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Lappas, D. G; L'Huillier, Anne

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PO Box 117
221 00 Lund
+46 46-222 00 00

Generation of attosecond xuv pulses in strong laser-atom interactions

Demetris G. Lappas and Anne L'Huillier

Department of Physics, Lund Institute of Technology, P.O. Box 118, S-221 00 Lund, Sweden

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The generation of radiation pulses that can be as short as 120 attoseconds is demonstrated theoretically. We have employed a two-dimensional exact solution of the Schrödinger equation that allows for arbitrary laser ellipticity. By manipulating the laser ellipticity in time, it is shown that one can control the wave-packet dynamics of the ejected atomic electron. The nonlinear interaction of the electron with the atomic core can thus be restricted to extremely short times. Photon energies up to 160 eV are generated. It is also shown that with a higher laser frequency, even shorter pulses can be produced. [S1050-2947(98)02211-2]

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I. INTRODUCTION

The interaction of a strong laser pulse with an atom results in a variety of phenomena that go beyond the traditional perturbative approach, creating new possibilities for technological advances and a new understanding of light-matter interactions [1]. The generation of very-high-order harmonics in scattered light has not only created a new source of xuv and soft x-ray radiation, but has also shown a way of producing extremely short radiation pulses [2–5]. Although the production of this short-wavelength and ultrashort pulsed radiation has been discussed and predicted in the past [5], only the recent technological advances may allow for an experimental realization. Coherent radiation with photon energies up to 0.5 keV and duration of the order of 10 fs has been produced recently [6,7], and some evidence for short pulses of the order of a femtosecond in the regime of soft x rays has been reported by various groups [7,8].

In the last few years decisive progress has been accomplished in the theoretical understanding of high-order harmonic generation. According to the standard strong-field approximation (SFA) theories, as applied to harmonic generation [9–11], the motion of the ejected electron in the continuum dominates the laser-atom interaction. It is not the electron motion inside the atom alone, but mainly the free-electron wave-packet dynamics that is the source of some typical strong-field phenomena, such as the plateau and the cutoff in the harmonic generation spectrum. In a quasistatic approach, the maximum probability that an electron tunnels into the continuum occurs around the time when the laser field reaches a maximum amplitude [12]. The ejected electron may return to the atomic core, where it scatters, generating a short pulse of bremsstrahlung radiation. The periodicity of this process results in the harmonic peaks in the radiation spectrum [4]. The maximum photon energy is determined by the maximum energy that an electron can release during its recollision with the atomic core, and is given approximately by the formula

$$\omega_{\max} = I_p + 3.2U_p, \quad (1)$$

where I_p is the ionization potential of the atom and U_p the ponderomotive energy that a free electron gains in the laser field [9–11].

An essential element of the generation of radiation in the above theory is the recollision of the electron wave packet with the atomic core. A small ellipticity of the laser would be enough to drive the returning electron wave packet away from the atomic core and significantly reduce the emission of radiation [13]. Efficient emission cannot occur if the laser does not remain linearly polarized for the minimum time that is needed for electron rescattering.

The numerical nonperturbative methods that have been employed for the calculation of harmonic generation spectra in the last few years have mainly dealt with the cylindrically symmetric case of linear laser polarization. First attempts were made with a one-dimensional model [14] and a full three-dimensional calculation with two spatial degrees of freedom [9]. These efforts have been quite successful in describing the strong-field dynamics. However, these approaches do not allow for elliptical polarization. Recent attempts, either with a large basis expansion [15], or based on the direct *ab initio* solution of the two-dimensional Schrödinger equation on a numerical grid that we employ here, have already started to address the case of elliptical polarization.

In the following we show how one can control the wave-packet dynamics in order to achieve emission of high-frequency radiation at well defined times. In Sec. II we explain the laser pulse and its ellipticity properties. In Secs. III and IV we investigate how this particular laser pulse generates ultrashort pulses, by solving the Schrödinger equation numerically. The onset of ionization is discussed in Sec. V, and we conclude in Sec. VI.

II. LASER PULSE WITH TIME-DEPENDENT ELLIPTICITY

The laser pulse that is used in this work consists of two laser pulses with perpendicular linear polarizations. Their frequencies are slightly different, creating a time-dependent phase difference and thus a time-dependent ellipticity. This laser pulse was proposed by Ivanov *et al.* [5], and some experiments have already been performed [16]. The electric field vector of our pulse is

$$\mathbf{E}(t) = E_0 f(t) [\sin(\omega - \Delta\omega)t \cdot \hat{\mathbf{x}} + \cos(\omega + \Delta\omega)t \cdot \hat{\mathbf{y}}], \quad (2)$$

where $f(t)$ is the pulse envelope function, E_0 is the maximum field strength, and ω is the mean carrier frequency of the resulting laser pulse. In a frame of coordinates (x',y') that is rotated by $\pi/4$ radians with respect to (x,y) , the pulse takes the form

$$\mathbf{E}(t) = -E_0 f(t) \sqrt{2} [\sin(\Delta\omega t - \pi/4) \cos(\omega t - \pi/4) \cdot \hat{\mathbf{x}}' + \cos(\Delta\omega t - \pi/4) \sin(\omega t - \pi/4) \cdot \hat{\mathbf{y}}']. \quad (3)$$

At $t=0$ the electric field $\mathbf{E}(t)$ points in the positive y direction and the ellipticity is circular, according to Eq. (3). The ellipticity is given by the ratio

$$\epsilon = \frac{\cos(\Delta\omega t - \pi/4)}{\sin(\Delta\omega t - \pi/4)}. \quad (4)$$

The values of zero or infinity correspond to linear polarization along either the y' or the x' axis.

The pulse shape $f(t)$ is trapezoidal, with two cycles of linear turn-on, followed by two cycles of constant amplitude and then two cycles of linear turn-off, for the field in the x direction. This shape ensures no drift motion in the free-electron displacement in the x direction, at the end of the pulse. For the field in the y direction the same pulse shape is used, although this results in a noninteger number of cycles for the turn-on/off and, therefore, in some free-electron drift motion in the y direction.

Such a short laser pulse is within the current experimental capabilities. A pulse with comparable duration (and constant ellipticity), but with a smoother time profile, has been produced recently by a table-top CPA laser system [8].

For $\Delta\omega = \omega/8$ and $\omega - \Delta\omega = 0.057$ a.u. (wavelength $\lambda = 800$ nm), we show in Fig. 1(a) the trajectory of the tip of the electric field vector, as it is traced in time, for a six-cycle pulse (16 fs, with FWHM ~ 10 fs). A typical, but rather fast-changing time-dependent ellipticity is shown in Fig. 1(b). The time scale is in periods (optical cycles) $T_x = 2\pi/(\omega - \Delta\omega) = 2.7$ fs of the laser in the x direction. We see that the absolute value of the ellipticity remains less than 0.1 for time intervals that are shorter than 1 fs. Linear polarization ($\epsilon = 0$) occurs twice along the y' axis and only once along the x' axis in the middle of the pulse ($t = 3T_x$). Given the fact that the efficiency of high-order harmonic generation decreases dramatically with increasing ellipticity [13,17], we expect the radiation pulses to be emitted only during time intervals that are shorter than 1 fs, when the polarization is approximately linear.

III. GENERATION OF ULTRASHORT PULSES

The time-dependent wave-packet dynamics is described from a direct exact solution of the two-dimensional Schrödinger equation, as in a previous work by Protopapas *et al.* [17]. The only approximations are the two space dimensions and the soft core of the atomic Coulomb potential that is given by

$$V(x,y) = -\frac{1}{\sqrt{a^2 + x^2 + y^2}}. \quad (5)$$

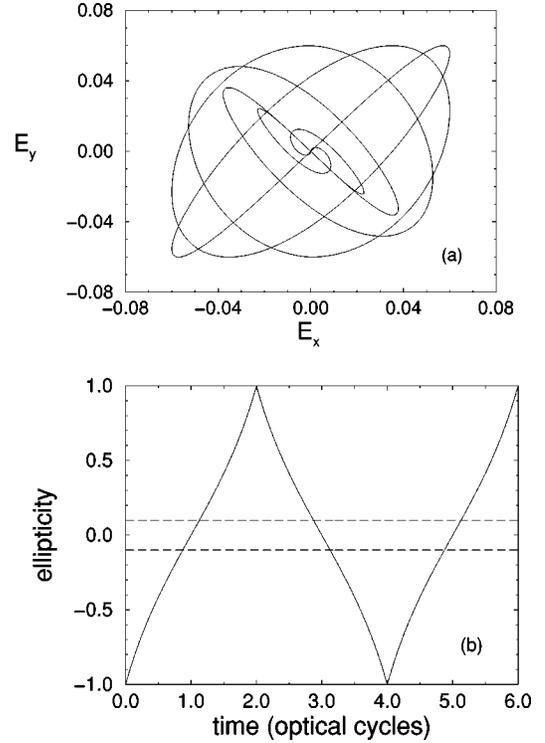


FIG. 1. (a) The trajectory of the tip of the electric field vector of the laser, as it is traced in time, for $\omega - \Delta\omega = 0.057$ a.u. ($\lambda = 800$ nm) and $\Delta\omega = \omega/8$ a.u. (b) The ellipticity of the laser pulse as a function of time. The unit of time, everywhere in the following, is the period of the laser in the x direction, $T_x = 2\pi/(\omega - \Delta\omega)$. The ellipticity starts from circular ($\epsilon = \pm 1$), when the electric field is zero, and ends with circular, again at zero field.

We use the smoothed Coulomb potential [18] in order to avoid numerical problems associated with the singularity at $x=y=0$. The parameter $a=0.8$ ensures that the ground state has the same binding energy as for the hydrogen atom, i.e., -0.5 a.u. We solve the time-dependent Schrödinger equation with the external field in the minimal coupling form. The time evolution is performed with a two-dimensional split-operator method, similar to the one used by Grobe and Eberly, and others [18,19], in their studies of two-electron systems. In this method, the second space derivatives of the kinetic energy operator are expressed exactly in momentum space [20]. An absorber is used to remove any part of the wave packet that reaches the boundaries, so that artificial reflections are avoided. It is assumed that any wave function reaching the end of the grid box represents ionization, and thus the remaining norm gives information about the amount of the resulting ionization. In this work the initial wave function is the ground state that is found by imaginary time integration. A more detailed discussion on various computational aspects can be found in Ref. [18].

For a maximum laser intensity of $I = 2.5 \times 10^{14}$ W/cm² ($E_0 = 0.06$ a.u.), and for the $\lambda = 800$ nm laser, we calculate the time-dependent intensity of the emitted radiation signal in a window of frequencies. The scattered light field is directly proportional to the atomic dipole acceleration, and the detected coherent light spectrum in the forward direction is given by the squared modulus of the Fourier amplitude of the acceleration [21]. After filtering all

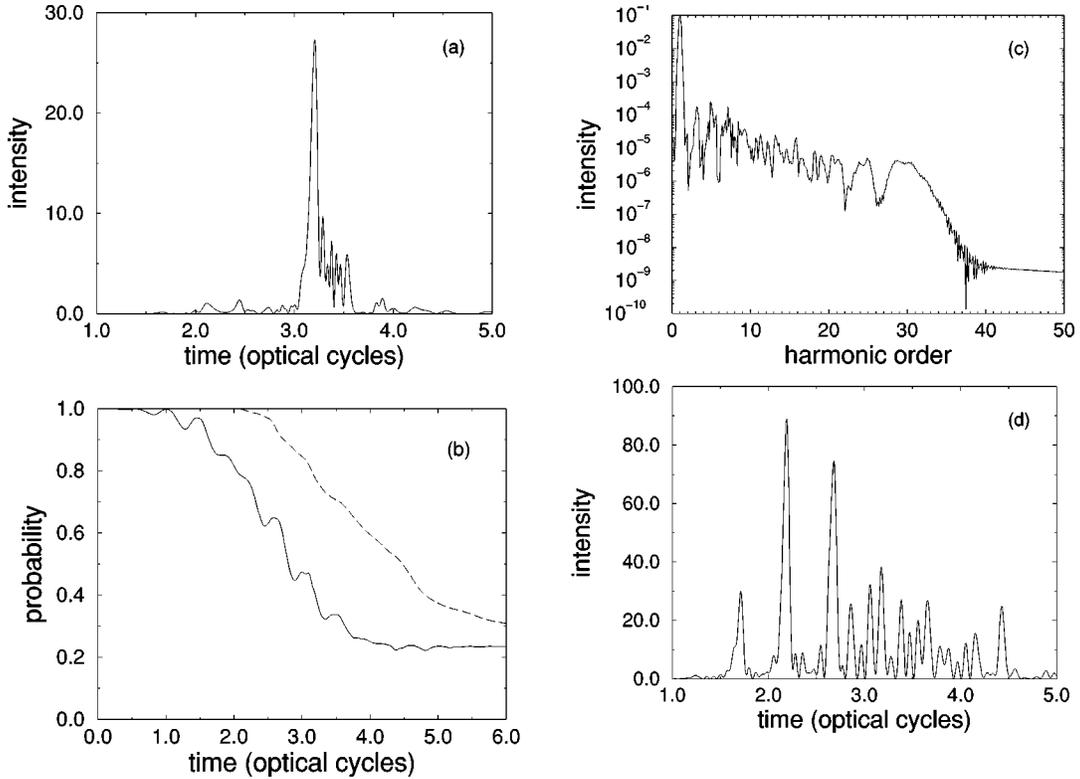


FIG. 2. (a) The instantaneous intensity of the radiation signal for all photon energies, except for the lowest 11 harmonic orders, as given by Eq. (6), along the x' axis. The time scale is in units of T_x . The laser frequency is 0.057 a.u. ($\lambda=800$ nm), and the maximum intensity at the peak of the pulse is $I=2.5 \times 10^{14}$ W/cm 2 . (b) The decay of the ground-state probability (solid curve) and the total norm of the wave function (dashed curve), as functions of time. (c) The total harmonic spectrum of the radiation field that is polarized in the x' direction (see text). The harmonic orders refer to the mean laser frequency ω . (d) Instantaneous intensity of the radiation signal along the x axis, including photon frequencies from the 11th to the 25th harmonic, when the y component of the laser pulse is missing, i.e., with constant linear polarization along the x axis.

photon energies below 11ω , we perform the inverse Fourier transform back to the time domain in order to obtain the generated filtered radiation field $E_f(t)$. The instantaneous intensity is

$$I_f(t) = E_f^*(t)E_f(t) \quad (6)$$

and is shown in Fig. 2(a). The generated pulse has a FWHM of 260 attoseconds and is polarized along the x' axis. In this case the pulse that was used is the one shown in Fig. 1. The polarization first becomes linear in the y' direction at time $t=1T_x$, then circular at $t=2T_x$, linear in the x' direction at $t=3T_x$, circular at $t=4T_x$, and again linear in the y' direction at $t=5T_x$. As we can clearly see from Fig. 2(a), the ‘attosecond pulse’ appears just after the time $t=3T_x$ and is generated by the rescattering of the electron wave packet along the x' direction. Although there is a significant amount of ionization, the remaining norm of the electron wave function is sufficient for a rather efficient and short emission of radiation. In Fig. 2(b) we show both the decay of the ground-state probability (solid line) and the decay of the norm of the electron wave function (dashed line) due to ionization. The norm is reduced to 31% at the end of the pulse. The radiation spectrum is shown in Fig. 2(c). We see the clear formation of a supercontinuum with no harmonic structure for the higher frequencies, especially in the cutoff regime. As has been pointed out by Protopapas *et al.* [4], this form of the har-

monic spectrum appears when there is no periodicity in the rescattering mechanism that could allow for the formation of well separated harmonic peaks. In Fig. 2(d) we show again the time-dependent intensity, but with constant linear polarization along the x axis. In this case, we have multiplied the x component of the electric field by a factor of $\sqrt{2}$, so that the intensity remains the same. In that figure we see the succession of attosecond pulses that is responsible for high-order harmonic generation [3,4]. The lack of apparent perfect periodicity is due to the nonadiabatic turn-on effects.

In this work, we want to isolate only one of these attosecond pulses, in the single-atom response. We achieve this goal by varying the ellipticity during the laser pulse. Furthermore, we do not expect that the propagation effects in our case could significantly alter the generated single-atom pulse. Indeed, it is always the same attosecond pulse that is selected from each atom during the laser pulse, regardless of the intensity, and therefore, independently of the particular atom in the interaction volume. We note that, by changing the characteristics of the incident laser pulse, it could be possible to select another attosecond pulse, i.e., a pulse that corresponds to another electron trajectory. In particular, it might be of interest to choose a trajectory that leads to efficient phase matching [2].

Since it appears that the absolute time duration of the single attosecond pulse depends mainly on the period of the laser field oscillations, it is reasonable to expect that higher

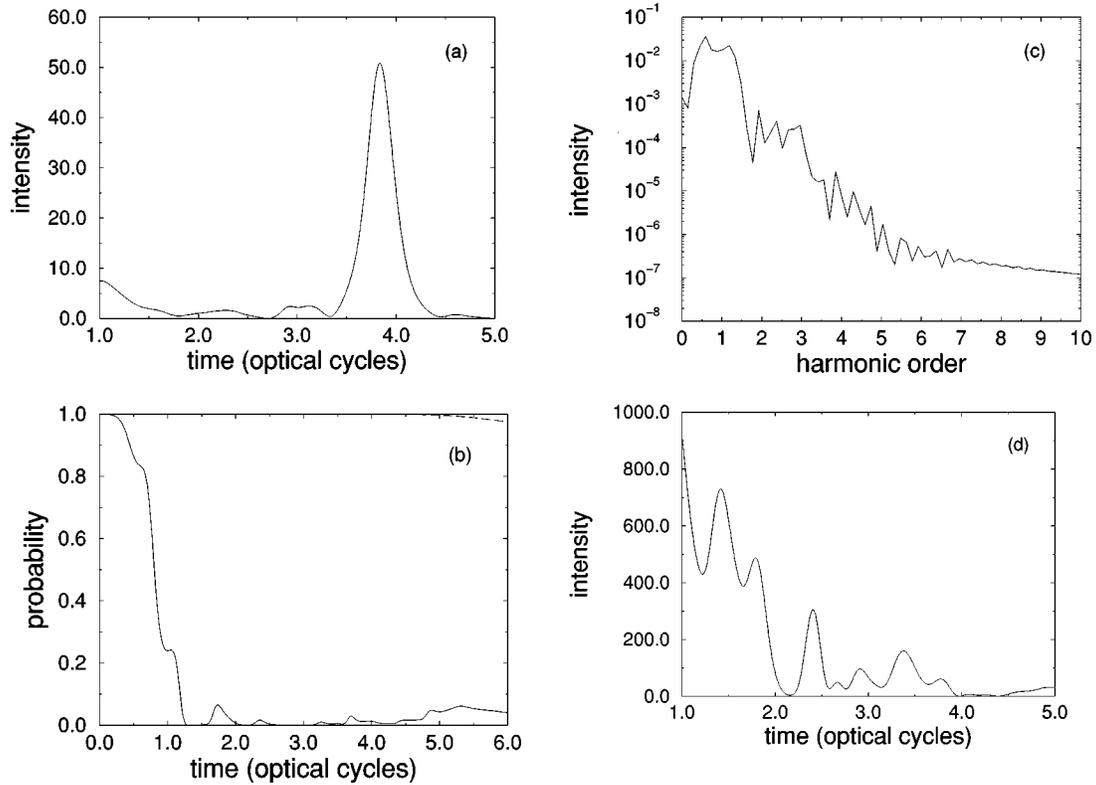


FIG. 3. Same as in Fig. 2, but with frequency 0.4 a.u. ($\lambda=114$ nm) and intensity $I=1.12\times 10^{16}$ W/cm². In (a) the lowest two harmonic orders (i.e., up to frequency 2ω) have been excluded. In (d) photons from 1.5ω to 6.5ω have been included.

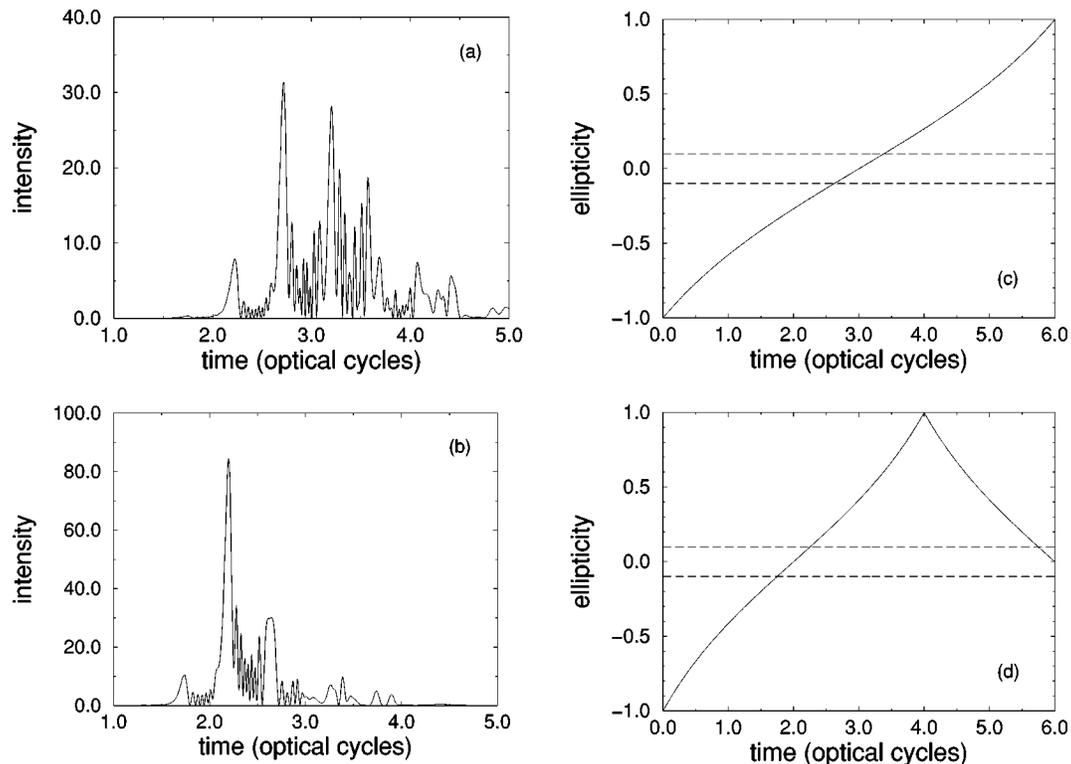


FIG. 4. (a) and (b) The instantaneous intensities of the radiation signal for all photon energies, except for the lowest 11 harmonic orders, as given by Eq. (6), along the y' axis. The time scale is in units of T_x . The laser frequency is 0.057 a.u. ($\lambda=800$ nm) and the maximum intensity at the peak of the pulse $I=2.5\times 10^{14}$ W/cm². (c) and (d) The corresponding ellipticities as functions of time, for the laser pulses of (a) and (b), respectively.

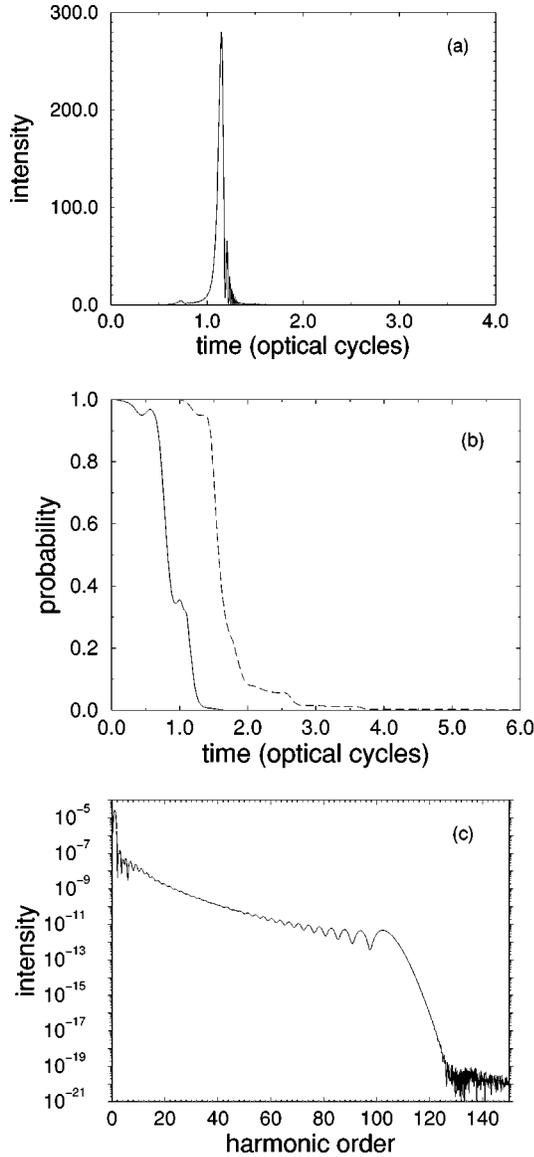


FIG. 5. (a) The instantaneous intensity of the radiation signal for photon energies between 10.5ω and 85.5ω , as given by Eq. (6), along the y' axis. The time scale is in units of T_x . The laser frequency is 0.057 a.u. ($\lambda=800$ nm) and the maximum intensity at the peak of the pulse $I=2.8\times 10^{15}$ W/cm². (b) The decay of the ground-state probability (solid curve) and the total norm of the wave function (dashed curve), as functions of time. (c) The total harmonic spectrum in the direction of the linear polarization along y' (see text). The harmonic orders refer to the mean laser frequency ω .

frequencies can generate even shorter pulses. Without departing from the strong-field tunneling regime, we employ a laser with wavelength $\lambda=114$ nm [$\omega-\Delta\omega=0.4$ a.u. and period $T_x=2\pi/(\omega-\Delta\omega)=0.38$ fs] that could be produced, for example, as the seventh harmonic of the 800-nm laser. In Fig. 3(a) we see the generation of a pulse that has a FWHM of only 120 attoseconds. The pulse is again generated in the x' direction, and only the frequencies above 2ω are included. The laser intensity is much higher than before, $I=1.12\times 10^{16}$ W/cm², to ensure tunneling. As Fig. 3(b) shows, the behavior of the wave packet for these parameters is remarkable: the depletion of the ground state (solid line) is

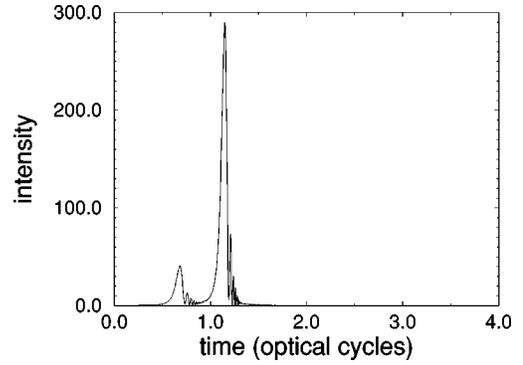


FIG. 6. Same as in Fig. 5(a), but with constant linear polarization along the x axis.

almost complete after only one optical cycle T_x , but the norm (dashed line) has not yet left the box; only 3% of ionization occurs by the end of the pulse. This is due to the fact that, although the Keldysh adiabaticity parameter that determines the tunneling time of the electron ejection out of the ground state is roughly the same as before (in Fig. 2), the displacement of the free electron motion in the high-frequency laser field, being of the order of E_0/ω^2 , is much smaller than in the low-frequency field. This results in a lower amount of ionization, a quantity that is defined here as the total norm that is missing from our numerical box. Therefore, a sufficient amount of probability of the wave-packet distribution is available for rescattering and emission of radiation within the duration of the pulse. Figure 3(c) shows the radiation spectrum in the direction of wave-packet rescattering; the absence of harmonics indicates the lack of periodicity. Finally, Fig. 3(d) illustrates the case of linear polarization along the x axis, where the laser intensity remains the same. We see no obvious periodic pattern, as the atomic ground state decays within one optical cycle. A quick comparison between the vertical scales of Figs. 3(a) and 3(d) shows that the time-dependent ellipticity method selects for rescattering only a very small fraction of the ejected electron packet, for this frequency, thus making the total radiation signal much weaker compared to the case of linear polarization.

IV. IMPORTANCE OF ELLIPTICITY VARIATION

In all of the cases that are described in the previous section the rate of ellipticity variation is rather high: the polarization changes from circular to linear within only one optical cycle. These conditions cannot be easily realized in experiments and could be major obstacles in experimental efforts for attosecond pulse generation with this method. Here, we investigate the effect of the rate of this ellipticity variation and show that the process of selecting a single attosecond pulse from the produced pulse train is indeed very sensitive to how rapidly the ellipticity changes.

When the ellipticity changes from 1 to 0 within three optical cycles, we can hardly separate a single attosecond pulse, as one can see from Fig. 4(a). Even with the slightly faster ellipticity variation of 1 to 0 within two cycles, a single pulse cannot be clearly extracted, as is shown in Fig. 4(b). The respective ellipticities as functions of time (in units of the optical period T_x in the x direction) are shown in Figs.

4(c) and 4(d). Only in the case of the fast variation of Fig. 1(b), with a change of 1 to 0 within one cycle, can one obtain the very “clean” and well distinguishable single radiation pulse that is shown in Fig. 2(a). One should note that in Fig. 4 only the pulse in the y' polarization axis is shown. This is the direction of the first linear polarization in the laser pulse, for all the above three cases, and the only one for the two cases of Fig. 4. Another way to select a single pulse, while keeping a rather slow ellipticity variation, could be the additional effect of the selective propagation and phase matching in the macroscopic medium [2].

V. EFFECT OF IONIZATION

It is well known that when the instantaneous electric field of the laser exceeds a critical value and the laser frequency is low enough to allow for a quasistatic approach [12], the atomic potential can be suppressed sufficiently for the electron to be transmitted over the barrier of the combined atomic and external potentials. This case of over-the-barrier ionization (OBI) is a very fast electron ejection process. Therefore, if one manages to achieve a single recollision before the electron is ionized, a single and extremely short radiation pulse can be emitted. For a maximum pulse intensity of $I = 2.8 \times 10^{15}$ W/cm², at a wavelength of 800 nm, we show in Fig. 5(a) the instantaneous intensity of the emitted radiation pulse in the frequency window from the 11th to the 85th harmonic. The ellipticity variation of the incident laser pulse is the same as in Fig. 2, i.e., the fastest variation that we have used in this work: the laser becomes linearly polarized along the y' axis at the end of the first optical cycle. Just after this moment, the ultrashort pulse is emitted due to the rescattering of the ejected electron wave packet with the atomic core. The FWHM of the pulse is only 160 attoseconds. After this first rescattering event the electron does not return again to the atomic core for more recollisions, but it is ionized very fast, as we can see from Fig. 5(b): the ground-state decays completely within the first optical cycle, and the total norm within the second cycle.

The precise timing of the recollision is consistent with the spectrum of the radiation pulse, shown in Fig. 5(c): the approximate cutoff frequency ω_{\max} corresponds to the laser intensity at the estimated time of the recollision, according to formula (1). However, the fact that the precise timing and the dynamics of the single recollision depend on the instantane-

ous intensity of the laser pulse, which has a spatial dependence in a macroscopic medium, indicates that the attosecond pulse can be generated at different phases of the incident laser pulse for different atoms in the medium. Therefore, a significant broadening of the pulse in time may be expected, and phase matching of this radiation is not guaranteed, unless a spatially homogeneous laser pulse can be used.

When no time-dependent ellipticity is introduced, one can still obtain a very short pulse of radiation before the atom is ionized. However, in this case more than one single subfemtosecond pulse can be generated. In Fig. 6 we show a pair of two pulses generated when the laser polarization is constantly linear along the x axis. Since we have the same intensity as for Fig. 5, the electric field of the laser along the x axis of polarization is multiplied by $\sqrt{2}$. We observe that the only significant difference between the signals of Figs. 5(a) and 6 is the presence of a second short and much weaker pulse that appears only in the case of constant linear polarization (see Fig. 6). We also observe that the strength of the dominant pulse in Fig. 6 is the same as of the single pulse in Fig. 5(a). These results may eliminate the need for time-dependent ellipticity for the generation of ultrashort pulses, if the propagation question is properly addressed.

VI. CONCLUSION

We have achieved the generation of a single 120-attosecond radiation pulse from a single atom interacting with an external laser pulse that has a time-dependent polarization and arbitrary time duration. Two major methods have been employed; one is based exclusively on the time-dependent ellipticity properties of the incident laser, while the second utilizes the ultrafast electron ejection mechanism in a strong external field. Various laser frequencies could be used for generating different time scales of single-pulse durations, with no apparent limit in sight.

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