Plasmonics History

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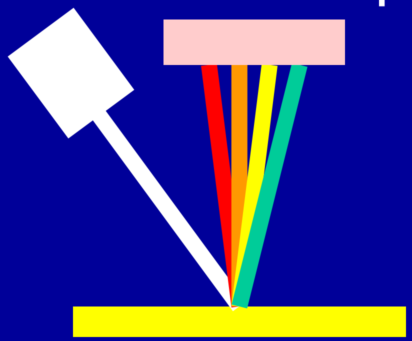
« One of the most interesting problems that I have ever met with » R.W. Wood, 1902

Incandescent lamp

Spectrometer



Ruled grating

« I was astounded to find that under certain conditions, the drop from maximum illumination to minimum, a drop certainly of from 10 to 1, occured within a range of wavelengths not greater than the distance between the sodium lines »

« The singular anomalies were exhibited only when the direction of electric field was at right angle to the ruling »

Summary

- Experimental and theoretical analyses of Wood anomalies: a survey

- What is a surface plasmon? (polariton, plasmon-polariton):
  flat interface, grating

- Why surface plasmons generate Wood anomalies? Phenomenology, total absorption of light by a grating

- Extraordinary transmission through subwavelength holes
- Plasmonics and metamaterials
- Plasmonics and near field microscopy

- Coherent thermic emission of light

- Surface plasmons on non-periodic surfaces: enhanced backscattering, Anderson localization of photons.

Wood anomalies: a survey

First explanation: Rayleigh, 1907

« I was inclined to think that the determining circumstance might perhaps be found in the passing-off of a spectrum on higher order »

Rayleigh observed small but significant discrepancies between the actual locations of anomalies and those deduced from his prediction but explained these discrepancies by a bad knowledge of the grating period

New experimental data for various metallic gratings, J. Strong, 1936

He showed Wood anomalies for various metallic gratings. The results implicitely showed the influence of the metal on the location and shape of the anomalies

(in contradiction with Rayleigh conjecture)

The theoretical breakthrough: U.Fano (1941)

« One may distinguish two anomalies:

- A sharp anomaly, that is, an edge of intensity, governed by a relation discovered by Rayleigh
- A diffuse anomaly extends for a wide interval from the edge towards the red and depends upon the optical constants »

- Fano explained the diffuse anomaly by a « forced resonance » related to the « leaky waves supportable by the grating »

Hessel and Oliner, 1965

They confirmed the conclusions of Fano and tried to propose a numerical demonstration of the existence of a diffuse anomaly.

In fact, the modeling tool was based on an impedance approximation and was unable to provide reliable quantitative results.

J. Hägglund and F. Sellberg, 1965

They compared their experimental measurements on various metallic gratings with numerical results deduced from the Rayleigh expansion method.

Unfortunately, when the numerical results converged, the agreement with experimental data was only qualitative

(location of anomalies)

End of 60’s and begining of 70’s: a double revolution

1- Technology and experimental tools:

Use of laser sources and photoresist layers permitted the discovery and construction of holographic gratings (D. Rudolph and G. Schmahl, 1967, A. Labeyrie and J. Flamand, 1969).

For the first time, the holographic technology provided a rapid and accurate tool for constructing gratings with sub-micronic periods

End of 60’s and begining of 70’s: a double revolution

2- Theory: The opportunity of using the first powerful computers and the strong development of the rigorous electromagnetic theory of gratings made it possible wide numerical studies of Wood anomalies and allowed the first successful quantitative comparisons between experiments and theory.

D. Maystre and R. Petit, 1972, 1973

I elaborated a rigorous integral theory of scattering from metallic diffraction gratings including the representation of the metal properties by its optical index, in contrast with the perfect conductivity model.

The first results showed:

-For s polarized light, the numerical efficiencies of the actual metallic grating deduce from those assuming a perfect conductivity of the metal through a multiplication factor close to the reflectivity of the metal plane.

- For p polarization, strong discrepancies appear.

Crucial consequence

Even though metals used to make gratings (Al, Ag, Au…) present high reflectivities in the visible and near IR, the model of perfect conductivity fails, at least for p polarized light (and then for natural light).

Unfortunately, this numerical demonstration was not considered as a definitive proof

(ICO IX, Santa Monica, 1972).

M.C. Hutley, 1973
M.C. Hutley and V.M. Bird, 1973

Hutley constructed holographic diffraction gratings, measured their profiles (profilometer = chisel shape stylus) and their efficiencies in different orders for various incidences, wavelengths, metals, profiles. Comparing his results to theoretical data deduced from a theory assuming the metal to be perfectly conducting, he noticed:

-For s polarized light, the experimental efficiencies deduce from the numerical ones through a multiplication factor close to the reflectivity of the metal plane

- For p polarization, strong discrepancies appear.

- He invoked 3 possible reasons to explain these discrepancies.

Conclusion: strong discrepancies observed for p-polarization between:

- The perfect conductivity model for gratings

-The finite conductivity model or the experimental measurements.

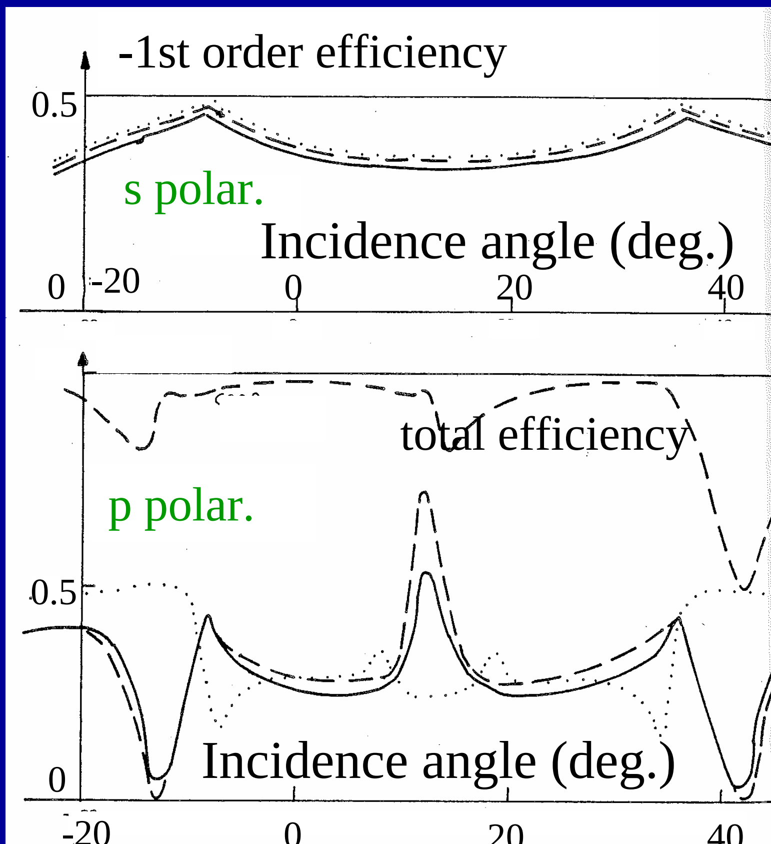
Two possible explanations:

1- The failure of the perfect conductivity model

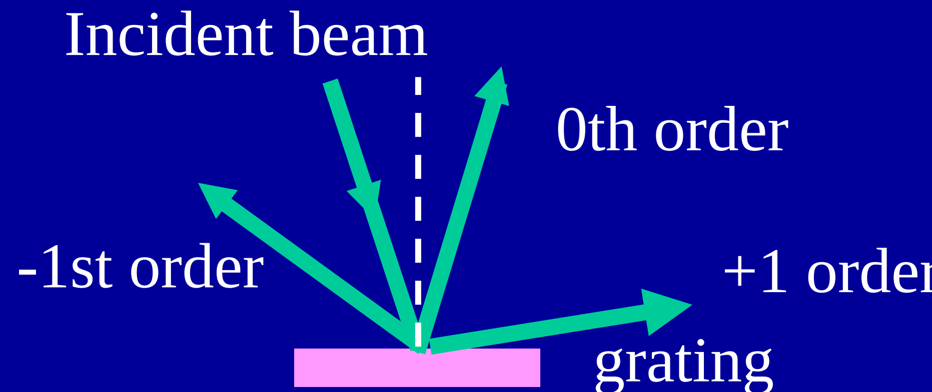
2- The failure of Maxwell equations and macroscopic theory of scattering for modeling the real properties of metallic gratings (it could be necessary to use a microscopic model of Solid State Physics)

Does Electromagnetic scattering model fail in Optics?

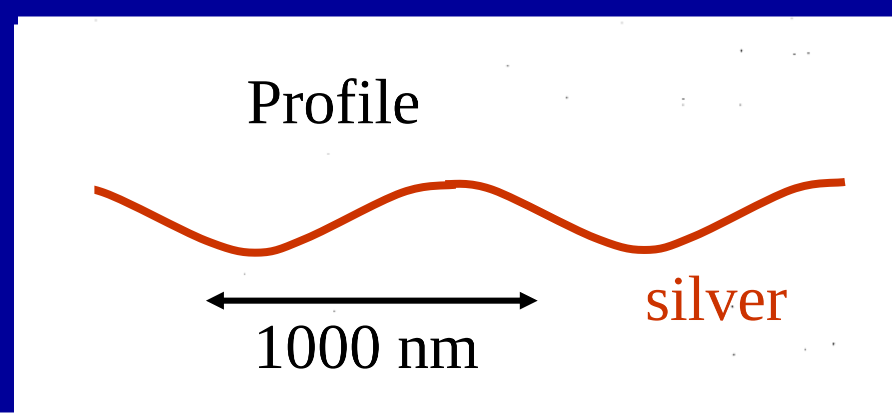
R.C. Mc Phedran and D. Maystre, 1974



experimental data
theory: Ag grating
theory: PC grating



Holographic grating with period 1205 nm illuminated for p polarization by a laser beam with λ=521 nm



Quantitative phenomenological theory (D. Maystre, 1973)

The results given by the integral theory of gratings were confirmed by the differential theory (M. Nevière and P. Vincent, 1974), even though this theory had strong problems of stability for metallic gratings.

Thus, the new opportunity to perform rapid and accurate computations of grating properties encouraged us to develop a quantitative phenomenological analysis of Wood anomalies.

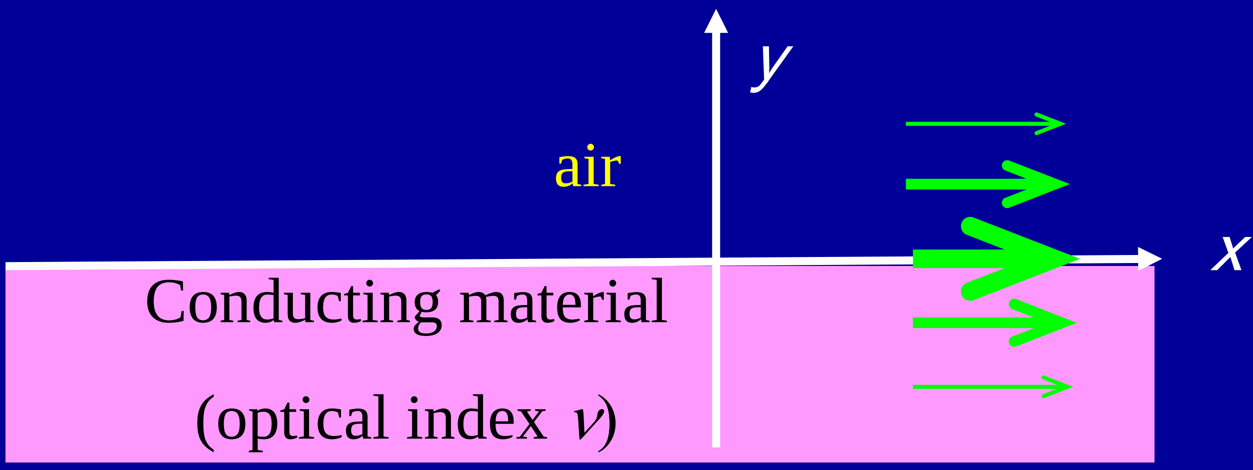
Phenomenology: using intuition then mathematics in order to describe quantitatively a phenomenon from the smallest number of parameters

It allowed the discovery of the phenomenon of total absorption of light (Maystre and Petit, 1976, Hutley and Maystre, 1976)

What is a surface plasmon?:

1- Surface plasmon on a flat interface

Surface plasmon on a flat interface



Complex amplitude with time dependence in $\exp(-i\omega t)$

A polarized surface wave $\mathbf{F} = F\,\mathbf{z}$ must satisfy the following conditions:

- Propagation in *x:* $F(x, y) = f(y)\exp(ik\alpha x)$ $(k=2\pi/\lambda)$

- Maxwell equations at any point and boundary conditions at $y = 0$

- Radiation condition: it must $\begin{cases} \text{vanish or propagate upwards as } y \to +\infty \\ \text{vanish as } y \to -\infty \end{cases}$

Question: can α be real?

$$F(x,y) = f(y)\exp(ik\alpha x) = f(y)\exp(ik\alpha' x)\exp(-k\alpha'' x)$$

$$\alpha = \alpha' + i\alpha''$$

If the surface wave propagates towards $x = +\infty$ on the surface of a

conducting dissipative material $(\alpha' > 0)$, its amplitude should decrease (in modulus) as *x* increases. Thus α must be complex with

$$\alpha'' > 0$$

We will assume that $\alpha'' \ll \alpha'$: the surface wave can

propagate at a distance $\delta \gg \lambda$ before extinction .

Maxwell equations in the air

$$F(x,y) = f(y)\exp(ik\alpha x)$$

Helmholtz equation: $\nabla^2 F + k^2 F = 0$

$$f'' + k^2\beta^2 f = 0 \quad , \quad \beta^2 = 1-\alpha^2$$

$$f(y) = a\exp(-ik\beta y) + b\exp(+ik\beta y) \qquad \beta(\alpha) = \sqrt{1-\alpha^2}$$

$$\boxed{F(x,y) = \cancel{a\exp(ik\alpha x - ik\beta y)} + b\exp(ik\alpha x + ik\beta y)}$$

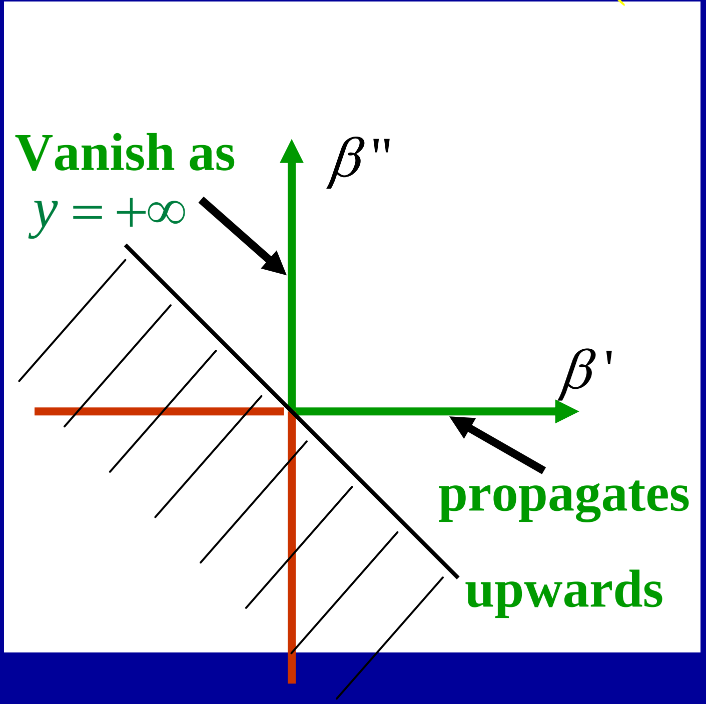
We must define the determination of $\beta(\alpha)$ in order to impose the radiation condition

Determination of $\beta(\alpha) = \sqrt{1-\alpha^2}$

$$\beta = \beta' + i\beta''$$

$$F(x,y) = b\exp(ik\alpha x + ik\beta y) =$$

$$= b\exp(ik\alpha x)\exp(ik\beta' y)\exp(-k\beta'' y)$$



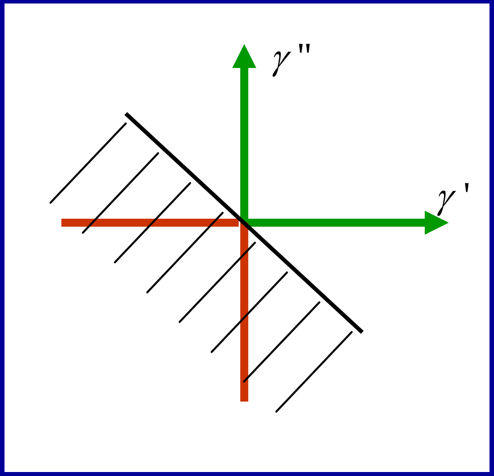
For complex α, the determination of β is given by

$$\beta' + \beta'' > 0$$

Maxwell equations in the conducting material

$$\nabla^2 F + k^2 \nu^2 F = 0$$

Imposing $F(x,y) = a' \exp(ik\alpha x - ik\gamma y) + \cancel{b' \exp(ik\alpha x + ik\gamma y)}$



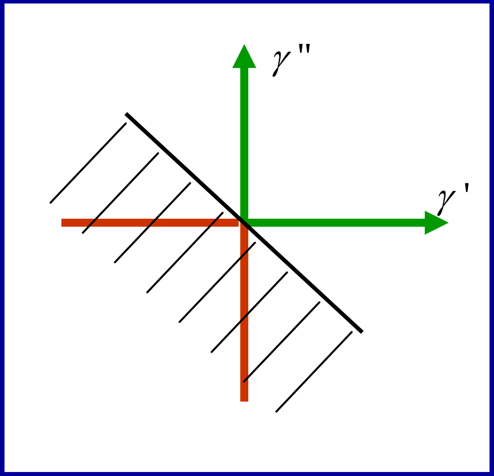
with $\gamma(\alpha) = \sqrt{\nu^2 - \alpha^2}$

The radiation condition is satisfied by keeping the same determination for γ as for β

$$\gamma' + \gamma'' > 0$$

Calculation of α

$$F(x,y)=\begin{cases} b\exp\left(ik\alpha x+ik\beta y\right) & \text{in the air} \\ a'\exp\left(ik\alpha x-ik\gamma y\right) & \text{in the conducting material} \end{cases}$$



Propagate along the *x*-axis and satisfies:

- Maxwell equations in both materials

-Radiation condition

-Last condition: boundary condition on the interface: continuity of the tangential components of the electric and magnetic fields at y = 0

Determination of α, s polarization

$$F(x,y)=\begin{cases} b\exp\left(ik\alpha x+ik\beta y\right) & \text{in the air} \\ a'\exp\left(ik\alpha x-ik\gamma y\right) & \text{in the conducting material} \end{cases}$$

s polarization,
F electric field

F and $\dfrac{\partial F}{\partial y}$ continuous at $y=0$

$$\begin{cases} b=a' \\ \beta b=-\gamma a' \end{cases} \Rightarrow \beta=-\gamma \quad \Rightarrow \quad \sqrt{1-\alpha^2}=-\sqrt{\nu^2-\alpha^2} \Rightarrow 1=\nu^2$$

Impossible!

Determination of α, p polarization

$$F(x,y)=\begin{cases} b\exp(ik\alpha x+ik\beta y) & \text{in the air} \\ a'\exp(ik\alpha x-ik\gamma y) & \text{in the conducting material} \end{cases}$$

p polarization, F magnetic field

F continuous and $\left.\frac{\partial F}{\partial y}\right|_{\text{air}}=\frac{1}{\nu^2}\left.\frac{\partial F}{\partial y}\right|_{\text{cond.}}$ at $y=0$

$$\begin{cases} b=a' \\ \beta b=-\dfrac{\gamma a'}{\nu^2} \end{cases} \Rightarrow \quad \nu^2\sqrt{1\text{-}\alpha^2}=\text{-}\sqrt{\nu^2\text{-}\alpha^2} \Rightarrow \alpha=\tilde{\alpha}^{plane}=\frac{\nu}{\sqrt{1+\nu^2}}$$

Surface wave: surface plasmon

Calculation of α for a metal in the visible region

$$\alpha = \tilde{\alpha}^{plane} = \frac{\nu}{\sqrt{1+\nu^2}}$$

Aluminum at 647 nm: $\nu = 1.3+i7.1$

$$\tilde{\alpha}^{plane} = 1.009 + i\,3.5\;\;10^{-3}$$

Real part slightly greater than unity

Very small imaginary part

$\Rightarrow$

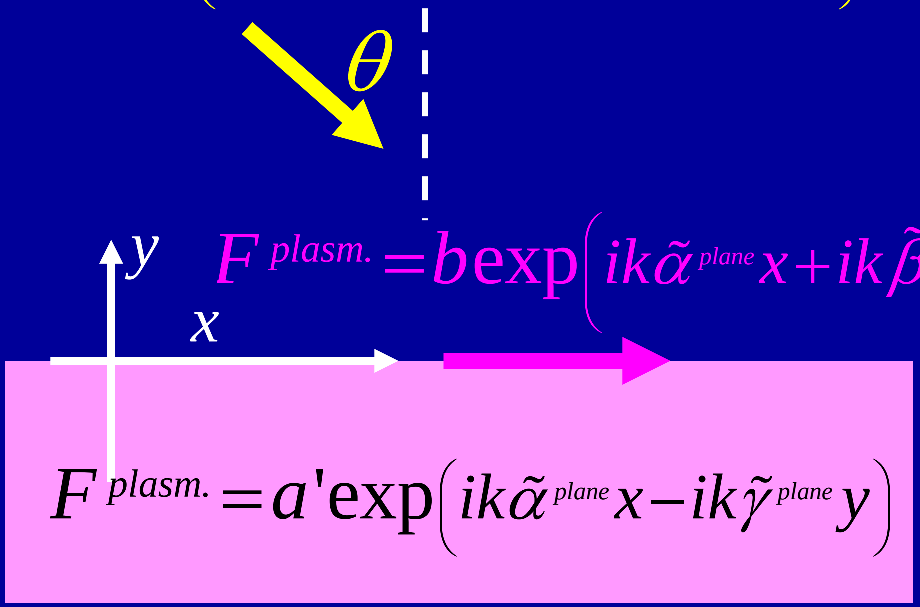
1- The surface plasmon can propagate at large distance before extinction

2- Since the real part of the propagation constant $k\tilde{\alpha}^{plane}$ is greater than k, the surface plasmon cannot be excited by a plane wave

Why a surface plasmon cannot be excited by a plane wave coming from the air?

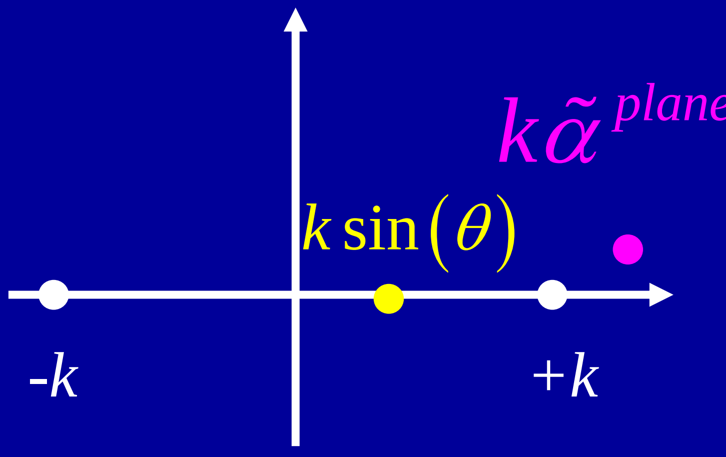
Incident plane wave

$$F^{i} = \exp\left(ik\sin\left(\theta\right)x - ik\cos\left(\theta\right)y\right)$$



$$F^{plasm.} = b\exp\left(ik\tilde{\alpha}^{plane}x + ik\tilde{\beta}^{plane}y\right)$$

$$F^{plasm.} = a'\exp\left(ik\tilde{\alpha}^{plane}x - ik\tilde{\gamma}^{plane}y\right) \Longrightarrow$$



The x-component of the propagation constant is conserved, thus the plasmon cannot be excited by a plane wave in the air

How to excite the surface plasmon?

1- Use of an electron beam
(C.J. Powell and J.B. Swan, 1959)

Striking thin films of metal with an electron beam, it has been observed peaks of absorption in the energy spectrum of the transmitted beam.

These losses are due to the collective excitation of conduction electrons at the surface of the metal. This is the description of surface plasmons in Solid State Physics.

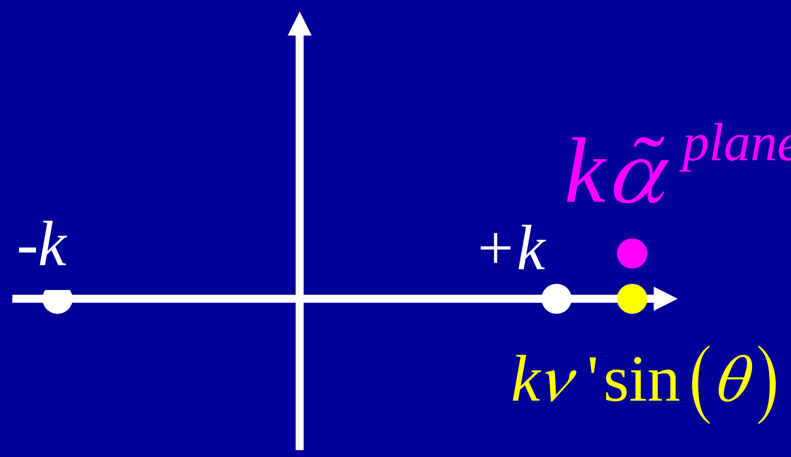
In the following of this talk, this microscopic interpretation of the existence of surface plasmons is ignored.

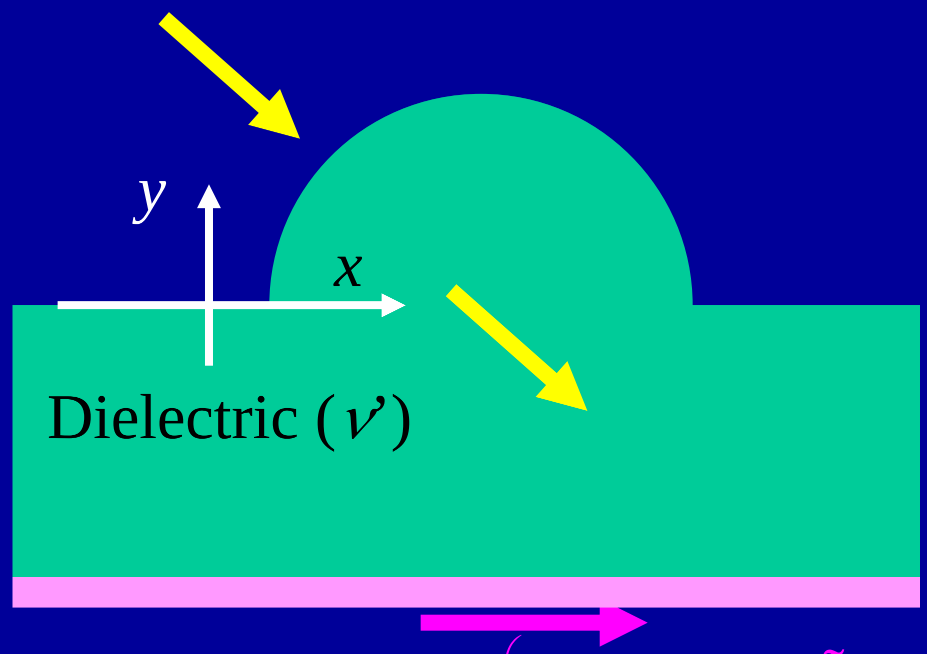
Question: How to describe a phenomenon caused by a collective resonance of electrons without electrons?

2- Use of a prism

Incident beam inside the dielectric

$$F^{i} = \exp\left(ik\nu'\sin\left(\theta\right)x - ik\nu'\cos\left(\theta\right)y \right)$$

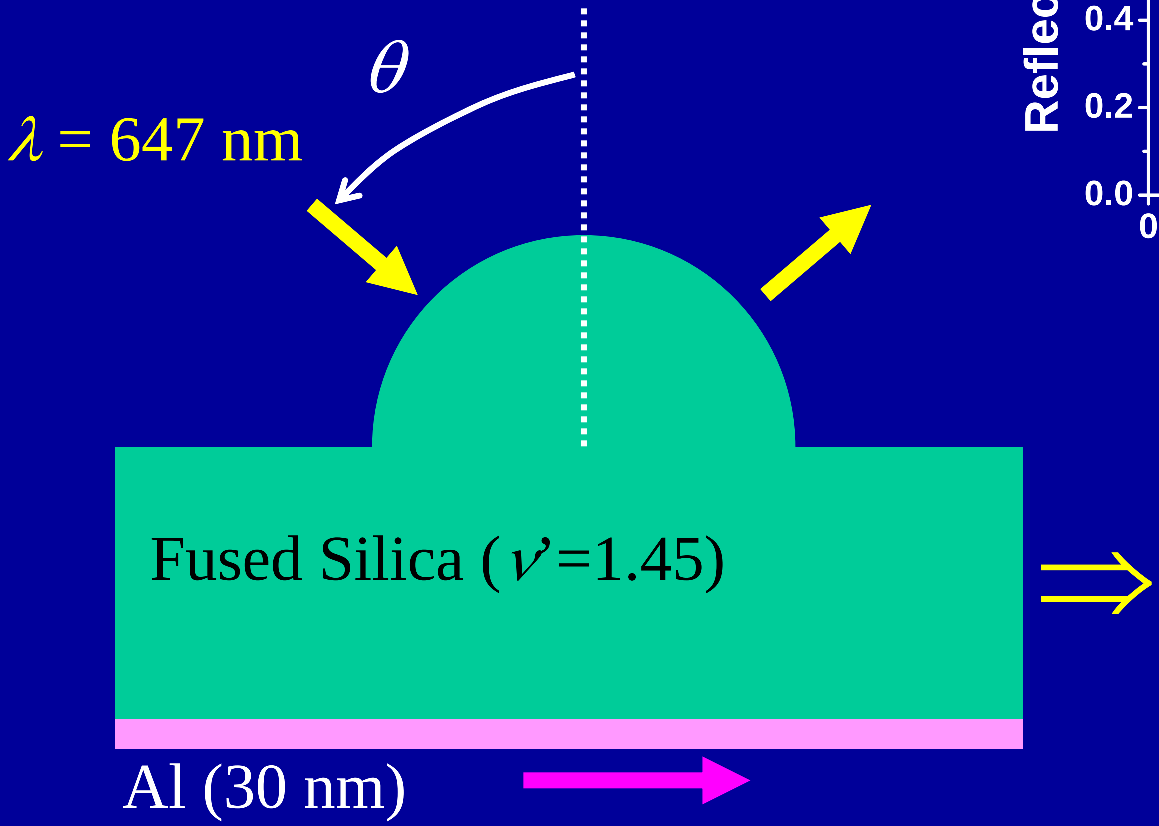


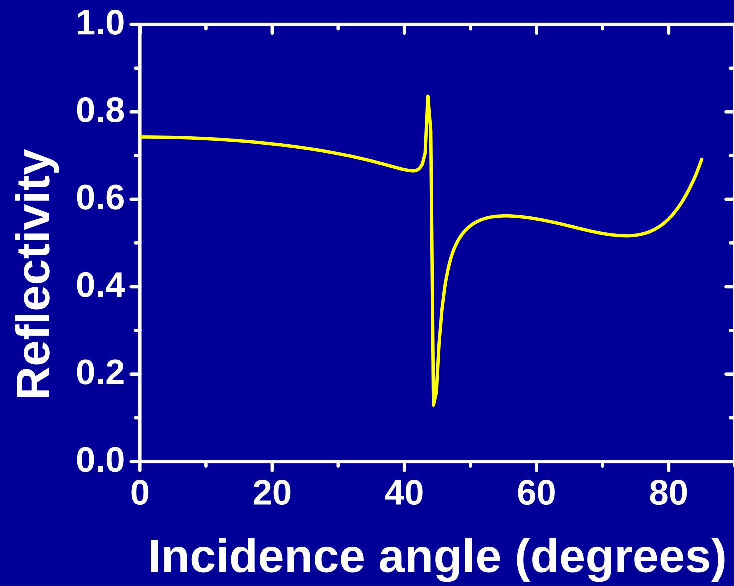


$$F^{plasm.} = a'\exp\left(ik\tilde{\alpha}^{plane}x - ik\tilde{\beta}^{plane}y \right)$$

$\Rightarrow$ The prism multiplies the *x*-component of the propagation constant of the incident plane wave by a factor ν'

2- Use of a prism: numerical calculation





Minimum at θ = 44.55

$\nu' \sin(\theta) = 1.017$

$\mathrm{Re}\left(\tilde{\alpha}^{plane}\right) = 1.009$

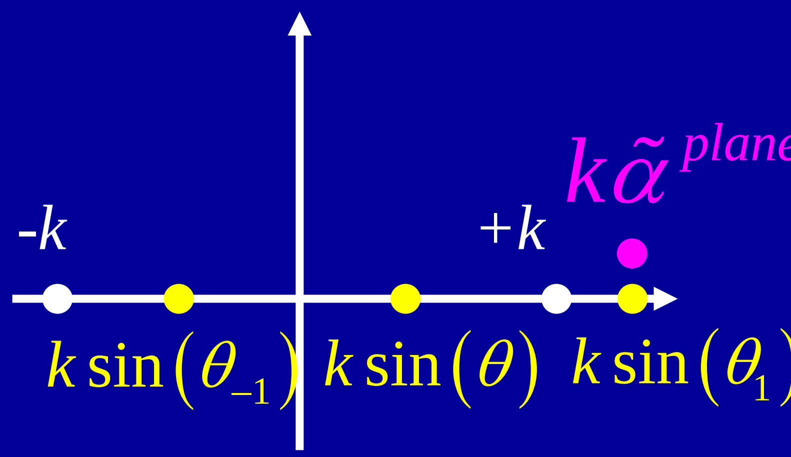
Resonance:

$\nu' \sin\left(\theta\right) = \tilde{\alpha}^{plane}$

3- Use of a grating, heuristic presentation

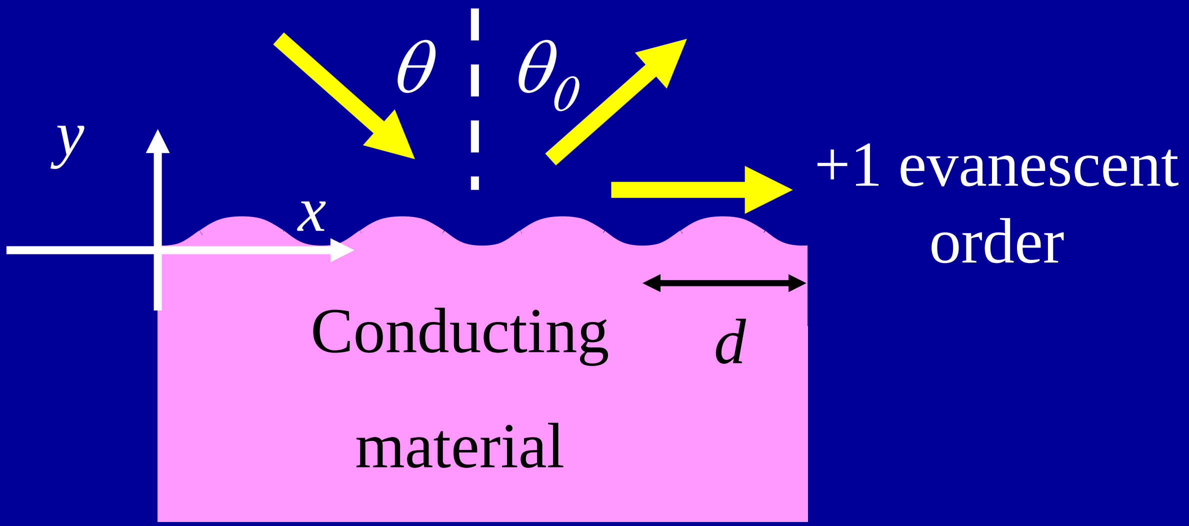
Incident field:

$$F^i = \exp\left(ik\sin\left(\theta\right)x - ik\cos\left(\theta\right)y\right)$$



Scattered field:

$$\sin\left(\theta_n\right) = \sin\left(\theta\right) + n\lambda / d$$



What is a surface plasmon?:

2- Surface plasmon on a grating

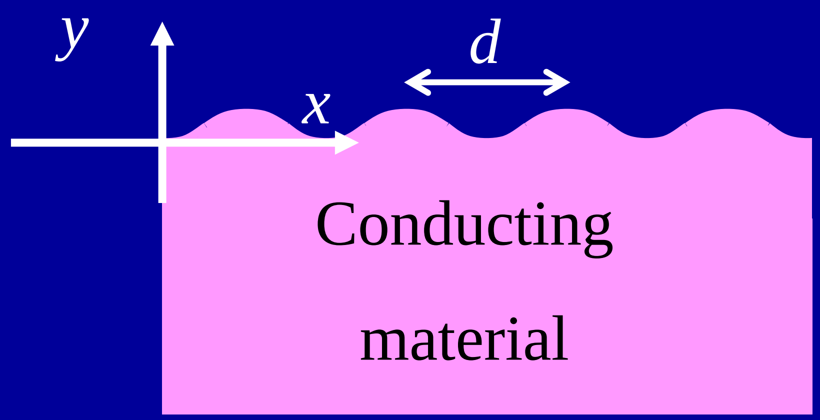
Grating: description of the surface plasmon

Floquet-Bloch theorem: a surface wave propagating along the grating surface takes the form:

$$F(x,y) = P(x,y)\exp(ik\tilde{\alpha}x),\ P(x,y) \text{ of period } d \text{ in } x$$

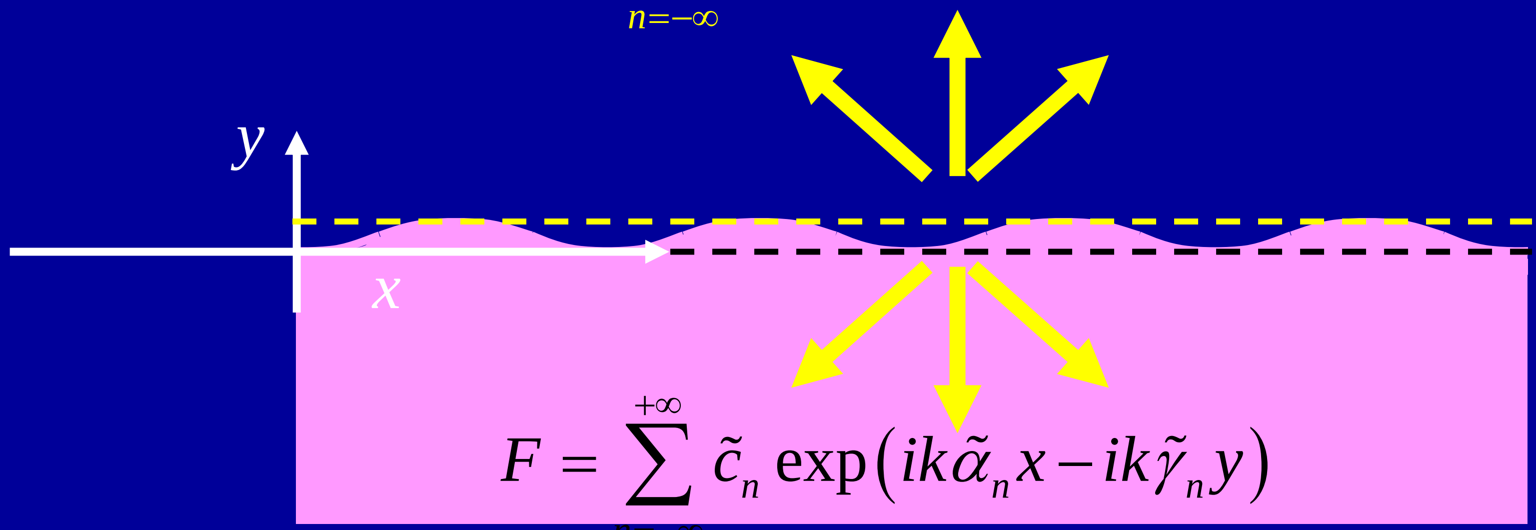
$$F(x,y) = \sum_{n=-\infty}^{+\infty} p_n(y)\exp(ik\tilde{\alpha}_n x),\ \tilde{\alpha}_n = \tilde{\alpha} + n\lambda / d$$

Flat surface: $p_n(y) = 0$ except for $n = 0$



Use of Maxwell equations and radiation conditions

$$F = \sum_{n=-\infty}^{+\infty} \tilde{b}_n \exp\left(ik\tilde{\alpha}_n x + ik\tilde{\beta}_n y\right)$$

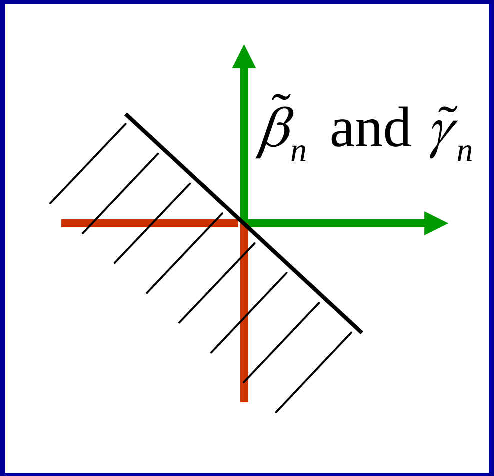


$$F = \sum_{n=-\infty}^{+\infty} \tilde{c}_n \exp\left(ik\tilde{\alpha}_n x - ik\tilde{\gamma}_n y\right)$$

$\tilde{\alpha}_n = \tilde{\alpha} + n\lambda / d,\ \tilde{\alpha}$ defined to within λ / d

Continuity: $\tilde{\alpha} \ \to \tilde{\alpha}^{plane} = \dfrac{\nu}{\sqrt{1+\nu^2}}$ when $h \to 0$

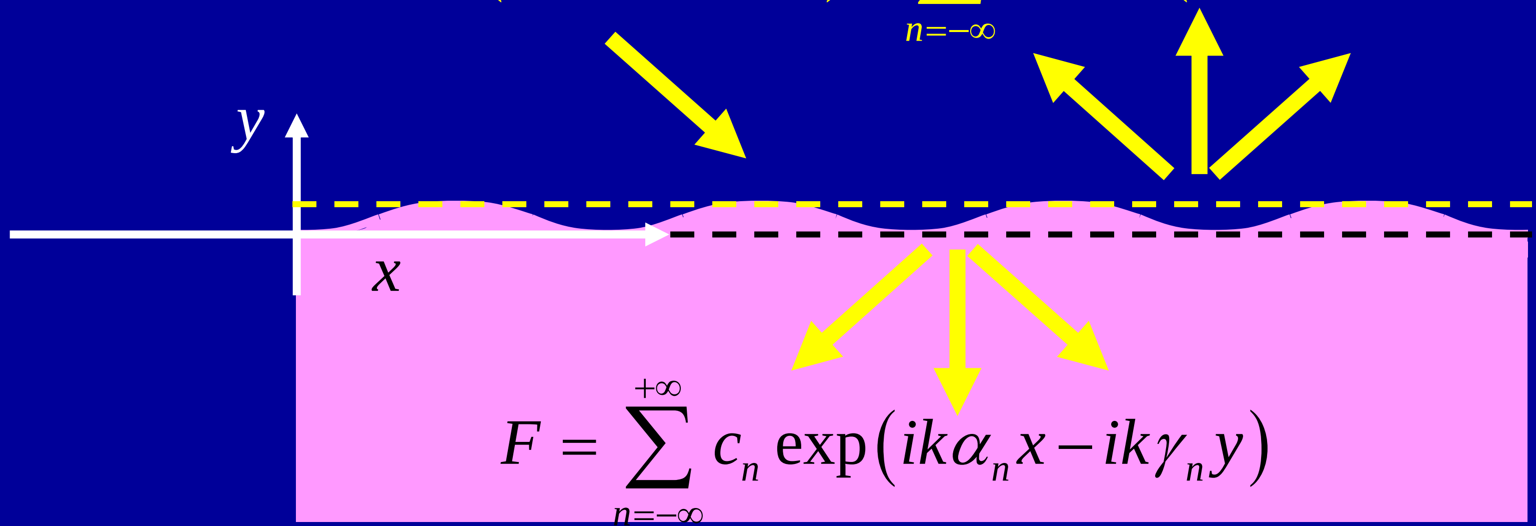
$$\tilde{\beta}_n(\alpha) = \sqrt{1-\tilde{\alpha}_n^2}, \quad \tilde{\gamma}_n(\alpha) = \sqrt{\nu^2 - \tilde{\alpha}_n^2}$$



Phenomenology: why surface plasmons generate Wood anomalies?

The scattering problem

$$F = a\exp\left(ik\alpha x - ik\beta y\right) + \sum_{n=-\infty}^{+\infty} b_n \exp\left(ik\alpha_n x + ik\beta_n y\right)$$



$$F = \sum_{n=-\infty}^{+\infty} c_n \exp\left(ik\alpha_n x - ik\gamma_n y\right)$$

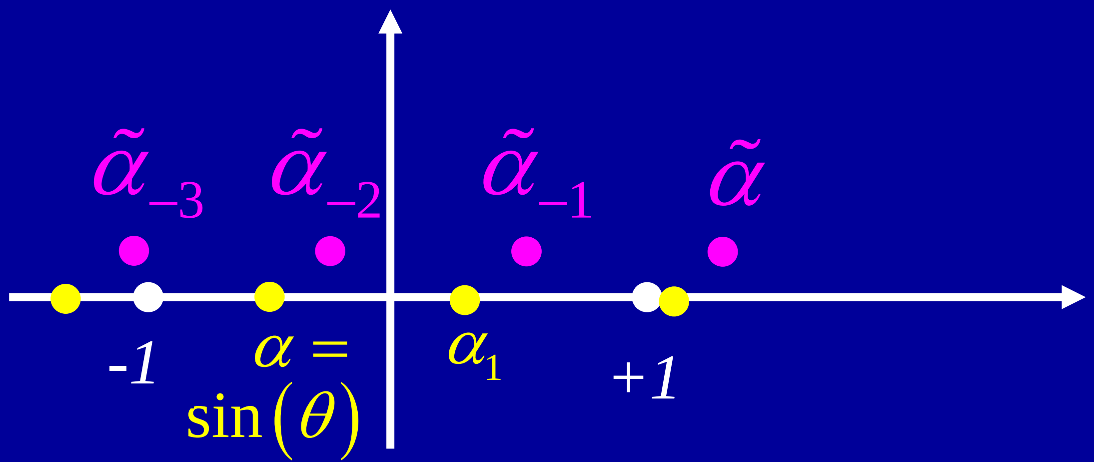
$$\alpha_n = \alpha + n\lambda / d, \quad \alpha = \sin\left(\theta\right), \quad \beta = \cos\left(\theta\right)$$

$$\beta_n\left(\alpha\right) = \sqrt{1-\alpha_n^2}, \quad \gamma_n\left(\alpha\right) = \sqrt{\nu^2 - \alpha_n^2}$$

Normalized amplitudes $\quad b_n^{norm} = b_n / a, \quad c_n^{norm} = c_n / a$

Field of surface plasmon = field of a scattering problem, but with $a = 0$ and α complex

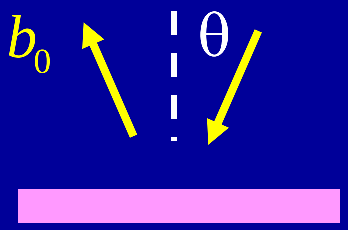
Vital consequence



The normalized amplitudes $b_p^{norm}(\alpha)$ and $c_p^{norm}(\alpha)$ being considered as functions of $\alpha = \sin(\theta)$ real , the constants of propagation $\tilde{\alpha}_n$ of all the space components of the surface plasmon are poles of the analytic continuation of any normalized amplitude in the complex plane of α :

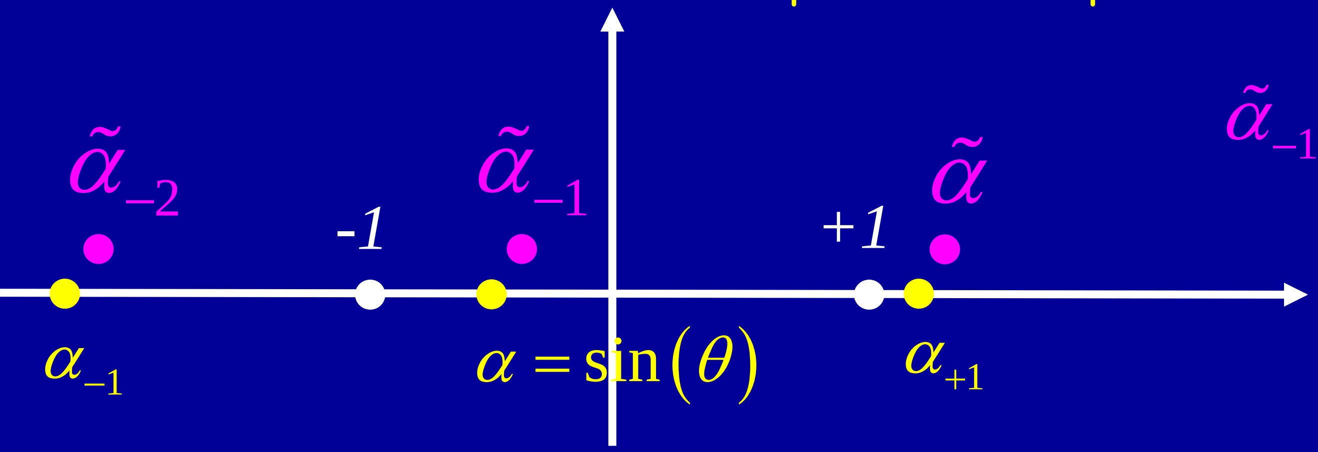
$$b_p^{norm}(\tilde{\alpha}_n) = \tilde{b}_n / 0 = \infty, \quad c_p^{norm}(\tilde{\alpha}_n) = \tilde{c}_p / 0 = \infty$$

When $\alpha = \sin(\theta)$ becomes close to one of them, a resonance phenomenon occurs



Particular case

$$\left|\mathrm{Re}\left(\tilde{\alpha}_n\right)\right| > 1 \text{ except } n = -1$$



$\tilde{\alpha}_{-1}$ is the pole of all the $b_p^{norm}(\alpha)$ and in particular of $b_0^{norm}(\alpha)$

$$b_0^{norm}(\alpha) = \frac{g_0}{\alpha - \tilde{\alpha}_{-1}} + g_1 + \cancel{u(\alpha)(\alpha - \tilde{\alpha}_{-1})}$$

$$b_0^{norm}(\alpha) \simeq \frac{g_0 + g_1(\alpha - \tilde{\alpha}_{-1})}{\alpha - \tilde{\alpha}_{-1}} = g_1 \frac{\alpha - \tilde{\alpha}'_{-1}}{\alpha - \tilde{\alpha}_{-1}} \text{ with } \tilde{\alpha}'_{-1} = \tilde{\alpha}_{-1} - \frac{g_0}{g_1}$$

Phenomenological theory: crucial result

$$b_0^{norm}(\alpha) \simeq r\,\frac{\alpha-\alpha^Z}{\alpha-\alpha^P}\ \text{, with:}$$

$$\alpha^P = \tilde{\alpha}_{-1} = \tilde{\alpha} - \frac{\lambda}{d},$$

$$\alpha^Z = \tilde{\alpha}_{-1} - \frac{g_0}{g_1} = \alpha^P - \frac{g_0}{g_1}$$

If $h \to 0$, $\tilde{\alpha} \to \tilde{\alpha}^{plane}$ thus $\alpha^p \to \alpha^z \to \tilde{\alpha}^{plane} - \frac{\lambda}{d} = \frac{\nu}{\sqrt{1+\nu^2}} - \frac{\lambda}{d}$

If h=0, the pole and the zero cancel out each other

thus r is close to the reflection coefficient of the metallic plane

Numerical search for poles and zeros

The numerical tools based on rigorous methods of scattering from gratings have been extended to complex values of $\alpha = \sin\left(\theta\right)$

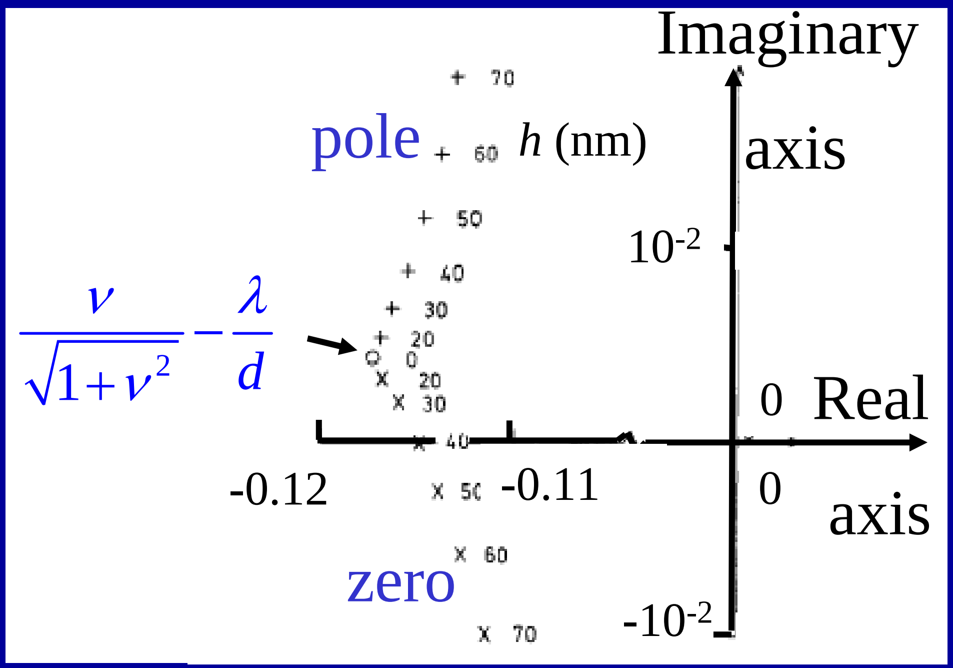
in order to compute numerically the location of poles and zeros in the complex plane

M.C. Hutley and D. Maystre, 1976:

Sinusoidal gold grating with a period *d*=555.5 nm,

Illuminated with p polarized light of wavelength 647 nm. For small incidences, the only non-evanescent order is the zeroth order.

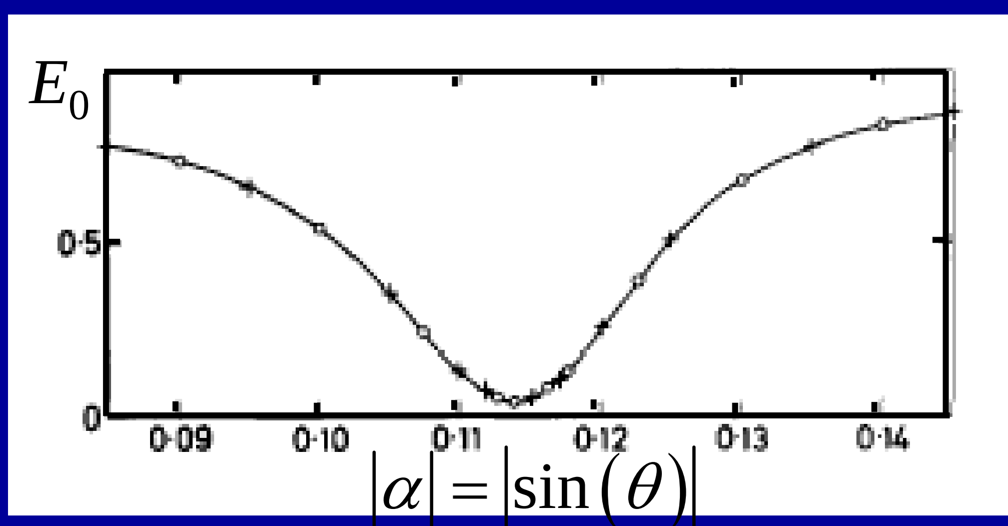
Trajectory of the pole α^p and of the zero α^z of $b_0^{norm}(\alpha)$ when the height h of the sinusoidal grating is increased



Comparison between the calculated efficiency in the zeroth order (crosses) and the phenomenological formula (circles) for h = 50 nm

$$E_0 = \left|b_0^{norm}\right|^2 \simeq |r|^2 \left|\frac{\alpha - \alpha^Z}{\alpha - \alpha^P}\right|^2 \text{ , with } |r|^2$$

reflectivity of a gold plane in normal incidence



Quantitative phenomenology:

Total absorption of light by a grating

Total absorption of light

$$E_0 = \left| b_0^{norm} \right|^2 \simeq \left| r \right|^2 \left| \frac{\alpha - \alpha^Z}{\alpha - \alpha^P} \right|^2$$

If α^Z crosses the real axis of α for h=h_0 then an incident plane wave with angle of incidence $\theta = \sin^{-1}\left(\alpha^z\right)$ will be absorbed in totality

Theoretical prediction for a sinusoidal gold grating at 647 nm: h_0=40 nm for $\theta = 6.6°$

(Maystre and Petit, 1976)

Verification on a sinusoidal grating (Hutley and Maystre, 1976)

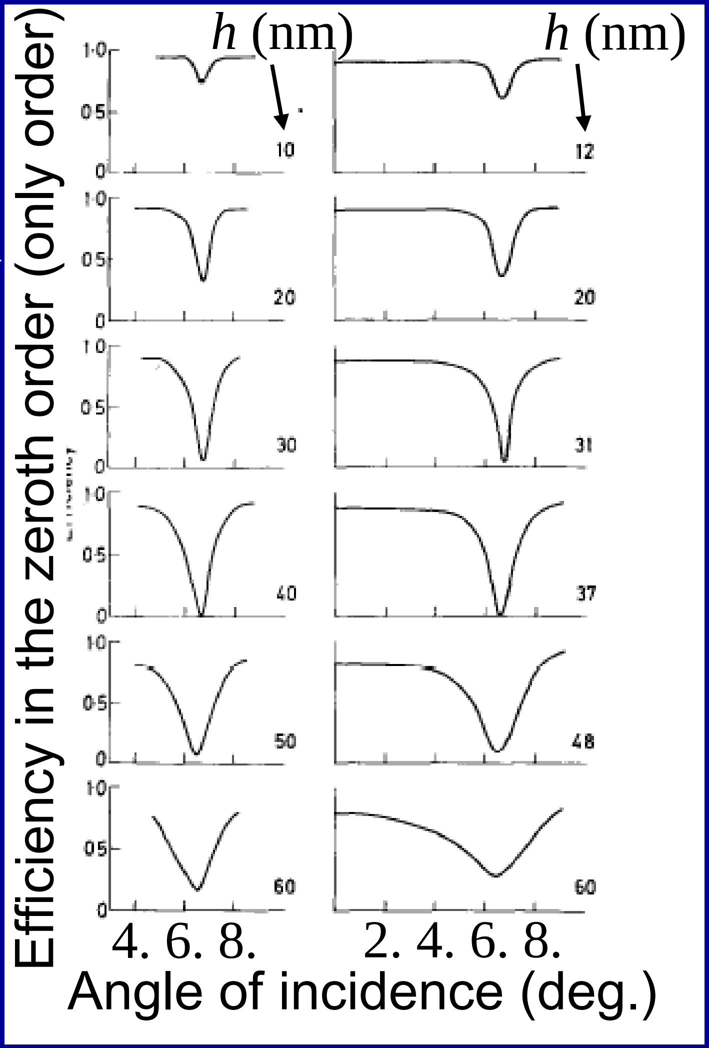
Numerical results:

d=555.5nm

h_0=40 nm

for

$\theta = 6.6°$



Experimental data: d=555.5 nm, for h_0=37 nm and $\theta = 6.6°$ the measured efficiency in the zeroth (specular) order was 0.5%

A very gentle undulation in the surface of a gold mirror causes the reflectance to fall dramatically from over 90% to 0 (theory) or below 1% (exp.)

Application: virus detection

A strong consequence of the phenomenological formula

$$E_0 = \left| b_0^{norm} \right|^2 \simeq \left| r \right|^2 \left| \frac{\alpha - \alpha^Z}{\alpha - \alpha^P} \right|^2$$

Width at half-height of the absorption peak: 2 $\mathrm{Im}(\alpha^P)$

Extinction length proportional to $1/\,\mathrm{Im}\left(\alpha^P\right)$

Since the imaginary part of a^P increases with the height of the grating, the surface plasmon becomes more and more localized and the width of the absorption peak is increased

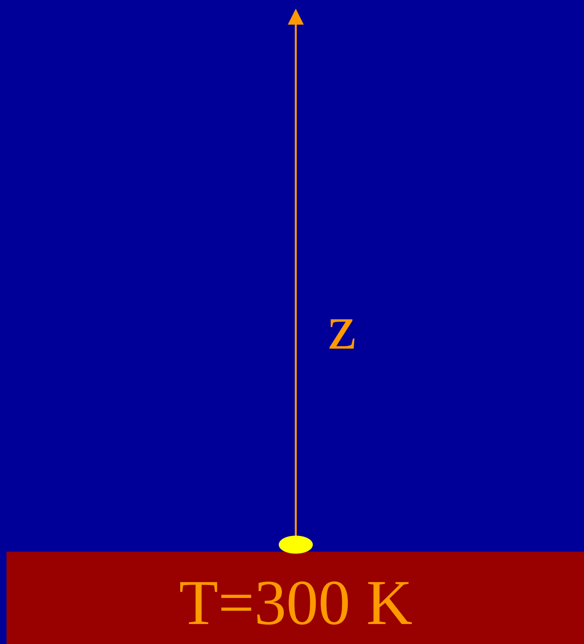
(example of localized plasmons: see T.V. Teperik et al., Nature Photonics, 2008)

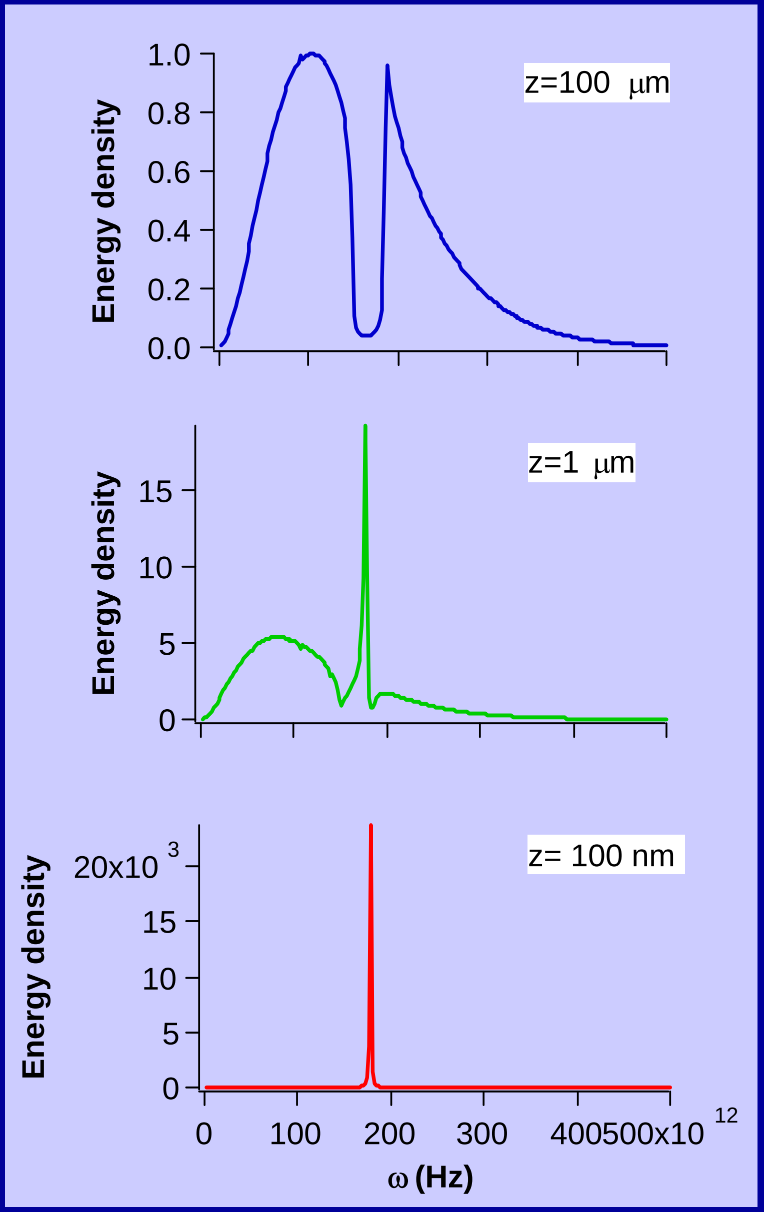
Coherent thermal radiation

See J-J. GREFFET and C. HENKEL
Contemporary Physics, Vol. 48, No. 4,
July – August 2007, 183 – 194

The figures on this subject were
kindly provided by J.J. Greffet

Density of energy near a SiC-vacuum interface





Origine of the phenomenon, coherence of the near field

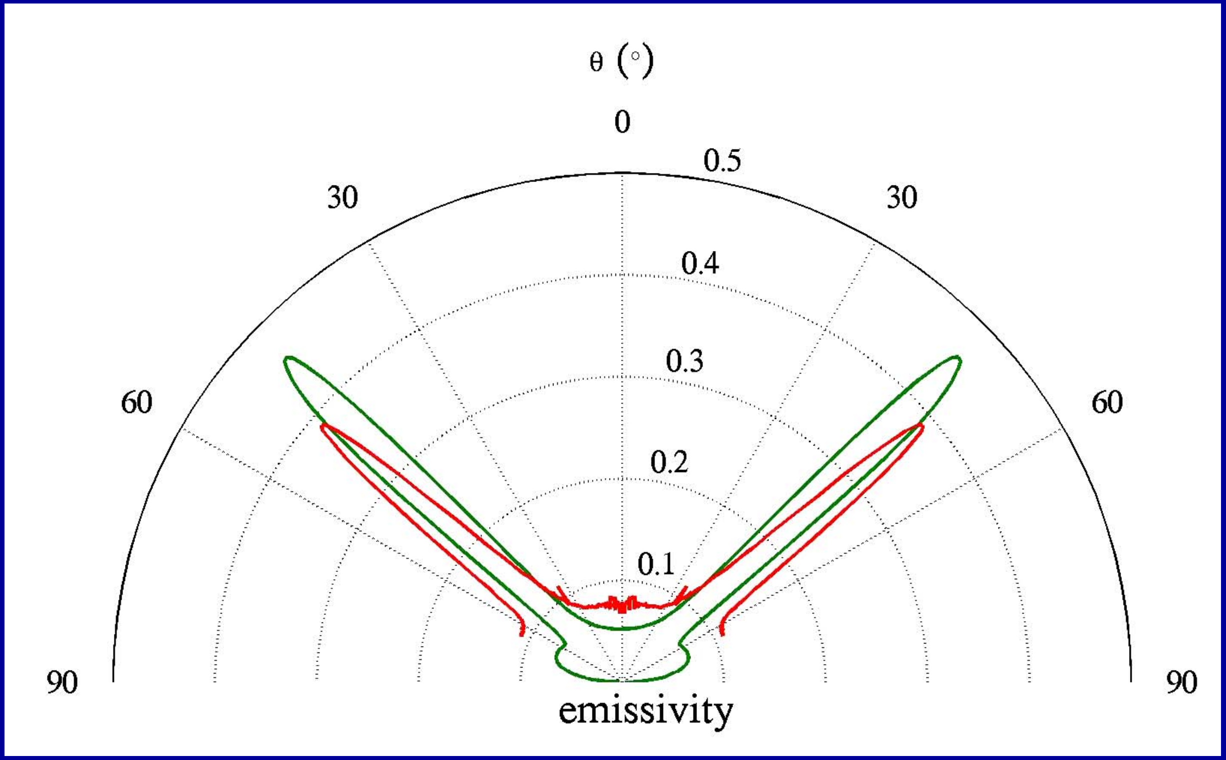
At a given temperature, it can be shown that surface plasmons can spontaneously propagate at the surface of a conducting material around a given frequency.

As a consequence, in contrast with the far field, the near field is nearly monochromatic. It is spatially and temporally strongly coherent.

Replacing the flat surface by a grating, the surface plasmons are scattered at infinity around given directions. In these directions, the far field is strongly coherent

Emission pattern of a SiC grating

Green line : theory
Red line : measurement



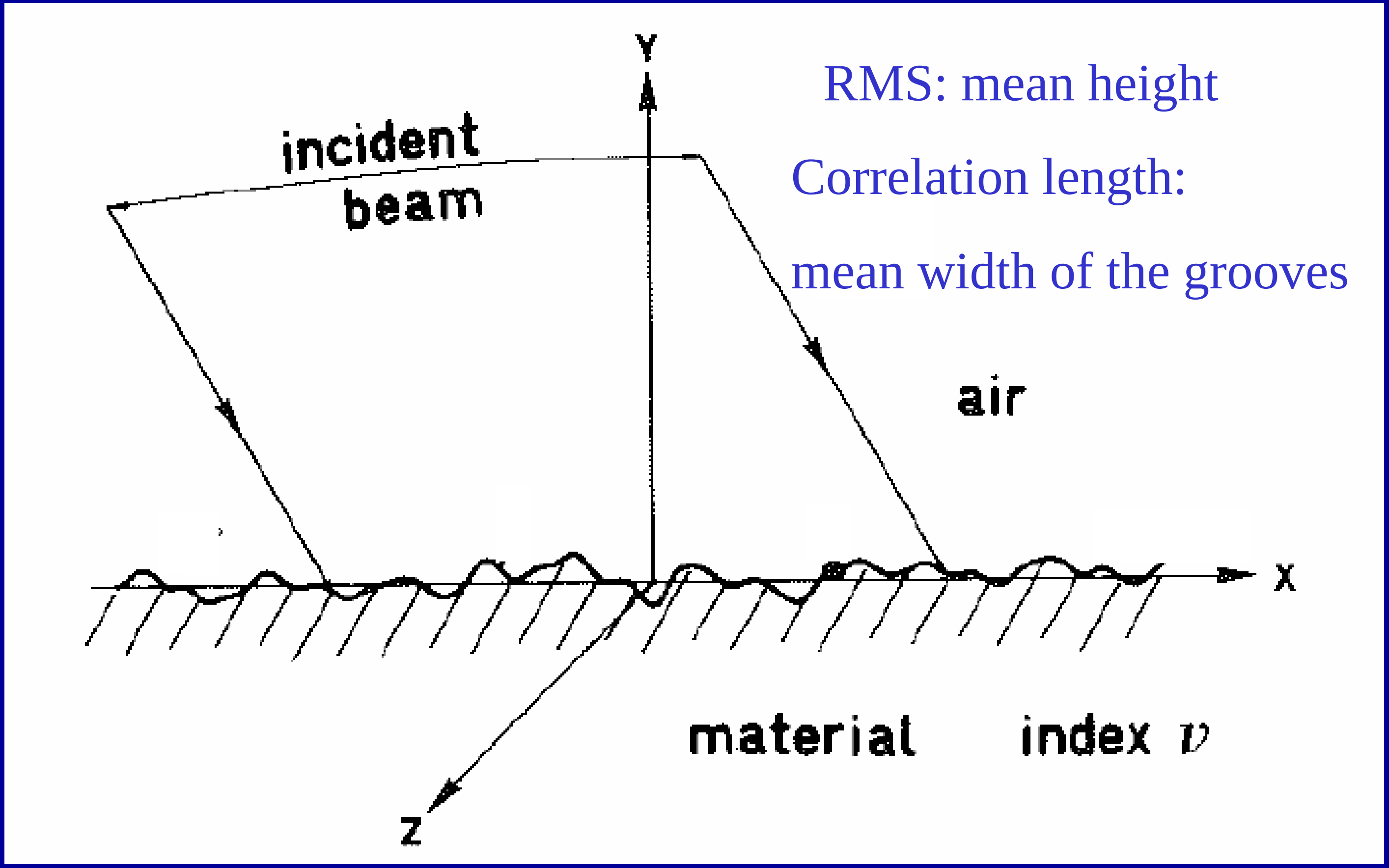
J.J. Greffet et al.
*Nature **416**, p 61 (2002)*

The emission pattern looks like an antenna emission pattern.
The angular width is a signature of the spatial coherence.

Plasmons on randomly rough surfaces

1-Enhanced backscattering (weak localization)

Device



Y
incident
beam
RMS: mean height
Correlation length:
mean width of the grooves
air
X
material index ν
Z

Numerical results for the mean intensity on 100 realizations (D. Maystre et al., 1995)

s polar. p polar.

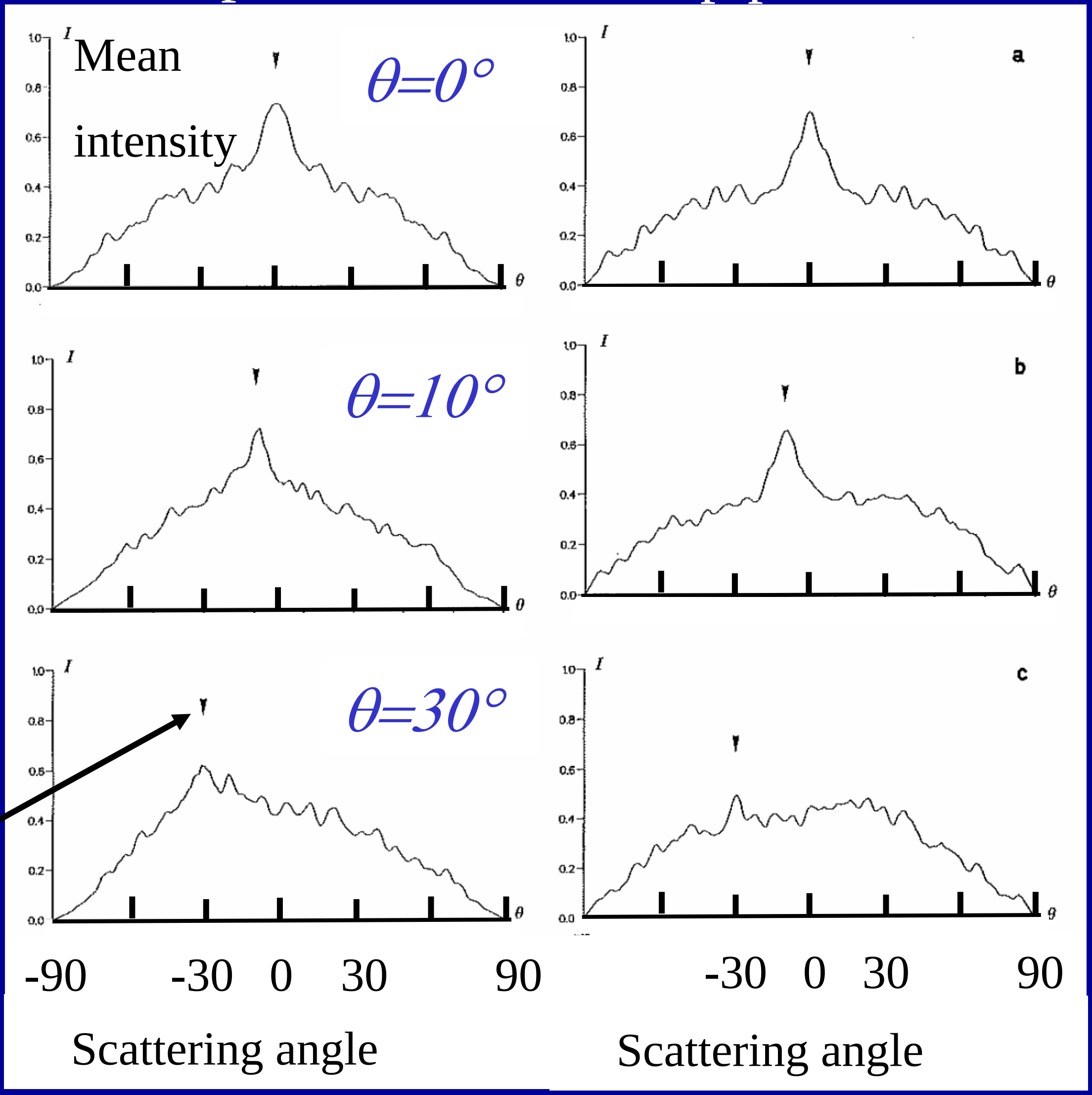
λ=3392nm

Material: Gold

RMS=1950nm

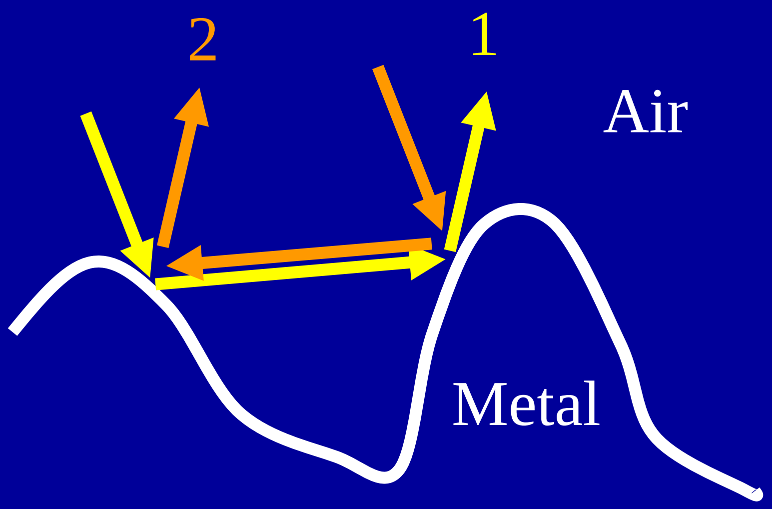
Correlation length = 3570nm

Backscattering angle



Heuristic interpretation

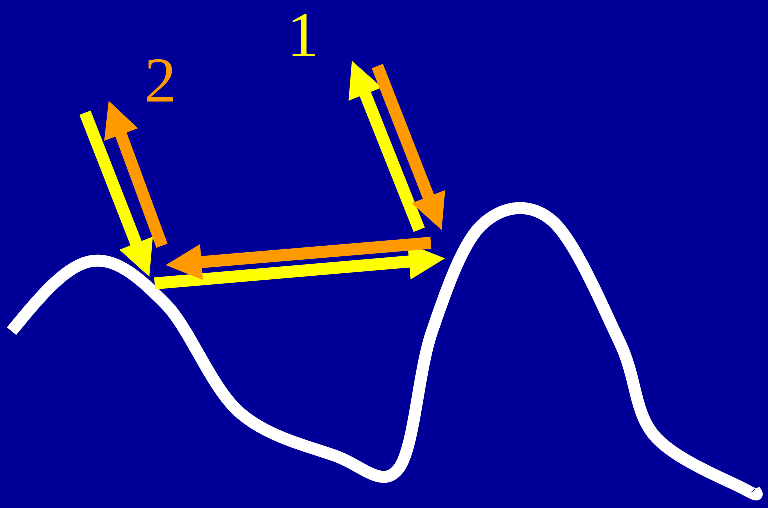
The emerging secondary fields are not in phase in an arbitrary direction



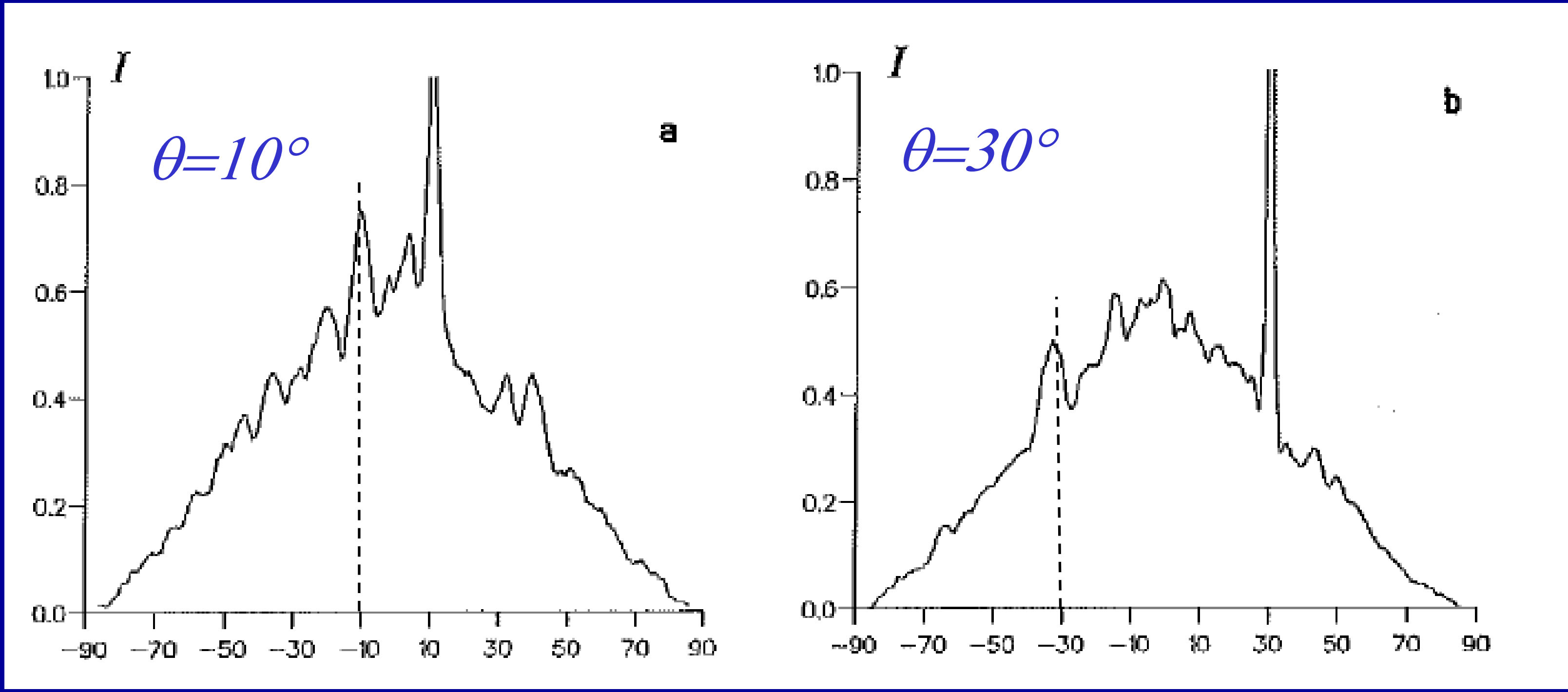
The emerging secondary fields are in phase in the backscattering direction

(reciprocity for reverse paths)

Enhancement by less than 2



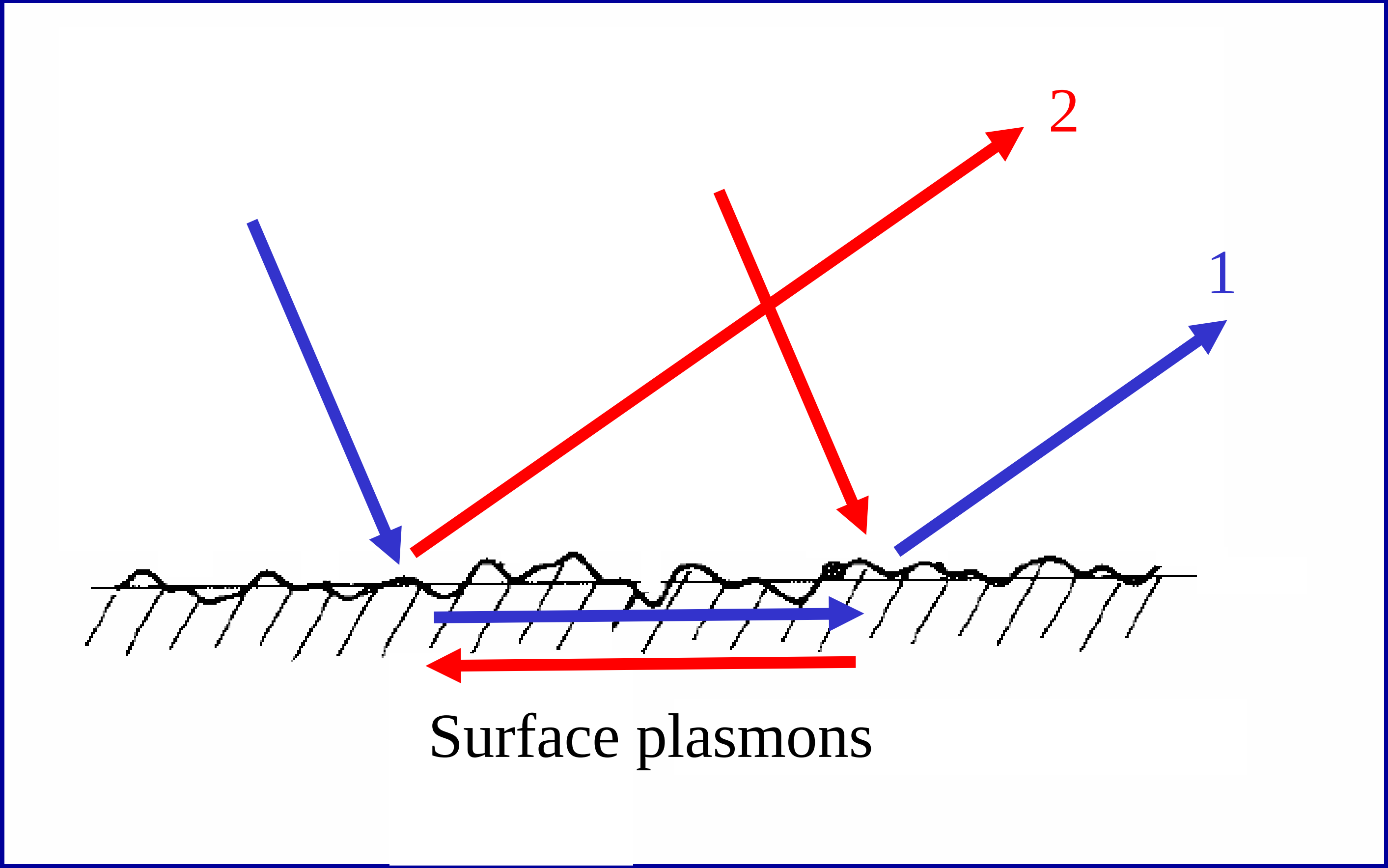
Case of a very flat surface with moderate correlation length
p polarization: mean intensity on 100 realizations



Ag , λ=400nm, RMS=8nm, Correlation length=100nm,

Heuristic interpretation

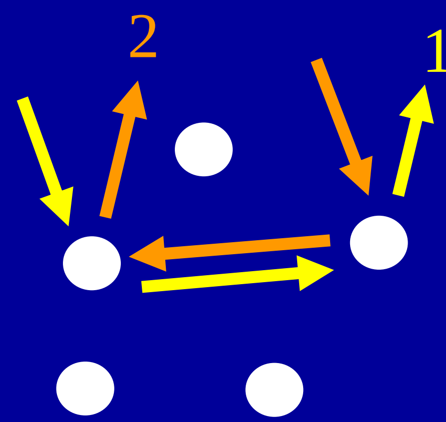
Emerging rays 1 and 2 are in phase in the backscattering direction



Ag, λ=400nm, RMS=8nm, Correlation length=100nm,

Remark:

Enhanced backscattering by a set of particles



Set of particles

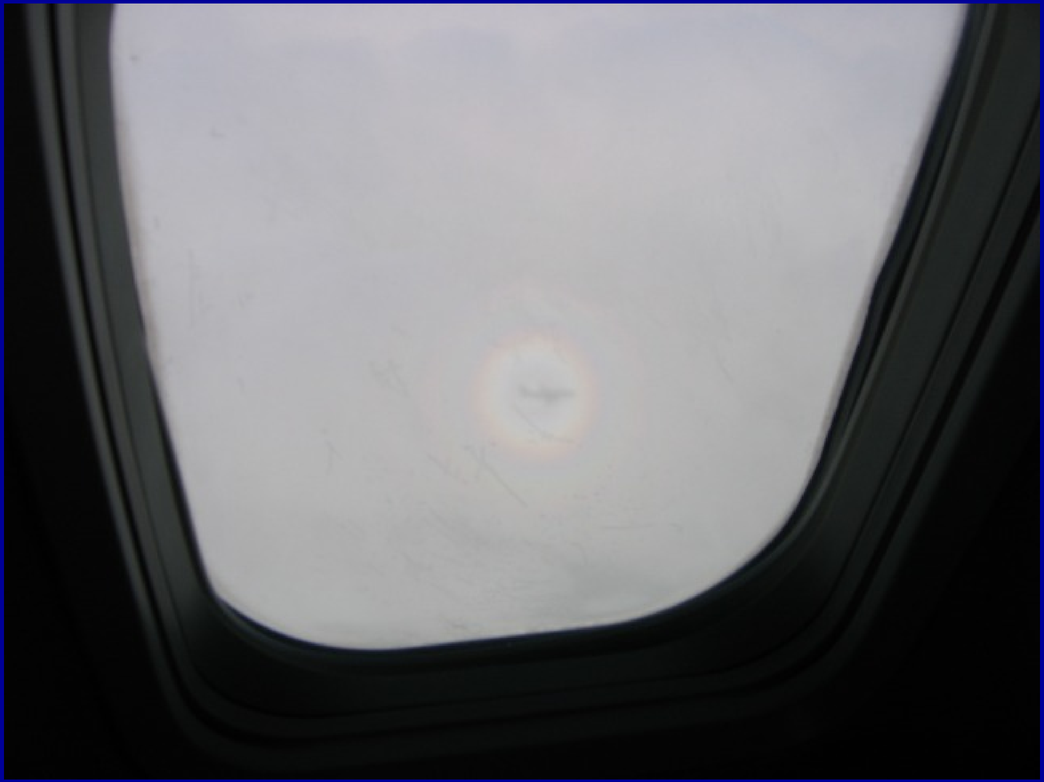
This phenomenon has a vital importance in radar observation or propagation of laser beams in turbulent media

Observation in every day life:

- bright halo (glory) around the shadow of an airplane onto a cloud layer

LEE Boon-Ying

Hong-Kong University



G. Tayeb

Institut Fresnel

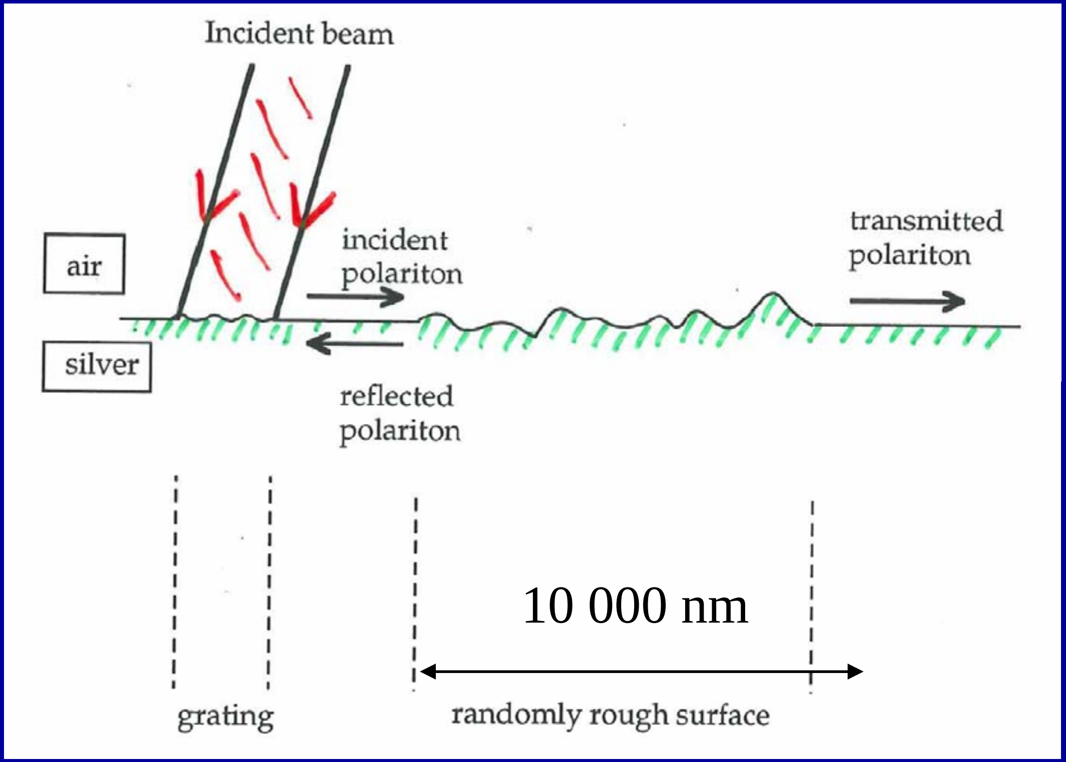
Plasmons on randomly rough surfaces

2-Anderson localization
(strong localization)
P.W. Anderson, 1958

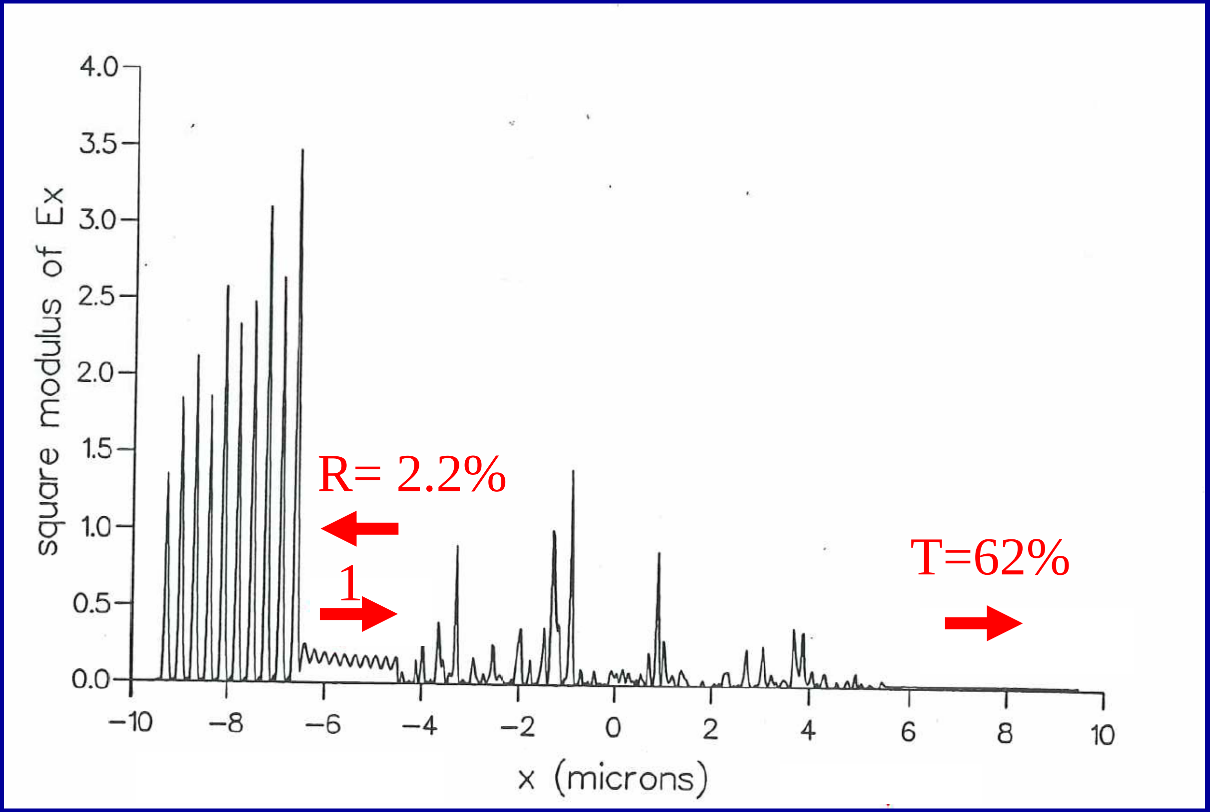
Numerical experiment (D. Maystre et al.,1995):

reflection and
transmission of a surface plasmon by a randomly rough surface

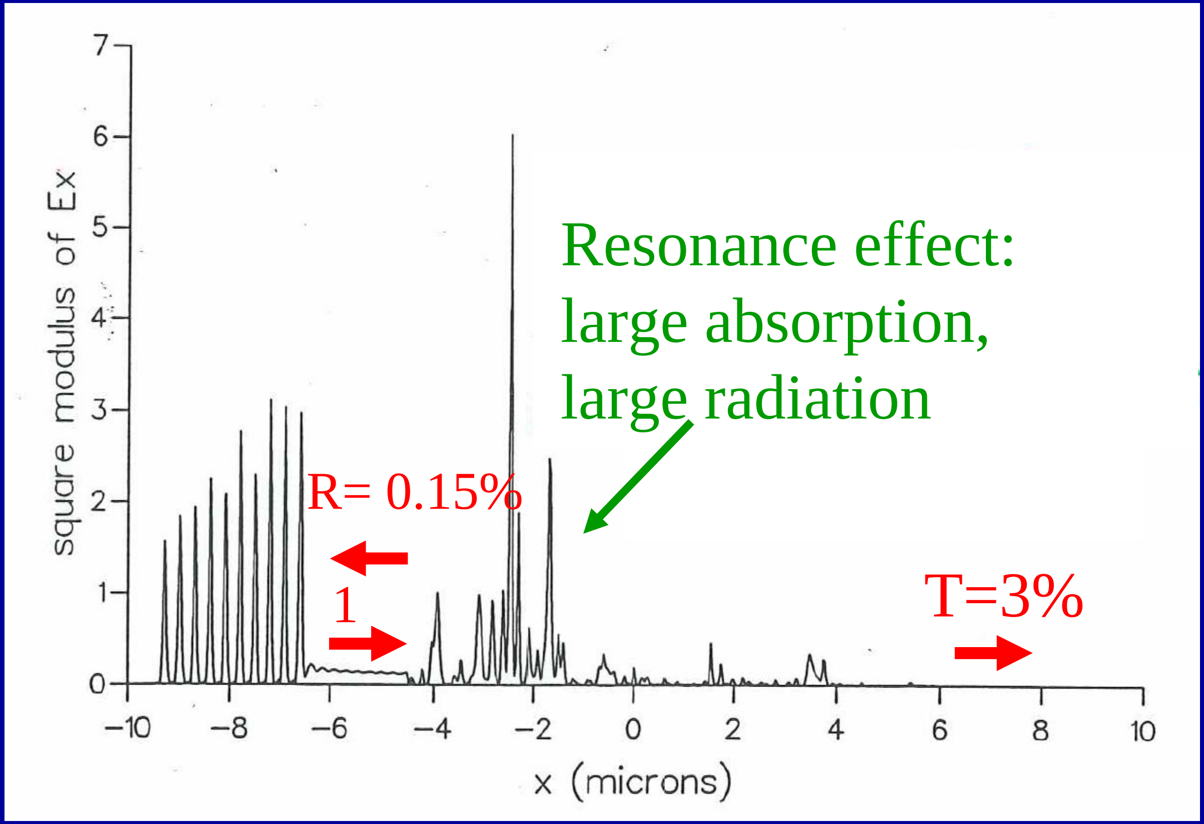
100 realizations, λ = 436 nm, correlation length= 65 nm, RMS= 14 nm



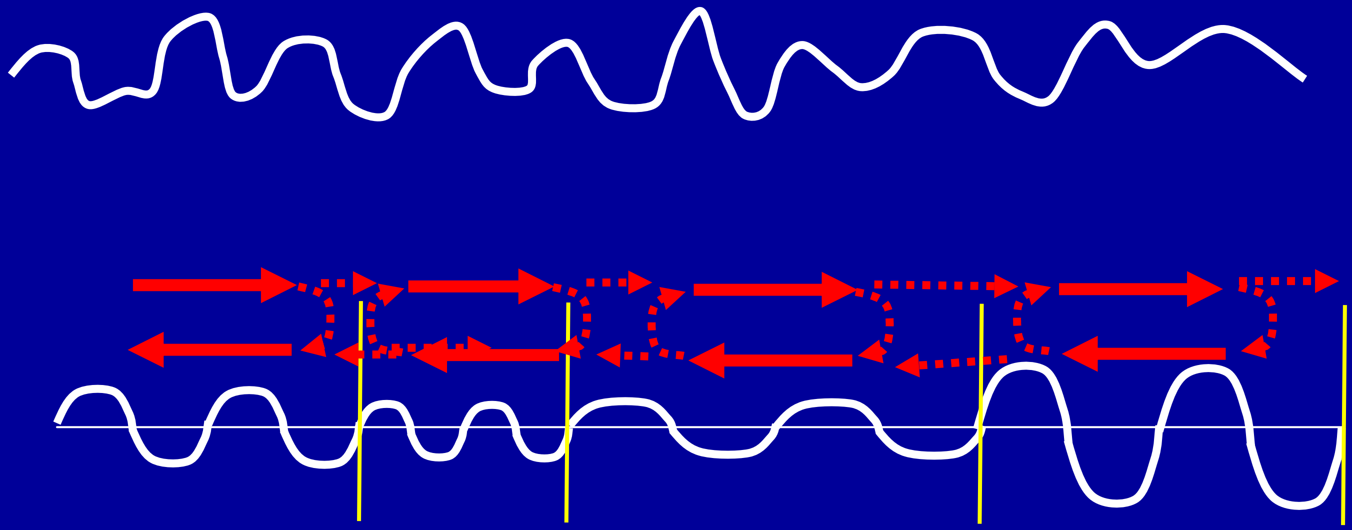
The greatest transmission on 100 realizations



The smallest transmission (and reflection)



Interpretation



In some parts of the surface, the local system of interference may be constructive, in such a way that the field becomes very strong and remains localized inside this region of resonance $\Rightarrow$ strong localization

Strong absorption and strong radiated field

Thanks

for your attention