Under-the-Tunneling-Barrier Recollisions in Strong-Field Ionization

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A new pathway of strong-laser-field-induced ionization of an atom is identified which is based on recollisions under the tunneling barrier. With an amended strong-field approximation, the interference of the direct and the under-the-barrier recolliding quantum orbits are shown to induce a measurable shift of the peak of the photoelectron momentum distribution. The scaling of the momentum shift is derived relating the momentum shift to the tunneling delay time according to the Wigner concept. This allows us to extend the Wigner concept for the quasistatic tunneling time delay into the nonadiabatic domain. The obtained corrections to photoelectron momentum distributions are also relevant for state-of-the-art accuracy of strong-field photoelectron spectrograms in general.

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Modern strong-field photoelectron spectroscopy has achieved unprecedented momentum resolution of the order of 0.01 atomics units (a.u.), see, e.g.. Refs. [1–3], due to advancement of the measurement technique with a reaction microscope [4]. Recently, the attoclock technique has been developed [5,6] based on the strong-field ionization of an atom in an elliptically polarized laser field, which attempts to map the photoelectron momentum at the detector into the time of the electron appearance in the continuum during strong-field ionization. In this way, the attoclock technique is assumed to extract information on the time-resolved dynamics of the electron released from the atomic bound state during strong-field ionization, and in particular, on the time-delay of the tunneling electron wave packet from the atom in a strong laser field [5–10]. Furthermore, the interference structures in the high-resolution photoelectron momentum distribution (PMD), created by the direct and recolliding trajectories, allow an interpretation as time-resolved holographic imaging of atoms and molecules, which admits attosecond time resolution and angstrom spatial resolution [11–14]. For a correct interpretation of imaging results of the PMD based attoscience applications, one needs to understand theoretically all PMD features in detail.

There are many theoretical approaches for the treatment of the tunneling delay time [15–17], leading to different solutions and to a debate on how to explain the photoelectron momentum distribution in attoclock experiments [17–19]. Although all alternative definitions of the tunneling delay time are equally valid theoretical concepts, the Wigner concept [20] is physically relevant to the measurement of the photoelectron momentum distribution in the attoclock setup in the quasistatic regime, as proven in a recent experiment [10]. However, the Wigner definition of the time delay via the derivative of the wave function phase, and its generalization for the strong-field tunneling problem [18,21–24] is applicable only in the quasistatic limit, i.e., when the laser-induced barrier is (quasi)static. Therefore, there is need for a generalization of the Wigner concept to the nonadiabatic regimes [25–27] of the strong-field ionization, which may explain the discrepancy between the theory and the attoclock experiment at large Keldysh parameters [10].

The main workhorse for the theoretical treatment of the strong-field ionization, the strong-field approximation (SFA) [28–30], in its common form does not provide a signature of the tunneling time in the asymptotic momentum distribution. The same is true for the Coulomb corrected SFA (CCSFA) [31,32], and the analytic R-matrix (ARM) theory [33–35], which include the Coulomb field of the atomic core for the continuum electron in the eikonal approximation [that is, in the Wentzel-Kramers-Brillouin (WKB) approximation combined with the perturbative accounting of the Coulomb field in the phase of the wave function]. Meanwhile ARM theory provides very good agreement with the time-dependent Schrödinger-equation simulations of PMDs for a hydrogen atom in the deep-tunneling regime [18,24], including the asymptotic momentum shifts (these shifts are not linked to delays during electron tunneling). To describe the Wigner tunneling time delay (emerging from the derivative of the phase of the wave function) within the SFA, one needs to account for the phase of the wave function during the under-the barrier dynamics, which is vanishing in the leading order of WKB approximation, but becomes non-negligible for sufficiently thin barriers, i.e., in the near-threshold tunneling regime [22]. For the sake of intuitive understanding within an analytical treatment, this conceptual problem is most easily addressed in the case of a short-range potential. It is well known that the qualitative description of many strong-field phenomena, such as above-threshold ionization [36,37], high-order harmonic generation [38,39], or nonsequential double ionization [40], have been successfully given first in a simplified approach using a short-range potential.

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In this Letter, we have modified the common SFA in the case of a short-range atomic potential, revealing and employing new quantum orbits for the ionizing electron, which describe rescattering of the electron at the atomic core during the under-the-barrier dynamics, see Fig. 1. We demonstrate that the interference of the direct and the under-the-barrier rescattering trajectories induces a phase shift of the wave function of the tunneling electron and a measurable shift of the peak of the momentum distribution. In the quasistatic regime the scaling of the momentum shift with respect to the laser and atom parameters is in accordance with the Wigner time delay theory, which allow us to interpret it accordingly. Moreover, the modified SFA provides a route for treating the Wigner time delay in nonadiabatic regimes of strong-field ionization.

We consider the ionization of an atom in a laser field of linear and elliptical polarization in the case of a short-range binding potential in the nonrelativistic regime. The Keldysh-parameter $\gamma \equiv \kappa\omega/E_0$ is not restricted, with $\kappa = \sqrt{2I_p}$, the ionization potential I_p, the laser field amplitude E_0 and frequency ω, describing the tunneling, the multiphoton, as well as the transition regimes. The field strength parameter $f \equiv E_0/\kappa^3$ is assumed to be small to avoid over-the-barrier ionization, and atomic units are used throughout. Having simplified the scenario to the basic physical process, we are able to calculate the photoelectron momentum distribution $w(\mathbf{p}) = |M(\mathbf{p})|^2$ analytically via a second order SFA amplitude [41]. For an improvement of the recollision treatment, the low frequency approximation [42,43] is employed, replacing the recollision matrix element in the Born approximation by the exact T matrix:

$$\begin{aligned} M(\mathbf{p}) &= M_0(\mathbf{p}) + M_1(\mathbf{p}) \\ &= -i\int dt \langle\psi_{\mathbf{p}}(t)|H_i(t)|\phi(t)\rangle \\ &\quad - \int dt' \int^{t'} dt'' \int d^3\mathbf{q} \langle\psi_{\mathbf{p}}(t')|T|\psi_{\mathbf{q}}(t')\rangle \\ &\quad \times \langle\psi_{\mathbf{q}}(t'')|H_i(t'')|\phi(t'')\rangle, \end{aligned} \tag{1}$$

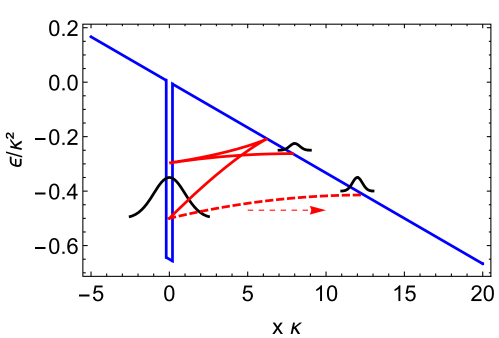

FIG. 1. Schematic picture of laser-induced tunneling ionization: (dashed line) the direct trajectory, and (solid line) the under-the-barrier recolliding trajectory.The Keldysh parameter is $\gamma = 1$, featuring nonadiabatic tunneling, i.e., when the energy is not constant during tunneling.

where M_0, M_1 are the direct and rescattering amplitudes, $|\psi_{\mathbf{p}}(t)\rangle = |\mathbf{p} + \mathbf{A}(t)\rangle \exp[iS_{\mathbf{p}}(t)]/\sqrt{2\pi}^3$ is the Volkov state in length gauge with the asymptotic momentum $\mathbf{p}$ and contracted action $S_{\mathbf{p}}(t) = \int_t^\infty ds[\mathbf{p} + \mathbf{A}(s)]^2/2$, $H_i(t) = -\mathbf{r}\cdot\mathbf{F}(t)$ the interaction Hamiltonian with the laser-field-induced force $\mathbf{F}(t) = E_0\mathbf{e}_x\cos(\omega t)$, $\partial_t\mathbf{A} = \mathbf{F}$, $\langle\mathbf{p}|H_i(t)|\phi\rangle$ the matrix element of the transition from the bound state into the continuum, $|\phi\rangle$ the initial bound state, and $\langle\mathbf{p}|T|\mathbf{q}\rangle$ the scattering T-matrix element.

First, we illustrate our theoretical approach in the 1D case and a laser field of linear polarization, and further extend the discussion to 3D and elliptical polarization. Thus, we begin considering the single active electron to be initially in its bound state in a 1D δ-potential $V(x) = -\kappa\delta(x)$, with the wave function of the bound state $\langle x|\phi(t)\rangle = \sqrt{\kappa}\exp(-\kappa|x| + i\kappa^2/2t)$ [44]. In 1D, $H_i = -xF(t)$ is $\langle p|H_i|\phi\rangle = 2\sqrt{2}ipF(t)/[\sqrt{\pi}(p^2+\kappa^2)^2]$, and the exact scattering T matrix is $\langle p|T|q\rangle = -(\kappa/2\pi)[\sqrt{p^2}/(\sqrt{p^2} - i\kappa)]$. The momentum amplitude of Eq. (1) in 1D case ($d^3\mathbf{q} \to dq$) has two terms, the 1D integral for the direct electron, and 3D integral for the rescattered electron. In the latter, rather than considering the rescattering during the continuum excursion, contribution of which is well investigated and takes place during at least two laser half cycles, we consider only rescattering during the under-the-barrier motion which appears already in one laser half cycle. Ionization induced by a half cycle is considered, circumventing interference effects from the ionization from neighboring half cycles.

For the physical interpretation of the recollision picture, we first apply the simultaneous 3D saddle-point integration analytically in the quasistatic case $\gamma \ll 1$, when the saddle point equations read $q_s = -E_0(t_r + t_i)/2$, which defines the intermediate momentum q_s via the return condition of the trajectory, $(p + E_0t_r)^2/2 = (q_s + E_0t_r)^2/2$, the recollision time t_r via the energy conservation at recollision, $(q_s + E_0t_i)^2/2 = -I_p$, and the ionization time t_i via the energy conservation at ionization. The saddle point equations yield the following physical solution: $t_r = (-p + i\kappa)/E_0$ and $t_i = (-p + 3i\kappa)/E_0$ (other solutions yield unphysical trajectories with increasing probabilities during propagation). Simplifying further for a moment with $p = 0$, one obtains $t_i = 3i\kappa/E_0$, $t_r = i\kappa/E_0$, and $q_s + A(t_i) = i\kappa$, accordingly $q_s + A(t_r) = -i\kappa$ and $p + A(t_r) = i\kappa$. The latter provides the trajectory of the recolliding electron up to the recollision point: $x(t) = i\kappa(t - t_i) + E_0(t - t_i)^2/2$. The trajectory starts at time t_i at the atomic core $x(t_i) = 0$, moves along the electric field through the barrier to the tunneling exit $x_e = I_p/E_0$, reaching it at $t = 2i\kappa/E_0$. Afterwards the electron is reflected and turns around, tunnels back to the core, where it recollides off the core $x = 0$ at t_r, and again tunnels to the exit, leaving the barrier at $t_e = 0$. In Fig. 1, the trajectory is visualized in the nonadiabatic regime at $\gamma = 1$, with numerical solution of the saddle-point equations, showing the electron energy gain during ionization. The accurate quantitative evaluation of the ionization amplitudes is carried out numerically [45]. The result for PMD is shown in Fig. 2(a). Whereas the direct $|M_0(p)|^2$ and the recolliding $|M_1(p)|^2$ PMDs are peaked at zero momentum, the coherent sum of the two distributions $|M_0(p) + M_1(p)|^2$ is slightly shifted towards positive momenta, i.e., the interference of the direct and the recolliding trajectories gives rise to a momentum shift δp of the PMD peak.

The behavior of the discussed momentum shift in the quasistatic and the nonadiabatic regimes is illustrated in Figs. 2(b) and 2(c), respectively (the momentum shift δp is equivalent to the time delay at the detector $\delta t = -\delta p/E_0$ and is positive and corresponds to the asymptotic negative time delay, see also Refs. [18,46]). In the quasistatic regime $\gamma \ll 1$ a significant tunneling time delay occurs when the field strength exceeds approximately 0.1 a.u. indicating that it is connected with near-threshold tunneling. For smaller field strengths the recolliding path is strongly suppressed and does not affect the momentum distribution. However, it is also possible to have a significant momentum shift for relatively small field strengths as long as the Keldysh parameter is large, see Fig. 2(c). The reason is that the electron gains energy during the tunneling process and can enter this way to the near-threshold strong-field ionization regime even for a small laser electric field strength. Additionally, we display in Fig. 2(d) the tunneling time delay vs the field strength for a fixed laser frequency in the nonadiabatic regime. When the frequency is fixed, a significant tunneling time delay occurs at large and small field strengths, where the latter can be associated with a large Keldysh parameter.

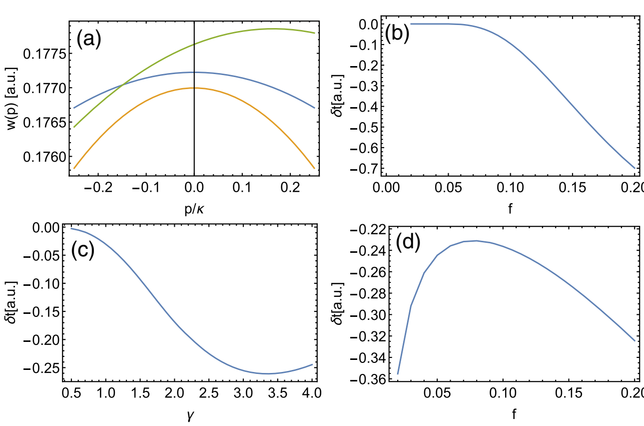

FIG. 2. Ionization of a 1D atomic system: (a) PMD for $f = 0.2$ and $\gamma = 0.2$, (blue) via the direct amplitude, (brown) via the recolliding amplitude, scaled by a factor of 434, and (green) via including the interference of the direct and recolliding trajectories. (b) Tunneling time delay vs the field strength f in the adiabatic regime of $\gamma = 0.2$, (c) vs the Keldysh-parameter γ in the nonadiabatic regime for $f = 0.05$, and (d) vs f in the non-adiabatic regime for $\omega = 0.2$ a.u..

Let us estimate the scaling of the momentum shift due to interference of the direct and under-the barrier trajectories in the quasistatic regime. Using the equivalence of the ionization matrix element $\langle p|H_i|\phi\rangle$ with $\langle p|V|\phi\rangle$ [41], the amplitudes of the direct electrons can be estimated as $M_0 \sim -i\delta t_i V_i \exp[-\kappa^3/(3E_0)]$ with the typical size of the volume element $\delta t_i \sim 1/\sqrt{\partial_t^2 S} \sim 1/\sqrt{\kappa E_0}$, and $V_i = \langle p|V|\phi\rangle \sim -\kappa^{3/2}$, which yields: $M_0 \sim i\exp[-\kappa^3/(3E_0)]/\sqrt{f}$.

The amplitude of the rescattered electrons can be estimated alike $M_1 \sim -\delta t_1 \delta t_2 \delta q V_i \langle i\kappa|T| - i\kappa\rangle \exp(-\kappa^3/E_0)/2$. The size of the volume element is $\delta t_1 \delta t_2 \delta q \sim 1/\sqrt{E_0 \kappa^3}$, which is estimated from the determinant of the matrix of the second order derivatives in the static regime, with $\partial_{qq} S = -i(t_r - t_i)$, $\partial_{qt_1} S = -iE_0(t_r - t_i)/2$, $\partial_{qt_2} S = -iE_0(t_r - t_i)/2$, $\partial_{t_1 t_1} S = iE_0^2(t_r + t_i)/2$, $\partial_{t_2 t_1} S = 0$, $\partial_{t_2 t_2} S = -iE_0^2(t_r - t_i)/2$. Further, we estimate $\langle i\kappa|T| - i\kappa\rangle \sim \kappa/\sqrt{f}$, with the typical size of the recollision momentum $p + A(t_r) \sim i(\kappa - \sqrt{E_0/\kappa})$ [47]. Note that the recollision amplitude via the T matrix is increased by a factor of $1/\sqrt{f}$, compared with the standard description in the Born approximation, due to the singularity of the T matrix at the recollision energy of $-\kappa^2/2$. Thus, the rescattering amplitude is estimated to be $M_1 \sim -\exp(-\kappa^3/E_0)/f$.

Applying the quasistatic approximation, i.e., replacing E_0 by the instantaneous electric field, and taking into account the time to momentum mapping, $E_0 \to F(p) = E_0\sqrt{1 - [\omega(-p + i\kappa)/E_0]^2}$ [48], we obtain

$$|M(p)|^2 \sim \frac{|i \exp\left(-\frac{\kappa^3}{3F(p)}\right) - \frac{1}{\sqrt{f}} \exp\left(-\frac{\kappa^3}{F(p)}\right)|^2}{f}. \quad (2)$$

The latter has a maximum at $\delta p^{(1D)} \sim (M_1/M_0)\kappa \sim \exp[-2/(3f)]\kappa/\sqrt{f}$, demonstrating the PMD shift. The amplitude of the recolliding electrons is smaller by a factor of $\exp[-2/(3f)]$ due to the three times longer tunneling distance. In fact, an estimation of the tunneling amplitude via the WKB tunneling exponent $S = \int p dx$ along the recolliding trajectory yields $S = -\kappa^3/E_0$. Note that the replacement of the recollision matrix element in the Born approximation by the exact T matrix is necessary, because $p = i\kappa$ for the considered under-the-barrier recollision, while the Born approximation requires $p \gg \kappa$. Thus, the amplitude of the additional recolliding path is rather small, $M_0/M_1 \approx 20$ in 1D, as Fig. 2(a) shows. Nevertheless, the momentum shift due to its interference with the direct trajectory is not negligible as $\delta p/\kappa \sim 0.05$. The momentum distribution in 3D is qualitatively the same as in the 1D case, however, the observed momentum shift is smaller by a factor of f: $\delta p^{(3D)} \sim \exp[-2/(3f)]\kappa\sqrt{f}$ [45].

Up to now we have discussed the case of a linearly polarized laser field. However, the experimental observation of the discussed momentum shift will require the attoclock setup, i.e., an elliptically polarized laser field close to circular, to avoid masking the effect by low-energy structures [1–3]. We have calculated PMD in an elliptically polarized laser field with a vector potential $\mathbf{A}(t) = E_0/\omega[\sin(\omega t)\mathbf{e}_x + e\cos(\omega t)\mathbf{e}_y]$, with $E_0 = 0.2$ a.u., $\omega = 0.057$ a.u., and ellipticity $e = 0.8$, including all relevant saddle points from two consecutive laser cycles, see Fig. 3, also Ref. [45]. The potential of the atomic core is modeled by a 3D-short-range potential $V = 2\pi/\kappa\delta(\mathbf{r})\partial_r r$ with $\kappa = 1$ a.u. ($\gamma \approx 0.3$ and $f = 0.2$). Here there exists only one saddle point per cycle describing direct electrons, where the corresponding coherent superposition leads to a photon interference structure along the radius of the PMD with an energy spacing of ω. We find in each cycle an additional saddle point describing the under-the-barrier recollision. When the latter are taken into account, the peaks of the PMD rings are rotationally shifted to positive angles, see Fig. 3. The angle of the maximum of the PMD indicated by a cross in Fig. 3(a), is at $\theta_{\max} \approx 3.4°$, which is consistent with the calculation of the time delay in a linearly polarized field, since $t = e(\pi/180°)\theta_{\max}/\omega \approx 0.8$ a.u. This is intuitively explainable, because there is no significant variation of the tunneling barrier and of the under-the-barrier recollisions in the quasistatic regime for both the linear and circular polarization cases. Thus, the under-the-barrier recollisions exist in an elliptically polarized laser field, close to circular. The interference of the direct and under-the-barrier rescattered trajectories induces a rotational shift of the peak of PMD, which is not masked by the photon interference rings in the radial direction. The magnitude of the rotational shift is reduced by a factor of $\sim f$, when the spreading of the electron wave packet during the under-the-barrier motion is accounted for, see Fig, 3(b).

The described momentum shift due to interference of the direct and the under-the-barrier rescattered trajectories is closely related to the Wigner tunneling time delay. To demonstrate this, we recall the Wigner formalism which accounts for the tunneling delay time during the laser-driven

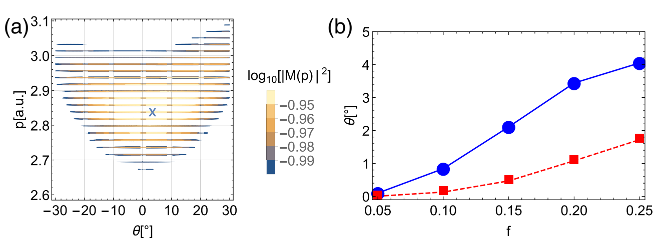

FIG. 3. Ionization in an elliptically polarized laser field: (a) PMD at $\gamma \approx 0.3$ and $f = 0.2$, with $\theta = \arctan(p_x/p_y)$ and $p = \sqrt{p_x^2 + p_y^2}$, spreading during tunneling is neglected. The cross indicates the maximum of the PMD. (b) The attoclock angle shift θ vs the field strength f with (red squares) and without (blue circles) accounting for electron wave packet spreading during the tunneling step.

ionization process from a short-range atomic potential. The time delay in the Wigner formalism is calculated as a derivative of the phase of the wave function. The continuum wave function in a slowly varying laser field (approximated by a constant electric field E_0), which has outgoing current and is matched with the bound state ϕ at the matching coordinate under the barrier $x = x_m$ [45], reads $\psi(x) = \mathcal{T}[\mathrm{Ai}(\Xi) - i\mathrm{Bi}(\Xi)]$, with the transition coefficient $\mathcal{T} = \phi(x_m)/[\mathrm{Ai}(\zeta) - i\mathrm{Bi}(\zeta)]$, $\zeta \equiv \sqrt[3]{2}(I_p - x_m E_0)/E_0^{2/3}$, and $\Xi \equiv \sqrt[3]{2}(I_p - E_0 x)/E_0^{2/3}$). The Wigner time delay is calculated as [22]

$$\delta t = \mathrm{Re}\left\{-i\frac{\partial}{\partial I_p}\{\ln[\psi(x)] - \ln[\psi^{qc}(x)]\}\right\}\Bigg|_{x\to\infty}, \quad (3)$$

where $\psi^{\mathrm{qc}}(x)$ is the quasiclassical wave function, i.e., $\psi(x)$ at the limit $E_0 \ll \kappa^3$, and the related momentum shift $\delta p = -E_0\delta t$ equals

$$\delta p = \frac{\sqrt[3]{2}\sqrt[3]{f}\kappa}{\pi[\mathrm{Ai}(\frac{1}{2^{2/3}\tilde{f}(x_m)^{2/3}})^2 + \mathrm{Bi}(\frac{1}{2^{2/3}\tilde{f}(x_m)^{2/3}})^2]} \sim \sqrt{\frac{\kappa^5}{E_0}}e^{-\frac{2}{3f(0)}}, \quad (4)$$

where $\tilde{f}(x) = E_0/(\kappa^2 - 2xE_0)^{3/2}$ is the reduced field, and the second equality is valid at small field asymptotics. The latter shows that the derived 1D Wigner momentum shift coincides with the momentum shift due to interference of the direct and the under-the-barrier recolliding trajectories discussed for elliptical fields (for 3D linear case see Ref. [45]).

In the present discussion the effect of the Coulomb field of the atomic core is neglected in the description of the ionization process. Including the Coulomb field, the tunneling process in the static regime takes place along one of the parabolic coordinates. In the transverse direction to the tunneling coordinate the electron dynamics is confined by a channel which would suppress the spreading and increase the recollision probability. We may therefore expect that in a realistic situation with a Coulombic atomic core in action, the under-the-barrier rescattering process would be more similar to the case when spreading is neglected for a short-range potential.

Concluding, we have found a new type of rescattering trajectory during the under-the-barrier dynamics in strong-field tunneling ionization, and demonstrate that interference of the direct and under-the-barrier recolliding trajectories induces a shift of the peak of the photoelectron momentum distribution. The momentum shift can be interpreted as a tunneling time delay. What is remarkable is that the time delay decreases with decreasing field strength, i.e., the thicker the barrier, the smaller the delay, or the longer the electron has to travel under the barrier, the smaller is the delay time. Consequently, the tunneling time delay can be observable only in the near-threshold regime of strong-field ionization that explains the calculated vanishing time delay in Ref. [18].

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