Coherent control of a short and intensive XUV laser pulse in the spherical symmetric box potential

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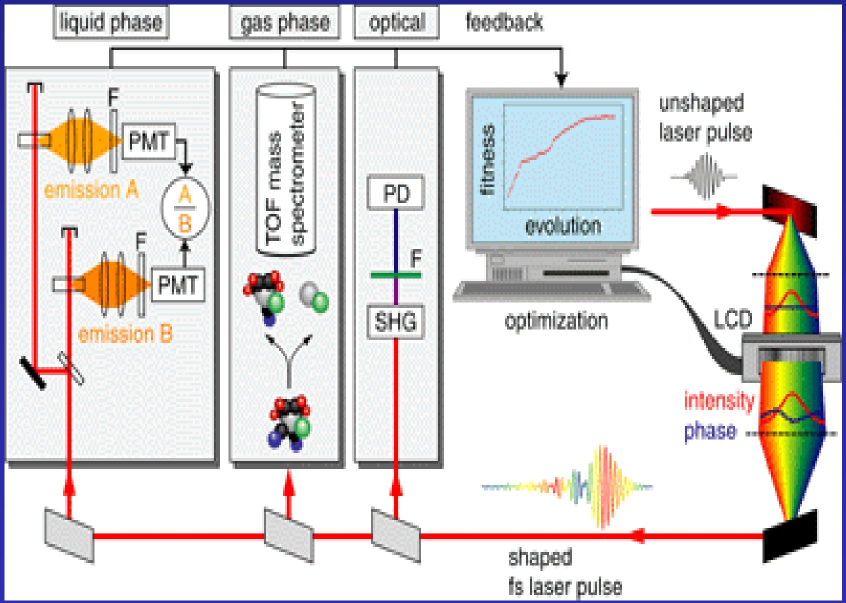
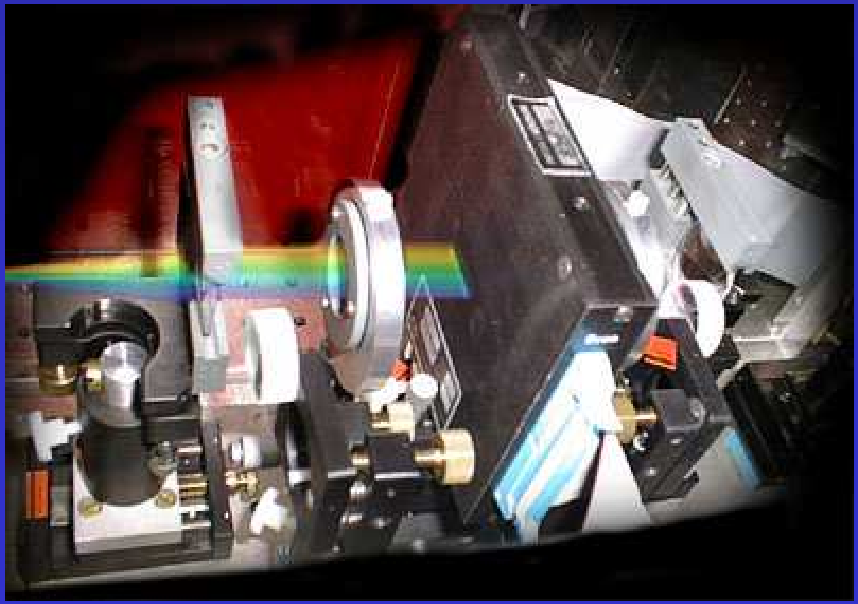
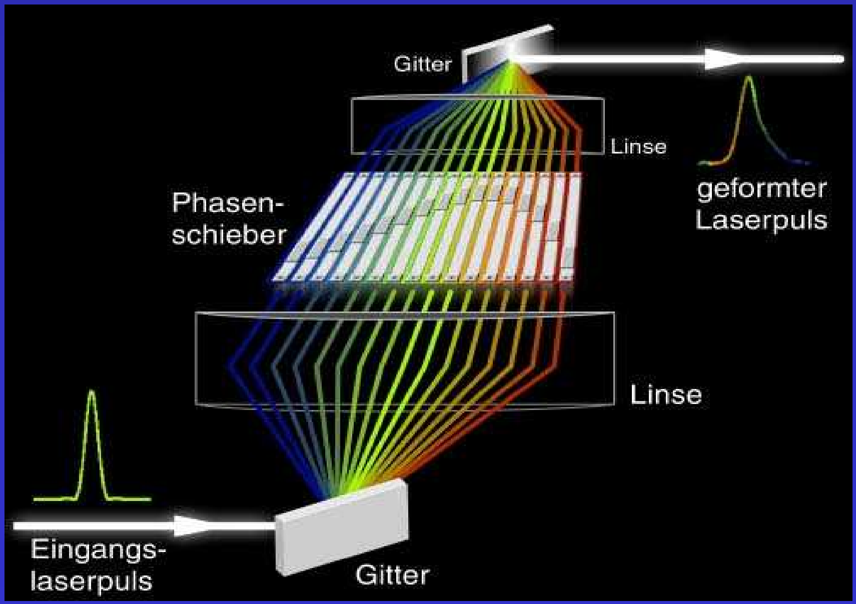
Motivation 1

Adaptive femtosecond quantum control can be applied for visible light

(The photos are from Gustav Gerber's Institute)
http://www.physik.uni-wuerzburg.de/femto-welt/







Motivation 2

it is experimentally possible to create XUV laser pulses with HHG or with Free Electron Laser

→ eg. two-photon double ionisation can be studied

→ electron-electron correlation can be investigated

OR/END coherent controlled laser pulses can be created

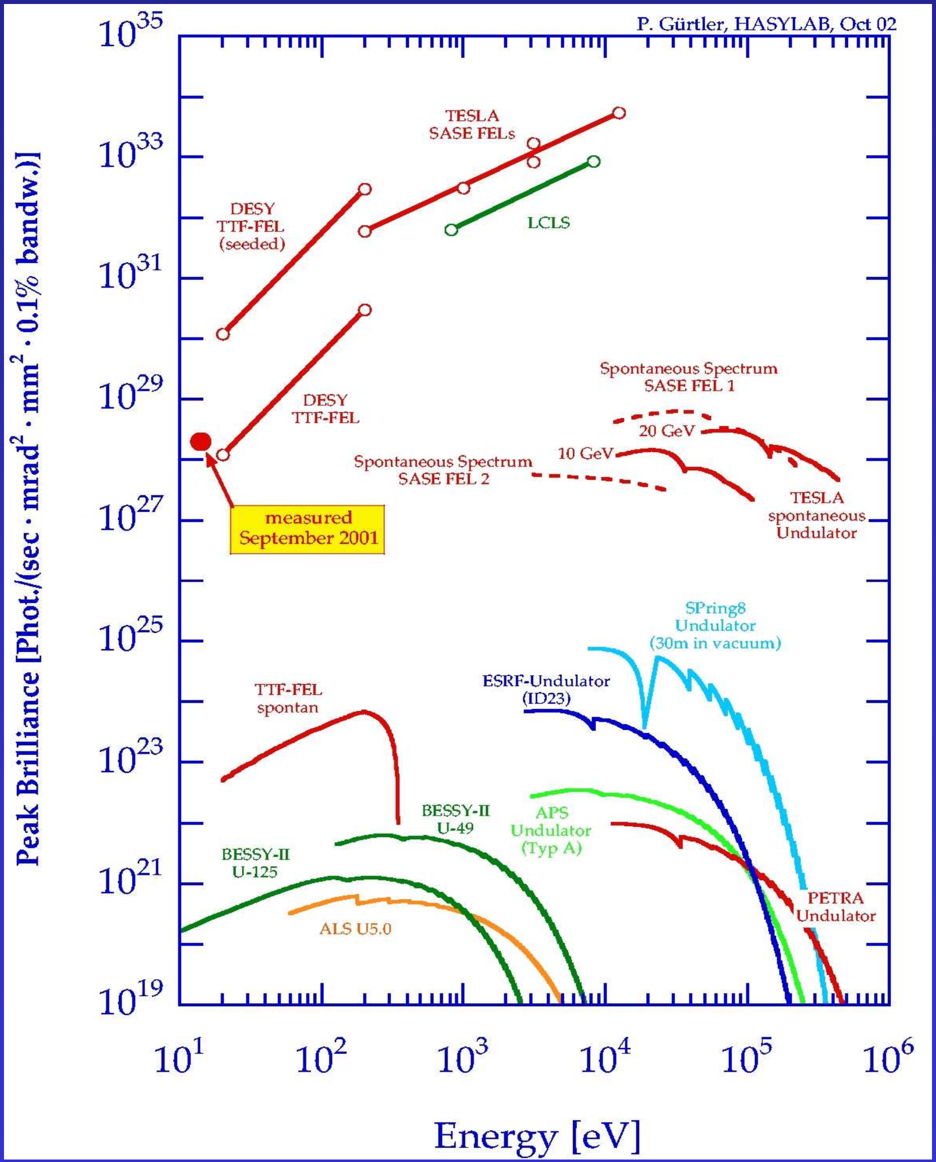


figure is from http://www.dasy.de

Our coupled-channel method

-- Our applied Coupled-Channel Method was originally developed for ion-He collision (details in I.F. Barna, Ionization of helium in relativistic heavy-ion collisions, Doctoral Thesis, University of Giessen (2002) „Giessener Elektronische Bibliothek“

http://geb.uni-giessen.de/geb/volltexte/2003/1036

I.F. Barna, N. Grün and W. Scheid, Eur. Phys. J. D 25, (2003) 239

I.F. Barna, K. Tőkési and J. Burgdörfer, J. Phys. B 38, (2005) 1

-- Later modified to study photoionization of He

I.F. Barna, J.M. Rost, Eur. Phys. J. D 27, (2003) 287

I.F. Barna, J. Wang and J. Burgdörfer, Phys. Rev. A 73, (2006) 023402

-- and coherent control

I.F. Barna
"Coherent control calculations for helium atom in short and intensive XUV laser pulses "
Eur. Phys. J. D 33, (2005) 307

The Hamiltonian of the system:

The time-dependent Schrödinger equation:

$$i\frac{\partial}{\partial t}\Psi(\vec{r}_1,\vec{r}_2,t) = (\hat{H}_{He} + \hat{V}(t))\Psi(\vec{r}_1,\vec{r}_2,t)$$

- $\hat{V}(t)$ laser-electron interaction
- $\Psi(\vec{r}_1,\vec{r}_2,t)$ CI. wavefunction of helium

The unperturbed helium Hamiltonian:

$$\hat{H}(\vec{r}_1,\vec{r}_2)_{He} = -\frac{\vec{\nabla}_1^2}{2} - \frac{\vec{\nabla}_2^2}{2} - \frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{|\vec{r}_1 - \vec{r}_2|}$$

- spin-spin, spin-orbit, and mass polarisation terms are neglected

Laser field

minimal coupling for the electromagnetic field to $\hat{H}_{He}$

$$\hat{H}(\vec{r}_1,\vec{r}_2,t) = \frac{(\vec{p}_1 - \vec{A}(\vec{r}_1,t)/c)^2}{2} + \frac{(\vec{p}_2 - \vec{A}(\vec{r}_2,t)/c)^2}{2}$$

$$-\frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{r_{12}} - \phi(\vec{r}_1,t) - \phi(\vec{r}_2,t) = \hat{H}_{He} + \hat{V}(t)$$

- dipole approximation: $\lambda_{laser} >> r_{atom}$
- velocity gauge: $\hat{V}(\vec{r},t) = \sum_{i=1,2} \vec{A}(t)\cdot\vec{p}_i$
- length gauge:
  $\hat{V}(\vec{r},t) = -\sum_{i=1,2} \vec{E}(t)\cdot\vec{r}_i, \qquad \vec{E}(t) = -\frac{1}{c}\frac{\partial}{\partial t}\vec{A}(t)$
- $\vec{E}(t) = E_0 \cdot F(t) \cdot \sin(\omega t) \cdot \vec{e}_z$ linearly polarised pulse
- envelope function: $F(t) = \sin^2\left(\frac{\pi t}{T}\right)$ T is the pulse duration
- or $F(t) = \exp\left(-\frac{(2\ln 2)t^2}{T^2}\right)$ T is the FWHM

The coupled-channel equation

Ansatz:

$$\Psi(\vec{r}_1, \vec{r}_2, t) = \sum_{j=1}^{N} a_j(t)\Phi_j(\vec{r}_1, \vec{r}_2)e^{-iE_j t}$$

Leads to a system of first-order-differential equations for the coefficients a_j :

$$\frac{da_k(t)}{dt} = -i\sum_{j=1}^{N} V_{kj}(t)e^{-i(E_j - E_k)t}a_j(t) \quad (k = 1 \ldots N)$$

$V_{kj}(t) = \langle\Phi_k|\hat{V}(t)|\Phi_j\rangle$ coupling matrix

Initial conditions:

$$a_k\,(t \to -\infty) = \begin{cases} 1 & k = 1 \\ 0 & k \neq 1 \end{cases}$$

the final probability for each channel:

$$P_k(b, t \to \infty) = |\, a_k(t \to \infty)\,|^2$$

The wavefunction

Configuration interaction(CI) expansion of $\Phi_j(\vec{r}_1, \vec{r}_2)$ in terms of two-particle basis functions f_μ

$$\Phi_j(\vec{r}_1, \vec{r}_2) = \sum_\mu b^j_\mu f_\mu(\vec{r}_1, \vec{r}_2).$$

where $f_\mu(\vec{r}_1, \vec{r}_2)$ are symmetric (S=0) products of

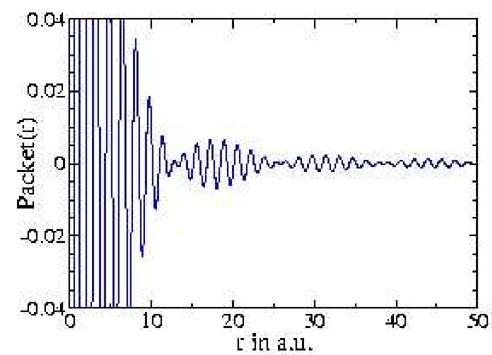
1. Slater-type orbitals:

$$\chi_{n,l,m,\kappa}(\vec{r}) = C(n, \kappa)r^{n-1}e^{-\kappa r}Y_{l,m}(\theta, \varphi)$$

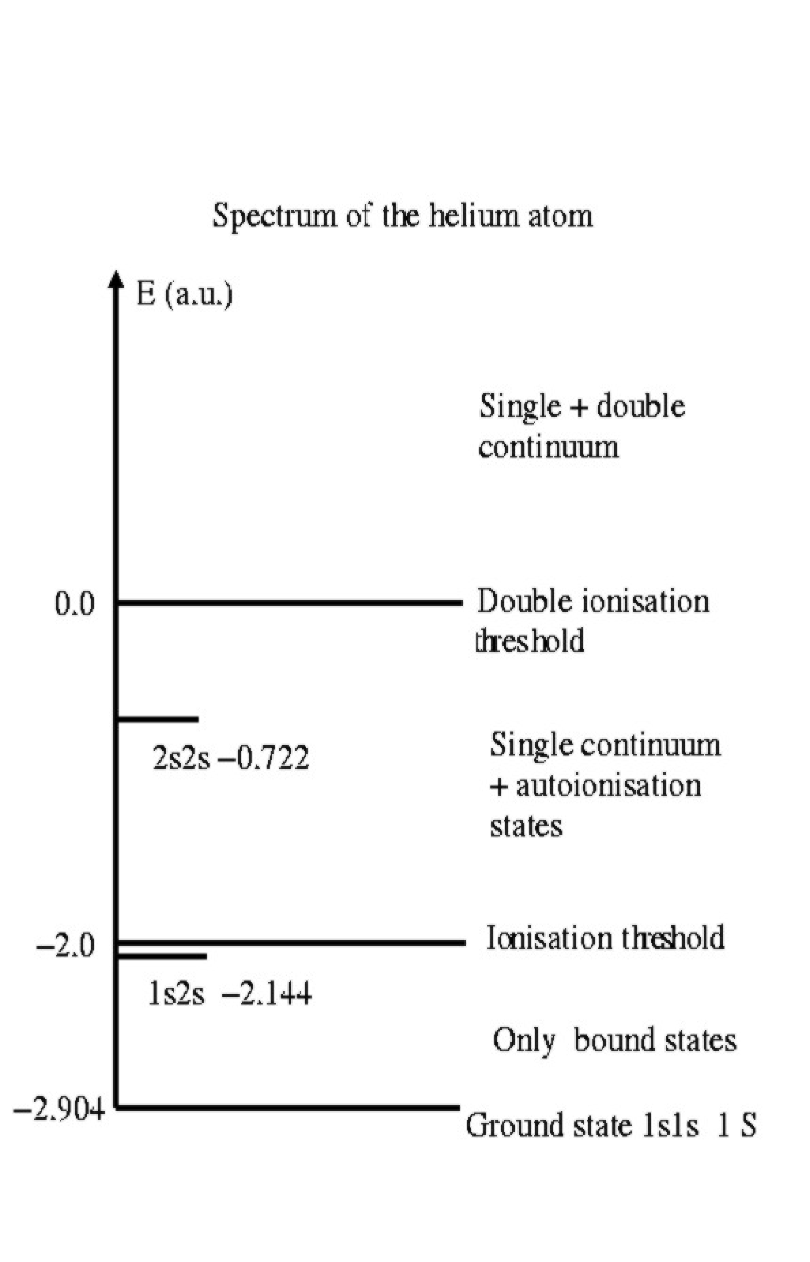
2. regular Coulomb wave packets:

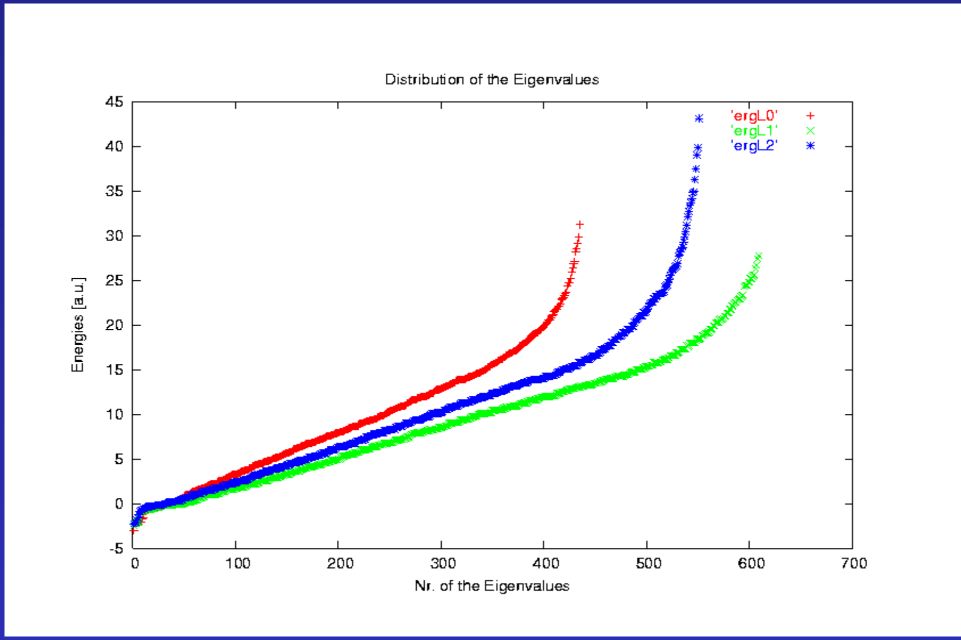
$$\varphi_{k,l,m,\tilde{Z}}(\vec{r}) = N(k, \Delta k)\int_k^{k+\Delta k} R_l(\eta, \rho)dk' Y_{l,m}(\theta, \varphi)$$

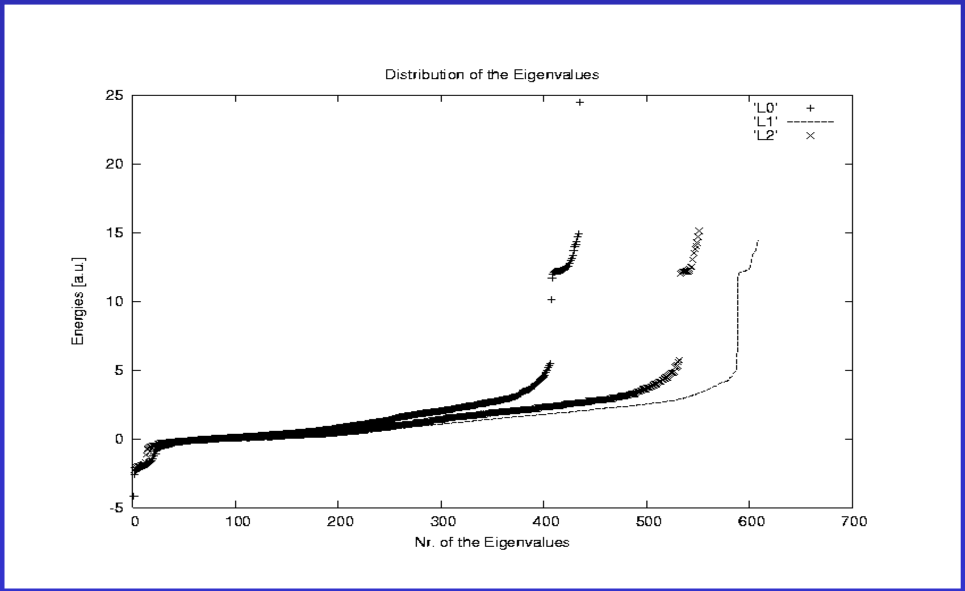
- $\eta = \tilde{Z}/k'$, $\rho = k'r$, $\tilde{Z}$ effective charge
- $N(k, \Delta k)$, $C(n, \kappa)$ normalisation constants



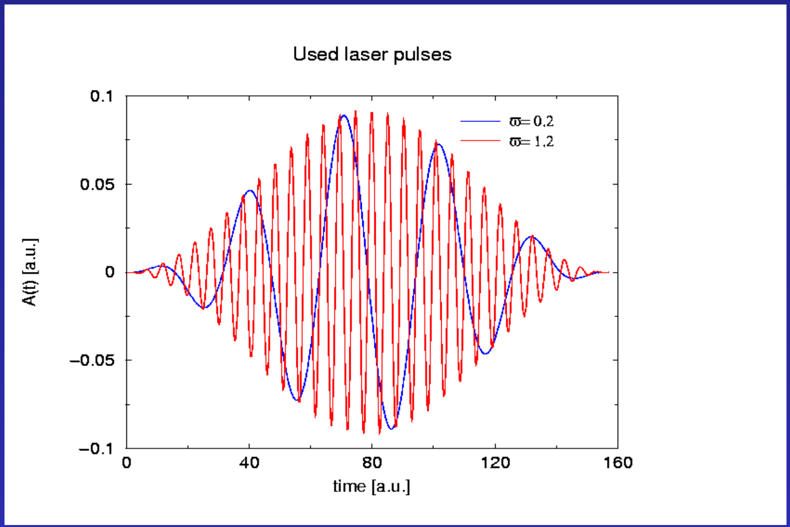
Spectrum & Continuum States



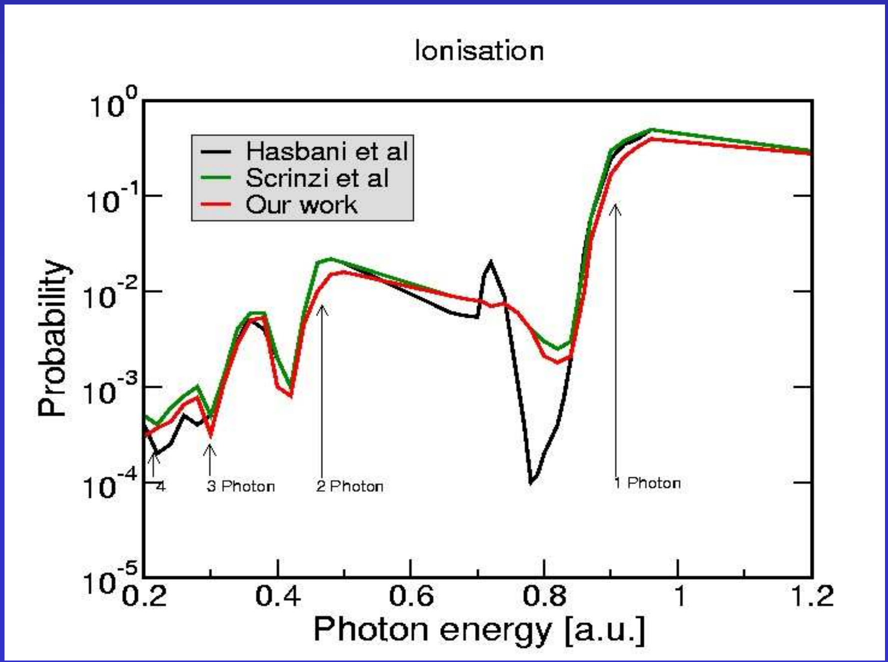


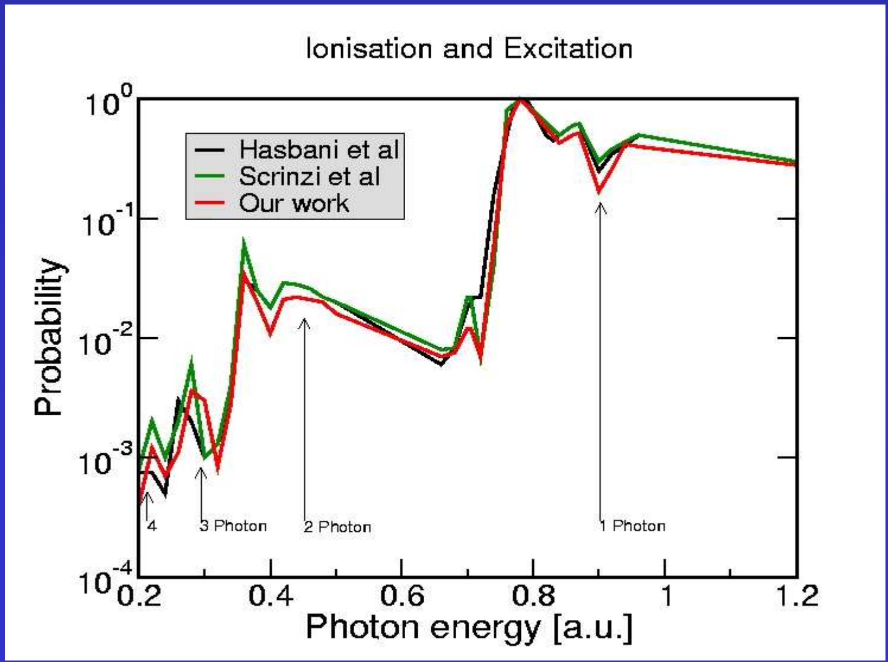


Laser pulse & Photoionization



A. Scrinzi *et al*. PRA **58**, 1310 (1998)
R. Hasbani *et al*. JPB **33**, 2101 (2000)
I. F. Barna *et al*. Eur. Phys. J. D **27,** 287 (2003)



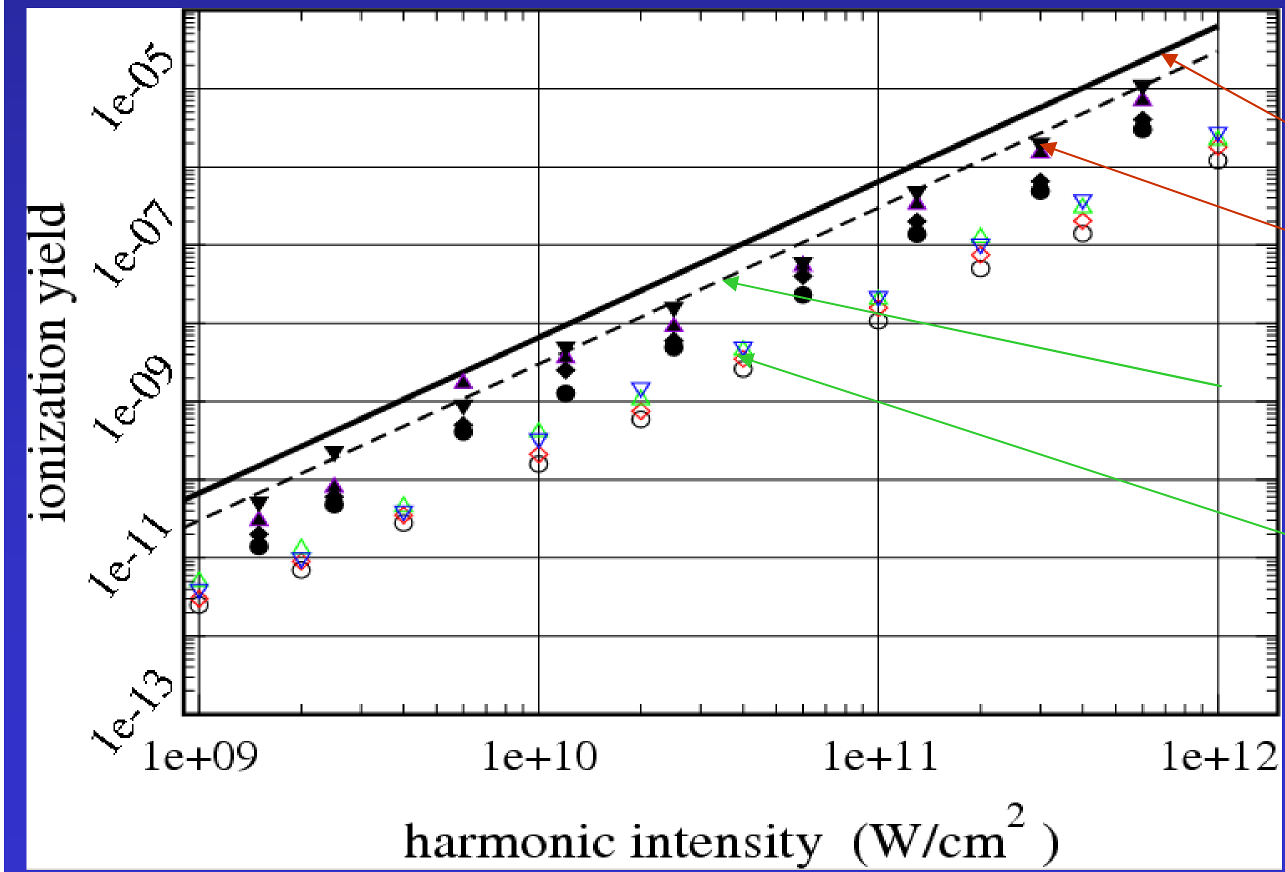


Two-Photon Ionization of He through a superposition of Higher Harmonics

Experiment & Theory (Coupled Channel method versus Spline Base)

N. A. Papadogiannis *et al.* ***PRL 90 133902-1 (2003)***

7^{th}, 9^{th}, 11^{th} and 13^{th} harmonics



36-49 fs pulse length
sin^2 envelope

our results

Papadogiannis

Genetic algorithm & Pulse Shaping

for coherent control for He atom

Genetic algorithm

nonlinear optimisation problem for N parameters

$\longrightarrow$ first construct K parameter sets randomly

$[a_1 \dots a_N]$, $[b_1 \dots b_N]$ ... $[k_1 \dots k_N]$

this is one generation of the parameters

$\longrightarrow$ transform all parameter sets into binary code

$[a_1 \dots a_N] \rightarrow [00010101_1 | \dots | 11010100_N]$

$\longrightarrow$ use genetic operations: *mutation, inversion, crossover*

01010101	01010101	10101111|10111000
↓	↓	↓
00110101	10101010	10101000|10111111

to change the parameters

$\longrightarrow$ calculates the fitness function which must be optimised (e.g. in our case: $1s1s \longrightarrow 1s2p$ transition probability)

$\longrightarrow$ check for the best parameter set

keep the best and skip the other '$\Rightarrow$ survival of the fittest'

$\longrightarrow$ generate new parameter sets and do it again!!

- method is successfully implemented in experiments
- global localisation procedure $\Rightarrow$ does not stick to a small local minimum (at least we hope)
- 30-60 generations are needed to achieve convergence
- not the best method for optimise large number of parameters

Phase modulation as pulse shaping

procedure:

1) Fourier transform the pulse: $E(\omega) = \int_{-\infty}^{+\infty} e^{i\omega t} E(t) dt$

2) multiplicate with a complex phase $\tilde{E}(\omega) = E(\omega) e^{ig(\omega)}$

3) inverse Fourier transform: $\tilde{E}(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} e^{-i\omega t} \tilde{E}(\omega) d\omega$

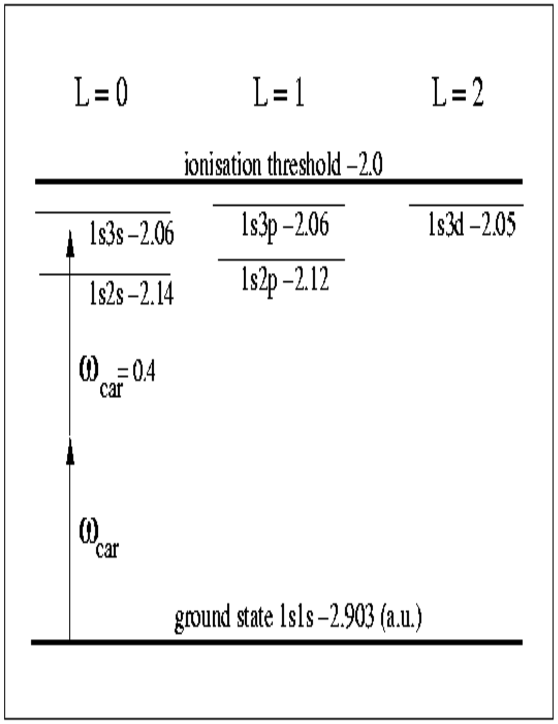
- where $g(\omega)$ is the phase function in practice it must be smooth, slow varying (e.g. polynomial)
- in our case $0 \leq g(\omega) \leq 2\pi$
  $\longrightarrow$ are the input parameters of the genetic algorithm
- Fast Fourier Transformation is applied in calculations

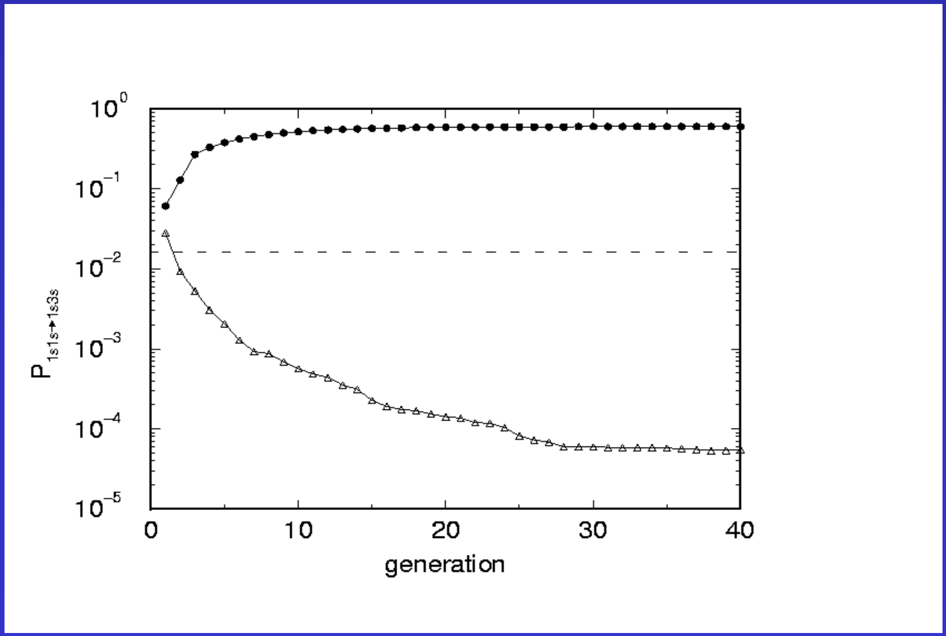
- each spectral component gets a new phase
- peak intensity and pulse duration are changed
- pulse energy remains the same,
  $\longrightarrow$ no extra (re)normalisation is needed
  $\int |f(t)|^2 dt = \int |\tilde{f}(t)|^2 dt$
- method is experimentally available for IR or visible laser pulses even with 128 different frequencies

Investigated system & results

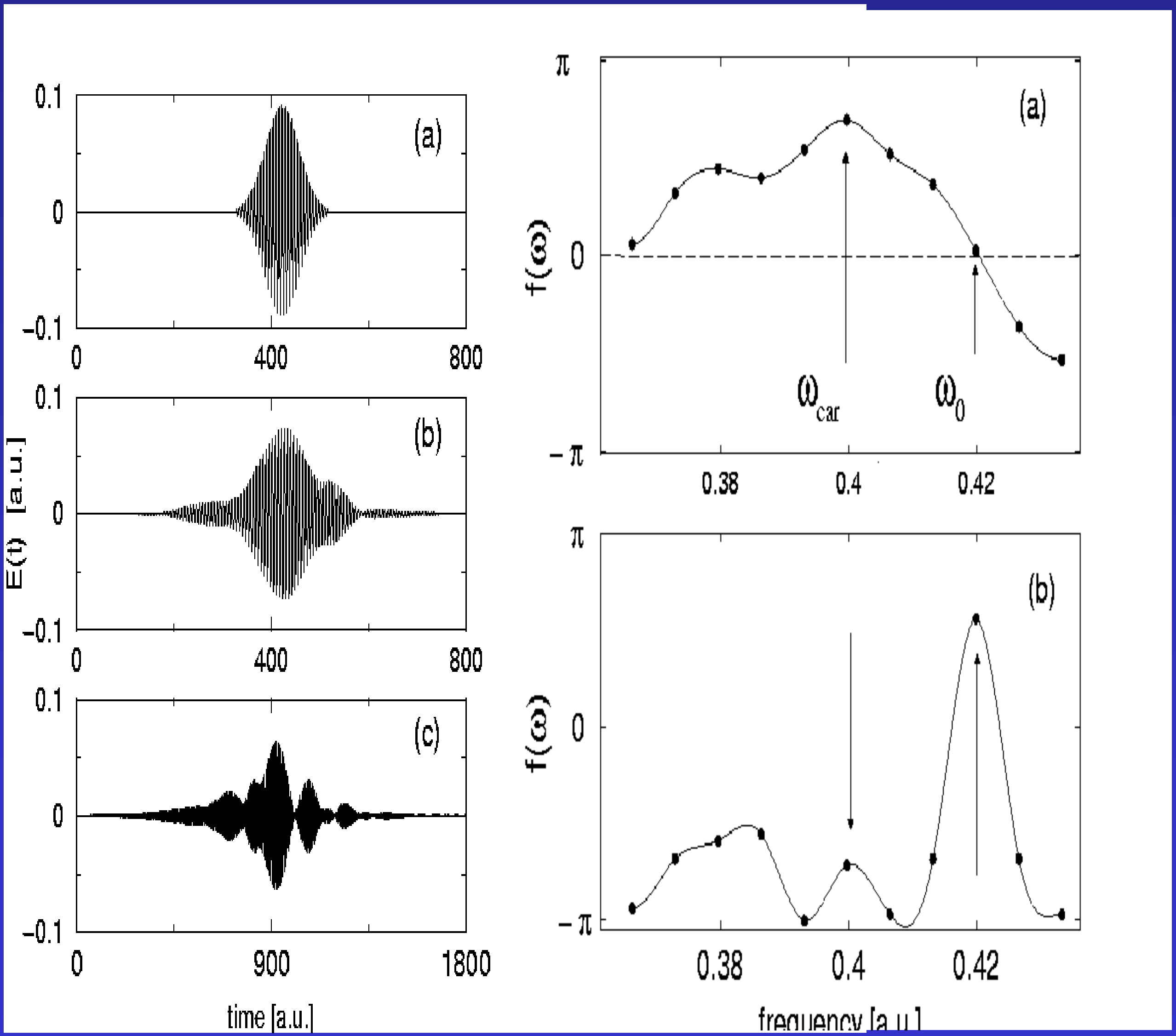
- the resonant two-photon excitation was investigated by D. Meshulach *et al*. Nature **396**, 239 (1998), PRA. 60, 1287, (1999)

- here the non-resonant two-photon excitation of the helium atom is examined
- at the non-perturbative intensity range, 10^14 W/cm^2, 40 channels are taken





Results



antisymmetric phase function
around the resonance frequency
for the best pulse

symmetric phase function
around the resonance frequency
for the worst (dark) pulse

Our model for the box potential

- three-dimensional, spherically symetric square-well box potential with 4 bound states, Energies, Eigenvalues are known

$$\Phi_\ell(\mathrm{r}) = \left(\frac{\pi}{2r}\right)^{\frac{1}{2}} \cdot J_{\ell+\frac{1}{2}}(rk) Y_{\ell,0}(\theta,\varphi).$$

- Linearly polarized pulse is taken, with 3 off-resonant carrier frequencies, pulse duration = 400 a.u.

$$\vec{E}(t) = E_n \cdot \sin^2\left(\frac{\pi t}{T}\right)\left[a_1 \sin(\omega_1 t + \delta_1) + a_2 \sin(\omega_2 t + \delta_2) + a_3 \sin(\omega_3 t + \delta_3)\right]\vec{e}_z.$$

- the full time-dependent quantum mechanical problem is solved

$$\Psi(\mathrm{r},t) = \sum_{\ell=0}^{3} a_\ell(t)\Phi_\ell(\mathrm{r})e^{-iE_\ell}$$

- the 3 phases and 3 pulse amplitudes are controlled with GE

Results

- the centre-of-mass of the time-dependent electron current is investigated

- best to worst pulse ratio is 82

$$\mathbf{j}(\mathbf{r},t) = \Psi(\mathbf{r},t)^* \vec{\nabla} \Psi(\mathbf{r},t)$$

$$r_{cm}(t) = \frac{\int j(r,t) \cdot r dr}{\int j(r,t) dr}$$

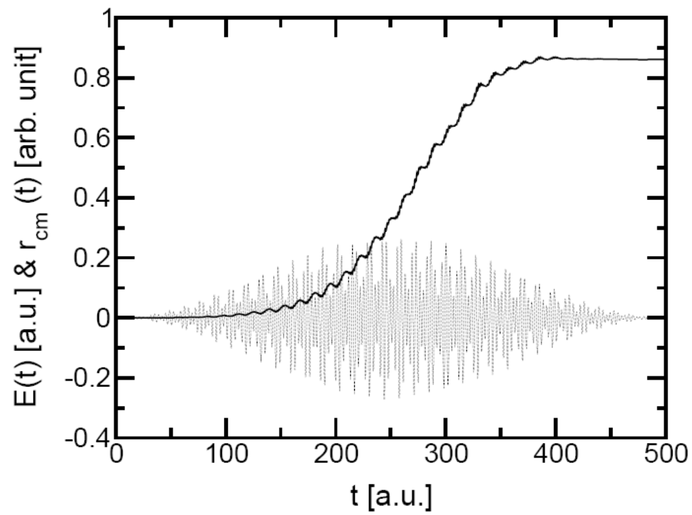


FIG. 1: The electric field strength E(t) of the optimized pulse (dashed line) with the corresponding center of mass of the electron current r_{cm}(t) solid line)

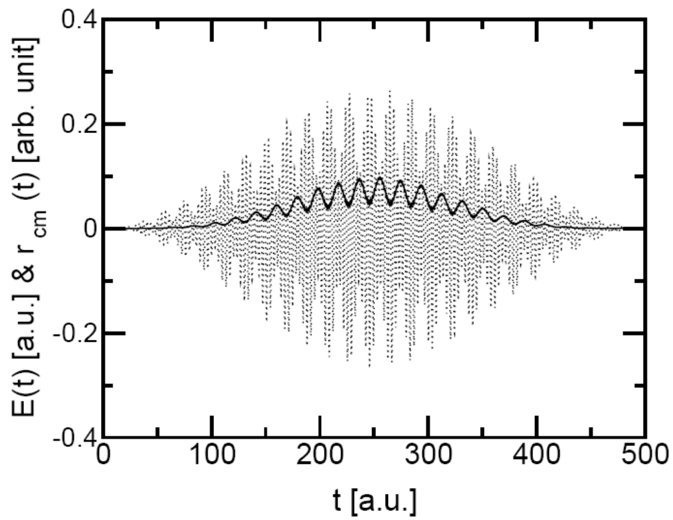


FIG. 3: The electric field strength E(t) of the neutralized pulse (dashed line) with the corresponding center of mass of the electron current r_{cm}(t) solid line)

Summary

- We presented our couppled channel method for He which is capable to describe one to four-photon ionization and coherent control
- As a new result we presented coherent control calculations for non-resonanat two photon excitation in a spherical box-potential