mpipks

Finite Systems

The envelope hamiltonian for electron interaction with ultrashort pulses

Jan Michael Rost

Max-Planck-Institute for the

Physics of Complex Systems, Dresden

Koudai Toyota (postdoc)

Qicheng Ning (predoc)

Ulf Saalmann

The situation…

- for electron dynamics with ultrashort pulses,
  essentially only numerical solution available

regime: $T \sim T_\nu > T_\omega$

T pulse length
T_ν electron period
T_ω optical period

observations: - one needs many eigenstates of H_0 to reproduce light induced shifts (length gauge)

Demekhin & Cederbaum, PRL 108, 253001 (12)

- many Fourier components contribute to represent ultra short pulse dynamics (Kramers-Henneberger frame)

Morales etal, PNAS 108, 16906 (11)

yet…

- *perturbative regime (despite $U_p >> E_b$)*
  in the sense that only a few photons exchanged with the target

An illustration…

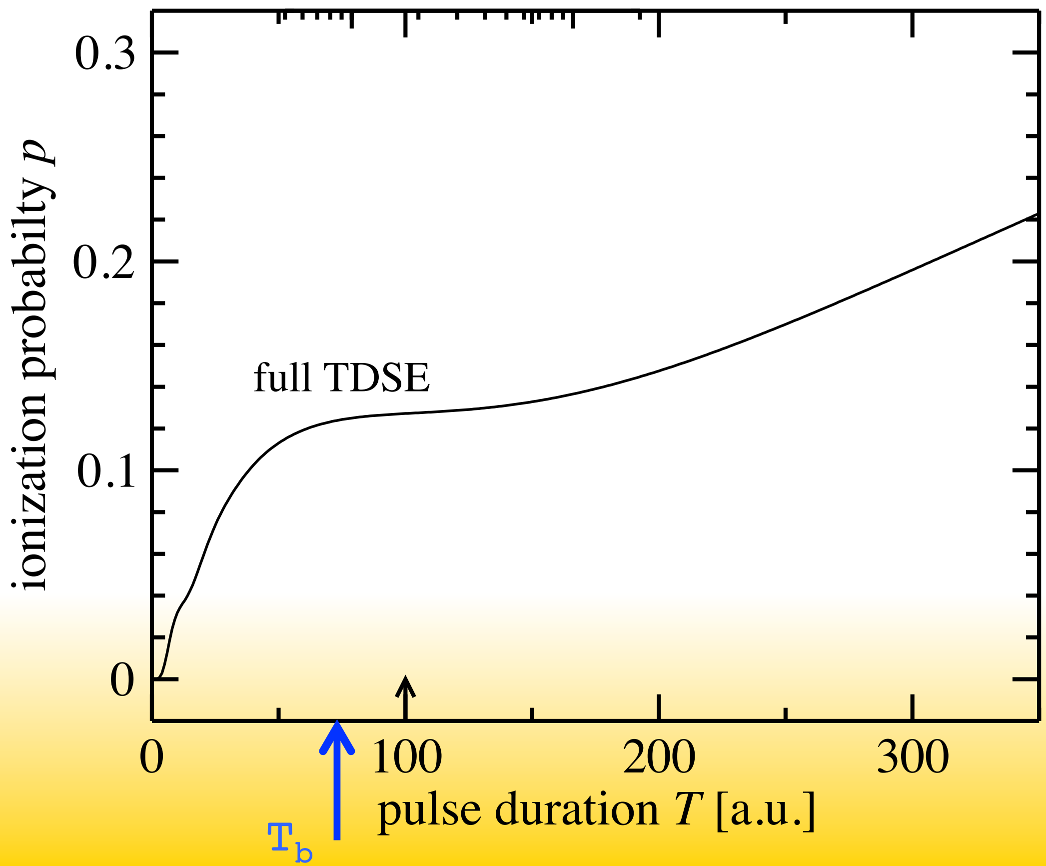
- Ionization probability for an atom as a function of pulse length ($F_0 = 0.5$, $\omega = 0.3$)

- 1D model
  for negative ion
  (short range potential)
  $E_b = -0.0277$

$$V(x) = -\frac{\exp[-c_1\sqrt{(x/c_1)^2 + c_2^2}]}{\sqrt{(x/c_1)^2 + c_3^2}}$$

T : variable
T_b = 70 au
T_ω = 20 au

$$T \sim T_\nu > T_\omega$$



Motivation.

T pulse length
T_ν electron period
T_ω optical period

- regime: $T \sim T_\nu > T_\omega$
- essentially perturbative regime
- *what are the perturbations and what is H_0 ?*
  can one isolate true n-photo absorption ?

The Hamiltonian and the light field

$$H = -\frac{1}{2}\nabla^2 + V\left(\mathbf{r} + \mathbf{e}_x\, x_\omega(t)\right),$$

$$F(t) = -\frac{d^2 x_\omega}{dt^2}$$

$$x_\omega(t) = \alpha(t)\cos(\omega t + \delta),$$

$$\alpha(t) \equiv \alpha_0\, \mathrm{e}^{-4\ln 2 (t/T)^2}.$$

characterize light field through
peak amplitude

$$F(0) = F_0 \cos\delta$$

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

$\alpha_0 \to 0$ for $\omega \to \infty$ and $T \to 0$

$\omega \to \infty$ is the standard high frequency adiabatic limit,
also valid in the "diabatic" limit of very short pulses ?

Realization of the parametric pulse shape dependence: *Single-cycle* Floquet representation

$$H_{n_{\max}}(t) = -\frac{1}{2}\nabla^2 + \sum_{n=-n_{\max}}^{+n_{\max}} V_n(\mathbf{r},t)\,\mathrm{e}^{-\mathrm{i}n\omega t}$$

$$V_n(\mathbf{r},t) = \frac{1}{T_\omega}\int_0^{T_\omega} \mathrm{d}t'\, V\big(\mathbf{r}+\mathbf{e}_x\,\alpha(t)\cos(\omega t'+\delta)\big)\mathrm{e}^{\mathrm{i}n\omega t'}$$

- $V_n(\mathbf{r},t)$, n =±1,±2,… are the interactions/perturbations which lead to 1,2,… photon emission and absorption
- $H_0(t)$ contains (envelope) dependent dynamics (e.g., the light shifted bound state) $H_0(t) = -\frac{1}{2}\nabla^2 + V_0(\mathbf{r},t)$
- formulate time-dependent perturbation theory, where the basis |β(t)> (solutions to $H_0(t)$) is also time-dependent.

Realization of the parametric pulse shape dependence: *Single-cycle* Floquet representation

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✓ light induced shifts in H_0

✓ pulse shape dependence separated (eliminates high Fourier components in KH)

? what is the minimal n_{max} needed ?

- V_n(**r**,t), n =±1,±2,… are the interactions/perturbations which lead to 1,2,… photon emission and absorption
- H_0(t) contains (envelope) dependent dynamics (e.g., the light shifted bound state)

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

- formulate time-dependent perturbation theory, where the basis |β(t)> (solutions to H_0(t)) is also time-dependent.

The time dependent basis and coupled equations

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

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

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

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

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

$$\sum\!\!\!\!\!\!\!\int |\beta(t)\rangle\langle\beta(t)| = 1$$

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

- For continuum states $|\beta\rangle = \mathbf{k}$:
  $E_{\mathbf{k}}(t) = \epsilon_{\mathbf{k}}$
- for bound states $|\beta\rangle = |j\rangle$:
  $E_j(t) = t^{-1}\int^t dt'\,\epsilon_j(t')$
- phase freedom $\chi(t)$ is time depenent

Representing the dynamics of H_{n_max} in the basis of H_0

$$H_{n_{\max}}(t) = -\frac{1}{2}\boldsymbol{\nabla}^2 + \sum_{n=-n_{\max}}^{+n_{\max}} V_n(\mathbf{r},t)\,\mathrm{e}^{-\mathrm{i}n\omega t}$$

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

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

Ionization probability:

$$\frac{dP}{d\mathbf{k}} = \lim_{t\to\infty} |c_{\mathbf{k}}^{(1)}(t)|^2$$
$$= \left|\sum_{n=-n_{max}}^{+n_{max}} \lim_{t\to\infty} M_n(\mathbf{k},t)\right|^2$$

$$M_n(\mathbf{k},t) = -\mathrm{i}\int_{-\infty}^{t} \mathrm{d}t' \left\langle \mathbf{k},t' \middle| V_n(x,t') \middle| \mathrm{b}(t') \right\rangle \mathrm{e}^{\mathrm{i}\phi_n(t')}$$ n-photon ionization

$$M_0(\mathbf{k},t) = -\int_{-\infty}^{t} \mathrm{d}t' \left\langle \mathbf{k},t' \middle| \frac{\partial}{\partial t'} \middle| \mathrm{b}(t') \right\rangle \mathrm{e}^{\mathrm{i}\phi_0(t')}$$ non-adiabatic ionization

$$\phi_n(t) = [k^2/2 - E_{\mathrm{b}}(t) - n\omega]t$$

The *pulse length dependent* ionization probability for an atom ($F_0 = 0.5$, $\omega = 0.3$)

$$\frac{dP}{d\mathbf{k}} = \left| \sum_{n=-2}^{2} \lim_{t\to\infty} M_{\mathbf{k}}^{(n)}(t) \right|^2$$

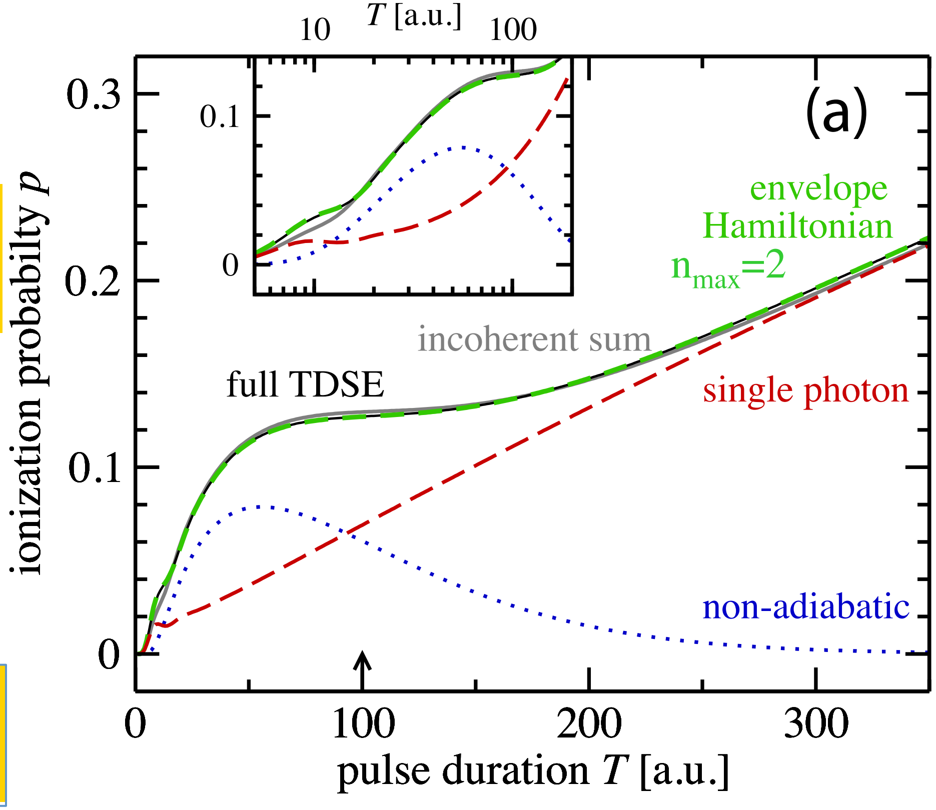
- why is the incoherent sum so accurate ?

incoherent sum:

$$\frac{dP}{dk} \approx \left|M_k^{(0)}\right|^2 + \left|M_k^{(1)}\right|^2$$

$$M_{\mathbf{k}}^{(0)}(t) = \int_{-\infty}^{t} \mathrm{d}t' \left\langle \mathbf{k}, t' \middle| \frac{\partial}{\partial t'} \middle| \mathrm{b}(t') \right\rangle e^{-\mathrm{i}\phi_0(t')}$$

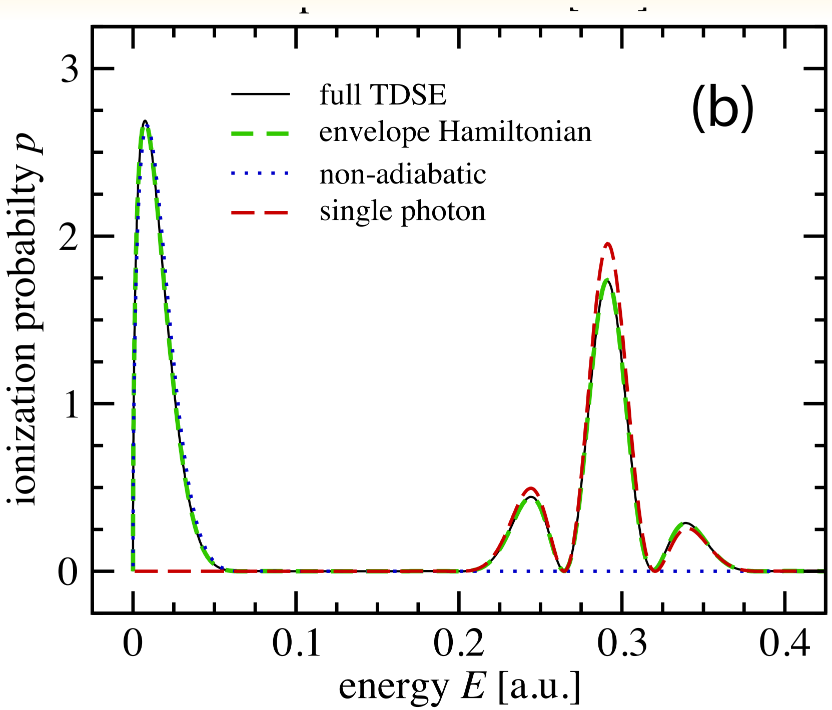
$$M_{\mathbf{k}}^{(n)}(t) = -i \int_{-\infty}^{t} \mathrm{d}t' \left\langle \mathbf{k}, t' \middle| V_n(x, t') \middle| \mathrm{b}(t') \right\rangle e^{-\mathrm{i}\phi_n(t')}$$



$$\phi_n(t) = [k^2/2 - E_\mathrm{b}(t) - n\omega]t$$

The photo electron spectrum

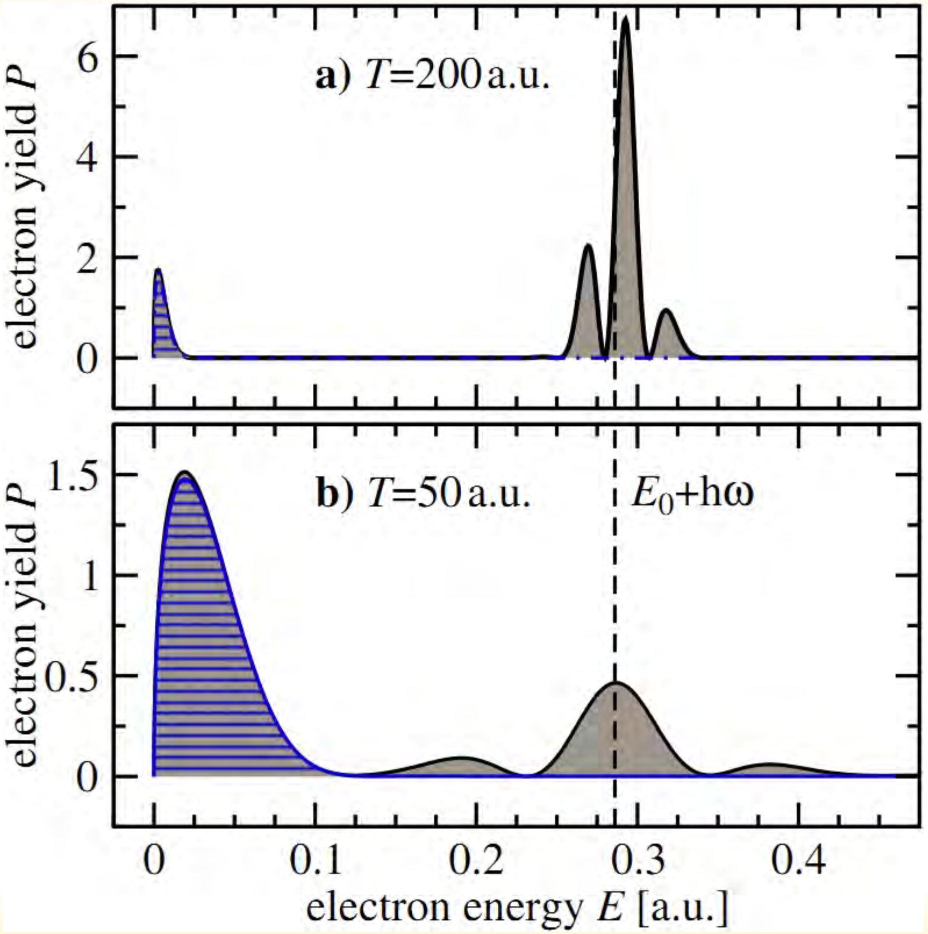
$$\frac{dP}{d\mathbf{k}} = \Big|\sum_{n=-2}^{2} \lim_{t\to\infty} M_{\mathbf{k}}^{(n)}(t)\Big|^2$$



- incoherent superposition due to very different energies for
- *non-adiabatic* electrons *(E~0)* and
- *single photon ionization (E~ $E_b+\omega$)*

- *only for very short pulses overlap…*

Width of peaks for small T make contributions overlap

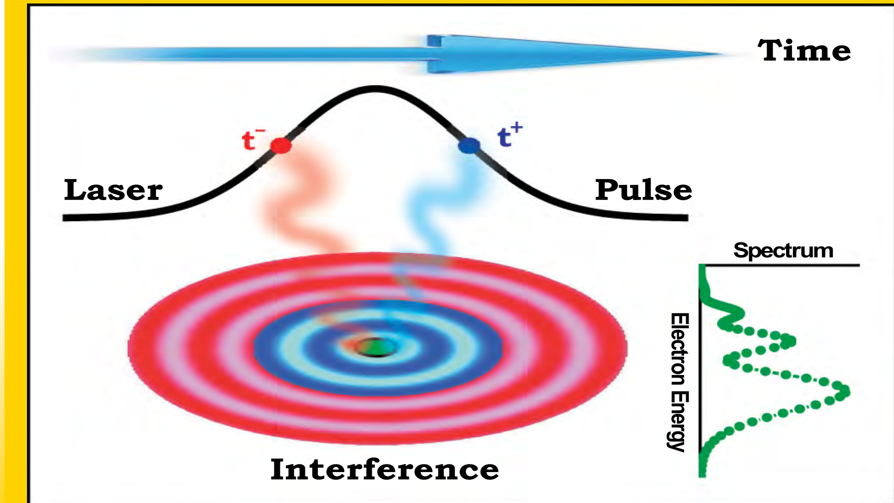


- interference of two main contributions, when final energy coincides with dressed light induced bound state energy

  Demekhin & Cederbaum, PRL 108, 253001 (12)

  Koudai etal, PRA 78, 033432, (08)

  light induced blue shift (AC Stark shift, intensity dependent)



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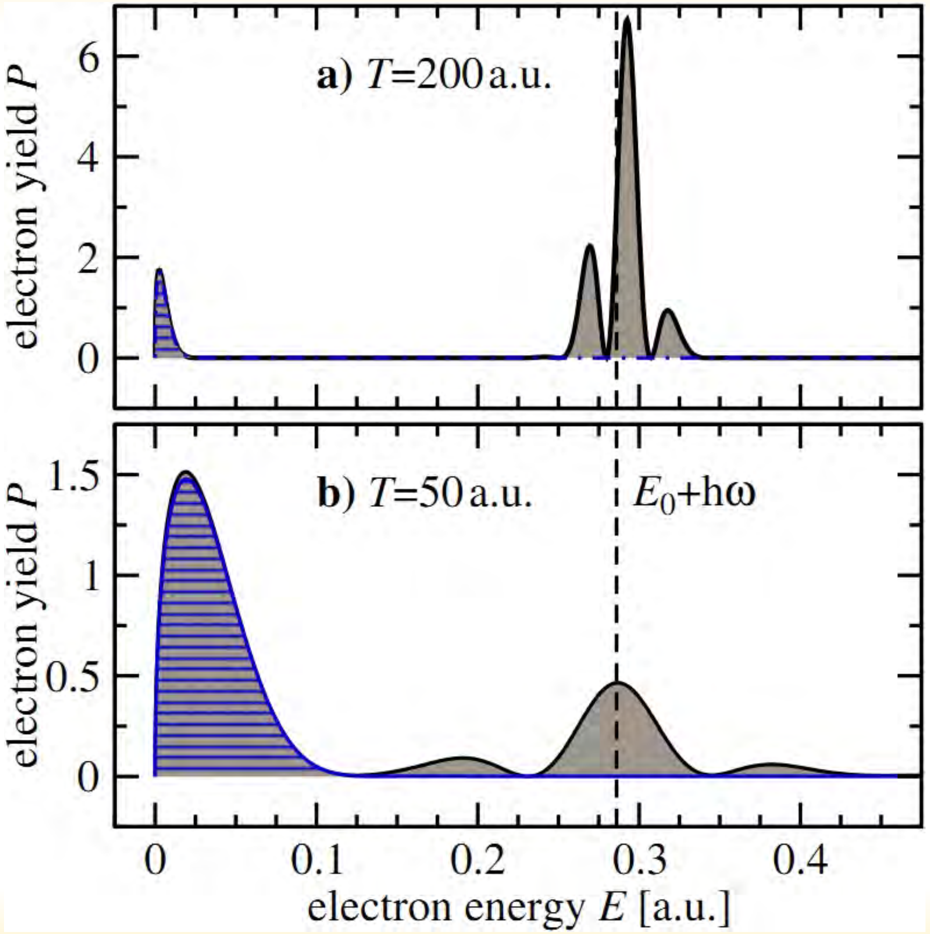

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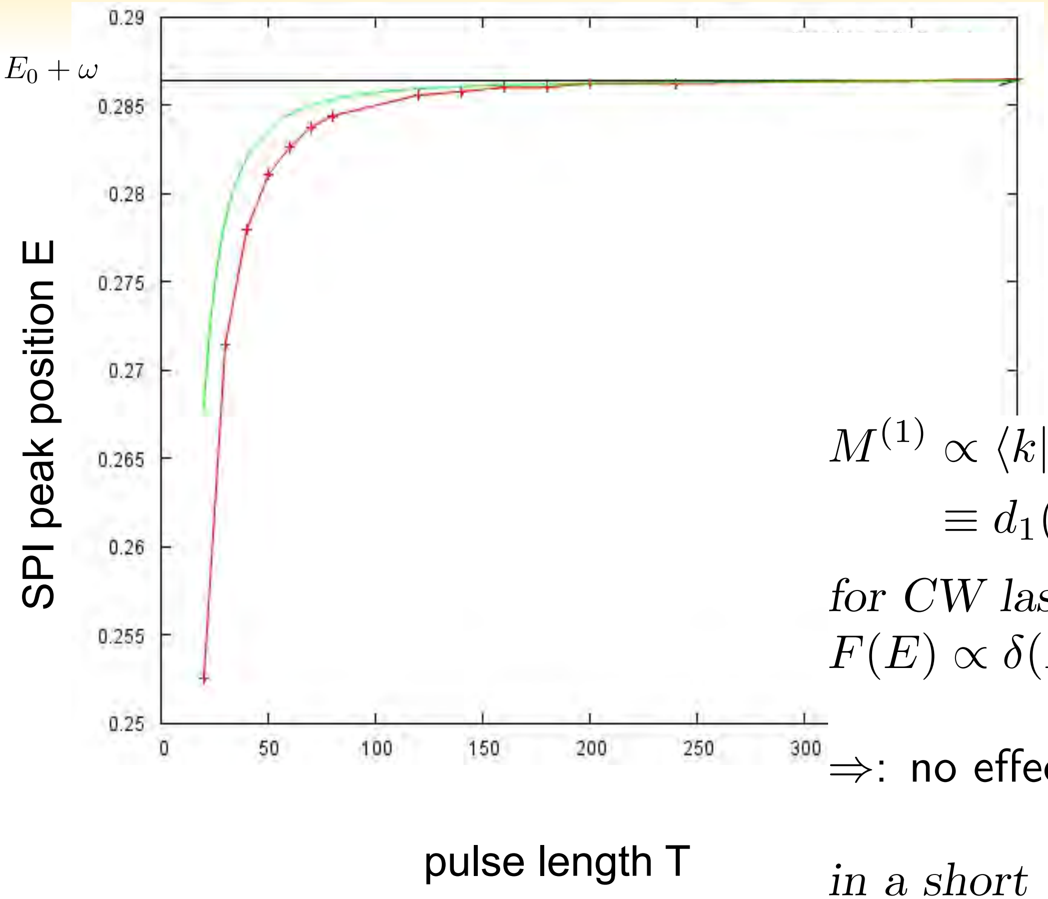
- **NEW:** pulse length dependent red shift

Pulse dependence of red shift



$$E \approx E_0 + \omega - \frac{2}{T^2} \frac{1}{E_0 + \omega}.$$

numerical

$$M^{(1)} \propto \langle k | V_1(\mathbf{r}, t) | b(t) \rangle F(k^2/2 - \omega - E_b(t))$$
$$\equiv d_1(E) F(E)$$

for CW lasers:
$F(E) \propto \delta(E - \omega - \epsilon_b)$

$\Rightarrow$: no effect of $d_1(E)$.

in a short pulse:
$F(E)$ has a broad peak and $d_1(E)$ decreases

$\Rightarrow$: redshift of the peak, should vanish for $T \to \infty$

Why does the envelope hamiltonian H_2 work for short pulses ?

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

$$H_{\rm env}(t) \equiv H_0(t) + \sum_{n=\pm1,\pm2} V_n(\mathbf{r},t)\, \mathrm{e}^{-\mathrm{i}n\omega t}$$

$$H_0(t) \equiv -\frac{1}{2}\boldsymbol{\nabla}^2 + V_0(\mathbf{r},t)\,,$$

expand the single period avaraged potentials $V_n(\mathbf{x},t)$ to second order in α_0:

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

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

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

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

$$\approx V(\mathbf{x} + \mathbf{e}_x\alpha(t)\cos(\omega t + \delta))$$

Why does the envelope hamiltonian H_2 work for short pulses ?

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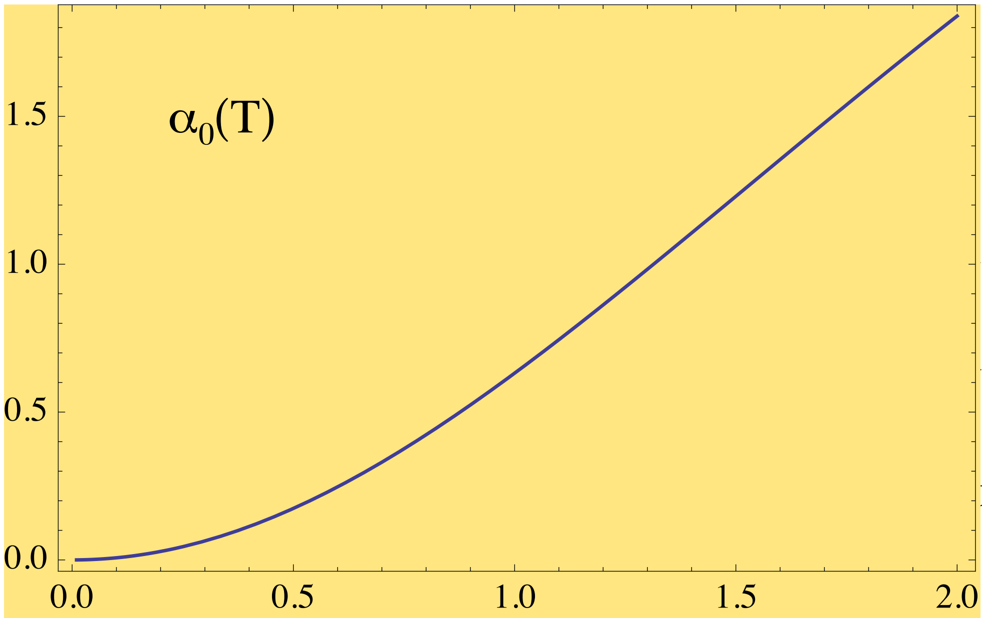


$V_n(\mathbf{x},t)$ to second order in α_0:

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

$)\mathrm{e}^{\mp i\delta},$

$\alpha^2(t)\mathrm{e}^{\mp 2i\delta}\,.$

$\delta) + \frac{1}{2}\frac{\partial^2 V}{\partial x^2}\alpha^2(t)\cos(\omega t+\delta)^2$

$))$

- $\alpha_0 < 1$ only for T < 1.5
- why is the envelope hamitonian so accurate for all pulse lengths T ?

View *envelope hamiltonian* as an
expansion in the number of photons
with a parametric pulse shape dependence

$$H_{\mathrm{env}}(t) \equiv H_0(t) + \sum_{n=\pm1,\pm2,\pm3,\pm4,\ldots} V_n(\mathbf{r},t)\,\mathrm{e}^{-\mathrm{i}n\omega t}$$

$$H_0(t) \equiv -\frac{1}{2}\nabla^2 + V_0(\mathbf{r},t)\,,$$

■ if the first photon puts the electron in the continuum, only non-adiabatic transitions and single photon ionization occurs…

n-photon ionization rates during the laser pulse ?

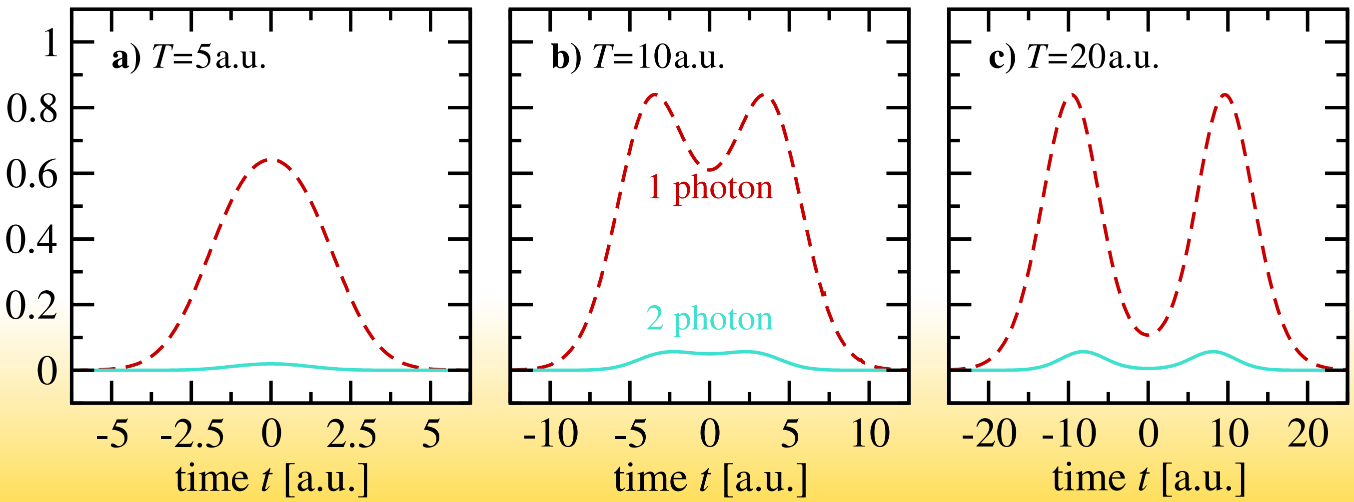
$$\frac{dP}{d\mathbf{k}} = \lim_{t\to\infty} |c_{\mathbf{k}}^{(1)}(t)|^2$$
$$= \left|\sum_{n=-n_{max}}^{+n_{max}} \lim_{t\to\infty} M_n(\mathbf{k},t)\right|^2$$

- just take $M_n(k,t)$ at times t during the pulse **NO!!!**
- only valid after the pulse is over: basis is time/pulse dependent and ionization not well defined during the pulse

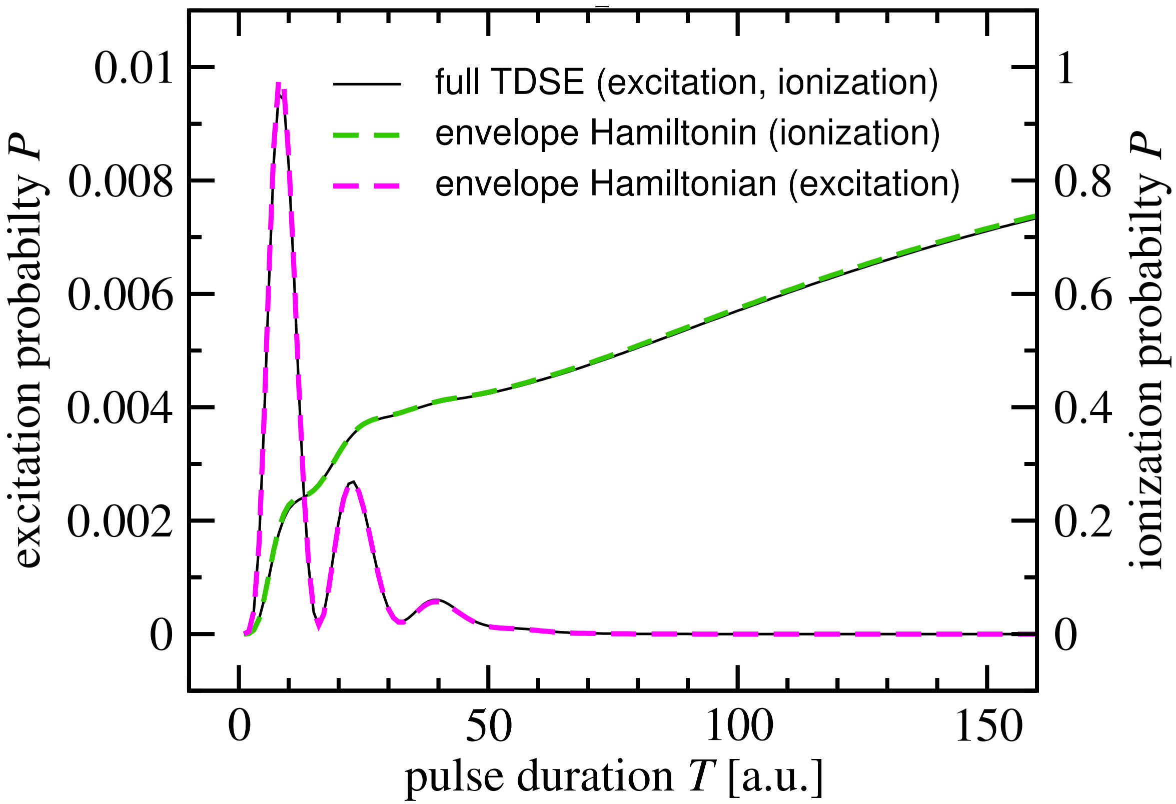
n-photon ionization rates during the laser pulse ?

… but we can define *cycle averaged* ionization rates during the pulse

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



For another potential....



$$W_g = -U_0 \exp(-(x/\sigma)^2) \text{ with } U_0 = 0.2 \text{ and } \sigma = 2.65$$

Summary

- ***The envelope hamiltonian*** separates the effect of the pulse envelope and (multiple) photo absorption
  - approximate, yet very accurate for realistic pulses
  - opens attosecond dynamics to perturbation theory
  - allows insight into the dynamics through separation of
    - *0^{th} order dynamics:* averaged KH-potential
      envelope dependent, includes naturally light shifts
    - *n-photon absorption*
      with so far not studied *single-cycle-averaged* interactions potentials $V_n(\mathbf{r},t)$

- Not only for atto-pulses,
  but whenever $T \sim T_\nu$, e.g. Femto pulses+ Rydberg systems

Thanks !