Attosecond time-delays in photoionisation

J. Marcus Dahlström

2023-06-06 Attosecond General Group Meeting (Freiburg).

Outline of lecture:

- *Review of attosecond pulse characterization*
  – Simple models based on SFA*
- *How large is the atomic response?*
  – Argon photoionization delay experiment
  – Delays in other noble gas atoms
- *How can we interpret the atomic delays?*
  – Coulomb potential and laser field
  – Many electron effects ("Feynman diagrams")
  – Autoionization processes
- *Conclusion and Outlook*

* SFA=Strong Field Approximation

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- Problems for the PhD-studends (*Task* : *i*)
  http://www.matfys.lth.se/staff/Marcus.Dahlstrom/

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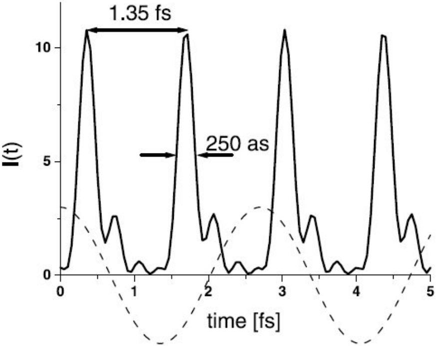
Observation of a Train of Attosecond Pulses from High Harmonic Generation

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)]

– “RABIT”, “RABBIT”, “RABITT” or “RABBITT”?

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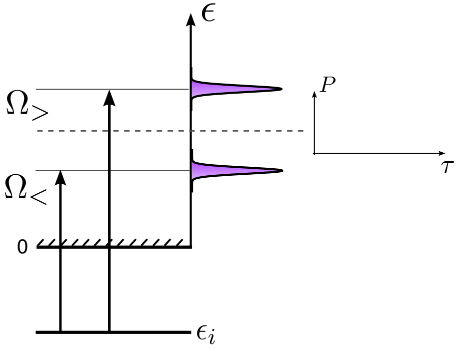
– Why is a laser field needed to characterize attopulses?

Linear interaction: $P(\epsilon) \sim |\Psi(\epsilon)|^2 \sim |E(\Omega)|^2$

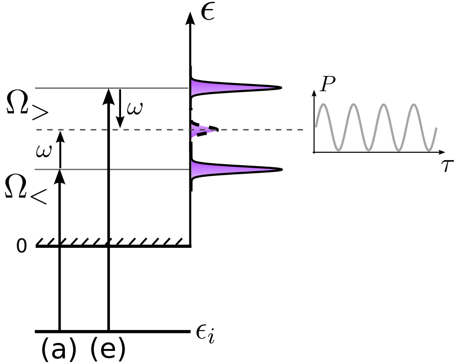
– No phase information about attopulses –



Photoelectron peaks due to absorption of one XUV harmonic photon

$$\Omega_{2q+1} = (2q+1)\omega$$

Spectral shearing by absorption/emission of laser photon
– How the phase of attopulse varies with energy –



Laser-induced sideband signal:

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

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

– How can the atomic delay, τ_{Atom}, be determined?
Is it important or negligible?

Model: atom in multi-color electromagnetic fields

Atomic units: $e = m = \hbar = 4\pi\epsilon_0 = 1$

Hamiltonian for interaction with field *and* ion:

$$H = H_V + V_A$$

Kinetic energy of electron in a *uniform* electromagnetic field:

$$H_V = \frac{1}{2}[\hat{\mathbf{p}} + \mathbf{A}(t)]^2$$

Vector potential of both attopulses and laser fields:

$$\mathbf{A}(t) = \mathbf{A}_X(t) + \mathbf{A}_L(t)$$

Atomic potential for hydrogen:

$$V_A(r) = -\frac{1}{r}$$

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Argon potential* within *single-active electron* approximation:

$$V_A(r) = -\frac{1}{r}(1 + 5.4e^{-r} + 11.6e^{-3.682r})$$

* **PT:** [E S Toma and H G Muller JPB **35**, 3435 (2002)] **TDSE:** [J Mauritsson et al. PRA **72**, 013401 (2005)]

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)			
	$(1,0) \rightarrow (1,0)$	$(1,0) \rightarrow (3,0)$	$(1,\pm 1) \rightarrow (1,\pm 1)$	$(1,\pm 1) \rightarrow (3,\pm 1)$
$E_0 + 12\hbar\omega$	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.

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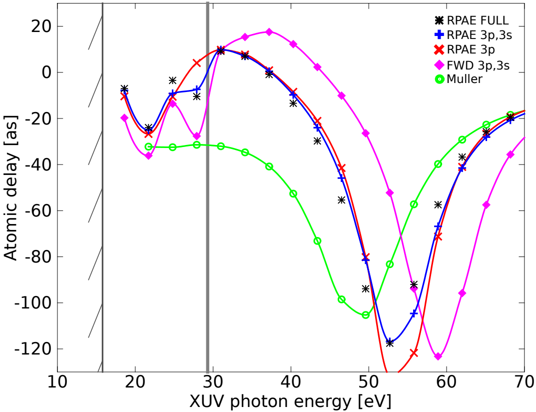
If we know the amplitudes and phases then we can compute τ_{Atom} and deduce the group delay of the attopulses τ_{GD} in experiments.
But how sure are we about this model? Can it be tested?

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

Study of correlation effects in $Ar3p^{-1}$

Experimental binding energies (not HF values):



- At 34.1 eV (SB22) the atomic delay is small (~ 5 as).
- The atomic delay exhibits a negative peak of ~ -120 as.

[J M Dahlström and E Lindroth JPB **47** 124012 (2014)]

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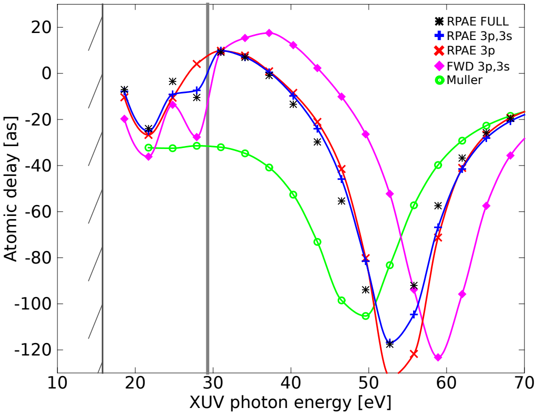
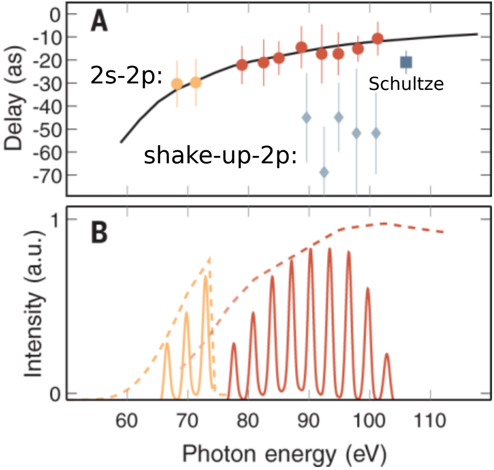


- At 34.1 eV (SB22) the atomic delay is small (~ 5 as).
- The atomic delay exhibits a negative peak of ~ -120 as.
- Electron correlation effects amount to ~ 40 as (Muller).

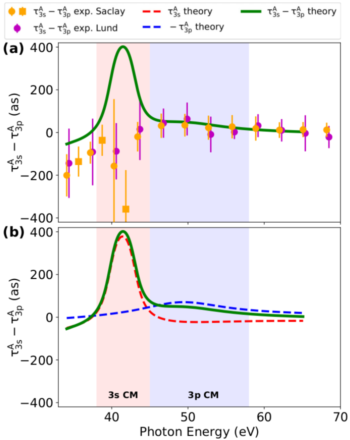
[J M Dahlström and E Lindroth JPB **47** 124012 (2014)]

Neon delay 2s-2p



M. Isinger et al., Science 10.1126/science.aao7043 (2017).

Argon delay 3s*-3p



*At low sidebands the shake-ups ($4p$ and $3d$) contribute to $3s$ signal.

C. Alexandridi et al. PRR 3, L012012 (2021).

– OK, "atomic delays" have been measured experimentally. Why is it so fascinating — what does it mean?

Probing Single-Photon Ionization on the Attosecond Time Scale

$$\tau_A = \tau_W + \tau_{CC}$$

''The determination of photoemission time delays requires taking into account the measurement process, involving the interaction with a probing infrared field. This contribution can be estimated using a universal formula and is found to account for a substantial fraction of the measured delay.''

[K. Klünder et al. PRL **106**, 143002 (5 April 2011)]

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Time-resolved photoemission by attosecond streaking: extraction of time information

''We show that attosecond streaking ... contain ... Eisenbud-Wigner-Smith time delay matrix ... if ... the streaking infrared (IR) field ... is properly accounted for ...'' [S Nagele et al. JPB. **44**, 081001 (11 April 2011)]

– Now back to the basics!

Simplest possible model for RABBIT

Atomic units: $e = m = \hbar = 4\pi\epsilon_0 = 1$

Assumption: Photoelectron is unaffected by atomic potential.

Plane wave:

$$\varphi_{\mathbf{k}}(\mathbf{r}) = \frac{1}{(2\pi)^{3/2}} \exp[i\mathbf{k}\cdot\mathbf{r}]$$

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Momentum eigenstate:

$$\hat{\mathbf{p}}\varphi_{\mathbf{k}} \equiv -i\nabla\varphi_{\mathbf{k}}(\mathbf{r}) = \mathbf{k}\varphi_{\mathbf{k}}$$

Solution to the free particle Schrödinger equation (SE):

$$H_0\varphi_{\mathbf{k}} = \frac{\hat{\mathbf{p}}^2}{2}\varphi_{\mathbf{k}} = \frac{k^2}{2}\varphi_{\mathbf{k}} \equiv \epsilon_k\varphi_{\mathbf{k}}$$

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Second-order perturbation theory*:

$$M_{\mathbf{k}}^{(2)} \approx \int d^3k' \frac{\langle\, \mathbf{k} \mid O \mid \mathbf{k}' \,\rangle\langle\, \mathbf{k}' \mid O \mid g \,\rangle}{(\epsilon_g + \omega - \epsilon_{\mathbf{k}'})}$$

Perturbation by external field (dipole approximation):

$$\text{Velocity}: O = \mathbf{A}(\omega) \cdot \hat{\mathbf{p}}$$
$$\text{Length}: O = \mathbf{E}(\omega) \cdot \mathbf{r}$$

Vector potential and electic field (uniform in space):

$$\tilde{\mathbf{E}}(t) = -\frac{\partial \tilde{\mathbf{A}}}{\partial t}$$

* In depth discussion: [A Jimenez-Galan, F. Martin and L. Argenti RPA **93**, 023429 (2016)]

Simplest possible model for RABBIT

Atomic units: $e = m = \hbar = 4\pi\epsilon_0 = 1$

$(Task : 1)$ Approximate two photon matrix element:

$$M_{\mathbf{k}}^{(2)} \approx -2A(\Omega)A(\omega)\frac{\epsilon_k}{\omega}\cos^2\theta_{\mathbf{k}}\,\langle\,\mathbf{k}\,|\,g\,\rangle$$

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(*Task* : 2) Projection of ground state ($1s$) on plane wave:

$$\langle\,\mathbf{k}\,|\,g\,\rangle = \frac{2^{3/4}}{\pi}\frac{I_p^{5/4}}{(I_p+\epsilon_k)^2},\quad I_p = \frac{Z^2}{2}$$

The two-photon matrix goes like $1/\epsilon_k$, $\epsilon_k \gg I_p$
and it is *real* within *plane-wave* approximation.

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$\rightarrow$ The atomic delay is zero!?

– What if the laser field is treated non-perturbatively?

Electron driven in a field (Volkov state)

Atomic units: $e = m = \hbar = 4\pi\epsilon_0 = 1$

Time-dependent Schrödinger equation (TDSE):

$$i\frac{\partial \psi}{\partial t} = H_V \psi(\mathbf{r}, t)$$

Volkov Hamiltonian (velocity gauge):

$$H_V = \frac{1}{2}\left[\mathbf{p} + \mathbf{A}(t)\right]^2$$

Ansatz using plane wave with time-dependent phase:

$$\psi_{\mathbf{k}}^{V}(\mathbf{r}, t) = \phi_{\mathbf{k}}(\mathbf{r}) \exp[-i\Phi_{\mathbf{k}}(t)]$$

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(*Task* : 3) Insert into TDSE to obtain the Volkov phase:

$$\Phi_{\mathbf{k}}(t) = \int_{\mathrm{ref.}}^{t} dt'\frac{1}{2}[\mathbf{k} + \mathbf{A}(t')]^2$$

Photoionization to laser dressed continuum

Laser-dressed time-dependent perturbation theory *

$$c_{\mathbf{k}}(t) = \frac{1}{i}\int_{-\infty}^{t} dt' A_X(t') \langle\, \Psi_{\mathbf{k}}^{V} \mid \hat{p}_z \mid \tilde{g}\, \rangle$$

where the conjugate Volkov state is

$$\Psi_{\mathbf{k}}^{V*}(\mathbf{r},t) = \phi_{\mathbf{k}}^{*}(\mathbf{r}) \exp[i\Phi_{\mathbf{k}}(t)]$$

and the ground state is with binding $I_p > 0$ is

$$\tilde{g}(\mathbf{r},t) = g(\mathbf{r}) \exp[-i\epsilon_g t] \equiv g(\mathbf{r}) \exp[iI_p t]$$

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

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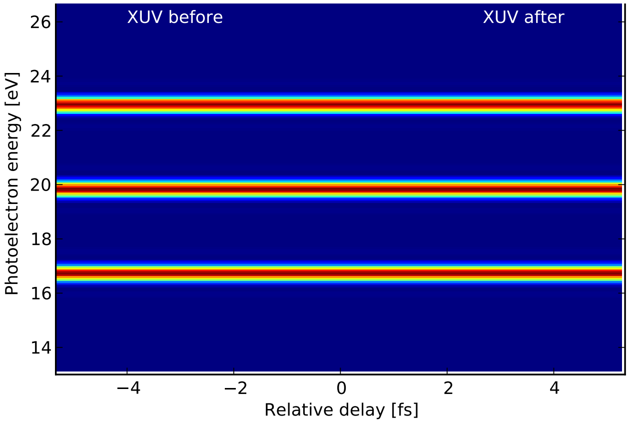
Amplitude for final momentum $\mathbf{k}$:

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

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

Photoelectron spectrogram

One photon absorption from XUV comb and dressing by laser field (Volkov approx.)



Redistribution of three harmonic peaks due laser dressing: Formation of sidebands.

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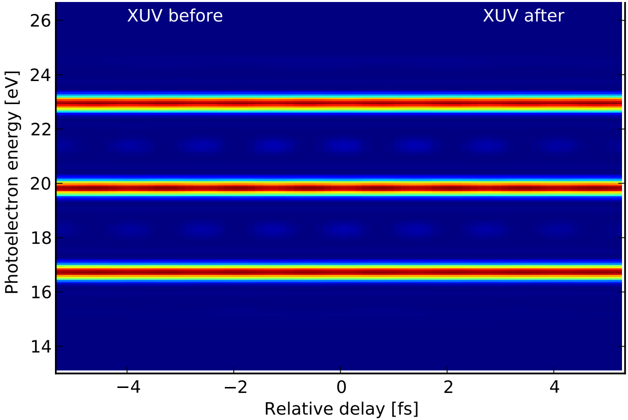

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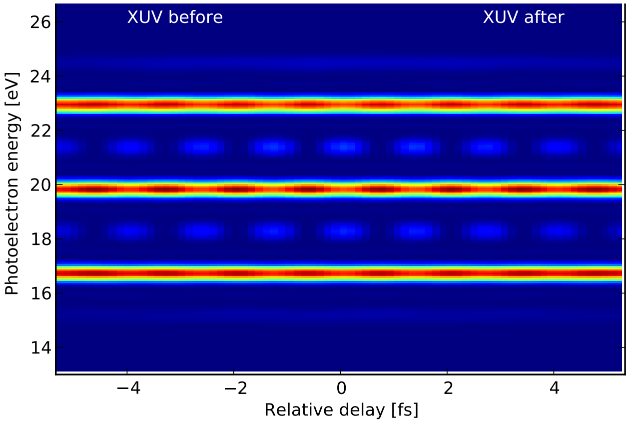

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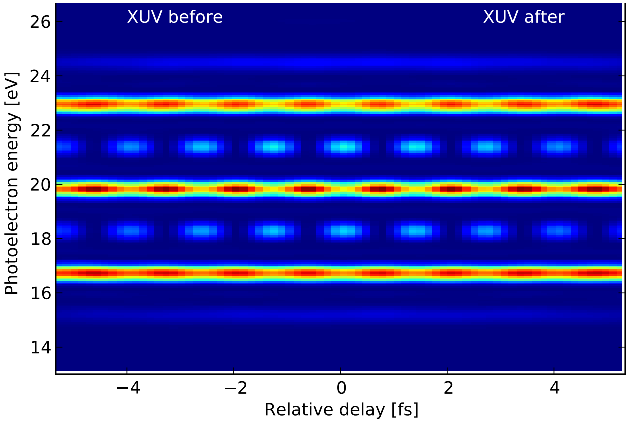

Redistribution of three harmonic peaks due laser dressing: Formation of sidebands.

– How does the photon picture arise?

Connection to the photon picture

Amplitude for laser-dressed one-photon ionization:

$$c_{\mathbf{k}}(t) = \frac{1}{i}\langle \varphi_{\mathbf{k}} | \hat{p}_z | g \rangle \int_{-\infty}^{t} dt' A_X(t') \exp i \int^{t'} dt'' \left\{ \frac{[\mathbf{k} + \mathbf{A}_L(t'')]^2}{2} + I_p \right\}$$

Assume weak laser $[\mathbf{k} + \mathbf{A}_L(t'')]^2 \approx k^2 + 2\mathbf{k} \cdot \mathbf{A}_L(t'')$ and slowly varying *laser* envelope $\Lambda_L(t)$ compared to laser oscillation ω_L with $A_L(t) = \Lambda_L(t) \sin \omega_L t$

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$$c_{\mathbf{k}}(t) \approx \frac{1}{i}\langle\,\varphi_{\mathbf{k}}\,|\,\hat{p}_z\,|\,g\,\rangle \int_{-\infty}^{t} dt' \frac{1}{2}\Lambda_X(t') \sum_{n=-\infty}^{\infty} (-i)^n J_n\left(\frac{\mathbf{k}\cdot\mathbf{\Lambda}_L(t')}{\omega_L}\right)$$
$$\times \exp[i(\epsilon_k + I_p - \omega_X + n\omega_L)t'] \quad (\textit{Task}: 4)$$

- Photon energy conservation given by exponential factor.
- Multiphoton transition determined by *real* Bessel function, J_n.

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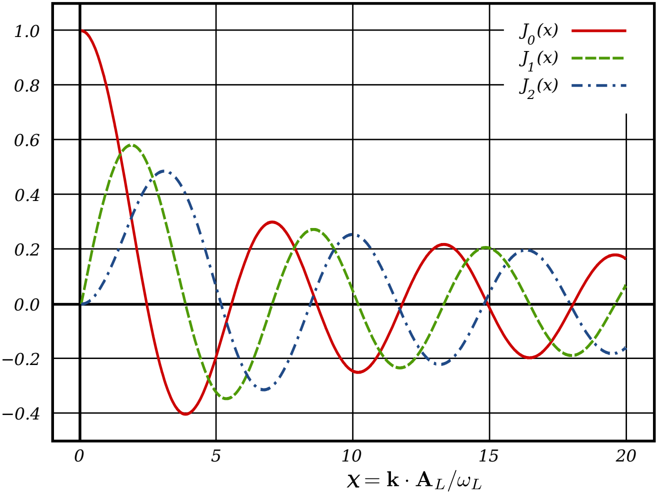
$\rightarrow$ The atomic delay is zero?!?!

Bessel functions



The Bessel functions describe the magnitude of laser stimulated continuum transitions as a function of $x = \mathbf{k} \cdot \mathbf{A}_L/\omega_L$.

Multiphoton interaction phase shifts

Assumptions:

- Rotating wave for XUV field.
- Constant IR envelope.

$$c_{\mathbf{k}} \approx \frac{1}{2} \sum_{n=-\infty}^{\infty} (-i)^n \exp[in\varphi] J_n \left(\frac{\mathbf{k} \cdot \mathbf{\Lambda}_{\omega,0}}{\omega} \right) \langle \mathbf{k} | \hat{p}_z | g \rangle \exp[i\delta]$$
$$\times \int \mathrm{d}t \; \Lambda_{\Omega}(t) \exp[i(\epsilon_k + I_p + U_p - \Omega - n\omega)t],$$

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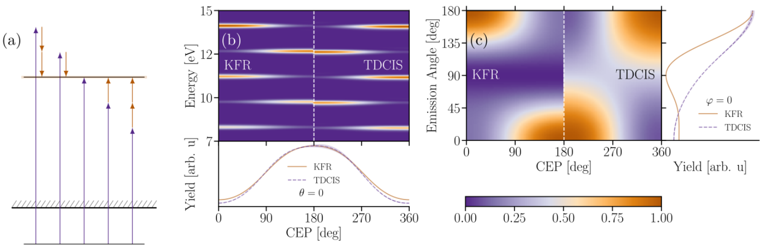
Model for photoelectron interferometry:

$$c_{\mathbf{k}}^{(n)} = (-i)^{|n|} J_{|n|}\left(\frac{\mathbf{k}\cdot\mathbf{\Lambda}_{\omega,0}}{\omega}\right) \exp[in\varphi] f_{\mathbf{k}g}(n).$$

$$f_{\mathbf{k}g}(n) = \frac{1}{2}\langle \mathbf{k}|\hat{p}_z|g\rangle \exp[i\delta] \int \mathrm{d}t\, \Lambda_\Omega(t) \exp[i(\epsilon_k + I_p + U_p - n\omega - \Omega)t],$$

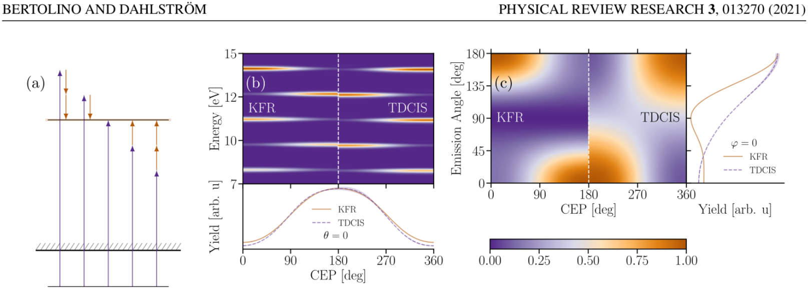
Coupling of ω harmonics (even and odd) from HHG

BERTOLINO AND DAHLSTRÖM PHYSICAL REVIEW RESEARCH 3, 013270 (2021)



Original experiment: Laurent et al. Phys. Rev. Lett. 109, 083001 (2012).

Coupling of ω harmonics (even and odd) from HHG



Simple model: $f_{odd} = f_{even} = 1$

$$P \approx |J_0 - iJ_1(e^{i\varphi} + e^{-i\varphi})|^2 = |J_0 - iJ_1 2\cos\varphi|^2$$

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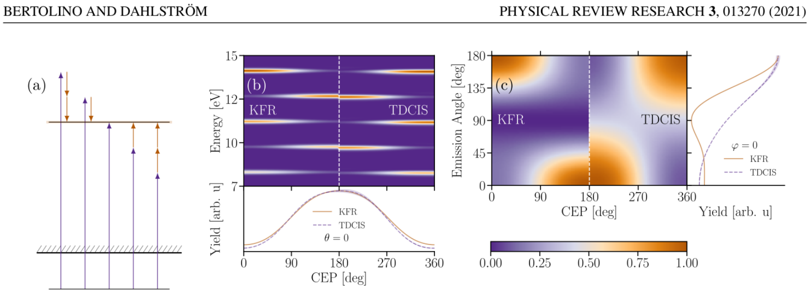

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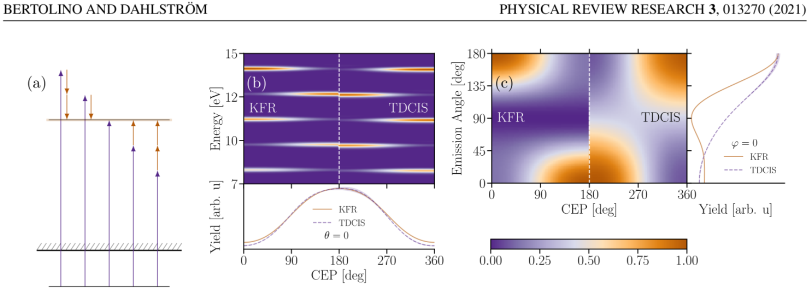

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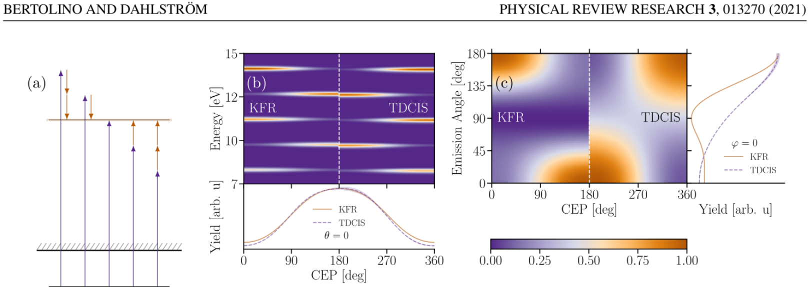

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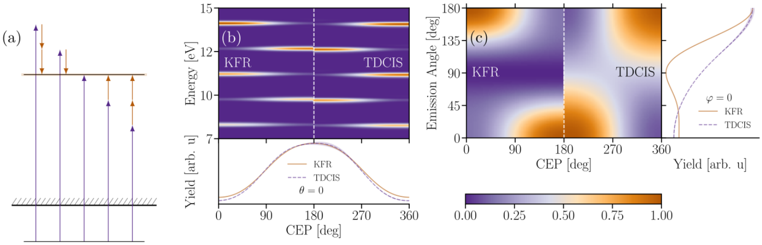
BERTOLINO AND DAHLSTRÖM PHYSICAL REVIEW RESEARCH 3, 013270 (2021)



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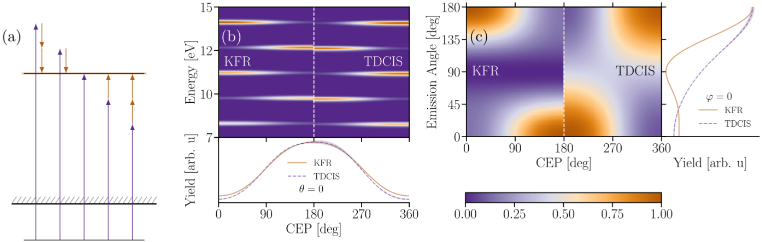

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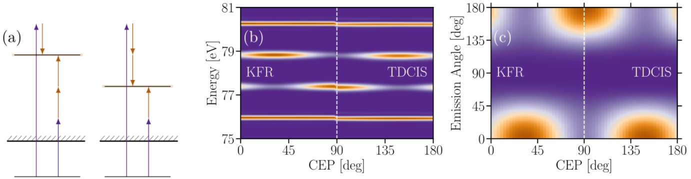
Coupling of 3ω harmonics from seeded FEL



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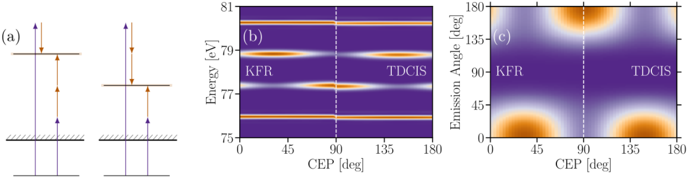
Coupling of 3ω harmonics from seeded FEL

MULTIPHOTON INTERACTION PHASE SHIFTS … PHYSICAL REVIEW RESEARCH **3**, 013270 (2021)



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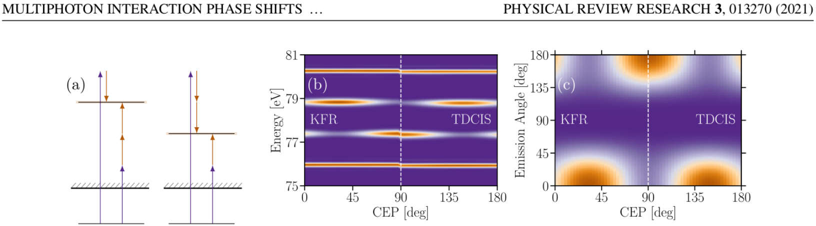

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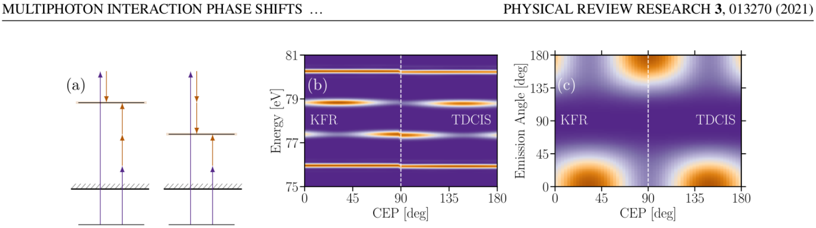

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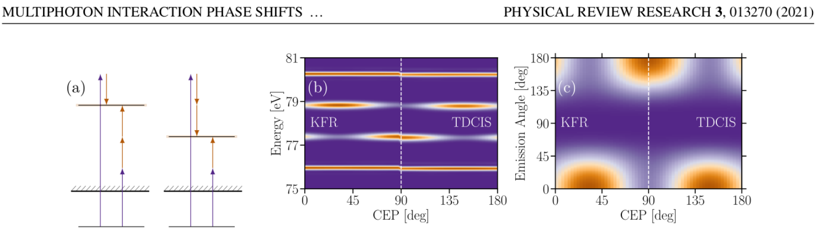

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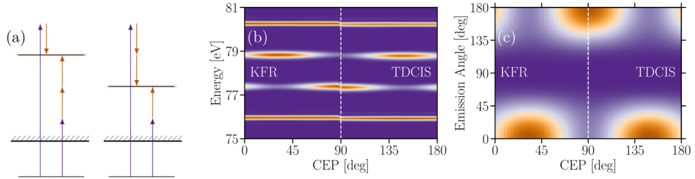

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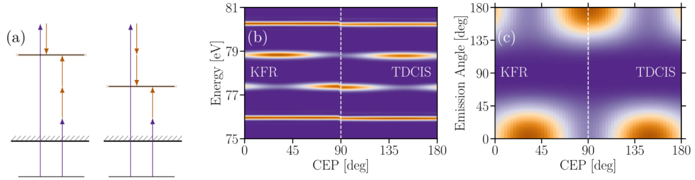

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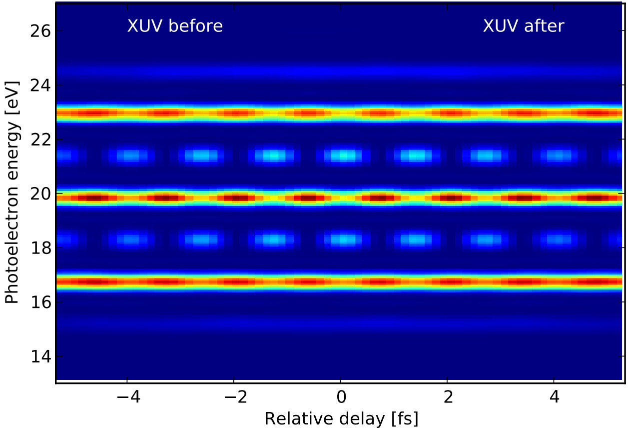
– Thank you!

See original lecture with all problems and solutions:
http://www.matfys.lth.se/staff/Marcus.Dahlstrom/

Photoelectron spectrogram

One photon absorption from XUV comb and dressing by laser field (Volkov approx.)



Redistribution of harmonics due to intense laser dressing.
Multi-photon processes amount to non-sinusoidal beating patterns.

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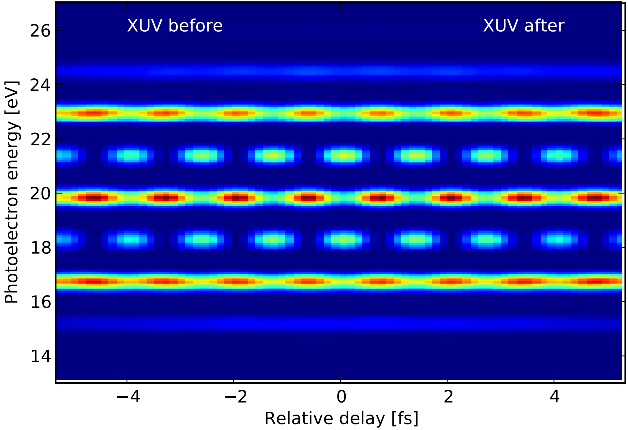

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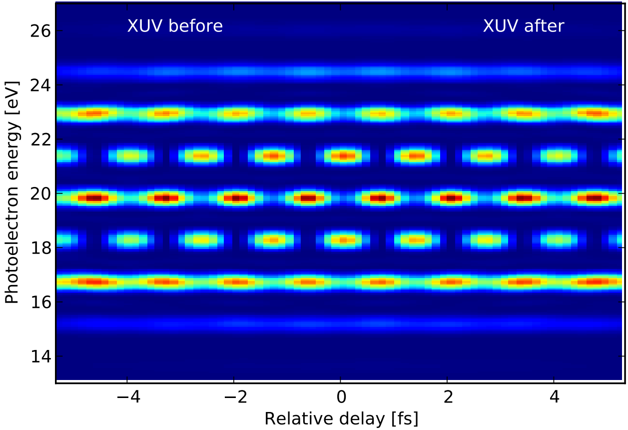

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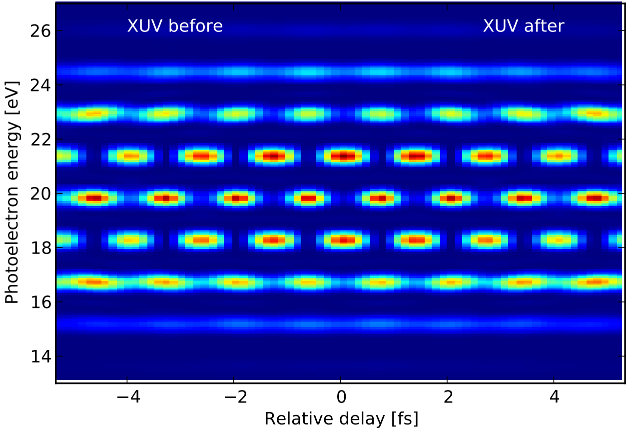

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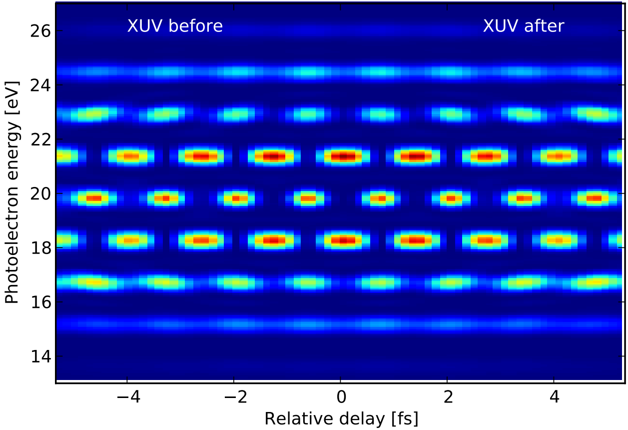

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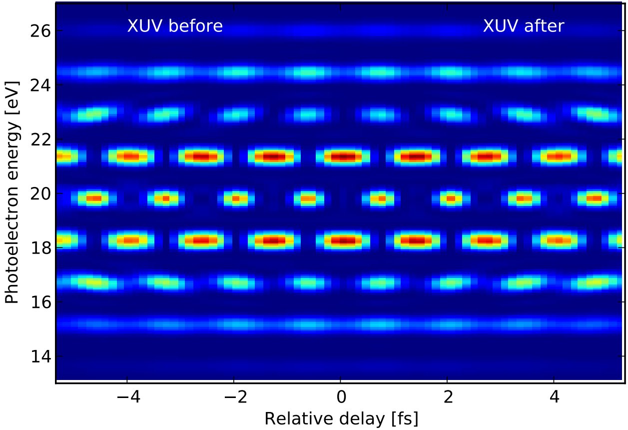

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Group-delay characterization of high-order harmonics

RABBIT method based on higher-order laser photon processes

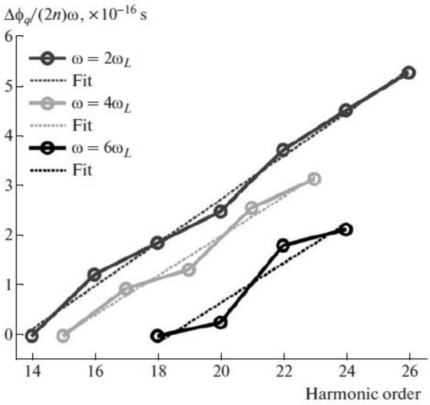


Fig. 8. Comparaison of the obtained phase differences for three different frequency components present in the experimental electron signal. The conventional RABITT includes contribution from sidebands 14 to 26. The $4\omega_R$-component has been extracted from harmonics 15 to 23 and the $6\omega_R$-modulation was obtained from sidebands 18 to 24. The curves have been shifted for better comparison.

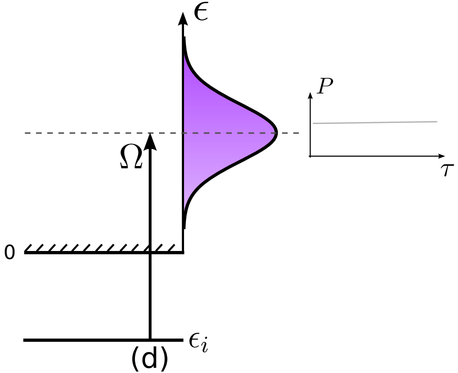
[Swoboda et al. Laser Physics **19** 1591 (2009)]

– What if a single attopulse is used? (instead of an attosecond pulse train)

Group-delay characterization of coherent XUV continuum

FROG-CRAB method (=...Complete Reconstruction of Attosecond Burst)

No temporal information by one-photon ionization

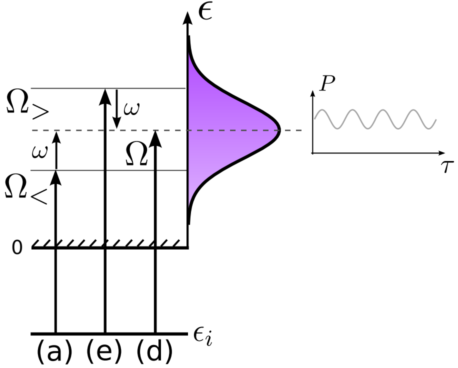


Broad photoelectron peak due to absorption of one XUV harmonic photon Ω

Group-delay characterization of XUV continuum

FROG-CRAB method (=...Complete Reconstruction of Attosecond Burst)

Laser field will induce complex inteference

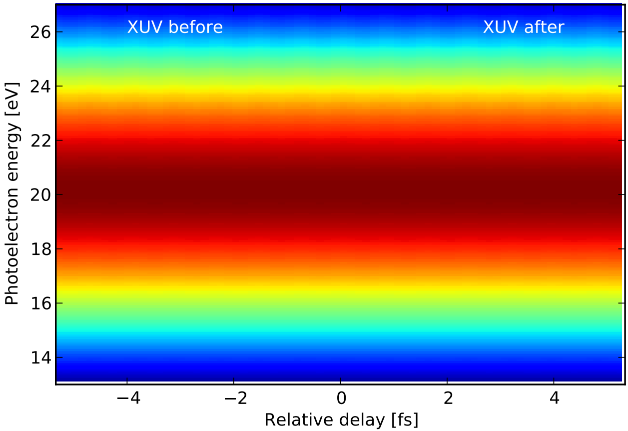


Classical picture

"Streaking of photoelectron" $p_f \approx p_0 - A(t_0)$

Photoelectron spectrogram

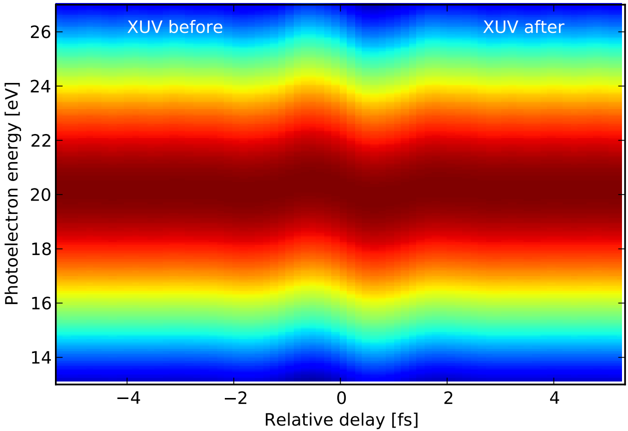
One photon absorption from XUV continuum and dressing by laser field (Volkov)



Redistribution of photoelectron probability due to vector potential of laser field

Photoelectron spectrogram

One photon absorption from XUV continuum and dressing by laser field (Volkov)



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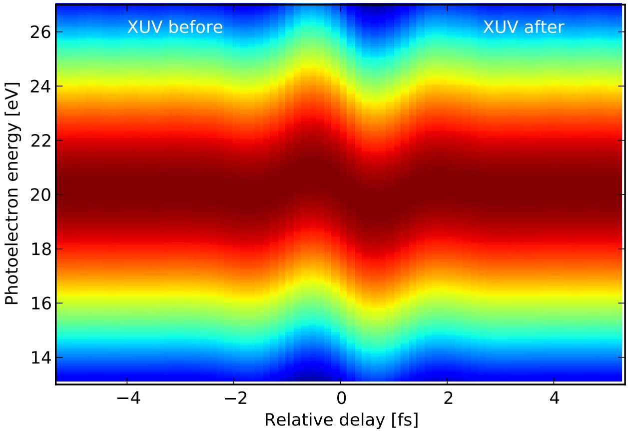

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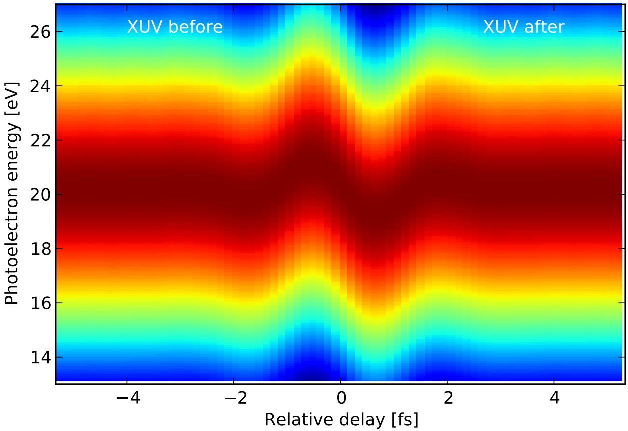

Redistribution of photoelectron probability due to vector potential of laser field

Connection to the streaking picture

Amplitude for laser-dressed one-photon ionization:

$$c_{\mathbf{k}}(t) = \frac{1}{i}\langle\varphi_{\mathbf{k}}|\hat{p}_z|g\rangle \int_{-\infty}^{t} dt' A_X(t') \exp\left[i \int^{t'} dt'' \frac{[\mathbf{k} + \mathbf{A}_L(t'')]^2}{2} + I_p \right]$$

Assume *short XUV pulse* given by $A_X(t) = \Lambda_X(t - t_0)\sin\omega_X t$,
then the laser vector potential *appears* static: $t'' \approx t' \approx t_0$.

Connection to the streaking picture

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$$c_{\mathbf{k}}(t) \approx \frac{1}{i}\langle\, \varphi_{\mathbf{k}} \mid \hat{p}_z \mid g \,\rangle \int_{-\infty}^{t} dt' \frac{1}{2} \Lambda_X(t' - t_0)$$
$$\exp\{ i[\epsilon_k + I_p - \omega_X + \mathbf{k} \cdot \mathbf{A}_L(t_0)]t' \} \quad (\mathit{Task} : 5)$$

- Quasi-static vector potential approximation: $A(t'') \approx A(t_0)$.
- Energy conservation determined by exponential factor. The shift is given by instantaneous laser vector potential!

Connection between multi-photon and streaking pictures

Identification of streaking mechanism as multi-photon processes:

$$\exp[i\mathbf{k}\cdot\mathbf{A}_L(t_0)t'] \leftrightarrow \sum_{n=-\infty}^{\infty}(-i)^n J_n\left(\frac{\mathbf{k}\cdot\mathbf{\Lambda}_L(t')}{\omega_L}\right)\exp[in\omega_L t']$$

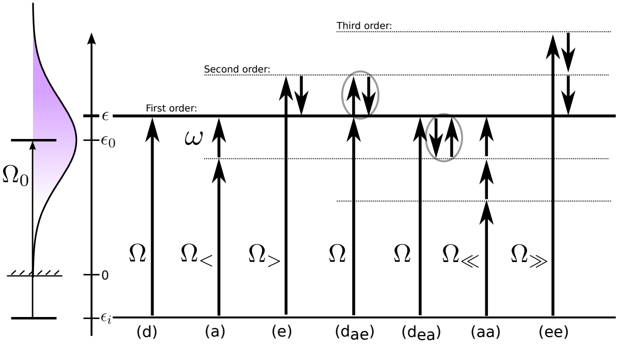
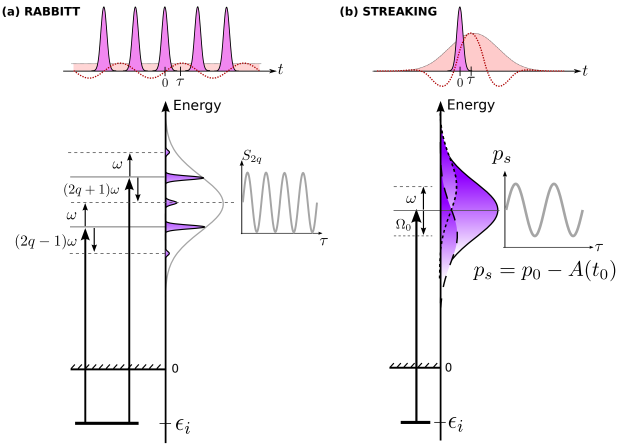


Figure: Multi-photon processes leading to the same final state.

Probing temporal structure of *as* pulse

Photoelectron is manipulated using an phase-locked laser field



- Spectral shearing interferometry (abs./emi. of laser photon)
- Frequency Resolved Optical Gating (laser sets "phase gate")

[Paul et al. Science **292**, 1689 (2001)]
[Mairesse and Quéré. PRA, **71** 011401, (2005)]

Can we measure a delay in photoionization?

– *Is it a delay of the attopulse or of the photoelectron!?*

Experimental breakthrough = Relative delay measurements

- Inter-orbital delay experiments ("between states")
- Inter-species delay experiments ("between atoms")
  using the *same* attosecond pulses.

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  using the *same* attosecond pulses.

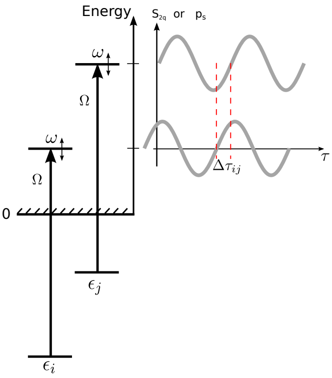
Theoretical proposal for latency-free pulse characterization

- Photoionization of coherent bound wave packets (PANDA)
- Pabst and Dahlström PRA **94**, 013411 (2016)
- PhD tutorial: arXiv:1808.08102 [quant-ph] (2018)

Inter-orbital photoionization delay experiment

Differential delay between initial orbitals *i* and *j*

Idea: Use the same attopulse to ionize from different orbitals!

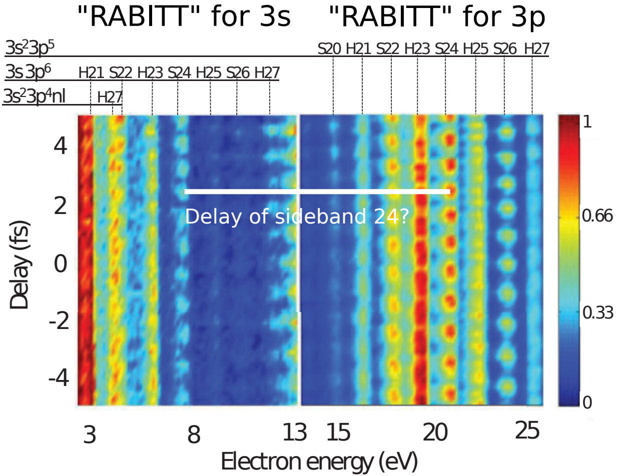


Ne: $2p - 2s$ [*Schultze et al.* Science **328** (2010) 1658]

Ar: $3p - 3s$ [*Klünder et al.* PRL **106** (2011) 143002]

Inter-orbital photoionization delay experiment

(in attoseconds, $1\,as = 10^{-18}\,s$)



Same sideband order but different ionic states $\sim 100\,as$ (@37 eV).

[Klünder et al. PRL 106 143002 (2011)] [Guenot et al. PRA **85**,053424 (2012)]

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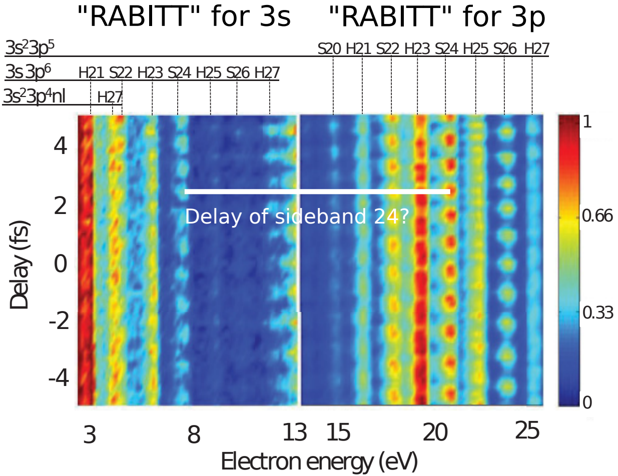

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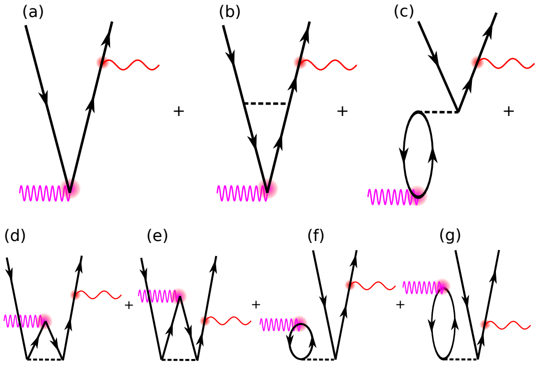
Experiment: -"Electrons from 3*p* are delayed relative 3*s*."

[Klünder et al. PRL 106 143002 (2011)] [Guenot et al. PRA **85**,053424 (2012)]

How to treat continuum transition in theory?

$$\lim_{\eta \to 0} \frac{1}{\Delta E + i\eta} = \mathrm{p.v.} \frac{1}{\Delta E} - i\pi\delta(\Delta E)$$

Calculation of correlated two-photon matrix elements:
(RPAE=Random Phase Approximation with Exchange)



- "Feynman diagrams": ↑=electron and ↓=hole
- Absorption of XUV photon with RPAE correlation effects.
- Stimulated electron continuum transition by IR field.

[Dahlström *et al.* Phys. Rev. A 86, 061402 (2012)] [J M Dahlström and E Lindroth JPB **47** 124012 (2014)]

Evaluation of IR-driven continuum transition

The perturbed wavefunction (PWF) is an outgoing wave

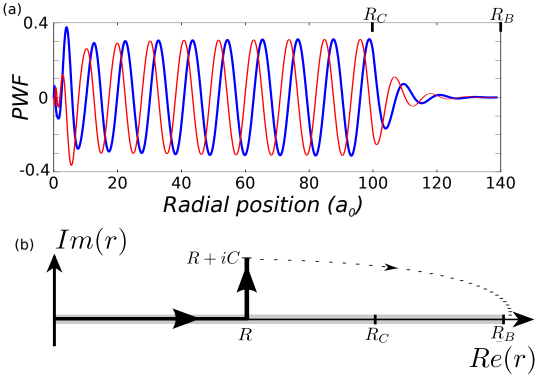
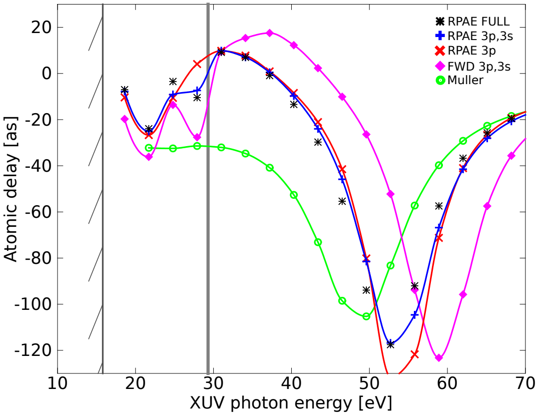


Figure: A perturbed wavefunction (PWF) is setup on **B-splines** (kord=7) with **exterior complex scaled** knot sequence (nknot=250). The PWF is **matched to Coulomb functions** before the scaled region ($x < 100$). The remaining analytical integral is evaluated along the imaginary axis.

Study of correlation effects in Ar$3p^{-1}$

Experimental binding energies (not HF values):



- At 34.1 eV (SB22) the atomic delay is small (~ 5 as).
- The atomic delay exhibits a negative peak of ~ -120 as.

[J M Dahlström and E Lindroth JPB **47** 124012 (2014)]

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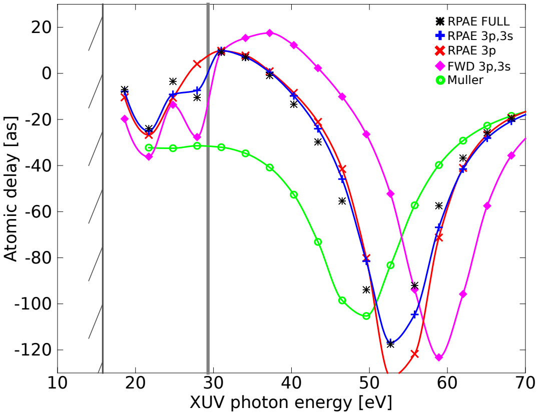
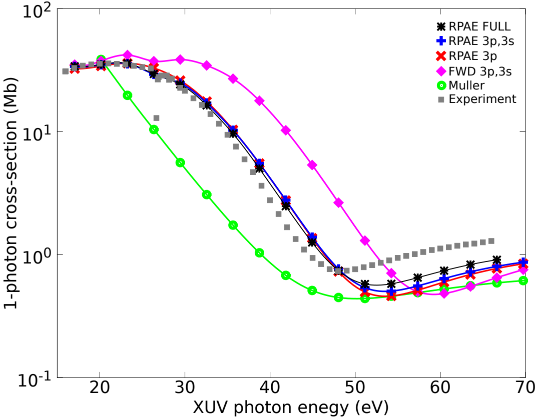


- At 34.1 eV (SB22) the atomic delay is small (~ 5 as).
- The atomic delay exhibits a negative peak of ~ -120 as.
- Electron correlation effects amount to ~ 40 as (Muller).

[J M Dahlström and E Lindroth JPB **47** 124012 (2014)]

One-photon ionization cross-section for argon $[3p^{-1}]$



- Cooper minimum because dipole matrix element vanishes.*
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*[J W Cooper Phys. Rev. **128** 681 (1962)] Fig: [J M Dahlström and E Lindroth JPB **47** 124012 (2014)]

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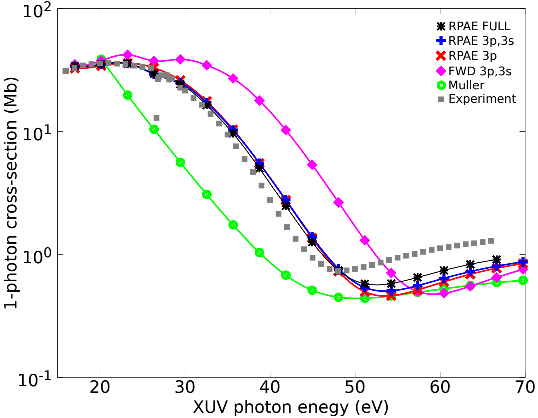

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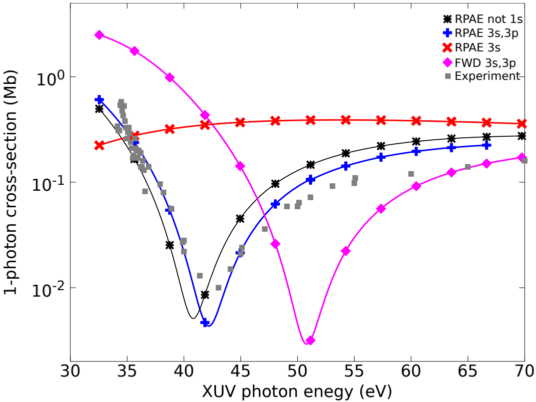
One-photon ionization cross-section for argon $[3s^{-1}]$



- Cooper minimum in 3s due to correlation with 3p.**
- Intra-orbital correlation between 3s and 3p is required.

[M Ya Amusia et al PHYS. LETT. **40A 361 (1971)] [J M Dahlström and E Lindroth JPB **47** 124012 (2014)]

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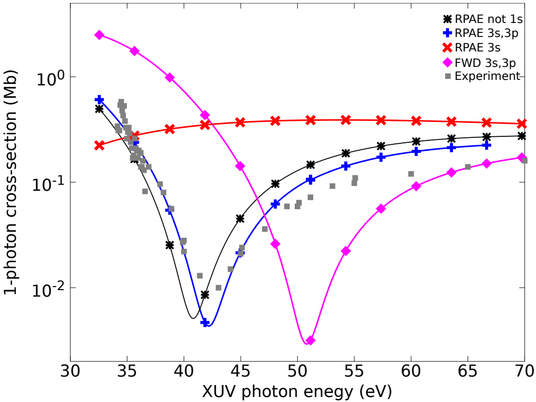

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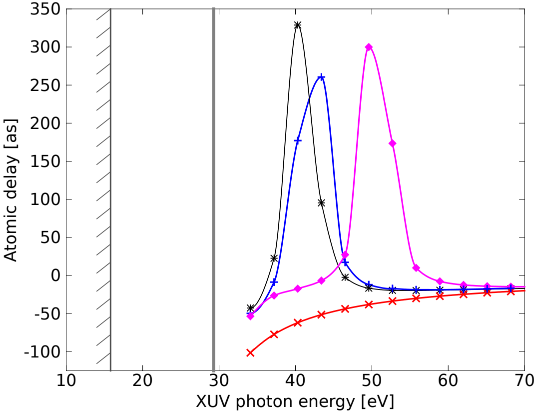
Study of correlation effects in Ar3s^{-1}

Experimental binding energies (not HF values):



- Large *positive* delay peak (~ 300 as) close to 40 eV*.
- Electron correlation effects amount to ~ 400 as.
- At 34.1 eV (SB22) the delay is ~ -50 as.

[J M Dahlström and E Lindroth JPB **47** 124012 (2014)] *[A S Kheifets PRA **87**, 063404 (2013)]

Comparison between theory and the argon experiment

Table of results for argon delays:

Experiment*	$\tau_{3s} - \tau_{3p} = -80 \pm 50\,\mathrm{as}$	(SB22)
Theory:	$\tau_{3s} - \tau_{3p} \approx -55\,\mathrm{as}$	

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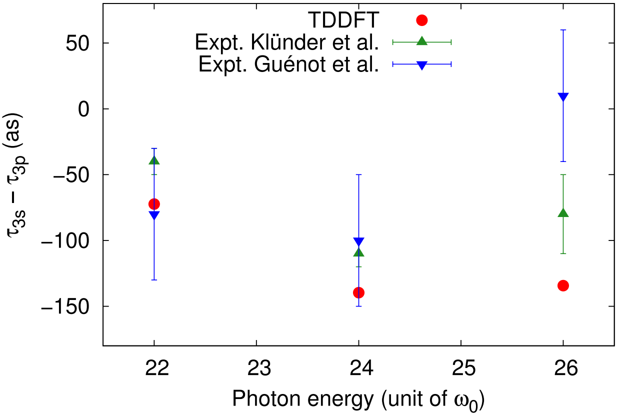
* Guénot et al. PHYSICAL REVIEW A **85**, 053424 (2012)

Other ideas?

- The $3s^{-1}$ is only 69% a single hole state.**
- Shake-up processes: $3s^{-1} \to 3p^{-2}n\ell$.
- Laser-stimulated hole transitions.
- Final state correlation (after absorption of IR).

** T Carette et al . PRA **87**, 023420 (2013)

Comparison between experiment and TDDFT



TDDFT: Sato et al. Eur. Phys. J. B **91**: 126 (2018)
(Group of Angel Rubio at CFEL)

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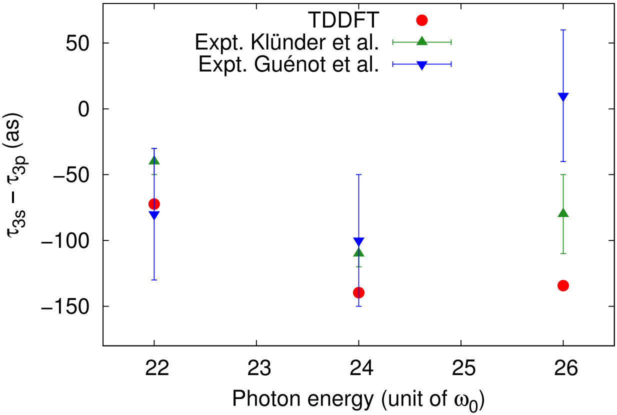

TDDFT: Sato et al. Eur. Phys. J. B **91**: 126 (2018)

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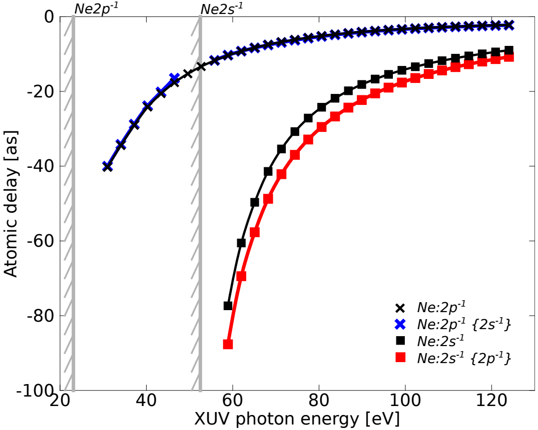
Position of 3s Cooper minimum using TDDFT?

– Time to revisit the simpler atom: NEON

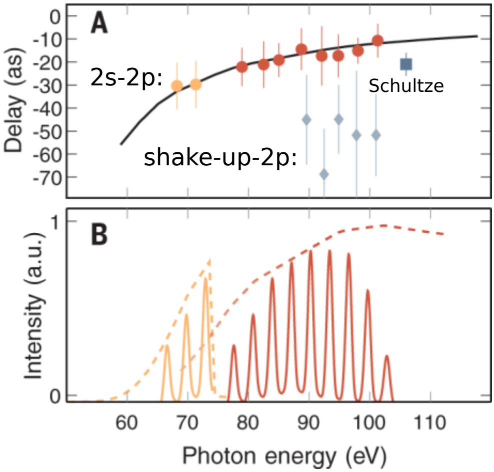
"Atomic delays" from 2p and 2s states in Ne



- **Small delay** in 2s due to **inter-orbital** correlation with 2p.
- Delay at $\sim 105\,\text{eV}$: $\Delta\tau_{p-s} =$12.4 as (Exp* ≈ 21 as)

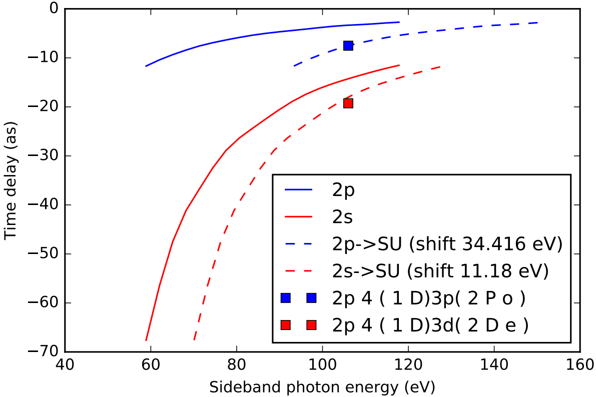
[Dahlström *et al.* Phys. Rev. A 86, 061402 (2012)], *[Schultze et al. Science 328, 1658 (2010)]

Neon delay 2s-2p revisited



M. Isinger et al., Science 10.1126/science.aao7043 (2017).

Delay in shake-up channels?



Simple model for shake-up based on RPAE
agrees with hybrid MCHF+CLC+DLC calculation
Feist et al., Phys. Rev. A 89, 033417 (2014),

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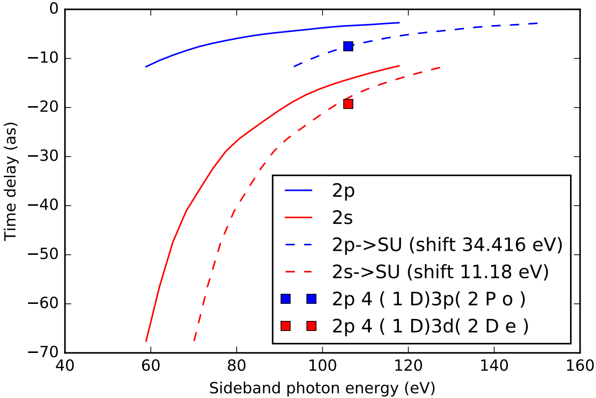

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Too small compared to experiment = OPEN QUESTION

– OK, “atomic delays” have been measured experimentally. Why is it so fascinating — what does it mean?

Probing Single-Photon Ionization on the Attosecond Time Scale

$$\tau_A = \tau_W + \tau_{CC}$$

``The determination of photoemission time delays requires taking into account the measurement process, involving the interaction with a probing infrared field. This contribution can be estimated using a universal formula and is found to account for a substantial fraction of the measured delay.''

[K. Klünder et al. PRL **106**, 143002 (5 April 2011)]

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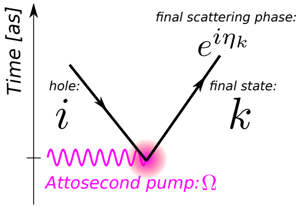
Time-resolved photoemission by attosecond streaking: extraction of time information

``We show that attosecond streaking ... contain ... Eisenbud-Wigner-Smith time delay matrix ... if ... the streaking infrared (IR) field ... is properly accounted for ...'' [S Nagele et al. JPB. **44**, 081001 (11 April 2011)]

Photoionization matrix elements



One-photon matrix element:

$$M^1(\vec{k}) = -\, iE_\Omega \langle\, \vec{k} \mid z \mid i \,\rangle$$
$$\sim \exp[i\eta_\ell(k)]$$

[J.M. Dahlström et al Chem.Phys.(2012)]

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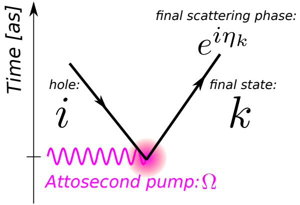

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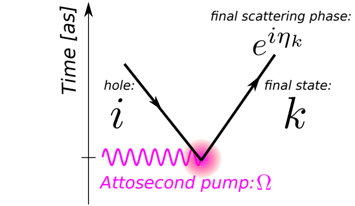
Scattering state expansion in partial wave basis:

$$\phi_{\vec{k}}^{(-)}(\vec{r}) = \sum_{\ell,m} i^\ell e^{-i\eta_\ell} Y^*_{\ell,m}(\hat{k}) Y_{\ell,m}(\hat{r}) R_{k,\ell}(r)$$

Scattering phase, η_ℓ, is specific to the target atom.

[J.M. Dahlström et al Chem.Phys.(2012)]

Photoionization matrix elements



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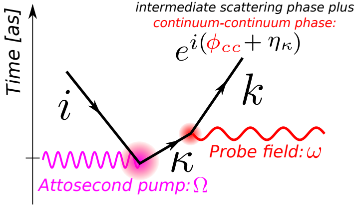


Two-photon matrix element:

$$M^2(\vec{k}) = -\, iE_\Omega E_\omega$$
$$\times \sum\!\!\!\!\!\!\int_{\kappa'} \frac{\langle\, \vec{k} \mid z \mid \kappa' \,\rangle\langle\, \kappa' \mid z \mid i \,\rangle}{\epsilon_i + \Omega - \epsilon_{\kappa'}}$$
$$\sim \exp[i\phi_{cc}(k,\kappa) + i\eta_\ell(\kappa)]$$

[J.M. Dahlström et al Chem.Phys.(2012)]

Continuum–continuum phases

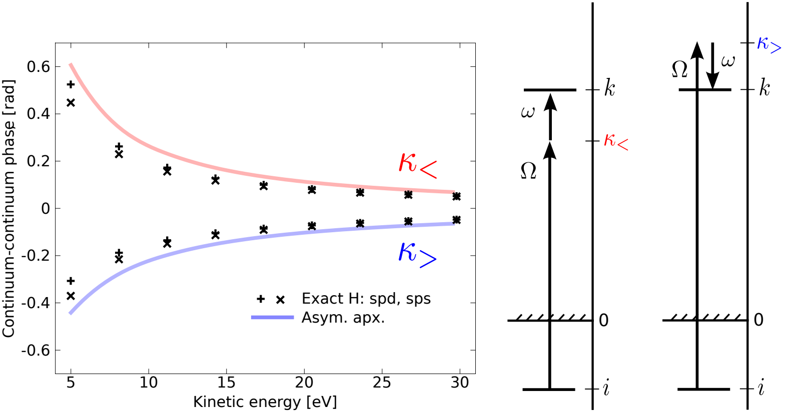
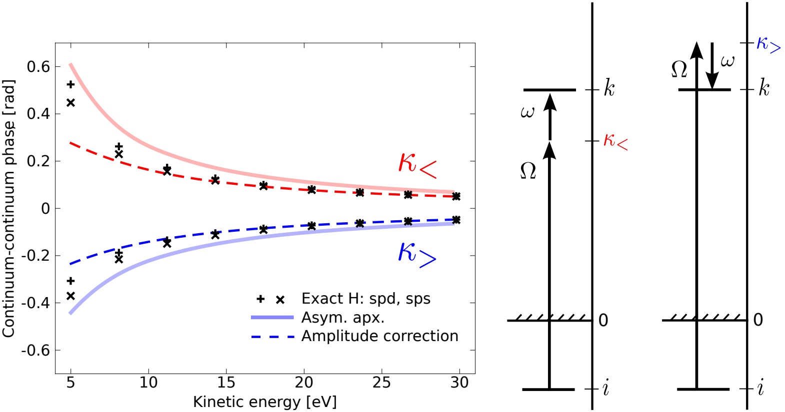


Figure: Exact vs. asymptotic values of $\phi_{cc}(k, \kappa)$.

[K. Klünder *et al.* PRL. (2011)]
Collaboration with A. Maquet and R. Taïeb at UPMC through COST.



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Phase of 2 photon ATI amplitude
(ATI=Above Threshold Ionization process)

Explicit phase of ATI transition: $i \rightarrow \vec{\kappa} \rightarrow \vec{k}$**:**

$$\arg[M^2(\vec{k})] \approx \pi + \arg[Y_{L,m_i}(\hat{k})] + \phi_\Omega + \phi_\omega - \frac{\pi\ell}{2} + \eta_\ell(\kappa) + \phi_{cc}(k,\kappa),$$

with XUV: Ω first, then continuum–continuum IR: ω.

(One intermediate angular momenta: ℓ.)

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-Now we apply this "ansatz" to experimental schemes!

Applications of ATI matrix elements: "RABBIT"

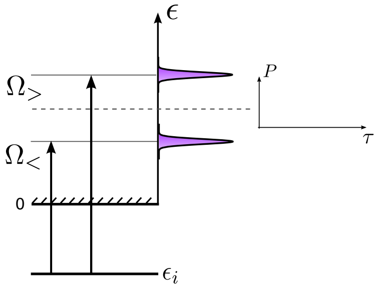

Figure: Ionization by APT.

Applications of ATI matrix elements: "RABBIT"

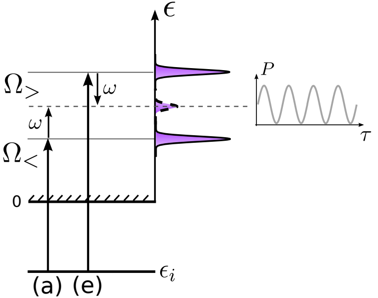

Figure: Ionization by APT+IR.

Probability of emission along $\hat{z}$:

$$P(\vec{k}) \approx |M_a + M_e|^2$$
$$= |M_e|^2 + |M_a|^2 + 2\Re\{M_e M_a^*\}$$

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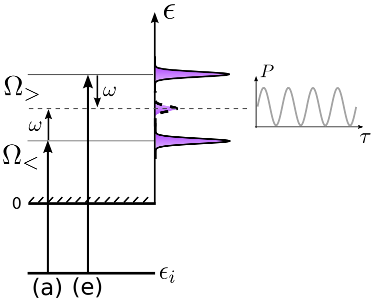

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The phase of the cross-term:

$$\arg\{M_e M_a^*\} \approx -2\omega \times \tau$$

$$+\phi_{\Omega_>} + \eta_{\kappa_>,\ell} + \phi_{cc}(k, \kappa_>)$$

$$-\phi_{\Omega_<} - \eta_{\kappa_<,\ell} - \phi_{cc}(k, \kappa_<)$$

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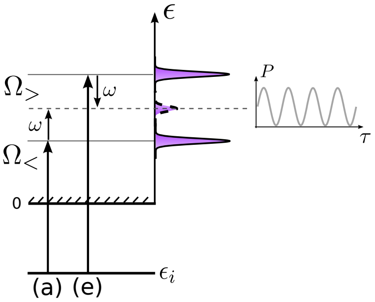

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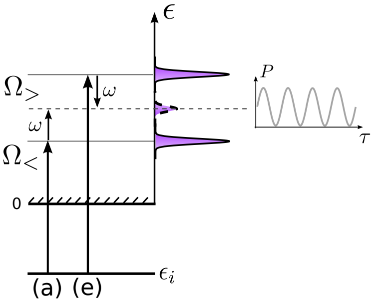

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Max of modulation!

$$\tau = \frac{\Delta\phi_\Omega}{\Delta\omega} + \frac{\Delta\eta_{\kappa,\ell}}{\Delta\omega} + \frac{\Delta\phi_{cc}}{\Delta\omega}$$

(Finite-difference derivatives)

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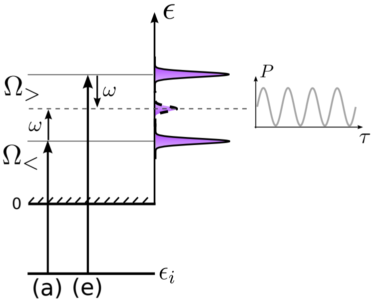

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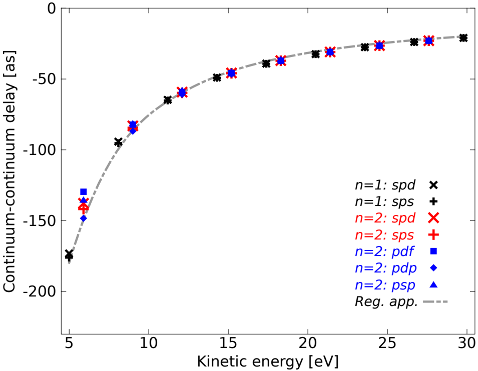
"RABBIT delays" (relation: max vector potential of probe field vs. arrival of XUV pulse)

$$\tau \approx \tau_\Omega + \tau_{k,\ell} + \tau_{cc}(k;\omega),$$

Group delay + Wigner delay + Continuum–continuum delay.

Continuum–continuum delays in hydrogen (H)



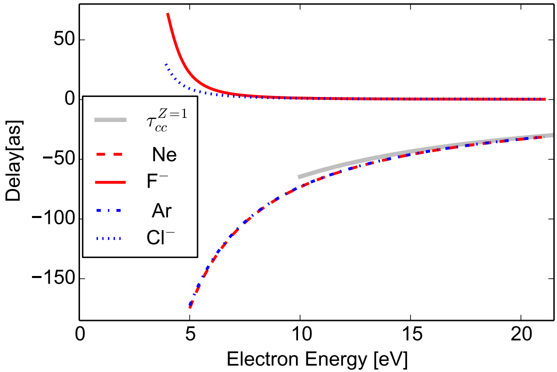
$$\tau_{cc}(k,\omega) \equiv \frac{\Delta\phi_{cc}}{\Delta\omega} \equiv \frac{\phi_{cc}(k,\kappa_>) - \phi_{cc}(k,\kappa_<)}{2\omega}.$$

Exact calculations by R. Taïeb (UPMC) for hydrogen using Sturmians.
[J. M. Dahlström and D. Guénot *et al.* Chem. Phys. (2012)]

What have we learned about "atomic delay" since 2001?

Interpretation:

"Atomic delay" $\approx$ Wigner delay + CC delay:



- Target-specific **Wigner delay** of photoelectron.
- **Noble gas universal CC delay** due to **laser transition**.

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

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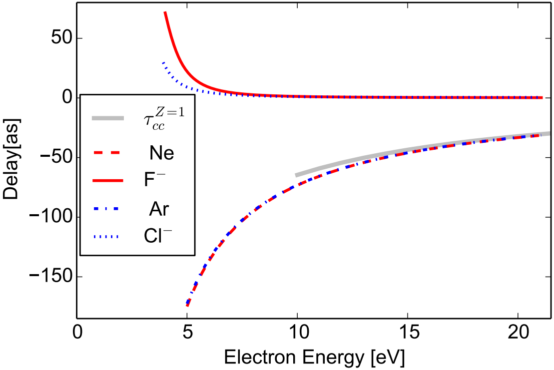
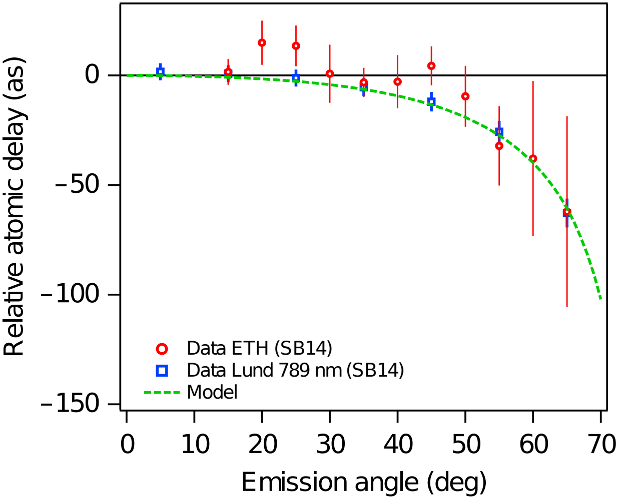


- Target-specific **Wigner delay** of photoelectron.
- In **negative ions** the CC delay is **small but not universal**!

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

The atomic delays vary over angle of emission

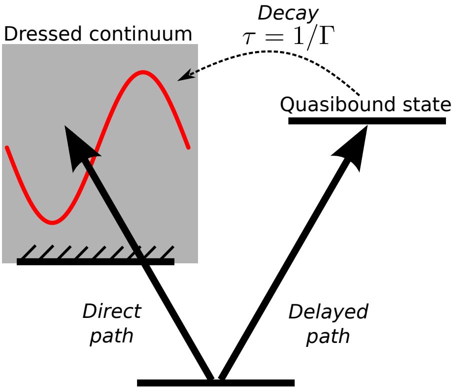


Cirelli et al. NATURE COMMUNICATIONS (2018) 9:955

What happens if a resonance is embedded in the continuum?

Streaking with a resonance

Direct and autoionizing processes



Asymmetric Fano peak

Photoelectron distribution depends on q-parameter

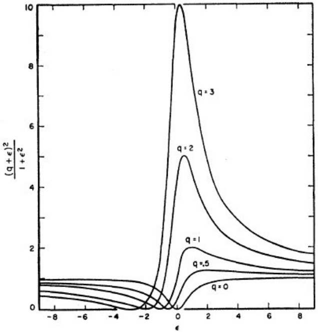


FIG. 1. Natural line shapes for different values of q. (Reverse the scale of abscissas for negative q.)

The parameter q measures the relative strength of the formation of the “bound” state and the direct continuum.

[U Fano Phys. Rev. **124** 1866 (1961)]

Streaking over a resonance

Direct and autoionizing processes

Fano theory transition probability ratio:

$$\frac{|\langle \Psi \mid T \mid g \rangle|^2}{|\langle \psi \mid T \mid g \rangle|^2} = \frac{(q+\epsilon)^2}{1+\epsilon^2}$$

where the $\epsilon = (E - E_r)/(\Gamma/2)$ and q describes the resonance.

Corresponding complex amplitude:

$$\langle \Psi \mid T \mid g \rangle = \underbrace{\frac{q+\epsilon}{1-i\epsilon}}_{f_F(E)} \langle \psi \mid T \mid g \rangle$$

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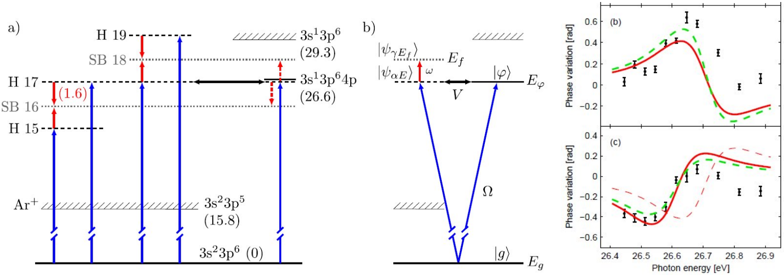
Can this phase shift be measured?

RABBIT with a resonance

Direct and autoionizing processes



Two-photon matrix element with two continuum and one resonance:

$$M = M^{(1)}\frac{q+\epsilon}{\epsilon+i} + M^{(2)}$$

[Kotur et al. NATURE COMMUNICATIONS — 7:10566 (2015)]

Fano theory in time domain

Corresponding complex amplitude:

$$\langle \Psi \mid T \mid g \rangle = \underbrace{\frac{q+\epsilon}{1-i\epsilon}}_{f_F(E)} \langle \psi \mid T \mid g \rangle$$

Find time domain representation: (*Task* : 6)

$$F_F(\tau) = \frac{1}{2\pi} \int dE \, f_F(E) \exp[-iE\tau] = i\delta(\tau) + \frac{\Gamma}{2}(q-i)e^{-iE_r\tau - \Gamma\tau/2}\Theta(\tau)$$

See: [Z X Zhao and C D Lin PRA **71**, 060702 (2005)]

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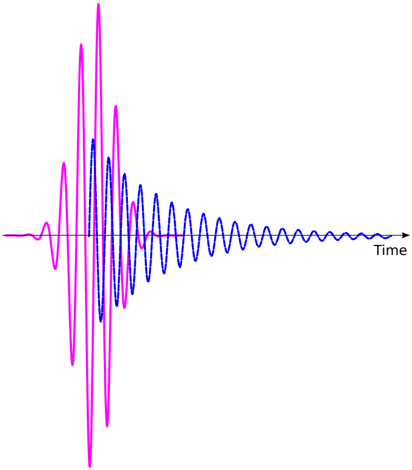
The Fano phase is proportional to ϵ for $q = 0$ (*Task* : 7)
which implies that the τ_W at the resonance is $2/\Gamma$.

See: [Z X Zhao and C D Lin PRA **71**, 060702 (2005)]

Streaking with a resonance

Direct and autoionizing processes

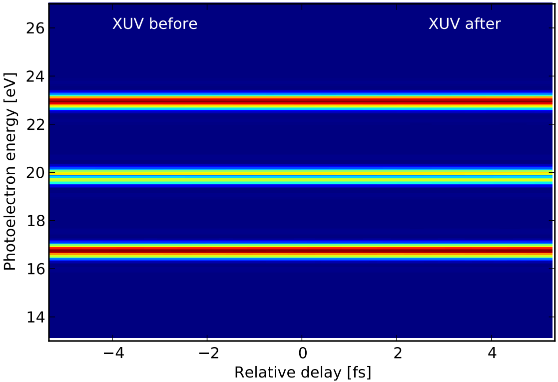


Electrons ejected into the continuum:

Direct path + Decay (exponential tail)

Photoelectron spectrogram

One photon absorption to dressed continuum with autoionization



Redistribution of three harmonic peaks due to laser dressing of window resonance ($q = 0$).

See also: RAINBOW RABBITT technique: Gruson et al. Science (2016)
Time scales of autoionization longer than laser period!

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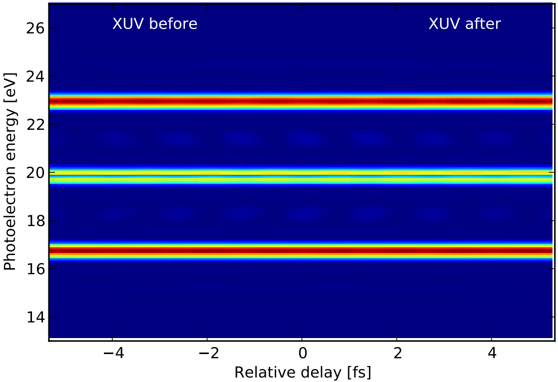

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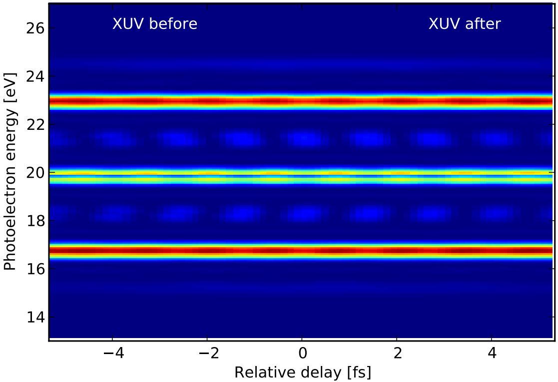

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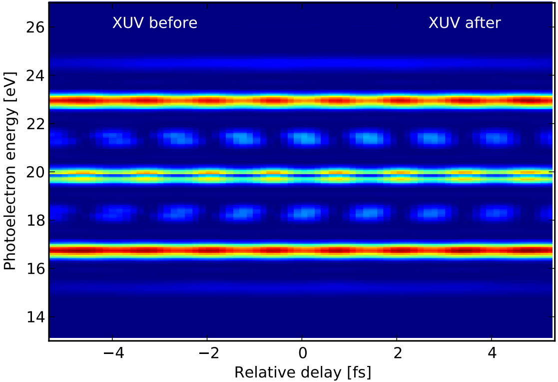

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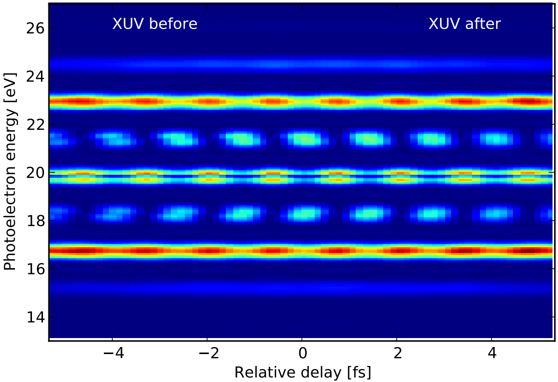

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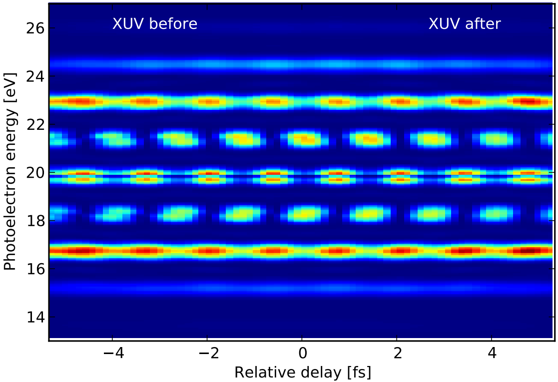

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Conclusion and Outlook:

- **Attosecond pulse metrology** has shifted focus to make connection with the field of **theoretical atomic physics**.
- The simple approximations based on SFA are not sufficient to describe attosecond photoelectron dynamics.
- The **Wigner delay can not be directly measured**, but it can be extracted based on assumptions regarding the interaction with the probe field.
- Inter-orbital delays can be used to test electron correlation effects.
- **Non-linear interaction with the fields** *and* **ion.**

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http://www.matfys.lth.se/staff/Marcus.Dahlstrom/
Thank you for your attention!