Time-dependent density functional theory

Lecture II

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Approximations for the static xc energy functional derived from TDDFT

ADIABATIC CONNECTION FORMULA

$$H(\lambda) = T + \sum_{i=1}^{N} v_\lambda(r_i) + \lambda \frac{e^2}{2} \sum_{\substack{i,k=1 \\ i \neq k}}^{N} \frac{1}{|r_i - r_k|} \qquad 0 \leq \lambda \leq 1$$

$$H(\lambda=1) = T + \sum_{i=1}^{N} v_{nuc}(r_i) + \frac{e^2}{2} \sum_{\substack{i,k=1 \\ i \neq k}}^{N} \frac{1}{|r_i - r_k|}$$

= Hamiltonian of fully interacting system

Choose $v_\lambda(r)$ such that for each λ the ground- state density satisfies $\rho_\lambda(r) = \rho_{\lambda=1}(r)$

Hence $v_{\lambda=0}(r) = v_{KS}(r)$ $v_{\lambda=1}(r) = v_{nuc}(r)$

Determine the response function $\chi^{(\lambda)}(r,r';\omega)$ corresponding to $H(\lambda)$, Then

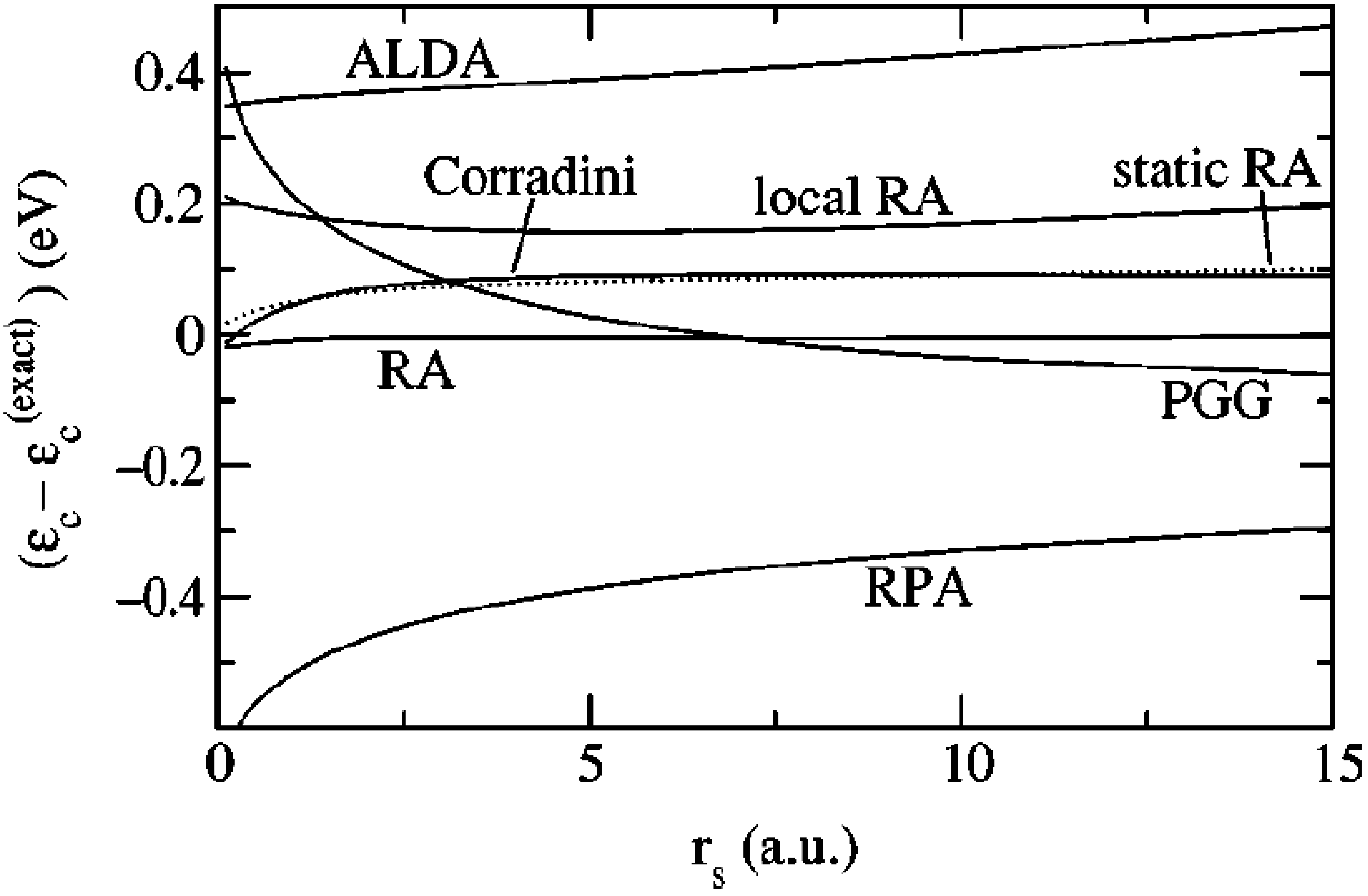
$$\boxed{E_{xc} = -\int_0^1 d\lambda \int_0^\infty \frac{du}{2\pi} \int d^3r \int d^3r' \frac{e^2}{|r-r'|} \left\{ \chi^{(\lambda)}(r,r';iu) + \rho(r)\delta(r-r') \right\}}$$

Second ingredient : TDDFT

$$\boxed{\chi = \chi_s + \chi_s \left[W_{Clb} + f_{xc} \right] \chi}$$

and for $0 \leq \lambda \leq 1$:

$$\boxed{\chi^{(\lambda)} = \chi_s + \chi_s \left[\lambda W_{Clb} + f_{xc}^{(\lambda)} \right] \chi^{(\lambda)}}$$



r_s-dependent deviation of approximate correlation energies from the "exact" correlation energy per electron of the uniform electron gas.

M. Lein, E. K. U. G., J. Perdew, Phys. Rev. B 61, 13431 (2000).

For finite systems, truncate after first iteration:

$$\chi^{(\lambda)} \approx \chi_s + \chi_s [\lambda W_{clb} + f_{xc}^{(\lambda)}]\chi_s$$

plug this approximation into adiabatic connection formula

$\Rightarrow$ Orbital functional for E_c

Resulting Atomic Correlation energies (in a.u.)

atom	LDA	new fctl	exact
He	-0.111	-0.048	-0.042
Be	-0.224	-0.13	-0.096
Ne	-0.739	-0.41	-0.394
Ar	-1.423	-0.67	-0.72

Resulting v.d.W. coefficients C_6

Lein, Dobson, EKUG, J. Comp. Chem. (‘99)

system	Calculated C_6	experiment
He-He	1.639	1.458
He-Ne	3.424	3.029
Ne-Ne	7.284	6.383
Li-Li	1313	1390
Li-Na	1453	1450
Na-Na	1614	1550
H-He	2.995	2.82
H-Ne	5.976	5.71
H-Li	64.96	66.4
H-Na	75.4	71.8

Time-Dependent Electron Localization Function
(TD-ELF)

Time-Dependent Electron Localization Function (TD-ELF)

GOAL

Time-resolved visualization of the breaking and formation of chemical bonds.

Time-Dependent Electron Localization Function (TD-ELF)

How can one give a rigorous mathematical meaning to chemical concepts such as

- **Single, double, triple bonds**
- **Lone pairs**

Note:

- **Density $\rho_\sigma(r)$ is not useful!**
- **Orbitals are ambiguous (w.r.t. unitary transformations)**

$$D_\sigma(\vec{r},\vec{r}') = \sum_{\sigma_3\sigma_4\ldots\sigma_N} \int d^3r_3 \ldots \int d^3r_N \left|\Psi\left(\vec{r}\sigma, \vec{r}'\sigma, \vec{r}_3\sigma_3 \ldots, \vec{r}_N\sigma_N\right)\right|^2$$

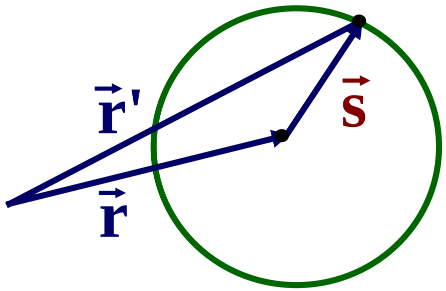
= diagonal of two-body density matrix

= probability of finding an electron with spin σ at $\vec{r}$ and another electron with the same spin at $\vec{r}'$.

$$P_\sigma(\vec{r},\vec{r}') := \frac{D_{\sigma\sigma}(\vec{r},\vec{r}')}{\rho_\sigma(\vec{r})}$$

= conditional probability of finding an electron with spin σ at $\vec{r}'$ if we know with certainty that there is an electron with the <u>same</u> spin at $\vec{r}$.

Coordinate transformation



If we know there is an electron with spin σ at $\vec{\mathbf{r}}$, then $P_\sigma\left(\vec{\mathbf{r}},\vec{\mathbf{r}}+\vec{\mathbf{s}}\right)$ is the (conditional) probability of finding another electron at $\vec{\mathbf{r}}+\vec{\mathbf{s}}$

Spherical average $$p_\sigma\left(\vec{r},|\vec{s}|\right)=\frac{1}{4\pi}\int_0^\pi \sin\theta d\theta\int_0^{2\pi} d\varphi P_\sigma\left(\vec{r},|\vec{s}|,\theta,\varphi\right)$$

If we know there is an electron with spin σ at $\vec{\mathbf{r}}$, then $p_\sigma\left(\vec{\mathbf{r}},\mathbf{s}\right)$ is the conditional probability of finding another electron at the distance **s** from .

Expand in a Taylor series:

$$p_\sigma\left(\vec{r},s\right)=\underbrace{p_\sigma\left(\vec{r},0\right)}_{0}+\underbrace{\left.\frac{dp_\sigma\left(\vec{r},s\right)}{ds}\right|_{s=0}}_{0}\cdot s+\frac{1}{3}C_\sigma\left(\vec{r}\right)s^2$$

The first two terms vanish.

$C_\sigma(\vec{r})$ is a measure of electron localization.

Why? $C_\sigma(\vec{r})$, being the s^2-coefficient, gives the probability of finding a second <u>like-spin</u> electron <u>very near</u> the reference electron. If this probability <u>very near</u> the reference electron is low then this reference electron must be very localized.

$C_\sigma(\vec{r})$ small means strong localization at $\vec{r}$

$\mathbf{C}_\sigma$ is always ≥ 0 (because $\mathbf{p}_\sigma$ is a probability) and $\mathbf{C}_\sigma(\vec{\mathbf{r}})$ is not bounded from above.

Define as a useful visualization of localization
(A.D. Becke, K.E. Edgecombe, JCP <u>92</u>, 5397 (1990))

$$\boxed{\mathrm{ELF} = \frac{1}{1+\left(\frac{C_\sigma(\vec{r})}{C_\sigma^{\mathrm{uni}}(\vec{r})}\right)^2}}$$

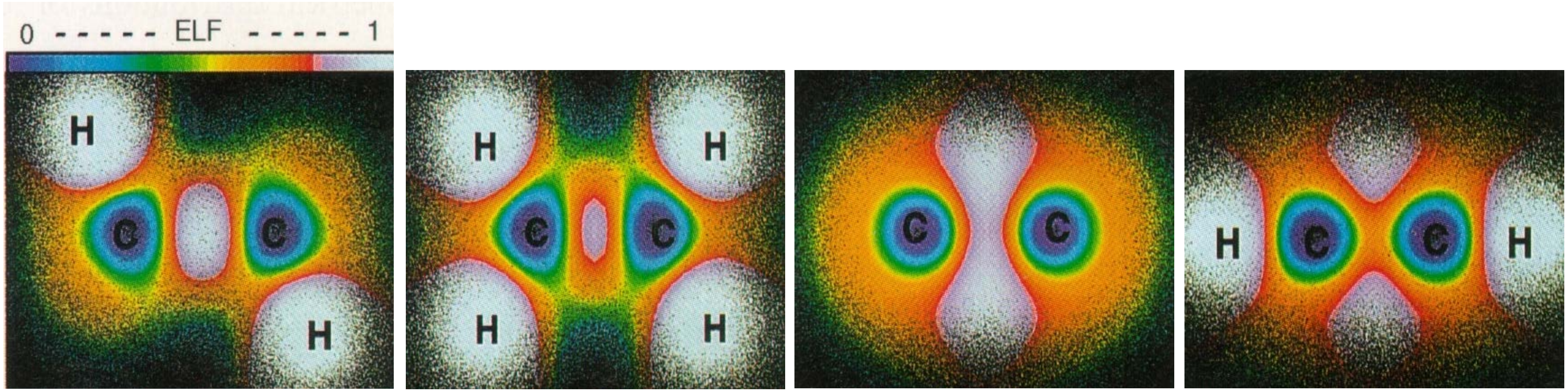
where

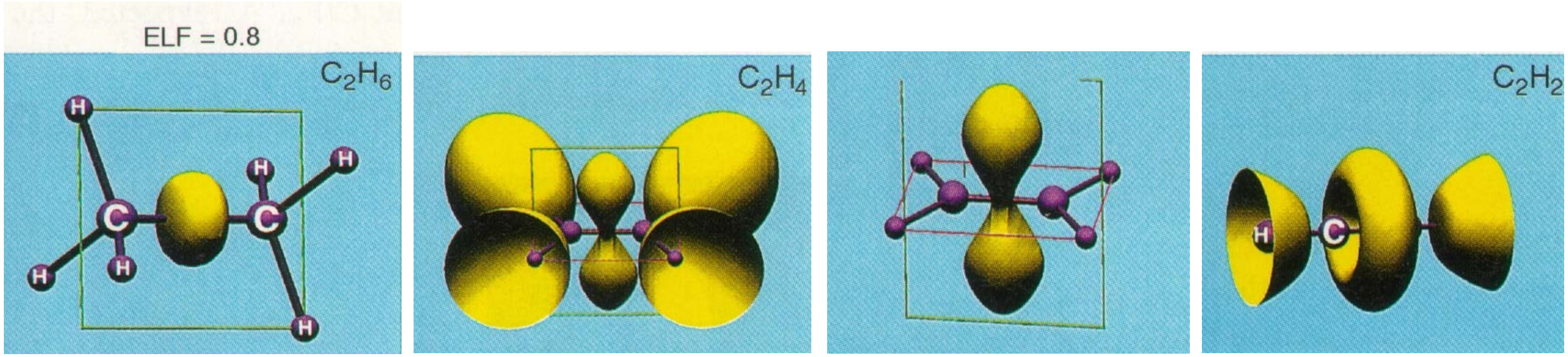
$$C_\sigma^{\mathrm{uni}}(\vec{r}) = \frac{3}{5}\left(6\pi^2\right)^{2/3}\rho_\sigma^{5/3}(\vec{r}) = \tau_\sigma^{\mathrm{uni}}(\vec{r})$$

is the kinetic energy density of the uniform gas.

Advantage: ELF is dimensionless and $0 \leq \mathrm{ELF} \leq 1$

ELF

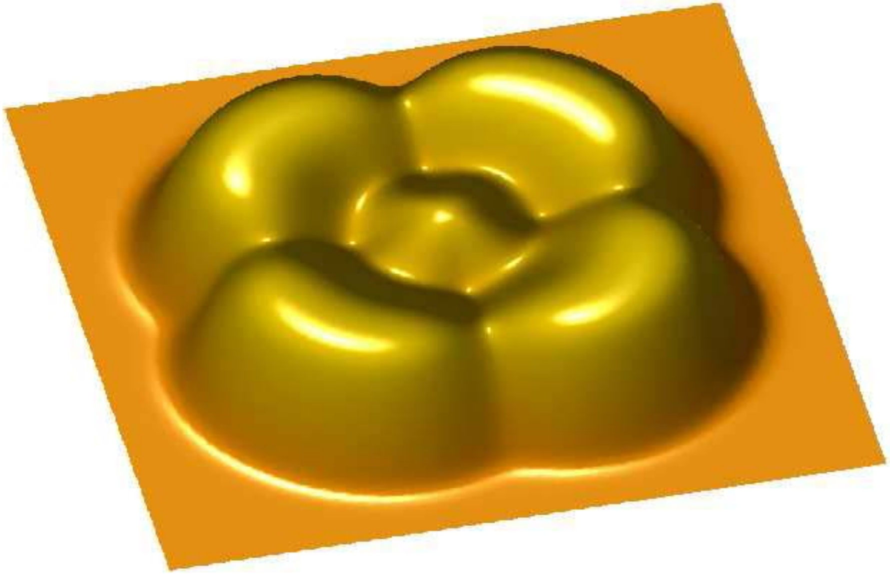




A. Savin, R. Nesper, S. Wengert, and T. F. Fässler, Angew. Chem. Int. Ed. 36, 1808 (1997)

12-electron 2D quantum dot with four minima

Density



ELF

E. Räsänen, A. Castro and E.K.U. Gross, Phys. Rev. B 77, 115108 (2008).

For a determinantal wave function one obtains

in the static case:

$$C_\sigma^{\mathrm{det}}(\vec{r}) = \sum_{i=1}^{N_\sigma} \left|\nabla\varphi_{i\sigma}(\vec{r})\right|^2 - \frac{1}{4}\frac{\left(\nabla\rho_\sigma(\vec{r})\right)^2}{\rho_\sigma(\vec{r})}$$

(A.D. Becke, K.E. Edgecombe, JCP 92, 5397 (1990))

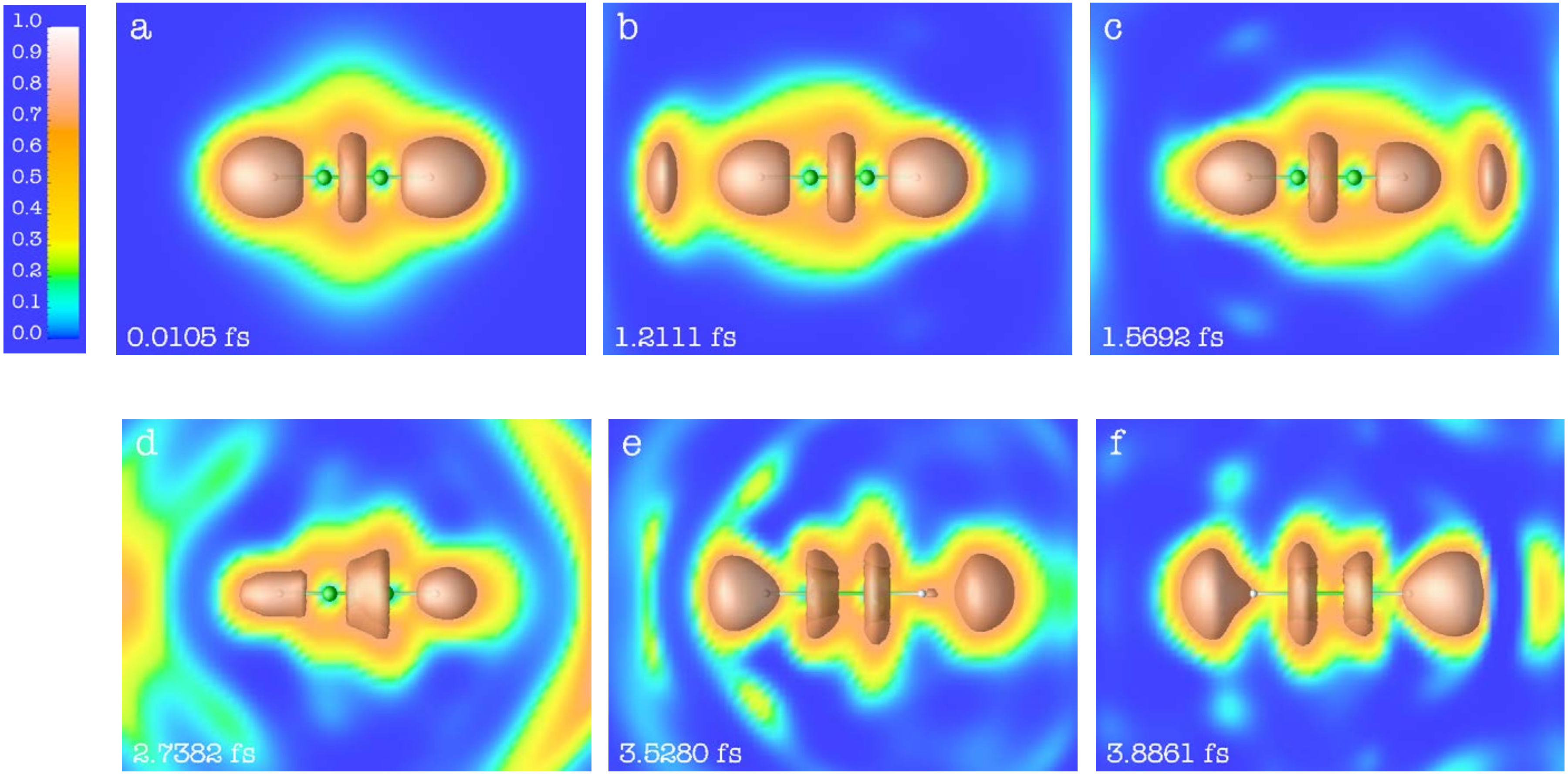
in the time-dependent case:

$$\boxed{C_\sigma^{\mathrm{det}}(\vec{r},t) = \sum_{i=1}^{N_\sigma} \left|\nabla\varphi_{i\sigma}(\vec{r},t)\right|^2 - \frac{1}{4}\frac{\left(\nabla\rho_\sigma(\vec{r},t)\right)^2}{\rho_\sigma(\vec{r},t)} - j_\sigma(\vec{r},t)^2 \big/ \rho_\sigma(\vec{r},t)}$$

(T. Burnus, M. Marques, E.K.U.G., PRA (Rapid Comm) 71, 010501 (2005))

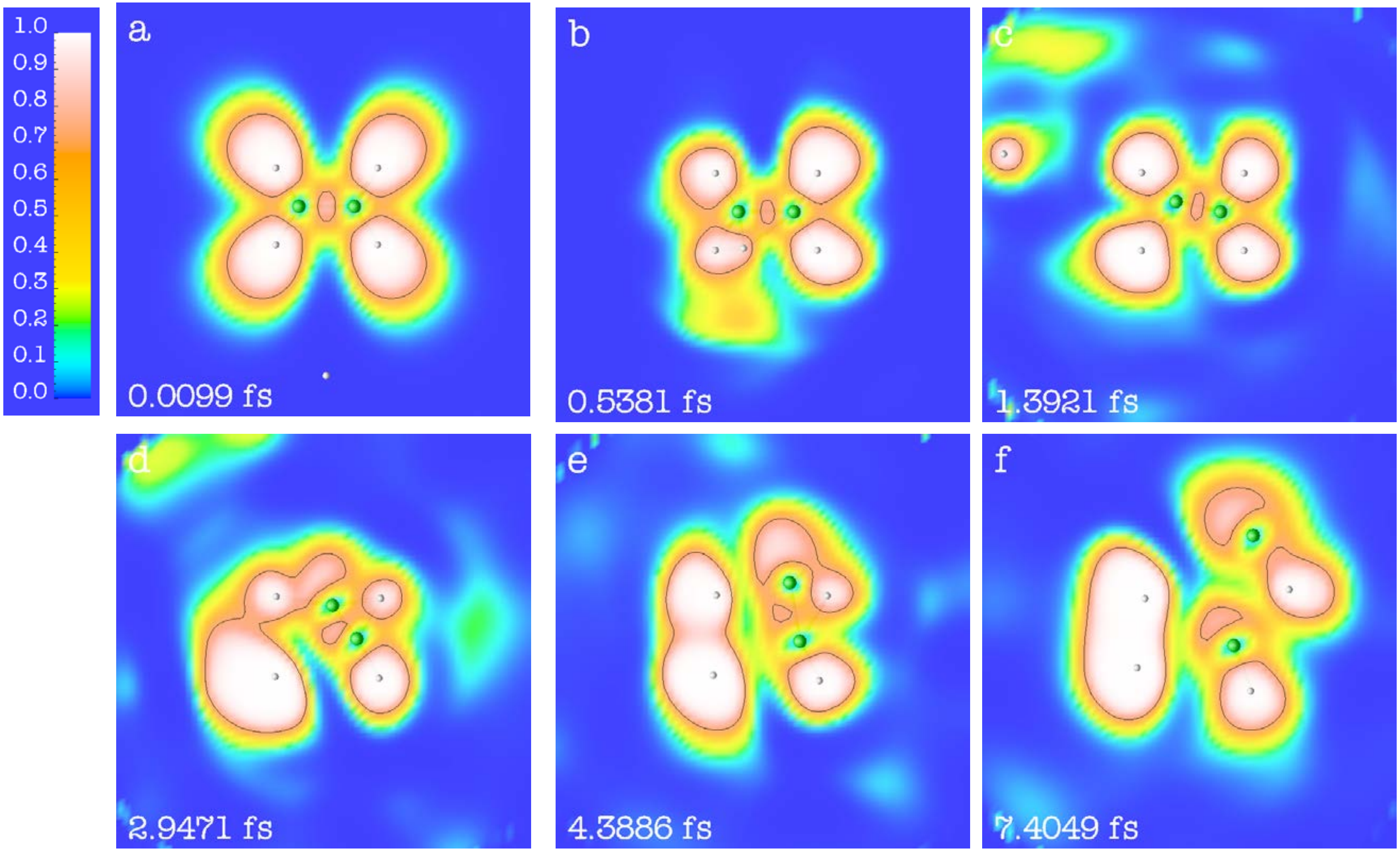
TDELF for acetylene in strong laser field

($\hbar\omega = 17.15$ eV, $I = 1.2\times10^{14}$ W/cm^2)



TDELF for scattering process

2 keV proton colliding with ethylene



Optimal control using short laser pulses

Review Article on Quantum Optimal Control Theory:
J. Werschnik, E.K.U. Gross, J. Phys. B 40, R175-R211 (2007)

Optimal Control Theory (OCT)

Normal question:

What happens if a system is exposed to a given laser pulse?

Inverse question (solved by OCT):

Which is the laser pulse that achieves a prescribed goal (target)?

possible targets:

a) system should end up in a given final state ϕ_f at the end of the pulse

b) wave function should follow a given trajectory in Hilbert space

c) density should follow a given classical trajectory r(t)

Optimal control of static targets
(standard formulation)

For <u>given</u> target state Φ_f , maximize the functional:

$$J_1 = \left| \langle \Psi(T) | \Phi_f \rangle \right|^2 = \langle \Psi(T) | \Phi_f \rangle \langle \Phi_f | \Psi(T) \rangle = \left\langle \Psi(T) \middle| \hat{O} \middle| \Psi(T) \right\rangle$$

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with the constraints:

$$J_2 = -\alpha \left[\int_0^T dt\, \varepsilon^2(t) - E_0 \right]$$

E_0 = *given* fluence

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E_0 = given fluence

$$J_3[\varepsilon, \Psi, \chi] = -2\,\mathrm{Im} \int_0^T dt \langle \chi(t) | -i\partial_t - [\hat{T} + \hat{V} - \mu\varepsilon(t)] | \Psi(t) \rangle$$

Optimal control of static targets (standard formulation)

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Optimal control of static targets (standard formulation)

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GOAL: Maximize $J = J_1 + J_2 + J_3$

Set the total variation of $J = J_1 + J_2 + J_3$ equal to zero:

Control equations

1. Schrödinger equation with initial condition:

$$\delta_\chi J = 0 \rightarrow \boxed{i\partial_t \psi(t) = \hat{H}(t)\psi(t), \quad \psi(0) = \phi}$$

2. Schrödinger equation with final condition:

$$\delta_\psi J = 0 \rightarrow \boxed{i\partial_t \chi(t) = \hat{H}(t)\chi(t), \quad \chi(T) = \hat{O}\psi(T)}$$

3. Field equation:

$$\delta_\varepsilon J = 0 \rightarrow \boxed{\varepsilon(t) = \frac{1}{\alpha}\operatorname{Im}\left\langle \chi(t) \middle| \hat{\mu} \middle| \psi(t) \right\rangle}$$

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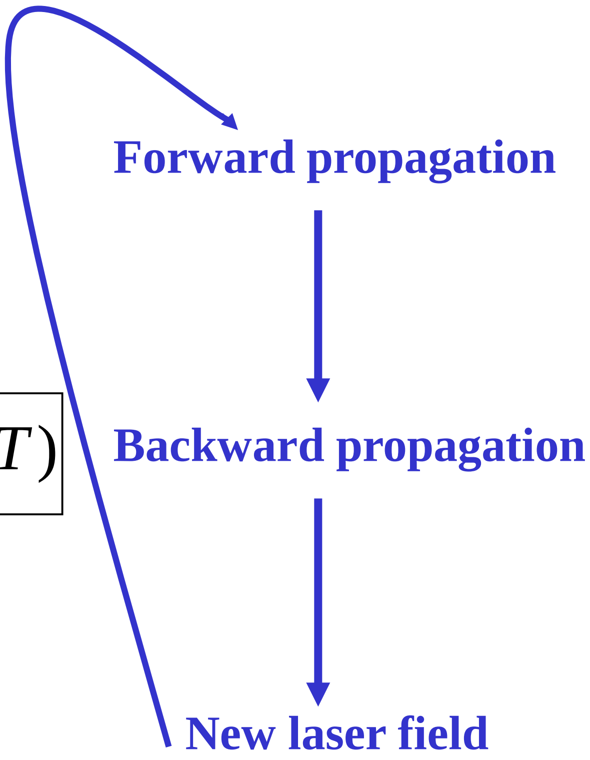
Algorithm



Forward propagation

Backward propagation

New laser field

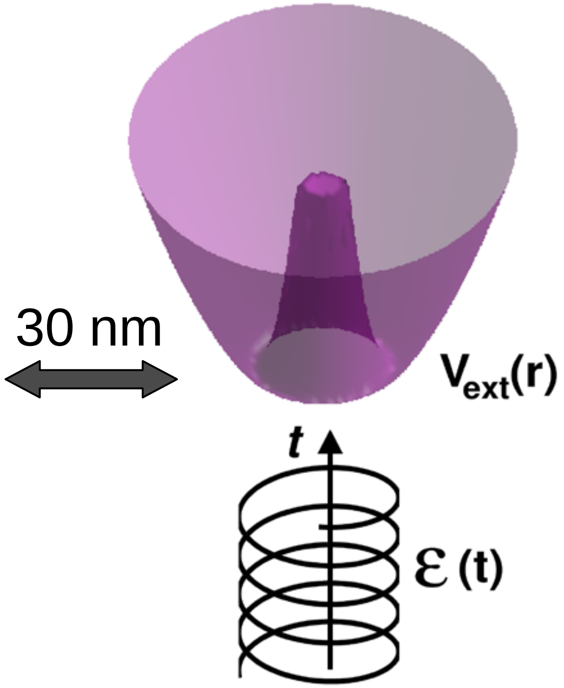
Algorithm monotonically convergent: W. Zhu, J. Botina, H. Rabitz, JCP 108, 1953 (1998))

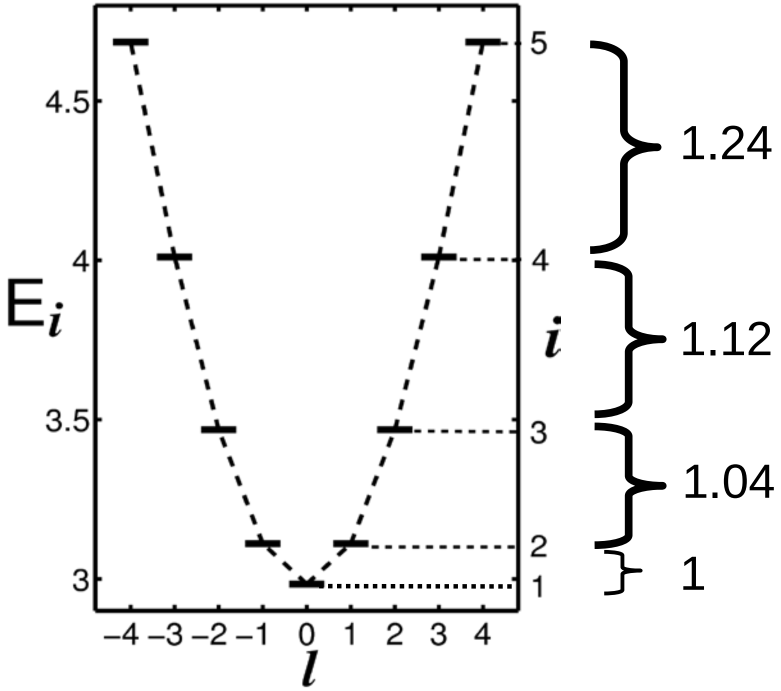
Quantum ring: Control of circular current

TDSE: $$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r},t) = \left[\hat{H}_0 + e\,\mathbf{r}\,\epsilon(t)\right]\Psi(\mathbf{r},t)$$

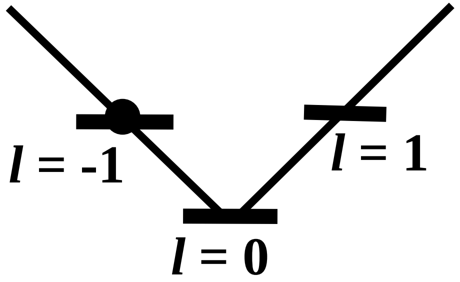
$$\hat{H}_0 = -\frac{\hbar^2}{2m^*}\nabla^2 + \underbrace{\frac{1}{2}m^*\omega_0^2 r^2 + V_0 e^{-r^2/d^2}}$$

$$\epsilon(t) = (\epsilon_x(t), \epsilon_y(t))$$

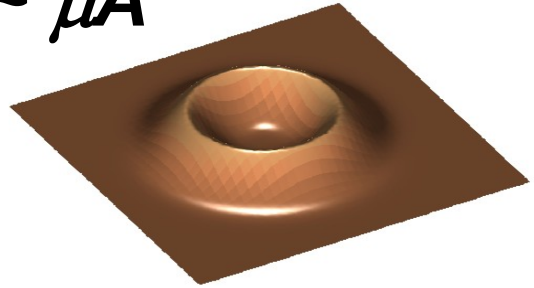




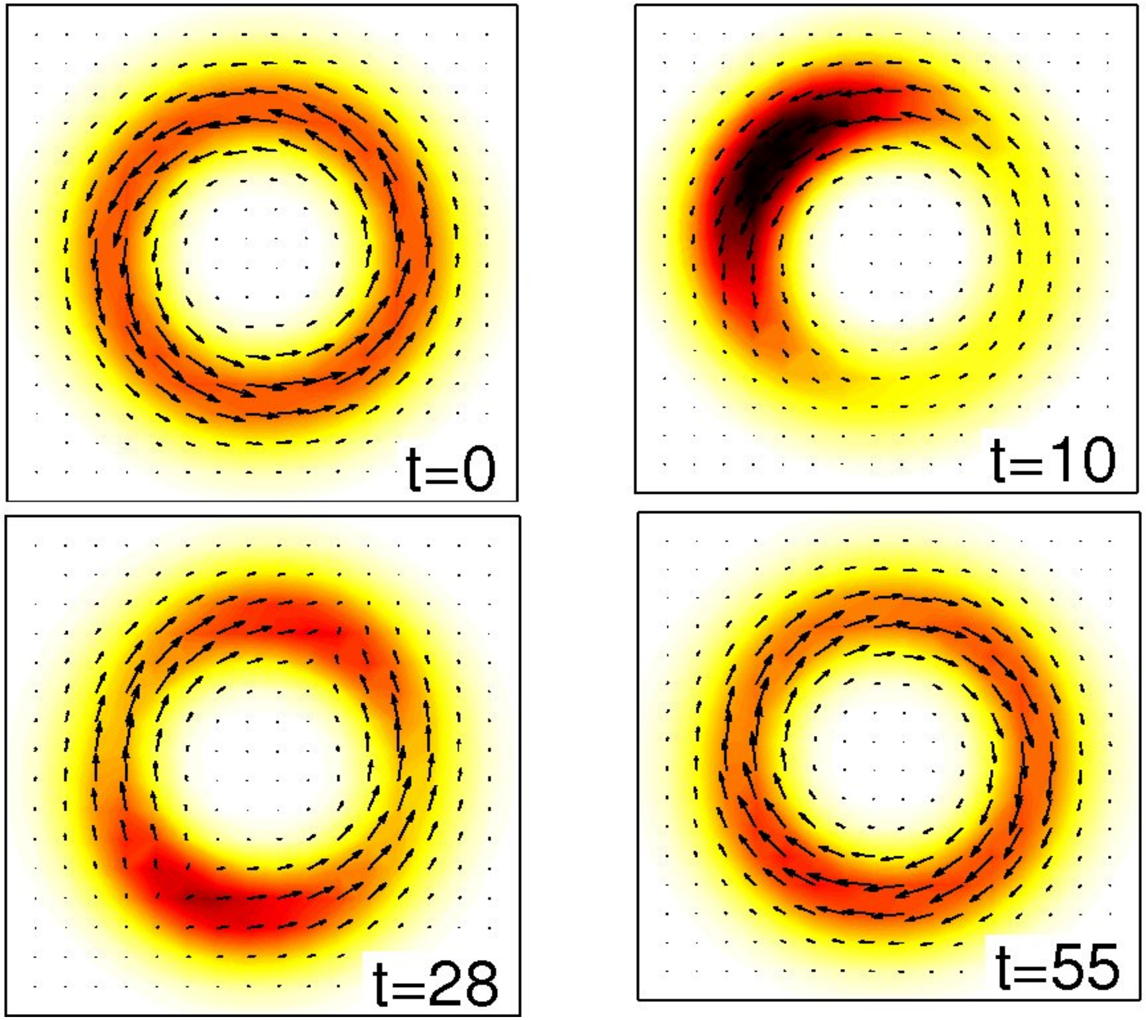
Control of currents



I* ~ *μA



$|\psi(t)|^2$ and $\mathbf{j}\ (t)$



E. Räsänen, A. Castro, J. Werschnik, A. Rubio, E.K.U.G., PRL 98, 157404 (2007)

OCT of ionization

- Calculations for 1-electron system H_2^+ in 3D
- Restriction to ultrashort pulses ($T<5$fs)
  - nuclear motion can be neglected
- Only <u>linear</u> polarization of laser (parallel or perpendicular to molecular axis)
- Look for enhancement of ionization by pulse-shaping only, keeping the time-integrated intensity (fluence) fixed

<u>Control target to be maximized</u>:

$$J_1 = \left\langle \Psi(T) \middle| \hat{O} \middle| \Psi(T) \right\rangle$$

with $$\hat{O} = \hat{1} - \sum_{i}^{\text{bound}} \left| \varphi_i \right\rangle \left\langle \varphi_i \right|$$

Standard OCT algorithm (forward-backward propagation) does not converge:

Acting with $\hat{O}$ before the backward-propagation eliminates the smooth (numerically friendly) part of the wave function.

Instead of forward-backward propagation, parameterize the laser pulse to be optimized in the form

$$\in(t) = f(t)\cos(\omega_0 t) \ ,$$ with $\omega_0 = 0.114$ a.u. ($\lambda = 400$ nm)

$$f(t) = \sum_{n=1}^{N}\left[f_n\sqrt{\frac{2}{T}}\cos(\omega_n t) + g_n\sqrt{\frac{2}{T}}\sin(\omega_n t)\right] \ ,$$ with $\omega_n = 2\pi n/T$

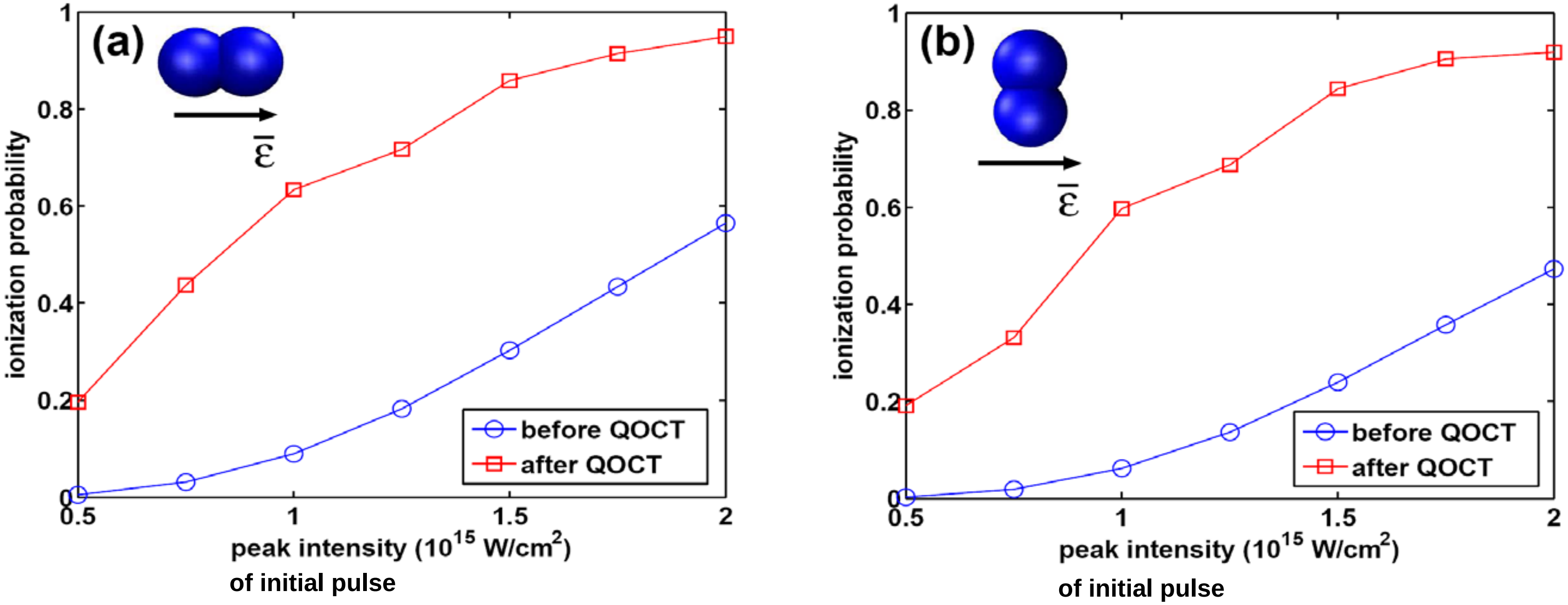
Choose N such that maximum frequency is $2\omega_0$ or $4\omega_0$. T is fixed to 5 fs.

Maximize J_1 ($f_1 \ldots f_N, g_1 \ldots g_N$) directly with constraints:

$$\text{(i)} \quad f(0) = f(T) = 0 \quad \Rightarrow \sum_{n=1}^{N} f_n = 0$$

$$\text{(ii)} \quad \int_0^T dt\,\varepsilon^2(t) = E_0.$$

using algorithm NEWUOA (M.J.D. Powell, *IMA J. Numer. Analysis* 28, 649 (2008))



Ionization probability for the initial (circles) and the optimized (squares) pulse as function of the peak intensity of the initial pulse.

Pulse length and fluence is kept fixed during the optimization.

OPTIMAL CONTROL OF TIME-DEPENDENT TARGETS

Maximize $J = J_1 + J_2 + J_3$

$$J_1[\Psi] = \frac{1}{T}\int_0^T dt \left\langle \Psi(t) \middle| \hat{O}(t) \middle| \Psi(t) \right\rangle$$

$$J_2 = -\alpha\left[\int_0^T dt\, \varepsilon^2(t) - E_0\right]$$

$$J_3[\varepsilon, \Psi, \chi] = -2\,\mathrm{Im}\int_0^T dt \left\langle \chi(t) \middle| -i\partial_t - \left[\hat{T} + \hat{V} - \mu\varepsilon(t)\right] \middle| \Psi(t) \right\rangle$$

Set the total variation of $J = J_1 + J_2 + J_3$ equal to zero:

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$$\delta_\chi J = 0 \rightarrow \boxed{i\partial_t \psi(t) = \hat{H}(t)\psi(t), \quad \psi(0) = \phi}$$

2. Schrödinger equation with final condition:

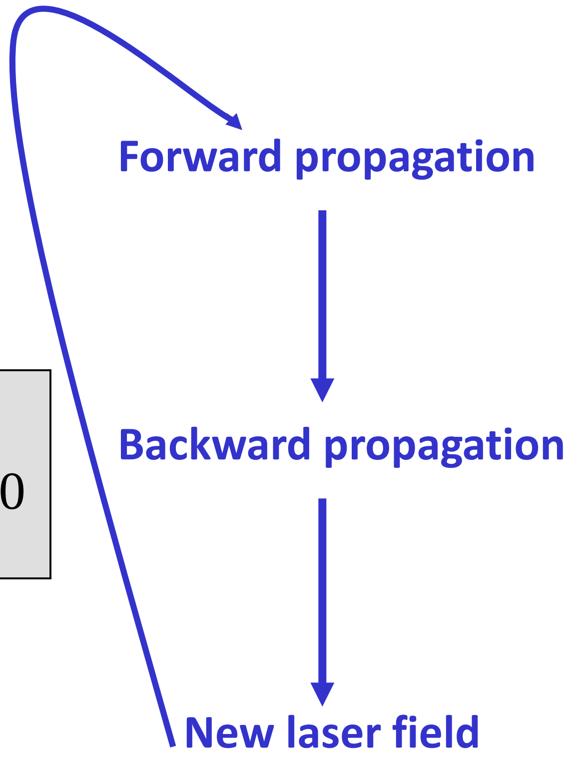
Inhomogenous TDSE :

$$\delta_\psi J = 0 \rightarrow \boxed{\left[i\partial_t - \hat{H}(t)\right]\chi(t) = -\frac{i}{T}\hat{O}(t)\psi(t), \quad \chi(T) = 0}$$

3. Field equation:

$$\delta_\varepsilon J = 0 \rightarrow \boxed{\varepsilon(t) = \frac{1}{\alpha}\,\mathrm{Im}\left\langle \chi(t)\middle|\hat{\mu}\middle|\psi(t)\right\rangle}$$

Algorithm



Forward propagation

Backward propagation

New laser field

Algorithm monotonically convergent:
I. Serban, J. Werschnik, E.K.U.G., Phys. Rev. A 71, 053810 (2005)

Control of path in Hilbert space

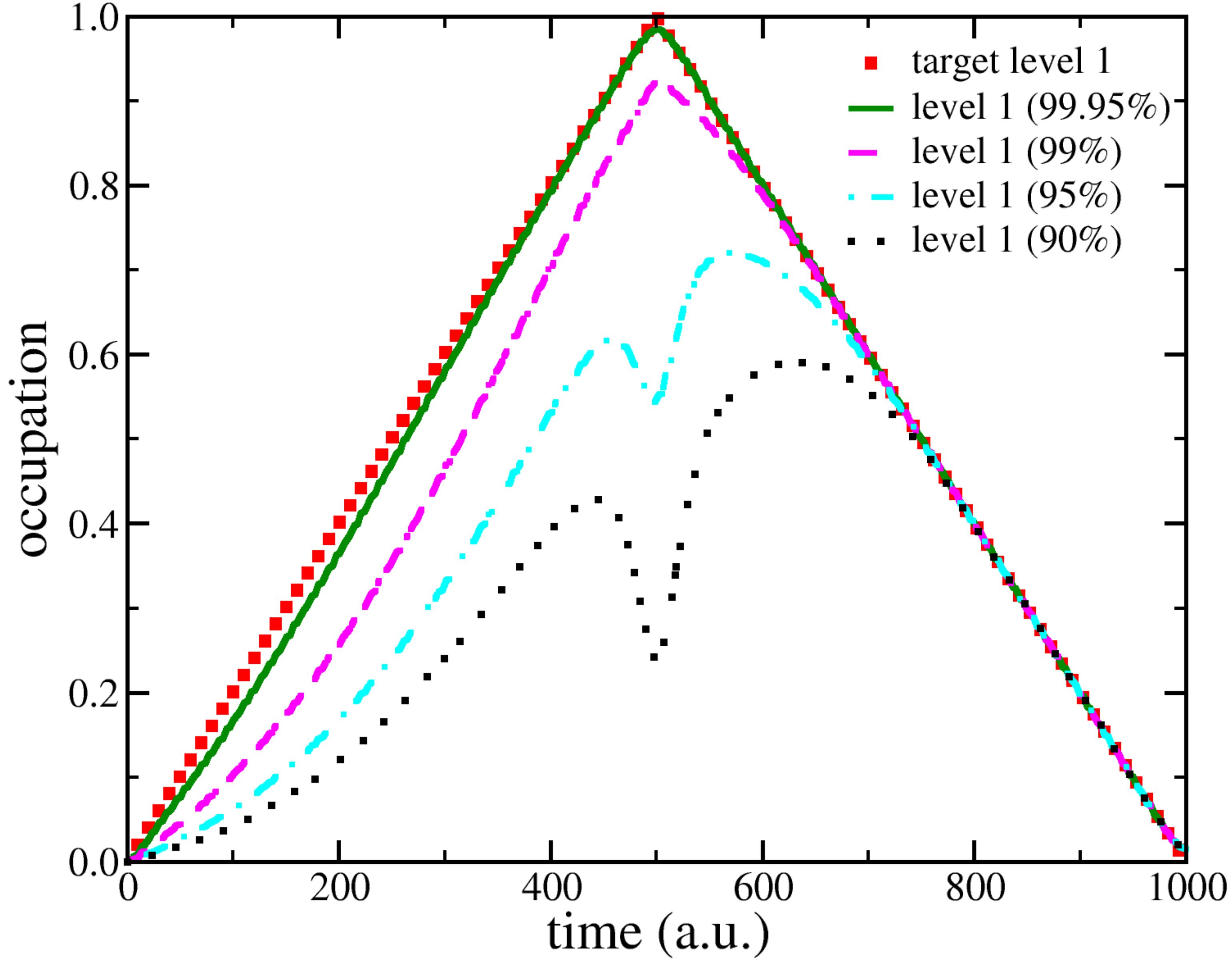
I. Serban, J. Werschnik, E.K.U.G. Phys. Rev. A 71, 053810 (2005)

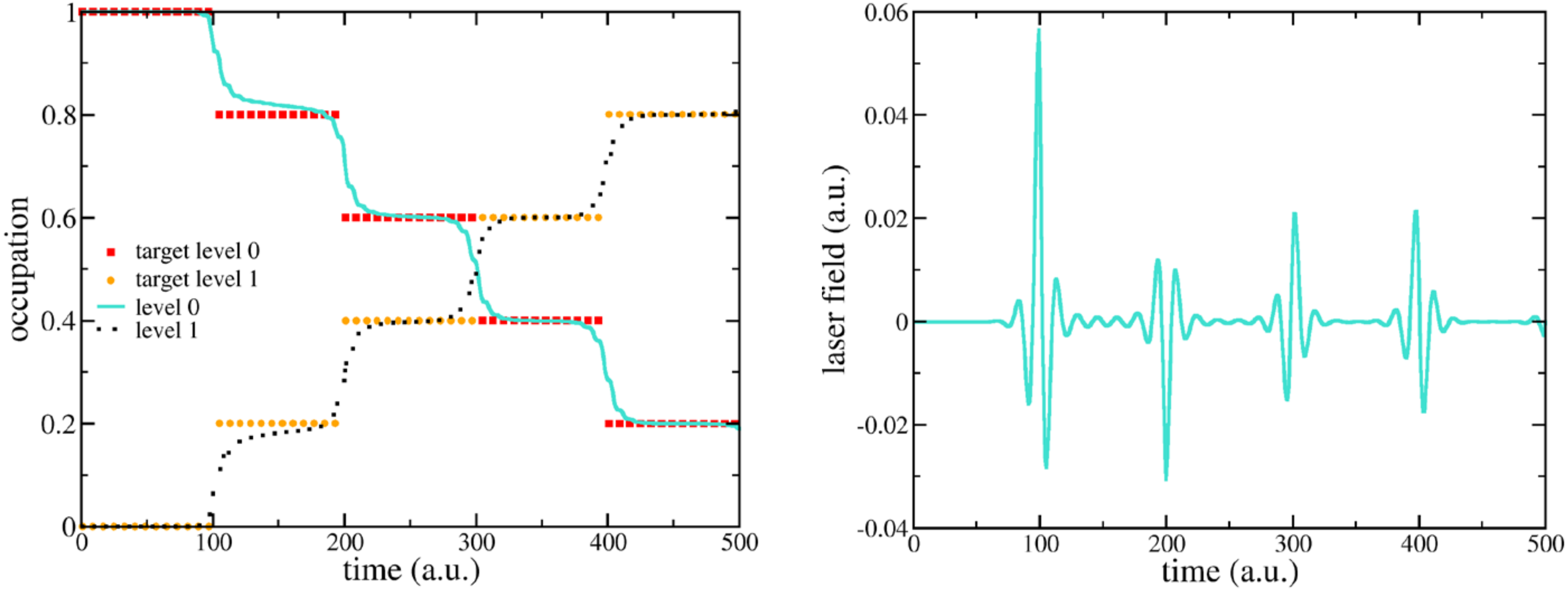
$$\hat{O}(t) = |\Phi(t)\rangle\langle\Phi(t)|$$

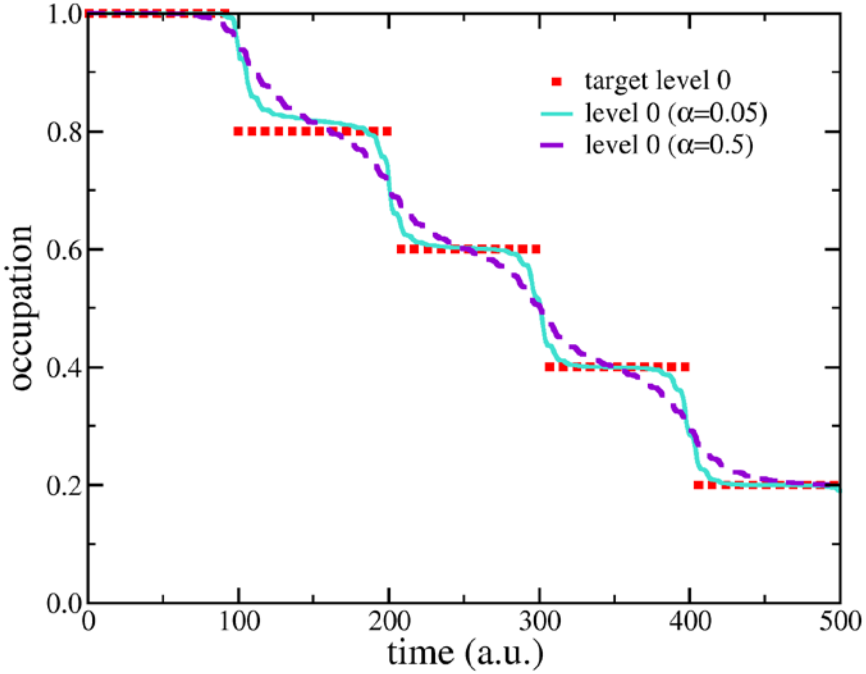
with $$|\Phi(t)\rangle = \alpha_0(t)e^{-i\varepsilon_0 t}|0\rangle + \alpha_1(t)e^{-i\varepsilon_1 t}|1\rangle$$

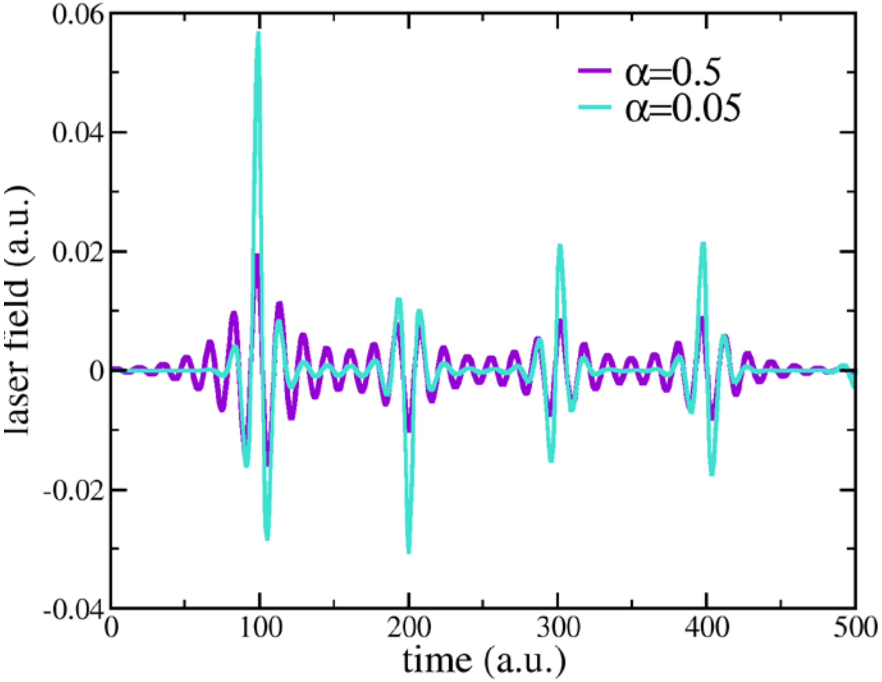
$|\alpha_0(t)|^2$ **<u>given</u> target occupation, and** $|\alpha_1(t)|^2 = 1 - |\alpha_0(t)|^2$

Goal: Find laser pulse that reproduces $|\alpha_o(t)|^2$


occupation
time (a.u.)
0.0
0.2
0.4
0.6
0.8
1.0
0
200
400
600
800
1000
target level 1
level 1 (99.95%)
level 1 (99%)
level 1 (95%)
level 1 (90%)


occupation
0
0.2
0.4
0.6
0.8
1
0
100
200
300
400
500
time (a.u.)
target level 0
target level 1
level 0
level 1
laser field (a.u.)
-0.04
-0.02
0
0.02
0.04
0.06
0
100
200
300
400
500
time (a.u.)


1.0
0.8
0.6
0.4
0.2
0.0
occupation
0
100
200
300
400
500
time (a.u.)
target level 0
level 0 (α=0.05)
level 0 (α=0.5)


0.06
0.04
0.02
0
-0.02
-0.04
laser field (a.u.)
0
100
200
300
400
500
time (a.u.)
α=0.5
α=0.05

Control path in real space

$$\hat{O}(t) = \delta(r - r_0(t)) \approx \frac{1}{\sqrt{2\pi\sigma^2}} e^{-(r-r_0(t))^2/2\sigma^2}$$

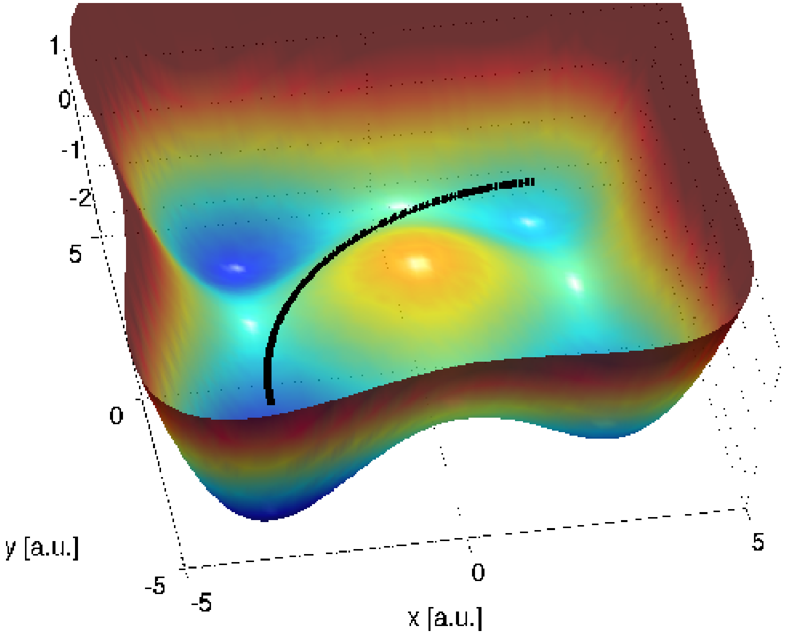
with <u>given</u> trajectory $r_0(t)$ **.**

Algorithm maximizes the density along the path $r_0(t)$:

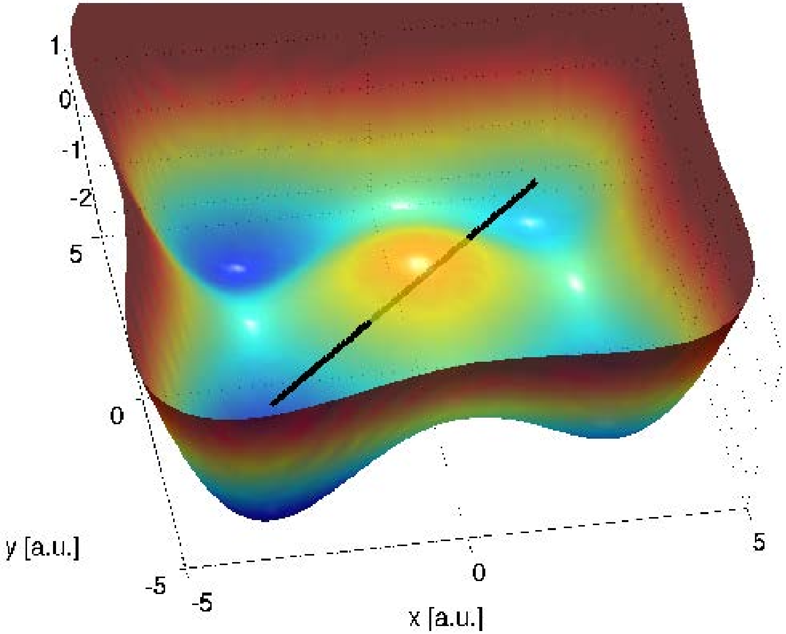
I. Serban, J. Werschnik, E.K.U.G. Phys. Rev. A 71, 053810 (2005)

Control of charge transfer along selected pathways

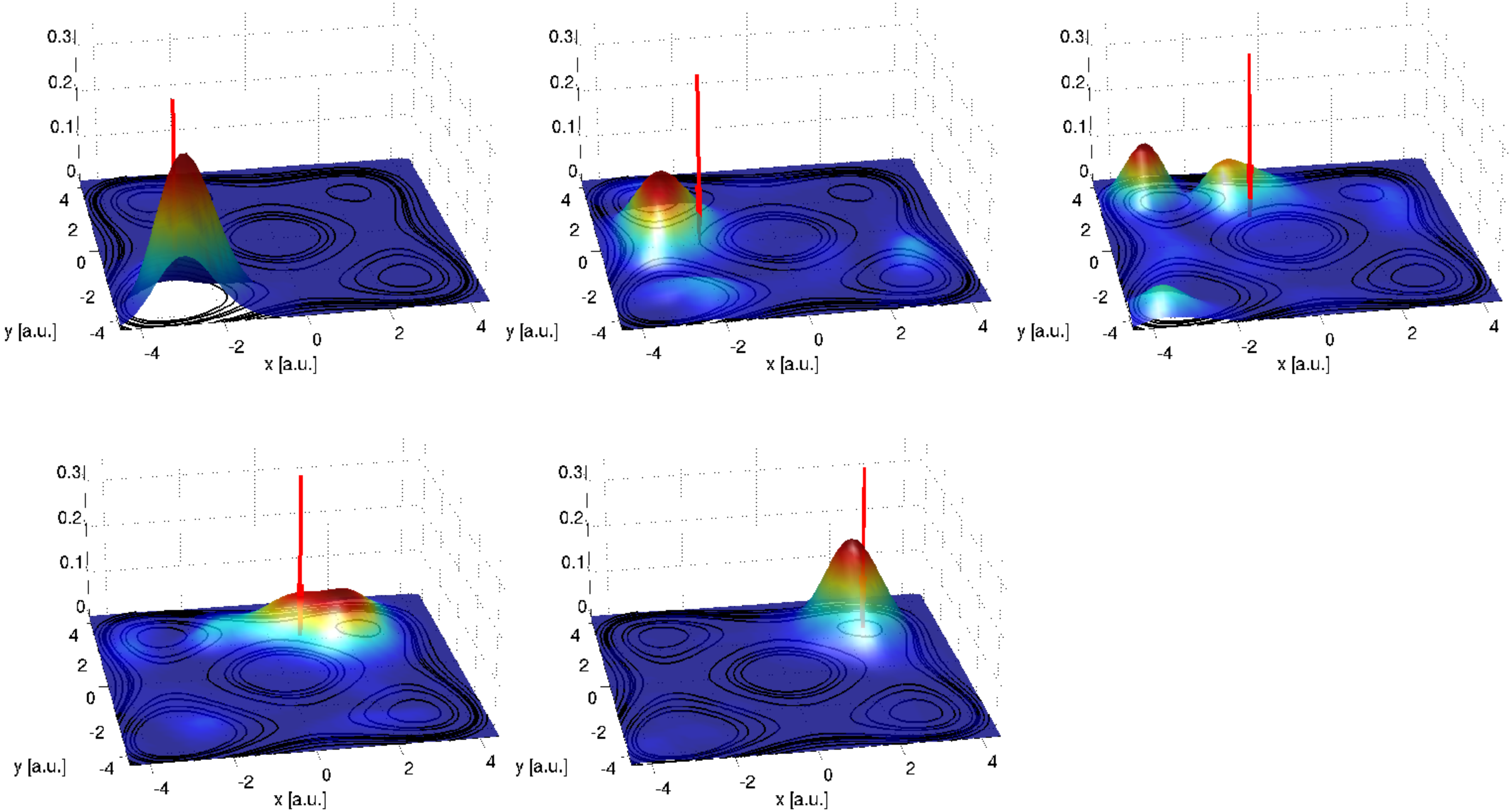
Trajectory 1



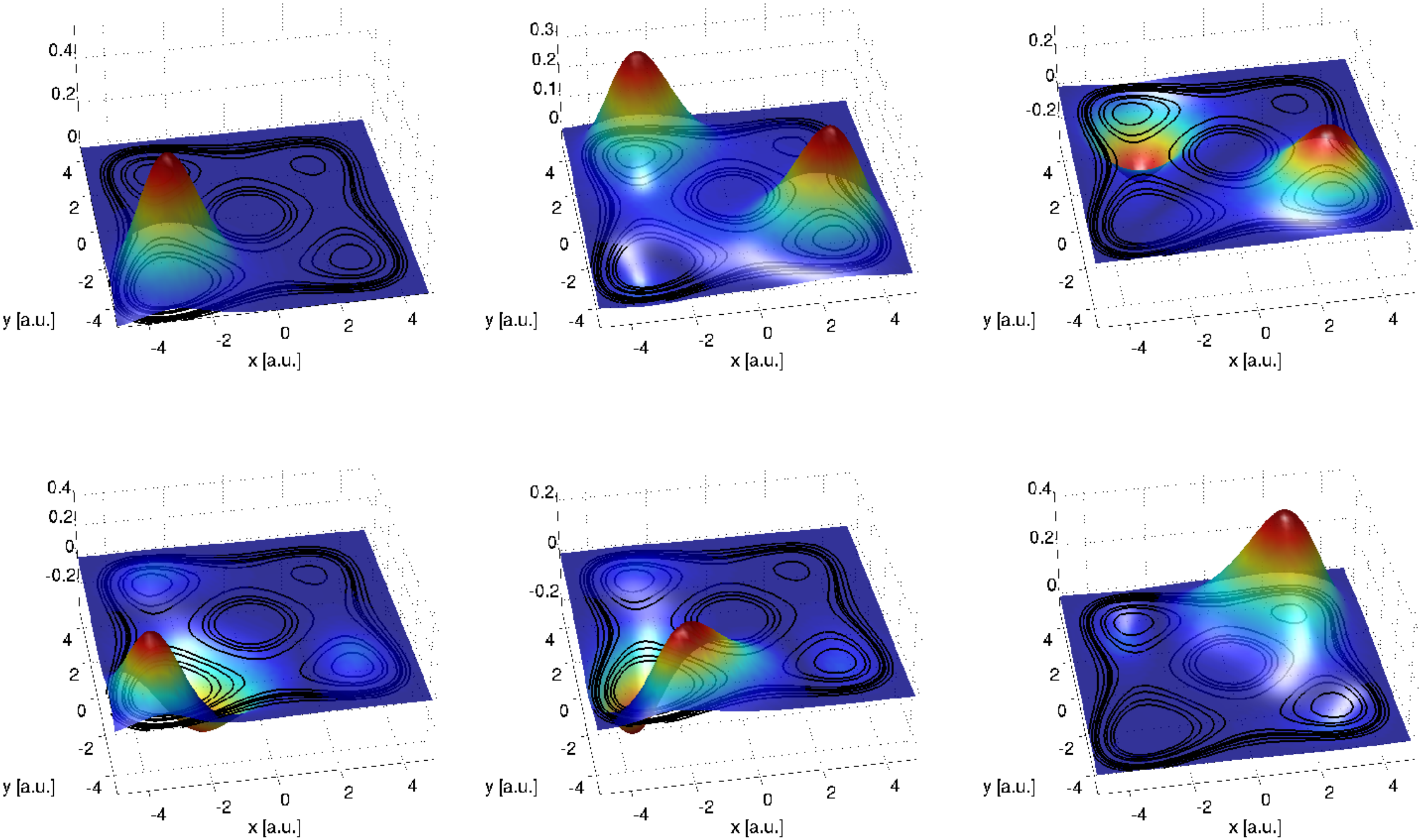
Trajectory 2



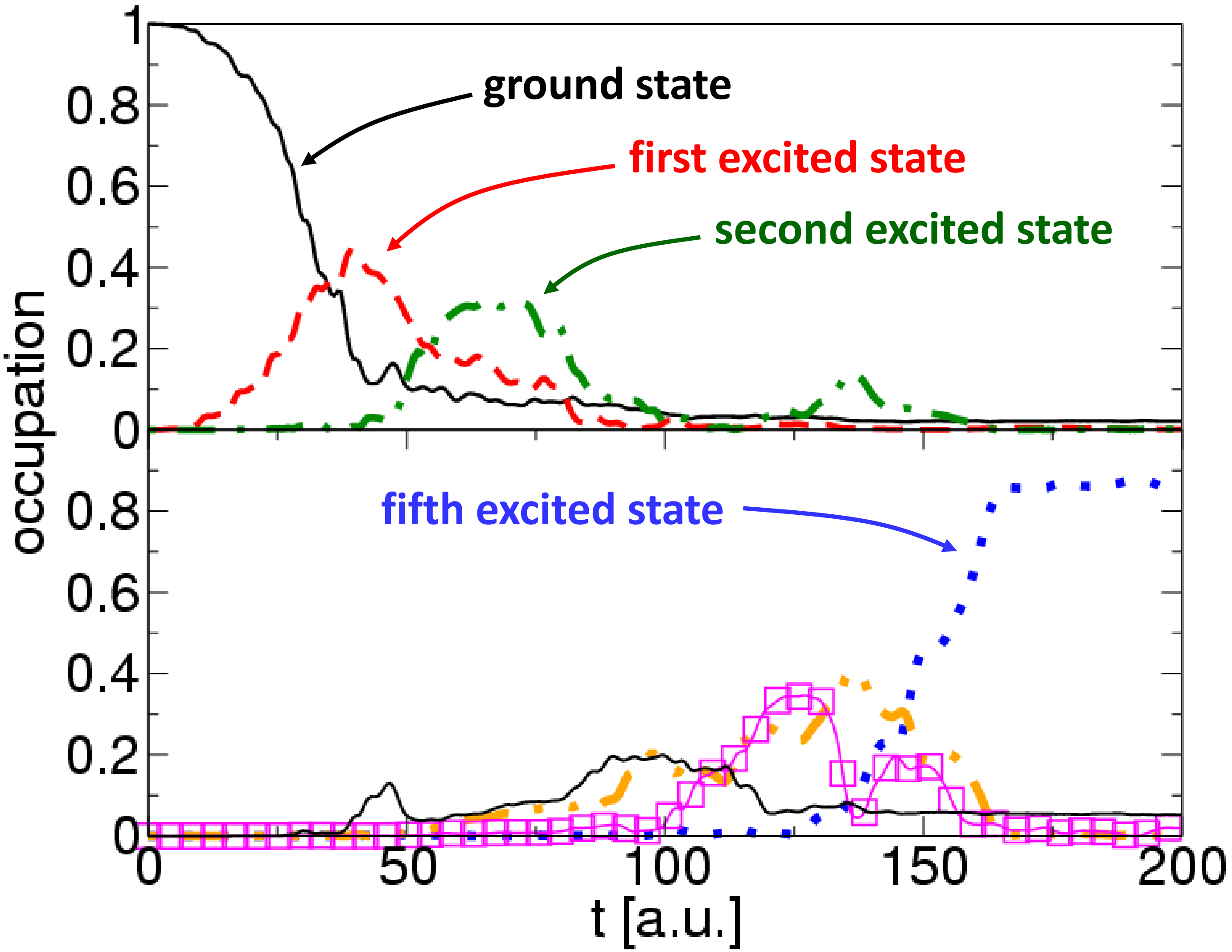
Time-evolution of wavepacket with the optimal laser pulse for trajectory 1



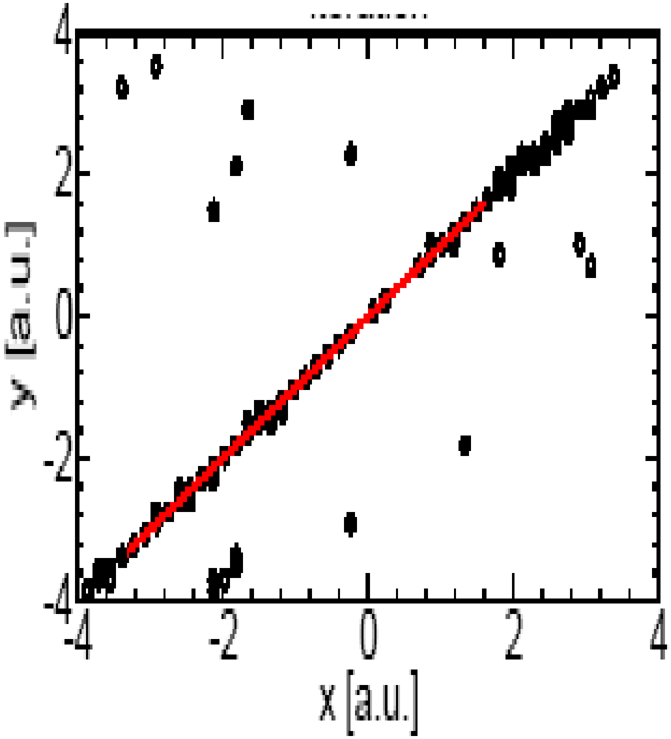
Lowest six eigenstates

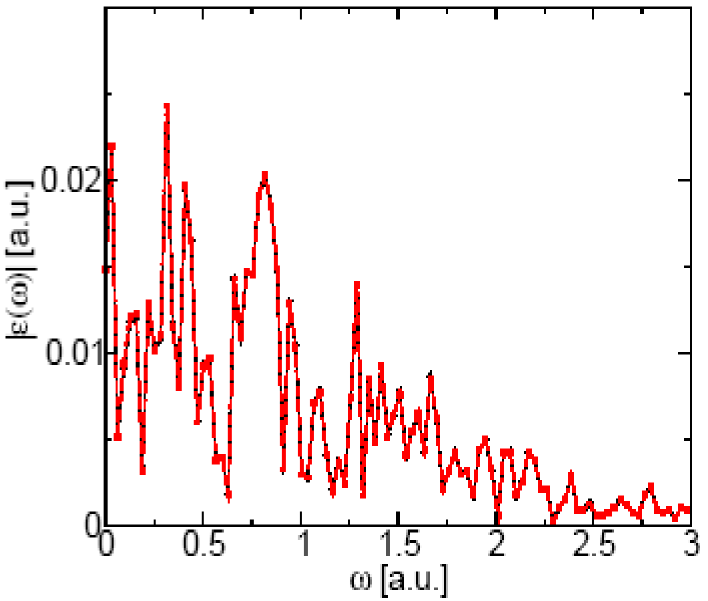


Populations of eigenstates



Trajectory 2





Control of many-body systems

- Formally the same OCT equations
- Problem: For 3 or more degrees of freedom, the full solution of the TDSE becomes computationally very hard

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A. Castro, J. Werschnik, E.K.U. Gross, PRL 109, 153603 (2012)

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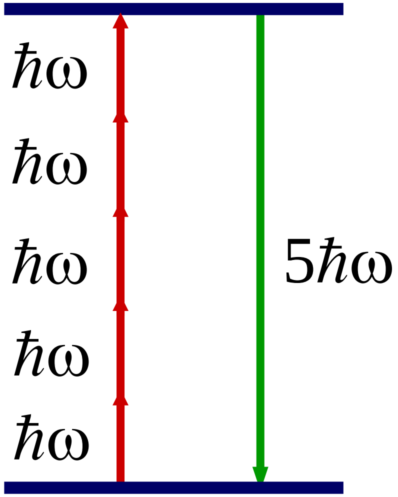
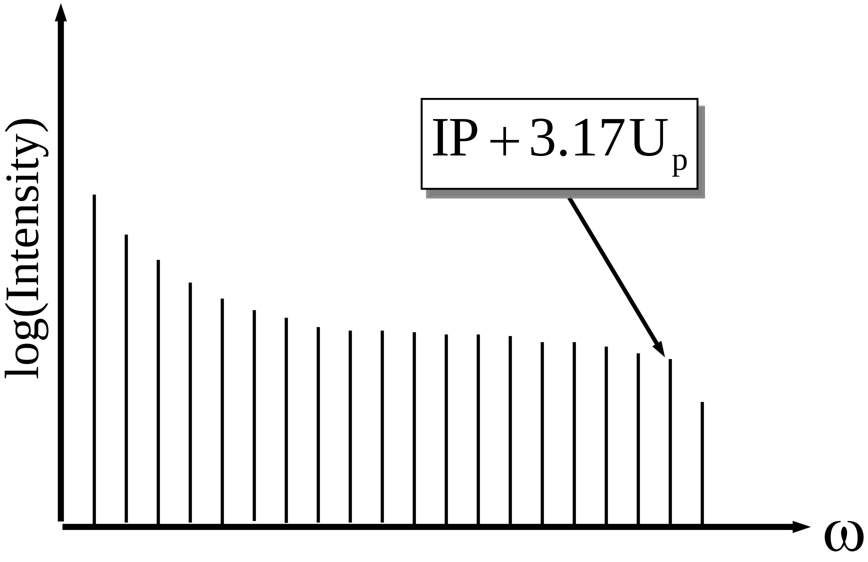
Important: Control target must be formulated in terms of the density!

Optimal Control of Harmonic Generation (example: Helium Atom)





Enhancement of a single harmonic peak

Harmonic Spectrum:

$$H(\omega) = \left| \int dt e^{i\omega t} \frac{d^2}{dt^t} \left\{ \int d^3 r z \rho(\vec{r}, t) \right\} \right|^2$$

Maximize:

$$F = \sum_k \alpha_k \max_{\omega \in [k\omega_0 - \beta, k\omega_0 + \beta]} H(\omega)$$

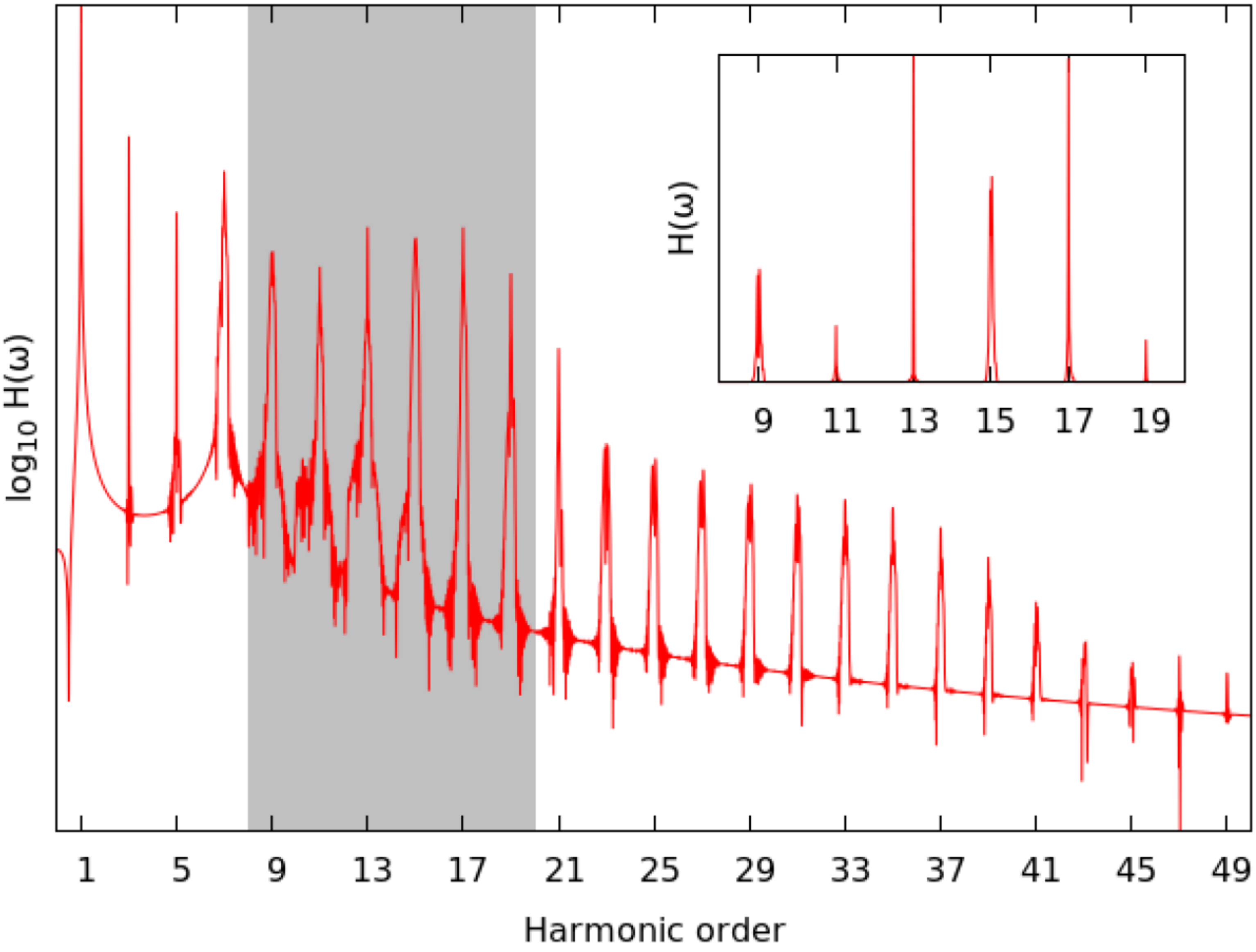
To maximize, e.g., the 7th harmonic of ω_0 , choose coefficients as
$\alpha_7 = 4$, $\alpha_3 = \alpha_5 = \alpha_9 = \alpha_{11} = -1$

<u>Measure of enhancement</u>: Compare with reference pulse:

$$\varepsilon_{\mathrm{ref}}(t) = \varepsilon_0 \cos\left(\frac{\pi}{2}\frac{2t - T}{T}\right)\cos(\omega t)$$

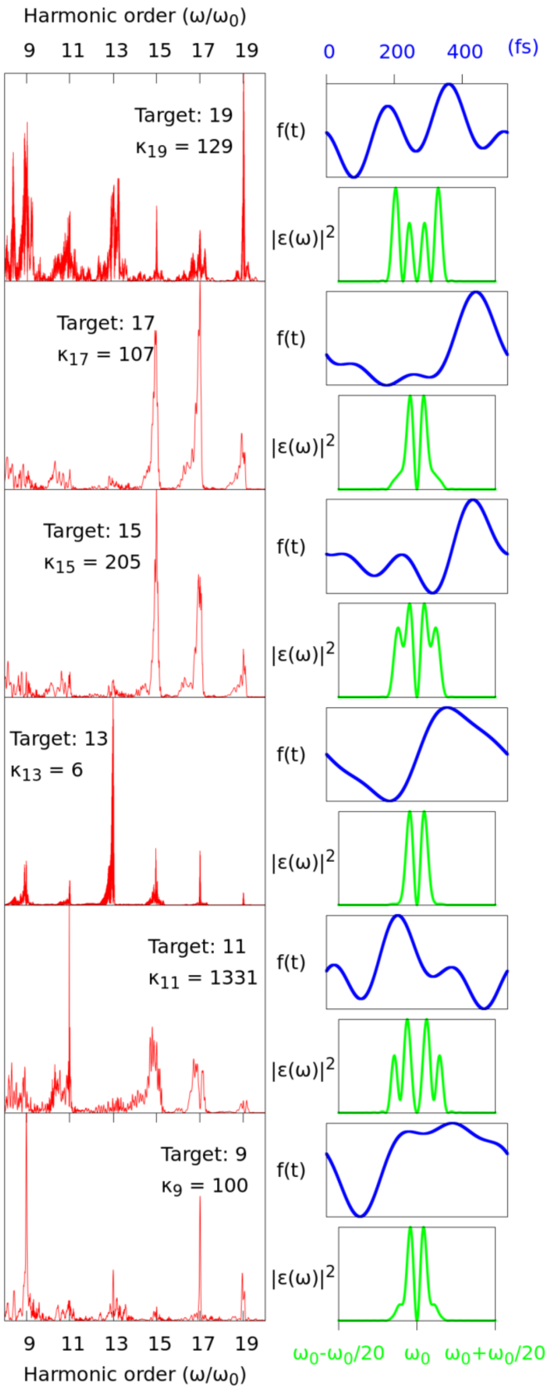
$$\kappa_j = \frac{\max_{\omega \in [j\omega_0 - \beta,\, j\omega_0 + \beta]} H(\omega)}{H_{\mathrm{ref}}(j\omega_0)}$$

Harmonic spectrum of reference pulse for hydrogen atom



Results for Hydrogen

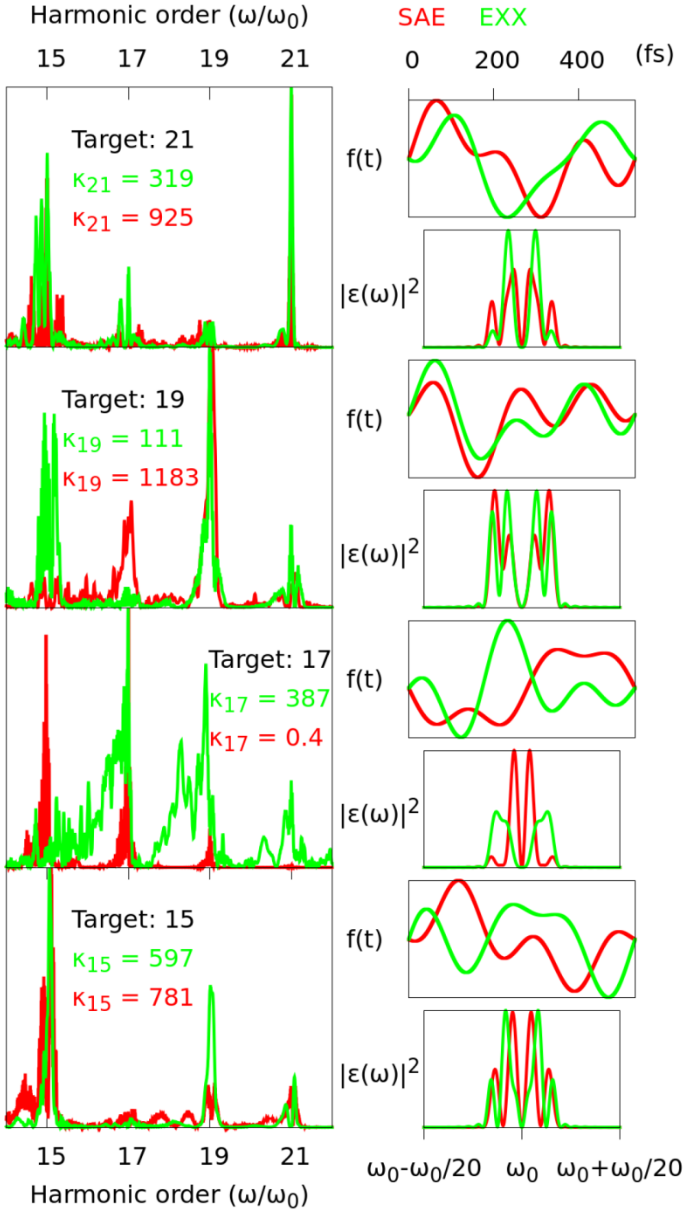
A. Castro, A. Rubio, E.K.U.Gross, Eur. Phys. J. B 88, 191 (2015).

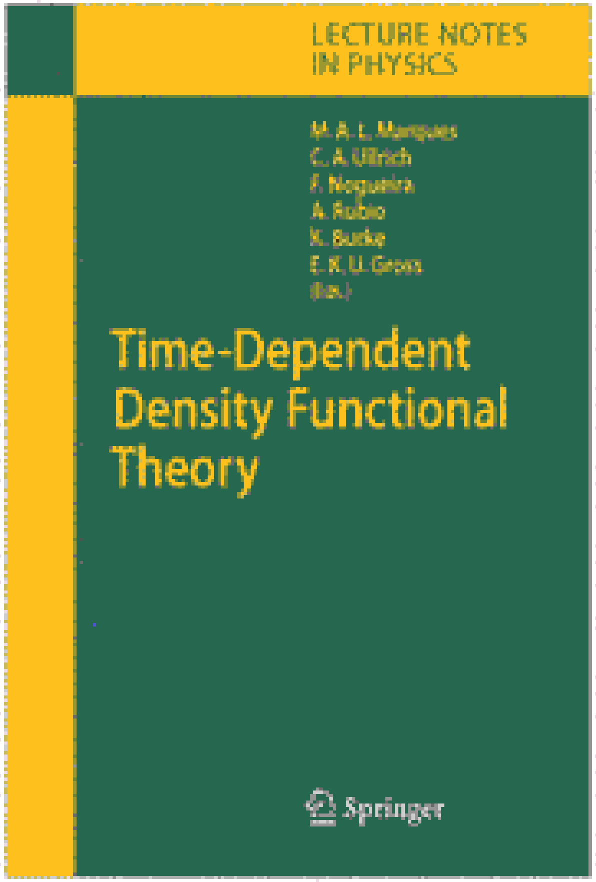


Results for Helium

(Using TDDFT with EXX functional)

A. Castro, A. Rubio, E.K.U.Gross,
Eur. Phys. J. B 88, 191 (2015).





LECTURE NOTES
IN PHYSICS

M. A. L. Marques
C. A. Ullrich
F. Nogueira
A. Rubio
K. Burke
E. K. U. Gross
(Eds.)

Time-Dependent
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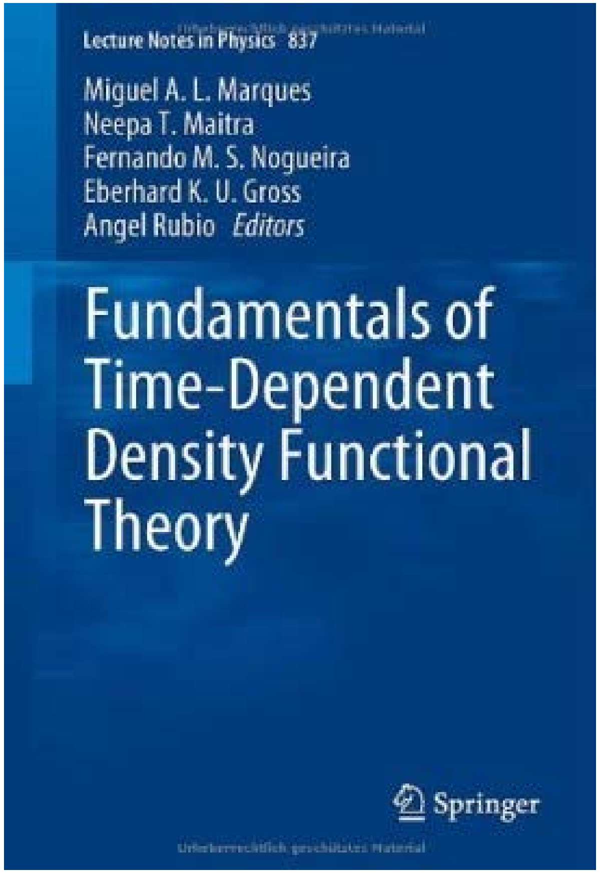
Theory

Springer

Lecture Notes in Physics 706
(Springer, 2006)



Lecture Notes in Physics 837

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Fundamentals of
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Thanks !