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Single-photon double ionization of negative hydrogen ions in the presence of a dc electric field

I. A. Ivanov[*] and A. S. Kheifets
Research School of Physical Sciences and Engineering, The Australian National University, Canberra Australian Capital Territory 0200, Australia

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We present an accurate numerical study of the influence an the external dc electric field on the process of single-photon double ionization of the negative hydrogen ion at photon energies from the threshold to 30 eV. We demonstrate that the presence of an external dc electric field of the order of 0.005 a.u. may considerably alter the cross section of the process.

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I. INTRODUCTION

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

The theoretical approaches mentioned above are based on different pictures of the two-electron escape. It may be a picture of two electrons having nearly identical velocities as in the classical Wannier theory or one electron being much faster than the other as in [4]. Whatever picture of the electron motion we adopt, the electron correlation plays a crucial role in the DPI process. Being driven by the correlation, two-electron escape is a fragile process which can be easily disrupted by an external agent.

In the present paper, as such an agent we consider an external dc field. We shall be interested in the influence of a dc field on the probability of single-photon double electron ionization for the negative hydrogen ion H^-. Study of the one-electron ionization in the presence of both ac and dc electric fields has a relatively long history, starting from [12,13]. In these works the effect of the external dc field on the single-photon one-electron ionization in solids and negative ions has been studied. It was found that an external dc field introduces oscillatory structures in the photoionization cross sections. When an external dc field is present, the photoionization cross section exhibits resonancelike behavior for photon frequencies above the absorption threshold. For a given ac field frequency, the photoionization cross section was found to be an oscillatory function of the external dc field strength. These theoretical predictions have been confirmed experimentally [14,15]. For negative ions, the oscillations in the photoionization cross section were interpreted [16] as the result of interference of two classical trajectories of an escaping electron.

The first theoretical work treated the electromagnetic field perturbatively. Subsequent theoretical progress consisted [17–22] in the development of nonperturbative techniques allowing description of single- and multiphoton one-electron ionization in strong fields and for various geometries of the external fields. In [18] a procedure was given allowing computation of the probabilities of multiphoton detachment of an electron from a short-range potential in the presence of a strong dc field. The so-called quasistationary, quasienergy state formalism was developed and applied in [21,22] to study the effect of a strong dc field on single- and two-photon ionization of an electron from negative ions. The physical model used in this work to represent negative ions was an electron bound in a short-range potential. This model neglects, of course, electron correlations in the target atom. Despite this fact, the short-range potential model seems to provide quite an adequate picture of the process of one-electron photoionization of negative ions in the presence of both ac and dc fields. In [20] an approach was proposed which took into account the electron correlations in the intermediate and final states of the process of two-photon one-electron detachment. This work, and the nonperturbative calculations of one-electron photoionization of H^- and Li^- [23,24], which take complete account of electron correlation effects, show the same oscillatory structures in the cross sections as the theories based on the short-range potential model of negative ions. Thus, the oscillations induced by the external dc field turn out to be a "robust" phenomenon which seems, at least as far as qualitative features of the phenomenon are concerned, to be little affected by the electron correlations.

As opposed to single-electron photoionization, double-electron photoionization is, as we discussed above, an archetypical example of a process where electron correlations play a predominant role. It would be of interest, therefore, to study the effect of an external dc field on the double-electron

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[*] Corresponding author. On leave from the Institute of Spectroscopy, Russian Academy of Sciences, Troitsk. Igor.Ivanov@anu.edu.au

photoionization of a negative ion. Below, we present such a study for the single-photon double-electron ionization of the hydrogen negative ion.

II. THEORY

Single- and double-electron photoionization of H$^-$ is a phenomenon studied by many authors [25–28]. To include the effect of the external dc field, we use the method described in Ref. [29].

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 field-free final states. To represent these states, in particular the states belonging to the double continuum of H$^-$, we use the so-called convergent close-coupling (CCC) method [30].

We first find a solution of the TDSE,

$$i\frac{\partial\Psi}{\partial t} = \hat{H}\Psi, \tag{1}$$

with the Hamiltonian

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

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

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

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 the period of the ac field. For $t\in(T,7T)$ $f(t)=1$; for $t\in(0,T)$ it grows monotonically, 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 an expansion on a square-integrable basis

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

Here

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

where the notation $|l_1(1)l_2(2)L\rangle$ is used for a spherical biharmonic function. The index j is used as a shortcut for the set n_1,l_1,n_2,l_2,L specifying a basis vector. The radial orbitals in Eq. (5) are pseudostates obtained by diagonalizing the hydrogen Hamiltonian in the Laguerre basis [30]:

$$\langle\phi^N_{nl}|\hat{H}_{\mathrm{H}}|\phi^N_{n'l'}\rangle = E_i\delta_{nn'}\delta_{ll'}.$$

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: an ac field of the order of 0.015 a.u. corresponding to 7.8×10^{12} W/cm^2 intensity, and a 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 the 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 these coefficients, we diagonalize the field-free Hamiltonian of H$^-$ in the basis set (5) and use the set of a_j for the ground state to impose the initial conditions. The energy of the ground state obtained as a result of this procedure was −0.527 62 a.u. which differs by approximately 1.3×10^{-4} a.u. from the well-known value −0.527 751 a.u. [31]. Such accuracy may not be very impressive by the usual standards of bound-state calculations, but it is quite sufficient for the present time-dependent calculation.

The second ingredient that we need to compute ionization probabilities is a wave function representing two electrons in the continuum. This is provided by the CCC method [30]. In the CCC formalism, the interacting two-electron state is represented by a close-coupling expansion over the channel states, each of which is composed of a target pseudostate j and a Coulomb wave $\boldsymbol{k}$:

$$\Psi_f(\boldsymbol{k}) = |\boldsymbol{k}f\rangle + \sum_{\boldsymbol{k}'j}\frac{\langle\boldsymbol{k}f|T|\boldsymbol{k}'j\rangle}{E - k'^2/2 - \varepsilon_j + i0}|\boldsymbol{k}'j\rangle. \tag{6}$$

Here $\langle\boldsymbol{k}f|T|\boldsymbol{k}'j\rangle$ is the half-on-shell T matrix, which is found by solving a set of coupled Lippmann-Schwinger equations [32]. If the target pseudostate f has positive energy, than the solution given by Eq. (6) can be used to construct a wave function $|\boldsymbol{k}_1,\boldsymbol{k}_2\rangle$ describing two electrons in the continuum. A more detailed description of this procedure can be found in [29].

We thus prepared a set of final states corresponding to various photoelectron energies E_1, and E_2. By projecting the solution of the TDSE on these states, we computed a probability distribution function $p(\boldsymbol{k}_1,\boldsymbol{k}_2)$ of finding the ionized system in the field-free state $\boldsymbol{k}_1,\boldsymbol{k}_2$ at the time $t=T_1$.

The total integrated cross section (TICS) of DPI is given by [29]

$$\sigma(\omega) = \frac{4\pi\omega}{c}\frac{1}{W}\int p(\boldsymbol{k}_1,\boldsymbol{k}_2)d\hat{\boldsymbol{k}}_1 d\hat{\boldsymbol{k}}_2 dk_1 dk_2, \tag{7}$$

where $W=2\int_0^{T_1}F^2_{\mathrm{ac}}(t)dt$ and $c\approx137$ is the speed of light in atomic units. By 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 the energy distribution function $P(E_1,E_2)$, which can be of use if more detailed information about the system than just the TICS is needed.

These are the key steps of the procedure we employ in the work. The results are presented below.

III. RESULTS

Before discussing the results it is worthwhile to provide an estimate of the accuracy we can hope to achieve in the present calculation. This can be done following the procedure adopted in [28] or the similar procedure we used in [29].

The present calculation consists of two main steps—solving the TDSE and projecting the solution obtained upon the set of CCC states representing two electrons in the continuum. The first step can be implemented accurately, requiring, in principle, only a large enough basis set used for the construction of the TDSE solution. The second step requires more care. The wave functions on which we project the solution of the TDSE and the basis set used to solve the TDSE are two unrelated sets of functions, which are furnished by two different procedures. This may lead to considerable limitations of accuracy for the following reason.

Let us consider the time evolution of the hydrogen negative ion after the moment of time T_1, when external fields are switched off. The wave function describing this evolution can be written as a superposition:

$$\Psi(t) = \sum c_k e^{-iE_k t}\Psi_k, \tag{8}$$

where Ψ_k and E_k are solutions of the eigenvalue problem for the field-free Hamiltonian constructed with the help of the basis set (5). If we project this wave function on a final state furnished by the CCC method, than since the eigenvectors Ψ_k are not strictly orthogonal to the CCC field-free states, the overlap of the solution of the TDSE and the CCC state will contain oscillating terms $\Sigma c_k\, e^{-iE_k t}\langle\Psi_{\mathrm{CCC}}|\Psi_k\rangle$. The nonorthogonality of the eigenstates Ψ_k and CCC vectors introduces thus unphysical oscillations or beats in the computed ionization probabilities for times larger than T_1, when external fields are absent. This circumstance may affect the accuracy of the calculation considerably. If, for example, instead of the CCC vectors we used uncorrelated final-state wave functions to project upon, then as demonstrated in [28] the oscillations of the probabilities could be quite significant, reaching a factor of 2. In our approach, the CCC final states and eigenvectors Ψ_k, though obtained by two different procedures, are solutions of the Schrödinger equation for the same Hamiltonian. They retain, therefore, at least approximate orthogonality, which reduces the beats mentioned above in the probabilities to an acceptable level.

This point is illustrated in Fig. 1 where we plot the squared overlaps $|\langle\Psi_{\mathrm{CCC}}|\Psi_k\rangle|^2$ between various P-symmetry eigenfunctions of the eigenvalue problem for the field-free Hamiltonian on the basis (5) and a final-state CCC wave function at the excess energy of 5.5 eV above the double-ionization threshold.

We see that overlaps of large absolute value peak narrowly around the excess energy and the other overlaps are insignificant on this scale. The narrow localization of the overlaps dampens the beats considerably. As a check, we performed a few calculations of the TICS, allowing the solution of the TDSE to propagate freely (evolution in time governed by the field-free Hamiltonian of the negative hydrogen ion) for some time after external fields are switched off. This procedure is completely analogous to the one performed in [29], and is described there in details. These calculations show that the oscillations mentioned above lead to variations of the TICS of the order of 20%. We adopted this figure as an accuracy estimate of the present calculation.

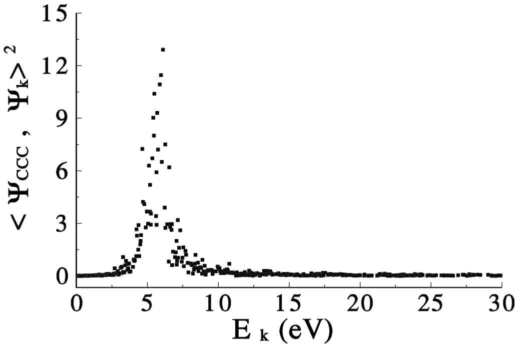

FIG. 1. Squared overlaps $|\langle\Psi_{\mathrm{CCC}}|\Psi_k\rangle|^2$ between various P-symmetry eigenfunctions of the eigenvalue problem for the field-free Hamiltonian on the basis (5) and a CCC wave function at the excess energy of 5.5 eV above the double-ionization threshold.

The issue of the dependence of the computed probabilities on time for times larger than T_1, when the laser field is switched off, which we described above, can also be looked at from a different angle. As discussed in [28], use of inadequate final-state vectors on which the TDSE solution is projected may lead to significant dependence of the computed probabilities upon the quantum-mechanical picture (e.g., the Schrödinger or interaction one) employed in the calculation. The origin of this phenomenon is, of course, exactly the same as the origin of the beats discussed above, lack of orthogonality of the eigenvectors of the field-free Hamiltonian and the final states on which the TDSE is projected.

Our results for the TICS in the absence of the external dc field are shown in Fig. 2. The data are compared with other results for the TICS for DPI in the hydrogen negative ion, available in the literature. Presented in Fig. 2 are results of a perturbative CCC calculation [11] (in length and velocity forms), a recent time-dependent calculation [28], a J-matrix

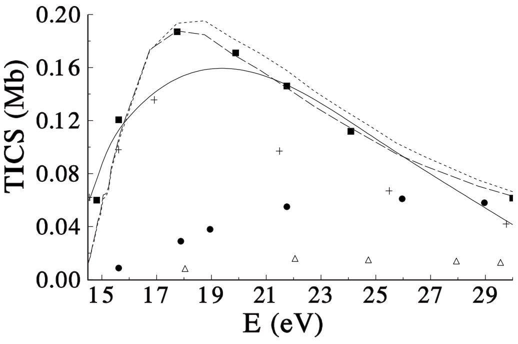

FIG. 2. Total integrated cross section of DPI of H$^-$ for zero external dc field; solid line (present results); data from [11], length form (short dashed), velocity form (long dashed); results of Foumouo *et al.* [28] (solid squares); data from [33] (pluses); results of Nicolaides *et al.* [34], length form (solid circles), velocity form (triangles).

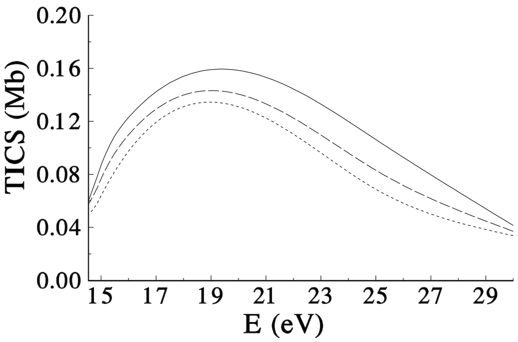

FIG. 3. Total integrated cross section of DPI of H^- for various values of external dc field; $F_{dc}=0$ (solid line), 0.002 (long dashed), and 0.003 a.u. (short dash).

calculation [33], and the results of a variational configuration-interaction calculation [34].

For the range of photon frequencies we consider in this paper, the values reported by Kheifets and Bray [11] (especially the velocity form results) are in very good agreement with the recent results given by Foumouo *et al.* [28]. Both calculations use a fully correlated representation for the final doubly ionized state of the hydrogen negative ion, and show good agreement between the results in the length and velocity forms (for the calculation [28] the velocity and length results are in fact indistinguishable on the scale of Fig. 2). The disagreement between these results and the calculation [33] may be due to the fact that an uncorrelated final state (represented as a product of a positive energy pseudostate and a plane wave) was employed in this work. The results of [34] exhibit some dependence on the gauge used.

The present data differ by approximately 20% from the results of [11,28], which is compatible with the accuracy estimate we gave above.

The effect of the external dc field on the TICS is illustrated in Fig. 3. As one can see from Fig. 3, for field strengths below 3×10^{-3} a.u., the external dc field tends to diminish the TICS for all photon energies. We found that this diminishing of the TICS with dc field occurs only for not very large values of the dc field strength. The region of the photon energies that is of most interest for us in this paper is the near-threshold region corresponding to the Wannier regime (15 eV or less). In Fig. 4, we present results for the TICS for the photon energy of 15 eV for various values of the dc field strength.

For not very large dc field strength, the behavior of the TICS for H^- is similar to that for He which we reported recently [29]. In the case of He, we found that for a dc field strength not exceeding 0.03 a.u. the TICS decreases slowly with increasing dc field. We did not attempt to study the domain of larger field strengths in that work, since for dc fields of the order of 0.03 a.u. or larger we could not achieve convergence of the calculation with respect to the number of angular momentum states included in the calculation. Proper treatment of such fields may require inclusion of terms with higher total angular momentum (and hence a much larger calculation) in the expansion Eq. (4). A much more weakly bound system, H^- offers a more promising subject of study.

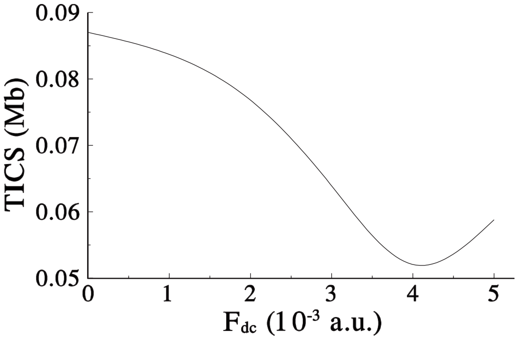

FIG. 4. Total integrated cross section of DPI of H^- as a function of the external dc field for the photon energy of 15 eV.

As one can see from Fig. 4, application of an external dc field with a strength of the order of 10^{-3} a.u. to H^- can produce considerable changes in the TICS.

As a function of the dc field strength, the TICS reaches a minimum at approximately $F_{dc}=4\times10^{-3}$ a.u. and then starts to grow. More detailed information can be obtained if we analyze the energy distribution of the ejected electrons shown in Fig. 5 as a contour plot. Darker shades of gray correspond to larger values of the energy distribution function $P(E_1,E_2)$.

One can see from Fig. 5 how the external dc field progressively erases the regular structure due to energy conservation. For small values of the dc field, the ionized electrons are to be found primarily in the region of the E_1,E_2 plane close to the energy conservation line. The external dc field smears this structure, making the 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. A somewhat ridged structure of the plot for $F_{dc}=0.005$ a.u. is a clear indication that at this stage the 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 $e^{i[E(\boldsymbol{k}_1,\boldsymbol{k}_2)-E_0]T_1}$ cannot be compensated by the oscillating term in the interaction Hamiltonian [here $E(\boldsymbol{k}_1,\boldsymbol{k}_2)$ and E_0 are, respectively, the energies of the final and initial states].

The growth of the TICS for dc fields exceeding 0.004 a.u. indicates therefore that ionization due to the dc field is becoming a dominant process. The role of electron correlations at this stage is probably not very significant. It is arguable, therefore, that for dc fields of the order of 0.004 a.u. or larger, we may consider the effect of the external dc field on the TICS of double photoionization as a predominantly one-electron phenomenon, which is not very sensitive to the electron correlation pattern.

As we mentioned in the Introduction, a nonmonotonic behavior of the ionization probability similar to that shown in Fig. 4 is characteristic of the process of one-electron ionization of negative ions in the presence of an external dc field. A theoretical explanation of this phenomenon can be obtained using an essentially single-particle picture of one-electron ionization from a short-range potential [21,22], neglecting electron correlations in the final state. Taking ac-

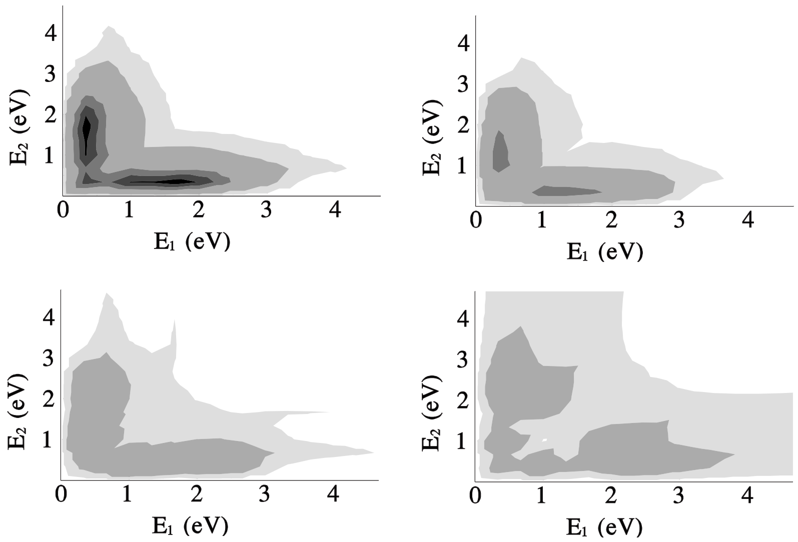

FIG. 5. Electron energy distribution $P(E_1,E_2)$ for $\omega=14.55$ eV, $F_{\mathrm{ac}}=0.015$ a.u., and $T_1=8$ T. The dc field strength F_{dc} is 0, 0.003, 0.004, and 0.005 a.u. (from left to right and top to bottom).

count of electron correlations [20,23,24] does not seem to modify the predictions of this theory qualitatively.

As we discussed above, the role of electron correlations in the DPI process may not be very important for sufficiently strong external dc field. If this surmise is correct, a simplified treatment of the DPI problem in an external dc field could be performed in essentially the same way as was done in the works treating the subject of one-electron ionization, with two noninteracting electrons escaping from a short-range potential in our case. The wave function of the final state of two electrons with momenta $\boldsymbol{\pi}_1$ and $\boldsymbol{\pi}_2$ can than be built as an uncorrelated product of one-electron states, each of which is described by a generalization of the well-known Volkov wave function [35]. In the present case, when we have both ac and dc external fields present, such a wave function, describing a state in which the electron has a kinetic momentum $\boldsymbol{\pi}=\boldsymbol{p}+A(T_1)$ at a time T_1 when the external field is switched off, is (the velocity gauge is used)

$$\Psi_{\boldsymbol{\pi}}(\boldsymbol{r},t)=\frac{1}{(2\pi)^{3/2}}\exp\left(\frac{i}{2}\int_t^{T_1}[\boldsymbol{\pi}+\boldsymbol{A}(\tau)-\boldsymbol{A}(T_1)]^2d\tau + i[\boldsymbol{\pi}-\boldsymbol{A}(T_1)]\boldsymbol{r}\right), \tag{9}$$

where the vector potential is $\boldsymbol{A}(t)=-\int_0^t\boldsymbol{F}(\tau)d\tau$; $\boldsymbol{F}(\tau)=f(\tau)(\boldsymbol{F}_{\mathrm{ac}}\cos\omega\tau+\boldsymbol{F}_{\mathrm{dc}})$.

The oscillating behavior of the ionization probablilities is an inherent property of amplitudes computed with the help of the Volkov states (9). We would have obtained such behavior for the TICS of the double photoionization had we used a two-electron final state composed of functions (9) for the calculation of the DPI of a two-electron system. This model may explain the dc field dependence of the cross section found in the present work.

IV. CONCLUSION

We have studied the influence of an external dc electric field on the total integrated cross section for the single-photon double-electron ionization of the negative hydrogen ion. The photon frequency range from the threshold to 30 eV was considered. The TICS, as a function of the dc field strength, exhibits a nonmonotonic behavior. Such field dependence of the cross section of double photoionization is reminiscent of the oscillating behavior of the total cross section of one-electron ionization of negative ions. The origin of this effect may possibly be attributed, just as in the case of one-electron ionization, to the presence of competing tunneling pathways through the dc and ac field-induced barriers.

To produce observable change in the TICS of DPI of the hydrogen negative ion one has to apply a dc field of the order of 10 MV/cm. Static fields of at least comparable strength can be obtained in the laboratory (e.g., a field of 3 MV/cm has been obtained in an experiment using a relativistic hydrogen atom beam [36]).

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