Probing attosecond pulses by XUV stimulated dynamics

J. Marcus Dahlström

PhD: Lund [LTH] -> Post-doc: Stockholm [SU] -> Guest res.: Hamburg [CFEL/MPGPKS]

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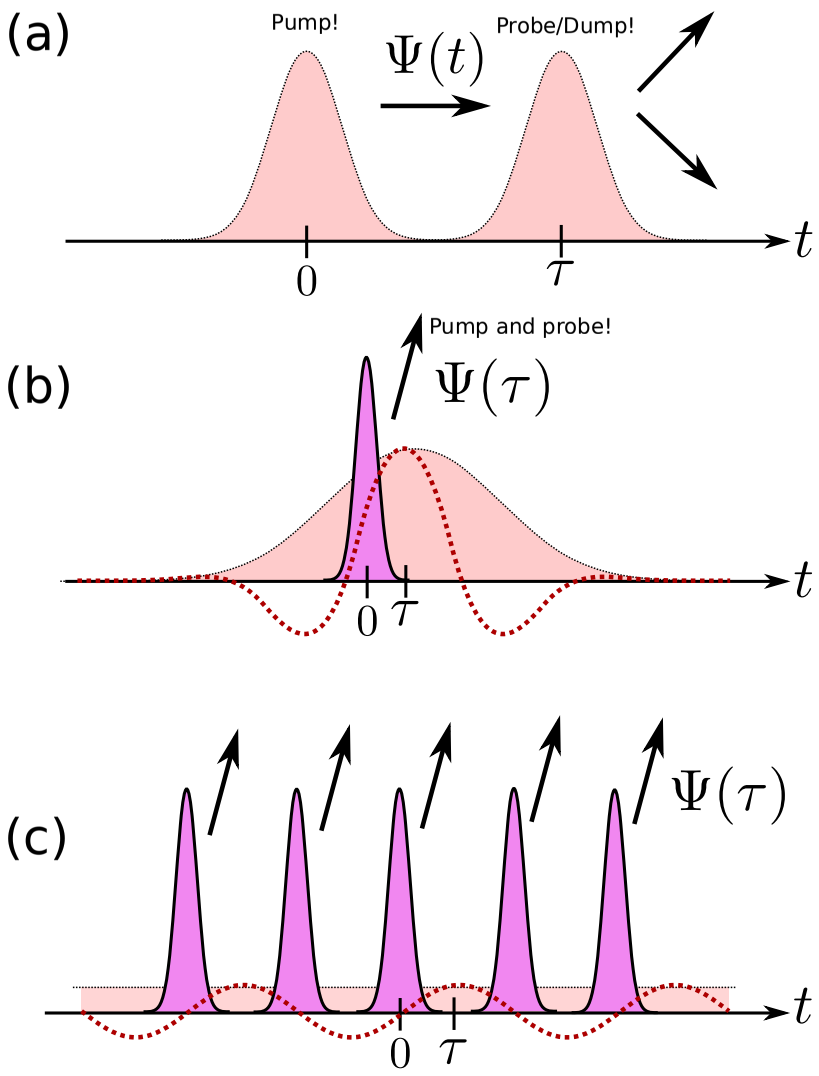
AlbaNova

MPG-PKS

Outline of talk:

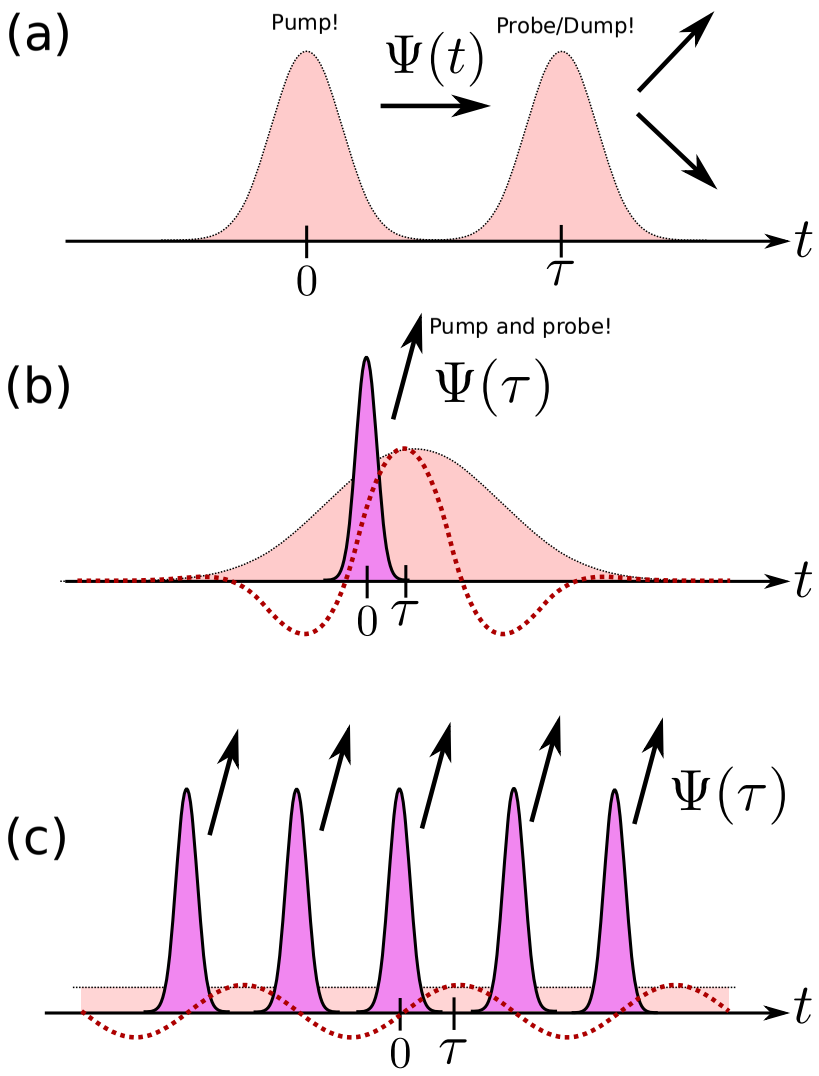
- Introduction to “attophysics”
- Details on computational method (SU: MBPT)
- New proposal:
  Attosecond interferometry based on stimulated hole dynamics

Introduction to "attophysics"
Overview of pump and probe setups



$(1\,as{=}10^{-3}\,fs{=}10^{-18}\,s)$

Traditional pump and probe with *femto-pulses* control of Born-Oppenheimer dynamics:



Traditional pump and probe with *femto-pulses* control of Born-Oppenheimer dynamics:

- *Tannor-Rice:* (t-domain)
- *Brumer-Shapiro:* (ω-dom.)

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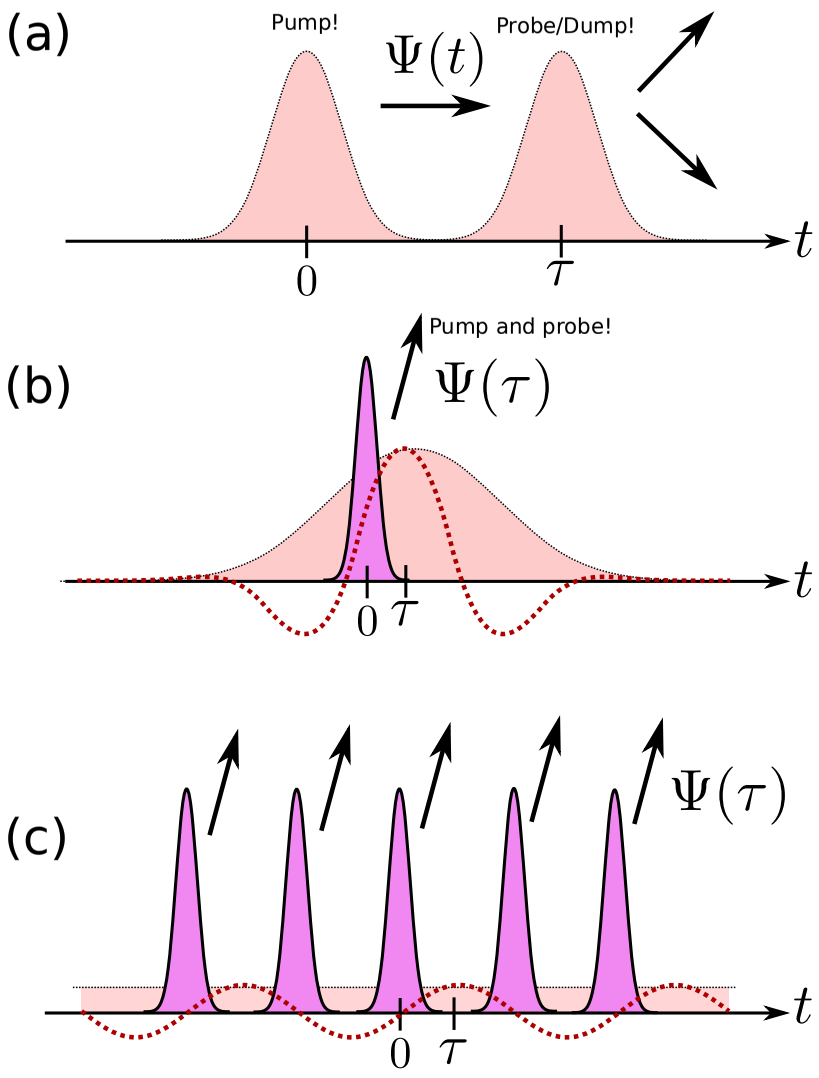

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Attosecond streak-camera: photoionization by XUV *atto-pulse* and an IR fs probe field. Control of e^- wave packet.

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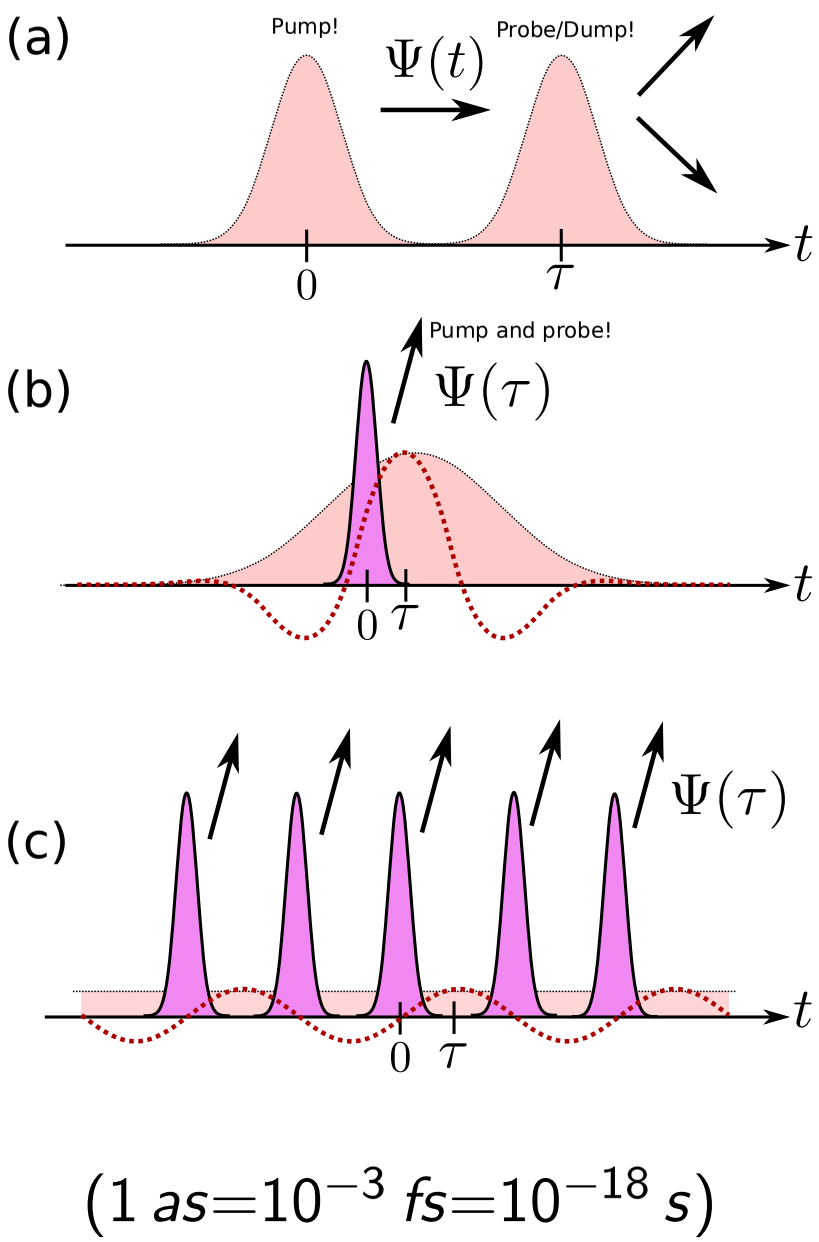

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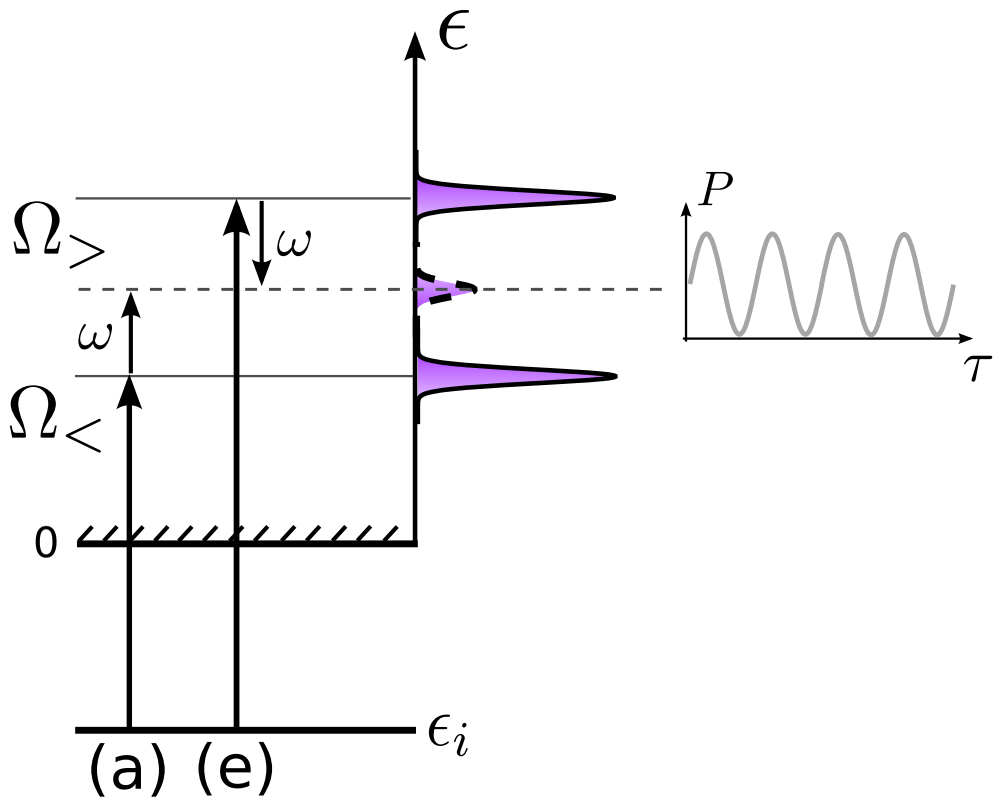
Attosecond streak-camera: photoionization by XUV *atto-pulse* and an IR fs probe field. Control of e^- wave packet.

RABBITT method: photoionization by *train of atto-pulses* and an IR fs probe field $\rightarrow$ interference in ω-domain.

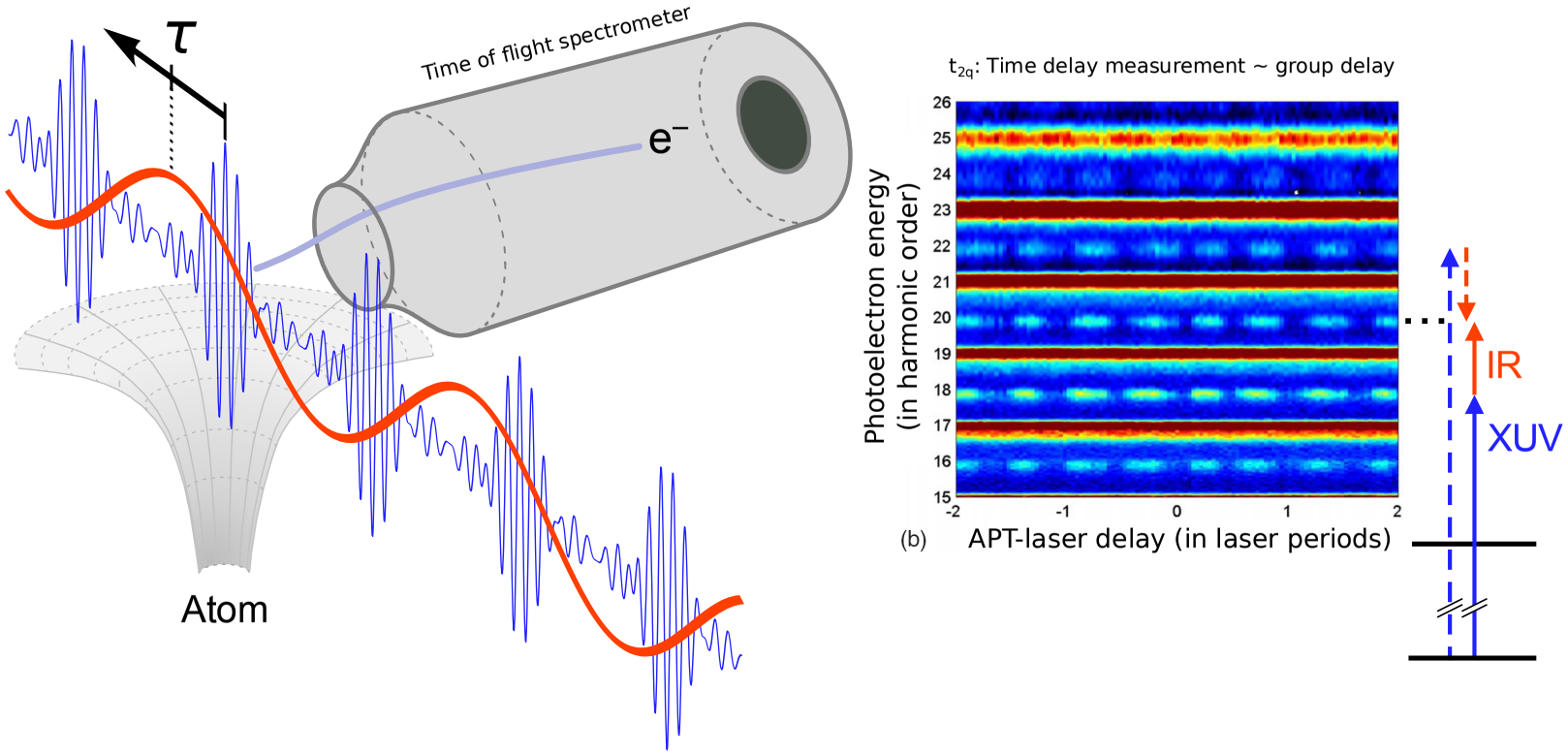
Attosecond pulse train $\rightarrow$ *odd* XUV harmonics: $(2q+1)\omega$.



Experiment: *Paul et al. Science (2001)* **292** *1689*
Theory: *Muller (2002) Appl. Phys. B* **74** *s17-21*

Atomic delay in *single* ionization

Sketch of the RABBITT setup



- *Time-of-flight* tube determines the *energy* of the electron.

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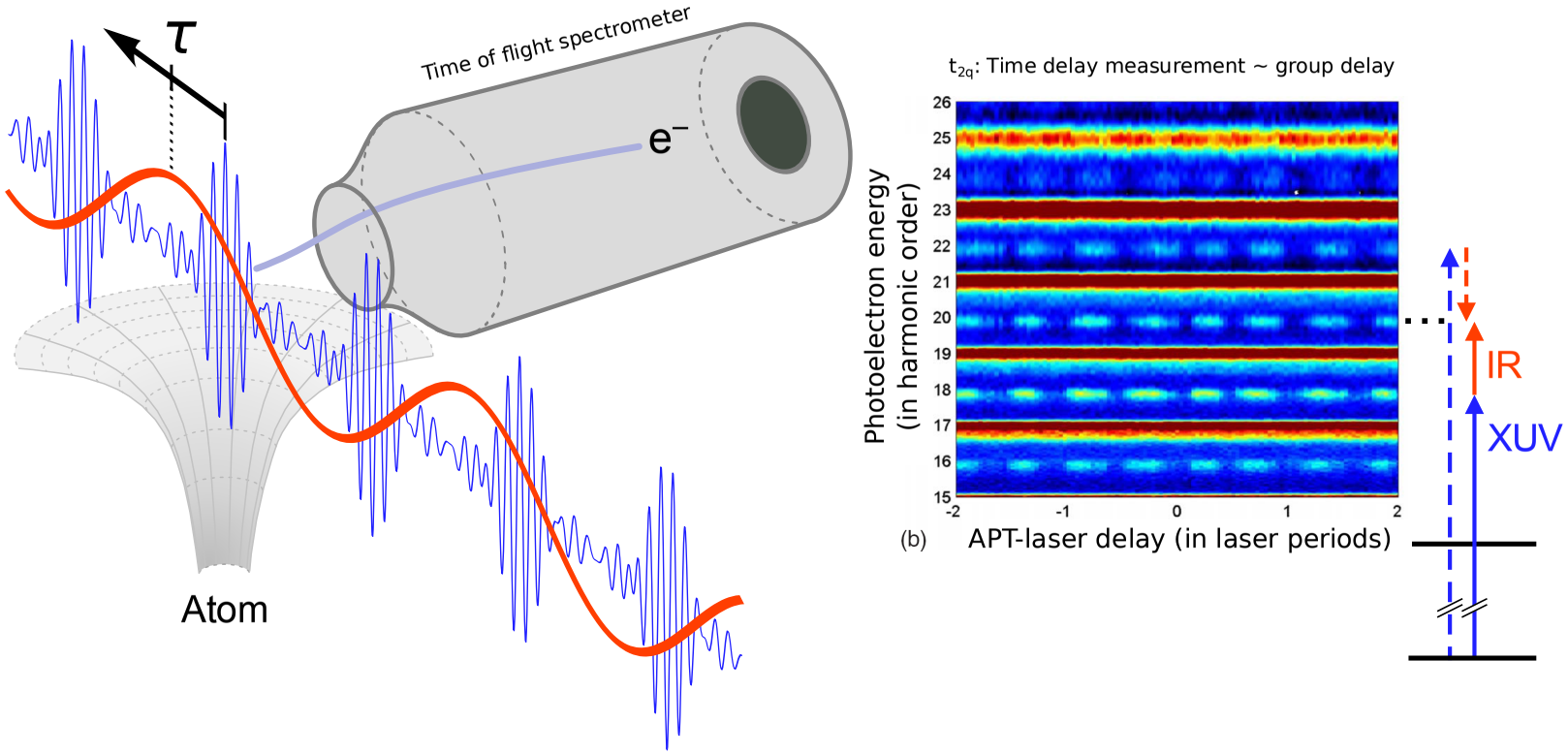

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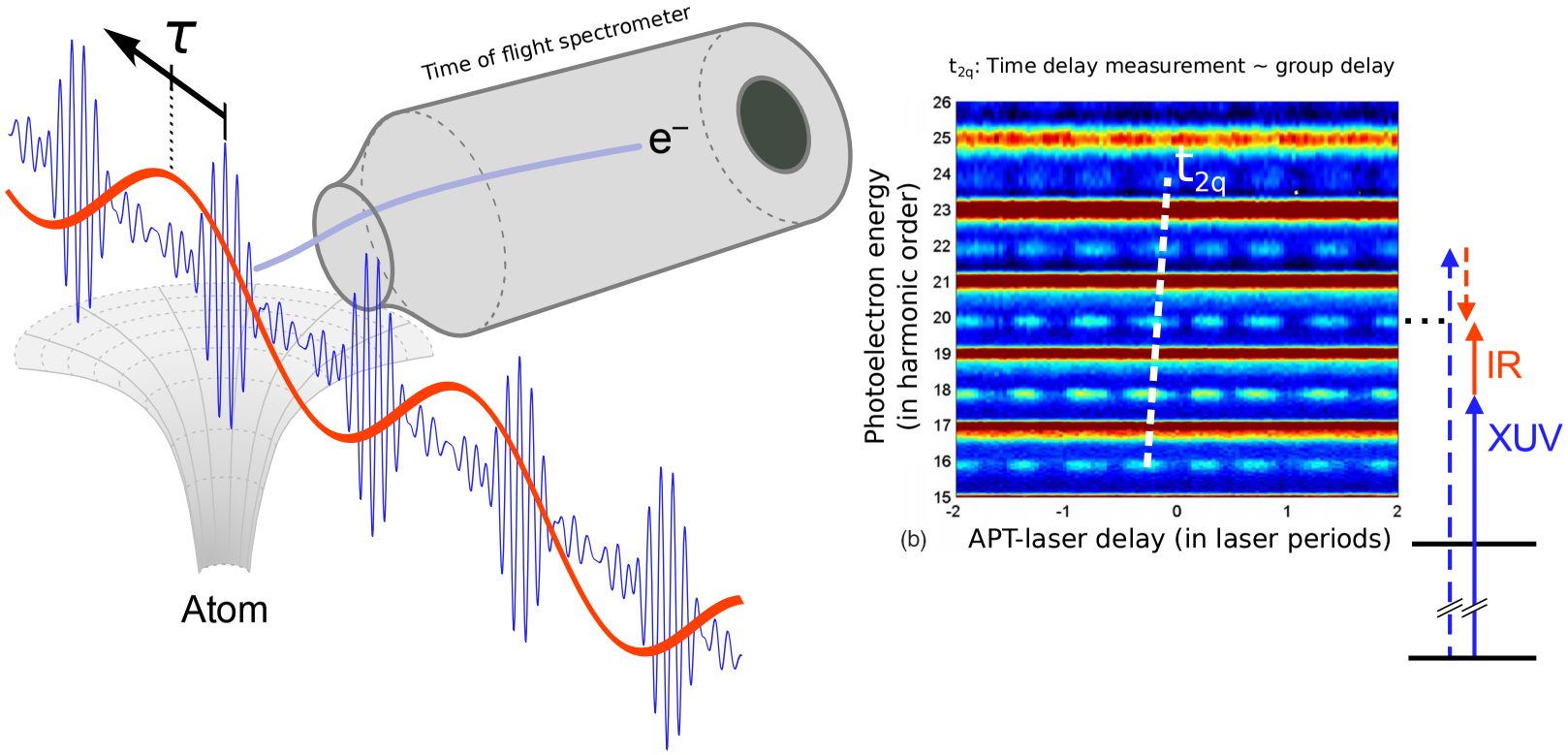

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Inter-species photoionization delay experiment
(in attoseconds, $1\,as = 10^{-18}\,s$)

Sideband	20	22	24
$\tau(Ar) - \tau(Ne)$	68 ± 15	70 ± 12	52 ± 25
Theory	60	51	40
$\tau(Ar) - \tau(He)$	82 ± 15	83 ± 22	71 ± 21
Theory	72	59	45
$\tau(Ne) - \tau(He)$	23 ± 4	12 ± 4	10 ± 8
Theory	12	8	4

The delay is relative to the same sideband order.

- Ar has a larger delay than both Ne and He.
- Theory shows slightly smaller delays.

[Guénot et al 2014 J. Phys. B: At. Mol. Opt. Phys. 47 245602]:

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The RABBITT method is *not* perfect for attosecond metrology because we measure *different* delays for different target atoms.

[Guénot et al 2014 J. Phys. B: At. Mol. Opt. Phys. 47 245602]:

Independent electron Hamiltonian:

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

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)

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

$$u_Q = -\hat{Q}\frac{1}{r}J_0(aa, r)\hat{Q},$$

$$\hat{P} = \sum_a^{occ} | P_a \rangle\langle P_a |$$

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

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

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

Method

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

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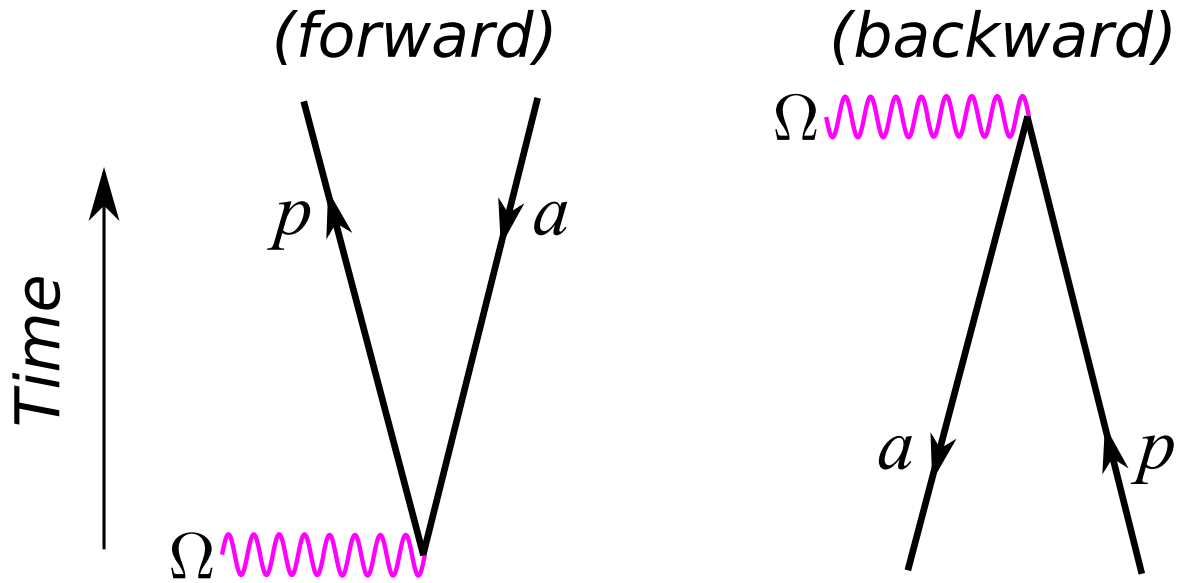
The **projected potential ensures Rydberg series** and Coulombic asymptotic wavefunctions for excited states.

Fermi vacuum (second quantization): $1 = \hat{P} + \hat{Q}$.
Holes ($\downarrow \in \hat{P}$ occupied) and electrons ($\uparrow \in \hat{Q}$ excited).

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

Diagrammatic perturbation theory

Setup of uncorrelated perturbed wavefunction



$$\rho_{fwd}(a) = \sum\!\!\!\!\!\!\int_p \frac{\ldots \mid p \rangle\langle\, p \mid d_\Omega \mid a \,\rangle}{\epsilon_a + \Omega - \epsilon_p}$$

$$\rho_{bwd}(a) = \sum\!\!\!\!\!\!\int_p \frac{\langle\, a \mid d_\Omega \mid p \,\rangle\langle\, p \mid \ldots}{\epsilon_a - \Omega - \epsilon_p}$$

[Mårtensson-Pendrill J. Phys. France 46, 1949 (1985); Dahlström *et al.* Phys. Rev. A 86, 061402 (2012)]

RPAE-type effects in XUV-IR processes:
(RPAE=Random Phase Approximation with Exchange)

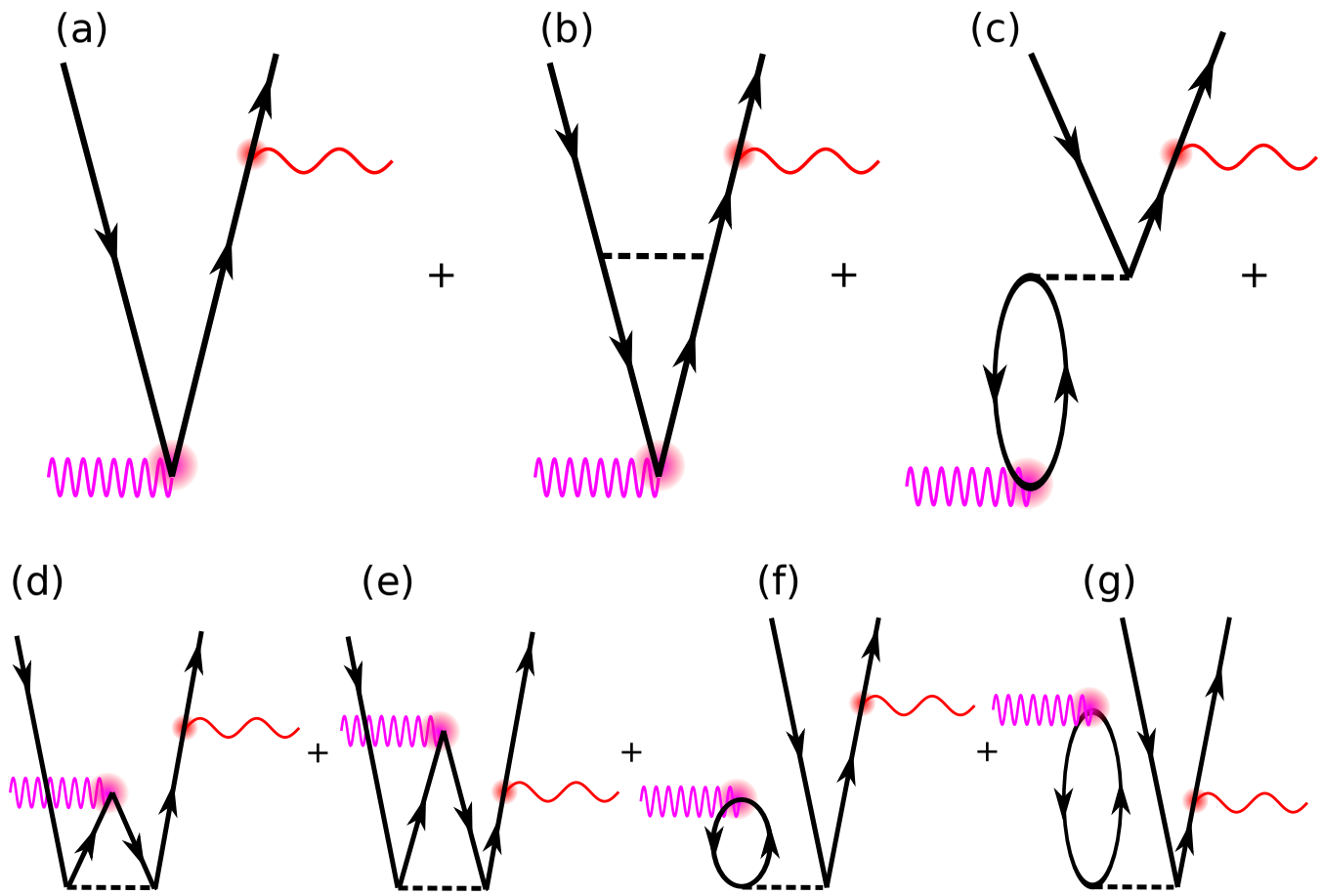
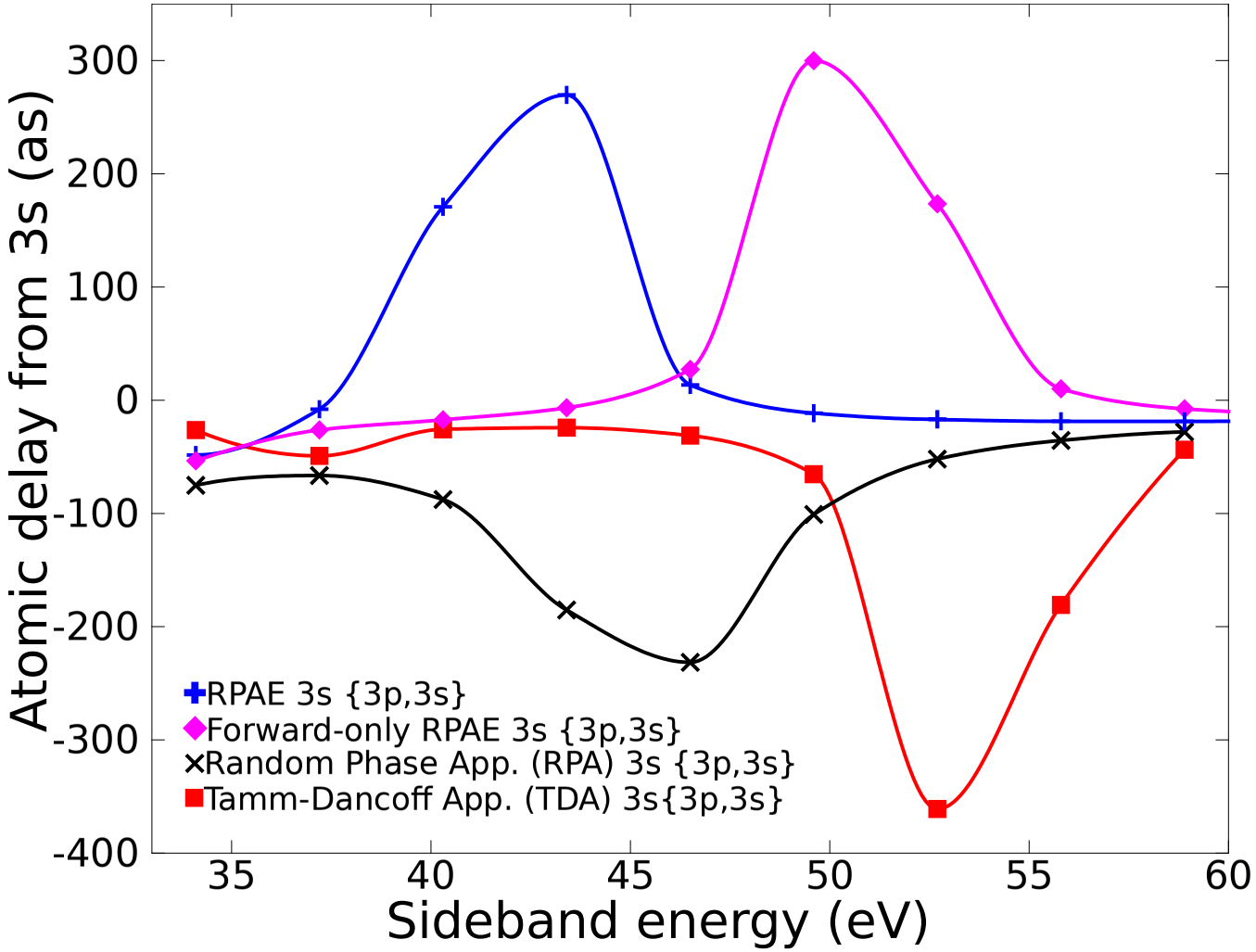


Figure: **XUV photon absorbed first** including **RPAE-type correlation**, and then interaction with the **IR field last**: (a) "Hartree-Fock"; (b) Direct; (c) Exchange; and (d)-(g) Ground-state correlations. *Final-state correlation is not included*.

"Worst case": Correlation effects in Ar3s^{-1}

RABBITT delay depends strongly on correlation effects:

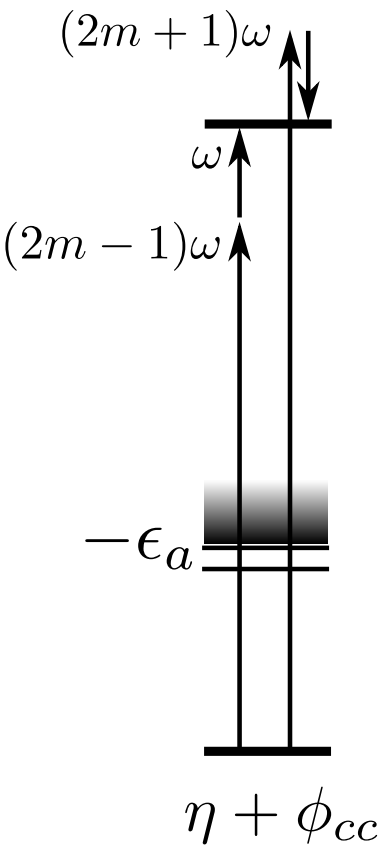


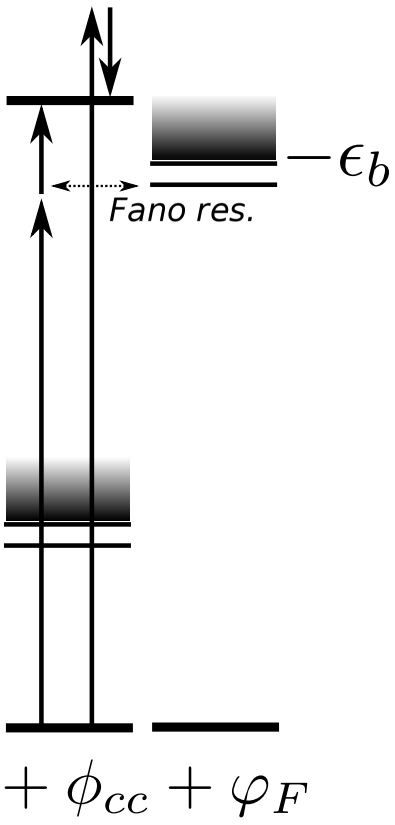
[Dahlström & Lindroth J.Phys.B **47** (2014) 124012]

Overview of interferometric schemes

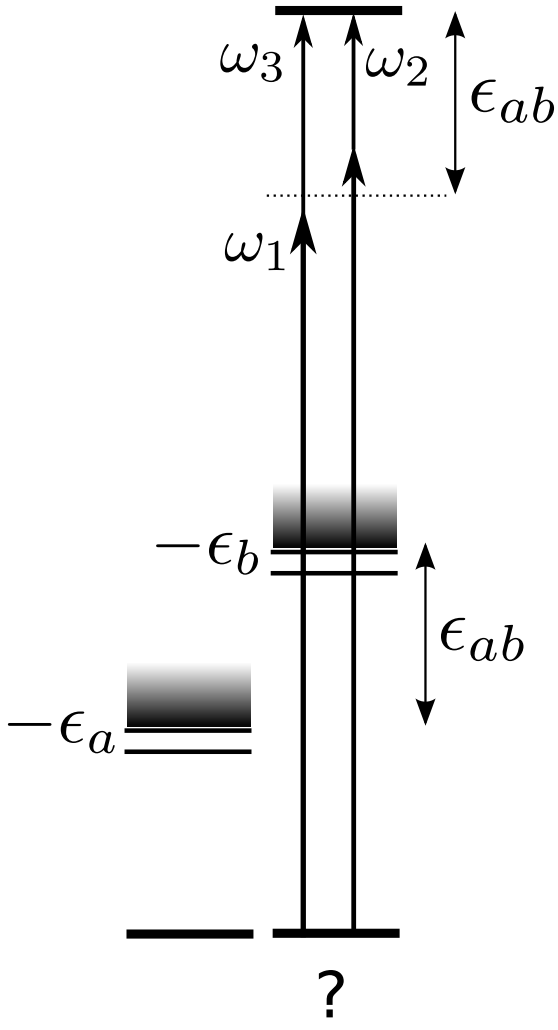
IR-driven electron dynamics → XUV-stimulated hole transition

XUV HARMONICS + IR PROBE:



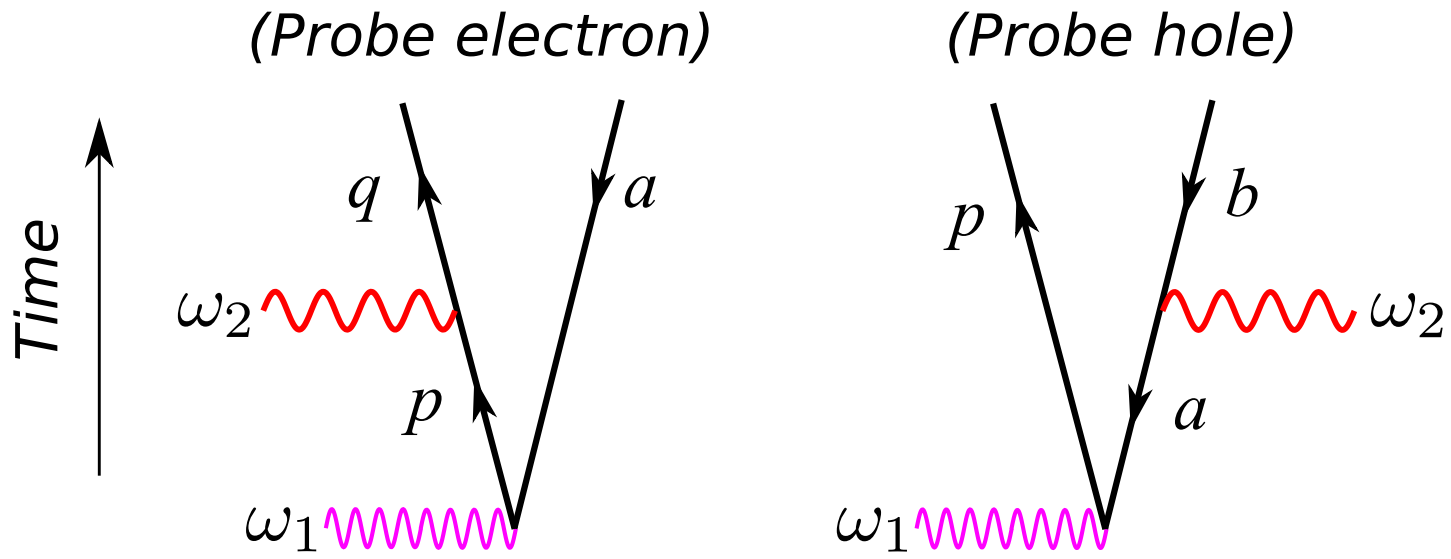


XUV PUMP AND XUV HARMONIC PROBE:



Considerations for a real probe scheme

Probe photoelectron *or* remaining hole in target?



$$M_{qa}^{(e)}(\omega_1,\omega_2) = \lim_{\varepsilon\to 0^+} \sum_n \frac{\langle\, q \mid O_2 \mid p \,\rangle\langle\, p \mid O_1 \mid a \,\rangle}{(\epsilon_a + \omega_1 - \epsilon_p + i\varepsilon)} \in \mathbb{C} \quad X$$

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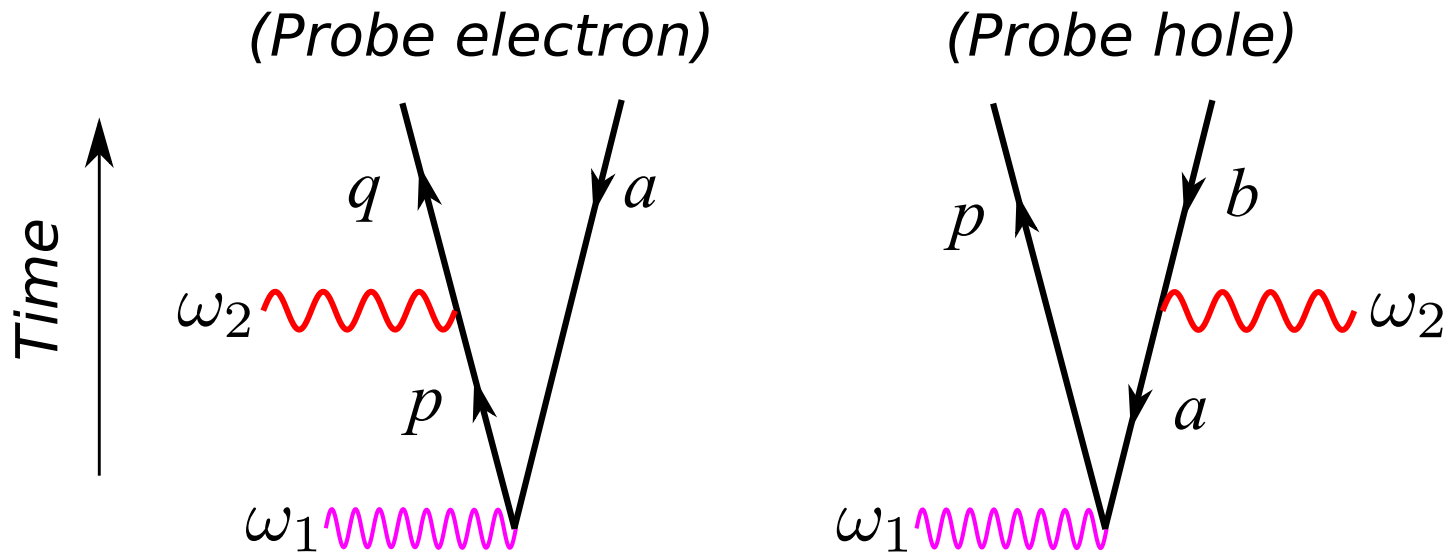

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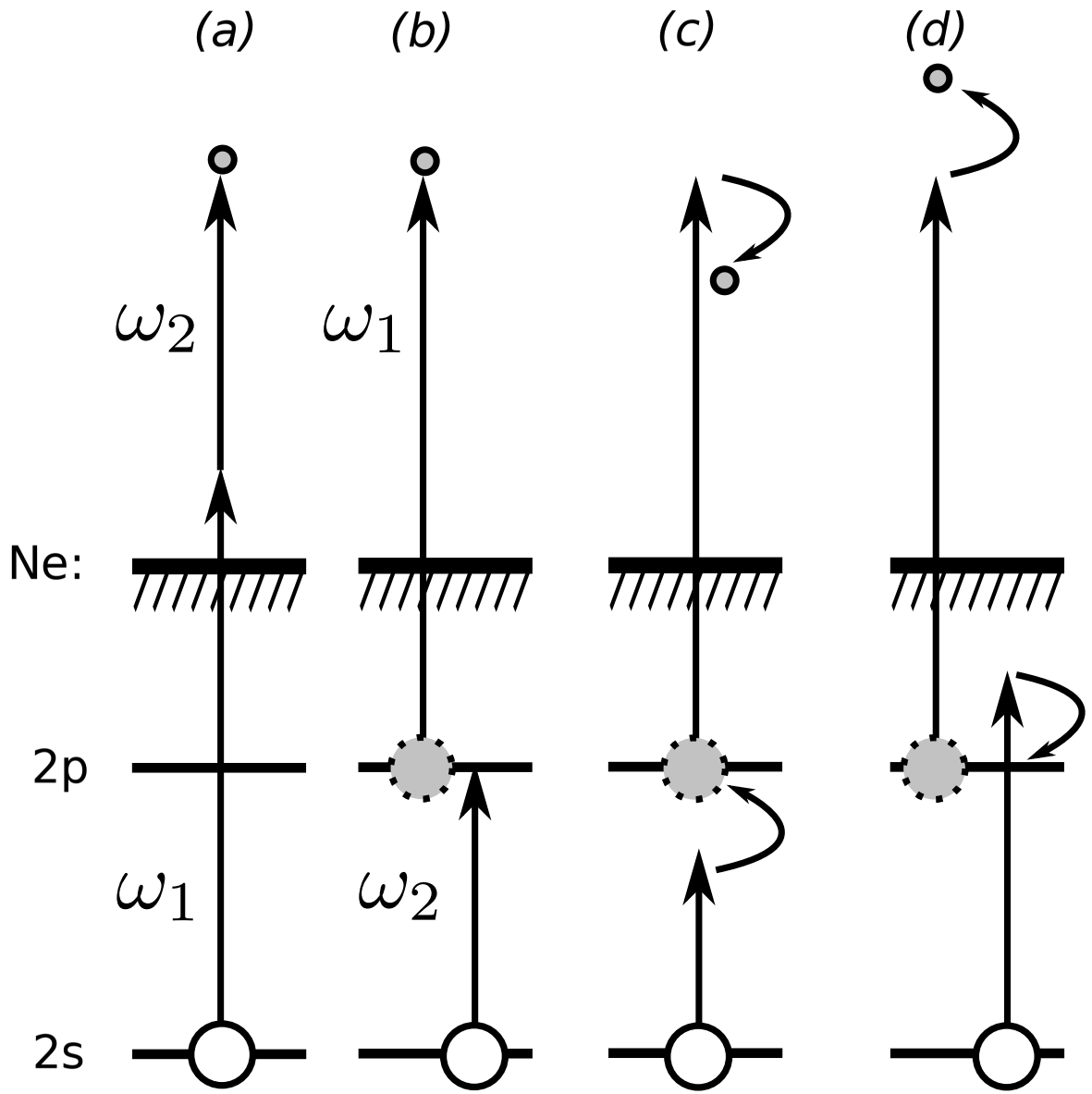
$$M_{pb}^{(h)}(\omega_1,\omega_2) = \frac{\langle\, a \mid O_2 \mid b \,\rangle\langle\, p \mid O_1 \mid a \,\rangle}{\underbrace{(\epsilon_a - \epsilon_b - \omega_2)}_{-\delta\omega}} \in \mathbb{R}, \qquad \surd$$

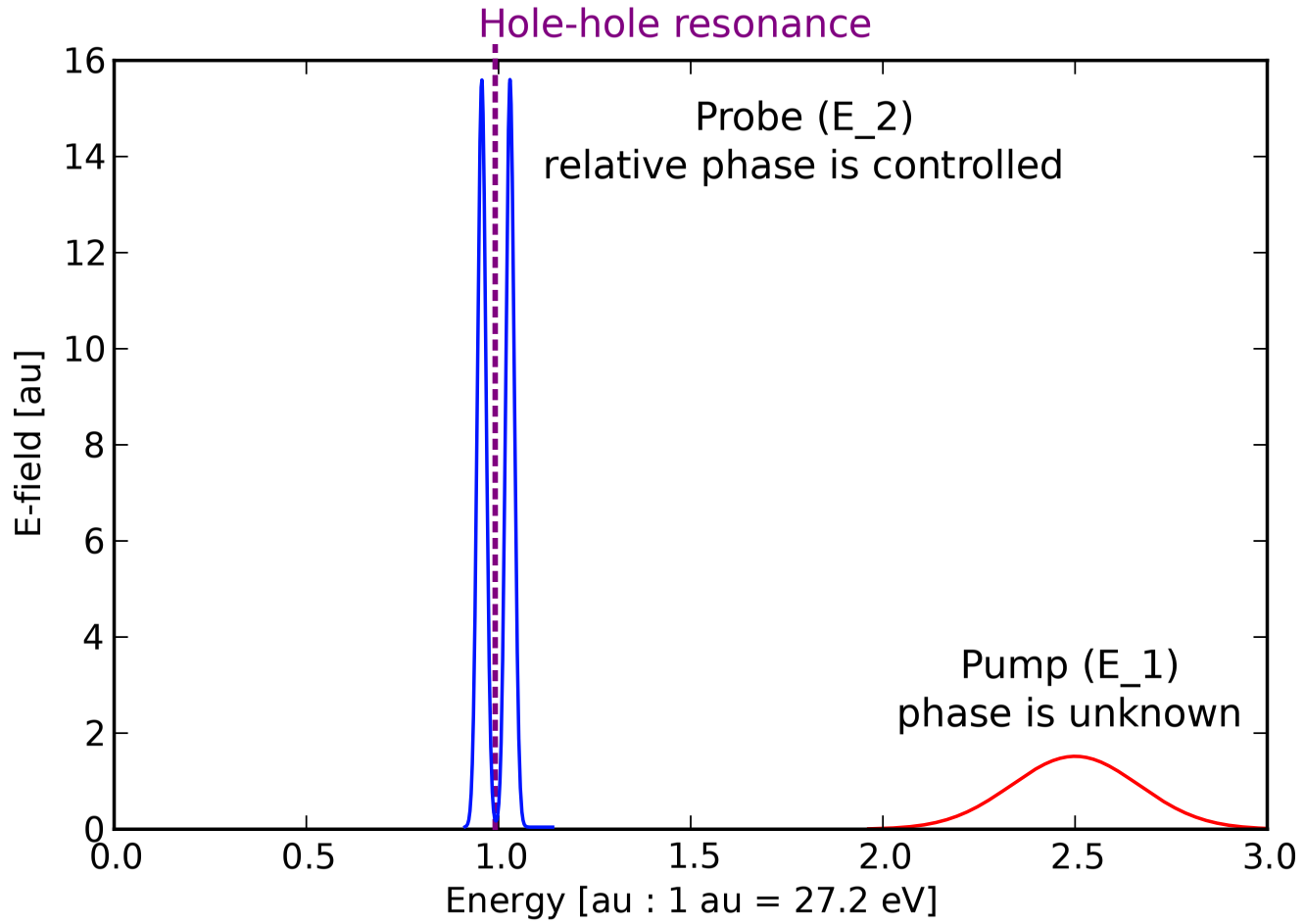
Cartoon of energy-shearing mechanism

Spectral shear of photoelectron due to energy sharing with hole



Setup of XUV fields

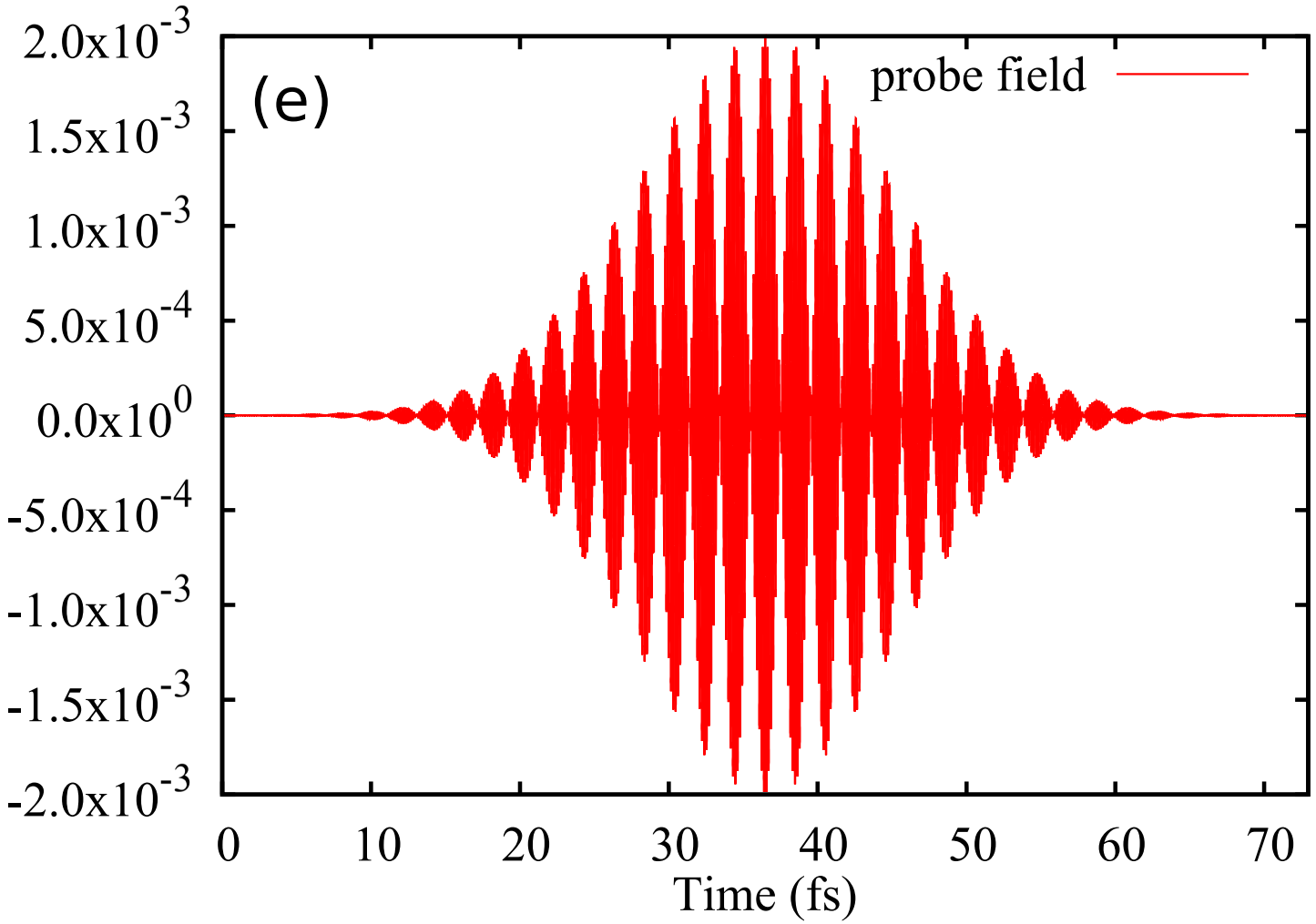
XUV field is divided in two parts as pump: E_1 and probe: E_2



- The pump E_1: broad bandwidth with unknown phase, $\phi(\omega)$.
- The probe E_2: has two components: above *and* below resonance $[\mathrm{Ne}2p^{-1} \leftrightarrow \mathrm{Ne}2s^{-1}]$ with relative phase, φ.

XUV probe field in the time-domain

Evelope-modulation on the attosecond timescale.

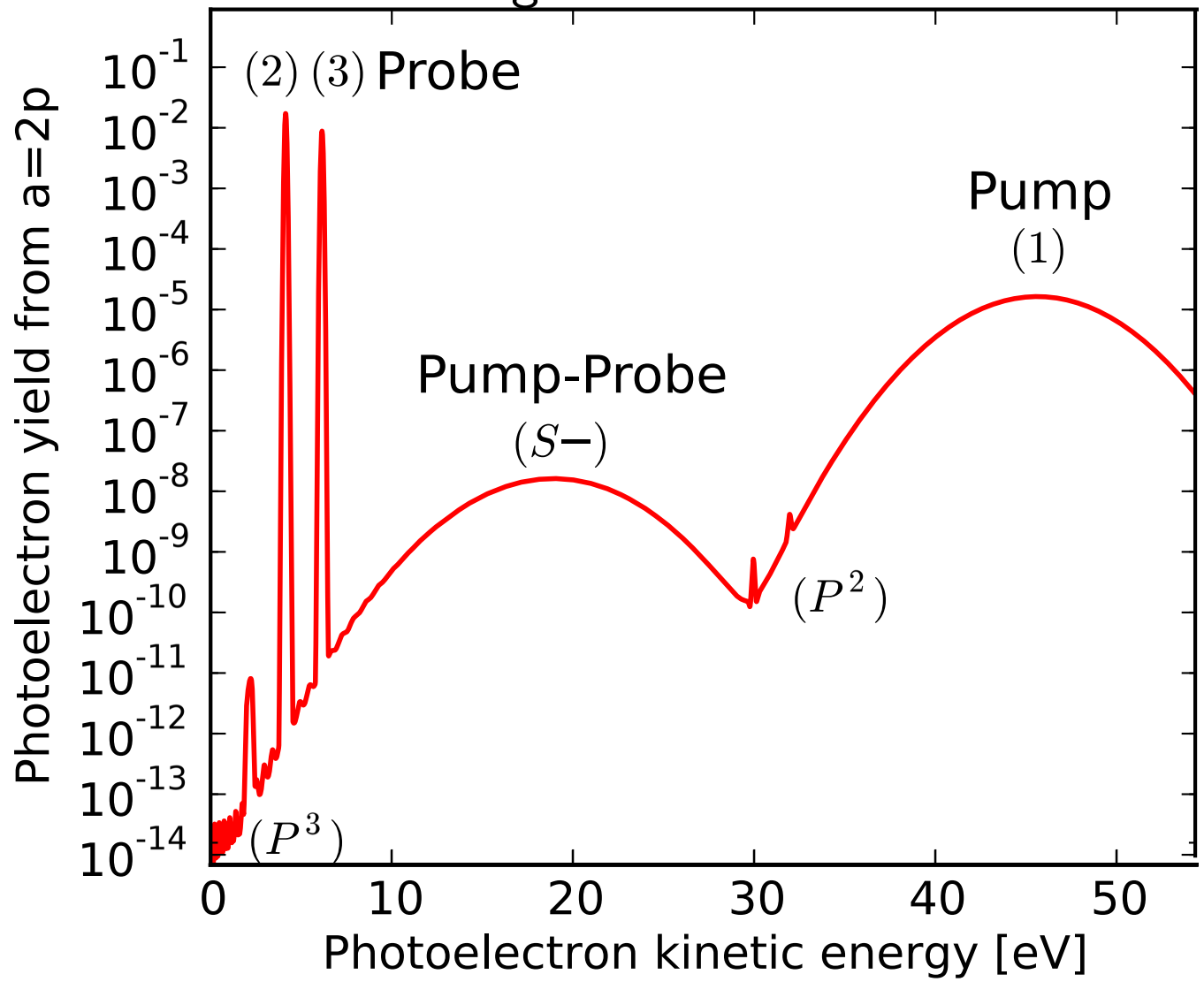


$$E_2(t) + E_3(t) \sim \cos[\delta\omega t - \tfrac{1}{2}(\phi_3 - \phi_2)] \cos[\omega_{ab} t - \tfrac{1}{2}(\phi_3 + \phi_2)]$$

Photoionization from outer orbital

Time-Dependent Configuration-Interaction Singles

TDCIS calculation using TSURFF for 1D neon model



[Calculation by *Jhih-An You* in the group of *Nina Rohringer*]

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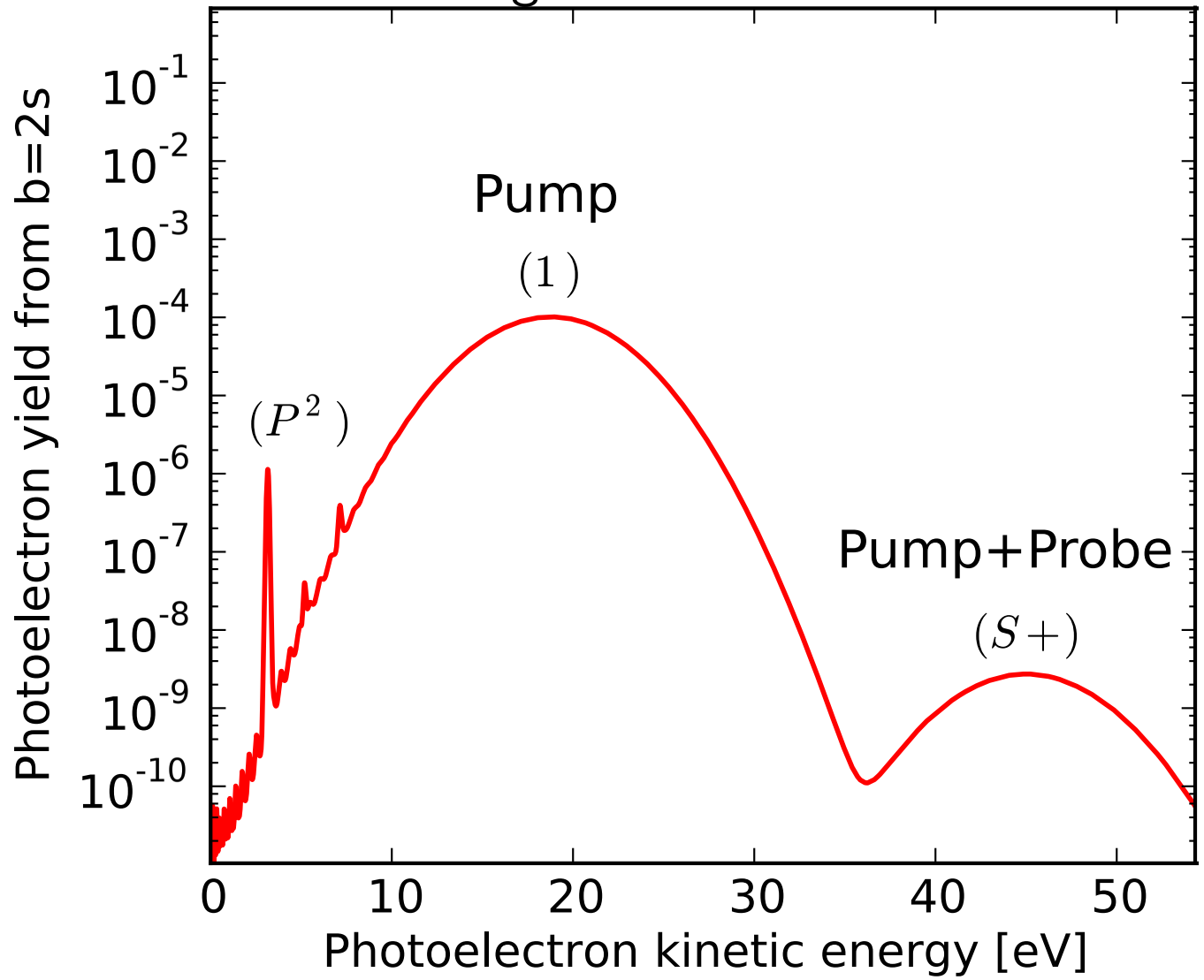
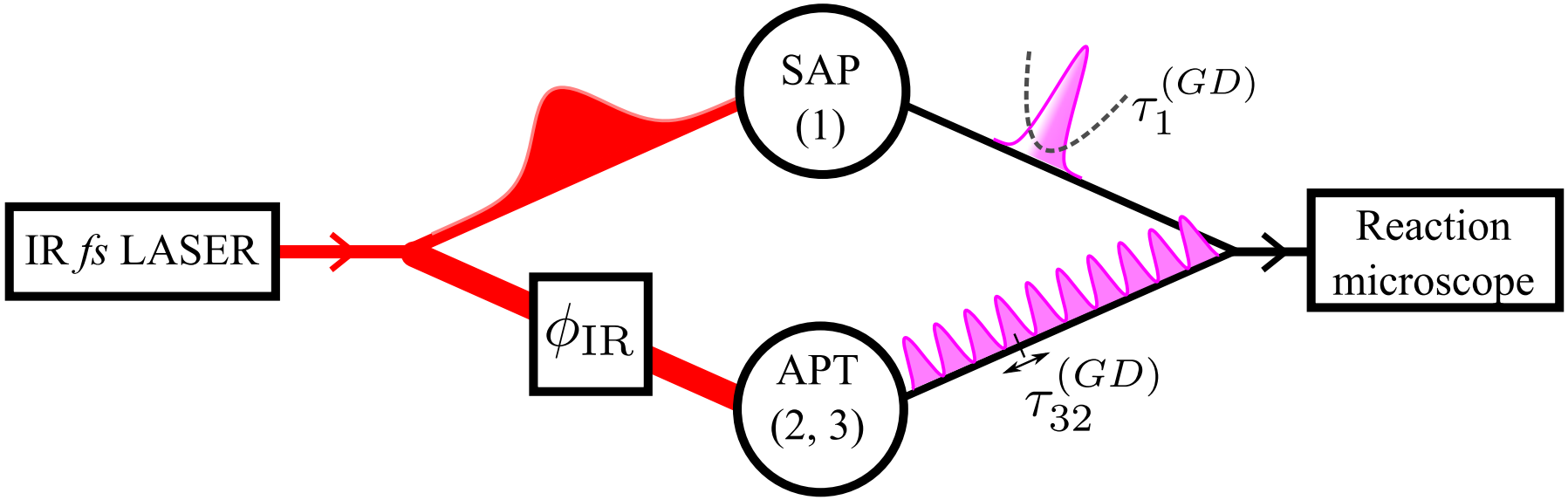


[Calculation by *Jhih-An You* in the group of *Nina Rohringer*]

Overview of proposal

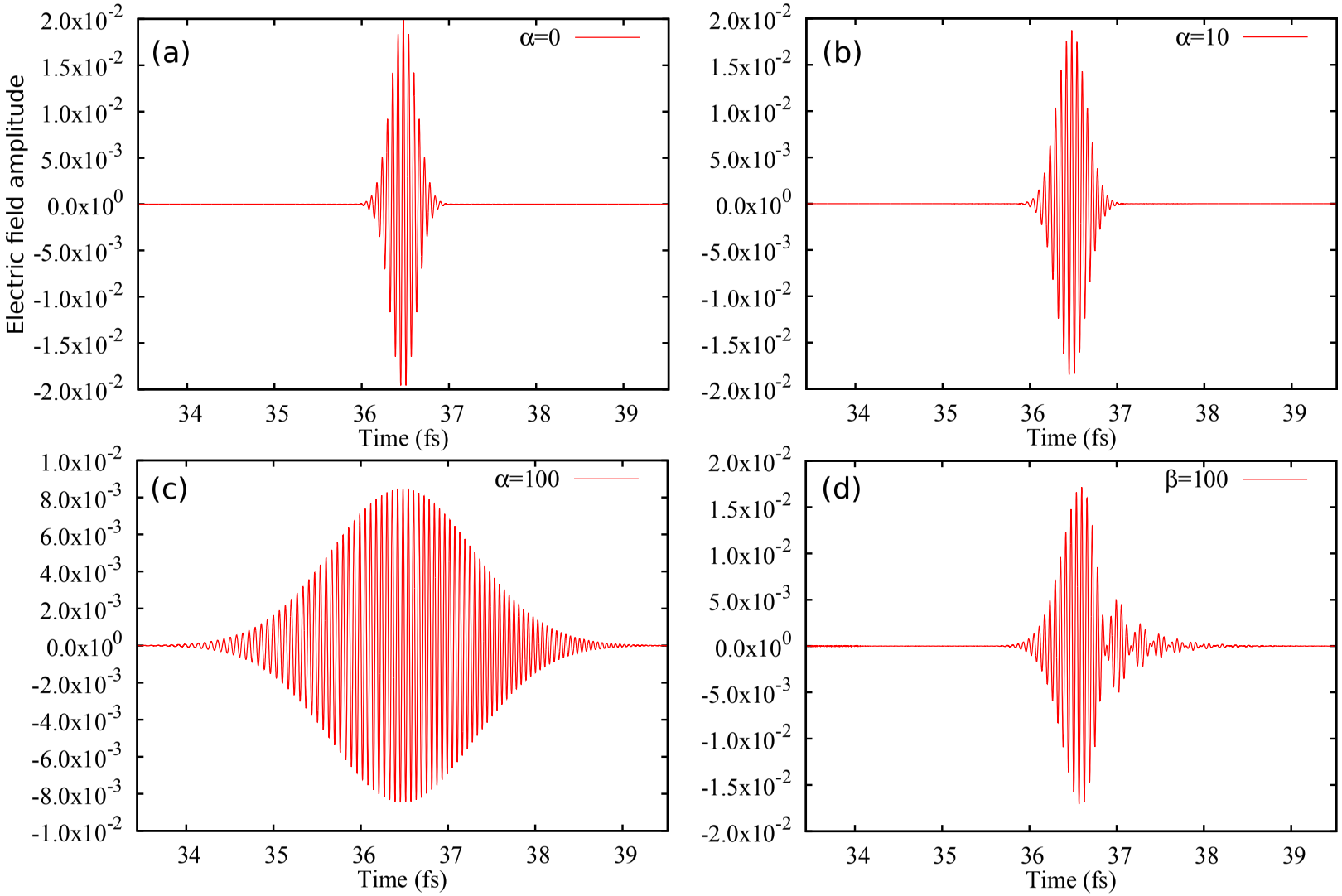
Coherent control of XUV pump and probe fields



Isolated attosecond pulse and XUV probe fields are generated by high-order harmonic generation (HHG).
IR phase, ϕ_{IR}, is used to control XUV probe field.

Isolated attosecond pulses in the time-domain

The spectral phase changes the temporal structure of the pulses.



$$\arg[E_1(\omega)] = \phi_1(\omega) = \alpha(\omega - \omega_1)^2 + \beta(\omega - \omega_1)^3$$

Photoionization from inner orbital: (S+) peak

Time-Dependent Configuration-Interaction Singles

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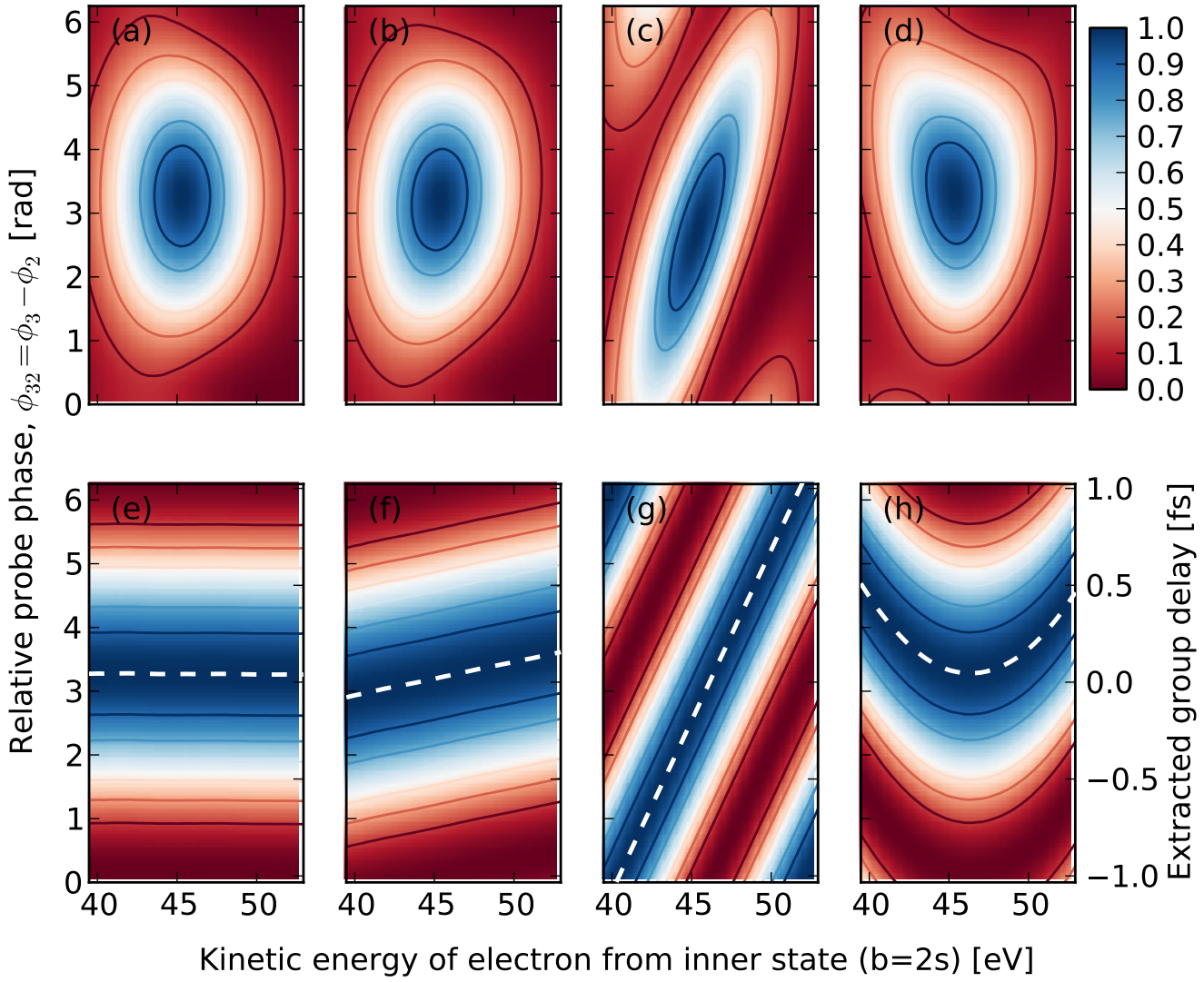
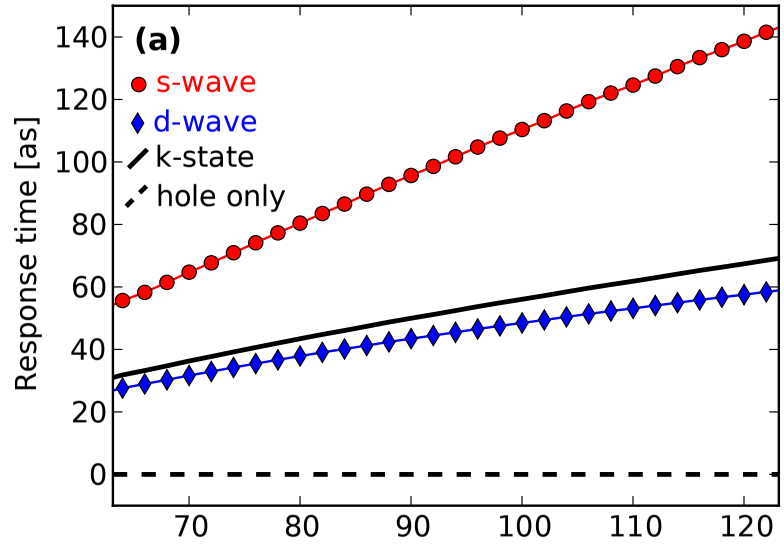
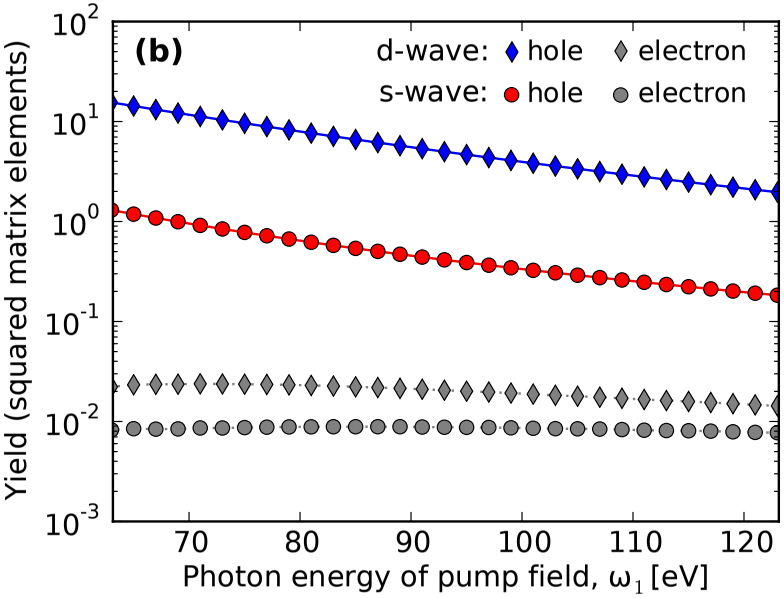
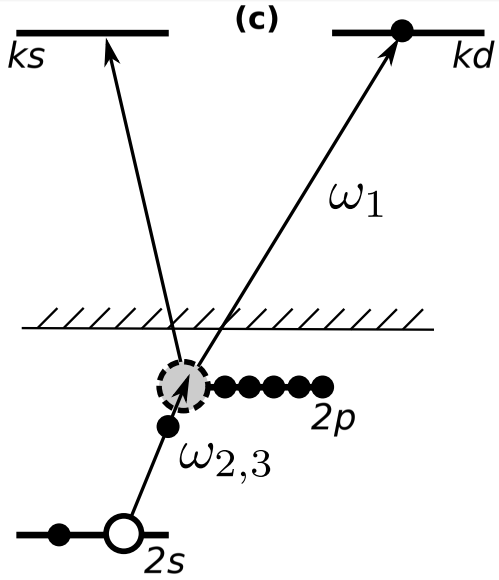


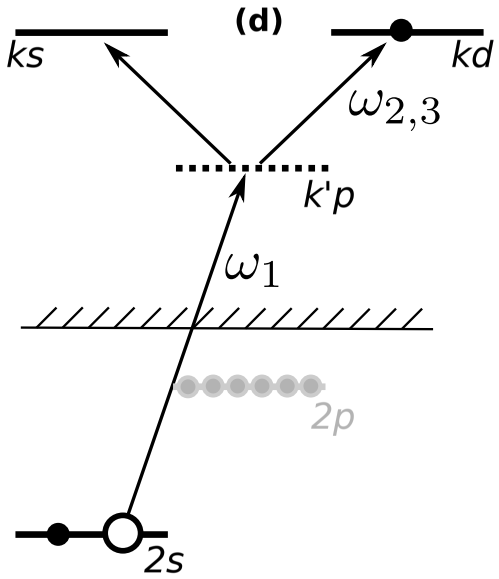
Results for independent-particle model of neon (3D)

Response times and yields.









$$W_\gamma(\mathbf{k}) = |A_\gamma| - |B_\gamma| \cos[2\delta\omega(\tau_1^{(\mathrm{GD})} - \tau_{3,2}^{(\mathrm{GD})} - \tau_\gamma(\mathbf{k}))]$$

The complex amplitude for a transition from α to γ by first absorption of a photon from the pump field, $E_1(\omega_1)$, and then a photon from either probe field, E_f where $f = [2, 3]$, is

$$S_\gamma \approx \frac{1}{2\pi i} \sum_{f=2}^{3} E_f E_1(\omega_{pc} - \omega_f) M_{pc}(\omega_{pc} - \omega_f, \omega_f), \tag{1}$$

where the two-photon matrix element contains two terms,

$$\begin{aligned} M_{pc}(\omega_1, \omega_f) &= M_{pc}^{(h)}(\omega_1, \omega_f) + M_{pc}^{(e)}(\omega_1, \omega_f) \\ &= \left[\sum_{b'} \frac{z_{b'c} z_{pb'}}{\omega_f - \omega_{b'c}} - \lim_{\xi \to 0^+} \sum_{p'} \frac{z_{pp'} z_{p'c}}{\omega_f - \omega_{pp'} - i\xi} \right], \end{aligned} \tag{2}$$

corresponding to stimulated hole (h) and electron dynamics (e), respectively. Total energy conservation requires that $\omega_1 + \omega_f = \epsilon_p - \epsilon_c$. In writing Eq. (2) we have used second quantization to approximate the N-body matrix elements by single-particle transitions,

$$Z_{\beta'\alpha} \approx z_{p'b'}, \quad Z_{\gamma\beta'} \approx -z_{b'c}\delta_{p,p'} + z_{pp'}\delta_{c,b'}.$$

Conclusion and Outlook

- Attosecond metrology using electron-laser interactions suffers from phase-lag and measurement-induced delays.
- Accurate calculation of these atomic delays is a big challenge [watch out for "TDLDA" (=linear response of LB94)*].
- New idea: Photoelectron shearing using stimulation of detuned hole-transition.
- The response time arises from the ratio of stimulated electron and hole dynamics by XUV fields.

Acknowledgement:
Eva Lindroth, Jhih-An You and Nina Rohringer.

* [M. Magrakvelidze, M. E. Madjet, G. Dixit, M. Ivanov, Himadri S. Chakraborty: arXiv:1505.01058]

Thank you for your attention!