Attosecond angular streaking

Claudio Cirelli[1], Petrissa Eckle[1], Mathias Smolarski[2], Adrian Pfeiffer[1], Philip Schlup[1], Jens Biegert[1], André Staudte[2], Markus Schöffler[2], Harm G. Muller[3], Reinhard Dörner[2], Ursula Keller[1]

[1] *Department Physik, ETH Zurich, Wolfgang-Pauli-Str. 16, 8093-Zurich, Switzerland*
[2]*Institut für Kernphysik, Johann Wolfgang Goethe Universität, Max-Von-Laue-Str. 1, 60438 Frankfurt am Main, Germany*
[3]*FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands*
cirelli@phys.ethz.ch

Abstract: We have demonstrated a new technique to achieve attosecond resolution without using attosecond pulses based on 'angular-streaking' with carrier envelope offset phase (CEP) controlled circularly polarized light. We resolved sub-cycle ionization dynamics with an accuracy of 23 as rms and temporal resolution of 150 as when compared to a semi-classical model.

"Angular-streaking" is a new technique to achieve attosecond time resolution using circularly polarized CEP-controlled intense pulses in the two-optical-cycle regime. The turning electric field vector of a circularly polarized pulse is used to deflect photoelectrons in angular spatial direction such that the instant of ionization is mapped to the final angular momentum in the polarization plane – similar to the "minute hand" of a clock. This 'atto-clock' runs over one 360-degree turn of the electric field which in our case corresponds to 2.3 fs. For normal optical pump-probe experiments temporal resolution is limited by the length of pump and probe pulses. Here we have demonstrated for the first time attosecond angular streaking, obtained a temporal resolution of ~150 as and an accuracy of 23 as without the need of any attosecond pulses.

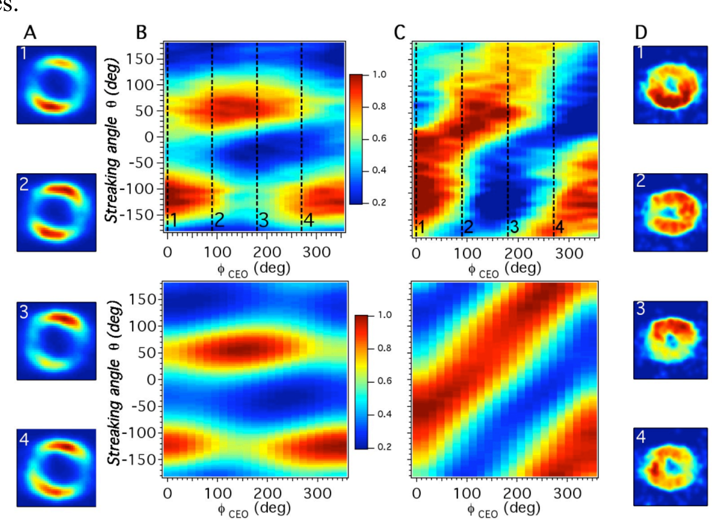

Fig. 1: Angular streaking measurements and semic-classical simulation (a) toroidal He ion momentum distributions projected onto the polarization plane for four different CEP values $\phi_{CEO} = 0$, $\pi/2$, π and $3/2\pi$ respectively. (b) radially integrated distributions as a function of CEP for the entire CEP scan (left) compared to simulations (right). (c) The data has been normalized to the CEP-averaged distribution (which effectively corresponds to the non-CEP-stabilized IR pulse), measurement (left), simulations (right). (d) analogous to (a) but for the normalized data.

Using attosecond angular streaking we fully characterized the sub-cycle photoionization dynamics of Helium generated by a minor amount of ellipticity in the intense infrared pulse. The small ellipticity (i.e. ≈0.92) results in sub-cycle oscillations of the electric field envelope with a period of approximately half the optical cycle of the carrier field. Shifting the CEP moves the pulse envelope through the sub-cycle oscillations as observed in Fig. 1a and b. Normalizing the measurement to the CEP-averaged data set results in a linear mapping of the CEP to the peak ionization streaking angle – as expected for an ideal circularly polarized streaking field (Fig. 1c and d). A different streaking technique with sub-femtosecond time resolution was demonstrated before, the so-called "attosecond-streaking" or "energy streaking" with which the electric field of a few-cycle pulse has been measured with 200 as time resolution [1]. Combining attosecond angular streaking with state of the art multi-electron detection techniques allows for recording the timing sequence of several steps in few electron processes by mapping the time of one electron to an angle in space and the time difference between subsequent ionization events to angular differences with an unsurpassed high accuracy (e.g. Fig. 2A and 2B). These measurements have the potential to reveal deeper insight in possible electron correlation effects in the timing difference of the individual tunneling ionization events of the two electrons. In addition, we expect that the time resolution in the molecular clock experiment [2] can be improved substantially. Furtherfore, this technique can also be used to measure the CEP [3] with an accuracy of 23 as, corresponding to an uncertainty of 0.06 rad.

For this measurement we used intense infrared (IR) pulses using a CEP-stabilized Ti:sapphire amplifier system with a two-stage filamentation compressor [4] to generate 5.5-fs pulses with a peak intensity of $I_p = 3.9 \cdot 10^{14}$ W/cm^2. Close to circularly polarized light was obtained with a custom-made ultrabroadband zero-order quarter wave plate. We measured the momentum distributions of the He^+ ions with a COLTRIMS (COLd Target Recoil Ion Momentum Spectroscopy) setup [5]. The CEP was ramped shot by shot by adding an additional offset in the CEP stabilization loop, such that the phase was shifted by 351° over a time duration of 120 min and the results are shown in Fig. 1 and 2.

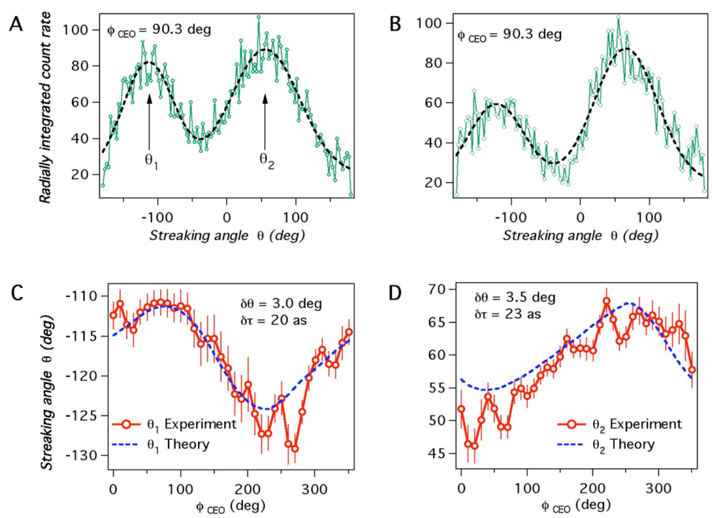

Fig. 2: CEP dependency of the electric field ionization in He using attosecond angular streaking: A), B) A double peak structure and peak position is clearly resolved within a measurement span of one optical cycle (i.e. 2.3 fs or 360° streaking angle span) for two different but constant CEP-values. C), D) The angular position of the two peaks as a function of CEP. The agreement with a semi-classical model is very good and results in a peak position accuracy of 23 as rms and a resolution of ≈150 as.

References

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