Attosecond time-delays in photoionisation

J. Marcus Dahlström

2018-08-31 ELISS Szeged, Hungary.
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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- **Review of attosecond pulse characterization** [*Basic level*]
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- **How large is the atomic response?** [*Intermediate level*]
  - Argon photoionization delay experiment
  - Delays in other noble gas atoms

- **How can we interpret the atomic delays?** [*State of the art*]
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  - Many electron effects (“Feynman diagrams”)
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- **Problems for the PhD-studends (Task : i)**
  
  [link](http://www.matfys.lth.se/staff/Marcus.Dahlstrom/)

* SFA=Strong Field Approximation
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. Agostini1

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”?
– Why is a laser field needed to characterize attopulses?
Group-delay characterization of high-order harmonics
RABBITT method

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

\[ P \approx A + B \cos[2\omega(\tau - \tau_{GD} - \tau_{Atom})], \]
where \( \tau_{GD} \approx (\phi_> - \phi_<)/2\omega \) is group delay of attopulse.
– How can the atomic delay, $\tau_{\text{Atom}}$, be determined? Is it important or negligible?
Model: atom in multi-color electromagnetic fields

Atomic units: $e = m = \hbar = 4\pi\varepsilon_0 = 1$

Hamiltonian for interaction with field and ion:

$$H = H_V + V_A$$

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

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

Vector potential of both attopulses and laser fields:

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

Atomic potential for hydrogen:

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

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

Vector potential of both attopulses and laser fields:

\[
A(t) = A_X(t) + A_L(t)
\]

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>
<thead>
<tr>
<th>Sideband</th>
<th>$\Delta \varphi_{\text{atomic}}^f$ (rad) / amplitude $A_f$ (arbitrary units)</th>
</tr>
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<tbody>
<tr>
<td>$E_0 + 12\hbar\omega$</td>
<td>$(1,0) \rightarrow (1,0)$: 0.438/6094, $(1,0) \rightarrow (3,0)$: 0.060/3659, $(1, \pm 1) \rightarrow (1, \pm 1)$: 0.125/1914, $(1, \pm 1) \rightarrow (3, \pm 1)$: 0.060/2440</td>
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<td>$E_0 + 14\hbar\omega$</td>
<td>$(1,0) \rightarrow (1,0)$: 0.292/5135, $(1,0) \rightarrow (3,0)$: 0.102/2311, $(1, \pm 1) \rightarrow (1, \pm 1)$: 0.125/1281, $(1, \pm 1) \rightarrow (3, \pm 1)$: 0.102/1541</td>
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<tr>
<td>$E_0 + 16\hbar\omega$</td>
<td>$(1,0) \rightarrow (1,0)$: 0.221/3645, $(1,0) \rightarrow (3,0)$: 0.100/1349, $(1, \pm 1) \rightarrow (1, \pm 1)$: 0.108/763, $(1, \pm 1) \rightarrow (3, \pm 1)$: 0.100/899</td>
</tr>
<tr>
<td>$E_0 + 18\hbar\omega$</td>
<td>$(1,0) \rightarrow (1,0)$: 0.192/2444, $(1,0) \rightarrow (3,0)$: 0.090/742, $(1, \pm 1) \rightarrow (1, \pm 1)$: 0.090/427, $(1, \pm 1) \rightarrow (3, \pm 1)$: 0.090/494</td>
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*If we know the amplitudes and phases then we can compute* $\tau_{\text{Atom}}$ *and deduce the group delay of the attopulses* $\tau_{\text{GD}}$ *in experiments.*

[Paul et al. SCIENCE 1690 292 (2001)]
Quite complex process...

Amplitude and phase of two-photon matrix elements

Table 1. The atomic phases $\Delta \varphi_{\text{atomic}}$ and the relative strengths $A_{\gamma}$ 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.

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

But how sure are we about this model? Can it be tested?

[Paul et al. SCIENCE 1690 292 (2001)]
Simplest possible model for RABBITT

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

Assumption: Photoelectron is unaffected by atomic potential.

Plane wave:
\[
\varphi_k(r) = \frac{1}{(2\pi)^{3/2}} \exp[i k \cdot r]
\]
Assumption: Photoelectron is unaffected by atomic potential.

Plane wave:

\[ \varphi_k(r) = \frac{1}{(2\pi)^{3/2}} \exp[ik \cdot r] \]

Momentum eigenstate:

\[ \hat{p}\varphi_k \equiv -i\nabla \varphi_k(r) = k\varphi_k \]

Solution to the free particle Schrödinger equation (SE):

\[ H_0 \varphi_k = \frac{\hat{p}^2}{2} \varphi_k = \frac{k^2}{2} \varphi_k \equiv \epsilon_k \varphi_k \]
Simplest possible model for RABBITT

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

Second-order perturbation theory*:

\[
M_k^{(2)} \approx \int d^3k' \frac{\langle k | O | k' \rangle \langle k' | O | g \rangle}{(\epsilon_g + \omega - \epsilon_{k'})}
\]

Perturbation by external field (dipole approximation):

Velocity: \( O = A(\omega) \cdot \hat{p} \)

Length: \( O = E(\omega) \cdot r \)

Vector potential and electic field (uniform in space):

\[
\tilde{E}(t) = -\frac{\partial \tilde{A}}{\partial t}
\]

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

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

(Task: 1) Approximate two photon matrix element:

$$M_k^{(2)} \approx -2A(\Omega)A(\omega)\frac{\epsilon_k}{\omega} \cos^2 \theta_k \langle k | g \rangle$$
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\[ M_k^{(2)} \approx -2A(\Omega)A(\omega) \frac{\epsilon_k}{\omega} \cos^2 \theta_k \langle k \mid g \rangle \]

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

\[ \langle k \mid g \rangle = \frac{2^{3/4}}{\pi} \frac{l_p^{5/4}}{(l_p + \epsilon_k)^2}, \quad l_p = \frac{Z^2}{2} \]

The two-photon matrix goes like \( 1/\epsilon_k, \epsilon_k \gg l_p \) and it is \textit{real} within plane-wave approximation.
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\langle 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.

\[\rightarrow\] 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):

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

Volkov Hamiltonian (velocity gauge):

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

Ansatz using plane wave with time-dependent phase:

$$\psi^V_k(r, t) = \phi_k(r) \exp[-i\Phi_k(t)]$$

Task 3: Insert into TDSE to obtain the Volkov phase:

$$\Phi_k(t) = \int_{t_{ref}}^t dt' \left[ k + A(t') \right]^2$$
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(Task : 3) Insert into TDSE to obtain the Volkov phase:

$$\Phi_k(t) = \int_{\text{ref.}}^t dt' \frac{1}{2} [\mathbf{k} + \mathbf{A}(t')]^2$$
Photoionization to laser dressed continuum

Laser-dressed time-dependent perturbation theory *

\[ c_k(t) = \frac{1}{i} \int_{-\infty}^{t} dt' A_X(t') \langle \psi^V_k | \hat{p}_z | \tilde{g} \rangle \]

where the conjugate Volkov state is

\[ \psi^V_k(r, t) = \phi_k^*(r) \exp[i\Phi_k(t)] \]

and the ground state is with binding \( l_p > 0 \) is

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

* [M Kitzler, N Milosevic, A Scrinzi, F Krausz, and T Brabec PRL 88, 173904 (2002)]
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Amplitude for final momentum \( k \):

\[
c_k(t) = \frac{1}{i} \langle \phi_k | \hat{p}_z | g \rangle \int_{-\infty}^{t} dt' A_X(t') e^{i \int^{t'} dt'' \left\{ \frac{[k + A_L(t'')]^2}{2} + I_p \right\}}
\]

* [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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Attosecond time-delays in photoionisation
Redistribution of three harmonic peaks due laser dressing: 
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One photon absorption from XUV comb and dressing by laser field (Volkov approx.)

Redistribution of three harmonic peaks due laser dressing:
Formation of sidebands.
– How does the photon picture arise?
Amplitude for laser-dressed one-photon ionization:

\[ c_k(t) = \frac{1}{i} \langle \varphi_k | \hat{p}_z | g \rangle \int_{-\infty}^{t} dt' A_X(t') \exp \left( i \int_{t'}^{t} dt'' \left\{ \frac{[k + A_L(t'')]^2}{2} + l_p \right\} \right) \]

Assume weak laser \([k + A_L(t'')]^2 \approx k^2 + 2k \cdot A_L(t'')\) and slowly varying laser envelope \(\Lambda_L(t)\) compared to laser oscillation \(\omega_L\) with \(A_L(t) = \Lambda_L(t) \sin \omega_L t\)
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\[ c_k(t) \approx \frac{1}{i} \langle \varphi_k | \hat{p}_z | g \rangle \int_{-\infty}^{t} dt' \frac{1}{2} \Lambda_X(t') \sum_{n=-\infty}^{\infty} (-i)^n J_n \left( \frac{k \cdot \Lambda_L(t')}{\omega_L} \right) \]

\[ \times \exp \left[ i(\epsilon_k + l_p - \omega_X + n\omega_L) t' \right] \quad (Task : 4) \]

- Photon energy conservation given by exponential factor.
- Multiphoton transition determined by real Bessel function, \(J_n\).
Amplitude for laser-dressed one-photon ionization:

\[ c_k(t) = \frac{1}{i} \langle \varphi_k | \hat{p}_z | g \rangle \int_{-\infty}^{t} dt' A_X(t') \exp i \int_{t'}^{t} dt'' \left\{ \frac{[k + A_L(t'')]^2}{2} + I_p \right\} \]

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

\[ \rightarrow \text{The atomic delay is zero?!?} \]
The Bessel functions describe the magnitude of laser stimulated continuum transitions as a function of $x = k \cdot A_L / \omega_L$. 
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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Redistribution of harmonics due to intense laser dressing. Multi-photon processes amount to non-sinusoidal beating patterns.
Group-delay characterization of high-order harmonics

RABBITT method based on higher-order laser photon processes

Fig. 8. Comparison of the obtained phase differences for three different frequency components present in the experimental electron signal. The conventional RABBITT 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)
No temporal information by one-photon ionization

Broad photoelectron peak due to absorption of one XUV harmonic photon $\Omega$
Group-delay characterization of XUV continuum
FROG-CRAB method (Complete Reconstruction of Attosecond Burst)

Laser field will induce complex interference

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

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Attosecond time-delays in photoionisation
Amplitude for laser-dressed one-photon ionization:

\[ c_k(t) = \frac{1}{i} \langle \varphi_k | \hat{p}_z | g \rangle \int_{-\infty}^{t} dt' A_X(t') \exp \left[ i \int_{t'}^{t''} dt'' \frac{[k + A_L(t'')]^2}{2} + l_p \right] \]

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: \( t'' \approx t' \approx t_0 \).
Amplitude for laser-dressed one-photon ionization:

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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: \( t'' \approx t' \approx t_0 \).

\[ c_k(t) \approx \frac{1}{i} \langle \varphi_k | \hat{p}_z | g \rangle \int_{-\infty}^{t} dt' \frac{1}{2} \Lambda_X(t' - t_0) \exp \{ i[\epsilon_k + I_p - \omega_X + k \cdot A_L(t_0)]t' \} \quad (Task : 5) \]

- 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!
Identification of streaking mechanism as multi-photon processes:

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

**Figure**: Multi-photon processes leading to the same final state.
Probing temporal structure of attosecond 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.
Experimental breakthrough = Relative delay measurements

- Inter-orbital delay experiments ("between states")
- Inter-species delay experiments ("between atoms") 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)
Inter-orbital photoionization delay experiment

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

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

\[\Delta \tau_{ij}\]

\[\begin{align*}
\epsilon_i & \quad \omega \\
\epsilon_j & \quad \Omega
\end{align*}\]

Energy

\[S_{2q} \text{ or } p_s\]

Ar: $3p - 3s$ [Klünder et al. PRL 106 (2011) 143002]
Inter-orbital photoionization delay experiment
(in attoseconds, $1 \text{ as} = 10^{-18} \text{ s}$)

"RABITT" for 3s  "RABITT" for 3p

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

[Klünder et al. PRL 106 143002 (2011)] [Guenot et al. PRA 85,053424 (2012)]
Inter-orbital photoionization delay experiment
(in attoseconds, $1 \text{ as} = 10^{-18} \text{s}$)

"RABITT" for 3s   "RABITT" for 3p

$3s^23p^5$  
$3s^3p^6$  
$3s^23p^4nl$

H21 H22 H23 H24 H25 H26 H27

Delay (fs)

Delay of sideband 24?

Electron energy (eV)

Same sideband order but different ionic states $\sim 100 \text{ as} (\sim 37 \text{ eV})$.

Experiment: -“Electrons from 3p are delayed relative 3s.”

[Klünder et al. PRL 106 143002 (2011)] [Guenot et al. PRA 85,053424 (2012)]
Calculation of correlated two-photon matrix elements:
(RPAE=Random Phase Approximation with Exchange)

“Feynman diagrams”: $\uparrow=$electron and $\downarrow=$hole
Absorption of XUV photon with RPAE correlation effects.
Stimulated electron continuum transition by IR field.

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.
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)]
Study of correlation effects in Ar3p\(^{-1}\)

Experimental binding energies (not HF values):

-120
-100
-80
-60
-40
-20
0
20
10
20
30
40
50
60
70

Atomic delay [as]
XUV photon energy [eV]

- RPAE FULL
- RPAE 3p,3s
- RPAE 3p
- FWD 3p,3s
- Muller

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)]
Cooper minimum because dipole matrix element vanishes. *

Intra-orbital correlation is enough for 3p (6 e\textsuperscript{−} in 3p orbital).

* [J W Cooper Phys. Rev. 128 681 (1962)] Fig: [J M Dahlström and E Lindroth JPB 47 124012 (2014)]
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*[J W Cooper Phys. Rev. 128 681 (1962)] Fig: [J M Dahlström and E Lindroth JPB 47 124012 (2014)]
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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**[M Ya Amusia et al PHYS. LETT. 40A 361 (1971)] [J M Dahlström and E Lindroth JPB 47 124012 (2014)]
Study of correlation effects in Ar3s$^{-1}$

Experimental binding energies (not HF values):

- Large positive delay peak ($\sim$ 300 as) close to 40 eV*.
- Electron correlation effects amount to $\sim$ 400 as.
- At 34.1 eV (SB22) the delay is $\sim$ −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:

<table>
<thead>
<tr>
<th>Experiment* ( \tau_{3s} - \tau_{3p} )</th>
<th>Theory: ( \tau_{3s} - \tau_{3p} \approx -55 \text{ as} )</th>
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* Guénot et al. PHYSICAL REVIEW A 85, 053424 (2012)
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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: $3s^{-1} \rightarrow 3p^{-2} n\ell$.
- Laser-stimulated hole transitions.
- Final state correlation (after absorption of IR).

** T Carette et al. PRA 87, 023420 (2013)
Comparison between experiment and TDDFT

(Group of Angel Rubio at CFEL)

(Group of Angel Rubio at CFEL)
Comparison between experiment and TDDFT

(Group of Angel Rubio at CFEL)

Position of 3s Cooper minimum using TDDFT?

![Graph showing comparison between experiment and TDDFT, with data points and error bars.]
– 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 \( \sim 105 \text{ eV} \): \( \Delta \tau_{p-s} = 12.4 \text{ as} \) (Exp* \( \approx 21 \text{ as} \))

Neon delay 2s-2p revisited

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), Too small compared to experiment = OPEN QUESTION
– OK, “atomic delays” have been measured experimentally. Why is it so fascinating — what does it mean?
\[ \tau_A = \tau_W + \tau_{CC} \]

‘‘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)]
Probing Single-Photon Ionization on the Attosecond Time Scale

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‘‘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)]

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:

\[ M^1(\vec{k}) = -iE_\Omega \langle \vec{k} | z | i \rangle \sim \exp[i\eta_\ell(\vec{k})] \]
One-photon matrix element:

\[ M^1(\mathbf{k}) = -iE_\Omega \langle \mathbf{k} | z | i \rangle \]
\[ \sim \exp[i\eta_\ell(k)] \]

Scattering state expansion in partial wave basis:

\[ \phi_k^{(-)}(\mathbf{r}) = \sum_{\ell, m} i^\ell e^{-i\eta_\ell} Y_{\ell, m}^*(\hat{k}) Y_{\ell, m}(\hat{r}) R_{k, \ell}(r) \]

Scattering phase, \( \eta_\ell \), is specific to the target atom.

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

One-photon matrix element:

\[ M^1(\vec{k}) = -i E_\Omega \langle \vec{k} | z | i \rangle \]
\[ \sim \exp[ i \eta_\ell(k) ] \]

Two-photon matrix element:

\[ M^2(\vec{k}) = -i E_\Omega E_\omega \]
\[ \times \sum_{\kappa'} \frac{\langle \vec{k} | z | \kappa' \rangle \langle \kappa' | z | i \rangle}{\epsilon_i + \Omega - \epsilon_{\kappa'}} \]
\[ \sim \exp[ i \phi_{cc}(k, \kappa) + i \eta_\ell(\kappa) ] \]
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.
Figure: Exact vs. asymptotic values of $\phi_{cc}(k, \kappa)$. 

Collaboration with A. Maquet and R. Taïeb at UPMC through COST.
Explicit phase of ATI transition: \( i \rightarrow \vec{\kappa} \rightarrow \vec{k} \):

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

with XUV: \( \Omega \) first, then continuum–continuum IR: \( \omega \).

( One intermediate angular momenta: \( \ell \). )
Explicit phase of ATI transition: $i \rightarrow \vec{k} \rightarrow \vec{k}$:

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

$$- \frac{\pi \ell}{2} + \eta_{\ell}(\kappa) + \phi_{cc}(k, \kappa),$$

with XUV: $\Omega$ first, then continuum–continuum IR: $\omega$.

(One intermediate angular momenta: $\ell$.)

-Now we apply this “ansatz” to experimental schemes!
Applications of ATI matrix elements: “RABBITT”

Figure: Ionization by APT.
Applications of ATI matrix elements: “RABBITT”

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

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

Figure: Ionization by APT+IR.
Applications of ATI matrix elements: “RABBITT”

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

\[
P(\mathbf{k}) \approx |M_a + M_e|^2 = |M_e|^2 + |M_a|^2 + 2 \Re \{M_e M_a^*\}
\]

The phase of the cross-term:

\[
\arg\{M_e M_a^*\} \approx -2\omega \times \tau + \phi_{\Omega>} + \eta_{\kappa>,\ell} + \phi_{cc}(k, \kappa>) - \phi_{\Omega<} - \eta_{\kappa<,\ell} - \phi_{cc}(k, \kappa<)
\]

Figure: Ionization by APT+IR.
Applications of ATI matrix elements: “RABBITT”

Figure: Ionization by APT+IR.

Probability of emission along \( \hat{z} \):

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Applications of ATI matrix elements: “RABBITT”

Probability of emission along \( \hat{z} \):

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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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Figure: Ionization by APT+IR.

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

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)]
What have we learned about “atomic delay” since 2001?

**Interpretation:**

“Atomic delay” $\approx$ Wigner delay + CC delay:

- 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
What happens if a resonance is embedded in the continuum?
Streaking with a resonance
Direct and autoionizing processes

Dressed continuum
Quasibound state
Direct path
Delayed path
Decay
\[ \tau = \frac{1}{\Gamma} \]
Asymmetric Fano peak
Photoelectron distribution depends on $q$-parameter

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:

\[
\left| \frac{\langle \psi \mid T \mid g \rangle}{\langle \psi \mid T \mid g \rangle} \right|^2 = \frac{(q + \epsilon)^2}{1 + \epsilon^2}
\]

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

Corresponding complex amplitude:

\[
\langle \psi \mid T \mid g \rangle = \frac{q + \epsilon}{1 - i\epsilon} \left[ f_F(E) \right] \langle \psi \mid T \mid g \rangle
\]
Fano theory transition probability ratio:

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

Can this phase shift be measured?
RABBITT 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)]
Corresponding complex amplitude:

\[
\langle \Psi | T | g \rangle = \frac{q + \epsilon}{1 - i\epsilon} \langle \psi | T | g \rangle
\]

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)]
Fano theory in time domain

Corresponding complex amplitude:

\[
\langle \psi | T | g \rangle = \frac{q + \epsilon}{1 - i\epsilon} \langle \psi | T | g \rangle_f F(E)
\]

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

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

The Fano phase is proportional to \(\epsilon\) for \(q = 0\) \((Task : 7)\)
which implies that the \(\tau_W\) at the resonance is \(2/\Gamma = \tau_R\).

See: [Z X Zhao and C D Lin PRA 71, 060702 (2005)]
Electrons ejected into the continuum:

Direct path + Decay (exponential tail)
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!
Redistribution of three harmonic peaks due to laser dressing of window resonance ($q = 0$).

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Photoelectron spectrogram
One photon absorption to dressed continuum with autoionization

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Time scales of autoionization longer than laser period!
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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- **Lund University (LTH)**
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  - Thomas Carette

- **Université Pierre et Marie Curie (UPMC)**
  - Alfred Maquet
  - Richard Taïeb

http://www.matfys.lth.se/staff/Marcus.Dahlstrom/

*Thank you for your attention!*