Photoionization Time Delay in Molecular Hydrogen

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Abstract: We extract the photoionization time delay difference between argon and molecular hydrogen with two complementary interferometric measurement techniques using attosecond pulses. The methods show different results, calling for different theoretical interpretations of the experimental observations.
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1. Introduction

In recent years, the great advance in ultrafast science, particularly the generation and use of XUV attosecond pulses, has allowed to start, control and probe electron dynamics in atomic and molecular systems. Among others, one of the most intriguing applications of attosecond science has been the determination of a time delay in single-photon ionization from atomic targets. Single Attosecond Pulses (SAP) using the attosecond streaking method [1] or Attosecond Pulse Trains (APT) in a RABBITT (Reconstruction of Attosecond Beating By Interference of Two-photon Transition) [2] experiment were employed to measure the relative time delay between electrons emitted from two distinct energy levels of neon and argon, respectively.

With the support of theoretical studies it is well accepted that in both experiments one part of the observed delay is caused by the influence of the infrared (IR) probe pulse. Analytical expressions for such a measurement induced time delay τ_{IR} can be derived and show a significant dependence on the kinetic energy of the emitted electrons, particularly at low energies [3,4]. Once this contribution is subtracted from the measured time delay τ_m, direct access to the single-photon ionization time delay, also called Wigner time τ_{WS}, is feasible [5].

Here, we present the first experiments addressing the single-photon ionization time delay for randomly oriented molecular hydrogen (H_2), the simplest molecular target. We measure the difference $\Delta\tau_m^{Ar,H2}$ in time delays between electrons emitted from the $3p^6$ shell of argon (Ar) and the Highest Occupied Molecular Orbital (HOMO) of H_2 by means of two distinct methods, employing attosecond streaking and RABBITT.

In contrast to previous experiments [1,2] we choose the approach of studying photoionization time delays for electrons detected with approximately the same kinetic energy emitted from two different species within the same gas target. Due to the coincidence capability of the detector setup and the gas target itself consisting of a proper H_2/Ar mixture, we are able to simultaneously obtain both RABBITT traces as well as both streaking traces, respectively, for H_2 and Ar in every single measurement.

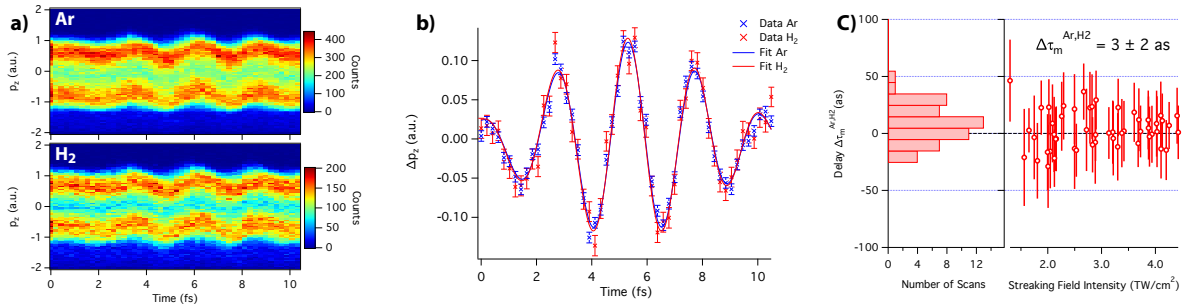

Fig. 1: a) Attosecond streaking momentum spectra corresponding to an electron energy of about 14 eV; b) For any delay step, average momentum (data points) and corresponding fit (solid line); c) Histogram of the time delay between the two species and its distribution as a function of the IR streaking field intensity. Data points are extracted from 52 independent measurements.

Since the IR-induced time delay τ_{IR} only depends on the electron kinetic energy [2], it is valid to assume that τ_{IR}^{H2} is equal to τ_{IR}^{Ar} and therefore vanishes once the difference is computed [5]: $\Delta\tau_m^{Ar,H2} \approx \Delta\tau_{WS}^{Ar,H2} = \tau_{WS}^{Ar} - \tau_{WS}^{H2}$. Therefore, if the contribution of the Ar delay τ_{WS}^{Ar} is known [6], the Wigner time of H_2 τ_{WS}^{H2} can be computed.

2. Setup

We generate APTs with a cut-off energy of about 40 eV by focusing 800 nm IR laser pulses of approximately 30 fs duration, with a repetition rate of 10 kHz into an Ar gas target. Just after the generation, a 300 nm thick

aluminum filter is used to remove the fundamental co-propagating IR and compensate the intrinsic chirp of the attosecond pulses. SAPs are obtained with driving IR pulses compressed to about 5 fs using a neon filled hollow core fiber and employing the polarization gating technique. With this generation scheme, SAPs centered at about 30 eV, with 10 eV bandwidth and a duration of less than 200 as, can be produced.

The XUV-pump beam is then recombined with the delayed IR-probe through a holey mirror and both beams are collinearly focused by a toroidal mirror into a supersonic gas target of a Reaction Microscope [7] detector. Here, ions and electrons are separated by the electric field of the spectrometer and guided towards space and time sensitive detectors. This allows to retrieve the 3D momentum vector of each individual particle at the moment of ionization and thus enables to measure electrons and ions in coincidence allowing to distinguish between electrons resulting from the photoionization of H_2 and Ar, although they energetically overlap. The gas target contains a proper mixture of H_2 and Ar such that in every measurement the experimental conditions for both species are the same thus minimizing potential sources of errors and ensuring the possibility of directly comparing both RABBITT traces and both attosecond streaking traces, respectively, with each other.

3. Results

In a first experiment, the attosecond streaking method was employed. Fig. 1 shows the results achieved with this technique. For each species the two momentum traces, corresponding to an electron kinetic energy centered at about 14 eV, represent electrons emitted along the z-axis, which is the SAP polarization direction (Fig. 1a). Electrons detected with positive (negative) momentum are those being emitted towards (away from) the electron detector.

For any delay step, a momentum average is calculated resulting in the data points shown in Fig. 1b. The photoionization time delay between the two species can be extracted from the phase differences obtained by the fits of the data. This procedure is repeated for 52 independent measurements and the distribution of the results is shown in Fig. 1c as a function of the streaking field intensity, estimated by the amplitude of the momentum modulations. The estimated $\Delta\tau_{WS}^{Ar,H2}$ is equal to 3 ± 2 as with its uncertainty representing the error of the mean value.

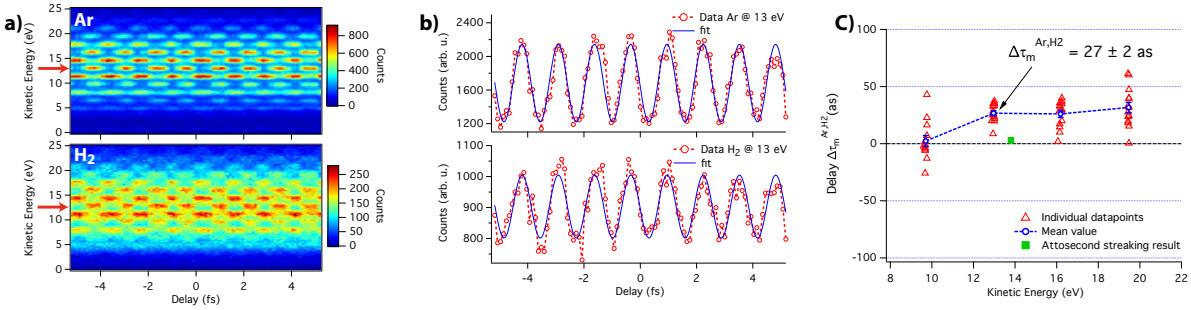

Fig. 2: a) RABBITT traces of Ar and H_2, respectively; b) SB oscillations for e.g. SB 3 at about 13 eV, marked by the red arrows in a) (data points) and corresponding fits (solid lines); c) Distribution of the individual time delays obtained by every of the 14 individual measurements (triangles) as well as their mean value (circles) as a function of the SB energy. The green square represent the result obtained by attosecond streaking measurements shown in Fig. 1.

In a second experiment, the RABBITT method was applied. Its results are shown in Fig. 2. The sideband (SB) oscillations of Ar and H_2 are clearly visible (Fig. 2a) and as an example the oscillation of SB 3 at an energy of about 13 eV is illustrated (Fig. 2b). The time delay $\Delta\tau_m^{Ar,H2}$ between Ar and H_2 for each SB is obtained by the phase differences resulting from the fits of the data for every of the 14 individual measurements. For SB 3 $\Delta\tau_m^{Ar,H2}$ results to be 27 ± 3 as. The uncertainty is the error of the mean value. This value disagrees by about 25 as with the result extracted from the attosecond streaking measurements, which have been performed at a similar electron kinetic energy.

In conclusion, both techniques give complementary access to the photoionization time delay difference between argon and molecular hydrogen. However, for an electron kinetic energy of about 13 eV, which is accessible using both methods, the extracted results differ by about 25 as, calling for different theoretical interpretations of the measured time delays.

4. References

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