# Single Circularly Polarized Attosecond Pulse Generation by Intense Few Cycle Elliptically Polarized Laser Pulses and Terahertz Fields from Molecular Media

Kai-Jun Yuan and André D. Bandrauk*

*Laboratoire de Chimie Théorique, Faculté des Sciences, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K 2R1*

(Received 31 July 2012; published 11 January 2013)

We present a method for producing a single circularly polarized attosecond pulse by an intense few cycle elliptically polarized laser pulse combined with a terahertz field from numerical solutions of the time-dependent Schrödinger equation for the molecular ion $H_2^+$. It is found that in the presence of a 62.5 THz ($\lambda = 4800$ nm) field at an intensity of $\sim 10^{14}$ W/cm$^2$, a single circularly polarized 114 as pulse can be generated by an elliptical polarized laser pulse at a wavelength of 400 nm with an ellipticity of $\epsilon = 0.59$. The efficiency of circular polarization attosecond pulse generation is interpreted based on a classical model of single electron recollision with the parent ion.

DOI: 10.1103/PhysRevLett.110.023003 PACS numbers: 33.80.Rv, 42.65.Ky

Generating coherent radiation from intense laser-matter interaction is currently being intensely studied in order to develop efficient methods of attosecond (1 as $= 10^{-18}$ s) pulse generation and metrology [1–3]. Attosecond pulses can be produced by high harmonic generation (HHG) from atoms, molecules, and plasmas in a driving laser pulse at high intensity [4,5]. Spectral filter [6] and polarization gating [7,8] have been employed for obtaining a single attosecond pulse from a pulse train. Recently, isolated 148 as pulses have been generated with a 28 fs laser pulse with a generalized double optical gating technique [9]. The shortest single attosecond pulses with a duration of 80 [10] and 67 as [11] have been generated recently from HHG with few cycle intense femtosecond pulses.

The HHG mechanism is based on the rescattering and recollision model in intense linearly polarized light [12]. Following tunneling ionization, the electron remains "controlled" by the laser field, returning to the parent ion after a phase (sign) change of the electric field. This simple classical model of laser induced recollision with the parent ion has led to the development of a consistent theory of HHG in atoms [13] and molecules [14], giving the maximum energy law $N_m\omega_0 = I_p + 3.17U_p$, where $I_p$ is the ionization potential, $U_p = E_0^2/4\omega_0^2$ is the ponderomotive energy for a pulse maximum amplitude $E_0$, corresponding to an intensity $I_0 = \frac{1}{2}c\varepsilon_0 E_0^2$ and an angular frequency $\omega_0$ [12–14] [atomic units (a.u.) $e = \hbar = m_e = 1$ are used, unless otherwise noted]. Another scheme for HHG in molecules is collision with neighboring ions which allows for extension of the cutoff order as a function of ion separation $R$ [15]. In both linearly and circularly polarized laser induced collisions with neighboring ions, the maximum harmonic energies are given from the initial zero velocity ionization model by up to $I_p + 8U_p$ [15–17]. Collision with the parent or a neighboring ion allows for refocusing of the electron wave packet thus enhancing the efficiency [18].

All previous attosecond pulse generation was obtained with intense linearly polarized HHG. Circularly polarized attosecond pulses are potential new tools for investigations of electron dynamics in atoms, molecules, and materials. In general for atomic systems exposed to circularly polarized laser fields, no circularly polarized HHG spectra can be produced because of the conservation of angular momentum and the suppression of the recollision resulting in a dramatic drop in photon energy yield except in certain cases of double ionization [19]. Using bichromatic circularly polarized laser fields with opposite rotation polarization directions, a method was proposed for circularly polarized HHG [20]. Recently, (near) circularly polarized high harmonics and attosecond pulses have been generated using ring-current states with angular momentum $|m| = 6$ in atoms [21] or by the combination of elliptically polarized laser and strong static fields in molecular ions [22]. Previous work on atomic HHG with combinations of elliptically polarized and static fields has shown the sensitivity of the HHG processes on interference processes [23]. We have also shown that a single circularly polarized attosecond pulse can be produced by an intense elliptically polarized laser pulse in an asymmetric molecular ion due to the asymmetry of the Coulomb potential [24]. In this Letter, we propose an efficient method for producing a single circularly polarized attosecond pulse in a molecular medium by an intense elliptically polarized laser pulse in the presence of an intense terahertz field. Studies of terahertz fields for controlling HHG spectra and generating attosecond pulses have been widely investigated, but most have focused on linear polarization (see, e.g., Refs. [25,26]). We find that with a combination of a circularly polarized laser pulse and an intense terahertz field, the active electron can recombine according to the standard three-step model [12,13]. Since the polarization properties of HHG depend on the trajectory of the returning electron with the parent ion [27], we show that with proper conditions single circularly polarized attosecond

 

pulses can be efficiently produced in the presence of intense elliptically polarized laser pulses and terahertz fields of comparable intensity.

In Fig. 1 we present numerical results from the time-dependent Schrödinger equation (TDSE) for fixed nuclei, $x$-aligned $H_2^+$ at equilibrium $R_e = 2$ a.u. by a five-point difference and second-order split-operator method [22]. We find that single circularly polarized attosecond pulses can be successfully produced when the generated $x$ and $y$ field components have the same maximum amplitude and time duration, i.e., a full width at half maximum of 114 as and the phase difference $\phi = \pi/2$. Due to the different parallel and perpendicular ionization rates of the molecular ion [17,22], ultrashort intense elliptically polarized laser pulses, $E_x(t) = E_x f(t)\cos(\omega_0 t + \varphi)$ and $E_y(t) = E_y f(t)\sin(\omega_0 t + \varphi)$, with ellipticity $\epsilon = E_x/E_y = 0.59$ have been employed, where the pulse wavelength $\lambda = 400$ nm ($\omega_0 = 0.114$ a.u.), the phase $\varphi = 0.1\pi$, and the intensity $I_0 = 5 \times 10^{14}$ W/cm$^2$ corresponding to a field amplitude of $E_0 = 0.1194$ a.u. are used. A temporal slowly varying envelope $f(t) = \sin^2(\pi t/6\tau)$ where one optical cycle $1\tau = 2\pi/\omega_0 = 1.33$ fs is adopted. We choose the terahertz field with linear polarization along the $y$ axis and the form $E_{\mathrm{THz}}(t) = E_{\mathrm{THz}0}\sin^2(\pi t/6\tau) = E_{\mathrm{THz}0}\sin^2(\omega_0 t/12)$, where $E_{\mathrm{THz}0} = 0.75E_0 = 0.09$ a.u. ($4.63 \times 10^8$ V/cm) is the strength of the terahertz field, for $t \le 6\tau$, corresponding to a frequency of $\omega_{\mathrm{THz}} = \omega_0/12$ ($\lambda = 4800$ nm), as shown in Fig. 1(c).

The attosecond pulse field amplitudes $\mathcal{I}_{x/y}(t)$ [see Fig. 1(a)] are obtained by performing an inverse Fourier transform of the corresponding HHG spectrum, by filtering away the low energy harmonics $\omega < \omega_c = 32\omega_0$. The HHG spectrum intensity $P(\omega)$ [see Fig. 2] is obtained from the square of the Fourier transform $a(\omega)$ of the electron dipole acceleration $a(t) = \langle\Psi(t)| - \partial H/\partial r|\Psi(t)\rangle$,

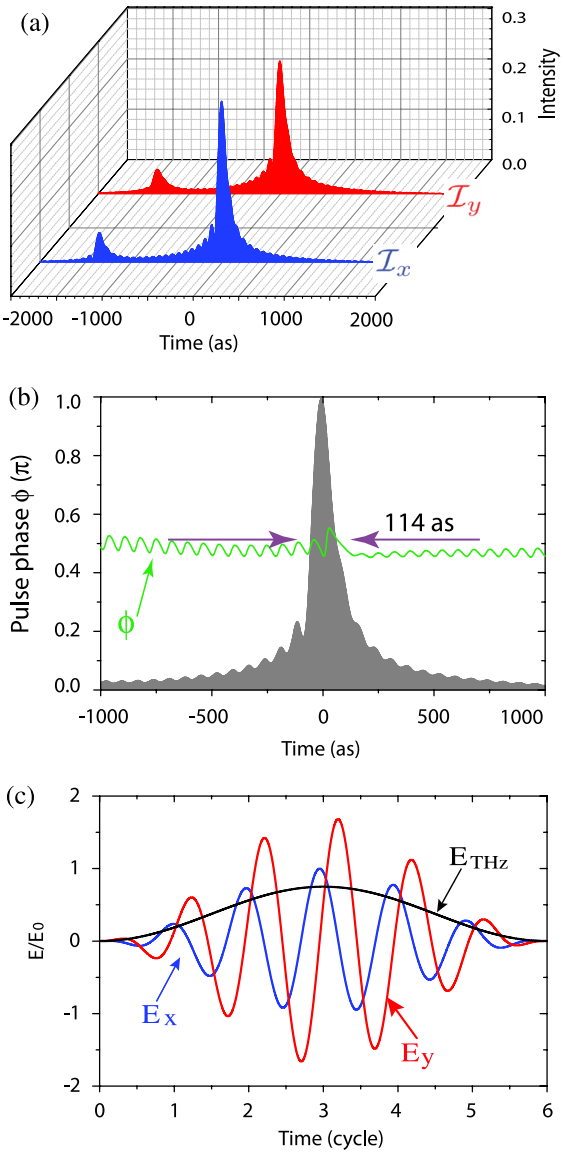


FIG. 1 (color online). (a) Temporal profiles of attosecond pulses obtained by a 6 cycle (8 fs) elliptically polarized $\epsilon = 0.59$ laser pulse at wavelength $\lambda = 400$ nm in the presence of a 62.5 THz ($\lambda = 4800$ nm) field and (b) the corresponding relative phase difference $\phi(t)$ (green line) between the $x$ and $y$ field components of attosecond pulses with intensities $\mathcal{I}_x$ and $\mathcal{I}_y$. (c) The elliptically polarized $E_x(t)$ and $E_y(t)$, and terahertz $E_{\mathrm{THz}}(t)$ laser fields for the circularly polarized HHG and isolated attosecond pulse generation ($I_0 = 5 \times 10^{14}$ W/cm$^2$).

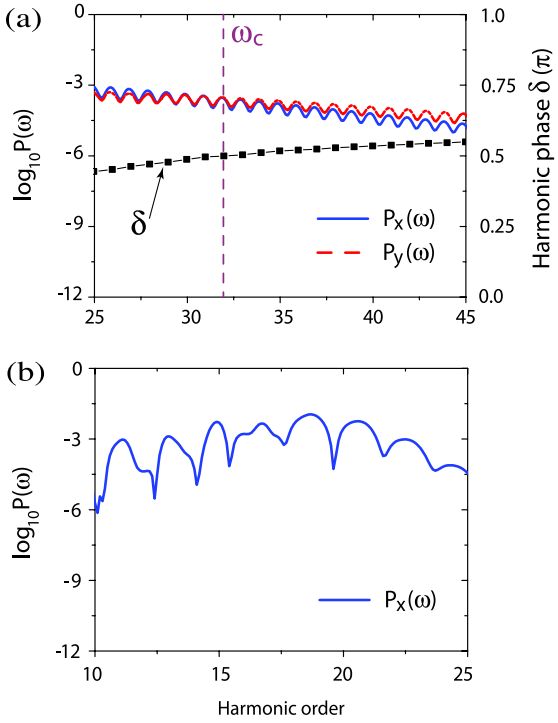


FIG. 2 (color online). (a) Harmonic intensities $P_x(\omega)$ and $P_y(\omega)$ for generations of attosecond pulses and corresponding phase difference $\delta(\omega)$ (black square) in the combination fields, corresponding to Fig. 1(c). (b) Molecular harmonics in an intense 6 cycle linearly polarized laser pulse at intensity $I_0 = 5 \times 10^{14}$ W/cm$^2$ and wavelength $\lambda = 400$ nm.

where the electron wave function $\Psi(t)$ is an exact solution of the TDSE and $r = x, y$. To describe the polarization properties of the emitted HHG spectra and generated attosecond pulses, we have also calculated the relative harmonic and attosecond pulse phase differences, $\delta$ and $\phi$. The complex integrals of $a(\omega)$ and $\mathcal{I}(t)$ have two $x$ and $y$ components, thus allowing us to extract the phase differences $\delta$ between the polarized components of the emitted harmonics, $\delta(\omega) = |\arg[a_x(\omega)] - \arg[a_y(\omega)]|$, and $\phi$ of the attosecond pulses $\phi(t) = |\arg[\mathcal{I}_x(t)] - \arg[\mathcal{I}_y(t)]|$.

Figure 2(a) shows molecular harmonics and their relative phases by combinations of the 6 cycle (8 fs) elliptically polarized $\epsilon = 0.59$ laser pulse and the 62.5 THz fields, where $P_x(\omega)$ and $P_y(\omega)$ are equivalent in intensity around $\omega_c$ with the phase $\delta(\omega) = \pi/2$. From such circularly polarized HHG spectra, circularly polarized attosecond pulses are therefore produced. However we find that in the plateau regime HHG spectra created with the assistance of the terahertz field are about 12 orders ($10^{12}$) of magnitude higher than those produced by the second collision process ($\sim 10^{-16}$) [24]. For comparison of HHG efficiency, in Fig. 2(b) we also display molecular harmonics near the cutoff regime by a few cycle linearly polarized laser pulse at an intensity of $I_0 = 5 \times 10^{14}$ W/cm$^2$. Results show that the efficiency of the circular polarization harmonics is nearly similar to that of the linear polarization with magnitude orders $\sim 10^{-3}$ since essentially the circularly polarized attosecond pulse is a superposition of two linear polarization laser induced electron recollision processes. Thus with such an efficiency of HHG it is possible to generate single circularly polarized attosecond pulses with current ultrashort laser technology (see, e.g., Refs. [4,5,9–11]).

To study the HHG mechanism of the present laser pulse and terahertz field scheme, in Fig. 3 we show time profiles of the $x$ and $y$ components of the generated circularly polarized harmonics near the cutoff region from which single circularly polarized attosecond pulses are produced. The time profile analysis [28] of the harmonics provides the recollision time of the continuum electron with parent ions as it is guided by the time dependent field and informs us about the depopulation of the state to which the electron recollides in the presence of the laser field. The time profiles of the harmonics are obtained via a Gabor transform [28] of the time dependent dipole acceleration which includes phase effects,

$$a_G(\omega, t) = \int_{-\infty}^{\infty} \exp(-i\omega t') \exp\left[-\frac{(t'-t)^2}{2\sigma_0^2}\right] a(t') dt', \quad (1)$$

where $\sigma_0 = 0.1$ fs is the width of the Gaussian time window in the Gabor transform. HHG mainly comes from single electron recollision trajectories, leading to isolated attosecond pulses [7]. Figure 3 shows that the electron recollision trajectory at time $t = 2.6\tau$ is dominant due to molecular photoionization at early times. Thus single recollision is the main mechanism of circularly polarized attosecond pulse generation as opposed to linear polarization.

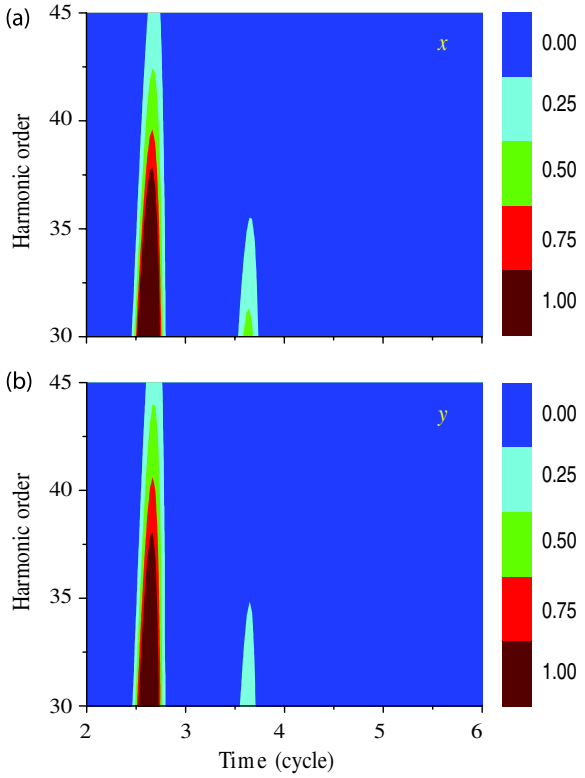


FIG. 3 (color online). Time profiles of the (a) $x$ and (b) $y$ components of the acceleration amplitude $a_G(\omega, t)$ using a Gabor time window in the cutoff region in the combined elliptically polarized laser and terahertz fields, corresponding to Fig. 1(c).

We have compared our numerical results with a classical model of recollision in the combined elliptically polarized laser and terahertz fields. Due to the slow variation of the terahertz field with time, we examine its influence at its peak strength, $E_{\mathrm{THz}} = s_0 E_0$. The classical field equations of motion, $\ddot{x}(t) = -E_0 \cos(\omega_0 t + \varphi)$ and $\ddot{y}(t) = -\epsilon E_0 \sin(\omega_0 t + \varphi) - s_0 E_0$, give the laser induced velocities

$$\dot{x}(t) = -\frac{E_0}{\omega_0}[\sin(\omega_0 t + \varphi) - \sin\varphi],$$
$$\dot{y}(t) = -\frac{\epsilon E_0}{\omega_0}[\cos\varphi - \cos(\omega_0 t + \varphi)] - s_0 E_0 t, \quad (2)$$

with the initial zero velocity conditions, $\dot{x}(0) = \dot{y}(0) = 0$ as the first step in tunneling ionization. The corresponding electron time-dependent displacements are

$$x(t) = -\frac{E_0}{\omega_0^2}[\cos\varphi - \cos(\omega_0 t + \varphi) - \omega_0 t \sin\varphi],$$
$$y(t) = -\frac{\epsilon E_0}{\omega_0^2}[\omega_0 t \cos\varphi + \sin\varphi - \sin(\omega_0 t + \varphi)] - \frac{1}{2} s_0 E_0 t^2. \quad (3)$$

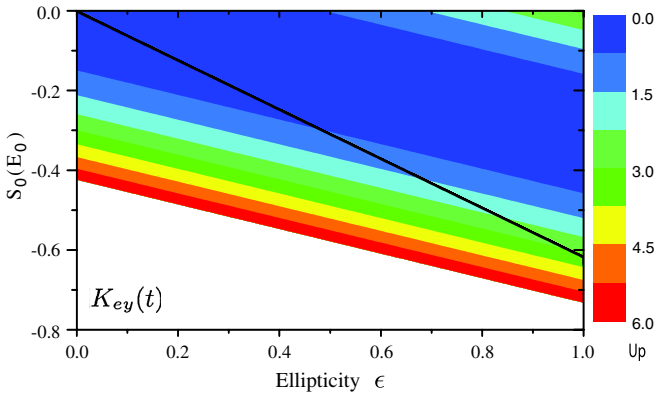


FIG. 4 (color online). Transverse kinetic energy $K_{ey}(t)$ as functions of ellipticity $\epsilon$ and terahertz field strength $s_0$ for the continuum electron with $\varphi = 0.1\pi$ and $t = 1.3\pi/\omega_0$ ($n = 0$). The black solid line denotes the kinetic energy $K_{ey}$ of the recollision electron with the parent ion, i.e., $x(t) = y(t) = 0$.

In the $x$ direction, since $E_x(t) = E_0 \cos(\omega_0 t + \varphi)$, the recollision conditions $\varphi = 2n\pi + 0.1\pi$ and $\omega_0 t = 2n\pi + 1.3\pi$, $n = 0, 1, 2, \ldots$, result in the maximum recollision kinetic energy $K_{ex} = \dot{x}^2/2 = 3.17U_p$ of the continuum electron at $x(t) = 0$ [14]. Substituting $\varphi$ and $\omega_0 t$ into Eqs. (2) and (3), we obtain the transverse velocity $\dot{y}(\epsilon, s_0)$ and position $y(\epsilon, s_0)$ at $t = (2n\pi + 1.3\pi)/\omega_0$. The corresponding kinetic energy $K_{ey} = \dot{y}^2/2$ as functions of ellipticity $\epsilon$ and $s_0$ with the pulse phase $\varphi = 0.1\pi$ and the recollision time $\omega_0 t = 1.3\pi$ ($n = 0$) is displayed Fig. 4. The transverse kinetic energy $K_{ey}$ is shown to be critically sensitive to the field parameters $\epsilon$ and $s_0$. To efficiently produce harmonics, the displacements must satisfy the conditions $x(t) = y(t) = 0$ in Eq. (3), for recollision of continuum electrons with the parent ion. In Fig. 4 we also plot the corresponding kinetic energy $K_{ey}$ (black solid line), where the pulse parameters are defined by the recollision conditions, i.e., $x(t) = y(t) = 0$ for $\varphi = 0.1\pi$ and $\omega_0 t = 1.3\pi$ ($n = 0$). The maximum transverse kinetic energy in the $y$ direction occurs at $\epsilon = 1$ and $s_0 = -0.617$ and $K_{ey} = 3.17U_p = K_{ex}$, or the total maximum kinetic energy $K_e = 6.34U_p$. For $\epsilon = 0$ and $s_0 = 0$, $K_{ey} = 0$, i.e., for a linearly $x$-polarized laser pulse, we recover the maximum linear polarization kinetic energy $K_e = K_{ex} = 3.17U_p$ [12–14]. It should be noted that in the classical recollision model, the effects of the Coulomb potential and the molecular structure, leading for example to orientation dependent ionization rates in the molecular photoionization processes [17,22], are ignored. In the numerical simulations such effects are included, showing that an elliptically polarized laser pulse and strong terahertz fields generate circularly polarized harmonics due to the control of the recolliding electron by the driving laser pulses, i.e., the electron recollision with the parent ions. Figure 3 illustrates that the circularly polarized harmonics near the cutoff region are mainly created by single trajectories at the approximate recollision times of $t_c = 0.65\tau$, which are in good agreement with the classical model predicted by Eqs. (2) and (3) where the recollision time is $t_c = 1.3\pi/\omega_0 = 0.65\tau \approx 1$ fs for $n = 0$ at $\omega_0 = 0.114$ a.u. ($\lambda = 400$ nm). This confirms that the harmonics mainly result from single recollision and rescattering of the electron wave packets induced by the elliptically polarized laser and terahertz fields. Electron recollisions can also occur at $\omega_0 t = 2n\pi + 1.3\pi$ for $n > 0$ to produce circularly polarized harmonics. From Eq. (3), one sees that much longer trajectories are created at such time, thus reducing the efficiency of high frequency HHG. Furthermore since circularly polarized HHG is produced with equal field intensities $P_x$ and $P_y$ and the relative phase $\pi/2$, short trajectories with $n = 0$ are the most efficient.

The present work focuses on the laser control electron recollision with the parent ions in the molecular plane to produce high order circularly polarized harmonics, thus generating single circularly polarized attosecond pulses. The refocusing of these electron trajectories by the two nuclear centers plays an essential role. To produce circularly polarization HHG, equal acceleration dipole amplitudes with the relative phase $\pi/2$ and energies of the returning electron are required for the resulting emitted radiation. Therefore the recombination of the electron determines the properties of the harmonics. For an atom with a spherical symmetric electron probability density, HHG with low ellipticity can be produced by intense elliptically polarized laser pulses (see, e.g., Ref. [29]). Excitation of an atom to high angular momentum states can also result in near circularly polarized attosecond pulses in the cutoff region [21]. Recent theoretical studies of atomic HHG by circularly and elliptically intense laser pulses show that up to four recollision trajectories can occur, i.e., two trajectories for each polarization direction [30,31]. Our detailed time profiles for the molecular ion in Fig. 3 show that only a single recollision trajectory occurs, thus implying essential control of the HHG process in molecules by both the highly nonspherical asymmetric molecular Coulomb potential and the presence of the intense terahertz field to redirect circular electron trajectories with large radii towards the molecular ion, according to Eq. (3). The present results suggest molecular media, rather than atoms, as promising sources to produce high order circularly polarized harmonics for the generation of single circularly polarized attosecond pulses. Such pulses should serve to create circular electron currents with resulting large internal magnetic fields in molecules [32] for new research in the nonlinear response of matter to intense fields.

The authors thank RQCHP and Compute Canada for access to massively parallel computer clusters and the CIPI (Canadian Institute for Photonic Innovations) for financial support in its ultrafast science program.

*andre.bandrauk@usherbrooke.ca


[1] P. B. Corkum and F. Krausz, Nat. Phys. **3**, 381 (2007).
[2] F. Krausz and M. Ivanov, Rev. Mod. Phys. **81**, 163 (2009).
[3] Z. Chang and P. Corkum, J. Opt. Soc. Am. B **27**, B9 (2010).
[4] T. Popmintchev, M.-C. Chen, P. Arpin, M. M. Murnane, and H. C. Kapteyn, Nat. Photonics **4**, 822 (2010).
[5] T. Popmintchev *et al.*, Science **336**, 1287 (2012).
[6] R. Kienberger, E. Goulielmakis, M. Uiberacker, A. Baltuska, V. Yakovlev, F. Bammer, S. Scrinzi, T. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) **427**, 817 (2004).
[7] P. B. Corkum, N. H. Burnett, and M. Y. Ivanov, Opt. Lett. **19**, 1870 (1994).
[8] I. J. Sola, E. Mével, L. Elouga, E. Constant, V. Strelkov, L. Poletto, P. Villoresi, E. Benedetti, J. P. Caumes, S. Stagira, C. Vozzi, G. Sansone, and M. Nisoli, Nat. Phys. **2**, 319 (2006).
[9] X. Feng, S. Gilbertson, H. Mashiko, H. Wang, S. D. Khan, M. Chini, Y. Wu, K. Zhao, and Z. Chang, Phys. Rev. Lett. **103**, 183901 (2009).
[10] E. Goulielmakis, M. Schultze, M. Hofstetter, V. S. Yakovlev, J. Gagnon, M. Uiberacker, A. L. Aquila, E. M. Gullikson, D. A. Attwood, R. Kienberger, F. Krausz, and U. Kleineberg, Science **320**, 1614 (2008).
[11] K. Zhao, Q. Zhang, M. Chini, Y. Wu, X. Wang, and Z. Chang, Opt. Lett. **37**, 3891 (2012).
[12] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[13] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, Phys. Rev. A **49**, 2117 (1994).
[14] A. D. Bandrauk, S. Chelkowski, and S. Goudreau, J. Mod. Opt. **52**, 411 (2005).
[15] A. Bandrauk, S. Barmaki, and G. L. Kamta, Phys. Rev. Lett. **98**, 013001 (2007).
[16] M. Lein and J. M. Rost, Phys. Rev. Lett. **91**, 243901 (2003).
[17] K. J. Yuan and A. D. Bandrauk, Phys. Rev. A **81**, 063412 (2010).
[18] M. V. Frolov, N. L. Manakov, T. S. Sarantseva, M. Yu. Emelin, M. Yu. Ryabikin, and A. F. Starace, Phys. Rev. Lett. **102**, 243901 (2009).
[19] F. Mauger, C. Chandre, and T. Uzer, Phys. Rev. Lett. **105**, 083002 (2010).
[20] D. B. Milosevic, W. Becker, and R. Kopold, Phys. Rev. A **61**, 063403 (2000).
[21] X. Xie, A. Scrinzi, M. Wickenhauser, A. Baltuška, I. Barth, and M. Kitzler, Phys. Rev. Lett. **101**, 033901 (2008).
[22] K. J. Yuan and A. D. Bandrauk, Phys. Rev. A **83**, 063422 (2011); J. Phys. B **45**, 074001 (2012).
[23] B. Borca, A. V. Flegel, M. V. Frolov, N. L. Manakov, D. B. Milosevic, and A. F. Starace, Phys. Rev. Lett. **85**, 732 (2000).
[24] K. J. Yuan and A. D. Bandrauk, Phys. Rev. A **84**, 023410 (2011).
[25] D. B. Milosevic and A. F. Starace, Laser Phys. **10**, 278 (2000).
[26] E. Balogh, K. Kovacs, P. Dombi, J. A. Fulop, G. Farkas, J. Hebling, V. Tosa, and K. Varju, Phys. Rev. A **84**, 023806 (2011).
[27] F. Morales, I. Barth, V. Serbinenko, S. Patchkovskii, and O. Smirnova, J. Mod. Opt. **59**, 1303 (2012).
[28] P. Antoine, B. Piraux, and A. Maquet, Phys. Rev. A **51**, R1750 (1995).
[29] M. Möller, Y. Cheng, S. D. Khan, B. Zhao, K. Zhao, M. Chini, G. G. Paulus, and Z. Chang, Phys. Rev. A **86**, 011401(R) (2012).
[30] C. H. Raymond Ooi, W. L. Ho, and A. D. Bandrauk, Phys. Rev. A **86**, 023410 (2012).
[31] M. V. Frolov, N. L. Manakov, T. S. Sarantseva, and A. F. Starace, Phys. Rev. A **86**, 063406 (2012).
[32] I. Barth, J. Manz, Y. Shigeta, and K. Yagi, J. Am. Chem. Soc. **128**, 7043 (2006); I. Barth and J. Manz, Phys. Rev. A **75**, 012510 (2007).