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Attosecond pulse formation via switching of resonant interaction by tunnel ionization

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We derive an analytical solution uncovering the origin of few-cycle attosecond pulse formation from vacuum-ultraviolet (VUV) radiation in an atomic gas simultaneously irradiated by a moderately strong infrared (IR) laser field, which does not perturb atoms in the ground state, but induces rapid quasistatic ionization from the excited states [*Opt. Lett.* **36**, 2296 (2011)]. The derived solution shows that the pulses are produced due to periodic switching of the resonant interaction between the incident VUV radiation and the atoms: turning it off near the crests of the IR-field strength and switching it back on near the IR field zero-crossings. We extend the method originally proposed in [*Opt. Lett.* **36**, 2296 (2011)] to non-hydrogenlike media and show that the pulses can be produced from resonant VUV radiation in a variety of atomic gases. The pulses are nearly bandwidth-limited without external adjustment of phases of the generated sidebands. Proximity of the carrier frequency of the produced pulses to intra-atomic resonances may allow their utilization for nondestructive steering of ultrafast dynamics of the bound electrons. The experimental possibilities for attosecond pulse formation from 58.4 nm VUV radiation in helium and from 73.6 nm VUV radiation in neon dressed by the 3.9 μm laser field, as well as from 122 nm VUV radiation in atomic hydrogen dressed by CO_2-laser field are discussed.

The first decade of the millennium was marked by the birth of attosecond science - the branch of physics devoted to direct investigation and control of the motion of charged particles in atoms, molecules, clusters, and solids, unfolding on the attosecond time scale [1–6].

In recent years, there has been a growing interest in investigation of the ultrafast dynamics of atoms, simultaneously irradiated by an intense low-frequency (LF) laser field, far detuned from all the atomic resonances, and a high-frequency (HF) radiation, quasi-resonant to a transition from the ground to an excited bound or autoionizing atomic state [6–15]. Utilization of quasi-resonant radiation substantially enriches the toolset of attosecond science, allowing for switching the pathways of multiphoton excitation and/or ionization of atoms by the LF field, and under certain conditions leading to quantum interference of these pathways [7–15]. Up to now, however, the experimental studies are mainly limited to investigation of helium atoms, possessing the highest ionization potential among the neutral atoms and the highest-frequency transitions between the bound states. This choice is primarily motivated by the aspiration to perform an intra-atomic excitation using high-order harmonics of laser radiation and attosecond pulses with sufficiently high carrier frequency generated in noble gases [1–3].

Recently, a method to produce extremely short pulses with relatively low carrier frequency in the vicinity of the resonances of hydrogen-like atoms was proposed [16–18] and discussed [19–22]. The pulses are produced due to *the resonant* interaction of an incident HF radiation with hydrogen-like atoms, dressed by a moderately strong LF laser field. While the resonant HF radiation excites atoms from the energy level n=1 to the energy level n=2 (where n is the principal quantum number), the LF field produces the *sub-LF-field-cycle* perturbation of the excited energy level. The perturbation constitutes (i) *time-dependent* splitting and shift of the excited energy levels via the Stark effect as well as (ii) *time-dependent* broadening of these levels via tunnel ionization from the correspond-

ing excited states. The *sub-LF-field-cycle* variation of the instantaneous values of splitting and/or broadening of the excited energy levels, determined by the instantaneous strength of the LF field [14], leads to a multifrequency atomic response to the quasi-monochromatic incident HF radiation. As shown in references [16-21], under the appropriate conditions this corresponds to formation of extremely short few-femto- and attosecond pulses with carrier frequency inherently determined by the frequency of the resonant atomic transition. Since most of the spectral components of the produced pulses lay below the threshold of ionization and dissociation of various neutral atoms and molecules, such a source of the resonant attosecond pulses constitutes a unique tool for the nondestructive (nonionizing) study and control of ultrafast dynamic of the bound atomic and molecular electrons. This essentially distinguishes the proposed technique from the commonly used approach to attosecond pulse formation via the high-order harmonic generation (HHG) of laser radiation in gases [1-3]. In the latter case, the pulses consist of high-order harmonics, corresponding to the plateau and/or cutoff regions of the generated spectrum, and possessing frequencies far above the ionization threshold of the generating atoms. Consequently, interaction of such pulses with atoms/ molecules, which ionization/dissociation potential is comparable to that of the generating medium, results primarily in photoionization or photodissociation, respectively. Another difference of the proposed technique for attosecond pulse formation from HHG in gases is the absence of attochirp [3]: the pulse formation does not imply adjustment of phases of the generated sidebands, since all of them are produced in-phase.

In the present paper, we extend the method of extremely short pulse formation from resonant radiation to various non-hydrogenlike atoms and show the possibility to produce attosecond pulses in helium and neon. We derive an analytical solution uncovering the origin of attosecond pulse formation from the resonant HF radiation in the case of a relatively strong LF field [17, 18, 20, 21]. In this case the probability of atomic ionization *from the excited state*, corresponding to the upper energy level of the resonant HF atomic transition, approaches unity over each half-cycle of the LF field. For hydrogen-like atoms this corresponds to amplitude of the LF field strength, E_C, exceeding $0.15\times Z^3/n^3 E_A$, where Z is the nucleus charge and $E_A = m_e^2 e^5/\hbar^4$ is the atomic unit of electric field (e and m_e are the charge and the mass of electron, respectively, $\hbar$ is the Planck's constant). Ionization from the ground atomic state remains unimportant as far as $E_C < 0.05\times Z^3 E_A$. The derived solution shows that the attosecond pulses are produced due to confinement of the resonant atomic response to the incident quasi-monochromatic HF radiation within the extremely short time-intervals in the vicinity of zero-crossings of the LF field. For the rest of the time the *resonant* interaction between the HF radiation and the atoms is effectively switched off (the resonance disappears) because of rapid ionization from the excited atomic state, produced by the LF field. Both analytically and numerically, we show that in this regime, the effect of extremely short pulse formation from the resonant radiation is insensitive to the particular dependences of (i) Stark shift / splitting of the excited atomic energy level as well as (ii) rate of quasistatic ionization from the corresponding excited state(s) on the LF field strength. Thus, the discussed mechanism can be used for attosecond pulse formation in a variety of atomic gases, possessing sufficiently high ionization potential. Extension of the proposed method to non-hydrogenlike media is important for an experimental realization, since both atomic hydrogen and hydrogen-like ions are unstable. Use of various media for attosecond pulse generation also substantially increases the practical value of the proposed technique, enabling formation of pulses at different carrier frequencies, suitable for ultrafast excitation of different atomic, molecular and solid-state systems.

The paper is organized as follows: after the introduction, we describe the model and derive the analytical solution for a resonant HF radiation, propagating through an atomic gas dressed by an intense LF field. We compare the analytical results to the results of numerical solutions, obtained within the more general models [17, 23] and discuss the possibilities to produce attosecond pulses from 58.4 nm vacuum-ultraviolet (VUV) radiation in helium dressed by 3.9 μm laser field [24], as well as from 73.6 nm VUV radiation in neon dressed by 3.9 μm laser field and 1-fs pulses from 122 nm VUV radiation in atomic hydrogen dressed by a CO_2-laser field [25].

Let us consider interaction of a HF radiation with an atomic gas. The incident HF radiation is monochromatic and propagates along the z-axis. At the entrance to the medium, z=0, it has the form

$$\vec{E}_{inc}(t) = \frac{1}{2}\vec{x}_0 E_0 \exp\{-i\omega t\} + \text{c.c.}, \qquad (1)$$

where E_0 is its amplitude, ω is the angular frequency, and c.c. stands for complex conjugation. The HF radiation (1) is chosen to be near-resonant to a transition $|1\rangle \leftrightarrow |2\rangle$ from the ground atomic state to one of the lowest excited states, $\omega \approx \omega_{21}^0$ (where ω_{21}^0 is the unperturbed frequency of the resonant transition).

The gas is simultaneously irradiated by a moderately strong linearly polarized LF laser field

$$\vec{E}_{LF}(\tau) = \frac{1}{2}\vec{x}_0 E_C \exp\left\{-i\left[\Omega(t - z/c) + \varphi_0\right]\right\} + \text{c.c.} \qquad (2)$$

Here E_C is the amplitude of the LF field, Ω is its angular frequency, φ_0 is its initial phase, and c is the speed of light in vacuum. The term low-frequency (LF) means that the frequency of the field is much smaller than the frequencies of all the transitions from both the ground and the resonantly excited by the HF radiation atomic states. Due to far-detuning from the relevant atomic resonances, the LF field (2) does not suffer from the atomic dispersion at the considered propagation distances and traverses the medium without substantial modification along the direction of propagation of the HF radiation. Since both HF and LF fields are polarized in the same direction, $\vec{E}_{inc} \,\|\, \vec{E}_{LF} \,\|\, \vec{x}_0$, their polarizations are not changed during propagation through an isotropic gas, so that the vector notations will be omitted, for now on.

The amplitude of the LF field is chosen to be under the threshold of atomic multiphoton excitation and/or ionization from the ground state $|1\rangle$, however, the LF field is strong enough to almost completely ionize atoms from the excited state $|2\rangle$ over *every* half-cycle, producing extreme broadening of the $|1\rangle \leftrightarrow |2\rangle$ transition line in the vicinity of its crests, Fig. 1. The Keldysh parameter of the LF field for the excited state $|2\rangle$ is much smaller than unity, $\gamma_{Keldysh} << 1$, so that the excited-state-ionization takes place via tunneling mechanism. If the state $|2\rangle$ corresponds to an eigenstate in spherical coordinates, then according to [26], the ionization rate from it is given by

$$w_{ion}^{(2)}(\tau) = \frac{m_e e^4}{\hbar^3} C_{\kappa l}^2 \kappa^2 (2l+1) \frac{4^{n^*-|m|}(l+|m|)!}{|m|!(l-|m|)!} |F_{LF}(\tau)|^{|m|+1-2n^*} \exp\left\{-\frac{2}{3|F_{LF}(\tau)|}\right\}, \qquad (3)$$

where $\tau \equiv t - z/c$ is the local time (in the reference frame, comoving with the LF wavefront), $\kappa = \sqrt{I_P^{(2)}/I_H}$, $I_P^{(2)}$ is the ionization potential from the excited atomic state $|2\rangle$, $I_H = \frac{m_e e^4}{2\hbar^2}$ is the ionization potential from the ground state of atomic hydrogen, m is the projection of the angular momentum l on the LF electric field, $n^* = Z/\kappa$ is the effective principal quantum number, Z is the atomic core charge, $C_{\kappa l}$ is the dimensionless asymptotic coefficient of the atom wave function, and $F_{LF}(\tau) \equiv \frac{E_{LF}(\tau)}{\kappa^3 E_A}$ is the reduced dimensionless LF field. The tunnel ionization rate from an eigenstate in parabolic coordinates differs from (3) only by the pre-exponential factor [27, 17, 18].

The LF field also produces instantaneous Stark shift of the energy level, corresponding to the excited atomic state $|2\rangle$, which energy E_2 takes the form

$$\mathrm{E}_2 = \mathrm{E}_2^{(0)} + \Delta\mathrm{E}_2(\tau), \qquad (4)$$

where $\mathrm{E}_2^{(0)}$ is the unperturbed energy of the excited state $|2\rangle$, and $\Delta\mathrm{E}_2$ is the instantaneous shift of the energy level, induced by the LF field. In the lowest order of perturbation theory one has $\Delta\mathrm{E}_2 \propto F_{LF}(\tau)$ for hydrogen-like atoms and $\Delta\mathrm{E}_2 \propto F_{LF}^2(\tau)$ for non-hydrogen-like atoms.

Propagation of the HF radiation through the medium is described by the wave equation

$$\frac{\partial^2 E_{HF}}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E_{HF}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P}{\partial t^2}, \tag{5}$$

where E_{HF} is the HF radiation strength and P is the *resonant* HF polarization of the gas. At the considered propagation distances, the nonresonant interaction of the HF radiation with atoms is unimportant and can be disregarded. Since the characteristic scales of spatial evolution of the HF radiation in any gas are much larger than the HF radiation wavelength, the substitution $t \to \tau \equiv t - z/c$ allows reducing wave equation (5) to

$$\frac{\partial E_{HF}}{\partial z} = -\frac{2\pi}{c}\frac{\partial P}{\partial \tau}, \tag{6}$$

Equation (6) implies the slowly-evolving-wave approximation [28, 29], $\left|\frac{\partial E_{HF}}{\partial z}\right| << \frac{1}{c}\left|\frac{\partial E_{HF}}{\partial \tau}\right|$. Within the additional approximation of slowly-varying-envelope, $E_{HF}(z,\tau) = \frac{1}{2}\tilde{E}_{HF}(z,\tau)\exp\{-i\,\omega\tau\} + \text{c.c.}$, $P(z,\tau) = \frac{1}{2}\tilde{P}(z,\tau)\exp\{-i\,\omega\tau\} + \text{c.c.}$, $\left|\partial\tilde{E}_{HF}/\partial\tau\right| << \omega$, $\left|\partial\tilde{P}/\partial\tau\right| << \omega$, $\left|\partial\tilde{E}_{HF}/\partial z\right| << \omega/c$, $\left|\partial\tilde{P}/\partial z\right| << \omega/c$, equation (6) has the solution

$$\tilde{E}_{HF}(z,\tau) = \tilde{E}_{HF}(0,\tau) + \tilde{E}_{Scatt}(z,\tau), \quad \tilde{E}_{Scatt}(z,\tau) \equiv i\frac{2\pi\omega}{c}\int_0^z \tilde{P}(z',\tau)dz', \tag{7}$$

where $\tilde{E}_{Scatt}(z,\tau)$ is the slowly-varying envelope of HF radiation, resonantly scattered by the atoms, which is entirely determined by the slowly-varying envelope of the resonant polarization $\tilde{P}$.

The envelope of polarization $\tilde{P}$, in turn, is proportional to the envelope a_{21} of the atomic coherence $\rho_{21} = a_{21}\exp\{-i\,\omega\tau\}$ at the resonant transition $|1\rangle \leftrightarrow |2\rangle$:

$$\tilde{P}(z,\tau) = 2Nd_{12}a_{21}(z,\tau), \tag{8}$$

where N is the atomic density, and d_{12} is the dipole moment of the resonant transition. The value a_{21} satisfies an equation

$$\frac{da_{21}}{d\tau} + \left(i\left(\omega_{21}(\tau) - \omega\right) + \gamma_{21}(\tau)\right)a_{21} = \frac{i}{2\hbar}n_{12}d_{21}\tilde{E}_{HF}, \tag{9}$$

where $\omega_{21}(\tau)$ is the instantaneous frequency of the transition $|1\rangle \leftrightarrow |2\rangle$, $\gamma_{21}(\tau)$ is its instantaneous decoherence rate, $\tilde{E}_{HF}$ is the envelope of the HF radiation, n_{12} is population difference between the states $|1\rangle$ and $|2\rangle$, $n_{12} \equiv \rho_{11} - \rho_{22}$ (in the following we consider a relatively weak HF field, which does not change populations of the atomic states considerably during the interaction time, $n_{12} \cong 1$), and $d_{21} = d_{12}^*$.

In general, $\omega_{21}(\tau) = \omega_{21}^{(0)} + \Delta\mathrm{E}_2(\tau)/\hbar$ and $\gamma_{21}(\tau) = \gamma_{21}^{(0)} + w_{ion}^{(2)}(\tau)/2$, where $\omega_{21}^{(0)}$ and $\gamma_{21}^{(0)}$ are the unperturbed transition frequency and decoherence rate, correspondingly. However, $\Delta\mathrm{E}_2 \propto F_{LF}$ or F_{LF}^2, while $w_{ion}^{(2)} \propto \exp\left\{-\frac{2}{3|F_{LF}|}\right\}$. Hence, in the case of a strong LF field, $F_{LF}^{\max} \succ 0.1$, considered in this paper, the peak value of ionization broadening of the resonant transition line considerably exceeds the depth of Stark sweeping of the resonant transition frequency,

$\max\{w_{ion}^{(2)}\} >> \max\{\Delta E_2/\hbar\}$. Therefore, in order to obtain an analytical solution one may neglect variation of frequency of the resonant transition as compared to variation of its linewidth, assuming $\omega_{21}(\tau) = \omega_{21}^{(0)}$. The opposite case of a relatively weak LF field, where $\max\{w_{ion}^{(2)}\} << \max\{\Delta E_2/\hbar\}$, has been considered (for the hydrogen-like atoms) in our recent paper [22].

Taking into account the strongly nonlinear dependence of the tunnel ionization rate (3) on the LF field strength (2), we adapt a stepwise approximation for the local-time-dependence of decoherence rate of the resonant transition:

$$\gamma_{21}(\tau) = \begin{cases} \overline{\gamma}_{\min}, & 0 \le \tau < \Delta t_{\text{zero}}, \\ \overline{\gamma}_{\max}, & \Delta t_{\text{zero}} \le \tau < \pi/\Omega, \end{cases} \quad \gamma_{21}(\tau + \pi/\Omega) = \gamma_{21}(\tau). \tag{10}$$

The decoherence rate $\gamma_{21}(\tau)$ takes it minimum value $\overline{\gamma}_{\min}$ near zero-crossing of the LF field at $\tau = \Delta t_{\text{zero}}/2$ (corresponding to $\varphi_0 = [\pi - \Omega \Delta t_{\text{zero}}]/2$ in (2)), and maximum value $\overline{\gamma}_{\max}$ at the rest of time, Fig. 1. Since the excited-state ionization rate (3) is a periodic function of time, which period equals the half-cycle of the LF field, the decoherence rate (10) possesses the same periodicity.

According to the results of our numerical calculations [17] extremely short pulses can be produced from the quasi-monochromatic HF radiation in an infinitely thin medium, while an increase of the medium thickness leads only to an increase of the peak amplitude of the pulses (due to increased intensity of the generated sidebands) at the cost of slight increase of the pulse duration (because of phase mismatching of the sidebands). Therefore, similarly to [22], in order to derive an analytical solution we assume that the length of the medium L is small enough, such that the incident HF spectral component dominates over the generated sidebands. In this case, the resonant atomic response can be calculated accounting only for the scattering of the incident HF radiation, and neglecting rescattering of the generated sidebands. This corresponds to assumption $E_{HF}(z,\tau) \approx E_{inc}(\tau)$ in right hand of (9). Substitution of (1) into (9) and use of the approximations (10) and $\omega_{21}(\tau) = \omega_{21}^{(0)}$ lead to the steady-state solution

$$a_{21}(\tau) = \begin{cases} a_{21}^{(1)}(\tau), & 0 \le \tau < \Delta t_{\text{zero}}, \\ a_{21}^{(2)}(\tau), & \Delta t_{\text{zero}} \le \tau < \pi/\Omega, \end{cases} \tag{11}$$

where $a_{21}^{(1)}(0) = a_{21}^{(2)}(\pi/\Omega)$ and $a_{21}^{(2)}(\Delta t_{\text{zero}}) = a_{21}^{(1)}(\Delta t_{\text{zero}})$. The dependences $a_{21}^{(1)}(\tau)$ and $a_{21}^{(2)}(\tau)$ are given by

$$\begin{cases} a_{21}^{(1)}(\tau) = C_1 \exp\{(i\Delta\omega - \overline{\gamma}_{\min})\tau\} + D_1, \\ a_{21}^{(2)}(\tau) = C_2 \exp\{(i\Delta\omega - \overline{\gamma}_{\max})(\tau - \Delta t_{\text{zero}})\} + D_2, \end{cases} \tag{12}$$

where $\Delta\omega \equiv \omega - \omega_{21}^{(0)}$ and

$$\begin{cases} D_1 = i \dfrac{n_{12} d_{21} E_0}{2\hbar(\overline{\gamma}_{\min} - i\Delta\omega)}, \\ D_2 = i \dfrac{n_{12} d_{21} E_0}{2\hbar(\overline{\gamma}_{\max} - i\Delta\omega)}, \end{cases} \tag{13}$$

$$\begin{cases} C_1 = (D_2 - D_1) \dfrac{\exp\{(i\Delta\omega - \overline{\gamma}_{\max})(\pi/\Omega - \Delta t_{\text{zero}})\} - 1}{\exp\{(i\Delta\omega - \overline{\gamma}_{\max})\pi/\Omega\} \exp\{(\overline{\gamma}_{\max} - \overline{\gamma}_{\min})\Delta t_{\text{zero}}\} - 1}, \\ C_2 = (D_1 - D_2) \dfrac{\exp\{(i\Delta\omega - \overline{\gamma}_{\min})\Delta t_{\text{zero}}\} - 1}{\exp\{(i\Delta\omega - \overline{\gamma}_{\max})\pi/\Omega\} \exp\{(\overline{\gamma}_{\max} - \overline{\gamma}_{\min})\Delta t_{\text{zero}}\} - 1}. \end{cases} \tag{14}$$

According to (7) and (8), the envelope of the resonantly scattered HF radiation has the form

$$\tilde{E}_{Scatt}(\tau)=\begin{cases}\tilde{E}^{(1)}_{Scatt}(\tau), & 0\le\tau<\Delta t_{\text{zero}},\\ \tilde{E}^{(2)}_{Scatt}(\tau), & \Delta t_{\text{zero}}\le\tau<\pi/\Omega,\end{cases}\quad \tilde{E}_{Scatt}(\tau+\pi/\Omega)=\tilde{E}_{Scatt}(\tau), \tag{15}$$

where $\tilde{E}^{(1)}_{Scatt}(\tau)=i\mathrm{A}\times a^{(1)}_{21}(\tau)$ and $\tilde{E}^{(2)}_{Scatt}(\tau)=i\mathrm{A}\times a^{(2)}_{21}(\tau)$; $\mathrm{A}\equiv 4\pi N d_{12}\dfrac{\omega}{c}L$.

Equations (15), (11)-(14) acquire especially clear form in the case of almost complete ionization from the excited state $|2\rangle$ in the vicinity of crests of the LF field, $\exp\{-\overline{\gamma}_{\max}(\pi/\Omega-\Delta t_{\text{zero}})\}<<1$, and relatively small instantaneous ionization rate near the LF field zero-crossings, $\exp\{-\overline{\gamma}_{\min}\Delta t_{\text{zero}}\}\approx 1-\overline{\gamma}_{\min}\Delta t_{\text{zero}}$, which is realized in a sufficiently strong LF field. Assuming that detuning of the incident HF radiation from the resonance is substantively smaller than the doubled LF field frequency, $\exp\{i\Delta\omega\tau\}\approx 1+i\Delta\omega\tau$, one obtains

$$\begin{cases}\tilde{E}^{(1)}_{Scatt}(\tau)=-\chi E_0\tau, & 0\le\tau<\Delta t_{\text{zero}},\\ \tilde{E}^{(2)}_{Scatt}(\tau)=-\chi E_0\Delta t_{\text{zero}}\exp\{-\overline{\gamma}_{\max}(\tau-\Delta t_{\text{zero}})\}, & \Delta t_{\text{zero}}\le\tau<\pi/\Omega.\end{cases} \tag{16}$$

where $\chi\equiv\dfrac{2\pi N n_{12}|d_{21}|^2\omega}{\hbar c}L$. As follows from (16), the scattered HF radiation has a form of a pulse train with sub-LF-field-cycle duration $\tau_{pulse}=\dfrac{\sqrt{2}-1}{\sqrt{2}}\Delta t_{\text{zero}}+\dfrac{\ln 2}{2\overline{\gamma}_{\max}}\approx 0.3\Delta t_{\text{zero}}+\dfrac{0.35}{\overline{\gamma}_{\max}}$ (the duration of pulses is defined as the full width at half-maximum of the scattered intensity $I_{Scatt}\propto\tilde{E}^2_{Scatt}(\tau)$).

The Fourier transform of the scattered radiation (15) has the form

$$\tilde{E}_{Scatt}(\tau)=i\mathrm{A}\cdot\sum_{n=-\infty}^{\infty}\alpha_{2n}\exp\{-i2n\Omega\tau\}. \tag{17}$$

In the particular case (16) the coefficients α_{2n} are determined by the following expressions:

$$\alpha_{2n}=i\frac{\chi E_0}{\mathrm{A}}\frac{\Omega}{\pi}\left[\frac{\exp\{i2n\Omega\Delta t_{\text{zero}}\}-1}{4n^2\Omega^2}+\frac{\overline{\gamma}_{\max}\Delta t_{\text{zero}}\exp\{i2n\Omega\Delta t_{\text{zero}}\}}{2n\Omega(2n\Omega+i\overline{\gamma}_{\max})}\right],\quad n\neq 0. \tag{18}$$

The direct component of the slowly-varying amplitude of the scattered field $\tilde{E}_{Scatt}(\tau)$ has the form

$$\alpha_0=i\frac{\chi E_0}{\mathrm{A}}\frac{\Omega}{\pi}\left[\frac{\Delta t^2_{zero}}{2}+\frac{\Delta t_{\text{zero}}}{\overline{\gamma}_{\max}}\right]. \tag{19}$$

In general case (15) the coefficients α_{2n} can be expressed as

$$\begin{aligned}\alpha_{2n}=\frac{\Omega}{\pi}\Bigg[&C_1\frac{\exp\{[i(2n\Omega+\Delta\omega)-\overline{\gamma}_{\min}]\Delta t_{\text{zero}}\}-1}{i(2n\Omega+\Delta\omega)-\overline{\gamma}_{\min}}+\\ &C_2\frac{\exp\{[i\Delta\omega-\overline{\gamma}_{\max}][\pi/\Omega-\Delta t_{\text{zero}}]\}-\exp\{i2n\Omega\Delta t_{\text{zero}}\}}{i(2n\Omega+\Delta\omega)-\overline{\gamma}_{\max}}+\\ &+(D_1-D_2)\frac{\exp\{i2n\Omega\Delta t_{\text{zero}}\}-1}{i2n\Omega}\Bigg],\end{aligned} \tag{20}$$

$$\alpha_0 = \frac{\Omega}{\pi}\left[C_1 \frac{\exp\left\{\left(i\Delta\omega - \overline{\gamma}_{\min}\right)\Delta t_{\text{zero}}\right\} - 1}{i\Delta\omega - \overline{\gamma}_{\min}} + D_1 \Delta t_{\text{zero}} + \right.$$
$$\left. C_2 \frac{\exp\left\{\left(i\Delta\omega - \overline{\gamma}_{\max}\right)\left(\pi/\Omega - \Delta t_{\text{zero}}\right)\right\} - 1}{i\Delta\omega - \overline{\gamma}_{\max}} + D_2\left(\pi/\Omega - \Delta t_{\text{zero}}\right) \right]. \quad (21)$$

where C_i, D_j are defined by equations (13), (14). The derived solution (7), (15)-(21) is valid for arbitrary atomic system. The quantitative features of field-atom interaction specific for a given medium and LF field are taken into account via parameters $\overline{\gamma}_{\min}$, $\overline{\gamma}_{\max}$, and Δt_{zero}. In the following, we compare the derived analytical solution to the results of numerical calculations within the more general models [17, 23], taking into account the ground and two excited energy levels, which instantaneous positions and widths are modulated by the LF field through the nonlinear (taking into account the linear, quadratic and cubic constituents) Stark effect and the tunnel ionization, respectively. The numerical calculations are performed without the slowly-varying amplitude and rotating-wave approximations. The model [23] is designed specifically for non-hydrogen-like medium. Contrary to the models, used in papers [17, 18, 20, 21], it takes into account not only the *adiabatic* influence of a LF field on the atomic system through the sub-LF-field-cycle shift and/or broadening of the energy levels, but also *non-adiabatic* transitions between the nondegenerate excited energy levels, induced by the LF field.

Let us apply the above model to study the possibility to produce few-cycle attosecond pulses from 58.4 nm VUV radiation due to the resonant interaction with 1s↔2p transition of helium dressed by 3.9 μm infra-red (IR) laser field from OPCPA source [24]. The incident VUV radiation can be generated in different ways. Firstly, it can be produced via frequency tripling of 175 nm radiation in a gas. The 175 nm radiation, in turn, can be generated via frequency doubling of 350 nm radiation in a nonlinear crystal with sufficiently wide transparency range, such as lithium triborate (LBO). The latter radiation can be produced by OPA/OPO or via frequency tripling of either Nd: YLF or Nd: YAG laser field. Secondly, the incident VUV radiation can be obtained via the resonantly enhanced HHG of Ti:Sa laser field in InP plasma plume [30, 31]. In both cases the efficiency of conversion of the radiation energy from visible/near-infrared to VUV spectral range normally exceeds the achieved conversion efficiency of attosecond pulse formation via HHG., This is because of (i)the absence of a tiny factor, determined by the probability of recombination of the detached electron [32], inherent to HHG process, and (ii) the possibility to resonantly enhance the atomic susceptibility. Under the action of the laser field with an intensity $I_{\text{IR}}=1.5\times10^{13}$ W/cm^2 the resonant response of helium atoms to the incident VUV radiation and, correspondingly, the resonantly scattered radiation, are confined to the sub-IR-field-cycle time intervals near zero-crossings of the IR field, Fig. 2. This can be understood referring to the dependence of the optical depth of a medium, G, on the linewidth of the resonant transition, γ: $G\sim1/\gamma$ [33]. Since in the vicinity of the IR field crests excited-state-ionization greatly (by orders of magnitude) broadens the resonant transition line, it leads to the proportional decrease of the optical depth, making the medium transparent for the resonant VUV radiation. On the contrary, in the vicinity of zero-crossings of the IR field (during the time intervals Δt_{zero}) the ionization broadening is small, providing conditions for the resonant absorption and scattering of the incident VUV radiation. The Fourier transform of the atomic response is plotted in Fig. 3. It corresponds to a broad comb of the equidistant spectral components, separated by the doubled frequency of the IR field. The generated sidebands are almost in-phase to each other and in-anti-phase to the incident VUV radiation, corresponding to burning of the extremely short dips in the time-dependence of the output VUV intensity because of the resonant absorption of the VUV radiation in the vicinity of zero-crossings of the IR field, Fig. 4. In an optically thin medium the output intensity satisfies the relation

$$I_{HF}(z,\tau)=\frac{c}{8\pi}\left|\tilde{E}_{HF}\right|^2\approx\frac{c}{8\pi}E_0^2\left(1+2\tilde{E}_{Scatt}(z,\tau)/E_0\right). \quad (22)$$

Naturally, the shape of the absorption dips in Fig. 4 almost coincides with the inversed shape of extremely short bursts of resonantly scattered radiation, plotted in Fig. 2. As is well known, the resonant absorption itself occurs due to the destructive interference of the incident and resonantly scattered radiation (which in turn is due to $\pi/2$ phase shift of the resonant component of polarization). Thus, elimination of the resonant component in the spectrum of the output VUV radiation (frequency of the resonant component coincides with the frequency of the incident VUV radiation) and slight attenuation of the nearest sidebands by a narrowband spectral filter results in the production of the attosecond pulses shown in Fig. 5. The pulses arise at the positions of the intensity dips, i.e. where the resonant absorption and scattering take place. As seen from Figs. 2 and 5, the produced pulses are shorter than the bursts of the resonantly scattered radiation. This is due to the facts, that (i) while Fig. 2 shows the radiation strength, Fig. 5 illustrates the time-dependence of the radiation intensity (squaring of the pulse envelope reduces the pulse duration), and (ii) suppression of the resonant component of the output VUV spectrum and attenuation of the most intense sidebands lead to effective broadening of the output spectrum and, hence, shortening of the produced pulses. The duration of pulses shown in Fig. 5 equals 420 as, corresponding to 2.15 cycles of the VUV carrier, the pulse repetition period is determined by the half-cycle of the IR field and equals 6.5 fs. The produced attosecond pulses are nearly bandwidth-limited without external adjustment of phases of the generated sidebands. The values of the parameters $\Delta t_{zero}=0.23\pi/\Omega$, $\overline{\gamma}_{\min}=0.1\times\Omega$, $\overline{\gamma}_{\max}=15\times\Omega$, used for the analytical calculations in Figs. 2–5, were chosen in order to support the best agreement between the analytical and numerical solutions. Although in this paper we consider analytically the limit of small optical depth of the medium, the numerical analysis [21] for the larger values of the optical depth shows that efficiency of transformation of the incident VUV radiation into the attosecond pulses can reach ~ 50%, while efficiency of transformation of the total (IR + VUV) radiation can reach ~ 5%.

The pulses of comparable duration, 460 as, shown in Fig. 6, can be produced from 73.6 nm VUV radiation via the resonant interaction with atoms of neon, dressed by 3.9 μm IR laser field. The incident VUV radiation is resonant to the transition from the ground atomic state, $2s^22p^6$, to one of the lowest excited states, $2s^22p^5$ ($^2P^0_{1/2}$)3s. The numerical model, used for calculations, also takes into account the $2s^22p^5$ ($^2P^0_{3/2}$)3s state and all the $2s^22p^53p$ states of neon. The transitions from the ground state to the excited $2s^22p^53s$ states are induced by the VUV radiation, while the transitions between $2s^22p^53s$ and $2s^22p^53p$ states are driven by the IR field. Simultaneously, the IR field induces rapid ionization from all the excited atomic states, which is taken into account via the time-dependent ionization rates (3). The intensity of the IR field, required for ionization switching of the resonant interaction in neon and generation of attosecond pulses, shown in Fig. 6, equals $I_{IR}=5\times10^{13}$ W/cm^2. The higher IR field intensity compared to the case of helium is primarily caused by the larger value of ionization potential from the resonant excited state of neon (which equals 4.71 eV vs. 3.37 eV in helium). In order to achieve the same value of reduced dimensionless laser field, which determines the ionization rate (3), $F_{LF}\propto 1/\kappa^3$ ($\kappa\propto\sqrt{I_P^{(2)}}$ and $I_P^{(2)}$ is the ionization potential from the chosen excited atomic state), the laser intensity in neon should be increased approximately 2.7 times in comparison to that in helium. The pulses, shown in Fig. 6, comprise 1.85 cycles of the VUV carrier and are nearly bandwidth-limited. The values of the parameters Δt_{zero}, $\overline{\gamma}_{\min}$, $\overline{\gamma}_{\max}$, used for the analytical calculation in Fig. 6, are $\Delta t_{zero}=0.25\pi/\Omega$, $\overline{\gamma}_{\min}=0.1\times\Omega$, $\overline{\gamma}_{\max}=15\times\Omega$.

In order to show that the effect of extremely short pulse formation from an incident VUV radiation via ionization switching of its resonant interaction with an atomic gas is insensitive to the choice of a specific medium we consider the pulse formation in atomic hydrogen. In Fig. 7 we plot the time-dependence of the VUV intensity resulted from the resonant interaction of 122 nm VUV

radiation with 1s↔2p transition of atomic hydrogen dressed by 10.65 μm CO_2-laser field with intensity I_{IR}=2.2×10^{13} W/cm^2 [25] and subsequent suppression of the resonant component of the output VUV spectrum. In such a case, the output radiation represents a train of pulses with duration 1.1 fs and repetition period 17.8 fs. The analytical solution in Fig. 7 corresponds to the parameters values $\Delta t_{\text{zero}} = 0.21\pi/\Omega$, $\bar{\gamma}_{\min} = 0.1\times\Omega$, $\bar{\gamma}_{\max} = 10\times\Omega$. From Figs. 5 and 7 it follows that the ratio between duration of pulses, produced in atomic hydrogen and helium, 1.1 fs / 0.42 fs ≈ 2.6, almost coincides with the ratio between the wavelengths of respective IR fields, used for interruption of the resonant interaction, 10.65 μm / 3.9 μm ≈ 2.7. This can be understood within the applied approximation (10), taking into account that the dependencies of ionization rates from the 2p states of helium and atomic hydrogen on the IR field strength are quite close to each other (the corresponding ionization potentials are 3.37 eV for helium and 3.40 eV for atomic hydrogen). In the quasistatic approximation, the instantaneous excited-state-ionization rate (3) is determined solely by the IR field strength, while variation of frequency (wavelength) of the IR field results in stretching / shrinking of the time scale (see Fig. 1). Therefore, since the pulse duration is proportional to the length of time-interval of the resonant interaction in the vicinity of zero-crossings of the IR field, $\tau_{pulse} \propto \Delta t_{\text{zero}}$, it is proportional to the IR wavelength, $\Delta t_{\text{zero}} \propto \Omega^{-1} \propto \lambda_{IR}$.

As follows from Figs. 2–7, regardless of the different dependencies of the instantaneous Stark shifts and excited-state-ionization rates on the IR field strength for helium, neon, and atomic hydrogen, the derived analytical solution even in its simplest form (7), (15)-(19) is in excellent agreement with the results of numerical calculations performed within the more general models [17, 23] without the slowly-varying amplitude and the rotating-wave approximations, taking into account time-dependencies of the instantaneous Stark shifts and ionization rates from two excited energy levels [17], nonadiabatic coupling of them [23], as well as complicated energy level structure in the case of neon. This agreement originates from the fact that the main effect of the IR field in the considered regime, - the periodic switching off the resonant interaction between the VUV radiation and the atoms near crests of the IR field strength and resumption of the resonant interaction near the IR field zero-crossings, is quite correctly described within the two-level model and stepwise approximation (10) for the time-dependence of the excited-state tunnel ionization rate.

In conclusion, in this paper we extended the method of extremely short pulse formation from the resonant radiation, originally proposed in [17], to non-hydrogenlike media, particularly, to noble gases. We derived the analytical solution for the envelope of extremely short atto- and femtosecond pulses produced from the resonant VUV radiation in an atomic gas, dressed by an IR laser field [17, 18, 20, and 21]. The IR field is chosen to be strong enough to almost completely ionize atoms from the upper state of the resonant VUV transition during each IR half-cycle but does not essentially perturb the ground state during the interaction time. The derived solution reveals the origin of pulse formation, which is the interruption of the resonant interaction between the VUV radiation and the atoms near the crests of the IR field strength and resumption of the resonant interaction within extremely short time-intervals in the vicinity of zero-crossings of the IR field. Comparison of the derived analytical solution to the results of numerical calculations within the more general models [17, 23] shows excellent agreement both for helium and atomic hydrogen. In such a way, the effect of extremely short pulse formation due to ionization switching of the resonant interaction is shown to be insensitive to the specific properties of an atom (such as dependence of Stark shift and excited-state ionization rate on the IR field strength), and can be realized under conditions of tunnel ionization in arbitrary atomic gas, possessing spectrally isolated VUV transition from the ground to a bound excited state and a sufficiently high ionization potential from this excited state. Since the carrier frequency of the pulses lies below the ionization potential of neutral atoms and can be varied via the choice of generating atoms and a particular transition, the proposed technique for extremely short pulse formation constitutes a promising tool for nondestructive steering of ultrafast dynamics of the bound electrons inside atoms, molecules, and solids. The discussed approach can be straightforwardly applied for the shortening of pulses of existing extreme-ultraviolet and soft-x-ray lasers [34] towards the attosecond duration. It is worthwhile to mention, that although the present

paper is devoted to an analytical description of the attosecond pulse train formation, as it was shown in our previous work [18], the same mechanism of interruption of the resonant interaction via tunnel ionization allows formation of a single attosecond pulse by means of techniques similar to those used for creation of isolated attosecond pulses via high harmonic generation, namely, by using a short (as compared to the period of the IR field) incident VUV radiation pulse, IR radiation pulse with a steep front edge, or fast polarization switch of the IR field.

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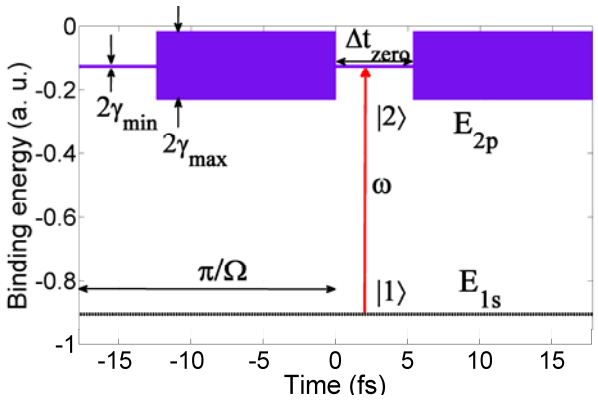

FIG. 1. (Color online) Sketch of the theoretical model used for derivation of the analytical solution. The incident HF radiation with frequency ω selects the lower, $|1\rangle$, and upper, $|2\rangle$, states of the resonant atomic transition, possessing the energy E_{1s} and E_{2p}, respectively. The LF field with frequency Ω rapidly ionizes atoms from the excited state $|2\rangle$ leading to periodic broadening of the corresponding energy level twice within the LF field cycle. At the same time, the ground atomic state $|1\rangle$ is unaffected by the LF field. The linewidth of the resonant transition $|1\rangle \leftrightarrow |2\rangle$ takes the minimum value $2\overline{\gamma}_{\min}$ during the time-intervals Δt_{zero} near zero-crossings of the LF field and the maximum value $2\overline{\gamma}_{\max}$ during the time-intervals $\pi/\Omega - \Delta t_{\text{zero}}$ near the LF field crests.

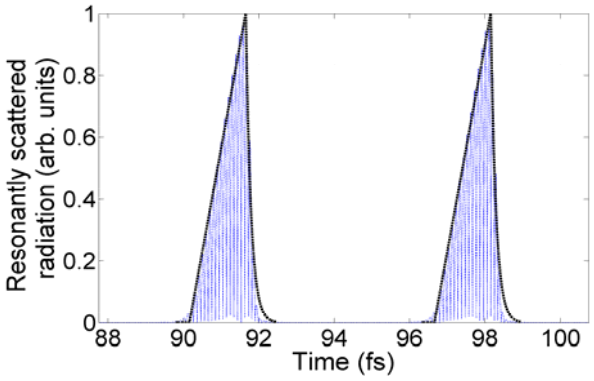

FIG. 2. (Color online) Time-dependence of radiation, resonantly scattered by the atoms of helium under the simultaneous action of the incident 58.4 nm VUV radiation, exciting 1s ↔ 2p atomic transition, and 3.9 μm IR radiation with intensity $I_{IR}=1.5\times10^{13}$ W/cm^2, rapidly ionizing atoms from the 2p state twice within the IR field cycle. The resonant scattering is confined to the time intervals Δt_{zero} near zero-crossings of the IR field. For the rest of time the resonant scattering is suppressed due to the huge broadening of the resonant transition line, see Fig. 1. The black solid line corresponds to the radiation envelope, calculated analytically (15), (16). The rapidly oscillating blue dashed line corresponds to absolute value of the radiation strength, calculated numerically within the three-level model [22], taking into account the nonadiabatic coupling between 2p and 2s states of helium, as well as instantaneous quadratic Stark shift of the corresponding energy levels and tunnel ionization from them.

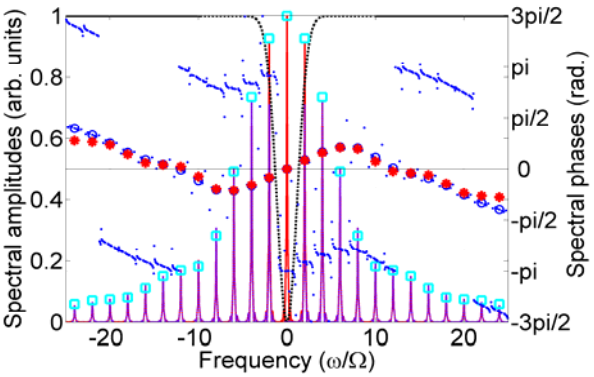

FIG. 3. (Color online) Fourier transform of the slowly-varying envelope of the resonantly scattered radiation plotted in Fig. 2. The result of analytical calculation for the spectral amplitudes of the scattered radiation using equations (18) and (19) is shown by cyan squares. Amplitudes of the spectral components calculated numerically within the three-level model [22] (see caption to Fig. 2) before and after spectral filtering (see the text) are shown by red solid and bold lavender dashed line, respectively (this lines almost overlap except for the three central spectral components). Transmission of the spectral filter, which is zero at the resonance frequency (zero frequency of the envelope) and unity far away from it, is shown by black dashed line. The result of analytical calculation for the spectral phases of the scattered radiation using equations (18) and (19) is shown by red stars. The numerically calculated [22] phases of the spectral components at the multiples of the doubled IR field frequency, ±2Ω, ±4Ω, ... are marked by blue cycles.

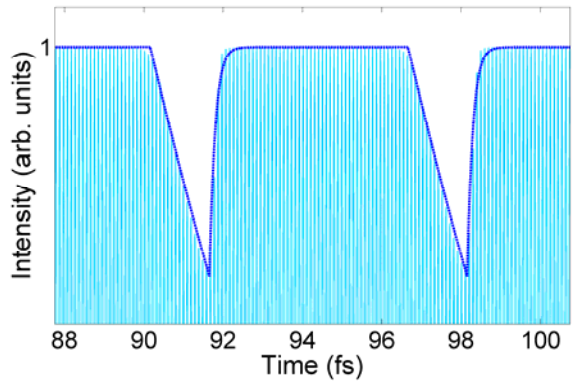

FIG. 4. (Color online) Intensity of the output VUV radiation, propagated through an optically thin medium of helium, simultaneously irradiated by the resonant 58.4 nm VUV radiation and the strong 3.9 μm IR field (see caption to Fig. 2). The output radiation results from coherent summation of the resonantly scattered radiation (Fig. 2) with the incident one. The blue solid line represents the result of analytical calculation for the envelope of the output VUV intensity. The rapidly oscillating dashed cyan curve corresponds to the square of the normalized VUV radiation strength, calculated numerically within the model [22]. It worth noting, that the presented calculations imply approximation of an infinitely thin medium, so that the depth of intensity dips in Fig. 4 is much smaller than unity.

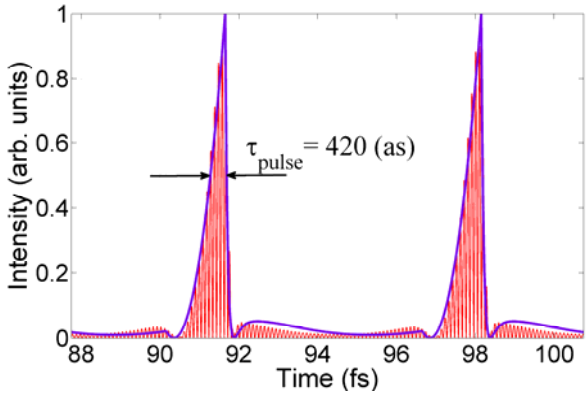

FIG. 5. (Color online) Intensity of the attosecond pulses produced from the output VUV radiation (see Fig. 4) via suppression of the resonant components of its spectrum according to Fig. 3. The lavender solid line represents the result of analytical calculation for the envelope of the pulses. The rapidly oscillating dashed red curve corresponds to the square of the normalized VUV radiation strength, calculated numerically within the model [22]. The pulse duration is τ_{pulse}=420 as, the pulse repetition period equals half-cycle of the IR field, T=6.5 fs.

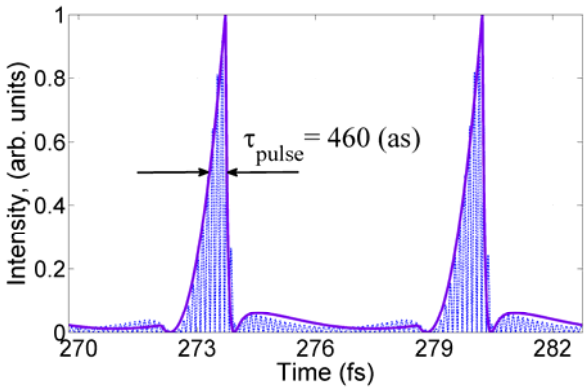

FIG. 6. (Color online) Time-dependence of intensity of the attosecond pulse train, produced from 73.6 nm VUV radiation in neon, dressed by 3.9 μm IR field with intensity $I_{IR}=5\times10^{13}$ W/cm^2, via the resonant interaction with $2s^22p^6 \leftrightarrow 2s^22p^5(^2P^0_{1/2})3s$ atomic transition and suppression of the resonant component of the output VUV spectrum. The solid lavender curve represents the analytical solution for the envelope of the pulses. The rapidly oscillating blue curve corresponds to the square of the normalized VUV radiation strength, calculated numerically within the eight-level model [23], which takes into account transitions between the ground atomic state $2s^22p^6$ and the excited states $2s^22p^53s$ and $2s^22p^53p$, coupled to each other by the VUV radiation and the IR field. The latter also induces time-dependent ionization from all the excited atomic states. The pulse duration is τ_{pulse}=460 as, the pulse repetition period equals half-cycle of the IR field, T=6.5 fs.

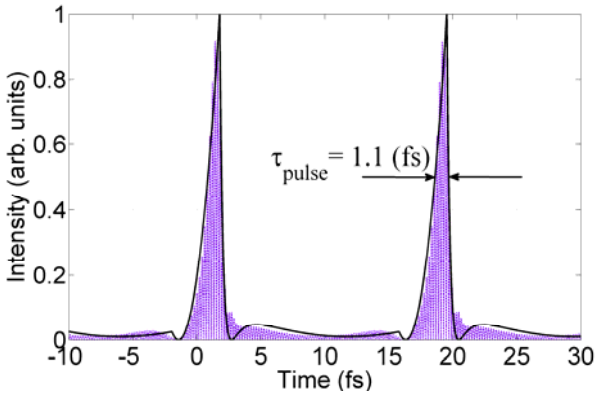

FIG. 7. (Color online) Intensity of the ultrashort pulses, produced from 122 nm VUV radiation due to the resonant interaction with $1s \leftrightarrow 2p$ transition of atomic hydrogen, dressed by 10.65 μm CO_2-laser field with intensity $I_{CO2}=2.2\times10^{13}$ W/cm^2, via suppression of the resonant component of the output VUV spectrum. The solid black curve represents the analytical solution for the envelope of the pulses. The rapidly oscillating lavender curve corresponds to the square of the normalized VUV radiation strength, calculated numerically within the three-level model [16], which takes into account both tunnel ionization from the excited atomic states $|2\rangle=(|2s\rangle+|2p\rangle)/\sqrt{2}$ and $|3\rangle=(|2s\rangle-|2p\rangle)/\sqrt{2}$, and Stark shift of the corresponding energy levels. The pulse duration is τ_{pulse}=1.1 fs, the pulse repetition period equals half-cycle of the CO_2-laser field, T=17.8 fs.