Nonadiabatic tunnel ionization: Looking inside a laser cycle

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We obtain a simple closed-form analytical expression for ionization rate as a function of instantaneous laser phase $\phi(t)$, for arbitrary values of the Keldysh parameter γ, within the usual strong-field approximation. Our analysis allows us to explicitly distinguish multiphoton and tunneling contributions to the total ionization probability. The range of intermediate $\gamma \sim 1$, which is typical for most current intense field experiments, is the regime of nonadiabatic tunneling. In this regime, the instantaneous laser phase dependence differs dramatically from both quasistatic tunneling and multiphoton limits. For cycle-averaged rates, our results reproduce standard Keldysh-like expressions.

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Traditionally, beginning with the pioneering paper by Keldysh [1], rates of intense-field ionization in both the multiphoton and tunneling limit are averaged over the laser cycle. Today, this is no longer sufficient. Subcycle electron dynamics is now known to play a key role in such processes as high harmonic generation and above-threshold ionization in intense low-frequency laser fields (see, e.g., Refs. [2,3]). It is also crucial for the understanding of correlated double multiphoton ionization of atoms in intense laser fields (see, e.g., [4] and references therein). In general, in few-cycle laser pulses, intensity changes from one cycle to the next, making cycle-averaging meaningless, especially for highly nonlinear processes. Subcycle dynamics of multiphoton ionization is the basis of the proposed approaches in [5,6] to measure the absolute carrier phase φ_0 of the electric-field oscillation under the envelope [7]. For linear polarization, the absolute phase φ_0 is defined as $\vec{\mathcal{E}} f(t)\cos(\omega_L t+\varphi_0)$ [$\mathcal{E}$ is the amplitude, $f(t)$ is the envelope, and ω_L is the frequency of the laser field]. Since pulse-to-pulse stability of φ_0 has now been experimentally achieved [7], measuring φ_0 remains the biggest challenge. Our results provide a simple way of evaluating the feasibility of various approaches to measuring φ_0, which are based on subcycle intense field ionization dynamics.

Experimentally, in intense-field multiphoton ionization one is typically dealing with intermediate values of the Keldysh parameter $\gamma \sim 1$. Here $\gamma^2 = I_p/2U_p$, I_p is the ionization potential, and $U_p = \mathcal{E}^2/4\omega_L^2$ is the average energy of electron oscillations in the laser field (atomic units are used throughout the paper). It is common to model ionization in this regime using quasistatic approximation to the rate of tunnel ionization:

$$\Gamma_{\rm qs}(t) = A_{n^*,l^*} B_{l,|m|} I_p \left(\frac{2(2I_p)^{3/2}}{\mathcal{E} f(t)|\cos\phi(t)|} \right)^{2n^*-|m|-1} \times \exp\left(-\frac{2(2I_p)^{3/2}}{3\mathcal{E} f(t)|\cos\phi(t)|} \right). \tag{1}$$

Here $\phi(t) = \omega_L t + \varphi_0$ is the instantaneous phase of the linearly polarized laser field. The coefficient A_{n^*,l^*} comes from the radial part of the wave function at $r \gg 1/\sqrt{2I_p}$ and depends on the effective principal quantum number $n^* = Z/\sqrt{2I_p}$ (Z is the ion charge) and the effective angular momentum l^*. The coefficient $B_{l,|m|}$ comes from the angular part of the wave function and depends on the actual angular momentum l and its projection m on the laser polarization vector. The corresponding expressions are [8–10]

$$A_{n^*,l^*} = \frac{2^{2n^*}}{n^*\Gamma(n^*+l^*+1)\Gamma(n^*-l^*)}, \quad B_{l,|m|} = \frac{(2l+1)(l+|m|)!}{2^{|m|}|m|!(l-|m|)!}, \tag{2}$$

where $\Gamma(z)$ is the gamma function. Averaging Eq. (1) over a laser half-cycle for a sufficiently smooth envelope $f(t)$, one obtains the well-known tunneling formula from Refs. [8–10].

Quasistatic approximation Eq. (1), which includes the effect of the Coulomb potential, is rigorously valid only in the limit $\gamma \ll 1$. The reasons for the frequent use of Eq. (1) for intermediate values of γ are (i) computational convenience, (ii) absence of simple closed-form analytical expressions for instantaneous ionization rates for $\gamma \sim 1$, and (iii) good accuracy of *cycle-averaged* tunneling ionization rates up to $\gamma \sim 0.5$ [11]. Here we provide simple closed-form analytical expressions for instantaneous ionization rates for arbitrary γ. We note that for cycle-averaged rates, an analytical expression that is valid in a broad range of γ has been derived in [8,9]. Its accuracy is excellent for many atoms (He, Ne, Ar, Kr, Xe) up to $\gamma = 3-4$ [12] and even many small molecules [13], and is only restricted by the condition $\omega_L \ll \omega_{\rm exc}$ (here $\omega_{\rm exc}$ is the characteristic energy of electronic excitation), which ensures adiabatic electron dynamics inside the potential well.

Dependence of ionization on the instantaneous phase of the laser field is apparent in *ab initio* numerical simulations and is present in intermediate expressions of many analytical approaches; see, e.g., [1,8,9,14–18]. These intermediate ex-

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pressions are given by multidimensional integrals, which obfuscate the subcycle dynamics of electron appearance in the continuum. The dynamics is revealed by the saddle-point analysis, which is possible in the low-frequency case $\omega_L \ll I_p$ and is most straightforward in the limits $\gamma \ll 1$ (quasistatic tunneling) and $\gamma \gg 1$ (multiphoton limit). Our analysis for arbitrary γ allows us to explicitly distinguish multiphoton and tunneling contributions to the total ionization probability and shows that for $\gamma \sim 1$, tunneling is still dominant but differs significantly from its quasistatic limit Eq. (1).

The population of continuum states at instant t is

$$W(t) = \int d^3v |a_{\mathbf{v}}(t)|^2, \tag{3}$$

where $a_{\mathbf{v}}(t)$ is the probability amplitude of populating the field-free continuum state labeled by the velocity $|\mathbf{v}\rangle$.

Using the Dykhne method [19,20] and the strong-field approximation, with exponential accuracy we obtain

$$a_{\mathbf{v}}(t) \sim \int_{-\infty}^{t} dt' \exp(-iS_{\mathbf{v}}(t,t')), \tag{4}$$

where

$$S_{\mathbf{v}}(t,t') = (I_p + \tfrac{1}{2}v_\perp^2)(t-t') + \tfrac{1}{2}\int_{t'}^{t} dt''[v_\parallel + v_0 f(t) \times \sin\phi(t) - v_0 f(t'')\sin\phi(t'')]^2 \tag{5}$$

is the action integral, $v_\parallel$ and $v_\perp$ are the velocity components parallel and perpendicular to $\vec{\mathcal{E}}$, and $v_0 = \mathcal{E}/\omega_L$ is the velocity amplitude of electron oscillations. With exponential accuracy, $v_\perp$ can be set to zero, since the Gaussian integral over $v_\perp$ only contributes to the preexponential factor.

Let us first assume that $f(t)$ is constant, $f=1$. Generalization for short pulses and the appropriate criterion are given below. The integral over t' contains many saddle points $t' < t$ given by the equation

$$[v_\parallel + \sin\phi(t) - \sin\phi(t')]^2 + \gamma^2 = 0. \tag{6}$$

For the saddle point closest to t, denoted as t_0', $\mathrm{Re}(t_0') \approx t$. Other t_n' are separated from t_0' by the integer number of cycles, $t_n' = t_0' - 2\pi n$, integer $n \geq 1$. For constant $f(t)$, the imaginary part of action is the same for all of them,

$$\mathrm{Im}[S_v(t,t_n')] = \mathrm{Im}[S_v(t,t_0')]. \tag{7}$$

The physical meaning of these saddle points is as follows.

(i) The contribution of the saddle point t_0' describes the population that has just appeared in the continuum at instant t; the complex t_0' is the moment when the electron enters the classically forbidden region (under the barrier) while t is ''the moment of birth'' in the continuum.

(ii) Contributions of the saddle points t_n', $n \geq 1$, describe the population created in the continuum one or more laser cycles ago; the corresponding action integrals contain contributions from the free-electron motion in the continuum.

(iii) Since we are interested in the contribution to ionization at instant t, we should only take into account the saddle point t_0'. We note that due to Eq. (7), exponential dependence is determined by t_0', while other t_n' with $n \geq 1$ contribute to the preexponential factor.

(iv) Physically, for t_0' the velocity $v_\parallel$ is the longitudinal electron velocity at the moment of birth in the continuum and, as known since [1], $v_\parallel^2/2 \ll I_p$. One can therefore expand the exponent in $a_{\mathbf{v}}(t)$ in powers of $v_\parallel$ up to the second order near $v_\parallel = 0$, with the saddle point t_0' of the inner integral calculated for $v_\parallel = 0$. Once again, the exponential dependence is determined predominantly by the contribution at $v_\parallel = 0$, while the Gaussian integral over $v_\parallel$ adds to the preexponential factor [1,8,9,20].

For convenience, we introduce the phase $\theta(t)$ defined as

$$\theta(t) = \phi(t) - \pi k = \omega_L t + \varphi_0 - \pi k \tag{8}$$

with the integer k chosen to ensure that

$$-\pi/2 \leq \theta(t) \leq \pi/2. \tag{9}$$

This phase is always equal to zero at the local peaks of the instantaneous electric field. It is easy to check that if the field envelope is constant during the half-cycle, the function $S_0(\theta(t)) = S_{v=0}(t,t_0')$ has the following properties:

$$\mathrm{Im}[S_0(-\theta)] = \mathrm{Im}[S_0(+\theta)], \qquad \mathrm{Re}[S_0(-\theta)] = -\mathrm{Re}[S_0(+\theta)]. \tag{10}$$

Therefore, for constant envelope the instantaneous ionization rate $\Gamma(t) \sim \exp(-2\,\mathrm{Im}[S_0(\theta(t))])$ is an even function of θ.

With exponential accuracy, the result for $\Gamma(t)$ is

$$\Gamma(t) \sim \exp\left(-\frac{\mathcal{E}^2 f^2(t)}{\omega_L^3}\Phi(\gamma(t),\theta(t))\right), \tag{11}$$

where the Keldysh parameter $\gamma(t) = \gamma/f(t)$ now includes the pulse envelope. The function $\Phi(\gamma,\theta)$ is given by the following expression:

$$\Phi(\gamma,\theta) = (\gamma^2 + \sin^2\theta + \tfrac{1}{2})\ln c - \frac{3\sqrt{b-a}}{2\sqrt{2}}\sin|\theta| - \frac{\sqrt{b+a}}{2\sqrt{2}}\gamma,$$

$$a = 1 + \gamma^2 - \sin^2\theta, \qquad b = \sqrt{a^2 + 4\gamma^2\sin^2\theta}, \tag{12}$$

$$c = \sqrt{\left(\sqrt{\frac{b+a}{2}} + \gamma\right)^2 + \left(\sqrt{\frac{b-a}{2}} + \sin|\theta|\right)^2}.$$

We have reintroduced the envelope into Eq. (11) by treating $f(t)$ as nearly constant during one-half of the laser cycle. Compared to such processes as the generation of high harmonics, for ionization the requirements of the envelope are less strict. In high harmonic generation, the electron spends one or more cycles in the continuum before recombining

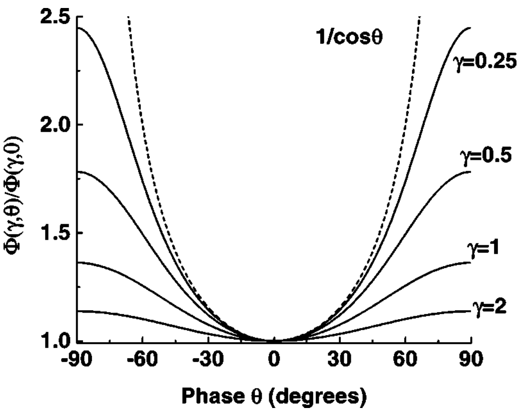

FIG. 1. Dependence of the exponent $\Phi(\gamma,\theta)$ in the instantaneous ionization rate on the phase $\theta=\omega_L t+\varphi_0-\pi k$. Data are normalized to $\Phi(\gamma,0)$. Labels near the solid curves indicate values of the Keldysh parameter γ. Dashed curve shows the quasistatic limit $\gamma/\cos\theta \ll 1$.

with the parent ion and adiabatic treatment of the envelope requires $f(t)$ to remain constant during all this time. In ionization, $f(t)$ should remain constant only during a much shorter *tunneling time* $\tau_{\rm tun}$ given by the imaginary part of $t-t_0'$:

$$\omega_L \tau_{\rm tun} = -\,{\rm Im}[\arcsin(\sin\theta - i\gamma)]. \tag{13}$$

The phase dependence of

$$R(\gamma,\theta)=\frac{\Phi(\gamma,\theta)}{\Phi(\gamma,0)} \tag{14}$$

[i.e., $\Phi(\gamma,\theta)$ normalized to $\theta=0$] for different γ is shown in Fig. 1, together with the quasistatic limit $1/\cos\theta$.

Several remarks on the results of Eqs. (11) and (12) and Fig. 1 are in order.

(i) As expected, there is no θ dependence in $\Phi(\gamma,\theta)$ in the multiphoton limit $\gamma \gg 1$.

(ii) As γ decreases, clear phase dependence appears, reflecting a tunneling contribution to the total ionization rate. The rate peaks at $\theta=0$ and $\Phi(\gamma,\theta)$ is a quadratic function of θ at small θ.

(iii) The original Keldysh exponent for the cycle-averaged ionization rate is obtained by setting $\theta=0$ in Eqs. (11) and (12). After the previous remark, this is no longer surprising. Indeed, at large $\gamma \gg 1$ (multiphoton limit), the θ dependence disappears and one can set $\theta=0$ in the average rate. At small and intermediate, γ, the factor $\mathcal{E}^2/\omega_L^3$ in Eq. (11) is large, $\mathcal{E}^2/\omega_L^3=2I_p/(\gamma^2\omega_L)\gg 1$, and averaging of $\Gamma(\gamma,\theta)$ over θ can be done using the saddle-point method. Since the corresponding saddle point is $\theta=0$ and $[\Phi(\gamma,\theta)-\Phi(\gamma,0)]\propto\theta^2$ for small θ, the exponential dependence is given by $\Phi(\gamma,0)$.

(iv) The multiphoton contribution to the total ionization rate can be defined as the phase-independent background, which is given by $\Phi(\gamma,\pm\pi/2)$. For $\gamma \gg 1$,

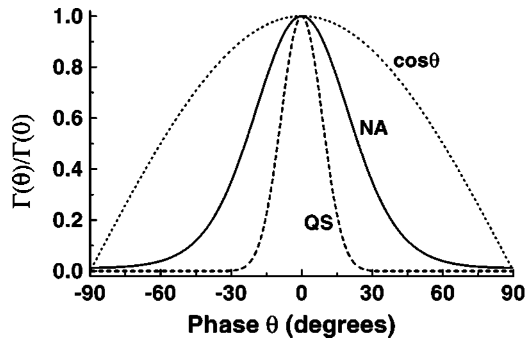

FIG. 2. Dependence of the instantaneous ionization rate $\Gamma(\theta)$ on θ. Data are normalized to $\Gamma(0)$. Labels ''QS'' (dashed curve) and ''NA'' (solid curve) stand for ''quasistatic'' and ''nonadiabatic.'' Dotted curve shows instantaneous electric field. Calculations are done for He and laser field with wavelength $\lambda=780$ nm and intensity $I=5\times10^{13}$ W/cm^2.

$$\Phi(\gamma,\theta)\approx\Phi(\gamma,\pm\pi/2) = (\gamma^2+\tfrac{3}{2})\ln(2\gamma)-\tfrac{1}{2}\gamma^2-1+\frac{35}{16\gamma^2}+\cdots. \tag{15}$$

For $\gamma \leq 2$, the following approximate expression is valid:

$$\Phi(\gamma,\pi/2)\approx\tfrac{4}{5}\gamma^2\sqrt{\gamma}(1+\tfrac{3}{50}\gamma-\tfrac{1}{80}\gamma^2). \tag{16}$$

The leading term in this expansion is exact for $\gamma \ll 1$ and is accurate up to $\gamma=1$ within 5%.

At $\gamma \sim 1$, the multiphoton contribution remains small, but tunneling differs significantly from the quasistatic limit. This is illustrated in Fig. 2, where $\Gamma(t)$ is calculated for a helium atom and the laser wavelength $\lambda=780$ nm. The curves show the ionization rate as a function of θ normalized to $\theta=0$, $\Gamma(\theta)/\Gamma(0)$, for intensity $I=5\times10^{13}$ W/cm^2 (when $\gamma\approx2$). Although the multiphoton contribution (phase-independent background) is small (approximately 3.3% in the total rate integrated over a half-cycle), the instantaneous rate clearly differs from the quasistatic limit.

So far, we have only looked at the exponential dependence in $\Gamma(t)$. One would like to add the preexponential factor, $N(t)$, to the exponential dependence Eq. (11):

$$\Gamma(t)=N(t)\exp\left(-\frac{\mathcal{E}^2 f^2(t)}{\omega_L^3}\Phi(\gamma(t),\theta(t))\right). \tag{17}$$

The subcycle dependence in the preexponential factor $N(t)$ can be ignored (up to the electric-field envelope). Indeed, at $\gamma \ll 1$ and $\gamma\approx1$, ionization is strongly peaked around $\phi(t)=\omega_L t+\varphi_0=\pi k$ and we only need to know $N(\phi=\pi k)$. At $\gamma \gg 1$, the subcycle dependence disappears, and knowing $N(\phi=\pi k)$ is again sufficient. Hence, it is sufficient to include the time dependence in $N(t)$ via the envelope $\mathcal{E}f(t)$ only.

The simplest way to define $N(t)$ in Eq. (17) is to match our result with the cycle-averaged result of [8], which is also valid for $\gamma>1$. The corresponding preexponential factor is

$$N(t)=A_{n*,l*}B_{l,|m|}\left(\frac{3\kappa}{\gamma^3}\right)^{1/2}C\,I_p\left(\frac{2(2I_p)^{3/2}}{\mathcal{E}f(t)}\right)^{2n*-|m|-1}, \tag{18}$$

$$\kappa=\ln(\gamma+\sqrt{\gamma^2+1})-\frac{\gamma}{\sqrt{\gamma^2+1}},$$

where $A_{n*,l*}$ and $B_{l,|m|}$ are given by Eqs. (2) and the factor $C=(1+\gamma^2)^{|m|/2+3/4}A_m(\omega_L,\gamma)$ is the Perelomov-Popov-Terent'ev (PPT) correction to the quasistatic limit $\gamma\ll 1$ of the Coulomb preexponential factor, with A_m given by Eqs. (55) and (56) of Ref. [8].

We would like to stress that the key difference from the quasistatic tunneling limit is in the exponent. The PPT correction C is in practice a slow function of γ. In the limit $\gamma\ll 1$, the factor $C=1$, while in the limit $\gamma\gg 1$ for $m=0$ one has $A_0\approx 1.2/\gamma^2$ and $C\approx 1.2/\sqrt{\gamma}$.

The subcycle ionization dynamics plays the key role in such processes as multiphoton correlated double ionization of noble gases (see, e.g., [4], and references therein) and intense-field ionization with single-cycle pulses, where it could be the basis for measuring the absolute carrier phase [5–7]. Such measurement can also be done with circular polarization where the instantaneous phase $\theta(t)$ determines the direction of the final drift velocity of the freed electrons. In this case, instantaneous ionization rates Eqs. (11) and (12) have the following phase-independent form:

$$\Gamma_{\rm circ}(t)\sim\exp\left(-\frac{\mathcal{E}^2 f^2(t)}{\omega_L^3}\Phi_{\rm circ}(\gamma(t))\right), \tag{19}$$

where the function $\Phi_{\rm circ}(\gamma)$ is

$$\Phi_{\rm circ}(\gamma)=(\gamma^2+2)\cosh^{-1}\left(1+\frac{\gamma^2}{2}\right)-2\gamma\sqrt{1+\frac{\gamma^2}{4}}. \tag{20}$$

In conclusion, simple expressions for the subcycle ionization dynamics derived in this paper can be used as a basis for evaluating the feasibility of different approaches to measuring the absolute carrier phase of few-cycle pulses in a typical experimental regime of $\gamma\sim 1$.

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