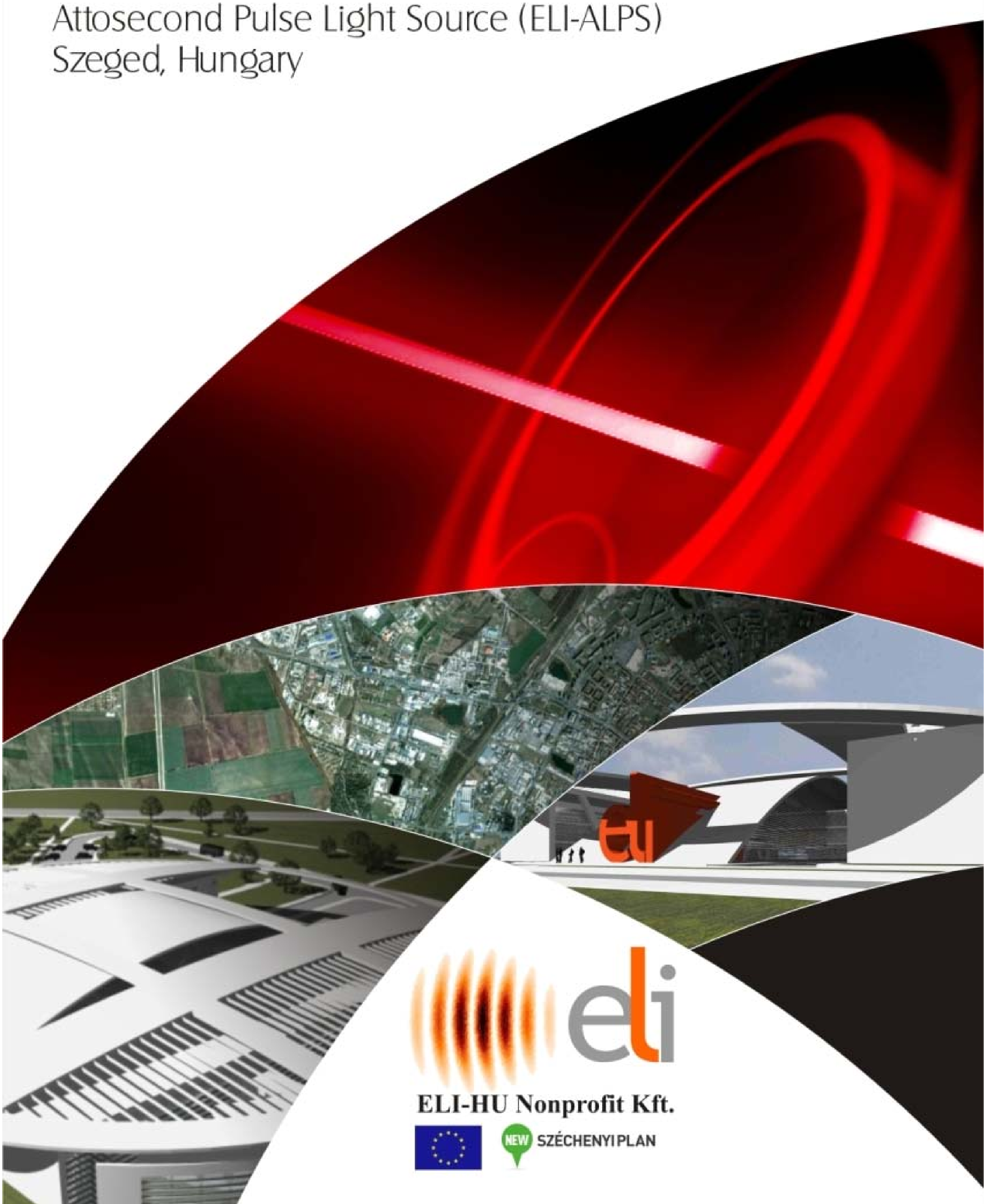
EXTREME LIGHT INFRASTRUCTURE

Attosecond Pulse Light Source (ELI-ALPS)
Szeged, Hungary



Frontiers in Ultrafast Photonics

The Attosecond Light Pulse Source (ALPS)

of the Extreme Light Infrastructure (ELI)

Szeged - Hungary

Science and Technology

Basis of the Planned Research Activity, that is

part of the Structural Funds Application

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VISION OF ELI ALPS

The frontiers of modern photonics are mainly defined by the characteristics of available photon sources. 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: the short-wavelength and high flux of third-generation synchrotron sources with the incomparable pulse duration of laser-driven harmonic sources. Thanks to this never-before-achieved combination of parameters, 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 creation of ELI-ALPS in its own right. However, ALPS will have much more to offer. Its attosecond XUV/X-ray pulses will come in synchronism with the controlled few-cycle to sub-cycle electric and magnetic 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 unequilibrium 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 ~60-page 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.

Envisioned main parameters of ELI-ALPS primary (laser) and secondary (laser-driven) sources

ELI-ALPS is aimed at defining the frontiers in ultrafast photonics. To this end, current technological limitations will be overcome by use of novel concepts. The main technological backbone of ELI-ALPS will be optical parametric chirped-pulse amplification (OPCPA) of few-cycle to sub-cycle laser pulses. Pumped by dedicated all-solid-state short-pulse (ps-scale) sources and their (low-order) harmonics, this approach will be competitive with conventional (Ti:sapphire-laser-based) femtosecond technology in terms of pumping efficiency and will dramatically outperform previous technologies in terms of average power, contrast, bandwidth, and – as a consequence – degree of control of the generated radiation.

The ELI-ALPS laser architecture will consist of three main pillars, operating at different regimes of repetition rates and peak powers: WB (wide-band): 10-100 kHz/TW scale, SYLOS (single-cycle OPCPA system): 1 kHz/ 10-TW scale, HF (high-field): 1-10 Hz/PW scale, see Table 1. The last two systems will draw on similar front ends and can be operated from a common architecture to ensure a maximum amount of flexibility, stability (duplicated front end to minimize downtime) and synchronism between them. All three pillars will deliver pulses with unique parameters: unparalleled fluxes, extreme broad bandwidths spanning up to several octaves (by use of multi-channel OPA technology) and sub-cycle control of the generated fields. This exceptional performance will give way to a set of secondary sources with incomparable characteristics, too.

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[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.

Table 1: ELI-ALPS Source Parameters (till the end of 2015)

Primary sources	Peak /average power	Repetition rate	Pulse Energy	Pulse Duration	Spectral Range	Secondary sources
ALPS-WB	**>0.1TW / ~100W**	**100kHz**	**~1mJ**	**<5fs**	**Up to 0.3-1.3µm**	**-T0 FIR/THz** 0.3-3THz, 100µm-1mm, 1.24-12.4meV **-T1 MIR** 3-30THz, 10-100µm, 12.4-124meV **-T2 NIR** 30-300THz, 1-10µm, 0.12-1.24eV **-P1 UV/XUV** 1-25PHz, 12-300nm 4-100eV **-P2 SXR** 25-100PHz, 3-12nm,100-400eV
SYLOS (*Single-cycle OPCPA System*)	**≥5TW / ≥50W**	**1kHz**	**≥50mJ**	**<9fs**	**Up to 0.5-1.3µm**	**-S1** 10-100eV, 12-120nm **-S2** 100-1000eV, 1.2-12nm **-H1** 1-10keV, 0.12-1.2nm **-H2** 10-100keV, 0.12-1.2A
ALPS-HF[2]	**PW scale / >>1W (PW scale** / ≥100W)[3]	**1Hz** (5Hz)	**>>1J** (≥20J)	**sub-10fs** (<20fs)	**Up to 0.5-1µm/ 0.7-1.3µm** (0.7-0.9µm)	

[2] Assuming 3 µm FWHM focus diameter.
[3] Technological feasibility of the HF pillar on the basis of OPCPA will be assessed in 2012. In the absence of a reliable implementation strategy, ALPS-HF will be realized on the basis of an OPCPA front and Ti:sapphire-based back end, resulting in system parameters summarized in parentheses.

Envisioned Timeline[4]

	2012	2013	2014	2015
Primary sources				
ALPS-WB				
SYLOS				
ALPS-HF				
Secondary sources - WB				
ALPS-WB-T0/1/2				
ALPS-WB-P1/2				
Secondary sources - SYLOS				
SYLOS-S1/2, -H1 atomic harmonics				
SYLOS-S1/2, -H1 surface harmonics				
SYLSO-H2				

The unique features of ELI-ALPS

i. Few-cycle to sub-cycle light waveforms from terahertz/infrared (WB-T1/T2) to petahertz/visible/ultraviolet (WB) frequencies permitting accurate sculpting of electric and magnetic fields on femto- to attosecond time scales, along with synchronized attosecond XUV probes (WB-P1/P2) at multi-kHz repetition rates;

ii. Millijoule-scale attosecond XUV (SYLOS-S1) and soft-X-ray (SYLOS-S2) pulses at kHz repetition rates;

iii. Microjoule-scale attosecond hard-X-ray pulses at kHz repetition rates (SYLOS-H1);

iv. Sub-femtosecond multi-10-keV hard-X-ray pulses (SYLOS-H2);

v. Controlled, ultra-relativistic, ultrahigh-contrast few-cycle/mono-cycle light fields;

vi. Attosecond synchronisation between (i)-(v).

[4] Light blue area: extended development time; dark blue area: min. required development time. The status of the systems at the end of 2015 is described in the corresponding laser chapter. The secondary sources include the required experimental infrastructure of the generation as well as application of them.

Synergies with ELI-BL and ELI-NP

During the ELI Preparatory Phase (ELI-PP) process three sites have been chosen to host the ultra-high intensity infrastructure(s). The multiple site structure is developed on the basis of specificity and complementarity. The main goal of the facilities is to develop and build ultra-high-power laser systems with focussed intensities and average powers reaching far beyond of what is currently available and to serve European users in a wide range of scientific, industrial and medical applications.

The three infrastructures have been decided to provide laser pulses all of world leading, but different characteristics, serving different applications: the ELI Beamline Facility (ELI-BL, Czech Republic) focuses on short pulse x-ray generation and acceleration of particles and their applications, the ELI Attosecond Facility (ELI-ALPS, Hungary) on generation and application of supershort (attosecond-range) pulses with very high repetition rates, whereas the ELI Nuclear Physics Facility (ELI-NP, Romania) will use ultra-intense optical and gamma ray pulses mainly for investigations of fundamental problems in the field of nuclear physics. Merging the individual scientific areas covered by these sites shall match the full scientific case of ELI.

The laser science and technology developments at the three sites will support each other in achieving the goals set by the individual projects, and at the same time contribute towards the technology development of generating 200 PW pulses, being the ultimate goal of the ELI project.

TECHNOLOGICAL BASIS AND DEVELOPMENTS

1 Laser infrastructure

Attosecond physics and the underlying technologies have been pushed to their limits in the recent years. Further scientific advancement calls for a breakthrough in one or other parameters of the primary sources / light sources / lasers applied in the production of these attosecond XUV and x-ray flashes or the generation process itself. This chapter concentrates on the planned primary sources and the corresponding added value that they provide compared to other conventional systems. The main quantum leap is the increase in repetition rate and associated tremendous boost in average power by about two orders of magnitude. This renders the primary and the associated secondary sources to promising and applicable tools not only in scientific research, but also in medicine and industry. Another unique feature is the much shorter duration of ultra-intense laser pulses, capable of generating a single attosecond pulse directly with significantly increased efficiency. As a consequence secondary sources with unmatched properties will be available such as much higher repetition rate, broader spectrum and correspondingly shorter duration down to the zeptosecond regime, many orders of magnitude higher x-ray yield and photon number, and larger photon energy. To this end three unique primary sources (ALPS-WB, SYLOS, ALPS-HF chapters) are designed, tailored to the applications and to the short wavelength secondary sources. As the demand on these primary sources is very high, much above the technical limitations of conventional solid-state lasers – as for example the 20-30 fs pulse duration – the optical parametric chirped pulse amplification (OPCPA) technique is utilized (State of the art chapter). As the ELI ALPS facility is aimed to put a hyperspectral radiation source as probing diagnostics or tool to produce excited states in atoms and molecules to the user's disposal, the project includes also the generation of longer wavelength as well as UV/VUV radiation (low order harmonics) and therefore some lasers serving these sources will be developed as well (Other primary sources chapter). Two challenging characteristics of state-of-the-art systems, the carrier-envelope phase (CEP) and the contrast, are discussed in the Diagnostics chapter.

1.1 State of the art

The optical parametric chirped pulse amplification (OPCPA) technique in non-collinear pump and seed geometry offer extremely broad – reaching octave spanning – bandwidth of amplification and the associated very short pulse duration down to a single optical cycle, which are not available from any laser medium and are a prerequisite of the successful operation of the ELI ALPS facility. Therefore, the basic primary sources will utilize this technology and it is discussed in more detail in this subchapter. A short description of Ti:sapphire systems will be presented later. OPCPA includes a stretching before and compression after amplification and so significantly diminishes nonlinear effects and eventual optical damage permitting operation at much higher energy levels than optical parametric amplification alone. Other important advantages[1] of OPCPA over conventional lasers are the huge single pass gain; low thermal load in the amplifier crystals; spectral tunability; scalability to high energy; support of high temporal intensity contrast by finite temporal window of amplification and reduced amplified spontaneous emission. OPCPA systems are developed (or are under development) at very high energies and corresponding highest power levels of 0.56 PW in Russia[2] as well as 10 PW[3] in United Kingdom supporting few-10-fs pulse duration using large size LBO or KDP crystals. A mixture of OPCPA and well developed chirped pulse amplification based on conventional solid state laser amplifiers, a so-called hybrid laser system[4], is also applied at these highest energy and power levels of 0.5-1.3 PW[5] in Japan, China and USA, utilizing the high gain and large bandwidth of OPCPA and the high efficiency and power scalability (i.e., the possibility to use more pump beams) of Ti:sapphire or Nd:glass as the final gain medium.

[1] A. Dubietis et al., IEEE J. Sel. Topics Quantum Electron.12, 163 (2006).
[2] V. V. Lozhkarev et al., Laser Phys. Lett. 4, 421 (2007).
[3] Y. Tang et al., Opt. Lett. 33, 2386 (2008); C. Hernandez-Gomez et al., CLF Annual Report 2007-2008.
[4] I. Jovanovic et al., Opt. Lett. 27, 1622 (2002).
[5] H. Kiriyama et al., Appl. Opt. 49, 2105 (2010); Wang et al., CLEO 2011, PDPA9; E. W. Gaul et al., Appl. Opt. 49, 1676 (2010).

Non-collinearly phase matched OPAs are producing µJ-scale few-cycle pulses down to the 3.9 fs duration[6]. Non-collinear OPCPA systems resolving these energy limitations and still supporting the huge bandwidth required for few-cycle pulses. This way multi-TW systems with 2-3 cycle duration (5.5-8 fs) became available[7]. Further decrease in pulse duration can be achieved by the application of multi-color pumping by, for example, the second and third harmonics of the Nd:YAG pump laser that provides about an octave spanning bandwidth and scalable to J energy scale[8]. It has been proven that the carrier envelope phase is preserved during OPCPA[7,9].

1.2 ALPS-WB

The wide band (WB) primary source is based on an OPCPA amplifier and partially on external spectral broadening. WB has very high repetition rate (100 kHz) – two orders of magnitude higher compared to state-of-the-art Ti:sapphire drivers in this field –, moderate laser energy (3-5 mJ) – still about a factor of ten higher than other comparable sources – and shortest pulse duration reaching the sub-optical cycle (1.5-3 fs), to push the envelope of attosecond science with popular moderate energy XUV and X-ray pulses from gas high harmonic generation and IR/visible/UV pulses in pump-probe configuration (chapter A).

The laser development is divided into two milestones corresponding to a basic design till the end of 2015 and an advanced design with longer development time:

a) basic WB OPCPA design (doable right now or with small risk R&D, till 2015):
100 kHz, >0.1 TW (<5 fs, ~1 mJ), P_{ave} ~100 W

b) advanced WB OPCPA design (need significantly more R&D, beyond 2015):
100 kHz, 2-3 TW (1.5-3 fs, 3-5 mJ), P_{ave} = 300-500 W

The basic components of the system (see also Figure 1.1.) and relevant technologies include:

- 100 kHz CEP stable front end. Ti: sapphire multipass amplifier systems are very advanced in this field and offer a solution up to about 10 kHz. As higher repetition rate is a challenge with Ti:Sa other technologies as external cavities or oscillators further amplified with fiber lasers[10] with external CEP stabilization seem to be suitable candidates.

- kW-scale diode pumped solid state ps pump laser. Various technologies hold out hopes of providing a pump laser with these properties, as Yb:YAG slab amplifier (ILT Aachen) or thin disk laser (Trumpf GmbH, developing institutes in Berlin[11] and Garching[12]). The kW power scale is within reach and all other properties as stability, duration, synchronism to a seed, and applicability are satisfactory for a small-scale high-repetition-rate OPCPA pump system as was proven in Garching (LWS-1 project[13]).

- 1-2 non-collinear OPCPA stages (ps, multi-color pump). This approach has been proven for a ps pump in various OPA / OPCPA systems as well as for a spectrum approaching an octave in an OPCPA test setup in Garching[8]. Although, damage and B-integral issues are limiting the applicability such a pump laser the aimed energy level is still feasible.

- Octave spanning stretcher and compressor. As the pump pulse duration is only about one ps the stretching and compression is less demanding than for other applications and specially designed chirped mirrors are ideal candidates for this purpose of the OPCPA part as well as the synthesizer part.

[6] A. Baltuska et al., Opt. Lett. 27, 306 (2002).
[7] S. Adachi et al., Opt. Express 16, 14341 (2008); S. Witte et al., Opt. Express 14, 8168 (2006); D. Herrmann et al., Opt. Lett. 34, 2459 (2009).
[8] D. Herrmann et al., Opt. Express 18, 18752 (2010); and later compression test experiments of the spectrum approaching an octave.
[9] C.P. Hauri et al. Opt. Lett. 29, 1369 (2004).
[10] Tavella et al., Opt. Express 18, 4689 (2010); Rotthard et al., Opt. Express 18, 12719 (2010), Hädrich et al., Opt. Lett. 36, 313 (2011).
[11] http://www.mbi-berlin.de/de/research/projects/1-02/
[12] T. Metzger et al., Opt. Lett. 34, 2123 (2009).
[13] X. Gu et al., Opt. Express 17, 62 (2009), and present developments.

However, a prism stretcher and a bulk compressor options are also suitable alternatives. Tunability of the OPCPA pulses can be provided by a spatial light modulator based shaper or matched wedge combinations.

- Partially hollow-core fiber broadening to generate UV components and a field synthesizer. Synthesizing optical fields of sub-/mono-cycle duration has been demonstrated on the sub-mJ energy scale in Garching. The technology is based on broadening in a hollow-core fiber with higher pressures to generate spectral components from UV to NIR. The compression is realized by separating the beam in three spectral regions using corresponding chirped mirrors and combining the different parts with interferometric accuracy and stability. This way a unique sub-cycle source is constructed with a waveform tunable in a certain range by tuning the channel parameters.

- Single shot (SS) CEP meter and stabilization / stabilization and temporal contrast, these properties are discussed in the Diagnostics subchapter.

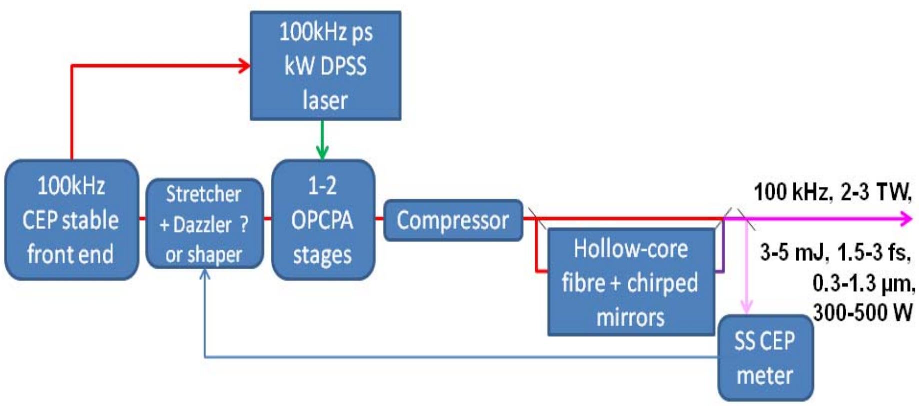

Figure 1.1

Schematics of ELI-ALPS-WB

1.3 SYLOS

The SYLOS (Single-cycle OPCPA system) system will be the main source of the ELI-ALPS facility with high enough power (100 TW) to drive secondary sources efficiently not only with atomic, but also with surface high harmonics at an unprecedented repetition rate of 1 kHz (chapter 2 Secondary sources). Furthermore, the tremendous laser energy compared to state-of-the-art systems and still quasi-single-cycle duration makes this source an ideal driver for ultra-intense, high average power, single attosecond pulses. Due to the exceptionally large XUV/X-ray energy this system opens up the route to nonlinear XUV and X-ray science (chapter B) as well as 4D imaging (chapter C) and industrial, biological and medical applications (chapter E). The two stages of the system development include:

a) basic SYLOS design (doable right now or with small risk R&D, till 2015):
1 kHz, ≥5 TW (<9 fs, ≥50 mJ), P_{ave} ≥50 W

b) advanced SYLOS design (need significantly more R&D, beyond 2015):
1 kHz, 100 TW (3-5 fs, 300-1000 mJ), P_{ave} = 300-1000 W

The basic components of the system (see also Figure 1.2) and relevant technologies include:

- ≥ 1 kHz CEP stable front end, which is available nowadays as a stand-alone system[14] as well as front end for OPCPA[15].

[14] A. L. Cavalieri et al., New J. Phys. 9, 242 (2007); Chen et al., Appl. Phys. B 99, 149 (2010).
[15] D. Herrmann et al., Opt. Lett. 34, 2459 (2009).

- Multi-kW diode pumped solid state 1-100 ps pump laser. The thin disk laser technology seems to be a suitable candidate as discussed by the pump sources for ALPS-WB. It holds the promise to deliver even multi-kW power on a longer term while having a tunable pulse duration in the required 1-100 ps range.

- 3-6 non-collinear OPCPA stages (1-100 ps multi-color pump). Light Wave Synthesizer 20, a pilot project at MPQ, delivered a lot of experience about 2-stage 8-fs OPCPA technology[15]. A system upgrade is in the final stadium of development with 4 stages and two-color pump to reach 5 fs duration with ~100 mJ energy. In the near future a further upgrade to 6 stages and 500 mJ, i.e. 100 TW is scheduled. A different OPCPA development project , the Petawatt Field Synthesizer, is based on 1 ps pump laser technology and provided a lot experience in this field as well[16]. A few-ps pump laser provides a gain bandwidth between 700-1300 nm, while two-color pumping even with multi-10-ps pulses supports a range about 580-1020 nm.

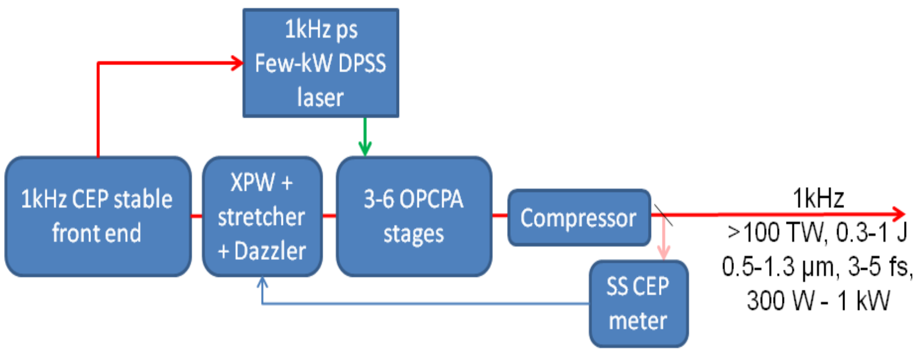

Figure 1.2

Schematics of SYLOS

- High transmission octave spanning stretcher and compressor. Depending on the pump pulse duration large size chirped mirrors with or without an extra single or mosaic bulk compressors comprise a possible solution. Spectral and temporal tunability is reliably provided by a Dazzler.

- By providing a two-beam option (for example 2x 500 mJ) a versatile tool with reduced efforts can be constructed for preparation of extreme states of matter and an intense probe beam in the same time.

- SS CEP meter / stabilization and temporal contrast, these properties are discussed in the Diagnostics subchapter.

1.4 ALPS-HF

The ELI-ALPS facility needs a HF arm reaching multi-PW peak power, even at the seemingly lower rep-rate of 1-10 Hz -in comparison to the kHz range of the earlier stages, because it complements the unique diagnostic capabilities of the facility as well as provides a mean of producing extreme states of matter. It is the necessary laser driver for generating interesting new relativistic laser-plasma interaction phenomena occurring only at those extremely high intensities (> 10^{20} W/cm^2) reachable via focusing the ALPS-HF beams –see the science sections discussing plasmas (chapter D) and inner-shell, core level atomic physics (chapter B). ALPS-HF will offer two novel features as compared to other high-power laser installations. First, the ultrahigh intensities and field strengths (both electric and magnetic), along with the associated pressure and ponderomotive potential, will be "switched on" at a never-before-demonstrated rate, from zero to maximum, thanks to the exceptional contrast offered by short-pulse-pumped OPCPA technology. Second, the ultrahigh fields from ALPS-HF will come in perfect synchronism to attosecond x-ray probes, opening the route to attosecond real-time observation of phenomena induced by ultrastrong laser fields. There are two approaches to reach the above-PW power range with the ALPS-HF system. The first option supporting few-cycle pulses at PW level, in harmony with the other

[16] Zs. Major et al., The Review of Laser Engineering 37, 431 (2009).

primary sources, is a fully OPCPA-based version. In contrast, the alternative is an OPCPA / Ti:Sa-based hybrid option, which relies on well understood / developed / commercialized Ti:Sa amplifier stages.

1.4.1 OPCPA option

The design parameters of the HF OPCPA system dictated by the extraordinary applications and limited by the state of the art of present laser technology (this system needs the demonstrated SYLOS OPCPA parameters at lower repetition rates therefore, its development starts later):

a) basic HF OPCPA design (doable right now or with small risk R&D, till 2015):
1 Hz, PW-scale (sub-10 fs, >>1 J), P_{ave} >>1 W

b) advanced HF OPCPA design (need significantly more R&D, beyond 2015):
1-10 Hz, 1-3 PW (3-5 fs, 3-10 J), P_{ave} = 10-100 W

The conceptual design of a PW OPCPA system (similar setup as by SYLOS) contains the following components:

- ≥1 kHz CEP stable front end, which is available nowadays as mentioned at the SYLOS system. The advantage of the 1 kHz repetition rate is that a synchronized probe pulse can be generated to the PW arm and its input state without interaction can be characterized in a multi-shot manner much faster than the 1-10 Hz laser repetition rate.

- 40-200 J diode pumped solid state few-(10) ps pump laser. However, the construction of such a pump laser is a challenge and significant R&D is required, flash lamp pumped few-10-ps systems are feasible at the 20-40 J levels nowadays.

- 6-8 non-collinear few(-10)-ps multi-color pumped OPCPA stages. As discussed by the SYLOS amplifier the Light Wave Synthesizer 20, a pilot project at MPQ, provided a lot of experience about the OPCPA technology up to the few-100-mJ level. Careful scaling and optimization of the amplifier crystal material –BBO, LBO, KDP, DKDP– is expected to support few-J energies. A shorter pump duration supports the gain bandwidth of 0.7-1.3 µm and a longer duration with two-color pumping delivers a range about 0.5-1 µm.

- High transmission octave spanning stretcher and compressor. The main challenge compared to the previous systems is the large power in the compressor. A grating based unit containing metal-dielectric gratings with improved efficiency or mosaic bulk compressor are possible alternatives. Spectral and temporal tunability is reliably provided by a Dazzler.

- Single shot CEP meter / stabilization and temporal contrast, these properties are discussed in the Diagnostics subchapter.

1.4.2 Alternative option for a PW-scale OPCPA / Ti:Sa hybrid system

Ti:sapphire (Ti:Sa-based) final amplifiers present an alternative technique for the ALPS-HF arm in comparison to the overall OPCPA system. Such a system is capable to boost the down-sampled (from 1 kHz to 1-10 Hz) series of the medium-level OPCPA pulses (~100 mJ, kHz reprate - same as SYLOS final output) to a compressed series of ≥20 J, 20 fs pulses to reach PW-scale at 1-10 Hz repetition rate. The conceptual design of the hybrid Ti:Sa arm presented here is based on recent advances in Ti:Sa laser technology and lessons learned at PW laser systems under construction world-wide (e.g. BELLA laser in Berkeley, USA; Apollon laser in Paris, France; Vulcan/Gemini lasers at RAL, UK).

In order to study these new interactions and relativistic laser-plasma physics phenomena, the driving hybrid laser should provide the following unique set of parameters –taken also into account the constructability of such lasers:

a) basic HF Ti:Sa design (doable right now or with small risk R&D, till 2015):
5 Hz, PW-scale (<20 fs, ≥20 J), P_{ave} = ≥100 W

b) advanced HF Ti:Sa design (need significantly more R&D, beyond 2015):
10 Hz, >2 PW (<20 fs, >40 J), P_{ave} = >400 W

In the following we will discuss the details of the Ti:Sa section of a hybrid OPCPA/Ti:Sa PW-class laser, using the goal parameters listed in the basic version (a).

How to reach the PW-scale goal parameters – conceptual amplifier design

The Ti:Sa section of the hybrid OPCPA/Ti:Sa laser system will amplify the medium-level OPCPA output pulses from 100 mJ to 40-80 J, and eventually compresses to 25-50 J, 20 fs duration pulses to reach 1-2 PW at 5-10 Hz. An additional stretcher after the ps OPCPA output will be needed to stretch the pulses to ~ 0.5-1.0 ns pulse duration in order to avoid optical damage in subsequent stages and optical components. This stretcher might reduce the maximum applicable OPCPA input energy, which will be compensated by the Ti:Sa amplifiers. The stretching and amplification estimation is based on widely accepted intensity (W/cm^2) and fluence (J/cm^2) damage threshold values, gain and saturation properties of typical Ti:Sa amplifier crystals, and also takes into account at least 1.5x safety factors relative to the theoretically achievable efficiency numbers for pumping and compression. After stretching two multi-pass amplifiers could boost the energy to the required value of about 40-80 J with a beam/crystal diameter of 100/150 mm in the second stage. The compression back to 20 fs is feasible with a conventional grating compressor supporting 200 nm bandwidth and a transmission of about 65%. This concept represents one of several –though highly realizable with relatively low-risk– potential configurations of a Ti:Sa amplifier system reaching ~1-2 PW peak power. Fine-tuning of the listed parameters should be performed during the final optimization and design, also taking into account other considerations, such as pumping architecture, CPA dispersion management, laser room and beam delivery constraints. The repetition rate is primarily depends on the available pump lasers; 5-10 Hz is realistic.

1.5 Other primary sources

The ELI ALPS facility will offer beyond the previous three systems a variety of smaller drivers to provide hyperspectral radiation for various investigations. Direct energetic IR sources between 1 and 10 μm are described in the IR OPCPA subchapter. A different spectral range, the THz generation, requires as an optimal driver more or less conventional ~J scale (sub-)ps systems. This will be available even at 1 kHz repetition rate as the pump systems for SYLOS so they are not discussed in detail here. Various studies and applications call for a few-fs UV/VUV source that is described in the Advanced sources in the UV/VUV subchapter.

1.5.1 IR OPCPA

The availability of energetic and well-controlled ultrashort pulses has opened up many new avenues of science, which is nearly exclusively based on current technology for the near-infrared spectral range below 1000 nm. Ultrafast technology at wavelengths longer than one micron, which are well suitable for example in medical applications (Chapter E), is however lacking seriously behind due to unavailability of suitable gain media, difficult to operate, or simply not available detectors, and underdeveloped optics and coatings; this lack of technology rapidly increases from NIR (near infrared: 0.9 – 1.7 μm) to SWIR (short-wavelength infrared: 1.7 – 3.0 μm) and to MIR (mid infrared: 3 – 10 μm). The most promising route to generate ultrashort pulses in the SWIR to MIR is nearly exclusively via three-wave-mixing in nonlinear crystals and specifically parametric amplification and difference frequency generation (DFG).

State of the art

Probably the most established method to access ultrashort IR pulses is via non-collinear OPA of some white light continuum or frequency shifted output, from Ti:sapphire or Yb-based fiber CPA systems[17]. For the broad spectra of hollow fiber broadened Ti:sapphire lasers DFG can also be used[18], followed by amplification in an OPA using the Ti:sapphire system as a pump. Different implementations of these various approaches have generated few-cycle pulses at 1.2 – 3 μm[15,19] and recently this has been extended to longer wavelengths, delivering 25 fs duration pulses at ~ 3 μm, with pulse energies of 2 μJ[20]. The latter system amplifies a white-light continuum with a Ti:sapphire pump laser to generate an amplified signal at 1.3 μm. In a second stage OPA this signal is amplified further, and the idler from the interaction is extracted, which has 2 μJ infrared energy at 3 μm. CEP stability of this system has yet to be proven, and scaling to higher energies is limited by the un-chirped nature of the OPA interaction. Mixing of amplified ultrashort pulses from a Ti:sapphire laser with longer pulses at around 1 μm wavelength has been shown to produce ultrashort pulses in the mid-IR[21], but is limited to roughly the duration of the driving laser pulse, and suffers from low efficiency. This technique has been a workhorse of ultrafast mid-IR spectroscopy, but is unlikely to be scalable to higher energy or few-cycle pulse durations, and does not provide CEP stable pulses.

An alternative approach to frequency shifting of NIR laser systems is direct amplification of ultrashort IR pulses using OPCPA, which involves the amplification of broad-bandwidth chirped seed pulses using a narrowband, typically pico-to-nanosecond pump laser. This approach allows amplification across a huge range of central wavelengths in the NIR to MIR with ultra-broad gain bandwidth that makes direct amplification of few-cycle pulses possible; first OPCPA systems have been reported in the SWIR (at 2.1 μm) and MIR (at 3.2 μm)[22].

Few-cycle CEP-stable 100 kHz MIR OPCPA

The proposed source is relying on a demonstrated[23] direct OPCPA concept which is based on a self-CEP stable seed through difference frequency generation (DFG) from a fiber-based master oscillator power amplifier (MOPA) frontend, pumping with high average power picosecond-duration pulses, and subsequent OPCPA. This strategy works since a demonstrated source[23] routinely delivers extremely stable (0.7% rms over 30 min), high repetition rate pulses (100kHz), with stable CEP, and few-cycle-duration (3-6 cycles). The OPCPA system is compact, reliable, and easy to operate and therefore a viable solution that can fulfill the key criteria needed across the wide range of applications within a user facility such as ELI-ALPS.

Without broadband oscillators operating in the IR, the seed pulse for our system must be generated from a shorter wavelength source and a nonlinear process. This in fact is very advantageous for an ultrashort long wavelength system, as it allows us to use a combination of standard, well-developed oscillator technology, and also to passively stabilize the CEP via DFG. In the experimental realization (Figure 1.3), the ultra broadband MIR seed for the OPCPA system is derived from a two-color Erbium fiber laser system, which delivers amplified, and phase-coherent ultrashort pulses at 1050 nm and 1550 nm. DFG between the arms producing an ultra-wideband seed spectrum[24] from 3000 to 4000 nm. Timing jitter between the MOPA arms was measured to be negligible with less than 21 as over 200 hours, corresponding to a CEP drift of less than 90 mrad over this time, without locking electronics or feedback loops[25]. The seed pulse is sent through a bulk stretcher to roughly match the pump pulse duration. The current implementation employs an electronically slaved (< 350 fs rms over 6 hours) MOPA pump system from Lumera Laser GmbH, able to deliver 40 W of average power (8 ps, 100 kHz, 1064 nm). Compression is achieved with a simple grating based setup. Previously reported results from three OPCPA stages[23] led to a record 6-cycle (67 fs) pulse, centered at 3.2 μm, with 3.8 μJ pulse energy at 100

[17] T. Wilhelm et al., Opt. Lett. 22, 1494 (1997); C. Schriever et al., Opt. Lett. 33, 192 (2008).
[18] C. Vozzi et al., Opt. Expr. 14, 10109 (2006).
[19] G. Cirmi et al., JOSA B 25, B62 (2008); C. Zhang et al., Opt. Lett. 34, 2730 (2009); G. Krauss et al., Nature Phot. 4, 33 (2010).
[20] D. Brida et al., Opt. Lett. 33, 2901 (2008).
[21] F. Rotermund et al., JOSA B 16, 1539 (1999).
[22] T. Fuji et al., Opt. Lett. 31, 1103 (2006); O. Chalus et al., Opt. Expr. 17, 3587 (2009).
[23] O. Chalus et al., Opt. Expr. 16, 21297 (2008); O. Chalus et al., Opt. Lett, 35, 3204 (2010).
[24] C. Erny et al., Opt. Lett. 32, 1138 (2007).
[25] F. Adler et al., Opt. Lett. 32, 3504 (2007).

kHz. Measurements also exhibited an unsurpassed stability of 0.7% rms over 1.8×10^8 pulses (30 min) and CEP stability to better than 100 mrad over 11 sec; proper CEP characterization is topic of current research.

Extended goal and ELI-ALPS target specification

ELI-ALPS targets within its "T2 specification" the following parameters: center wavelength of 1-10µm, pulse energy >100µJ at 10-100 kHz repetition rate. Based on the fact that 25 µJ pulse energy has been already demonstrated (unpublished) we are extremely confident that the 100 µJ pulse energy level can be reached within the following months and without any change to the existing source.

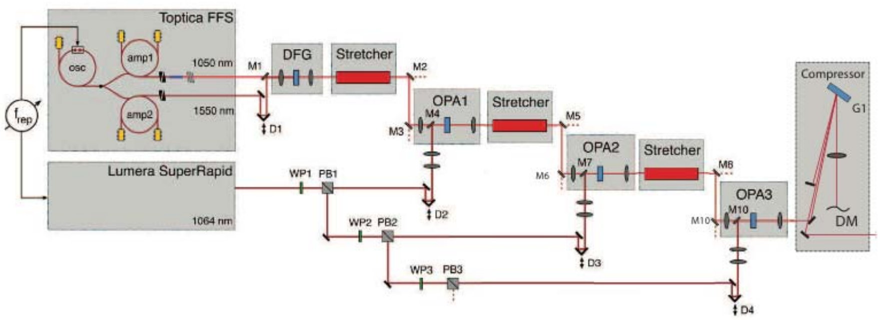

Fig. 1.3

MIR OPCPA source layout

Energy scaling of the OPCPA is feasible and fully supported by 3D simulations. At a high repetition rate of 100 kHz, 100 W (1 mJ) pump sources are already operating and scaling to the 10-mJ-level (1 kW) should be relatively straightforward with slab or thin disk pump laser technology. We foresee MIR pulse energies in the millijoule level and beyond within the timeframe of ELI-ALPS without measurable or visible parametric super-fluorescence as in the previous demonstrations. The possibility of upgrading the source to higher energy and lower repetition rate is there, simply through the addition of extra amplifier stages and/or exchange of pump lasers; this is an important advantage for ELI-ALPS meaning that as pump laser technology evolves, straightforward upgrades will be possible.

Wavelength scaling from MIR to SWIR is feasible but requires frequency doubling of the fiber MOPA output to shorter wavelength. Scaling to longer wavelengths, from the demonstrated 3.2 µm to 10 µm is possible without even changing the OPCPA crystals; only change of optics, or using metal optics is required.

1.5.2 Advanced sources in the UV/VUV

Atoms molecules and condensed matter after absorbing ultraviolet radiation are electronically excited. Whereas important molecular processes initiated by electronic excitation have been extensively studied in the past years the limited capabilities of existing sources in terms of bandwidth –to enable the excitation of electronic manifolds of states rather than isolated states– as well as duration –to permit coherent excitation– hinders broad range of applications. Recent progress in generating few-cycle pulses in the deep[26] and vacuum ultraviolet with the potential of extension to the attosecond domain[27] comprises the basis of what is proposed here as an important addition to the ELI sources. We propose to generate deep (200-400 nm) and vacuum ultraviolet (100-200 nm) pulses via the direct upconversion of few-cycle pulses (up to several mJ at 800 nm) loosely focused into a pressurized gas (1 - 10 bar) cell filled with Ne. We anticipate conversion efficiencies on the order of 2% for the deep ultraviolet and on the order of 0.5 % for the vacuum ultraviolet. The efficiency of the conversion benefits from the broad bandwidth of the few-cycle pulse which enables a sequence of nonlinear cascades other than direct harmonic generation. The pulses can be generated in vacuum and do not require additional compression and are inherently synchronized with the few-cycle pulse, a part of which can be used to generate XUV attosecond pulses (see chapter 2) for pump-probe experiments. For the cases in which narrower bandwidth

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[26] U. Graf et al., Opt. Exp. 16, 18956 (2008); F. Reiter et al., Opt. Lett. 35, 2242 (2010).
[27] F. Reiter et al., Phys. Rev. Lett. 105, 243902 (2010).

but more powerful UV excitation is needed we propose to spectrally broaden optically synchronized KrF UV pulses[28] via hollow-core fiber/chirped mirror compressors[29].

1.6 Diagnostics

This chapter deals with the characterization and control of two properties of few-cycle ultra-high intensity lasers: the carrier-envelope phase (CEP) and the temporal contrast. As these features bear a challenge even in less demanding systems they have a high priority in system design and construction from the beginning. Temporal characterization of the laser pulses is also important, but it is more routine[1] nowadays.

1.6.1 CEP measurement and stabilization

The characterization and control of CEP of an ultrashort laser pulse is fundamental for all extreme non-linear phenomena that will be accessible according to the parameters envisioned for the three primary sources (ALPS-WB, SYLOS and ALPS-HF) planned for ELI-Hungary. In particular the measurement and control of the CEP is a prerequisite for the reliable generation of isolated attosecond pulses both from gases and from solid surfaces.

The unique characteristics of the ALPS driving laser sources in terms of repetition rate (100 kHz for ALPS-WB) and of peak intensities and pulse durations (>100 TW in 3-5 fs for SYLOS and 1-3 PW in 3-5 fs for ALPS-HF) poses severe challenges for the control of CEP and calls for the development and implementation of new stabilization. In particular the ideal feedback loop should operate on a single shot basis and with a bandwidth approaching the repetition rate of the driving laser source. For the primary sources of ALPS, we propose to combine cutting-edge technologies in order to implement a feedback loop operating on a single shot basis up to few tens of kHz, matching the high repetition rate of ALPS-WB. This scheme could be implemented also on SYLOS and ALPS-HF for, either a more precise control of fast CEP fluctuations, or to perform CEP-sensitive experiments without the need to stabilize the driving pulse train. The envisioned feedback loop will be based on the combination of a fast measurement technique (a) and a fast control device (b) to correct for CEP fluctuations.

a) In situ determination of the CEP requires an extreme nonlinear optics phenomena, as the outcome of these processes depends on the precise electric field form. The absorption of a number of photons by an atom or a molecule exceeding the ionization potential, depends on the precise electric field form and is usually indicated as Above-Threshold Ionization (ATI). For few-cycle pulses the number and energy of electrons emitted in opposite directions depends on the CEP and these information can be used to characterize the CEP in the interaction region[30]. It was demonstrated that this technique can work on a single shot basis up to several kHz repetition rates, assigning to each laser pulse its corresponding CEP[31]. Characterization of the CEP drift is based on a more conventional nonlinear process, as realized in a nonlinear interferometer (f-2f interferometer) where the combination of second and third order nonlinearities, allows one to retrieve the drift of the CEP. Using a fast photodiode and/or fast photomultiplier to acquire different regions of the spatially resolved interference pattern, the CEP drift can be characterized on a single shot basis at kHz repetition rates[32], with the prospective to extend this approach up to few hundreds of MHz. Phase stabilization of ALPS large scale laser systems will require single shot measurement of the CEP or its drift, but most primarily the laser system should exhibit a high mechanical and optical stability, providing that the shot-to-shot phase change will exhibit a tendency and can be compensated by a fast feedback loop. As so far only table-top lasers have been phase-stabilized the scalability of stabilization to several orders higher peak powers (ALPS-HF) mainly for stability reasons is foreseen as a great technical challenge. However CEP sensitive experiments can be performed without the need for stabilization, simply by measuring the CEP of each laser shot and associating the experimental outcome with the corresponding CEP value, on single shot basis. This approach currently employed on high repetition rate low intensity few-cycle systems reveal itself an alternative way and can facilitate phase sensitive experiments at ELI-ALPS before stabilized pulses will be available.

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[28] S. Szatmári et al., Landolt-Börnstein New Series VIII/1B1, 215 (2007).
[29] T. Nagy et al., Opt. Lett. 34, 2300 (2009).
[30] G. G. Paulus et al., Nature 414, 182 (2001).
[31] T. Wittmann et al., Nature Phys. 5, 357 (2009).
[32] S. Koke et al., Opt. Lett. 33, 2545 (2008).

b) Fast control of the CEP requires the implementation of a control device that can be updated at kHz or higher repetition rate. Feedback loops based on moving parts are usually limited to few tens of Hz because of the intrinsic limitation introduced by the mass of the moving components. An acousto-optic programmable dispersive filter (DAZZLER) has already been used at low repetition rates to control the CEP of amplified laser systems and represents a promising solution for the realization of a feedback loop at higher repetition rates. Such a filter is already present in the ALPS primary sources for compensating the high-order spectral phase terms introduced in the amplification process and dispersion management. The acousto-optic modulator acts as a phase filter by diffracting the optical wave by means of an acoustic wave sent into the crystal. The CEP phase of the acoustic wave ϕ_{ac} is linked to the CEP of incoming ϕ_{in} and outgoing ϕ_{out} optical pulses by the relation[33]: $\phi_{out} - \phi_{in} - \phi_{ac} = \pi/2$. It is clear that by controlling the CEP of the acoustic wave, it is possible to stabilize the CEP of the outgoing pulse. It is important to observe that the DAZZLER itself introduces a CEP noise due to the time jitter between the acoustic wave and the reference clock that drives the acoustic wave generator modulator. However recent development in the synchronization of the driving electronic with an external signal have reduced such jitter to about 25 mrad that is well below the value of 100 mrad that represents the state of the art for CEP stabilization reported in the literature. The DAZZLER can be used up to repetition rate of about 30 kHz.

The combination of a fast measurement (Stereo-ATI and/or f-2f interferometer) and a DAZZLER will allow a superior control of the CEP stability of the ALPS sources. Only the ALPS-WB system might need a different CEP control device based for example on the novel an acousto-optic frequency shifter technique[34].

1.6.2 Contrast

Relativistic-intensity laser interaction with solid-state matter needs clean and well controlled experimental situations. A crucial condition is a confined, steep, high-density plasma at the arrival of the super-intense laser pulse requiring a clean, prepulse- and pedestal-free temporal structure, characterized by the contrast – the main pulse intensity divided by the light intensity at a given time instant eventually much before or after the laser pulse. This nontrivial feature of ultrahigh-intensity laser systems is dominantly influences the interaction prohibiting the generation / investigation of numerous phenomena as surface high harmonic generation, which is one of the most important secondary source – as discussed in Chapter 2. High-dynamic-range third-order correlators[35] are popular and widespread devices to determine the laser pulse contrast in a multi-shot mode. However, OPA correlators offer a reliable characterization approach, too. Depending on various factors as laser intensity, target material, laser pulse preceding temporal structure, etc., the plasma threshold of about 10^{10} W/cm^2 requires a contrast from 8 to 14 orders of magnitude on the ps timescale and even better on the ns scale. Although, conventional Ti:sapphire lasers –correspondingly the systems intended to use as front end in the ELI ALPS primary sources as well– provide without extra measures only a contrast between 10^6 and 10^8. Therefore, various contrast improvement techniques were developed to reach the desired contrast leading to an experimental situation devoid of preplasma[35]. These techniques can be arranged into three categories:

- as internal active contrast improvement (cross-polarized wave generation (XPW), saturable absorber, unbalanced SHG, self-diffraction)
- internal passive contrast improvement (short pulse OPCPA, OP(CP)A preamplifier for idler generation, unsaturated OP(CP)A)
- external contrast improvement (frequency doubling, single/double plasma mirror)

The first category denotes strategies, which typically need pulse compression in the laser system after the first amplifiers, an active nonlinear cleaning stage and a stretching again, demanding a double CPA architecture. The

[33] N. Forget et al., Opt. Lett. 34, 3647 (2009).
[34] S. Koke et al., Nature Phot. 4, 462 (2010).
[35] L. Veisz, "Contrast Improvement of Relativistic Few-Cycle Light Pulses," in Coherence and Ultrashort Pulse Laser Emission, ISBN 978-953-307-242-5, ed. F. J. Duarte (2010), http://www.intechopen.com/articles/show/title/contrast-improvement-of-relativistic-few-cycle-light-pulses

listed methods have a significant improvement factor (10^3-10^5), but reduced efficiency (~10%) that is typically compensated for in the last laser amplifiers. The internal passive strategies are special amplification techniques leading to a significant gain and contrast increase (gain dependent 10^5 is feasible), but the cost is the implementation of the different type of amplifier. The external strategies have high efficiency (35-80 %) and reduced improvement (10^2-10^4) at the end of the laser leading inevitable to a loss in final system energy. A detailed experimental study of the relevant techniques at MPQ involving a high quality Ti:Sa front end with 10^8 contrast plus an XPW stage for seed improvement with the 80 ps OPCPA pump laser of LWS-20 and a plasma mirror provided a contrast of 10^{19}, 10^{14} and 10^9, when measured 50, 25, and 1ps before the 8-fs main pulse. This study proved that the relevant techniques are compatible with few-cycle pulse duration and can fulfill even the highest expectations. Future investigations of novel spatial and temporal cleaning techniques[36] hold out a promise of even more effective filters. Based on this short and general overview of contrast measurement and correction approaches the following solutions are recommended for the ELI-ALPS systems:

ALPS-WB is mainly devoted to high harmonic generation in atomic medium, which is fairly insensitive to the contrast and normal system parameters deliver a suitable temporal structure that a dominant part of the laser energy is contained within the few-fs pulse envelope. As the pump source is a laser system with ps duration this primary source will naturally deliver high-contrast pulses suitable even for surface high harmonic generation surpassing the requirements.

SYLOS will deliver >100 TW pulses focusable to >10^{20} W/cm^2 and therefore a contrast of 10 orders of magnitude or better is required for surface high harmonic generation or Thomson scattering. Although, this contrast is a challenge with a single-cycle pulse duration, the application of XPW after a Ti:Sa front end and a short pump pulse OPCPA even with tens of ps duration can provide the required value if the system is developed carefully.

ALPS-HF has the highest power in the >1-PW range and correspondingly the highest focused intensity in principle on the order of 10^{23} W/cm^2. The high demand on the contrast of about 14 orders of magnitude might necessitate even a plasma mirror if tens of ps long pump laser is utilized as parametric super-fluorescence is expected to generate a worse preceding pedestal. The plasma mirror has spatial cleaning effects allowing a better focusing of the beam and increases the steepness of the rising edge, which is relevant in some cases as nanometer thick foil targets. Other possible solution is a few-ps pump laser up to the mJ energy scale and stretcher and few-10-ps laser pumped final amplifiers. If the alternative hybrid system is realized the short pulse OPCPA part of the system provides the same excellent contrast as the full OPCPA option.

At the end of the laser chapter the final goals of the primary and their secondary sources are summarized in Table 2.

[36] based on for example a plasma mirror with a conjugate beam block pair, see: S. Szatmári et al., Opt. Commun. 134, 199-204 (1997).

Table 2: Final ELI-ALPS Source Parameters (beyond 2015)

Primary sources	Peak /average power	Repetition rate	Pulse Energy	Pulse Duration	Spectral Range	Secondary sources
ALPS-WB	**2-3TW / 300-500W**	**100kHz**	**3-5mJ**	**1.5-3fs**	**0.3-1.3µm**	**-T0 FIR/THz** 0.3-3THz, 100µm-1mm, 1.24-12.4meV / 0.1-1 mJ, 10-30 MV/cm **-T1 MIR** 3-30THz, 10-100µm, 12.4-124meV / 10-100µJ, 3-300 MV/cm **-T2 NIR** 30-300THz, 1-10µm, 0.12-1.24eV / >100µJ, 0.3-30 GV/cm; as an alternative an IR OPCPA system can provide similar parameters **-P1 UV/XUV** 1-25PHz, 12-300nm 4-100eV / 100-10nJ **-P2 SXR** 25-100PHz, 3-12nm,100-400eV / >1nJ
SYLOS (*Single-cycle OPCPA System*)	**100TW / 300-1000W**	**1kHz**	**0.3-1J**	**3-5fs**	**0.5-1.3µm**	**-S1** 10-100eV, 12-120nm / 100-1µJ **-S2** 100-1000eV, 1.2-12nm / 1-0.1µJ **-H1** 1-10keV, 0.12-1.2nm / 100-1nJ **-H2** 10-100keV, 0.12-1.2A / >1nJ
ALPS-HF[1]	**1-3PW / 10-100W** (>2PW / >400W)[2]	**1-10Hz** (10Hz)	**3-10J** (>40J)	**3-5fs** (<20fs)	**0.5-1µm/ 0.7-1.3µm** (0.7-0.9µm)	**Controlled electric field:** (3-5)x10^{14} V/m **Normalized vector potential:** 60-110/80-140 **Peak intensity:** (1-3)x10^{22} W/cm^2 **Ponderomotive potential:** (0.6-2)x10^{22} / (1-3)x10^{22} W/cm^2 × µm^2

[1] Assuming 3 µm FWHM focus diameter.
[2] Technological feasibility of the HF pillar on the basis of OPCPA will be assessed in 2012. In the absence of a reliable implementation strategy, ALPS-HF will be realized on the basis of an OPCPA front and Ti:sapphire-based back end, resulting in system parameters summarized in parentheses.

2 Secondary sources

ALPS-WB will drive – via low-order (perturbative) nonlinear interactions – infrared sources, ALPS-WB-T0, ALPS-WB-T1 and ALPS-WB-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-WB, 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-WB-P1 and ALPS-WB-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-WB 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 and ALPS-SYLOS-S2, and ALPS' incomparable attosecond hard-X-ray source: 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 H1-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 for example the Yb:YAG pump source of ALPS-SYLOS off few-femtosecond relativistic electron bunches accelerated by the >100TW 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 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.

2.1 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, the attosecond light pulses may be used to produce electron wave packet 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[1] or high-order harmonic generation in bulk crystals[2].

[1] J. Seres et al., Nat. Phys. 6, 455 (2010)
[2] S. Ghimire et al., Nat. Phys. 7, 138 (2010)

2.1.1 Attosecond pulse production via gas harmonics generation

Principle of operation

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[3]. High-order harmonic generation (HHG) in gas is a widely used technique for the production of coherent extreme ultraviolet (XUV) radiation[4]. 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[5]. 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 freed electron may 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.

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[6,7] or in hollow-core fibers[8,9]. 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 fulfill 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 driving pulses, with an energy of 14 mJ, focused into a long cell (xenon) by a 5-m-focal length lens[10].

Source development

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[11]. All-optical schemes have been also developed, which employ weak counter-propagating pulses[12]. Another QPM approach is based on the use of a multi gas cell target[13]: it has been demonstrated that it is possible to increase the conversion efficiency by using n consecutive gas cells

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[3] A. L'Huillier and Ph. Balcou, Phys Rev Lett 70, 774 (1993).
[4] 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.
[5] K. J. Schafer and K. C. Kulander, Phys. Rev. Lett. 70, 1599 (1993), P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).
[6] J.-F. Hergott et al., Phys. Rev. A 66, 021801(R) (2002).
[7] E. Takahashi et al., Phys. Rev. A 66, 021802 (2002).
[8] A. Rundquist et al., Science 280, 1412 (1998).
[9] R. Bartels et al., Nature 406, 164 (2000).
[10] E. J. Takahashi et al., Opt. Lett. 27, 1920 (2002).
[11] E.A. Gibson et al., Science 302, 95 (2003).
[12] T. Popmintchev et al., Nature Phot. 4, 822 (2010).
[13] J. Seres et al., Nature Phys. 3, 878 (2007).

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 n^2. 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 two main classes of generation schemes. The first method, called amplitude gating, is based on spectral selection of the cutoff region of the HHG radiation[14]. 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. 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[15]. (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[16,17]; 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 pulses by using sub-30-fs driving pulses[18]. By using the interferometric polarization gating technique[19] 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[20].

Envisioned characteristics

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 10^{14}– 10^{15} W/cm^2 (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 bellow 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-WB and the ALPS-SYLOS beamlines.

The parameters listed in Table 1 for the SYLOS S-band and the H1-band sources have been estimated under the assumption of attosecond pulses emerging from quasi-phase-matched atomic high-order harmonic generation.

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2.1.2 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.*

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[1]. 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 probably 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[2] for intensities beyond $\approx 10^{21}$ W/cm^2. 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[3].
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[4], at intensities of a few 10^{20} W/cm^2. 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[1,5], for intensities slightly above 10^{19} W/cm^2. 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[6].

Requirements for the driving pulses

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. The "wavelength cubed" approach is one possible method for obtaining isolated attosecond pulses with unprecedented peak-power.

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.

Source development needed for upscaling

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.

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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 multi-Joule 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.

Envisioned characteristics

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[7] are summarized in Figure 2.1. 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.

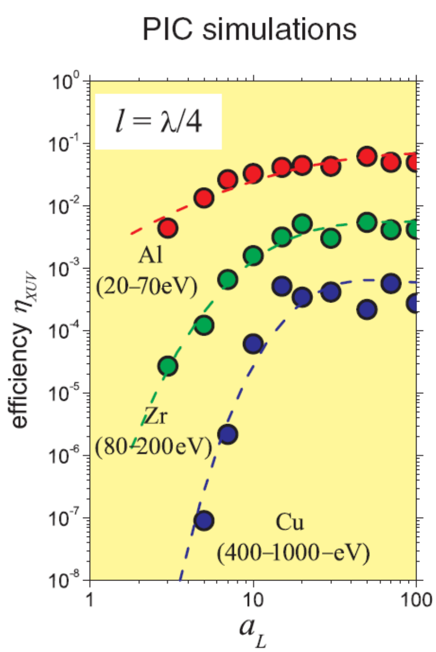

Figure 2.1.
Variation of theXUV pulse efficiency η_{XUV} with the normalized vector potential a_L for three different spectral ranges determined by the indicated thin filter used. From ref. 7

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

2.1.3 Attosecond pulse production via Thomson scattering

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) may 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.

Improvement by double foil target

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 $p_\perp = 0$, cruising with constant $\gamma_x = \gamma$ exactly in normal direction and ready for coherent backscattering with full Doppler factor $4\gamma^2$ [1].

Requirements for drive laser pulses and targets

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 would be very attractive. Of course, it needs to be studied how target systems for repetitive use can be made, in particular, the double layer version[1].

Envisioned characteristics of the secondary source

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 may limit 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[1]. 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 may become a superior tool for attosecond science.

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2.1.4 XUV optics for high-energy attosecond pulses

The generation of high-energy attosecond pulses poses a number of constraints for the design of the XUV beamline.

One of the main issue that has to be solved and that is unique for high-harmonic generation is the co-propagation of the high-energy infrared pumping beam with the generated XUV radiation. This problem is very severe in terms of application of the harmonics: the high-intensity fundamental beam co-propagating with the harmonics may damage the XUV optics of the beamline or even destroy the sample if focused on it. The first optical element of the beamline is then a suitable system that separates the infrared fundamental beam and the XUV radiation. The main requirements for the separation are high damage threshold, high attenuation of the pump pulse, high throughput in the XUV, no alteration of the pulse temporal structure. The study of such a system has a crucial role in the definition of the elements of the beamline. The simplest beam-separation system is the use of a thin metal foil that is almost totally opaque to the IR and partially transparent to the XUV[1]. The drawback is the damage threshold of the filter, which limits the maximum incident IR radiation. A different approach consists in the use of an annular driving beam[2]. The main drawback is the loss of a considerable portion of the pump energy. High-throughput and high damage-threshold beam separators for linearly-polarized 800-nm laser pulses can be realized by a pair of plane plates set at the Brewster angle with respect to the pump wavelength. The design is simple and effective, although it can be used only with linearly-polarized pulses in p-reflection mode. Both Si plates and NbN films have been demonstrated to be effective in the XUV[3,4]. Also plates with multilayer (ML) antireflection coatings for 800 nm working at extreme grazing angles (4°-6°) are available.

Even in the case of very high rejection efficiency of the separation system, the huge difference in intensity between the fundamental and the harmonics (that may be as high as 10^6-10^8) imposes normally the use of so-called solar-blind detectors, which are not sensitive at all to the visible and infrared radiation, e.g. microchannel-plates with suitable photocathodes or metallic photodiode. The use of solid-state detectors (typically silicon photodiodes or CCD cameras) has to be carefully evaluated, since the efficiency of these detectors in the visible and near infrared is comparable or even higher than the efficiency in the XUV and the presence of some radiation in the infrared not completely eliminated by the separation system may add a substantial noise to the useful signal in the XUV.

Suitable optical elements are then required to compensate for the intrinsic chirp of the attosecond pulses. Various techniques have been proposed and employed, based on the use of thin metallic filters[5]; ML mirrors[6]; attosecond XUV-grating compressors[7].

Another important issue for high-energy attosecond beamlines in order to achieve a high intensity on target is the proper design of the XUV focusing optics. There are two main focusing techniques: ML-coated optics and grazing-incidence mirrors. Spot sizes of the order of few tens of micrometers are easily achieved using a ML-coated spherical mirror at near normal incidence[8]. The aberrations can be fully eliminated by using a Cartesian surface (parabola or ellipse) to focus the beam. Spot sizes smaller than 1 um have been obtained by using a SiC-Mg off-axis parabola with short focal length[9]. MLs are designed to work in a defined band. The focusing in a broad and tunable band requires the use of metallic coatings at grazing incidence. The simplest way to focus the beam with a single optical element and no astigmatism is the use of a toroidal mirror with unity magnification. When a strong demagnification is required, coma aberration is unavoidable. Spot sizes of 7 μm have been

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[9] H. Mashiko et al., Opt. Lett. 29, 1927 (2004).

reported, but at the expenses of reducing the XUV beam aperture[10]. The aberrations can be almost eliminated using an ellipsoidal surface at grazing incidence. Spot sizes of 2.4 um have been reported using a platinum-coated ellipsoidal mirror[11]. A different configuration for tight focusing, extensively used for synchrotron applications, is the use of a pair of crossed elliptical or spherical mirrors in the Kirkpatrick-Baez (KB) configuration, where each of the mirrors focuses the beam in one direction. It gives exceptional performances in terms of sub-micrometer focusing of X-ray radiation[12]. Particularly important is the choice of the coating: metallic coatings, primarily gold and platinum, are normally used in the XUV. An alternative coating that is being widely used at present in free-electron-laser (FEL) facilities is highly-dense graphite, since it gives a considerable increase in reflectivity over metallic coatings especially at extreme grazing angles and for wavelengths lower than 10 nm. Based on the strong expertise achieved with FELs, such a coating could be a valid candidate also for the use with attosecond pulses[13].

Depending on the experiment, the design of the beamline may require either the use of all-grazing-incidence elements or a proper combination of grazing-incidence optics to transport the radiation and normal-incidence optics to focus the beam.

Strictly related to the optical design of the beamline is the use of suitable photon diagnostics techniques, that are essential to fully characterize the performance of the source. An ideal detector for ELI source has to cover the full dynamic range of gas and plasma harmonic sources, to be suitable for single-pulse measurements, to exhibit low degradation under radiant exposure in the XUV and soft X-ray and to be ultra-high vacuum compatible and suitable for being assembled under clean room conditions. Furthermore, some experiments may require online diagnostics with minimum interference with the XUV beam: this means that the diagnostics should not block the XUV beam or alter some crucial beam properties such as coherence, temporal duration and intensity to an undesirable extent. The development of novel photon diagnostic techniques for the ELI facility may benefit of the large experience on the existing XUV free-electron-laser (FEL) facilities. In particular, on-line monitoring techniques are widely used to provide real-time information to the users about the spectrum and the intensity of FELs and may be applied also to high-order harmonic XUV radiation.

The measurement of the spectrum is essential both to characterize the source during the development phase and to give to the users the necessary spectral information during the experiment. The use of flat-field grazing-incidence gratings gives the possibility of having single-shot spectrometers which acquire the spectrum on a broad-band with a 2D flat detector. This gives the possibility of measuring simultaneously the spectrum and the divergence of the source[14]. In case of generation at low repetition rates with high laser pulse energies, the user may require the knowledge of the spectrum in the interacting radiation. In such a case, the spectrum can be measured on-line without altering any of the properties of the photon beam but with a small reduction of the flux to the experiment, adopting the same solution as in the case of FELs[15].

Also the measurement of the intensity is of crucial importance both to optimize the photon flux through the control of the generation process and to give to the users the parameters of the experimental conditions. The intensity monitor has to cover the full XUV spectral range of the source. Several options are available, among them we can cite calibrated metallic photodiodes and MCP calibrated detectors. The users may require the on-line measurement of the intensity of the incoming radiation. In such a case, the method already developed and currently used for FELs, i.e. the gas ionization detector, may be applied also to high-order harmonics[16].

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2.1.5 Characterisation of attosecond pulses

The applicability of the generated attosecond pulses relies on the full characterization of the produced radiation: to exploit the pulses to their full potential, their intensity together with spectral and temporal characteristics needs to be determined. On-line and off-line photon diagnostic techniques for the ELI-ALPS facility may benefit of the large experience acquired in the attosecond science community and in the existing XUV free-electron-laser (FEL) facilities

Measuring the temporal characteristics of attosecond pulses is a non-trivial task. The demonstrated techniques are applicable in certain wavelength ranges for pulse durations between 50 – 1000 attoseconds. Currently, the "tool box" for the temporal characterization of the attosecond pulses contains diagnostic tools based on autocorrelation or cross-correlation with a fraction of the generating laser pulse.

The autocorrelation approach used in 2nd-order volume autocorrelation technique[1][2][3] relies on a two-XUV-photon process. It requires a high XUV photon number in the pulse, which is expected to be fulfilled at the facility. In return it provides a direct measurement of the duration of the attosecond pulses. The method has been demonstrated to be applicable for photon energies up to 30 eV, and pulse durations as short as 320 attosecond for gas harmonics2. This method has also been applied for the characterization of XUV attosecond pulses generated from a laser-plasma interaction of 900 attosecond duration and photon energy up to 30 eV 3.

There are different approaches to the cross-correlation measurements:

(1)The RABITT technique[4] is based on the interference between ionization quantum paths XUV + IR and XUV – IR providing information on the relative delay of discrete harmonics, therefore it is only applicable for attosecond pulse trains generated by long driving pulses. The technique has been demonstrated to characterise pulses from harmonic radiation up to 50 eV down to 150 attoseconds[5].

(2) The XUV adaptation of the FROG method, known as FROG CRAB[6] uses a retrieval algorithm for full reconstruction of the harmonic radiation. The method was demonstrated to work for single-cycle isolated attosecond pulses of 130 attoseconds at 36 eV photon energy[7].

(3) In the Attosecond Streak Camera[8], the single XUV pulses is used to create an identical electron wavepacket replica of the pulse through single-photon ionization, which is than streaked by the IR pulse providing a time-to-energy transformation. This method, in combination with the FROG-CRAB technique, has been applied for up to ~100 eV, 80 attosecond pulses[9].

Although the "tool box" of attosecond diagnostics – also containing the recently developed methods of XUV-SPIDER[10] ; Transmission Grating Interferometer[11] and In-Situ attosecond metrology[12] [20] – appears rich, it is not as mature as femtosecond pulse metrology. For a proper exploitation of the ultra-high intensity attosecond pulses, to be delivered by the ELI facility, in a broad area of applications, further enrichment of the existing tool box is required.

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2.2 THz source

State of the art – applications requiring THz radiation

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[1]. 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[2]. Up to 5 μJ energy was demonstrated with 1×10^{-4} efficiency[3]. However, the further scalability of this technique is limited. Air plasmas can also be used for detection of THz pulses[2]. 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[4]. By using $LiNbO_3$ (LN) pumped with fs pulses with tilted pulse fronts[5] (near) single-cycle THz pulses in the 1-THz frequency range with energies up to 50 μJ[6,7] and focused electric field strengths exceeding 1 MV/cm were demonstrated[8].

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.

Source development for upscaling

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, cryogenic crystal temperatures (to suppress THz absorption) in combination with a large-area (~5 cm diameter) contact grating on the surface of LN[9,10]. 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.

Driving pulses

The THz sources of ELI-ALPS are going to be driven by the WB and possibly by the SYLOS primary sources. Since these sources deliver single- or few-cycle optical pulses, they are not optimal for driving the synchronized THz sources directly. For OR a more optimal solution is to use a dedicated synchronized driving laser source for THz generation, such as a diode-pumped Yb-based amplifier chain with ~500 fs pulse duration (which can in its moderate-energy part coincide with the pump chain of the primary sources). Shorter pump pulses can be used with THz generation from air plasma.

Envisioned characteristics

WB

The key parameter determining the characteristics of the THz source is the high repetition rate, together with the experimental requirements. Assuming 1 kW pump power from an Yb-based amplifier, the available pump energy is 100 mJ and 10 mJ for 10 kHz and 100 kHz repetition rate, respectively. In the few-THz frequency

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range THz pulse energies (focused peak electric field strengths) on the 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 THz frequency can be higher, but still high THz field strengths are required, difference-frequency generation in GaSe[11] or THz generation from air are possible alternatives. Especially the latter is compatible with very high repetition rates; also shorter pump pulses (20–30 fs) can be used.

SYLOS or other primary source

By driving OR with 1 J, 500 fs pump pulses (1 kW pump power at 1 kHz) 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 if such a primary source is available.

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3 Periphery (beam steering, targetry, detection systems)

The term periphery includes beam-steering, targetry, experimental area instrumentation and background infrastructure. These are not part of the primary or the secondary sources, however they are essential for users and the successful operation of the facility. Points addressed are: state of the art, requirements, challenges, development needs, possible solutions and layout.

3.1 Beam delivery

In order to provide a flexible operation of the ELI-ALPS system the propagation of laser beams from the primary sources toward the secondary sources and to the experiments – the so-called laser beamlines – have to be optimized. The beamlines from the secondary sources to the experiments are described in detail in the secondary sources chapter. Users should have a wide variety of possibilities to carry out experiments with the different secondary sources. Since the facility is meant for simultaneous use of many experimental stations by different groups of users, who may have conflicting requirements, the maximum flexibility corresponds to a system of fast switches, able to direct individual or multiple laser beams to one or the other stations.

The beams will be directed from the primary sources (in most cases) in vacuum tubes toward the secondary sources and the experiments. Vacuum cubes are to be used in which externally controlled mirrors (flip- and turning mirrors) provide the switching from one primary source to the other one and also the beam-steering from one-secondary source to the other one on a sub-minute time scale. The cubes will be equipped with beam-steering diagnostic markers which allow the continuous control and stabilization of the alignment. Active stabilization will be used to compensate slowly varying pointing instability. High repetition rate or cw pilot lasers will be used for this compensation in the high-field, low-repetition rate beamline. Vacuum valves should allow to open part of the beamlines while delivering laser beams through other ones. Safety interlock systems will be installed to provide equipment and personal protection.

The synchronization of different beams with an accuracy comparable to the laser pulse duration at a secondary source or an experimental chamber is the greatest challenge. The temporal accuracy of 3-5 fs corresponds to 1-1.5 μm in space, which means at least 10^{-7} precision. Slow processes as thermal and vibration effects can be controlled by a feedback after monitoring the beams (e.g. with spectrally and spatially resolved interferometry or spectral comb interferometry). Note that thermal effects can be detrimental, therefore the beamlines should be preferentially temperature-stabilized with high accuracy (< 0.5°C) and the beams to be unified are ideally guided inside the same tubes, parallel to each other. The use of materials with low thermal expansion coefficients, as e.g. invar, are preferred. We expect that these measures will allow the accurate synchronization of the beamlets from a single laser source. It is even more challenging to synchronize laser pulses from the SYLOS and HF primary sources[1] having independent amplifier chains and different repetition rates, even if both are seeded from the same front end.

In order to provide an attosecond beam synchronized with the HF interactions with few-fs accuracy – one of the most important features of ELI-ALPS – the possibility of obtaining the attosecond source from a part of the HF beam before or after the last amplifier is also considered (besides synchronizing SYLOS and HF). In this case the shorter beam path difference will allow a more accurate synchronization than by using a pulse selected from the kHz pulse train of the SYLOS beam. As the laser energy is limited in all cases and the transportation of the high-harmonics radiation introduces significant losses, the synchronized attosecond sources should be placed as close as possible to the HF experiments. Generally, in order to reduce jitter, the experiments/applications must be as close as possible to the secondary sources.

All the beam-steering lines and the experimental chambers have to be pre-pumped by oil-free vacuum pumps in order to reduce contaminations of samples, mirrors, dispersion elements and detectors. At present 3 to 5 secondary sources are planned, and for each laser beam (i.e. WB, SYLOS and HF) 3 to 5 vacuum chambers are

[1] The synchronism with the WB system can be substituted by splitting a part of the front end of SYLOS or HF and generating a low energy kHz repetition rate secondary source comparable to the WB generated pulses. More energy is also available by splitting a small portion of a few mJ from these systems and producing a secondary source with it.

foreseen. A user friendly control should be established which enables the highest level of combinations of sources and experiments. A part of the chambers will be used with permanent experimental setups, whereas temporal experimental installations will be possible in other ones. Some of the chambers will be reserved for the continuous development of sources, some of them will be reserved for user experiments. As this is a rapidly developing area where new ideas are arising on a daily basis with novel user requirements, the laser beam-steering must be prepared as flexible as possible in order to meet the changing user requirements.

3.2 Targetry

Target areas

Target areas (TA) are building sections housing most of the secondary sources and all experimental chambers or stages. The organization of individual secondary sources and experimental stages into different TAs is determined by radiation protection measures, the needs of experiments, and practical aspects of laser and secondary beam delivery. According to the expected radiation dose and the required shielding three groups of TAs are identified (see: figure 3.1).

1. Experiments with no radiation hazard can be carried out in the **non-shielded environment**, which is also the most flexible in terms of experimental arrangement and beamtime distribution. The secondary sources available in these TAs include GHHG sources with moderate photon energy and longer wavelength radiation such as THz / MIR / IR.
2. Experiments producing low radiation doses can be carried out in the TAs with **moderate shielding**. Maximum flexibility will be maintained by using movable shielding walls and blocks. The secondary sources available in these areas are high-photon-energy x-ray sources (surface-HHG, and possibly also incoherent x-ray radiation) and sources of laser-accelerated protons and ions.
3. Experiments with high radiation risk will be located in the **highly protected** TAs. These include kHz-repetition-rate electron sources and electron sources with large bunch charge or higher energy (>150 MeV) but with lower (10 Hz or 0.1 Hz) repetition rate. To allow independent work the shielding geometry should allow access to the experimental stages while experiments are running at other stages.

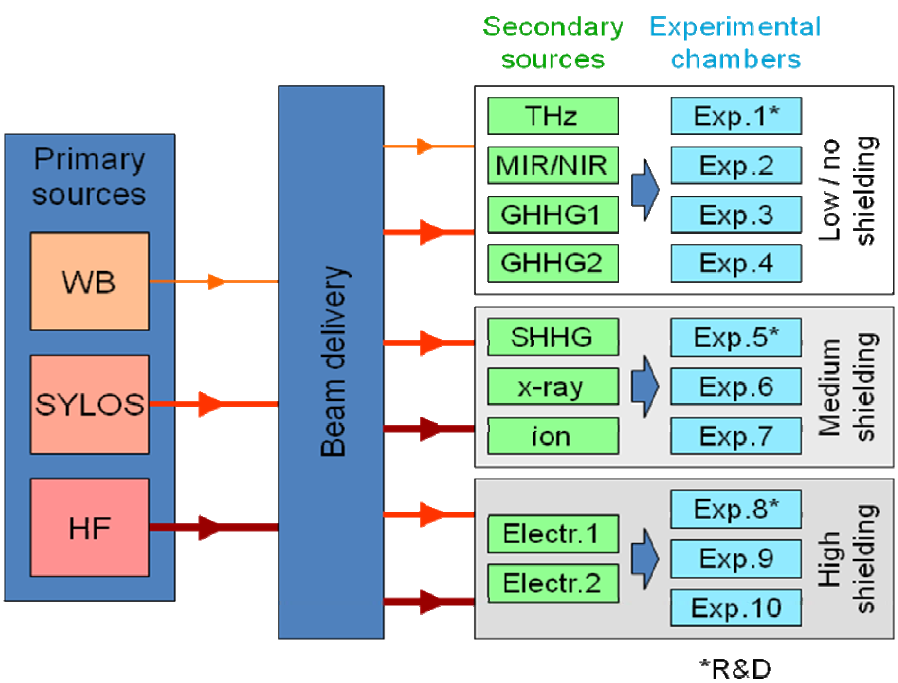


Figure 3.1
Target area layout

In order to make time-resolved pump-probe studies possible with a wide range of radiation and particle sources, a set of design criteria has been established. The criteria are also directly connected to the beam delivery system, therefore some of the following issues were already mentioned in that part.

The layout of the target areas should offer the highest degree of modularity in terms of secondary source and user station layout. Since the installation of the most secondary source target chambers and the subsequent user stations will take place around 2015, one has to consider the option of complying with secondary source schemes that are currently not fully developed. TAs should offer further extendibility including future chamber extensions, additional user stations, movable radiation shielding, etc.. Flexible laser beam delivery system is required in order to enable laser beams at different amplifier stages from any of the laser systems to be transported into different target areas. Separated user-access and source development units are required to allow effective technology developments. State-of-the-art of user-accessible secondary source options will have to be re-assessed in 2013 to enable the finalization of the target areas. Synchronized input from the SYLOS and HF lasers should be possible. Active and passive measures will have to be taken to enable this synchronization, including vibration isolation, the full enclosure of beam paths, active path length stabilization in different laser units, temperature stabilization of the experimental halls, etc. Target areas with different radiation protection should be set up. The less demanding TAs will enable a more versatile layout that can be reconfigured more easily for different types of experiments. TAs for electron acceleration will necessitate complex measures for beam dumping and shielding. The TAs should meet cleanroom conditions, equipped with aerosol monitoring.

Target technology

Various target types will be required for primary and secondary sources (plasma mirror for laser contrast improvement, GHHG, SHHG, particle acceleration) as well as for experiments. GHHG requires continuous or pulsed gas jet targets. Ionization limits the available HHG yield. One way to increase the yield is to use quasi phase matching structures with multiple gas jets or with external fields[2]. Challenges include the development of fast pulsed gas jets, and complying with high vacuum requirements.

An important issue with SHHG is target damage when high pump energies at high repetition rates are used. Several leading groups are intensively working on different schemes to find a suitable target for high repetition rate SHHG. Large-area moving targets with active position stabilization were used in recent experiments. The current developments (4.3 h lifetime with 0.4 mm steps @ 10 Hz, >100 mJ at MPQ, and 1.7 h lifetime with 50 µm steps @ 1 kHz, 1.5 mJ at LOA[3]) hold promise to meet the requirements for the SYLOS (1 kHz, 0.3–1 J) source by further increasing the target size and solving possible mechanical stability problems. Other approaches rely on using metal strip targets (pursued at RAL), or liquid jet targets (vacuum compatibility is questionable). Thin foil targets are required to create a large amount of laser-accelerated electrons for Thomson-scattering to produce high-photon-energy x-ray radiation.

Advanced secondary source designs and many experiments will require even more complex target structures including microstructured species. Significant challenges arise for target fabrication by moving from low to high repetition rates. The challenges fall in two categories: (i) the production of (complex) solid targets at high rates and (ii) accurately place them at the beam interaction point at high repetition rates. Considerable work has already been done in addressing these two general issues, for example at the GEMINI laser in the UK. Furthermore the extensive targetry developments which have taken place within projects such as HiPER and LIBRA are also applicable to the needs of ELI-ALPS.

Specifically, by moving from manual production processes to wafer-based processes production rates of very high accuracy targets and components have been demonstrated along with cost reductions. However, experience shows that (i) initial costs are incurred with both mask manufacture and process development and (ii) the cost savings are most significant for high target numbers (which would generally be the case for ELI-ALPS). It should also be noted that wafer-based production allows for some types of complex targets to be produced.

However, it will not be possible to use wafer production for all types of complex targets and to make such targets several programmes are currently active in automated/robotic microassembly. With the inclusion of automated vision and feedback it will be possible to provide high complexity targets or clusters of targets, but not in very large numbers in the near term.

[2] C. Serrat and J. Biegert, Phys. Rev. Lett. 104, 073901 (2010).
[3] A. Borot et al., Opt. Lett. 36, 1461 (2011).

Mass production techniques suitable for high accuracy microcomponent/microtarget manufacture at the rate of tens of Hz are demonstrably near maturity. Advanced schemes are also currently being pursued in next generation liquid target sources which are compatible with very high (tens-of-kHz) operation rates.

Traditionally targets are transferred to the laser interaction point using a mechanical support (i.e. inserted). In the case of solid targets this is typically done using (i) a tape system, (ii) a wheel carrying targets around its edge and (iii) a mechanical inserter arm which picks up individual targets and places them on a holder in the interaction chamber. Although all of these techniques can be extended beyond the current state of the art it is difficult (although not impossible) to see how they could be used continuously at high repetition rates (kHz).

An additional requirement is the capability to change targets, more realistically carriers holding many targets, under vacuum conditions. This will reduce system downtime caused by changing targets. Target Inserter concepts based on this principle have already been developed and partly demonstrated.

Both HiPER and LIBRA are developing target delivery schemes in which targets are propelled to the interaction point without mechanical support (injected) where a timed laser pulse interacts with them (engagement). For LIBRA a wafer-based injector is being tested with targets which are of a similar scale to some ELI-ALPS primary and secondary targets.

From experience with high repetition rate targetry it will be highly advantageous on ELI-ALPS to solve the production and insertion/injection demands in an integrated solution. The importance for real time target production and placement characterization should not be forgotten, especially for high repetition rates and production numbers, and automated schemes are currently being developed. Finally, suitable target stream quality control processes will be established and implemented.

To serve the above requirements an advanced in-house target fabrication laboratory should be established.

3.3 Target area instrumentation and diagnostics

TA instrumentation will be provided for user and source development groups using the facility. This will include standardized vacuum elements, pumps, control systems and instrumentation (gauges, data acquisition and signal processing electronics, amplifiers, oscilloscopes etc.) to be worked out together with the Czech and Romanian ELI teams. Diagnostics includes that of the laser sources, that of the secondary sources, and the diagnostics of the different experiments. The first two must be standardized for all stations, or the same diagnostics are to be used. An emphasis will be also put on diagnostics development as for example single shot temporal characterization technique for attosecond pulses allowing a much faster measurement approach utilizing the unique high secondary source properties as exceptionally high energy.

The availability of diagnostic tools especially for the secondary sources are of crucial importance for the user experiments. Since such devices or setups are integral and indispensable parts of all experiments, the detailed design of the secondary-source/experimental chambers should take this requirement into account. Some of the diagnostic tools might need considerable space (e.g. electron spectrometer) or additional chambers, therefore they may affect the design of the target areas, too.

3.4 Background infrastructure

A. Service units

To support experimental and theoretical research for users and staff members it is necessary to establish a complex network of in-house service units, which should be supported by highly educated technical staff. The key requirements for all of these units is highest quality, sufficient capacity, short delivery times, creative engineering support for design, and the ability to cooperate with the other service units as well as to interact with research to solve interdisciplinary technological challenges (e.g. vacuum systems, micromachining, nanotechnology). Since the amount of user requests might fluctuate significantly, it can be necessary to contract several external companies, complying with the same high standards, for cases of in-house work overload.

The **optical preparation and coating laboratory** should be capable to design and produce custom multilayer optics up to large apertures covering different wavelength ranges from IR to X-ray. As the requirements for optics of the three ELI facilities will be similar, solving the optical coating requirements within ELI should be a joint effort of the three sites. A facility for electron beam lithography and plasma etching should also be available.

The **optical metrology laboratory** should be equipped with a suite of quality assurance diagnostics. For the characterization of substrate surface and bulk quality and that of optical coatings, laser interferometry can be used. The measurement suite should include: a phase-shifting interferometer for the determination of the 3D wavefront topography; a pulsed-laser-driven interferometer for the characterization of gas jets; a travelling microscope for the measurement of bubble/inclusion content, and coating defects; a spectrophotometer for the optical characterization of coatings; a Stylus type profiler system; a scatter measurement device; a white-light interferometer for group delay dispersion characterization; and a femtosecond damage threshold testbed with a dedicated short-pulse light source. The z-scan technique can be used for characterization of bulk, nonlinear and laser materials by measuring the linear and nonlinear absorption coefficients and the nonlinear refractive index. After a standardized quality assessment procedure to be developed for each type of optical element, the components can be delivered for usage at ELI-ALPS.

To cover the need for customized mechanical components a **mechanical workshop** is needed, equipped with precision tools (CNC machines, etc.), and having a technician team also capable to design, build, test and maintain **vacuum systems**. An **electronics workshop** should be able to design and build complex devices for source and measurement (computer) control, and data acquisition (DAQ). Cost analysis should decide to what extent the above-mentioned workshops should be realized in-house, and what activity would be more economical to be outsourced. A state-of-the-art **target fabrication laboratory** should enable the production of targets for demanding applications, including those with intense high-repetition-rate sources. A **microscope laboratory** equipped with optical microscopes, AFM/STM, SEM, and possibly NMR should complete and complement the optical metrology and target fabrication capabilities. A **computer cluster** with significant computational resources is required by numerical simulations supporting for example laser-plasma experiments or multi-layer mirror development. Additional service units/teams will include a **chemistry laboratory**, a **DAQ & IT group** to support users in DAQ, measurement evaluation and modeling, as well as to maintain the computer facility. The **TA operation and user support group** will be responsible for the daily operation of the experimental infrastructure as well as for developing and running the control system, and serve user needs during specific experimental campaigns. The **laser safety and radiation protection team** will be responsible for the safe access and operation of lasers and TAs, and for the safe design and running of experiments (see next Section).

B. Laser safety and radiation protection

Laser safety and radiation protection is a key issue for ELI, as it is discussed in Chapter 8 of Ref.[4]. Due to the unique features of ELI-ALPS this challenging task requires careful design, and continuous interaction with source and building design. An international expert team will be organized at an early stage of planning the infrastructure to deal with this challenge. The expertise from other fields of science such as that from accelerator and synchrotron community should be utilized as well. The radiation protection system shall be fitted to the requirements of highest flexibility in user facilities. Modular and flexible shielding blocks will be used.

Radiation protection measures will be planned according to the ALARA principle (as low as reasonably achievable). This means that the justifiable radiation exposures resulting from practice must be reduced to the lowest level possible considering the cost of such a reduction in dose. Corresponding to the radiation safety rules dose limits will be taken into account and enforced which involves setting upper limits on the dose that may be received by any member or guest of the institute from all man-made exposures.

[4] G. A. Mourou, G. Korn, W. Sandner, J. L. Collier (Eds.), “ELI – Extreme Light Infrastructure. Science and Technology with Ultra-Intense Lasers. Whitebook” (2011).

After determining the laser input parameters, one can simulate light-matter interaction with a Particle-In-Cell code (for example OSIRIS) including energetic particles (electrons, protons, ions). Beam dumps and shielding elements can be designed and optimized afterwards with the FLUKA code accounting for the neutron radiation as well. Some sophisticated beam dump solutions have already been developed for the Czech ELI facility and for MPQ, the results of which can be adapted to the TAs of the ELI-ALPS facility, too. One has to consider the possible activation of shielding materials, beam dumps and carry out life cycle analysis as well. The amount and handling procedure of potential nuclear waste produced in the facility has to be determined with preliminary calculations. Particles accelerated during high-intensity light-matter interactions can also activate air, water and solid materials which have to be considered in the building engineering designs, especially in that of venting and heating of the experimental halls. Extraordinary events, such as unexpected power loss resulting in the switching off of guide or spectrometer magnets (preferably permanent magnets will be used) and short-lived particle beams hitting the vacuum chamber walls etc. also have to be considered. During normal operation particle beams will be absorbed by a complex and individually designed system of beam dumps and shielding layers made up of multiple materials including steel, graphite, borated polyethylene, aluminum and concrete.

The facility must be equipped with a full radiation protection monitoring system including active and passive detectors and counters. By the time of facility commissioning, staff and user access rules to all the areas including laser bays, experimental areas, TAs and workshops have to be determined in order to minimize exposure. Training has to be provided for all those involved in radiation protection relevant experiments.

4 Strategic partners / network for the pursuit of technological developments

The following institutions are directly or indirectly related to the ALI-ALPS facility:

Hungarian Academy of Sciences KFKI Atomic Energy Research Institute (AEKI), Budapest, Hungary
Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), Debrecen, Hungary
Bern University, Bern, Switzerland
Atomic Energies Commission (CEA) Bordeaux, France
Atomic Energies Commission (CEA) Saclay, France
Central Laser Facility (CLF), Rutherford Appleton Laboratory (RAL), United Kingdom
National Institute for R&D of Isotopic and Molecular Technologies, Cluj-Napoca, Romania
German Electron Synchrotron (DESY), Hamburg, Germany
University of Debrecen, Debrecen, Hungary
Swiss Federal Institute of Technology Zurich (ETH), Zurich, Switzerland
Foundation for Research and Technology Hellas (FORTH), Heraklion, Greece
Friedrich Schiller University of Jena (FSU), Jena, Germany
Göttingen Laser Laboratory, Göttingen, Germany
Heinrich Heine University of Düsseldorf (HHUD), Düsseldorf, Germany
Heidelberg University, Germany
Institute of Photonic Sciences (ICFO), Barcelona, Spain
Institut de la Lumière Extrême (ILE) ENSTA Palaiseau, France
Imperial College, London, United Kingdom
Saclay Institute of Matter and Radiation (IRAMIS), Saclay, France
Instituto Superior Técnico (IST), Lisbon, Portugal
Laboratoire d'Optique Appliquée (LOA), Palaiseau, France
Lawrence Berkeley National Laboratory (LBNL), Berkeley, California, USA
Ludwig Maximilian University of Munich (LMU), Munich, Germany
Waves and Acoustics Laboratory (LOA), Paris, France
Lasers, Plasmas et Procédés Photoniques (LP3), CNRS, Marseille, France
Laboratory for Intense Lasers (LULI), Palaiseau, France
Lund University, Lund, Sweden
Max Born Institute (MBI), Berlin, Germany
Lomonosov Moscow State University, Moscow, Russia
Max Planck Institute for Quantum Optics (MPQ), Garching, Germany
KFKI Research Institute for Particle and Nuclear Physics of HAS (MTA KFKI-RMKI), Budapest, Hungary
Research Institute for Solid-State Physics and Optics of HAS (MTA SZFKI RISSPO), Budapest, Hungary
National Institute for Laser, Plasma & Radiation Physics (INFLPR), Magurele, Romania (NLFPR)
National Research Council (NRC), Canada
Prague Asterix Laser System (PALS), Prague, Czech Republic
University of Padova, Padova, Italy
Politecnico di Milano, Milan, Italy
University of Pécs (PTE), Pécs, Hungary
Queen's University Belfast, UK
University of Strathclyde, Glasgow, UK
University of Stuttgart, Germany
Biological Research Centre (SzBK), Szeged, Hungary
University of Szeged (SZTE), Szeged, Hungary
Budapest Technical University, Budapest, Hungary
Technical University Munich, Munich, Germany
Technical University of Vienna, Vienna, Austria
University of Bordeaux, France
University of Toronto, Canada
Uppsala University, Uppsala, Sweden
Vilnius University, Vilnius, Lithuania
Weizmann Institute of Science, Rehovot, Israel
Fraunhofer Institute for Laser Technology (ILT, Aachen, Germany)
Lawrence Livermore National Laboratory (LLNL), California, USA

MAIN APPLICATION AREAS – UNIQUE RESEARCH OPPORTUNITIES

A. Valence electron science

Motivation / state of the art

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 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.

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 may lead to ultrafast electron transfer processes in extended molecules on the attosecond or few-femtosecond timescale[1,2], which may 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[3]. 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 photoelectron angular distributions that are reminiscent of the structures in the photoionization efficiency measured in EXAFS experiments[4].

Research directions / examples

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 two-color pump-probe experiments where an attosecond pump or probe laser

[1] A.I. Kuleff et al., Journal of Chemical Physics 123, 044111 (2005).
[2] F. Remacle et al., Proceedings of the National Academy of Sciences of the United States of America 103, 6793 (2006).
[3] R. Santra et al., Phys Rev Lett 85, 4490 (Nov 20, 2000).
[4] F. Krasniqi et al., Phys Rev A 81, 033411 (Mar, 2010).

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 not so much the unprecedented pulse energy, but rather 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-WB sources.

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[5]. 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[6]. 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[7], the introduction of attosecond transient absorption in recent experiments on strong-field ionization of Kr[8], and the recent use of wavepacket interferometry in two-color XUV+IR ionization of He[9].

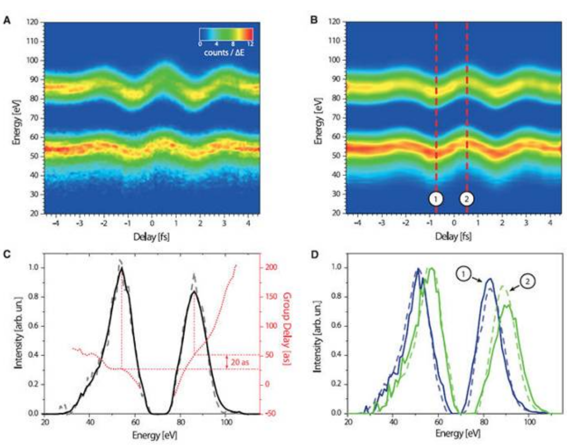


Figure A.1.
Delay between the emission of 2s and 2p electron in XUV photo-emission of Ne, measured by means of attosecond streaking

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[6]. This is where the ELI-ALPS facility allows enormous progress to be made. Using ELI-ALPS it will be possible to carry out these experiments at a hundred-fold increased repetition rate of 100 kHz, providing a

[5] J. Itatani et al., Phys Rev Lett 88, 173903 (2002).
[6] M. Schultze et al., Science 328, 1658 (Jun 25, 2010).
[7] M. Uiberacker et al., Nature 446, 627 (2007).
[8] E. Goulielmakis et al., Nature 466, 739 (Aug 5, 2010).
[9] J. Mauritsson et al., Phys Rev Lett 105, 053001 (Jul 27, 2010).

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.

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[10]. 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 depended with 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.

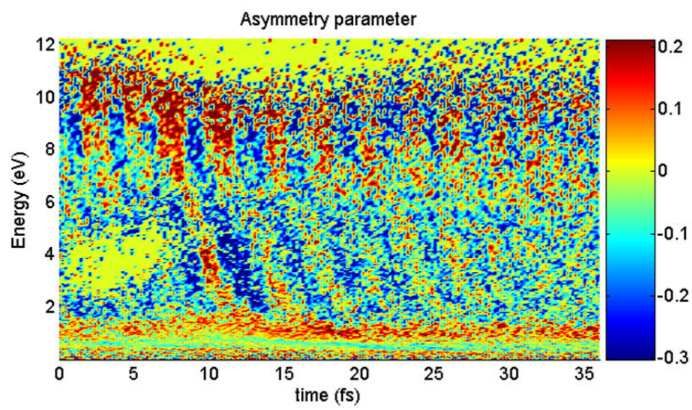


Figure A.2.
Asymmetry in the ejection of D^+ in two-color dissociative ionization of D_2, revealing the attosecond time dependence of two electron localization mechanisms that rely on coupling of electronic and nuclear degrees of freedom

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-WB 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-WB 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.

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] may be viewed as a first example where electron dynamics has been observed in what some have called a "post Born-Oppenheimer

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

regime"[11]. Upon instantaneous (attosecond) photo-excitation, a regime may 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 may 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 WB-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[1,2]. 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 surface science

Surface science represents an area where the ELI-ALPS facilities can have a huge impact. Attosecond surface science is in its infancy, the only results having been reported so far a pioneering experiment which revealed a delay in the photo-emission originating from localized core states in tungsten, as opposed to photo-emission involving delocalized conduction-band states[12]. However, the promise of attosecond surface science is enormous. As has been recently demonstrated (see Figure A.3), angle-resolved photo-emission from molecular crystals allows the reconstruction of static molecular orbital densities that are in good agreement with DFT calculations[13]. Using the ELI-ALPS facilities it will be 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 benefit from the high repetition rate and photon energy range that will be accessible in the ELI-ALPS attosecond sources.

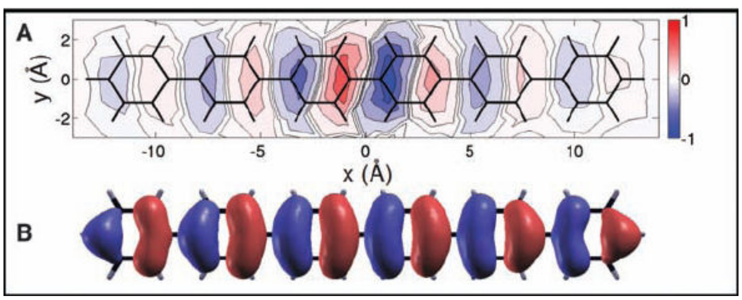


Figure A.3. Comparison between experimentally determined orbital densities for the sexiphenyl molecule and theoretical results obtained by DFT

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[14].

Another area of interest in condensed phase research where the ELI-ALPS facility can make a significant impact is plasmonics. In the last few years this field has attracted huge attention, due – in part - to the promise of plasmonic nano-structures to connect the world of (fiber)optical communication to that of ultrafast nano-electronics. This could speed up electronics by up to 6 orders of magnitude. The ELI-ALPS facility provides unique opportunities for the time-resolved investigation (and thereby optimization) of nano-photonic elements, by carrying out studies that map local field enhancements in nano-structures in real time. In this hugely important area no experiments have been performed to date, although theoretical predictions have been reported[15]. Given that these experiments would most advantageously be performed making use of Photoelectron

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

[12] A.L. Cavalieri et al., Nature 449, 1029 (2007).
[13] P. Puschnig et al., Science 326, 702 (2009).
[14] A. Fohlisch et al., Nature 436, 373 (2005).
[15] M.I. Stockman et al., Nature Phot. 1, 539 (2007).

Emission Microscopy (PEEM), the preferred light source would combine a very high repetition rate with very high photon energies (>>100 eV), i.e. a combination of requirements that will be almost impossible to realize elsewhere.

Outlook / future implications

As described above, the advantages of the ELI-ALPS facilities and particularly the ALPS-WB source for studying valence electron science are manifold. The high repetition rates of the ALPS-WB source allows (i) massive increases in the S/N ratios of atomic experiments, thereby allowing to shed light for the first time on the role of electron correlation in atomic ionization experiments, (ii) 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 and (iii) the wide-spread use of the attosecond technology on surfaces and towards studies of plasmonic structures, 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.

The attosecond sources of ELI-ALPS, in combination with the widely tunable and uniquely controllable wideband primary sources allow unique investigations on molecular and condensed phase systems 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. In these experiments the wideband primary sources serve as the active agent that seeks to control the electron dynamics, while the attosecond source serves as a monitor of the electron dynamics and as a read-out of the final result that is accomplished. If successful, the end result will herald the arrival of waveform-controlled electronics in the PHz regime, with huge impacts on the future of information processing and transferal.

B. Core electron science

Introduction

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.

Two-color time resolved studies of core electron dynamics

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 may 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 WB-P2 beamline photo-ionizes atomic core shells. The synchronized MIR radiation of the beamline WB-T1, with wavelengths 10 µm – 100 µm, may 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 possible but sufficiently high temporal resolution. The combination of these two sources serves perfectly the study of electron relaxation dynamics in the few fs range.

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-S1,S2,H1 or WB-P2 – IR/visible probe by SYLOS or WB) 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[1]. The delayed XUV pulse provided a way of following molecular photodissociation of I_2 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, 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.

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

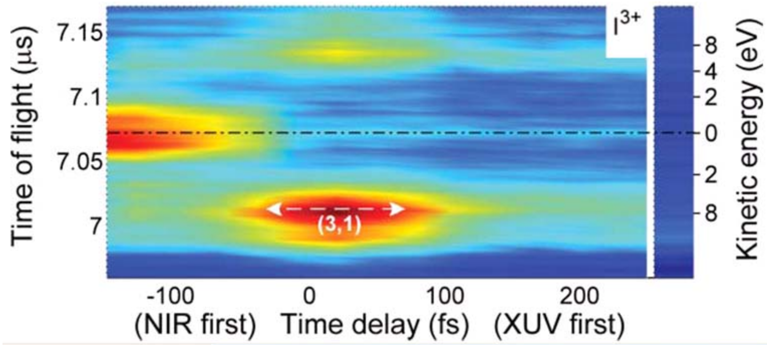


Figure B.1.
Momentum resolved transient I3+ ion yields as a function of the time delay between XUV and NIR pulses. The axis at the right indicates the fragment kinetic energy for ions

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 may overcome this type of limitations. To give an example, the WB-P2 beamline XUV radiation at 3 nm -12 nm may create highly excited transient electronic states, while the WB-P1 UV pulses at 12 nm – 300 nm may select ionization / fragmentation channels.

Another example 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[2]. This experiment requires $\geq 10^{11}$ 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.

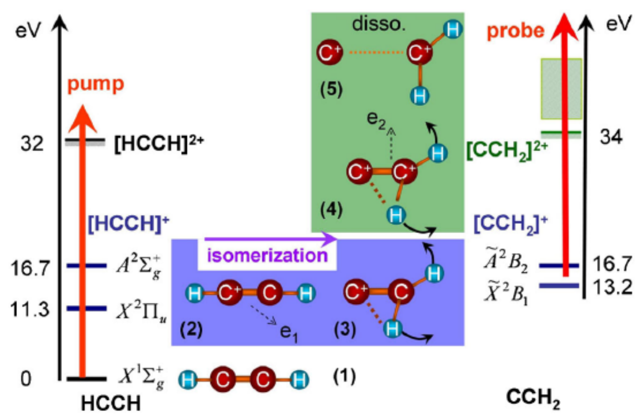


Figure B.2.
Time-resolved imaging of izomerisation dynamics in C_2H_2 cation studied with the XUV-XUV pumpe-probe scheme

Performing this kind of experiments with soft X-rays above carbon K-edge would allow 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[3]. 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.

[2] Y. Jiang et al., Phys. Rev. Lett. 105 263002 (2010).
[3] T. Osipov et al., Phys. Rev. Lett. 90 233002 (2003).

Time-resolved photoelectron diffraction and holography

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.

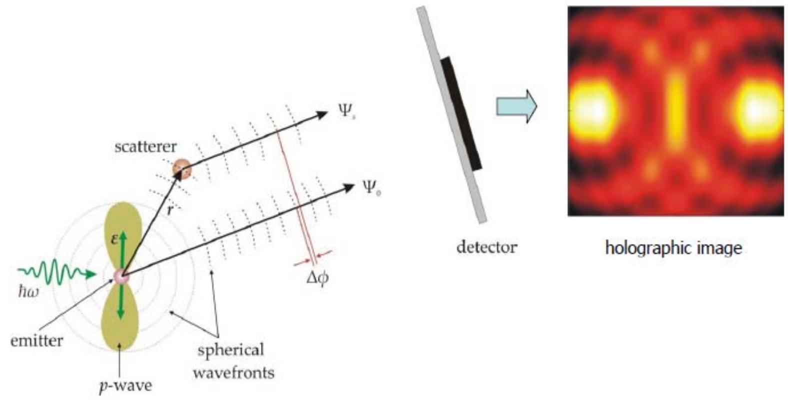


Figure B.3. The sketch of the photoelectron holography approach

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 WB/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 Fig. 3[4].

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.

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

Multi-photon vs strong field interactions

Due to the short wavelengths of the radiation and consequently to the restricted ponderomotive energy interactions are mainly of multiphoton character. Fig. 4 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

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

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, potentially available at ALPS, XUV wavelengths may 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.

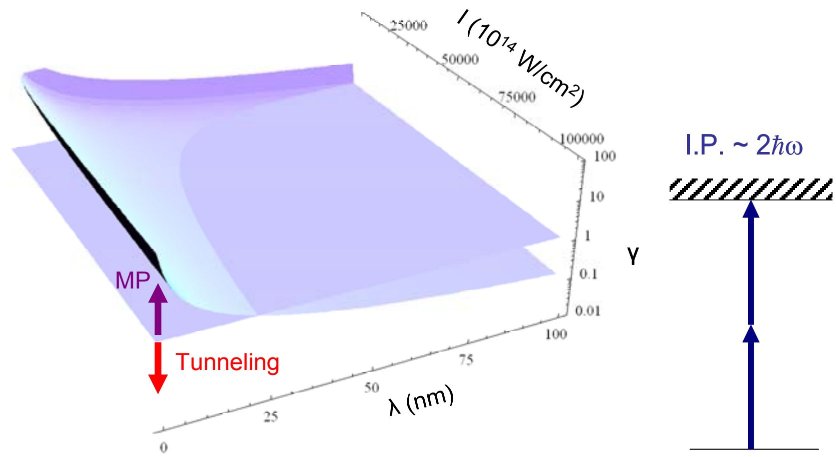
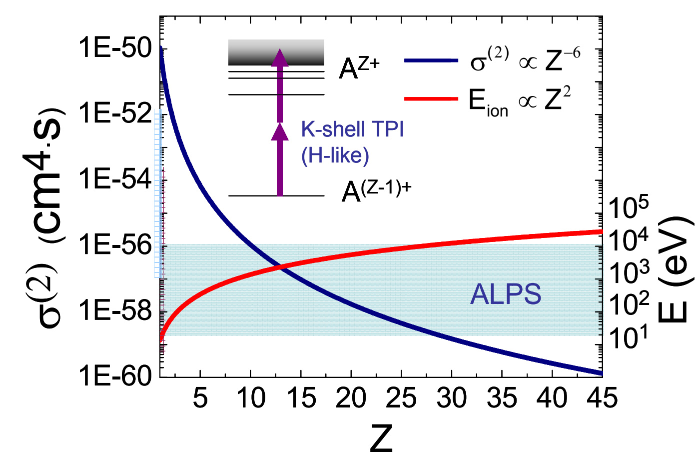


Figure B.4. Adiabaticity (Keldysh) parameter as a function of laser intensity and wavelength for a two-photon ionization scheme

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. Fig. 5 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.

Figure B.5.



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 may 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.

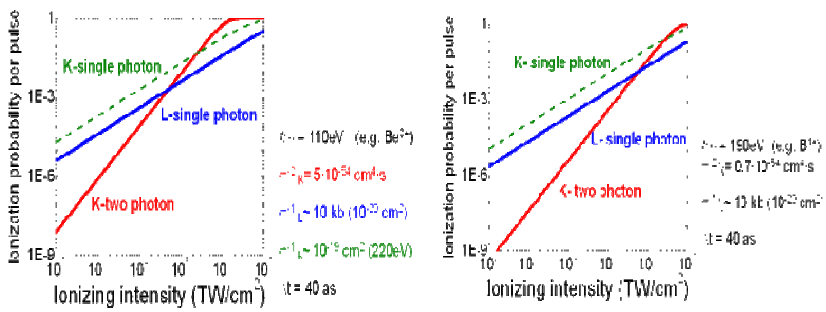


Figure B.6. Two-photon K-shell, single-photon L-shell and single photon K-shell ionization probability of H-like Be and B

Fig. 6 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 5•1016 W/cm2 and 5•1017 W/cm2 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 m3 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.

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 would be 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. 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 would 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. Presumably, these and new under development sources will improve, it is not clear to what extent and when. Reproducible XUV pulses with predictable temporal shape at ALPS will enable conclusive data interpretation in these type of experiments.

The ALPS-ELI 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 cm2, it gives 1019W/cm2. The corresponding photon flux would then be about 1035photons/cm2sec. Taking, to be conservative, a cross section of 10-20 cm2, 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 JPB[5] 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. Presumably the high repetition rate would 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 1019 W/cm2 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 would be a d→f transition with the double well, quite novel behavior in unknown territory can be expected. The above is an extreme case of more modest direct multielectron ejection processes that have been introduced and discussed recently in PRA[6].

5) The above multi-core-electron ejection would be 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).

[5] J Phys B 38, 2119 (2005)

[6] Phys. Rev. A 83, R021407 (2011)

C. Attosecond imaging in 4D

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[1], by high-resolution 'flash' imaging in free electron lasers [2], or by table-top experiments with laser-excited plasmas[3]. Recent findings include the visualization of atomic pathways during phase transformations and melting processes[4,5], nanostructural transformations[6], or the dynamics during explosion of particles/foils[7,8]. 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 Figure C.1), 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.

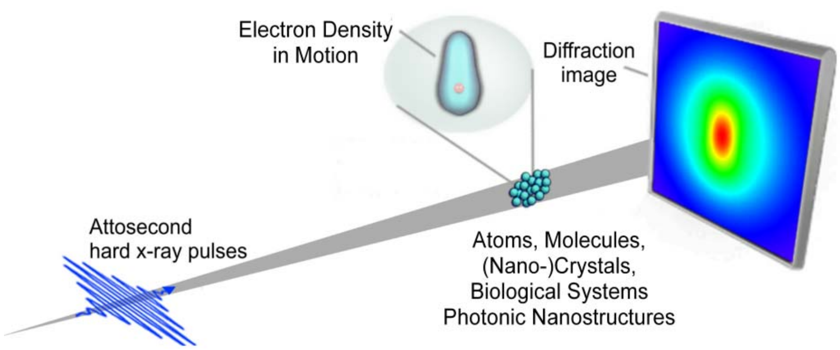


Figure C.1.
General concept for visualizing the attosecond dynamics of charge density motions (see text)

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[1] A.H. Zewail, Science 328, 187 (2010).
[2] H.N. Chapman et al., Nature 470, 73 (2011).
[3] M. Wörner et al., J. Chem. Phys. 133, 064509 (2010).
[4] P. Baum et al., Science 318, 788 (2007).
[5] R.J.D. Miller et al., Acta Crystal. A 66, 137 (2010).
[6] O.H. Kwon et al., Science 328, 1668 (2011).
[7] H.N. Chapman et al., Nature 448, 676 (2007).
[8] C.M. Günther et al., Nature Photonics 5, 99 (2011).

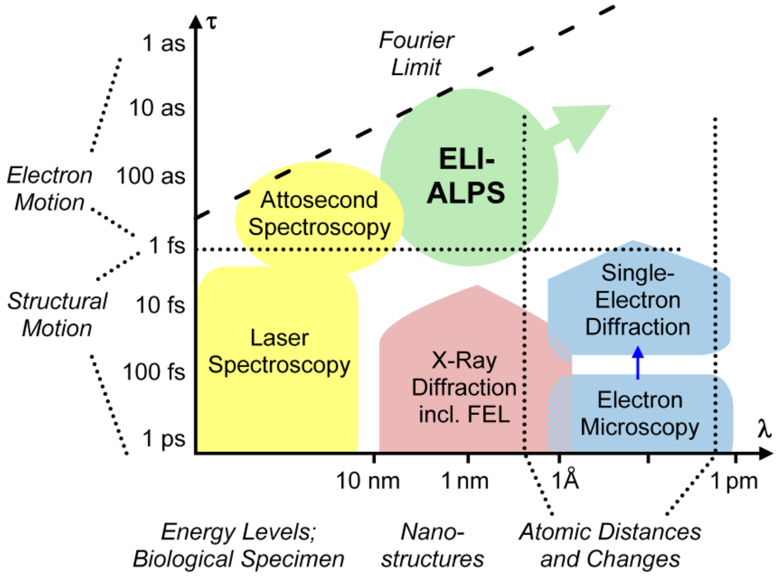


Figure C.2.
Current approaches for visualizing structural dynamics

State-of-the-art

Figure C.2 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 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.

Attoseconds and 4D – general considerations

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 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[9]. Motion of charge densities with femtosecond resolution has already been experimentally resolved with plasma-generated x-ray pulses[3]; 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.

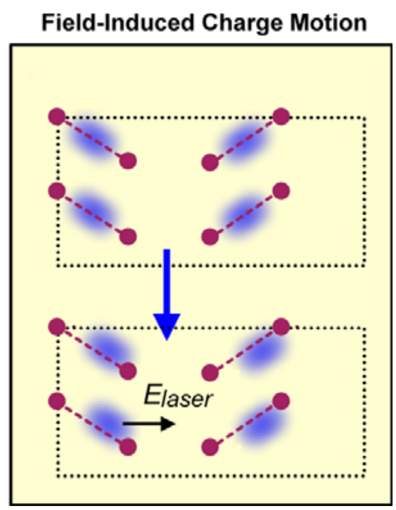
[9] P. Baum et al., Chem. Phys. 366, 2 (2009).

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.

Research directions / examples

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[10,11]. 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, 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 may open up a route towards directed engineering of novel functional materials for optics and lasers.

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[12]. Or, the light-energy conversions process in Bacteriorhodopsin involves electron and proton transfer on ultrashort time scales[13,14], 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.

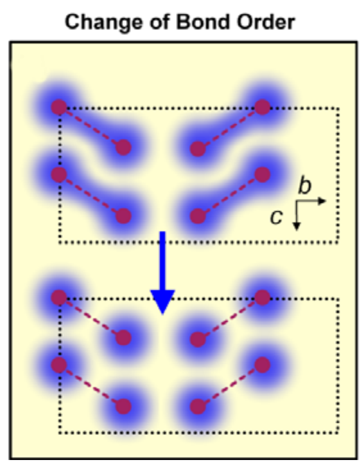
[10] P. Baum et al., Science China G 53, 987 (2010).
[11] H.C. Shao et al., Phys. Rev. Lett. 105, 263201 (2010).
[12] B. O'Regan et al., Nature 353, 6346 (1991).
[13] G.I. Groma et al., PNAS 105, 6888-6893 (2008).
[14] K. Edman et al., Nature 401, 822-826 (1999).

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 could be done at ALPS-SYLOS-S2, maybe -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 may 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 may provide the possibility to image the flow and oscillation of charge within such structures. The nanostructure would be excited with visible, infrared or THz fields, and XUV pulses would 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[15]. 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.

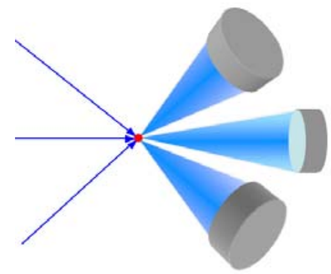
Spatio-temporal inner-shell dynamics

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 possible 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 would consist of a sample of atoms, either as gas or in the condensed phase. We 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

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

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 a possible 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[16,17]. 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[17]. 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.

Outlook / Implications

Historically, one of chemistry's most fundamental questions was solved by time-resolved imaging: the making and breaking of bonds. With our studies, we foresee a similar impact for physics: understanding light-matter interaction on a field↔electron level. 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 we regard our research as fundamental science, we clearly foresee its importance in industrial and medical applications over the next decades.

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

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

Research directions / examples

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

Fast dynamical processes in overdense plasma

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 $T_p / T_L = \sqrt{n_{\text{crit}} / n_e}$, where the critical density is about $n_{\text{crit}} \approx 10^{21}\,\text{cm}^{-3}$ for laser light of period $T_L \approx 3$ fs and the actual electron density $n_e \approx 10^{23} - 10^{24}\,\text{cm}^{-3}$. 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 may play a major role and need to be controlled.

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 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[1].

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

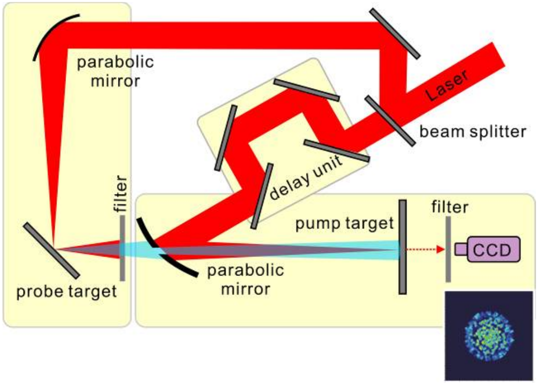

Figure D.1
Schematic of a possible setup for the study of the filamentation instability in dense plasmas

This is susceptible to instabilities leading to strong filamentation of the electron beam thus deteriorating its effectiveness. As the whole process takes place in densities up to 1000 time solid density the corresponding characteristic time for the instability to develop is on attosecond time scale. A thorough study must provide this temporal resolution. A schematic of a possible setup utilizing the combined capabilities of the ELI-ALPS is shown in Figure D.1. Although a simple shadowgraphy is here implied, a more sophisticated technique that will take full advantage of the attosecond temporal resolution like Longitudinal Frequency-domain Spectroscopy[2] or polarimetry[3] can be used.

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 possible setup for this purpose that takes advantage of the capabilities of ELI-ALPS is shown in Figure D.2.

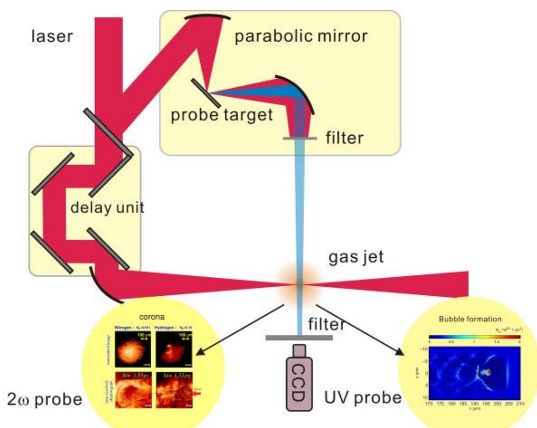

Figure D.2
Schematic of a simple shadowgraphy setup for the investigation of relativistic self-focusing (left inset) or "bubble" formation (right inset)

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[4]. The use of synchronized attosecond pulses in combination with more advanced techniques like Longitudinal Frequency-domain Spectroscopy[2] or polarimetry[3] will provide by far higher temporal and spatial resolution.

Probing nanometer thick foil targets

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[2] N.H. Matlis et al., Nature Phys. 2, 749 (2006).
[3] A. Buck et al., Nature Phys. doi:10.1038/nphys1942, (2011).
[4] R. Fedosejevs et al., Phys. Rev. E 56, 4615 (1997).

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.

D.3a

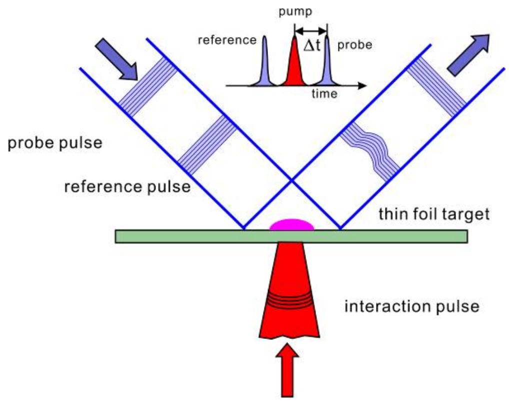
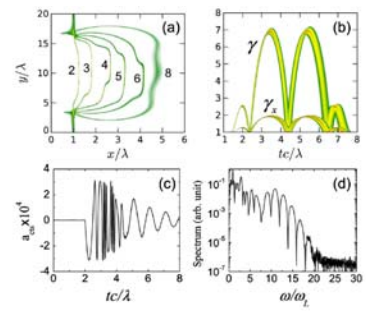


Figure D.3a
The technique of Frequency-domain Interferometry for measuring the density profile of the expanding plasma[7]

D.3b



D.3c

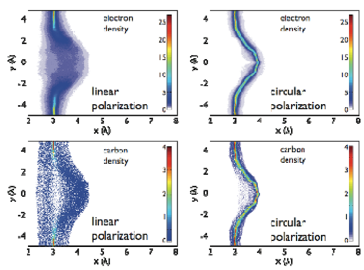


Figure D.3b
Simulation results depicting the generation of relativistically moving electron sheath[5]

Figure D.3c
Two-dimensional PIC simulations of thin foil targets interacting with linearly (left) compared to circularly polarized light (right)[6]

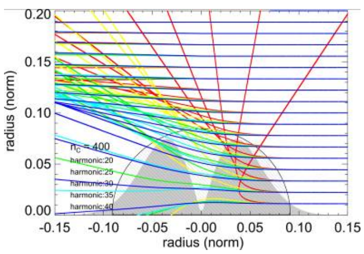
A new concept has been recently proposed in which using nanometer thick foil targets one can disengage the electrons from the ionic background and produce an electron sheath moving at relativistic velocities (see Figure D.3b). This is the ideal „mirror" on which a laser pulse can be coherently reflected to obtain Doppler shifted photons reaching far into gamma rays[5]. 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 (see Figure D.3c), which relies on accelerating the electrons that in turn pull the ions by using circularly polarized laser pulses[6].

In both concepts it is important to be able to ascertain with high temporal resolution the state of the accelerated electron sheath (Figure D.3b) or that of the thin foil target (Figure D.3c). The availability of synchronized attosecond pulses with the PW laser pulses makes this requirement amenable to the well known technique of Frequency-domain Interferometry[7]. This is schematically depicted in Figure D.3a 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.

[5] H.-C. Wu et al., Phys. Rev. Lett. 104, 234801 (2010).
[6] A. Henig et al., Phys. Rev. Lett. 103, 245003 (2009).
[7] J.P. Geindre et al., Opt. Lett. 19, 1997 (1994).

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.



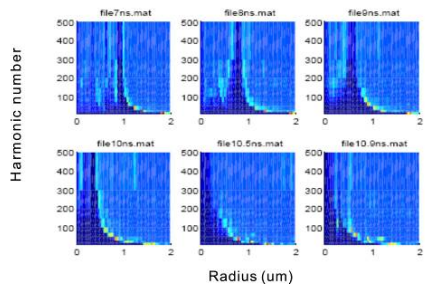


Figure D.4a
Ray-tracing of the indicated harmonic order through a hollow pellet with peak density of 400 overcritical

Figure D.4b
Hydrodynamic simulations depicting the mapping out of the evolution of the compression phase by recording the harmonic signal after passage through the pellet

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 Figure D.4a 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 follow a different path (see Figure D.4a). 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 Figure D.4b.

Strong-field coherent control of nonlinear optical processes.

Due to ac Stark shifts of the atomic levels in the ultra-strong laser fields, the resonance conditions are changed drastically and are function of the applied laser fields. As a result such atomic states may be achieved that are non achievable in weak fields. The picture is complicated further when the ultra-high peak intensity of the laser pulses is combined with ultra-short, atto-second duration of the pulses. The system begins to be highly nonlinear and strong-field coherent control of it opens rich possibilities for enhancing different nonlinear processes including multi-photon ionization, high harmonics generation and others.

In such and similar applications, the initial preparation of atoms (molecules) in coherent superposition of states is of high importance. For example, the importance of the coherent preparation of atoms in the superposition of the ground state and an excited state for high-order harmonic generation and multi-photon ionization in gases was shown already more than a decade ago[8]. It has been demonstrated that such initial preparation of the atoms leads to drastic (by orders of magnitude) decrease of the multi-photon ionization threshold and may lead to significant increase of the efficiency of nonlinear processes (e.g. high order harmonics or Raman sidebands generation). Application of laser pulses of ultra-short duration to such physical problems will generate additional possibilities for coherent control in such prepared atomic ensembles when every laser pulse will simultaneously interact with a number of superposition states.

New and interesting physical effects may be anticipated when the atto-second pulses are frequency chirped. It is known that frequency chirp allows controlling selective population of quantum states which otherwise are not resolved due to the wide frequency spectrum of the pulse. Similar effects may be anticipated for attosecond frequency-chirped laser pulses.

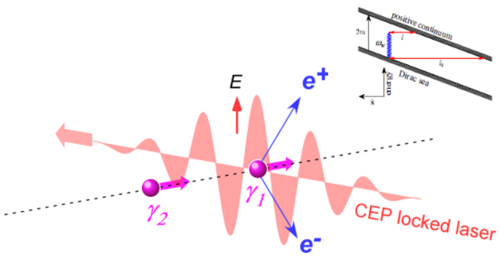
[8] B. Watson et al., Phys. Rev. A, 53, R1962 (1996).

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 $\boldsymbol{E_{SN} = E_{S\sigma}(m_\sigma c^2/\hbar\omega)}$
Schwinger field $E_{Se} = \alpha^{-3} E_K$

Figure D.5
Streaking the vacuum



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

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

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 may 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 may 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[9,10]) including light diffraction on a standing electromagnetic wave[11], four-wave mixing[12], generation of optical harmonics[13] and others. 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 may 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.

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 may survive the gradual increase of the laser pulse intensity up to values that lead to relativistic interactions. Evidence of relativistic interactions was

[9] M. Marklund et al., Rev. Mod. Phys. 78, 591 (2006).
[10] G.A. Mourou et al., Rev. Mod. Phys. 78, 308 (2006).
[11] A. Di Piazza et al., Phys. Rev. Lett. 97, 083603 (2006).
[12] E. Lindstrom et al., Phys. Rev. Lett. 96, 083602 (2006).
[13] A. Di Piazza et al., Phys. Rev. D 72, 085005 (2005).

obtained in high charge states of Xe (Xe^{19+}, Xe^{20+}) through the suppression of direct multiple electron ejection[14], 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.*[15], 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 would 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 may 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.

Outlook / future implications

The contributions expected from the operation of the ELI-ALPS facility would 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 would 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 would open up the route to new ideas that can be tested using this facility. In short, it would provide the scientific community with an additional "eye" with which it would be possible to "see" in real time processes that were held obscure up to now.

[14] M. Dammasch et al., Phys. Rev. A 64, 061402 (R) (2001).
[15] M. Uiberacker et al., Nature 446, 627 (2007).

E. Compact high-brilliance photon sources for biological, medical, and industrial applications

from proof-of-concept experiments at Synchrotron/FEL installations to real-world applications of compact brilliant short wavelength sources

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 experiments, which are currently run or developed at these large scale installations, may 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 $\Delta\omega/\omega$:

$$B = \Delta P/(\Delta A*\Delta\Omega*\Delta\omega/\omega)$$

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.

Laser-based soft-to-hard X-ray sources delivering coherent radiation of high spectral brightness may be realized by (not all of them will be used at ELI-ALPS):

a) Efficient High Harmonic Generation from atoms or surfaces (GHHG, SHHG)[1]

(+) technology matured in attosecond physics, high degree of coherence (HHG)
(–) very low efficiency, currently limited to the (very) soft X-ray range

b) Undulator radiation from laser-driven GeV electron bunches (Table-top undulator/FEL)[2]

(+) undulator radiation experimentally demonstrated, potential for hard X-ray emission
(–) bunch charge limitations, SASE FEL operation not demonstrated yet

c) Relativistic laser-electron Thomson scattering[3]

(+) highest spectral brightnesses theoretically achievable, hard X-ray capability
(–) experimentally most demanding, highest pulse energy laser requirements (~1 J)

d) Betatron radiation from laser-plasma accelerators[4]

(+) high spectral brightness, application experimentally demonstrated
(–) noisy experimental environment, temporal resolution is limited

The envisioned downsizing from large scale facilities to laboratory scale (in spatial size scale as well as potentially in costs) would 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

[1] F. Krausz and M.Ivanov, Rev. Mod. Phys. 81, 163 (2009).
[2] M. Fuchs et al., Nature Phys. 5, 826 (2009).
[3] R. Schoenlein et al., Science 274, 236 (1996); S. Kashiwagi et al., J. Appl. Phys. 98, 123302 (2005).
[4] S. Kneip et al., Nature doi:10.1038/npre.2011.5946.1 (2011).

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.

We have identified three major application areas which benefit from the existence of compact and brilliant X-ray sources:

Biological applications

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 may 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[5]. Furthermore, soft-X-ray microscopy can be complimented by light microscopy in an integrated dual-mode instrument.

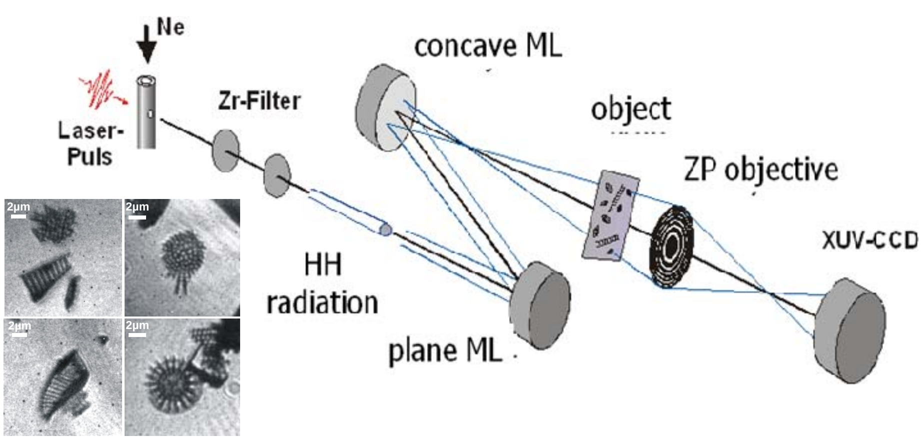


Figure E.1. Principle of an imaging zoneplate soft X-ray microscope using laboratory based coherent High Harmonic radiation + multilayer monochromator and sample images of diatom algae[5]

Figure E.1 displays an experimentally realized HHG zone plate microscope for full-field imaging microscopy by selecting a single High Harmonic 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[6].

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 powerful tool for structural investigation of macromolecular crystals, while recently new coherent x-ray diffraction techniques (Figure E.2) combined with oversampling refinement algorithms have been applied to the structural investigation of non-crystallized macromolecules[6,7]. These new coherent diffractive imaging technologies can be especially useful for structural investigations of membrane molecules, which are hard to

[5] W. Chao et al., Opt. Express 17, 17669 (2009).
[6] 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).
[7] A. Ravasio et al., Phys. Rev. Lett. 103, 028104 (2009).

crystallize and thus only a fraction of membrane protein structures are known from protein crystallography data[8].

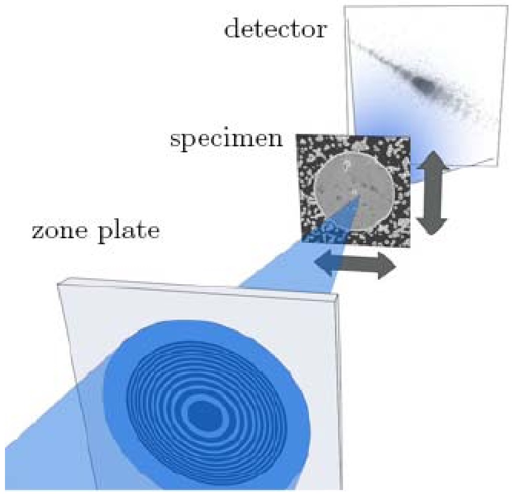


Figure E.2.
Principle of a STXM/CDI experiment[9]

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

Medical applications

Medical applications of brilliant X-rays are found in diagnostics as well as in therapy.

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[10]. The betatron source has been used to experimentally demonstrate phase contrast imaging with a laser-driven X-ray source[4].

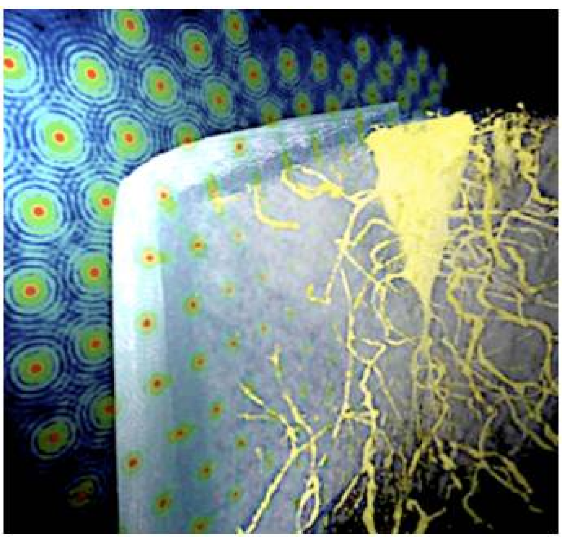


Figure E.3.
Ptychographic hard X-ray image of bone structure using multiple coherent diffraction images and high-resolution (~ 100 nm) 3D image reconstruction[11]

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.

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[8] M. Bergh et al., Quarterly Review of Biophysics 41, 181 (2008).
[9] P. Thibault et al., Science 321, 379 (2008).
[10] T. Donath et al. , Inv. Radiology 45, 445 (2010).
[11] M. Dierolf et al., Nature 467,436 (2010).

Besides phase contrast imaging further x-ray/matter interactions can be utilized for imaging, like small angle x-ray scattering[12], which is also relying on a coherent low-divergence x-ray beam illumination.

The mid-infrared (MIR) spectral region achieved with the planned IR OPCPA primary source (see chapter 1) 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[13] in Laser Dentistry, Laser Angioplasty, Endoscopic Submucosal Dissection and Laser Lithotripsy might benefit from short duration of the MIR pulses as well.

Material science applications

Application of brilliant and wavelength tunable short wavelength radiation in material 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) and X-ray Photoelectron Emission Microscopy (X-PEEM).

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[14]. A highly-monochromatic source in the soft to the hard X-ray regime (best tunable) would be the optimum requirement.

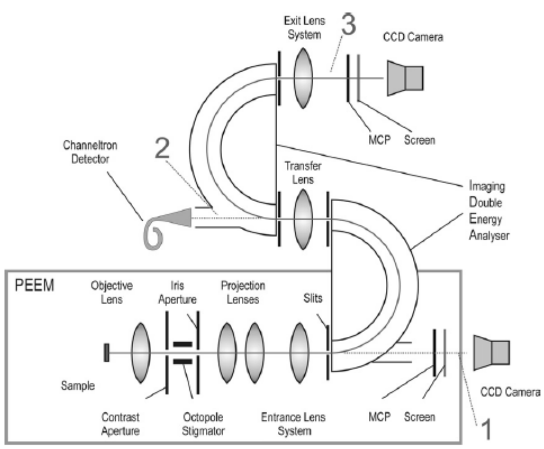


Figure E.4.
Nano-ESCA system for surface nanospectroscopy[12]

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 would perform measurement tasks, which currently can only be run at Synchrotron Radiation facilities due to the high source brightness requirements of the instrument.

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 Extreme Ultraviolet source, condenser optics and all-reflective multilayer-coated imaging objective. One of the most critical issues of EUVL is the

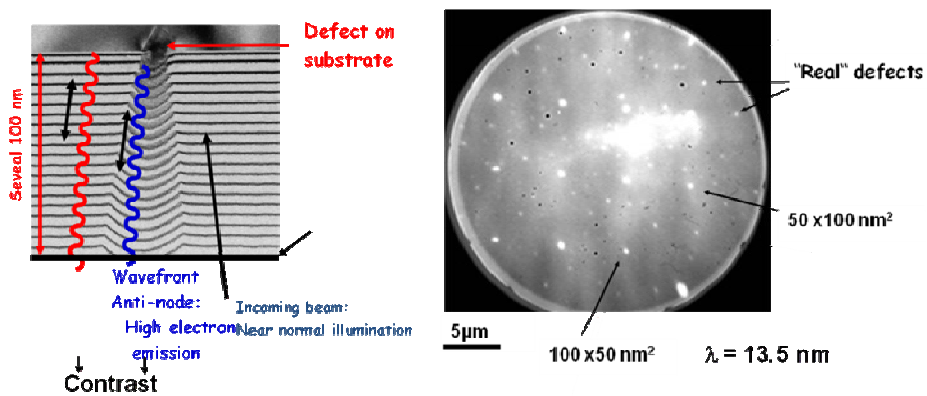
[12] T.H. Jensen et al., Phys. Med. Biolog. 56, 1717 (2011).
[13] K. Ishii et al., Proc. SPIE 6854, 685418 (2008).
[14] M. Escher et al., J. Phys. Cond. Matter 17, 1329 (2005).

realization of defect-free multilayer coated reflection masks. It has been proved, that many critical defects can only be found by *at-wavelength* (actinic) inspection at the lithography wavelength (13.5 nm) and are hidden from already establishedinspection 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 6inch mask within a reasonable time (~ few h) in a laboratory-based inspection tool requires very fast data acquisition (10 nm voxel number ~ 10E14 !) 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)[15] [15, 16]. The photoelectron yield measured from the sample surface by the PEEM is controlled by the electrical field intensity (Fermis 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.

Figure E.5
Schematic setup of a standing wave PEEM and defect image of 50 nm buried defects[13]



This technique (together with other techniques based on darkfield scattering of nao-focused EUV beams) could up to now 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 EUV metrology tool. This integration depends on the availability of a high-brightness laboratory-based EUV source, which may be realized in the future on the basis of high repetition rate High Harmonic Generation.

Outlook / Implications

A large number of biological, medical and industrial applications will profit from and novel application areas will arise due to the hyperspectral radiation sources realized at ELI-ALPS. Most of the advantages is cannot even looked over due to the broad research field and expected rapid scientific evolution triggered by this project.

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