Numerical time-of-flight analysis of the strong-field photoeffect

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(Dated: August 17, 2020)

Short-time filtering of the photoionization amplitude extracted straight from the numerical solution of the time-dependent Schrödinger equation (TDSE) is used to identify dominant pathways that form photoelectron spectra in strong fields. Thereby, the “black-box nature” of TDSE solvers only giving the final spectrum is overcome, and simpler approaches, e.g., semi-classical based on the strong-field approximation, can be tested and improved. The approach also allows to suppress intercycle quantum interference between pathways removing patterns that are usually washed out in experiments.

In order to predict photoelectron spectra that are measured in intense laser-matter interaction experiments, theorists proposed a plethora of approaches, from first-principle numerical [1–7] to simplified, semi-classical [8–11]. In general, the simpler and the more analytical a model is, the more insight into the photoionization process it gives while precise quantitative predictions are hardly possible. On the other hand, the most accurate results are obtained from ab initio solutions of the time-dependent Schrödinger equation (TDSE). However, as in an experiment, it is hard to disentangle in TDSE simulations all the various processes the photoelectron undergoes on its way to the detector. The strong-field approximation (SFA) [12–14] took the lead as far as insight into the strongly nonlinear electron dynamics is concerned, especially when formulated in terms of semi-classical quantum orbits [8, 15–18]. The investigation of these orbits and their weights allows to identify the dominant ionization pathways [15, 16, 19, 21]. However, photoelectron spectra calculated by such approaches can be quantitatively orders of magnitude off or have a qualitatively wrong shape because of Coulomb effects [22–25]. The influence of the Coulomb potential on the photoelectron can be easily accounted for within classic-trajectory Monte-Carlo simulations [11, 26], which, however, lack interference effects intrinsic to quantum systems. In the present work, we propose ways to identify the relevant ionization pathways in ab initio solutions to the TDSE. Our method combines the accuracy of the TDSE with the insight offered by quantum-orbit methods. Moreover, simple, semi-analytical models can be benchmarked and improved using the TDSE-based results.

We start with a brief introduction to the time-dependent surface flux method (tSURFF) for the calculation of photoelectron spectra [7, 27]. Consider an electron described by the state $|\Psi(t)\rangle$ and a laser field $E(t)$ defined via its vector potential $A(t) = -\int_{t'}^{t} E(t')dt'$ in dipole approximation. Initially, a bound state of the atomic potential $|\psi_{\text{bound}}(t)\rangle$, transitions to continuum states $|k(t)\rangle$ due to interaction with the laser may occur, contributing to the free part $|\psi_{\text{free}}(t)\rangle$.

$$|\Psi(t)\rangle = |\psi_{\text{bound}}(t)\rangle + |\psi_{\text{free}}(t)\rangle,$$

$$|\psi_{\text{free}}(t)\rangle = \int |k(t)\rangle \langle k(t)| \psi_{\text{free}}(t) \rangle dk.$$  \hspace{1cm} (2)

The momentum-resolved photoelectron spectrum is defined as

$$Y(k) = |a_k(T)|^2 = |\langle k(T)|\psi_{\text{free}}(T)\rangle|^2$$  \hspace{1cm} (3)

where time $T \rightarrow \infty$. Since all bound states are negligible at large enough distances where the photoionized part of the wavefunction is localized,

$$Y(k) = |a_k(T)|^2 \approx |\langle k(T)|\Theta(r-R)|\Psi(T)\rangle|^2$$  \hspace{1cm} (4)

with the Heaviside step function $\Theta(r-R)$ cutting away the contributions from distances $r < R$. With $a_k(0) = 0$, the amplitudes for the continuum states of momentum $k$ at the final time can be written as

$$a_k(T) = \int_0^T \partial_t a_k(t) dt.$$  \hspace{1cm} (5)

FIG. 1. (a) Time-energy-resolved photoelectron spectrum in the laser propagation direction computed from (7) and normalized to its maximum. Laser wavelength $\lambda = 2000$ nm, intensity $I = 10^{13}$ W/cm$^2$. Flux-capturing surface is at $R = 350$ atomic units. Colored lines indicate times at which the electrons that originate from times when $E(t)$ peaks (red and black lines for $k > 4U_p$) or equals zero (green lines at $E_k > 4U_p$) arrive at the surface according to (10). (b) Laser profile $E(t)$ with points referring to colored lines in (a).

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Using \( \partial_k \bullet = i [\hat{H}(t), \bullet] \), it is straightforward to show that the amplitudes \( a_k(T) \) can be expressed as a time integral over the flux \( j_k \) through the surface \( S_R \) at \( r = R \),

\[
a_k(T) = \int_0^T \left( \int_{S_R} j_k(r, t) dS_r \right) dt
\]

with the normal vector on the surface \( dS_r = n_r dS \). The particular form of the surface flux \( j_k(r, t) \) used in this work is described in the Appendix and in [2].

**Time-energy photoelectron distribution.** Before tSURFF became widely used the window-operator method (WOM) [28] was a common way to obtain the photoelectron spectrum from the final wavefunction calculated by some TDSE solver. While WOM gives the correct total electron spectrum, momentum or angle-resolved spectra are only approximate [29]. Nevertheless, phase-space distributions after the laser pulse were obtained using WOM, which proved useful for comparisons with semi-classical theories [30, 31]. The main technical advantage of tSURFF over WOM is that there is no need to keep track of the full wavefunction, which rapidly spreads over a huge area due to strong ionization. Instead, the photoionization amplitude \( a_k(T) \) is expressed as a time integral, as mentioned above. Albeit a mathematical trick in the first place, this provides the opportunity to study the build-up of the photoelectron spectrum in a time-resolved way. In [32], the integrand itself has been investigated, which, however, is highly oscillatory. Instead, we introduce a Gaussian time window in the integrand,

\[
\partial_t \tilde{a}_k(t_0) = \int_0^T e^{-\frac{(t-t_0)^2}{2\tau_w^2}} \partial_t a_k(t) dt, \tag{7}
\]

which amounts to the zero-frequency component of a Gabor transform [33]. Note that the transform (7) conserves the total amplitude and the yield because integration over \( t_0 \) gives back [5] up to a constant \( \sqrt{2\pi\tau_w^2} \), and

\[
\tilde{Y}(k) = \left| \int_{-\infty}^{\infty} \int_{0}^{T} e^{-\frac{(t-t_0)^2}{2\tau_w^2}} \partial_t a_k(t) dt \right| dt_0^2 = 2\pi\tau_w^2 Y(k). \tag{8}
\]

We choose \( \omega t_w = 0.2\pi \), i.e., a tenth of a laser cycle, so that only intracycle interference is captured at this subcycle time resolution. An example for such a time-resolved spectrum is shown in Fig. [1] for a 6-cycle laser pulse with an “1-4-1 envelope” (i.e., \( A(t) \) has a 4-cycle flat-top central part and an 1-cycle sin²-ramping on each side).

When applying the tSURFF method to calculate photoelectron spectra in TDSE calculations, one usually adopts a classical picture for laser-driven photoelectrons to estimate the time limit \( T \) for integration as the time that the slowest electrons of interest need to reach the flux-capturing surface,

\[
T \geq T_{\text{pulse}} + \frac{R}{k_{\text{min}}}. \tag{9}
\]

The density map in Fig. [1] does not only support the classical estimate [9] but additionally allows to resolve times when electrons with a certain energy most likely arrive at the detector. Typically, strong-field photoelectron spectra consist of so-called “direct” electrons with energies \( E_k < 2 \div 4U_p \) (where \( U_p = I/4\omega^2 \) is the ponderomotive energy in atomic units), and “rescattered” electrons at energies \( E_k < 10U_p \). The direct electrons are emitted at times with high absolute value of the electric field \( |E(t)| \) while the final energy of the rescattered electrons is determined at scattering times when the absolute value of the vector potential \( |A(t)| \) approaches its maximum (we consider first-order returns only) [15]. Indeed, from Fig. [1] it is clear that the curves \( t_{\text{reg}}(k, t_0) \) obtained from

\[
R = \int_{t_0}^{t_{\text{reg}}} A(t) dt + k(t_{\text{reg}} - t_0) \tag{10}
\]

match the corresponding times of highest time-resolved yield. Note that for registration times \( t_{\text{reg}} > T_{\text{pulse}} \) this equation simplifies so that the curves \( t_{\text{reg}}(k, t_0) \) are less “wiggly”. In any case, the time-resolved yield is nicely aligned along these curves of arrival.

A way to visualize the build-up of intercycle interference and the rescattering plateau was proposed in [34]. At times \( t_E \) where \( E(t_E) = 0 \), the contribution \( |\psi_{\text{bound}}(t_E)|^2 \) was projected out of the state \( |\Psi(t_E)|^2 \). The calculated spectra were then equal to those where the laser pulse was suddenly terminated at times \( t_E \) because the authors worked in the length gauge and, thus, eigenstates of their Hamiltonian at times \( t_E \) were eigenstates of the laser-free Hamiltonian. Analogously, within the velocity gauge this approach is valid at times \( t_A \) such that \( A(t_A) = 0 \). After obtaining \( |\Psi(t_A)|^2 \) one may set \( A(t > t_A) = 0 \) and apply the iSURFV method to efficiently post-propagate according to the field-free Hamiltonian.
The particular form of $J$ can be interpreted as the complex ionization times because similar build-ups are observed in SFA-based theories [1, 32, 35], where the photoelectron amplitude $M$ is also given by a time integral.

In quantum-orbit theory, this time integral is evaluated using the saddle-point method, and the saddle points can be interpreted as the complex ionization times $t_s$ that contribute predominantly,

$$a_s^{\text{SFA}}(T) = \sum_s M(k(t_s)).$$

A sudden shutdown of the laser in the TDSE simulations corresponds to discarding ionization times with $\text{Re}(t_s) > t_E$ (or $\text{Re}(t_s) > t_A$ in velocity gauge) [15, 38, 39].

Continuing exploring features accessible in SFA-based theories, we address the possibility to exclude intercycle interference (see, e.g., [38, 39]). In quantum orbit theory, this can be achieved by considering an incoherent sum over saddle points,

$$Y(\theta) = \sum |M(k(t_s))|^2.$$
while it is violated for a hydrogenic binding potential (see Figs. 4(a,b)) where a deviation in the arrival time \( \omega \Delta t_{\text{reg}} = -\Delta \phi \sim 0.12\pi \) is observed and illustrated in Fig. 5. The mapping of registration, ionization or tunneling-time delays to angular shifts is the basic idea behind so-called “attoclock” experiments [41,44].

**Conclusions.** We showed how the dominant pathways that constitute photoelectron spectra in strong-field laser ionization experiments are revealed by applying a short-time filter to the ionization amplitude that is calculated anyway within the efficient ISURFF approach incorporated in state-of-the-art strong-field TDSE solvers. Our method allows to connect ab initio TDSE simulations with simple and intuitive semi-analytical theories, thus providing insight and a way to benchmark and improve simple models. We also pointed out an efficient way to remove intercycle interference, revealing the envelopes of photoelectron spectra without ATI peaks.

Future work may concentrate on regimes where simple, semi-classical theories actually do not work, e.g., in the over-barrier-ionization regime or if excited states play a role.

This work was supported by the project BA 2190/10 of the German Science Foundation (DFG).

**Appendix.** All TDSE solutions in the present paper were obtained with the QPROP software [1,3]. For the examples with linear polarization an argon pseudo potential

\[
U_{Ar}(r) = \frac{1+17e^{-cr}}{r}, \quad r < R_{co}
\]

with \( c = 2.2074 \) was used. This leads to an ionization potential of \( I_p = 15.8 \text{eV} \) for the 3p initial state on a radial grid of resolution \( dr = 0.1 \). The flux-capturing surface was put at \( R = 350 \).

For the example with elliptical polarization the hydrogenic potential

\[
U_{Hy}(r) = \frac{1}{r}, \quad r < R_{co}
\]

and a short-range potential

\[
U_{sr}(r) = -C_1 e^{-c_2 r}, \quad r < R_{co}
\]

with \( C_1 = 5.2074 \) and \( c_2 = 5.0 \) were used. Both lead to a ground-state ionization potential \( I_p = 13.6 \text{eV} \). The flux-capturing surface was put at \( R = 200 \). In all calculations the long-range Coulomb tail was removed by matching it at \( r = R_{co} = 50 \) to a linear roll-off that reaches zero at \( r = 2R_{co} \). Since \( R > 2R_{co} \), this approximation allows to use Volkov functions for \( |k(t)| \) and the Volkov Hamiltonian

\[
\hat{H}(t) = \frac{(-i\nabla + A(t))^2}{2} - \frac{A^2(t)}{2},
\]

(with the purely time-dependent \( A^2(t)/2 \) term transformed away), leading to the density flux

\[
j_k(r, t) = \frac{i}{2} (k(t)|\nabla + iA(t)|r)(r|\Psi(t))
- \frac{i}{2} (k(t)|r)(r|\nabla + iA(t)|\Psi(t))
\]  

FIG. 4. (a,c) Integrated flux according (18) vs angle and time. Wavelength \( \lambda = 800 \text{nm} \), intensity \( I = 10^{14} \text{W/cm}^2 \), ellipticity \( \epsilon = 0.882 \). Flux-capturing surface is put at \( R = 200 \). Gray lines correspond to angles of \( -A(t) \), black lines on top of them indicate angles where \( |E(t)| > 0.986E_{\text{max}} \). (b,d) Total angular distributions, i.e., sections through \( t = T \) of (a,c). (a,b) for hydrogenic binding potential, (c,d) for short-range potential with same ionization potential \( I_p = 13.6 \text{eV} \).

FIG. 5. A section through the time and angle resolved distributions shown in Fig. 4 at angles \( \phi \) where the total yield is maximum, i.e., \( \phi = -0.495\pi \) (solid black) and \( \phi = 0.373\pi \) (solid gray) for hydrogen, \( \phi = -0.305\pi \) (dashed) and \( \phi = 0.495\pi \) (dashed gray) for short-range potential. All four curves are normalized to their maxima.
needed in equation (6). Unless the iSURFV method was used, the upper limit for the time integration was chosen $T = T_{\text{pulse}} + 500$. The time window

$$w(t) = 1 - e^{-\frac{(t - T)^2}{2T^2}} \quad (27)$$

with $T = 100$ was added to suppress the effects of finiteness of $T$. In the expression (12), where we use the iSURFV approach [11, 32, 35], the time-integrated flux density is

$$J_k^{(\infty)}(r, t_A) = \frac{i}{2} \langle \langle k(t_A) | \nabla | r \rangle \langle \frac{1}{E_k - H_0} | \Psi(t_A) \rangle - \frac{i}{2} \langle k(t_A) | r \rangle \langle \frac{1}{E_k - H_0} | \Psi(t_A) \rangle \rangle. \quad (28)$$

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