Above threshold ionization of Argon atoms by multicolor XUV radiation

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Abstract. We analyse theoretically the Argon photoelectron spectra produced by strong and extreme ultraviolet radiation of six colors: from the 11\textsuperscript{th} to the 16\textsuperscript{th} harmonics of $\omega_0$ (800 nm). In particular we concentrate in the range of the spectra where absorption of two photons occurs. The combination of photons of different frequencies results in eleven peaks that are separated by $\omega_0$. We point out that their relative intensities are very sensitive to the laser pulse parameters and target description. We also compare the theoretical description with experimental results finding good qualitative agreement.

1. Introduction  
High Harmonic Generation (HHG) is one of the most interesting phenomena discovered for intense lasers interacting with matter. Since the first observation of the surprising intensity plateau for a wide range of harmonic orders, great progress has been made concerning the understanding and control of the process. Nowadays, the HHG radiation is commonly used as a light-source due to its attractive properties of coherence, very short duration (femto or attoseconds), high intensities and frequencies (see for example [1, 2, 3]). These characteristics of the radiation are necessary for initiating and probing ultrafast electron motion in matter.

In a recent experiment the energy-resolved photoelectron (PE) spectra produced via extreme-ultraviolet-(XUV)-photon above-threshold ionization (ATI) of argon were observed [2]. The XUV radiation consisted of higher-order harmonics (11\textsuperscript{th} to 16\textsuperscript{th}) of the original laser $\omega_0$ ($\lambda = 800$ nm) generated by the process of the relativistic oscillating mirror (so-called ROM) in high-peak-power laser-pulse interaction with solid targets. In Ref. [2] it was possible to measure the ATI peaks in the PE spectra corresponding to different combinations of two harmonic photons leading to the same excess energy.

The theoretical description of this type of process with few-cycles pulses is commonly obtained neglecting the multielectronic nature of the target. For example, the numerical solution of the Time Dependent Schröedinger Equation (TDSE) is mostly performed in the single active electron approximation, also the Strong Field Approximation (SFA) neglects the interaction of the emitted electron with the residual multielectronic target [4]. Alternatively, in the Coulomb-Volkov approach (CV) [6] it is possible to include this interaction in the final state. Additionally, when multiphotonic regime is reached the PE spectra can be factored into two contributions separating the roles of the laser pulse and the parent ion. This approximation, which is based on
the CV approach is here called DipA [5]. The yield factorization allows for the study of complex atoms or molecules since the target contribution, that is proportional to the monochromatic one-photon cross section, can be computed accurately independently of the laser-matter process involved.

In this work we study the above-threshold ionization (ATI) of argon atoms in the context of the DipA approximation and use approximately the same light-pulse parameters as the experiment [2]. Atomic units are employed, except where otherwise stated.

2. Theory

In this section we summarize the CV and DipA approximations [5, 6]. The transition matrix in length gauge reads:

\[ T_{fi} = -i \int_0^\tau \langle \Psi'_f(t) | \mathbf{F}(t) \cdot \mathbf{r} | \phi_i(t) \rangle \, dt \]  

where \( \Psi'_f(t) = \phi'_f(r) \exp[-i(S(k, t) - \mathbf{A}(t) \cdot \mathbf{r} + E_i t)] \) is the CV wave function for the final electronic state with ingoing boundary conditions, \( \phi_i(t) \) is the asymptotic target wave function with energy \( E_i \) in absence of external fields and \( S \) is part of the Volkov phase: \( S(k, t) = k \int_0^t \mathbf{A}(t') dt' + \frac{1}{2} \int_0^t \mathbf{A}^2(t') dt' \). The ejected electron has momentum \( k \equiv (k, \Omega) \) corresponding to an energy of \( E_f = k^2/2 \).

The vector potential is obtained from the electric field of the ionizing radiation pulse as \( \mathbf{A}(t) = -\int_0^t \mathbf{F}(t') dt' \). In the present work we consider the sum of the 11th to the 16th harmonics of \( \omega_0 \) (\( \lambda = 800 \text{ nm} \)), each with an amplitude \( F_n \), a delay \( t_n \) and a duration \( \tau_n \):

\[ \mathbf{F}(t) = \hat{\varepsilon} \sum_{n=1}^{16} F_n \cos(n\omega_0(t - t_n - \tau_n/2)) \sin^2(\pi(t - t_n)\tau_n). \]  

Each contribution vanishes outside the time interval \( t_n < t < \tau_n \) and has its maximum amplitude at the middle of the symmetric pulse. The linear polarization is given by the vector \( \hat{\varepsilon} \). The duration \( \tau \) of all the pulse is defined as the mayor value of \( \tau_n + t_n \). For a coherent pulse we fix \( t_n = 0, \tau_n = \tau \) and \( F_n = F_0 \) for all \( n \).

The differential ionisation probability in energy is obtained from the transition matrix magnitude as

\[ \frac{dP^{\text{CV}}}{dE} = k \int d\Omega |T_{fi}|^2 \]  

In the high frequency regime \( A \sim F_0/\omega \) is small, therefore the term \( \mathbf{A}(t) \cdot \mathbf{r} \) in the Volkov phase can be neglected given rise to the DipA approximation. In this case temporal and radial integrals are decoupled leading to \( T \simeq \mathbf{L}(k) \cdot \mathbf{M}(k) \). Here \( \mathbf{L} = \langle \varphi'_f | \mathbf{r} | \varphi_i \rangle \) depends only on the target structure. Then, the approximate spectra can be obtained as:

\[ \frac{dP^{\text{DipA}}}{dE} = k \int d\Omega |\mathbf{L}(k)|^2 |\mathbf{M}(k)|^2 \]  

both terms inside the integral depend on the electronic momentum, however an additional approximation can be made to achieve total decoupling (DipA2); that is to average over the angular dependence of the factor \( \mathbf{M} \):

\[ \frac{dP^{\text{DipA}2}}{dE} = k \int d\Omega |\mathbf{L}(k)|^2 \frac{1}{4\pi} \int d\Omega |\mathbf{M}(k)|^2 \]  

Finally, the contribution that accounts for the effect of the electromagnetic field on the free-electron final state is:

\[ \mathbf{M}(k) = \int_0^\tau dt \mathbf{F}(t) e^{iS(k, t)} e^{i\omega_f t} \]
The square of the dipole matrix element, $|L(k)|^2$ in Eq. (5), which depends on the atomic structure is calculated within the Random-Phase approximation, as implemented in Ref. [7]. This approximation, which includes certain classes of many-body effects such as target polarization and ground state correlation, is known to reproduce the main feature of the one-photon cross section for rare gas atoms.

3. Results

3.1. Checking CV, DipA and DipA2 approximations

In a previous work the accuracy of the CV description was analyzed by comparison with TDSE results for ionization of H(1s) [5]. In fact the DipA-approximation reproduces exactly the ionization spectra in the region where the one-photon process is dominant (first ATI peak), and therefore also describes accurately the total rate. However, it underestimates the high-order ATI peaks.

For the present study $A \sim F_0/\omega \ll 1$ and DipA should be applicable. To check this, we present in Fig. 1 the PE spectra for ionization of H from the $3p_0$ orbital with an effective charge $Z_{\text{eff}}$ (producing the correct Argon ionization potential). We compare the CV, DipA and DipA2 results. DipA and DipA2 reproduce exactly the first CV-ATI peaks and underestimate with one and two order of magnitude the 2nd and 3rd ATI peaks respectively. Despite this, the shape of the spectra remains unchanged: when we add an arbitrary factor in each region to fit the spectra, the CV results are exactly reproduced. Thus, we expect that both DipA and DipA2 should yield reasonable spectra, although lacking the correct relative normalization for the different multi-photon processes.

![Figure 1. Comparison of CV, DipA and DipA2 spectra for H(3p₀) ionization due to a coherent laser pulse eq. (2) with $\omega_0$ corresponding to 800nm, $F_n = 0.01$ and $\tau = 8\text{fs}$. The frequencies varies from $11\omega_0 = 0.63$ to $16\omega_0 = 0.91$ a.u. An effective charge $Z_{\text{eff}} = 3.23$ a.u. was used. Three regions can be distinguished: from 0 to 10eV, 15 to 35eV and 35 to 55eV, corresponding to process of absorption of only one, two and three photons respectively.](image)

3.2. Argon spectra

In Fig. 2 we present the Argon spectra obtained with same conditions as above. We show in colors the ionization spectra for each harmonic. The first six peaks are positioned at $E = n\omega_0 - 15.76\text{eV}$ corresponding to the absorption of only one photon of frequency $n\omega_0$. The first ionization potential of Argon is 15.76eV. As explained in [2], the first peak with energies ranging from 22 to 32 $\omega_0$ correspond to absorption of two photons, $E_{nn'} = n\omega_0 + n'\omega_0 - 15.76\text{eV}$ where different combinations of $n$ and $n'$ lead to the same excess energy, thus contributing to the same ATI peak. In the right figure we have plotted this part of the spectra in linear scale. The results are presented in the DipA approach for H-like (as in previous section) and Argon. The two spectra show clearly different relative heights of the peaks, showing the sensitivity to the target description. On the other hand, DipA2 and DipA differ only with a global factor. This indicates that the laser field couples uniformly with electrons ejected in different angles.
The Argon spectrum, (red line in the right panel of Fig 2) qualitatively agrees with the experimental result of Ref. [2].

4. Conclusions
In the present work we analyze theoretically the argon PE spectra, using experimental light-pulse parameters. Due to the high frequency of the multicolor XUV radiation, the DipA approximation in the context of Coulomb Volkov approach should be adequate. We have shown that the applicability of DipA and DipA2 is well justified. These approximations allow to describe the spectra as the contribution of two factors: one accounts for the target information and the other for the electron-laser interaction. In the context of DipA approximation we have calculated the Argon spectra, in particular in the range where absorption of two photons occurs. The combination of photons of different frequencies results in eleven peaks that are separated by $\omega_0$. We observe a variation in the relative intensity of these peaks depending on the description of the target. The theoretical description shows good qualitative agreement with recent experimental results for two-extreme-ultraviolet-photon above-threshold ionization of Argon atoms.

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