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Resonant-enhanced above-threshold ionization of atoms by XUV short laser pulses

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ABSTRACT

Above-threshold ionization of atoms by XUV short laser pulses is investigated close to the resonant 1s–2p transitions. Both ab initio TDSE and a theoretical Coulomb–Volkov like theory are used to study the enhancement in the ionization probabilities. Our modified Coulomb–Volkov theory, fully accounting for the important 1s–2p transition is able to explain the spectrum as well as the total ionization cross sections.

1. Introduction

The development of lasers capable of delivering short pulses of very intense radiation over a wide frequency range has led to the discovery of new non-perturbative effects in atomic and molecular systems interacting with lasers. Sometimes, these pulses are strong enough to compete with the Coulomb forces in controlling the electron dynamics in atomic systems. As a result, atoms and molecules in intense laser fields exhibit new properties which have been discovered via the study of multiphoton processes. In order to understand the observed effects new theoretical tools beyond the strong field approximation [1–3], which take into account the electron interaction with the laser and the nucleus field on equal footing, are needed.

In recent works [4–6], it was established that a simple theoretical approach called CV2$^-$, which is based on Coulomb–Volkov-type (CV) states, can supply reliable predictions of atomic ionization by extreme ultraviolet laser pulses in the subfemtosecond regime when compared with time-dependent Schrödinger equation simulations (TDSE). When photon energies are lower than the ionization potential it has been introduced a modified form of CV2$^-$ (called MCV2$^-$) that aims at accounting for the new features revealed in the electron spectrum by the above-mentioned TDSE calculation [7]. The MCV2$^-$ theory takes into account a pathway through intermediate bound states within a first Born approximation.

Failures on this theory arise when the field becomes strong or even when the laser frequency is close to a resonant transition irrespective of the strength of the laser field [7,8]. Here we want to improve this theory even further by considering an initial state given by a combination of a finite set of bound-state wave functions with time-dependent coefficients obtained from solving the corresponding close coupling equations. The spirit of the procedure is in the line of the employed by Geltman [9], however, differences arise as [9] is focused on avoiding divergencies in the perturbative expansion due to the resonances. Our approach here continues the effort to improve Coulomb–Volkov basic theory looking for the required changes to provide the theory for a predictive value.

We analyze hydrogen ionization by a few-cycle laser when the laser frequency is close to resonance with the first excitation level $2p_0$. Resonantly enhanced above-threshold ionization REATI [10] has been previously studied considering an infrared 608 nm laser light. In that case the resonant character derived from Stark shift time-dependent resonances, so-called Freeman resonances [11]. We would like here a direct study of REATI by tuning the laser frequency across the 1s–$2p_0$ resonance for the hydrogen atom. For recent work on hydrogen ionization by laser pulses we refer to [12] and references therein. The TDSE results have been obtained with a code described in [13].

In Section 2 the theory is summarized and in Section 3 the results are presented. Conclusions are given in Section 4. Atomic units are used throughout unless otherwise stated.

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2. Theory

We consider here the ionization of a one-active electron atom by an external laser radiation whose electric field is $\mathbf{F}(t)$. Under non-relativistic conditions and within the electric dipole approximation the electron wave function $\Psi(\mathbf{r},t)$ satisfies the length-gauge time-dependent Schrödinger equation:

$$i\frac{d\Psi(\mathbf{r},t)}{dt} = \left[-\frac{1}{2}\nabla^2 + V(r) + \mathbf{r}\cdot\mathbf{F}(\mathbf{t})\right]\Psi(\mathbf{r},t), \tag{1}$$

where $\mathbf{r}$ is the position of the electron with respect to the nucleus identified with the center-of-mass, $\mathbf{F}(t)$ is the laser field in the volume of the atom, $V(r)$ is the interaction between the electron and the rest of the target and $r = |\mathbf{r}|$. The finite pulse length τ is featured by a sine-square envelope:

$$\mathbf{F}(t) = \mathbf{F}_0 \sin(\omega t + \varphi)\sin^2\left(\frac{\pi t}{\tau}\right). \tag{2}$$

The transition amplitude from the state i at $t = 0$ to the state f at $t = \tau$ may be approximated by the prior form of the following variational expression [14]:

$$a_{fi}^- = \lim_{t\to 0}\langle\chi_f^-(t)|\chi_i^+(t)\rangle - i\int_0^\tau dt\left\langle\chi_f^-(t)\left|\overleftarrow{H - i\frac{d}{dt}}\right|\chi_i^+(t)\right\rangle, \tag{3}$$

where the arrow on the left-hand side indicates the state on which the non-hermitian operator applies; $\chi_f^-(t)$ and $\chi_i^+(t)$ are trial functions to the exact solutions of the Eq. (1), subject to the asymptotic conditions:

$$\chi_f^-(t) \underset{t\to\tau}{\to} \phi_f^-(\mathbf{r},t) = \varphi_f^-(\mathbf{r})\exp(-i\epsilon_f t), \tag{4}$$

$$\chi_i^+(t) \underset{t\to 0}{\to} \phi_i^+(\mathbf{r},t) = \varphi_i^+(\mathbf{r})\exp(-i\epsilon_i t), \tag{5}$$

where $\varphi_f^-(\mathbf{r})$ and $\varphi_i^+(\mathbf{r})$ are eigenfunctions of the atomic Hamiltonian associated with the eigenenergies ϵ_f and ϵ_i, respectively. Expression (3) provides exact transition amplitudes when one of the two trial functions are exact solutions of (1). The Coulomb–Volkov wave function is used as the trial wave function [4,5]

$$\chi_f^-(\mathbf{r},t) = \phi_f^-(\mathbf{r},t)\exp\left[i\mathbf{A}^-(t)\cdot\mathbf{r} - i\,\mathbf{k}\cdot\int_\tau^t dt'\mathbf{A}^-(t') - \frac{i}{2}\int_\tau^t dt'\mathbf{A}^{-2}(t')\right], \tag{6}$$

where $\mathbf{A}^-(t) = \int_t^\tau dt'\ \mathbf{F}(t')$ is the vector potential. After an easy algebra using Eq. (6), the expression (3) may be transformed into:

$$a_{fi}^- = \int_0^\tau dt\exp\left\{i\frac{k^2}{2}t + i\,\mathbf{k}\cdot\int_\tau^t dt'\mathbf{A}^-(t') + \frac{i}{2}\int_\tau^t dt'\mathbf{A}^{-2}(t')\right\} \times\int d\mathbf{r}\ \chi_i^+(\mathbf{r},t)\exp[i\mathbf{A}^-(t)\cdot\mathbf{r}]\mathbf{A}^-(t)\cdot[i\mathbf{k} + \nabla]\varphi_f^{-*}(r). \tag{7}$$

The choice of the trial wave function $\chi_i^+(\mathbf{r},t)$ in Eq. (7) is still open. It should account for most of the bound state part of the exact wave function of a given problem. For instance, when both the photon energy is greater than or equal to the ionization potential and the ionization process is not saturated, it looks reasonable to replace $\chi_i^+(\mathbf{r},t)$ by the unperturbed wave function $\phi_i^+(\mathbf{r},t)$. Then the so-called CV2$^-$ approximation is obtained. Electron energy spectra as predicted by CV2− has an excelent agreement with TDSE computations as long as the photon energies are above the Hydrogen ionization potential and the laser intensity is small enough to avoid the saturation regime [4,5]. On the other hand, significant differences showed up for all ATI peaks predicted by CV2$^-$ were smaller than TDSE ones, while TDSE presented a new set of secondary peaks that were unexplained by the CV2$^-$ theory [7]. In fact, due to energy conservation, the main ATI peaks were found at the energies $E_p = \epsilon_i + p\omega$ where p is the number of absorbed photons. In the considered case, the ponderomotive energy, that is the kinetic energy of the electron due to its quiver motion in the oscillatory external field, is negligible because of both the low laser intensity and the high photon frequency. Further, it was noticed that the positions of secondary peaks follow a similar rule, $E_{n,s} = \epsilon_n + s\omega$, where $s > 0$ and $n > 1$ are integers. This suggests that the secondary peaks can be traced back to the absorption of s photon(s) from excited states $n = 2, 3, \ldots$, etc. [15]. This may be physically understood by realizing that a short laser pulse has a broad spectrum, thus making possible to populate a wide range of atomic excited states through the absorption of a single photon. It is worth insisting on the fact that this intermediate transition may occur even though the laser frequency ω is not in tune. Hence, improving the theory CV2$^-$ implies taking into account a pathway through intermediate bound states.

A simple way to improve the CV2$^-$ consists of a different choice for the trial wave function[7]:

$$\chi_i^+(\mathbf{r},t) = \sum_j b_{ji}(t)\phi_j(\mathbf{r},t), \tag{8}$$

where $b_{ji}(t)$ is the transition amplitude at time t from the initial state i to the intermediate state $j = (n, l, m)$. In [7], $b_{ji}(t)$ was estimated by a first Born approximation giving place to the called MCV2$^-$ approximation. In this case, if the laser field is linearly polarized in the z-direction and if the initial state i is the ground state 1s, electric dipole selection rules impose $l = 1$ and $m = 0$. Although MCV2$^-$ allows to describe the position of the emergent secondary peaks, some sizeable quantitative differences with TDSE remains. In this work, we improve MCV2$^-$ further, computing $b_{ji}(t)$ in an exact $N + 1$-states (the initial plus N excited states) close coupling scheme. Thus, the transition amplitude for the new CC–CV2$^-$ theory takes the form:

$$a_{fi}^{\mathrm{CC-CV2^-}} = a_{fi}^{\mathrm{CV2^-}} + \sum_{j>1}^{N}\int_0^\tau dt\ b_{ji}(t)\ \exp\{i(k^2/2 - \epsilon_j)t + i\mathbf{k} \times\int_\tau^t dt'\mathbf{A}^-(t') + i/2\int_\tau^t dt'\mathbf{A}^{-2}(t')\}\mathbf{A}^-(t') \times\int d\mathbf{r}\ \varphi_j(r)\exp[i\mathbf{A}^-(t)\times\mathbf{r}]\mathbf{A}^-(t)\times[i\mathbf{k} + \nabla]\varphi_f^*(r), \tag{9}$$

where j stands now for the principal quantum numbers of the first N bound states. We observe that the first term accounts for the usual ATI peaks as given by the standard theory CV2$^-$, while the second term represents a series of CV2$^-$ amplitudes for transitions starting from intermediate states j. These amplitudes are weighted by the close-coupling transition amplitudes at any time during the laser pulse. Their meaning is quite simple: to undergo a transition into a continuum state, the system can first be excited into an upper bound state through a full although finite basis-set evolution, and then undergo a final multiphoton transition into the final state. The first step, which is described by the close-coupling transition amplitude, is possible, even in non-resonant situations, because the finite duration of the pulse results in a broadening of the laser frequency. In the subsequent applications, we have checked that the convergence is achieved in Eq. (9) for $N < 5$. In fact, secondary peaks for higher intermediate states vanish in the background.

3. Results

In this section, we apply the CC–CV2$^-$ theory to study the ionization of hydrogen atom by a laser with frequencies near the $1s–2p_0$ resonant one. Fig. 1 show the ionization electron spectrum for three different frequencies: (a) below, (b) at and (c) above the resonant $\omega_0 = 0.375$ a.u. case. In both non-resonant cases, the simple CV2$^-$ roughly agree in the background with the more elaborated theories MCV2$^-$, CC–CV2$^-$ and with the TDSE