Attosecond ionization dynamics

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Invited Talk, Wednesday 3. Sept. 2014

Frontiers of Intense Laser Physics
The Kavli Institute for Theoretical Physics
July 21 – Sep. 19, 2014, Santa Barbara, USA

Attoscience team members, ETH Zurich

Attoclock and Atto-COLTRIMS
Dr. Claudio Cirelli
Robert Boge
Sebastian Heuser
Mazyar Sabbar

VMIS
Dr. Jochen Maurer
Andre Ludwig

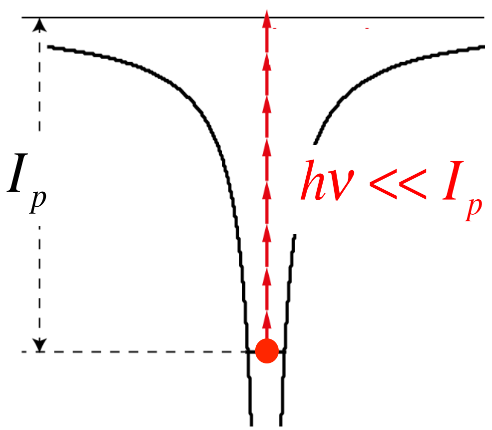
Dr. Matteo Lucchini
Dr. Lukas Gallmann

Theory
Cornelia Hofmann
Dr. Alexandra Landsman

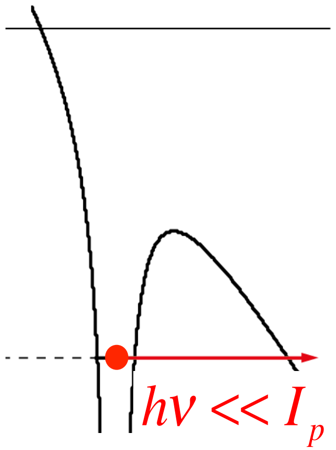
Attoscience: we ask the question ...

How long does it take for light to remove an electron from an atom, molecule and surface?

Multi-photon ionization (MPI) and

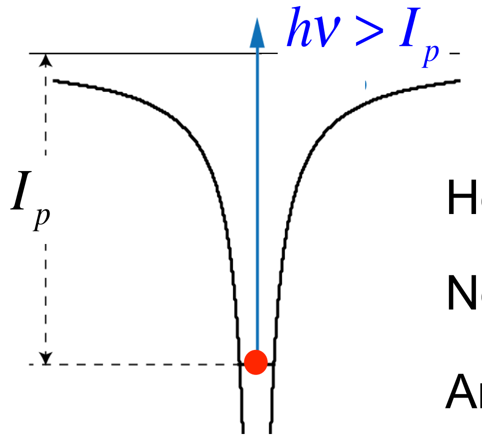


tunnel ionization



photoemission

absorption of a single photon



He: I_p = 24.59 eV

Ne: I_p = 21.56 eV

Ar: I_p = 15.76 eV

What is the scaling from 1 photon to n photon absorption?

Nature Photonics, March 2014

COMMENTARY | FOCUS

What will it take to observe processes in 'real time'?

Stephen R. Leone, C. William McCurdy, Joachim Burgdörfer, Lorenz S. Cederbaum, Zenghu Chang, Nirit Dudovich, Johannes Feist, Chris H. Greene, Misha Ivanov, Reinhard Kienberger, Ursula Keller, Matthias F. Kling, Zhi-Heng Loh, Thomas Pfeifer, Adrian N. Pfeiffer, Robin Santra, Kenneth Schafer, Albert Stolow, Uwe Thumm and Marc J. J. Vrakking

Even for simple systems, the interpretations of new attosecond measurements are complicated and provide only a glimpse of their potential. Nonetheless, the lasting impact will be the revelation of how short-time dynamics can determine the electronic properties of more complex systems.

Challenge 1: Time is not an operator in quantum mechanics!

Challenge 2: All at the same time: strong IR laser field, XUV pulse, Coulomb potential, many electrons ...
What approximations are ok when?

Attosecond streaking and interferometric techniques

Attosecond energy streaking (Krausz group, Munich)

Hentschel M. et al., Nature 414, 509, 2001

M. Schultze et al, Science 328, 1658, 2010 – for ionization delay

Attosecond angular streaking: attoclock (Keller group, Zurich)

P. Eckle et al., *Science* **322**, 1525, 2008 – for tunnel ionization delay

Attosecond interferometer: RABBITT (L'Huillier group, Lund)

P. M. Paul *et al.*, *Science* **292**, 1689, 2001 – for pulse characterization

K. Klünder et al, PRL 106, 143002, 2011 – for ionization delay

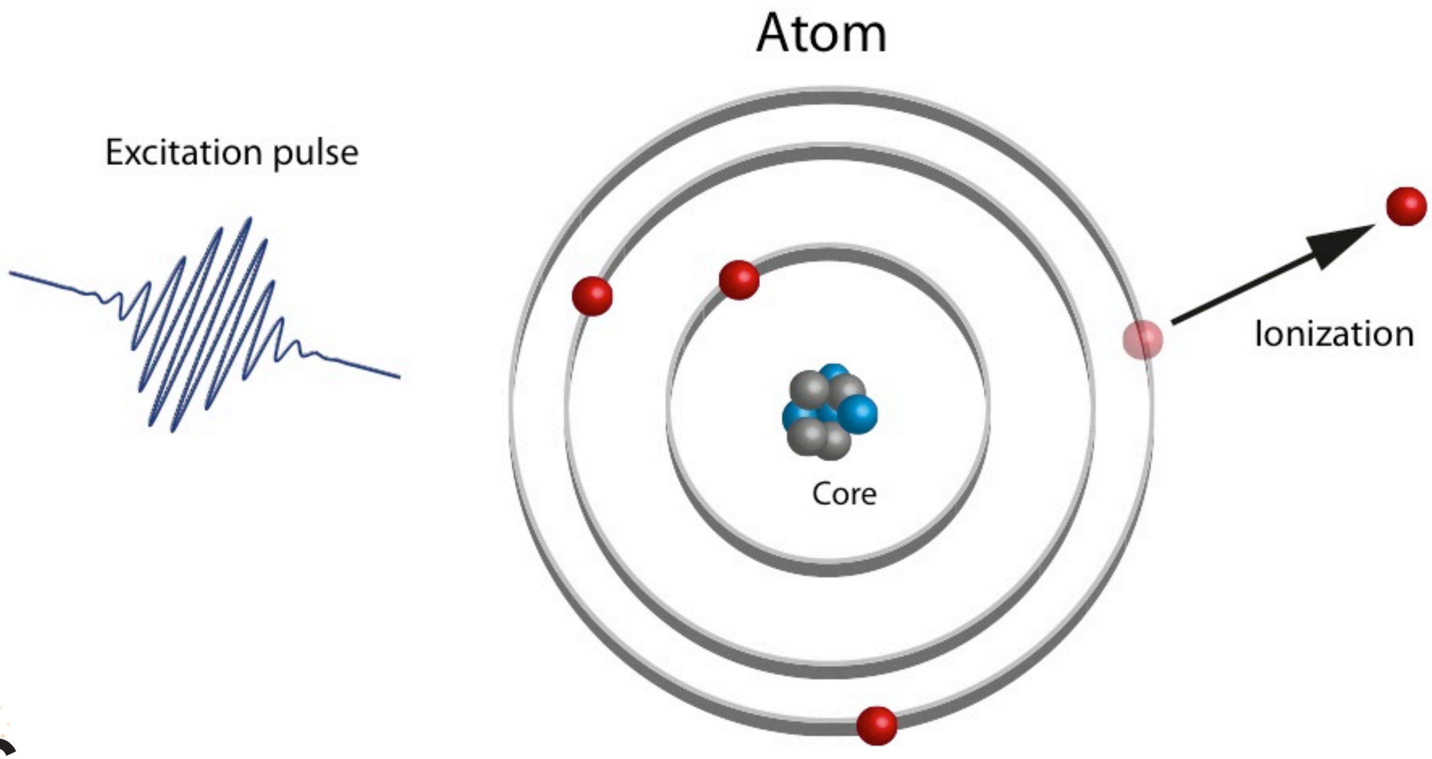
Do we in all cases measure the same time delays?

Time is not an operator in quantum mechanics – so ionization time and tunneling time is a highly debated topic.

Time delay for photoemission (single photon event)

Electron motion in atoms: attosecond time scale (10^{-18} s)

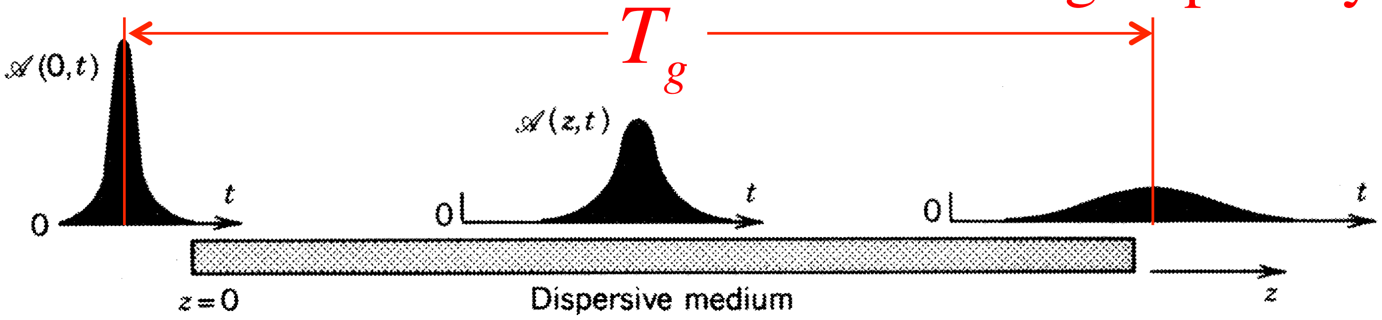
How long does it take to remove an electron from an atom
with one XUV photon pulse?



Analogy between photon and electron wave packet

photon wave packet (laser pulse)

group delay: $T_g = \frac{z}{\upsilon_g} = \frac{d\phi}{d\omega}$

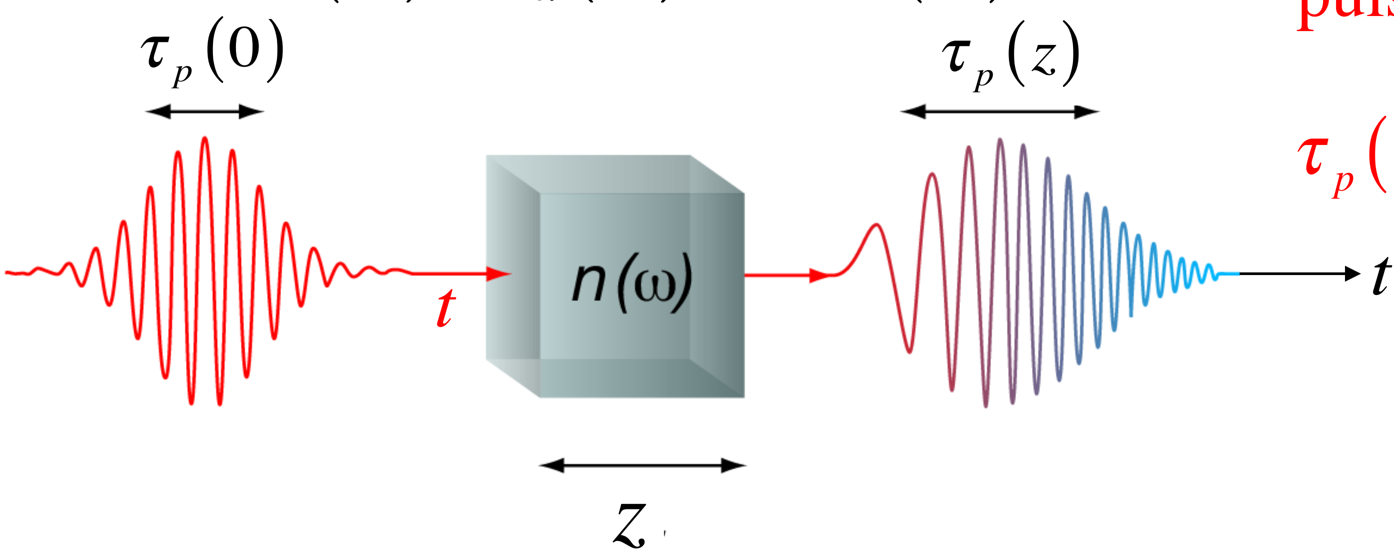


Dispersive medium

$$\phi(\omega) = k_n(\omega) z = k\, n(\omega) z$$

pulse broadening:

$$\tau_p(z) \approx \frac{d^2\phi}{d\omega^2} \Delta\omega_p$$



Analogy between photon and electron wave packet

photon in vacuum

$$E = cp \quad \Leftrightarrow \quad \omega(k) = ck$$

Phase velocity

$$\upsilon_p = \frac{\omega}{k} = c$$

Group velocity

$$\upsilon_g = \frac{d\omega}{dk} = c$$

electron in vacuum

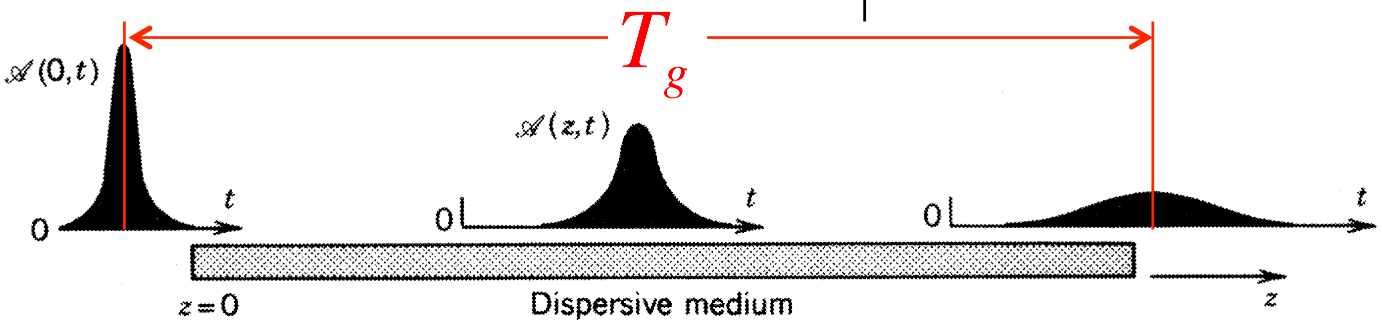
$$E = \frac{p^2}{2m} \quad \Leftrightarrow \quad \omega(k) = \frac{\hbar}{2m} k^2$$

$$\upsilon_p = \frac{\omega}{k} = \frac{\hbar k}{2m}$$

$$\upsilon_g = \frac{d\omega}{dk} = \frac{\hbar k}{m}$$

$$\upsilon_g = \frac{\hbar k}{m} = \frac{p}{m} = \frac{m\upsilon_{class}}{m} = \upsilon_{class}$$

group delay: $T_g = \frac{z}{\upsilon_g} = \frac{d\phi}{d\omega}$

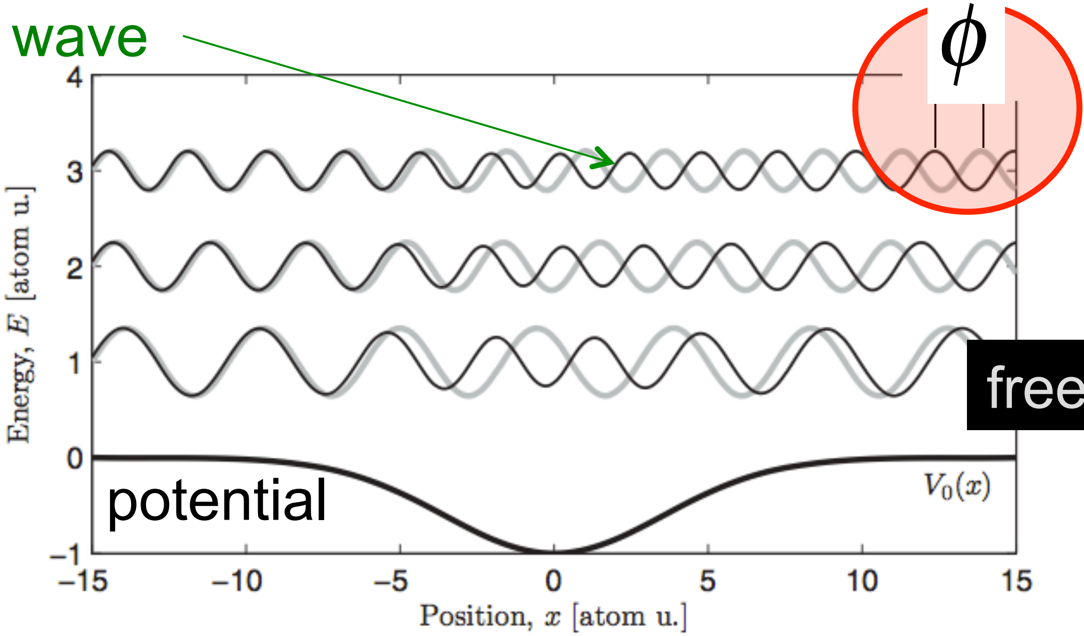


What time delay do we measure?

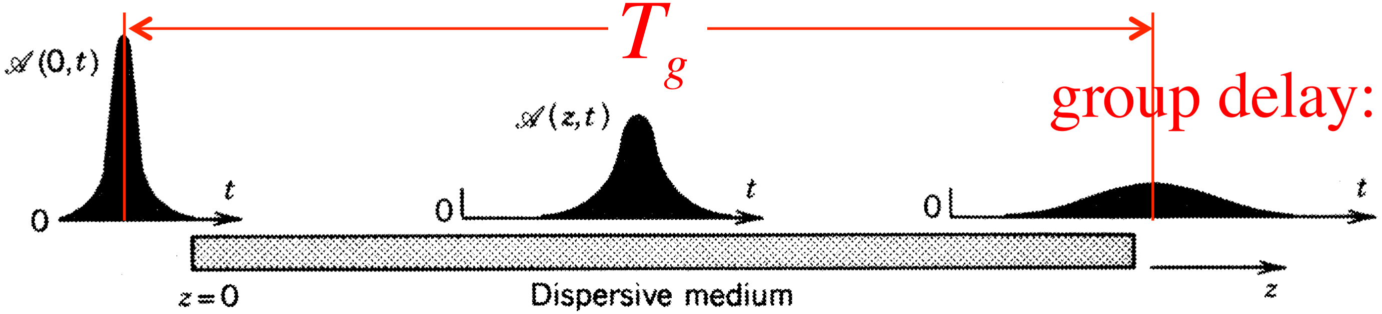
From scattering theory: Wigner delay

E. P. Wigner, Physical Review 98, 145 (1955)

$$\tau_W \equiv \frac{\partial \phi}{\partial \omega} \xrightarrow{E=\hbar\omega} = \hbar \frac{\partial \phi}{\partial E}$$



J. M. Dahlström, Alfred Maquet, Anne L'Huillier, Joachim Burgdörfer, et al.



group delay: $T_g = \frac{z}{\upsilon_g} = \frac{d\phi}{d\omega}$

Comparison: RABBITT & Energy streaking

Attosecond interferometer: RABBITT

$$\tau_{RABBITT} = \tau_{Wigner} + \tau_{CC}$$

Attosecond energy streaking

$$\tau_{Streaking} = \tau_{Wigner} + \tau_{CLC}$$

$$\tau_{CC} = \tau_{CLC}$$

A. S. Kheifets, PRA **87**, 063404 (2013)

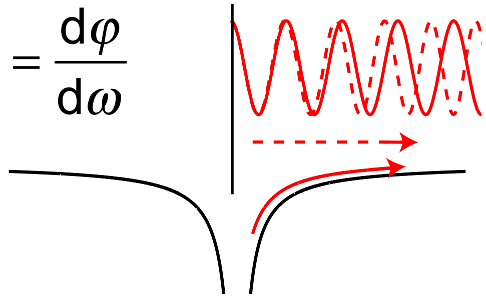
J. Mauritsson *et al.*, PRA **72**, 013401 (2005)

J. M. Dahlström *et al.*, PRA **86**, 061402(R) (2012)

τ_{Wigner}: Wigner delay

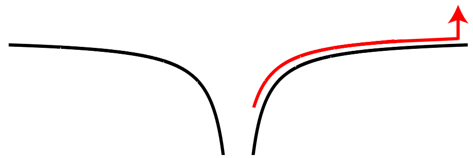
Electron escapes potential, no laser

$$\tau_{Wigner} = \frac{d\varphi}{d\omega}$$



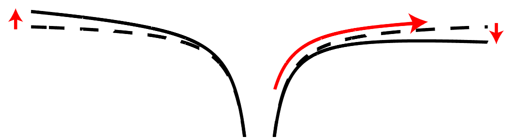
τ_{CC}: Continuum-Continuum transition:

IR photon absorption



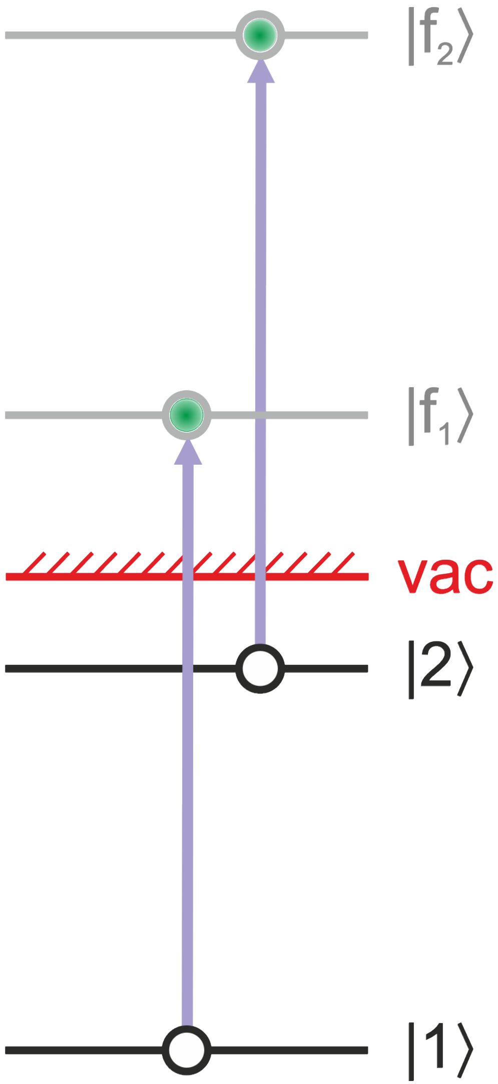
τ_{CLC}: Coulomb-Laser Coupling

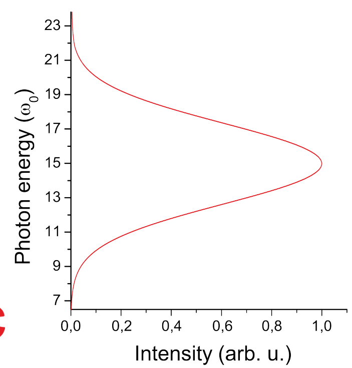
Streaking field interplay with Coulomb potential

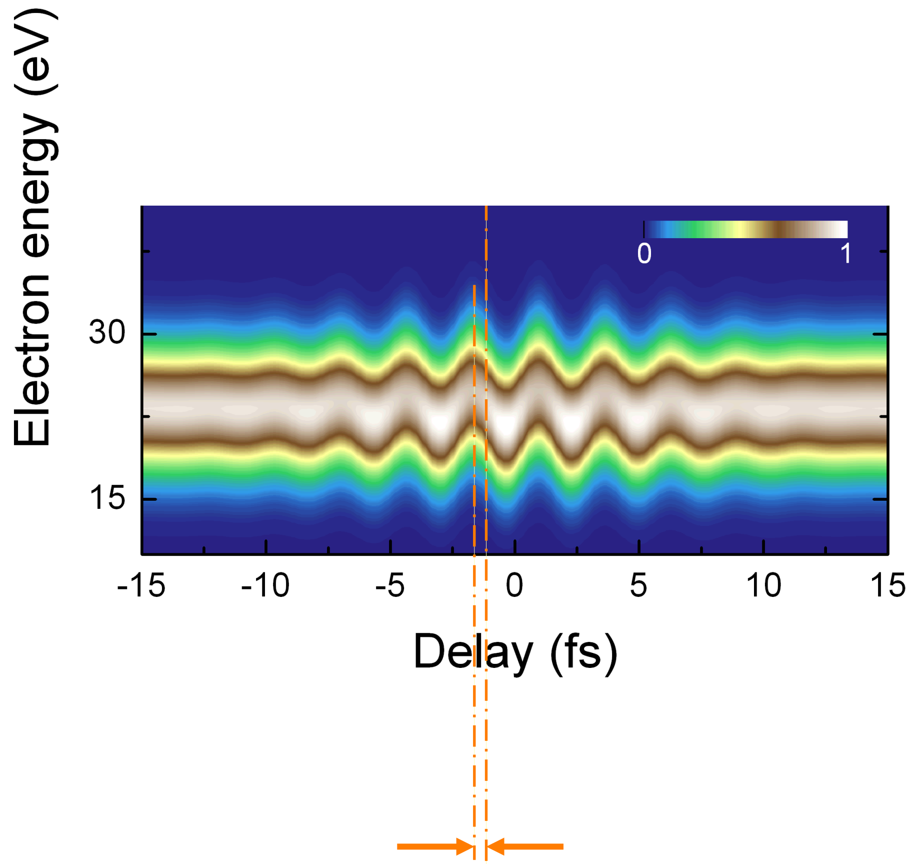


Single attosecond pulse and photoemission delays

Attosecond energy streaking
(attosecond streak camera)



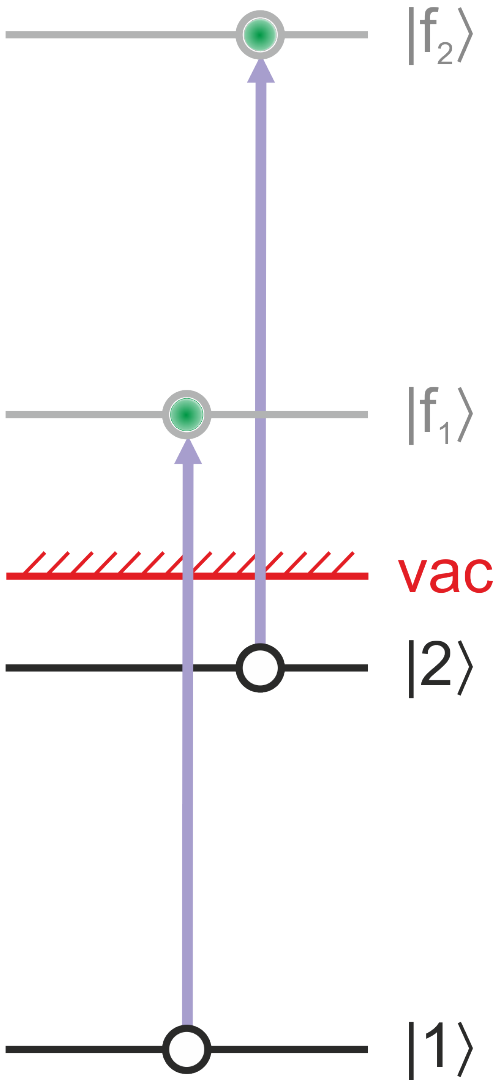


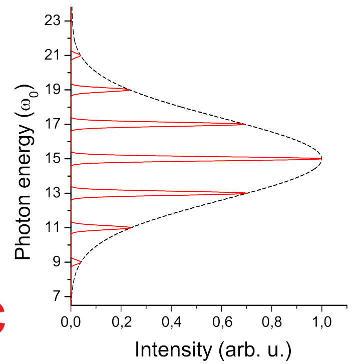


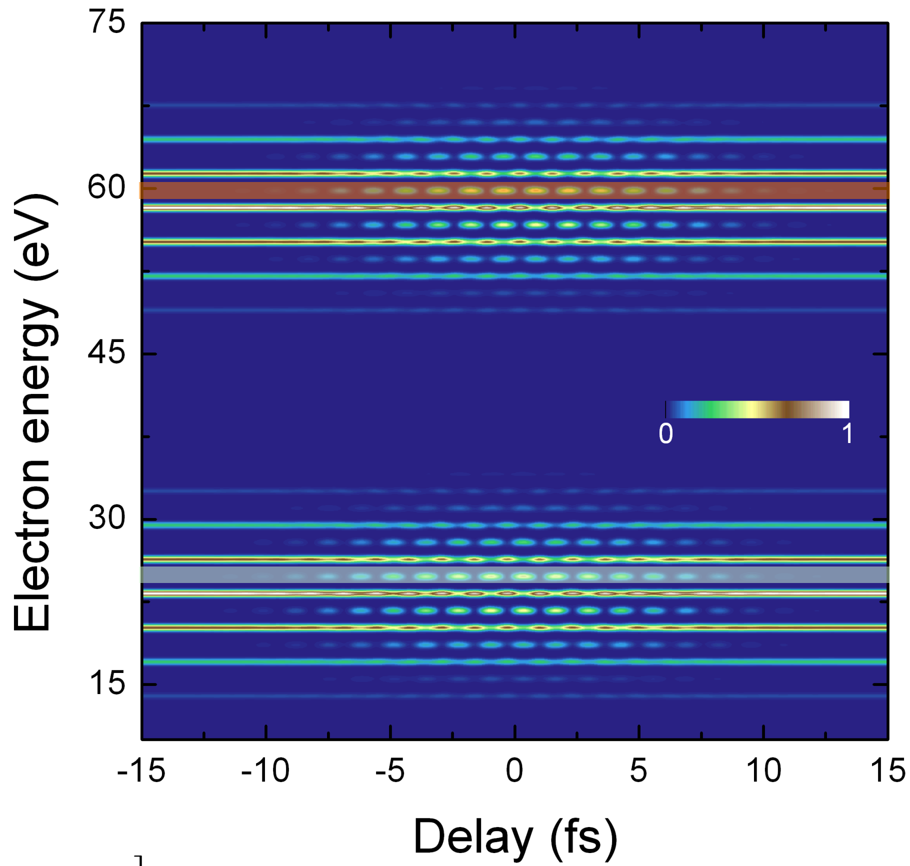
Relative delay τ
between the two states

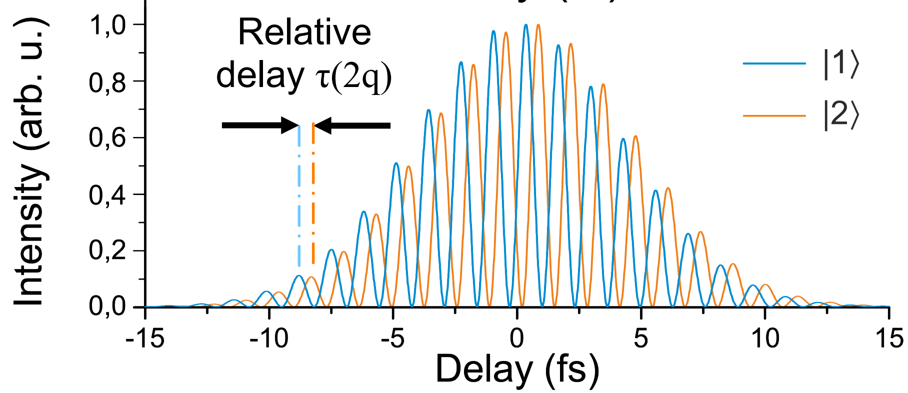
Attosecond pulse trains and photoemission delays

Attosecond interferometer: RABBITT









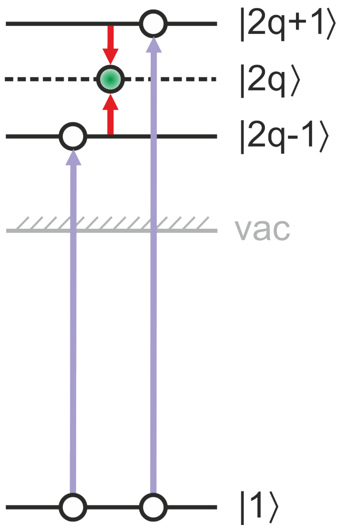
RABBITT formalism

Emission path: $$\varphi_{2q}^{(e)} = \varphi_{2q+1} - \omega_0\tau + \varphi_{Wig,\,2q+1} + \varphi_{cc,\,2q+1}$$

harmonic phase — IR phase — transition amplitude phase — continuum-continuum phase

Absorption path: $$\varphi_{2q}^{(a)} = \varphi_{2q-1} + \omega_0\tau + \varphi_{Wig,\,2q-1} + \varphi_{cc,\,2q-1}$$

Interference: $$\propto 1+\cos\left(\varphi_{2q}^{(a)} - \varphi_{2q}^{(e)}\right) = 1+\cos(2\omega_0\tau - \phi_{2q})$$



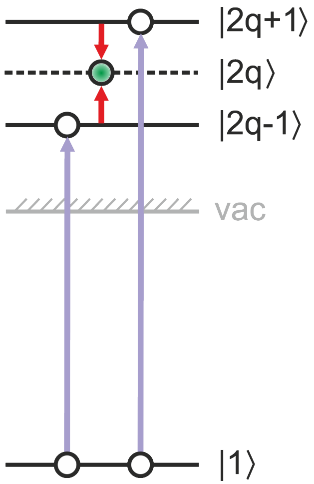
$$\phi_{2q} = \phi_{2q}^{(e)} - \phi_{2q}^{(a)} = \Delta\varphi_{2q} + \boxed{\Delta\varphi_{Wig,\,2q} + \Delta\varphi_{cc,\,2q}} + \varphi_0$$

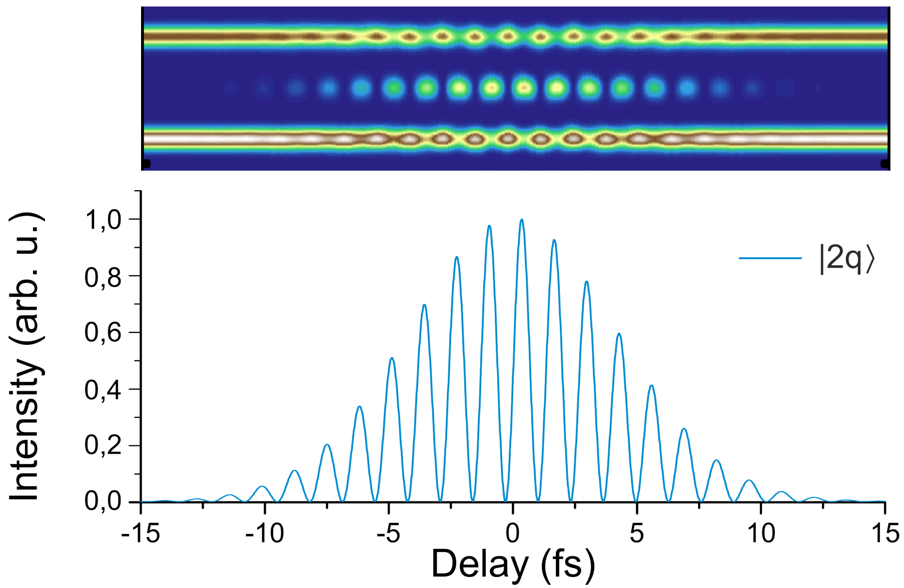
offset phase

$$\Delta x_{2q} = x_{2q+1} - x_{2q-1}$$

Principle of RABBITT

Reconstruction of Attosecond Beating By Interference of Two-photon Transitions





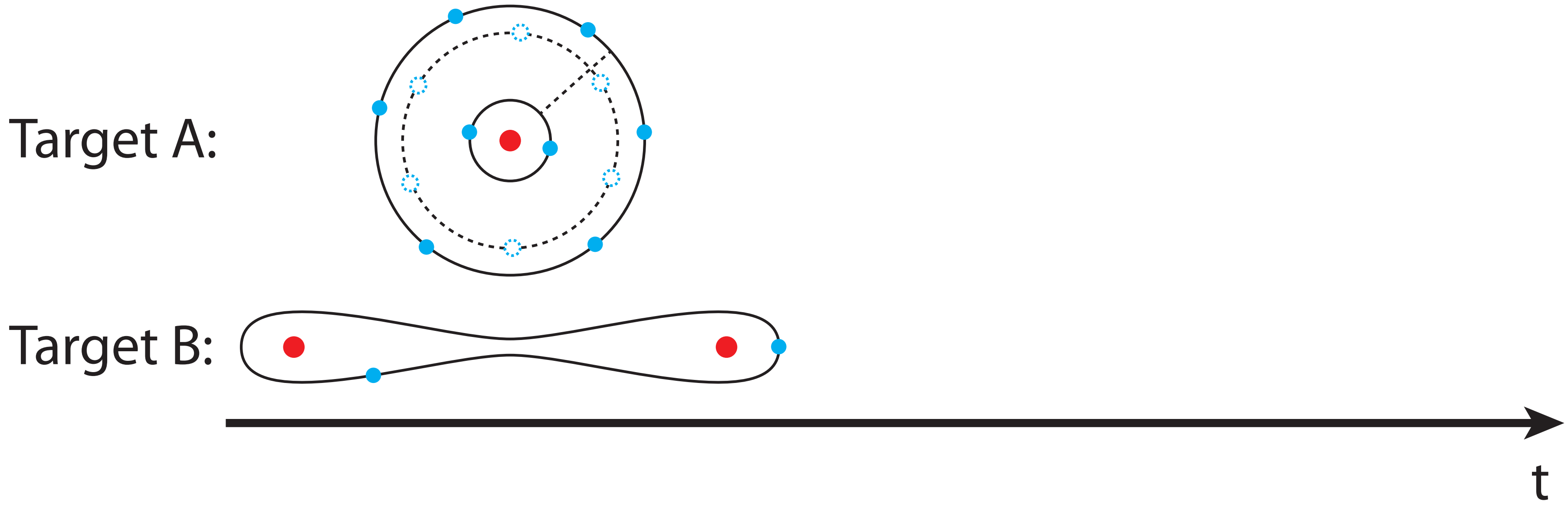
$$SB_{2q} \sim cos\left(2\omega_0\tau - \Phi_{2q}\right) = cos\left(2\omega_0\tau - \Delta\theta_{2q} - \Delta\varphi_{2q}^{At}\right)$$

$$\frac{\Delta\varphi_{2q}^{At}}{2\omega_0} = \tau_{2q}^{At} = \tau_{2q}^{Wig} + \tau_{2q}^{cc}$$

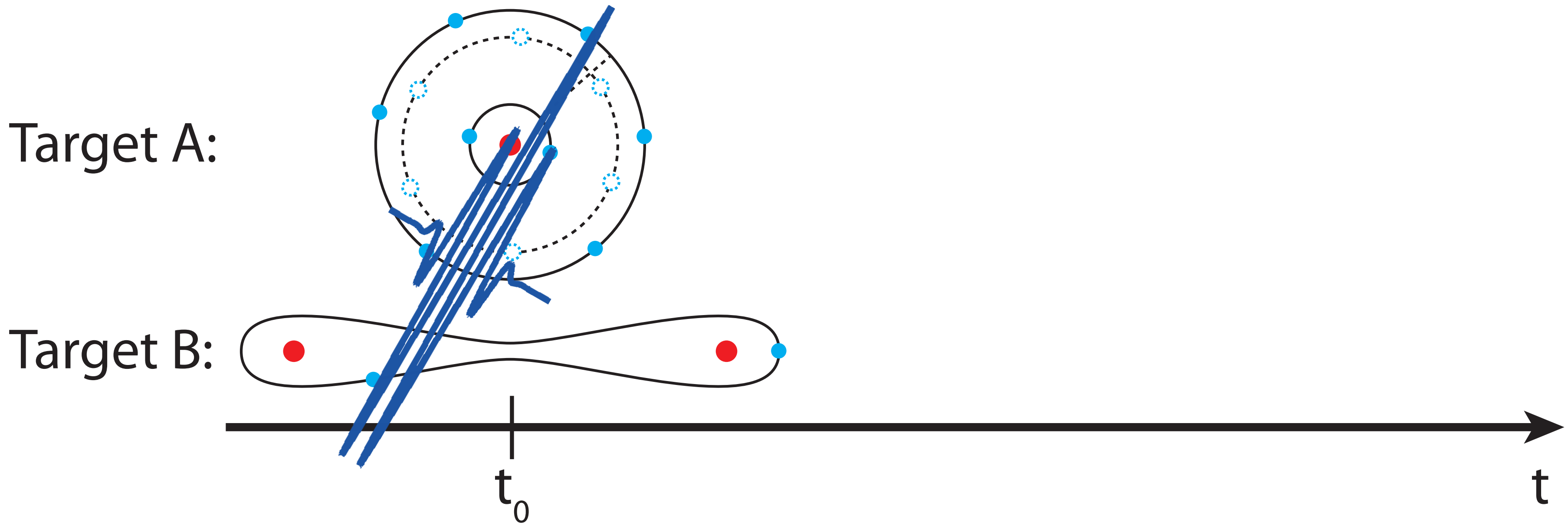
P. M. Paul et al., Science 292, 1689 (2001)

J. M. Dahlström et al., Chem. Phys. 414, 53-64 (2013)

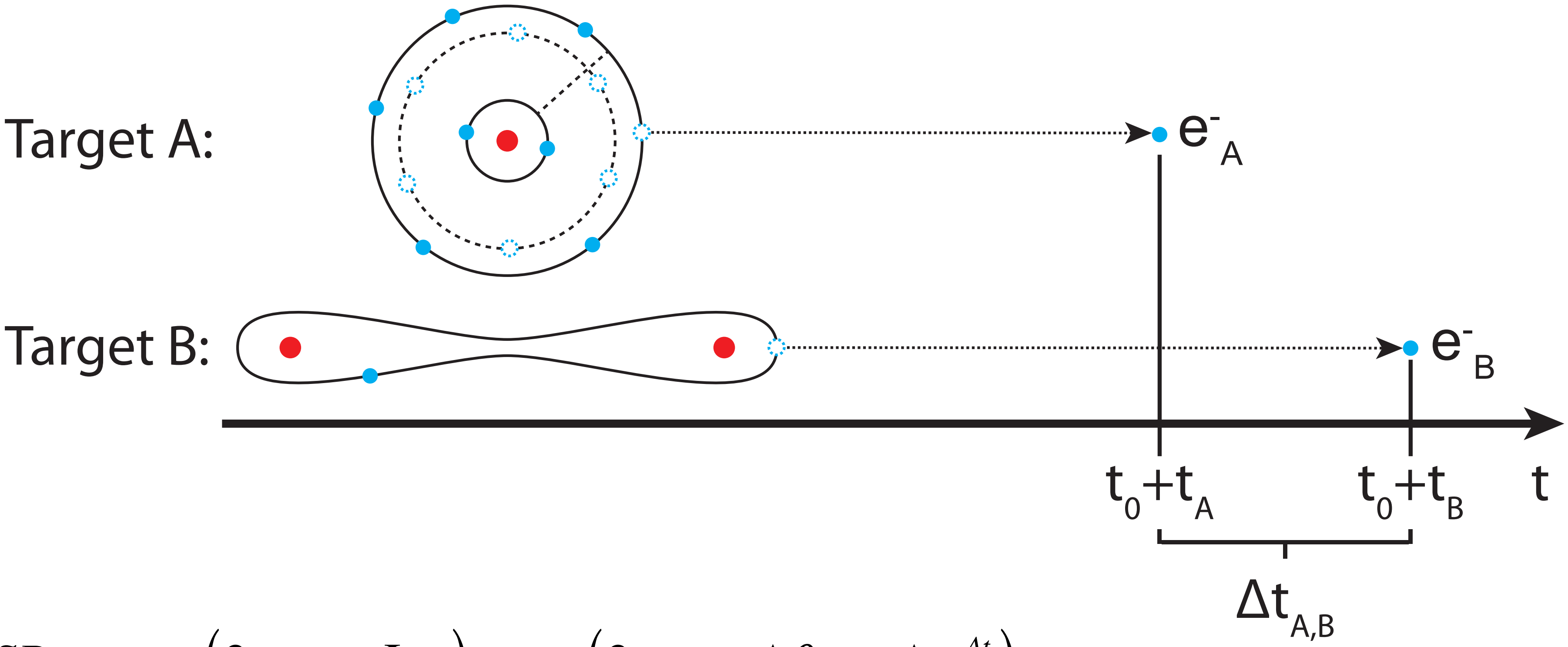
Single photon ionization delay



Single photon ionization delay



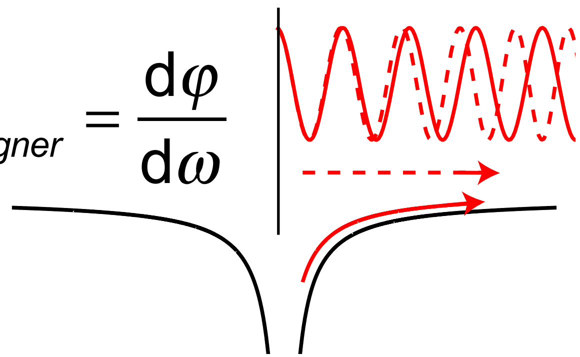
Single photon ionization delay



$$SB_{2q} \sim cos\left(2\omega_0\tau - \Phi_{2q}\right) = cos\left(2\omega_0\tau - \Delta\theta_{2q} - \Delta\varphi_{2q}^{At}\right)$$

$$\frac{\Delta\varphi_{2q}^{At}}{2\omega_0} = \tau_{2q}^{At} = \tau_{2q}^{Wig} + \tau_{2q}^{cc}$$

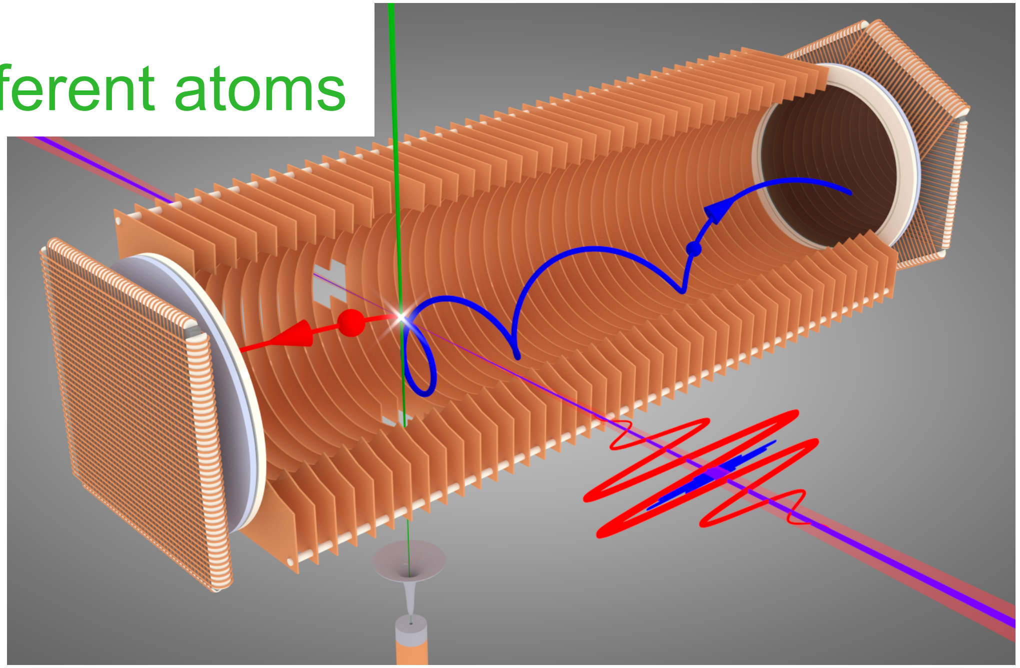
$$\tau_{Wigner} = \frac{d\varphi}{d\omega}$$



Single photon ionization delay

Reaction Microscope (COLTRIMS): R. Dörner *et al.*, Phys. Rep. **330**, 95, 2000

Gas target:
mixture of different atoms

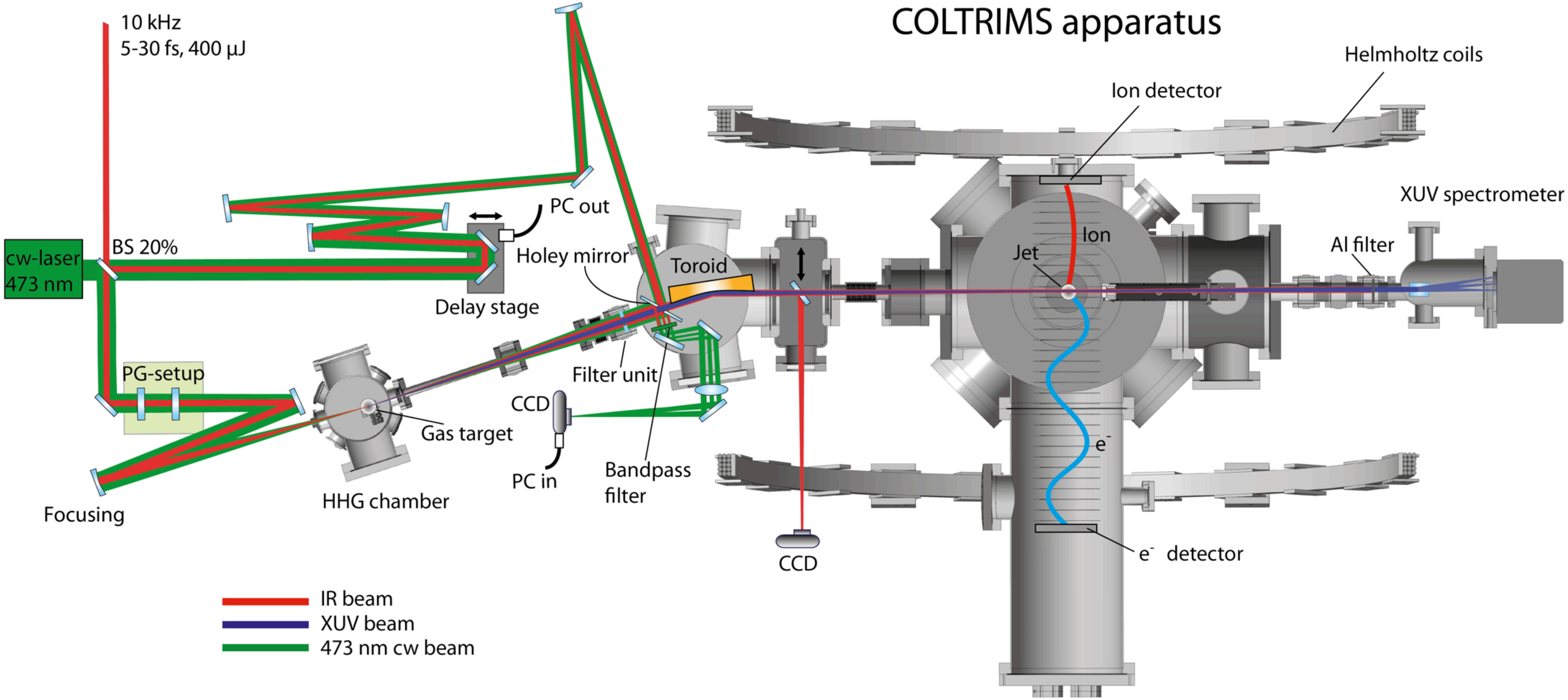


- Reconstruction of 3D momentum
- Full solid angle for electrons and ions
- Coincidence measurement of ions and electrons

M. Sabbar, S. Heuser, R. Boge, M. Lucchini, L. Gallmann, C. Cirelli, U. Keller
Phys. Rev. Lett., submitted 20. July 2014

The AttoCOLTRIMS apparatus

AttoCOLTRIMS = Attosecond beamline + COLTRIMS detector

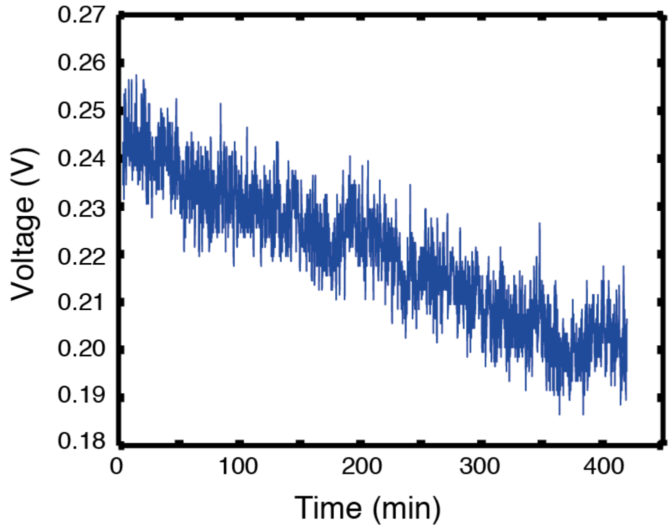


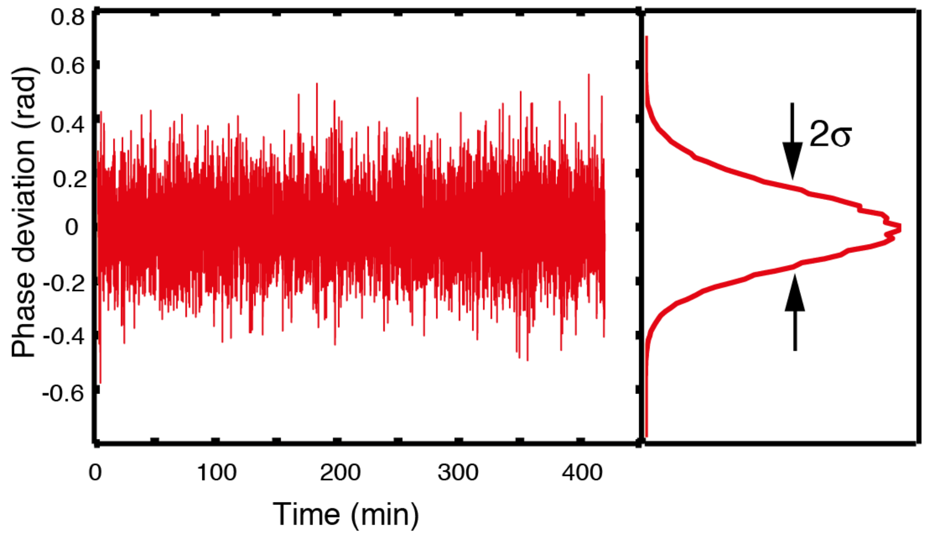
Sabbar and Heuser *et al.*, submitted to Rev. Sci. Instrum.

The AttoCOLTRIMS apparatus

Active interferometer stabilization

(for a constant delay position)

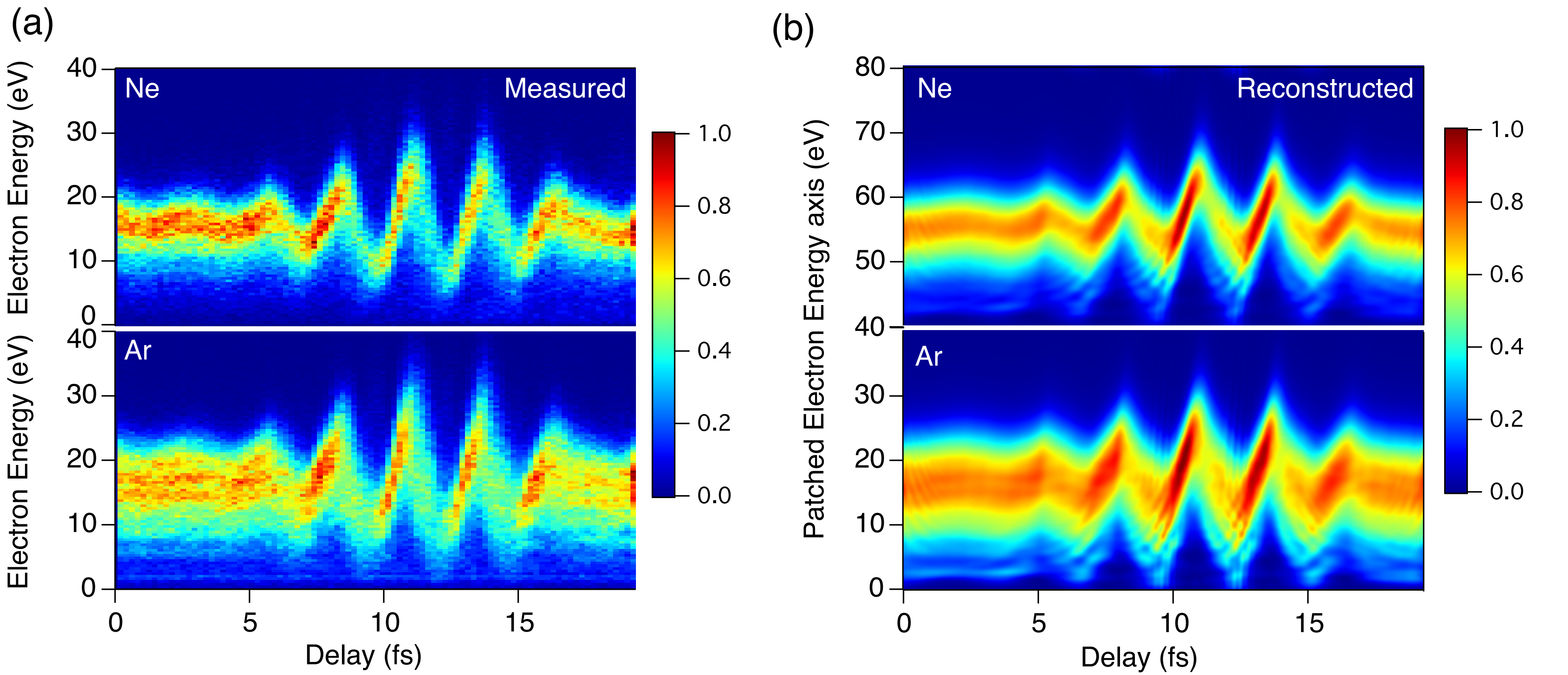




Compensates short- and long term drifts over more than 7 hours

rms < 60 as

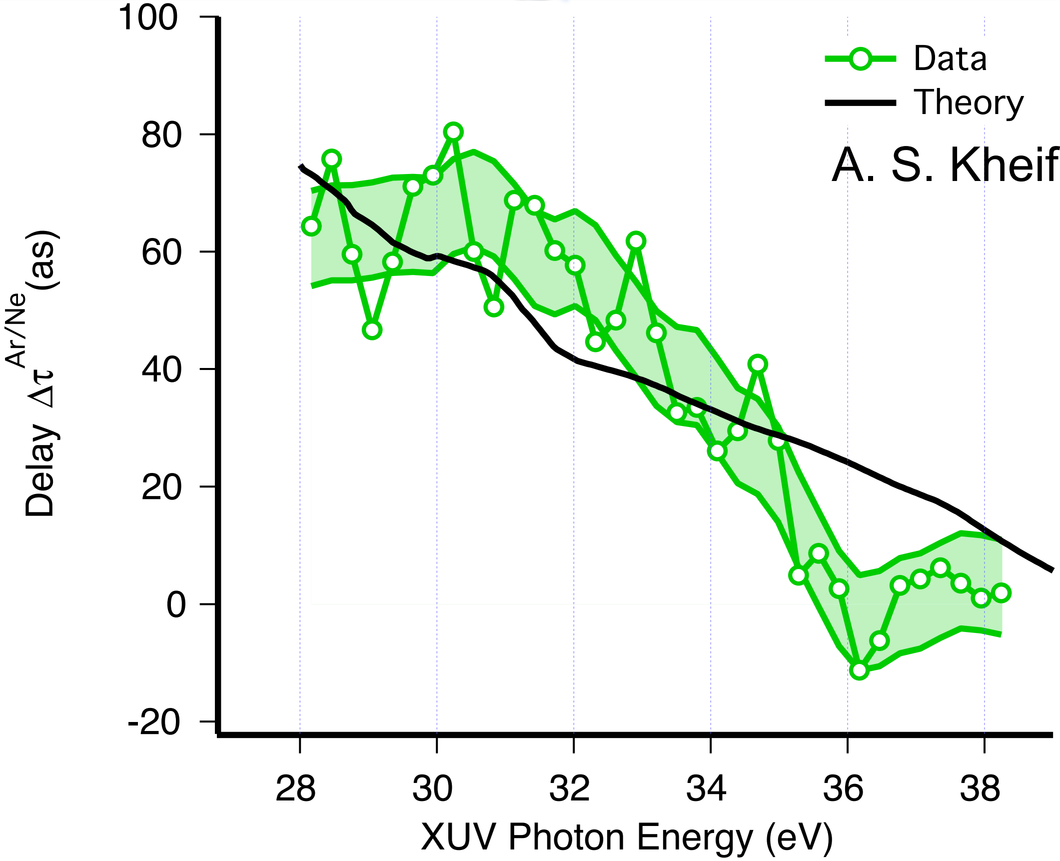
Single photon ionization delay



- Attosecond energy streaking in coincidence
- Single attosecond pulses, center energy 35 eV, 10 eV bandwidth
- Correction for attochirp and frequency dependent ionization crosssection using the FROG-CRAB algorithm

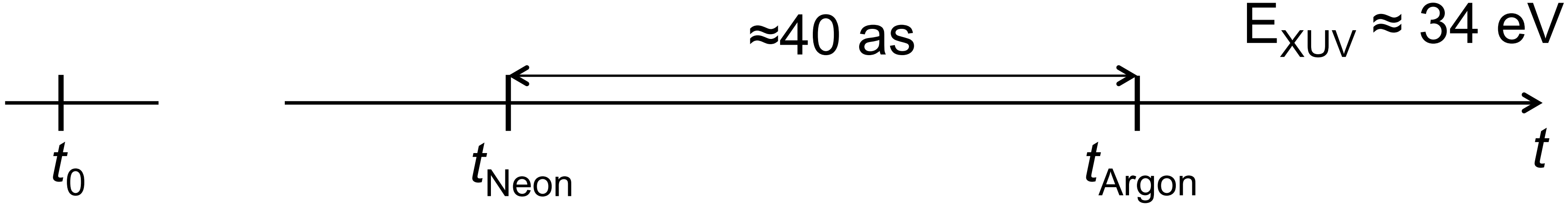
M. Sabbar, S. Heuser, R. Boge, M. Lucchini, L. Gallmann, C. Cirelli, U. Keller
Phys. Rev. Lett., submitted 20. July 2014 and arXiv:1407.6623 (2014)

Single photon ionization delay



A. S. Kheifets, Phys. Rev. A **87**, 063404 (2013)

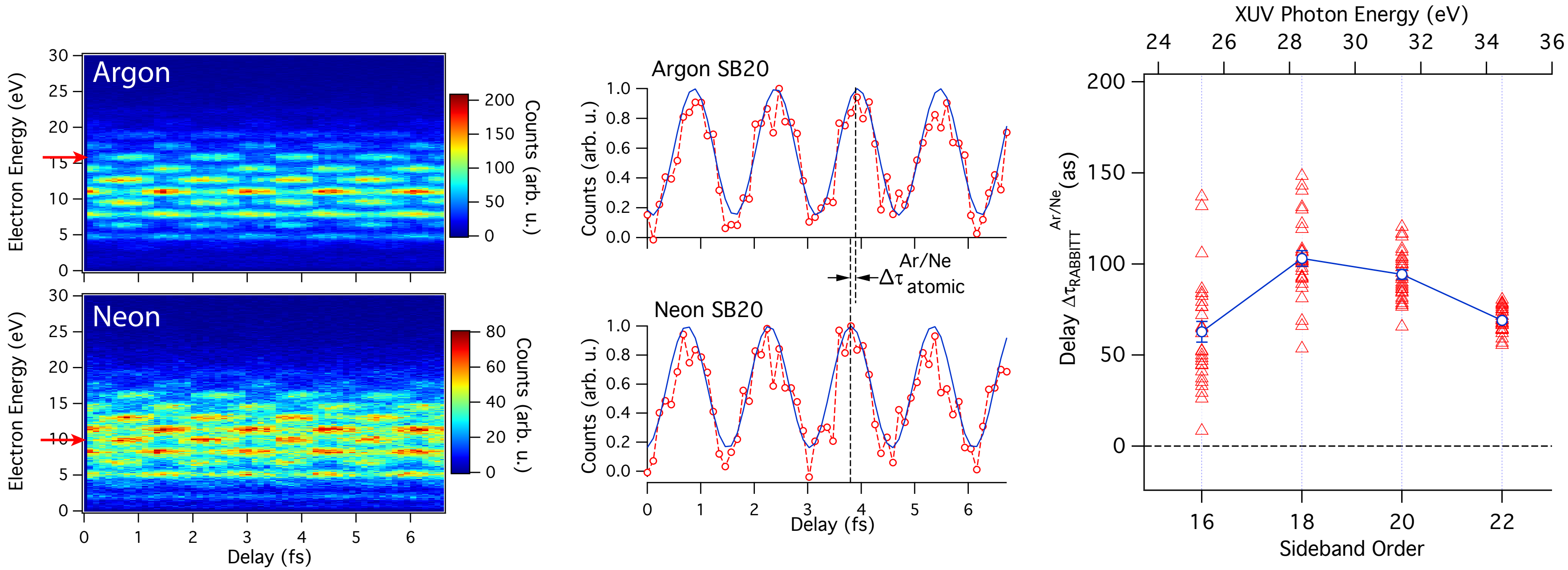
$E_{XUV} \approx 34$ eV



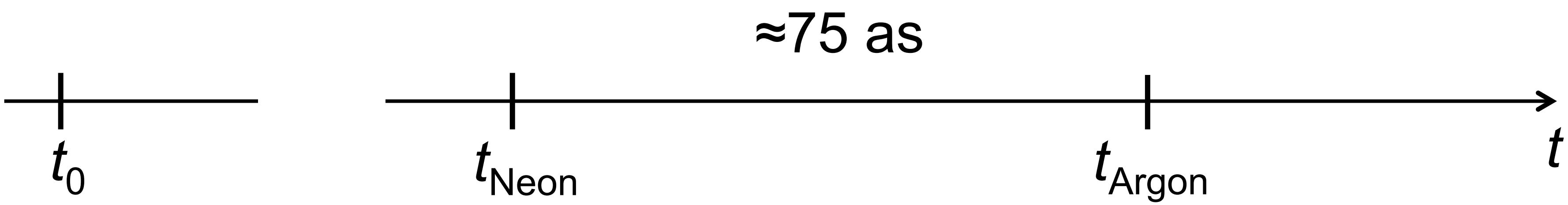
M. Sabbar, S. Heuser, R. Boge, M. Lucchini, L. Gallmann, C. Cirelli, U. Keller
Phys. Rev. Lett., submitted 20. July 2014

Coincidence RABBITT

- Simultaneous measurement with argon and neon gas mixture
- Extract relative delay between sideband pairs



Neon ionizes earlier than argon: $\Delta\tau^{Ar/Ne}_{RABBITT} \approx +75\,\text{as}$



RABBITT data from Lund

Keller visit in Lund 24. April 2014

Realized that Anne L'Huillier's group has done similar measurements with different noble gas atoms ...

Measurements of relative photoemission time delays in noble gas atoms

D. Guénot[1], D. Kroon[1], E. Balogh[2], E. W. Larsen[1], M. Kotur[1], M. Miranda[1], T. Fordell[1], P. Johnsson[1], J. Mauritsson[1], M. Gisselbrecht[1], K. Varjù[2], C. L. Arnold[1], T. Carette[3,6], A. S. Kheifets[7], E. Lindroth[5], A. L'Huillier[1] and J. M. Dahlström[3,4,5]

[1] Department of Physics, Lund University, P. O. Box 118, SE-22100 Lund, Sweden

[2] Department of Optics and Quantum Electronics, University of Szeged, Dóm tér 9, 6720, Szeged, Hungary

[3] Department of Physics, Stockholm University, AlbaNova University Center, SE-106 91 Stockholm, Sweden

[4] Center for Free-Electron Laser Science, Luruper Chaussee 149, 22761 Hamburg

[5] Max Planck Institute for the Physics of Complex Systems, Noethnitzerstr. 38, 01187 Dresden

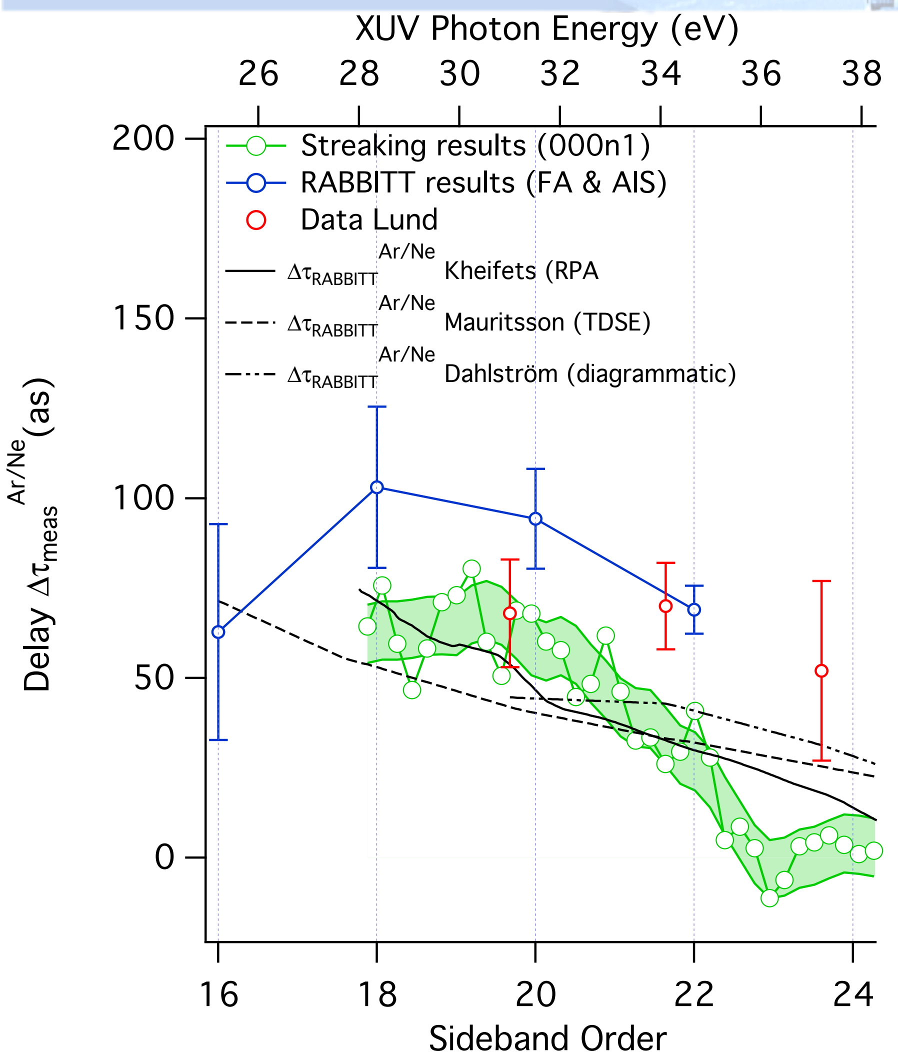
[6] Laboratoire de Chimie quantique et photophysique, CP160/09, Universite Libre de Bruxelles, B 1050 Brussels, Belgium

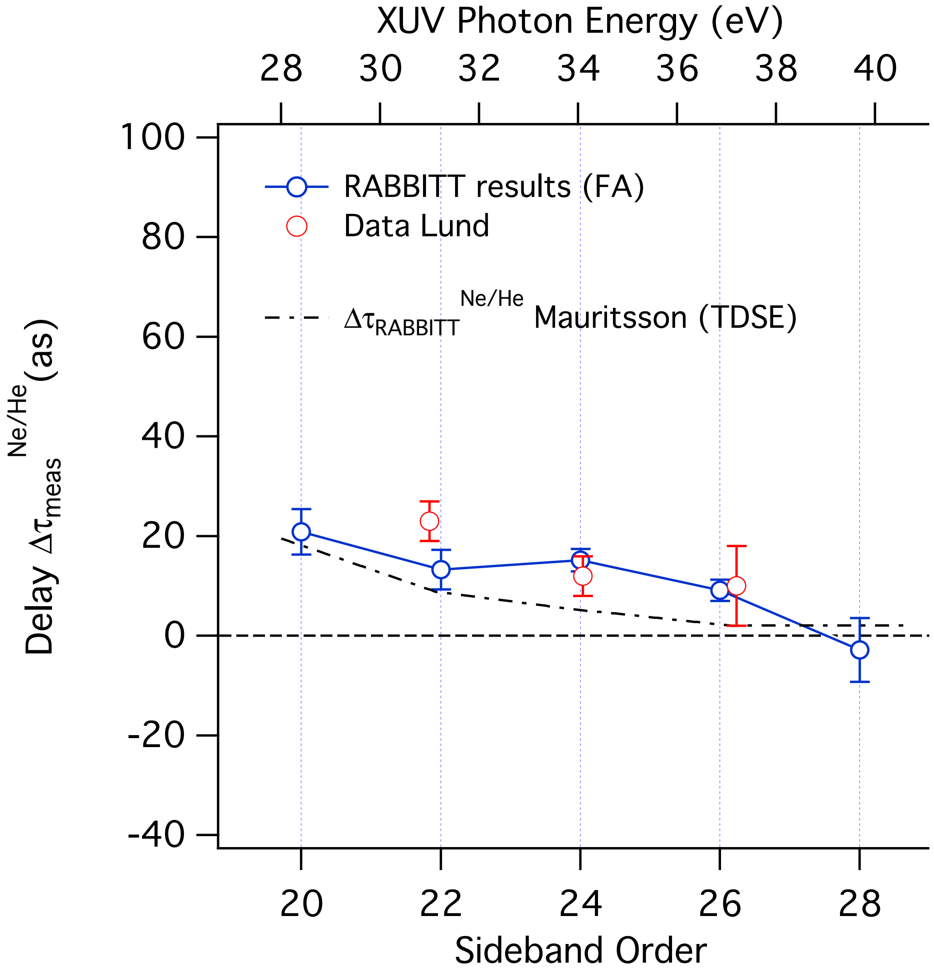
[7] Research School of Physical Sciences, The Australian National University, Canberra ACT 0200, Australia

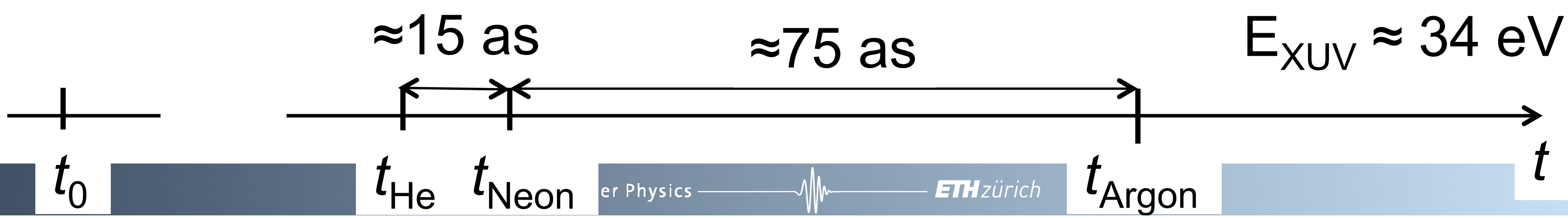
E-mail: diego.guenot@fysik.lth.se

Submitted June 2014

Single photon ionization delay







What next?

We will continue to do more measurements
(angle resolved, molecules etc.)

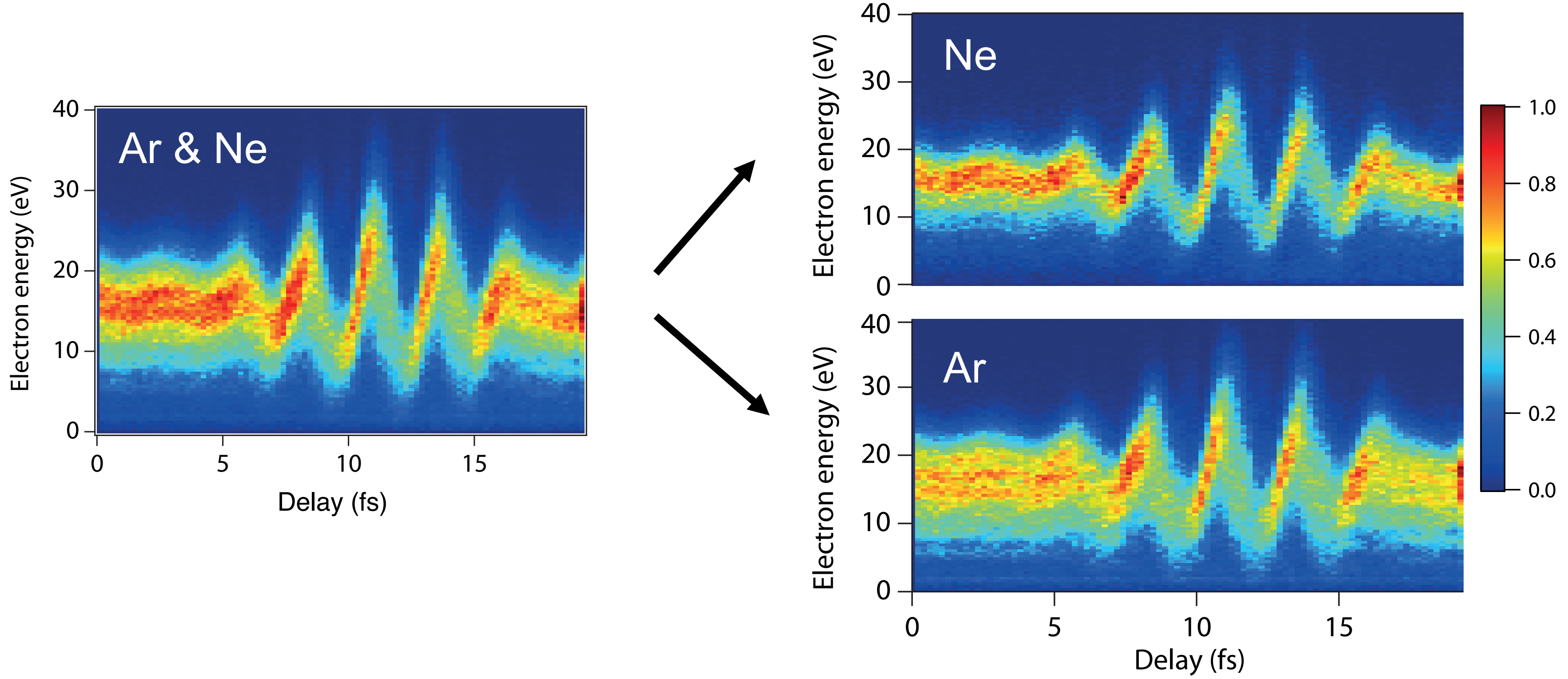
We need to sort out streaking and RABBITT differences

- theoretically they should be the same, maybe streaking IR fields too strong?
- attochirp effects?
- energy dependent absorption cross-section?
- angle dependence?

Attochirp-corrected ionization time delays

Powerful method: Extend it by coincidence streaking

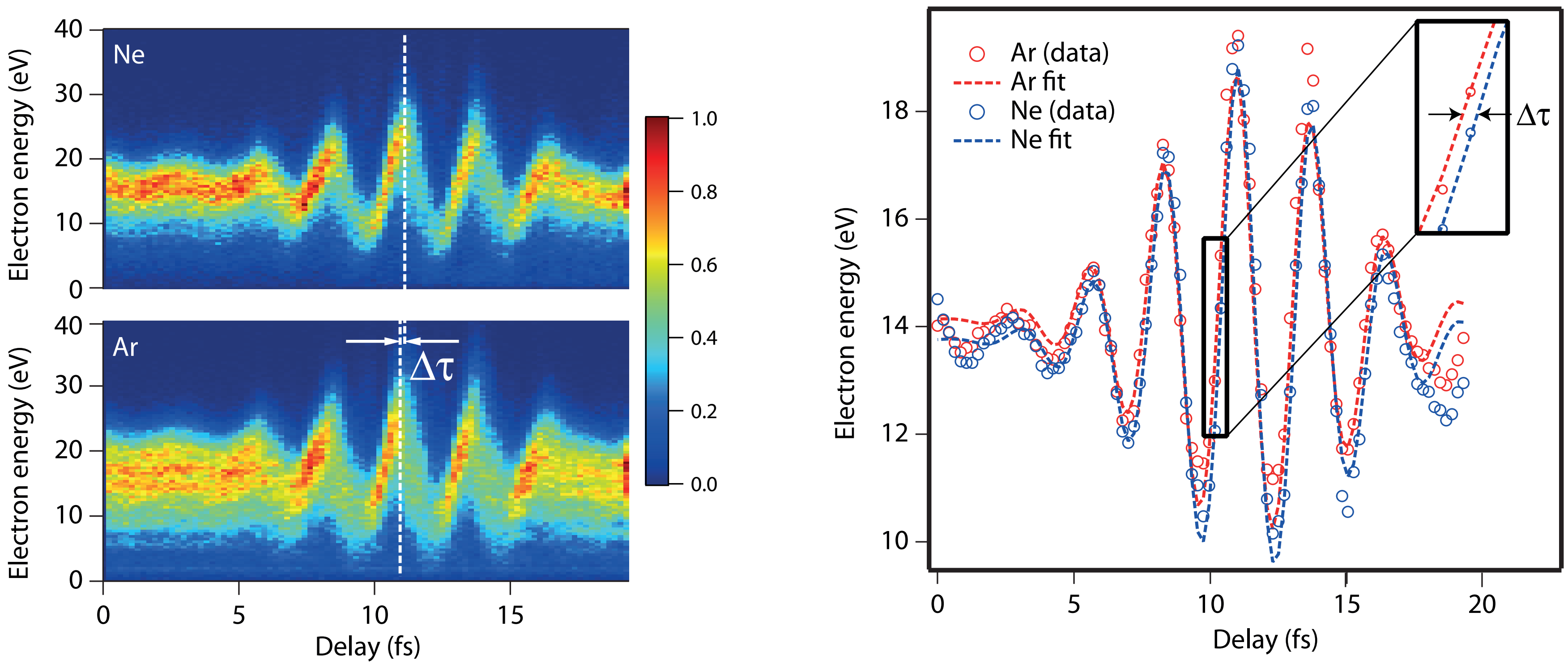
Streaking with multiple species target



M. Sabbar, S. Heuser, R. Boge, M. Lucchini, L. Gallmann, C. Cirelli, U. Keller, arXiv:1407.6623 (2014)

Attochirp-corrected ionization time delays

COM analysis of Ar/Ne data

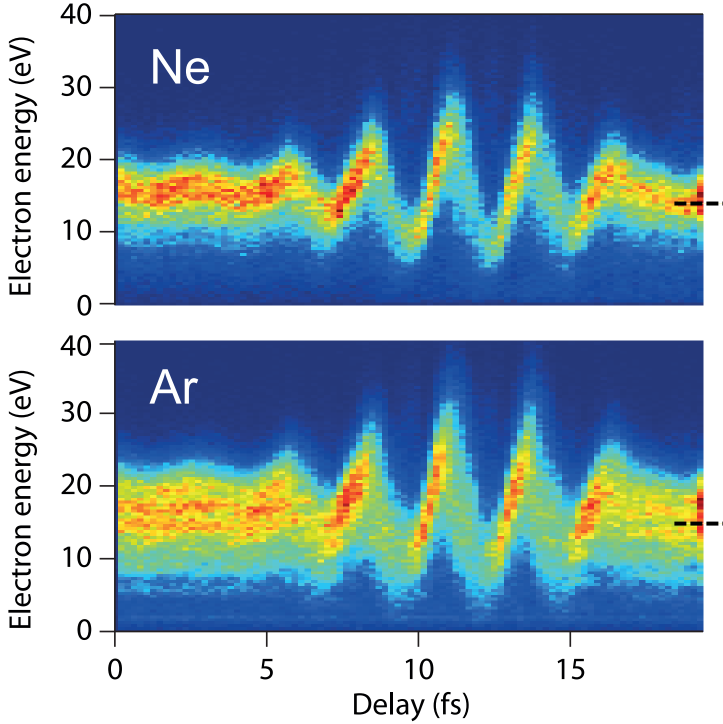


$$\Delta\tau \doteq \tau^{\mathrm{Ar}} - \tau^{\mathrm{Ne}} \approx -113\,\mathrm{as}$$

Attochirp-corrected ionization time delays

COM analysis is in general misleading

Attochirp = photons of different energy arrive at different instances in time



$E_e^{COM} \approx 15\text{ eV}$

$E_e^{COM} \approx 15\text{ eV}$

$$E_{XUV}^{COM} = E_e^{COM} + I_p$$

$$I_P^{Ne} = 21.56\text{ eV}$$

$$I_P^{Ar} = 15.76\text{ eV}$$

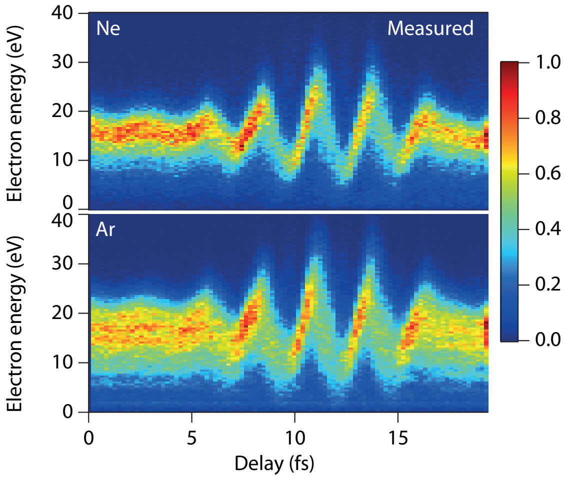
$$\left(E_{XUV}^{COM}\right)_{Ar} \neq \left(E_{XUV}^{COM}\right)_{Ne}$$

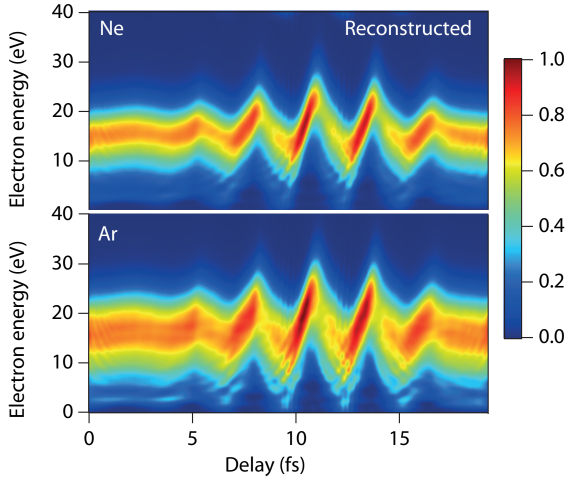
Attochirp-corrected ionization time delays

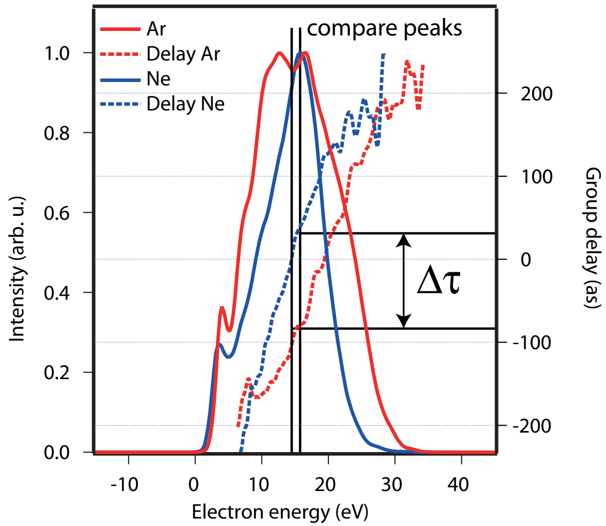
FROG-CRAB analysis

(using 33 measured traces)

Comparing electron peaks





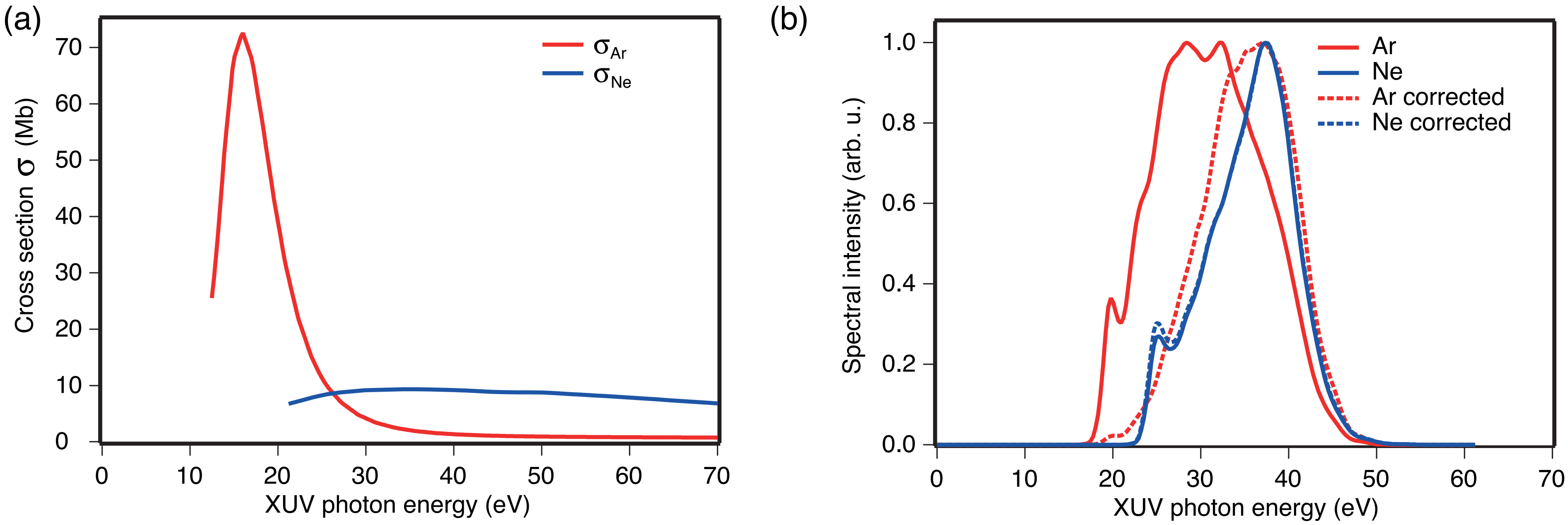


Comparing peaks: $\Delta\tau \doteq \tau^{\mathrm{Ar}} - \tau^{\mathrm{Ne}} \approx -117\ \mathrm{as}$

(COM analysis: -113 as)

Energy dependent cross-sections

Cross-sections of Ar & Ne

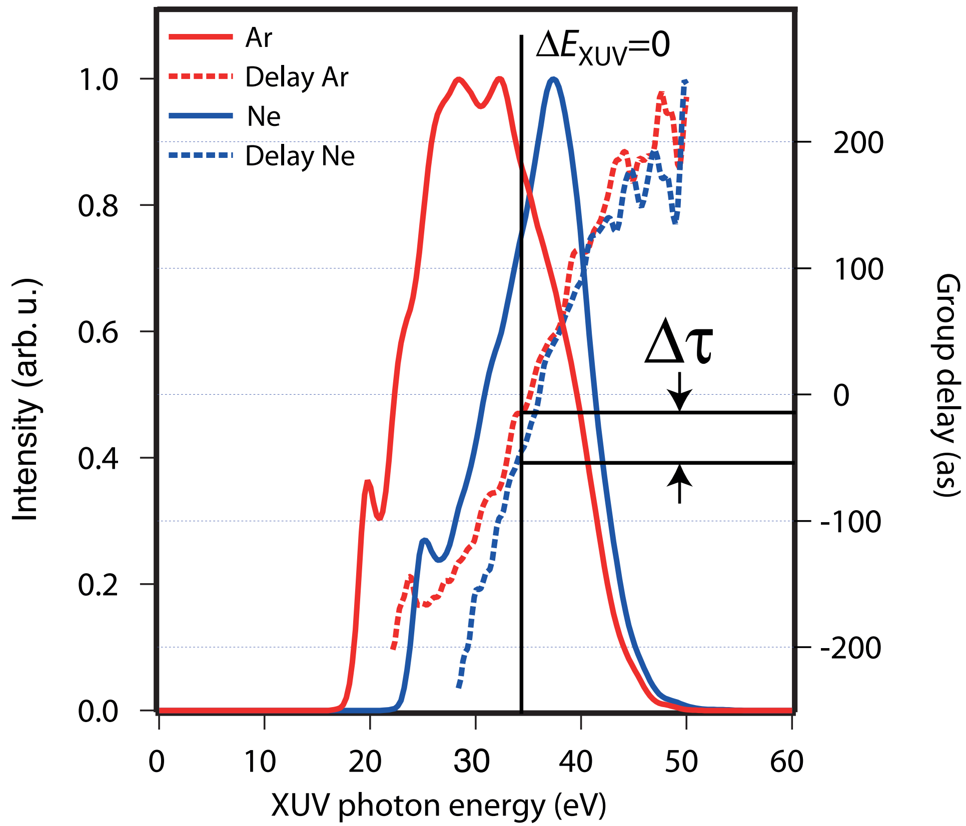


Attochirp-corrected ionization time delays

FROG-CRAB analysis

(using 33 measured traces)

Correction: XUV photon axis

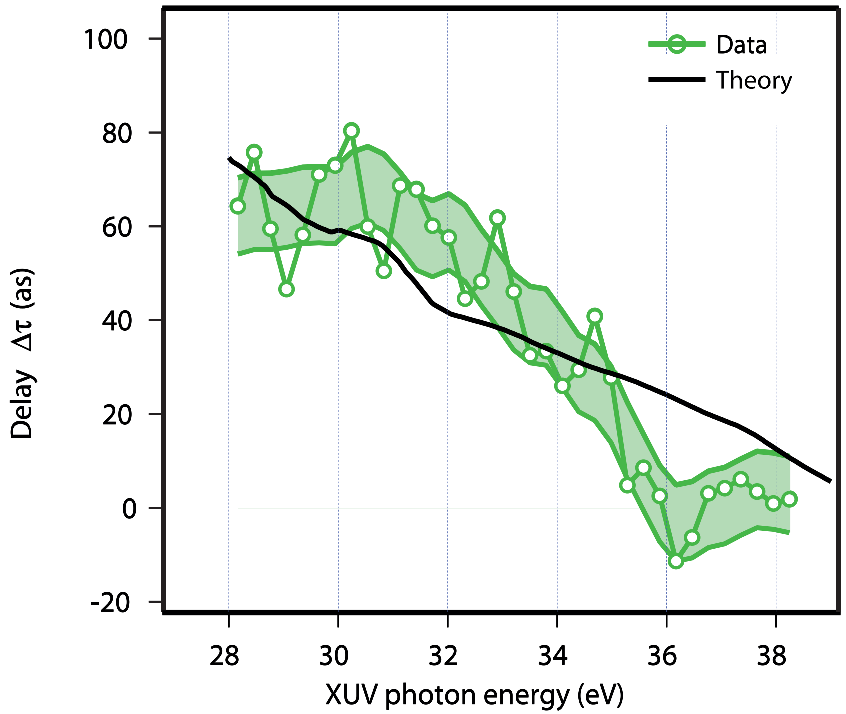


Attochirp-corrected ionization time delays

FROG-CRAB analysis

(using 33 measured traces)

Sabbar *et al.*, arXiv:1407.6623 (2014)



Result agrees with theory:

$$\tau_S = \underbrace{\tau_W}_{\text{Wigner}} + \underbrace{\tau_{CLC}}_{\text{taken from perturbation theory}^2}$$

τ_W: calculated with the random-phase approximation[1]

τ_{CLC}: Can be taken from perturbation theory[3]

[1] A. S. Kheifets, Phys. Rev. A **87**, 063404 (2013)

[2] J. M. Dahlström *et al.*, Phys. Rev. A **86**, 061402(R) (2012),

[3] R. Pazourek *et al.*, Faraday Discussions **163**, 353 (2013)

What next?

We will continue to do more measurements
(angle resolved, molecules etc.)

We need to sort out streaking and RABBITT differences

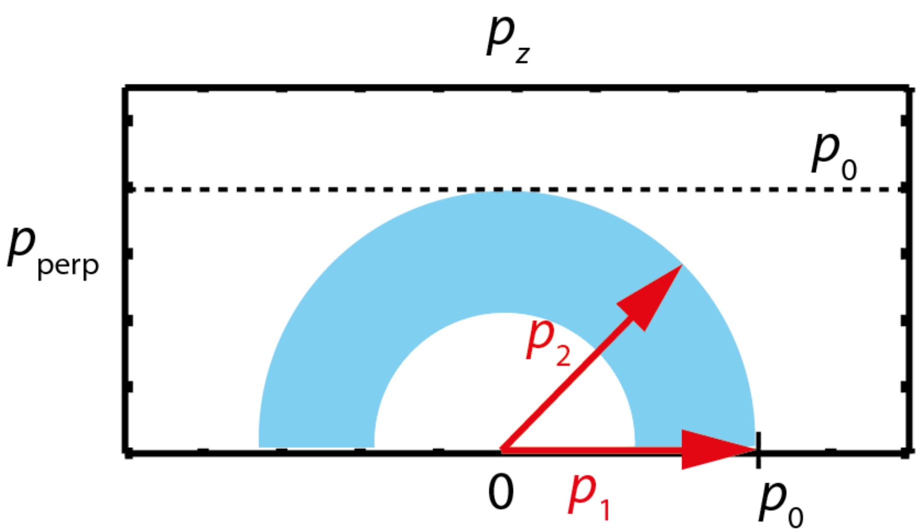
- theoretically they should be the same, maybe streaking IR fields too strong?
- attochirp effects?
- energy dependent absorption cross-section?
- angle dependence?

Angle dependence

Attosecond streaking with a COLTRIMS detector

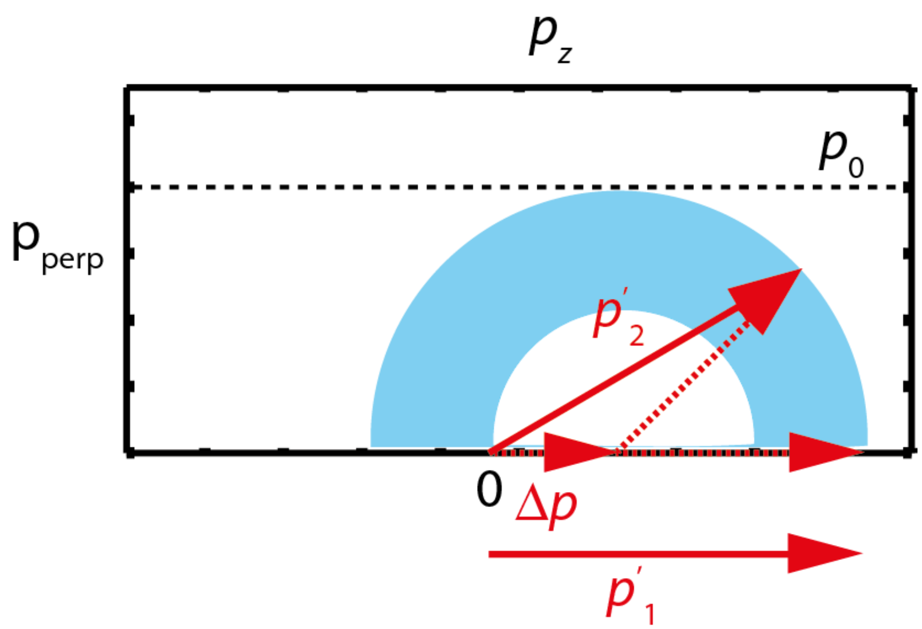
(Angular discrepancy)

no streaking field



$$E = \boldsymbol{p}_1^2 / 2m_e = \boldsymbol{p}_2^2 / 2m_e = \boldsymbol{p}_0^2 / 2m_e$$

streaking field shifts the distribution



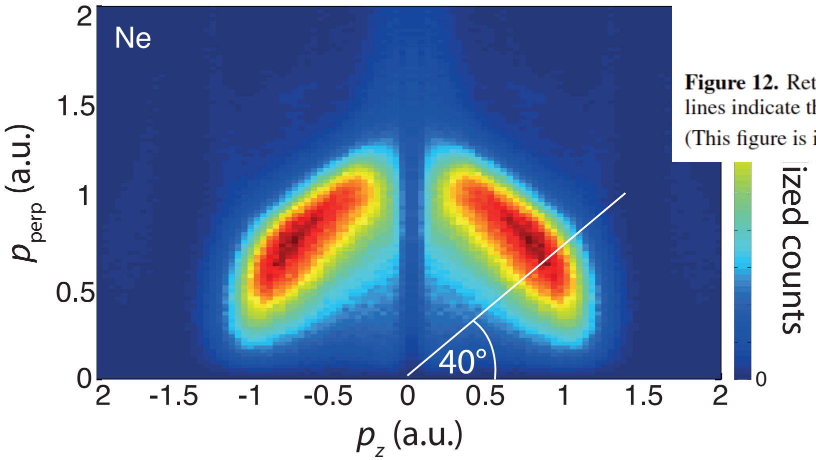
$$\boldsymbol{p}_1^{'2} / 2m_e \neq \boldsymbol{p}_2^{'2} / 2m_e$$

Angle dependence

Attosecond streaking with a COLTRIMS detector

(Restrict angle)

Used a cone filter with 40° opening for streaking measurement.



XUV polarization

IR polarization

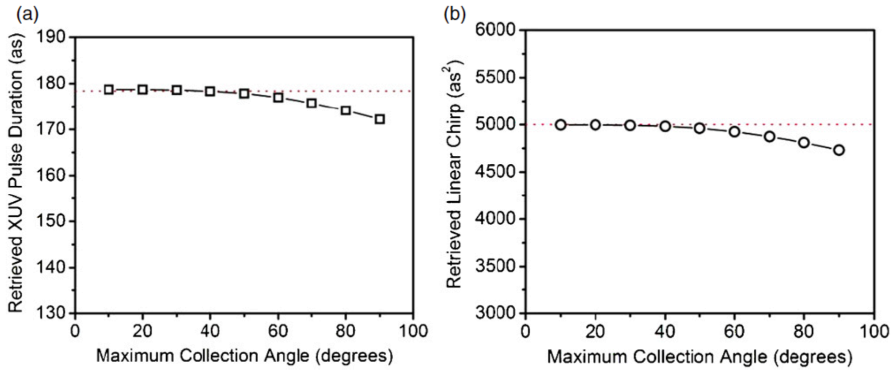


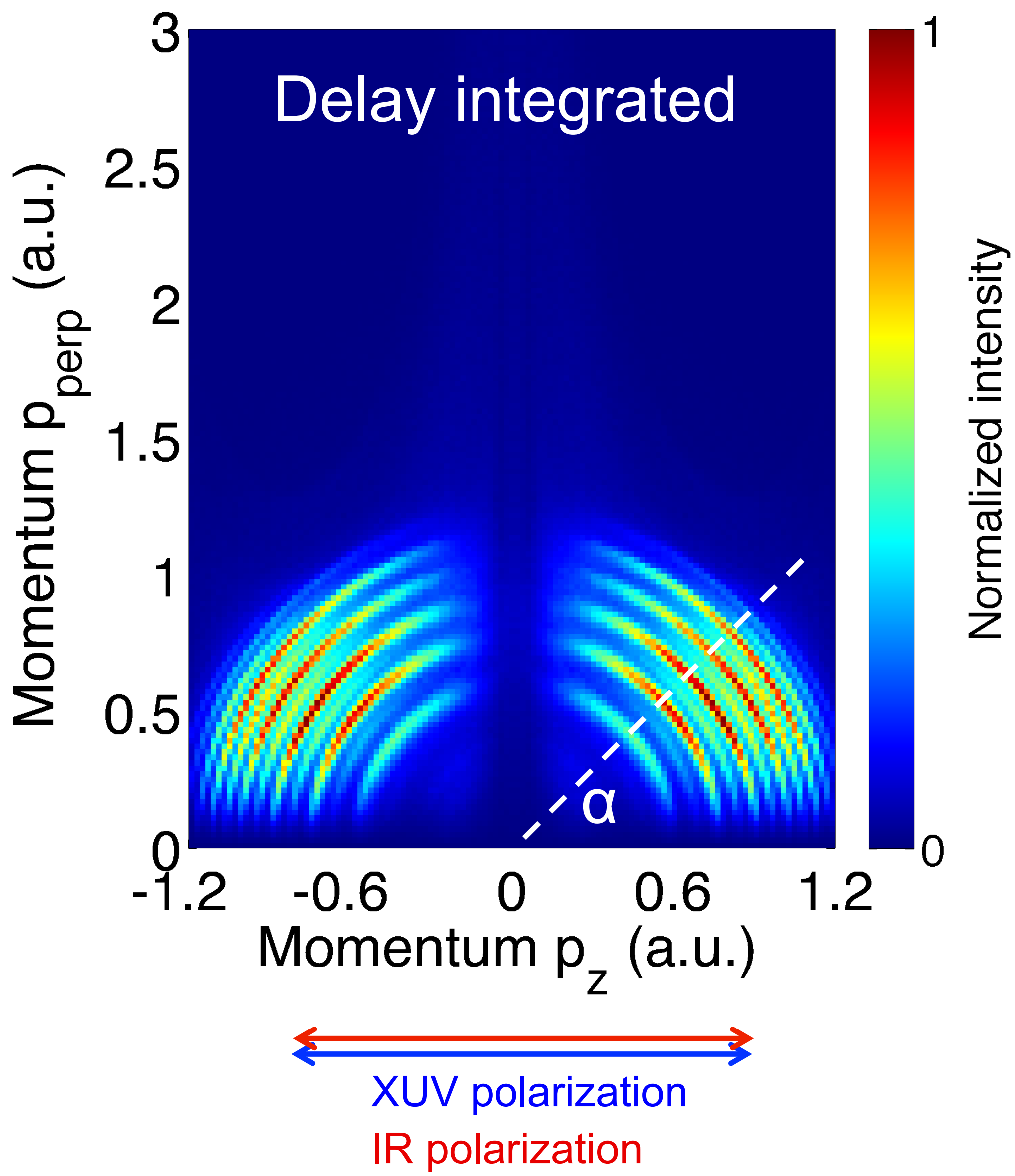
Figure 12. Retrieved (a) XUV pulse duration and (b) XUV linear chirp as a function of the maximum collection angle θ_0. The red dashed lines indicate the actual values, whereas the black open shapes represent the retrieved values.
(This figure is in colour only in the electronic version)

'As shown in figures 12 (a) and (b), when the maximum collection angle is below 40° , the XUV pulse duration and chirp can be retrieved exactly.'

H. Wang *et al.*, J. Phys. B-At. Mol. Opt. Phys. **42** (2009)

Angular dependent delays

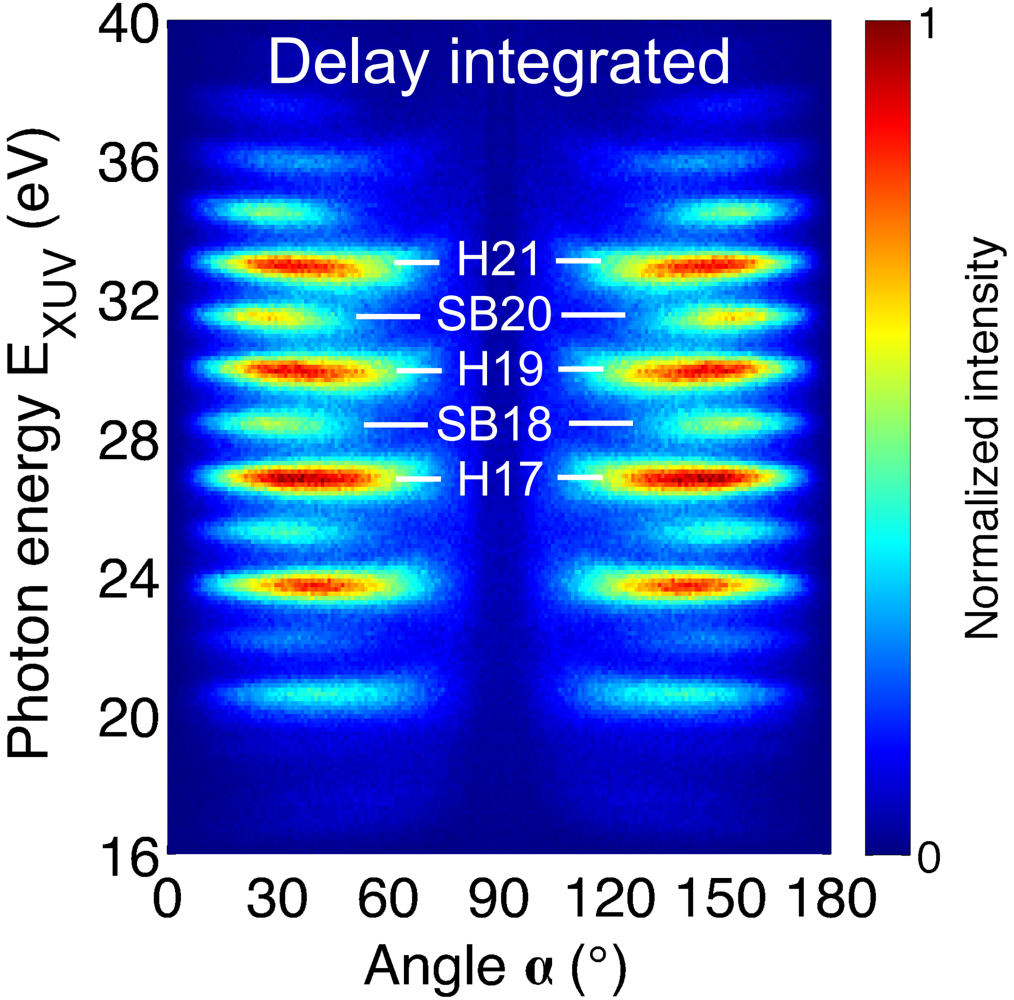
- p_z is the momentum parallel to the XUV/IR polarization direction
- p_{perp} is the absolute momentum perpendicular to the XUV/IR polarization direction



Angular dependent delays

- $E_e = (p_{perp}^2 + p_z^2)/2m_e$
- $E_{XUV} = E_e + I_p$
- Emission angle α is given by the electron emission direction relative to the laser polarization direction:

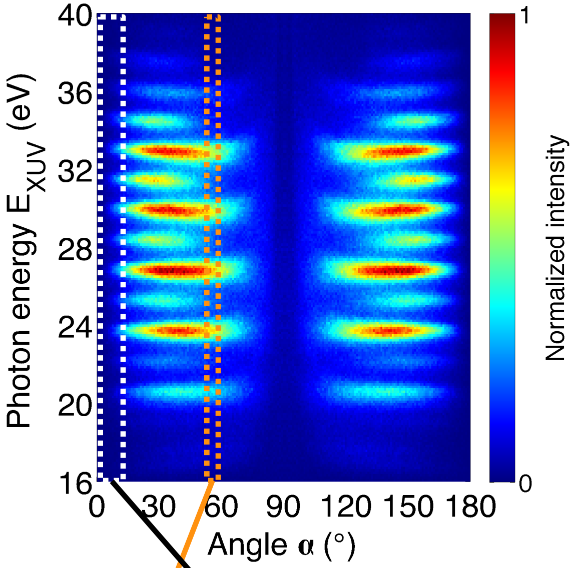
$$\alpha = \text{atan}\,(p_{perp} / p_z)$$



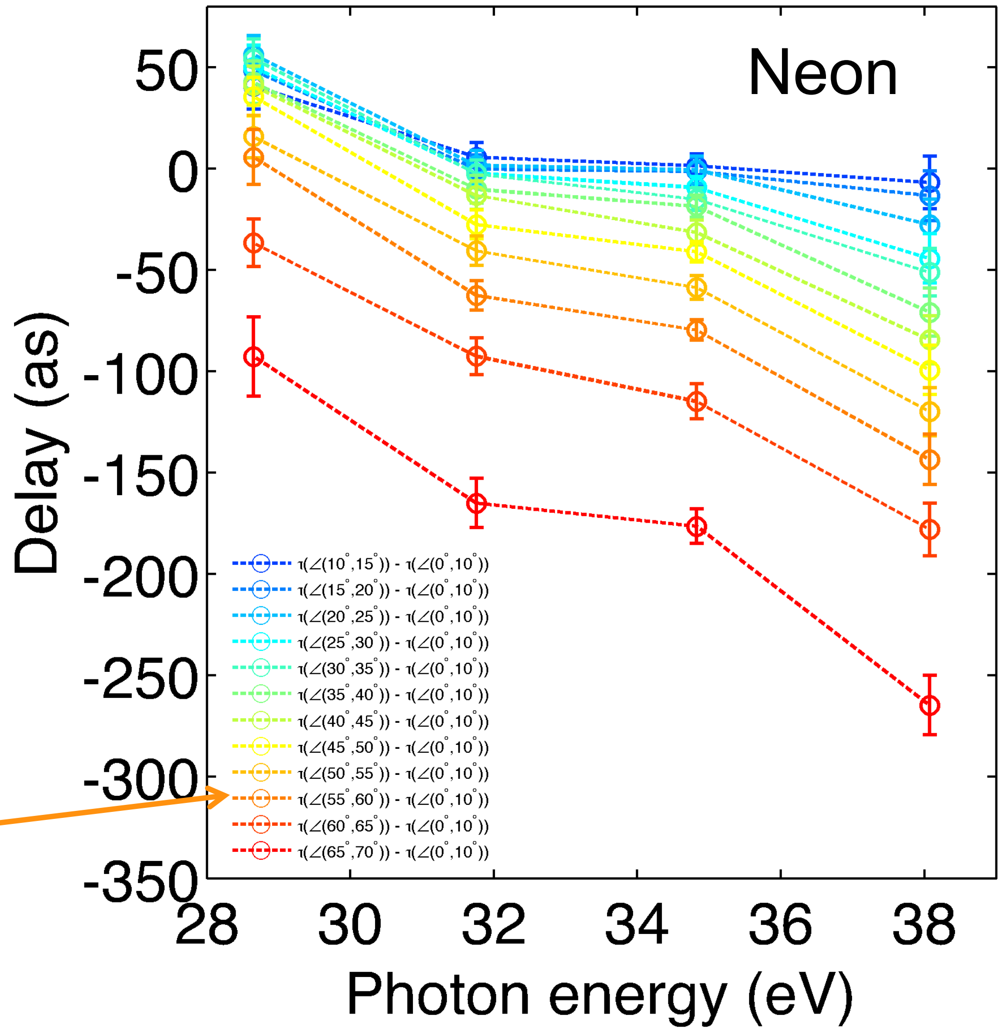
Is the phase of the SB oscillation dependent on the electron emission angle α?

Angular dependent delays: Ne results

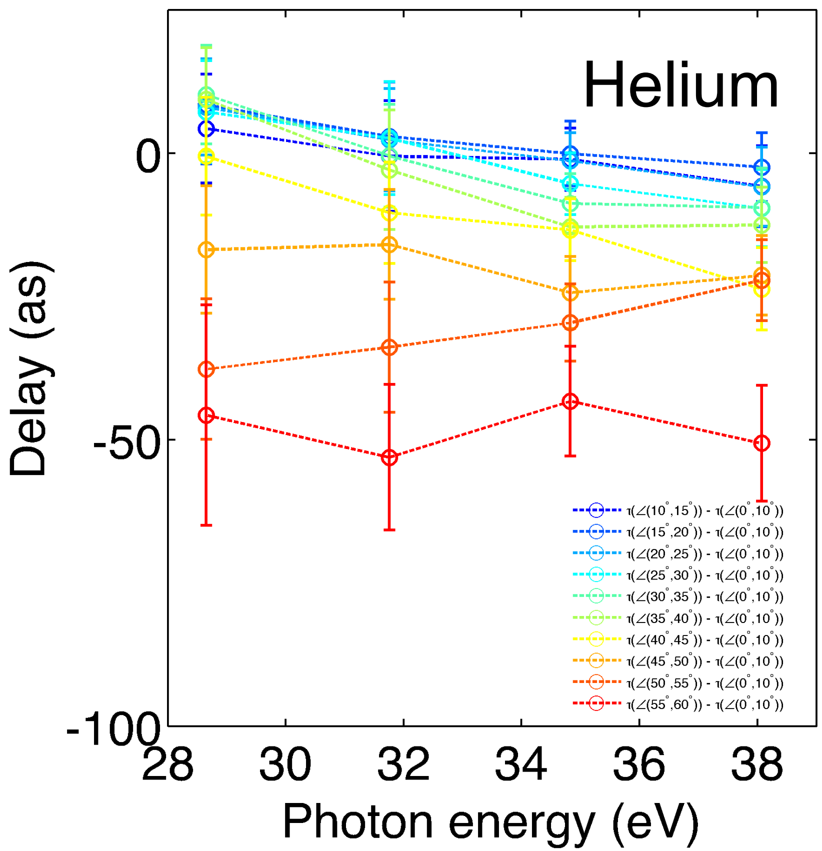
Angular dependent delays referenced to the delay determined along XUV/IR polarization direction (0 < α < 10°)

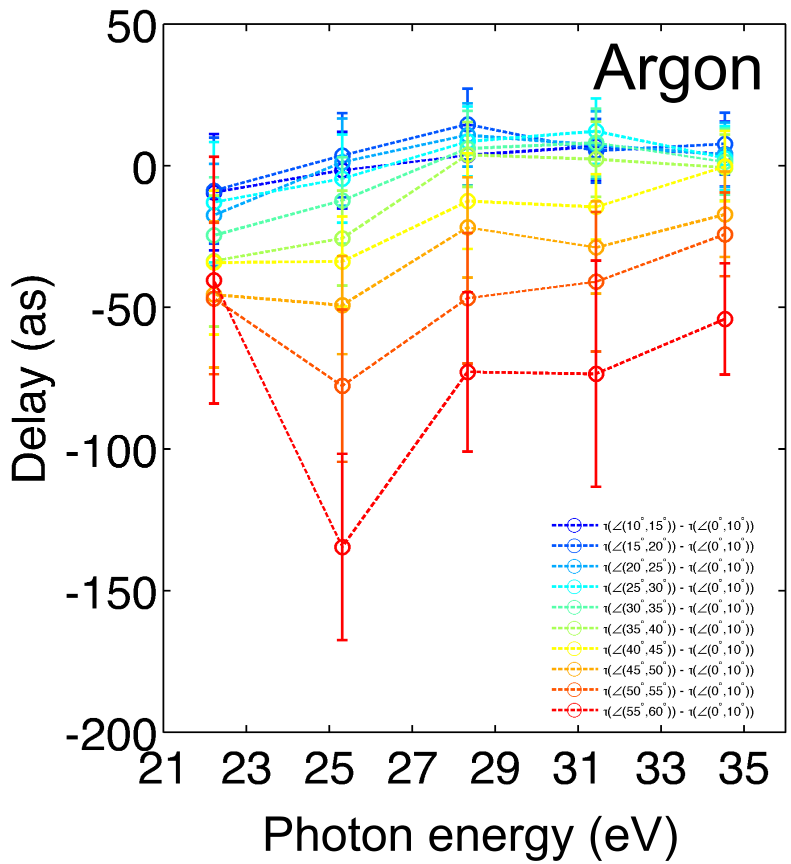


$$\tau^{Ne}_{\alpha\in(55^\circ,60^\circ)} - \tau^{Ne}_{\alpha\in(0^\circ,10^\circ)} = \Delta\tau^{Ne}_{atomic}$$



Angular dependent delays: He and Ar results

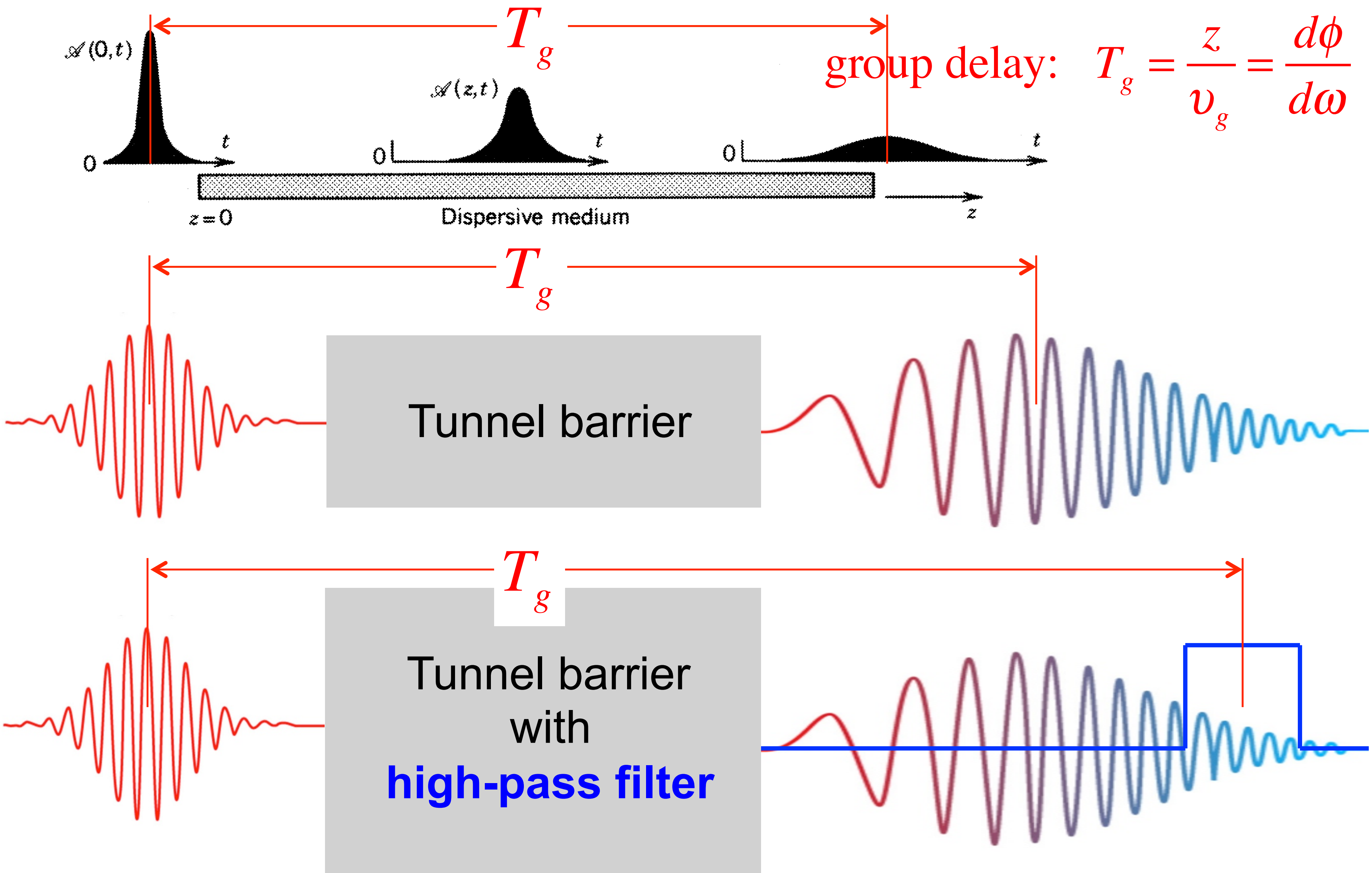




- First experiments resolving angle-dependent photoionization time delays.
- Preliminary data. Ongoing theoretical collaboration with J. Bürgdorfer group in Vienna (S. Nagele) to explain the trend observed in the experiments.

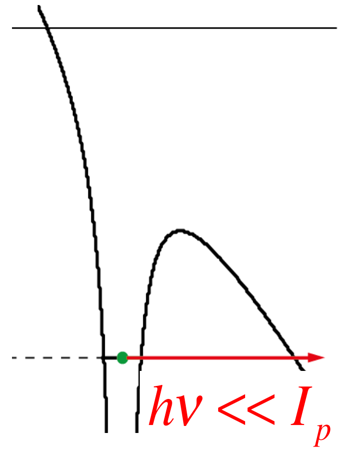
Wigner time is problematic with tunneling

group delay: $T_g = \frac{z}{\upsilon_g} = \frac{d\phi}{d\omega}$

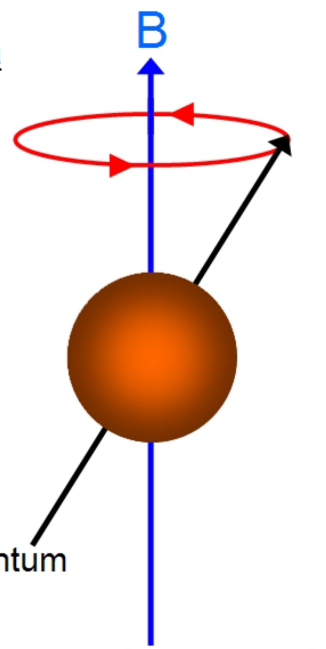


Attoclock measurement: consistent with Larmor time

tunnel ionization

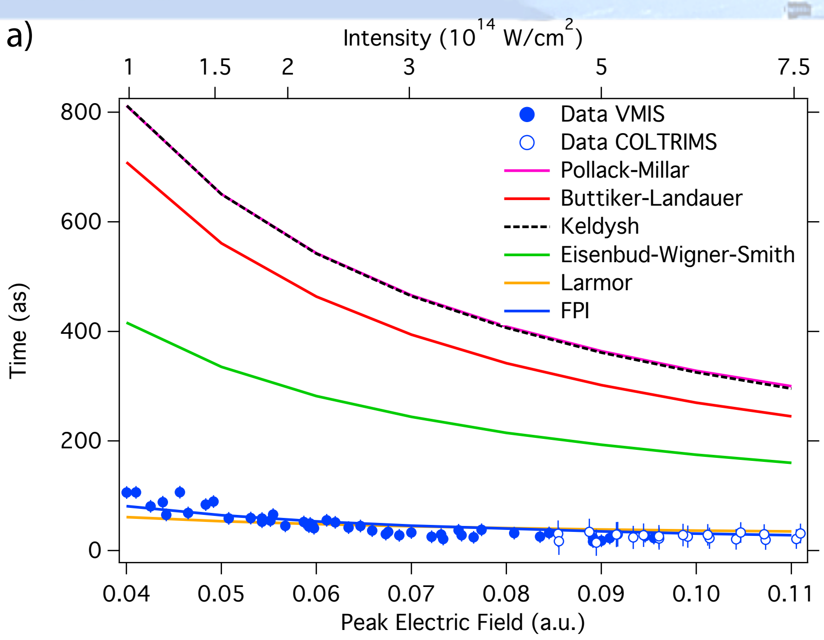


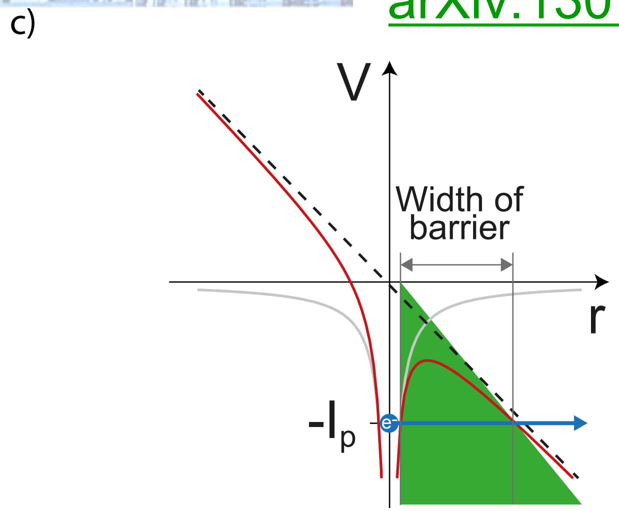
Tunnel barrier
with
an uniform
magnetic field *B*



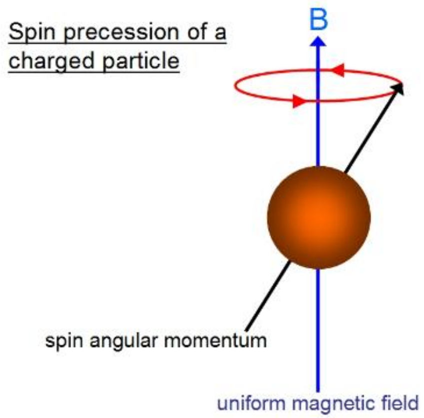
Attoclock results from Helium

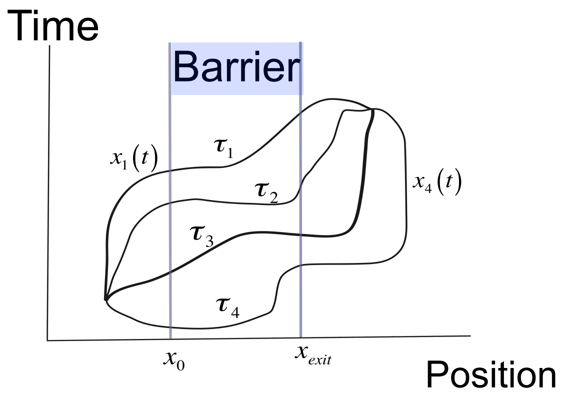
A. S. Landsman et al., arXiv:1301.2766





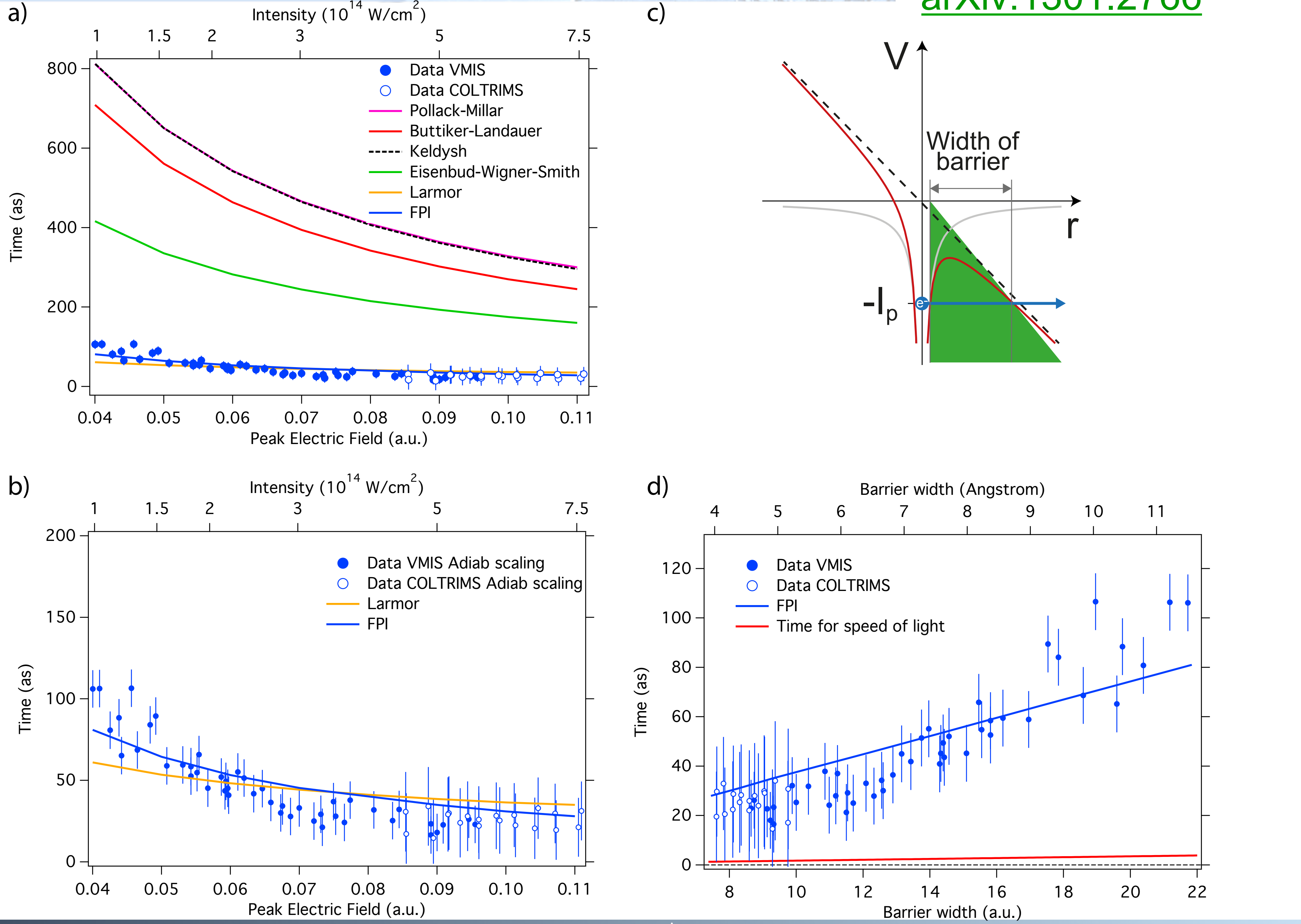
Best agreement with Larmor tunneling time
and Feynman path integral calculation (Alexandra Landsman)





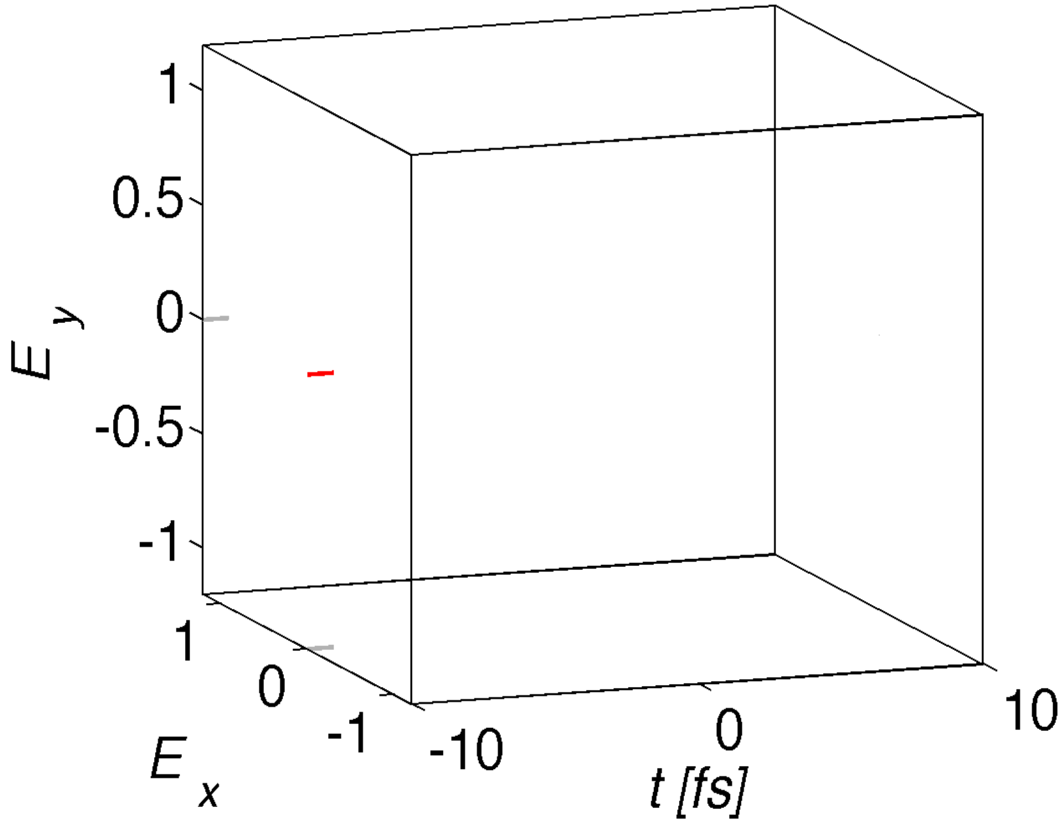
Attoclock results from Helium

A. S. Landsman et al., arXiv:1301.2766



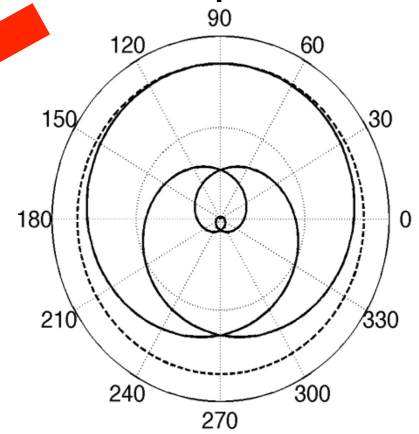
Circular polarization of a two-cycle infrared pulse

Rotation in space instead of oscillation in time

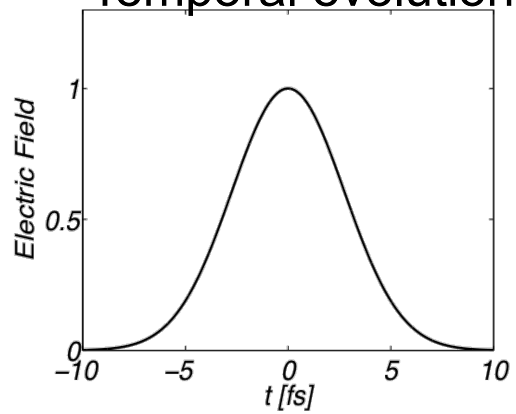


Projection onto

polarization plane



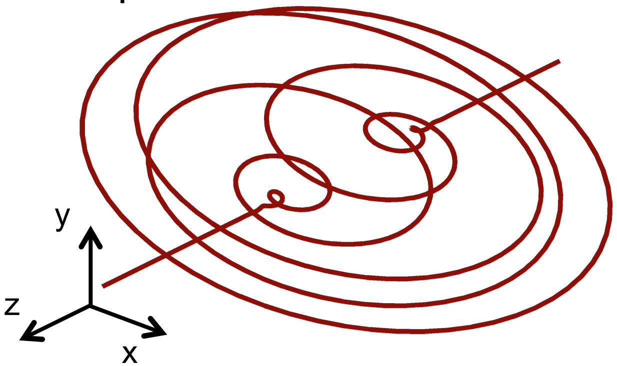
Temporal evolution:

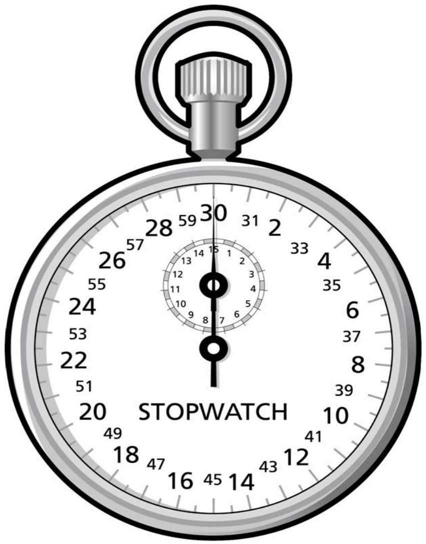


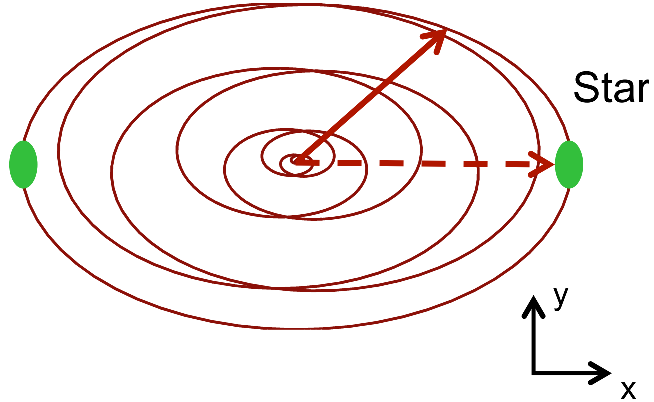
Attoclock: maps time to angle

Rotating infrared electric field of laser pulse streaks electron, mapping angle to moment of ionization:

Laser pulse







For λ = 800 nm:
1° equal to 7.5 as

Time zero? — Is **self referenced** or given by a additional reference such as an attosecond pulse

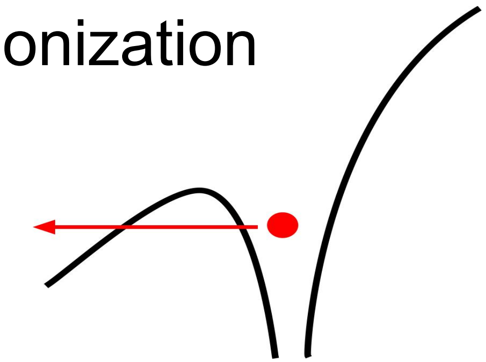
What do we measure? — The **final electron momentum** after the short pulse

What do we want to know? — **Tunneling delay time**

Attoclock: angular streaking

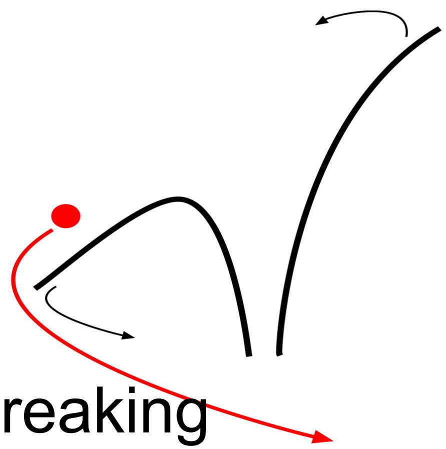
2-step model:

ionization

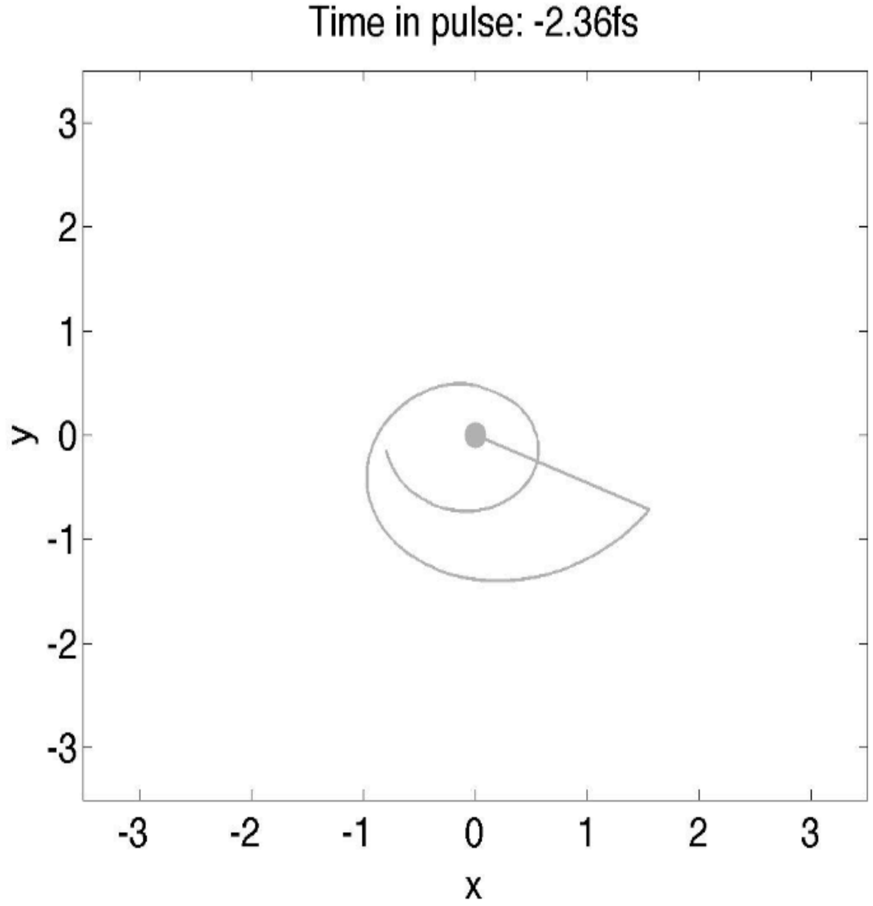


$t_{0,ion}$

time electron exits the tunnel



streaking



$$m_e \frac{d^2 x}{dt^2} = q\vec{E}_L$$

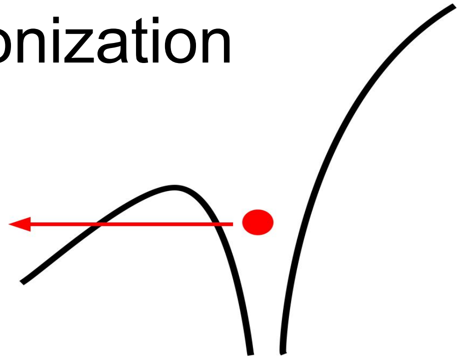
Assuming: electron liberated at maximum electric field

- laser pulse short enough that electron is not moving out of laser focus
- classically calculated trajectory in the electric field of the pulse

Attoclock: angular streaking

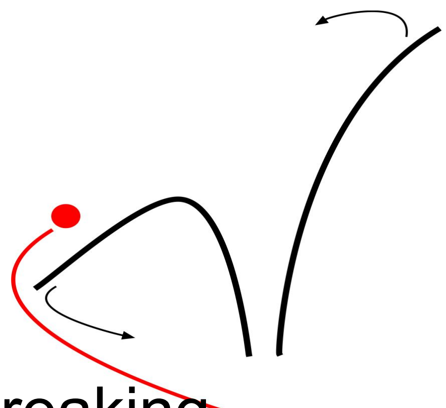
2-step model:

ionization



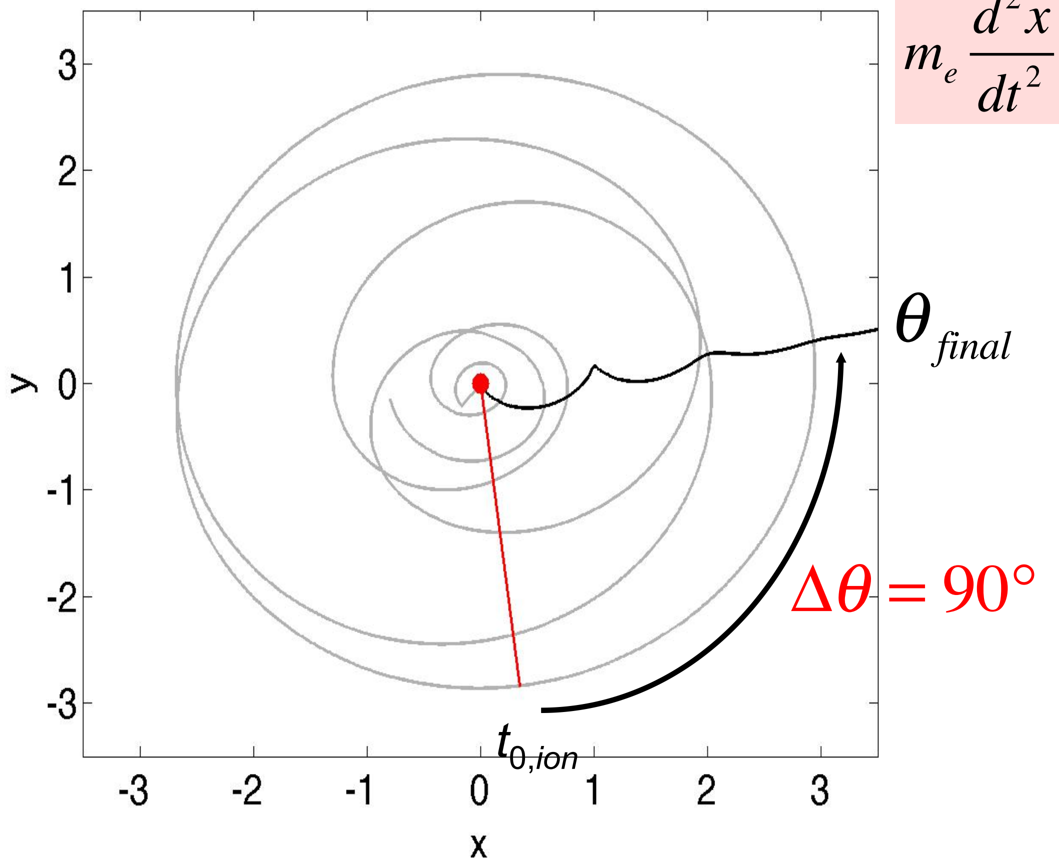
$t_{0,ion}$

time electron exits the tunnel



streaking

$$m_e \frac{d^2 x}{dt^2} = q\vec{E}_L$$



Assuming: electron liberated at maximum electric field

- laser pulse short enough that electron is not moving out of laser focus
- classically calculated trajectory in the electric field of the pulse

Tunnel geometry of Coulomb potential and laser field

Effective potential – parabolic coordinates

$$V = -\frac{1}{z} + zE_L$$

Parabolic coordinates

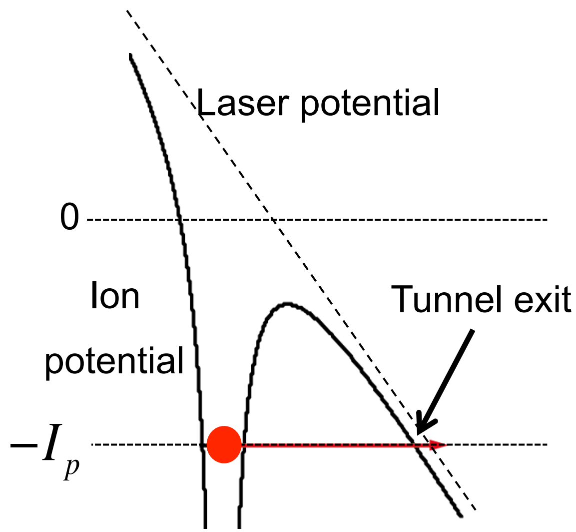
$$\xi = r + z \qquad \eta = r - z \qquad \phi = \arctan\left(\frac{y}{x}\right)$$

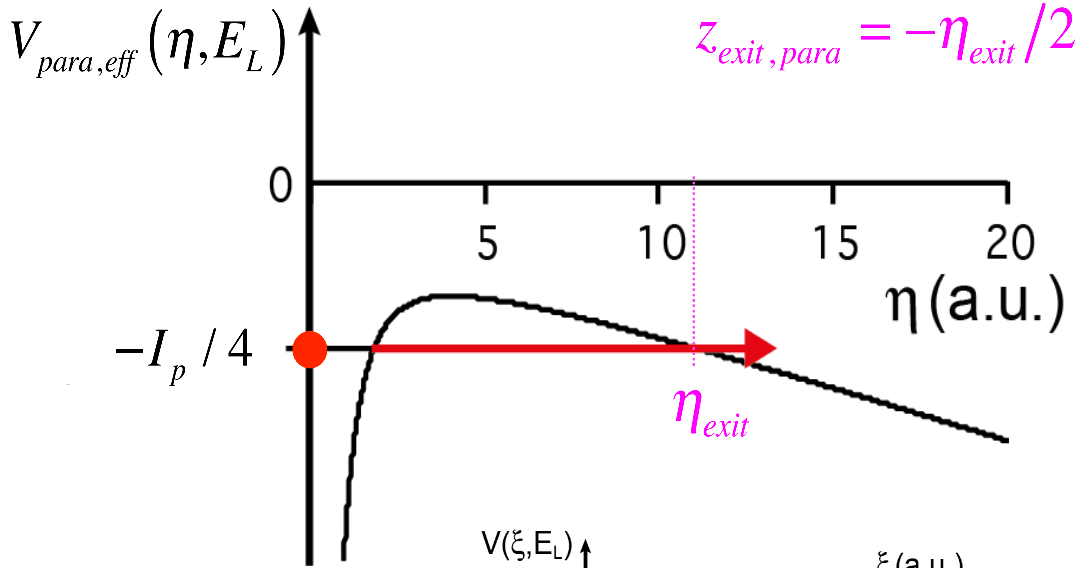
$$V_{para} = -\frac{2}{\xi+\eta} + \frac{1}{2}(\xi-\eta)E_L$$

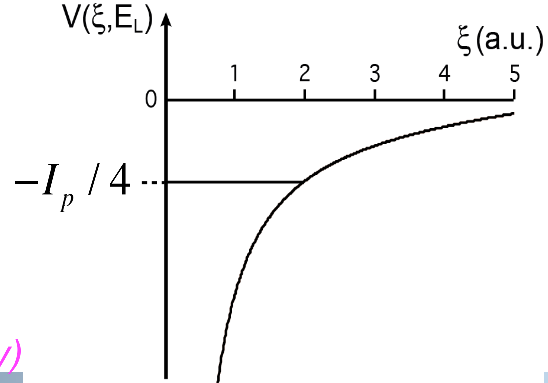
Separation of Schrödinger Equation $\psi = f_1(\xi) f_2(\eta) e^{im\varphi}$

$$V_{para,eff} = -\frac{\left(1-\sqrt{2I_p}/2\right)}{2\eta} - \frac{1}{8}\eta E_L + \frac{m^2-1}{8\eta^2}$$

Field-direction model





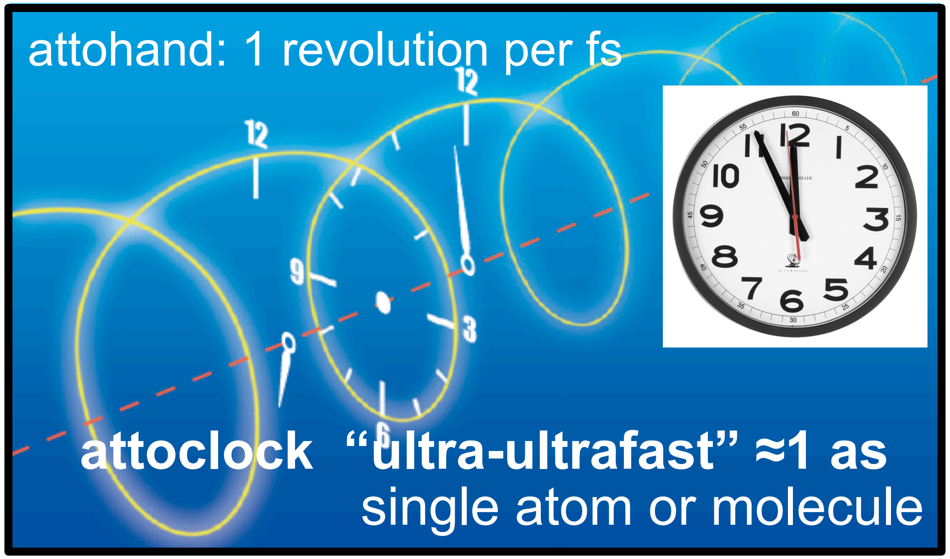


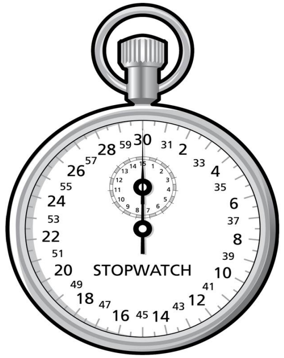
L. D. Landau, E. M. Lifschitz, *Quantum Mechanics (non-relativistic theory)*

Nature Physics **8**, 76, 2012

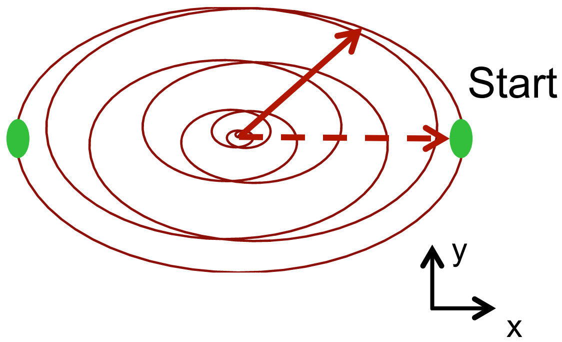
Attoclock for tunneling delay time

infrared field has **close to circular** polarization: "map time to angle – like a clock"
as soon as an electron is liberated it can be accelerated by the rotating electric field and final momentum vector of electron and/or ion maps time to angle





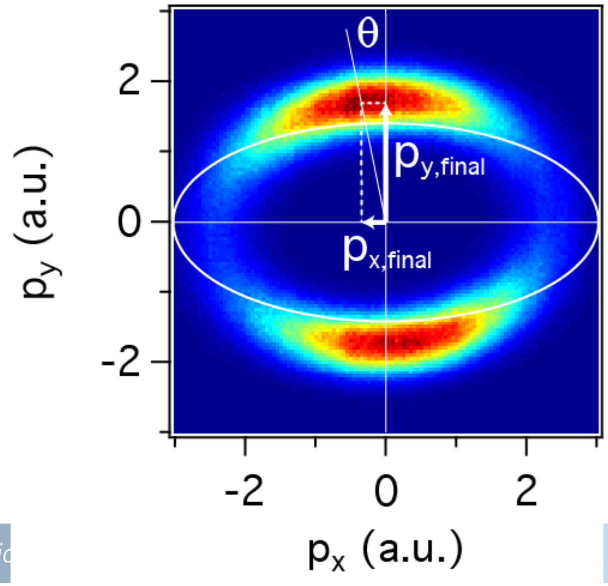
For λ = 735 nm:
1° equal to 7 as



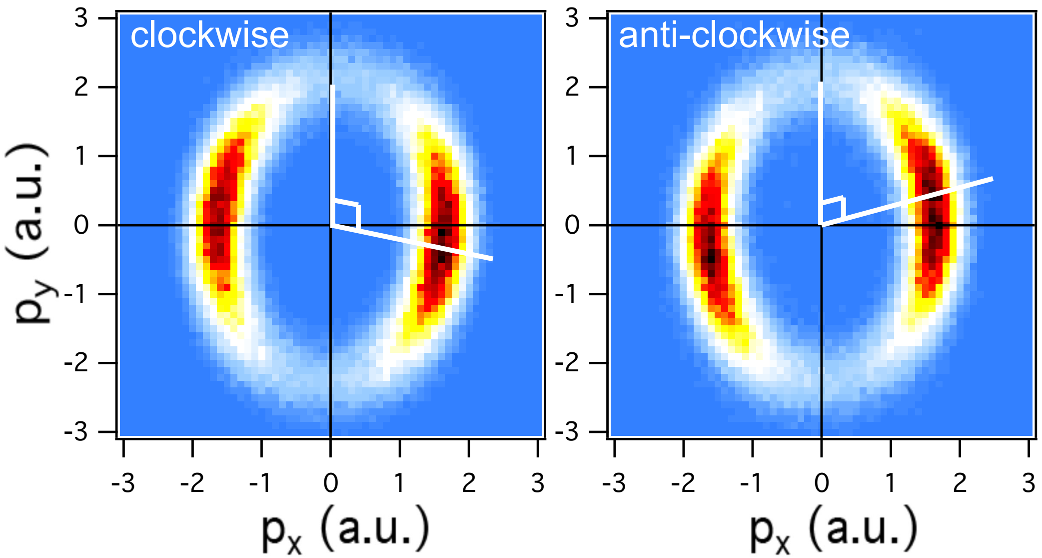
Time zero? Is **precisely defined** with the peak of the electric field (i.e. the most probable trajectory)

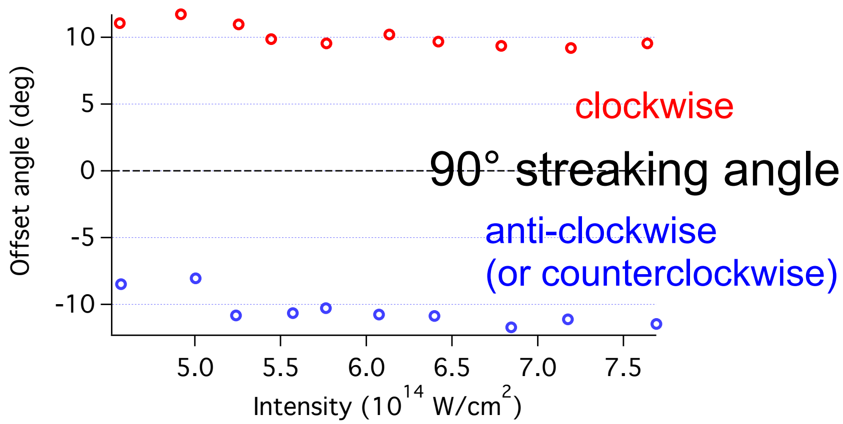
Time measurement? Like a clock which is the **same** microscopically and macroscopically

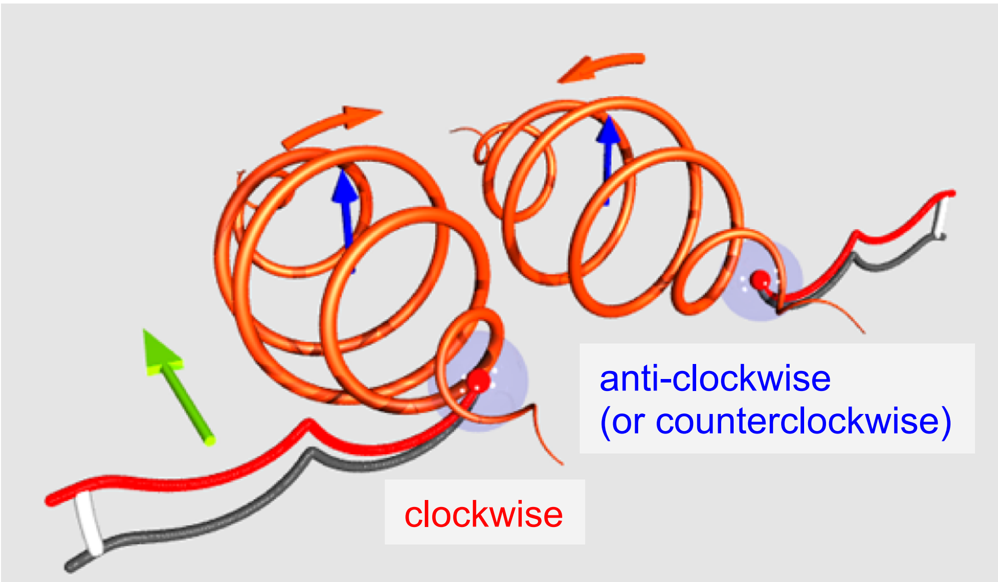
What do we measure? The **final electron momentum vector with the highest electron count** after the short pulse (i.e. the most probable trajectory)

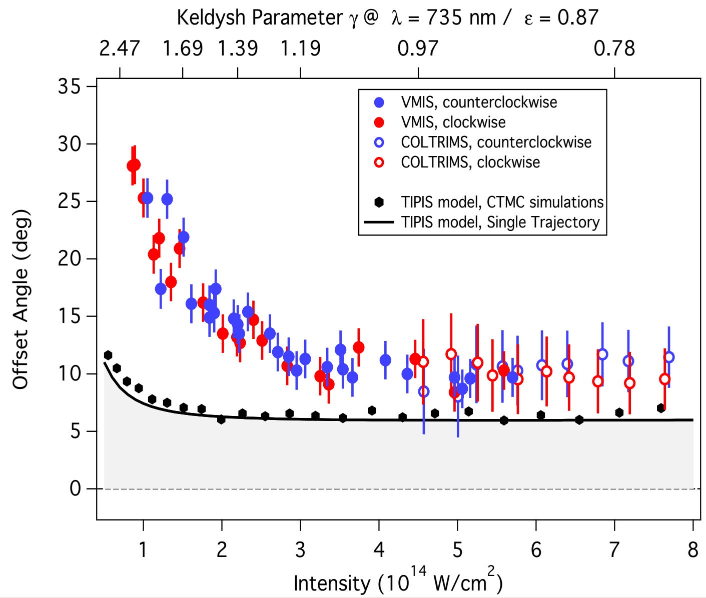


Attoclock: clockwise and anticlockwise





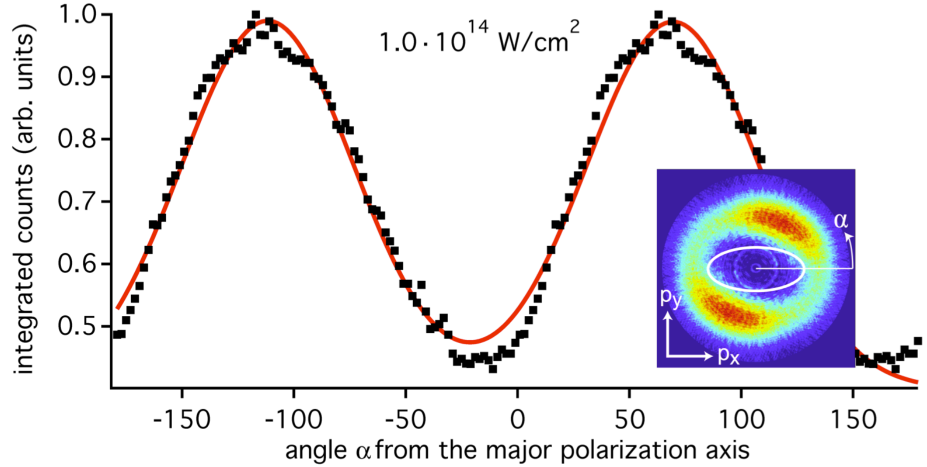




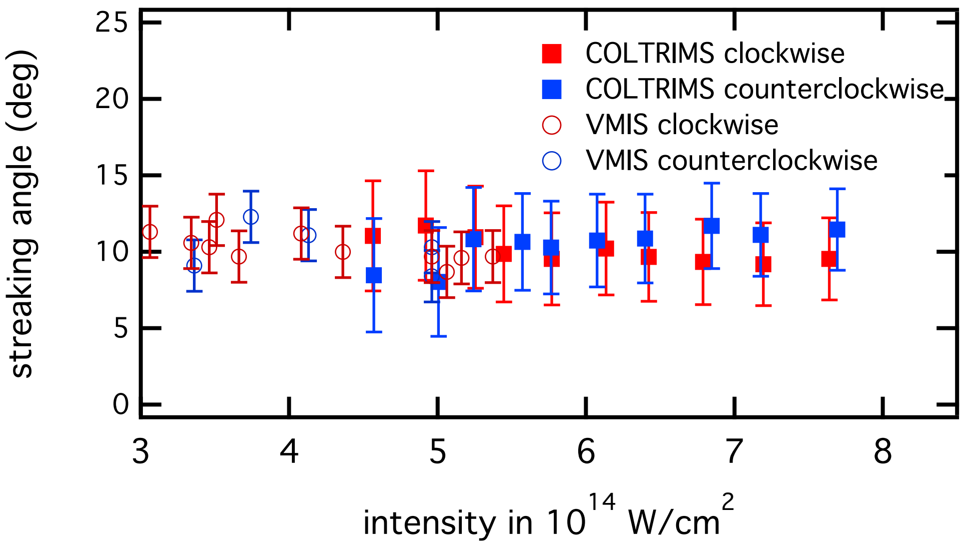
Nature Physics **8**, 76, 2012

Comparison of clockwise and anti-clockwise rotating laser pulses reduces systematic errors

Results: Comparison with COLTRIMS



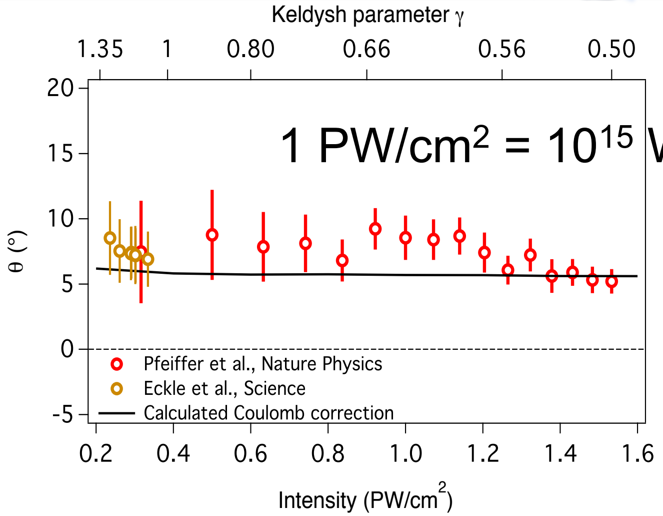
Extraction of the streaking angle of the electron with a double gaussian fit of the photoelectron angular distribution



- COLTRIMS data confirm the agreement of both methods
- High statistics and good momentum resolution result in small error bars

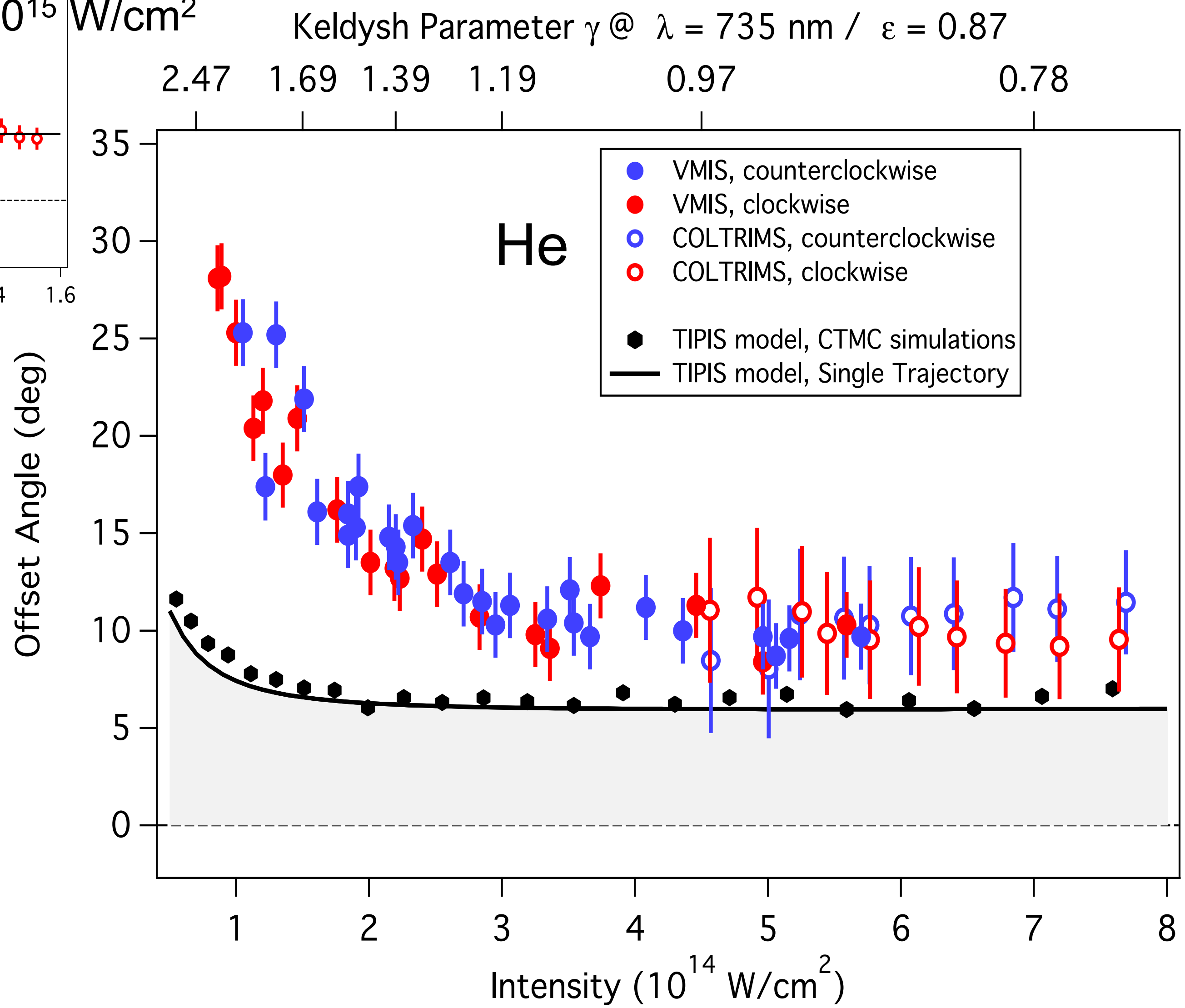
M. Weger, J. Maurer, A. Ludwig, L. Gallmann, U. Keller, "Transferring the attoclock to velocity map imaging" *Optics Express* **21**, 21981, 2013

Attoclock results with both VMIS and 10 kHz COLTRIMS

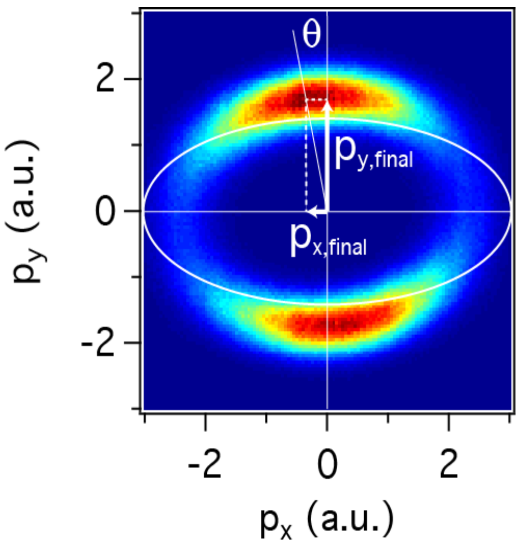


1 PW/cm² = 10^{15} W/cm²

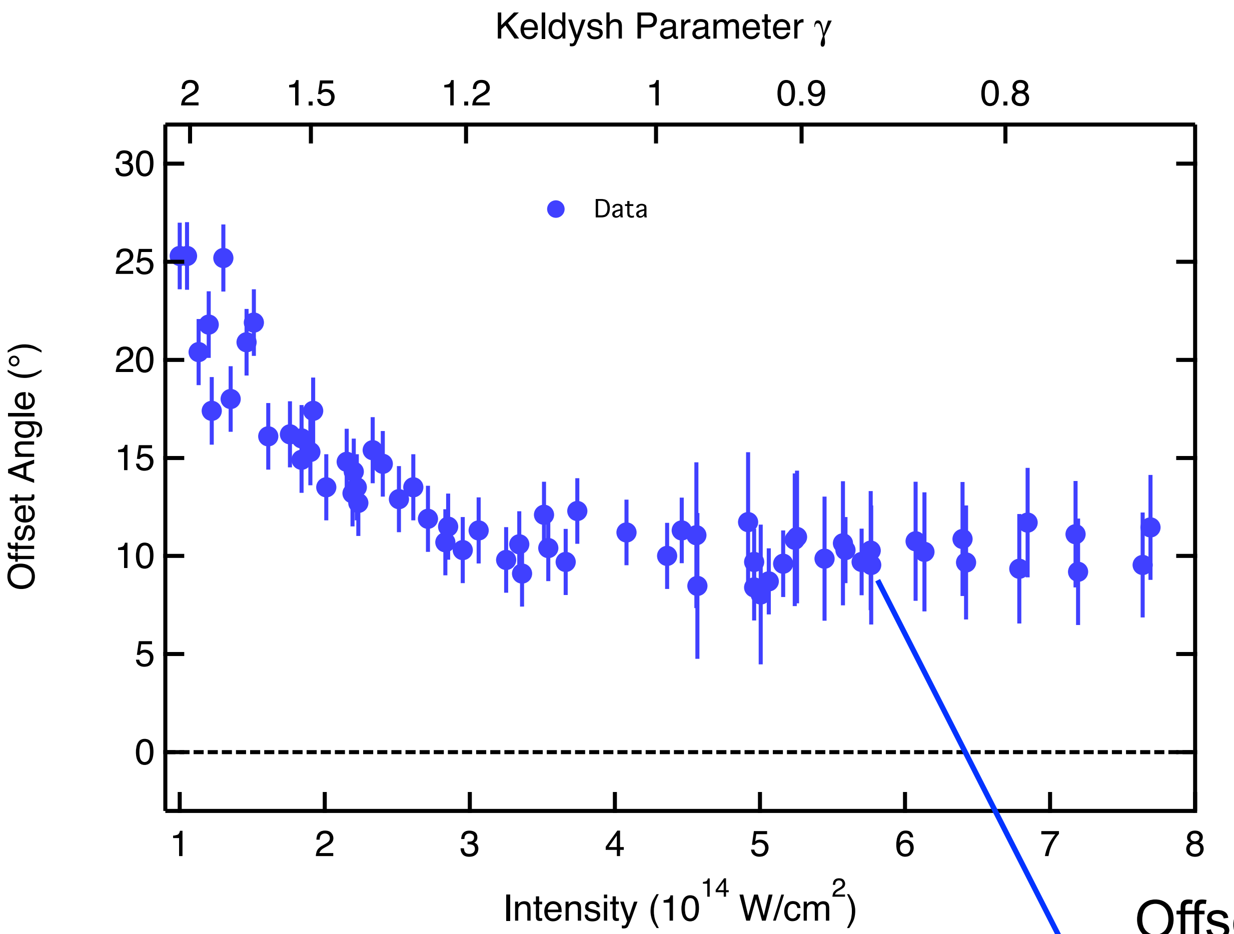
VMIS and 10 kHz COLTRIMS

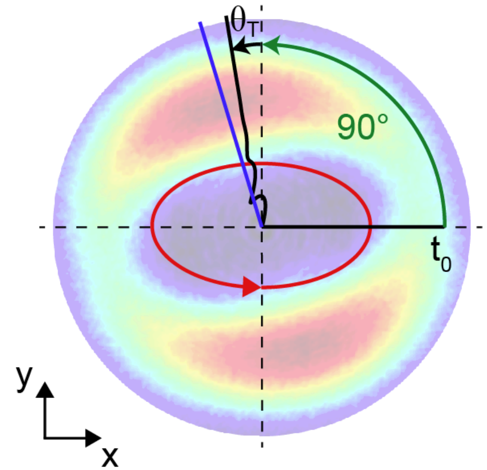


10 kHz COLTRIMS



Results over broad intensity range





Two corrections

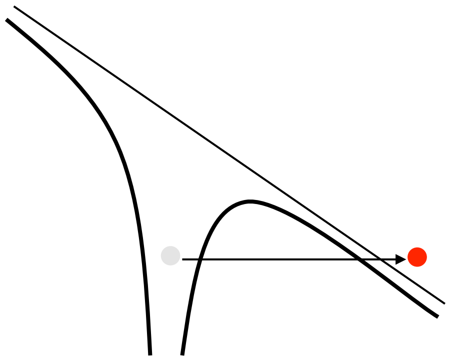
Offset from 90°

$$\omega\tau = \theta_m - \Delta\theta_{str} - \theta_T$$

Adiabatic versus non-adiabatic

TABLE I. Overview over characteristics of theories

Name	Type	r_{exit}	Field-dependence	v_{initial}	$\sigma_{\perp}$	$\sigma_{\|\|,\text{final}}$	Ref.
Keldysh/ADK	adiab.	I_p/F	$\propto \exp\left\{-\frac{2(2I_p)^{3/2}}{3F}\right\}$	0	$\sqrt{\frac{\omega}{2\gamma}}$	$\sqrt{\frac{3\omega}{2\gamma^3(1-\epsilon^2)}}$	[1–3]
parabolic	adiab.	$\frac{I_p+\sqrt{I_p^2-4\beta_2F}}{2F}$	$\propto \exp\left\{-\frac{2(2I_p)^{3/2}}{3F}\right\}$	0	$\sqrt{\frac{\omega}{2\gamma}}$	$\sqrt{\frac{3\omega}{2\gamma^3(1-\epsilon^2)}}$	[4–6]
PPT	non-adiab.	$\leq I_p/F$	$>$	$v_{\perp,i} \neq 0$	$>$	$>$	[7, 8]



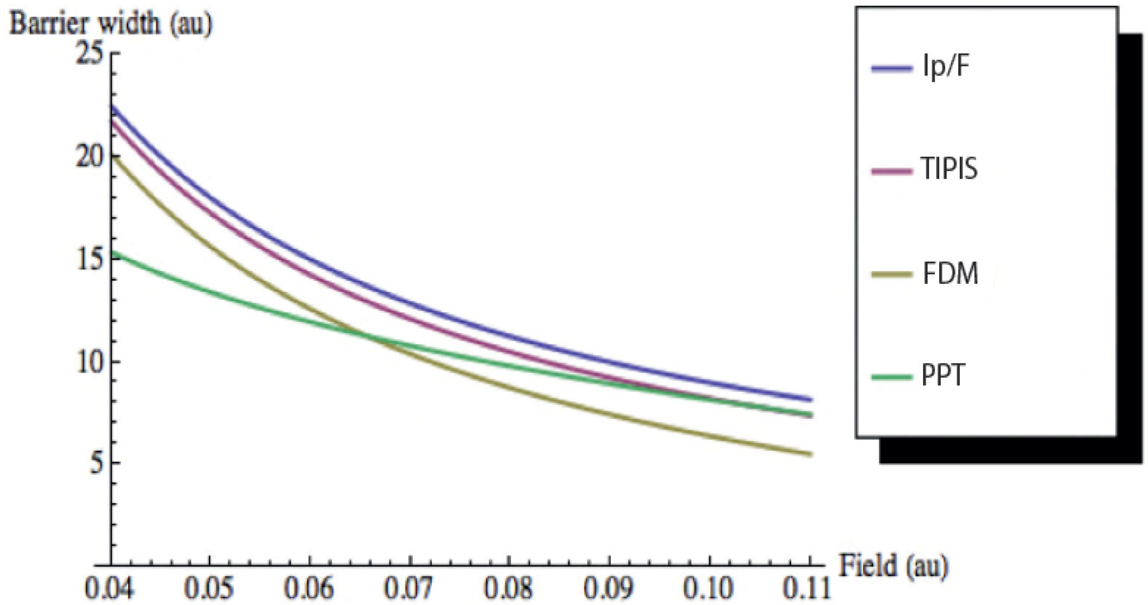
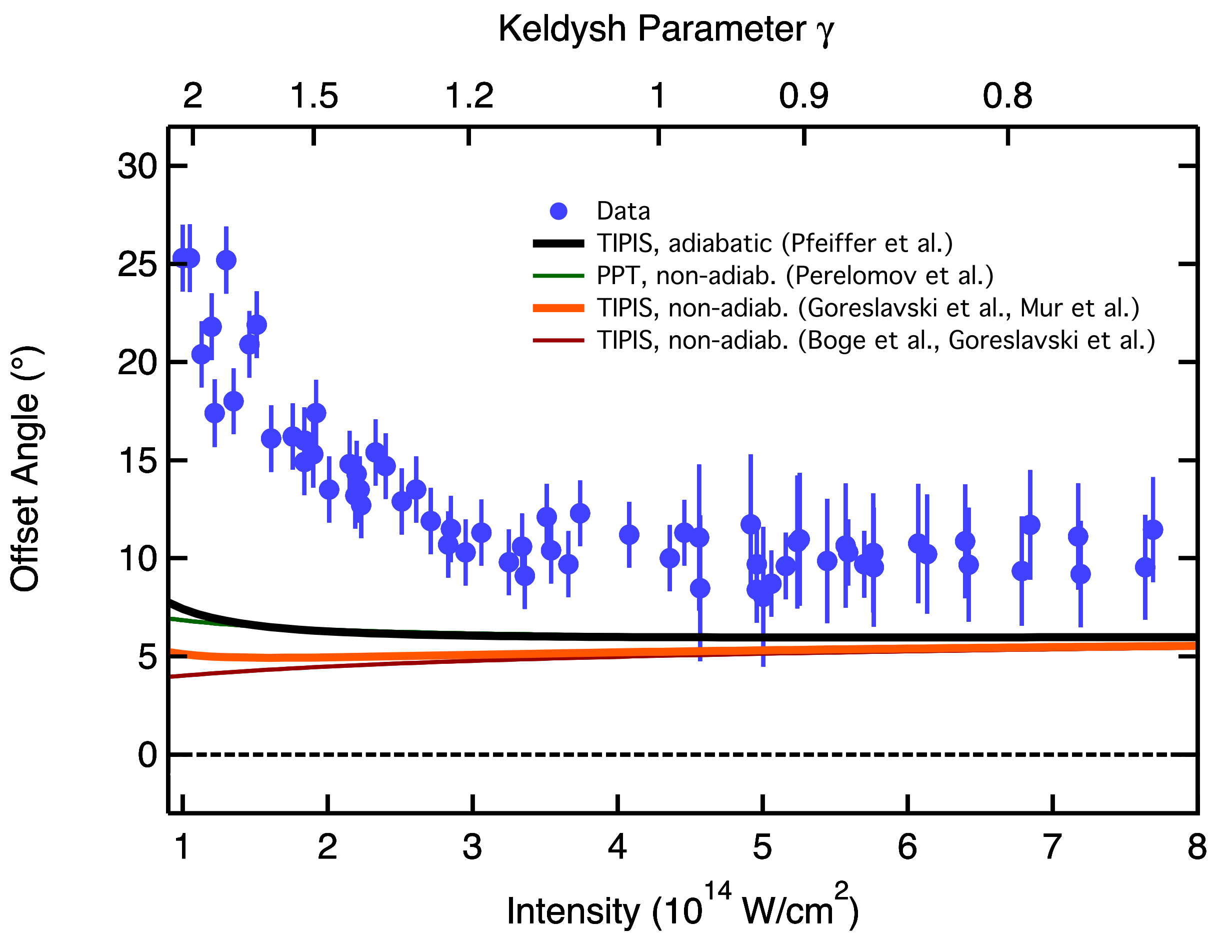


FIGURE 3. Barrier widths predicted by different models: PPT, TIPIS, and FDM (field-direction model). At high field strengths, corresponding to $\gamma \ll 1$, the PPT curve approaches the I_p/F curve.

A. S. Landsman, U. Keller, "Tunneling time in strong field ionization", *J. Phys. B*, in press

Different models for non-adiabatic corrections



Models differ in:

- Starting point $\vec{x}_0$
- Initial velocity $\vec{v}_0$

Adiabatic: $\vec{v}_0 = 0$

Non-adiabatic: $\vec{v}_0 \neq 0$

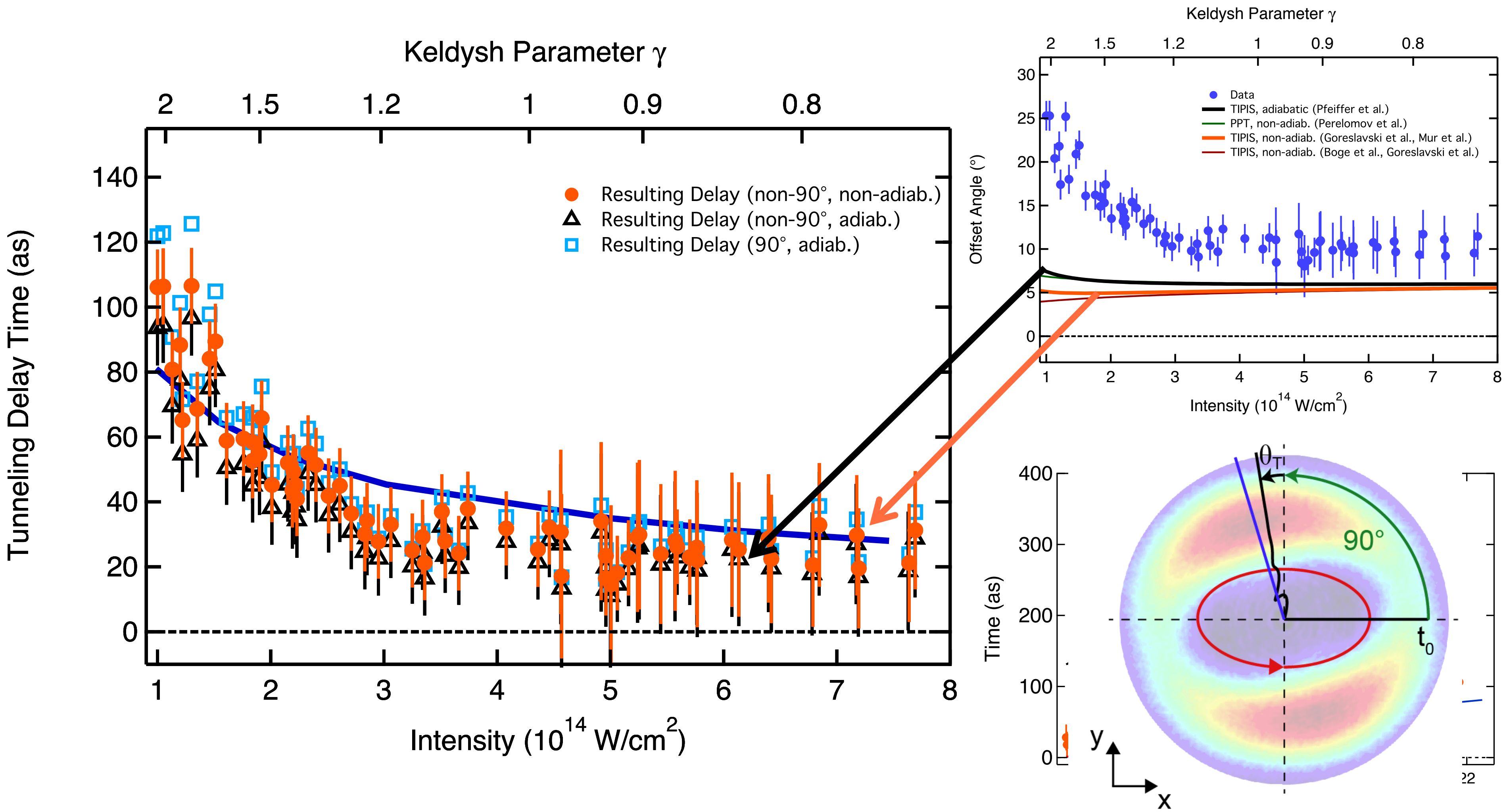
Classical trajectories after ionization:

$$m\ddot{\vec{x}} = \vec{F}_{laser} + \vec{F}_{Coul}$$

$$\omega\tau = \theta_m - \Delta\theta_{str} - \theta_T$$

A. N. Pfeiffer et al., *Nature Physics* **8**, 76 (2012)
N. I. Shvetsov-Shilovski et al., *Phys. Rev. A* **85**, 023428 (2012)
A. M. Perelomov et al., *Zh. Eksp. Teor. Fiz.* **50,** 1393 (1966)
S. P. Goreslavski et al., *PRL* **93,** 233002 (2004)
V. D. Mur et al., *J. Exp. Theor. Phys.* **92**, 777-788 (2001)
R. Boge et al., *PRL* **111**, 103003 (2013) – Keller group, „Probing non-adiabatic effects in strong-field tunnel ionization“

Resulting tunneling delay time



$$\omega\tau = \theta_m - \Delta\theta_{str} - \theta_T$$

Theoretical Description

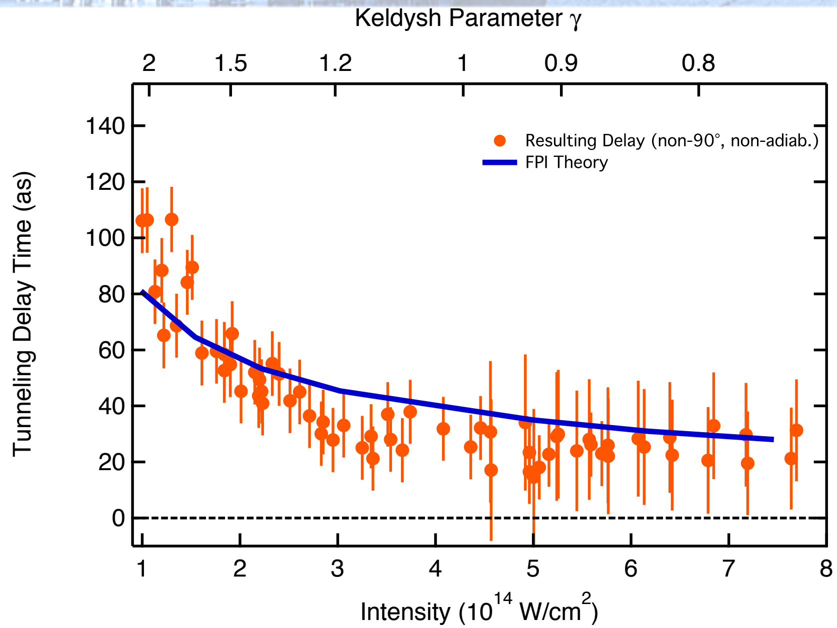
Calculation of tunneling delay time distributions with a Feynman Path Integral (FPI) formalism

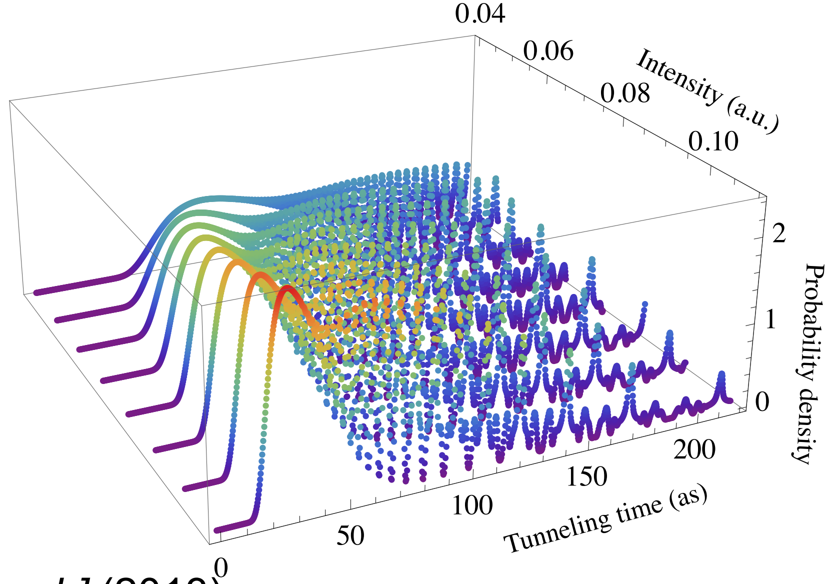
Good agreement with experimental data

Data consistent with tunneling picture

Implications for attoscience:

- Instantaneous tunneling is in general not a good assumption
- When do hole dynamics really begin to evolve? Not instantaneously!

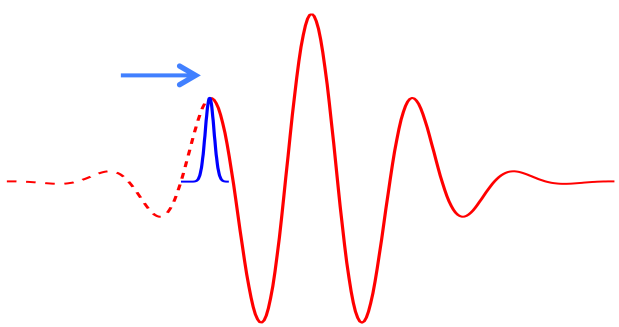


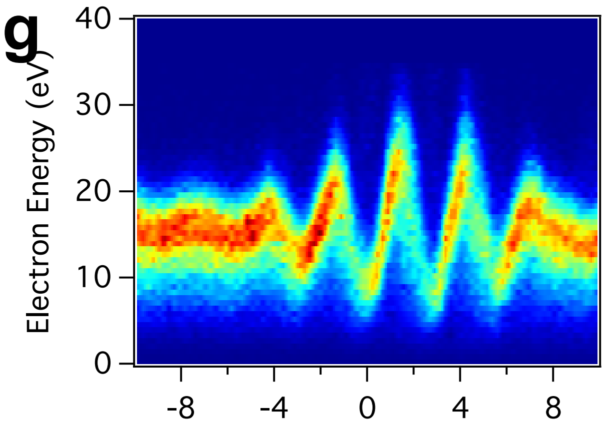


A. Landsman et al., *arXiv:1301.2766 [physics.atom-ph]* (2013)

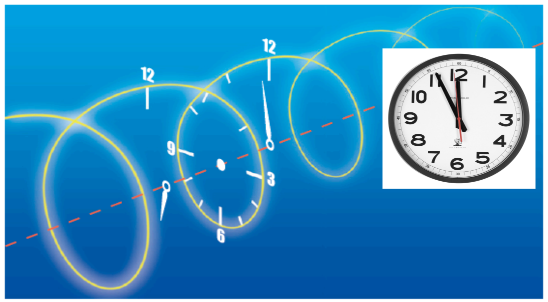
Exciting new results in attosecond science ...

Attosecond energy streaking



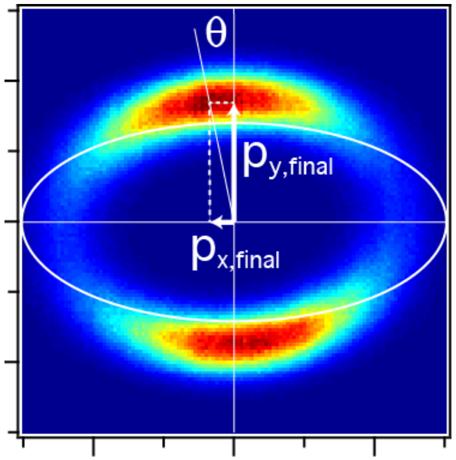


Attoclock

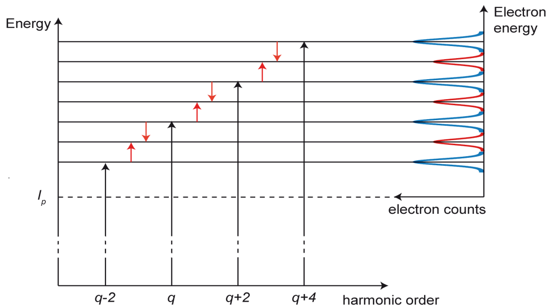


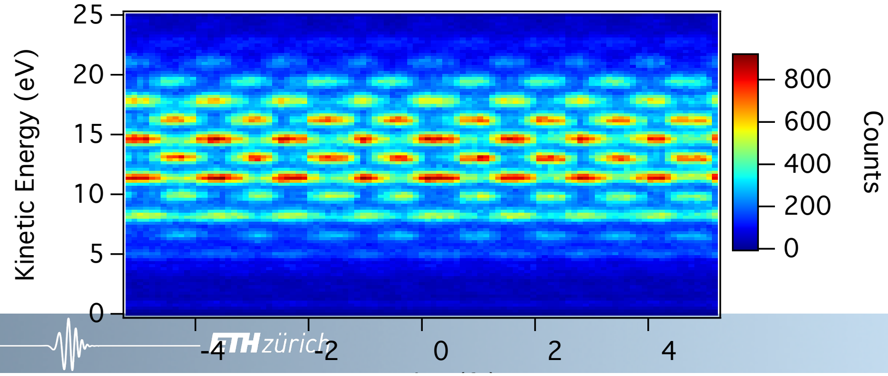
For λ = 735 nm:

1° equal to 7 as



Attosecond interferometer: RABBITT





What next?

We will continue to do more measurements
(angle resolved, molecules etc.)

We need to sort out streaking and RABBITT differences

theoretically they should be the same ...

What about the attoclock technique?

Can also be used for single photon emission:

IR field low such that no tunnel ionization occurs
and single attosecond pulse for single photon absorption on two gas targets.

We need more agreements on theory

We can provide more systematic measurements with changing parameters

What approximations do we agree with?

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What approximations do we agree with?
Example: breakdown of the electric dipole approximation

Application – Strong-field ionization

VMIS: Velocity Map Imaging Spectrometer

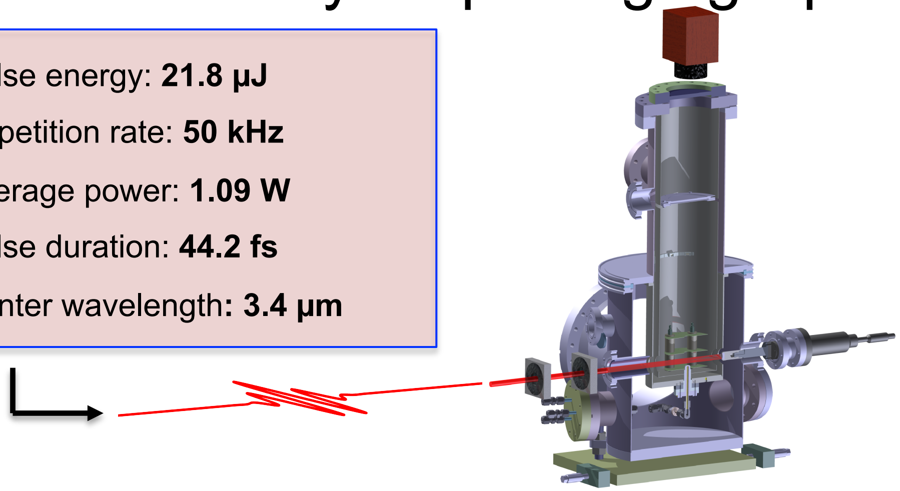
Pulse energy: **21.8 µJ**

Repetition rate: **50 kHz**

Average power: **1.09 W**

Pulse duration: **44.2 fs**

Center wavelength: **3.4 µm**



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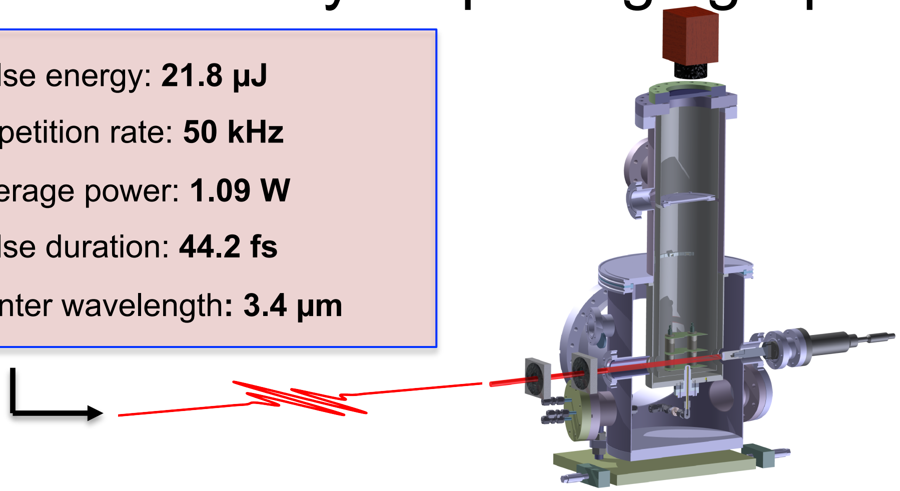

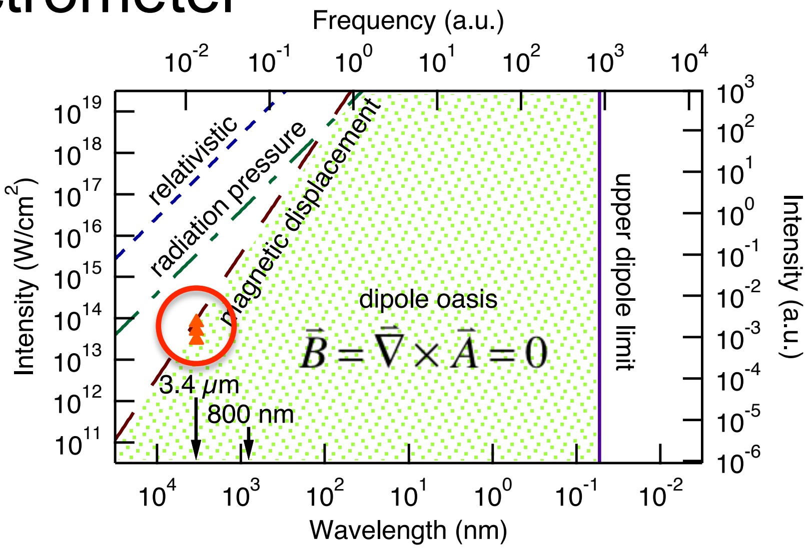

Dipole approximation

$$\vec{B} = \vec{\nabla} \times \vec{A} = 0$$

$$\vec{A}(\vec{r}, t) \equiv \vec{A}(t)$$

H. R. Reiss, *Phys. Rev. Lett.* **101,** 043002 (2008)

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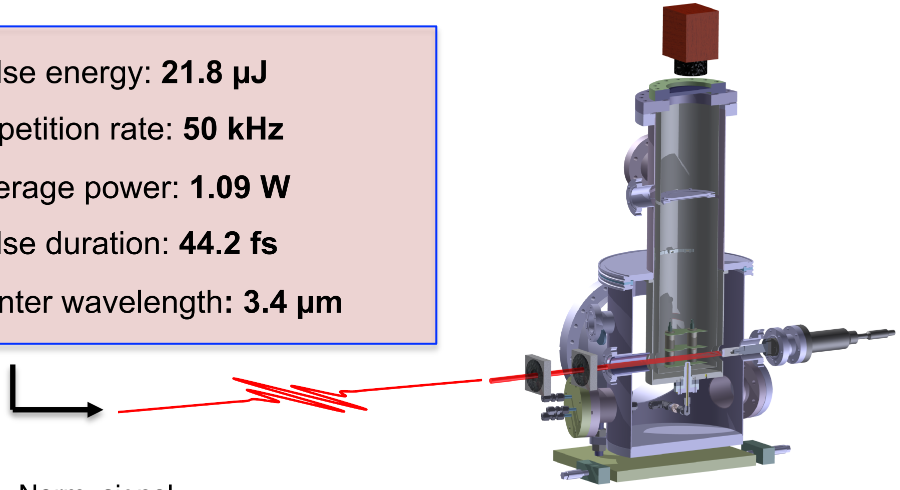

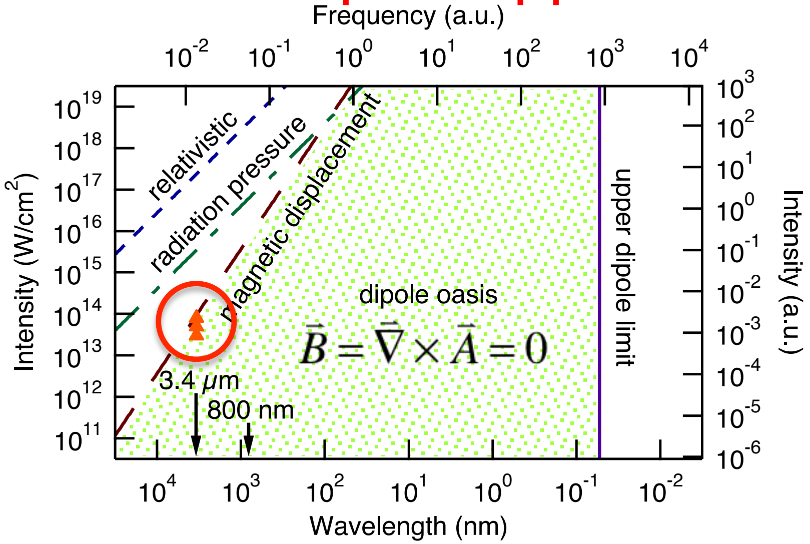

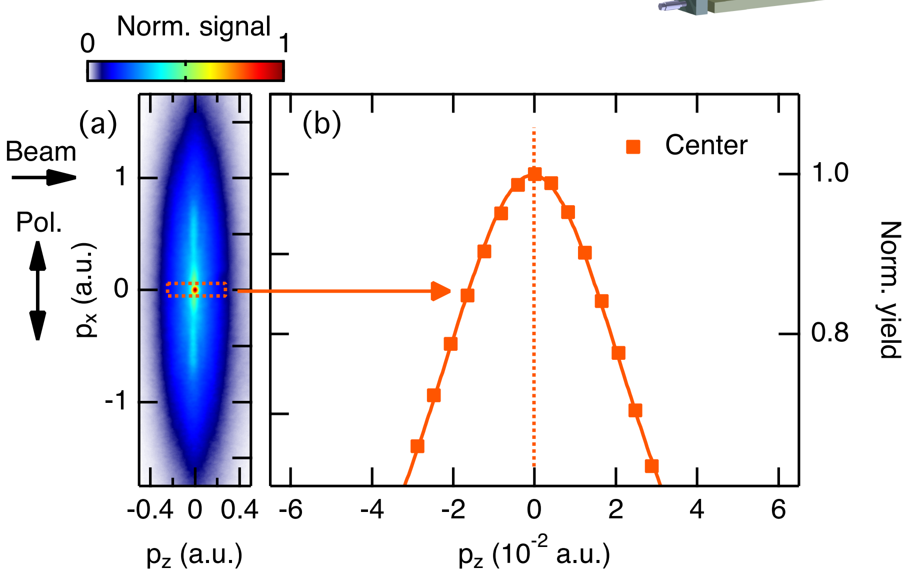

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J. Maurer, A. Ludwig, B. W. Mayer, C. R. Phillips, L. Gallmann, and U. Keller, *CLEO 2014: Postdeadline Paper*, FTh5A.10.

A. Ludwig, J. Maurer, B. W. Mayer, C. R. Phillips, L. Gallmann, and U. Keller, arXiv:1408.2336 [physics.atom-ph]

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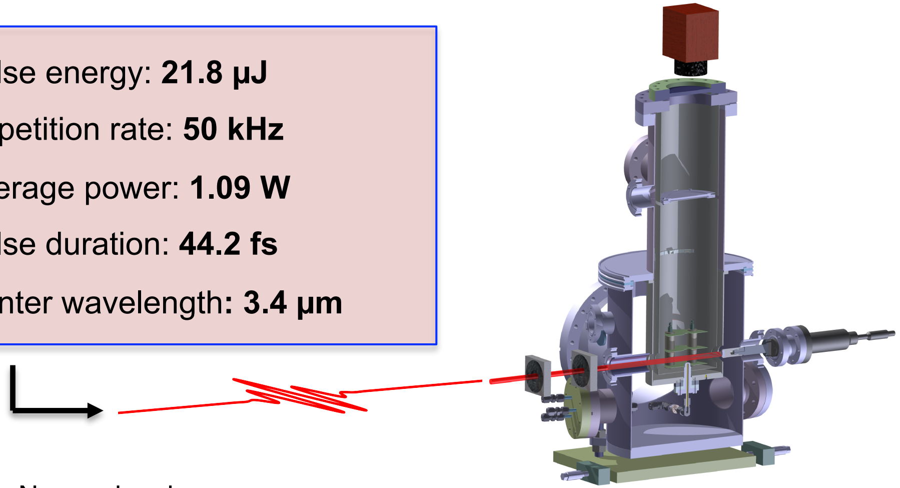

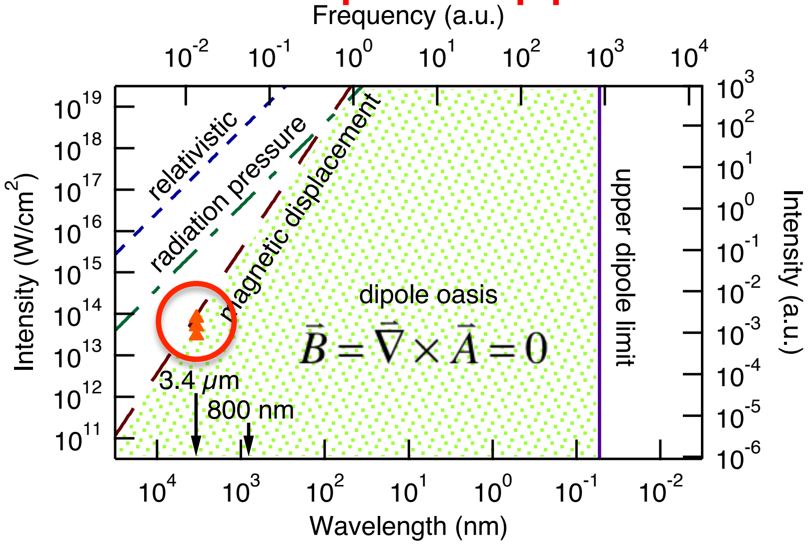

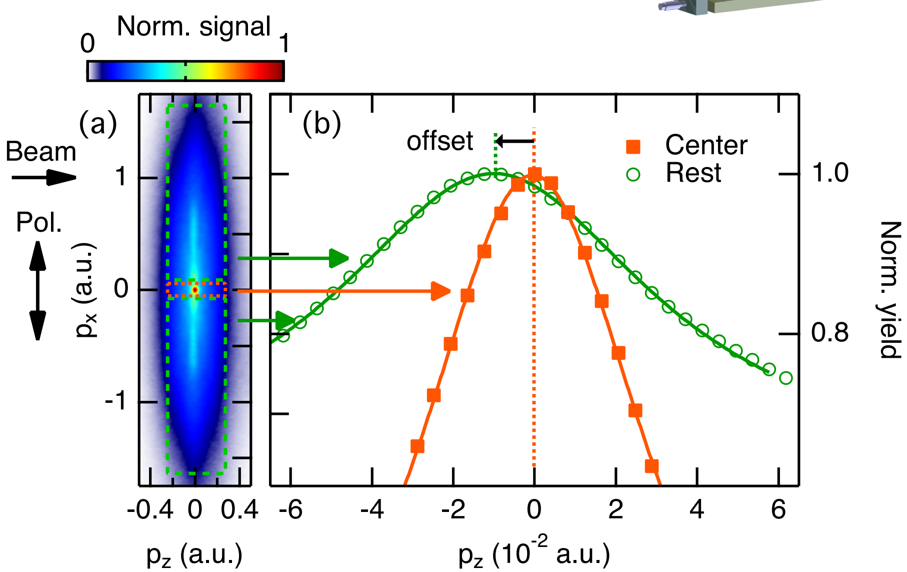

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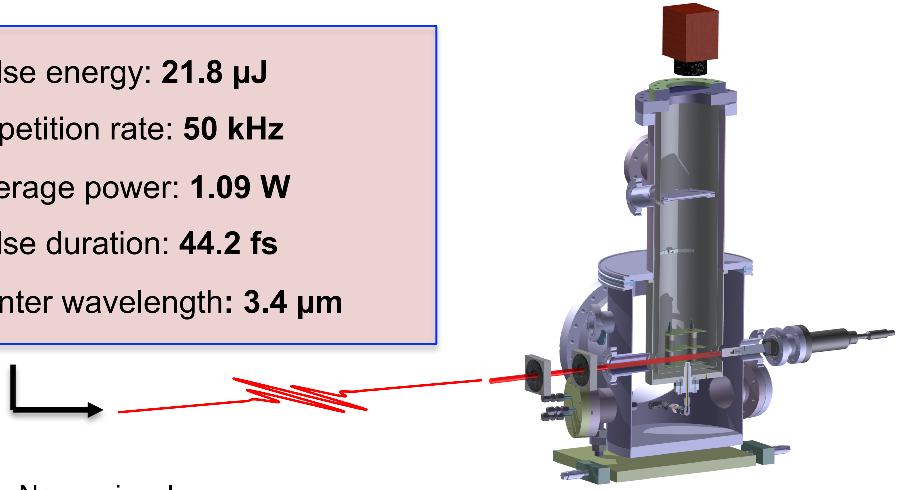

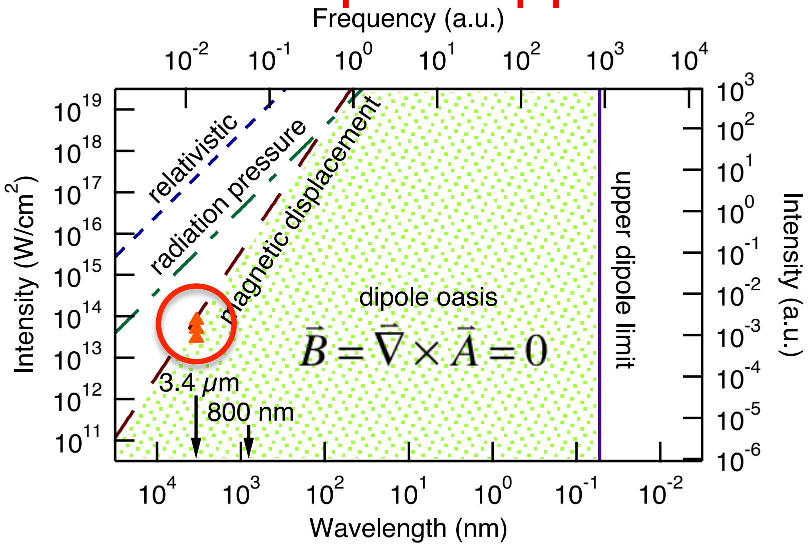

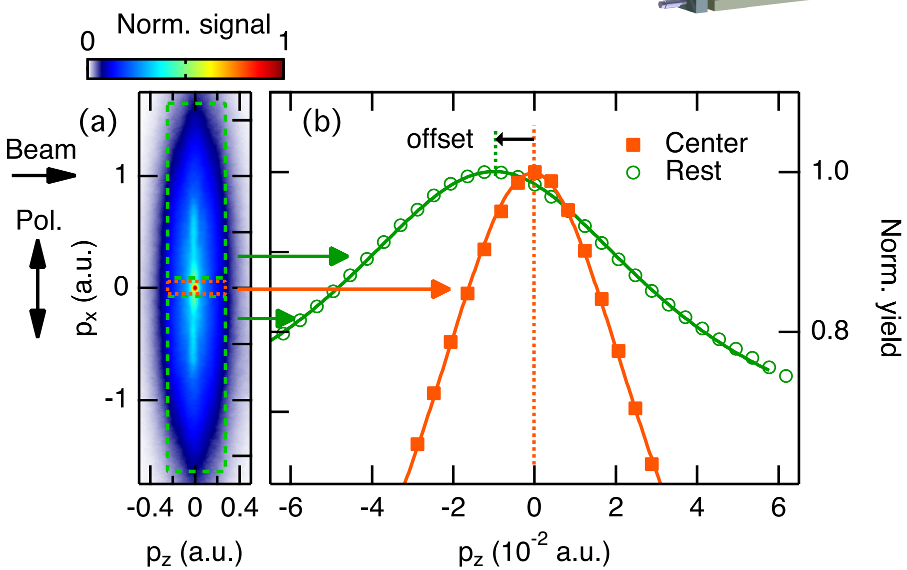

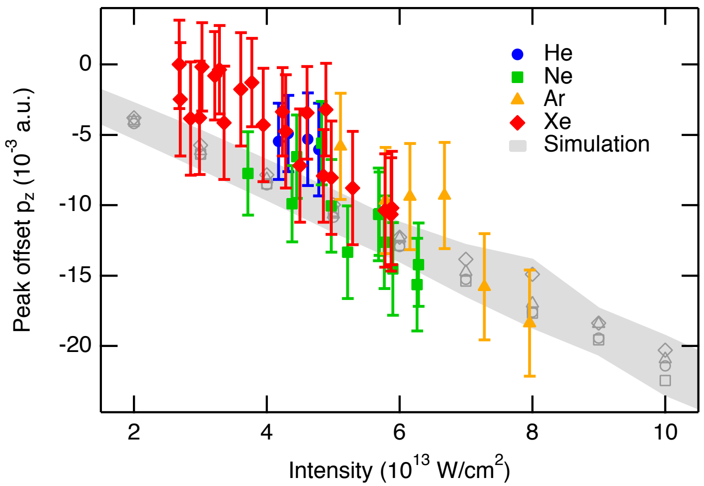

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