PHYSICAL REVIEW A **71**, 021402(R) (2005)

Low-order above-threshold ionization in intense few-cycle laser pulses

C. C. Chirilă and R. M. Potvliege
Department of Physics, University of Durham, Durham DH1 3LE, United Kingdom
(Received 1 August 2004; published 11 February 2005)

The low-energy end of the spectrum of photoelectrons detached from hydrogenic ions exposed to an intense low-frequency few-cycle pulse is calculated within the strong-field approximation (SFA). The effect on the detached photoelectron of the Coulomb field of the nucleus is taken into account quasiclassically. The results are compared with those of an *ab initio* solution of the time-dependent Schrödinger equation, for the case of an He^+ ion irradiated by a 400-nm pulse of 1×10^{16} W cm^{-2} peak intensity. Many of the features of the *ab initio* spectra can be understood within the SFA.

DOI: 10.1103/PhysRevA.71.021402 PACS number(s): 32.80.Rm, 32.80.Wr, 42.50.Hz

Most of the electrons ejected from atoms by strong, low-frequency, linearly polarized laser pulses have a relatively low energy, somewhat below twice the ponderomotive energy. It is well established that the ejection of electrons of higher energy, belonging to the plateau part of the above-threshold ionization (ATI) spectra, can in many respects be described within the strong-field approximation (SFA) and be explained by the simple semiclassical model with which the SFA is associated [1,2]. The question thus arises as to how well can the ejection of slow electrons be understood within this approximation. Of central importance in this problem is the role of the long-range Coulomb interaction between the outgoing photoelectron and the residual core. It has been known for a long time that the accuracy with which this interaction can be taken into account within the SFA is sufficient for obtaining correct total probabilities of ionization [3–6]. The SFA has also been shown to be reliable for low-order above-threshold detachment from negative ions, for which there is no long-range Coulomb interaction in the final state [7,8]. However, there are also indications that the low-energy photoelectrons ejected from atoms or positive ions in strong fields might be so much affected by the electron-core interaction that SFA-type theories would be unable to explain their angular and energy distributions, even in the adiabatic regime [9]. In this Rapid Communication, we show that a good description of their spectra can nonetheless be achieved within this approximation for a wide range of angles of ejection, if the binding energy of the initial state is sufficiently large and the intensity sufficiently high. Our approach is based on Keldysh's length gauge formulation of the SFA. We neglect backscattering but make allowance for the effect of the Coulomb interaction on the continuum electron through a simple quasiclassical approximation [10]. The corresponding expression of the ionization amplitude is derived in the following paragraphs. Atomic units are used throughout the paper, except where specified otherwise.

We assume the dipole approximation and describe the laser pulse by a vector potential of the form $\mathbf{A}(t) = (F_0/\omega)\hat{\boldsymbol{\epsilon}}a(t)\sin(\omega t+\phi)$, with $\hat{\boldsymbol{\epsilon}}$ the polarization vector, ω the carrier's angular frequency, ϕ the carrier-envelope phase, and $a(t)$ a function varying between 0 and 1 and defining the envelope of the pulse. The latter is taken to be a $\sin^2$ function in the numerical calculation, with a total width (measured at the basis) of either 4 or 2 optical cycles. The corresponding electric-field component of the pulse is $\mathbf{F}(t)=-\partial_t\mathbf{A}(t)$. We also consider that the pulse lasts from a time t_i to a time t_f such that $\mathbf{A}(t)$ and $\mathbf{F}(t)$ are both negligible for $t\leqslant t_i$ and $t\geqslant t_f$. We aim at calculating the amplitude for the electron to be at a time $T\geqslant t_f$ in a field-free continuum state $|\Phi_{\mathbf{p}}(T)\rangle$ of asymptotic momentum $\mathbf{p}$ if for $t<t_i$ it is in the field-free bound state $|\Phi_0(t)\rangle$. This amplitude can be written in terms of the dipole operator and of the time-evolution operator for the atom in the field, as [1]

$$A_{\mathbf{p}0} = -i\int_{t_i}^{t_f} dt\langle\Phi_{\mathbf{p}}(T)|U(T,t)H_{\mathrm{dip}}(t)|\Phi_0(t)\rangle. \tag{1}$$

The matrix element $\langle\Phi_{\mathbf{p}}(T)|U(T,t)H_{\mathrm{dip}}(t)|\Phi_0(t)\rangle$ is the projection of $H_{\mathrm{dip}}(t)|\Phi_0(t)\rangle$ on the state vector which, at time T, reduces to $|\Phi_{\mathbf{p}}(T)\rangle$. Assuming the normalization $\langle\Phi_{\mathbf{p}}(T)|\Phi_{\mathbf{p}'}(T)\rangle=\delta(\mathbf{p}-\mathbf{p}')$, the density of probability that an electron is detached by the pulse with a final kinetic energy $E=p^2/2$ in the direction of the vector $\mathbf{p}$ is $P(E,\hat{\mathbf{p}})=p|A_{\mathbf{p}0}|^2$.

In Keldysh's formulation of the SFA, the interaction of the electron with the field is described within the length gauge, $\langle\mathbf{r}|\Phi_{\mathbf{p}}(T)\rangle$ is taken to be a plane wave of momentum $\mathbf{p}$, and the interaction of the unbound electron with the residual core is neglected between times t and T. Accordingly, $\langle\Phi_{\mathbf{p}}(T)|U(T,t)|\mathbf{r}\rangle$ is taken to be the complex conjugate of the Volkov wave function

$$\Psi_{\mathbf{p}}(\mathbf{r},t) = \frac{1}{(2\pi)^{3/2}}\exp\left[i\boldsymbol{\pi}(\mathbf{p},t)\cdot\mathbf{r} + \frac{i}{2}\int_t^T dt'\,\boldsymbol{\pi}^2(\mathbf{p},t')\right], \tag{2}$$

where $\boldsymbol{\pi}(\mathbf{p},t)=\mathbf{p}+\mathbf{A}(t)$ is the kinematical momentum of the electron. Within this approach, the interaction of the electron with the core is taken into account only in the initial wave function, $\Phi_0(\mathbf{r},t)\equiv\langle\mathbf{r}|\Phi_0(t)\rangle$. As noted by Krainov and Shokri [5], the effect of this interaction during tunneling can also be taken into account, approximately, by multiplying $\Psi_{\mathbf{p}}(\mathbf{r},t)$ by the factor $I(r)=[4I_p/(F_0 r)]^{Z/\sqrt{2I_p}}$, with Z the charge of the residual ion and I_p the binding energy of the initial state. Adopting this correction yields the "tunneling corrected" ionization amplitude

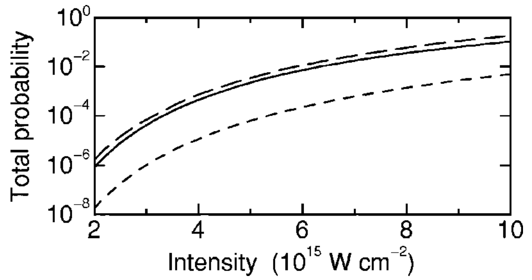

FIG. 1. The total detachment probability for an He^+ ion irradiated by a 4-cycle 800-nm pulse of zero carrier-envelope phase, as calculated within the SFA with (long-dashed curve) or without (short-dashed curve) the factor $I(r)$, vs the peak intensity of the pulse. Also shown is the prediction of Eq. (8) (solid curve).

$$A_{\mathbf{p}0}^{(\mathrm{TC})} = -i\int_{t_i}^{t_f} dt \int d\mathbf{r}\ \Psi_{\mathbf{p}}^*(\mathbf{r},t) I(r)[\mathbf{r}\cdot\mathbf{F}(t)]\Phi_0(\mathbf{r},t). \quad (3)$$

In this work, we concentrate on the case of a hydrogenic ion initially in the ground state, for which $\Phi_0(\mathbf{r},t) = B\exp(-\kappa r)/\sqrt{4\pi}\exp(I_p t)$, with $\kappa=(2I_p)^{1/2}$ and $B=2\kappa^{3/2}$. Integration over time using the saddle-point method gives [11]

$$A_{\mathbf{p}0}^{(\mathrm{TC})} = \frac{B}{i\pi\sqrt{8}}\left(\frac{2\kappa^2}{F_0}\right)\sum_{t_s}\sqrt{\frac{2\pi i}{S''(\mathbf{p},t_s)}}\exp[iS(\mathbf{p},t_s)]. \quad (4)$$

In this equation,

$$S(\mathbf{p},t) = -\frac{1}{2}\int_t^T dt'\,\boldsymbol{\pi}^2(\mathbf{p},t') + I_p t \quad (5)$$

and the (complex) times t_s are the saddle points of $S(\mathbf{p},t)$, i.e., the times at which $\boldsymbol{\pi}^2(\mathbf{p},t)/2+I_p=0$.

Had we omitted the factor $I(r)$ in Eq. (3), the ionization amplitude would have reduced to a short-pulse form of the familiar Keldysh amplitude, namely

$$A_{\mathbf{p}0}^{(\mathrm{K})} = -i\int_{t_i}^{t_f} dt \int d\mathbf{r}\ \Psi_{\mathbf{p}}^*(\mathbf{r},t)[\mathbf{r}\cdot\mathbf{F}(t)]\Phi_0(\mathbf{r},t). \quad (6)$$

This integral is also amenable to saddle-point integration, although the integrand is singular at the saddle times for our initial wave function [8]. The result is

$$A_{\mathbf{p}0}^{(\mathrm{K})} = \frac{\kappa B}{\sqrt{8}}\sum_{t_s}\frac{1}{S''(\mathbf{p},t_s)}\exp[iS(\mathbf{p},t_s)]. \quad (7)$$

The effect of the correction factor $I(r)$ is illustrated by Fig. 1, in which the total detachment probability, $P_{\mathrm{tot}} = \int dE\, d\hat{\mathbf{p}} P(E,\hat{\mathbf{p}})$, calculated using Eq. (4), is compared to that calculated using Eq. (7). Results are presented for an He^+ ion irradiated by a 4-cycle 800-nm pulse. Also shown in Fig. 1 is the probability obtained under the assumption that detachment proceeds at any time as if the electric field was static. Within that model,

$$P_{\mathrm{stat}} = 1 - \exp\left\{-\int_{t_i}^{t_f} dt\ \Gamma[|\mathbf{F}(t)|]\right\}, \quad (8)$$

where $\Gamma[F]$ is the rate of ionization by a static electric field of strength F. The Keldysh adiabaticity parameter $\gamma = \omega(2I_p)^{1/2}/F_0$ ranges from 0.48 at 2×10^{15} W cm^{-2} intensity down to 0.21 at 1×10^{16} W cm^{-2}. Given that it is small, one can expect P_{stat} to be a good approximation of the exact detachment probability. As seen from the figure, at the intensities considered, the SFA probability without the correction factor is somewhat smaller than P_{stat}. (Depletion, which is not taken into account in our estimate of P_{tot}, is not negligible at intensities above 1×10^{16} W cm^{-2}.) The correction factor brings the SFA probability much closer to P_{stat}.

As is well known, at high intensity the complex saddle times t_s that contribute most to the ionization amplitude (4) have a small imaginary part and differ little from the real detachment times t_d of the semiclassical model, which are the times at which $\boldsymbol{\pi}(\mathbf{p},t)\cdot\hat{\boldsymbol{\epsilon}}=0$. Neglecting terms of order $(t_s-t_d)^4$ in $S(\mathbf{p},t_s)$ yields [12]

$$A_{\mathbf{p}0}^{(\mathrm{TC})} \approx -i\sum_{t_d} a(p_\perp,t_d)\exp[iS(\mathbf{p},t_d)], \quad (9)$$

where $p_\perp$ is the component of $\mathbf{p}$ normal to the direction of polarization and

$$a(p_\perp,t_d) = \frac{B}{\pi\sqrt{8}}\left(\frac{2\kappa^2}{F_0}\right)\sqrt{\frac{2\pi}{\zeta|\mathbf{F}(t_d)|}}\exp\left[-\frac{1}{3}\frac{\zeta^3}{|\mathbf{F}(t_d)|}\right], \quad (10)$$

with $\zeta=(2I_p+p_\perp^2)^{1/2}$. Equation (9) can also be written as

$$A_{\mathbf{p}0}^{(\mathrm{TC})} \approx \sum_{t_d}\langle\Phi_{\mathbf{p}}(T)|U(T,t_d)|\Psi_{\mathbf{p}0}(t_d)\rangle, \quad (11)$$

with $\langle\Phi_{\mathbf{p}}(T)|U(T,t_d)|\mathbf{r}\rangle$ approximated by $\Psi_{\mathbf{p}}^*(\mathbf{r},t_d)$ and

$$\langle\mathbf{r}|\Psi_{\mathbf{p}0}(t_d)\rangle = -i(2\pi)^{3/2}a(p_\perp,t_d)e^{iI_p t_d}\delta(\mathbf{r}-\mathbf{r}_d(t_d)). \quad (12)$$

Equations (9) and (11) are equivalent if the vectors $\mathbf{r}_d(t_d)$, which are otherwise arbitrary, fulfill the condition $\boldsymbol{\pi}(\mathbf{p},t_d)\cdot\mathbf{r}_d(t_d)=0$. By analogy with the semiclassical model, we take, for each detachment time t_d, the vector $\mathbf{r}_d(t_d)$ to be the position vector of the outer turning point of the potential barrier, $-I_p\mathbf{F}(t_d)/|\mathbf{F}(t_d)|^2$. The state vectors $|\Psi_{\mathbf{p}0}(t_d)\rangle$ can thus be viewed as representing the nascent photoelectron at the possible detachment times.

In order to take into account the interaction of the unbound electron with the core, we replace, in Eq. (11) , $\langle\Phi_{\mathbf{p}}(T)|U(T,t)|\mathbf{r}\rangle$ by the complex conjugate of a quasiclassical Coulomb-corrected Volkov wave recently discussed by Gordienko and Meyer-ter-Vehn [13], $C_{\mathbf{p}}(\mathbf{r},t) \equiv \Psi_{\mathbf{p}}(\mathbf{r},t)\exp[i\sigma(\mathbf{r},t)]$. Here,

$$\sigma(\mathbf{r},t) = -Z\int_t^T dt'\left|\mathbf{r} + \int_t^{t'} dt''\boldsymbol{\pi}(\mathbf{p},t'')\right|^{-1}. \quad (13)$$

As shown by Gordienko and Meyer-ter-Vehn, this wave function can be expected to be accurate if $|\boldsymbol{\pi}(\mathbf{p},t)| \gg |\nabla\sigma(\mathbf{r},t)|$, i.e., if the electron's velocity resulting from its

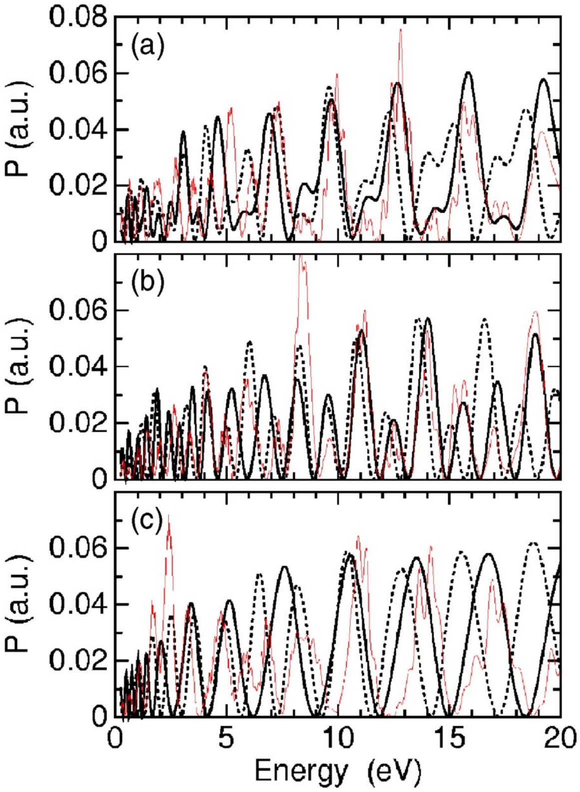

FIG. 2. The density of probability $P(E,\hat{\mathbf{p}})$, in atomic units, that an electron is ejected from an He^+ ion irradiated by a 4-cycle laser pulse of 400-nm wavelength and 1×10^{16} W cm^{-2} intensity. Thin solid curves: *ab initio* spectra. Dotted curves: predictions of the tunneling corrected SFA. Thick solid curves: predictions of the continuum and tunneling corrected SFA. (a): $\theta=10°$, $\phi=0$; (b): $\theta=10°$, $\phi=\pi/2$; (c): $\theta=170°$, $\phi=\pi/2$.

acceleration by the laser field is much larger than its additional velocity resulting from its interaction with the core. The replacement results in a "continuum and tunneling corrected" (CTC) ionization amplitude [14],

$$A_{\mathbf{p}0}^{(\mathrm{CTC})} = -i\sum_{t_d} a(p_\perp,t_d)e^{i[S(\mathbf{p},t_d)+\delta S(\mathbf{p},t_d)]}, \tag{14}$$

with $\delta S(\mathbf{p},t_d)\equiv -\sigma(\mathbf{r}_d(t_d),t_d)$. Thus $\delta S(\mathbf{p},t_d)$ is the contribution of the Coulomb potential to the classical action calculated over the trajectory of an electron accelerated only by the laser field and which, at time t_d, is at position $\mathbf{r}_d(t_d)$ and has a momentum $\boldsymbol{\pi}(\mathbf{p},t_d)$. Clearly, Eqs. (13) and (14) lose their meaning if the electron approaches the Coulomb singularity too closely.

Illustrative energy spectra are presented in Figs. 2 and 3 for the case of an He^+ ion irradiated by a few-cycle pulse with a carrier wavelength of 400 nm and a peak intensity of 1×10^{16} W cm^{-2}. The corresponding value of the Keldysh parameter γ is 0.42 and the ponderomotive energy is 149 eV. The figures give the density of probability $P(E,\hat{\mathbf{p}})$ as obtained using either Eq. (9) or Eq. (14) , for different values of

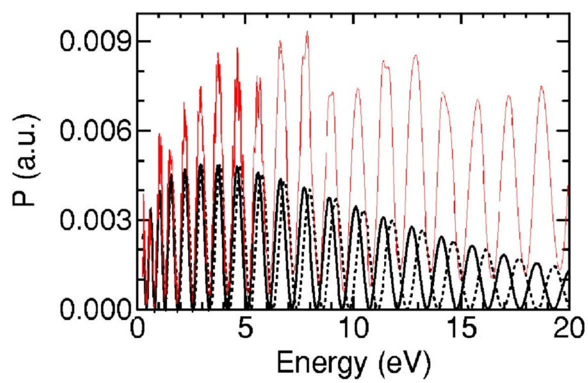

FIG. 3. The same as Fig. 2(b), but for a laser pulse encompassing only 2 optical cycles.

the angle of ejection θ and of the carrier-envelope phase ϕ. (The angle θ is measured with respect to the polarization vector $\hat{\boldsymbol{\epsilon}}$.) The SFA results are compared with the momentum distribution of the photoelectron at the end of the pulse calculated *ab initio* by solving the time-dependent Schrödinger equation numerically [15]. Correspondingly, we set $T=t_f$ in the SFA calculations.

In the cases considered in Fig. 2, the SFA results are, on the whole, in quantitative agreement with the *ab initio* results, although the position and the shape of the peaks differ at very low energy. Note that these peaks are not ATI peaks in the usual sense, which would be found for longer laser pulses. When correctly predicted by the SFA, they can be ascribed to quantum inferferences between photoelectrons with a same final kinetic energy but different trajectories [8]. They are not regularly spaced and their position varies with the angle of ejection. In the CTC-SFA, the Coulomb force acting on the continuum electron merely changes the phase relationship of the different trajectories. The change can be considerable, as some of the trajectories (corresponding to the "indirect wave packets" of Ref. [9]) remain in the vicinity of the nucleus for much longer than others. It affects the spectrum mainly by shifting and deforming the peaks. The net effect, in Fig. 2, is to bring the CTC-SFA results close to the *ab initio* results above 10 eV [16]. In the case of Fig. 3, the position of the peaks is accurately predicted by the SFA calculations over the entire energy range considered, particularly when the Coulomb correction in the continuum is included, but their relative magnitude is not as well reproduced as in Fig. 2.

For ejection between 10° and 170° from the direction of polarization, the level of agreement between the CTC-SFA results and the *ab initio* results is comparable to that shown by Figs. 2 and 3, if not better. The agreement deteriorates below 10° and above 170°. This is not surprising, since electrons ejected close to the polarization direction may pass close to the Coulomb singularity.

The authors thank B. Piraux for useful discussions and providing the programs used for obtaining the *ab initio* results, and EPSRC for their financial support.

[1] W. Becker *et al.*, Adv. At., Mol., Opt. Phys. **48**, 35 (2002).

[2] Recall that in this model, the photoelectron enters the continuum at some detachment time t_d, e.g., by tunneling through the effective potential barrier formed by the instantaneous electric field of the laser. Once in the continuum, the electron follows a classical trajectory under the acceleration imparted by the field.

[3] A. M. Perelomov and V. S. Popov, Zh. Eksp. Teor. Fiz. **52**, 514 (1967) [Sov. Phys. JETP **25**, 336 (1967)]; A. M. Perelomov, V. S. Popov, and V. P. Kuznetsov, Zh. Eksp. Teor. Fiz. **54**, 841 (1968) [Sov. Phys. JETP **27**, 451 (1968)].

[4] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)]; F. A. Ilkov, J. E. Decker, and S. L. Chin, J. Phys. B **25**, 4005 (1992).

[5] V. P. Krainov and B. Shokri, Zh. Eksp. Teor. Fiz. **107**, 1180 (1995) [JETP **80**, 657 (1995)]; V. P. Krainov, J. Opt. Soc. Am. B **14**, 425 (1997). See also A. Becker and F. H. M. Faisal, Phys. Rev. A **59**, R1742 (1999).

[6] A. Becker *et al.*, Phys. Rev. A **64**, 023408 (2001); H. R. Reiss, *ibid.* **65**, 055405 (2002).

[7] R. Shakeshaft *et al.*, Phys. Rev. A **42**, 1656 (1990).

[8] G. F. Gribakin and M. Yu. Kuchiev, Phys. Rev. A **55**, 3760 (1997).

[9] A. de Bohan *et al.*, Phys. Rev. Lett. **89**, 113002 (2002).

[10] Various methods have been proposed for taking the Coulomb interaction into account within the SFA. See, e.g., Refs. [3–6,12,13]; S. V. Fomichev and D. F. Zaretsky, Phys. Lett. A **144**, 91 (1990); H. R. Reiss and V. P. Krainov, Phys. Rev. A **50**, R910 (1994); M. H. Mittleman, *ibid.* **50**, 3249 (1994), and references therein; T. Brabec, M. Yu. Ivanov, and P. B. Corkum, *ibid.* **54**, R2551 (1996); D. B. Milošević and F. Ehlotzky, J. Phys. B **32**, 1585 (1999), and references therein; H. W. van der Hart and K. Burnett, Phys. Rev. A **62**, 013407 (2000); J. Bauer, J. Phys. B **34**, 1343 (2001); G. L. Yudin and M. Yu. Ivanov, Phys. Rev. A **63**, 033404 (2001); V. T. Platonenko, Kvantovaya Elektron. (Moscow) **31**, 55 (2001) [Quantum Electron. **31**, 55 (2001)]; K. Mishima *et al.*, Phys. Rev. A **66**, 053408 (2002); B. A. Zon and A. S. Kornev, Zh. Eksp. Teor. Fiz. **123**, 991 (2003) [JETP **96**, 870 (2003)]; P. A. Macri, J. E. Miraglia, and M. S. Gravielle, J. Opt. Soc. Am. B **20**, 1801 (2003); V. D. Rodriguez, E. Cormier, and R. Gayet, Phys. Rev. A **69**, 053402 (2004), and references therein; S. P. Goreslavski *et al.*, Phys. Rev. Lett. **93**, 233002 (2004); C. C. Chirilă *et al.*, *ibid.* **93**, 243603 (2004). See Ref. [1] for calculations of recollisional ATI based on the Born approximation.

[11] It is convenient to integrate by parts over time, so as to express the integral in terms of the Fourier transform of $I(r)\Phi_0(\mathbf{r},t)$ with respect to the momentum $\boldsymbol{\pi}$. Contributions from the end points t_i and t_f arise both from the integration by parts and from the asymptotic expansion involved in the saddle-point integration. However, it can be shown that their sum is negligibly small if $\mathbf{F}(t)$ vanishes sufficiently smoothly at t_i and t_f. In this case, the ionization probability does not depend on the detail of the variation of the vector potential at the very beginning and the very end of the pulse, in agreement with one's intuition.

[12] M. Y. Ivanov, T. Brabec, and N. Burnett, Phys. Rev. A **54**, 742 (1996).

[13] S. Gordienko and J. Meyer-ter-Vehn, in *Laser Interaction with Matter*, Vol. 5228 of *Proceedings of SPIE*, edited by O. N. Krokhin, S. Yu. Gus'kov, and Yu. A. Merkul'ev (SPIE, Bellingham, WA, 2003), p. 416.

[14] The phase correction δS has been derived in other ways and used in the framework of the SFA in Refs. [3] and [12]. For use in the numerical integration of the Schrödinger equation, see A. M. Ermolaev *et al.*, Phys. Rev. A **60**, 4831 (1999); A. M. Ermolaev and A. V. Selin, *ibid.* **62**, 015401 (2000). For use in the path integral formulation of the transition amplitude, see, e.g., G. van de Sand and J. M. Rost, Phys. Rev. A **62**, 053403 (2000); M. Spanner, Phys. Rev. Lett. **90**, 233005 (2003); M. W. Walser and T. Brabec, J. Phys. B **36**, 3025 (2003).

[15] The *ab initio* calculations follow the method described by E. Huens *et al.*, Phys. Rev. A **55**, 2132 (1997).

[16] We could not verify whether the agreement persists at energies higher than 20 eV due to difficulties in obtaining reliable *ab initio* results.