Progress report:

Precise attosecond pulse characterization

How to use coherent bound wave packets

J. Marcus Dahlström

Atomic Physics at Stockholm University [SU]

$\longrightarrow$

Mathematical Physics at Lund University [LU]

`2017-07-31 Cairns Convention Center, Australia.`

Outline of Progress Report:

- **Introduction to attosecond pulse characterization**
- **Recent progress:**
  - *Noble gas atoms and negative ions*
- **Novel approach: Ionization of bound wave packets**
- **Test cases:**
  - *Alkali atoms and noble gas atoms*
- **Conclusions**

"Eliminating the dipole phase in attosecond pulse characterization using Rydberg wave packets"

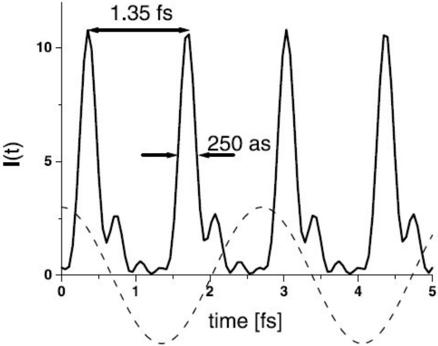
[*Pabst and Dahlström, PRA* **94**, *013411 (2016)*]

(NORDITA + ITAMP VISITOR PROGRAM)

Observation of a Train of Attosecond Pulses from High Harmonic Generation

P. M. Paul,[1] E. S. Toma,[2] P. Breger,[1] G. Mullot,[3] F. Augé,[3]
Ph. Balcou,[3] H. G. Muller,[2*] P. Agostini[1]

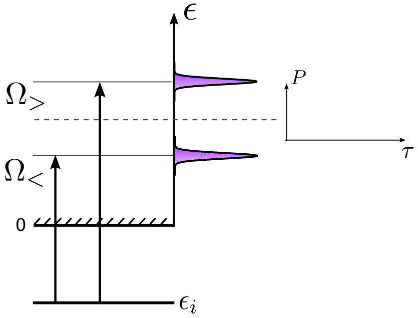
In principle, the temporal beating of superposed high harmonics obtained by focusing a femtosecond laser pulse in a gas jet can produce a train of very short intensity spikes, depending on the relative phases of the harmonics. We present a method to measure such phases through two-photon, two-color photoionization. We found that the harmonics are locked in phase and form a train of 250-attosecond pulses in the time domain. Harmonic generation may be a promising source for attosecond time-resolved measurements.



[Paul *et al.*, SCIENCE 1690 **292** (2001)]

Introduction to attosecond pulse characterization

Photoelectrons in energy domain: $P(\epsilon) \sim |E(\Omega)|^2 |\Psi(\epsilon)|^2$



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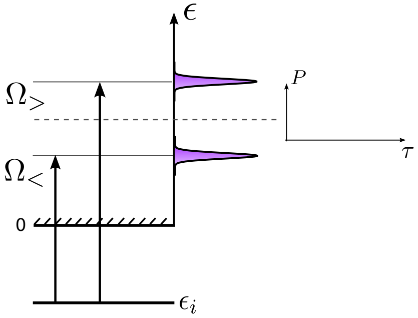


XUV harmonic comb for High Harmonic Generation:

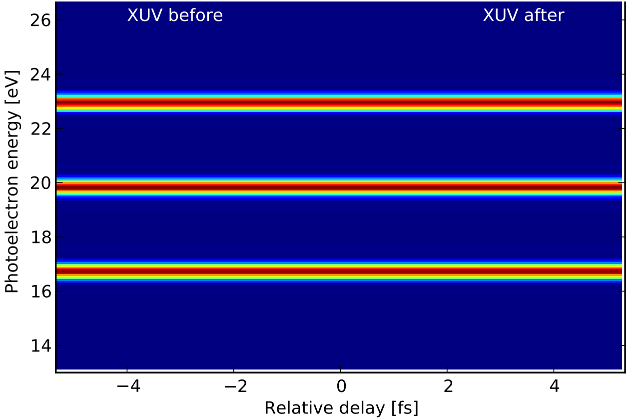
Odd high-order **harmonics** of laser: $\Omega_{2q+1} = (2q+1)\omega$

Discrete peak of photoelectrons: $\epsilon_{2q+1} = \hbar\Omega_{2q+1} - I_p$,

GOAL: **Measure the spectral phase** between harmonics.

Photoelectron spectrogram

Overlap XUV comb and phase-locked laser light:



Laser-assisted redistribution of the photoelectron comb peaks.

Formation of **sidebands** at **even** harmonic energies.

Photoelectron spectrogram

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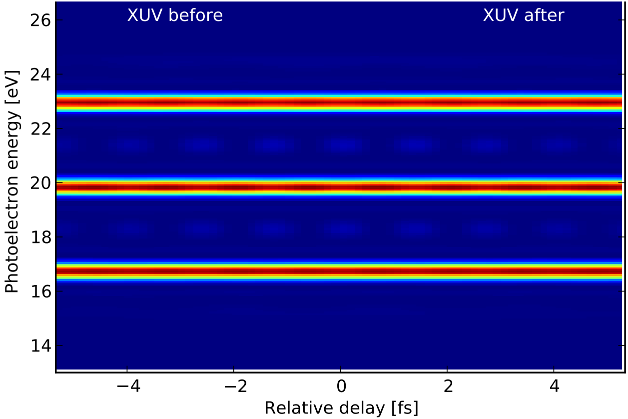

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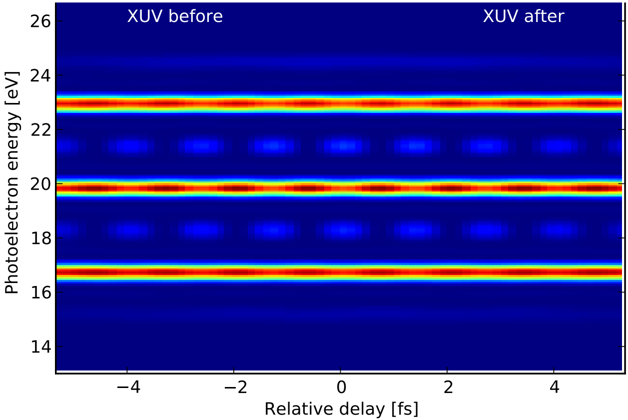

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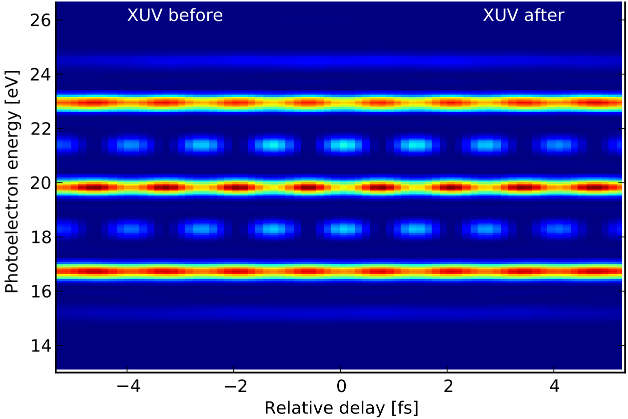

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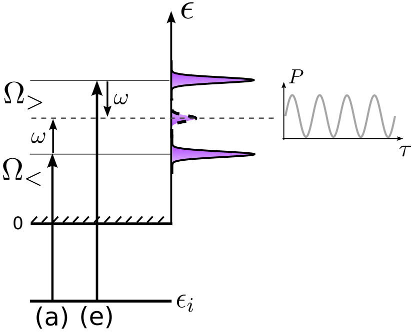
Temporal characterization of high-order harmonics

Spectral shearing interferometry by laser field:

$\rightarrow$ Extract the group delay (GD) of attopulse.



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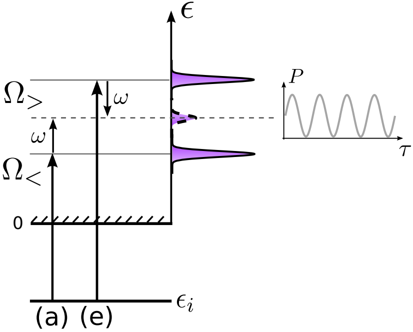


Interference of quantum paths:

$$P \approx A + B\cos[2\omega(\tau - \tau_{\rm GD} - \tau_{\rm Atom})],$$

where $\tau_{\rm GD} \approx (\phi_> - \phi_<)/2\omega$ is **group delay** (GD) of attopulse.

RABBITT method due to H.G. Muller.

Quite complex process...

Amplitude *and* phase of two-photon matrix elements

Table 1. The atomic phases $\Delta\varphi^{f}_{\text{atomic}}$ and the relative strengths A_f of each two-photon transition responsible for the sideband peaks. The numbers within the parentheses represent the values of the angular and magnetic quantum numbers of the initial 3p state and the final continuum state of the listed energy.

Sideband	$\Delta\varphi^{f}_{\text{atomic}}$ (rad) / amplitude A_f (arbitrary units)			
	$(1,0) \rightarrow (1,0)$	$(1,0) \rightarrow (3,0)$	$(1,\pm1) \rightarrow (1,\pm1)$	$(1,\pm1) \rightarrow (3,\pm1)$
$E_0 + 12\hbar\omega$	0.438/6094	0.060/3659	0.125/1914	0.060/2440
$E_0 + 14\hbar\omega$	0.292/5135	0.102/2311	0.125/1281	0.102/1541
$E_0 + 16\hbar\omega$	0.221/3645	0.100/1349	0.108/763	0.100/899
$E_0 + 18\hbar\omega$	0.192/2444	0.090/742	0.090/427	0.090/494

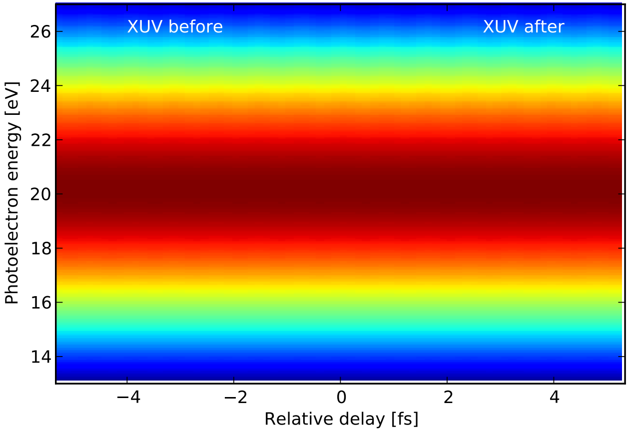
If we know the amplitudes and phases then we can compute τ_{Atom} and deduce the group delay of the attopulses τ_{GD} in experiments.

RABBITT METHOD: [Paul et al. SCIENCE 1690 **292** (2001)]

What if we have isolated attosecond pulses?

Photoelectron spectrogram

Overlap XUV continuum and phase-locked laser light:



Laser-assisted redistribution of the photoelectron peak.
Depends on the instantaneous laser field.

See for instance: [Kienberger *et al.*, NATURE **427**, 817 (2004)] where isolated 250-as XUV pulses were detected.

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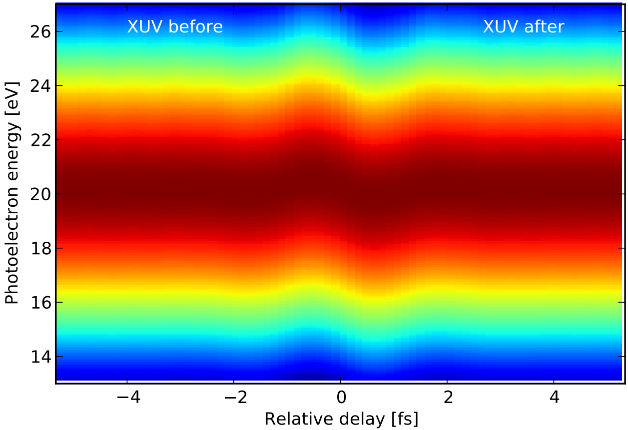

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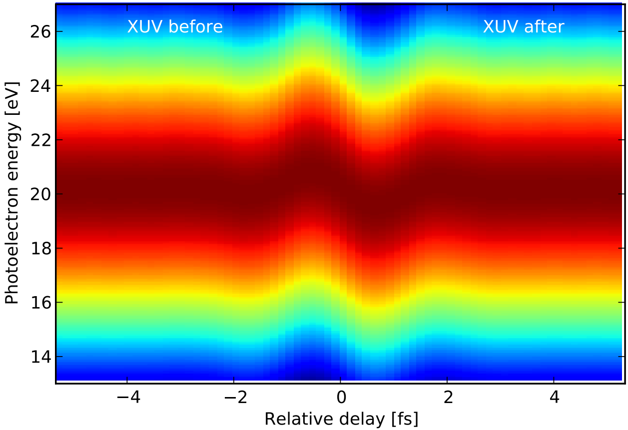

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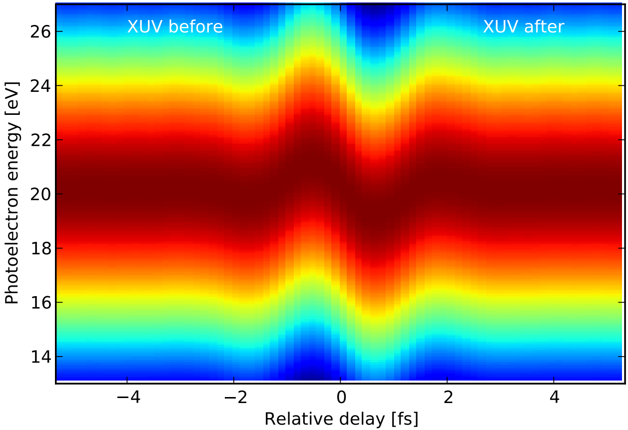

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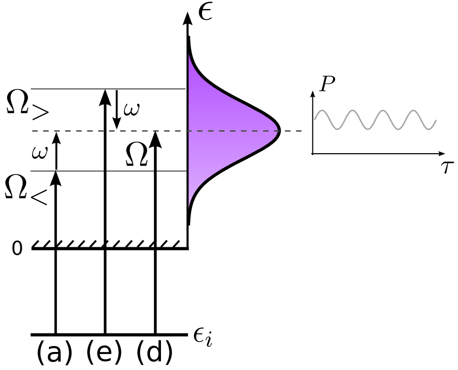
Temporal characterization of XUV continuum

Laser field will induce complicated interference effects



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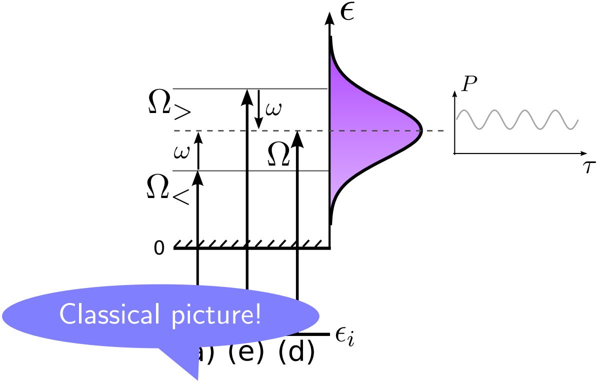


Laser-driven "streaking" of photoelectrons

$p_f \approx p_0 - A(t_0)$, where $A(t)$ is the vector potential.

See for instance: [Kienberger *et al.*, NATURE **427**, 817 (2004)] where isolated 250-as XUV pulses were detected.

Quantum mechanical treatment of Laser-assisted Photoionization by XUV pulse

- **Let's approximate the continuum states by Volkov states!**

Amplitude for final momentum k:

$$c_{\mathbf{k}}(t) = \overbrace{\frac{1}{i}\langle\, \phi_{\mathbf{k}} \mid \hat{p}_z \mid g \,\rangle}^{\text{Indep. of } t \text{ and } k?} \int_{-\infty}^{t} dt' \underbrace{A_X(t')}_{\text{XUV at } t'} \, e^{i\int^{t'} dt'' \overbrace{\left\{\frac{[\mathbf{k}+\mathbf{A}_L(t'')]^2}{2} + I_p\right\}}^{\text{Instantaneous energy}}}$$

* [M Kitzler, N Milosevic, A Scrinzi, F Krausz, and T Brabec PRL **88**, 173904 (2002)]

FROG-CRAB = Frequency Resolved Optical gating for Complete Reconstruction of Attosecond Bursts

[PRA, **71**, 11401 (2005) (Mairesse* and Quéré)] *IUPAP winner

Problem solved?

<u>FROG-CRAB</u> = Frequency Resolved Optical gating for Complete Reconstruction of Attosecond Bursts

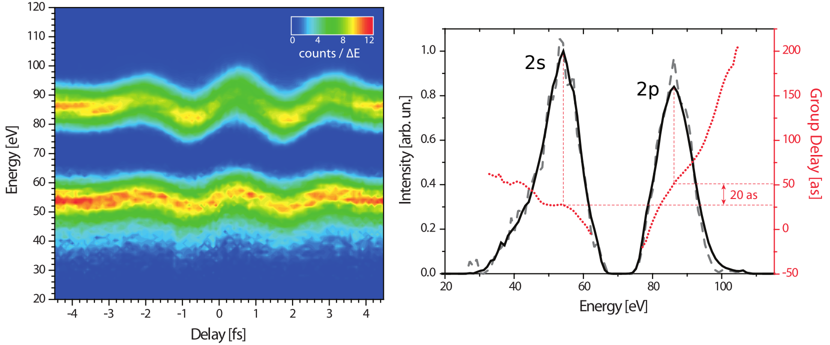
Iterative black-box method from ultra-fast optics adaptation using the Strong Field Approximation (SFA).

[PRA, **71**, 11401 (2005) (Mairesse* and Quéré)] *IUPAP winner

Is it OK to neglect all atomic effects?

Experiment: Laser-assisted photoionization delay in neon

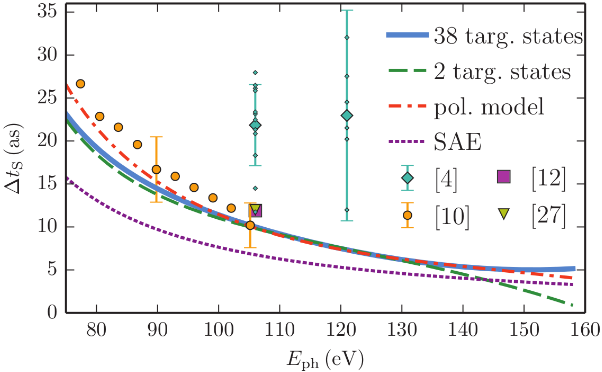
Test case for FROG-CRAB with relative $2p/2s$ measurement:



Reconstructed attosecond pulses
were **not the same**!

[Science **328**, 1658 (2010) (Schultze *et al.*)]

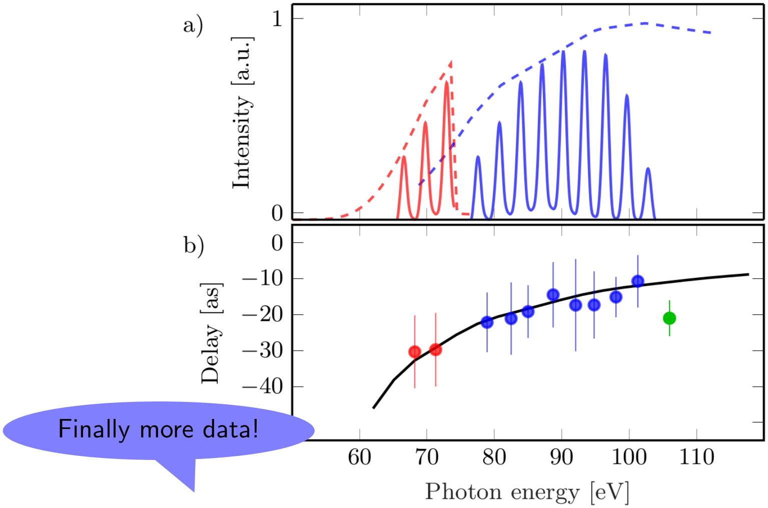
Comparison of neon experiment with theory



[4]	Schultze *et al.*	(2010)	Experiment with FROG-CRAB
[10]	Moore *et al.*	(2011)	Time-dependent R-matrix method
[27]	Dahlström *et al.*	(2012)	Many-body perturbation theory
[12]	Kheifets	(2013)	Hybrid: RPAE+CLC
[–]	Feist *et al.*	(2014)	Hybrid: MCHF+CLC+dLC

[Feist* *et al.*, Phys. Rev. A **89**, 033417 (2014)] *IUPAP winner

Neon $2p/2s$ delays revisited with RABBITT method



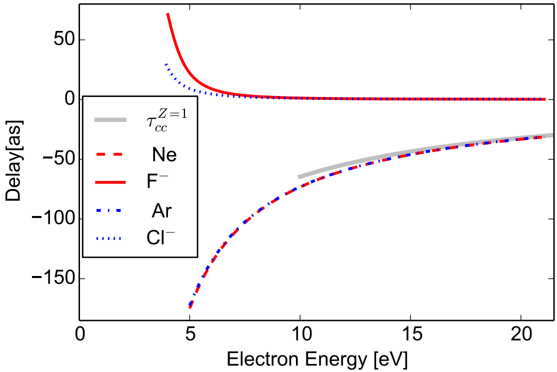
Experimental data from the group of **Prof. Anne L'Huillier (LTH)**.

Theory by Dahlström and Lindroth: Many-body perturbation theory.

What have we really learned since 2001?

Interpretation of the "atomic delay":

Atomic delay ≈ Wigner delay + CLC delay:



- Target-specific **Wigner delay** of photoelectron.
- **Universal CLC (or CC) delay** in **noble gas atoms**.

[Dahlström, L'Huillier and Maquet, JPB **45**, 183001 (2012)] [Lindroth and Dahlström, PRA **96**, 013420 (2017)]

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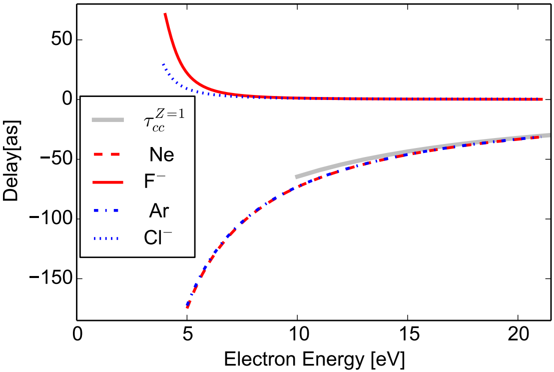
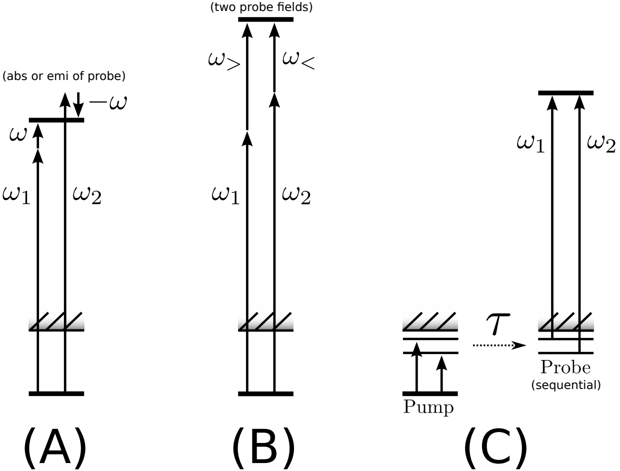


- Target-specific **Wigner delay** of photoelectron.
- In **negative ions** the CC delay is **small but not universal**!

[Dahlström, L'Huillier and Maquet, JPB **45**, 183001 (2012)] [Lindroth and Dahlström, PRA **96**, 013420 (2017)]

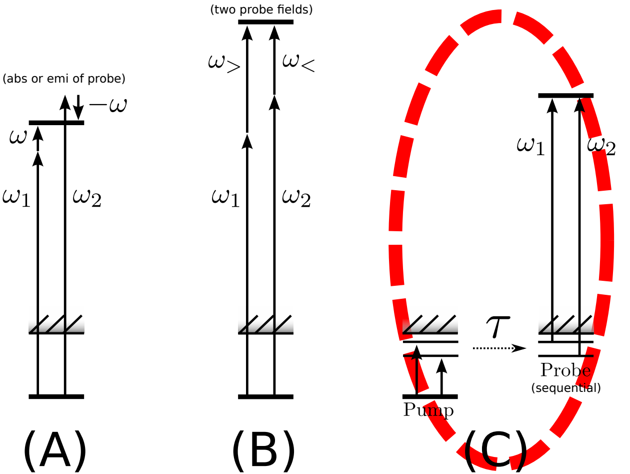
Can we think of a process with even shorter response time — to be able to measure the ultra short pulses of tomorrow?

Phase-sensitive multi-photon processes



- (A) Laser-assisted photoionization: RABBITT, FROG-CRAB...
- (B) Bichromatic probe fields: [You et al. PRA **93**, 033413 (2016)]
- (C) Pump+Probe: [Pabst and Dahlström PRA **94**, 013411 (2016)]

Phase-sensitive multi-photon processes



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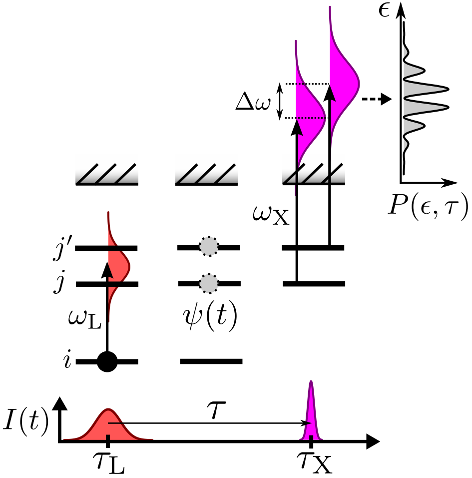
PANDA: Photoionization of bound wave packet



Basic steps:

- Bound wave packet.
- Sequential ionization (by attopulse at τ)
- All angles detection of photoelectron.

Spectral shearing interferometry → Precise characterization

[PRA, **94**, 013411 (2016)(Pabst and Dahlström)]

Spectral shearing by two-state wave packet

Phase difference between wave packet states j and j':

$$\Delta\phi = \underbrace{\omega_{j'j}\tau}_{\text{Propagation}} + \underbrace{\phi_X^{(jj')}(\epsilon_f) + \phi_D^{(jj')}(\epsilon_f)}_{\text{Ionization}}$$

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$$\text{Spectral phase difference}: \phi_X^{(jj')}(\epsilon_f) = \phi_X(\omega_{fj}) - \phi_X(\omega_{fj'})$$

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$$\text{Dipole phase difference}: \phi_D^{(jj')}(\epsilon_f) = \arg[d_{fj}] - \arg[d_{fj'}]$$

Spectral shearing by two-state wave packet

Phase difference between wave packet states j and j':

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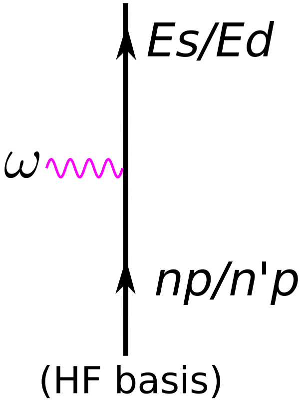
$$\text{Dipole phase difference}: \phi_D^{(jj')}(\epsilon_f) = \arg[d_{fj}] - \arg[d_{fj'}] \ \sim ???$$

Case study: Alkali atoms

Example: Potassium (K)
$[\mathrm{Ar}]4s^1 \to 4p/5p \to E\ell,\ m=0$

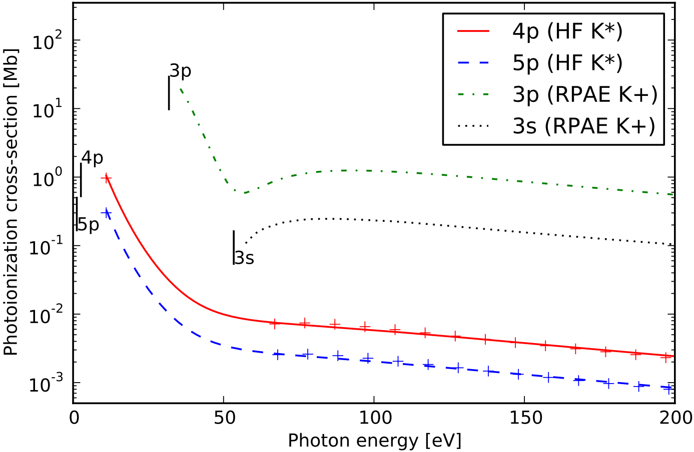
HF energies*:

$$
(\mathrm{K}^+\ \text{core}) \begin{cases} \ldots \\ \epsilon_{2p} = -319.4\,\mathrm{eV} \\ \epsilon_{3s} = -53.40\,\mathrm{eV} \\ \epsilon_{3p} = -31.86\,\mathrm{eV} \\ \hline \epsilon_{4s} = -3.996\,\mathrm{eV} \\ \epsilon_{4p} = -2.600\,\mathrm{eV} \\ \epsilon_{5p} = -1.240\,\mathrm{eV}, \end{cases}
$$



***STOCKHOLM CODE:** (JM Dahlström and E Lindroth (2014) J. Phys. B: At. Mol. Opt. Phys. **47** 124012)

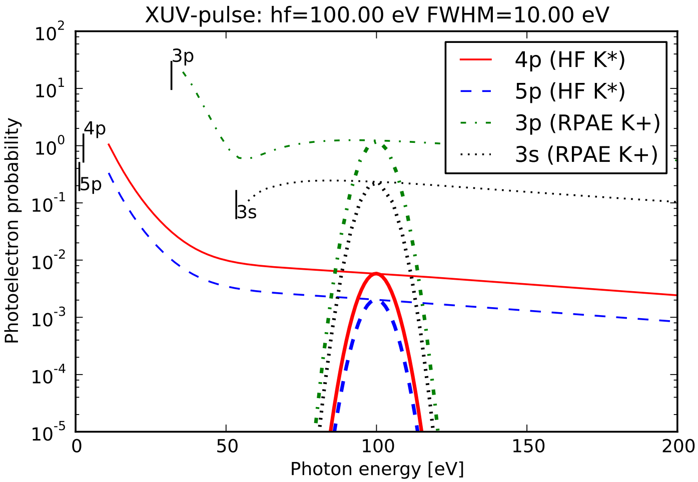
Partial photoionization cross section for K*



Photoionization of excited valence states ($4p$ and $5p$) is *not* strongly affected by inner-core polarization ($3p$ and $3s$).

'Characterization of attosecond pulses in the soft x-ray regime' [JPB, **50**, 104002 (2017)(Pabst and Dahlström)]

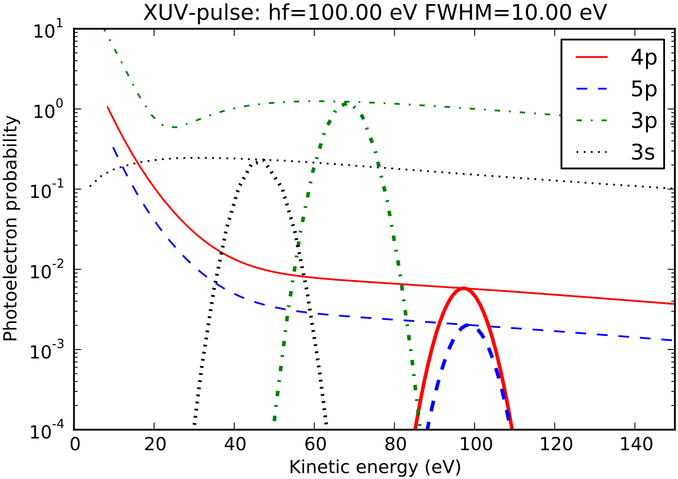
Partial absorption for K*



Example: 100 eV pulse with 10 eV bandwidth: Most of the ionization comes from inner-shell.

'Characterization of attosecond pulses in the soft x-ray regime' [JPB, **50**, 104002 (2017)(Pabst and Dahlström)]

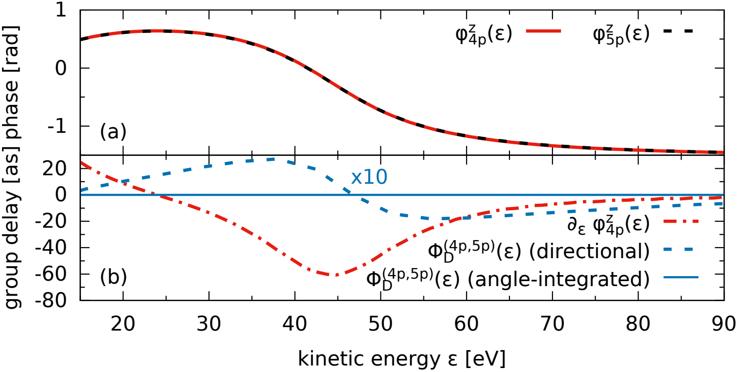
Partial photoelectron distributions for K*



Inner-shell contributions separate in energy and show *no* delay modulations (for independent particles).

'Characterization of attosecond pulses in the soft x-ray regime' [JPB, **50**, 104002 (2017)(Pabst and Dahlström)]

Summary of results for K*



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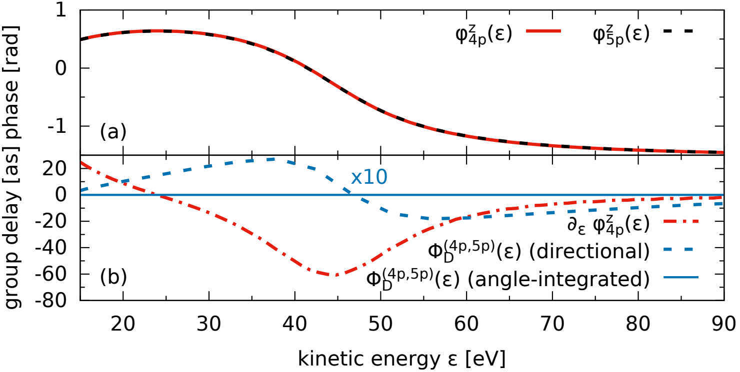


$$\text{Wigner delay}: \tau_W^{(4p)} \equiv \partial_\epsilon \varphi_{4p}^z$$

$$\text{z} - \text{direction}: \tau_z^{(4p,5p)} \equiv \frac{\varphi_{4p}^z - \varphi_{5p}^z}{\omega_{5p} - \omega_{4p}}$$

$$\text{all} - \text{directions}: \tau^{(4p,5p)} \equiv 0$$

Various attosecond test pulses

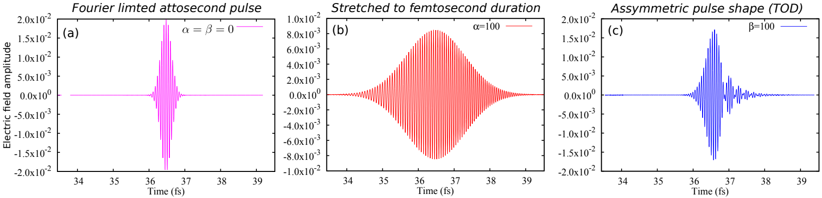
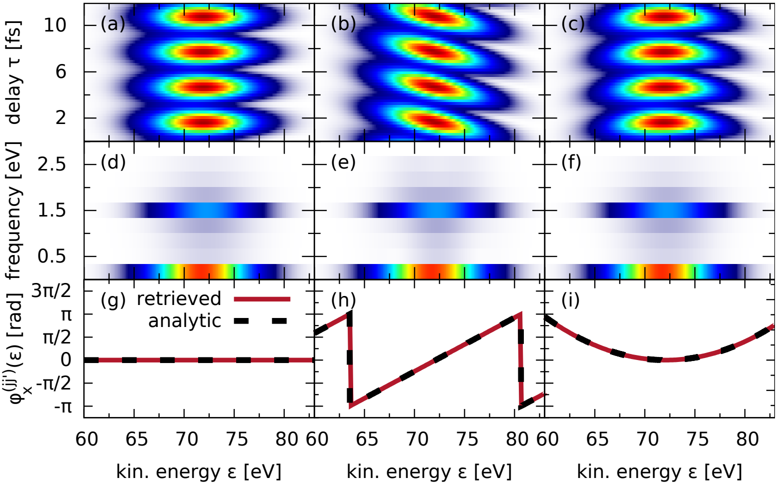


Figure: (a) Fourier limited (b) Linear chirp (c) 3^{rd} order dispertion.

Time-dependent PANDA calculations



Ionization of the K^* : $4p/5p$-wave packet
used to **characterize different attosecond pulses**.

Calculation using **XCID** program within the HF approximation.

Effects beyond HF

- **Core polarization - complex dipole elements:**
  K*: RPAE $(3p^{-1}E\ell)4p/5p \to E\ell$ kick out to valence (∼as).
- **Coupling to autoionizing state - Fano resonance (∼fs):**
  Ne*: TDCIS $2p^{-1}\,3s/4s \to 2p^{-1}\,Ep \leftrightarrow 2s^{-1}\,3s$ (∼as)
- **Auger delay/Fluorescence - decay of inner-shell hole:**
  Breit-Wigner distribution $\ll$ w.k.p. energy separation.
- **Shake up (soft x-ray range):**
  Kr*: Hartree-Slater $(3d^{-1}E\ell)4p^{-1}5s/6s \to 6s/7s$ (30%)

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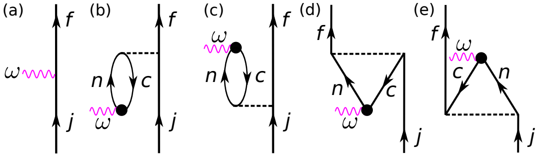
- **Shake up (soft x-ray range):**
  Kr*: Hartree-Slater $(3d^{-1}E\ell)4p^{-1}5s/6s \rightarrow 6s/7s\,(30\%)$
  Two ways to reach $(3d^{-1}E\ell)4p^{-1}6s$ implies interference!

'Characterization of attosecond pulses in the soft x-ray regime' [JPB, **50**, 104002 (2017)(Pabst and Dahlström)]

Effects beyond HF: Core polarization (RPAE in K*):

K wave packet: $4p$ and $5p$
photoionized with virtual excitation of $3p$ and $3s$ core electrons:

$$D_{fj} \approx \overbrace{d_{fj}^{(\mathrm{HF})}}^{\mathrm{Real}} + \overbrace{\Delta d_{fj}^{(\mathrm{RPAE})}}^{\mathrm{Complex}}$$



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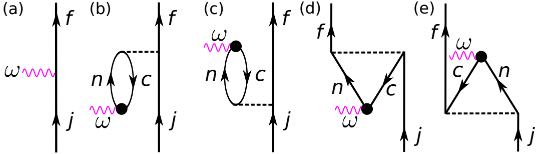
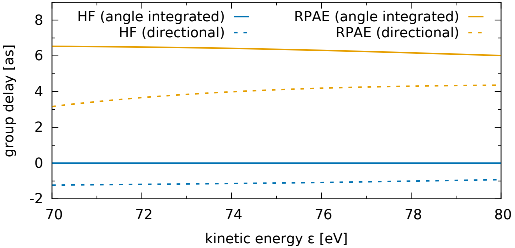
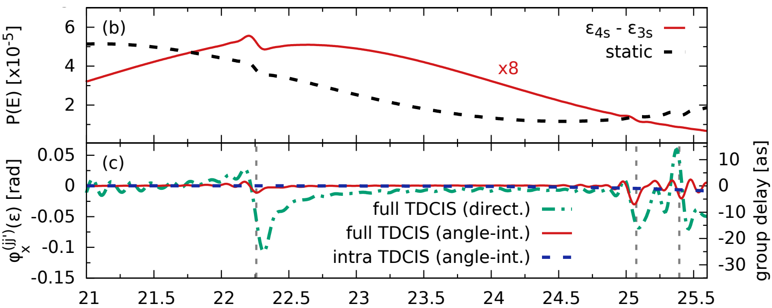




Effects beyond HF: Fano resonance

Ne*: $2p^{-1}\,3s/4s \rightarrow 2p^{-1}\,Ep \leftrightarrow 2s^{-1}\,3s$:



Comparison: Response $<$ 3 as — resonance lifetime 6.4 fs.

$2p^{-1}3s$	16.8 eV
$2p^{-1}4s$	19.7 eV
$2p^{-1} \rightarrow 2s^{-1}$	25.9 eV

Calculation using **XCID** program within the TDCIS approximation.

[PRA, **94**, 013411 (2016) (Pabst and Dahlström)]

Role of Fano resonance: *Interpretation*

Fano theory (one resonance $2s^{-1}3s$ and one continuum $2p^{-1}Ep$):

$$D_{fj} = \underbrace{d_{fj}}_{\text{Real}} \times \underbrace{\frac{(q_j + \epsilon)}{(1 - i\epsilon)}}_{\text{Complex}}, \;\; \epsilon = (E - E_\varphi - F)/(\Gamma/2)$$

where $q_j \in R$ is the Fano parameter.

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Phase difference between initial $j = 2p^{-1}3s$ or $j' = 2p^{-1}4s$:

$$\arg\left[\frac{D_{fj}}{D_{fj'}}\right] = \left[\frac{d_{fj}}{d_{fj'}} \times \frac{(q_j + \epsilon)}{(q_{j'} + \epsilon)}\right] = N\pi, \text{ if } q, d \in R.$$

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According to Fano theory there should be no delay!

Conclusions:

- *Long-standing Ne attosecond delay problem solved?*
- **Attosecond pulse characterization** by **photoionization** of a **bound wave packet**.
- Within the independent particle approximation the **method is lag free**.

Future directions:

- Photoionization delays:
  -**Negative ions** → **Wigner delay** [Lindroth & Dahlström PRA (2017)]
- Pulse characterization using bound wave packets:
  -**Soft x-ray regime** (*e.g.* shake-up) [Pabst & Dahlström JPB (2017)]
  -**Transient Abs. Spec.?** [Dahlström, Pabst & Lindroth Submitted (2017)]

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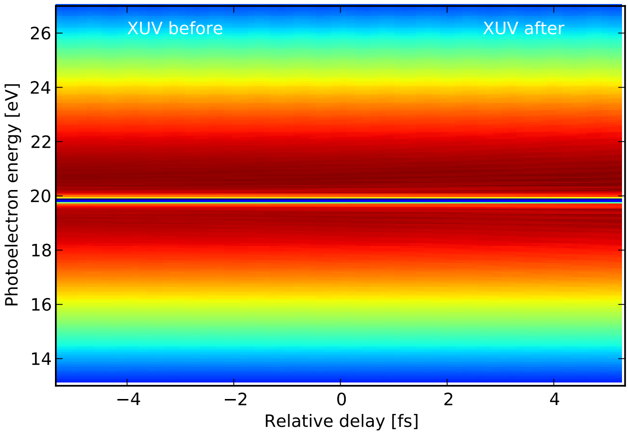
PANDA:

Pulse Analysis by Delayed Absorption

Photoelectron spectrogram with window resonance



Laser-assisted redistribution of autoionization.

Theory of laser-assisted autoionization by attosecond light pulses

[PRA, **71**, 060702 (2005) (Zhao and Lin)]

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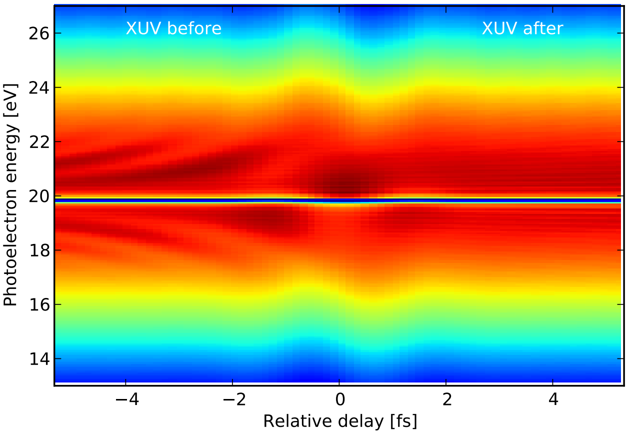

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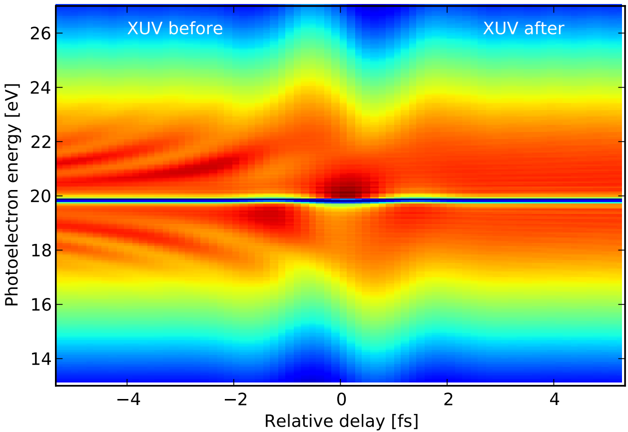

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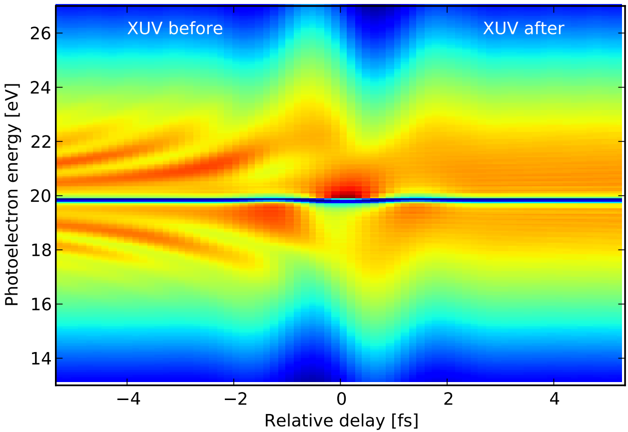

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Explicit form of Fano parameter:

$$q_j = \frac{\langle \varphi \mid T \mid j \rangle + \mathrm{p.v.} \int dE' \langle \varphi \mid H \mid \psi_{E'} \rangle \langle \psi_{E'} \mid T \mid j \rangle / (E - E')}{\pi \langle \varphi \mid H \mid \psi_E \rangle \langle \psi_E \mid T \mid i \rangle}$$

$$F(E) = \mathrm{p.v.} \int dE' \frac{|V_{E'}|^2}{E - E'}$$

Why is the quantum beat-delay removed completely with angle-integrated photoelectron detection?

Quantum beats with / without angular resolution

With angular resolution:

(wave packet: i and i' are $4p, 5p$, $m_L = 0$)

$$P(\mathbf{k},\tau) = \frac{1}{k} \sum_{i} \sum_{i'} c_i c_{i'}^* \sum_{L=s,d} \sum_{L'=s,d} i^{L-L'} Y_{L'0}(\hat{\mathbf{k}}) Y_{L0}^*(\hat{\mathbf{k}})$$

$$\times\, e^{-i\eta_L + i\eta_{L'}} d_{fi} d_{f'i'}^* E_X(\omega_{fi},\tau) E_X^*(\omega_{fi'},\tau)$$

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Without angular resolution:

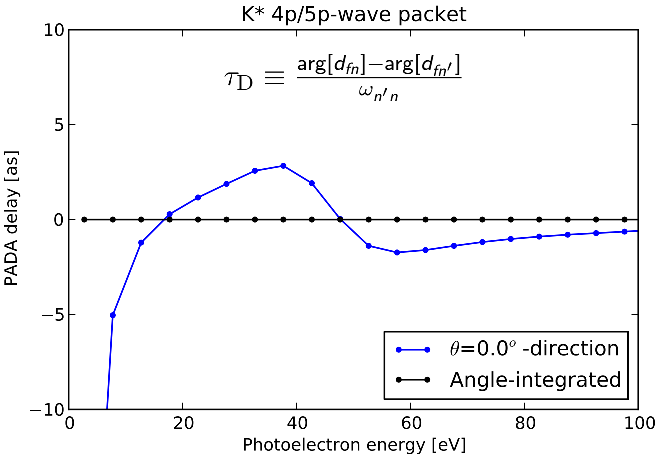
(terms with $L \neq L'$ are removed)

$$P(\epsilon,\tau) = \int d\Omega_{\mathbf{k}} P(\mathbf{k},\tau)$$

$$\sum_i \sum_{i'} \sum_{L=s,d} c_i c_{i'}^* d_{fi} d_{fi'}^{(*)} E_X(\omega_{fi}, tau) E_X(\omega_{fi'},\tau),$$

no dependence on the scattering phases $\eta_L(\epsilon)$!

Delay in K* with 4p/5p-wave packet



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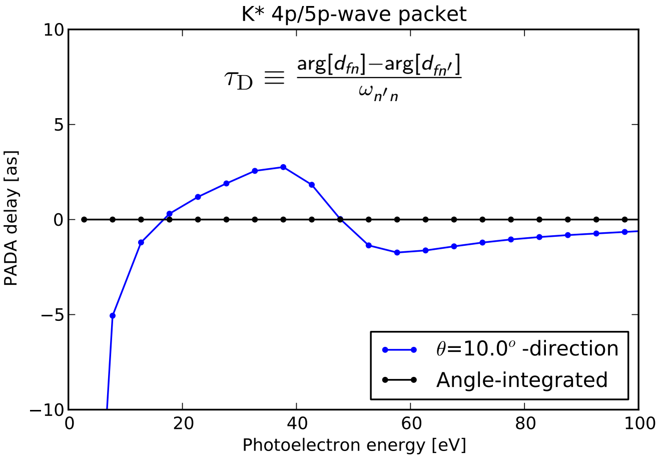

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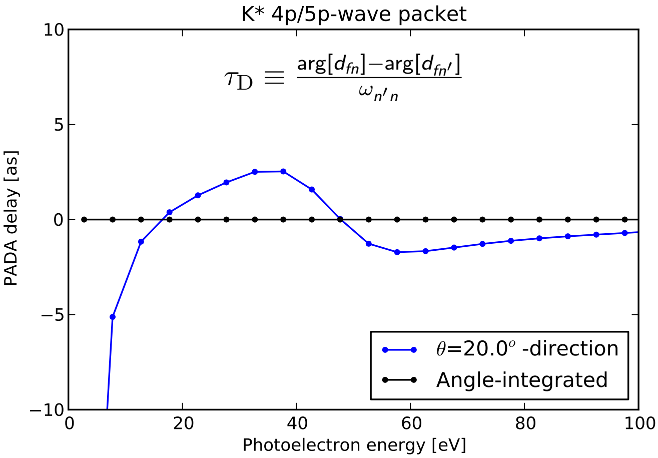

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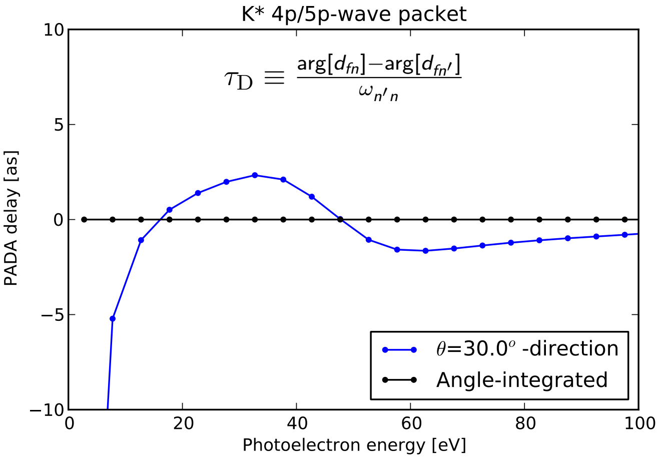

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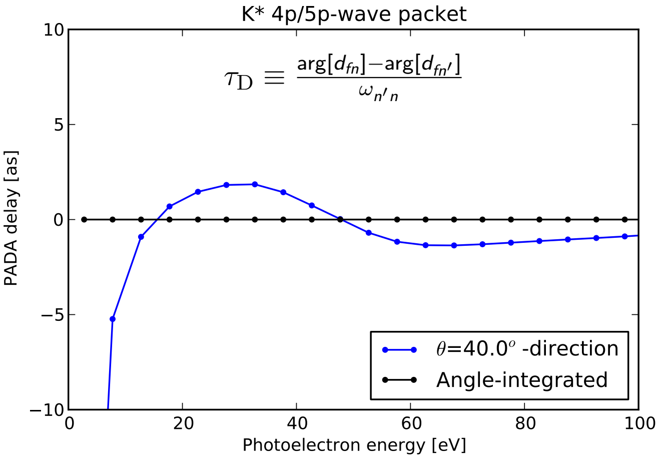

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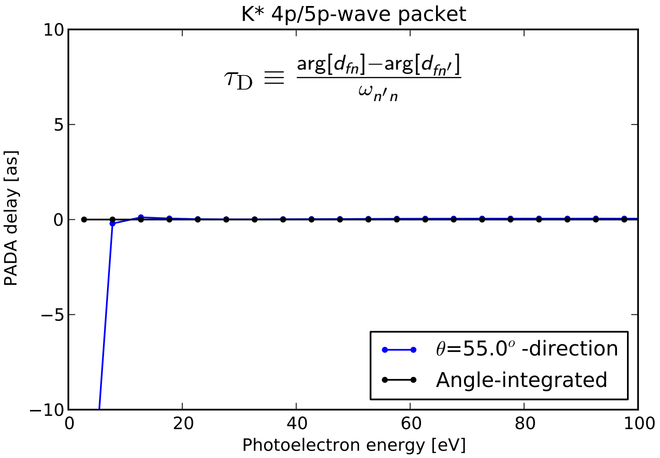

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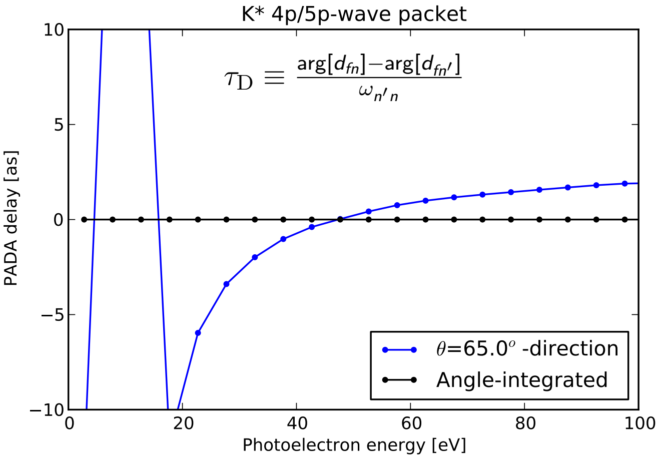

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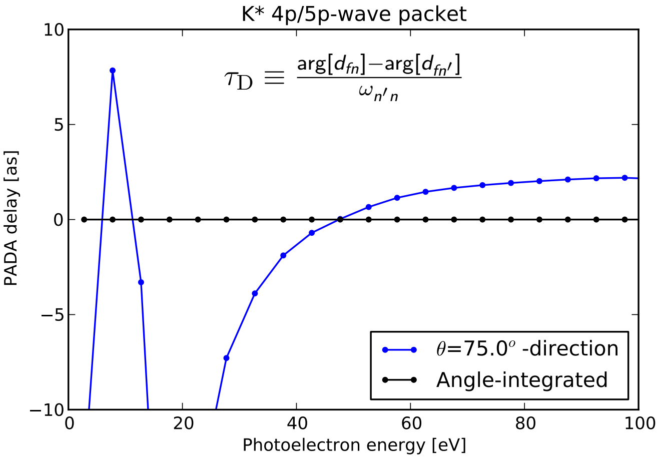
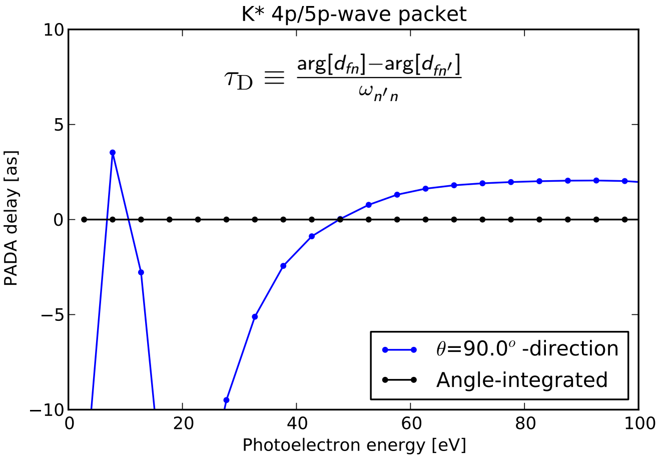
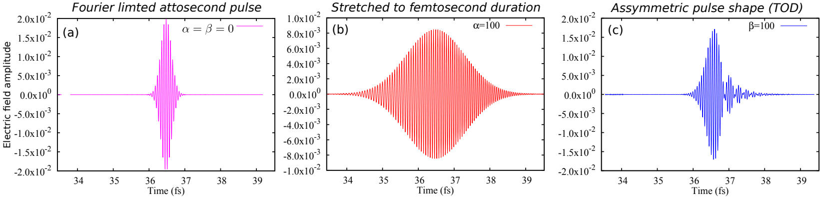


Delay in K* with 4p/5p-wave packet



Case study with different attosecond pulses

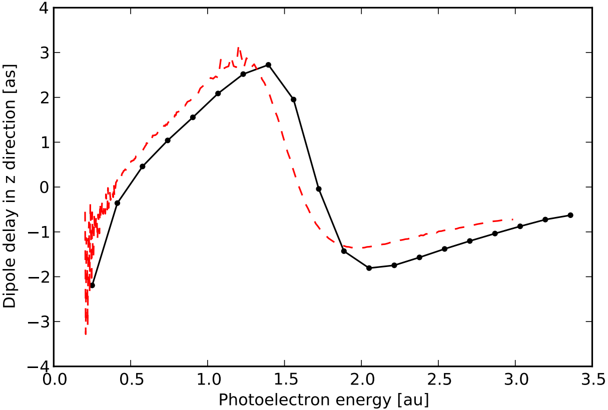
Q: Can the spectral phase difference be retrived by quantum beats?



$$E_1(\omega) = E_1^{(0)} \exp[-(\omega-\omega_1)^2/\Delta\omega_1^2]$$
$$\times \exp[i\alpha(\omega-\omega_1)^2 + i\beta(\omega-\omega_1)^3]$$

Delay of quantum beats from 4p/5p wave packet in K*

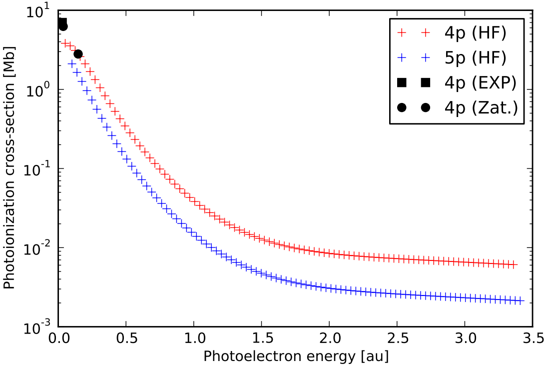
Comparison of static HF and time-dependent [XCID] methods



[Time-dependent picture] Delay extracted from **XCID** simulations.

[Static picture •] Delay: $\tau_D \equiv \frac{\arg[d_{fj}] - \arg[d_{fj'}]}{\omega_{j'j}}$ with $\hat{\mathbf{k}}_\mathbf{f} = \hat{\mathbf{z}}$.

Photoionization from excited Potassium

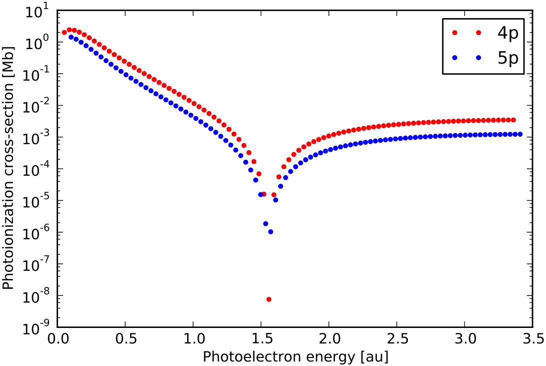


Photoionization cross-section:

$$\sigma_{\rm ph}(E)[{\rm Mb}] = a_0^2 \times 4\pi^2 \alpha\omega \sum_{L_f = s,d} |z_{fi}|^2$$

Experiment: Petrov at al. Eur. Phys. J. D **10**, 53-65 (2000)
Theory (pol. pot.): Zatsarinny and Taval PRA **81**, 043423 (2010)

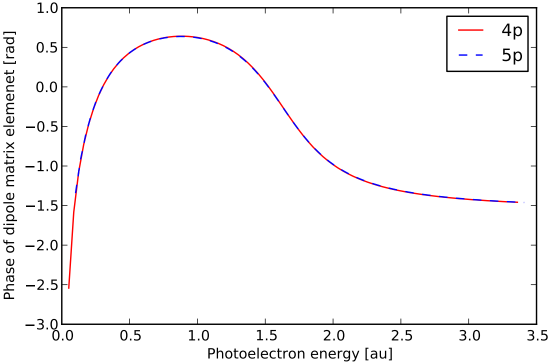
Partial d-wave ionization from excited Potassium



Cooper minimum in cross-section for partial d-wave:

$$\sigma_{\mathrm{ph}}(E)[\mathrm{Mb}] = a_0^2 \times 4\pi^2 \alpha\omega |z_{fi}|^2, \; L_f = d$$

Photoionization *phase* from excited Potassium

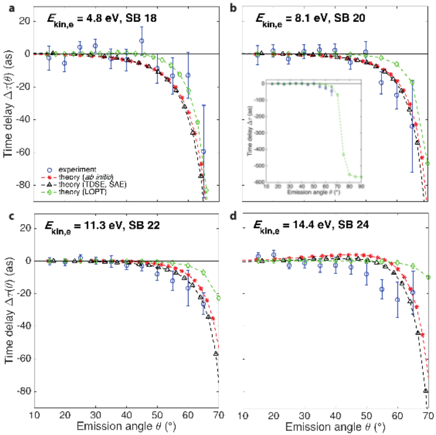


Phase of $\langle f \mid z \mid i \rangle$, $i = 4p, 5p$ to final momentum state:

$$| f \rangle = \psi^-_{\mathbf{k}}(\mathbf{r}) = \frac{1}{k^{1/2}} \sum_{L=0}^{\infty} \sum_{M=-L}^{L} i^L e^{-i\eta_L} Y^*_{LM}(\hat{\mathbf{k}}) Y_{LM}(\hat{\mathbf{r}}) R_E(r)$$

with energy E and scattering phases $\eta_L(E)$ with $\hat{\mathbf{k}} = \hat{\mathbf{z}}$.

Helium 1s angle-resolved delays with RABBITT method



Experimental data from the group of **Prof. Ursula Keller (ETH)**.
Theory by Dahlström and Lindroth: LOPT = Many-body pert. theory.

[REFERENCE!]

When can the atomic response be neglected?

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It depends strongly on the target and detection method!

Estimate for neon from 2p state at 50 eV:

$c \approx 0.5\,\mathrm{as/eV} \rightarrow \delta t_{\mathrm{crit}} \approx 30\,\mathrm{as}$

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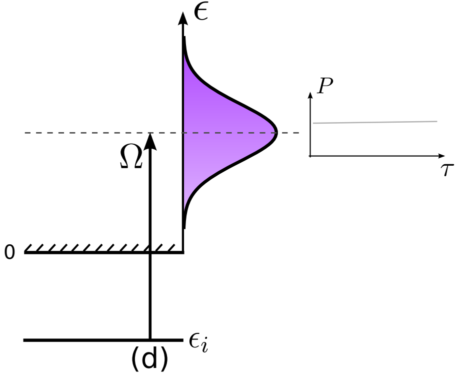
Actual duration	Reconstructed duration
100 as	100.4 as
30 as	42.4 as

[Pabst and Dahlström PRA **94**, 013411 (2016)]

Temporal characterization of coherent XUV continuum

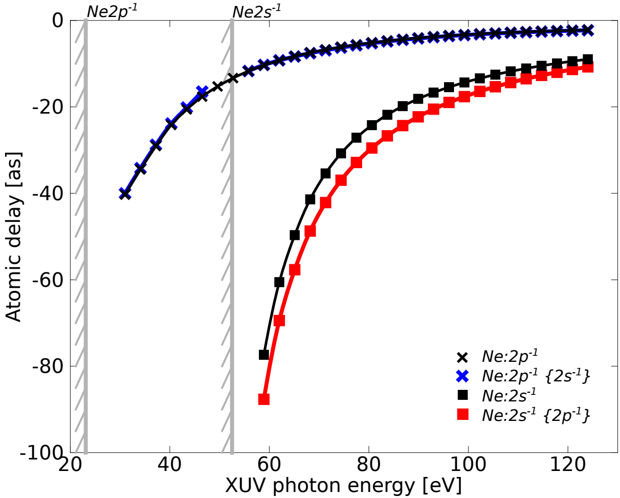
No temporal information by one-photon ionization



Broad photoelectron peak:

Centered at $\epsilon = \Omega - I_p$ with $\Delta\Omega > \omega$.

Theory: Laser-assisted photoionization delay in neon



[Dahlström, Carette and Lindroth, Phys. Rev. A, **86**, 061402(R) (2012)]