Double Ionization of Atoms in Intense Laser Fields

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- Calvin College Research Fellowship (release time)
- National Science Foundation (RUI grant)
- J.H. Eberly and R. Panfili of the University of Rochester
- Student assistants Z. Smith, A. Karim, L. Breen

Background--Calvin College

- Calvin College is a comprehensive college of 4,000 students in Grand Rapids, MI
- Physics and Astronomy Department has 8 tenure-track faculty (7.5 postns)
  - 5 experimental physicists
  - 2 astronomers
  - 1 theorist
- Graduate ~5 physics majors per year. About half immediately go on to grad school.

My background

- I started at Calvin in 1983, fresh from a postdoc
- Spent at least part of each of the first 3 summers back at Colorado, where I'd postdocked.
- Received my first grant for research at Calvin in 1987 (Research Corp + NSF-RUI), and hired my first undergraduate assistant in Summer 1987
- Continuous NSF support since then

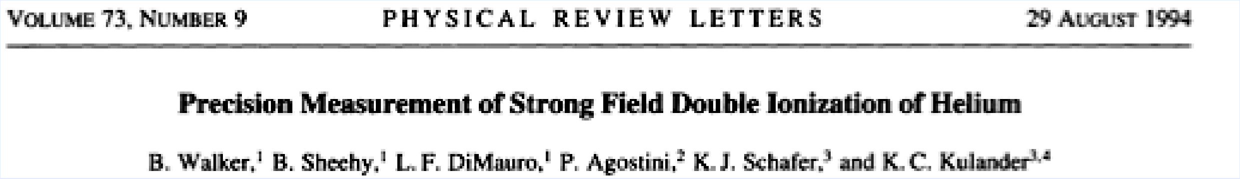
Professional Development

- Sabbatical 1992-93 to University of Rochester
  - Funded in part through NSF ROA program
  - Collaboration (with J.H. Eberly) still ongoing!
- Sabbatical also marked a transition from work that was analytically based to work that was more computer based.
  - Brought “supercomputer techniques” back to Calvin
  - Now do my work on desktop machines

Usual Procedures

- I like to hire 2 students each summer.
- Ideally, one of the students is experienced and can help the other
- The students work on closely related threads of the same project
- I challenge each student to take ownership of the thread, and to see it through to completion by working on a part-time basis through the academic year

The Motivation



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Precision Measurement of Strong Field Double Ionization of Helium

B. Walker,[1] B. Sheehy,[1] L. F. DiMauro,[1] P. Agostini,[2] K. J. Schafer,[3] and K. C. Kulander[3,4]

Experiments in the early 1990s showed considerably more double ionization occurring than would be expected for independently behaving electrons.

- Somehow the electrons share energy and escape together

first ionization 16 photons; second 34 more

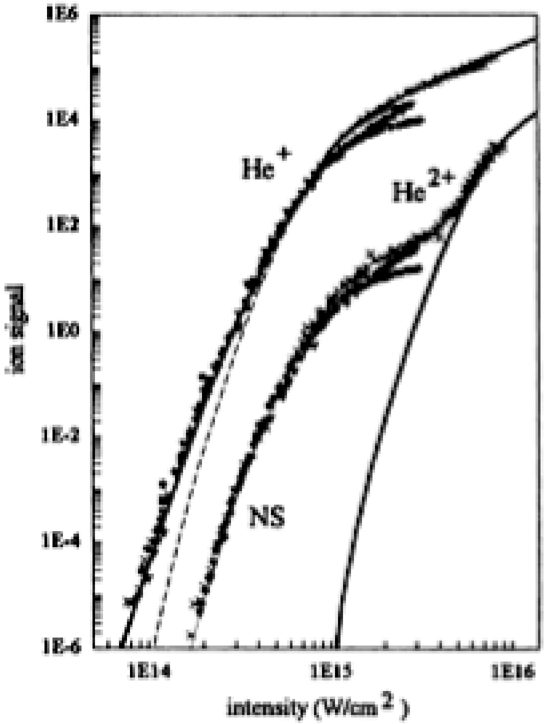


FIG. 1. Measured He ion yields for linear polarized, 100 fsec, 780 nm light. Calculations are shown as solid (SAE) and dashed (ac-tunneling) lines. The measured intensities are multiplied by 1.15. The solid curve on right is the calculated sequential He^{2+} yield.

The Challenge

We can't just solve the 3-d Time-Dependent Schrödinger Equation for two electrons in an oscillating electric field, even on the best computers.

Instead, we (& several other groups) worked with a one-dimensional model of the helium atom

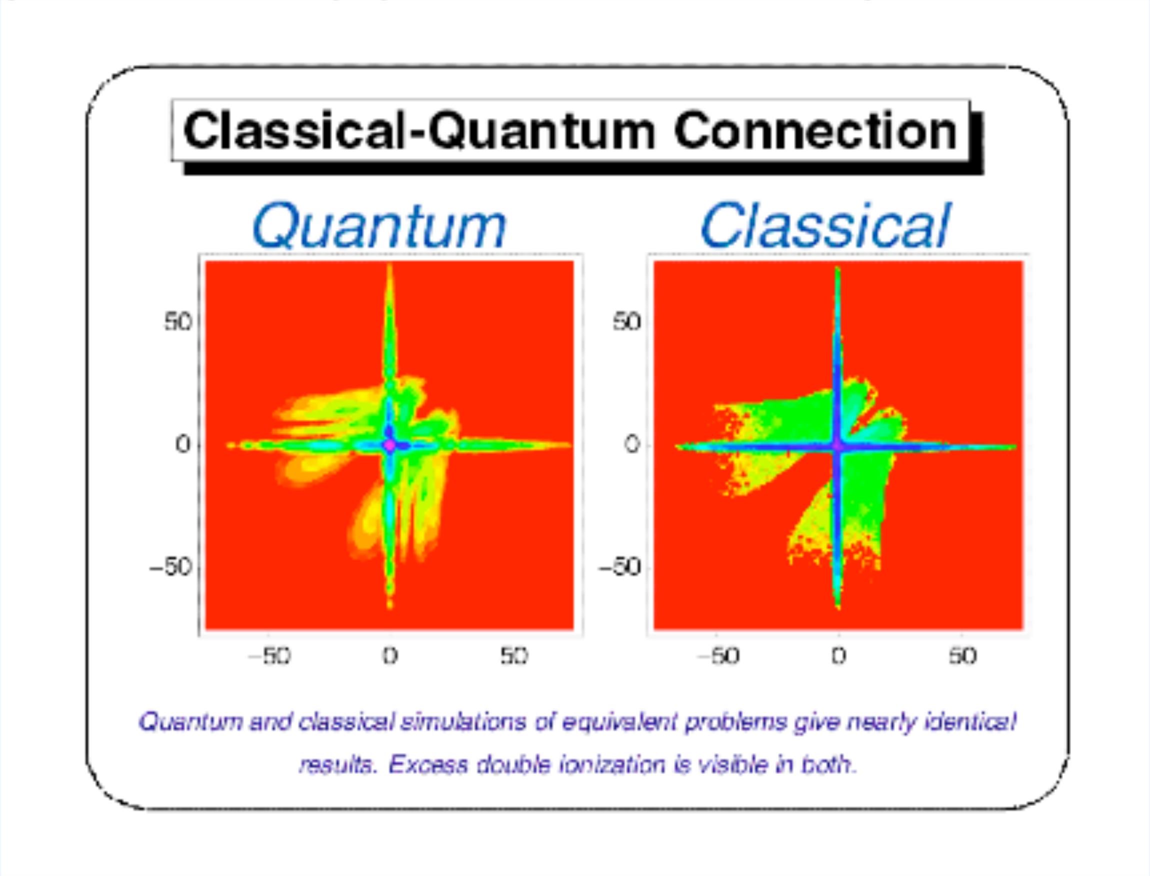
- In the model, each electron could move only parallel or antiparallel to the laser polarization
  - Picture the electrons as being on railroad tracks
- A full quantum solution is possible.
- We learned a lot,
  - including that the electron behavior seemed very classical.
- So we tried classical modeling of the one-dimensional system,
  - and we found that the classical results reproduced all the important quantum results.

Overview of Classical Method

- We set up an ensemble of classical two-electron atoms.
  - Each atom has slightly different initial conditions, so that collectively they imitate the quantum ground state of the atom.
  - Ensemble sizes typically 400,000.
- We evolve each two-electron atom in time through a laser pulse, using Newton’s laws of motion.
  - Account for electron-electron interaction, electron-nucleus interaction, and influence of laser field (in dipole approximation).
- After each run, we can sort the trajectories and
  - Study statistical behavior;
  - Backtrack individual trajectories to learn their history.

Classical simulations

- Classical ensembles exhibit behavior very similar to the quantum systems.
- Example below shows population distributions at a particular time.



Classical-Quantum Connection

Quantum and classical simulations of equivalent problems give nearly identical results. Excess double ionization is visible in both.

R. Panfili, J.H. Eberly and S. Haan, Opt. Express 8, 431-435 (2001).

Extension to 3D

The success of 1D classical simulations gives us motivation to pursue the 3D classical case.

The classical advantage--

In classical studies we find the time development for each classical atom separately.

The QM wavefunction incorporates, at each time, everything that can happen

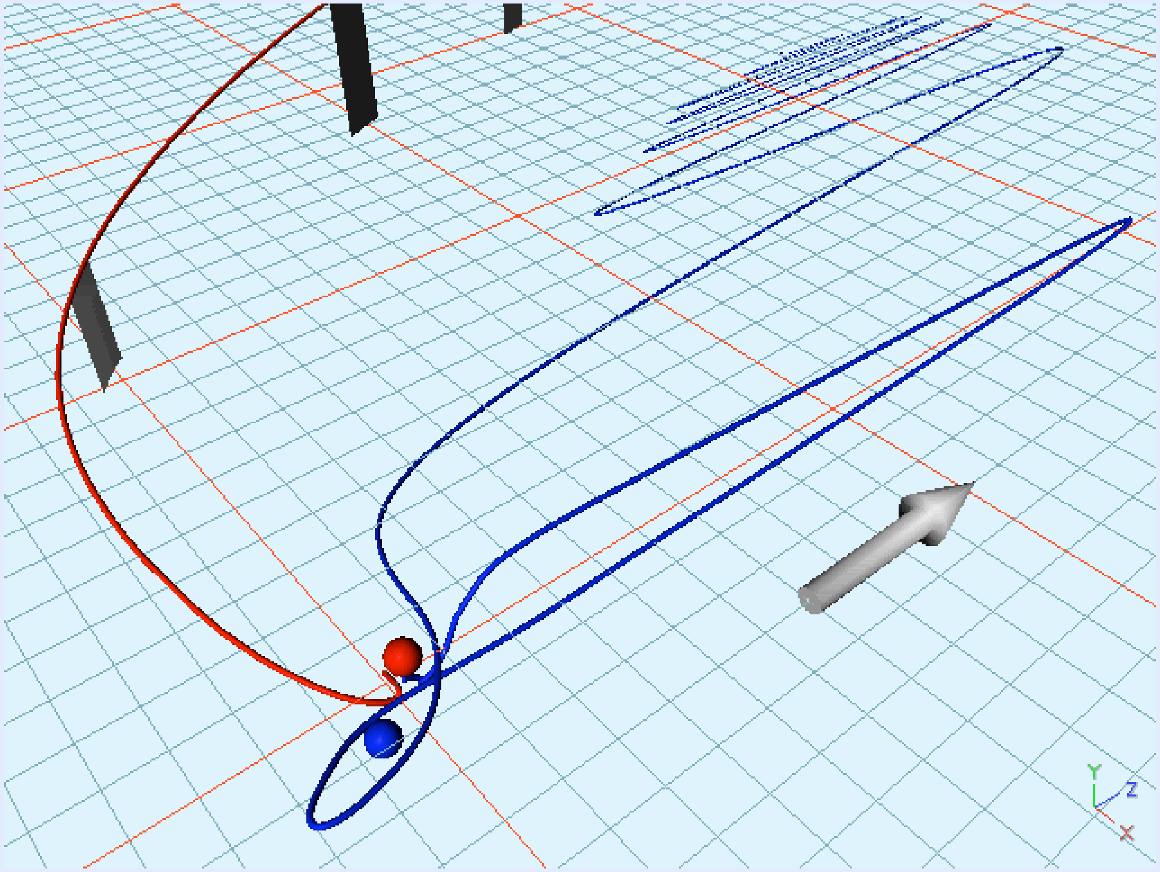
R. Panfili (Rochester grad student) wrote a first draft of a computer program for the 3d classical case.

He graduated & took a job in industry
without getting the kinks out of it.

My students and I got the kinks out

Example double ionization trajectory :

Recollision ionization, with brief recapture of recolliding electron

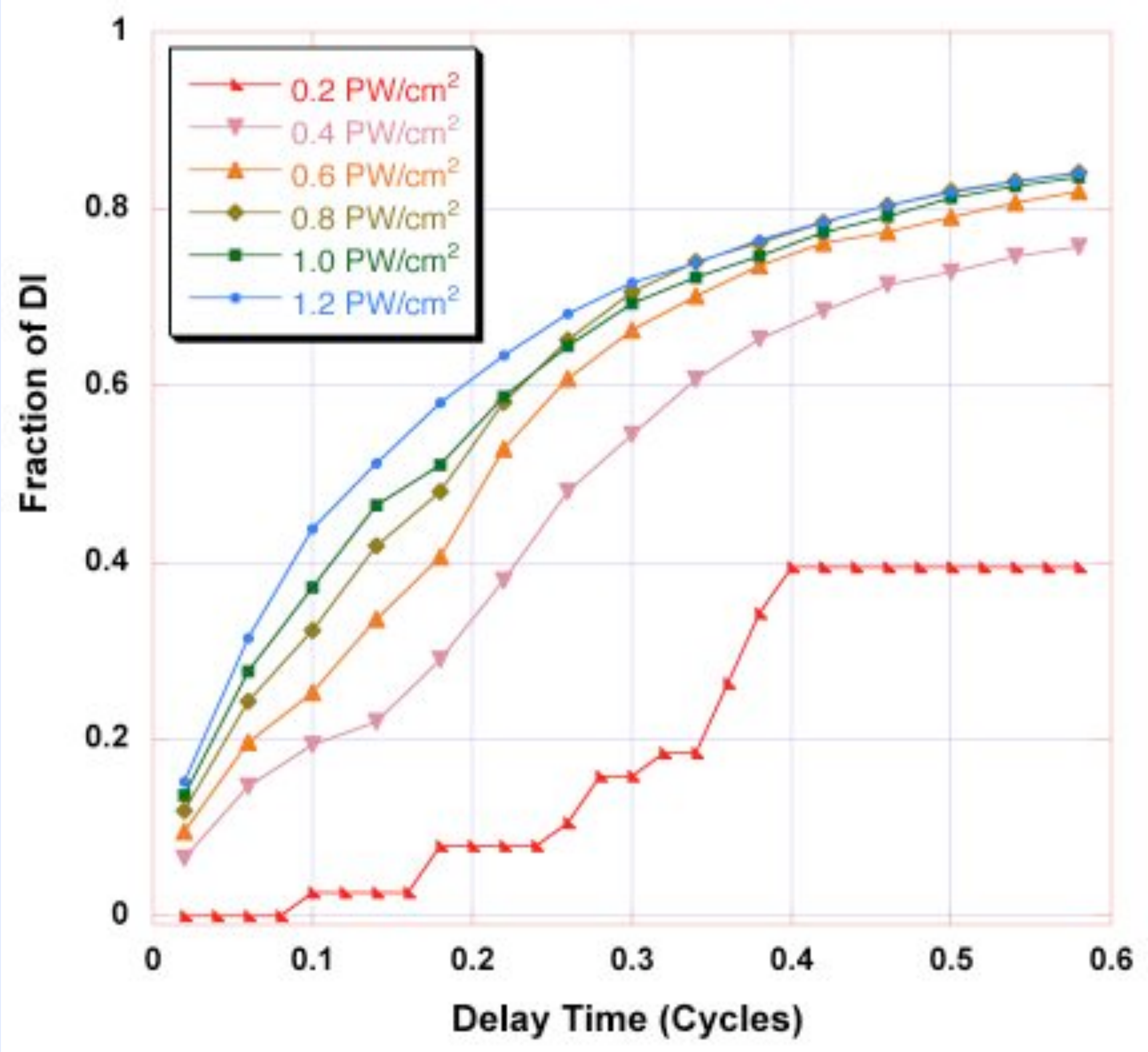


Recollision had been proposed from the outset (by others), but most everyone overlooked this possibility of having one electron bound for a portion of a cycle after recollision.

Delay time between recollision & double ionization

- Most DI trajectories show a part-cycle phase delay between recollision and double ionization

- Runs at right are all for laser wavelength 780 nm.



Final longitudinal momenta sorted by: delay times from recollision to ionization **and** by final direction relative to recollision direction

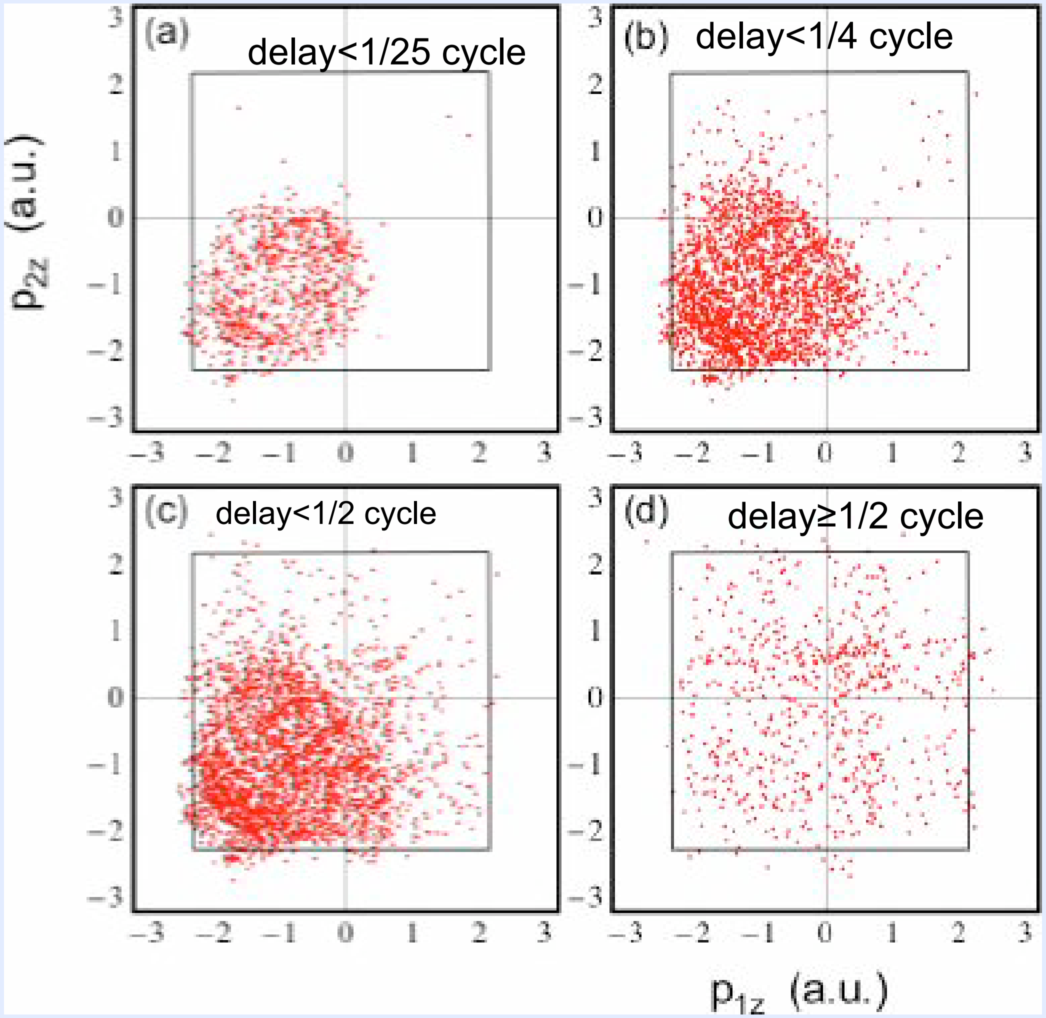
I=6x10^{14} W/cm^2

RE directions--adjust signs of momenta so all collisions occur with returning electron traveling in the +z direction.

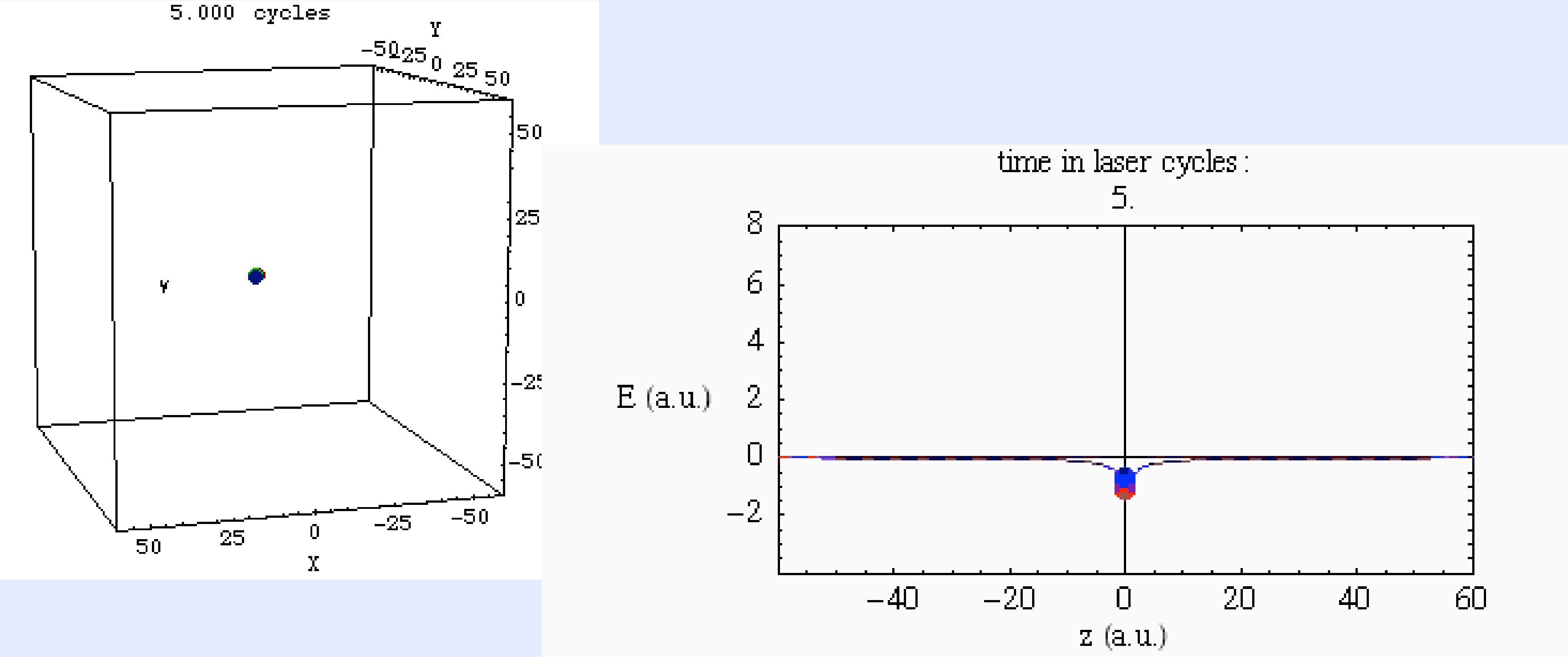
- •For small delay times, almost all final z-momenta are *opposite* from the recollision direction.

- •With increased delay times, there is increased spillover into the 2nd and 4th quadrants.

S.L. Haan, Calvin College



- If, to first approximation, second electron ionizes after the field peaks, the electrons can have drift velocities in opposite momentum hemispheres.



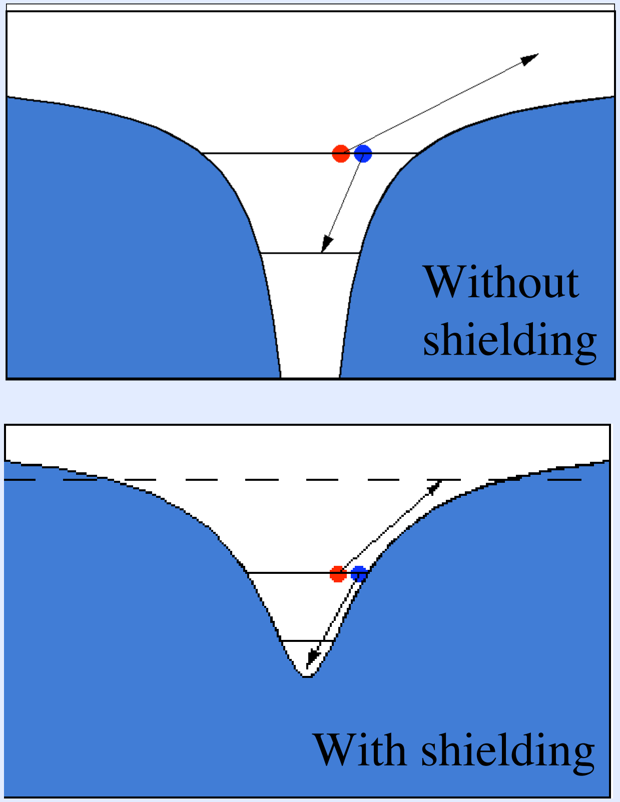
- These results all summarized in two recent papers
    - S.L. Haan, L. Breen,* A. Karim,* and J.H. Eberly, "Recollision Dynamics and Time Delay in Strong-Field Double Ionization," Optics Express **15**, 767-778 (February 2007).
    - S.L. Haan, L. Breen,* A. Karim,* and J.H. Eberly, "Variable Time Lag and Backward Ejection in Full-Dimensional Analysis of Strong-Field Double Ionization," Phys. Rev. Lett. **97**, 103008 (8 September 2006). Republished in Journal of Ultrafast Science **5**, Issue 10 (October 2006).

A challenge for 3D: Classical 3-D Helium is unstable, even without external fields

- One electron can dive deep into potential-energy well, letting the other escape.
- The well is bottomless if use Coulomb potentials.
- Can stabilize by screening the Coulomb interaction:

$$1/r \rightarrow 1/\sqrt{r^2 + a^2}$$

- For initial energy = -2.9035 au, setting a ≥ 0.7 au prevents self-ionization
  - Results I've shown a=0.825 au



Plots show only the nuclear.

Populating initial ensemble to mimic 3D ground state?

There are various possibilities related to energy and angular momentum.

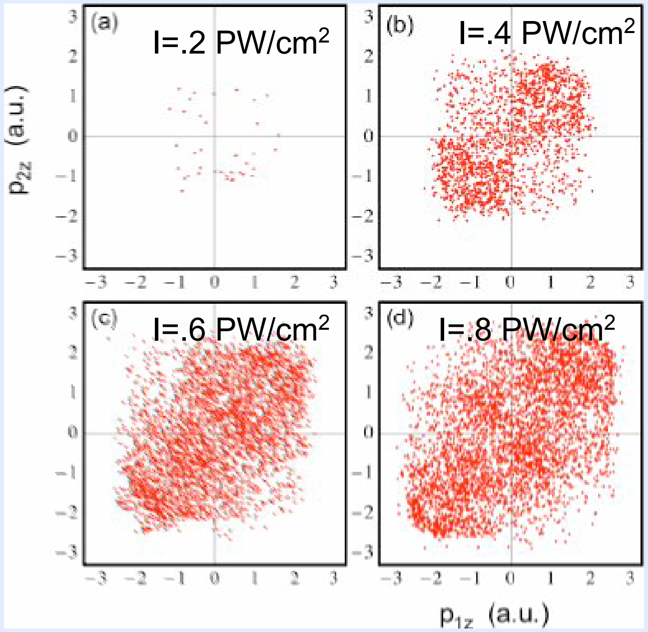
We simply:

- -Set electron-nuclear shielding at 0.825 au, and electron-electron shielding at 0.05 au
- -Fixed the energy at -2.9035 au (He ground state)
- -Choose classically allowed positions
  - Employed several methods--He quantum ground state, Gaussian, random
  - --but always spherically symmetric
- -Randomly divided the available KE between the two electrons
  - (subject to constraint that each electron have total energy <-1 au)
- -Gave each electron zero angular momentum.
  - (If don't constrain angular momentum, we get very little double ionization.)
- -Allow the system to evolve for the equivalent of one laser cycle (~100 au) with no laser field
  - Total energy and total angular momentum conserved
  - Position and momentum distributions stabilize

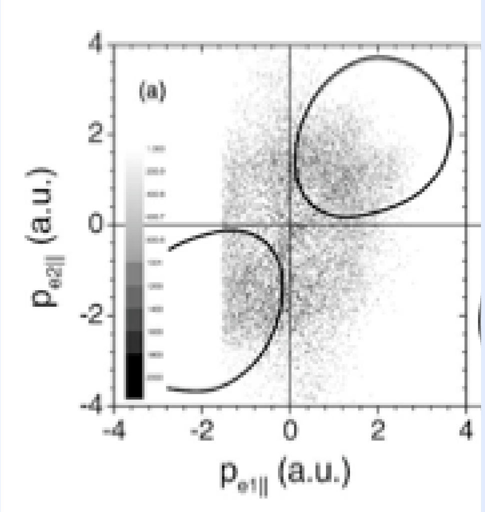
A difficulty

- The screening of the nucleus works well for stabilizing our initial state
  - It can be justified on the basis of the uncertainty principle--the system starts in a state with low uncertainty for energy and momentum, and electron isn’t allowed to know very precisely its position relative to the nucleus
- But what about at recollision?
  - We’ve found that *details of the final electron momentum & energy distributions depend on the form of the interaction with the nucleus*
    - One electron can scatter off nucleus at recollision
      - This electron may be either free or still bound
    - Small impact parameter ⇒ large deflection, if nucleus unshielded
  - In order to reproduce experimental results, we need to partly unscreen the nucleus.

Our results:



Experiment (I=.5 PW/cm²)

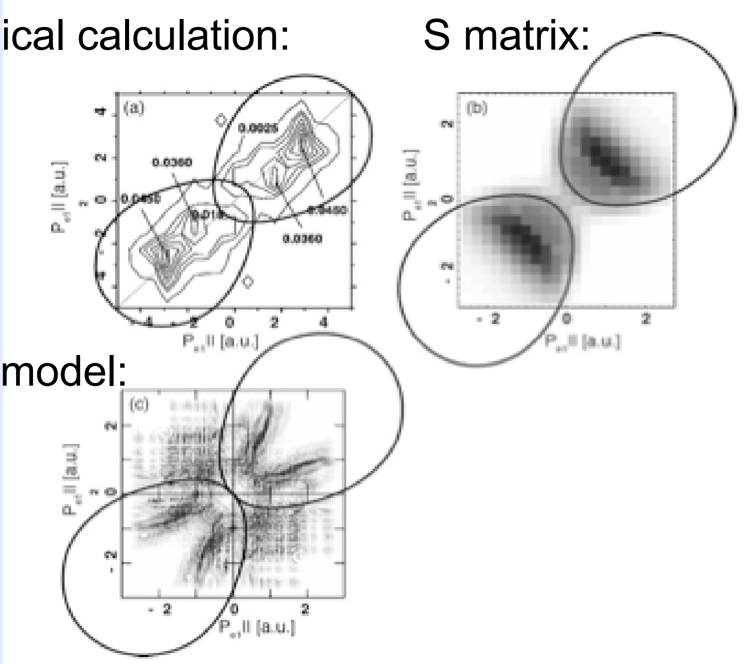


Other theoretical results

Classical calculation:

S matrix:

1-d model:



Egg-like shapes show kinematically allowed regions after recollision ionization

Experimental results--V.L.B. de Jesus, et al., Journal of Electron Spectroscopy 141, 127 (2004);

theoretical--J. Chen et al PRA 63,011404R (2001); A. Becker and F.H.M. Faisal, PRL89, 193003 (2002); Lein et al PRL 85, 4707 (2000)

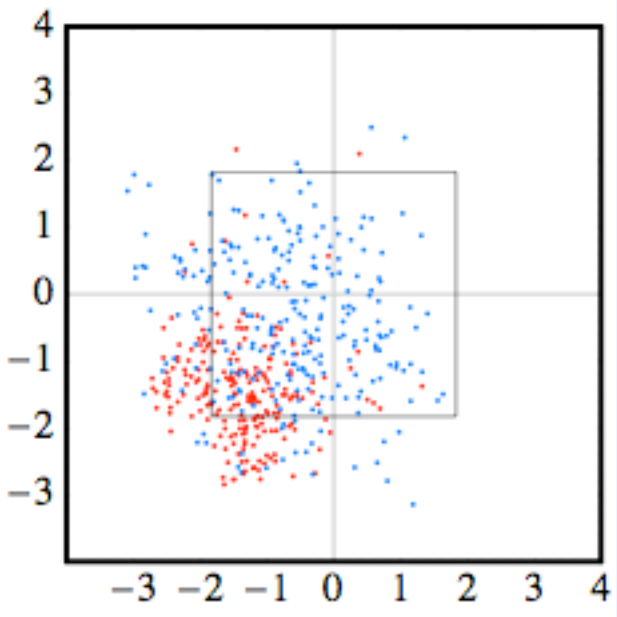
Current work

- This summer my students and I have revised our program with a “toggle switch” on the nuclear screening

  –When one electron ionizes, we reduce the nuclear shielding for both electrons

    •We adjust the electrons’ radial kinetic energies to preserve total energy

  -Now we get trajectories with higher final energy.



Present Status

- We’re sytematically adjusting the shielding at return
- I’m working on a “dynamic shielding,” in which the screening of the nucleus for each electron depends on the energy of that electron
  - How, precisely, should the screening depend on the energy?
  - Should I add terms to the equation of motion to account for the “velocity dependent potential”?
- Research projects can lead us into new areas we know nothing about!