EPJ Web of Conferences **205**, 02002 (2019) https://doi.org/10.1051/epjconf/201920502002
UP 2018

The molecular attoclock: sub-cycle control of electronic dynamics during H_2 double ionization

Václav Hanus[1,*], *Sarayoo* Kangaparambil[1], *Seyedreza* Larimian[1], *Xinhua* Xie[1], *Markus* Schöffler[2], *André* Staudte[3], *Gerhard* Paulus[4], *Andrius* Baltuska[1] and *Markus* Kitzler[1]

[1]Photonics Institute Technische Universität Wien, Gusshausstrasse 27, 1040 Wien, Austria
[2]Institut für Kernphysik, J.W. Goethe-Universität, Max-von-Laue-Strasse 1, 60438 Frankfurt am Main, Germany
[3]Joint Attosecond Science Lab. of the National Research Council and the University of Ottawa, 100 Sussex Drive, Ottawa, Ontario, Canada
[4]Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany

Abstract. We introduce and employ the molecular attoclock method. This allows us to simultaneously trace the nuclear and electron dynamics during H_2 fragmentation, and to CEP-control the two-electron emission dynamics on sub-cycle time scales.

1 Introduction

The laser-sub-cycle mapping between the ionization time of an electron and its final momentum established by the rotations of the electric field vector of circularly or elliptically polarized laser pulses can be exploited for investigating double-ionization dynamics [1,2]. In this angular streaking concept, also known as the attoclock method, the absolute values of the momenta of the electrons and their emission angle, associated with the hour respectively the minute hand of the clock, encode the release times of the electrons. To obtain absolute timing, the clock needs to be calibrated by the carrier-envelope phase (CEP) of the laser pulses, as has been shown and exploited for measurements on atoms [2]. However, to apply the clock to molecules, an additional hand for encoding the nuclear motion that may take place during double ionization needs to be added.

Here, using H_2 as a model, we show that the kinetic energy release (KER) can serve as this hand of the clock. Using the resulting molecular attoclock we experimentally trace the sub-cycle dynamics of two-electron emission that underlies the fragmentation dynamics into two protons. In our experiments, we use a reaction microscope to measure in coincidence the momenta of protons and electrons created upon interaction of a cold beam of H_2 with strong, elliptically polarized laser pulses with a duration of ~5 fs. CEP sensitivity was obtained with a phase-meter [2].

* Corresponding author: vaclav.hanus@tuwien.ac.at

2 Timing of nuclear motion

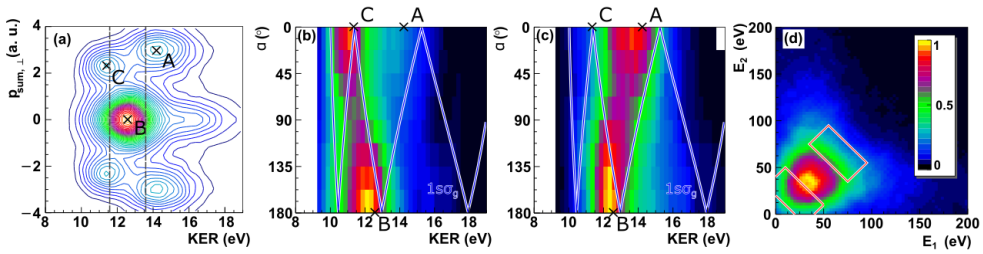

Fig. 1. Yield of protons created upon double ionization of H_2 as a function of kinetic energy release (KER) and the sum momentum of the two emitted electrons (a), the relative angle of the electrons (b, c). Distributions in (b, c) are made of events with low and high electron energy, respectively, as indicated in (d) which shows first and second electron energy correlation. Points marked by A, B, C indicate parallel (A and C) and anti-parallel (B) emission of the two electrons.

Double ionization of H_2 consists of two steps. The first step, the removal of the first electron, prepares the molecular ion and initiates nuclear stretch motion. The second ionization step, which takes place with a certain delay to the first one, catches the nuclear motion at some inter-nuclear distance R and induces molecular fragmentation into two protons. The Coulomb explosion of the two protons results in a KER that constitutes a precise measure for R. The energy of emitted electrons localizes the ionization event within the laser envelope. At the same time, the emission angle of the electrons corresponds to the direction of the electric field vector at the time of ionization. Thus, the relative angle between the first and second electron contains sub-cycle timing information. If the electrons are emitted parallel to each other (with ionization delays of an even number of laser-half-cycles) their momenta add up and thus the electron sum momentum will be large. Contrarily, if the two electrons are emitted in anti-parallel manner (with delays of an odd number of half-cycles) they will exhibit small sum momentum. Therefore, the direction of sum momentum provides timing reference in multiples of half-cycles, see Fig. 1(a). Moreover, the emission angle of the two electrons, constitutes finer time reference for the ionization. By relating this angle to the nuclear motion, characterized by KER as described above, the nuclear motion can be timed on sub-cycle time scales. This is demonstrated in Fig. 1(b, c). Additionally, it turns out that long ionization delays produce electrons of lower energy than short delays. Indeed, selecting low energy electron events as indicated in Fig. 1(d) reveals projections of the wave-packet between 1.5 and 2 laser cycles (Fig. 1(b)) while high energy electrons show the dynamics between 1 and 1.5 laser cycles (Fig. 1(c)). Timing values extracted from the figure are summarized in Tab. 1. Including the inter-nuclear distance encoded in KER via the 1/R relation.

Table 1. Timing and KER values extracted from Fig. 1(a) for the points marked A, B and C together with the inter-nuclear distance calculated from KER.

Peak	Delay (cycles)	t_{21} [fs]	KER	Inter-nuclear distance (a.u.)
A	1.0	2.50	14.2	1.99
B	1.5	3.75	12.6	2.23
C	2.0	5.00	11.3	2.46

3 Sub-cycle control of ionization

Let us consider the effect of the nuclear motion during on the double ionization dynamics. As the nuclei evolve and the inter-nuclear distance R increases in between the first and

second ionization steps, the ionization potential strongly decreases. As a result, the double ionization probability is very small for ionization delays shorter than one half-cycle, but strongly increases within only a few laser half-cycles [3]. Thus, for a pulse consisting of less than two cycles, the double ionization dynamics is well confined around a delay of about 1.5 half-cycles and KER of 12.6 eV (see Fig. 2(b)). However, the dynamics of two-electron emission can be fine-tuned on a sub-cycle time-scale using the CEP. To demonstrate this, we plot in Fig. 2(a) the asymmetry of two-electron emission in the polarization frame as a function of CEP and the ionization delay (quantified by KER). For a given ionization delay (the example of 1 cycle is indicated in the figure), the double ionization dynamics can be tuned from the case where both electrons are emitted into the upper/lower hemisphere (positive respectively negative asymmetry), and to an equal emission probability into both hemispheres (zero asymmetry).

To visualize the underlying sub-cycle double-ionization timing we performed semi-classical simulations of the ionization process. The simulation included the dependence of the ionization rate on the inter-nuclear distance. The numerical results well reproduce the asymmetry map of Fig. 2(a) (not shown) and allow us to extract the ionization times of the first and second electron. As an example, we show the case of large asymmetry for an ionization delay of one cycle in Fig. 2(c). The figure clearly shows that the experimentally observed asymmetry is due to the sub-cycle localization of the ionization steps. For different ionization delays the CEP needs to be adapted to obtain the largest asymmetry. This explains the CEP and KER dependence of the asymmetry map in Fig. 2(a).

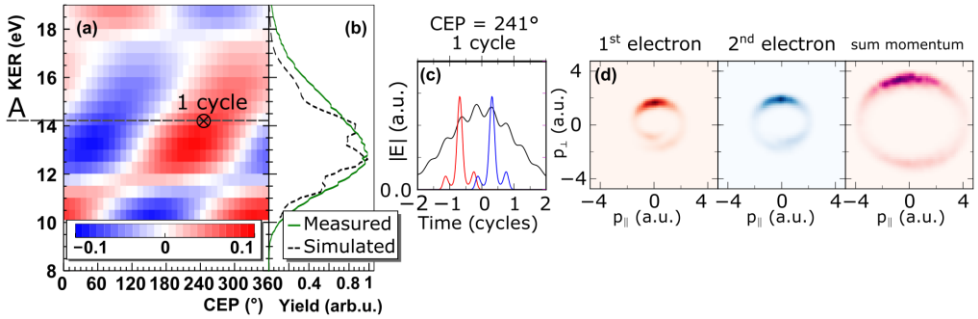

Fig. 2. (a): Measured polarization-frame asymmetry of two-electron emission calculated as the normalized difference between the ionization yields where both electrons are emitted into either the upper or lower hemisphere. The line marks the KER value corresponding to an ionization delay of 1 cycle. (b): Yield of double ionization as a function of KER (measured and simulated). For the point of maximum asymmetry marked in (a) we show the simulated ionization probabilities in (c) for first (red) and second (blue) electron on top of the magnitude of the electric field (black line). The 2D momentum distributions of first and second electron in polarization plane as well as the distribution of the sum momentum are given in (d).

References

1. C. M. Maharjan, A. S. Alnaser, X. M. Tong, B. Ulrich, P. Ranitovic, S. Ghimire, Z. Chang, I. V. Litvinyuk, and C. L. Cocke, Phys. Rev. A - At. Mol. Opt. Phys., vol. **72**, no. 4, pp. 1–4 (2005).
2. M. S. Schöffler, X. Xie, P. Wustelt, M. Möller, S. Roither, D. Kartashov, A. M. Sayler, A. Baltuska, G. G. Paulus, and M. Kitzler, Phys. Rev. A, vol. **93**, 63421 (2016).
3. S. Erattupuzha, C. L. Covington, A. Russakoff, E. Loetstedt, S. Larimian, V. Hanus, S. Bubin, M. Koch, S. Graefe, A. Baltuska, X. Xie, K. Yamanouchi, K. Varga, and M. Kitzler, J. Phys. B At. Mol. Opt. Phys., vol. **50**, 125601 (2017).