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Recent attoclock measurements of strong field ionization

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ABSTRACT

The attoclock is a powerful, new, and unconventional experimental tool to study fundamental attosecond dynamics on an atomic scale. We have demonstrated the first attoclock with the goal to measure the tunneling delay time in laser-induced ionization of helium and argon atoms, with surprising results. It was found that the time delay in tunneling is zero for helium and argon atoms within the experimental uncertainties of a few 10's of attoseconds. Furthermore we showed that the single active electron approximation is not sufficient even for atoms such as argon and the parent-ion interaction is much more complex than normally assumed. For double ionization of argon we found again surprising results because the ionization time of the first electron is in good agreement with the predictions, whereas the ionization of the second electron occurs significantly earlier than predicted and the two electrons exhibit some unexpected correlation.

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1. Introduction

The key process underlying attosecond physics is strong field ionization by femtosecond laser pulses [1–3]. The strong electric field of the laser pulse bends the binding potential of the ion such that the electron is separated from the continuum by a potential barrier, through which it can escape by tunneling. Depending on the phase of the external electric field, the tunneled electron may recollide with the parent ion. Possible processes upon recollision are scattering to higher energies, which leads to an extended plateau in the photoelectron spectrum [3,4], or scattering with other electrons leading to double or multiple ionization [5]. Another possibility is that the electron recombines with the parent ion, which leads to the emission of high-energy photons. This is the so-called process of high-order harmonic generation (HHG) [4,6–9], which is the fundamental process for the generation of attosecond pulses [10,11].

Attosecond pulses and laser-controlled electron trajectories have been used for direct imaging of the electric field of a laser pulse [12], to observe quantum path control and interference within the HHG process [13,14], to image molecular orbitals [15], and to probe molecular dynamics on an ultrafast time scale [16–18]. In this sense progress in attoscience, aiming at the resolution of the dynamics of the electron motion on its natural timescale, requires a detailed understanding of tunnel ionization.

In attosecond streaking [19,20], a weak attosecond pulse ionizes an atomic [21], molecular [22] or solid target [23]. The attosecond pulse is overlaid with a relatively intense femtosecond infrared laser pulse. The electron gains a final energy that depends on the instant it appears in the continuum where it is accelerated by the femtosecond pulse. Therefore the photoelectron spectrum depends on the delay between the atto- and the femtosecond pulse. Attosecond streaking experiments have addressed very fundamental questions in quantum mechanics such as: how fast can light remove a bound electron from an atom [21,24,25] or from a surface [23]?

The attoclock is a powerful, new, and unconventional tool to study fundamental attosecond dynamics on an atomic scale. The attoclock is also an attosecond streaking technique but here the time reference is given by a close-to-circularly polarized laser field. In this way it is possible to obtain attosecond time resolution by employing a femtosecond pulse [26]. However, the attoclock technique could also be used with attosecond pulses. The close-to-circularly polarized laser field with the rotating electric field vector gives the time reference similar to the hands of a clock and is based on the definition of "time" by "counting cycles and/or fractions of cycles". In case of a "normal clock" the hour hand rotates the full cycle over 360° in 12 h, the minute hand in 60 min and the second hand in 60 s. Thus, the faster the rotating hand the more accurate is the time measurement. In case of the attoclock, the "minute hand" is the rotating electric field vector of the laser pulse which rotates a full cycle in 2.7 fs (for a center wavelength of 800 nm).

For the attoclock we measure the laser pulse polarization, and the momentum vectors of ions and electrons in coincidence, using

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"COLd Target Recoil Ion Momentum Spectroscopy" (COLTRIMS) [27,28]. Depending on the data acquisition time, an accuracy of a few attoseconds can be achieved because the measurement is based on a peak search for the exact angle at which the highest electron and ion count rate are observed [24]. The attoclock cycle, the "time zero", the laser polarization and the exact time evolution of the streaking laser field are fully characterized independently. In addition, using both clockwise and anti-clockwise polarized pulses we can minimize systematic errors in the angular streaking.

Because of the close-to-circular polarization, the probability for re-scattering of the liberated electron with the parent ion is very low and the ionization event is very well isolated. After the tunneling event, the liberated electron is considered to propagate classically in the combined laser field and the potential of the parent ion, so that the instant of ionization can be mapped to the angle of the final momentum of the electron in the polarization plane, measured with the COLTRIMS apparatus. The ion and electron momenta are measured in coincidence, which means that they can be grouped according to their atom of origin. This opens up the possibility to explore correlations between the involved particles.

2. Outline

In this paper we review recent experiments that made use of the attoclock technique. First it is discussed how the attoclock provides timings on a coarse and on a fine scale, similar to the hour and the minute hand of a watch face. This principle can be used to measure the "ionization time", i.e., the time zero of the electron trajectory. We found that the ionization time of the first electron in double ionization of argon is in good agreement with the model predictions, whereas the ionization of the second electron occurs significantly earlier than predicted [29].

A comparison of the maximum in the distribution of the electron emission angle to the angular orientation of the polarization ellipse has allowed us to resolve the timing of the ionization process. We measured the time in between the maximum of the electric field and the maximum of the ionization rate, referred to as "tunneling delay time". We found that the tunneling delay time is zero within the experimental uncertainties of a few 10's of attoseconds.

A modified semi-classical model was introduced which explained our experimental results taking into account a more complex electron parent-ion interaction with multi-electron effects and the Stark shift during the streaking process [30]. The tunnel exit points calculated with the new model are generally larger than in the standard model, and the threshold intensity for over-the-barrier ionization is increased substantially.

3. Initial conditions for the semi-classical model

A very simplified and very useful model to describe ionization in an intense laser pulse is the so-called semi-classical model [4,31,32]. This semi-classical picture uses a scalar potential for the laser field interaction, which corresponds to the length gauge in the dipole approximation. This is generally considered to be a valid picture for a center wavelength of around 800 nm and the laser intensities discussed here (e.g. shaded area in [33]). This semi-classical model in the length gauge then results in two distinct steps in the ionization process with an elliptically polarized laser field: the first step is the tunnel event which liberates the electron from the bound state in the atom (i.e. a clear quantum mechanical event) and the second step is the classical trajectory of the liberated electron in the combined potential of the parent ion and the strong laser field.

The electric field of the laser pulse given by

$$\mathbf{F}(t) = \begin{pmatrix} F_x(t) \\ F_y(t) \end{pmatrix} = \frac{f(t)\sqrt{I}}{\sqrt{\varepsilon^2+1}} \begin{pmatrix} \cos(\omega t + \varphi_{\mathrm{CEO}}) \\ \varepsilon \sin(\omega t + \varphi_{\mathrm{CEO}}) \end{pmatrix} \tag{1}$$

where I is the peak intensity, $f(t)$ is the normalized field envelope, ε is the ellipticity, and φ_{CEO} is the carrier-envelope-offset (CEO) phase [34]. With this convention, the xy-plane is the polarization plane and the major polarization axis is oriented along the x-axis. Atomic units are used throughout the paper and the atomic unit of intensity is $3.509 \times 10^{16}\,\mathrm{W/cm^2}$.

In the following electron motion the laser field exerts the dominant force, but also the Coulomb attraction by the parent ion acts on the electron. However, the electron is pulled far away from the parent ion within a fraction of the optical cycle and the Coulomb attraction becomes a small perturbation compared to the laser field. In the simplest approximation the Coulomb field is neglected after ionization. In this case, the final momentum after the laser pulse of an electron that was liberated at a time t is approximately given by:

$$\begin{pmatrix} p_x(t) \\ p_y(t) \end{pmatrix} \approx \frac{f(t)\sqrt{I}}{\omega\sqrt{\varepsilon^2+1}} \begin{pmatrix} \sin(\omega t + \varphi_{\mathrm{CEO}}) \\ -\varepsilon \cos(\omega t + \varphi_{\mathrm{CEO}}) \end{pmatrix} \tag{2}$$

Even though the simple approach of neglecting the interaction of the liberated electron with its parent ion potential is very successful for many purposes, effects have been discovered revealing that the Coulomb force influences the electron trajectory in the continuum or even under the barrier [35]. These effects include Coulomb focusing [36,37], Coulomb asymmetry in above-threshold ionization [38] and the recently discovered low energy structure at mid-infrared laser wavelengths [39,40]. More recently we clearly demonstrated that the Coulomb potential alone is not a sufficient correction. We even need to take into account multi-electron effects in an atom such as Argon. Even more complicated interactions are expected for molecules and surfaces [30].

To include the influence of the parent ion potential, the electron trajectory $\mathbf{r}$ is calculated by solving numerically the equation of motion in the combined potential of the laser pulse and the ion potential. The difference of this trajectory compared to the electron trajectory in the laser field only is mainly a change in the final electron emission direction (Fig. 1). This angular offset θ in helium is dominated by the Coulomb correction. This Coulomb correction is especially sensitive to the ion-electron attraction at the beginning of the electron trajectory. In general however this angular offset θ is much more complicated and not fully explored and understood to date [30].

The transition from the tunneling process into a classically moving particle raises serious issues both from a conceptual definition and from a measurement point of view. The link between the first and the second step of the semi-classical model, i.e. between tunneling and classical propagation, is still debated. Specifying the initial conditions for the classical electron movement after the tunnel event provides this link. The initial conditions for the electron trajectory are the ionization time, the tunneling delay time, the exit of the tunnel, and the initial momentum of the electron. In addition for the electron trajectory dynamics we need to have a clear model for the combined potential of the parent ion and the strong laser field. In a first approximation of the semi-classical picture we assume a zero initial momentum of the electron at the exit of the tunnel. This approximation can be further refined with an initial electron wavepacket spread [41]. This could be verified experimentally [42,43]. A discussion about the starting time and starting point for the electron trajectory follows in the next sections.