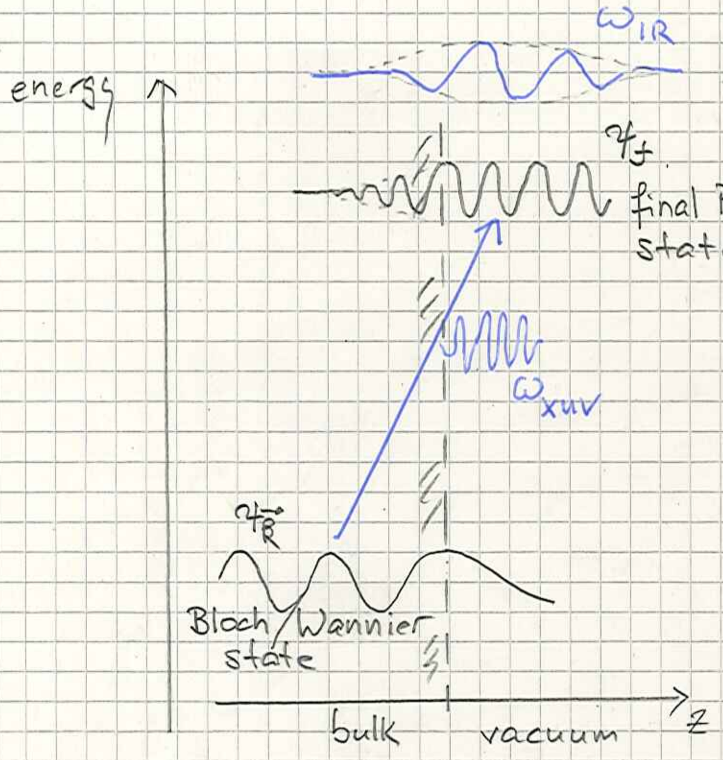
Numerical example : Comparison of Coulomb + laser induced streaking amplitudes and photoemission delays with SFA and numerical TDSE results.

$\downarrow$

hand out

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

Streaked photoemission from metal surfaces



recall :

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

emission amplitude

incoherent sum over occupied initial states $\rightarrow$ 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 :

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

$$k_{f,z} = Re(k_{f,z}) - i \underbrace{\lambda(k_f)}_{\text{damping factor}} / 2$$

damping factor (e^- mean-free path)

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

(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

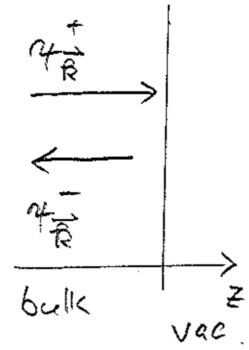
$$u_{\vec{k}}(\vec{r}) = u_{\vec{k}}(\vec{r} + \vec{R}_{\vec{n}})$$

↑ crystal lattice

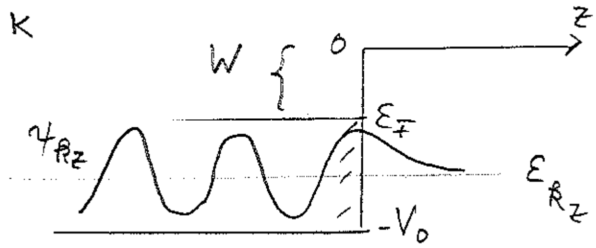
- can be applied to localized & delocalized initial states
- localized states: Bloch waves are superpositions of core levels that are localized at lattice sites $\{\vec{R}_n\}$ with binding energies ε_i

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

$$\psi_{\vec{k}} = \underset{\text{incident}}{\psi^{+}_{\vec{k}}} + \underset{\text{surface reflected}}{\psi^{-}}$$



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



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

momentum / position in surface plane

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

$$\psi_{k_z}(\vec{r}) \sim e^{i k_z \cdot z} + \underset{\substack{\uparrow \\ \text{reflection coeff}}}{R(k_z)} e^{-i k_z z}$$

reflection coeff : $R(V_0; k_z)$

$$\downarrow$$

$$\varepsilon_F + W$$

W ↑ work function

examples ↓ handout : a) tungsten :

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

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

4f – conduction-band delay :

$$\Delta\tau^{exp.} = (110 \pm 70) \text{ as}$$

70 ↑ later improved

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

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

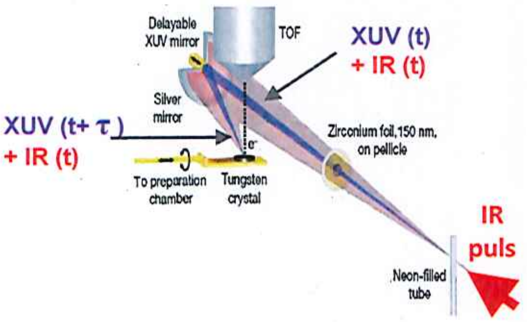
c) adsorbate – covered tungsten : Mg/W(110)

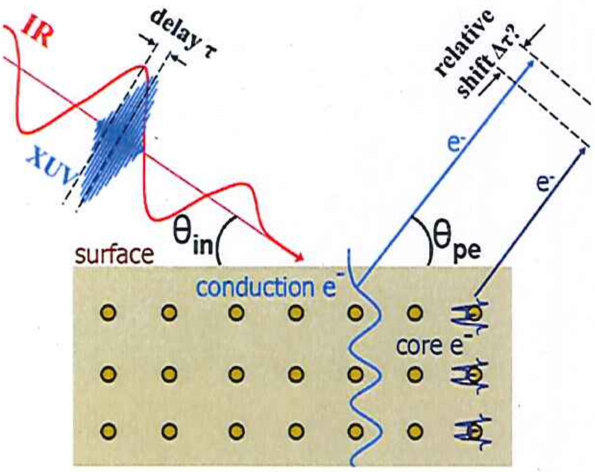
experiment : Neppl et al., Nature 517, 342 (2015)

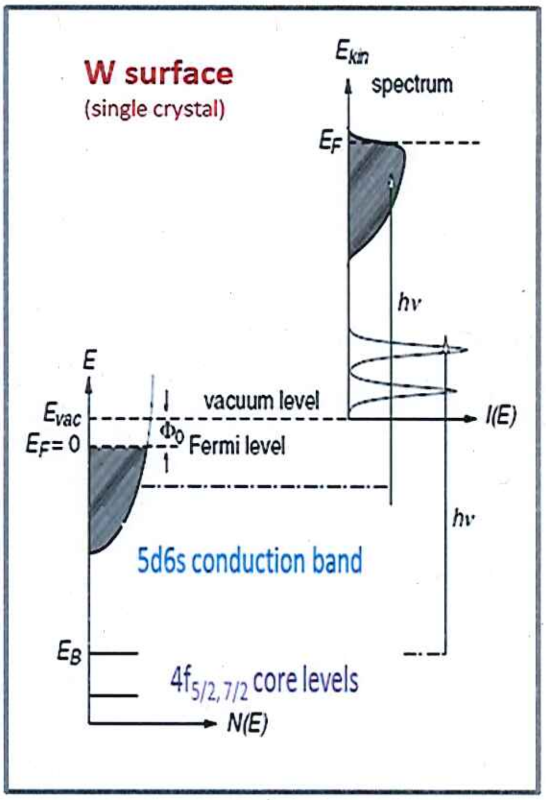
theory : Liao & U.T., Phys. Rev. A 92, 031401 (2015)

Time-resolved photoemission from metal surfaces

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

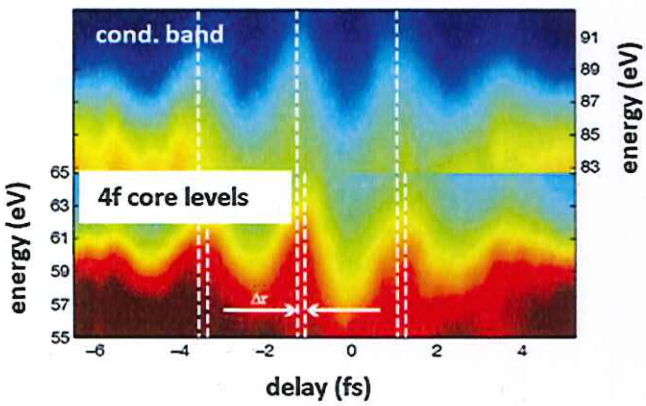






Experiment: $\Delta\tau = 110 \pm 70\,\mathrm{as}$

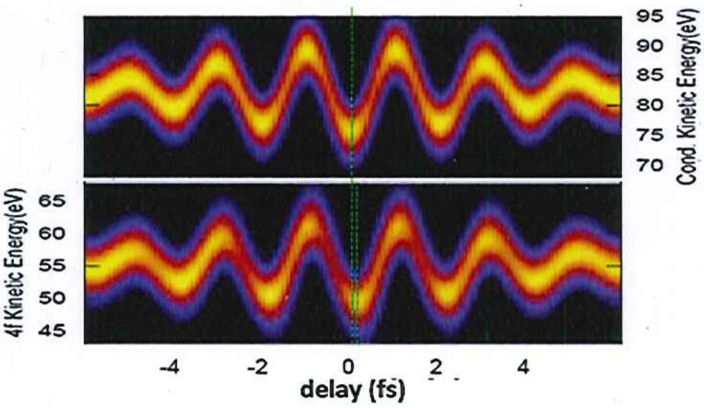
Cavalieri *et al.*, *Nature* **449**,1029(2007)



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

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

PRA **84**, 063403 (2011)



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

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

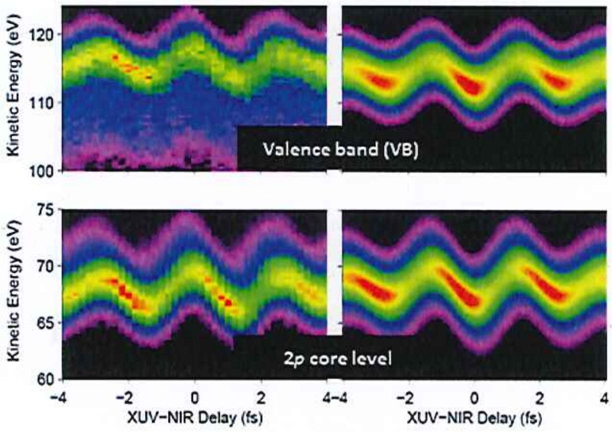
Experiment Neppl *et al.*, *PRL* **109**, 087401 (2012)

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

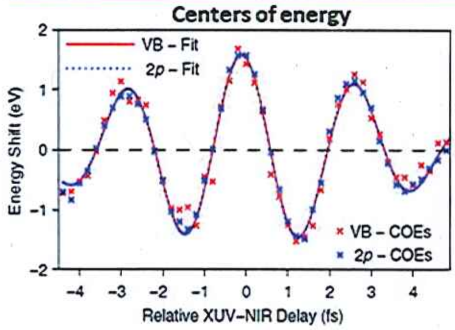
XUV: 435 as, 118 eV

NIR: 800 nm, 5 fs
CEP = 0

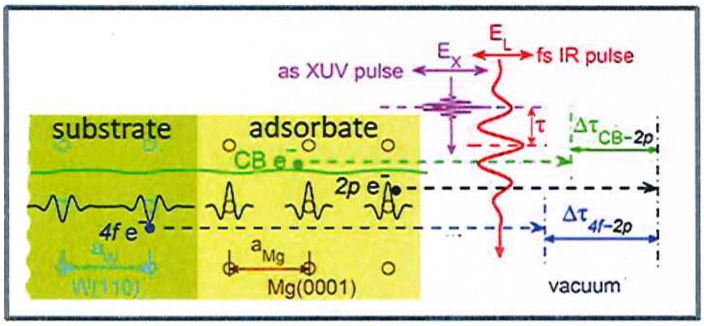
IR-skin depth = 2 Å
MFPs: 4.9 Å for VB
3.7 Å for Mg(2p)

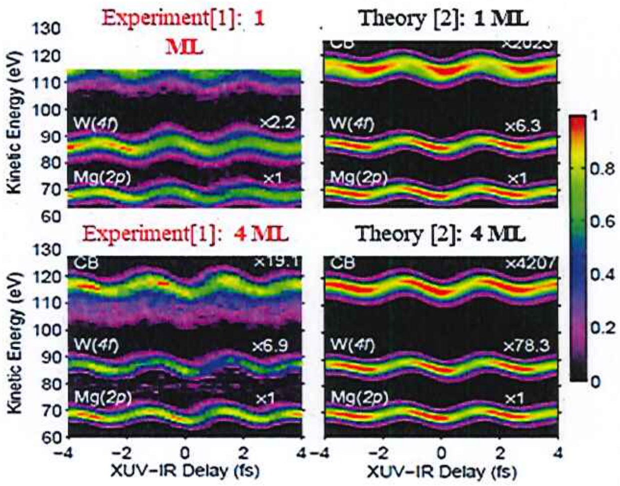


VB – 2p streaking time delay = 0

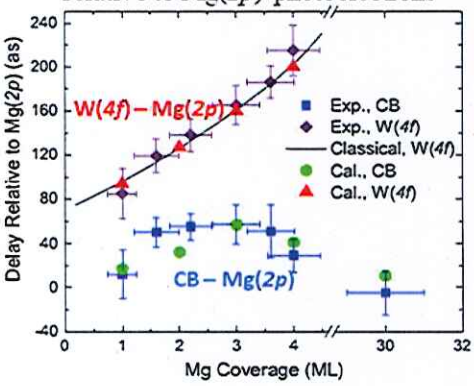


Streaked photoemission from heterogeneous structures: *Mg/W(110)*





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

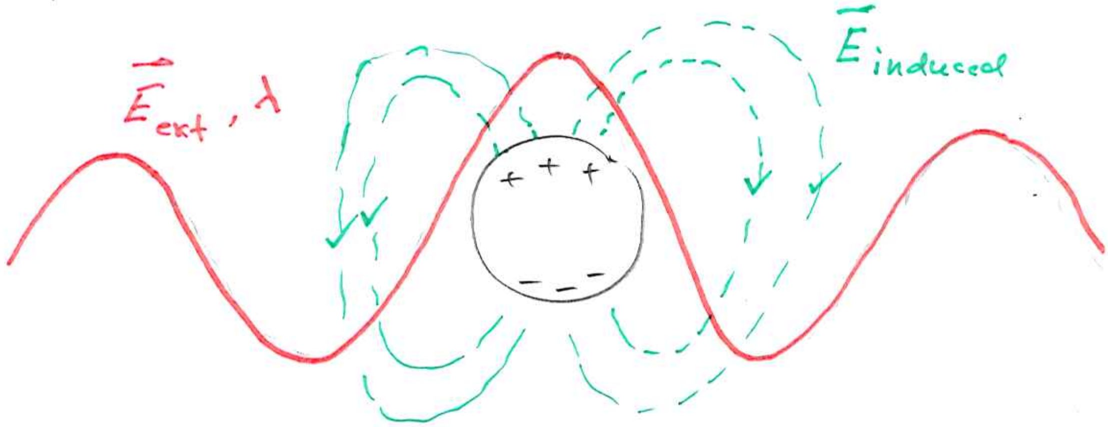
- W (4f) - Mg (2p) delay : increases with adsorbate thickness (as expected)

- 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 solids

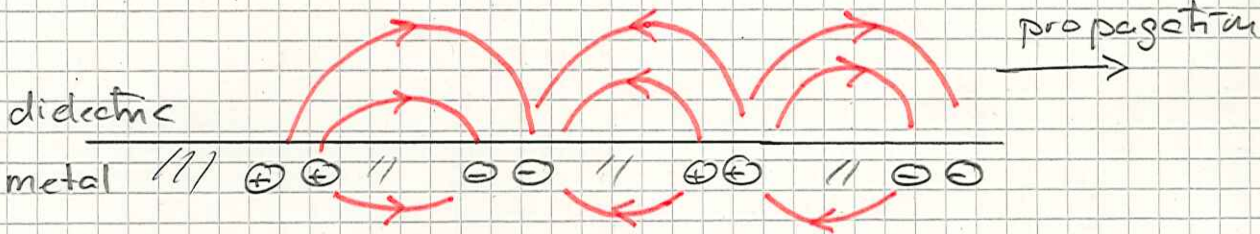
E-M waves can excite modes of collective e^- motion in matter. Examples are :

(i) Localized surface plasmons on metal nanoparticles (with diameter < $\lambda/2$)

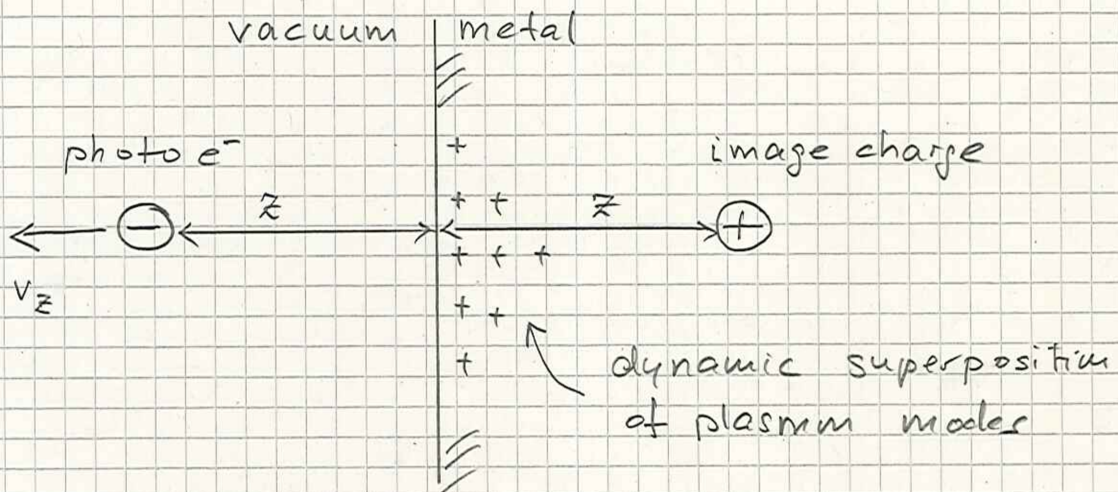


⇒ strong induced (plasmonic) E-M field.

(ii) Traveling surface plasmon polariton at metal - dielectric/vacuum interface



(iii) XUV surface photoemission. Surface & bulk plasmon excitation (surface wake potential)



Contribution of dynamical surface charge (image charge) build-up to photo e⁻ streaking delay

Compare

$$V_{im}^{static}(z) = -\frac{1}{4z}$$

static image charge potential $\hat{=}$ instantaneous electron self-interaction

$$V_{im}^{dyn}(z, v_z) \underset{\substack{\text{large } z \\ \text{small } v_z}}{\longrightarrow} V_{im}^{static}(z)$$

effective potential on PE due to plasmon response

Calculation of plasmon response :

Hamiltonian

creation / annihilation operators

$$H_0 = \underbrace{\sum_{\vec{k},\, k_z \geq 0} \omega_k \, b^+_{\vec{k}} b_{\vec{k}}}_{\text{bulk plasmons}} + \underbrace{\sum_{\vec{Q} \,\in\, \text{surface plane}} \sigma_Q \, a^+_{\vec{Q}} a_{\vec{Q}}}_{\text{surface plasmons}}$$

Dispersion relations (including: plasmon + single-particle excitation)

$$\omega_k = \left(\omega_p^2 + 3 k_F^2 k^2 + k^4/4 \right)^{1/2}$$

decay into e^--hole pairs

$$\sigma_Q = \left(\omega_s^2 + \sqrt{3}\, k_F \omega_s Q / \sqrt{5} + \beta Q^2 + Q^4/4 \right)^{1/2}$$

where

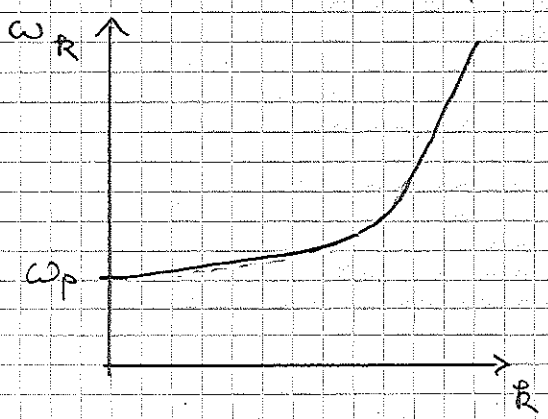
$\omega_p = 4\pi n$ (bulk) plasmon frequency

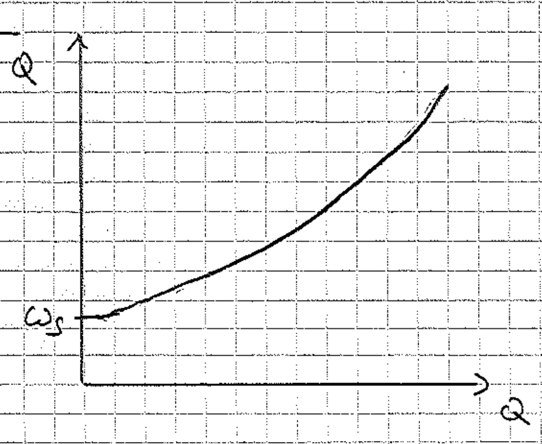
$\omega_s = \omega_p / \sqrt{2}$ surface —"—

n : bulk e^- density

$k_F = (3\pi^2 n)^{1/3}$ Fermi momentum

β : fit parameter, determined to make surface & bulk dispersion merge e^--hole continuum at same momentum and energy





- assume classical photo electron

$$\varrho(\vec{r},t) = -\underbrace{\delta(\vec{r}_{\parallel})}_{\substack{\text{for emission} \\ \perp \text{ to surface}}}\,\delta(z,t) \quad \text{charge density}$$

Interaction by plasmon creation / annihilation

$$H_{int} = \int d\vec{r}\;\varrho(\vec{r},t)$$

$$\times\Bigg[\underbrace{\sum_{\vec{Q}}\left(\frac{\pi\,\omega_s^2}{A\,Q\,\sigma_Q}\right)^{1/2} e^{-Q|z|}\, e^{i\vec{Q}\cdot\vec{r}_{\parallel}}\left(a^{+}_{\vec{Q}} + a_{-\vec{Q}}\right)}_{\text{surface plasmon excitation}}$$

$$+\;\underbrace{\Theta(-z)\sum_{\vec{k},\,k_z\geq 0}\left(\frac{8\pi\,\omega_p^2}{V\,k^2\,\omega_k}\right)^{1/2}\sin(k_z z)\,e^{i\vec{k}\cdot\vec{r}_{\parallel}}\left(b^{+}_{\vec{k}_{\parallel},k_z} + b_{-\vec{k}_{\parallel},k_z}\right)}_{\text{bulk plasmon excitation}}\Bigg]$$

where

A/V = quantization surface / volume

Evolution of initial plasmon field $|\psi_0\rangle = |\psi(t=-\infty)\rangle$
(= vacuum state of plasmon field)

$$|\psi(t)\rangle = \underset{\substack{\uparrow \\ \text{time} \\ \text{ordering}}}{T}\,\exp\left[-i\int_{-\infty}^{t}dt'\;\underbrace{e^{iH_0 t}\,H_{int}\,e^{-iH_0 t}}_{H_{int}\text{ in interaction picture}}\right]|\psi_0\rangle$$

this requires some algebra, but can be calculated analytically.

Finally, the dyn. image potential is obtained :

$$V_{im}^{dyn}(z, v_z(t)) = \frac{1}{2} \langle \psi(t) | e^{iH_0 t} H_{int} e^{-iH_0 t} | \psi(t) \rangle$$

$$= \Sigma_r(z, v_z(t)) + i \, \Sigma_i(z, v_z(t))$$

$$\Sigma_r \xrightarrow[v_z \to 0]{z \to \infty} -1/(4z), \qquad \Sigma_i \xrightarrow[v_z \to 0]{z \to \infty} 0$$

Handout : Num. example for Al surface,
from [1]=Zhang + U.T., Phys. Rev. A <u>84</u>, 063403 (2011)

(Fig. 1:) - v_z dependent "wake" potential on vacuum side

- wake amplitude ↗ as v_z ↗

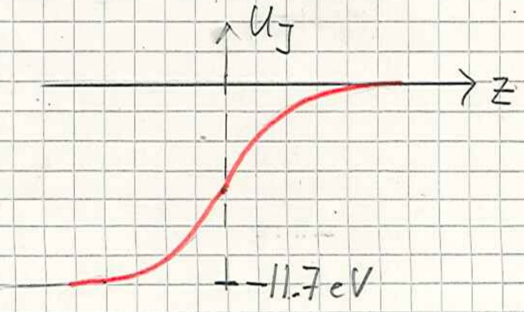
- wake amplitude → 0 as $v_z \to 0$.

Compute streaked spectrum for static & dyn. image potentials:

(i) Find eigenfunction for thick Al slab
(typical thickness in num. calc. = 300 a.u.)

$$\left[-\frac{1}{2}\frac{d^2}{dz^2} + U^{static}(z) - \varepsilon_k \right] \psi_k = 0 \quad (\text{short } k \text{ for } k_z)$$

$$U^{static} = \underbrace{\frac{-11.7\,\text{eV}}{1 + \exp(z/2.8)}}_{=: U_J(z), \text{ adjusted to DFT results}} + V_{im}^{static}(z)$$

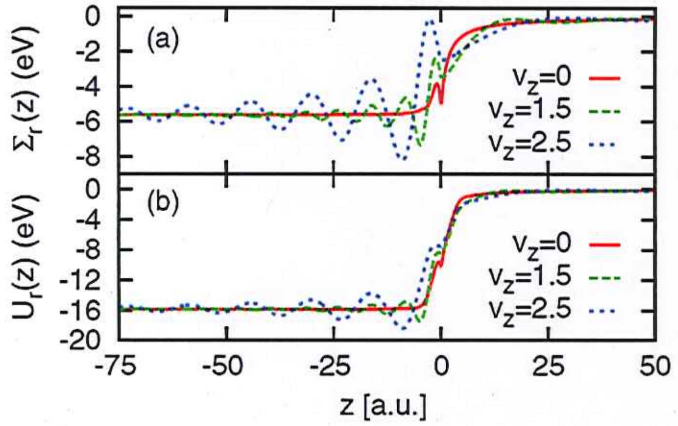


smeared-out step ("jellium") potential

$$\Rightarrow \{ \psi_k(z,t) = \psi_k(z) e^{-i\varepsilon_k t} \}$$

47a

Towards the time-resolution of collective excitations in solids



(a) Dynamic wake potential V_{im}^{dyn} for Al for different PE velocities.
(b) Real part of the total surface potential U^{dyn}. The

Streaked XUV photoemission from Al surface

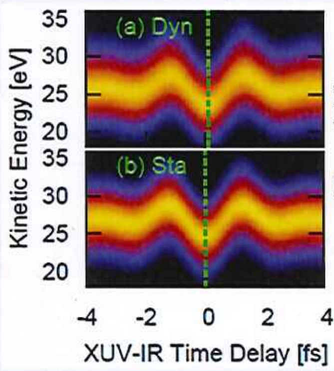
$$\Delta\tau_{dyn} - \Delta\tau_{sta} = 100\,\text{as}$$

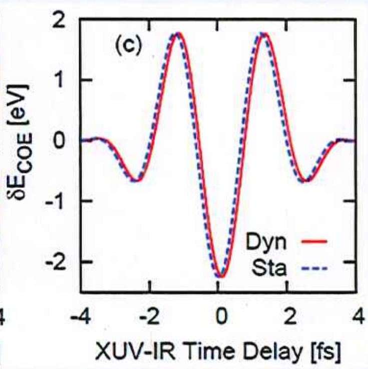
Dynamic surface-charge rearrangement $V_{im}(z, v_z)$ surf.+bulk plasmon excit.

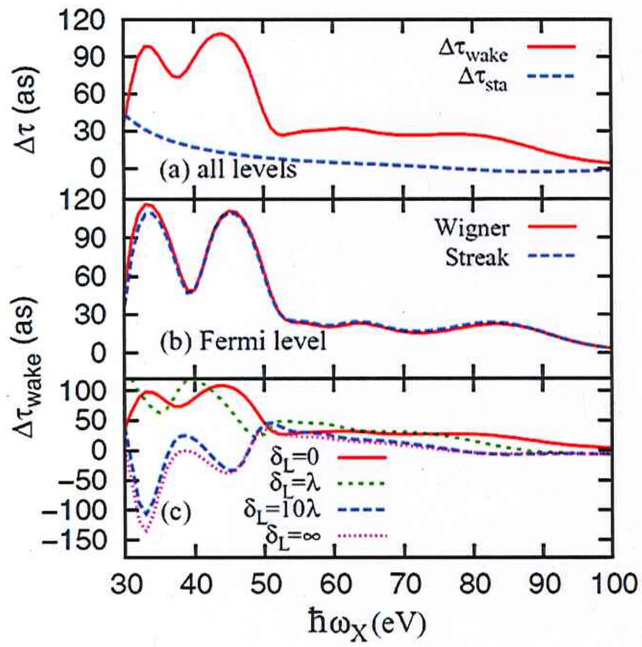
Instant (static) surface charges $V_{im}^{static}(z) = -\frac{1}{4z}$

$\hbar\omega_X = 40\,\text{eV}, \hbar\omega_s = 10\,\text{eV}$

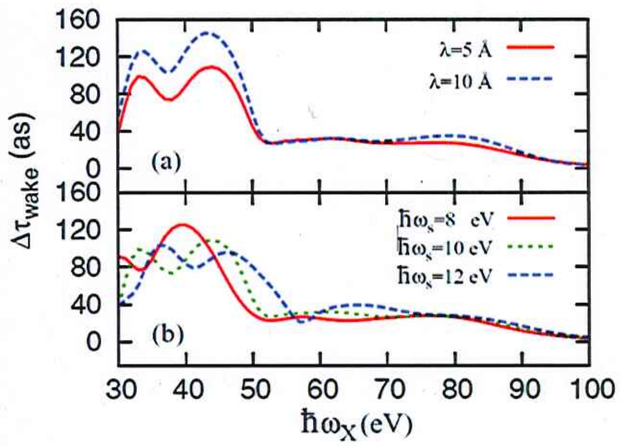
$\lambda = 10\,a.u., \delta_L = 0$







Dependence on the IR skin depth δ_L. "Wigner" refers to the extraction of relative time delays from the PE wave packet center.



Dependence on (a) the electron mean-free path and (b) surface plasmon energy.

Zhang & U.T., *PRA* **84** (2011)
U.T., Liao, *et al.*, Handbook of Photonics, Vol.**1**, Chap. XIII (Wiley, 2015)

(ii) PE propagation from occupied substrate states $\{\psi_k\}$:

$$i\frac{\partial}{\partial t}\phi_k^{\substack{\text{static}\\ \text{dyn}}}(z,t) = \Bigg[\frac{1}{2}\Big(-i\frac{\partial}{\partial z} + A_{IR}(z,t)\Big)^2 + \begin{Bmatrix} U^{\text{static}}(z) \\ U^{\text{dyn}}(z, v_z) \end{Bmatrix}\Bigg]\phi_k^{\substack{\text{static}\\ \text{dyn}}} + z E_{XUV}(t+\tau)\,\psi_k(z,t)$$

surface screened IR streaking field

$k \leq k_F$ at 0K

where

$$U^{dyn} = U_J(z) + V_{im}^{dyn}(z, v_z)$$

(iii) Photoelectron emission probability ($\hat{=}$ streaked spectrum)

Fourier transform:

$$\tilde{\phi}_k(q,\tau) = (2\pi)^{-1/2}\int dz \lim_{t\to\infty} \phi_k(z,t)\, e^{-iqz}$$

Incoherently add contributions from occupied states:
(here for 0K)

$$P(\varepsilon,\tau) = \sum_{\varepsilon=\varepsilon(k)<\varepsilon_F} \int dq\, |\tilde{\phi}_k(q,\tau)|^2$$

Time delays

Separately fit centers of energy for calculations with U^{static} & U^{dyn}:

$$E_{COE}^{\substack{\text{static}\\ \text{dyn}}}(\tau) = \ddot{a} + \ddot{b}\,\underbrace{A_{IR}(t+\ddot{\tau})}_{\text{no } z\text{-dependence for } z>0}$$

$$\Delta\tau_{wake} = \tau^{dyn} - \tau^{\text{static}}$$

Handout: Num. example for Al surface (from [1])

(Fig. 2:) - streaked spectra

- $E_{COE}^{\substack{\text{static}\\ \text{dyn}}}(\tau) \Rightarrow \Delta\tau_{wake} = 100$ as