Laser-driven recollisions under the Coulomb barrier

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Photoelectron spectra obtained from the ab initio solution of the time-dependent Schrödinger equation can be in striking disagreement with predictions by the strong-field approximation (SFA) not only at low energy but also around twice the ponderomotive energy where the transition from the direct to the rescattered electrons is expected. In fact, the relative enhancement of the ionization probability compared to the SFA in this regime can be several orders of magnitude. We show for which laser and target parameters such an enhancement occurs and for which the SFA prediction is qualitatively good. The enhancement is analyzed in terms of the Coulomb-corrected action along analytic quantum orbits in the complex-time plane, taking soft recollisions under the Coulomb barrier into account. These recollisions in complex time and space prevent a separation into sub-barrier motion up to the “tunnel exit” and subsequent classical dynamics. Instead, the entire quantum path up to the detector determines the ionization probability.

Various surprises in strong-field photoelectron spectra (PES) have been found recently, especially at low energies and for long wavelengths [1]. Here, by “surprise” we mean “in disagreement with the strong-field approximation” (SFA) [2–4] or tunneling theories [5]. The SFA is the theorists’ work horse for intense-laser ionization problems and has been particularly insightful thanks to its formulation in terms of quantum orbits [6–9]. However, in plain SFA, for the so-called direct SFA matrix element, the Coulomb interaction of the outgoing photoelectron with its parent ion is neglected. Laser-driven recollisions can be taken into account in an extended SFA via a Born-like rescattering matrix element [4, 10, 11], leading to good agreement with experimental results or ab initio solutions of the time-dependent Schrödinger equation (TDSE) for high photoelectron momenta or emission angles where the direct SFA matrix element alone yields exponentially small probabilities [12]. The strongest discrepancies between SFA and TDSE or experiment were found in the low [13, 14], very low [15, 16], and zero energy [1, 17] regime where surprisingly high ionization probabilities for particular final electron momenta were observed. All these low-energy structures were found to originate from soft (multiple) laser-driven recollisions [18, 19]. Their locations in momentum space are even encoded in the rescattering SFA matrix element [12, 20] but they do not stick out probability-wise without taking interaction with the parent ion into account.

In this Letter, we examine the energy regime around twice the ponderomotive energy. The ponderomotive energy \( U_p \) is the time-averaged kinetic energy of a free electron in a laser field. For a linearly polarized laser field of the form \( E(t) = E_0 \cos \omega t \) (in dipole approximation) the velocity \( v \) of a classical electron emitted at time \( t_0 \) with \( v(t_0) = 0 \) is, at \( t \to \infty \), when the laser is off, \( v = \left(E_0 / \omega \right) \sin \omega t_0 \) (atomic units are used unless noted otherwise). The classically highest kinetic energy \( E^2 / (2m^3) = 2U_p \) thus results for ionization times \( t_0 \) where \( \sin \omega t_0 = 1 \). Since at such times \( E(t) = 0 \), tunneling rate formulas [5] will give zero weight to such electrons. However, as shown in Fig. 1 below, \( 2U_p \)-photoelectrons are clearly observed in spectra obtained solving numerically the TDSE. The SFA, as a quantum mechanical approach, has no abrupt cut-off. In fact, the SFA has not even a cut-off-like feature around \( 2U_p \) such as a change in slope. Apart from the well understood intra and inter-cycle interferences [21], SFA spectra obtained using the direct matrix element alone just roll-off featureless. At some photoelectron energy—typically around \( (2–4)U_p \)—the rescattering SFA matrix element takes over, up to the rescattering cut-off around \( 10U_p \) from where on the photoelectron yield ceases quickly with increasing energy. The SFA rescattering matrix element could be made very strong between zero and \( 2U_p \) energy by reducing the screening in the model potential. In fact, for potentials with a Coulomb tail, the rescattering probability for energies between zero and \( 2U_p \), can be comparable to the probability for direct ionization. The SFA rescattering matrix element then generates an overpronounced, step-like plateau up to \( 2U_p \), which is in even worse agreement with the TDSE spectrum than the probability from the direct SFA matrix element alone.

The main purpose of this Letter is threefold: (i) to predict for which laser and target parameters the direct SFA spectrum is in good or bad agreement with the TDSE result concerning the yield around \( 2U_p \), (ii) to highlight the mechanism that leads to an enhancement in the yield employing Coulomb-corrected quantum orbits in complex space and time, and (iii) to show that the notion of a “tunnel exit” after which the electron dynamics can be treated classically is erroneous in this case. In fact, it turns out that none of the commonly applied Coulomb corrections of orbits starting from the tunnel exit in real space and time (incl. our own used in Ref. [22]) makes up for the enhanced yield around \( 2U_p \), simply because the ionization probability is determined once the electron arrived at the tunnel exit in such approaches. Instead, the proper incorporation of soft recollisions via navigating the quantum orbits through Coulomb-generated branch cuts in the complex-time
plane is required, as was pointed out in Refs. 23, 24.

A prominent example for a distinct $2U_{\text{p}}$-plateau in the photoelectron yield can be found in Fig. 3 of Ref. 25. In this experiment, a short, intense 7-micron laser pulse ionized Xe atoms prepared in the 6s excited, metastable state of ionization potential $I_\text{p} = 0.14$. In Fig. 4(a) a momentum-resolved PES in laser polarization direction from the numerical solution of the TDSE is shown for the laser parameters of Ref. 25. In order to reproduce $I_\text{p}$ in single-active-electron approximation the 2s state in an effective potential $V_{\text{eff}}(r) = -[Z + (Z_{\text{ion}} - Z)e^{-r/r_\text{s}}]/r$ was used as the initial state, where $Z = 1$ and $Z_{\text{ion}} = 54$ are the asymptotic ion charges as $r \to \infty$ and $r \to 0$, respectively, and a screening length $r_\text{s} = 0.026$ was tuned to match $I_\text{p}$. As the SFA cannot be expected to give correct ionization probabilities in absolute numbers, we are free to shift the corresponding SFA spectrum vertically. We may adjust it to match the TDSE result in the rescattering-plateau region or at low energies. In either case we observe a discrepancy between TDSE and SFA of more than an order of magnitude for energies between $U_\text{p}$ and $4U_\text{p}$. The prediction of a “simple man’s model” (SMM, see, e.g., 23) where each ionization time $t_0$ introduced above is weighted by a tunneling rate is included in Fig. 4(a). The SMM spectrum drops $\sim \exp[-2Z(Z_{\text{ion}} - Z)/r]$ where $F = E/(2I_\text{p})^{3/2}$ is the reduced electric field and $q = p/\sqrt{4U_\text{p}}$, i.e., even faster with increasing momentum than the SFA, as it approaches zero at the cut-off momentum $p = \sqrt{4U_\text{p}}$.

In order to determine the relevant parameters that govern the (dis)agreement between TDSE and SFA let us consider first the TDSE for the wavefunction $\Psi(r, t)$ describing the electron in a hydrogen-like ion and a linearly polarized laser field. Expressed in dimensionless time $\tau = \omega t$ and position $\tilde{r} = \sqrt{\omega} r$ (in SI units see 22) the TDSE reads

$$i \frac{\partial}{\partial \tau} \tilde{\Psi} = \left[ -\frac{1}{2} \Delta_{\tilde{r}} - \frac{Z}{\sqrt{\omega} r} + \frac{\tilde{E}}{\omega^{3/2}} \bar{z}f(\tau) \cos \tau \right] \tilde{\Psi} \tag{1}$$

with $\tilde{\Psi} = \Psi[r(\tilde{r}), t(\tau)]$ and $f(t) \leq 1$ a dimensionless envelope function. The scaling was chosen such that the left hand side (energy) and the kinetic energy (first term on the right hand side) keep their form, i.e., do not acquire an extra prefactor. Then the Coulomb potential scales with a factor $\alpha_C = Z/\sqrt{\omega}$ and the laser-field with $\alpha_L = \tilde{E}/\omega^{3/2} = \sqrt{\omega}$, where $\sqrt{\omega}$ is the strong-field parameter 4. For hydrogen-like ions $I_\text{p} = Z^2/(2n^2)$ where $Z = \sqrt{\omega}$ and $n$ is the principal quantum number. For ground states $n = 1$ in hydrogen-like ions, the ratio $\alpha_C/\alpha_L$ thus equals the Keldysh parameter $\gamma = \sqrt{I_\text{p}/(2U_\text{p})}$ 2.

For fixed $\alpha_C$, $\alpha_L$, and initial-state quantum numbers but otherwise different laser and target parameters the same PES are obtained when plotted vs the dimensionless momentum $p/\sqrt{\omega}$. This is demonstrated in Fig. 4(b) for $\alpha_C = \alpha_L = 4$ (the other parameters are given in the figure caption).

The plain SFA is known to depend on only two dimensionless parameters as well, e.g., any pair of the set $\gamma$, $\sqrt{\omega}$, the reduced electric field $F$, and the multiquantum parameter $K_0 = I_\text{p}/\omega$. As a consequence, the plain SFA predicts the same scaled spectrum too for the two cases in Fig. 4(b) (plotted only once). The intra-cycle interference pattern is overpronounced in the SFA PES, which can lead to orders-of-magnitude discrepancies with the TDSE around certain photon energies. Expressed in terms of the ionization potential for hydrogen-like ions $I_\text{p} = Z^2/(2n^2)$ we obtain $\alpha_C = n\sqrt{2T_\text{p}}/\sqrt{\omega}$, i.e., $\alpha_C = 2n^2K_0$. Fixing $I_\text{p}$ and $Z$ independently is like changing the principal quantum number $n$ of the initial state in a hydrogen-like ion accordingly. This variable effective principal quantum number is a third dimensionless parameter that comes into play when working with non-hydrogen-like effec-

![FIG. 1. (Color online). TDSE spectra around the $2U_{\text{p}}$ cut-off compared to plain-SFA spectra. (a) same $I_\text{p} = 0.14$ (3.8 eV) and laser parameters as in Ref. 25, i.e., $\omega = 0.0065$ (7 microns), $E = 0.0045$ ($I = 7.1 \times 10^{13}$ W/cm$^2$), $\alpha_C = 12.4$ (asymptotic charge $Z = 1$), $\alpha_L = 8.6$, using a 6-cycle sin$^2$-shaped pulse envelope. The shaded area between TDSE and SFA spectrum highlights the order-of-magnitude discrepancy. Vertical lines indicate 1, 2, and $4U_\text{p}$. The SMM prediction is plotted bold black. Panel (b) shows two TDSE PES vs scaled momentum, both for the groundstate as initial state and both with $\alpha_C = \alpha_L = 4$ but one for $Z = 1$, $\omega = 0.0625$, $E = 0.0625$ (purple) and the other for $Z = 2$, $\omega = 0.25$, $E = 0.5$ (green, shifted for better comparison). The SFA result is included (cyan). Panel (c) shows two TDSE PES for similar $I_\text{p}$, realized via an excited state (purple) and an effective potential (green), respectively (see text). In both cases $\alpha_C = 11.3$ and $\alpha_L = 5.7$. The corresponding SFA spectrum is in striking disagreement. Panel (d) shows TDSE and SFA PES in good agreement for H(1s) as in (b) but for $\alpha_L = 6$.](image-url)
tive potentials (or when starting from different initial states \([28]\)).

Figure \([1]\)c shows a TDSE PES for \(\alpha_c = 11.3\) and \(\alpha_L = 5.7\) for \(Z = 1, n = 2\) (2s-state, \(I_p = 0.125\)), \(\omega = 0.0078\), and \(\hat{E} = 0.0039\). The second TDSE spectrum in this panel was calculated for the groundstate of \(I_p = 0.143\) (cesium) using the effective potential introduced above with \(Z_{\text{full}} = 0.1\) and \(r_e = 3.13\) to match \(I_p\). The TDSE spectra are very similar, showing that mainly \(I_p\) and the asymptotic \(-Z/r\) matter for the overall shape of the PES. Note that the TDSE spectra are rather flat up to \(2U_p\), in striking disagreement with the SFA [29] for an s-state with \(I_p = 0.125\) despite the fact that the Keldysh parameter is still \(\gamma = 1\), as in Fig. \([1]\)b where the agreement with the SFA is good. This shows that the Keldysh parameter alone is not sufficient to characterize the importance of Coulomb corrections.

Figure \([1]\)d shows an example where both TDSE and SFA PES display a rather flat \(2U_p\) plateau and a slow roll-off down to the rescattering plateau. The same parameters as for the H(1s) case in Fig. \([1]\)b were used, apart from the increased field amplitude \(\hat{E} = 0.09375\), i.e., \(\alpha_L = 6\).

The TDSE solutions yield little insight as to how the binding potential modifies PES. The SFA, on the other hand, can be interpreted (and evaluated) using quantum orbits in complex spacetime. Starting point is the direct SFA matrix element in saddle-point approximation [8, 9]

\[ M_{p}^{(\text{SFA})} = \sum_{\alpha} f_{\Psi_0}(p,t_{0\alpha}) e^{i S_{p,t_{0\alpha}}(t_{0\alpha})} \] where the sum runs over all complex solutions \(t_{0\alpha}\) of the saddle-point equation \([p + A(t_{0\alpha})]^2/2 = -I_p\) for a given final photoelectron momentum \(p\). The vector potential \(A(t)\) is related to the electric field according \(\hat{E}(t) = -\partial_t A(t)\). The phase \(S_{p,t_{0\alpha}}\) is the classical action along an electron orbit in the laser field \(A(t)\),

\[ S_{p,t_{0\alpha}}(t_{0\alpha}) = -\int_{t_{0\alpha}}^{T_p} \left\{ \frac{1}{2} |p + A(t)|^2 + I_p \right\} dt, \] but evaluated for complex times, making the orbit “quantum.” At the upper integration limit \(T_p\) the laser pulse is off again. For \(t > T_p\) all interfering trajectories with momentum \(p\) evolve equally and the propagation to the detector at \(t \rightarrow \infty\) yields a constant phase factor only. The pre-exponential factor \(f_{\Psi_0}(p,t_{0\alpha})\) is proportional to the matrix element \(\langle p + A(t_{0\alpha})|E(t_{0\alpha})\rangle\) and thus acts like a form factor that depends on the initial state \(\Psi_0\).

We Coulomb-correct the plain SFA along the lines of Refs. [9, 22, 30] (Coulomb-corrected SFA) or [31, 32] (analytical \(R\)-matrix method). When applied in the perturbative regime both methods consider the change in the action due to the binding potential by integrating \(V(r(t))\) along the unperturbed, complex plain-SFA quantum orbit

\[ r(t) = p(t - t_{0\alpha}) + \int_{t_{0\alpha}}^{t} A(t') dt', \] leading to

\[ \Delta S_{\text{CC}}(t_{0\alpha}) = -\int_{t_{0\alpha}}^{T_p} V[r(t)] dt. \] The singularity of \(V(r) = -Z/r\) at the initial position \(r(t_{0\alpha}) = 0\) is regularized through matching to the field-free initial-state wavefunction [5, 9, 30].

Without Coulomb correction the action \([3]\) can be evaluated along any integration path in the complex-\(t\) plane from complex \(t_{0\alpha}\) to real \(T_p\), thanks to the analyticity of the integrand. When including the Coulomb-correction in \([4]\) for \(V(r) = -Z/\sqrt{r^2}\) one has to take into account the branch points \(r^2 = 0\) and branch cuts originating from them along, e.g., \(\text{Im} r^2 = 0\), \(\text{Re} r^2 < 0\), and make sure that the integration path remains on a fixed Riemann sheet. The topology of branch cuts has been analyzed in Refs. [23, 24]. Proper integration pathways and their effect on PES have been investigated in [24] to reveal the origin of low-energy structures. To identify the mechanism behind the enhanced ionization yield around \(2U_p\) we performed a similar analysis. First, we calculated (for each final momentum \(p\) of interest) the positions of the branch points determined by the plain-SFA quantum orbit \([3]\) for \(r^2(t) = 0\). The roots of this equation come in pairs, see Fig. \([2]\)a. For so-called short orbits that do not return closely to the core, no obstacles in the form of branch cuts are put in the way of the “standard” integration path, which is down to the real-time axis at \(t = \text{Re} t_{0\alpha}\) (interpreted as the “tunnel exit” \(\text{Re} r(\text{Re} t_{0\alpha})\) in position space) and then along the real axis to \(t = T_p\), as in Fig. \([2]\)b. For so-called long orbits with close approaches to the core the pair of branch points are closer together, leaving a narrow gap only at finite imaginary-time values to navigate the integration path through, see Fig. \([2]\)a. As a consequence, the standard integration path is blocked by a branch cut. The algorithm used to find an analytic integration path first determines all returns on the real-time axis, then computes the corresponding branch point pair or “gate” as solutions of \(r^2(t) = 0\). These are used to divide solutions of \(r(t) \cdot v(t) = 0\)—the principal waypoints—into times of closest approach (which represent the center of a gate) and intermediate turning points (corresponding to classical turning points) [24]. These waypoints are then used to construct a valid, analytic integration path passing perpendicularly through the gates.

The effect of the Coulomb-correction to the action \([4]\) for the dominating orbits is shown in Fig. \([2]\)c. For the plain SFA the contributions of short and long trajectories are equal. The Coulomb correction for short trajectories increases the ionization probability uniformly. This is consistent with previous Coulomb-corrected total ionization rates [5] which are found by calculating the Coulomb correction along the unperturbed, dominant \(p = 0\)-trajectory analytically. For long trajectories the spectrum is qualitatively modified. For small momenta low-energy structures are recovered (purple curve in Fig. \([2]\)c), as in Ref. [24]. Around \(2U_p\), the yield is significantly increased compared to the short-orbit contribution, generating a plateau in the spectrum. The magnitude of this increase depends on the angle \(\vartheta = \arccos(p_{\parallel}/p)\). For \(\vartheta = 0\), i.e., in polarization direction, this correction diverges logarithmically, and for very small \(\vartheta\) it becomes large [see \(\vartheta = 10^{-3}\pi\) in Fig. \([2]\)c, yellow]. This is not surprising, as our perturba-
FIG. 2. (Color online). How long and short orbits are affected by the Coulomb correction [same parameters as in Fig. 1(a)]. Panels (a) and (b) show integration contours (cyan) in the complex-time plane for a long and a short orbit at the cut-off momentum, respectively. The color scale indicates $\text{Re} \sqrt{r^2(t)}$, the colored lines constant $\text{Im} \sqrt{r^2(t)}$ (red, yellow = 0, green > 0). White lines indicate branch cuts starting at branch points. For the short trajectory in (b) the integration contour can be chosen in the standard way because the recollission branch points and cuts are far away from the real axis, i.e., outside the complex-time domain shown in (b). For the long trajectory in (a) the integration path needs to be deformed in order to navigate through the branch cut gate. Panel (c) shows partial spectra from long and short trajectories for different angles $\vartheta$ and a plain SFA spectrum (calculated with only one trajectory per momentum to inhibit interferences). The angular dependence of the short-trajectory spectrum is negligible. Panel (d) displays the trajectory weights $\text{Im} \Delta S_{CC}$, measured along the integration contours in (a) and (b). Note that every kink in (d) is related to a change in the integration direction in (a), (b).
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