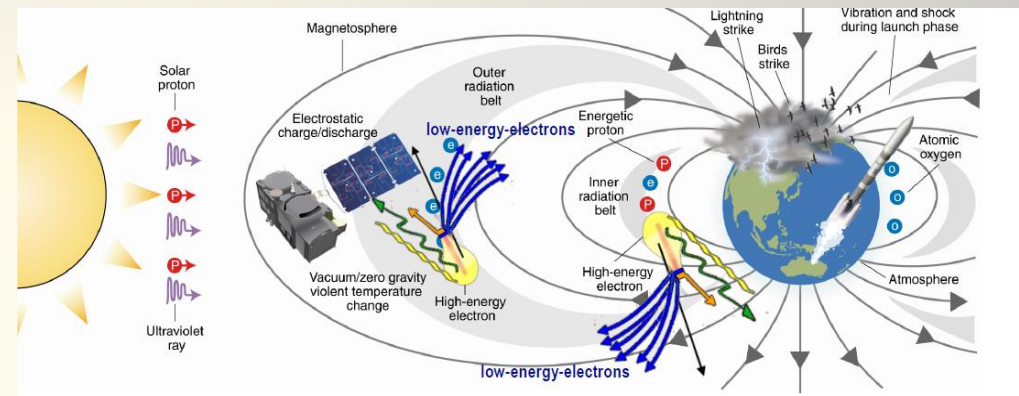


LEE2022

a brainstorming meeting on relevance of
Low Energy Electrons in aerospace



Contribution of plasmon decay to secondary electron emission

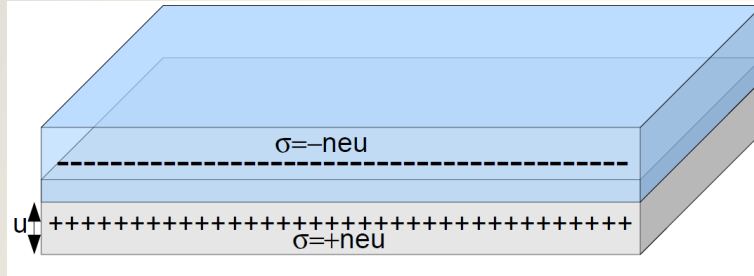
Alessandro Ruocco
Università degli Studi Roma Tre



Cosa sono i plasmoni

I plasmoni sono oscillazioni di carica collettive che si possono manifestare in sistemi diversi:

Elettroni all'interno di un solido



Supponiamo che gli elettroni subiscano uno spostamento longitudinale u :

$$u(t) = u_0 e^{i\omega t}$$

$$m\ddot{u}(t) = -eE(t) \quad E = 4\pi\sigma \quad \sigma = Neu$$

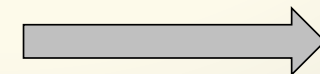
$$m\ddot{u}(t) + 4\pi Ne^2 u = 0$$

Equazione di un oscillatore armonico con frequenza propria:

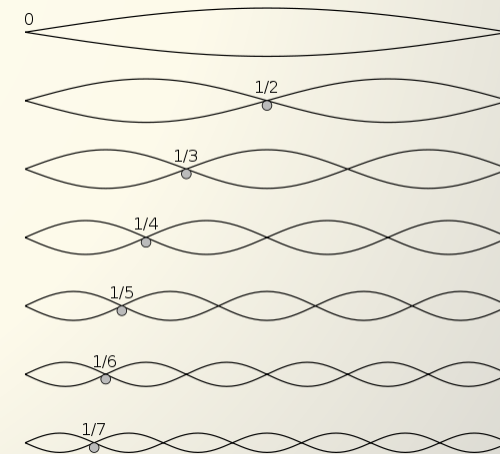
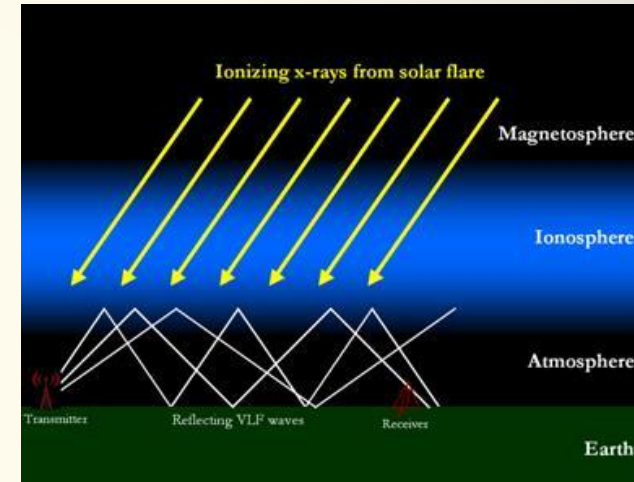
$$\omega_p^2 = \frac{4\pi Ne^2}{m}$$

Frequenza di plasma nel limite di lunghezza d'onda infinita

Le oscillazioni di plasma esistono anche per $q \neq 0$



Ionosfera



$$\omega(q) = \omega_p \left(1 + \frac{3v_F^2}{10\omega_p^2} q^2 \right)$$

Excitation mode compatible with Maxwell equations

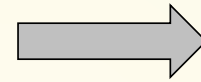
Plasmon is an e.m. excitation that must satisfy Maxwell equations

Maxwell equations

$$\begin{aligned} \text{div}(\mathbf{E}) &= 4\pi(\rho_{\text{pol}} + \rho_{\text{ext}}) \\ \text{div}(\mathbf{B}) &= 0 \\ \text{rot}(\mathbf{E}) &= -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t} \\ \text{rot}(\mathbf{B}) &= 4\pi(\mathbf{j}_{\text{ext}} + \mathbf{j}_{\text{pol}}) + \frac{1}{c} \frac{\partial \mathbf{E}}{\partial t} \end{aligned}$$

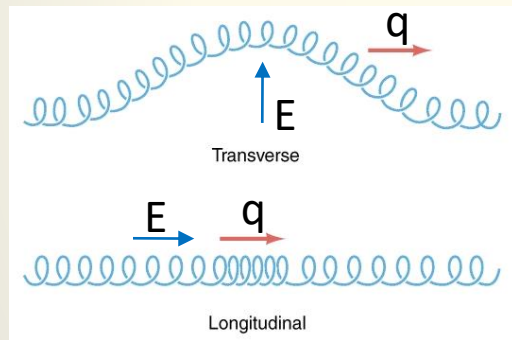
Maxwell equations in the Fourier space

$$\begin{aligned} \mathbf{q} \cdot \hat{\mathbf{D}} &= -4\pi i \hat{\rho}_{\text{ext}} \\ \mathbf{q} \cdot \hat{\mathbf{B}} &= 0 \\ \mathbf{q} \wedge \hat{\mathbf{E}} &= \frac{\omega}{c} \hat{\mathbf{B}} \\ \mathbf{q} \wedge \hat{\mathbf{H}} &= \hat{\mathbf{j}}_{\text{ext}} - \frac{\omega}{c} \hat{\mathbf{D}}. \end{aligned}$$



In the absence of external charge and current from Maxwell equations we obtain:

$$\mathbf{q} \cdot (\mathbf{q} \cdot \mathbf{E}) - q^2 \mathbf{E} = -\epsilon(\mathbf{q}, \omega) \frac{\omega^2}{c^2} \mathbf{E}$$



$$q^2 = \frac{\omega^2}{c^2} \epsilon(\mathbf{q}, \omega) \quad \text{(transverse wave)}$$

$$0 = \frac{\omega^2}{c^2} \epsilon(\mathbf{q}, \omega) E \quad \epsilon(\mathbf{q}, \omega) = 0 \quad \text{(longitudinal wave)}$$

$$D = \epsilon(\mathbf{q}, \omega) E$$

If external field is zero, $D=0$
The electric field E can be different from zero only if $\epsilon = 0$

Bulk plasmon in metal

Drude model for a simple metal:

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma}$$

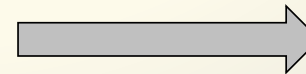
$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2}$$

$$\epsilon_1 = 0 \text{ per } \omega = \omega_p$$

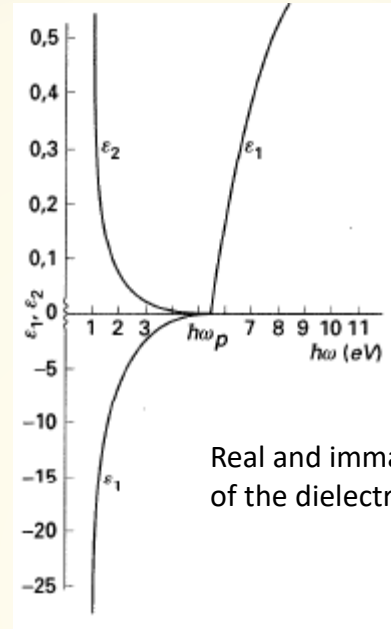
$$\omega_p^2 = \frac{4\pi N e^2}{m}$$

Plasma frequency in the limit of infinite wave length

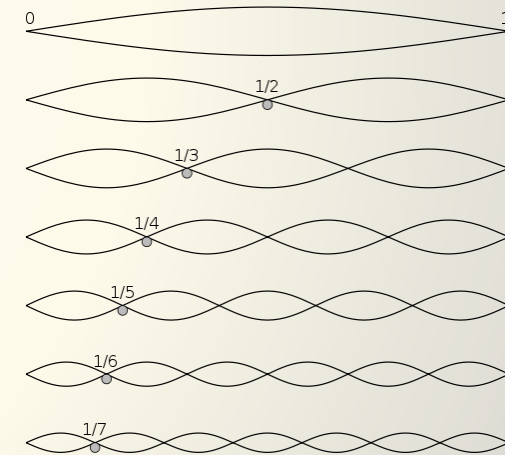
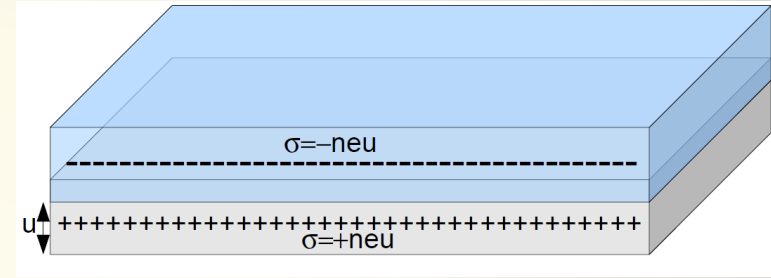
Dispersion relation for plasma excitation for $q \neq 0$



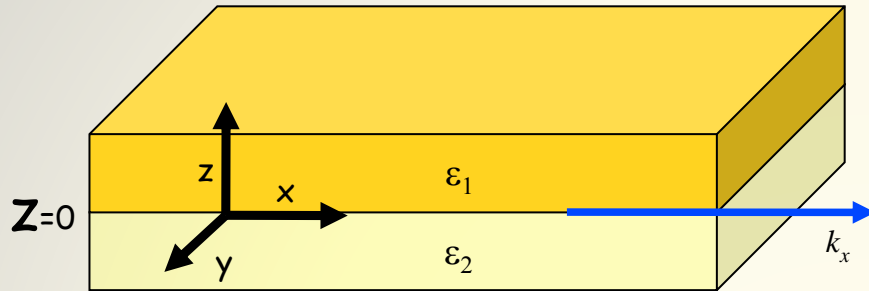
$$\omega(q) = \omega_p \left(1 + \frac{3v_F^2}{10\omega_p^2} q^2 \right)$$



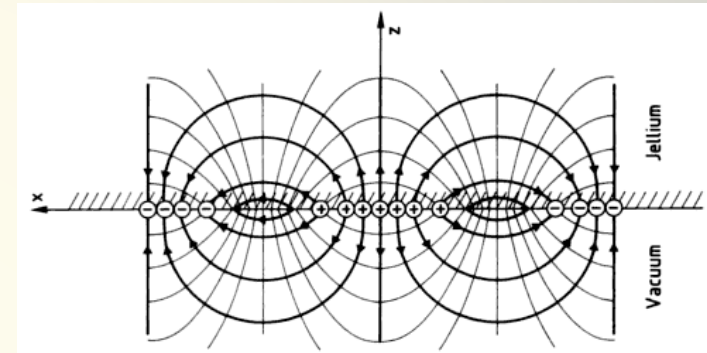
Real and imaginary part of the dielectric function



Surface plasmon



We consider an e.m. excitation propagating along x and strongly damped in the z direction inside the two media



TMW $E_i = (E_{ix}, 0, E_{iz}) e^{-\kappa_i |z|} e^{i(q_i x - \omega t)}$ $H_i = (0, H_{iy}, 0) e^{-\kappa_i |z|} e^{i(q_i x - \omega t)}$

Continuity condition across the surface separating the two media give rise to the following equation for the existence of a surface plasmon polariton

$$\epsilon_1 + \epsilon_2 = 0$$

$$\frac{\epsilon_1}{k_1} + \frac{\epsilon_2}{k_2} = 0$$



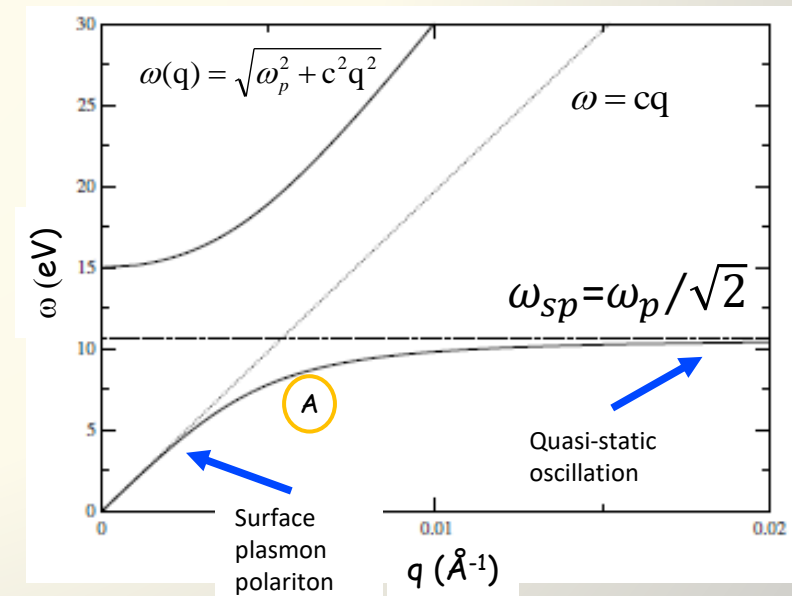
$$q(\omega) = \frac{\omega}{c} \sqrt{\frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2}}$$

If $\epsilon_2 = 1$ (vacuum) and ϵ_1 metal described by Drude model

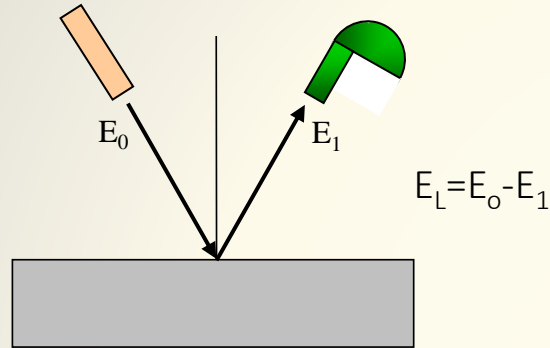
$$\epsilon_1 = 1 - \frac{\omega_p^2}{\omega^2}$$

$$\omega^2(q) = \omega_p^2/2 + c^2 q^2 - \sqrt{\omega_p^4/4 + c^4 q^4}$$

(A)



Plasmon excitation by electron scattering



$$\dot{W} = \frac{e^2}{4\pi^2 \epsilon_0 v} \left\{ \log \left[\left(\frac{\omega}{v} \right)^2 + q_{\perp}^2 \right] \right\} \Big|_0^{q_c} \int \omega d\omega \text{Im} \left\{ \frac{-1}{\epsilon(\omega)} \right\}$$

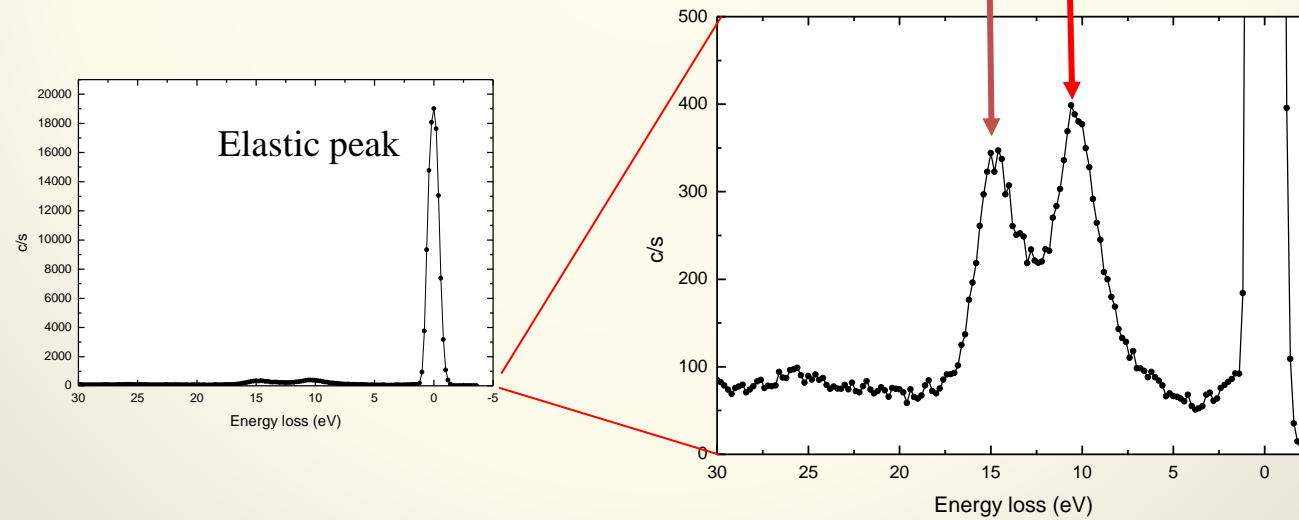
Bulk Loss function:

$$\text{Im} \left\{ \frac{-1}{\epsilon(\omega)} \right\} = \frac{\epsilon_2(\omega)}{\epsilon_1^2(\omega) + \epsilon_2^2(\omega)}$$

Surface Loss function:

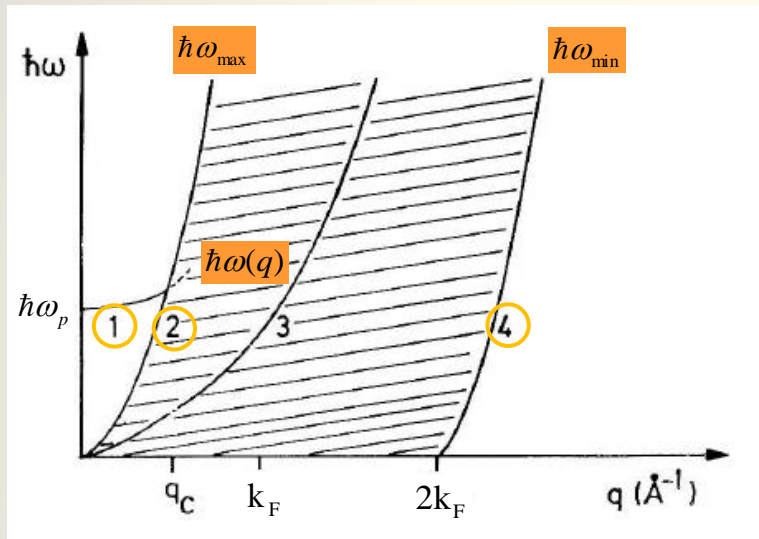
$$\text{Im} \left\{ -\frac{1}{\epsilon(\omega) + 1} \right\} = \frac{\epsilon_2(\omega)}{[\epsilon_1(\omega) + 1]^2 + \epsilon_2^2(\omega)}$$

Al(100) Reflection Energy Loss Spectrum



Plasmon excitation by electron scattering

Il processo di eccitazione di un plasmone da parte di un fascio incidente di elettroni è in competizione con l'eccitazione di un singolo elettrone all'interno di un solido



$$\mathbf{k}_f = \mathbf{q} + \mathbf{k}$$

$\hbar\omega, \hbar\mathbf{q}$ impulso ed energia persi dall'elettrone esterno
 \mathbf{k}, \mathbf{k}_f impulso iniziale e finale dell'elettrone nel solido

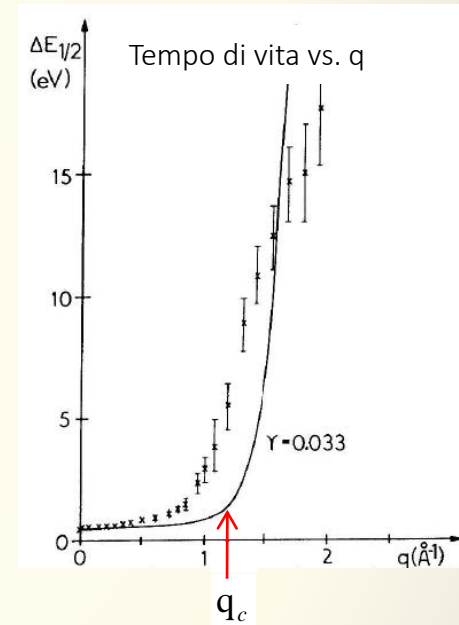
$$\hbar\omega = \underbrace{\frac{\hbar^2}{2m}(\mathbf{q} + \mathbf{k})^2}_{E \text{ finale}} - \underbrace{\frac{\hbar^2}{2m}k^2}_{E \text{ iniziale}} = \frac{\hbar^2}{2m}(q^2 + 2\mathbf{q} \cdot \mathbf{k})$$

$$\textcircled{4} \quad \hbar\omega_{\min} = \frac{\hbar^2}{2m}(q^2 - 2qk_F) \quad \textcircled{2} \quad \hbar\omega_{\max} = \frac{\hbar^2}{2m}(q^2 + 2qk_F)$$

Relazione di dispersione per il plasmone

$$\hbar\omega(q) = \hbar\omega_p \left(1 + \frac{3v_F^2}{10\omega_p^2} q^2\right)$$

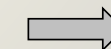
1



Stima di q_c ovvero quando la curva di dispersione del plasmone è degenera con l'eccitazione di singolo elettrone

$$\frac{\hbar^2}{2m}(q^2 + 2qk_F) = \hbar\omega_p \left(1 + \frac{3v_F^2}{10\omega_p^2} q^2\right)$$

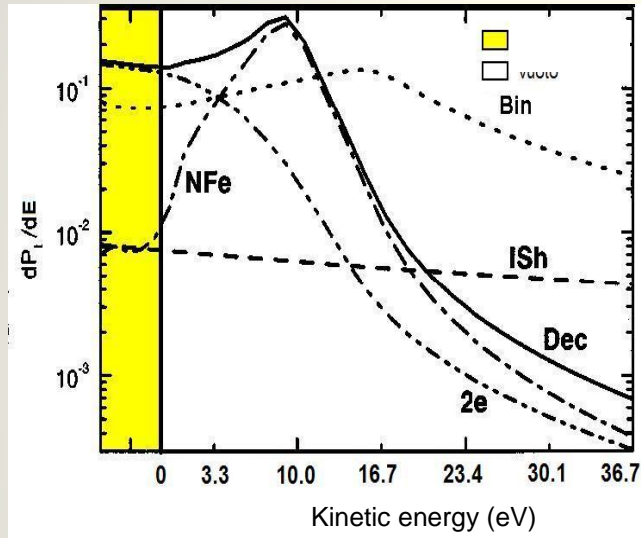
trascurando i termini in q^2



$$q_c = \frac{\omega_p}{v_F} \approx 1 \text{ \AA}^{-1}$$

Decay channels of plasma oscillations

Calculation of secondary emission after the decay of volume plasmon in Aluminum



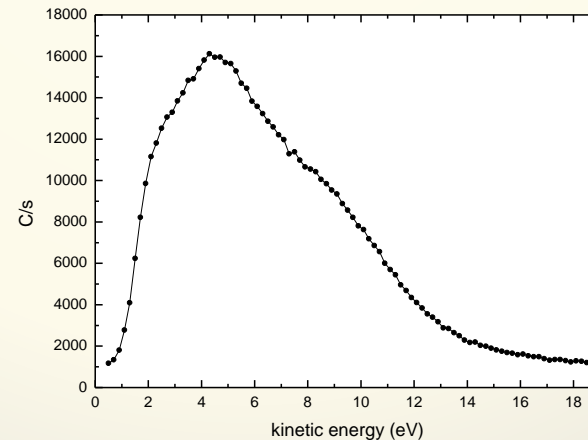
Bocan et al. Phys. Rev. A **69** (2004) 012901

- Two interacting electrons (2e) is important only at low energy
- The dominant process is the interband transition (NFe)

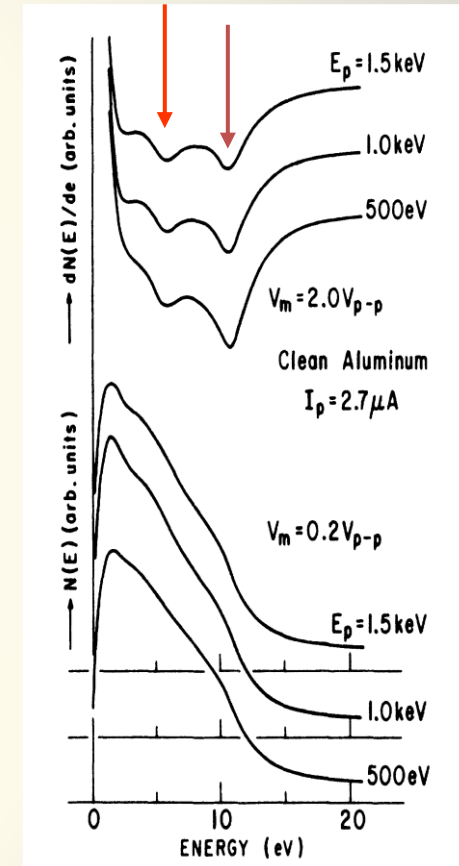
$$\hbar\omega_p \approx 5 \div 20 \text{ eV}$$

- understand how plasmons decay
- literature: plasmon transfer its energy and momentum to an electron of the solid
- emission of an electron in the secondary region (plasmon energy: few eV up to 20 eV)

Al(100) Secondary electron energy region

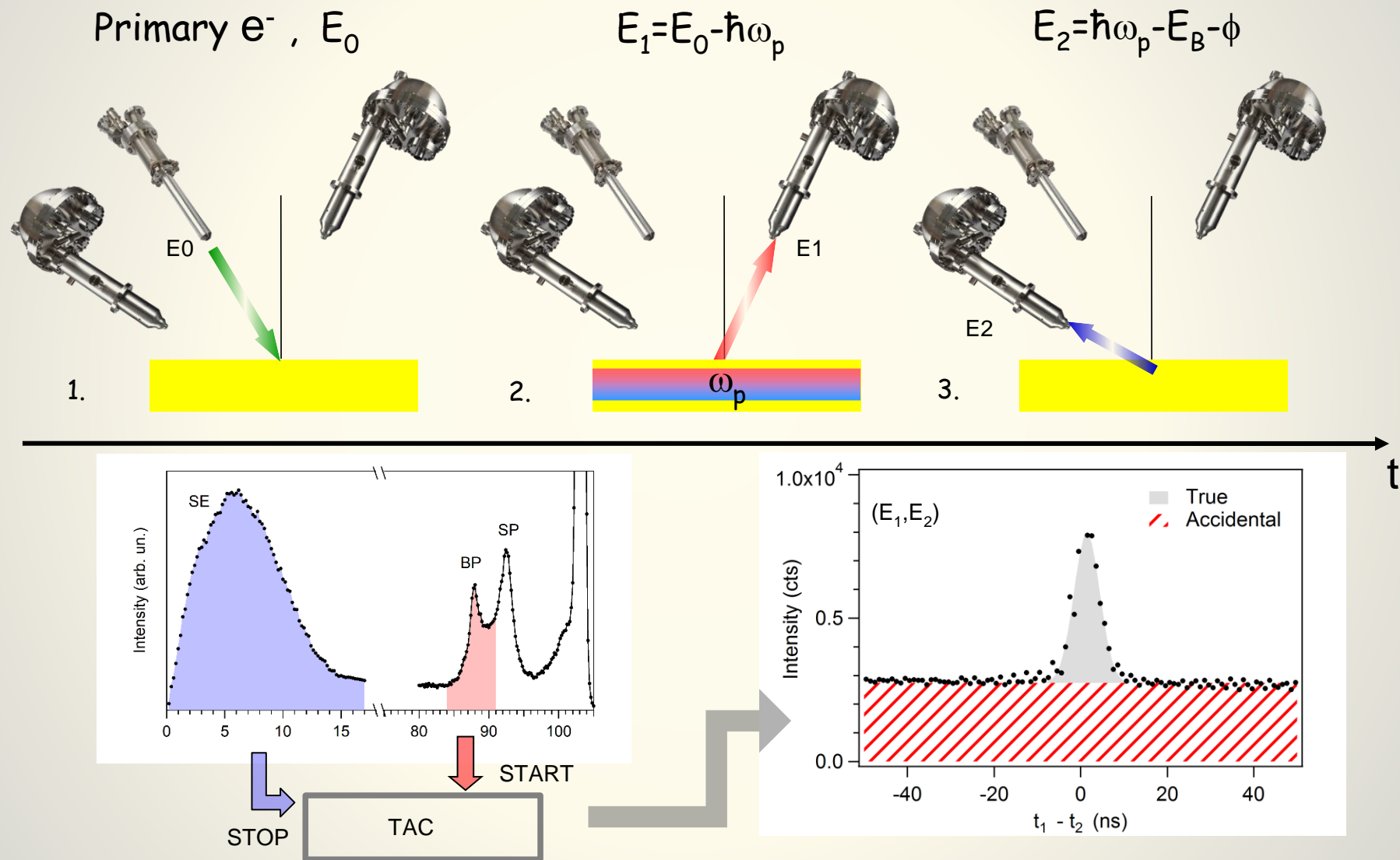


surface bulk



Chung and Everhart
Phys. Rev. B **15** (1977) 4699

Electron-electron coincidence spectroscopy



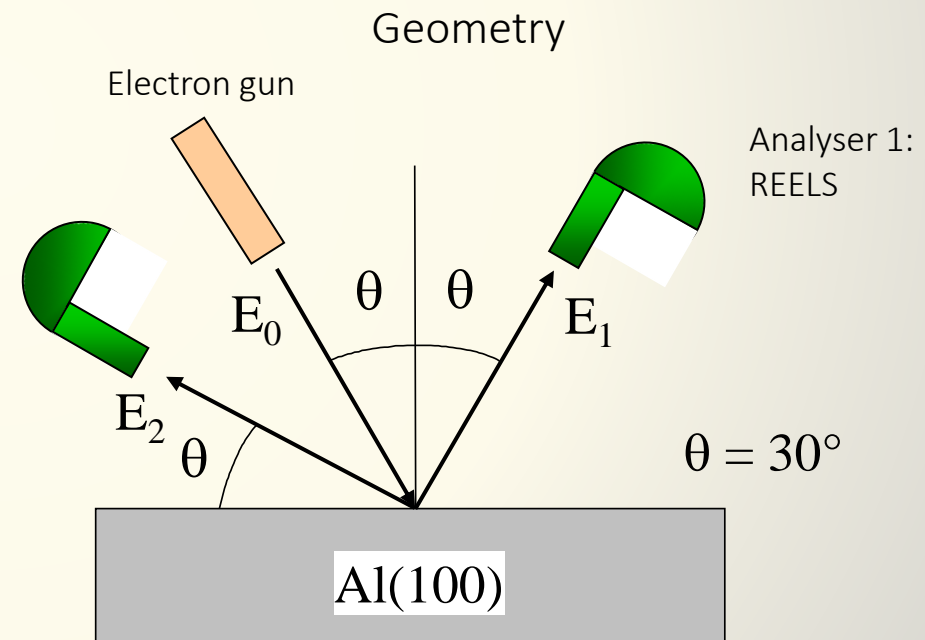
Experimental apparatus



LASEC laboratory
Department of Physics
University of Roma Tre

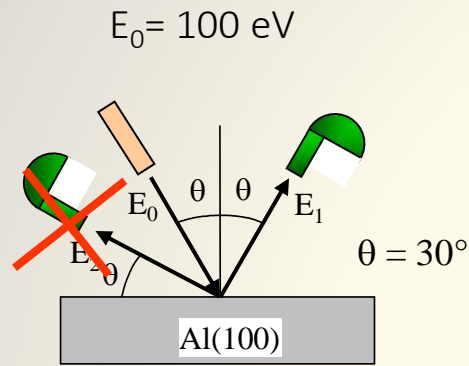
Analyser 2:
secondary
electrons

- UHV apparatus (base pressure $2 \cdot 10^{-10}$ mbar)
- Al(100) prepared each day by sputtering and annealing
- Magnetic field compensated by Helmutz coil

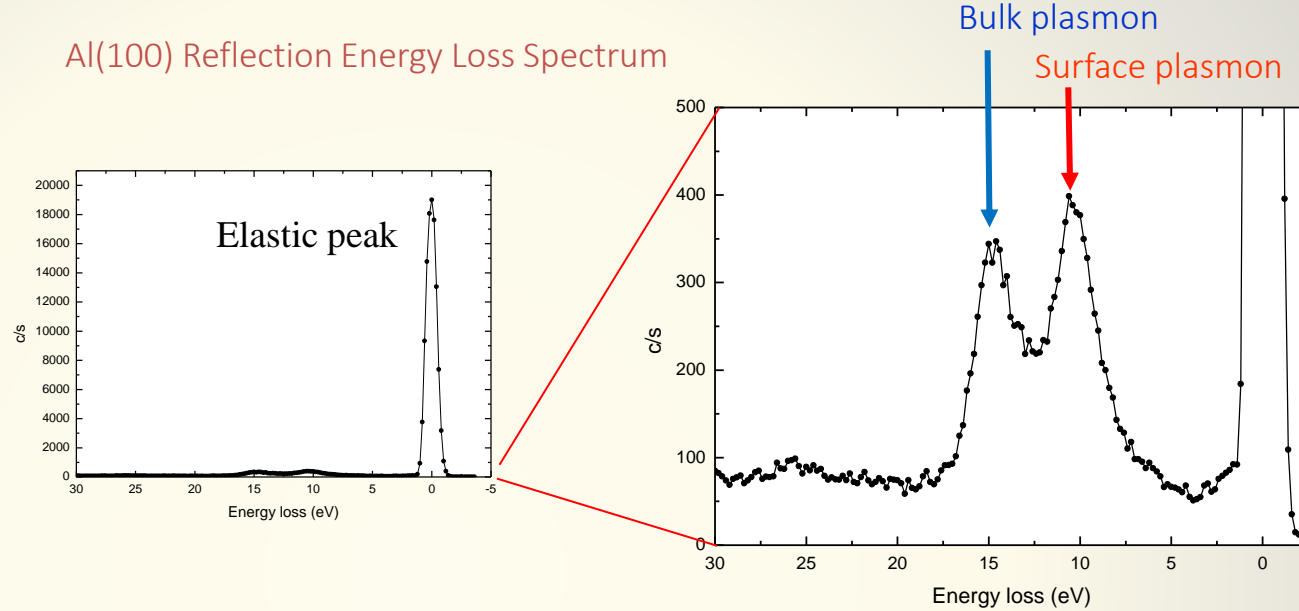


In this geometry plasmon is created with a well defined q not far from zero

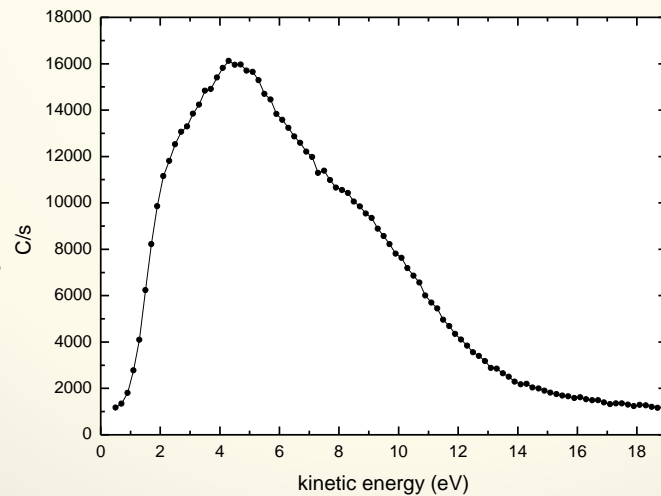
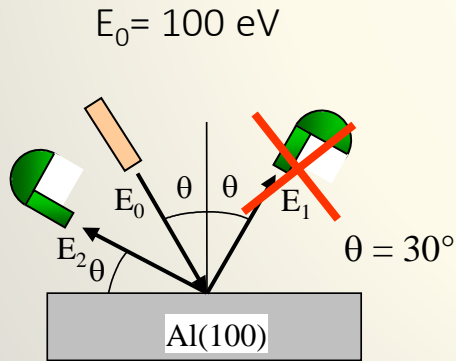
Single channel: REELS and secondary electron region



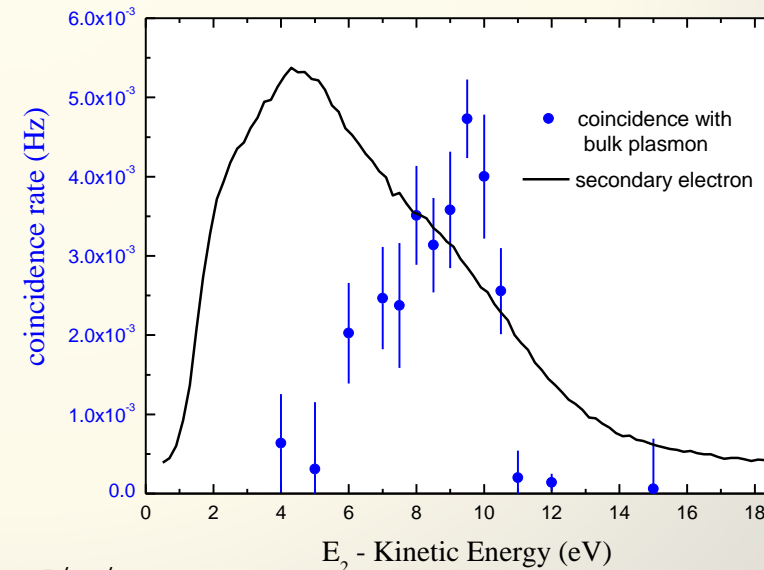
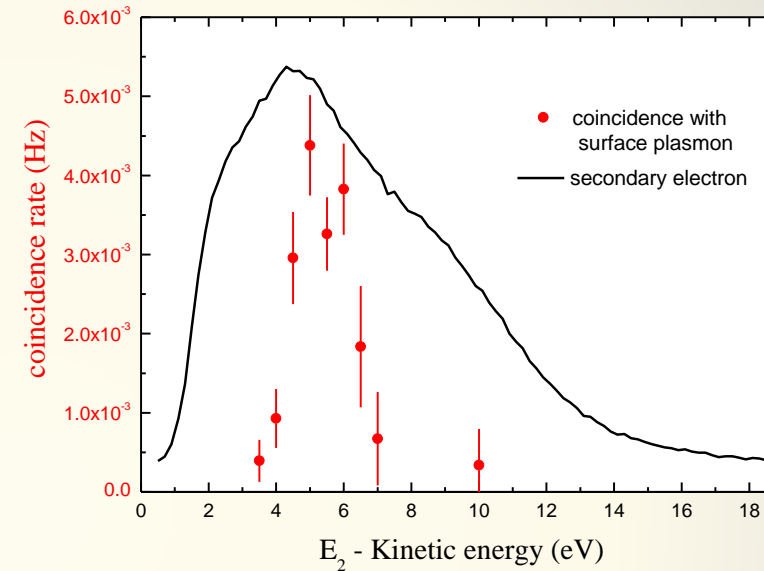
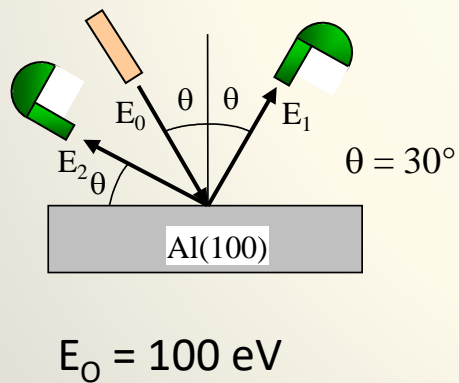
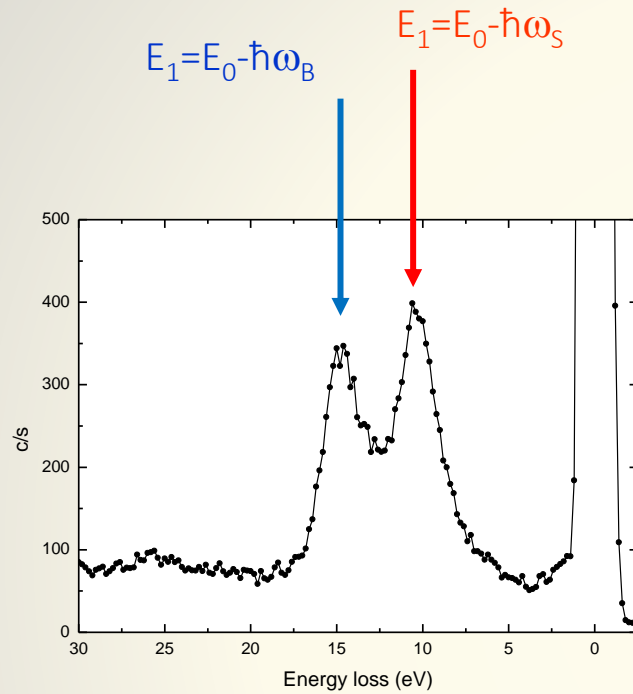
Al(100) Reflection Energy Loss Spectrum



Al(100) Secondary electron energy region



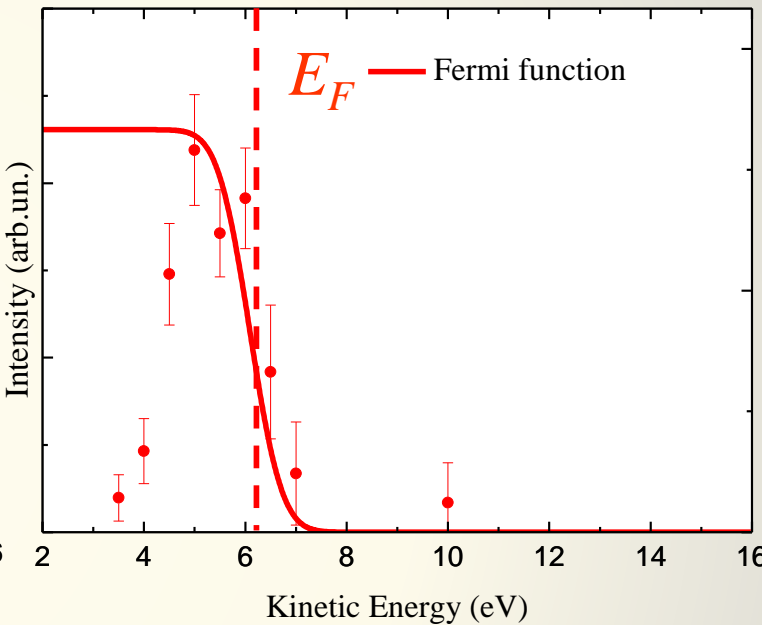
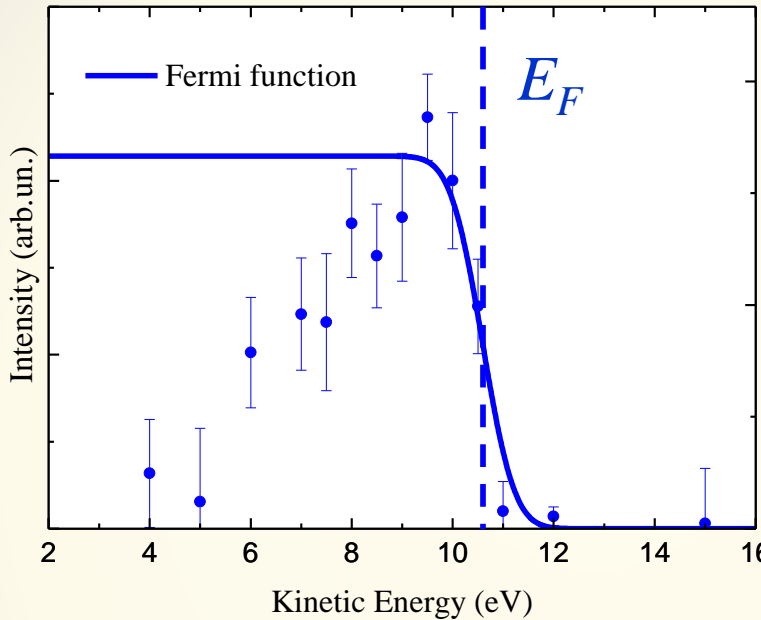
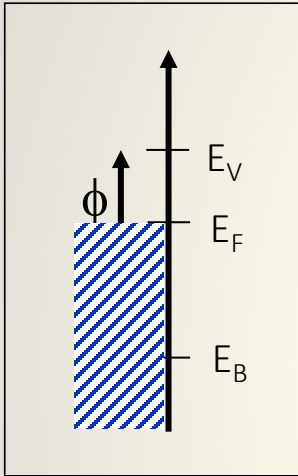
Coincidence: decay of the bulk and surface plasmon in Al(100)



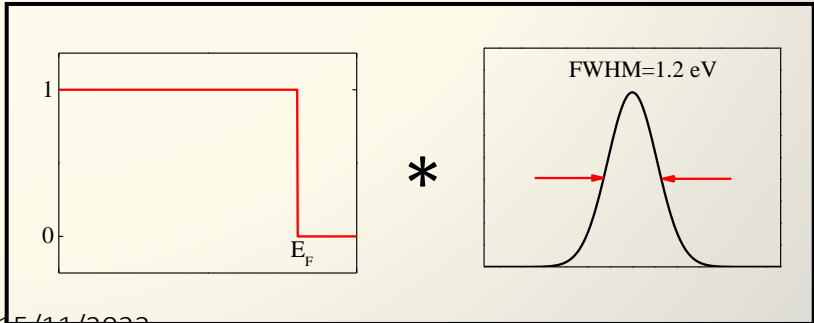
Plasmon-emission

$$E_F = \hbar\omega_{BP} - \Phi = 10.6 \text{ eV}$$

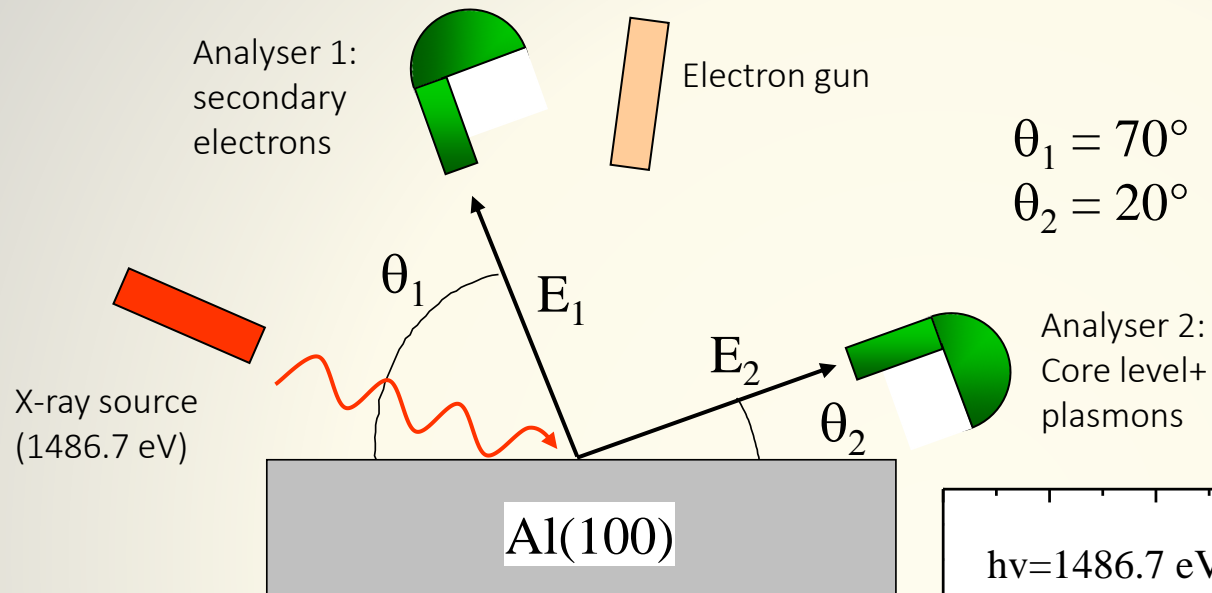
$$E_F = \hbar\omega_{SP} - \Phi = 6.1 \text{ eV}$$



The Fermi energy is derived from the experiment: the plasmon energy from EELS and the analyser work function from independent measurements (photoemission)

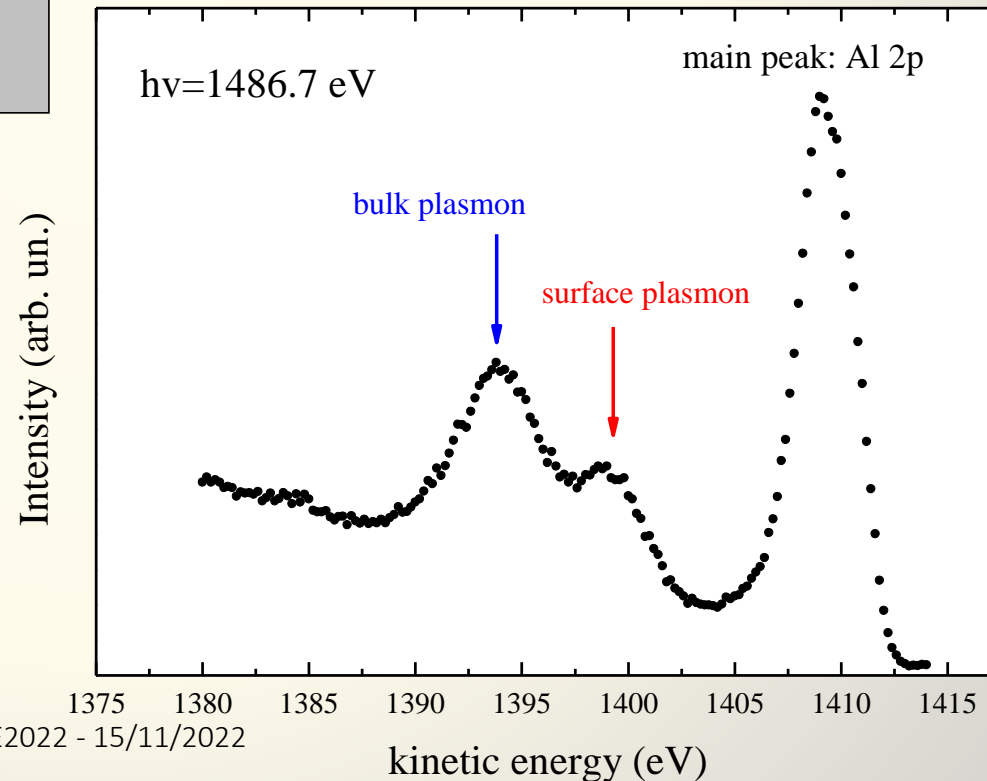


Plasmons decay in ($\gamma, 2e$) experiment: the role of the band structure

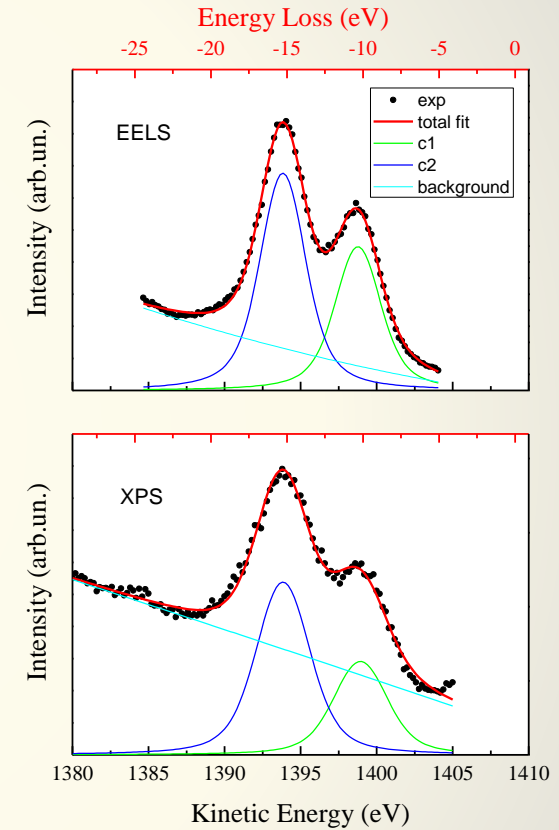
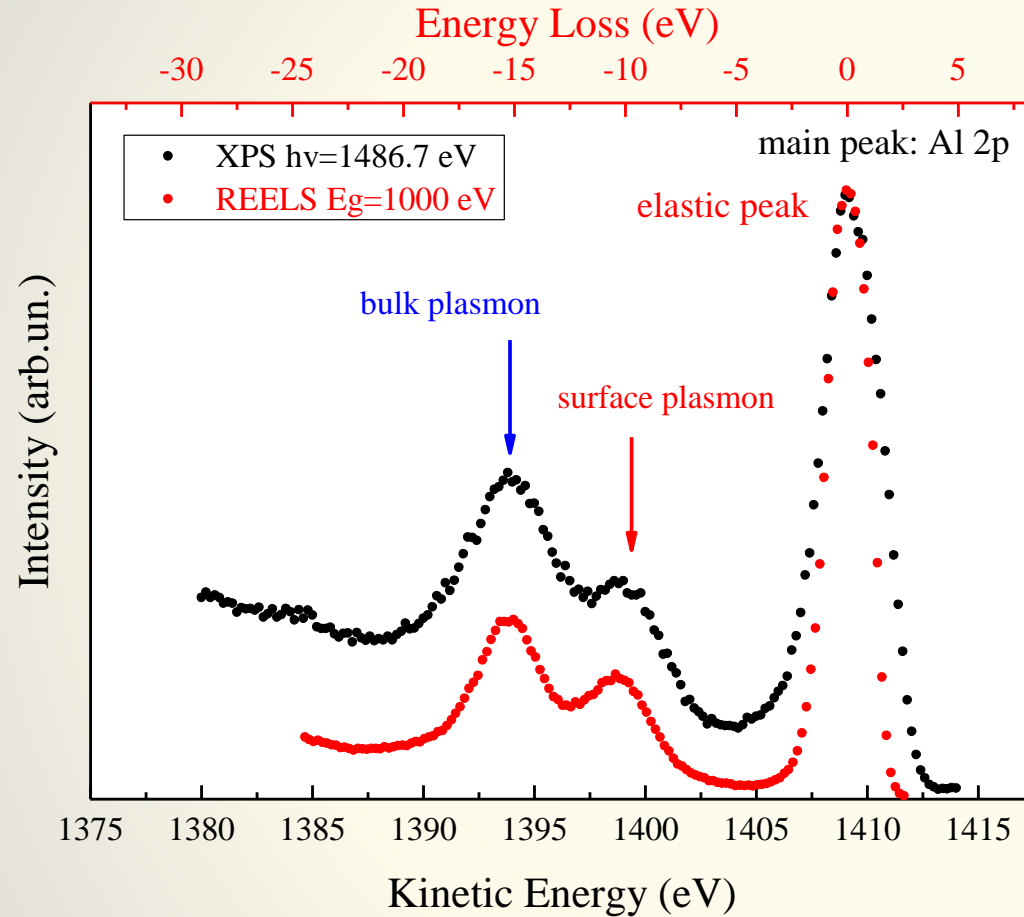


X-ray photoemission and plasmon creation, two possible channels:

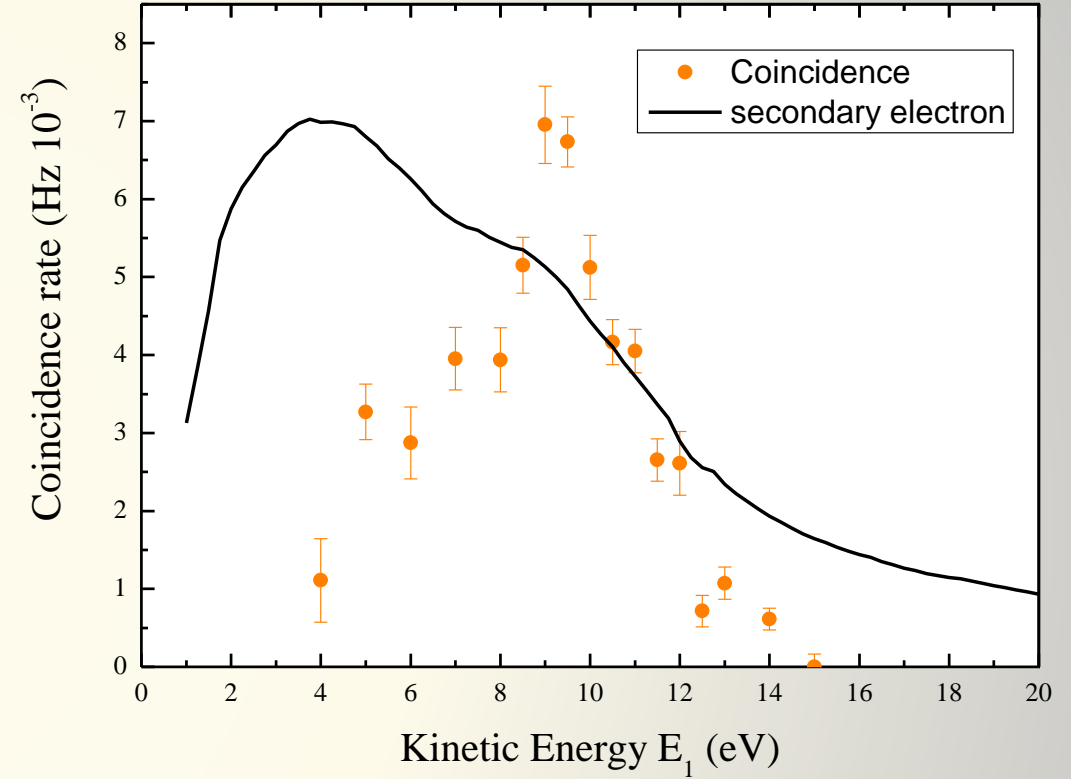
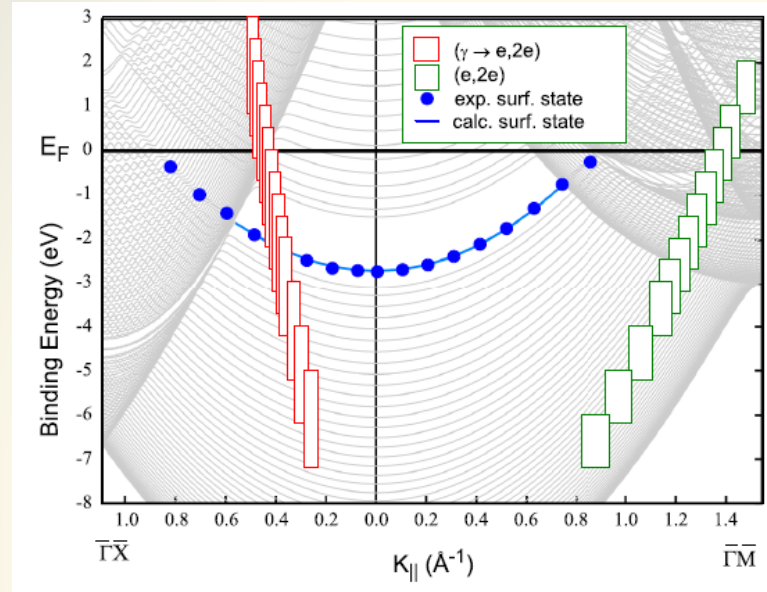
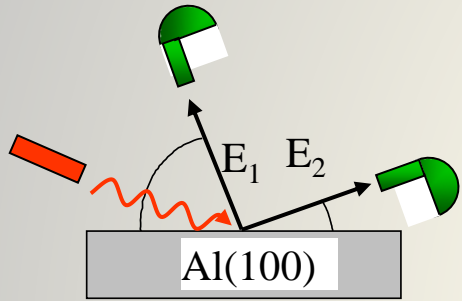
- 1) intrinsic plasmon
- 2) extrinsic plasmon



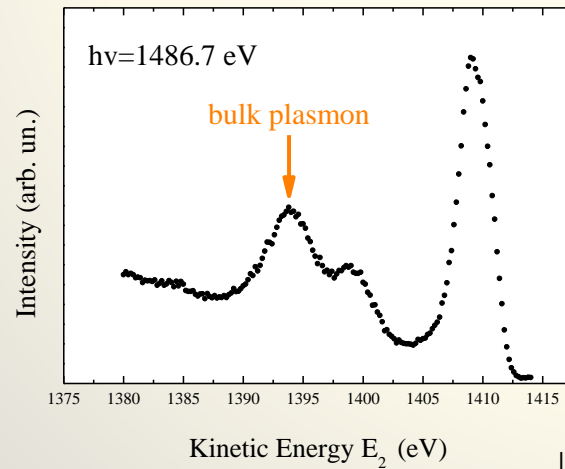
Very similar EELS and XPS spectra



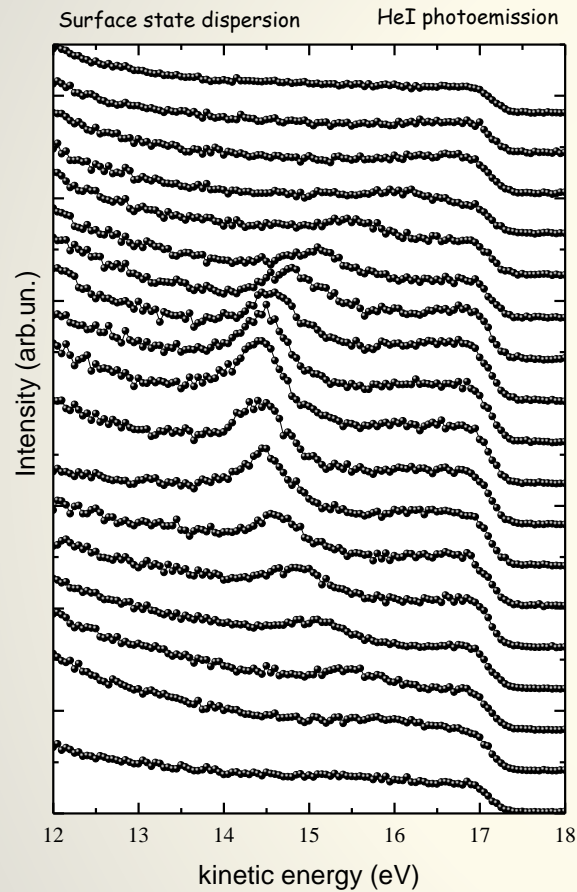
$(\gamma, 2e)$ coincidence: decay of the bulk plasmon



E_2 fixed at bulk plasmon excitation energy

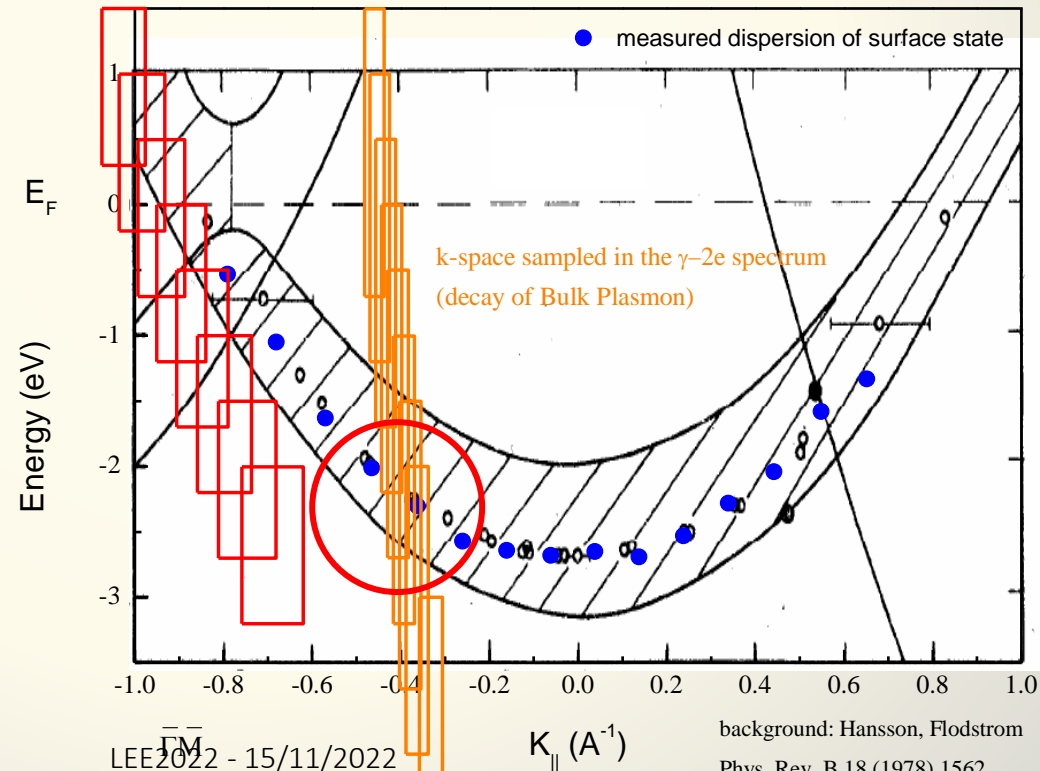
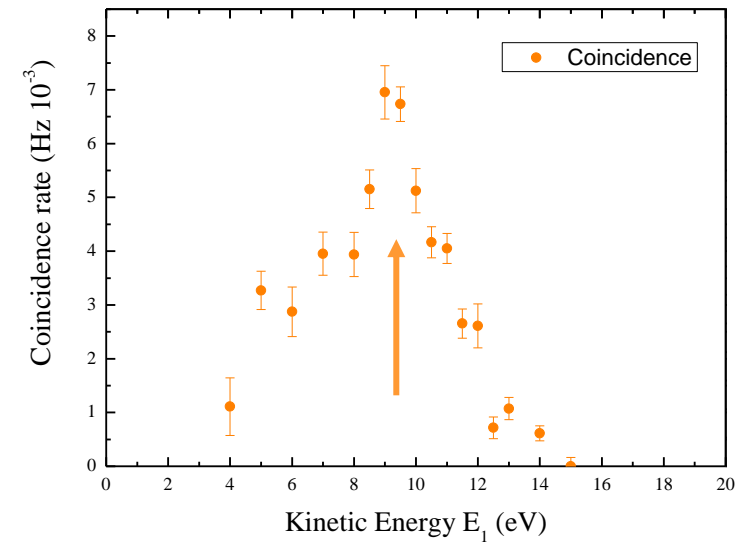


Sampled K-space in coincidence spectrum

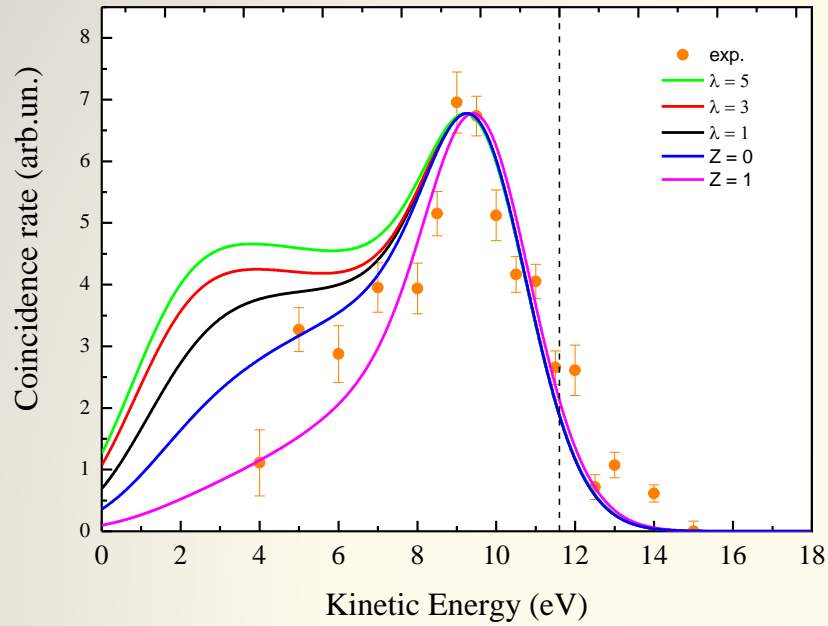


$$E_k = \hbar\omega_p - E_B - \Phi$$

$$\mathbf{k}_{\parallel}^f = \mathbf{k}_{\parallel}^i + \mathbf{k}_{\parallel}^p$$

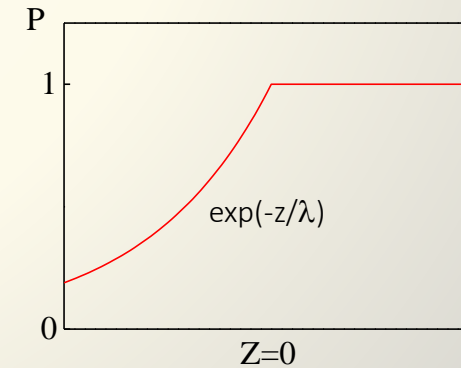
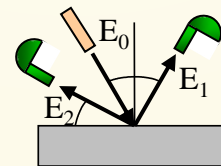
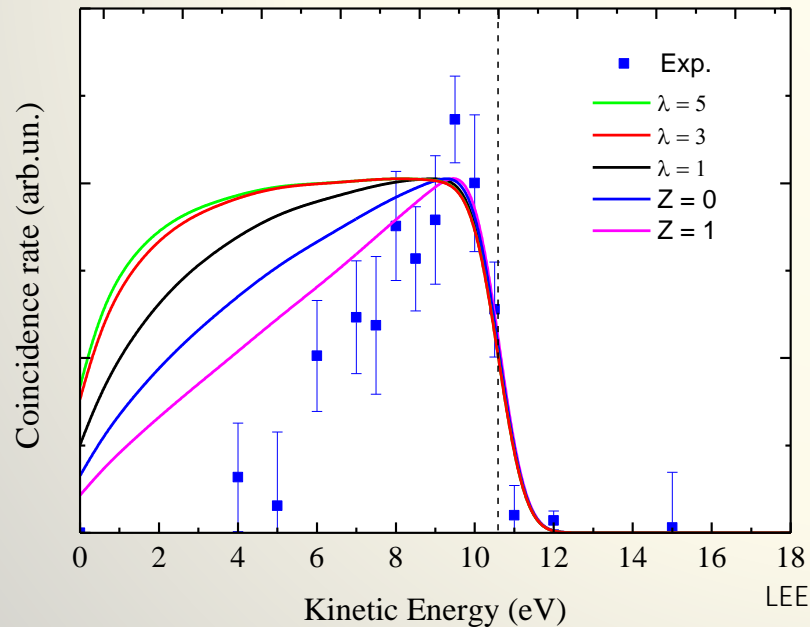
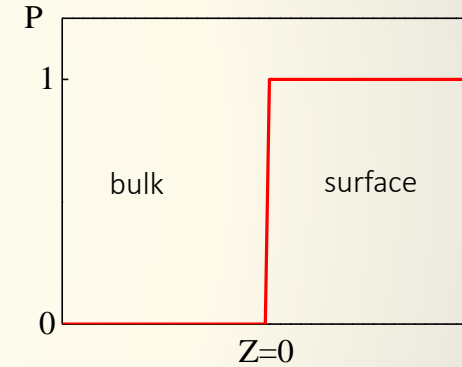
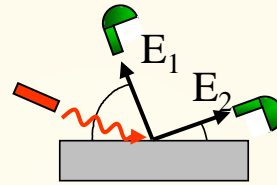


Comparison with calculated density of state



Calculation:

- bulk DOS (jellium) + surface state @ selected K
- refraction of outgoing electron
- experimental broadening
- probing depth



Conclusions

- Experimental evidence of the decay of bulk and surface plasmons in the region of secondary electron
- large contribution of surface plasmon decay to secondary region
- Agreement with theory seems good but better refinement and check are needed
- Interband transition not assisted by other particle is the dominant decay channel (Plasmo-emission)
 - plasmon with fixed momentum
 - lineshape of the emitted electrons

In collaboration with:

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F. Offi	RomaTre
D. Sbaraglia	RomaTre
G. DiFilippo	Roma Tre
S. Iacobucci	ISM-CNR
W. Werner	TU Wien
W. Smekal	TU Wien