“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

$$H_0(t) = -\frac{1}{2}\boldsymbol{\nabla}^2 + V_0(\mathbf{x},t)\,, \tag{1}$$

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

$$H_0(t)|\mathrm{j}(t)\rangle = |\mathrm{j}(t)\rangle\,\varepsilon_\mathrm{j}(t) \tag{2a}$$

$$H_0(t)|\mathbf{k},t\rangle = |\mathbf{k},t\rangle\,\varepsilon_\mathbf{k} \qquad \text{with} \quad \varepsilon_\mathbf{k} = \frac{\mathbf{k}^2}{2}. \tag{2b}$$

Together the $|\mathrm{j}(t)\rangle$ and $|\mathbf{k},t\rangle$ form a complete orthonormal basis set

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

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

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

$$\left[H_0(t) + Q(x,t)\right]\big|\psi(t)\big\rangle = 0 \qquad \text{with} \quad Q(x,t) \equiv U(x,t) - i\partial/\partial t \tag{4}$$

as

$$\big|\psi(t)\big\rangle = e^{-\mathrm{i}\chi(t)} \sum\!\!\!\!\!\!\!\int\; |\beta(t)\rangle\, c_\beta(t)\, e^{-\mathrm{i}tE_\beta(t)}\,, \tag{5}$$

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_β. For continuum states $\beta = \mathbf{k}$ we have $E_\mathbf{k}(t) \equiv \varepsilon_\mathbf{k}$ as usual, but for the bound states $\beta = \mathrm{j}$ the energies for the phase factor are given by $E_\mathrm{j}(t) \equiv t^{-1}\int^t \mathrm{d}t'\,\varepsilon_\mathrm{j}(t')$.

If we insert the ansatz (5) into Eq. (4) and project from the left onto $|\beta\rangle$ we obtain

$$\mathrm{i}\,\dot{c}_\beta(t) = -c_\beta(t)\,\dot{\chi}(t) + \sum\!\!\!\!\!\!\int Q^{\beta\beta'}(t)\,c_{\beta'}(t)\,e^{-\mathrm{i}t[E_{\beta'}(t)-E_\beta(t)]}\,, \tag{6a}$$

where

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

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 $|\mathrm{b}\rangle$ and all other states unoccupied it is

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

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

$$\mathrm{i}\,\dot{c}_\mathrm{b}^{(1)}(t) = \left[Q^\mathrm{bb}(t) - \dot{\chi}(t)\right]c_\mathrm{b}^{(0)}(t) + \sum\!\!\!\!\!\!\int_{\beta\neq\mathrm{b}} Q^{\mathrm{b}\beta}(t)c_\beta^{(0)}(t)e^{-\mathrm{i}t[E_\beta(t)-E_\mathrm{b}(t)]} \tag{8a}$$

$$\mathrm{i}\,\dot{c}_\beta^{(1)}(t) = -\dot{\chi}(t)c_\beta^{(0)}(t) + Q^{\beta\mathrm{b}}(t)c_\mathrm{b}^{(0)}(t)e^{-\mathrm{i}t[E_\mathrm{b}(t)-E_\beta(t)]} + \sum\!\!\!\!\!\!\int_{\beta'\neq\mathrm{b}} Q^{\beta\beta'}(t)c_{\beta'}^{(0)}(t)e^{-\mathrm{i}t[E_{\beta'}(t)-E_\beta(t)]}\,. \tag{8b}$$

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

$$c_\beta^{(1)}(t) = -\mathrm{i}\int_{-\infty}^{t} \mathrm{d}t'\; Q^{\beta\mathrm{b}}(t')e^{-\mathrm{i}t'[E_\mathrm{b}(t')-E_\beta(t')]}. \tag{9}$$

The result (9) of this aTDPT agrees formally with that of the standard TDPT execpt 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., $\beta = \mathrm{b}$.

The lowest-order (time-dependent) correction to the bound states is $\mathcal{O}(\alpha_0^2)$, where α_0 is the effective quiver amplitude, see Eq. (12) below. Taking only terms up to order α_0 we get

$$\tilde{c}_\beta(t) = -\mathrm{i}\int_{-\infty}^{t} \mathrm{d}t'\,\langle\beta|U(x,t')|\mathrm{b}\rangle \mathrm{e}^{-\mathrm{i}(E_\mathrm{b}-\varepsilon_\beta)t'}, \tag{10}$$

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 $|\mathrm{b}\rangle$.

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

$$-\mathrm{i}\int_{-\infty}^{t}\mathrm{d}t'\,Q^{\beta\mathrm{b}}(t')e^{-\mathrm{i}t'[E_{\mathrm{b}}(t')-E_{\beta}(t')]}=\sum_{n=-n_{\max}}^{+n_{\max}}M_n(\mathbf{k},t) \tag{11}$$

with the $M_n(\mathbf{k},t)$ given in Eq.(9) of the main manuscript. For the dynamics discussed there, it has been sufficient to include a maximal exchange of $n_{\max}=2$ 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]

$$\alpha_0=\frac{F_0}{\omega^2}\frac{1}{1+8\ln 2/(T\omega)^2} \tag{12}$$

small α_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 α_0 with the exact dynamics in the Kramers-Henneberger frame. To see this we expand the single-period-averaged potentials $V_n(\mathbf{x},t)$ to second order in α_0:

$$V_0(\mathbf{x},t)\approx V(x)+\frac{1}{4}\frac{\partial^2 V}{\partial x^2}\alpha^2(t), \tag{13a}$$

$$V_{\pm1}(\mathbf{x},t)\approx \frac{\partial V}{\partial x}\alpha(t)\mathrm{e}^{\mp i\delta}, \tag{13b}$$

$$V_{\pm2}(\mathbf{x},t)\approx \frac{1}{8}\frac{\partial^2 V}{\partial x^2}\alpha^2(t)\mathrm{e}^{\mp 2i\delta}\,. \tag{13c}$$

With the interactions from Eq.(6) of the main manuscript the full potential without single-cycle averaging is recovered to order $\alpha_0{}^2$:

$$\begin{aligned}\sum_{n=-2}^{+2}V_n(\mathbf{x},t)e^{-in\omega t}&\approx V(\mathbf{x})+\frac{\partial V}{\partial x}\alpha(t)\cos(\omega t+\delta)+\frac{1}{2}\frac{\partial^2 V}{\partial x^2}\alpha^2(t)\cos(\omega t+\delta)^2\\ &\approx V(\mathbf{x}+\mathbf{e}_x\alpha(t)\cos(\omega t+\delta))\,.\end{aligned} \tag{14}$$

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 $n_{\max}=2$ in Eq. (11) to obtain the correct asymptotic limit for small α_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_ω,

$$P_n(t) = \int \frac{dk}{2\pi} \left| \int_0^{T_\omega} \mathrm{d}t' \, \langle k,t|V_n(x,t)|\mathrm{b(t)}\rangle \mathrm{e}^{\mathrm{i}t'(\mathrm{k}^2/2-\mathrm{n}\omega-\varepsilon_\beta(\mathrm{t}))} \right|^2 , \tag{15}$$

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

$$\Gamma_n(t) = \frac{P_n(t)}{T_\omega} = \frac{1}{k} \left(|\langle +k(t)|V_n(x,t)|\mathrm{b}(t)\rangle|^2 + |\langle -k(t)|V_n(x,t)|\mathrm{b}(t)\rangle|^2 \right) \tag{16}$$

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