Simulation of Streaking Experiments at Surfaces

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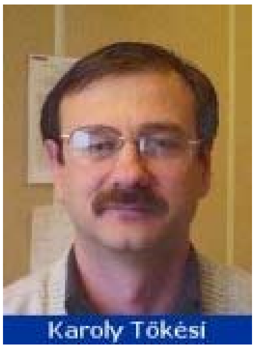
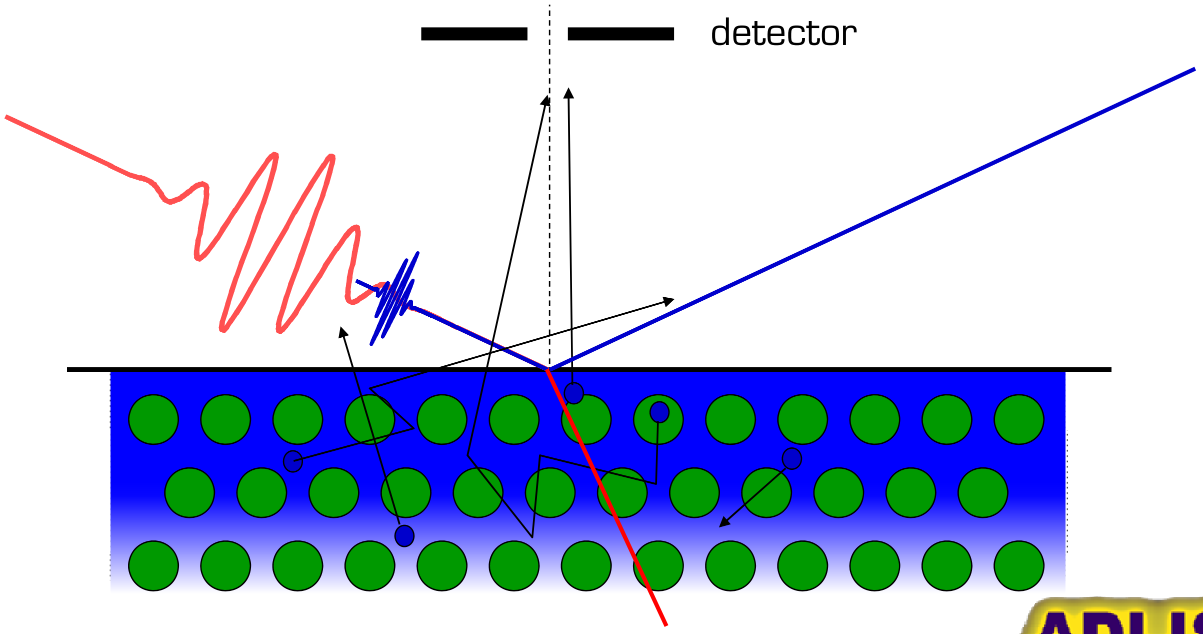

Karoly Tőkési

Simulated System



Outline

- Review of Simulation Methods
- Classical-Transport Model
  - Electron excitation by XUV pulse
    - depth and energy distribution of excited electrons
    - delayed excitation of inner shell electrons?
  - Transport of electrons in metals
    - elastic and inelastic scattering processes
    - dielectric function
- Comparison to experimental results
- Summary – Outlook

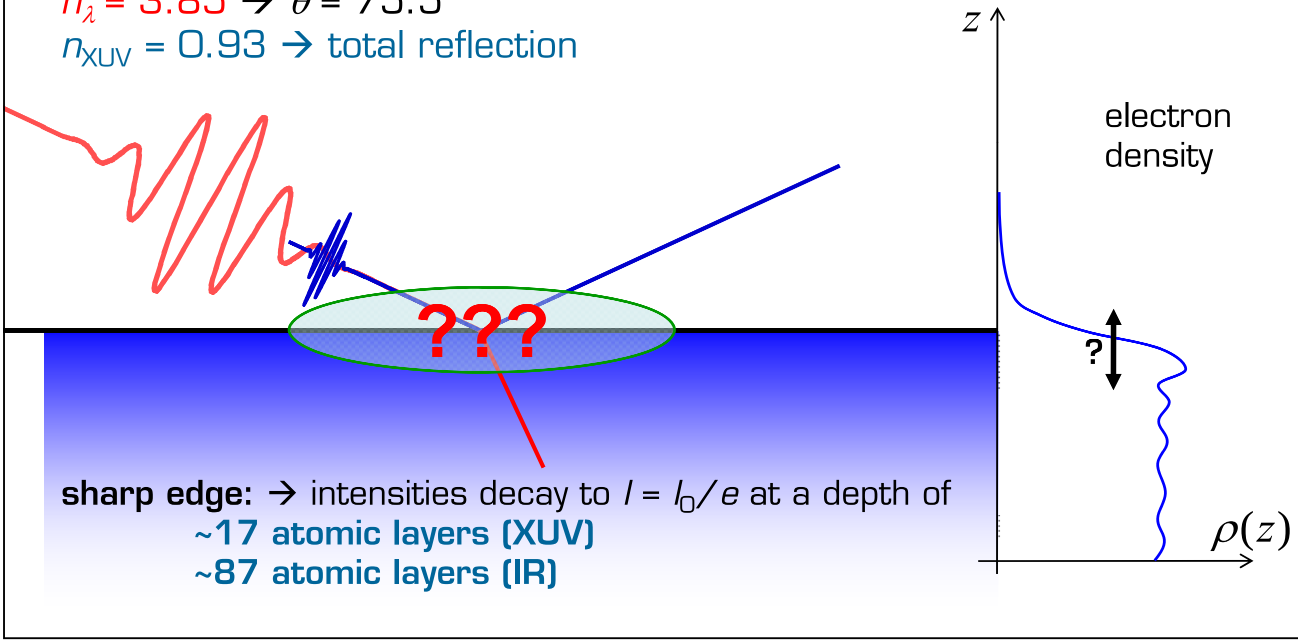
Methods

- Delay due to crystal structure
  - Cavalieri et al.: dispersion relation perpendicular to surface (Bragg scattering)
  - Zhang and Thumm: “structure factor” for core electrons, jellium for CB electrons + fitting to experimental result
- Delay due to transport
  - Baggesen and Madsen: Boltzmann transport equation, anisotropy due to surface neglected
  - Kazansky and Echenique: wave function propagation of core and CB electrons; delay due to interaction with (shielded) core hole (presentation later today)
  - Lemell et al.: classical transport

Combined XUV and IR fields

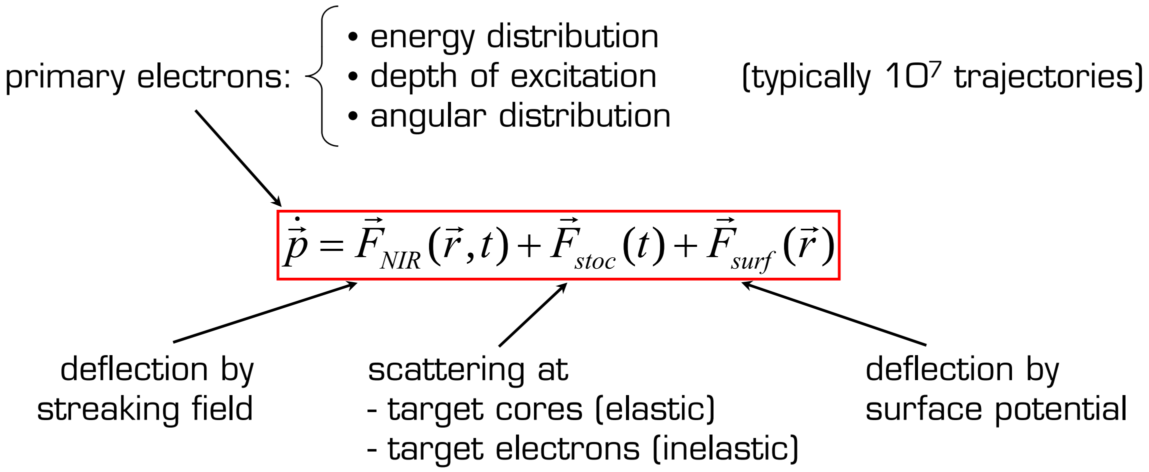
λ = 702 nm
n_λ = 3.85 → θ = 75.5°
n_{XUV} = 0.93 → total reflection

$$\varepsilon = \varepsilon_1 + i\varepsilon_2 = \left(n^2 - k^2\right) + i\,2nk$$



sharp edge: → intensities decay to $I = I_0/e$ at a depth of
~17 atomic layers (XUV)
~87 atomic layers (IR)

Classical Transport Simulation



primary electrons:
- energy distribution
- depth of excitation
- angular distribution

(typically 10^7 trajectories)

$$\dot{\vec{p}} = \vec{F}_{NIR}(\vec{r},t) + \vec{F}_{stoc}(t) + \vec{F}_{surf}(\vec{r})$$

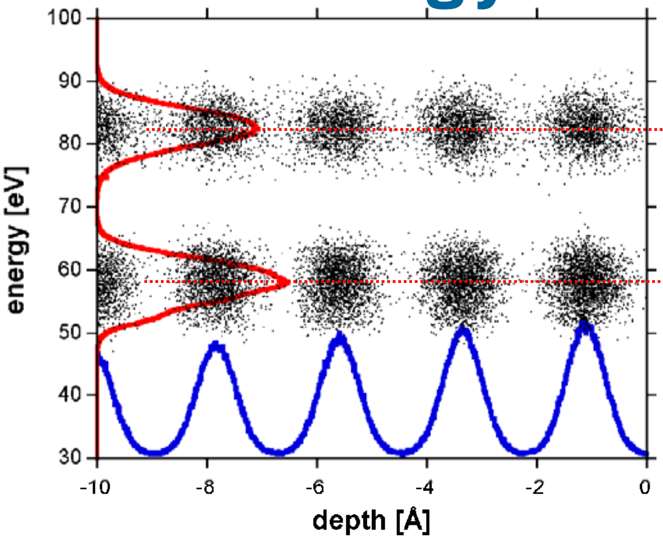
deflection by streaking field

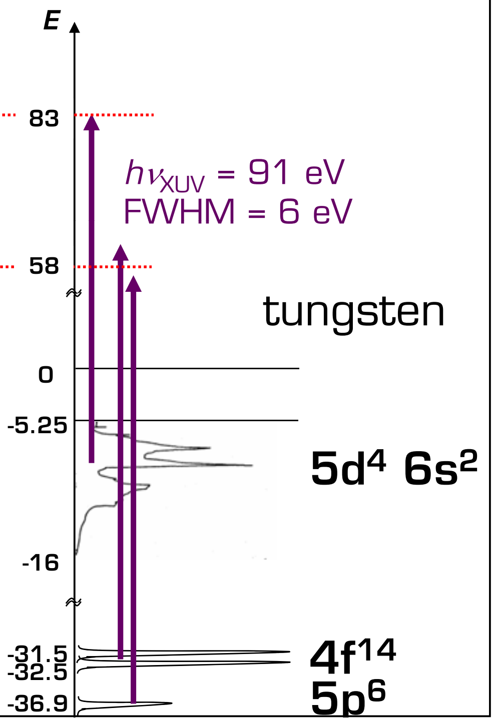
scattering at
- target cores (elastic)
- target electrons (inelastic)

deflection by surface potential

transport of secondary electrons included
→ particle number not conserved

Energy distribution



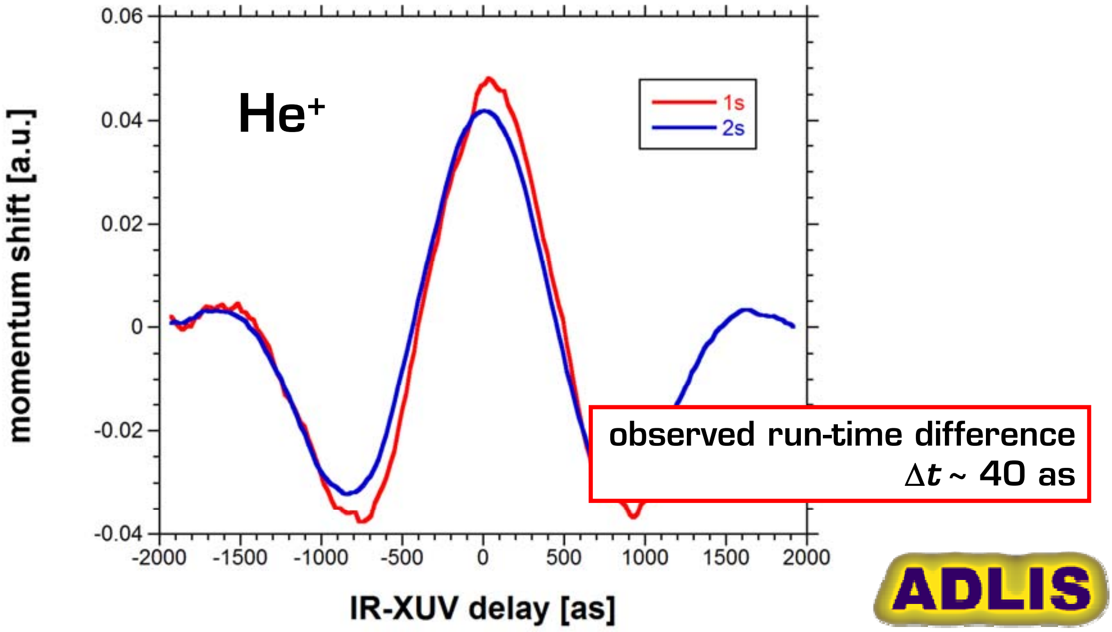


evanescent wave: $I \sim \exp(-2\kappa z)$
$\kappa \sim 0.013/$Å **~17 atomic layers**

tungsten:
a = 3.16 Å
$d_{(110)}$ = 2.24 Å

Excitation delay?

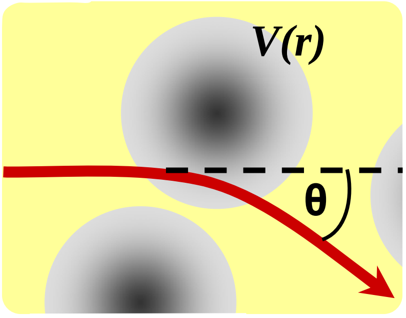
Experiments with Ne → observed delay for different *n*,*ℓ*

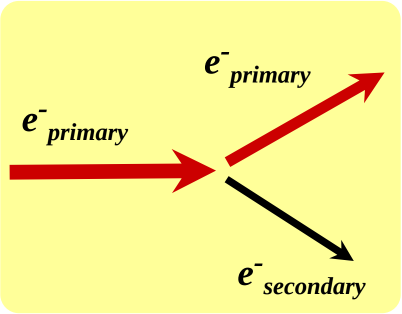


observed run-time difference
$\Delta t \sim 40$ as

ADLIS

Electron transport





elastic scattering:
muffin-tin potentials for W atoms
partial wave expansion
→ doubly differential scattering cross section

inelastic scattering:
from dielectric function
→ inelastic mean free path

$$E'_{prim} = E_{prim} - \Delta E$$

$$E_{sec} = \Delta E$$

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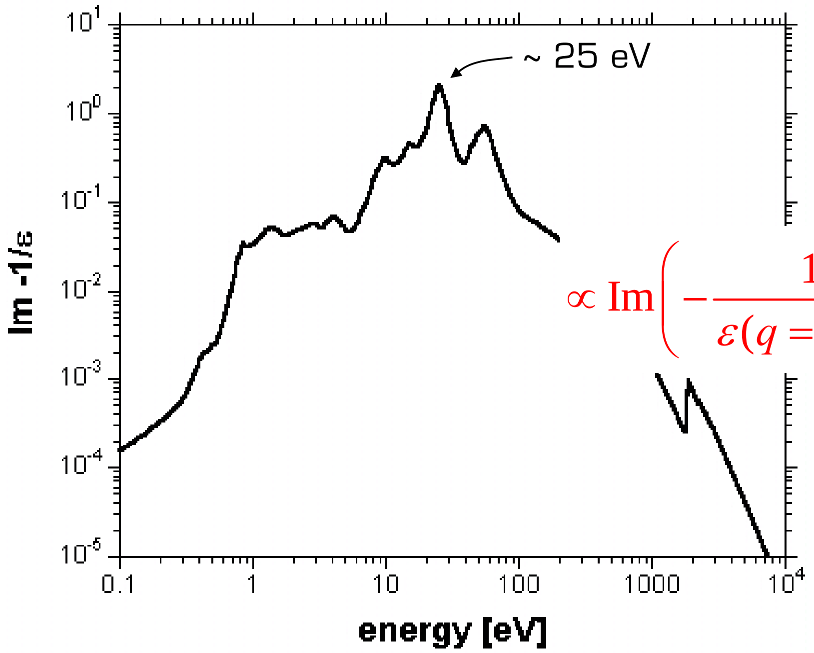
Energy loss



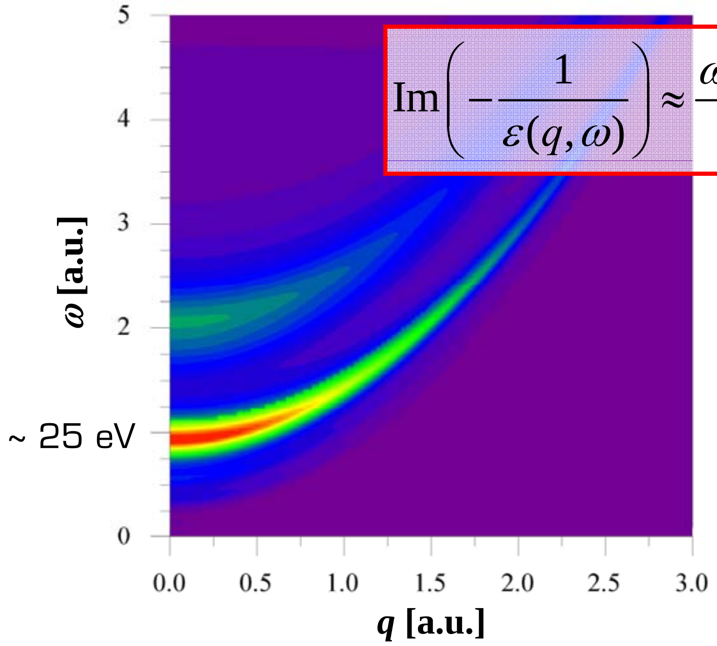
optical data for W

loss function:

$$\propto \mathrm{Im}\left(-\frac{1}{\varepsilon(q=0,\omega)}\right)=\mathrm{Im}\left(-\frac{1}{\varepsilon_{opt}(\omega)}\right)$$

Dielectric function for W

extrapolation of optical data to *q*–*ω* plane

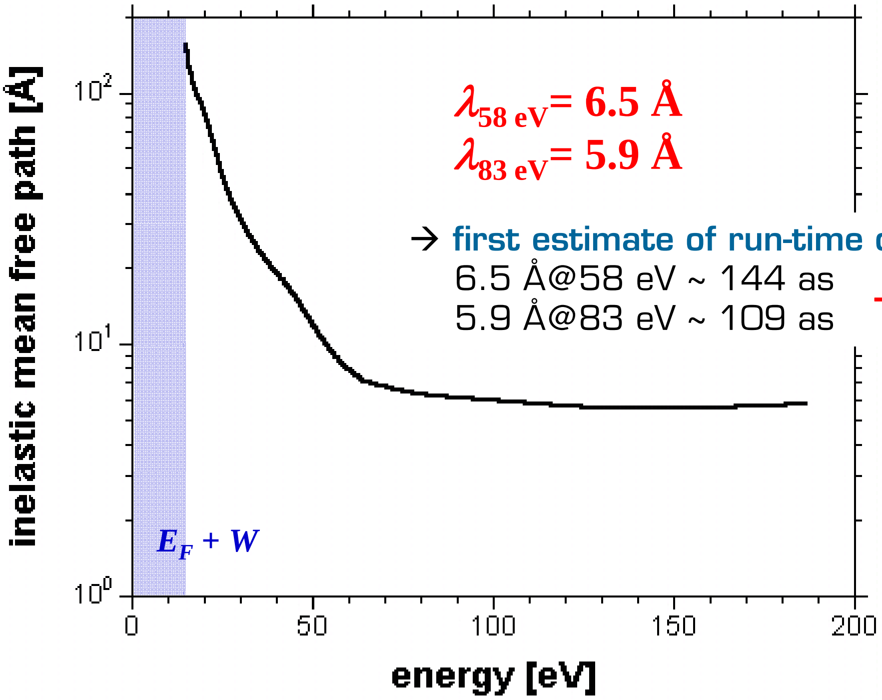


$$\boxed{\mathrm{Im}\left(-\frac{1}{\varepsilon(q,\omega)}\right)\approx\frac{\omega-q^2/2}{\omega}\mathrm{Im}\left(-\frac{1}{\varepsilon_{opt}(\omega-q^2/2)}\right)}$$

neglects creation of
electron-hole pairs;
good approximation for
electron energies $E \gg \omega_p$ ~ 25 eV

$$\boxed{\frac{d^2\lambda_{inel}^{-1}}{dq\,d\omega}\propto\mathrm{Im}\left(-\frac{1}{\varepsilon(q,\omega)}\right)}$$

Inelastic mean free path



$\lambda_{58\ \mathrm{eV}} = 6.5$ Å
$\lambda_{83\ \mathrm{eV}} = 5.9$ Å

→ **first estimate of run-time difference:**
6.5 Å@58 eV ~ 144 as
5.9 Å@83 eV ~ 109 as

→ Δt ~ 35 as

Streaking

streaking field: $E(t) = E_0 \sin^2[t\pi/2\tau]\cos[\omega_{NIR}(t-\tau)]$

momentum change within target: $\Delta p = \int_{t_{start}}^{t_{escape}} E(t)\,dt$

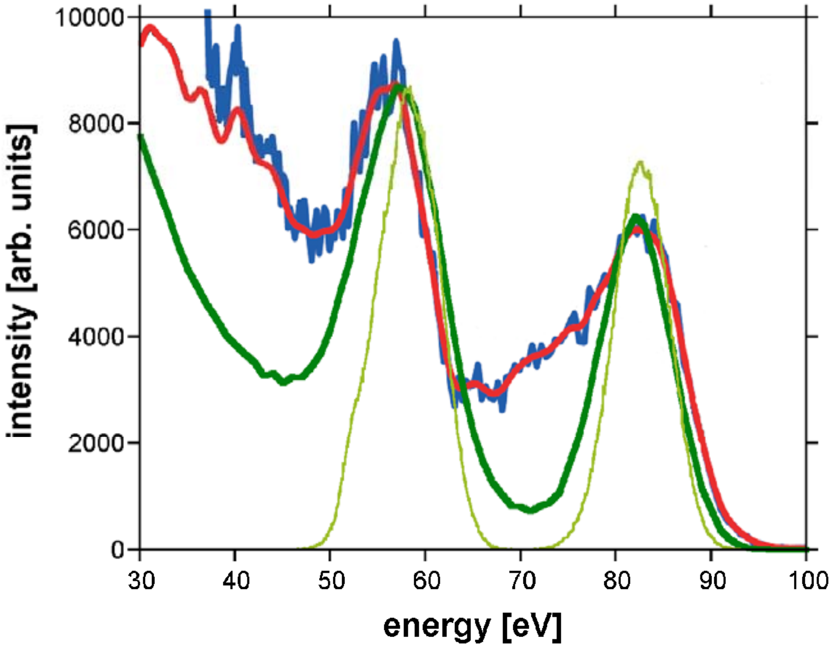
almost perpendicular to observation direction, Δt much shorter than oscillation period
→ small sub-surface contribution

momentum change after escape: $\Delta p = \int_{t_{escape}}^{\infty} E(t)\,dt$

parameters:

- XUV-pulse: FWHM τ_{XUV} ~ 300 as (defines t_{start})
- streaking laser wavelength λ = 702 nm
- FWHM τ_{NIR} = 5.7 fs
- $I = 2 \times 10^{12}$ W/cm² → E_0 = 0.075 a.u.

Electron energy spectra

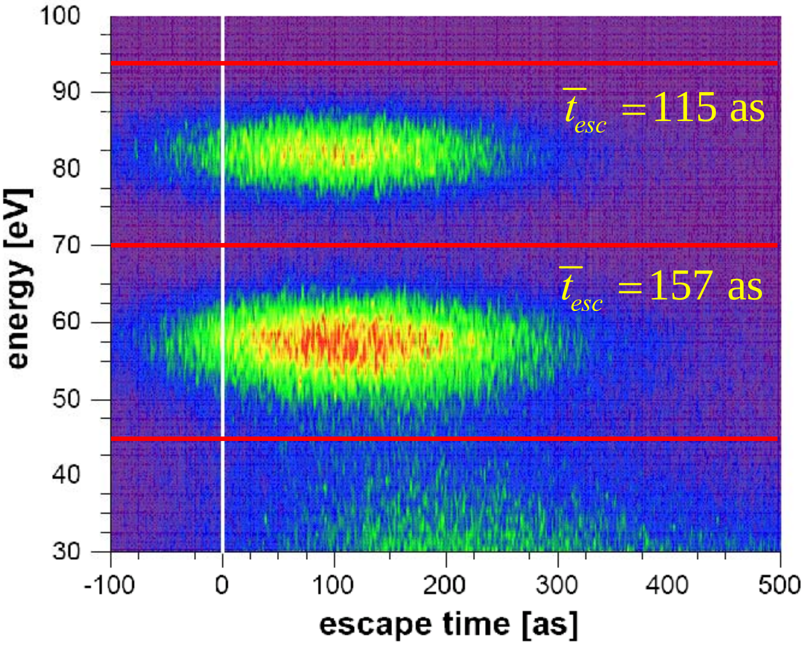


maximum of XUV pulse at start of NIR pulse; free electron disp. rel.

experiment:
raw spectrum
smoothed spectrum

simulation:
excitation spectrum
energy spectrum of escaped electrons

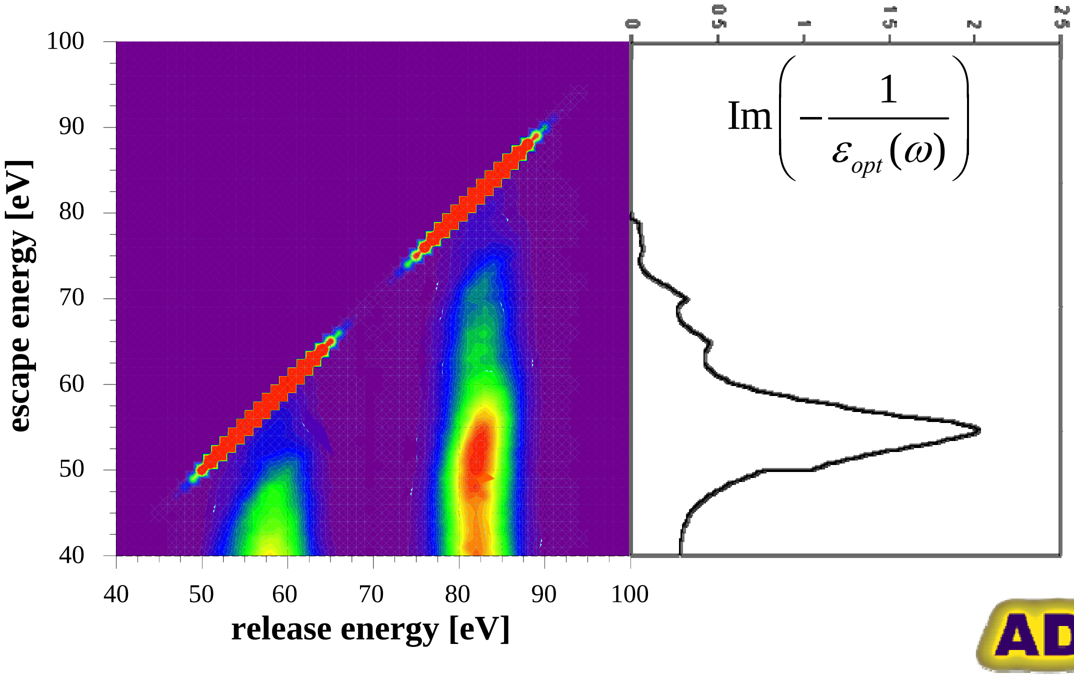
Escape time of electrons



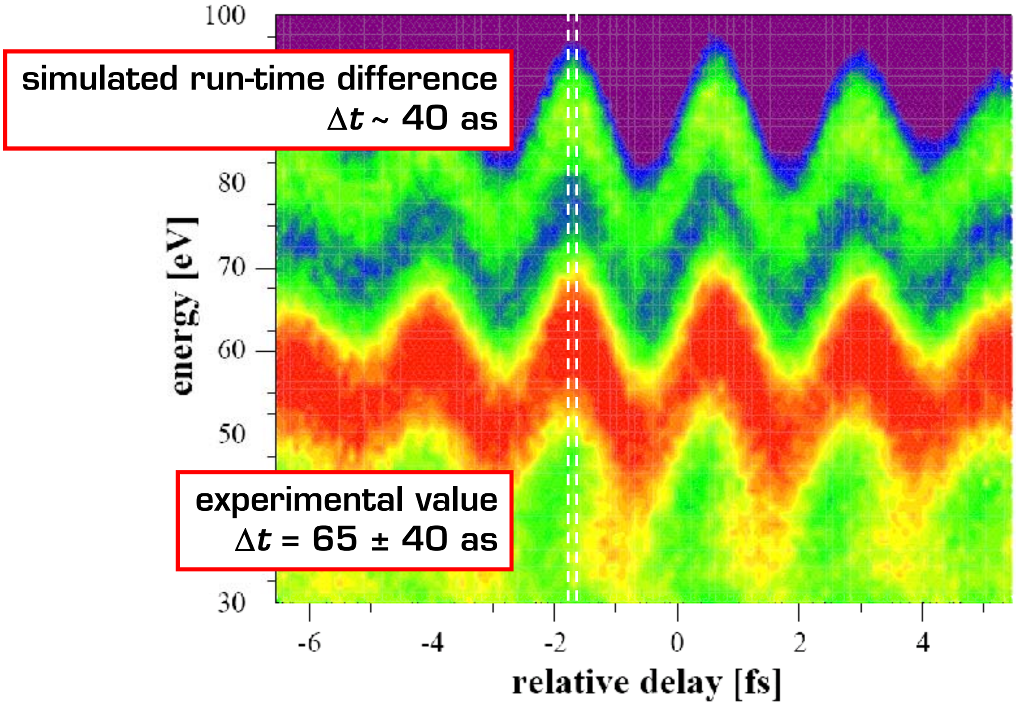
maximum of XUV pulse at $t = 0$

run-time difference: $\Delta t = 42$ as

Energy loss and delay



Streaked spectra



simulated run-time difference
$\Delta t \sim 40$ as

experimental value
$\Delta t = 65 \pm 40$ as

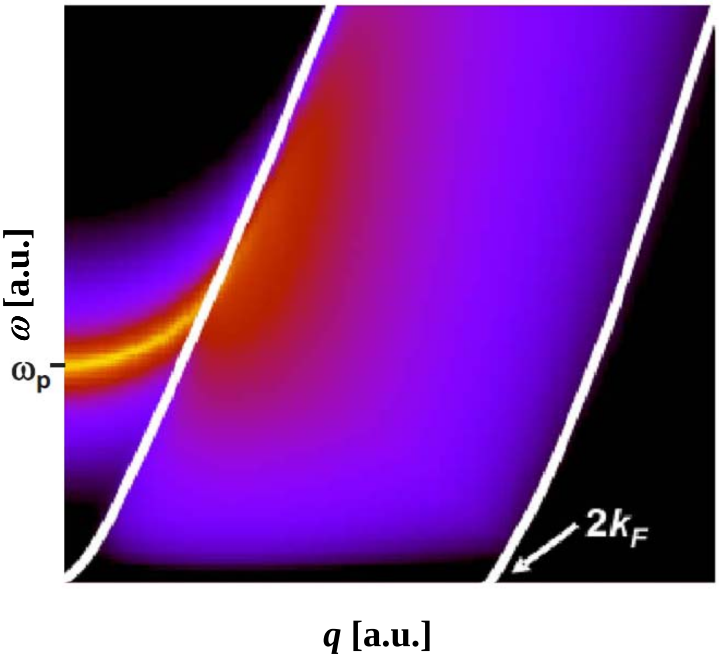
Other materials

theorists choice: Al target; Mg experiments in preparation

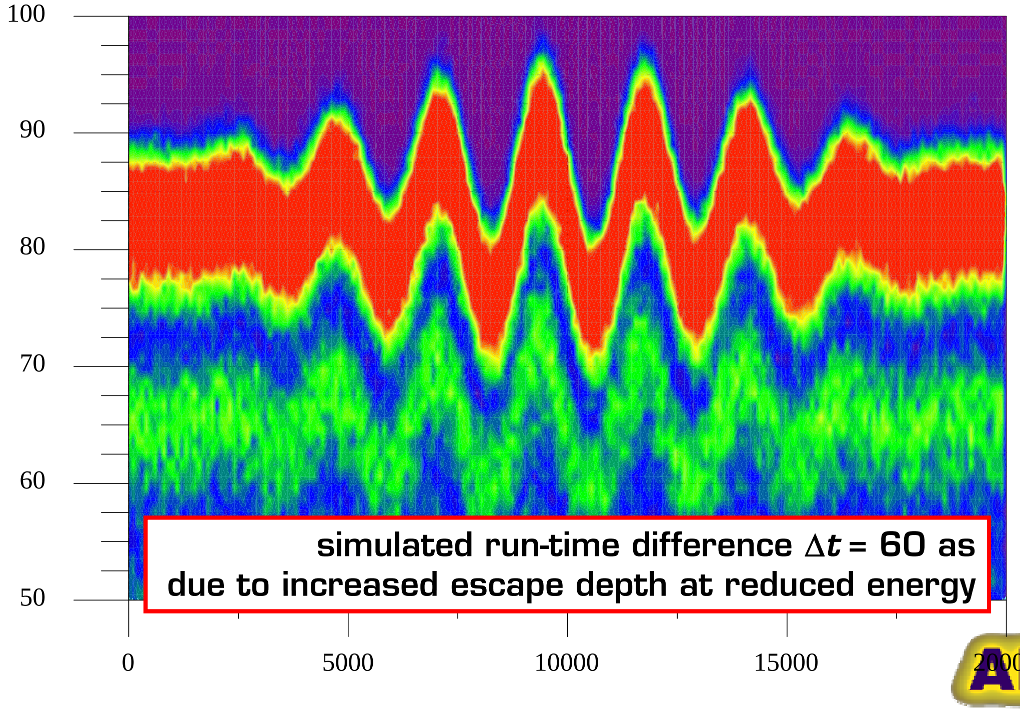
advantages:
- (almost) free electron gas
- strong plasmon loss
- deeply bound core electrons

disadvantage: hard to clean (AlO-layer)

Dielectric function of Al



Streaking of Al



simulated run-time difference Δt = 60 as
due to increased escape depth at reduced energy

Summary

- Various models for photoelectron streaking at surfaces
- Classical transport model
  - good agreement with measured energy spectra
  - delayed emission of deeply bound electrons?
- Simulated run-time difference too small

Remaining differences

- Intrinsic delay for core electrons?
- Crystal effect on excitation and transport?
- Localized surface induced fields?