Single sub-fs XUV pulses: generation and measurement

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The change from a zero transition to the maximum amplitude of the electric field of visible light lasts shorter than one femtosecond (1 fs = 10^{-15} s). By precisely controlling the hyperfast electric field oscillations in a short laser pulse we developed a measuring apparatus – the Atomic Transient Recorder (ATR) - like an ultrafast stopwatch. This apparatus is capable of measuring the duration of atomic processes with an accuracy of less than 100 attoseconds (1 as = 10^{-18} s) which is the typical duration of electronic processes (transients) deep inside atoms. A 250-attosecond X-ray pulse initiates the atomic process to be measured and the attosecond stopwatch at the same time. This new measuring method now allows for the first time the observation of ultrafast processes in the electron shell of atoms.

Efforts to access ever shorter time scales are motivated by the endeavour to explore the microcosm in ever smaller dimensions. Recently, femtosecond laser techniques have allowed control and tracing of molecular dynamics and the motion of atoms on the length scale of internuclear separations without the need for resolving the objects of scrutiny in space[1]. Here we demonstrate that laser light consisting of a few, well-controlled field oscillations[2] extends these capabilities to the interior of atoms, allowing control and measurement of electronic motion on an atomic scale of time. The measurement of ever shorter intervals of time and tracing dynamics within these intervals relies on the reproducible generation of ever briefer events and on probing

26th International Congress on High-Speed Photography and Photonics, edited by D. L. Paisley, S. Kleinfelder, D. R. Snyder, B. J. Thompson, Proc. of SPIE Vol. 5580 (SPIE, Bellingham, WA, 2005) · 0277-786X/05/$15 · doi: 10.1117/12.580483

techniques of corresponding resolution. The briefest events produced until recently have been pulses of visible laser light, with durations of around 5 fs (1 fs = 10^{-15} s) [3,4,5]. Recently sub-femtosecond bunching of femtosecond (> 10 fs) extreme ultraviolet light (XUV) was observed in two-colour[6,7] and two-photon[8] ionization experiment and evidence of sub-fs confinement of XUV emission from few-cycle-driven (ionizing) atoms was also obtained[9]. In this paper we review most recent advances in ultrafast science. These led to the controlled, reproducible generation and accurate measurement of isolated sub-femtosecond X-ray pulses, which – along with novel measurement techniques – now open to way to tracing electronic dynamics deep in the interior of atoms directly in the time-domain with a resolution well within the classical Bohr orbit time in the inner orbit of the hydrogen atom (150 as).

Basic concepts for attosecond pulse generation and time-resolved atomic spectroscopy on an attosecond scale

Intense laser pulses consisting of a few, precisely-controlled oscillations of the electric field[2] constitute the key to both the generation of isolated attosecond pulses and time-resolved measurements on an attosecond time scale. The electric field of linearly-polarized femtosecond laser pulses, if sufficiently strong, induces – in a highly nonlinear interaction – gigantic dipole oscillations by pulling an electron out of the atom and smashing it back towards its core half a cycle later. The oscillations are not sinusoidal but contain very high frequency components extending into the extreme ultraviolet and soft-X-ray regime[3]. In a laser field containing many oscillation cycles, the oscillations are repeated quasi-periodically, resulting in the emission of a series of high-energy bursts of sub-femtosecond duration and high-order harmonics of the laser radiation in the spectral domain. For a few-cycle laser driver only a few dipole oscillations, different in amplitude occur. The oscillation with the highest amplitude has been predicted to produce a single burst in the spectral range of the highest emitted photon energies[2].

First evidence for the emission of an isolated sub-femtosecond pulse from this interaction was reported in 2001[9] but the reproducible generation of single sub-femtosecond pulses had not been possible before the giant atomic dipole oscillations could be precisely controlled with intense waveform-controlled few-cycle laser pulses in 2003[2]. With waveform-controlled few-cycle light, the few giant atomic dipole oscillations induced can be precisely controlled and reproduced from one laser shot to the next. This is expected to result in an x-ray burst with parameters well reproduced from one shot to the next which is a basic prerequisite for these tools being used for time-resolved measurements on an attosecond time scale.

The synchronism of the X-ray burst to the field oscillations of the generating laser pulse offers the potential for using the X-ray burst *in combination with* the oscillating laser field for attosecond spectroscopy. This is essential because these laser-produced X-ray bursts are too weak to be used for *both* triggering *and* probing electronic dynamics (X-ray-pump/X-ray-probe spectroscopy). Instead the oscillating laser field, which changes its strength from zero to maximum within some 600 attoseconds in a 750-nm laser wave, can take over the role of the probing X-ray pulse. In the following chapter we shall see how the hyperfast electric field oscillations of the few-cycle visible laser pulse that previously produced the sub-femtosecond X-ray burst can be employed for taking "tomographic images" of the time-momentum distribution of electrons ejected from atoms following an impulsive excitation by the synchronized X-ray burst. From these images the temporal evolution of *both* emission intensity *and* initial momentum of freed electrons can be retrieved on a sub-femtosecond time scale. Probing photo- or Auger–electrons yields insight into excitation and subsequent relaxation processes, respectively. The transients can be triggered by an isolated attosecond electron or photon burst synchronized to the probing light field oscillations. The technique draws on the basic operation principle of a streak camera[10-14] where a light pulse generates an electron bunch having exactly the same temporal structure. The deflection of the electrons in an electric field allows the reconstruction of the duration of the electron bunch. By measuring the temporal evolution of

emission intensity *and* momentum distribution of positive-energy electrons, the atomic transient recorder (ATR) provides direct temporal insight into the rearrangement of the electronic shell of excited atoms on a sub-femtosecond scale[15].

From electron-optical chronoscopy to recording atomic transients

The sudden deflection of a light flash produces a streaked spot of the beam, allowing the measurement of the flash duration[10]. Implementation of the same concept with photoelectrons allowed electron-optical chronoscopy by image-tube streak cameras[11-12]. From the streaked image of the bunch of photoelectrons the temporal structure of the light pulse can be inferred with sub-picosecond resolution. The time resolution of these devices is ultimately limited by the spread of the electron transit time due to a spread of their initial momentum. In this work we report temporal characterization of sub-femtosecond electron emission from atoms by drawing on the same basic concept.

The three-orders-of-magnitude improvement in time resolution results from several essential modifications of image-tube streak cameras: (i) the electric field of light is used for affecting the electrons' motion, the field (ii) is virtually jitter-free and (iii) applied along the direction of electron motion – implying their acceleration or deceleration instead of their deflection – (iv) directly at the location and instant of emission. Whereas (i) implies "only" a dramatically-enhanced streaking speed, the consequences of (ii)-(iv) are much more profound: (ii) allows to systematically vary the timing of the probing field with an accuracy within the electron bunch length and owing to (iii) this capability resuls in "projecting" the initial time-momentum distribution of electron emission into a series of different final (measurable) momentum distributions, while (iv) prevents the initial momentum spread from introducing any measurement error. Inspired by the physics of the first sub-femtosecond experiment[9], the basic concept for ATR metrology was put forward by Corkum and coworkers[13] and analyzed with a quantum theory by Brabec and coworkers[14]. Consider electron emission from atoms exposed to

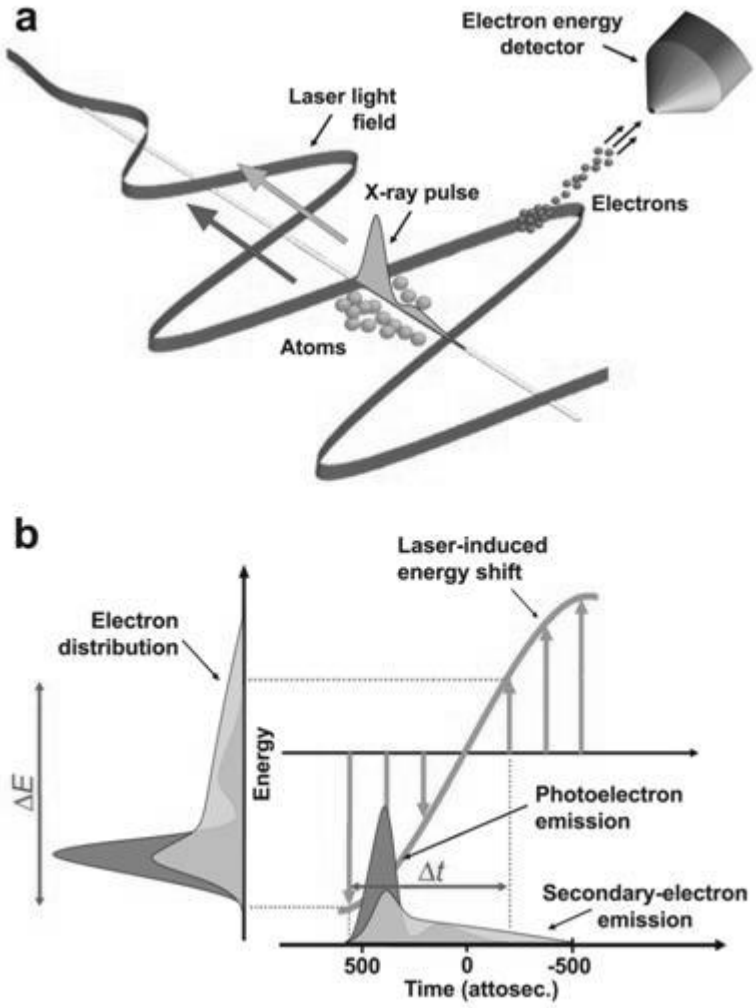

Figure 1 Light-field-controlled streak camera. The attosecond x-ray flash excites the atoms, which then emit electrons. The electrons ejected in the direction of the electric field of the laser light pulse simultaneously beamed in are detected. They undergo - depending on the time of their emission within half the oscillation period of the laser light - a change of velocity: in the case illustrated the electrons emitted first are decelerated, while those released on termination of the X-ray flash are accelerated. In this manner the successively emitted electrons are detected separately, but not spatially on a screen but alongside one another *on the energy scale*. The width ΔE and shape of the measured energy distribution (vertical axis in Fig. 1 b) 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, which opens the way to measurement in the attosecond region.

a sub-fs XUV burst in the presence of an intense, linearly-polarized, few-cycle laser field $E_L(t) = E_0(t)cos(\omega_L t + \varphi)$. The momentum of the freed electrons is changed by $\Delta p = eA_L(t)$ along the laser field vector. Here $A_L(t_r) = \int_{t_r}^{\infty} E_L(t)\,dt$ is the vector potential of the laser field, e and m stand for the charge and rest mass of the electron, respectively, and t_r is the release time of the

electron. This momentum transfer (arrows in Fig. 1b) maps the temporal emission profile into a similar distribution of final momenta $p_f = p_i + \Delta p$ within a time window of $T_0/2 = \pi/\omega_L$, if the electrons' initial momentum p_i is constant in time and their emission terminates within $T_0/2$. Under these conditions the temporal evolution of the electron emission (profiles on the horizontal axis in Fig. 1b) can be unambiguously retrieved from a single "streaked" momentum distribution (profiles on the vertical axis in Fig. 1b). However, any sweep of the electrons' initial momentum revokes the unique correspondence between the electron's final momentum and release time, preventing the retrieval of accurate temporal information from single streak records.

In general, the initial momentum spectrum of electrons detached from atoms by an impulsive excitation is varying in time during emission resulting in a time-momentum distribution $n_e(p, t)$ of electron emission rate. The final electron momentum spectrum, $\sigma(p) = \int_{-\infty}^{\infty} n_e(p,t)\,dt$, can be viewed as the projection of the time-dependent momentum distribution on the momentum space along lines of constant p. In the classical description of the freed electrons' motion in the strong laser field, the final spectra obtained in the presence of a strong laser field are generalized projections along lines where $p_f = p_i + eA_L(t)$ is constant. By delaying the laser field with respect to the the excitation that triggers electron emission, we obtain a set of tomographic records (briefly: streaked spectra),

$$\sigma_A(p) = \int_{-\infty}^{\infty} n_e(p - eA_L(t), t)\,dt, \qquad (1)$$

from which, with a suitable set of $A_L(t - \Delta t_n)$, the complete distribution $n_e(p, t)$ can be reconstructed. The method is closely related to frequency-resolved optical gating[16,17] with the oscillating field as a gate. In the simplest cases two streaked spectra (in addition to the field-free spectrum) may be sufficient. In the absence of a nonlinear momentum sweep the streak records obtained near the zero transitions of $A_L(t)$ with opposite slopes together with the field-free spectrum allow determination of all relevant characteristics: the temporal profile, duration and

monentum chirp of emission. Note that a linear momentum sweep leads to an asymmetric broadening of the streaked spectra at these delays and it is this asymmetry that makes the measurement highly sensitive to the momentum sweep, i.e. highly sensitive to deviations of the pulse duration from the Fourier-limit.

Experimental setup

We sample electron emission from atoms using the experimental setup that was described in Ref. 9 and employed recently for probing Auger electrons on a few-femtosecond time scale[18]. Now, the waveform-controlled few-cycle light provides a reproducible excitation burst for accurate triggering and a reproducible streaking field for capturing sub-fs electron emission from atoms. For excitation, XUV bursts are produced from Ne atoms ionized by intense, few-cycle waveform-controlled light pulses[2] in a process giving rise to high-order harmonics of the incident light for periodic (multi-cycle) pumping[19,20]. The x-ray pulses exiting the neon harmonic source co-propagate with the laser pulses down the beam delivery tube (Fig. 2). A zirconium foil mounted on a nitrocellulose membrane produces an annular laser beam with the x-ray beam in the center. The collinear XUV and laser beams are focused into a neon gas jet and delayed with respect to each other for the ATR measurements with a two-component Mo/Si broadband multilayer. A fine tuning of the cut-off of the X-rays can be carried out by fine tuning the energy of the fundamental laser beam. The reflecitivity band of the multilayer extends from 85 eV to 100 eV with a peak reflectance of ~ 30% and a full width of half maximum (FWHM) of ~ 9 eV.

In the first experimental demonstration of the ATR concept[15] two types of experiments were implemented. First, the internal part of the Mo/Si mirror was used to focus both the XUV and the laser beam to eliminate any external source of fluctuations in the timing between the excitation and probing pulses. In the second type of studies, the XUV and the laser beam were reflected by different sections of the focusing mirror: The central piece is sitting on a piezo stage adjustable in transverse and longitudinal direction. In this manner the two pulses can be

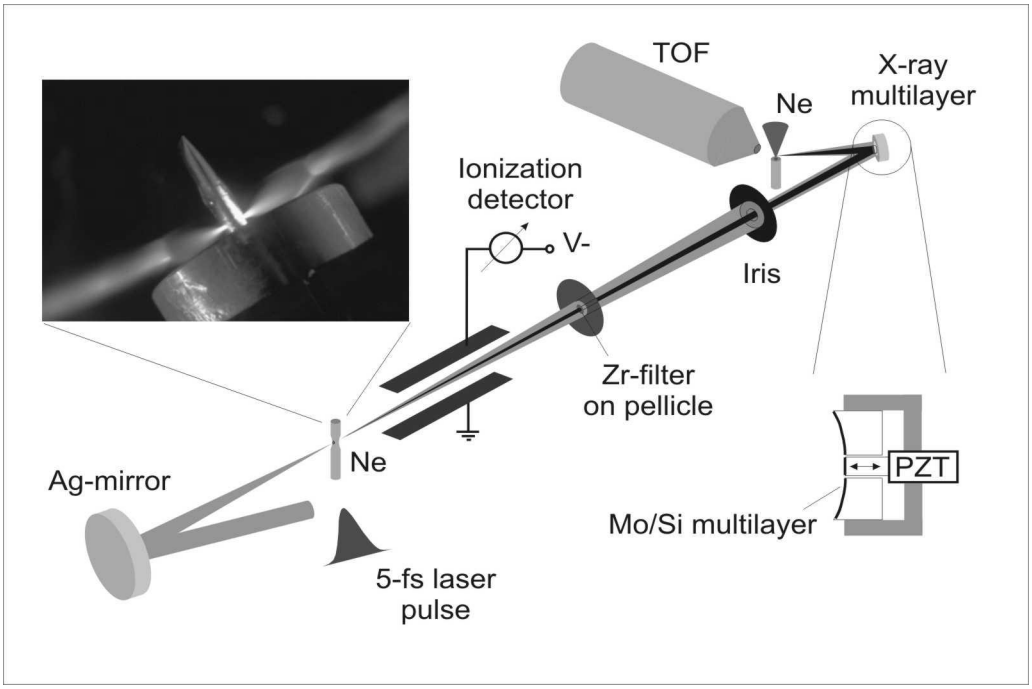

Figure 2 The schematic of the experiment. The focused 5-fs laser beam interacts with neon atoms to produce high-harmonic radiation. The laser and the highly-collimated x-ray beam co-propagate collinearly through a 2-m beamline towards the measurement. In the beamline the laser and harmonic beams pass through a 200-nm-thick, 3-mm-diameter zirconium foil placed on a 5-μm-thick nitrocellulose pellicle to cover a hole of 2-mm diameter. The energy transported by the resulting annular beam can be adjusted with a motorized iris between a fraction of a microjoule and a few tens of microjoules. The Mo/Si multilayer consists of an annular part having an outer diameter of 10 mm with a concentric hole of 3-mm diameter hosting a miniature mirror of slightly smaller diameter. Both parts originate from the same substrate, ensuring identical radii of curvature (R= -70 mm). The miniature central mirror is mounted on a piezo stage (PZT), allowing alignment and translation with respect to the external part.

overlapped spatially and temporally in the focal plane, exactly where the tip of a nozzle emitting the atoms to be photoionized is placed and the probing laser field could be delayed with respect to the XUV excitation pulse. Electrons ejected following the XUV excitation are detected with a time-of-flight spectrometer.

Isolation of a single sub-fs XUV excitation pulse

Fig. 3 summarizes representative results of a series of measurements of streaked XUV photoelectron spectra recorded with the XUV and laser pulse impinging with a fixed relativ timing set in the XUV generation process[15]. According to an intuitive one-active-electron model

of Schafer *et al.*[21,22], Corkum[23] and Lewenstein *et al.*[24,25] ionization of atoms by a linearly-polarized laser field is accompanied by the emission of extreme ultraviolet radiation due to recombination of the detached electron into its original ground state upon recollision with the parent ion. These theories along with a number of numerical simulations[25-29] predict the emission of the highest-energy (cutoff) photons to occur near the zero transition(s) of the electric field following the most intense half cycle(s) at the pulse peak in a few-cycle driver. In a cosine waveform ($\varphi = 0$) cutoff emission is expected to be confined to a single bunch emitted at the zero transition of $E_L(t)$ following the pulse peak. The photoelectrons knocked out in the direction of the peak electric field at this instant suffer maximum increase of their momentum and hence of their energy. The streaked spectrum in Fig. 3d, obtained with 5-fs,750-nm cosine pulses used for both XUV generation and subsequent electron streaking corroborates this prediction. The photoelectron spectrum peaking at $\hbar\omega_{xuv} - W_b \approx 72$ eV (where $\hbar\omega_{xuv} \approx 93.5$ eV is the center of the XUV spectrum selected by the Mo/Si mirror and $W_b = 21.5$ eV the binding energy of the most weakly bound valence electrons in Ne) in the absence of the laser field is upshifted by some 10 eV with only a few electrons scattered outside the shifted band. The clear upshift is consistent with the XUV burst coinciding with the zero transition of the laser electric field (see Fig. 3d). Possible satellites would appear at the adjacent zero transitions of $E_L(t)$ and suffer an energy downshift. The absence of a downshifted spectral peak of substantial intensity indicates a clean single sub-fs pulse generation. With the phase adjusted to yield a sine waveform ($\varphi = -\pi/2$) cutoff emission is predicted to come in twin pulses (Fig. 3c) [2]. The observation of the double-peaked streaked spectrum reflects clearly this time structure. Instead of being confined to a single burst the filtered high-energy XUV photons are now distributed in two bursts, each of which is less than half as intense as the isolated burst produced by the cosine waveform (Fig. 3d). These measurements demonstrate how control of the waveform of few-cycle laser light allows to control the temporal profile of XUV emission within the half wave cycle, i.e. on a sub-femtosecond time scale.

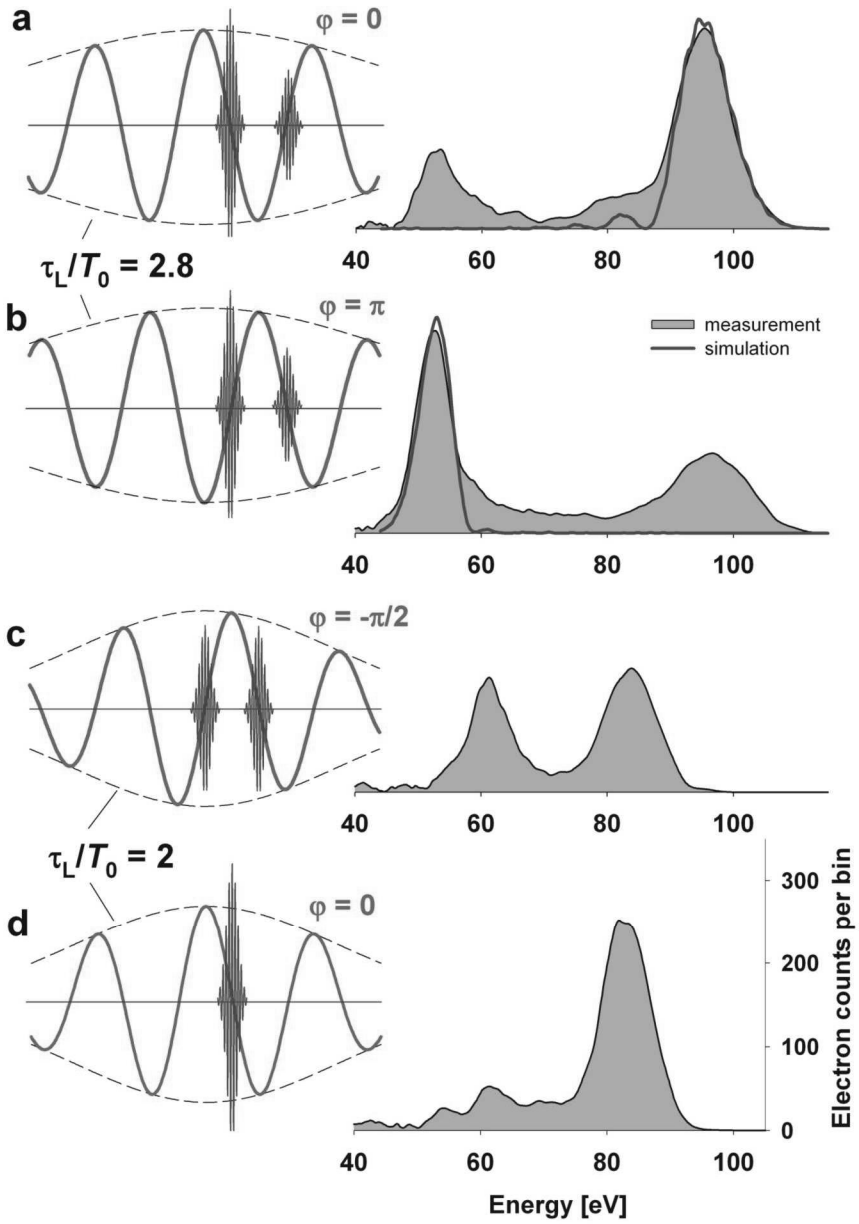

Figure 3 Streaked photo electron spectra recorded at a fixed delay of probing light. Energy distribution of photoelectrons emitted from neon atoms excited a sub-fs XUV pulse carried at a photon energy of ≈ 93 eV. The XUV bursts originate from the spectrally-filtered cutoff energy range of recombination emission from another target of neon atoms pumped by intense few-cycle 750-nm laser pulses; a), b) streaked spectra obtained with laser pulses of a normalized duration of $\tau_L / T_0 = 2.8$ having a cosine and –cosine waveform. The bold lines on the right-hand side depict calculated spectra obtained under the assumption of zero spectral phase, the calculations do not model the satellite pulse because the corresponding modulation of the (unperturbed) photoelectron spectrum is insufficiently resolved. However, the asymmetric broadening is well accounted for. c), d) streaked spectra obtained with laser pulses of a normalized duration of $\tau_L / T_0 = 2$ having a sine and cosine waveform, respectively.

For the generation of a single sub-fs XUV burst the normalized pulse duration τ_L/T_0 other than the carrier-envelope phase φ plays a crucial role. This is demonstrated by repeating the measurements with slightly longer, 7-fs laser pulses ($\tau_L/T_0 = 2.8$). The corresponding streaked spectrum obtained for $\varphi = 0$ (Fig. 3a) exhibits a sizeable downshifted spectral feature in addition to the upshifted main peak, indicating the appearance of a substantial satellite pulse ($\approx$ 30% of the principal burst) in the cosine-wave-driven recombination emission. A reduced difference in intensity of the adjacent half cycles of the few-cycle wave implies that the highest-energy spectral components of adjacent bursts reach the energy band selected by our 15%-bandwidth bandpass filter. Shifting φ by π results in the same emission but the streaking is reversed (Fig. 3b). In these measurements (Figs. 3a and b) we increased the strength of the streaking laser field by approximately a factor of two. The increased streaking reveals a pronounced asymmetry of the up- and down-shifted spectra, which is consistent with a near-quadratic temporal energy sweep of the XUV burst. This results from the asymmetric spectral intensity profile filtered by the Mo/Si mirror. The (near) quadratic momentum sweep of electron emission yields an asymmetric streaking at the adjacent maxima of $A_L(t)$ in a similar way as a linear chirp does at subsequent zero transitions of $A_L(t)$.

Measurement of the time-momentum distribution of atomic electron emission

With isolated sub-fs XUV pulses at our disposal atomic transients can now be triggered and their subsequent evolution be captured by probing electron emission with a synchronized wave of laser light. In the ATR measurements reported so far[15] the objects of scrutiny were photo electrons. The streaking field in these experiments was produced by blocking the internal part of the laser beam with a zirconium filter (transmitting the XUV pulse) and focusing the transmitted annular beam on the target with the external section of the Mo/Si mirror[9], which can be delayed with respect to the internal section that focuses the XUV beam. Fig. 4 shows a series

of streaked spectra of photoelectrons emitted from neon as a function of Δt. The excitation pulse at 93 eV was produced by a cosine waveform with $\tau_L/T_0 = 2$.

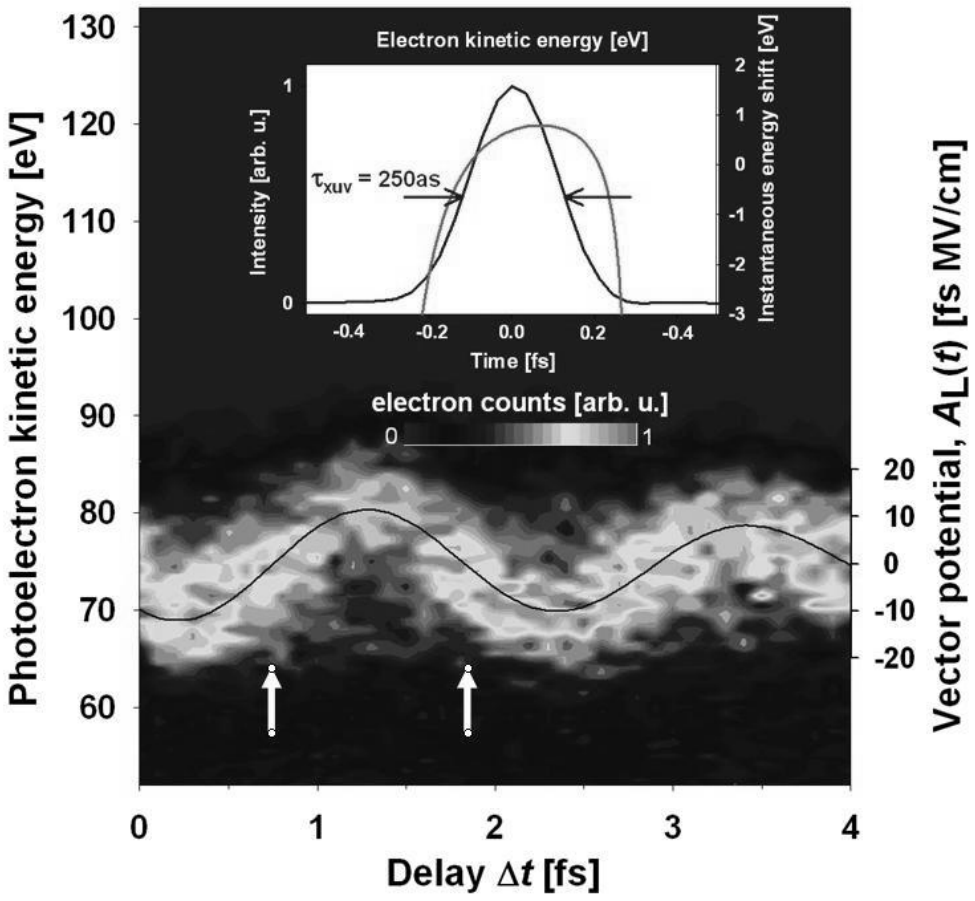

Figure 4 ATR measurement: a series of tomographic projections (streaked kinetic energy spectra) of the initial time-momentum distribution of photoelectrons knocked out by a single sub-fs XUV pulse (in false-colour representation). A few-cycle laser pulse with a cosine waveform and a normalized duration of $\tau_L/T_0 = 2$ was used for both generating the single sub-fs excitation pulse and for probing photo electron emission in the atomic transient recorder. Black line: $A_L(t)$ of the probing field evaluated from the peak shift of the streaked spectra (see scale on the right hand side). Inset: Temporal intensity profile and energy sweep of the sub-femtosecond XUV excitation pulse evaluated from the ATR measurements. Basis of the calculation was the unperturbed and two streaked spectra at the zero-transitions of the laser electric vector potential marked by the two white arrows.

If the electrons are emitted with an initial kinetic energy much larger than their average quiver energy in the laser field, and temporally confined to a fraction of the half oscillation cycle, theory predicts that their energy shift is linearly proportional to Δp and hence to the vector potential at the instant of release of the wavepacket, $\Delta W(t_r) \approx (p_i/m)\Delta p(t_r) = (ep_i/m)A(t_r)$, where p_i

is the initial momentum of the electron. As a consequence, from the peak shifts of the spectra – without having to analyze their detailed structure – $A_L(t)$ and hence $E_L(t)$ can be accurately determined. The result is shown by the black line in Fig. 4, constituting the first direct (time-resolved) measurement of a visible light field. From this measurement we can also evaluate the half oscillation period of the electromagnetic field as $T_0/2 \approx 1$ fs. This latter value indicates a significant blue shift to a carrier wavelength of 600 nm near the pulse peak (origin: ionization-induced self-phase modulation in the XUV source), in agreement with previous observations[9].

With the streaking field $A_L(t)$ known, full temporal (time-momentum) characterization of sub-femtosecond electron emission is now becoming feasible. This capability strictly relies on light waveform control: In its absence the streak records smear out beyond redemption due to irreproducible excitation and probing. From a few selected streaked spectra the duration, the chirp and the timing jitter of the XUV burst can be determined. From this analysis it was found that XUV pulse is near Fourier-limited with a duration (full width at half maximum of the intensity envelope) of τ_{xuv} = 250 (-5/+30) as and a timing jitter (with respect to the laser field oscillations) of τ_{jitter} = 260 (± 80) as. The remarkable accuracy of τ_{xuv} relies on using several (in this case 3) tomographic projections of the time-momentum distribution of photoelectrons for the XUV pulse retrieval, which is the essence of the ATR concept. Restricting our analysis to only one of the two streaked spectra – in the spirit of streak camera measurements – would only allow setting an upper limit of 500 as on the XUV pulse duration**.** These measurements provide evidence for the emergence of isolated, bandwidth-limited XUV bursts over a relative spectral band as broad as 15% (15 eV at 100 eV) from recombination emission driven by a cosine waveform with $\tau_L/T_0 = 2$. This has important implications for scaling the technology.

The experiments reported so far[15] indicate that there is room for further improvement. The absence of spectral phase modulation in the cutoff energy range of few-cycle driven recombination emission over an energy band as broad as 15% and the confinement of this

emission to a single burst holds promise for the generation of single 50-attosecond pulses from the same source upscaled to 500 eV photon energies[30]. Extension of the presented experiments to exploring the sub-femtosecond temporal variation of emission intensity and momentum distribution of primary (photo) *and* secondary (Auger) electron emission simultaneously will provide unprecedented insight into the excitation and relaxation dynamics of the electronic shell of atoms and molecules. The currently used time window of $T_0/2 \approx 1$ fs can be extended to several 10 fs by difference frequency generation with the few-cycle laser pulse whilst keeping the resolution of the ATR in the sub-fs regime. At near keV excitation energies, the atomic transient recorder presented here will allow time-domain metrology of atomic dynamics with a resolution approaching the atomic unit of time (24 as).

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30. Terawatt-scale few-cycle pulses recently produced intense recombination emission at 500 eV in the authors' laboratory.

Sponsored by the Fonds zur Förderung der wissenschaftlichen Forschung (Austria, grants Z63 and F016), the Deutsche Forschungsgemeischaft (Germany, grants SPP1053, HE1049/9, and KL1077/1) and by the European Union's Human Potential Programme under contract HPRN-2000-00133 (Atto). R.K. acknowledges an APART fellowship of the Austrian Academy of Sciences.