Numerical methods
in
Strong field dynamics II

Correlated Multielectron Dynamics in Intense Light Fields
PhD School of the INT-network CORINF – 11 - 15 June 2012

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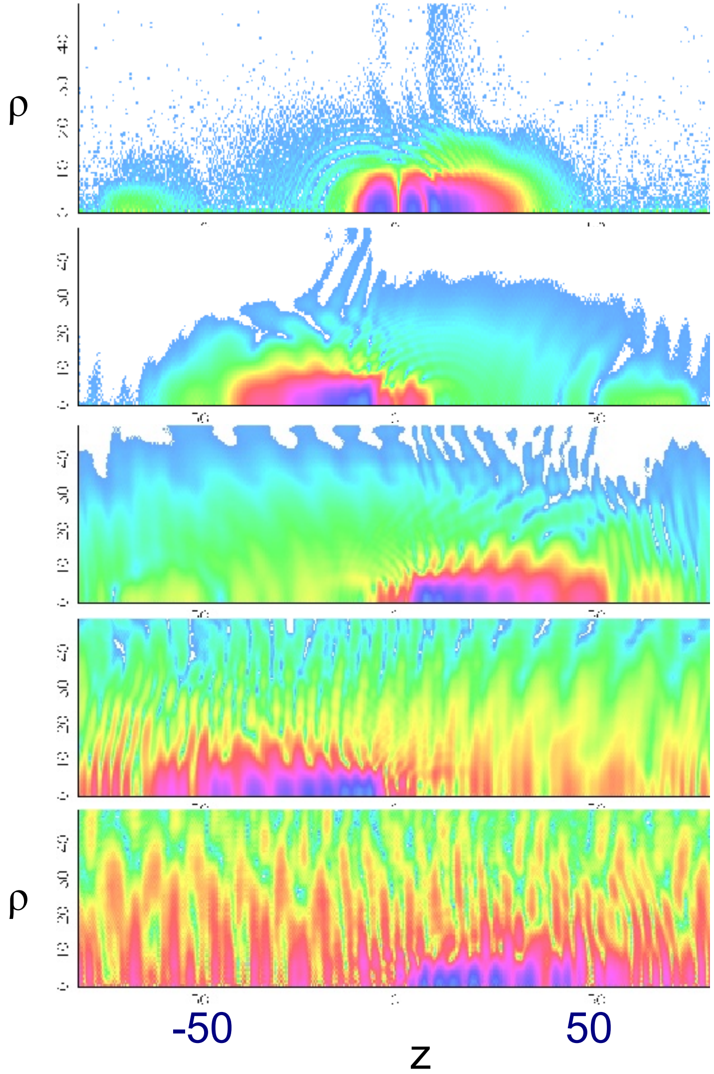
Strong pulse hits an atom

Electron density of the Hydrogen atom
In a 2-cycle pulse @ 2 x 10^{14} W/cm^2, 800nm



Complex appearance – simple basic physics

- tunnel (or so) ionization,
- free electron in the laser field,
- dipole matrix elements

Free motion is exactly described by the
Volkov solutions
of the time-dependent Schrödinger equation

Ionization can be (reasonably well) described
by rate formulae

Dipole matrix elements are known
from electronic structure calculations

We believe we understand the physics.

Why struggle to solve the TDSE?

Why solve the TDSE?
(Except for producing colorful pictures)

Test models:
We think we have understood – have we?

Numerical experiments:
Try, instead of thinking too hard
can be simple and inspiring

Predict:
Use models for trustworthy predictions of phenomena (not numbers)
Here we are doing physics at its best
High harmonic cutoff-energy ~ $I_p + 3.2\ U_p$
was first seen in simulation!

Get the numbers right, get the details:
ionization rates, photo-electron spectra, high harmonics
In the end, numbers is all that matters

Reproduce experimental findings:
The ultimate (but only ultimate) prove of correct experiment&computation
(very hard, if taken seriously!)

The solution – t-SURFF

The time-dependent SURFace Flux method

Idea:

We know the time-evolution beyond radius R_c ("channel radius"), e.g.Volkov solutions
Integrate flux through a surface for obtaining the asymptotic solutions

Related to:

R-matrix theory (in spirit, not technically)
Spectra from auto-correlation (for time-independent Hamiltonians)

Requires:

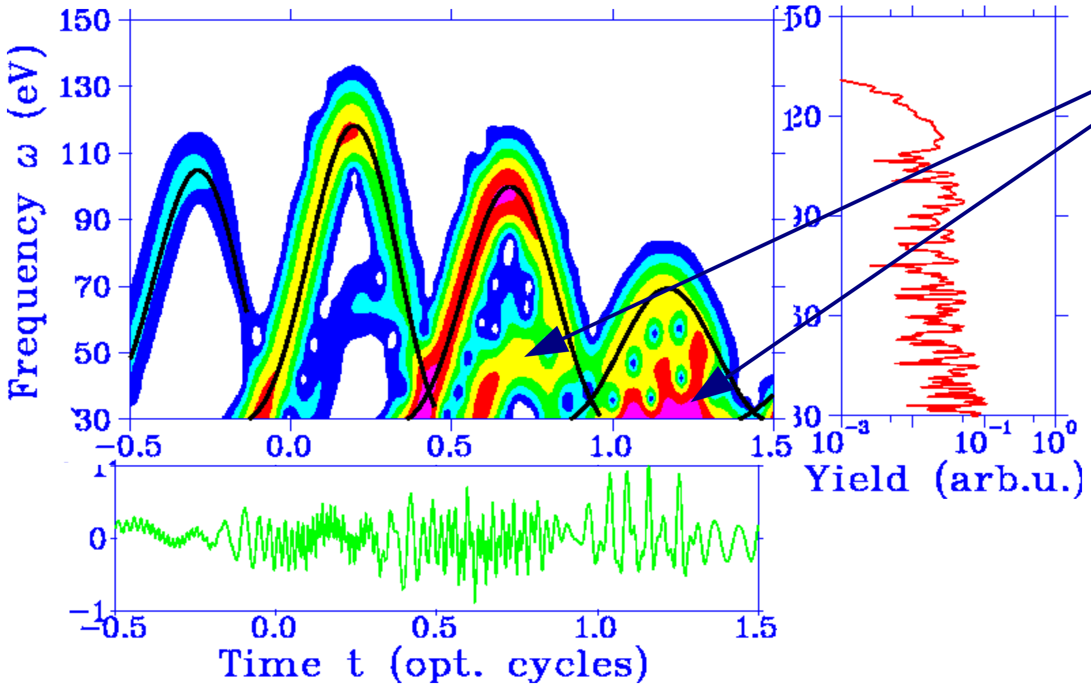
- wave function and derivative on $|\mathbf{r}|=R_c$: $\Psi|_{Rc}$, $\partial_r\Psi|_{Rc}$
- asymptotic solution (exact or numerically "cheap")
- perfect (or very good) absorption beyond R_c

Can do:

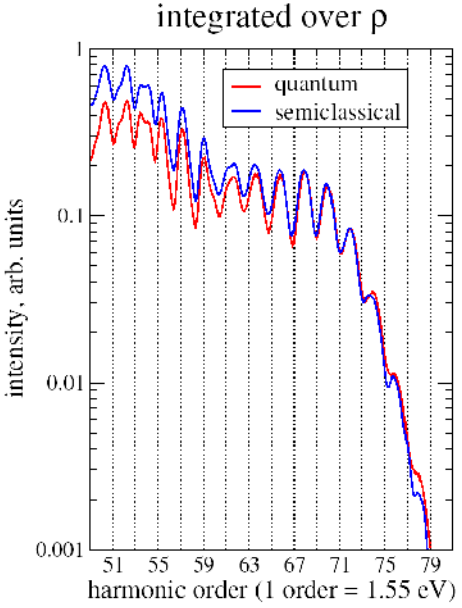
- fully differential strong-field IR single photo-electron spectra any polarization
- shake-up processes in two-electron systems
- fully differential strong-field IR double photo-electron spectra

We have understood: HHG

Time-frequency analysis of the high harmonic response

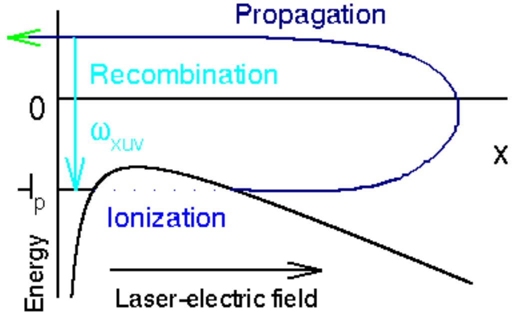


More structures
beyond naïve recollision



Colors: complete numerical solution

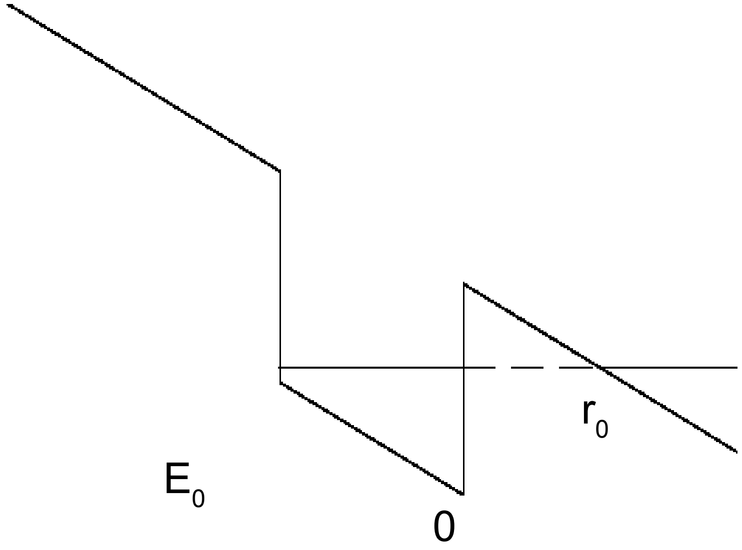
Solid lines: classical recollision energies



Tunnel ionization - principle

Gamov factor

$$\Gamma_{\text{tunnel}} \sim \exp\left[-2^{3/2}\int_0^{r_0}\sqrt{V(r)-E_0 dr}\right]$$



$$\int_0^{r_0}(-E_0-\vec{r}\cdot\vec{\mathcal{E}}_0)^{1/2} = -\frac{2}{3}\frac{(-E_0)^{3/2}}{\mathcal{E}_0}$$

$$\Gamma_{\text{tunnel}} \propto \exp\left[-\frac{2}{3}\frac{(-2E_0)^{3/2}}{\mathcal{E}_0}\right]$$

Strongly non-linear (exponential !) dependence on parameters

Ammosov-Delone-Krainov (ADK) formulae

Get the numbers right: field-ionization rates of He

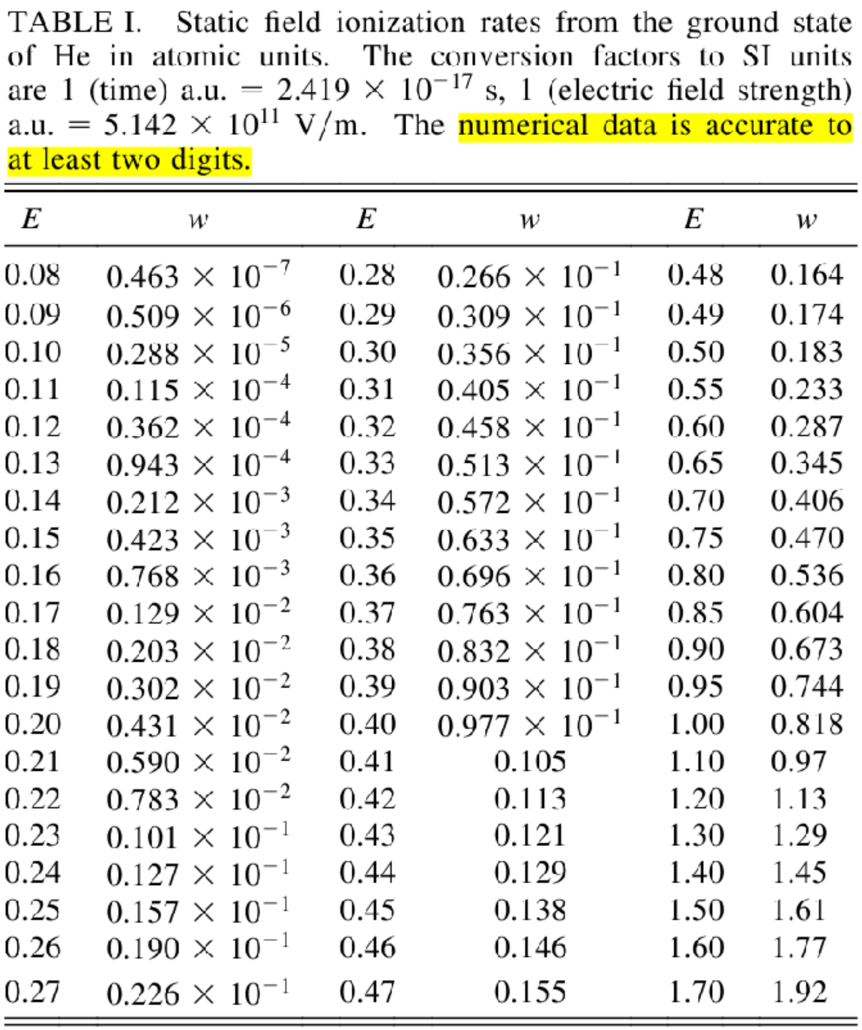
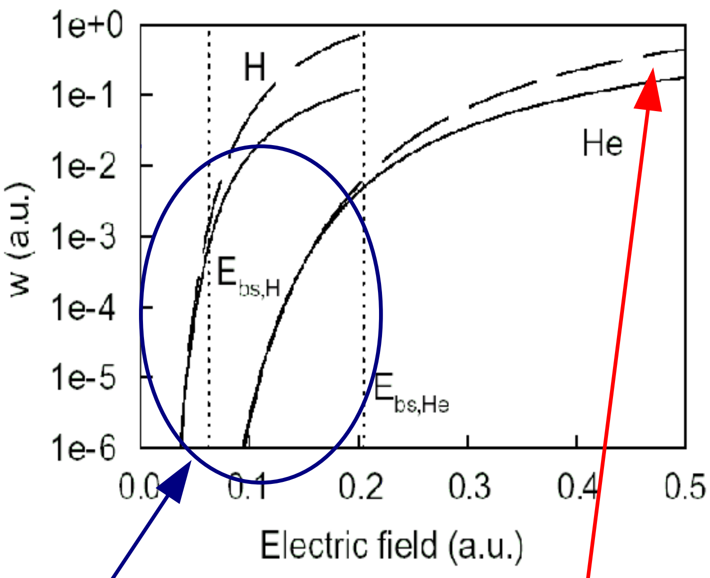


TABLE I. Static field ionization rates from the ground state of He in atomic units. The conversion factors to SI units are 1 (time) a.u. = 2.419 × 10^{-17} s, 1 (electric field strength) a.u. = 5.142 × 10^{11} V/m. The numerical data is accurate to at least two digits.

E	w	E	w	E	w
0.08	0.463×10^{-7}	0.28	0.266×10^{-1}	0.48	0.164
0.09	0.509×10^{-6}	0.29	0.309×10^{-1}	0.49	0.174
0.10	0.288×10^{-5}	0.30	0.356×10^{-1}	0.50	0.183
0.11	0.115×10^{-4}	0.31	0.405×10^{-1}	0.55	0.233
0.12	0.362×10^{-4}	0.32	0.458×10^{-1}	0.60	0.287
0.13	0.943×10^{-4}	0.33	0.513×10^{-1}	0.65	0.345
0.14	0.212×10^{-3}	0.34	0.572×10^{-1}	0.70	0.406
0.15	0.423×10^{-3}	0.35	0.633×10^{-1}	0.75	0.470
0.16	0.768×10^{-3}	0.36	0.696×10^{-1}	0.80	0.536
0.17	0.129×10^{-2}	0.37	0.763×10^{-1}	0.85	0.604
0.18	0.203×10^{-2}	0.38	0.832×10^{-1}	0.90	0.673
0.19	0.302×10^{-2}	0.39	0.903×10^{-1}	0.95	0.744
0.20	0.431×10^{-2}	0.40	0.977×10^{-1}	1.00	0.818
0.21	0.590×10^{-2}	0.41	0.105	1.10	0.97
0.22	0.783×10^{-2}	0.42	0.113	1.20	1.13
0.23	0.101×10^{-1}	0.43	0.121	1.30	1.29
0.24	0.127×10^{-1}	0.44	0.129	1.40	1.45
0.25	0.157×10^{-1}	0.45	0.138	1.50	1.61
0.26	0.190×10^{-1}	0.46	0.146	1.60	1.77
0.27	0.226×10^{-1}	0.47	0.155	1.70	1.92

Comparison of
accurate numerical data to
ADK ionization rates

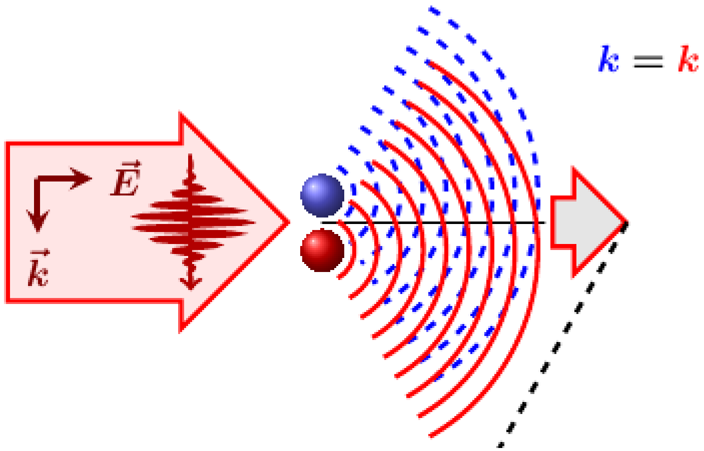


ADK works
nicely here

Factor 6 discrepancy!

We have not (fully) understood: ionization

Laser ionization of H_2



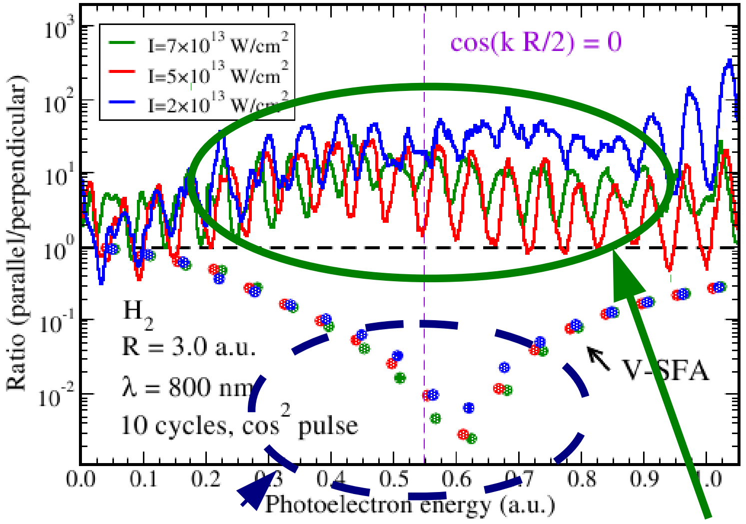
Electrons coherently leave
from the two atoms of H_2

When atomic separation matches
½ of the phase difference

Destructive interference, no ionization?

Predicted by molecular ADK formula

Solve the TDSE for H_2



Truth
(well, of TDSE)

Molecular ADK prediction

Thanks to A. Saenz
[Y.V.Vanne, A. Saenz PRA 82, 11404(R), (2010)]

Numerical experiments

Two-photon double ionization of He

Basic idea:

Two photons
well separated in time ("long" pulse)
Ionic ground state after 1st ionization

2 well separated photo-electron energies

Two photons
at the same time (short pulse)
Both from the correlated neutral state

Both in the same energy range

Just where exactly would we expect the Effect and how strongly?

Very large scale simulation of TDSE

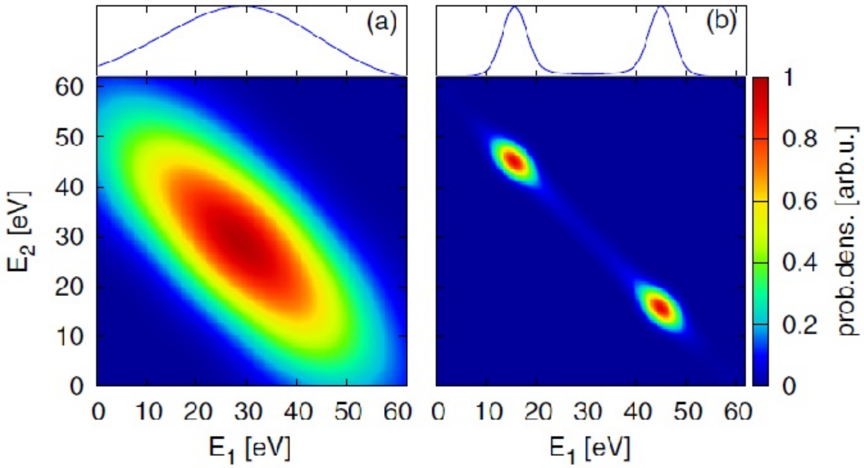


FIG. 1 (color online). TPDI electron spectra $P(E_1, E_2)$ at $\hbar\omega = 70$ eV for different pulse durations (FWHM): (a) $T_p = 150$ as, (b) $T_p = 750$ as. The top shows the spectrum integrated over one energy, i.e., the one-electron energy spectrum $P(E_1) = P(E_2)$.

[J. Feist et al., PRL 63002 (2009)]

Understand experiments in detail...

Photo-electron emission and spectra
- basis for all re-scattering type experiments

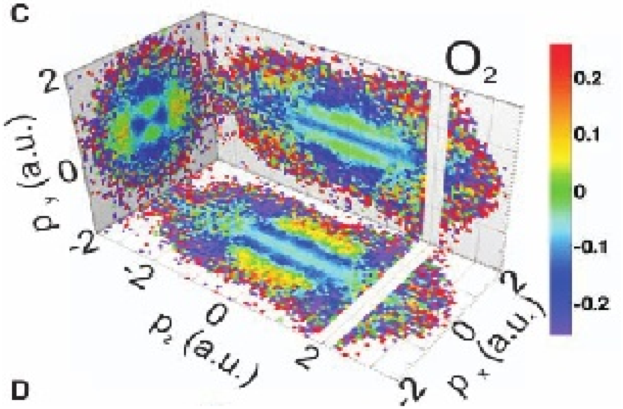
- High harmonic generation
- IR photo-electron spectra
- “Re-scattering imaging”

Surprisingly little is known

- Hydrogen
- Helium (to some degree)
- Single electron models

IR photo-electron spectra (COLTRIMS)

[Meckel et al., Science 2008]



Phantastic detail – difficult interpretation!

Experimentalists rely on ADK and its derivatives
Known to be quite wrong where we can check it:
e.g. Hydrogen atom and molecule at 800 nm, [shown by Alejandro Saenz]

Things to come...

- Approaches to solving the TDSE
- Simplifying models: 1d and restricted dynamics
- The 2-electron problem
- The few-electron problem and beyond
- Absorption of outgoing flux & spectra

The TDSE

potential

kinetic

field-interaction

Single electron, length gauge

$$i\frac{d}{dt}\Psi_L(\vec{r},t) = \left[-\frac{1}{2}\Delta - \frac{1}{r} - \vec{r}\cdot\vec{\mathcal{E}}(t)\right]\Psi_L(\vec{r},t)$$

Multi-electron

$$H(t) = \sum_i -\frac{1}{2}\Delta_i + \frac{Z}{r_i} + \vec{\mathcal{E}}\cdot\vec{r}_i + \sum_{i>j}\frac{1}{|\vec{r}_i - \vec{r}_j|}$$

Gauge-transformation

$$\Psi_L(\vec{r},t) = e^{+i\vec{A}\cdot\vec{r}}\Psi_V(\vec{r},t) \qquad \vec{A}(t) = \int_t^\infty \vec{\mathcal{E}}(t')dt'$$

~ laser vector potential

Velocity gauge

$$i\frac{d}{dt}\Psi_V(\vec{r}) = \left[\frac{1}{2}\left(-i\vec{\nabla} + \vec{A}(t)\right)^2 - \frac{1}{r}\right]\Psi_V(\vec{r})$$

field-interaction

Linear partial differential equation (parabolic)

Dimensions 1 (time) + 3^n (space), n... number of particles

Simple time-dependence

Approaches to solutions

Simplified analytic models – the Volkov propagator

Classical models – classical trajectory Monte-Carlo

Single-electron models

Models in reduced dimensions

Full fledged quantum dynamics calculations

Classical trajectory methods

Classical trajectory methods

Motion of free electron in the field is nearly classial
→ simulate by classical trajectories

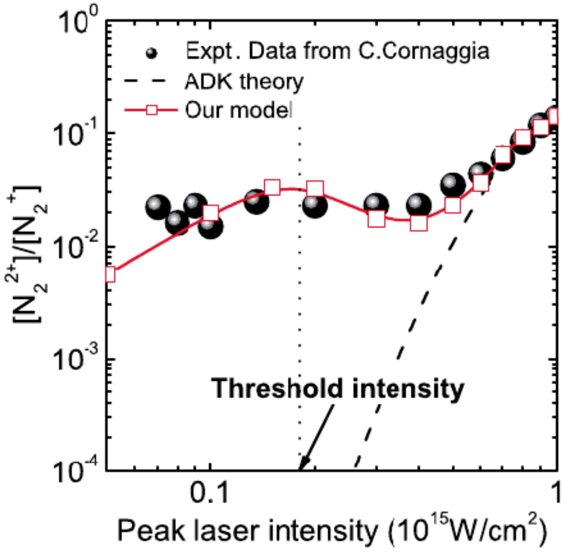
Advantages:
- Can handle 2-e situation relatively easily
- Follow the trajectories – “see what happens”

Disadvantages:
- “Auto-ionization” of the ground state
- Crucial choice of initial distributions in phase space
- Difficult to assess correctness

Reproduce
- Double-differential photo-electron emission patterns
- Double-to-single ratios at 800nm

Geyer, Rost, J. Phys. B 36, L107 (2003)
Ye, Chen, Liu, PRA 77, 013403 (2008)
Shaaran, Nygren, Faria, PRA 81, 063413 (2010)

N_2 Single-to-double ionization ratio



Ye et al. 2008
Exp. : Cornaggia 2000

Numerical solutions

Numerical solutions

What we need to do

In mathematical terms:

Solve the initial value problem
for a (parabolic) partial differential equation

Discretization

approximately represent Ψ by a finite set of numbers c_i

Turns the partial differential equation
into a set of ordinary differential equations

Time-propagation

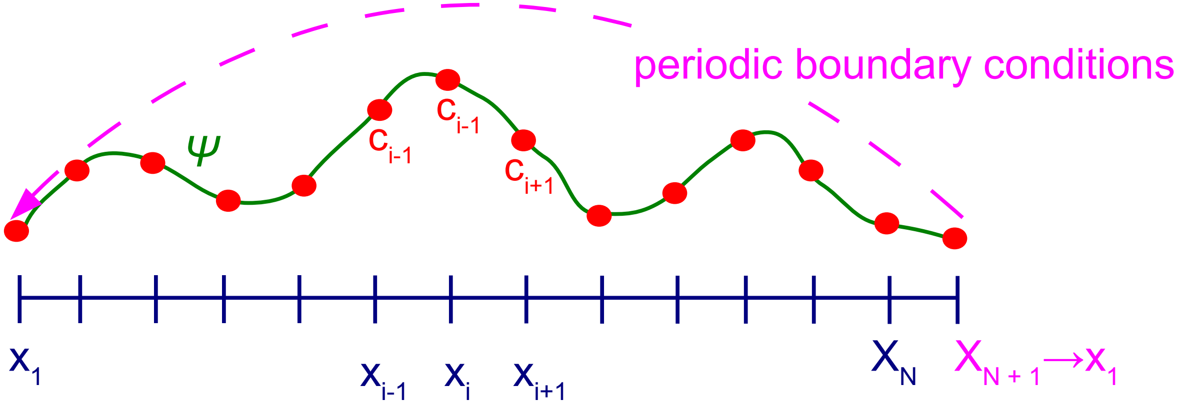
Determine an initial state
Solve the set of ordinary differential equations in time
Obtain $\Psi(t)$ *in terms of* $c_i(t_k)$ for (a set of) times t_k

Analyze

Extract the information from the $c_i(t)$

– electron spectra, ionization rates, high harmonic responses, …

Grid representation of $\hat{T}$ and $\hat{V}$



$$\hat{T}$$

$$\left(\frac{1}{\Delta x}\right)^2 \begin{pmatrix} 2 & -1 & 0 & \dots & 0 & -1 \\ -1 & 2 & -1 & 0 & \dots & 0 \\ 0 & -1 & 2 & -1 & \dots & 0 \\ & & \ddots & \ddots & \ddots & \\ 0 & \dots & 0 & -1 & 2 & -1 \\ -1 & 0 & \dots & 0 & -1 & 2 \end{pmatrix}$$

Finite differences rep. of -Δ

$$\hat{V}$$

$$\begin{pmatrix} V(x_1) & 0 & 0 & \dots & 0 \\ 0 & V(x_2) & 0 & \dots & 0 \\ \vdots & & \ddots & \vdots & \\ 0 & \dots & 0 & V(x_{N-1}) & 0 \\ 0 & 0 & \dots & 0 & V(x_N) \end{pmatrix}$$

Storage ~ 3 N
Accuracy *T* ~ $\sim \mathcal{O}[(\Delta x)^n]$

Storage ~ N
Accuracy *V* ~ $\sim \mathcal{O}[(\Delta x)]$

Simple & straight forward, very limited accuracy

Basis representation of $\hat{T}$ and $\hat{V}$

General basis φ_i

Full matrices

$$\hat{T}_{ij} = -\int dx \varphi_i(x) \frac{\partial^2}{\partial x} \varphi_j(x)$$

Matrix elements

$$\hat{V}_{ij} = \int dx \varphi_i(x) V(x) \varphi_j(x)$$

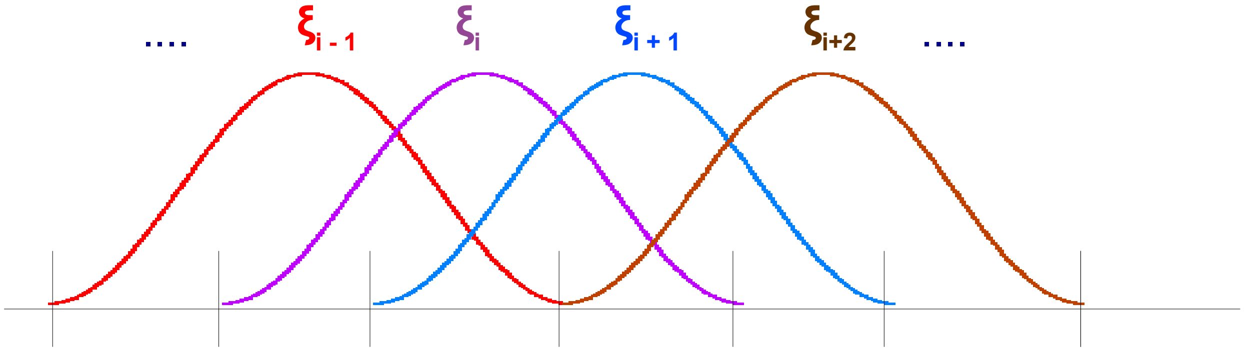
Storage ~ N^2, accuracy can be exponential in N $\sim \mathcal{O}(h^N)$

Rapid convergence, more elaborate setup (integrations), large storage

Local basis representation of $\hat{T}$ and $\hat{V}$

Local basis ξ_i

(Example B-splines)



Band matrices

$$d_n = -\int dx \xi_i(x) \frac{\partial^2}{\partial x} \xi_{i+n}(x)$$

Half-bandwidth n ~ order of splines

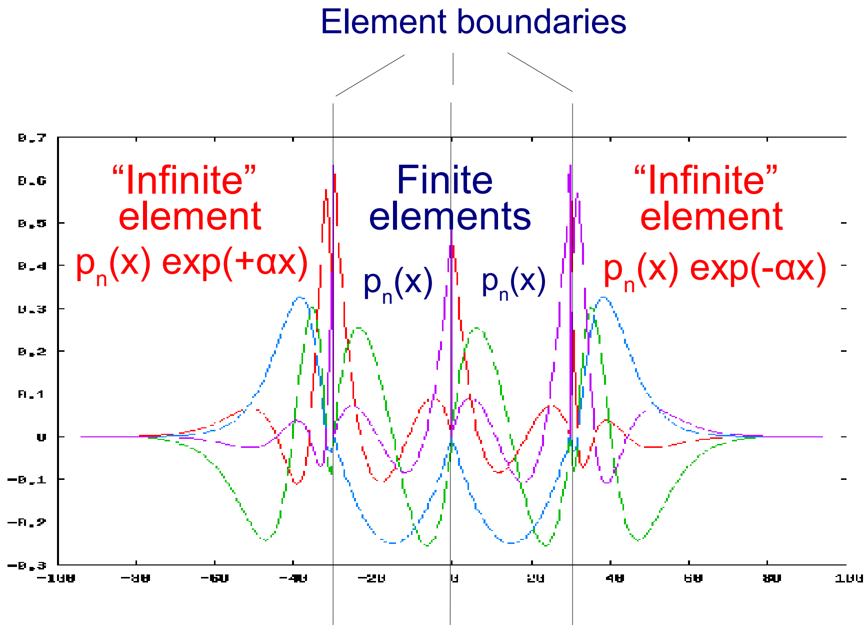
Storage ~ nN

Accuracy ~ $\mathcal{O}[(\Delta x)^n]$

$$\begin{pmatrix} d_0 & d_1 & d_2 & 0 & \ldots & 0 & d_{-2} & d_{-1} \\ d_{-1} & d_0 & d_1 & d_2 & 0 & \ldots & 0 & d_{-2} \\ d_{-2} & d_{-1} & d_0 & d_1 & d_2 & 0 & \ldots & 0 \\ 0 & d_{-2} & d_{-1} & d_0 & d_1 & d_2 & 0\ldots & 0 \\ 0 & 0 & \ddots & \ddots & \ddots & \ddots & \ddots & 0 \\ 0 & 0 & d_{-2} & d_{-1} & d_0 & d_1 & d_2 & 0 \\ 0 & \ldots & 0 & d_{-2} & d_{-1} & d_0 & d_1 & d_2 \\ d_2 & 0 & \ldots & 0 & d_{-2} & d_{-1} & d_0 & d_1 \\ d_1 & d_2 & 0 & \ldots & 0 & d_{-2} & d_{-1} & d_0 \end{pmatrix}$$

Easy setup (simple integrations), tunable accuracy

Local basis sets: finite elements



FEM – finite elements method

Piece-wise approximation
by a smooth basis:

- polynomials $p_n(x)$
- others, e.g. $p_n(x)\,e^{-\alpha x}$

Can be combined with Gauss quadratures to give the

"FEM-DVR" method

finite elements in "discrete variable representation"

Mathematically and computationally
very similar to B-splines
Possibly more flexible...

Time-propagation

Solve a set of ordinary (usually linear) differential equations

$$i\frac{d}{dt}\vec{C} = \hat{H}(t)\vec{C}$$

Formal solution

$$\vec{C}(t+\tau) = T\exp\left[-i\int_t^{t+\tau} dt'\,\hat{H}(t')\right]\vec{C}(t)$$

Highly symbolic notation:

T … time-ordered product!

How to obtain the exponential of the matrix?

Make numerical sense of it:

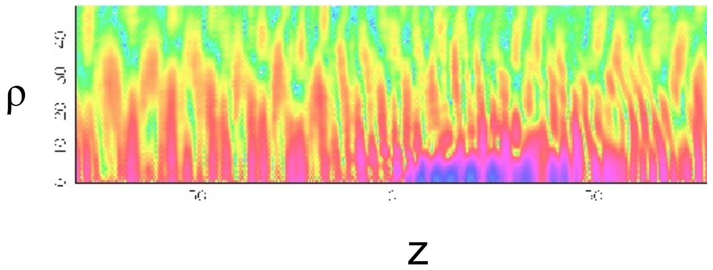
Runge-Kutta
Crank-Nicolson
Split-step
… and many more, more elaborate methods

Absorption of outgoing flux

What is the problem?

During a few-cycle pulse,
electrons can move VERY far
much further than just the quiver amplitude

either
reflection from simulation box boundaries



<= **bad (aka wrong) results**

or
VERY large simulation box

<= **VERY costly**

or
<u>traceless</u> absorption of flux

<= **non-trivial**

Extracting information from Ψ(x,t): excitation and HHG

Excitation

Probability of finding an excited *state after the pulse is over*

$$P_n = |\langle \Psi(T) | \Phi_n \rangle|^2 \quad \text{or} \quad P_n = |\vec{C}^*(T) \cdot \vec{C}_n|^2$$

Straight forward and easy to compute

High harmonic spectra

Time-dependent dipole $\vec{d}(t) = \langle \Psi(t) | \vec{r} | \Psi(t) \rangle = \vec{C}^*(t) \cdot \hat{X} \vec{C}(t)$

Harmonic power spectrum $h(\omega) = |\mathcal{F}[\vec{d}(t)](\omega)|^2$

Reasonably simple, some care for density of time-grid, smooth beginning and end of field

Extracting information from Ψ(x,t): ionization

Fundamentally, ionization is an asymptotic concept
– well defined only at large times

Easy to compute only when we know
the stationary states of the field free system

Single ionization yield

Easy, when all bound states C_n are available:

$$Yield = 1 - \sum P_n = 1 - \sum |\vec{C}^*(T) \cdot \vec{C}_n|^2$$

Double ionization yield

Harder: need to know bound and single-electron scattering states

Alternative definition

Yield ~ probability of finding the electron(s)
outside a small box surrounding the nucleus
(simple and efficient, reasonably reliable)

Extracting information from Ψ(x,t): photo-electron spectra

Fundamentally, photo-electron energy is an asymptotic concept
– well defined only at very large times and distances

Generally hard
(1) Need to keep the complete wave function until the pulse is over
=> Very large simulation boxes needed
(2) Need to asymptotically analyze Ψ(x,T)

Note: we do not have the exact double-photoelectron states!

$$\nexists\ \Phi_{E_1,E_2}: \quad \xcancel{P(E_1,E_2) = |\langle\Psi(T)|\Phi_{E_1,E_2}\rangle|^2}$$

Ways out:

(1) Analyze only asymptotic part of wave function using free-electron states
=> need to wait even longer until also slow electrons come there
Long after the end of the pulse
=> need an even larger box

(2) Keep track of current through a surface, perfectly absorb beyond that surface
=> need to wait long, but at least can keep the box small

It's no mystery: try it in Python...

Your choice of the discretization
 – for simplicity start from finite differences

Choose a grid: $(x_0,x_1,\ldots,x_N)$

Set up the matrices T, V, X, H=T+V

Solve the eigenproblem for an initial state:
 “C = eig(H)” (eig is a standard SciPy eigensolver)

Choose a laser pulse:
 e.g. “function f = field(t)
 f = cos(ωt) cos(π t/T)”

Define the derivative
 “function d = hamiltonian(c,t)
 D = (T + V + field(t)*X) * c”

Time-propagate from t = -T to t = T
 function “ode()” [solver for moderately stiff ODEs]

Analyze:
 Determine ionization
 High harmonic response
 Excited state populations

Python:
 It's free!
 It's a programming language
 It's great!

Numerical approaches:

More than 1 electron

Scaling of the computational effort

N ...number of discretization points ~ phase space covered by the solution

$\sim P_{max} \times X_{max} \times$ Pulse duration

Electron in the laser field:

$P_{max} \sim E_{max} \times \lambda_{laser}$ and $X_{max} \sim E_{max} \times (\lambda_{laser})^2$

Pulse durations grow $\sim \lambda_{laser}$

Total effort

$\sim (\lambda_{laser})^4$ (linear polarization)

$\sim (\lambda_{laser})^7$ (general polarization)

Number of particles: N grows exponentially !

Limitations

- Long wave length
- Several “active” electrons
- Very strong pulses

The curse of dimensions

The quantum wave function Ψ

(tells us everything we can know about the system)

$\Psi(x_1,y_1,z_1,x_2,y_2,z_2,\ldots,x_n,y_n,z_n)$ n...number of electrons

n = 1 – one electron (3 dimensions) 100 points in each dimension	100^3 points 8 MB	store on a memory stick
n = 2 – two electrons (6 dimensions)	100^6 points 8 TB	hard disk (rather large)
n = 3 – three electrons (9 dimensions)	100^9 points 8 ExaB	e.g. 10^9 DVDs (4000 metric tons)

The “curse of dimensionality” strikes early on!

Resources needed

1d, single electron problem – try run it on your smartphone

3d problem, cylindrical symmetry – laptop, desktop (minutes – hours)

3d problem, no symmetry: good workstation, small cluster (hours – days)

Larger (2 or more electrons): really large computers

Simplifying models

Lower-dimensional models

“1d-Helium”

$$H(x_1, x_2; t) = \sum_{i=1,2} \left[-\frac{1}{2}\frac{\partial^2}{\partial x_i^2} - \frac{2}{\sqrt{x_i^2 + a}} \right] + \frac{1}{\sqrt{(x_1 - x_2)^2 + b}}$$

Adjust *a* and *b* to obtain spectra similar to 3d Helium
NOTE: a=1/2 give the exact He$^+$ ground state energy E_0= -2 (atomic units)

Easy to program, fast solving:

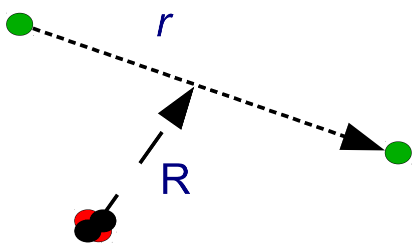
E.g. $x_{1,}x_2$ – grid of 1000 x 1000 ~ 16 MB for the wave function
FFT and split step very efficient for time propagation
Very nice toy!

Lower-dimensional models: restricted dynamics

Restrict dynamics **A. Becker & group**

e.g. only center-of-mass motion of two electrons

$$H(\mathbf{R},\mathbf{r},t) = \frac{\mathbf{P}^2}{4} + \mathbf{p}^2 - \frac{Z_{\text{nucl}}}{|\mathbf{R}+\frac{\mathbf{r}}{2}|} - \frac{Z_{\text{nucl}}}{|\mathbf{R}-\frac{\mathbf{r}}{2}|} + \frac{1}{r} - \frac{\mathbf{P}\cdot\mathbf{A}(t)}{c}$$



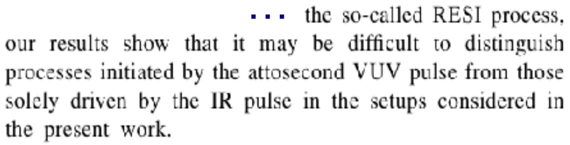
Jacobi coordinates...

R → Z

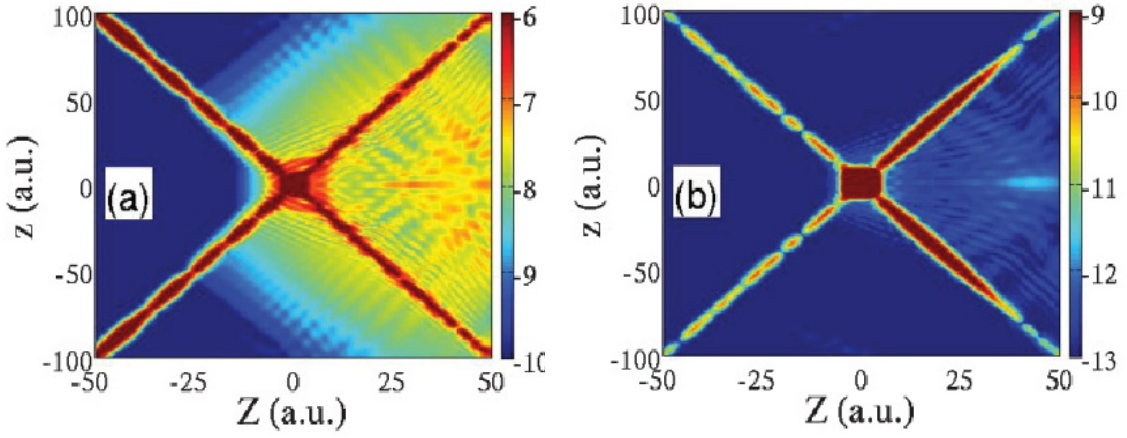
$$H(Z,\rho,z,t) = \frac{P_Z^2}{4} + p_\rho^2 + p_z^2 + \frac{1}{\sqrt{\rho^2+z^2}} - \frac{2}{\sqrt{\rho^2/4+(Z+z/2)^2+a^2}} - \frac{2}{\sqrt{\rho^2/4+(Z-z/2)^2+a^2}} - \frac{P_Z\,[A_1(t)+A_2(t)]}{c},$$

Investigated XUV + IR ionization

Chen et al., PRA 82, 033626 (2010)


... the so-called RESI process, our results show that it may be difficult to distinguish processes initiated by the attosecond VUV pulse from those solely driven by the IR pulse in the setups considered in the present work.

Probability distribution



XUV + IR IR only

But...

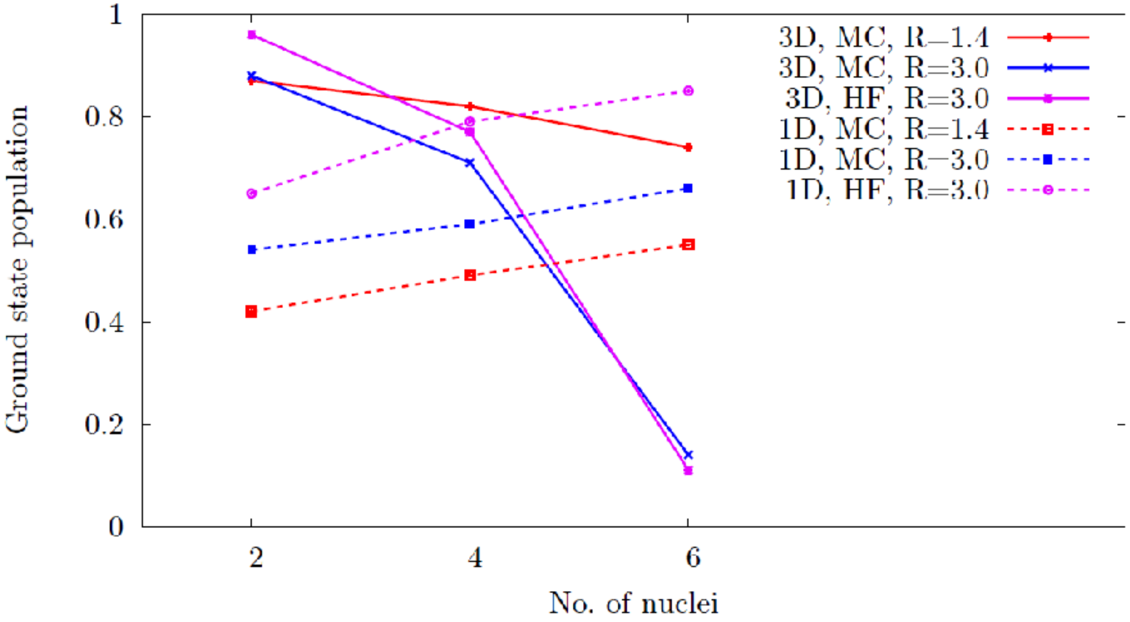
A note of caution for lower-dimensional models

Ionization of molecules

With increasing size
2 – 4 – 6 atoms
2 – 4 – 6 electrons

A simple process!
Shouldn't 1-d models give us
a feeling for what happens?

3-d calculations vs. 1-d calculations
(solid lines vs. dashed lines)



1-d models can be qualitatively wrong

Simple models can be inspiring, but do not base conclusions on them!

Single active electron approximation

Not very precisely defined: somehow “freeze” all electrons except for one

[since 1987: Kulander, Schafer,...]

Build a potential V_{eff} that simulates the effect of the frozen electrons

$$H(t) = -\frac{1}{2}\Delta + V_{eff}(\vec{r})$$

“invent” a good V_{eff}: match excitation energies, scattering properties...

Slater determinant, n-1 fixed functions, let one be time-dependent

Use exact Hamiltonian $$H(t) = \sum_i -\frac{1}{2}\Delta_i + \frac{Z}{\vec{r}_i} + \vec{\mathcal{E}}\cdot\vec{r} + \sum_{i>j}\frac{1}{\vec{r}_i - \vec{r}_j}$$

Ansatz: $\Psi(r_1,r_2,\ldots,r_n;t) = \det[\Phi_1(r_1;t)\ \Phi_2(r_2)\ \ldots\ \Phi_n(r_n)]$

Only least bound orbital evolves in time

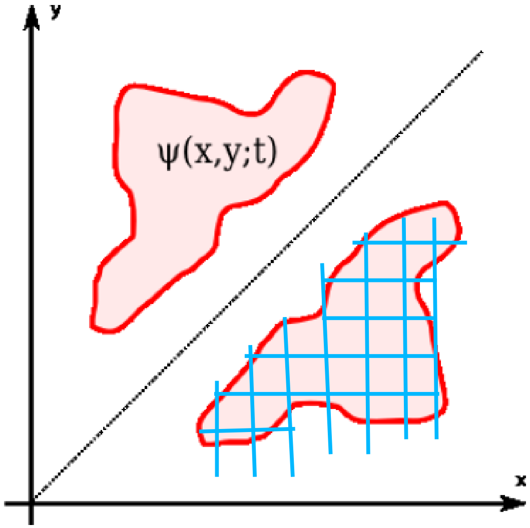
Field free HF orbitals, frozen

Note: gauge question arises!

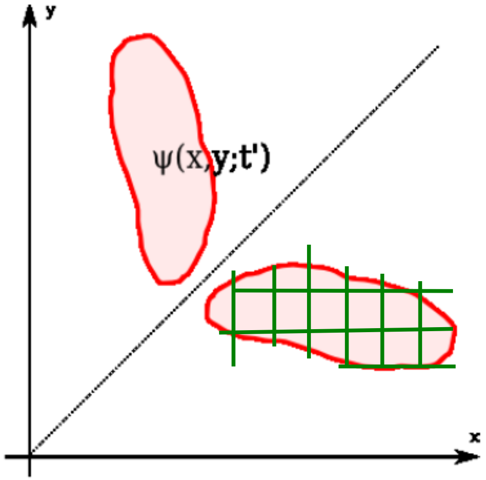
Neither is exact, errors of both are difficult to estimate
Make the simplest choice for your purpose

Non-linear approximations to the TDSE

As the wave function evolves, different discretizations would be optimal



Some time t_0...



...some later time t_1

Adjust the discretization to the evolution of the solution

The feed-back introduces non-linearity into the equations

The current solution acts back on its representation

→ NONLINEAR EQUATIONS

Examples:
- Adaptive meshes
- Time-dependent Hartree-Fock

The two-electron problem

He in linearly polarized fields – fixed discretization

5-dimensional problem, just at the limit where full discretizations are still possible

Combine spherical harmonics with grids on r_1 and r_2

Finite-differences grid

[K. Taylor & group]

Double-ionization photo-electron spectra
at wavelength ~ 400 nm
one result @ 800 nm

FEM-DVR grid + split-step propagator

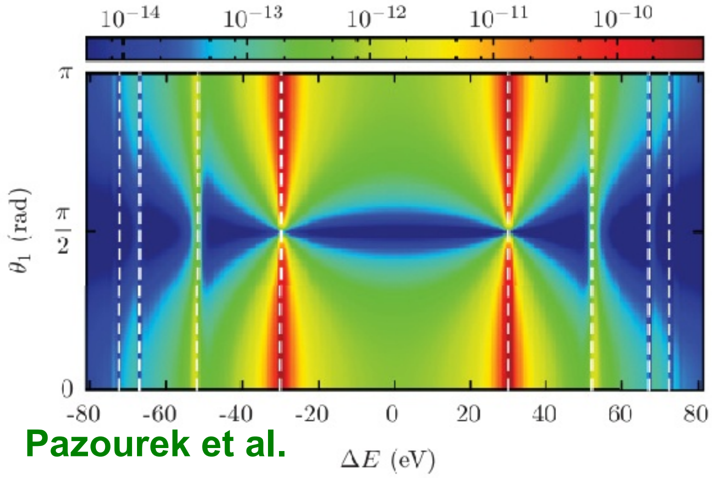
[J. Feist et al. PRA 77, 43420 (2008)]

mostly at shorter wave length
numerically very solid results

Two-photon double-ionization

Very large computer resources

PHYSICAL REVIEW A **83**, 053418 (2011)



Pazourek et al.

FIG. 3. (Color online) Two-dimensional energy-angular differential distribution $P^{\mathrm{DI}}(\Delta E,\theta_1)$ for an xuv pulse with $T = 4.5$ fs and $\hbar\omega = 80$ eV as a function of the energy difference $\Delta E = E_1 - E_2$ between the two electrons, and emission angle θ_1, relative to the polarization axis. The vertical dashed white lines show the expected positions of the peaks for the sequential process; see Fig. 1. The horizontal nodal line $\theta_1 = \pi/2$ is visible except near shake-up resonances.

Tackling the multi-electron problem

Multi-electron systems

Isn't the single-electron picture sufficient?

NOT for

- Inherently two- or more-electron processes:
  - Auger processes in the field
  - Double ionization

- Severe “perturbations” by multi-electron effects
  - As in molecular orbital tomography

- Getting the numbers right

Approaches that work:
CI and R-matrix

CI – configuration interaction

[A. Saenz & group]:

Get a basis from a dedicated CI calculation
Select by physical insight, energy range
Expand time-dependent problem in this basis

Very efficient, very precise in some parameter ranges

Possible limitations:
    Continuum difficult to include
    Plays an important role in strong-field polarization

R-matrix Floquet and time-dependent R-matrix

[Burke, Joachain, H.van der Hart]

Exploit existing structure codes to compute inner region

R-matrix:
    match value and derivative at the boundary
    Between outer region scattering solutions to inner region

Question of different gauges:
    inner region must be length gauge
    outer region is more efficient in velocity gauge

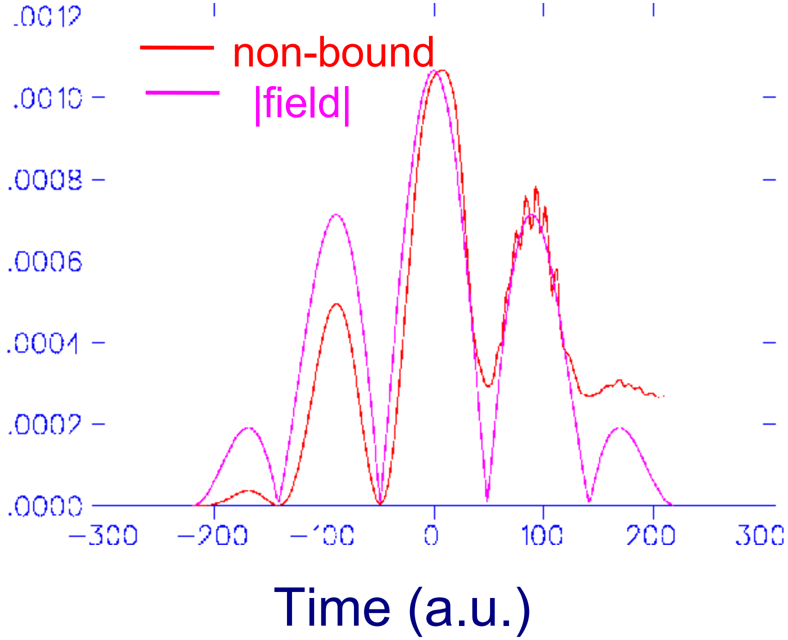
Possibile limitations:
    Similar as CI

Continuum states and polarization

Bound/unbound is an asymptotic concept

Spectral distinction between bound and unbound only for time-independent Hamiltonians

In CI, continuum states are excluded from the core → play a role in polarization!



Contents of field-free non-bound states during ionization of H (length gauge)

“Deep tunneling regime”:
Keldysh γ ~ 0.3 !

Conceptual question:

Can we look into the laser cycle?

How to define an ionization rate during the cycle??

There is no “adiabatic” Hamiltonian to distinguish bound-continuum during the pulse

Approaches that work:
Very large scale full discretizations

Carry the multi-surface Born-Oppenheimer picture to the extreme (F. Martin & group)

$$\Psi(\vec{r}_1,\vec{r}_2,R) = \sum_i \chi_i(R)\Phi_R^{(i)}(\vec{r}_1,\vec{r}_2)$$

$$h_R(\vec{r}_1,\vec{r}_2)\Phi_R^{(i)}(\vec{r}_1,\vec{r}_2) = E_R^{(i)}\Phi_R^{(i)}(\vec{r}_1,\vec{r}_2)$$

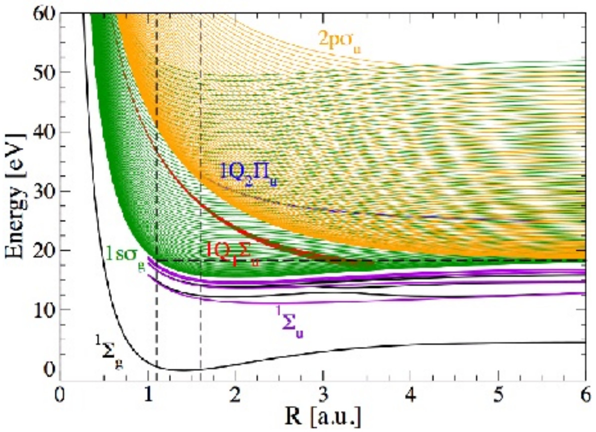
Use a very large number of $E_R^{(i)}$
including: continuum states
doubly excited states
relevant couplings

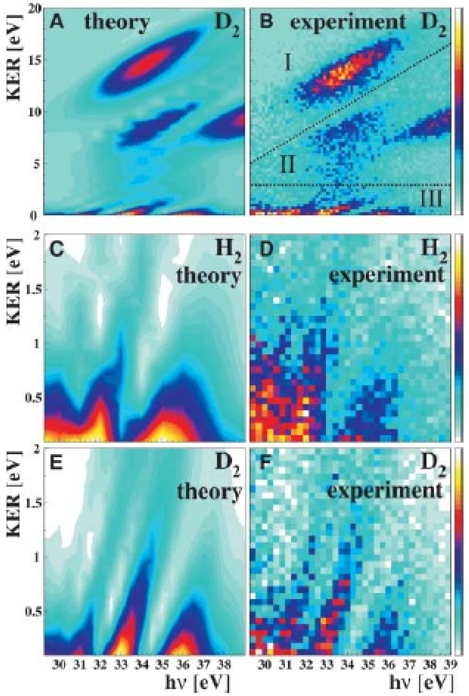
Obtain $\Phi_R^{(i)}$ by
(B-spline) discretization on a very large box

Advantages:
best results results on laser-dissociation of H_2

Disadvantages:
Limited to H_2
Enormous computer needs

Very large computer resources





[F. Martin et al., Science 315, 629 (2007)]

Approaches that do not work: TDHF and TDDFT

(Time-dependent) Hartree-Fock method – TDHF

Approximate the electron wave function by
a single determinant of inter-dependent three-dimensional orbitals

$$\Psi(r_1,r_2,...,r_n;t) = \det[\Phi_1(r_1;t)\ \Phi_2(r_2;t)\ ...\ \Phi_n(r_n;t)]$$

Orbitals Φ are optimal for each time t

We hope/assume that the determinant ansatz is suitable

+ compact representation of the wave function (some 10 MB)
+ straight forward extraction of experimental observables

- large computational effort (much larger than TDDFT)
- no checks of accuracy
- **fails for strong time-dependent interactions**

Time-dependent density functional theory - TDDFT

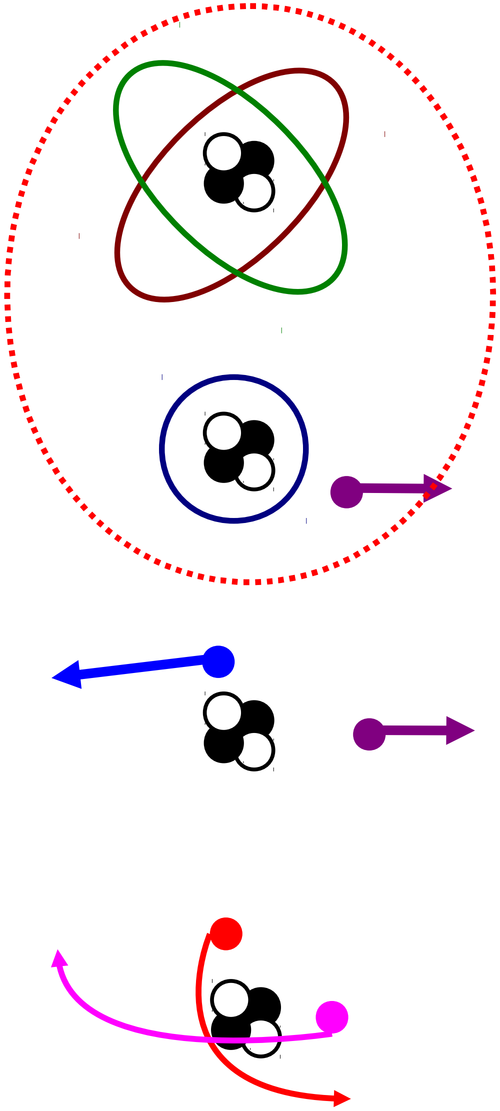
suffers from similar problems, possibly more severely

Why time-dependet Hartree-Fock fails

A two-electron system in various states

Symbolic representation

Quantum wave $\Psi(r_1,r_2)$



Superposition state

$$\Psi(r_1,r_2) \approx$$

Initial state (a configuration)

$$A_{ab}\Phi_a(r_1)\Phi_b(r_2)$$

Partial ionization

$$+$$

Ionized state (another configuration)

$$A_{cd}\Phi_c(r_1)\Phi_d(r_2)$$

$$+$$

Another state (more configurations)

$$A_{ef}\Phi_e(r_1)\Phi_f(r_2)$$

Yet another state....

$$+$$

$$\vdots$$

A non-linear method: MCTDHF

Solve the time-dependent Schrödinger equation for a few particles

Multi-Configuration Time-dependent Hartree -Fock

Hartree: Few-particle wave function ≈ Product of single particle wave function

$$\Psi(\boldsymbol{x}_1,\ldots,\boldsymbol{x}_f) \approx \phi_1(\boldsymbol{x}_1)\phi_2(\boldsymbol{x}_2)\ldots\phi_f(\boldsymbol{x}_f)$$

Multi-Configuration: use linear combination of products

$$\Psi(\boldsymbol{x}_1,\ldots,\boldsymbol{x}_f) \approx \sum_{j_1=1}^{n}\ldots\sum_{j_f=1}^{n} A_{j_1,\ldots,j_f}\phi_{j_1}(\boldsymbol{x}_1)\phi_{j_2}(\boldsymbol{x}_2)\ldots\phi_{j_f}(\boldsymbol{x}_f)$$

Time-dependent: single-particle functions $\phi_j(\boldsymbol{x},t)$ and

linear coefficients $A_{j_1,\ldots,j_f}(t)$ depend on time

[Review of MCTDH: Beck et al. Physics Reports 324, 1 (2000)]

Fock: restrict to anti-symmetric:

$$A_{\ldots j_\kappa \ldots j_\mu \ldots} = - A_{\ldots j_\mu \ldots j_\kappa \ldots}$$

System of non-linear PDEs

$$i\frac{d}{dt}A_{l_1\dots l_f}(t) = \sum_{l_1\dots l_f} \underbrace{\langle\phi_{j_1}\dots\phi_{j_f}|H(t)-F^{(1)}(t)|\phi_{l_1}\dots\phi_{l_f}\rangle}_{\text{Galerkin matrix}} A_{l_1\dots l_f}$$

$$i\frac{d}{dt}\phi_j = F^{(1)}(t) + \overbrace{P_\perp[\boldsymbol{\phi}]}^{\text{Projector}} \sum_{l,m=1}^{N} \overbrace{(\rho[\mathbf{A}])^{-1}_{jl}}^{\text{Density matrix}} \overbrace{\langle((H-F^{(1)})[\mathbf{A},\boldsymbol{\phi}]\rangle_{lm}}^{\text{Mean field matrix}}\phi_m$$

“Gauge fixing” $F^{(1)}(t) = \sum_{i=1}^{f} F(\boldsymbol{x}_i,t)$ arbitrary self-adjoint single-particle op. fixes non-uniqueness of the ansatz

Hamiltonian with two-body interactions

$$H = \sum_{k=1}^{f}\sum_{k<n} H(x_k,x_n,t)$$

Projector

$$P_\perp = 1 - \sum_{n=1}^{N} |\phi_n\rangle\langle\phi_n|$$

Single particle density matrix

$$\rho_{jl} = \sum_{j_2\dots j_f} A^*_{jj_2\dots j_f} A_{lj_2\dots j_f}$$

Two particle density matrix

$$\rho^{(2)}_{j_1j_2l_1l_2} = \sum_{j_3\dots j_f} A^*_{j_1j_2j_3\dots j_f} A_{l_1l_2j_3\dots j_f}$$

Mean field operators

$$\hat{H}_{mn} = \frac{(f-1)(f-2)}{2}\sum_{m_2n_2}\langle\phi_{m_2}|H|\phi_{n_2}\rangle\rho^{(2)}_{mm_2,nn_2} \quad \text{for gauge} \quad F=0$$

Example

HHG – a dynamic multi-electron process

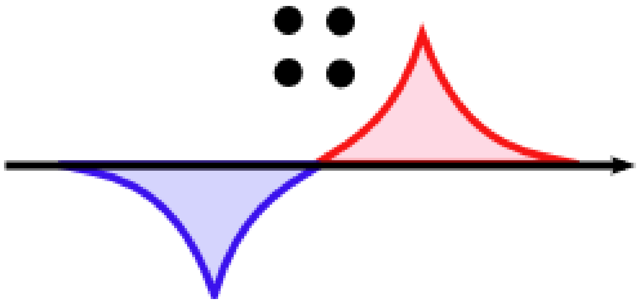
HHG for the following system:

- Diatomic, homonuclear molecule
Internuclear distance R = 2.8 a.u.
3d with cylindrical symmetry

- Screened Coulomb potential
I_p = 0.58 a.u. (N_2)

- Laser pulse:
800 nm, 1 cycle, 3 x 10^{14} W/cm^2

4 active electrons
ungerade outer orbital



[G. Jordan et al., NJP 10, 025035 (2008)]

Do single active electron models work?

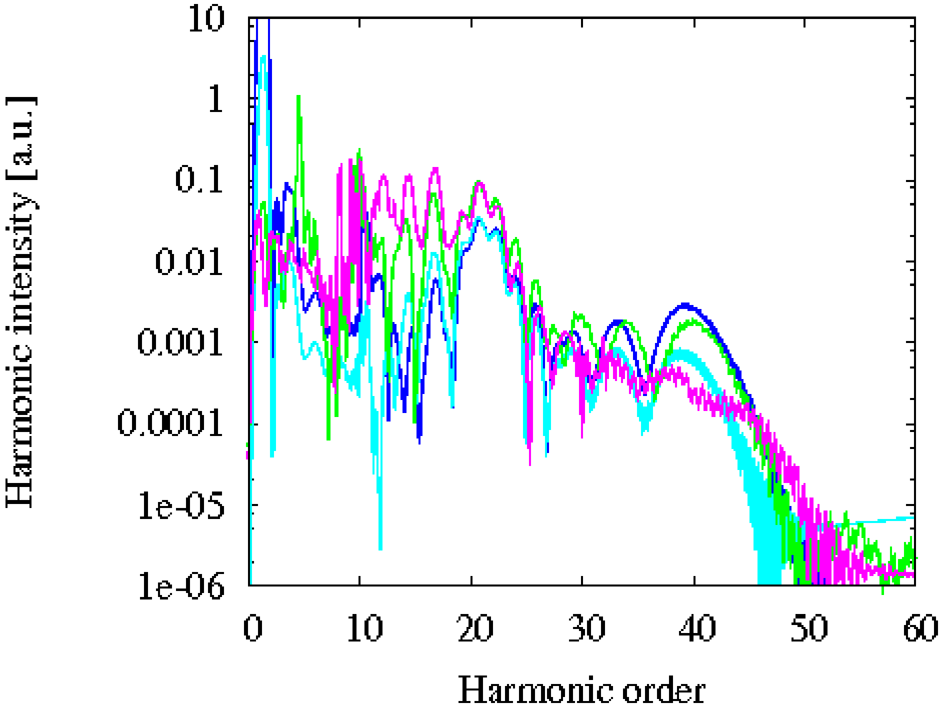
MCTDHF

SFA: HOMO + plane waves

Effective 1e Hamiltonian

$H_{eff} = |0\rangle E_0\langle 0| + (1-|0\rangle\langle 0|)H_m(1-|0\rangle\langle 0|)$

Multi-electron corrections to the transition dipole [Patchkovskii et. al



(Known) single active electron models do not work.

More than one electron moves!

Factor MCTDHF wave function into (core) x (single electron)

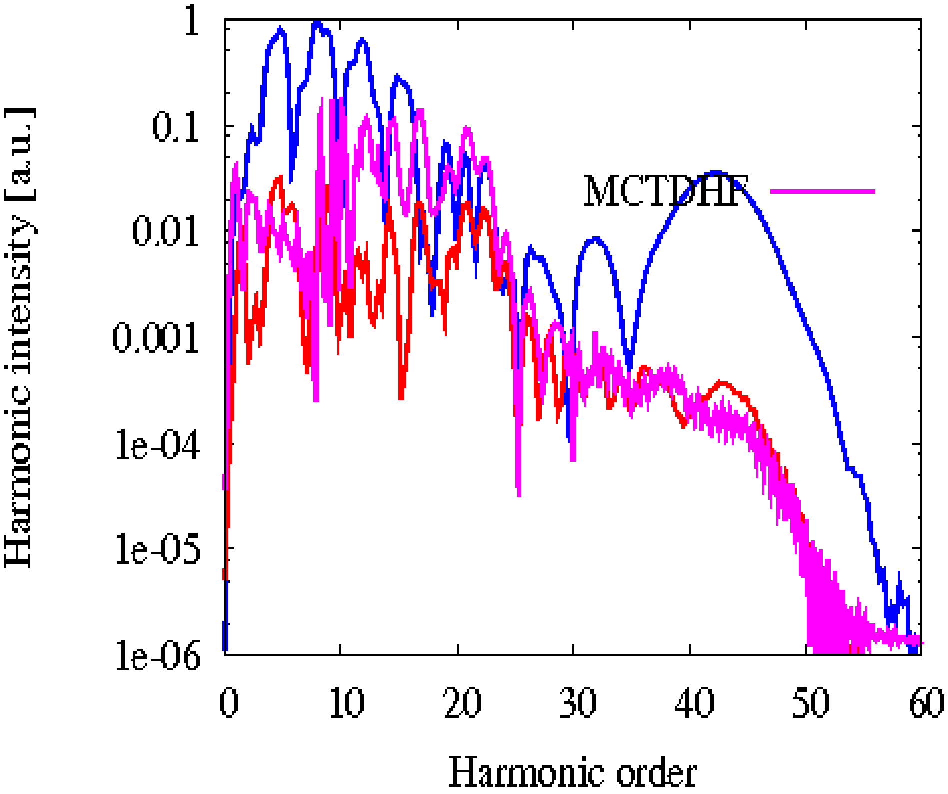
$$\Psi_{MCTDHF} \sim \Psi_{factor} = \Phi_{core}\varphi_e$$

choose Φ_{core}

determine φ_e

Static core:
Φ_{core} = ion without field

Dynamic core:
$\Phi_{core}(t)$ = ion in the field



[G. Jordan et al., NJP 10, 025035 (2008)]

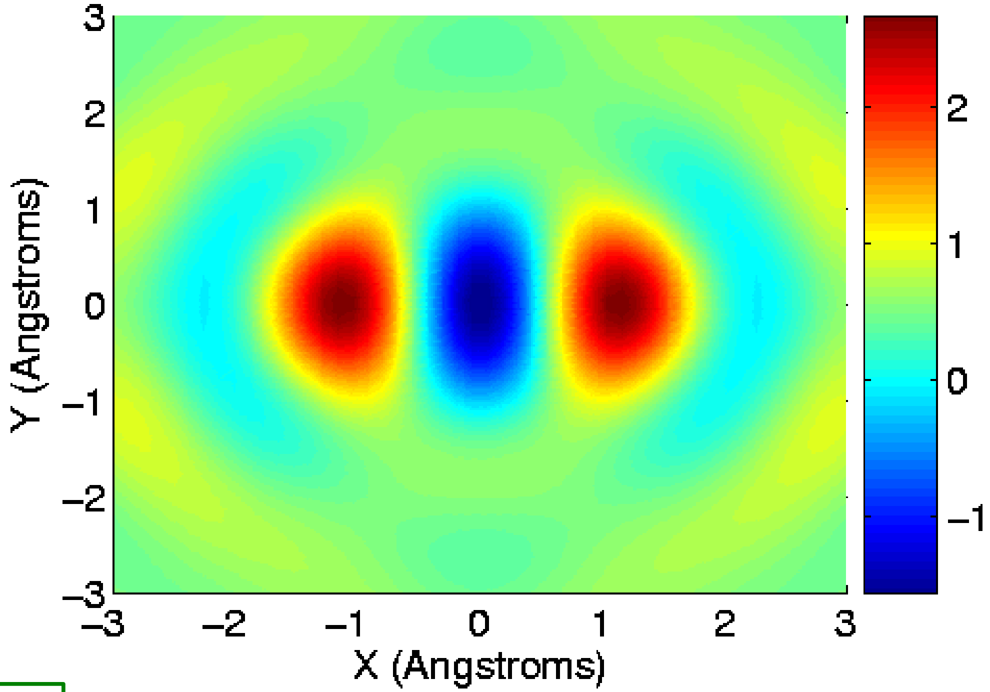
Why does it matter?

If we want to learn from harmonics about electronic structure and dynamics...

Molecular orbital tomography

Idea: extract electronic structure
measured under different polariz

$2\sigma_g$ HOMO orbital of N_2 ?



[Itatani, Nature **432,** 867 (2004)]

Do we have "single electron" Information in the harmonics?

Electron energy spectrum: $2U_p$

Assume release at time t_0

Momentum at $t_0 \sim 0$

Acceleration by the laser field

$$\vec{p}(\infty) - \vec{p}(t_0) = -\int_{t_0}^{\infty} \vec{\mathcal{E}}(t) \quad = -\vec{A}(t_0)$$

$$\vec{p}(t_0) = \vec{p}(\infty) + \vec{A}(t_0) \quad = 0$$

Maximal acceleration = $\pm A_0 = \pm \varepsilon_0/\omega$

Maximal electron energy ~ 2Up = $2\varepsilon_0^2/4\omega^2$

“direct electrons”

The 10 U_p cutoff

Acceleration of the electron

from t_0 until t_1 $$-\int_{t_0}^{t_1} \mathcal{E}(t)dt = A(t_0) - A(t_1)$$

from t_1 until end of pulse $$-\int_{t_1}^{\infty} \mathcal{E}(t)dt = A(t_1)$$

final momentum without scattering $A(t_0)$

final momentum if it
reverses momentum at t_1 $$|p_{final}(t_0)| = |A(t_0) - 2A(t_1)|$$

maximal momentum $$p_{\max} = \max(t_0)|A(t_0) - 2A(t_1)|$$

numerically determine p_{max}

=> $$\boxed{E_{max} = 10.00..\ U_p}$$

Absorption of outgoing flux

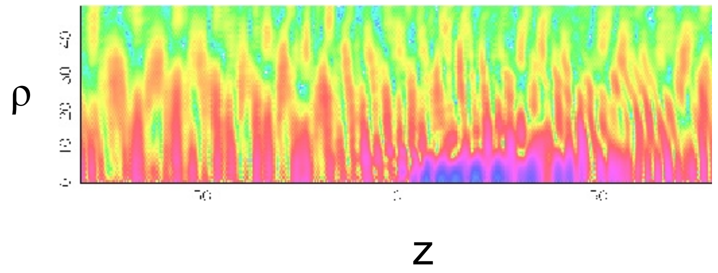
Photo-electron spectra

Absorption of outgoing flux

What is the problem?

During a few-cycle pulse,
electrons can move VERY far
much further than just the quiver amplitude

either
reflection from simulation box boundaries



<= bad (aka wrong) results

or
VERY large simulation box

<= VERY costly

or
<u>traceless</u> absorption of flux

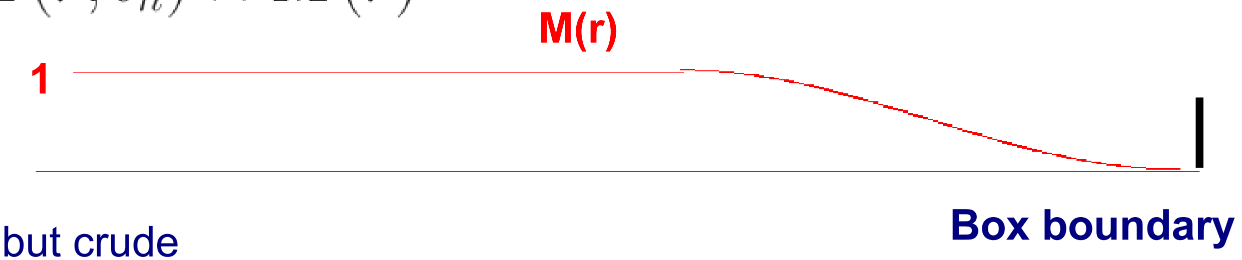
<= non-trivial

Methods for absorption

Absorption by mask

After each time step, multiply the wave function that comes close to the boundary
By some function that goes gently to 0 towards the box boundaries

$$\Psi(\vec{r}, t_n) \longrightarrow \Psi(\vec{r}, t_n) \times M(\vec{r})$$



Straight forward, but crude
Produces artefacts

Complex absorbing potentials

Add a potential with a (negative) imaginary part near the box boundary

$$H(t) \longrightarrow H(t) + V_{CAP}(\vec{r}), \quad \text{e.g.} \quad V_{CAP}(\vec{r}) = -i\alpha(|\vec{r}| - R)^n \text{ for } |\vec{r}| > R$$

Closer analysis shows that this is the differential version of an absorption by mask

Straight forward, less crude
Produces artefacts

A perfect absorber

irECS

infinite-range Exterior Complex Scaling

[A.S., Phys. Rev. A81, 53845 (2010)]

Triggered by COST STMS visit to F. Martin & group

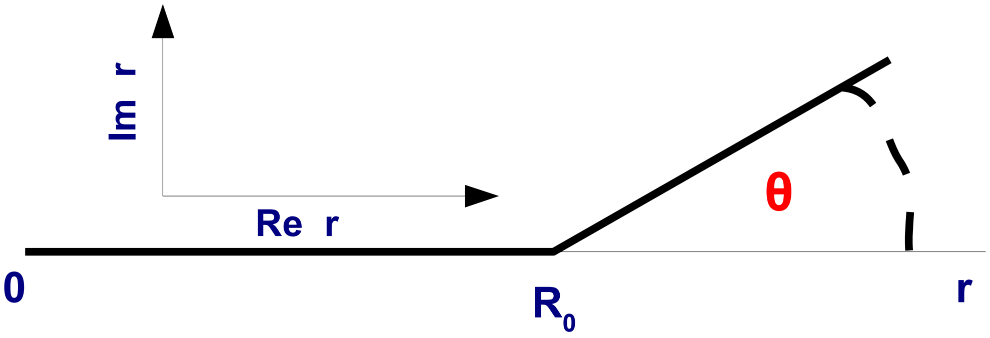
Exterior complex scaling

Hamiltonian is **analytic only beyond R_0**

complex scale only
beyond a finite distance R_0

$r \to r$ for $r < R_0$

$r \to R_0 + e^{i\theta}(r - R_0)$ for $r > R_0$



Outgoing wave boundary conditions

$\exp(ikx) \rightarrow \exp(i\cos\theta\, kx - \sin\theta\, kx)$

Outoing waves	$k > 0$	→	exponentially decaying functions, L^2 integrable
Ingoing waves	$k < 0$	→	exponentially exploding functions, not in L^2

Implementation of exterior complex scaling

Important technical complication

Bra and ket functions are not from the same space!!!

Exterior scaled Laplacian $\Delta_{R0,\Theta}$ is defined on discontinuous functions

$$\Psi(R_0 - 0) = e^{3i\theta/2}\Psi(R_0 + 0)$$

Discontinuity arises because we need to keep the norm constant

Discontinuity is reversed for the left hand functions

$$\Psi^*(R_0 - 0) = (e^{-3i\theta/2}\Psi)^*(R_0 + 0)$$

Matrix elements of $\Delta_{R0,\Theta}$

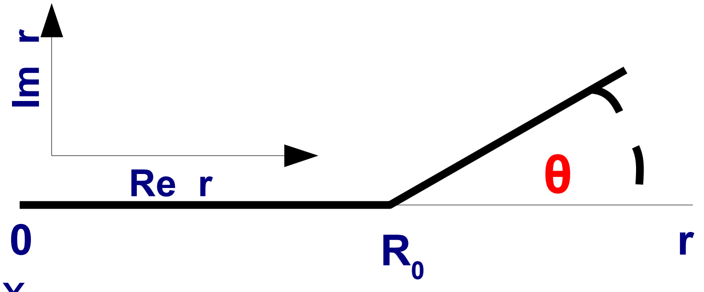
are computed by piece-wise integration $[0,R_0] + [R_0,\infty)$

Conditions easy to implement with a local basis set

IrECS – infinite range exterior complex scaling

[A.S., Phys. Rev. A81, 53845 (2010)]

Exterior complex scaling

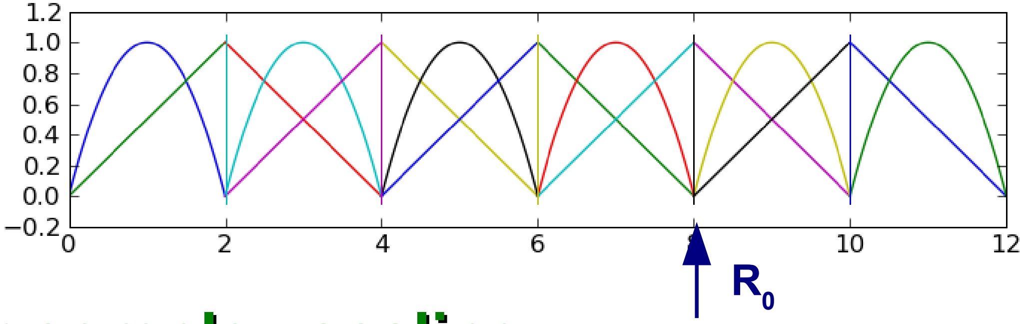


$r \to r$ for $r < R_0$

$r \to R_0 + e^{i\theta}(r-R_0)$ for $r > R_0$

Usually done with a finite discretization box...

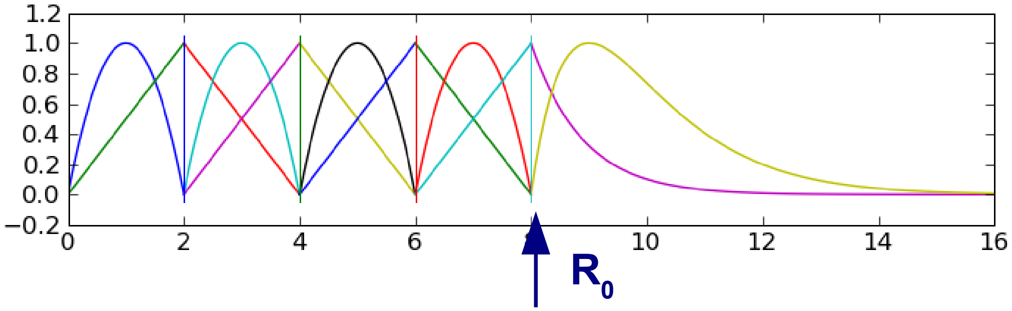
Example:
order 2 finite elements



Infinite range exterior complex scaling

Use an infinite size last element $[R_0,\infty)$

Example:
order 2 infinite last element



In practice: 20 ~ 30 functions on last element

Does it work?

Exterior complex scaling for the TDSE

Solve

$$i\frac{d}{dt}\Psi_{\theta R_0}(x,t) = H_{\theta R_0}(t)\Psi_{\theta R_0}(x,t)$$

Hope that

$\Psi_{\theta R0}(x) = \Psi(x)$ for $|x| < R_0$

Does it work?

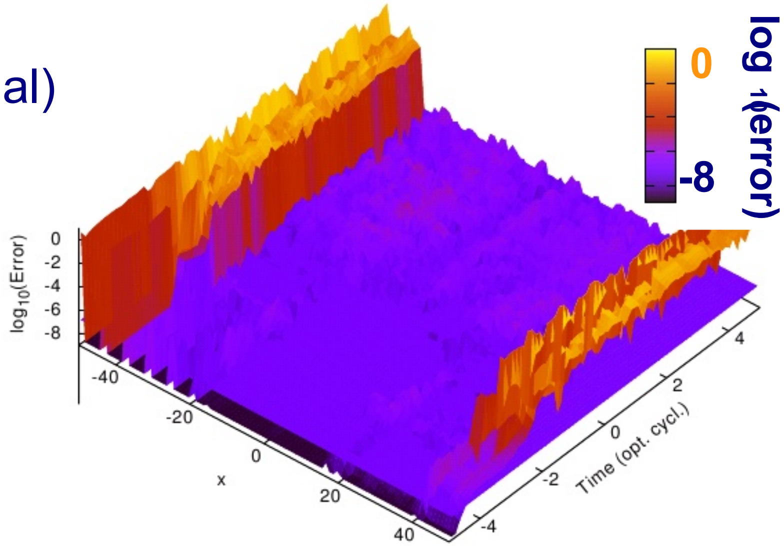
Atom in a strong dipole field

$$i\frac{d}{dt}\Psi(\vec{x},t) = \left[-\frac{1}{2}\Delta_{\vec{x}} + i\vec{A}(t)\cdot\vec{\nabla}_{\vec{x}} + V(\vec{x})\right]\Psi(\vec{x},t)$$

Pulse parameters: 2 x 10^{14} W/cm^2, 760 nm, 5 opt. cycl. FWHM

Massive ionization:
At the end of the pulse ~ 80% of the probability is outside [-40,40]

Accuracy of the solution (1-dimensional)



Relative error

$$|\Psi_{\theta R0}(x) - \Psi(x)| \,/\, |\Psi_{\theta R0}(x) + \Psi(x)|$$

Inside box ~ 10^{-7}

Works very nicely!
[Phys. Rev. A81, 53845 (2010)]

irECS is a perfect absorber

Errors are determined by the accuracy of the discretization

Mathematically speaking there are NO errors introduced by ECS itself

ch line +++ punch line +++ punch line +++ punch line +++ punch line +++ punch line +++ pun

How efficient is irECS?

How many discretization points do we need for perfect absorption?

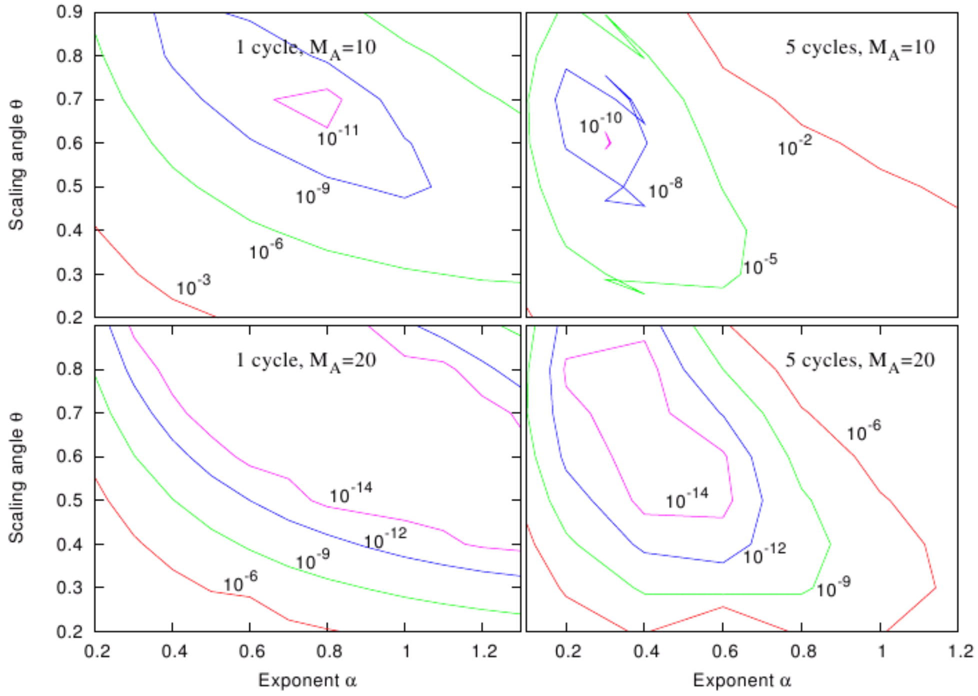
How many discretization points are needed?

Error measure: $||\Psi_{\theta R0}(x) - \Psi(x)||^2_{x<R0} / ||\Psi(x)||^2_{x<R0}$

Points in unscaled region 160

Points for absorption
$M_A = 10$

$M_A=20$



20 Absorption points are perfect!

Quite insensitive to scaling angle θ and exponential exp(-αx)

[Phys. Rev. A81, 53845 (2010)]

Compare to complex absorbing potentials (CAPs)

Errors of ECS and CAPs with identical discretizations

Method	M_A	A	θ or σ	q	$\mathcal{E}[-R_0, R_0]$
irECS	21	∞	0.6	—	2×10^{-15}
ECS	20	10	0.6	—	2×10^{-4}
ECS	40	20	0.5	—	1×10^{-7}
CAP	20	10	10^{-4}	4	3×10^{-3}
CAP	20	10	2×10^{-6}	6	4×10^{-3}
CAP	40	20	4×10^{-6}	4	3×10^{-4}
CAP	60	30	6×10^{-7}	4	1×10^{-5}

Number of points

Accuracy

[Phys. Rev. A81, 53845 (2010)]

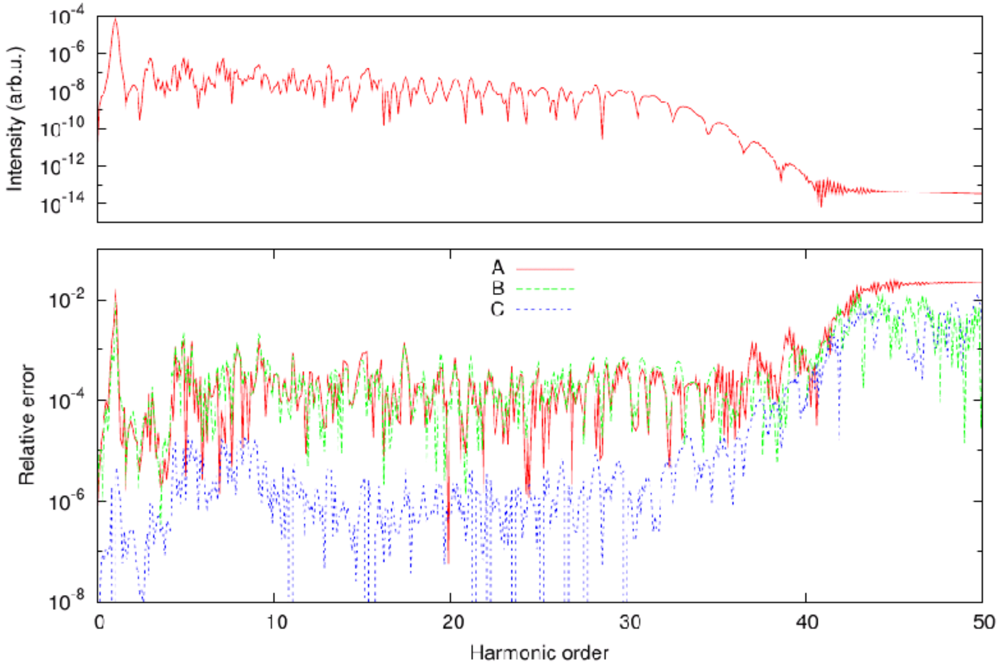
In 3 dimensions

Works just the same way and equally robust and efficient

Perfect high harmonic responses for H and model Ne

Very stable wave functions when varying scaling parameters R_0 and θ

High harmonic spectrum of the Hydrogen atom



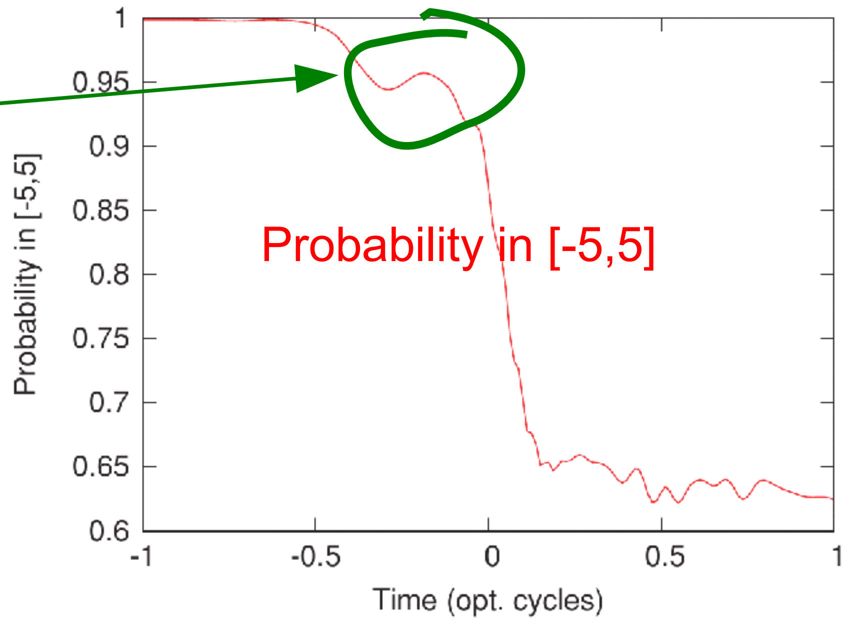
Relative error

Can we hope to backscale the irECS solution?

Observation:
Wave function is not just absorbed in the scaled region
It returns from the exterior scaled region without distortions

~ 1% probability returns
from scaled region $|x|>5$
into unscaled region $|x|<5$

Wave-function in $|x|<5$
accuracy $< 10^{-11}$ at all times



Dynamics is correctly encoded also in the scaled region!

Yes, we can hope... in practice: neither easy nor very important

Computation of photo-electron spectra

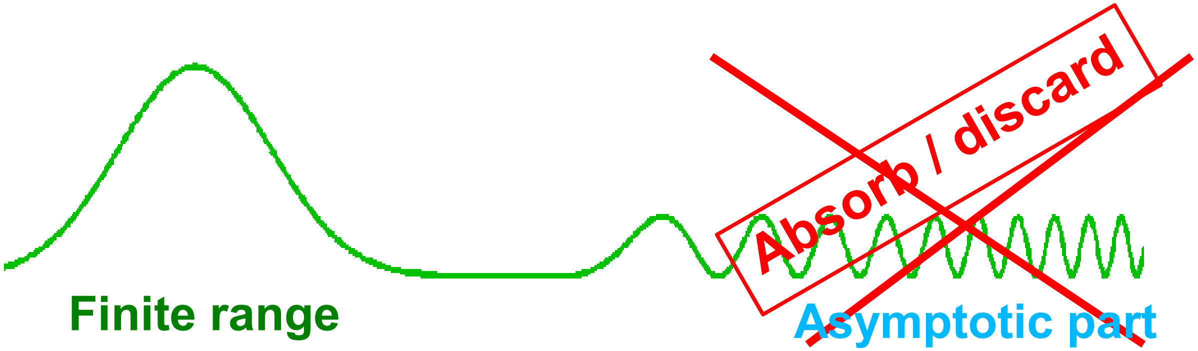
t-SURFF

Time-dependent surface flux method

[L. Tao and A.S., New. J. Phys. 14, 013021 (2012)]

Spectra from a finite range wave function

Scattering spectra = asymptotic information by definition



Problem:

If we solve only on a finite range, exactly the asymptotic information is missing

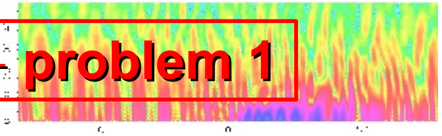
Solution

Continue beyond the box using some known solution - Volkov

[Caillat et al,, Rev. A 71 , 012712 (2005)]
[L. Tao and A.S., New. J. Phys. 14, 013021 (2012)]

How we usually calculate spectra from TDSE

Get Ψ(r,t) at the end of the pulse t=T: ***Ψ(r,T)***



← problem 1

Scattering solution ψ_k

$$H(T)|\psi_{\vec{k}}\rangle = \frac{\vec{k}^2}{2}|\psi_{\vec{k}}\rangle$$

← problem 2

With asymptotics

$$\psi_{\vec{k}}(\vec{r}) \sim (2\pi)^{-3/2}\exp(i\vec{k}\cdot\vec{r})$$

Spectrally analyze Ψ(x,T)

$$b(\vec{k}) = \langle\psi_{\vec{k}}|\Psi(T)\rangle$$

Spectral density

$$\boxed{\sigma(\vec{k}) \propto |b^2(\vec{k})|}$$

Solve problem 2: only use asymptotic states

Choose distance R_c where we (approximately) know the scattering solution

$$\psi_{\vec{k}}(\vec{r}) \sim (2\pi)^{-3/2} \exp(i\vec{k} \cdot \vec{r})$$

Propagate until large T where bound Ψ_b and scattering Ψ_s parts separate

$$\Psi(\vec{r}, T) = \Psi_{\mathrm{b}}(\vec{r}, T) + \Psi_{\mathrm{s}}(\vec{r}, T)$$

$$\begin{aligned} \langle \psi_k | \Psi_{\mathrm{s}}(T) \rangle &\approx \langle \psi_k | \theta(R_c) | \Psi_{\mathrm{s}}(T) \rangle \\ &= \langle \chi_k | \theta(R_c) | \Psi_{\mathrm{s}}(T) \rangle \end{aligned}$$

$$\theta(R_c) = \begin{cases} 0 & |\vec{r}| \leq R_c \\ 1 & |\vec{r}| > R_c \end{cases}$$

Only need χ_k
asymptotic scattering solution

$$\boxed{\sigma(\vec{k}) \propto \left| \langle \chi_{\vec{k}} | \theta(R_c) | \Psi(T) \rangle \right|^2}$$

Solves problem 2, but worsens problem 1: even larger *T* needed

Solve problems 1 & 2:

t-SURFF – time-dependent surface flux method

Same R_c as before

Asympotitic “comparison Hamiltonian” $H_c(t)$

$$H_c(t) = H(t) \quad \text{for } |\vec r| > R_c \quad \text{and} \quad \forall t$$

with known solutions χ_k $$i\frac{d}{dt}|\chi_{\vec k}(t)\rangle = H_c(t)|\chi_{\vec k}(t)\rangle$$

E.g. $$H_c(t) = \frac{1}{2}[-i\vec\nabla - \vec A(t)]^2$$ **with χ_k ...Volkov solutions**

Convert volume-integral → time-integral + surface-integral

$$\begin{aligned}\langle\chi_k(T)|\theta(R_c)|\Psi_s(T)\rangle &= \int_0^T dt\frac{d}{dt}\langle\chi_k(t)|\theta(R_c)|\Psi_s(t)\rangle\\ &= i\int_0^T dt\langle\chi_k(t)|H_c(t)\theta(R_c)-\theta(R_c)H(t)|\Psi_s(t)\rangle\\ &= i\int_0^T dt\langle\chi_k(t)|\left[-\frac{1}{2}\Delta + i\vec A(t)\cdot\vec\nabla, \theta(R_c)\right]|\Psi_s(t)\rangle\end{aligned}$$

TDSE for χ_c and Ψ_s

$H_c \equiv H$ for r > R_c

Commutator depends only on $\Psi(R_c,t)$!!!

Commutator in terms of the angular basis

$$
\begin{aligned}
&4\pi^{-1} e^{-i\Phi(\vec{k},t)} R_c^{-2}\langle\vec{k}|[H_c(t),\theta(R_c)]|\Psi(t)\rangle \\
&\quad= \sum_{lmn}(-i)^l Y_{lm}(\hat{k})\langle Y_{lm}|Z_{ln}\rangle(\partial_r j_l c_{ln} - j_l d_{ln}) \\
&\qquad +2A_z \Big[(-i)^l Y_{l+1m}(\hat{k})\langle Y_{l+1,m} j_{l+1}|\cos\theta|Z_{l,n}\rangle c_{ln} \\
&\qquad\qquad - (-i)^l Y_{l-1m}(\hat{k})\langle Y_{l-1,m} j_{l-1}|\cos\theta|Z_{l,n}\rangle c_{ln}\Big]
\end{aligned}
$$

$\Phi(\mathbf{k},t)$..Volkov phase
Y_{lm} ...spherical harmonics
Z_{ln} ...angular expansion of Ψ
j_l ...spherical Bessel functions at $|\mathbf{r}|=R_c$
c_{ln} ...radial expansion of Ψ at $|\mathbf{r}|=R_c$
d_{ln} ...radial expansion of $\partial_r\Psi$ at $|\mathbf{r}|=R_c$

$$
= \sum_{ln} B_{ln}(\hat{k})c_{ln}(t) + C_{ln}(\hat{k})d_{ln}(t) + A(t)D_{ln}(\hat{k})c_{ln}(t)
$$

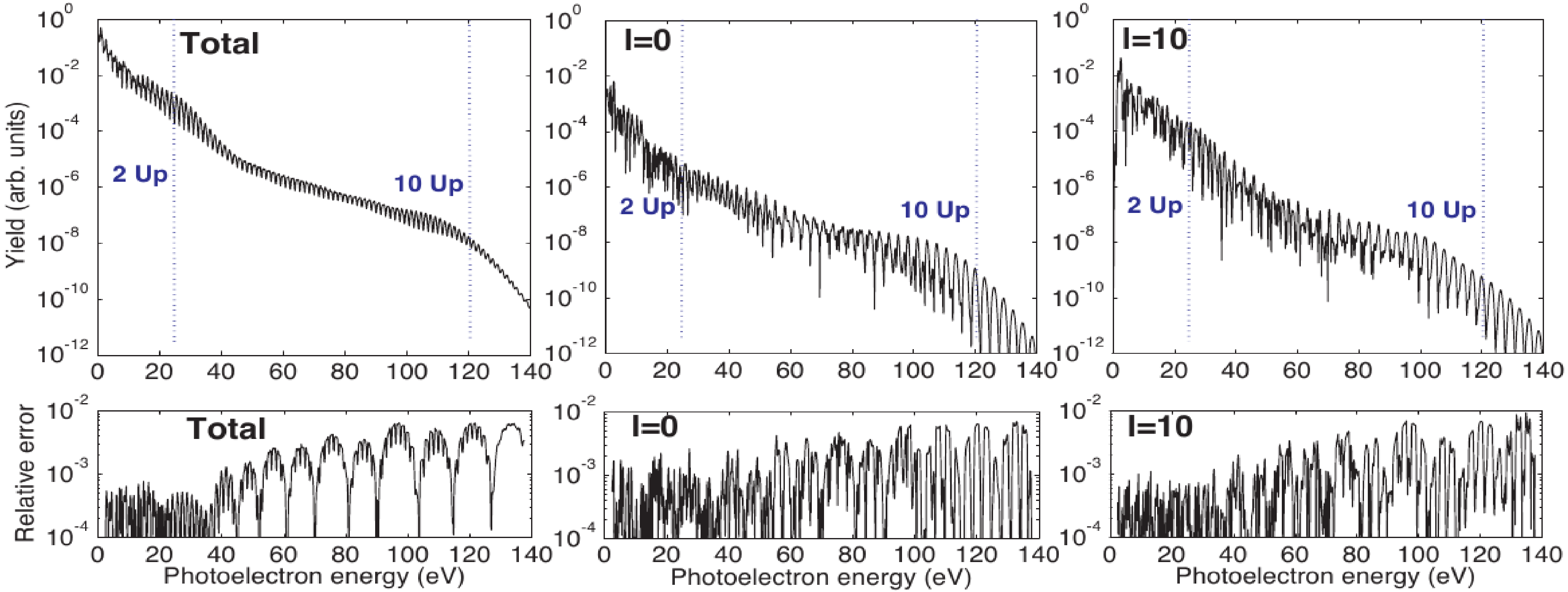
Note: B_{ln}, C_{ln}, D_{ln} are time-independent

Photo-electron spectra – short range potential

Truncated Coulomb potential

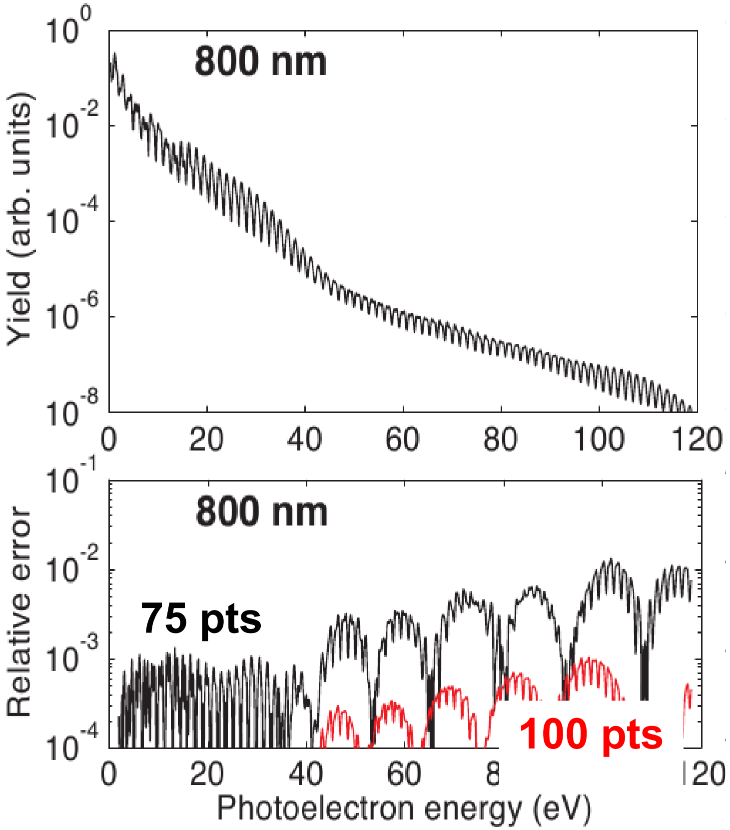
$$V(r)=\begin{cases} c\left[-1/r-r^2/(2R)^3+3/(2R)\right] & \text{for } r<R \\ 0 & \text{for } r>R. \end{cases}$$

10 optical cyles, 800 nm, 2 x 10^{14} W/cm^2



Errors for 90 discretization points relative to a well-converged calculation

Total box must accommodate quiver radius



Quiver radius ~ 23 a.u.

Potential range R= R_0 = 15
Box size ~ 25 a.u.

Radial points 75 (black line)

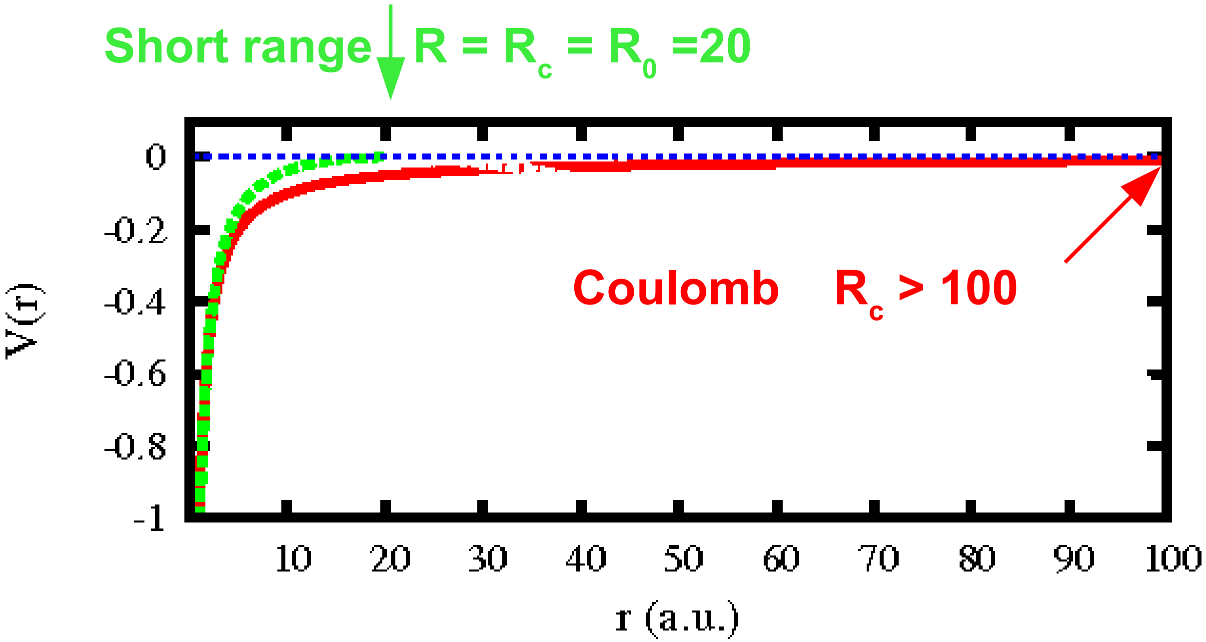
100 (red line)

Hydrogen atom

Infinite range of the Coulomb potential

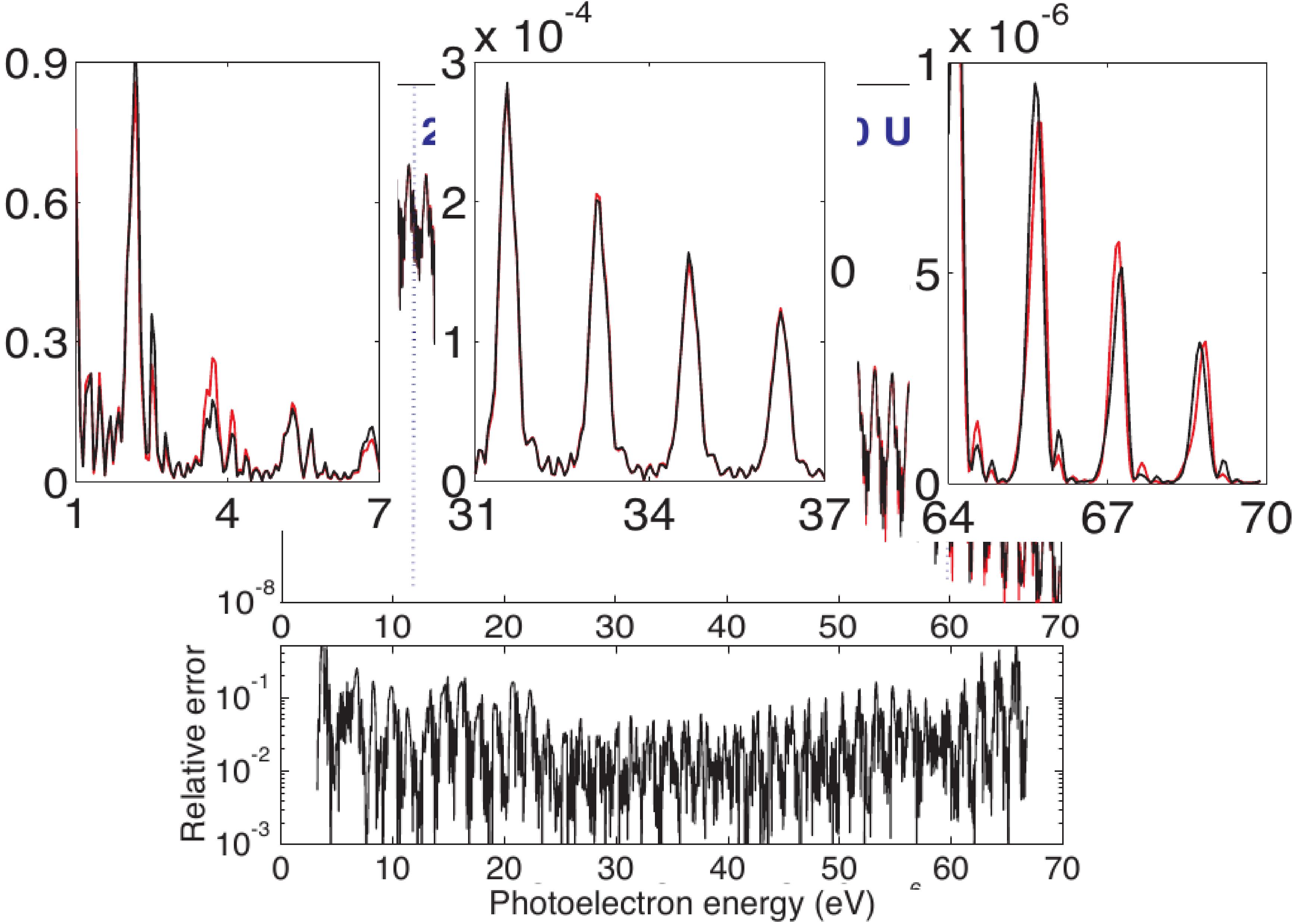
→ Volkov solutions never become exact

Choose larger R_c



Note: R_c can be reduced with significant extra effort – maybe useful for multi-electron

Spectra: 800nm, 10^{14}W/cm^2, 40 opt. cycl.



Photoelectron spectra

If you like glossy pictures...

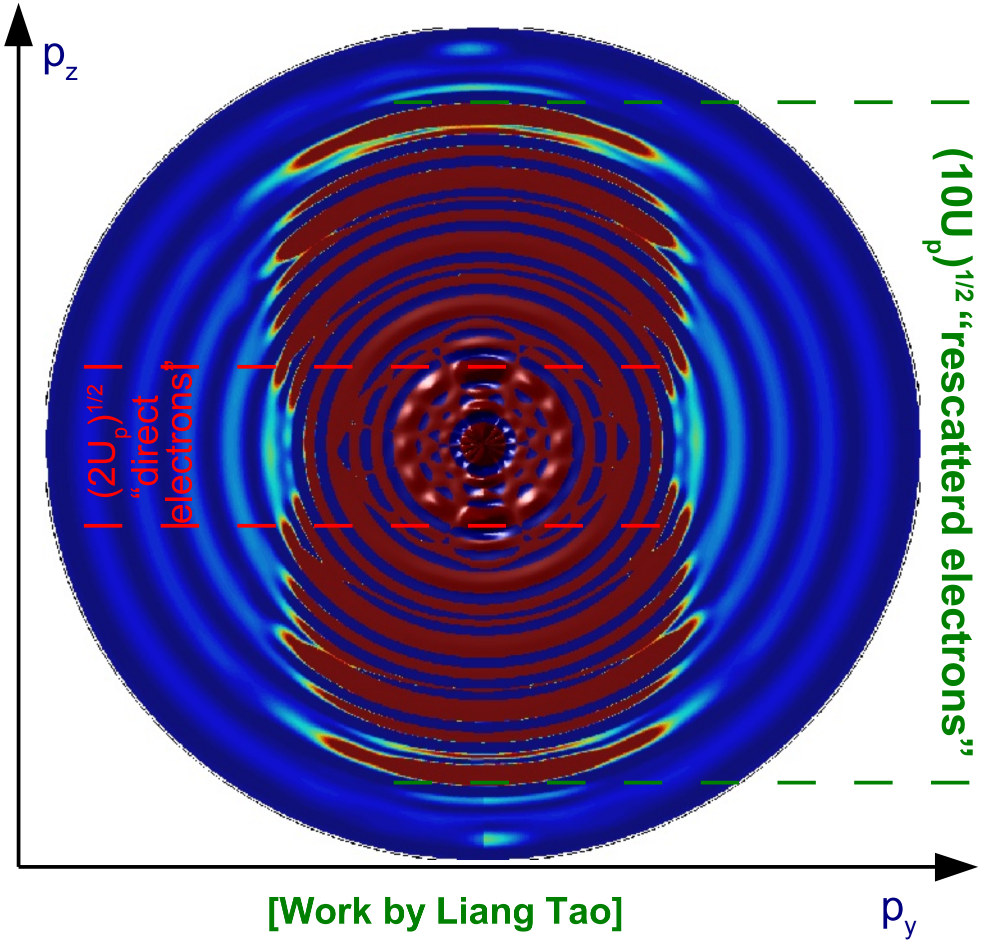
Photo-electron momentum distribution
20 optical cycles FWHM
800 nm @ 2 x 10^{14} W/cm^2

Fastest electrons move to ~ 4000 au!

Simulation box size 30 au !

- All visible structures are accurate to ~ 1% in density (mostly much better)
- Dynamical range is ~ 10^8

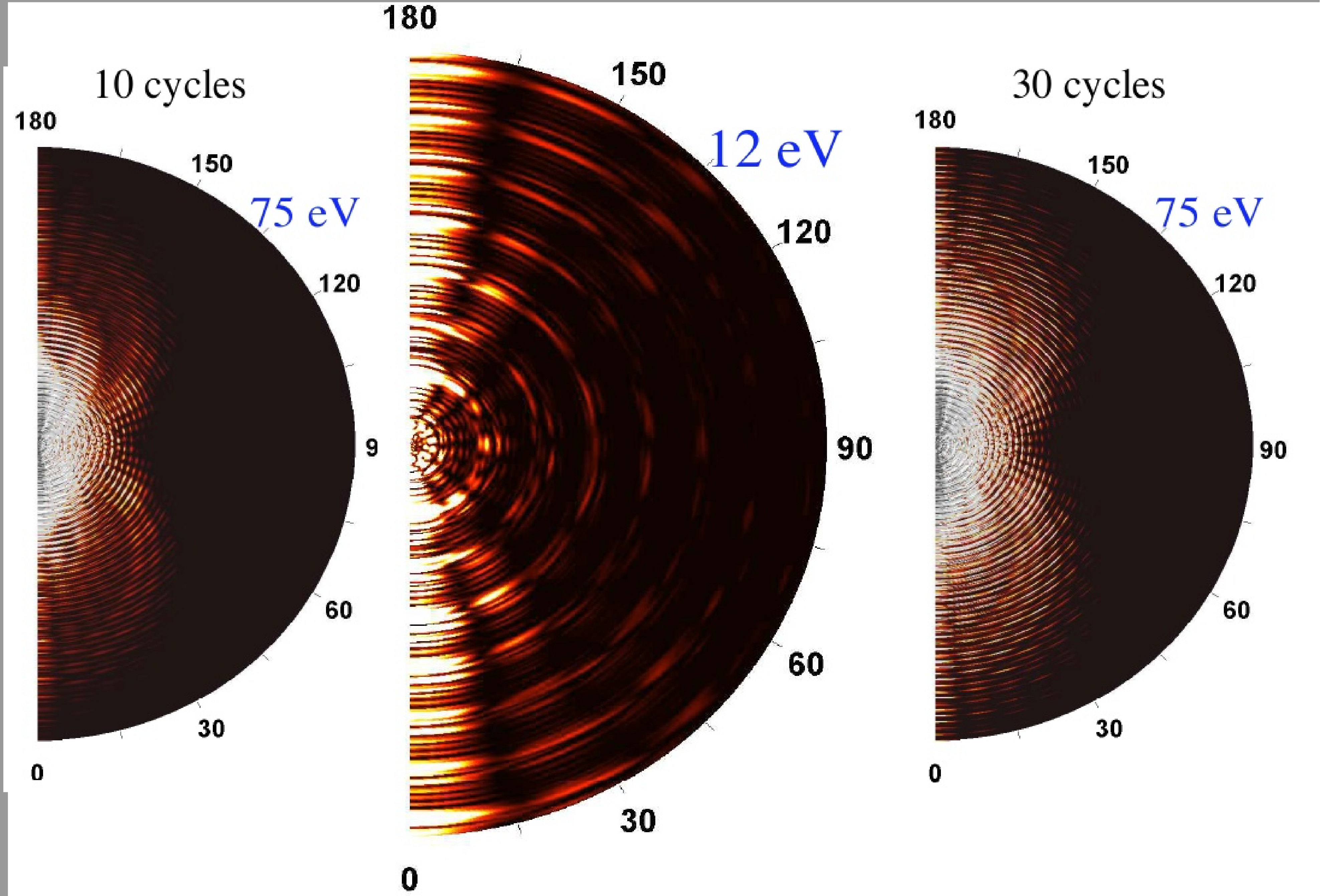
Photo-electron momentum density



[Work by Liang Tao]

Note: Coulomb causes trouble, requires larger boxes ~ 100 au

Angle-resolved spectra



General polarization

Problem grows from 2 to 3 dimensions

But only for |r| < R_c!

For I = 2 x 10^{14} W/cm^2 @ 800 nm

Needs l_{max}=30, i.e. ~ 900 angular functions and ~ 90 radial functions
→ Workstation size problem

Additional term in the commutator:

$$= \sum_{ln} B_{ln}(\hat{k})c_{ln}(t) + C_{ln}(\hat{k})d_{ln}(t) + [A_z(t)D_{ln}(\hat{k}) + \boxed{A_y(t)E_{ln}(\hat{k})}]\, c_{ln}(t)$$

In the process of implementation...

Two-electron systems

Multi-channel single ionization

Shake-up

[A. S., submitted to New. J. Phys., arXiv:1201.3590v1]

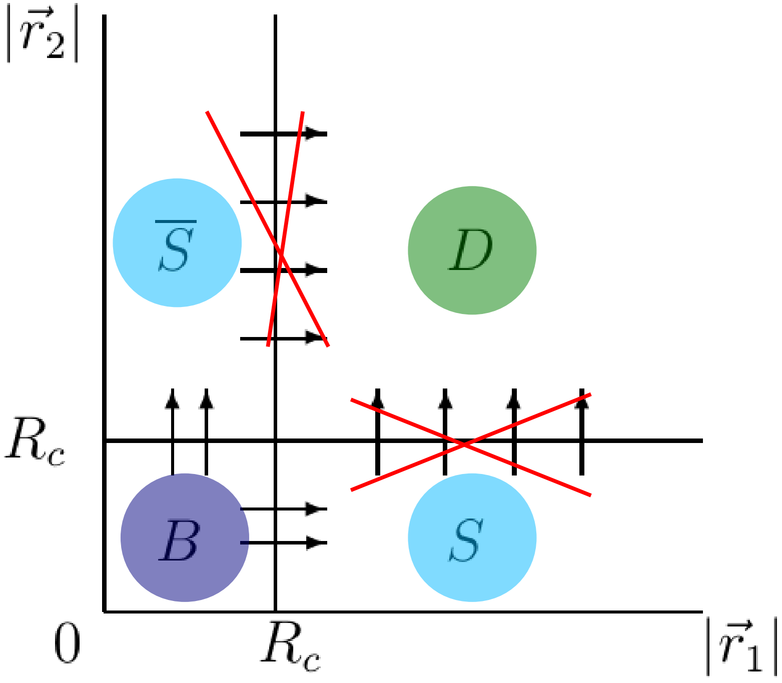
Defining surfaces...

Split two-electron coordinate space

B... $|\mathbf{r}_1|,|\mathbf{r}_2| < R_c$ “bound” region
Numerical solutions on $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$

S... $|\mathbf{r}_2| < R_c$, $|\mathbf{r}_2| > R_c$ “singly asympotic” region
Numerical ionic solution on $\boldsymbol{r}_2$: $\Phi_c(\boldsymbol{r}_2,t)$
Volkov solution on $\boldsymbol{r}_1$

D... $|\mathbf{r}_1|,|\mathbf{r}_2| > R_c$ “doubly asymptotic” region
Volkov solutions on $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$



Simplest case: neglect double ionization

Channel solution

$$\chi_{c,\vec{k}}(t) = (2\pi)^{-3/2} e^{-i\Phi(t)} e^{i\vec{k}\vec{r}} \otimes \phi_c(t)$$

Ionic TDSE

$$i\frac{d}{dt}\phi_c(t) = H_{ion}(t)\phi_c(t)$$

Multi-channel case

E.g. different ionic states channels

Play the same trick as before
“Channel asymptotic functions” $\chi_{c,k}$

Channel spectral density $\sigma_c(\vec{k}) = |\langle \chi_{c,\vec{k}}(T)|\theta(R_c)|\Psi_{\mathrm{s}}(T)\rangle|^2$

Channel asymptotic TDSE $i\frac{d}{dt}\chi_{c,\vec{k}}(t) = H_c(t)\chi_{c,\vec{k}}(t)$

Channel Hamiltonian $H_c(t) = \frac{1}{2}[-i\vec{\nabla} - \vec{A}(t)]^2 \otimes H_{ion}(t)$

… further the same reasoning as before

Numerical demonstration: 2 x 1d model

$$H(t) = \sum_{\alpha=1,2} \frac{1}{2}\left[-i\frac{\partial}{\partial x_\alpha} - A(t)\right]^2 - \frac{2M(x_\alpha)}{\sqrt{x_\alpha^2 + 1/2}} + \frac{M(x_1)M(x_2)}{\sqrt{(x_1 - x_2)^2 + 0.3}}$$

M(x) … mask function turning off all potentials for $|\boldsymbol{r}_\alpha| > C_p$

→ Volkov solution exact beyond C_p

Neutral energy: -2.88 a.u.

Ion energy: -2.0 a.u.

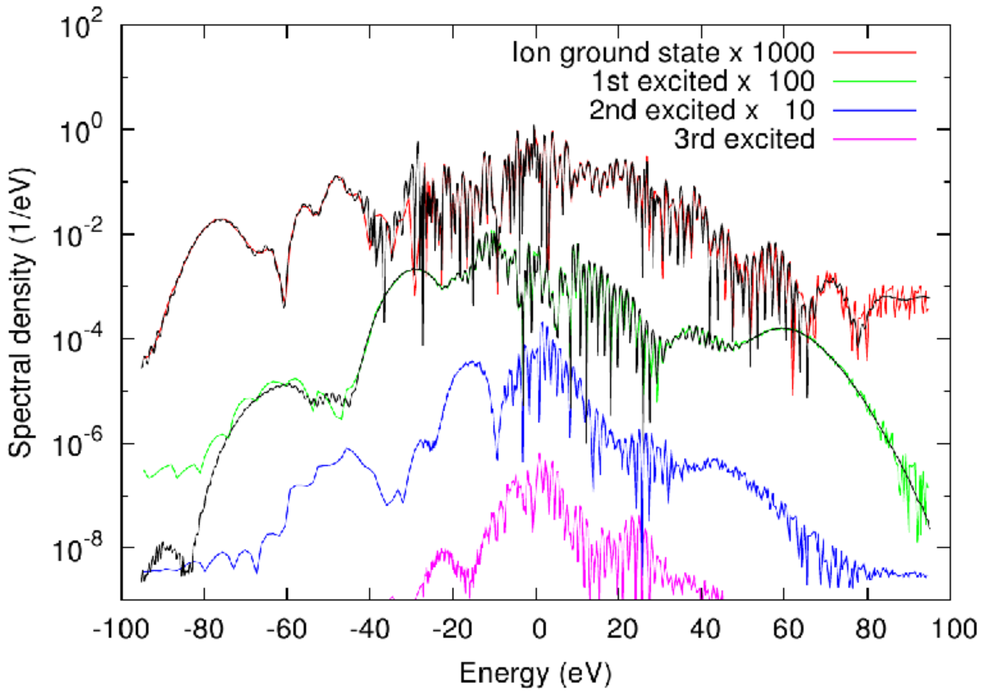
"helium"

Shake-up photo-electron spectra

Single cycle, 2 x 10^{14}W/cm^2 @800 nm

Black lines:
Channel method,
double ionization neglected

Color:
including double ionization
(discussed below)



Two-electron systems

Double ionization

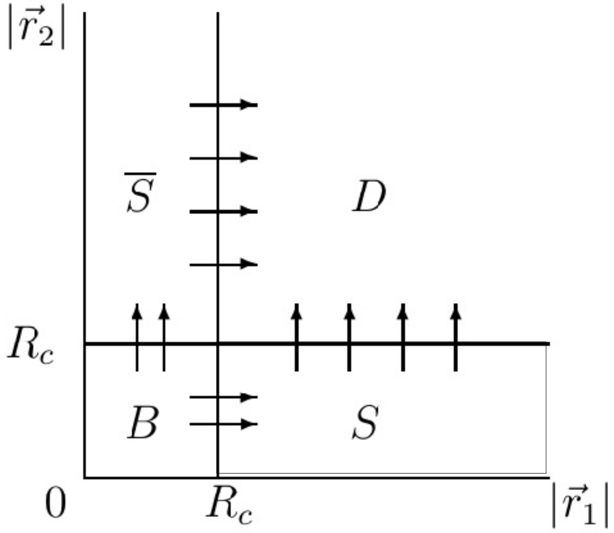
[A. S., submitted to New. J. Phys., arXiv:1201.3590v1]

Extending t-SURFF to double-ionization

Do not neglect flux S → D

For Ψ and $\partial_r\Psi$ on interface S|D
on area S
expand into Volkov $\chi_k(r_1,t)$ x basis $\xi_n(r_2,t)$

$$\Psi(\vec{r}_1,\vec{r}_2,t) = \int d^3k \sum_n \chi_{\vec{k}}(\vec{r}_1,t)\xi_n(\vec{r}_2) b'(\vec{k},n)$$



Equation for the expansion coefficients $b(k_1,n,t)$

$$i\frac{d}{dt}b(\vec{k}_1,n,t) = \sum_m \langle\xi_n|H_{ion}(t)|\xi_m\rangle b(\vec{k}_1,m,t)$$ …ionic time-evolution

$$-\langle\vec{k}_1,t|[H_v(t),\theta_1]\langle\xi_n|\Psi(t)\rangle\rangle$$ …flux B → S

For each $\boldsymbol{k}_1$, solve one ionic problem in $|\boldsymbol{r}_2|<R_c$!

Integrate flux S → D by t-SURFF

Spectrum of $\boldsymbol{k}_2$ for each $\boldsymbol{k}_1$

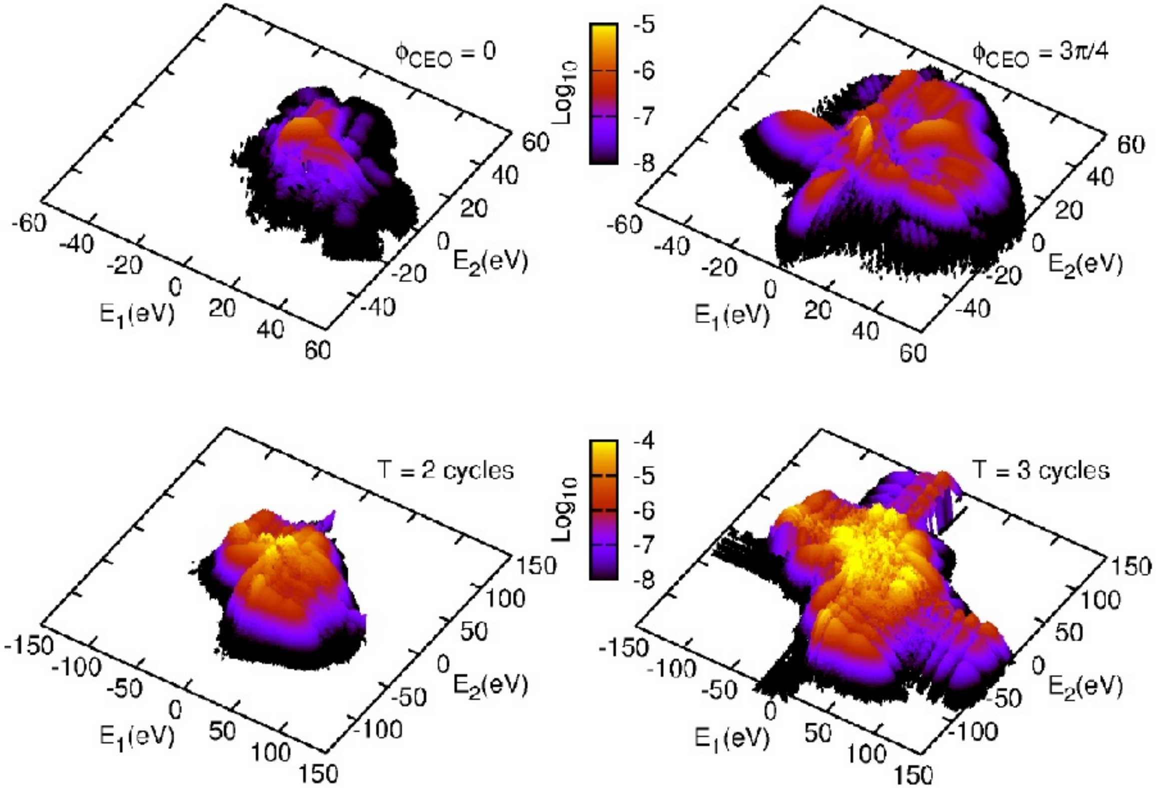
Numerical demonstration for 2 x 1d model

$$H(t) = \sum_{\alpha=1,2} \frac{1}{2}\left[-i\frac{\partial}{\partial x_\alpha} - A(t)\right]^2 - \frac{2M(x_\alpha)}{\sqrt{x_\alpha^2 + 1/2}} + \frac{M(x_1)M(x_2)}{\sqrt{(x_1 - x_2)^2 + 0.3}}$$

$\cos^2$ – pulse @ 800nm, I=2 x 10^{14} W/cm^2, FWHM = 1,2,3 cycles

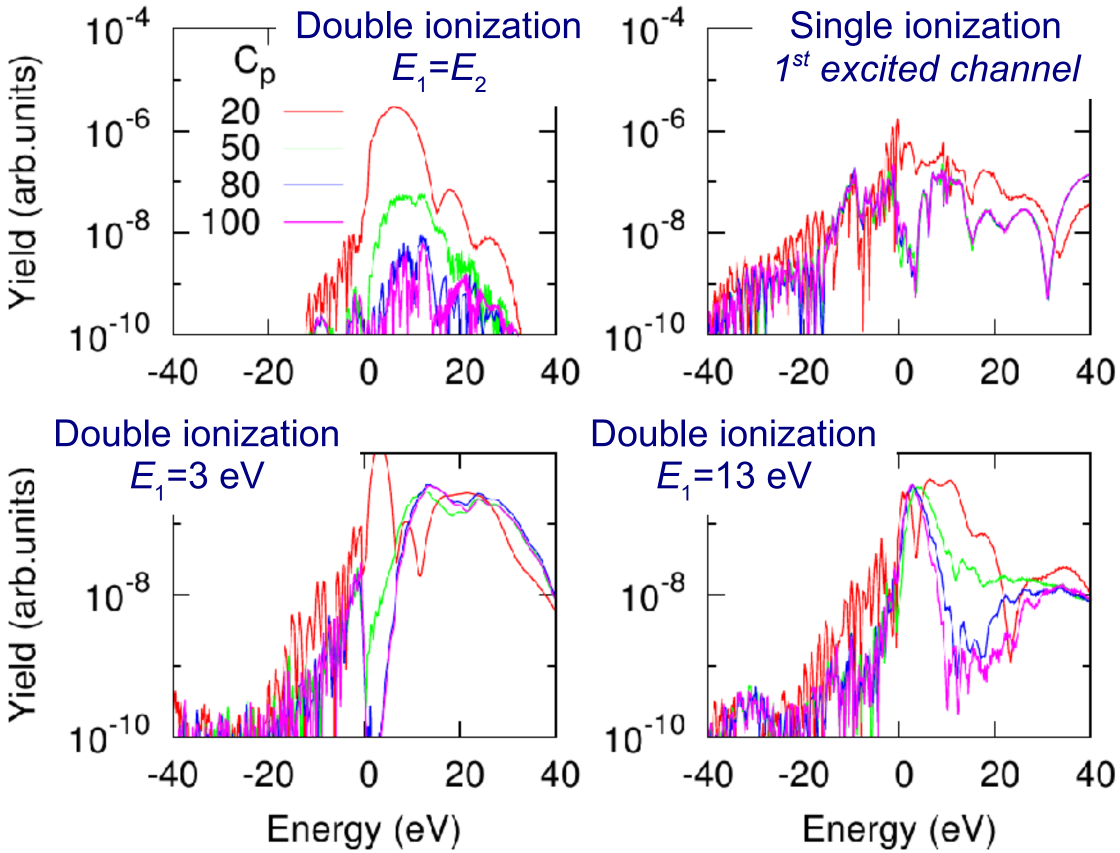
Discretization size

49 points on [0,∞)
total of 97 x 97 points



Coulomb potential: importance of long-range

M(x) restricts the potential to $|r_\alpha| < C_p$



Satisfactory approximation at $C_p \approx 80$ a.u.

Scaling to 2 x 3 dimensions

In 2 x 1d two-electron calculations

→ 2 directions -x and +x
→ 50 linear coefficients for x in [0,∞)

Discretization size: $2 \times 2 \times 50 \times 50 = 10^4$
Single Intel i5 core @ 2.6GHz ~ 2 CPU hours per pulse

From 3d single electron calculations

Discretization at laser parameters 800nm, I=2×10^{14} W/cm^2

→ $L_{max} \approx 30$ angular momenta
→ 100 ~ 200 linear coefficients for r in [0,∞) (Coulomb potential!)

Discretization size: $30 \times 30 \times 30 \times 100 \times 100 \approx 3 \times 10^8$

Resource estimate for accurate fully differential 2-electron spectra

6 x 10^4 CPU hours

~ 2 days on 1000 CPUs
Not small... but rather high accuracy standards

Summary & outlook

Prospects are that
t-SURFF completely solves IR single and double ionization

- single-electron spectra in linear polarization

- single-electron spectra in general polarization (in preparation)

- shake-up and double ionization in 2x1d (demonstrated)

- 3d double-ionization @ 800 nm: large but feasible problem

~ Thanks ~

[A.S., Phys. Rev. A81, 53845 (2010)]

[L. Tao and A.S., New. J. Phys. 14, 013021 (2012)]

[A. S., submitted to New. J. Phys., arXiv:1201.3590v1]