Double Ionization of Atoms in Intense Laser Fields

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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 postdoced.
- 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
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
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.

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


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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

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.

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• 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

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:
  \[ \frac{1}{r} \rightarrow \frac{1}{\sqrt{r^2 + a^2}} \]
- For initial energy = -2.9035 au, setting \( a \geq 0.7 \) 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 $\Rightarrow$ large deflection, if nucleus unshielded
  – In order to reproduce experimental results, we need to partly unscreen the nucleus.
Our results:

Experiment ($I = 0.5 \text{ PW/cm}^2$)

Other theoretical results

Classical calculation:

1-d model:

Egg-like shapes show kinematically allowed regions after recollision ionization

S matrix:

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