Strong-field atomic ionization with XUV pulses:

A finite displacement effect

Free-electron displacement

$$x = \int_0^T \boldsymbol{A}(t)\,dt \neq 0$$

Free-electron kick

Lawson–Woodward theorem, 1979.

$$p = \int_0^T \boldsymbol{E}(t)\,dt \propto \frac{\omega}{c} \to 0$$

Anatoli Kheifets

THE AUSTRALIAN NATIONAL UNIVERSITY

KITP 2014

Outline

- Strong-field ionization in XUV
  - Resonant ATI spectra of atomic hydrogen
    - *Autler-Townes doublet*
- Pulse shape effects
  - Ramp-on/ramp-off
    - *$\sin^2$ vs. trapezoidal*
  - Photo-electron spectra
    - *Line distortion*
    - *Angular momentum / asymmetry*
- Kramers-Henneberger atom
  - Final displacement effect
- Implications
  - Theory, experiment

Effects of numerical approximations in the treatment of short-pulse strong-field ionization of atomic hydrogen

Phys. Rev. A **88**, 055401 – Published 5 November 2013

Alexei N. Grum-Grzhimailo, Mikhail N. Khaerdinov, and Klaus Bartschat

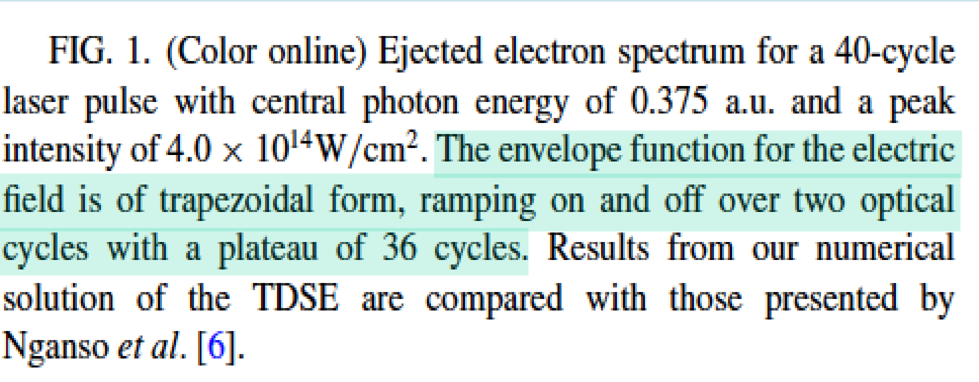
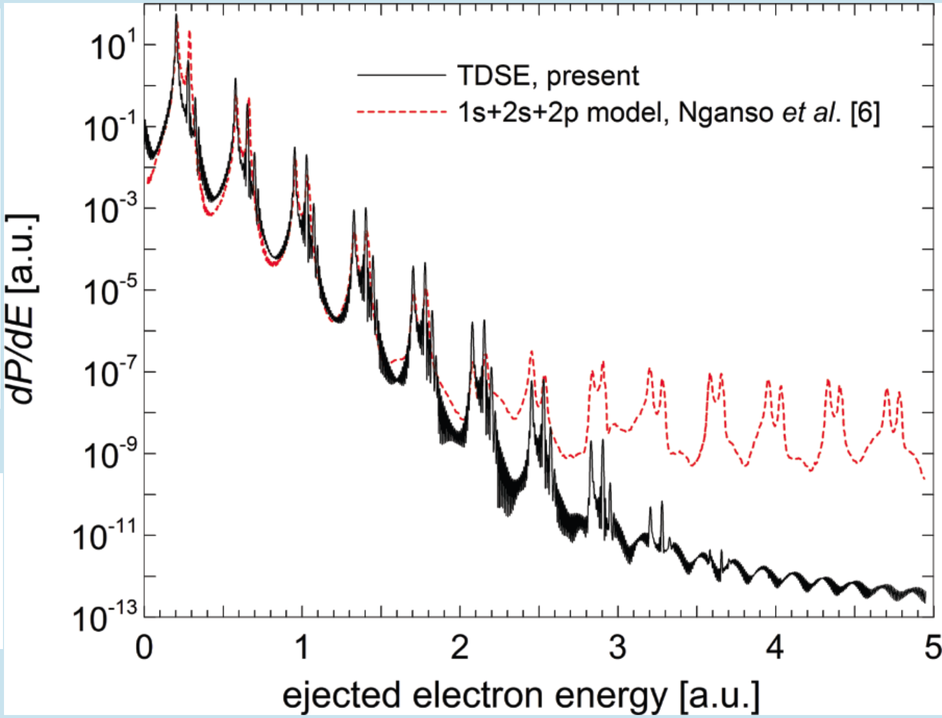
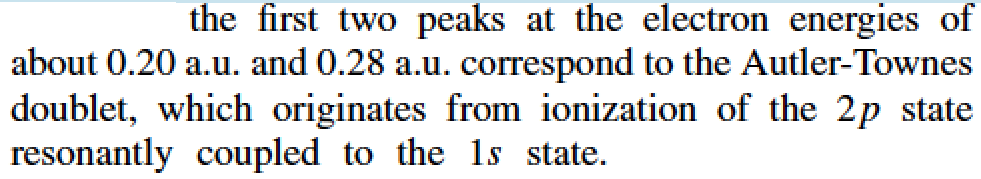
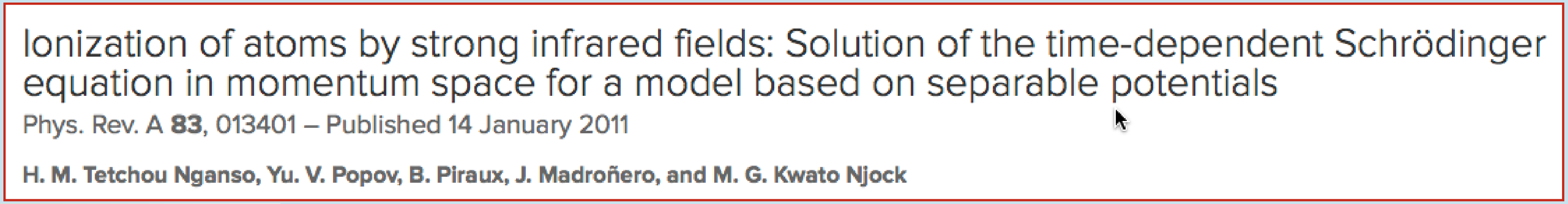


FIG. 1. (Color online) Ejected electron spectrum for a 40-cycle laser pulse with central photon energy of 0.375 a.u. and a peak intensity of $4.0 \times 10^{14} \mathrm{W/cm^2}$. The envelope function for the electric field is of trapezoidal form, ramping on and off over two optical cycles with a plateau of 36 cycles. Results from our numerical solution of the TDSE are compared with those presented by Nganso *et al.* [6].





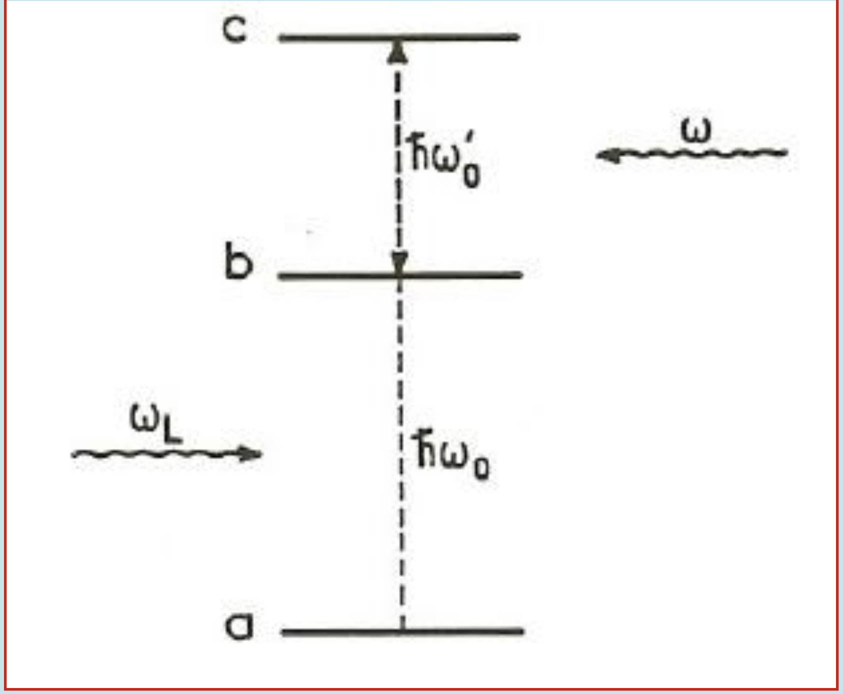
the first two peaks at the electron energies of about 0.20 a.u. and 0.28 a.u. correspond to the Autler-Townes doublet, which originates from ionization of the $2p$ state resonantly coupled to the $1s$ state.

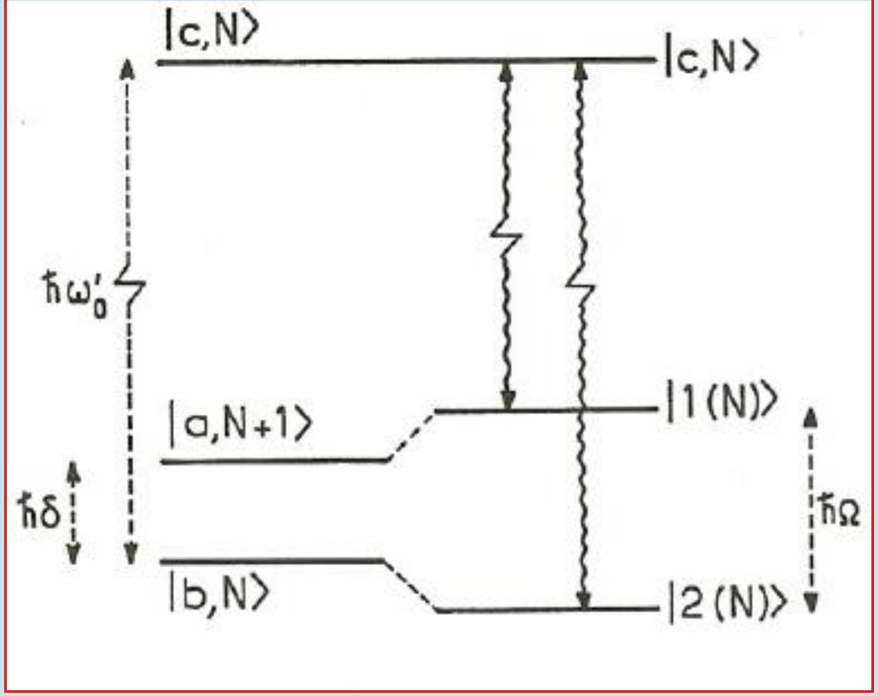


Ionization of atoms by strong infrared fields: Solution of the time-dependent Schrödinger equation in momentum space for a model based on separable potentials

Phys. Rev. A **83**, 013401 – Published 14 January 2011

H. M. Tetchou Nganso, Yu. V. Popov, B. Piraux, J. Madroñero, and M. G. Kwato Njock



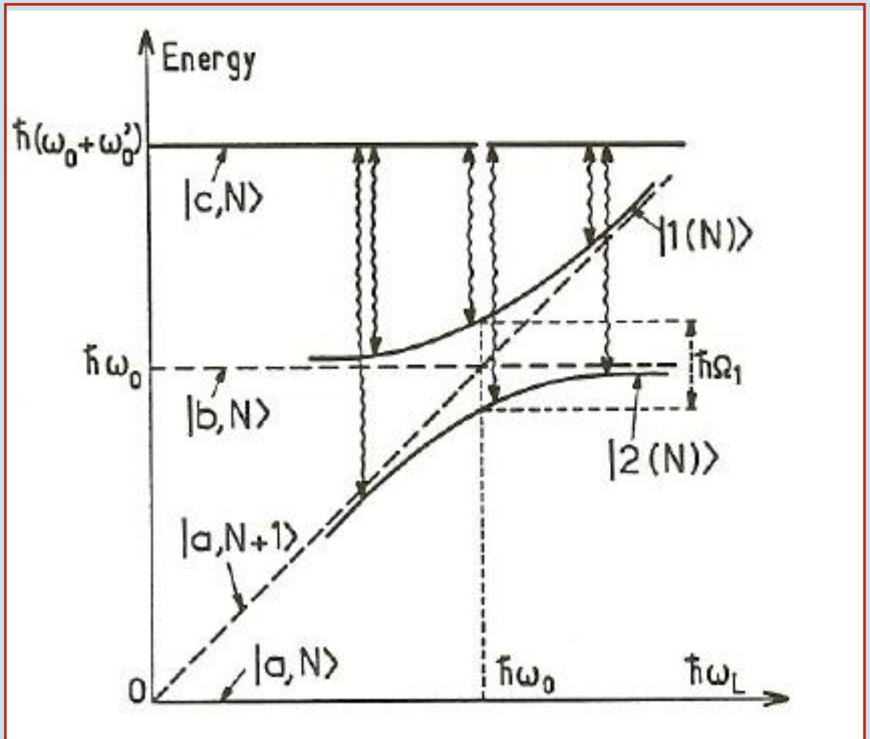


C. Cohen-Tannoudji,

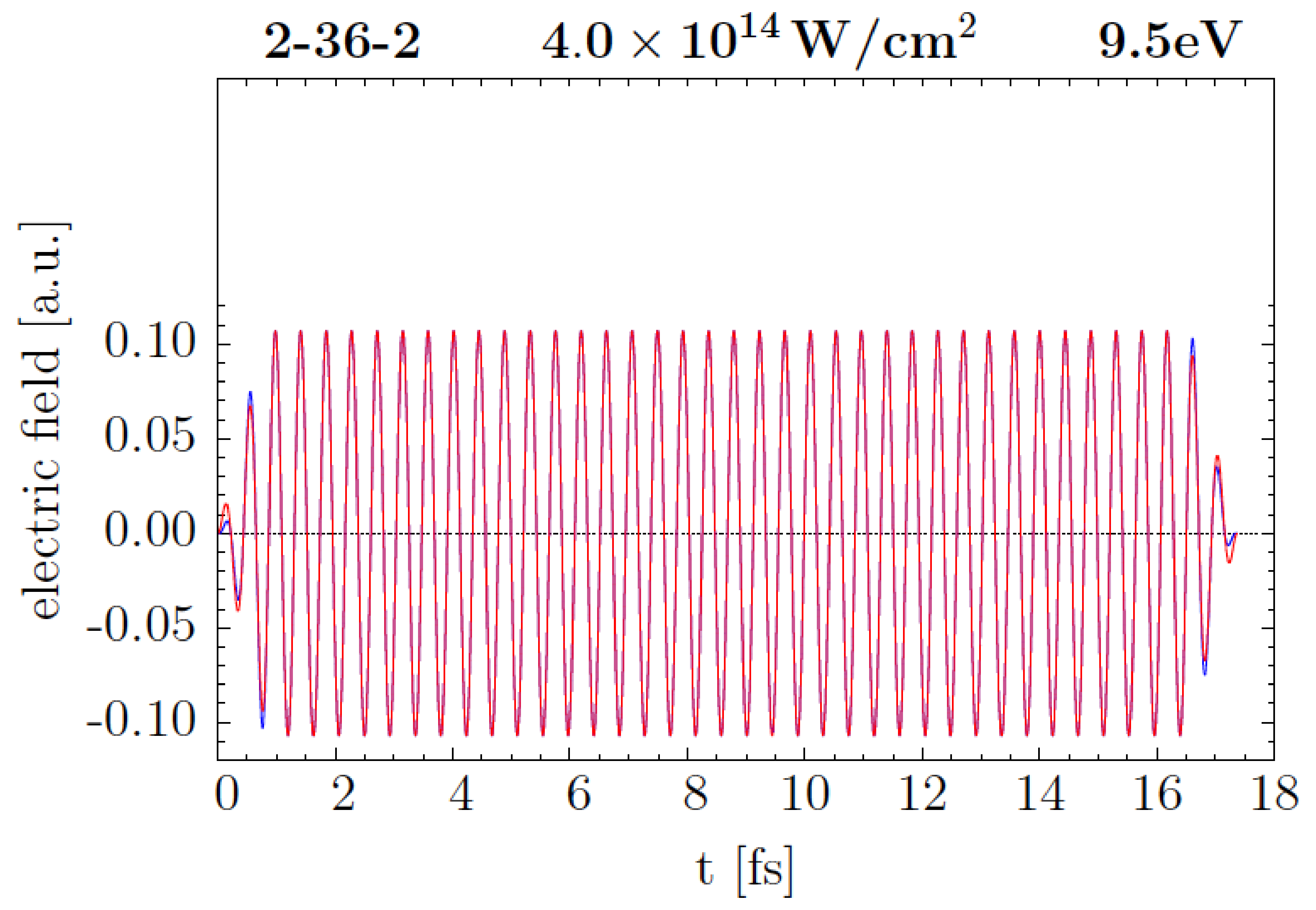
The Autler-Townes effect revisited

In *Amazing Light : a volume dedicated to Charles Hard Townes on his 80th birthday*,

ed. by R. Y. Chiao, p. 109 (Springer, 1996)

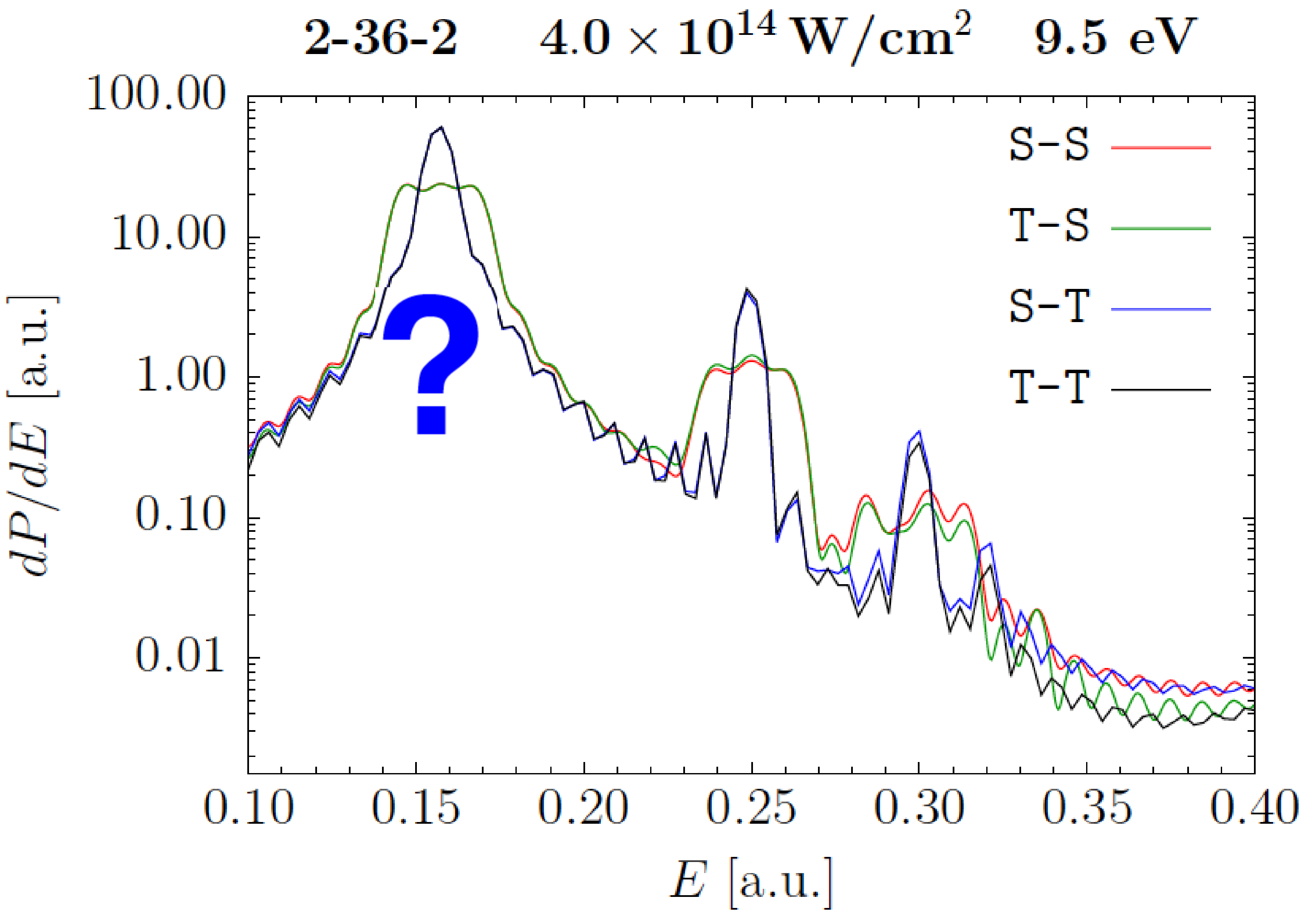


Pulse shape effect



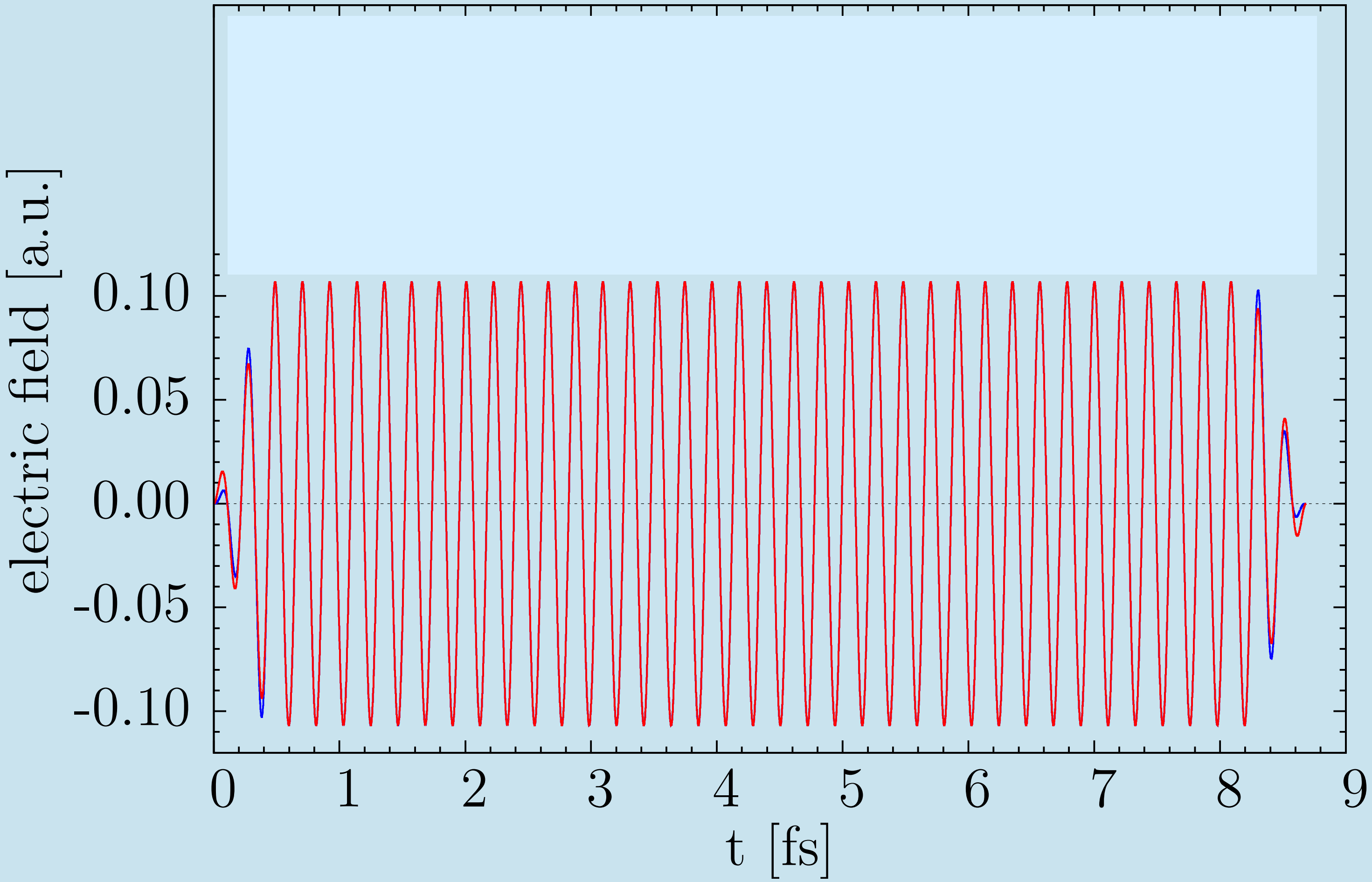
S-S = $\sin^2$ ramp-on/off; T-T = linear ramp-on/off.

Photoelectron spectrum

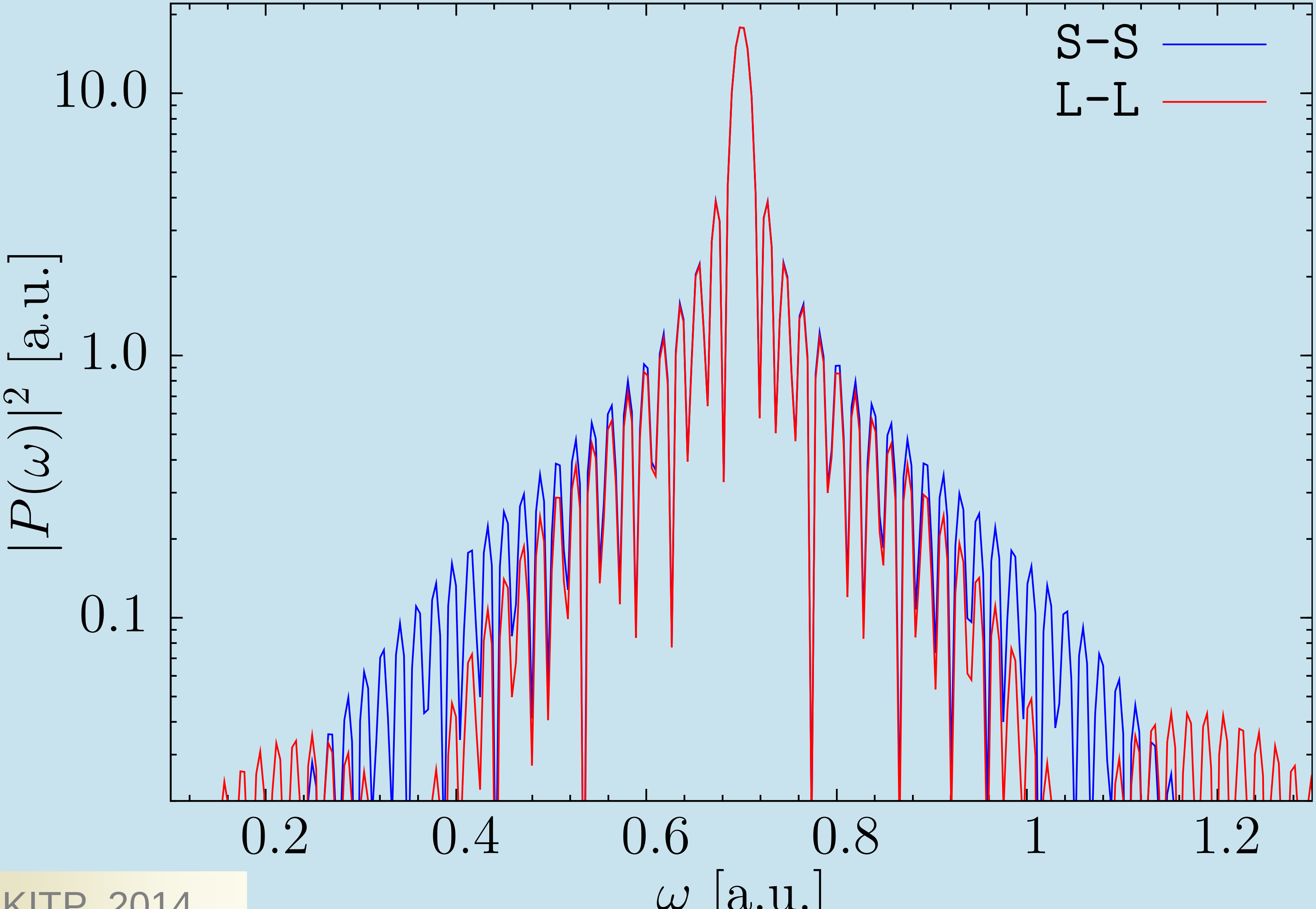


Pulse shape effect: away from resonance

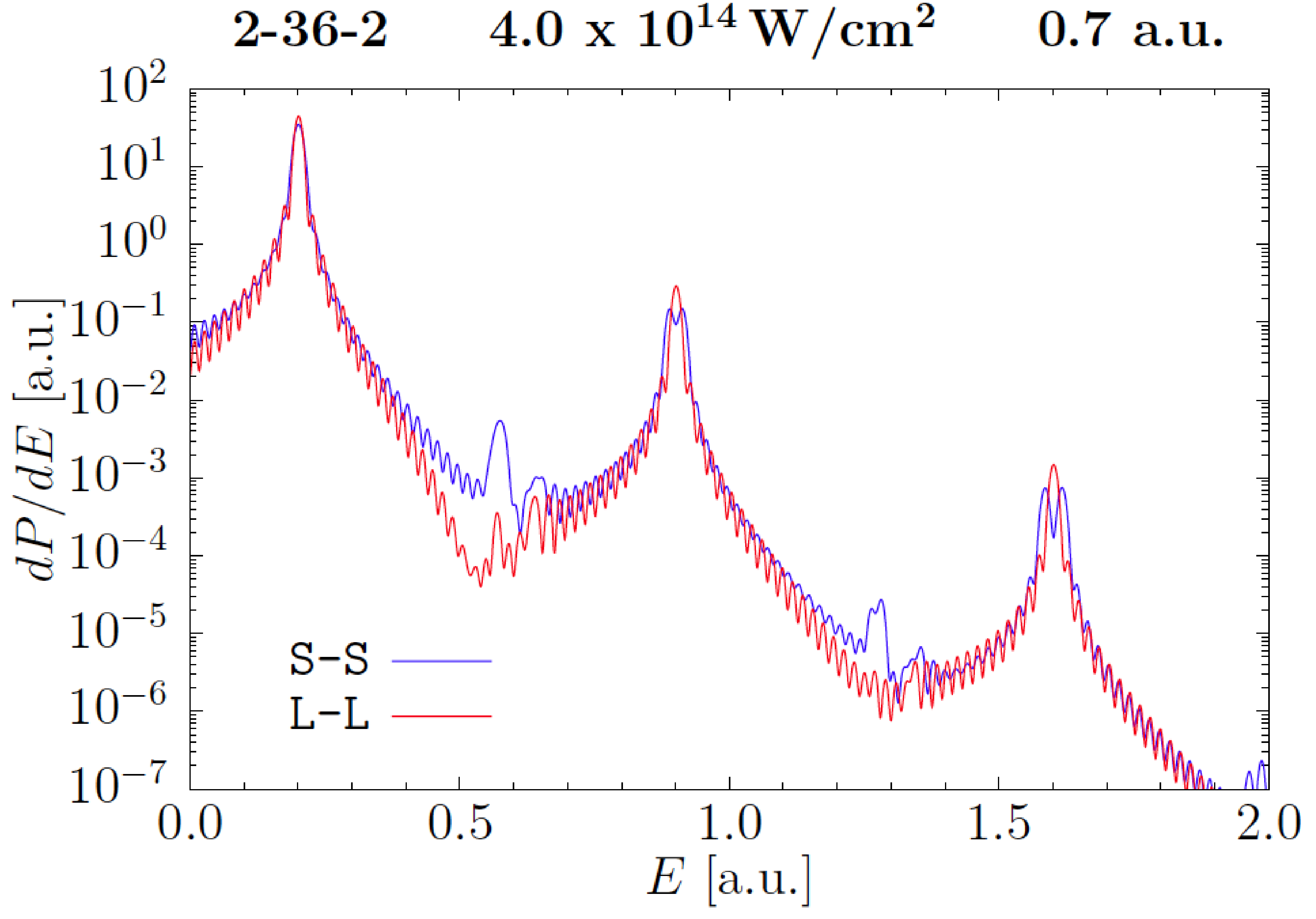
2-36-2 **4.0×10^{14} W/cm^2** **0.7 a.u.**



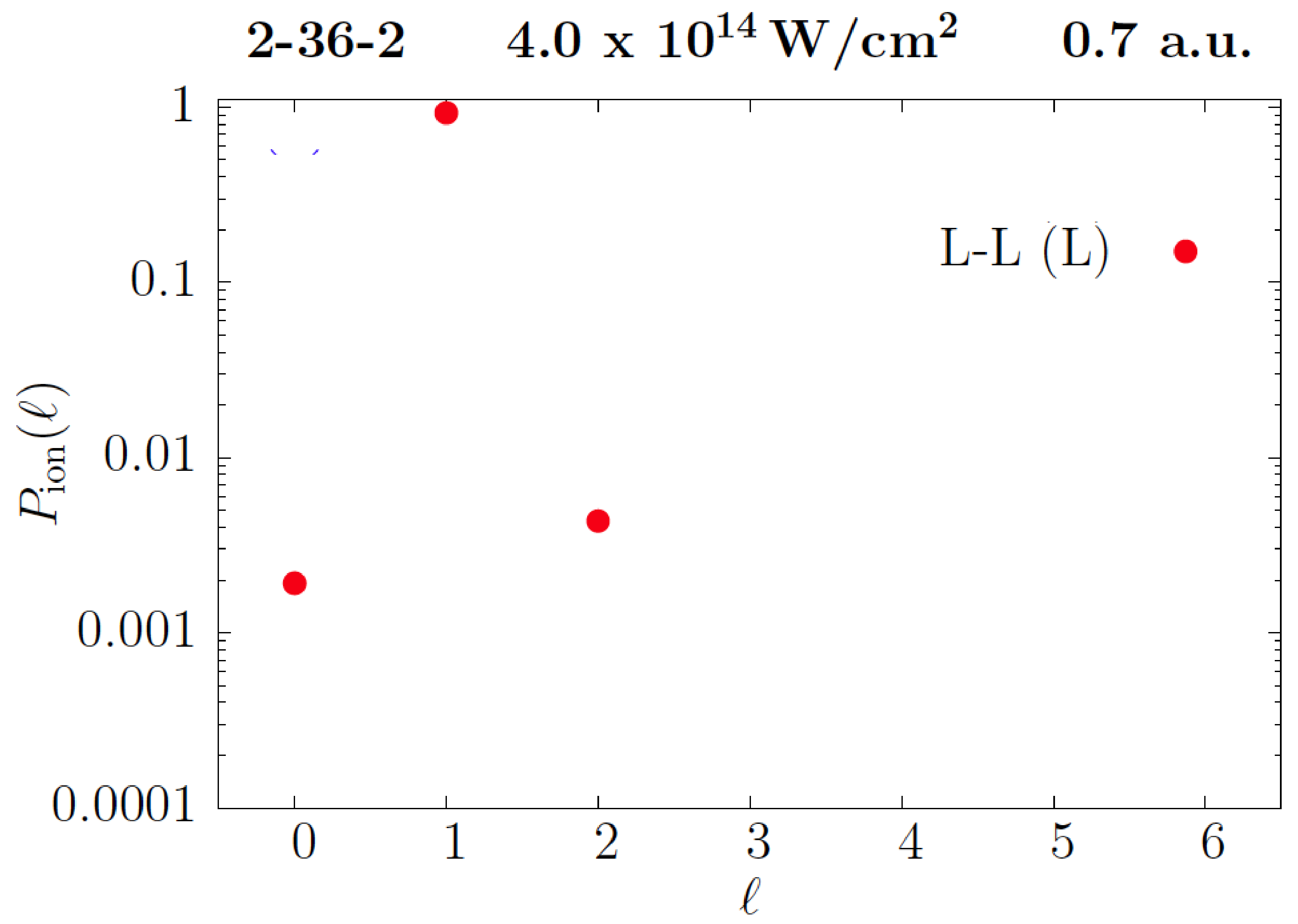
Spectral content



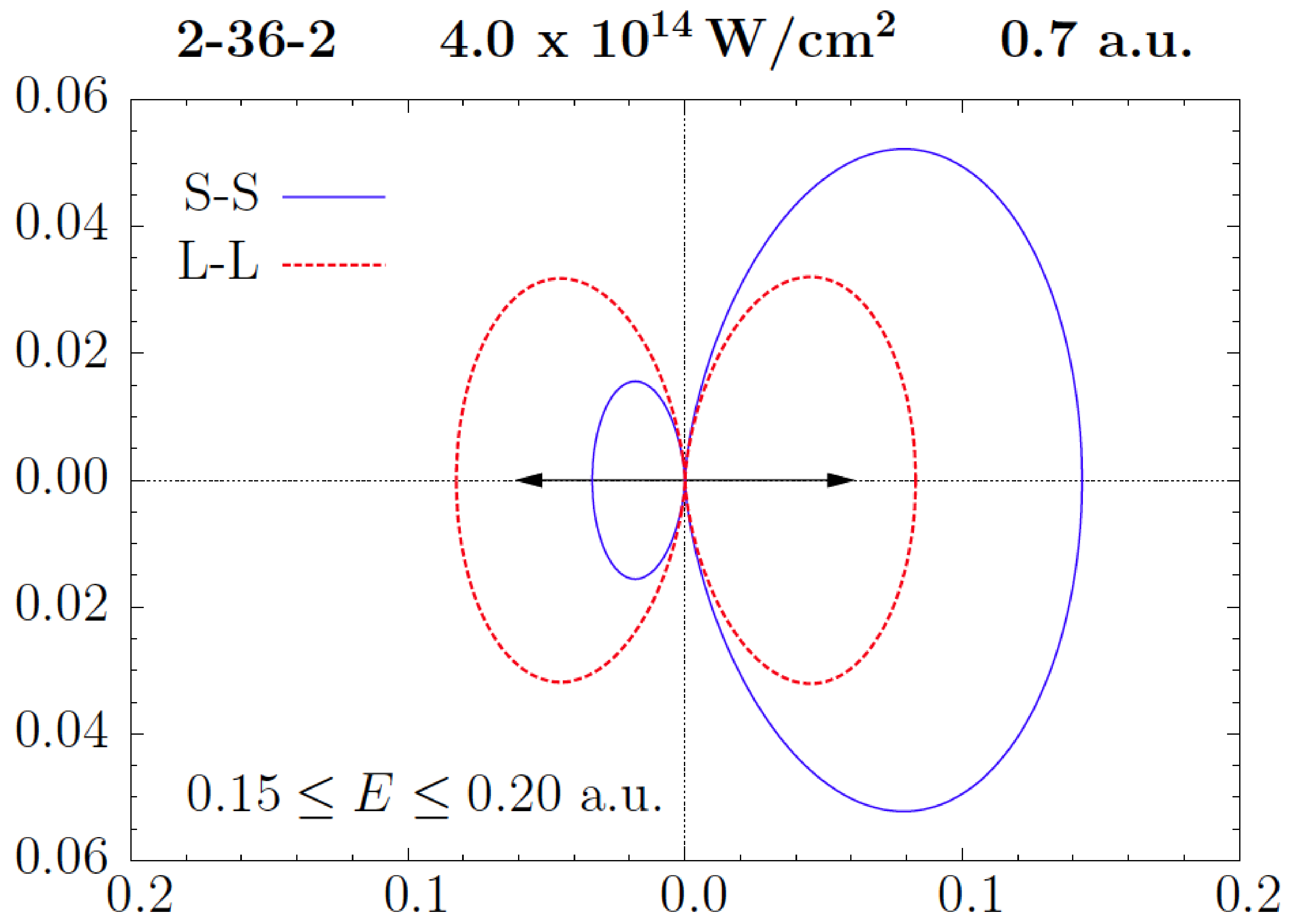
Photoelectron spectrum



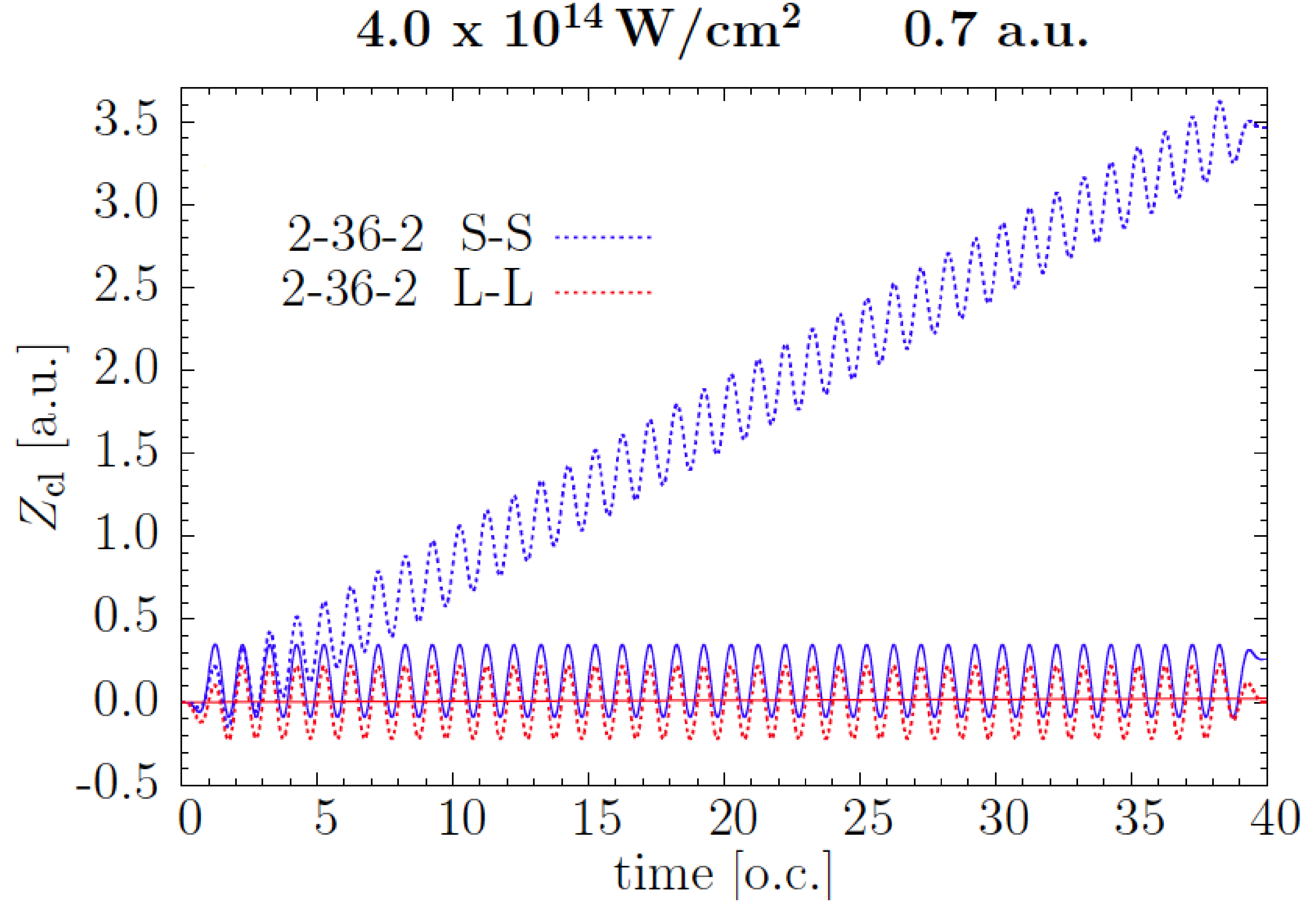
Angular momentum composition



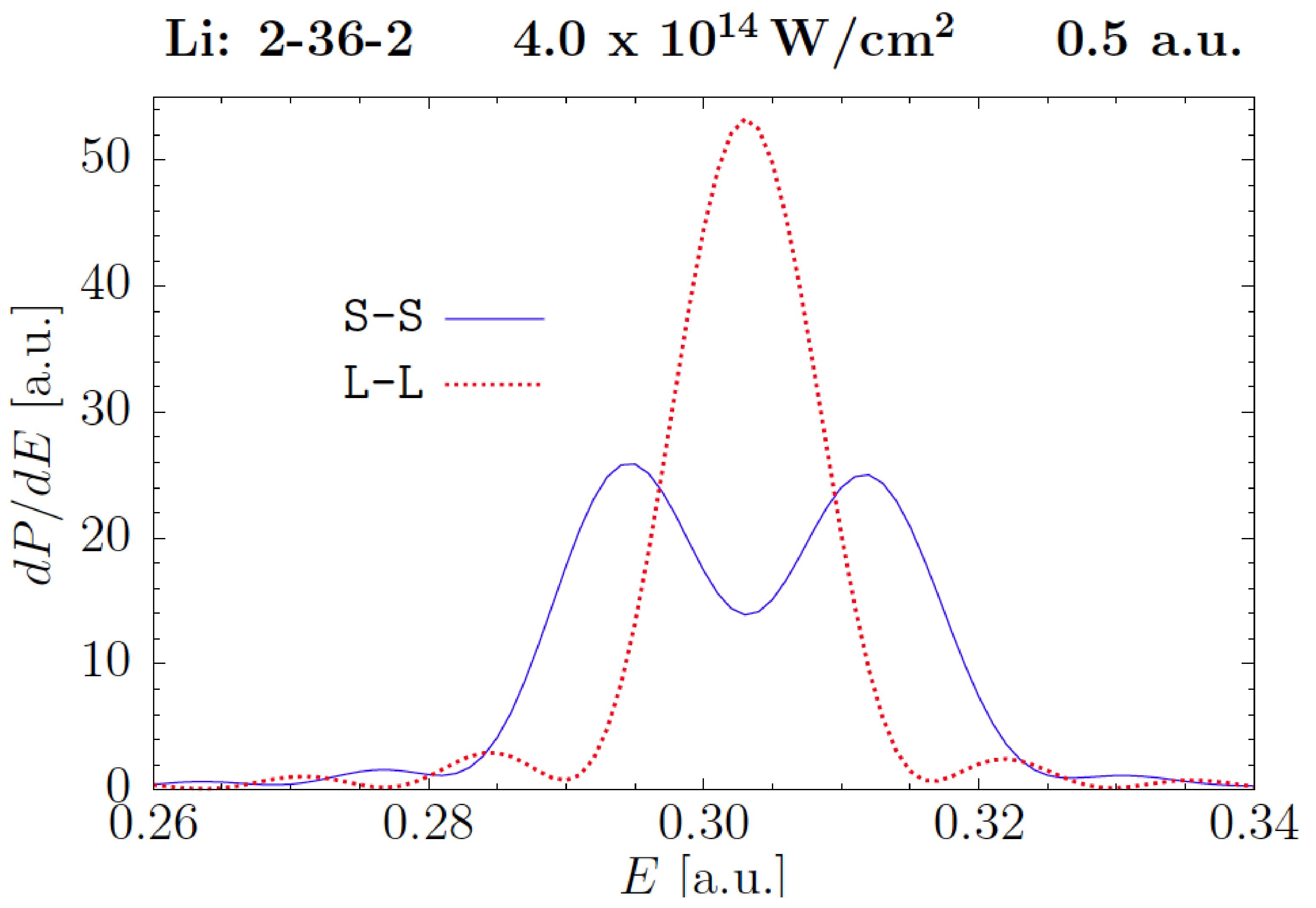
Photoelectron angular distribution



Free-electron displacement



Photoelectron spectrum: Li



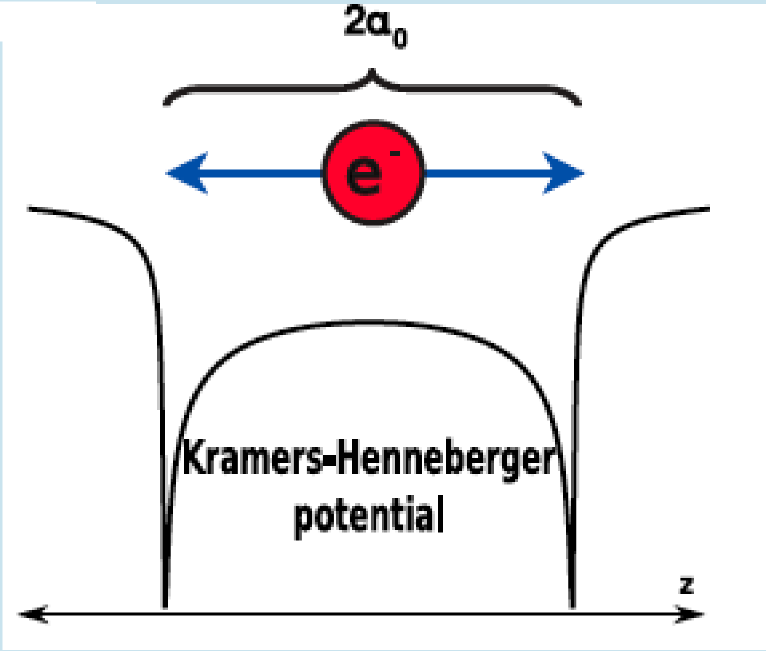
PNAS PNAS PNAS PNAS

Imaging the Kramers–Henneberger atom

Felipe Morales[1], Maria Richter[1], Serguei Patchkovskii[2], and Olga Smirnova[3]

[a]Max-Born Institute for Nonlinear Optics, Max-Born-Strasse 2A, D-12489 Berlin, Germany

Edited* by Paul B. Corkum, University of Ottawa, Ottawa, Canada, and approved July 25, 2011 (received for review April 14, 2011)



Atom reference frame:

$$i\partial\Psi/\partial t = [-\nabla^2/2 + U_{\rm ion}(\mathbf{r}) + \mathbf{E}(t)\mathbf{r}]\Psi$$

$$\mathbf{E}(t) = \mathbf{e}_z E\cos(\omega t)$$

KH atom reference frame:

$$i\partial\Psi_{\rm KH}/\partial t = [-\nabla^2/2 + U_{\rm ion}(\mathbf{r}_{\rm KH} + \alpha(t)\mathbf{e}_z)]\Psi_{\rm KH}$$

$$\alpha(t) = \alpha_0\cos(\omega t) \quad \alpha_0 = E/\omega^2$$

KH potential

$$U_{\rm ion}(\mathbf{r}_{\rm KH} + \alpha(t)\mathbf{e}_z) = V_0(\mathbf{r}_{\rm KH}) + \sum_{n=1}^{\infty} V_n(\mathbf{r}_{\rm KH})\cos(n\omega t)$$

Kramers-Henneberger potential

Time-Dependent Schrödinger equation

$$i\partial\Psi(\boldsymbol{r})/\partial t = \left[\hat{H}_{\text{atom}} + \hat{H}_{\text{int}}(t)\right]\Psi(\boldsymbol{r}),$$

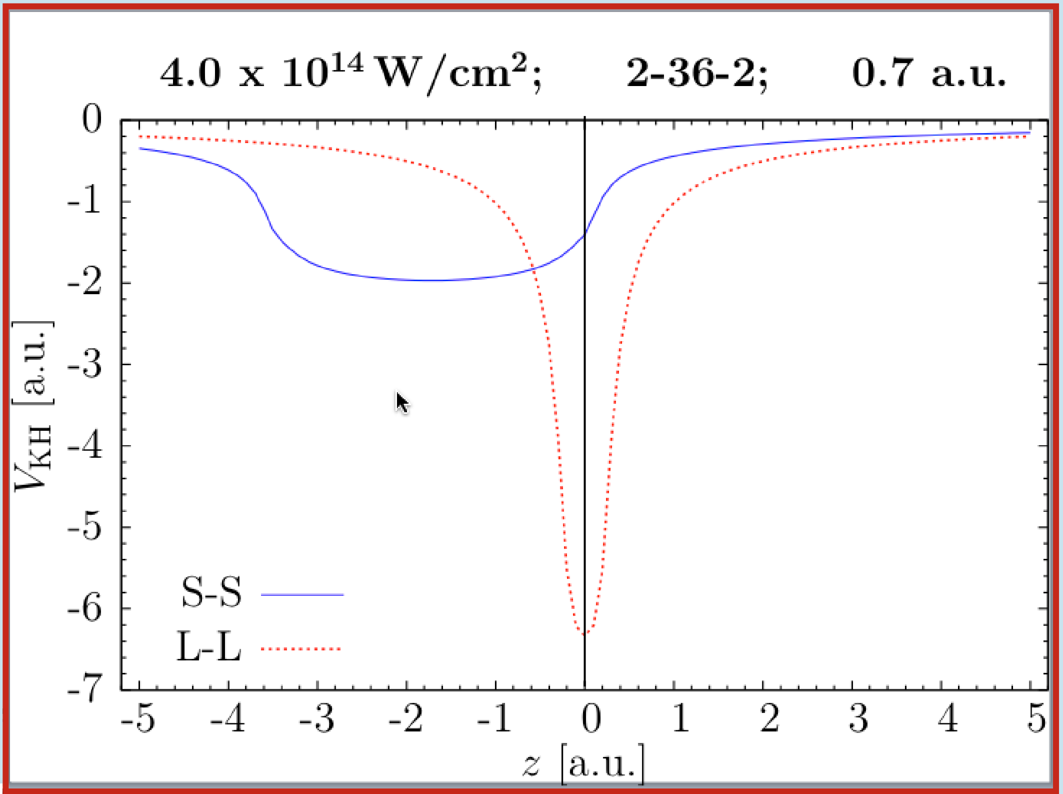
Dipole electromagnetic interaction

$$\hat{H}_{\text{int}}(t) = \begin{cases} \boldsymbol{E}(t)\cdot\hat{\boldsymbol{r}} \\ \boldsymbol{A}(t)\cdot\hat{\boldsymbol{p}} \quad , \quad \boldsymbol{A}(t) = -\int\limits_0^t \boldsymbol{E}(\tau)\,d\tau \end{cases}$$

Kramers-Henneberger gauge

$$\hat{H}_{\text{KH}} = e^{i\hat{T}}\hat{H}_{\text{V}}e^{-i\hat{T}} - \frac{\partial\hat{T}}{\partial t} = \frac{\hat{\boldsymbol{p}}^2}{2} + V(\boldsymbol{r} + \boldsymbol{x}(t))$$

$\hat{T} = \int\limits_0^t \boldsymbol{A}(\tau)\cdot\hat{\boldsymbol{p}}\,d\tau$, where $\boldsymbol{A}(\tau)$ is the vector potential.

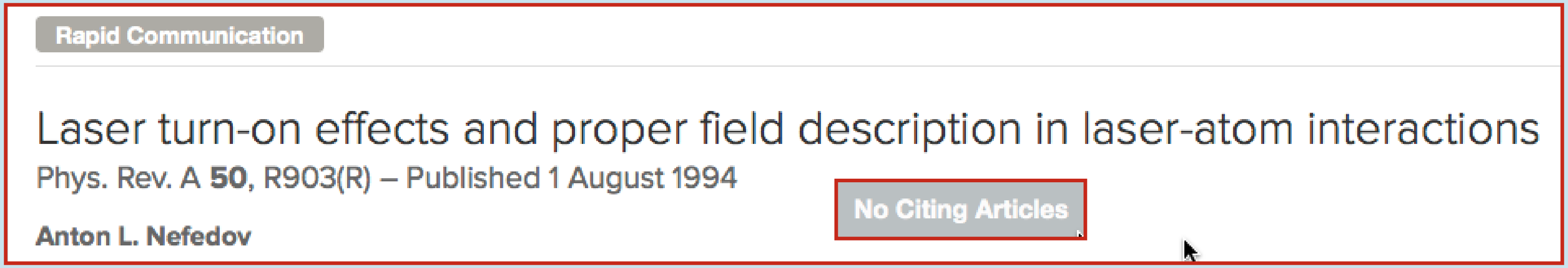


Finite displacement

leads to strongly asymmetric KH potential

$$x = \int\limits_0^T \boldsymbol{A}(t)\,dt \neq 0$$

Theorists warnings

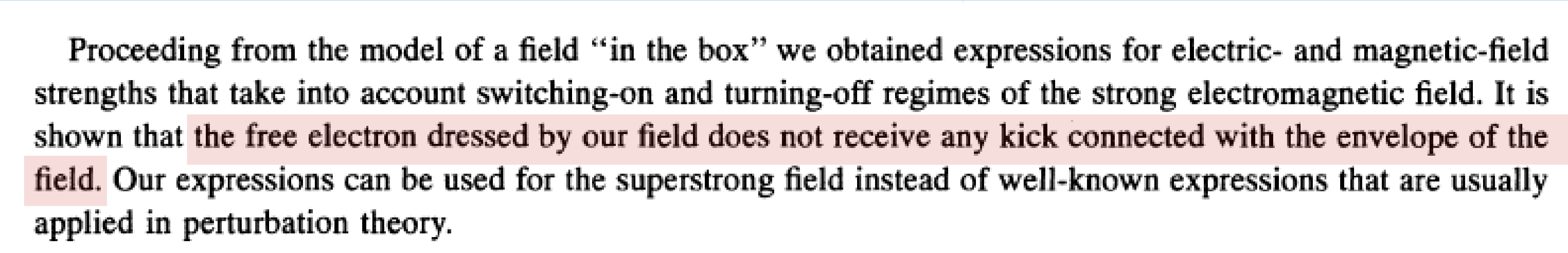


Rapid Communication

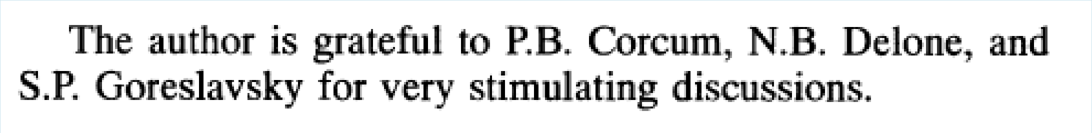
Laser turn-on effects and proper field description in laser-atom interactions

Phys. Rev. A **50**, R903(R) – Published 1 August 1994

Anton L. Nefedov

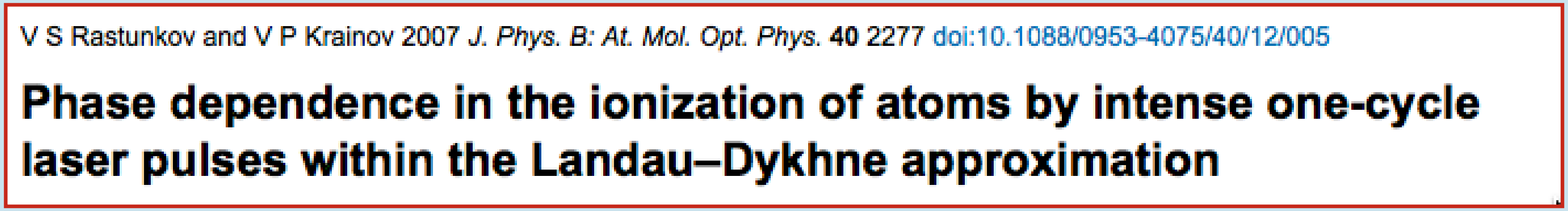


Proceeding from the model of a field “in the box” we obtained expressions for electric- and magnetic-field strengths that take into account switching-on and turning-off regimes of the strong electromagnetic field. It is shown that the free electron dressed by our field does not receive any kick connected with the envelope of the field. Our expressions can be used for the superstrong field instead of well-known expressions that are usually applied in perturbation theory.



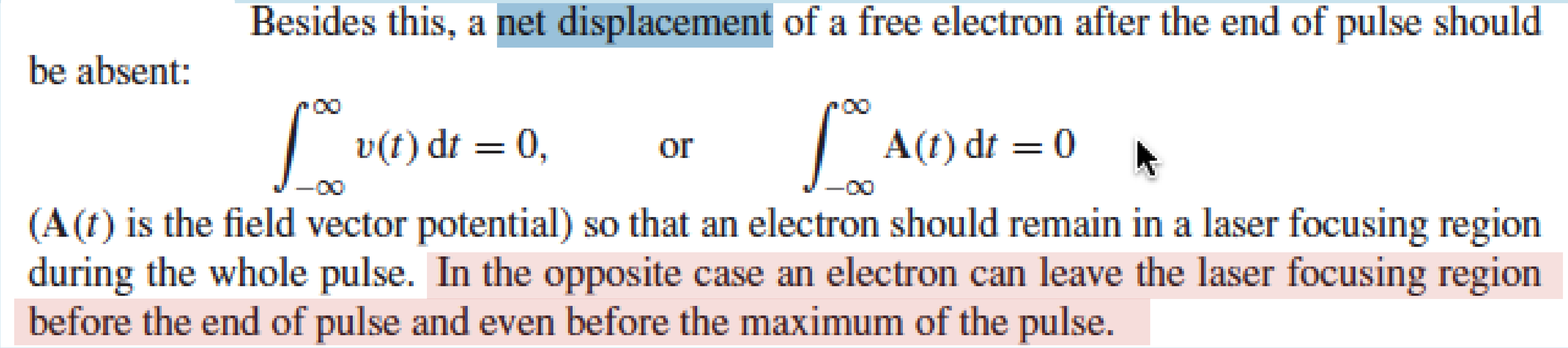
The author is grateful to P.B. Corcum, N.B. Delone, and S.P. Goreslavsky for very stimulating discussions.

Theorists warnings



V S Rastunkov and V P Krainov 2007 *J. Phys. B: At. Mol. Opt. Phys.* **40** 2277 doi:10.1088/0953-4075/40/12/005

Phase dependence in the ionization of atoms by intense one-cycle laser pulses within the Landau–Dykhne approximation



Besides this, a net displacement of a free electron after the end of pulse should be absent:

$$\int_{-\infty}^{\infty} v(t)\,\mathrm{d}t = 0, \qquad \text{or} \qquad \int_{-\infty}^{\infty} \mathbf{A}(t)\,\mathrm{d}t = 0$$

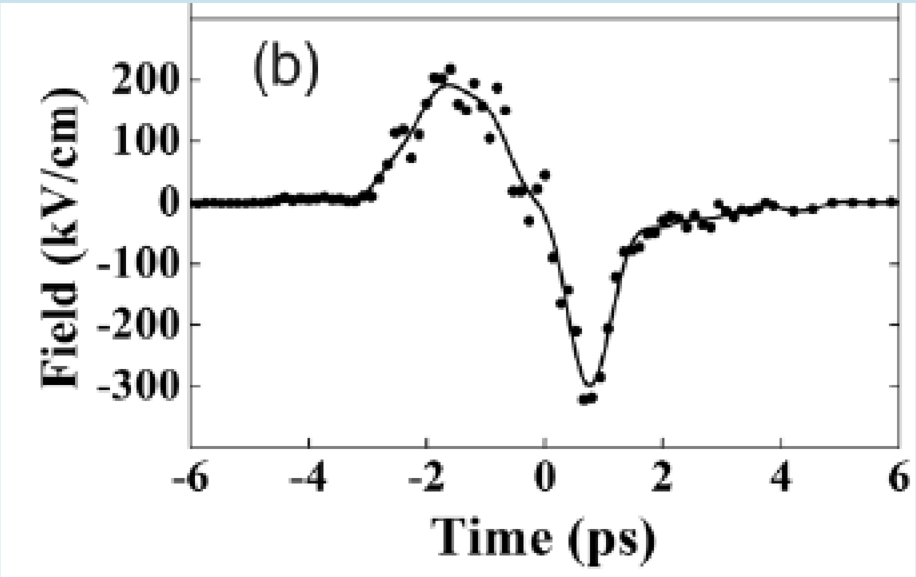
($\mathbf{A}(t)$ is the field vector potential) so that an electron should remain in a laser focusing region during the whole pulse. In the opposite case an electron can leave the laser focusing region before the end of pulse and even before the maximum of the pulse.

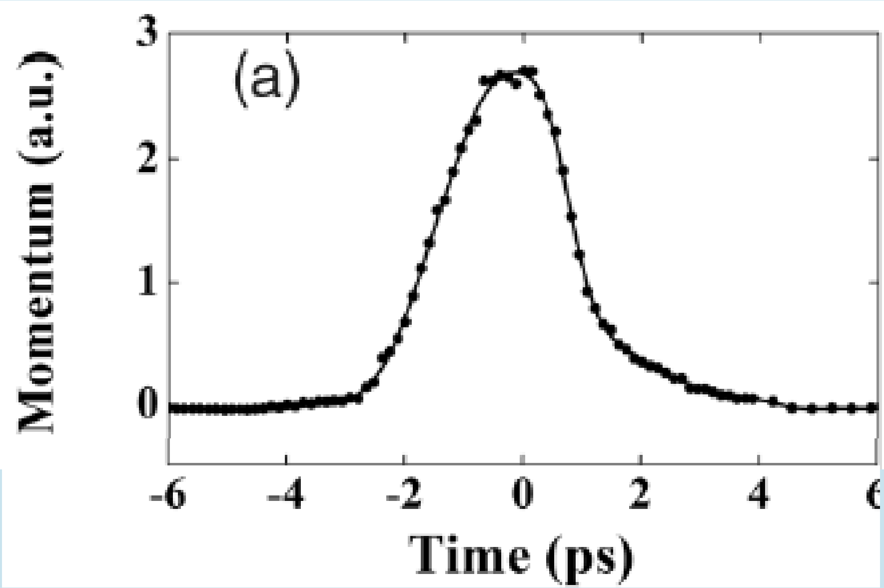
Experimental observations

Ionization of Excited Atoms by Intense Single-Cycle THz Pulses

Phys. Rev. Lett. **112**, 143006 – Published 9 April 2014

Sha Li and R. R. Jones





Displacement is finite:

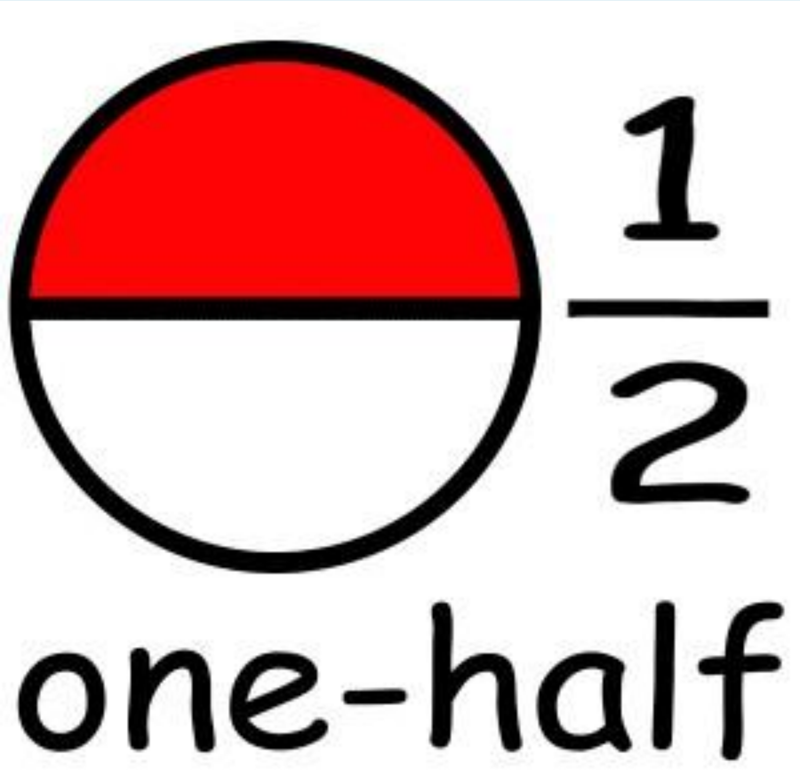
$$x = \int_0^T \boldsymbol{p}(t)\,dt > 0$$

Summary

- Do the pulses with non-zero displacement exist?
  - Maxwell equations do not forbid $\Delta x = \int_0^T \mathbf{A}(t)\, dt \neq 0$
  - Theorists recommend to avoid such pulses:
    - *Kicks and displacements make calculations unstable*
    - *Electron may leave the laser focus*
  - Single-cycle THz pulses have non-zero displacement

- If pulses with non-zero displacement are allowed:
  - Measurement & interpretation require extra care:
    - *Spectral lines*
    - *Angular distributions*

- If zero displacement is enforced:
  - Theorists are restricted in their pulse shapes:
    - *CEP and the envelope function are coupled*
    - *Strong impact on setups for quantum control.*


1
2
one-half

Consistency tests for the attosecond time delay measurements

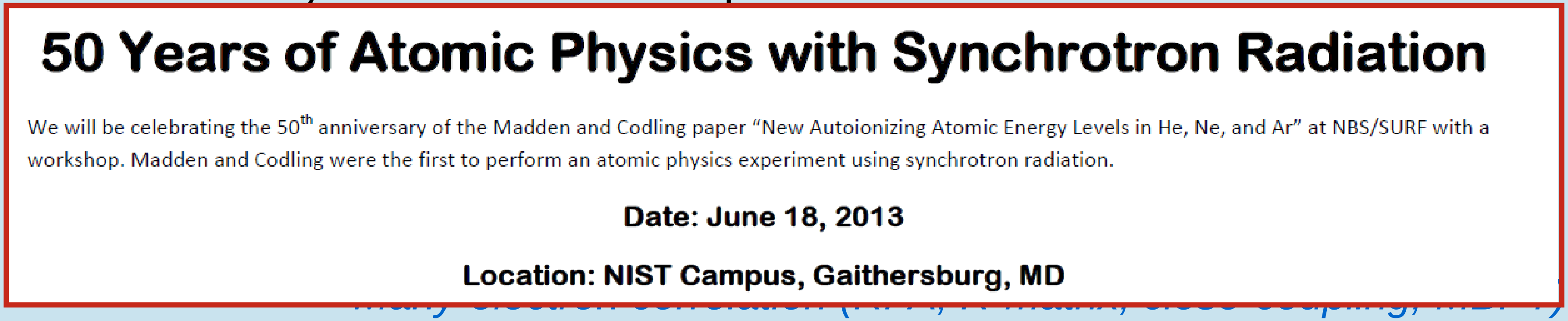
Harsh reality check for attosecond physics

Anatoli Kheifets

THE AUSTRALIAN NATIONAL UNIVERSITY

From conventional to new physics

- *Conventional* atomic physics
    - Synchrotron based experiments



50 Years of Atomic Physics with Synchrotron Radiation

We will be celebrating the 50th anniversary of the Madden and Codling paper "New Autoionizing Atomic Energy Levels in He, Ne, and Ar" at NBS/SURF with a workshop. Madden and Codling were the first to perform an atomic physics experiment using synchrotron radiation.

Date: June 18, 2013

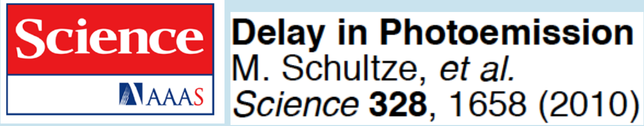
Location: NIST Campus, Gaithersburg, MD

- *New* atomic physics
    - *Intense and ultra-short laser pulses*
        - *Photoelectron group delay* $\tau \sim \partial \arg \Sigma \langle i|D|f_{l\pm1}\rangle / \partial E$
        - *Coulomb-laser coupling corrections*
    - *Theoretical methods*
        - *Time-dependent Schrödinger equation*
        - *Single active electron approximation*

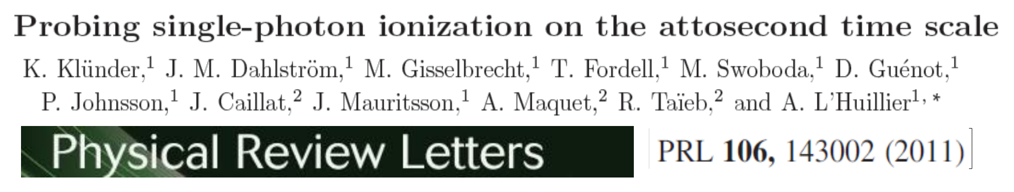
Motivation

- **Measurements of photoemission time delay**

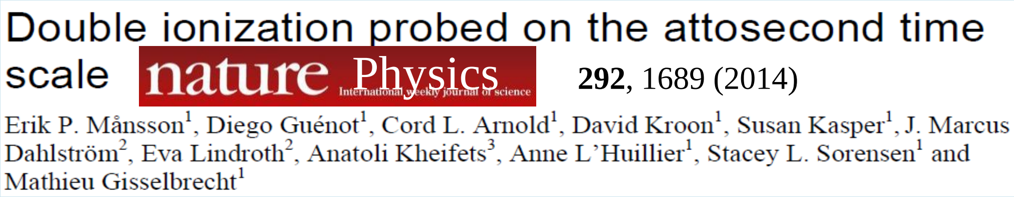
– Attosecond Streaking Ne $2s^{-1}/2p^{-1}$


Science AAAS
Delay in Photoemission
M. Schultze, *et al.*
Science **328**, 1658 (2010)

– RABITT Reconstruction of Attosecond Bursts by Ionization of Two-photon Transitions
  - ✓ Ar $3s^{-1}/3p^{-1}$


Probing single-photon ionization on the attosecond time scale
K. Klünder,[1] J. M. Dahlström,[1] M. Gisselbrecht,[1] T. Fordell,[1] M. Swoboda,[1] D. Guénot,[1] P. Johnsson,[1] J. Caillat,[2] J. Mauritsson,[1] A. Maquet,[2] R. Taïeb,[2] and A. L'Huillier[1,*]
Physical Review Letters PRL **106**, 143002 (2011)

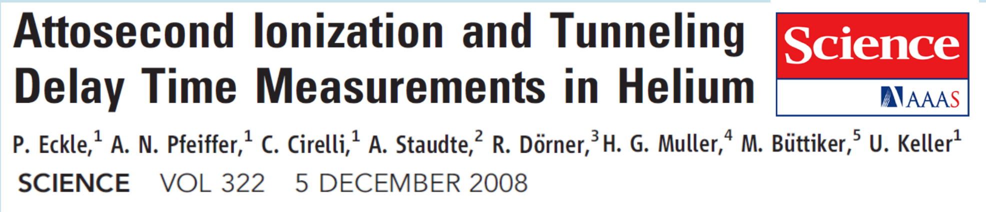
  - ✓ Xe $5p^{-1}/5p^{-2}$


Double ionization probed on the attosecond time scale
nature Physics International weekly journal of science **292**, 1689 (2014)
Erik P. Månsson[1], Diego Guénot[1], Cord L. Arnold[1], David Kroon[1], Susan Kasper[1], J. Marcus Dahlström[2], Eva Lindroth[2], Anatoli Kheifets[3], Anne L'Huillier[1], Stacey L. Sorensen[1] and Mathieu Gisselbrecht[1]

– HHG+RABITT

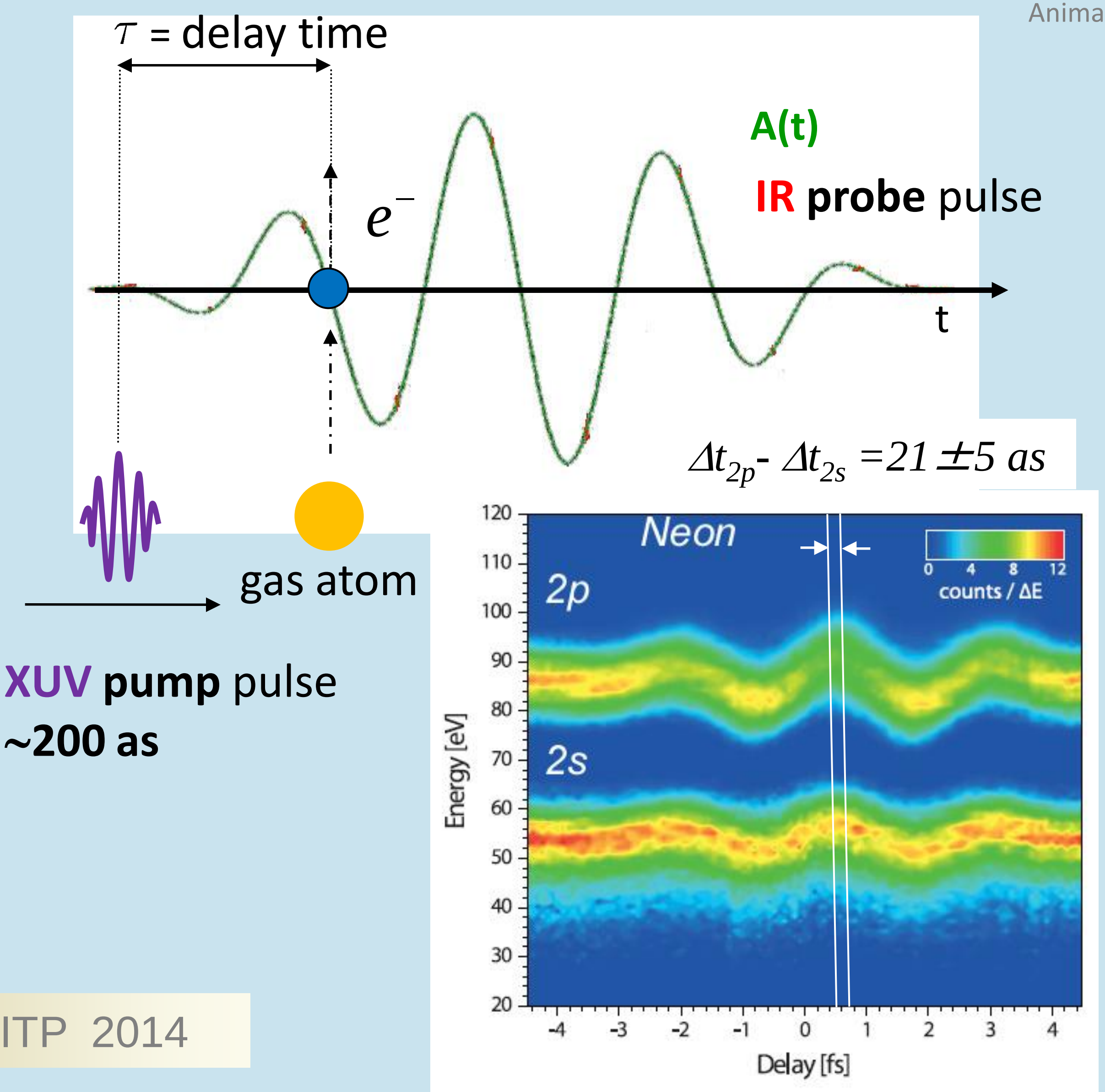

Attosecond pulse shaping around a Cooper minimum
S. B. Schoun,* R. Chirla, J. Wheeler, C. Roedig, P. Agostini, and L. F. DiMauro
Department of Physics, The Ohio State University, Columbus, OH 43210, USA
Physical Review Letters PRL **112**, 153001 (2014)

– ATTO-Clock


Attosecond Ionization and Tunneling Delay Time Measurements in Helium
Science AAAS
P. Eckle,[1] A. N. Pfeiffer,[1] C. Cirelli,[1] A. Staudte,[2] R. Dörner,[3] H. G. Muller,[4] M. Büttiker,[5] U. Keller[1]
SCIENCE VOL 322 5 DECEMBER 2008

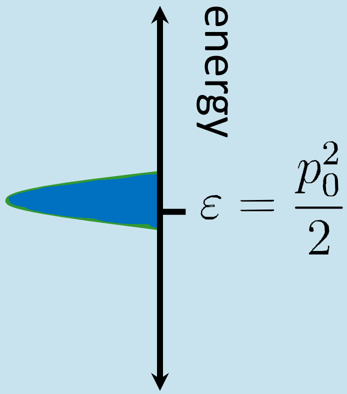
Attosecond Streak Camera

Animation: Renate Pazourek



$\Delta t_{2p} - \Delta t_{2s} = 21 \pm 5$ as

XUV pump pulse
~200 as



$$\varepsilon = \frac{p_0^2}{2}$$

$$\vec{p}(\tau) \approx \vec{p}_0 - \vec{A}(\tau)$$

Mapping of (release) time to momentum

M. Schultze *et al.*, *Science* **328**, 1659 (2010)

Multiphoton regime / linear light

- **Attosecond Streaking**
  - Relative time delay in Ne $\Delta\tau$ $2s^{-1}/2p^{-1}$

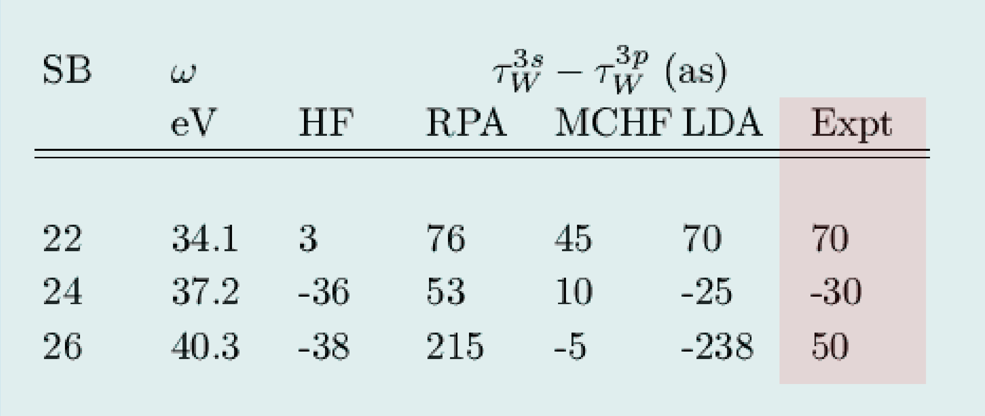
Schultze *et al,* Science **328**, 1658 (2010)	21±5 as	Experiment
Kheifets & Ivanov, PRL **105**, 233002 (2010)	8.4 as	Theory, TDSE
Moore *et al*, PRA **84**, 061404 (2011)	10.2±1.3 as	Theory, R-matrix
Dahlström *et al,* PRA **86**, 061402(R) (2012)	12 as	Theory, MBPT
Kheifets, PRA **87**, 063404 (2013)	11.9 as	Theory, RPA
Feist *et al,* PRA **89**, 033417 (2014)	10.0 as	Theory, B-splines

Multiphoton regime / linear light

- RABITT
  - Relative time delay in Ar $\Delta\tau$ $3s^{-1}/3p^{-1}$

Multiphoton regime / linear light

- RABITT
  - Relative time delay in Ar $\Delta\tau$ $3s^{-1}/3p^{-1}$

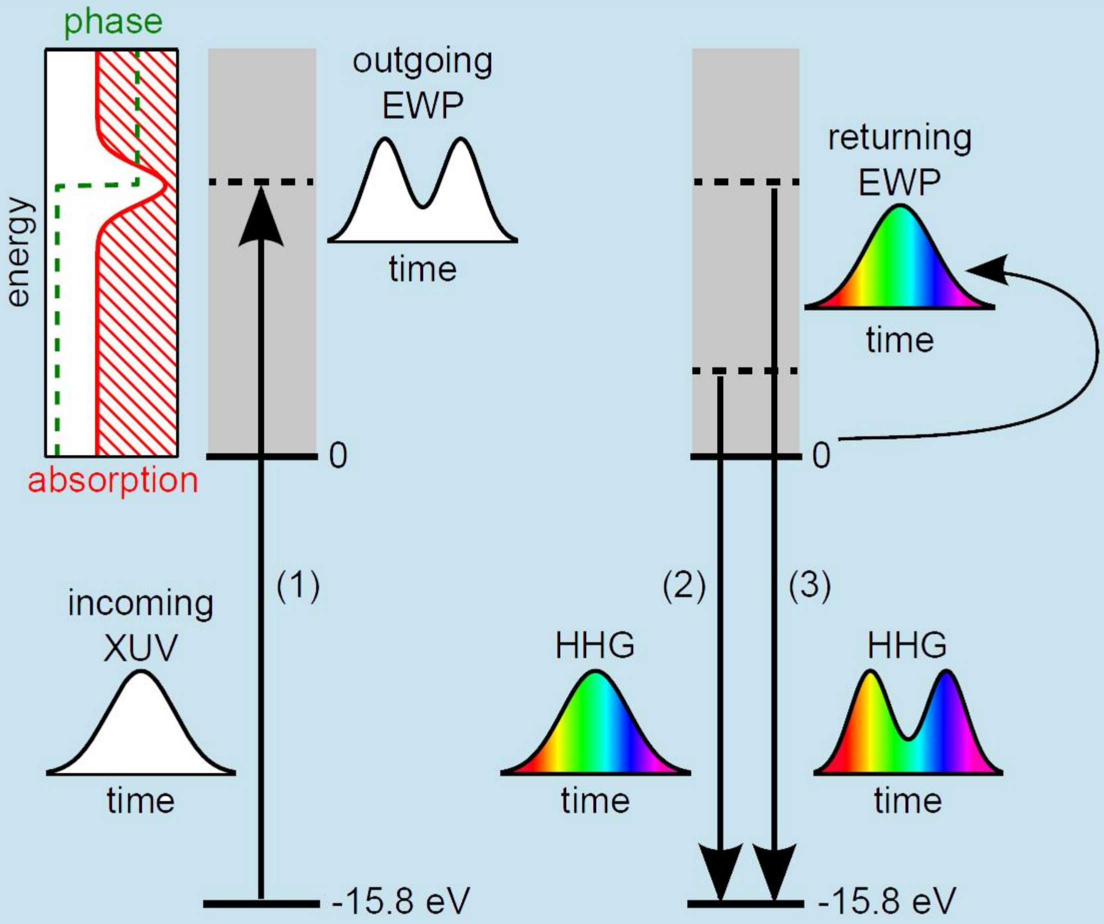


SB	ω	$\tau_W^{3s} - \tau_W^{3p}$ (as)				
	eV	HF	RPA	MCHF	LDA	Expt
22	34.1	3	76	45	70	70
24	37.2	-36	53	10	-25	-30
26	40.3	-38	215	-5	-238	50

Guénot *et al,* PRA 85, 053424 (2012) Experiment
Carette *et al,* PRA **87**, 023420 (2013) Theory, MCHF
Kheifets, PRA **87**, 063404 (2013) Theory, HF,RPA
Dixit et al, PRL **113**,203003 (2013) Theory, LDA

Multiphoton regime / linear light

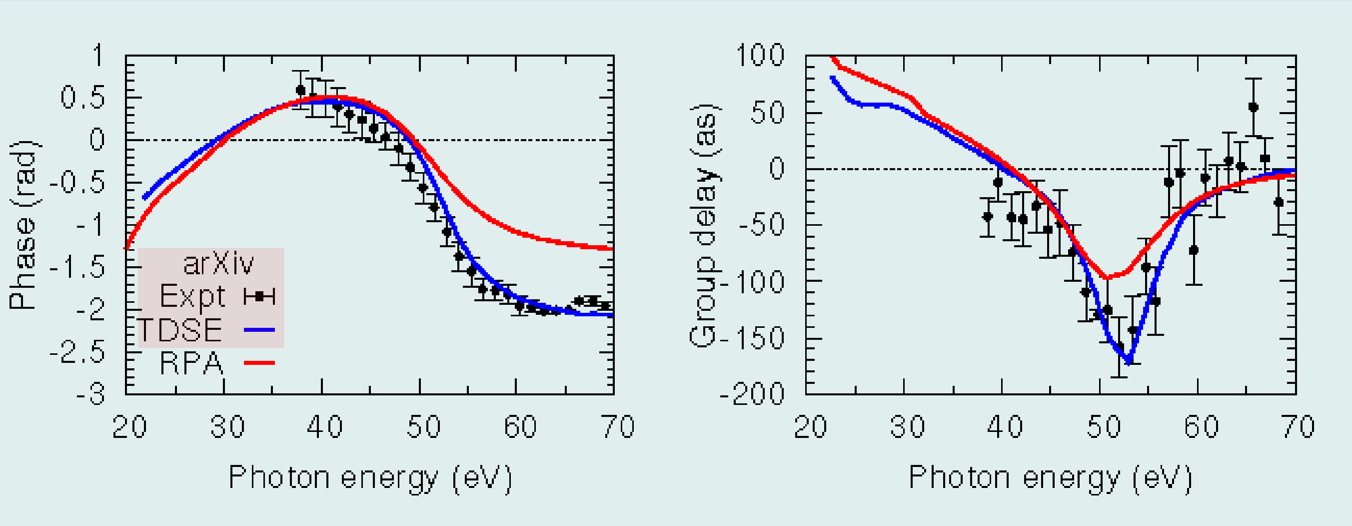
- **HHG + RABITT**
  - Phase variation and group delay near Cooper minimum in Ar ←→ $3p^{-1}$



Schoun *et al*, PRL **112**, 153001 (2014)

Multiphoton regime / linear light

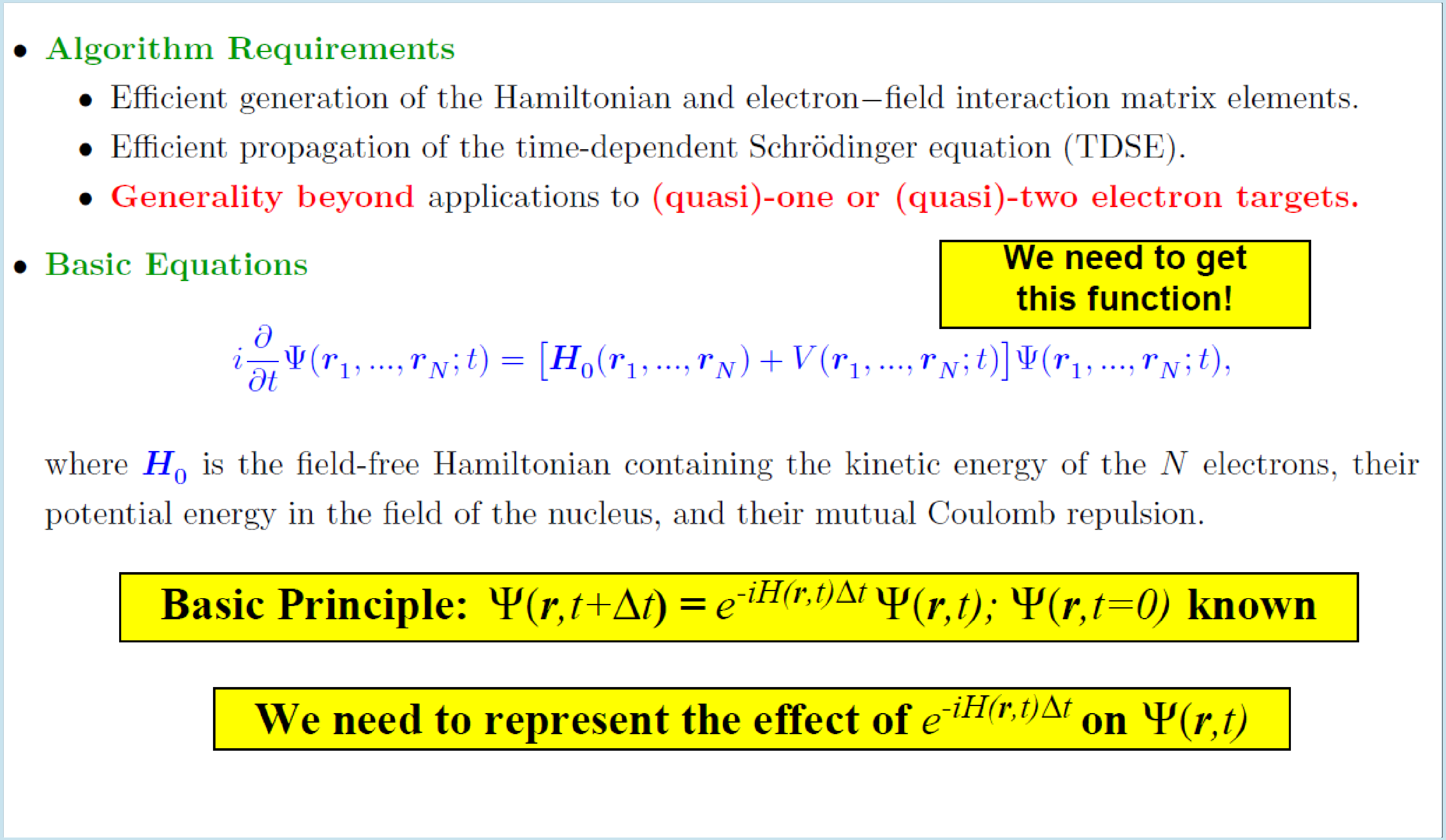
- HHG + RABITT
  - Phase variation and group delay near Cooper minimum in Ar $3p^{-1}$



Schoun *et al,* PRL **112**, 153001 (2014)	2.6±0.3 rad	Experiment, TDSE
Kheifets, PRA **87**, 063404 (2013)	1.9 rad	Theory, HF,RPA

Time-Dependent Theory

Solving Time-Dependent Schrödinger Equation



- **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!

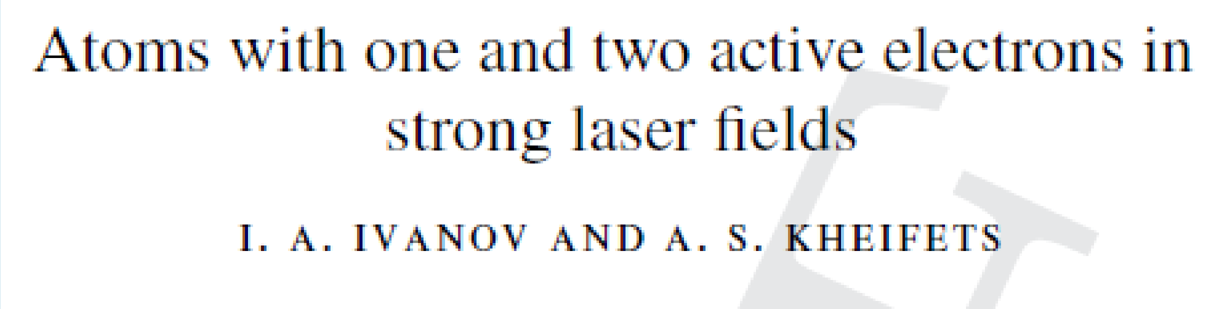
$$i\frac{\partial}{\partial t}\Psi(\boldsymbol{r}_1,...,\boldsymbol{r}_N;t) = \left[\boldsymbol{H}_0(\boldsymbol{r}_1,...,\boldsymbol{r}_N) + V(\boldsymbol{r}_1,...,\boldsymbol{r}_N;t)\right]\Psi(\boldsymbol{r}_1,...,\boldsymbol{r}_N;t),$$

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.

Basic Principle: $\Psi(\boldsymbol{r},t+\Delta t) = e^{-iH(\boldsymbol{r},t)\Delta t}\,\Psi(\boldsymbol{r},t);\ \Psi(\boldsymbol{r},t=0)$ **known**

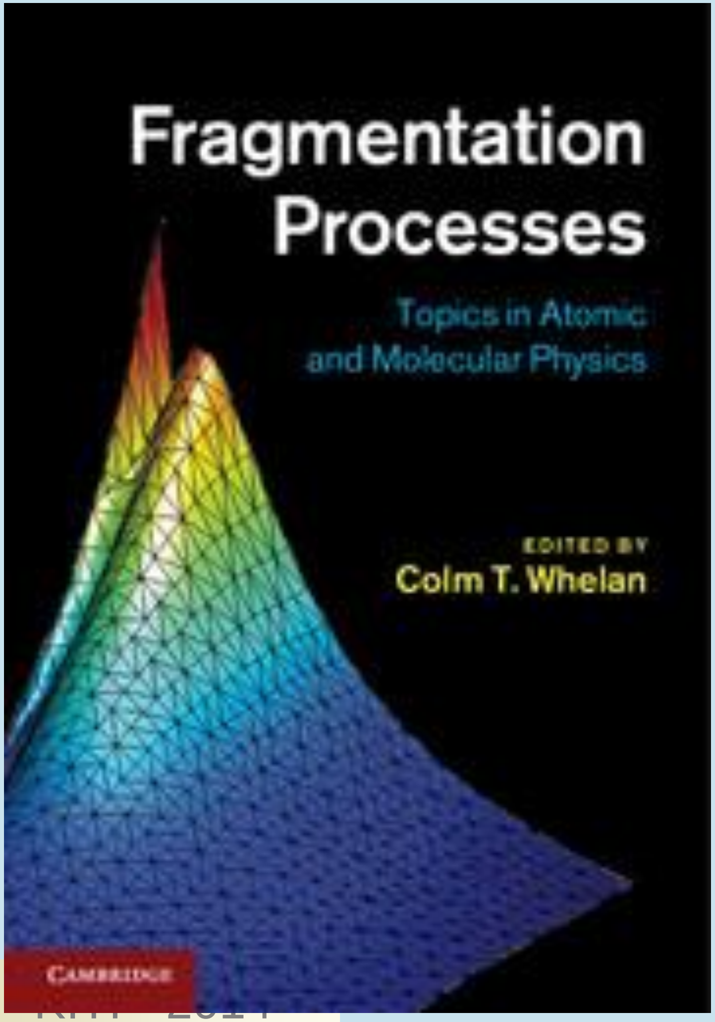
We need to represent the effect of $e^{-iH(\boldsymbol{r},t)\Delta t}$ **on** $\Psi(\boldsymbol{r},t)$

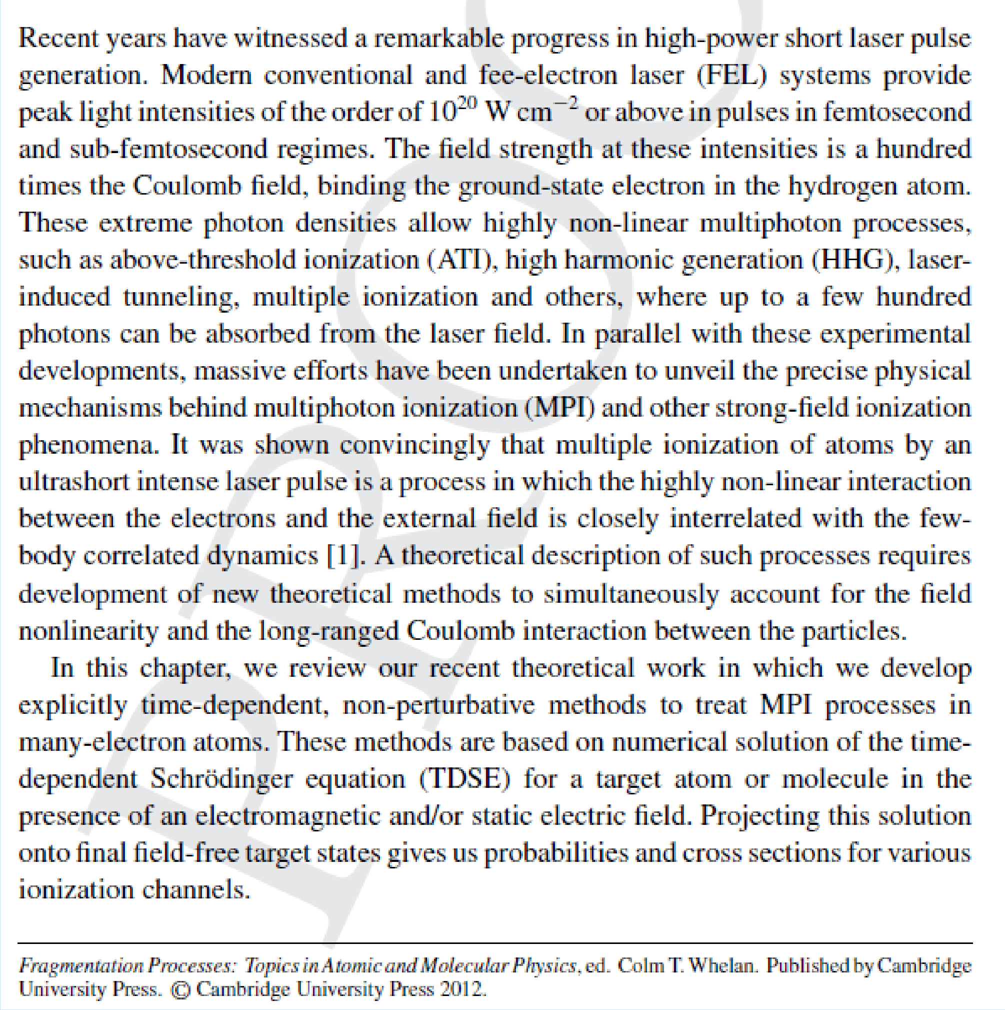
From Klaus Bartschat Wednesday talk



Atoms with one and two active electrons in strong laser fields

I. A. IVANOV AND A. S. KHEIFETS





Recent years have witnessed a remarkable progress in high-power short laser pulse generation. Modern conventional and fee-electron laser (FEL) systems provide peak light intensities of the order of 10^{20} W cm^{-2} or above in pulses in femtosecond and sub-femtosecond regimes. The field strength at these intensities is a hundred times the Coulomb field, binding the ground-state electron in the hydrogen atom. These extreme photon densities allow highly non-linear multiphoton processes, such as above-threshold ionization (ATI), high harmonic generation (HHG), laser-induced tunneling, multiple ionization and others, where up to a few hundred photons can be absorbed from the laser field. In parallel with these experimental developments, massive efforts have been undertaken to unveil the precise physical mechanisms behind multiphoton ionization (MPI) and other strong-field ionization phenomena. It was shown convincingly that multiple ionization of atoms by an ultrashort intense laser pulse is a process in which the highly non-linear interaction between the electrons and the external field is closely interrelated with the few-body correlated dynamics [1]. A theoretical description of such processes requires development of new theoretical methods to simultaneously account for the field nonlinearity and the long-ranged Coulomb interaction between the particles.

In this chapter, we review our recent theoretical work in which we develop explicitly time-dependent, non-perturbative methods to treat MPI processes in many-electron atoms. These methods are based on numerical solution of the time-dependent Schrödinger equation (TDSE) for a target atom or molecule in the presence of an electromagnetic and/or static electric field. Projecting this solution onto final field-free target states gives us probabilities and cross sections for various ionization channels.

Fragmentation Processes: Topics in Atomic and Molecular Physics, ed. Colm T. Whelan. Published by Cambridge University Press.

National Computational Infrastructure

National Computational Infrastructure

NCI National Facility

nf.nci.org.au

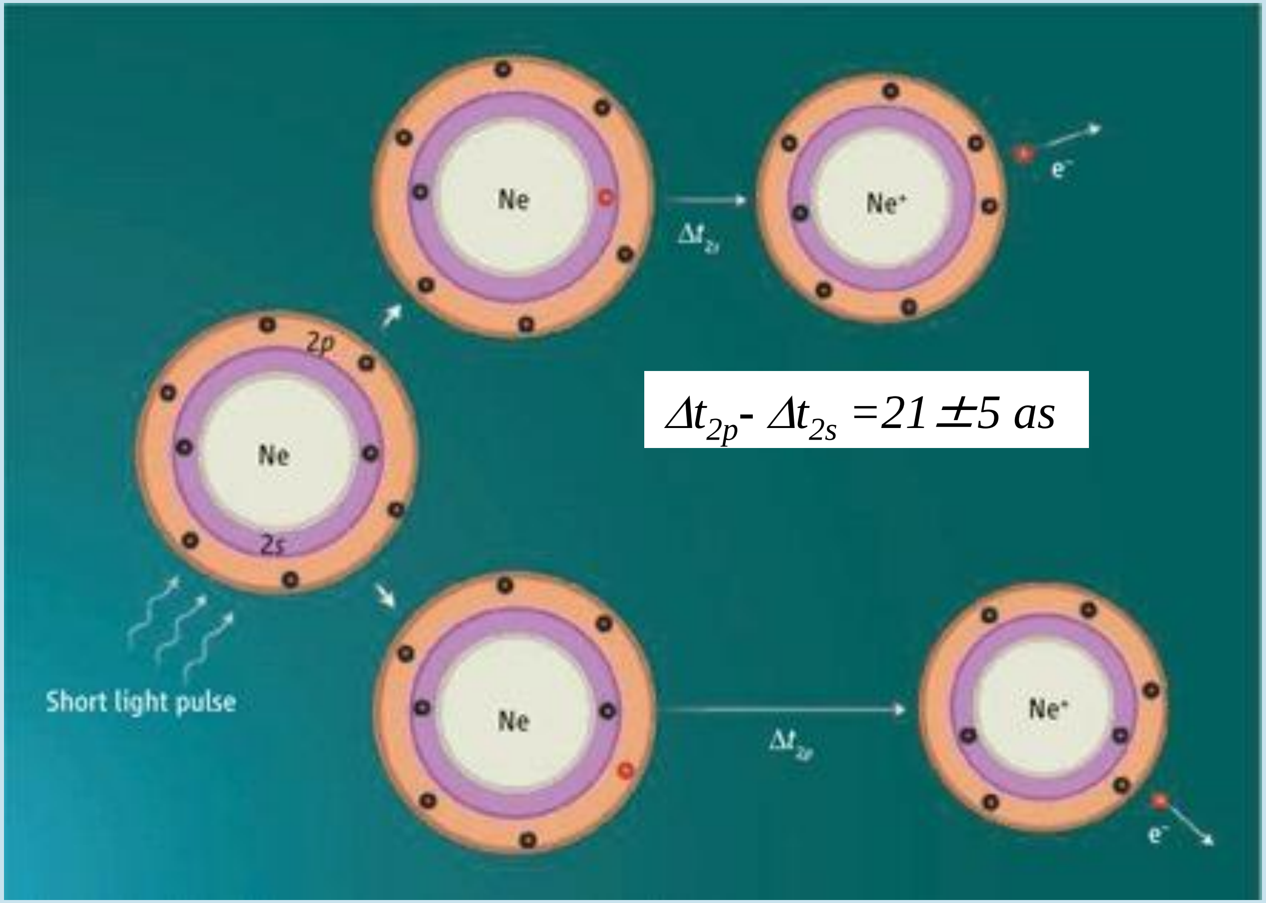
Raijin – Thunder God in Japanese, launched officially on July 31, 2013

Peak performance of 1.2 PetaFlops – 1,200,000,000,000,000 floating point operations per second

Number 24 on the World Top 500 Supercomputer ranking

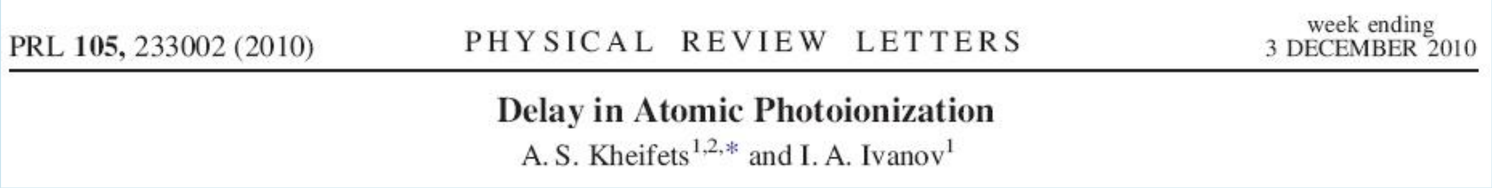
Experiment

Delay in Photoemission
M. Schultze, *et al.*
Science **328**, 1658 (2010)



$\Delta t_{2p} - \Delta t_{2s} = 21 \pm 5$ as

Theoretical interpretation: solution of TDSE

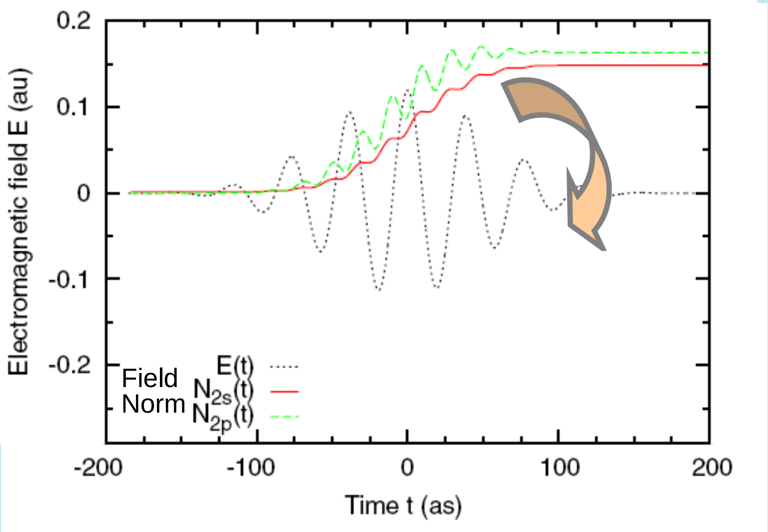


PRL **105**, 233002 (2010) PHYSICAL REVIEW LETTERS week ending 3 DECEMBER 2010

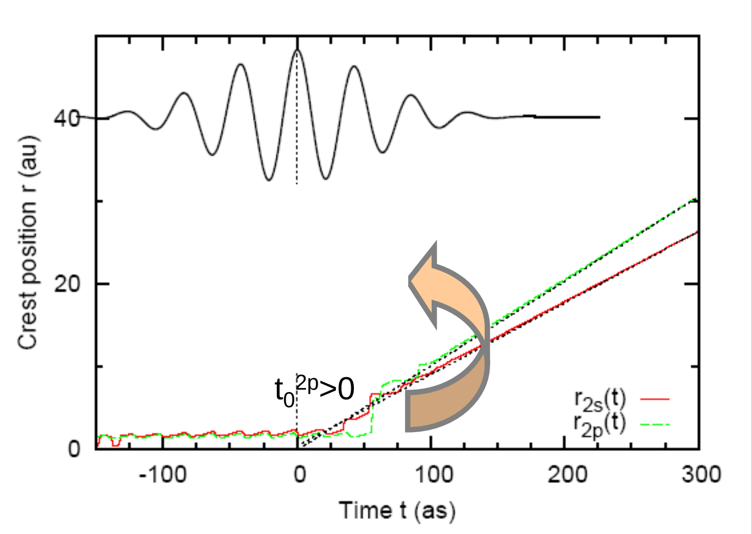
Delay in Atomic Photoionization

A. S. Kheifets[1,2,*] and I. A. Ivanov[1]

Wavepacket norm



Crest position



–*Photoelectrons are set in motion with the end of the pulse*

–*Delay is negative for 2s and positive for 2p*

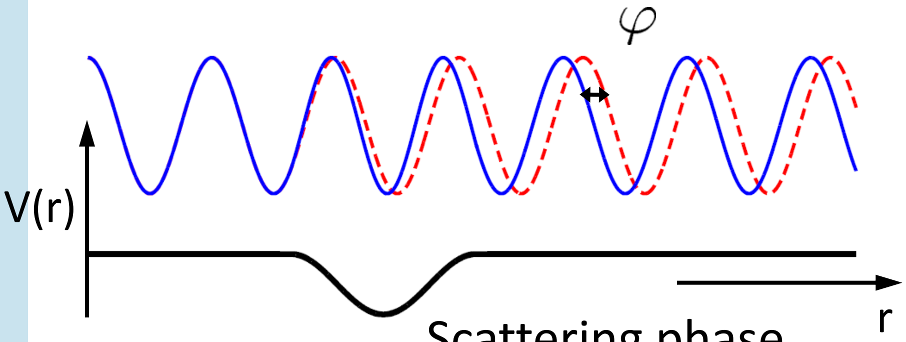
–*Relative time delay is only ≈ 6 as*

Wigner Time delay

Time delay

in quantum mechanics

$$\psi_{in} = e^{\mathrm{i}kx - \mathrm{i}\omega t} \quad \longrightarrow \quad \psi_{out} = e^{\mathrm{i}kx - \mathrm{i}\omega t + \varphi}$$

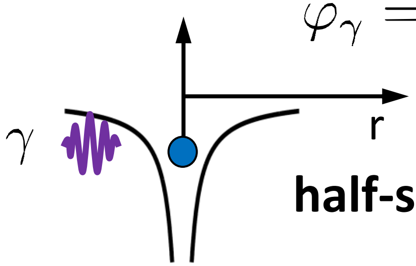


Scattering phase

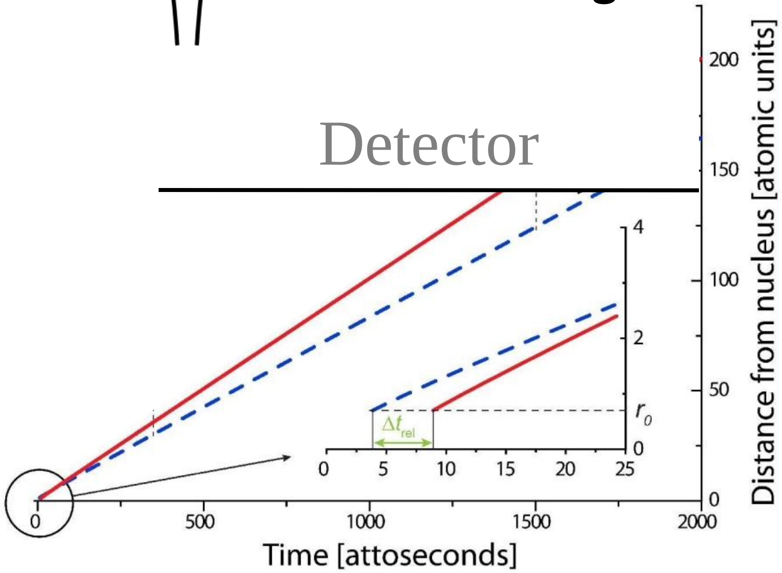
$$\boxed{t_{EWS} = \hbar \frac{d\varphi}{dE}}$$

Wigner time delay

$$\varphi_\gamma = \arg(\langle \vec{p_0} | z | \phi_i \rangle)$$

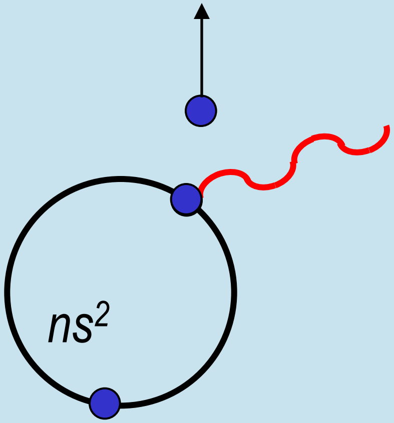


half-scattering



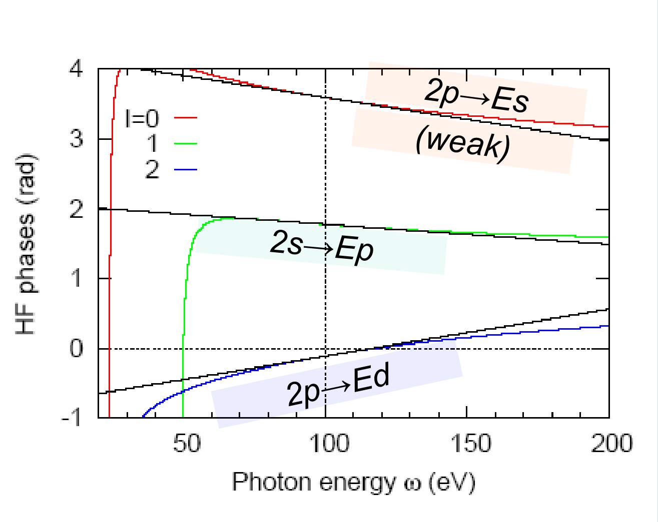
E.P. Wigner. Phys. Rev. 98, 145 (1955)

Quantum-mechanical interpretation



Ne: n=2
Ar: n=3

Coulomb ↓



$$\delta_l(0) - \delta_l(\infty) = n\pi$$

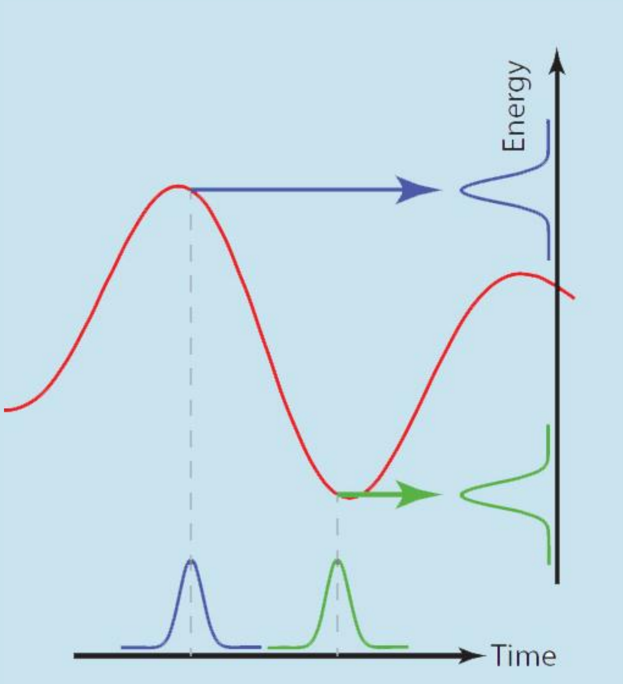
N. Levinson 1949

Levinson →

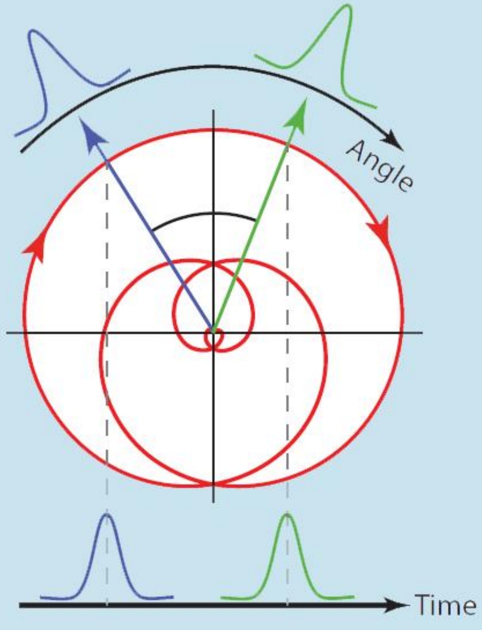
Atto-clock

- Linear streaking vs. angular streaking

Linear streaking



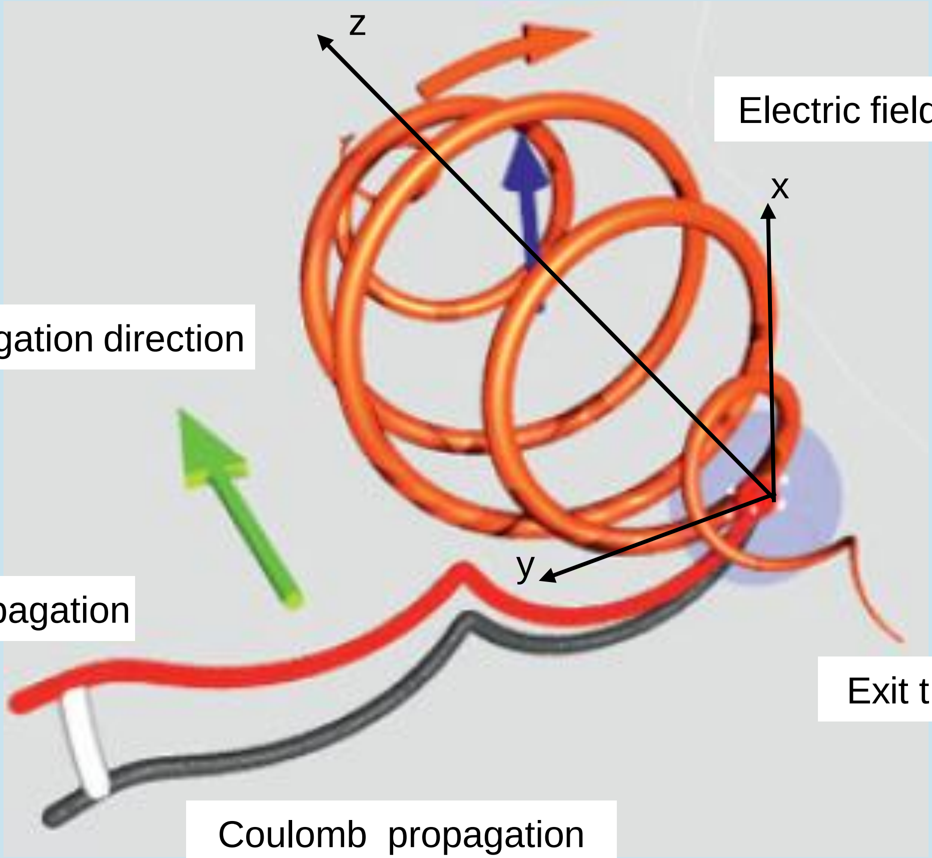
Angular streaking



Annu. Rev. Phys. Chem. 2012. 63:447–69

Atto-clock

- Offset angle θ



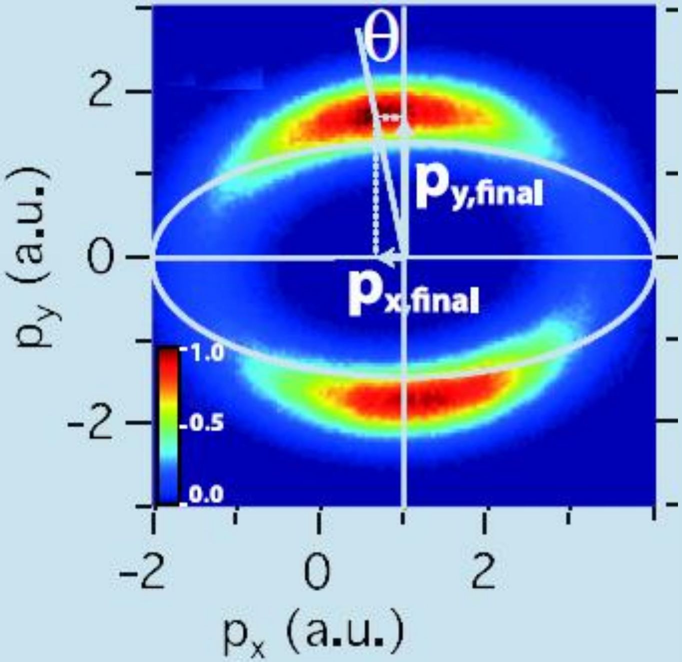
Electric field

Propagation direction

Free propagation

Exit the tunnel

Coulomb propagation



LETTERS Pfeiffer, Cirelli, Smolarski et al.
PUBLISHED ONLINE: 23 OCTOBER 2011 | DOI: 10.1038/NPHYS2125
nature physics

Atto-clock measurement in He

- *Angular off-set*

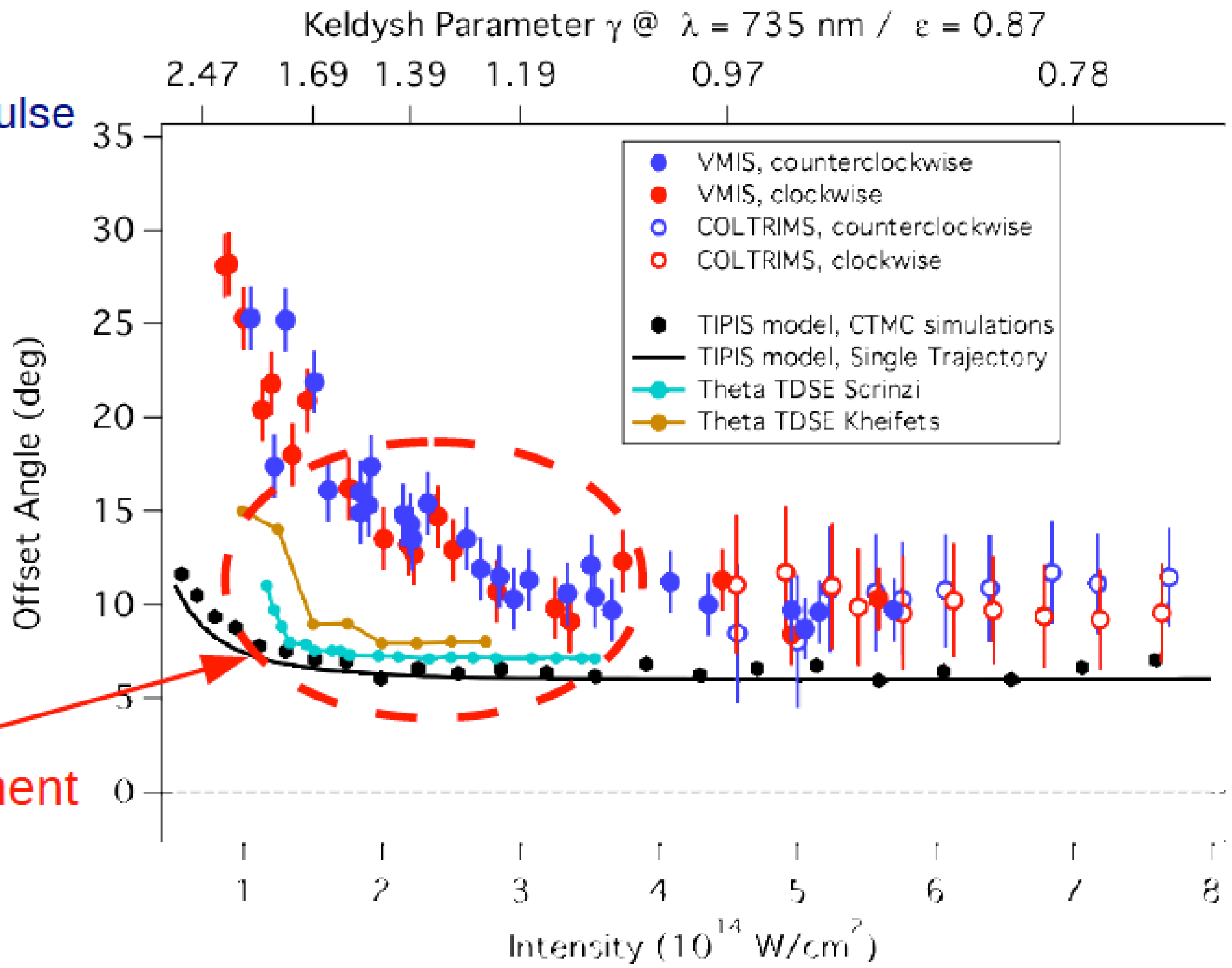
Helium, elliptically polarized pulse

Measurements:

C. Cirelli et al. ETH

Calculations:

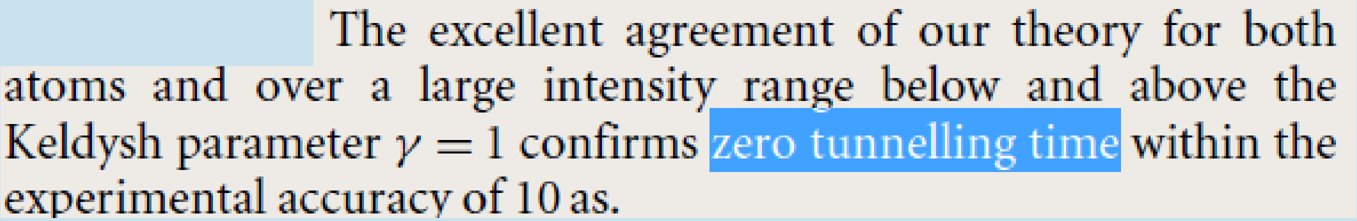
L. Madsen(TIPIS)
Kheifets/Ivanov
A. Zielinski/A.S.



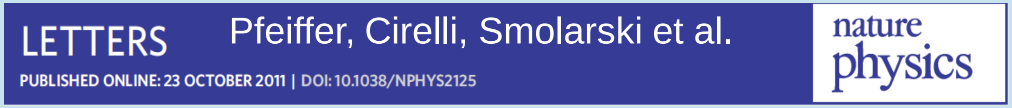
Very disquieting disagreement

ATTO-clock

- Tunneling time implications
  - Zero tunneling time with COLTRIMS $\gamma>1$, $I>3\times10^{14}$ W/cm^2
  - Finite tunneling time with VM



The excellent agreement of our theory for both atoms and over a large intensity range below and above the Keldysh parameter $\gamma = 1$ confirms zero tunnelling time within the experimental accuracy of 10 as.



LETTERS Pfeiffer, Cirelli, Smolarski et al.

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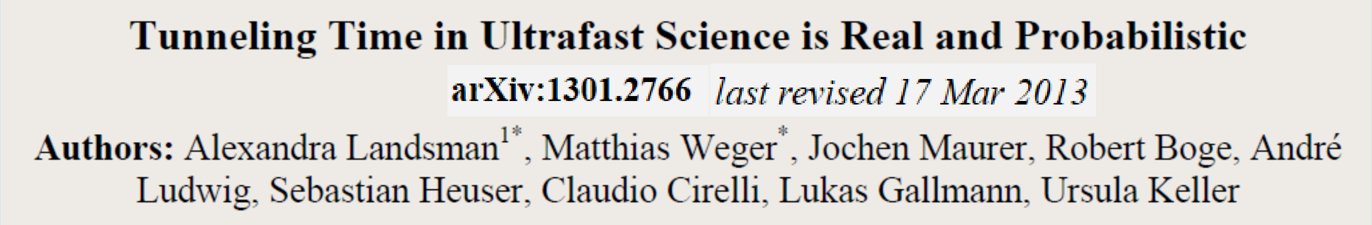


Tunneling Time in Ultrafast Science is Real and Probabilistic

arXiv:1301.2766 *last revised 17 Mar 2013*

Authors: Alexandra Landsman[1*], Matthias Weger[*], Jochen Maurer, Robert Boge, André Ludwig, Sebastian Heuser, Claudio Cirelli, Lukas Gallmann, Ursula Keller

Physics > Atomic Pl **arXiv:1402.5620**

nature Photonics

Interpreting Attoclock Measurements of Tunnelling Times

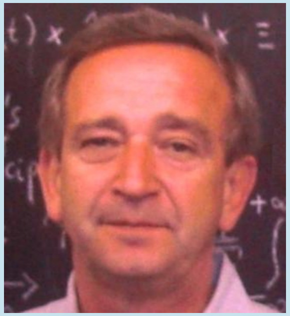
Lisa Torlina, Felipe Morales, Jivesh Kaushal, Harm Geert Muller, Igor Ivanov, Anatoli Kheifets, Alejandro Zielinski, Armin Scrinzi, Misha Ivanov, Olga Smirnova

Conclusion

- Probing single-photon ionization on the attosecond time scale
  - Experiment
    - *Attosecond streaking*
    - *RABBITT*
    - *HHG*
    - *ATTO-clock*
  - *TDSE calculations*
    - *Qualitatively similar*
    - *Numerically can be very different*
  - *Time delay*
    - *Multi-photon regime: understood*
    - *Tunneling regime: still an open question*
- Outlook:
    - *New attosecond devices: Larmor clock*

In collaboration with — theory:

Igor Ivanov, ANU



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Griffith University

Thomas Pfeifer, Robert Moshammer

MPI Heidelerg

Louis DiMauro, Pierre Agostini

Ohio State University, Columbus


Thank you