the ion to its next ionization stage.

Probing nanometer thick foil targets

In the quest for small scale schemes of particle acceleration laser plays an important role. The original idea of laser wakefield acceleration has advanced to the point that multi-GeV quasi-monoenergetic electrons are routinely generated (see figures below).

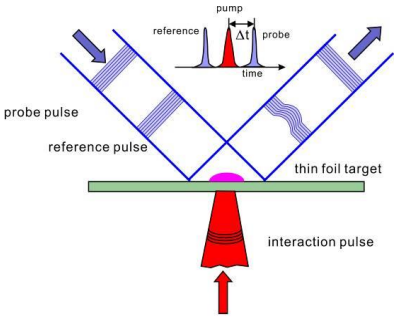


Figure46. The technique of frequency-domain interferometry for measuring the density profile of the expanding plasma

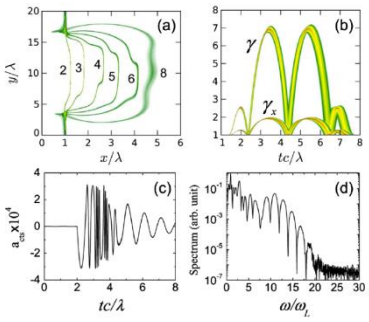


Figure47. Simulation results depicting the generation of relativistically moving electron sheath

[128] J.J. Honrubia and J. Meyer-ter-Vehn, Nucl. Fusion 46, L25 (2006).

[129] M. Ueberacker et al., Nature **446**, 627 (2007)

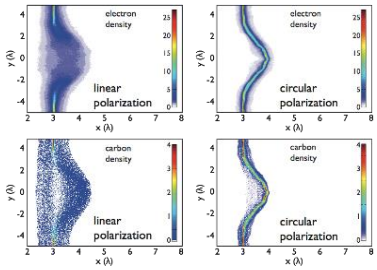


Figure48. Two-dimensional PIC simulations of thin foil targets interacting with linearly (left) compared to circularly polarized light (right)

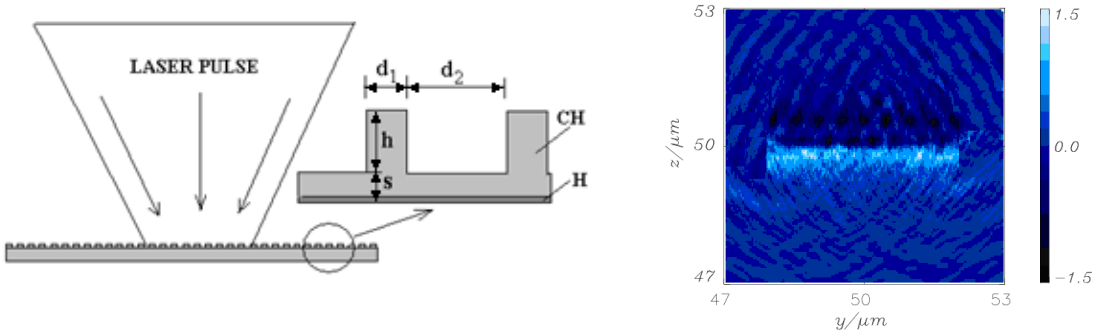
A new concept has been recently proposed in which using nanometer thick foil targets one can disengage the electrons from the ionic background and produce an electron sheath moving at relativistic velocities (see figures above). This is the ideal „mirror" on which a laser pulse can be coherently reflected to obtain Doppler shifted photons reaching far into gamma rays[130]. Similarly, ion acceleration using laser pulses has become an important alternative to large scale conventional sources for medical applications. Different mechanisms have been investigated to date but one that appears the most promising is the so called Radiation Pressure Acceleration, which relies on accelerating the electrons that in turn pull the ions by using circularly polarized laser pulses[131].

In both concepts it is important to be able to ascertain with high temporal resolution the state of the accelerated electron sheath (see figures above) or that of the thin foil target (see figures above). The availability of synchronized attosecond pulses with the PW laser pulses makes this requirement amenable to the well known technique of Frequency-domain Interferometry[132]. This is schematically depicted in Figure 47a and used to map out the expansion of the critical density surface of a femtosecond laser-produced plasma with subnanometer spatial resolution along the laser axis. The temporal resolution is primarily determined by the duration of the probe pulse.

Efficiency enhancement by structured targets

Typically, the efficiency of such a secondary source conversion is orders of magnitude lower than the percent level if simple targets as for example plane foils are irradiated. Thus, the main goal of this research activity is to achieve as high as possible conversion efficiencies where plasma structures will provide a basis for. Towards this goal in particular, the process of ion acceleration in mass-limited targets (MLT) has been studied.[133]. The kinetic energy of the accelerated ions is enhanced compared to that of a plane foil target and the ion beam divergence can be kept similar by applying a chain of MLT[134]. Careful control of lowest or a distinct pre-pulse level is an ultimate prerequisite for the experiment and the precise knowledge of this parameter is necessary for successful simulation and model development of the interaction scenario. As an example a very high intensity contrast ensures that the ion energy may be increased by using very thin targets, where hot electrons are refluxing[135] , but high contrast leads to steep density gradients and consequently, laser absorption decreases. We suggested resolving this dilemma by the application of well-adapted nano-structures on the foil front side. Optimum structure of a foil target permits to get almost total absorption of a laser pulse and transfers nearly all laser energy into hot electrons[136] .

[130]H.-C. Wu et al., Phys. Rev. Lett. 104, 234801 (2010).
[131] A. Henig et al., Phys. Rev. Lett. 103, 245003 (2009).
[132]J.P. Geindre et al.,Opt. Lett. 19, 1997 (1994).
[133] A. Andreev et al., Eur. Phys. J. Special Topics **175**, 123 (2009)
[134] A. Andreev et al., Laser and Particle Beams, **27**, 449 (2009)
[135] A. Andreev et al., Phys. Rev. Lett., **101**, 155002 (2008), ibid **105**, 015005 (2010)
[136] A. Andreev et al., Phys. Plasmas **18**, 103103 (2011)



The energy of the accelerated particles is increased without significant loss of its quality. Modification of the field distribution is the steering process for following energy transfer as the energy gain of electrons in the field and released secondary processes as radiation emissions by electrons or ion acceleration. Most important are the characteristic scales of the profile. These are parameters which are accessible with our laser structuring method and which we want to study and optimize for applications. An optimum thick and a laterally limited target together with an optimum structure would exhibit highest laser to line radiation conversion efficiency[137] . Beside the question of optimizing the laser absorption also envisioned short X-ray pulses need short bunch duration of fast electrons. The specific double foil arrangement was proposed such that the generated fast electrons interact with a secondary foil and can produce short pulses of K-α radiation with high efficiency.[138]. A novel scheme of short, energetic and dense electron bunch generation was offered for the interaction at large angles of incidence for the laser radiation. If a specific target is considered three streams of electron bunches arise, which propagate in different directions. The conversion efficiency into fast electrons and atto-pulses is a percent range. Additionally structured targets can permit a certain directionality of atto-pulses[139].

Summarizing we can state that high intensity laser pulses can generate nanometer sized bunches of fast electrons and thus attosecond pulses of soft X-ray radiation are already released from plane targets. We are aiming to use specific nano-structuring of the target surface which increases the efficiency of the process and permits further manipulation methods. Nano-structuring is based on a self organized and pure laser interaction process which is connected to a variety of phenomena which deserve further investigation.

Ray-tracing through overdense plasma

In inertial fusion energy the goal is the uniform implosion of a pellet containing the fuel to densities and temperatures appropriate for the ignition. This can be achieved only when the pellet irradiation is to very high degree symmetric and the implosion occurs free of instabilities.

The ELI-ALPS facility will provide the unique capability of visualizing the temporal evolution of imploding plasma. The simple technique of “harmonic backlighting” can be used to this end. The principle is outlined in the above figures where a synchronized pulse consisting of discrete harmonics illuminates an overdense pellet having a hollow density profile. Simple ray-tracing calculation reveals that depending on the harmonic wavelength, the different harmonics will

[137] A. Andreev et al., Quant. Electron., **41**, 515 (2011)
[138] A. Andreev et al., *Appl. Sci.* **3**, 94 (2013)
[139] A. Andreev et al., Optics and Spectroscopy, **114**, 138 (2013)

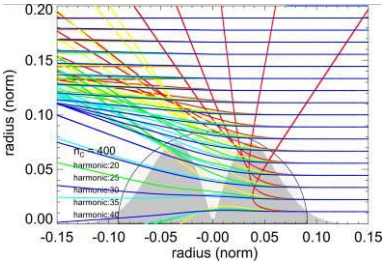


Figure 49

Ray-tracing of the indicated harmonic order through a hollow pellet with peak density of 400 overcritical

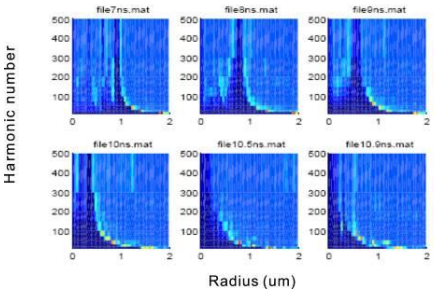


Figure 50

Hydrodynamic simulations depicting the mapping out of the evolution of the compression phase by recording the harmonic signal after passage through the pellet

follow a different path (see figures above). The density profile of the imploding pellet can then be deduced by unfolding the records at different times of the image thus obtained. Examples of such images from realistic hydro-simulations are shown in the above figures.

Underdense plasma

Intense laser pulse interaction with underdense plasma

In a number of applications involving multi-TW laser pulses like electron acceleration it is desirable to be able to follow the propagation of the plasma wave through the underdense plasma medium. The visualization of the accelerating structure is of outmost importance for the optimization of the plasma wave formation and loading. A proposed setup for this purpose that takes advantage of the capabilities of ELI-ALPS is shown in the figure below.

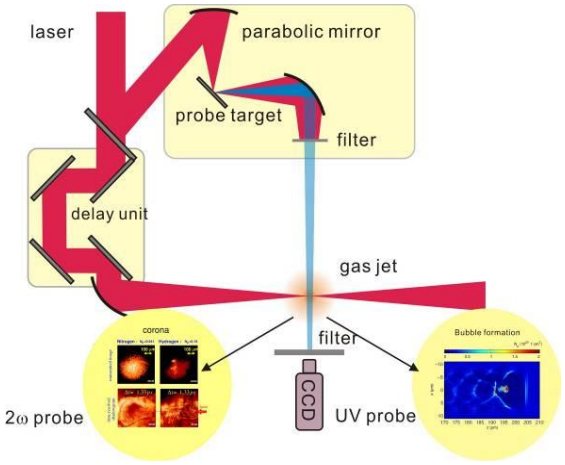


Figure 44. Schematic of a simple shadowgraphy setup for the investigation of relativistic self-focusing (left inset) or "bubble" formation (right inset)

This configuration has been used in the past to visualize the occurrence of the relativistic self-focusing in gaseous media using the second harmonic of the laser light[140]. The use of synchronized attosecond pulses in combination with more advanced techniques like Longitudinal Frequency-domain Spectroscopy or polarimetry will provide by far higher temporal and spatial resolution.

Multiple field-ionization of heavy ions

The interaction of an atom with a laser field is conventionally non-relativistic because of the depletion of the atomic ground state through ionization before the field increases to strengths leading to relativistic electron velocities. However, in multiple atomic ionization, higher charge states can survive the gradual increase of the laser pulse intensity up to values that lead to relativistic interactions. Evidence of relativistic interactions was obtained in high charge states of Xe (Xe^{19+}, Xe^{20+}) through the suppression of direct multiple electron ejection[141], when interacting with 60fs long IR pulses. Within the reclusion model, in order for simultaneous electron ejection to occur, the electron set free in the continuum has to revisit the atom in order to kick further electrons out. For intensities at which the $\vec{\upsilon} \times \vec{B}$ term is not negligible the electron trajectory may miss the origin, leading to a suppression of the e-e collision and thus of the simultaneous electron rejection.

ALPS-ELI provides the unique opportunity in performing time resolved studies of relativistic interactions. First of all due to the much shorter pulse durations of the different light sources at ALPS the effect will be observable at lower charge states due to their increased ionization saturation intensities. The degree of suppression of the "simultaneous" ionization will depend on the phase of the laser field at which the re-colliding electron will be born. This introduces interesting dynamics in the ionization process of much different character than the dynamics in the non-relativistic regime. The sub-cycle relativistic ionization dynamics can then be probed by an attosecond pulse, which when absorbed will lead to the production of a higher charge state. Similarly to the work by M. Uiberacker et al.[142], variation of the delay between the multiply ionizing pulse and the attosecond pulse will reveal the relativistic ionization dynamics.

Besides "simultaneous" multiple ionization, relativistic interaction introduce a modified dependence of the partial sequential ionization cross-section on the on the electron ejection angle on a plane containing the propagation axis of the ionizing field. The $\vec{\upsilon} \times \vec{B}$ term introduces velocity components along the propagation axis and thus a modified azimuthal angular distribution. Measurement of the electron angular distribution on a plane containing the propagation axis will reveal the effect. Again the process is dynamic and depends on the phase of the ionizing field at which the electron is ejected. ALPS provides the tools for the study of these dynamics. For a few cycle laser field, relativistic interactions can be restricted to the central cycle having the highest field amplitude. Probing of the ionizing phase can be through an attosecond pulse of appropriate central wavelength that will promote the ion to its next ionization stage.

Vacuum

The applications to high-field physics that can be done in ELI-ALPS for ultra high intensities (e.g. reaching the Schwinger limit, pair creation, etc.) will depend on focusing the attosecond pulses. This will require curved plasma sheets, and time resolved control of how they form and propagate will be crucial for success[143].

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[140] R. Fedosejevs et al., Phys. Rev. E 56, 4615 (1997).
[141] M. Dammasch et al., Phys. Rev. A 64, 061402 (R) (2001).
[142] M. Uiberacker et al., Nature 446, 627 (2007).
[143] T. Okada, A. Andreev et al., Phys. Rev. E **74**, 026401 (2006)

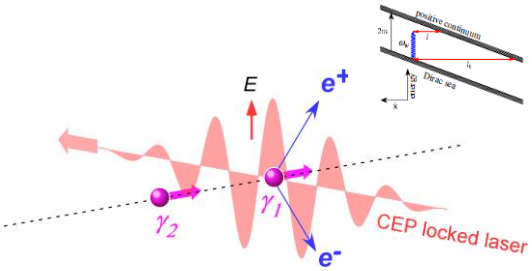
Following the vacuum breakdown and photon-photon scattering

The similarity between atomic physics and QED vacuum physics has not escaped our eye but over the half century work since the invention of laser this has been now more clearly recognized. This reflects, for example, in the similarity between theories of Schwinger and Keldysh.

Vacuum:

Schwinger/Nishikov field $E_{SN} = E_{S\sigma}(m_\sigma c^2/\hbar\omega)$

Schwinger field $E_{Se} = \alpha^{-3} E_K$



$\gamma_{V\sigma} = m_\sigma \omega c /eE = 1/a_0$

where $\sigma = e$, or q (quark)

Figure 50 Streaking the vacuum

The rapidly developing discipline of attosecond metrology and atom streaking is now capable of capturing the atomic dynamics in the time-domain measurement. Utilizing the combination of an ionizing XUV photon and CEP controlled intense laser pulse, it has been demonstrated that the electron time-of-flight can realize the attosecond atomic streaking. Here we suggest a parallel scheme of zeptosecond metrology composed of a gamma photon and intense CEP stable laser (or equivalent coherent CEP stable XUV pulse) to streak the vacuum and its dynamics out of Dirac's sea of vacuum. The created pair of electron-positron can be traced with the time-of-flight measurement, as is the case of the atom streaking. The parameter regime that realizes this streaking naturally produces the necessary space-time resolution of vacuum structure and dynamics in non-perturbative QED in the time domain for the first time, rather than in the frequency (or momentum) regime. Classically, photon-photon scattering cannot take place in vacuum. However, according to quantum electrodynamics (QED), vacuum can be polarized in strong external electromagnetic fields due to the exchange of virtual electron-positron pairs. Different theoretical schemes were proposed to detect elastic and non-elastic photon-photon scattering in QED vacuum (see review papers[144,145]) including light diffraction on a standing electromagnetic wave[146], four-wave mixing[147], generation of optical harmonics[148] and others. The demanding laser intensity in these schemes is higher than our case but "cascade" process[149] combined with influence of heavy ions [150] promises to get a significant yield of electron-positron pairs even for our maximal laser intensity. The laser created pairs in very low density plasmas can be traced with the time-of-flight measurement, as is the case of the atom streaking. The parameter regime that realizes this streaking naturally

[144]M. Marklund et al., Rev. Mod. Phys. 78, 591 (2006).
[145]G.A. Mourou et al.,Rev. Mod. Phys. 78, 308 (2006).
[146]A. Di Piazza et al., Phys. Rev. Lett. 97, 083603 (2006).
[147]E. Lindstrom et al., Phys. Rev. Lett. 96, 083602 (2006).
[148]A. Di Piazza et al.,Phys. Rev. D 72, 085005 (2005).
[149] A. Bell, J. Kirk, PRL **101**, 200403 (2008).
[150] A. Andreev, Generation and Application of Ultra-High Laser Fields, NOVA Science Publishers, Inc., New York (2001).

produces the necessary space-time resolution of plasma structure and dynamics in non-perturbative QED in the time domain for the first time, rather than in the frequency (or momentum) regime. The ELI-ALPS facility will open completely new and unique basis for theoretical and experimental consideration of nonlinear photon-photon scattering in QED vacuum. Ultra-short duration combined with ultra-high peak intensity of the pulses can have major effect on the output of the nonlinear interactions. Application of time-synchronized attosecond laser pulses from secondary ELI-ALPS sources will open a unique possibility for analyzing the dynamics of vacuum polarization in the elastic and non-elastic photon-photon interactions. This field puts high requirements on the laser system and this is where ELI-ALPS shows its lifelong renewability, because for such experiments 200 PW lasers are needed, meaning that ELI-ALPS could host the fourth pillar of the ELI project.

Outlook, future implications

The contributions expected from the operation of the ELI-ALPS facility will have far reaching consequences not only to fundamental physical questions but also to concrete applications. It is a unique combination of advanced technological development that will open up the prospects to original and path breaking advances in science with tangible implications to societal role of the resulting knowledge. For example issues relevant to particle acceleration for medical applications or to better understanding of the approach towards inertial fusion energy are only few to be mentioned. In addition, research in purely scientific topics will inevitably result in new technological developments with yet imperceptible contributions to new application. In particular the planned operation of a laser system of PW class in combination with a synchronized attosecond pulse source will open up the route to new ideas that can be tested using this facility. In short, it has the potential to provide the scientific community with an additional "eye" with which it will be possible to "see" in real time processes that were held obscure up to now.

4.3 Surface and condenced matter scienece

Motivation, state of the art

In traditional spectroscopy a continuous excitation of a solid sample results in a response of dipole (or in certain cases higher order multipole) oscillations, originating from the constituents of the medium, and producing a secondary electromagnetic wave. This signal is then Fourier transformed by the spectroscopic device itself, and yields, say, the absorption spectrum of the material. With the advent of short laser pulses the complementary possibility has become available, namely to observe the relevant processes directly in the time domain. In similarity with *microscopy*, which deals with high resolution observations of spatial structures, techniques that yield fine temporal details of a process have been termed as *chronoscopy*.

One of the methods of chronoscopy is the pump-probe technique: the pump pulse sets off some dynamical process and the probe examines it after some time delay. During the development of short pulse laser technology this approach was first applied to investigate chemical or biochemical kinetics with time constants already in the nanosecond range. In the last two decades of the past century the motion of atoms in molecules could be analysed in the time window from about 1 picosecond down to around tens of femtoseconds: this is the field called femtochemistry.

More recently phenomena connected with the dynamics of the electrons -- particles with the largest mobility in ordinary matter -- have become the subject of explicitly time dependent studies. The atomic unit of time $t_a \simeq 24$ *attoseconds* emerges as the natural scale in atomic problems where the quantum mechanical motion of the electrons play a decisive role. (Note that the time of one revolution in the ground-state Bohr orbital of the H atom is $2\pi t_a$ and the inverse of the Bohr frequency $(\Delta E/\hbar)^{-1}$ belonging to a typical atomic transition is also about of this order of magnitude.) In case of solids the relevant temporal scales are similar, as the time needed for an electron to travel

a distance of a lattice constant with the Fermi velocity $v_F \simeq 2\times10^6$ m/s is also around 100 *as*. Therefore, to reveal and understand the dynamics of electrons in atomic or condensed matter systems a time resolution in this range is necessary. Detailed time-resolved measurements of the motion of electrons in solids (as well as in atoms) used to be out of reach before 2005, and the first rudimentary attempts to study this problem have begun less than about 10 years ago. The importance of understanding the electronic processes and the possibility of even controlling them was the main reason of the decision to build a scientific facility that can resolve the details of such phenomena. The implementation of the ELI ALPS laboratories will respond this challenge, as it is going to enable its users to perform experiments with electromagnetic pulses in the attosecond range with appropriate intensity, repetition rate and control.

One of the most important tools in the investigation of solid surfaces by ultrafast pulses is laser-assisted-photoemission (LAPE). An attosecond XUV pulse originating as a high harmonic (HH) pulse produced in a scattering process from a noble gas jet is directed to the surface of a material and generates photoelectrons. Part of the strong near infrared (NIR) few cycle pulse, which is used to trigger HHG, accompanies the XUV photons and accelerates the photoelectrons depending on the time delay between the pulses. The emission time of the electron can be determined by measuring its velocity depending on the relative synchronization of the two pulses. This novel technique has been termed also as attosecond streaking[151]. A less drastic perturbation is if the ultrashort pulse results only in a temporary redistribution of the electrons in the sample but without triggering electron emission. In that case just the observation of the scattered field can be a witness of the processes in the solid.

Similarly to such studies on isolated atoms in the gas phase, these methods are to be made possible by the ELI-ALPS infrastructure, and promise to obtain information on (i) the structure and dynamics of delocalized valence and conduction-band states, (ii) on complex correlated electronic systems (iii) on electron transport properties in solids, and (iv) on the decay of collective excitations near solid surfaces and of novel plasmonic structures. It is important to note that these kinds of experiments implicate another possibility in perspective. The electronic effects triggered by the electromagnetic field of the laser will be characteristic of the XUV pulses themselves providing thereby information on the spatial and temporal structure of the light pulses themselves.

Surface science represents an area where the ELI-ALPS HR and SYLOS laser systems can have a huge impact. Attosecond surface science is in its early days, with the only results having been reported so far representing some pioneering experiment which revealed a delay in the photo-emission originating from localized core states in tungsten and the delay of electrons passing through magnesium adlayers, with respect to photo-emission involving delocalized conduction band states[152 153]. The unique combination of high-pulse energy laser sources available at ELI-ALPS will offer unique scientific opportunities to study the coupling of THz-driven coherent excitations of the lattice and spin systems to the dynamics of long-range valence orders in strongly correlated electron systems. These couplings are at the heart of many key properties of such systems, and the new information offered by these new experiments may lead to the design of better materials with wide-ranging applications.

Research directions and activities

The promise of attosecond surface science is enormous. It ranges from electron dynamics in metals and dielectrics, the real time monitoring of dynamical modifications of orbitals and electron transfer on surfaces, though time resolved Angularly Resolved PhotoEmission Spectroscopy (ARPES) to the study of a manifold of new phenomena that occur in materials due to the strong electron-electron correlations that they exhibit.

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[151] F. Krausz , M. Ivanov, Rev. Mod. Phys. **81**, 163 (2009) DOI: 10.1103/RevModPhys.81.163
[152] A.L. Cavalieri et al., Nature **449**, 1029 (2007).
[153] S. Neppl. et al., Nature **517**, 342 (2015).

Quasi-particles versus the one electron picture in solids

The structure of a covalently bound solid is a complicated system consisting fundamentally of interacting electrons and ionic cores. The main properties of these solids depend mostly on their electronic structure determined by the laws of quantum mechanics. The energy eigenstates, or more generally the dynamics of the huge number of interacting electrons are described by an extremely complicated many particle entangled quantum state. In many cases it turns out, however, that the behaviour of these particles can be understood -- at least approximately -- as if any of them had an individual quantum state, that is why a single particle (or independent particle) approximation picture works in several cases: e.g. one-electron band theory reproduces well the properties of simple metals. This was explained more deeply by Landau's Fermi liquid theory, which stated that in spite of the definitely collective properties, the low energy (elementary) excitations of a many electron system behave very much like independent particles, they carry the same charge and spin as electrons, but have an effective mass which is different from that of a free electron.

These types of excitations called quasi-particles[154] emerge in all many-body systems, and it is important that they have only a finite lifetime. In metals these quasi-particles are either the electrons themselves, or plasmons which are collective excitations in the many body density of the electrons. In dielectrics and in semiconductors the relevant quasi-particles are excitons, which is again a concept to replace the complicated many body excited state of the system by an independent particle, by an electron-hole pair. Here one thinks of an electron elevated to the conduction band, and the redistribution of the remaining electrons is considered as if a hole was created in the rest of the sea of electrons. If in addition some of these quasi-particles are coupled to an electromagnetic wave in the medium, one usually speaks of a polariton. These, and other quasi-particles like magnons (spin waves) and polarons (polarization waves) are treated usually in the framework of a stationary theory, mainly because their dynamics in the time domain has been out of reach for experiments so far. Time dependent studies with femtosecond pulses of the the spin degree of freedom in ferromagnetic materials[155] [156] and especially in heterostructures have begun only very recently[157]. The theory of their dynamical behaviour is now in an intensively developing stage which goes in parallel with the experimental possibilities opened recently by the developments in femtosecond-attosecond measurement techniques.

Time scales determining the dynamics of collective excitations and quasi-particles are shown in the figure below. Further impetus in the understanding of their properties and new discoveries in this field are expected from measurements to be done with the facilities of the ELI ALPS laboratories, which shall be ideally suited for such experiments opening the subfemtosecond range, where quantum coherences of all these excitations are maintained, and dissipation effects play no role any more. This means that besides studying the collective excitations themselves, the investigation of the explicit time dependence of their decoherence mechanisms will become possible.

[154] Bovensiepen U, Petek H, Wolf M, Eds. *Dynamics at Solid State Surfaces and Interfaces*, Vols. 1 and 2, Wiley VCH (2010)
[155] Chan La-O-Vorakiat, et al., Phys. Rev. Lett. **103**, 257402 (2009)
[156] Bigot JY, Vomir M, Beaureaire E, Nat. Phys. **5**, 515 (2009)
[157] Kampfrath T, et. al. Nature Nanotechnology **8**, 256 (2013)

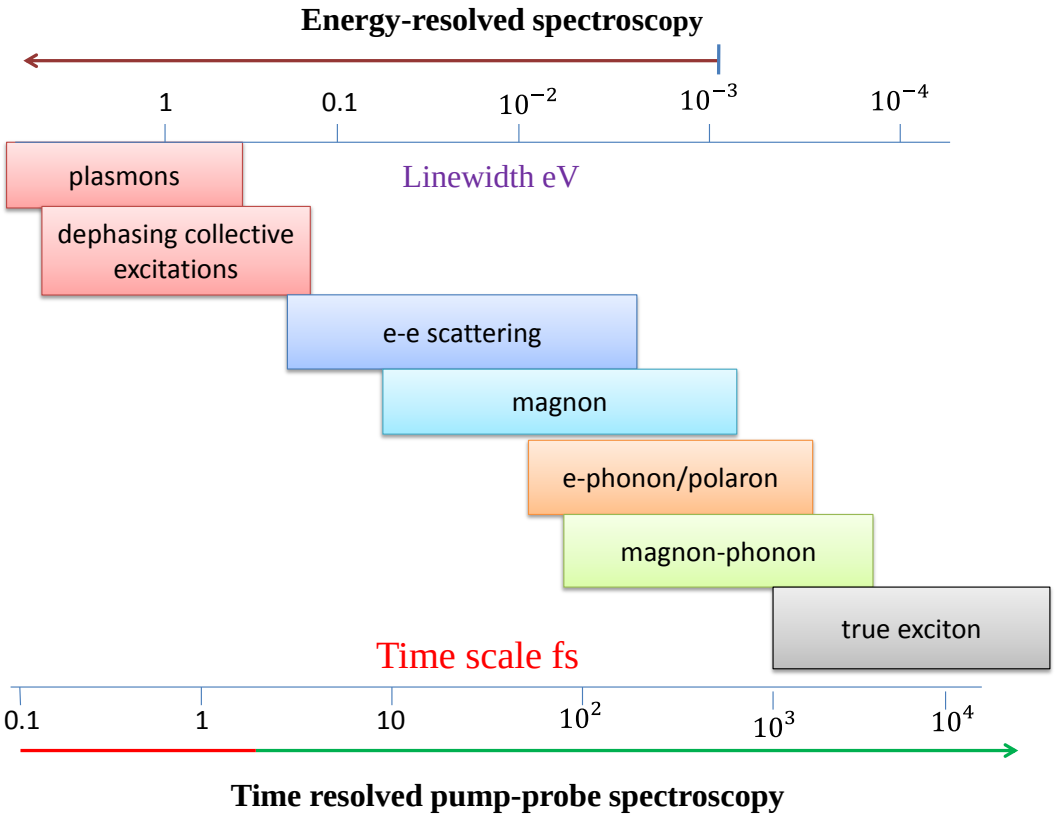


Fig 51 Energy and time scales of quasiparticles and collective excitations. Adapted from Ref [158]

Electrons from metals

Single electron emission

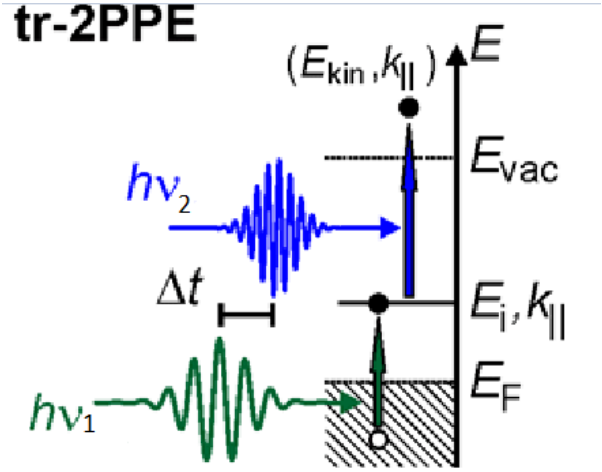
An excitation that leads to electron emission from a metal is a complicated process, and a number of various mechanisms will determine the actual dynamics[159]. (i) The ionic background of the lattice will change the group velocity of the electrons starting their motion. (ii) As a consequence of electron ejection a positively charged hole is created in the sample to be screened by the rest of the electrons remaining in the metal. (iii) Inelastic scattering of the ejected electrons with other ones will determine the depth from which the ejected electrons can reach the surface and thus carry direct information on the processes in the bulk. (iv) The normal component of the laser field decreases in the bulk abruptly to a very small value. The first experiments aiming to resolve the time dependent motion of a photoelectron from a metallic surface by *fs* pulses have begun less than 10 years ago. In one of these experiments[160], a Pt surface was simultaneously irradiated by XUV and intense IR pulses, and conclusions about the photoemission process could be drawn by analysing how the infrared laser modified the XUV photoelectron spectrum. In the first time-resolved photoemission experiment[161] photoelectron emission was observed from a tungsten surface, which was triggered by a pulse of 300 *as* duration with 95 *eV* photon energy. This XUV pulse was accompanied again by an additional near infrared (NIR) 750 *nm*, 5-*fs* pulse. The energy acquired by the ejected electron from this second field depends on the time of the electron passage across the metal surface. Thus, measuring the dependence of the ejected electron energy spectrum on the time delay between the

[158] Petek H, Ogawa S, Prog. Surf. Sci. **56**, 239 (1997)
[159] Kazhansky AK, Echenique PM, Phys. Rev. Lett. **102**, 177401 (2009)
[160] Miaja-Avila L, et al, Phys. Rev. Lett. **97**, 113604 (2006) doi: 10.1103/PhysRevLett.97.113604
[161] Cavalieri AL, et al, Nature **449**, 1029 (2007)

pulses, one can keep track of the process in the time domain; this is the idea of the "streak camera", mentioned earlier. With this technique a time delay of 110 *as* in the emission of the 4f core electrons of tungsten was measured compared to the ejection of conduction electrons from the Fermi edge. With the facilities of ELI ALPS this streaking technique can be extended, and provide a much better resolution if the second pulse will be similar to the first, with a precisely synchronized delay in the timing of the two pulses. In this way the femtosecond-attosecond technique will be replaced by an *attosecond-attosecond* one, and should become a standard tool for time-resolved investigation of photoemission from solids.

Time resolved multi-photon photoemission

One of the methods of getting information about the ultrafast excitations and their decay in a solid is time resolved two-photon photoemission (tr-2PPE), see the figure below.



It is a pump-probe method and it is appropriate for metals and dielectrics, as well. Two pulses of different frequencies are applied to excite and probe unoccupied states between the highest occupied level (Fermi level) and the vacuum level. The first pulse populates the intermediate state of energy E_i while the second one generates the actual photoemitted electron from this intermediate level. The energy of the latter as the function of the delay between the individual pulses provides information about the lifetime of the excitation. Pulses with photon energies of a few *eV*-s in the visible and in the UV regime are sufficient to cover the energy range in question, as the value of the work function $E_{vac} - E_F$ is of the order of 4-5 *eV*-s. Similar experiments on magnetic materials could be especially interesting, where spin dependent density of states could be mapped.

The method can be extended to measure the full band structure as in the photoemission process the momentum of the electron is conserved, due to the negligible photon-momentum in the visible range. The momentum components of the electron can be determined by resolving the angular distribution of the electron energies with a method called angularly resolved photon emission spectroscopy (ARPES).

A natural extension of tr-2PPE would be tr-*n*PPE with *n*, i.e. with more than two photons with variable delays. Various multiphoton experiments without time resolution have been performed so far on metal surfaces. They enlarge the energy range and provide further information on the photoexcitation process, but require more intense optical fields. The possibility of extending these band mapping methods will be provided by ELI-ALPS with its short and intense pulses in the femtosecond-attosecond range, allowing to observe coherent evolution of electron states in detail.

Dielectrics

Insulators or dielectrics are materials where (in the one electron picture) the valence band -- containing the highest energy electrons -- is full, and an energy gap, much larger than the thermal energy at room temperature, separates this band from the next empty band above it. SiO_2 being one of the best insulators has e.g. a band gap as large as about 9 *eV*. In the first experiment[162] where a valence band-conduction band transition was observed in some transparent dielectric materials, a secondary optical signal resulting from the emerging attosecond transition dynamics could be recorded.

In a more recent experiment[163], the resulting electric current has been directly observed, so it was proven, that the material can be transformed into a conductor for a very short time by a 4 *fs* NIR pulse with a peak electric field of 20 GV/m. In order to expose a sample to such an intense laser field without destroying it, the medium must be transparent, so that little energy should remain in the medium after the interaction with the pulse. This implies that the laser photon energy should be much smaller than the band gap. It took more than five laser photons that the near IR pulse bridged the large band gap of SiO_2. This few-cycle optical wave reversibly increased -- free from breakdown -- the AC conductivity of the material by more than 18 orders of magnitude within a few femtoseconds. In the experiment of Schiffrin et al. an AC current was induced in the material and a net charge transfer could be registered between two gold electrodes on the surface of the silica sample. Furthermore, it has been found that the transferred charge could be steered by controlling the carrier--envelope phase (CEP) of the laser pulses. The findings of this experiment open the possibility of switching electric current at rates 10^{15} cycles per second. In order to make use of this effect in technology, both the interaction of the light field with the exciton, and the relaxation mechanisms which settle the final charge transfer in the process must be well understood. In a closely related work[164] attosecond absorption spectroscopy and wave-cycle-resolved optical reflectivity measurements provided information on the inverse effect, how the induced electron current influenced the optical properties of the material on the subfemtoscond time scale.

A phenomenological model of the electron-phonon interaction which influences the actual behaviour of the oscillating current and the resulting charge difference between the electrodes can be found in[165]. In order to understand the role of the electron-phonon coupling on this very short time scale further experiments with generating and switching currents in the femto- and attosecond range are necessary. When the external electric field strong enough, then, in spite of scattering, the electron can acquire a crystal momentum to reach an edge of the first Brillouin zone. In the case of such a strong oscillating field Ref. 18 predicts so called dynamical Bloch oscillations and Wannier-Stark localization[166], which are still to be confirmed by probing valence band-conduction band transitions with subfemtosecond resolution.

ELI-ALPS will certainly yield an excellent opportunity for such technologically important investigations, where one would exploit the electric field of laser light to control the flow of electrons in dielectric materials, and switch currents in electronic circuits at light frequencies, which, in turn, may modulate the transmitted light.

Molecular orbital and electron transfer dynamics on surfaces

As has been recently demonstrated (see figure below), angle-resolved photo-emission from molecular crystals allows the reconstruction of static molecular orbital densities that are in good agreement with DFT calculations[167]. Using the ELI-ALPS facilities it will become possible to perform these experiments in the time-domain and to observe and monitor in real-time how molecular orbitals adapt under the influence of incident radiation. These experiments greatly

[162] Mitrofanov A, et al., Phys. Rev. Lett. **106**, 147401 (2011)
[163] Schiffrin A, et al., Nature **493**, 70 (2013) doi:10.1038/nature11567
[164] Schulze M, et al., Nature **493**, 75 (2013) doi:10.1038/nature11720
[165] Földi P, Benedict MG, Yakovlev VS, New J. Phys. **15**, 063019 (2013), doi:10.1088/1367-2630/15/6/063019
[166] Apalkov V, Stockman MI, Phys. Rev. B **86,** 165118 (2012)
[167] P. Puschnig et al., Science 326, 702 (2009).

benefit from the high repetition rate and photon energy range that will be accessible in the ELI-ALPS attosecond sources.

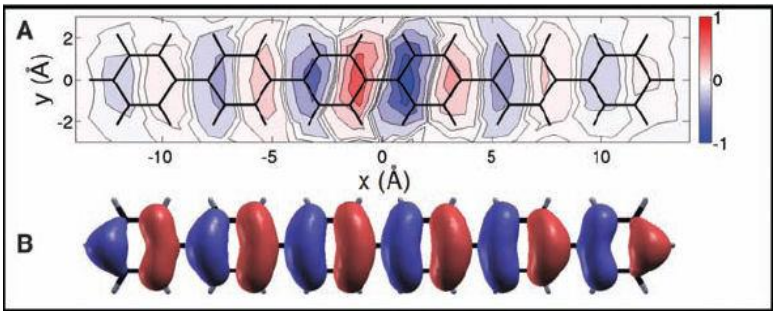


Figure 52. Comparison between experimentally determined orbital densities for the sexiphenyl molecule and theoretical results obtained by DFT

Besides these experiments which address the electron densities in pure crystals and/or molecular assemblies, the ELI-ALPS facilities will also be uniquely suited for the investigation of electron transfer processes on surfaces, allowing to confirm and significantly extend the work that is thus far carried out at synchrotron facilities using the core hole clock method where, for example, electron transfer times from absorbed sulphur atoms to a ruthenium substrate as short as 320 attoseconds have been inferred[168].

Ultrafast nanoscience and technology

Localized optical near fields on the nanoscale play an important role in many contemporary applications in scanning probe microscopy, enhancement of photovoltaic elements, sensorics, highly sensitive spectroscopic methods and many more. The common element in these approaches is that they involve and exploit so-called surface plasmons that are localized or propagating charge density oscillations bound to a metal surface. The quasi-free electrons move against the restoring force of the ionic cores, these are called plasma oscillations. Their angular frequency in the bulk is $\omega_p=(n_e e_0^2/\varepsilon_0 m_{ef})^{1/2}$, where n_e is the electron density and m_{ef} is the effective mass of the electrons in the metal. For gold e.g. $\omega_p=1.36\text{x}10^{16}$ 1/s which falls into the UV range. Due to the finite skin depth of electromagnetic fields in this domain, observable plasmonic effects arise only at the surface of the sample. The nonequilibrium electric charge density created by the external field becomes a source of a secondary near field in the vicinity of the surface. This field -- which is rapidly decaying with the distance away from the surface -- can be more than 10-100 times stronger than the incident generating field. These particular modes of the coupled motion of the charge inhomogeneity and the field generated by them are known as surface-plasmon polaritons but often shortened as just surface plasmons. The lifetime of a surface plasmon is around 10 *fs*, while due to the phase sensitivity of the electron wave with respect to any disturbance, the coherent lifetime of a plasmon is estimated to be around 100 *as*. Because of the short lifetimes, the only way to study and control plasmons is to apply subfemtosecond sources to be delivered in ELI-ALPS: To understand the mechanism of the generation of surface plasma waves and their relaxation needs extensive experimental and theoretical studies. Light scattering as well as angularly resolved photemission spectroscopy (ARPES) from such structures should reveal important information on the plasmon properties.

Propagating surface plasmons (PSPs) typically run along a metal thin film while localized surface plasmons (LSPs) are charge density oscillations of metal nanoparticles. They both can be efficiently induced by visible laser pulses and come together inherently with i) ultrahigh localization of the electromagnetic field of light to nanoscale volumes (along one or all spatial

[168] A. Fohlisch et al., Nature 436, 373 (2005).

dimensions) and ii) substantial enhancement of the electric field amounting to as much as several hundreds [169]. Fig. 1. depicts a resonantly excited LSP on a metal nanosphere.

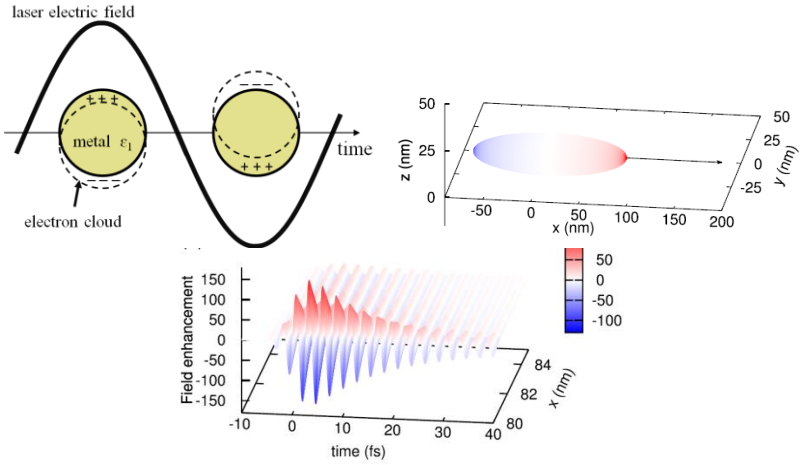


Fig. 53. Left panel: resonant excitation of a localized surface plasmon on a metal nanosphere by a laser beam. Right panel: electric field distribution on a resonantly excited ellipsoidal plasmonic nanoparticle. Bottom panel: temporal and spatial evolution of the plasmonic near-field on a resonantly excited ellipsoidal nanoparticle along the long axis[170].

It is of special interest that in metallic nanoparticles -- whose dimensions are smaller than the skin depth -- electromagnetic fields can be localized in a modal volume of the order of 10 $(nm)^3$, which is the size of the particle itself. This volume is about six orders of magnitude smaller than the cubed half-wavelength of visible light, the size to which the field of an electromagnetic wave can be constrained in vacuum. We note that this local, short range and strong field is responsible for the important effect of Surface Enhanced Raman Scattering (SERS), that makes plasmonic structures to be very sensitive detectors of certain molecules adsorbed on the surface of a nanoparticle[171] [172]. In addition to that, ordered two-dimensional periodic plasmonic structures have been fabricated and their unprecedented fast response to optical fields at a very large bandwidth seems to offer a tremendous potential for circuitry, for optical switching and possibly for optical memory[173]

In light of the numerous applications of plasmonic nanosystems, ELI-ALPS can offer several unique possibilities to test their fundamental properties and optimize them for various real-life scenarios. Plasmonic field enhancement together with the high repetition rate of the HR source (and its secondary sources) is an unparalleled opportunity to perform time-resolved experiments on plasmonic phenomena with attosecond precision. This possibility is currently not available anywhere else. With attosecond tools, it will become feasible to directly test the build-up and decay and of surface plasmon oscillations and once time-resolved information is available, one can also optimize nanoarchitectures for the spatiotemporal control of these phenomena on unprecedented scales. The secondary near field distribution on the nanoscale can be precisely controlled with the shape of the nanoparticle evoking an interesting connection between nanoplasmonics and ultrafast photoelectron emission. The field enhancement due to the near field of the charge distribution in the metal particles increases the electron emission probability from the surface and causes a strong initial acceleration to the emitted electrons. They are found to escape along straight trajectories with orientations governed by the particle geometry. Measurements of

[169] J. A. Schuller et al., Nat. Mater. 9, 193−204 (2010).
[170] P. Földi, I. Márton, N. Német, V. Ayadi, P.Dombi, Appl. Phys. Lett. 106, 013111 (2015).
[171] Kneipp K, Phys. Today, p. 40 Nov. 2007.
[172] Stockman M, Phys. Today, p. 39 Feb. 2011.
[173] Ebessen T, Genet C., Bozhevolny SI, Phys Today, p. 44 May, 2008.

the electron energy spectrum from an ordered array of gold nanorods (typical dimensions 160×80×40 nm³) have been performed[174] with pulses of duration in the 100 *fs* range. Correlation between the surface plasmon resonance condition, tuned by the size and shape of the particles and the cut-off in the electron spectra have been observed. In the experiments performed so far the excitation has been realized either with relatively long, multicycle laser pulses or with shorter few-cycle femtosecond pulses, but without carrier-envelope phase control. The opportunities provided by ELI-ALPS with phase-stabilized few-cycle pulses will allow one to go down to the femtosecond or subfemtosceond range, and may lead to observe predictable steering of electrons on the nanoscale[175]. One expects to establish quantitative correlations with the optical spectra of plasmonic structures, and thereby to understand the real time dynamics of the plasmons themselves. One proposed method for this is attosecond near-field microscopy with a modified version of a photoelectron emission microscope (PEEM)[176]. The goal is to resolve the evolution of plasmonic near-fields in time. Even without the attosecond sources of the facility, one has the chance to characterize the distribution of plasmonic near-fields with state-of-the-art photoelectron spectroscopic tools[177] with an unprecedented resolution. This requires the proper adaptation of PEEM and/or velocity map imaging (VMI) tools of the facility for this purpose and using high repetition rate femtosecond laser sources due to the relatively low yield of rescattering photoelectrons necessary for this application.
Very recently an interesting experiment has been performed[178] hinting at the possibility of electron pair formation assisted by surface plasmons. The phenomenon was observed in gold films with exciting laser pulses of duration of 100 *fs*. As this value is longer than the lifetime of the plasmon, one might assume that the attraction between the electron pairs is initiated by the plasmon field, but to investigate the detailed mechanism of this possible effect needs shorter pulses with controlled and variable strength. High energy and faster laser pulses to be delivered in the ELI ALPS will allow to continue such experiments with a much better temporal resolution. Plasmonically enhanced near-fields are also believed to play a significant role in the conversion of laser light to various other types of radiation from the THz[179] to the XUV domain[180]. Mechanisms responsible for the observed phenomena are still debated in the literature and attosecond tools are expected to shed light on the underlying phenomena. Once these are known, nanoarchitectures can be optimized for various frequency conversion schemes in the facility.

In terms of applications of ultrafast plasmonic technology, ELI-ALPS will be particularly suitable for focusing on the field of so-called lightwave electronics. The limitation of current electronics circuitry to the multi-gigahertz range is caused by fundamental bottlenecks in their architecture. In order to achieve a paradigm shift in the design of nanoelectronic devices, one needs to use tailored ultrafast light fields to control electron currents in all-solid-state nanocircuits. An ideal lightwave-controlled nanocircuit accepts data as light and in the form of plasmons which travel at speeds literally approaching the speed of light. Furthermore, the highly non-linear response of nanomaterials to strong laser fields permits the switching of light and currents with frequencies approaching the PHz domain. With the help of novel, ultra-broadband light sources of ELI-ALPS, one expects to be able to control suitably designed nanoarchitectures and later on, build ultrafast switches. In order to characterize and control these structures, one needs a combination of ultrabroad and/or attosecond sources with high repetition rate together with novel photoelectron detection tools. This instrumentation

[174] Dombi P, et al., Nano Lett. **13**, 674 (2013), dx.doi.org/10.1021/nl304365e
[175] Földi P, et al., Appl. Phys. Lett.**106**, 013111 (2015), dx.doi.org/10.1063/1.4905464
[176] M. I. Stockman, M. F. Kling, U. Kleineberg, F. Krausz, Nature Phot. 1, 539 (2007)
[177] S. Thomas, M. Krüger, M. Förster, P. Hommelhoff, Nano Lett. 13, 4790 (2013).
[178] Kroó N, Rácz P, Varró S, Europhys. Lett. **105**, 67003 (2014), dx.doi.org/10.1209/0295-5075/105/67009
[179] D. Polyushkin, E. Hendry, E. K. Stone and W. L. Barnes, Nano Lett. 11, 4718 (2011).
[180] [6] I-Y. Park et al., Nature Phot. 5, 677 (2011).

background is expected to be available at the ELI-ALPS facility and after developing suitable methodologies, we expect to demonstrate proof-of-principle nanoarchitectures of signal processing in close collaboration with leading nanofabrication facilities.

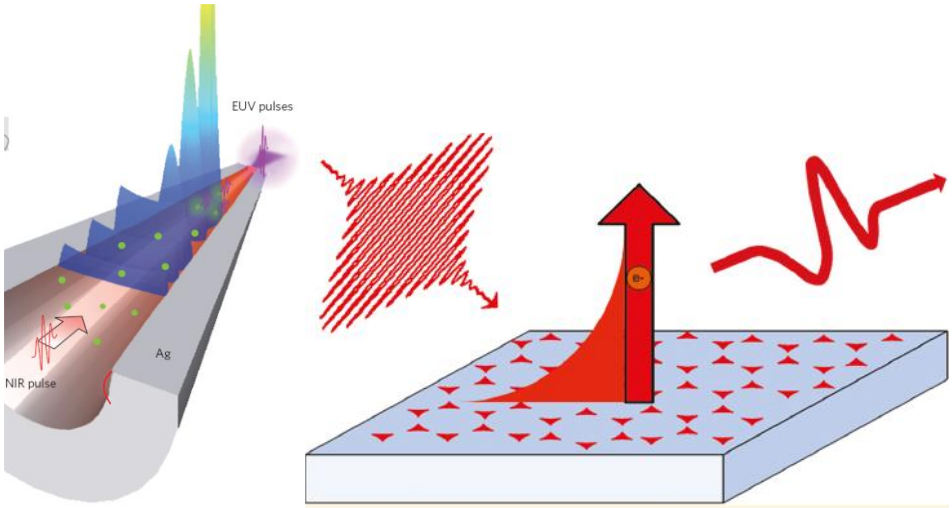


Fig. 54. XUV (left panel) and THz (right panel) conversion schemes with ultrashort near-infrared laser pulses interacting with plasmonic nanostructures.

In summary, the ELI-ALPS facility is expected to shed light on a number of unresolved questions about the dynamics of plasmonic oscillations with an unprecedented spatiotemporal resolution. Once known, tools shall be developed to control these phenomena in order to gain better access to already existing applications of plasmonics in various areas. There are also some unprecedented directions including, for example, the construction of ultrafast nanointegrated plasmonic circuitry where the unique primary and secondary sources of the facility together with state-of-the-art end-stations can bring substantial advances for pioneering technologies.

Strongly correlated materials

Many aspects of the electronic structure and conductance properties of simple metals and semiconductors like Si can be relatively easily interpreted in terms of one particle band theory. In contrast, *strongly correlated materials* (SCM) represent the class of solids, where the usual one-electron picture loses its validity. As the explanation of their unconventional properties should contain more advanced quantum many body principles, it has become customary to call such substances as *quantum materials*. There is a wide variety of compounds that belong to this category: transition metal oxides exhibiting metal-insulator transition, cuprate based high temperature superconductors, the recently discovered iron-based superconductors (FeSC), topological insulators, multiferroics, magnetic substances, etc., each providing a rich research field for fundamental understanding of correlated electron systems. In spite of their promising future technological applications, to obtain a satisfactory explanation of their fundamental properties has been a long lasting and in many respects a still unsolved challenge for condensed matter physics.

Traditionally the behaviour of these materials has been investigated in the static regime, and the aim was to characterize their thermodynamic phases by changing the temperature, as well as by varying the pressure or the original stochiometry through chemical substitution of isolectronic elements. Quite recently several research groups initiated the study of time dependent dynamics of excitations in SCM by ultrashort laser pulses. As it turned out, femtosecond laser pulses can selectively excite modes of strongly correlated electron systems and controllably push materials

from one ordered phase to another[181]. The knowledge of reliable dynamical phase structures should be utilized for ultrafast and sensitive (thermodynamic) phase control via pure photonic channels, free from thermal effects.
We mention now a few of such materials and studies.

A photoinduced insulator-to-metal transition (IMT) is a "sudden" collapse of the band gap present in the insulator phase, which becomes "filled in" by available electron states as a consequence of a laser pulse. There are a few types of such materials. One of them is a so called *Peierls insulator*, where the IMT mechanism is connected with electron-phonon coupling. The characteristic times for such transitions have been measured in the 100*fs*-1*ps* range, due to the large ionic masses that play a role here. Starting from the low temperature insulator phase the photoinduced transition to the metallic state is followed by the equilibration of the electron and phonon subsystems. The reverse process, the metal-to-insulator transition takes place spontaneously below the transition temperature as a result of an instability of the ionic background, which leads to a symmetry breaking Charge Density Wave (CDW) in the insulator phase.

In the case of *Mott insulators* the gap is the consequence of electron-electron repulsion, and as being a pure electronic process, the characteristic time of IMT is estimated here to be around 1 femtosecond or below. As it results purely from electron correlation a Mott transition has not been resolved so far due to the present experimental limitations. ELI-ALPS will offer the possibility to overcome this limit and yield the possibility of a deeper understanding of this paradigmatic process. The return to the equilibrium (Mott insulator) state is a relatively slower process, as it is mainly due to electron-phonon interactions. In an experiment[182] using femtosecond pulses several such channels have been explored in the Mott transition of the layered material TaS_2.

Another kind of photoinduced insulator-metal transition can take place in certain organic materials called charge-ordered insulators where coherent oscillations of the correlated electrons and their coupling to intramolecular vibrations could be observed in the time domain with resolution in the 10-50 *fs* interval[183]. As shown by these and other experiments, the changes in the structural and electronic properties of the materials are strongly correlated and due to the differences in the relevant degrees of freedom (electron, phonon etc.) there are several mechanisms playing role in such transformation. The fastest among them is the motion of the electrons, and while the dynamics of the vibrations could be revealed, the finest and most definitive role played by electronic motion was still beyond the reach of experiments.

The mechanism of the creation of Cooper pairs in high-T_C cuprate or iron based superconductors is still an undecided issue, after more than two decades of research. Time and angularly resolved photoemission (trARPES) studies have been the subject of experiments in such materials in the last few years[184] [185] [186]. The applied laser pulses were in the hundred femtosecond domain, where mainly the destruction of the superconductive pairs could be studied, as it is assumed to be caused by electron-phonon interactions. It is very probable that the formation of the pairs is an effect mediated by the specific correlation due to the presence of *d-bands* in these materials. Being a purely electronic process, this mechanism should be much faster than phonon mediated phenomena. However, such correlated electron dynamics immediately after the photoexcitation have remained hidden, because the time resolution is not sufficiently high to detect the electronic degree of freedom before it is significantly affected by the phonon motion. Therefore the final word on this question can be reliably expected to be given only by experiments that can go below the femtosecond range , to be provided by ELI-ALPS.

[181] Orenstein J, Phys. Today p. 44 Sept. 2012; doi: 10.1063/PT.3.1717
[182] Perfetti L, et al., New Journal of Physics **10** (2008) 053019
[183] Kawakami Y, et al., Phys Rev. Lett. **105**, 246402 (2010)
[184] Cortes R, et al. Phys Rev. Lett. 107, 097002 (2011)
[185] Smallwood CL, et al. Science 336, 1137 (2012)
[186] Rameau JD, et al. Phys Rev B 89, 115115 (2014)

Let us also note here that in a different time domain strong-field single-cycle terahertz pulses have been used[187] to gate interlayer coupling in a high T_C cuprate based layered superconductors $La_xSr_{1-x}CuO_4$. In-plane superconductivity remained unperturbed, revealing a non-equilibrium state in which the dimensionality of the superconductivity was time-dependent and the gating frequency is determined by the electric field strength. Such non-dissipative, bi-directional gating of superconductivity is of interest for device applications in ultrafast nanoelectronics and is a possible research field to make use of the THz source of ELI-ALPS.

As we see, the experiments performed so far with SCMs demonstrate that there exist transient phases which can be achieved by photoexcitation and thus transforming matter to states not accessible by manipulating simple thermodynamic parameters. Exploration of such short living phases are now restricted to a temporal resolution at best around a few tens of femtoseconds or longer, while theoretical models predict the *subfemtosecond* scale as relevant from the point-of-view of the essential physical effects in these materials. Calculations of the density fluctuations of strongly correlated electron systems under the excitation of a pulse of a few *tens of attoseconds* have been performed by genuinely many body methods[188], and a clear distinction could be made when contrasting it to one-electron uncorrelated calculations.

The possibilities of the ELI-ALPS with its *subfemtosecond* pulses will enable its users to reveal these finest details of electronic dynamics, and will help to understand the processes of controlled modification of quantum materials which -- beyond their high scientific interest -- are expected to be exploited in future ultrafast optoelectronic devices.

THz-driven dynamics in strongly correlated electron systems

Materials that exhibit strong correlations between electrons can exhibit many emergent phenomena due to the failure of the independent electron picture. These phenomena often manifest themselves in the form of exotic and sometimes highly useful properties. Probably the most well-known example of such a property is high-temperature superconductivity in cuprates, but other examples include colossal magnetoresistance, orbital and charge ordering, multiferroicity and unusual metal-insulator transitions. The microscopic mechanisms for these phenomena are a strong and persistent challenge for both experiments and theory, largely due to the strong competition among many different degrees of freedom operating on similar energy scales.

In many cases the interesting emergent properties can depend on a particular kind of interaction between different degrees of freedom that are normally considered as separate. An example would be the strong coupling of the lattice and charge that strongly influences many of the unusual valence ordering in manganites[189]. To understand such interactions on an experimental level, pump-probe methods offer considerable potential. The idea here is to selectively drive dynamics in one degree of freedom (e.g. exciting a particular vibrational mode of the lattice) and observe the system as it evolves in time to see how the energy deposited in that part of the system couples to other parts. One of the main difficulties in actually performing such experiments is a lack of selectivity in experimentally accessible, short-time-duration perturbations as well as a lack of selectivity in ultrafast time-resolved probes.

Both these problems can potentially be overcome using ELI-ALPS, which offers a wide spectrum of ultrashort pulses form the THz to the x-ray frequency range. This is highly attractive since THz and mid-IR pulses can in many systems selectively drive phonon and magnon resonances, which typically range from about 1 THz to 30 THz. Complementary to this is the ability to use synchronized ultrashort soft and hard x-ray pulses as a probe of the resulting dynamics of long range order, which is a highly selective probe of particular valence properties. Very recently several

[187] Dienst A, et al, Nature Photonics 5, 485--488 (2011),1038/nphoton.2011.124).
[188] Kwong NH, Bonitz M, Phys Rev. Lett. 84, 1768 (2000).
[189] E. Dagotto. Nanoscale Phase Separation and Colossal Magnetoresistance (Springer, 2003).

proof-of-principle experiments have demonstrated the feasibility of these kinds of experiments, currently performed at x-ray free electron lasers[190].

Figure 55 shows a generic schematic for such a pump-probe measurement that would take advantage of the wide spectral range of ultrashort pulses availalble at this facility. On the pump side of the experiment, recent theoretical and experimental investigations have suggested that several strongly correlated systems of interest show a dramatic sensitivity to large-amplitude excitations of infrared-active phonon modes[191]. One prominent example is the recent claim of enhanced superconductivity in cuprates from mid-IR excitation[179]. Furthermore, the magnetic field of the EM pulses also offer the possibility for more direct excitation of spins[192].
On the probe side, the range of x-ray frequencies covered by ELI-ALPS includes the transition metal L-edges for photon energies between 500 eV and 1000 eV. Resonant scattering at these edges allow for direct and quantitative measurements of the long range order of many different

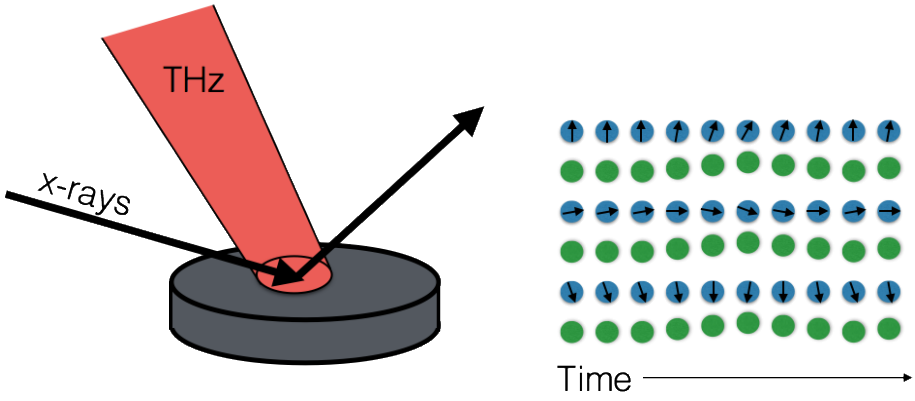


Fig. 55: Concept of a possible experiment to drive a vibrational mode with THz radiation and to observe the resulting dynamics of spin with resonant x-ray diffraction. The left shows the experiment: a few-cycle pulse resonant with a vibrational mode excites an infrared-active phonon mode in the material. As a result, the lattice distorts coherently with time. At various points along this distortion, an x-ray pulse resonant with a core level transition selectively probes the long-range spin order of the system. The result is a quantitative measure of the coupling between that particular vibrational mode and the spin dynamics.

valence properties, including spin, orbital and charge orders [9]. When combined with the selective pumping offered by the THz and mid-infrered pulses, soft x-ray scattering offers a unique look at how different kinds of structures within a material can interact.

ELI-ALPS offers a unique combination of light sources that enable these experiments: high pulse energies over a wide frequency range with precision synchronization. The fact that these sources derive completely from optical paths makes the inherent synchronization in principle superior to that offered by SASE free-electron-laser facilities, where some level of timing jitter is unavoidable. In addition, the highly limited access to free-electron lasers also makes ELI-ALPS an attractive facility for these types of measurements

[190] T. Kubacka et al. Science 343, 6177 (2014); P. Beaud et al. Nature Materials 13, 923 (2014); M. Först et al. Phys. Rev. Lett. 112, 157002 (2014).
[191] M. Först et al. Nature Physics 7, 854-856 (2011);D. Fausti et al. Science 331, 189-91(2011);R. Mankowsky et al. Nature 516, 71 (2014).
[192] T. Kampfrath, K. Tanaka & K. A. Nelson, Nature Photonics 7, 680-690 (2013)

Outlook and future implications

The advantages of the ELI-ALPS facilities are manifold. The high repetition rates, high photon fluxes and ultra short pulse duration of the ElI-ALPS sources allow (i) massive increases in the signal-to-noise ratios of atomic experiments, thereby allowing to shed light for the first time on the role of electron correlation in atomic ionization experiments as well as to ultrafast dynamical processes of charge migration in large molecules, (ii) understanding light-matter interaction on a field↔electron level. Historically, one of chemistry's most fundamental questions was solved by time-resolved imaging: the making and breaking of bonds. A similar impact is now foreseen for physics. Such insight will provide a long-sought basis for technologies to come, be it electronics at lightwave frequencies, design of advanced materials, molecular motors, switches, photonic crystals, biological nano-machines or organic solar cells. Although the research presented in this chapter is regarded as fundamental science, its importance in industrial and medical applications over the next decade is clearly foreseeable (iii) opening up the so far unreachable field of inner shell non-linear processes at highest spatiotemporal resolution, in particular thanks to the substantial increase of intensities combined with the shortest ever durations of energetic of the XUV/x-ray pulses (iv) the use of sophisticated correlation spectroscopies in attosecond time-resolved studies of molecular electron dynamics, thereby setting the stage for studies of charge-directed reactivity in molecules and the realization of the long-standing goals of optical control of molecular reactivity, (v) will have far reaching consequences in plasma physics not only to fundamental physical questions but also to concrete applications. The unique combination of advanced technological development will open up the prospects to original and path breaking advances in science with tangible implications to societal role of the resulting knowledge. For example issues relevant to particle acceleration for medical applications or to better understanding of the approach towards inertial fusion energy are only a few of these to be mentioned. (vi)The wide-spread use of the attosecond technology at high repetition rates on surfaces and condensed matter towards studies of molecular orbital dynamics on surfaces, electron dynamics at interfaces, plasmonic structures,spin/magnetization dynamics, dynamics of correlated complex systems areas where synchrotron studies have already clearly suggested the promise of attosecond time-resolved measurements, but where the main promise of attosecond exploration remains to be fulfilled as one of the key areas where the ELI-ALPS facility can have a major impact. These research areas are of highes technological interest as e.g. in IT applications not only because of the increased speed of control of charge flows but also because of the prospect for large amounts of information transport, due to the nano scale size of the components involved and of the spatial selectivity.

The attosecond sources of ELI-ALPS, in combination with the widely tunable and uniquely controllable wideband primary sources and state of the art end stations allow unique investigations in all states of mater that allow answering the fundamental question to what extent it is possible to control and exploit electronic dynamics on the attosecond and few-femtosecond timescale nanometer or sub-nanometer spatial scale. If successful, the end result will herald the arrival of next generation technology pillars, such as waveform-controlled electronics in the PHz regime, with huge impacts on the future of information processing and transferral, novel energy sources, light harvesting devices, ultrafast nonospintronics.

5 Applied research activities

Motivation, state of the art

With the realization of highly brilliant laser-based X-ray sources with photon parameters (partly) comparable to those of large-scale third-generation synchrotron radiation sources or even fourth-

generation SASE free electron lasers many applications, which are currently run or developed at these large scale installations, can be performed on a laboratory scale in the foreseeable future.

The relevant figure of merit with respect to the source is the *spectral brightness*, defined as the radiated power ΔP per unit solid angle $\Delta\Omega$ and unit source area ΔA (perpendicular to the emission direction) in a given spectral bandwidth $\Delta\omega/\omega$: $B = \Delta P/(\Delta A \times \Delta\Omega \times \Delta\omega/\omega)$
Typical spectral brightness parameters of highly brilliant X-ray sources are given in units of [ph/sec×mm^{2}mrad^{2}0.1%] and reach numbers in excess of 10^{18} for third and fourth generation sources. Brightness is a conserved quantity in lossless optical systems, thus providing very high focused intensities in small focal spot areas, when starting with a small source size and a low divergence source (> high brightness source), which makes it the relevant source figure-of-merit for investigating micro- to nanoscaled samples in spectro-microscopy or micro-spectroscopy experiments.

The envisioned downsizing from large scale facilities to laboratory scale (in spatial size scale as well as in costs) will also open the door to many biological, medical or even industrial applications, which are currently hindered by the limited accessibility of beamtime at international large-scale facilities. Even more, many biological or even medical applications require very specialized laboratory environment (e.g. biological safety hutches, medical patient treatment environment etc.) or dedicated cleanroom environment (e.g. in case of semiconductor industrial applications), which cannot be realized at large-scale facilities, but requires a genuine integration of lab-scaled sources and experiments into the laboratory areas of university (biological), hospital (medical) or industrial sites.

5.1 Biological applications

Research Topics

(i) X-ray microscopy

High resolution nanometer imaging of biological material (organelles, cells, sub-cellular structures) under functional conditions (living or at least in-vitro) is a key technology to understand structure-function relationship in biological soft matter. This includes understanding of cell metabolism, transport across cell membranes, cell-cell communication and many more tasks. Currently, high resolution electron microscopy or advanced confocal light microscopy are state-of-the art techniques to perform imaging of biomaterial down to ~ 1 nm spatial resolution, but require extensive cryo-fixation and/or staining of samples which is known to alter and poison the sample structure. As a complimentary technique, soft X-ray microscopy using short-wavelength radiation in the “water-window” spectral range between the carbon K-edge (4.3 nm) and the oxygen K-edge (2.4 nm) allows for in-vitro imaging of unstained (but usually cryogenically cooled) samples without creating imaging artifacts by staining[193]. Furthermore, soft-X-ray microscopy can be complemented by light microscopy in an integrated dual-mode instrument.

[193] W. Chao et al., Opt. Express 17, 17669 (2009).

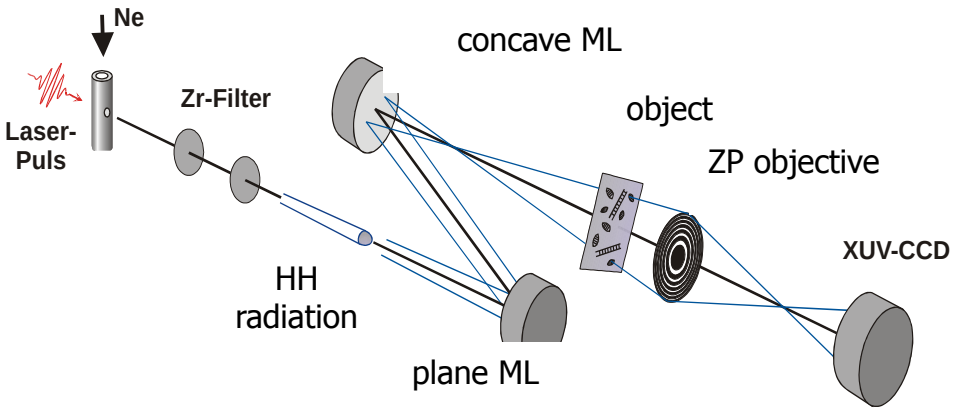


Figure 56 . Principle of an imaging zoneplate soft X-ray microscope using laboratory based coherent high harmonic radiation in combination with a multilayer monochromator and sample images of diatom algae

The Fig. 56 displays an experimentally realized HHG zone plate microscope for full-field imaging microscopy by selecting a single high harmonic order at 93 eV by means of a multilayer monochromator and using an imaging zone-plate with moderate resolution limit (~ 150 nm) for full-field imaging . First test images acquired on dried diatom algae exhibit ~ 200 nm spatial resolution and display the proof-of-concept[194]. Besides cellular imaging structure investigation of biological macro-molecules with sub-Angstrom spatial resolution is of major importance in proteomics and pharmaceutical science. Understanding the structure-function relationship of proteins and enzymes is key to the biochemical understanding of diseases and the development and discovery of new drugs by pharmaceutical industry. Protein hard X-ray crystallography has developed to a powerful tool for structural investigation of macromolecular crystals, while recently new coherent x-ray diffraction techniques (see figure below) combined with oversampling refinement algorithms have been applied to the structural investigation of non-crystallized macromolecules[195]. These new coherent diffractive imaging technologies can be especially useful for structural investigations of membrane molecules, which are hard to crystallize and thus only a fraction of membrane protein structures are known from protein crystallography data[196].

[194] M. Wieland et al., Appl. Phys. Lett. 81, 2520 (2003); M. Wieland et al., Proc. 8th Intern. Conf. X-ray Microscopy, IAP Conf. Ser. 7 (2002).
[195] A. Ravasio et al., Phys. Rev. Lett. 103, 028104 (2009).
[196] M. Bergh et al., Quarterly Review of Biophysics 41, 181 (2008).

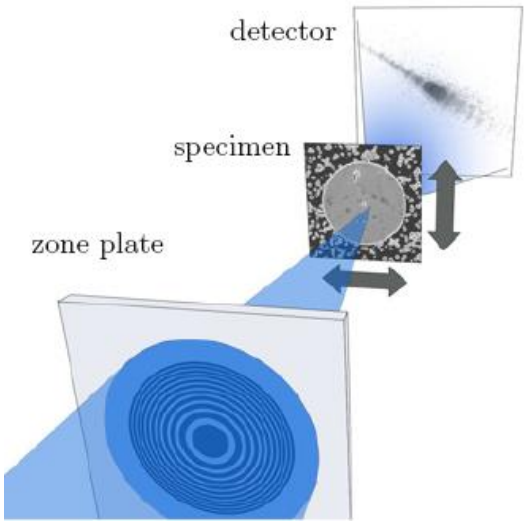


Figure57. Principle of a STXM/CDI experiment[197]

A more detailed description of Coherent Diffractive Imaging for the investigation of macromolecular structure can be found in a separate chapter of this report.

Coherent X-ray beams enable the utilization of phase contrast shadowgraphy or 3D tomography in order to get high-resolution insight into tissue density structure or tumor tissue. While amplitude contrast lacks in signal to noise sensitivity for small tumors (~ 1 mm) and tissue with small electron density variation, phase contrast imaging bears the potential of detecting even pre-cancerous stages of degenerated tissue in a non-metastasized stage[198]. The betatron source has been used to experimentally demonstrate phase contrast imaging with a laser-driven X-ray source.

Furthermore, the realization of a laser-based coherent phase contrast tomography also allows for easy implementation of image-guided radiotherapy, e.g. for the integration of laser accelerated proton or carbon beams.

[197] P. Thibault et al., Science 321, 379 (2008).
[198] T. Donath et al. , Inv. Radiology 45, 445 (2010).

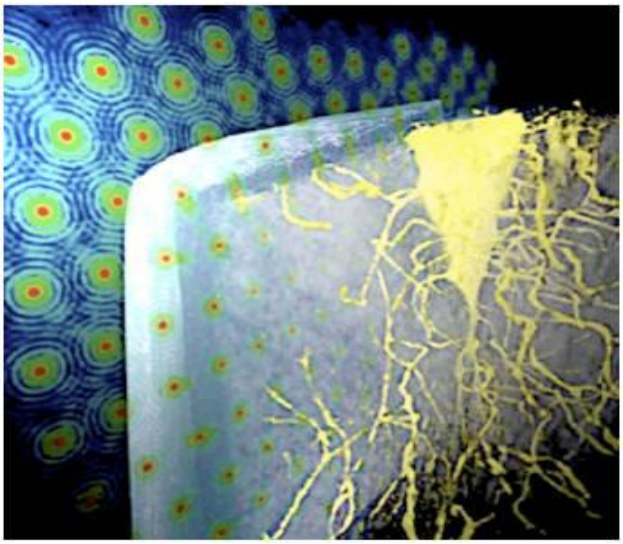


Figure 58. Ptychographic hard X-ray image of bone structure using multiple coherent diffraction images and high-resolution (~ 100 nm) 3D image reconstruction[199]

The mid-infrared spectral region achieved with the planned IR OPCPA primary source is well suited to a large number of medical applications. Bio-molecules have specific absorption lines in the 3-15 μm region and so a well-tuned laser can selectively excite and so detect or destroy molecules. Applications[200] in Laser Dentistry, Laser Angioplasty, Endoscopic Submucosal Dissection and Laser Lithotripsy might benefit from short duration of the MIR pulses as well.

(ii) Serial femtosecond X-ray crystallography (SFX)

SFX is a new approach for structural biology that uses of in vivo–grown crystals[201] to record diffraction data delivered by coherent X-ray pulses from an XFEL. Using the 'diffraction before destruction' principle, diffraction patterns are collected with single, ultrafast pulses that essentially terminate before the onset of significant structural changes occurs, and the X-ray pulse finally vaporizes the sample. The first successful structural investigation of the protein complex up to a resolution of 8.5 Å demonstrates the viability of using nanocrystals and the SFX method for structure determination.[202]

Although the current brightness of the GHHG and SHHG sources are well behind that of provided by XFELs, the adoptation of this very promising method to the forseenable parameters of ELI-ALPS may be an exciting, high risk high gain scientific journey.

(iii) Utrafast electron motions in biomolecules

Charge motions play a crucial role in biological processes – an area to be intensively studied in ELI-ALPS. In a basic experiment it was observed that upon light excitation of the C terminal of a small polypeptide chain, ionic fragmentation occurred on the N terminal[203]. According to model calculations this is caused by correlated electron migration on the attosecond time scale[204]. Such

[199] M. Dierolf et al., Nature 467,436 (2010).
[200] K. Ishii et al., Proc. SPIE 6854, 685418 (2008).
[201] Ijiri et al., Biomaterials 30, 4297 (2009)
[202] Chapman et al., Nature 470, 73 (2011)
[203] R. Weinkauf et al. Journal of Physical Chemistry**99**(28): 11255-11265 (1995).
[204] A. Kuleff et al., Chemical Physics**414**: 100-105 (2013).; F. Remacle, F. and R. D. Levine PNAS **103**(18): 6793-6798 (2006).

fast charge migrations may also occur on DNA molecules[205]. Recently, the phenomenon has been demonstrated on phenylanaline molecules with XUV-pump, visible/NIR probe experiments[206].

(iv) THz absorption and emission spectroscopy

THz spectroscopy has the potential to become a valuable tool for the study of exciton relaxation in pigment-protein complexes. In these complexes the excitation energy is usually delocalized over a number of pigments and it relaxes between such delocalized states towards the low-energy states, which have large contributions from the target pigments, e.g. pigments close to the reaction center, where the energy is ultimately transformed into chemical energy[207]. During the relaxation of excitons (quasi particles that carry the excitation) their excess energy, that is the energy difference between the two delocalized states the exciton relaxes, has to be dissipated by protein vibrations[208]. It turns out that these energy differences are in the THz spectral range. If now there are multiple pathways for exciton relaxation it will be possible to manipulate the pathways of excitons by bringing certain vibrational modes of the protein out of equilibrium by THz excitation. In that case the ratio of absorption and emission of these vibrational modes is changed and thereby the ratio of up- and downward exciton relaxation, which in thermal equilibrium fulfils detailed balance[209], recorded THz radiation from 2D crystalline arrays of bacteriorhodopsin molecules, upon their light-induced ultrafast electron and proton translocation processes, and provided evidence for the role of electron polarization in the initiation of the subsequent proton pump in the protein. Using the same principle, vectorial electrogenic events can be identified in other systems, such as the photochemical reaction centers (RCs) of different photosynthetic organisms or artificial reaction center complexes. Recently, Raoul Freese and coworkers using Langmuir-Blodgett deposition technique, have assembled monolayers of purple bacterial LH1-RC complexes with uniform orientation – a sample which might be suitable for THz spectroscopy[210]. benefiting from the substantially increased THz photon flux offered by ELI-ALPS sources.

(v) The mechanism of charge seperation as studied viaTHz-field induced effects

Charge separation in photosynthetic reaction centers occurs on the femtosecond timescale and the ultrafast electron transfer processes are coupled to protein dynamics – the details of which are not fully understood[211]. Since electric fields affect the processes of charge separation in the reaction centers (and all charge movements in many other biological systems) external electric fields are broadly used for characterizing the change in dipole moment and polarizability for electronicand vibrational transitions[212]. The large electric fields which can be exposed on the sample by THz radiation, and the fact that this can be applied in well defined phase and time-delay with respect to the VIS-NIR light - which can be used either for pumping or for monitoring the charge separated state - provides a potentially unique tool toward the better understanding of the processes in photochemical reaction centers. This can be achieved via using optical pump – THz probe spectroscopy, but it looks equally possible to induce charge separation in the reaction center by intense THz electric field (probably in the range of 0.1 – 1 THz), and monitored via ultrafast absorption and/or fluorescence spectroscopy, thus using THz pump - optical probe spectroscopy.

[205] E. W. Schlag, , et al. PNAS 97(18): 9849-9854 (2000).
[206] L. F. Belshaw, . Phys. Chem. Lett.**3**(24): 3751-3754 (2012).
[207] Y. Shibata et al. J. Am. Chem. Soc. 135: 6903-6914 (2013);H. Van Amerongen et al. Photosynthetic Excitons. World Scientific, Singapore (2000).
[208] T. Renger Phys. Chem. Chem. Phys. 15: 3348-3371 (2013).
[209] G.I. Groma et al. . Proc. Natl. Acad. Sci. USA 105: 6888–6893 (2008).
[210] D. Delgado COST Action TD1102 Book of Abstracts p. 10. April 9-11, 2014, Istanbul.
[211] B. Gobets, R. van Grondelle R. Biochim. Biophys. Acta - Bioenergetics 1507: 80-99 (2001); A. O. Goushcha Biophys. J. 79: 1237-1252 (2000); N. P. Pawlowicz et al. Biophys. J. 95: 1268-1284 (2008); G. Renger T., Renger Photosynth. Res. 98: 53—80 (2008).
[212] S. G. Boxer J. Phys. Chem. B 113: 2972-2983 (2009).

Several groups have synthesized artificial complexes capable for photochemical charge separation[213]), a development which is important for solar energy conversion technologies. Given the large internal and external field strengths, it can be envisioned that these studies on natural and artificial photosynthesis will advance our knowledge on „charge-directed reactivity" of molecules.

(vi) Multiple coherent projections for single-particle imaging

As pointed out in the original document, „simultaneous illumination from multiple directions and with synchronized/coherent pulses could provide a full 3D view of an object and when combined with a synchronized light field for the initiation of dynamics, 4D-structural movies of single particles seem feasible to be obtained". This type of imaging is of great potential interest in biology where motions on the relevant time scale are of functional importance. E.g. in photosynthesis the main light-harvesting complex of plants (LHCII) has been shown to act as a molecular switch between light energy conversion and dissipation[214]. Multiple coherent projections on grana membranes could contribute to the elucidation of the structure and structural flexibility of the semi-crystalline protein arrays.
It would be of great significance to apply laser-induced electron diffraction[215] or time-resolved holography[216] to observe and characterize the structural relaxations following the ionization of macromolecules.

(vii) Pump-probe X-ray absorption spectroscopy

Advanced X-ray spectroscopy techniques are used for the characterization of molecular and electronic structures and dynamics of metalloenzymes with high temporal and spatial resolution[217]. These methods have provided novel insights into the functioning of photosynthetic oxygen evolution and of hydrogenases[218] (Lambertz et al. 2014).The mechanisms and reaction intermediates of some of the key processes in photosynthesis are still unknown. Understanding the elementary mechanisms at work requires direct structural information about very short-lived intermediate states. This is potentially achievable by way of ultrafast time-resolved X-ray spectroscopy, such as optical (laser) pump – X-ray probe absorption and emission spectroscopy. The information that is in principle available from X-ray emission, XANES or EXAFS measurements – metal coordination numbers and geometry, oxidation state changes, bond lengths, type of ligands etc., is potentially of crucial value for validating any mechanistic model of the process under investigation. Of special interest is here also soft X-ray spectroscopy on transition metal L-edges and light atom K-edges. Due to the smaller natural linewidth L-edge transiton metal absorption spectroscopy is richer in features compared to K-edges and provides direct information about the electronic structure of the element studied. Due to the fs nature of the ELI-APS pulses conventional damage mechanisms, hampering soft X-ray spectroscopy of biological samples at synchrotrons, are not occuring and observation of metal L-edge and oxygen or nitrogen K-edge features under close to physiological conditions becomes possible[219] (Mitzner et. Al 2013). These studies can be coupled with optical or THz pumps to populate excited states and study the evolution of these states in the fs to ms time domain (from excited state dynamics down to protein rearrangements influencing the active site environment).

(viii) Alignment of samples by intense THz fields

[213] J. E. Bullock J. Am. Chem. Soc. 131: 11919-11929 (2009).
[214] A.A. Pascal et al. Nature 436: 134-137 (2005).
[215] C. I. Blaga et al. Nature**483**(7388): 194-197 (2012).
[216] Y. Huismans et al. Proc. Natl. Acad. Sci. USA 108: 13516–13521 (2011).
[217] L.X. Chen Annu. Rev. Phys. Chem. 56: 221-254 (2005).
[218] C. Lambertz Chem. Sci. 5, 1187-1203 (2014).
[219] R. Mitzner J. Phys. Chem. Lett.4**,** 3641 (2013).

Alignment of (induced or permanent) polar molecules by the strong external quasi-dc field will allow studies of novel types of coherent or collective phenomena (for gas phase, see 220 . It has also been shown that using relatively weak linearly polarized infrared laser beam, used for trapping, even large objects, such as whole chloroplasts and chromosomes can be aligned221 . It is anticipated that for small, anisotropic molecular assemblies larger fields, which are available at ELI-ALPS could be used in applications where anisotropy is of interest. Based on these results and those obtained on self-assembling films of small molecules with linearly polarized pulsed infrared laser beam222, the alignment of membrane proteins in Langmuir troughs, and/or the formation of 2D ordered arrays on solid surfaces seemfeasible.

Advanced spectroscopic techniques for biological application

A number of advavnced techniques and associate instrumentation and devicesserve a class of experiments in bilological applications. These arrangements serve experiments that are expected to be conducted and thus are considered to be provided by ELI-ALPS by a large number of users. They include:

(i) XUV-pump XUV/visible/NIR probe true attosecond pump-probe spectroscopy

The true attosecond pump-probe technique is still in its infancy[223], however, this is an area where real breakthorughs are expected, specifically suited to ELI-ALPS. Among the most impressive results of attosecond spectroscopy is the demonstration that the molecule is able to localize the electrons even following ionization, i.e. it is possible to experimentally observe the quantum mechanical density function[224], the electron wave packet[225] or proton dynamics. In the near future it is not expected that the procedure can be extended to macromolecules, however, biologically relevant molecules of moderate size (NADH, FAD, retinal, amino acids, nucleotides) can be observed this way.

(ii) Femtosecond transient UV/VIS absorption spectroscopy and multidimensional coherent ultrafast optical spectroscopy

Femtosecond transient absorption spectroscopy is widely used in photosynthesis and photobiology and photochemistry, in femtobiology and femtochemistry[226], which dictates that set-ups should be made available at ELI-ALPS.

The role of coherence in excitation dynamics and its influence on the efficiency of energy transfer and photoinduced charge separation in molecular complexes is intensively discussed during the last years. This discussion originates from two-dimensional (2D) coherent electronic spectroscopy data, which recently demonstrated the oscillatory behavior in the 2D spectra of various molecular systems. However, it still remains unclear what is actually being observed: either excitonic and/or vibrational wavepacket motion[227]. To solve this problem and to identify the possible role of the coherence (quantumness) in energy transfer and charge separation various coherent spectroscopic (one-colour and two-color) approaches should be used. For instance, by using two-color (VIS and IR) measurements the possible relationship between electronic and vibrational degrees of freedom could be resolved. 2D spectroscopy has also proved to be a valuable tool in providing a wealth of information on the pigment interactions, energy and electron transfer pathways and dynamics of

[220] S.Fleischer et al. Phys. Rev. Lett. 107: 163603 (2011).
[221] G. Garab et al. Eur. Biophys. J. Biophys. Lett. 34: 335-343 (2005).
[222] A.Birman Angew. Chem. Int. Ed. 122: 2404–2407 (20101).
[223] P. Tzallas et al. Nat Phys**7**(10): 781-784 (2011); P. Carpeggiani et al. Phys. Rev. A89, 023420 (2014)
[224] D. Shafir et al. Nature Physics **5**(6): 412-416 (2009); E. Goulielmakis et al. Nature **466**(7307): 739-U737. (2010);
[225] S. Haessler et al. (2010) Nature Physics **6**(3): 200-206. (2010).
[226] A. H. Zewail J. Phys. Chem. A, 104: 5660-5694 (2000).
[227] V. Butkus Chem. Phys. Lett. 587: 93-98 (2013); F. D. Fuller Nature Chem., **6**, 706-711 (2014).

LHCII[228]. This technique, along with the emerging 3D electronic spectroscopy, will most certainly contribute to the mapping of primary energy and electron transfer processes in natural and artificial systems designed for light harvesting and photochemical energy conversion.

(iii) Time-resolved Raman and IR spectroscopy

As a light source, ELI-ALPS offers a wide range of tunable UV light which could certainly be used to UV-Raman and developed to pulsed UV Raman, which may be of high interest in biology. Time–resolved Raman spectroscopy has been proved to be very successful in the better understanding of the photophysical and photochemical properties of carotenoids[229]. A transient-Raman spectrometer for pump-probe experiments could readily be installed at ELI-ALPS. There are many questions left open in photosynthesis on the structure of the excited states, e.g. the photoprotective mechanism of non-photochemical quenching of the singlet excited state of chlorophyll-a in plant light harvesting antennae[230]. Time-resolved Resonance Raman may provide direct access to this problem. It may also help one to record the Raman signal before the fluorescence, which may of great help in a number of systems.

Transient IR spectroscopy would be of special interest when monitoring the dissipation processes in photosynthesis, i.e. the fate and effect of excitation energy that cannot be utilized for photochemistry. Reconstituted light-harvesting complexes have been designed and constructed for such studies[231] (Lambrev et al. 2013). Although transient IR devices are accessible in different laser centers[232] , a top-notch instrument at ELI-ALSP would most certainly be of use by the research community.

(iv) Transient Multi Probe Spectroscopy

In the newly developed ultrafast spectroscopy technique following a femtosecond pump signal using multiple probes relaxation processes up to milliseconds can be followed in a single scan, giving the possibility to characterize complex dynamic phenomena[233].

5.2 Biomedical applications

Radiobiology

Motivation, state of the art

The installation of ultrafast, high-energy lasers with high repetition rate at ELI-ALPS, opens the possibility for development of revolutionary approaches in radiation oncology. The motivation behind is the continuous growing of the cancer incidence. The rapid development of radiation therapy in the recent decades improved remarkably the efficiency of cancer treatment[234]. One of the main reasons was the broad introduction of compact linear accelerators providing electron and photon beams with energies up to 20 MeV and of advanced beam delivery techniques like intensity modulated radiation therapy (IMRT) and imaged guided radiotherapy which allows selective dose delivery. Further increase in dose precision can be achieved by the advantageous physical feature (energy deposition characterized by the Bragg peak) of accelerated heavy charged particles (protons and heavier ions). In addition to the

[228] K. L. Wells Phys. Chem. Chem. Phys. 16: 11640-11646 (2014).
[229] H. Hashimoto et al. Biochim. Biophys. Acta – Bioenergetics1847: 69–78 (2015).
[230] A.A. Pascal et al. Advances in Photosynthesis and Respiration Vol. 40. Springer Dordrecht. Heidelberg, New York, London (2014).
[231] P. H. Lambrev et al. J. Phys. Chem. B 117: 11260-11271 (2013).
[232] M.-L. Groot et al. Photochem. Photobiol. Sci. 6: 501-507 (2007).
[233] G. M. Greetham et al. Rev. Sci. Instrum. 83, 103107 (2012).
[234] C. Allemani et al. the CONCORD Working Group. Lancet 2014 Nov 26; doi:10.1016/S0140-6736(14)62038-9.

precise dose delivery, heavy ions are „densely ionizing" particles with increased relative biological effectiveness (RBE). However, at present less than 1% of the patients have access to charged particle therapy facilities due to their large size and complexity resulting in extreme high construction and operation costs[235]. The development of new, cost-effective heavy charged particle accelerators could increase the availability of these most advanced therapeutic methods to the wider public[236]. Apart from more compact accelerators due to ion acceleration on micrometer scale, high-intensity lasers allow the generation of a wide range of radiation qualities which are advantageous for medical application. In addition to therapeutic irradiation, laser-based radiation provide extraordinary imaging tools, e.g. for basic subcellular research and monitoring of radiation delivery to patient. Moreover, high-intensity laser systems may provide synchronously several radiation qualities for multiple related functions, such as imaging and therapy, which possibility could be explored for complex biomedical applications. ELI-ALPS will provide not only primary laser light over a wide spectral and temporal range but also secondary sources for soft and hard X-rays (1 keV to 100 MeV), high-energy electrons (up to 2 GeV) and protons (up to 250 MeV), appropriate for medical and biological research already from the first phase of implementation. This will be an exceptional basis for outstanding research toward the development of compact, laser-driven, energy and intensity modulated, image guided, complex multi-particle radiation therapy facilities. Additionally, laser-driven beams have the particular property of ultra-short, very intense pulses resulting in an extreme high pulse dose rate (>10^{10} Gy/second) which may introduce a new dimension in radiation therapy[237]. The variety of available radiation sources at ELI-ALPS will allow to study radiobiological effects at atomic, molecular, (sub)cellular, tissue and organ level. For translation of the new, laser-based ionizing radiation sources to clinical application, a wide range of multidisciplinary research including radiation monitoring, dose calculation, dosimetry and extensive radiobiological investigations on different biological models is necessary.
The research infrastructure offered by ELI-ALPS is supplemented by the regional biology and medical institutions providing an excellent basis for highly innovative, interdisciplinary approach for basic as well as applied radiobiology and medical research. The scientific activity in the field of radiation biology at ELI-ALPS is integrated in the regional laser associated biomedical and comprehensive anticancer research programs and intended to be harmonized with international research on biophysics and radiation biology at laser-driven beams.

Radiation biology research will benefit from both the primary and secondary sources at ELI-ALPS as highlighted bellow.

- Primary laser sources

Mid-infrared laser line (MIR)

The mid-infrared spectral region achieved with the planned IR OPCPA primary source is well suited to a large number of medical applications. Bio-molecules have specific absorption lines in the 3-15 μm region and so a well-tuned laser can selectively excite and so detect or destroy molecules. Applications in Laser Dentistry, Laser Ophtalmology, Laser Dermatology, Laser Angioplasty, Endoscopic Submucosal Dissection and Laser Lithotripsy might benefit from short duration of the MIR pulses as well[238].

[235] A. Peeters et al. Radiother Oncol 95(1), 45-53 (2010).
[236] S.S. Bulanov Med Phys 35(3), 1770-1776 (2008); J. M. Schippers and A. J. Lomax, Acta Oncol 50, 838-850 (2011); K.M. Hofmann et al. J Biophotonics 5(11-12), 903-911 (2012).
[237] M. Fernet et al. Int J Radiat Biol 76(12),1621-1629 (2000);T. Prempree at al. Int J Radiat Biol Relat Stud Phys Chem Med15(6), 571-574 (1969); V. Favaudon et al. Sci Transl Med 6(245), 245ra93 (2014).
[238] J. Qiu et al. J Biomed Opt 15(2), 028001 (2010); B. Pajic et al. Clin Ophthalmol 8, 2485-2489 (2014); J. Ilgner et al. J Biomed Opt. 11(1), 014004 (2006); J. L. Calvo-Guirado et al. Clin Implant Dent Relat Res (2013) doi:10.1111/cid.12162; A.S. Kabas et al. J Biomed Opt18(9), 098003 (2013).

Femtosecond infrared (IR) laser filamentation

High-power ultra-short infrared laser pulses produce high-density avalanches of low energy electrons via laser filamentation, selectively depositing large doses deep inside an adjustable, well-controlled macroscopic volume in a spatial energy density and temporal dose rate that both exceed by orders of magnitude the most intense clinical radiotherapy systems. A focused femtosecond pulse induces optical breakdown with significantly less pulse energy than is required with a longer pulse. The use of femtosecond pulses therefore minimizes the amount of energy deposited into the targeted region of the sample, minimizing mechanical and thermal effects that lead to collateral damage in adjacent tissues. The effect at the molecular level is comparable to that of conventional ionizing radiation, and the first results on animal cancer show a clear therapeutic benefit, which warrant further investigations on that highly promising technique[239].

- Secondary sources

Laser driven X-ray beam

Clinical X-ray imaging presently relies on X-ray generators that deliver a broad polychromatic spectrum. Coherent X-ray beams enable the utilization of phase-contrast shadowgraphy or 3D tomography in order to get high-resolution insight into the density structure of normal and tumour tissue. Phase-contrast X-ray imaging offers significantly higher contrast in soft tissue for detection of morphological changes, than conventional X-ray radiography. Phase-contrast tomography theoretically allows more accurate 3D detection of tumour growth and regression as well as normal tissue radiogen damages in the *in vivo* biological models, than conventional X-ray CT. The monochromatic X-ray imaging bears the potential of detecting cancer in early-, even pre-cancerous stages[240]. Moreover, a laser-based coherent phase-contrast tomography will also allow for easy implementation of image-guided radiotherapy, e.g., by combined use of the laser for generation of laser-driven therapeutic particle beam and diagnostic X-rays.

The radiation effect of pulsed mode and ultra-high dose rate of laser-based X-rays could be studied on cancer and normal tissues. Up to now the few radiobiology studies performed with laser driven X-ray sources suggest that there is no increase in the cytotoxic effects of laser-based X-rays at dose rates as high as 10^{13} Gy/second[241].

High -energy electron beam

Laser-driven electron beams reach very high energies (>100 MeV) and can be beneficial for medical purposes, providing relevant advantages over currently available (≤20 MeV) electron and photon beam treatment. The high beam energy allows an increase of depth dose[242]. Moreover, laser-driven electron beams could be considered as monoenergetic with low divergence and potentially are of interest for electron beam therapy of deep seated tumours[243]. Small diameter beamlets can be scanned, thereby producing a better resolution for intensity modulated treatment than achievable with photon beams. Additionally, the laser-based electron acceleration results in ultra-high dose rate, delivering very high instantaneous doses within a time interval shorter than many chemical reactions. First experiments with tumour cell cultures[244] and tumour xenograft on small animals[245] were carried out with laser-accelerated electron beams. Very recently, a favourable increase of the differential response between

[239] R. Meesat et al. Proc Natl Acad Sci USA 109, E2508-E2513 (2012).
[240] S.A. Zhou et al. 139(1-3), 334-338 (2010).
[241] K. Shinohara et al. J Radiat Res 45(4), 509-514 (2004).
[242] O. Lundh et al. Med Phys 39(6), 3501-3508 (2012).
[243] C. DesRosiers et al. Proc. of SPIE 6881, 688109/1-688109/14 (2008).
[244] E. Beyreuther et al. Med Phys 37(4), 1392-1400 (2010); L. Laschinsky et al. J Radiat Res 53, 395-403 (2012).
[245] K. Brüchner et al. Radiat Oncol 9, 57 (2014); M. Oppelt et al. Radiat Environ Biophys (2015) doi:10.1007/s00411-014-0582-1.

normal and tumour tissue has been shown in mice for ultra-high dose rate flash irradiation at a conventional accelerator[200]. Further translational research on the effects of laser-driven very high energy electron radiation using differently complex biological models including the detection of molecular basis of action is a very promising approach.

Heavy charged particle beam

Proton and ion beams provide the highest physical selectivity and millimeter accuracy in radiation dose delivery. Furthermore, the biological effectiveness of heavy charged particles could be 2-4 times higher than that of photons and electrons. The laser-plasma based particle acceleration may provide different nuclear particles, which could be exploited for improved cancer treatment. The laser approach may offer considerable cost savings in facility construction, operation and long term service.

ELI-ALPS HF with advanced, high-repetition-rate laser technologies, providing high intensity, high energy pulses in the short-pulse regime (<20 fs), open the wide research avenue toward the development of laser-based compact ion accelerators for medical application. Recent *in vitro* data showed similar effects of laser-accelerated versus conventional (continuous) proton beams[246] and some early small animal experiments with pulsed proton beams at a conventional accelerator supported these observations for tumour models[247]). Further extensive investigations are needed in order to study the biological effects of laser-driven ion beams out of the Bragg peak and within the Spread Out Bragg Peak (SOBP), i.e. both for healthy and tumour tissues, obtaining clinically relevant data and exploiting the biomedical processes behind as well.

Research directions and activities

I. Studies on cancer cell properties and molecular processes of ionizing radiation

In recent years investigations focused increasingly on molecular processes inherent in normal and cancer cells, yielding basic understanding on cancer development and introduction of highly effective therapeutic strategies. Radiation-related research activity turned as well as towards molecular processes and their impact on the effect of radiation on normal and malignant tissues. Studies on activation of multiple signal transduction pathways by ionizing radiation has indicated the dependence on dose, dose rate, radiation quality, cell type and many other variables, which a lot remained to be explored from. Subcellular spatial detection and imaging methods with ultra-high resolution at ELI-ALPS could lead to further results on tumour metabolism and stem cell biology, immunology and inflammation, clinical translation and biomarkers, differential effect of ultra-high dose rate and signaling pathways, DNA repair of normal and pathologic tissue, tumour microenvironment and hypoxia, radiation sensitizers and protectors, genomics and epigenetics in radiation oncology.

II. Microdosimetry

The dependence of biological effectiveness on radiation quality is attributed to the energy deposition pattern on atomic, molecular and cellular level. Information about the spatial distribution of energy deposition and its correlation with primary and secondary particles from models (validated by measurements) are important, for a fundamental understanding of microscopic dose and dose concepts and the associated uncertainties. An adequate detailed

[246] A. Yogo et al. Appl Phys Lett 94, 181502 (2009); C. Richter et al. Phys Med Biol 56, 1529-1543 (2011); D. Doria et al. AIP Advances 2, 011209/1-011209/6 (2012); J. Bin et al. Appl Phys Lett 101, 243701 (2012);K. Zeil et al. Appl Phys B110, 437-444 (2013).

[247] O. Zlobinskaya et al. Radiat Res 181(2), 177-183 (2014).

description of the energy deposition in living matter by studying the properties of the particle track structure can give answers for different radiobiological problems, such as radiation quality dependency of the dose-response relation for a particular biological system; differences between biological systems showing different susceptibilities for producing radiation-induced effects; nonlinearity of many biological systems; differences between overall response of a biological system for inhomogeneous exposure; the effects of dose rate and dose fraction[248] . Research related to nanometric/micrometric energy deposition pattern in combination with short pulses of laser-driven ionizing radiation sources may improve the understanding of spatio-temporal correlations of radiation interaction events and of radiochemical processes involved in development of biological effects and may reveal and quantify correlation between spatio-temporal track structure and radiation damage. The following experiments anc schemes could be developed and established for research at ELI-ALPS to answer the above questions: (i) single particle – single cell ion microbeams, (ii) dynamics studies, (iii) track structure imaging systems, and (iv) microdosimety systems including microcalorimeters, silicon-based high-resolution solid-state dosimeters, tissue-equivalent proportional counters, electrical circuits built from DNA molecules, and 3D dosimetry techniques.

III. Beam monitoring and dosimetry

The main condition of reliable, reproducible radiobiological experiments and of any clinical application is a stable, reliable, precise radiation dose delivery. To fulfill this condition extensive research is needed which covers dosimetry of all planned radiation sources at ELI-ALPS (X-ray, electron, ion). The characteristics of laser-driven ionizing radiation, in particular the short, ultra-high intensity pulses, does not allow the direct application of established dosimetry protocols. New approaches are challenged by the large range of target volume size which covers orders of magnitude, from micrometer scale for single cell irradiation experiments to tumours of several centimeters size in patients. First dosimetry systems have been developed for cell culture and small animal experiments with laser-driven beams, combining different dosimeters in a dedicated integrated system to provide spatial distribution, absolute values and online information for the delivery of prescribed doses to predefined target volumes. The overall dose uncertainty of about 10% achieved so far has to be reduced below 5% as required for radiotherapy application.

New methods for off line and on line characterization of laser-generated beams, by diagnostic of the laser conditions and monitoring of the secondary source parameters at the laser target, are proposed, in order to control the energy spectrum, the transverse profile including the divergence, and the intensity of the laser-driven beam. Additional, accurate direct dosimetric measurements are required at irradiation site. From the various detector types like radiochromic films, scintillator-based detectors, chemical dosimeters with polymer gels, silicon-based solid-state pixel detectors, ionization chambers, calorimeters, Faraday cups, Rogowski coils and integrated current transformers, suitable components have to be identified and combined in dedicated systems depending on application and radiation quality. Methods for "quenching correction" are essential for many dosimeters to correctly measure the dose also at high local energy-deposition (e.g. at high LET in the Bragg peak region of ion beams, at ultra-high dose rate of laser-driven radiation pulses) and have to be developed and established. Dedicated phantoms (from simple plastic or water to antropomorphic phantoms) should be defined for dosimetric purposes. Apart from dose measurement, beam and dose calculation using CT based 3D treatment planning software or Monte Carlo codes like MCNP are necessary to provide data for definition of field size, source to object distance, and time or monitor units for irradiation

[248] W. Rühm et al. EURADOS Report 2014-01, Braunschweig, (2014); M. Desrosiers et al. J Res Natl Inst Standards Technol 118, 403-418 (2013).

of biological systems with prescribed doses. The laser-driven secondary radiation sources at ELI-ALPS will provide unique opportunity for testing and verifying dosimetric components and approaches.

IV. Radiation biology research

Pioneering research on the biological effects of laser-accelerated ionizing particles is under progress in laser centers in Europe, in the US and in Japan. With laser-driven photons and electrons, *in vitro* tumour cell irradiation experiments indicated no major difference in biological effectiveness. Similar results have been derived from investigations on laser-accelerated protons by groups producing 0.8-23 MeV energy beams for tumour cell irradiations. *In vivo* studies have recently been started for laser-driven electrons using a small animal tumour model[8]. Studies of the response of normal tissue cells to laser-driven beams are very rare (*in vitro*) or still completely missing (*in vivo*).

Possible ultra-high dose rate effects should be associated with specific early-time physical interactions such as ionizations and excitations, and subsequently generated chemical radicals. Various *in vitro* and *in vivo* studies have been performed at conventional pulsed radiation sources, which could mimic laser-based radiation quality to some extent. Authors have shown, e.g., that short pulses of X-rays or protons[249]produce fewer dicentric chromosomes than do conventional irradiation in human blood lymphocytes and human-hamster hybrid cells, and fewer micronuclei in human cervical cancer cells and human keratinocytes in a reconstructed skin model. Differential G2 arrest was also reported after pulsed compared to continuous irradiation[2], suggesting that intense pulses generate a smaller amount of clustered DNA damage sites than do continuous irradiation. Moreover, short pulses of radiation, each given at a conventional dose rate and repeated at a few minutes interval, reduced late normal tissue toxicity compared to the same total dose given in a single fraction[250]. Recently, a promising increase of the differential response between normal and tumour tissue has been shown in mice for short high-intensity electron pulses.

Exploring the mechanism of the differential biological effect induced by short intense laser-based radiation pulses and exploiting for better clinical efficacy is a highly promising research field. It requires translational research including *in vitro* cell irradiation to animal experiments and finally clinical trials and studying the radiobiological processes and effects at atomic, molecular cellular, tissue and organ level for the different laser-driven ionizing particles and tumour entities. For this, the use of hierarchical biological systems including yeast, classical cell cultures of tumour and normal cell lines, 3D cell co-cultures and animal models, and the investigation of a wide range of biological endpoints including morphologic, functional, histopathologic, proteomic and genomic examinations are necessary. Apart from traditional systems of *in vitro* cell cultures with clonogenic cell survival assay or comet and H2AX assay for DNA damage detection, extensive and complex preclinical examinations in small animals are necessary. Classical small animal models to study acute mouse skin reaction[251], intestinal crypt assay[252], locomotor disorders of extremities due to spinal cord radiation[253] are established both for low and high LET radiation. New models of wild type, transgenic and transparent zebrafish lines should advance the research on the effects of ionizing radiation, e.g. for combined modalities with radio-sensitizing and -protective agents. Apart from tumour response, particular focus is on the

[249] S. Auer et al. Radiat Oncol 6, 139 (2011); T. E. Schmid et al. Radiat Res 172, 567-574 (2009); T. E. Schmid et al. Radiat Res 175, 719-727 (2011); T. E. Schmid et al. Radiother Oncol 95, 66-72 (2010).
[250] Y. Rong et al. Int J Radiat Oncol Biol Phys 79(3), 934-942 (2011).
[251] K. Kagawa et al. Int J Radiat Oncol Biol Phys 54(3), 928-938 (2002).
[252] J. Gueulette et al. Radiother Oncol 61(2),177-184 (2001).
[253] P. M. Medin et al. Int J Radiat Oncol Biol Phys 79(1), 226-232 (2011).

effects on normal tissues in order to ensure safe clinical introduction of innovative laser-driven radiation approaches both for diagnostic and therapy application.

5.3 Materials science

Computationally assisted chemical & materials research

Modern technology entails the manipulation of matter on ultrashort scales, and measurement of the dynamic processes in ultrafast domain. Thus "ultrafast science" impacts multiple areas of modern physics, chemistry, biology, materials science, engineering etc. Formation and breaking of chemical bonds occur in femtosecond time scale, and thus, elementary molecular processes can be observed and utilized by freezing the transition states of chemical processes at ultrashort time scale, even shorter than the vibrational and rotational periods in matter.

The ELI-ALPS infrastructure with many unique features such as ultrahigh intensity and electric field, ultrashort pulses of high energy radiations in the femto-zeptosecond regime, and electrons with ultra-relativistic energies would help expand the frontiers of knowledge of ultrafast phenomena at the nanoscale and in complex materials. For example, nonlinear optical processes in the XUV region[254] are of extreme importance in the field of quantum electronics and ultrafast optics.

Such advanced research in materials science progressively facilitates from the support of theoretical modeling and computational research, resulting in explosion of activities in areas dealing with electronic structures, molecular dynamics, nonlinear spectroscopies etc. For example, from the comparison of the potential energy surfaces of reactive systems in their ground state and excited states, it is feasible to locate and analyse the particular regions of surface where transition states play the most crucial role for advanced applications. The domain of computational research and modeling has varieties of tools. Electronic structures, molecular orbitals etc are best described with ab-initio techniques, such as Hartree-Fock methods, Density functional theory, Time-dependent density functional theory, and ab-initio molecular dynamics. Using Time-dependent DFT, it is possible to address many interesting issues relevant to ELI-ALPS facilities, such as, bonding and transition state dynamics, photo-ionization, energy loss/absorption spectra and high harmonic generation.

Exploration of ultra-fast timescales using time dependent density functional theory and quantum optimal control theory to address chemical processes[255] in different time scales is of paramount importance in modern materials research[256]. While bond rotations are comparatively slower, bonding through lattice vibration and subsequent electron redistribution is an ultrafast process occurring at femtosecond time scale. The dynamics and the speed of these processes can be tuned further by probing with intense laser. Such studies will be easily feasible in ELI-ALPS research facilities under materials research in strong field science research, interesting from both fundamental perspective, as well as from the point of view of wide-ranging applications. The diagram below categorizes the spectrum of possible applications of ELI-ALPS facilities in some

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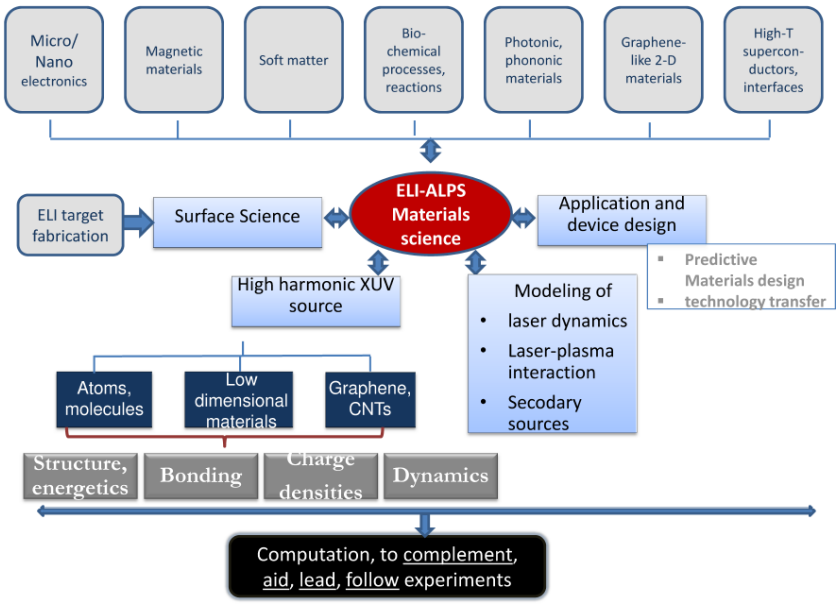
[254] N.A. Papadogiannis, L.A.A. Nikolopoulos, D. Charalambidis, P. Tzallas, G. Tsakiris and K. Witte *Phys. Rev. Lett.* **90**, 133902 (2003); T. Sekikawa, A. Kosuge, T. Kanai, and S. Watanabe, Nonlinear optics in the extreme ultraviolet, Nature **432**, 605 (2004).

[255] S. Wall, B. Krenzer, S. Wippermann, S. Sanna, F. Klasing, A. Hanisch-Blicharski, M. Kammler, W. G. Schmidt, and M. H. Hoegen, Atomistic picture of charge density wave formation at surfaces, Phys. Rev. Lett. **109**, 186101 (2012); L. X. Yang, G. Rohde, T. Rohwer, A. Stange, K. Hanff, C. Sohrt, L. Rettig, R. Cortes, F. Chen, T. Wolf, B. Kamble, I. Eremin, T. Popmintchev, M. Murnane, H. C. Kapteyn, L. Kipp, J. Fink, M. Bauer, U. Bovensiepen, and K. Rossnagel, Ultrafast modulation of the chemical potential in $BaFe_2As_2$ by coherent phonons, Phys. Rev. Lett. **112**, 207001 (2014).

[256] I. Radu, K. Vahaplar, C. Stamm, T. Kachel, N. Pontius, H. A. Durr, T. A. Ostler, J. Barker, R. F. L. Evans, R. W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, Th. Rasing, and A. V. Kimel, Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins, Nature **472**, 205 (2011); J. Conyard, K. Addison, I. A. Heisler, A. Cnossen, W. R. Browne, B. L. Feringa, and S. R. Meech, Ultrafast dynamics in the power stroke of a molecular rotary motor, Nature Chemistry **4**, 547 (2012).

important areas of materials science. Our proposed theoretical and computational activities mainly center around probing the ultrafast phenomena in materials science, from two different main



Short overview of materials research and some of the possible applications in ELI-ALPS: experiments and theoretical modeling can complement each other.

perspective: (i) addressing different structures and dynamics at nanoscale with primary and secondary sources. (ii) and, modeling secondary sources.

Modification of the electronic structures of the system like changes of the reaction barrier or energetic shifts in the potential energy landscape result in a very rich reaction dynamics, interesting from both fundamental or application point of view. In low-dimensional structures those are heart to micro and nano-electronics, these dynamics are even more fascinating, and can be thoroughly probed with ELI-ALPS facilities, as well as time-dependent density functional theory.

Modern technology massively uses magnetic switching to encode information in hard drives, magnetic random access memory and other computing devices. Recent research activities[257] have paved a new route to address today's technological demand to push the switching speed limit manyfold than currently used in magnetic memory technologies. Femto- and sub-femtosecond laser-excited coherence between molecular electronic states (and their suitable superpositions) can switch magnetic order through sudden manipulation of competing phases of correlated materials[258], resulting in ultrafast magnetic switching. Such photo-induced magnetic phase switching can be better understood through the underlying photosynthetic dynamics, and study of transient effect of an applied electric field on the magnetic order, with potential applications in novel switching devices. Femto- and sub-Femto-second transient polarization of condensed matter systems during a laser pulse can thus be effectively utilised to manipulate spin or change magnetic order. These research activities would be a highly interesting domain of application for ELI-ALPS facilities, particularly accessable through the theoretical and computational modeling.

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[257] J. Y. Bigot, M. Vomir, and E. Beaurepaire, Coherent ultrafast magnetism induced by femtosecond laser pulses. Nature Phys. **5**, 515 (2009); T. Li, A. Patz, L. Mouchliadis, J. Yan, T. A. Lograsso, I. E. Perakis, and J. Wang, Femtosecond switching of magnetism via strongly correlated spin-charge quantum excitations, Nature **496**, 69 (2013).

[258] P. Brumer, and M. Shapiro, Controlling chemical reactions in the quantum regime. Phys. Today **64**, 11 (2011).

Investigation of conformer-specific attosecond dynamics in complex molecules[259], experimental and theoretical developments in manipulation of such reaction dynamics, selective bonding and controlled reaction rate have multiple biochemical applications. For example, by changing the conformation of the deliverable drug and making sure it attaches to the right place, we can target efficient drug delivery. While computational tools can help us to easily access the site selectivity of a molecule[260] (for example antibiotics) during chemical and biochemical processes, by irradiation with an XUV pulse, the bond dynamics and transition states are altered. Whether this promotes or hinders the desired reaction, can be addressed through ELI-ALPS facilities, and also can be easily modeled through our theoretical research. From TD-DFT we can find the transition barrier and associated dynamics under the time dependent driving field and can capture the associated nonlinear response of the system, and the whole process in greater detail. We can have the bonding dynamics details through calculating Electron Localization function or ELF. ELF, an output of TDDFT, provides the mathematical description of chemical bonding, in a scale of perfect localization to total absence of electron cloud. In contrast to charge density, ELF gives the true nature of the bond from the localized density. ELI-ALPS facilities would be highly useful to address these ultrafast electron dynamics of complex molecules (and their different possible structural isomers), and associated structure-function relationship with respect to the electronic, i.e., chemical, properties of the molecules.

Thus, by utilising the extreme light, such as from ELI-AlPS laser, it is possible to develop techniques to probe and control the inherent primary events in biochemical and chemical processes/reactions. It seems possible to “grab" and “chanell" the electrons around an excited molecule with ultrafast attosecond pulses, passing them from one pulse to another until we guide them to where we want them. By applying the laser expertise of the in-house experimental groups to the catalytic methods from the user-end, in a number of novel feasibility studies, it is possible in ELI-ALPS to explore the potential future role for sub-femtosecond laser technology over a range timescales as a either a diagnostic of or a participant in catalytic reactions. This will find manyfold applications in materials science, such as to improve the efficiency of current industrial processes, developing interesting photonic materials and new energy sources for the future. For example, when a solar cell first absorbs sunlight, an electron is excited: a process too fast to measure without ultrafast pulses. A possible “film"ing of the process through attosecond pulses will provide clues to enhance the process efficiency. Even more fascinating is the photosynthesis in plants. There are evidences that quantum processes are important in the plants, and with attosecond pulses we could unravel them, and maybe even copy them for our own solar cells.

ELI-ALPS facilities together with the in-house computational activities, also provide access to understanding how femtosecond pulses of light are being used to produce shorter pulses of light that are of attosecond duration, for example, from gas molecules, or graphene, fullerene, nanotubes etc. The information we gain about the molecules and the generation of attosecond pulses come as a result of High Harmonic Generation (HHG). The detailed picture of the process provides the knowledge of the nuclear dynamics going on in the molecule and its electron structure. Probing the attosecond dynamics and utilising the HHG frequency spectrum to understand the elementary energy transfer and charge transfer processes induced by photoexcitation in materials (e. g., donor-acceptor materials, organic semiconducting polymers, chromophore-nanotube assemblies, DNA, various other systems), molecular understanding and application feasibilities can be massively improved. It seems realistic to predict HHG from novel sources utilizable for multiple applications.

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[259] C. R. Becer, R. Hoogenboom, and U. S. Schubert, Click chemistry beyond metal-catalyzed cycloaddition, Angew. Chem. Int. Ed. **48**, 4900 (2009); F. Lepine, M. Y. Ivanov, and M. J. J. Vrakking, Attosecond molecular dynamics: fact or fiction? , Nature Photonics **8**, 195 (2014).

[260] F. Calegari, D. Ayuso, A. Trabattoni, L. Belshaw, S. De Camillis, S. Anumula, F. Frassetto, L. Poletto, A. Palacios, P. Decleva, J. B. Greenwood, F. Martin, and M. Nisoli, Ultrafast electron dynamics in phenylalanine initiated by attosecond pulses, Science **346**, 336 (2014).

For example, recent theoretical work predicts that graphene offers multiple recombination centers[261] for single ionization, allowing possibilities for high harmonic generation. Through addition of dopant or intercalant, and thus tuning the doping type, level and concentration in graphene and similar systems, we can achieve a proper control on the possible lasing and high harmonic generation from these systems. Such study would be feasible in ELI-ALPS materials science division.

The experimental demonstration of graphene in 2004 has triggered a surge of activities on various 2D crystals including single layers of hexagonal-boron nitride (h-BN), several dichalcogenides (such as MoS_2 and WSe_2), and complex oxides, which have been successively prepared using the micromechanical exfoliation technique employed for graphene. Auger processes in two-dimensional atomic materials play very important role. Electron correlation in these two dimensional structures is an important topic in condensed matter and materials research, manifesting itself in multiple applications of materials, e.g., metal-insulator transitions, high-temperature superconductivity, colossal magneto-resistance etc. How electrons interact, their different degrees of freedom superpose in such systems to manipulate the overall reponse of the matter is tremendously interesting and can be approached with pump-probe time-resolved spectroscopy: pumping energy into a particular degree of freedom and measuring the time required for a response to appear in the others. With ELI facilities, it is possible to experiment with and simulate these interesting areas through the attosecond time-resolved spectroscopy of electron-hole dynamics, applicable to ultrafast electron or hole migration, carrier redistribution and more.

Application of brilliant and wavelength tunable short wavelength radiation in materials science

Applications can be found in the fields of chemical/elemental surface analysis e.g. by X-ray photoelectron spectroscopy (XPS, ESCA) or X-ray fluorescence analysis.

High requirement with respect to the high source brilliance (“photon hungry” experiments) are especially given, when the above spectroscopic techniques are coupled to nanoscale spatial resolution in nanospectroscopy or spectromicroscopy, like in the case of Nano-ESCA (XPS with nanoscaled spatial resolution), attosecond spin-ARPES and X-ray Photoelectron Emission Microscopy (X-PEEM).

Nano-ESCA

Nano-ESCA based on electrostatic PEEM imaging and double hemispherical energy analysis enables various measurement modes like non-energy filtered PEEM imaging (detector 1), nano-XPS of selected image regions (detector 2) and energy-filtered ESCA imaging (3), the latter measurement mode imposing the highest requirements on the source brightness[262]. A highly-monochromatic source in the soft to the hard X-ray regime (best tunable) is the optimum requirement.

[261] S. A. Sorngard, S. I. Simonsen, and J. P. Hansen, High-order harmonic generation from graphene: Strong attosecond pulses with arbitrary polarization, Phys. Rev. A **87**, 053803 (2013).

[262] M. Escher et al., J. Phys. Cond. Matter 17, 1329 (2005).

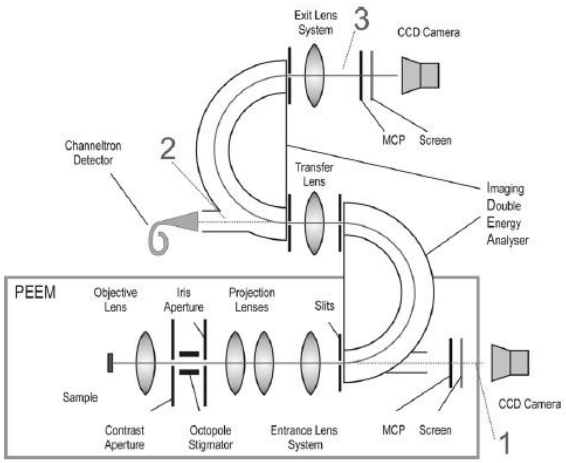

Figure 59. Nano-ESCA system for surface nanospectroscopy

A laboratory-scale Nano-ESCA instrument provides a powerful tool for a wide variety of different measurement tasks in material science, ranging from semiconductor physics and electronic band structure imaging to the investigation of magnetic materials and magnetic domains when using circularly polarized x-ray radiation. Such an instrument has the potential to perform measurement tasks, which currently can only be run at synchrotron radiation facilities due to the high source brightness requirements of the instrument.

Attosecond Magnetism With spin-ARPES

The speed at which a magnetic state can be manipulated and, hence, data can be magnetically stored depends ultimately on the elementary spin-photon interaction, spin-scattering, and spin-transport processes. Until the mid-1990s, dynamics in magnetic systems were believed to occur on time scales of ~100 ps or longer, determined by the interaction of the spins with the lattice. However, studies using femtosecond (fs) laser pulses starting from 1996 revealed the presence of other processes beyond this simple spin-lattice relaxation picture. In magneto-optical Kerr effect (MOKE) experiments, Beaurepaire et al. found that after absorption of an optical pump pulse, the magnetization in Ni sharply decreased on timescales shorter than a picosecond[263]. Since this pioneering experiment on ferromagnetic nickel in 1996, the field of ultrafast magnetization dynamics has seen a constant progress in experimental and theoretical studies. It is now an experimentally well-established fact that the 3d-ferromagnets Fe, Ni and Co show characteristic demagnetization timescales on the order of some 100 fs after optical excitation. On the theoretical side several models based on spin- dependent scattering, transport and magnon generation have been proposed to explain the available experimental data. However, despite being the subject of intense research for over a decade, the underlying mechanisms that govern the demagnetization remain unclear or at least highly controversial[264]. A very recent novel research field that has opened up in the area of ultrafast magnetism is the study of coherent magnetization dynamics effects after an ultrashort laser pulse excitation. First theoretical considerations[265] and one indirect experimental work indicate that the material polarization induced by the photon field interacts coherently with the spins[266]. The corresponding mechanism has its origin in relativistic quantum electrodynamics, beyond the spin–orbit interaction involving the ionic potential. The study of these coherent

[263] E. Beaurepaire et al., PRL 76, 4250 (1996)

[264] J.-Y . Bigot and M. Vomir, Annalen der Physik 525, 2 (2013); A. Kirilyuk, A. V. Kimel, and T. Rasing, Reviews of Modern Physics 82, 2731 (2010)

[265] G. Zhang and W. Hübner, APB 68, 495 (1999). G. Zhang, W. Hubner, PRL 85, 3025 (2000).

[266] J.-Y. Bigot et al., Nat Phys 5, 515 (2009)

magnetization dynamics would allow us to fundamentally understand spin-photon interaction in solids, which would be a huge step in the elucidation of ultrafast magnetic processes. However, this field was so far experimentally not accessible, since the timescale of the process requires light pulses in the very low femtosecond regime to fully resolve the according dynamics. Also, in order to study the light- induced polarization in a direct manner, one needs to be able to follow the photo-induced re-distribution of the spin-polarized carriers in the material, and the subsequent spin-resolved energy vs. momentum band- structure dynamics. In summary, an experimental approach to the novel field of coherent magnetization dynamics requires (1) ultrahigh time-resolution and the capability of (2) spin-resolved ARPES, which are exactly the experimental possibilities enabled by the realization of ELI with a spin-resolved NanoESCA end-station.

Photoelectron spectroscopy for the study of the dynamics in correlated electron systems

Photoemission is an established spectroscopic technique omnipresent at the synchrotron light sources. Its popularity steams from its ability to deliver information on the chemical composition of solid state samples (via monitoring the core levels in atoms) and the structure of the electron bands close to the Fermi energy (by performing angle-resolved photoelectron spectroscopy). A well-conceived photoemission instrument stationed at the beamline spanning the photon energies from vacuum ultraviolet to soft x-rays can deliver a great deal of data on samples belonging to the every corner of the condensed matter physics. Moreover, implementing user-friendly experimental stations for the photoelectron spectroscopy at the new light source facilities with well-defined and controllable timing structure of the photon pulses (FEL`s or HHG`s) will ensure access to the time-resolved photoemission data by a wide pool of users interested in understanding dynamics of the electronic correlations in solids in connection with the studies of magnetism, spintronics, superconductivity, and other phenomena governed by the electron correlations (giant magneto-resistance for example).

Manipulation of matter by light might emerge as one of the central research themes at such photoemission instruments. The topics might include photo-induced superconductivity[267] and optical switching of magnetization which along with the switching by electric fields is posed to revolutionize information technologies. So far the magnetization switching has been mainly studied using X-ray microscopy and X-ray magnetic circular dichroism[268]. Photoemission instrument will deliver the new data on the photo-induced modifications of the electronic structure which are naturally behind all these fascinating phenomena. As far as studies of the magnetism are concerned, it will be crucial to fit the photoemission spectrometer with the detector sensitive to the spin of the photoelectrons. Recent improvements in the efficiency of such detectors[269] shall enable previously impossible experiments aimed at the ultrafast studies of the magnetization dynamics.

Actinic inspection tools in Extreme Ultraviolet Lithography as next generation lithography

Extreme Ultraviolet Lithography (EUVL) is considered to be the most promising next generation lithography technology for high-volume production of logic and memory semiconductor devices with critical dimensions of 32 nm and below. The technology is based on an incoherent high-power XUV source, condenser optics and all-reflective multilayer-coated imaging objective. One of the most critical issues of EUVL is the realization of defect-free multilayer coated reflection masks. It has been proven, that many critical defects can only be found by at-wavelength (actinic) inspection

[267] D. Fausti *et al*., Science **331**, 189 (2011), J. D. Rameau *et al*., Physical Review B **89**, 115115 (2014)

[268] I. Radu *et al*., Nature **472**, 205 (2011); L. Le Guyader *et al*., Nature Communications **6**, 5839 (2015); T. Zhao *et al*., Applied Physics Letters **90**, 123104 (2007)

[269] I. Radu *et al*., Nature **472**, 205 (2011); L. Le Guyader *et al*., Nature Communications **6**, 5839 (2015); T. Zhao *et al*., Applied Physics Letters **90**, 123104 (2007)

at the lithography wavelength (13.5 nm) and are hidden from already established inspection methods in the DUV. Therefore, different actinic microscopic or scattering techniques have been developed to locate and detect defects on EUVL mask blanks (unstructured multilayer coatings) or EUVL reticles ("ready" masks) down to the sub-30 nm size level.

However, finding all defects on a full 6 inch mask within a reasonable time (~ few h) in a laboratory-based inspection tool requires very fast data acquisition (10 nm voxel number ~ 10^{14} !) and a very high brightness illumination source at the EUVL wavelength of currently 13.5 nm.

One technical actinic mask inspection system (displayed in Figure E.5) is based on standing-wave assisted Photoelectron Emission Microscopy, where a standing wave is induced inside and on-top of the multilayer reflection coating, when resonantly illuminated at the lithography wavelength (13.5 nm) and at near-normal incidence angle (5 deg)[270]. The photoelectron yield measured from the sample surface by the PEEM is controlled by the electrical field intensity (Fermi's golden rule) and thus by the relative phase position of the standing wave. Defects on-top as well as sub-surface buried defects inside the multilayer coating result in little local phase distortions, which alter the local photoelectron yield at defect positions and thus give rise to an image contrast in standing-wave PEEM.

Up to now this technique (together with other techniques based on darkfield scattering of nano-focused XUV beams) could only be implemented at synchrotron radiation storage rings (BESSY 2, ALS), but integration into the cleanroom environment of semiconductor lithography laboratories is ultimately needed to convert these proof-of-concept instruments into a commercial XUV metrology tool. This integration depends on the availability of a high-brightness laboratory-based XUV source, which can be realized in the future on the basis of high repetition rate HHG.

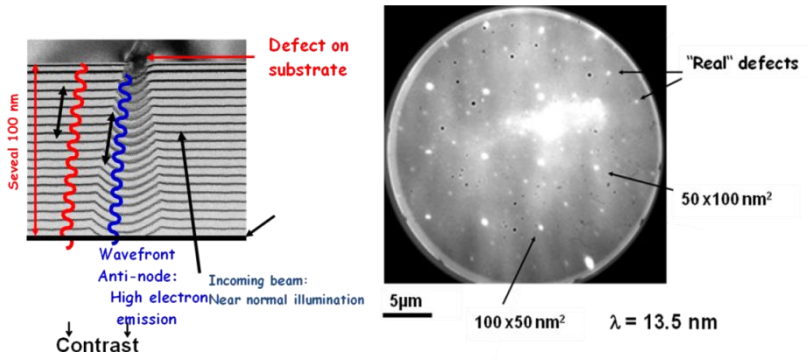

Figure 60. Schematic setup of a standing wave PEEM and defect image of 50 nm buried defects

5.4 Manipulation of matter by intense THz fields

Motivation, state of the art

Pulsed electromagnetic radiation from the THz frequency range with extremely high peak intensities offers unique application possibilities with immense scientific discovery and application potential. These derive, on the one hand, from the specific degrees of freedom in matter (molecules,

[270] J. Lin et al., Microelectronic Engineering 85, 922 (2008); J. Lin et al., Opt. Express 16, 15343 (2008).

nanostructures, condensed matter – including biomaterials) accessible by THz radiation. On the other hand, the THz wavelength range is conveniently fitted to many innovative applications allowing to build devices practically impossible by using other types of radiation. This latter type of applications includes especially the manipulation of charged particles.

- Nonlinear THz spectroscopy

Nonlinear THz spectroscopy requires μJ-level THz pulse energies and peak electric field strengths on the order of 100 kV/cm. The spectacular increase in the energy of THz pulses in the last few years enabled the investigation of nonlinear THz response[271] and ultrafast carrier dynamics in semiconductors by *THz pump—THz probe* measurements[272,273,274]. Picosecond carrier dynamics in InSb following excitation by below band gap broadband far-infrared radiation was investigated[275]. Carrier heating and impact ionization dynamics was observed. THz pump—THz probe measurements also revealed the strong saturation and subsequent recovery of free-carrier absorption in n-type semiconductors in the THz frequency range[276]. Ultrafast high-field transport of electrons was studied in n-type GaAs with ultrashort THz pulses having electric field amplitude up to 300 kV/cm[277]. Observation of nonequilibrium BCS state dynamics induced by THz pulses in a superconducting NbN film was reported[278]. High-field THz bulk photovoltaic effect was recently observed in $LiNbO_3$[279].

THz pump—optical probe spectroscopy enabled the investigation of highly nonlinear THz light-matter interaction in single-walled carbon nanotubes[280] and the study of the interaction of excitons with THz electric fields in ZnSe/ZnMgSSe multiple quantum wells[281]. In the latter case the dependence of the excitonic absorption resonance energy on the THz field was observed to follow the Stark effect for smaller THz fields, while for larger THz fields the interaction enters the nonperturbative regime. High-field single-cycle THz pulses have been used for bi-directional ultrafast electric-field gating of interlayer charge transport in a cuprate superconductor[282].

The unique combination of ultrashort light sources of ELI-ALPS spanning the range from infrared to x-rays with precise synchronization possibility to THz sources providing the highest achievable field strengths allows novel types of pump-probe studies. These include THz pump—THz probe, and THz pump—optical probe spectroscopy, where intense THz pulses are used to initiate changes in the sample and THz or optical pulses are used for detecting these changes.

- Multi-spectral imaging

By using intense THz sources with typical pulse energy on the order of 10 μJ time-domain THz spectroscopy (TDTS) can be combined with imaging techniques, a method called multi-spectral imaging (MSI). This allows, for example, to investigate large samples using 2D electro-optic sampling, without the need for time-consuming scanning over the sample surface[283]. MSI is an interesting new tool for non-destructive testing, recording the spatial patterns of various

[271] P. Gaal et al., Phys. Rev. Lett. 96, 187402 (2006).
[272] L. Razzari et al. , Phys. Rev. B 79, 193204 (2009).
[273] J. Hebling et al., Phys. Rev. B 81, 035201 (2010).
[274] H. Hirori et al., Phys. Rev. B 81, 081305 (2010).
[275] M. C. Hoffmann et al., Phys. Rev. B 79, 161201 (2009).
[276] J. Hebling et al., Phys. Rev. B 81, 035201 (2010).
[277] W. Kuehn et al., Phys. Rev. B 82, 075204 (2010).
[278] R. Matsunaga and R. Shimano, Phys. Rev. Lett. 109, 187002 (2012).
[279] C. Somma et al., Phys. Rev. Lett. 112, 146602 (2014).
[280] S. Watanabe et al., Opt. Express 19, 1528 (2011).
[281] H. Hirori et al., Phys. Rev. B 81, 081305R (2010).
[282] A. Dienst et al., Nat. Photon. 5, 485 (2011).
[283] T. Yasui et al., Opt. Express 16, 1208 (2008).

chemicals[284], security screening, and biomedical applications (for example the detection of skin cancer), where the analysis of the chemical composition of the test object is also important besides visualizing the geometrical shapes of its internal structures. MSI can also be used for fast 3D THz computed tomography[285]. Since many materials have characteristic spectral fingerprints in the THz region, the spectroscopic analysis in this region is of interests as a new tool for material characterization. ELI-ALPS will provide an advanced MSI system.

- Control of matter by intense THz fields

The recent progress in further increasing the power of intense THz sources paved the way for high-field THz science. It is becoming possible to enter the regime of extreme nonlinear optics also in the THz frequency range. New insight will be gained on molecules, clusters, nanostructured, and bulk materials by exposing them to extremely intense THz fields and investigating the physical, chemical and biological processes they are involved in. The control of material properties and processes under the influence of extremely strong THz fields will open up new engineering possibilities.

Intense THz pulses may be used to trigger transitions between various conformational states of biomolecules, and use THz, optical, or XUV pulses for time-resolved studies. Recently, it was shown that exposure of artificial human skin tissue to intense THz pulses affects expression levels of numerous genes associated with skin cancer, which suggests potential therapeutic applications[286]. Orientation and alignment of molecules (with induced or permanent dipole moment) by strong THz fields[287] will allow studies of novel types of coherent or collective phenomena. Controlling the pathways of chemical reactions may also become possible as a longer perspective. Resonant control of ionic lattice dynamics was demonstrated by driving the ferroelectric soft mode in $SrTiO_3$ to the anharmonic regime by intense THz pulses[288]. This indicates that THz control over macroscopic order becomes possible, for example by switching the direction of ferroelectric polarization[289].

Control over degrees of freedom associated with spins have been demonstrated in antiferromagnetic[290], ferromagnetic[291], and multiferroic[292] materials. Ultrafast control of magnetization in ferromagnetic materials with a strong THz field suggests new opportunities for ultrafast data storage. Multiferroic materials exhibiting magnetoelectric effect are on the threshold to initiate a new generation of memory devices, the electrically controlled magnetic memories, due to the unique potential to manipulate their magnetic state via electric fields and their electric properties via magnetic fields. Recently, it has also been demonstrated that magnetoelectric materials offer new optical functionalities such as the one-way transparency and an efficient control of light polarization[293,294]. The synthesis of efficient magnetoelectric materials is limited by our present understanding of the coupling between magnetization and electric polarization on the microscopic level. THz pump−optical/THz probe spectroscopy can be used to gain a deeper insight into the coupled magnetization-polarization dynamics in magnetoelectric materials, with particular attention to the nature of electromagnon excitations that are a new class of elementary excitation only present in magnetoelectric media.

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- Post-acceleration of ion beams using THz radiation for hadron therapy

Laser-driven sources of accelerated ion beams offer a promising route towards compact accelerator structures. In case of protons a maximum energy of 60 MeV was reported[295]. Carbon ions with 185 MeV maximum energy were generated in the relativistic transparency regime[296]. Monoenergetic ion beams with 100 to 200 MeV energy are required by hadron therapy. High-energy proton beams with low energy spread and high number of particles are also needed for radiography, injection into conventional accelerators, and inertial confinement. However, the energy spectrum of laser-driven ion sources, usually using a thin-film target, is not monoenergetic, and the presently available energy is not sufficient for medical applications.

Various structures have been proposed for particle acceleration driven by optical fields. One example is to use evanescent waves[297]. However, the short wavelengths of visible and infrared lasers require scaling the geometry below practical limits imposed by typical sizes of particle beams. One solution for generating monoenergetic ion beams with sufficient energy for hadron therapy can be to use intense THz radiation[298,299]. This new approach holds promise to realize a compact, cost-effective ion source for hadron therapy with a much narrower energy spectrum as that of purely laser-based accelerators, and an easy spatial alignment of the accelerated beam. The same (sub-ps) laser can be used to drive the THz source as well as to generate a laser-accelerated ion beam to seed the THz post-accelerator segment. The generation of proton beams with high bunch charge may become also possible by generating gas plasma directly with high-energy THz pulses. The post-acceleration or direct acceleration of ions will require multi-mJ THz pulse energy at low (<0.5 THz) frequencies.

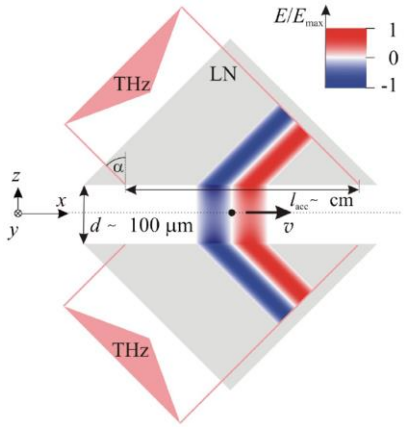

Fig. 61. Scheme of the THz-driven evanescent-wave post-accelerator (from Pálfalvi et al., Phys. Rev. ST Accel and Beams 17, 031301 (2014).

Outlook and future implications

ELI-ALPS will provide unique sources of ultraintense THz pulses and advanced THz spectroscopic tools with synchronization possibility to other light and particle sources of the facility spanning the wavelength range from infrared to x-rays. Such a unique source will significantly increase the discovery potential of ELI-ALPS by enabling (i) an unprecedented variety of spectroscopic and

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imaging experiments and the development of new technologies, (ii) a significant improvement of the characteristics of attosecond sources, (iii) the manipulation of relativistic electron bunches, which might also open up the field for new types of ultrafast x-ray sources, and (iv) the construction of compact sources of accelerated ion beams based on new approaches suitable for hadron therapy.

5.5 Innovative Laser-Based Accelerators for Diagnostic on Cultural Heritage

Motivation, state of the art

In the last decades, strong effort has been put into research of innovative techniques in the field of Physics and Chemistry applied to Cultural Heritage for both, diagnostic and conservations, based on the applications of new materials and investigation methods. The main challenge within the field of cultural heritage is to obtain the greatest number of information without provoking damage to the artifacts, i.e. having an efficient method for diagnostics. Another challenge is the conservation of artifacts without modifying its aesthetical appearance. Chemical and morphological information on artwork are mainly, obtained using surface spectroscopies or other methods based on particle accelerators and are most effective and sensible in laboratory.

Moreover, great interest and economical supports has been devoted from many International Institution to the preservation of historical, artistic, architectural, and cultural heritage. The European Commission and the UNESCO actions for Cultural Heritage are well-known, many historical and cultural sites in Europe benefit of Restoration and Conservation actions of both Institutions, with allocation of tens of billions of Euros for these actions. As example, in the upcoming HORIZON2020 Program, the European Parliament has recently included 'Cultural Heritage' as a priority in the proposed funding program of €80 bn from 2014-2015. The inclusion of cultural heritage in HORIZON2020 takes the form of a series of amendments focusing on the contribution of culture to research excellence, social cohesion and growth. A key section reads: "Accessibility and preservation of cultural heritage is needed for the vitality of engagement within and across European cultures by also considering the importance of cultural heritage as strong economic driver in a post-industrial economy and its contribution to sustainable economic growth." The sections of European interest in Cultural Heritage are specified in the "Carvalho Report – Specific Program". In particular, it can be read in the CA 18 (Industrial Leadership) the objective: "Applying design and the development of converging technologies to create new business opportunities, including the preservation of Europe's heritage and materials with historical or cultural value. Protecting the cultural heritage: assessment, monitoring and choice of conservation materials and techniques, with reference to the environment and energy management, use and maintenance, and integration into contemporary and historical urban surroundings and archaeological and cultural contexts".

In the last decades, strong effort has been put into research of innovative techniques in the field of Physics and Chemistry applied to Cultural Heritage for both diagnostic and conservations, based on the applications of new materials and methods. Many groups currently explore the possibility to realize equipment for diagnostic and conservation to be used directly on side in the archeological sites or in museums. The main challenge of the science applied to cultural heritage is to obtain as more as possible information without provoking damage to the artifacts, i.e. having an efficient method for diagnostics. The classical techniques of diagnostics and conservation (restoration and consolidation) require, generally, to move the artworks from museum, or archeological site, into a laboratory, or to make micro sampling. Chemical information on artworks (ceramic, bronzes, metals, pigments) is mainly obtained using surface spectroscopies (such as Photoluminescence, Raman, X-ray photoelectron spectroscopy (XPS), X-Ray-Fluorescence (XRF), Energy Dispersive X-ray Fluorescence (EDX)) in SEM), while morphological information can be gained with SEM. Complete chemistry of material bulks is known using sophisticated techniques of nuclear physics such as Proton Induced X-ray and Gamma Emission (PIXE and PIGE), fig. 62 shows a sketch of physical phenomena involved on

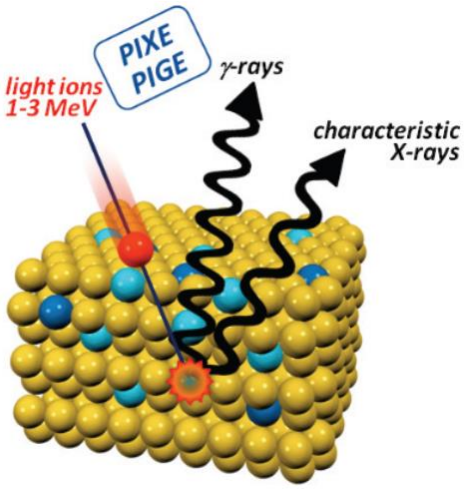

Figure 62. Sketch of Physical Phenomena involved in PIXE and PIGE Spectroscopies

both spectroscopies. All these methods are based on particle accelerators and are most effective and sensible in laboratory conditions (for example, in ultra high vacuum conditions and controlled temperature). Moreover, the use of conventional accelerators, with their very narrow range of proton energy, allows to study only the first microns of material's bulk, often confining the analysis to the corrosive patina or to the decoration on surface. Raman and Photoluminescence spectroscopy techniques, on the other side, require sophisticated spectrometers and lasers. SEM and XPS must be taken under vacuum conditions and PIXE and PIGE require the employment of large particle accelerators. Moreover, all these methods allow realizing only local analysis on artworks (beam spot sizes are generally in the order of mm^2). Several portable instrumentations, such as XRF and Raman, have been realized to make diagnostics directly in situ. However, all this portable equipment has efficiencies lower than when used in laboratory conditions.
Fig. 63 shows an example of a PIXE scan on a paint from "Uffizi" of Florence, the scan has been made by conventional proton gun (energy ranging from 1 to 3 MeV). The analysis with a smaller spot size (order of microns) allow analysing only small region of paint and necessitates scanning the paint in hundreds of point to have complete information. This elongates the analysis time and the possibility to damage the artifacts (i.e. increases of accumulate dose, long time in nonprotective atmosphere, etc…). Moreover, the elemental depth profiles obtained by classical PIXE and PIGE have a maximum depth of 2 - 20 microns, which are often the thickness of surface patina, preventing, then the correct analysis of material bulk In conclusions, conventional methods for diagnostic on cultural heritage present the limiting factor of not being very tunable and adaptable, making their use limited only to a certain field of energy range and to areas of

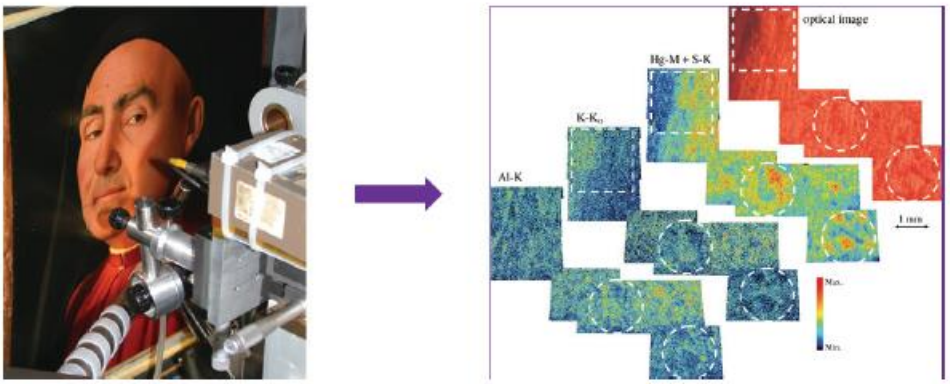

Figure 63. Exampleof PIXE to analyze the pigment's composition of the Trivulzio portrait by Antonello da Messina: The combined use of differential PIXE, fixed beam PIXE and external

microbeam scanning PIXE on some areas of the painting was essential in identify the different pigments present on different areas of the paint.

about few microns on surfaces. The advent of lasers, which can produce compact bunches of coherent photons, has opened up possibilities of laser-based particle acceleration, as foreseen in reference[300].The investigation of laser-driven proton acceleration and its use is currently challenging many research laboratories worldwide, with some significant results, in particular for the improved characteristics of laserbased particle sources such as compactness, its efficiency and its versatility and its tunability. Laserbased particle beam, moreover, presents the advantage of having a high current, strong laminarity at the source, short duration, and small source side. Focusing on laser-driven proton acceleration, today, existing multi-hundred-TW table-top laser systems generating on-target intensities of $\sim 10^{19}$- 10^{20} W/cm² can routinely reach proton energies of ~15-20 MeV with a typical laser-to-proton energy conversion efficiency of 1-6 % [301], a current in the kA regime and a laminarity at the source 100 times better than conventional accelerators. These parameters make them a desired candidate for innovative applications requiring one or more of the above mentioned properties, e.g. in the medical field[302], for inertial confinement fusion[303], warm dense matter studies[304] or hybrid accelerators[305] and allow to project their use in the field of Cultural heritage.

Laser driven ion beams for PIXE and PIGE diagnostics can allow obtaining:

- Complete chemical analysis on large area of artworks (order of cm^2) in opposite to smaller investigated surface of conventional accelerator (order of microns)
- "Layer by Layer" analysis, obtained tuning the beam energy from few MeV to tens of MeV (with few MeV of conventional guns it is possible analyzing simply the first micron of materials)
- Small damage for artifacts. The short duration of laser-driven beams and their higher current ensure sort duration of measurements and, then, small absorbed dos
- Nondestructive and multi-elemental analysis of trace elements with an excellent detection limit of up to 20 ppb.
- Elemental composition of magnetic films in which other methods don't have an enough mass resolution to resolve Mn-Fe-Co-Ni elements.
- Lattice location of impurities in single crystalline samples.

While there has been already many attempts to use and compare laser based protons to conventional accelerated particle in some applications (medical field, etc), no effort has currently been put to test the efficiency and usage of laser-driven proton acceleration for purposes of cultural heritage. This

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opens up new opportunities to apply the field of laser-driven particle accelerators on this research domain and to test and develop innovative and improved methods of diagnostics with new particle sources. The current proposal comes very timely, since ELI facilities in secondary source are currently pursuing the setup of laser-driven beamlines but have not yet prepared applications (at least not in the field of cultural heritage) of those innovative sources. This proposal could therefore perform breakthrough results showing the innovative character of the source and more over in a very sensitive field such as cultural heritage.

Research directions and activities

The aim of this project is to improve the field of Diagnostic in Cultural Heritage using Laser-accelerated beams. The main objectives are the development of much sensitive and performing equipment for diagnostics on the different materials of interest for cultural heritage (i.e., stones, bronzes, marbles, pottery, etc…). As methodology, the project will make use of laser-driven proton sources for generating X and Gamma ray on materials, the analysis of produced radiation will allow to obtain a complete chemical analysis of material's bulk, with a spatial resolution of few microns. Laser-based particles have great advantages compared to conventional proton accelerators:
1) the possibility to tune proton energy in a broad range from few MeV to tens of MeV, allowing a "layer by layer analysis",
2) a high flux of particles, allowing to deposit more energy,
3) shorter pulse duration, lowering the damage during the analysis. All these properties will allow distinguishing the corrosive patina from main component of artifacts or from pigments and decoration on surface with enormous benefits for better design restoration and conservation project. Finally, the results of proposed experiments can open the field of new laser-driven particle accelerators to the research field of Cultural Heritage driving ELI-ALPS to be an European reference facility for analysis on Artworks and Archeological Artifacts.
In this proposal laser-generated protons will be uses to study their applicability in the domain of Cultural Heritage. To our knowledge, only little research has been currently conduced in the field of applied laser-driven ion beams, with most of the application dedicated to radiography, astrophysics and warm dense studies. However, no attempt has been made to test if laser-driven ions are suitable for irradiating historical artifacts. Compared to "classical" proton accelerators used in Cultural Heritage, laser-driven ion beams have the strong interest of being
more versatile – the energy can be easily selected with a selector and adapted for every shot **higher in current** - selected particles can generate currents of nC/ps, i.e. in the kA regime. This might evidently be too strong for the requested application, but on the other side it is quite easy to downscale the current limiting the number of particles. The number of particles also allows to irradiate in a single shot a larger surface and thus cover a larger surface in the analysis than using conventional small scale accelerators.
shorter in duration - the short pulse characteristics of laser-driven protons has never been tested on samples. Due to longer relaxation timescales of most reactions within the sample, it is likely that the sample will suffer less from a shorter-compacter laser-driven irradiation (in the ps scale) than from a conventional accelerator.

In detail, the project addresses the following research topics:
01 Study the use of laser-driven particle sources for cultural heritage applications and compare them to conventional accelerators

02 Perform numerical simulations for investigating the phenomena occurring during irradiation, using both Particle In Cell and hybrid codes

03 Measure the improvement and the tunability/versatility of laser-driven sources (e.g. regarding stopping power and penetration depth)

04 Study the influence of particle (protons) and X-ray irradiation on the stability of the protective structure often applied on artwork surface

05 Study of the effect of accumulated radiative dose during particle irradiation on artifacts materials

Preliminary studies and methodology

Preliminary simulations have been performed on several materials of interest for Cultural Heritage (i.e., bronze, marble, glass, noble metals, ceramics, etc….) to verify if the required onditions for diagnostic can be obtained on the ELI laser facility. For that, a Hydrodynamic Code and Mathematica scripts is used in which as proton heating source

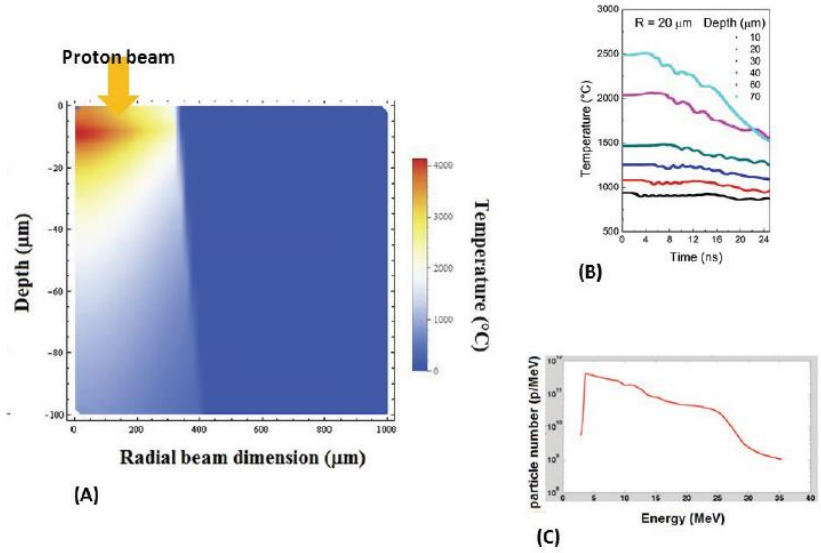

Figure 64.(A)2D Temperature map, obtained with the Hydrodynamic code, 100 ps after irradiation, the 0 level indicates the target surface in front of the proton beam;
(B) Temperature evolution in time, monitored in different target depth positions. The temperature has been measured at a fixed radial position of R=20 μm;
(C) Laser---generated proton spectrum as obtained by the TITAN Laser during the campaign October 2012 from a 15 μm gold foil.

from laser-generated protons obtained in previous high-power laser experiments are inserted (reference: a proton spectrum obtained on a hundred TW system in delivering ~150 J in 800 ps (TITAN laser at LLNL)). Although the laser-parameters of the TITAN laser are not the same as those of the ELI-ALPS laser, it is expected proton parameters to be sufficiently performing to generate similar proton parameters and thus guarantee the sufficient parameters to perform the experiment.
In order to take this into account the proton yield was lowered by a factor of 10 in the spectrum. For testing bronze metals, as sample a 100 μm target using the chemical composition of modern bronze (90% Copper and 10% Tin)is used . It is tested if the proton beam irradiation generates temperature higher than melting point (about 1100 °C for modern bronze and ranging between 1100 and 1500 °C for ancient bronzes, which have different chemical composition) and checked the penetration depth levels reached by proton with different energies . Figure 64A shows the temperature map, obtained with our code of a 100 μm bronze target irradiated by the laser-generated proton beam, 100 ps after irradiation. Images are taken at different instants of time from 1 ps to 50 ns and the obtained time evolution of temperature on the sample is illustrated in fig. 64B. The temperature reached in different regions of the target is strictly dependent by the proton energy and on its distribution, the energy profile and temperature as function of depth and radial dimensions of the proton beam are showed in fig. 65.The temperature generated by the high energetic protons

(with E > 5 MeV) is higher than the melting point of bronzes (about 1200 °C). However, high temperatures are confined in an area relatively narrow respect to the entire area covered by the proton beam (red area in fig 64A) while for large zones on the target surface and bulk, the temperature remains below the melting point.
The red circles in fig. 65 indicate the energy intervals available for reaching temperatures less than melting point and the corresponding depths in target bulk. With proton energy ranging between 1 and 5 MeV the temperature is lower than the melting point and the concerned layers in the bronze have a depth ranging from 0 (surface layer) up to 40 microns (compared to 20 microns, maximum reached by conventional proton accelerator). It is therefore possible to take an analysis of different layers of the target bulk opportunely selecting the mean energy of the proton beam. Using an energy selector, i.e. a device that captures only a part of the beam and reduces the energy-spread it is possible to perform an analysis on different layers considering a layer-thickness precision of a few microns, and scanning layer by layer from the surface to the inner region of the bronze artworks.
The detectable depth range changes with materials and reaches values of about 80 microns for materials with high melting point such as silicate. Since the total proton number generated by the laser-driven sources is limited, one needs to find the best working point for the energy selector as trade-off between energy spread and number of usable particles. Reducing the energy spread means reducing the total number of particles impinging the target. However, a too-large energy spread means irradiating a larger layer of material which makes the diagnostic and analysis less precise.
It has been verified that a proton beam with mean energy 5 MeV, energy spread of 0.5 MeV (full width at half maximum (FWHM)), represents a good compromise between available particles (of the order of 1011 particle/MeV) and reachable layer-depth (until 100 microns). Figure 66 shows the results obtained by Hydrodynamic simulations selecting the proton with energy of 5±0.5 MeV, the temperature reaches the maximum value of 350 °C while proton beam reach a depth of 100 microns in the target with a fluence of about 5*1011 particles/pulse.
The experiments shall be carried out on high-power lasers that are suitable for laser-driven ion acceleration, such as the ion-beamline available on ELI-ALPS. With its foreseen proton parameters (energy ranging between 1 MeV and 40 MeV, current of the order of nC, and pulse <fs) the beamline can ensure the versatility, high current, and short duration properties necessary for developing an effective diagnostic on artifacts and artworks.
For testing the feasibility of these experiments, it is planed to operate in three different phases:
1- Simulation of interaction between energetic proton beams and matter for better define the beam parameters
2- Tests on commercial materials of interest for cultural heritage (bronzes, ceramics, marbles, stone, etc,…) and realization of "database" of proton beam condition for each material of interest for Cultural Heritage
3- Convention with the much important European Museums for tests on artworks

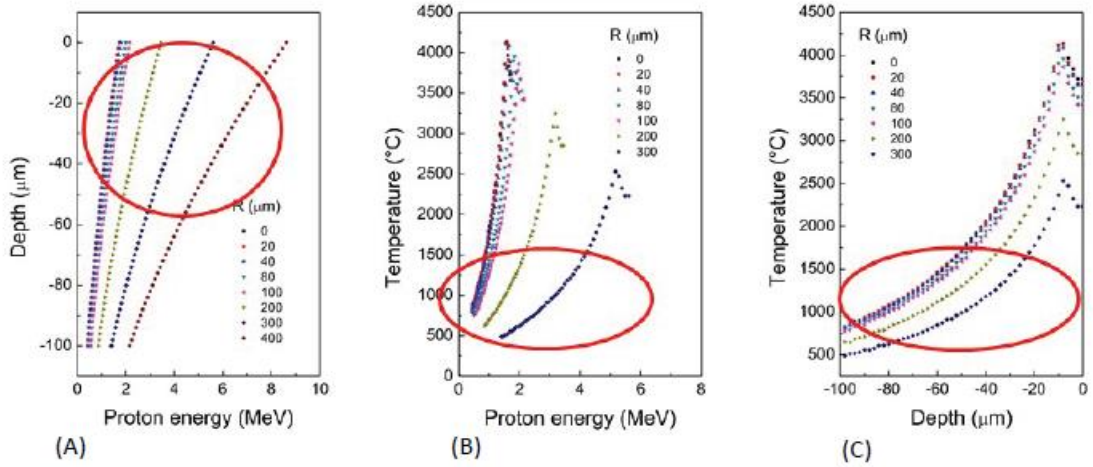

Figure 65. (A)Depth reached by protons as functions of energy for several value of beam radial dimension. The proton energy has been estimated in the hypothesis that the protons are in linear uniform motion between source and target. The 0 level indicates the target surface in front of proton beam(B). Temperature distribution on depth after 100 ps of irradiation as evaluated from Hydrodynamic simulations. (C) Temperature profile as function of proton energy, as obtained comparing the data in A and B. The red circle in all figures indicate the ranges of energy, temperature and depth available for PIXE and PIGE diagnostic.

In the first phase, shall be studied all details of the laser-matter interactions and contribute in preparing the experiments. Simulations shall identify the required laser and proton conditions in order to find best interaction conditions, ensuring that the artifact is not damaged but at the same time the ion flux is sufficient to ensure the usability of the diagnostics. The simulations, as showed in the preliminary simulations above, must identify:
1- Energy range and pulse duration for the very different materials of interest (pigments, marbles, bronzes, etc…)
2- The best energy range for analyzing different layers of materials (form few microns to few cm)
3- The absorbed doses for each layer of materials and the temperature reached on artifacts surface and bulk

A complete simulation can be realized using a PIC code.

The objective of the second phase is to study the best way to generate laser-driven ions that are suitable with the required application in the Cultural Heritage field. The main object will take care, based on simulation results, of which target is most suitable for generating the best interaction between proton and the materials of interest for cultural heritage. It has been shown that different interaction target can yield to different ion spectra. As an example, plastic targets enhance the lower energy part of the spectra, but don't allow achieving very high proton energies. Moreover, the produce more filamentation, the proton beam could therefore result less homogeneous. More homogeneous protons can be achieved with metallic targets or exploded foil targets (but in that case the setup might become more complex).
As already mentioned, the irradiation of artifacts by using laser-generated protons might require some beam shaping. While the divergence of the laser-generated particles does not

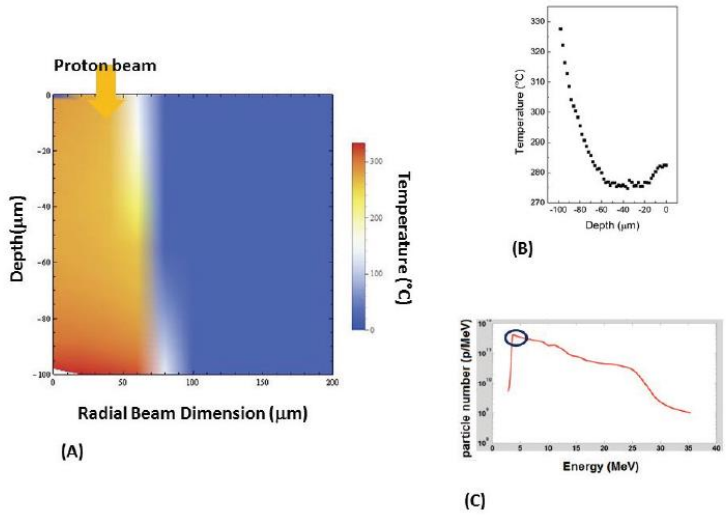

Figure 66. (A) 2D Temperature map, obtained with the Hydrodynamic code, 100 ps after irradiation, selecting only particle with energy of 5±0.5 MeV; (B)Temperature changes as function of depth. The temperature has been measured at a fixed radial position of R=0 μm; (C) Typical

laser--generated proton spectrum as obtained by a high--power laser (TITAN) from a 15 mm gold foil, the blue circle indicates the proton range used for this simulation.

particularly affect the measurements, since the artifacts need to be irradiated with less flux than typically generated on laser-driven ion beams, it might be needed to perform an energy selection in order to irradiate the artifact with only a specific energy (with its energy-spread). There are currently several easy solutions for performing this. One is using a beam selector that is generated by a classical "chicane"[306] and that can be used to tune the beam energy for investigating the different layers of materials.

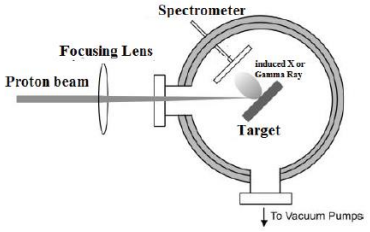

Figure 67. Sketch of experimental setup for in vacuum experiments of phase 2

Performing experiments on laser driven proton acceleration to irradiate artifacts is the core of proposal. A sketch of proposed experimental setup is illustrated in fig. 67, where the spectrometer indicated for diagnostic will be an X-ray or Gamma ray detector as we perform PIXE or PIGE experiments.

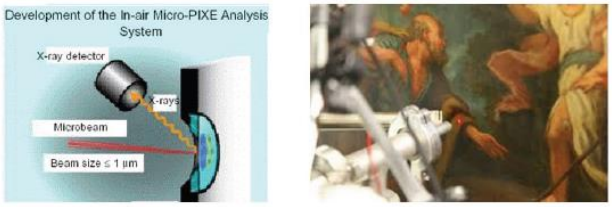

Figure 68. (A) Sketch of experimental setup for "in atmosphere"measurements in phase 3; (B) Example of application to an analysis of paints

The experiments of phase 2, will performed in under vacuum conditions to better calibrate the proton beam parameters while the experiments of phase 3, performed on artworks, will be integrated in air conditions (fig. 68 shows an example of classical PIXE conducted in atmospherically conditions).

5.6 Nanomaterial growth by laser-driven proton irradiation

Motivation, state of the art

The enhancement and control of growing techniques for nano and nanostructured materials with well controlled properties (i.e., crystallinity, shape, dimensions, band edge, etc..) is one of the current key challenges in nanoscience and nanotechnology. The *a-priori* definition of material properties and the setting of exact growth parameters is crucial for obtaining materials suitable for applications.

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Nanomaterials are of strong importance in many fields, ranging from material science (mechanics), energy production (nuclear plants and green energy), medicine (surgery) and aero spatial application[307,308]. The possibility to obtain routinely nanocrystals with controlled shape, dimensions and crystallinity is a challenge that many research groups are pursuing and that is considered strategically important for manifold applications. The most effective example is the application to research in new materials for photovoltaic devices, field of strong interest for European Commission in the last decades. A budget of €6 million has been allocated to non-nuclear energy research for the period 2014-2020 in the photovoltaic program of Horizon 2020, while out of this figure, more than €200 million is earmarked to support European Institute of Innovation and Technology activities[309]. In this field many projects have been submitted to European Commission actions, involving Research Institutes and Companies of all European countries and the 90% of those are based on the controlled growth of nanomaterials and nanostructures. The realization of mesoporous or nanostructured electrodes can, in fact, strongly improve the efficiency of a photovoltaic device from the currently 15% to 20-25% (enhancement of 30-50%).

The main problem in the definition of a standard growth protocol is the individuation of parameters to generate the conditions of temperature and pressure required to produce well-defined structures in very short timescales (ps-fs), necessary for the nucleation of particles with dimensions of tens of nm.

A way to achieve this is the irradiation of matter, in vacuum or in solution, by an energetic proton beam with short duration. The irradiation of a bulk target by high energetic particles (in particular protons) can generate the temperature and pressure conditions required to grow crystalline structure, while the short pulse duration limits the nucleation time to the range of ps-ns ensuring the stop of nucleation at crystallinity phase without aggregation of amorphous structures.

In the other hands, ultra-intense high-energy short laser pulses, such as those obtained at ELI-ALPS, enable the production of multi-MeV protons and ions beams of low divergence, short duration (<1ps) and high current. Compared to conventional proton accelerators, they have the advantage of being much shorter and higher in flux. In recent times, much effort has been put in order to control the proton production at the source and to tailor the beam, including increasing its energy, in order to make it suitable for many potential applications. For some applications, such as in the medical field, proton beam parameters still need to be improved in order to become a valid candidate as substitute of classic accelerator. However, little has been done in order to use laser-generated protons for multidisciplinary applications (with industrial interest), such as material science or nanotechnology.

In this proposal, we want to use laser-generated protons as obtained on the ELI-ALPS HF ion beamline in order to explore these fields based on the results of theoretical simulations and of preliminary experiments on laser-driven proton irradiation of a silicon wafer performed on Lawrence Livermore National Laboratory (California). From our calculations and experiments, laser-driven proton beams represent an ideal candidate to perform irradiation of nanomaterials since they can be tuned in energy, flux and have the advantage of being short and intense. This yields to achieving high temperatures and high pressure in very short timescales, a key requirement in the "nano-world".

The main objective of this proposal is, then, to use laser-generated protons to grow and obtain nanostructured surfaces and colloidal solutions where the constituent nanomaterials are nanocrystals with well defined shape, dimensions and crystallinity. We plan to control the

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[309] Horizon 2020 call for research in green energy: http://ec.europa.eu/programmes/horizon2020/en/h2020-section/secure-clean-and-efficient-energy

nanocrystals properties tuning the laser-accelerated proton beam characteristics (energy, pulse duration and fluence).

Scientific Context (methods for nanomaterial growth and application of laser driven proton beam irradiation)

In the past two decades, nanowires, nanoparticle, nanocrystals, have been used for a wide variety of materials; among those, metals and semiconductor play a key role. The great interest in these new materials is related not only to their properties and potential applications but also in the fact that many properties can be tuned by tailoring the size, shape, surface functionality or crystallinity of nanocrystals[310]. A typical example of the size-dependent properties of nanocrystals is the continuous fluorescent emission of quantum dots, such as CdSe or CdTe, which covers the entire size of a visible region as function of the particle size. Many results clearly demonstrate that the controlling of the size, shape and crystallinity of nanocrystals is critical for tailoring their properties and optimizing their performance. As example, the band edge of 2D or 3D bulk semiconductors can be easily modulated and controlled realizing 2D, 1D, or 0D nanostructures[311,312]. Several theoretical and experimental works[313,314,315] demonstrated that the dimensions of nanoparticles are strictly related to the shift of band edge compared to those of bulk semiconductors.

In this scenario, a well-defined control of the particle's shape and dimensions is, then, the main aim and objective of researchers. However, the growth processes of nanomaterials are, often, related to chemical reactions without the possibility to define a-priori shapes and dimensions and, then, without control on the electronic and crystallinity. At current, the control synthesis of nanostructures is still in an embryonic state and, often, related to a-posteriori morphological analysis on grown nanomaterials. Empirical procedures have been developed for the preparation of a wide range of semiconductor, ceramic, and metallic nanocrystals and the choice of methods and growth parameters are often specific of a single research group. The syntheses methods are strictly related to experimental parameters (i.e. pH, precursors, temperature, time, capping agent, etc…) and a little variation in each of them can strongly influence the properties of the samples[316] resulting in non-uniform samples. For example, changes in temperature or time conditions can strongly modify the shape and the dimensions of produced nanomaterials and the roughness of nanostructured surfaces preventing their insertion in electronic devices (high roughness causes high contact resistance) or changing their mechanical properties (i.e. heat conduction, mechanical resistance, etc…). Moreover, for high reaction times (> ms) the Ostwald – ripening mechanism starts and big amorphous aggregates

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with dimensions of a few microns appear in the solution, the size distribution becomes highly broad, and the quantum size confinement effect (dimension < 10 nm) is completely lost. Finally, the chemical methods cause the intrinsic formation of a surfactant shell around nanoparticles or nanostructures which strongly influences the electronic properties (i.e., band edge).

On the other hand, "chemical-free" methods, as Pulsed Laser Ablation (PLD) and laser Ablation in Solution (LASiS), are employed for nanoparticles growth from mono-atomic target, with a control in particles dimensions ranging, in the best cases, from 20% to 25%. Furthermore, the application of laser methods at insulating and semiconductors is only studied little and implemented, because it is difficult to obtain uniform colloidal nanoparticles from a bi-atomic or multi-atomic target. In general, High Energetic UV lasers (i.e. KrF), in controlled atmosphere or in vacuum, induce material ablation from surface and a formation of a high energetic plasma plume. In these conditions, the irradiation causes the evaporation from surface of high energetic and separated atoms, molecules and clusters, which reach the deposition surface and aggregate randomly forming nanoparticles of different atomic and molecular species without crystal shape.

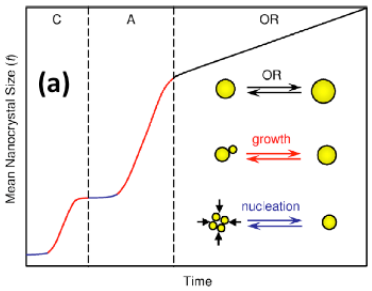
A general kinetic profile depicting the regimes of nanocrystals growth is given in Figure 1. An initial induction period and size increase corresponds to classical nucleation and growth (regime C), which produces primary nanocrystals in diameter range of 1-3 nm. That regime may be followed by a second induction and growth period associated with aggregative nucleation and growth (region A). For long aggregation time, nanoparticles aggregate forming greater amorphous cluster (region OR). Hence, great efforts have been necessary in defining a simple and repeatable synthesis process with a perfect control of produced nanocrystals.

To date, achieving a fine control over nanocrystals monodispersivity, structure, composition and defects is still a strong challenge.

Objectives

As stated in the summary section, the main objective of this proposal is the definition of a controlled synthesis process driven by proton irradiation of precursor components.

We plan to realize a controlled growth process for nanocrystals with dimensions less than 10 nm (quantum dots) based on two well-established methods for growing crystals and amorphous nanomaterials: (i) "melt methods" for macrocrystals induced by laser-generated proton beam irradiation, and (ii) the nucleation of particles in colloidal solutions irradiated by laser-generated proton beams. The pressure and temperature condition produced in our bulk sample by high energetic proton irradiation will ensure the required conditions for growth of crystals while the short duration of the irradiating pulse will narrow the growth in the A - C region in fig. 69 stopping the amorphous nucleation of big particles. The shapes, dimensions, and crystallinity can be controlled varying the beam parameters and monitoring the growth in the first nucleation steps (regions C and A in fig. 69).



(b)

Growth condition	Condition A	Condition B	Condition C
Temperature gradient (°C/cm)	20	20	20
Growth temperature T_G (°C)	640	640	690
Vapor control temperature T_V (°C)	380	440	520
Growth time (days)	14	14	14
Se vapor pressure P_G (Torr)	3.4	14.2	70.4

Figure 69a) Schematic representation of nanocrystal growth including classic nucleation and growth, aggregative nucleation and bog cluster formation ; b) Table - Details of crystal growth conditions for a crystal of GaSe

The artificial synthesis of macro-crystals can be roughly classified into three groups, i.e. solid-solid, liquid-solid and gas-solid processes, depending on which phase transition is involved in the crystal formation[317]. Among these three categories, the liquid-solid process (including the Czochralski or Bridgman methods) is the most popular method for growing single crystals (perfectly controlled) at large scale. More than half of the industrially made crystals are nowadays obtained by this technique including the crystals of semiconductor of II-VI compounds (CdS, ZnS,…). Melt techniques involve heating polycrystalline material in a container above its melting point and slowly cooling it down from one end where a seed crystal is located. Single crystal material is progressively formed along the length of the container. Crystal growth is performed under several vapor pressures and growth time was kept to be 15-20 days for a crystal with thickness in the order of hundreds of microns. The table in **Hiba! A hivatkozási forrás nem található.** shows details of crystal growth conditions for GaSe[318].

The irradiation of matter with a high-energy laser-generated proton beam opens the possibility of reproducing the temperature and pressure conditions for crystal growth in the very short time required for nanocrystal growth[319]. As an example, the irradiation of a solid target of Al with a proton beam of 20 MeV, 10 protons and a pulse of few ps generates a heating at temperature of about a few eV in 20 ps[320]; at this temperature, the matter is in a plasma state where the atoms can start to nucleate forming nanoparticles. We have performed simulations showing that the requested temperature and pressure ranges for growing nanomaterials can easily be achieved using laser-generated protons (see later). The nucleation time will be in the order of μs if in this temporal transient the sample temperature rapidly decreases below the melting point. If in this time interval the pressure is in the order of GPa, the nucleation is made along the crystallographic axes and nanocrystals growth on the target surface. This methods can obtain nanostructured surfaces highly ordered and controlled but without possibility of detaching nanoparticles from substrates.

On the other hands, one of the most useful and promising methods to growth nanoparticles is the ablation of a bulk target in solution[321]. In general, this growth technique is achieved irradiating by a laser with a pulse in the range of ps and ns a metal target immersed in an a-polar solvent (acetone or ethanol). The irradiation causes the desorption of atoms from surface to solution, in a warm region where the particles nucleates and aggregate in times of the order of 10^{-6} – 10^{-4} s. That high nucleation times collocates the particles growth in the region

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[318] F. P. Doty, J. F. Butler, J. F. Schetzina, K. A. Bowers, Journal of Vacuum Science and Technology B 10, 1418 (1992)
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[321] V. Amendola, M. Meneghetti, J. phys. Chem. C, 113, 4277 (2009); V. Amendola, V. Meneghetti,Phys. Chem. Chem. Phys., 15, 3027 (2013)

OR of graph in fig. 1 preventing the formation of ordered crystallinity structures. Therefore, it has been found that the proton beam irradiation can be used as a powerful alternative reducing source for the synthesis of amorphous nanoparticles in solution: gold nanocrystals and nanorods were achieved through the proton beam irradiation (energy of about 20 MeV) of a solution containing gold ions. The heating of the solution and the very short heating time generated by the laser-driven proton beam irradiation can confine the nucleation time in the region C and A blocking the amorphous aggregation.

In the scenario described above, the main scientific mission of the current proposal can be summarized as follows:

1. Direct growth of nanocrystals on metallic and semiconducting surfaces by induction of a "melt state" with temperature and pressure conditions for crystal growth;
2. Realization of colloidal solution of nanocrystals by irradiation of metallic or semiconducting target immersed in a-polar solvent;
3. Exact control of crystal shape, dimensions, and crystallinity tuning the proton beam parameters (energy, pulse, and fluence)

Methodology

The proposed experiments could be structured in two independent series of measurements: (i) the study of controlled growth of nanocrystals directly on target surface and (ii) the realization of controlled colloidal solution. Both experiments will be devoted to the realization of metallic and semiconducting nanocrystals, using as probe amorphous target of noble metals (Au and Ag), two of the most common semiconductor (Si, CdS and ZnS) and Carbon based materials (amorphous graphite and carbon nanotubes). These target materials are most common in several applications and their production with different chemical and physical methods is well studied, so that it is possible to compare our results with those existing at present in literature or industry, immediately defining the advantages and disadvantages of the new defined methods and the quality of produced nanocrystals.

We plan two series of experiments. In the first series we will irradiate a metallic, carbon based, or semiconductor target (in vacuum) with laser-generated protons. In the second series we will use the laser-generated proton beam to irradiate a target immersed in a-polar solvent (acetone or ethanol) in a cuvette. For each series – depending on the results – we will change the irradiation characteristics by playing on the proton beam parameter.

We plan to vary for each target, in both series of experiments, mean proton energy, pulse duration and fluence. The nanocrystals growth will be monitored, in situ, by two spectroscopic techniques: X-ray diffraction (XRD) to verify the crystallinity and the measurements of the bulk (or solution) optical absorption. This technique allows estimating, directly "on-line", i.e. after the shot, shape and dimensions of nanocrystals (following the exact Mie electrostatic theory). Droplet of colloidal solution will be deposed on a Silicon Dioxide surface immediately after irradiation and optical absorption measurements, to prevent the possible aggregation in solution. All the produced samples will be analyzed, in successive stages, with the most common spectroscopic and microscopic techniques to better understand the chemical and morphological properties of produced samples and the exact influence of proton beam parameters.

We have performed preliminary simulations and experiments to verify if the required conditions for the nanomaterials can be obtained on the ELI laser facility. For that, we used a Hydrodynamic Code and matlab scripts in which we have inserted as proton heating source laser-generated protons obtained in previous experiments (proton spectrum obtained during a Titan (LLNL) campaign in October 2012). Although the laser-parameters of the TITAN laser are not the same as those of the ELI-ALPS laser, we expect proton parameters to be sufficiently

performing to generate similar proton parameters and thus guarantee the sufficient parameters to perform the experiment. We took as sample a 15 μm Si foil, which can easily be stand-alone mounted on a target holder. In order to achieve temperatures within the target close to the fusion temperature of Si (~1500 °C), we need to put the target 2.5 cm away from the proton source (which is a 15 μm Au foil), geometrically diluting the proton flux of the source. 70 shows the temperature map, obtained with our code, of a 15 μm Silicon foil irradiated by the laser-generated proton beam. Images are taken at different instants of time. Adapting the proton flux, one can see that we are able to heat the Silicon foil (and its back surface, where we plan to check the nanomaterials), to temperatures around ~1500 °C within a few tens of ps. We also see that the temperature stays constant and homogenous for a sufficient period of time in order to catalyze and process the nanomaterial reaction (reaction time <1 μs). We see that after a few ns the temperature of the target has cooled down to temperatures below the melting point (stopping the nanomaterial reaction). Playing on the distance and on the spectrum of the laser-generated proton spectrum we are able to adjust the desired temperatures within the target in order to adapt it to the requirements. We have performed similar simulations with Silicon, where the gap between the melting and the boiling point is much higher. This will warrant that – even if we have fluctuations in the proton spectra and hence in the obtained target temperature – our target/sample will be heated above the melting temperature and below the boiling temperature.

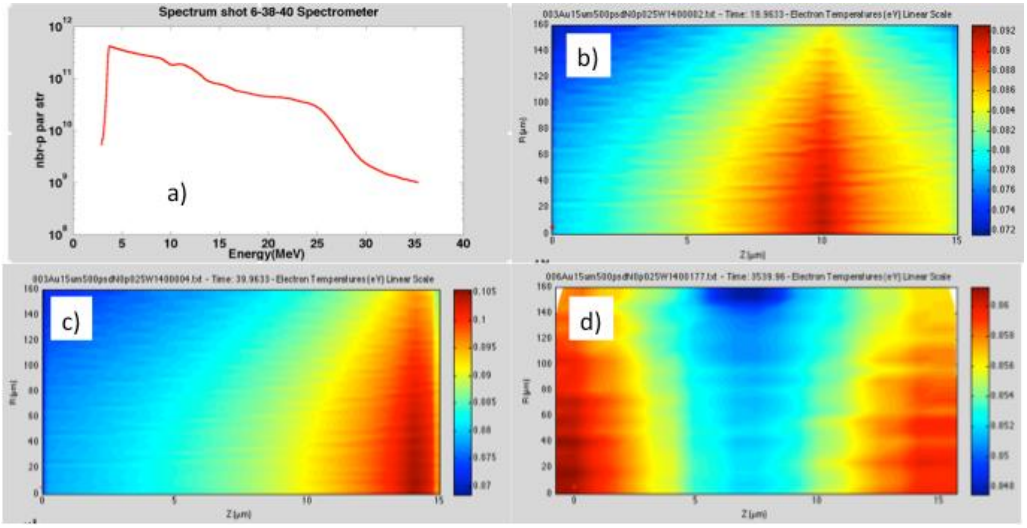

Figure 70 a) Laser-generated proton spectrum as obtained on the TITAN (LLNL) laser in October 2012 from a 15 μm gold foil. b) temperature map, obtained with the DUED code, of a 15 μm gold foil irradiated by the laser-generated proton beam. The images shows the temperature in eV, 20 ps after irradiation. The target is located at 13.5 cm in order to adapt the proton flux to the requested temperature conditions. c) same as in b), but 40 ps after irradiation. d) same as c), but after 3.5 ns.

Regarding the realization of colloidal solution of nanocrystals by irradiation of metallic or semiconducting target immersed in a-polar solvent, we have checked that the laser-generated protons on ELI have sufficient energy to pass the solvent and irradiate the sample. Regarding this kind of targetry/sample, we plan either 1) to use a longer "feed-through-finger" that can be put into the beam path of the laser-generated protons and that can be "in air". This structure can be made by 1-2 mm thick aluminium. Stopping power tables for protons, such as obtained by the code SRIM, indicate that proton energies of ELI are sufficiently energetic to pass through the aluminum structure and the solvent in order to irradiate the sample (20 MeV protons are stopped in less than 2 mm of Aluminum, whereas 8 MeV protons can penetrate through 1 mm of solvent (e.g. acetone)).

Alternatively, we can put a vacuum proof "minichamber" inside the target chamber, with the same material/wall thickness. In other experimental runs, we were able to use a 50 μm window of diameter 2 mm which was able to withstand the pressure of the vacuum. Since the stopping power conditions wouldn't change, we are sure to have sufficient proton energy and flux for irradiating the target/sample.
Since we might be interested in increasing the pressure of the irradiated sample, we plan to use a "split-beam" confirmation with two focusing parabolas. One laser-beam would be used for the proton generation, while the second beam path would be used for generating pressures on the back side of the target. A sketch of the foreseen experimental setup is indicated in 71. First part of the beam (most of it) is used for generating the laser-accelerated protons. For the direct growth of nanocrystals by induction of a "melt state" the second part of the beam is used to generate a pressure on the back side of the target (if really needed). This pressure can be adjusted using a partially reflecting mirror and selecting the amount of split-beam energy going to the second beam-path. Alternatively we could also use the East-beam for achieving this purpose, although its temporal duration is much longer and therefore less adapted for generating pressure on a quick timescale on the target rear surface. Previous experiments on high-power lasers have reported achievements of pressures of >10 GPa on the target rear surface[322] nanomaterial synthesis, which is between 2-5 atm, i.e. 200-500 kPa. Since the synthesis process is not very sensitive on the obtained pressure, we have sufficient margin and precision on this beam-path to achieve the desired objective. In the case of realization of colloidal solution of nanocrystals by irradiation of metallic targets immersed in a-polar solvent, the second beam path (generating the pressure) will not be used since it is not necessary for the required experiment.

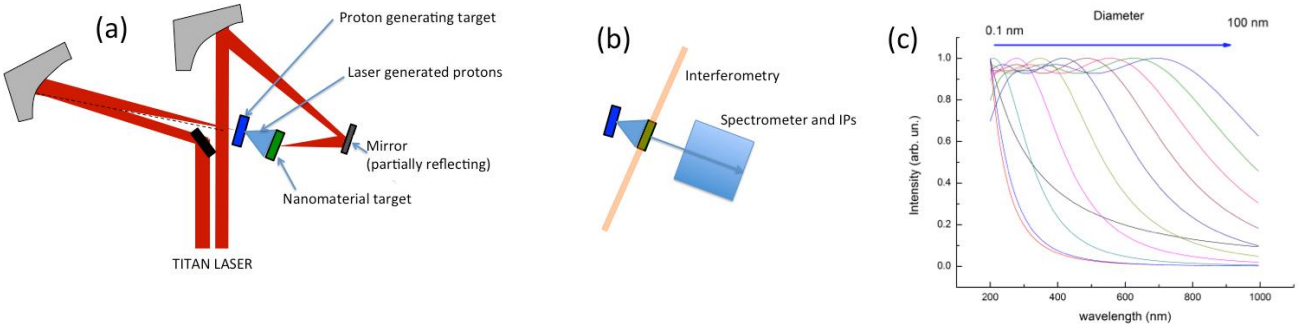

Figure 71 a) Sketch of the experimental setup for the proposed experiment (part one), making use of the spit-beam configuration. One beam path will generated the laser-accelerated protons and the second beampath a pressure on the back side of the target. b) sketch of the used diagnostics. c) extinction cross section for Al nanoparticles with diameters ranging from 0.1 to 100 nm.

As diagnostic, we plan to use a Thomson parabola and IPs for checking/determining the obtained proton spectrum impinging the sample to be irradiated. We also plan to use interferometry for the first part of the experiment in order to check the backside of the target and the pressure generation from the second laser. With both diagnostics we have extensive experience to measure the requested parameters.

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6 Scientific prospects of using Mid-IR laser sources at ELI-ALPS

In this chapter we present scientific applications envisioned to be done with mid-IR ELI-ALPS laser and X-Ray sources, one being already described in chapter 9.4.1 (charge migration in complex molecules). The activities range from AMO physics and physical chemistry to attosecond magnetism and condensed matter science.

Motivation, state of the art

Following the rapid advances of laser technology, the development of table-top extreme ultraviolet (XUV) sources has provided novel ways to achieve real-time manipulation of electron dynamics in atoms and molecules in gas phase, and more recently in condensed matter. These laser-driven strong-field processes are based on the rescattering model that explains how an electron, born at the peak of the laser pulse, can be driven back and can interact with the parent ion[323]. There are two major processes occurring at the instant of the rescattering: the electron can recombine with the parent ion and radiate a short burst of XUV radiation, or it can scatter off elastically (inelastically). Thus far, both attosecond XUV radiation and ultrashort electron pulses have been used as novel spectroscopy tools. To extend these applications to higher energies, though, long-wavelength laser sources have to be used. The cut-off energy of the X-Ray beam radiated in the process of recombination is proportional to the ponderomotive energy (Up) of the driving laser field and it scales as 3.17Up, while the scattered electrons can gain an extra energy from the laser pulse and can reach a cut-off energy as high as 10Up. Ponderomotive potential, on the other hand, scales as λ^2, where λ is the wavelength of the diving laser field.

Thus far, the majority of applications done with laser-driven electron and XUV sources have been utilizing lasers operating in the near IR part of the spectrum with a typical wavelength of 800 nm. Only recently, though, high-power mid-IR sources, with the wavelengths ranging from 1300 nm to 3900 nm, have been used as driving sources of high-energy electrons[324,325] and coherent soft X-Rays[326]. A simple calculation, comparing the energy an electron can gain in a typical 800 nm laser field versus a 3.2 nm laser driver, shows that the electron cut-off energy in the mid-IR field is 16 times higher for the same laser intensity. This means that, for a laser intensity of $5x10^{14}$ W/cm^2, the X-ray cut-off energy changes from 110 eV in case of 800 nm to 1530 eV in a 3200 nm laser field driving the harmonic generation process in argon. So far only a handful of experiments have been done that utilize mid-IR-driven electron pulses to self-image the structure of small atoms and molecules, while no experiments have been reported that utilize broadband X-Ray sources driven by mid-IR sources. The main reason for the lack of scientific applications done with coherent, tabletop X-Ray sources is a low photon flux, limited by low-repetition-rate mid-IR sources. Here, we present a road-map of experimental activities planned at ELI-ALPS that would allow for a new class of experiments to be performed in gases and condensed mater, extending the attosecond scientific application to the water window range. In the first phase, this would be done with the ELI-ALPS 100 kHz, 3200 nm laser source.

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[326] T. Popminchev et al. *Science* 336, 1287 (2012)

6.1 Mid-IR driven attosecond X-Ray sources

In the first experiments reporting on the ultra-broadband, coherent X-Ray spectra, a 3900 nm laser driver was used in combination with a high-pressure helium-filled waveguide. The experimental approach for generating coherent X-Ray radiation with the 1.6 keV cut-off, was to use a high-energy (10 mJ), low-reprate mid-IR laser system (20 Hz), in a loose focus geometry, where laser pulses as intense as $4x10^{14}$ W/cm^2 were focused in a large-diameter capillary filled with helium gas with pressures up to 80 atm. These high pressures were crucial for observing a significant photon flux in the keV spectral region. However, the 20 Hz reprate laser could not generate a photon flux necessary for using such X-Ray beam as a spectroscopy tool. At ELI-ALPS, a 3200 nm laser driver, operating at 100 kHz, would be used to generate broadband X-Ray pulses. Our moderate pulse energy of 0.15 mJ would require a completely different frequency upconverting approach for reaching the keV photon energy range. A tight-focusing geometry, necessary to reach the intensities up to 5 $x10^{14}$ W/cm^2, would be used either in a horizontal gas-filled capillary or a vertical waveguide with a 40 microns diameter. By coupling the laser beam into a capillary with a 40 microns diameter, and keeping the pressure of He at 30-80 atm, we would be able to generate the X-Ray pulses with energies as high as 1.5 keV and increase significantly the X-Ray flux due to the high reprate of the ELI-ALPS mid-IR laser source. More recently, it was shown that isolated attosecond pulses, with a cut-off energy of 300 eV, were produced in a tight focusing geometry, with 1800 nm laser drivers and moderate gas pressures (3.8 atm), and 0.2 mJ pulse energies[327]. These results are making us optimistic that we would be able to extend the cut-off to a keV energy range with the current mid-IR laser source, and produce X-Rays with enough flux to use them as spectroscopy tools.

6.2 Time-resolved attosecond X-Ray applications

Coherent, broadband X-Ray pulses posses an extremely high temporal resolution. Unfortunately, the broadband structure prevents us from using these novel spectroscopy tools in a traditional way, e.g., as monochromatic X-Ray pulses from synchrotron and FEL sources are used in XPS experiments. At ELI-ALPS, we will develop experimental setups capable of performing time-resolved XPS experiments with a moderate time/energy resolution utilizing multilayer mirrors and grating monochromators. On the other hand, when used in a transient-absorption/reflection geometry, these broadband X-Ray sources would allow for a completely new class of experiments with unprecedented temporal and spectral resolution in the soft X-Ray region. Below, we give a couple of examples for utilizing attosecond X-Ray radiation both in gas phase and condensed matter experiments.

Gas-phase X-Ray transient absorption

The ability to control the excitation of a neutral molecule on attseocnd time scales[328] and monitor electronic respone over a broad, X-Ray energy range would allow for controlling electron dynamics with unprecedented spectral and temporal resolution. In case of ozone, for example, by doing attosecond UV-pump/X-Ray-probe transient absorption experiments, where attosecond X-Ray generation is driven by a 3200 nm laser field, we would be able to measure a rapid dissociative process of an excited O_3^* molecule by monitoring a chemical shift of the central and terminal O1s electrons. As shown in fig. 72 b), this chemical shift is 4.7 eV, and the dissociation process would be followed by measuring how the relative amplitudes of the O_C1s and O_T1s peaks change as a

[327] F. Silva et al., Nature Communications, DOI: 10.1038/ncomms7611 (2015)
[328] P. Ranitovic et al. PNAS, 111 (3), page 912-917 (2014)

function of UV-pump/X-Ray-probe delay time.

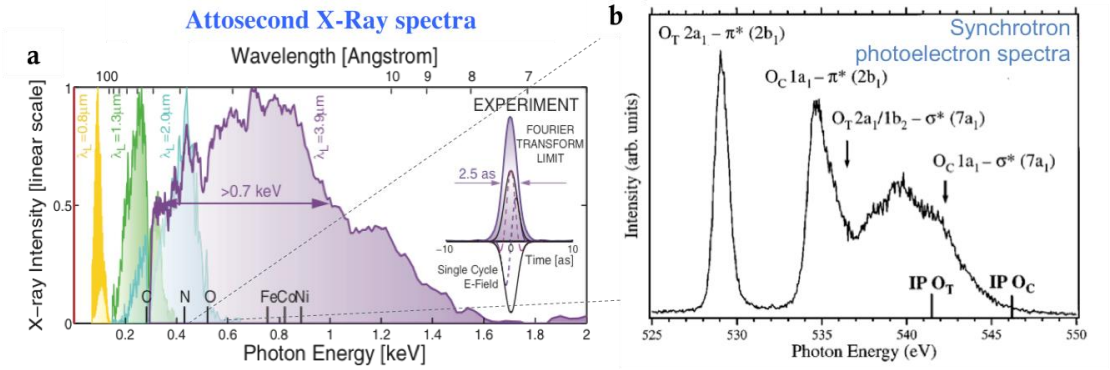


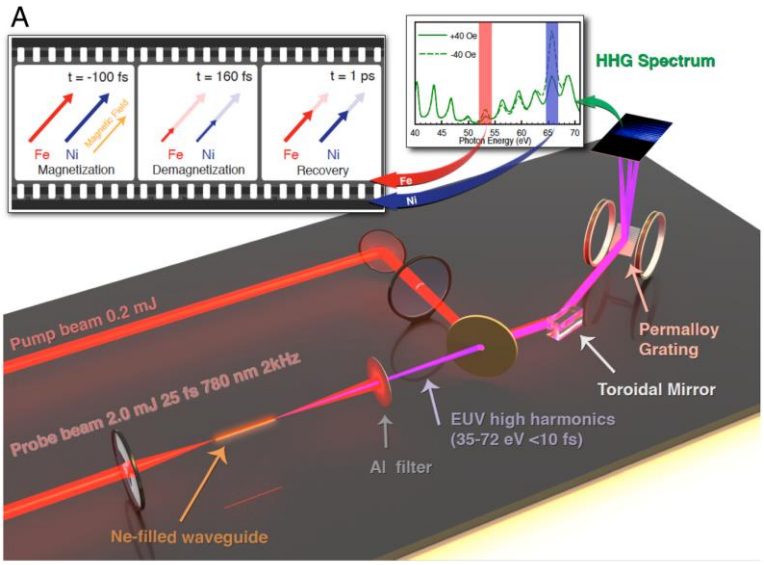
Figure 72 a) Broadband X-Ray spectra b) Synchrotron photoelectron spectra of ozone

Compared with the synchrotron experiments (Fig. 72b), where the X-Ray energy has to be scanned and photoelectron spectra measured, such a spectrum can be obtained in a couple of seconds by measuring transient absorption X-Ray spectra. Fig. 72a) shows a typical broadband spectrum obtained with different mid-IR laser drivers[3] that can be used for such experiments. Similar experiments can be done in a lower energy range as well, by measuring a chemical shift in a process of acetylene-vinylidene isomerization. As seen in Fig. 72 a), C1s binding energy is in the 300 eV range where a significantly higher photon flux could be obtained. By using the same technique, we would excite a neutral C_2H_2 (or C_2H_4) molecule by means of attosecond VUV radiation (i.e. 5th harmonic) and probe the isomerization dynamics by recording the X-Ray transient absorption spectra. The broadband X-Ray pulses centered at the C1s electron energy (~285 eV) would allow for monitoring the ultrafast process of acetylene isomerization by observing the chemical shift over a broad energy range as the molecule isomerizes from the H-C-C-H to the 2H-C-C configuration.
At ELI-ALPS these X-Ray transient absoprtion experiments are invisioned to be extended to solvents in liquid water jets, and surface chemistry experiments where we would be able to apply these novel attosecond tehcniques to, e.g., uderstand and control simple chemical reactions on metal-organic interfeces.

Condensed-phase X-Ray transient absorption

One of the missing key ingredients for understanding magnetism on microscopic level is the lack of tools that allow for simultaneously accessing ultrafast dynamics of many different - element-specific - magnetic domains. Recently, it was demonstrated that coherent XUV beams from high-harmonic sources could be used to probe ultrafast, element-specific magnetization dynamics in permalloy (FeNi)[329]. It was shown that the magnetic birefringence at the M-edge in a transition metal could be used to simultaneously follow dynamics of both Ni and Fe. Figure 73 shows a typical transient absorption/reflection setup for ultrafast magnetization experiments done with table-top XUV sources. Several striking features are obvious from those pioneering experiments. First, the use of 30 fs IR laser drivers prevents the investigation of the magnetic domain dynamics above the XUV energies of 100 eV. Second, 10 fs long XUV pulses comprise of individual harmonic combs separated by 3.2 eV, which dramatically decreases the spectral resolution and ability to access site-specific energies. And third, while 30 fs IR and 10 fs XUV pulses have a good temporal resolution,

[329] S. Mathias, PNAS 109 (13), page 4792-4797 (2009)



compared with synchrotron and to

Figure 73 Transient absorption/reflection setup for ultrafast magnetism experiments

FEL sources, these pulse durations are still not short enough to perform first attosecond magnetism experiments. With the ELI-ALPS mid-IR laser source, we would be able to dramatically improve all three of these parameters: photon cut-off energy, spectral and temporal resolution. As discussed in the previous paragraph, broadband X-Ray spectra have a superior temporal (i.e. attosecond), and spectral resolution, and would be able not just to extend the photon energy cut-off to the Fe and Ni L edges (see Fig. 72a in the 800 eV range), but also to give a complete picture of the magnetization dynamics of multiple elements with energies ranging from valence to core electrons. This example shows a full power of ultrashort and ultrabroad X-Ray pulses when used in a transient-absorption/reflection geometry in condensed phase. Similar to the magnetism experiments, at ELI-ALPS we envision using these transient absorption methods not just in magnetic, but also in strongly correlated materials.

Time-resolved X-Ray HHG spectroscopy

The availability of high intensity mid-IR laser pulses will also open new avenues to high-harmonic spectroscopy (HHS). Whereas HHS driven by 800-nm laser pulses is largely limited to species with ionization potentials above 10 eV, promising progress has already been made with the use of 1300-1800 nm sources[330,331]. Using laser pulses centered at 3200 nm opens up a door for an entirely new set of applications where HHS can be applied to electronically excited states with binding energies as low as a few eV. Building on studies of photochemical excited-state dynamics[332,333], all requirements for performing time-resolved tomography of the evolution of electronic structure in photochemical reactions will be fulfilled. Moreover, novel schemes of HHS will become feasible, such as X-ray-initiated HHG (XiHHG)[334]. We propose to implement XiHHG using soft-X-ray attosecond pulses to ionize small organic molecules from the K-shell of C, N or O (280-540 eV range) and use the synchronized mid-IR laser field to probe the ultrafast electronic and nuclear dynamics through high-harmonic spectroscopy. This method will give access to dynamics independent of the laser field, such as Auger decay and charge migration, but additionally to

[330] C. Vozzi et al., Nat. Phys. 7, 822 (2011)

[331] A. Rupenyan et al., Phys. Rev. A 87, 033409 (2013)

[332] H. J. Wörner et al., Nature 466, 604 (2010)

[333] H. J. Wörner et al., Science 334, 208 (2011)

[334] J. Leeuwenburgh et al., Phys. Rev. Lett. 111, 123002 (2013)

dynamics induced or controlled by the external field, such as laser-enabled Auger decay or laser-controlled charge migration.

6.3 Time-resolved electron diffraction and self-imaging applications

As mentioned in the introduction, in a recollision process of an electron with its parent ion, significantly higher electron energies can be reached when this process is driven by a mid-IR laser, compared with a typical 800 nm laser field[335]. Pioneering experiments using 1800-2200 mm laser pulses have recently been reported, measuring the structural dynamics of O_2^+ following strong-field ionization[336]. Higher kinetic energies correspond to shorter de-Broglie wavelengths that will offer subatomic resolution. We propose to perform time-resolved electron diffraction experiments to resolve both structural and electronic dynamics in molecular cations triggered by strong-field ionization. These experiments will be carried out in a dedicated velocity-map-imaging spectrometer designed to resolve electrons with kinetic energies up to 2 keV. We propose to study the coupled electronic and nuclear dynamics taking place in CH_4^+ following strong-field ionization. Ionization of CH_4 leaves the cation in a geometric configuration that corresponds to a triple conical intersection of CH_4^+. A very strong Jahn-Teller effect subsequently drives a coupled electronic and nuclear rearrangement that takes place on a sub- to few-femtosecond time scale. Measuring angle-resolved photoelectron spectra for several central wavelengths provided by the tunable ELI-ALPS mid-IR laser source is equivalent to tuning the excursion time of the continuum electron and is therefore expected to provide the differential scattering cross section of dynamically evolving CH_4^+ on a sub-femtosecond time scale. Similar studies will also be carried out on CD_4^+ which represents a useful reference system to discern electronic from vibrational dynamics. These studies will be extended to the investigation of ionization-induced proton transfer in molecules and clusters. This is another fundamental process that plays an important role in the understanding of chemical and biological function and could not be studied on its natural few-femtosecond time scale so far because of the lack of suitable experimental methods.

6.4 Attosecond electron beams

We propose to investigate the generation of energetic monochromatic electron bunches with a radially polarized midIR beam[337]. Focusing a TM01 transverse magnetic laser mode can produce a strong longitudinal field. At the focus, constructive superposition of the electric field will take place along the propagation axis, unlike when a TEM_{00} or TE_{01} mode is used. At high intensities, electrons positioned in the focal region can be accelerated along the propagation axis by the longitudinal electric field component, while the transverse field components (electric and magnetic) help to maintain the particles close to the beam propagation axis. The intensity of the resulting longitudinal field is dependent upon the peak power of the pulse, the laser central wavelength, and the numerical aperture (NA) of the focusing optics. It has been demonstrated the generation of 10^6 electrons per shot at 20 keV ($\Delta E/E$ of 10%) with a few cycle pulse of 0.5 mJ at 1.8 μm[338]. The longitudinal field a_z scales as λ^2 thus, illuminating with a mid IR beam at 5 μm allows to increase a_z by a factor of 8 compared to use of 1.8 μm and 40 compared to the use of 800 nm radiation. With a few cycle pulse of 10 mJ (0.2 TW), it would be possible to generate longitudinal fields as high as $a_z = 20$.

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[335] D. Ray, Phys Rev Lett., 100 (14), p 143002 (2008)

[336] C. I. Blaga et al., Nature 483, 194 (2012)

[337] C. Varin *etal.*, Appl. Phys. B 74, s83–s88 (2002), Q. Kong *etal.*, Phys. Rev. E 69, 056502 (2004).

[338] S. Payeur et al, APPLIED PHYSICS LETTERS 101, 041105 (2012)

6.5 Nano-plasmonic with midIR sources

These applications exploit the lamda-square scaling of the achievable electron energy of photoemitted and photoaccelerated electrons at plasmonic nanoparticles (novel photocathode applications, diffraction experiments etc.) and the possibility of investigating strong-field physics phenomena at unprecedentedly low laser intensities in the closest nanoscale vicinity on metal nanoparticles. It is carrier-envelope phase-stable few-cycle or single-cycle pulses with 3-8 μm central wavelength with at least 100 μJ pulse energy with up to 100 kHz repetition rate that would be particularly interesting for this purpose.

Outlook, future implications

Mid-IR lasers are one of the most promising light sources to be used for extending attosecond science from the XUV to the soft X-Ray spectral domain. In this chapter we gave several examples of how these novel sources would be used at ELI-ALPS with applications in AMO physics, physical chemistry and condensed matter physics. In the first phase of the project, we are planning to use a high-reprate, low-energy mid-IR source in a tight-focusing geometry for doing proof-of-principle experiments. In the second phase, we would increase the photon flux of the soft X-Ray beam by increasing the average power of the driving mid-IR source and adjusting the X-Ray HHG sources accordingly. By the end of the implementation phase, we are envisioning a fully developed attosecond soft X-Ray beamline to be offered to user.