Endpoint contribution to the instantaneous ionization rate for tunneling ionization

I. A. Ivanov,[1,2,*] Chang Hee Nam,[1,3] and Kyung Taec Kim[1,3]
[1]*Center for Relativistic Laser Science, Institute for Basic Science, Gwangju 500-712, Republic of Korea*
[2]*Research School of Physics and Engineering, The Australian National University, Canberra ACT 0200, Australia*
[3]*Advanced Photonics Research Institute, GIST, Gwangju 500-712, Republic of Korea*

(Received 8 April 2015; published 8 June 2015)

We examine the instantaneous ionization amplitudes and instantaneous ionization rates for the process of tunneling ionization. We show that the endpoint contribution usually neglected in the asymptotic evaluation of the amplitudes, may be significant. For weak fields the instantaneous ionization rate is largely defined by this contribution. For higher field strengths of the order of 0.1 a.u., the account of this contribution allows one to reproduce numerically computed instantaneous ionization rates with higher accuracy.

DOI: 10.1103/PhysRevA.91.063404 PACS number(s): 32.80.Rm, 32.80.Fb, 42.50.Hz

I. INTRODUCTION

The pioneering work by Keldysh [1], and its subsequent developments [2,3], known collectively as the Keldysh-Faisal-Reiss (KFR) theory, laid out a framework on which our current understanding of the ionization phenomena is based. Depending on the value of the Keldysh parameter $\gamma = \omega\sqrt{2|\varepsilon_0|}/E$, where ω, E, and $|\varepsilon_0|$ are the frequency, field strength, and ionization potential of the target atom (here and below we use the atomic units), ionization processes can be broadly divided in two classes: multiphoton and tunneling processes. We shall be interested below in the process of the tunneling ionization. Part of the appeal of the tunneling ionization theory to a theorist is probably due to the fact that under typical experimental conditions the motion of an ionized electron in the laser field is essentially classical. This allows one to use classical and semiclassical methods, and often permits obtaining the solution in a closed analytical form. This feature is common both for the KFR theory, its generalizations [4–7], and alternative developments, such as the recently proposed adiabatic approach [8]. On the other hand, this essentially classical character of the motion of the ionized electron allows a physically transparent interpretation of experimental data. Thus, the well-known techniques of attosecond streaking and angular attosecond streaking [9,10] are based on the analysis of the classical electron trajectories.

An important ingredient in such an analysis is the so-called instantaneous ionization rate. Although some doubts have been expressed as to the very possibility of a meaningful definition of the subcycle ionization rate [11], this notion proved to be extremely fruitful. It is a key ingredient, for example, in the derivation of the well-known and widely used ADK [7] formula. The instantaneous ionization rate is often used in simulations of the experimental results on tunneling ionization [12,13], which rely on the fact of the essentially classical character of the electron motion after the ionization event. In high harmonic generation, for example, the quantum path interference of harmonic radiation generated through different classical electron trajectories is clearly observed in the experiments, which are well reproduced in the simulation [14]. The instantaneous ionization rate provides the quantum ingredient to this picture; different electron trajectories come with the weights given by the instantaneous ionization rate [12].

The simplest approximation to the instantaneous ionization rate is the so-called quasistatic formula [5], which is just an ionization rate in the constant electric field. More general expressions with the correct pre-exponential factor, valid for all values of the Keldysh parameter γ, have been obtained in [15,16]. Expressions for the instantaneous ionization rate obtained in these works allow one to gain insight into the development of the ionization process on a subcycle scale. Derivation of these expressions rely on a technique often employed in the theories of tunneling ionization—the saddle point method [17]. It is this technique, which allows one to obtain in many cases closed analytical results in theories of tunneling ionization. It is often implicitly assumed that the saddle points contributions are the only contributions we have to consider to obtain estimates for the integrals defining the ionization amplitudes. While this is true if we consider ionization on the interval of the whole pulse duration, the situation may be different when we consider amplitudes as functions of time for the moments of time inside the laser pulse. Estimating the integrals defining the ionization amplitudes, we have to consider in this case the contribution of the endpoint—the current moment of time t. This contribution is often neglected, as it was done, e.g., in [15], and indeed it is, as we shall see, insignificant in many cases. Nevertheless, as we shall show, there is a range of the laser field parameters (weak laser fields), when this contribution is, in fact, dominant. Even for stronger fields (intensities of the order of several units of 10^{14} W/cm^2) an account of the endpoint contribution may still prove significant.

II. THEORY

We consider a system described by the Hamiltonian operator:

$$\hat{H}(t) = \hat{H}_{\text{atom}} + \hat{H}_{\text{int}}(t), \tag{1}$$

where $\hat{H}_{\text{atom}}$ is the Hamiltonian of the field-free atom, and operator $\hat{H}_{\text{int}}(t)$ describes the interaction of the atom with an electromagnetic EM field. We use the length gauge for this operator and assume that the EM field is polarized along the z

*igor.ivanov@anu.edu.au

direction:

$$\hat{H}_{\text{int}}(t) = E(t)z, \tag{2}$$

where $\boldsymbol{E}(t)$ is the electric field of the laser pulse. We assume that the laser pulse is present on the interval of time $(0,T_1)$. We are interested in the time dependence of the total ionization rate on the interval $t \in (0,T_1)$. The total ionization rate may be defined as

$$w(t) = \frac{dP(t)}{dt}, \tag{3}$$

where

$$P(t) = \int |a_{\boldsymbol{p}}(t)|^2\, d\boldsymbol{p} \tag{4}$$

is the total instantaneous ionization probability, and $a_{\boldsymbol{p}}(t)$ the instantaneous ionization amplitude. Let us recapitulate briefly the procedure used to calculate the ionization amplitude in the Keldysh theory [also known as the strong field approximation (SFA) theory]. The well-known SFA expression for the ionization amplitude in the length gauge can be written as [1,5,18]

$$a_{\boldsymbol{p}}(t) = -i\int_0^t E(\tau)\langle \boldsymbol{p} + \boldsymbol{A}(\tau) - \boldsymbol{A}(t)|\hat{U}(t,\tau)z\phi_0\rangle e^{-i\varepsilon_0\tau}\, d\tau, \tag{5}$$

where ϕ_0 is the initial atomic state with energy ε_0, $\boldsymbol{A}(t)$ vector potential of the laser pulse, and $\hat{U}(t,\tau)$ is the propagator, which in the SFA takes the form,

$$\langle \boldsymbol{p}|\hat{U}^{\text{SFA}}(t,\tau)|\boldsymbol{p}'\rangle = \exp\left\{-\frac{i}{2}\int_\tau^t (\boldsymbol{p} + \boldsymbol{A}(x) - \boldsymbol{A}(t))^2\, dx\right\} \times \delta(\boldsymbol{p} - \boldsymbol{p}'). \tag{6}$$

Upon substituting this expression into Eq. (5), one obtains an expression for the amplitude,

$$a_{\boldsymbol{p}}(t) = -i\int_0^t g(\tau)e^{-iu(\tau)}\, d\tau, \tag{7}$$

with

$$g(\tau) = E(\tau)\langle \boldsymbol{p} + \boldsymbol{A}(\tau) - \boldsymbol{A}(t)|z|\phi_0\rangle, \tag{8}$$

and

$$u(\tau) = \frac{1}{2}\int_\tau^t (\boldsymbol{p} + \boldsymbol{A}(x) - \boldsymbol{A}(t))^2\, dx + \varepsilon_0\tau. \tag{9}$$

Further development of the SFA theory is based [1,5] on the observation, that if $I/\omega \gg 1$ (where $I = -E$ the ionization potential) the function $u(\tau)$ defined in Eq. (9) is large. The integral in Eq. (7) contains then a rapidly oscillating exponential function, which invites application of the saddle point method [5]. This strategy was used in [15] to evaluate integral (5) for the instantaneous ionization rate.

We should note, however, that for t inside the interval $(0,T_1)$ (where T_1 is the total pulse duration), the contributions due to the saddle points are not the only contributions we have to consider to find the asymptotic behavior of the integral (5). The contributions of the endpoints of the integration interval have to be considered, too. We can use the stationary phase method [17] to elucidate this point.

Let us first consider a simple integral which models expression (7) for the amplitude:

$$I(t) = \int_{-\frac{\pi}{2}}^t \cos x\; e^{i\lambda(\frac{x^3}{3}+bx)}, \tag{10}$$

where $b > 0$, λ is a large parameter (we use $\lambda = 10$ below) and we chose the lower limit of integration so that as in Eq. (7) the integrand vanishes there. The integrand has stationary points at $x_s = \pm i\sqrt{b}$. Taking into account that the derivative of the expression in the exponential function is nonzero on the interval $(0,t)$, and integrating (10) by parts, we obtain in a usual way [17] the first term of the asymptotic expansion of the integral (10) in powers of λ^{-1}:

$$I(t) \approx \frac{\cos t}{i\lambda(t^2+b)}e^{i\lambda(\frac{t^3}{3}+bt)}. \tag{11}$$

In Fig. 1 we present numerically computed quantity $W(t) = |I(t)|^2$ and $w(t) = \frac{dW(t)}{dt}$ as functions of time and compare them with the results obtained using the first term of the asymptotic expansion (11). We see, that for the values of the parameter $b \approx 1$ we obtain very good agreement between the results of the numerical calculation and results of the calculation using the first term of the asymptotic expansion (11). Agreement is getting worse when b decreases and becomes very poor for $b \approx 0.05$. The reason for this is, ultimately, a well-known

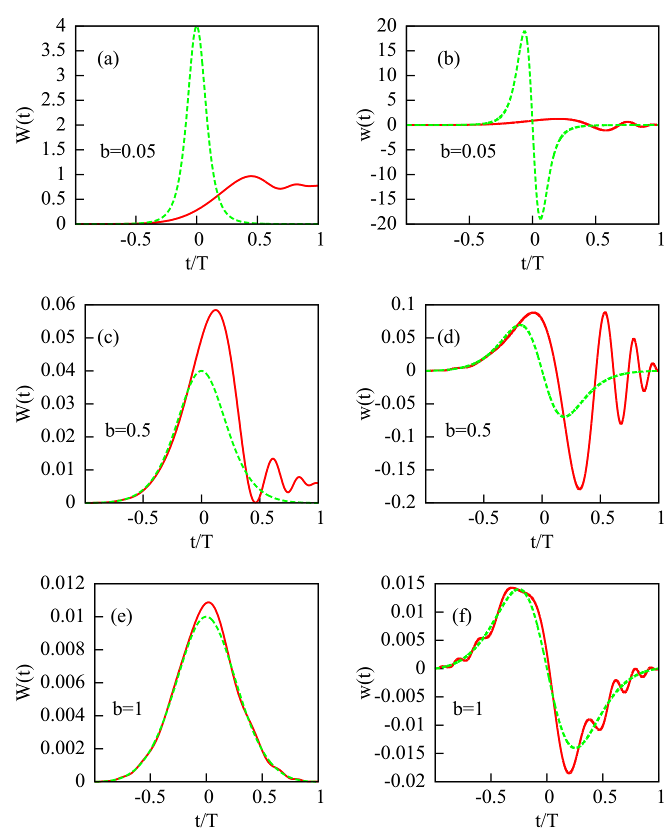

FIG. 1. (Color online) Contribution of the endpoint to the value of $W(t) = |I(t)|^2$ and $w(t) = \frac{dW(t)}{dt}$, where $I(t)$ is an integral (10) for different values of the parameter b. (Red solid line) Numerical calculation; (green dashed line) calculation using the first term of the asymptotic expansion (11). Parameter $T = 2\pi$.

feature [19] of the asymptotic expansions; their accuracy is limited. Equation (11) represents the first term of the asymptotic expansion of the amplitude in powers of a small quantity λ^{-1}. The account of the higher order terms [obtainable by repeated integration by parts in Eq. (10)] can give terms of higher order in λ^{-1}. We can thus obtain the asymptotic series in powers of λ^{-1} for the integral. There is, however, another contribution to the integral, due to the saddle points at $x_s = \pm i\sqrt{b}$. This contribution, as can be easily seen, is (we write only the exponential factor) proportional to $\exp(-\frac{2\lambda}{3}b^{\frac{3}{2}})$. Because of the limited accuracy inherent to the asymptotic series [19], asymptotic expansion in inverse powers of λ cannot reproduce terms decaying exponentially when $\lambda \to \infty$ in true asymptotic of the integral in Eq. (10). If, for a given λ, these terms are small (i.e., parameter b is such that $\lambda b^{\frac{3}{2}} \gg 1$), the endpoint contribution is much larger than the saddle point contribution, and the first term of the asymptotic expansion in powers of λ^{-1} provides a good approximation to the integral (10). If $\lambda b^{\frac{3}{2}} \approx 1$, the saddle point contribution may be significant and Eq. (11) provides a poor approximation to the integral (10). We see, thus, that depending on the value of the parameter b in (10), we may have both the case when the saddle point gives the dominating contribution, and then saddle point analysis would provide an accurate asymptotic estimate, and the case, when the endpoint contribution dominates, and then we have to use the stationary phase method to get an accurate asymptotic estimate.

Let us consider now the more realistic case of the integral in Eq. (7) defining the ionization amplitude. Taking into account that $u'(\tau) \neq 0$ on the interval $(0,T_1)$, and integrating (7) by parts, we obtain the first term of the asymptotic expansion of the integral in Eq. (7) in inverse powers of the quantity $\lambda = |u'(t)|$ which is assumed to be large:

$$a_{\boldsymbol{p}}^{ep}(t) = -i\int_0^t g(\tau)e^{-iu(\tau)}\,d\tau \approx g(t)\frac{e^{-iu(t)}}{u'(t)} = -\frac{2E(t)\langle \boldsymbol{p}|z|\phi_0\rangle}{\boldsymbol{p}^2 - 2\varepsilon_0}e^{-i\varepsilon_0 t}. \quad (12)$$

The corresponding contribution from the endpoint $\tau = 0$ vanishes since the electric field vanishes there, and hence $g(0) = 0$. We use notation $a_{\boldsymbol{p}}^{ep}(t)$ in Eq. (12) to emphasize the fact that $a_{\boldsymbol{p}}^{ep}(t)$ is an endpoint contribution to the amplitude.

Equation (12), in fact, does a very good job in approximating the SFA amplitude for the laser field parameters such that Keldysh parameter $\gamma \approx 1$. As an example, we compare in Fig. 2 results of the numerical evaluation of the SFA amplitude $a_0(t)$ for zero electron momentum and results given by the asymptotic formula (12) for pulses with the base frequency $\omega = 0.057$ a.u. and field strength of 0.05 a.u. For better visibility we present in Fig. 2 the amplitude $b_0 = a_0 e^{i\varepsilon_0 t}$, which does not oscillate so fast in time. We consider a model atom with $\varepsilon_0 = -0.5$ a.u. and assume that the matrix element $\langle \boldsymbol{p}|z|\phi_0\rangle = 1$.

We observe very good agreement between the numerical calculation and the asymptotic formula for the real part of the amplitude b_0. The imaginary part of the amplitude, which for the field parameters we consider is much smaller than the real part, cannot be represented by the asymptotic formula (12).

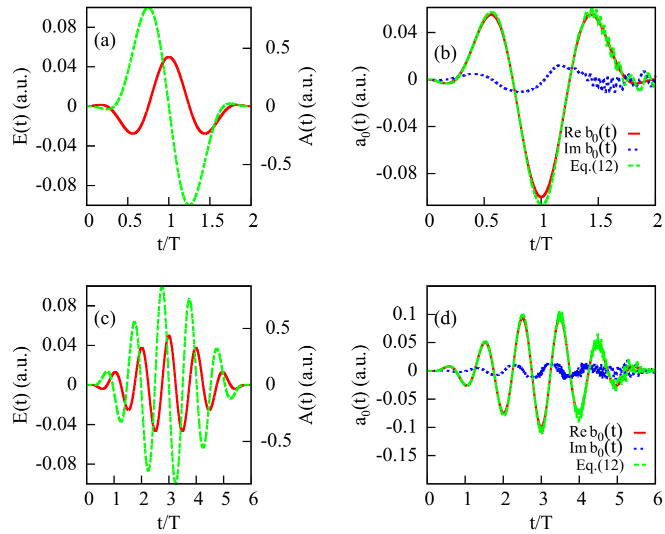

FIG. 2. (Color online) (a) and (c) Electric field (red solid line), and vector potential (green dashed line) of the laser pulse used in Eq. (12). (b) and (d) (Red solid line) Numerically computed real part of the amplitude $b_0(t)$; (green dashed line) amplitude $b_0(t)$ computed according to Eq. (12); (Blue dotted line) numerically computed imaginary part of $b_0(t)$. (Total pulse duration) Two optical cycles (a) and (b), six optical cycles (c) and (d); base frequency $\omega = 0.057$ a.u., peak electric field strength $E_0 = 0.05$ a.u.

The reason for this is the same as in the example we considered above; this is a manifestation of the limited accuracy of the asymptotic expansions. Equation (12) represents the first term of the asymptotic expansion of the amplitude in powers of the small quantity $\lambda = 1/|u'(t)|$. Repeating the integration by the parts procedure, we can obtain higher order terms in λ^{-1}, and thus obtain an asymptotic series in powers of λ^{-1} for the real part of the amplitude b_0. The imaginary part of the amplitude cannot be obtained by this procedure; since it shows parametrically (as a function of λ) different behavior, it is an exponentially small quantity for $\lambda \to \infty$. To obtain the imaginary part, we should use the saddle point method, as was done in [15]. For $\lambda \to \infty$ the imaginary part thus obtained is smaller than the real part at all points of the interval of the laser pulse duration except the points where the laser field is zero. Indeed, Eq. (12) shows that the endpoint contribution vanishes at this point. We arrive, thus, at the following picture. For the large values of the parameter λ introduced above, asymptotic behavior of the instantaneous ionization amplitude at the moment of time t can be can be represented as a sum of two terms:

$$a_{\boldsymbol{p}}(t) \sim a_{\boldsymbol{p}}^{sp}(t) + a_{\boldsymbol{p}}^{ep}(t) \quad (\lambda \to \infty), \quad (13)$$

where $a_{\boldsymbol{p}}^{sp}(t)$ is the contribution due to the saddle points, and $a_{\boldsymbol{p}}^{ep}(t)$ is the endpoint contribution. Depending on the choice of the laser field parameters the relative contributions of the two terms in Eq. (13) may vary. For small values of the Keldysh parameter γ SFA predicts [5] that the saddle point lying in the upper half of the complex t plane (these are the points that matter for the asymptotic evolution of the amplitude) should satisfy $\mathrm{Im}\, t_s \approx \frac{1}{|E_0|}$, where E_0 is the peak field strength. This leads to the well-known law of the exponential decay for the amplitudes $a_{\boldsymbol{p}}^{sp}(t)$ in Eq. (13) when $E_0 \to 0$. We can consider this exponential decay as a signature of the stationary point in Eq. (7). Amplitudes $a_{\boldsymbol{p}}^{ep}(t)$, on the other hand, are rational functions of E_0. For sufficiently small E_0 the contribution of $a_{\boldsymbol{p}}^{ep}(t)$ in Eq. (13) will dominate for all intervals of the laser pulse duration except the point $t = T_1$, where $a_{\boldsymbol{p}}^{ep}(t)$ vanish according to Eq. (12). Inverse electric field strength, in fact, plays the role of the parameter b in the example (10) we considered above. For small E_0 (large b) the saddle point is a complex number with a large imaginary part, its contribution is exponentially small, and endpoint analysis based on the stationary phase method gives an accurate estimate for the amplitude (7) [integral (10)].

Using Eq. (13) we can write for the total instantaneous ionization probability,

$$P(t) \sim P_1(t) + P_2(t) \quad (\lambda \to \infty), \tag{14}$$

where

$$P_1(t) = \int [|a^{sp}(\boldsymbol{p})|^2 + a^{sp*}(\boldsymbol{p})a^{ep}(\boldsymbol{p}) + a^{ep*}(\boldsymbol{p})a^{sp}(\boldsymbol{p})]\, d\boldsymbol{p}, \tag{15}$$

and

$$P_2(t) = \int |a^{ep}(\boldsymbol{p})|^2\, d\boldsymbol{p} = CE^2(t). \tag{16}$$

Expression for C can easily be obtained from Eq. (12), but we will not need its explicit form below. What was said above about the relative roles of the contributions due to the saddle points and endpoints applies to the instantaneous ionization probabilities $P_1(t)$ and $P_2(t)$ and their time derivatives, which give the saddle-point and endpoint contributions to the instantaneous ionization rates. We can expect that for weak fields $P_2(t)$ should represent total instantaneous ionization probability quite accurately at all points of the interval of the laser pulse duration $(0,T_1)$ except the point $t = T_1$. In the next section we verify these statements, comparing asymptotic results for the endpoint contribution with the results obtained by solving the time-dependent Schrödinger equation (TDSE). Before presenting analysis of these results, we should make the following remark. We used the SFA expression to evaluate the ionization amplitude. The exact instantaneous ionization amplitude is given by an expression similar to Eq. (5) with the SFA propagator replaced by the exact propagator, and the plane wave state $|\boldsymbol{p}\rangle$ replaced by the exact scattering state of the system. Repeating the steps of the derivation Eq. (9), Eq. (8), which led to Eq. (12) for the amplitude and Eq. (16) for the ionization probability, we would obtain essentially the same result. The difference with the SFA result would be that factor C in Eq. (16) would generally depend on time. Indeed, time independence of C in the derivation based on the SFA approach was a result of the fact that the endpoint contribution (12) was a function of $\boldsymbol{p}$ only and did not contain the vector potential $\boldsymbol{A}(t)$. If we do not use SFA, in particular if the plane wave in Eq. (5) is replaced with the true continuum scattering state of the system, the endpoint contribution to the amplitude becomes a function of the two arguments $\boldsymbol{p}$ and $\boldsymbol{A}(t)$. Therefore, in the general case the factor C in Eq. (16) may depend on time.

III. RESULTS

We performed a set of numerical calculations of the instantaneous ionization probabilities and ionization rates. The time-dependent Schrödinger equation (TDSE) was solved numerically for the hydrogen atom for pulses of various shapes, duration, and field strength. We use linearly polarized pulses and the geometry with the polarization vector pointing in the z direction. The method we employed to solve TDSE has been described in [20,21]. We shall give, therefore, only a brief description of the procedure. To treat spatial variables TDSE is discretized on the grid with the stepsize $\delta r = 0.05$ a.u. in a box of the size $R_{\max} = 1000$ a.u. The wave function is represented as

$$\Psi(\boldsymbol{r},t) = \sum_l f_l(r,t)Y_{l0}(\theta), \tag{17}$$

where summation in Eq. (17) is restricted to $l = 0 - L_{\max}$. The particular value of the parameter $L_{\max}$ is determined by the convergence properties of Eq. (17). The value we used in the calculations reported below was $L_{\max} = 60$. We have performed several checks to ensure that these value of $L_{\max}$ and $R_{\max}$ were sufficient to solve accurately TDSE for the EM fields we consider below. To propagate the wave function (17) in time we use the matrix iteration method (MIM) developed in [22].

To compute the function $P(t)$ defined in Eq. (4), we project the solution of the TDSE $\Psi(t)$ at various instances of time in the course of evolution on the set of the continuous spectrum wave functions of the hydrogen atom $|klm\rangle$, obtaining a set of the coefficients $a_{klm}(t) = \langle klm|\Psi(t)\rangle$ (for the geometry we use and the 1s state of hydrogen, which we use as the initial state, only the coefficients with $m = 0$ have nonzero values, of course). Using the set of the coefficients $a_{kl0}(t)$ we computed $P(t)$ in Eq. (4) as

$$P(t) = \sum_{l=0}^{L_{\max}} \int_0^\infty |a_{kl0}(t)|^2 dk. \tag{18}$$

Instantaneous ionization rate $w(t)$ was found by differentiating $P(t)$. In Fig. 3 we show results for the instantaneous total ionization probability for the ionization process driven by the laser pulse with the electric field:

$$E(t) = E_0 \sin^2\left(\frac{\pi t}{T_1}\right) \cos \omega t, \tag{19}$$

for $t \in (0,T_1)$, and $E(t) = 0$ outside this interval. Base frequency of the pulse was $\omega = 0.057$ a.u. (wavelength of 800 nm), total pulse duration $T_1 = 2T$, where $T = 2\pi/\omega$ is the optical cycle corresponding to the base frequency ω. We performed calculations for various values of the pulse peak strengths E_0.

We see, that in agreement with the discussion we presented above and Eq. (16), instantaneous ionization probability for weak fields in Figs. 3(a) and 3(b), when the saddle-point contribution is exponentially small, mimics almost exactly the squared instantaneous electric field of the pulse. For these fields the endpoint contribution $P_2(t)$ in Eq. (14) is a dominating contribution, which actually defines the instantaneous ionization probability for times inside the interval of the pulse

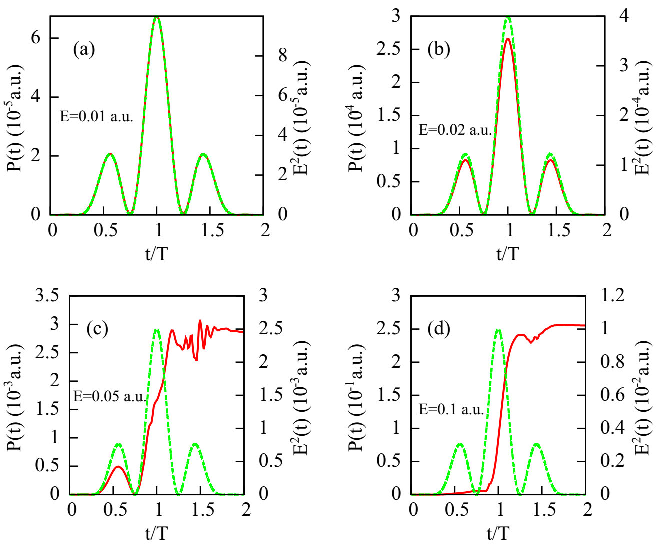

FIG. 3. (Color online) (Red solid line) TDSE results for the instantaneous ionization probability as a function of time for the laser pulse given by Eq. (19) with total pulse duration $T_1 = 2T$, where $T = 2\pi/\omega$, an optical cycle, and peak field strengths of 0.01 a.u., 0.02 a.u., 0.05 a.u., 0.1 a.u. (a)–(d). (Green dashed line) Squared electric field of the pulse $E^2(t)$.

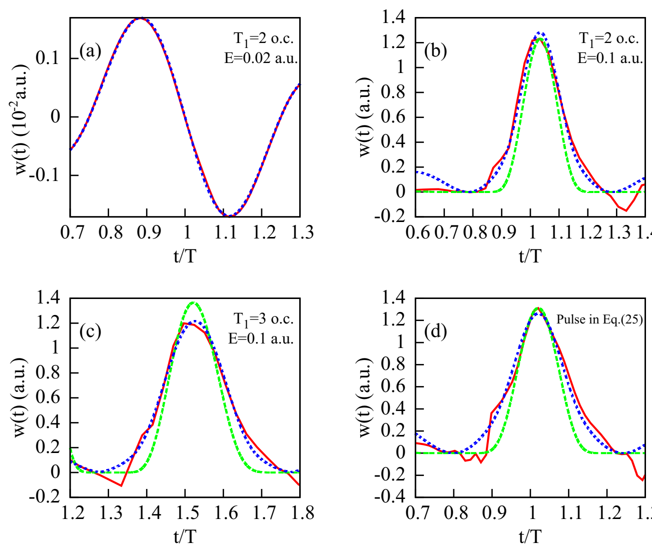

FIG. 4. (Color online) Instantaneous ionization rate as a function of time for the laser pulse (19) with total pulse duration $T_1 = 2$ optical cycles and the peak field strength $E_0 = 0.02$ a.u. (a), $T_1 = 2$ o.c. and $E_0 = 0.1$ a.u. (b), $T_1 = 3$ o.c. and $E_0 = 0.1$ a.u. (c). (d) Instantaneous ionization rate as a function of time for the laser pulse (25) with total pulse duration $T_1 = 2$ o.c. and $E_0 = 0.1$ a.u. (Red solid line) TDSE results; (green dashed line) quasistatic formula; (blue dotted line) results of the fits based on Eqs. (21) and (24).

duration. Of course, the endpoint contribution vanishes at the end of the laser pulse, so that total ionization probability at the end of the laser pulse is always determined by the saddle-point contribution. For stronger electric fields, exceeding 0.05 a.u. in Figs. 3(c) and 3(d) the contribution of the saddle point is a dominating one. Nevertheless, even for such fields the account of the endpoint contribution may prove important. Let us consider the instantaneous ionization rate $w(t)$.

Taking the derivative with respect to time of both sides of Eq. (14) and using Eq. (16) we find the following formula for $w(t)$:

$$w(t) = \frac{dP_1(t)}{dt} + \frac{dC(t)}{dt}E^2(t) + 2C(t)E(t)\frac{dE(t)}{dt}. \quad (20)$$

Relative contributions of the three terms in Eq. (20) depend, of course, on the field strength we employ. We have seen that for the fields of the order of 0.02 a.u. the first term on the right-hand side can be safely neglected. One can argue that the term quadratic in the field strength may also be small, and can be neglected, too. That this is indeed the case can be seen from Fig. 4. In the figure we show ionization rates given by the TDSE calculation. For the weak field of 0.02 a.u. $w(t)$ is perfectly fitted by the formula following from Eq. (20), assuming we keep only the third term on the right-hand side of this equation and treat $C(t)$ as a constant fitting parameter:

$$w(t) \approx CE(t)\frac{dE(t)}{dt}, \quad (21)$$

where $E(t)$ is the laser pulse electric field given by Eq. (19).

An expression often used to estimate the instantaneous ionization rate is the so-called quasistatic formula, which for the hydrogen atom we consider reads

$$w_{qs}(t) = \frac{4}{|E(t)|}e^{-\frac{2}{3|E(t)|}}, \quad (22)$$

where $E(t)$ is the instantaneous electric field. This formula results from the evaluation of the ionization amplitude using the saddle-point method for the case of the constant electric field, and can be interpreted, therefore, as a saddle-point contribution. Other formulas have been proposed [7,23] to describe behavior of the instantaneous ionization rate. They give different pre-exponential factors in Eq. (22), but essential for the purpose of the present paper is the exponential factor in Eq. (22), which can be regarded as a signature of the saddle point causing the exponential behavior of the amplitude.

Figure 4(a) shows that Eq. (16) and the expression for the ionization rate derived from it indeed work well in the weak field case. In this regime the behavior of the instantaneous ionization rate as a function of time can in no way be represented by quasistatic (or analogous) formulas. Indeed, one can see from Fig. 4 that the instantaneous ionization rate can assume negative values, which are not reproduced by the quasistatic expression. The negative instantaneous ionization rate describes a process of the electron's return to the atom. More precisely, the total instantaneous ionization probability and rate were defined above in terms of the norm of the projection of the wave function on the positive energy part of the spectrum of the atomic Hamiltonian. The negative ionization rate means, therefore, a decrease of this norm and a corresponding increase of the norm of the projection of the wave function on the discrete energy eigenstates. As we saw, the account of this process can prove important for weak electric fields.

Consider the case of the stronger field with the peak strength $E_0 = 0.1$ a.u. (corresponding to the intensity of 3.5×10^{14} W/cm^2).

By differentiating Eq. (15) we can see that the time derivative $\frac{dP_1(t)}{dt}$ in Eq. (20) is generally a sum of two contributions: the contribution due to the saddle point and the term due to the interference between saddle-point and endpoint amplitudes. One can argue that if we want to describe the ionization rate curve $w(t)$ for the moments of time t at some distance from the maxima of the electric field $E(t)$, the interference term can be neglected. Indeed, the saddle-point contribution to the amplitude decays exponentially when the electric field decreases, while the contribution due to the endpoint is a rational function of the electric field strength. The endpoint contribution to the instantaneous ionization rate will, therefore, eventually dominate. Approximating the rate $\frac{dP_1(t)}{dt}$ in Eq. (20) by the quasistatic formula, we arrive at the following equation for the instantaneous ionization rate:

$$w(t) = Aw_{qs}(t) + \frac{dC(t)}{dt}E^2(t) + 2C(t)E(t)\frac{dE(t)}{dt}, \quad (23)$$

where we scale the quasistatic rate to achieve better agreement. Assuming further that for strong enough fields the third term in Eq. (23) can be neglected, and considering $B = \frac{dC(t)}{dt}$ as a constant factor, we obtain the fitting formula:

$$w(t) \approx Aw_{qs}(t) + BE^2(t). \quad (24)$$

Results of the fits based on Eq. (24) are shown in Fig. 4 for the pulse given in Eq. (19) for the peak strength of $E_0 = 0.1$ and the total duration of two and three optical cycles. One can see that adding the endpoint contribution $BE^2(t)$ improves the fits considerably. To make sure that the good agreement between numerically computed $w(t)$ and the ionization rate obtained using Eq. (24) does not depend on the particular shape of the laser pulse, we performed a calculation using the laser pulse defined in terms not of the electric field as in Eq. (19), but in terms of the vector potential:

$$A(t) = \frac{E_0}{\omega}\sin^2\left(\frac{\pi t}{T_1}\right)\sin\omega t, \quad (25)$$

for $t \in (0,T_1)$, and $A(t) = 0$ outside this interval. As before, the peak field strength is $E_0 = 0.1$ a.u., and base frequency $\omega = 0.057$ a.u. Results of the fit based on Eq. (24) for this pulse are shown in Fig. 4.

IV. CONCLUSION

We considered the contribution of the endpoint to the integral (7) defining the instantaneous ionization amplitude. This contribution should be added to the contribution of the saddle point, which is usually used to evaluate the ionization amplitude. We saw that depending on the field strength, relative contributions of the saddle-point and endpoint contributions may vary. The saddle-point contribution becomes exponentially small in the weak field limit. The endpoint contribution, on the other hand, is a rational function of the electric field strength. Therefore, for the weak fields (field strength of the order of 0.02 a.u. in the examples we considered above), the endpoint contribution plays a dominant role. For stronger fields, with the field strength of the order of 0.1 a.u., we may still need the saddle-point contribution to describe the behavior of the instantaneous ionization amplitude in the region not in the intermediate vicinity of the maximum of the instantaneous electric field.

Equation (14) represents total instantaneous ionization probability as a sum of the contributions due to the saddle point and the endpoint. The endpoint contribution vanishes at the moment of time when the electric field is zero. In particular, the endpoint contribution vanishes at the end of the laser pulse, so that total ionization probability at the end of the laser pulse is always determined by the saddle-point contribution. In particular, if we took the expression for the ionization rate (20), which contains the endpoint contribution, and integrated it over the pulse duration (or any integer number of cycles of the laser pulse), the integral would have the same value we would have obtained had only the saddle-point contribution been taken into account.

It may seem, therefore, that the endpoint contribution we discussed in the paper is, to a degree, unphysical. We believe it is not quite so. The instantaneous ionization rate is often used in modeling the ionization phenomena in complex systems, where *ab initio* TDSE calculations become prohibitively complex. An example is the TIPIS model [12,13], where the ionization process is modeled by launching classical trajectories at different moments of time, contributions of different trajectories being weighted using an appropriate expression for the instantaneous ionization rate. As we have seen, the account of the endpoint contribution may considerably modify the instantaneous ionization rate.

ACKNOWLEDGMENTS

The authors acknowledge support from the Institute for Basic Science, Gwangju, Republic of Korea. I.A.I. benefited from useful discussion with Dr. O. I. Tolstikhin.

[1] L. V. Keldysh, Sov. Phys. JETP **20**, 1307 (1965).
[2] F. H. M. Faisal, J. Phys. B **6**, L89 (1973).
[3] H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).
[4] A. M. Perelomov, V. S. Popov, and M. V. Terentiev, Sov. Phys. JETP **23**, 924 (1966).
[5] V. S. Popov, Physics-Uspekhi **47**, 855 (2004).
[6] S. V. Popruzhenko, J. Phys. B **47**, 204001 (2014).
[7] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Sov. Phys. JETP **64**, 1191 (1986).
[8] O. I. Tolstikhin and T. Morishita, Phys. Rev. A **86**, 043417 (2012).
[9] A. Baltuška, T. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V. S. Yakovlev, A. Scrinzi, T. W. Hänsch *et al.*, Nature (London) **421**, 611 (2003).
[10] P. Eckle, A. N. Pfeiffer, C. Cirelli, A. Staudte, R. Dörner, H. G. Muller, M. Büttiker, and U. Keller, Science **322**, 1525 (2008).
[11] A. Saenz and M. Awasthi, Phys. Rev. A **76**, 067401 (2007).

[12] N. I. Shvetsov-Shilovski, D. Dimitrovski, and L. B. Madsen, Phys. Rev. A **85**, 023428 (2012).
[13] R. Boge, C. Cirelli, A. S. Landsman, S. Heuser, A. Ludwig, J. Maurer, M. Weger, L. Gallmann, and U. Keller, Phys. Rev. Lett. **111**, 103003 (2013).
[14] K. T. Kim, C. Zhang, A. D. Shiner, S. E. Kirkwood, E. Frumker, G. Gariepy, A. Naumov, D. M. Villeneuve, and P. B. Corkum, Nat. Phys. **9**, 159 (2013).
[15] G. L. Yudin and M. Y. Ivanov, Phys. Rev. A **64**, 013409 (2001).
[16] O. Smirnova, M. Spanner, and M. Ivanov, J. Phys. B **39**, S307 (2006).
[17] N. Bleistein and R. Handelsman, *Asymptotic Expansions of Integrals* (Dover, New York, 1975).
[18] I. Dreissigacker and M. Lein, Chem. Phys. **414**, 69 (2013).
[19] G. H. Hardy, *Divergent Series* (Oxford University Press, London, 1973).
[20] I. A. Ivanov, Phys. Rev. A **83**, 023421 (2011).
[21] I. A. Ivanov, Phys. Rev. A **90**, 013418 (2014).
[22] M. Nurhuda and F. H. M. Faisal, Phys. Rev. A **60**, 3125 (1999).
[23] Y. V. Vanne and A. Saenz, Phys. Rev. A **75**, 063403 (2007).