Ionization Time and Exit Momentum in Strong-Field Tunnel Ionization

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Tunnel ionization belongs to the fundamental processes of atomic physics. The so-called two-step model, which describes the ionization as instantaneous tunneling at the electric field maximum and classical motion afterwards with zero exit momentum, is commonly employed to describe tunnel ionization in adiabatic regimes. In this contribution, we show by solving numerically the time-dependent Schrödinger equation in one dimension and employing a virtual detector at the tunnel exit that there is a nonvanishing positive time delay between the electric field maximum and the instant of ionization. Moreover, we find a nonzero exit momentum in the direction of the electric field. To extract proper tunneling times from asymptotic momentum distributions of ionized electrons, it is essential to incorporate the electron's initial momentum in the direction of the external electric field.

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In 1932, MacColl [1] studied the time which may be associated with the process of a particle approaching from far away a potential barrier of a height larger than the particle's energy and eventually tunneling through the barrier. Many efforts have been directed toward defining [2–4] and measuring [5–9] tunneling times. A related open problem is the question of how long it takes to ionize by tunneling through a binding potential. Keller *et al.* have conducted experiments using the angular streaking technique aiming to measure tunneling times for ionization from a bound state, the so-called attoclock experiments [10,11]. In the tunnel ionization case, a Coulomb-bound electron is ionized by a strong electromagnetic field, and a potential barrier can be defined via the electron's binding energy and the Coulomb potential bent by the electric field's potential; see Fig. 1. Since the attoclock experiments have been performed, many renewed efforts have been directed toward defining a tunnel ionization time, because a consensus on a suitable theoretical definition of tunneling time and the interpretation of experimental results is still lacking [10–17].

In the following, we study the time delay τ_A between the instant of ionization, i.e., when the electron exits the barrier, and the instant of electric field maximum t_0. One may reconstruct the moment of ionization from the electron's asymptotic momentum $\boldsymbol{p}(\infty)$, which reads with the electron's charge q and the time-dependent electric field $\boldsymbol{E}(t)$ (applying the dipole approximation and neglecting Coulomb corrections)

$$\boldsymbol{p}(\infty) = \boldsymbol{p}(t_0 + \tau_A) + q \int_{t_0+\tau_A}^{\infty} \boldsymbol{E}(t')dt'. \tag{1}$$

In attoclock experiments, electrons are ionized by elliptically polarized light, which makes the direction of the asymptotic momentum very sensitive to the ionization time $t_0 + \tau_A$. In the extraction of τ_A from attoclock experiments, it is state of the art to treat the ionized electron classically, to take into account Coulomb corrections, and to assume that the electron's initial momentum follows from some semiclassical theory [18,19]. For a reliable reconstruction of the attoclock time τ_A, however, suitable initial conditions have to be identified as pointed out in Refs. [20,21]. In particular, assumptions about the initial momentum bias the reconstructed value for τ_A.

The popular two-step model of tunnel ionization assumes a maximal ionization rate at the instant of maximal electric field strength, i.e., $\tau_A = 0$, and that free electrons have zero initial momentum, i.e., $\boldsymbol{p}(t_0 + \tau_A) = 0$. Within the two-step model, the electron's asymptotic momentum follows by solving the classical equations of motion for the electron's motion in the combined electromagnetic field of the binding potential and the ionizing external light pulse. This model, however, cannot be used as a benchmark for experiments, because a possible match or mismatch of experimental data and the theoretical prediction by the two-step model may be explained by various pairs of nonzero delay τ_A [22] and nonzero initial momentum $\boldsymbol{p}(t_0 + \tau_A)$,

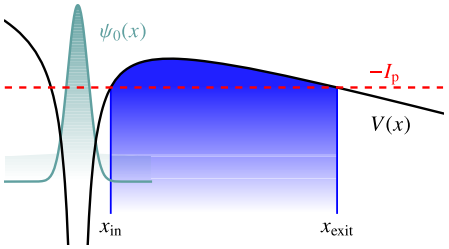

FIG. 1. The one-dimensional Coulomb potential, which is bent by an additional electric field, defines the effective potential $V(x)$ and the tunneling barrier between the points x_{in} and x_{exit} for an electron, which is given by the wave function $\psi_0(x)$, with the ground state energy $-I_p$.

which may a have nonzero component parallel to the electric field direction [23,24].

Neither the delay τ_A nor the initial momentum $\boldsymbol{p}(t_0+\tau_A)$ are directly accessible by experiments, and it is also challenging to calculate them analytically. Therefore, we employ *ab initio* quantum calculations and a virtual detector [25,26] at the tunnel exit. The virtual detector technique allows us to determine directly the electron's time of arrival at the tunnel exit as well as its exit momentum.

We analyze theoretically an initially bound electron ionized by an electric field pulse. To determine the time delay τ_A and the exit momentum, we solve the time-dependent Schrödinger equation and place a virtual detector at the tunneling exit. The virtual detector is realized by calculating the probability current at the exit. The most probable time delay τ_A is determined by comparing the instant of maximum probability current at the potential barrier exit and the instant of maximum electric field strength. The exit momentum is determined by the space-resolved momentum distribution at the tunnel exit at the instant of ionization. By separating the wave function into a tunneled part and a bound part after the interaction with the laser pulse, we can calculate the momentum distribution of the tunneled electron, from which one can determine the most probable asymptotic momentum.

Considered system.—In experiments, a Coulomb-bound electron is usually excited by a laser pulse with a wavelength much bigger than the atomic dimensions such that the laser pulse is nearly homogeneous over the size of the atom. Furthermore, relativistic effects and effects due to the magnetic field component set in only for tunneling from highly charged ions [27]. Thus, we will apply the electric dipole approximation. The laser pulse is modeled by a time-varying homogeneous electric field $E(t) = E_0 \exp[-\omega^2(t-t_0)^2/2]$, where t_0 denotes the instant of the maximum field strength E_0 and $\tau_E = \sqrt{2}/\omega$ is the time scale of the rise and decay of the electric field. The linear polarization of the electric field and neglecting the magnetic field component render the motion of the electron quasi-one-dimensional, allowing us to investigate general features of tunneling times in a one-dimensional scenario. Furthermore, tunneling in the fully three-dimensional Coulomb problem can be described by an effective one-dimensional tunneling barrier via introducing parabolic coordinates [28]. Thus, we restrict ourselves to one-dimensional systems and consider an electron bound to the soft-core potential $-Z/\sqrt{x^2+\alpha(Z)}$ [29–32] to model the essential features of an electron in a three-dimensional Coulomb potential. Here, Z is the atomic number, and the softening parameter $\alpha(Z) = 2/Z^2$ is chosen such that the ground state energy of the soft-core potential is $-I_p = -Z^2/2$, which equals the ground state energy of the Coulomb potential [33]. Thus, the Schrödinger equation (in atomic units)

$$i\frac{\partial\psi}{\partial t} = \hat{H}(t)\psi = \left(-\frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{Z}{\sqrt{x^2+\alpha(Z)}} - E(t)x\right)\psi \qquad (2)$$

with the Hamiltonian $\hat{H}(t)$ and the effective potential $V(x,t) = -Z/\sqrt{x^2+\alpha(Z)} - E(t)x$ will be solved numerically [34]. The so-called Keldysh parameter $\gamma = \omega\sqrt{2I_p}/E_0$ [12] characterizes the ionization process as dominated by tunneling for $\gamma \ll 1$ and by multiphoton ionization for $\gamma \gg 1$. Thus, simulation parameters will be set such that $\gamma < 1$ in the following.

Time delay τ_A.—The time delay τ_A is based on the time-dependent ionization rate. In our one-dimensional model, the probability current $j(x,t) = [\psi(x,t)^*\partial_x\psi(x,t) - \psi(x,t)\partial_x\psi(x,t)^*]/(2i)$ represents the average net number of electrons passing a given point at a specific time. Thus, we determine the ionization rate via the probability current at the exit x_{exit} as a function of time, where x_{exit} is defined by the maximum electric field strength E_0 via $V(x_{\text{exit}}, t_0) = -I_p$. Monitoring the probability current at a fixed position is justified, because the tunnel probability is maximal for $E(t) = E_0$ and it is exponentially suppressed for lower electric fields. Furthermore, $E(t) = E_0[1 - \Delta t^2/\tau_E^2 + O(\Delta t^4/\tau_E^4)]$ for $\Delta t = t - t_0$ with $|\Delta t| < \tau_E = 2\sqrt{I_P}/(\gamma E_0)$. Thus the tunneling barrier does not change substantially if times close to t_0 are considered. To further ensure that the calculated exit x_{exit} is where the electron exits the barrier, we place a virtual detector at different points between x_{in} and x_{exit} and calculate $j(x,t)$ at each point as a function of time. As shown in Fig. 2, the probability current has a positive peak as well as a negative one for $x < x_{\text{exit}}$, indicating the tunneling and reflection dynamics, respectively. This tunneling and reflection dynamics corresponds to under-the-barrier dynamics. As reflection is absent for $x \geq x_{\text{exit}}$, the particle leaves the barrier at x_{exit}.

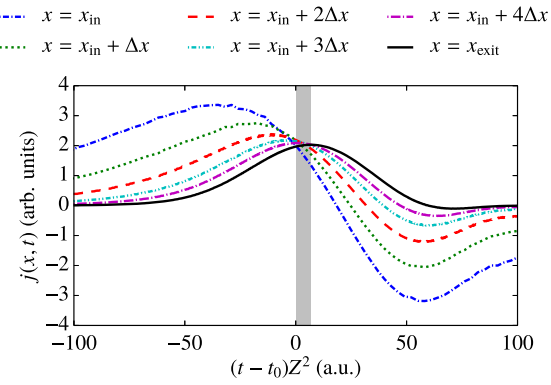

FIG. 2. The probability current $j(x,t)$ as a function of time at different positions x between the barrier entry $x = x_{\text{in}}$ and the barrier exit x_{exit} separated by $\Delta x = (x_{\text{exit}} - x_{\text{in}})/5$ for the parameters $E_0/Z^3 = 0.048$ and $\gamma = 0.25$. The shadowed area represents the time between the instant of maximum electric field strength t_0 and the instant of maximum probability at x_{exit}.

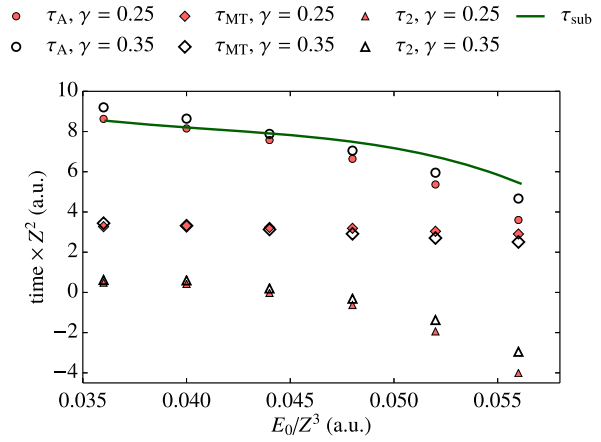

FIG. 3. The time delay τ_A, the Mandelstam-Tamm time τ_{MT}, and the times τ_2 and τ_{sub} plotted for different electric field strengths E_0 and for different Keldysh parameters γ. Definitions of the various times are given in the main text.

The most probable time delay τ_A is calculated by subtracting the instant of the maximum field strength from the instant of the maximum current, i.e., $\tau_A = \mathrm{argmax} j(x_{\mathrm{exit}}, t) - \mathrm{argmax} E(t)$, which yields the positive time delay shown in Fig. 3. Note that, for the parameters used in Fig. 3, $\tau_A < \tau_E$, and thus the electric field remains almost constant for times $|\Delta t| \lessapprox \tau_A$, justifying our choice for x_{exit}.

The origin of the time delay τ_A can be understood by considering the time-energy uncertainty principle and following its interpretation given by Mandelstam and Tamm [14,35–37]. As a consequence of the time-energy uncertainty principle, the time, which a wave function ψ of a system with a time-independent Hamiltonian $\hat{H}$ needs to change significantly, is bounded from below by $1/(2\sigma_{\hat{H}})$, where $\sigma_{\hat{H}} = \sqrt{\langle\psi|\hat{H}^2|\psi\rangle - \langle\psi|\hat{H}|\psi\rangle^2}$. As outlined in the previous section, the Hamiltonian in (2) can be considered as time independent for times $|\Delta t| < \tau_E$. This allows us to define the Mandelstam-Tamm time

$$\tau_{\mathrm{MT}} = \frac{1}{2}(\langle\psi(t_0)|\hat{H}(t_0)^2|\psi(t_0)\rangle - \langle\psi(t_0)|\hat{H}(t_0)|\psi(t_0)\rangle^2)^{-1/2}, \tag{3}$$

which is indeed a lower bound to the time delay τ_A as indicated in Fig. 3. The time delay τ_A is close to its lower bound τ_{MT}, which indicates that the delay τ_A is a consequence of the wave function's inertia, i.e., its inability to adopt instantaneously to the field. The time delay τ_A decreases as E_0 increases at fixed Keldysh parameter γ and matches approximately τ_{MT} when the regime of over-the-barrier ionization is approached, which is for $E_0/Z^3 \approx 0.06$ a.u.

The observed decrease of the delay τ_A with growing electric field strength (but constant γ) is consistent with calculations for the case of a sudden turn-on of the electric field, which show that the time for the wave function to adopt to the electric field is proportional to the Keldysh time $\tau_K = \sqrt{2I_p}/E_0$ [14,37], although the functional dependence on E_0 is different for continuously changing electric fields. For a fixed maximum electric field strength E_0 but increasing γ, the time delay τ_A increases. As γ increases, the pulse duration decreases, granting the wave function less time $\sim\tau_E$ to evolve and to adopt to the time-varying Hamiltonian during the rise of the electric field. Thus, the wave function has less time to develop the necessary components for tunneling and thus needs more time to reach the maximum ionization rate.

The times τ_A and τ_{MT} are intractable by analytical methods. As we will demonstrate in the following, however, one may employ the Wigner time as an approximation for τ_A. Comparing the quantum mechanical Wigner trajectory $t_{\mathrm{W}}(x)$ [24,27] to the trajectory $t_c(x)$ of a classical particle, which travels instantaneously from x_{in} to x_{exit} and then moves according to the classical equations of motion, we can calculate the time delay

$$\tau_{\mathrm{sub}} = t_W(x_{\mathrm{exit}}) - t_c(x_{\mathrm{exit}}), \tag{4}$$

where the Wigner and the classical trajectories $t_W(x)$ and $t_c(x)$ are determined such that both coincide at the entry point x_{in}.

The delay τ_A describes the time interval that the electron spends under the barrier *after the field maximum*. Interpreting τ_A as a tunneling time [18] is justified only if the classical forbidden region is entered at the field maximum. The Wigner delay (4), however, represents the total time spent under the barrier, since the relation $t_c(x_{\mathrm{in}}) = t_c(x_{\mathrm{exit}})$ holds in the classical model. Also, the time delay τ_A is often understood as the time under the barrier. Our numerical results show that τ_{sub} is indeed close to the time delay τ_A, as shown in Fig. 3, where τ_{sub} is calculated for the one-dimensional Coulomb potential. Although τ_{sub} and τ_A agree well, it is open if τ_A should be interpreted as time spent under the barrier, because the interpretation as under-the-barrier time is based on the assumption that tunneling starts at the instant of the maximal electric field, but the wave function could penetrate the tunneling barrier earlier.

For the one-dimensional Coulomb potential and at field intensities approaching the over-the-barrier threshold, the Wigner time is given by $\tau_{\mathrm{sub}} \approx 14.29\sqrt{1 - 16E_0/Z^3}/Z^2$, and by $\tau_{\mathrm{sub}} \approx 9.0\sqrt{1 - 9.5E_0/Z^3}/Z^2$ for the three-dimensional case. Furthermore, the Wigner formalism can be extended [38] to include the Stark shift and also multielectron effects. The resulting Wigner time τ_{sub} for helium is shown exemplarily in Fig. 4 as a function of the laser's peak intensity.

Exit momentum.—In the following, we determine the quantum mechanical exit momentum p_0 in the direction of

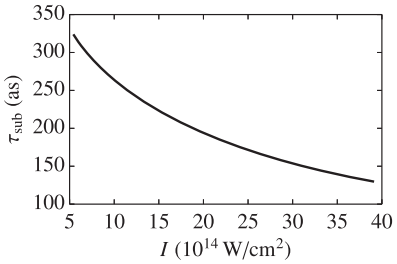

FIG. 4. Wigner time τ_{sub}, which is an estimate for τ_A, for the three-dimensional Coulomb potential including Stark shift and multielectron effects as a function of the electromagnetic field's peak intensity I for ionization of helium atoms by linearly polarized laser fields. The applied wavelength is $\lambda = 800$ nm, and the Keldysh parameter varies from 0.6 to 0.2.

the electric field at the tunnel exit x_{exit} at the instant of ionization $t = t_0 + \tau_A$ by two different methods. In the first method, we calculate a space-resolved momentum distribution around $x = x_{exit}$ by multiplying the wave function $\psi(x, t_0 + \tau_A)$ by a Gaussian window function with mean x_{exit} and width $\delta x = (x_{exit} - x_{in})/20$ and calculating its Fourier transform $\tilde{\psi}_{exit}(t_0 + \tau_A)$. The most probable exit momentum p_0 can be inferred by the momentum where $|\tilde{\psi}_{exit}(t_0 + \tau_A)|^2$ is maximal. In the second method, the most probable initial momentum p_0 is determined from the probability current $j(x_{exit}, t_0 + \tau_A)$ at the exit at the instant of the maximum probability current. Following Refs. [25,26], the local velocity of the wave function's probability flow at x_{exit} equals $v = j(x_{exit}, t_0 + \tau_A)/|\psi(x_{exit}, t_0 + \tau_A)|^2$. Both methods yield very similar results for the moment p_0 as shown in Fig. 5. The initial momentum p_0 is almost independent of the parameter γ and depends only weakly on the electric field strength E_0.

As p_0 does not depend on the parameters of the external electric field, it must result from the initial quantum state, i.e., the ground state of the binding potential. In fact, the ground state of the employed soft-core potential has in

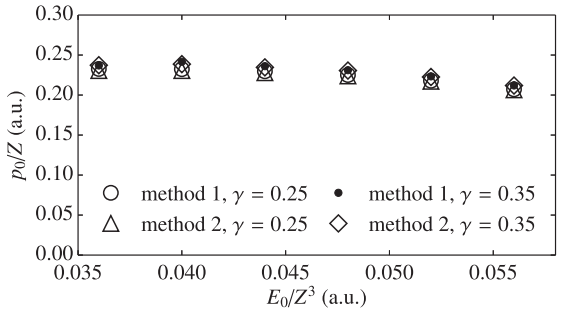

FIG. 5. The momentum p_0 at the time $t_0 + \tau_A$ at the tunnel exit x_{exit} for different electric field strengths E_0 and different Keldysh parameters γ as determined by two different methods. Method 1 is based on the space-resolved momentum distribution, while method 2 utilizes the velocity of the probability flow; see the main text for details on both methods.

momentum space a width of about $0.38 \times Z$, which is of the same order as p_0; see Fig. 5. The fact that p_0 scales with the width of the ground state's momentum distribution and not with its mean (which is zero) may be interpreted as momentum components, which propagate into the ionization direction, being ionized preferably.

Implications.—The above considerations are relevant for every high-precision laser-induced tunneling experiment. Here we discuss as an example attoclock measurements due to their currently high attention. Attoclock experiments aim to determine the time delay τ_A between the instant of the electric field maximum and the instant of tunneling, which is not directly accessible experimentally. Instead, the asymptotic momentum of the tunneled electron is measured. As it depends on the exit momentum and on the moment at which the electron starts to propagate freely in the field, one can infer τ_A from the electron's asymptotic momentum provided that the exit momentum is known. The delay τ_A is commonly reconstructed by assuming zero initial momentum in the electric field direction. Our numerical simulations and the virtual detector approach, however, indicated a nonzero initial momentum in the direction of the electric field. How does the zero-initial-momentum assumption affect the reconstruction of τ_A from the asymptotic momentum?

To answer this, we determine the final momentum of the tunneled electron by propagating the wave function $\psi(t)$ till some final time t_f such that $t_f - t_0 \gg 1/\omega$ and separate the tunneled part $\psi_{free}(t_f)$ from the bound part of the wave function by projecting out all bound eigenstates of $\hat{H}(t)$ in Eq. (2) for $E(t) = 0$ from $\psi(t_f)$. From the resulting probability density in momentum space representation $\tilde{\psi}_{free}(t_f)$, the most probable momentum $p_{f,q}$ can be inferred by the position where $|\tilde{\psi}_{free}(t_f)|^2$ is maximal. Using the Newton equations of motion for an electron in the effective potential $V(x, t)$, we can calculate at which instant of time the electron must exit the barrier at x_{exit} with zero initial momentum in the electric field direction such that its asymptotic momentum equals $p_{f,q}$. The result is the time delay τ_2, which is also shown in Fig. 3 and that does *not* coincide with the delay τ_A. The delay τ_2 is close to zero or even negative depending on the electric field strength as found in Refs. [20–22,24,27]. Consequently, the delay τ_A and the instant of tunneling cannot be determined on the basis of the standard assumption of zero initial momentum in the electric field direction. The exit momentum has to be included.

Similarly to the delay τ_2, also the time delays determined from measured asymptotic momenta are close to zero (of the order of experimental uncertainties) [10,11]. As our numerical calculations indicate that the reconstruction of the delay τ_A from asymptotic momenta is sensitive to the electron's exit momentum, we argue that a possible nonzero exit momentum into the electric field direction has to be included for a reliable reconstruction of τ_A in attoclock

experiments, not only a nonzero exit momentum in the direction perpendicular to the electric field as recently proposed [20,39].

Conclusions.—We reexamined tunneling times in strong field ionization by an *ab initio* solution of the time-dependent Schrödinger equation. Our calculations show that there is a delay τ_A between the maximum of the ionization rate and the maximum of the electric field strength and a nonzero exit momentum p_0 in the electric field direction. The delay τ_A can be explained as the response time needed by the wave function to react to the change of the driving electric field. The initial momentum can be estimated from the width of the ground state's momentum distribution, which is Z for the three-dimensional Coulomb potential. The time τ_A may be estimated by the Wigner time τ_{sub}. Note that our results differ from the vanishing tunneling delay as obtained by the recently introduced analytical R-matrix method [21], which is based on the assumption that this quasiclassical model provides a good description of the quantum-mechanical tunneling dynamics also in the vicinity of the tunneling barrier.

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