PHOTOIONIZATION DYNAMICS WITH ATTO-SECOND PULSE TRAINS

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Division of Atomic Physics

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Atto group in Lund

Dynamics in real time

To capture a moving object we need...

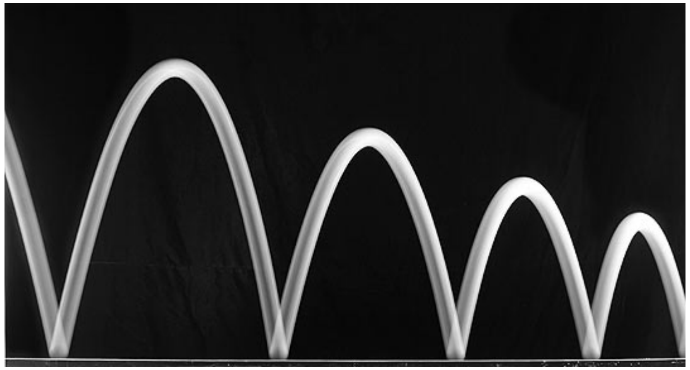
Camera with long exposure time

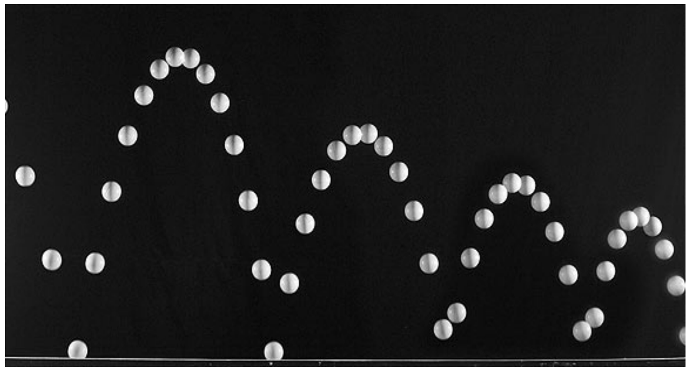
an exposure time /shutter faster than the motion !

Sequential approach

Dynamics in real time

Camera with long exposure time and a strobe light



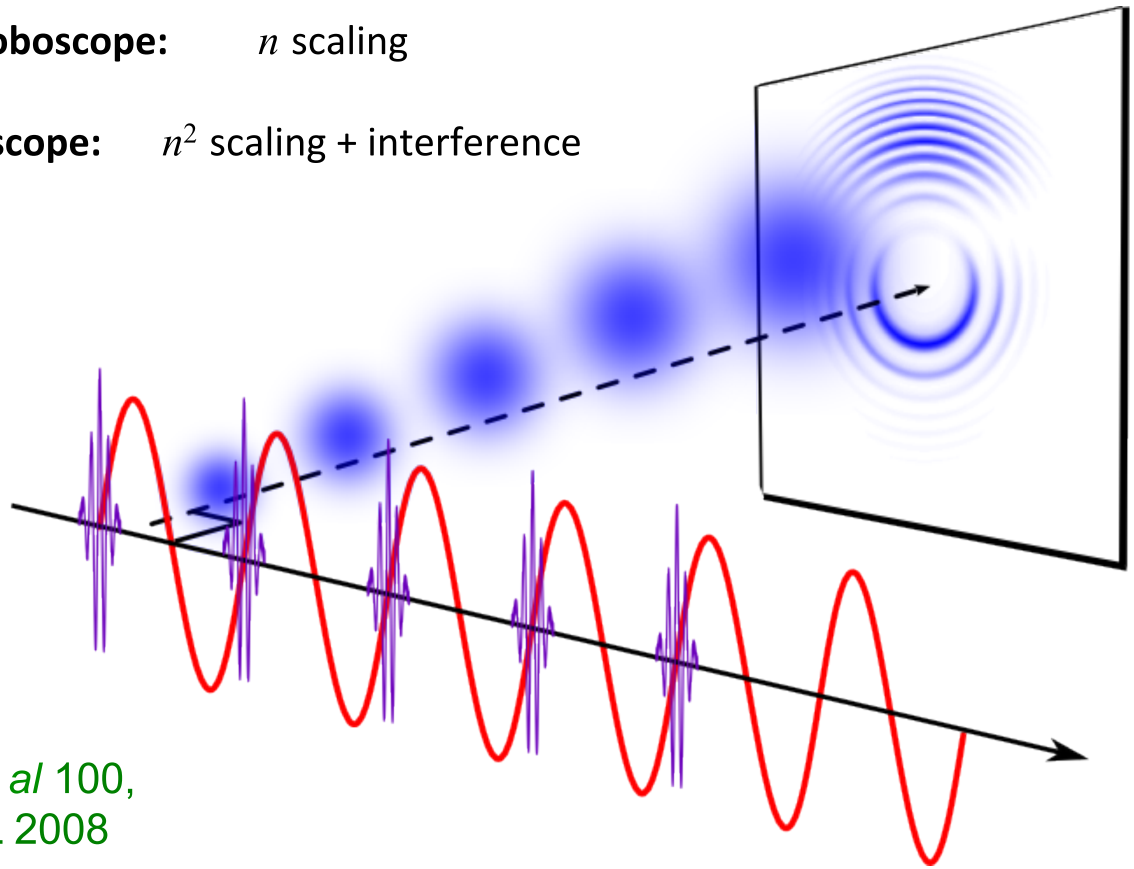


Stroboscope approach

The quantum stroboscope with Attosecond Pulse Trains (APTs)

Conventional stroboscope: n scaling

Quantum stroboscope: n^2 scaling + interference



Mauritsson *et al* 100, 073003 PRL 2008

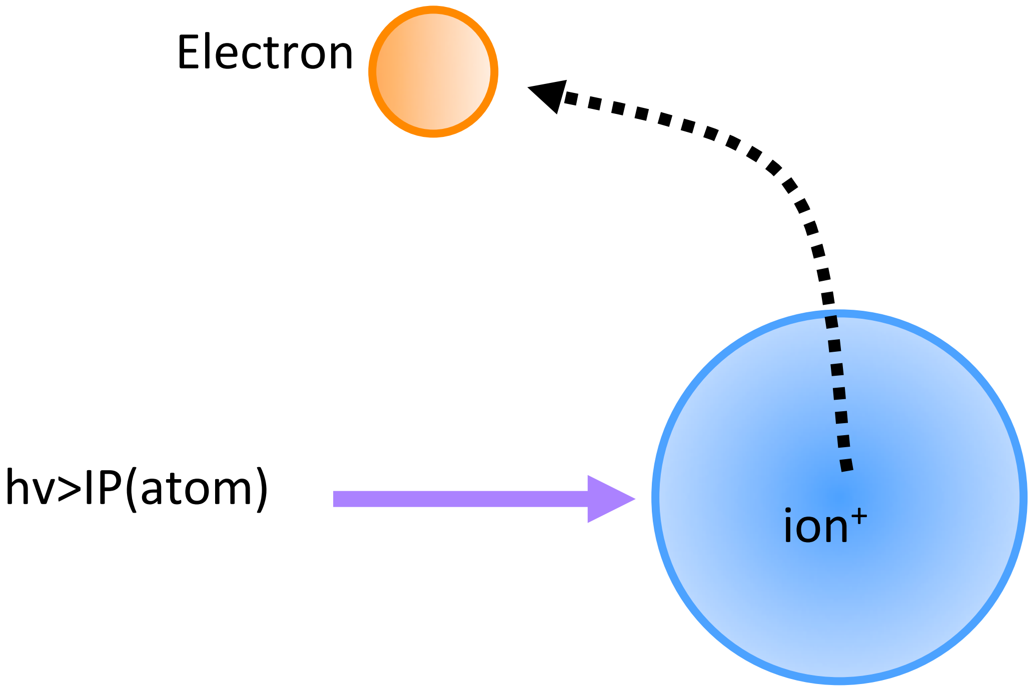
Applications

Atto-second Pulse Trains

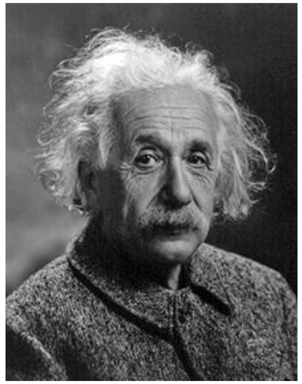
Weak IR field → Interference in the frequency domain

Strong IR Field → Interference in the time domain

Photo-ionization

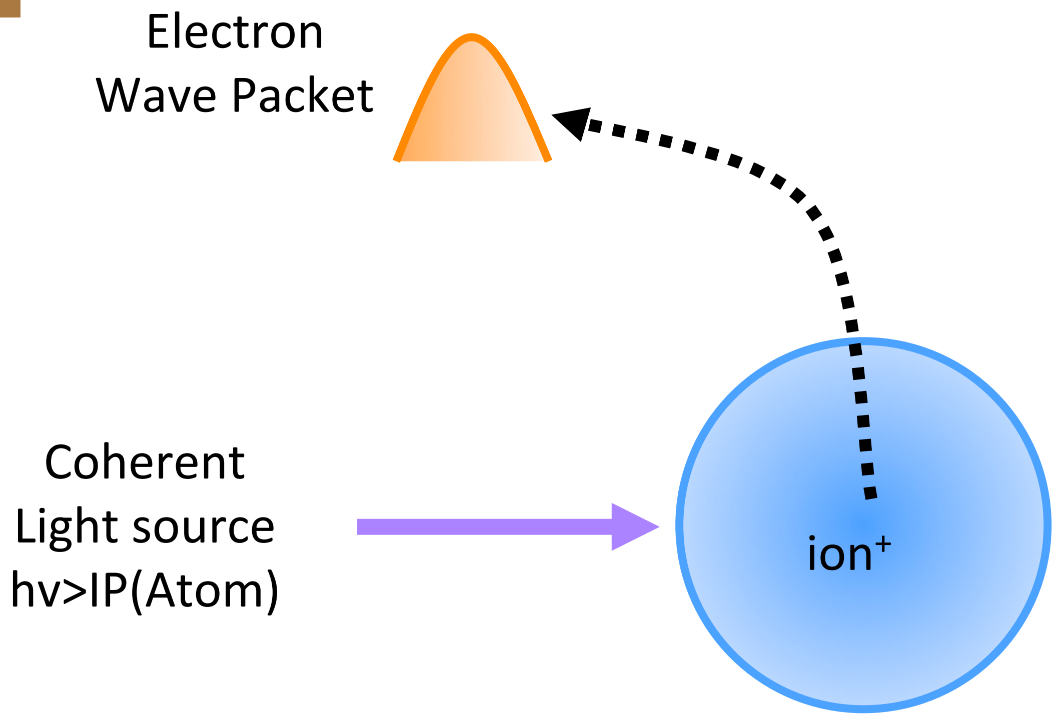


H.R Hertz - 1887



A. Einstein - 1905

Photo-ionization with APTs



Atto-second regime

$\Delta t<1fs \longleftrightarrow \Delta E>2$ eV

Wave-particle duality

Photo-ionization with APTs

Electron

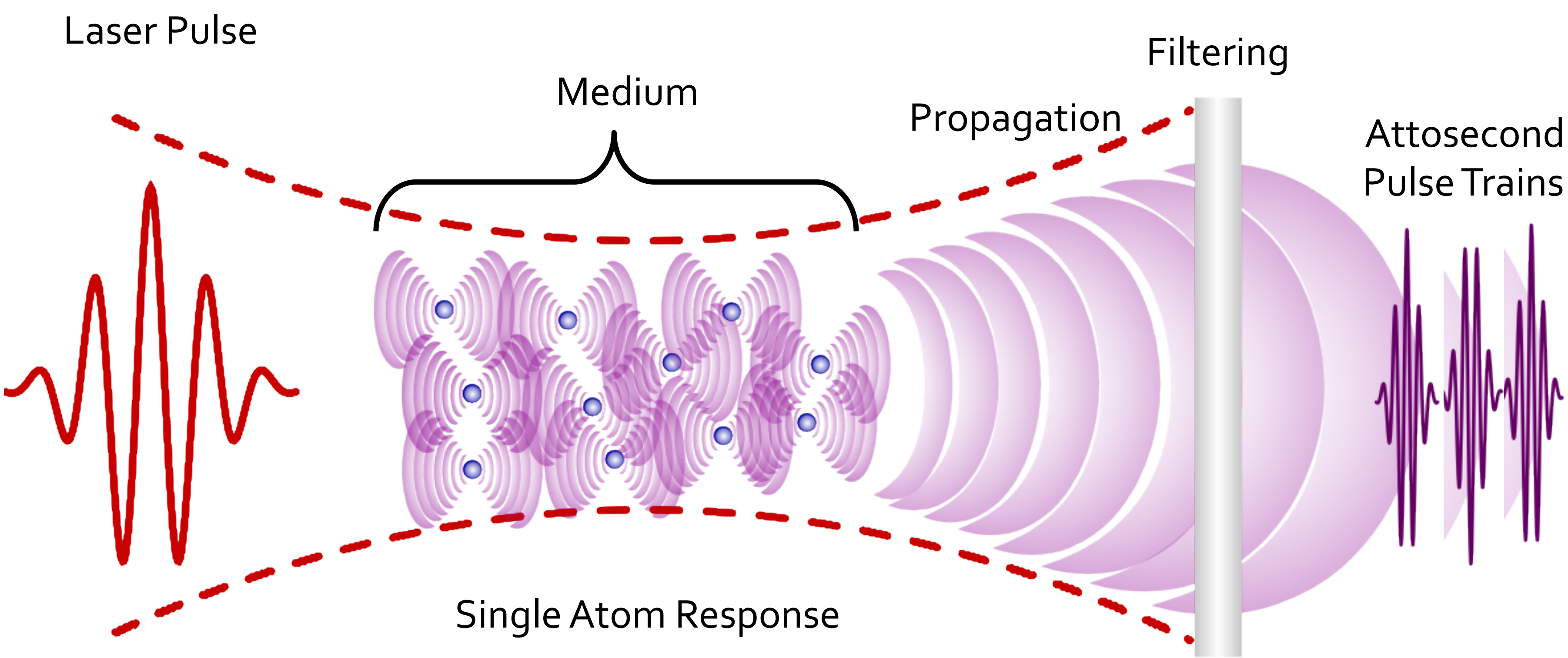
Ligh
hv>

Can we ”read-out” properties
of **one**-photon ionization
in the Electron Wave Packet ?

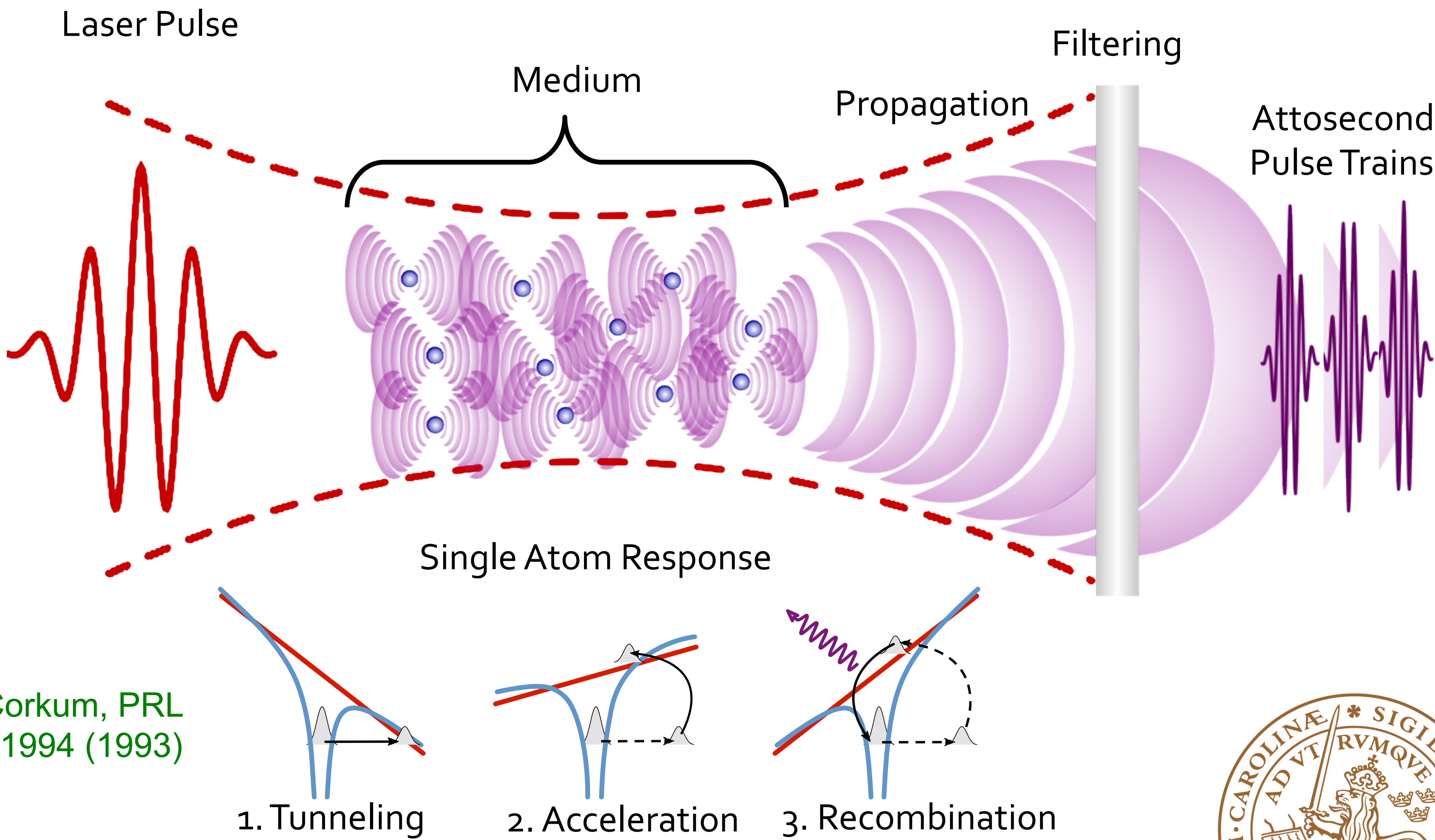
Wave-particle duality

TOOLS

Attosecond Pulse Trains

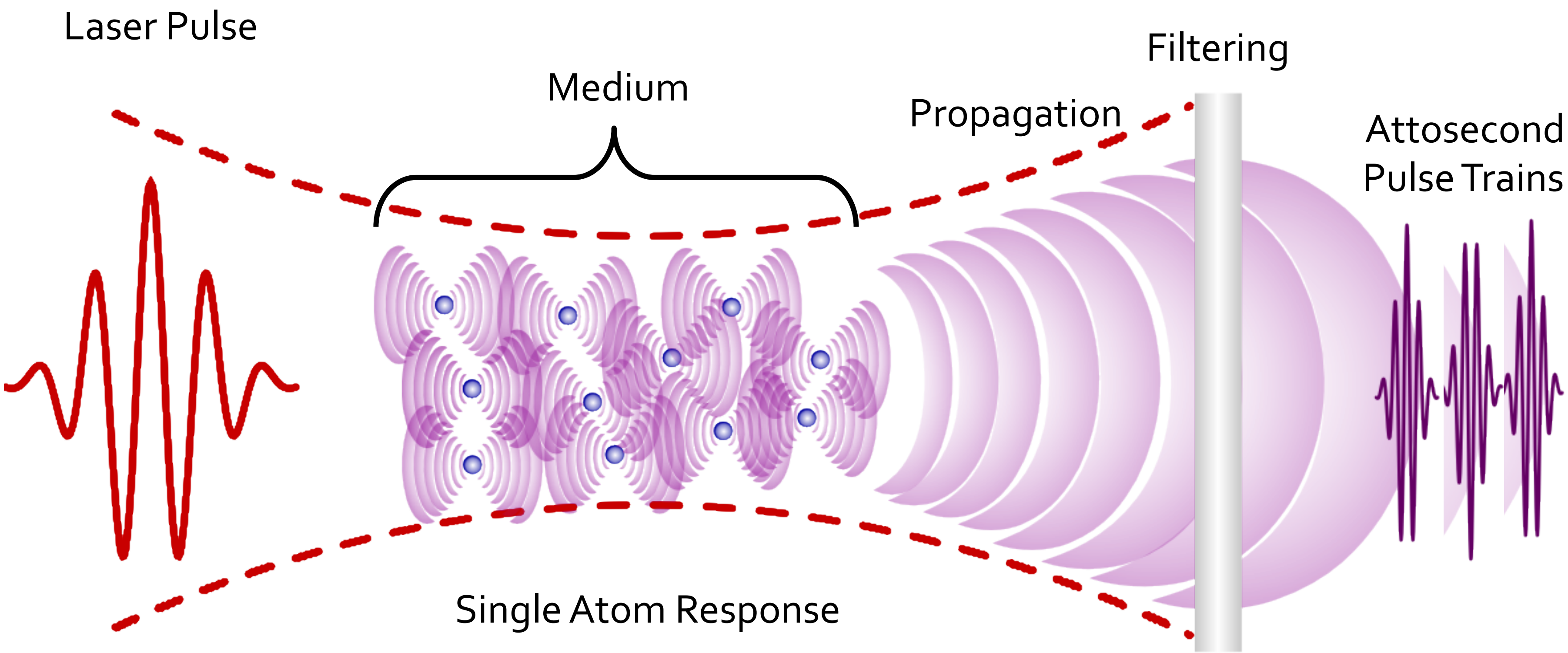


Attosecond Pulse Trains



P. Corkum, PRL **71**, 1994 (1993)

Attosecond Pulse Trains



$$x(t) \propto i\int_{-\infty}^{t} d\tau \int d\vec{p}\; d^{*}(\vec{p}+e\vec{A}(t))\, e^{-iS(\vec{p},t,\tau)/\hbar} E(t-\tau)\, d(\vec{p}+e\vec{A}(t-\tau))$$

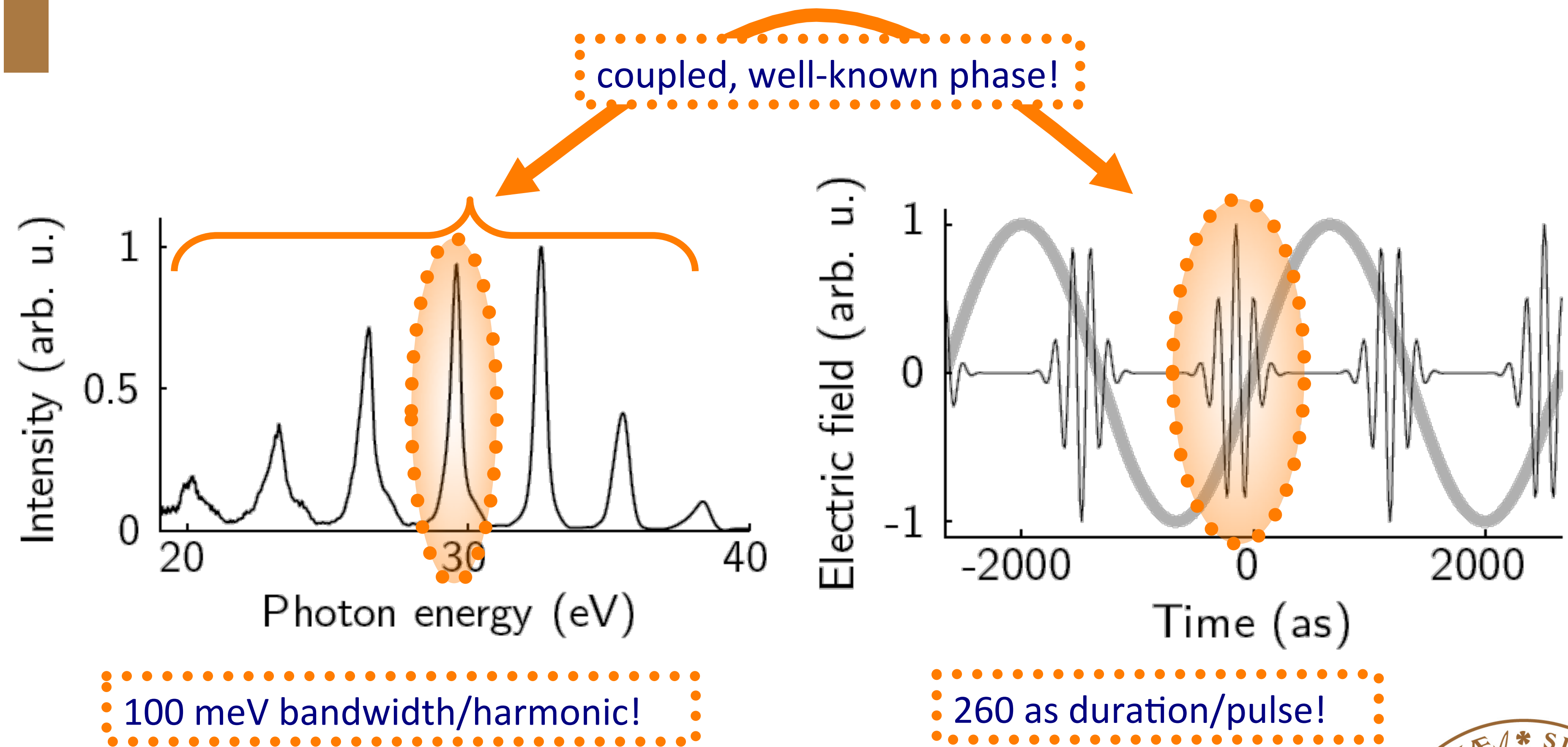
Lewenstein, 49, 2117, PRA 1994

Recombination

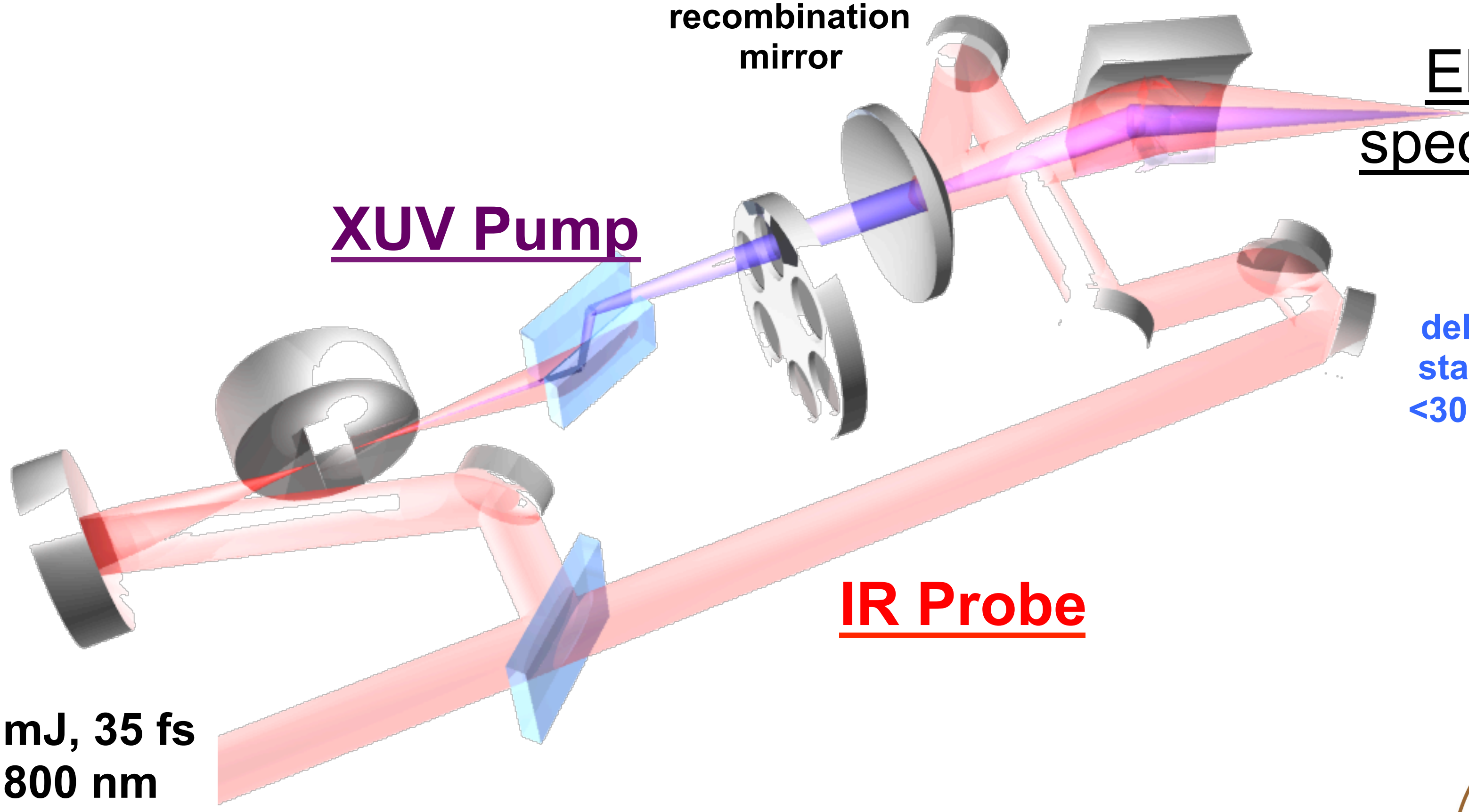
Acceleration in the continuum

Transition to the continuum

Attosecond Pulse Trains



Pump-probe setup

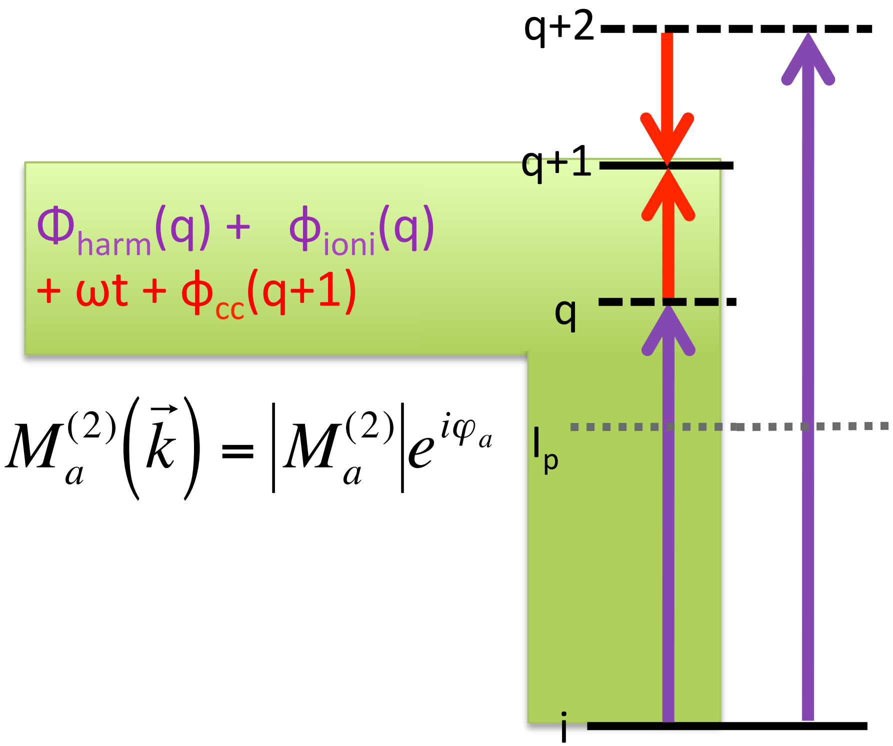


4 mJ, 35 fs
800 nm
Ti:Saph

INTERFEROMETRY IN FREQUENCE DOMAIN

Interferometric measurement

Ionization with two harmonics in the presence of a weak infrared field

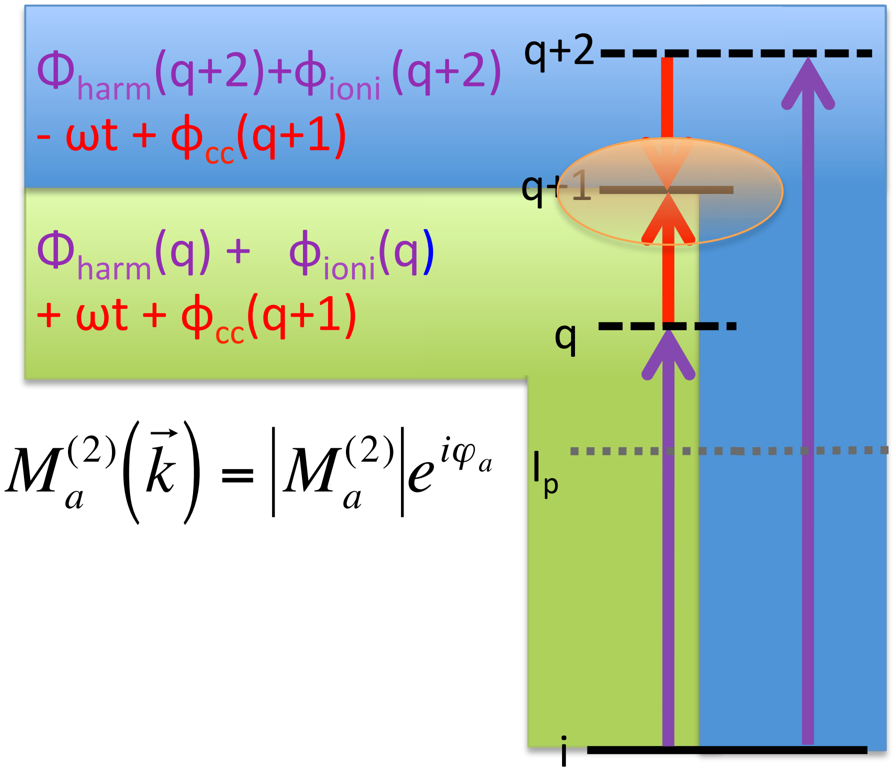


$$M_a^{(2)}\left(\vec{k}\right) = \left|M_a^{(2)}\right| e^{i\varphi_a}$$

Interferometric measurement

Ionization with two harmonics in the presence of a weak infrared field

$$M_e^{(2)}(\vec{k}) = \left|M_e^{(2)}\right| e^{i\varphi_e}$$



$$M_a^{(2)}(\vec{k}) = \left|M_a^{(2)}\right| e^{i\varphi_a}$$

Interference between two quantum paths

$$S_{q+1} = \left|M_a^{(2)}(\vec{k}) + M_e^{(2)}(\vec{k})\right|^2$$

$$S_{q+1} \propto \cos\left(2\omega t - \Delta\Phi_{harm} - \Delta\Phi_{ioni}\right)$$

Interferometric measurement

Ionization with two harmonics in the presence of a weak infrared field

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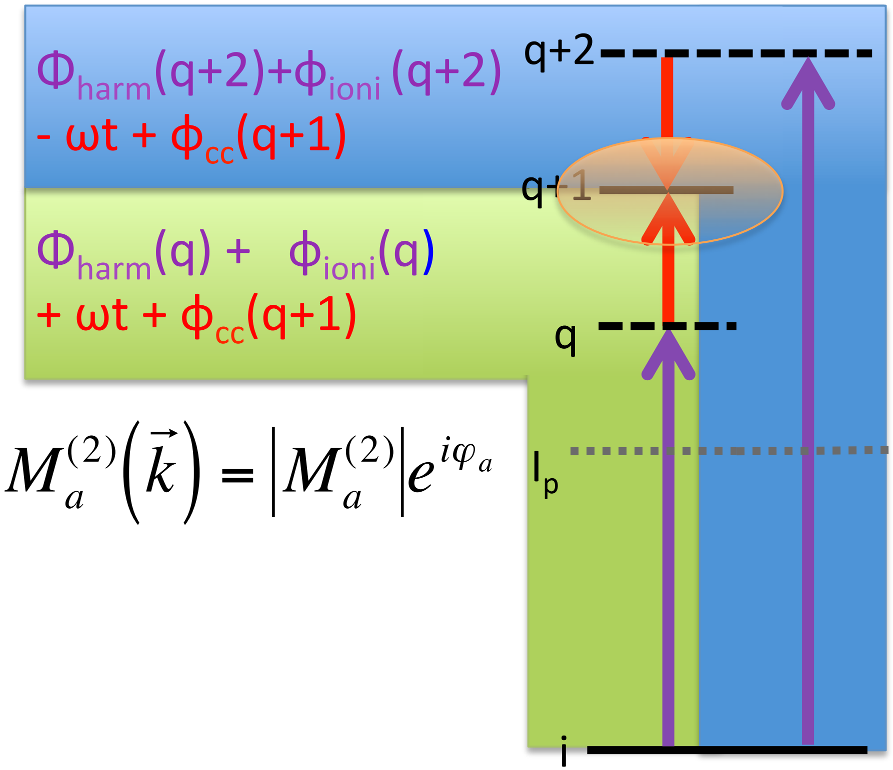

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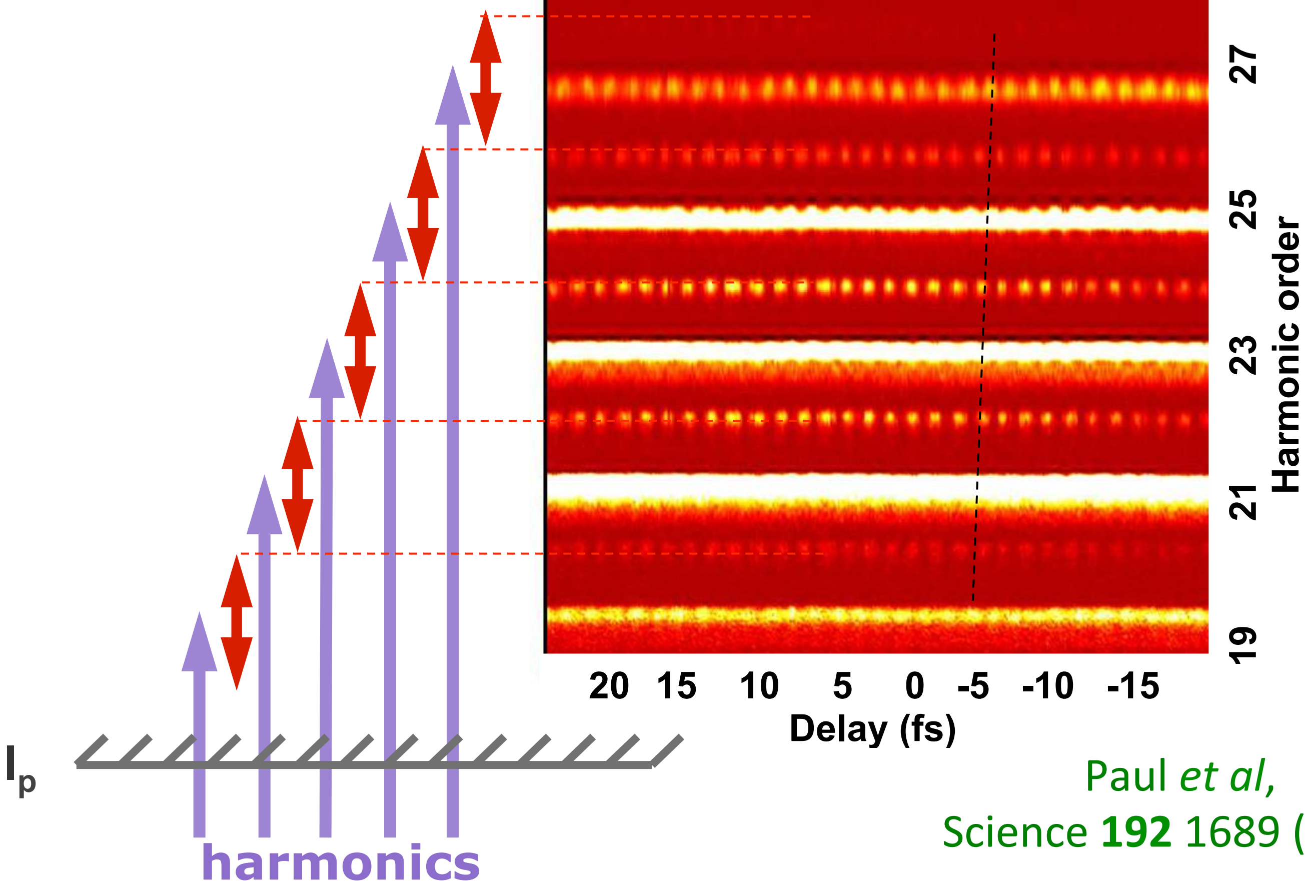
Interference between two quantum paths

$$S_{q+1} = \left|M_a^{(2)}(\vec{k}) + M_e^{(2)}(\vec{k})\right|^2$$

$$S_{q+1} \propto \cos\left(2\omega t - \Delta\Phi_{harm} - \overset{\approx 0}{\Delta\Phi_{ioni}}\right)$$

Reconstruction of Attosecond Beating by Interference of Two-photon Transition

$A + h\nu_{H19\text{-}H27} + h\nu_{IR} \rightarrow A^{+} + e^{-}$



Paul *et al*,
Science **192** 1689 (2001)

Reconstruction of Attosecond Beating by Interference of Two-photon Transition

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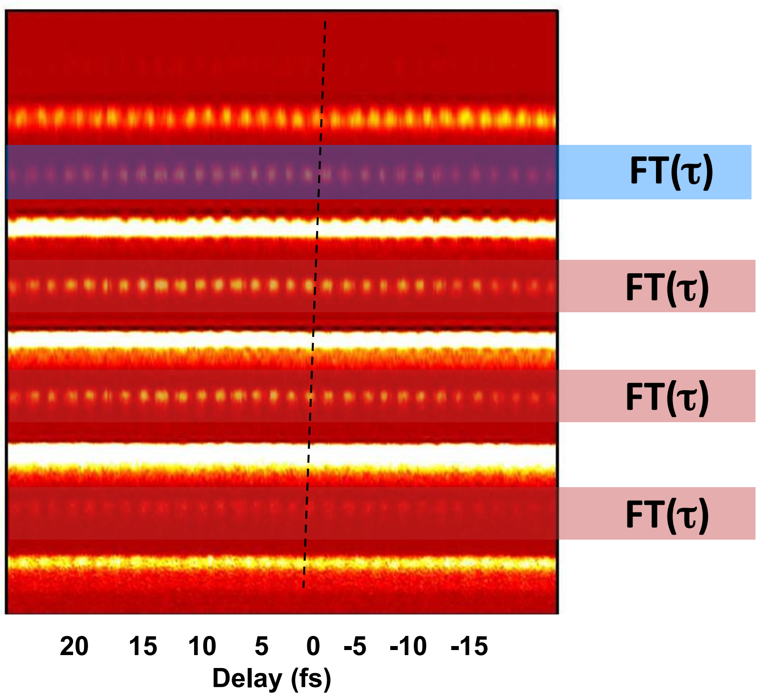
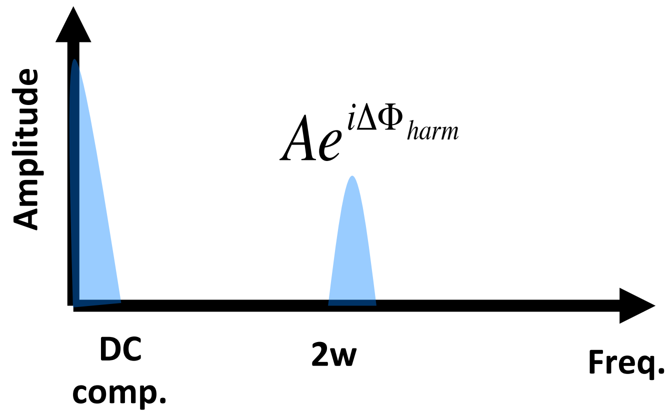
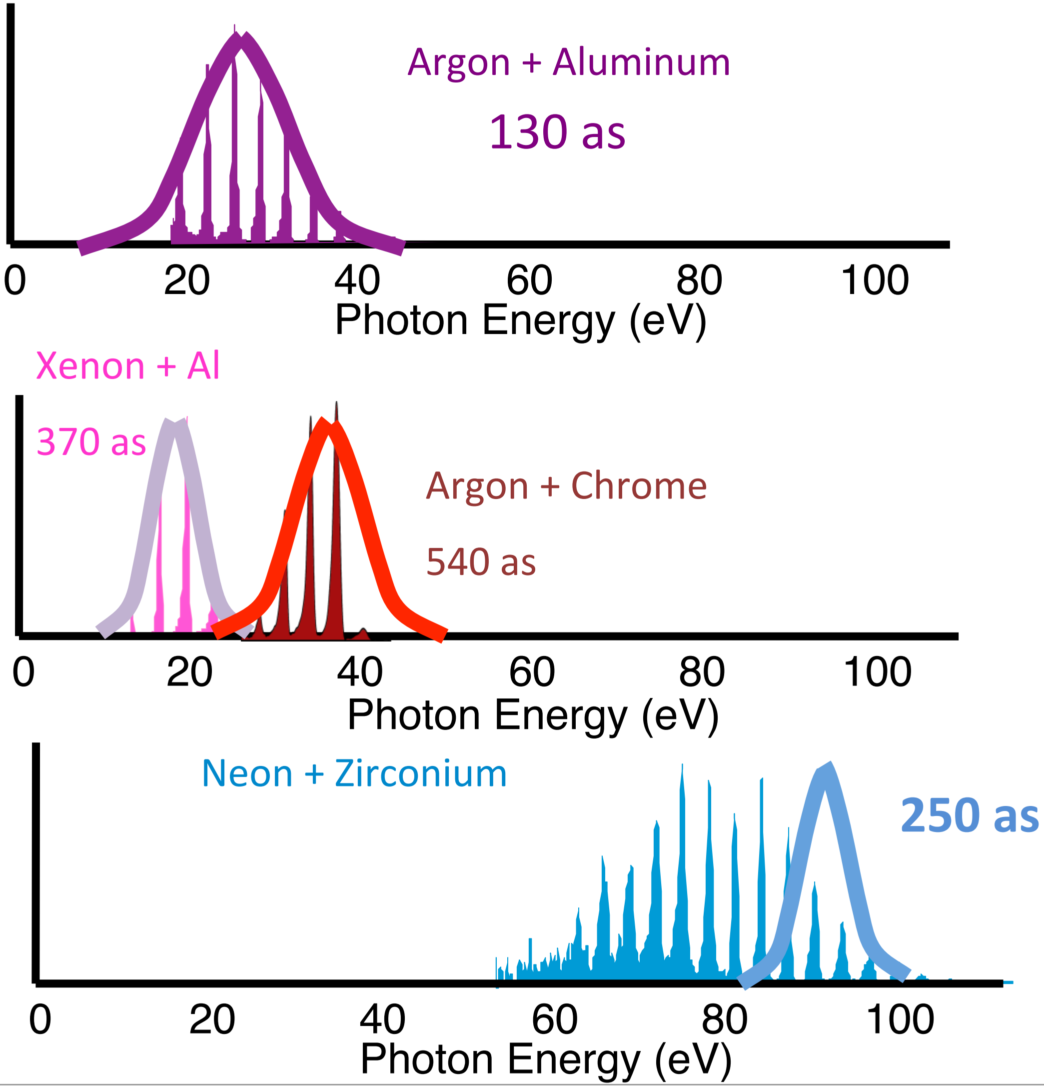


$$S_{q+1} \propto \cos\left(2\omega t - \Delta\Phi_{harm}\right)$$



Phase determination down to 20 as !

Reconstruction of Attosecond Beating by Interference of Two-photon Transition



…

López-Martens *et al*,
Phys Rev Lett **94** 3301 (2005)

…

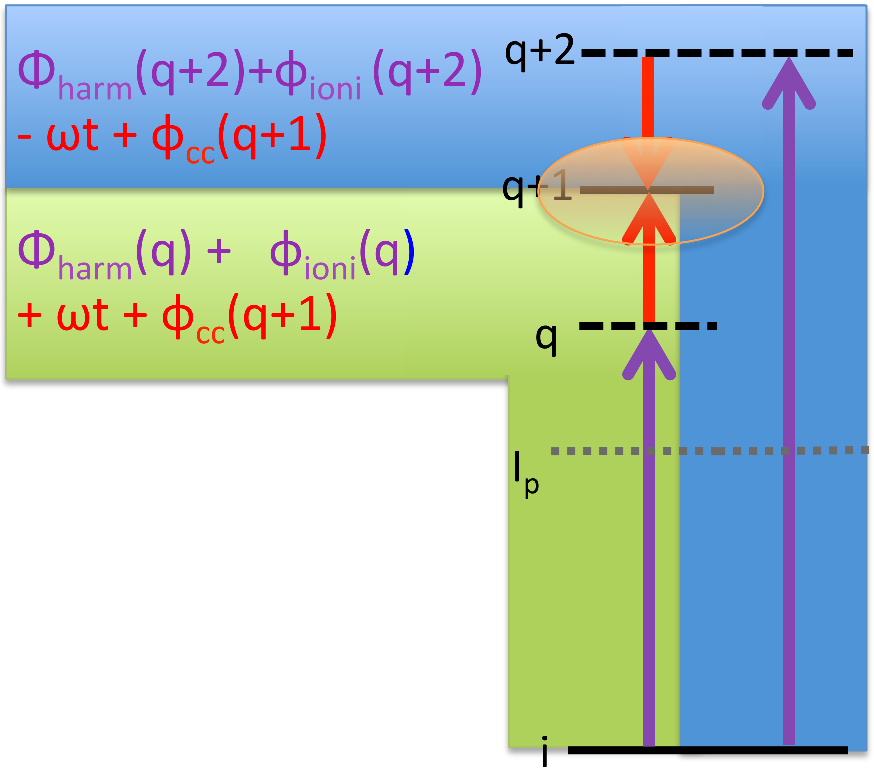
E. Gustafsson *et al*,
Optics Letters 32, 1353 (2007)

…

Resonant photo-excitation – atomic case

Interferometric measurement

Ionization with two harmonics in the presence of a weak infrared field



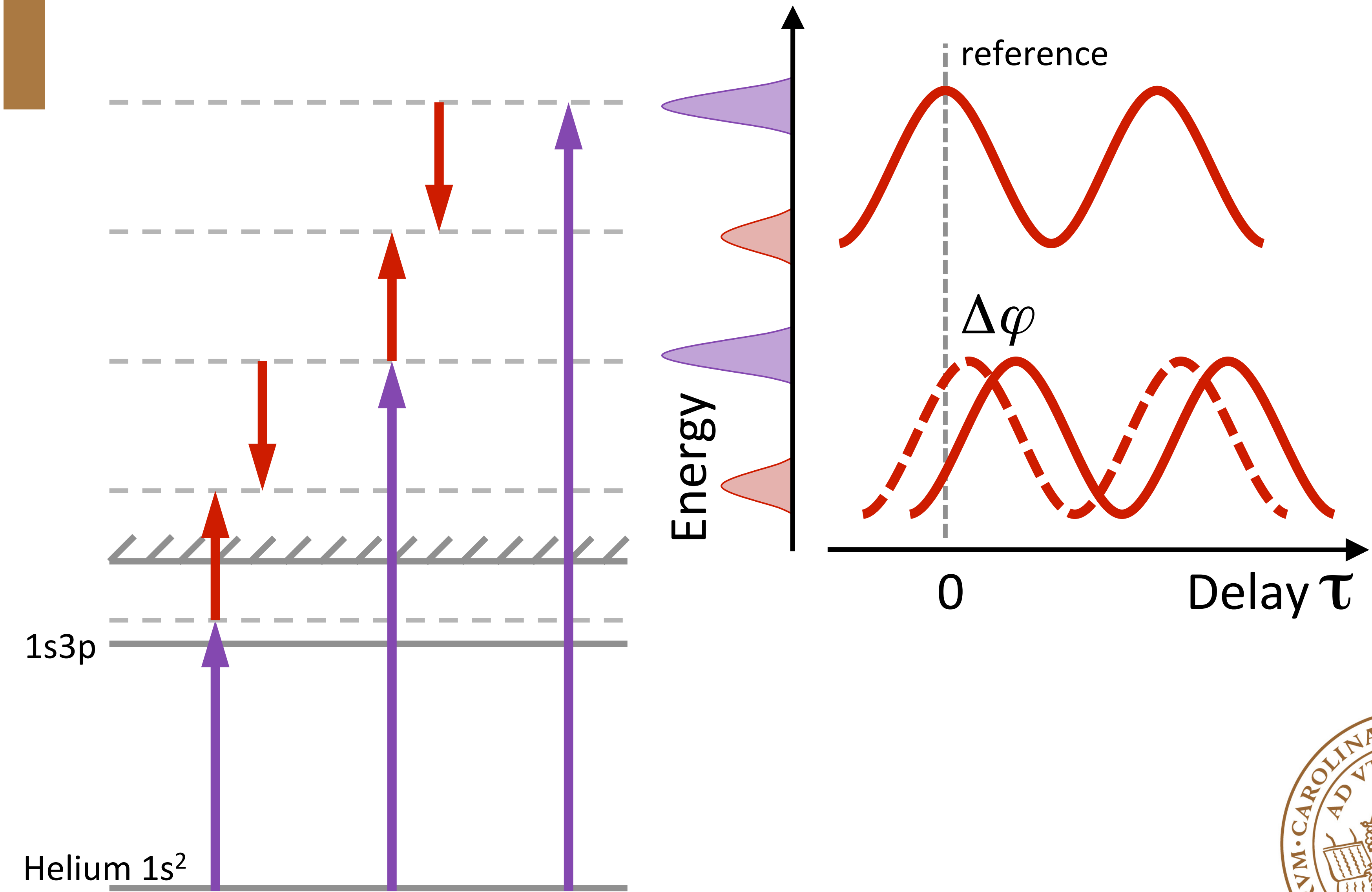
Interference between two quantum paths

$$S_{q+1} \sim \cos(2\omega t - \Delta\phi_{harm} - \Delta\phi_{ioni})$$

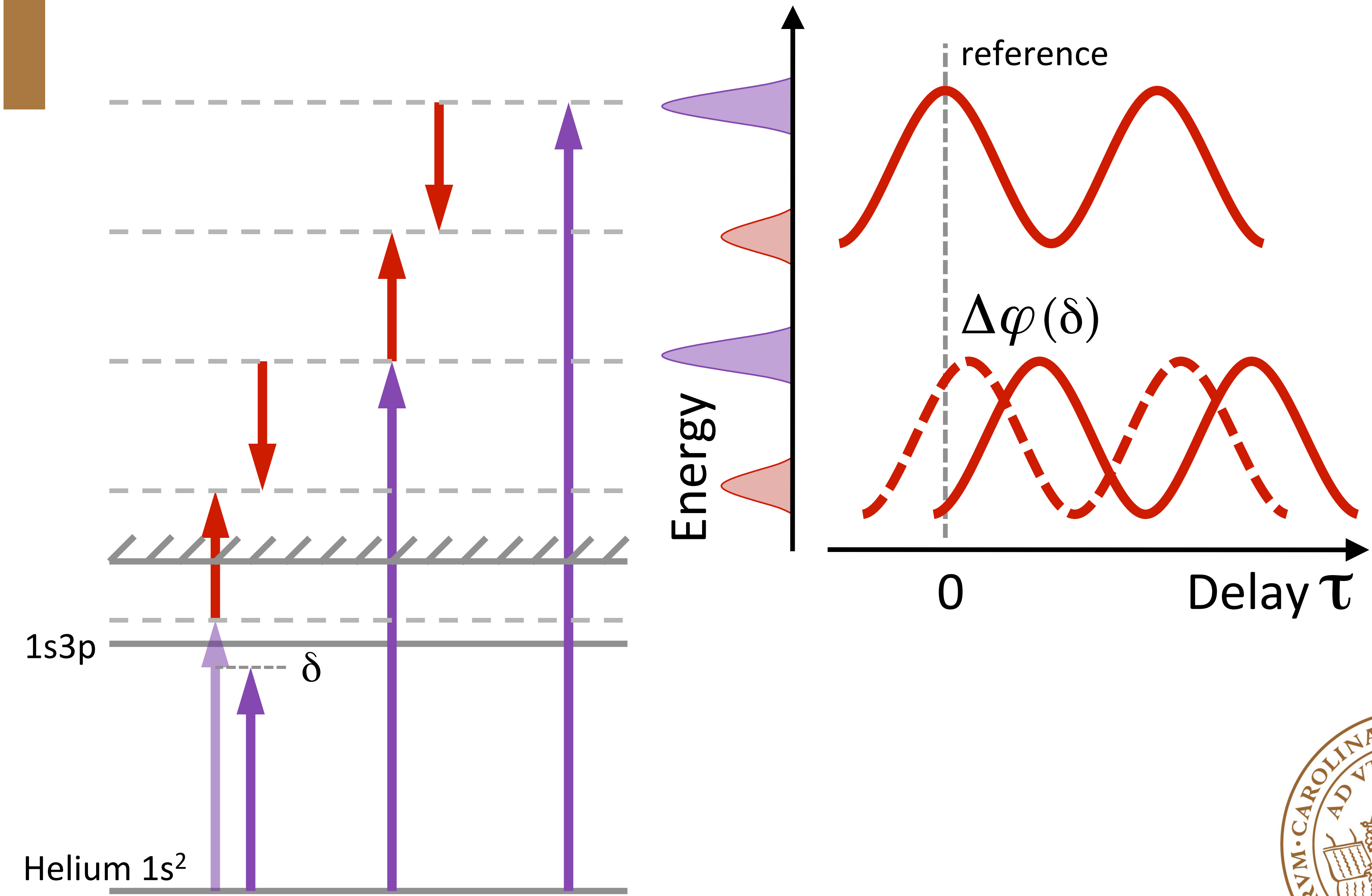
Idea

"Two photon Resonant the photo-ionization pathway will introduce a phase jump of π"

Two-Color Two-Photon Ionization

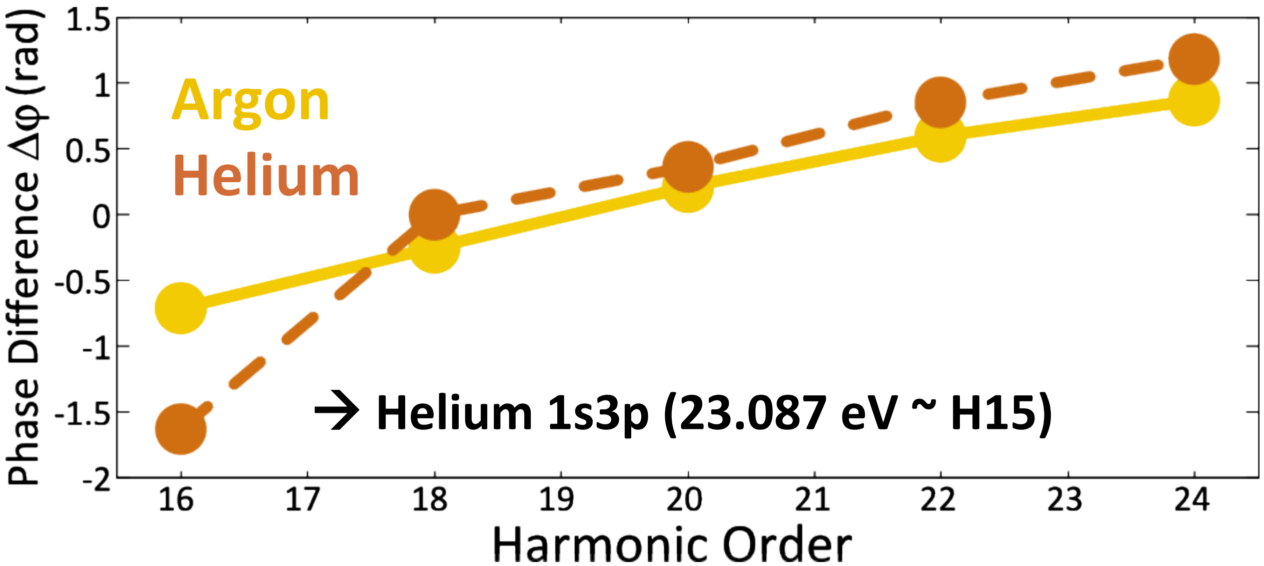


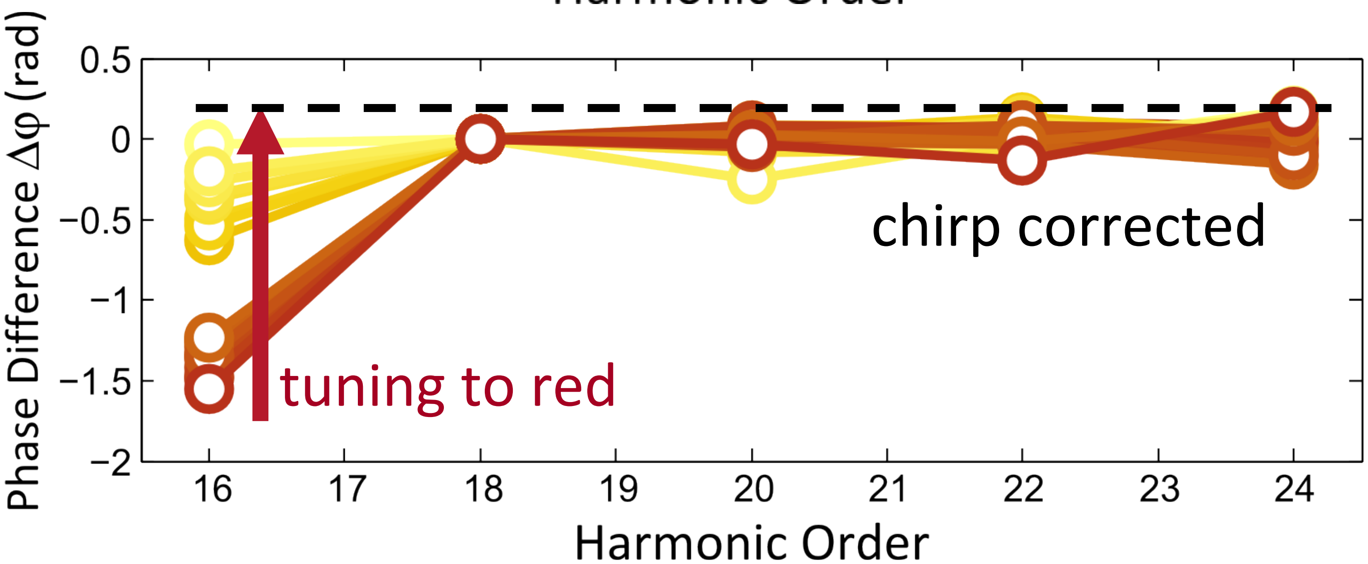
Two-Color Two-Photon Ionization



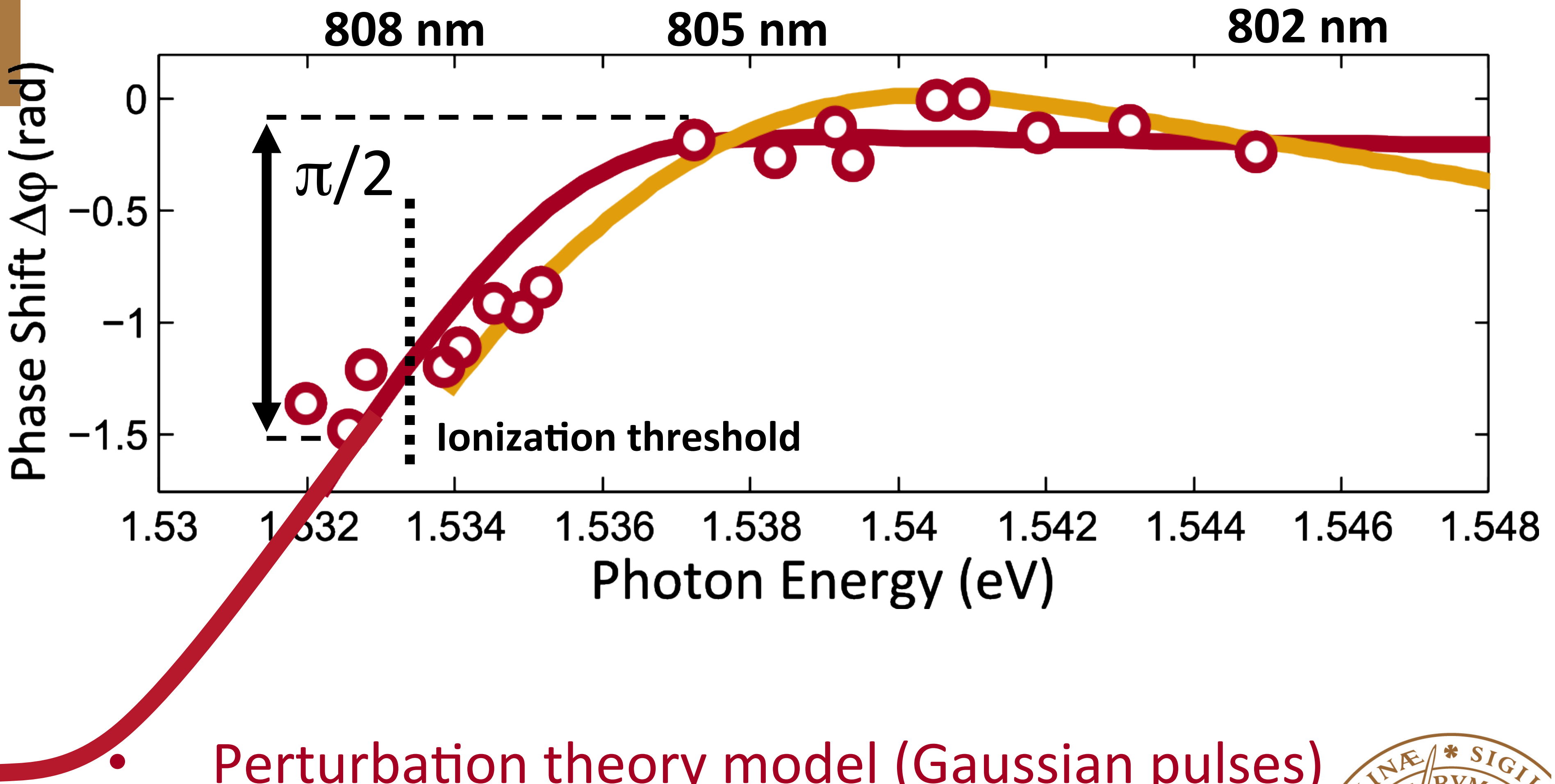
Harmonic Δφ:Argon vs Helium

Swodoba *et al,* PRL **104**, 103003 (2010)





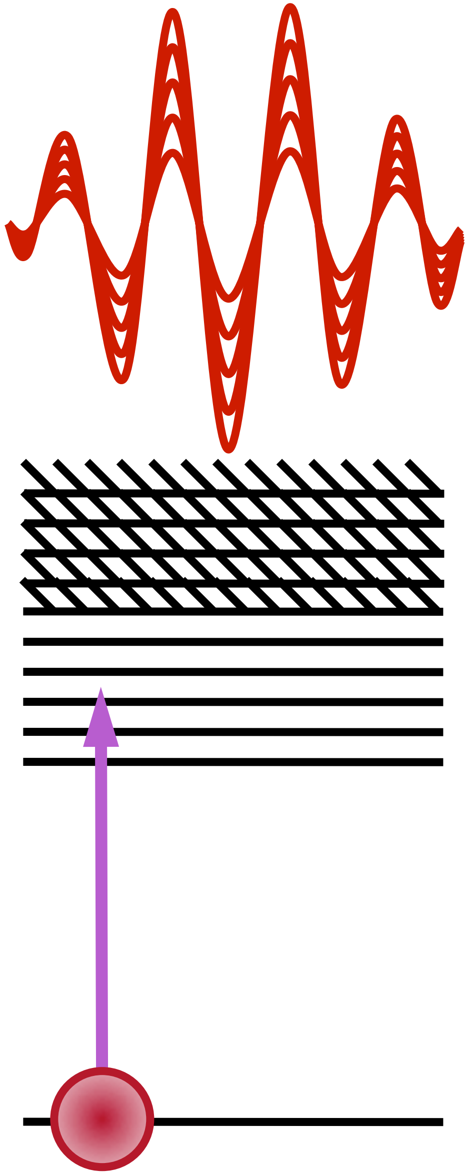
Wavelength Dependence

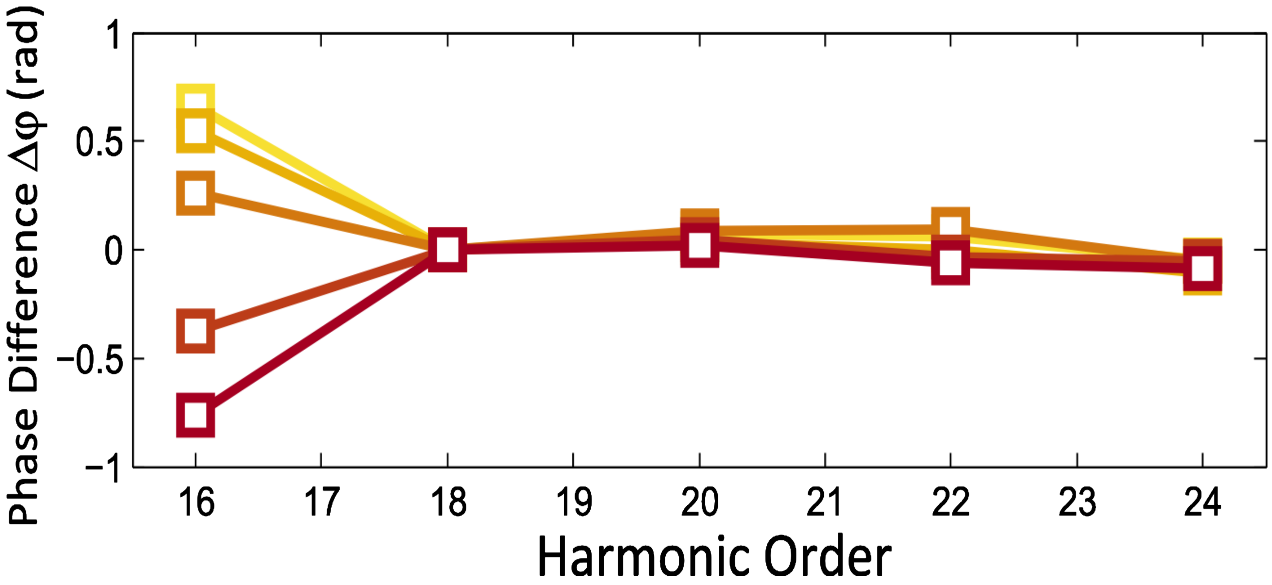


- Perturbation theory model (Gaussian pulses)
- TDSE simulation (@ 1e11 W/cm^2)

Intensity Dependence

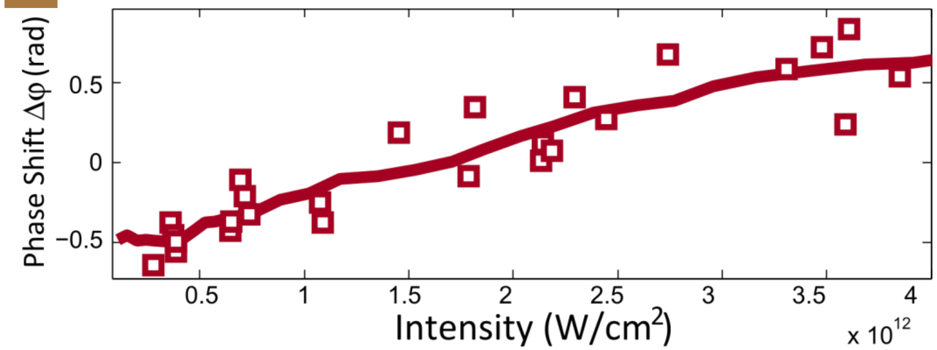
Move the state instead of the photon frequency

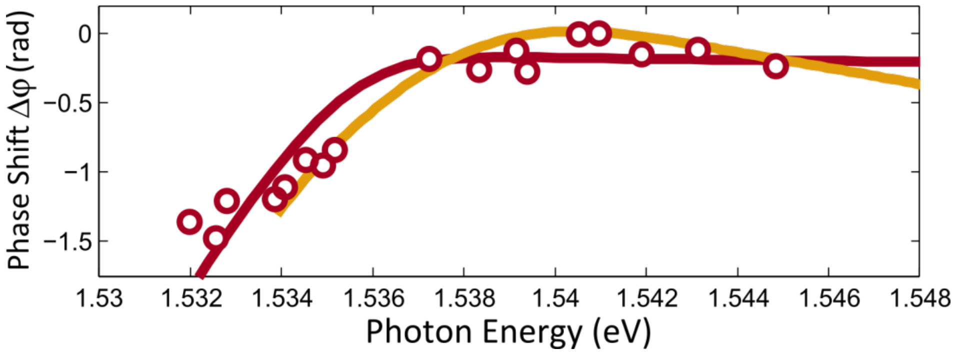




Measure the AC Stark Shift

- combine intensity & wavelength dependence of $\Delta\varphi$:





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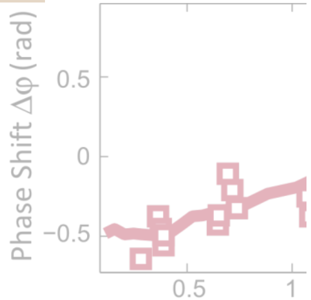
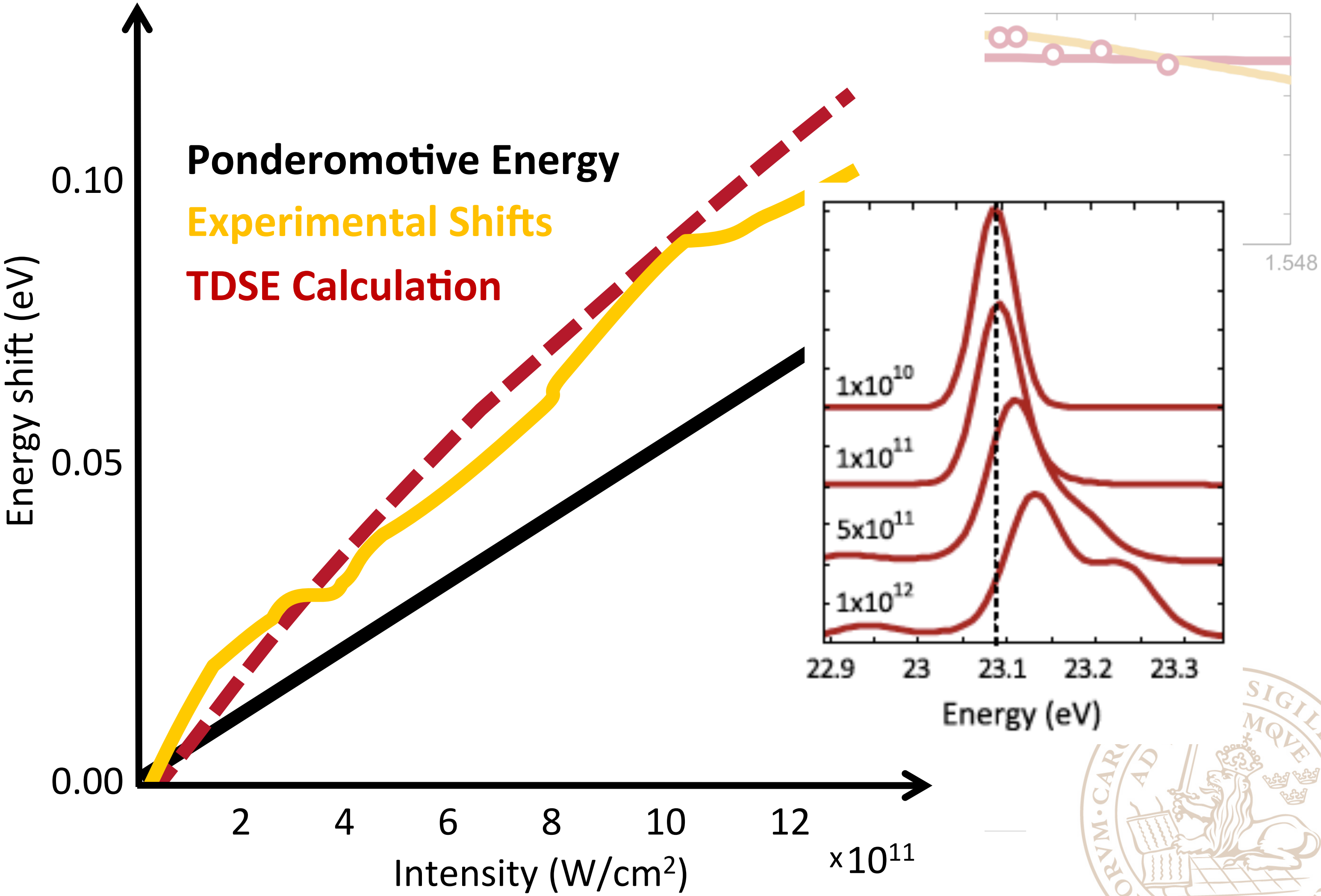
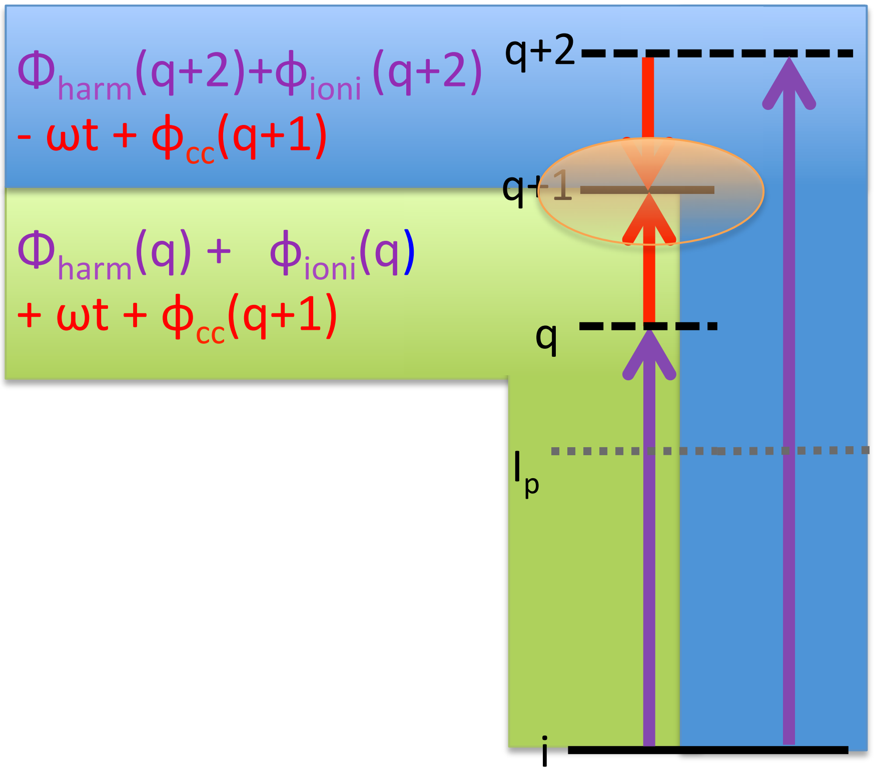




Emission time in photo-ionization

Interferometric measurement

Ionization with two harmonics in the presence of a weak infrared field



Interference between two quantum paths

$$S_{q+1} \sim \cos(2\omega t - \Delta\phi_{harm} - \Delta\phi_{ioni})$$

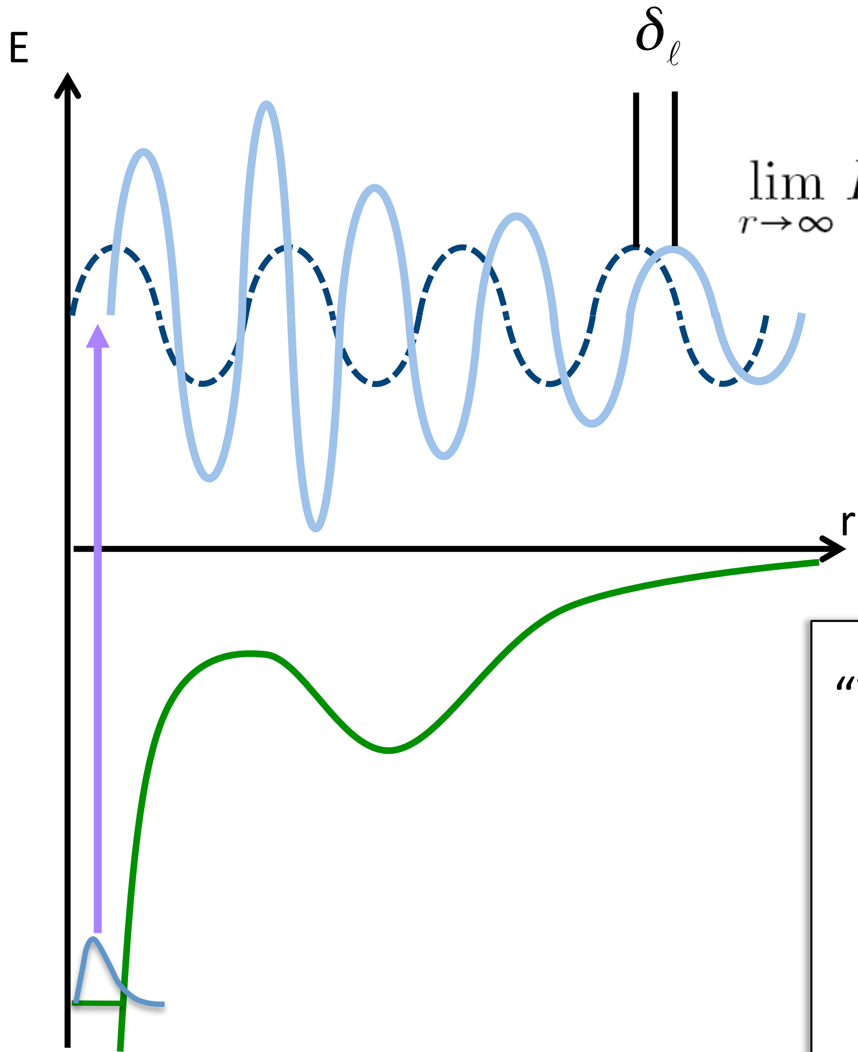
$$\Delta\phi_{ioni} = \phi_{ioni}(q+2) - \phi_{ioni}(q)$$

$$\sim 2\omega_0 * d\phi_{ioni}(q+1)/d\omega$$

$$\sim 2\omega_0 * GD$$

(GD: Group Delay)

Dwell time in scattering experiment



Divergent long-range phase

$$\lim_{r\to\infty} R_{k\ell}(r) \propto \sin\left[kr - \frac{\ell}{2}\pi + \frac{1}{k}\ln(2kr) + \eta_\ell(k)\right]$$

Centrifugal term

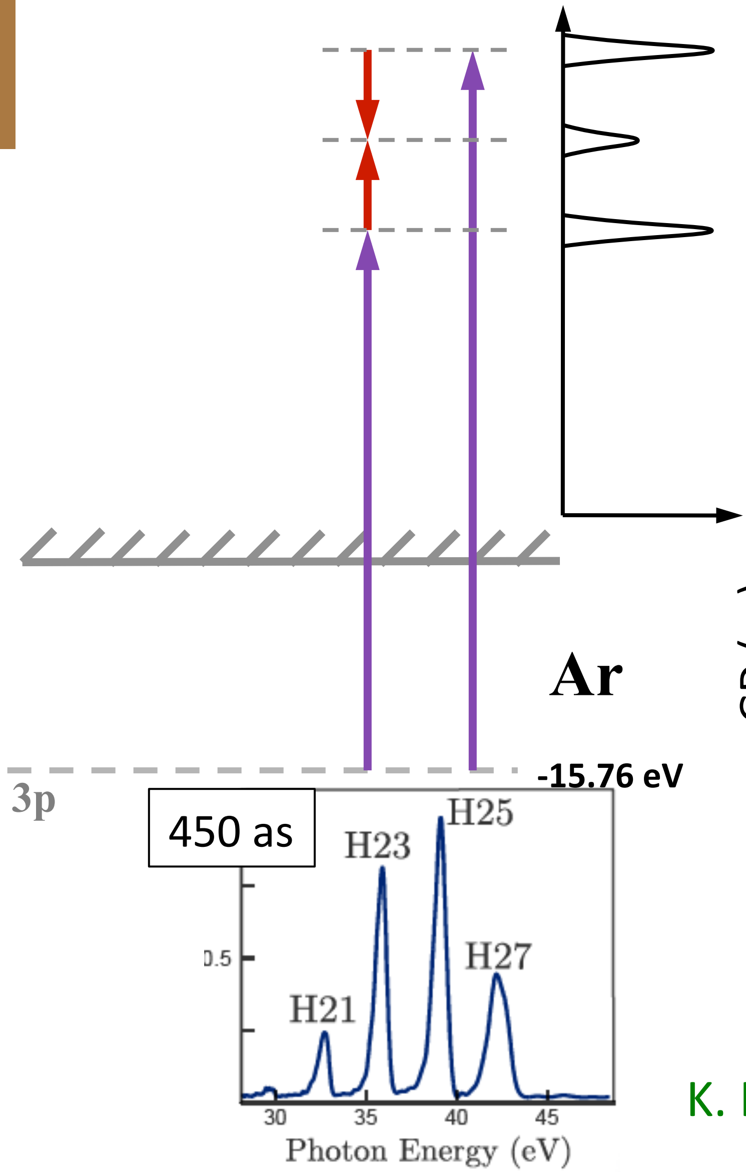
Scattering phase

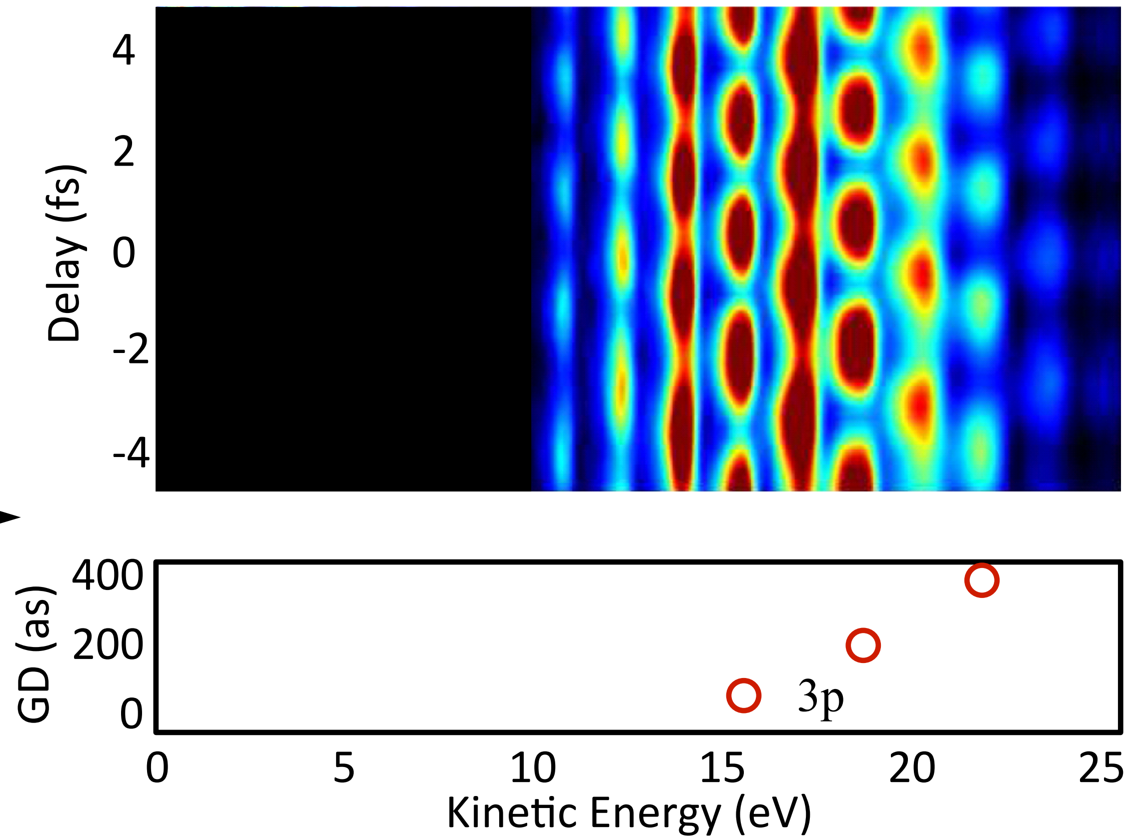
“Wigner time delay” for photo-ionization

Wigner, Phys Rev **98**, 145 (1955)

$$\tau_W = \hbar \frac{d\eta_\ell}{d\varepsilon}$$

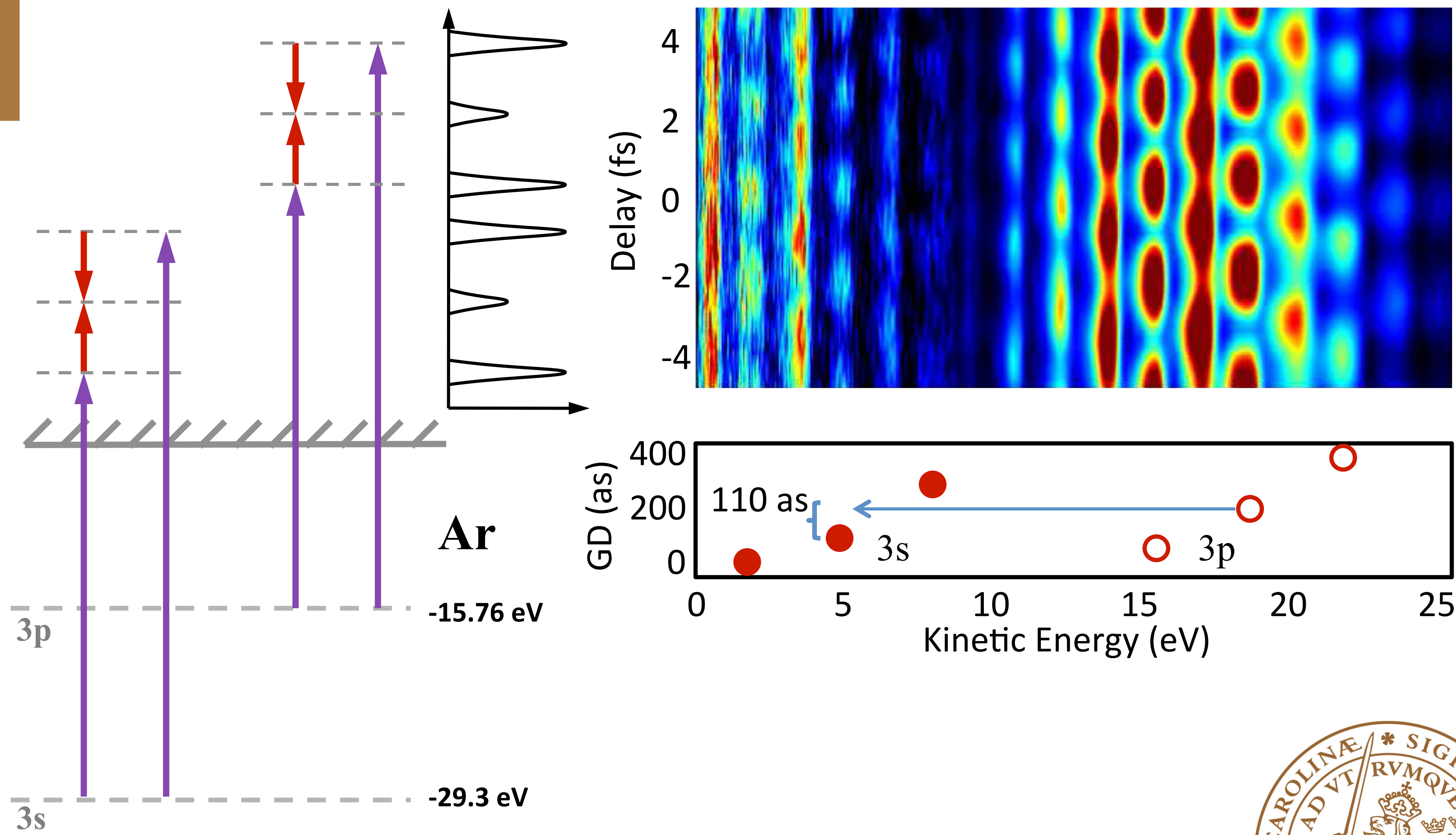
Ionization from the 3s and 3p shells





K. Klünder *et al*, PRL **106**, 143002 (2011)

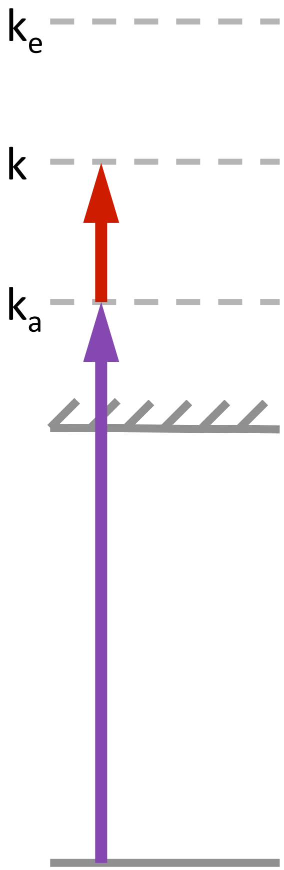
Ionization from the 3s and 3p shells



K. Klünder *et al*, PRL **106**, 143002 (2011)

Interpretation

Perturbation theory - Independent particle model

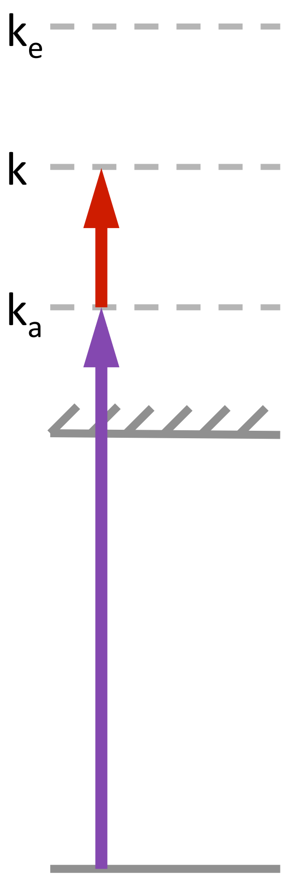


All intermediate states, *n*, must be considered

$$M_a^{(2)}(\vec{k}) = -iE_{\mathrm{L}}E_{\mathrm{H}} \lim_{\varepsilon \to 0^+} \sum\!\!\!\!\!\!\int_n \frac{\langle \varphi_{\vec{k}} | \vec{\epsilon} \cdot \vec{r} | \varphi_n \rangle \langle \varphi_n | \vec{\epsilon} \cdot \vec{r} | \varphi_i \rangle}{\epsilon_i + \omega_{\mathrm{H}} - \epsilon_n + i\varepsilon}$$

Interpretation

Perturbation theory - Independent particle model



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Partial wave expansion

$$M_a^{(2)}(\vec{k}) = -iE_{\mathrm{L}}E_{\mathrm{H}} \sum_{\ell=0,2} C_{\ell 0} Y_{\ell 0}(\hat{k})\, e^{i\eta_\ell(k)} T_a^{(2)}(k)$$

Scattering phase of the final state

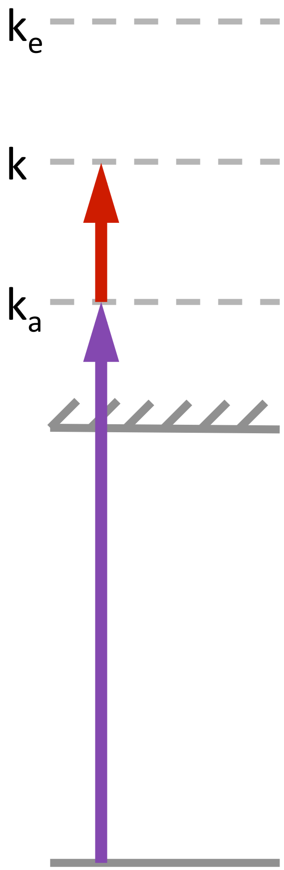
Radial part

Define the pertubated wavefunction ρ

$$T_a^{(2)}(k) = \sum\!\!\!\!\!\!\int_n \frac{\langle R_{k\ell}|r|R_{n1}\rangle\langle R_{n1}|r|R_{i0}\rangle}{\epsilon_i + \omega_{\mathrm{H}} - \epsilon_n + i\varepsilon} = \langle R_{k\ell}|r|\rho_{k_a 1}\rangle$$

Interpretation

Perturbation theory - Independent particle model



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Using asymptotic development
R_{kl} and ρ_{ka1}

$$M_a^{(2)}(k) \propto \underbrace{e^{i\eta_1(k_a)}}_{(\mathrm{I})} \times \underbrace{\left(\frac{i}{k_a - k}\right)^{iz} \frac{(2k_a)^{\frac{i}{k_a}}}{(2k)^{\frac{i}{k}}} \Gamma(2+iz)}_{(\mathrm{II})}$$

(I) Intermediate scattering phase
(II) Continuum-Continuum phase

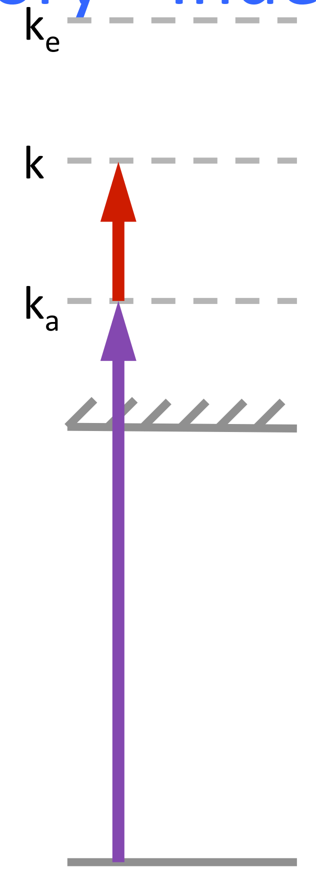
Interpretation

Perturbation theory - Independent particle model

$$M_a^{(2)} \propto e^{i\eta(k_a)} \times e^{i\varphi_{cc}(k_a,k)}$$

one-photon transition

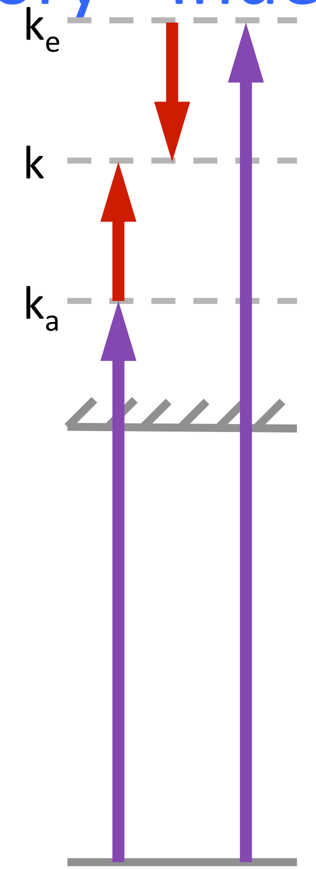
continuum-continuum transition



Interpretation

Perturbation theory - Independent particle model

$$M_a^{(2)} \propto \underbrace{e^{i\eta(k_a)}}_{\text{one-photon transition}} \times \underbrace{e^{i\varphi_{cc}(k_a,k)}}_{\text{continuum-continuum transition}}$$



$$M_e^{(2)} \propto \underbrace{e^{i\eta(k_e)}}_{\text{one-photon transition}} \times \underbrace{e^{i\varphi_{cc}(k,k_e)}}_{\text{continuum-continuum transition}}$$

$$\Rightarrow \quad GD = \frac{\eta(k_e)-\eta(k_a)}{2\omega} + \frac{\varphi_{cc}(k,k_e)-\varphi_{cc}(k_a,k)}{2\omega}$$

Interpretation

Perturbation theory - Independent particle model

k_e

$$M^{(2)} \propto e^{i\eta(k_e)} \times e^{i\varphi_{cc}(k,k_e)}$$

continuum-continuum transition

$$M_a^{(2)} \propto e^{i\eta(k_a)} \times$$

one-photon transition

Group Delay

"Wigner-Eisenberg time delay" for photo-ionization

Wigner, Phys Rev **98**, 145 (1955)

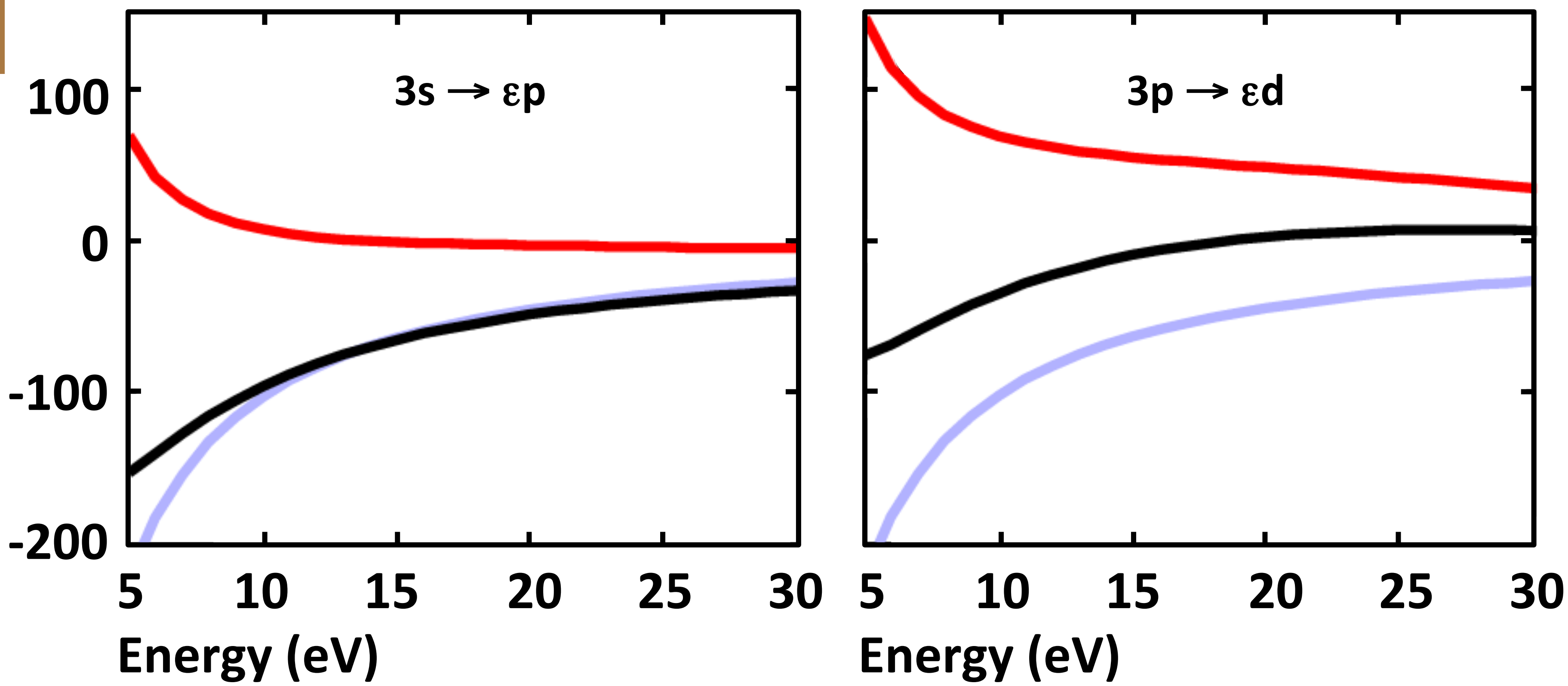
$$\tau_W = \hbar \frac{d\eta}{d\varepsilon}$$

$$GD = \frac{\eta(k_e) - \eta(k_a)}{2\omega} + \frac{\varphi_{cc}(k,k_e) - \varphi_{cc}(k_a,k)}{2\omega}$$

Wigner time delay **Continuum-continuum delay**

Calculated Group Delay

Independent particle model



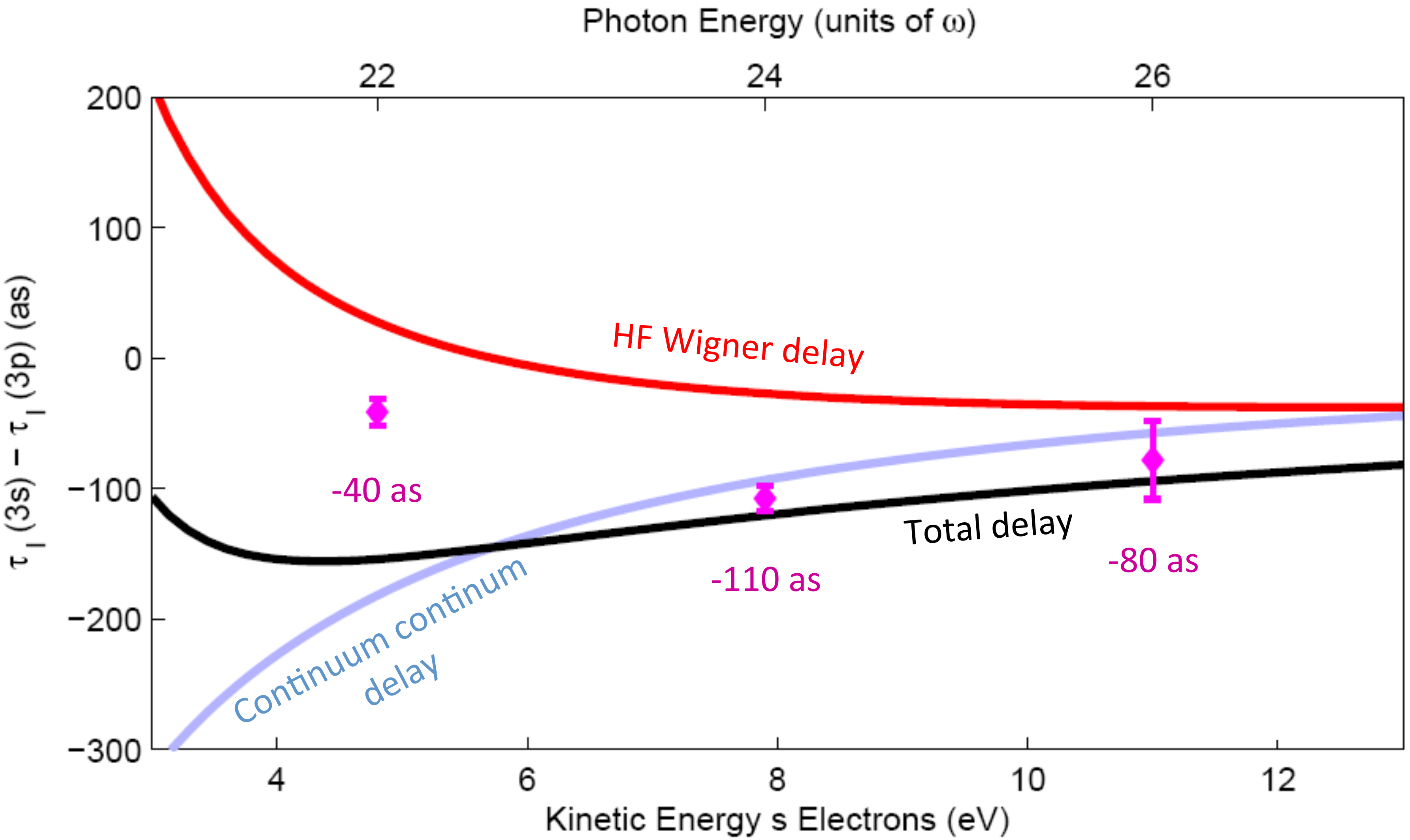
HF Wigner time delay (1-photon transition)

Continuum-continuum delay

Total

Kennedy and Manson, Phys. Rev. A **5**, 227 (1972)

Comparison with HF



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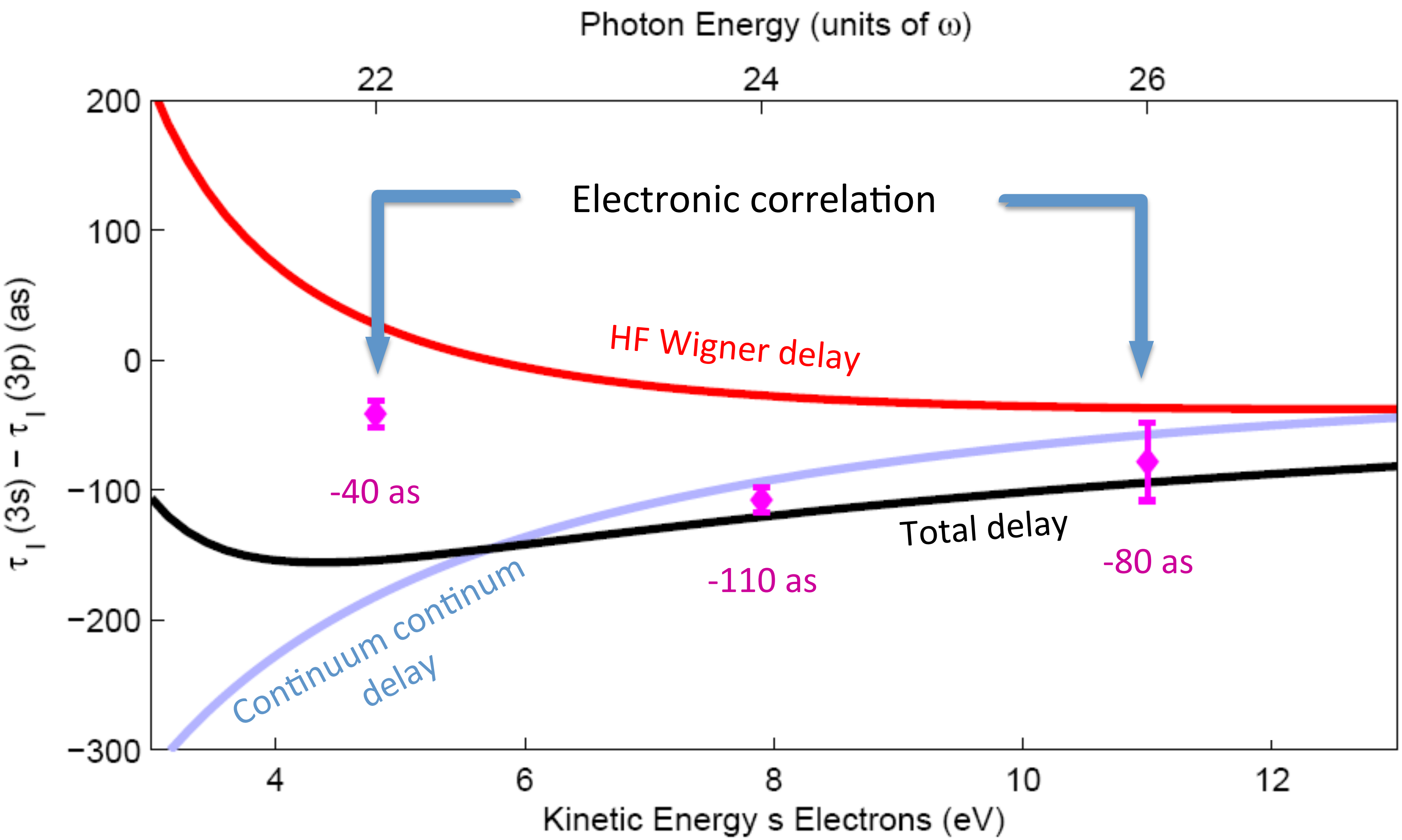
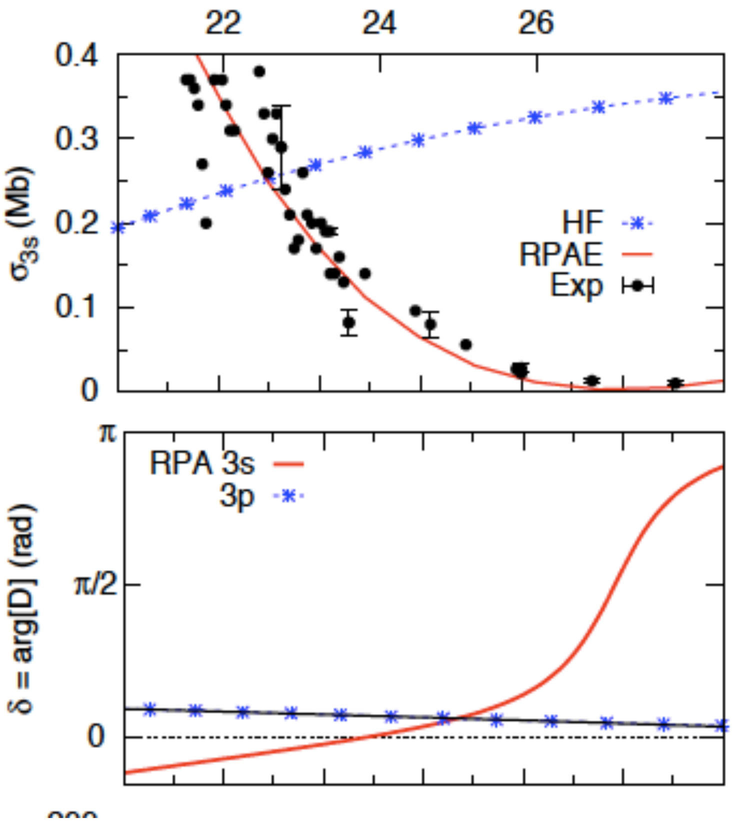
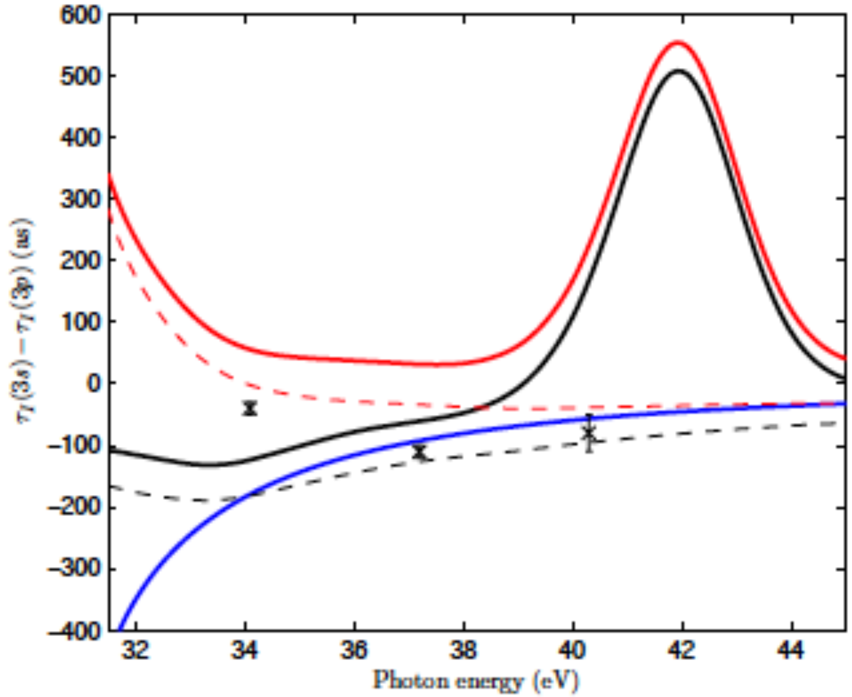


Comparison with RPAE

”Cooper minimum” in the 3s channel





With the courtesy of A. Kheifets

Conclusion

- Interferometric methods enables to read out the
  Group Delay (GD) of the Electronic Wave Paquet

- The GD encodes information on:
  One - photon transition phase

- The measurement introduces a contribution to the GD
  Continuum - continuum transition phase

⇨ New insight into electronic correlation

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