4.2 Project part P02

Attosecond X-ray pulse generation and metrology with phase controlled light

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Allocation to technical disciplines (according to code of OeStat*)

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4.2.1 Summary

Project P02 ‘Attosecond X-ray pulse generation and metrology with phase controlled light’ had four goals:

(A) compression and carrier-envelope phase (henceforth: C-E phase) stabilization of TW laser pulses
(B) reproducible generation and full, accurate characterization of the temporal shape of the sub-fs harmonic radiation
(C) reducing the duration of the XUV bursts to less than 300-as
(D) measurement of the driving field's C-E phase.
(E) All of them can be regarded as – at least party - fulfilled.

(A) The unique Advanced Femtosecond Source (AFS) developed in the first period of ADLIS was combined with control and diagnostic techniques for the C-E laser phase. Compression to less than 10 fs was realized. While the 1 TW frontier could not quite be reached the laser system was used to generate high energy soft-x-ray photons in a regime never reached so far.

(B) A new measurement system called Attosecond Streak Camera was used to fully characterize the sub-fs XUV bursts. With this method at hand one is able to determine the spectral chirp of the XUV pulse and hence for the duration of the pulse to a remarkable accuracy (+/- 8 attoseconds at 250 as pulse duration).

(C) Using new extremely broadband multilayer mirrors for filtering harmonic radiation in the cut-off region we were able to generate single, isolated 250 attoseconds pulses which we could fully characterize using the method mentioned above.

(D) The C-E phase of the XUV pulse’s driving field was measured on the one hand by utilizing the phase sensitivity of above-threshold ionization. On the other, hand the electric field waveform of the laser pulses was traced completely using the sub-fs XUV bursts giving the full information on all parameters of the field including, of course, the C-E phase.

4.2.2 Results and discussion

Measurement of microscopic processes in real time requires a definition of the zero of time (when the process gets going) with sufficient accuracy and subsequent probing of the phenomenon with sufficient resolution. Prerequisite for real-time measurements is a physical quantity that varies on the relevant time scale in a controlled and reproducible manner. The
variation of this quantity allows elapsed time to be measured just as the direction of the hand of a stopwatch does, provided that the fast-varying physical quantity is able to interact with the microscopic process to be investigated. The hyperfast oscillating electric field of visible or near-infrared light is ideally suited to studying electronic dynamics in real time. The electric field exerts a force on the electrons, providing the necessary coupling, and varies on the relevant, atomic, time scale, affording the potential for attosecond control and measurement of electronic processes.

For this potential to materialize, control and measurement must be performed under well-defined and well-reproducible conditions. To this end, a single oscillation period needs to be isolated in the light wave and precisely reproduced from one laser shot to the next. Isolation of an oscillation period is feasible if and only if individual cycles of the light wave are distinguishable. This applies only if the light pulse comprises merely a few wave cycles (henceforth: few-cycle pulse). Pulse-to-pulse reproduction of the electric field evolution in a selected (e.g. the most intense, central) cycle calls for precise reproduction of the electric-field waveform of a few-cycle laser pulse. This was accomplished for the first time by our group in collaboration with T. Udem, R. Holzwarth, Ch. Gohle, and T. W. Hänsch by drawing on a technique first proposed by T. W. Hänsch in 1997 and is now in widespread use for optical frequency-comb synthesis with mode-locked lasers. As a result, intense, waveform-controlled few-cycle pulses of visible/near-infrared laser light are now available for exciting, controlling, and measuring electronic processes on an attosecond time scale.

The controlled generation and precise measurement, i.e. synthesis, of few-cycle, monocycle, and possibly sub-cycle waveforms of intense laser light composed of many different colors (spanning more than an octave) are of key importance for attosecond science coming of age. The controlled electromagnetic forces the synthesized light waveforms exert on charged particles afford promise of controlling quantum transitions of electrons in atoms and molecules to an extent never achieved before. A prerequisite is the reliable generation of isolated soft-X-ray (0.1 - 10 keV) pulses of atomic-scale duration (<100 attoseconds) synchronized to their few-cycle laser driver and their measurement. These emerging attosecond tools and measurement techniques allow time-resolved measurements of electronic transitions in atoms and molecules.

(A) Compression and carrier-envelope phase stabilization of TW laser pulses

One of the most challenging tasks in ultrafast laser technology is to cross the 1-TW-border with few-cycle (i.e. < 7 fs at 800 nm) laser pulses. Huge efforts have been done in this direction. Finally, with more than 1.5 mJ in 5 fs pulses we approached this limit (J. Seres 2003, A.J. Verhoef 2005). With such a powerful source at hand we started to push the cut-off
energy of high-order harmonic generation towards higher photon energies. Using absorption edges for calibration we found radiation far beyond the titanium L-edge indicating harmonic radiation exceeding 1 keV photon energy (J. Seres 2005).

(B) Reproducible generation and full, accurate characterization of the temporal shape of the sub-fs harmonic radiation

Control of the evolution of the electromagnetic field of visible/near-infrared light has provided the rapidly-varying physical quantity required for measuring brief time intervals and fast processes and opened the door to time-resolved studies on an attosecond time scale. The precisely controlled and known variation of the electric field in a selected oscillation cycle of a waveform-controlled few-cycle light pulse (e.g. in the most intense cycle at the peak of the pulse) can provide the reference “clock” for temporal measurements. However, attosecond metrology relies on our ability not only to measure attosecond processes but also to create them in a reproducible manner and in perfect synchronism with the “clock”. Clearly, the latter requirement can be met only if dynamics is triggered by the “clock” itself.

To this end, we have exposed atoms to the synthesized few-cycle laser field. The field is linearly-polarized and strong enough to ionize the atoms with substantial probability. However, before the most-weakly-bound (valence) electron is detached from the atom and becomes free once and for all, the center of gravity of its wave function performs a few giant oscillations near the atomic core. The amplitude of the oscillation driven by the most intense field cycle can exceed the Bohr radius by several orders of magnitude. The electron’s wiggling center-of-gravity motion is harmonic with the driving laser field, except for instants when the electron gets close to the atomic core. Here the motion becomes strongly inharmonic due to the influence of the atomic Coulomb potential. As a result, a high-frequency burst is emitted whenever the electron returns to the core. In a few-cycle light field with a cosine waveform the wavepacket recolliding near the zero transition of the laser field following the pulse peak is significantly more energetic than the others, resulting in the creation of a burst at higher photon energies than other recollision events. Filtering these photons with a suitable broad-band bandpass filter isolates a single pulse of sub-femtosecond duration in the emitted XUV/soft-X-ray radiation. This synchronized sub-femtosecond XUV pulse emerging from giant atomic dipole oscillations together with its generating few-cycle light wave constitute our key tools for attosecond metrology and spectroscopy.

By drawing on these tools in collaboration with M. Drescher, U. Kleineberg, T. Westerwalbesloh, and U. Heinzmann from the Univ. of Bielefeld and with P. B. Corkum from NRC Canada we developed an attosecond measurement technique that reverts to the well-
known concept of streak imaging. In our experiments electrons ejected from atoms upon irradiation with an XUV or X-ray pulse are deflected with a light field - varying millions of times as fast as the deflecting voltage ramp in a conventional streak camera. The light field imparts its effect in space and time immediately on release of the electrons. In a detection geometry where the laser electric field is parallel to the direction of detection, the initial momentum of the electrons ejected along the light field vector is increased or decreased by the laser field, depending on the instant of birth of the electrons. In this manner the successively emitted electrons are detected separately again, but this time not spatially on a screen but alongside one another on the energy scale. The width $\Delta E$ and shape of the measured energy distribution of the electrons reflect the duration and evolution of the electron emission, just as their spatial distribution in conventional streak imaging. In this case, however, „deflection” occurs within half a light period, resulting in attosecond time resolution.

The emission of photoelectrons instantly reflects the temporal variation of the excitation process, in our case that of the X-ray intensity. As a consequence, the “streak images” of the photoelectrons recorded at different timings with respect to the “streaking” laser field allow temporal characterization of the exciting XUV/X-ray pulse (R. Kienberger 2004). By measuring the emission time of the photoelectrons by this new streak method we were able to determine the duration of our $\sim$100-eV XUV pulses as 250 attoseconds (Fig. 1).

With these tools and techniques the door is now open to attosecond spectroscopy of electronic processes in atoms and molecules. Limited by a maximum streaking of +/- several 10 eV for $\sim$100 eV excitation, our streak camera currently has a resolving power of $\sim$100 attoseconds. Increasing the excitation photon energy towards (and beyond) 1 keV, which was reported in the section above, will permit streaking of several hundred eV and thereby result in a resolution that is comparable to the atomic unit of time, 24 attoseconds.

(C) Reducing the duration of the XUV bursts to less than 200-as

Reducing the duration of attosecond XUV harmonic pulses to less than 200-as requires the generation process to be confined to a single oscillation of the driving field for a large fraction of the harmonic spectrum. Compression of the laser pulses to 4.3 fs was a step in this direction and performs – with current techniques - practically the ultimate step towards the natural limit of one oscillation cycle, which is about 2.7 fs in the 800 nm wavelength range. Multilayer mirrors for the XUV regime being extremely broad in spectrum (>10 eV around 100 eV) provided by our collaboration partners from Bielefeld, M. Drescher, U. Kleineberg, T. Westerwalbesloh, and U. Heinzmann paved the way for the generation of 250-attosecond pulses. As it was pointed out in the previous section we were able to fully
characterize these pulses including the spectral chirp which led to the remarkable accuracy of +/- 8 as. These are the shortest (isolated) pulses ever observed and also the shortest time span measured hitherto. Figure 1 shows a trace of streaked electron spectra being used to retrieve pulse duration and chirp of the sampling attosecond pulse.

![Figure 1: Trace of electron spectra streaked by the electric field of a few-cycle laser pulse. The insert shows the retrieved pulse duration and chirp of the XUV pulse.](image)

**(D) Measurement of the driving field's C-E phase.**

The first source of waveform-controlled light uses Ti:sapphire as the broadband gain medium and a hollow-fiber/chirped-mirror pulse compressor along with a couple of phase control loops for the generation of 5-fs, 0.1-TW pulses at a carrier wavelength of 750 nm and a repetition rate of 1 kHz. The laser system delivers pulses with a constant waveform, as a result of the constancy of the amplitude envelope, the carrier frequency, and the timing of the field oscillations with respect to the pulse peak. In the carrier-envelope representation of the light field, this timing is mathematically denoted by $\phi$, the carrier-envelope phase. While the phase control loops allow $\phi$ to be stabilized its value to be shifted by a known amount, its absolute value is still unknown. For synthesisization of a specific (e.g. “cosine”-shaped for $\phi = 0$ or “sine”-shaped for $\phi = \pi/2$, waveform, stabilization of $\phi$ is not sufficient and its exact value also has to be known.
To this end, we ionized atoms with our linearly-polarized few-cycle light pulses and detected the liberated electrons with two electron spectrometers aligned with the laser polarization on opposite sides, in a scheme invented by G. G. Paulus and H. Walther. (G. Paulus 2003) The optical-field ionization experiments were conducted in collaboration with G. G. Paulus, F. Lindner, M. Schätzel, and H. Walther. After their liberation the electrons interact with the strong laser field and pick up a drift momentum that depends sensitively on the waveform of the few-cycle pulse. As a result, the energy distribution observed on the left- and right-hand sides are generally different and from the left-right asymmetry the absolute value of $\phi$ can be unambiguously determined by comparison with a simple semi classical model of optical-field ionization. These experiments, along with the evaluation of the amplitude envelope and carrier frequency from standard pulse diagnostics, allowed for the first time complete determination of the evolution of the electric field of a light pulse (including its direction: $\phi = 0$ implies an electric field vector pointing towards the right detector on the pulse peak).

**Fig. 2:** False-color representation of the energy distribution of a 250-attosecond electron probe versus delay between the few-cycle laser pulse and the 95-eV XUV pulse releasing the electron probe.

The 250-as photoelectron bursts knocked free by the XUV pulse also permits the evolution of the laser electric field to be traced. This is because the momentum imparted to the electron is proportional to the instantaneous value of the vector potential of the field at the instant of electron release. Hence, the temporal evolution of the vector potential can be
tracked by measuring the energy shift of the sub-femtosecond electron wavepacket versus delay between the few-cycle laser pulse and the XUV pulse releasing the probe electron (Fig. 2). The data clearly reveal – without any cumbersome analysis – that a single XUV pulse of sub-femtosecond duration synchronized with sub-femtosecond accuracy to the laser field was at work in this measurement. Furthermore, the temporal evolution of the electron energy shift (displaying that of the laser field’s vector potential) appears to be sinusoidal, implying a “cosine”-shaped electric-field waveform (Fig. 3), which was found to be a prerequisite for creation of a single XUV burst. The experiment can be regarded as the first direct and complete measurement of light: direct, because the field is directly measured, and complete, because not only is its temporal variation obtained, but also its strength and direction (E. Goulielmakis 2004).

![Graph showing electric field of a 4.3 fs, 750-nm laser pulse measured directly by an attosecond electron probe.](image)

**Fig. 3:** Electric field of a 4.3 fs, 750-nm laser pulse measured directly by an attosecond electron probe.

### 4.2.3 Collaboration within and beyond the SFB

- Cooperation with P03 – Ch. Spielmann: A couple of experiments in the framework of P03 were done at the Advanced Femtosecond source using the same cross-correlation setup as in P02.

- Cooperation with P12 – G. Reider, M.Drescher: First experiments for solid state surface photo emission were performed and are going on at the moment under improved vacuum conditions.

- Cooperation with P16 – M.Lezius: Preliminary experiments on time-resolved dissociation in H2 were done. The stringent vacuum conditions necessary for a
successful experiment forced us to build a completely new beamline which is being commissioned at the moment to be used for a further cooperation with P16.

- Cooperation with P17 – M. Drescher, F.Krausz: An experimental stove providing metal vapor for the Coster-Kronig experiments is under final construction. As soon as it is finished these experiments can carry on.

- Cooperation with P11 – A. Scrinzi: Theoretical research in P11 provided invaluable support in evaluating the full XUV pulse characterization form the Attosecond Streak Camera measurements.

- Collaboration with University of Bielefeld: Multilayer mirrors for the XUV regime being extremely broad in spectrum (>10 eV around 100 eV) provided by our collaboration partners from Bielefeld, M. Drescher, U. Kleineberg, T. Westerwalbesloh, and U. Heinzmann paved the way for the generation of 250-attosecond pulses.

- Collaboration with Max Born Institute, Berlin: We set up a so-called bulk-compression scheme to compress multi millijoule pulses at the MBI laser system and got close to the Fourier-limit. This experiment was pre-experiment for the compression of the Advanced Femtosecond Source pulses