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Electronic excitations in thin metallic films

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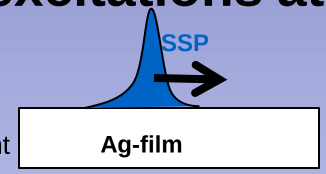
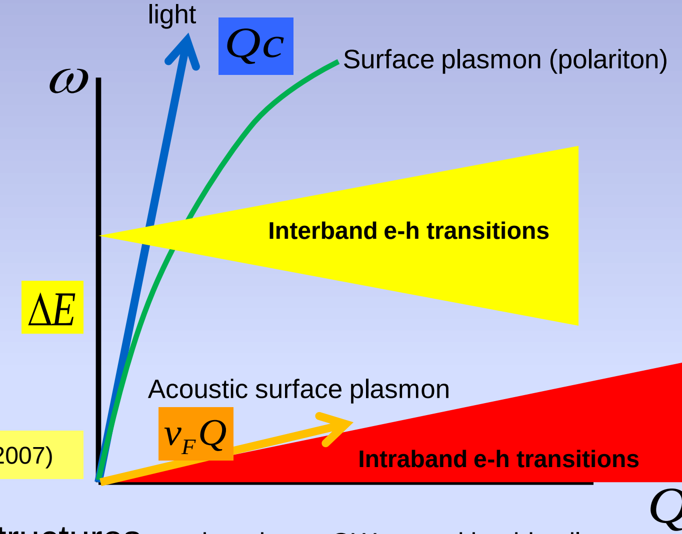
1. Introduction and motivation

Why is proper knowledge of the electronic excitations at metallic surface important?



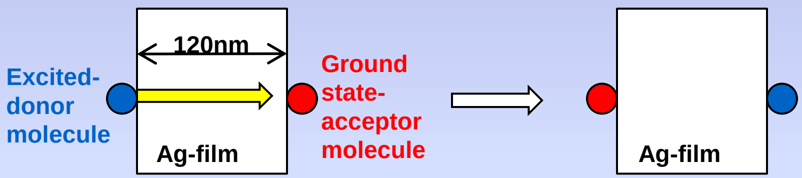
1.Plasmonic circuits-guided surface plasmons (polariton) could be subject to significant Landau damping.

Berni P. et al, J. Appl. Phys. **98** 043109 (2005)



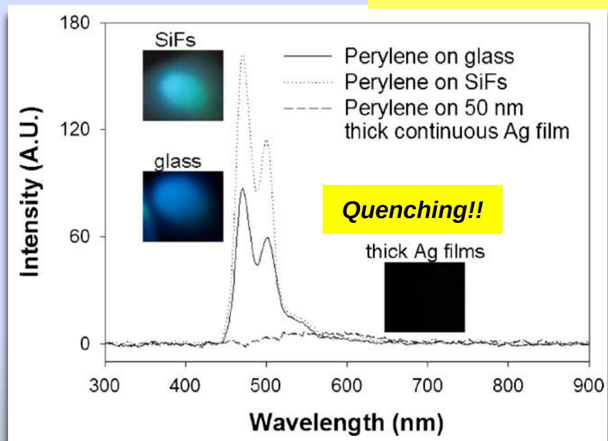
2.Plasmonic devices-thin silver film covered from both sides by thin polimer films with molecular chromophores. **ENERGY TRANSFER ACROSS A METAL FILM MEDIATED BY SURFACE PLASMON POLARITONS**

Andrew P. and Barnes W. L., Science **306** 1002 (2004)



3.Biosensing –Metal-enhanced fluorescence: Surface plasmon can radiate a Fluorophore' s structured emission.

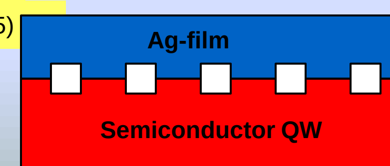
Yongxia Zhang, *et al* , Appl. Phys. Lett. **90,**053107 (2007)



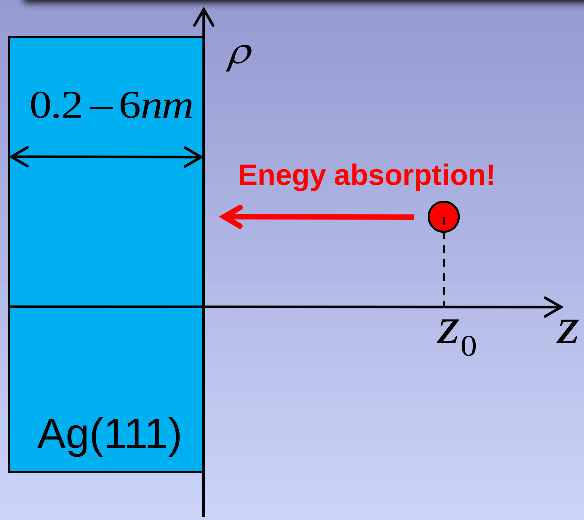
4.Light-emitting structures semiconductor QW coated by thin silver films increse spontaneous emission in QW. Efficient energy transfer from electron-hole pair recombination (in QW) to a surface plasmon (in silver film). Energy is converted into light by using corrugated metal-semiconductor interface.

A. Neogi, *et al*, Phys. Rev. **B 66**, 153305 (2002)

K. Okamoto, *et al*, Appl. Phys. Lett. **87** 071102 (2005)



2. Mathematical formulation and modeling of the system



Sudden created point charge near Ag(111) surface,
V.M. Silkin, Poster **90**

$$V^{ext} = \frac{2\pi e}{Q} e^{-Q|z-z_0|} \cos(\omega t)$$

$$P_{loss}(Q,\omega) \propto \mathrm{Im} W^{ind}(Q,\omega,z_0,z_0)$$

$$W^{ind}(Q,\omega_0,z_0,z_0) = e^{-2Qz_0} g(Q,\omega)$$

Surface response function

$$g(Q,\omega) = \int dz_1 dz_2 e^{2Q(z_1+z_2)} R(Q,\omega,z_1,z_2)$$

$-\mathrm{Im}\, g(Q,\omega)$ **Intensity of the real excitations in the system!**

Response function-RPA

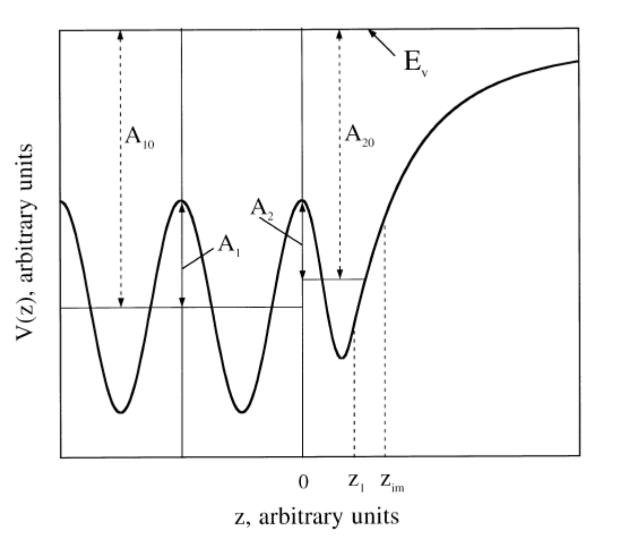
$$R = \frac{R_0}{1 - vR_0}$$

Free electron response function

$$R^0\left(\mathbf{Q},\omega,z,z'\right)=\sum_{n,m,\mathbf{K}}\frac{f\left(E_{n,\mathbf{K}}\right)-f\left(E_{m,\mathbf{K+Q}}\right)}{\omega-\left(E_{m,\mathbf{K+Q}}-E_{n,\mathbf{K}}\right)+i\eta}\Phi_n\left(z\right)\Phi_m\left(z\right)\Phi_n\left(z'\right)\Phi_m\left(z'\right)$$

Band structure and wave function-**1D model potential**

E.V. Chulkov, V. M. Silkin and P.M. Echenique, Surf.Sci**. 437** (1999) 330-352



Parameters of the potential are set up to fit experimental (first principle) datas for surface state, first image state and postion and width of band gap at $\overline{\Gamma}$ point

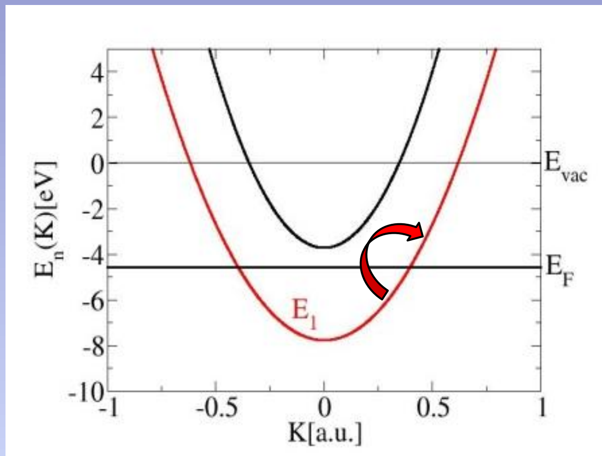
Band structure and wave functions- solutions of 1D Schrodinger equation:

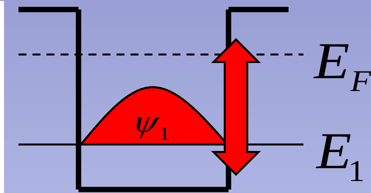
$$\left[-\frac{\hbar^2}{2m^*}\frac{d^2}{dz^2}+V_{MP}\left(z\right)\right]\Phi_n\left(z\right)=E_n\Phi_n\left(z\right)$$

$$E_{n,\vec{K}}=E_n+\frac{\hbar^2K^2}{2m^*}$$

3. Excitation spectra inside ultra thin silver films

1ML(0.2nm)- Ag(111)





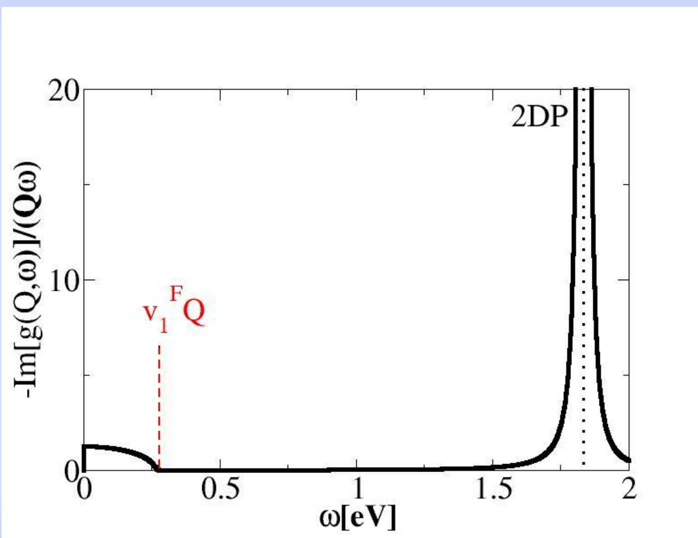
1ML-one quasi 2D electron gas

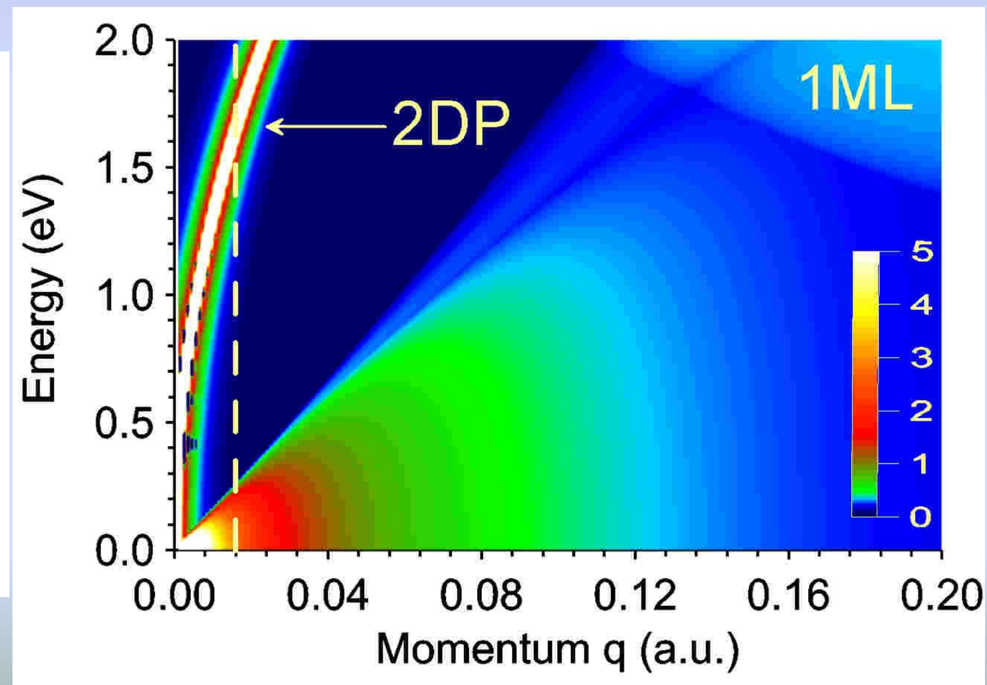
$$\varepsilon_{eff}^{2D}(Q,\omega)=1-V_Q R_1^{2D}(Q,\omega)$$

$$V_Q=v_Q\int_{-\infty}^{\infty} dz_1 dz_2 \psi_1^2(z_1) e^{-Q(z_1-z_2)}\psi_1^2(z_2)\xrightarrow{Q\to 0}\frac{2\pi e}{Q}$$

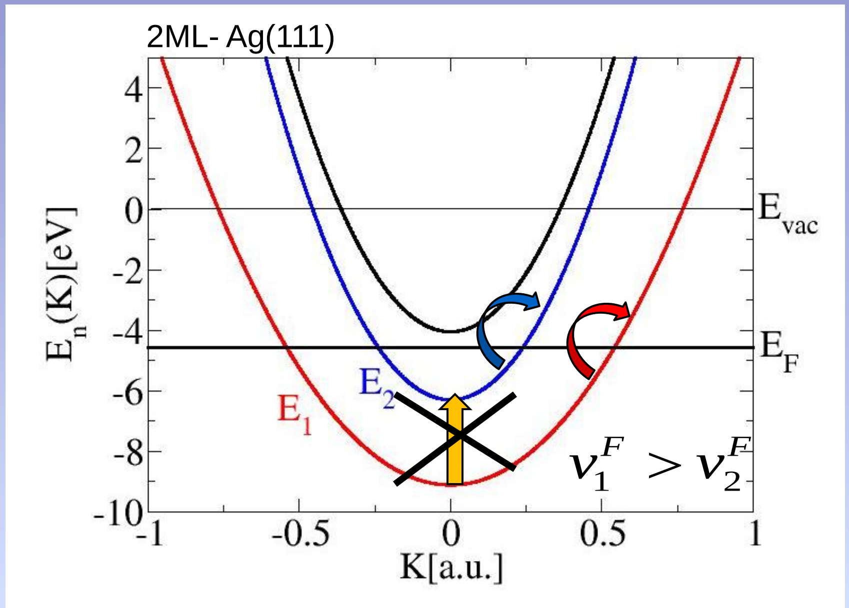
$$\mathrm{Im}\,\varepsilon_{eff}(Q,\omega)=0 \qquad \mathrm{Re}\,\varepsilon_{eff}(Q,\omega)=0$$

$$\omega_{2D}=\frac{k_1^F}{\sqrt{m^*}}\sqrt{Q}$$

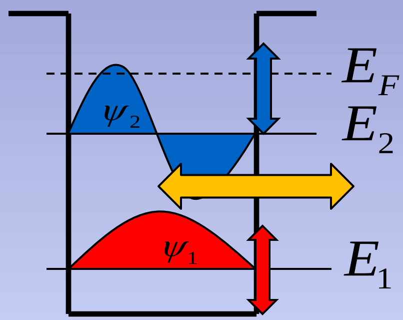




2ML(0.4nm)- Ag(111) film



2ML-two quasi 2D electron gases



Approximationes:
a) Longwavelenght limit $Q \to 0$
b) There are no transitions between bands E_1 **and** E_2

$$\varepsilon_{eff}^{2D}(Q,\omega) = 1 - v_Q\left[R_1^{2D}(Q,\omega) + R_2^{2D}(Q,\omega)\right]$$

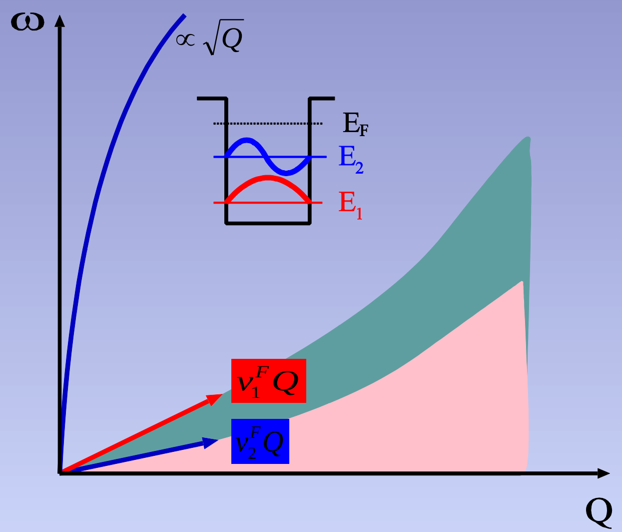
$$\omega_{ASP} = v_S Q \qquad v_S = \frac{2}{\sqrt{3}} v_1^F < v_2^F \qquad v_{1,2}^F = \sqrt{\frac{2}{m_{1,2}*}(E_F - E_{1,2})}$$

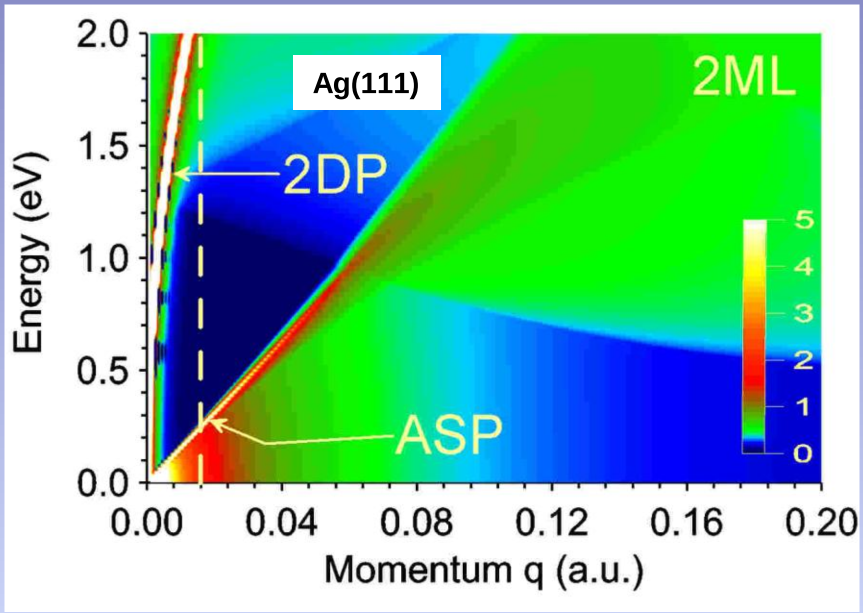
2ML-Two component quasi 2D electron plasma-Egzistence of fast and slow electron plasmas allow forming of Acoustic Surface Plasmon (ASP) . D. Pines, Can J. Phys. **34**, 1379 (1956)

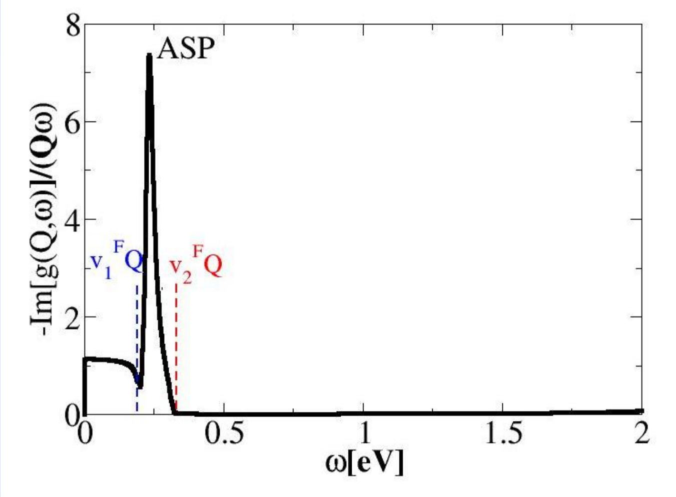
3ML, 4ML, . . . Multiplication of acoustic surface plasmon branches

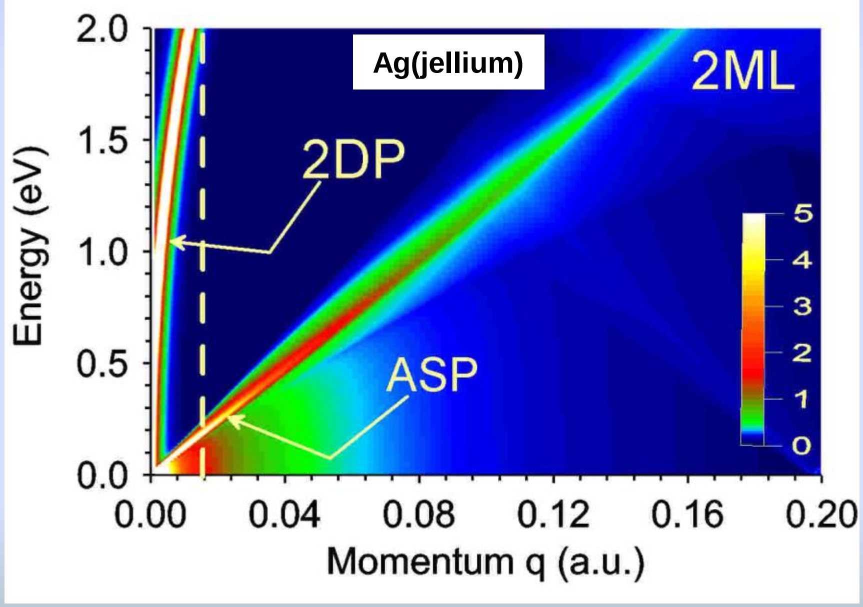
$$\omega_{ASP}^i = v_S^i Q \qquad v_1^F < v_S^1 < v_2^F < v_S^1 < v_2^F < ....$$

2ML(0.4nm)- Ag(111) film

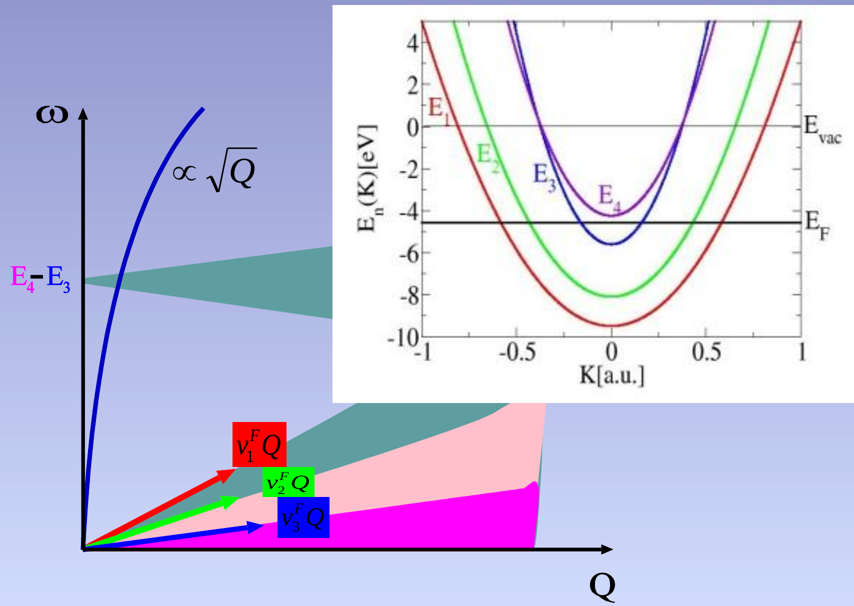


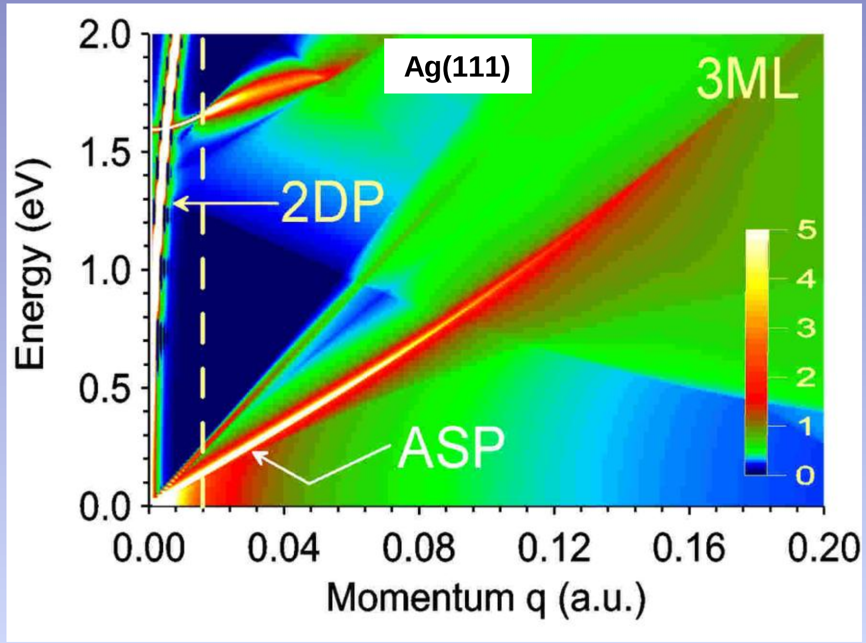


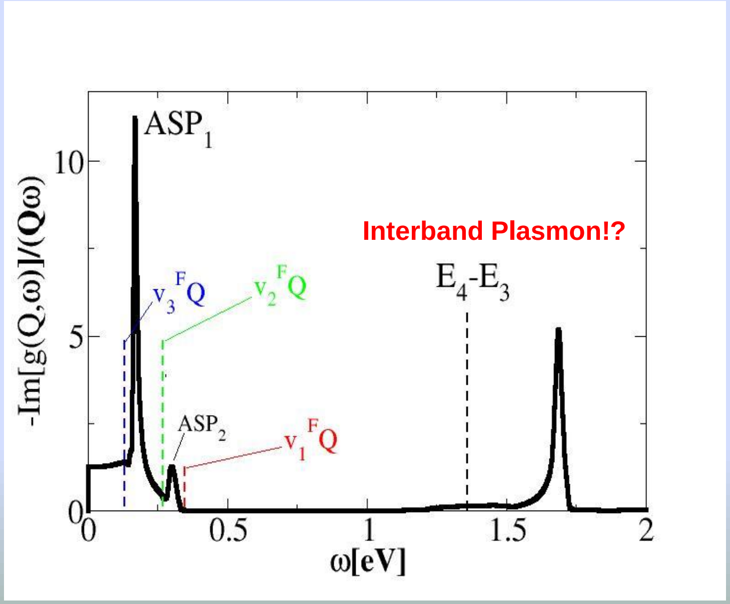


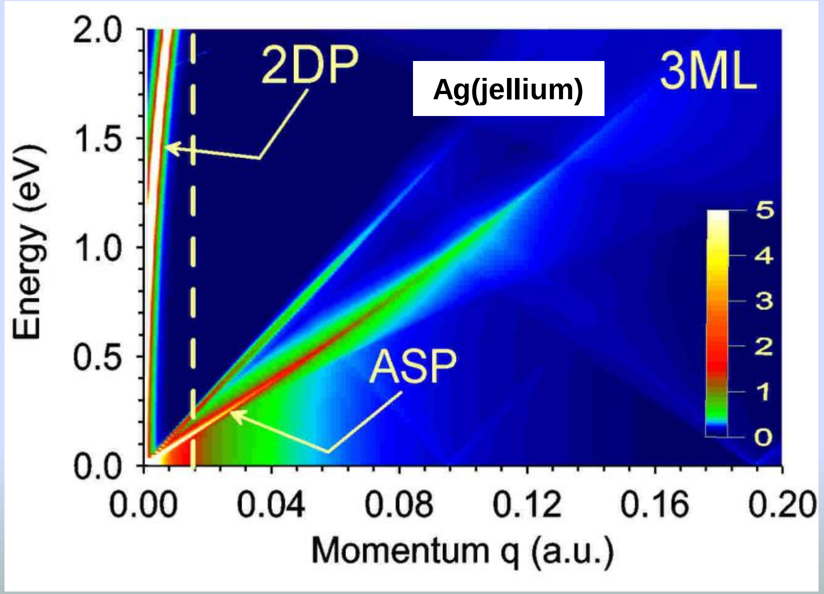


3ML(0.6nm)- Ag(111) film

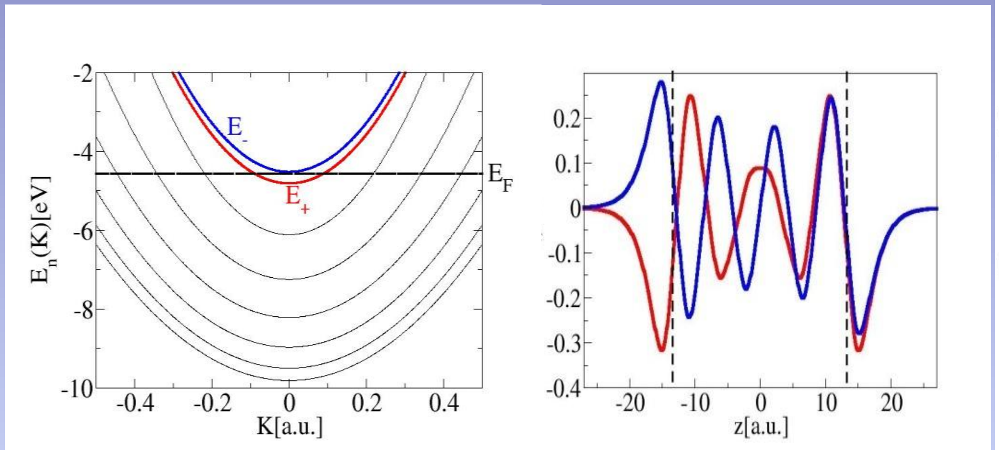


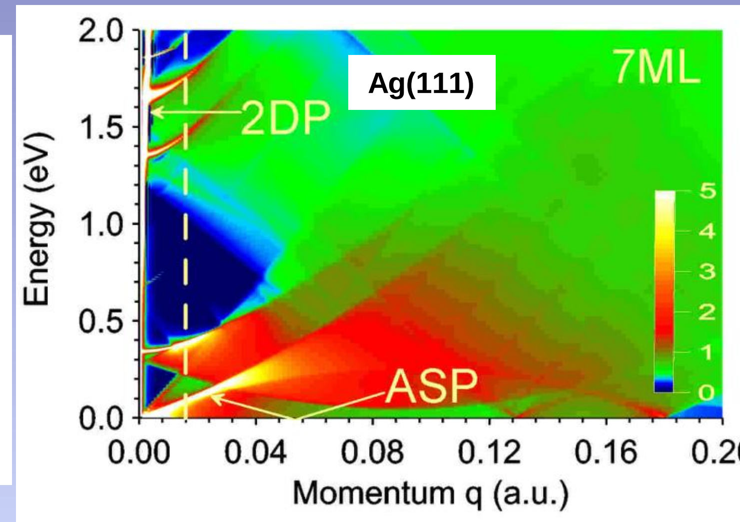


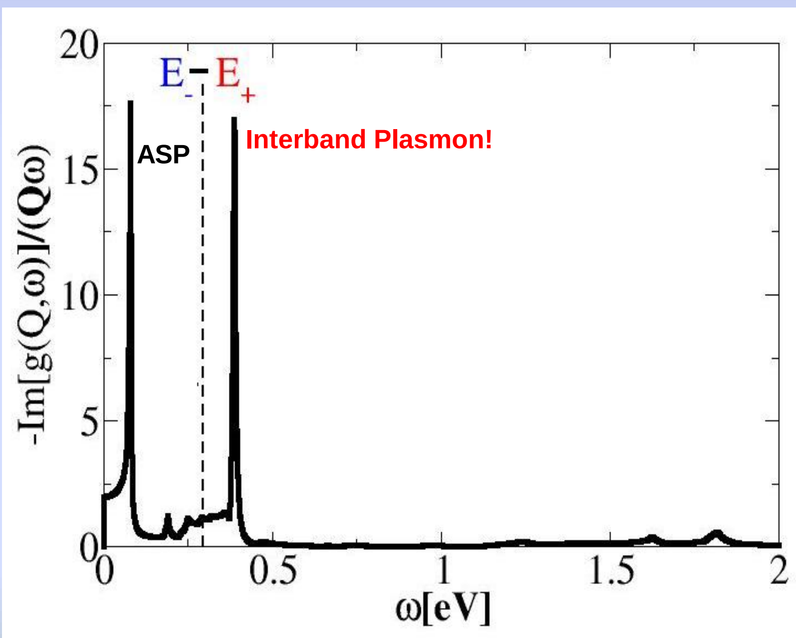


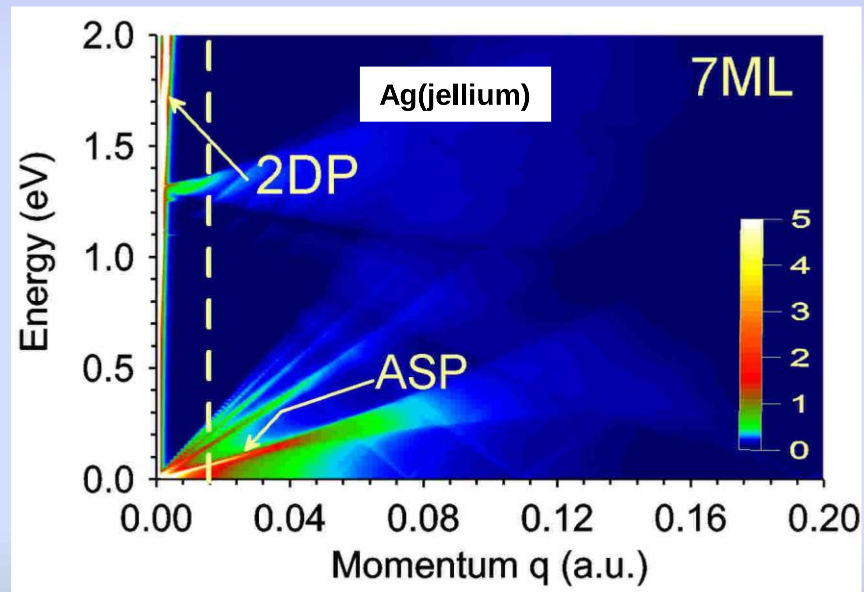


7ML(1.4nm)- Ag(111) film



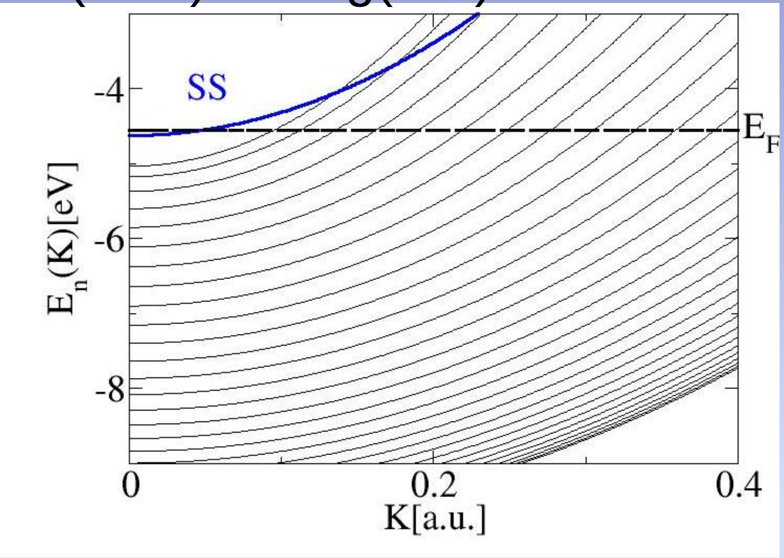


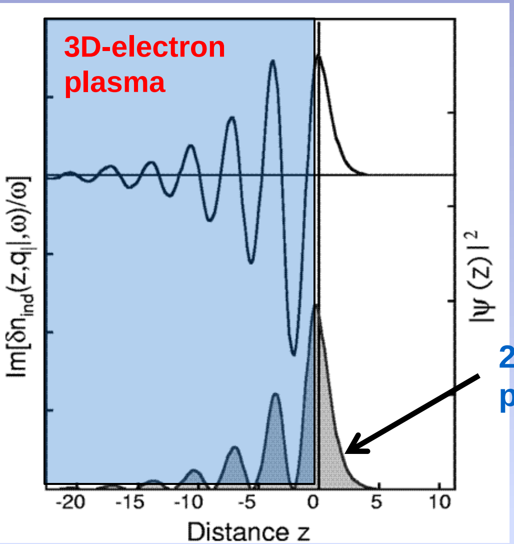




4. Excitation spectra inside thick silver films and transition to spectra in semiinfinite metals

31(6nm)ML- Ag(111) film





J. M. Pitarke, V. U. Nazarov, V. M. Silkin, E. V. Chulkov, E. Zaremba, and P. M. Echenique Phys. Rev. **B 70**, 205403 (2004)

V. M. Silkin ,J.M. Pitarke, E. V. Chulkov and P. M. Echenique Phys. Rev. **B 72**, 115435 (2005)

$$\varepsilon_{eff}^{2D}(Q,\omega)=1-W^{3D}(Q,\omega)R_{SS}^{2D}(Q,\omega)$$

Dispersion relation equation

$$\varepsilon_{eff}^{2D}(Q,\omega)=0$$

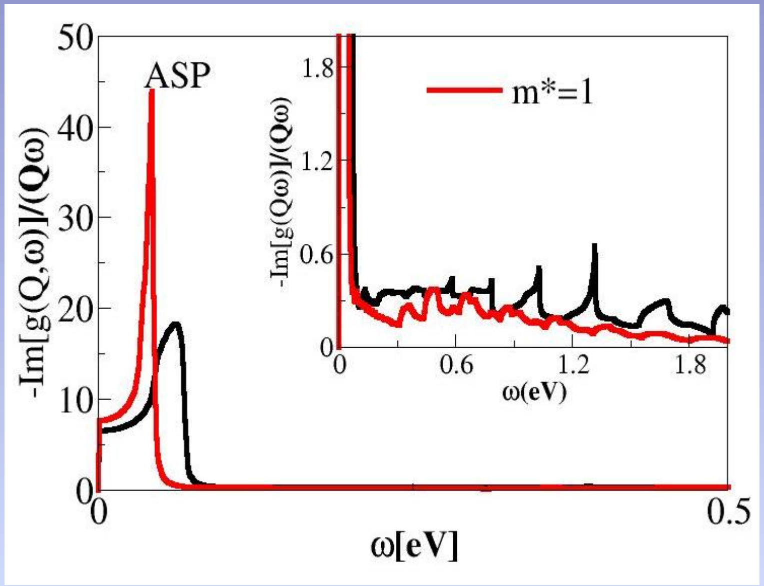
$$\omega_{ASP}=\alpha v_{SS}^{F}Q$$

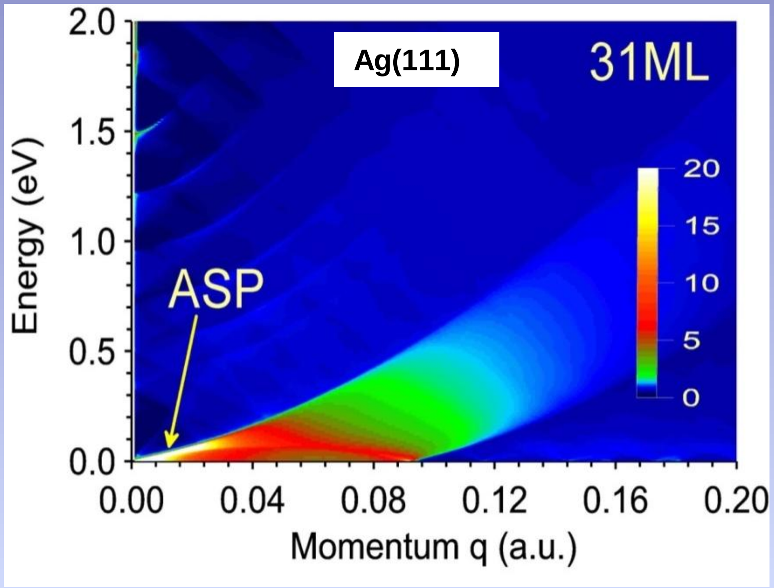
$$\alpha\approx 1$$

$$W^{3D}(Q,\omega)=\int_{-\infty}^{\infty}dz_1dz_2\psi_{SS}^2(z_1)W^{3D}(Q,\omega,z_1,z_2)\psi_{SS}^2(z_2)$$

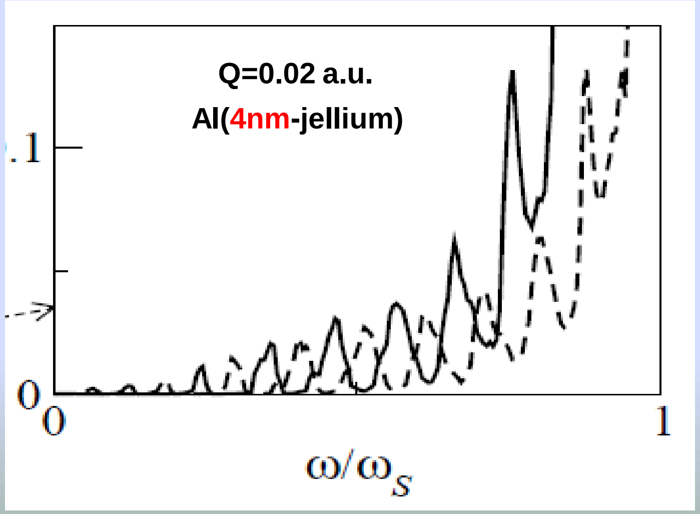
Without 3D electron plasma $W^{3D}(Q,\omega)\rightarrow v_Q=\dfrac{2\pi e}{Q}\Leftrightarrow$ **One** quasi 2D electron gas $\omega_{2D}\propto\sqrt{Q}$

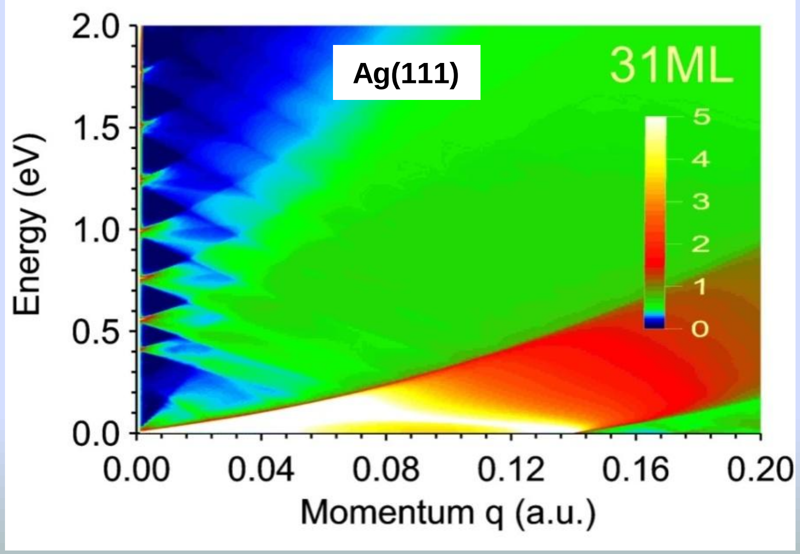
31(6nm)ML- Ag(111) film





L. Marušić, V. Despoja and M. Šunjić, J. Phys.: Condens. Matter **18** (2006) 4252

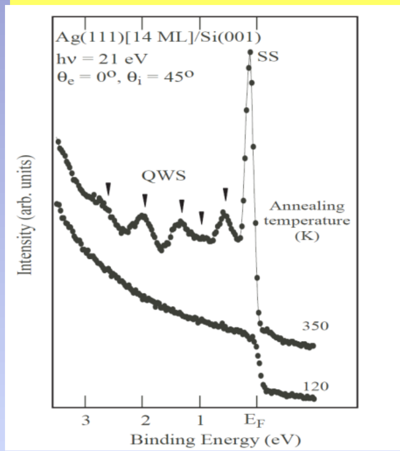


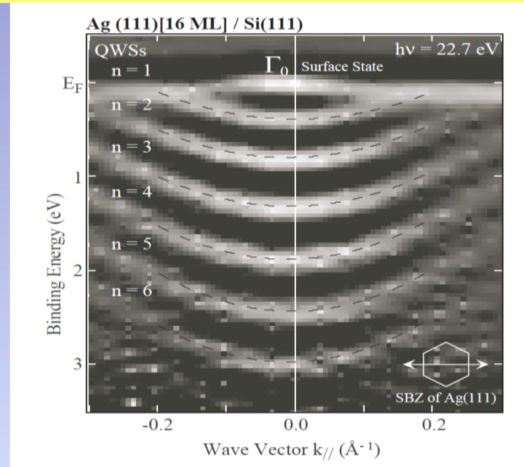


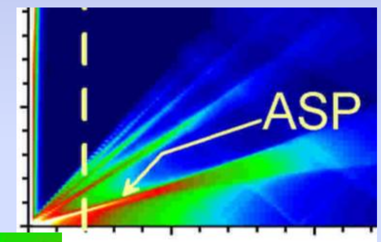
5. Conclusion

ARPES measurements of quantum size effects

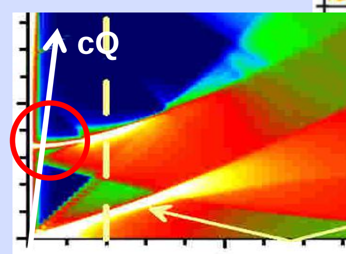
I. Matsuda *et al*, J. Surf. Sci. Nanotech. Vol. **2** (2004) 169-177







1.Ultra thin metallic films → quantum size effect → multicomponent 2D plasma → multiplication of ASP branches.



2.Thin metallic slabs → interband plasmon.
It would be nice to investigate how it couples to light.

3.Semiinfinite noble metals (111) surfaces.
Slow 2D plasma (surface state band) + fast 3D plasma (bulk state bands) =Acoustic Surface Plasmon

Thank you for your attention!