SINGLE AND DOUBLE IONIZATION OF ATOMS AND MOLECULES BY SHORT-PULSE INTENSE LASERS AND CHARGED-PARTICLE IMPACT

Klaus Bartschat, Drake University

Moscow State University
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Collaborators:

Xiaoxu Guan, Oleg Zatsarinny, Brant Abeln, Ethan Secor, Dan Weflen (Drake University)
Barry Schneider (National Science Foundation)
Cliff Noble (Daresbury Laboratory)
Johannes Feist (Technical University of Vienna)
Alexei Grum-Grzhimailo (Moscow State University)

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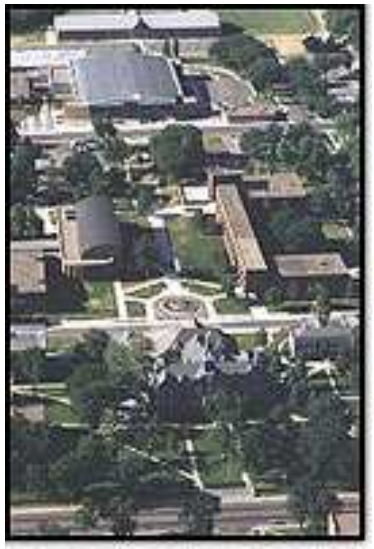

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OVERVIEW:

I. **Motivation: Why Attosecond Physics?**

II. **A Computational Challenge: Ionization of Atomic Hydrogen by an Infrared Pulse**
- Numerical Method and Difficulties
- The Matrix Iteration Method
- Example Results (experiment at Griffith University)
- Quasi-One-Electron Targets: Li (experiment at Heidelberg)

III. **Many-Electron Targets**
- The Situation for **Two-Electron Atoms and Molecules**
- **Extension to Complex Atoms**

IV. **The Time-Dependent *B*-Spline *R*-Matrix (TDBSR) Approach**
- **Theoretical Formulation**
- Connection to Electron Scattering and Atomic Structure
- Time Propagation
- **Numerical Aspects**

V. **Results: Excitation and Single Ionization of Neon and Argon**
- The One-Photon Case
- Multi-Photon Cases

VI. **More Results: Two-Photon Double-Ionization of Helium**
- Test Calculations for Single Pulse
- Two-Color Pulses with Delay

VII. **Recent Extensions**
- Application to Molecules: H_2^+ and H_2
- Charged-Particles as Projectiles: Antiprotons and Protons

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VIII. Conclusions and Outlook

No worries! I'll cut these parts short and show you just a few nice examples :):):)

Attosecond/Femtosecond Physics

- **1 attosecond** is defined as one-millionth of one millionth of one millionth (10^{-18}) of a second.
- There are **twice as many attoseconds in one second than there are seconds in the age of the universe (15 billion years)!**
  - Atomic unit of time:

$$\frac{0.529 \times 10^{-10}\,\mathrm{m}}{3 \times 10^{8}\,\frac{\mathrm{m}}{\mathrm{s}}/137} \approx 24 \text{ attoseconds}$$

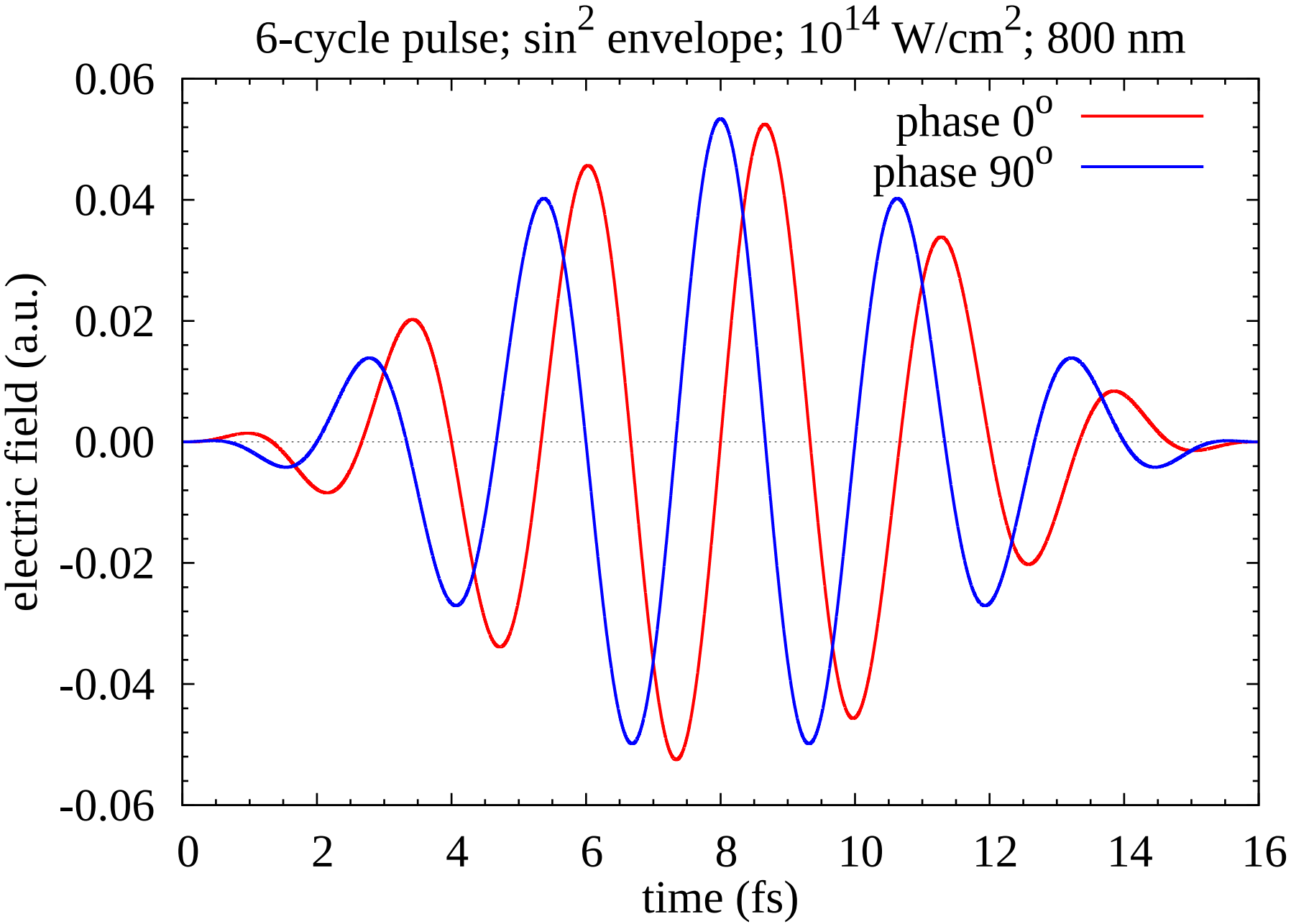
  - period for the $n = 1$ orbit in atomic hydrogen: ≈ 150 attoseconds (Bohr model)
- **Attosecond laser pulses provide a window to study the details of (valence) electron interactions in atoms and molecules**.
- These capabilities promise a **revolution in our microscopic understanding of matter.**
- A major role for **theory** in attosecond science is to **elucidate novel ways to investigate and to control electronic processes in matter on such ultra-short time scales.**
- If we could **control the behavior of valence electrons**, this may open up new avenues to:
  - **manipulate the outcome of chemical reactions**
  - **make novel materials**
  - **do many other fancy things we aren't even thinking of yet**
- Some experiments have been performed with attosecond pulses or pulse trains, but most single-pulse durations are in the femtosecond (1 fs = 1,000 as) regime (though getting shorter fast).

Note: 900 attoseconds sounds better than 0.9 femtoseconds!

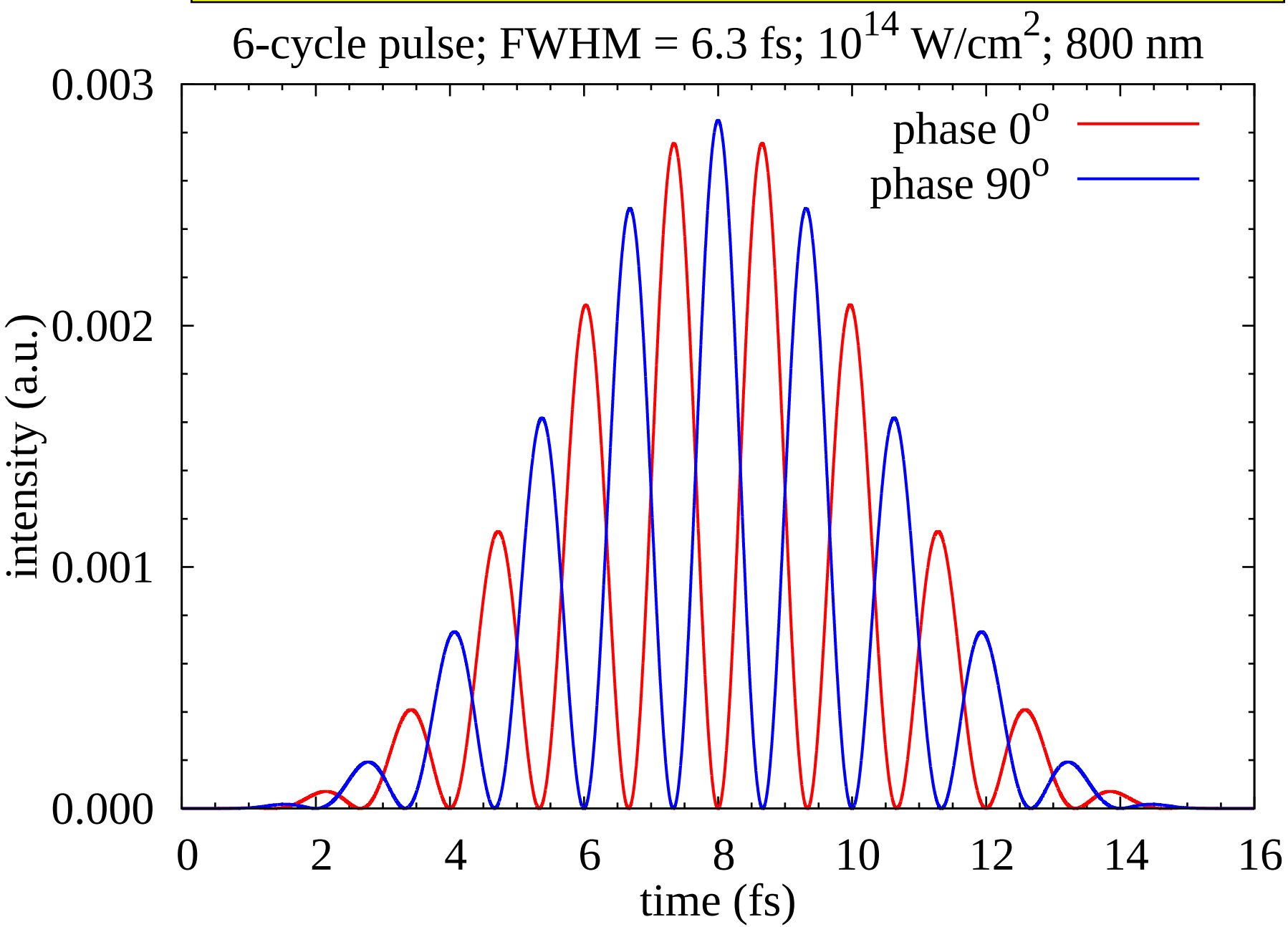
A Computational Challenge: Ionization of Atomic Hydrogen by an Infrared Pulse

- We study the hydrogen atom under the influence of an **intense femtosecond laser pulse**.

Example of a short pulse (Griffith experiment)



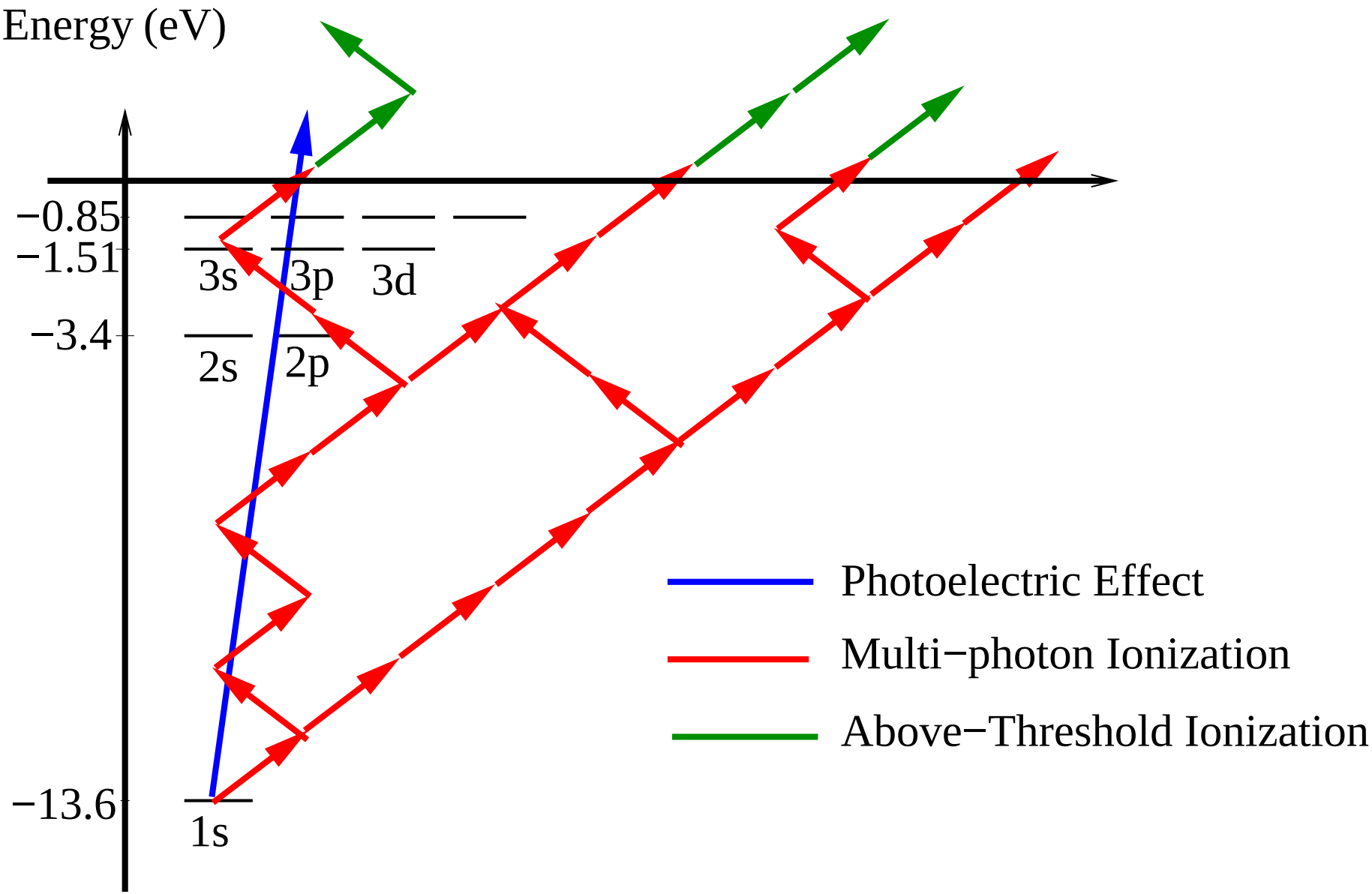
carrier envelope (CEP) phase can have an effect; measure it – or average over it!



A Computational Challenge: Ionization of Atomic Hydrogen by an Infrared Pulse

- We study the hydrogen atom under the influence of an **intense femtosecond laser pulse**.
- The intensities range from $10^{12} - 10^{15}\,\mathrm{W/cm^2}$ concentrated on a tiny area (less than $1\,\mathrm{mm^2}$).
- **$10^{14}\,\mathrm{W/cm^2}$ is a million billion times stronger than the radiation that the Earth gets from the Sun directly above us on a clear day**.
- Such intensities can rip electrons away from atoms in a very different way from the standard photoeffect:
  - Several photons can collaborate in a **multi-photon ionization process**.
  - **Above-threshold ionization** can give additional energy to the ejected electron.

Single vs. Multi−Photon Ionization in Atomic Hydrogen



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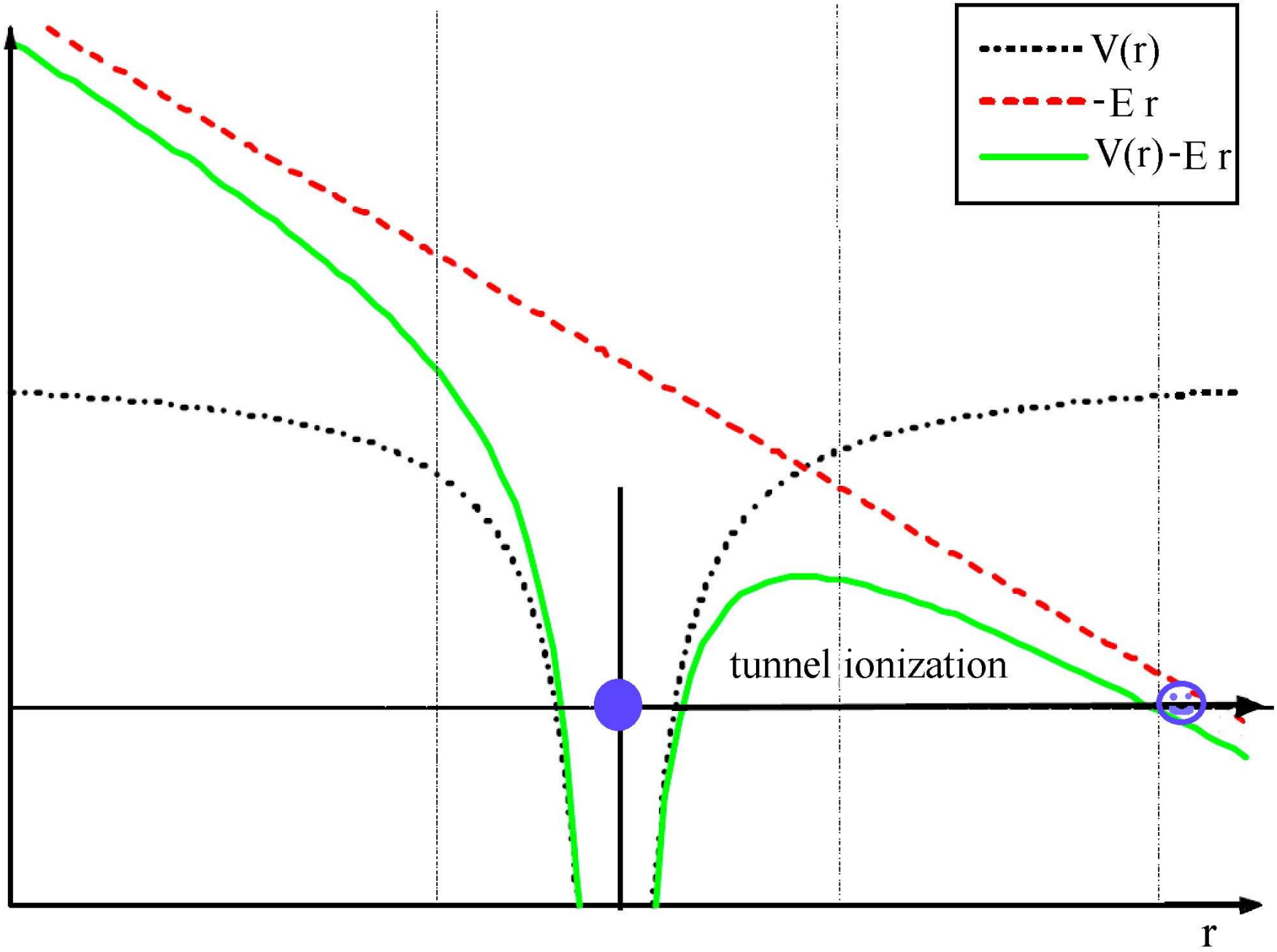
  - Several photons can collaborate in a **multi-photon ionization process**.
  - **Above-threshold ionization** can give additional energy to the ejected electron.
  - **Field (tunnel) ionization** may be possible as well.

Field (Tunnel) Ionization



Why Study Atomic Hydrogen?

- **Theorists:**
  - This is, in principle, an "exactly" (to machine accuracy) solvable problem.
  - We know the non-relativistic field-free states.
  - We can learn about
    - time propagation
    - extraction of results
- **Experimentalists:**
  - This is technically difficult, because of:
    - atomic hydrogen
    - producing and controlling short intense pulses
    - detection issues – what electrons are really detected?
  - Once we can handle and understand all this, we are ready for more complex targets.
  - **Show these arrogant theorists that they don't know everything (yet)!**
  - **Get assistance from theory – find out the actual parameters in the experiment.**

Why is this Interesting and Challenging?

- **Keldysh Parameter**: $\gamma \equiv \sqrt{E_b/2U_p}$, where $U_p = I/4\omega^2$ is the "ponderomotive energy".
- The classical interpretation of U_p is the average energy of an electron "wiggling" (quivering) in the oscillating field.
- This effectively increases the binding energy E_b (without the field) in the above formula.
- For high intensities I and large wavelengths (small ω), the ponderomotive energy can be many eV and thus totally change the physics of the problem.

- **Keldysh Parameter (for atomic hydrogen)**:

$$\gamma \approx \frac{850}{\lambda[\mathrm{nm}]\sqrt{I[10^{14}\mathrm{W/cm^2}]}}$$

- **What does it mean?**
  - $\gamma \gg 1 \rightarrow$ multi-photon ionization ($\rightarrow$ Floquet-theory; somewhat trivial time dependence can be factored out)
  - $\gamma < 0.5 \rightarrow$ tunnel ionization $\rightarrow$ strong-field approximation (SFA)
  - $\gamma \approx 1 \rightarrow$ no clear picture and treatment becomes very difficult ($\rightarrow$ **solve the TDSE**)
  - $\gamma = 1.06$ **for** $\lambda = 800\,\mathrm{nm}$ **and** $I = 10^{14}\,\mathrm{W/cm^2}$**!!!**

Numerical Method

- We solve the **time-dependent Schrödinger equation**

$$i\frac{\partial \Psi(\mathbf{r},t)}{\partial t} = H(\mathbf{r},t)\Psi(\mathbf{r},t)$$

with the atomic Hamiltonian and a **linearly polarized laser field**:

$$H(\mathbf{r},t) = -\frac{\Delta}{2} + V(\mathbf{r}) + r\cos\vartheta\, E(t)$$

r•E "length form"

E(t) only: dipole approximation

by the time-dependent close-coupling method:

$$\Psi(\mathbf{r},t) = \frac{1}{r}\sum_{\ell=0}^{\infty} a_\ell(r,t)\sqrt{\frac{2\ell+1}{4\pi}}P_\ell(\cos\vartheta).$$

- The coefficients $a_\ell(r,t)$ satisfy the set of close-coupling equations:

$$i\frac{\partial a_\ell(r,t)}{\partial t} = \left[-\frac{1}{2}\frac{\partial^2}{\partial r^2} + \frac{\ell(\ell+1)}{2r^2} + V(r)\right] a_\ell(r,t) + rE(t)\sum_{\ell'=\ell\pm1}\nu_{\ell\ell',1}\, a_{\ell'}(r,t);$$

$$\ell = 0, 1, ..., \ell_{max}.$$

- This is a **coupled system of partial differential equations**; we sometimes have up to **100 functions**, each of which is defined on up **200,000 points** in space.
- We typically propagate the initial solution for **20,000 - 100,000 time steps**.

Observables of Interest

- **Photoelectron spectrum:**

$$\frac{d\sigma(E)}{dE} = \sum_{\ell} |Z_{E\ell}|^2 ,$$

where

$$Z_{E\ell} = \lim_{t\to\infty} \int_0^\infty P_{E\ell}(r)\, a_\ell(r,t) dr$$

- **Angular distribution of photoelectrons:**

$$\frac{d^2\sigma}{dE d\Omega_{\mathbf{k}}} = \left| \langle\, \Phi^-_{\mathbf{k}}(\mathbf{r}) | \Psi(\mathbf{r},t)\, \rangle \right|^2_{t\to\infty} = \frac{\sigma(E)}{4\pi} \left[1 + \sum_L \beta_L(E)\, P_L(\cos\theta) \right] ,$$

with the photoelectron wavefunctions of the atomic Hamiltonian:

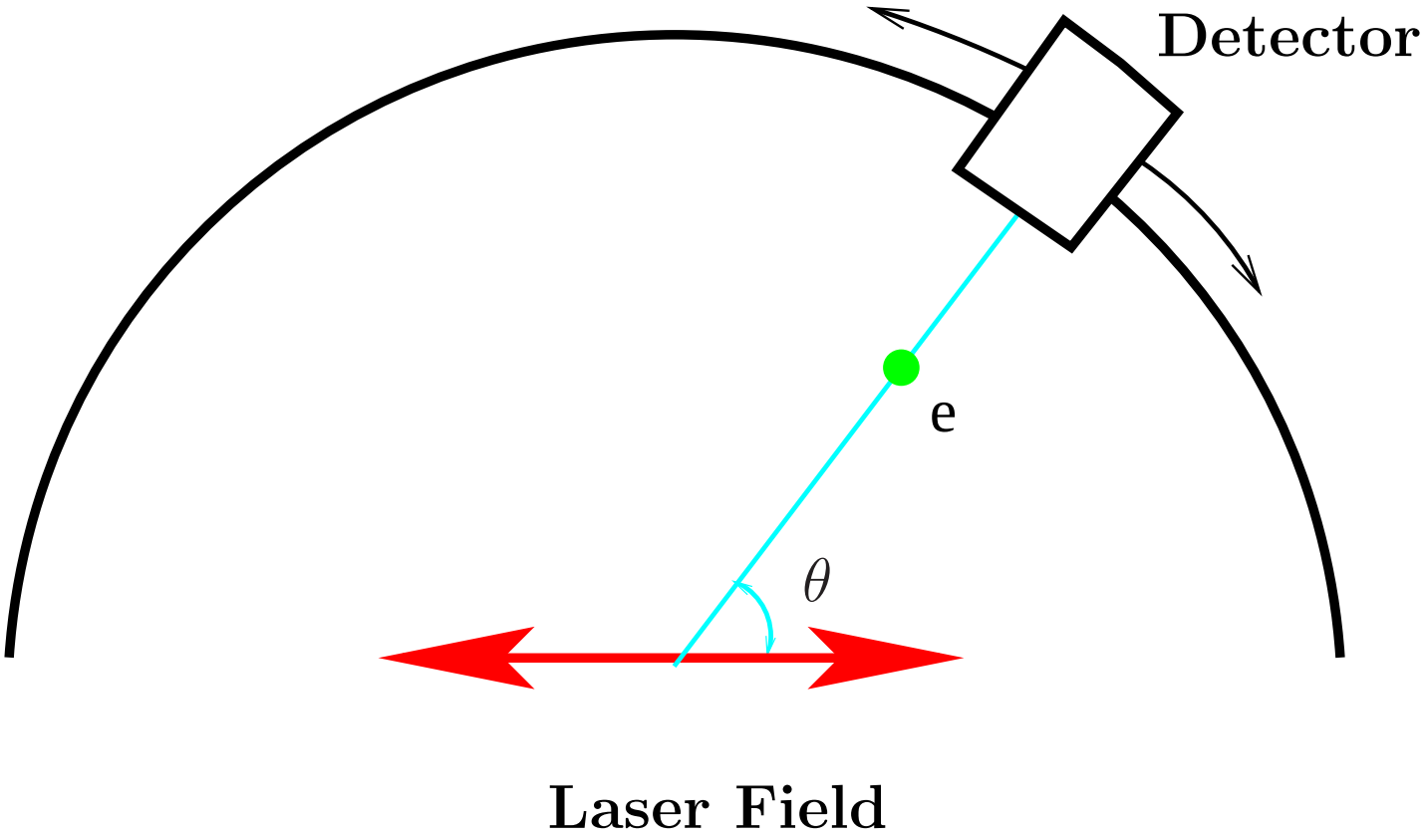
$$\Phi^-_{\mathbf{k}}(\mathbf{r}) = 4\pi r^{-1} \sum_{\ell m} i^\ell e^{-i\delta_{E\ell}} P_{E\ell}(r) Y^*_{\ell m}(\hat{\mathbf{r}}) Y_{\ell m}(\hat{\mathbf{k}}).$$

- **Anisotropy parameters:**

$$\beta_L(E) = (2L+1) \sum_{\ell\ell'} i^{\ell'-\ell} e^{i(\delta_{E\ell} - \delta_{E\ell'})} \nu_{\ell\ell',L}\, Z_{E\ell}\, Z^*_{E\ell'} \Bigg/ \sum_{\ell} |Z_{E\ell}|^2 ,$$

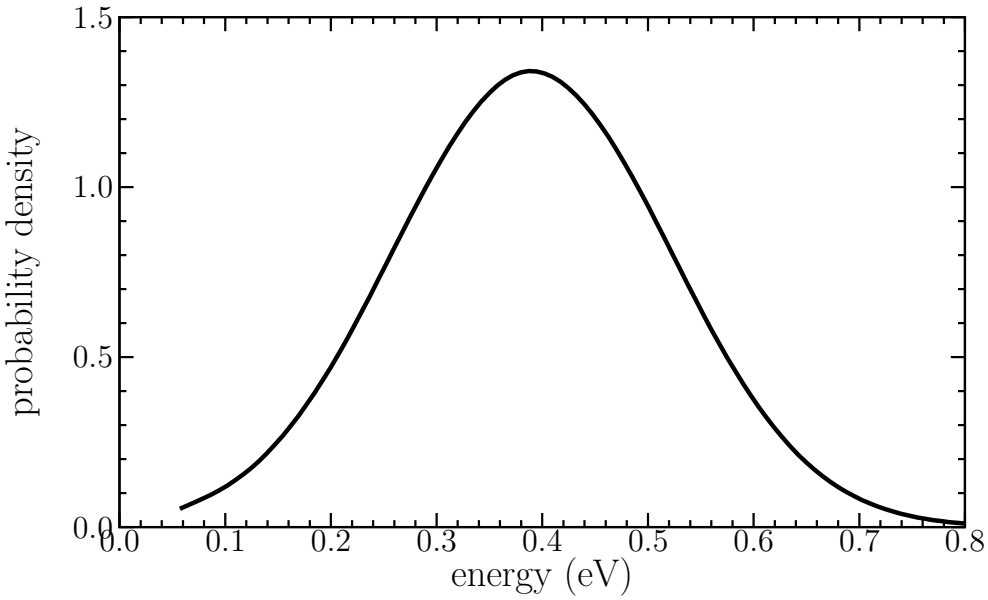
where $\nu_{\ell\ell',L} = \nu_{\ell'\ell,L} = \sqrt{(2\ell+1)/(2\ell'+1)}\, (\ell 0, L 0 \,|\, \ell' 0)^2$.

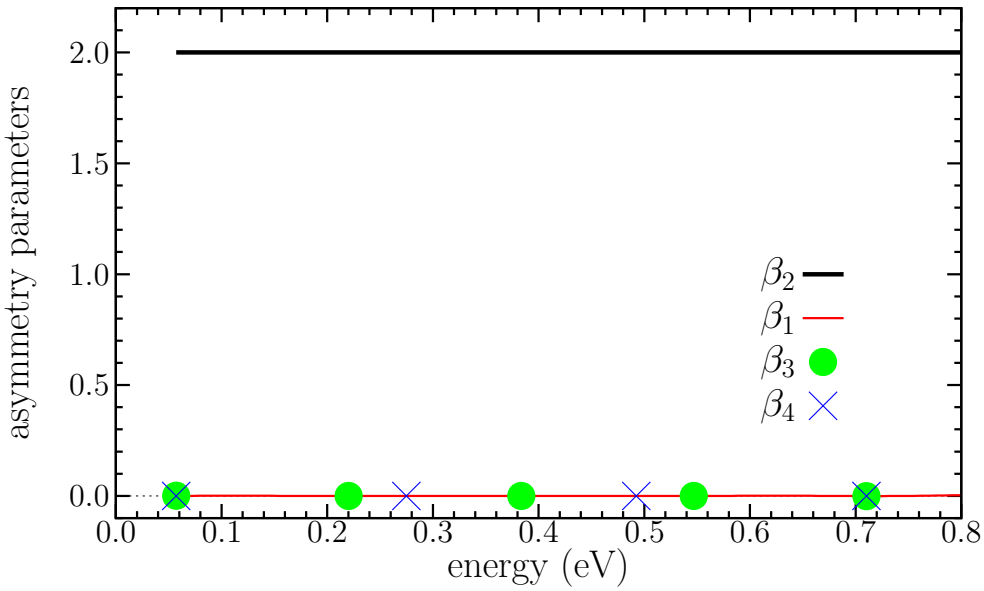
Scheme of an Angular-Distribution Experiment

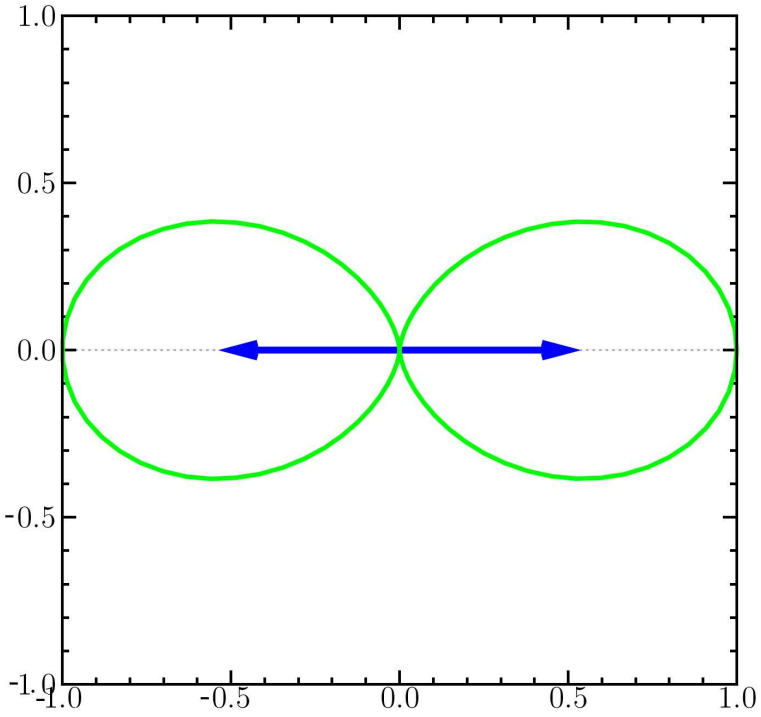


First Test: UV light, 88 nm, 10^{15} W/cm^2

Single-Photon Ionization by Short Pulse with Energy 14.0 eV





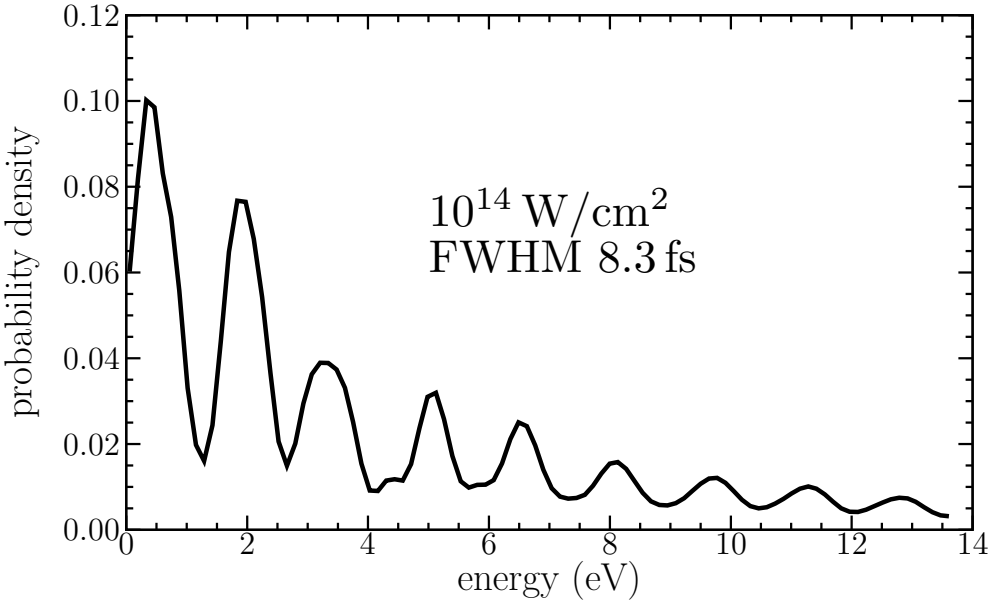


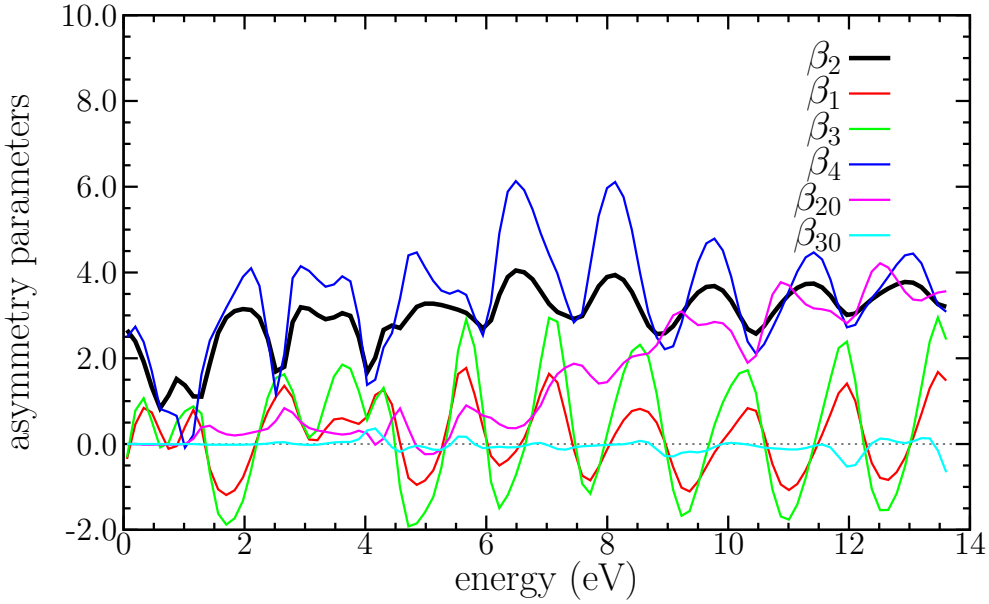
nice, but kind of boring

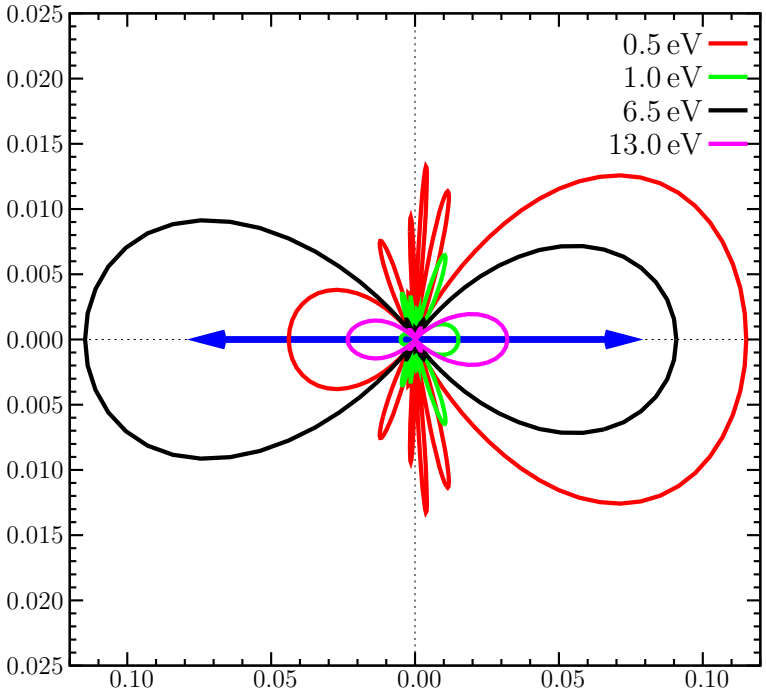
and easy to calculate

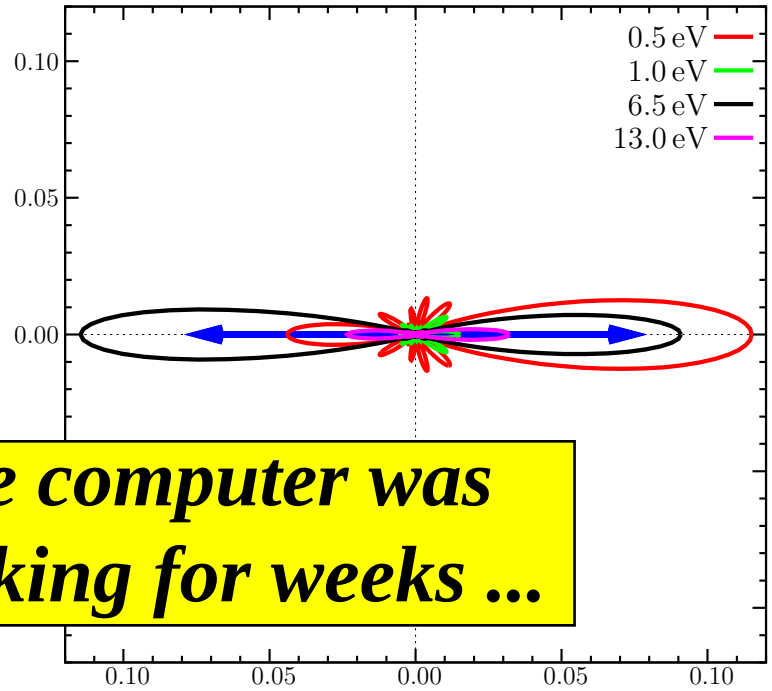
IR light, 800 nm, 10^{14} W/cm^2

Multi-Photon Ionization by Short Pulse with Energy ≈ 1.5 eV









the computer was working for weeks ...

Difficulties for Higher Intensities

- Results in the length form of the electric dipole operator converge **very s l o w l y.**
- The velocity form is expected to be more appropriate, but it carries its own challenges:
  - The interaction term $\boldsymbol{A} \cdot \boldsymbol{p}$ rather than $\boldsymbol{E} \cdot \boldsymbol{r}$ involves a derivative.
  - While $\boldsymbol{E} = \partial \boldsymbol{A}/c\partial t$, "field-free" does not mean that the vector potential vanishes.
  - This causes numerical and interpretation problems if $\boldsymbol{A}$ is not zero at the end of the pulse (residual **static $\boldsymbol{E}$ field**)
  - While the mathematics (**gauge invariance**) is fine,
    - **theorists avoid the problem by setting $\boldsymbol{A}$**

      BUT
    - **experimentalists control $\boldsymbol{E}$ rather than $\boldsymbol{A}$!**
- This is a problem of ongoing research.

Which gauge is best?

J. Phys. B: At. Mol. Opt. Phys. **29** (1996) 1667–1680. Printed in the UK

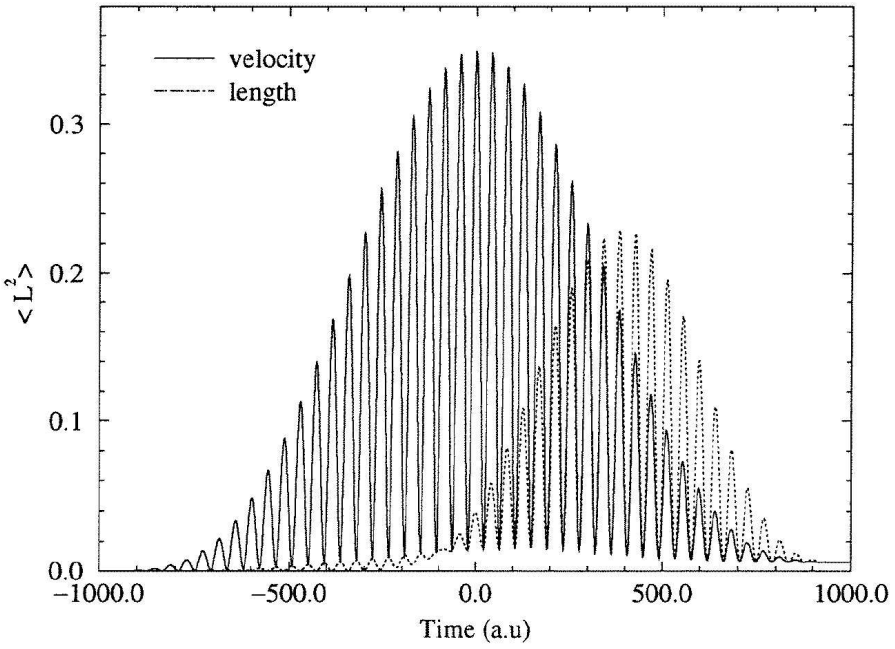
Optimal gauge and gauge invariance in non-perturbative time-dependent calculation of above-threshold ionization

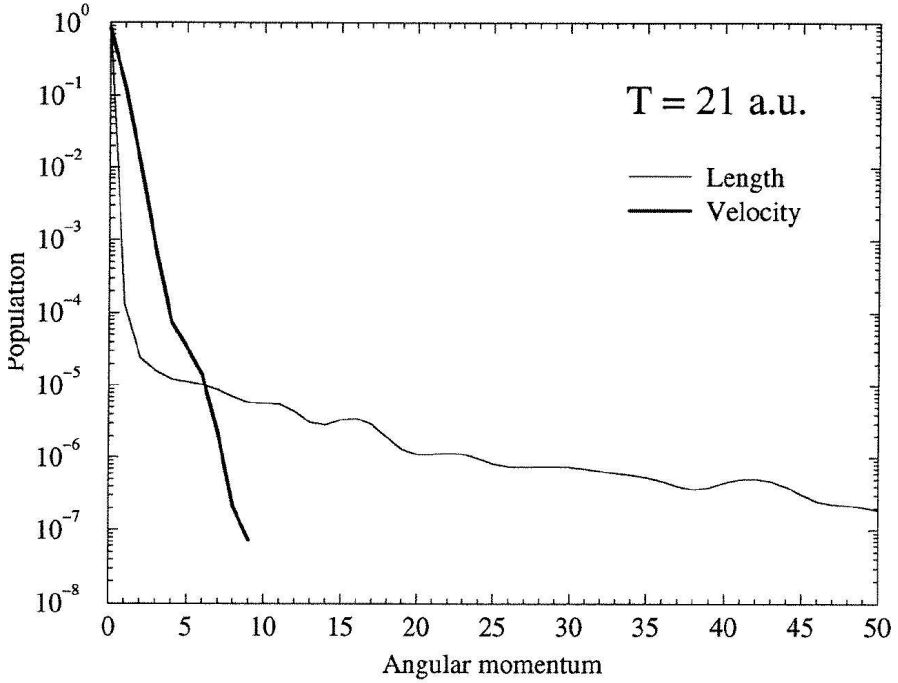
E Cormier† and P Lambropoulos

Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-strasse 1, D-85748 Garching, Germany

Abstract. We discuss the problem of the choice of the gauge in which to represent the electromagnetic field in the non-perturbative time-dependent study of the interaction of atoms with intense laser fields. Even though quantum mechanics is gauge invariant, the velocity gauge is more adopted then the length gauge for dynamical reasons. This property is even more severe when the problem is solved by expanding the wavefunction in spherical harmonics. The point is illustrated through the calculation of the above-threshold ionization photoelectron energy spectrum produced by atomic hydrogen under an intense laser pulse.

Velocity form of dipole operator converges [A LOT] faster





Since there is no observable physics in the partial-wave expansion, different convergence rates are possible!

Difficulties for Higher Intensities

- Results in the length form of the electric dipole operator converge **very s l o w l y.**
- The velocity form is expected to be more appropriate, but it carries its own challenges:
  - The interaction term $\boldsymbol{A} \cdot \boldsymbol{p}$ rather than $\boldsymbol{E} \cdot \boldsymbol{r}$ involves a derivative.
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  - This causes numerical and interpretation problems if $\boldsymbol{A}$ is not zero at the end of the pulse (residual **static $\boldsymbol{E}$ field**)
  - While the mathematics (**gauge invariance**) is fine,
    - **theorists avoid the problem by setting $\boldsymbol{A}$**

      BUT
    - **experimentalists control $\boldsymbol{E}$ rather than $\boldsymbol{A}$!**
- This is a problem of ongoing research.
- **Be careful if somebody tells you that "We solved this problem a long time ago!"**
- Chances are they made their life easy by choosing the parameters in a convenient way.

Problem Solved?

PHYSICAL REVIEW A **74**, 053405 (2006)

Analysis of two-dimensional photoelectron momentum spectra and the effect of the long-range Coulomb potential in single ionization of atoms by intense lasers

Zhangjin Chen,[1] Toru Morishita,[1,2] Anh-Thu Le,[1] M. Wickenhauser,[3] X. M. Tong,[4] and C. D. Lin[1]

[1]*J. R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, Kansas 66506-2604, USA*

[2]*Department of Applied Physics and Chemistry, The University of Electro-Communications, 1-5-1 Chofu-ga-oka, Chofu-shi, Tokyo 182-8585, Japan*

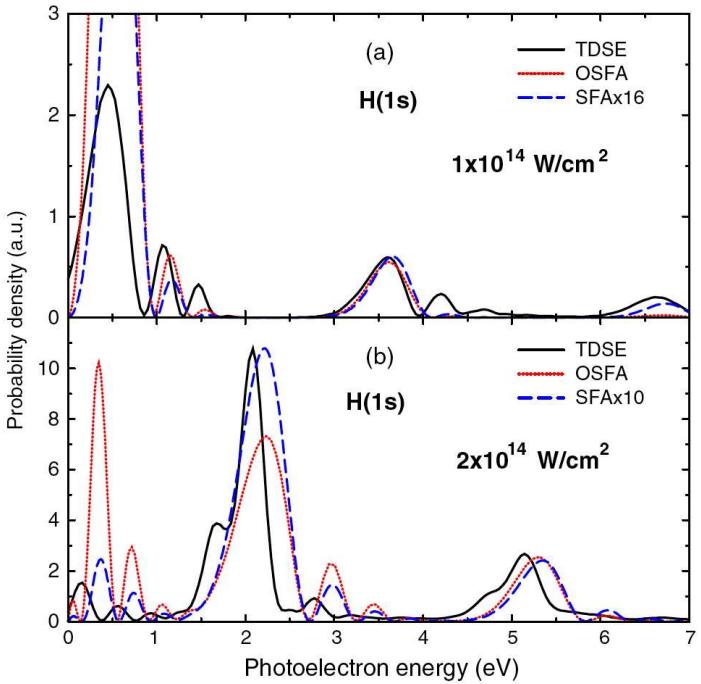
[3]*Institute for Theoretical Physics, Vienna University of Technology, A-1040 Vienna, Austria*

[4]*Institute of Materials Science, Graduate School of Pure and Applied Science, and Center for Computational Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8577, Japan*

(Received 26 September 2006; published 6 November 2006)

Two-dimensional (2D) electron momentum distributions and energy spectra for multiphoton ionization of atoms by intense laser pulses, calculated by solving the time-dependent Schrödinger equation (TDSE) for different wavelengths and intensities, are compared to those predicted by the strong-field approximation (SFA). It is shown that the momentum spectra at low energies between the TDSE and SFA are quite different and the differences arise largely from the absence of a long-range Coulomb interaction in the SFA. We further found that the low-energy 2D momentum spectra from the TDSE exhibit ubiquitous fanlike features where the number of stripes is due to a single dominant angular momentum of the low-energy electron. The specific dominant angular momentum in turn has been found to be decided by the minimum number of photons needed to ionize the atom only. The electron momentum spectra predicted by models modified from the SFA are also examined and found to lack the fanlike features as in the SFA.

Not really – the results are for 400 nm!



CHEN *et al.*

PHYSICAL REVIEW A **74**, 053405 (2006)

FIG. 10. (Color online) Comparison of photoelectron spectra of H ($1s$) by a 10-fs (FWHM) laser pulse with wavelength of 400 nm at peak intensity of (a) 1.0×10^{14} W/cm^2 and (b) 2.0×10^{14} W/cm^2, calculated by different theoretical models. Note that the results from the SFA have been normalized, as shown on the labels. See text.

This problem is A LOT easier for 400 nm than for 800 nm!

A very recent paper

PHYSICAL REVIEW A **80**, 033409 (2009)

Explicit time-propagation method to treat the dynamics of driven complex systems

Javier Madroñero
Laboratoire de Physique Atomique, Moléculaire et Optique (PAMO), Université catholique de Louvain, 2 chemin du Cyclotron, 1348 Louvain-la-Neuve, Belgium
and Physik Department, Technische Universität München, James-Franck-Straße, 85747 Garching, Germany

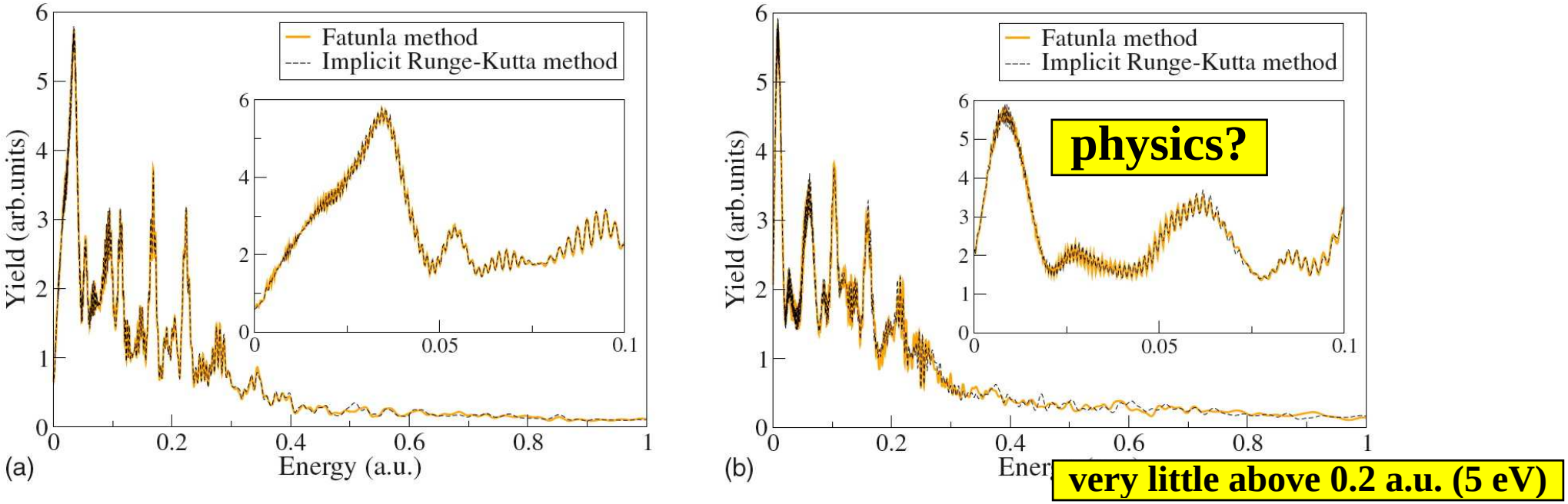
Bernard Piraux
Laboratoire de Physique Atomique, Moléculaire et Optique (PAMO), Université catholique de Louvain, 2 chemin du Cyclotron, 1348 Louvain-la-Neuve, Belgium

(Received 15 June 2009; revised manuscript received 27 July 2009; published 9 September 2009)

We describe the efficient implementation of an *explicit* method to solve systems of stiff differential equations either on a grid or within a spectral approach. This method is based on an ansatz that approximates the solution. This ansatz depends on stiffness parameters that are shown to be related to the eigenfrequencies of the system. The accuracy and the performance of the method are tested in three different cases. First, we treat a highly stiff single differential equation, where explicit schemes converge rather slowly. Then, we solve the stationary Schrödinger equation associated to the quantum reflection of an ultracold atom by a surface. Finally, we consider the interaction of atomic hydrogen with a strong low-frequency laser pulse whose duration is of the order of 25 fs. We focus on the calculation of the above-threshold ionization electron spectrum, a problem which, under such realistic physical conditions, is computationally very demanding.

10-cycle pulse; 800 nm; I_0 = 4 and 6 x 10^{14}W/cm^2 —TOUGH!

Lots of structure – plus some wiggles …

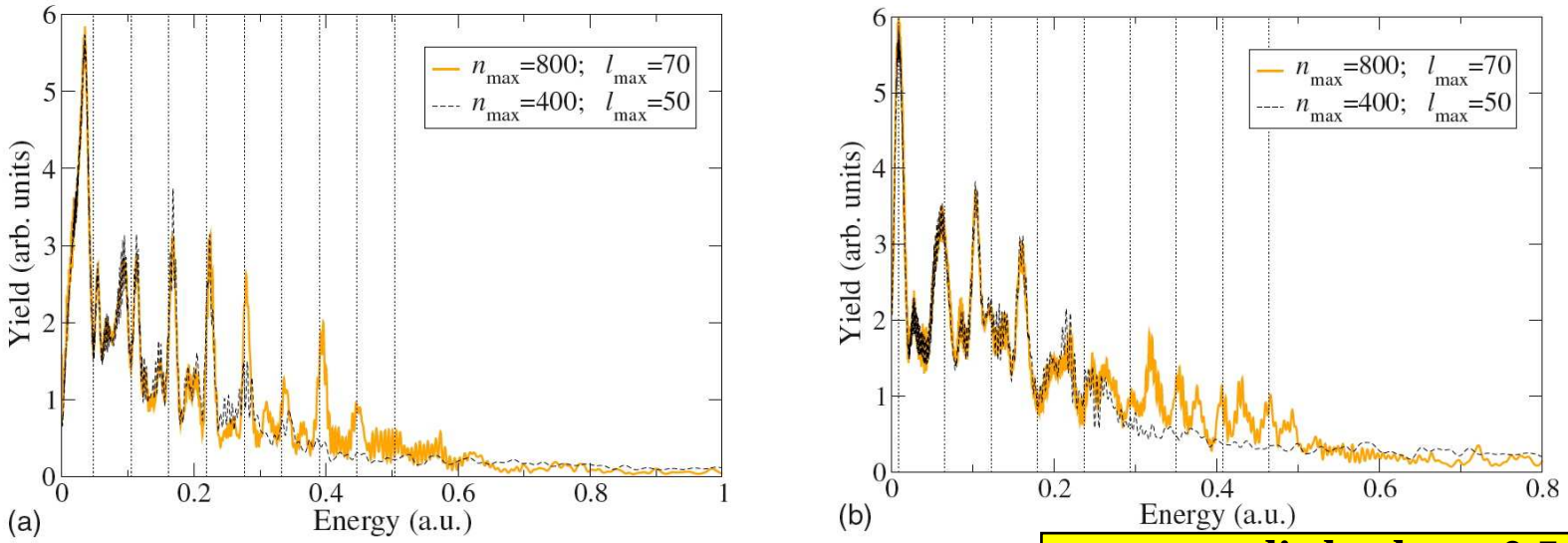


very little above 0.2 a.u. (5 eV)

FIG. 5. (Color online) Energy spectrum for hydrogen driven by a field with frequency ω=0.057 a.u. and intensities I=4 $\times 10^{14}$ W/cm^2 (left) and I=6$\times 10^{14}$ W/cm^2 (right). Results obtained for several n_{sup}=400, ℓ_{max}=50, and α=2.5 (κ=1/α=0.4. The dashed curve was obtained using a diagonally implicit RK propagator. The solid curve was obtained using Fatunla's method.

10-cycle pulse; 800 nm; I_0 = 4 and 6 x 10^{14}W/cm^2 —TOUGH!

Convergence problems ...
... number of basis functions and/or angular momenta?



now very little above 0.5 a.u. (14 eV)

FIG. 6. (Color online) Electron energy distributions for single ionization of hydrogen by 25 fs pulses at $I=4\times10^{14}$ W/cm^2 (left) and $I=6\times10^{14}$ W/cm^2 (right) . Convergence of the results as a function of the basis size: $n_{max}=800$ and $\ell_{max}=70$ (solid line); $n_{max}=400$ and $\ell_{max}=50$ (dashed line). The dotted vertical lines denote the expected position of the peaks according to Eq. (41).

The Matrix Iteration Method

PHYSICAL REVIEW A VOLUME 60, NUMBER 4 OCTOBER 1999

Numerical solution of time-dependent Schrödinger equation for multiphoton processes: A matrix iterative method

M. Nurhuda[1,2] and F. H. M. Faisal[1,*]
[1]*Fakultät für Physik, Universität Bielefeld, Postfach 100131, D-33501 Bielefeld, Germany*
[2]*Physics Department, Brawijaya University, Malang 65144, Indonesia*
(Received 30 March 1999)

An implicit algorithm for integration of the three-dimensional (3D) time-dependent Schrödinger equation of an atomic system interacting with intense laser pulses is developed. It is based on a matrix iteration of the Crank-Nicholson approximant to the short-time propagator using the *total* Hamiltonian (unsplit) of the system directly. To test the method, 3D Schrödinger wave-packet propagation is carried out, and so-called above-threshold ionization and high-harmonic generation spectra for atomic hydrogen irradiated by intense laser pulses are obtained. They are also compared with that obtained using the popular split-operator method. The present algorithm is shown to provide an alternative to the the split-operator method, and proves to be more efficient in all the cases studied here. A procedure for optimizing the maximum grid size is also given, and its usefulness is illustrated. [S1050-2947(99)06409-4]

- Basic idea: **approximation to $e^{-iH(\boldsymbol{r},t)\Delta t}$**

$$\Psi(\boldsymbol{r}, t+\Delta t) \approx [1 - i\hat{H}\Delta t/2][(1 + i\hat{H}\Delta t/2)]^{-1}\Psi(\boldsymbol{r}, t)$$

  - Split $(1 + i\hat{H}\Delta t/2)$ into diagonal (O_D) and non-diagonal (O_{ND}) parts.
  - Do a series expansion of the inverse ("denominator") in terms of "O_{ND}/O_D".
- **It works very well ($\rightarrow$ examples), but may have been forgotten for a decade.**

approximating $e^{-iH(\boldsymbol{r},t)\Delta t}$ is actually THE PROBLEM!

We know that there are other ways ...
I am sure your favorite one is sooo... much better!

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Nineteen Dubious Ways to Compute the Exponential of a Matrix, Twenty-Five Years Later

Cleve Moler† Charles Van Loan‡

Abstract. In principle, the exponential of a matrix could be computed in many ways. Methods involving approximation theory, differential equations, the matrix eigenvalues, and the matrix characteristic polynomial have been proposed. **In practice, consideration of computational stability and efficiency indicates that some of the methods are preferable to others, but that none are completely satisfactory.**

10. Method 20: Krylov space methods. If we were to revise thoroughly *Nineteen Ways*, we would have to revise the title because Krylov space methods constitute a twentieth approach!

Examples from Recent Work

PHYSICAL REVIEW A **81**, 043408 (2010)

Ionization of atomic hydrogen in strong infrared laser fields

Alexei N. Grum-Grzhimailo,[*] Brant Abeln,[†] Klaus Bartschat,[‡] and Daniel Weflen[§]
Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA

Timothy Urness[‖]
Department of Mathematics and Computer Science, Drake University, Des Moines, Iowa 50311, USA
(Received 27 January 2010; published 14 April 2010)

We have used the matrix iteration method of Nurhuda and Faisal [Phys. Rev. A **60**, 3125 (1999)] to treat ionization of atomic hydrogen by a strong laser pulse. After testing our predictions against a variety of previous calculations, we present ejected-electron spectra as well as angular distributions for few-cycle infrared laser pulses with peak intensities of up to 10^{15} W/cm^2. It is shown that the convergence of the results with the number of partial waves is a serious issue, which can be managed in a satisfactory way by using the velocity form of the electric dipole operator in connection with an efficient time-propagation scheme.

DOI: 10.1103/PhysRevA.81.043408 PACS number(s): 32.80.Rm

4-cycle pulse; 152 nm; $I_0 = 10^{16}$W/cm^2

A Simple Test Case

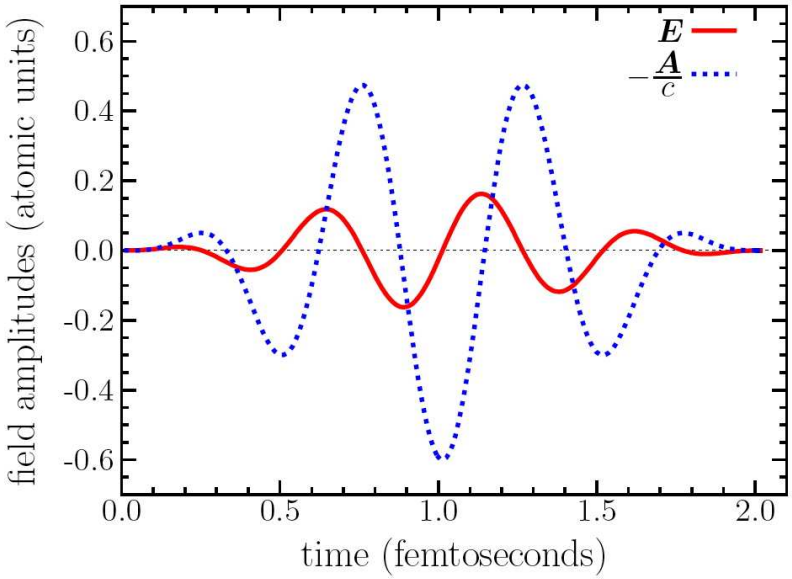


FIG. 1: (Color online) Electric field $\boldsymbol{E}(t)$ and vector potential $-\boldsymbol{A}(t)/c$ for a 4-cycle laser pulse with $\sin^2$ envelope, peak intensity of 1×10^{15} W/cm^2, and a central frequency of 0.3 a.u., corresponding to a wavelength of 152 nm.

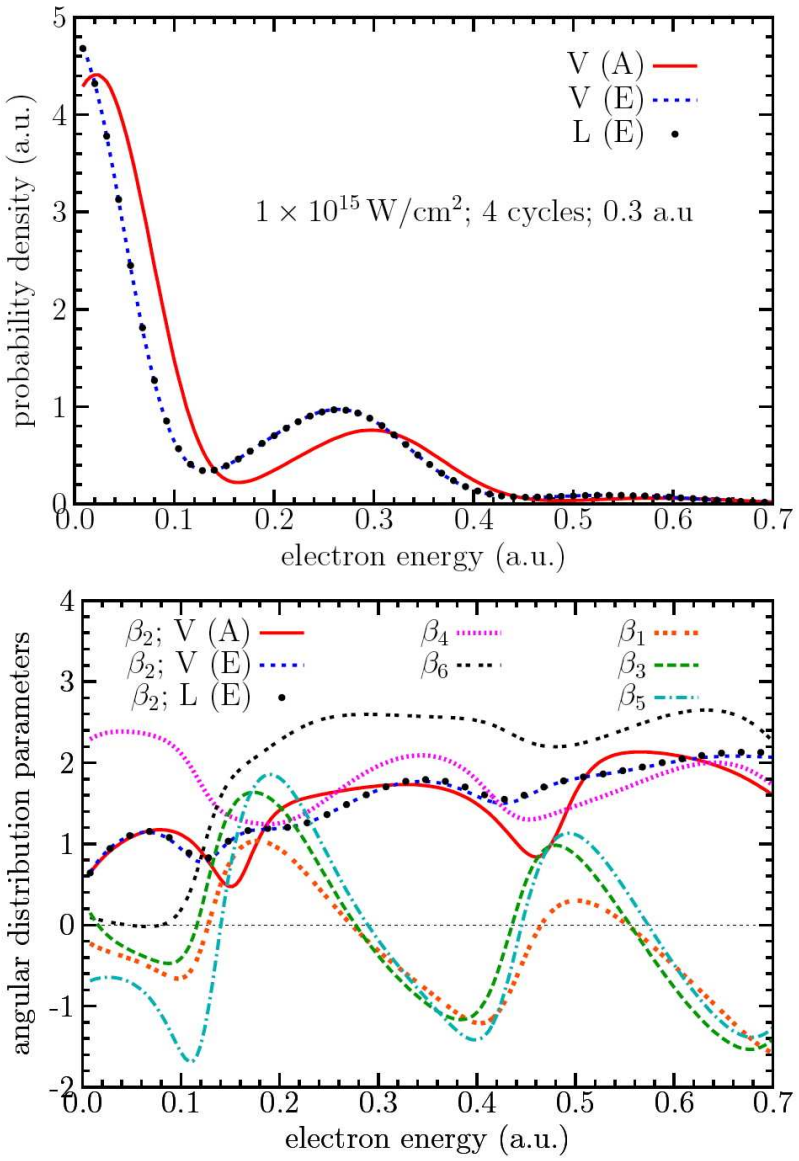
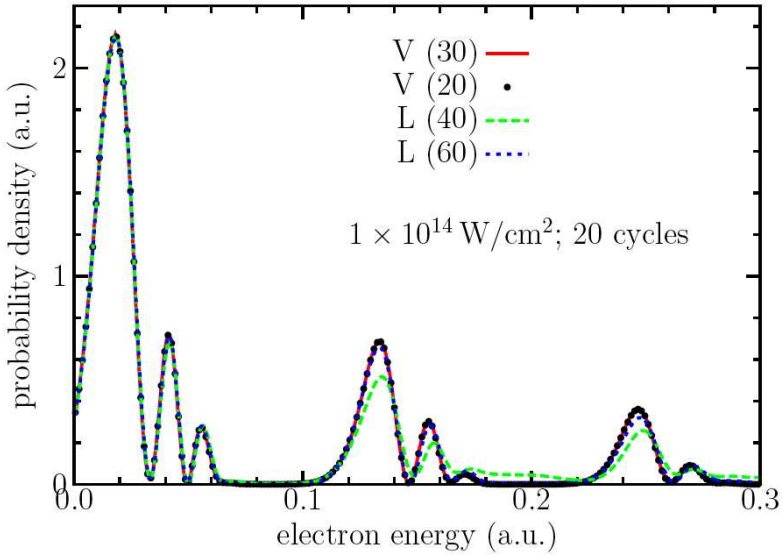


FIG. 2: (Color online) Photoelectron spectrum (top) and several asymmetry parameters (bottom) for ionization of atomic hydrogen in a 4-cycle laser pulse with $\sin^2$ envelope, peak intensity of 1×10^{15} W/cm^2, and a central frequency of 0.3 a.u., corresponding to a wavelength of 152 nm. The calculations were performed with angular momenta up to $\ell_{max} = 20$ in the length (L) and velocity (V) form of the electric dipole operator. Either the electric field ($\boldsymbol{E}$) or the vector potential ($\boldsymbol{A}$) were set to the $\sin^2$ envelope for the spectrum and the parameter β_2. For all other parameters, the $\sin^2$ envelope was used for the vector potential (see text for details).

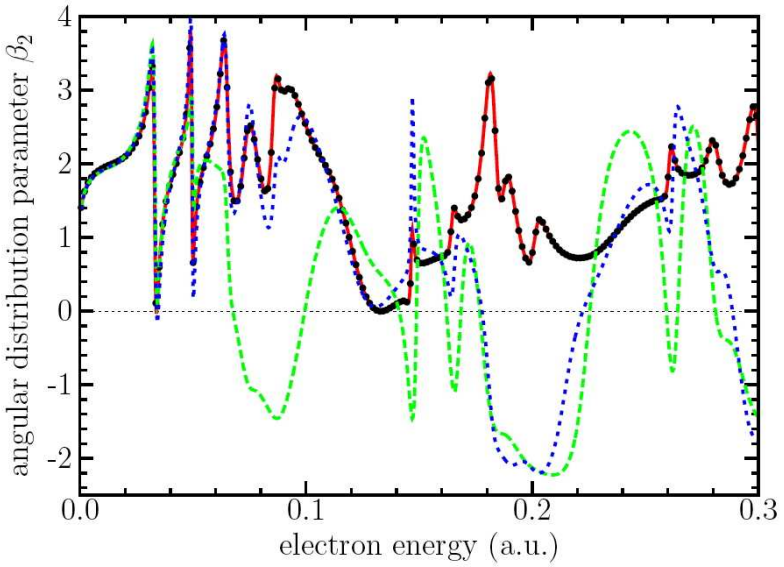
It matters whether we set *E* or *A* for a short pulse!

20-cycle pulse; 400 nm; $I_0 = 10^{14}$W/cm^2

A Little More Difficult ...



This is the case that C.D. Lin's group solved.



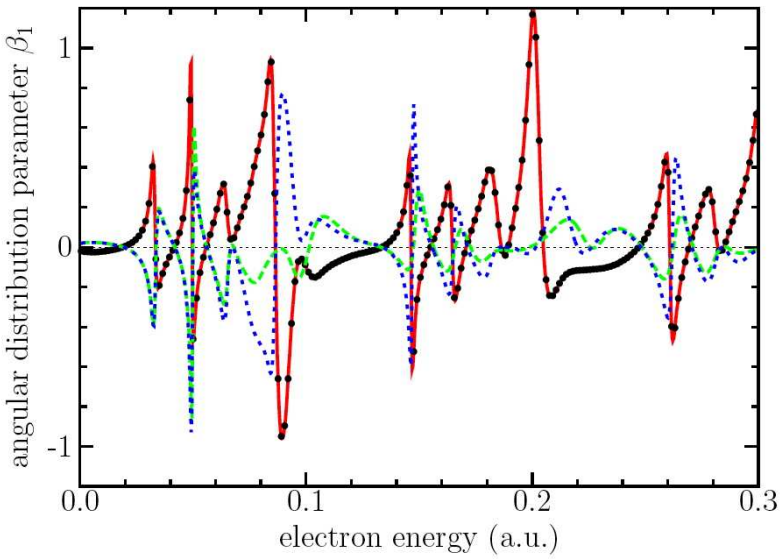


FIG. 4: (Color online) Photoelectron spectrum (top) and asymmetry parameters β_2 (center) and β_1 (bottom) for ionization of atomic hydrogen in a 20-cycle laser pulse with $\sin^2$ envelope for the electric field, peak intensity of 1×10^{14} W/cm^2, and a central frequency of 0.114 a.u., corresponding to a wavelength of 390 nm. The calculations were performed with the length (L) and velocity (V) forms of the electric dipole operator for angular momenta up to the values ℓ_{max} indicated in the parentheses.

One can still get converged results in the length form.

Recall from Madronero and Piraux

Lots of structure – plus some wiggles ...

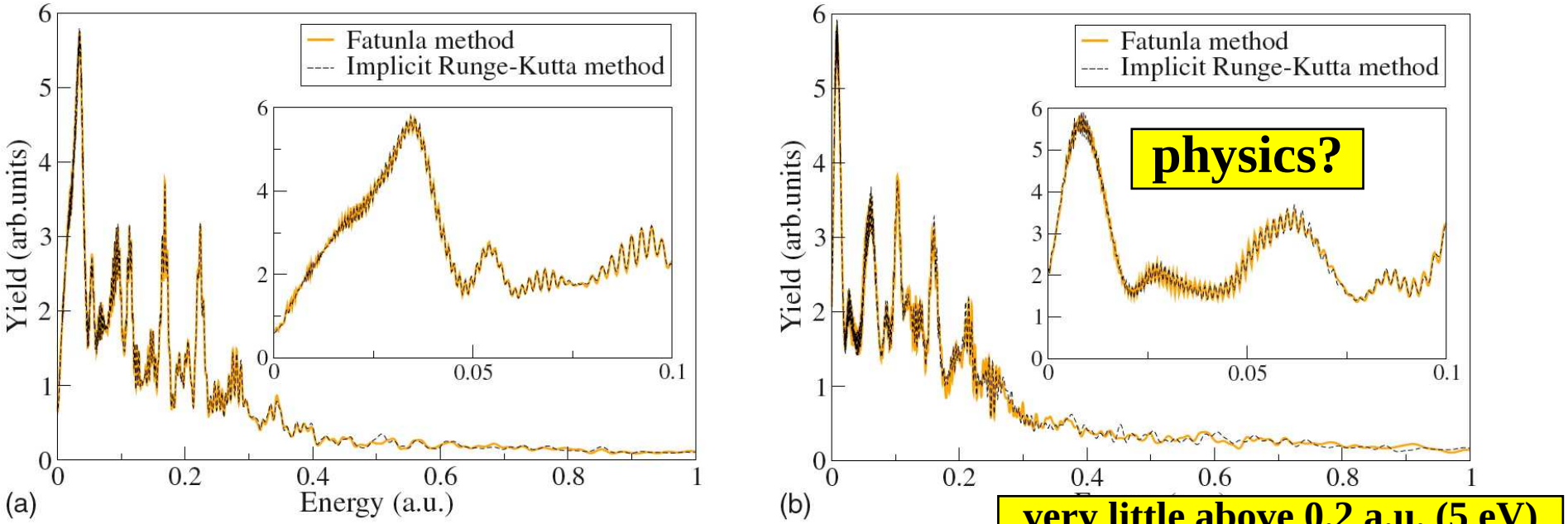
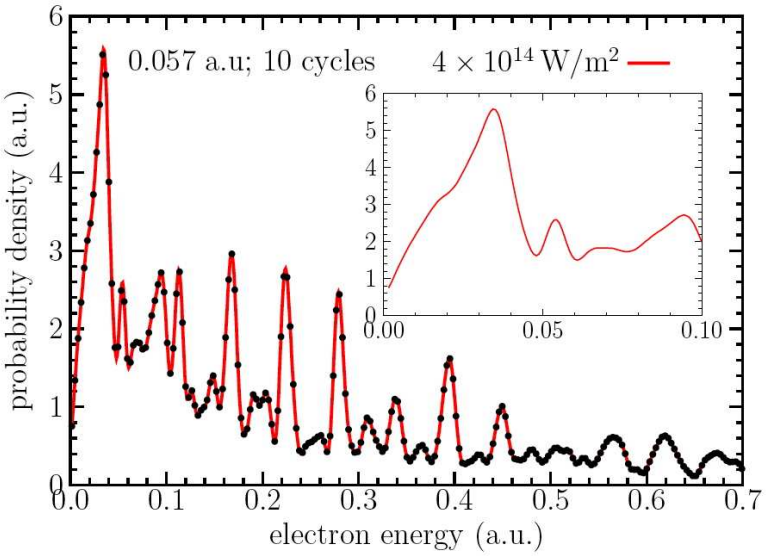
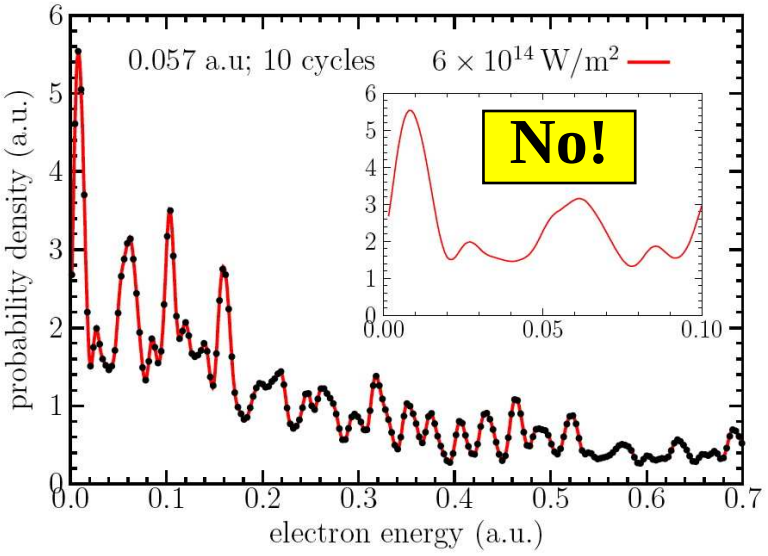


FIG. 5. (Color online) Energy spectrum for hydrogen driven by a field with frequency $\omega=0.057$ a.u. and intensities $I=4\times10^{14}$ W/cm^2 (top) and $I=6\times10^{14}$ W/cm^2 (bottom). Results obtained for several $n_{sup}=400$, $\ell_{max}=50$, and $\alpha=2.5$ ($\kappa=1/\alpha=0.4$. The dashed curve was obtained using a diagonally implicit RK propagator. The solid curve was obtained using Fatunla's method.

10-cycle pulse; 800 nm; I_0 = 4,6,10 x 10^{14}W/cm^2

The REAL TEST





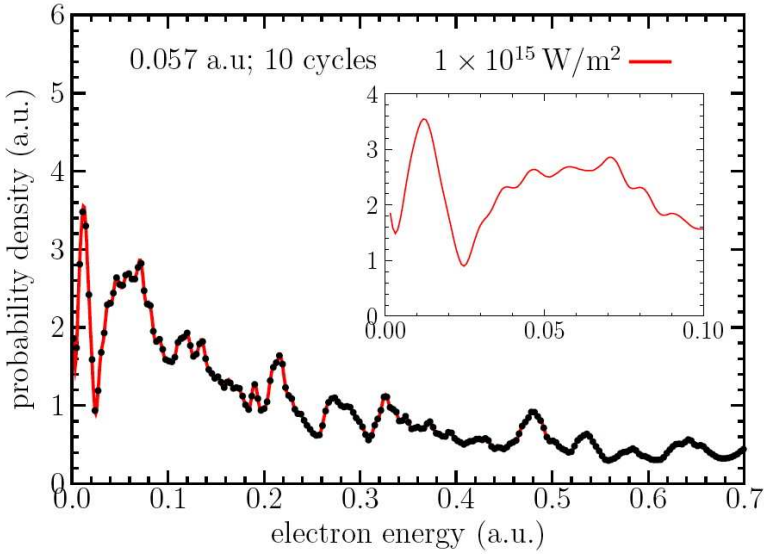
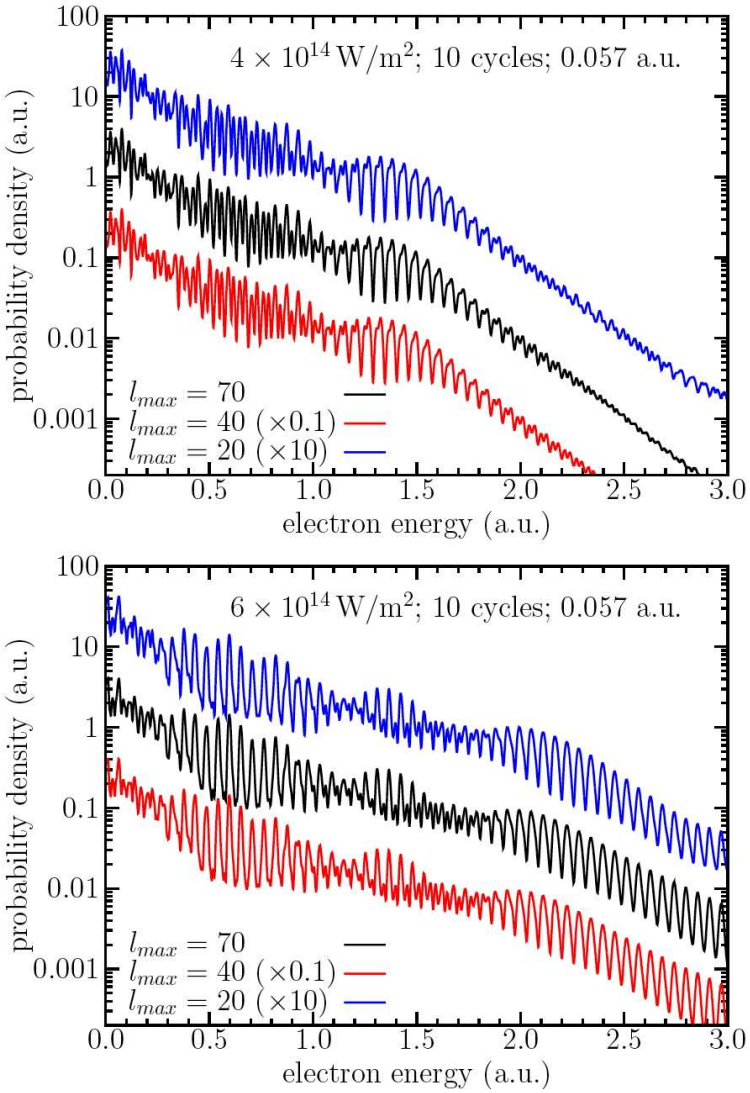


FIG. 5: (Color online) Photoelectron spectrum for ionization of atomic hydrogen in a 10-cycle laser pulse with $\sin^2$ envelope for the vector potential, peak intensities of 4×10^{14} W/cm^2 (top), 6×10^{14} W/cm^2 (center), and 1×10^{15} W/cm^2 (bottom), and a central frequency of 0.057 a.u., corresponding to a wavelength of 780 nm. The calculations were performed with the velocity form of the electric dipole operator for angular momenta up to $\ell_{max} = 70$ (solid line) and $\ell_{max} = 50$ (dots). The inserts show the low-energy regime on an extended scale.

lots of structure up to 0.7 a.u. (20 eV) ...

It works! No unphysical wiggles!

Pushing Further ...



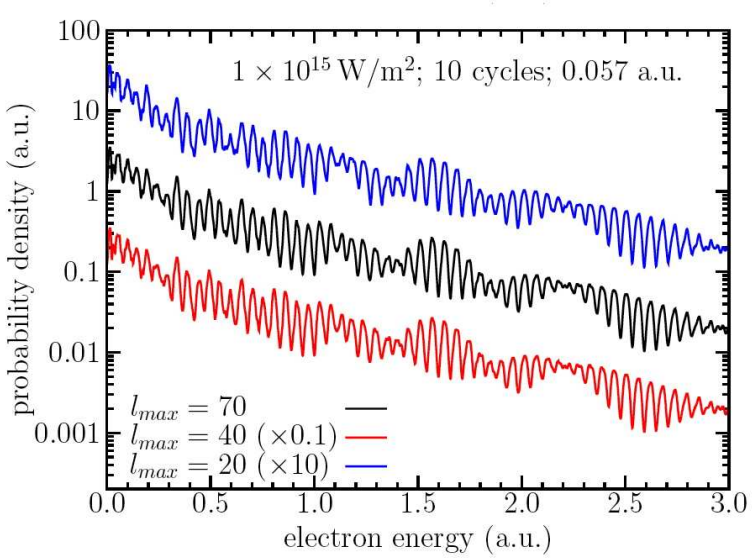


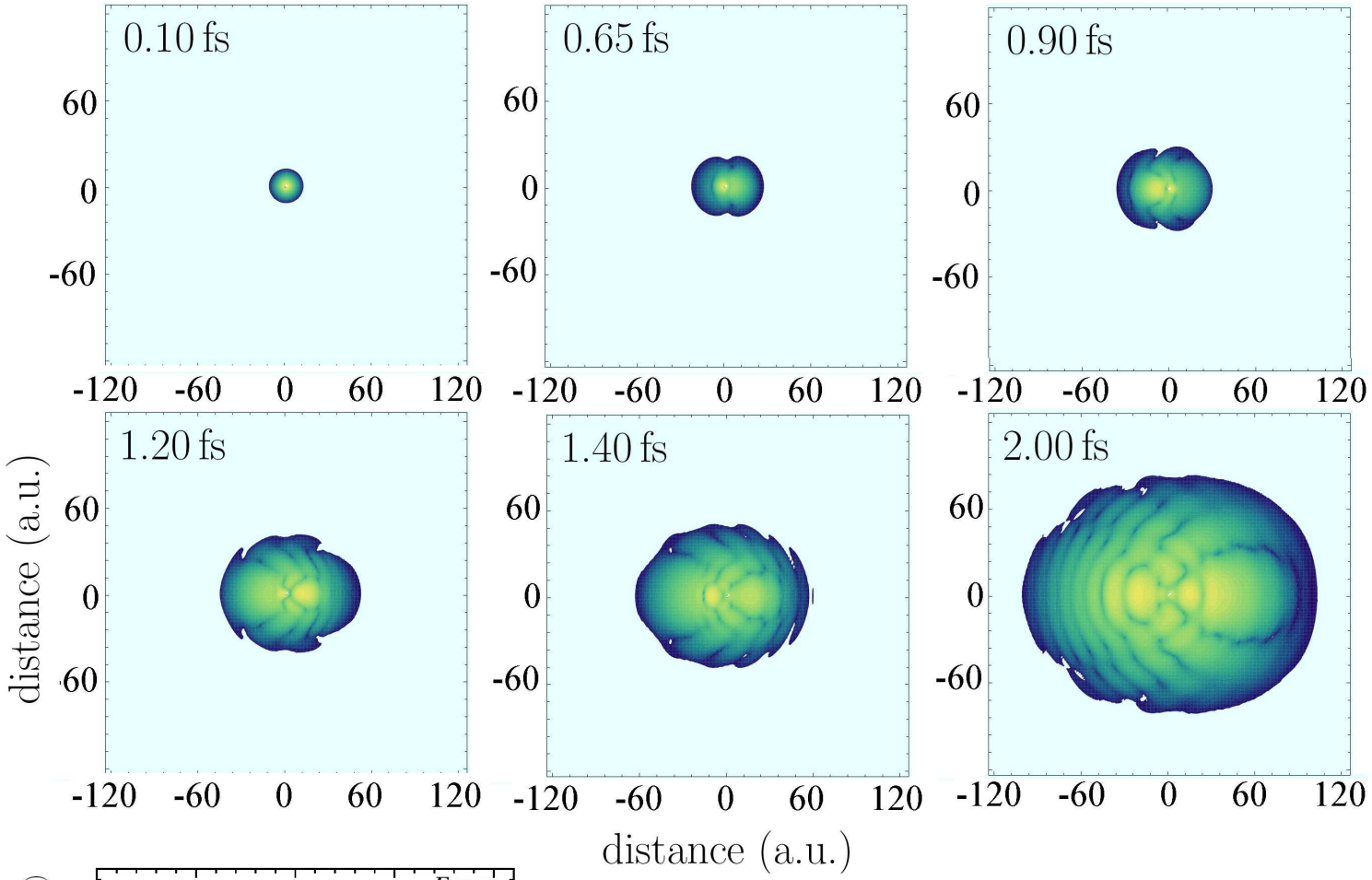
FIG. 6: (Color online) Photoelectron spectrum for ionization of atomic hydrogen in a 10-cycle laser pulse with $\sin^2$ envelope for the electric field, peak intensities of $4 \times 10^{14}\,\mathrm{W/cm^2}$ (top), $6 \times 10^{14}\,\mathrm{W/cm^2}$ (center), and $1 \times 10^{15}\,\mathrm{W/cm^2}$ (bottom), and a central frequency of 0.057 a.u., corresponding to a wavelength of 780 nm. The calculations were performed with the velocity form of the electric dipole operator for angular momenta up to $\ell_{max} = 70$, 40, and 20. The results for $\ell_{max} = 40$ (20) were multiplied by 0.1 (10) to make them distinguishable from the $\ell_{max} = 70$ results.

... and beyond (> 80 eV)

Excellent convergence in the velocity form :):):)

Visualization of the Results

Probability density to find the electron at different times in the pulse



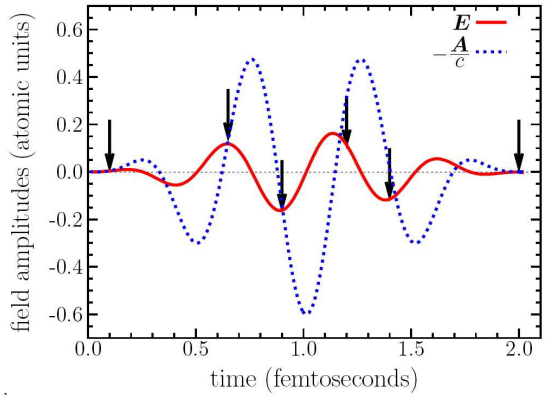
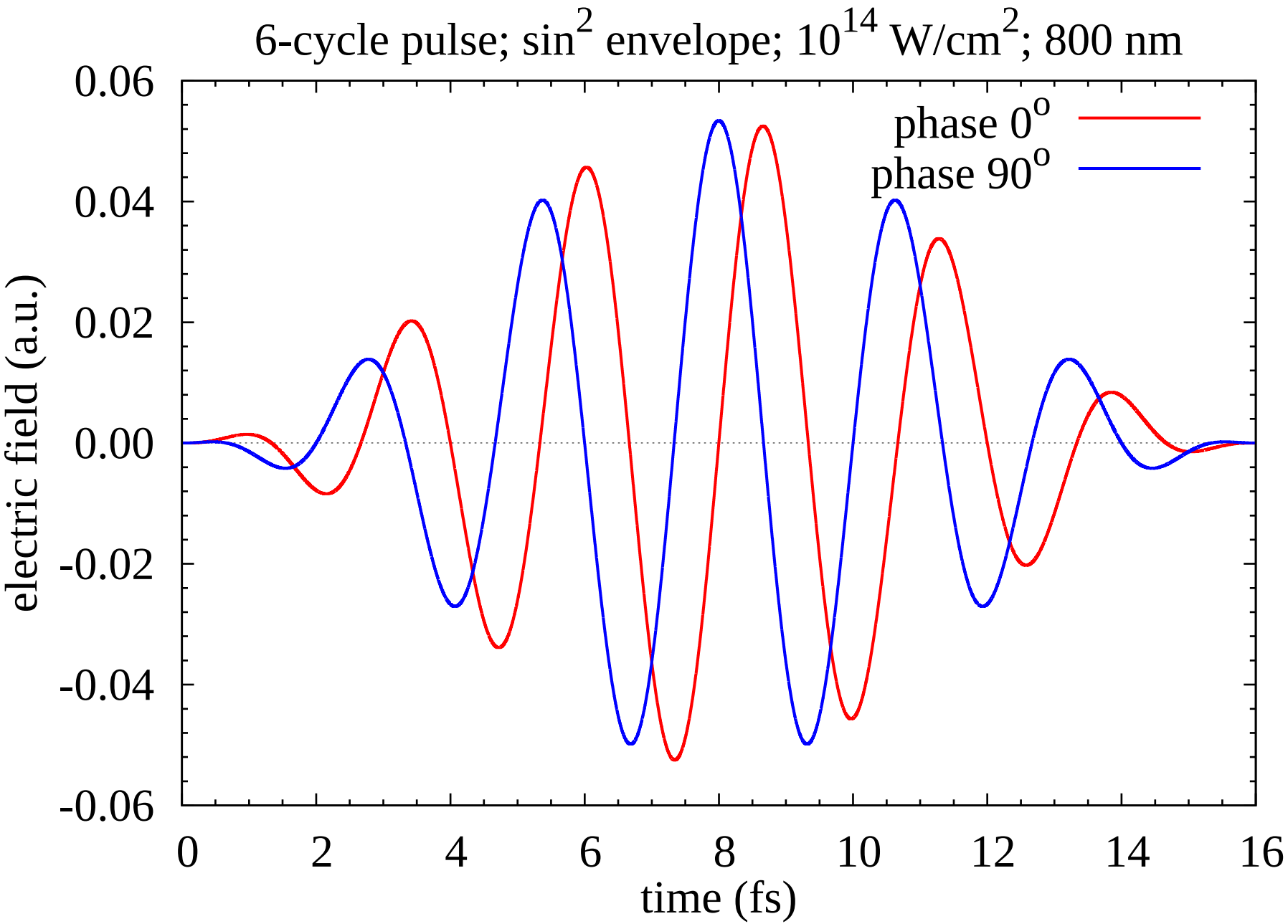
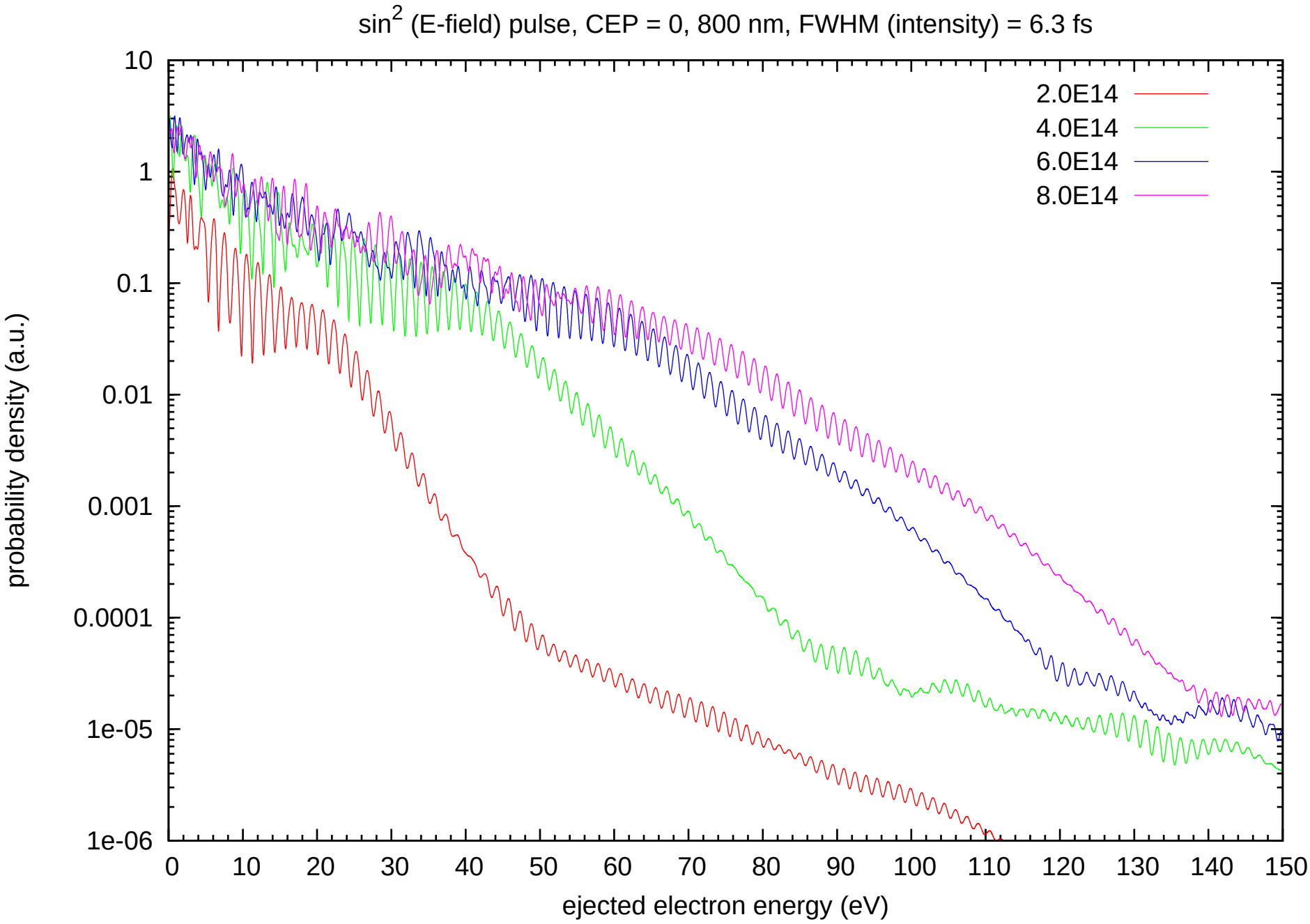


FIG. 9: (Color online) Electric field $\boldsymbol{E}(t)$ and vector potential $-\boldsymbol{A}(t)/c$ for a 4-cycle laser pulse with $\sin^2$ envelope, peak intensity of 1×10^{15} W/cm^2, and a central frequency of 0.3 a.u., corresponding to a wavelength of 152 nm. This is the same as Fig. 1, with additional arrows marking the times for which the electron density is shown in the panels below. The axes indicate the spatial extension of the packet plotted.

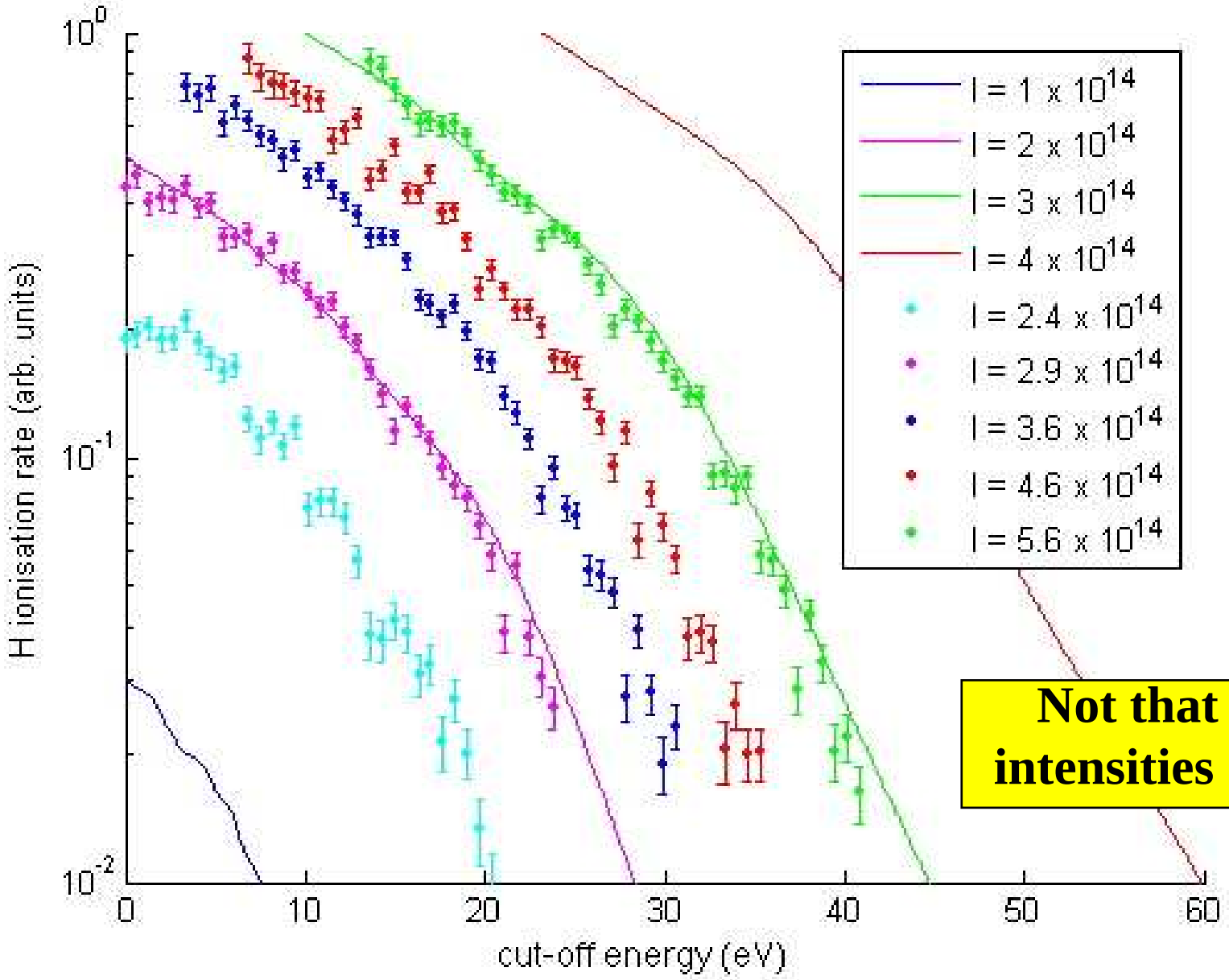
Recall: Example of a short pulse (Griffith experiment)



Predictions for the "cut-off spectrum" in the experiment at Griffith

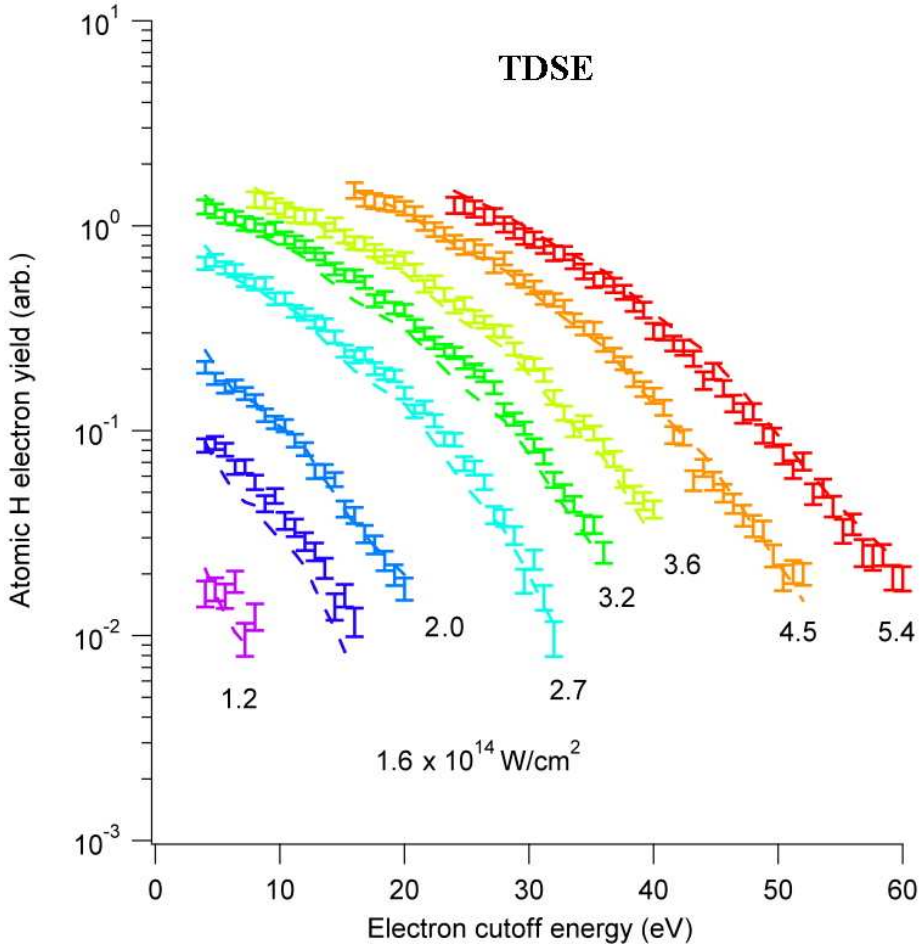


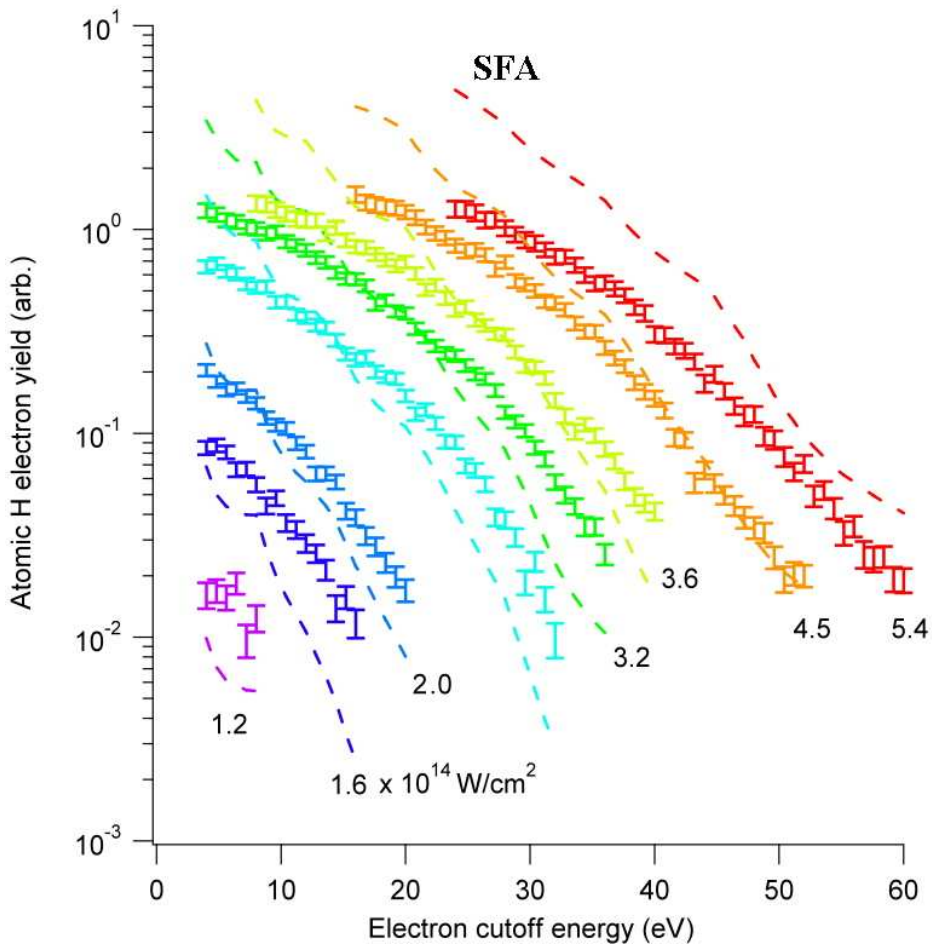
First Comparison with Experiment



Not that great – the intensities don't match!

Revised Comparison





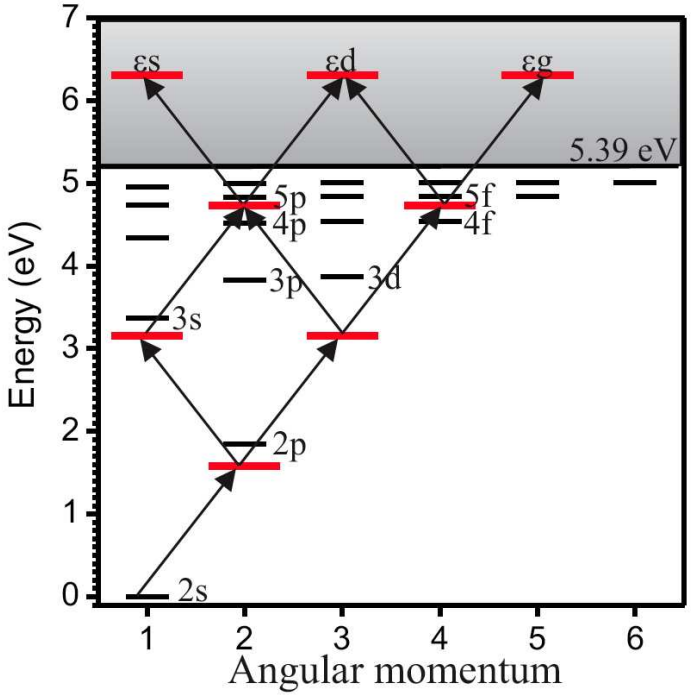
- The **remaining discrepancies** are most likely due to:
  - actual pulse envelope is not really $\sin^2$
  - FWHM not exactly known
  - intensity profile not exactly known
  - not all electrons are detected
  - likelihood of saturation effects

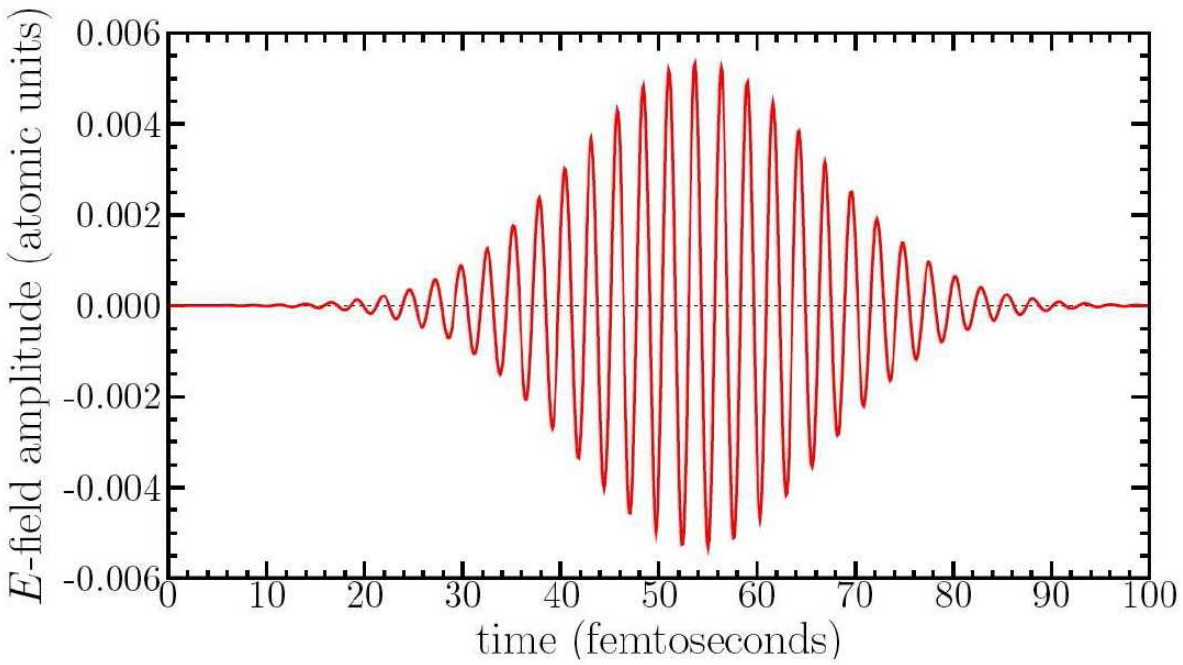
Much better after fixing (some of) the experimental kinks!

A Quasi-One-Electron System

Multi-Photon Ionization of Li (2s)

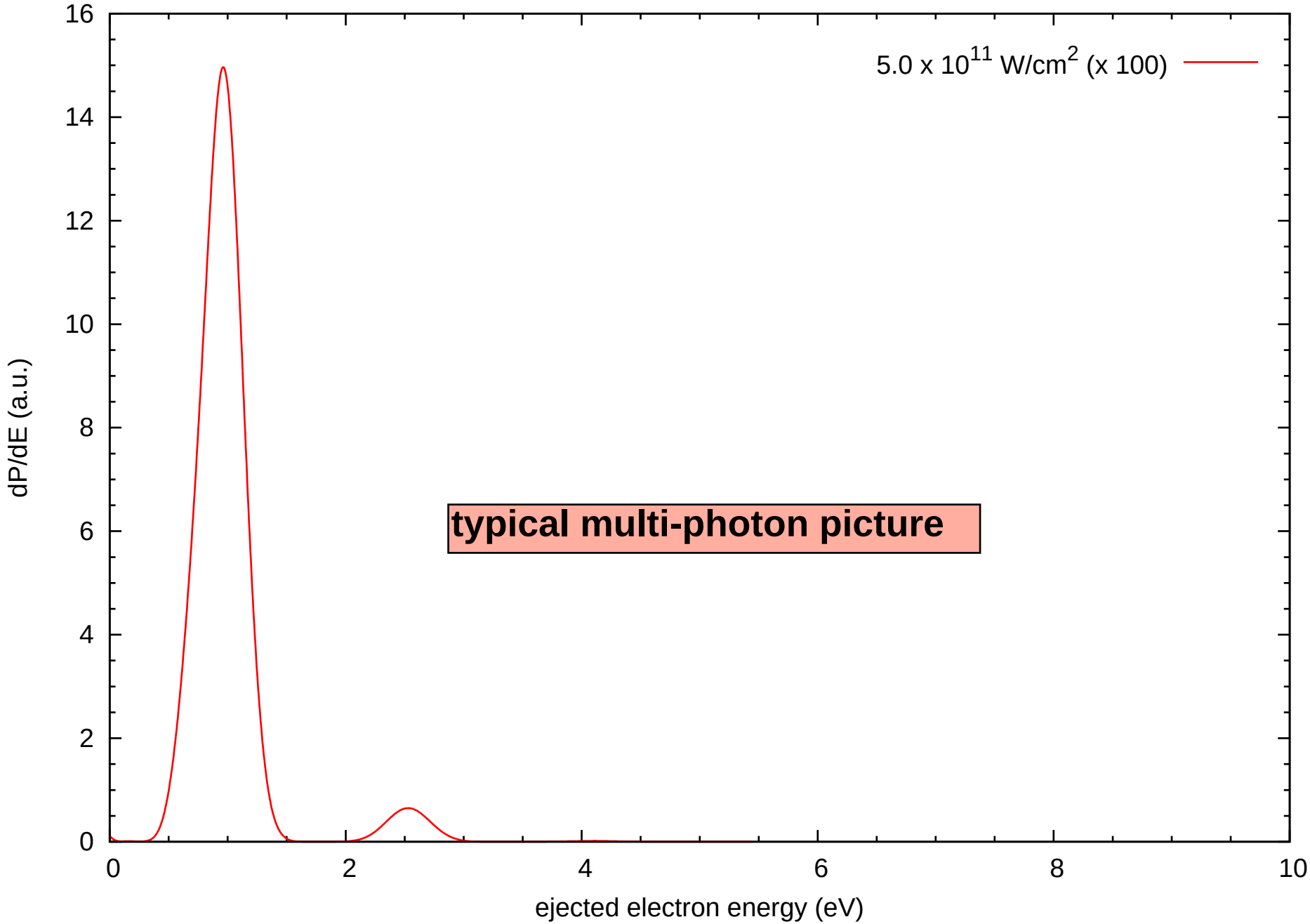
Experiment: Heidelberg Group



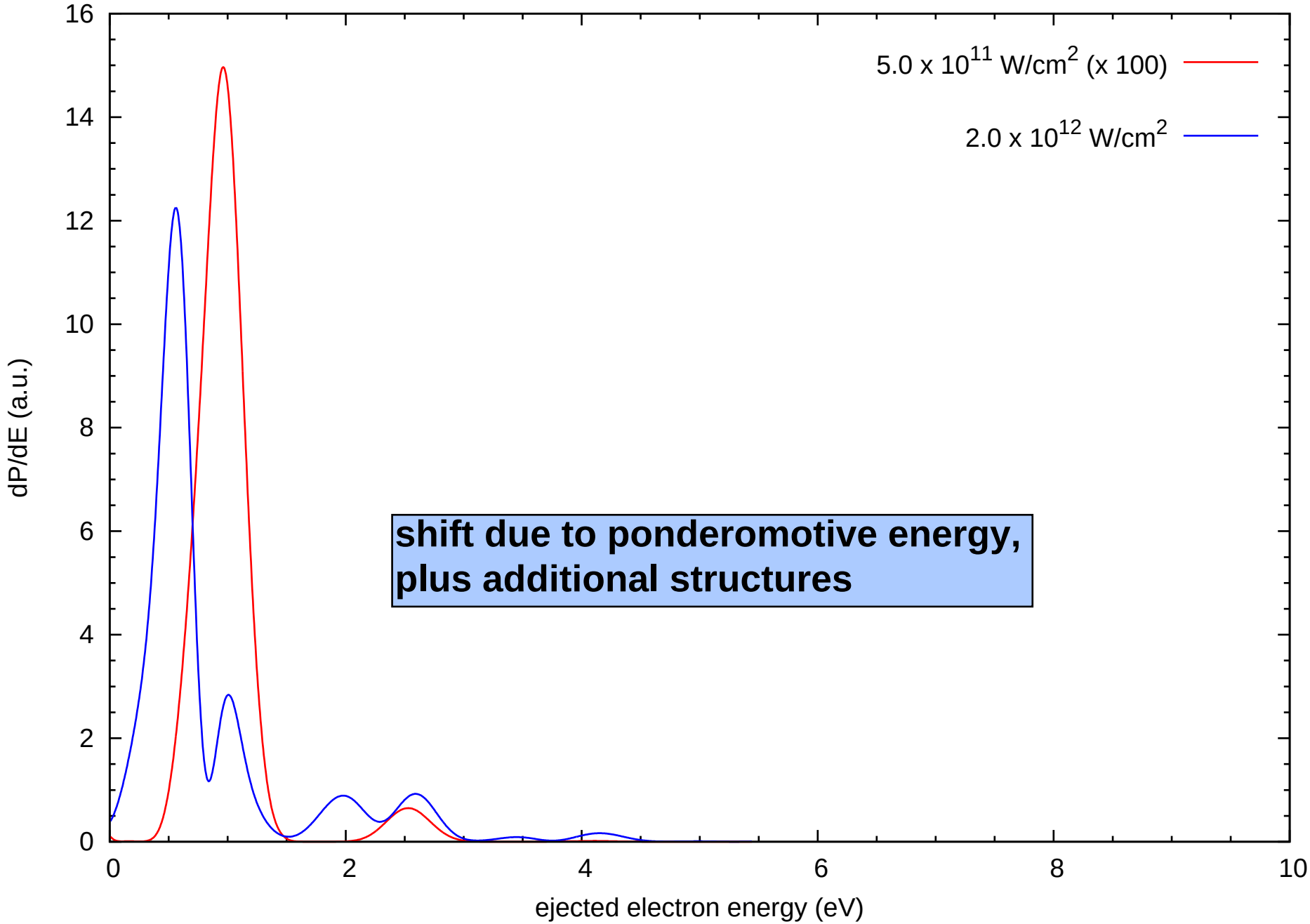


- **Practical issues:**
  - Pulse does not look that nice.
  - Intensity varies over the interaction region (→ focal averaging).
  - The pulse has an energy width (→ increased chance to hit a "stepping stone").
  - Details of the Li structure may have an effect. **They do!**
- As a result, **the direct comparison with experiment is by no means straightforward!**

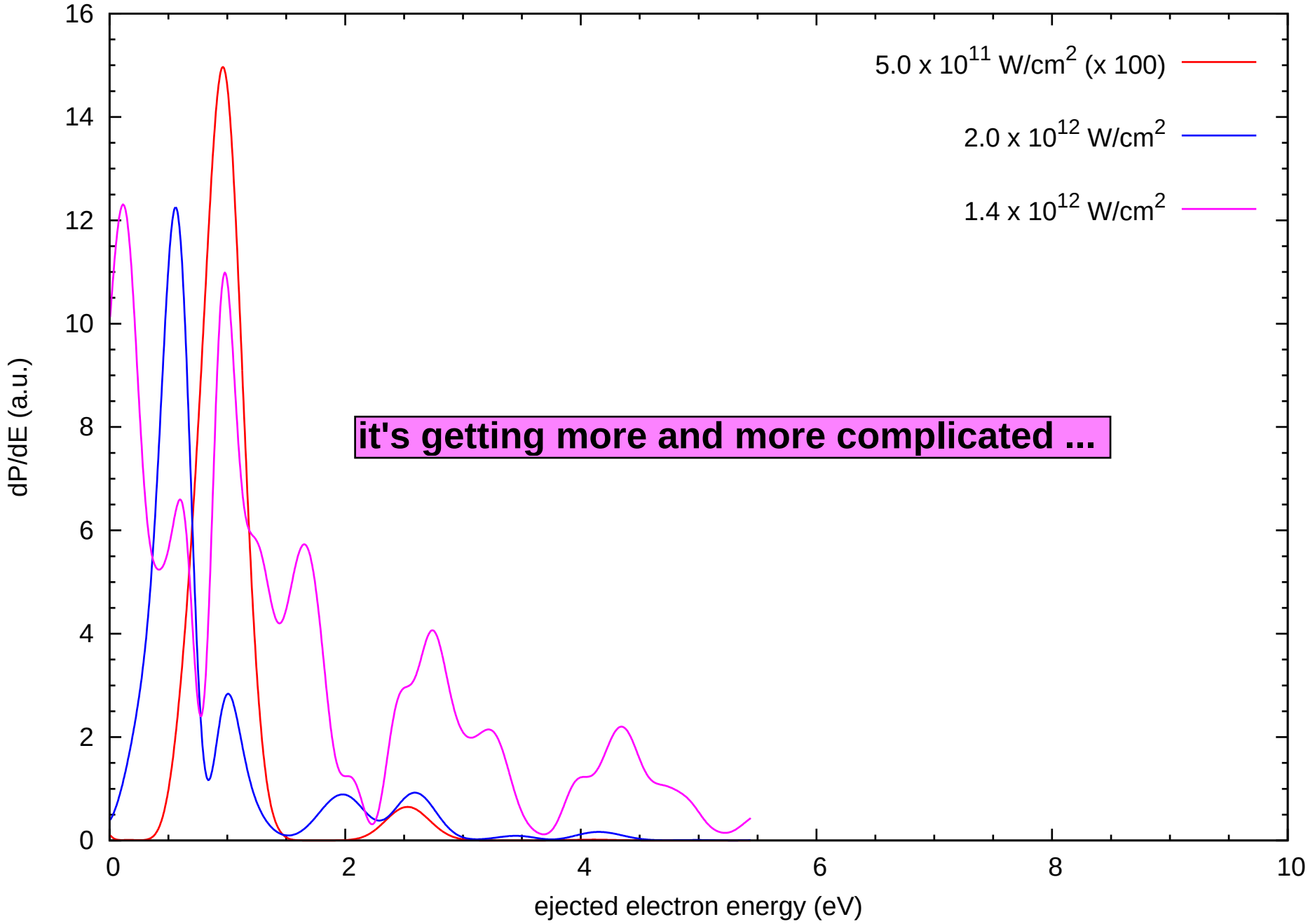
Ejected Electron Spectra



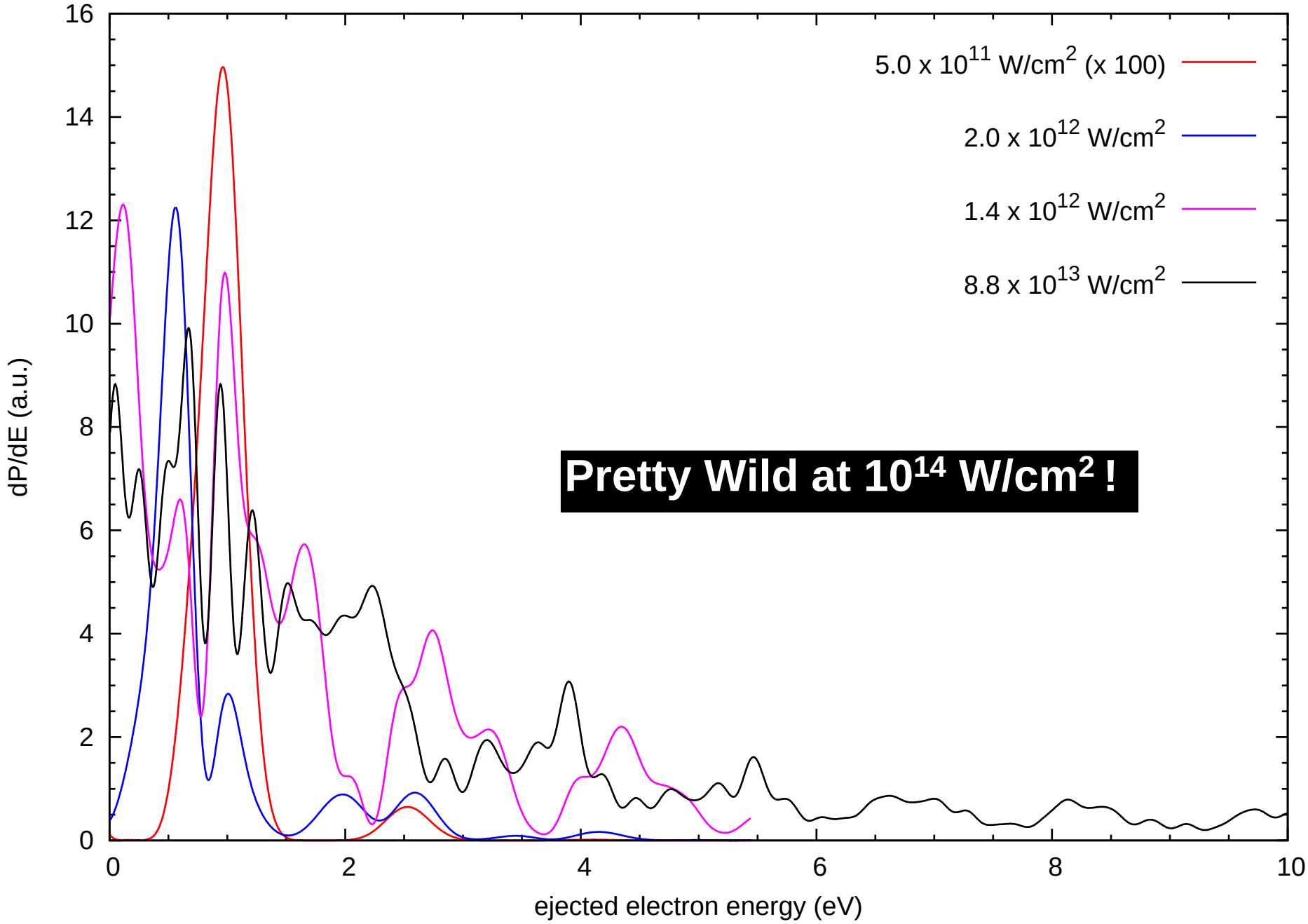
Ejected Electron Spectra

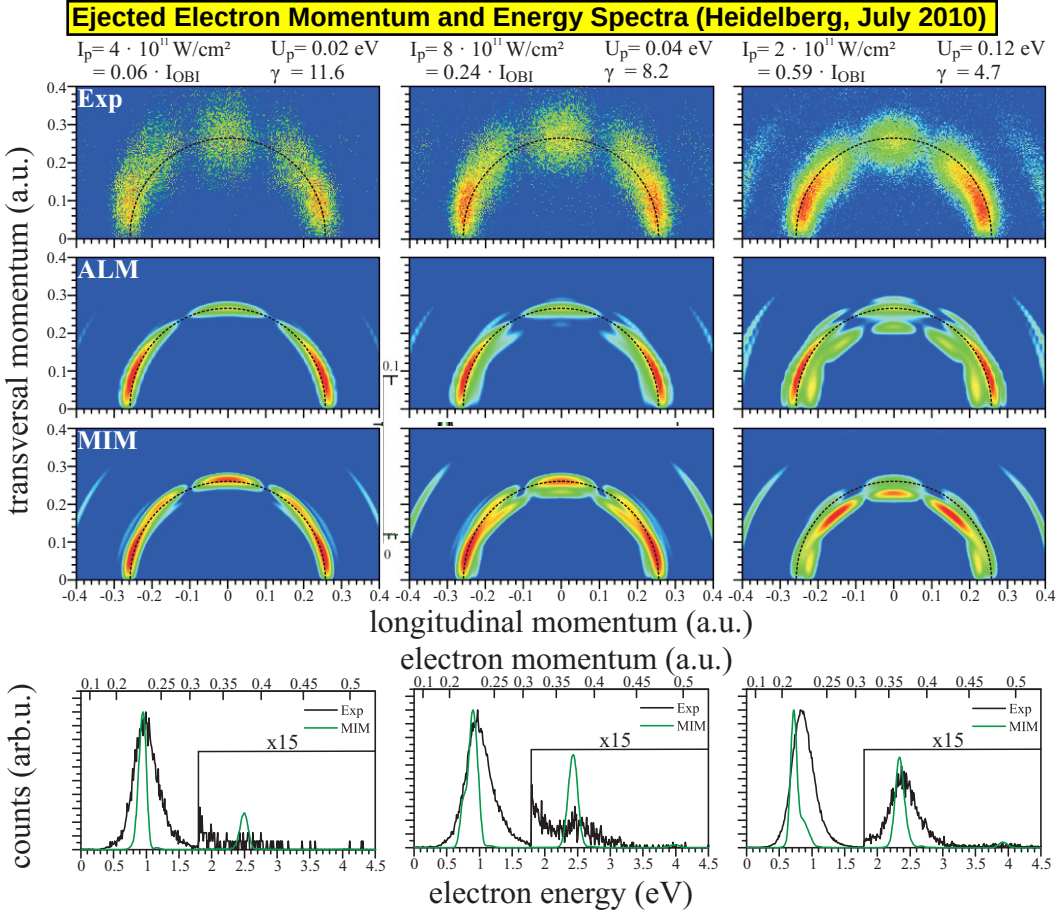


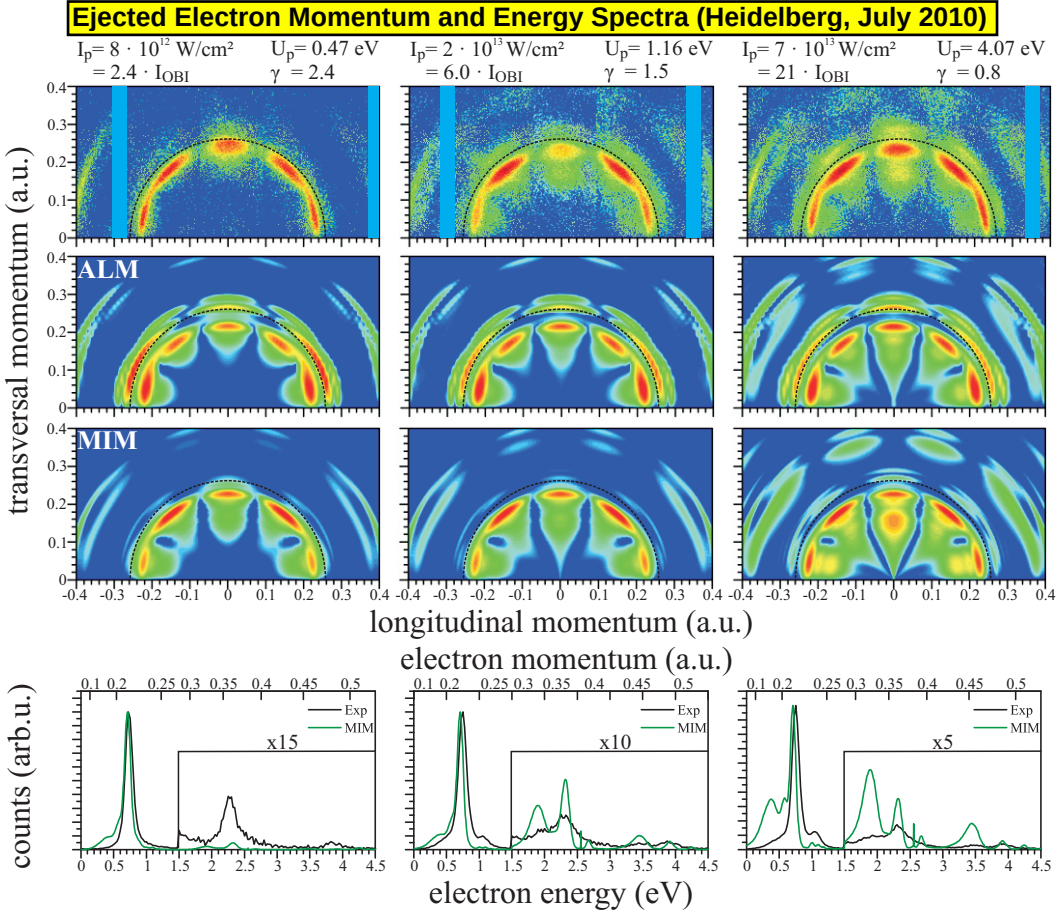
Ejected Electron Spectra



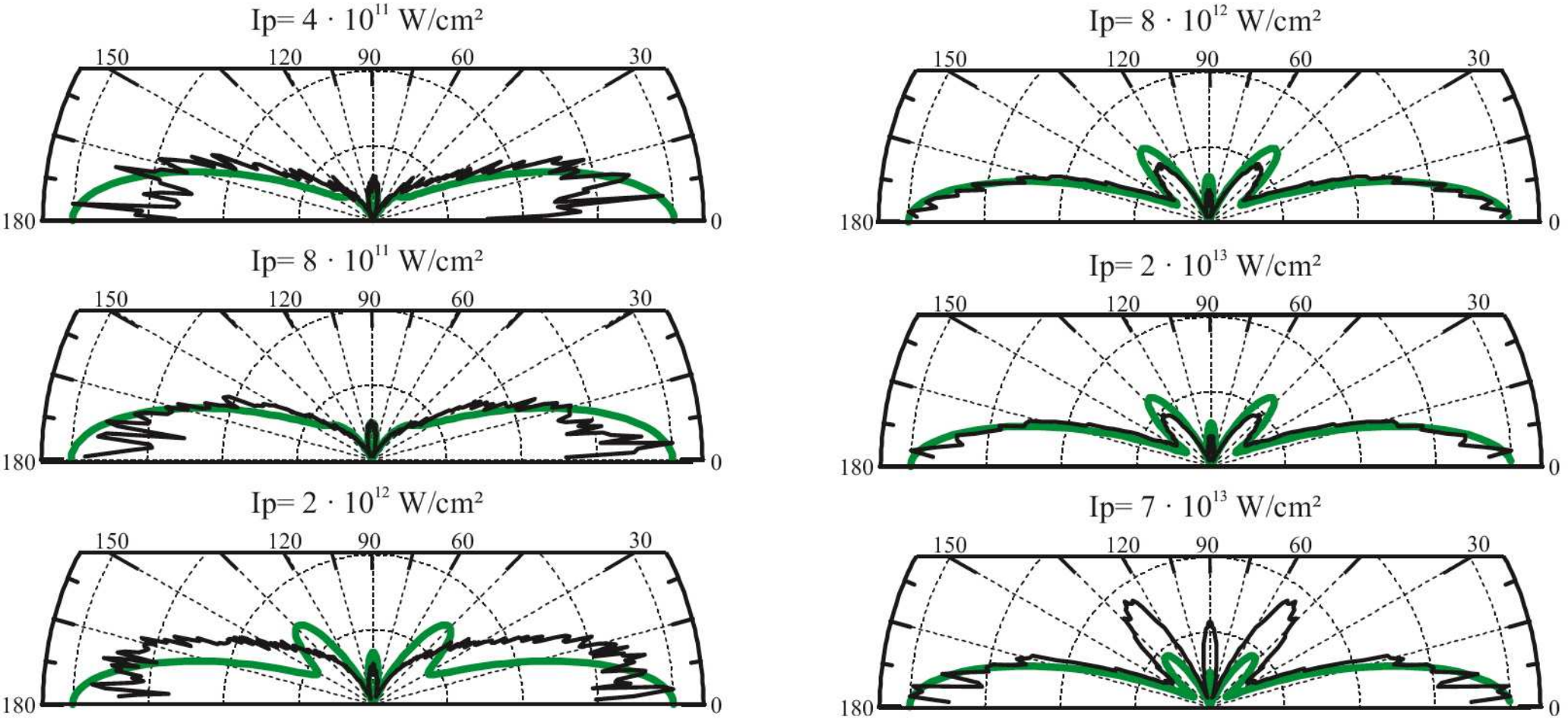
Ejected Electron Spectra




Ejected Electron Momentum and Energy Spectra (Heidelberg, July 2010)
$I_p = 4 \cdot 10^{11}$ W/cm² = 0.06 · I_{OBI}
U_p = 0.02 eV
γ = 11.6
$I_p = 8 \cdot 10^{11}$ W/cm² = 0.24 · I_{OBI}
U_p = 0.04 eV
γ = 8.2
$I_p = 2 \cdot 10^{11}$ W/cm² = 0.59 · I_{OBI}
U_p = 0.12 eV
γ = 4.7
Exp
ALM
MIM
transversal momentum (a.u.)
longitudinal momentum (a.u.)
electron momentum (a.u.)
counts (arb.u.)
Exp
MIM
x15
electron energy (eV)


Ejected Electron Momentum and Energy Spectra (Heidelberg, July 2010)
$I_p = 8 \cdot 10^{12}$ W/cm² = 2.4 · I_{OBI}
U_p = 0.47 eV
γ = 2.4
$I_p = 2 \cdot 10^{13}$ W/cm² = 6.0 · I_{OBI}
U_p = 1.16 eV
γ = 1.5
$I_p = 7 \cdot 10^{13}$ W/cm² = 21 · I_{OBI}
U_p = 4.07 eV
γ = 0.8
ALM
MIM
transversal momentum (a.u.)
longitudinal momentum (a.u.)
electron momentum (a.u.)
counts (arb.u.)
Exp
MIM
x15
x10
x5
electron energy (eV)

Angular Distributions in the Main Line



We seem to be on the right track, but there is room for improvement!

For more, see Schuricke *et al.*, Phys. Rev. A 83 (2011) 023413

Hang in there for a little bit longer ...

Part 2

Many-Electron Systems

Those are REALLY hard!

But one can see two (or even more) electrons play together :-) :-) :-)

A pioneering calculation ...

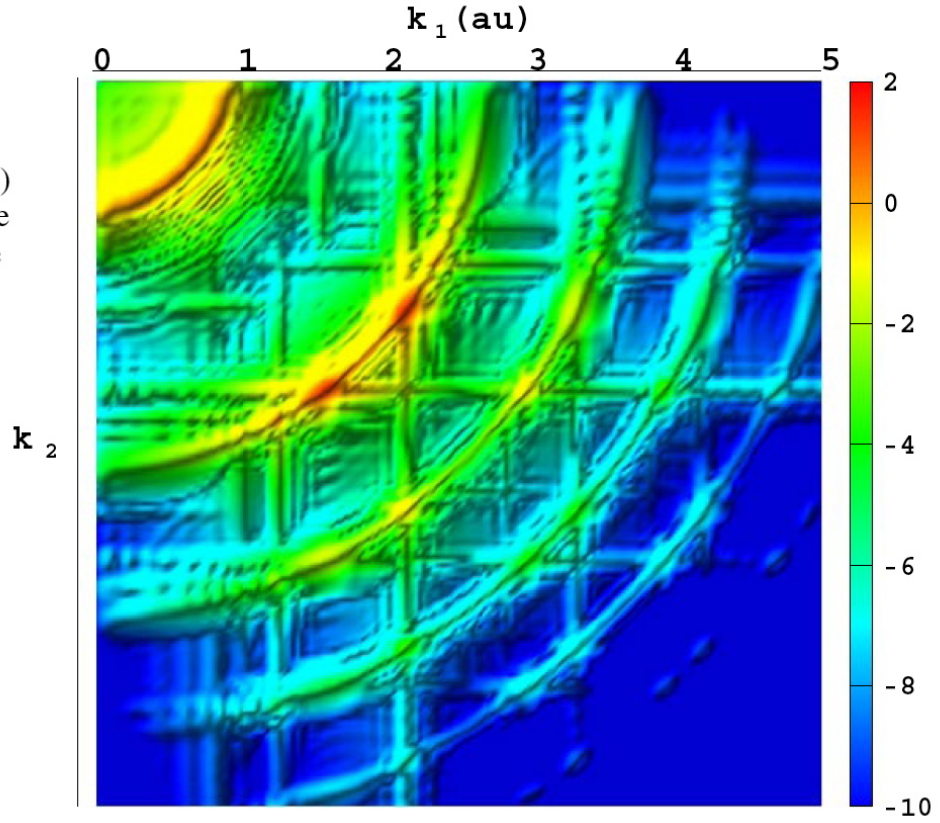
J. Phys. B: At. Mol. Opt. Phys. **34** (2001) L69–L78

Double-electron above threshold ionization of helium

J S Parker, L R Moore, K J Meharg, D Dundas and K T Taylor

Department of Applied Mathematics and Theoretical Physics, The Queen's University of Belfast, Belfast BT7 1NN, UK

Log plot of the probability distribution $P(k_1, k_2)$ of doubly ionizing electrons in momentum space after excitation with a 46 field period laser pulse of frequency 3.2 Hartrees and of peak intensity 2.0×10^{16} W cm^{-2}.



Another one ... [Science 310 (2005) 1787]

Complete Photo-Induced Breakup of the H_2 Molecule as a Probe of Molecular Electron Correlation

Wim Vanroose,[1] Fernando Martín,[2] Thomas N. Rescigno,[3] C. William McCurdy[3,4]

Despite decades of progress in quantum mechanics, electron correlation effects are still only partially understood. Experiments in which both electrons are ejected from an oriented hydrogen molecule by absorption of a single photon have recently demonstrated a puzzling phenomenon: The ejection pattern of the electrons depends sensitively on the bond distance between the two nuclei as they vibrate in their ground state. Here, we report a complete numerical solution of the Schrödinger equation for the double photoionization of H_2. The results suggest that the distribution of photoelectrons emitted from aligned molecules reflects electron correlation effects that are purely molecular in origin.

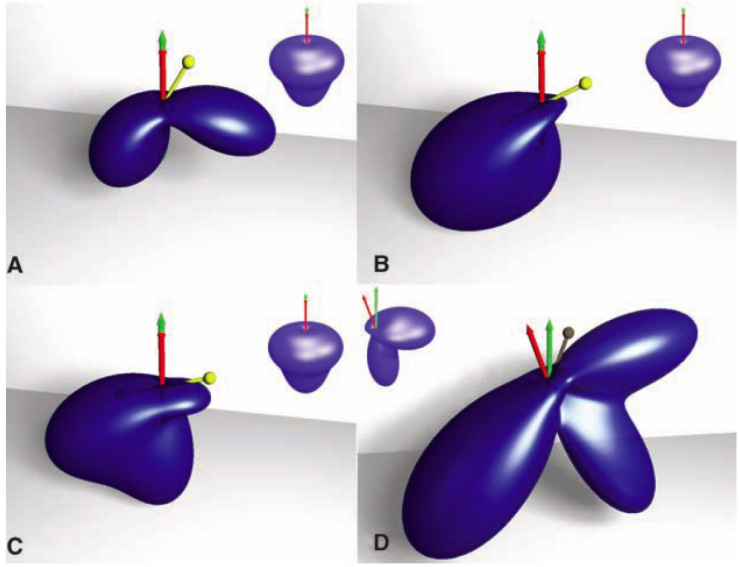


Fig. 3. Effects of molecular orientation on the angular distribution of ejected electrons. For the fixed electron ejected along the polarization direction with 90% of the kinetic energy, the molecule makes an angle with the polarization of (**A**) 30°, (**B**) 60°, and (**C**) 75°, and splits the corresponding pattern for the helium atom (insets) into two lobes which vary in size and ultimately show a tendency to align along the molecular axis as in Fig. 1B. The cross section in (A) is about one-fourth the magnitude of (B) and (C). (**D**) A case in which the molecule and fixed electron have 10% of the kinetic energy, both at 20° from the polarization vector but on opposite sides, yielding an ejection pattern markedly different from the corresponding atomic one.

PERIODIC TABLE

Atomic Properties of the Elements

NIST
National Institute of Standards and Technology
Technology Administration, U.S. Department of Commerce

Physics Laboratory
physics.nist.gov

Standard Reference Data Group
www.nist.gov/srd

Frequently used fundamental physical constants

For the most accurate values of these and other constants, visit physics.nist.gov/constants

1 second = 9 192 631 770 periods of radiation corresponding to the transition between the two hyperfine levels of the ground state of ^{133}Cs

speed of light in vacuum	c	299 792 458 m s^{-1}	(exact)
Planck constant	h	6.6261×10^{-34} J s	($\hbar = h/2\pi$)
elementary charge	e	1.6022×10^{-19} C	
electron mass	m_e	9.1094×10^{-31} kg	
	$m_e c^2$	0.5110 MeV	
proton mass	m_p	1.6726×10^{-27} kg	
fine-structure constant	α	1/137.036	
Rydberg constant	R_∞	10 973 732 m^{-1}	
	$R_\infty c$	$3.289\,842 \times 10^{15}$ Hz	
	$R_\infty hc$	13.6057 eV	
Boltzmann constant	k	1.3807×10^{-23} J K^{-1}	

Solids
Liquids
Gases
Artificially Prepared

Group 1 IA | 2 IIA | 3 IIIB | 4 IVB | 5 VB | 6 VIB | 7 VIIB | 8 | 9 VIII | 10 | 11 IB | 12 IIB | 13 IIIA | 14 IVA | 15 VA | 16 VIA | 17 VIIA | 18 VIIIA

Period 1

1 $^2S_{1/2}$ **H** Hydrogen 1.00794 1s 13.5984																	2 $^1S_0$ **He** Helium 4.002602 $1s^2$ 24.5874
3 $^2S_{1/2}$ **Li** Lithium 6.941 $1s^2 2s$ 5.3917	4 $^1S_0$ **Be** Beryllium 9.012182 $1s^2 2s^2$ 9.3227											5 $^2P^o_{1/2}$ **B** Boron 10.811 $1s^2 2s^2 2p$ 8.2980	6 $^3P_0$ **C** Carbon 12.0107 $1s^2 2s^2 2p^2$ 11.2603	7 $^4S^o_{3/2}$ **N** Nitrogen 14.0067 $1s^2 2s^2 2p^3$ 14.5341	8 $^3P_2$ **O** Oxygen 15.9994 $1s^2 2s^2 2p^4$ 13.6181	9 $^2P^o_{3/2}$ **F** Fluorine 18.9984032 $1s^2 2s^2 2p^5$ 17.4228	10 $^1S_0$ **Ne** Neon 20.1797 $1s^2 2s^2 2p^6$ 21.5645
11 $^2S_{1/2}$ **Na** Sodium 22.989770 [Ne]3s 5.1391	12 $^1S_0$ **Mg** Magnesium 24.3050 $[Ne]3s^2$ 7.6462											13 $^2P^o_{1/2}$ **Al** Aluminum 26.981538 $[Ne]3s^2 3p$ 5.9858	14 $^3P_0$ **Si** Silicon 28.0855 $[Ne]3s^2 3p^2$ 8.1517	15 $^4S^o_{3/2}$ **P** Phosphorus 30.973761 $[Ne]3s^2 3p^3$ 10.4867	16 $^3P_2$ **S** Sulfur 32.065 $[Ne]3s^2 3p^4$ 10.3600	17 $^2P^o_{3/2}$ **Cl** Chlorine 35.453 $[Ne]3s^2 3p^5$ 12.9676	18 $^1S_0$ **Ar** Argon 39.948 $[Ne]3s^2 3p^6$ 15.7596
19 $^2S_{1/2}$ **K** Potassium 39.0983	20 $^1S_0$ **Ca** Calcium 40.078	21 $^2D_{3/2}$ **Sc** Scandium 44.955910	22 $^3F_2$ **Ti** Titanium 47.867	23 $^4F_{3/2}$ **V** Vanadium 50.9415	24 $^7S_3$ **Cr** Chromium 51.9961	25 $^6S_{5/2}$ **Mn** Manganese 54.938049	26 $^5D_4$ **Fe** Iron 55.845	27 $^4F_{9/2}$ **Co** Cobalt 58.933200	28 $^3F_4$ **Ni** Nickel 58.6934	29 $^2S_{1/2}$ **Cu** Copper 63.546	30 $^1S_0$ **Zn** Zinc 65.409	31 $^2P^o_{1/2}$ **Ga** Gallium 69.723	32 $^3P_0$ **Ge** Germanium 72.64	33 $^4S^o_{3/2}$ **As** Arsenic 74.92160	34 $^3P_2$ **Se** Selenium 78.96	35 $^2P^o_{3/2}$ **Br** Bromine 79.904	36 $^1S_0$ **Kr** Krypton 83.798
37 $^2S_{1/2}$ **Rb** Rubidium 85.4678 [Kr]5s 4.1771	38 $^1S_0$ **Sr** Strontium 87.62 $[Kr]5s^2$ 5.6949	39 $^2D_{3/2}$ **Y** Yttrium 88.90585 $[Kr]4d5s^2$ 6.2173	40 $^3F_2$ **Zr** Zirconium 91.224 $[Kr]4d^2 5s^2$ 6.6339	41 $^4F_{3/2}$ **Nb** Niobium 92.90638 $[Kr]4d^4 5s$ 6.7589	42 $^7S_3$ **Mo** Molybdenum 95.94 $[Kr]4d^5 5s$ 7.0924	43 $^6S_{5/2}$ **Tc** Technetium (98) $[Kr]4d^5 5s^2$ 7.28	44 $^5D_4$ **Ru** Ruthenium 101.07 $[Kr]4d^7 5s$ 7.3605	45 $^4F_{9/2}$ **Rh** Rhodium 102.90550 $[Kr]4d^8 5s$ 7.4589	46 $^1S_0$ **Pd** Palladium 106.42 $[Kr]4d^{10}$ 8.3369	47 $^2S_{1/2}$ **Ag** Silver 107.8682 $[Kr]4d^{10} 5s$ 7.5762	48 $^1S_0$ **Cd** Cadmium 112.411 $[Kr]4d^{10} 5s^2$ 8.9938	49 $^2P^o_{1/2}$ **In** Indium 114.818 $[Kr]4d^{10} 5s^2 5p$ 5.7864	50 $^3P_0$ **Sn** Tin 118.710 $[Kr]4d^{10} 5s^2 5p^2$ 7.3439	51 $^4S^o_{3/2}$ **Sb** Antimony 121.760 $[Kr]4d^{10} 5s^2 5p^3$ 8.6084	52 $^3P_2$ **Te** Tellurium 127.60 $[Kr]4d^{10} 5s^2 5p^4$ 9.0096	53 $^2P^o_{3/2}$ **I** Iodine 126.90447 $[Kr]4d^{10} 5s^2 5p^5$ 10.4513	54 $^1S_0$ **Xe** Xenon 131.293 $[Kr]4d^{10} 5s^2 5p^6$ 12.1298
55 $^2S_{1/2}$ **Cs** Cesium 132.90545 [Xe]6s 3.8939	56 $^1S_0$ **Ba** Barium 137.327 $[Xe]6s^2$ 5.2117	72 $^3F_2$ **Hf** Hafnium 178.49 $[Xe]4f^{14} 5d^2 6s^2$ 6.8251	73 $^4F_{3/2}$ **Ta** Tantalum 180.9479 $[Xe]4f^{14} 5d^3 6s^2$ 7.5496	74 $^5D_0$ **W** Tungsten 183.84 $[Xe]4f^{14} 5d^4 6s^2$ 7.8640	75 $^6S_{5/2}$ **Re** Rhenium 186.207 $[Xe]4f^{14} 5d^5 6s^2$ 7.8335	76 $^5D_4$ **Os** Osmium 190.23 $[Xe]4f^{14} 5d^6 6s^2$ 8.4382	77 $^4F_{9/2}$ **Ir** Iridium 192.217 $[Xe]4f^{14} 5d^7 6s^2$ 8.9670	78 $^3D_3$ **Pt** Platinum 195.078 $[Xe]4f^{14} 5d^9 6s$ 8.9588	79 $^2S_{1/2}$ **Au** Gold 196.96655 $[Xe]4f^{14} 5d^{10} 6s$ 9.2255	80 $^1S_0$ **Hg** Mercury 200.59 $[Xe]4f^{14} 5d^{10} 6s^2$ 10.4375	81 $^2P^o_{1/2}$ **Tl** Thallium 204.3833 [Hg]6p 6.1082	82 $^3P_0$ **Pb** Lead 207.2 $[Hg]6p^2$ 7.4167	83 $^4S^o_{3/2}$ **Bi** Bismuth 208.98038 $[Hg]6p^3$ 7.2855	84 $^3P_2$ **Po** Polonium (209) $[Hg]6p^4$ 8.414	85 $^2P^o_{3/2}$ **At** Astatine (210) $[Hg]6p^5$	86 $^1S_0$ **Rn** Radon (222) $[Hg]6p^6$ 10.7485
87 $^2S_{1/2}$ **Fr** Francium (223) [Rn]7s 4.0727	88 $^1S_0$ **Ra** Radium (226) $[Rn]7s^2$ 5.2784	104 $^3F_2$? **Rf** Rutherfordium (261) $[Rn]5f^{14} 6d^2 7s^2$? 6.0 ?	105 **Db** Dubnium (262)	106 **Sg** Seaborgium (266)	107 **Bh** Bohrium (264)	108 **Hs** Hassium (277)	109 **Mt** Meitnerium (268)	110 **Uun** Ununnilium (281)	111 **Uuu** Unununium (272)	112 **Uub** Ununbium (285)		114 **Uuq** Ununquadium (289)			116 **Uuh** Ununhexium (292)		

Note that there are other atoms than He!

Lanthanides

57 $^2D_{3/2}$ **La** Lanthanum 138.9055 $[Xe]5d6s^2$ 5.5769	58 $^1G^o_4$ **Ce** Cerium 140.116 $[Xe]4f5d6s^2$ 5.5387	59 $^4I^o_{9/2}$ **Pr** Praseodymium 140.90765 $[Xe]4f^3 6s^2$ 5.473	60 $^5I_4$ **Nd** Neodymium 144.24 $[Xe]4f^4 6s^2$ 5.5250	61 $^6H^o_{5/2}$ **Pm** Promethium (145) $[Xe]4f^5 6s^2$ 5.582	62 $^7F_0$ **Sm** Samarium 150.36 $[Xe]4f^6 6s^2$ 5.6437	63 $^8S^o_{7/2}$ **Eu** Europium 151.964 $[Xe]4f^7 6s^2$ 5.6704	64 $^9D^o_2$ **Gd** Gadolinium 157.25 $[Xe]4f^7 5d6s^2$ 6.1498	65 $^6H^o_{15/2}$ **Tb** Terbium 158.92534 $[Xe]4f^9 6s^2$ 5.8638	66 $^5I_8$ **Dy** Dysprosium 162.500 $[Xe]4f^{10} 6s^2$ 5.9389	67 $^4I^o_{15/2}$ **Ho** Holmium 164.93032 $[Xe]4f^{11} 6s^2$ 6.0215	68 $^3H_6$ **Er** Erbium 167.259 $[Xe]4f^{12} 6s^2$ 6.1077	69 $^2F^o_{7/2}$ **Tm** Thulium 168.93421 $[Xe]4f^{13} 6s^2$ 6.1843	70 $^1S_0$ **Yb** Ytterbium 173.04 $[Xe]4f^{14} 6s^2$ 6.2542	71 $^2D_{3/2}$ **Lu** Lutetium 174.967 $[Xe]4f^{14} 5d6s^2$ 5.4259

Actinides

89 $^2D_{3/2}$ **Ac** Actinium (227) $[Rn]6d7s^2$ 5.17	90 $^3F_2$ **Th** Thorium 232.0381 $[Rn]6d^2 7s^2$ 6.3067	91 $^4K_{11/2}$ **Pa** Protactinium 231.03588 $[Rn]5f^2 6d7s^2$ 5.89	92 $^5L_6$ **U** Uranium 238.02891 $[Rn]5f^3 6d7s^2$ 6.1941	93 $^6L_{11/2}$ **Np** Neptunium (237) $[Rn]5f^4 6d7s^2$ 6.2657	94 $^7F_0$ **Pu** Plutonium (244) $[Rn]5f^6 7s^2$ 6.0260	95 $^8S^o_{7/2}$ **Am** Americium (243) $[Rn]5f^7 7s^2$ 5.9738	96 $^9D^o_2$ **Cm** Curium (247) $[Rn]5f^7 6d7s^2$ 5.9914	97 $^6H^o_{15/2}$ **Bk** Berkelium (247) $[Rn]5f^9 7s^2$ 6.1979	98 $^5I_8$ **Cf** Californium (251) $[Rn]5f^{10} 7s^2$ 6.2817	99 $^4I^o_{15/2}$ **Es** Einsteinium (252) $[Rn]5f^{11} 7s^2$ 6.42	100 $^3H_6$ **Fm** Fermium (257) $[Rn]5f^{12} 7s^2$ 6.50	101 $^2F^o_{7/2}$ **Md** Mendelevium (258) $[Rn]5f^{13} 7s^2$ 6.58	102 $^1S_0$ **No** Nobelium (259) $[Rn]5f^{14} 7s^2$ 6.65	103 $^2P^o_{1/2}$? **Lr** Lawrencium (262) $[Rn]5f^{14} 7s^2 7p$? 4.9 ?

Atomic Number | Ground-state Level | Symbol | Name | Atomic Weight† | Ground-state Configuration | Ionization Energy (eV)

58 $^1G^o_4$ **Ce** Cerium 140.116 $[Xe]4f5d6s^2$ 5.5387

†Based upon ^{12}C. () indicates the mass number of the most stable isotope.

For a description of the data, visit physics.nist.gov/data

NIST SP 966 (September 2003)

Let's go for generality!

This is where we need to know a lot about electron scattering and atomic structure!

Theoretical Formulation

- **Algorithm Requirements**
  - Efficient generation of the Hamiltonian and electron−field interaction matrix elements.
  - Efficient propagation of the time-dependent Schrödinger equation (TDSE).
  - **Generality beyond** applications to **(quasi)-one or (quasi)-two electron targets.**

Sorry – here comes the cut!

But we did a lot of (good) work on this ...
You can hear about some of it tomorrow!

List of early calculations with the BSR code (rapidly growing)

***hν* + Li**	Zatsarinny O and Froese Fischer C *J. Phys. B* **33** 313 (2000)
***hν* + He**	Zatsarinny O, Gorczyca T W and Froese Fischer C *J. Phys. B.* **35** 4161 (2002)
***hν* + C**	Gibson N D *et al.* Phys. Rev. A **67**, 030703 (2003)
***hν* + B**	Zatsarinny O and Gorczyca T W Abstracts of XXII ICPEAC (2003)
***hν* + O**	Zatsarinny O and Bartschat K *Phys. Rev. A* **73** 022714 (2006)
***hν* + Ca**	Zatsarinny O *et al. Phys. Rev. A* **74** 052708 (2006)
e + He	Stepanovic *et al. J. Phys. B* **39** 1547 (2006)
	Lange M *et al. J. Phys. B* **39** 4179 (2006)
e + C	Zatsarinny O, Bartschat K, Bandurina L and Gedeon V *Phys. Rev. A* **71** 042702 (2005)
e + O	Zatsarinny O and Tayal S S *J. Phys. B* **34** 1299 (2001)
	Zatsarinny O and Tayal S S *J. Phys. B* **35** 241 (2002)
	Zatsarinny O and Tayal S S As. J. S. S. **148** 575 (2003)
e + Ne	Zatsarinny O and Bartschat K *J. Phys. B* **37** 2173 (2004)
	Bömmels J *et al.* Phys. Rev. A **71**, 012704 (2005)
	Allan M *et al. J. Phys. B* **39** L139 (2006)
e + Mg	Bartschat K, Zatsarinny O, Bray I, Fursa D V and Stelbovics A T *J. Phys. B* **37** 2617 (2004)
e + S	Zatsarinny O and Tayal S S *J. Phys. B* **34** 3383 (2001)
	Zatsarinny O and Tayal S S *J. Phys. B* **35** 2493 (2002)
e + Ar	Zatsarinny O and Bartschat K *J. Phys. B* **37** 4693 (2004)
e + K (inner-shell)	Borovik A A *et al. Phys. Rev. A,* **73** 062701 (2006)
e + Zn	Zatsarinny O and Bartschat K *Phys. Rev. A* **71** 022716 (2005)
e + Fe^+	Zatsarinny O and Bartschat K *Phys. Rev. A* **72** 020702(R) (2005)
e + Kr	Zatsarinny O and Bartschat K *J. Phys. B* **40** F43 (2007)
e + Xe	Allan M, Zatsarinny O and Bartschat K *Phys. Rev. A* 030701(R) (2006)
Rydberg series in C	Zatsarinny O and Froese Fischer C *J. Phys. B* **35** 4669 (2002)
osc. strengths in Ar	Zatsarinny O and Bartschat K *J. Phys. B: At. Mol. Opt. Phys.* **39** 2145 (2006)
osc. strengths in S	Zatsarinny O and Bartschat K *J. Phys. B: At. Mol. Opt. Phys.* **39** 2861 (2006)
osc. strengths in Xe	Dasgupta A *et al. Phys. Rev. A* **74** 012509 (2006)

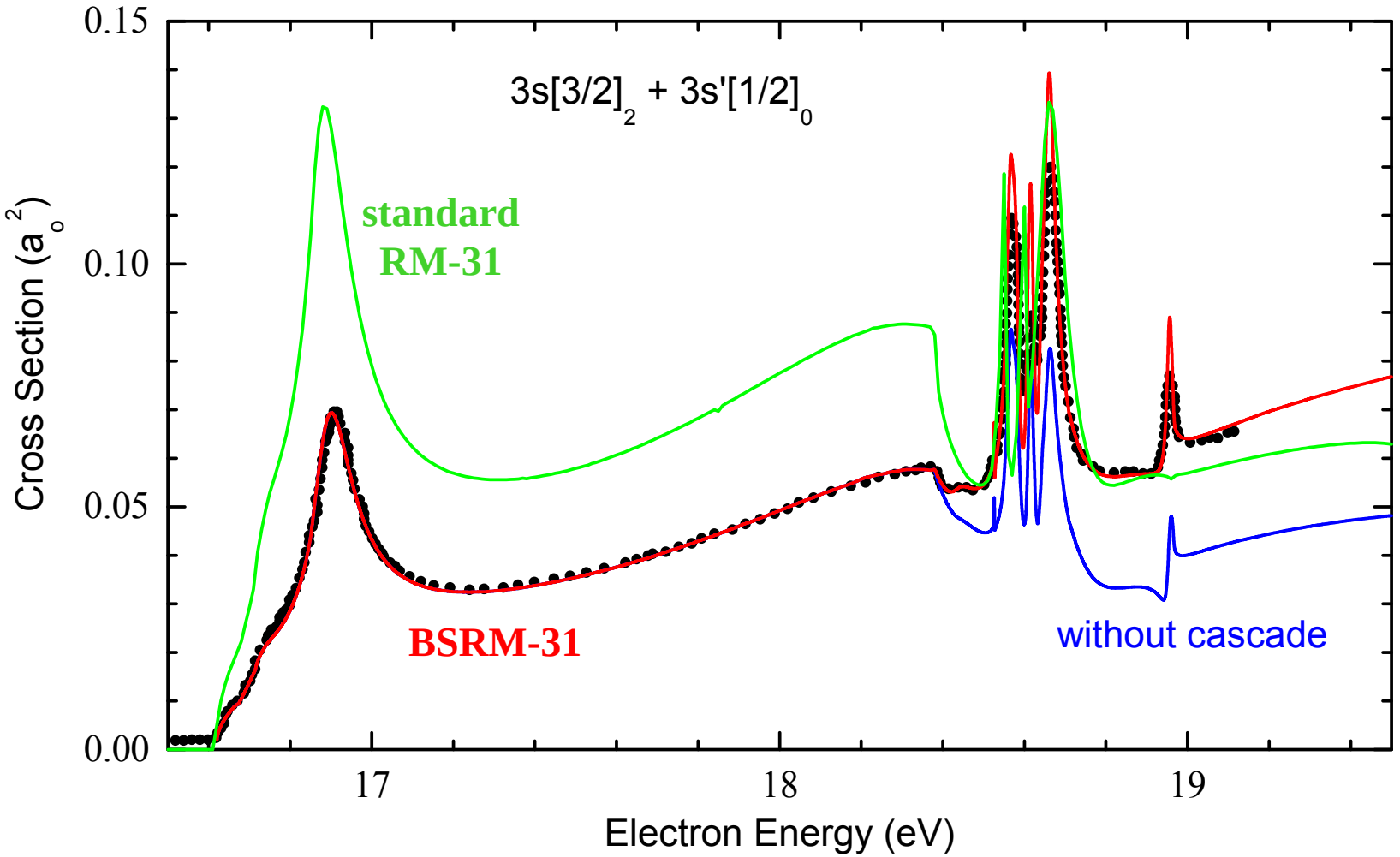
at least 40 more since 2006

Metastable yield in e-Ne collisions

Experiment: Buckman *et al.* (1983) [x 0.78]

Theories: 31-state Breit-Pauli R-matrix (Zeman & Bartschat 1997)

31-state B-spline R-matrix (Zatsarinny & Bartschat 2004)



Theoretical Formulation

- **Algorithm Requirements**
  - Efficient generation of the Hamiltonian and electron−field interaction matrix elements.
  - Efficient propagation of the time-dependent Schrödinger equation (TDSE).
  - **Generality beyond** applications to **(quasi)-one or (quasi)-two electron targets.**

- **Basic Equations**

We need to get this function!

$$i\frac{\partial}{\partial t}\Psi(\boldsymbol{r}_1,...,\boldsymbol{r}_N;t) = \left[\boldsymbol{H}_0(\boldsymbol{r}_1,...,\boldsymbol{r}_N) + V(\boldsymbol{r}_1,...,\boldsymbol{r}_N;t)\right]\Psi(\boldsymbol{r}_1,...,\boldsymbol{r}_N;t),$$

where $\boldsymbol{H}_0$ is the field-free Hamiltonian containing the kinetic energy of the N electrons, their potential energy in the field of the nucleus, and their mutual Coulomb repulsion.

Numerical Method of Solution

- We use a B-spline R-matrix method to generate all required matrices.
- The **primary advantages of this method** are:
  - B-splines have excellent numerical properties.
  - The use of non-orthogonal orbital sets allows for high flexibility in the target description.
- We use the **Arnoldi-Lanczos** method to propagate the TDSE in time by approximating the operator $\exp(-i\boldsymbol{H}\Delta t)$ in Krylov space.
- Because of the non-orthogonality of the primitive and the physical basis, we **[used to] transform the original matrices** and generate

$$\boldsymbol{H}_0' = \boldsymbol{S}^{-1/2}\boldsymbol{H}_0\boldsymbol{S}^{-1/2}; \quad \boldsymbol{D}' = \boldsymbol{S}^{-1/2}\boldsymbol{D}\boldsymbol{S}^{-1/2}.$$

Phys. Rev. A 76, 053411 (2007)

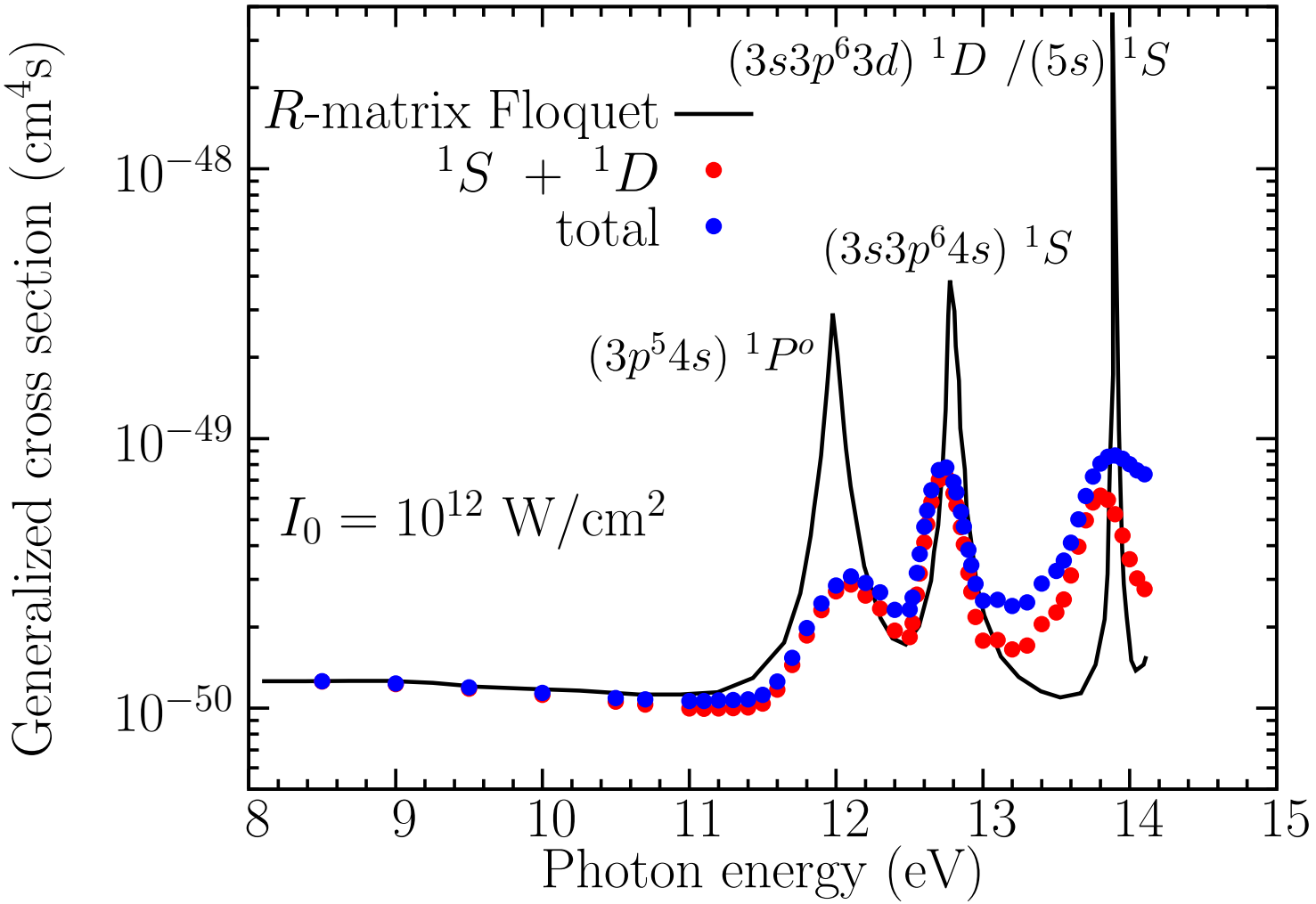
- Since $\boldsymbol{H}_0$, $\boldsymbol{D}$, and the overlap matrix $\boldsymbol{S}$ are all time-independent, this only **requires the diagonalization of $\boldsymbol{S}$ once** and matrix-vector multiplications at every time step.
- It is **much simpler to solve the generalized eigenvalue for each field-free partial-wave symmetry and transform the entire problem to the eigenbasis**. Then:
  - The field-free hamiltonian is diagonal.
  - High-energy states that would not be reached can be removed to improve the numerics.
  - It is very easy to extract the information.

Phys. Rev. A 78, 053402 (2008)

- **Observables presented** include:
  - Survival probability of the ground state
  - Probability for excitation
  - Probability for ionization

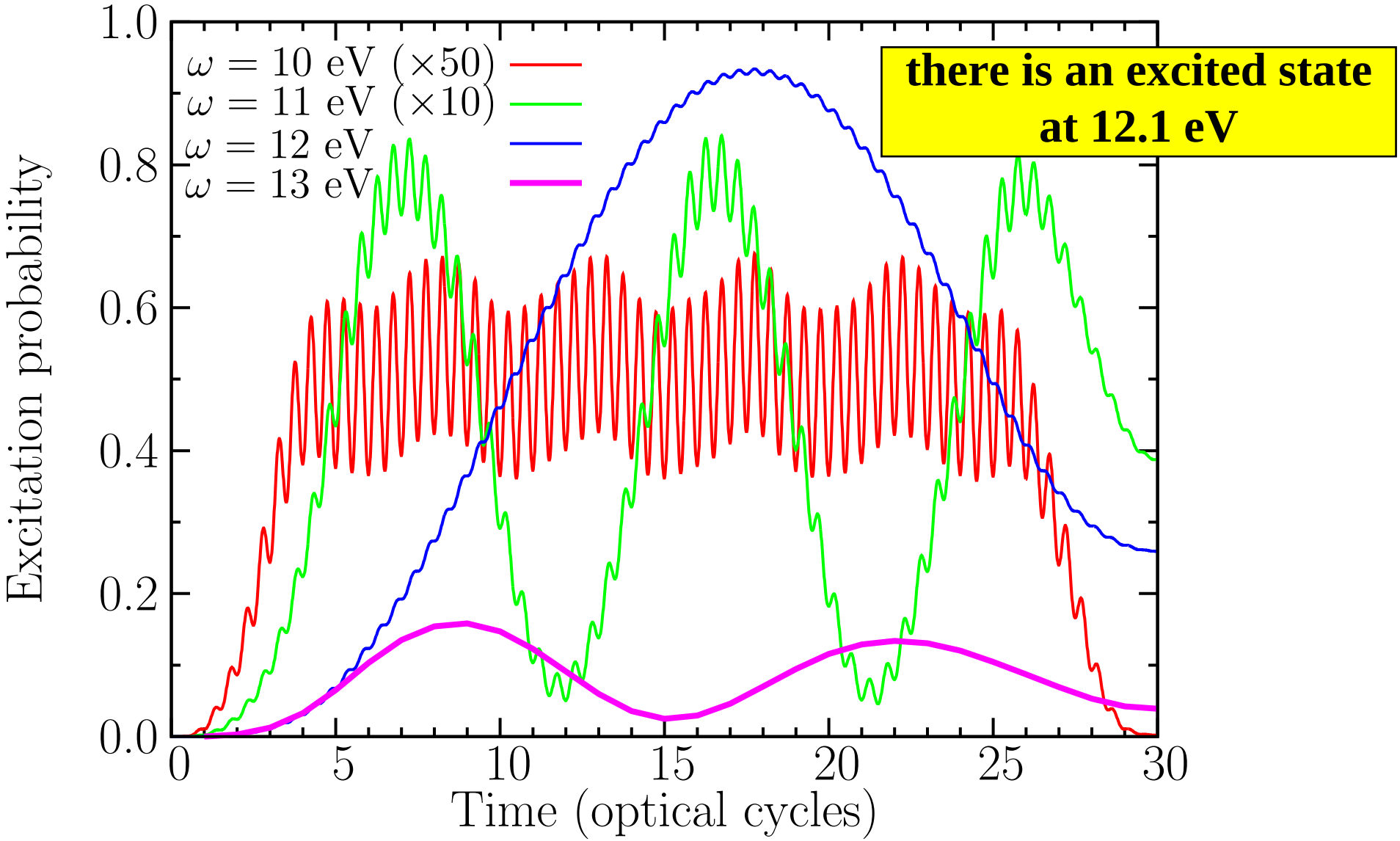
Phys. Rev. A 78, 053402 (2008)

Two-Photon Ionization of Argon



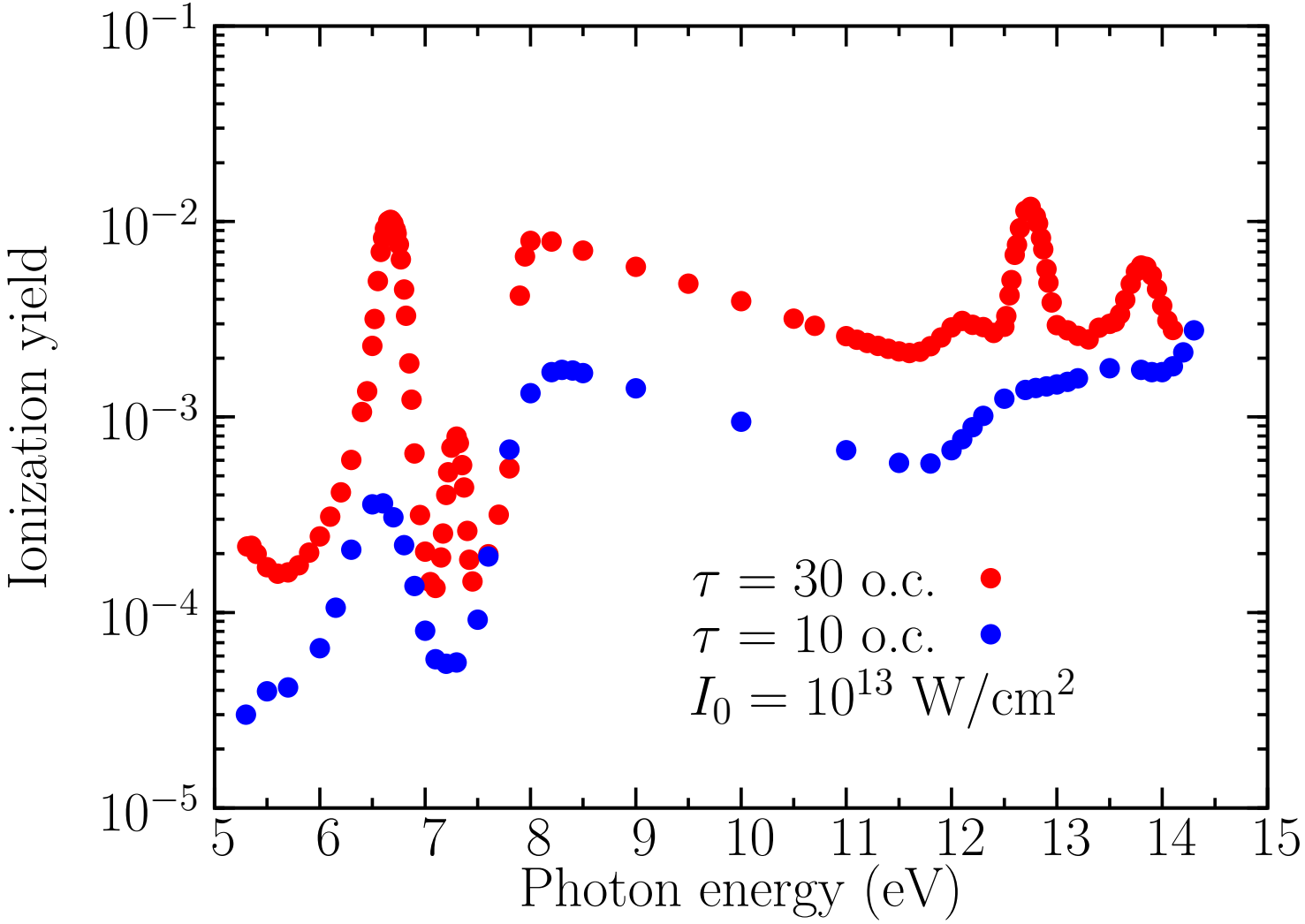
- Generalized cross section for two-photon ionization of Ar$(3p^6)^1S$
- 30-cycle laser pulse with a peak intensity of 10^{12} W/cm^2
- Floquet-results: **McKenna and van der Hart (2004)**

Rabi Oscillations in the Excitation Probability for Ar



- 30-cycle laser pulse with a peak intensity of 10^{12} W/cm^2
- **Note the different scales!**

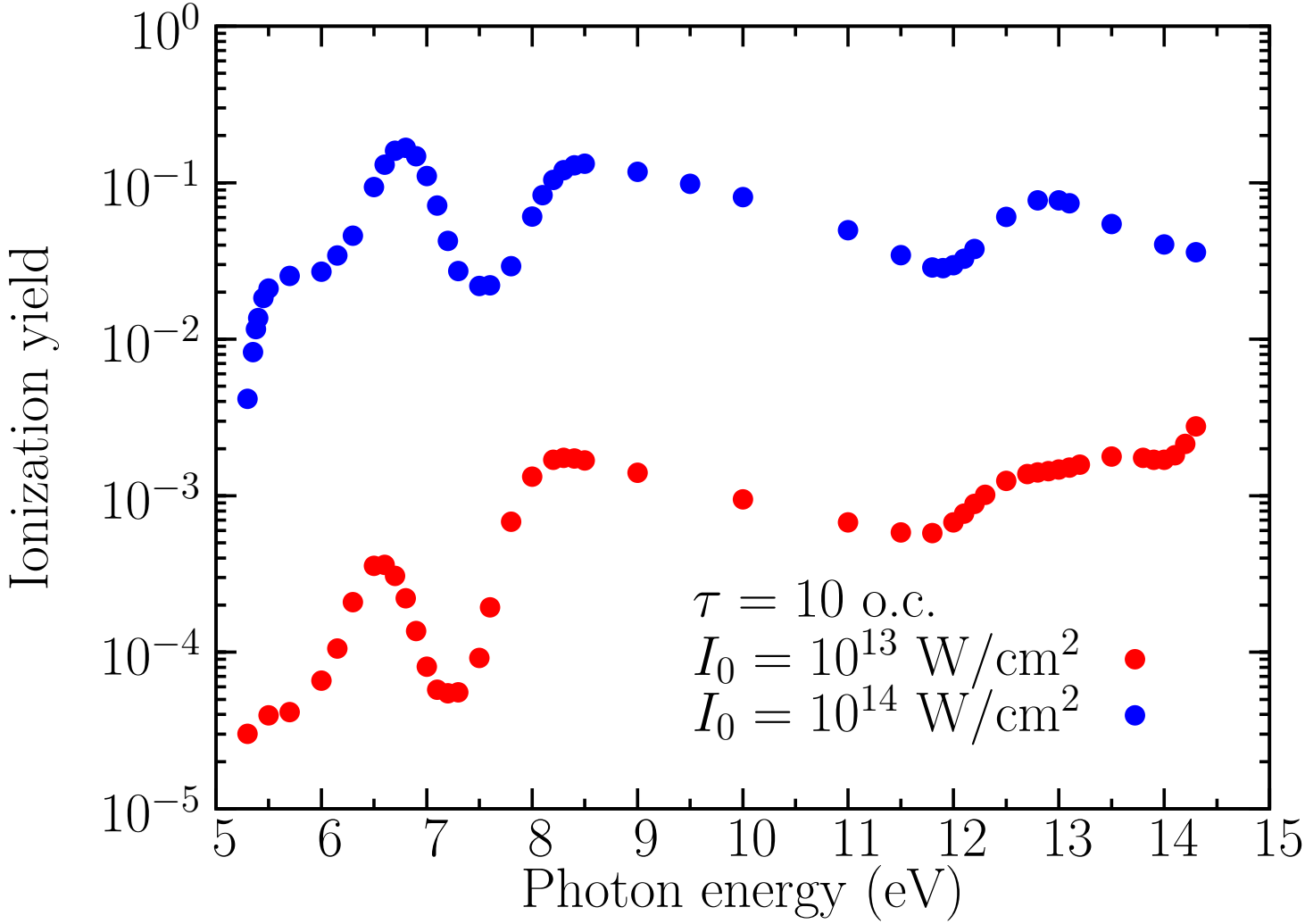
Ionization Yield for Two-Photon and Three-Photon Ionization of Argon



- 10 cycle and 30-cycle laser pulses with a peak intensity of 10^{13} W/cm^2

experimentalists play a lot with pulse length and intensity

Ionization Yield for Two-Photon and Three-Photon Ionization of Argon

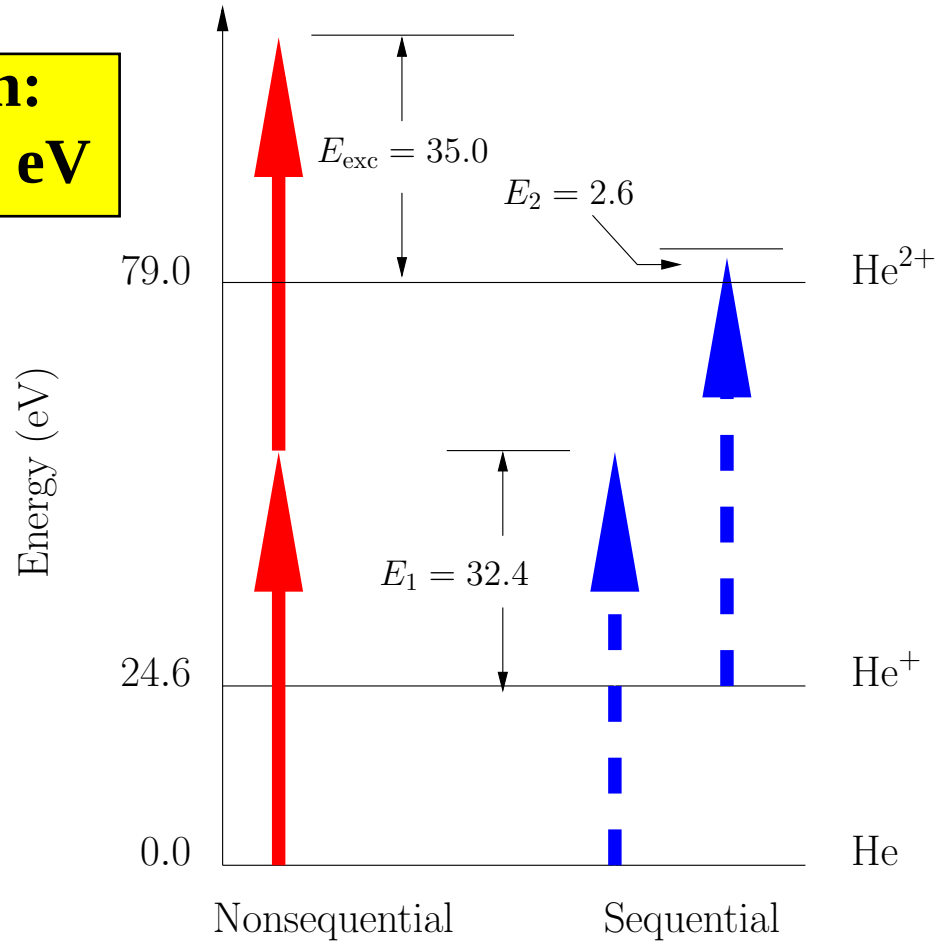


- 10 cycle laser pulses with peak intensityies of $10^{13}\,\mathrm{W/cm^2}$ and $10^{14}\,\mathrm{W/cm^2}$

Application to Two-Photon Double Ionization of Helium

- This is currently a **very controversial topic!**
- Questions include:
  - **Sequential or non-sequential process?**
  - **Final-state correlations** between the two escaping electrons?

illustration:
E_{photon} = 57 eV



A lot of other people like two-electron systems as well ... (this is just a small selection)

J. Phys. B: At. Mol. Opt. Phys. **38** (2005) L35–L45

Triple-differential cross-sections for two-photon double ionization of He near threshold

S X Hu, J Colgan and L A Collins

PRL **96,** 133001 (2006)

High-Energy Cutoff in the Spectrum of Strong-Field Nonsequential Double Ionization

J. S. Parker,[1] B. J. S. Doherty,[1] K. T. Taylor,[1] K. D. Schultz,[2] C. I. Blaga,[2] and L. F. DiMauro[2]

J. Phys. B: At. Mol. Opt. Phys. **40** (2007) 1347–1357

Time-dependent theory of double ionization of helium under XUV radiation

L A A Nikolopoulos[1] and P Lambropoulos[2]

PHYSICAL REVIEW A **75**, 033411 (2007)

Two-photon double ionization of helium in the region of photon energies 42–50 eV

I. A. Ivanov* and A. S. Kheifets

PHYSICAL REVIEW A **73**, 052706 (2006)

Electron-impact ionization of H_2 using a time-dependent close-coupling method

M. S. Pindzola, F. Robicheaux, S. D. Loch, and J. P. Colgan

PHYSICAL REVIEW A **77**, 043420 (2008)

Nonsequential two-photon double ionization of helium

J. Feist,[1,*] S. Nagele,[1] R. Pazourek,[1] E. Persson,[1] B. I. Schneider,[2,3] L. A. Collins,[4] and J. Burgdörfer[1]

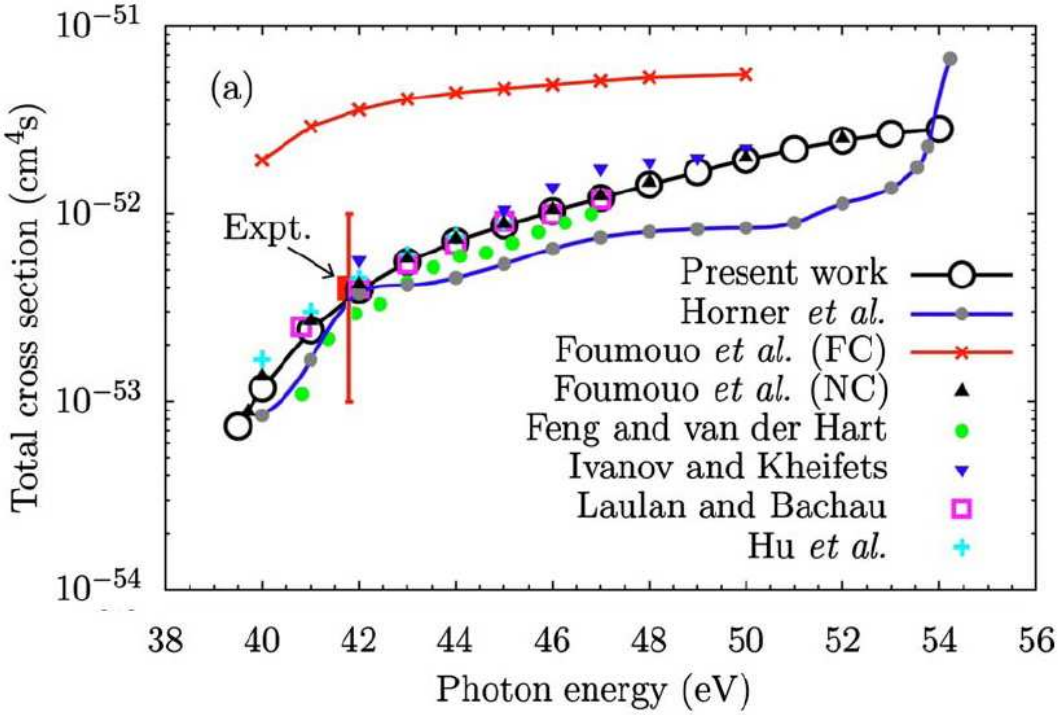
PHYSICAL REVIEW A **77**, 043421 (2008)

Dynamics of two-photon double ionization of helium in short intense xuv laser pulses

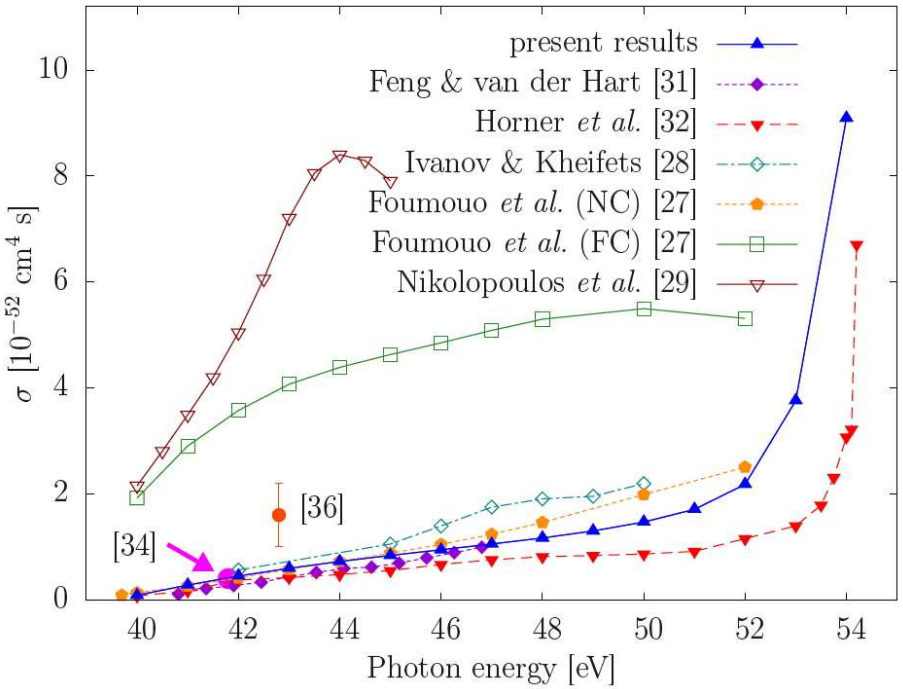
Xiaoxu Guan,[1] K. Bartschat,[1] and B. I. Schneider[2]

Total Cross Section for Two-Photon Double Ionization of Helium

Guan *et al.*, Phys. Rev. A **77**, 043421



Feist *et al.*, Phys. Rev. A **77**, 043420



- There are **enormous differences** between results from various calculations.
- The producers (and defenders) of the large numbers **claim the importance** of final-state correlations.
- The producers (and defenders) of the small numbers **dismiss the importance** of final-state correlations.
- The **experimental uncertainties are also large**, and they **depend on whom you ask!**
- Is the rapid increase of near the sequential threshold a **bug in the formulation** or a **signature of a channel to open soon?**

An example of the controversy – a few people were quite unhappy ...

PHYSICAL REVIEW A **78**, 055402 (2008)

Direct versus sequential double ionization in atomic systems

P. Lambropoulos,[1,2] L. A. A. Nikolopoulos,[3] M. G. Makris,[1] and Andrej Mihelič[1,4]
[1]*Institute of Electronic Structure and Laser, FORTH, P.O. Box 1527, 711 10 Heraklio, Crete, Greece*
[2]*Physics Department, University of Crete, P.O. Box 2208, 710 03 Heraklio, Crete, Greece*
[3]*Centre for Theoretical Atomic, Molecular and Optical Physics, DAMTP, The Queen's University of Belfast, BT7 1NN Belfast, United Kingdom*
[4]*Jožef Stefan Institute, Jamova cesta 39, SI-1000 Ljubljana, Slovenia*
(Received 9 July 2008; published 19 November 2008)

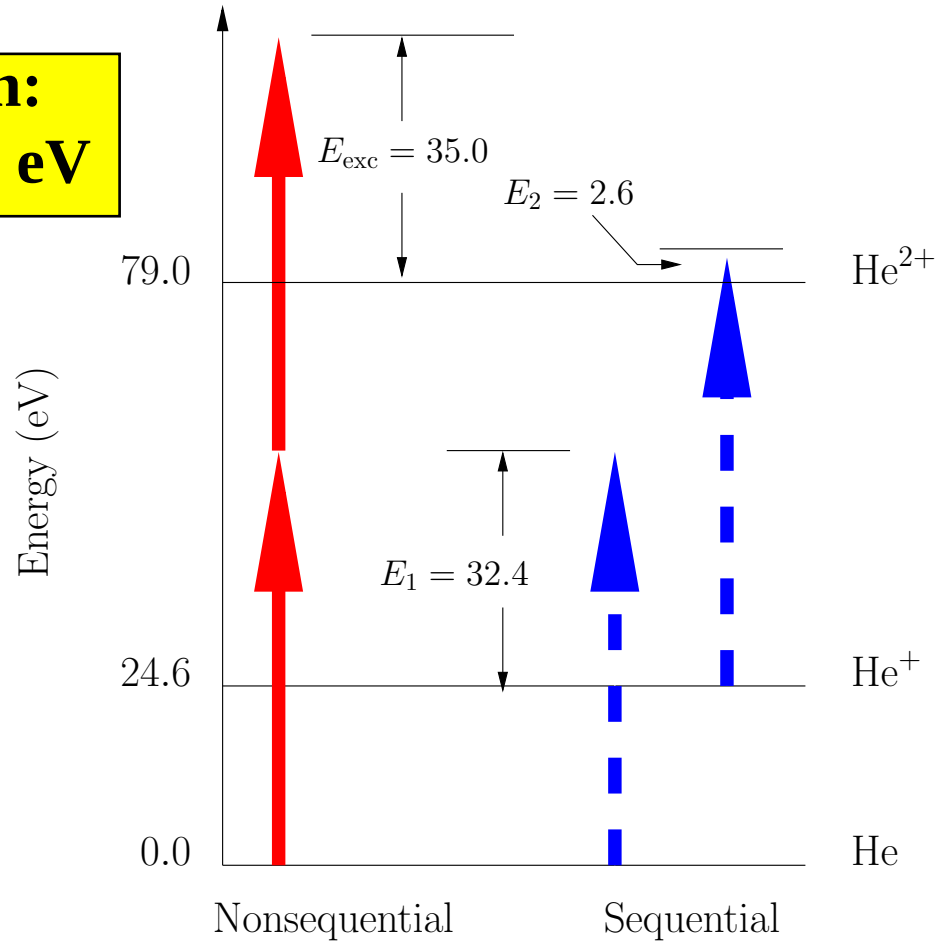
In view of recent papers on two-photon double ionization of helium, pertaining to a perceived anomaly in the behavior of the sequential process and its influence on the direct process, as well as the dependence of both on the laser pulse duration, we show that upon the proper formulation, the sequential is well defined and free of divergence, and that the dependence on pulse duration is considerably more intricate. We also argue that the apparent sharp rise of the cross section for the direct process, around 54.4 eV, is due to the unintended inclusion of the sequential process, which in any case is not properly describable in terms of a single cross section.

DOI: 10.1103/PhysRevA.78.055402 PACS number(s): 32.80.Rm, 32.80.Fb

Application to Two-Photon Double Ionization of Helium

- This is currently a **very controversial topic!**
- Questions include:
  - **Sequential or non-sequential process?**
  - **Final-state correlations** between the two escaping electrons?

illustration:
E_{photon} = 57 eV

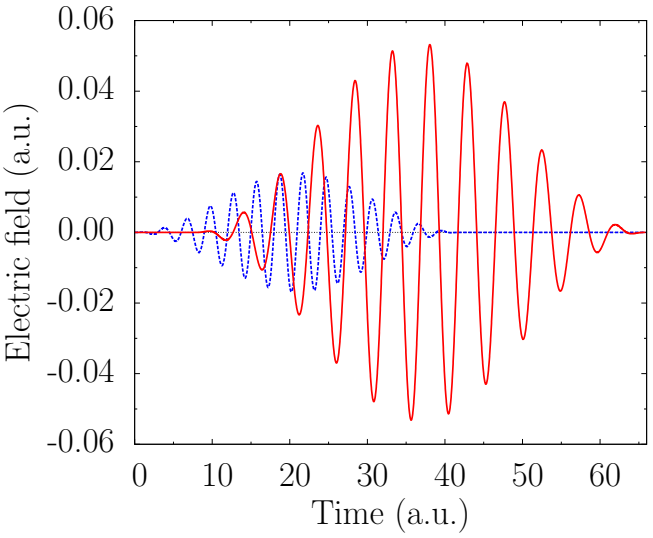


Question:

How long does it take the remaining electron to realize that it is in the wrong ("screened" 1s) quantum state after the first one has left?

This "experiment" can tell us!

Two-Color Double Ionization of Helium with Delay



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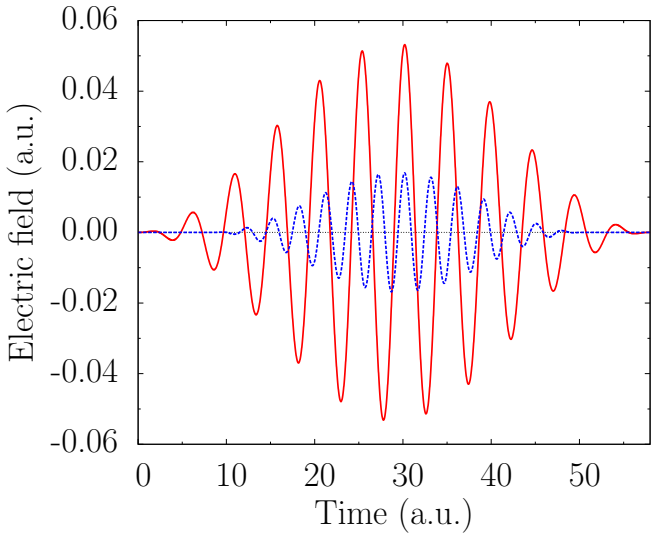

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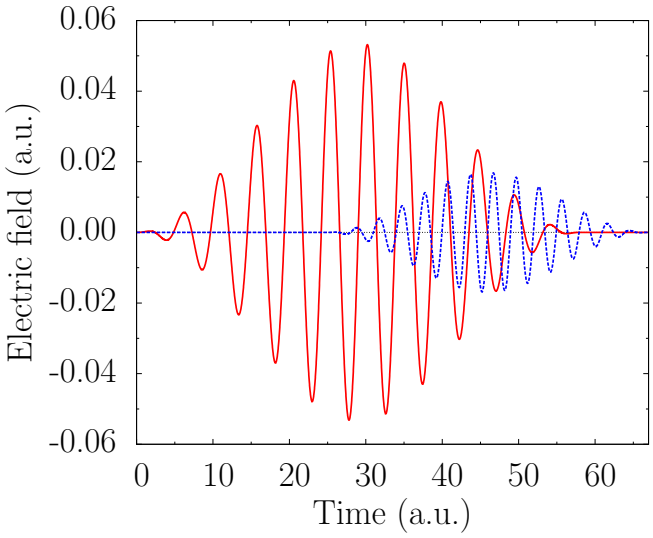
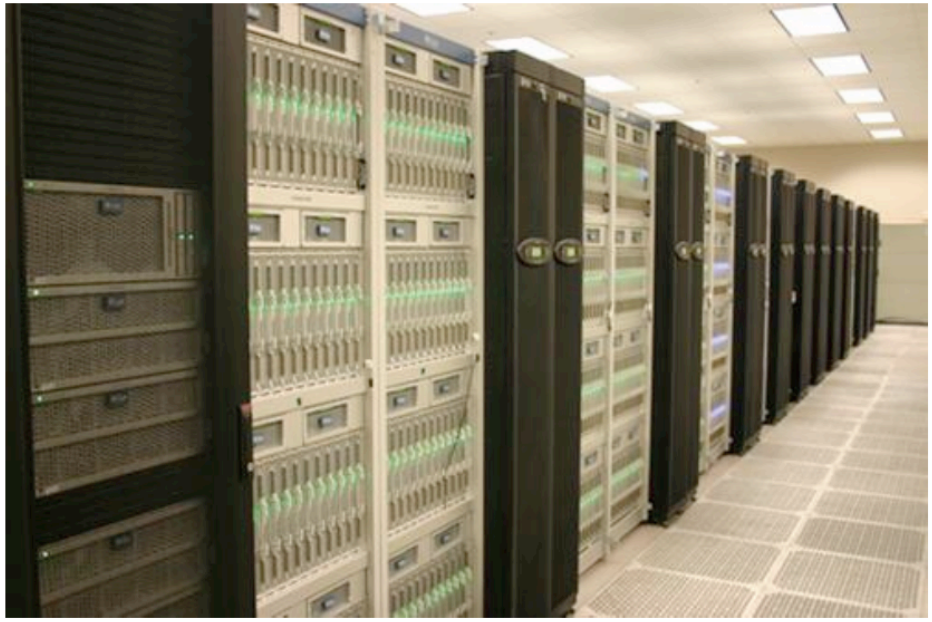


- $\omega_1 = 35.3\,\text{eV}$; $\tau_1 = 12\,\text{o.c.}$; peak intensity of $10^{14}\,\text{W/cm}^2$
- $\omega_2 = 57.1\,\text{eV}$; $\tau_2 = 14\,\text{o.c.}$; peak intensity of $10^{13}\,\text{W/cm}^2$
- The time delays (left to right): -16.5, 0.0, and 16.5 a.u. (≈ 400 atto-seconds)

our apparatus ...

TACC TEXAS ADVANCED COMPUTING CENTER

Ranger User Guide



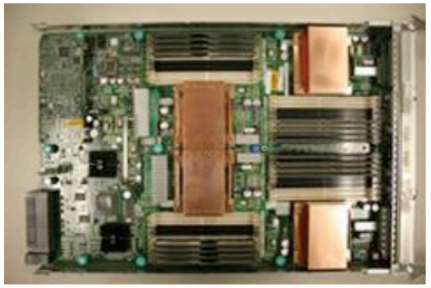
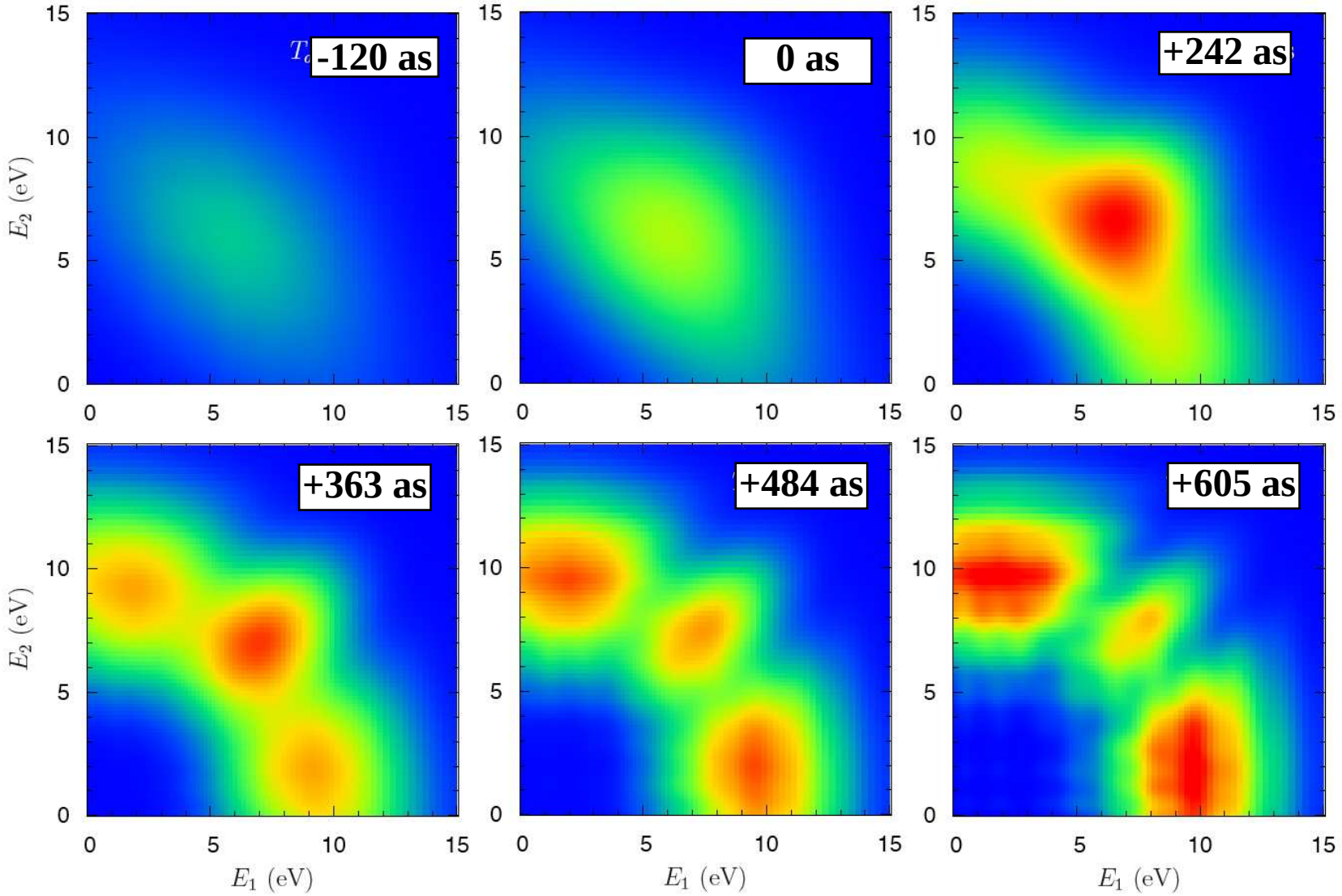


Figure 2. SunBlade x6420 motherboard (compute blade).

Operations		579 TFLOPS (Peak)
Nodes(blades)	Four Quad-Core AMD Opteron processors	3,936 Nodes / 62,976 Cores
Memory	Distributed	123 TB Aggregate
Shared Disk	Lustre, parallel File System	1.7 PB Raw

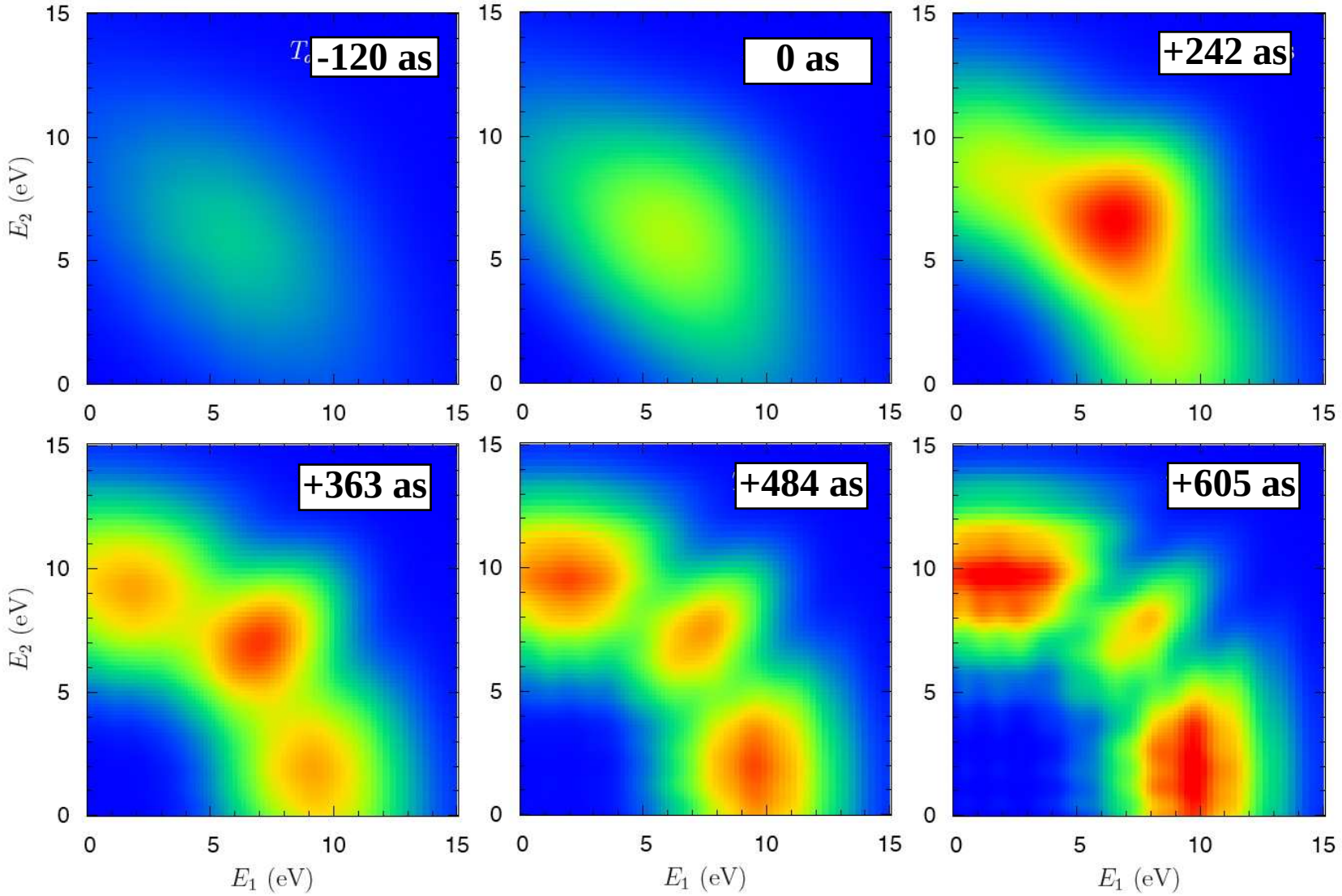
Two-Color Double Ionization of Helium with Variable Delay



- $\omega_1 = 35.3\,\mathrm{eV}$; $\tau_1 = 10\,\mathrm{o.c.}$; peak intensity of $10^{14}\,\mathrm{W/cm^2}$
- $\omega_2 = 57.1\,\mathrm{eV}$; $\tau_2 = 10\,\mathrm{o.c.}$; peak intensity of $10^{13}\,\mathrm{W/cm^2}$
- Time delays between -121 and 605 atto-seconds

expected energies of 10.7 and 2.7 eV for sequential process

Two-Color Double Ionization of Helium with Variable Delay



- $\omega_1 = 35.3\,\mathrm{eV}$; $\tau_1 = 10\,\mathrm{o.c.}$; peak intensity of $10^{14}\,\mathrm{W/cm^2}$
- $\omega_2 = 57.1\,\mathrm{eV}$; $\tau_2 = 10\,\mathrm{o.c.}$; peak intensity of $10^{13}\,\mathrm{W/cm^2}$
- Time delays between -121 and 605 atto-seconds

Answer: about 450 as!

Move on to Double Ionization of the H_2 Molecule in Strong Laser Fields

Single-Photon Absorption $\longrightarrow$ Double Ionization

Science 310 (2005) 1787

Complete Photo-Induced Breakup of the H_2 Molecule as a Probe of Molecular Electron Correlation

Wim Vanroose,[1] Fernando Martín,[2] Thomas N. Rescigno,[3] C. William McCurdy[3,4]

Despite decades of progress in quantum mechanics, electron correlation effects are still only partially understood. Experiments in which both electrons are ejected from an oriented hydrogen molecule by absorption of a single photon have recently demonstrated a puzzling phenomenon: The ejection pattern of the electrons depends sensitively on the bond distance between the two nuclei as they vibrate in their ground state. Here, we report a complete numerical solution of the Schrödinger equation for the double photoionization of H_2. The results suggest that the distribution of photoelectrons emitted from aligned molecules reflects electron correlation effects that are purely molecular in origin.

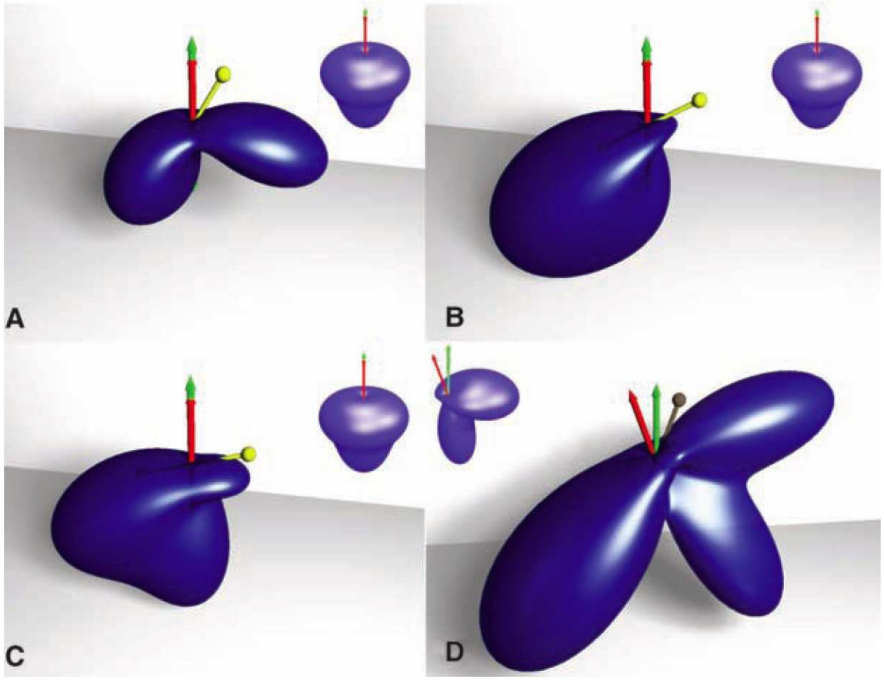


Fig. 3. Effects of molecular orientation on the angular distribution of ejected electrons. For the fixed electron ejected along the polarization direction with 90% of the kinetic energy, the molecule makes an angle with the polarization of (**A**) 30°, (**B**) 60°, and (**C**) 75°, and splits the corresponding pattern for the helium atom (insets) into two lobes which vary in size and ultimately show a tendency to align along the molecular axis as in Fig. 1B. The cross section in (A) is about one-fourth the magnitude of (B) and (C). (**D**) A case in which the molecule and fixed electron have 10% of the kinetic energy, both at 20° from the polarization vector but on opposite sides, yielding an ejection pattern markedly different from the corresponding atomic one.

Two-Photon Absorption ⟶ Double Ionization

IOP Publishing | Journal of Physics B: Atomic, Molecular and Optical Physics

J. Phys. B: At. Mol. Opt. Phys. **42** (2009) 134013 (8pp) doi:10.1088/0953-4075/42/13/134013

Two-photon double ionization of H_2 at 30 eV using exterior complex scaling

F Morales[1], F Martín[1], D A Horner[2], T N Rescigno[3] and C W McCurdy[3,4]

[1] Departamento de Química C-9, Universidad Autónoma de Madrid, 28049 Madrid, Spain
[2] Los Alamos National Laboratory, Theoretical Division, Los Alamos, NM 87545, USA
[3] Lawrence Berkeley National Laboratory, Chemical Sciences, Berkeley, CA 94720, USA
[4] Departments of Applied Science and Chemistry, University of California, Davis, CA 95616, USA

E-mail: felipe.morales@uam.es

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Online at stacks.iop.org/JPhysB/42/134013

IOP FTC

IOP Publishing | Journal of Physics B: Atomic, Molecular and Optical Physics

J. Phys. B: At. Mol. Opt. Phys. **41** (2008) 121002 (6pp) doi:10.1088/0953-4075/41/12/121002

FAST TRACK COMMUNICATION

Two-photon double ionization of the hydrogen molecule

J Colgan[1], M S Pindzola[2] and F Robicheaux[2]

[1] Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
[2] Department of Physics, Auburn University, Auburn, AL 36849, USA

Received 13 May 2008
Published 9 June 2008
Online at stacks.iop.org/JPhysB/41/121002

Move on to Double Ionization of the H_2 Molecule in Strong Laser Fields

II. Theoretical Formulation

- **FE-DVR: Two-Center and Two-Electron Problem in Prolate Spheroidal Coordinates:** $\xi = (r_1 + r_2)/R$, $\eta = (r_1 - r_2)/R$, and $\varphi = \tan^{-1}(y/x)$.
- **The Time-Dependent FE-DVR Approach: Arnoldi-Lanczos propagation**
  Comp. Phys. Commun. **180** (2009) 2401 (imaginary or real time scale)
- **Fixed-Nuclei Approximation**

Move on to Double Ionization of the H_2 Molecule in Strong Laser Fields

Theoretical Formulation

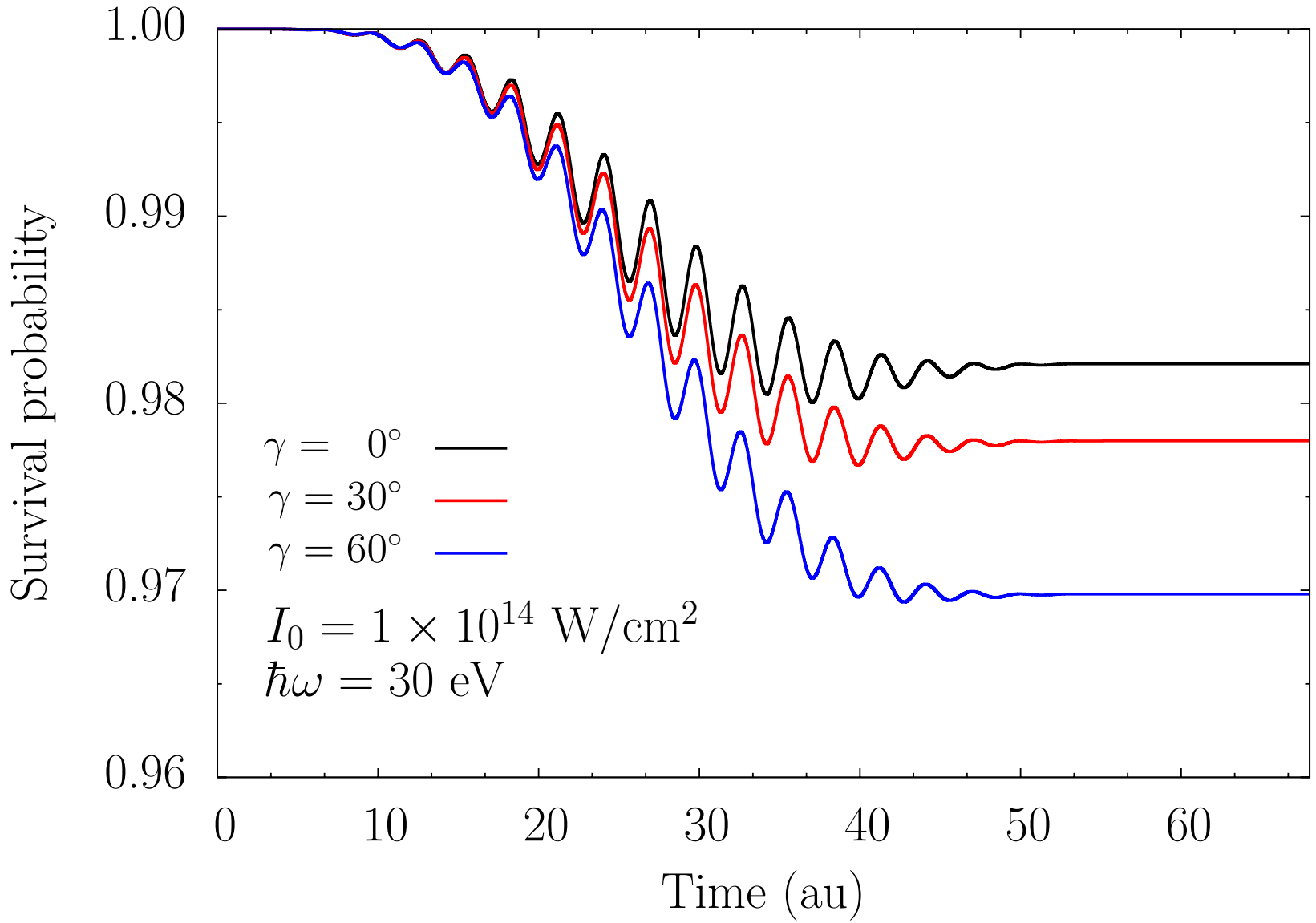
- **FE-DVR: Two-Center and Two-Electron Problem in Prolate Spheroidal Coordinates:** $\xi = (r_1 + r_2)/R$, $\eta = (r_1 - r_2)/R$, and $\varphi = \tan^{-1}(y/x)$.
- **The Time-Dependent FE-DVR Approach: Arnoldi-Lanczos propagation**
  Comp. Phys. Commun. **180** (2009) 2401 (imaginary or real time scale)
- **Fixed-Nuclei Approximation**

Results:

- **Numerical Aspects**
- $\hbar\omega = 30$ **eV,** $I_0 = 1 \times 10^{14}$ **W/cm^2, and** $\tau = 10$ **optical cycles**
- **Survival Probability of the Initial State**
- **Triple-Differential Cross Section**

RESULTS:

Dependence on relative orientation between laser polarization axis and molecular axis



Note the scale factors! ECS x 0.5; TDCC x 2.0 !!!

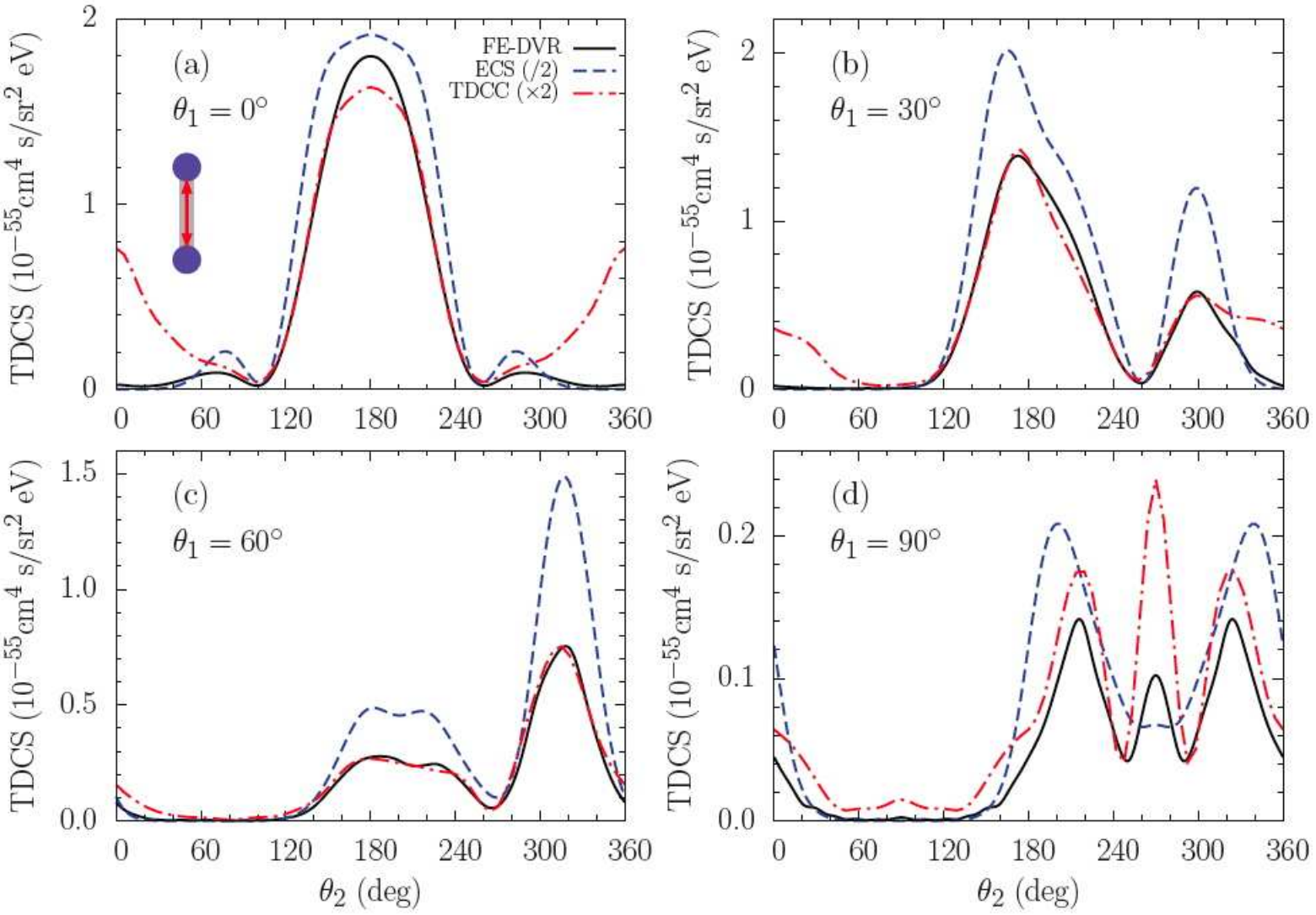


FIG. 2. (Color online) Coplanar TDCS for two-photon DI of H_2 at equal energy sharing ($E_1 = E_2 = 4.3$ eV) of the two ejected electrons in the parallel geometry.

BIG DISCREPANCIES for the small TDCS values!

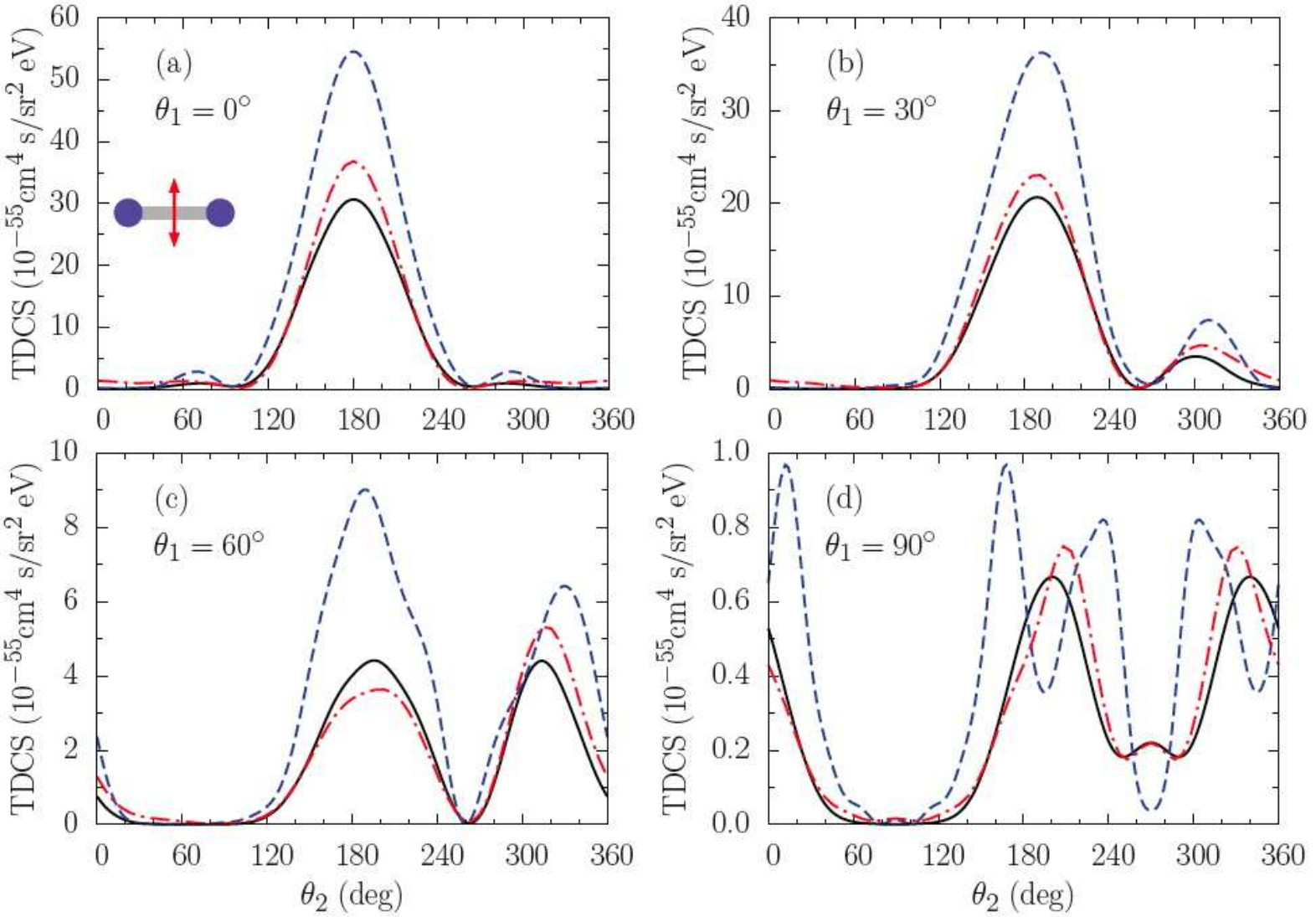


FIG. 3. (Color online) Same as Fig. 2, but for the perpendicular geometry. No scaling factors were applied to compare the various predictions.

RAPID COMMUNICATIONS

PHYSICAL REVIEW A **82**, 041404(R) (2010)

Two-photon double ionization of H_2 in intense femtosecond laser pulses

Xiaoxu Guan,[1] Klaus Bartschat,[1] and Barry I. Schneider[2]

[1]*Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA*

[2]*Physics Division, National Science Foundation, Arlington, Virginia 22230, USA*

(Received 23 July 2010; published 20 October 2010)

Triple-differential cross sections for two-photon double ionization of molecular hydrogen are presented for a central photon energy of 30 eV. The calculations are based on a fully *ab initio*, nonperturbative approach to the time-dependent Schrödinger equation in prolate spheroidal coordinates, discretized by a finite-element discrete-variable representation. The wave function is propagated in time for a few femtoseconds using the short, iterative Lanczos method to study the correlated response of the two photoelectrons to short, intense laser radiation. The current results often lie in between those of Colgan *et al.* [J. Phys. B **41**, 121002 (2008)] and Morales *et al.* [J. Phys. B **42**, 134013 (2009)]. However, we argue that these individual predictions should not be compared directly with each other, but preferably with experimental data generated under well-defined conditions.

DOI: 10.1103/PhysRevA.82.041404 PACS number(s): 33.80.Wz, 31.15.A−

Even the one-photon case is suddenly less clear ...

Phys. Rev. A 83, 043403

Breakup of the aligned H_2 molecule by XUV laser pulses: A time-dependent treatment in prolate spheroidal coordinates

Xiaoxu Guan,[1] Klaus Bartschat,[1,*] and Barry I. Schneider[2]
[1]*Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA*
[2]*Physics Division, National Science Foundation, Arlington, Virgina 22230, USA*
(Received 7 January 2011; published xxxxx)

We have carried out calculations of the triple-differential cross section for one-photon double ionization of molecular hydrogen for a central photon energy of 75 eV, using a fully *ab initio*, nonperturbative approach to solve the time-dependent Schrödinger equation in prolate spheroidal coordinates. The spatial coordinates ξ and η are discretized in a finite-element discrete-variable representation. The wave packet of the laser-driven two-electron system is propagated in time through an effective short iterative Lanczos method to simulate the double ionization of the hydrogen molecule. For both symmetric and asymmetric energy sharing, the present results agree to a satisfactory level with most earlier predictions for the absolute magnitude and the shape of the angular distributions. A notable exception, however, concerns the predictions of the recent time-independent calculations based on the exterior complex scaling method in prolate spheroidal coordinates [L. Tao *et al.*, Phys. Rev. A **82**, 023423 (2010)]. Extensive tests of the numerical implementation were performed, including the effect of truncating the Neumann expansion for the dielectronic interaction on the description of the initial bound state and the predicted cross sections. We observe that the dominant escape mode of the two photoelectrons depends dramatically on the energy sharing. In the parallel geometry, when the ejected electrons are collected along the direction of the laser polarization axis, back-to-back escape is the dominant channel for strongly asymmetric energy sharing, while it is completely forbidden if the two electrons share the excess energy equally.

DOI: 10.1103/PhysRevA.00.003400 PACS number(s): 33.80.Wz, 31.15.A−

New ECS predictions differ from the rest; revised TDCC agree well with time-dependent FEDVR (Phys. Rev. A 83, 043403)

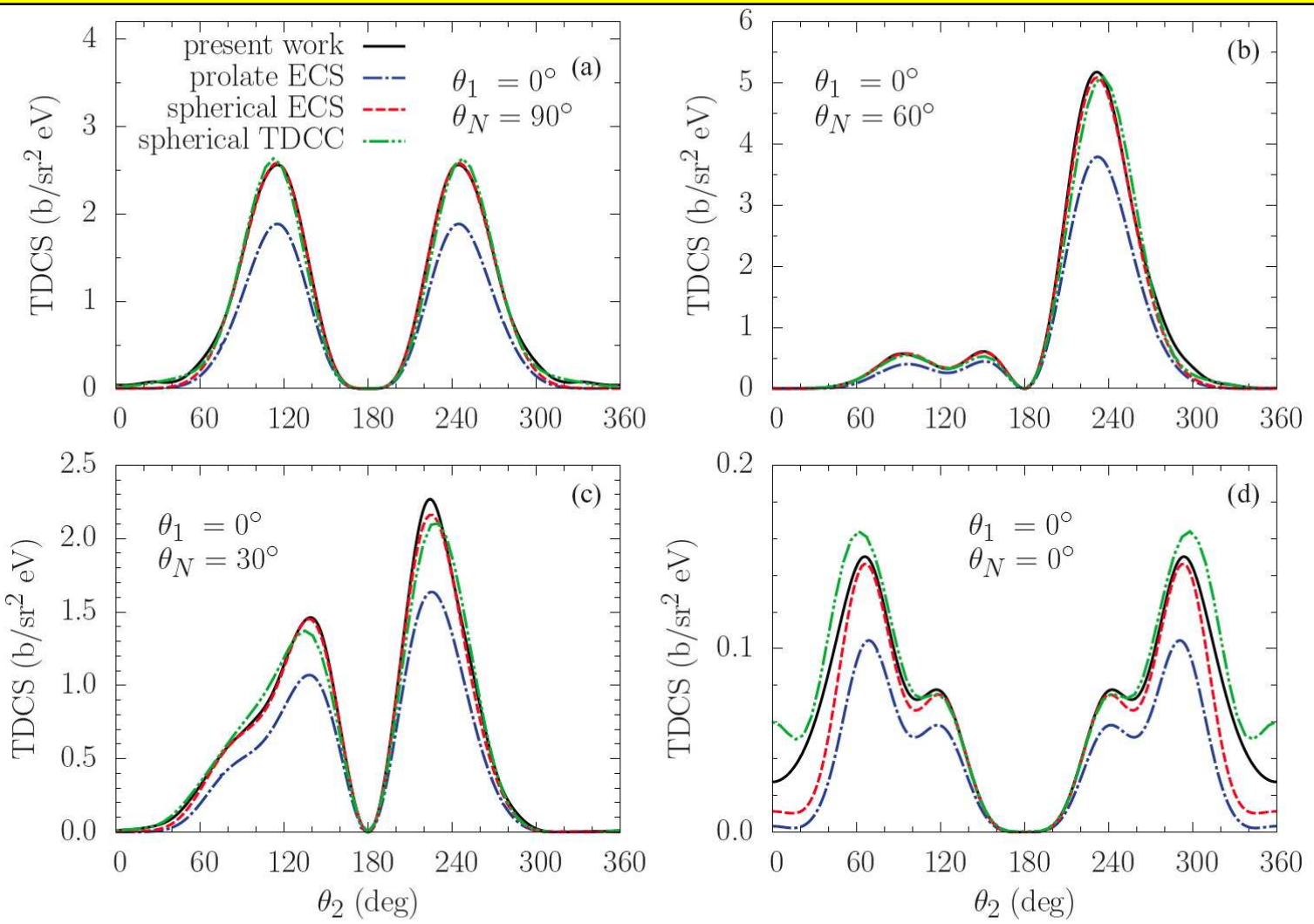


FIG. 5. (Color online) Coplanar TDCS of the aligned hydrogen molecule for equal energy sharing ($E_1 = E_2 = 11.8$ eV). The central photon energy is 75 eV. One electron is detected at the fixed direction of $\theta_1 = 0°$ with respect to the laser polarization axis. Also shown are the one-center spherical ECS results [11], two-center prolate spheroidal results [15], and one-center spherical TDCC results [14].

Finally (this is the last bit): Charged-Particle Impact

Complete Breakup of the Helium Atom by Proton and Antiproton Impact

Xiaoxu Guan* and Klaus Bartschat†

Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA

(Received 5 June 2009; published 17 November 2009)

We present a fully *ab initio*, nonperturbative, time-dependent approach to describe single and double ionization of helium by proton and antiproton impact. The problem is discretized by a flexible finite-element discrete-variable representation on the radial grid. Good agreement with the most recent experimental data for absolute angle-integrated cross sections is obtained for projectile energies between 3 keV and 6 MeV. Also, angle-differential cross sections for two-electron ejection are predicted for a proton impact energy of 6 MeV. The time evaluation of the ionization process is portrayed by displaying the electron density as a function of the projectile location.

DOI: 10.1103/PhysRevLett.103.213201 PACS numbers: 34.50.Fa, 25.40.Ep, 25.43.+t, 36.10.−k

total cross section for single and double ionization of He by anti-proton impact

PRL **103,** 213201 (2009)

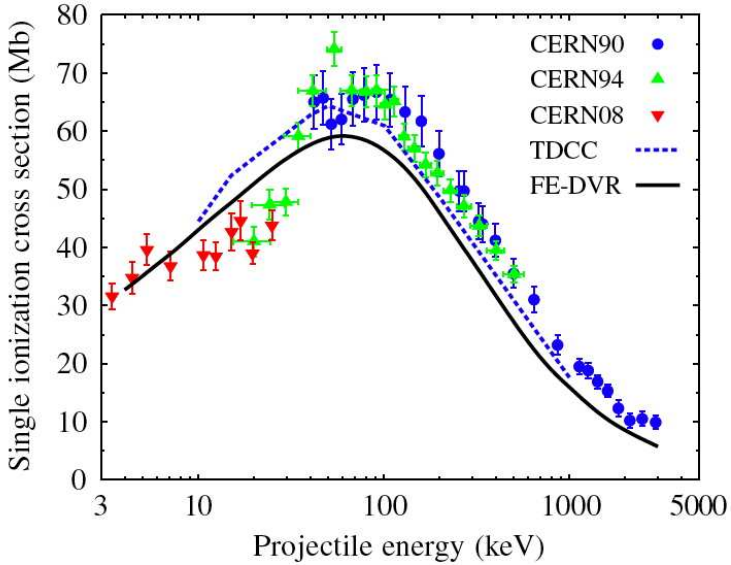


FIG. 1 (color online). Cross section for single ionization helium by antiproton impact. Experimental data obtained CERN by Andersen *et al.* [12] (CERN90), Hvelplund *et* [13] (CERN94), and Knudsen *et al.* [1] (CERN08) are compa with TDCC [3] and the present FE-DVR predictions. Since t were retracted in Ref. [1], the two lowest energy points measu in Ref. [13] are not shown.

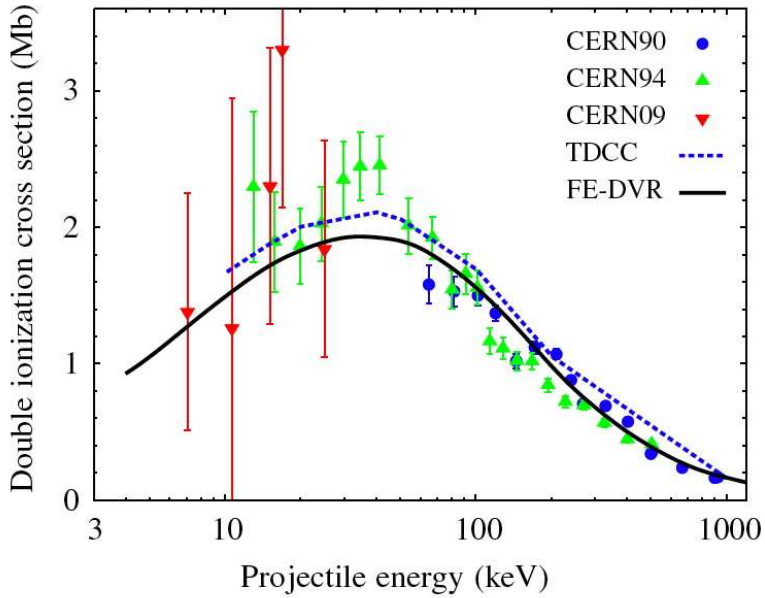
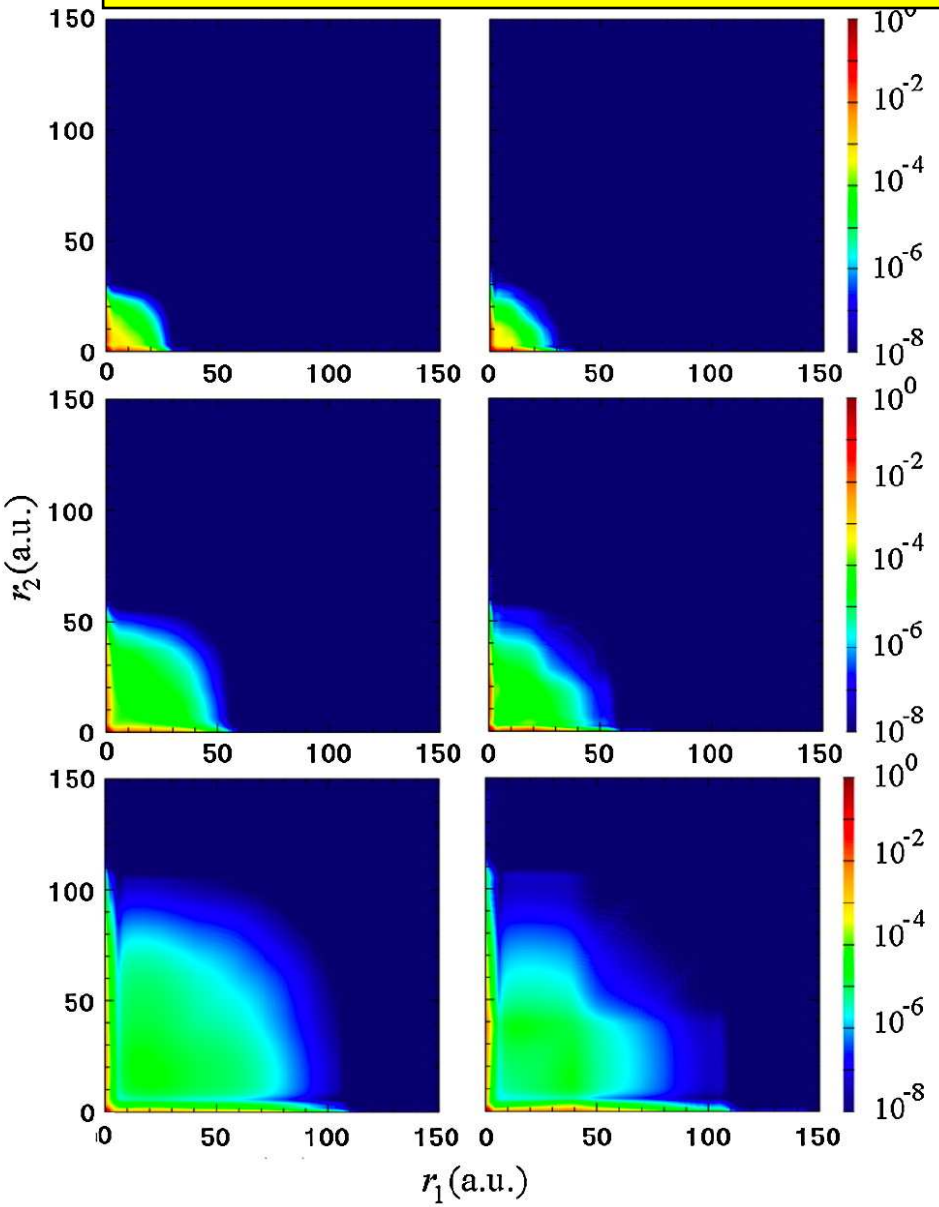


FIG. 2 (color online). Cross section for double ionization of helium by antiproton impact. The experimental data obtained at CERN by Andersen *et al.* [12] (CERN90), Hvelplund *et al.* [13] (CERN94), and Knudsen *et al.* [2] (CERN09) are compared with TDCC [3] and the present FE-DVR predictions.

Electron density distributions as a function of anti-proton position (10, 20, 40 bohr behind the target)

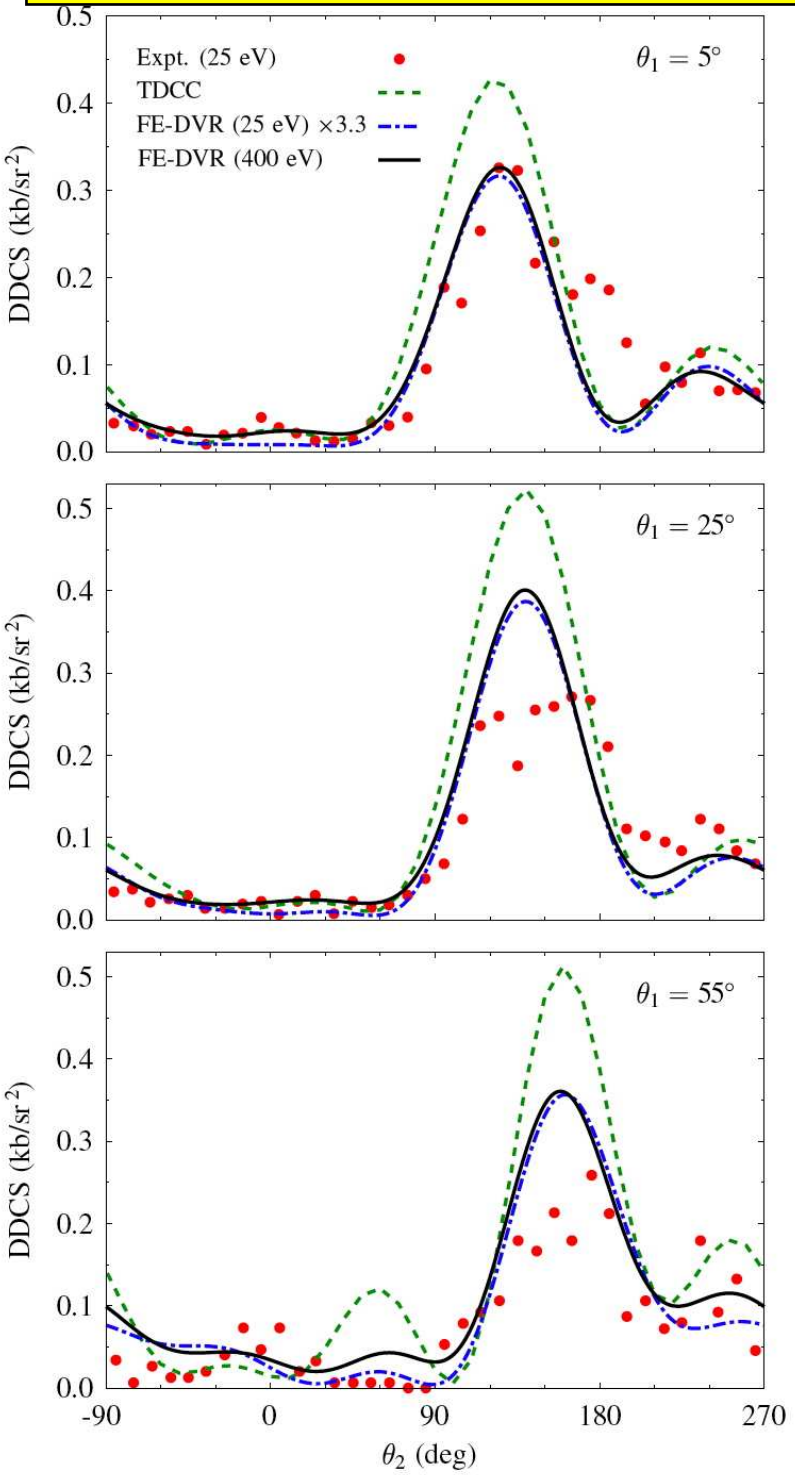


anti-protons **protons**

PRL **103,** 213201 (2009)

FIG. 3 (color online). Radial electron density after antiproton (left panels) and proton (right panels) impact on helium at an energy of 100 keV for an impact parameter of $0.5a_0$. Starting from an initial distance of $-50a_0$, the positions of the projectile shown in the snapshots, from top to bottom, are $+10$, $+20$, and $+40a_0$ relative to the center of the target, respectively.

DDCS for double ionization of He by proton impact (experiment and analysis: Schulz and collaborators)



PRL **103,** 213201 (2009)

FIG. 4 (color online). DDCS for proton impact double ionization of helium for an incident energy of 6 MeV, a fixed detection angle θ_1 for one of the electrons, and a variable detection angle θ_2 for the second electron. The experimental measurements of Refs. [17,18] were normalized to our converged DDCS at the large peak for $\theta_1 = 5°$. Also shown are the corrected TDCC results [19].

Conclusions and Outlook

- Our implementation of the matrix iteration method allows for the calculation of **numerically stable results** for the interaction of a short-pulse laser with the hydrogen atom.
- Although the principle of attacking this problem is well known, it remains a **challenge for the case of intense infrared radiation**, due to the large number of photons that need to be absorbed for the electron to be ionized.
- After confirming results from previous work, **we extended the parameter space into previously unchartered territory.**

- We are further developing a **general method for treating the interaction of a strong attosecond laser pulse with a complex atom.**
- The approach **combines** a highly flexible ***B*-spline *R*-matrix method** for the time-independent problem **with an efficient Arnoldi-Lanczos scheme** for the time propagation of the TDSE.
- The **major advantages** of the method are:
  - its **generality** and applicability to any complex many-electron target;
  - the possibility of generating **highly accurate target and continuum descriptions** with relatively small configuration interaction expansions.

- We are further developing an **FE-DVR approach to treat one- and two-electron systems.**
- The **major advantages** of the method include its **flexibility and numerical accuracy.**

Future Plans

- **Computational and Numerical Aspects:**
  - continuously analyze and improve the **numerical efficiency** of the method;
  - optimize the time propagation:
    - how much of the matrix do we really need?
    - non-orthogonal Arnoldi-Lanczos?
    - it seems **unlikely that a single method will do everything.**
- **Physics:**
  - investigate other **pump-probe processes** as a function of the time delay
  - tackle **multi-photon single and double ionization** of a complex target atom
  - investigate other **alignment effects in molecules**
  - move on to **diatomic molecules beyond H_2**
  - investigate possible effects of **nuclear motion**

Thank You!