Time dependent models for strong field physics

Workshop
Model Systems in Quantum Mechanics

January 11th–12th 2024

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Laboratoire PASTEUR

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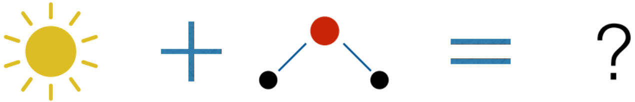
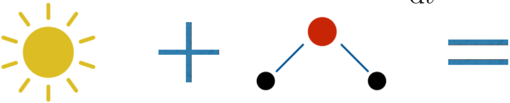
Simulation of strong-field–matter interaction



Simulation of strong-field–matter interaction

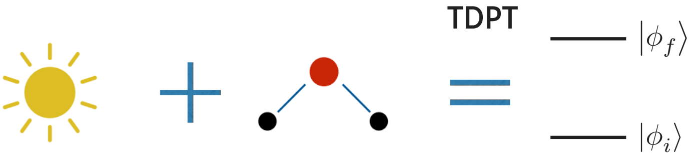
$$i\frac{\mathrm{d}\left|\Psi\right\rangle}{\mathrm{d}t} = H\left|\Psi\right\rangle$$



$$\left|\Psi(t)\right\rangle$$

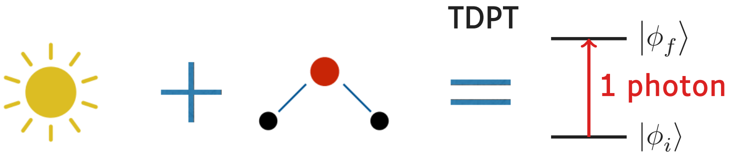
- Time-dependent Schrödinger Equation (hard in general)

Simulation of strong-field–matter interaction



- Time-dependent Schrödinger Equation (hard in general)
- Time Dependent Perturbation Theory $H = H_0 - \vec{D} \cdot \vec{F}(t)$

Simulation of strong-field–matter interaction



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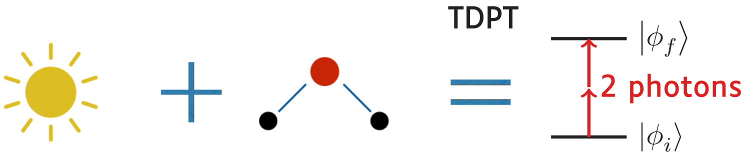

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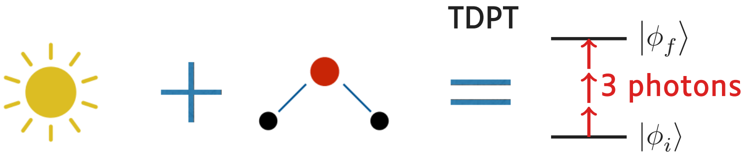

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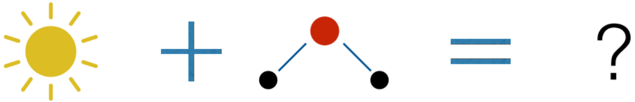
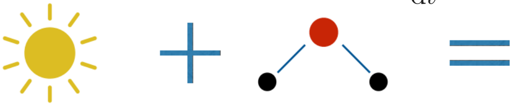
Simulation of strong-field–matter interaction



- Time-dependent Schrödinger Equation (hard in general)
- Time Dependent Perturbation Theory $H = H_0 - \vec{D} \cdot \vec{F}(t)$
- What about strong field?

Simulation of strong-field–matter interaction

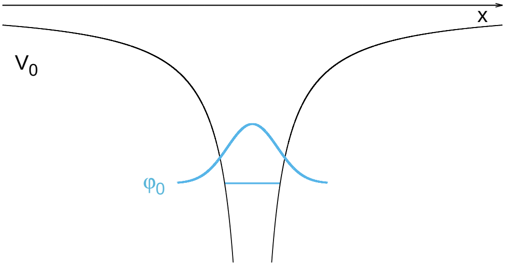
$$i\frac{\mathrm{d}\left|\Psi\right\rangle}{\mathrm{d}t} = H\left|\Psi\right\rangle$$



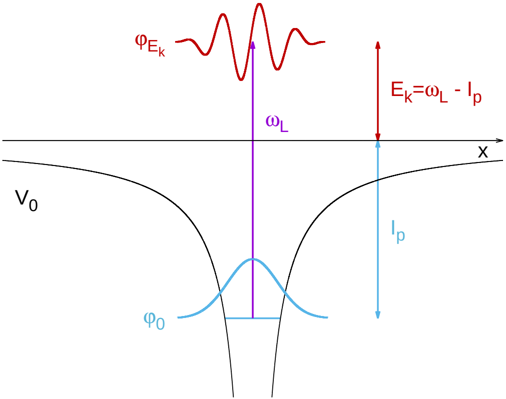
$$\left|\Psi(t)\right\rangle$$

- Time-dependent Schrödinger Equation (hard in general)
- Time Dependent Perturbation Theory $\quad H = H_0 - \vec{D}\cdot\vec{F}(t)$
- What about strong field? Model systems!

Photoionization



one photon absorption



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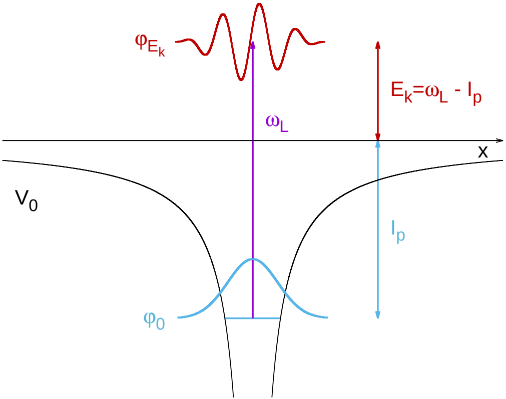
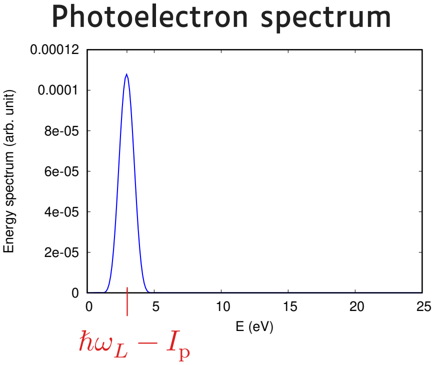
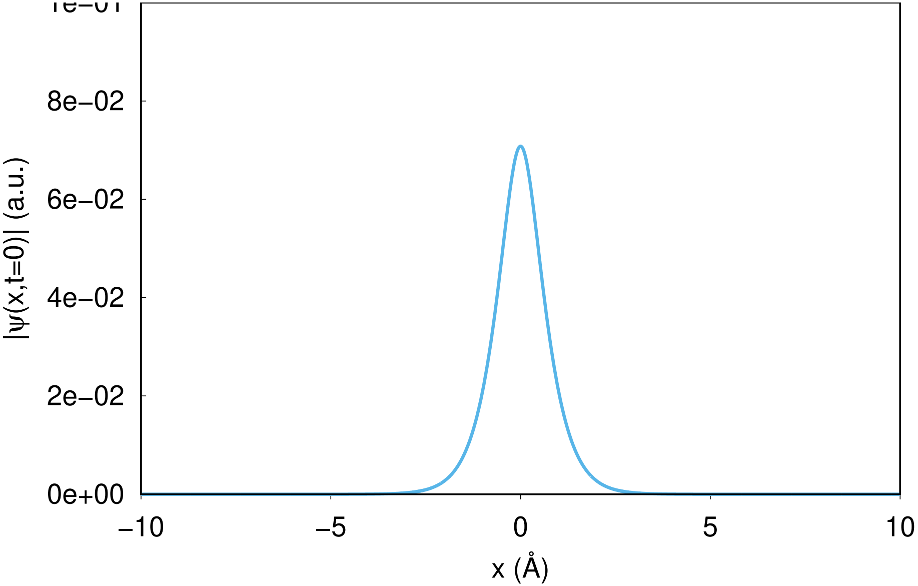
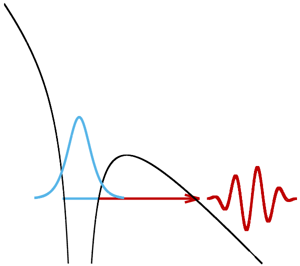





1e−01
8e−02
6e−02
4e−02
2e−02
0e+00
|ψ(x,t=0)| (a.u.)
−10
−5
0
5
10
x (Å)

Three-step Model

1. Tunnel ionization



Three-step Model

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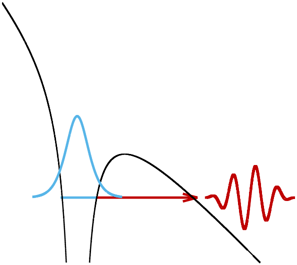
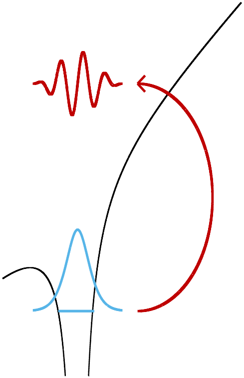
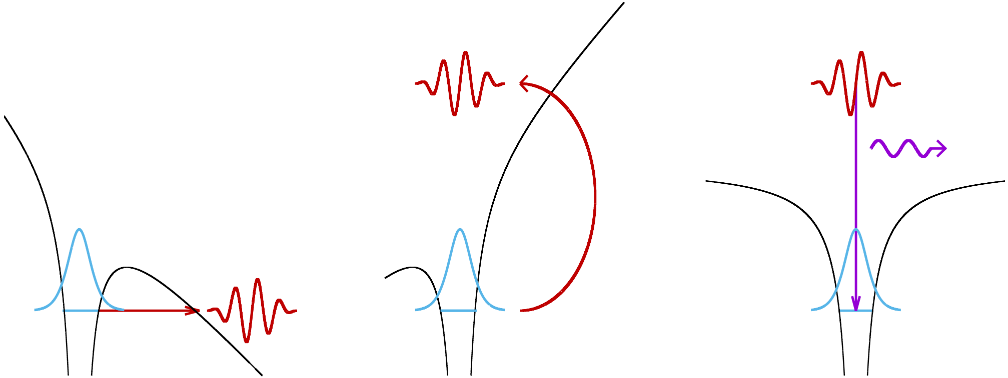


2. Propagation

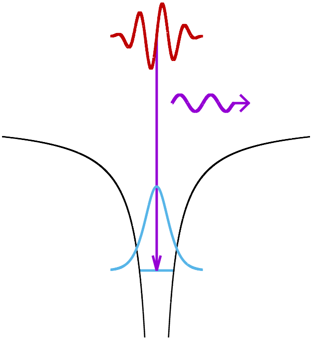


Three-step Model

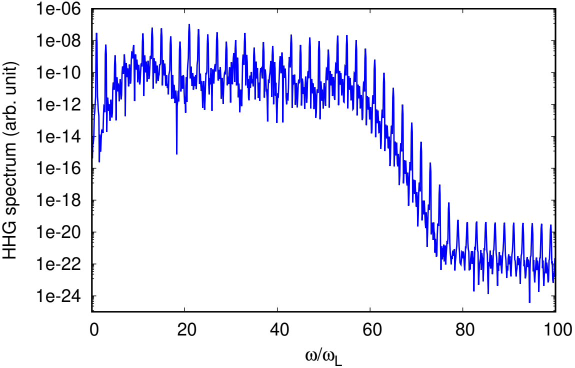
1. Tunnel ionization
2. Propagation
3. Recombination



3. Recombination



typical HHG spectrum



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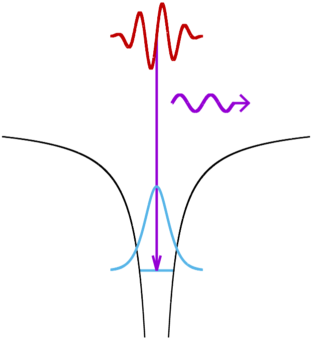

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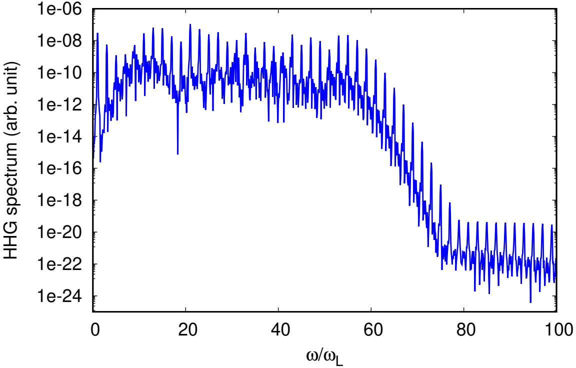
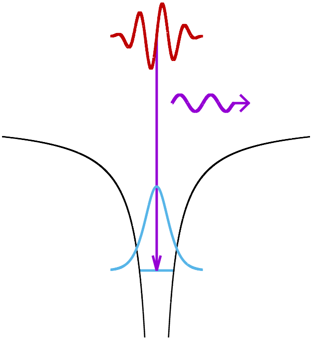


Incident 800nm IR Laser ∼1 eV → XUV emission (up to 1keV)

3. Recombination



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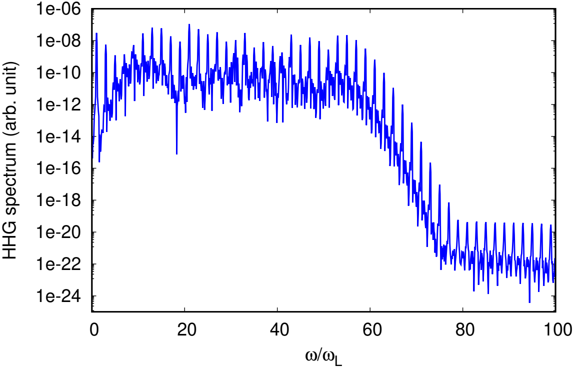

Incident 800nm IR Laser $\sim$1 eV $\rightarrow$ XUV emission (up to 1keV)

Coherent XUV light source!

Model systems for HHG

Analytical models

- Classical trajectories
- Lewenstein model

Analytical models

- Classical trajectories
- Lewenstein model

Numerical models

- Single Active Electron with pseudo-potential
- Low-dimensional model
- TD quantum chemistry methods (TDCI, real-time TDDFT)

Classical trajectories

Hypotheses

- After tunnel ionization, a classical free electron is "born"
  - with zero initial velocity
  - at nucleus position
  - it is "ionized" $\rightarrow$ does not feel the potential
- This classical electron oscillates in the field $F(t) = -F_0 \cos(\omega_\mathrm{L} t)$
- The ionization can happen at anytime t_i
- The field has a constant amplitude

Equation of motion

$$m_e \ddot{x}(t) = eF_0 \cos(\omega_\mathrm{L} t)$$

Classical trajectories

$$\dot{x}(t) = \frac{eF_0}{m_e \omega_{\mathrm{L}}} \left[\sin(\omega_{\mathrm{L}} t) - \sin(\omega_{\mathrm{L}} t_i) \right]$$

$$x(t) = \frac{eF_0}{m_e \omega_{\mathrm{L}}^2} \left[\cos(\omega_{\mathrm{L}} t) - \cos(\omega_{\mathrm{L}} t_i) - \omega_{\mathrm{L}} (t - t_i) \sin(\omega_{\mathrm{L}} t_i) \right]$$

Classical trajectories

$$\dot{x}(t) = \frac{eF_0}{m_e\omega_\mathrm{L}}\left[\sin(\omega_\mathrm{L}t) - \sin(\omega_\mathrm{L}t_i)\right]$$

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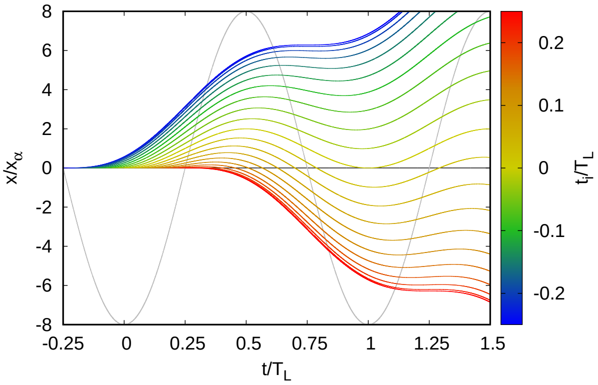
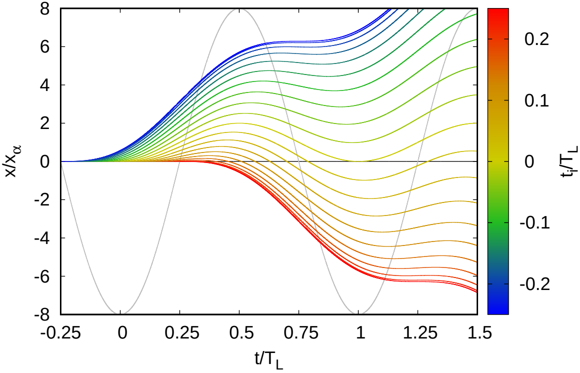


Classical trajectories

Remarks

- Only some trajectories return to the nucleus after ionization

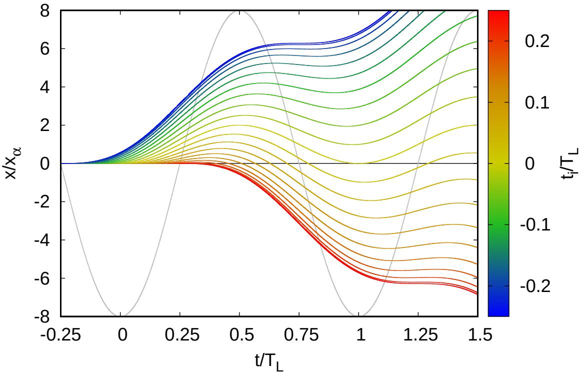


Classical trajectories

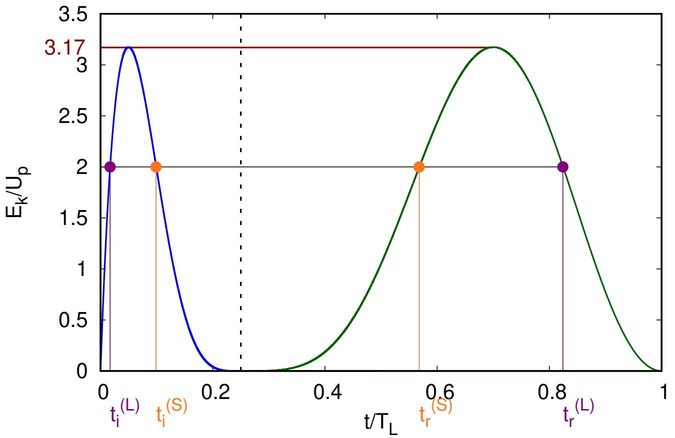
Remarks

- Only some trajectories return to the nucleus after ionization
- Maximal excursion length

$$2x_\alpha = \frac{2eF_0}{m_e\omega_L^2}$$



Kinetic Energy at return



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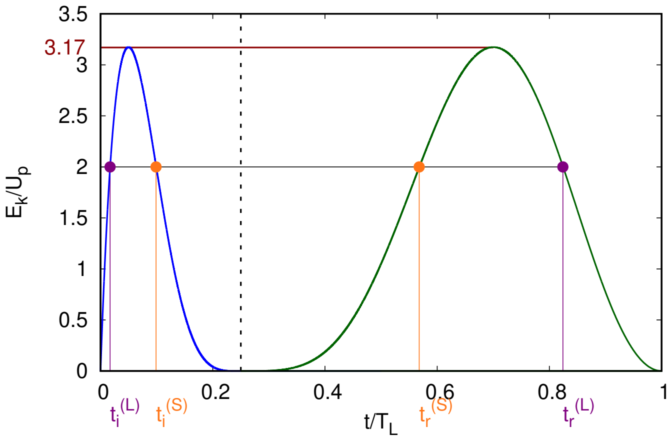
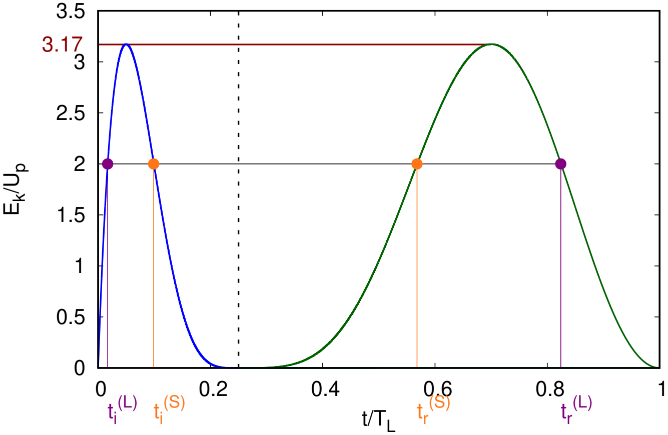


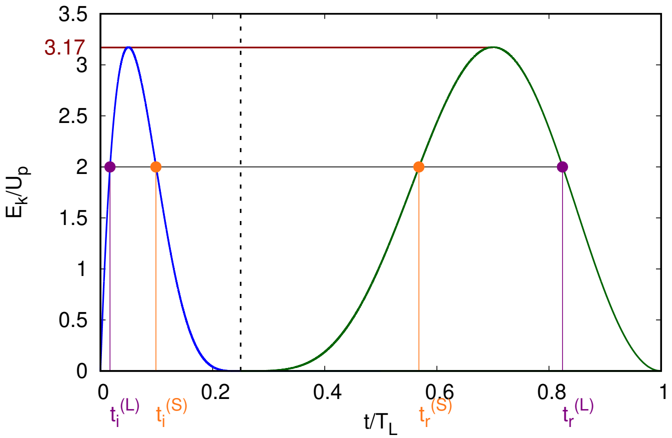
- Kinetic energy at return $\in [0, 3.17U_\mathrm{p}]$

Kinetic Energy at return



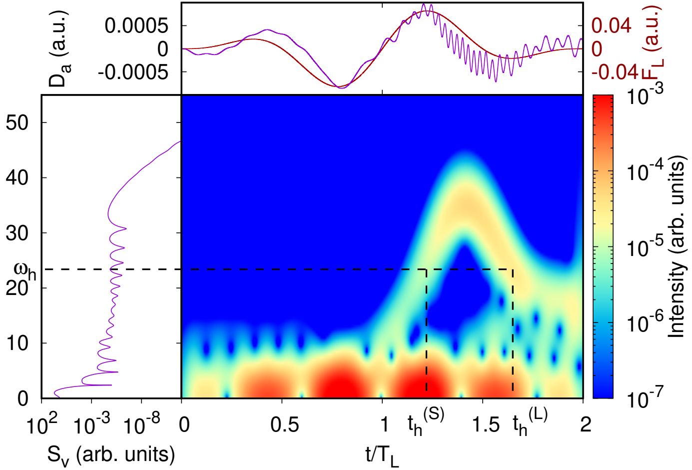
- Kinetic energy at return $\in [0, 3.17U_\mathrm{p}]$
- Two classes of trajectories: short and long

Kinetic Energy at return



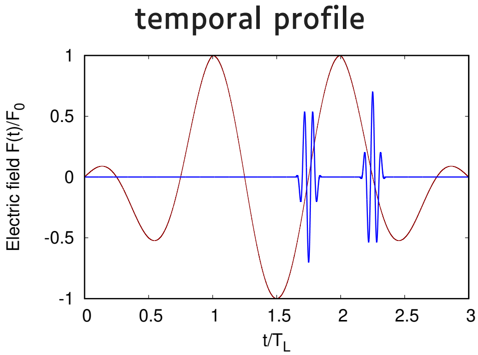
- Kinetic energy at return $\in [0, 3.17U_{\mathrm{p}}]$
- Two classes of trajectories: short and long
- In the quantum world, these "trajectories" can "interfere"

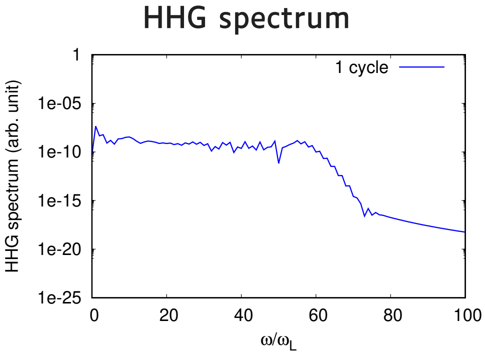
Time-Frequency Gabor transform of HHG emission signal



Harmonics?

1 generating laser cycle

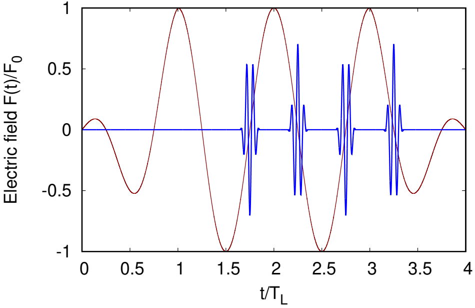




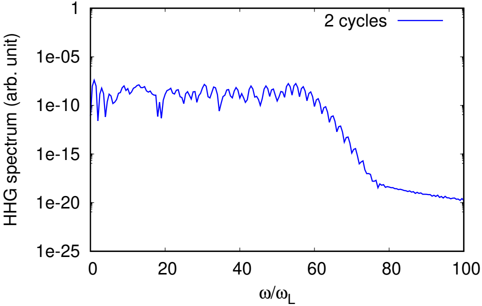
Harmonics?

2 generating laser cycle

temporal profile



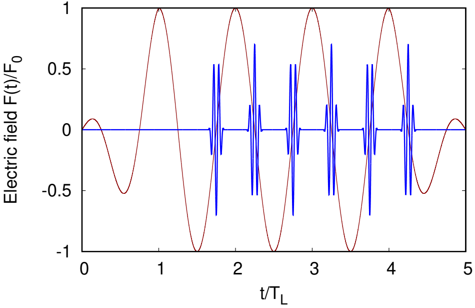
HHG spectrum



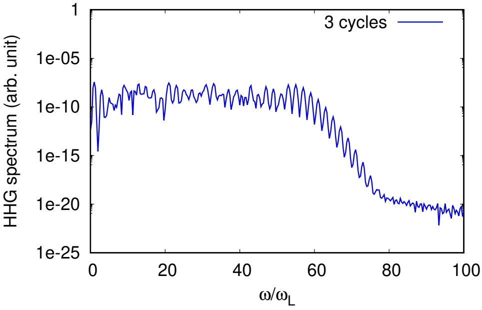
Harmonics?

3 generating laser cycle

temporal profile

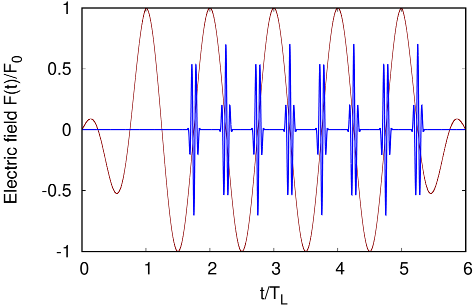


HHG spectrum

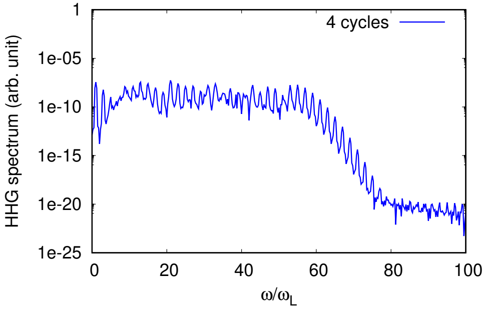


4 generating laser cycle

temporal profile

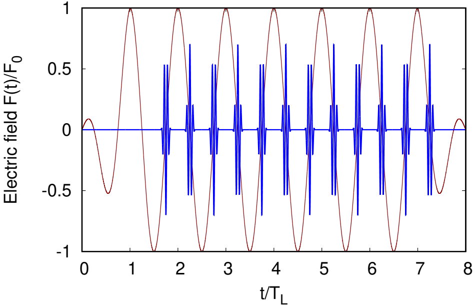


HHG spectrum

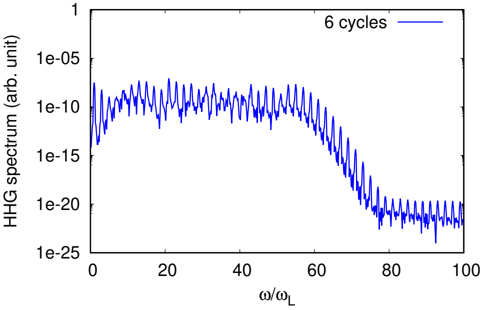


6 generating laser cycle

temporal profile

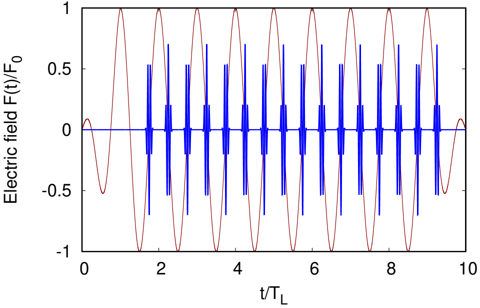


HHG spectrum

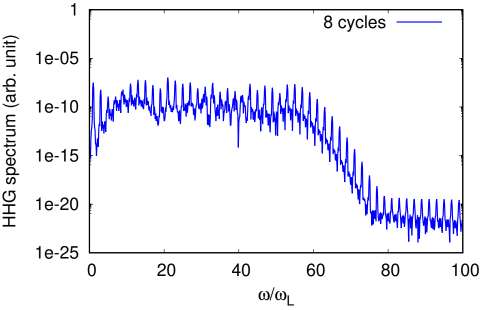


8 generating laser cycle

temporal profile

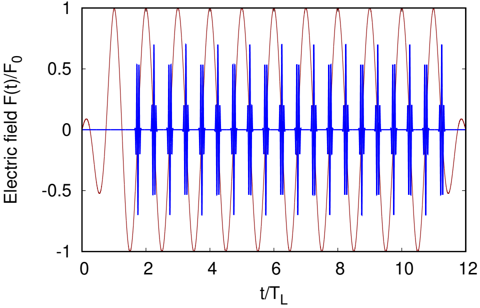


HHG spectrum

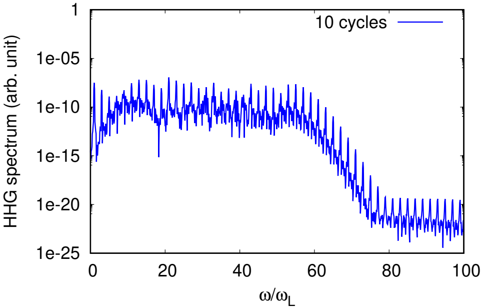


10 generating laser cycle

temporal profile



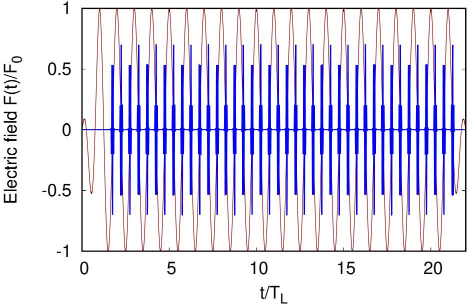
HHG spectrum



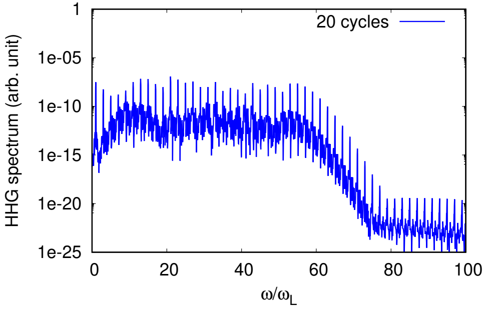
Harmonics?

20 generating laser cycle

temporal profile



HHG spectrum



Lewenstein Model

PHYSICAL REVIEW A VOLUME 49, NUMBER 3 MARCH 1994

Theory of high-harmonic generation by low-frequency laser fields

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(Received 19 August 1993)

We present a simple, analytic, and fully quantum theory of high-harmonic generation by low-frequency laser fields. The theory recovers the classical interpretation of Kulander *et al.* in [*Proceedings of the SILAP III Workshop*, edited by B. Piraux (Plenum, New York, 1993)] and Corkum [Phys. Rev. Lett. **71**, 1994 (1993)] and clearly explains why the single-atom harmonic-generation spectra fall off at an energy approximately equal to the ionization energy plus about three times the oscillation energy of a free electron in the field. The theory is valid for arbitrary atomic potentials and can be generalized to describe laser fields of arbitrary ellipticity and spectrum. We discuss the role of atomic dipole matrix elements, electron rescattering processes, and of depletion of the ground state. We present the exact quantum-mechanical formula for the harmonic cutoff that differs from the phenomenological law $I_p + 3.17U_p$ where I_p is the atomic ionization potential and U_p is the ponderomotive energy, due to the account for quantum tunneling and diffusion effects.

Hypotheses

- Single Active Electron
- Bound states contributions are neglected
- Ground state depletion is neglected
- Plane Wave Approximation for continuum states

HHG emission dipole

$$\mathbf{d}(\omega) = \int \mathrm{d}t_r \int_0^{t_r} \mathrm{d}t_i \int \mathrm{d}\mathbf{p}\, \mathbf{d}_{\mathrm{rec}}(\mathbf{p}+\mathbf{A}(t_r))d_{\mathrm{ion}}(\mathbf{p}+\mathbf{A}(t_i), t_i)\, \mathrm{e}^{-i[S(\mathbf{p},t_r,t_i)-\omega t_r]} + \tilde{c}c.$$

$$S(\mathbf{p}, t_r, t_i) = \int_{t_i}^{t_r} \mathrm{d}\tau \left(\frac{[\mathbf{p} + \mathbf{A}(\tau)]^2}{2} + I_{\mathrm{p}} \right)$$

$$d_{\mathrm{ion}}(\mathbf{k}) = \langle \mathbf{k} | - \mathbf{D} \cdot \mathbf{F}(t) | \phi_0 \rangle$$

$$\mathbf{d}_{\mathrm{rec}}(\mathbf{k}) = \langle \phi_0 | \mathbf{D} | \mathbf{k} \rangle$$

Compute the integral with Saddle Point Approximation:
$\nabla(S(\mathbf{p}, t_r, t_i) - \omega t_r) = 0$

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$\nabla(S(\mathbf{p}, t_r, t_i) - \omega t_r) = 0$

$$\begin{aligned}
\int_{t_i}^{t_r} [\mathbf{p} + \mathbf{A}(\tau)]\,\mathrm{d}\tau &= 0 \\
\frac{[\mathbf{p} + \mathbf{A}(t_r)]^2}{2} + I_\mathrm{p} - \omega &= 0 \\
\frac{[\mathbf{p} + \mathbf{A}(t_i)]^2}{2} + I_\mathrm{p} &= 0
\end{aligned}$$

Compute the integral with Saddle Point Approximation:

$\nabla(S(\mathbf{p}, t_r, t_i) - \omega t_r) = 0$

$$\int_{t_i}^{t_r} [\mathbf{p} + \mathbf{A}(\tau)]\, \mathrm{d}\tau = 0$$

classical trajectory with $\mathbf{r}(t_i) = \mathbf{r}(t_r)$

$$\frac{[\mathbf{p} + \mathbf{A}(t_r)]^2}{2} + I_\mathrm{p} - \omega = 0$$

$$\frac{[\mathbf{p} + \mathbf{A}(t_i)]^2}{2} + I_\mathrm{p} = 0$$

Compute the integral with Saddle Point Approximation:

$\nabla(S(\mathbf{p}, t_r, t_i) - \omega t_r) = 0$

$$\int_{t_i}^{t_r} [\mathbf{p} + \mathbf{A}(\tau)] \, \mathrm{d}\tau = 0$$

classical trajectory with $\mathbf{r}(t_i) = \mathbf{r}(t_r)$

$$\frac{[\mathbf{p} + \mathbf{A}(t_r)]^2}{2} + I_\mathrm{p} - \omega = 0$$

energy conservation at recombination

$$\frac{[\mathbf{p} + \mathbf{A}(t_i)]^2}{2} + I_\mathrm{p} = 0$$

Compute the integral with Saddle Point Approximation:
$\nabla(S(\mathbf{p}, t_r, t_i) - \omega t_r) = 0$

$$\int_{t_i}^{t_r} [\mathbf{p} + \mathbf{A}(\tau)]\,\mathrm{d}\tau = 0$$

classical trajectory with $\mathbf{r}(t_i) = \mathbf{r}(t_r)$

$$\frac{[\mathbf{p} + \mathbf{A}(t_r)]^2}{2} + I_\mathrm{p} - \omega = 0$$

energy conservation at recombination

$$\frac{[\mathbf{p} + \mathbf{A}(t_i)]^2}{2} + I_\mathrm{p} = 0$$

energy conservation at ionization

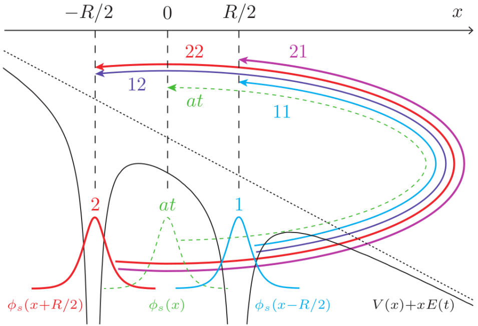
Low-dimensional Models

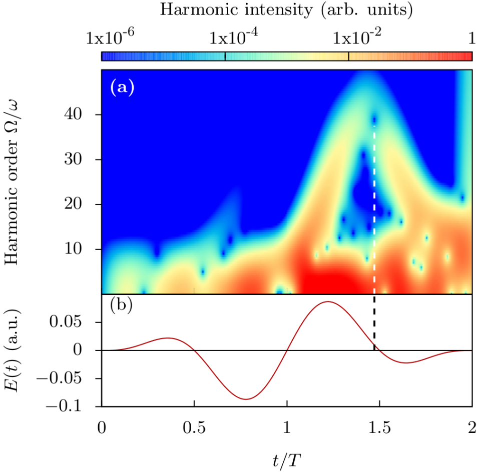
1D models

1D electron on a grid

- Soft-Coulomb potential $V(x) = -\frac{Z}{\sqrt{a^2 + x^2}}$ with $Z = 1$ for (neutral) atoms
- Molecular Soft-Coulomb $V(x) = -\frac{Z_1}{\sqrt{a_1^2 + (x - R_0/2)^2}} - \frac{Z_2}{\sqrt{a_2^2 + (x + R_0/2)^2}}$
  with $Z_1 + Z_2 = 1$
- Adjust regularization parameter a to match ionization potential
- 2nd-order Laplacian approximation $\rightarrow$ Tridiagonal Hamiltonian
- Crank-Nicolson algorithm
  $$\left[1 + iH\left(t + \frac{\Delta t}{2}\right)\frac{\Delta t}{2}\right]|\psi(t + \Delta t)\rangle = \left[1 - iH\left(t + \frac{\Delta t}{2}\right)\frac{\Delta t}{2}\right]|\psi(t)\rangle + O(\Delta t^3)$$

Molecules act like Young's two slits!





[Labeye et al., PRA (2019)]

2D Models

- **1 electron in 2D** $V(x,y) = -\frac{Z}{\sqrt{a^2 + x^2 + y^2}}$

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2D Models

Versatile!

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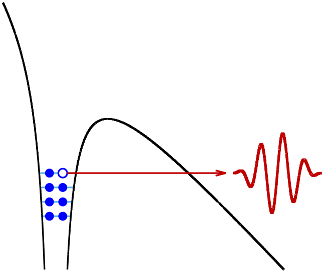
Multielectronic models

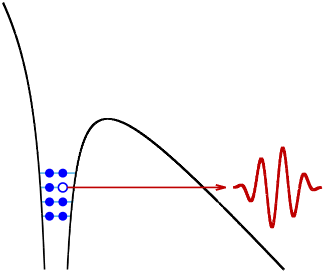
Limits of the Single Active Electron approximation

Ground state of the ion



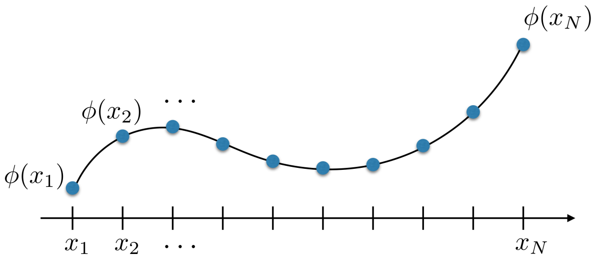
+

Excited state of the ion



Choice of the Basis

- **Grid:**
  - Good representation of the bound states
  - Good representation of the continuum states
  - Costly! (ok for low dimensional systems)



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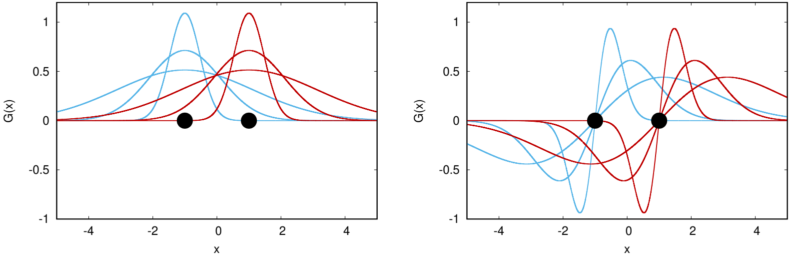

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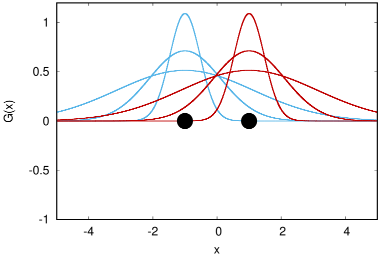
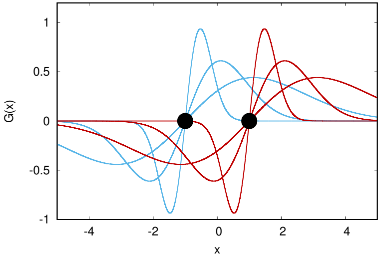
  - What about continuum states ? Kaufmann optimized gaussians [J.Phys.B (1989)]





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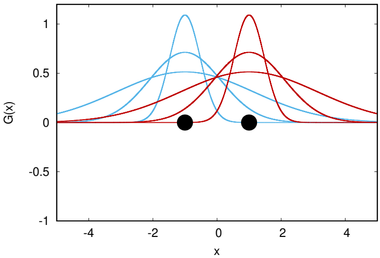
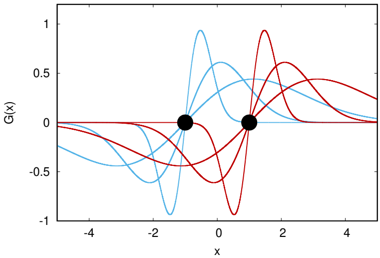
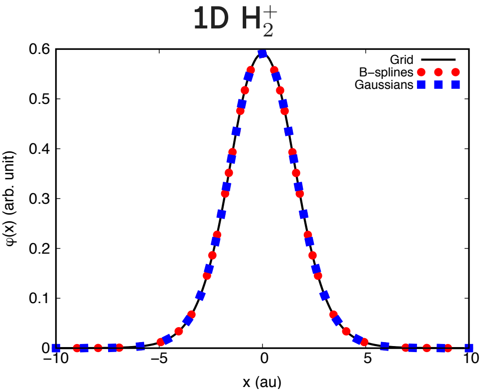
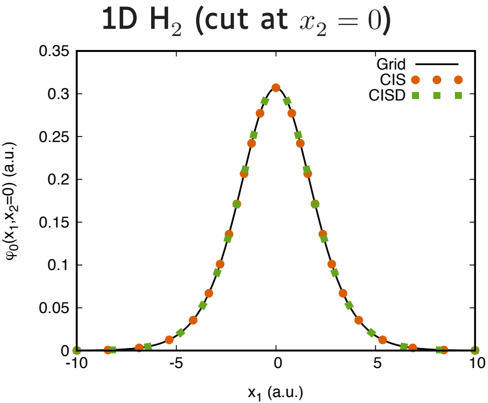




Low dimensional systems can be used to benchmark the Gaussian-based method!

Ground state





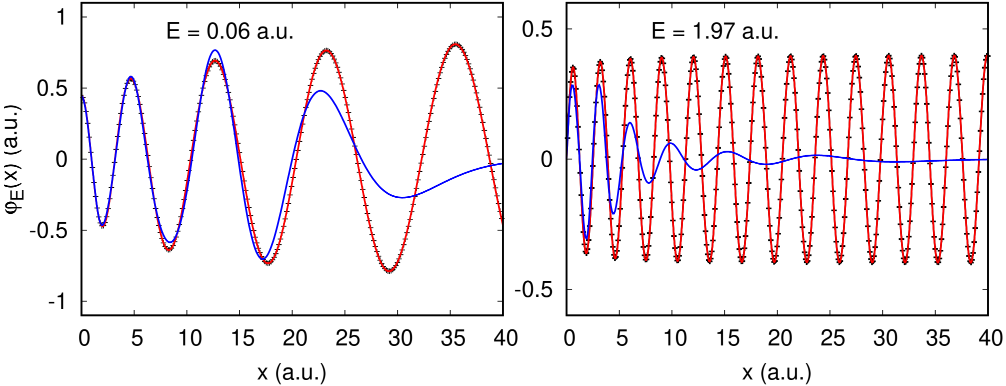
Good representation of the ground state

[Labeye et al. JCTC (2018)]

Continuum states

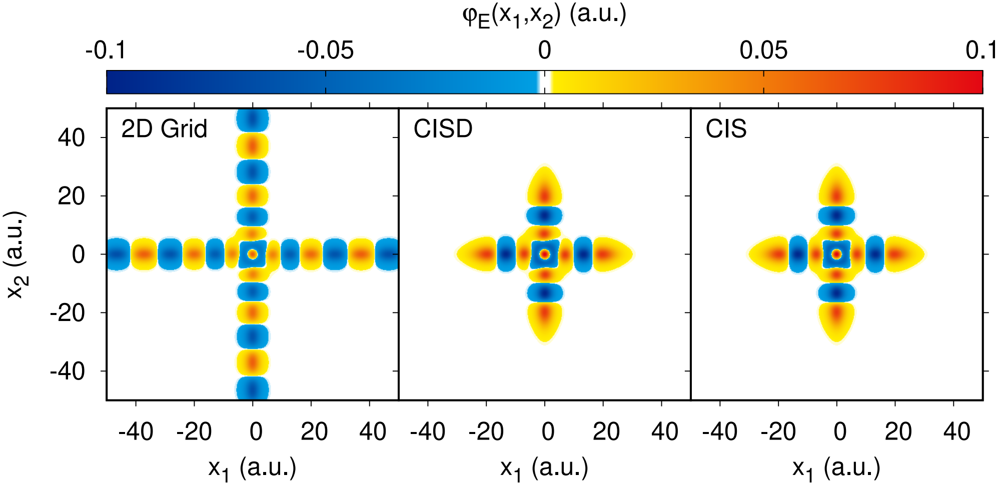
1D H_2^+

$2x_\alpha = 32\text{a.u.}$ **for** 800nm, 10^{14}W.cm^{-2}



Continuum states

1D H_2 E=0.02 a.u.



10^6 **grid points** 276 **determinants** 23 **determinants**

Continuum states lifetime

The continuum states are centered around the nuclei
The ionized electron "cannot leave"

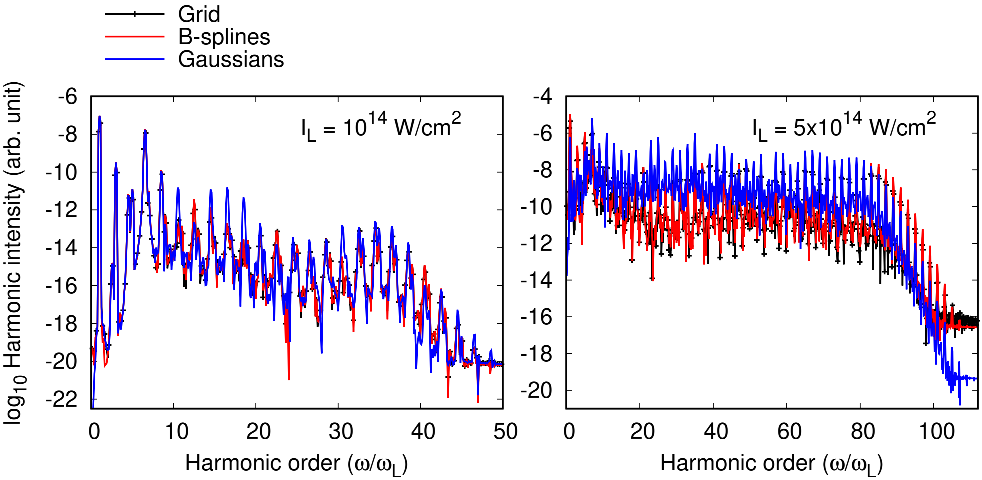
Heuristic Lifetime Model

- Enforce a decrease of population to model ionization
- Add an imaginary part to the energy of continuum states $\rightarrow$ finite lifetime
- The lifetime is computed as $\tau = \frac{d}{\sqrt{2E}}$

[Coccia et al. Int.J.Q.Chem. 2016]

HHG spectrum

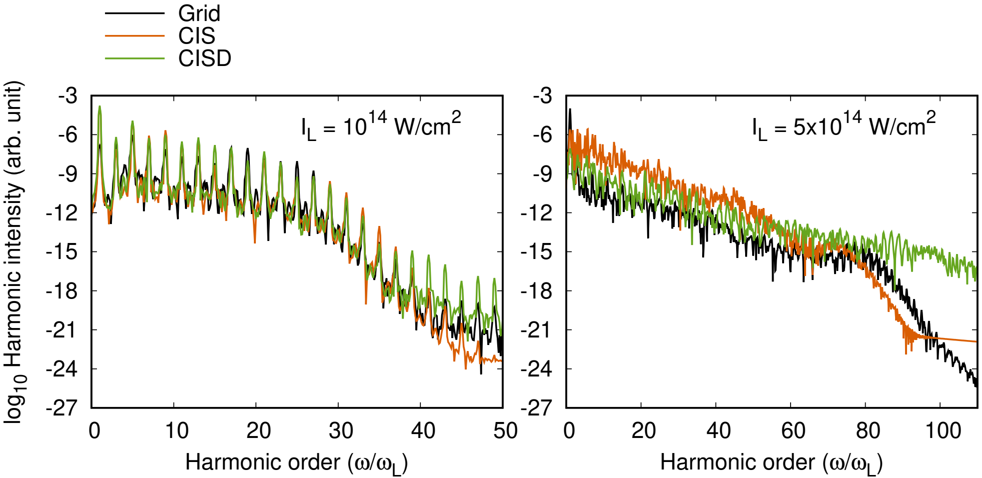
1D H_2^+



Good agreement at medium intensity

HHG spectrum

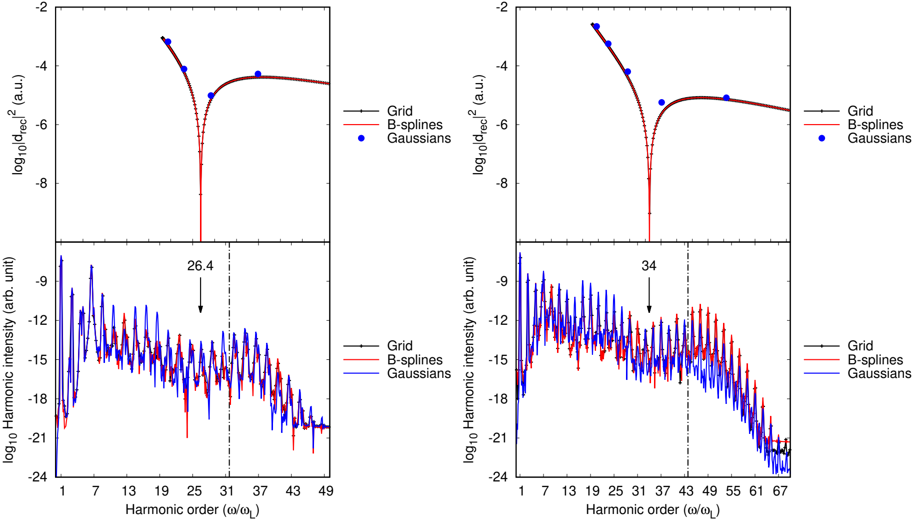
1D H_2



Good agreement at medium intensity

2-center interference

1D H_2^+



Conclusions

Conclusions

- Strong field processes require to solve the full TDSE
- Approximations have to be made
- Low dimensional systems are versatile tools to model complex processes
- Gaussian-based method can model strong field processes at medium intensities