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Multilevel model of multiphoton processes in a helium atom in a strong laser field: ionization description

© I.R. Khairulin[1], V.A. Antonov[1], M.Yu. Emelin[1], M.M. Popova[1,2,3], E.V. Gryzlova[1,3], M.Yu. Ryabikin[1]

[1] Institute of Applied Physics, Russian Academy of Sciences,
603950 Nizhny Novgorod, Russia
[2] Department of Physics, Lomonosov Moscow State University,
119991 Moscow, Russia
[3] Skobeltsyn Institute of Nuclear Physics,
Lomonosov Moscow State University,
119991 Moscow, Russia

e-mail: khairulinir@ipfran.ru

Received November 29, 2022
Revised December 30, 2022
Accepted January 28, 2023

A multilevel model that makes it possible to describe multiphoton processes taking into account ionization in a multielectron atom irradiated by an intense laser field is proposed. Using the He atom as an example, it is shown that this model reproduces the main regularities of multiphoton ionization of an atom by an intense high-frequency laser field.

Keywords: atom in a strong field, high-order harmonic generation, multiphoton processes, atom ionization.

DOI: 10.61011/EOS.2023.02.55772.5-23

Introduction

The appearance of sources of coherent radiation in vacuum ultraviolet and X-ray ranges based on high-order harmonic generation (HHG) of the laser field in gas media in tunnel ionization regime results in the emergence of attosecond physics — a field of science dedicated to the investigation and control of physical and chemical processes in atoms, molecules, nanostructures and solids on attosecond time scales [1].

The first HHG experiments were performed on noble gases and established the main properties of this process [2]. A three-step semiclassical rescattering model [3] became the basis of a qualitative understanding and theoretical description of the HHG process on a microscopic scale. Quantum-mechanical analogs of this model [4,5] made it possible to provide an analytical description of the process in the tunnel and above-barrier ionization regimes. One of the distinguishing features of these HHG regimes is that in these regimes the dynamics of the released electron in the continuum plays a leading role. At the same time, since, in terms of quantum mechanics, the high-frequency nonlinear polarization responsible for HHG is induced due to the interference of the wave function components corresponding to the released and bound electrons, the wave function features of the bound valence electron play an important role, which is used in HHG spectroscopy of atoms and molecules [6]. Moreover, it was found that the efficiency of HHG in a moderate-intensity laser pulse can increase dramatically in the presence of resonances of the generated harmonics with transitions between certain excited atomic states and the ground state [7]. However, HHG description in multiphoton and intermediate ionization regimes is a complicated task, since the approximations used for the HHG description in tunnel and above-barrier regimes become inapplicable. In this case, it is important to accurately take into account the real structure and properties of atomic states which differ greatly for different atoms.

In the present paper, we propose a multilevel model allowing to describe multiphoton processes in an atom taking into account ionization of its bound states.

Theoretical model

Consider a He atom irradiated by an intense laser pulse linearly polarized along the z axis, whose electric field has the form

$$\mathbf{E}(t) = \mathbf{z}_0 E(t) = \mathbf{z}_0 E_L f(t) \sin(\Omega_L t), \qquad (1)$$

where E_L and Ω_L are the amplitude and carrier frequency of the laser pulse, $f(t)$ is the laser pulse envelope. The evolution of the electron state in a He atom, $|\Psi\rangle$, in the external laser field is described by the time-dependent Schrödinger equation (hereinafter, atomic units are used):

$$i \frac{\partial}{\partial t} |\Psi\rangle = [\hat{H}_0 + \hat{V}]|\Psi\rangle, \qquad (2)$$

where $\hat{H}_0$ is the He atom Hamiltonian in the absence of interaction with the laser field (1), $\hat{V}$ is the operator of interaction between the atom and laser field, which in the electric dipole approximation has the form

$$\hat{V} = \hat{d}_z E_L f(t) \sin(\Omega_L t),$$

where $\hat{d}_z$ is the projection of the atomic dipole moment operator on the z axis. The electron wave function in He can be represented as an expansion in a finite number of bound two-electron states $|k\rangle$ with energies E_k and two-electron continuum states $|\varepsilon, l\rangle$ characterized by energy $\varepsilon > 0$ and orbital quantum number l:

$$|\Psi\rangle = \sum_{k=1}^{K} a_k(t)|k\rangle + \sum_{l=0}^{\infty} \int_0^{\infty} d\varepsilon b_l(\varepsilon, t)|\varepsilon, l\rangle, \qquad (3)$$

where $a_k(t)$ and $b_l(\varepsilon, t)$ are the excitation amplitudes of the states $|k\rangle$ and $|\varepsilon, l\rangle$, respectively, K is the number of bound states taken into account. In this case, the set of bound states is determined by the necessity to take into account the most probable paths of multiphoton atom excitation, while the continuum states allow to take into account the atom ionization. It should be noted that, since He atom experiences a dipole interaction with the linearly polarized field (1), the magnetic quantum number of atom states M does not change. Thus, if at the initial moment of time the atom was in a state with zero magnetic quantum number, for example, in the ground state of He atom $|1s^2\rangle$, then the magnetic quantum number of each state in (3) will be equal to zero. It is worth noting that the number of bound states in (3) can be arbitrary and, in certain calculations, is determined by the existing data on the energy structure of an atom (generally, a multielectron one), as well as by the available computational capabilities. However, the number of bound states taken into account determines the maximum order of multiphoton processes that can be correctly described within the model: generally, the maximum order does not exceed $K - 1$. As an example below, we will consider the first six bound states of the He atom ($K = 6$) with $M = 0$, namely $|1\rangle = |1s^2\rangle$, $|2\rangle = |1s2s\rangle$, $|3\rangle = |1s2p\rangle$, $|4\rangle = |1s3s\rangle$, $|5\rangle = |1s3p\rangle$, $|6\rangle = |1s3d\rangle$, using the spectroscopy data on the He atom (energies E_k and dipole moments of transitions between the bound states, as well as between the bound and continuum states) obtained using MCHF software package based on the solution of the stationary Schrödinger equation by the multiconfiguration Hartree-Fock method [8]. In this case, only states with energies below 1 a. u., that correspond to the detachment of one electron, are taken into account in the continuum. Thus, the set of two-electron states of an atom used in the calculations below allows to correctly describe the processes with excitation and/or ionization of one electron.

By substituting (3) into (2) and calculating scalar products of the derived expression and vectors of states $|k\rangle$ and $|\varepsilon, l\rangle$ step by step, one can obtain a system of equations for the amplitudes $a_k(t)$ and $b_l(\varepsilon, t)$, which fully describes the dynamics of the atomic state in an external field. When describing multiphoton processes primarily caused by transitions between the bound atomic states, this system can be simplified using the approximation of adiabatic elimination of the continuum states [9]. This allows to switch from the complete system of equations for $a_k(t)$ and $b_l(\varepsilon, t)$ to a closed system of equations for Fourier components of excitation amplitudes $a_{k,n}(t)$, which are defined as follows

$$a_k(t) = \sum_{n=-\infty}^{\infty} a_{k,n}(t) e^{in\Omega_L t}.$$

By neglecting additionally the connection between the bound states through the continuum states, in other words, by neglecting the influence of bound-free-bound transitions (i.e., recombination) on the wave function evolution, the following system of equations for $a_{k,n}(t)$ can be obtained:

$$\frac{da_{k,n}}{dt} = -i[E_k + n\Omega_L - iw_{k,n}(t)]a_{k,n} - \sum_{k'} \left[\frac{E_L d_{z,k'k}}{2} f(t) a_{k',n-1} - \frac{E_L d_{z,k'k}}{2} f(t) a_{k',n+1} \right], \qquad (4)$$

where $d_{z,k'k} = \langle k|\hat{d}_z|k'\rangle$, $w_{k,n}(t)$ is the ionization rate of the nth Fourier component of the state $|k\rangle$:

$$w_{k,n}(t) = \frac{\pi}{4} E_L^2 f^2(t) \big[D_{kk}(E_k - (n-1)\Omega_L)\theta(E_k - (n-1)\Omega_L) + D_{kk}(E_k - (n+1)\Omega_L)\theta(E_k - (n+1)\Omega_L) \big], \qquad (5)$$

where $\theta(x)$ is the Heaviside step function,

$$D_{kk}(\varepsilon) = \sum_l \left| \langle \varepsilon, l|\hat{d}_z|k\rangle \right|^2.$$

In (4) summation over k' is performed over the entire set K of bound states into which dipole transition is allowed, and $n = [-\infty; \infty]$. In addition, when calculating the ionization rate (5), principal value integrals over energy ε were calculated due to which Heaviside step functions occurred.

To solve system (4), it is necessary to determine the initial conditions for $a_{k,n}$. Since initially only excitation amplitudes of bound states $a_k(t = 0) = a_k^{(0)}$ are determined, there is uncertainty in the choice of initial conditions for the Fourier components. Below, we assume, that only Fourier components of amplitudes with $n = 0$ are non-zero in the initial moment of time:

$$a_{k,n}(t = 0) = a_k^{(0)} \delta_{n0}, \qquad (6)$$

where δ_{n0} is the Kronecker symbol. The system (4) together with the initial conditions (6) allows to describe intraatomic dynamics excited by the laser field (1) taking into account the atom ionization. Ionization in this model is taken into account by the appearance of decay rates $w_{k,n}(t)$ of the Fourier components of the bound state excitation amplitudes. In this case, ionization may be represented as an „irreversible flow“ of an electron wave packet during the multiphoton excitation into some „reservoir“ (into the continuum).

Comparison with numerical solution of the Schrödinger equation

To test the method of ionization accounting in the above model, we have performed calculations for the He atom on the basis of the numerical solution of the time-dependent Schrödinger equation:

$$i\frac{\partial}{\partial t}|\Psi(x,z,t)\rangle = \left[-\frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{1}{2}\frac{\partial^2}{\partial z^2} + \frac{i}{c}A_z(t)\frac{\partial}{\partial z} + U(x,z)\right]|\Psi(x,z,t)\rangle, \qquad (7)$$

where

$$A_z(t) = c\int\limits_{-\infty}^{t} E(t')dt'$$

is the vector potential of the field (1), c is the speed of light. In these calculations, the He atom was described by means of a 2D single-electron model with the effective potential allowing to reproduce the energies of the first three states of real He atom with zero magnetic quantum number ($|1\rangle$, $|2\rangle$, $|3\rangle$):

$$U(x,z) = U(r) = -\frac{1+(1+8.125r)e^{-8.125r}}{\sqrt{r^2+0.01}} + \frac{0.6r^6}{r^8+10^{-4}}. \qquad (8)$$

Equation (7) with potential (8) was solved by the split-step method with fast Fourier transform [10].

In order to exclude the influence of multiphoton resonant excitation of an atom during its ionization and, thus, to carry out direct comparative study of atom ionization from the selected bound state, we consider the case of a nonresonant laser field whose frequency is not in resonance of any multiplicity with the transitions between the ground and excited states of the He atom: $\Omega_L = 0.33$ a.u. ($\lambda_L = 138$ nm). As a field envelope (1), $f(t)$, we consider a trapezoidal envelope with full duration of 36 field periods $T_L = 2\pi/\Omega_L$ and with uniform turn on and turn off during $3T_L$ (Figure 1, right y-axis).

Figure 1 (left y-axis) shows a typical time dependence of the average number of electrons $N(t)$ in a He atom irradiated by the laser field (1) with the peak intensity $I_L = 2.86\cdot10^{-3}$ a.u. (10^{14} W/cm^2, the Keldysh parameter $\gamma = 16.8$). It was assumed that at the initial moment of time the atom was in $|2\rangle$ state. In the quantum-mechanical calculation based on (7), (8) $N(t)$ is calculated as follows:

$$N(t) = \iint\limits_R dxdz|\Psi(x,z,t)|^2 + 1 \equiv N_{2D}(t), \qquad (9)$$

where integration is performed inside the circle with $R = 20$ a.u. around the He atom. In this case, the wave functions of the first three stationary states of an active electron in problem (7), (8) fall within the integration circle. Therefore, the first term in (9) corresponds to the atoms whose active electron is not detached when interacting with

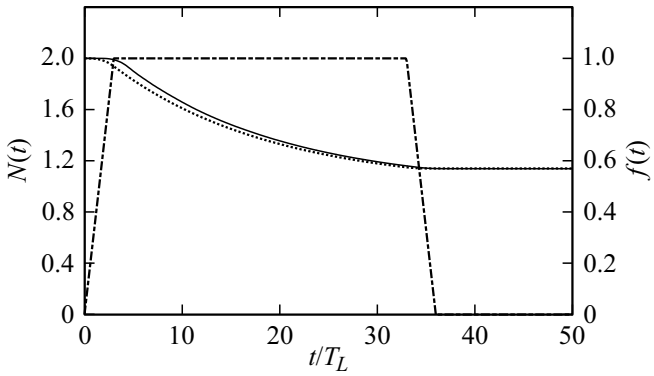

Figure 1. Time dependence of the average number of electrons in He atom (left y-axis, solid and dotted lines), as well as the shape of the field envelope (1) (right y-axis, dash-dotted line). The solid line corresponds to the numerical solution of equation (7) with potential (8) and the initial condition $|\Psi(x,z,t=0)\rangle = |2\rangle$, the dashed line corresponds to the solution of the system (4), (5) with initial conditions (6) with $a_2^{(0)} = 1$ and $a_k^{(0)} = 0$ for $k \neq 2$. The figure is plotted with $\Omega_L = 0.33$ a.u. ($\lambda_L = 138$ nm) and $I_L = 2.86\cdot10^{-3}$ a.u. (10^{14} W/cm^2, the Keldysh parameter $\gamma = 16.8$).

the field (1) at time t, while the second term corresponds to the second „frozen" electron that produces an effective potential (8) together with the nucleus. In the proposed model (4), (5) $N(t)$ corresponds to the following value:

$$N(t) = 2\sum_{k=1}^{K}|a_k(t)|^2 + \sum_{l=0}^{\infty}\int\limits_0^{\infty} d\varepsilon|b_l(\varepsilon,t)|^2 = \sum_{k=1}^{K}|a_k(t)|^2 + 1 \equiv N_{ML}(t), \qquad (10)$$

where the first term in the first equality corresponds to the probability of detecting two electrons in the He atom and the second term corresponds to the probability of detecting one electron in the He atom; condition of normalization of wave function (3) is also taken into account in the second equality:

$$\sum_{k=1}^{K}|a_k(t)|^2 + \sum_{l=0}^{\infty}\int\limits_0^{\infty} d\varepsilon|b_l(\varepsilon,t)|^2 = 1. \qquad (11)$$

Figure 1 shows that the time dependence $N_{ML}(t)$ (dotted line) is qualitatively and quantitatively close to $N_{2D}(t)$ (solid line): at the initial moments of time at the leading edge of the pulse (1), the average number of electrons in the He atom is close to two; then in the time interval with a constant field intensity (1), a smooth decrease in $N(t)$ is observed due to multiphoton ionization of the atom (the Keldysh parameter $\gamma = 16.8$); after the end of the pulse, the average number of electrons in the atom remains constant. At the same time, at the leading edge of the laser pulse, $N_{ML}(t)$ decreases slightly faster than $N_{2D}(t)$, which is caused by a

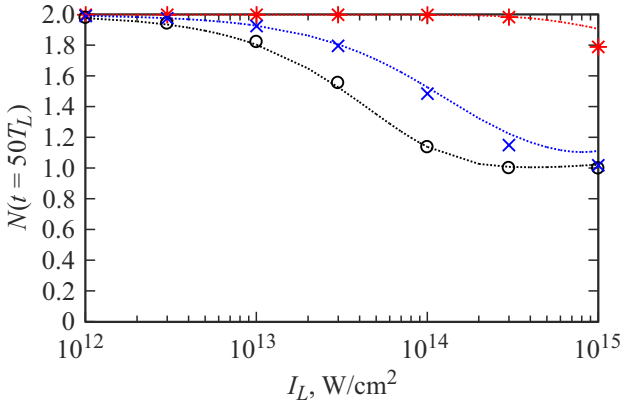

Figure 2. Dependence of the average number of electrons in He atom after the impact of the pulse (1) (at time $t = 50T_L$) on its peak intensity. The cases, when initially the atom is in the state $|1\rangle$ (red line), $|2\rangle$ (black line) or $|3\rangle$ (blue line), are shown. Dotted lines correspond to the data obtained on the basis of the solution of the system (4), (5), and markers (asterisks, crosses and circles) — on the basis of the numerical solution of the Schrödinger equation (7) with potential (8) with the appropriate initial conditions. The Keldysh parameter varies from 168 (at 10^{12} W/cm^2) to 5.31 (at 10^{15} W/cm^2).

delay in numerical calculations required for the free part of the wave packet to leave the integration circle. With further increase in t, the decay rate of $N_{2D}(t)$ tends to the decay rate of $N_{ML}(t)$, and then exceeds it. As a result, for a sufficiently long field pulse (1), starting from a certain moment of time in a range of constant intensity, $N_{ML}(t)$ becomes greater than $N_{2D}(t)$, and with further increase t the difference between $N_{ML}(t)$ and $N_{2D}(t)$ will increase. Such behavior of the dependences $N_{ML}(t)$ and $N_{2D}(t)$ is caused both by the approximations used in the derivation of the system (4), (5) and by the approximations made in the numerical solution of the Schrödinger equation (7), (8). However, for the pulse width shown in Figure 1, the initial difference between $N_{ML}(t)$ and $N_{2D}(t)$ and their decay rates is so small that after the end of the field pulse (1), for example, at $t = 50T_L$, $N_{2D}(t) \simeq N_{ML}(t)$.

Figure 2 shows the dependences of the average number of electrons in the He atom after the end of the pulse (at $t = 50T_L$) on the intensity of the field (1) with frequency $\Omega_L = 0.33$ a. u. for different initial states of the atom calculated on the basis of the numerical solution of equation (7) with potential (8) (colored markers), as well as on the basis of the proposed model (4), (5) (dotted lines). As it can be seen, in the considered range of laser field intensities, in which one electron participates in the process of ionization of the He atom [11], a good quantitative agreement is observed between solutions obtained from the model (4), (5) and numerical solution of the Schrödinger equation, which indicates the reasonableness of the assumptions used in the derivation of the system (4), (5).

Conclusion

In the present work, we proposed a multilevel model allowing to describe multiphoton processes, including resonant ones (for example, HHG with photon energies less than or on the order of the ionization potential) in an atom irradiated by a strong laser field taking into account the depletion of bound states in the multiphoton ionization regime. The description of multiphoton processes in this model is reduced to solving a linear system of equations for the Fourier components of the bound state excitation amplitudes, each of which is characterized by its own ionization rate. In this case, ionization can be represented as a process of „irreversible flow“ of an electron wave packet into continuum during multiphoton excitation. Using a He atom as an example, by comparing with the numerical solution of the time-dependent Schrödinger equation with an effective potential, which allows to reproduce the structure of low-lying atom states, it is shown that the proposed model describes the main regularities of the multiphoton ionization of an atom in a strong laser field. An important feature of the proposed model is the possibility of its application for calculations of the internal dynamic of multielectron atoms (provided that the bound state properties are known — binding energies and dipole moments of transitions), which is a computationally difficult task when using other approaches..

Funding

This work was supported by the Russian Science Foundation, grant № 22-12-00389.

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by E.Ilyinskaya