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Introduction to Surface Plasmon Theory

Jean-Jacques Greffet

Institut d'Optique Graduate School
Institut Universitaire de France, CNRS

Outline

A Plasmons

B Surface plasmons, description

C Near-field optics : fundamentals

D A few examples of surface plasmons

E Surface waves

Definition, Polarization properties, Dispersion relation, History of surface waves , Lateral wave.

F Surface plasmon polariton (SPP)

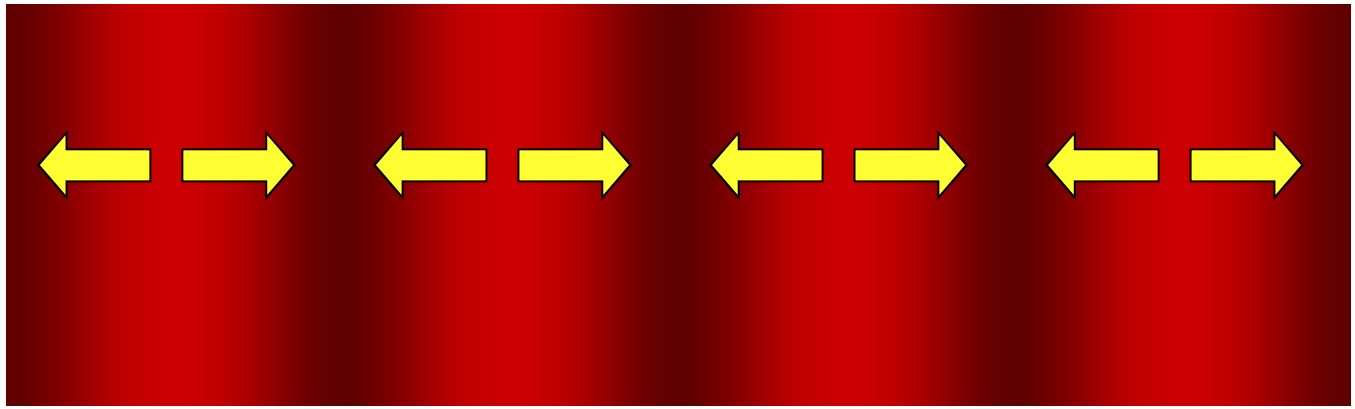
G Key properties of SPP

H SPP in lossy metals

I Fourier optics for surface plasmons

What is a (bulk) plasmon polariton?

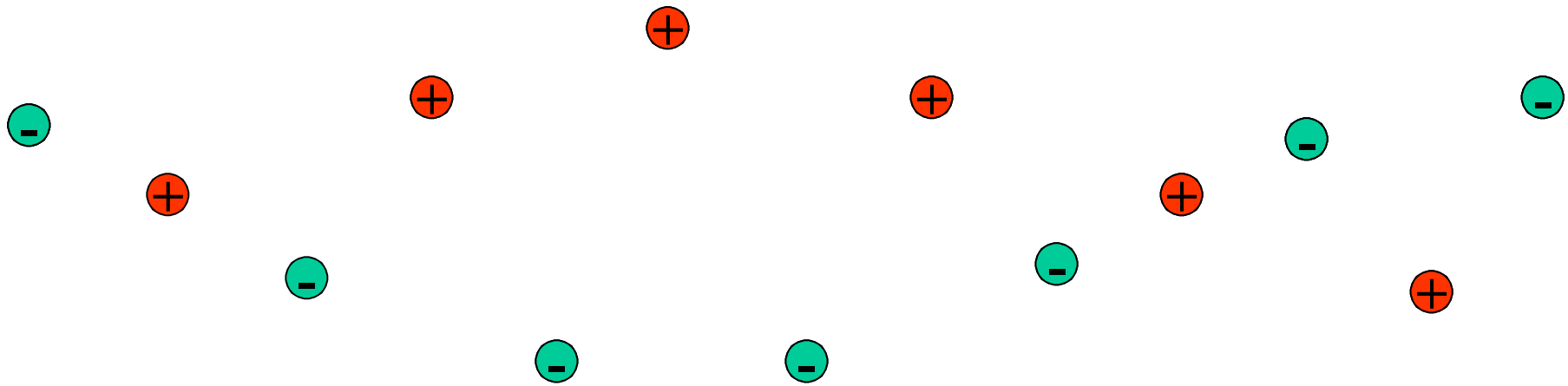
Acoustic wave in an electron gas :
photon+ phonon = polariton



An electron gas has a mechanical vibration eigenmode that generates a longitudinal EM mode.

Phonon polariton

Phonon polariton



(Bulk) Plasmon dispersion relation

Hydrodynamic model

$$div\ \mathbf{E} = -\frac{(n-n_0)e}{\varepsilon_0}$$

$$div\ n\mathbf{v} + \frac{\partial n}{\partial t} = 0$$

$$nm\left[\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v}\cdot\nabla\mathbf{v}\right] = -\nabla P - ne\mathbf{E}$$

$$\frac{\partial P}{\partial n} = m\frac{v_F^2}{3}$$

$$\omega^2 = \omega_p^2 + v^2k^2 \approx \omega_p^2$$

Electrodynamic point of view

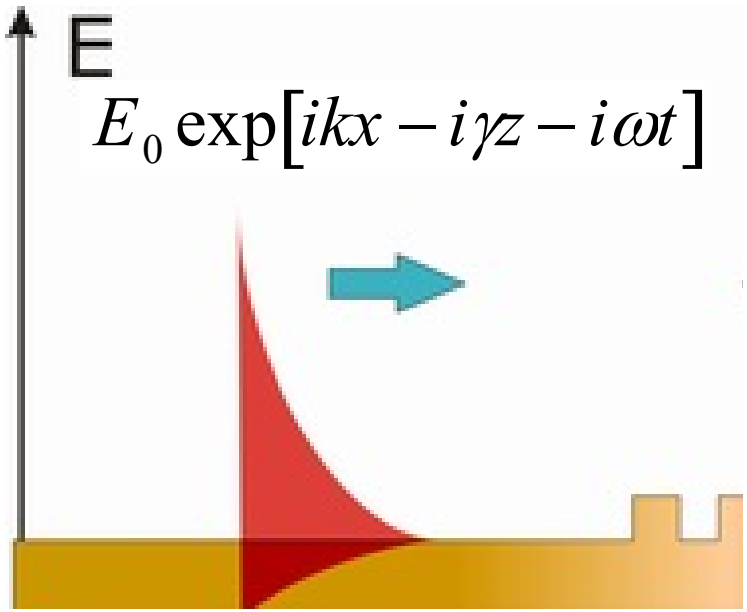
$$div\ D = div\ \varepsilon_0\varepsilon_r(\omega)\ E = 0$$

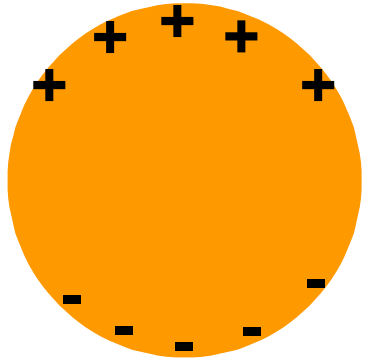
$$\varepsilon_r(\omega)\ k\cdot E(k,\omega) = 0$$

$$\varepsilon_r(\omega) = 1 - \frac{\omega_p^2}{\omega^2} = 0 \Rightarrow \omega = \omega_p$$

Plasmon is a coupled material/field mode: POLARITON.

What is a *surface* plasmon polariton?

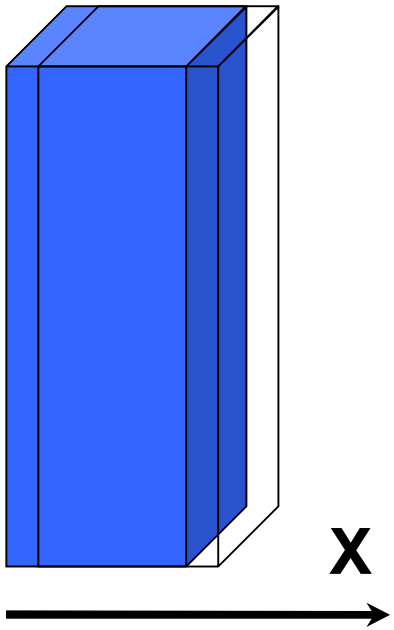




What is a surface plasmon?

Example : a thin film

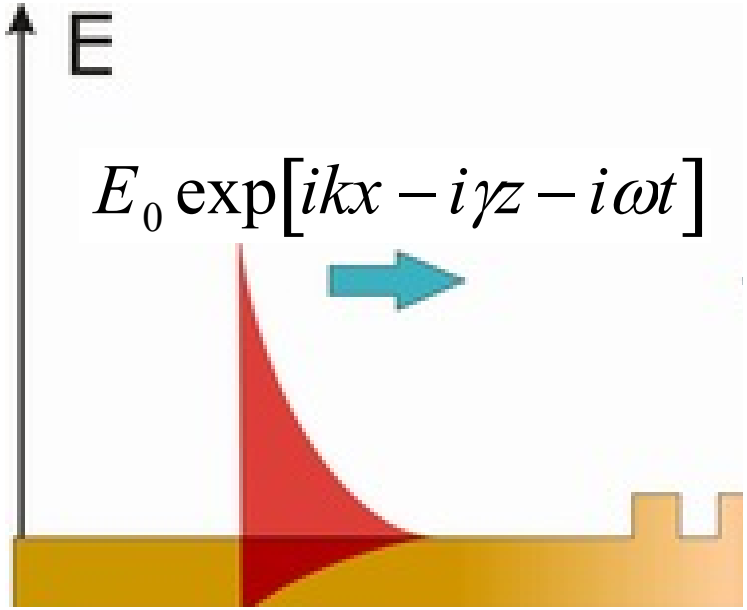
vibrational collective mode of oscillation of electrons



$$\omega_p^2 = \frac{ne^2}{m\varepsilon_0}$$

$$m\ddot{x} = -eE - \gamma m\dot{x} = -n\frac{e^2}{\varepsilon_0}x - \gamma m\dot{x}$$

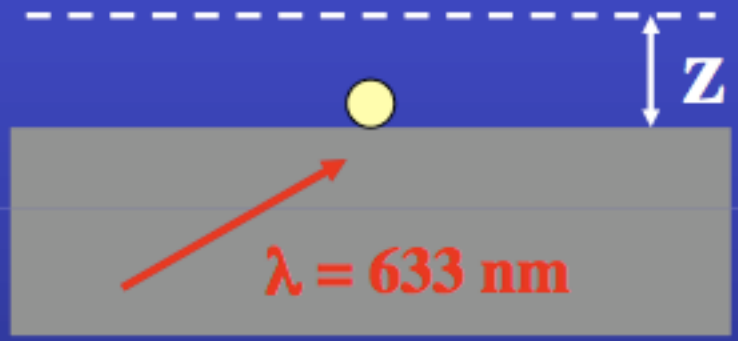
Typical length scales

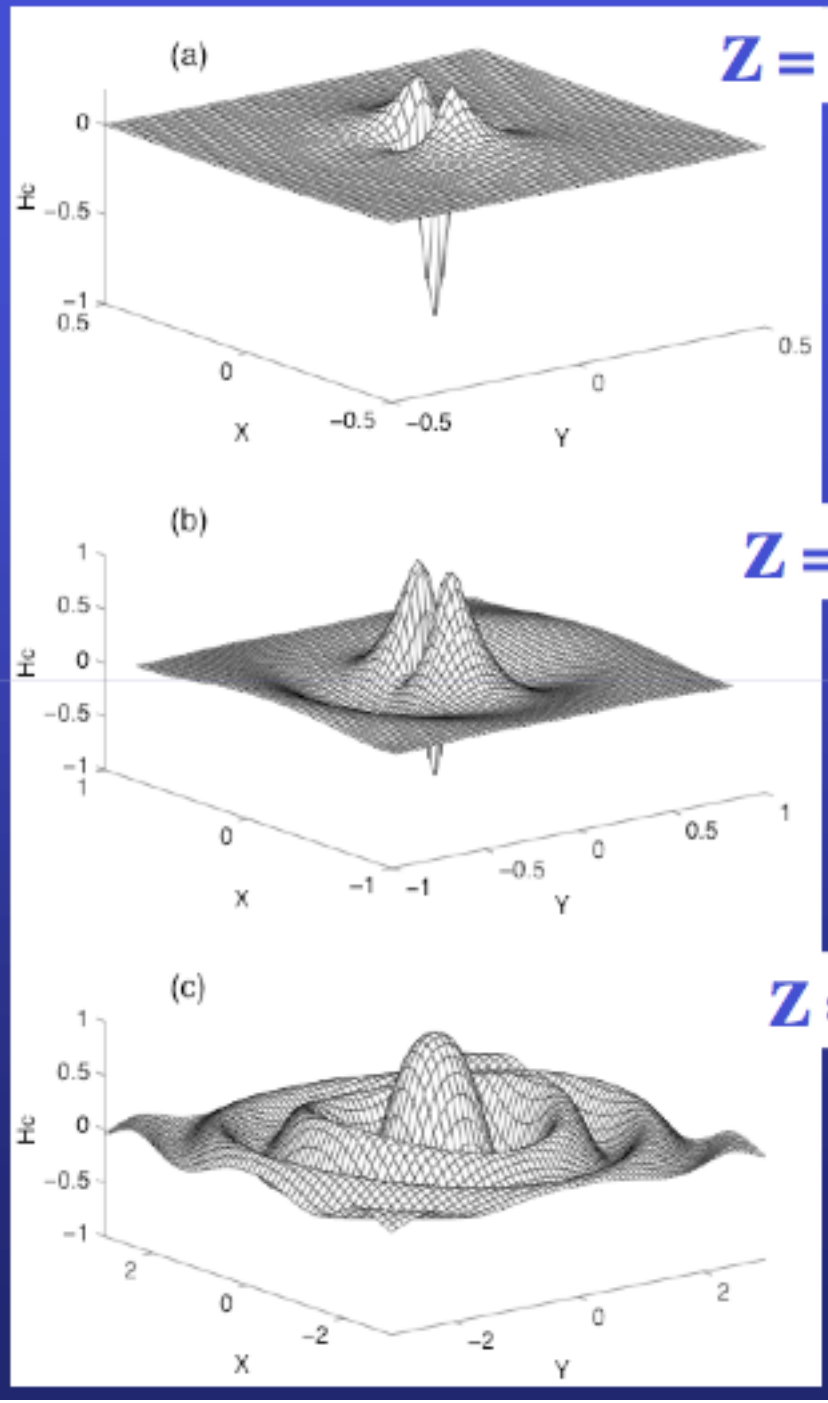


λ_i (μm)	0.633	1	10	36
δ_x (μm)	9.8	91.6	38 880	504 243
δ_{z1} (μm)	0.165	0.51	57.3	702.67
δ_{z2} (μm)	0.014	0.012	0.011	0.013

Fundamentals of near-field optics

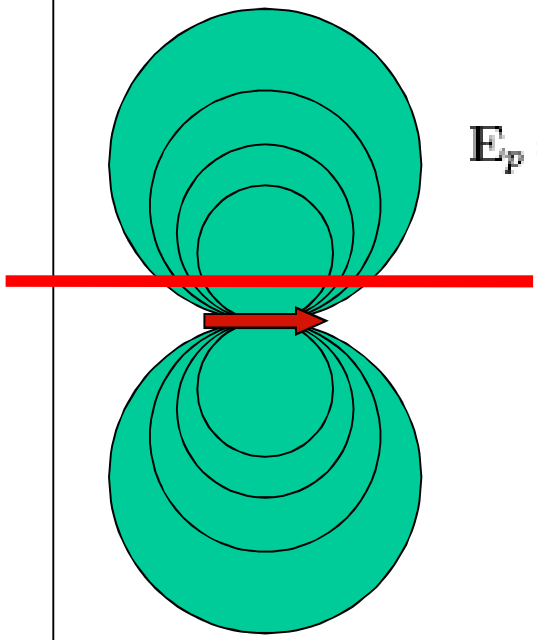
Spatial Filtering





Near field imaging

Back to basics : dipole radiation



$$\mathbf{E}_p = \frac{k_0^2}{4\pi\epsilon_0}\frac{e^{ik_0 r}}{r}\left[(\mathrm{I}-\mathrm{uu}) + \left(\frac{i}{k_0 r} - \frac{1}{(k_0 r)^2}\right)(\mathrm{I}-3\mathrm{uu})\right]\mathrm{p}$$

1/r terms
Radiation

$1/r^2$ and $1/r^3$ terms
Near field

Radiation dominates when $k_0 r>1$, $k_0=2\pi/\lambda$

Take home message :

In the region $r \ll \lambda$, retardation effects are negligible.

Although fields are time-dependent, their spatial structure can be derived using electrostatics with the complex dielectric constant $\varepsilon(\omega)$.

Near-field optics : fundamentals

The Fourier point of view : introducing evanescent waves

$$\frac{\exp\left[i\frac{\omega}{c}R\right]}{R} = \frac{i}{2\pi}\iint \frac{dk_x dk_y}{k_z}\exp\left[i(k_x x + k_y y + k_z z)\right]$$

$$k_\perp < \frac{\omega}{c} \quad ; \quad k_z = \sqrt{\frac{\omega^2}{c^2} - k_\perp^2} \quad ; \quad k_\perp = (k_x,\ k_y)$$

$$k_\perp > \frac{\omega}{c} \quad ; \quad k_z = i\sqrt{\left|\frac{\omega^2}{c^2} - k_\perp^2\right|}$$

$$k_\perp >> \frac{\omega}{c} \quad ; \quad k_z = ik_\perp \qquad resolution: \ \Delta x_{\min} \approx 2\pi / k_\perp \approx 2\pi z$$

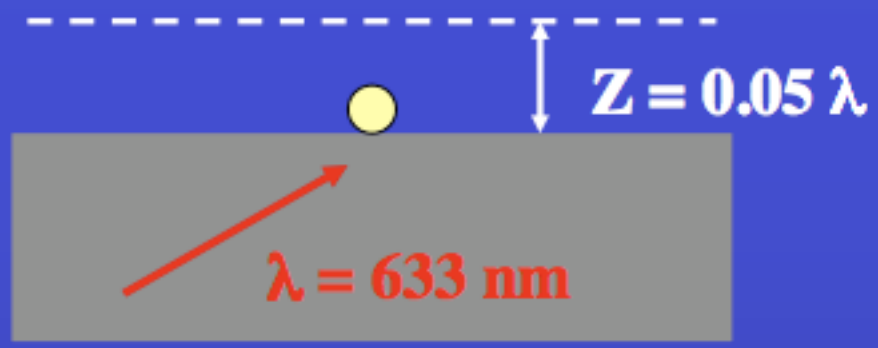
Near-field optics : fundamentals

The Fourier point of view

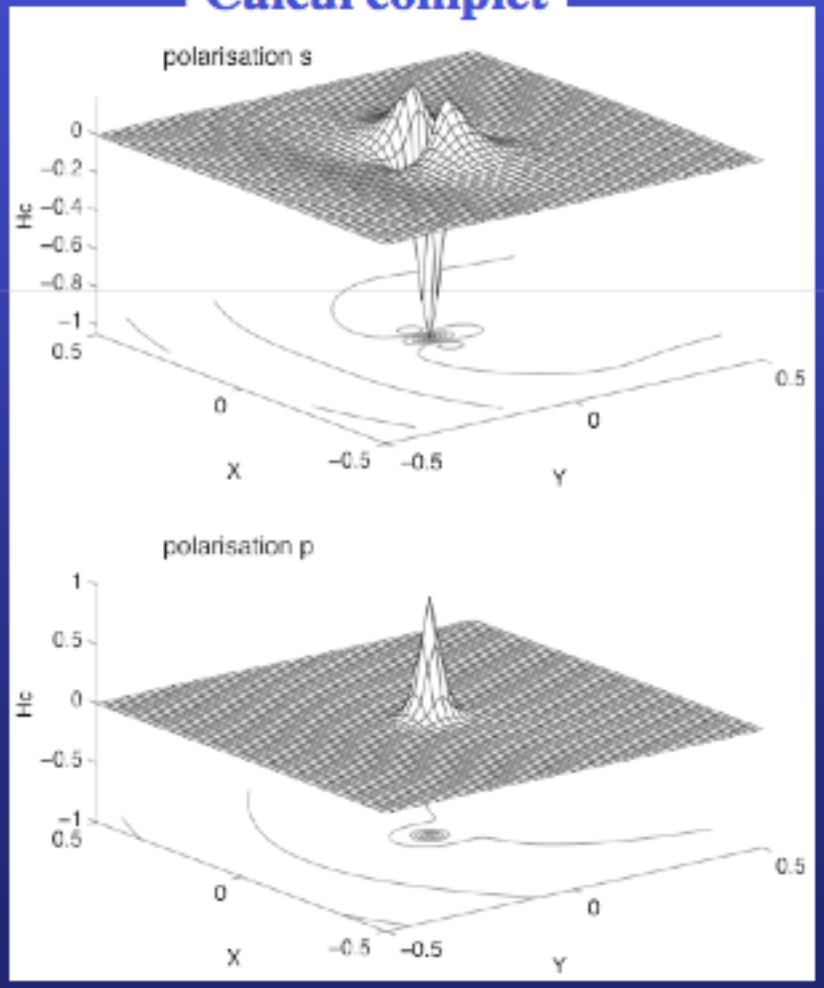
$$\frac{\exp\left[i\frac{\omega}{c}R\right]}{R} = \frac{i}{2\pi}\iint \frac{dk_x dk_y}{k_z}\exp\left[i(k_x x + k_y y)\right]\exp\left[-k_\perp z\right]$$

$$k_\perp >> \frac{\omega}{c} \quad ; \quad k_z = ik_\perp \qquad resolution: \boxed{\Delta x_{\min} \approx 2\pi / k_\perp \approx 2\pi z}$$

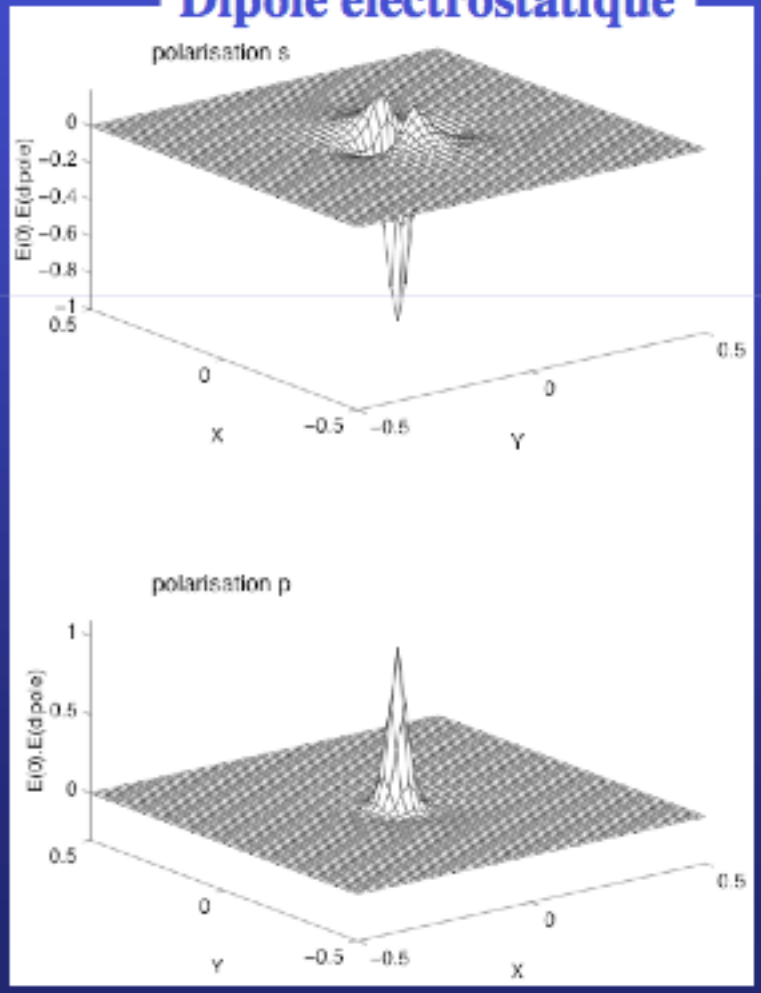
Limite quasi-statique



Calcul complet

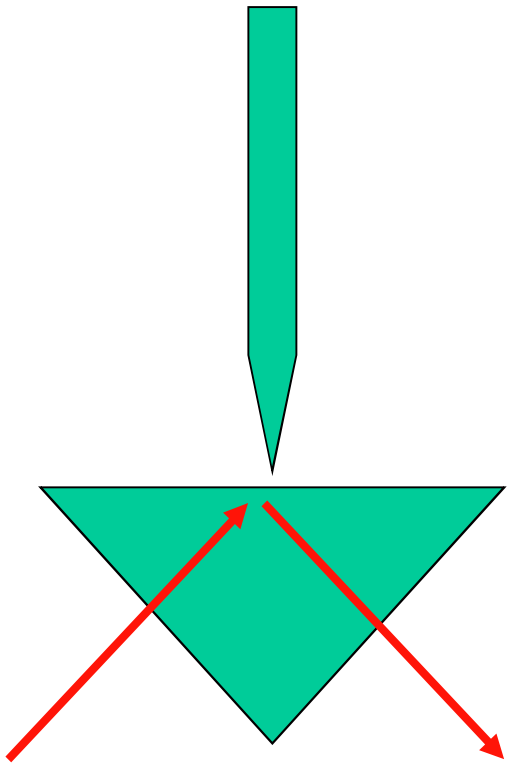


Dipôle électrostatique



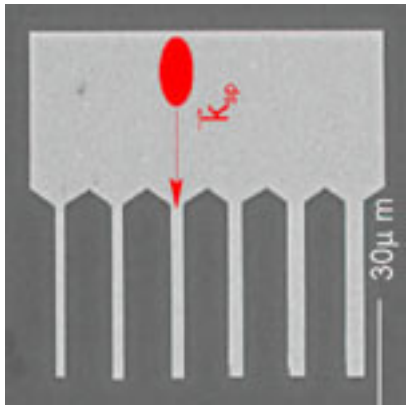
D. Some images of SPP

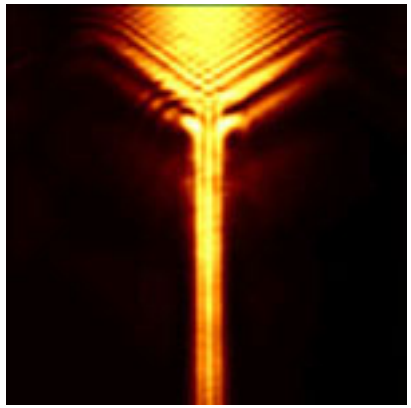
Image of a SPP



Dawson PRL 94

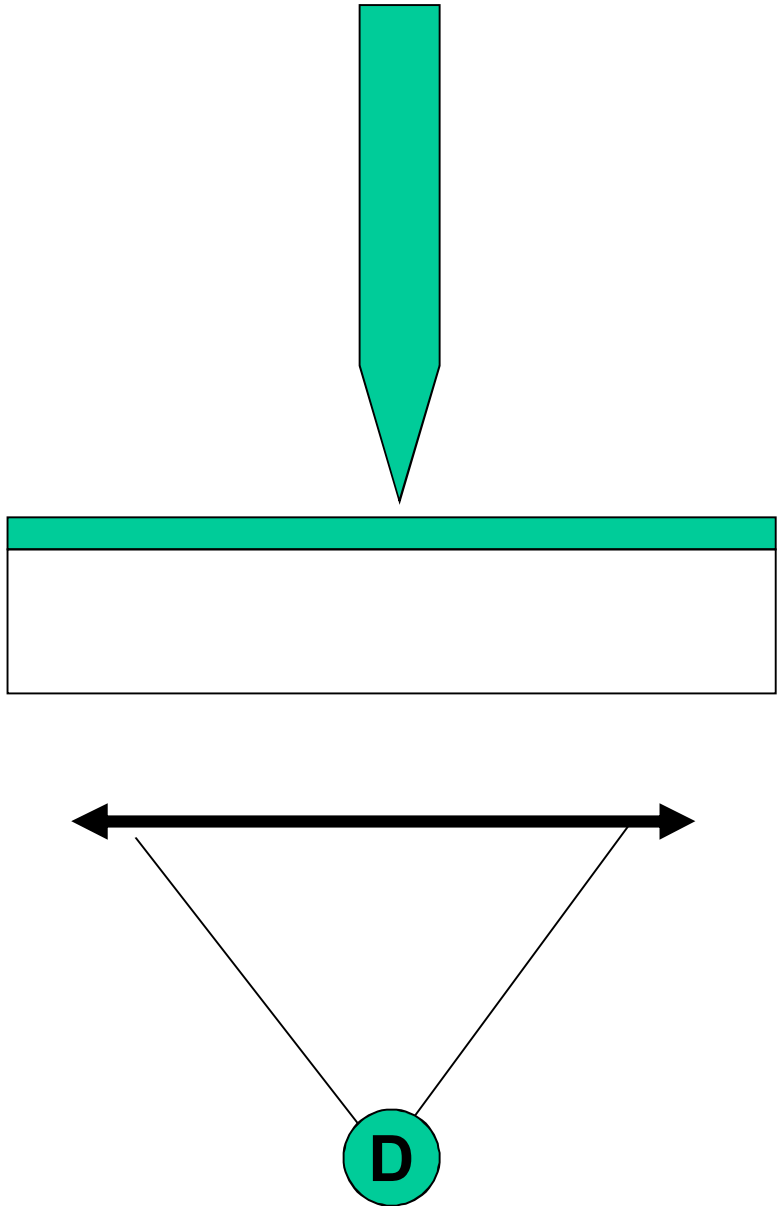
Metal stripes as SPP guides





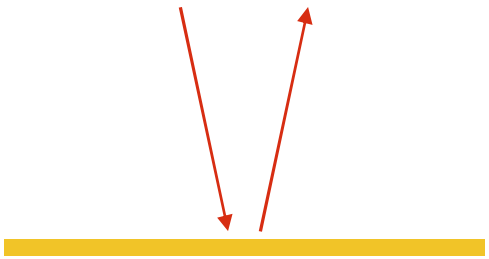
Université de Bourgogne

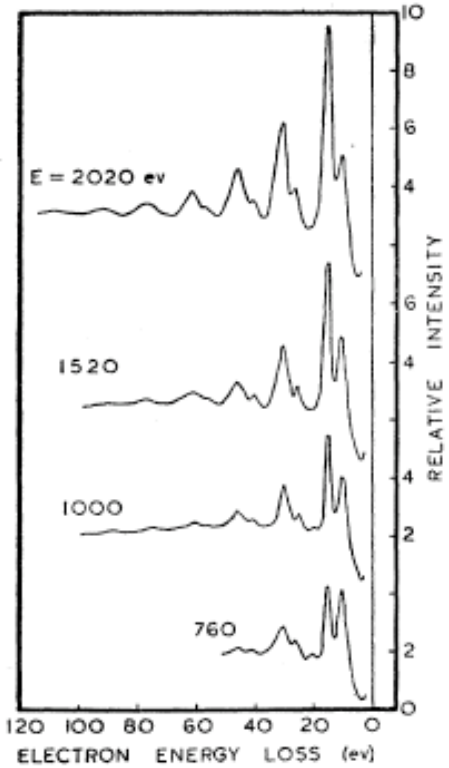
Excitation using a sub-λ source



Experimental proof of the existence of SPP

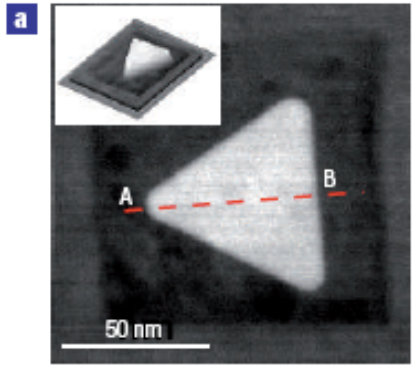
EELS (Electron Energy Loss Spectroscopy) of reflected electrons.

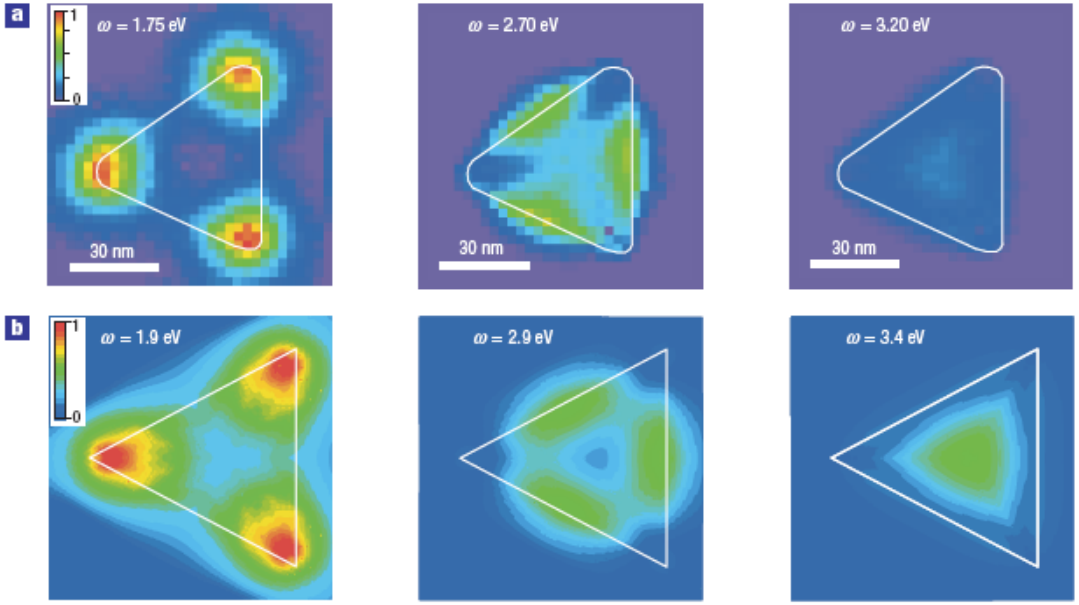




Powell, Phys.Rev. 1959

Observation of the LDOS using EELS





Nelayah et al. Nature Physics, (2007)

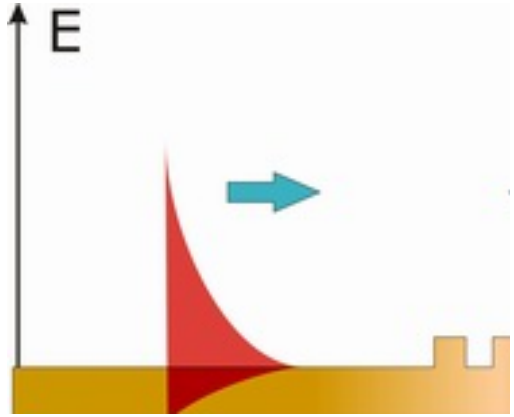
Surface waves

What is a Surface Wave (1)?

1. Solution of a homogeneous problem

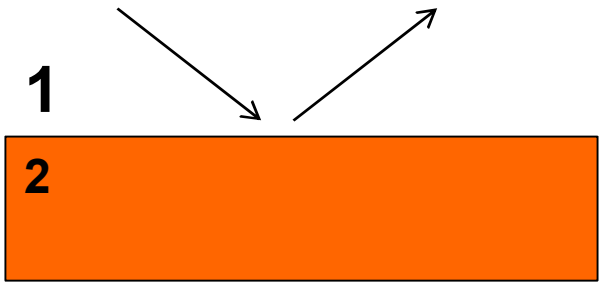
Derivation of the dispersion relation of the surface wave

- Helmholtz equation
- Boundary conditions
- exponential decay



$$K^2 = \frac{\omega^2}{c^2}\frac{\varepsilon}{\varepsilon+1}$$

2. Pole of a reflection factor



Poles and zeros

Dispersion relation

$$\varepsilon_2 k_{z1} + \varepsilon_1 k_{z2} = 0$$

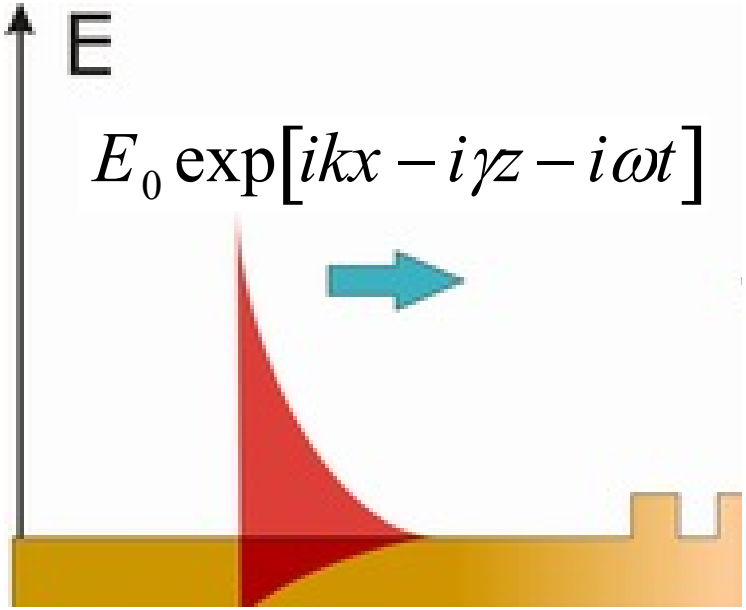
Reflection factor

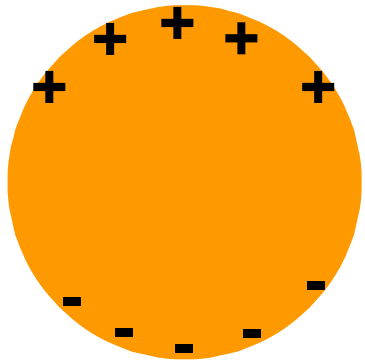
$$r_F = \frac{\varepsilon_2 k_{z1} - \varepsilon_1 k_{z2}}{\varepsilon_2 k_{z1} + \varepsilon_1 k_{z2}}$$

$K^2 = \frac{\omega^2}{c^2} \frac{\varepsilon}{\varepsilon + 1}$ **is a solution of** $\left(\varepsilon_2 k_{z1}\right)^2 = \left(\varepsilon_1 k_{z2}\right)^2$

Two cases : Brewster propagating wave and surface wave

What is a surface plasmon polariton?





$$r_F = \frac{\varepsilon_2 k_{z1} - \varepsilon_1 k_{z2}}{\varepsilon_2 k_{z1} + \varepsilon_1 k_{z2}}$$

$$\alpha = 4\pi a^3 \frac{\varepsilon_m(\omega) - 1}{\varepsilon_m(\omega) + 2}$$

$$\varepsilon_m(\omega) + 2 = 0$$

$$\varepsilon_2 k_{z1} + \varepsilon_1 k_{z2} = 0$$

$$\varepsilon_m(\omega) + \frac{l+1}{l} = 0$$

What is a Surface Wave (2)?

Long distance radio wave propagation :
The Zenneck's wave.

Dipole emission above an interface : the pole contribution and the Sommerfeld surface wave.

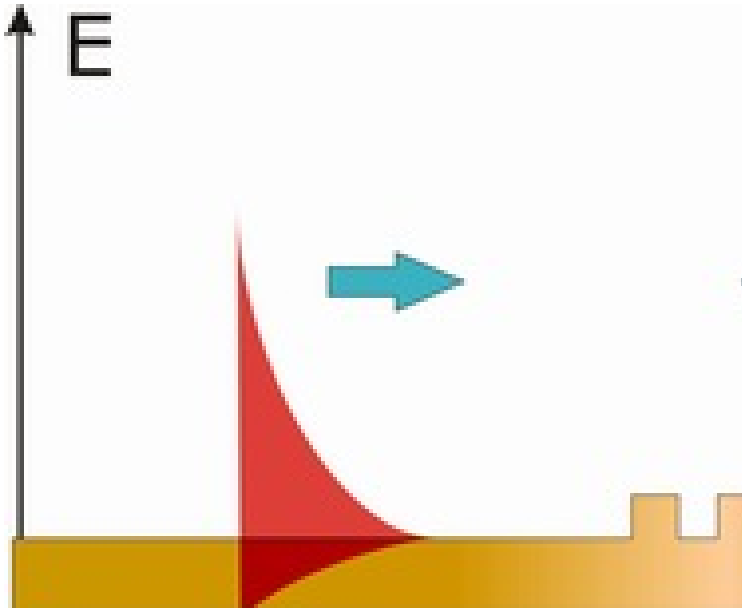
Norton approximate formula

Banos contribution

Lateral wave

What is a Surface Wave (3)?

1. Case of a good conductor (RF, microwaves, THz)



$$\varepsilon_r = \frac{i\sigma}{\omega\varepsilon_0}$$

$$k_{II} = \frac{\omega}{c}\left(1 + \frac{i\omega\varepsilon_0}{2\sigma}\right)$$

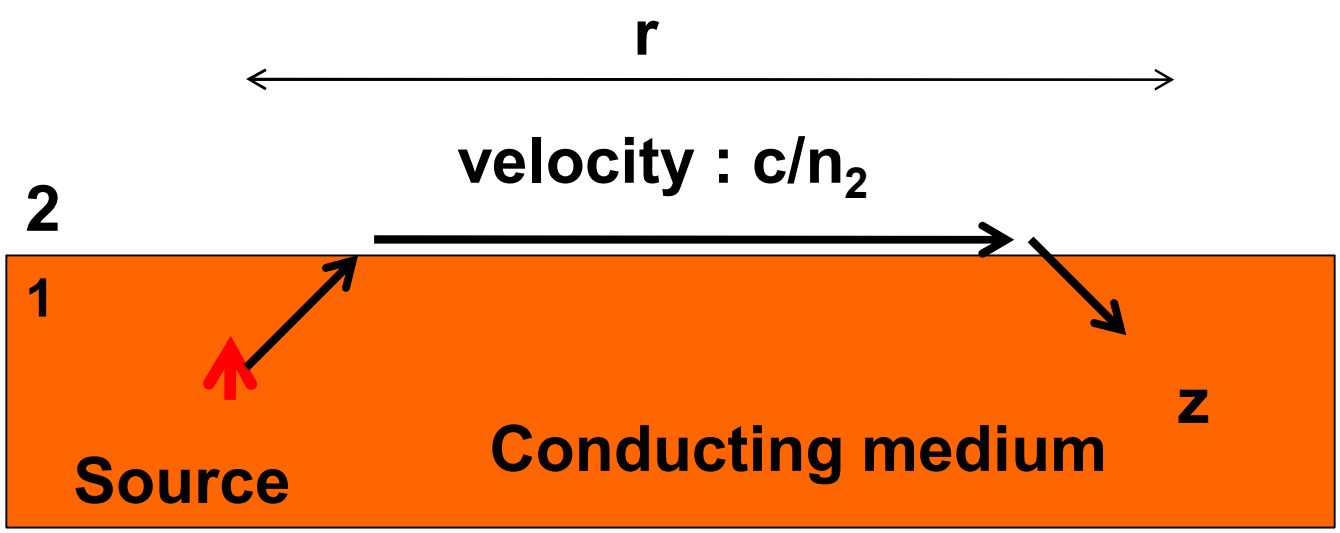
$$k_z = \frac{\omega}{c}\frac{i-1}{\sqrt{2}}\sqrt{\frac{\omega\varepsilon_0}{\sigma}}$$

This wave is due to the pole. Plasma frequency does not show up. Known as surface waves in radiowaves.

What is a lateral wave ?

In the ***far field***, the field decay as

$$\frac{\exp(ik_1 h + ik_2 r - ik_1 z)}{r^2}$$



This wave is NOT due to the pole

References

A. Banos, Dipole radiation in the presence of a conducting half space Pergamon Press, NY, 1966

L. Brekhovskikh Waves in layered media NY Academic Press 1980

A.Boardman Electromagnetic surface modes J. Wiley, NY 1982

R. King, Lateral electromagnetic waves, Springer Verlag, NY, 1992

Lalanne, Garcia Vidal, Collin. (quasi-cylindrical wave)

Optical properties of a metal

Optical properties of a metal

Drude model

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

Metal or dielectric ?

$\omega > \omega_p$ dielectric
$\omega < \omega_p$ metal

Plasmon or surface wave ?

$\omega > \gamma$ plasmon
$\omega < \gamma$ surface wave

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

oscillation

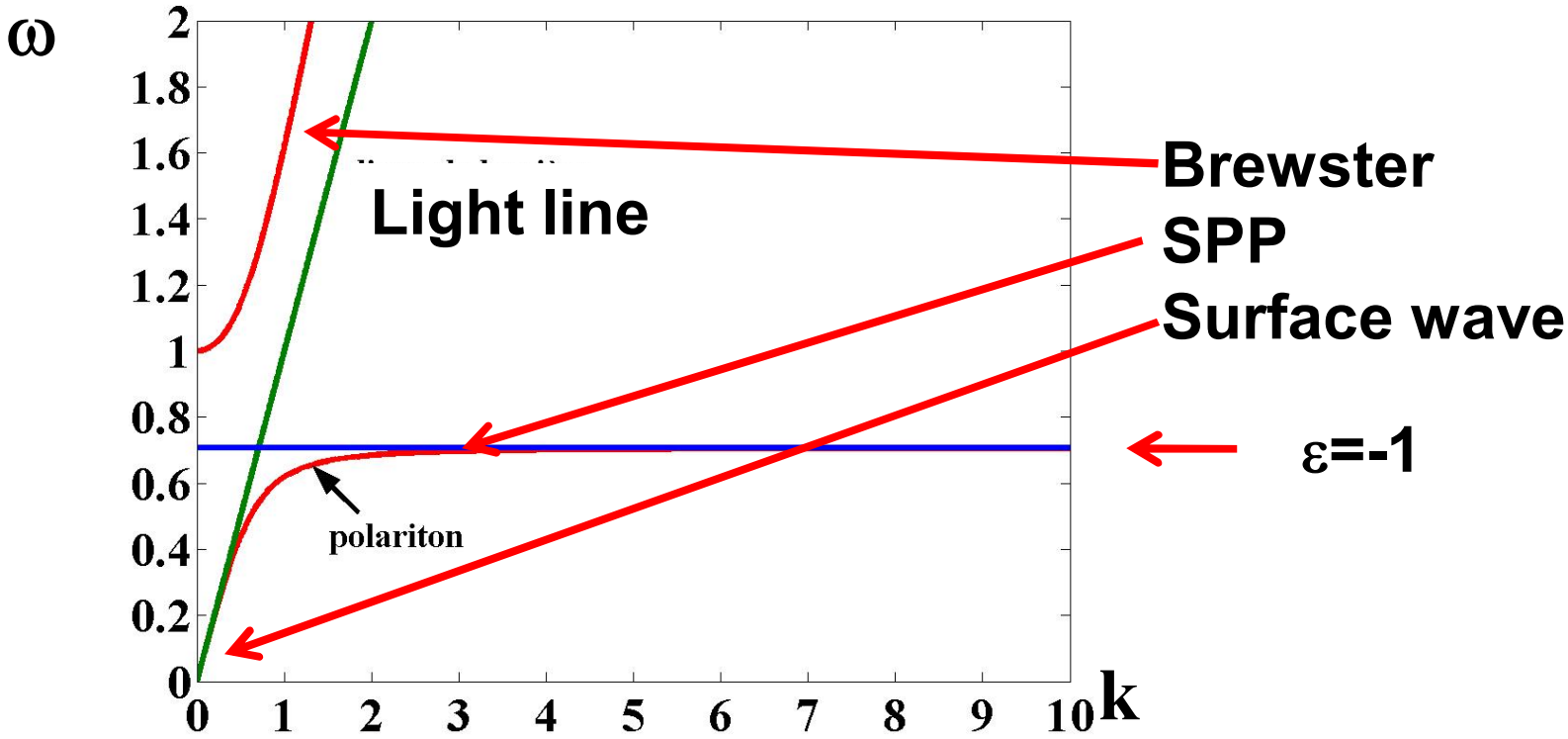
$$\varepsilon_r(\omega) = 1 - \frac{\omega_p^2}{i\omega\gamma} \approx i\frac{\omega_p^2}{\omega\gamma} = i\frac{\sigma}{\omega\varepsilon_0}$$

Overdamped oscillation

Surface plasmon polariton?

$$k^2 = \frac{\omega^2}{c^2}\sqrt{\frac{\varepsilon}{\varepsilon+1}} = \frac{\omega^2}{c^2}\sqrt{\frac{\omega^2-\omega_p^2}{2\omega^2-\omega_p^2}}$$

Drude model



Remark : no surface plasmon in metals at THz frequencies

Non local effects

How good is a macroscopic analysis of the problem?
What are the relevant length scales ?

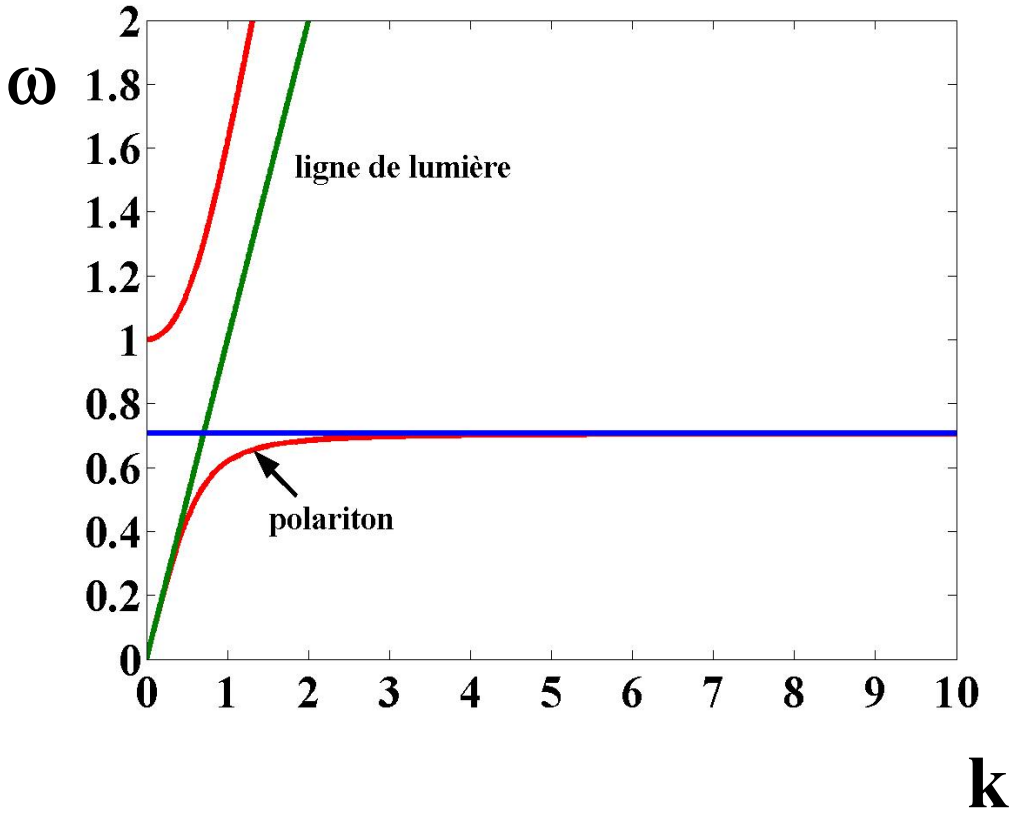
Definition of a non-local model
Origin of the non-locality :
- Thomas Fermi screening length
- Landau Damping

$$\hbar\omega = \frac{\hbar^2}{2m}[(\mathrm{q}+\mathrm{dq})^2 - \mathrm{q}^2] \approx \frac{\hbar^2 q \mathrm{d}q}{m} \approx \hbar v_F \mathrm{d}q \qquad \cdot\, k \approx \omega / v_F$$
$$\hbar\mathrm{k} = \hbar\,\mathrm{dq}$$

The Landau mechanism introduces a cutoff k.

Specific properties of SPP

1. Large density of states
2. Fast relaxation/broad spectrum
3. Confined fields



SPP Key properties 1

1. Large local density of states

Local Density of States

Energy point of view

$$U = \frac{\omega^2}{\pi^2 c^3} \frac{1}{\exp\left(\frac{\hbar\omega}{kT}\right) - 1} \hbar\omega$$

Lifetime point of view

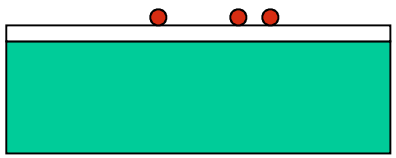
$$A_{21} = B_{21} \frac{\hbar\omega^3}{\pi^2 c^3} = \left[B_{21}\hbar\omega\right] \frac{\omega^2}{\pi^2 c^3}$$

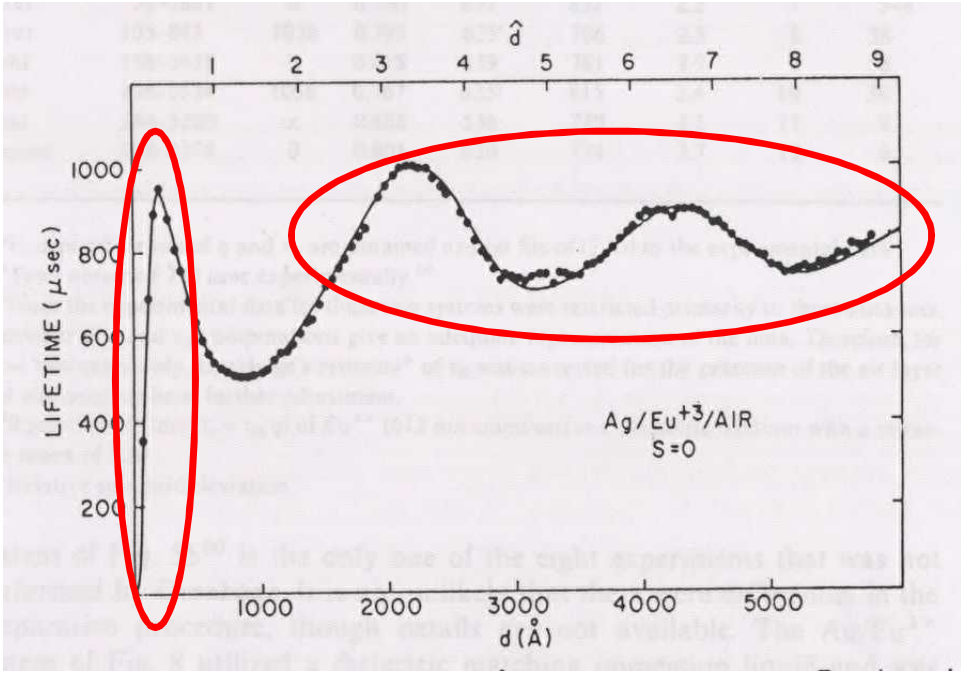
Larger LDOS means : i) shorter lifetime, ii) larger energy at thermodynamic equilibrium

LDOS and Spontaneous emission

Lifetime

SPP





Interferences

Drexhage (1970)
Chance, Prock, Silbey (1978)

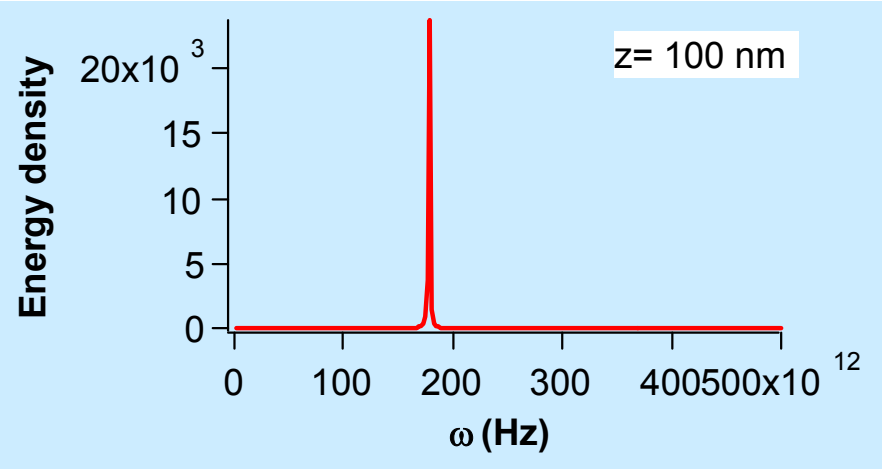
Lifetime is not intrinsic but depends on the environment

Asymptotic form of the EM- LDOS

Near-field form

$$N(\omega) = \frac{1}{16\pi^2 \omega z^3} \frac{\mathrm{Im}[\varepsilon(\omega)]}{|1+\varepsilon(\omega)|^2}$$

- **Resonance for ε(ω)→ -1**
- **Lorentzian shape**
- **The near-field effect exists without SPP !!**



Signature of the SPP ?

PRL 85, 1548 (2000)

PRB 68, 245405 (2003)

Where are the *new* modes coming from?

Estimate of the number of EM states with frequency below ω:

$$\frac{N}{V} = \int_0^{\omega} g(\omega')d\omega' = \frac{\omega^3}{3\pi^2 c^3} \qquad N \approx \frac{V}{\lambda^3}$$

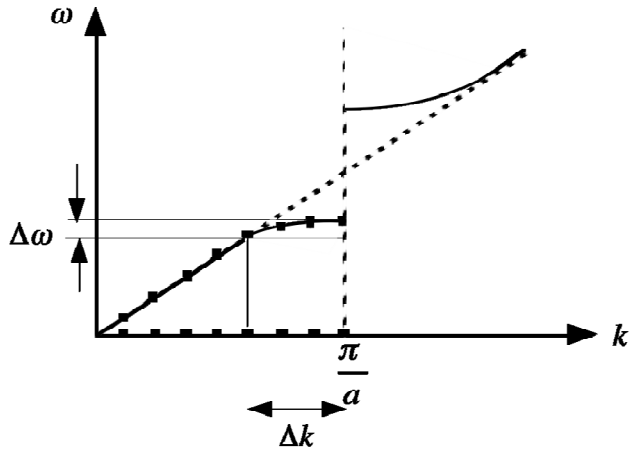
Estimate of the number of electrons/phonons:

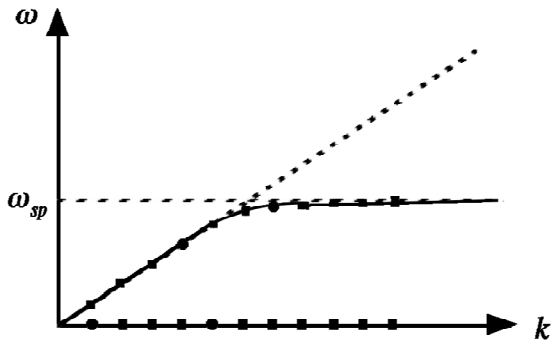
$$N \approx \frac{V}{a^3}$$

The EM field inherits the large DOS of matter.

Large LDOS : Plasmon or photonic crystal ?

Photonic crystals ar often used for increasing the LDOS taking advantage of the low group velocity

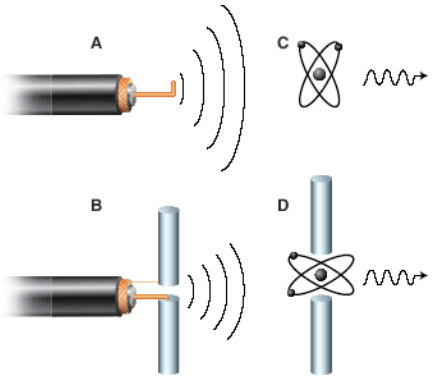




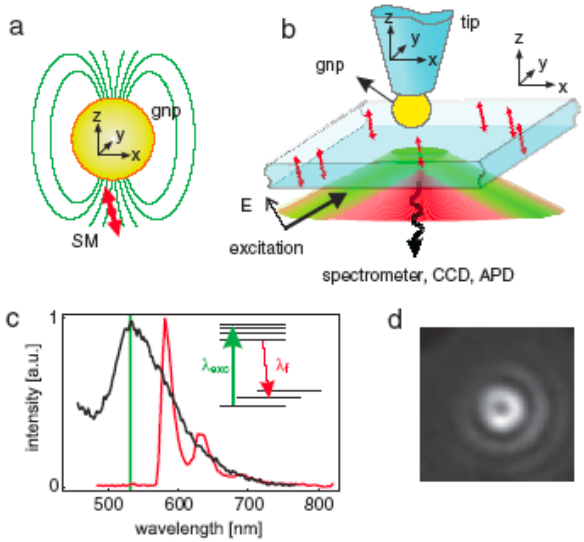
photonic crystal **Surface plasmon**

Conclusion : Plasmons are orders of magnitude more Efficient in increasing the LDOS.

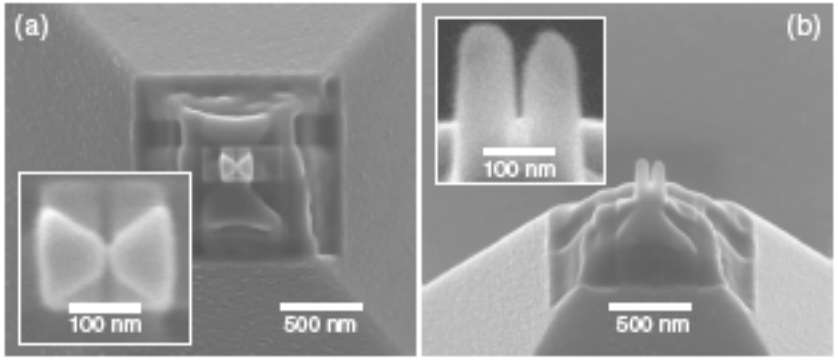
Application : nanoantenna



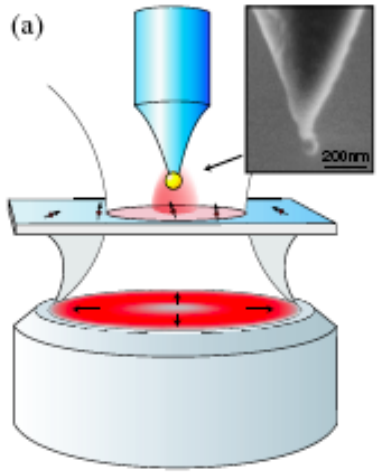
Mühlschlegel et al. Science 308 p 1607 (2005)
Greffet, Science 308 p (2005) p 1561



Kühn et al. PRL 97, 017402 (2006)



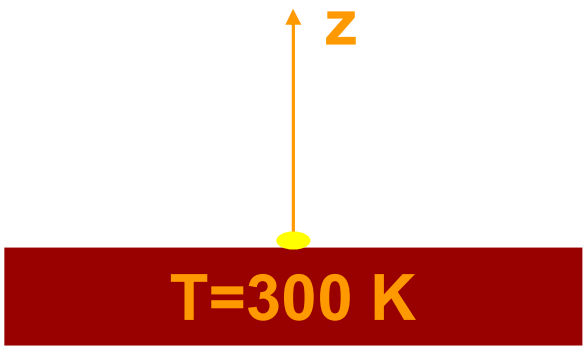
Farahani et al., PRL 95, 017402 (2005)

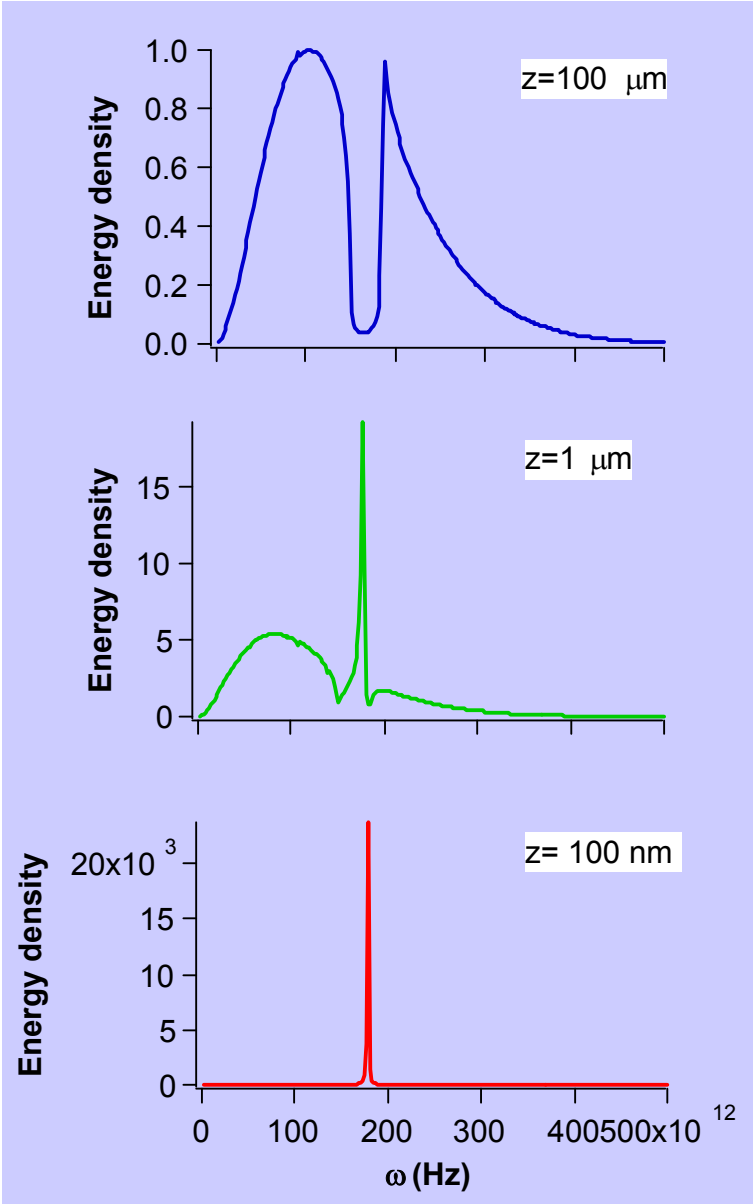


Anger et al., PRL 96, 113002 (2006)

LDOS and energy

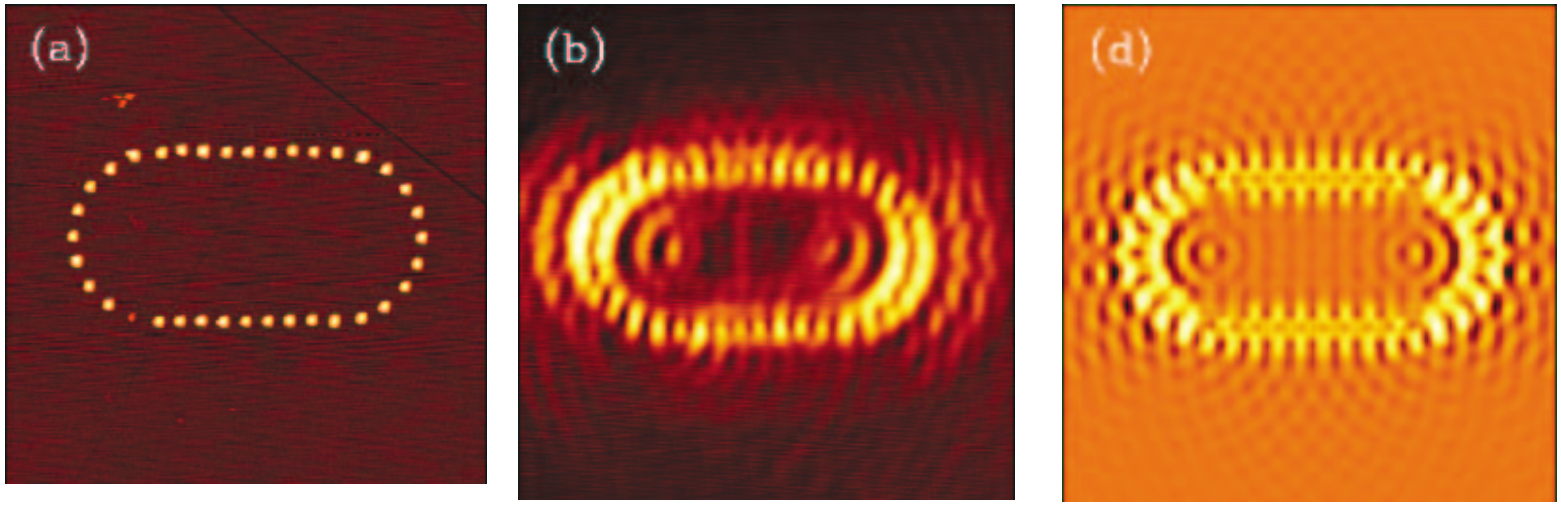
Energy density close to the surface

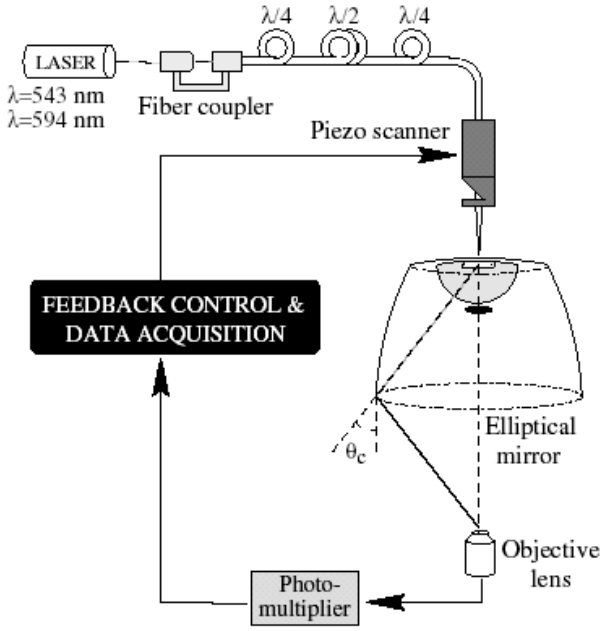




Shchegrov PRL, 85 p 1548 (2000)

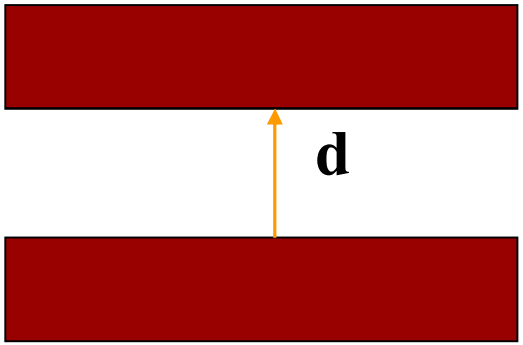
Observation of the SPP LDOS

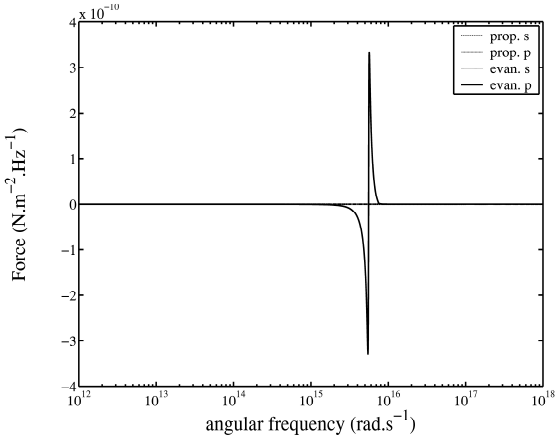




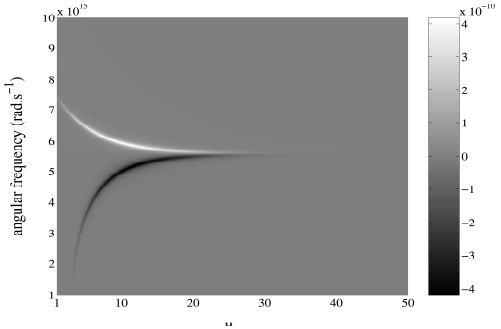
C. Chicanne et al., Phys. Rev. Lett. 88, 97402 (2002)

SPP LDOS and Casimir force

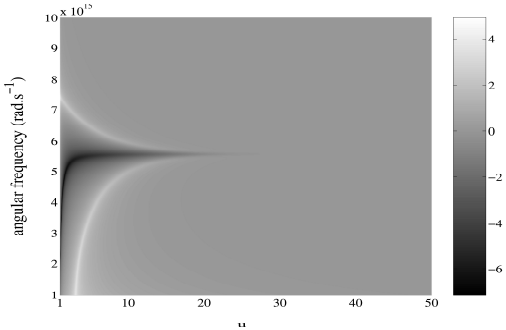




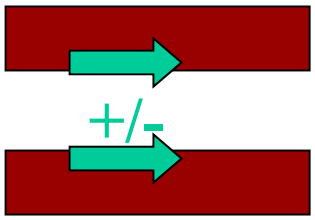
$$F = \int F(k,\omega)\, d^3k\, d\omega$$



Force



Dispersion relation



Joulain, Phys. Rev.A 69, 023808 (2004)

SPP key properties 2

Fast relaxation/Broad spectrum

Losses in noble metals (1)

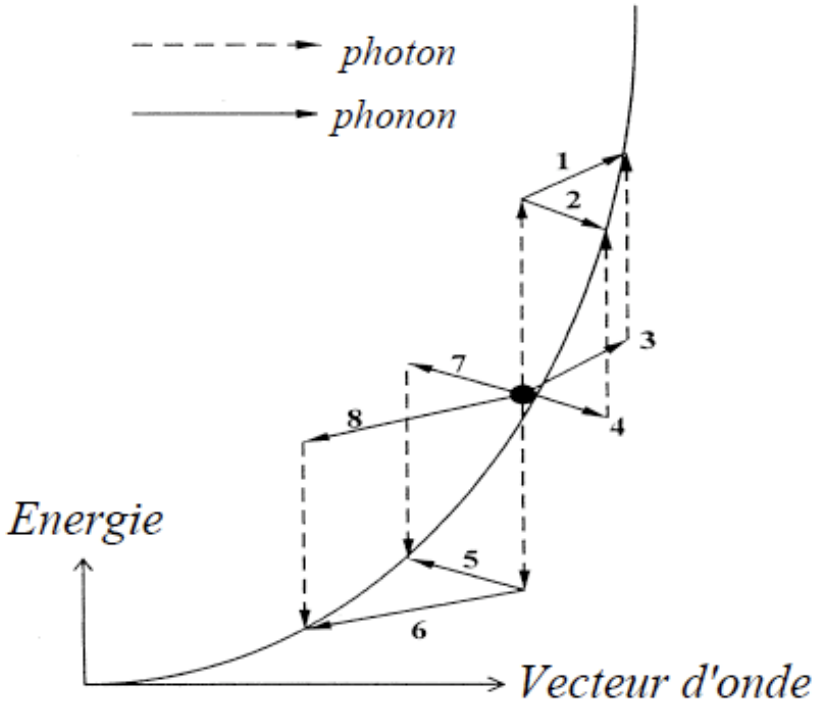
$$\varepsilon_r(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma}$$

Collisions	Relaxation time
Electron-phonon DC	α Te
Electron-phonon at optical frequency	17 fs weak dependence on Te
Electron-electron	170 fs

Adv. in Phys. 33 p 257 (1984)
Phys.Rev.B 25 p 923 (1982)
Phys.Rev.B 3 p 305 (1971)

Losses in noble metals (2)

Intraband loss Mechanism



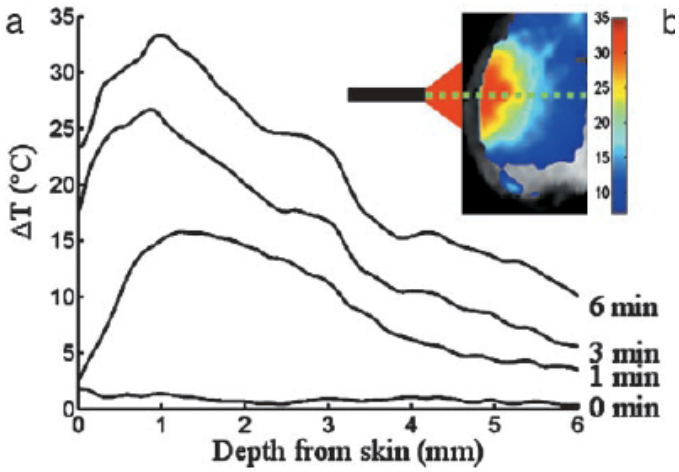
Different mechanisms at high frequency and low frequency

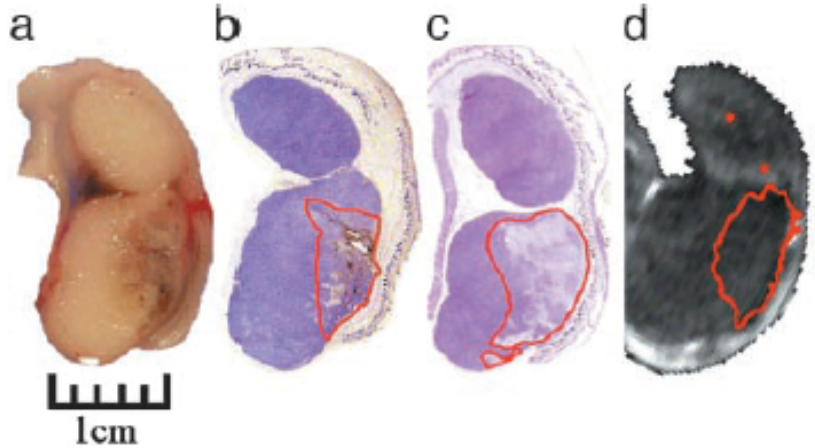
DC-GHZ : 2 bodies interaction
optics : 3 bodies interaction

Applications :

- Broad spectrum antenna
- Fast hot spot
- Absorber
- Local Heater

Tumor therapy





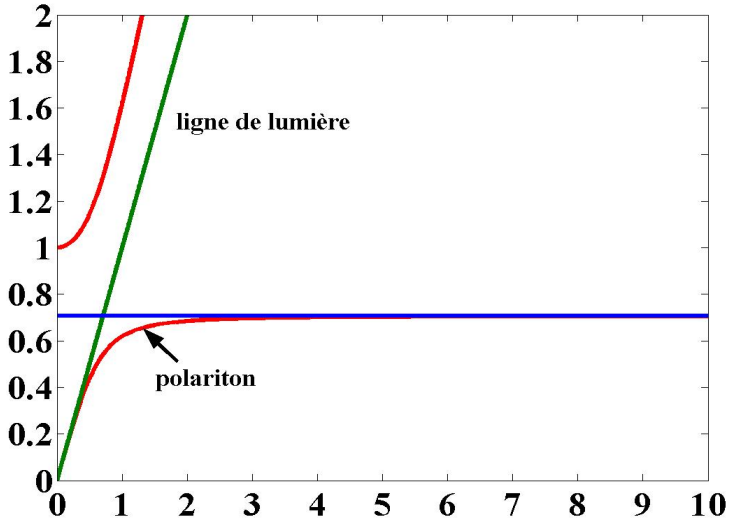
Nanoshell-mediated near-infrared thermal therapy of tumors under magnetic resonance guidance

L. R. Hirsch*, R. J. Stafford†, J. A. Bankson†, S. R. Sershen*, B. Rivera‡, R. E. Price‡, J. D. Hazle†, N. J. Halas§, and J. L. West*¶

PNAS | November 11, 2003 | vol. 100 | no. 23 | 13549–13554

SPP key properties 3

Field confinment



Field confinment by particles, tips

Electrostatic or SPP confinment ?

Examples : bow-tie, antennas, lightning rod, particles, superlens

Field confinment by particles, tips

Electrostatic or SPP confinment ?

Examples : bow-tie, antennas, lightning rod, particles

$$N(\omega) = \frac{1}{16\pi^2 \omega z^3} \frac{\mathrm{Im}\left[\varepsilon(\omega)\right]}{\left|1+\varepsilon(\omega)\right|^2}$$

Look for a resonance close to ω_p.

H. Dispersion relation in lossy metals

Archambault Phys. Rev. B 79 195414 (2009)

Surface plasmon

Solution for a non-lossy medium

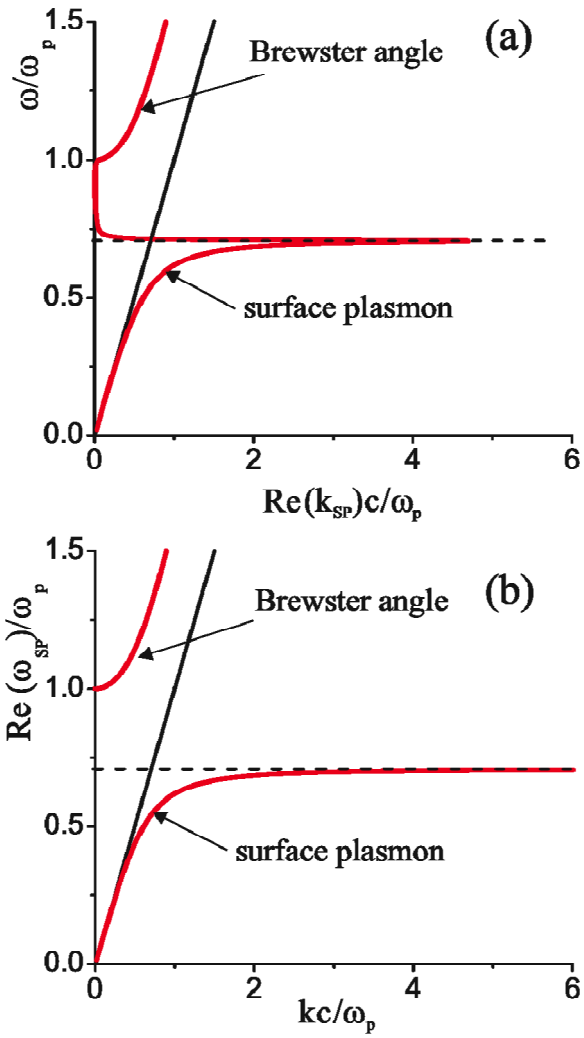
$$\mathbf{E}(z)\exp[i(K_x x + K_y y - \omega t)]$$

Dispersion relation

$$K^2 = \frac{\omega^2}{c^2}\frac{\varepsilon}{\varepsilon+1}$$

If ε is complex, there is no solution with real K and ω.

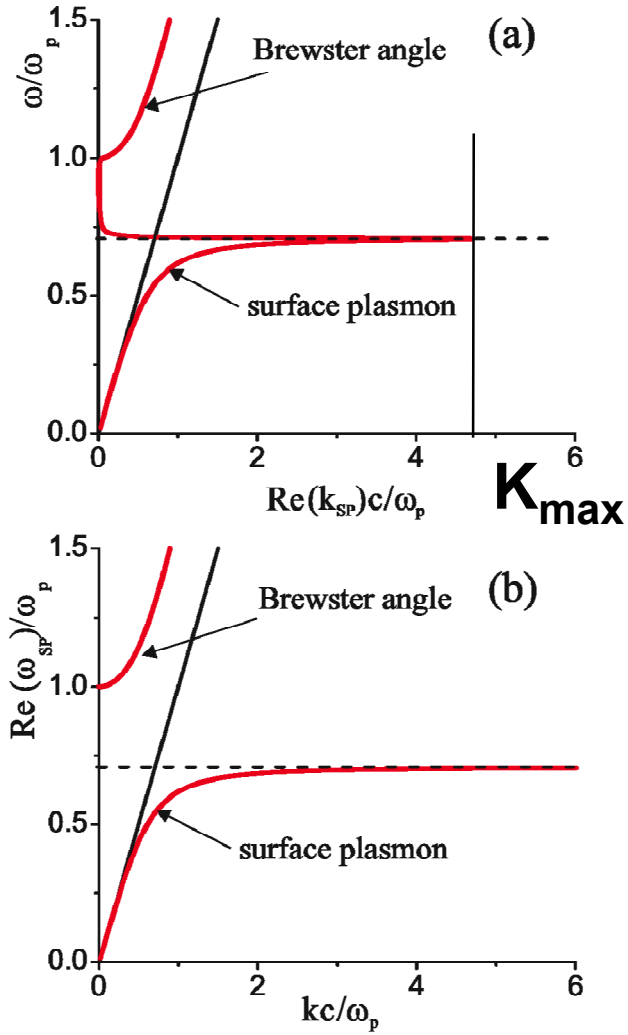
Surface plasmon dispersion relation



Real ω and complex K

Real K and complex ω.

Maximum confinment of the field ?

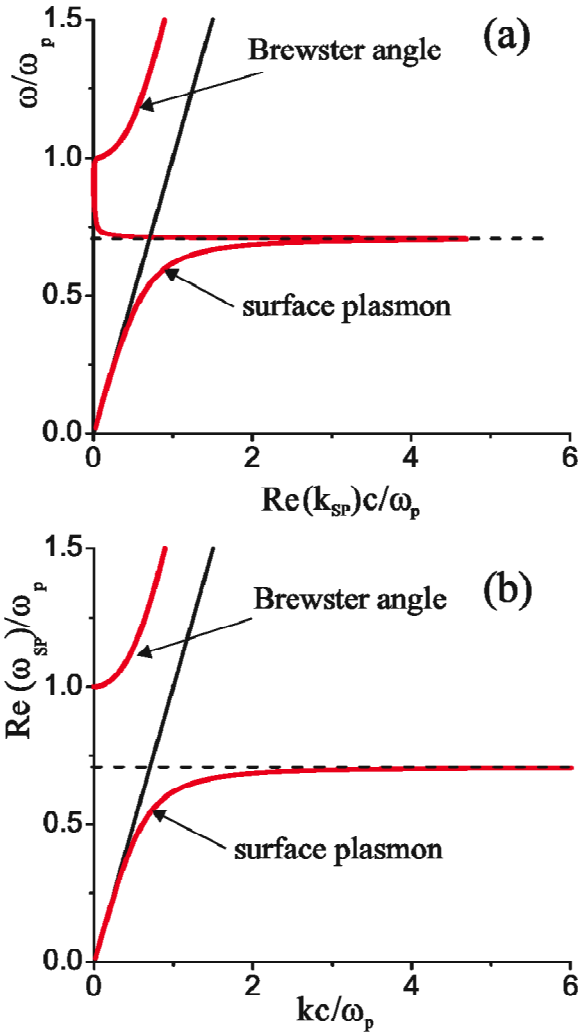


K_{max}

$1/K_{max}$

No limit !

Local Density of states?



Finite value of the LDOS

Divergence of the LDOS

The two dispersion relations predict different results regarding resolution and LDOS

Yet, these are well defined physical quantities !

What is the relevant dispersion relation ?

General representation of the field

We start from the general representation of the field generated by an arbitrary source distribution.The field is given explicitly by the Green tensor.

$$\mathbf{E}(\mathbf{r},t) = -\mu_0 \int \mathrm{d}t' \int \mathrm{d}^3\mathbf{r}' \, \overleftrightarrow{G}(\mathbf{r},\mathbf{r}',t-t') \frac{\partial \mathbf{j}(\mathbf{r}',t')}{\partial t'},$$

The Green tensor has a Fourier representation :

$$\overleftrightarrow{G}(\mathbf{r},\mathbf{r}',t-t') = \int \frac{\mathrm{d}^2\mathbf{K}}{4\pi^2} \int \frac{d\omega}{2\pi} \overleftrightarrow{g}(\mathbf{K},z,z',\omega) \, e^{i[\mathbf{K}(\mathbf{r}-\mathbf{r}')-\omega(t-t')]},$$

It includes Fresnel reflection factor and therefore poles representing surface plasmons.

General representation of the field

Following Sommerfeld, we define the **surface wave** as the **pole contribution to the field**

$$\overleftrightarrow{G} = \overleftrightarrow{G}_{reg} + \overleftrightarrow{G}_{sp},$$

$$\mathbf{E}_{sp}(\mathbf{r},t) = -\mu_0 \int \mathrm{d}t' \int \mathrm{d}^3\mathbf{r}' \; \overleftrightarrow{G}_{sp}(\mathbf{r},\mathbf{r}',t-t') \frac{\partial \mathbf{j}(\mathbf{r}',t')}{\partial t'}.$$

General representation of the field

Evaluating the pole contribution :

We can **choose** to integrate either over ω or over K_x

$$\overleftrightarrow{g}_{sp}(\mathbf{K}, z, z', \omega) = \frac{\overleftrightarrow{f}_{\omega_{sp}}(\mathbf{K}, z, z')}{\omega - \omega_{sp}} + \frac{\overleftrightarrow{f}_{-\omega^*_{sp}}(\mathbf{K}, z, z')}{\omega + \omega^*_{sp}},$$

$$\overleftrightarrow{g}_{sp}(\mathbf{K}, z, z', \omega) = \frac{\overleftrightarrow{f}_{K_{\mathrm{x,\,sp}}}(K_{\mathrm{y}}, z, z', \omega)}{K_{\mathrm{x}} - K_{\mathrm{x,\,sp}}} + \frac{\overleftrightarrow{f}_{-K_{\mathrm{x,\,sp}}}(K_{\mathrm{y}}, z, z', \omega)}{K_{\mathrm{x}} + K_{\mathrm{x,\,sp}}}$$

Due to losses, poles are complex !

General representation of the field

We obtain two different representations of the SP field :

Complex K

$$\mathbf{E} = \int \frac{\mathrm{d}\omega}{2\pi} \int \frac{\mathrm{d}K_\mathrm{y}}{2\pi} (\hat{\mathbf{K}} - \tfrac{K_{sp}}{\gamma_\mathrm{m}} \mathbf{n}_\mathrm{m}) E_>(K_\mathrm{y}, \omega) \, e^{i(\mathbf{K}\cdot\mathbf{r} + \gamma_\mathrm{m}|z| - \omega t)}$$

Complex ω

$$\mathbf{E}_\mathrm{sp} = 2\Re\mathfrak{e} \int \frac{\mathrm{d}^2\mathbf{K}}{(2\pi)^2} E(\mathbf{K}, t)(\hat{\mathbf{K}} - \tfrac{K}{\gamma_\mathrm{m}} \mathbf{n}_\mathrm{m}) \, e^{i(\mathbf{K}\cdot\mathbf{r} + \gamma_\mathrm{m}|z| - \omega_{sp} t)},$$

Each representation has its own dispersion relation

General representation of the field

Which representation should be used ?

Complex K

$$\mathbf{E} = \int \frac{\mathrm{d}\omega}{2\pi} \int \frac{\mathrm{d}K_\mathrm{y}}{2\pi} (\hat{\mathbf{K}} - \frac{K_{sp}}{\gamma_\mathrm{m}} \mathbf{n}_\mathrm{m}) E_>(K_\mathrm{y}, \omega)$$

$$e^{i(\mathbf{K}\cdot\mathbf{r} + \gamma_\mathrm{m}|z| - \omega t)}$$

The amplitude $E_>$ depends on x *in the sources*. It does not depend on x outside the sources.

The complex k representation is well suited for localized stationary sources. The dispersion relation has a backbending.

Archambault Phys. Rev. B 79 195414 (2009)

General representation of the field

Which representation should be used ?

Complex ω

$$\mathbf{E}_{\mathrm{sp}} = 2\Re\mathfrak{e} \int \frac{\mathrm{d}^2\mathbf{K}}{(2\pi)^2} E(\mathbf{K},t)(\hat{\mathbf{K}} - \frac{K}{\gamma_{\mathrm{m}}}\mathbf{n}_{\mathrm{m}}) e^{i(\mathbf{K}\cdot\mathbf{r}+\gamma_{\mathrm{m}}|z|-\omega_{sp}t)},$$

The amplitude $E_>$ depends on t when the source is active. It does not depend on time after the sources have been turned off. The imaginary part of the frequency describes the decay of the wave.

The complex frequency formulation is well suited for pulse excitations. The dispersion relation has no backbending.

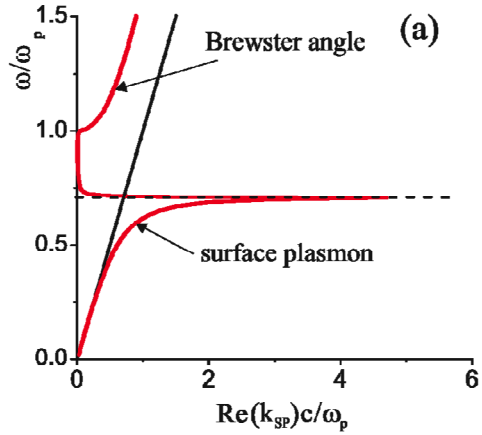
Archambault Phys. Rev. B 79 195414 (2009)

Discussion

What is the best confinment ?

Localized sources and stationary regime :
complex K and real ω

$$\mathbf{E} = \int \frac{\mathrm{d}\omega}{2\pi} \int \frac{\mathrm{d}K_\mathrm{y}}{2\pi} \left(\hat{\mathbf{K}} - \frac{K_{sp}}{\gamma_\mathrm{m}} \mathbf{n}_\mathrm{m}\right) E_>(K_\mathrm{y}, \omega) \, e^{i(\mathbf{K}\cdot\mathbf{r} + \gamma_\mathrm{m}|z| - \omega t)}$$



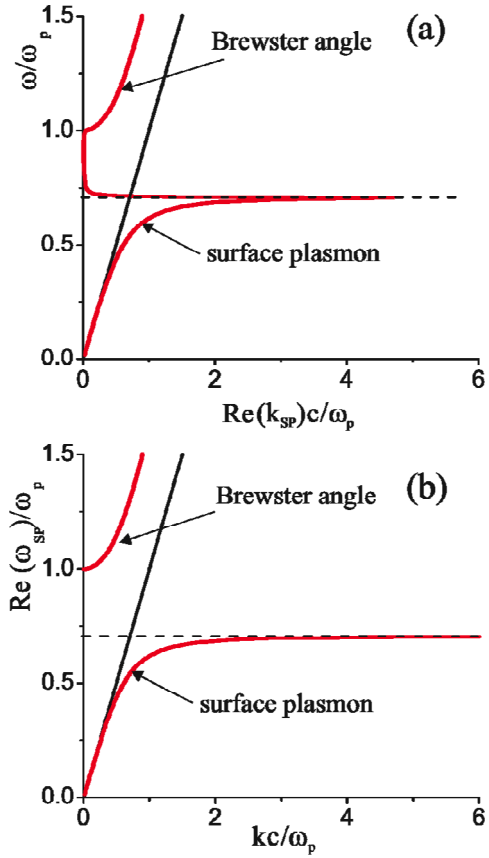
There is a spatial frequency cut-off for imaging applications!

Discussion

Local density of states
Which choice ? Real or complex K?

-i) The Green's tensor gives the answer : the LDOS diverges

ii) When counting states in k-space, K is real. We use modes with real K. It follows that the dispersion relation diverges.



Archambault Phys. Rev. B 79 195414 (2009)

Huygens-Fresnel principle for surface plasmons

Huygens-Fresnel principle for SP

$$\mathbf{E}^{SP}(x,y) = \int \frac{dk_y}{2\pi} \boxed{\mathbf{E}^{SP}(k_y)} \boxed{e^{i\sqrt{k_{SP}^2 - k_y^2}x + ik_y y}}.$$

$$\mathbf{E}^{SP}(x,y) = \int dy' \; E_z^{SP}(x=0,y')\mathbf{K}(x,y,y'),$$

$$\mathbf{K}(x,y,y') = \begin{bmatrix} K_x \\ K_y \\ K_z \end{bmatrix} = -\frac{1}{2} \begin{bmatrix} \frac{k_z}{k_{SP}^2} \frac{\partial^2}{\partial x^2} H_0^{(1)}(k_{SP}\rho) \\ \frac{k_z}{k_{SP}^2} \frac{\partial^2}{\partial x \partial y} H_0^{(1)}(k_{SP}\rho) \\ i\frac{\partial}{\partial x} H_0^{(1)}(k_{SP}\rho) \end{bmatrix}$$

The SPP is completely known when the z-component is known

Teperik, Opt. Express (2009)

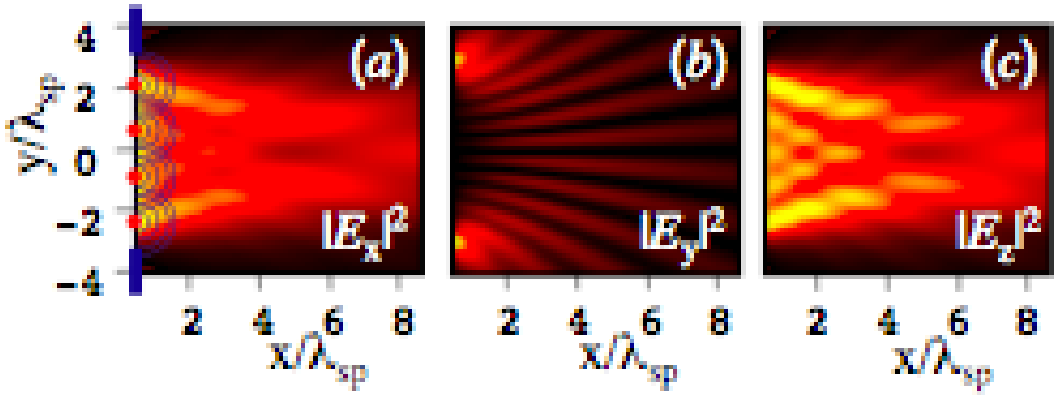
Huygens-Fresnel principle for SP

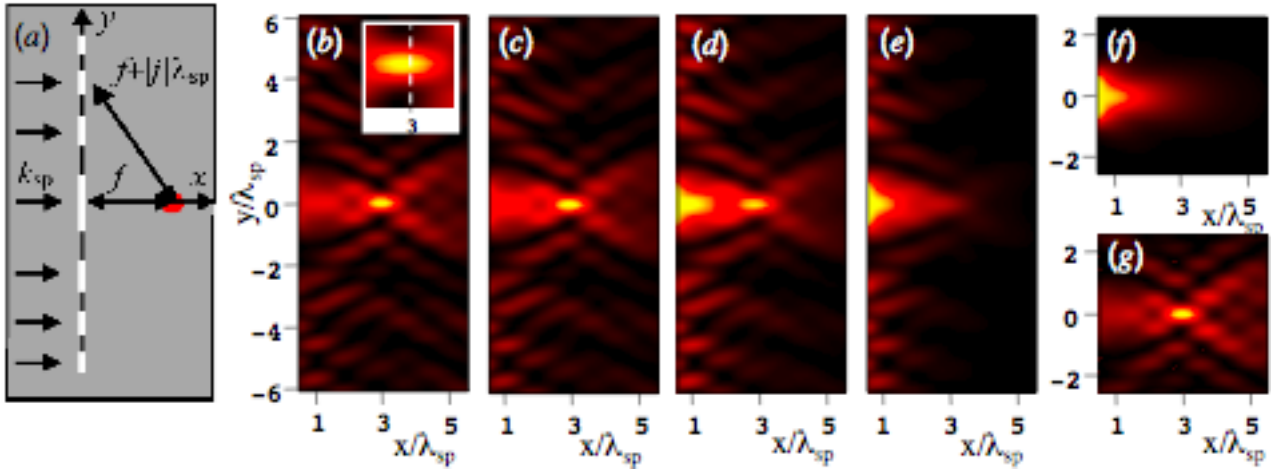
Asymptotic form

$$E_z^{SP}(x,y) =$$

$$= -\frac{i}{\sqrt{\lambda_{SP}}} \int dy' \ \cos\theta \ E_z^{SP}(x=0,y') \frac{e^{ik_{SP}\rho}}{\sqrt{\rho}} e^{i\pi/4}$$

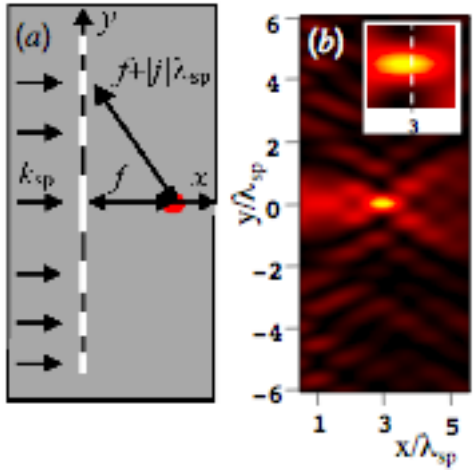
Huygens-Fresnel principle for SP

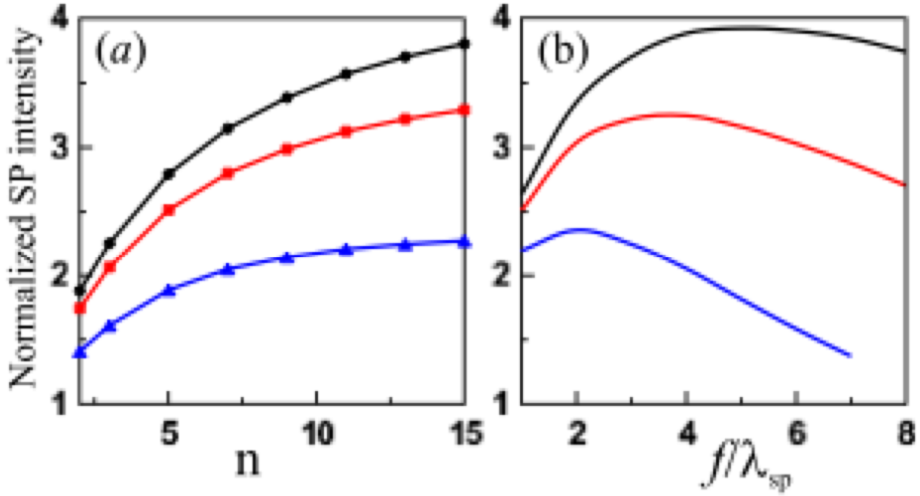




Huygens-Fresnel principle for SP

Influence of the number of apertures and the focal distance on the intensity at focus





Thank you !