Diagrams for resonances in RABITT:

Resummation of ATI matrix elements in second quantization

J. Marcus Dahlström

PhD: Lund [LTH] -> Post-doc: Stockholm [SU] -> Guest researcher: Hamburg [CFEL/MPGPKS]
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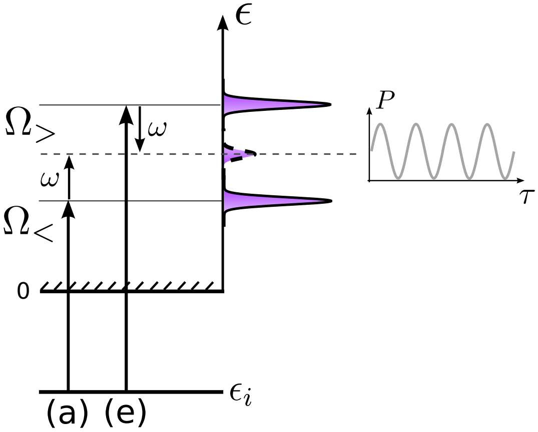
2014-09-11 KITP: Frontiers of Intense Laser Physics, Santa Barbara, USA

AlbaNova MPG-PKS

The RABITT method

Reconstruction of Attosecond Beating by Interference of Two-photon Transitions:

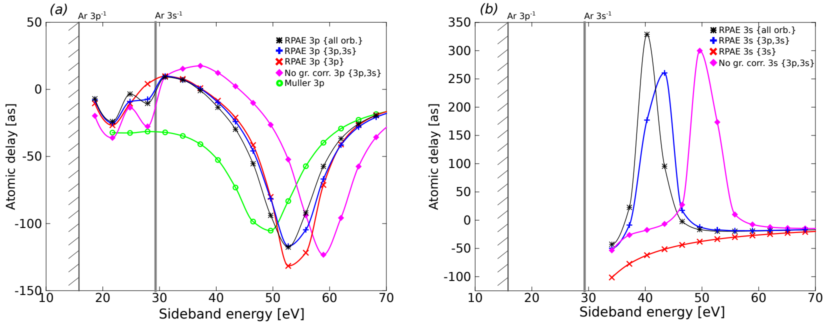


Experiment: *Paul et al. Science (2001)* **292** *1689*

Theory: *Muller (2002) Appl. Phys. B* **74** *s17-21*

Argon: Atomic delay depends on correlation effects

RPAE correlation with experimental binding energies (not HF values):



- Ionization from 3*p*: *intra-orbital* correlation (3p only).
- Ionization from 3*s*: *inter-orbital* correlation (M-shell).
- Ground-state correlation is always important (beyond CIS).

Method: *Dahlström and Lindroth J. Phys. B (2014)* **47** *124012*

Experimental work using XUV attosecond pulse trains and IR probe field:

- Above-threshold resonance in N_2:
  $(B^2\Sigma^+(3d\sigma_g)^1\Sigma^+ = 16.7\,\text{eV} \approx 11\hbar\omega)$

  Haessler et al. PRA (2009)

- Below-threshold resonance in *He*:
  $(1s3p = 23.1\,\text{eV} \approx 15\hbar\omega)$

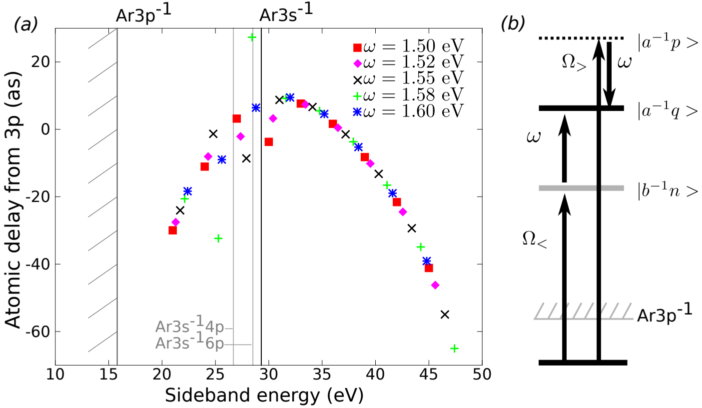
  Swoboda et al. PRL (2011)

- Above-threshold resonance in *Ar*:
  $(3s3p^6 4p = 26.6\,\text{eV} \approx 17\hbar\omega)$

  Kotur et al. Manuscript in preparation (Lund).

Resonance effect in RABITT by RPAE screening

Atomic delay in Ar with final hole in 3*p*



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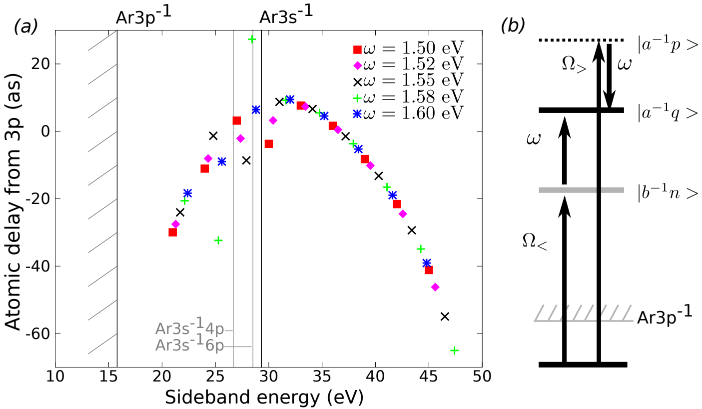

- Random Phase Approximation (RPAE) for XUV absorption: *Amusia (1990) Atomic Photoeffect*

Resonance effect in RABITT by RPAE screening

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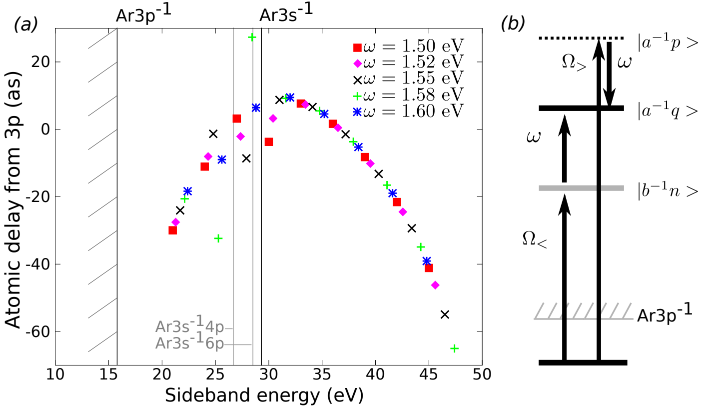
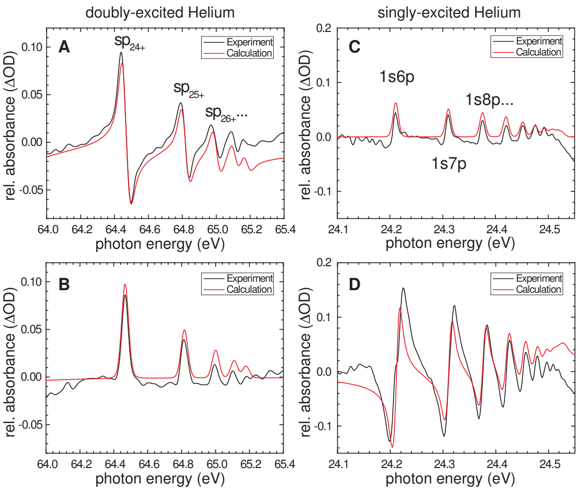


- Random Phase Approximation (RPAE) for XUV absorption: *Amusia (1990) Atomic Photoeffect*
- Fano for *Ar* $3s^{-1}4p$: $q_{RPAE} = 1.15 \neq q_{exp} = -0.22$. *Amusia and Kheifets (1981) Phys. Lett. A* **82** *407*

Motivation

Dynamical control of Fano's $q-$parameter in **attosecond transient absorption**:



Experiment and theory: *Ott et al. Science (2013)* **340** *716*

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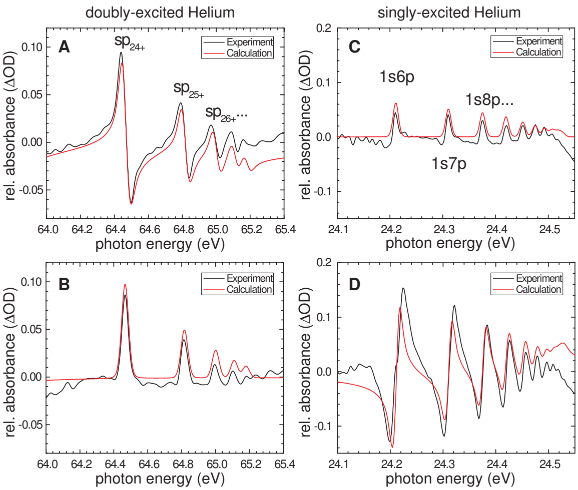

Experiment and theory: *Ott et al. Science (2013)* **340** *716*

Can the phase be probed/controlled using **RABITT**?

Independent electron Hamiltonian:

$$h_\ell(r) = -\frac{1}{2}\frac{d^2}{d^2 r} + \frac{\ell(\ell+1)}{2r^2} - \frac{Z}{r} + u_{HF}(r) + u_P(r)$$

Restricted Hartree-Fock (sum orbitals b with occupancy q_b):

$$u_{HF}(r) = \sum_b q_b[J_b(r) - \frac{1}{2}K_b(r)],$$

Lindgren and Morisson Atomic Many-Body Theory, Springer (1982)

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$$J_b(r)P_a(r) = \sum_k c(abk)\frac{1}{r}Y_k(bb,r)P_a(r),$$

$$K_b(r)P_a(r) = -2\sum_k d(abk)\frac{1}{r}Y_k(ab,r)P_b(r),$$

$$Y_k(ab,r) = r\int_0^\infty dr' \frac{r_<^k}{r_>^{k+1}} P_a(r')P_b(r')$$

Lindgren and Morisson Atomic Many-Body Theory, Springer (1982)

Method

Projected potential $u_P(r)$ alters excited electron orbitals

Excited electrons must see a long range $-1/r$ potential:

$$u_P = -P\frac{1}{r}J_0(aa,r)P,$$

$$P = \sum_p^{exc} |\, P_p \,\rangle\langle\, P_p \,|,$$

$$B = \sum_b^{occ} |\, P_b \,\rangle\langle\, P_b \,|$$

The **projected potential ensures Rydberg series** and Coulombic asymptotic wavefunctions for excited states.

Details: *Dahlström and Lindroth J. Phys. B (2014)* **47** *124012*

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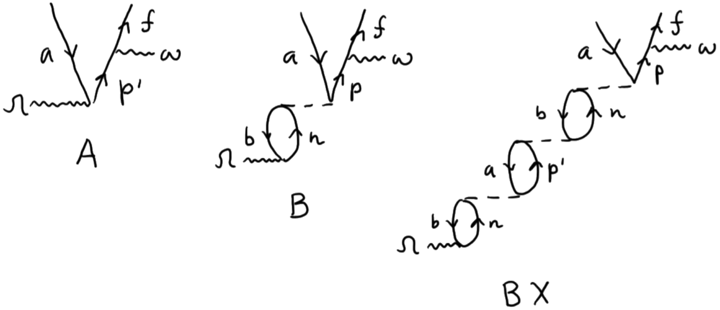
Fermi vacuum (second quantization): $1 = B + P$.
Holes ($\downarrow$: B occupied) and electrons ($\uparrow$: P excited).

Details: *Dahlström and Lindroth J. Phys. B (2014)* **47** *124012*

Diagrams for RABITT: A, B and BX

A simple correlation model based on the Tamm-Dancoff approximation:



- A single resonance coupled to a continuum: $b^{-1}n \leftrightarrow a^{-1}p$.
- $b =$ inner hole; and $a =$ outer hole (closer to Fermi surface).
- Example: $Ar\,3s^{-1}4p \leftrightarrow Ar\,3p^{-1}kd$

Diagram A: Uncorrelated RABITT
Formation of outgoing intermediate wave packet:

Direct transition to final state via virtual orbitals p:

$$A = E_\omega E_\Omega \lim_{\xi\to 0} \sum\!\!\!\!\!\!\!\int_p \frac{\langle\, f \mid z \mid p \,\rangle\langle\, p \mid z \mid a \,\rangle}{(\epsilon_a + \Omega_0 - \epsilon_p + i\xi)} \equiv \sum\!\!\!\!\!\!\!\int_p \frac{z_{fp} z_{pa}}{a + \Omega - p}$$

Tutorial: *Dahlström et al. J. Phys. B (2012)* **45** *183001*

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The boundary condition ensures that:
the intermediate electron wavepacket is outgoing:

$$\sum\!\!\!\!\!\!\!\int_p \frac{P_p z_{pa}}{(a + \Omega - p)} \propto \exp[i(k_{p_0} r + \Phi(k_{p_0})] z_{p_0 a}, \;\; r \to \infty$$

where $p_0 = \Omega + a = \Omega - I_p^{(a)} > 0$ is the "on-shell energy".

Tutorial: *Dahlström et al. J. Phys. B (2012)* **45** *183001*

Resummation of diagram B

The lowest-order term is divergent for $\Omega_0 \approx n - b$.

Lowest-order process (1 Coulomb interaction):

$$B = \sum\!\!\!\!\!\!\!\int_p \frac{z_{fp} c_{bpna} z_{nb}}{(a + \Omega - p)(b + \Omega - n)}$$

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Higher-order process (3 Coulomb interactions):

$$BX = \sum\!\!\!\!\!\!\!\!\int_p \frac{z_{fp} c_{bpna}}{(a + \Omega - p)} \left(\sum\!\!\!\!\!\!\!\!\int_{p'} \frac{c_{anp'b} c_{bp'na}}{(b + \Omega - n)(a + \Omega - p')} \right) \frac{z_{nb}}{(b + \Omega - n)}$$

Resummation of diagram B

The lowest-order term is divergent for $\Omega_0 \approx n - b$.

Higher-order process: $2N + 1$ (odd Coulomb interactions)

$$BX^N = \sum\!\!\!\!\!\!\int_p \frac{z_{fp} c_{bpna}}{(a + \Omega - p)} \left(\sum\!\!\!\!\!\!\int_{p'} \frac{c_{anp'b} c_{bp'na}}{(b + \Omega - n)(a + \Omega - p')} \right)^N \frac{z_{nb}}{(b + \Omega - n)}$$

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Geometric series (each term is divergent but sum is finite):

$$\frac{B}{1-X} = B + BX + BX^2 + \ldots = \sum\!\!\!\!\!\!\int_p \frac{z_{fp} c_{bpna} z_{nb}}{(a+\Omega-p)(b+\Omega-n-\Sigma_{nb})}.$$

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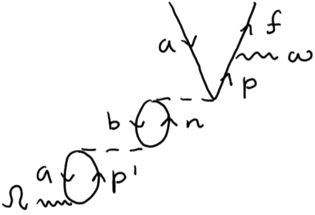
Complex energy of resonance (no divergence):

$$(n - b) \to (n - b + \Sigma_{nb}), \qquad \Delta = \mathrm{Re}\Sigma_{nb}, \qquad \Gamma/2 = -\mathrm{Im}\Sigma_{nb}.$$

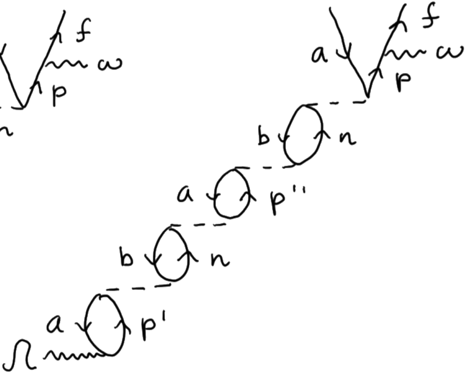
$$\Sigma_{nb} = \sum_{p'} \frac{c_{anp'b} c_{bp'na}}{(a + \Omega - p')} = \mathrm{p.v.} \sum_{p'} \frac{c_{anp'b} c_{bp'na}}{(a + \Omega_0 - p')} - i\pi c_{anp_0 b} c_{bp_0 na}$$

Diagrams for RABITT: C and CX

A simple correlation model based on the Tamm-Dancoff approximation:



C



CX

- The electron may directly go to the continuum and then flicker through the resonance multiple times.
- Even number of Coulomb interactions.

Resummation of diagram C

The lowest-order term is divergent for $\Omega_0 \approx n - b$.

Higher-order process: $2N+2$ (even Coulomb interactions)

$$CX^N = \sum_{p,p'} \frac{z_{fp} c_{bpna} c_{anp'b} z_{p'a}}{(a+\Omega-p)(b+\Omega-n)(a+\Omega-p')} \times \left(\sum_{p''} \frac{c_{anp''b} c_{bp''na}}{(b+\Omega-n)(a+\Omega-p'')} \right)^N$$

Geometric series gives resummed resonance energy:

$$\frac{C}{1-X} = \sum_{p,p'} \frac{z_{fp} c_{bpna} c_{anp'b} z_{p'a}}{(a+\Omega-p)(b+\Omega-n-\Sigma_{nb})(a+\Omega-p')}$$

Atomic physics resummation: *Pan and Kelly PRA* **39** *6232 (1989)*

Correlated two-photon matrix element

"Almost as easy as A, B, C":

Correlated two-photon matrix element with one resonance:

$$M^1_{fa} = A + \frac{B}{1-X} + \frac{C}{1-X}$$

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$$= \sum\!\!\!\!\!\!\int_p \frac{z_{fp}}{(a+\Omega-p)} \underbrace{\left(z_{pa} + \frac{c_{bpna}}{(b+\Omega-n-\Sigma_{nb})} \left[z_{nb} + \sum\!\!\!\!\!\!\int_{p'} \frac{c_{anp'b} z_{p'a}}{(a+\Omega-p')} \right] \right)}_{\text{Effective one-photon matrix element: } z^{eff}_{pa}}$$

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$$= \sum\!\!\!\!\!\!\int_p \frac{z_{fp} z^{eff}_{pa}}{(a+\Omega-p)}$$

Fano's theory for autoionization
Do we recover the same q parameter?

Eq.(21): Ratio of transition rates (Ψ_E = corr., ψ_E =uncorr.):

$$\frac{|\langle \Psi_E \mid T \mid i \rangle|^2}{|\langle \psi_E \mid T \mid i \rangle|^2} = \frac{(q+\epsilon)^2}{1+\epsilon^2} \rightarrow \frac{\langle \Psi_E \mid T \mid i \rangle}{\langle \psi_E \mid T \mid i \rangle} = \frac{q+\epsilon}{\epsilon+i} = 1 + \frac{q-i}{\epsilon+i}$$

Configuration interaction: *Fano Phys. Rev. (1961)* **124** *1866*

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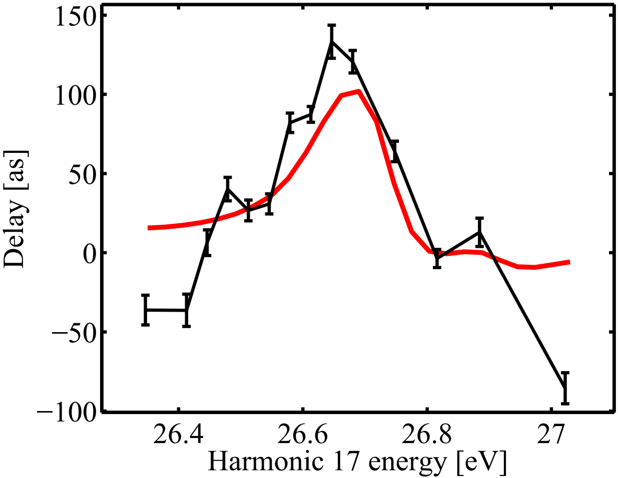
Fano's q–parameter for $z^{eff}_{p_0 a}$ from our correlation model:

$$q = \frac{z_{nb} + \mathrm{p.v.}\sum\!\!\!\!\!\!\int_{p'} \frac{c_{anp'b} z_{p'a}}{a+\Omega-p'}}{\pi c_{anpb} z_{pa}}$$

Derivation: *Dahlström and Lindroth J. Phys. B (2014)* **47** *124012*

RABITT measurement of Fano phase

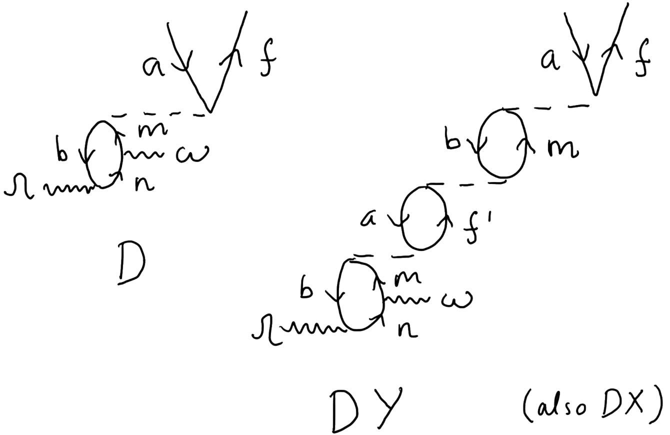
Phase extraction of Ar: $3s^{-1}4p$, lifetime ~ 8 *fs*



Experiment: *Kotur et al. Manuscript in preparation (Lund).*
Theory (MCHF): *Carette et al. PRA (2013)* **87** *023420*

Resonance in intermediate *and* final state: D, DX and DY

Resonance (b,m) is of opposite parity compared to (b,n)



- When b hole: IR photon induces transition: $m - n \approx \omega$.
- Resummation of both $(b - m)$ and $(b - m)$:
  $(D + DY + ...) + ...(D + DY + ...)X + ... = D/(1 - X)(1 - Y)$.

Resonance in intermediate *and* final state: E and EY

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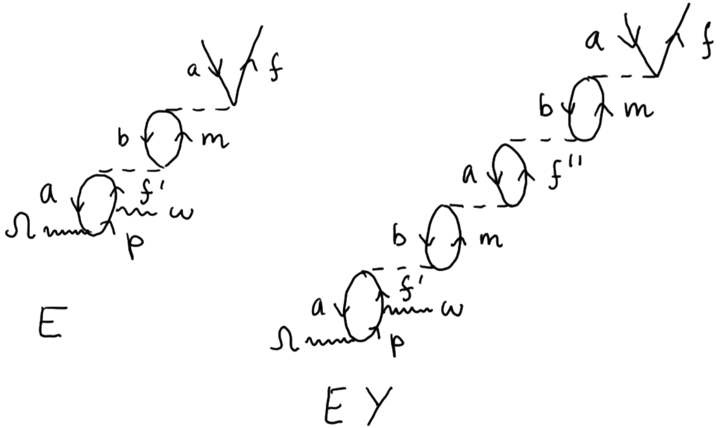
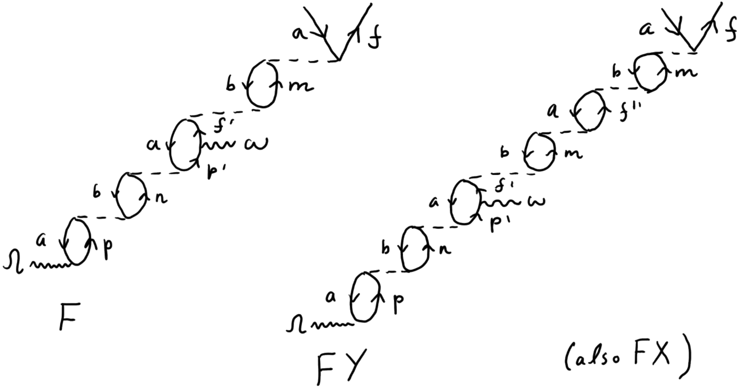


- When *a* hole: IR photon (ω) induces transition from p to f'.
- Resummation of $(b - m)$:
  $(E + EY + ...) = E/(1 - Y)$.

Resonance in intermediate *and* final state: F, FX and FY

Resonance (b,m) is of opposite parity compared to (b,n)



- When a hole: IR photon (ω) induces transition from p' to f'.
- Resummation of both $(b-m)$ and $(b-m)$:
  $(F+FY+...)+...(F+FY+...)X+...=F/(1-X)(1-Y)$.

Correlated two-photon matrix element

There are six distinct processes:

Correlated two-photon matrix element with two resonances:

$$M_{fa}^2 = \underbrace{A + \frac{B+C}{1-X}}_{\text{"Direct" path to final state: } f} + \underbrace{\frac{1}{1-Y}\left\{E + \frac{D+F}{1-X}\right\}}_{\text{Path through resonance: } m \to f}$$

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$$= M^1_{fa} + \frac{1}{(b+\Omega+\omega-n-\Sigma_{mb})}$$

$$\times \left\{ \sum\!\!\!\!\!\!\int_{pf'} \frac{c_{bfma} c_{amf'b} z_{f'p} z_{pa}}{(a+\Omega+\omega-f')(a+\Omega-p)} \quad \text{(continuum – continuum)} \right.$$

$$+ \frac{c_{bfma} z_{mn} z_{nb}}{(b+\Omega-n-\Sigma_{nb})} \quad \text{(bound – bound : 3 – level system)}$$

$$\left. + \sum\!\!\!\!\!\!\int_{pp'f'} \frac{c_{bfma} c_{amf'b} z_{f'p'} c_{bp'na} c_{anpb} z_{pa}}{(a+\Omega+\omega-f')(a+\Omega-p')(b+\Omega-n-\Sigma_{nb})(a+\Omega-p)} \right\}.$$

- Found analytical expression for correlated two-photon amplitudes based on Tamm-Dancoff approximation.
- The divergence of lowest orders was removed by geometric series (Pan and Kelly 1989).
- Above-threshold ionization with two resonances of different parity leads to new quantum paths for ionization through laser-driven bound-bound transitions followed by autoionization.
- Prospect: q parameters for two-photon transitions?

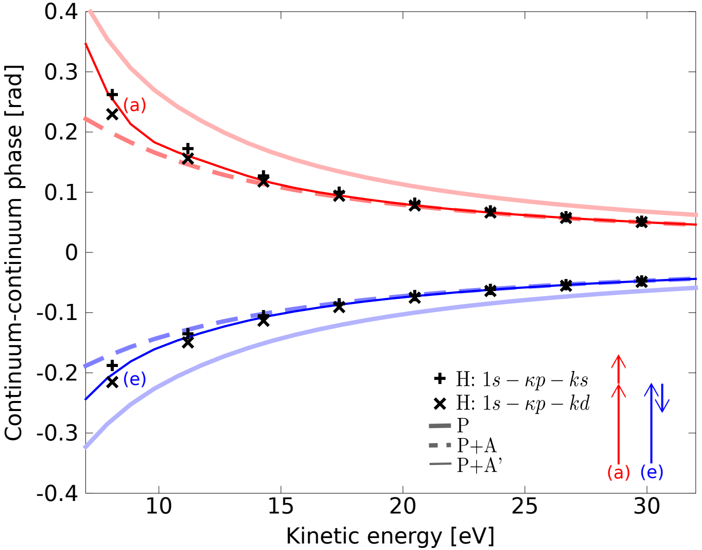
More details about our method:
Dahlström and Lindroth J. Phys. B (2014) **47** *124012*

Independent work on RABITT resonances:
Jiménez-Galán, Argenti and Martín (2014) arXiv:1405.4732v1

Thank you for your attention!

Phases induced in the transition: $k_0 \rightarrow k_f$

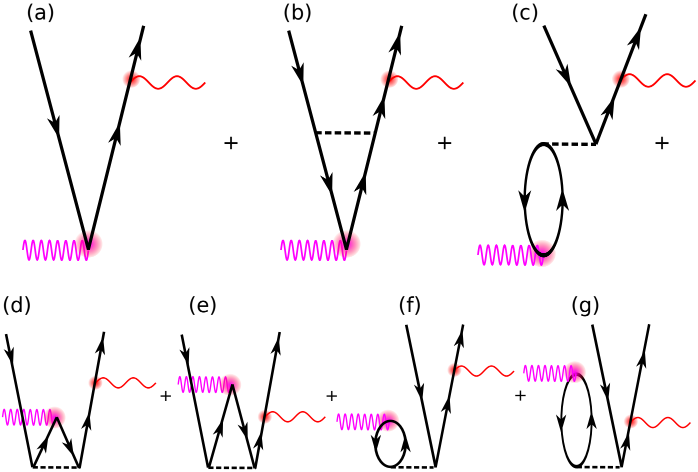
Continuum–continuum (cc) phase compared to exact calculation in H:

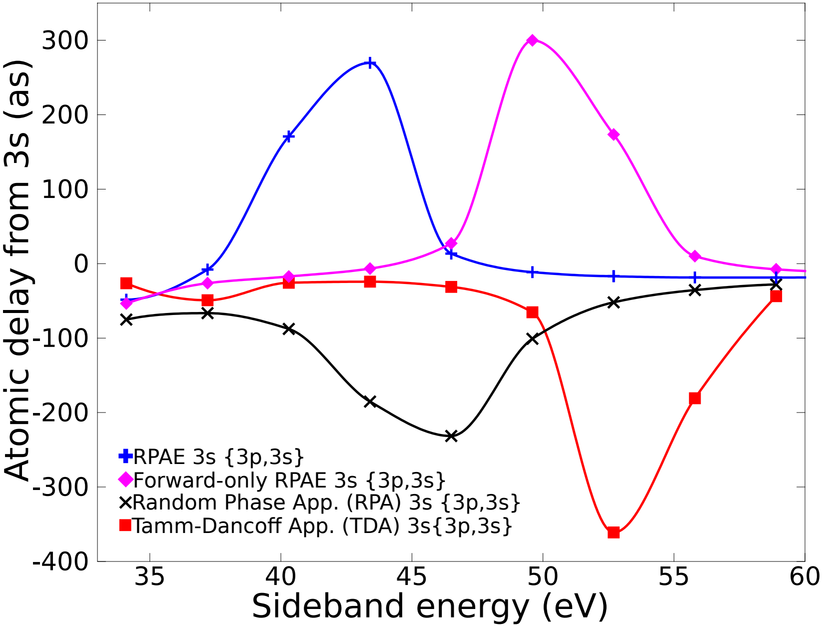


Exact calculation by R. Taïeb.

Two-photon diagrams with RPAE screening of XUV

Coulomb interactions are summed by iteration





Atomic delay from 3s (as)
300
200
100
0
-100
-200
-300
-400
35
40
45
50
55
60
Sideband energy (eV)
RPAE 3s {3p,3s}
Forward-only RPAE 3s {3p,3s}
Random Phase App. (RPA) 3s {3p,3s}
Tamm-Dancoff App. (TDA) 3s{3p,3s}