Many-electron effects in secondary radiation generation during the interaction of atoms with intense laser pulses

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Abstract. A parallel program has been developed for numerically simulating the interaction of intense laser pulses with many-electron atoms based on non-stationary Kohn–Sham equations. With the use of this program, we calculate the nonlinear electron currents excited during ionization of argon atoms by intense two-color laser pulses. Based on the comparison with the solution of the time-dependent Schrödinger calculations for hydrogen atoms, the importance of taking into account many-electron effects in simulating the generation of secondary radiation in atomic gases under the action of ultrashort laser pulses is shown.

1. Introduction

The interaction of an intense laser field with atoms and molecules is accompanied by different phenomena that are interesting from a scientific and applied point of view. These phenomena include, in particular, high-order harmonic generation, which is the result of the acceleration of freed electrons and their recollisions with the parent ions [1]. The excited electron currents can also contain a low-frequency component responsible for the generation of radiation with a frequency much lower than the optical one, in particular, in the terahertz and mid-ir ranges [2–6]. In the latter case, to achieve high generation efficiency, it is necessary that the laser pulse is extremely short [7, 8] or contains several components with a frequency ratio close to a rational fraction with an odd sum of the numerator and denominator [5].

Although most of the atoms and molecules are essentially many-electron quantum systems, numerical studies of the above-mentioned ionization-induced phenomena are traditionally based on the single-active-electron approximation [1, 4–6, 8–13]. Within the framework of this approximation, all the electrons except one are frozen in their orbitals, and the field of the parent ion is described by a static potential well. However, among neutral atoms, this approach is directly applicable only to a hydrogen atom. In this paper, we demonstrate the importance of taking into account many-electron dynamics when calculating electron currents responsible for the generation of secondary radiation during gas ionization by intense laser pulses. For this purpose, we develop computer code for numerical simulation of many-electron dynamics based
on the time-dependent Kohn–Sham equations, in which the Hamiltonian for individual orbitals includes the interaction with the atomic nucleus, electron–electron Hartree and exchange-correlation interaction, as well as interaction with the electric field of the laser pulse [14]. Particularly, we show that for an argon atom, which has a large number of electrons in the outer shell, the ionization lasts significantly longer than for hydrogen atom, which leads to a significantly higher spectral intensity of both the low-frequency and high-frequency electron currents. An interpretation of this effect is given taking into account the difference in ionization potentials and the influence of the dynamic polarization of the atom on the probability of transition of electrons to the continuum.

2. Statement of the problem

Suppose that a many-electron atom interacts with a linearly polarized laser-pulse electric field of strength \( \mathbf{E}(t) = z \mathbf{E}(t) \). We assume that gas density is small enough to neglect the interaction between neighboring atoms. The intensity and wavelength of the laser pulse correspond to the dipole approximation in which the action of the magnetic field in the calculation of the atom dynamics can be neglected [9]. We also limit ourselves here to the consideration of atoms in which orbitals initially are occupied by electrons of the opposite spin. Due to the weak influence of the magnetic field, the zero spin polarization is conserved during interaction with the laser pulse and the time-dependent Kohn–Sham equations describing the dynamics of a many-electron system is written down as follows (atomic units (a.u.) are used throughout this paper, unless specified otherwise):

\[
\frac{i}{\hbar} \frac{\partial \psi_n(\mathbf{r}, t)}{\partial t} = \hat{H}\psi_n(\mathbf{r}, t), \quad n = 1, \ldots, N/2, \tag{1}
\]

\[
\hat{H} = -\frac{1}{2} \nabla^2 - \frac{Z}{r} + z\mathbf{E}(t) + V_{ee}[\rho(\mathbf{r}, t)]. \tag{2}
\]

Here \( \psi_n \) is the wave function of the \( n \)-th orbital of Kohn–Sham; \( Z \) is the nuclear charge; \( N \) is the number of electrons, which is even for non-spin polarized atoms; \( \rho \) is the electron density; \( V_{ee}[\rho(\mathbf{r}, t)] \) is the potential of electron–electron interaction. The electron density is related to the Kohn–Sham orbitals by equality

\[
\rho(\mathbf{r}, t) = 2 \sum_{n=1}^{N/2} |\psi_n(\mathbf{r}, t)|^2. \tag{3}
\]

The electron–electron interaction potential consists of the Hartree potential

\[
V_{H}[\rho(\mathbf{r}, t)] = \int \frac{d^3 \mathbf{r}'}{r - \mathbf{r}'} \frac{\rho(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|} \tag{4}
\]

describing the electron repulsion in the framework of the mean field, and the exchange-correlation potential \( V_{xc} \) for which the spin-unpolarized form of LB94 approximation [15] is used:

\[
V_{xc}^{LB94}[\rho(\mathbf{r}, t)] = V_{xc}^{LDA}[\rho(\mathbf{r}, t)] = \frac{2^{1/3} \beta \chi(\mathbf{r}, t) \rho^{1/3}(\mathbf{r}, t)}{1 + 3 \times 2^{1/3} \beta \chi(\mathbf{r}, t) \sinh^{-1}
\left[2^{1/3} \beta \chi(\mathbf{r}, t)\right]. \tag{5}
\]

Here \( V_{xc}^{LDA}[\rho(\mathbf{r}, t)] \) is the exchange-correlation potential in the local density approximation [14] and \( \chi(\mathbf{r}, t) = [\nabla \rho(\mathbf{r}, t)]/\rho^{1/3}(\mathbf{r}, t) \). The initial condition at time moment \( t = 0 \) corresponds to the ground state of the atom, which is described by the stationary Kohn–Sham orbitals \( \psi_{n,0}(\mathbf{r}) \):

\[
\hat{H}_0 \psi_{n,0}(\mathbf{r}) = E_n \psi_{n,0}, \quad \hat{H}_0 = -\frac{1}{2} \nabla^2 - \frac{Z}{r} + V_{ee}[\rho_0(\mathbf{r})], \tag{6}
\]

where \( E_n \) is the energy of the \( n \)-th orbital, \( \rho_0(\mathbf{r}) = 2 \sum_{n=1}^{N/2} |\psi_{n,0}(\mathbf{r})|^2 \) is the initial electron density. Since gas consisting of non-interacting atoms is considered, equations (2) are complemented by...
standard boundary conditions for $\psi_n$ at infinity, which follow from the finiteness of the integral of $|\psi_n|^2$ and $|\nabla \psi_n|^2$ over the entire space. In numerical calculations, the spatial domain is finite; therefore, it is impossible to satisfy the mathematical boundary conditions directly. To find the approximate solution near the nucleus, we introduce an absorbing layer of width $l_{abs}$ near the grid boundary to suppress the reflected waves. The absorbing layer is set using the multihump imaginary potential method [16], which provides a wide range of effectively absorbed de Broglie wavelengths $\lesssim l_{abs}$. To achieve the convergence of calculations, the spatial domain boundary must be set at a distance $r_{max} > r_{ret} + l_{abs}$ from nucleus, where $r_{ret}$ is the maximum deviation of returning electrons. In further numerical calculations we use $r_{max} = 250$ a.u. and $l_{abs} = 50$ a.u.

The time-dependent dipole acceleration of the atomic system can be found as [17]

$$a(t) = -E(t)N - Z \int d^3r \frac{r}{r^3} \rho(r, t).$$

(7)

Then the macroscopic electron current density in the produced plasma is $j(t) = -N_0 \int_0^t a(t')dt'$, where $N_0$ is the gas density before the start of the ionization process. To study the temporal dynamics of ionization, we calculate the concentration of electrons normalized to gas density in outside of the sphere of radius $r_0 = 10$ a.u.:

$$n_{free}(t) = N - \int_{r < r_0} \rho(r, t) d^3r.$$ (8)

The radius $r_0$ is chosen large enough to exclude most populated excited bound states therefore $n_{free}$ estimates free electrons concentration.

For the numerical solution of the time-dependent Kohn–Sham equations, orbitals $\psi_n(r, t)$ are decomposed into spherical harmonics

$$Y_{lm}(\theta, \varphi) = \frac{\sqrt{(2l + 1)(l - m)!}}{4\pi (l + m)!} P_l^m(\cos \theta) e^{im\varphi},$$

(9)

where $l$ and $m$ are the orbital and magnetic quantum numbers, respectively; $P_l^m(\cos \theta)$ are the associated Legendre polynomials; $\theta$ is the polar angle with respect to $z$-axis directed along the external field; $\varphi$ is the azimuthal angle. The initial conditions for the Kohn–Sham orbitals correspond to the electronic configuration of the atom, which determines the set of quantum numbers $l_n$ and $m_n$, and are given in the form

$$\psi_{n,0}(r, \theta, \varphi) = \frac{1}{r} \Psi_{n,0}(r) Y_{l_n m_n}(\theta, \varphi),$$

(10)

where $\Psi_{n,0}(r)$ describes the radial part of the $n$-th orbital. Since the external electric field has linear polarization, the azimuthal quantum number $m_n$ of each $n$-th orbitals is conserved in time [11], therefore in the expansion of orbitals $\psi_n(r, t)$ over spherical harmonics $Y_{lm}(\theta, \varphi)$ only the summands with $m = m_n$ exist:

$$\psi_n(r, \theta, \varphi, t) = \frac{1}{r} \sum_{l=0}^{\infty} \Psi_{nl}(r, t) Y_{lm_n}(\theta, \varphi).$$

(11)

The potential of the electron–electron interaction $V_{ee}$ is expanded in spherical harmonics up to a quadrupole term:

$$V_{ee}(t, r, \theta) \approx V_{ee}^{(0)}(t, r) + V_{ee}^{(1)}(t, r) \cos \theta + V_{ee}^{(2)}(t, r) \cos^2 \theta.$$ (12)

The evolution of the radial component of the $n$-th Kohn–Sham orbital $\Psi_{nl}(r, t)$ is performed by the split-operator method [11] with a combination of the matrix diagonalization, the Crank–Nicolson approximation for exponential operators, and the Numerov approximation for the second-order derivative with respect to the radial coordinate.
3. Results of calculations

To obtain the high low-frequency response of atomic system, we assume that the electric field of the laser pulse represents a superposition of the strong field on the main angular frequency $\omega_1$ and weak frequency-detuned second-harmonic field with angular frequency $\omega_2 = 2\omega_1 + \Delta \omega$ [3, 5, 6]:

$$E(t) = -\frac{\partial A}{\partial t},$$
$$A(t) = -\sin^2\left(\frac{\pi t}{\tau_p}\right)\left[\frac{E_1}{\omega_1}\sin(\omega_1 t) + \frac{E_2}{\omega_2}\sin(\omega_2 t)\right], \quad 0 < t < \tau_p. \quad (13)$$

The detuning frequency is set as $\Delta f = \Delta \omega/(2\pi)$, where $\Delta \omega = -0.2\omega_1$. The main angular frequency $\omega_1$ corresponds to the wavelength of 800 nm; intensities of frequency components are $I_1 = 8 \times 10^{14}$ W/cm$^2$ and $I_2 = 3.2 \times 10^{13}$ W/cm$^2$, respectively. The pulse duration is $\tau_p = 60$ fs.

Figure 1 shows a comparison of the squared modulus $a_\omega = \int_0^\infty a(t)e^{i\omega t}dt$ of Fourier spectrum of the dipole acceleration calculated for the two-color field (13) for the argon and hydrogen atoms. In order to calculate the dipole acceleration for Ar we use the developed program for the solution of Kohn–Sham equations, while in the case of H we use the same program but for the only one electron bounded by Coulomb potential.

As can be seen in figure 1, the spectra for H and Ar contain large peaks at the laser field frequencies, $\omega_1/(2\pi)$ and $\omega_2/(2\pi)$, a broadband high-frequency part and a low-frequency part with the maximum near the detuning frequency $|\Delta f|$ in accordance with the paper [6]. Unlike hydrogen, in argon the low-frequency maximum has greater amplitude and a narrower spectrum.

We have calculated the time derivative of the low-frequency current density $\bar{j}$ using a spectral filter near the detuning frequency:

$$\frac{\partial \bar{j}}{\partial t} = -Na_\omega \frac{1}{2\pi} \int_{-\infty}^{\infty} a_\omega g(\omega)e^{-i\omega t}d\omega, \quad (14)$$

where $g(\omega)$ is a mask-function equal to unity for $|\omega| < 2\Delta \omega$ and smoothly vanishing outside this interval. The corresponding waveforms for Ar and H are shown in figure 2(a). As is seen, the...
maximum amplitude of the low-frequency current for H and Ar is approximately the same, but in the case of H, the generation of the low-frequency current lasts shorter time.

To explain this discrepancy, one should take into account that the generation of a current of low frequency occurs in the interval of increasing of free-electrons concentration [6]. As can be seen in figure 2(b), which shows the concentration of free electrons $n_{\text{free}}(t)$, the concentration increase interval for the H atom is on the leading edge of the laser pulse and is significantly shorter than in the case of the Ar atom. The H atom that has an ionization potential of $I_p = 0.5$ a.u., becomes fully ionized when intensity reaches $2 \times 10^{14} \text{ W/cm}^2$. As a result, the depletion of neutral atoms and the generation of low-frequency current occurs over a short time interval of $\approx 10$ fs at the leading edge of the pulse. When a laser pulse interacts with Ar, ionization lasts longer time ($\approx 20$ fs) due to the much weaker effect of atomic levels depletion. At the leading edge of a laser pulse, an increase in the concentration of free electrons in Ar is smaller due to the greater ionization potential ($I_p = 0.57$ a.u.) than in H, as well as due to the effect of screening the external field of the laser pulse under the influence of the dynamic polarization of the outer shell of the atom [18]. However, the Ar atom has doubly degenerate active orbital $3p_0$ with
respect to spin. Therefore, at high field intensities (above \(7 \times 10^{14} \text{ W/cm}^2\)), double ionization of the atom occurs, which significantly increases the duration of ionization and generation of a low-frequency current.

Returning to figure 1 it is important to note, that the dipole acceleration spectrum for H and Ar also contains a high-frequency part lying in the vacuum ultraviolet range, which is known as high-order harmonic generation (HHG) and is associated with rescattering of the photoelectrons on the parent ion [1, 9, 10]. The shape of the spectrum depends strongly on the type of atom. The HHG spectrum for the case of Ar contains a Cooper minimum located approximately at 51 eV, which is in good agreement with experiments [19, 20]. The HHG spectrum for the H atom has a smoother plateau, which ends at a much lower frequency than for Ar. In addition, the spectral intensity at the plateau cutoff for the H atom is substantially lower than that for Ar. This fact is also explained in the framework of analysis of the dynamics of the atomic levels population. In the case of the H atom, most electrons are ionized at the leading edge of a laser pulse, and there is no generation of harmonics at the center of the laser pulse. In the case of the Ar atom, on the contrary, external orbitals remain almost non-ionized at the maximum of the laser-pulse envelope and therefore make a large contribution to the HHG.

4. Conclusions
A computational program has been developed for the numerical simulation of the interaction of many-electron atoms with an intense field of the ultrashort laser pulse. Based on the developed program, we have calculated the electron current density and the concentration of free electrons generated during the ionization of the argon and hydrogen atoms by an intense two-laser laser pulse consisting of a field at the main frequency and frequency-detuned second harmonic. It is shown that the increase of free-electron concentration in Ar lasts a significantly longer time than in H due to the double degeneracy of the most active orbital and due to the effect of the outer shell polarization. At high laser-pulse peak intensities, this leads to a higher spectral intensity of both the low-frequency and high-frequency electron currents in Ar compared with H.

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