Introduction to Surface Plasmon Theory

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Outline

A A few examples of surface plasmons

B Surface waves

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

C Plasmons

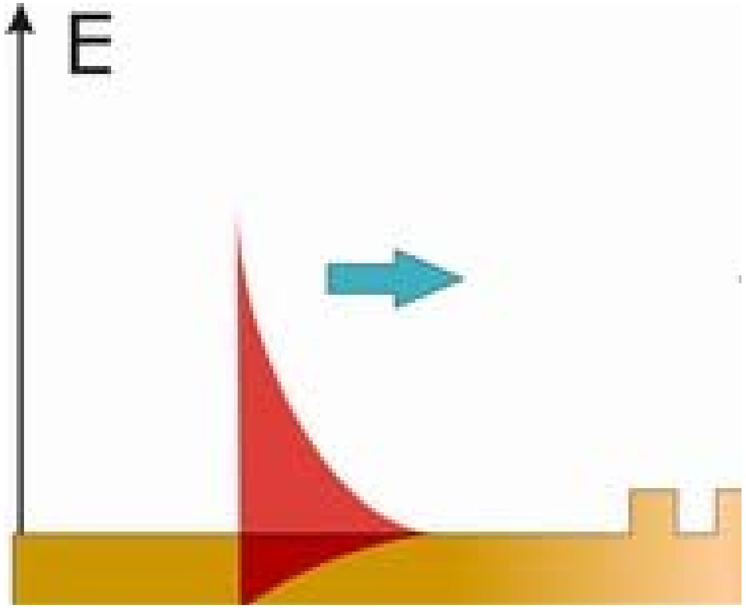
D Surface plasmon polariton (SPP)

F Key properties of SPP

G SPP in lossy metals

J Fourier optics for surface plasmons

What is a surface plasmon polariton?



$$E_0 \exp[ikx - i\gamma z - i\omega t]$$

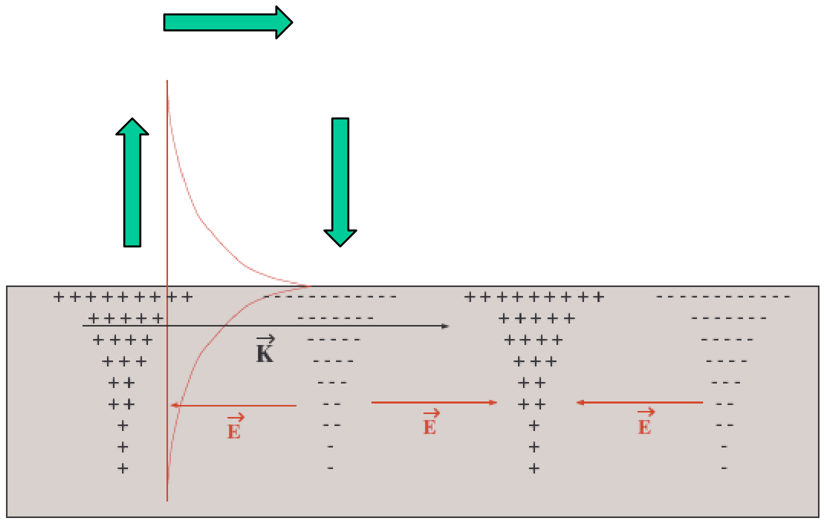
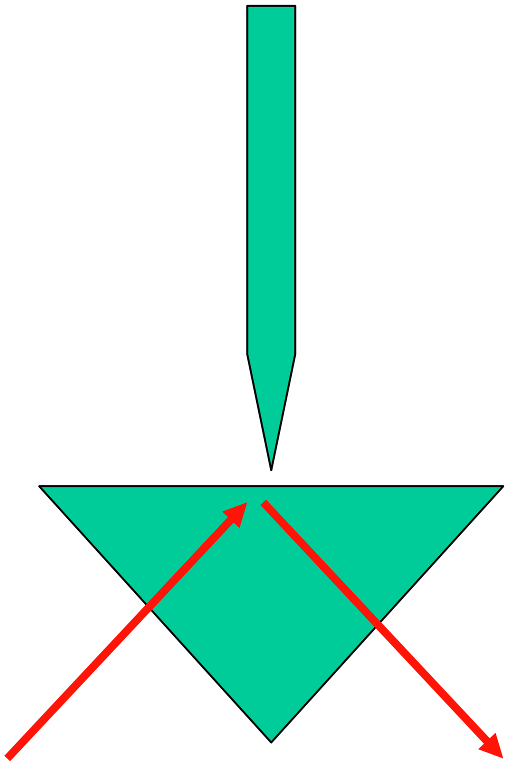
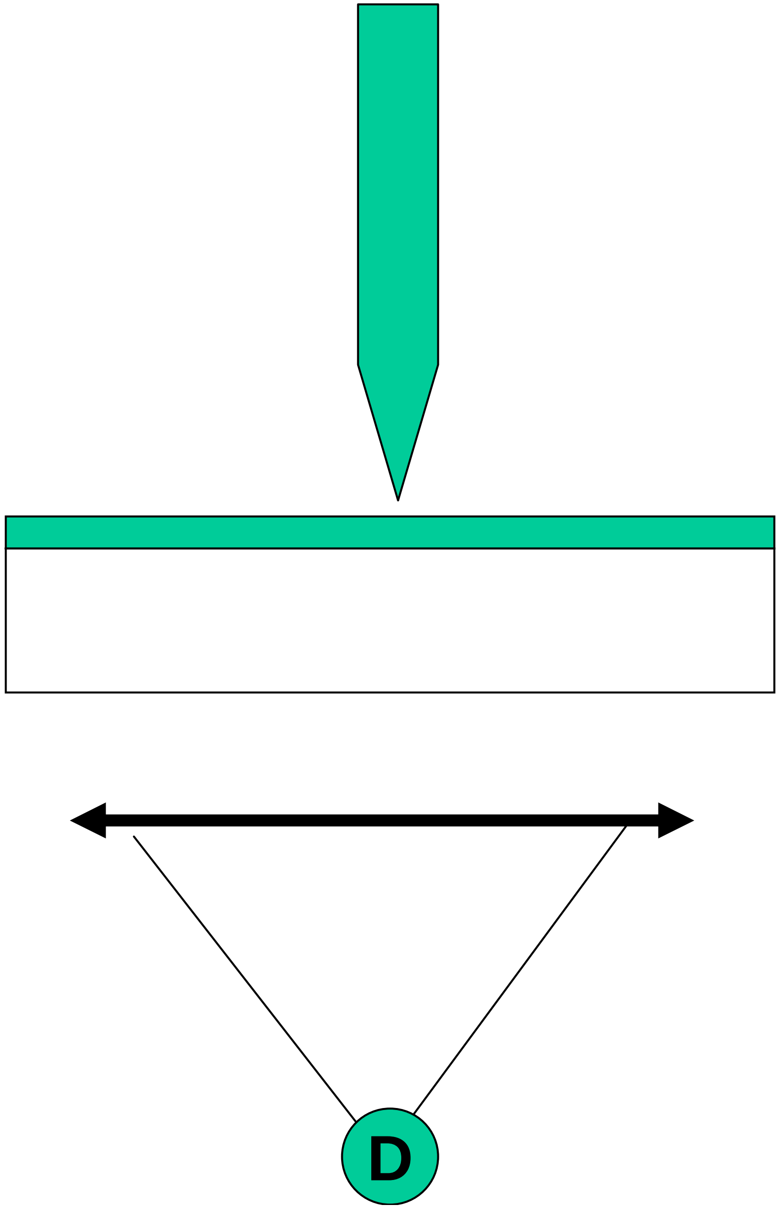


Image of a SPP

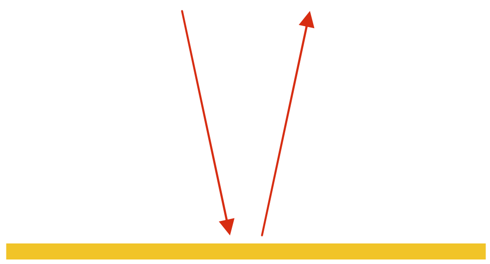


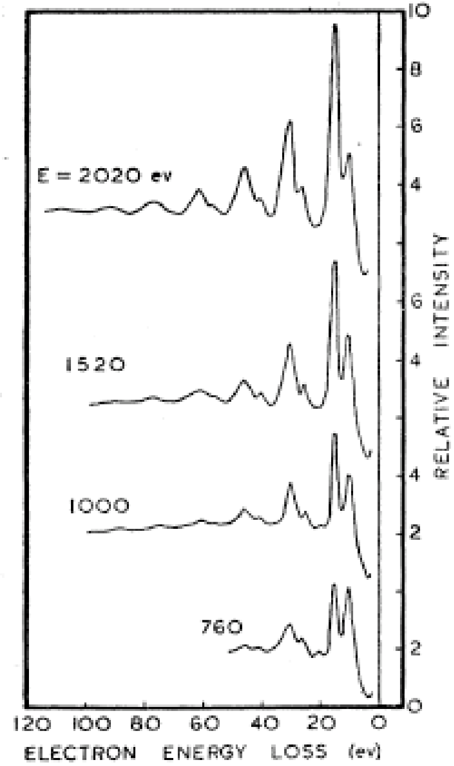
Dawson PRL 94

Excitation using a sub-λ source



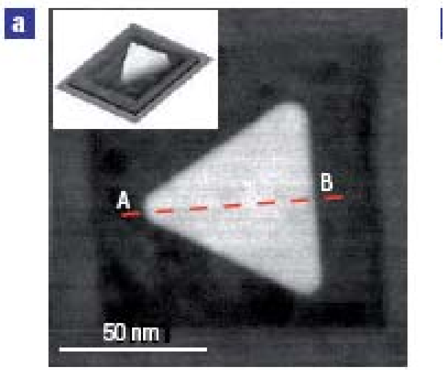
EELS (Electron Energy Loss Spectroscopy) of reflected electrons.

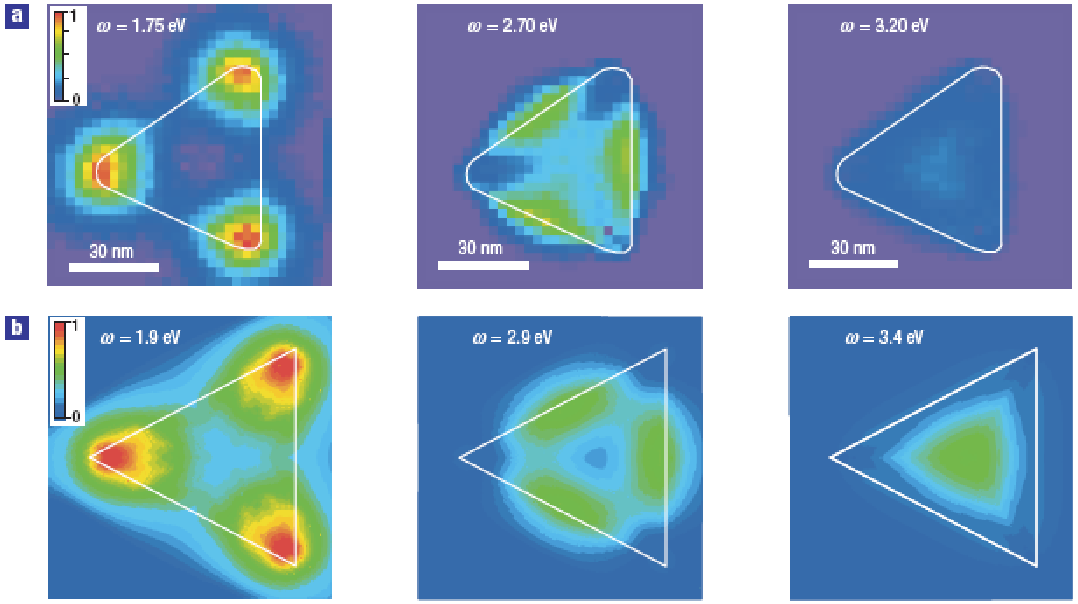




Powell, Phys.Rev. 1959

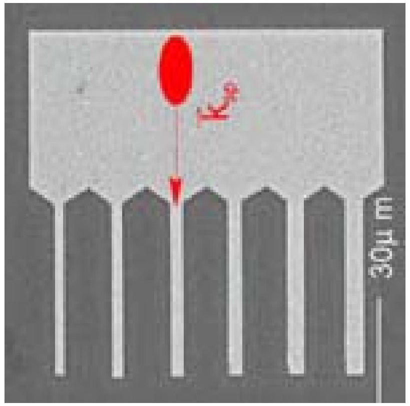
Observation of the LDOS using EELS





Nelayah et al. Nature Physics, (2007)

Metal stripes as SPP guides



Université de Bourgogne

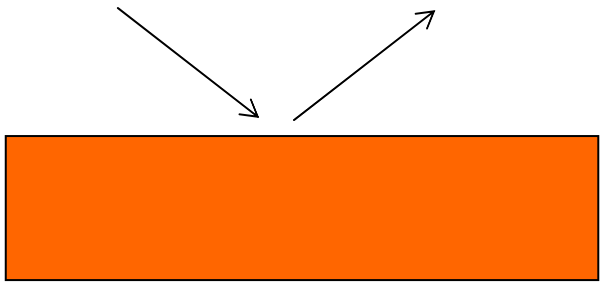
What is a Surface Wave (1)?

Derivation of the dispersion relation

0. Surface wave

1. Solution of a homogeneous problem

2. Pole of a reflection factor



Poles and zeros

Dispersion relation

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

Reflection factor

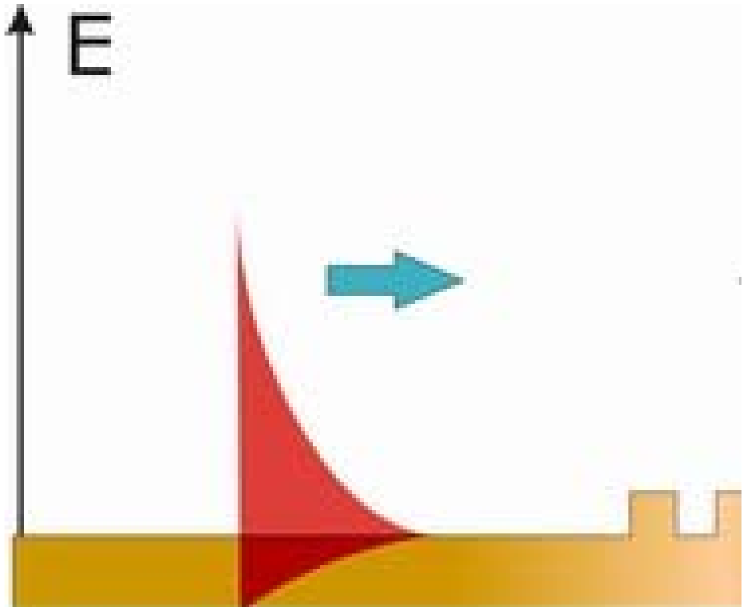
$$r_F = \frac{\varepsilon_2 k_{z1} - \varepsilon_1 k_{z2}}{\varepsilon_2 k_{z1} + \varepsilon_1 k_{z2}}$$

$$K^2 = \frac{\omega^2}{c^2}\frac{\varepsilon}{\varepsilon+1}$$ **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 Wave (2)?

Structure of the wave



$$E_x \exp\left[ikx - i\gamma z - i\omega t\right]$$

What is a Surface Wave (3)?

1. **Case of a good conductor**

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

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

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

What is a Surface Wave (4)?

Historical account of the surface wave concept

Long radio wave propagation : the hypothesis of Zenneck

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

Norton approximate formula

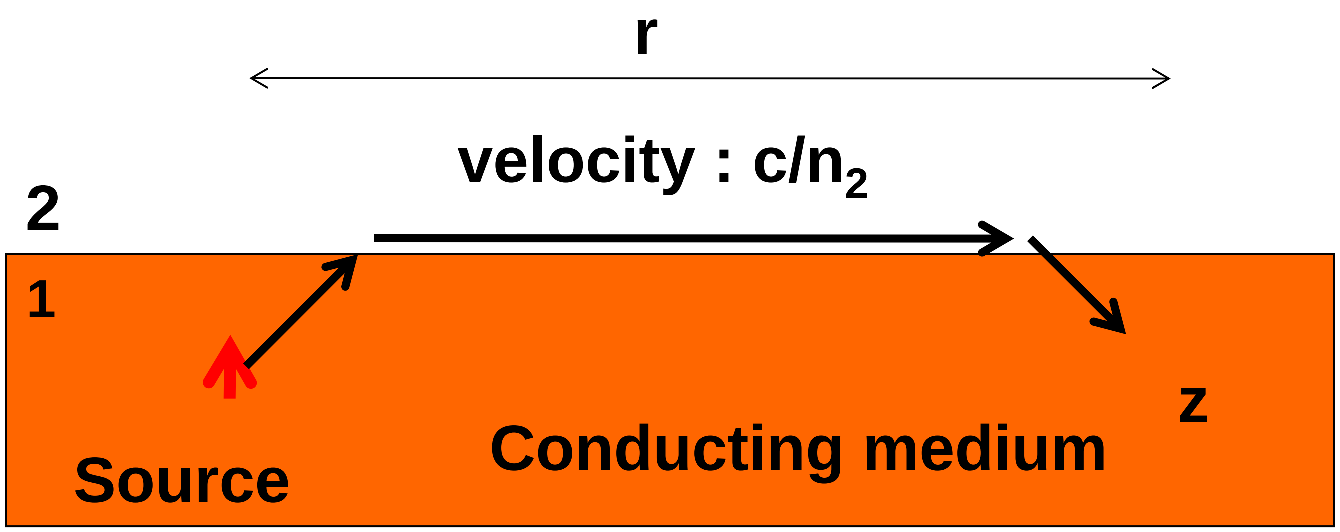
Banos contribution

Lateral wave

What is a lateral wave ?

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

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



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

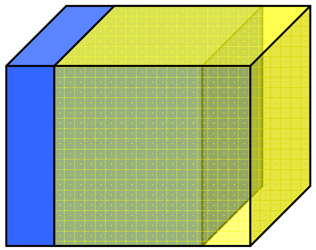
R. King, Lateral electromagnetic waves, Springer Verlag, NY, 1992

Surface wave and surface plasmon

Question : when a surface wave is a surface plasmon ?

What is a plasmon?

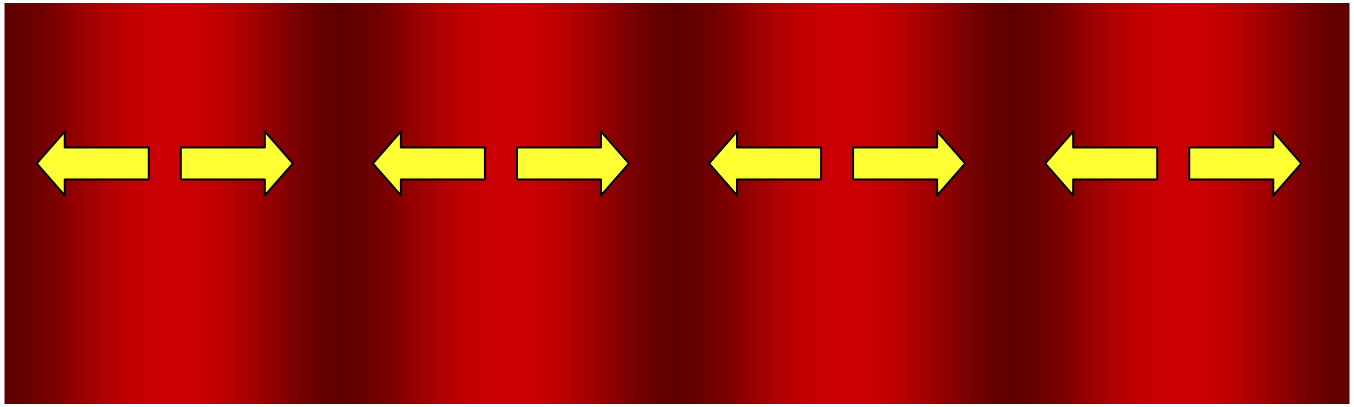
First example : a thin film
vibrational collective mode of oscillation of electrons



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

What is a (bulk) plasmon polariton?

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



(Bulk) Plasmon dispersion relation

Hydrodynamic model

$$div\ \mathbf{E} = \frac{\rho}{\varepsilon_0}$$

$$\frac{\partial \rho}{\partial t} + \mathbf{v} \cdot \nabla \rho = -\nabla P - \rho \mathbf{E}$$

$$P = -\frac{\rho}{e} k_B T$$

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

Electrodynamic point of view

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

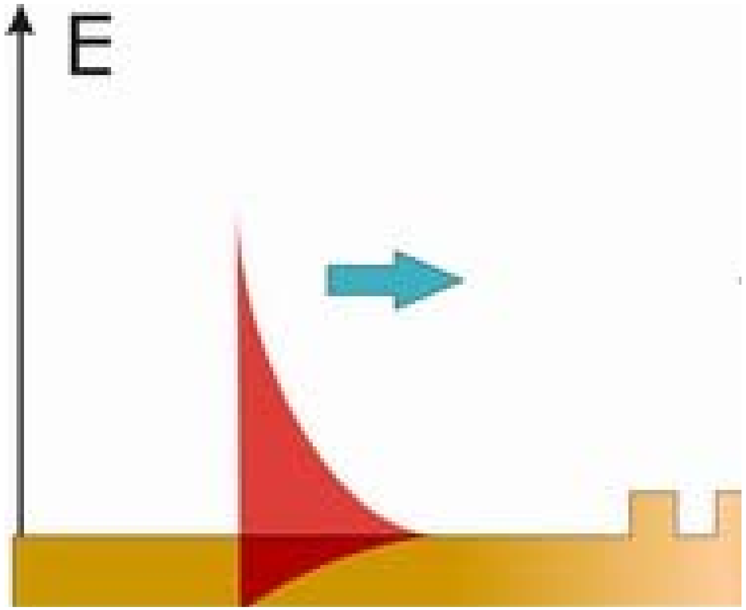
$$\varepsilon_r(\omega)\ k \cdot E(k,\omega) = 0$$

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

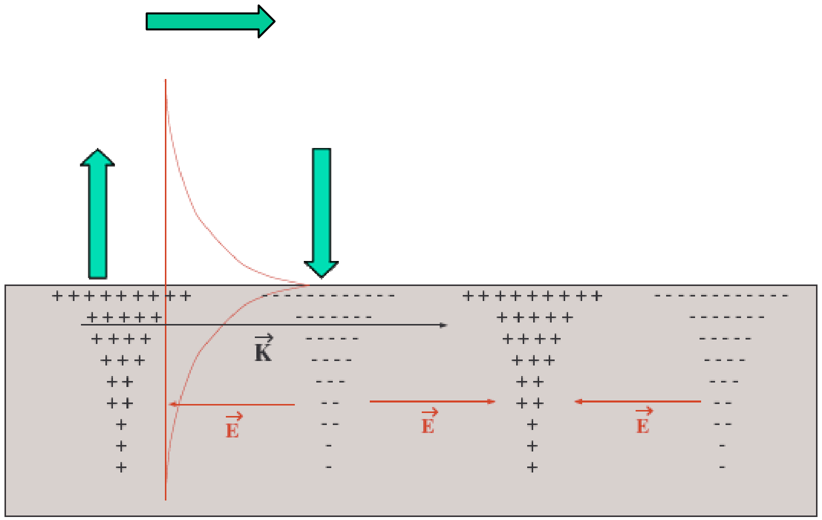
An electron gas has a mechanical vibration eigenmode that generates a longitudinal EM mode.
Key idea : plasmon is a material resonance.

What is a Surface Wave (2)?

Structure of the wave



$$E_x \exp\left[ikx - i\gamma z - i\omega t\right]$$



Elliptic polarization with a (geometrically) longitudinal component. (but transverse wave)

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

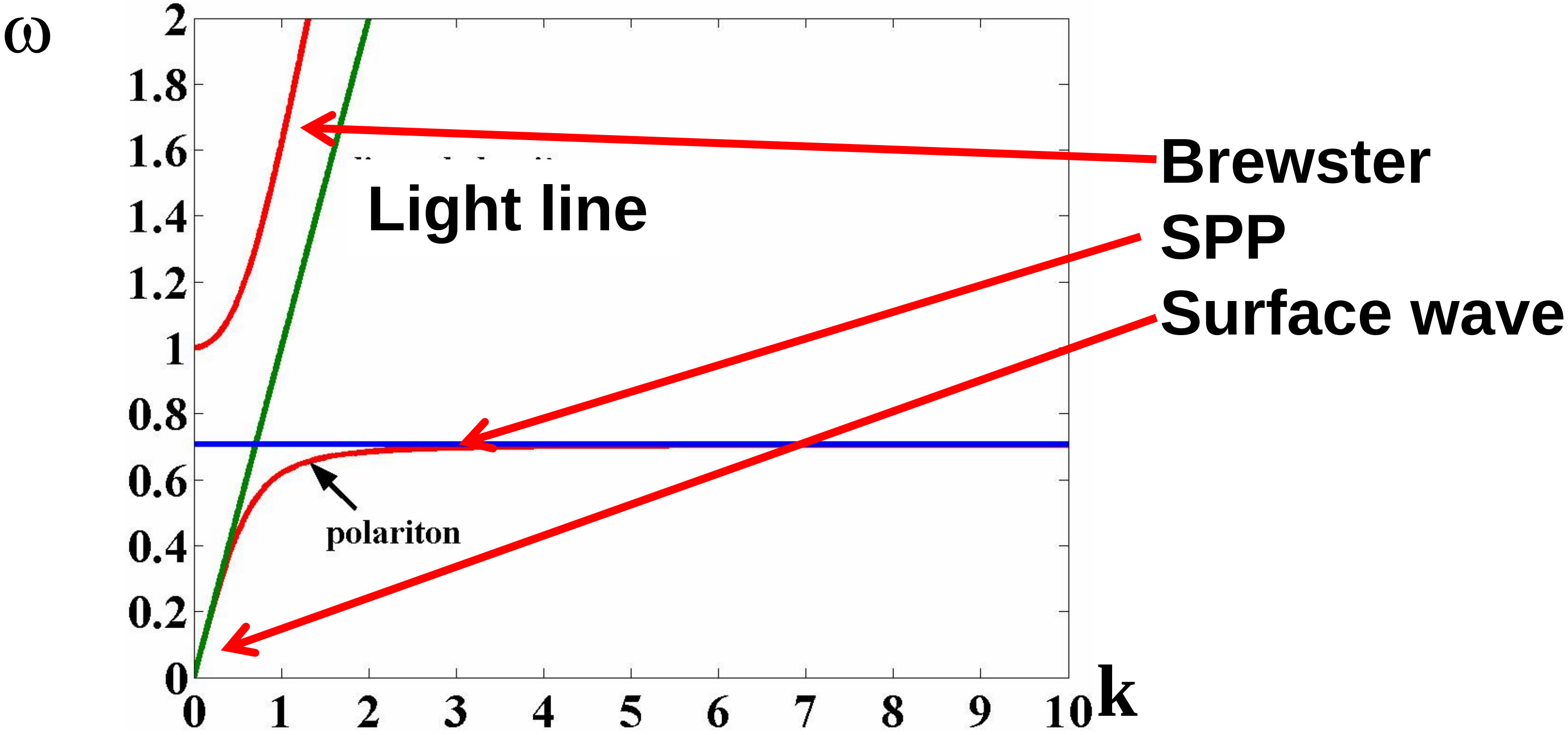
$$\varepsilon_r(\omega) = 1 - \frac{\omega_p^2}{i\omega\gamma} \approx i\frac{\omega_p^2}{\omega\gamma} = i\frac{\sigma}{\omega\varepsilon_0}$$

Overdamped oscillation

Surface plasmon polariton?

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

Drude model



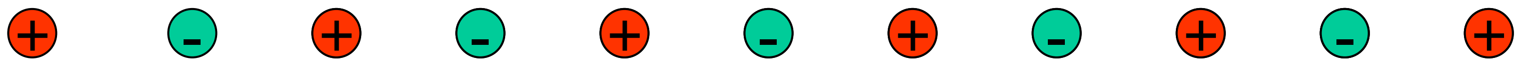
Remark : no surface plasmon in metals at THz frequencies

Non local correction

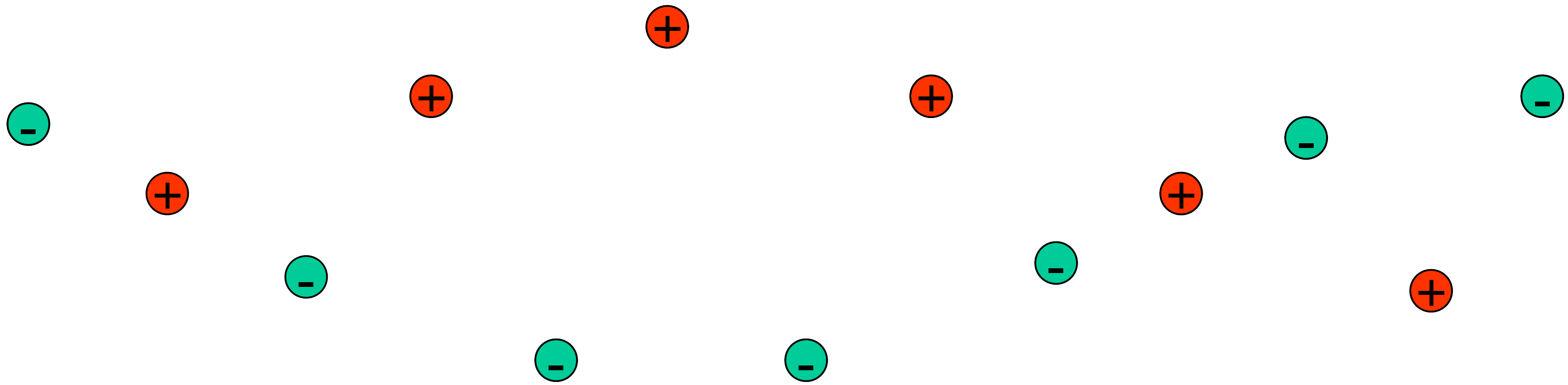
How good is a macroscopic analysis of the problem?
What are the relevant length scales ?

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

Phonon polariton

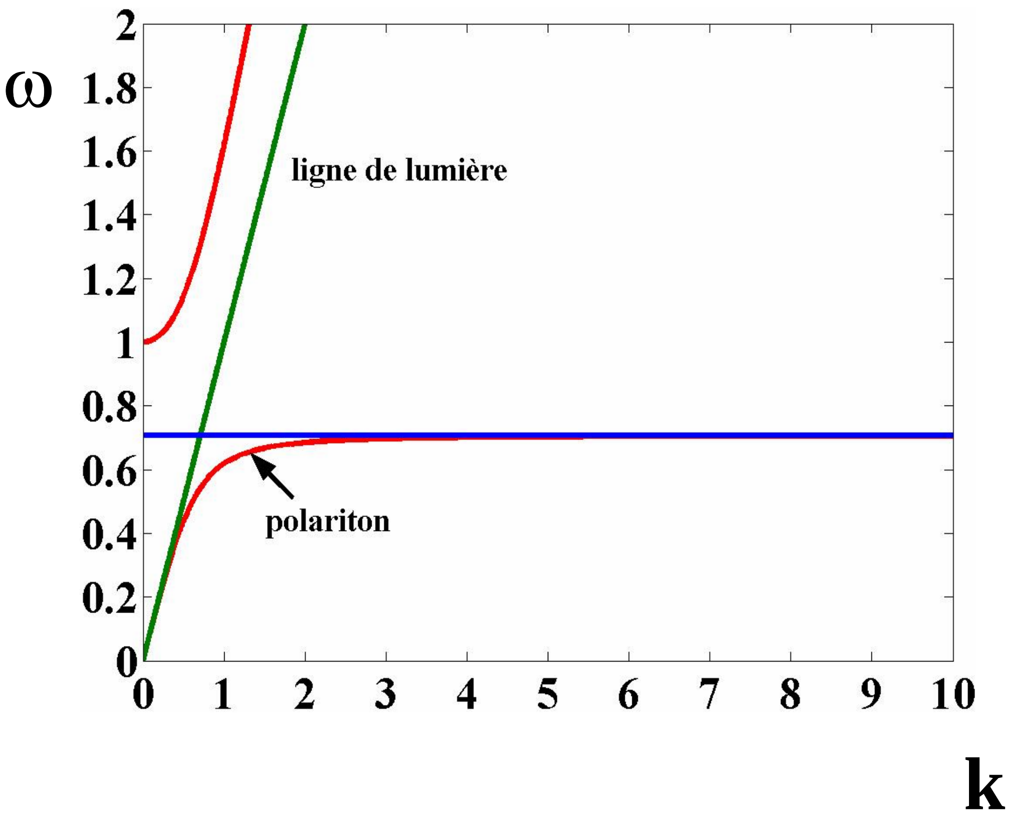


Phonon polariton



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

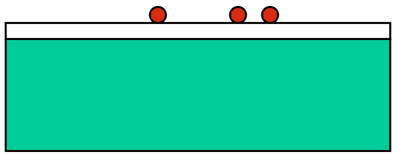
$$A_{21} = B_{21} \frac{\hbar\omega^3}{\pi^2 c^3} = \left[B_{21}\hbar\omega\right] \frac{\omega^2}{\pi^2 c^3}$$

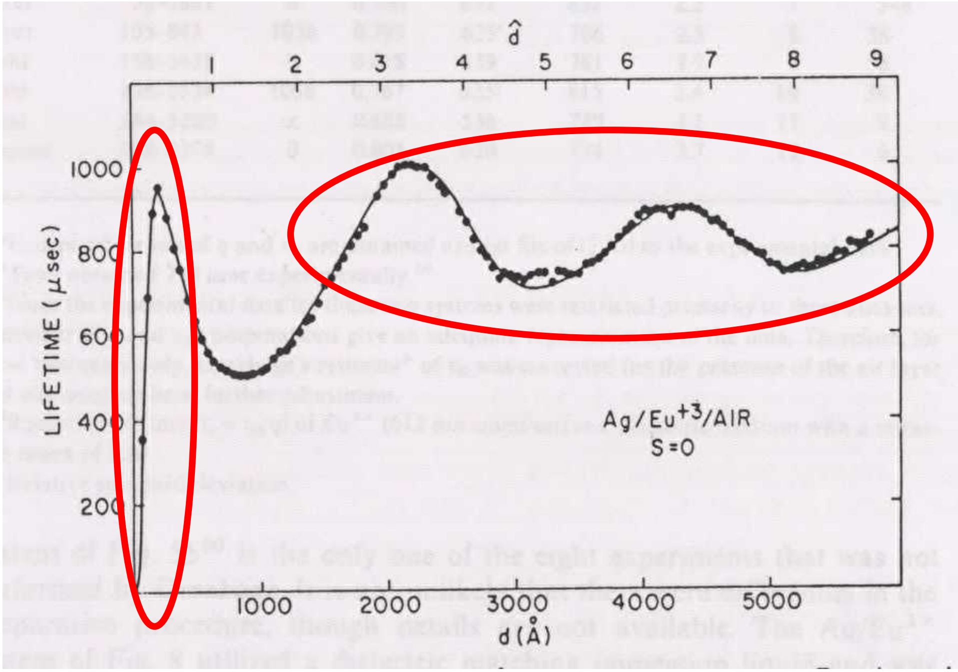
Larger LDOS means : i) shorter lifetime, ii) larger energy at thermodynamic equilibrium

LDOS and Spontaneous emission

Lifetime

SPP





Interferences

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

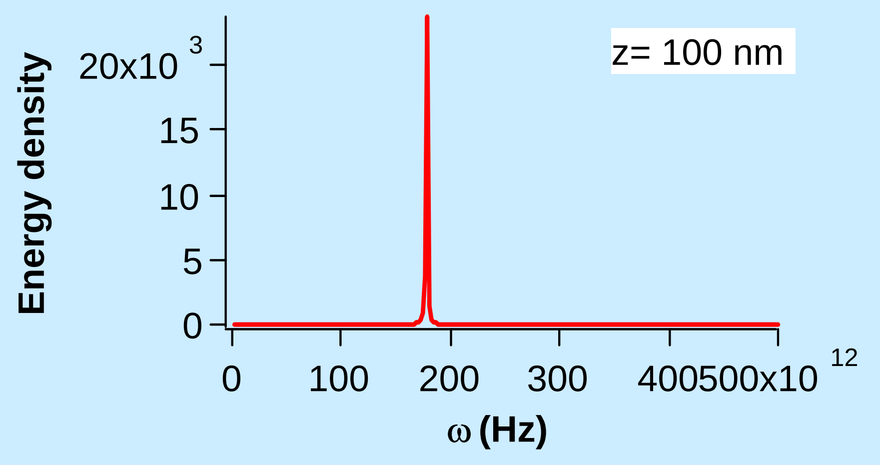
Lifetime is not intrinsic but depends on the environment

Asymptotic form of the EM- LDOS

Near-field form

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

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



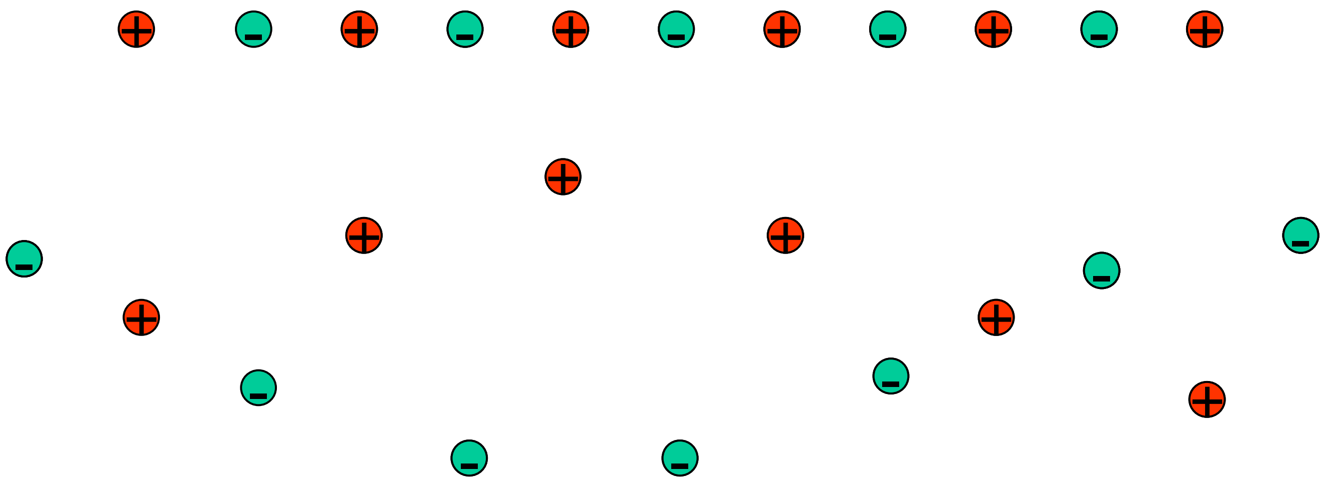
Signature of the SPP ?

PRL 85, 1548 (2000)

PRB 68, 245405 (2003)

Where are the *new* modes coming from?

The EM field inherit the density of states of matter : SPP are polaritons !



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

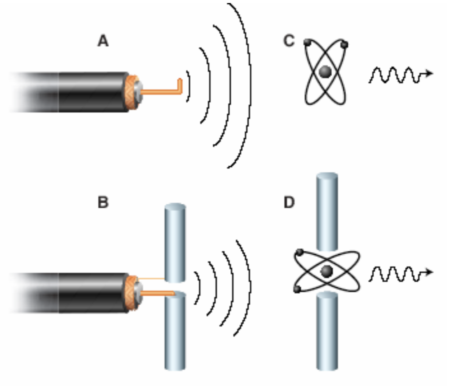
$$\frac{N}{V} = \int_0^{\omega} g(\omega')d\omega' = \frac{\omega^3}{3\pi^2 c^3} \qquad N \approx \frac{V}{\lambda^3}$$

Estimate of the number of electrons/phonons:

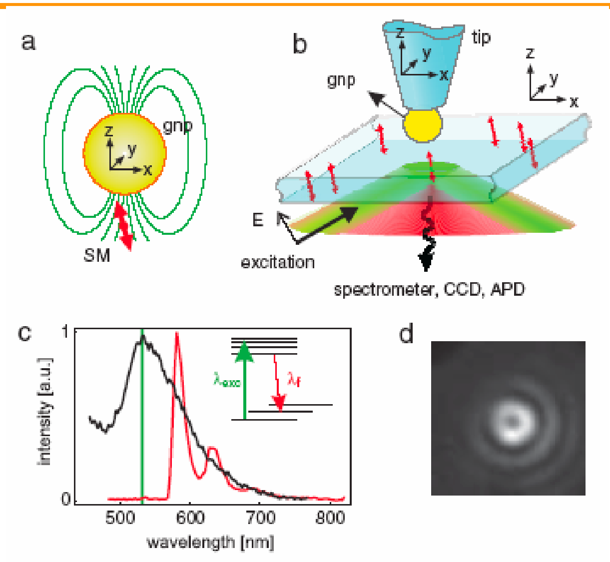
$$N \approx \frac{V}{a^3}$$

The EM field inherits the large DOS of matter.

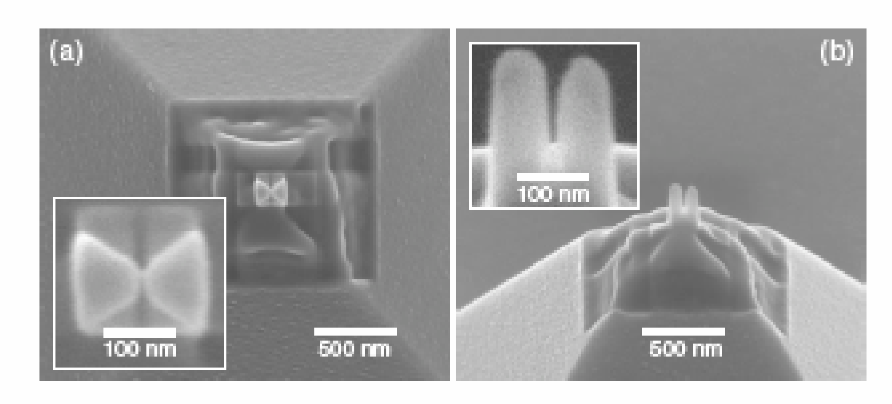
Application : nanoantenna



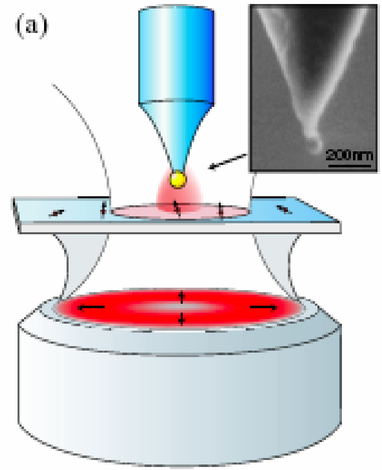
Mühlschlegel et al. Science 308 p 1607 (2005)
Greffet, Science 308 p (2005) p 1561



Kühn et al. PRL 97, 017402 (2006)



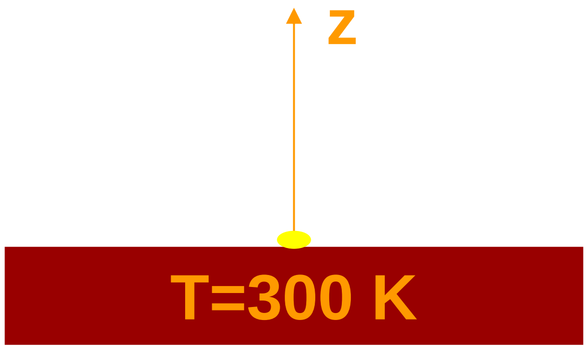
Farahani et al., PRL 95, 017402 (2005)

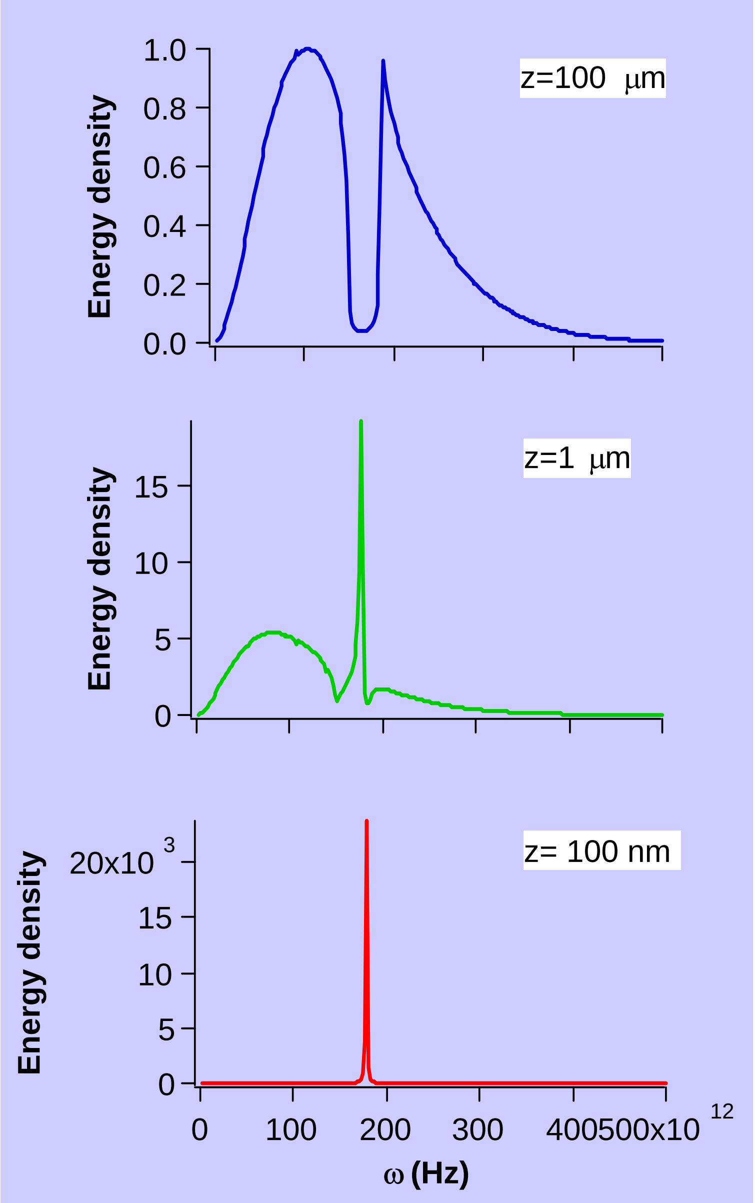


Anger et al., PRL 96, 113002 (2006)

LDOS and energy

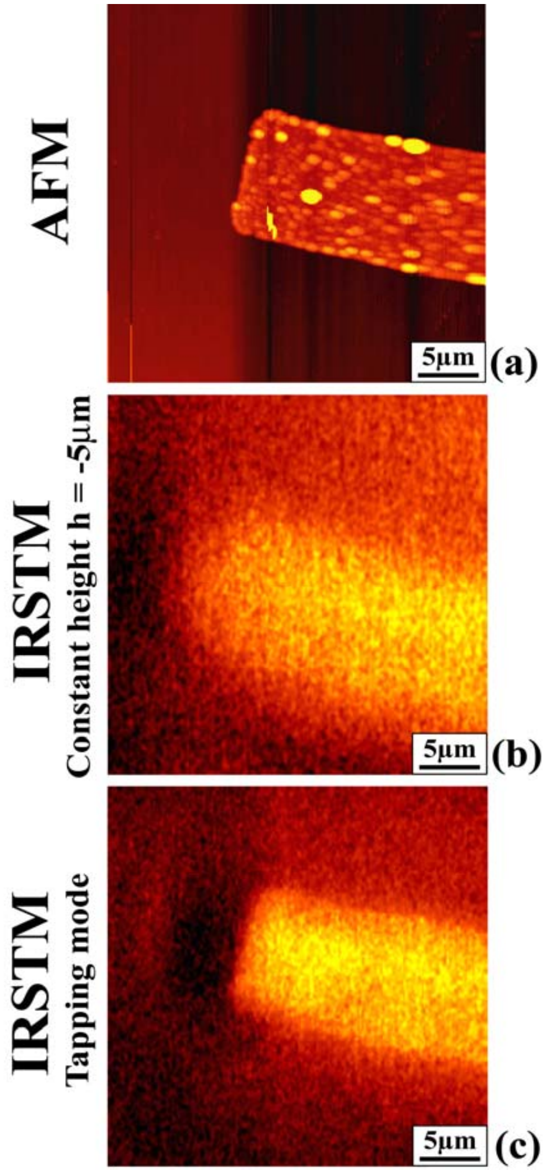
Energy density close to the surface



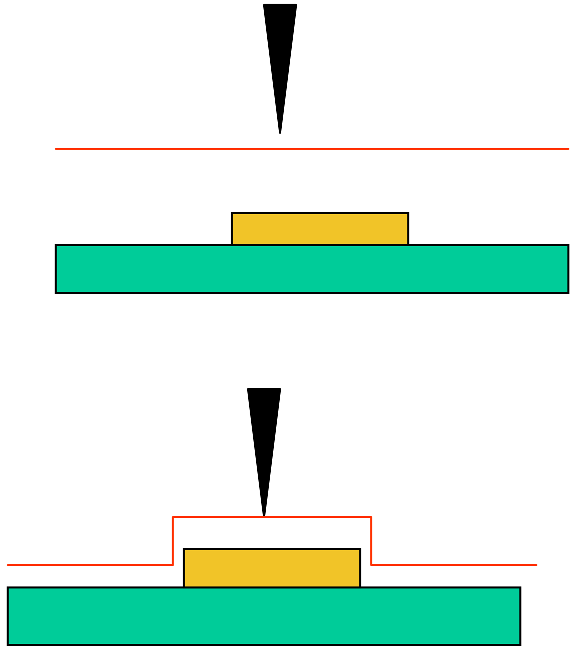


Shchegrov PRL, 85 p 1548 (2000)

LDOS and energy

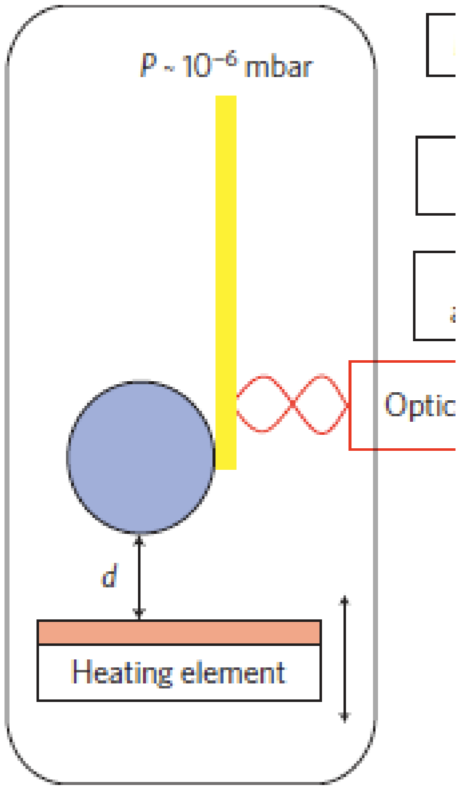


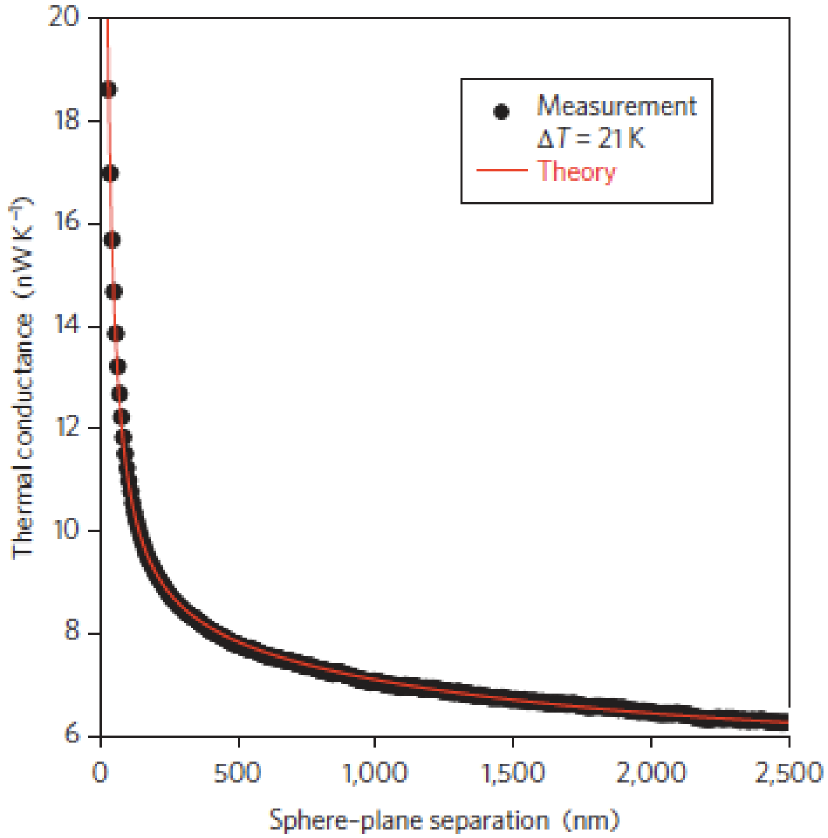
Observation of the thermal near field



De Wilde et al., Nature 444 p 740 (2006)

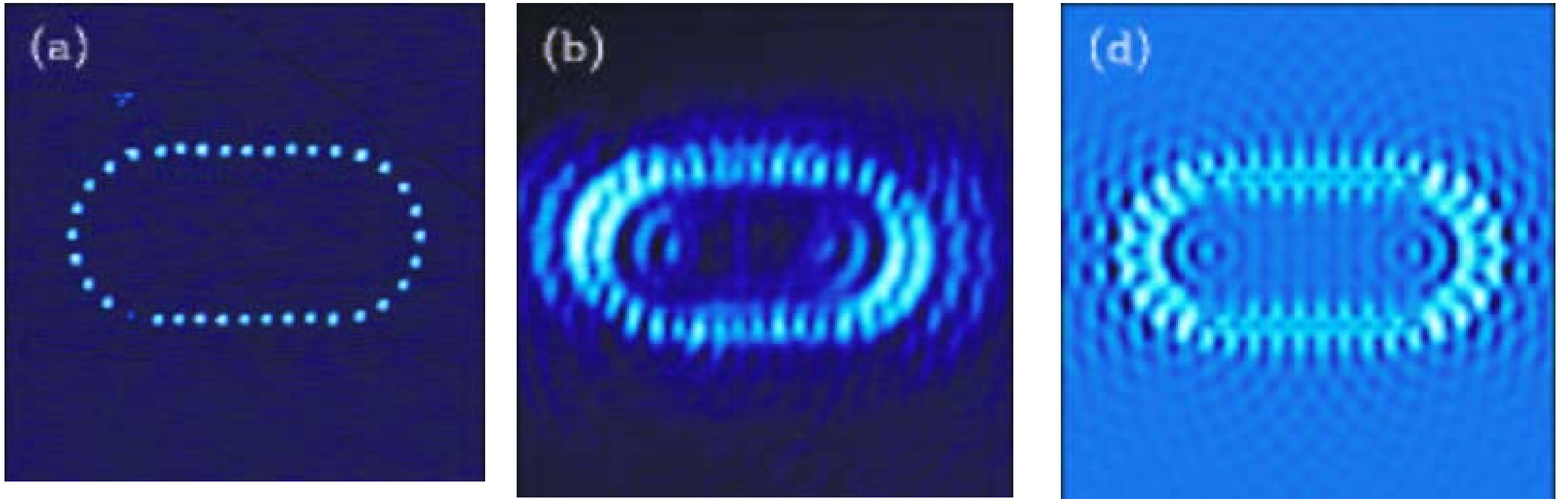
Application : nanoscale heat transfer

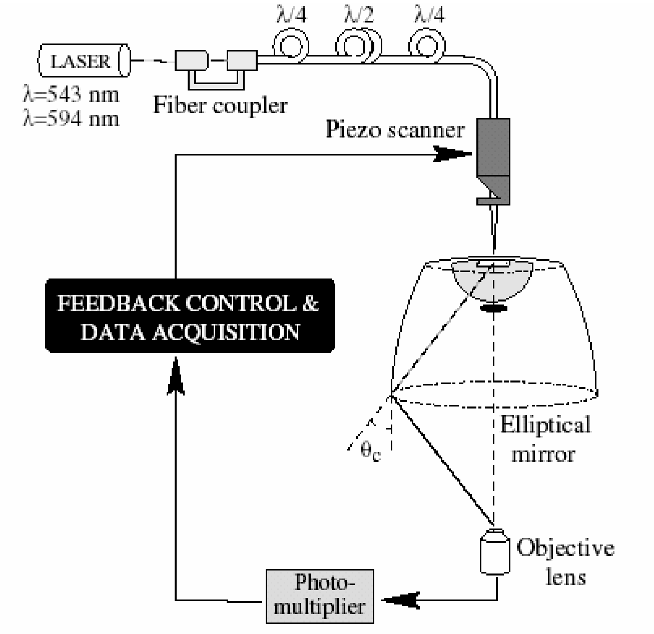




E. Rousseau, Nature Photonics, (2009), DOI 10.1038/Nphoton.2009.144

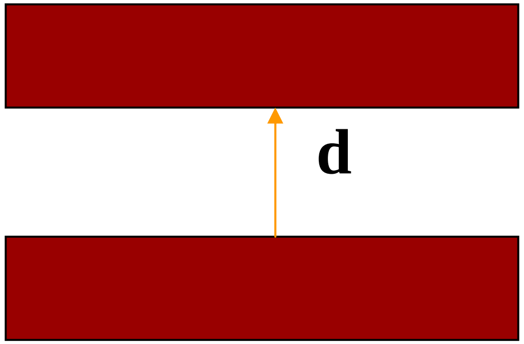
Observation of the SPP LDOS

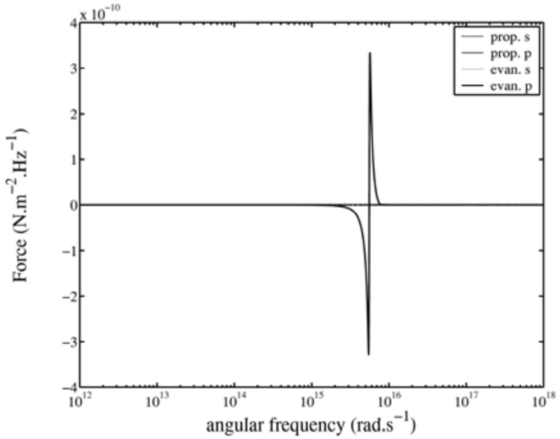




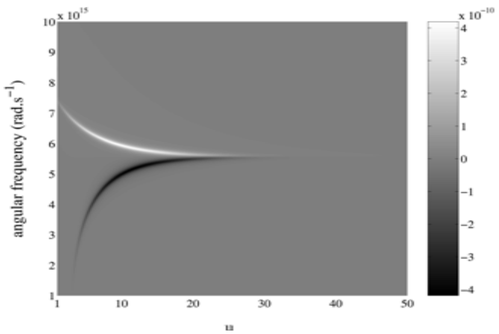
C. Chicanne et al., Phys. Rev. Lett. 88, 97402 (2002)

SPP LDOS and Casimir force

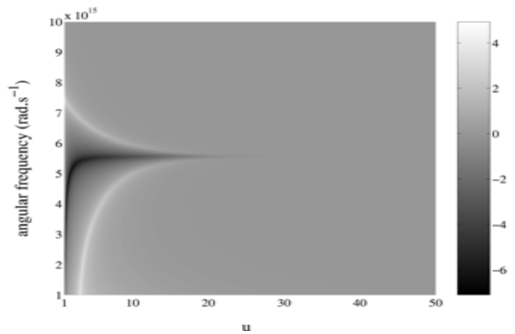




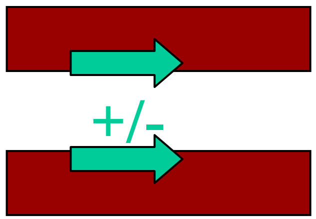
$$F = \int F(k,\omega)\, d^3k\, d\omega$$



Force



Dispersion relation



Joulain, Phys. RevA 69, 023808 (2004)

Remark

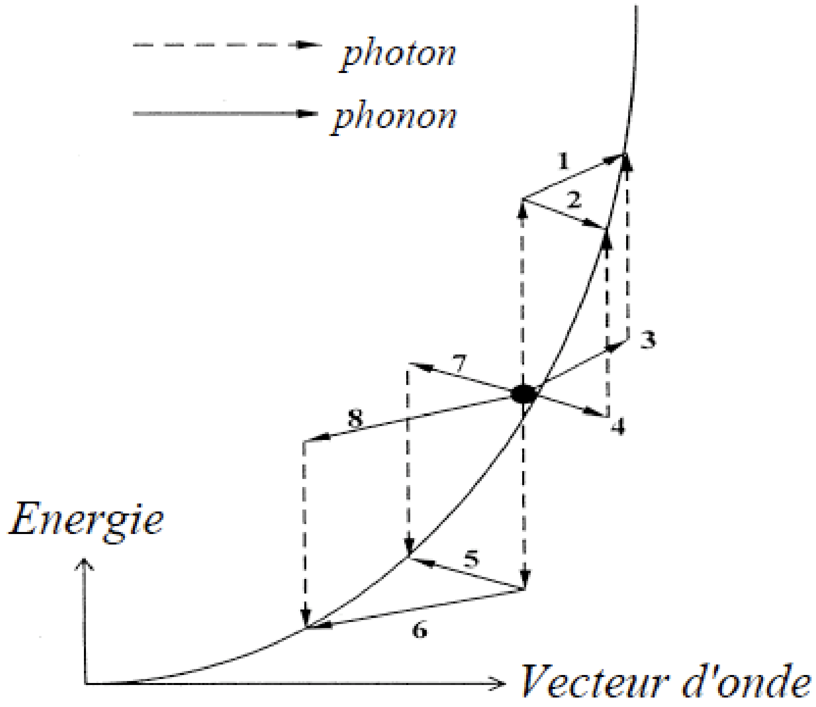
LDOS and projected LDOS

SPP key properties 2

Fast relaxation/Broad spectrum

Losses in noble metals (1)

Intraband loss Mechanism



Different mechanisms at high frequency and low frequency

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

Losses in noble metals (2)

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

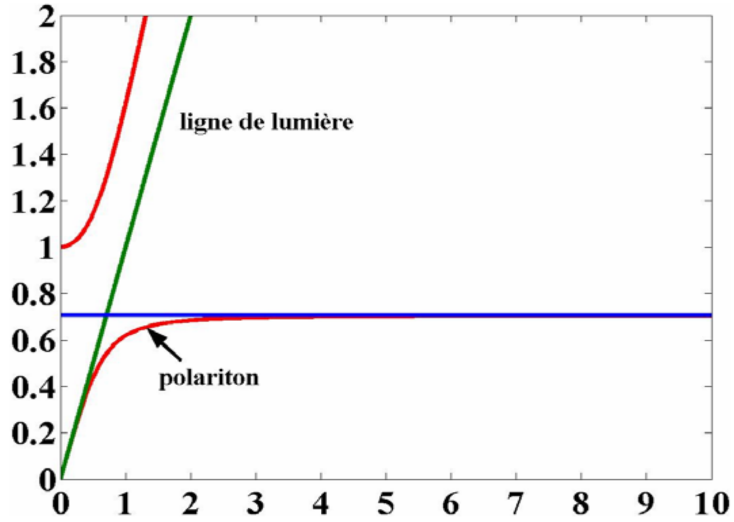
Adv. in Phys. 33 p 257 (1984)
Phys.Rev.B 25 p 923 (1982)
Phys.Rev.B 3 p 305 (1971)

Applications :

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

SPP key properties 3

Field confinment



Field confinment by particles, tips

Electrostatic or SPP confinment ?

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

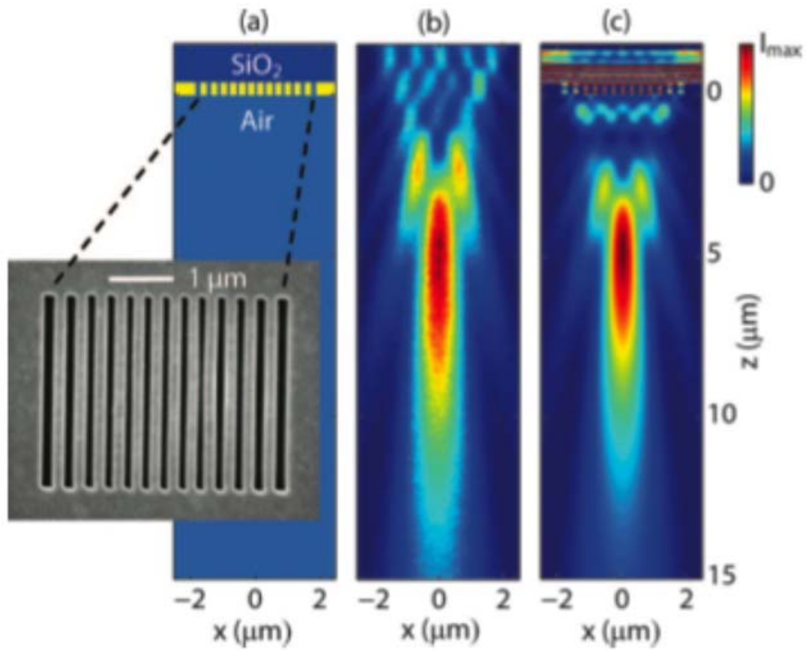
Field confinment by particles, tips

Electrostatic or SPP confinment ?

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

Look for a resonance close to ω_p.

SPP focusing



Nano Lett., 2009, 9 (1), 235-238• DOI: 10.1021/nl802830y

Fourier optics of surface plasmons

Archambault Phys. Rev. B 79 195414 (2009)

Surface plasmon

Solution for a non-lossy medium

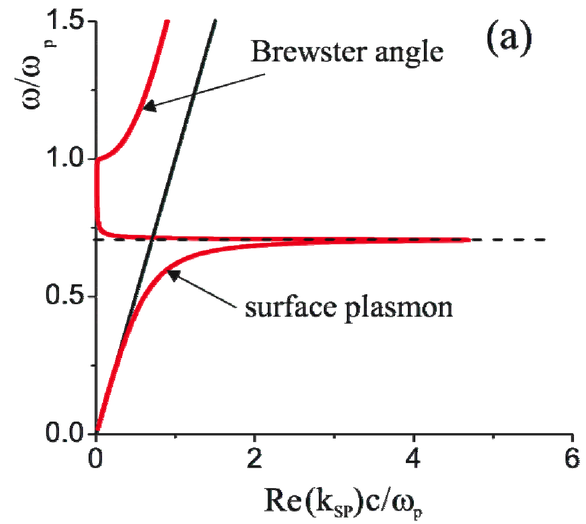
$$\mathbf{E}(z)\exp[i(K_x x + K_y y - \omega t)]$$

Dispersion relation

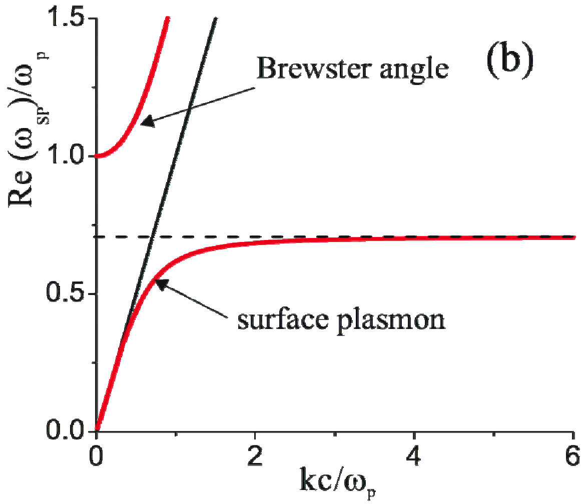
$$K^2 = \frac{\omega^2}{c^2}\frac{\varepsilon}{\varepsilon+1}$$

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

Surface plasmon dispersion relation

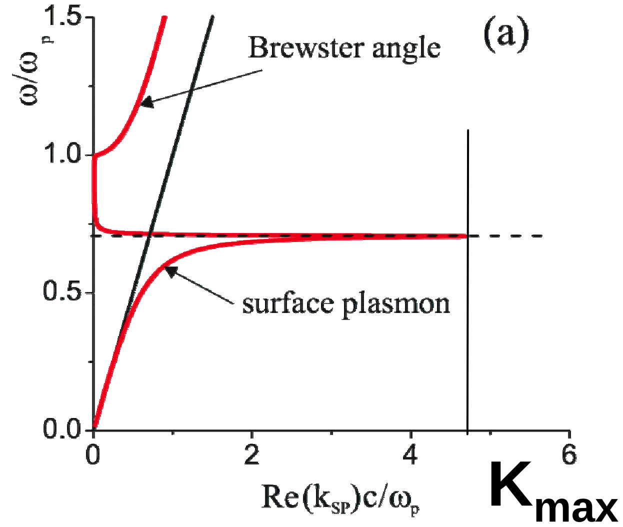


Real ω and complex K

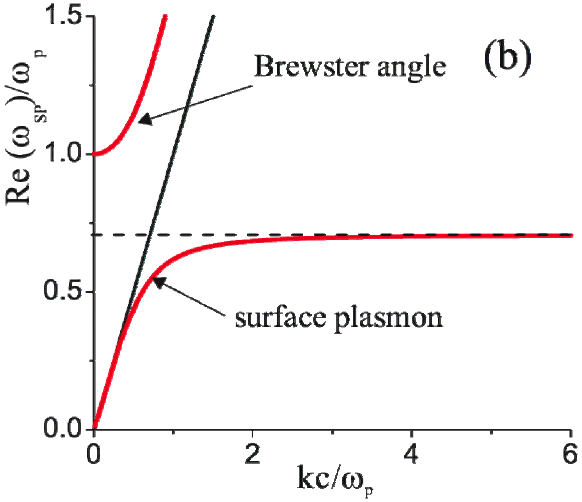


Real K and complex ω.

Maximum confinment of the field ?

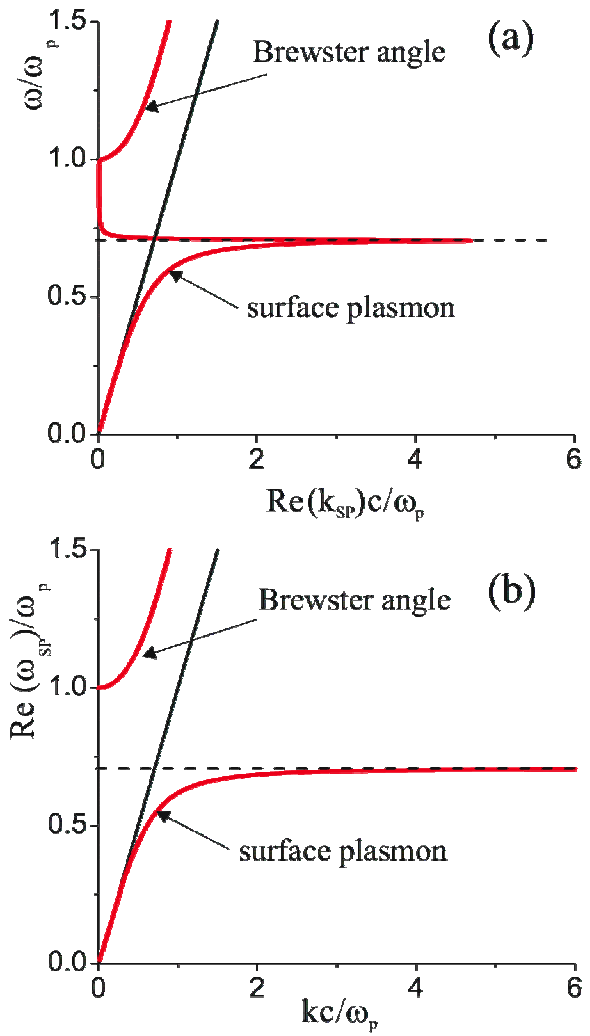


$1/K_{max}$



No limit !

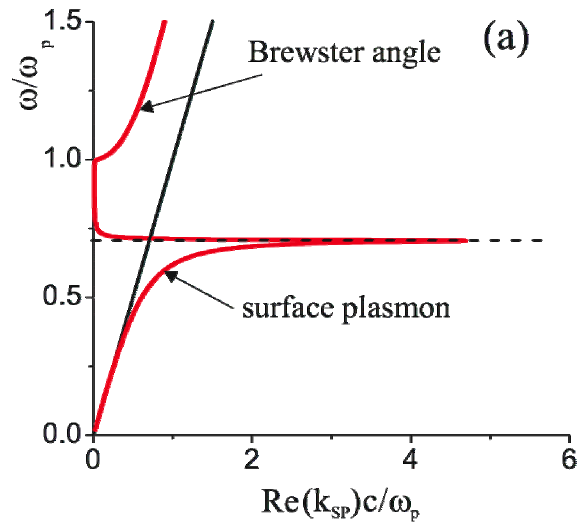
Local Density of states?

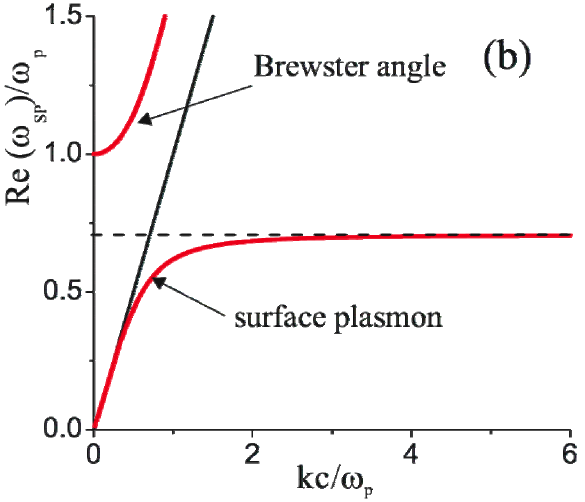


Finite value of the LDOS

Divergence of the LDOS

First analysis of the backbending





Analysing ATR experiments

Data taken at fixed angle while varying K

Data taken at fixed K while varying the frequency.

Fundamentals of Fourier Optics

The field is a superposition of plane waves:

$$\Psi(x,y,z) = \iint \Psi(\alpha,\beta,0)e^{i(\alpha x+\beta y+\gamma z)}\frac{\mathrm{d}\alpha}{2\pi}\,\frac{\mathrm{d}\beta}{2\pi}$$

$$\alpha^2+\beta^2+\gamma^2 = \frac{\omega^2}{c^2}$$

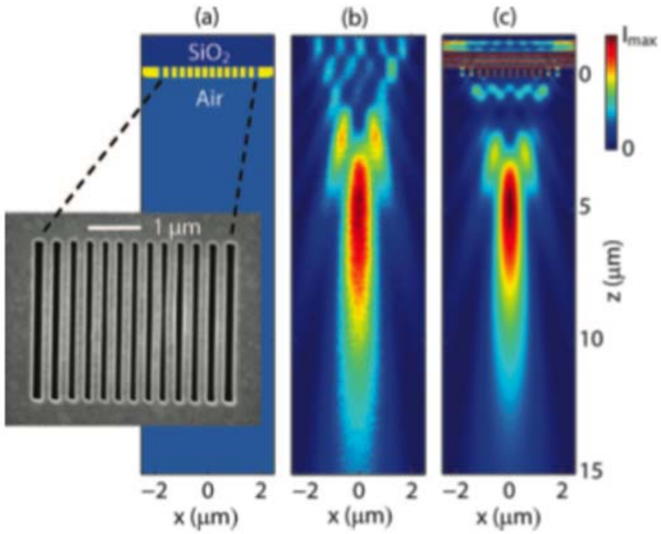
Propagation and diffraction can be described as linear operations on the spatial spectrum.

Propagation is a low-pass filter : resolution limit.

Equivalent (Huygens-Fresnel) form :

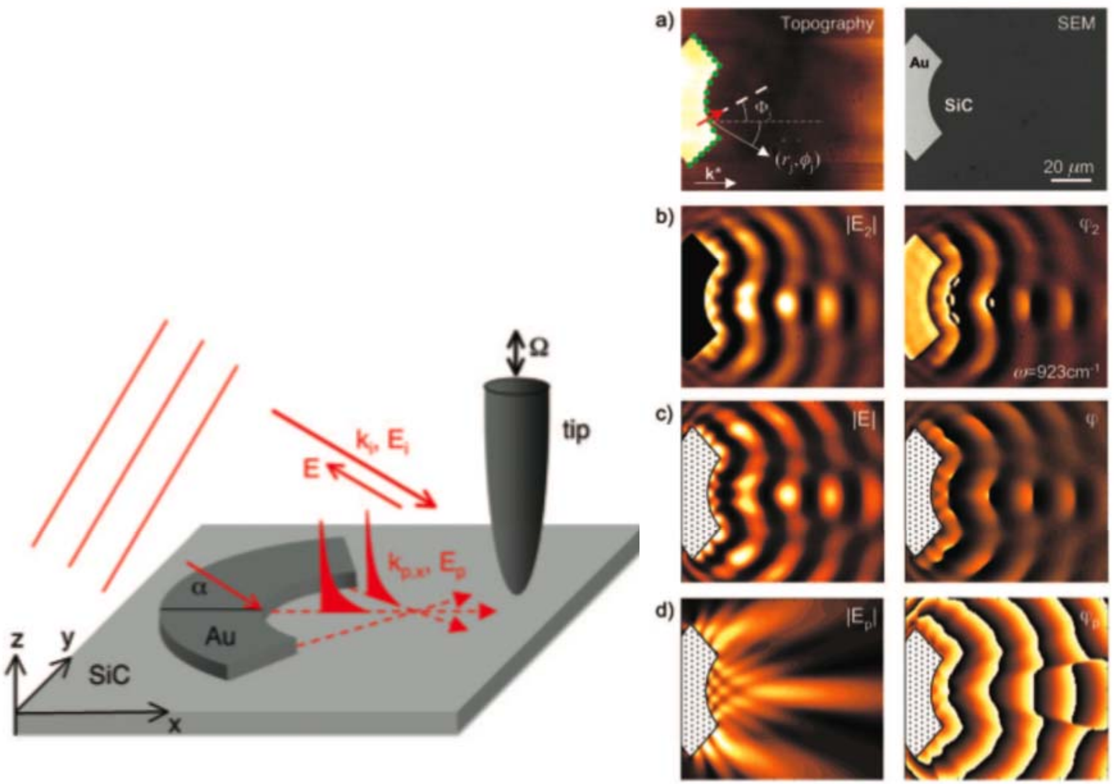
$$\Psi(x,y,z) = -\frac{1}{2\pi}\iint \Psi(x',y',0)\frac{\partial}{\partial z}\left[\frac{\exp(ikr)}{r}\right]\mathrm{d}x'\,\mathrm{d}y'$$

Surface plasmon Fourier optics



Brongersma, Nanolett, 2009

Hillenbrand, APL08



Key issues

Huygens-Fresnel propagator for surface plasmon ?

Linear superposition of modes with complex K ?
Linear superposition of modes with complex ω ?

Implication for the maximum confinment.
Implication for the LDOS.

Link with Fourier optics.

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

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

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

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

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

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

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

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.

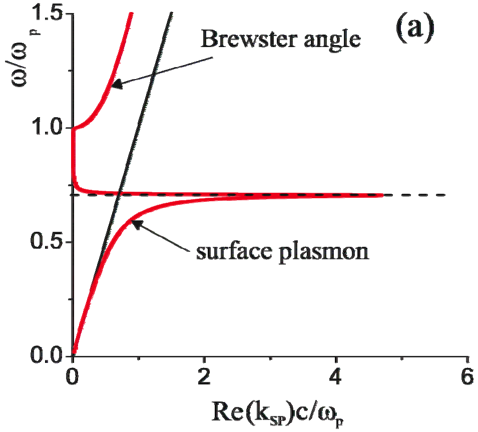
Archambault Phys. Rev. B 79 195414 (2009)

Discussion

What is the best confinment ?

Localized sources and stationary regime :
complex K and real ω

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



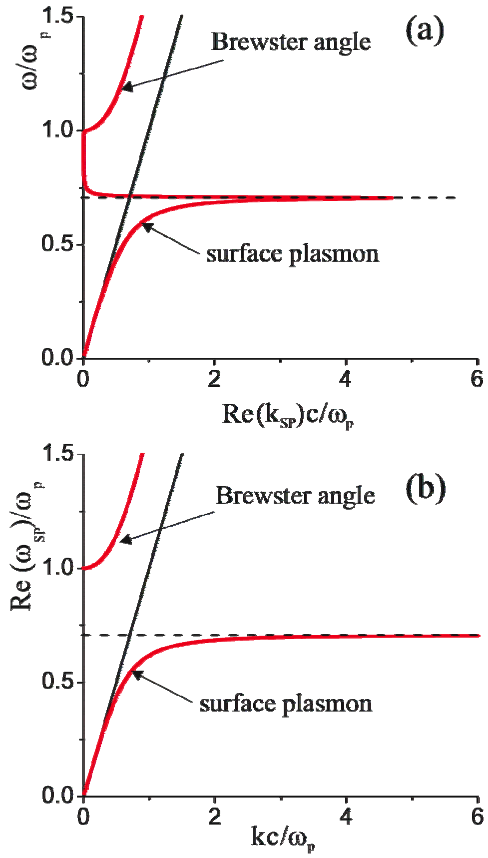
There is a spatial frequency cut-off for imaging applications!

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.



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Huygens-Fresnel principle for surface plasmons

Huygens-Fresnel principle for SP

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

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

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

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

Teperik, Opt. Express (2009)

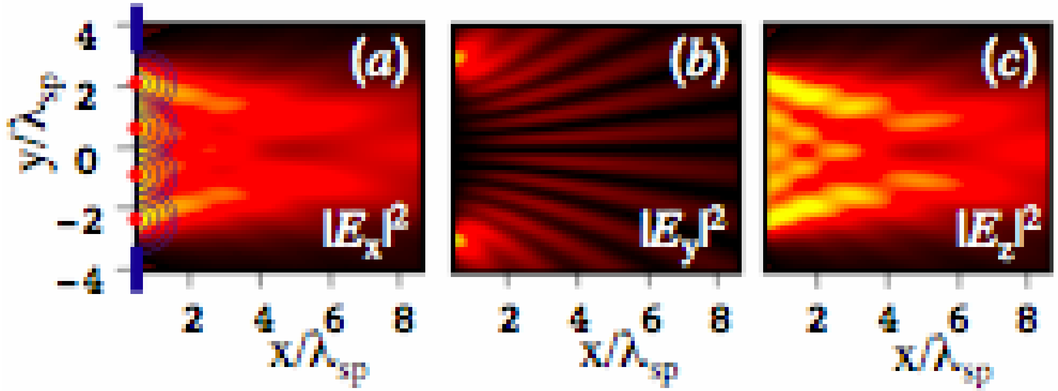
Huygens-Fresnel principle for SP

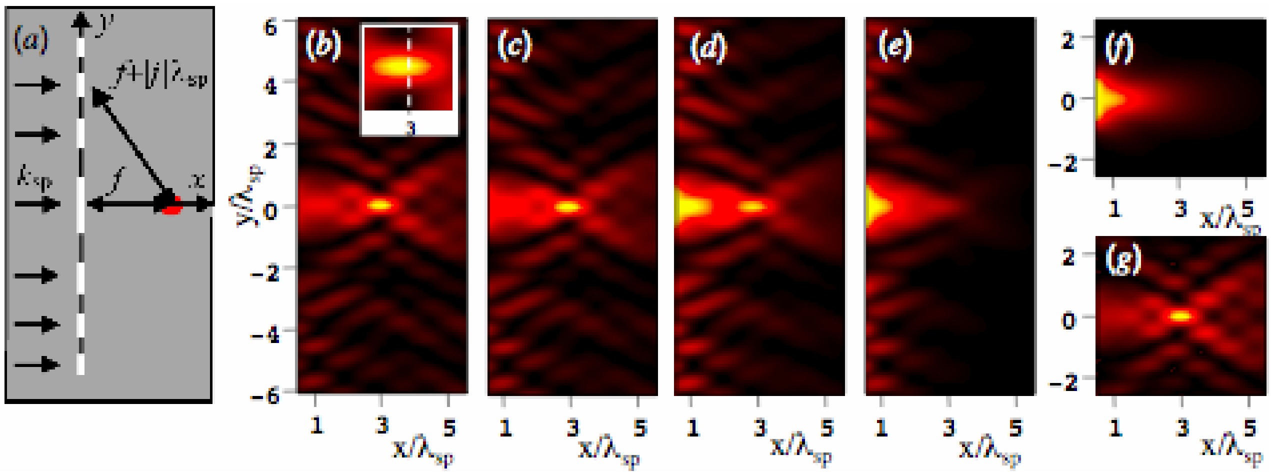
Asymptotic form

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

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

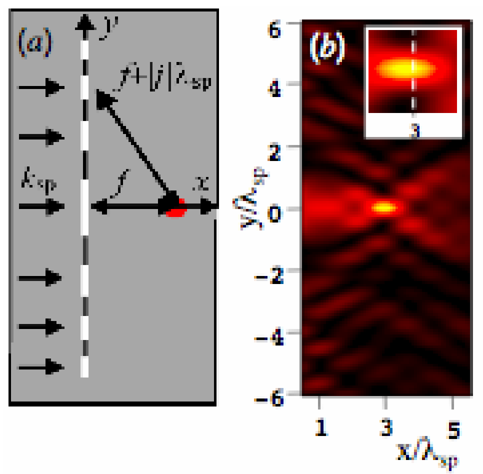
Huygens-Fresnel principle for SP

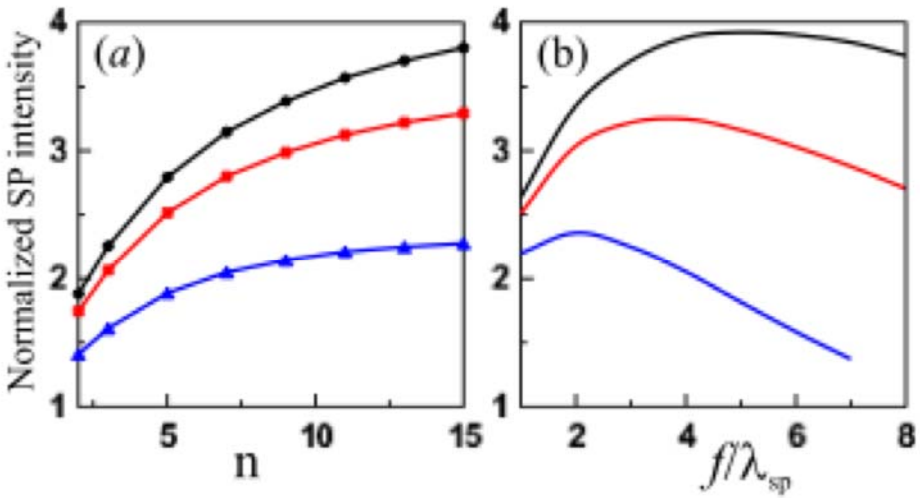




Huygens-Fresnel principle for SP

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





Thank you !