Single-photon double ionization of negative hydrogen ion in presence of DC electric field.

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We present a study of the influence of the external DC electric field on the process of single-photon double ionization of negative hydrogen ion at photon energies 14.55-30 eV. We show that presence of even moderate dc electric field (of the order of 0.005 a.u.) may considerably alter cross-section of the process.

Double electron ionization of two-electron systems has been an intriguing subject of research for the last few decades. At least partially responsible for this is the fact, that near the threshold of the complete fragmentation the process allows analytic description, a rather rare occurence for the three-body Coulomb problem. The first, entirely classical theory, providing such a description has been proposed in the pioneering work of Wannier [1]. Subsequently, a number of quantum-mechanical modifications of this theory have been developed [2–6]. Not all these theories agree completely with each other. For example, approach developed in [4] predicts presence of oscillations in the cross-section near the threshold of the complete fragmentation. Such oscillations were found experimentally [7, 8] for the cross-sections of double photo-ionization of lithium and beryllium. For helium, however, experiment [9] did not reveal oscillatory structure, neither was it found in a recent extensive time-dependent theoretical calculation [10] or earlier theoretical work [11].

The mentioned above theoretical approaches are based on different pictures of two electrons leaving the atom. It may be a picture of two electrons having nearly identical velocities like in classical Wannier theory or one electron being much faster than the other like in the work [4]. It is doubtless, however, that electron correlations play a crucial role in this motion. Double electron escape is a fragile balance which can be easily disrupted by an external agent.

As such an agent we consider in the present Letter external dc field. We shall be interested in the study of the influence of the dc field on probability of the the one-photon double electron ionization for the negative hydrogen ion.

Single and double electron photoionization of negative hydrogen ion is a well-studied phenomenon [12–15]. To include external dc field in the calculation we use the method described in [16].

The method is based upon numerical integration of the time-dependent Schrödinger equation (TDSE) by expanding the solution on a square integrable basis. Once the solution of the TDSE is constructed various ionization probabilities can be obtained by projection of the TDSE wave function on a set of the field-free final states of the negative hydrogen atom. To represent these states (in particular the states belonging to double continuum of H^-) we use the so-called convergent close-coupling (CCC) method [17].

We seek a solution of the TDSE in the form:

$$i\frac{\partial \Psi}{\partial t} = \hat{H}\Psi, \qquad (1)$$

with the Hamiltonian

$$\hat{H} = \hat{H}_{H^-} + \hat{H}_{int}(t), \qquad (2)$$

where $\hat{H}_{H^-}$ is Hamiltonian of the negative hydrogen ion. Interaction with the external electromagnetic and static fields is written in the length gauge (atomic units are used):

$$\hat{H}_{int}(t) = f(t)(\boldsymbol{r}_1 + \boldsymbol{r}_2) \cdot (\boldsymbol{F}_{ac}\cos\omega t + \boldsymbol{F}_{dc}), \qquad (3)$$

Here $f(t)$ is a smooth switching function which is chosen in such a way that it is zero for $t = 0$ and $t = 8T$, where $T = 2\pi/\omega$ is a period of the ac field. For $t \in (T, 7T)$ $f(t) = 1$, for $t \in (0, T)$ it grows monotonously, so that $f(t)$ and its derivative are continuous. Analogously, for $t \in (7T, 8T)$ $f(t)$ smoothly decays from 1 to zero. The total duration of the atom-field interaction is therefore $T_1 = 8T$.

The solution of the TDSE is sought in the form of expansion on a square-integrable basis

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = \sum_j a_j(t) f_j(\boldsymbol{r}_1, \boldsymbol{r}_2). \qquad (4)$$

Here

$$f_j(\boldsymbol{r}_1, \boldsymbol{r}_2) = \phi^N_{n_1 l_1}(r_1)\phi^N_{n_2 l_2}(r_2) \ |l_1(1)l_2(2) \ L\rangle, \qquad (5)$$

where notation $|l_1(1)l_2(2) \ L\rangle$ is used for a spherical biharmonic function. Index j is used as a shortcut for the set of quantum numbers n_1, l_1, n_2, l_2, L specifying a basis vector. The radial orbitals in Eq. (5) are pseudostates

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obtained by diagonalizing the hydrogen Hamiltonian in a Laguerre basis [17]:

$$\langle \phi_{nl}^N | \hat{H}_{\text{He}^+} | \phi_{n'l'}^N \rangle = E_i \delta_{nn'} \delta_{ll'} .$$

Here E_i is the energy of a pseudostate and N is the size of the basis.

In the present work, we consider modestly strong electric fields: the ac field of the order of 0.01 a.u. corresponding to 3.5×10^{12} W/cm^2 intensity, and the dc field not exceeding 0.005 a.u. This allows us to retain terms with total angular momentum $J = 0 - 2$ in expansion (4). For all S, P, D total angular momentum states we let l_1, l_2 vary within the limits $0 - 3$. The total number of pseudostates participating in building the basis states was 20 for each l. To represent $J = 0, 1, 2$ singlet states in expansion (4), we used all possible combinations of these pseudostates. Such a choice gave us 840 basis states of S-symmetry, 1200 basis states of P-symmetry and 1430 states of D-symmetry, resulting in a total dimension of the basis equal to 3470.

To solve TDSE (1) we rewrite it as a system of differential equations for the coefficients $a_j(t)$ in Eq. (4). To obtain initial values for the a_j's we diagonalize the field-free Hamiltonian of H$^-$ in the basis set (5) and use the set of a_j's for the ground state to impose initial conditions. The energy of the ground state obtained as a result of this procedure was -0.52762 a.u. which differs by approximately 1.3×10^{-4} a.u. from the well-known value -0.527751 a.u. [18]. Such accuracy may not be very impressive by usual standards of bound state calculations, but it is quite sufficient for the present time-dependent calculation.

The second ingredient which we need to compute ionization probabilities is a wavefunction of a field-free negative hydrogen ion representing two electrons in continuum. These are provided by the CCC method [17], details of their use to represent the doubly ionized electron states can be found in [16].

A set of the final states corresponding to various photoelectron energies E_1, E_2 was prepared. Projecting the solution of the TDSE on the states of this grid we computed a probability distribution function $p(\boldsymbol{k}_1, \boldsymbol{k}_2)$ of finding system in a field-free state $(\boldsymbol{k}_1, \boldsymbol{k}_2)$ at the time $t = T_1$.

The total integrated cross-section (TICS) of double photoionization (DPI) is given by [16]:

$$\sigma(\omega) = \frac{4\pi\omega}{Wc} \int p(\boldsymbol{k}_1, \boldsymbol{k}_2) \; d\hat{\boldsymbol{k}}_1 d\hat{\boldsymbol{k}}_2 dk_1 dk_2, \qquad (6)$$

where $W = 2 \int_0^{T_1} F_{\text{AC}}^2(t) \; dt$ and $c \approx 137$ is the speed of light in atomic units.

Integrating the distribution function $p(\boldsymbol{k}_1, \boldsymbol{k}_2)$ over directions of the momenta $(\hat{\boldsymbol{k}}_1, \hat{\boldsymbol{k}}_2)$, we obtain energy distribution function $P(E_1, E_2)$.

TICS obtained as a result of the procedure outlined above are shown on Figure 1 for various values of the
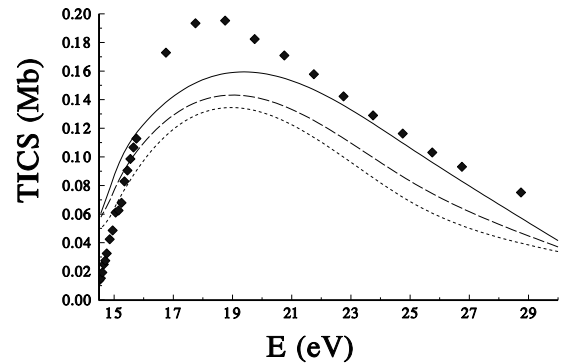

FIG. 1: Total integrated cross-section of DPI of H$^-$ for various values of external dc field; $F_{\text{dc}} = 0$ solid line (present) and solid circles (data from [11]); $F_{\text{dc}} = 0.002$ a.u. (long dash); $F_{\text{dc}} = 0.003$ a.u. (short dash).

external dc strength. As a reference data we use results given by perturbative CCC calculation [11].

As one can see from the Figure 1 the present data and perturbative CCC results [11] agree fairly well for all photon energies considered. One can see also, that for the dc field strengths presented (not exceeding 0.003 a.u.) external dc field tends to diminish TICS for all photon energies. In the region close to the threshold of DPI present calculation gives larger values for TICS than CCC calculation [11]. This is related to the fact that present calculation is based on a numerical solution of TDSE for a finite time $T_1 = 8T$. Energy conservation and hence a notion of a strictly defined threshold for the process is restored only in the limit of infinitely large T_1. Therefore, even for the zero external dc field the threshold in the time-dependent calculation is necessarily smeared out. Inclusion of nonzero dc external field adds to this smearing, since ionization due to dc field is a process which proceeds without energy conservation. Therefore even if we could attain in our numerical calculation the limit of very large T_1, the notion of a threshold of a process is somewhat ill-defined in the presence of the external dc field.

We found that this diminishing of TICS with dc field occurs only for not very large values of the dc field strength. Region of the photon energies which is of most interest for us in this Letter is the region corresponding to the Wannier regime (15 eV or less). On Figure 2 we present results for TICS for the photon energy of 15 eV for various values of the dc field strength.

TICS as a function of dc field strength reaches a minimum at approximately $F_{\text{dc}} = 0.004$ a.u., and then starts growing. More detailed information can be obtained if we analyze energy distribution of the ejected electrons, shown on Figure 3 as a contour plot. Darker shades of gray correspond to larger values of energy distribution
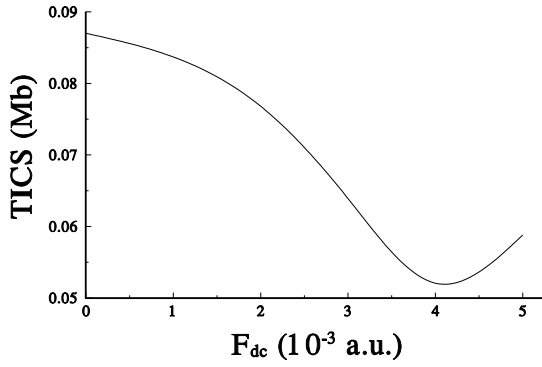
FIG. 2: Total integrated cross-section of DPI of H^- as a function of external dc field for photon energy of 15 eV.
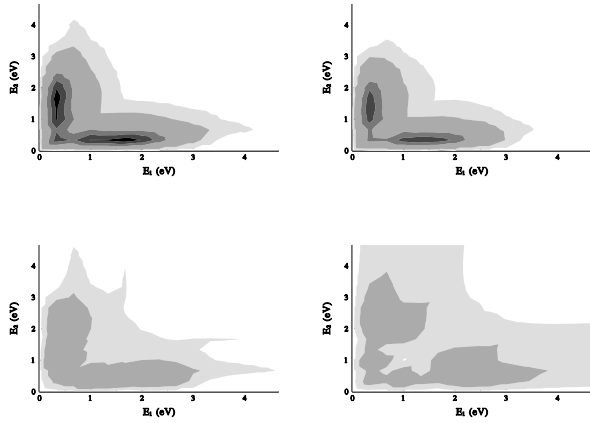
function $P(E_1, E_2)$.

FIG. 3: Electron energy distribution $P(E_1, E_2)$ for $\omega = 15$ eV, $F_{ac} = 0.015$ a.u. and $T_1 = 8T$. The dc field strength F_{dc} is 0, 0.003, 0.004, and 0.005 a.u. (from left to right and top to bottom).

One can see from the Figure 3 how the external dc field progressively erases the regular structure due to the energy conservation. For small values of the dc field, ionized electrons are to be found primarily in the region of the E_1, E_2 plane close to the energy conservation line. External dc field smears this picture, making electron distribution more homogeneous. This is to be expected, being a tunneling process, ionization due to the dc field does not conserve energy and should act therefore as a homogenizing agent. The somewhat ridged structure of the plot for $F_{dc} = 0.005$ a.u. is a clear indication that at this stage dc field contributes significantly to the ionization process. This structure results from the fact that in the expression for the transition amplitude the rapid

oscillations in the exponential function $e^{i(E(\mathbf{k}_1, \mathbf{k}_2) - E_0)T_1}$ cannot be compensated by the oscillating term in the interaction Hamiltonian (here $E(\mathbf{k}_1, \mathbf{k}_2)$ and E_0 are, respectively, energies of the final and initial states).

The growth of TICS for the dc fields exceeding 0.004 a.u. indicates therefore, that ionization due to dc field is becoming a dominant process. Role of electron correlations at this stage is probably not very significant. Indeed. a picture similar to Figure 2 is obtained already for a simple Keldysh-type model of ionization in presence of both dc and ac fields.

In the framework of the Keldysh approximation the amplitude to find electron in a state with momentum **p** at time T_1 when external field is switched off, is [19]:

$$a_{\mathbf{p}} = -i\langle \mathbf{p}|\hat{P}|\Psi_0\rangle \int_0^{T_1} e^{-\frac{i}{2}\int_\tau^{T_1}[\mathbf{p}+\mathbf{A}(\tau_1)]^2\ d\tau_1 - iE_0\tau}\ \mathbf{A}(\tau)\ d\tau,$$
(7)

where $\mathbf{A}(\tau)$- the vector potential, which in the present case, when two external fields are present, reads: $\mathbf{A}(\tau) = \mathbf{F}_{ac}\dfrac{\sin\omega\tau}{\omega} + \mathbf{F}_{dc}\tau$

This approximation is, strictly speaking, applicable to short range potentials [19]. For example, for a delta-function potential (ionization potential 0.7 a.u., ac field frequency 1 a.u.) the calculation of total ionization probability as function of dc field leads to the results shown on Figure 4.
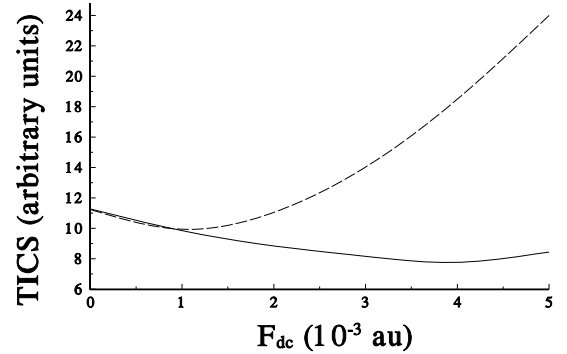

FIG. 4: Total integrated probability from short-range potential, $I_p = 0.7$ a.u., $\omega = 1$ a.u. $F_{ac} = 0.015$ a.u. (dashed line) and 0.05 a.u. (solid line).

The behavior shown on the Figure 4 is qualitatively similar to that of TICS DPI on Figure 2. Of course, Keldysh approximation neglects completely all electron correlations (and even electron-nucleus interaction) in the final state. Yet, it does not seem totally unreasonable to suppose, that similar effects may have similar causes in this case. Behavior of ionization probability from Figure 4 is a result of interplay of two fields, for some dc field strengths increase of dc field makes the barrier which

particle has to penetrate to escape, thinner. Analogous mechanism may lead to the behavior of TICS DPI of the negative hydrogen ion found in the present work.

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