High-flux ultrafast extreme-ultraviolet photoemission spectroscopy at 18.4 MHz pulse repetition rate

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Laser-dressed photoelectron spectroscopy, employing extreme-ultraviolet attosecond pulses obtained by femtosecond-laser-driven high-order harmonic generation, grants access to atomic-scale electron dynamics. Limited by space charge effects determining the admissible number of photoelectrons ejected during each laser pulse, multidimensional (i.e. spatially or angle-resolved) attosecond photoelectron spectroscopy of solids and nanostructures requires high-photon-energy, broadband high harmonic sources operating at high repetition rates. Here, we present a high-conversion-efficiency, 18.4-MHz-repetition-rate cavity-enhanced high harmonic source emitting $5 \times 10^5$ photons per pulse in the 25-to-60-eV range, releasing $1 \times 10^{10}$ photoelectrons per second from a 10-µm-diameter spot on tungsten, at space charge distortions of only a few tens of meV. Broadband, time-of-flight photoelectron detection with nearly 100% temporal duty cycle evidences a count rate improvement between two and three orders of magnitude over state-of-the-art attosecond photoelectron spectroscopy experiments under identical space charge conditions. The measurement time reduction and the photon energy scalability render this technology viable for next-generation, high-repetition-rate, multidimensional attosecond metrology.
At the beginning of this century, a series of seminal technological developments in the field of ultrafast lasers (reviewed, e.g., in ref. 1), led to the first photoelectron spectroscopy42 (PES) experiments performed with extreme-ultraviolet (XUV) radiation obtained by high-order-harmonic generation (HHG) driven by intense, visible/near-infrared (VIS/NIR) pulses in gases, in the presence of the (delayed) driving field43,44. In these experiments, the sub-optical-cycle temporal structure of HHG radiation, and its synchrony with the driving field providing a temporal reference, enabled laser-dressed XUV-PES with sub-femtosecond temporal resolution, marking the birth of attosecond metrology43. The two major PES-based tools employed in this emerging field, attosecond streaking4,9-12, using isolated attosecond pulses, and RABBITT (reconstruction of attosecond harmonic beating by interference of two-photon transitions)-based techniques,45,46 using attosecond pulse trains,49,15, rely on analysing the evolution of photoelectron spectra with varying delay between the two optical fields. Both approaches have been widely applied for measurements of attosecond photoemission delays from gases, providing insights into electron dynamics deep inside the atom (see e.g., refs. 1,7,13,14,16 and references therein). During the past decade, considerable efforts have addressed the extension of these techniques to solids9,12–14 and nanostructures17,18. For instance, very recently the combination of angle-resolved PES (ARPES) and RABBITT has enabled first hand-specific measurements of correlated electron dynamics on metal surfaces9,10. Another example is the ongoing effort to combine attosecond streaking with photoemission electron microscopy (PEEM) for the spatiotemporal study of complex plasmonic fields propagating on nanostructured metal surfaces17-21. Fundamental studies of correlated electron dynamics in these dense atomic systems are necessary for the development of novel materials and high technologies.

A severe shortcoming of laser systems employed in state-of-the-art attosecond-PES experiments, however, arises from their relatively low pulse repetition rate rep, usually well below 1 MHz, in the context of space charge22,23 (SC). Coulomb interaction of multiple photoelectrons released from the sample during a single pulse affects their velocities and trajectories towards the detector, leading to distortions of the observables, i.e., kinetic energy (and momentum or position)17,22,23. In fact, in the vast majority of setups for (multidimensional—i.e., energy, and angularly or spatially resolved) attosecond-PES on solids, space charge effects demand a significant attenuation of the HHG output9,17. At a space-charge-limited photoelectron flux and at typical rep values in the kHz range, the acquisition of (multidimensional) photoelectron spectra with sufficient signal-to-noise ratio necessitates impractically (or even prohibitively) long acquisition times of several hours10 to several tens of hours17. Over such periods, laser instabilities and sample contamination24 constitute severe technological challenges. This shortcoming can potentially be circumvented by HHG sources with repetition rates in the multi-MHz range. Fig. 1a shows an overview of state-of-the-art HHG sources illustrating that to date, however, the combination of high photon energies, high photon flux and high repetition rate has remained an unmet challenge. Although large-scale technologies such as FELs also offer high flux at high photon energies and repetition rates22 to date, they do not enable measurements with attosecond time resolution.

Here, we close this gap with a high-conversion-efficiency cavity-enhanced HHG source. Driving HHG at rep = 18.4 MHz with sub-40-fs, 1-μm pulses in argon results in XUV pulse trains with photon energies between 25 and 60 eV and, in particular, a record flux at >40 eV for high rep (Fig. 1a). In experiments demonstrating the potential of this source for time-resolved PES, a 10-μm-diameter spot on a tungsten surface was illuminated, generating between two and three orders of magnitude more photoelectrons than affordable in previous state-of-the-art, high-photon-energy attosecond-PES experiments under the same space-charge-imposed restrictions (Fig. 1b). Time-of-flight (ToF) detection allows for the simultaneous collection of photoelectrons from a large solid angle and in a broad range of kinetic energies, with a temporal detection duty cycle close to 100% (i.e., the time of flight dispersion corresponds to the pulse repetition period). Unlike other detection schemes (e.g., hemispherical or retarding-field analyzers), ToF spectrometers allow for the simultaneous detection of electron emission momentum (or spatial distribution) and kinetic energy without the need to scan over one of these parameters at the expense of a reduced flux. Furthermore, attosecond temporal resolution is inferred from the sideband modulation of laser-assisted photoemission46.

Results

Cavity-enhanced HHG. Modern MHz-repetition-rate ytterbium-based lasers (see refs. 26,27 and references therein) currently hold average power records for single-pass HHG with photon energies up to around 40 eV27–29, obtained from krypton and xenon (Fig. 1a), and have been successfully employed for narrowband PES measurements at these photon energies, both in single-pass26 and in cavity-enhanced HHG configuration30,31 (Fig. 1b).

However, with these technologies significantly higher photon energies (for which gases with a higher ionisation potential like argon or neon are typically employed) were so far only achieved at the cost of the repetition rate in single-pass HHG32,33 or of the photon flux in cavity-enhanced HHG34,35, see Fig. 1a. Here, we demonstrate that a combination of state-of-the-art fibre laser technology, nonlinear pulse compression and cavity-enhanced HHG can overcome these limitations.

The experimental setup is depicted in Fig. 2. The HHG source (see also Methods) consists of a master-oscillator-power-amplifier (MOPA) femtosecond frontend followed by a multi-pass bulk-compression stage and a femtosecond enhancement cavity (EC) housing the HHG gas target. The initial pulses were delivered by a titanium-sapphire oscillator with a repetition rate of 73.6 MHz. By means of a pulse picker after the oscillator, the repetition rate of the MOPA output36 was reduced to a quarter to permit sufficient energy dispersion in ToF-PES. The output of the MOPA was an 18.4-MHz train of 5.4-μJ, 250-fs pulses, spectrally centred at 1030 nm. Multi-pass nonlinear spectral broadening in bulk and subsequent chirp removal compensation with dispersive mirrors allowed for the temporal compression of these pulses down to sub-40-fs duration37 at an energy level up to one order of magnitude higher than in previous cavity-enhanced HHG setups with comparable pulse durations (see refs. 34,35 and references therein). For magnitude of phase distortion increases in HHG with stronger nonlinear interactions. When using a feed-forward phase-stabilised Ti:Sapphire oscillator to seed the Yb-fibre amplifier37 (see Fig. 3b) and actively controlling the repetition frequency of the seed oscillator and the EC length with a fast and a slow loop, respectively. High harmonics of the circulating NIR radiation were generated in an argon gas target at the focus, and coupled out through a 340-μm-diameter on-axis opening in the resonator mirror following the focus34,35. Fig. 3c, d show the autocorrelation and the spectra of the pulses impinging and circulating in the EC, respectively.

Argon gas was delivered through an end-fire quartz nozzle with a 200-μm-diameter opening. The gas target position and the backing pressure were optimised for highest XUV yield around

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Flux of state-of-the-art high-harmonic generation sources and PES experiments. a Generated XUV photons per pulse versus pulse repetition rate for different energy ranges (colour-coded, see legend) in state-of-the-art HHG sources (representative selection). The shape of the symbols (top of the figure) indicates the underlying femtosecond laser technology. Full and empty symbols indicate HHG sources which have and which have not been used for PES, respectively. The diagonal lines represent lines of constant XUV flux at a given energy: 30 eV (orange) and 50 eV (green). Data from refs. 17, 26–35, 45, 46, 51, 62–65, 68–71. b Photoelectron flux and estimated space-charge-induced kinetic energy distortion \( \Delta E_{\text{ESC}} \) (i.e., spectral broadening and energy shift of the same order, see Methods) in state-of-the-art ultrafast PES experiments versus pulse repetition rate. The colours of the symbols indicate different photon energy ranges (see legend). For each setup, the spot diameter employed in the experiment is given, where available. For comparison, the energy distortion (heat map) is calculated relating the number of released photoelectrons to a 50-µm-diameter spot (typical size). For photon energies higher than 40 eV (typical for attosecond-PES), a space-charge-limited spectral resolution of 0.1 eV corresponds to a number of \( \sim 10^4 \) photoelectrons released from the sample for each XUV pulse, resulting in an admissible number of \( \sim 1.5 \times 10^5 \) impinging photons (see Methods). Data from refs. 17, 26, 30, 31, 45, 46, 70. Note, that the photoelectron flux in refs. 26, 30, 31, 46, 70 is generated by single-harmonic XUV excitation.

Fig. 2 Experimental setup. The MOPA generates a train of 18.4-MHz-repetition-rate, 250-fs pulses centred at 1030 nm. The output pulses are nonlinearly compressed to <40 fs via spectral broadening upon multiple-pass self-phase modulation in fused silica (FS) followed by a chirped-mirror (CM) compressor, and coherently enhanced in a femtosecond enhancement cavity. HHG is driven in a 25 × 32-µm²-radius focus between two curved mirrors, one of which is pierced for on-axis XUV output coupling. IC: input coupler, PM: pierced mirror. Intracavity pulse diagnostics: autocorrelator (AC), optical spectrum analyser and power meter. The output-coupled XUV can either be sent to an XUV spectrometer or to the PES experiment. A 96%-NIR-transmission beam splitter (BS) can be used to separate the NIR radiation from the co-propagating XUV beam. Alternatively, the BS can be exchanged with an Au mirror, resulting in \( \sim 10^5 \) W/cm² NIR intensity on the sample for laser-dressed PES. In the ultra-high-vacuum PES chamber (<10⁻⁹ mbar) the XUV beam is focused onto the tungsten (110) crystal with a 250-mm radius-of-curvature multilayer (scandium-silicon) double mirror. The inner segment of the mirror can be delayed with a piezoelectric-ceramic stage. A moveable on-axis aluminium beam is focused onto the tungsten (110) crystal with a 250-mm radius-of-curvature multilayer (scandium-silicon) double mirror. The inner segment of the inner mirror segment. Fine-tuning the NIR power is achieved with a variable aperture. The electrons are detected by a time-of-flight spectrometer equipped with a retarding grid. Two operation modes are available, the drift mode without any manipulation of the electron trajectory and the low-angular-dispersion mode with capture in a larger solid angle (see Methods).

40 eV after output coupling. An output coupled XUV flux of \( \sim 1 \times 10^{13} \) photons/second was measured, with energies ranging from 25 to 60 eV (Fig. 4). By combining state-of-the-art fibre laser technology with a novel nonlinear pulse compression scheme, a pulse energy of 4.5 µJ was available as a seed to the enhancement cavity, leading to 0.15-mJ intracavity pulses. Importantly, the moderate cavity finesse alleviates ionisation-related intensity limitations (clamping) that arise from the blueshift caused by self-phase-modulation in the gas target38–40. Together with relatively weak cumulative ionisation effects in the gas target (each atom traverses the beam diameter within 4 laser shots), this enabled an intracavity conversion efficiency around 10 times
higher than in 34,35 and, for the first time, approaching that obtained with single-pass HHG in argon (see Methods). This allowed for a highly efficient use of the EC concept, in particular leading to – to our knowledge – the highest usable XUV flux for photon energies beyond 40 eV at $f_{\text{rep}} > 1$ MHz reported so far (see Fig. 1).

**Ultrafast photoelectron spectroscopy.** For PES experiments, the XUV beam and, optionally, the NIR beam, were directed through a differentially pumped beamline and focused to a 10-μm-diameter spot onto the surface of a single-crystalline (110) tungsten target with a multilayer XUV mirror (Fig. 2). Photoelectrons were collected with a ToF spectrometer (equipped with a retarding grid) with two settings—drift mode (DM) and large-angular-dispersion mode (LAD)—differing in their capture angle and energy resolution (see Fig. 5a and Methods).

A first PES experiment was aimed at measuring the number of photoelectrons released from the sample and scrutinising the space-charge-induced spectral distortions. To this end, the XUV beam was separated from the 6-W average power NIR beam passing through the opening in the output-coupling mirror with a beam splitter. Thus, all of the detected photoelectrons stem from single-photon XUV photoemission. Although the use of XUV transmission optics was avoided, the reflective optics attenuated the XUV photon flux by 97%, resulting into $3 \times 10^{11}$ photons/second at the sample, releasing an estimate of around $1 \times 10^{10}$ photoelectrons/second. This value is in good agreement with that calculated from the ToF count rate, and results in an estimated space charge distortion of $\Delta E_{\text{SC}} = 50$ meV (see Methods). In accordance with the instrument resolution of 0.13–0.36 eV (limited by the spectral width of the harmonics, see Methods), no distortions of the spectra were discernible for both acquisition modes (see Fig. 5b, c).

To evaluate the statistics of the PES measurements we calculated the relative standard deviation $\sigma$ of several measurements, for varying integration time $T$ of each measurement. The results are shown in Fig. 6a. A statistical behaviour (decrease of $\sigma$ with $\sqrt{T}$) was observed for $T < 160$ s, resulting in $\sigma < 1\%$. Long-term drifts led to an increase of $\sigma$ to $\sim 2.5\%$ after $T = 15$ min.

To demonstrate the suitability of the HHG source for attosecond-resolution PES, we performed XUV-pump-NIR-probe measurements. The XUV-NIR beam splitter was replaced by a gold mirror, enabling NIR intensities in the $10^{11}$-W/cm² range at the target. For introducing a mutual delay, the XUV and the NIR beams leaving the EC were spatially separated by a coaxial optic featuring a small 300-nm-thick aluminum filter covering the centre of the beam, held in position by thin metal wires. The outer part is transparent for the NIR pulses, whereas the central part is only transparent for the XUV. A variable temporal delay was introduced by a two-segment spherical mirror (Fig. 2). The fixed outer gold-coated ring reflects the NIR radiation and a movable, 5-mm-diameter multilayer-coated inner segment reflects the time-delayed XUV pulse trains with a peak reflectivity of 14% at 44 eV and in a bandwidth of 10 eV. This
common-path geometry renders the XUV-NIR interferometer inherently stable. Fig. 6b shows the intensity of the sideband peak at 37.9 eV as a function of the pump-probe delay. Its delay-dependent oscillation at twice the fundamental laser frequency is caused by the interference of two different two-photon-transitions. This sinusoidal curve contains information on the relative time delay between the NIR and XUV fields, on the spectral phase of the adjacent harmonics, as well as on the delay of the photoemission process itself and, thus, on the underlying electron dynamics in the sample. Using the RABBITT-technique, this information can be extracted from the phases of these sideband modulations. In order to determine an upper bound of the time resolution of our instrument, a sine curve with three free parameters (amplitude offset and phase) at a fixed wavelength of 1030 nm was fitted to the data. The resulting fit error of the phase indicates a timing precision of the interferometer of 36 as, which is comparable to state-of-the-art attosecond-PES experiments. Notably, the...
common-path XUV-NIR interferometer does not necessitate active stabilisation\textsuperscript{10,11} of the delay line. In the photoelectron spectra at a kinetic energy of 12 eV, a distinct peak that we attribute to the tungsten 4f doublet state is visible (Fig. 7). Its energy separation from the highest valence band peak corresponds well to the literature value of the binding energy of the tungsten 4f\textsubscript{7/2} state of 31.4 eV\textsuperscript{42}. Since it is difficult to discern peaks below 12 eV due to the background of secondary electrons, we generated significantly higher harmonics in neon and used a multilayer mirror centred at 65 eV to spectrally filter them without changing anything within the cavity but the gas, its pressure and the position of the gas nozzle. As it can be seen in Fig. 7, the higher photon energies of neon HHG allow to clearly observe several photoelectron peaks originating from the 4f-state. Due to spin-orbit coupling, the 4f-state is split into two peaks separated by 2.2 eV which differs by only 0.2 eV from the...
separation of 2.4 eV between two high harmonics. Consequently, a photoelectron peak from the deeper bound 4f 5/2 state excited by photons of a specific high harmonic would be separated by only 0.2 eV from the photoelectron peak resulting from the 4f 7/2 state and photons from the next lower harmonic. Although this can hardly be distinguished by eye in this particular measurement, techniques to separate such features reliably in time-resolved measurements (e.g., in the Fourier space) have recently been developed and applied successfully.

The different appearance of the valence band electrons of neon and argon high harmonics can be explained by the better separation from the secondary electron background in combination with the narrower bandwidth of the XUV mirror.

Discussion

We have presented a HHG source affording a combination of high repetition rate ($f_{\text{rep}} = 18.4$ MHz), high photon energies (up to 60 eV) and high photon flux ($5 \times 10^3$ photons per pulse). Proof-of-principle PES measurements on tungsten evidence the advantages of this parameter regime for PES-based attosecond metrology, over state-of-the-art HHG sources employed in this field. In particular, we demonstrated an improvement of the photoelectron flux by more than two orders of magnitude in space-charge-limited attosecond-PES. Considering the relatively strong attenuation of the XUV flux between the HHG source and the sample (almost two orders of magnitude), in principle an even higher photoelectron flux could be achieved with this HHG source. In addition, PES statistics attest the stability of this source for a comparatively very large number of laser shots. For measurement times of 160 s and 15 min, photoelectron spectra with standard deviations of less than 1% and ~2.5%, respectively, were taken. At our repetition rate, these two durations correspond to $2.9 \times 10^6$ and $1.7 \times 10^{10}$ shots, respectively. For comparison, at a repetition rate of 10 kHz, typical for state-of-the-art attosecond-PES experiments, this would correspond to 3.4 and 19 days of continuous measurement, respectively. A particularly important aspect of the decrease in measurement time is the fact that this large number of shots can be recorded over a duration shorter than the typical sample contamination time under ultra-high vacuum conditions. Typically, at a pressure of $10^{-10}$ mbar, a monolayer of residual gas particles builds up in around 2 h. This requires periodic cleaning, which considerably increases the overall measurement time and experiment complexity.

Furthermore, the measurement time reduction is expected to lead to a corresponding improvement of the temporal resolution in attosecond-PES experiments. In particular, the measurement of the sinusoidal oscillation of the laser-assisted XUV-photoemission sideband shown in Fig. 6b was done within 105 s which lies well within the time that the system behaves statistically (Fig. 5a), implying that multidimensional PES measurements can be repeated multiple times in the absence of drifts. The high flux and repetition rate of our system could even enable delay scans within less than a millisecond, which would decrease the impairment of the time resolution by acoustic vibrations. As was recently demonstrated, averaging the results of a large number of measurements can enable the determination of photoionisation delays with absolute errors in the sub-attosecond range.

The current configuration of our apparatus is particularly suited for RABBITT-based attosecond metrology. For measurements of photoionisation delays, these techniques exhibit a few distinct advantages. For instance, they combine high temporal resolution, given by the individual attosecond bursts in the XUV pulse train, with high spectral resolution afforded by the discrete nature of the harmonics, and they offer high signal-to-noise ratios due to the evaluation of sideband modulations rather than shifts of broad photoelectron spectra. In particular in combination with ARPES, very recently broadband RABBITT measurements have led to a number of insights into band-specific multi-electron attosecond correlation dynamics in solids.

In contrast to previous multi-MHz HHG sources, our apparatus generates photons with energies of several tens of eV at a significantly higher flux. In addition, due to geometrical XUV output coupling, the architecture is photon-energy scalable to 100 eV or more. Besides the general advantages of rendering PES less sensitive to external fields and of affording a better temporal resolution and a better energy separation of primary from secondary electrons, high photon energies are crucial for accessing deeply bound states of the system under study. This enables the comparison of photoemission delays from different initial states for self-referencing, and is desirable for experiments on X-ray magnetic circular dichroism at the M-edge of nickel or cobalt. We demonstrated that the high photon energies of our setup enable addressing such a deeply bound state in tungsten, as it can be seen in Fig. 7. This may allow one to compare photoemission delays between the valence band and a deeper bound state in a solid material utilizing the RABBITT-technique.

The femtosecond-laser technologies presented here can be extended both to longer and to shorter pulses. Driving HHG with longer intracavity pulses is a way towards higher spectral resolution for RABBITT-based measurements. Shorter pulses, together with cavity-enhanced gating methods offer the prospect of the generation of temporally isolated attosecond pulses at multi-MHz repetition rates. Such pulses could be used, for example, in multidimensional attosecond-streaking-based measurements, such as attosecond photoelectron emission microscopy of nanoplasmonic fields, beyond attosecond metrology the high-power multi-MHz-repetition-rate HHG source presented here could benefit applications such as XUV frequency comb spectroscopy for precision-measurements of electronic energy levels of atoms, or for referencing to nuclear transitions for future nuclear clocks.

Methods

Estimation of space charge effects

The effect of space charge on the photoelectron spectra is estimated by using the Long-Itchkawiz-Käbber (LIK) model. The model is based on the assumption of a radially expanding spherical, initially monoenergetic charge distribution and yields an energy broadening $\Delta E_{\text{ESC}}$ of the spectrum given by $\Delta E_{\text{ESC}}[\text{eV}] = 3 \times 10^{-3} N_e/(0.5 d_{\text{focus}}[\mu \text{m}])^{22,24}$, where $N_e$ denotes the number of emitted electrons per pulse and $d_{\text{focus}}$ is the diameter of the emission spot. The validity of the model has been confirmed by classical many-body calculations. It has been shown to also be applicable to photoelectrons uniformly distributed in energy by changing the factor on the right-hand side of the above equation to $0.5 \times 10^{-3} (\pm 0.12 \times 10^{-3})$ for electrons on the high-energy side of the spectrum. Furthermore, the simulations showed that the broadening is usually accompanied by an energy shift of roughly the same magnitude. We used this modified relation to estimate the space-charge-caused broadening. To relate the number of XUV photons to the number of emitted electrons, and to calculate the expected space charge effect for the various experiments shown in Fig. 1b (for a fixed spot size), the total photoemission probability including scattered and secondary electrons is needed. In the case of our experiment, we determined a photoemission probability of 3.3% from the measurement of the number of emitted electrons per pulse and taking into account the attenuation of ~97% of the output coupled XUV radiation along the beam path to the tungsten surface (the attenuations due to one Nb$_2$O$_5$ beam splitter, one gold mirror and a scandium-silicon XUV multilayer mirror amount to 50%, 40 and 90%, respectively). This value is in satisfactory agreement with the quantum efficiency of tungsten photoelectron rates experimentally determined previously to decrease from roughly 10% at 25 eV to about 6% at 50 eV. The discrepancy can be attributed to the uncertainty of the attenuation estimation (including geometrical losses at the
5-mm-diameter XUV mirror, angle uncertainties, surface roughness etc.). For typical PES experiments, an energy resolution on the order of 100 meV is sufficient to distinguish different partial states in the electron solid, which justifies the choice of the admissible space-charge-induced energy broadening in the discussion. Furthermore, we assumed a typical focal spot diameter of 50 μm for the comparison, while in our experiment, to scrutinise the presence of space charge effects, we utilized a smaller spot size of 10 μm in order to increase the sensitivity to these effects. In atto- and picosecond-PES, in order to attain homogeneous conditions for all emitted electrons, the focal spot of the XUV focus is restricted to be smaller than the spot size of the pump beam. The latter, in

...for the output coupling efficiency (Fig. 4) was estimated with the 3+1 D HHG model described in ref. 68 by varying the target gas density and selecting the value with optimum flux around 40 eV, and assuming a gas target position of one Rayleigh range before the focus. This target position was experimentally determined by scanning the nozzle position and observing the clamping of the intracavity power due to plasma-induced blueshift.

**Time-of-flight photoelectron detection.** Three different principles are common in electron spectroscopy: retardation of the electrons by a potential barrier (retarding field analyser), dispersion of electrons in a magnetic (or in some cases an electric) field (e.g., hemispherical analysers) and time-of-flight (ToF) analysis. The first two options necessitate a scan over the energy range that is to be analysed, whereas ToF detection allows for the simultaneous detection of electrons of all accepted kinetic energies. More importantly, when combined with a spatially resolving detector, only ToF spectrometers enable the detection of 3D data at once: electron kinetic energy and two dimensions either in space (PEEM) or angle (ARPES). ToF spectrometers are ideal in combination with HHG sources due to the pulsed nature of the latter. The maximum admissible temporal dispersion of electrons is limited by the time between two subsequent laser pulses, which is 54 ns at our repetition rate. Secondary electrons with times of flight far above this value (for any reasonable drift distance) are filtered out by an additional retarding grid as a high-pass filter for electrons. For our drift distance of 880.5 mm (Themis 1000, SPECTS GmbH) this typically results in a detectable window of 3.3–50 eV electron kinetic energy with a resolution of around 100 eV (determined from the measured temporal resolution of our 2D-delayline-detector). This energy range can be increased by accelerating electrons before or within the spectrometer at the expense of energy resolution. Without applying any additional field, our spectrometer collects a solid angle of 0.0016 sr in its so-called drift mode (DM). In its low angular dispersion mode (LAD), a complex multi-element electronic lens system within the spectrometer is used to image a larger solid angle of 0.047 sr on the detector. Since the electron lens system inevitably suffers from chromatic aberration, only a limited energy window can be correctly imaged, and a loss of energy resolution cannot be prevented. Although the earth magnetic field is compensated for, there remains a residual field of around 2 μT using Helmholtz-coils, this also can have a negative impact on the resolution. We experimentally determined the energy resolution in LAD mode for our settings to be ~1 eV (cf. Fig. 6b). Our instrument resolution can be calculated as the convolution of the detector resolution (30 meV in DM) and the width of the harmonics in electron spectroscopy: retardation of the electrons by a potential barrier (retarding field analyser), dispersion of electrons in a magnetic (or in some cases an electric) field (e.g., hemispherical analysers) and time-of-flight (ToF) analysis. The first two options necessitate a scan over the energy range that is to be analysed, whereas ToF detection allows for the simultaneous detection of electrons of all accepted kinetic energies. More importantly, when combined with a spatially resolving detector, only ToF spectrometers enable the detection of 3D data at once: electron kinetic energy and two dimensions either in space (PEEM) or angle (ARPES). ToF spectrometers are ideal in combination with HHG sources due to the pulsed nature of the latter. The maximum admissible temporal dispersion of electrons is limited by the time between two subsequent laser pulses, which is 54 ns at our repetition rate. Secondary electrons with times of flight far above this value (for any reasonable drift distance) are filtered out by an additional retarding grid as a high-pass filter for electrons. For our drift distance of 880.5 mm (Themis 1000, SPECTS GmbH) this typically results in a detectable window of 3.3–50 eV electron kinetic energy with a resolution of around 100 eV (determined from the measured temporal resolution of our 2D-delayline-detector). This energy range can be increased by accelerating electrons before or within the spectrometer at the expense of energy resolution. Without applying any additional field, our spectrometer collects a solid angle of 0.0016 sr in its so-called drift mode (DM). In its low angular dispersion mode (LAD), a complex multi-element electronic lens system within the spectrometer is used to image a larger solid angle of 0.047 sr on the detector. Since the electron lens system inevitably suffers from chromatic aberration, only a limited energy window can be correctly imaged, and a loss of energy resolution cannot be prevented. Although the earth magnetic field is compensated for, there remains a residual field of around 2 μT using Helmholtz-coils, this also can have a negative impact on the resolution. We experimentally determined the energy resolution in LAD mode for our settings to be ~1 eV (cf. Fig. 6b). Our instrument resolution can be calculated as the convolution of the detector resolution (30 meV in DM) and the width of the harmonics

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**Data availability**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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