A parallel MCTDHF code for multi-electron systems in strong fields

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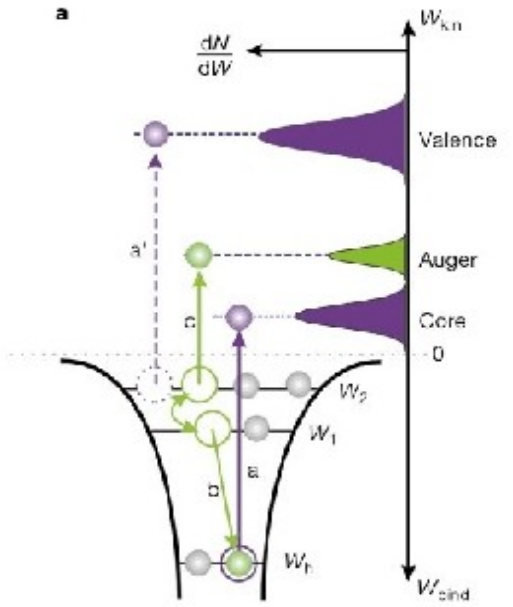
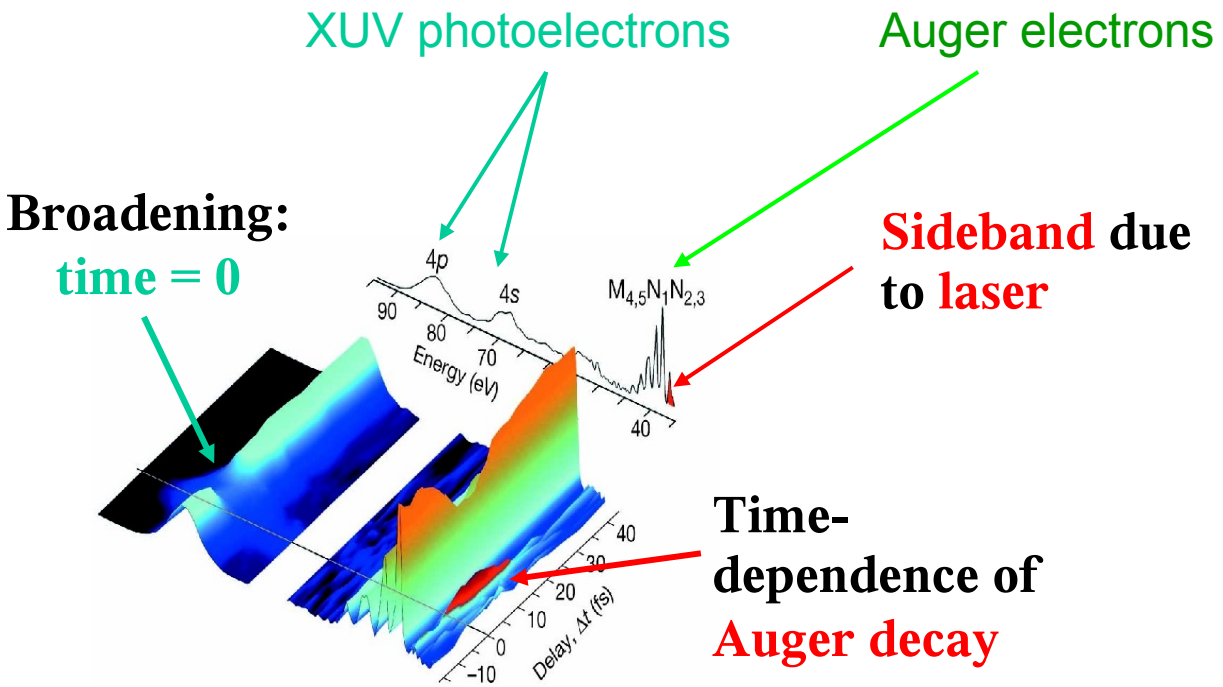
Motivation:

Attosecond pump-probe: Auger decay

Scheme of measurment



Core-hole formation
by attosecond XUV
Probe electron emission
by few-cycle laser



[Drescher et al., Nature (2002)]

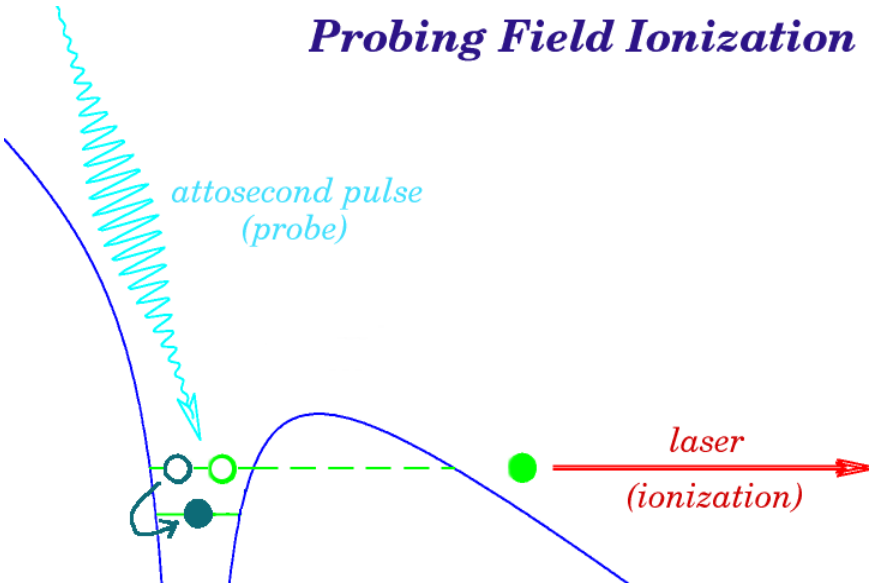
Motivation:

Attosecond pump-probe: ionization dynamics

Watch an atom
while it is being ionized

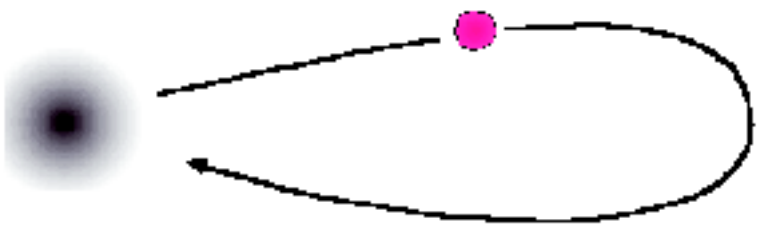
a strong laser ionizes
during a single field cycle
(2.6 fs @ 800 nm)

- depletion of neutral
- appearance of ion
- intermediate states ?

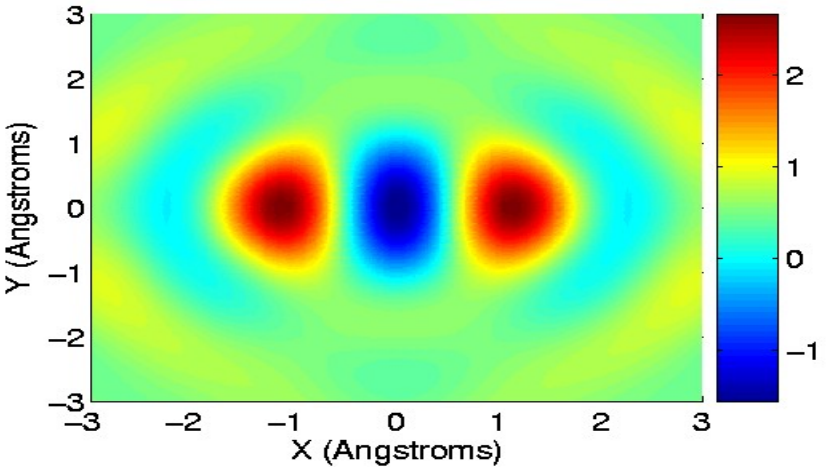


Motivation:

Rescattering imaging of a molecular orbital



(1) Laser detaches electron from molecule
(2) The electron is directed back by the laser
(3) Scattering produces harmonics
(4) Harmonics contain a
tomographic image of the HOMO



Measured image of the HOMO of N_2
[Itatani et al., Nature, (2004)]

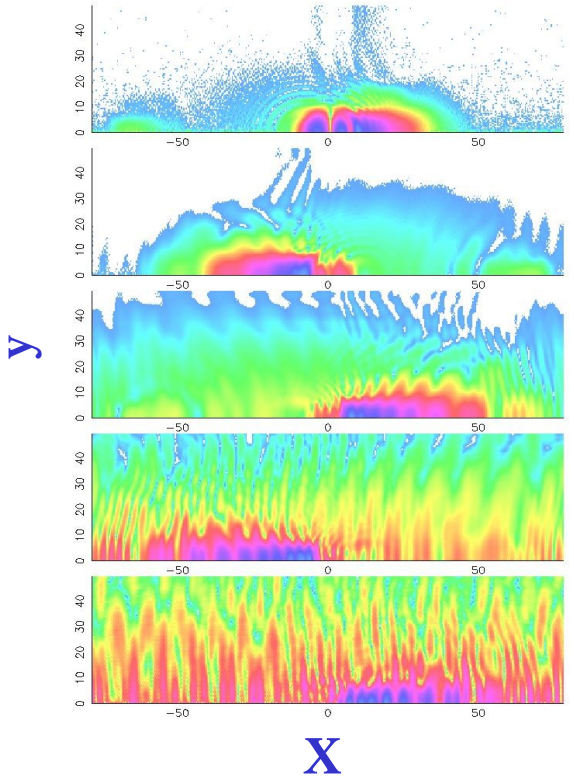
How exactly does the electron come back ?
What does one actually measure ?

Strong fields:

Hydrogen electron density during two laser cycles

4×10^{14} W/cm^2
5 fs FWHM
(simulation)

Total explosion during a few femtoseconds



Time (femtoseconds)

-2.6

-1.3

0

1.3

2.6

Key characteristics of the systems

Laser electric field ~ atomic field strength
=> highly non-perturbative
=> large simulation volumes

Short time scales: 100 attoseconds ~ electron orbit time
=> non-stationary, wave-packet like situation

Several electrons are involved:
-- Auger process
-- strong field ionization
-- rescattering
-- molecules

Both, continuous and bound, parts of the system
=> both, quantum and near classical, behavior

Hamiltonian

$$H(t) = \sum_{l=1}^{f} \frac{1}{2} \left[\frac{1}{\mathrm{i}} \vec{\nabla}_l - e\vec{A}(t) \right]^2 + V_n(\vec{r}_l) + \sum_{k=l+1}^{f} \frac{1}{|\vec{r}_l - \vec{r}_k|}$$

V_n ... nuclear potential: Coulomb or model

A(t) ... laser vector potential,
velocity gauge is <u>better</u> in very strong fields

<u>Observables</u>

High harmonic radiation

Ionization yields

Electron spectra

Our implementation of MCTDHF:

Basics: MCTDHF = MCTDH + F

$$\Psi(\vec{x}_1,\ldots,\vec{x}_f;t) = \sum_{j_1=1}^{n}\cdots\sum_{j_f=1}^{n} A_{j_1\ldots j_f}(t)\,\varphi_{j_1}(\vec{x}_1;t)\ldots\varphi_{j_f}(\vec{x}_{f;t})$$

All differences to MCTDH are "technical" (but important)

- an orbital carries all single particle properties
  3 spatial + 1 spin coordinates ("3d mode combination")
- $A_{j1\ldots jf}$ are strictly anti-symmetric with resp. to their indices
  many fewer (independent) A's (<1000) than in MCTDH
- there are only two-particle interactions
- use Slater rules for the calculation of mean fields etc.
- choose between restricted and unrestricted orbitals

New code developed from scratch

Our implementation of MCTDHF:
Typical numbers

Box sizes:

200 atomic units in laser polarization direction

20 atomic units perpendicular

(absorbing boundaries)

Spatial grid points: 10^5 ~ 10^6

Number of particles: 2 – 8

Strict cylindrical symmetry (to be extended to full 3d)

Run times: hours (but on a parallel computer)

Memory: ~ 500 MB (can be seriously improved)

Our implementation of MCTDHF:

Discretization and related stuff

Spatial discretization:

- Finite elements on cylinder coordinates (ρ,z)
- Product grid ~ 1000 x 100
- FFT method on z (only on single-processor)

Integrations:

transformation to quadrature grid

Time-integration:

- Runge-Kutta self-adaptive time-step and order up to 6
- CMF (“Constant Mean Field”):

  more function calls for given accuracies (!?)

Our implementation of MCTDHF:

Discretization and related stuff (cont'd)

Two-particle potential - low rank approximation:

$$\frac{1}{|\vec{r}_1 - \vec{r}_2|} \approx \sum_{m=1}^{M} U_m(\vec{r}_1) U_m(\vec{r}_2)$$

M ~ 200

Schmidt-decomposition (present)
H-matrix techniques (planned, good for parallel code !)

Initial state calculation: imaginary time propagation
(to be improved)

More technical details: Caillat et al., Phys. Rev. A (2005)

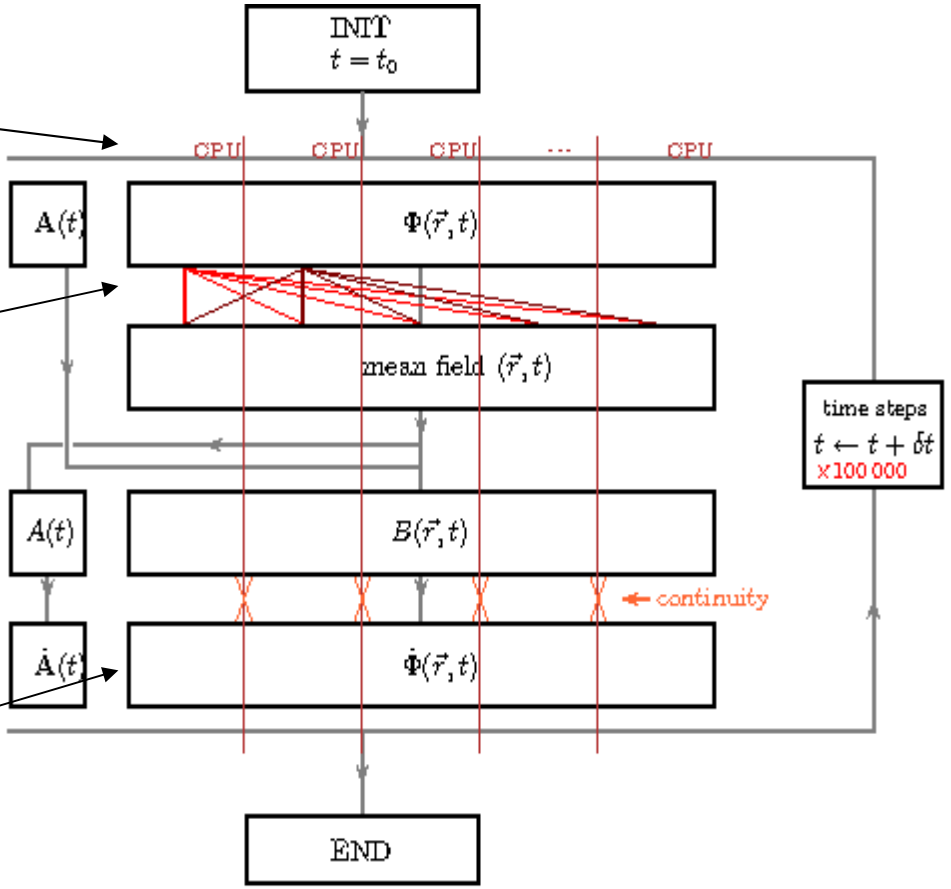
Parallelization

Scatter orbitals over cpu's

Calculation of mean fields:
non-local interaction reduced
by low-rank approximation

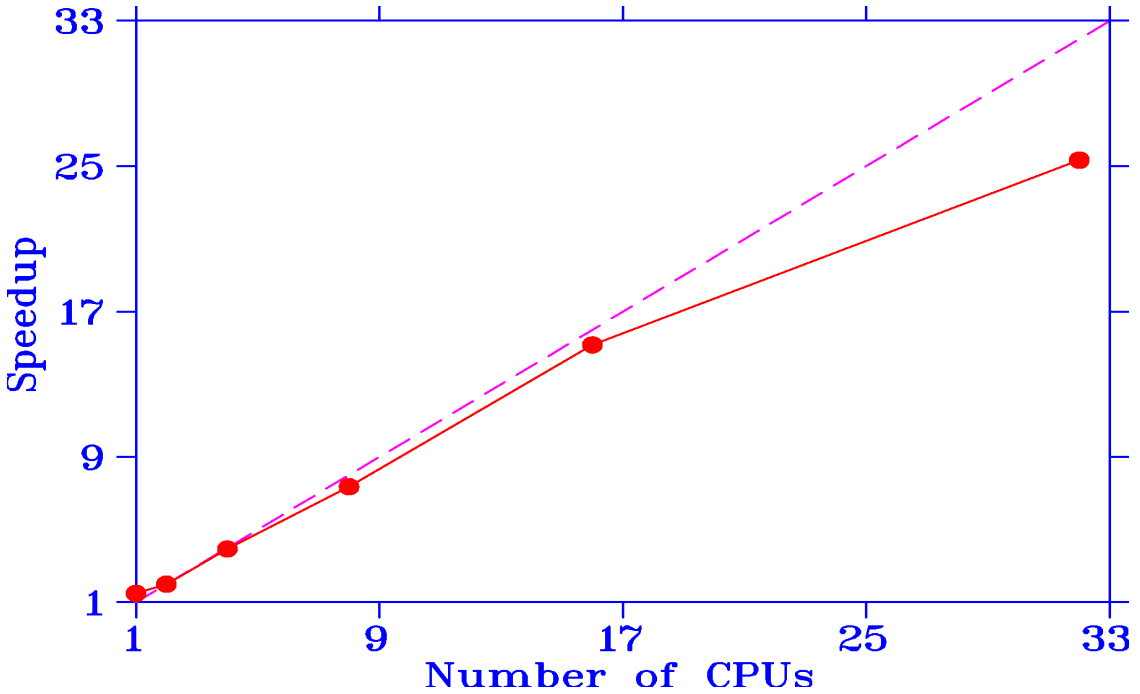
Application of mean fields:
strictly local

Differential operators have
negligible communication



Nearly linear scaling up to 32 CPUs (and beyond ?)

Scaling of the parallel code



Deviations from linear scaling mostly due to scalar calculation of d/dt A_J

NOTE: speedup is given relative to the 2-CPU calculation as the scalar code also partially uses 2 CPUs

NOTE: loss at 32 CPU not understood, maybe hardware ?

Checks

He and H_2 ground state energies

Helium ground state

(restricted MCHF)

n, f	energy
2,2	-2.8589
4,2	-2.8751
6,2	-2.8819
8,2	-2.8827
(exact	-2.9037)

H_2 energy at R=1.4

(restricted MCHF)

n, f	energy
2, 2	-1.8466
4, 2	-1.8652
6, 2	-1.8725
8, 2	-1.8732
(exact	-1.8887)

Acceptable accuracies
Difference to exact due to single-electron discretization (?)

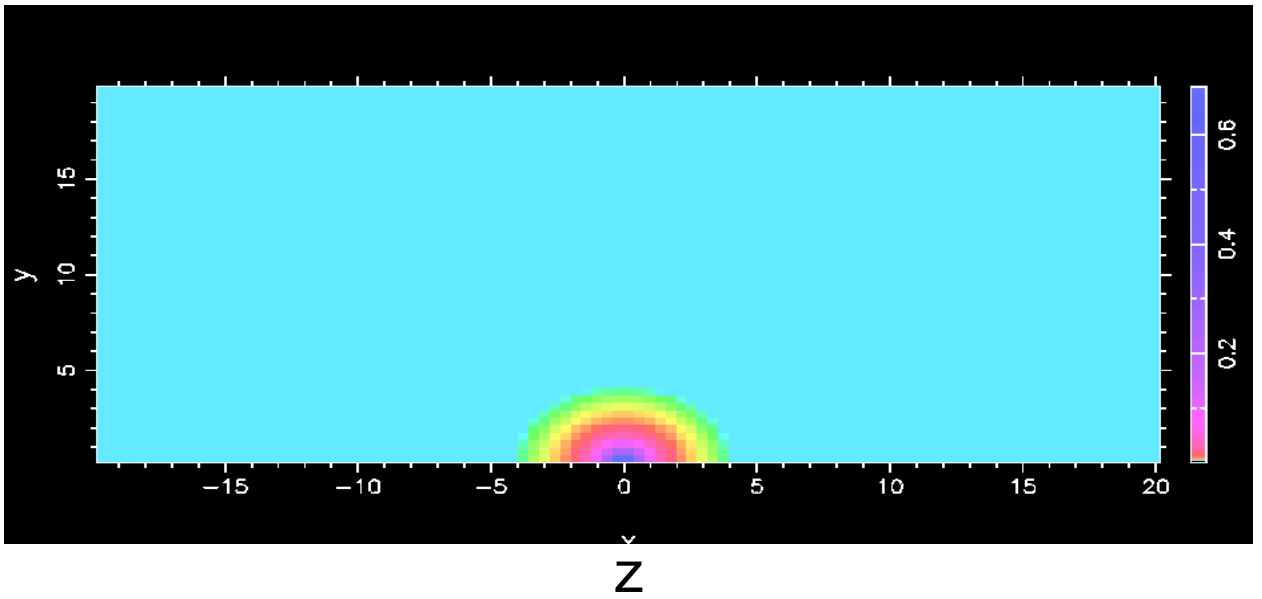
A two electron model of “Argon”

Ionization potential 0.57 a.u., second ionization pot. 1.2 a.u.
two active electrons

Laser
single-cycle laser pulse a 800 nm, peak intensity 3×10^{14} W/cm^2 (~ field 0.1 a.u.) [~ experimental parameters]

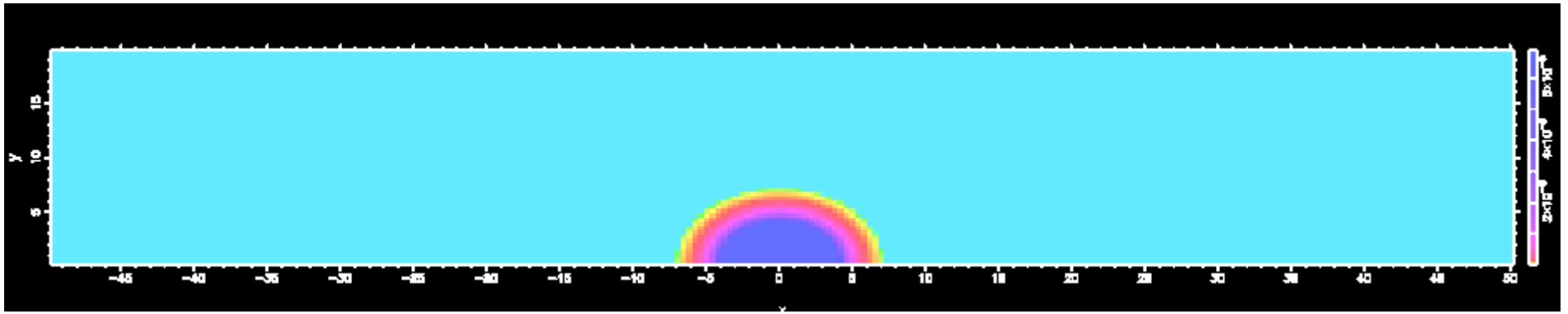
MOVIE

Electron density as ρ a function of time
range $1 - 10^{-4}$



Electron density of Ar below 10^{-4}

MOVIE: same as before, but range 10^{-4} – 10^{-7}



Our effect is a very small effect on top of a large effect

Need high accuracies !

(Is that why high order Runge-Kutta wins ?)

Calculation for "N_2"

- Two nuclei at separation of 2 a.u., same ionization potential as Ar, two active electrons, same laser parameters as before
- Similar picture, somewhat more ionization...

Does an electron tunnel ionize
from N_2 in the same way as from Ar ?

Are "all tunnels alike" ?

Are all tunnels alike ?

Key hypotheses of the molecular imaging experiment:

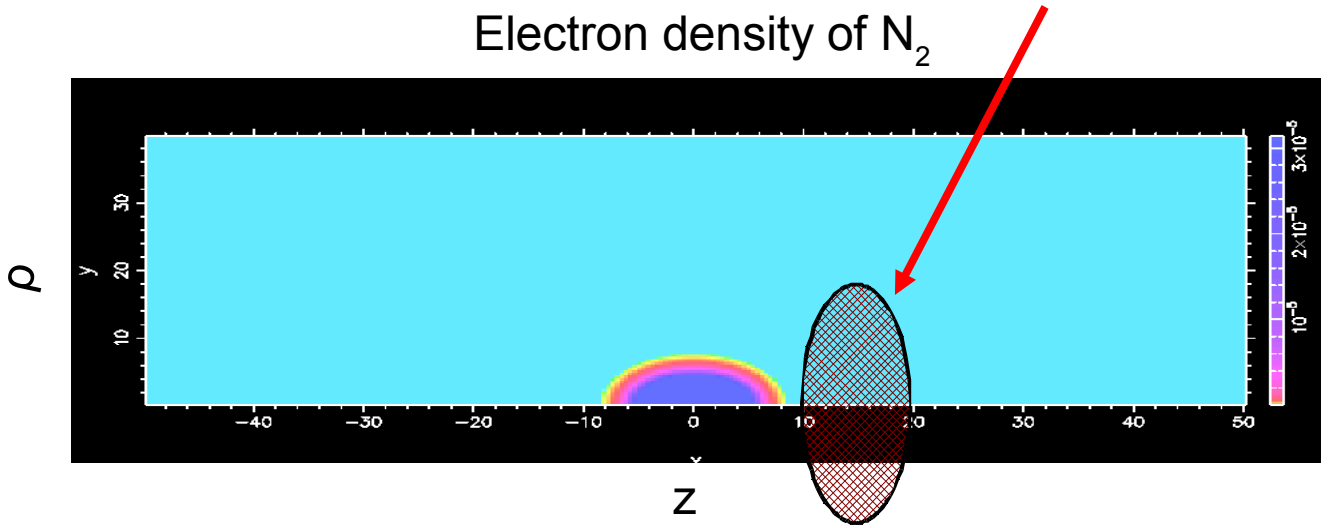
all electrons tunnel in the same way

depend only on the ionization potential

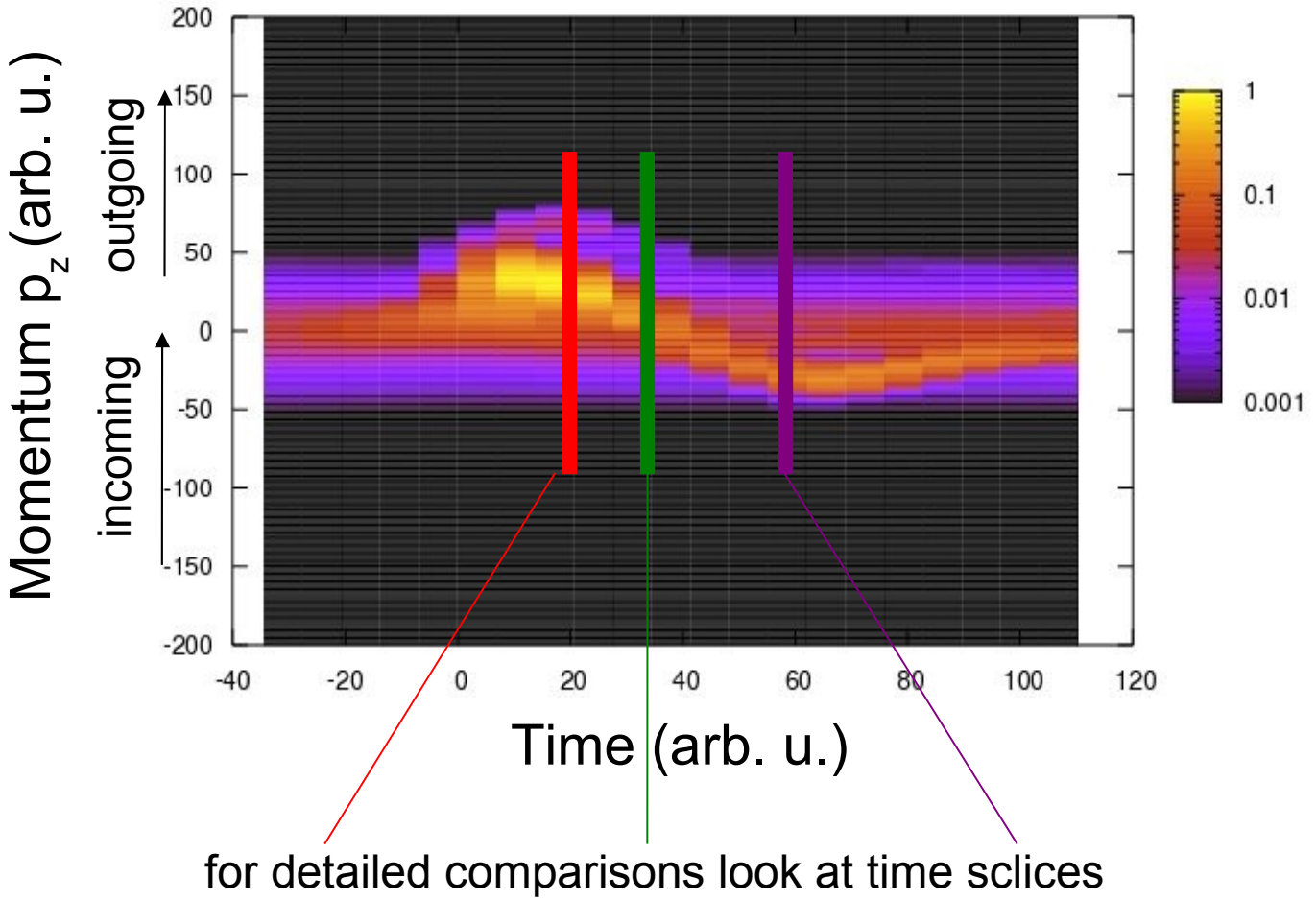
Put a “probe” into the electron flux

some 15 a.u. away from the system

“Measure” the electrons passing through a barrier



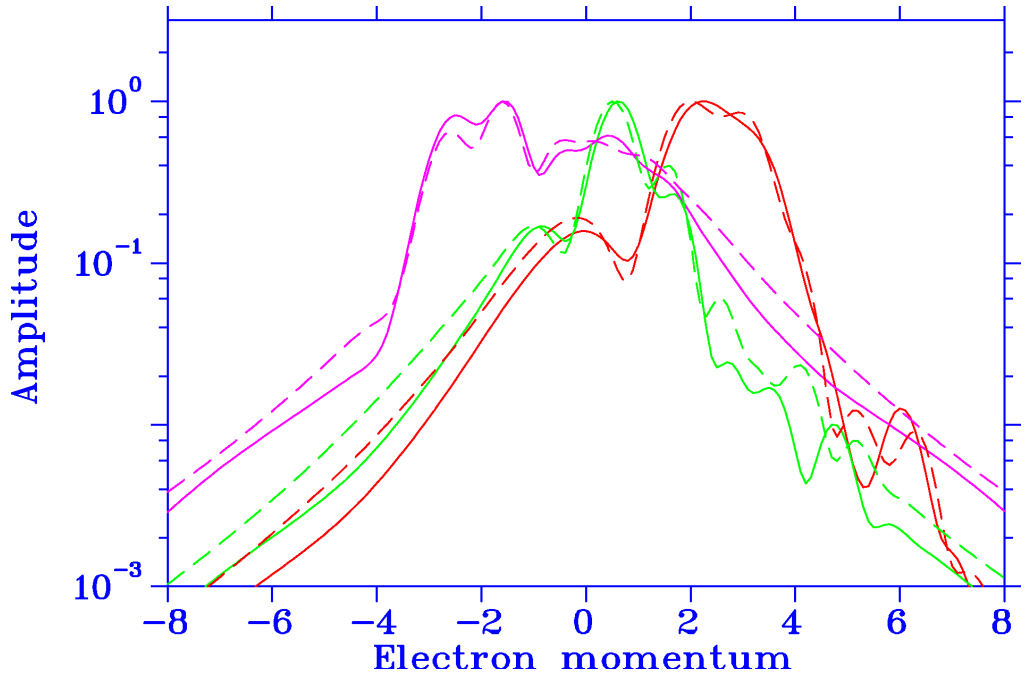
Rescattering p_z spectra as a function of time



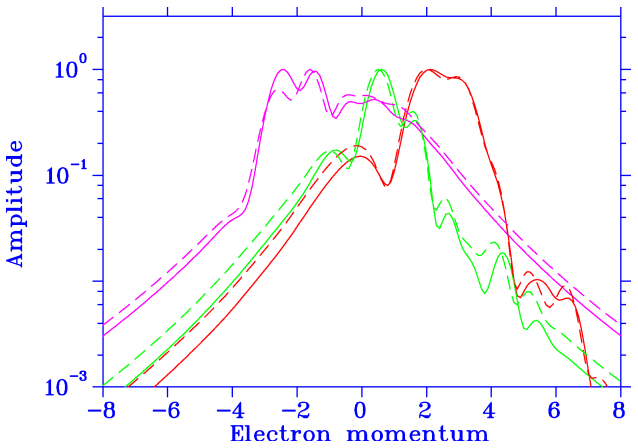
Are all tunnels alike ?

Electron momenta through the barrier

"Ar" vs. "N_2":
three different time slices



Importance of correlation:
4 vs. 8 orbitals



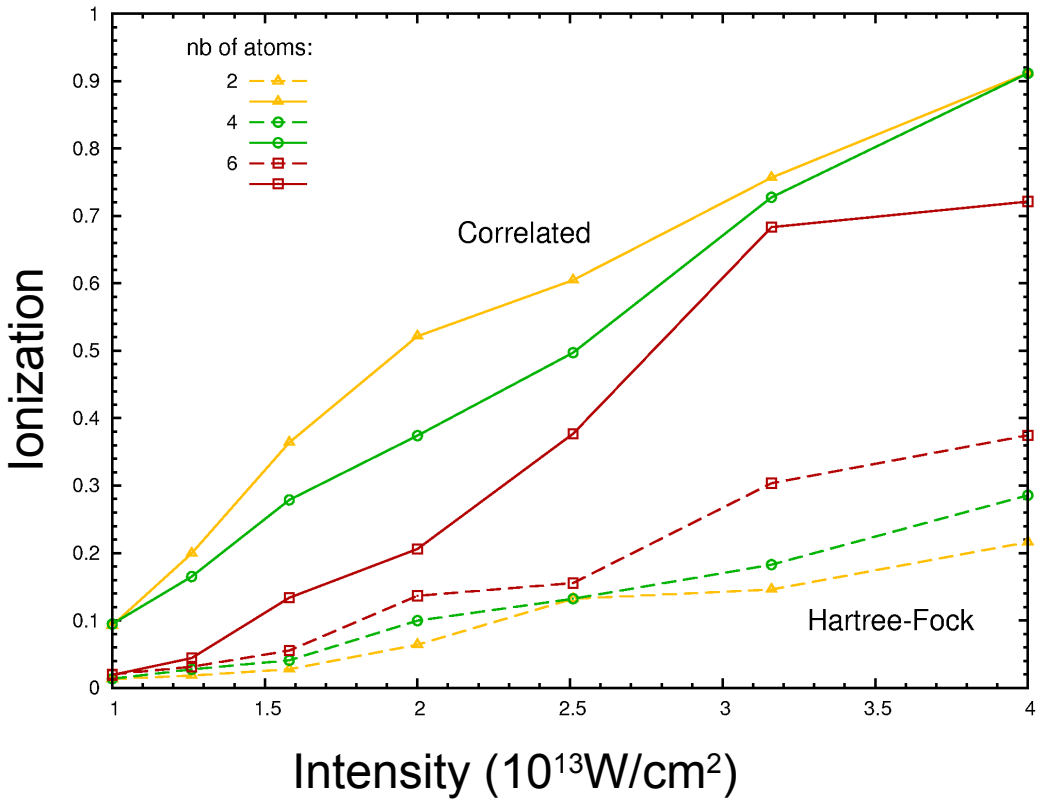
Basic picture correct with 4 orbitals

Qualitatively "all tunnels are alike" !
Quantitative consequence for orbital imaging
remain to be investigated

Strong field ionization of large molecules

1-d model molecules
Dependence of ionization on

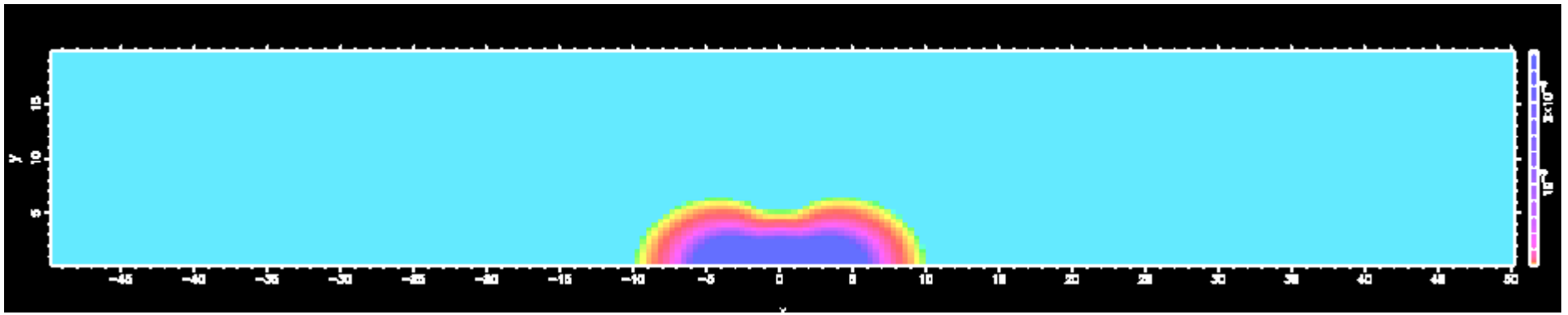
- laser intensity
- size of the molecule
  = number of active electrons



Multiconfiguration quantitatively and qualitatively differs from single-configuration Hartree-Fock

Ionization of a molecule with 6 active electrons

6 nuclei, 1 active electron/nucleus,
ionization potential: 0.3 a.u.
laser intensity: 3×10^{14} W/cm^2



NOTE: ~ 80 % ionization

First results:
More stable than 1d: comparable depletion at 10 times the intensity

Summary

- *Ab initio* time-dependent code for cylindrically symmetric systems
- Highly scalable parallel implementation
- Arbitrary potential shapes
- Non-perturbatively strong external fields
- Realistic applications to strong-field laser-atom and laser-molecule interactions

Outlook

- Technical improvements
  - Space discretization (e.g. “cascading”)
  - Time-integration methods: CMF (accuracy ?)
  - H-matrix representation of $1/(r_1-r_2)$
- Extend applications:
  - stacks of quantum dots
  - introduce nuclear motion
  - non-cylinder symmetric systems

People

Juergen Zanghellini: 1d code (now U. Graz)

Markus Kitzler: 1d code (now doing experiments)

Jeremie Caillat: 3d, cylinder coordinates (now CNRS, Paris)

Gerald Jordan: recent calculations

Christopher Ede: visualization

Money:

Austrian Science Foundation:

SFB ADLIS – Advanced Light Sources

SFB AURORA – High Performance Computing