“The envelope hamiltonian for electron interaction with ultrashort pulses”:
Supplemental material.

A. Adiabatic time-dependent perturbation theory

Here, we sketch an adiabatic time-dependent perturbation theory (aTDPT) for \( H = H_0(t) + U(x,t) \), split into an unperturbed Hamiltonian

\[
H_0(t) = -\frac{1}{2} \nabla^2 + V_0(x,t),
\]

which is itself parametrically time-dependent and a time-dependent perturbation \( U(x,t) \). Let \(| j(t) \rangle\) and \(| k, t \rangle\) be an eigenstate of the Hamiltonian \( H_0(t) \) for fixed time \( t \) from the discrete and continuos part of the spectrum, respectively

\[
H_0(t)| j(t) \rangle = | j(t) \rangle \varepsilon_j(t)
\]

\[
H_0(t)| k, t \rangle = | k, t \rangle \varepsilon_k \quad \text{with} \quad \varepsilon_k = \frac{k^2}{2}.
\]

Together the \(| j(t) \rangle\) and \(| k, t \rangle\) form a complete orthonormal basis set

\[
\langle j(t)| j'(t) \rangle = \delta_{jj'}, \quad \langle j(t)| k, t \rangle = 0, \quad \langle k, t| k', t \rangle = (2\pi)^3 \delta(k-k'),
\]

which we label in the following for simplicity with greek letters. Consequently \( \sum | \beta(t) \rangle \langle \beta(t) | = 1 \) holds. Note that the eigenenergies \( \varepsilon_j(t) \) as well as all basis functions are time-dependent but the continuum energies \( \varepsilon_k = k^2/2 \) of course not.

We expand the solution \( \psi(t) \) of the Schrödinger equation

\[
[H_0(t) + Q(x,t)] | \psi(t) \rangle = 0 \quad \text{with} \quad Q(x,t) \equiv U(x,t) - i\partial/\partial t
\]

as

\[
| \psi(t) \rangle = e^{-i\chi(t)} \sum \beta(t) \rangle c_\beta(t) e^{-itE_\beta(t)},
\]

where \( \chi(t) \) is the usual phase freedom which is in our case time-dependent and will be chosen later to obtain a simple form of the differential equations for the coefficients \( c_\beta \). For continuum states \( \beta = k \) we have \( E_k(t) \equiv \varepsilon_k \) as usual, but for the bound states \( \beta = j \) the energies for the phase factor are given by \( E_j(t) \equiv t^{-1} \int^t dt' \varepsilon_j(t') \).
If we insert the ansatz (5) into Eq. (4) and project from the left onto $|\beta\rangle$ we obtain

$$i \dot{c}_\beta(t) = -c_\beta(t) \tilde{\chi}(t) + \sum_{\beta' \neq \beta} Q^{\beta \beta'}(t) c_{\beta'}(t) e^{-i t[E_{\beta'}(t) - E_\beta(t)]}, \quad (6a)$$

where

$$Q^{\beta \beta'}(t) \equiv \langle \beta(t)|Q(x, t)|\beta'(t)\rangle \quad (6b)$$

with $Q$ from Eq. (4).

The coupled Eqs. (6a) provide a full solution to Eq. (4). However, if $U(x, t)$ is only a weak perturbation, we can solve Eq. (4) to a good approximation by a single iteration, where we assume that only first order transitions (linear in $U$ or in $Q$, respectively) occur. With an initial occupation of a bound state $|b\rangle$ and all other states unoccupied it is

$$c_\beta^{(0)}(t) = 1, \quad c_{\beta \neq b}^{(0)}(t) = 0, \quad (7)$$

and we obtain by a single iteration of Eq. (6a)

$$i \dot{c}_b^{(1)}(t) = [Q^{bb}(t) - \tilde{\chi}(t)] c_b^{(0)}(t) + \sum_{\beta \neq b} Q^{\beta b}(t) c_\beta^{(0)}(t) e^{-i t[E_b(t) - E_\beta(t)]} \quad (8a)$$

$$i \dot{c}_\beta^{(1)}(t) = -\tilde{\chi}(t) c_\beta^{(0)}(t) + Q^{\beta b}(t) c_b^{(0)}(t)e^{-i t[E_b(t) - E_\beta(t)]} + \sum_{\beta' \neq b} Q^{\beta \beta'}(t) c_{\beta'}^{(0)}(t) e^{-i t[E_{\beta'}(t) - E_\beta(t)]}. \quad (8b)$$

If we choose $\tilde{\chi} = Q^{bb}$ we obtain from Eq. (8a) $c_b^{(1)}(t) = 0$ implying $c_b^{(1)}(t) = c_b^{(0)}(t) = 1$ and from Eq. (8b) for $\beta \neq b$

$$c_\beta^{(1)}(t) = -i \int_{-\infty}^{t} dt' Q^{\beta b}(t') e^{-i t'[E_b(t') - E_\beta(t')]} \quad (9)$$

The result (9) of this aTDPT agrees formally with that of the standard TDPT except for two (subtle) differences: (i) the basis states entering the matrix element $Q^{\beta \beta'}$, cf. Eq. (6b), are explicitly time-dependent and (ii) so are the energies $E_\beta(t)$ for the bound states, e.g., $\beta = b$.

The lowest-order (time-dependent) correction to the bound states is $\mathcal{O}(\alpha_0^2)$, where $\alpha_0$ is the effective quiver amplitude, see Eq. (12) below. Taking only terms up to order $\alpha_0$ we get

$$\bar{c}_\beta(t) = -i \int_{-\infty}^{t} dt' \langle \beta|U(x, t')|b\rangle e^{-i(E_b - E_\beta)t'}, \quad (10)$$

which coincides with the result of standard time-dependent perturbation theory in textbooks. In general, the population of a state $|\beta(t)\rangle$ at any time $t$ is given in aTDPT by Eq. (9), provided that the system was initially in state $|b\rangle$. 


B. Expansion of the envelope Hamiltonian in terms of the number of photons exchanged

In the manuscript, we are interested to split the transition operator $Q$ of the envelope Hamiltonian [Eq. (6) of the main manuscript] into contributions according to the number of photons emitted or absorbed. Hence, we write

$$-i \int_{-\infty}^{t} dt' Q^{b} (t') e^{-i [E_{b} (t') - E_{\beta} (t')] t} = \sum_{n=-n_{\text{max}}}^{+n_{\text{max}}} M_{n}(k, t)$$

with the $M_{n}(k, t)$ given in Eq.(9) of the main manuscript. For the dynamics discussed there, it has been sufficient to include a maximal exchange of $n_{\text{max}} = 2$ photons. This is also the minimal number required to have a consistent limit for very weak pulses $\alpha_{0} \ll 1$. Through the relation [Eq. (5) of the main manuscript]

$$\alpha_{0} = \frac{F_{0}}{\omega^{2} 1 + 8 \ln 2/(T \omega)^{2}}$$

small $\alpha_{0}$ is realized through a short pulse $T \to 0$ or high frequency $\omega \to \infty$.

In this limit the time-dependent Schrödinger equation formulated with the envelope Hamiltonian agrees for small effective quiver amplitudes $\alpha_{0}$ with the exact dynamics in the Kramers-Henneberger frame. To see this we expand the single-period-averaged potentials $V_{n}(x, t)$ to second order in $\alpha_{0}$:

$$V_{0}(x, t) \approx V(x) + \frac{1}{4} \frac{\partial^{2} V}{\partial x^{2}} \alpha^{2}(t),$$

$$V_{\pm 1}(x, t) \approx \frac{\partial V}{\partial x} \alpha(t) e^{\mp i \delta},$$

$$V_{\pm 2}(x, t) \approx \frac{1}{8} \frac{\partial^{2} V}{\partial x^{2}} \alpha^{2}(t) e^{\mp 2i \delta}.$$

With the interactions from Eq.(6) of the main manuscript the full potential without single-cycle averaging is recovered to order $\alpha_{0}^{2}$:

$$\sum_{n=-2}^{+2} V_{n}(x, t)e^{-in\omega t} \approx V(x) + \frac{\partial V}{\partial x} \alpha(t) \cos(\omega t + \delta) + \frac{1}{2} \frac{\partial^{2} V}{\partial x^{2}} \alpha^{2}(t) \cos(\omega t + \delta)^{2}$$

$$\approx V(x + e_{x} \alpha(t) \cos(\omega t + \delta)) .$$

Since already the non-adiabatic term Eq. (13a) with zero-photon exchange contains the same interaction potential as the term Eq. (13c) with two-photon exchange, it is necessary to have a minimum expansion length of $n_{\text{max}} = 2$ in Eq. (11) to obtain the correct asymptotic limit for small $\alpha_{0}$.
C. Pulse-dependent photo-ionization rates

The adiabatic perturbation theory for parametrically time-dependent perturbations allow one easily to formulate photo-ionization rates (involving true photon absorption) during the laser pulse as photo-ionization rates per optical cycle. To this end we simply define the probability for single-photon ionization (here for clarity in the 1D case as in the main paper) at time $t$ by integrating the single photon transition matrix element $M_n(k,t)$ over energy and a laser period $T_\omega$,

$$P_n(t) = \int \frac{dk}{2\pi} \left| \int_0^{T_\omega} dt' \langle k, t | V_n(x,t) | b(t) \rangle e^{i k' (k^2/2 - n\omega - \varepsilon_b(t))} \right|^2,$$

where we have fixed all pulse-envelope related time dependencies including that of the bound state energy $\varepsilon_b$ as a parameter. Then $E_b(t) = \varepsilon_b(t)$, since $E_b(t') = 1/t' \int_0^{t'} dt'' \varepsilon_b(t) = \varepsilon_b(t)$. The residual time dependence $t'$ in the phase of the integral in Eq. (15) produces a $\delta$-function $2\pi \delta(k^2/2 - n\omega - \varepsilon_b(t))$ while the second (complex conjugate) integral gives then trivially $T_\omega$. The final result for the single-photon ionization rate is then

$$\Gamma_n(t) = \frac{P_n(t)}{T_\omega} = \frac{1}{k} \left( |\langle +k(t)| V_n(x,t) | b(t) \rangle|^2 + |\langle -k(t)| V_n(x,t) | b(t) \rangle|^2 \right)$$

with $k(t) = [2n\omega + 2\varepsilon_b(t)]^{1/2}$. 