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

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

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

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

where

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

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

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

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

ii) <u>Coulomb-laser interactions</u>

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

Taking the SFA final-state wavefunction

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

as a guide, we seek final states

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

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

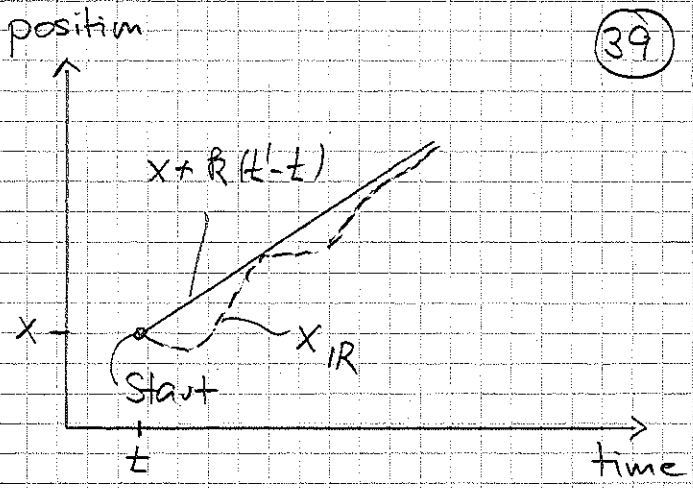
A semi-classical (eikonal) approximation allows us us to find S^{CL}:

cf. Zhang & Thumm, Phys Rev. A <u>82</u>, 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 S^{CL} in V_{tar} along x_{IR}:

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

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

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

$S^{c}(x)$: 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 in

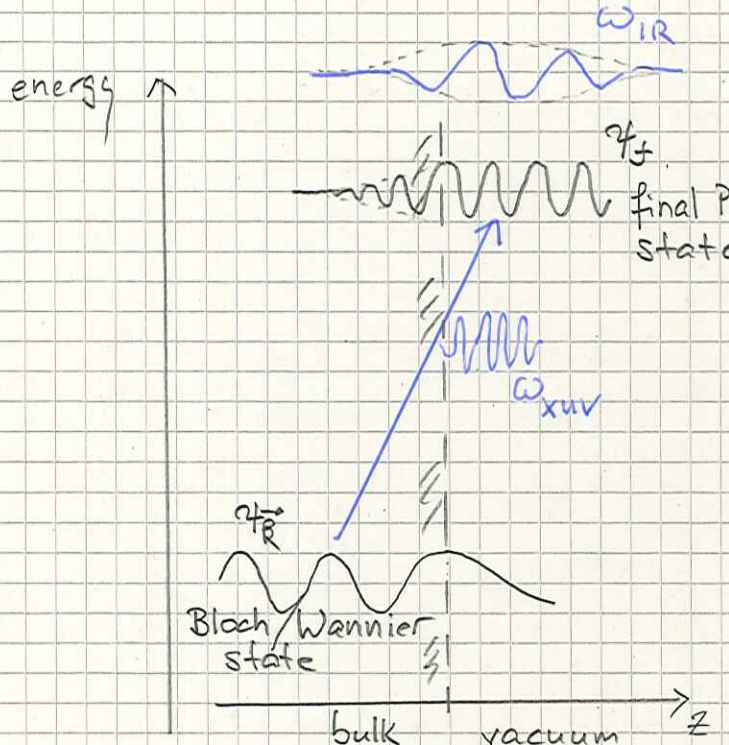
$$\delta E^{EA}(T) = K \, A_{IR}\left(T - \underbrace{\delta T(x)}_{\text{temporal shift due to simultaneous PE talk with target \& laser}}\right)$$

K: change in streaking amplitude (from R)

Numerical example : Comparison of Coulomb + laser induced streaking amplitudes and photoemission delays with SFA and numerical TDSE results.
Zhang & Thumm, Phys. Rev. A 82, 043405 (2010).

↓ hand out

Streaked photoemission from metal surfaces



recall :

$$T_{\vec{k}} \sim \int_{-\infty}^{\infty} dt \, \langle \psi_f(t) | \vec{r} \cdot \vec{A}_{XUV} | \psi_{\vec{k}} \rangle$$

emission amplitude

incoherent sum over occupied initial states → emission probability

$$P = \int d\vec{k} \, f(k, T) \, |T_{\vec{k}}|^2$$

f : distribution function (Fermi-Dirac), T : abs. temp.

(simplest) final-state model : damped Volkov state :

$$\psi_f(\vec{r}, t) \sim \exp\left\{ i \left[\vec{k}_f + \vec{A}_{IR}(t) \right] \cdot \vec{r} + i \, \phi^{Volkov}(t) \right\}$$

$$k_{f,z} = \mathrm{Re}\left(k_{f,z}\right) - i \, \underbrace{\lambda(k_f)}_{\tau} / 2$$

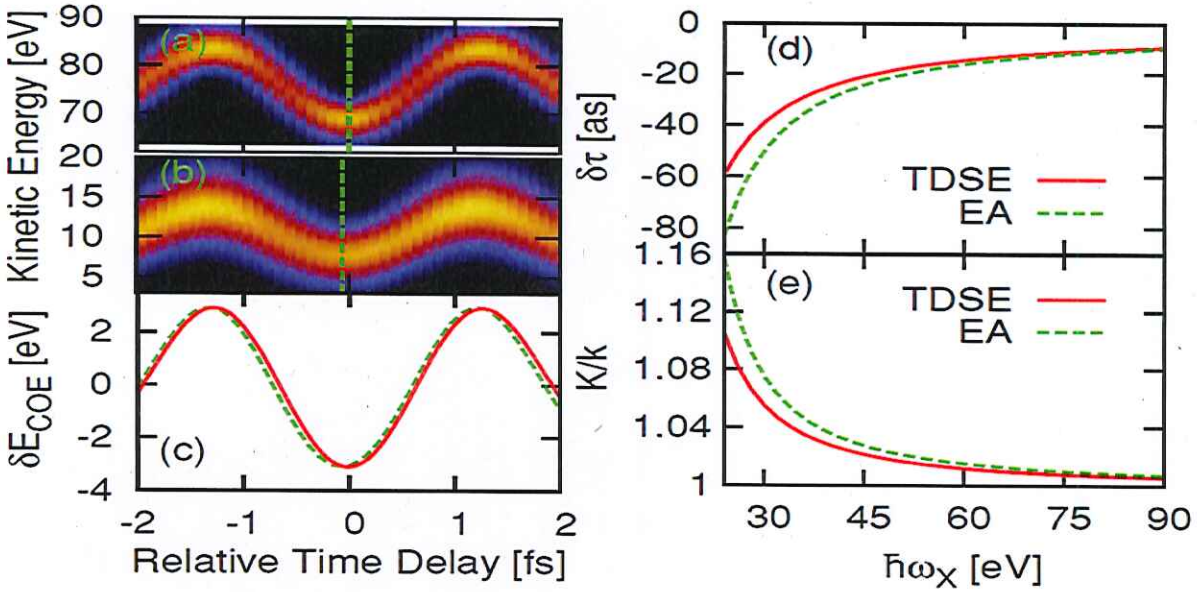
damping factor (e^- mean-free path)

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

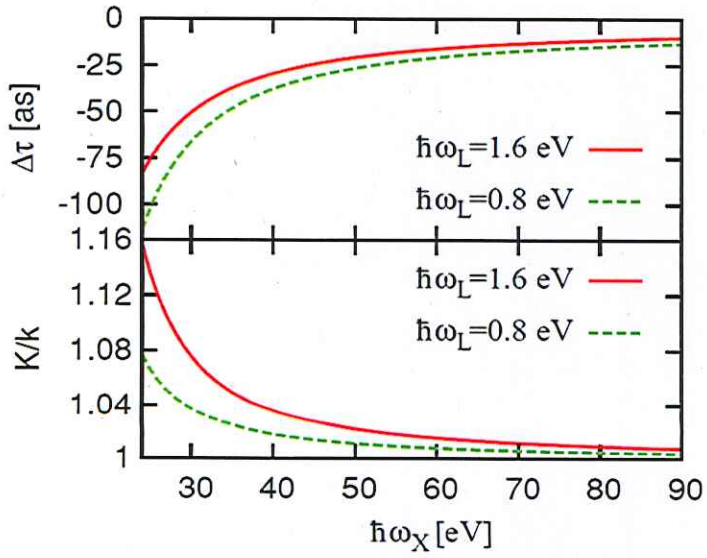
Electron-ion interaction effects in attosecond time-resolved photoelectron spectra

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.

(a simple) initial-state model :

$$\psi_{\vec{k}}(\vec{r},t) \sim \exp\{-i\varepsilon_{\vec{k}}t + i\vec{k}\cdot\vec{r}\}\, u_{\vec{k}}(\vec{r})$$

+ surface reflected wave

$$u_{\vec{k}}(\vec{r}) = u_{\vec{k}}(\vec{r} + \vec{R}_{\vec{n}})$$

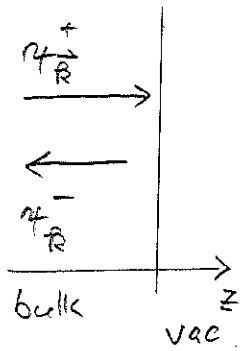
↑ crystal lattice

- can be applied to localized & delocalized initial states
- localized states : Bloch waves are superpositions of core levels that are localized at lattice sites $\{\vec{R}_n\}$ with binding energies ε_i

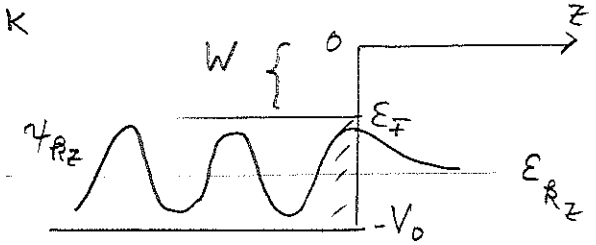
$$\psi^{\pm}_{\vec{k}}(\vec{r},t) = e^{-i\varepsilon_i t} \sum_{\vec{n}} e^{i\vec{k}\cdot\vec{R}_{\vec{n}}}\, \psi_{cl}(\vec{r}-\vec{R}_{\vec{n}})$$

$$\psi_{\vec{k}} = \psi^{+}_{\vec{k}} + \psi^{-}$$

incident surface reflected



- hierarchy of initial-state approximation :
  - Sommerfeld model : $T = 0K$ (step-up potential)



$$\psi_{\vec{k}}(\vec{r},t) = \psi_{k_z}(\vec{r})\, \exp\{i\vec{k}_{\parallel}\cdot\vec{r}_{\parallel} - i\varepsilon_k t\}$$

↑ momentum / ↑ position in surface plane

$$\varepsilon_k = \varepsilon_{k_z} + \varepsilon_{\vec{k}_{\parallel}}$$

$$\psi_{k_z}(\vec{r}) \sim e^{i k_z \cdot z} + R(k_z)\, e^{-i k_z z}$$

↑ reflection coeff : $R(V_0; k_z)$

↓

$\mathcal{E}_F + W$

↑ work function

examples : a) tungsten :

↓ handout

experiment : Cavalieri et al., Nature 449, 1029 (2007)

theory : Zhang & U.T., Phys. Rev. A 84, 063403 (2011), and refs.

4f – conduction-band delay :

$\Delta T^{exp.} = (110 \pm 70)$ as ← later improved

$\Delta T^{theory} = 110$ as for $\lambda = 5$ a.u.

b) magnesium

experiment : Neppl et al., Phys. Rev. Lett. 109, 087401 (2012)

theory : Liao & U.T. Phys. Rev. Lett. 112, 023602 (2014)

2p – valence-band delay = 0

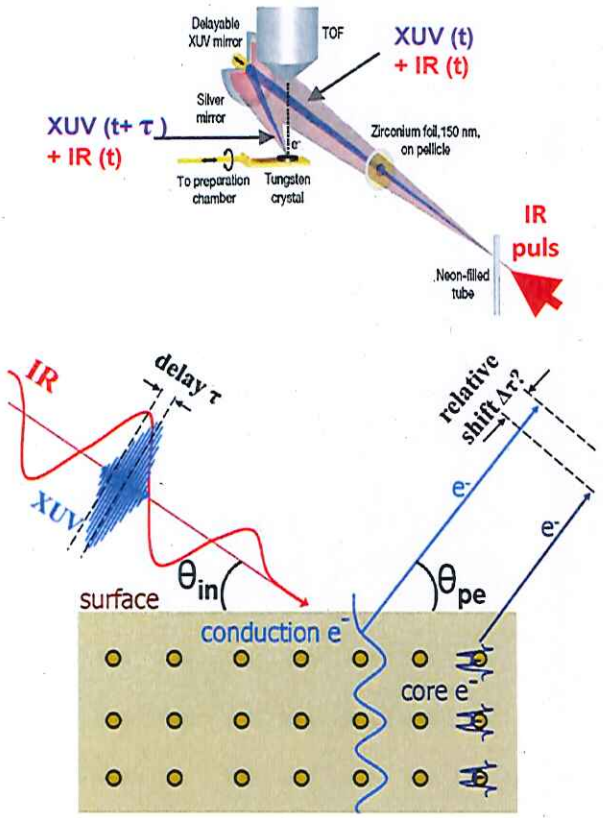
c) adsorbate-covered tungsten : Mg/W(110)

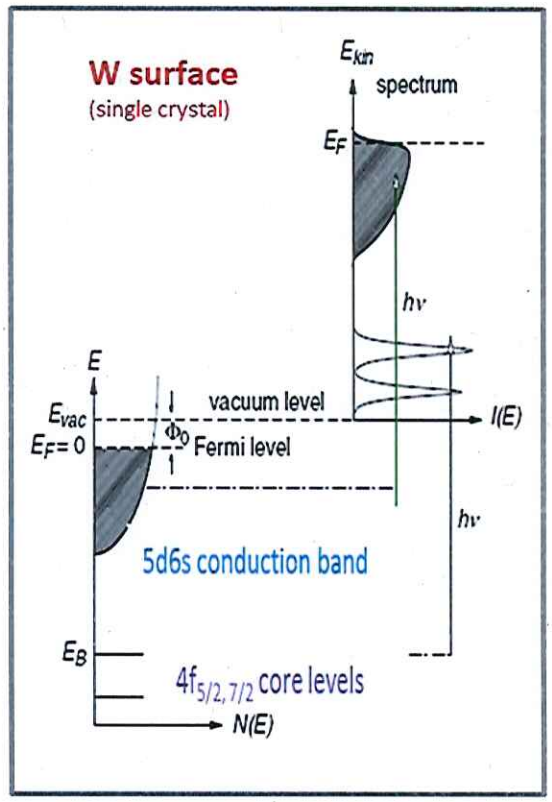
experiment : Neppl. et al., Nature 517, 342 (2015)

theory : Liao & U.T., Phys. Rev. A 92, 031401 (2015)

Time-resolved photoemission from metal surfaces

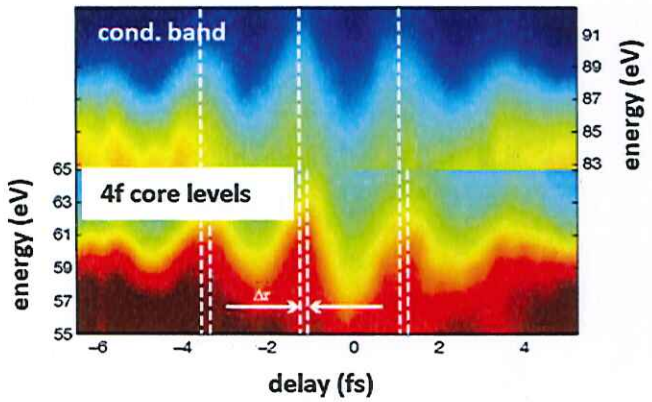
W(110): 4f core level relative to conduction-band emission





Experiment: $\Delta\tau = 110 \pm 70\,\text{as}$

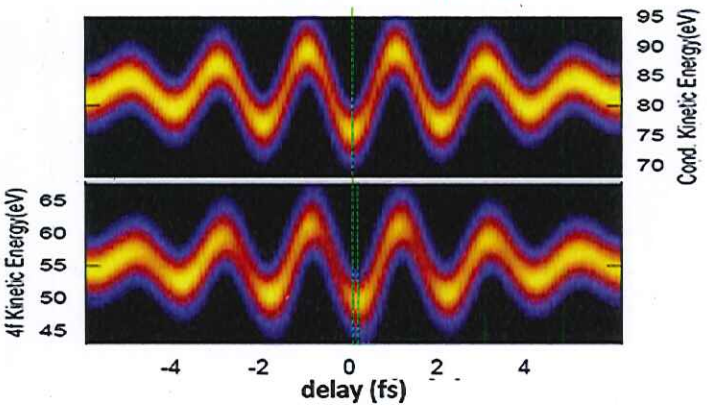
Cavalieri *et al.*, *Nature* **449**,1029(2007)



Theory: $\lambda \sim 5\,\text{a.u.} \Rightarrow \Delta\tau = 110\,\text{as}$

Zhang, U.T., *PRL* **102**, 123601 (2009)

PRA **84**, 063403 (2011)



$E_F = 4.35\,\text{eV}, \Phi = 4.5\,\text{eV}, I_p = 33.6\,\text{eV}, a = 0.31\,\text{nm}$

Mg(0001): 2p core level relative to valence-band emission

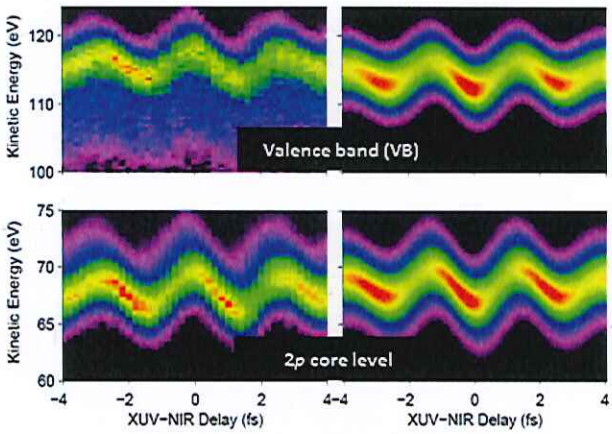
Experiment Neppl *et al.*, *PRL* **109**, 087401 (2012)

Theory Liao, U.T., *PRL* **112**, 023602 (2014)

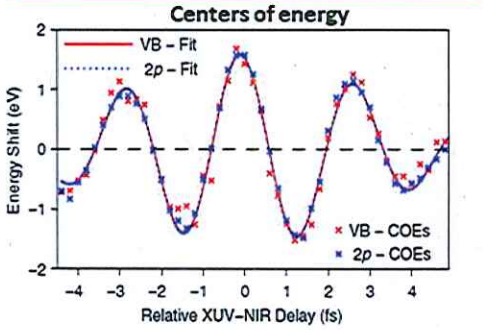
XUV: 435 as, 118 eV

NIR: 800 nm, 5 fs
CEP = 0

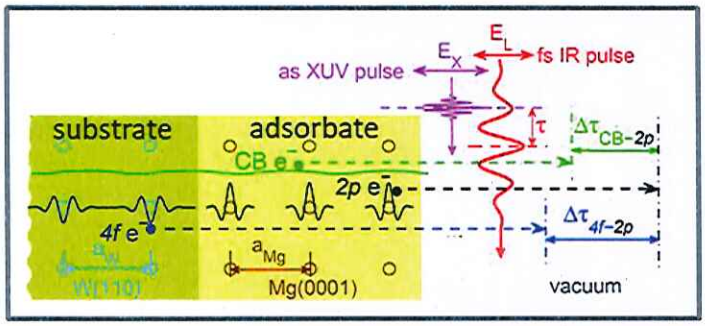
IR-skin depth = 2 Å
MFPs: 4.9 Å for VB
3.7 Å for Mg(2p)

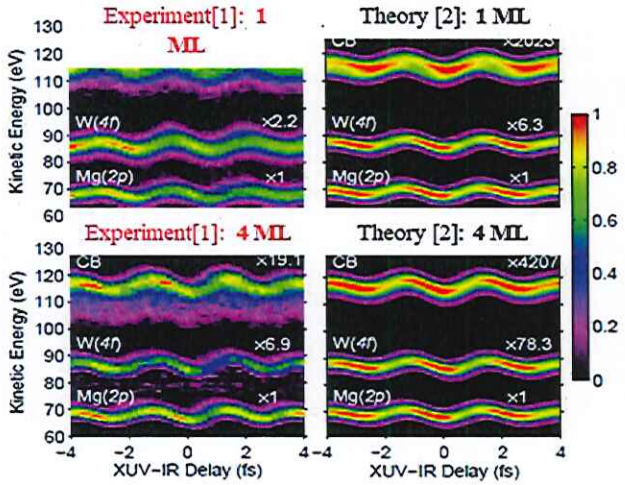


VB – 2p streaking time delay = 0

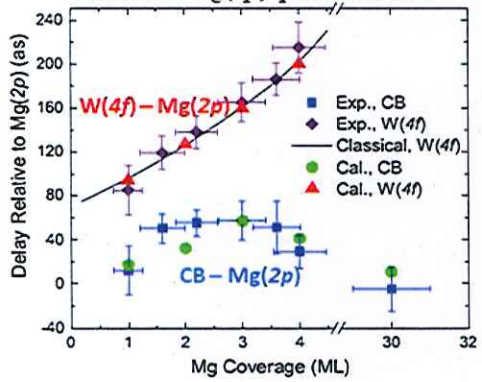


Streaked photoemission from heterogeneous structures: *Mg/W(110)*





Mg-coverage-dependent streaking delays
relative to Mg(2*p*) photoelectrons



Spectra & delays are sensitive to

- **electron dispersion in adsorbate**
- **substrate-adsorbate-interface properties**

[1] Neppl *et al.*, *Nature* **517**, 342 (2015)
[2] Liao, U.T., *PRL* **112**, 023602 (2014); *PRA* **92**, 031401(R) (2015)

layer thickness dependence of :

- W (4f) - Mg (2p) delay : increases with adsorbate thickness (as expected)

- Conduction - Mg (2p) delay : increases up to ~ 3ML (due to increasing travel time from W substrate. Decreases to 0 : fewer substrate electrons emitted; delay approaches Mg (2p - valence band delay) as # MLs ↑.

Towards the time resolution of collective excitations in solid