Formation of a Single Attosecond Pulse via Interaction of Resonant Radiation with a Strongly Perturbed Atomic Transition

V. A. Antonov, Y. V. Radeonychev, and Olga Kocharovskaya

Phys. Rev. Lett. **110**, 213903 — Published 21 May 2013

DOI: 10.1103/PhysRevLett.110.213903

Formation of a single attosecond pulse via interaction of resonant radiation with a strongly perturbed atomic transition

V. A. Antonov[1,2], Y. V. Radeonychev[1,2], Olga Kocharovskaya[2]

[1] *Institute of Applied Physics, Russian Academy of Sciences, 46 Ulyanov Street, Nizhny Novgorod, 603950, Russia,*
[2] *Department of Physics & Astronomy, Texas A&M University, College Station, TX 77843-4242, USA.*

Abstract – We propose a technique to form a single few-cycle attosecond pulse from vacuum ultraviolet (VUV) or extreme ultraviolet (XUV) radiation via resonant interaction with hydrogen-like atoms, irradiated by a high-intensity far-off-resonant laser field. The laser field strongly perturbs excited atomic energy levels via the Stark effect and ionizes atoms from the excited states. We show that an isolated attosecond pulse can be formed using either a short incident femtosecond pulse of the resonant radiation or a steep front edge of the laser field. We propose an experimental realization of a single subfemtosecond pulse formation at 121.6 *nm* in atomic hydrogen and a single sub-100 *as* pulse formation at 13.5 *nm* in Li^{2+} plasma.

Remarkable progress has been achieved in the last decade in understanding and control of the electron dynamics in atoms, molecules, and solids unfolding on the attosecond time scale [1, 2]. This progress became possible due to development of the attosecond pulse formation technique, based on high harmonic generation (HHG) of laser radiation in gases [1, 2]. This technique resulted in birth and flourishing of the attosecond physics, but is characterized by low efficiency on the order of 10^{-6} and requires an external adjustment of phases of the generated spectral components and a spectral filtering of the output radiation [3].

Another method of sub-femtosecond pulse generation is based on stimulated Raman scattering (SRS) of a biharmonic laser radiation [4, 5]. It provides a remarkably high efficiency up to 10^{-2}, but does not allow production of an isolated subfemtosecond pulse, which severely limits the scope of its spectroscopic applications. Similarly to the HHG technique, the SRS method relies on the external adjustment of phases of the generated spectral components.

A different approach to extremely short pulse formation is based on interaction of an ultraintense laser radiation with an overdense plasma mirror [6]. It is promising for generation of zeptosecond hard-X-ray pulses, but requires an extremely high intensity, short duration and high contrast of the laser radiation pulse.

Recently, a method to produce a train of few-cycle femto- and attosecond pulses from VUV or XUV radiation resonant to an atomic transition in a hydrogenlike medium, irradiated by a high-intensity low frequency (LF) laser field, was proposed [7-10]. It is based on time-dependent perturbation of the excited atomic energy levels over the LF field cycle due to Stark effect and ionization, produced by the LF field. The pulses are formed from the VUV or XUV radiation without external adjustment of phases of the generated sidebands. The efficiency of the method can be as high as in the case of SRS.

In this paper we show, that a *single* extremely short almost bandwidth-limited attosecond pulse can be produced from VUV or XUV radiation via resonant interaction with a transition of hydrogen-like atoms, strongly perturbed by an intense LF laser field, using either a short incident femtosecond pulse of the resonant radiation or a steep front edge of the laser field. In particular, this method, unlike the other techniques, is promising for generation of an isolated attosecond pulse in the soft-VUV spectral range at the photon energy around 10 *eV*. Since the ionization potentials of various chemical compounds lay just in this domain, such pulses are especially important for the attosecond nonlinear and photoionization-based spectroscopy. Another straightforward application could be shortening of the pulses of existing XUV-, X-ray-, and free-electron-lasers [11].

Let us consider propagation of a high frequency (HF) linearly polarized VUV/XUV radiation through a gas of hydrogen-like atoms. At the front edge of the medium, z=0, it has the form

$$\vec{E}_{inc}(t) = \frac{1}{2}\vec{x}_0 A_{inc}(t)\exp\{-i\omega t\} + C.C. \quad (1)$$

where ω is the angular frequency and $C.C.$ stands for complex conjugation. The incident radiation is quasi-monochromatic, $|dA_{inc}/dt| << \omega$, and close to resonance with the transition from the ground to the first excited energy level n=1↔n=2 (n is the principal quantum number): $|\omega - \omega_{21}^0| << \omega_{21}^0$, ω_{21}^0 is the unperturbed transition frequency.

The medium is simultaneously irradiated by an intense linearly polarized LF laser field,

$$\vec{E}_{LF}(t) = \frac{1}{2}\vec{x}_0 A_C(t - z/c_{LF})\exp\{-i\Omega(t - z/c_{LF})\} + C.C., \quad (2)$$

where $c_{LF} = c/\sqrt{\varepsilon_{LF}}$, c is the speed of light in vacuum, $\varepsilon_{LF} \cong 1$ is the dielectric permittivity of the medium at the frequency Ω. The amplitude of the LF field is insufficient for ionization or multiphoton excitation of atoms from their ground state, while the frequency of the LF field is much smaller than the frequencies of all atomic transitions from both the ground and the first excited energy levels. The length of the medium, L, is substantively smaller than the lengths of dispersive broadening, self-focusing, nonlinear spectral broadening, and the Rayleigh length of the LF beam. These conditions allow neglecting the difference between the phase and group velocities of the LF field (2) as well as distortions of the LF field during its propagation through the medium. Hereinafter we omit the vector character of the fields, since their polarization is defined by the incident radiation. The HF and LF radiation pulses propagate along the same axis and overlap in space and time.

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

$$\frac{\partial^2 E_{HF}}{\partial z^2} - \frac{1}{c_{HF}^2}\frac{\partial^2 E_{HF}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P}{\partial t^2}, \quad (3)$$

where $c_{HF} = c/\sqrt{\varepsilon_{HF}}$, $\varepsilon_{HF} \cong 1$ is the dielectric permittivity caused by non-resonant interaction with the medium, and P is the resonant polarization, induced at the atomic transition n=1↔n=2. Equation (3) strictly describes both the resonant absorption and dispersion of the medium, while the non-resonant effects are not important at the considered propagation length.

According to the selection rules, the VUV/XUV radiation, polarized along the polarization of the LF field, couples the ground state, $|1\rangle = |100\rangle$, and the excited atomic states $|2\rangle = (|200\rangle + |210\rangle)/\sqrt{2}$ and $|3\rangle = (|200\rangle - |210\rangle)/\sqrt{2}$ [7-10], which are the eigenstates of hydrogen-like atom in an external electric field [12] (numerals $|nlm\rangle$ label principal, orbit, and magnetic quantum numbers, respectively). Transitions to the upper energy levels, n=1↔n≥3, are not important, since (i) they are nonresonant, (ii) their dipole moments are substantively smaller than the dipole moment of the resonant transition, and (iii) the highly excited bound atomic states are demolished by ionization induced by the LF field. The resonant polarization is given by

$$P(z,t) = N\{2d_{tr}\,\mathrm{Re}\{\rho_{21} - \rho_{31}\} + d_{ex}(\rho_{22} - \rho_{33})\}, \quad (4)$$

where N is the concentration of the atoms, $d_{tr} = d_{21} = -d_{31} = 2^7 e a_0/(3^5 Z)$ is the dipole moment of the resonant transitions $|1\rangle$↔$|2\rangle$ and $|1\rangle$↔$|3\rangle$ (e is the charge of electron, a_0 is the Bohr radius, Z is the atomic number), $d_{ex} = d_{22} = -d_{33} = 3ea_0/Z$ is the mean dipole moment of atom in the excited state $|2\rangle$ or $|3\rangle$ [12], and ρ_{ab} are the elements of the density matrix, $a,b = 1,2,3$.

Dynamics of the density matrix elements is described by the equations

$$\frac{\partial \rho_{ab}}{\partial t} + (i\omega_{ab}(z,t) + \gamma_{ab}(z,t))\rho_{ab} = \frac{i}{\hbar}\sum_s (\rho_{sb}d_{as} - \rho_{as}d_{sb})\cdot E_{HF}, \quad (5)$$

where $a,b,s=1,2,3$; ω_{ab} is the frequency of atomic transition $|a\rangle\leftrightarrow|b\rangle$, d_{as} and γ_{ab} are the dipole moment and the decoherence rate of the corresponding transitions, and $\hbar$ is the Plank's constant.

The LF field causes quasi-static Stark splitting of the excited atomic energy level and tunnel ionization from the excited states $|2\rangle$ and $|3\rangle$, resulting in the *space-time variation* of frequencies and decoherence rates of the atomic transitions over the LF-field cycle:

$$\begin{aligned}\omega_{ab}(z,t)&\equiv\left(\mathrm{E}_a(z,t)-\mathrm{E}_b(z,t)\right)/\hbar,\\ \gamma_{ab}(z,t)&\equiv\gamma_{ab}^0+\frac{1}{2}\left(w_{ion}^{(a)}(z,t)+w_{ion}^{(b)}(z,t)\right).\end{aligned}\tag{6}$$

Here $\mathrm{E}_s(z,t)$ is the instantaneous energy of the $|\mathrm{s}\rangle$ state, $w_{ion}^{(s)}(z,t)$ is the instantaneous ionization rate from it, γ_{ab}^0 is the decoherence rate of transition $|a\rangle\leftrightarrow|b\rangle$ in the absence of the LF field. Since the LF field frequency is much smaller than the frequencies of all the atomic transitions from the first excited energy level, the instantaneous values of energies of the excited states $|2\rangle$ and $|3\rangle$ and ionization rates from them are determined by the instantaneous strength of the LF field. Within the perturbation theory, energies of the states $|2\rangle$ and $|3\rangle$ are given by [13]

$$\mathrm{E}_{2;3}(z,t)=-\frac{m_e e^4 Z^2}{8\hbar^2}\left(1\pm 3F_{LF}+\frac{21}{2}F_{LF}^2\pm\frac{195}{8}F_{LF}^3\right),\tag{7}$$

where m_e is the mass of electron, $F_{LF}(z,t)=\left(2/Z\right)^3 E_{LF}(z,t)/E_A$ is the normalized LF field strength and $E_A=m_e^2e^5/\hbar^4$. The rates of tunnel ionization from the excited states $|2\rangle$ and $|3\rangle$ have the form [13]

$$w_{ion}^{(2;3)}(z,t)=\frac{m_e e^4 Z^2}{8\hbar^3}\left(\frac{4}{|F_{LF}|}\right)^{2\pm s}\exp\left\{-\frac{2}{3|F_{LF}|}\mp 3s\right\},\tag{8}$$

where $s=\mathrm{sgn}\{F_{LF}(z,t)\}$. The Stark shift of the ground atomic state $|1\rangle$ and the ionization from it are negligible [12, 13].

The resulting system (2)-(8) is solved numerically using the initial conditions $\rho_{11}(z,t=0)=1$ and $\rho_{ab}(z,t=0)=0$ if $(a,b)\neq(1,1)$, assuming that the atoms are initially at the ground state, and the boundary condition $E_{HF}(z=0,t)=E_{inc}(t)$. The solution implies proximity of nonresonant permittivities at the frequencies of the VUV/XUV and the LF radiation, $\frac{\Omega}{c}\left(\sqrt{\varepsilon_{HF}}-\sqrt{\varepsilon_{LF}}\right)L<<\pi$. This inequality generally holds for a rarefied neutral gas. The role of the plasma dispersion is discussed below.

The sub-LF-field-cycle variation of the resonant transition frequencies and decoherence rates makes the atomic response to the VUV/XUV radiation ultra-broadband, providing a possibility to enrich the spectrum of radiation by phase matched sidebands and produce extremely short pulses. In the case of a relatively weak LF field, $\max\{F_{LF}\}\le 0.05$, pulse formation relies on Stark splitting of the first excited atomic energy level (7) [7, 9], which in this case is much larger than the peak ionization rates from the excited states (8). In the case of a strong LF field, $\max\{F_{LF}\}\ge 0.15$, which is considered below, the peak values of ionization rates (8) substantively exceed Stark splitting of the excited energy level (7), Fig. 1. In this case, the ultimately short pulses can be produced from the incident VUV/XUV radiation due to opening a channel for the resonant atomic transitions (otherwise suppressed because of overwhelming ionization from the excited states) only for extremely short periods of time near zero-crossings of the LF field (where excited-state-ionization rates are close to zero), Fig. 1, [8,10]. The produced sidebands are phase-matched, if the Rabi frequency of the incident VUV/XUV radiation does not exceed inverse duration of an extremely short time-

interval near zero-crossing of the LF field, where the resonant interaction takes place, Fig. 1. This condition is generally fulfilled for all the sources of HF radiation, considered below.

In order to produce an isolated attosecond pulse, the broadband atomic response should be confined within a time interval shorter than the half-cycle of the LF field. Below we discuss two options to meet this condition: (i) a short femtosecond incident VUV/XUV radiation pulse or (ii) a steep front edge of the LF field pulse. In the former case, the resonant atomic response occurs only within a short time interval, while in the latter case, the resonant atomic response is broadband during only a short time interval. Possible experimental realizations in atomic hydrogen and in a gas of Li^{2+} ions are discussed below.

First, let us consider the possibility to produce an isolated subfemtosecond pulse from 121.6 *nm* VUV radiation in atomic hydrogen irradiated by the mid-IR laser field [14]. The 90 GW pulse at 3900 *nm*, loosely focused to peak intensity $I_{LF}=2.2\cdot10^{13}$ W/cm^2, rapidly ionizes atoms from the excited states $|2\rangle$ and $|3\rangle$ but does not perturb the unexcited atoms. In accordance with (8), the excited-state-ionization rates rise up to extremely high values and drop to zero during each half-cycle of the IR field, Fig. 1, resulting in switching on and off the resonant interaction between the atoms and the VUV radiation twice within the IR field cycle [8]. Let the incident VUV radiation has a form of the pulse centered at zero-crossing of the IR field, with duration equal to the half-IR-field-cycle, 6.5 *fs* [15, 16]. Then it resonantly excites atoms during only a single extremely short time interval between the adjacent peaks of the excited state ionization rates, Fig. 1. The incident VUV radiation energy is partially re-emitted into a much broader spectrum, partially transferred to the excited atoms and carried away by the ionized electrons. The generated sidebands are almost in-phase with each other and in antiphase with the incident radiation, resulting in vanishing VUV radiation strength in the vicinity of zero-crossing of the IR field after propagation through 1.4 *mm* long medium with concentration of atomic hydrogen $N_H=10^{18}$ cm^{-3}, Fig. 2. The bandwidth of the generated spectrum, Fig. 3, is determined by inverse time-scale of the extremely short dip in the output VUV radiation strength, Fig. 2 resulting from extremely fast switch on and off the resonant absorption near zero-crossing of the IR field due to the highly nonlinear dependence of ionization rates on the IR field strength. The output subfemtosecond pulse, shown in Fig. 4, arises after suppression of the resonant component of the VUV spectrum by a narrowband spectral filter, Fig. 3. The pulse duration is 800 *as*, corresponding to 1.15 of its Fourier limit, and enclosing 2 cycles of the VUV carrier. The peak pulse intensity $I_{max}=6\cdot10^9$ W/cm^2 equals 0.68 of peak intensity of the incident VUV radiation $I_0=9\cdot10^9$ W/cm^2. At this intensity, on average, only one of 10^4 atoms is excited by the VUV radiation and subsequently ionized by the IR field, and hence the dispersion of produced plasma is negligible.

Another way to form an isolated almost bandwidth-limited attosecond pulse is based on use of a steep front edge of the laser field. We consider the possibility of a sub-100 *as* pulse formation from 13.5 *nm* XUV radiation, resonant to the transition $n=1\leftrightarrow n=2$ of Li^{2+} hydrogen-like ions. The sources and optics at this wavelength are extensively developed in application to the XUV lithography [11, 17]. Apart from radiation of the Li^{2+} H-like laser at 13.5 *nm*, the Ni-like Ag-laser or the Ni-like Cd-laser radiation at 13.9 *nm* and 13.2 *nm*, respectively, can be used as the quasi-resonant field [11], while ultrashort visible or near-infrared pulses can serve as the LF field. A quasi-cw arbitrary shaped XUV radiation should enter the medium prior to the LF field and induce a resonant response of the ions. When the amplitude of the resonant polarization reaches its maximum value, the LF pulse enters the medium and ionizes majority of the excited Li^{2+} ions by its leading edge, canceling the resonant interaction. If the front edge of the LF pulse is steep enough, the transient response of the ions to the XUV radiation is extremely short, corresponding to generation of the phase matched XUV sidebands. In such a case, suppression of the resonant component of the output spectrum, antiphased to the sidebands, results in formation of an extremely short attosecond pulse. We consider the LF field at carrier wavelength 400 *nm* with duration 4 *fs*, enclosing 3-cycles under the 4-th power super-Gaussian envelope, with peak intensity $I_{LF}=1.6\cdot10^{16}$ W/cm^2 [18, 19]. The incident 13.5 *nm* XUV radiation intensity is $I_0=6\cdot10^{12}$ W/cm^2. The attosecond pulse, shown in Fig. 5, is produced as a result of propagation of the XUV radiation through 100 μm region with concentration of Li^{2+} ions

N_{Li2+}=2.1·10^{18} *cm*$^{-3}$ and suppression of the central component of the output spectrum by a narrow-band Si:Mo multilayer mirror [17]. The peak pulse intensity I_{max}=1.3·10^{12} *W*/*cm*^{2} equals 0.2 of the incident XUV intensity I_0. The pulse duration is equal to 80 *as*, enclosing less than 2 cycles of the XUV carrier. The dispersion of doubly ionized plasma with concentration of free electrons N_e=4.2·10^{18} *cm*$^{-3}$ results in the pulse broadening $\Delta\tau_{\text{pulse}} = \frac{L}{c}\left(\sqrt{\varepsilon_{HF}} - \sqrt{\varepsilon_{LF}}\right) \cong \frac{2\pi N_e e^2 L}{m_e \Omega^2 c} \approx 100$ *as*, not shown in Fig. 5. The broadening can be reduced to $\Delta\tau_{\text{pulse}}$=20 *as* via fivefold decrease of the density of Li^{2+} ions or the interaction length at the cost of reduction of the peak pulse intensity from I_{max}=0.2·I_0 to I_{max}=0.01·I_0. Another possibility relies on quasi-phase-matching: use of the quasi-phase-matching structure [20] would allow one to reduce the pulse broadening below 20 *as* without its accompanying attenuation and to produce a sub-100 *as* pulse, similar to that, shown in Fig. 5.

It is worth noting another scenario of an isolated attosecond pulse formation, based on rapid change of polarization state of the LF field, similar to the polarization gating [21]. In a circularly polarized LF field, the resonant interaction between the HF radiation and the atoms is *constantly* suppressed since the excited energy levels are extremely broadened via *time-independent* ionization. In a linearly polarized LF field, instead, the resonant interaction resumes in the vicinities of zero-crossings of the LF electric field, leading to generation of the HF sidebands. Thus rapid change of the polarization state of the LF field from circular to linear and then back to circular might allow one generation of an isolated attosecond burst.

The discussed approach to form a single extremely short attosecond pulse from resonant radiation is not limited to a hydrogen-like medium. It may be implemented in noble gases, where a sub-femtosecond pulse can be produced from a VUV radiation in the wavelength range 60-125 *nm*.

We are grateful to Mikhail Yu. Ryabikin and Paul B. Corkum for the fruitful discussions. This research was supported by RFBR, grants # 10-02-01250, # 12-02-31325, # 12-02-12101-ofi_m, Ministry of Science and Education of Russian Federation, contract # 11.G34.11.0011, Federal Target Program "Research and development in priority directions of development of Russian scientific-technological complex" for 2007-2012 contract # 07.514.11.4162, and NSF grant # 0855688. V.A. Antonov acknowledges a personal grant for young scientists from the Foundation "Dynasty".

1 P. Agostini, L. F DiMauro, Rep. Prog. Phys. **67**, 813 (2004).
2 F. Krausz, M. Ivanov, Rev. Mod. Phys. **81**, 163 (2009).
3 E. Gustafsson *et. al.*, Opt. Lett. **32**, 1353 (2007).
4 M. Y. Shverdin, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **94**, 033904 (2005).
5 W.-J. Chen *et al.*, Phys. Rev. Lett. **100**, 163906 (2008).
6 C Thaury and F Quéré, J. Phys. B: At. Mol. Opt. Phys. **43**, 213001 (2010).
7 Y. V. Radeonychev, V. A. Polovinkin, O. Kocharovskaya, Phys. Rev. Lett. **105**, 183902 (2010).
8 V. A. Polovinkin, Y. V. Radeonychev, O. Kocharovskaya, Opt. Lett. **36**, 2296 (2011).
9 Y. V. Radeonychev, V. A. Polovinkin, O. Kocharovskaya, Las. Phys. **21**, 1243 (2011).
10 Y. V. Radeonychev, V. A. Antonov, O. Kocharovskaya, JOSA B, submitted.
11 D Attwood, *Soft X-Rays and Extreme Ultraviolet Radiation: Principles and Applications*. (Cambridge University Press, New York, 2007).
12 L. D. Landau and E. M. Lifshitz, *Quantum mechanics. Non-relativistic theory.* (Pergamon Press, New York, 1991), p. 129.
13 R. J. Damburg and V. V. Kolosov, in *Rydberg States of Atoms and Molecules,* edited by R. F. Stebbings and F. B. Dunning (Cambridge University Press, Cambridge 1983).
14 G. Andriukaitis *et al.*, Opt. Lett. **36**, 2755 (2011).
15 C. E. M. Strauss, D. J. Funk, Opt. Lett. **16**, 1192 (1991).
16 F. Reiter *et al.*, Opt. Lett. **35**, 2248 (2010).
17 M. G. Pelizzo et al., Opt. Exp. **16**, 15228 (2008).

18 O. Dühr et al., Opt. Lett. **24**, 34 (1999).
19 Eiichi Matsubara et al., JOSA B **24**, 985 (2007).
20 A. Willner *et al.*, Phys. Rev. Lett.**107**, 175002 (2011).
21 I. J. Sola *et al.*, Nature Phys. **2**, 319 (2006).

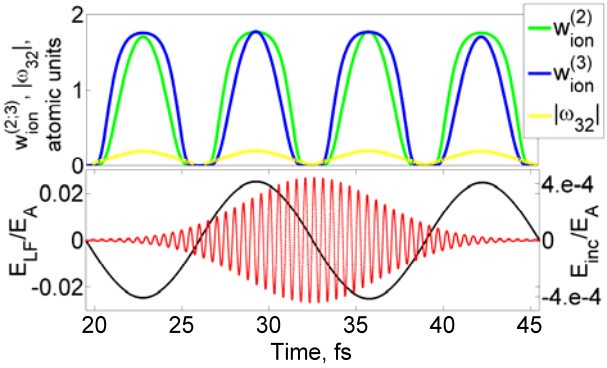

FIG. 1. (Color online) Electric field of the incident VUV radiation pulse, E_{inc} (rapidly oscillating red curve), and of the mid-IR radiation, E_{LF} (bold black curve) in atomic units (bottom panel). Rates of ionization from the excited states $|2\rangle$ and $|3\rangle$ (solid curves) and Stark splitting of the excited energy level, $|\omega_{32}|$, (dashed curve), induced by the mid-IR field (upper panel).

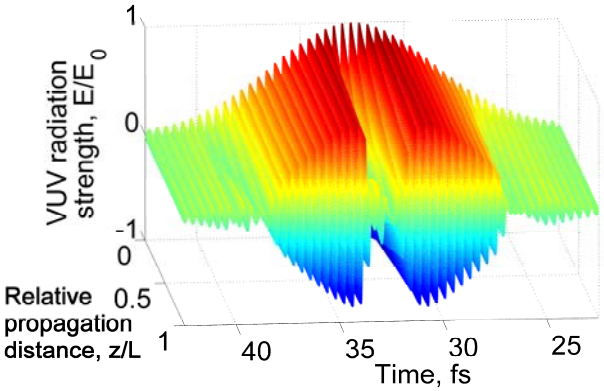

FIG. 2. (Color online) Space-time evolution of the VUV radiation, E, normalized to the peak incident VUV radiation strength, E_0, during its propagation through the medium.

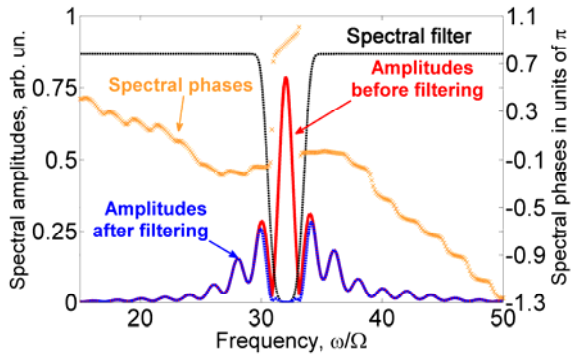

FIG. 3. (Color online) Amplitude spectrum of the output VUV radiation (plotted in Fig. 2, $z=L$) before (red curve) and after (blue curve) spectral filtering. Gold crosses mark spectral phases.

Black dashed curve shows transmission of the spectral filter, which is zero at resonance and unity far away from the resonance.

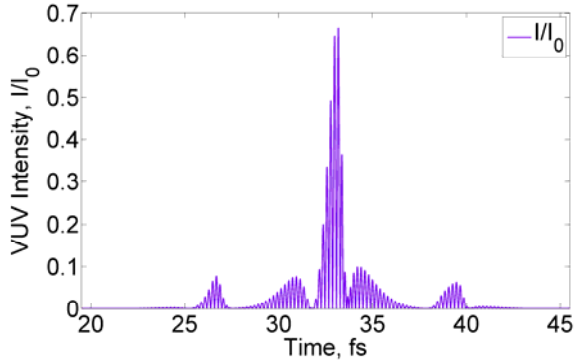

FIG. 4. (Color online) Intensity of the produced subfemtosecond pulse, normalized to the peak incident VUV intensity. The pulse duration is 800 *as*. The peak pulse intensity is $6 \cdot 10^9$ *W/cm*^{2}.

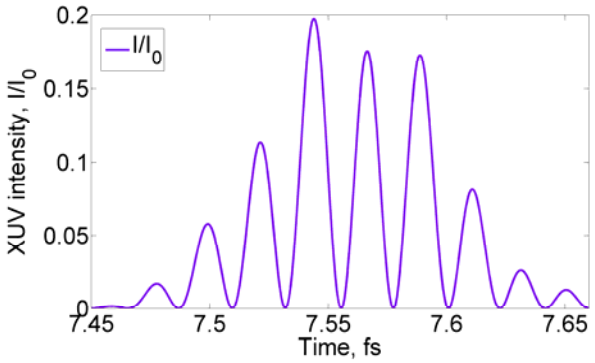

FIG. 5 (Color online) Intensity of the sub-100 *as* pulse, produced from 13.5 *nm* XUV radiation as a result of resonant interaction with Li^{2+} ions (see the text), normalized to the incident XUV radiation intensity. The pulse duration is 80 *as*. The peak intensity of the pulse is $1.3 \cdot 10^{12}$ *W/cm*^{2}.