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IMCN

Ultrafast laser-atom interaction processes in the long wavelength limit

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Outline

- What do we mean by long wavelength limit ?
- Low-energy behaviour of the ATI spectra
- The strong field approximation revisited
- Development of a separable potential model to treat one-electron processes
- Generalization of the model to complex systems exposed to ultra-short pulses
- Conclusions and perspectives

What do we mean by the long wavelength limit ?

Objectives of this contribution

To analyse the interaction of a one active electron system with an ultra-short laser pulse composed of 2 optical cycle, a fixed peak intensity of 10^{14} W/cm^2 and a frequency << ionisation potential

→ IONISATION PROCESS + SUBSEQUENT DYNAMICS

↓ Dipole approximation OK

↓ Non dipole corrections become important

Ponderomotive potential $U_p = E^2/4\omega^2$ allows to quantify non dipole couplings

- Magnetic field is important when the displacement $\beta = U_p/2c\omega$ of the electron in the laser field propagation direction is about 1 a.u.
- Radiation pressure produces a momentum in the laser field propagation direction given by U_p/c.
- Relativistic effects occur when $U_p \approx mc^2$.

Our study of the complex electron dynamics in presence of the electric, magnetic and Coulomb fields is based on the Pauli equation

High order Pauli equation

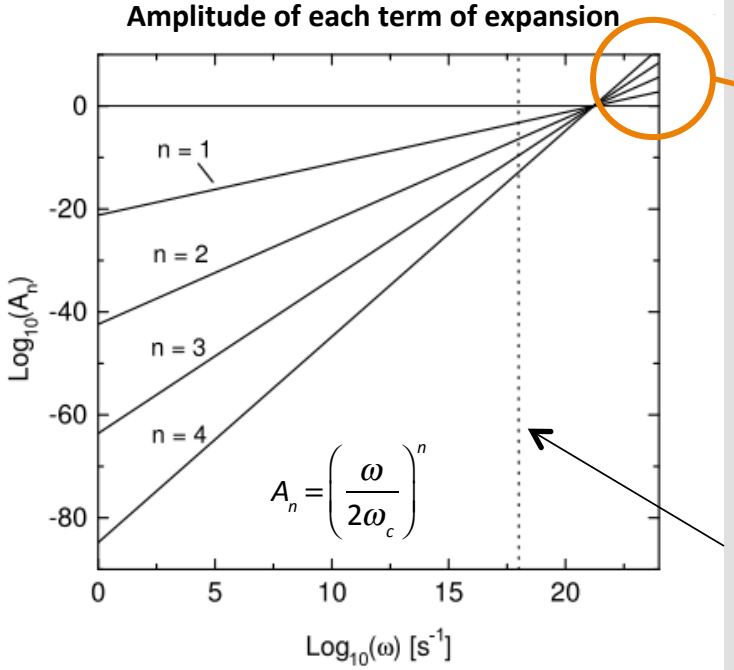
DIRAC EQUATION → PAULI EQUATION

Non-relativistic limit

Foldy – Wouthuysen transformation

Hinschberger & Hervieux (PLA 376, 2012) + Meyap Soh → **Pauli equation contains terms that couple successive time derivatives of the external fields to the spin**

$$-\frac{e\hbar}{2m}\vec{\sigma}\cdot\left(\vec{B}+\frac{1}{2}\sum_{n=1,2,\ldots}\left(\frac{1}{2i\omega_c}\right)^n\frac{\partial^n\vec{B}}{\partial t^n}+\frac{1}{2mc^2}\sum_{n=0,2,\ldots}\left(\frac{i}{2\omega_c}\right)\frac{\partial^n\vec{E}}{\partial t^n}\wedge(\vec{p}-e\vec{A})\right)$$



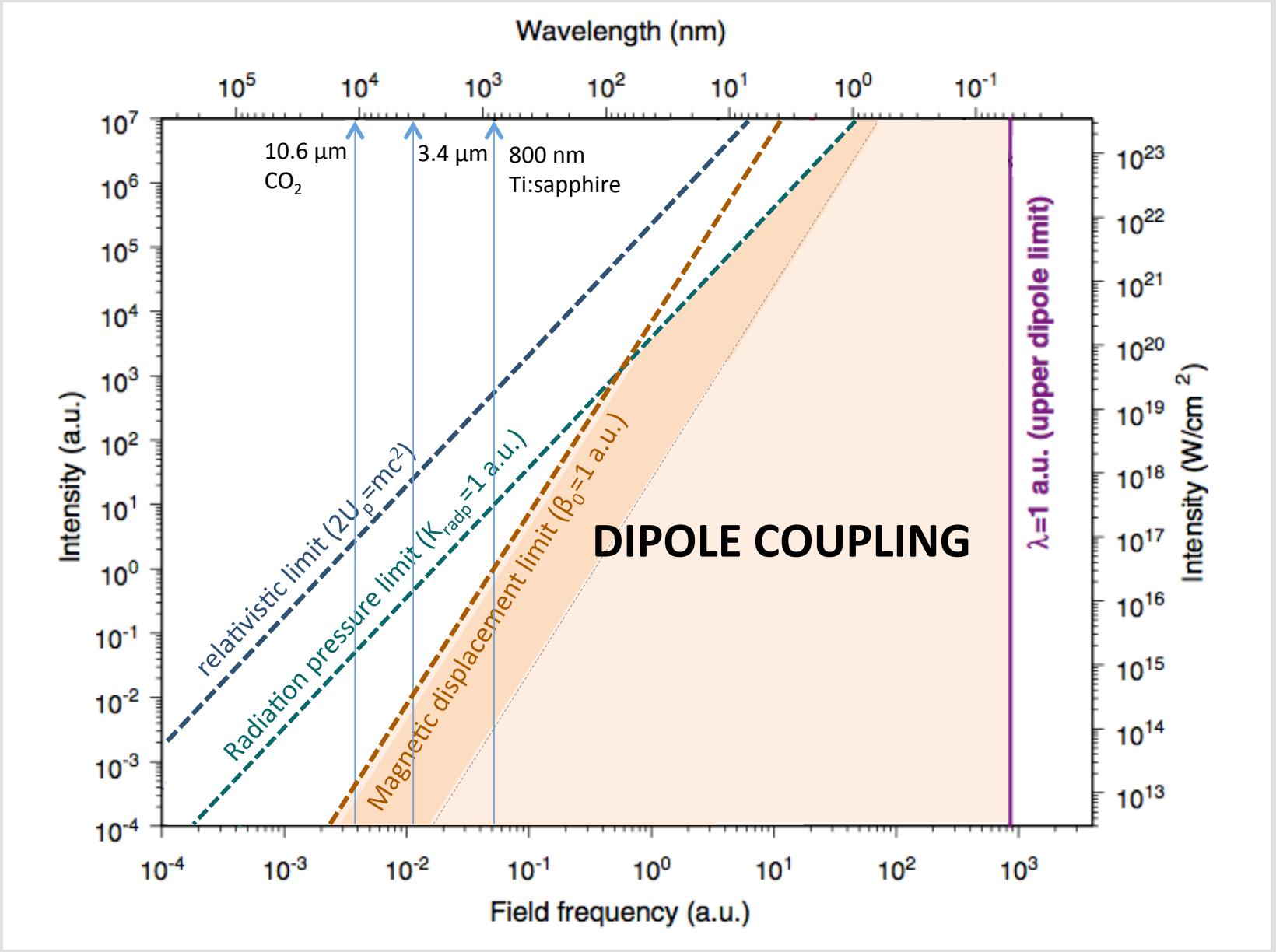
Attosecond temporal regime

For a plane wave modulated by a square envelope, each order is higher than the previous one. The re-summation of all orders gives a result proportional to:

$$1-\frac{\omega}{\omega_c}$$

→ Significant effects if $\omega\approx\omega_c=\frac{2\pi c}{\lambda_c}$, $\lambda_c=\frac{h}{mc}$

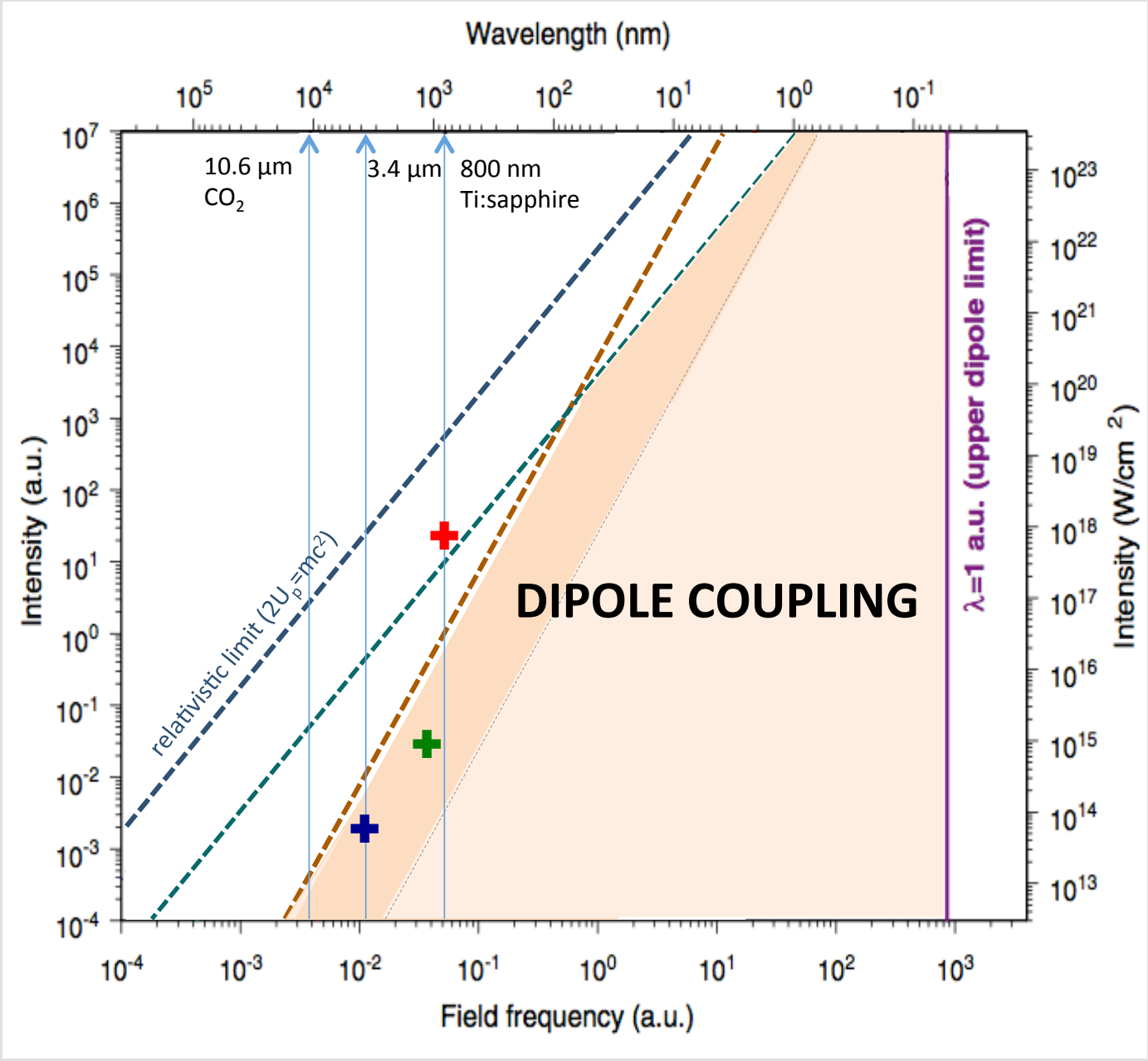
Note that Pauli equation is not valid in that regime !!



: $U_p > \omega$ → non-perturbative treatment

: $U_p < \omega$ → perturbation theory is valid

Experimental evidences



✚ : magnetic displacement effect on the electron momentum distribution projected on the axis of propagation.

Ludwig *et al.*,
PRL 113, 243001 (2014)

✚ : radiation pressure due to an intense circularly polarized field.

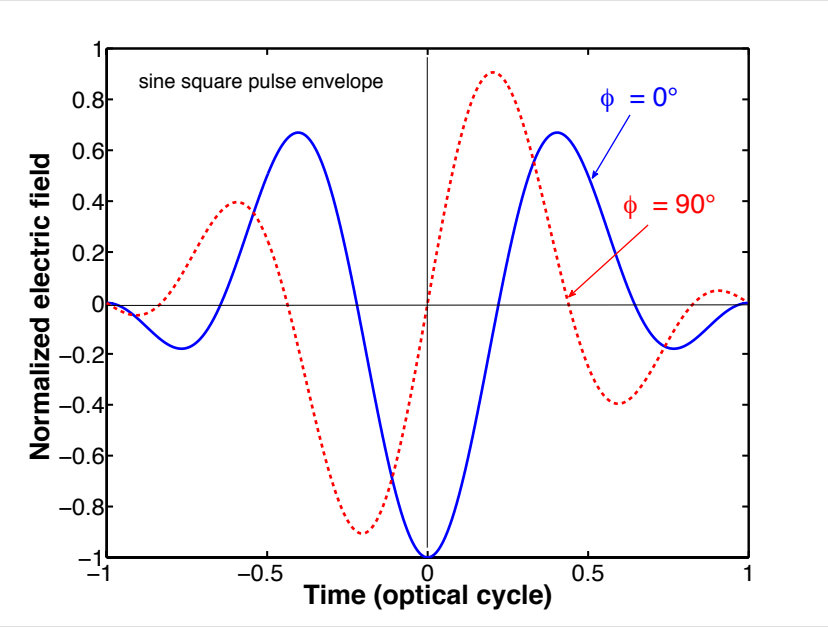
Smeenk *et al.*,
PRL 106, 193002 (2011)

✚ : effect of magnetic field in the interaction of multi-charged neon at very high intensity

Palaniyappan *et al.*,
PRL 94, 243003 (2005)

Low energy behaviour of the ATI spectra

Interaction of atomic hydrogen with an ultra-short laser pulse



Linear polarization

Dipole approximation

Velocity gauge

Pulse duration = 2 cycles

Peak intensity = 10^{14} W/cm^2

0.8 μm ≤ λ ≤ 3.6 μm

Numerical treatment of TDSE

Spectral method

- real coulomb sturmian functions
- complex coulomb sturmian functions (equivalent to a global complex rotation)
- B-splines

Time propagation

- Arnoldi algorithm
- Crank-Nicolson algorithm
- two-stage diagonally implicit Runge-Kutta method of second order

Useful (complex) norm for controlling accuracy of time propagation scheme

$\left|\widetilde{\Phi}(t)\right\rangle = \mathcal{T}\left|\Phi(-t)\right\rangle$ → the norm $\left\langle\widetilde{\Phi}(t)\middle|\Phi(t)\right\rangle$ is conserved in time

Properties of the time reversal operator

$\left\langle\widetilde{\Phi}(\infty)\middle|\Phi(\infty)\right\rangle = \left\langle 1s\middle|\Phi(\infty)\right\rangle$ → $\left\langle\widetilde{\Phi}(0)\middle|\Phi(0)\right\rangle = \left\langle 1s\middle|\Phi(\infty)\right\rangle$ = amplitude to stay in 1s state

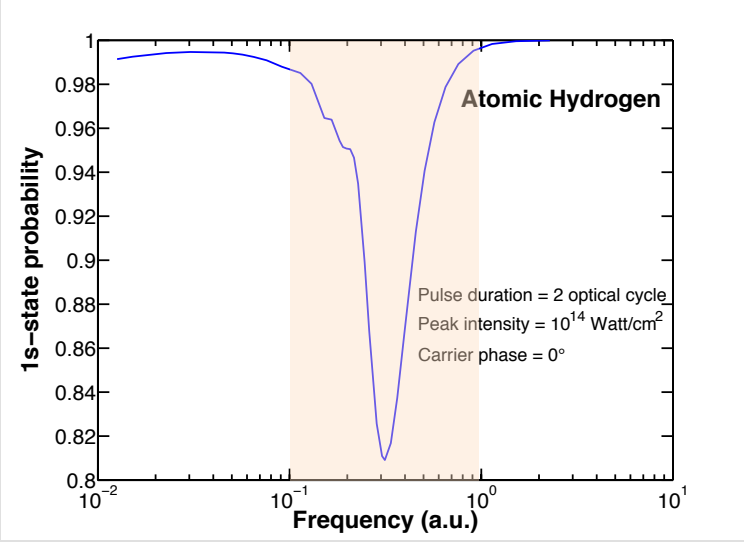
symmetric pulse

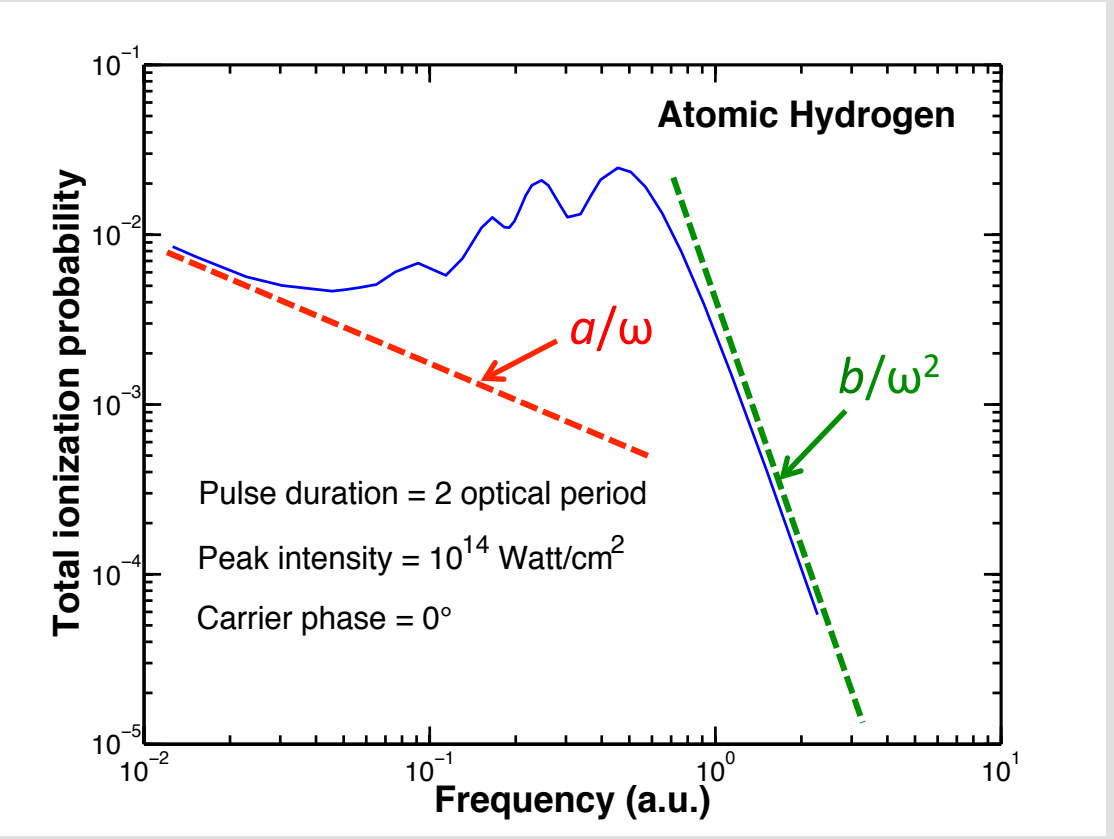
No need to propagate until the end of the pulse

Few comments about the numerical calculations

- Energy spectra are calculated by projecting on Coulomb functions $\Psi_c^{(-)}(\vec{r},\vec{k})$ or pseudo-states
- Size of sturmian bases: 6000 functions/angular momentum ℓ with ℓ_{max}= 160
- Size of the grid for B-splines : about 1000 a.u.

Results



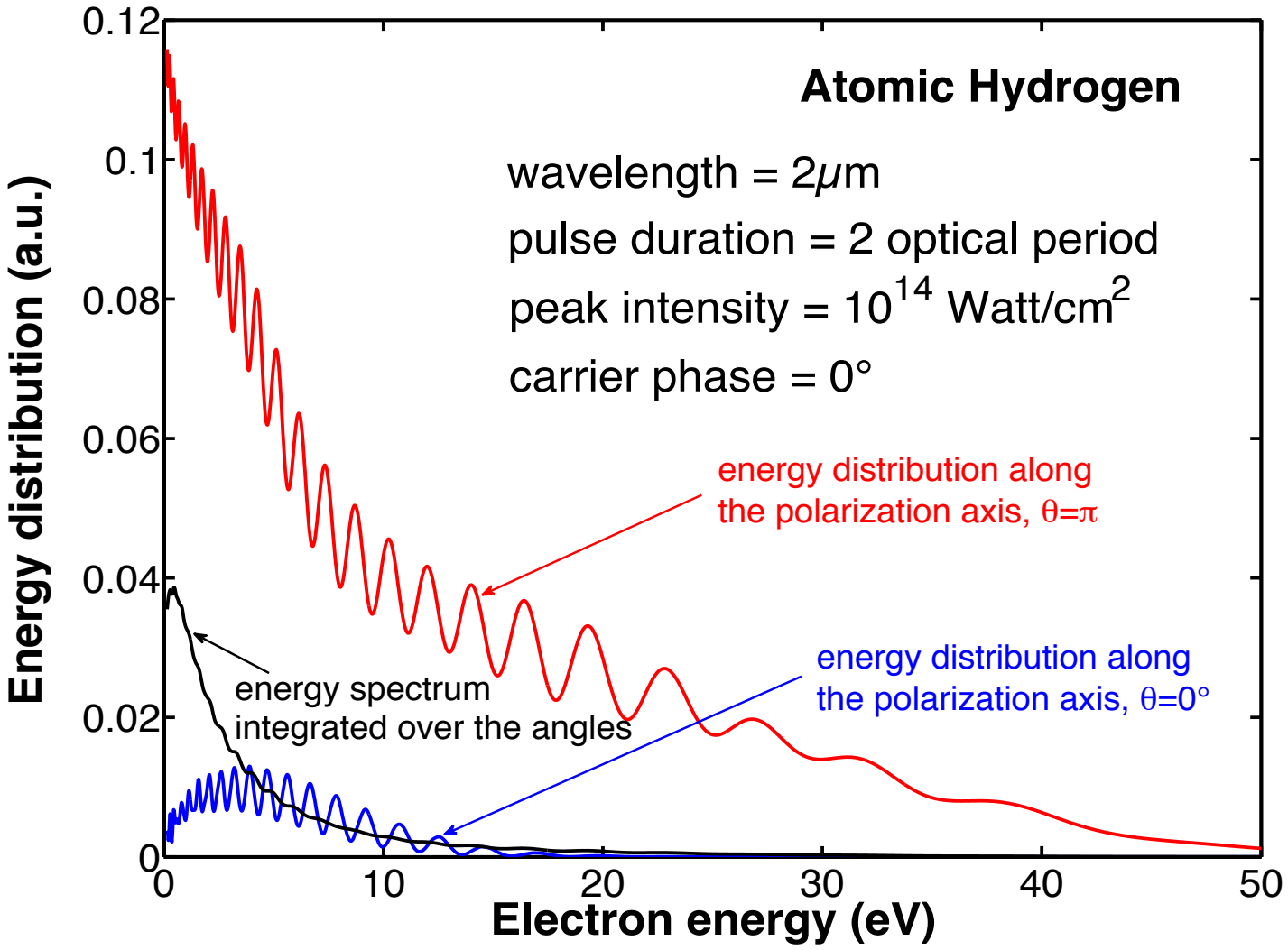


- There are 3 frequency regimes :
  - $0.01 < \omega < 0.1$
    - very weak ionisation probability
    - stabilization with increasing intensity ?
  - $0.1 < \omega < 1$

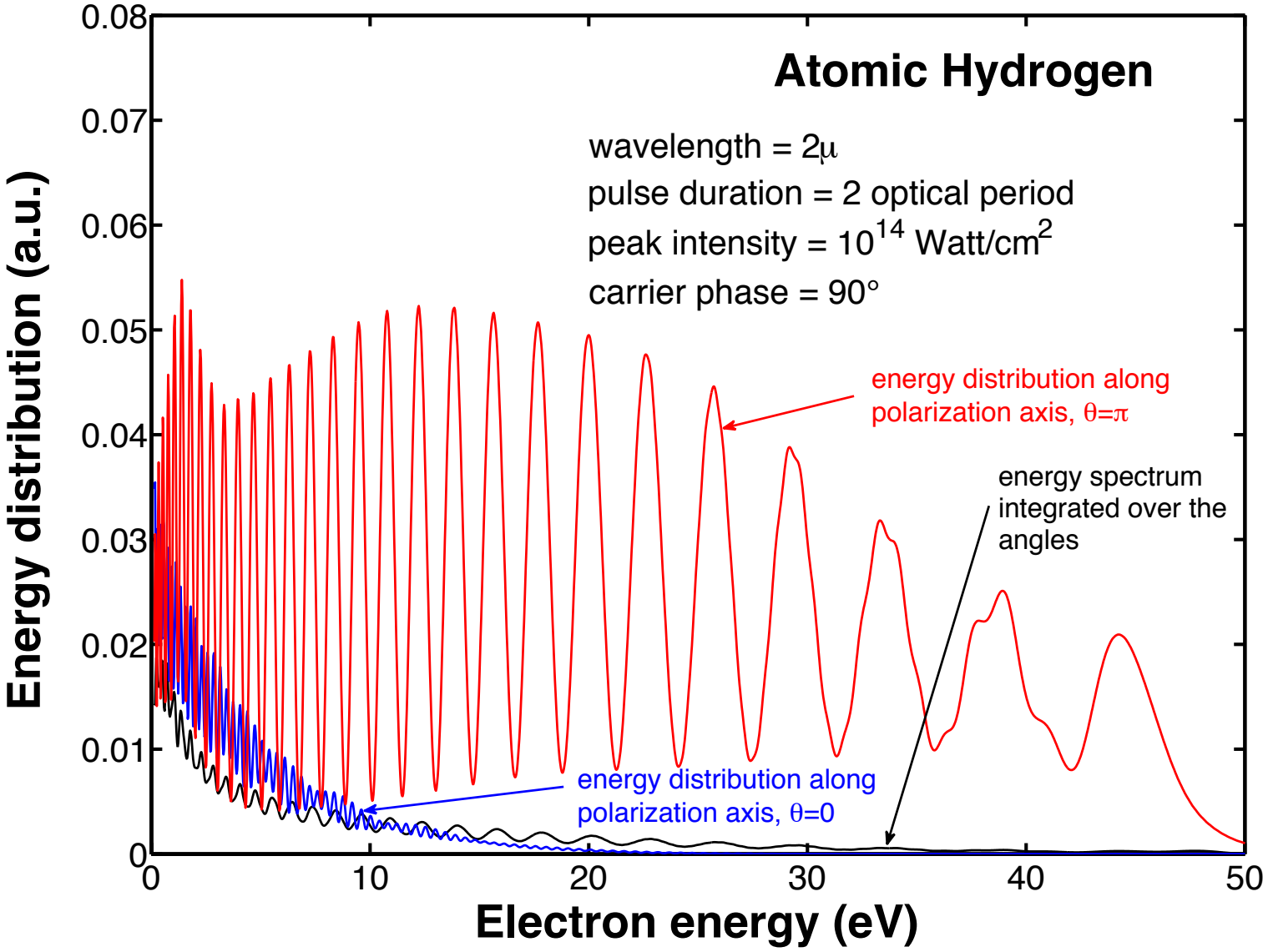
    regime of intermediate resonances
  - $\omega > 1$

    pulse too short -> no ionisation

- In the low frequency regime, the ionization yield becomes constant.
- Dimitrovski *et al*. showed that in the case of a half cycle pulse, such "asymptote" exists and can be determined from the tunnelling probability expressed in terms of the width of the ground state for static field.
- At high frequencies, lowest order perturbation theory is valid.

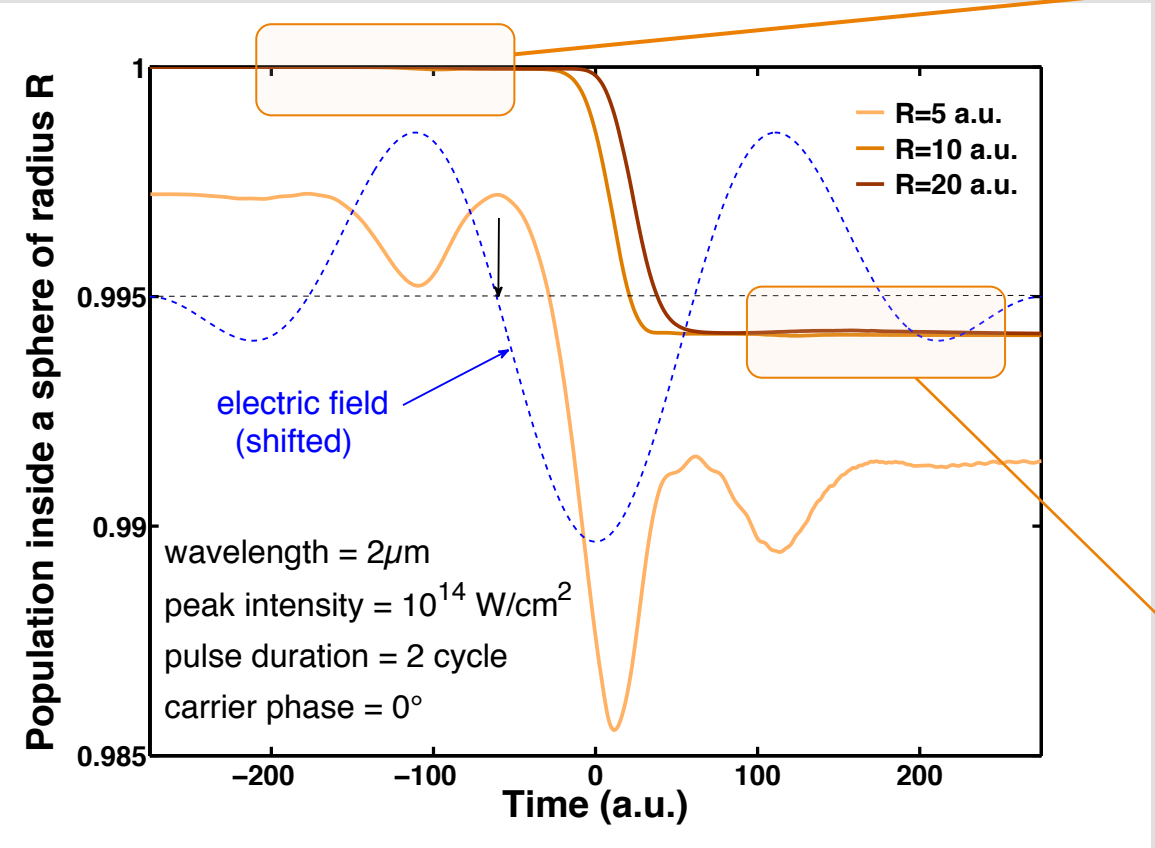


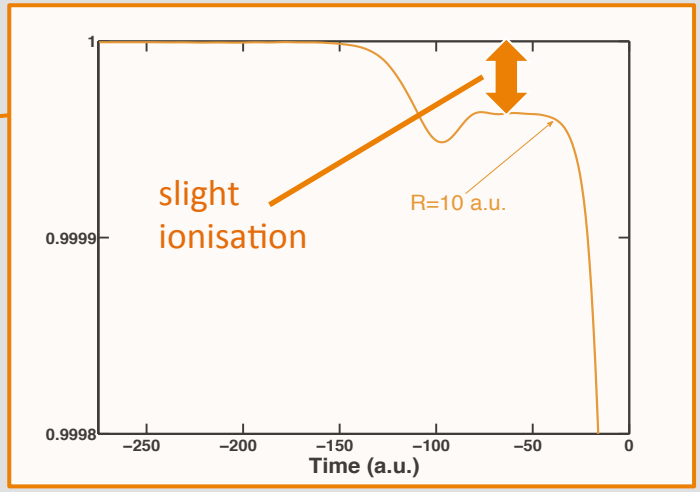
- The increasing distance in energy between the peaks results from the interference between an emitted wave packet and a returning wave packet emitted one cycle before (Arbo *et al.*)
- Important dissymmetry between backward and forward spectra (LES)

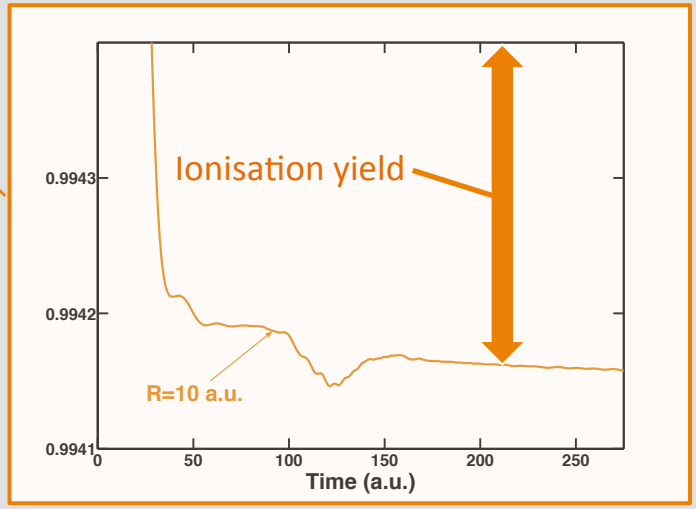


- The interference pattern is more pronounced = result of the field time behaviour
- The amplitude of the peaks is significantly reduced with respect to the $\phi=0°$ case

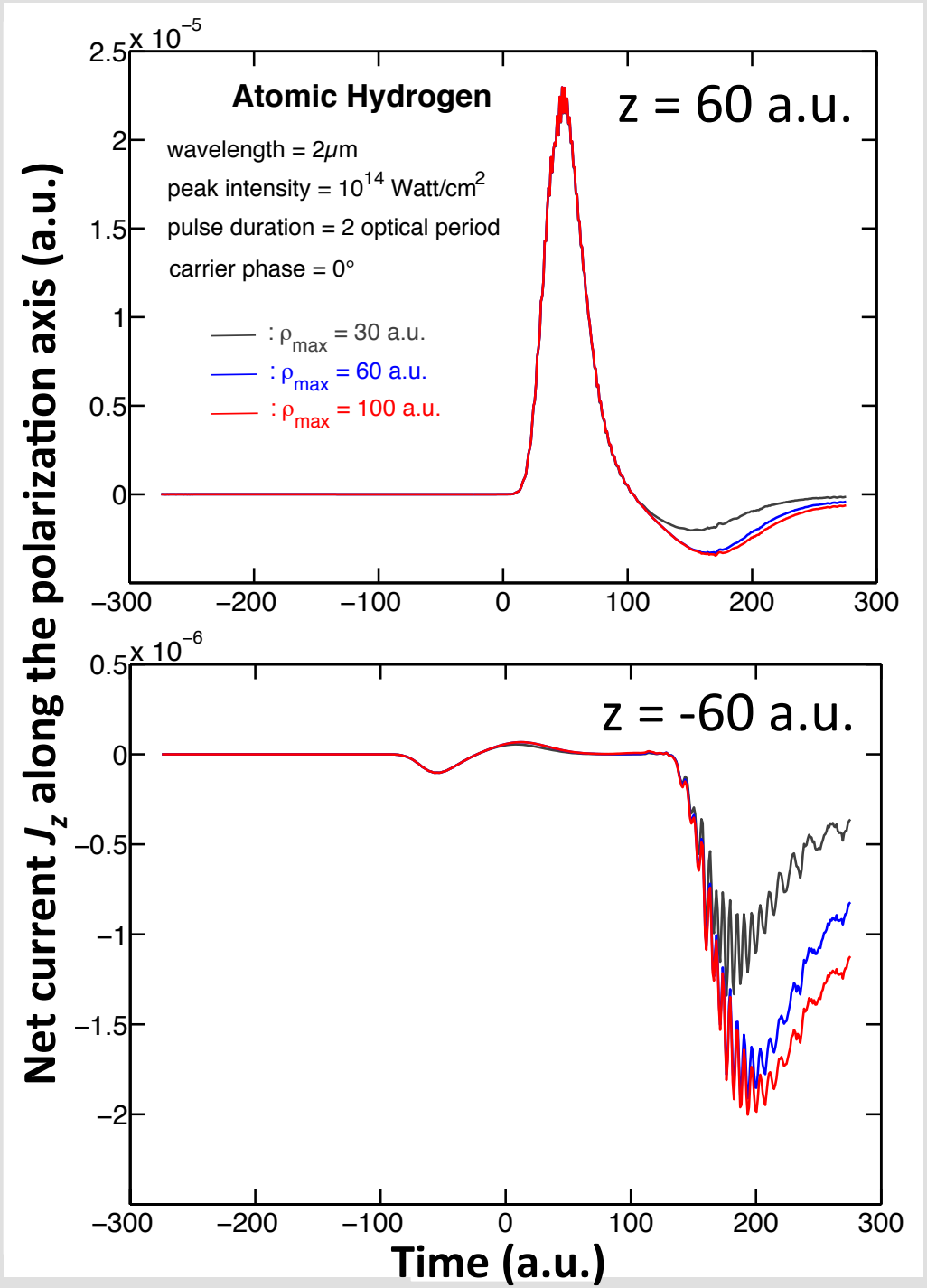
Population in sphere = gauge independent quantity !







- Around *t*=-100 a.u., polarization of the electron cloud + slight ionisation in the backward direction. Polarization follows the field and involves p-states.
- Around *t*=-50 a.u. a significant ionization starts when the electric field is zero (A(t) is maximum). Electron are emitted in the forward direction.

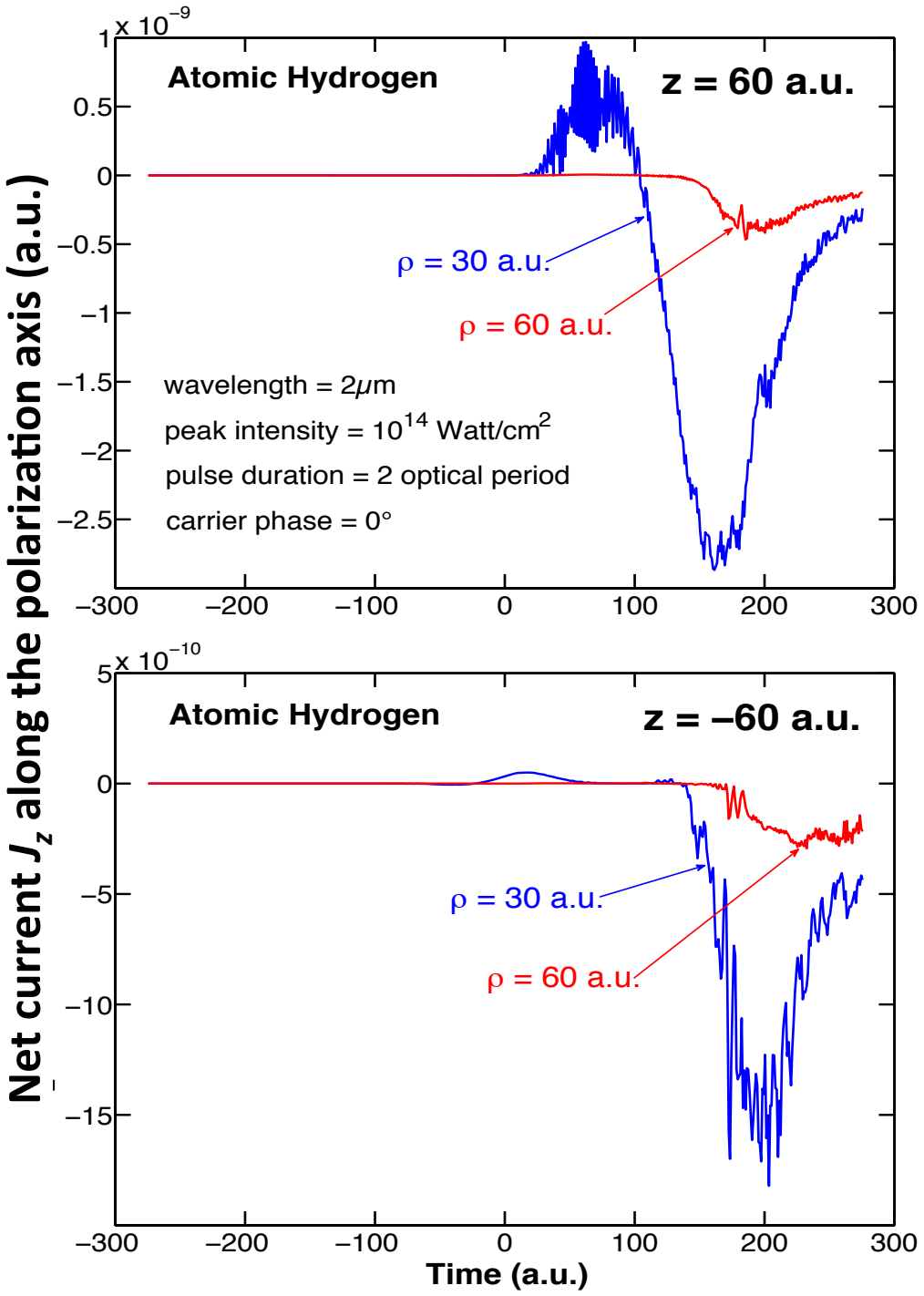


$$J_z(z,\rho_{max},t) = \int_0^{\rho_{max}} \rho\,[\vec{e}_z \cdot \vec{j}_V(z,\rho,t)]\,d\rho$$

with : $\vec{J}_V = \vec{J}_L - \vec{A}(t)|\Psi(\vec{r},t)|^2$

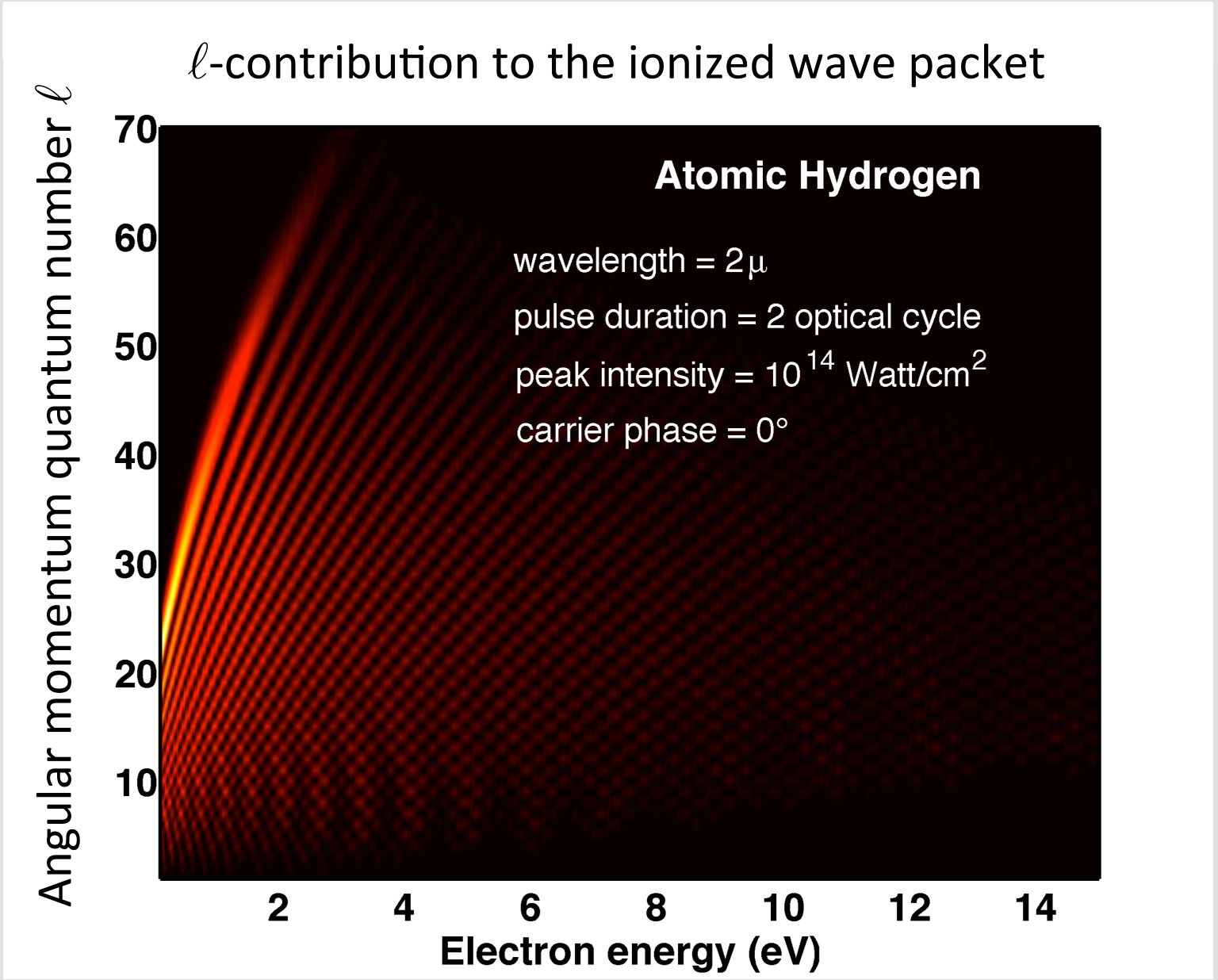
$$\vec{J}_L = \mathrm{Im}(\Psi^*(\vec{r},t)\nabla\Psi(\vec{r},t))$$

- At t=50 and z=60 a.u., emission of electrons in the forward direction along the polarization axis (all curves are on top of each other).
- At t=150 and z=60 a.u., most of the electrons are coming back except for the fastest (direct electrons)
- At t=200 and z=-60 a.u., electrons are leaving in the backward direction after one forward rescattering by the Coulomb potential

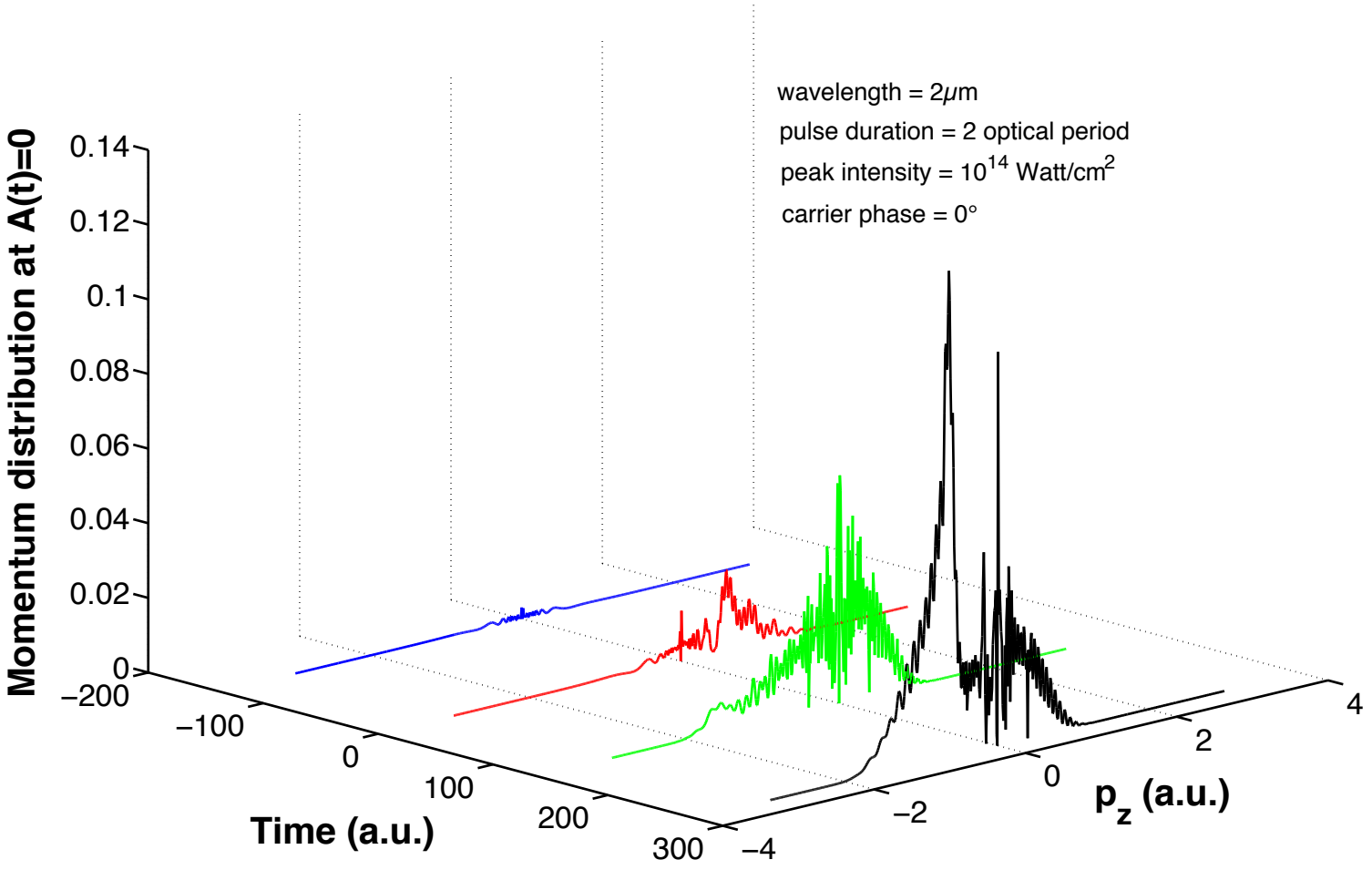


- At t=50 a.u. and z=60 a.u., the current at ρ=30 a.u. is 5 orders of magnitude smaller than the current along the axis (ρ=0).
- Between z=60 a.u. et z=-60 a.u., the dispersion of the wave packet in a direction perpendicular to the polarization axis is small because of the focusing effect of the Coulomb potential (Rutherford cross section diverges in the forward of backward direction).

Not a soft collision



- For threshold electrons, ℓ_{max} corresponds **exactly** to the number of photons to be absorbed to go from the 1s state to the threshold (without considering U_p)



- The momentum distribution along the polarization axis is consistent with the spectrum along this axis.
- The rich structure observed for positive p_z characterizes **direct electrons** involving the ionisation process only.

The simple man's theory

Main idea

Un electron is set free at time t_{ion} in $x(t_{ion})$=0 with no velocity. From thereon, it is driven by the field along the polarization axis (1D) without interacting with the ionic potential anymore.

Basic classical equations :

$$\dot{x}(t) = \dot{x}(t_{ion}) + A(t) - A(t_{ion})$$

$$x(t) = x(t_{ion}) + \int_{t_{ion}}^{t} A(t')\mathrm{d}t' - A(t_{ion})(t - t_{ion})$$

Look for re-collision time t_r

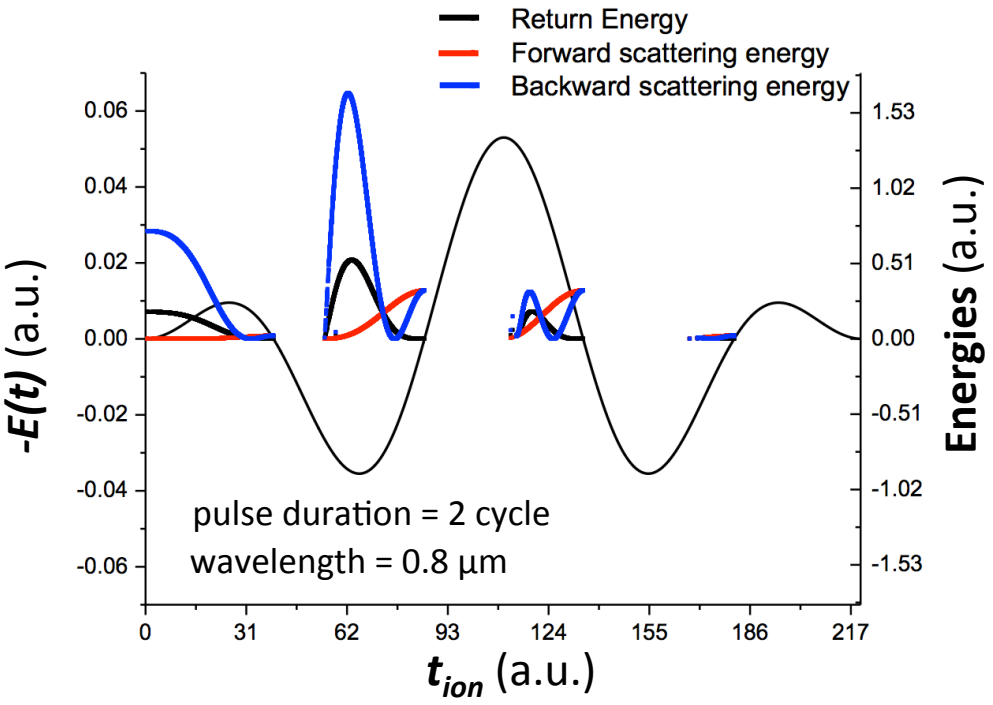
$$x(t_r) = \int_{t_{ion}}^{t_r} A(t')\mathrm{d}t' - A(t_{ion})(t_r - t_{ion}) = 0$$

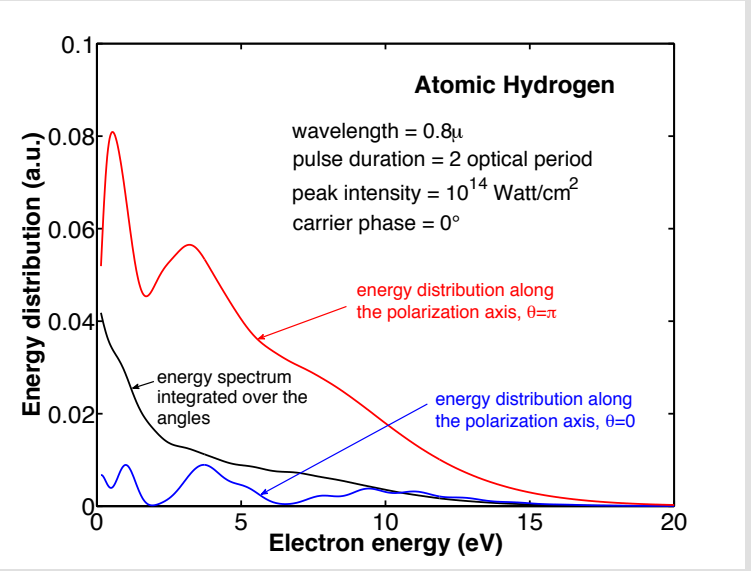
Definitions

Returning energy : $E_r = \frac{1}{2}\left(A(t_r) - A(t_{ion})\right)^2$

Forward energy = energy at the end of pulse if scattering in forward direction: $E_f = \frac{1}{2}A^2(t_{ion})$

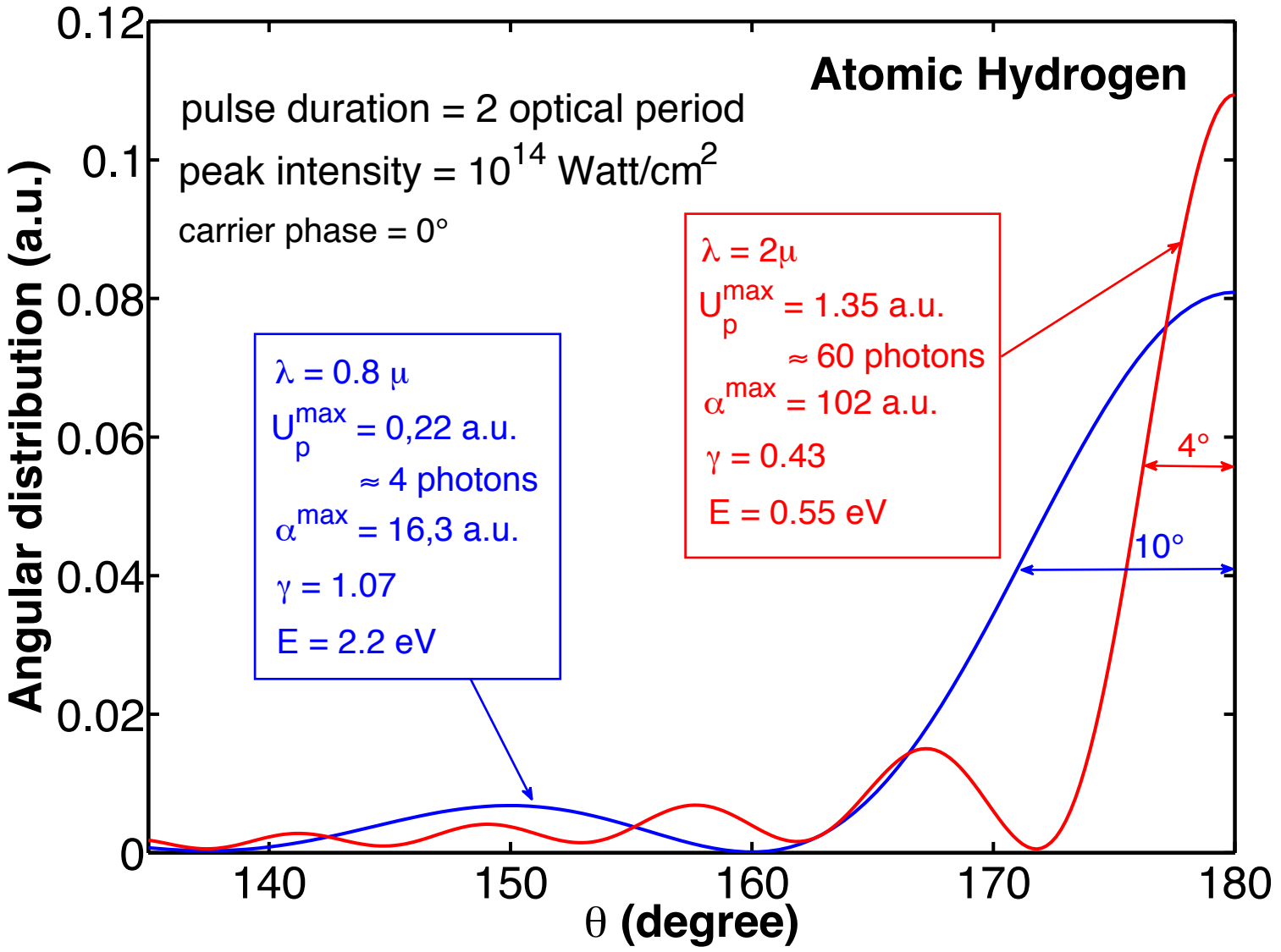
Backward energy = energy at the end of the pulse if backscattering occurs : $E_b = \frac{1}{2}\left(2A(t_r) - A(t_{ion})\right)^2$





- Electrons that are emitted at t_{ion} = 62 a.u. and that are re-scattered in the forward direction come out with an energy of about 0.08 a.u. Their return energy is about 0.6 a.u. which is consistent with 3.2 U_p >< an analysis that does not take the pulse envelop into account and which predicts that in the case of a soft collision, the peak close to zero energy cannot be observed in the case of two cycle pulses.

- The returning energy is proportional to λ^2



- The width at half maximum scales as 1/λ ⟺ the momentum transfer in the direction perpendicular to the polarization axis decreases as λ increases.

The strong field approximation revisited

The SFA and its variants exist for about 50 years

Keldysh published his first paper in 1964 !

The SFA is very popular

- It establishes a bridge between the classical and quantum descriptions of strong field processes
- It is consistent with the "simple man's theory"
- It provides a clear physical interpretation of the HOHG spectra and qualitatively explains most of their main features

The main idea

SFA's assumption consists basically in considering the binding potential dominant *until* ionization whereas the laser field takes over *after* the ionization has occurred.

The generalized SFA treats the effects of the possible electron re-scatterings from its parent ion core in a systematic perturbation series.

Basic equations

Definitions : $\frac{1}{c}\vec{A}(t) = -b'(t)\vec{e}_z, \quad \vec{E}(t) = b''(t)\vec{e}_z$ with $b(0) = b'(0) = b''(0) = b(T) = b'(T) = b''(T) = 0$

The wave packet satisfies the following TDSE :

- in the velocity gauge

$$\left[\mathrm{i}\frac{\partial}{\partial t} + \frac{1}{2}\Delta_r + \frac{Z}{r} - \mathrm{i}b'(t)(\vec{e}_z \cdot \nabla_r) \right] \Phi_v(\vec{r},t) = 0, \qquad \Phi_v(\vec{r},0) = \varphi_0(\vec{r})$$

- in the length gauge

$$\left[\mathrm{i}\frac{\partial}{\partial t} + \frac{1}{2}\Delta_r + \frac{Z}{r} - b''(\vec{e}_z \cdot \vec{r}) \right] \Phi_\ell(\vec{r},t) = 0, \qquad \Phi_\ell(\vec{r},0) = \varphi_0(\vec{r})$$

The exact wave packets satisfy :

$$\Phi_\ell(\vec{r},t) = e^{-\mathrm{i}b'(t)(\vec{e}_z \cdot \vec{r}) - \mathrm{i}\varsigma(t)} \Phi_v(\vec{r},t), \qquad \varsigma(t) = \frac{1}{2}\int_0^t \mathrm{d}\xi \, (b'(\xi))^2$$

Gauge invariant Born series (in the Coulomb potential)

Start from TDSE (velocity gauge) :

$$\left[\mathrm{i}\frac{\partial}{\partial t}+\frac{1}{2}\Delta_r-\mathrm{i}b'(t)(\vec{e}_z\cdot\nabla_r)\right]\Phi_v(\vec{r},t)=-\frac{Z}{r}\Phi_v(\vec{r},t),\quad \Phi_v(\vec{r},0)=\varphi_0(\vec{r})$$

Solution of the l.h.s. of this equation = Volkov wave : $\chi_v(\vec{r},\vec{p},t)=e^{[\mathrm{i}\vec{p}\cdot\vec{r}-\mathrm{i}(p^2/2)t+\mathrm{i}b(t)(\vec{e}_z\cdot\vec{p})]}$

Introduce the Green function : $G_v(\vec{r},t;\vec{r}',t')=-\mathrm{i}\theta(t-t')\int\frac{\mathrm{d}\vec{p}}{(2\pi)^3}\chi_v(\vec{r},\vec{p},t)\chi_v^*(\vec{r}',\vec{p},t')$

The general solution of the TDSE equation writes :

$$\Phi_v(\vec{r},t)=\underbrace{\mathrm{i}\int\mathrm{d}\vec{r}'G_v(\vec{r},t;\vec{r}',0)\varphi_0(\vec{r}')}-Z\int_0^t\mathrm{d}t\int\frac{\mathrm{d}\vec{r}'}{r'}G_v(\vec{r},t;\vec{r}',t')\Phi_v(\vec{r}',t')$$

Initial wave packet dispersing
with or without electric field

The zero order term writes : $\Phi_v^{(0)}(\vec{r},t)=\theta(t)\int\frac{d\vec{p}}{(2\pi)^3}\chi_v(\vec{r},\vec{p},t)\varphi_0(\vec{p})$

This term is the starting zero order term for the Born series. The second term writes :

$$4\pi iZ\int\frac{d\vec{p}}{(2\pi)^3}\chi_v(\vec{r},\vec{p},t)\int_0^t dt'\int\frac{d\vec{p}'}{(2\pi)^3}\frac{e^{it'(p^2-p'^2)/2-ib(t')\vec{e}_z\cdot(\vec{p}-\vec{p}')}}{|\vec{p}-\vec{p}'|^2}\varphi_0(\vec{p}')$$

- This Born series is gauge invariant term by term
- This Born series is valid for any arbitrary initial wave packet
- At time *t*=0, the initial wave packet coincides with the initial atomic state. At times *t*>0, it will inevitably disperse contrary to any stationary state.
- The dispersion of the initial wave packet is not fully compensated at each order

USELESS !

Strong field approximations

To avoid the problem of the dispersion of the initial wave packet, it is necessary to introduce an ansatz for $\Phi_\ell(\vec{r},t)$ and $\Phi_v(\vec{r},t)$. In fact, the gauge invariance constraint allows one to defining various families of ansatz.

1st example of family

Ansatz for velocity-gauge : $\Phi_v(\vec{r},t) = e^{-i\varepsilon_0 t}\varphi_0(\vec{r}) + F_v(\vec{r},t)$

with : $\left[i\frac{\partial}{\partial t} + \frac{1}{2}\Delta_r - ib'(t)(\vec{e}_z \cdot \nabla_r) + \frac{Z}{r} \right] F_v(\vec{r},t) = ib'(t) e^{-i\varepsilon_0 t}(\vec{e}_z \cdot \nabla_r)\varphi_0(\vec{r})$

Ansatz for length-gauge : $\Phi_\ell(\vec{r},t) = e^{-ib'(t)(\vec{e}_z\cdot\vec{r}) - \zeta(t) - i\varepsilon_0 t}\varphi_0(\vec{r}) + F_\ell(\vec{r},t)$

with : $\left[i\frac{\partial}{\partial t} + \frac{1}{2}\Delta_r - b''(t)(\vec{e}_z \cdot \nabla_r) + \frac{Z}{r} \right] F_\ell(\vec{r},t) = ib'(t) e^{-ib'(t)(\vec{e}_z\cdot\vec{r}) - i\varsigma(t) - i\varepsilon_0 t}(\vec{e}_z \cdot \nabla_r)\varphi_0(\vec{r})$

2nd example of family ➡ Keldysh formula

Ansatz for length-gauge : $\Phi_\ell(\vec{r},t) = e^{-i\varepsilon_0 t}\varphi_0(\vec{r}) + F_\ell^{'}(\vec{r},t)$

with : $\left[i\frac{\partial}{\partial t} + \frac{1}{2}\Delta_r - b''(t)(\vec{e}_z\cdot\vec{r}) + \frac{Z}{r} \right] F_\ell^{'}(\vec{r},t) = b''(t)e^{-i\varepsilon_0 t}(\vec{e}_z\cdot\vec{r})\varphi_0(\vec{r})$

Ansatz for velocity-gauge : $\Phi_v(\vec{r},t) = e^{-ib'(t)(\vec{e}_z\cdot\vec{r}) - \zeta(t) - i\varepsilon_0 t}\varphi_0(\vec{r}) + F_v^{'}(\vec{r},t)$

with : $\left[i\frac{\partial}{\partial t} + \frac{1}{2}\Delta_r - ib'(t)(\vec{e}_z\cdot\nabla_r) + \frac{Z}{r} \right] F_v^{'}(\vec{r},t) = b''(t)e^{ib'(t)(\vec{e}_z\cdot\vec{r}) + i\zeta(t) - i\varepsilon_0 t}(\vec{e}_z\cdot\vec{r})\varphi_0(\vec{r})$

- In a given family, each function *F* satisfies a non homogeneous TDSE that can be solved iteratively thereby leading to a Born series (in the Coulomb potential) for each *F* and hence for each Φ .

- Within a given family, all terms of the Born series for each Φ are gauge invariant. In a given gauge, each term of the Born series for Φ belonging to different families, differ but the sum of all terms of these Born series remains gauge invariant.

SFA – velocity gauge (1st family)

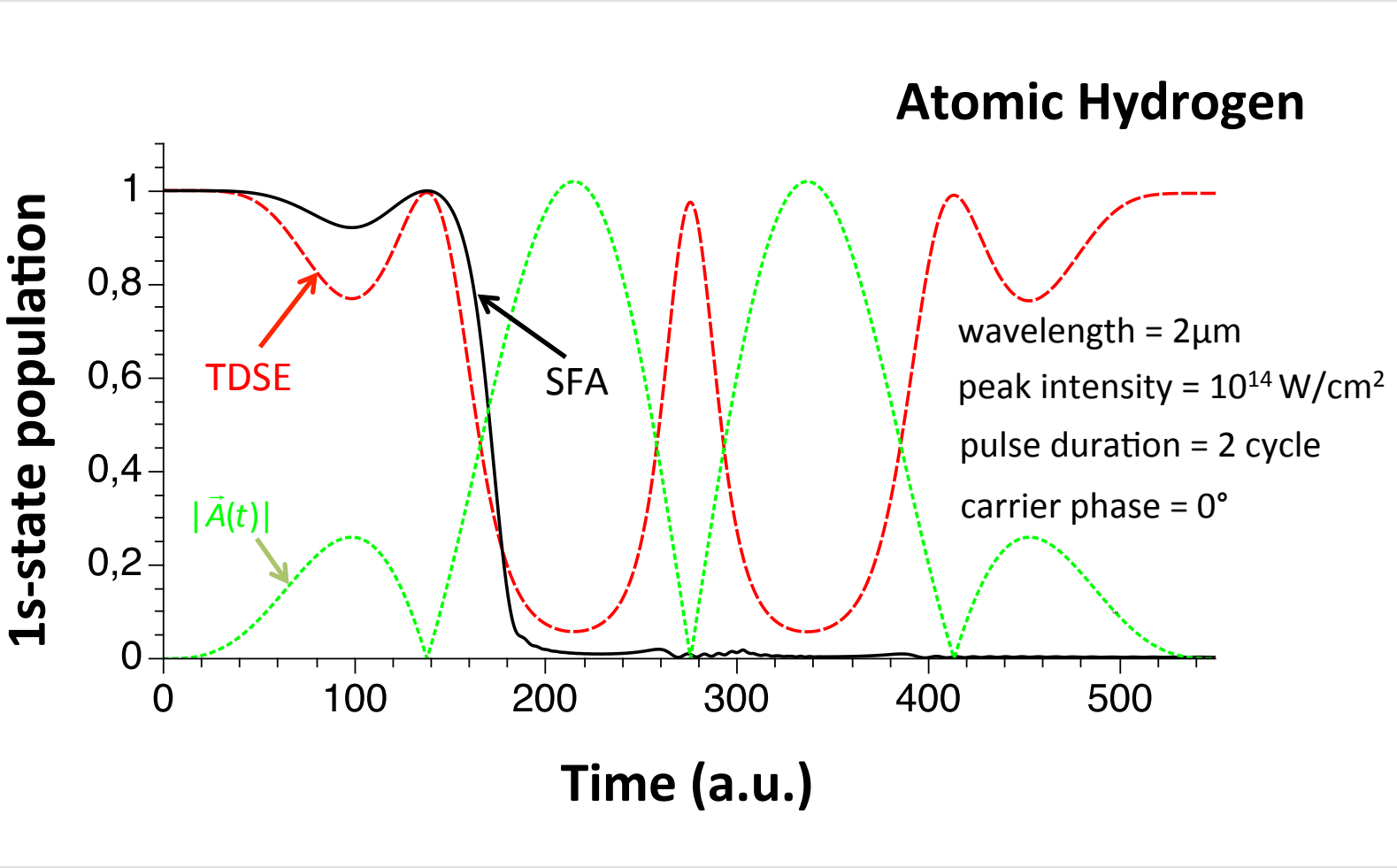
Ansatz : $\Phi_v(\vec{r},t) = e^{-i\varepsilon_0 t}\varphi_0(\vec{r}) + F_v(\vec{r},t)$ with $F_v(\vec{r},0) = 0$

Equation for zero order $F_v^{(0)}(\vec{r},t)$: $\left[i\frac{\partial}{\partial t} + \frac{1}{2}\Delta_r - ib'(t)(\vec{e}_z \cdot \nabla_r) \right] F_v^{(0)}(\vec{r},t) = \underbrace{ib'(t)e^{-i\varepsilon_0 t}(\vec{e}_z \cdot \nabla_r)\varphi_0(\vec{r})}_{\text{Source term}}$

Equation for nth order $F_v^{(n)}(\vec{r},t)$: $\left[i\frac{\partial}{\partial t} + \frac{1}{2}\Delta_r - ib'(t)(\vec{e}_z \cdot \nabla_r) \right] F_v^{(n)}(\vec{r},t) = -\frac{Z}{r} F_v^{(n-1)}(\vec{r},t)$

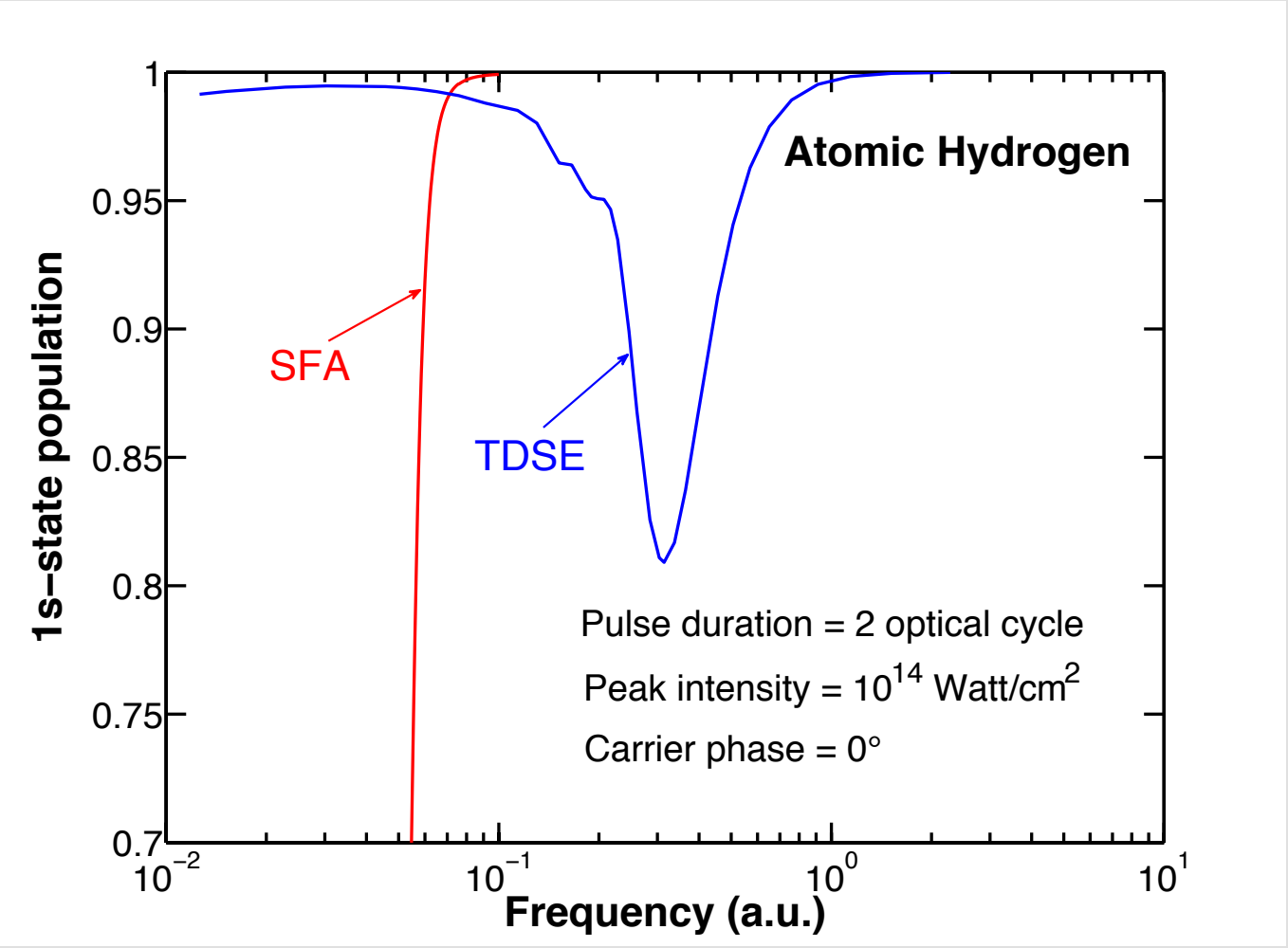
- $F_v^{(0)}(\vec{r},t)$ can be calculated analytically
- $F_v^{(n)}(\vec{r},t)$ for $n \geq 0$ can be obtained numerically ⟹ convergence study
- comparison with TDSE results imposes a proper normalization of $\Phi_v(\vec{r},t)$
- Spectrum is obtained by projecting on Coulomb functions or plane waves after subtraction of the initial state contribution

1s-state population as a function of time : $P_{1s}(t) = \frac{\left| \left\langle \varphi_{1s} e^{-i\varepsilon_{1s} t} \middle| \Phi_v(t) \right\rangle \right|^2}{\left\langle \Phi_v(t) \middle| \Phi_v(t) \right\rangle}$



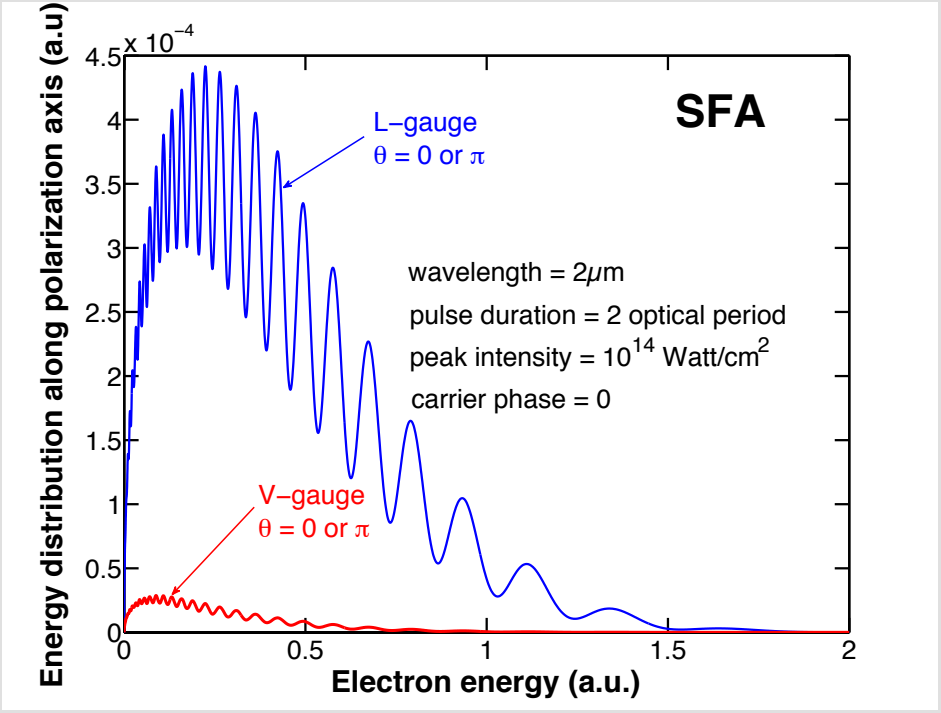
- SFA leads to full ionisation at the beginning of the pulse

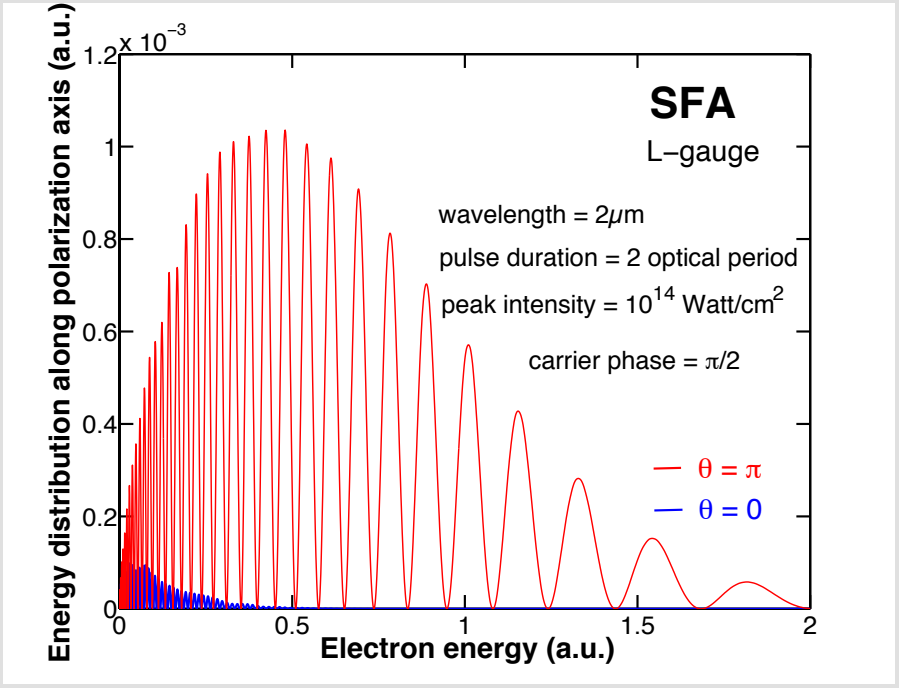
1s-state population at the end of the pulse as a function of the frequency

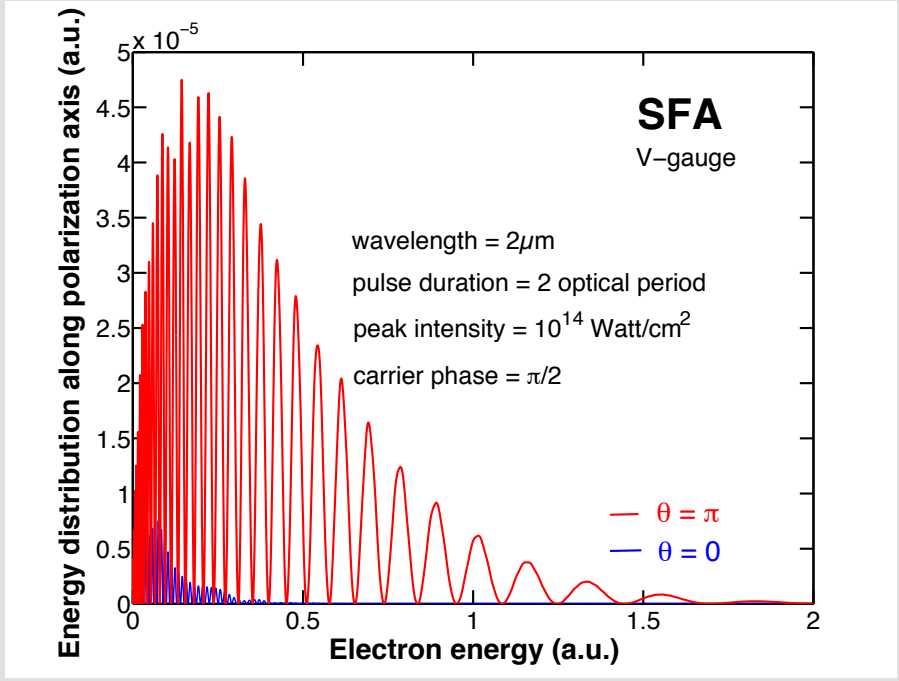


- In the low frequency regime, SFA leads to a full ionisation >< TDSE

Unormalized spectra

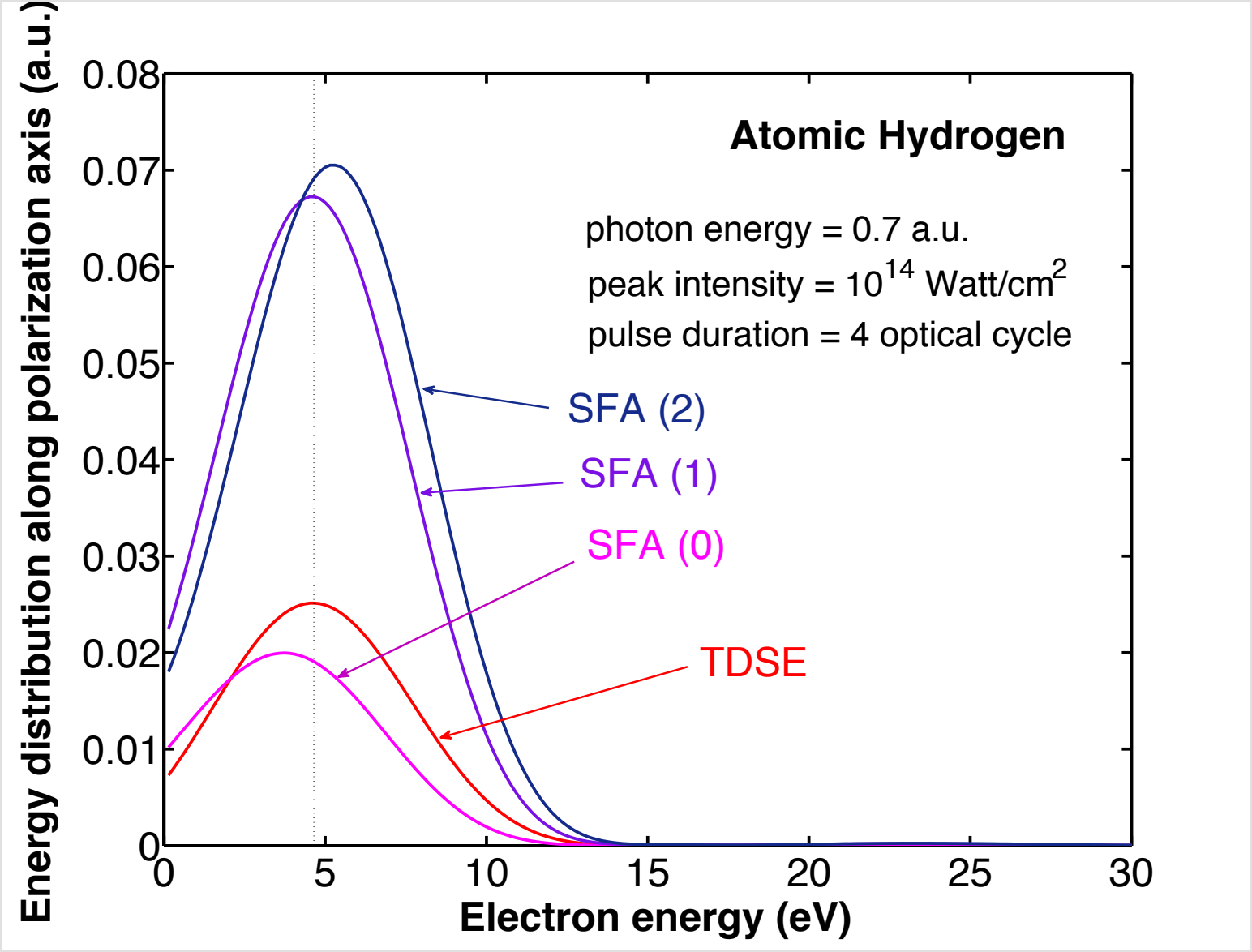






- When the carrier phase = 0, there is no dissymmetry and SFA describes only the direct electrons.
- The important differences between gauges is most likely due to the projection on plane waves (instead of Coulomb waves).

High order SFA (preliminary results)



Development of a separable potential model to treat one-electron processes

Main objective

To simplify the numerical solution of the TDSE in very demanding physical situations

- the long wavelength limit
- complex atomic and molecular targets
- fields of arbitrary polarization

Ideally, this model should

- allow a treatment that avoids a partial wave analysis of the solution wave packet
- provide information on the dynamics
- allow one to test the approximation schemes such as SFA
- Apply within the mean field approximation to complex atomic or molecular systems

Preliminary remark

In the case of coulomb systems it is more natural to work in the velocity gauge and therefore in momentum space. This is a consequence of Ehrenfest's theorem :

Length gauge: $$\frac{\mathrm{d}}{\mathrm{d}t}\langle \boldsymbol{\pi} \rangle = \frac{1}{\mathrm{i}\hbar}\left\langle \left[\boldsymbol{\pi}, \mathrm{H}_{length} \right] \right\rangle = -\langle \nabla_{\mathbf{r}} V \rangle + q\mathbf{E}(t)$$

➡ From the point of view of the forces acting on the electron, the role of the Coulomb potential is decoupled from the electric field

Velocity gauge: $$\frac{\mathrm{d}}{\mathrm{d}t}\langle \mathbf{p} \rangle = \frac{1}{\mathrm{i}\hbar}\left\langle \left[\mathbf{p}, \mathrm{H}_{velocity} \right] \right\rangle = -\langle \nabla_{\mathbf{r}} V \rangle$$

➡ The time evolution of the canonical momentum **p** results only from the gradient of the Coulomb potential.

In the limit where $\langle \nabla_{\mathbf{r}} V \rangle \to 0$, the canonical momentum becomes a constant of motion which reduces to the drift velocity (in a.u.) of the ionized electron.

General formulation in the case of atomic hydrogen

In momentum space and in the velocity gauge, the TDSE writes:

$$\left[\mathrm{i}\frac{\partial}{\partial t} - \frac{\mathbf{p}^2}{2} - A(t)(\boldsymbol{e}_z \cdot \mathbf{p}) \right] \Phi(\mathbf{p},t) - \int \frac{\mathrm{d}^3\mathbf{p}'}{(2\pi)^3} V(\mathbf{p}-\mathbf{p}')\, \Phi(\mathbf{p}',t) = 0$$

Replace the non-local potential by a sum of N symmetric separable potentials supporting N bound states of the atom:

$$V(\mathbf{p}-\mathbf{p}') = -\frac{4\pi Z}{|\mathbf{p}-\mathbf{p}'|^2} \approx -\sum_{n=1}^{N} v_n(\mathbf{p}) v_n^*(\mathbf{p}')$$

The TDSE becomes:

$$\left[\mathrm{i}\frac{\partial}{\partial t} - \frac{\mathbf{p}^2}{2} - A(t)(\boldsymbol{e}_z \cdot \mathbf{p}) \right] \Phi(\mathbf{p},t) + \sum_{n=1}^{N} v_n(\mathbf{p}) \int \frac{\mathrm{d}^3\mathbf{p}'}{(2\pi)^3} v_n^*(\mathbf{p}') \Phi(\mathbf{p}',t) = 0$$

$$\left[i\frac{\partial}{\partial t} - \frac{\mathbf{p}^2}{2} - A(t)(\boldsymbol{e}_z \cdot \mathbf{p}) \right] \Phi(\mathbf{p},t) + \sum_{n=1}^{N} v_n(\mathbf{p}) \underbrace{\int \frac{d^3\mathbf{p}'}{(2\pi)^3} v_n^*(\mathbf{p}') \Phi(\mathbf{p}',t)}_{F_n(t)} = 0$$

The solution writes:

$$\Phi(\mathbf{p},t) = \exp\left(-i\frac{p^2}{2}t + i\,b(t)\,p_z \right) \left[\Phi(\mathbf{p},0) + \sum_{n=1}^{N} v_n(\mathbf{p}) \int_0^t d\xi\; F_n(\xi) \exp\left(i\frac{p^2}{2}\xi - i b(\xi) p_z \right) \right]$$

$b(t) = -\int_0^t A(\tau)\,d\tau$

Initial state of the atom in the **p**-space

The $F_n(t)$ components are solution of a system of coupled Volterra integral equations of the second kind:

$$\mathbf{F}(t) = \mathbf{F}_0(t) + \int_0^t \mathbf{K}(t,t')\,\mathbf{F}(t')\,dt'$$

- The 4D TDSE reduces to a system of *N* 1D Volterra equations
- This approach is equivalent to solve the TDSE in an infinite box

Determination of the symmetric separable potentials

The sum of the *N* symmetric separable potentials supports *N* bound states of the atom

The wave functions associated to these bound states (in the **p**-space) are exact and satisfy the stationary Schrödinger equation:

$$\left(\varepsilon_j - \frac{1}{2}\mathbf{p}^2\right)\varphi_j(\mathbf{p}) + \sum_{n=1}^{N}\left(\int \frac{d^3\mathbf{p}'}{(2\pi)^3} v_n^*(\mathbf{p}')\varphi_j(\mathbf{p}')\right) v_n(\mathbf{p}) = 0$$

$$\int \frac{d^3\mathbf{p}'}{(2\pi)^3} v_n^*(\mathbf{p}')\varphi_j(\mathbf{p}') = a_{jn}$$

$$\mathbf{\Phi} + \mathbf{AV} = 0 \Leftrightarrow \mathbf{V} = -\mathbf{A}^{-1}\mathbf{\Phi}$$

If **A** exists

How to solve this equation ?

Write : $\Gamma = \mathbf{A}\,\mathbf{A}^{\mathsf{T}}$ with $\Gamma_{ij} = \int \frac{\mathrm{d}^3\mathbf{p}}{(2\pi)^3} \varphi_i^* \left(\varepsilon_j - \frac{1}{2}\mathbf{p}^2 \right) \varphi_j$

If the new atomic potential supports two bound states :

$$|a_{11}|^2 + |a_{12}|^2 = \Gamma_{11}$$
$$|a_{21}|^2 + |a_{22}|^2 = \Gamma_{22}$$
$$a_{11}a_{21} + a_{22}a_{12} = \Gamma_{12}$$

→ **Infinite number of solutions** → need a prescription

Examples :

$\mathbf{A}^{-1}$ is triangular

wrong choice because the new potential may change sign

$\mathbf{A}^{-1}$ is symmetric

Best (empirical) choice but not sufficient to guarantee that the solution is unique. Conditions must be imposed on various parameters.

Since the wave functions associated to the bound states are exact, a wrong choice of the separable potentials leads to a bad description of the continuum.

Illustration in the case where the atomic potential is the Coulomb potential

1. **One-state (1s) model** (the solution is unique)

Momentum space wave function : $\varphi_{1s}(\mathbf{p}) = \dfrac{8\sqrt{\pi}}{(p^2+1)^2}$

Separable potential : $v(\mathbf{p},\mathbf{p}') = v_{1s}(\mathbf{p}) v_{1s}^{*}(\mathbf{p}')$ with $v_{1s}(\mathbf{p}) = \dfrac{4\sqrt{\pi}}{p^2+1}$

Momentum space **Configuration space**

$$\hat{V}\left|\Phi\right\rangle = -\frac{16\pi}{p^2+1}\int \frac{\mathrm{d}^3\mathbf{p}'}{(2\pi)^3}\frac{\Phi(\mathbf{p}')}{p'^2+1} \quad \xrightarrow{\text{Inverse Fourier transform}} \quad \hat{V}\left|\Psi\right\rangle = -\frac{1}{\pi}\frac{e^{-r}}{r}\int \mathrm{d}^3\mathbf{r}'\,\Psi(\mathbf{r}')\frac{e^{-r'}}{r'}$$

2. **Two-state (1s and 2s) model** (the solution is not unique)

Separable potentials : $v(\mathbf{p},\mathbf{p}') = v_{1s}(\mathbf{p}) v_{1s}^{*}(\mathbf{p}') + v_{2s}(\mathbf{p}) v_{2s}^{*}(\mathbf{p}')$

$$\mathbf{V} = -\mathbf{A}^{-1}\Phi \quad \text{with} \quad \Gamma = \mathbf{A}\,\mathbf{A}^{\mathsf{T}}$$

$$\begin{cases} v_{1s}(\mathbf{p}) = \alpha_{11}\left(\varepsilon_{1s} - \frac{1}{2}p^2\right)\varphi_{1s}(\mathbf{p}) + \alpha_{12}\left(\varepsilon_{1s} - \frac{1}{2}p^2\right)\varphi_{2s}(\mathbf{p}) \\ v_{2s}(\mathbf{p}) = \alpha_{21}\left(\varepsilon_{1s} - \frac{1}{2}p^2\right)\varphi_{1s}(\mathbf{p}) + \alpha_{22}\left(\varepsilon_{2s} - \frac{1}{2}p^2\right)\varphi_{2s}(\mathbf{p}) \end{cases}$$

Gauge invariance

The present formulation is not gauge invariant. How to make it gauge invariant ?

1. *Write the TDSE in configuration space in the absence of the field.*

$$\left[\mathrm{i}\frac{\partial}{\partial t}-\frac{\mathbf{p}^2}{2}\right]\widetilde{\Phi}(\mathbf{r},t)+\sum_{n=1}^{N}\xi_n(\mathbf{r})\int \mathrm{d}^3\mathbf{r}'\xi_n^*(\mathbf{r}')\widetilde{\Phi}(\mathbf{r}',t)=0$$

2. *The non-local potential may be expressed as a local potential that is a function of* **r** *and* **p**.

$$\sum_{n=1}^{N}\xi_n(\mathbf{r})\int \mathrm{d}^3\mathbf{r}'\xi_n^*(\mathbf{r}')\widetilde{\Phi}(\mathbf{r}',t)$$

$$=\sum_{n=1}^{N}\xi_n(\mathbf{r})\int \mathrm{d}^3\mathbf{r}'\xi_n^*(\mathbf{r}')\,e^{\mathrm{i}(\mathbf{r}'-\mathbf{r})\cdot\mathbf{p}}\,\widetilde{\Phi}(\mathbf{r},t)$$

Translation operator

$$=\widetilde{V}(\mathbf{r},\mathbf{p})\widetilde{\Phi}(\mathbf{r},t)$$

3. *Introduce the electromagnetic field by applying the minimal substitution* **p** -> **p** + **A** *both in the kinetic energy operator and the potential.*

$$\left[i\frac{\partial}{\partial t} - \frac{(\mathbf{p}+\mathbf{A})^2}{2} \right] \widetilde{\Phi}(\mathbf{r},t) + \hat{V}(\mathbf{r},\mathbf{p}+\mathbf{A})\widetilde{\Phi}(\mathbf{r},t) = 0$$

$$\left[i\frac{\partial}{\partial t} - \frac{\mathbf{p}^2}{2} - A(t)(\boldsymbol{e}_z \cdot \mathbf{p}) \right] \widetilde{\Phi}(\mathbf{r},t) + \sum_{n=1}^{N} \xi_n(\mathbf{r}) e^{i\,\mathbf{A}\cdot\mathbf{r}} \int \frac{d^3\mathbf{r}'}{(2\pi)^3}\, \xi_n^*(\mathbf{r}') e^{-i\,\mathbf{A}\cdot\mathbf{r}'} \widetilde{\Phi}(\mathbf{r}',t) = 0$$

Do we need to impose the gauge invariance or to choose a particular gauge in which the physics is well described (while making some approximation) ? The debates are not over …

The continuum states

The continuum state wave functions with an asymptotic outgoing (+) or ingoing (-) spherical wave behaviour are solution of the following stationary Schrödinger equation:

$$\frac{\mathbf{p}^2}{2}\phi^{(\pm)}(\mathbf{p},\mathbf{k}) - \sum_{n=1}^{N} v_n(\mathbf{p}) \underbrace{\int \frac{d^3\mathbf{p}'}{(2\pi)^3} v_n^*(\mathbf{p}')\phi^{(\pm)}(\mathbf{p}',\mathbf{k})}_{\chi_n^{(\pm)}(\mathbf{k})} = \frac{\mathbf{k}^2}{2}\phi^{(\pm)}(\mathbf{p},\mathbf{k})$$

Solution of the equation:

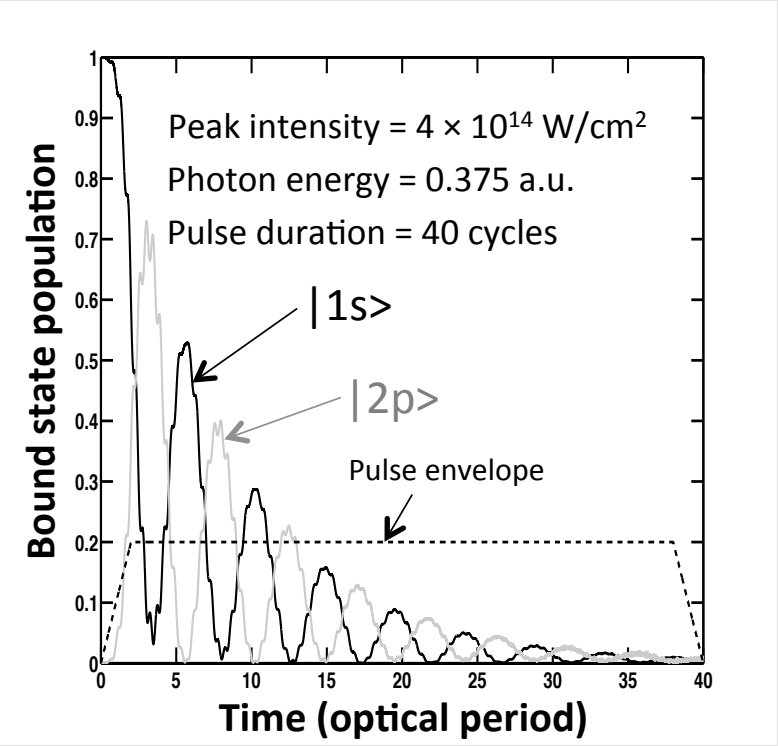
$$\phi^{(\pm)}(\mathbf{p},\mathbf{k}) = (2\pi)^{3/2}\delta(\mathbf{p}-\mathbf{k}) - \frac{2}{[(\mathrm{k}\pm i\varepsilon)^2 - \mathbf{p}^2]}\sum_{n=1}^{N} v_n(\mathbf{p})\chi_n^{(\pm)}(\mathbf{k})$$

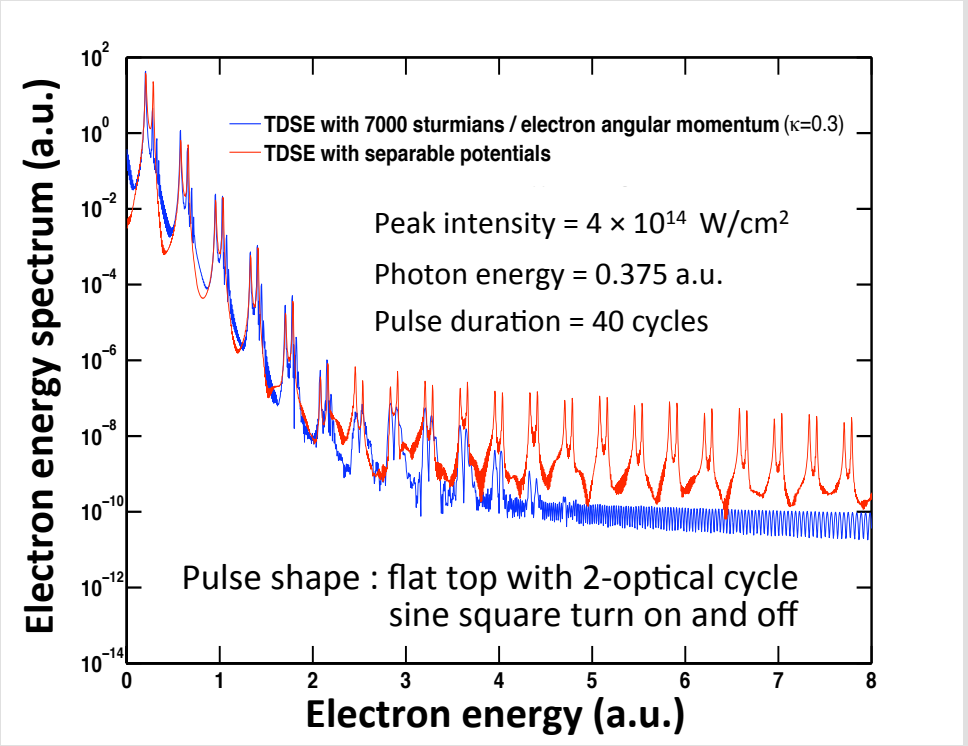
Substitute $\phi^{(\pm)}(\mathbf{p},\mathbf{k})$ in the expression of $\chi_n^{(\pm)}(\mathbf{k})$ to get these coefficients $\chi_n^{(\pm)}(\mathbf{k})$

Validity of the model

The present formulation is not gauge invariant but gives results that are qualitatively and quantitatively in very good agreement with those obtained by solving the TDSE in situations where the dynamics implies a small number of bound states.

- For $\omega > I_p$ and $U_p < \omega$ (perturbative regime) the one-bound state model provides accurate yields.
- At the *1s-2p* resonance frequency, the model reproduces well the Rabi oscillations of the populations and the corresponding Autler-Townes doublets in the electron energy spectrum.

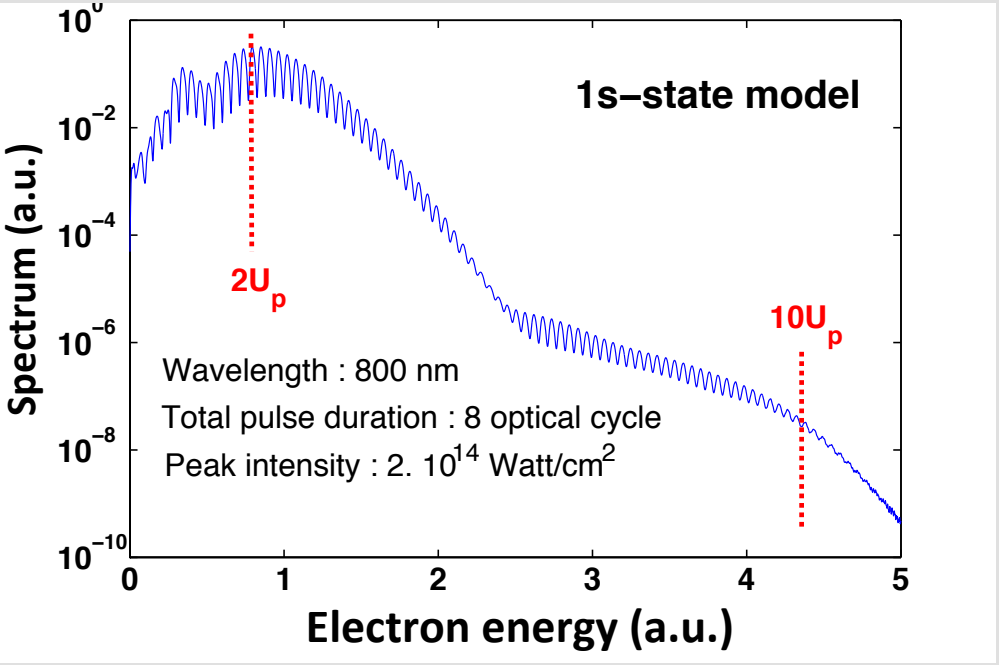




- Exact 1s- and 2p-wave function ➡ exact 1s-2p oscillator strength ➡ exact Rabi frequency of 1s and 2p populations.

- The spectrum is correct over 10 orders of magnitude. The extra peaks in each Autler-Townes doublet are due to np states with n>2. The discrepancies for energies > 2.5 a.u. result from the approximate description of the continuum.

- In the low frequency limit, the ATI spectra exhibits all typical characteristics of the strong field spectra.

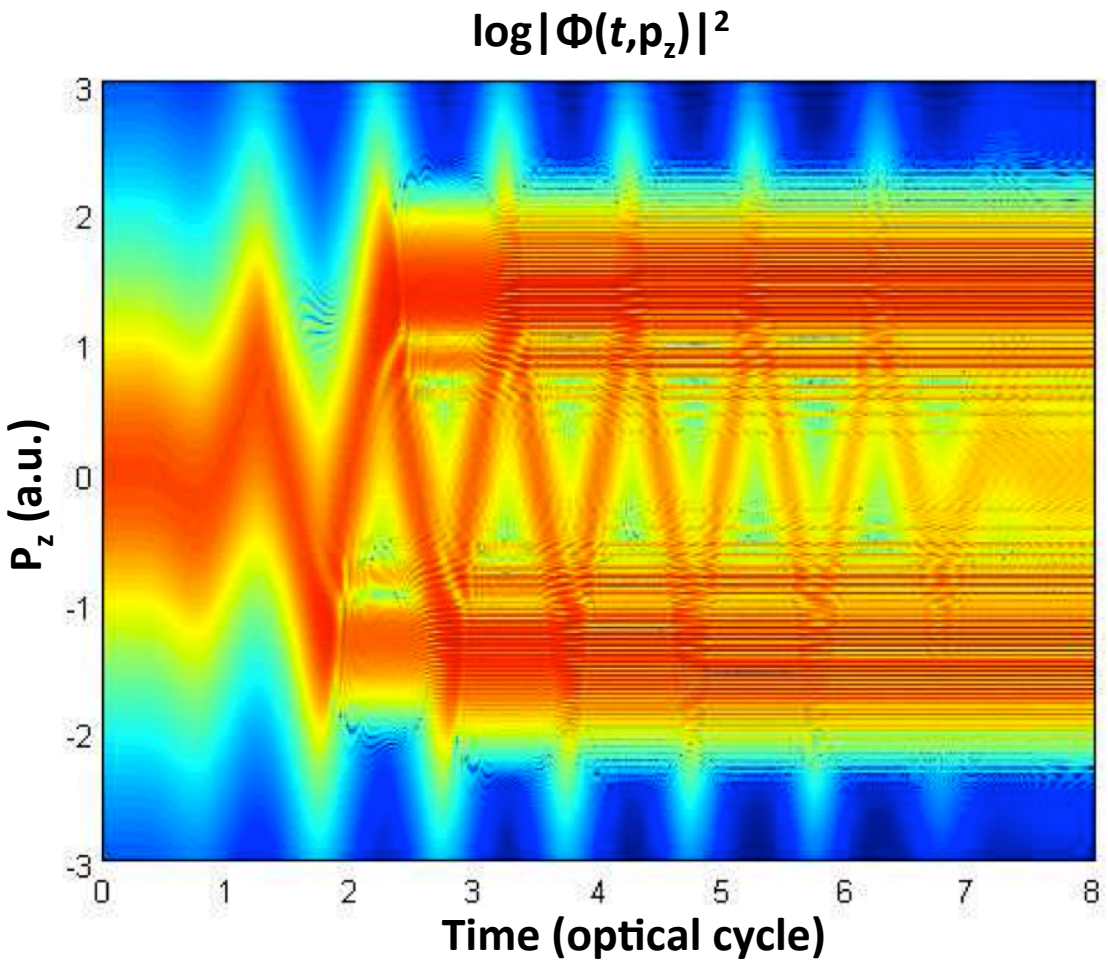


What do we learn about the dynamics with a one-bound state model ?

Preliminary remark

$$\langle \nabla_r V \rangle \approx 0 \quad \Rightarrow \quad \langle \boldsymbol{p} \rangle \approx \text{constant}$$

$$\langle v_z \rangle_{\text{bound}} \approx 0 \quad \Rightarrow \quad \langle p_z \rangle_{\text{bound}} \approx -A(t)$$



I_{peak} = 2.5 x 10^{14} Watt/cm^2

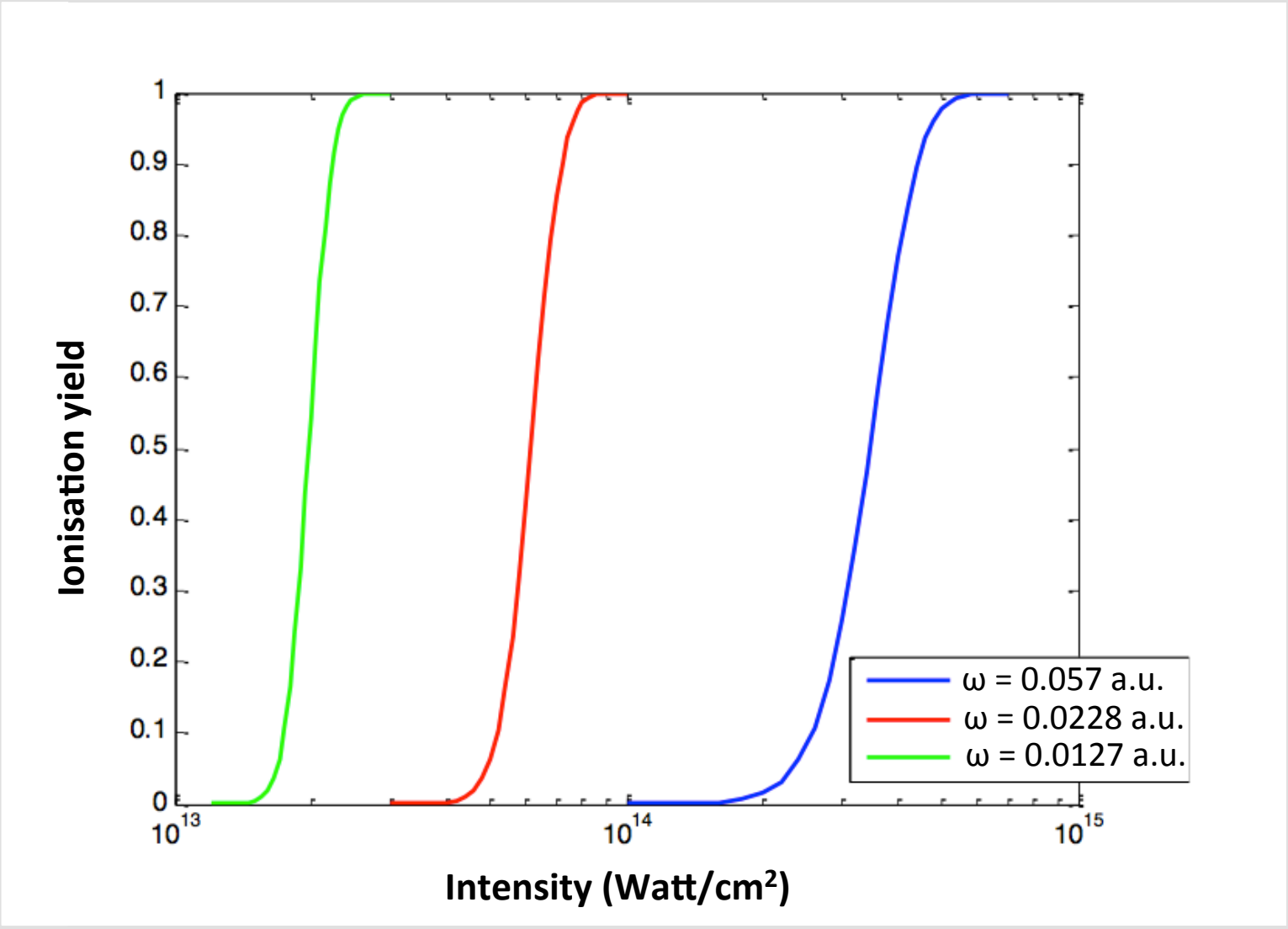
Pulse duration = 8 cycle

Photon energy = 0.057 a.u.

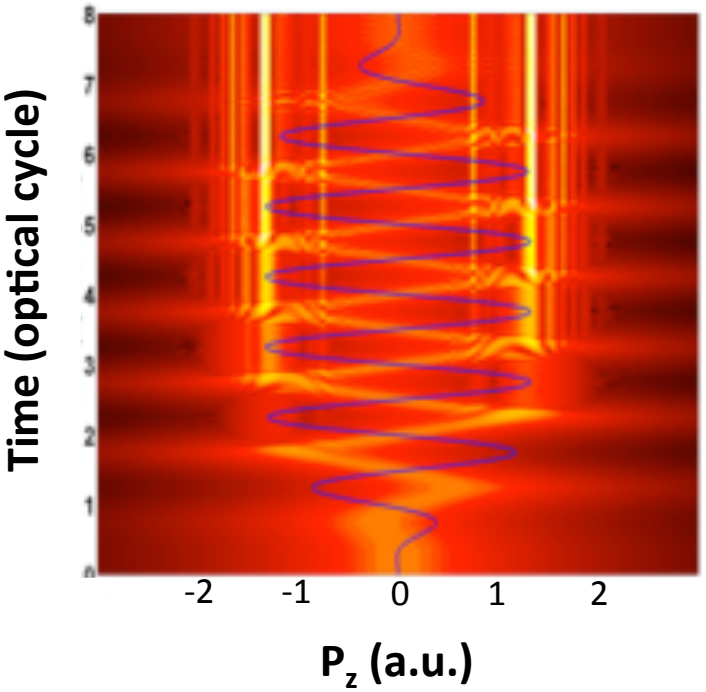
- The oscillating part corresponds to the probability density associated to the 1s state. It oscillates in phase opposition to the vector potential.
- The horizontal stripes (p_z=constant) corresponds to Volkov states (in fact linear superposition of Volkov states and the 1s state)

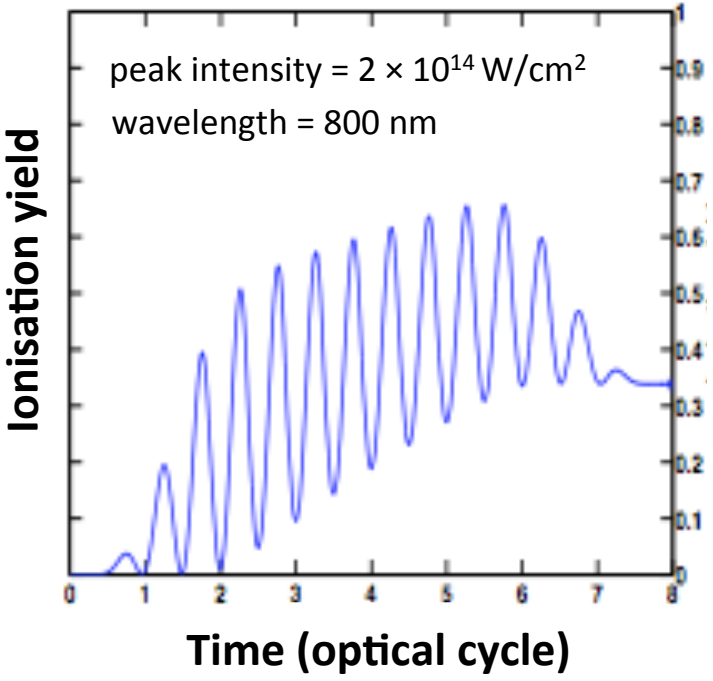
- Ionisation starts at p_z=-A(t_{ion}) where t_{ion} ≈2 optical cycle but the stripes start one cycle further ➡ the ATI peaks result from the interference between wave packets emitted during two successive cycles. The stripes = interference fringes.
- Electrons are emitted around the maximum of A(t) so when the electric field is minimum.

Two-cycle pulse (carrier phase = 0°)

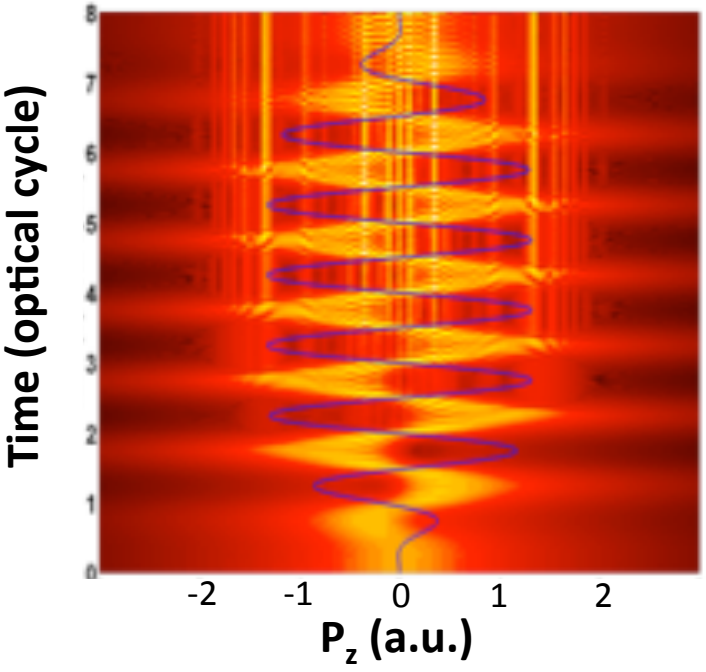


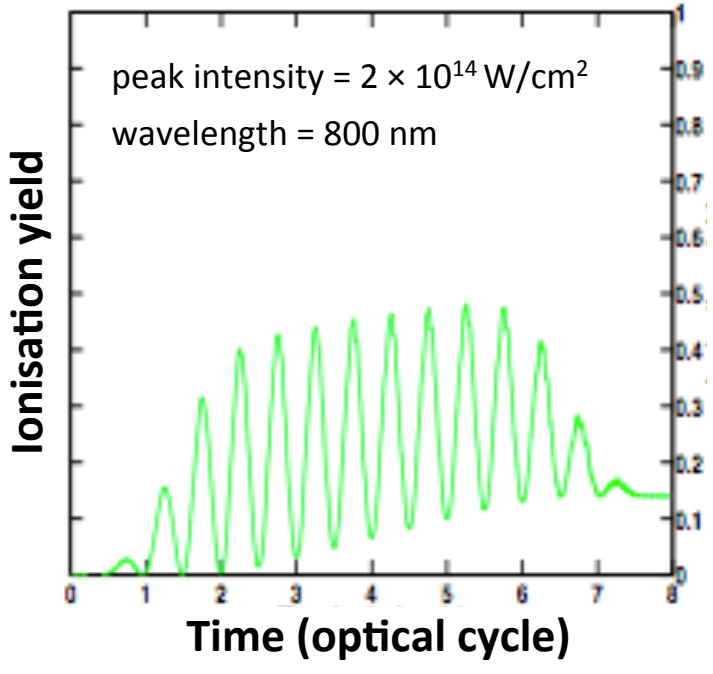
1s - model





(1s, 2s, 2p) - model



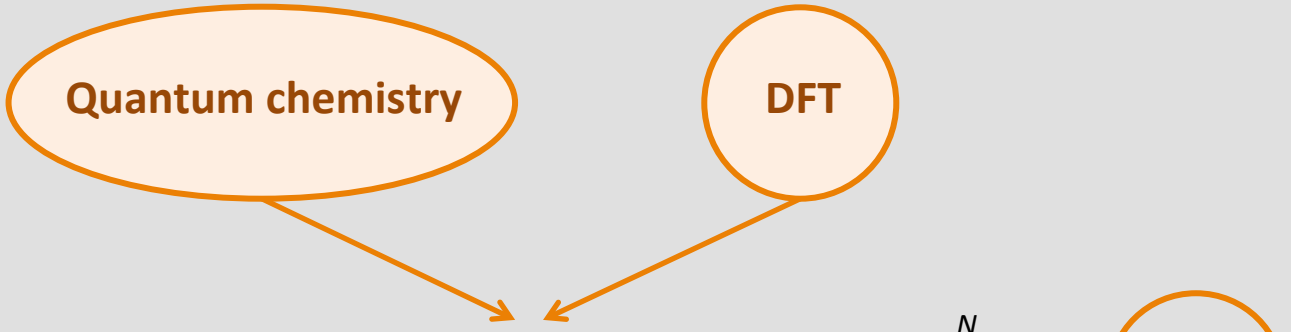


Generalization of the model to complex systems exposed to ultra-short pulses

Main idea

To build separable potentials from the wave functions associated to the essential states or calculated in the mean field approximation

Illustration : molecular system



$$\psi_i(\mathbf{r},\mathbf{R}) = \sum_i d_{ij} N_{lmn}(\alpha_{ij}) x^l y^m z^n \exp(-\alpha_{ij}|\mathbf{r}-\mathbf{R}|^2)$$

Procedure

- $\Phi_\gamma(\mathbf{r},\mathbf{R}) \xrightarrow{\text{F.T.}} \widetilde{\Phi}_\gamma(\mathbf{p},\mathbf{R}) = \sum_{i=1}^{N} c_i(\gamma)\widetilde{\psi}_i(\mathbf{p},\mathbf{R})$

- To build the nonlocal and separable molecular potentials from the molecular orbitals in momentum space as in the case of atomic hydrogen.

$$V(\mathbf{p},\mathbf{p}';\mathbf{R}) = -\sum_{n=1}^{N} v_n(\mathbf{p};\mathbf{R}) v_n^*(\mathbf{p}';\mathbf{R})$$

- To solve the TDSE

$$\left[i\frac{\partial}{\partial t} - \frac{\mathbf{p}^2}{2} - A(t)(\boldsymbol{e}_z \cdot \mathbf{p}) \right] \widetilde{\Phi}(\mathbf{p};\mathbf{R},t) + \sum_{n=1}^{N} v_n(\mathbf{p};\mathbf{R}) \underbrace{\int \frac{d^3\mathbf{p}'}{(2\pi)^3} v_n^*(\mathbf{p}';\mathbf{R}) \widetilde{\Phi}(\mathbf{p}';\mathbf{R},t)}_{F_n(\mathbf{R},t)}$$

Initial condition:

$$\widetilde{\Phi}(\mathbf{p};\mathbf{R},t=0) = \widetilde{\Phi}_{\text{HOMO}}(\mathbf{p};\mathbf{R})$$

$$\mathbf{F}(\mathbf{R},t) = \mathbf{F}_0(\mathbf{R},t) + \int_0^t \mathbf{K}(\mathbf{R};t,\tau)\, \mathbf{F}(\mathbf{R},\tau) d\tau$$

Conclusions and perspectives

- In the long wavelength regime considered here, our results indicate that:
  - the ionisation rate tends to a constant
  - the low energy structures in the ATI spectra results from a hard collision
  - the rich structure present in the momentum distribution near zero seems to result from the ionisation process itself (direct electrons)
  - the polarization of the electron cloud that involves p-states has to be described properly; it prevents the atom from ionizing too quickly
  - there is an obvious analogy with ion-atom collisions

- It is possible to define various families of strong field approximations by introducing some ansatz. In one given family, there is full gauge invariance but not between different families.

- All SFA schemes overestimate systematically the ionization yield in the long wavelength regime because the polarization of the target is only taken into account through continuum p-states.

- The separable potential model clearly confirms the crucial role of the polarization of the target in the long wavelength regime.