MAX BORN-INSTITUTE

Time and Spin in Attosecond Spectroscopy

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Max-Born Institute, Berlin

The Roadmap

- Defining ionization time: from one to many photons
  - The Spin-Orbit Larmor clock for ionization
  - Connection to Larmor time for tunnelling
  - Connection to quantum orbits and WKB analysis

- Measuring strong-field ionization times
  - The atto-clock setup
  - The transient absorption setup: IR pump – XUV probe. Effects of electron-hole entanglement
  - The high harmonic spectroscopy setup
  - Connection between different measurement protocols

Theory:

PhD students

Lisa Torlina, MBI, Berlin

Jivesh Kaushal MBI, Berlin

Felipe Morales, MBI Berlin

Harm Geert Muller

Misha Ivanov, MBI Berlin

Experiment:

Yann Mairesse , CELIA, Bordeaux

Nirit Dudovich , Weizmann

Dror Shafir , Weizmann Institute,

Hadas Soifer, Weizmann Institute,

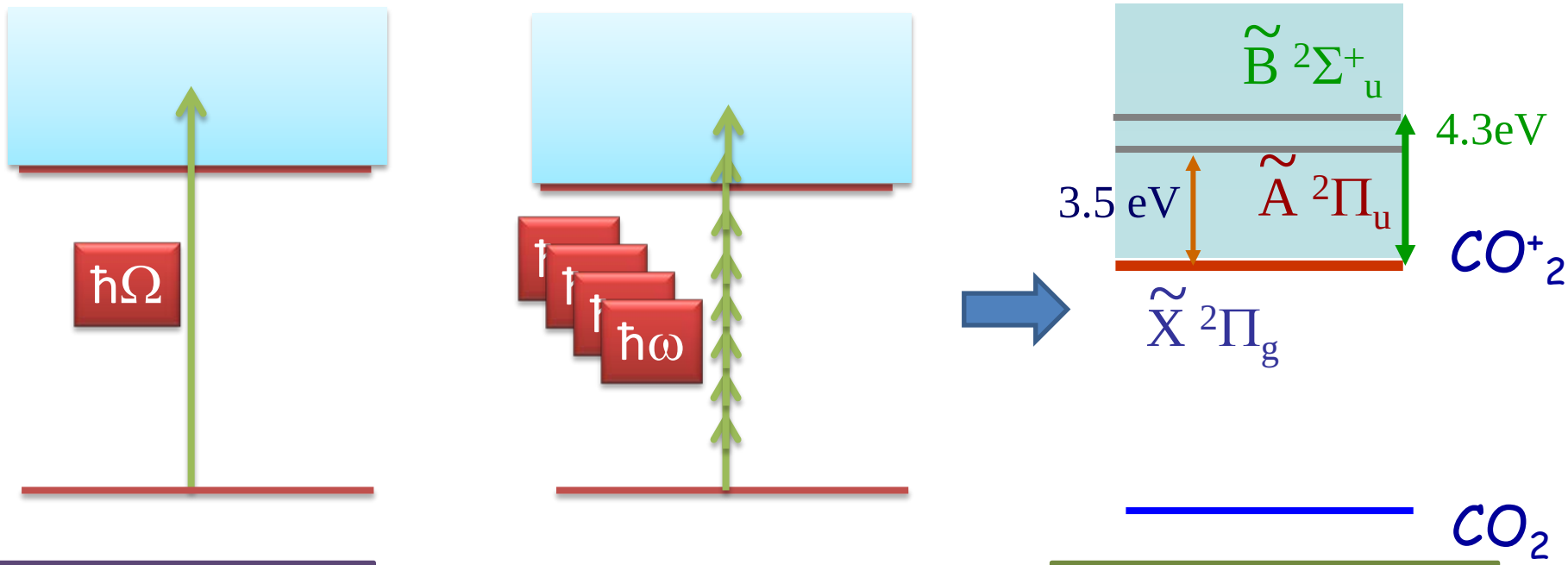
Attosecond spectroscopy: Goals & Challenges

Goal:

Observe & control electron dynamics at its natural time-scale (1asec=10^{-3}fsec)

One of key challenges:

- Observe non-equilibrium many-electron dynamics
  - This dynamics can be created by photoionization
  - Electron removal by an ultrashort pulse creates coherent hole



Ionization by XUV

Ionization by IR

Coherent population of several ionic states

Attosecond spectroscopy: Questions

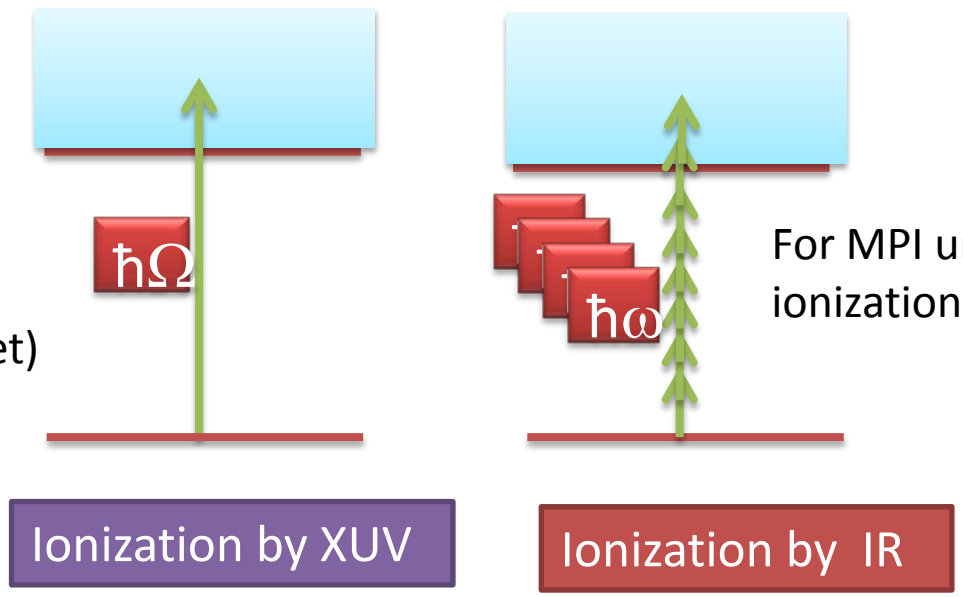
How long does it take to remove an electron with light and create a hole?

Experiment & Theory:

Eckle, P. et al Science 322, 1525–1529 (2008).
Goulielmakis, E. et al, Nature 466 (7307), 700-702 (2010)
Schultze, M. et al. Science 328, 1658–1662 (2010).
Klunder, K. et al., Phys. Rev. Lett. 106, 143002 (2011)
Pfeiffer, A. N. et al. Nature Phys. 8, 76–80 (2012)
Shafir, D. et al. Nature 485 (7398), 343-346 (2012)

Wigner-Smith Time (group delay of the electron wave-packet)

J. M. Dahlström et al,
J. Phys B,45,183001 (2012)



For MPI unique definition of ionization time is missing

Today: New questions

- How does this time depend on the number of absorbed photons (strong IR vs weak XUV)?
- What is the connection between different measurement protocols, e.g. atto-clock vs HHG vs IR pump-XUV-probe transient absorption ?
- How does electron-hole entanglement affect electron rearrangement and its time-scale?

Can we find a clock to define this time?

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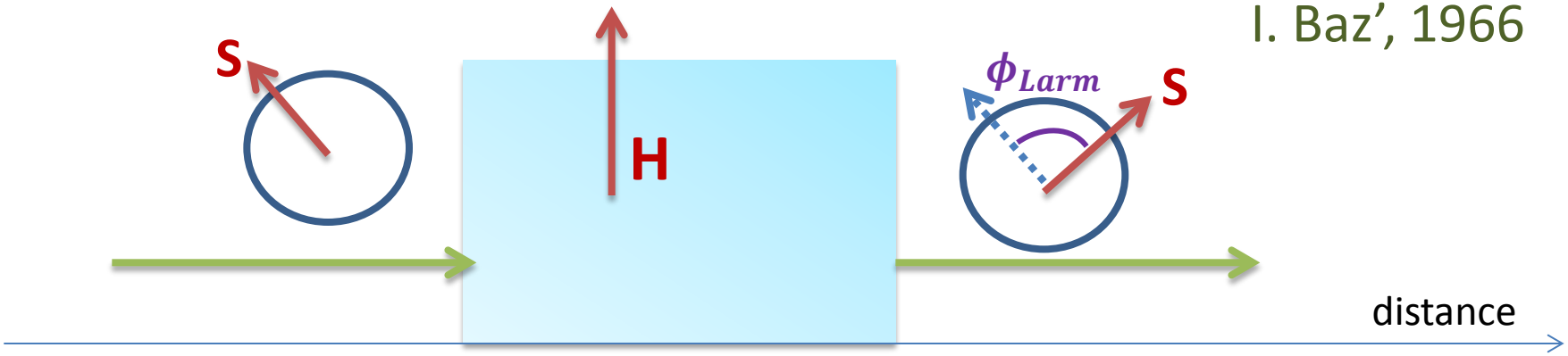
The Larmor clock for tunnelling

I. Baz', 1966



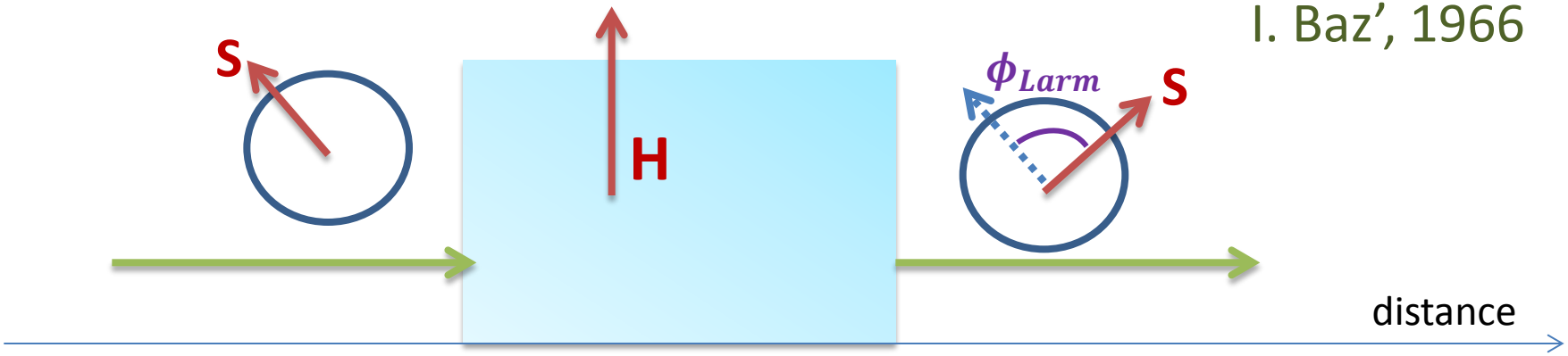
Today:

There is a built-in Larmor-like clock in atoms!

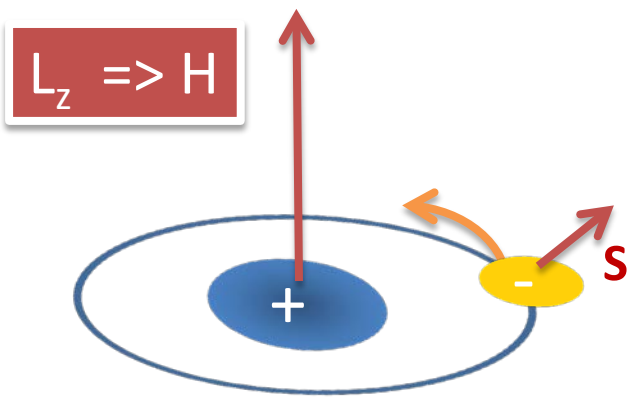
- Based on Spin-Orbit Interaction
- Good for any number of photons N
- We will use it to define ionization time

The Larmor clock for tunnelling

I. Baz', 1966



Spin-orbit interaction



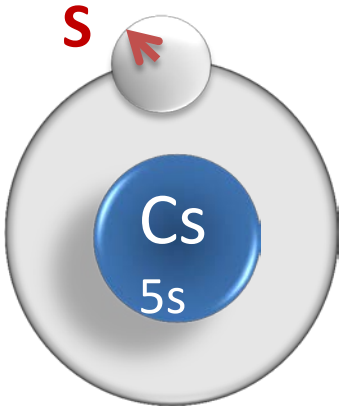
For e^-, the core rotates around it

- Rotating charge creates current
- Current creates magnetic field
- Electron's spin precesses in this field

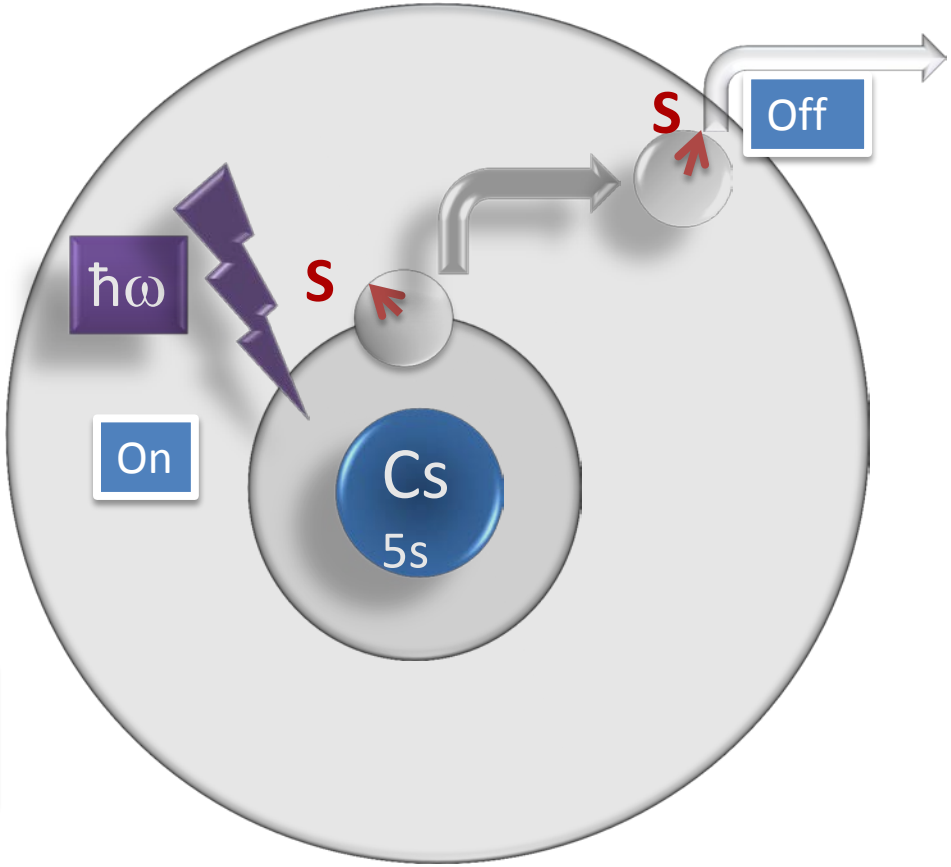
- The clock must be calibrated

Gedanken experiment for Calibrating the clock

One-photon ionization of Cs by right circularly polarized pulse



No SO interaction in the ground state



Detect electron spin

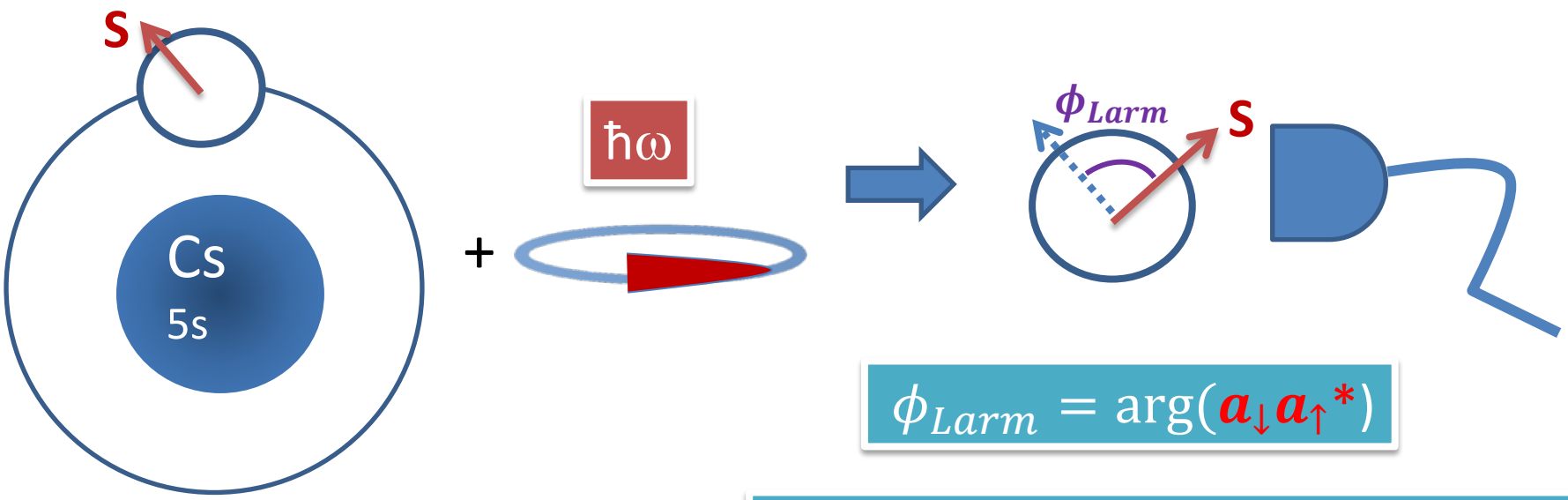
Photon absorption turns on SO interaction

Gedanken experiment for Calibrating the clock

One-photon ionization of Cs by right circularly polarized pulse
Define angle of rotation of electron spin during ionization

$$|s_{in}\rangle = \alpha|\downarrow\rangle + e^{i\varphi}\beta|\uparrow\rangle$$

$$|s_f\rangle = \boldsymbol{a}_{\downarrow}\alpha|\downarrow\rangle + \boldsymbol{a}_{\uparrow}e^{i\varphi}\beta|\uparrow\rangle$$

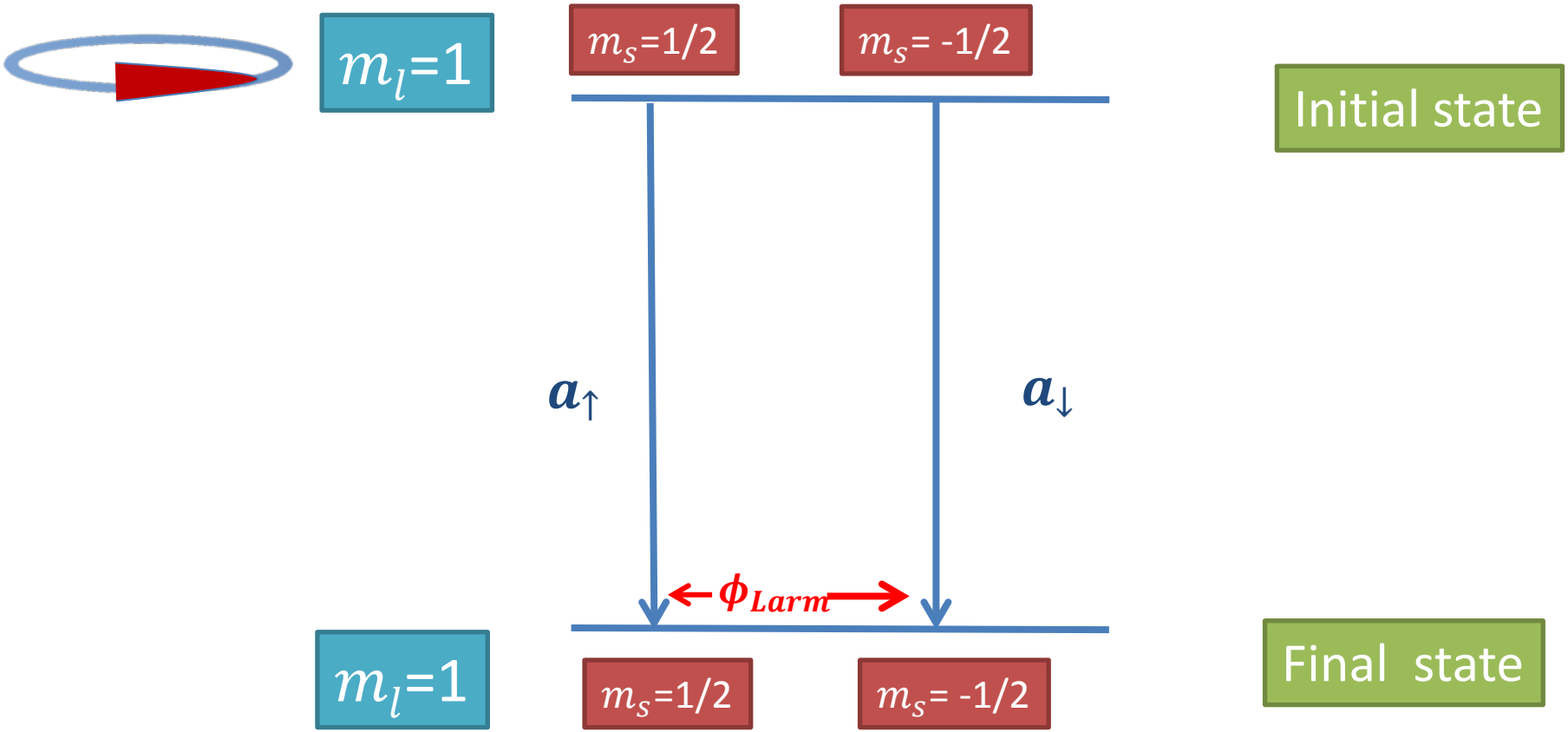


$$\phi_{Larm} = \arg(\boldsymbol{a}_{\downarrow}\boldsymbol{a}_{\uparrow}^{*})$$

No SO interaction in the ground state

ionization amplitude:
$\boldsymbol{a}_{\downarrow}$ for spin-down component
$\boldsymbol{a}_{\uparrow}$ for spin-up component

SO Larmor clock as Interferometer



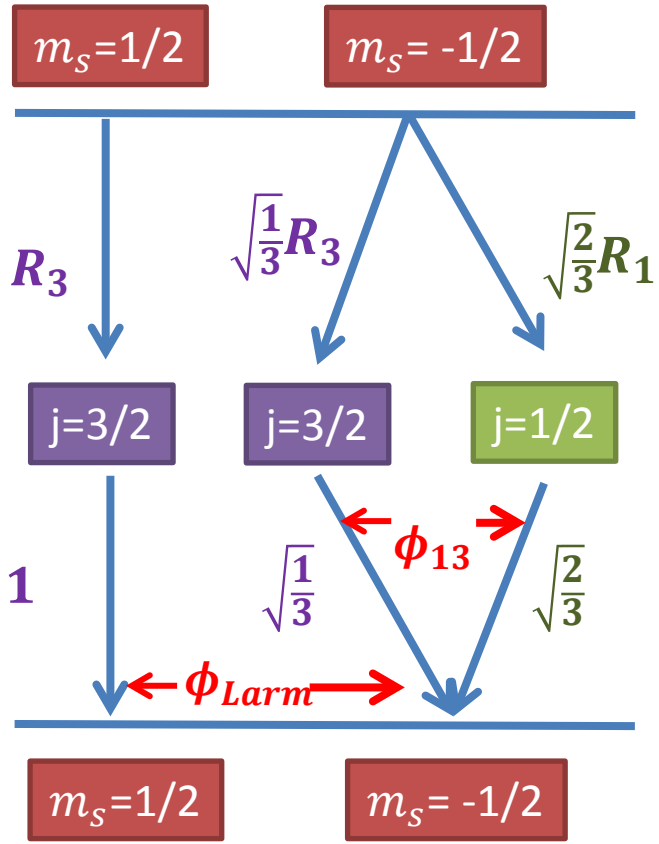
- Record the phase between the spin-up and spin-down pathways

- Looks easy, but … -- the initial and final states are not eigenstates, thanks to the spin-orbit interaction

SO Larmor clock as Interferometer

$R_{1,3} = |R_{1,3}| e^{i\phi^R_{1,3}}$

Radial photoionization matrix element



U. Fano, 1969

Phys Rev 178,131

A crooked interferometer: arm + double arm

$$a_\uparrow = R_3$$

$$a_\downarrow = \frac{1}{3}(R_3 + 2R_1)$$

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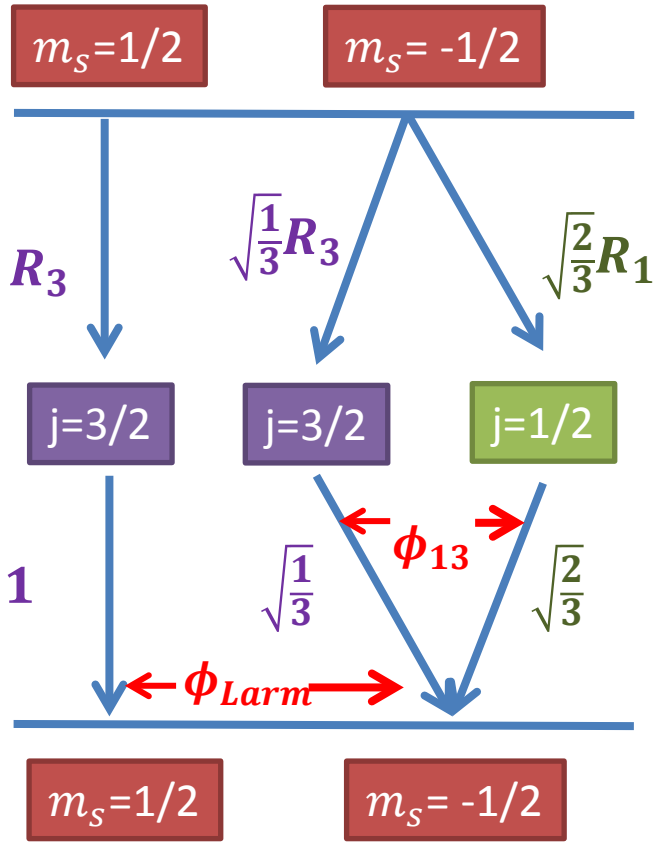
Radial photoionization matrix element



A crooked interferometer: arm + double arm

$$a_\uparrow = R_3$$

$$a_\downarrow = \frac{1}{3}(R_3 + 2R_1)$$

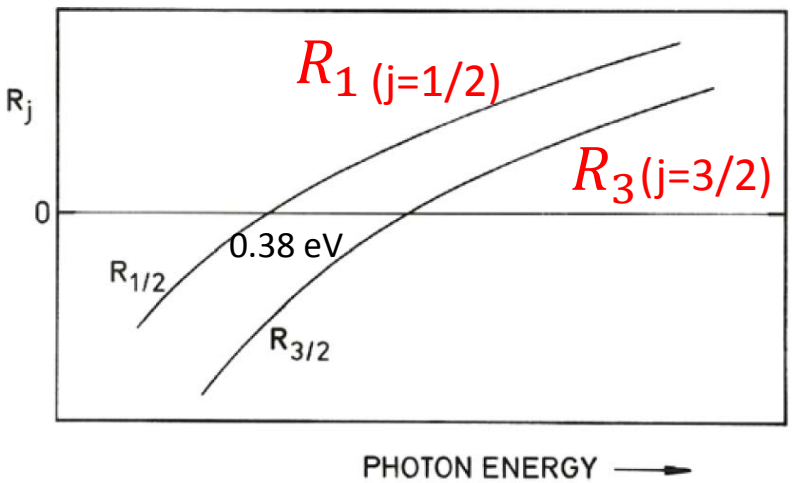
$$\tan\phi_{LARM} = -\frac{\sin(\phi_1^R - \phi_3^R)}{0.5|R_3|/|R_1| + \cos(\phi_3^R - \phi_1^R)}$$

Wigner-Smith time hides here

The appearance of Wigner-Smith time

$$\phi_1^R - \phi_3^R = ?$$

J. Cond. Matter 24 (2012) 173001



$$\phi_1^R - \phi_3^R = \phi\,(E + \Delta E_{SO}) - \phi(E)$$
$$= \tau_{WS}\Delta E_{SO}$$

$$\tau_{WS} = \partial\phi(E)/\partial E$$

Wigner-Smith time

$$\tan\phi_{LARM} = -\frac{\sin(\tau_{WS}\Delta E_{SO}^e)}{0.5|R_3|/|R_1| + \cos(\tau_{WS}\Delta E_{SO}^e)}$$

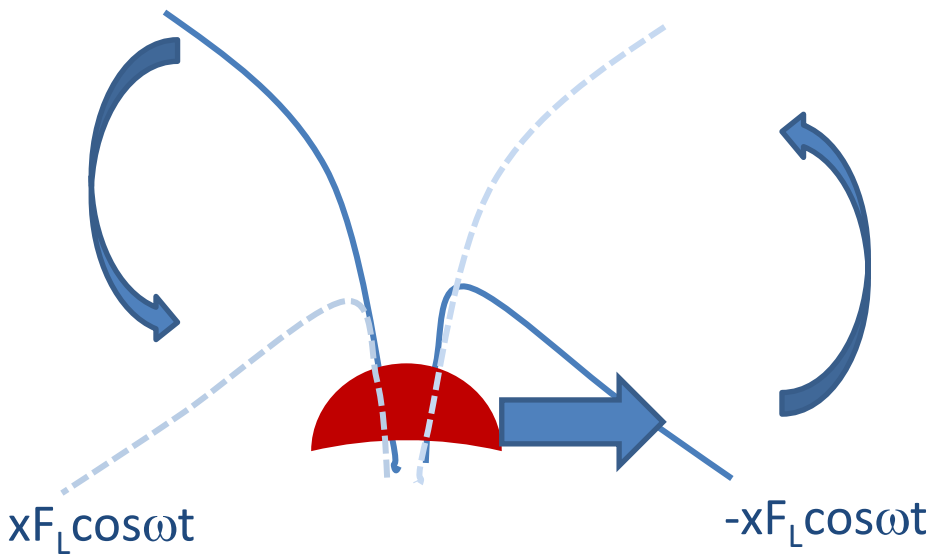
We have calibrated the clock

Strong Field Ionization in IR fields

Keldysh, 1965

Multiphoton Ionization: N>>1

Adiabatic (tunnelling) perspective ($\omega/I_p << 1$)

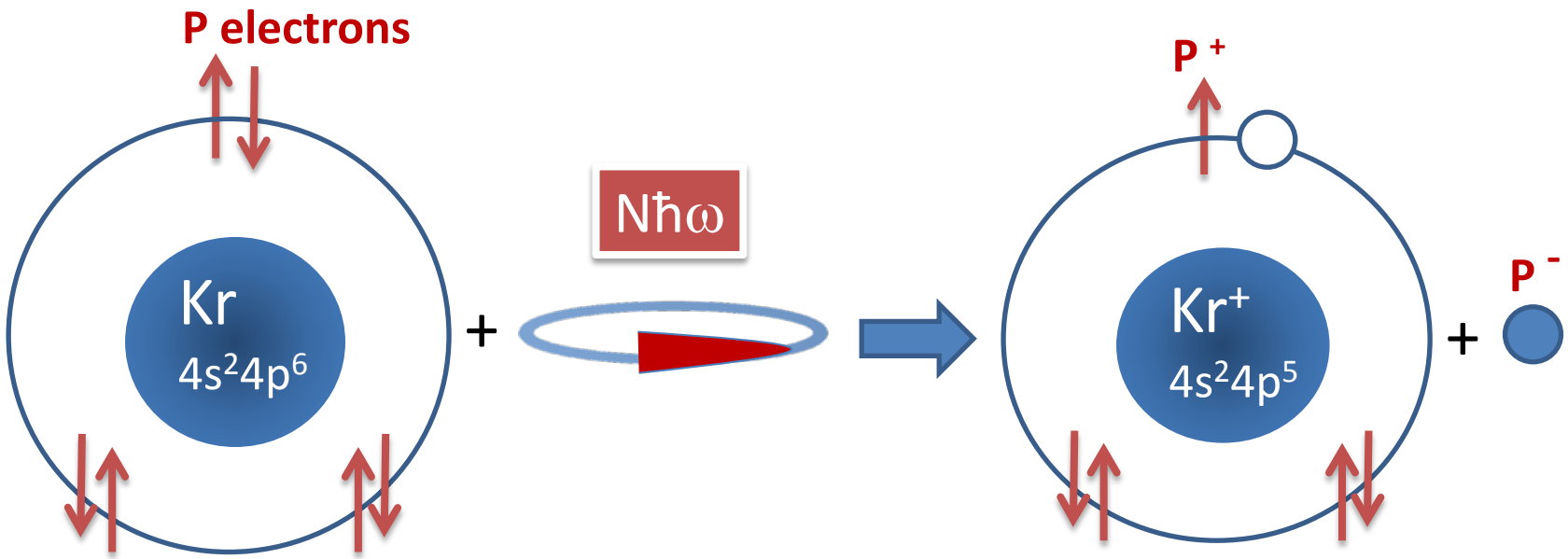


Find time it takes to create a hole in general case for arbitrary Keldysh parameter

Strong-field Ionization in IR fields: Circular polarization

N>>1 ionization preferentially removes p^- (counter-rotating) electron

- Theoretical prediction: Barth, Smirnova, PRA, 2011
- Experimental verification: Herath et al, PRL, 2012



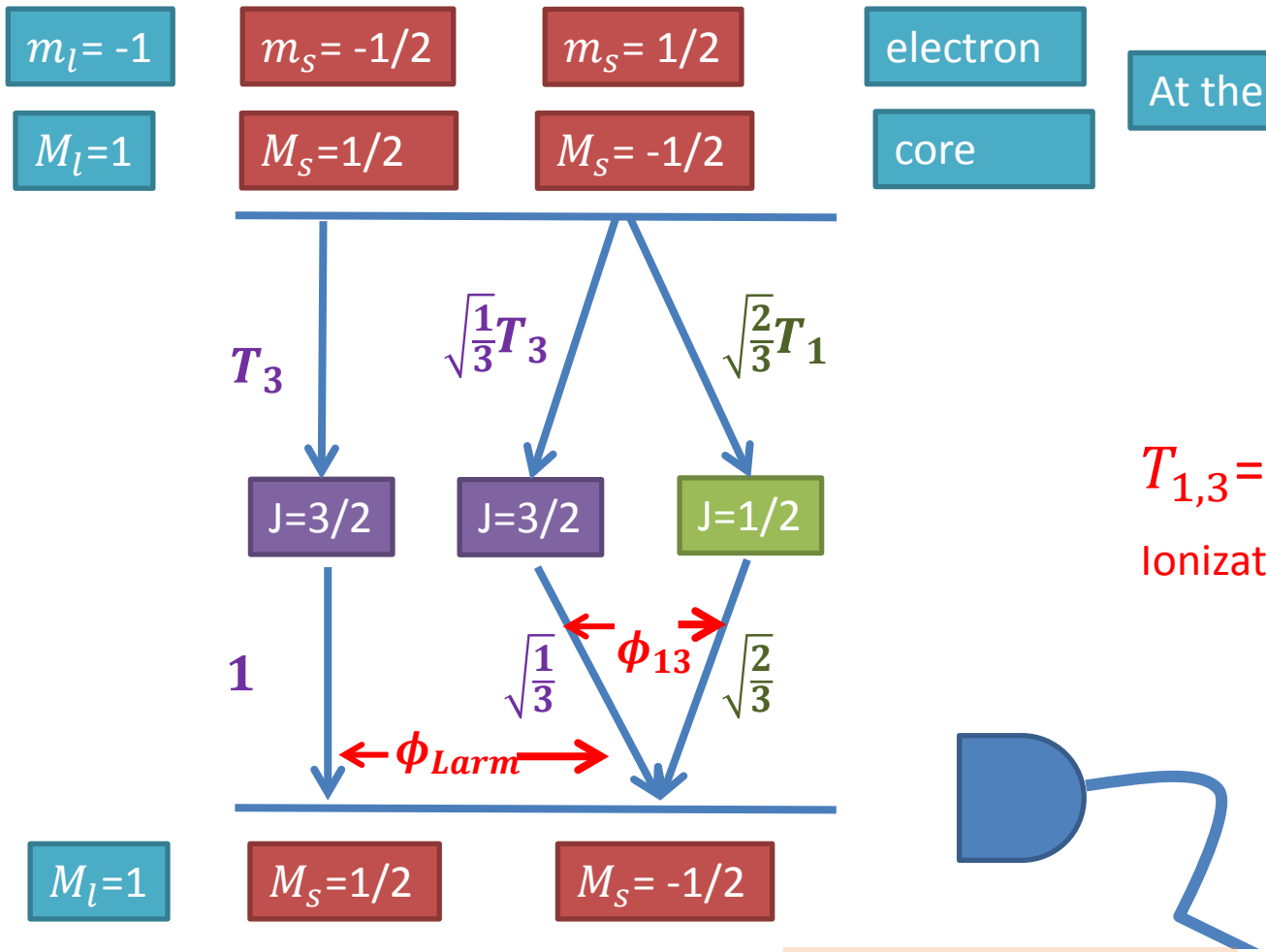
Closed shell, no Spin-Orbit interaction

Open shell, Spin-Orbit interaction is on

Ionization turns on the clock in Kr^+

Clock operates on core states: $P_{3/2}$ ($4p^5$,J=3/2) and $P_{1/2}$ ($4p^5$,J=1/2)

SO Larmor clock operating on the core



At the moment of separation

$T_{1,3} = |T_{1,3}| e^{i\phi^T_{1,3}}$

Ionization amplitude

$$\tan\phi_{LARM} = \frac{\sin(\Delta E^h_{SO} t - \phi_{13})}{0.5|T_3|/|T_1| + \cos(\Delta E^h_{SO} t - \phi_{13})}$$

The SFI Time

- One photon, weak field

$$\tan\phi_{LARM} = -\frac{\sin(\tau_{WS}\Delta E^{e}_{SO})}{0.5|R_3|/|R_1| + \cos(\tau_{WS}\Delta E^{e}_{SO})}$$

- Many photons, strong field

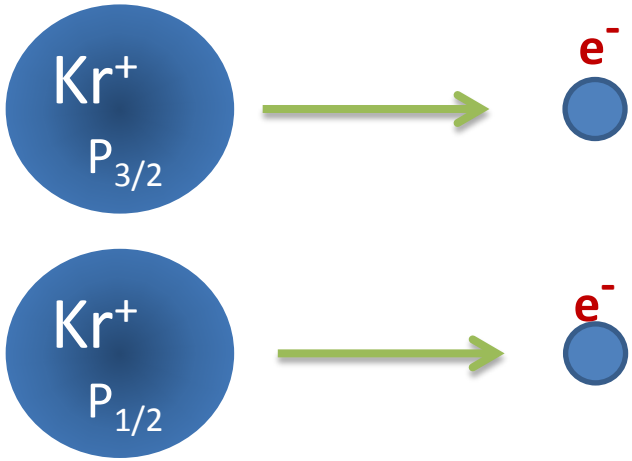
$$\tan\phi_{LARM} = \frac{\sin(\Delta E^{h}_{SO}t - \phi_{13})}{0.5|T_3|/|T_1| + \cos(\Delta E^{h}_{SO}t - \phi_{13})}$$

$$\phi_{13} = \phi^{T}_{1} - \phi^{T}_{3}$$

- Looks like a direct analogue of $\tau_{WS}\Delta E_{SO}$

- Does $\phi_{13}/\Delta E_{SO}$ correspond to time?

The appearance of SFI time



$$\phi_3 = \phi_c(I_p) + \phi_{3,SR}$$

$$\phi_1 = \phi_c(I_p + \Delta E_{SO}) + \phi_{1,SR}$$

$$\phi_{13} = \underbrace{\phi_c(I_p + \Delta E_{SO}) - \phi_c(I_p)}_{\frac{\partial \phi_c}{\partial I_p}\Delta E_{SO}} + \Delta\phi_{13,SR}$$

$$\tau_{SFI} = \frac{\partial \phi_c}{\partial I_p}$$

- Part of ϕ_{13} due to the common U yields Strong Field Ionization time
- Part of ϕ_{13} due to the different U_{SR} is a trace of e-h entanglement

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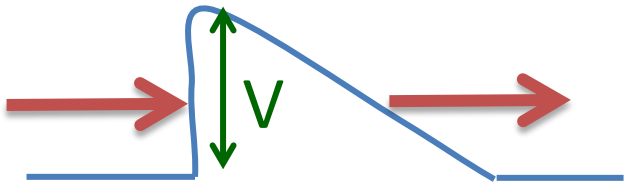
Strong-field ionization time & tunnelling time

Larmor tunneling time :

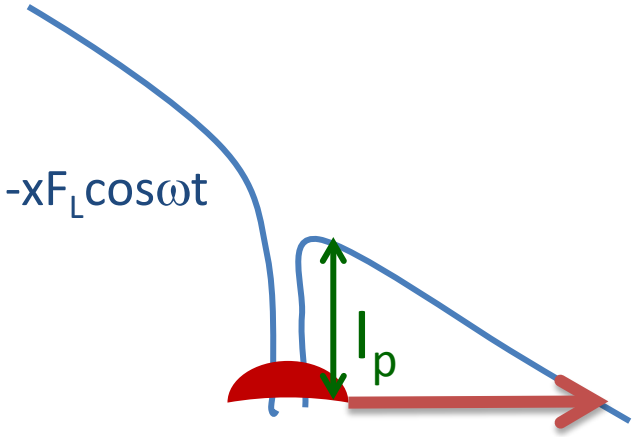
$$\tau_L = \frac{\partial \phi_c}{\partial V}$$



Hauge,E. H. et al, Rev. Mod Phys, 61, 917 (1989)

SFI time:

$$\tau_{SFI} = \frac{\partial \phi_c}{\partial I_p}$$



- Derivation has never relied on the tunnelling picture
- SFI time is consistent with the Larmor time for tunnelling through a static barrier of height $V=I_p$

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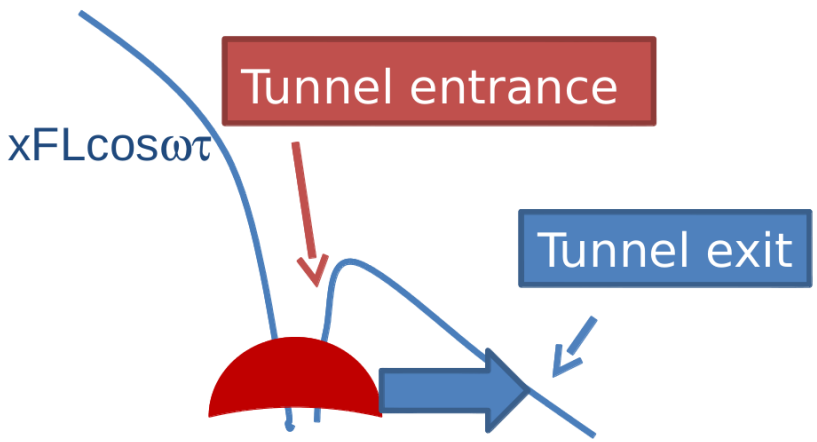
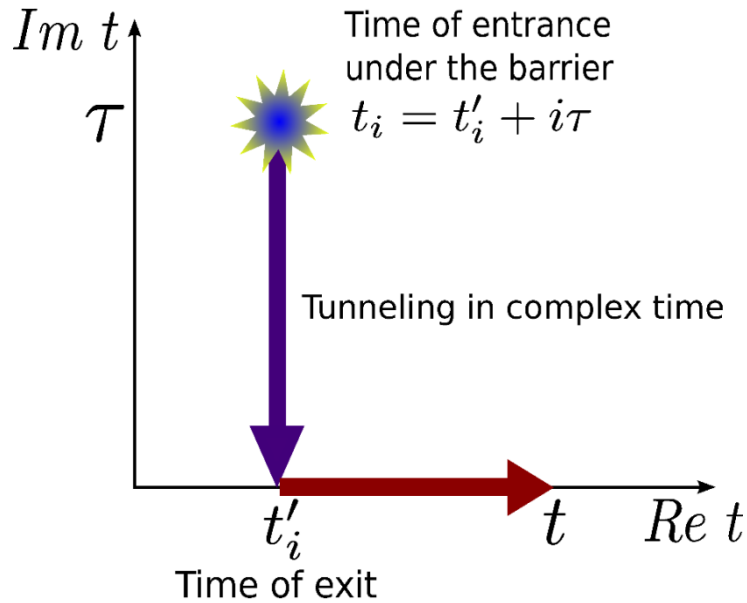
Strong-field ionization time & semiclassical time





Key result:

$$\tau_{SFI} = \frac{\partial \phi_c}{\partial I_p}$$

Coincides with the semiclassical 'exit time', if the semiclassical approximation is used for ϕ_c

- Can we measure the SFI time τ_{SFI}?

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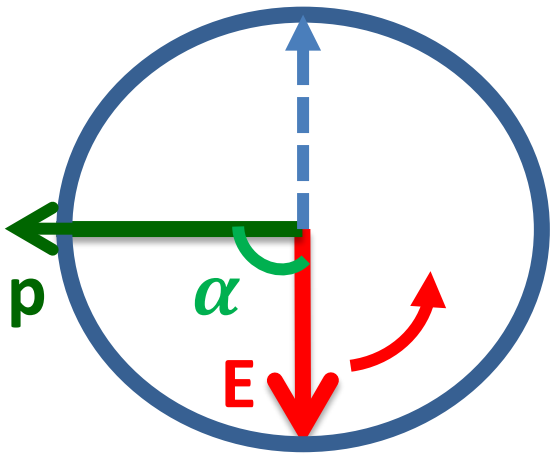
Ionization time measurements with Atto-clock

Atto-clock principle : ionization in strong circular IR fields maps ionization *time* on the electron detection *angle*

(U. Keller et al)

Example: Short-range potential

$$\alpha = \pi/2$$



- Electron is detected with final **p** ⊥ **E** at the ionization time t_{ion}
- Observable: angle between **p** and $\mathbf{E}_{max}$ for a 'single-cycle' pulse

SFI time and Atto-clock measurements

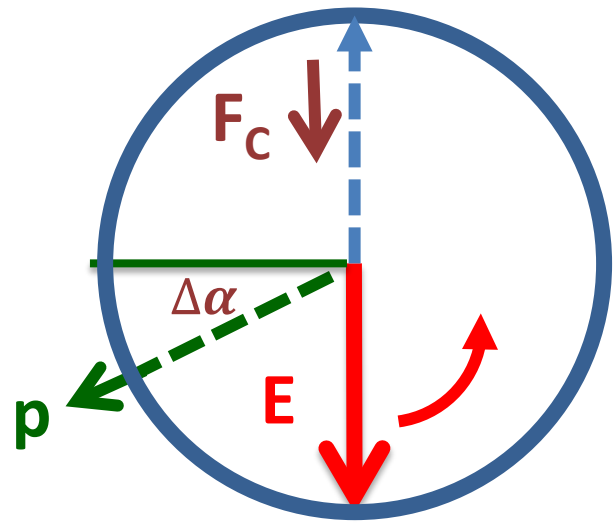
Ionization from a **long-range** potential: $\Delta\alpha$

$\Delta\alpha$ can be converted into time

$$\Delta t_{AC} = \frac{\Delta\alpha}{\omega}$$

Key result:

$$\frac{\Delta\alpha}{\omega} = \frac{\partial\phi}{\partial I_p}$$

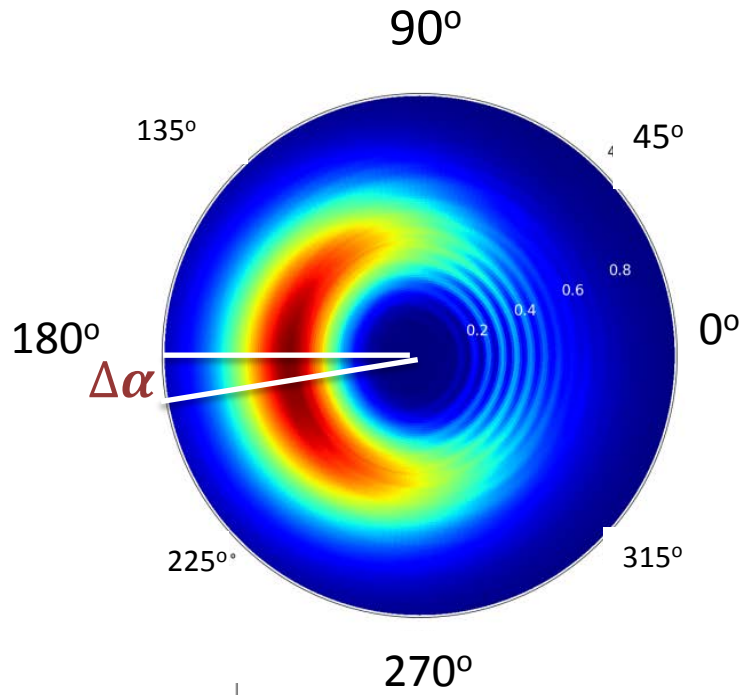


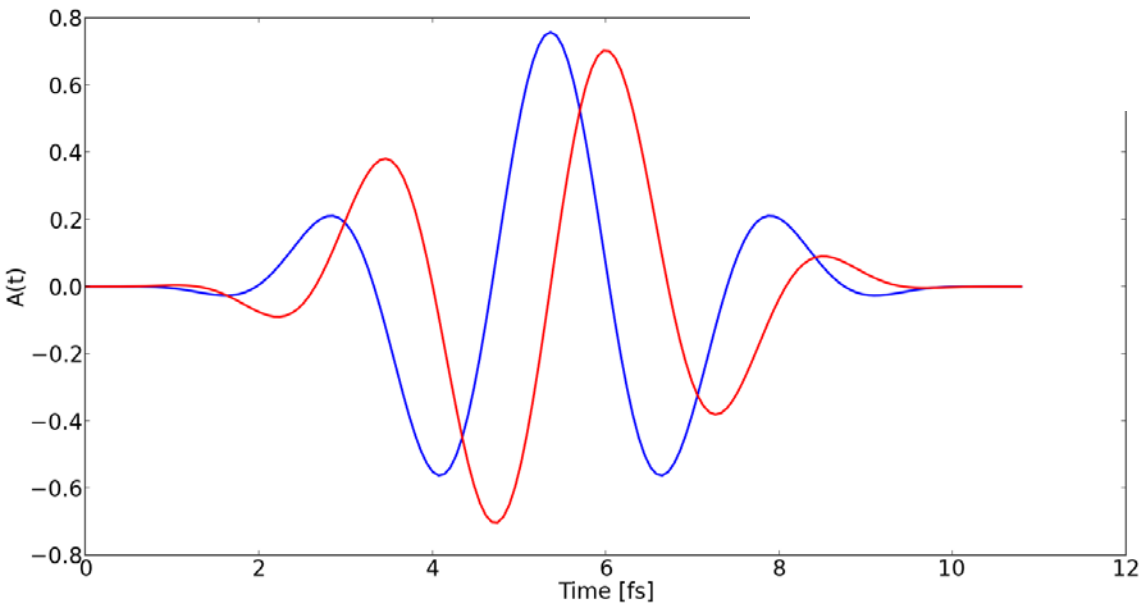
- Attoclock measures the SFI time that we have introduced.
  - ϕ is the total phase.

- Is there a phase accumulated under the barrier? Our analytical theory says 'no'
- Let us compare with numerical experiment

Atto-clock ‘measurements’: TDSE

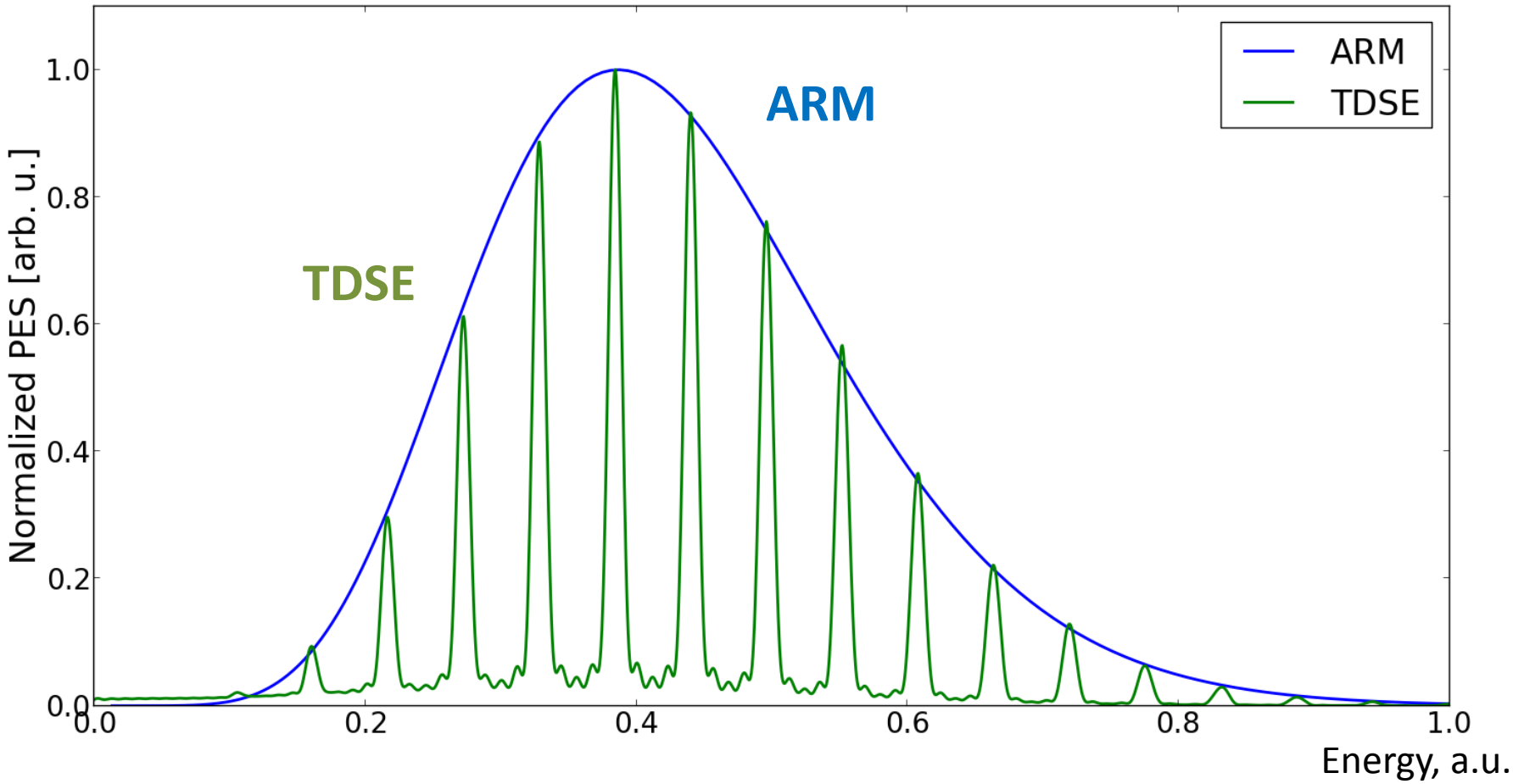
H-atom,
λ=800 nm,
Circular polarization
FWHM =2.8 fsec (just over one cycle)





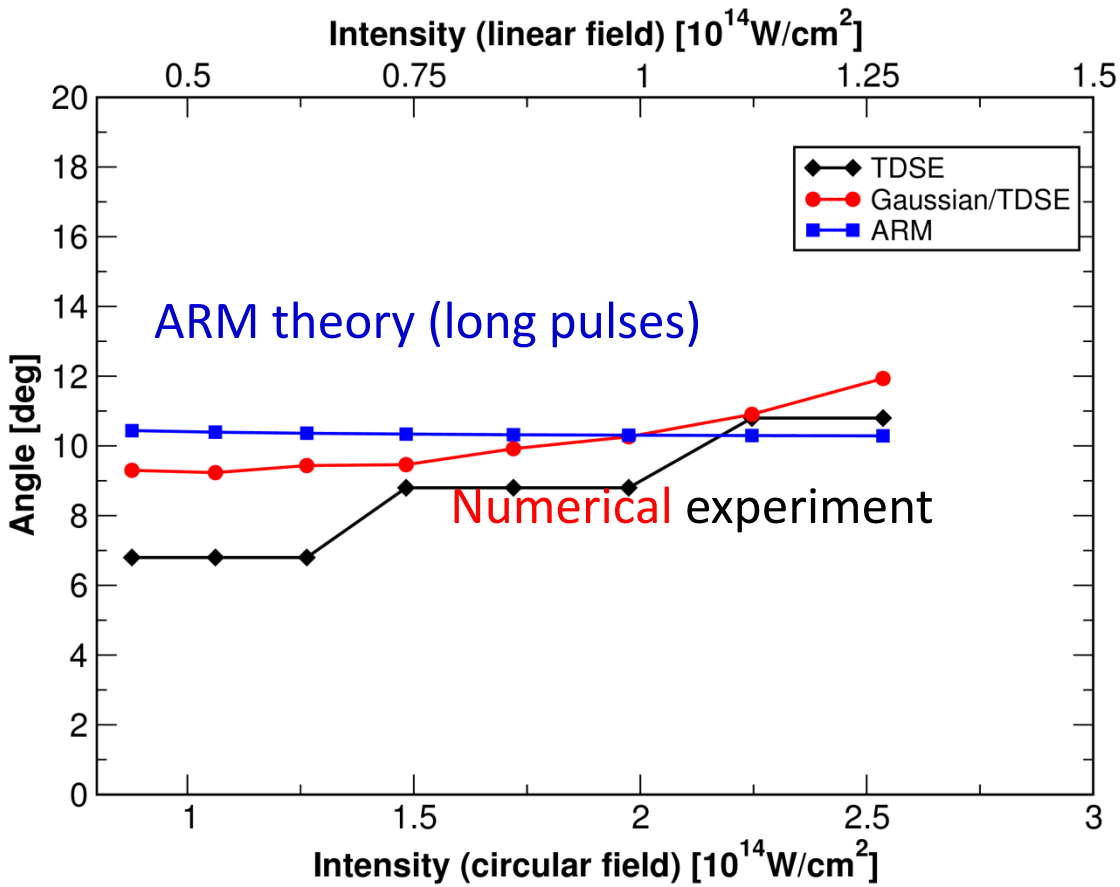
ARM vs TDSE (long pulse)

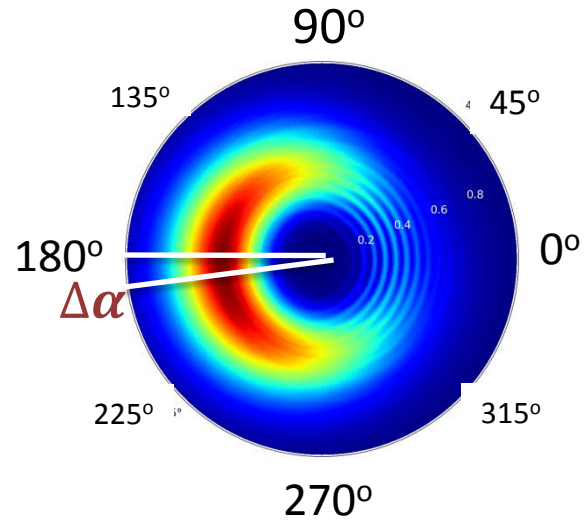
H-atom,
λ=800 nm,
Circular polarization
(6 cycles flat top)



Atto-clock measurement of SFI time

Angle $\Delta\alpha$



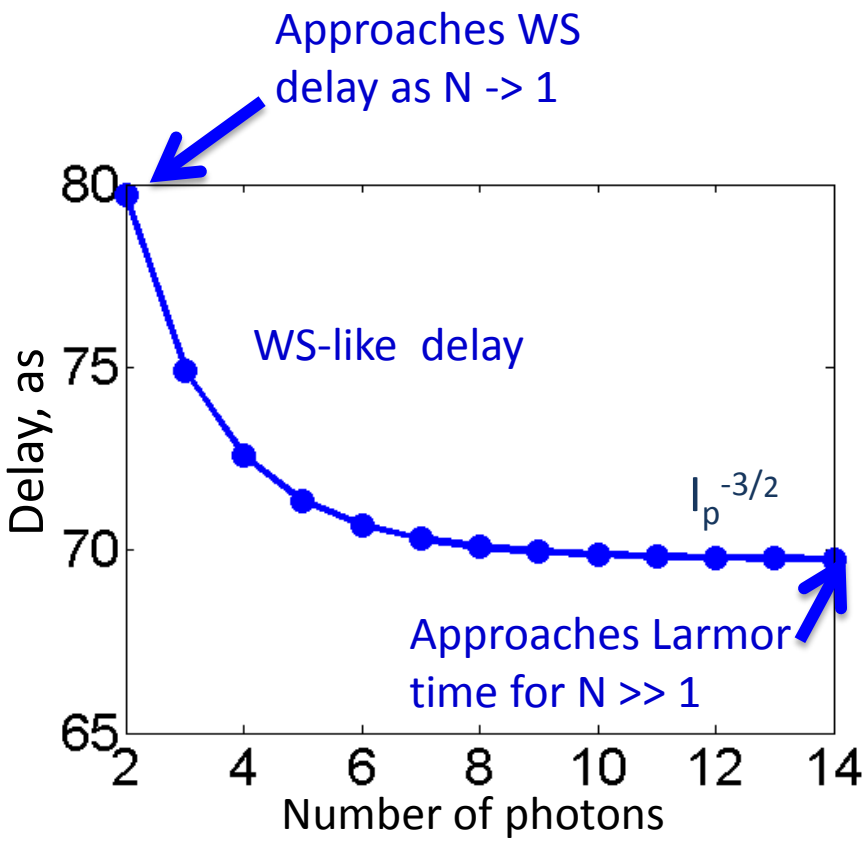


H-atom, 800 nm,
Circular polarization
Sin^2 pulse envelope,
4 cycles base to base

Our results do not show tunnelling delays for this case.

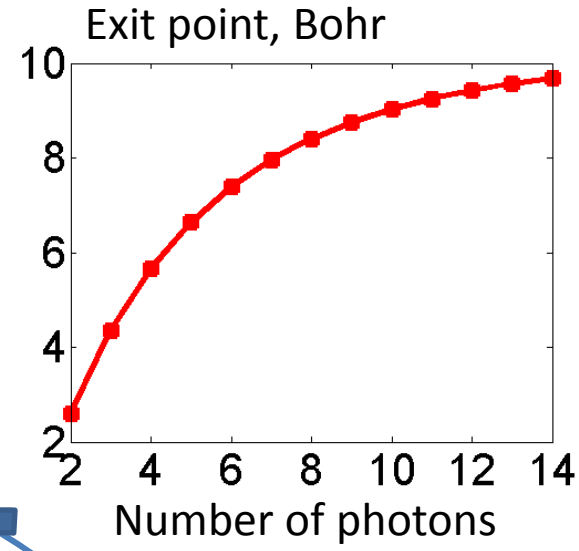
How does ionization time depend on N photons?

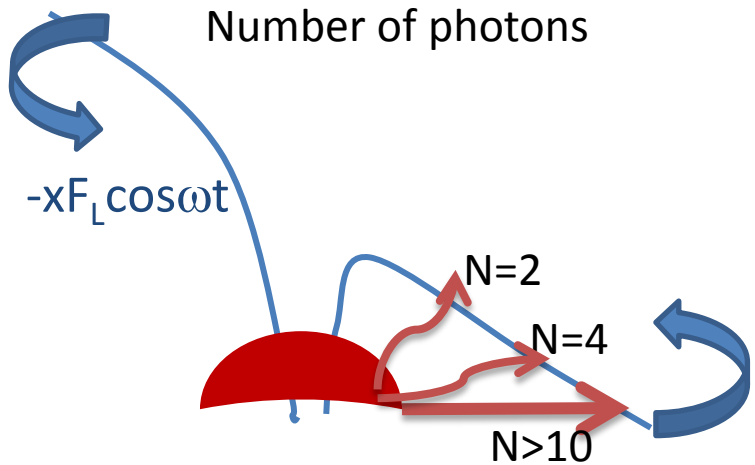
Delays : Results and physical picture



Kr atom:
Ip=14 eV

Circular field:
$2.5 \times 10^{14} W/cm^2$





- Phase and delays are accumulated after exiting the barrier
- Larger N – more adiabatic, exit further out

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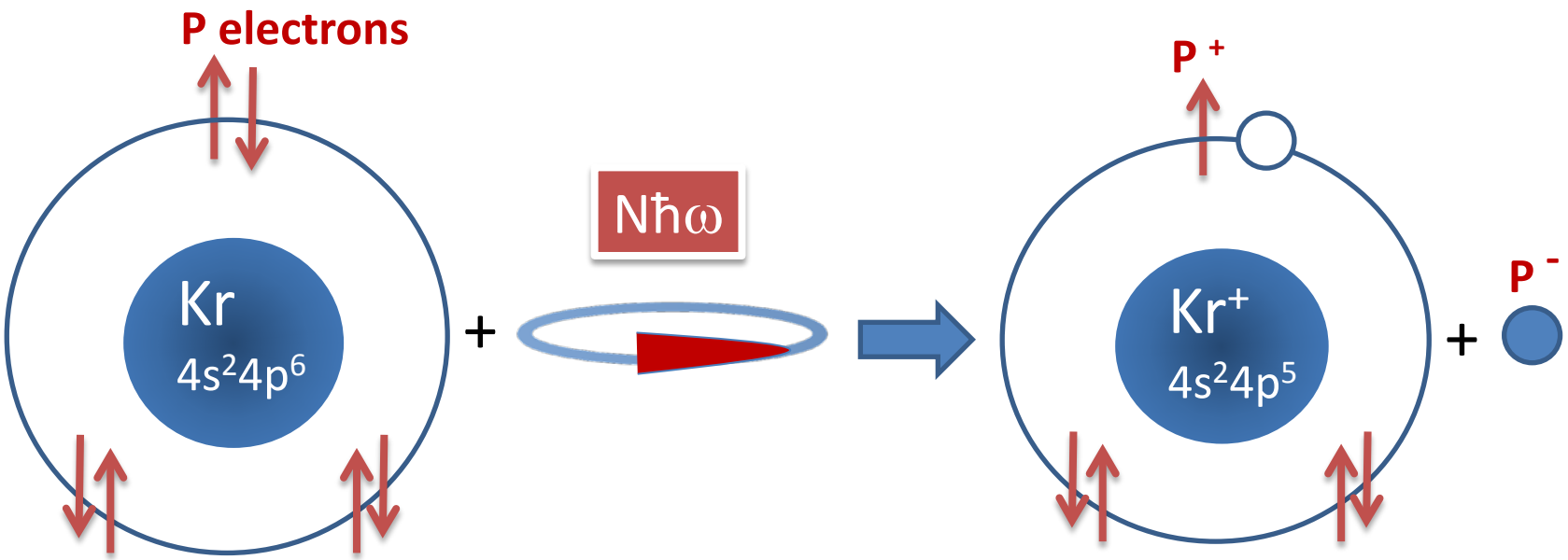
Ionization times in transient absorption

Experiment: use a few cycle circularly polarized IR pulse as a pump



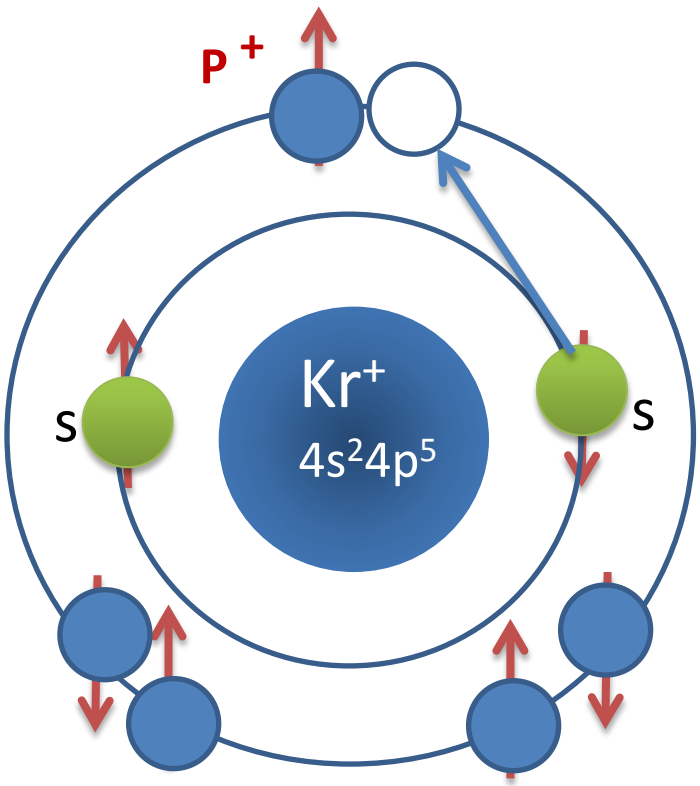
Closed shell, no Spin-Orbit interaction

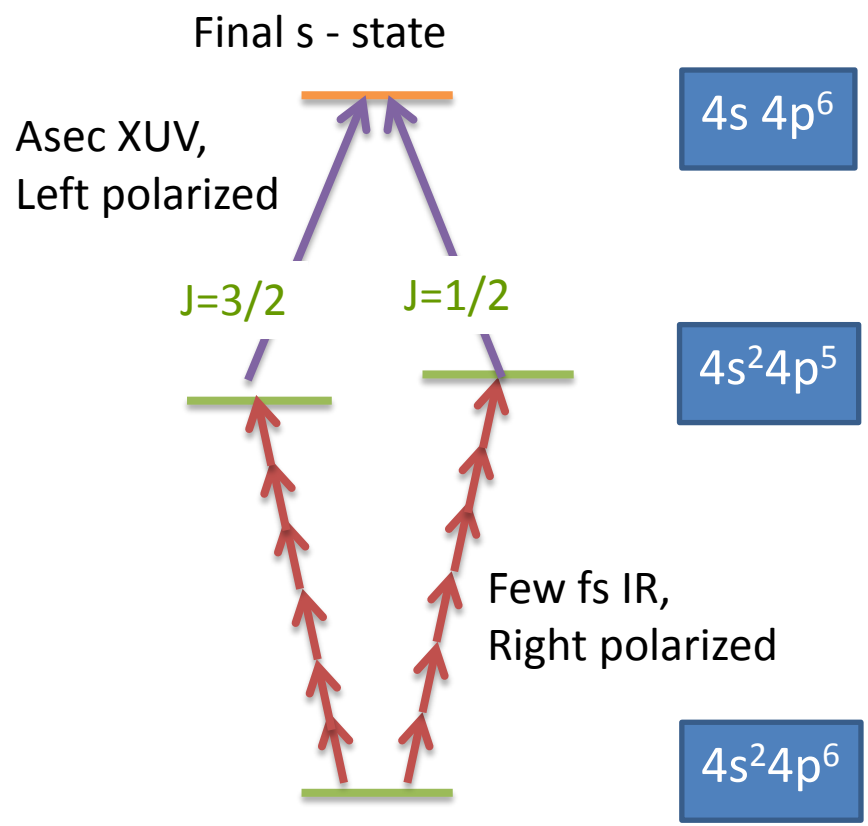
Open shell, Spin-Orbit interaction is on

Ionization turns on the clock in Kr^+

Clock operates on core states: $P_{3/2}$ ($4p^5$,J=3/2) and $P_{1/2}$ ($4p^5$,J=1/2)

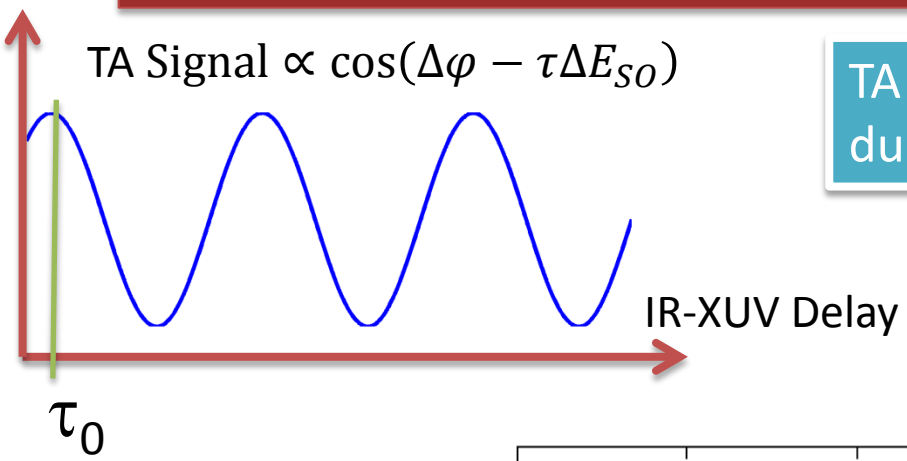
Measuring ionization times with transient absorption



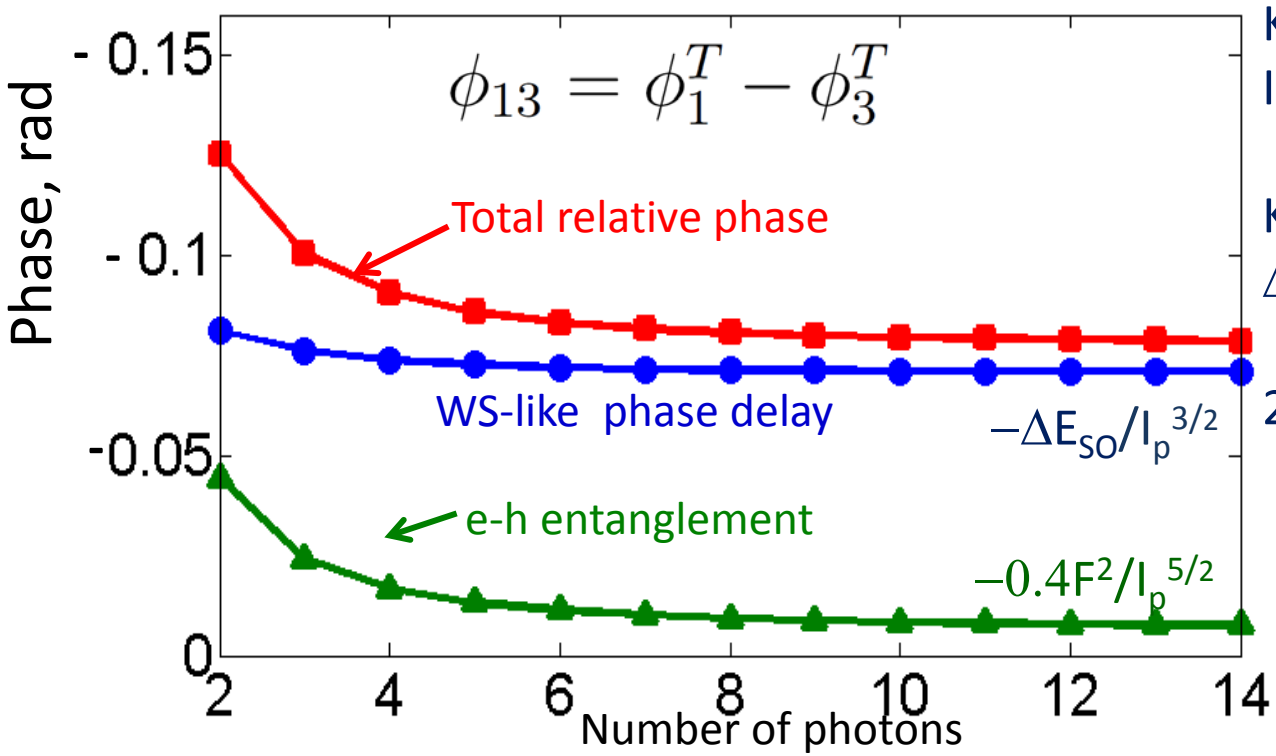


- Pump: Few fs IR creates p-hole and starts the clock
- Probe: Asec XUV pulse fills the p-hole and stops the clock
- Observe: Read the attosecond clock using transient absorption measurement

Transient absorption measurements



TA signal shows the phase difference $\Delta\varphi$ due to ionization into J=3/2 and J=1/2



Kr atom:
Ip=14 eV

Kr^+
ΔE_{SO}=0.67 eV

2.5×10^{14}W/cm^2

The Roadmap

- Defining ionization time: from one to many photons
  - The Spin-Orbit Larmor clock for ionization
  - Connection to Larmor time for tunnelling
  - Connection to quantum orbits and WKB analysis

- Measuring strong-field ionization times
  - The atto-clock setup
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  - The high harmonic spectroscopy setup
  - Connection between measurement protocols

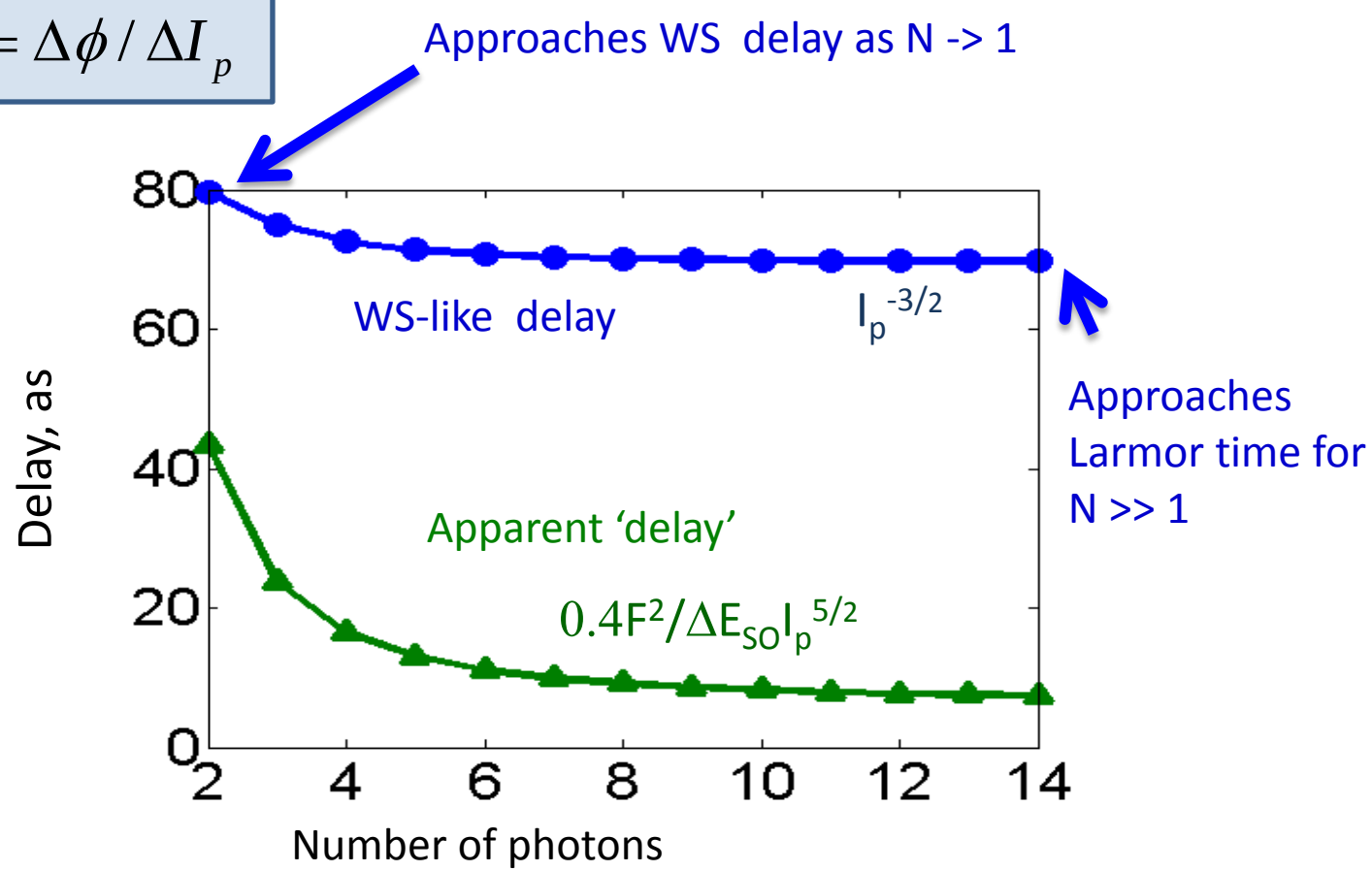
Delays : pump-probe vs Atto-clock

$$\tau_0 = \Delta\phi / \Delta E_{SO} = \Delta\phi / \Delta I_p$$

Kr atom:
Ip=14 eV

Kr^+
ΔE_{SO}=0.67 eV

Circular field:
$2.5 \times 10^{14} W/cm^2$



- 'Apparent delay' shows-up in Transient Absorption measurement
- 'Apparent delay' is due to electron –hole entanglement
- 'Apparent delay' is not present in Attoclock measurement

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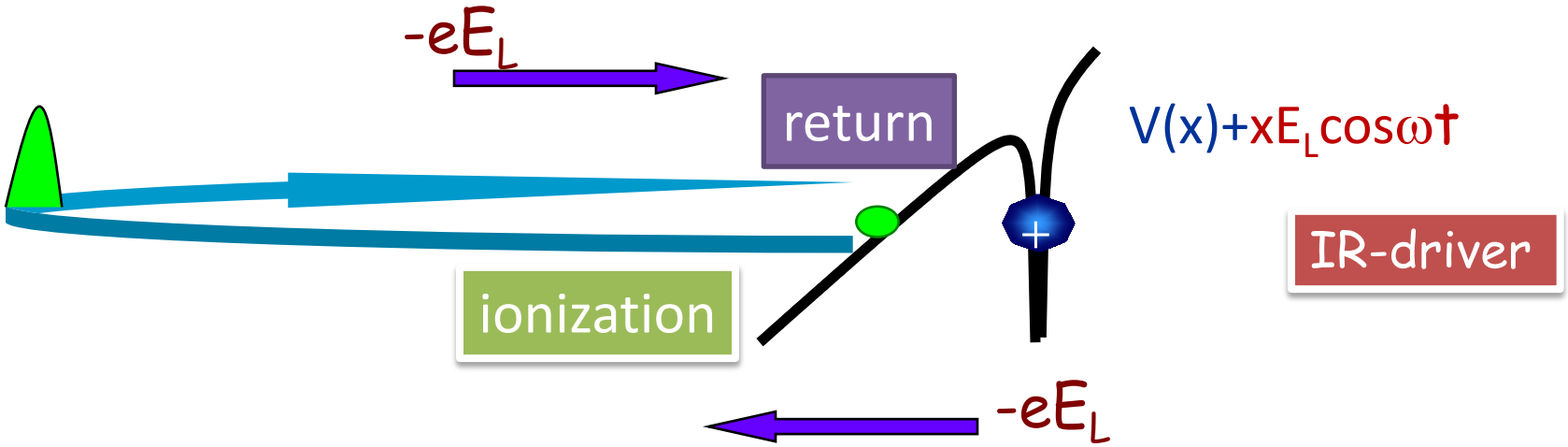
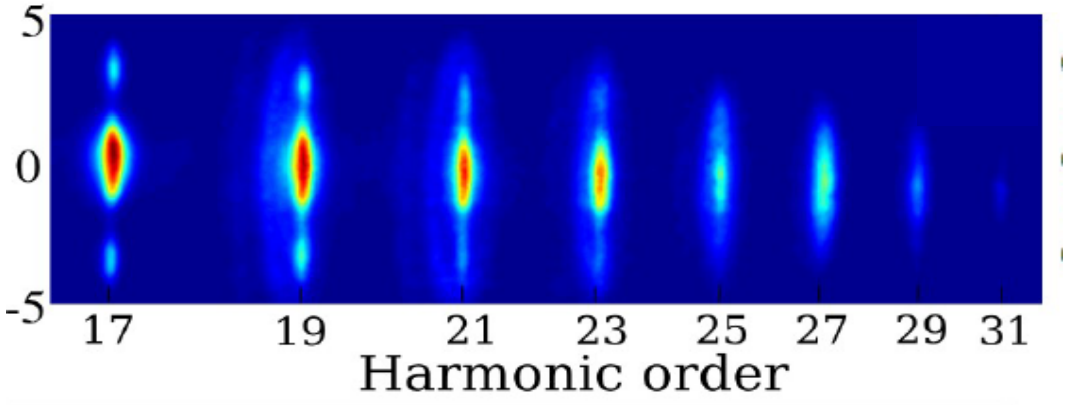
High harmonic spectroscopy of ionization times

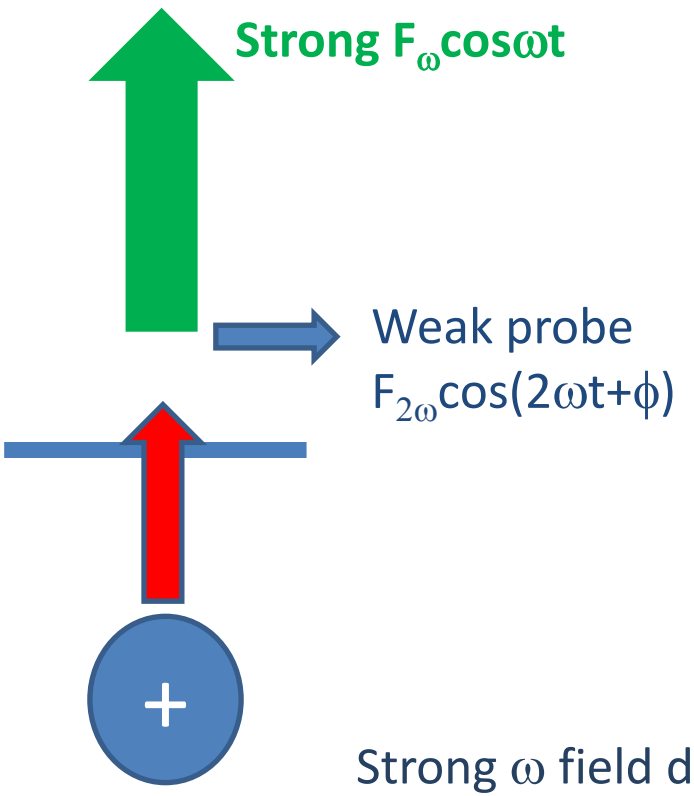


Oscillating field brings the electron back

Electron-ion recombination produces emission: high harmonics of IR



Measuring ionization times: Key idea



$$\Delta v_\perp(t,t_i) = -\int_{t_i}^{t} F_{2\omega}(t')dt' = A_{2\omega}(t) - A_{2\omega}(t_i)$$

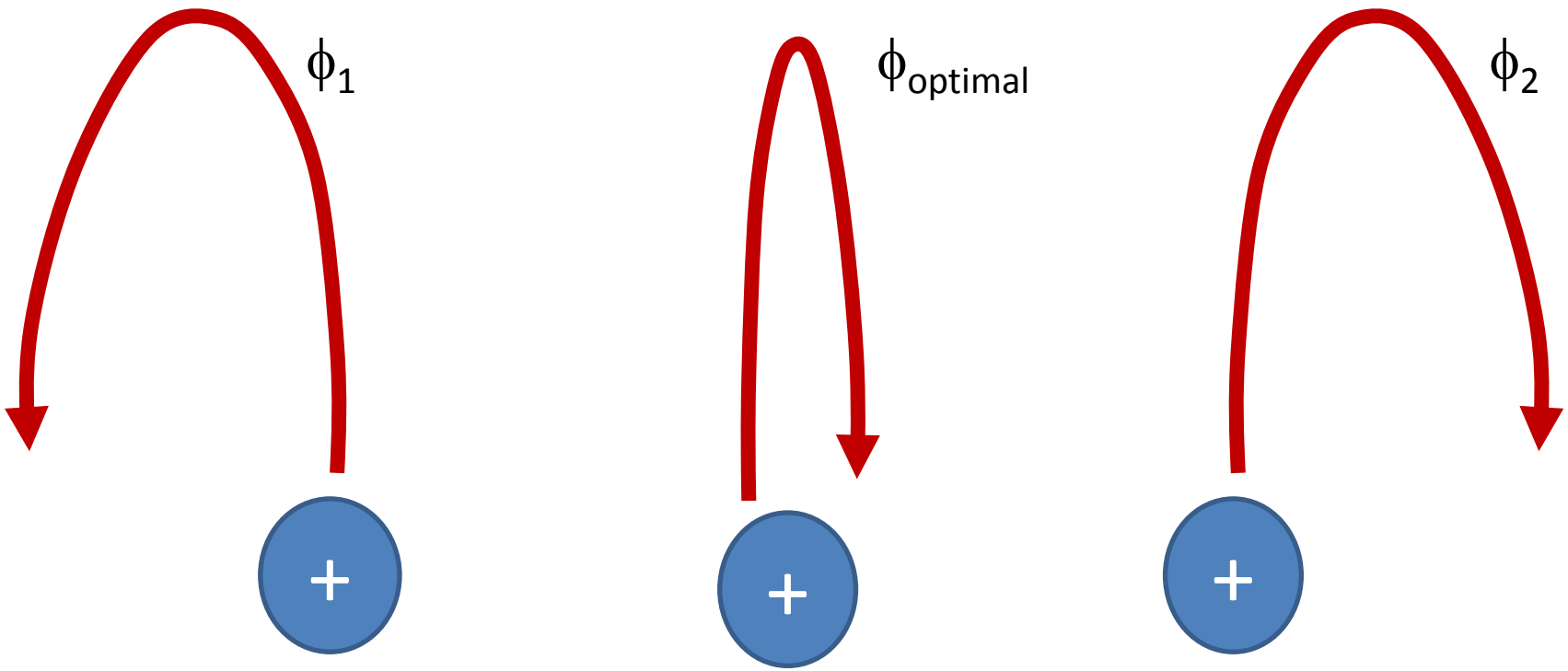
$$\Delta r_\perp(t,t_i) = \int_{t_i}^{t} \Delta v_\perp(t',t_i)dt'$$

Strong ω field drives tunnelling
Weak 2ω tags the electron
Dependence of Δv and Δr on ϕ encodes ionization times

How can we measure Δv and Δr ? –
where to find two independent observables?

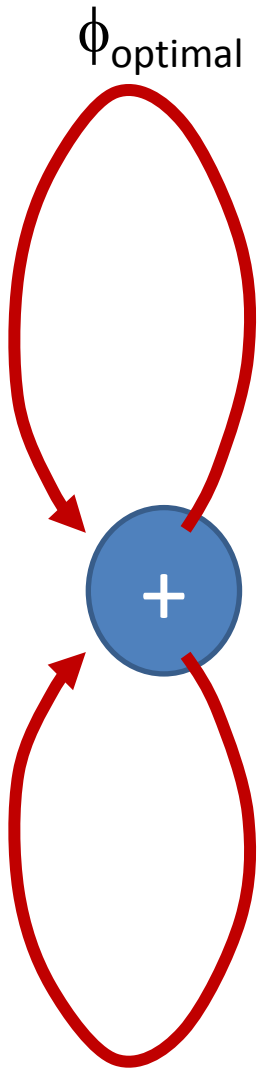
Measuring position shifts: 2D High harmonic spectroscopy

Oscillating field brings the electron back – harmonic emission
Parent ion is a perfect measurement device



Odd harmonic intensities maximize for minimal Δr (ϕ)
Odd harmonics measure Δr (ϕ)

Measuring velocity shifts: 2D High harmonic spectroscopy



Presence of 2ω leads to even harmonics

Harmonic emission means that electron has returned

Even harmonic intensity maximizes for maximal asymmetry between two subsequent half-cycles

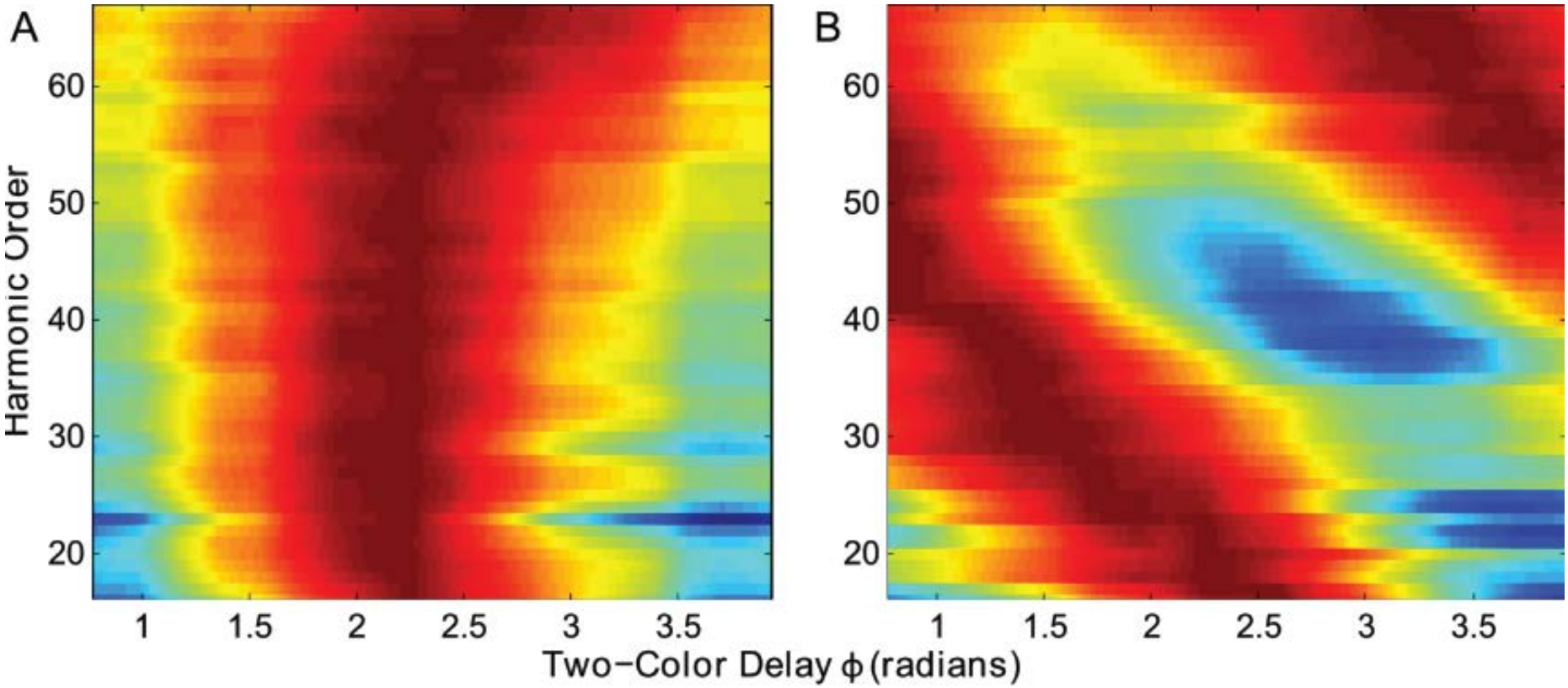
Maximal asymmetry means maximal Δv (ϕ)

Even harmonics measure Δv (ϕ)

Measurements in Helium

Helium, 40 fsec pulse, 800 nm at 4 10^{14} W/cm^2, 400 nm at 1-2% intensity level

Odd harmonics **Even harmonics**

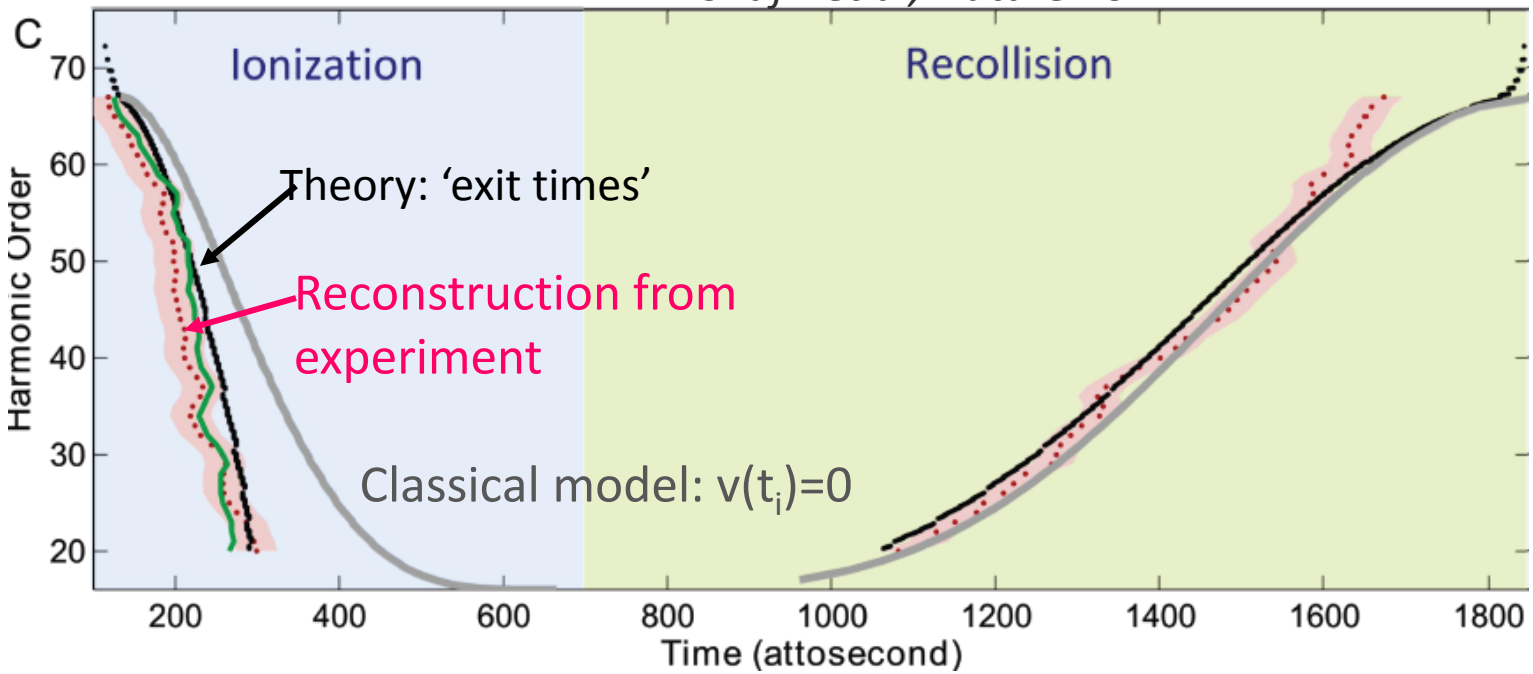


D. Shafir, H. Soifer, B. Bruner, S. Patchkovskii, Y. Mairesse, M. Ivanov, O. Smirnova, N. Dudovich, Nature (2012)

Results of reconstruction in Helium

Helium, 40 fsec pulse, 800 nm at 4 10^{14} W/cm^2, 400 nm at 1-2% intensity level

D. Shafir et al, Nature 2012



Results are:

- in agreement with the 'exit times'
- consistent with the theory predicting no tunnelling delay, within < 30 asec accuracy

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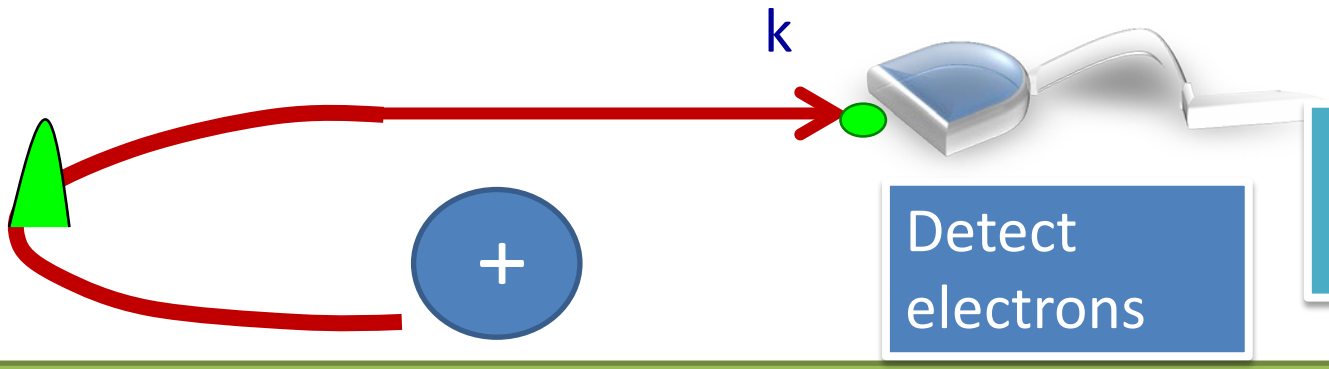
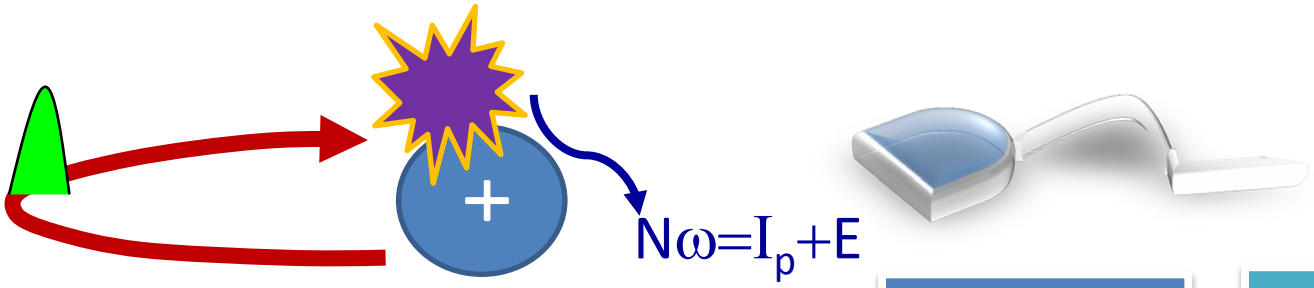
Ionization time measurements: Attoclock vs HHG



Detect electrons

Electron drift momentum must be real

Photoelectron spectroscopy - PES (Attoclock in circular fields)



HHG spectroscopy

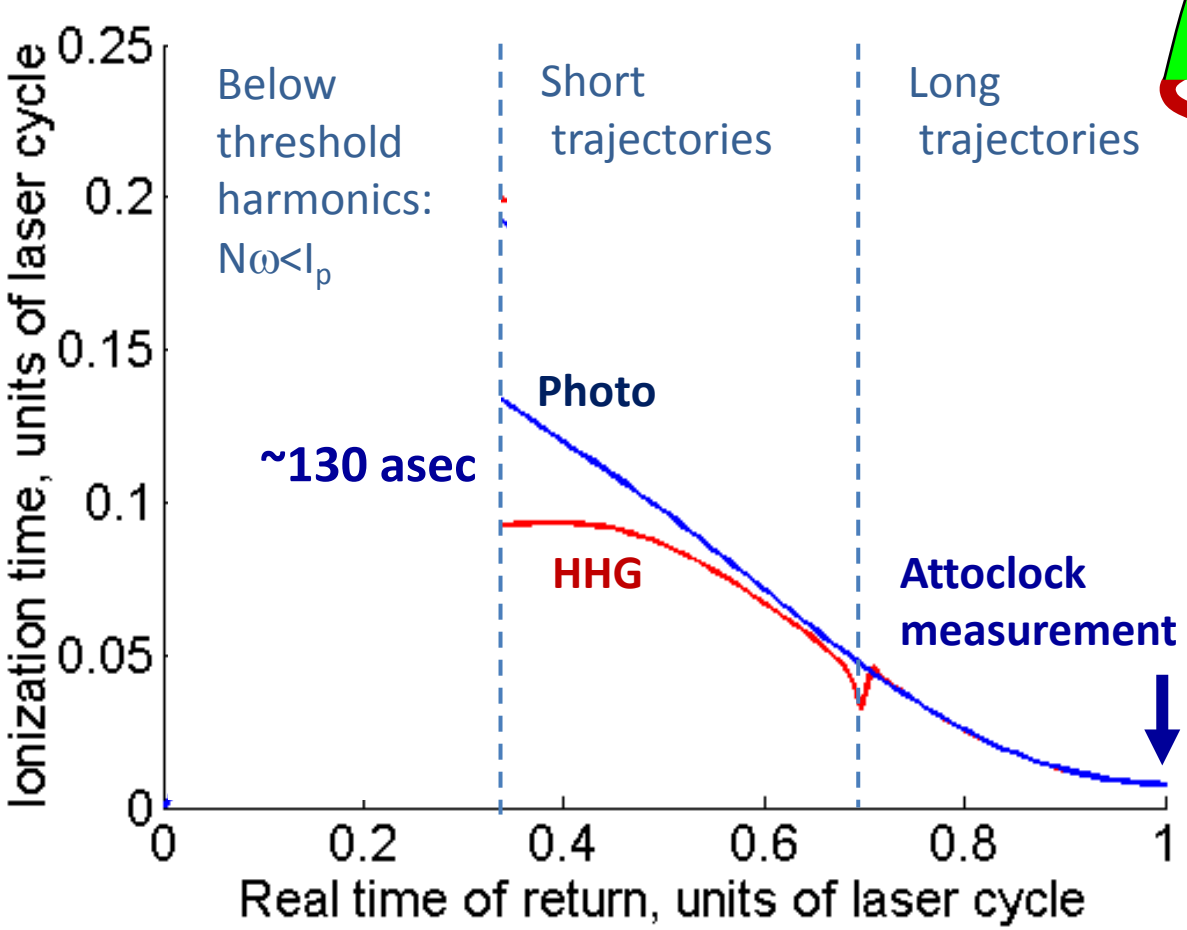
Detect photons

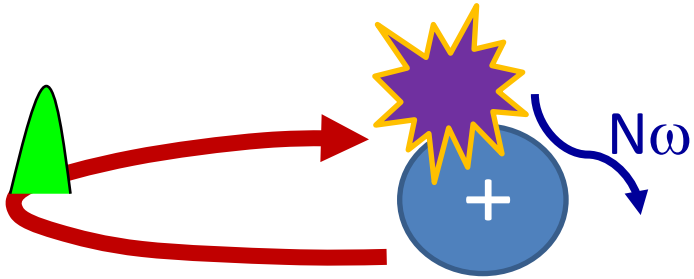
Electron energy is real, But its *drift momentum* may be complex

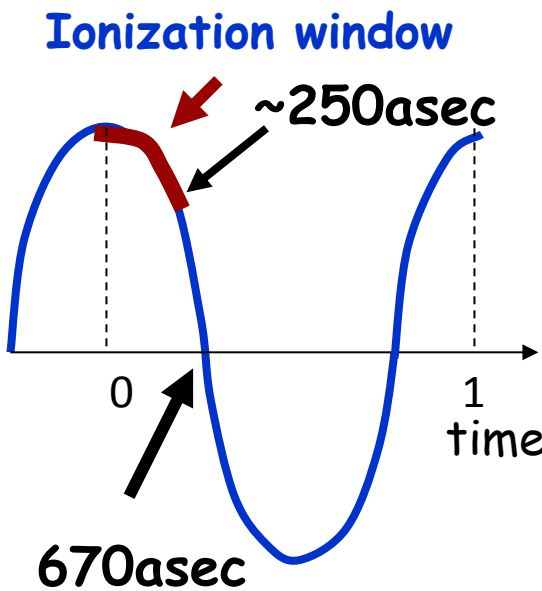
Do they measure same ionization times?

Ionization time measurements with HHG

Example: Short-range potential







For long trajectories ionization times are the same in HHG and PES

Conclusions

- Using SO Larmor clock we defined delays in hole formation:

- Actual delay in formation of hole wave-packet
  - Larmor- and Wigner-Smith – like,
  - Applicable for any number of photons, any strong-field ionization regime
- Apparent 'delay' – trace of electron-hole entanglement:
  - Clock-imparted 'delay' (encodes electron – hole interaction)
  - Analogous to spread of an optical pulse due to group velocity dispersion
  - does not depend on clock period
- Absorbing many photons takes less time than absorbing few photons, but not zero

- The SO interaction has been used to derive τ_{SFI} but is not needed to measure it!

- Directly related to times measured in IR-pump-XUV-probe scheme
  (e.g. Transient absorption, tiny difference due to entanglement)
- Directly related to ionization times in HHG (tiny difference)
- Directly measured by Atto-clock