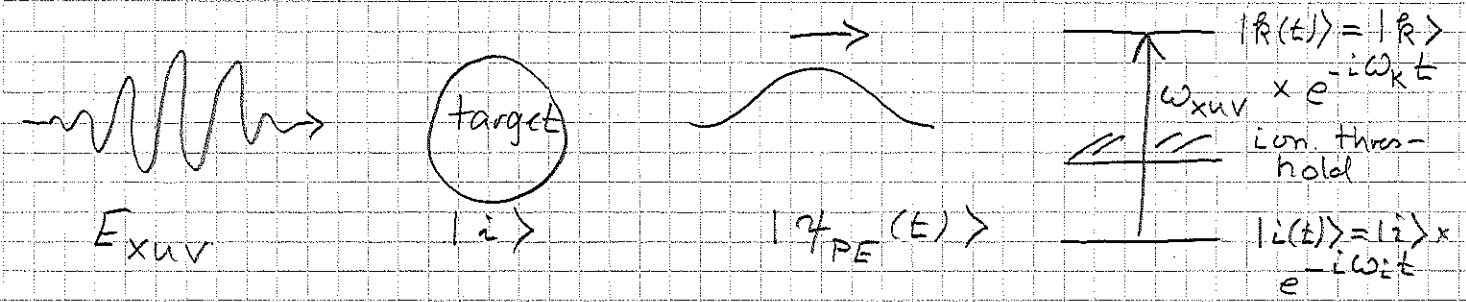
2. Time-resolved electron emission

C. Streaking spectroscopy

C-1 Photoemission phase shifts and time delays



Basic scenario of photoionization by an electromagnetic pulse from an initial state $|i\rangle$, generating a photoelectron wavepacket $|\Psi_{PE}(t)\rangle$.

- Initial / final state:

$$H_{tar}|i\rangle = \omega_i |i\rangle \quad ; \quad H_{tar}|k\rangle = \omega_k |k\rangle$$

Neglect all bound target states, except $|i\rangle$:

$$|\Psi(t)\rangle = g(t)\,|i(t)\rangle + \int dk\; a_k(t)\,|k(t)\rangle$$

initial conditions: $g(-\infty) = 1$; $a_k(-\infty) = 0$

- Solve TDSE for "weak" external field (single-photon ionization):

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle = \left(H_{tar} + V_{xuv}(t)\right)|\Psi(t)\rangle$$

$\langle k'|V_{xuv}|k\rangle \approx 0$ (no cont.-cont. coupling in "weak" fields)

$\Rightarrow$ (1. order TD perturbation theory)

$$g(t) = 1 \quad \text{(no initial-state depletion)}$$

$$a_k(t) = -i\int_{-\infty}^{t} dt' \langle k(t')|V_{xuv}(t')|i(t')\rangle$$

$\downarrow$ lin. polarization

$-z\,E_{xuv}(t)$ (length form)

For sufficiently long times, the photoelectron wave packet no longer overlaps with the target

$$|\psi_{PE}(t)\rangle \approx \int dk\; a_k(t)\,|k(t)\rangle$$

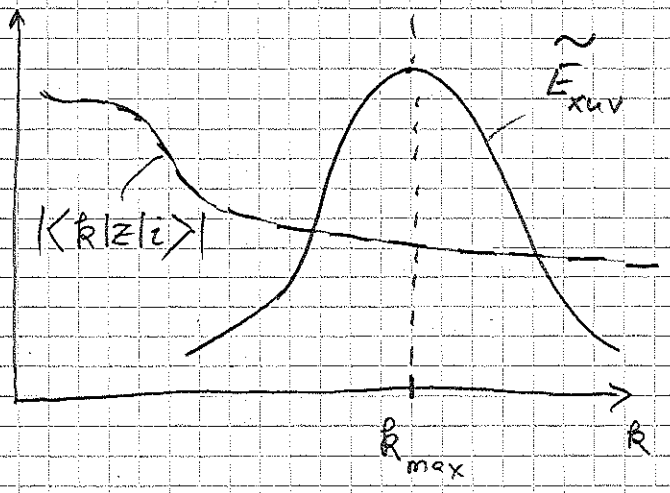
$$= i\int dk\,|k\rangle e^{-i\omega_k t}\langle k|z|i\rangle \int dt' \times \underbrace{(2\pi)^{-1/2}\int d\omega\,\tilde{E}_{XUV}(\omega)e^{-i\omega t'}}_{=E_{XUV}(t')} e^{\underbrace{i(\omega_k-\omega_i)t'}_{=:\omega_{ki}}}$$

$$\sim \sqrt{2\pi}\,i\int dk\,|k\rangle\langle k|z|i\rangle\,\tilde{E}_{XUV}(\omega_{ki})e^{-i\omega_k t}$$

- the (XUV) pulse spectral profile gets imprinted on the photoelectron wave packet
- photoelectron dispersion ω_k matters (for free electron only: $\omega_k = \frac{1}{2}k^2$)

Assume:

- $\tilde{E}_{XUV}(\omega_{ki})$ centered at k_{max}
- $|\langle k|z|i\rangle|$ monotoneously decreasing (for "large" k)



- dominant spectral component of $|\psi_{PE}\rangle$ slightly red-shifted from k_{max}.

In position representation:

$$\psi_{PE}(z,t) = \langle z|\psi_{PE}(t)\rangle$$

$$\sim \int dR \, |\langle R|z|i\rangle| \, \tilde{E}_{xuv}(\omega_{Ri}) \, e^{i \arg\langle R|z|i\rangle} \times \underbrace{\langle z|R\rangle}_{\sim e^{iRz}} e^{-i\omega_R t}$$

(PE assumed asymptotically free)

Taylor expand dipole phase about R_{max}:

$$\arg\langle R|z|i\rangle = \arg\langle R_{max}|z|i\rangle + \underbrace{\frac{d}{dR}\arg\langle R|z|i\rangle\Big|_{R_{max}}}_{=:\bar{z}} (R-R_{max}) + \dots$$

$$\Rightarrow |\psi_{PE}(z,t)|^2 \sim \left| \int dR \, |\langle R|z|i\rangle \tilde{E}_{xuv}(\omega_{Ri}) \times e^{iR(z-\bar{z}) - i\omega_R t} \right|^2$$

- Interpretation:

$\bar{z}$: PE wavepacket displacement from hypothetical reference wavepacket with $\bar{z}_{ref} = 0$

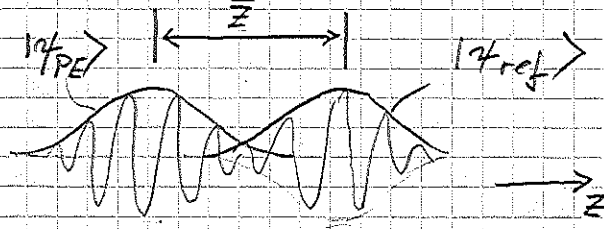
$\bar{t} = \bar{z}/v_g$: time needed to catch up with reference wavepacket

$v_g = \left.\frac{d\omega_R}{dR}\right|_{R_{max}}$: group velocity

For $\omega_R = \frac{R^2}{2}$:

$v_g = R_{max}$

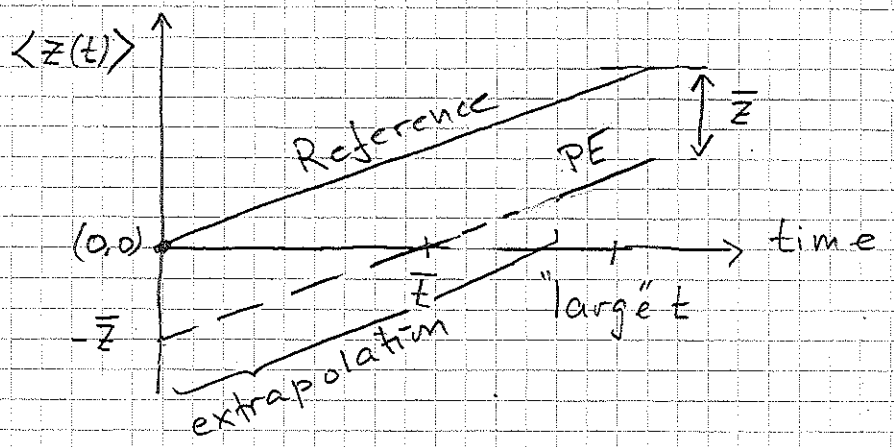
$\bar{t} = -\frac{d}{d\omega}\arg\langle R|z|i\rangle\Big|_{R_{max}}$



$\langle z\rangle = \lim_{\text{large } t} \langle z\rangle_{ref} - \bar{z}$: $\bar{t}$ as delayed start time relative to reference wavepacket

$\downarrow$

$\langle \psi_{PE}(t)|z|\psi_{PE}(t)\rangle$

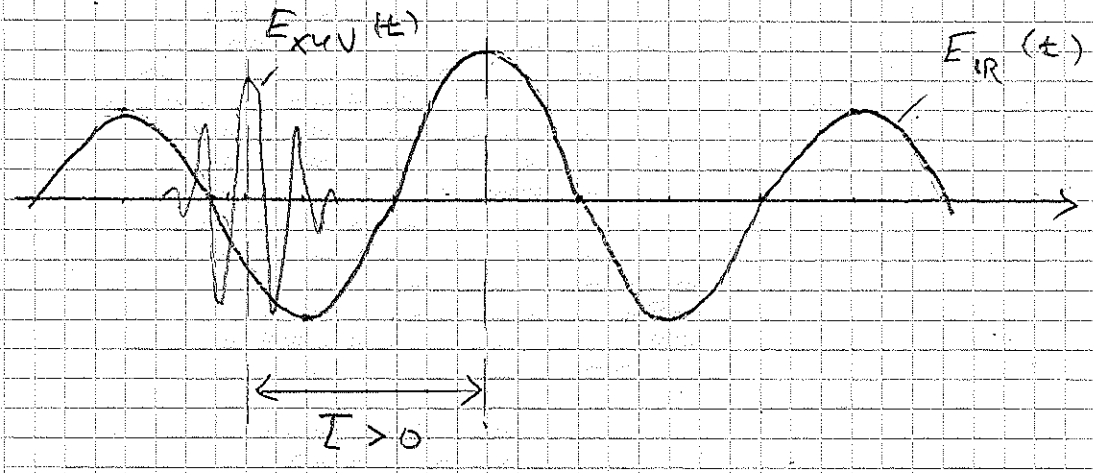


cf. U. Thumm, ..., R. Kienberger, "Attosecond physics: streaking spectroscopy", in Foundamentals of photonics & physics, D. L. Andrew (ed.), Chapter XIII (Wiley & Sons, New York 2015).

Streaking: PE energy → time conversion

Record XUV photoionization yield by adding a delayed IR probe pulse

τ : XUV − IR pulse delay



For sufficiently low IR intensity: target disturbance by probe pulse can be neglected

Classical analysis:

- assumed PE is instantaneously "born" into IR field at time τ

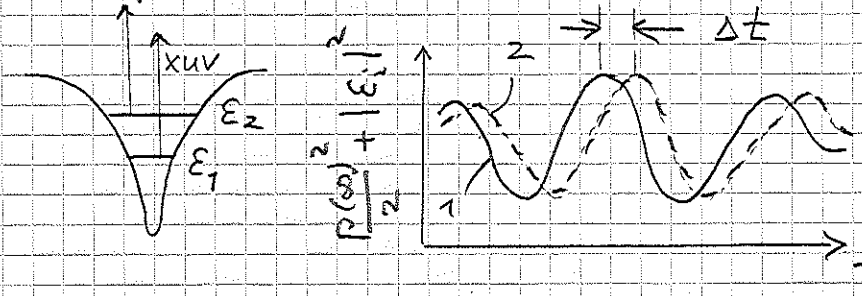
$$\frac{dp}{dt} = -E_{IR} \Rightarrow p(t) - p(\tau) = -\int_{\tau}^{t} dt' E_{IR}(t')$$

$$\Rightarrow \quad p(\infty) = p(I) - \underbrace{\int_{I}^{\infty} dt' \, E_{IR}(t')}_{A_{IR}(I)}$$

The detected PE momentum oscillates with A_{IR}, i.e., 180° out of phase with the streaking IR field.

In practice, relative streaking delays Δt_x are measured by XUV emission from

- different state of the same target



- emission from different targets

Example: IR streaked photoemission from Ne(2s) & Ne(2p): $\Delta t_{2p-2s} = (21 \pm 5)$ as
↓ hand out — Schultze et al., Science 328, 1658 (2010)

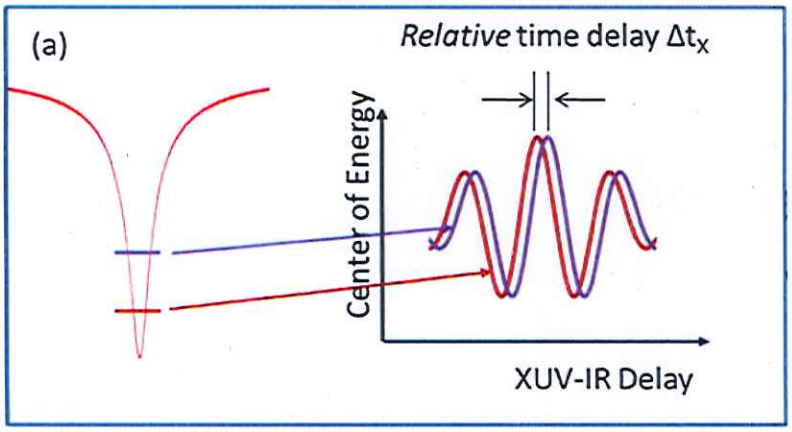
Initial-state distortion

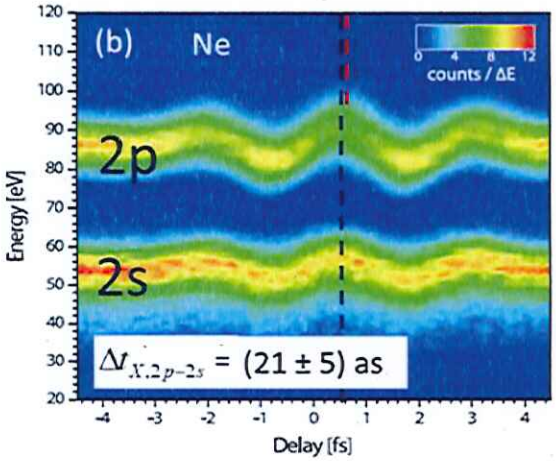
The streaking amplitude of the detected PE momentum is proportional to the IR electric field amplitude. To allow for acceptable time resolution in Δt, the IR intensity cannot be made too small and may affect the target electronic structure.

$$E_{IR}(t) = \bar{E}_{IR,0}(t) \cos(\omega_{IR} t + \phi_{CEP})$$

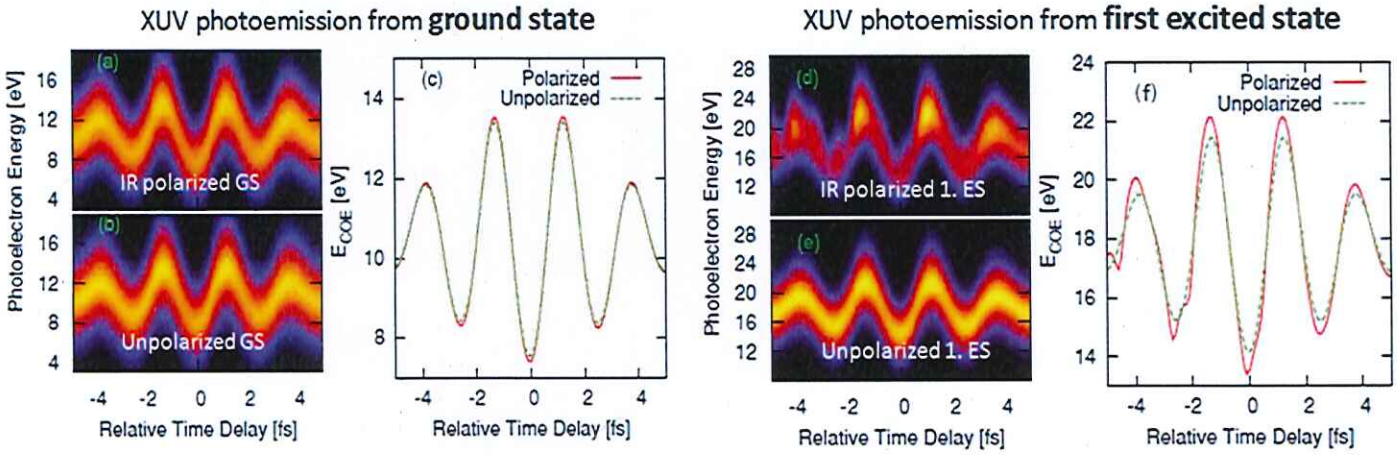
This distortion can be viewed as target polarization by the IR pump laser, which

IR streaked XUV photoemission from gaseous atoms





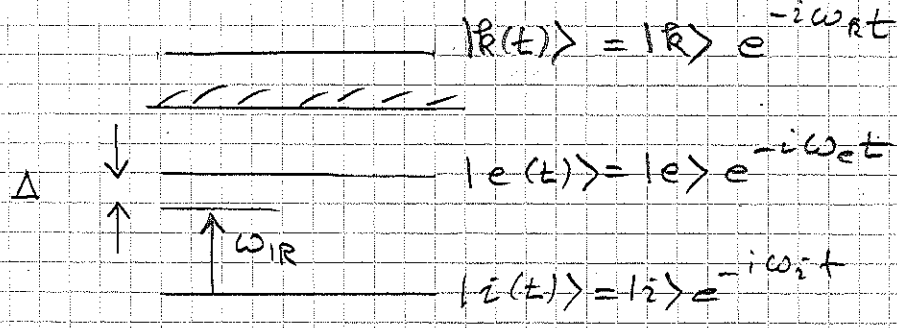
(a) Relative photoemission time delay for photoionization from two energetically separate atomic levels, yielding energetically separated streaking traces in the photoelectron streaking spectrum. The relative delay is obtained from the phase difference between the centers of energy of the two traces. **(b)** Measured photoemission streaking spectrum for XUV photoionization of Ne. The relative photoemission time delay for emission out of 2s and 2p levels is determined as (21 ±5) as. From Schultze *et al.,* Science **328**,1658 (2010).



Initial-state-perturbation effect ("polarization") by the IR streaking field on XUV photoemission from the ground state of a 1D model H atom for 25 eV, 300 as XUV pulses. Spectrogram **(a)** including and **(b)** without initial state polarization. **(c)** Corresponding central energies obtained as first moments in energy from the streaking traces. **(d,e,f)**: same as **(a,b,c)** for XUV photoionization from the first excited state of the model H atom. Adapted from Zhang & Thumm, Phys. Rev. A **82**, 043405 (2010).

we can examine by allowing for excitation to at least one excited state:

$$|i(t)\rangle \xrightarrow[\text{IR pulse}]{} |\Psi_i(t)\rangle = g(t)\,|i(t)\rangle + h(t)\,|e(t)\rangle$$



initial conditions:

$g(-\infty) = 1$

$h(-\infty) = 0$

$$i\frac{d}{dt}|\Psi_i(t)\rangle = \left(H_{tar} - z\,E_{IR}(t)\right)|\Psi_i(t)\rangle$$

$\Rightarrow$ (1. order TD perturb. theory)

$g(t) = 1$ (no initial state depletion accounted for)

$$h(t) = i\,E_{IR,0}\,\langle e|z|i\rangle \underbrace{\frac{\sin(\Delta t/2)}{\Delta}\,e^{i(\Delta t/2 - \phi_{CEP})}}_{=:\,b(t)}$$

compare with the PE wavepacket we obtained without allowing for initial-state distortion:

$$\langle R|z|i\rangle \longrightarrow \langle R|z|i\rangle\,(1 + B(\tau))$$

$$B(\tau) := \frac{\langle R|z|e\rangle\langle e|z|i\rangle}{\langle R|z|i\rangle}\cdot b(\tau)$$

$$\bar{t} \xrightarrow{\sim} \bar{t} - \frac{\mathrm{Re}\,B}{v_g}\frac{d}{dR}\arg\left\{\langle R|z|e\rangle\langle R|z|i\rangle\right\}\Big|_{R_{max}}$$

(neglecting distortion of v_g & R_{max})

Example ↓ handout: One-dim. model calculation for soft-core atom ("1D H atom"): $V_{tar} = \frac{-1}{(z^2+a^2)^{1/2}}$

Zhang & Thumm, Phys. Rev. A 82, 043405 (2010).

Final-state interactions

Deriving photoemission time delays analytically on p. 33, we assumed the PE to be free.

We now relax this assumption by allowing the PE to interact with the streaking field. In this

i) "strong-field approximation (SFA)" the PE is described by an analytical solution of the TDSE as a "Volkov state" (as we already discussed in Sec. A.3)

$$|\vec{k}(t)\rangle^{\text{Volkov}} = |\vec{k} + \vec{A}_{IR}(t)\rangle \, e^{i\phi^{\text{Volkov}}(t)}$$

$$\phi^{\text{Volkov}}(t) = -\frac{1}{2}\int_t^\infty dt' \left\{ \vec{k}^2 + 2\vec{k}\vec{A}_{IR}(t') + \vec{A}_{IR}(t')^2 \right\}$$

- $2\vec{k}\vec{A}_{IR}(t')$: PE energy oscillation in streaked spectra
- $\vec{A}_{IR}(t')^2$: Can be neglected for typical streaking intensities

We thus replace $|\vec{k}(t)\rangle$ on p. 33 by $|\vec{k}(t)\rangle^{\text{Volkov}}$:

$$\arg\langle \vec{k}|z|i\rangle \xrightarrow[\text{IR pulse}]{} \arg\langle \vec{k} + \vec{A}_{IR}(\tau)|z|i\rangle$$

$$= -\{\bar{z} + \Delta z_{IR}(\tau)\}k + \mathcal{O}\left(A_{IR,0}^2; |k - k_{max}|^2\right)$$

↑ expand in $A_{IR,0}$ & $|k - k_{max}|$

where

$$\Delta z_{IR} = \frac{d^2}{dk^2} \arg\langle \vec{k}|z|i\rangle \Big|_{k_{max}} \vec{A}_{IR}(\tau)$$

(displacement by A_{IR}, in addition to field-free displacement $\bar{z}$)

$$\bar{t} \longrightarrow \frac{\bar{z} + \Delta z_{IR}(\tau)}{v_g + A_{IR}(\tau)} = \bar{t} + \Delta t_{IR}(\tau) + \mathcal{O}\left(A_{IR,0}^2\right)$$

where

$$\Delta t_{IR}(\tau) = \frac{\bar{z}}{v_g} - \bar{t} - \frac{A_{IR}(\tau)}{v_g}$$

oscillates in τ. For sufficiently long IR pulses, these oscillations vanish and

$$\Delta t_{IR}(\tau) \xrightarrow[\text{pulse}]{\text{long IR}} \bar{t}$$

While the streaking delay is not changed by long pulses, the shape of the streaking traces in PE spectra remains distorted.

ii) Coulomb-laser interactions

To further improve the modeling of the final PE state, we allow for PE interaction with both, IR laser field and the residual (assumed charged) target.

Taking the SFA final-state wave function

$$\langle x | k(t) \rangle^{\text{Volkov}} \sim e^{-i\frac{k^2}{2}t - i S^{SFA}(x,t)}$$

as a guide, we seek final states

$$\langle x | k(t) \rangle^{CL} \sim e^{-i\frac{k^2}{2}t - i S^{EA}(x,t)}$$

$$S^{EA} = S^{SFA} + S^{CL}$$

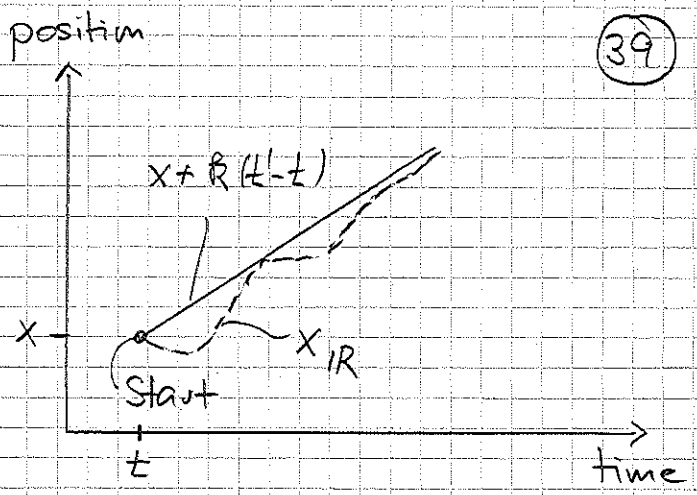
A semi-classical (eikonal) approximation allows us us to find S^{CL}:

cf. Zhang & Thumm, Phys Rev. A 82, 043405 (2010) and refs. therein.

Main ideas:

- IR field induced drift of free photo-electron

$$\Delta X(t,t') = \int_t^{t'} dt'' A_{IR}(t'')$$



$\Rightarrow$ PE trajectory

$$x_{IR}(t',t,x) = \underbrace{x + R(t'-t)}_{\text{free PE}} + \Delta X(t,t')$$

- Integrate class. action along in V_{tar} along x_{IR}:

$$S^{CL}(x,t) = \int_t^{\infty} dt' \, V_{tar}[x_{IR}(t',t,x)]$$

For typical streaking intensities ΔX is "small". Expand in orders of ΔX:

$$= S^{C}(x) - \int_t^{\infty} dt' \left\{ \underbrace{\left[-\frac{\partial}{\partial x} V_{tar}[x + R(t-t')] \right]}_{F_{tar}} \times \Delta X(t,t') + \mathcal{O}(\Delta X^2) \right\}$$

$S^{C}(x)$: IR-laser free phase accumulation in V_{tar} (e.g., Coulomb phase)

$F_{tar} \times \Delta X(t,t')$: PE interaction with the target (e.g., Coulomb scattering) while PE releases/absorbs IR photons.

$\Rightarrow$ Streaking (center of energy) oscillations relative to SFA results

$$\delta E^{EA}(t) = K \, A_{IR}(T - \delta T(x))$$

K: change in streaking amplitude (from R)

$\delta T(x)$: temporal shift due to simultaneous PE talk with target & laser.

Numerical example : Comparison of Coulomb + laser induced streaking amplitudes and photo-emission delays with SFA and numerical TDSE results.

↓

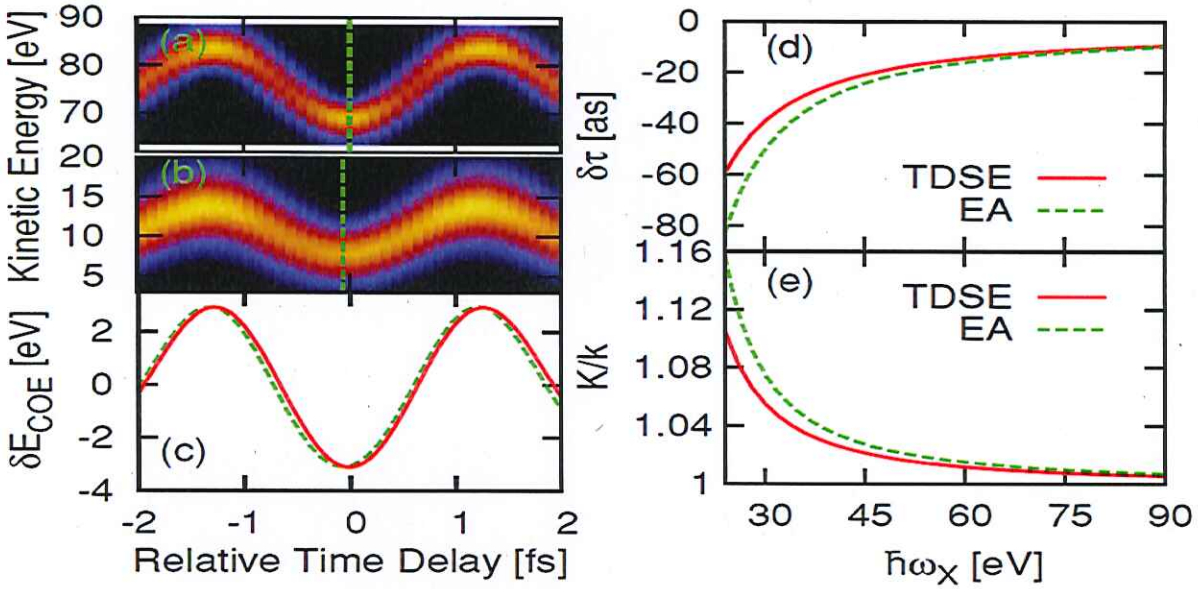
hand out

Zhang & Thumm, Phys. Rev. A 82, 043405 (2010).

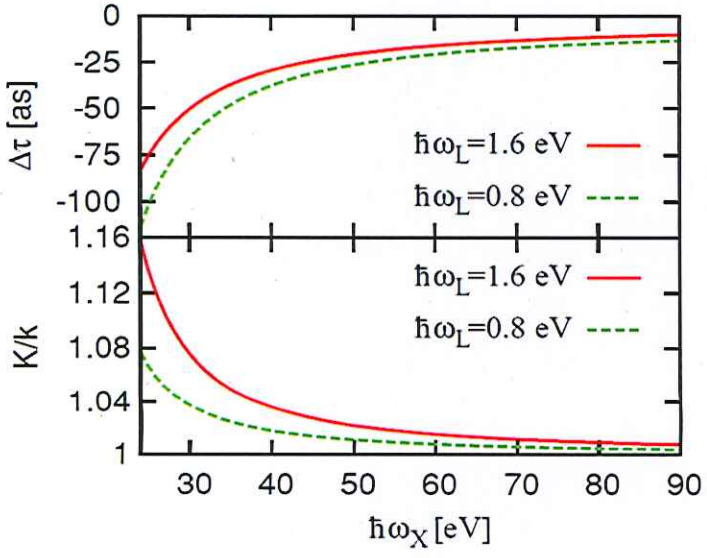
Electron-ion interaction effects in attosecond time-resolved photoelectron spectra

Zhang and Thumm, PHYSICAL REVIEW A **82**, 043405 (2010)

Photoionization by attosecond extreme ultraviolet (xuv) pulses into the laser-dressed continuum of the ionized atom is commonly described in strong-field approximation, neglecting the Coulomb interaction between the emitted photoelectron (PE) and the residual ion. By solving the time-dependent Schrödinger equation, we identify a temporal shift $\delta\tau$ in streaked PE spectra, which becomes significant at low PE energies. Within an eikonal approximation, we trace this shift to the combined action of Coulomb and laser forces on the released PE, suggesting the experimental and theoretical scrutiny of their coupling in streaked PE spectra. Further, we examined the effect of initial state polarization by the laser pulse on the xuv streaked spectrum.



Streaked photoemission from 1D model H atoms. TDSE calculations for XUV pulse central energies of (a) 90 eV and (b) 25 eV. (c) Corresponding centers of energy for 90 eV (solid line) and 25 eV (dashed line) pulses. To facilitate the identification of the relative temporal shifts $\delta\tau$, $\delta E_{COE}(\tau, \hbar\omega_X = 90\ \text{eV})$ is normalized to the $\hbar\omega_X = 25$ eV result. (d) $\delta\tau$ and (e) oscillation amplitude relative to the SFA for TDSE (solid line) and eikonal approximation (dashed line) calculations.



Comparison of eikonal temporal shift $\delta\tau$ and oscillation amplitude ratio K/k at two IR laser photon energies.