Strong-Field Ionization of Hydrogen

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Griffith UNIVERSITY

AASF

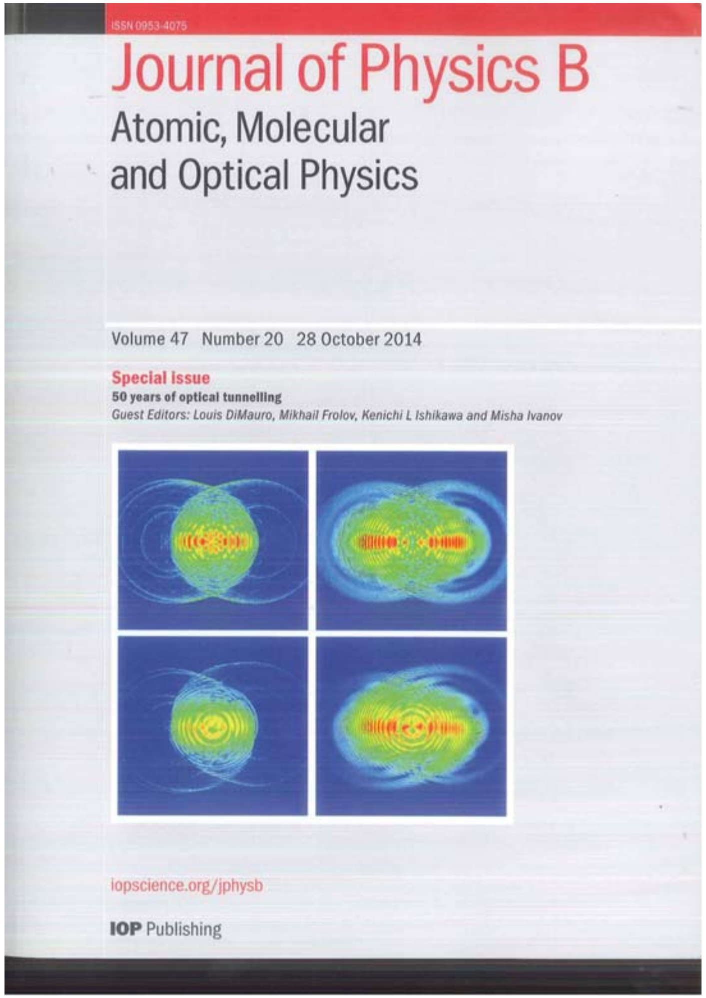
ICPEAC Cairns 26 July 2017

Outline

- Motivation
- Experiments with atomic hydrogen
  - Quantitatively accurate ionization measurements
  - Absolute CEP determination
  - Precise intensity calibration
  - Attosecond streaking and tunneling time
- Experiments with molecular hydrogen
  - Isotope effect in tunneling ionization of H_2/D_2

Motivation: Benchmarking and validating theoretical models

- To achieve quantitative agreement of theory and experiment
- To validate approximate models by comparison with accurate measurements



IOP Publishing Journal of Physics B: Atomic, Molecular and Optical Physics
J. Phys. B: At. Mol. Opt. Phys. **47** (2014) 204003 (18pp) doi:10.1088/0953-4075/47/20/204003

Review Article

Benchmarking strong-field ionization with atomic hydrogen

D Kielpinski[1,2], R T Sang[1,2] and I V Litvinyuk[2]

[1] ARC Centre of Excellence for Coherent X-Ray Science, Centre for Quantum Dynamics, Griffith University, Nathan QLD 4111, Australia
[2] Australian Attosecond Science Facility, Centre for Quantum Dynamics, Griffith University, Nathan QLD 4111, Australia

2014 - 50 years since Keldysh's paper

2018 – 25 years since Corkum's paper

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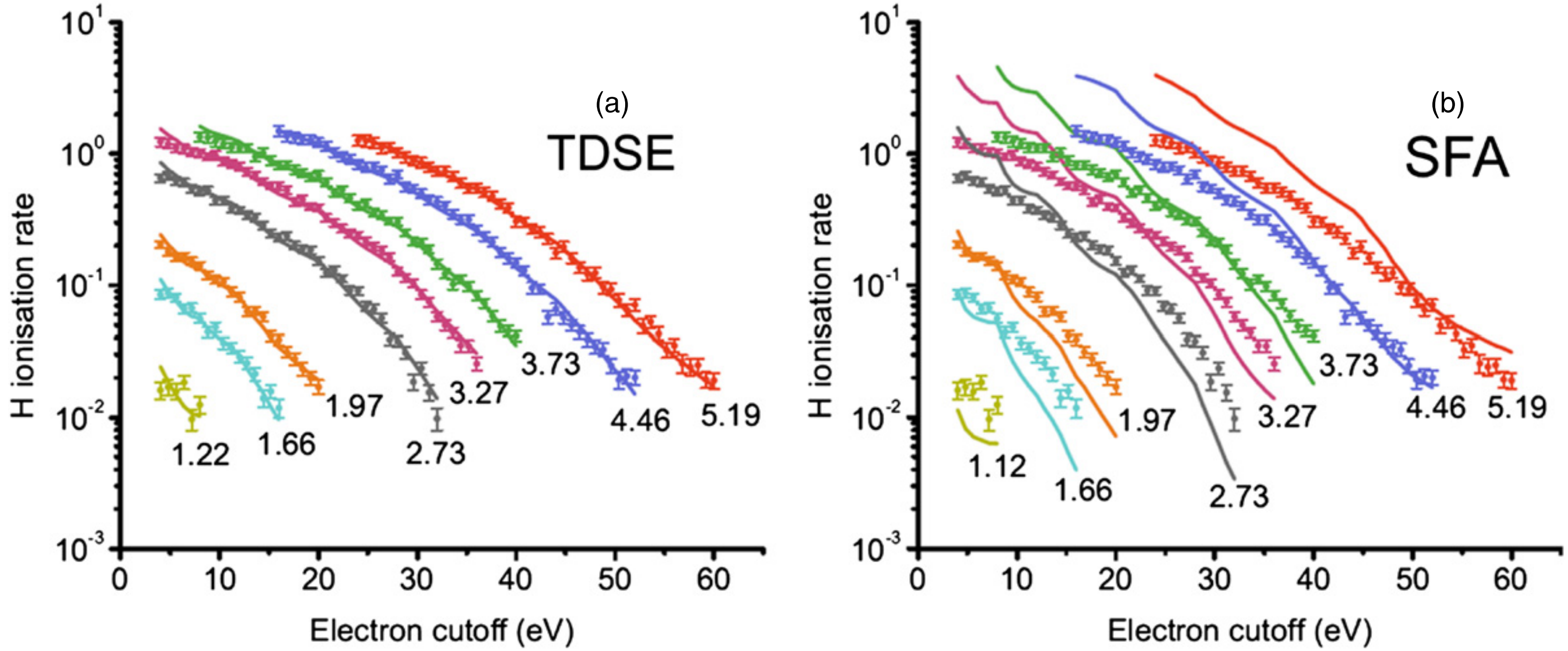
Why H?

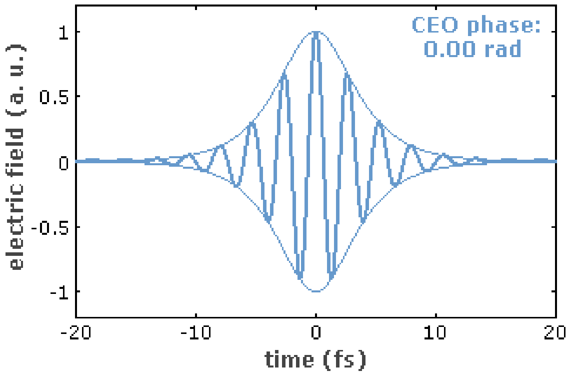
It is the only system where accurate *ab initio* modeling with strong fields is currently possible

Strong-field ionization of atomic hydrogen: results



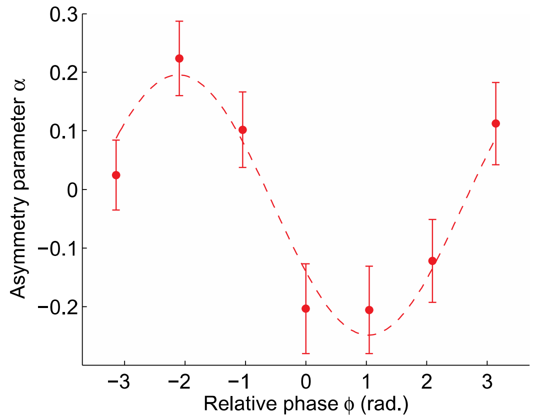
Pullen et al., Opt. Lett. 36, 3660 (2011)

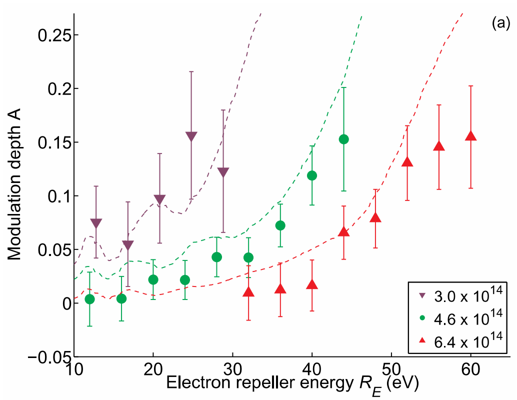
Strong-field ionization of atomic hydrogen: CEP effects

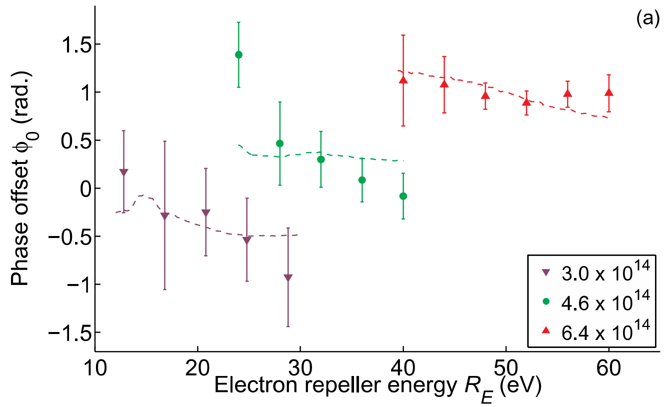


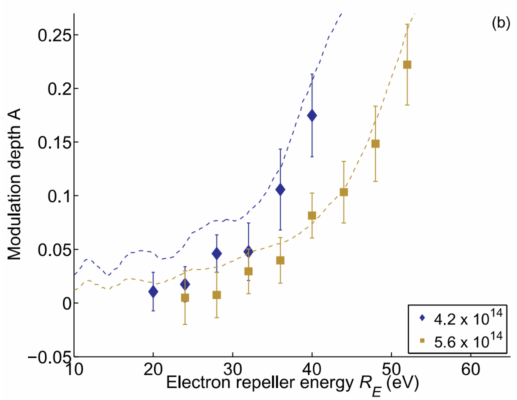
$$A(\phi_{\mathrm{CEP}}) = \frac{Y_T(\phi_{\mathrm{CEP}}) - Y_B(\phi_{\mathrm{CEP}})}{Y_T(\phi_{\mathrm{CEP}}) + Y_B(\phi_{\mathrm{CEP}})}$$

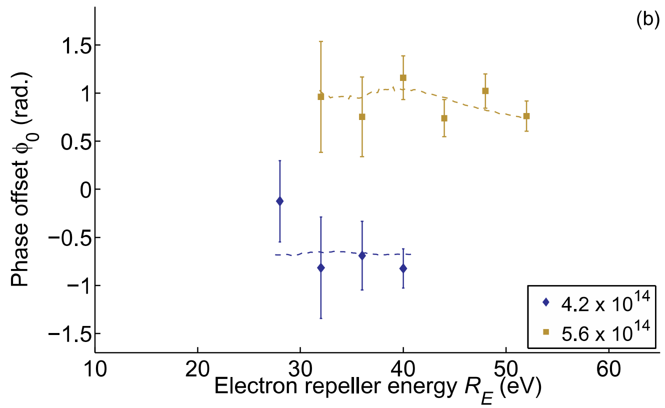
$$\approx A_0 \sin(\phi_{\mathrm{CEP}} - \phi_0).$$





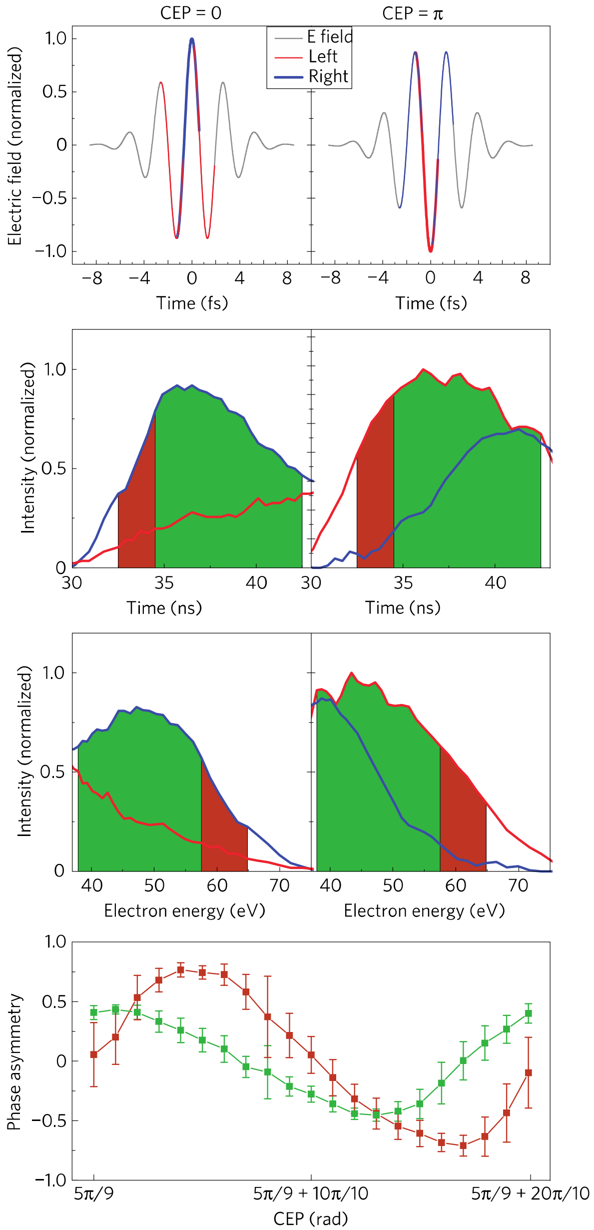






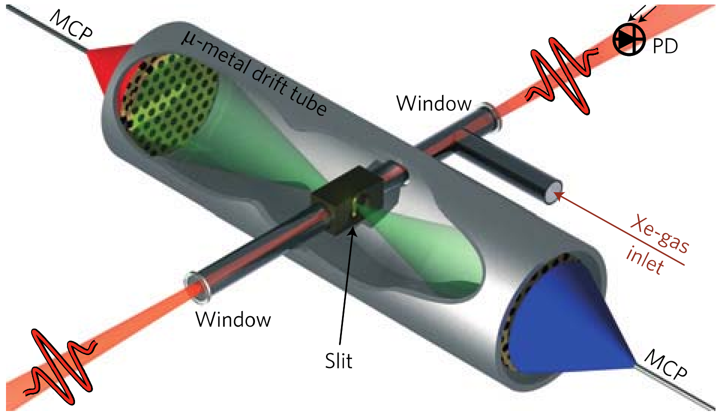
Wallace et al.,
New J. Phys. 15, 033002 (2013)

CEP measurement: stereo phasemeter

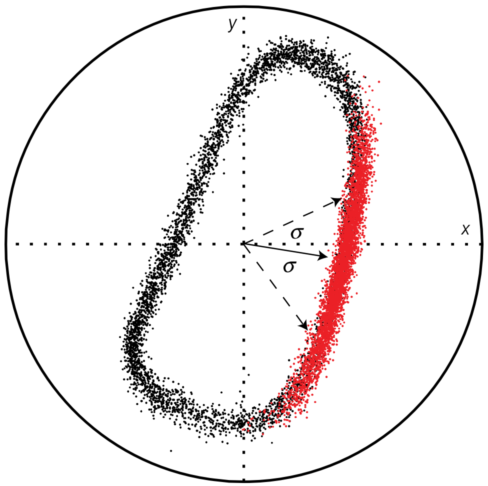


Single-shot carrier–envelope phase measurement of few-cycle laser pulses

T. Wittmann[1], B. Horvath[1], W. Helml[1], M. G. Schätzel[1], X. Gu[1], A. L. Cavalieri[1], G. G. Paulus[2,3] and R. Kienberger[1,4]★



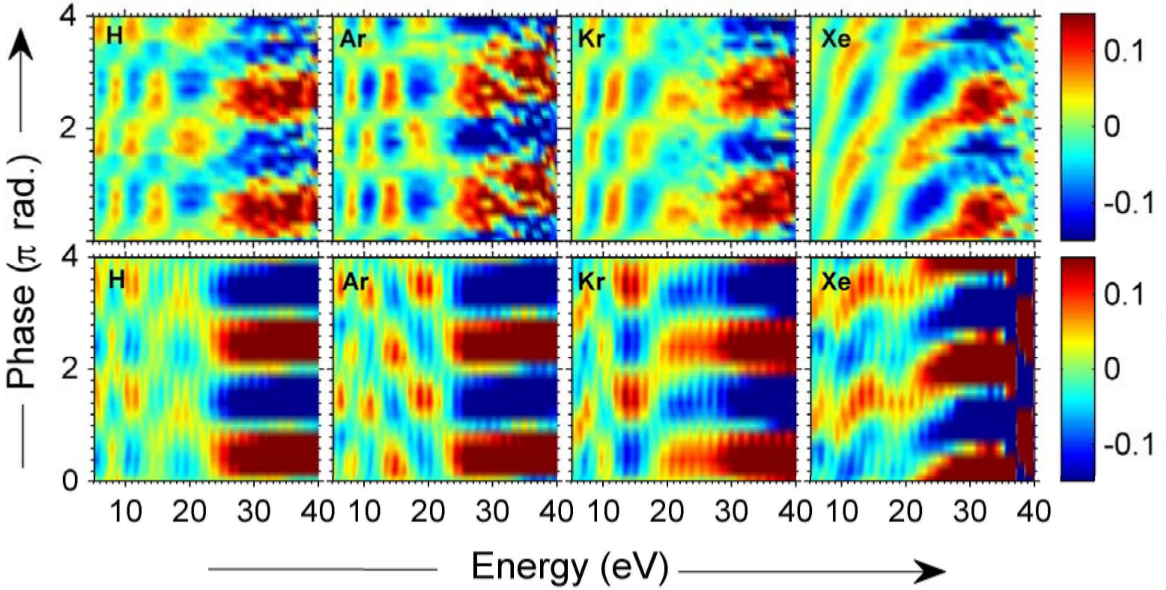
a



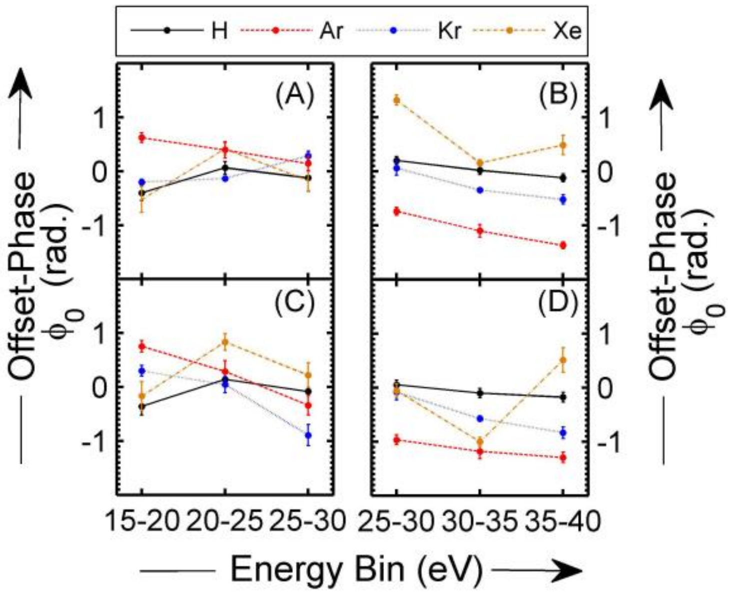
Wittmann et al., Nature Physics 5, 357 (2009)

Absolute CEP calibration with atomic H

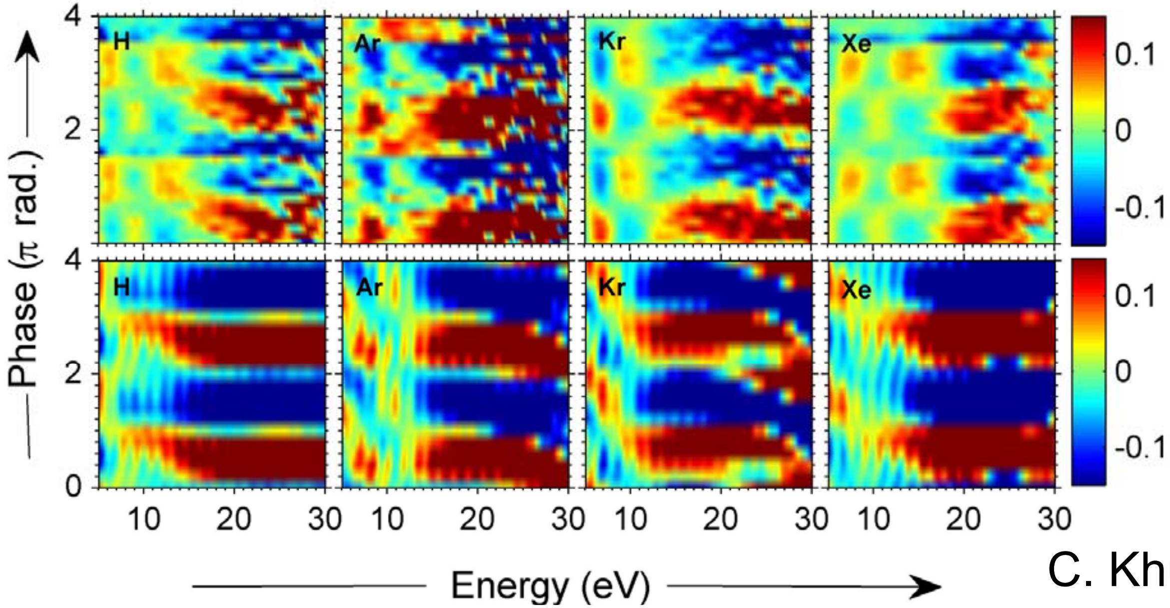
2.5×10^{14} W/cm^2



1.2 2.5



1.2×10^{14} W/cm^2

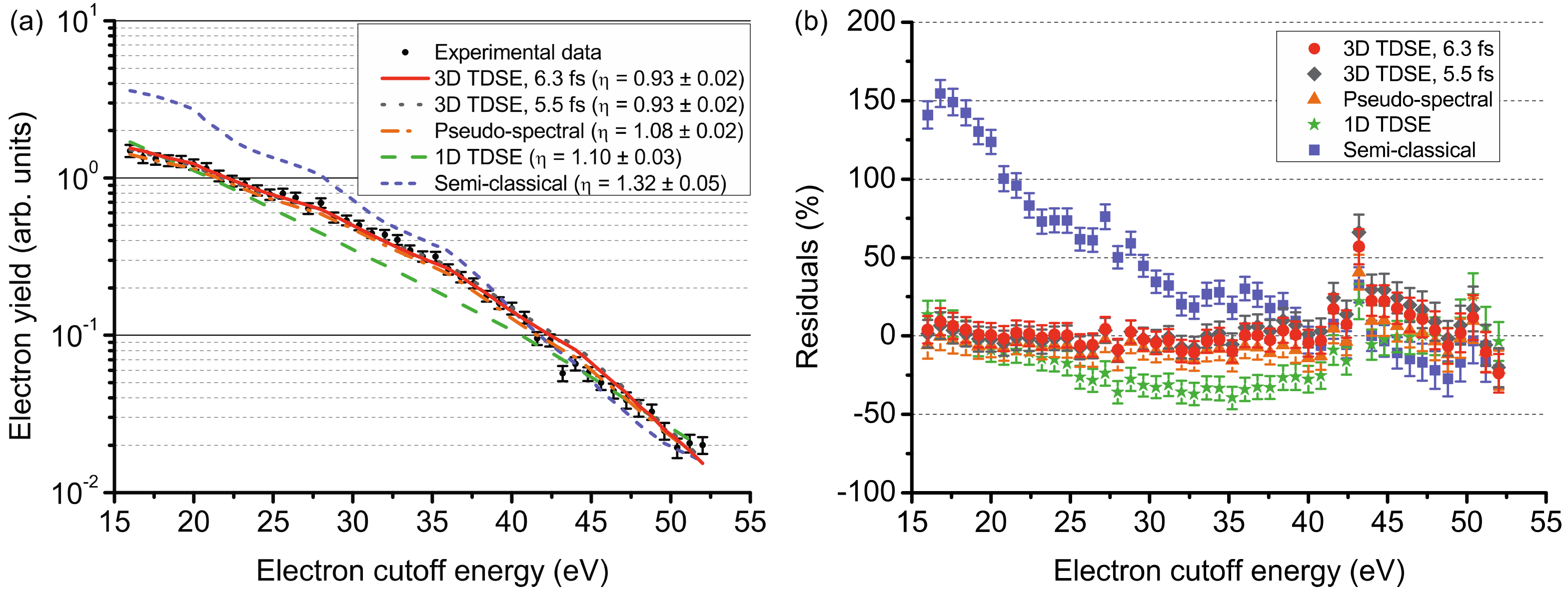


A, B – phase offset

C,D – difference between experiment and theory

C. Khurmi et al, PRA **96** 013404 (2017)

Precise intensity calibration

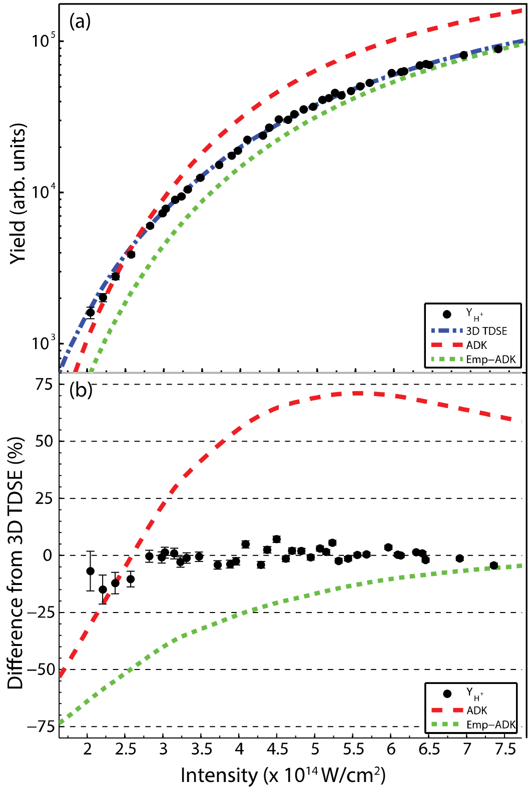


Measurement accuracy – 1%!

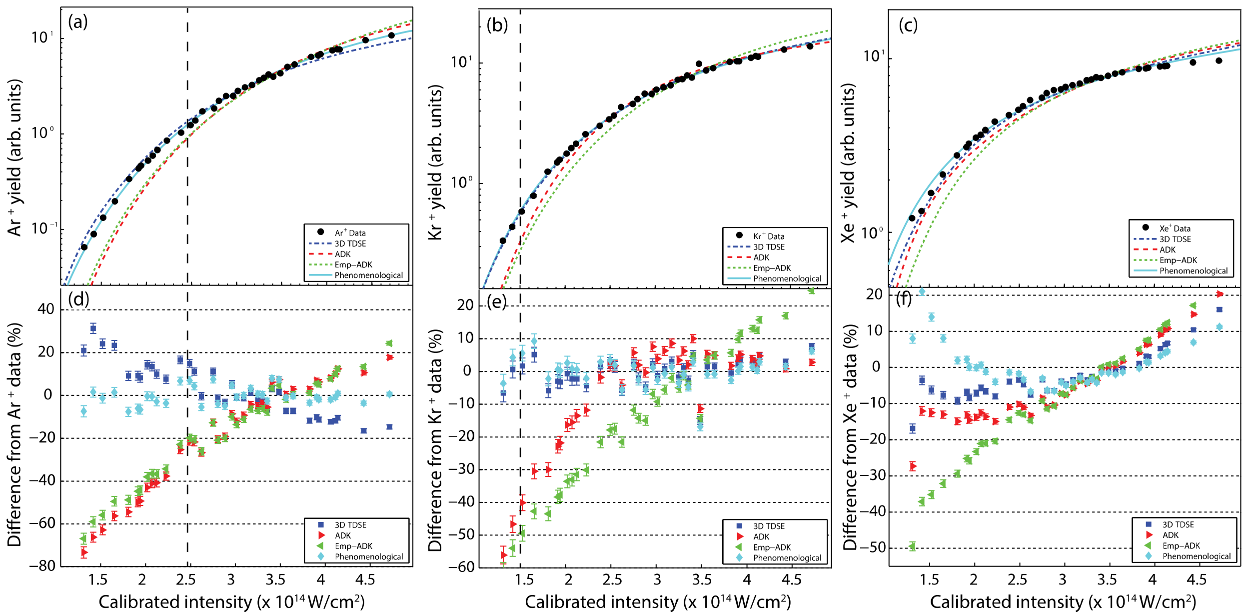
Pullen et al., Phys. Rev. A 87, 053411 (2013)

Transferable intensity calibration standard

H



Ar Kr Xe



$$S_{\text{phenom}}(\eta_2 I_{\text{est}}) = \frac{\exp[-\alpha(\eta_2 I_{\text{est}}/I_c)^{-1/2}]}{1+(\eta_2 I_{\text{est}}/I_c)^{\gamma}}$$

Fit parameter	Ar^+	Kr^+	Xe^+
α (arb. units)	2.84	4.24	3.71
γ (arb. units)	−3.03	−2.49	−2.69
I_c ($\times 10^{14}$ W/cm^2)	3.86 ± 0.05	2.06 ± 0.03	1.18 ± 0.03

By fitting phenomenological curves intensities could be determined with accuracies 1.3% using Ar, 1.5% using Kr and 2.5% using Xe.

Wallace et al., Phys. Rev. Lett 177, 053001 (2016)

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  - Attosecond streaking and tunneling time
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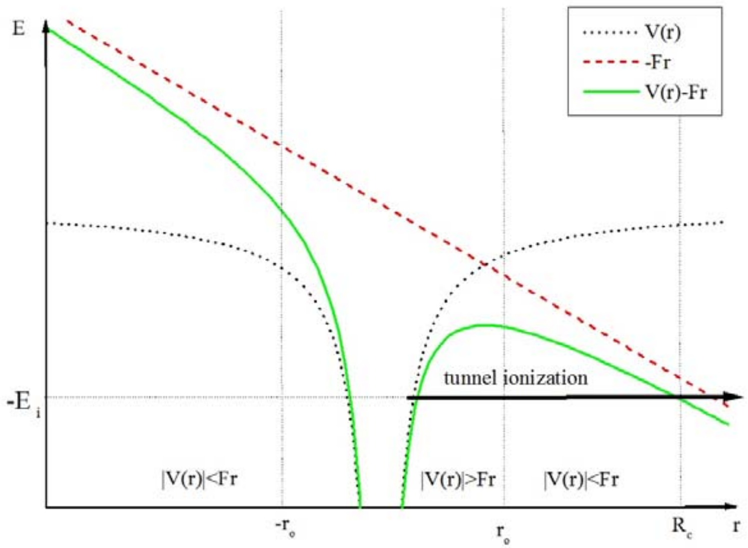
Attosecond streaking and tunneling time

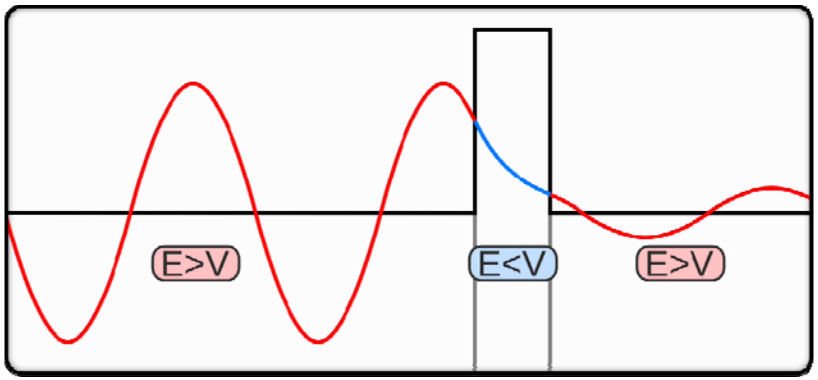


What is tunneling time?

Is it real? **Is it finite?**

Can it be measured?

Keldysh time



$$\tau_k = l/|v|$$

$$v = i\sqrt{|E_0|/2}$$

$$\gamma_k = 2\pi\,\tau_k/T_0$$

Tunneling regime $\gamma_k << 1$

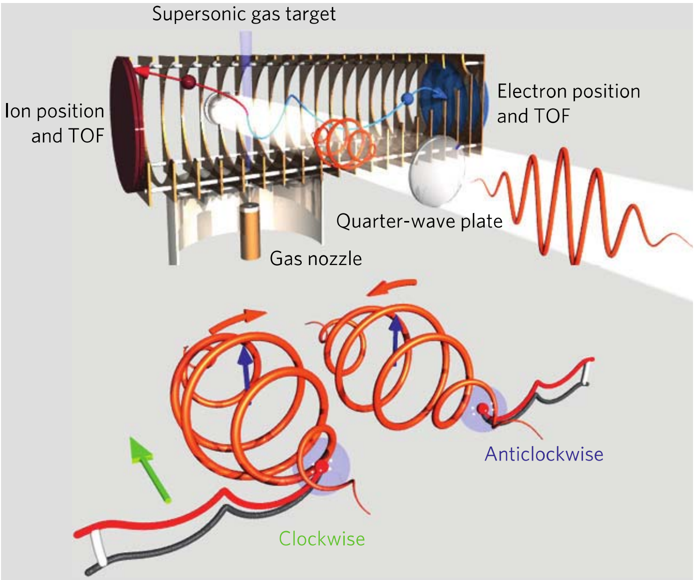
$\tau_k << T_0$

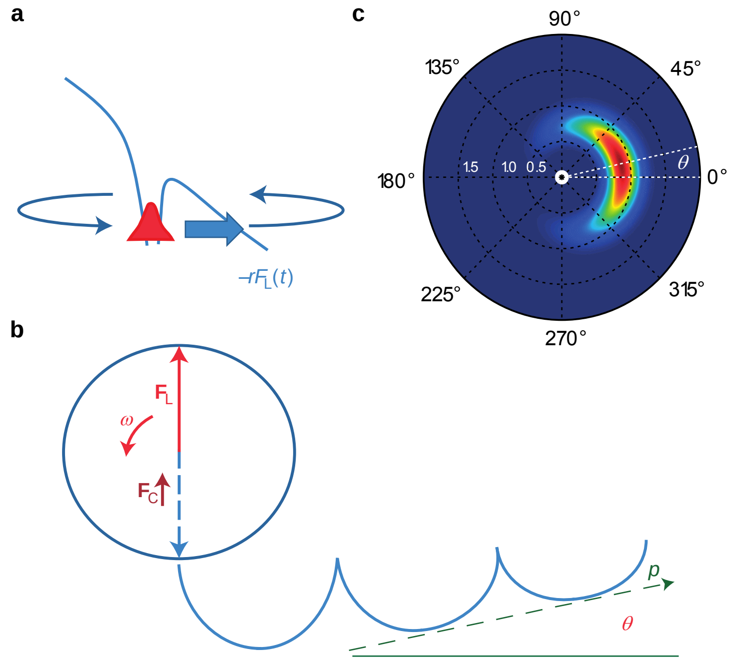
Attoclock experiments

LETTERS
PUBLISHED ONLINE: 23 OCTOBER 2011 | DOI: 10.1038/NPHYS2125
nature physics

Attoclock reveals natural coordinates of the laser-induced tunnelling current flow in atoms

Adrian N. Pfeiffer[1]*, Claudio Cirelli[1], Mathias Smolarski[1], Darko Dimitrovski[2]*, Mahmoud Abu-samha[2], Lars Bojer Madsen[2] and Ursula Keller[1]





Pfeiffer et al, Nature Phys. 47, 204003 (2014)

Attockclock interpretation

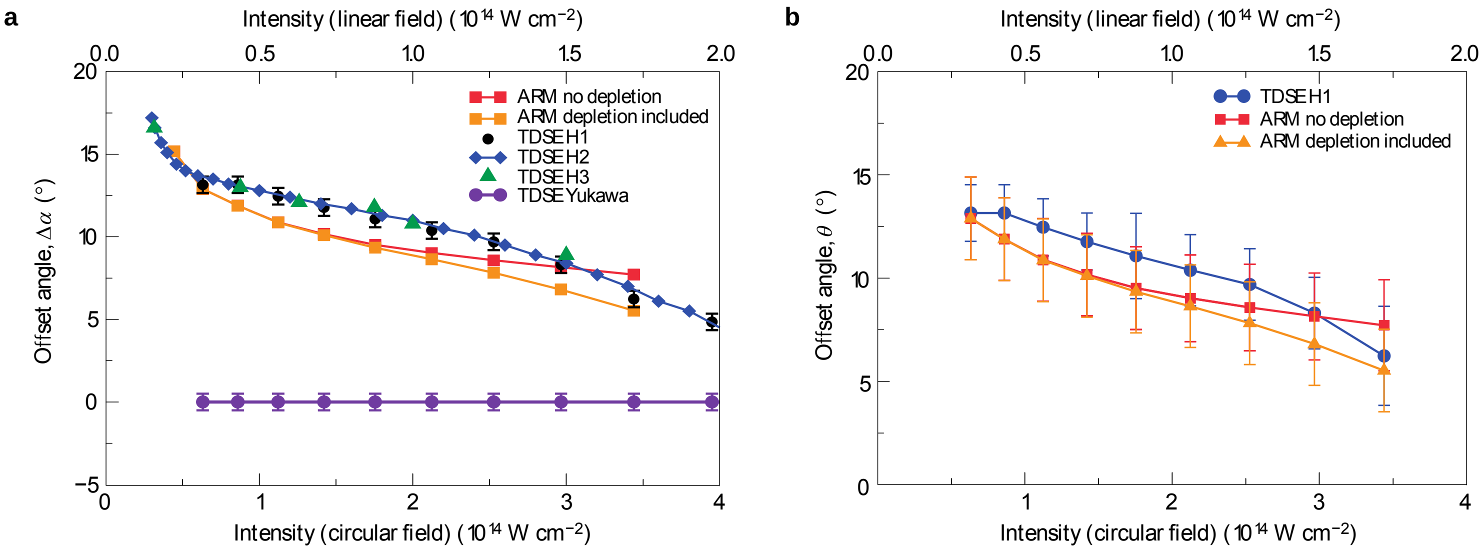
nature physics

ARTICLES

PUBLISHED ONLINE: 25 MAY 2015 | DOI: 10.1038/NPHYS3340

Interpreting attoclock measurements of tunnelling times

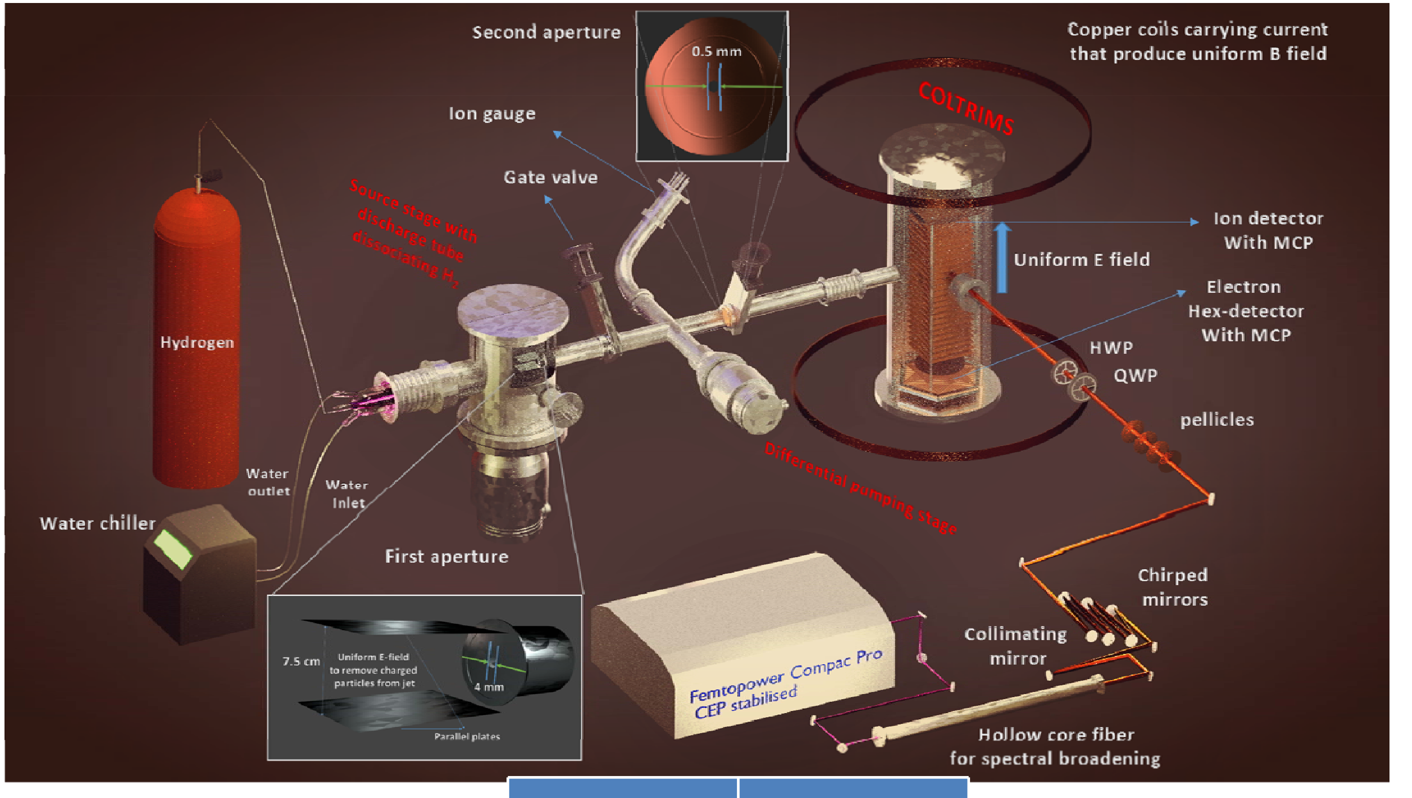
Lisa Torlina[1†], Felipe Morales[1†], Jivesh Kaushal[1], Igor Ivanov[2], Anatoli Kheifets[2], Alejandro Zielinski[3], Armin Scrinzi[3], Harm Geert Muller[1], Suren Sukiasyan[4], Misha Ivanov[1,4,5] and Olga Smirnova[1*]



Tunneling is instantaneous!

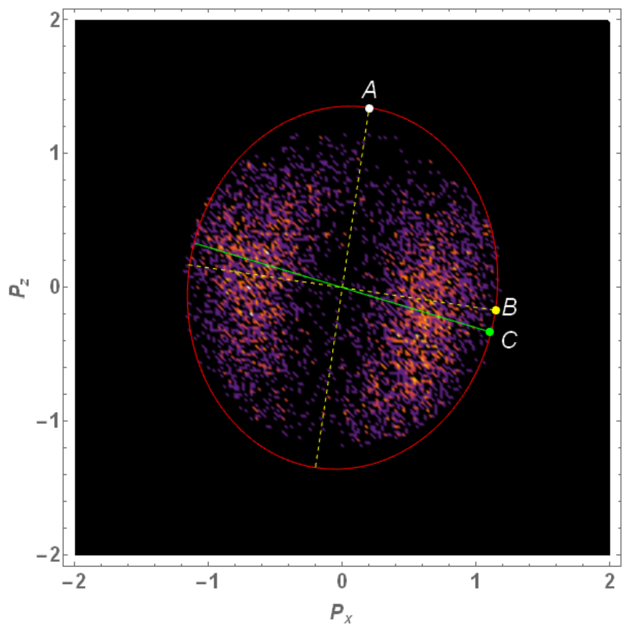
Torlina et al, Nature Phys. 11, 503 (2015)

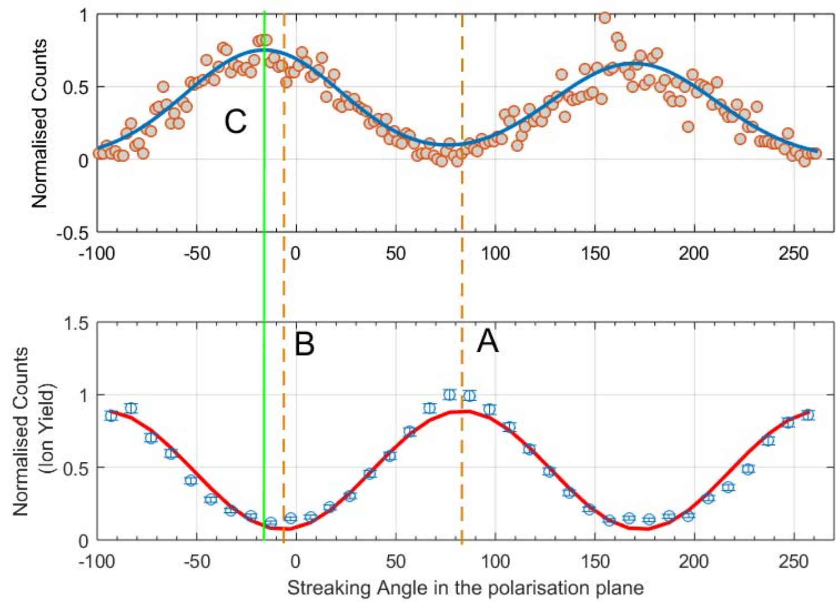
Attoclock with atomic hydrogen: experimental setup



H dissociation fraction	50%
λ	~800nm
Pulse duration	6.0 fs
ε	0.84±0.01

Attoclock with atomic hydrogen

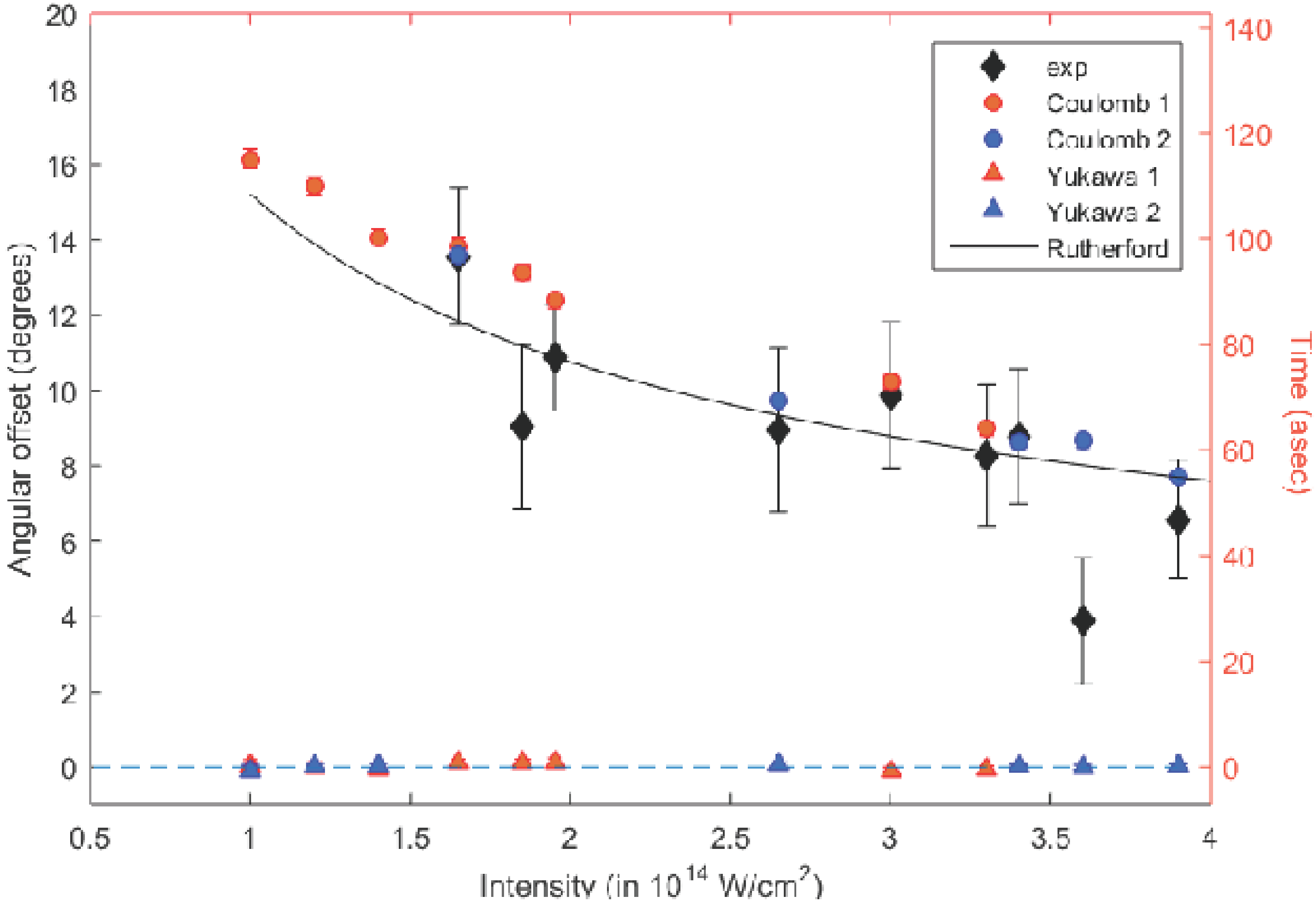




$$\theta_{tunnel} = \theta_{offset} - \theta_{streak} - \theta_{Coulomb}$$

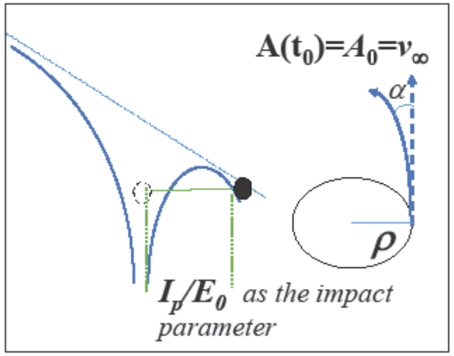
$\theta_{streak} = 90°$ $\theta_{Coulomb} = ?$

Attoclock with atomic hydrogen: experimental and theoretical results

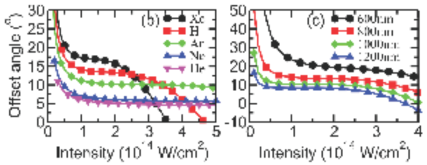


Attoclock with atomic hydrogen: Conclusions

- **Excellent agreement between experiment and 3D-TDSE (Coulomb) calculations.**
- **Angular offsets from theory, $\theta_{offset} \propto 1/\sqrt{Intensity}$.**
- **A simple Rutherford scattering model[5] for the electron in a Coulomb potential (Z/r) explains it qualitatively.**



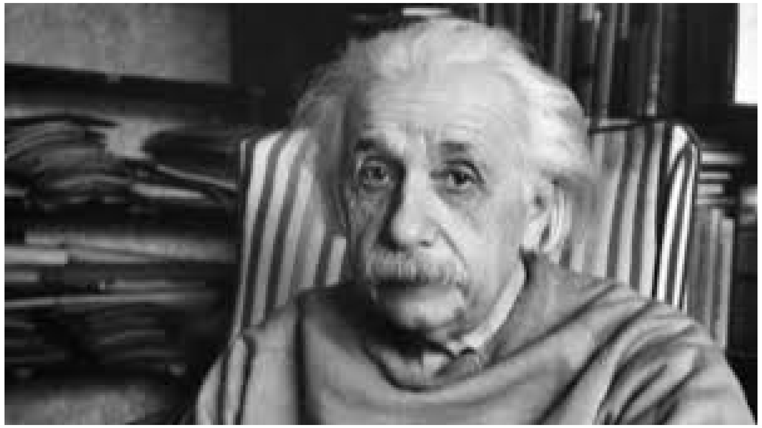
$$\boxed{\tan\left(\frac{\alpha}{2}\right)=\frac{1}{v_\infty^2}\frac{Z}{\rho}=\frac{1}{A_0^2}\frac{E_0}{I_p}=\frac{\omega^2}{E_0 I_p}}$$



Journal of Physics B: Atomic, Molecular and Optical Physics **50**, 055602 (2017).

- **Simulations with short-range Yukawa potential confirm that the tunnelling is instantaneous (within the numerical uncertainties).**
- **This puts an upper limit of 1.8 *asec* on possible delays due to tunnelling.**

For more details see Satya's poster MO-049 and *arXiv:1707.05445 (2017)*



“Everything should be made as simple as possible, but not simpler.”
(Albert Einstein)

A model should neglect as much as possible, but not more.

Commonly used approximations:

strong-field approximation (SFA), quasi-static approximation (QSA), single-active-electron approximation (SAE), dipole approximation, Born-Oppenheimer approximation (BOA), **frozen-nuclei approximation (FNA)**

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Ionization of molecular hydrogen

RAPID COMMUNICATIONS

PHYSICAL REVIEW A **87**, 041401(R) (2013)

Effect of nuclear motion on tunneling ionization rates of molecules

Oleg I. Tolstikhin,[1,2] Hans Jakob Wörner,[3] and Toru Morishita[4]

[1]*National Research Center "Kurchatov Institute," Kurchatov Square 1, Moscow 123182, Russia*
[2]*Moscow Institute of Physics and Technology, Dolgoprudny 141700, Russia*
[3]*Laboratorium für Physikalische Chemie, ETH Zürich, Wolfgang-Pauli-Strasse 10, 8093 Zürich, Switzerland*
[4]*Department of Engineering Science, The University of Electro-Communications, 1-5-1 Chofu-ga-oka, Chofu-shi, Tokyo 182-8585, Japan*

(Received 29 January 2013; published 9 April 2013)

We show that the observable rate of tunneling ionization of a molecule in an intense low-frequency laser field is affected by nuclear motion and can essentially differ from a bare electronic characteristic calculated for fixed nuclei. Both the absolute value of the rate and the shape of its orientation dependence are affected. The effect is significant for $I \sim 10^{14}$ W/cm^2 and becomes more pronounced at lower intensities. An isotope effect in tunneling ionization of H_2 and D_2 is predicted. The results are compared with available experiments.

$$\frac{\Gamma_0(H_2)}{\Gamma_0(D_2)} \approx e^{(1-1/\sqrt{2})\zeta(H_2)} \geqslant 1$$

$$\zeta(H_2) = 0.0017/F^2$$

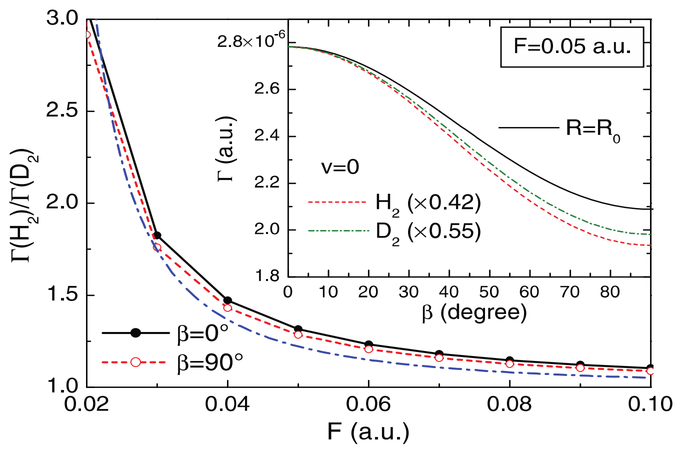
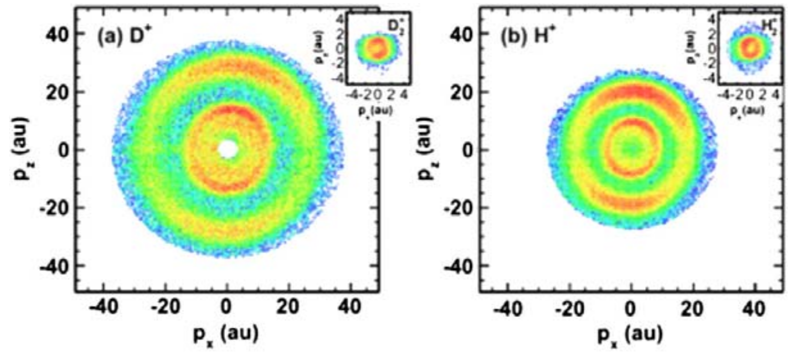


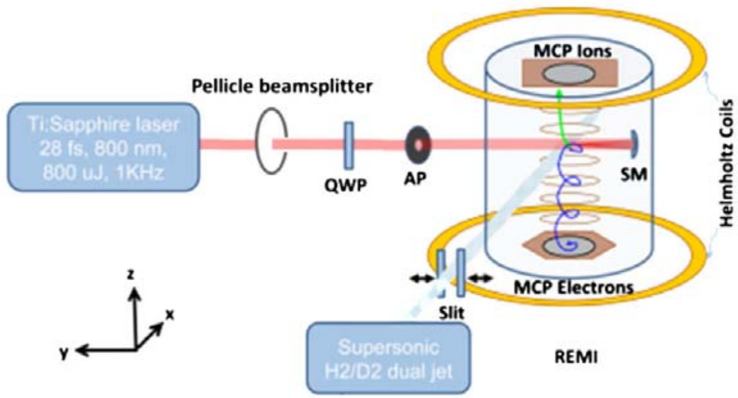
FIG. 3. (Color online) The ratio of the ionization rates $\Gamma_{v=0}(\beta)$ of H_2 and D_2 as a function of F for $\beta = 0^\circ$ (solid back line) and 90° (dashed red line). Dashed-dotted blue line: Results from Eq. (14). The inset shows $\Gamma_e(R_0,\beta)$ (solid line) and $\Gamma_0(\beta)$ for H_2 and D_2 (dashed lines) multiplied by the indicated factors.

Frozen nuclei approximation fails!

Tolstikhin, Worner, Morishita, Phys. Rev. A. 87, 041401(R) (2013)

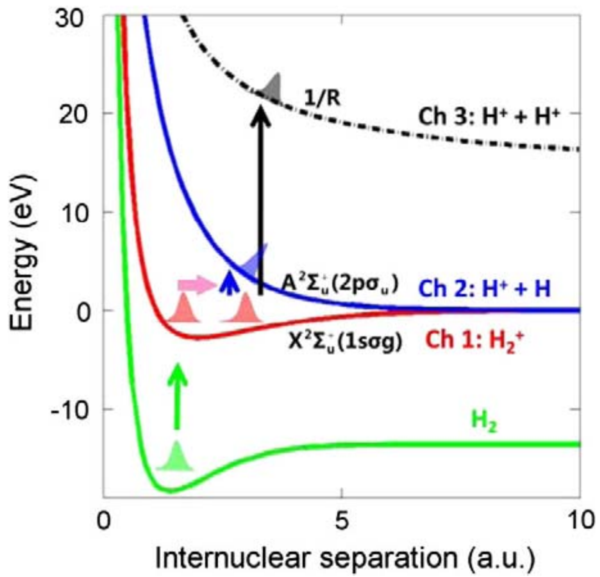
Isotope effect in tunneling ionization





QWP: Quarter-Wave Plate; AP: Aperature; SM: Spherical Mirror

REMI: Reactoion Microspcopes



(1): $H_2 \rightarrow H_2^+$

(2): $H_2 \rightarrow H_2^+ \xrightarrow{\text{diss}} H + H^+$

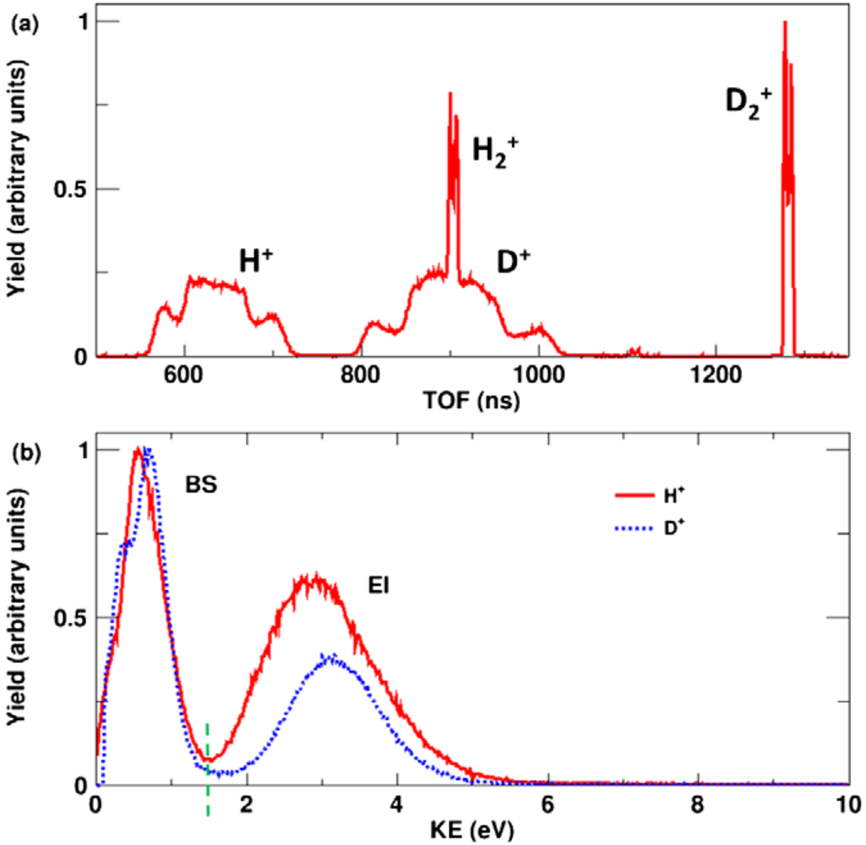
(3): $H_2 \rightarrow H_2^+ \rightarrow H_2^{++} \xrightarrow{\text{CE}} H^+ + H^+$

$$N_{\text{tot}} = N_1 + N_2 + 0.5N_3$$

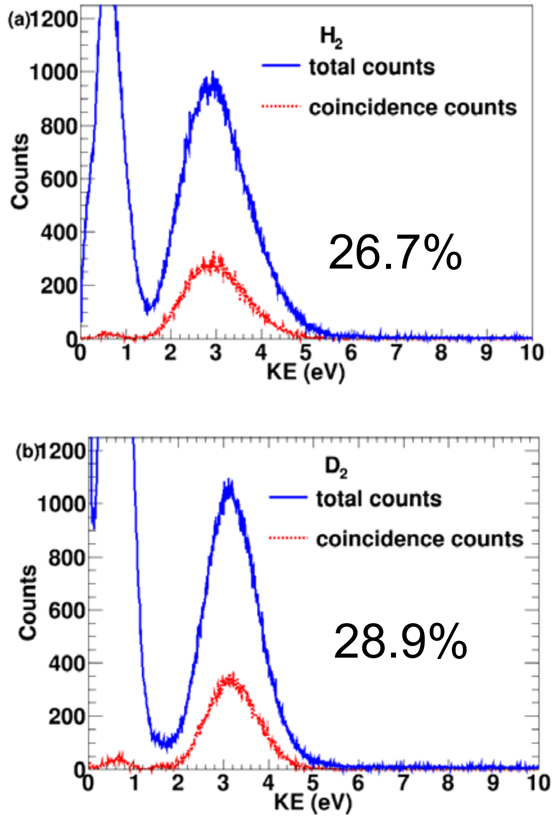
Wang et al., Phys. Rev. Lett. 117, 083003 (2016)

Experimental results

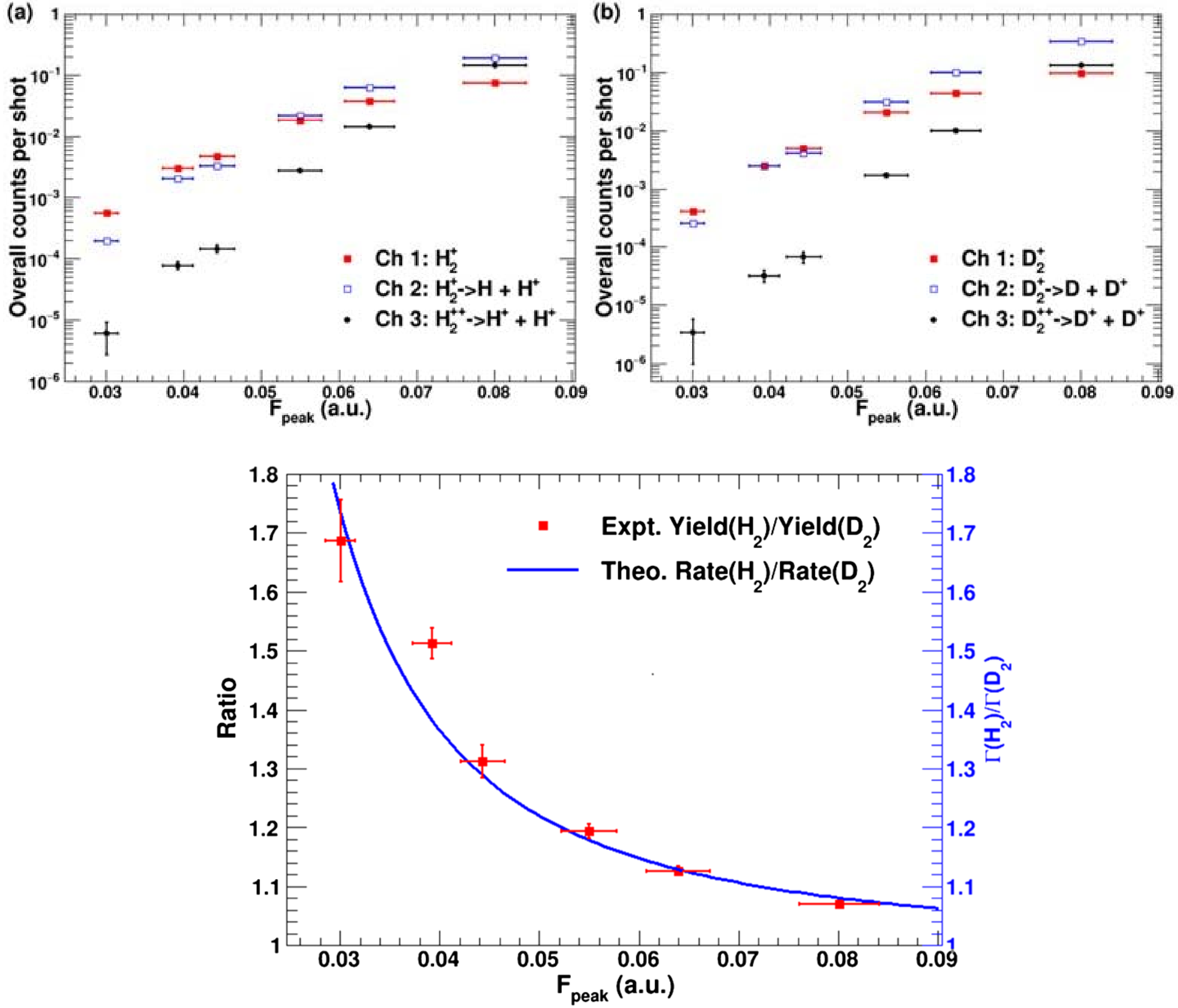
Separation of channels



Detector efficiency



Wang et al., Phys. Rev. Lett. 117, 083003 (2016)



Wang et al., Phys. Rev. Lett. 117, 083003 (2016)

Acknowledgments

Experiments

Griffith University

Han Xu
Satya Undurti
Atia-tul-Noor

Mick Pullen
Xiaoshan Wang
William Wallace
Champak Khumri
Dave Kielpinski

Prof Robert Sang

Theory

Shanghai Jiao Tong University

Feng He

Australian National University

Anatoli Kheifets
Alex Brey

University of Tsukuba

Xiaomin Tong

Drake University

Klaus Bartschat
Nicolas Douguet

Gwanju Institute of Science and Technology

Igor Ivanov

More research from Griffith group

Strong-field atomic ionization and excitation
Nida Haram **TH-016** *Intensity-dependent shift in transverse electron momentum distribution for strong field ionization*
Rohan Glover **MO-026** *Metastable argon production via strong-field excitation*

Laser-ablated plasmas
Smijesh N. **TU-031** *Optimization of laser plasma dynamics towards high order harmonic generation applications*
Kavya Rao **TU-032** *Plume dynamics of a laser produced plasma:Single and double pulse schemes*

Ultrafast molecular dynamics
Han Xu **WE-033** *Observing electron localization in a dissociating molecule in real time*
Atia-Tul-Noor **FR-039** *Enhanced ionization of* C_2H_2

High harmonics interferometry
Mumta Mustary **FR-024** *Attosecond time delay in harmonic emissions of* H_2 *and* D_2