PRAMANA — journal of physics

Vol. 82, No. 1
January 2014
pp. 79–85

# Attosecond-correlated dynamics of two electrons in argon

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**DOI:** 10.1007/s12043-013-0645-x; ***e*Publication:** 11 January 2014

**Abstract.** In this work we explored strong field-induced decay of doubly excited transient Coulomb complex $Ar^{**} \rightarrow Ar^{2+} + 2e$. We measured the correlated two-electron emission as a function of carrier envelop phase (CEP) of 6 fs pulses in the non-sequential double ionization (NSDI) of argon. Classical model calculations suggest that the intermediate doubly excited Coulomb complex loses memory of its formation dynamics. We estimated the ionization time difference between the two electrons from NSDI of argon and it is $200 \pm 100$ as (N Camus *et al*, *Phys. Rev. Lett.* **108**, 073003 (2012)).

**Keywords.** Non-sequential double ionization; doubly excited Coulomb complex; ultrashort pulse; correlated momentum map.

**PACS Nos 32.80.Fb; 42.50.Hz; 78.47.J–; 32.80.Rm**

## 1. Introduction

Electrons play a central role in almost all fields of science and technology. Reactions like dissociation and formation of bonds involve multielectron dynamics, and hence understanding and ultimately controlling the quantum motion of correlated electrons is a big challenge. Of particular interest is non-sequential double ionization (NSDI) which involves pure two-electron motion and is a classic example of two-electron dynamics in the presence of a strong laser field ([1], and references therein). Our specific interest is to first generate a doubly excited intermediate Coulomb complex $A^{**}$ at specific laser intensity which decays via $A^{**} \rightarrow A^{2+} + 2e$ into the double ionization continuum

involving a highly correlated electronic transition state. This requires an ultrashort laser pulse that essentially confines the electron emission to one optical cycle (∼2.7 fs for 800 nm). Additionally, a field profile is required that can be controlled or measured on a subcycle level, recently realized by carrier–envelope phase (CEP) stabilization or tagging [2, 3]. Furthermore, a meaningful recipe for experimentally tracing the time of two electrons in combined Coulomb and external laser fields has to be developed and verified ([2] and references therein).

## 2. Experimental set-up

The schematic of the experimental set-up is shown in figure 1. In the experiment, linearly polarized ultrashort (6 fs, 760 nm), intense ($0.9 \times 10^{13}$ W/cm$^2$) laser pulses were used. A part of the beam was sent to Stereo-ATI spectrometer [4] so that the CEP of each single pulse could be tagged. The residual beam was focussed by a spherical silver mirror ($f = 60$ mm) inside a vacuum chamber ($2 \times 10^{-11}$ mbar) onto a supersonic argon jet. The cold target argon gas beam density in the interaction region was estimated to be $\sim 10^8$ particles/cm$^3$. The reaction volume along the pulse propagation was well defined via two slits placed in the supersonic jet. This avoids integration over different CEP due to the Gouy effect [5]. The vacuum chamber also houses the reaction microscope. The charged particles formed in the reaction microscope were guided by weak electric and magnetic fields along the laser polarization. The reaction microscope collects the photofragments with near 4pi collection efficiency for both electrons and ions generated in each laser shot (for details, see ref. [4]).

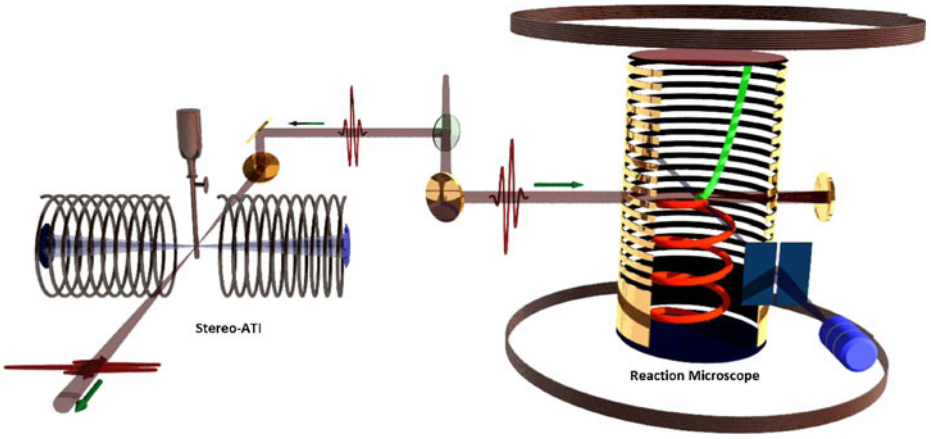


**Figure 1.** Schematic of the experiment. A 6 fs laser pulse is split into two, one part of the beam goes to Stereo-ATI to tag the CEP of the pulse and the other part goes in the reaction microscope and being focussed by a gold mirror to a supersonic gas jet. The ions and electrons formed are detected by the respective detectors. Time-of-flight and position information from the respective detectors are used to reconstruct three-dimensional momentum of electrons and ions in coincidence.

The three-dimensional momentum vectors of charged particle are constructed from measured time of flight and positions on the detector allowing us to extract kinematically complete electron–ion coincidences, and, thus, obtain information on channel separation, emission angular dependences, etc., not accessible by any other technique.

## 3. Results and discussion

In NSDI, the first electron is ionized by the electron and it travels in the field of intense laser and is driven back to the parent ion causing ionization of second electron as described in figure 2. There can be two possibilities depending on the intensity of laser pulse. First is recollision-induced excitation with subsequent ionization (RESI), where the recolliding electron excites the parent ion, which is ionized by the laser field subsequently as shown in figure 2. In the second process, the recolliding electron essentially loses all its kinetic energy during the recollision, the first electron, we call it $e_{1t1}$, is recaptured such that a doubly excited Coulomb complex (DEC) is formed. Since the intensity of the laser is low, the recolliding electron can have a maximum energy of about 15 eV which is not sufficient to ionize the second electron from $Ar^+$ ion. A theoretical calculation in ref. [2] shows that the dominant contribution to the formation of doubly charged ions is through the DEC. In the calculations it has been assumed that during the recollision, because of strong electron–electron interaction, the memory of the formation dynamic of DEC is lost.

In our experiment we used 6 fs pulses and the advantage of using the 6 fs pulses is that the DEC decays during one cycle and also multiple electron recollisions are suppressed. Moreover, fixing the CEP guarantees identical recollision energies and also DEC with similar excitation energies. In the experiment we traced the motion of electrons as a function of time. To trace out the motion we looked into the emission direction of the $Ar^{2+}$ ions in coincidence with the two electrons and defined an asymmetry parameter which depends on the CEP. The definition of asymmetry is given by the following equation:

$$A = \frac{N_+ - N_-}{N_+ + N_-},$$

where $N_+$ ($N_-$) is the number of $Ar^{2+}$ with positive (negative) momentum along the laser polarization.

In figures 3 and 4 the dependence of asymmetry of the two correlated electrons from $Ar^{2+}$ ion over CEP is plotted. The asymmetry in the emission of ions with CEP has been observed and reported [6]. However, to the best of our knowledge, our group was the first to observe and report the asymmetry in electrons [2,7]. We observed that the electrons from $Ar^{2+}$ also show a strong dependence on CEP. Figures 4a–4c are the experimental plots for asymmetry, while figures 4d–4f are plots after calculation. Figures 4b and 4e are for the least asymmetry, but figures 4c and 4f are for the maximum asymmetry. Experimentally, we can only have relative values of CEP. The absolute value of CEP was determined by comparing the data with the theory.

We presumed that the DEC is formed and then we estimated the instant of ionization of electrons from the transition state (for calculation details, see [2]). In calculations we define the ionization time difference between the two electrons. We look into

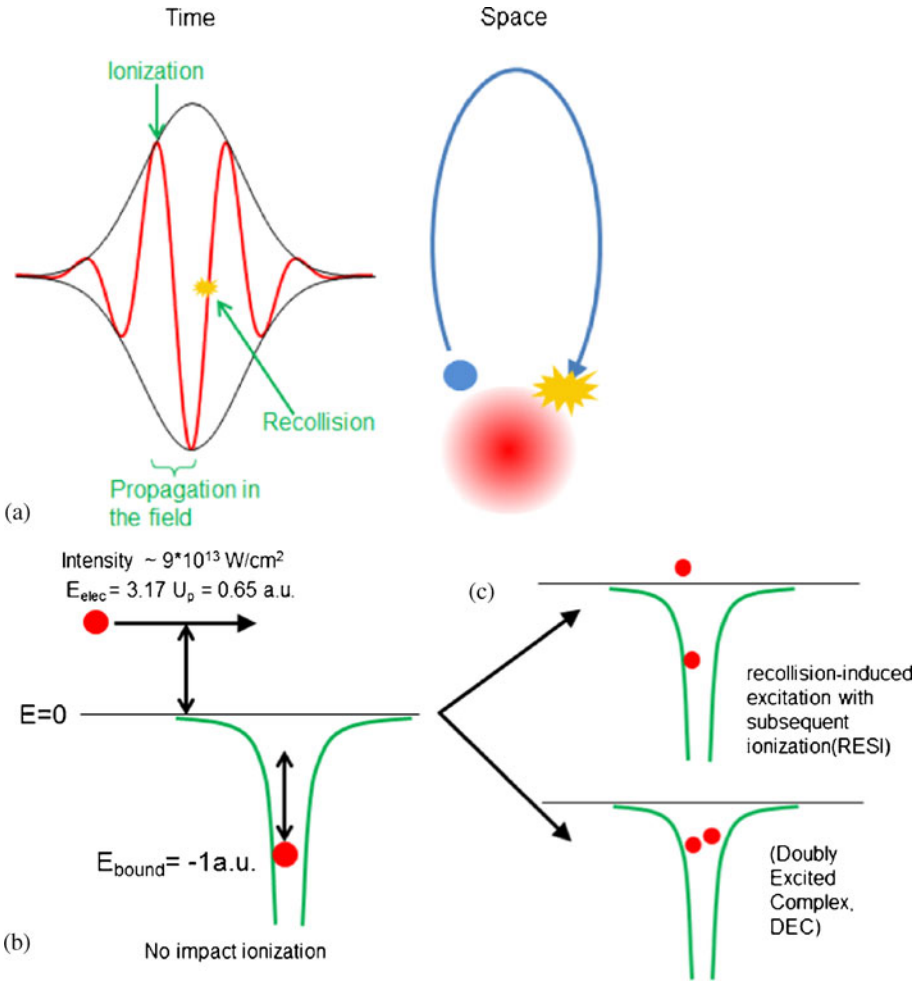


**Figure 2.** (**a**) A 6 fs laser pulse. The ionization happens at the peak of the laser pulse and electron then is said to be free and follow the laser field. As the sign of the laser field changes the electron may come back and revisit the ion. There are three possibilities: (i) Recombination of electron and hence the generation of harmonic radiations, (ii) elastic and (iii) inelastic recollision of electron with its ion core. The inelastic collision depending on the energy of the returning electron can lead to multiple ionization of parent ion core. (**b**) The returning electron carries energy less than the ionization energy of parent ion core and so the direct electron impact ionization is not possible. (**c**) In the case of low electron energy there can be two possibilities: (i) The returning electron will excite the parent ion core which subsequently will ionize which we call as recollision-induced excitation with subsequent ionization (RESI) or (ii) the returning electron is recaptured by the ion core and forms a doubly excited complex (DEC).

$\Delta t = t_{2e} - t_{1e}$ for each individual event $e$. We calculate single electron energy by considering Coulomb interaction with the ion. We neglected the electron–electron repulsion and the interaction with the laser field. In figure 5, $\Delta t$ distribution is shown for the

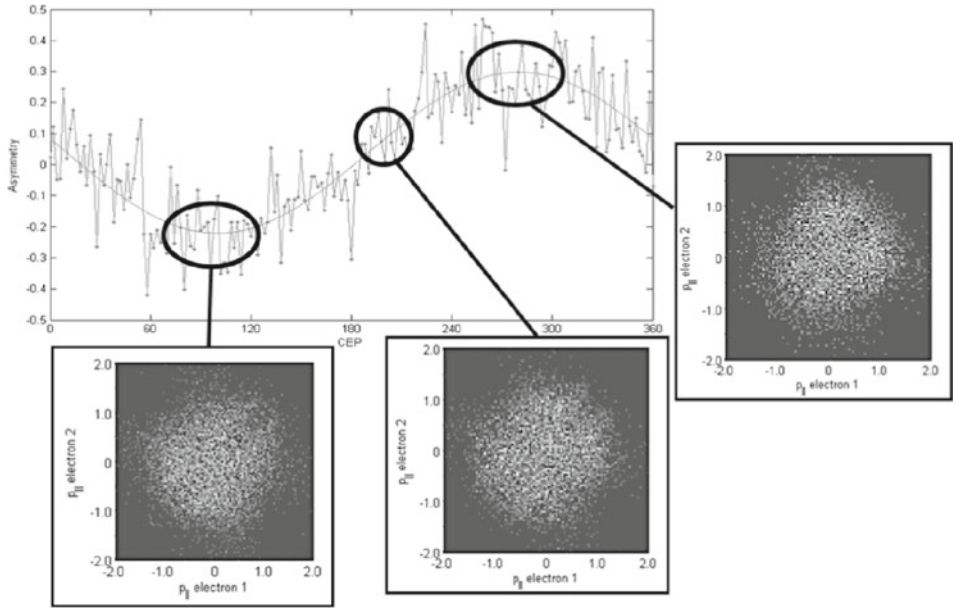


**Figure 3.** Asymmetry of electrons vs. CEP. The momentum correlation for the two electrons at different CEP is plotted in 2D momentum map.

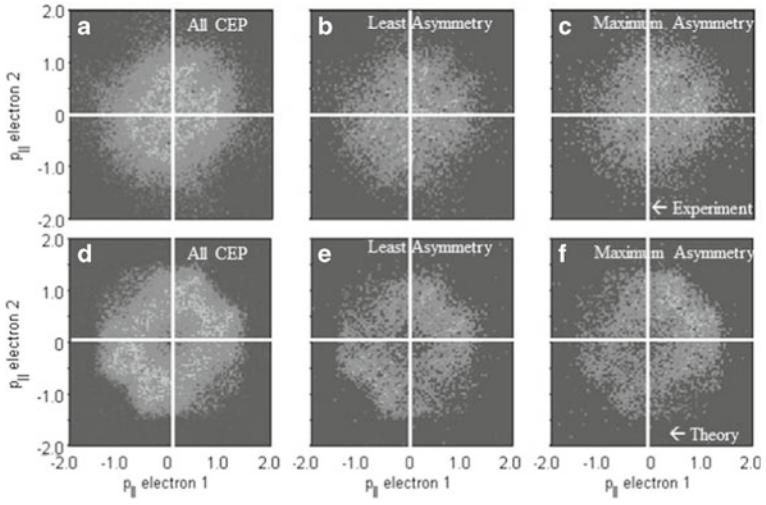


**Figure 4.** Momentum along the polarization axis of electron 1 ($P_{\|e1}$) vs. that of electron 2 ($P_{\|e2}$). (**a**, **b**, **c**) are the experiment and (**d**, **e**, **f**) are theory. (**a**) and (**d**) are averaged over all CEP, (**b**) and (**e**) for no asymmetry (at CEP $= 30°$) and (**c**) and (**f**) for maximum asymmetry (at CEP $= +60°$).

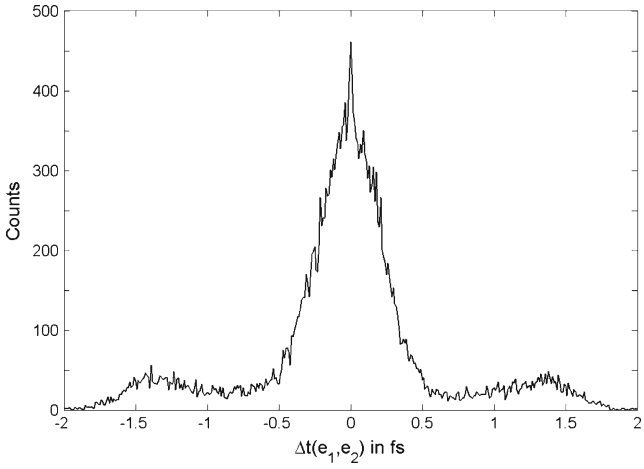


**Figure 5.** Calculated distribution for ionization time difference of electrons.

calculation. The main peak containing 80% of events at $\Delta t = 0$ (ionization time difference between the two electrons) corresponds to those events when the two electrons are ionized in the same half-cycle [2].

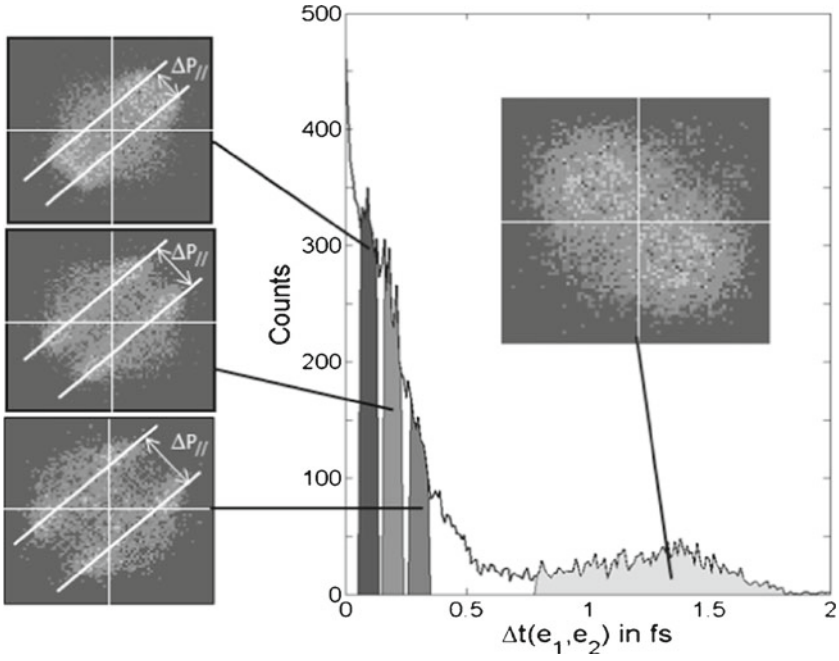


**Figure 6.** Distribution of difference of ionization time for DEC for all CEP. Inset shows the correlation map for a particular (shaded) time difference which essentially is momentum difference between the two electrons.

Now the question is: How can we measure the time difference between the ionization of the two electrons from DEC? The experimentally observable quantity for the two electrons is the drift momentum, which essentially is related to the time of birth in the laser field. We, therefore, inspect the correlated momentum map of the two electrons which is plotted in figure 5 and plot the time difference for the momentum difference (see figure 4 of ref. [2]) which in turn is $\Delta t$ (inset of figure 6). The events, which ionize in the same half-cycle of the laser, appear in the main peak and show correlated emission of electrons in the same hemisphere. On the other hand, for the events at peak 1.3 fs, the ionization of events is in the successive half-cycles and show anticorrelated momentum distribution, meaning the ionized electrons are emitted into opposite hemisphere. With this momentum distribution we calculated the (for details, see [2]) time difference between the ionization which shows a good agreement with the experiment and is equal to $200 \pm 100$ as (for details, see figure 4 of ref. [2]).

## 4. Conclusion

In conclusion, we found that a DEC was formed in NSDI complex of argon, and we related the measured momentum difference between the two electrons to the ionization time between the two electrons, and estimated the time difference.

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