

Plasmonics: a few basics

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- Photons and nanosystems
- Complex nanostructures
- Cold atoms, matter waves
- Biophotonic
- Optics & numerics (virtual reality)

Laboratoire Photonique, Numérique et Nanosciences (LP2N)

outline

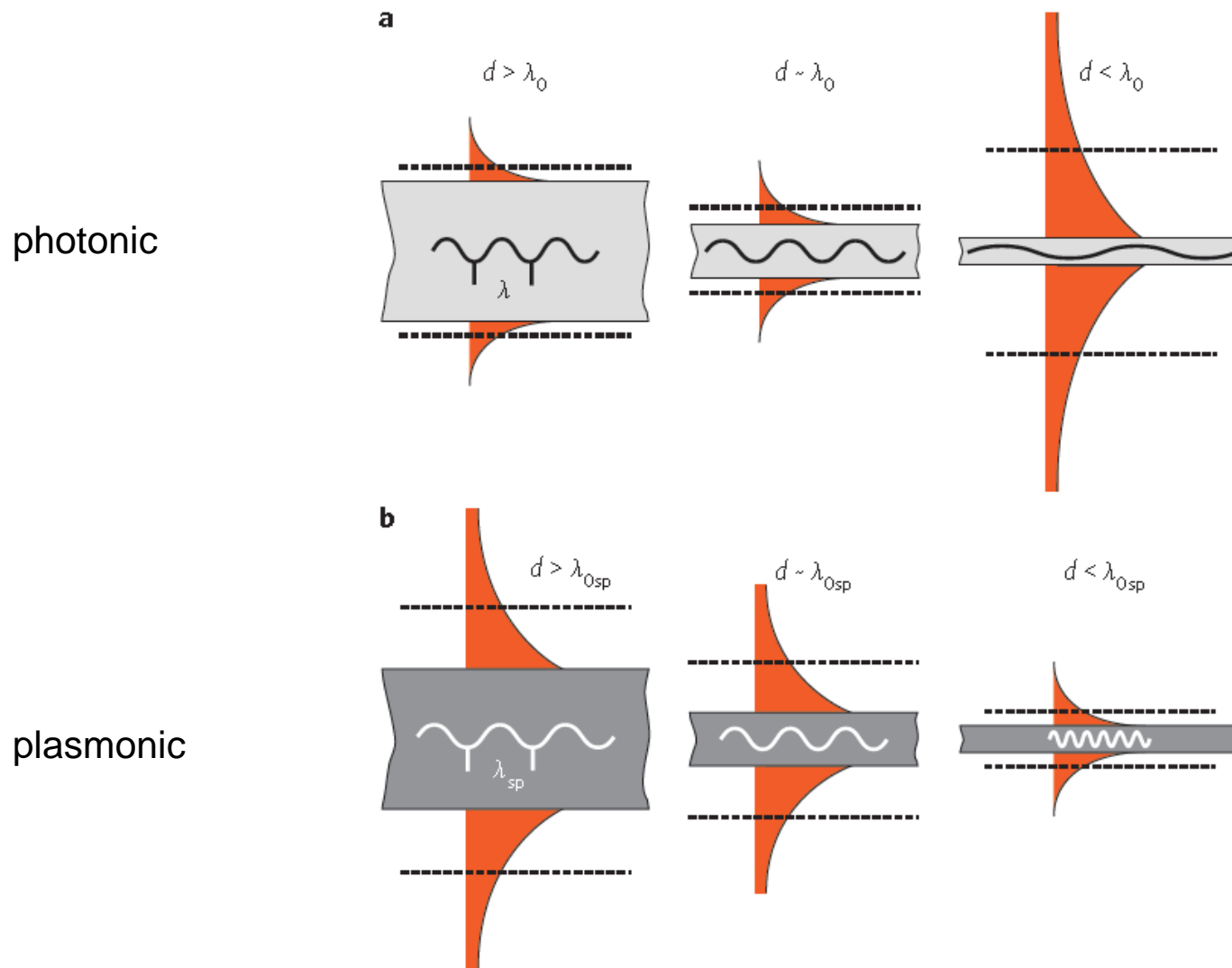
optics

physics

chemistry

- **Field localization (10mn)**
- Delocalized surface plasmons on metal surfaces
- Wood anomaly
- Localized plasmon
- The « end » of the plasmon

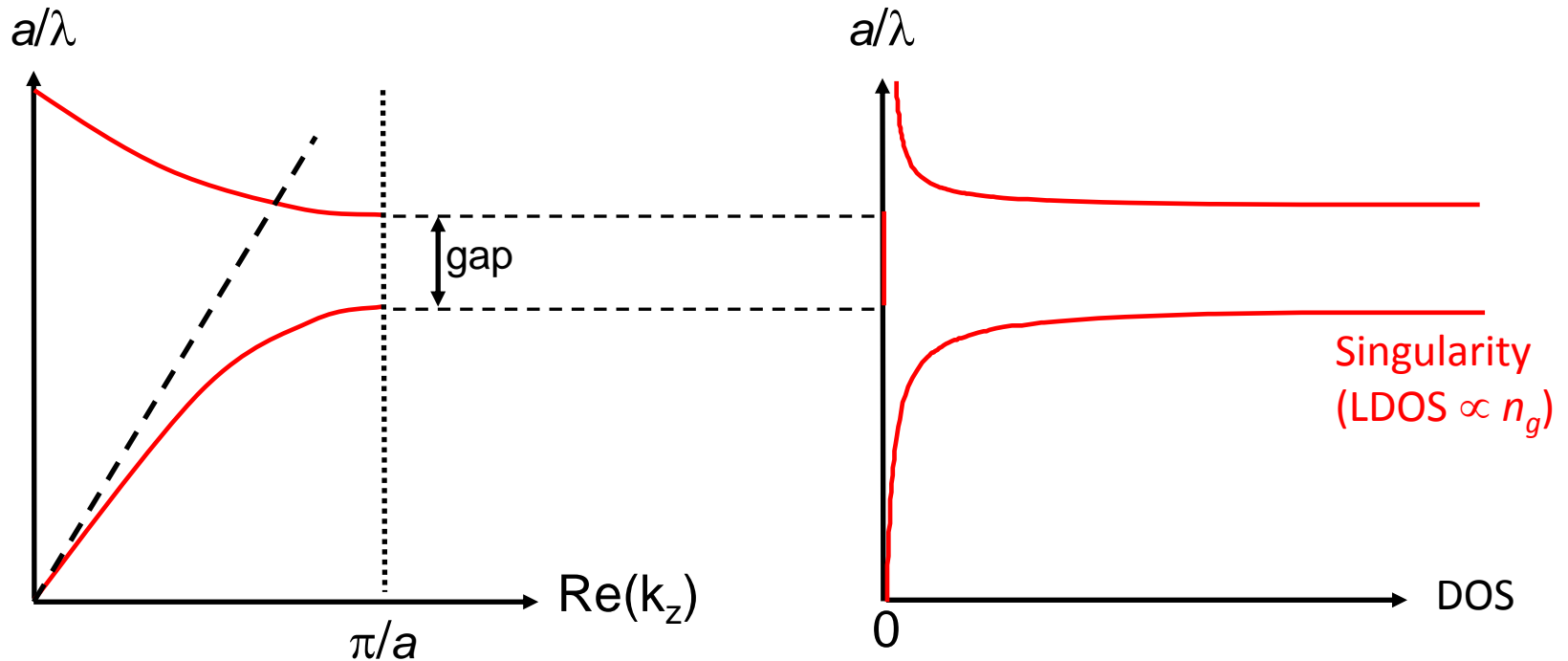
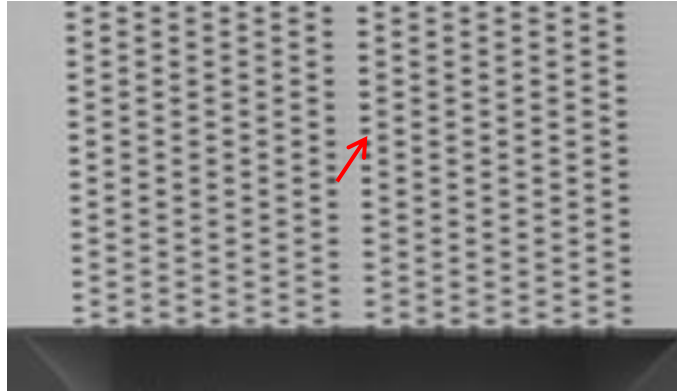
The magic confinement



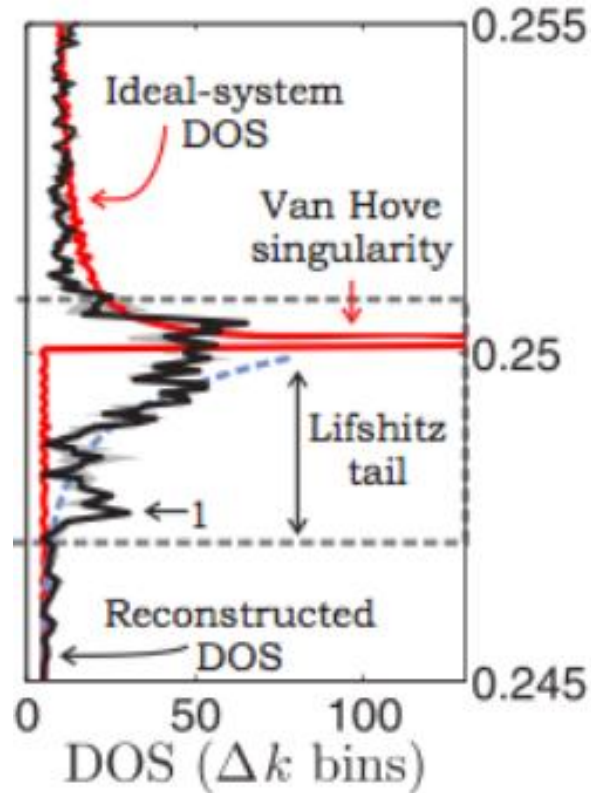
J. Takahara et al., Proc. SPIE 5604, 158 (2004).

D.K. Gramotnev and Sergey I. Bozhevolnyi, *plasmonics beyond the diffraction limit*, Nat. Photon. **4**, 83-91 (2010).

LDOS singularity in periodic systems



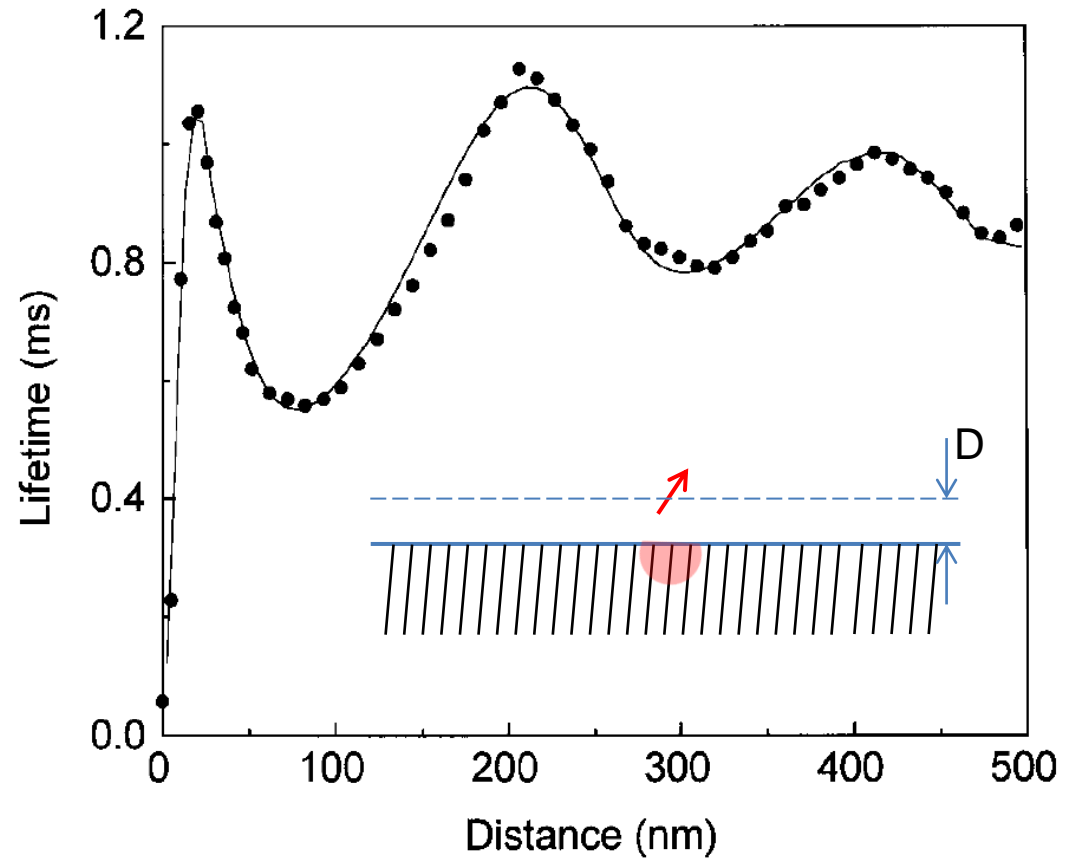
No singularity for real waveguide



Experimental reconstruction of the DOS in a photonic crystal waveguide

Quenching

Quenching is predicted by classical electromagnetic theory \rightarrow quenching is simply described by the dielectric constant of the substrate material.



The electron sea

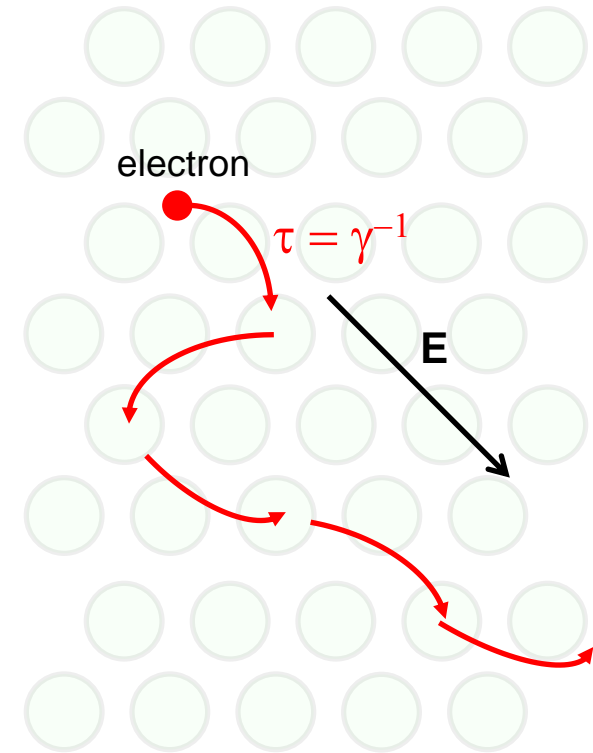
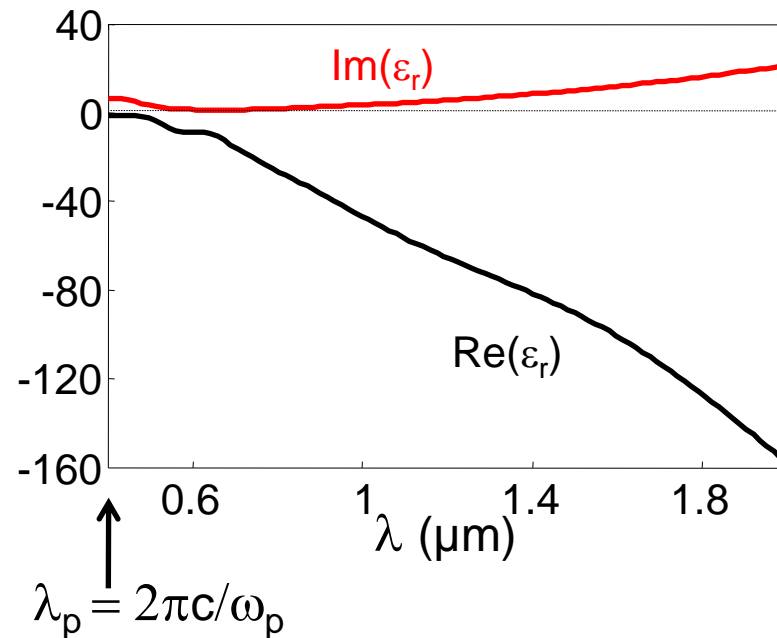
$$\ddot{\mathbf{x}} + \dot{\mathbf{x}}/\tau = -(e/m) \mathbf{E}_0 \exp(-i\omega t)$$

Dipole momentum

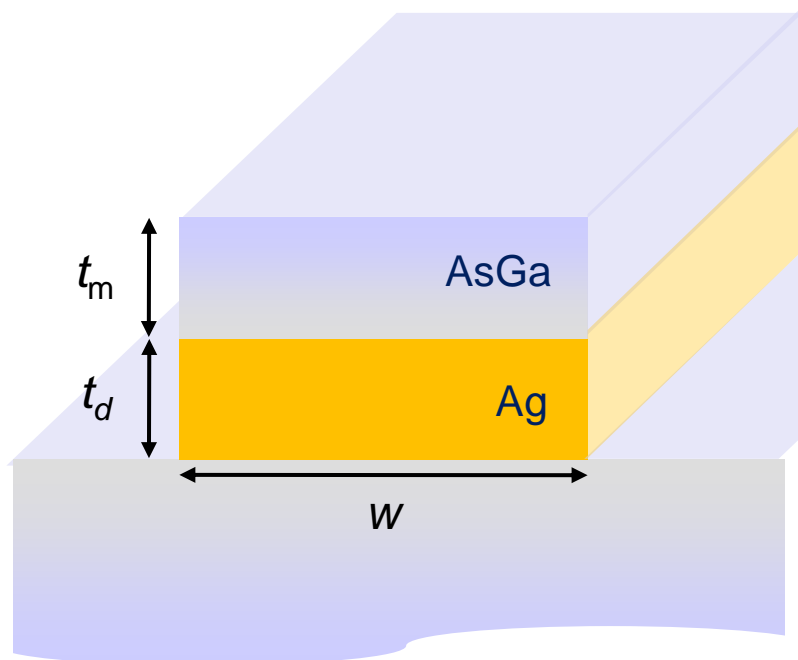
$$\mathbf{p}(\omega) = -e\mathbf{x}(\omega) = \varepsilon_0\alpha(\omega)\mathbf{E}(\omega)$$

Dielectric constant:

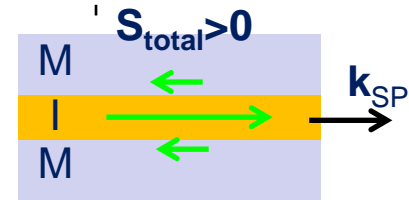
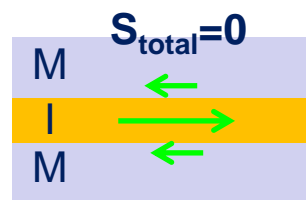
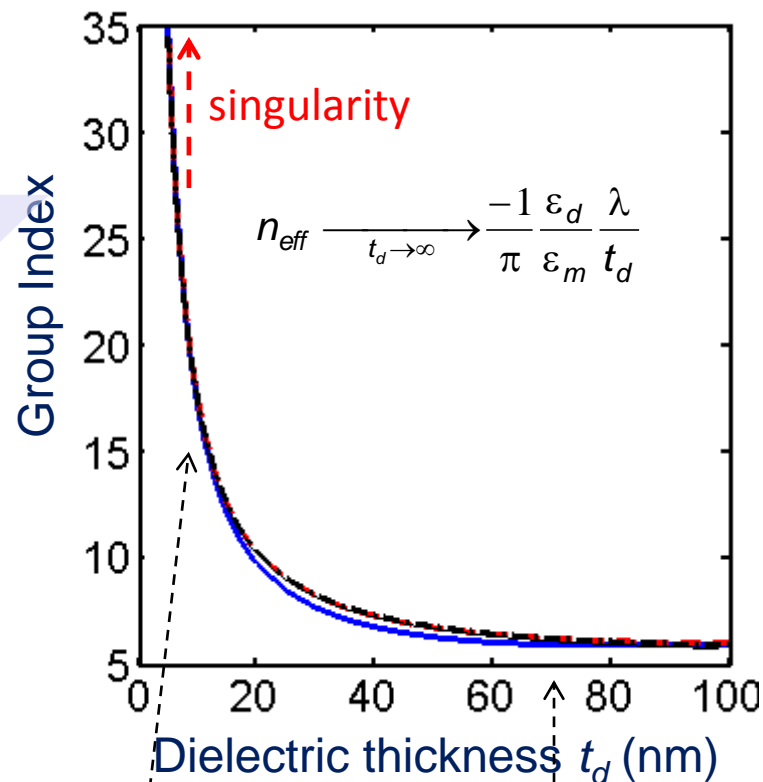
$$\varepsilon(\mathbf{r}) = 1 + N\alpha(\omega) = 1 - \frac{Ne^2}{m\varepsilon_0} \frac{1}{\omega^2 - i\omega\tau}$$



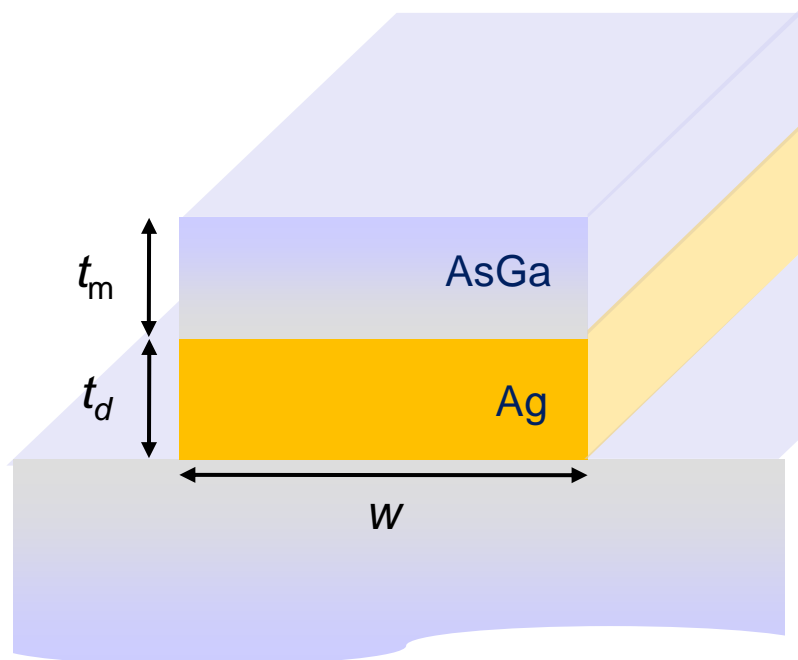
LDOS of MIM waveguides



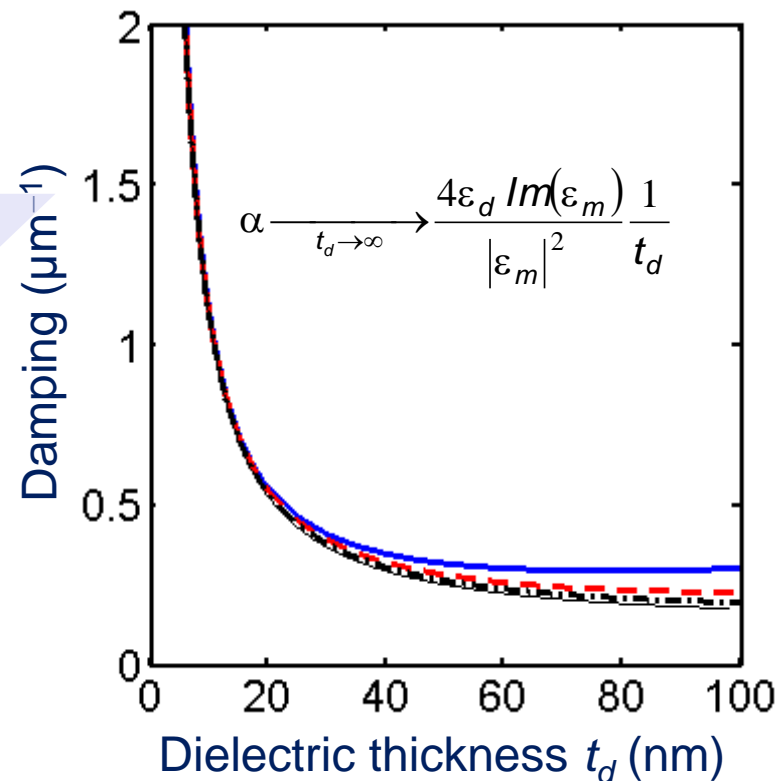
- $w=40$ nm
- - - $w=100$ nm
- · - · $w=350$ nm
- $w=\infty$



LDOS of MIM waveguides

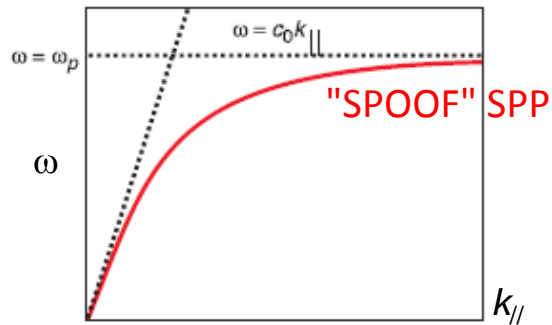


- $w=40$ nm
- - - $w=100$ nm
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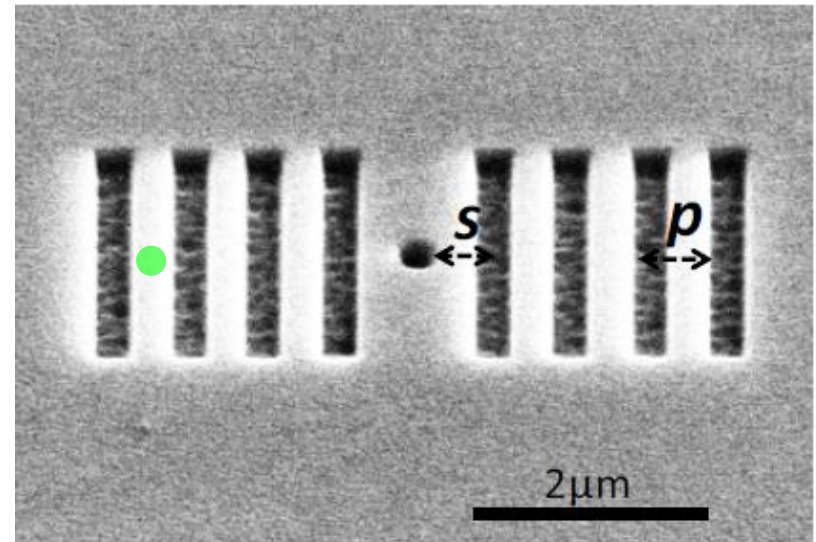


outline

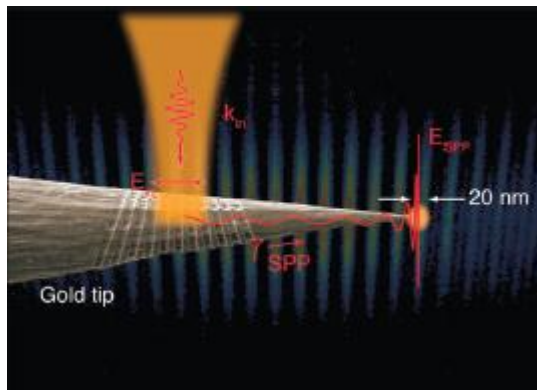
- Field localization
- **Delocalized surface plasmons on metal surfaces**
- Wood anomaly
- Localized plasmon
- The « end » of the plasmon



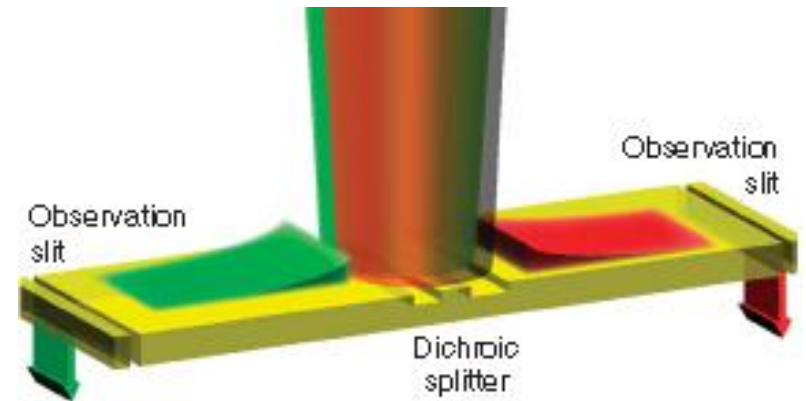
J. Pendry et al., Science **305**, 847 (2004).
 R. Ulrich and M. Tacke, APL **22**, 251 (1973).



Dark-field nanoscope: G.A. Zheng et al, PNAS **107**, 9043-48 (2010) .



Plasmonic nanofocussing for near-field spectroscopy: S. Berweger et al., Phys. Chem. Lett. **3**, 945 (2012) .



Submicron dichroic splitter: J. Liu et al, Nat. Comm., Nov. 2011.

Génération de plasmons avec des nanostructures

Questions:

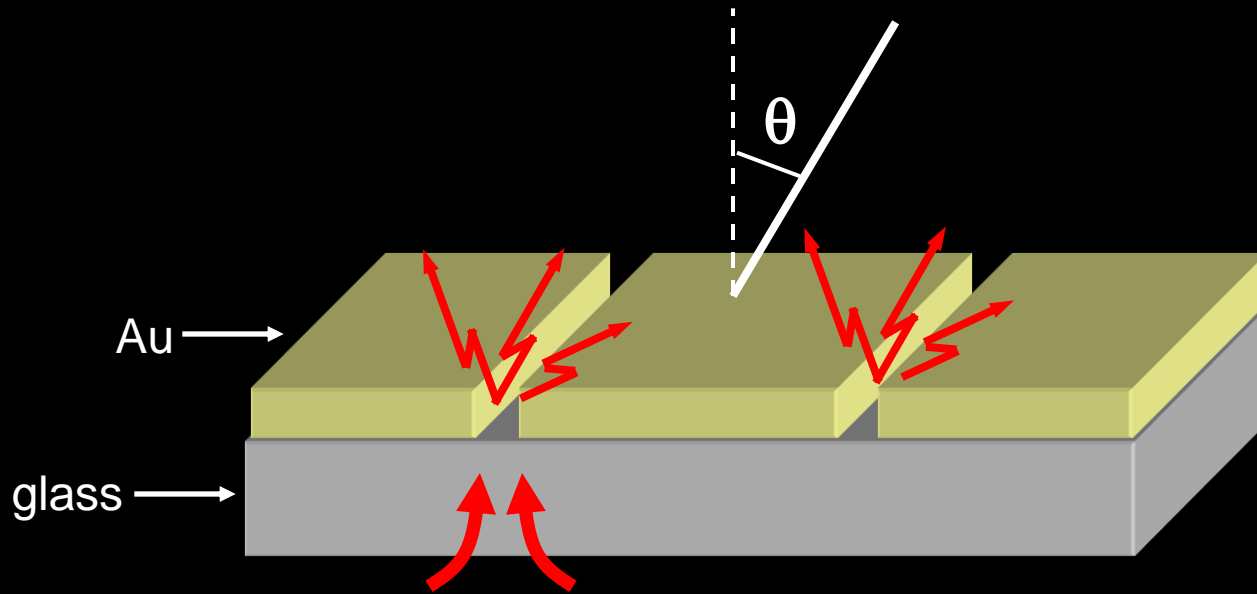
Comment peut-on mesurer ou calculer l'efficacité de génération des plasmons?

Comment exciter efficacement les plasmons de surface?

Comment cette efficacité varie avec les principaux paramètres?

Young slit experiment

(with a single slit illuminated)

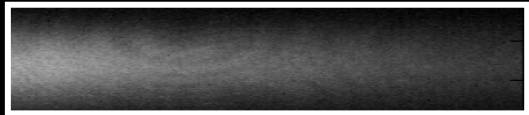


Kuzmin et al., *Opt. Lett.* **32**, 445 (2007).

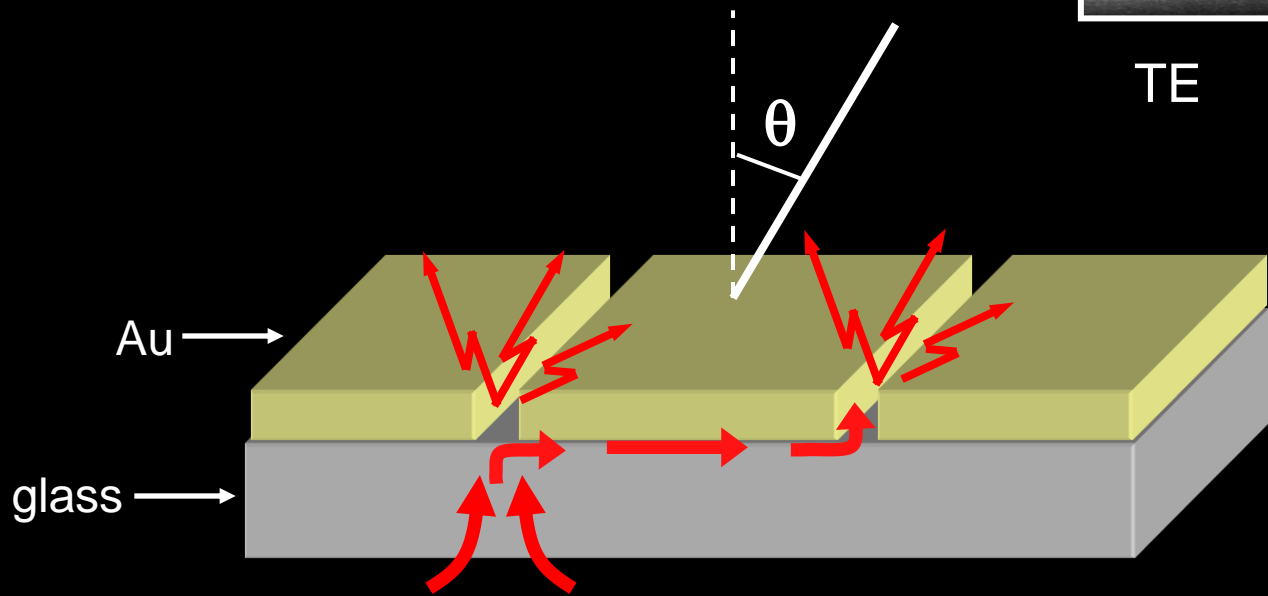
S. Ravets et al., *JOSA B* **26**, B28 (2009).

-40 -30 -20 -10 0 0 10 20 30

θ

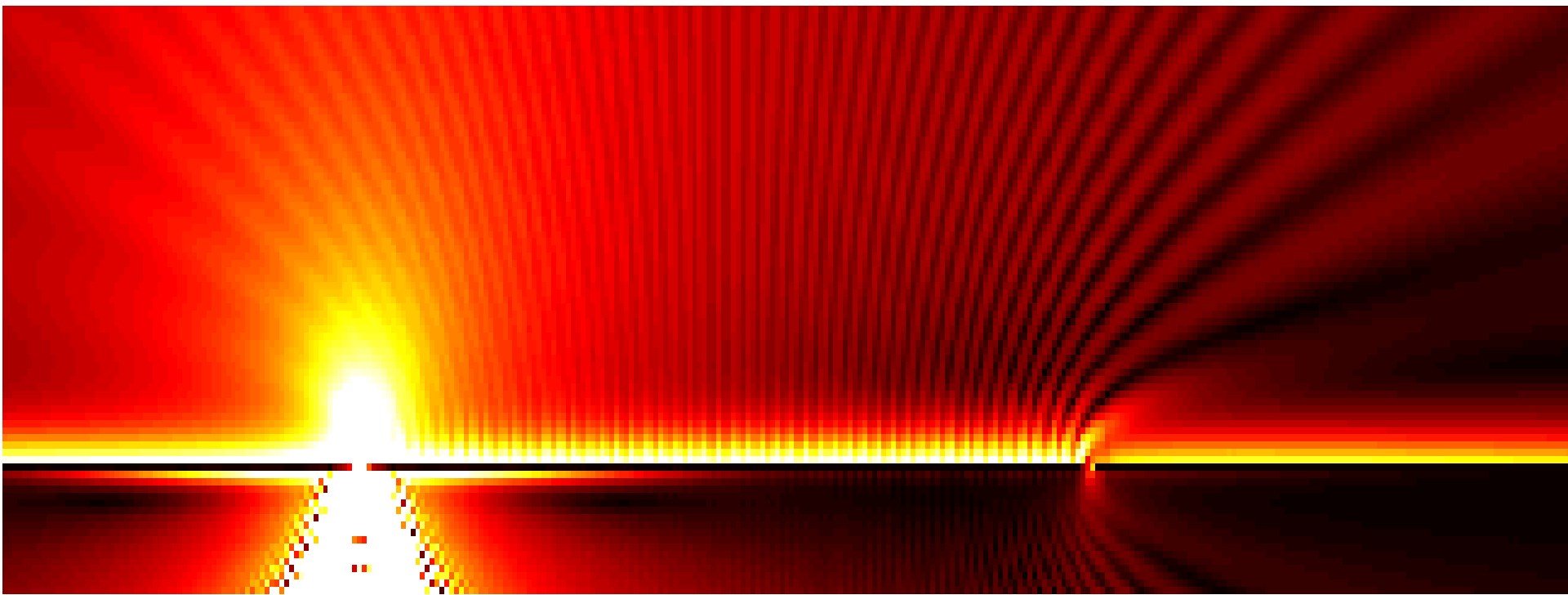


TE

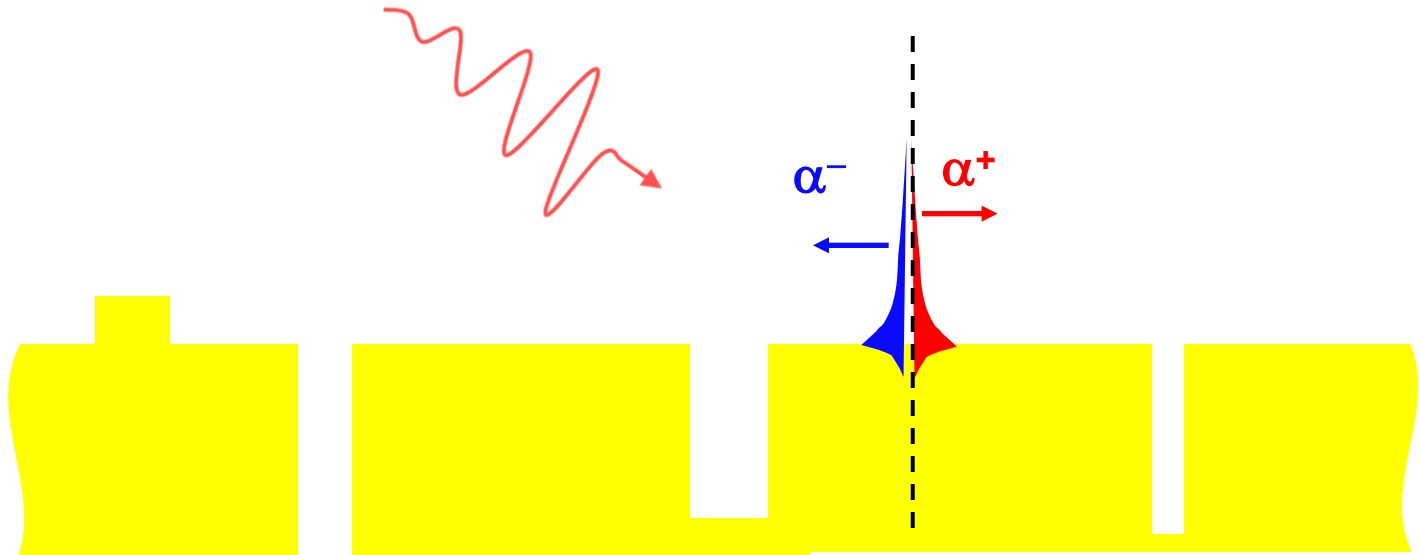


Kuzmin et al., *Opt. Lett.* **32**, 445 (2007).
S. Ravets et al., *JOSA B* **26**, B28 (2009).

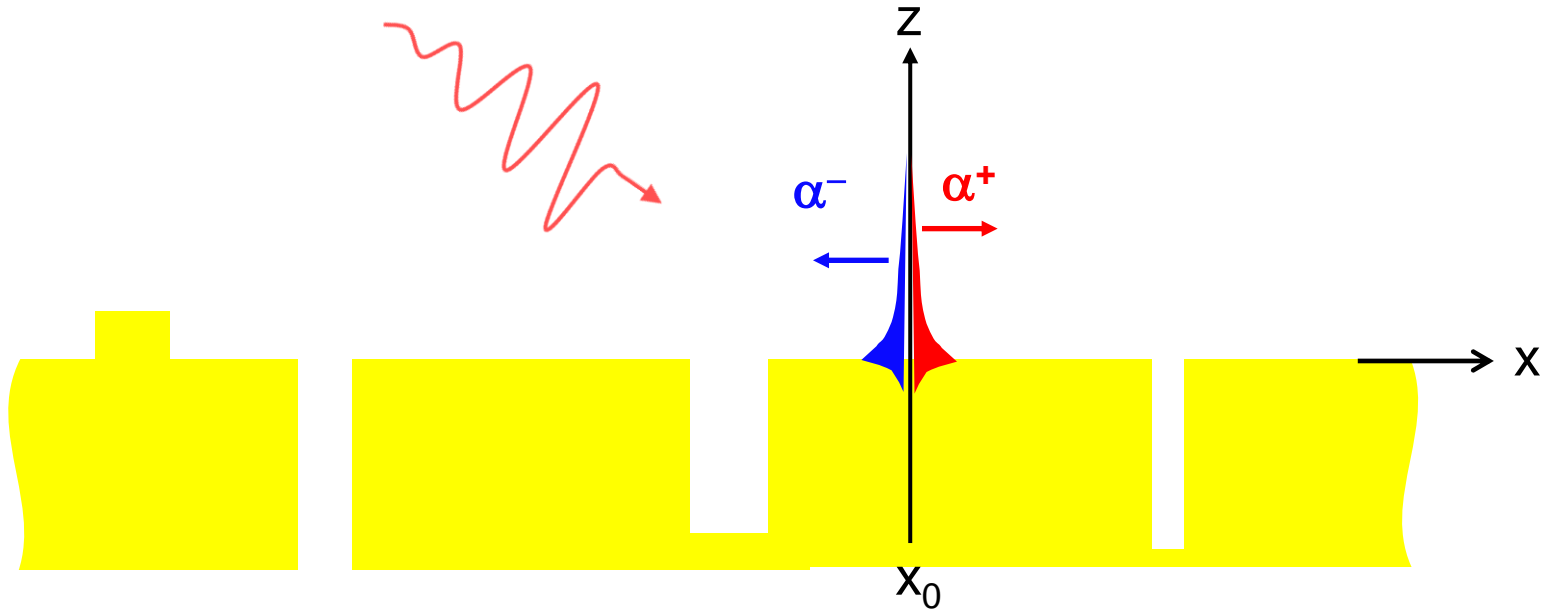
How to calculate the amount of SPP generated on the surfaces?



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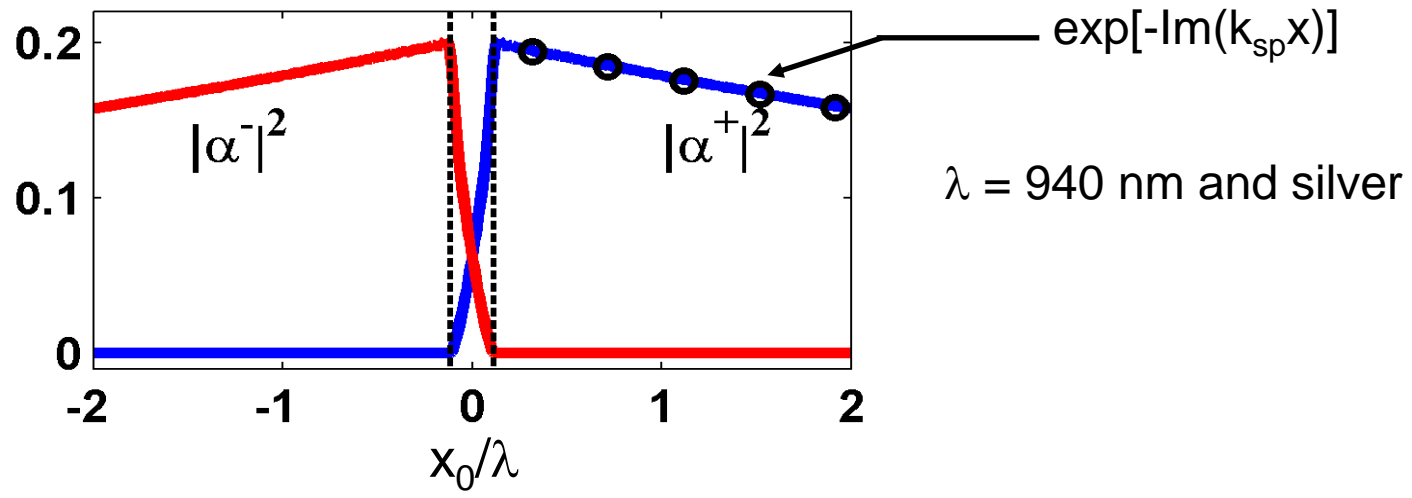
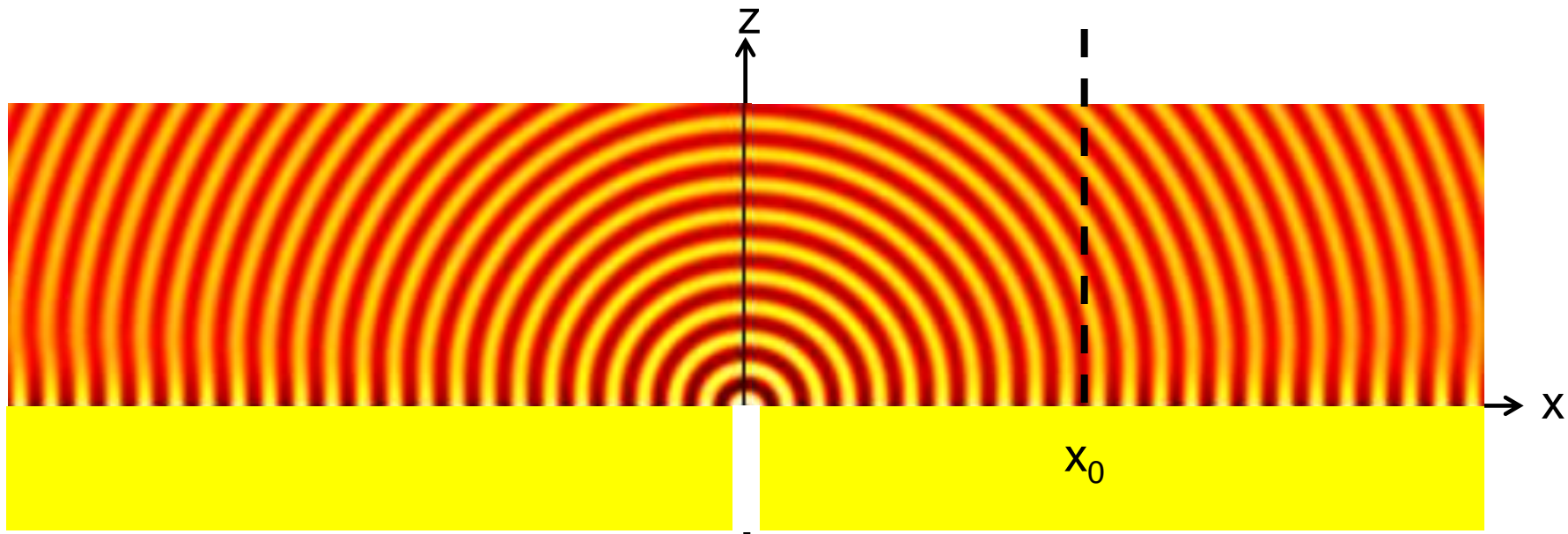
« Overlap integral »



$$\int_{-\infty}^{\infty} H_y(x_0, z) E_{SP}(z) dz = 2(\alpha^+(x_0) + \alpha^-(x_0))$$
$$\int_{-\infty}^{\infty} E_z(x_0, z) H_{SP}(z) dz = 2(\alpha^+(x_0) - \alpha^-(x_0))$$

Orthogonality is not implemented with EH^* products but with EH products

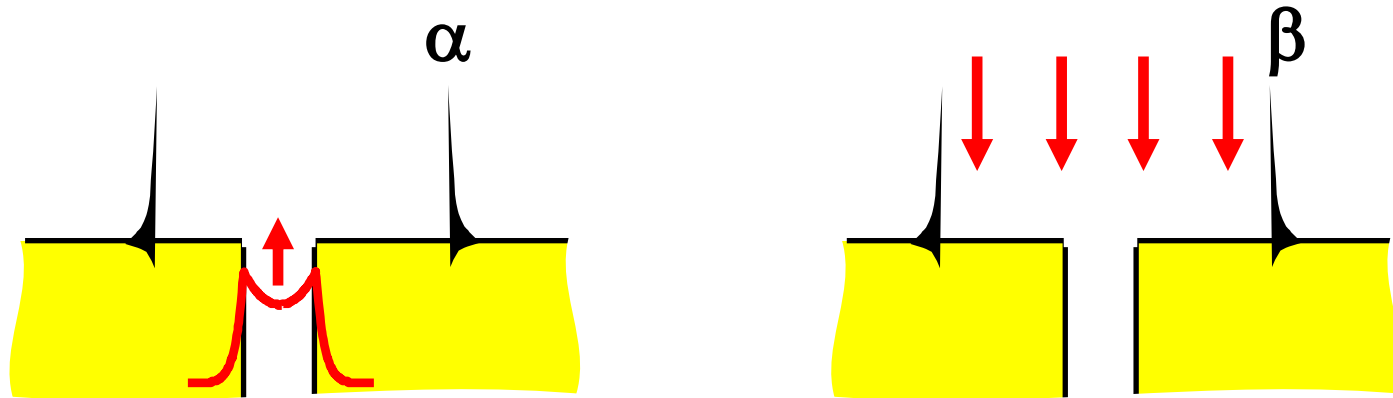
test



Il est bon de disposer de formule approcher pour mieux comprendre; ces formules ont été établie surtout pour les fentes.

Les résultats sont probablement généraux.

SPP generation by slits

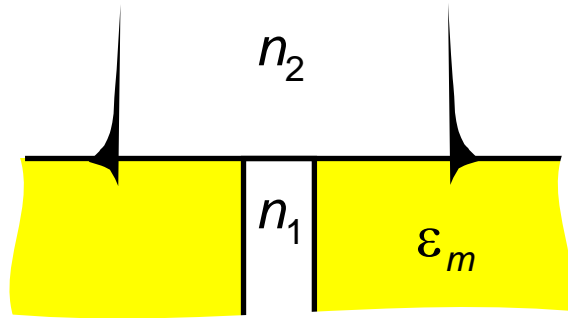


Normalization:

-incident field $E=1 \rightarrow$ effective SPP cross section

-intensity incident on the slit = 1 \rightarrow efficiency

Analytical model



$$|\alpha|^2 = |\beta|^2 = \underbrace{f(w/\lambda)}_{\text{blue}} \underbrace{\frac{n_2}{n_1} |\epsilon_m|^{-1/2}}_{\text{red}}$$

describe geometrical properties

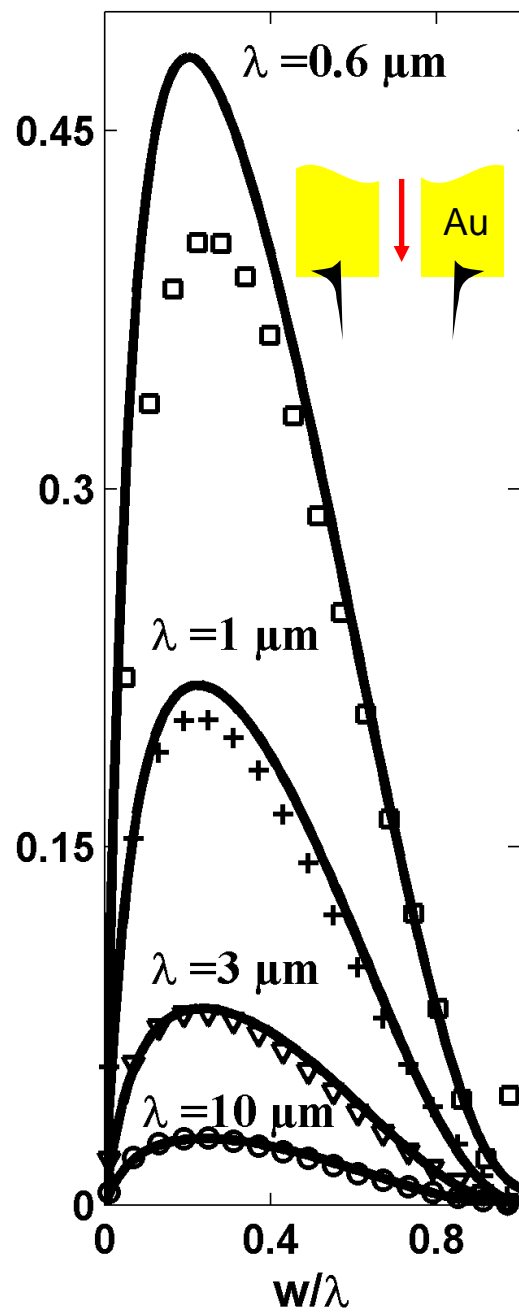
- the SPP excitation peaks at a value $w \approx \lambda/4$
- for visible frequency, $|\alpha|^2$ reach 0.5, which means that of the power coupled out of the slit half goes into heat

Expliquer avec Huygens pourquoi et dire que ce resultat devrait être vrai pour bcp de géométries

describe material properties

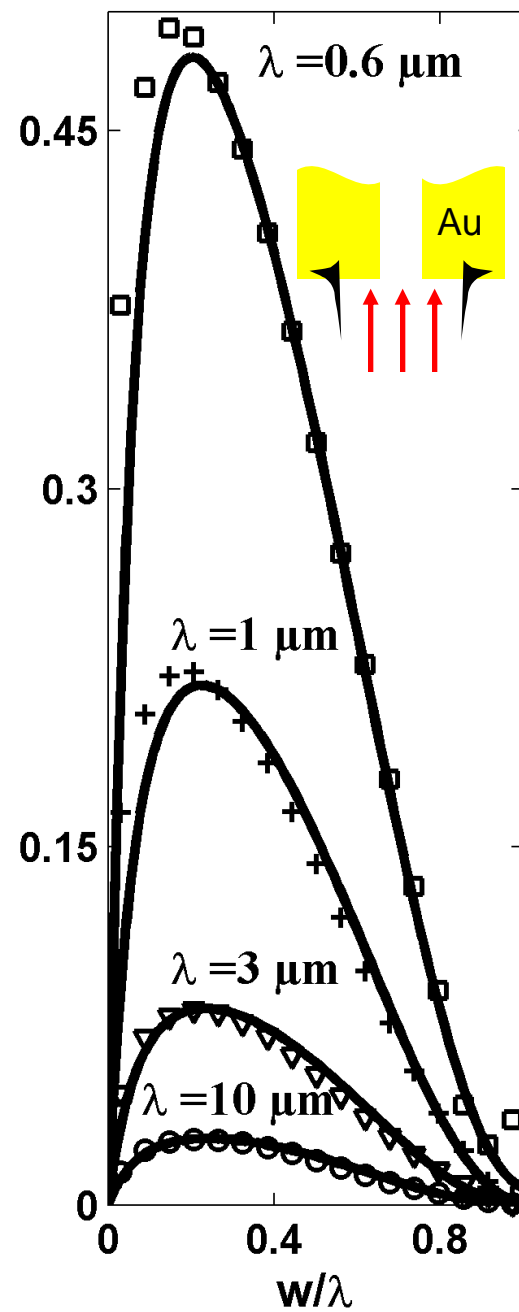
- Immersing the sample in a dielectric enhances the SP excitation ($\propto n_2/n_1$)
- The SPP excitation efficiency $|\alpha|^2$ scales as $|\epsilon_m(\lambda)|^{-1/2}$

Expliquer pourquoi avec l'intégrale de recouvrement et avec les mains

$|\alpha|^2$ 

solid curves
(analytical model)

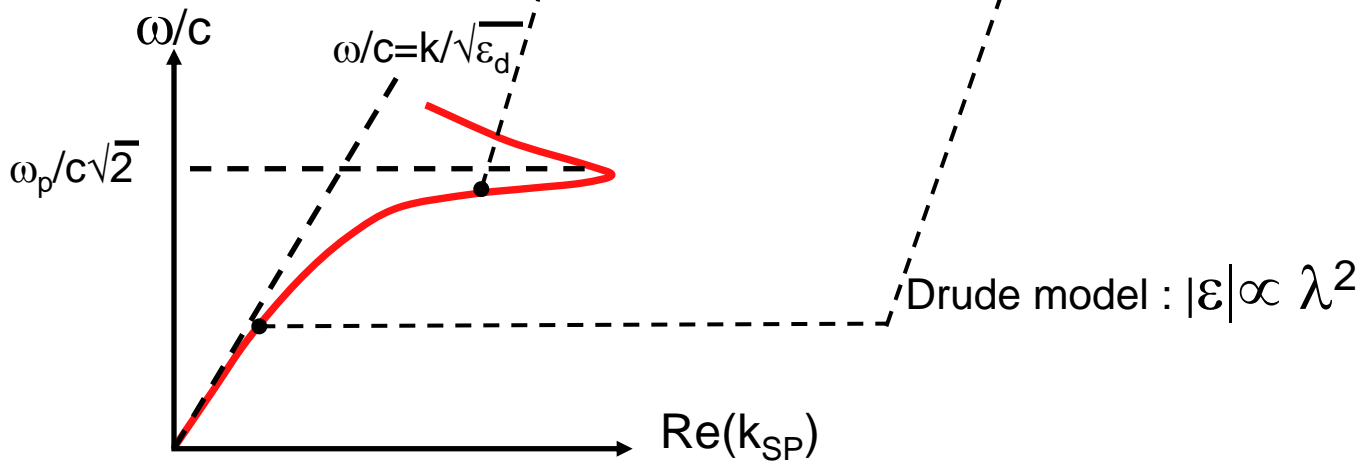
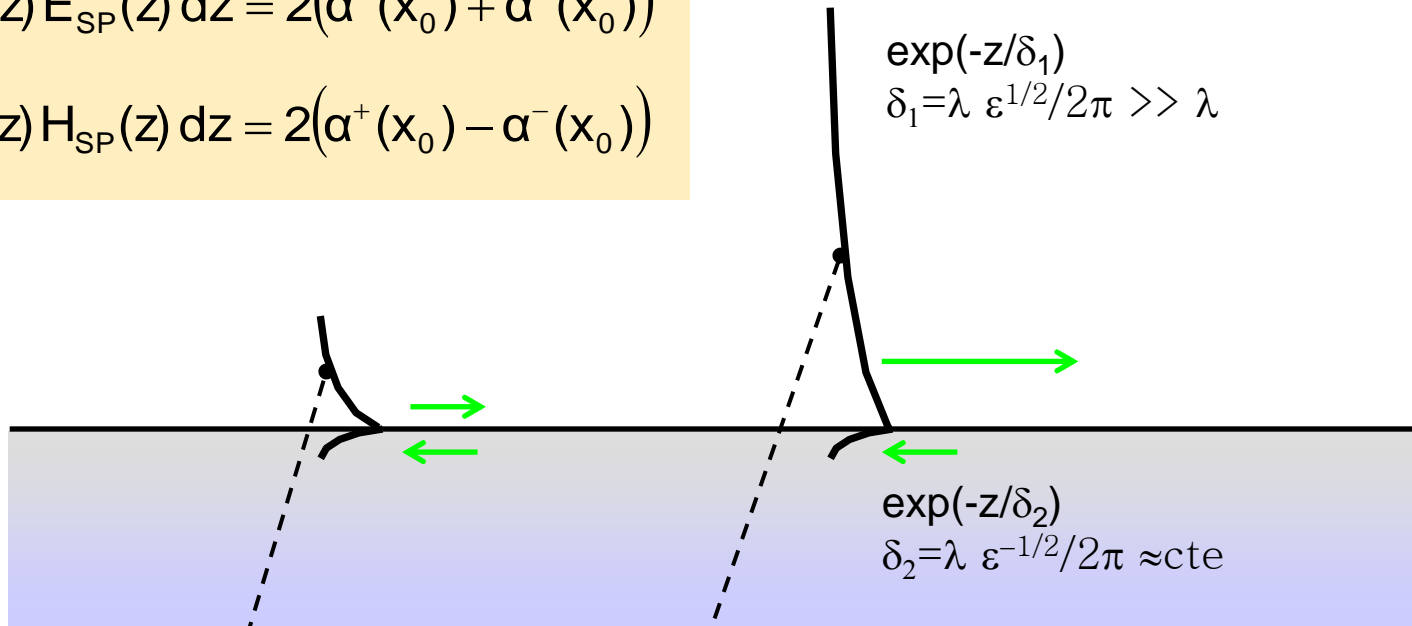
marks
(overlap integral)

 $|\beta|^2$ 

Surface plasmon polariton

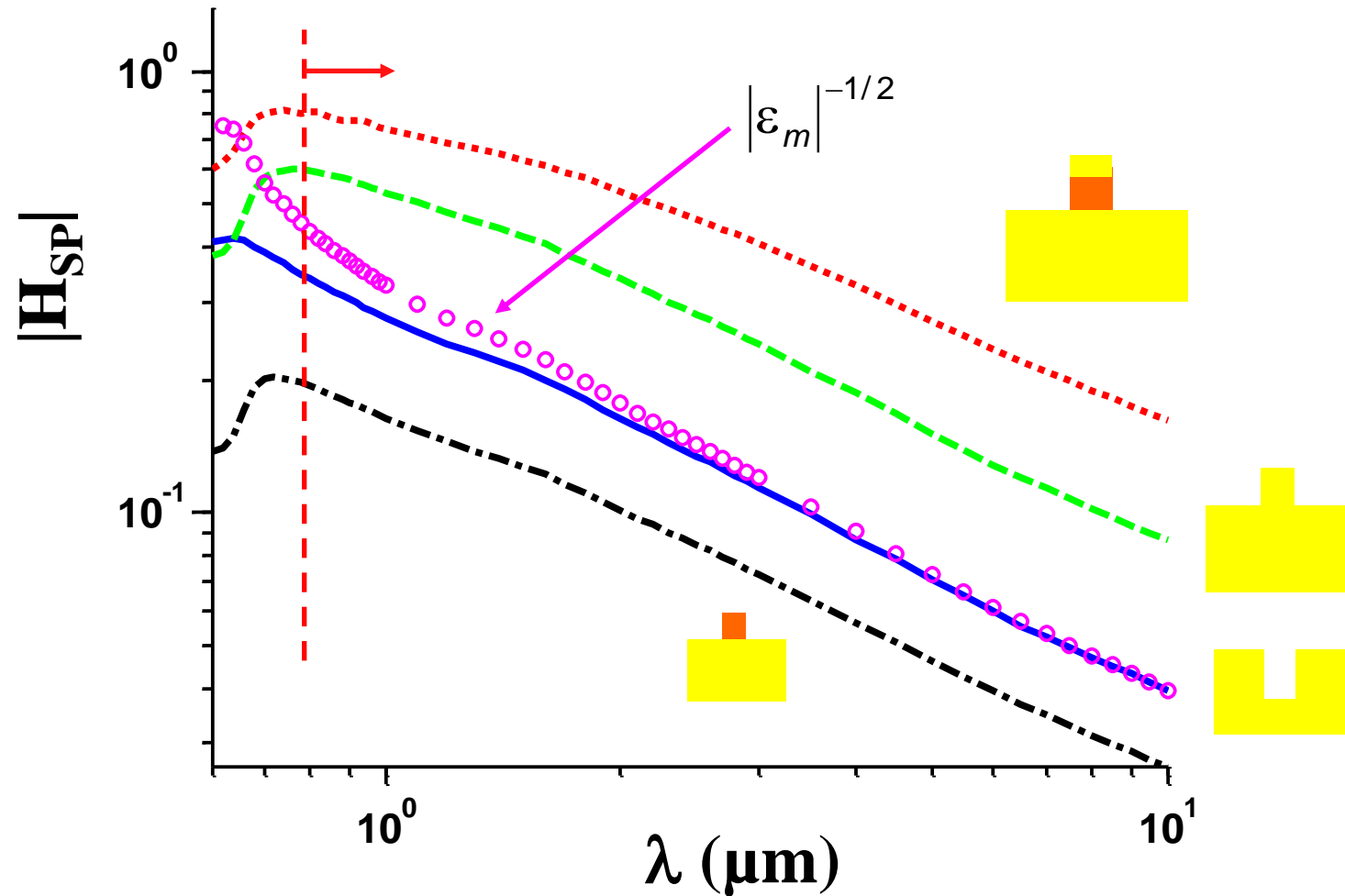
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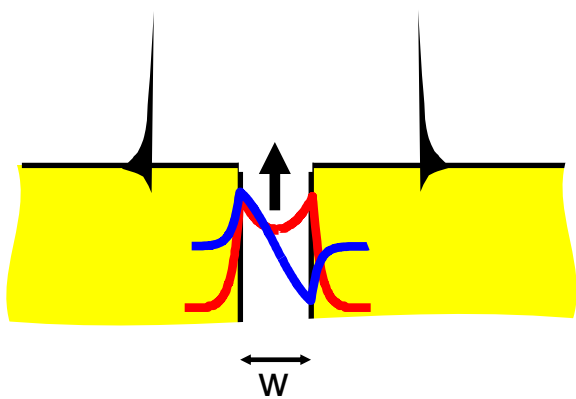


Valid for all subwavelength indentations

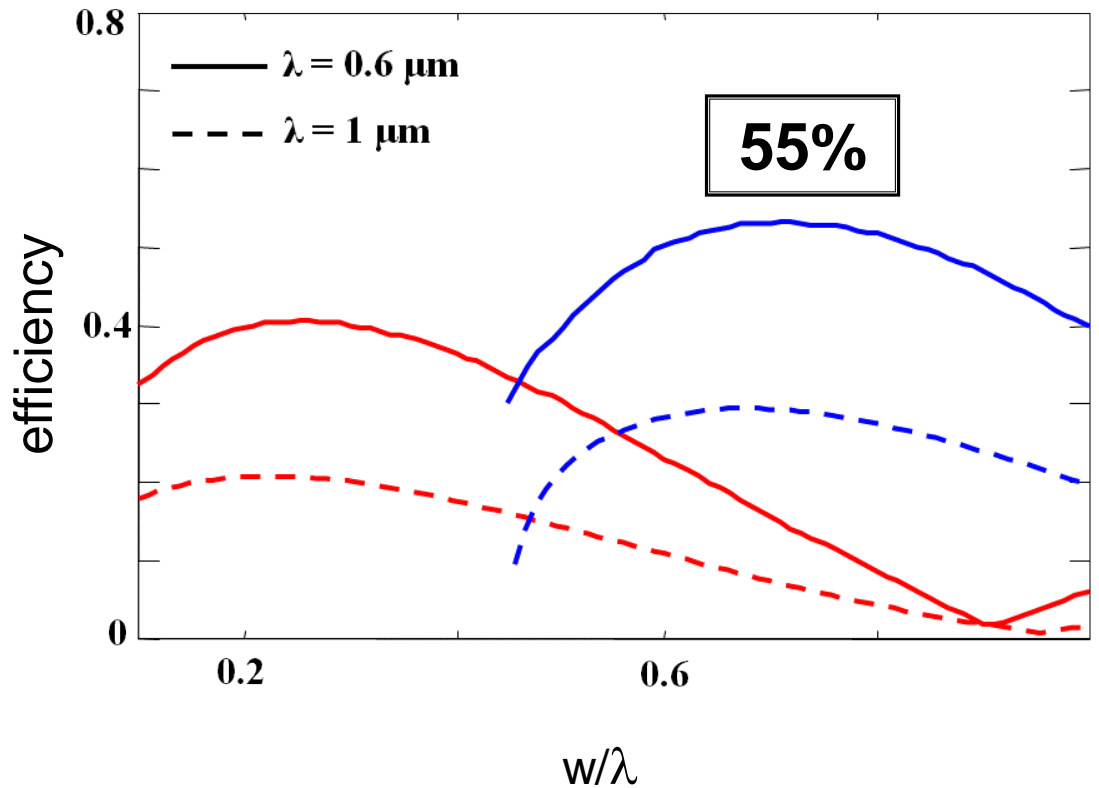
(results obtained for gold)



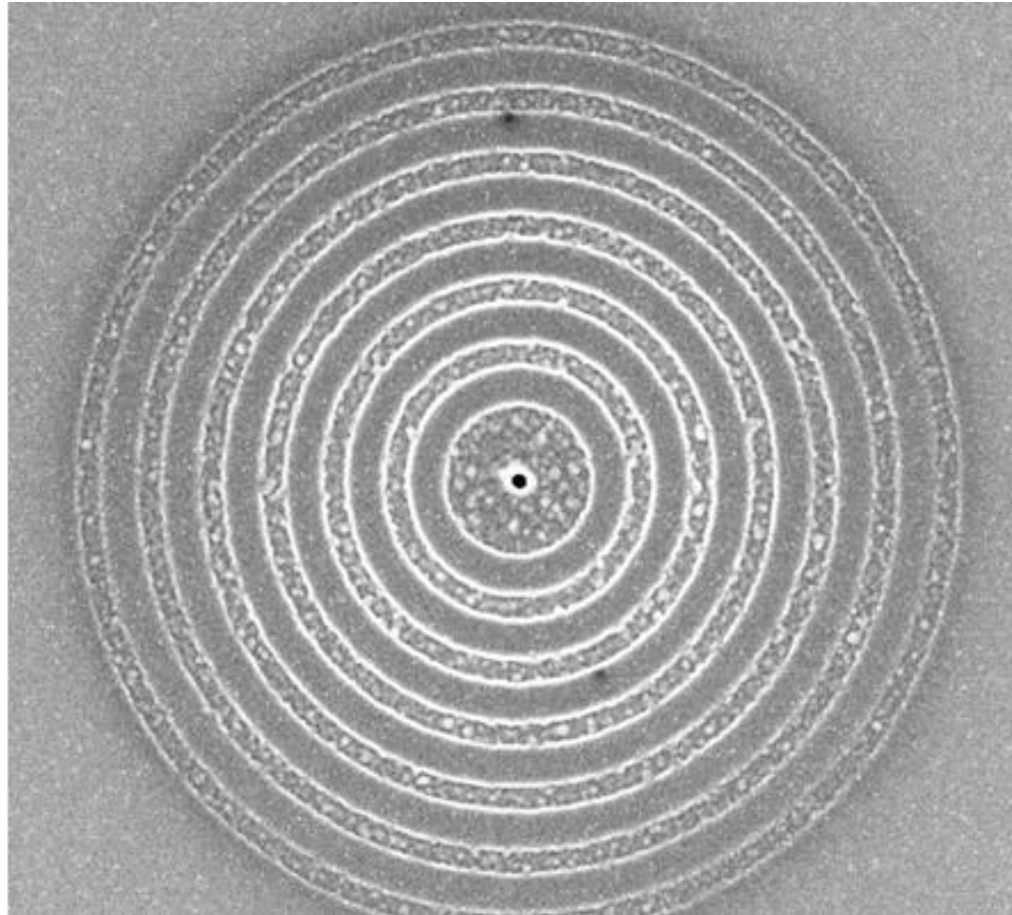
Anti-symmetric illumination (never mind!)



(results obtained for gold)



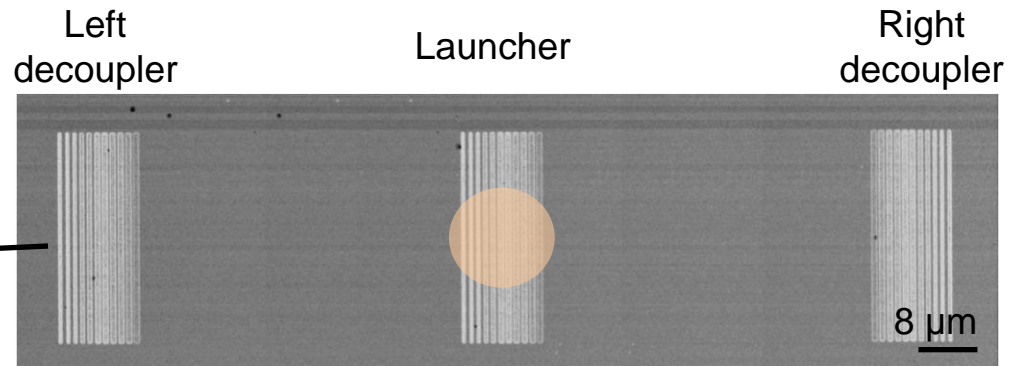
Unidirectional SPP launching with grooves arrays



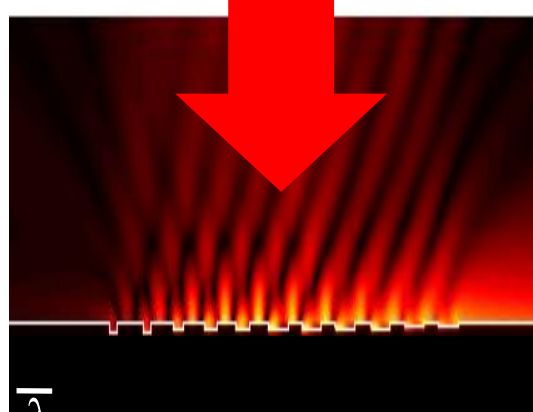
2 μm

Bull eye : H. Lezec et al., Science 297, 802-804 (2002).

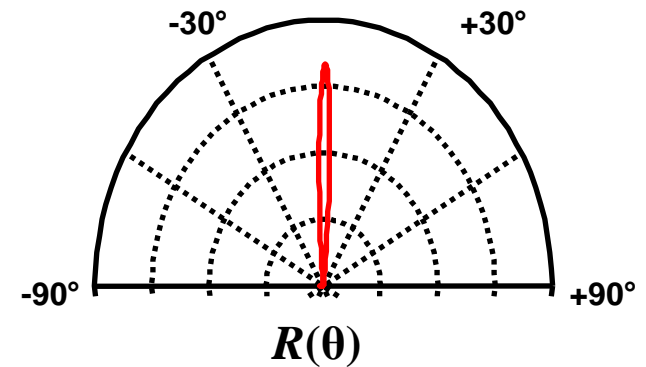
Unidirectional SPP launcher



Gaussian beam ($\lambda = 800 \text{ nm}$ waist = 6λ)



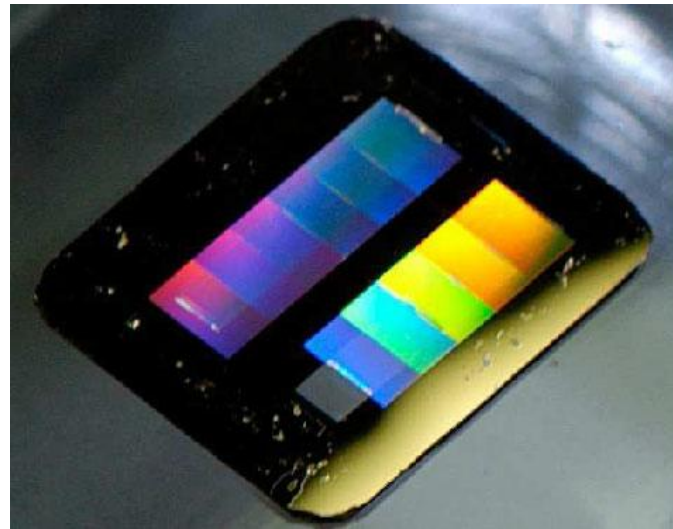
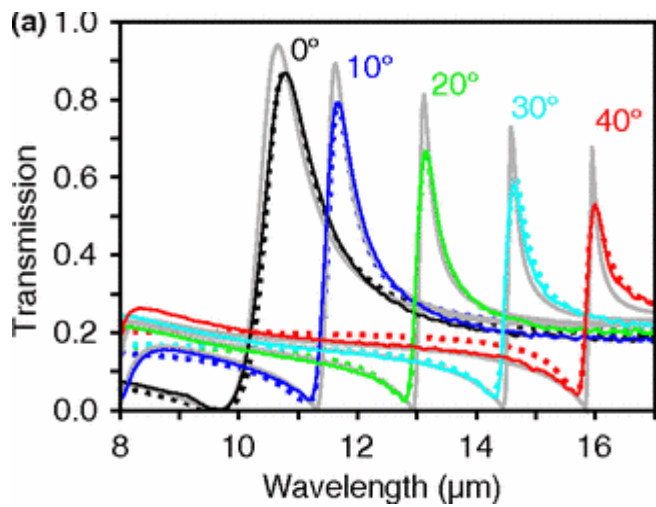
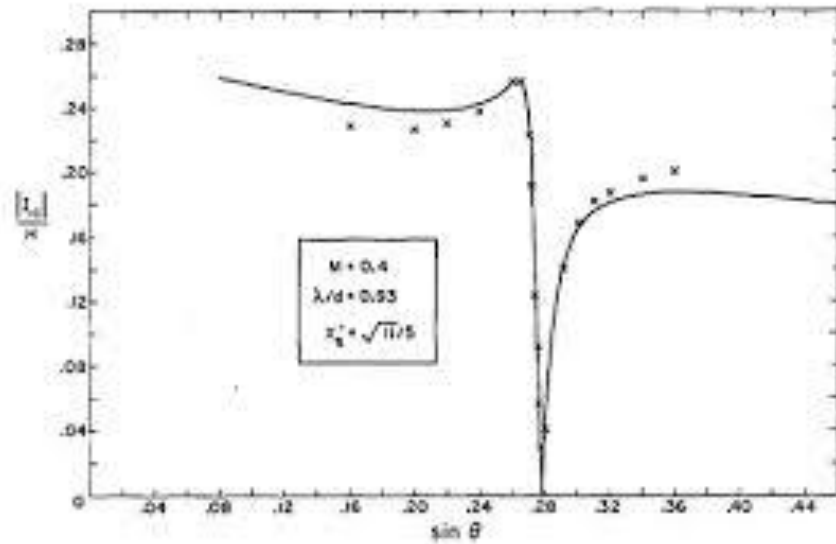
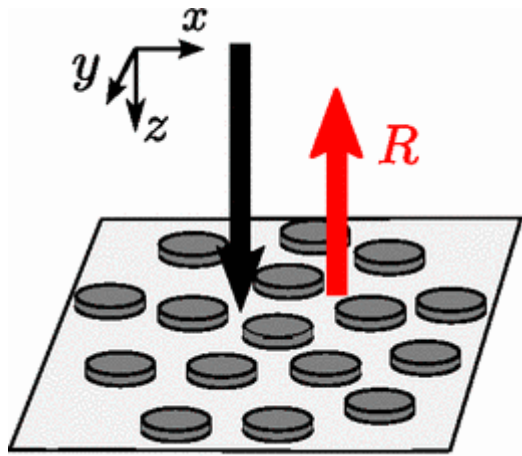
- Launching efficiency: $\eta_c^+ = 60\%$
- Contrast > 50



- Decoupling efficiency: $\eta_d = 75\%$
- Radiation cone: $< 10^\circ$

outline

- Field localization
- Delocalized surface plasmons on metal surfaces (30mn)
- **Wood anomaly**
- Localized plasmon
- The « end » of the plasmon



S. Collin et al., PRL 104, 027401 (2010).

- Historique de l'anomalie de Wood
- La description plasmonique de l'anomalie
- deux types d'onde sont mises en jeu: les plasmons et les ondes quasi-cylindriques
- Quelle est l'influence de la longueur d'onde sur le rôle de chacune des ondes?
- Commentaire sur le spoof plasmon

Rapid survey of Wood's anomalies

Discovery of the anomaly

R. W. Wood, *Philos. Mag.* 4, 396 (1902).

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

First explanation attempt by Lord Rayleigh

Rayleigh, *Proc. Royal Society (London)* 79, 399 (1907)

$$k_{//} + mK = k_0$$

The forced resonance explanation of Fano

U. Fano, *JOSA* 31, 213 (1941).

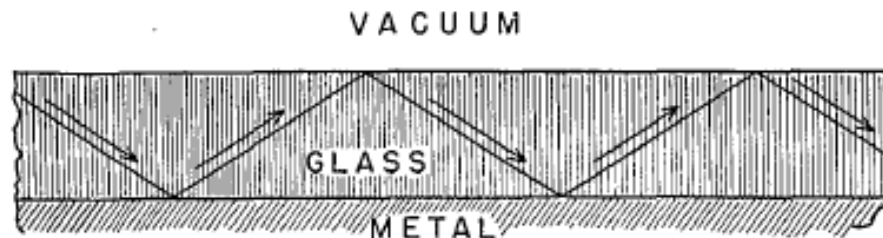


FIG. 4. Schematic path of a light wave progressing within a glass plate between a metal and a vacuum.

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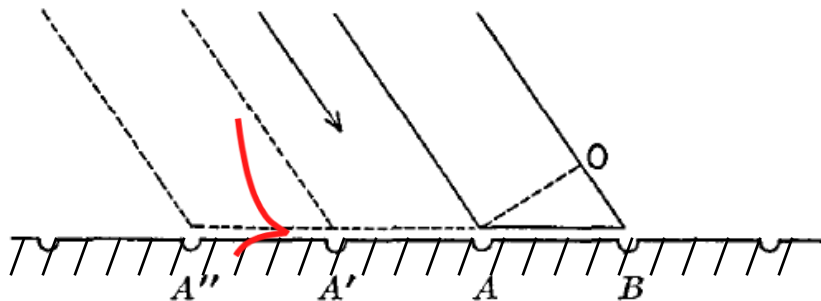
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$$k_{//} + mK = k_{SPP} (>k_0)$$

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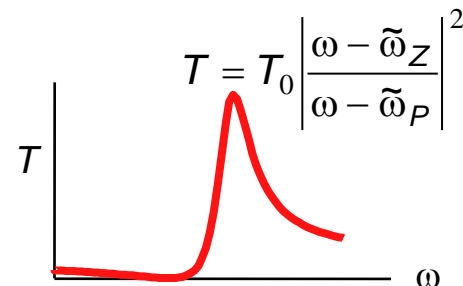
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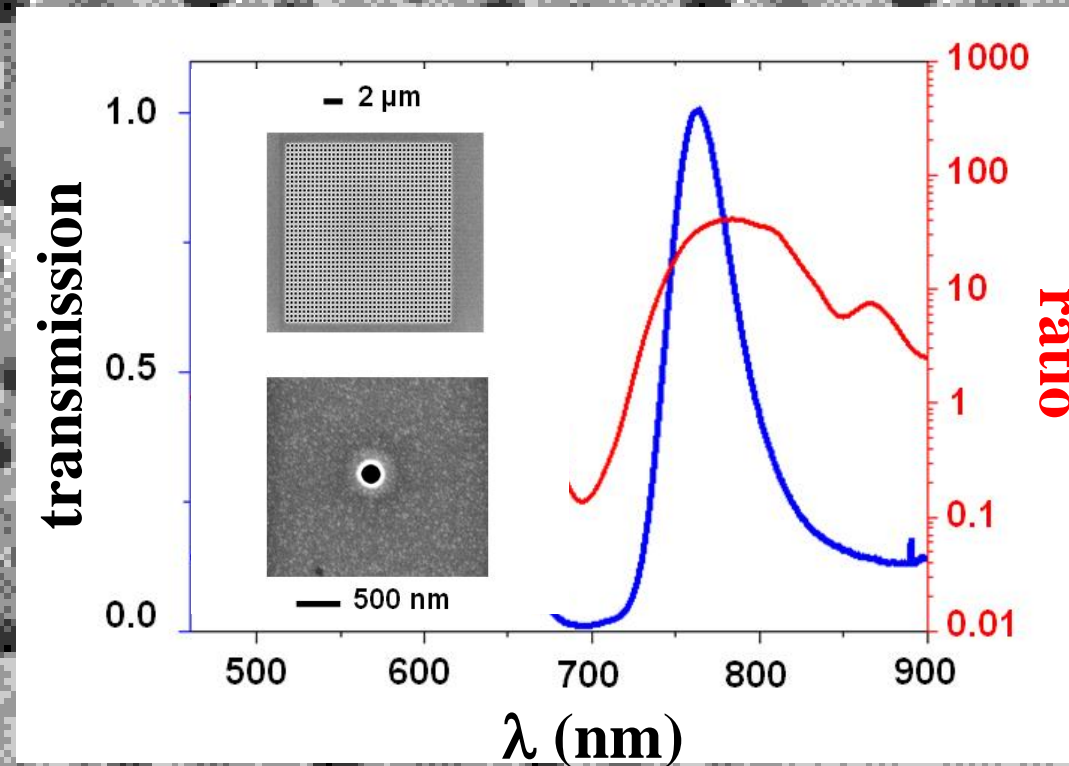
Modern theory of grating diffraction

✓ Fully-vectorial numerical tools: Integral, differential methods, RCWA, ...

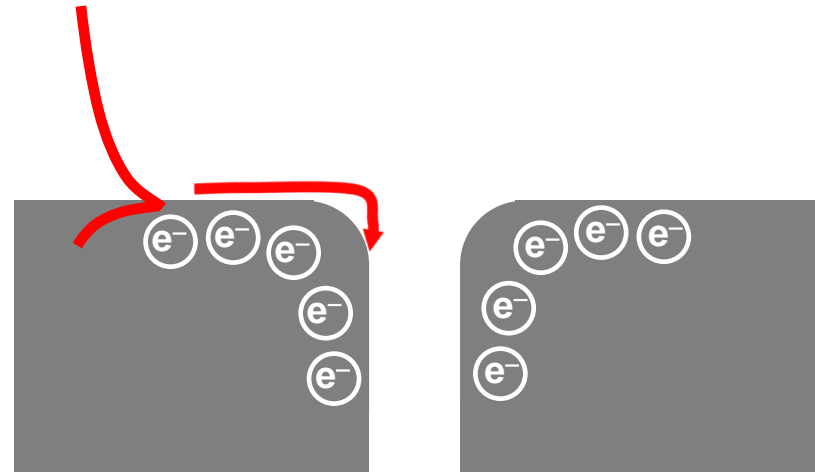
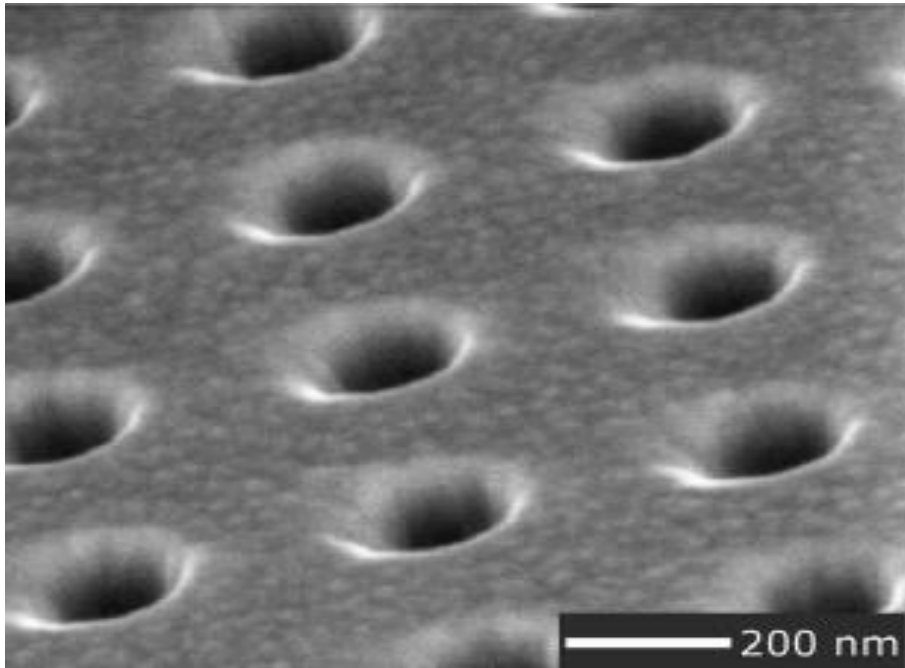
✓ Advanced conceptual tool: « polology »



The extraordinary optical transmission



SPP-assisted transmission?



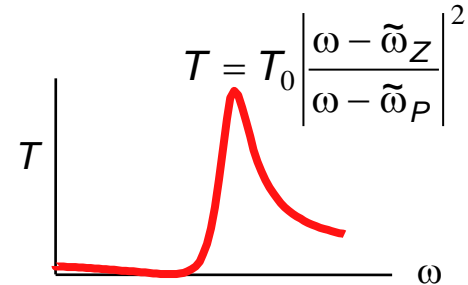
Debated hypothesis: The electronic character of SPP helps the coupling of the energy from the surface to the holes?

Main results from mode theory

Phenomenological polology

E. Popov et al., PRB **62**, 16100 (2000).

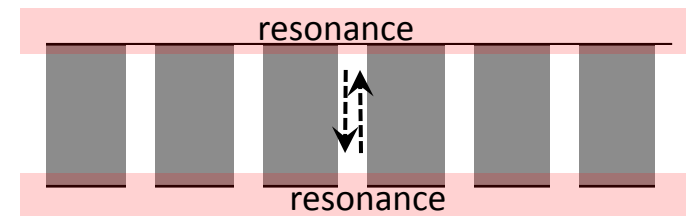
The Fano-type formula is very elegant as it well reproduce the spectral lineshape with o,nly 5 real parameters. It additionnally shows that the EOT is a resonance phenomenon.



Resonance-assisted tunneling

L. Martín-Moreno et al., PRL **86**, 1114 (2001).

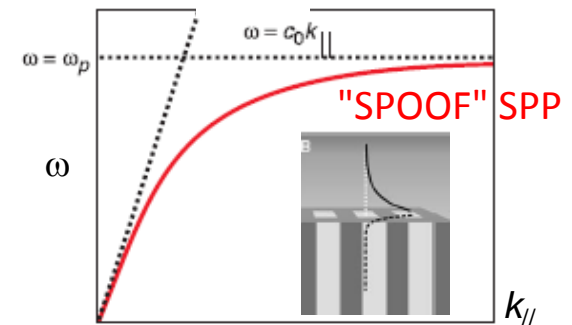
More insight has been provided by Martin-Moreno who showed that the resonance occurs at interfaces and that they boosts an evanescent tuneling.



Spoof plasmon

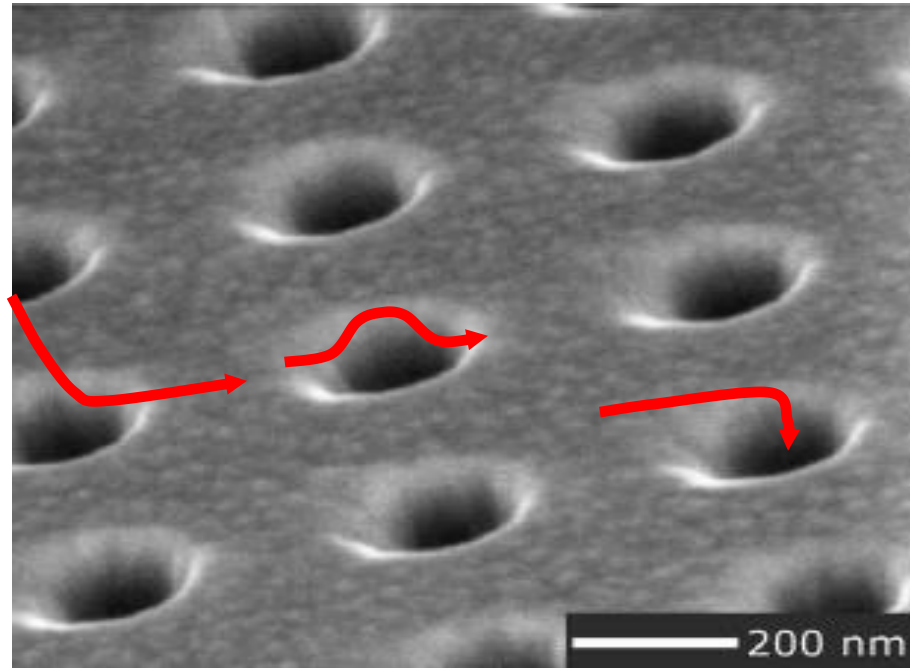
J. Pendry et al., Science **305**, 847 (2004).

Pendry et al. showed that the same resonant-assisted mechanism occurs at low frequencies, and introduced the concept of spoof plasmons.



All these analysis relies on physical 'GLOBAL' quantities attached to periodic ensembles; they give a good insight into the macroscopic mechanisms responsible for the transmission, but nothing is known about the individual plasmons that are launched inbetween the holes of the array.

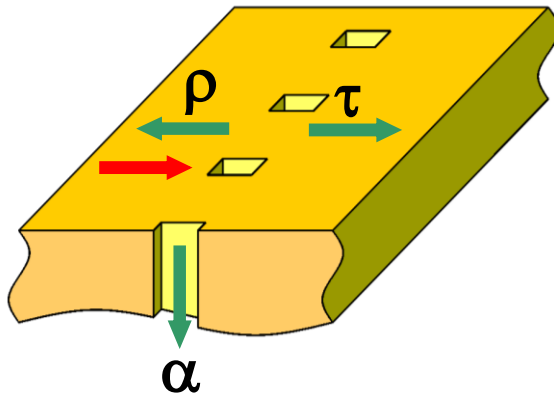
SPP-assisted transmission?



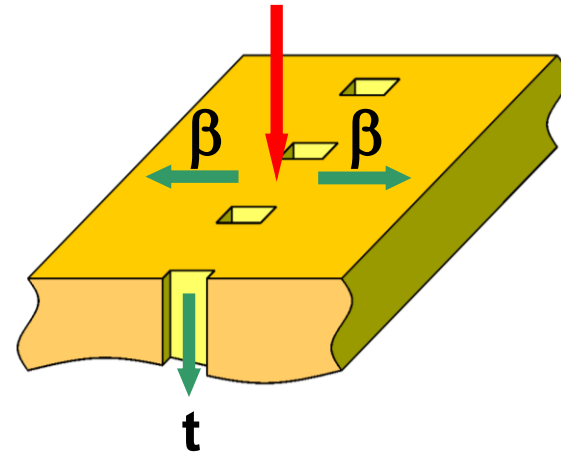
If one derives a model of the EOT where only SPP are assumed to carry the energy between adjacent hole chains and compares with fully-vectorial computations, then one should allow us to quantify what is really due to SPP in the EOT.

Microscopic SPP model

in-plane reflection-
transmission of SPP



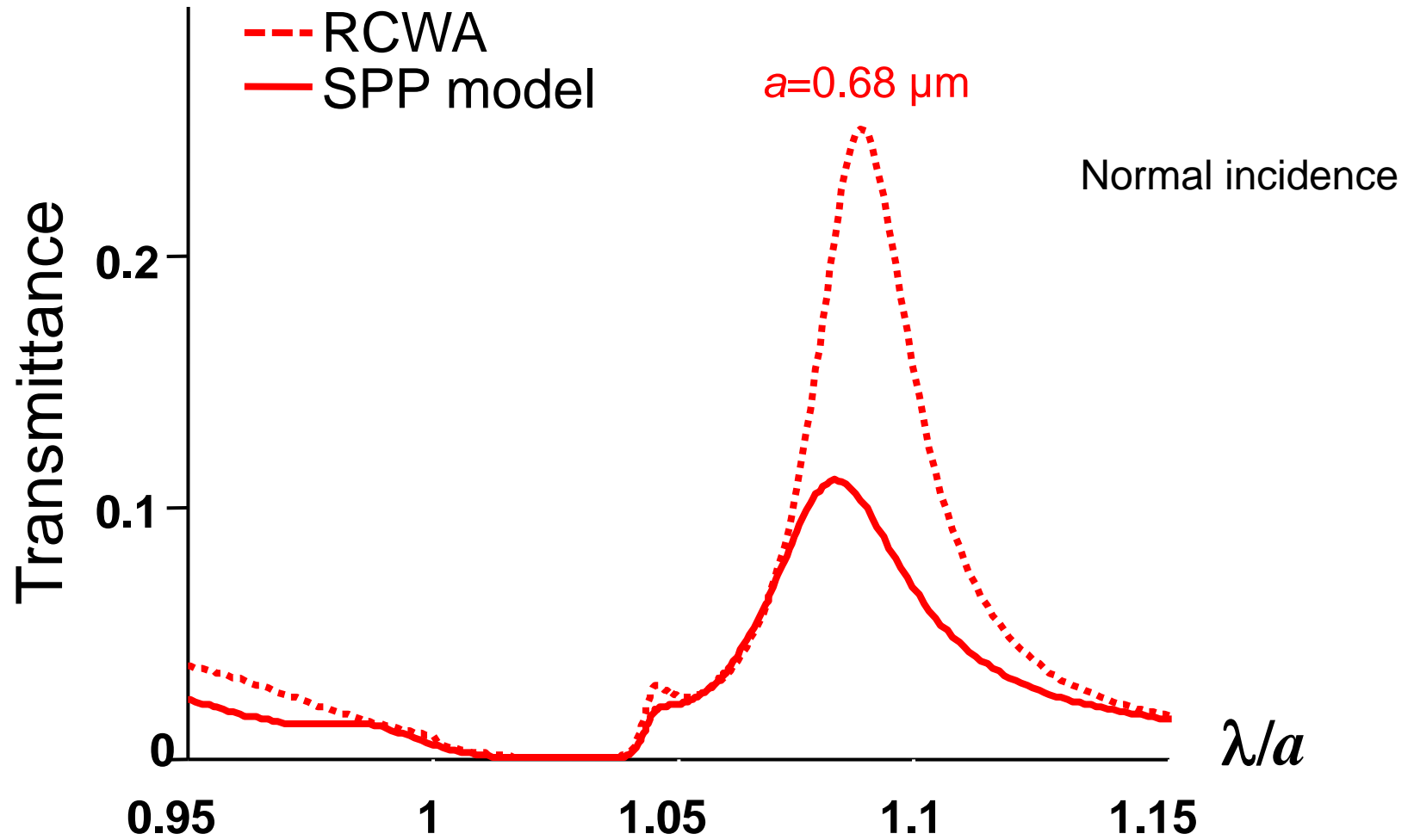
coupling of SPP to
free-space



$$T = \frac{\rho_1 \lambda_0^2}{\lambda^2} \left| 1 + \frac{\lambda_0^2}{\lambda^2} \frac{\alpha\beta}{\exp(ik_{SPP}a) - (\rho + \tau)} \right|$$

(for periodic arrays)

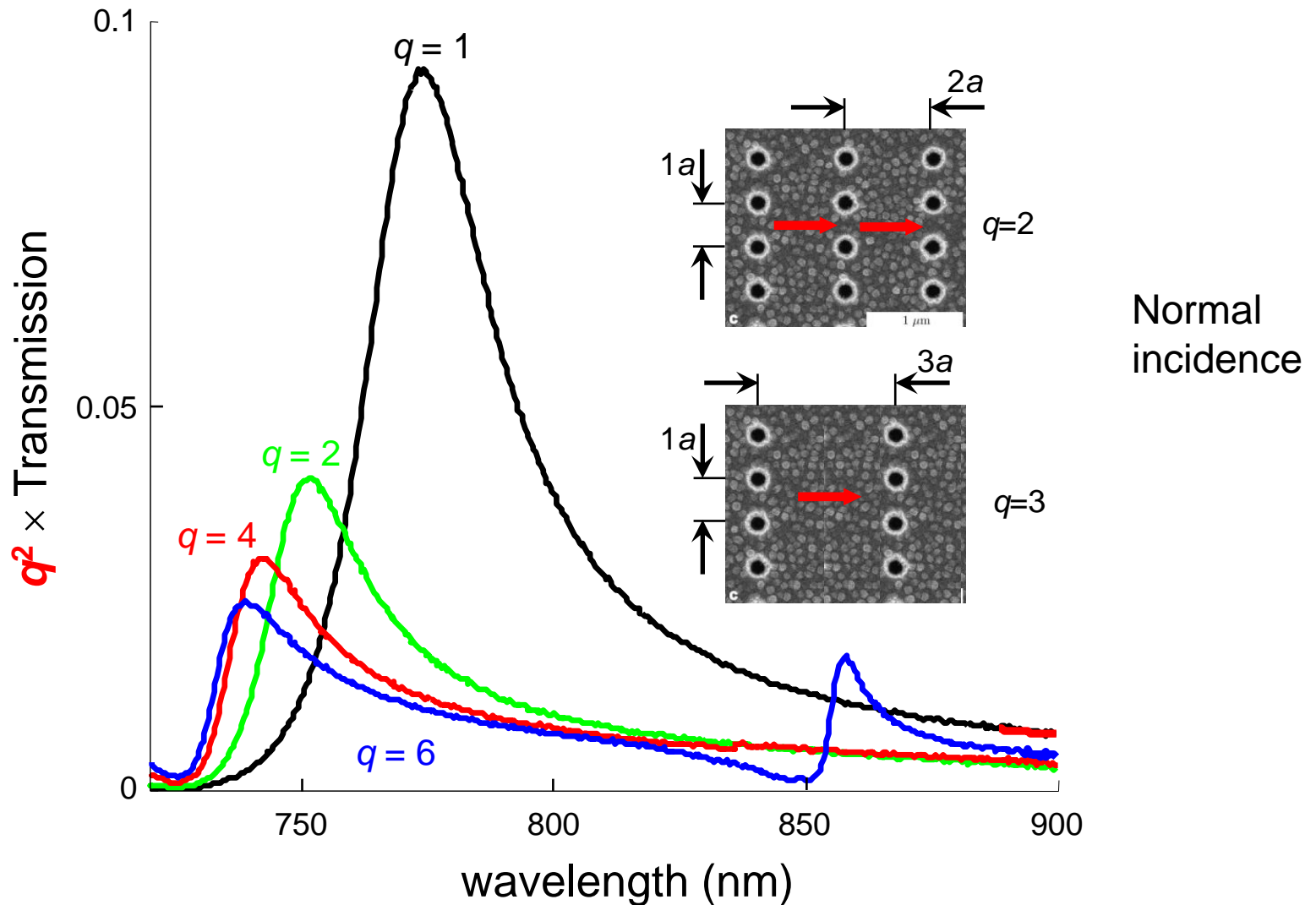
Actual SPP role in the EOT



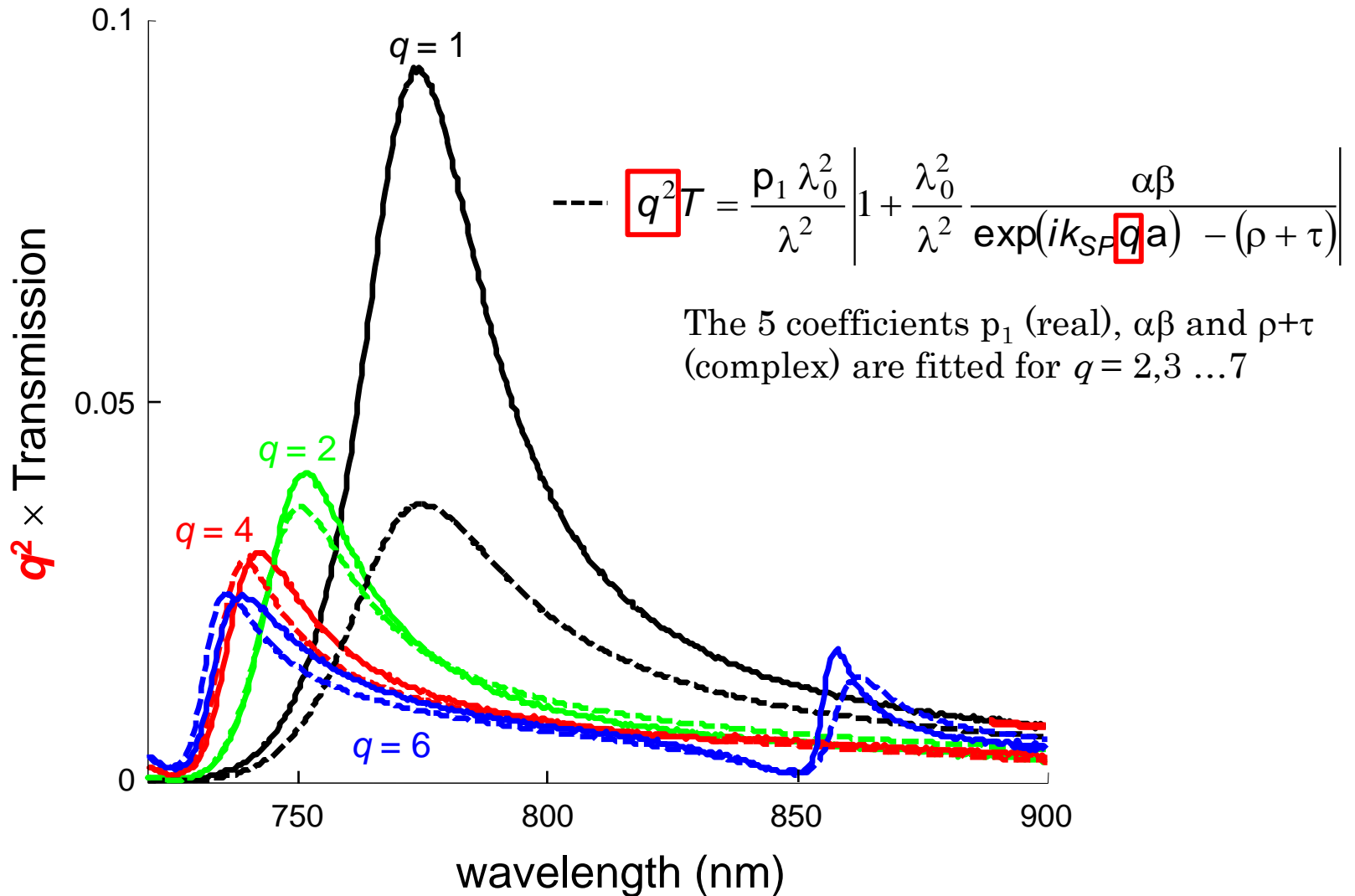
H. Liu and PL, Nature (London) **452**, 448 (2008).

Experimental evidence: F. van Beijnum et al., Nature **492**, 411 (Dec. 2012).

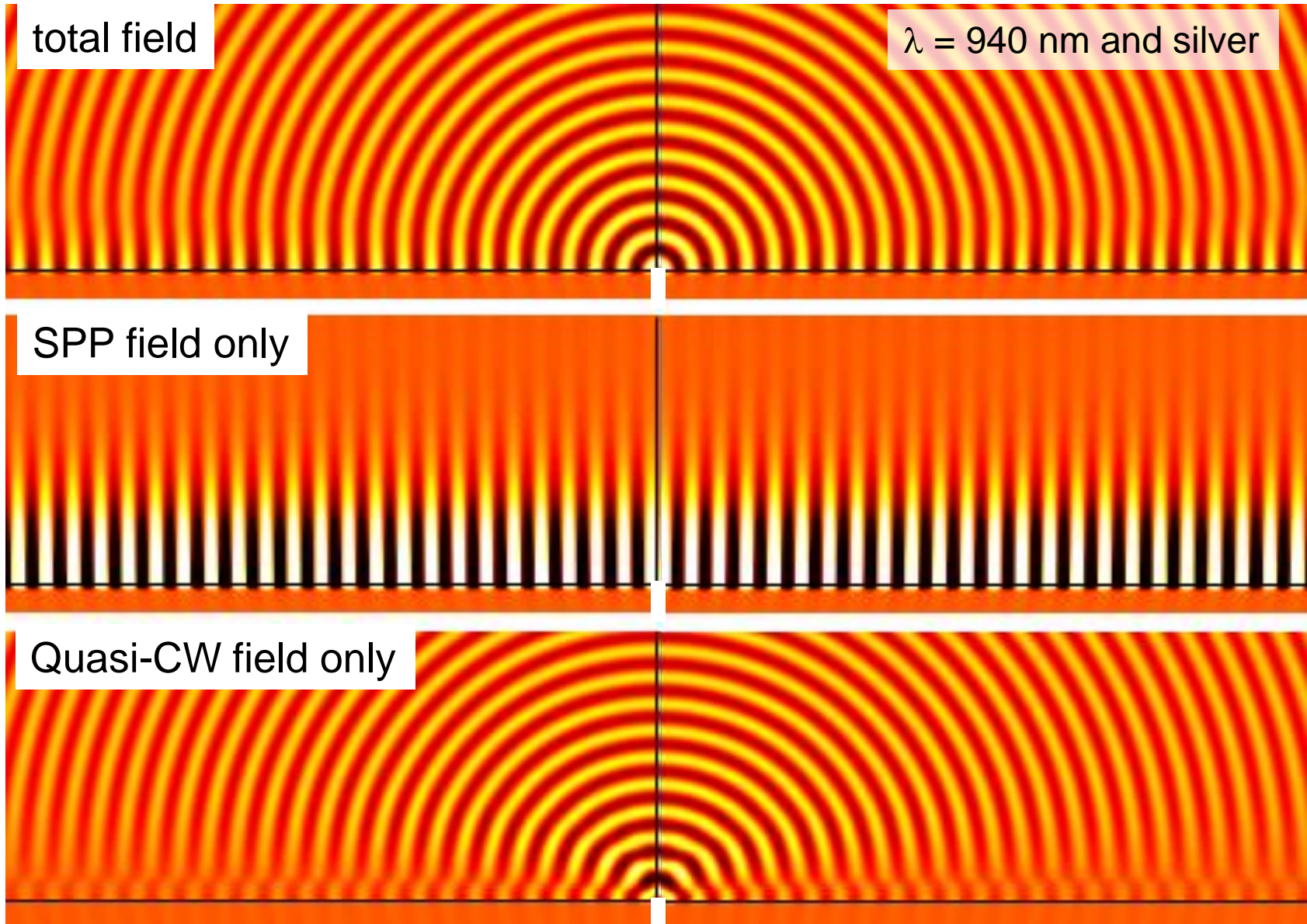
Direct experimental proof



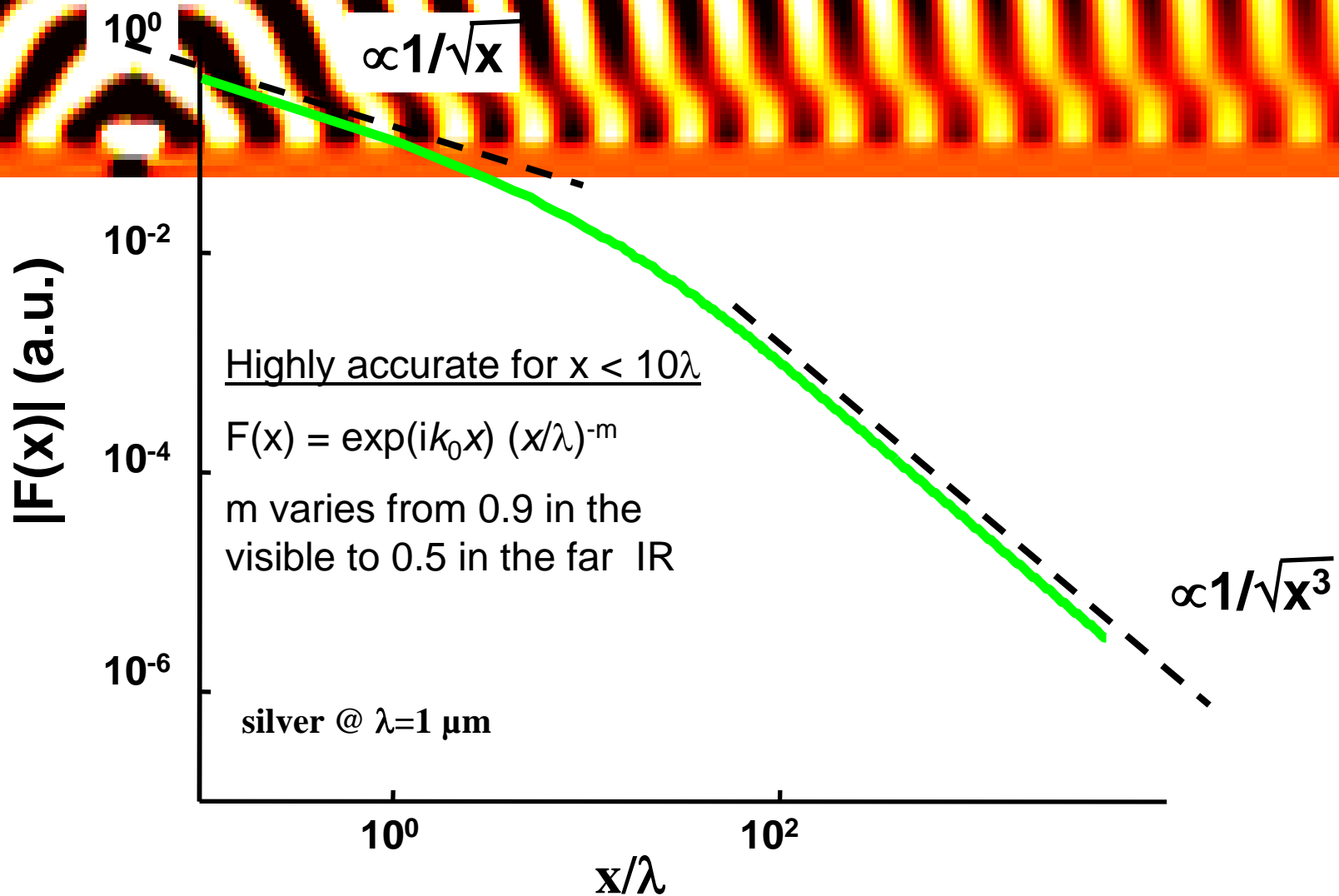
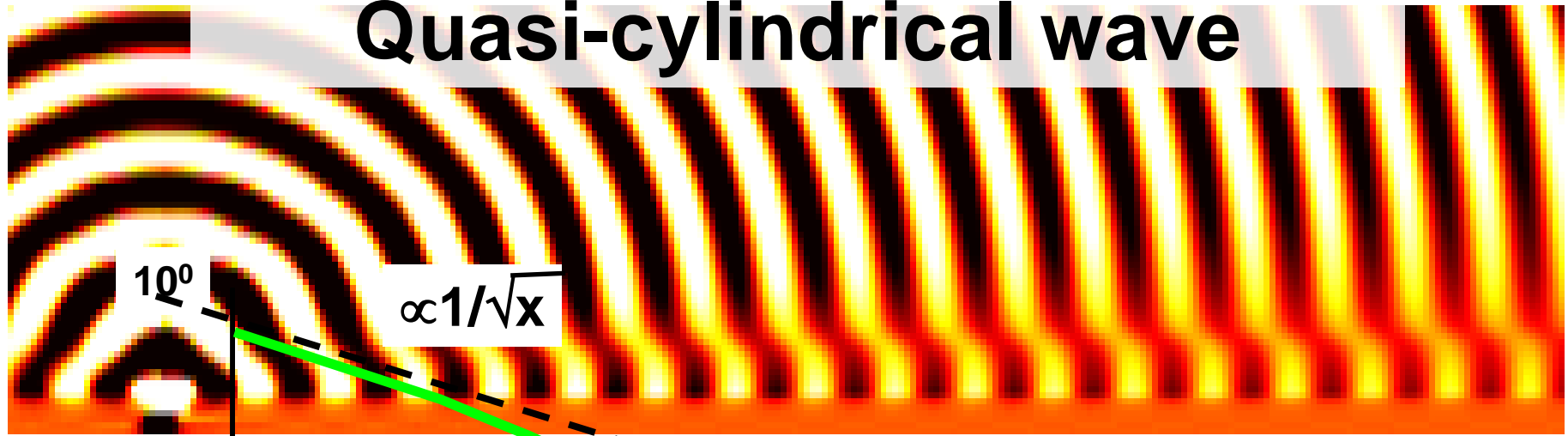
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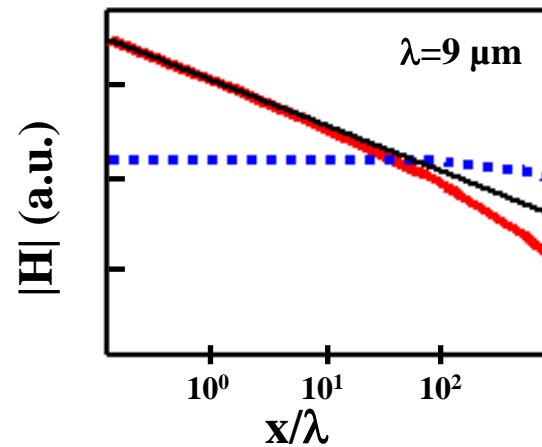
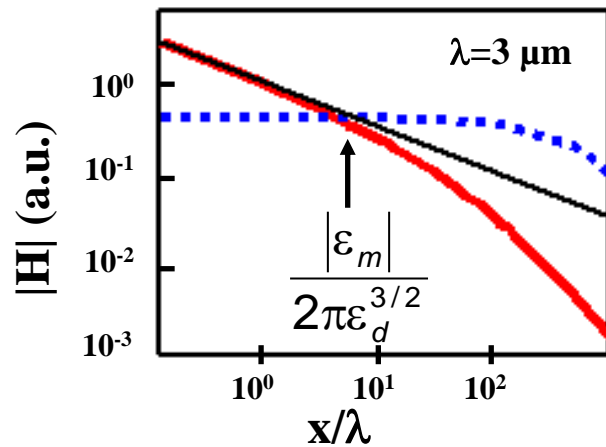
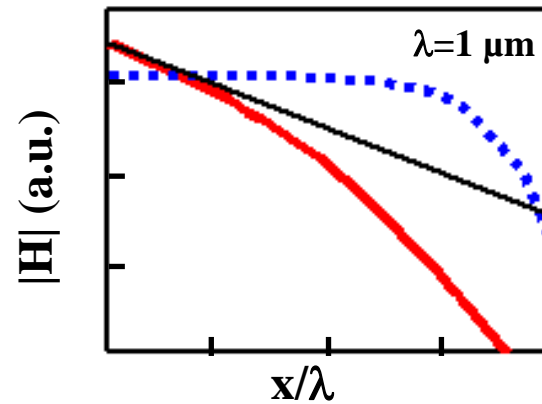
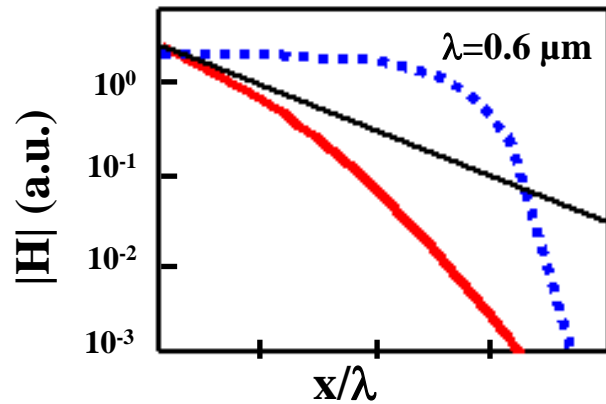
Quasi-cylindrical wave



Quasi-cylindrical wave



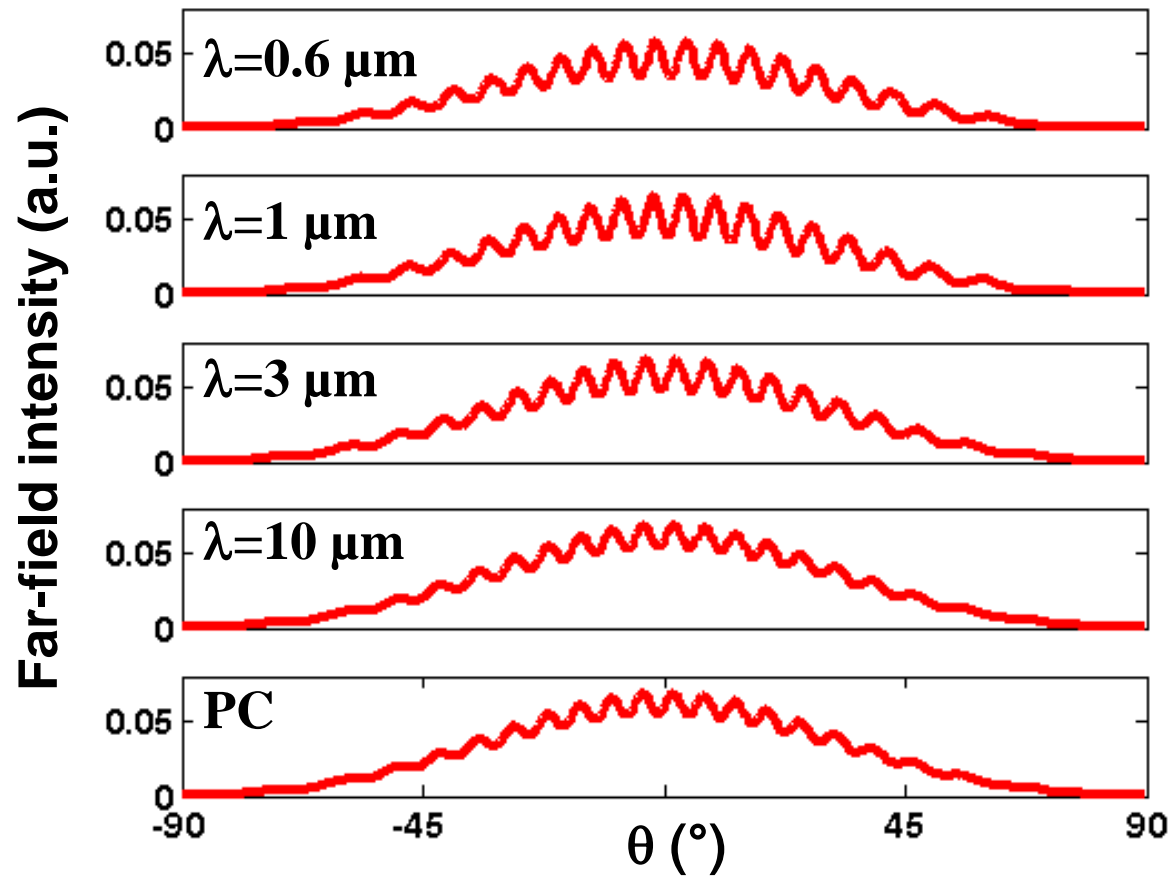
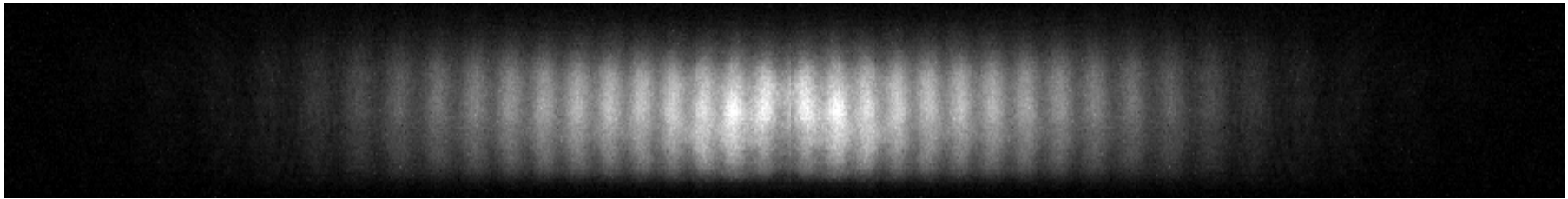
Frequency dependence (important)



⋯ H_{SP}
— H_{CW}
— $(x/\lambda)^{-1/2}$
 (perfect metal)

$H_{SP} \propto 1/|\epsilon_m|^{1/2}$
 $H_{CW} \propto \text{cte}$

(result for silver)



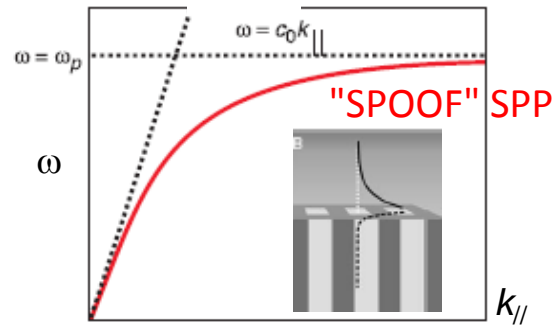
SPP mainly

Quasi-CW & SPP

Quasi-CW mainly

Quasi-CW only

Spoof=coherent interaction of holes
with quasi-cylindrical waves

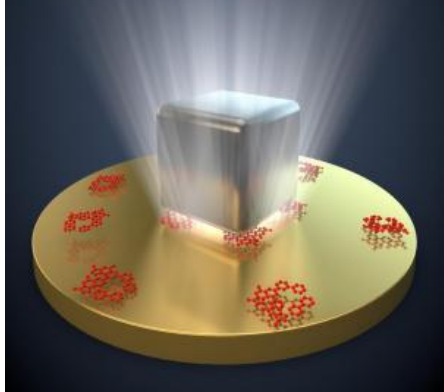
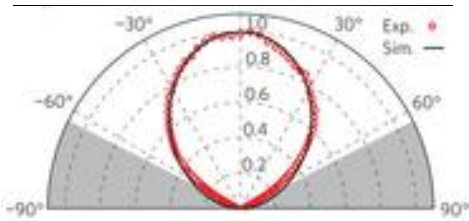


J. Pendry et al., Science **305**, 847 (2004).

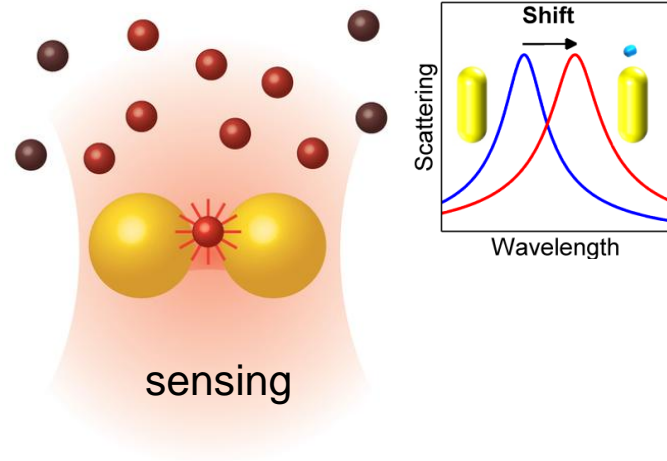
outline

- Field localization
- Delocalized surface plasmons on metal surfaces
- Wood anomaly (60 nm)
- **Localized plasmon**
- The « end » of the plasmon

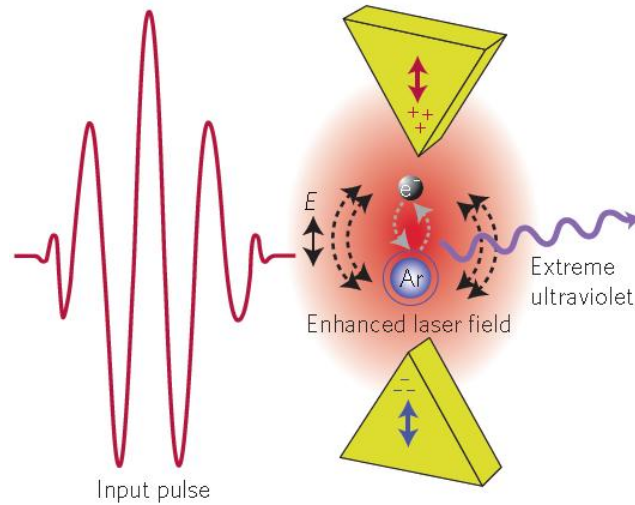
Metallic resonance



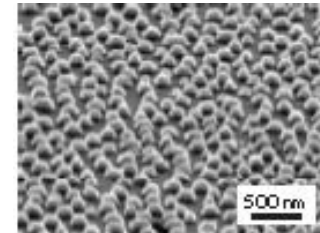
nanoantenna



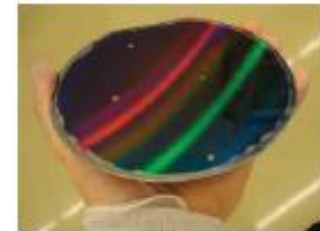
sensing



Frequency conversion



Solar cell



Metallic resonance

What is the resonance mode?

- how to define it « properly »?
- What is the mode volume?
- What are the limiting quantities for Q?

How efficiently can you excite it?

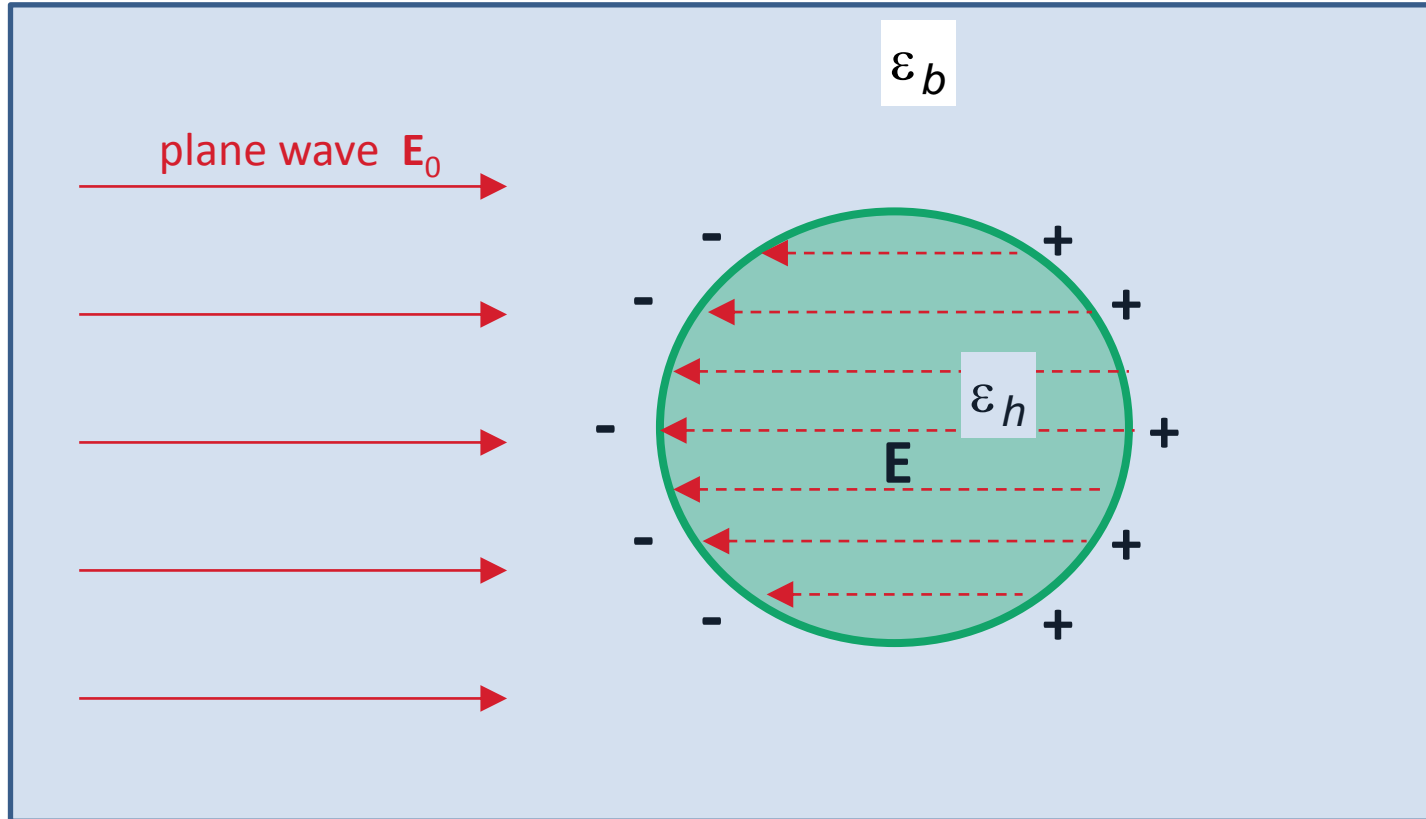
- from the near field? Purcell?
- from far field?

Application to sensing

- analytical formula of the resonance shift

Why tiny metallic NP resonate?

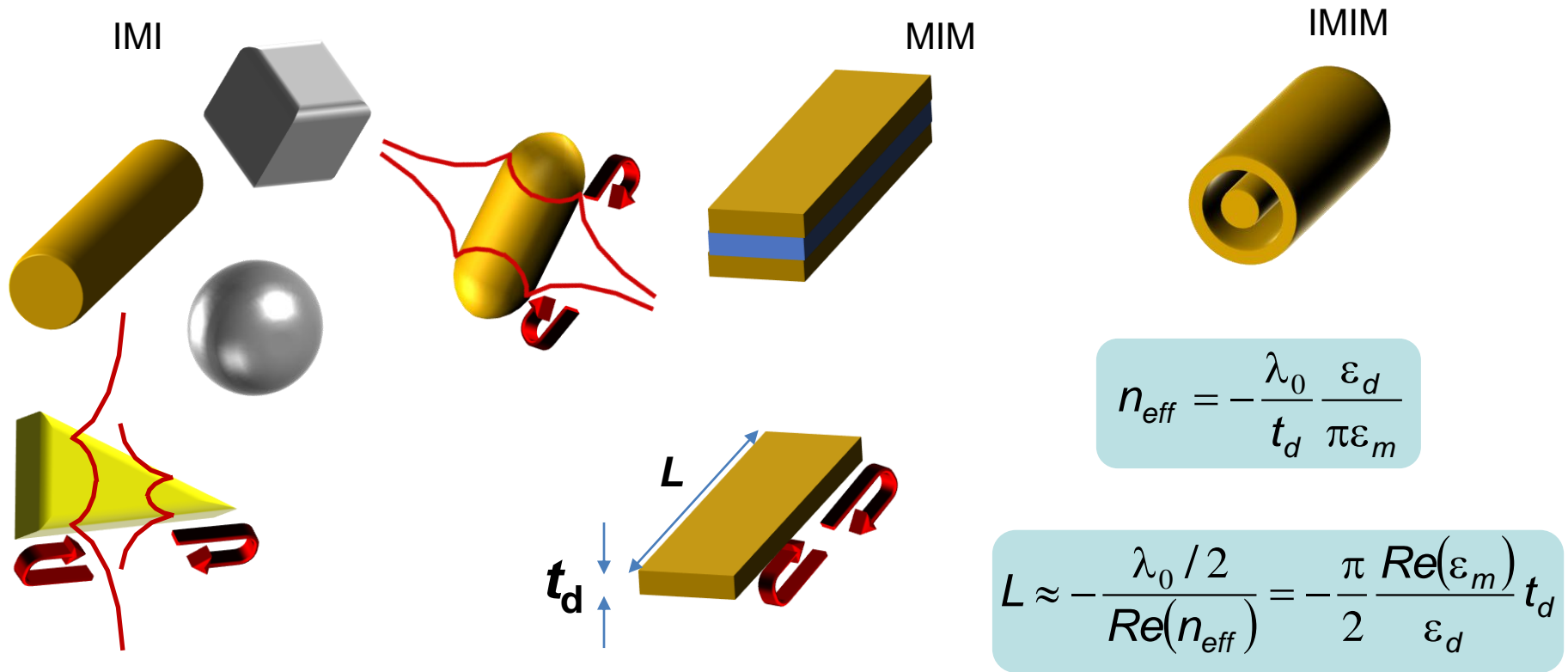
$$V \ll \lambda^3$$



$$\alpha = 3V \frac{\epsilon_h - \epsilon_b}{\epsilon_h + 2\epsilon_b} \longrightarrow \epsilon_h + 2\epsilon_b = 0$$

Resonance is achieved for a single fixed wavelength, such that $\epsilon_h + 2\epsilon_b = 0$

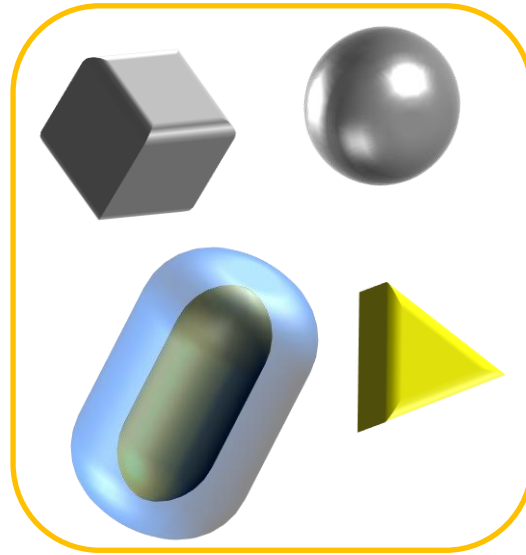
Why tiny metallic NP resonate?



Cross-section shrinks to zero, n_{eff} of the plasmonic mode diverges, and L shrinks!
 (The Fabry-Perot electric-dipole resonance mode scales down (no cutoff))

Resonance is achieved for any wavelength, just by scaling down dimensions.

Q factor at deep sub- λ scale



Quasi-static limit : Q factor of a localized plasmon resonance is determined **only** by ϵ_{metal} .

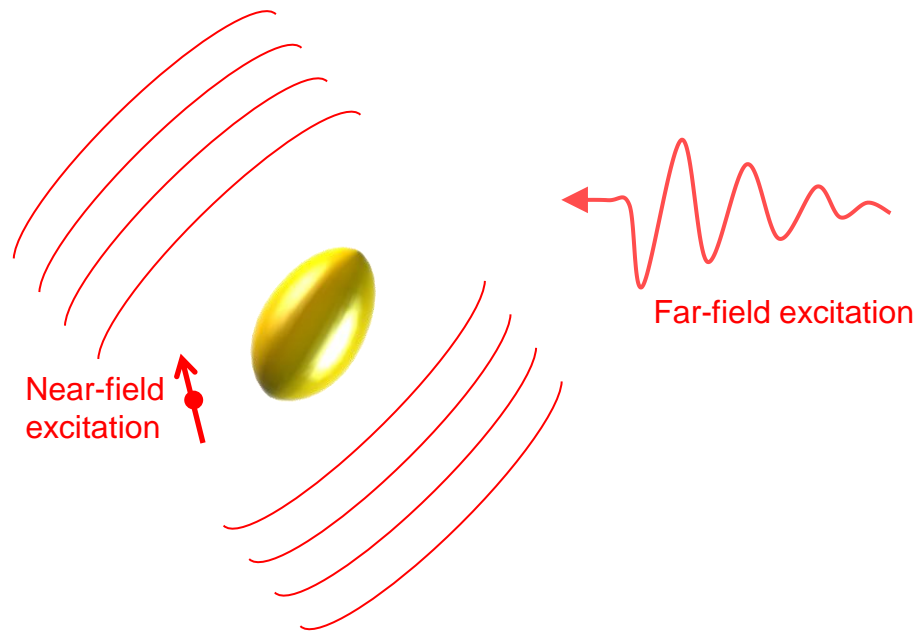
$$Q^S = \frac{\omega_0 \frac{\partial \text{Re}(\epsilon_{\text{metal}})}{\partial \omega}}{2 \text{Im}(\epsilon_{\text{metal}})} \quad (\text{Ag: } Q \sim 70 \text{ @ } \lambda=600\text{nm})$$

F. Wang et al., Phys. Rev. Lett. **97**, 206806 (2006).

J. Yang et al., Opt. Express 20, 16880-16891 (2012)

"Ultrasmall metal-insulator-metal nanoresonators: impact of slow-wave effects on the quality factor"

Excitation of metal resonance



What is a metallic resonance?



The quasi-normal modes $(\tilde{\mathbf{E}}_m, \tilde{\mathbf{H}}_m)$ are solutions of Maxwell's equations without source for a complex frequency $\tilde{\omega}_m$ ($Q = \text{Re}(\tilde{\omega})/2\text{Im}(\tilde{\omega})$)

$$\nabla \times \tilde{\mathbf{E}}_m = i \tilde{\omega}_m \mu_0 \tilde{\mathbf{H}}_m$$

$$\nabla \times \tilde{\mathbf{H}}_m = -i \tilde{\omega}_m \varepsilon(\mathbf{r}, \tilde{\omega}_m) \tilde{\mathbf{E}}_m$$

$$\tilde{\omega} \text{ complex} \Rightarrow \tilde{\mathbf{k}} \text{ complex} \Rightarrow \exp(i\tilde{\mathbf{k}}\mathbf{r}) \rightarrow \infty \text{ as } |\mathbf{r}| \rightarrow \infty$$

Excitation coefficient α

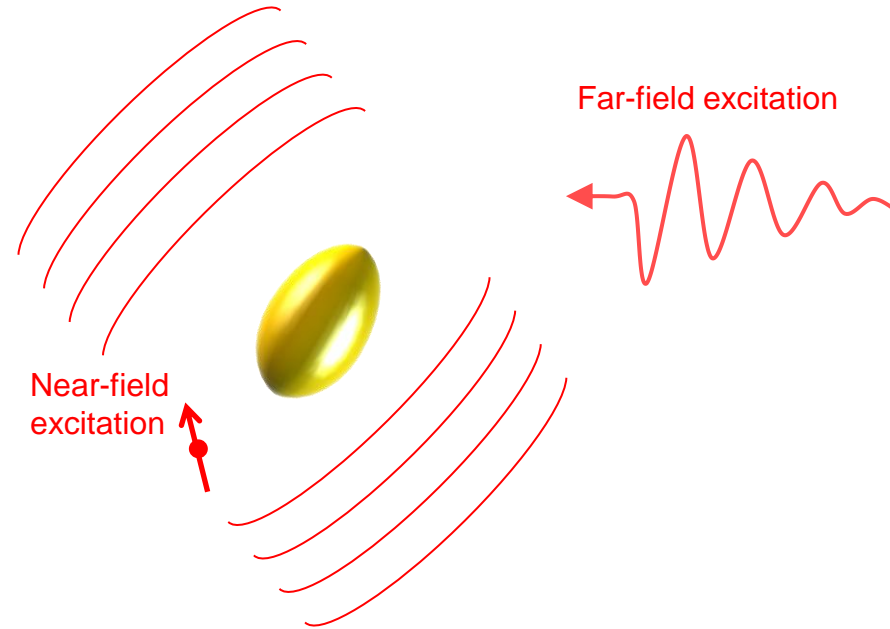
Only a single hypothesis : material is reciprocal

The scattered field is expanded in the QNM basis

$$\mathbf{E}(\mathbf{r}, \omega) = \mathbf{E}_{inc}(\mathbf{r}, \omega) + \sum_m \alpha_m(\omega) \tilde{\mathbf{E}}_m(\mathbf{r})$$

$$\alpha_m(\omega, \mathbf{r}) = -\frac{\omega}{\omega - \tilde{\omega}_m} \int d^3 \mathbf{r} \Delta\epsilon(\omega, \mathbf{r}) \mathbf{E}_{inc}(\omega, \mathbf{r}) \cdot \tilde{\mathbf{E}}_m(\mathbf{r})$$

$$\text{if } \iiint (\tilde{\mathbf{E}} \cdot \partial(\omega\epsilon)/\partial\omega \tilde{\mathbf{E}} - \tilde{\mathbf{H}} \cdot \partial(\omega\mu)/\partial\omega \tilde{\mathbf{H}}) d^3 \mathbf{r} = 1$$



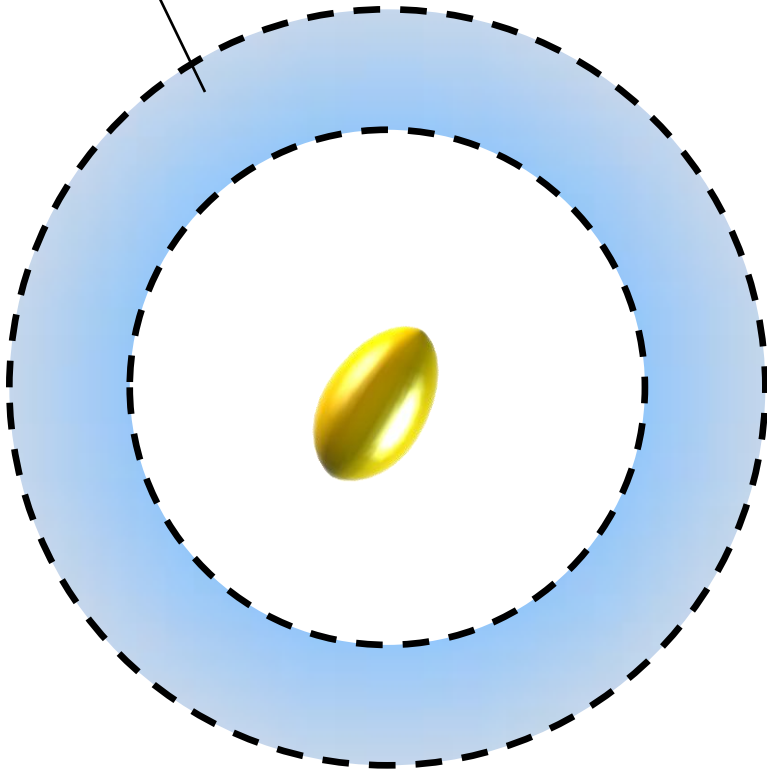
Very easy!

Energy of a dispersive material? yes but only when absorption is small. No energy consideration in the derivation.

The normalization issue

Complex coordinate transform (PML)
 $X = \alpha(1+im) x$
 $Y = \alpha(1+im) y$
 $Z = \alpha(1+im) z$

Analytical continuation in the complex plane with PMLs



remove the divergence problem for suitable m 's by transforming the exponentially diverging field into an exponentially damped field

$$\exp(i\tilde{\mathbf{k}}\mathbf{r})$$



∞

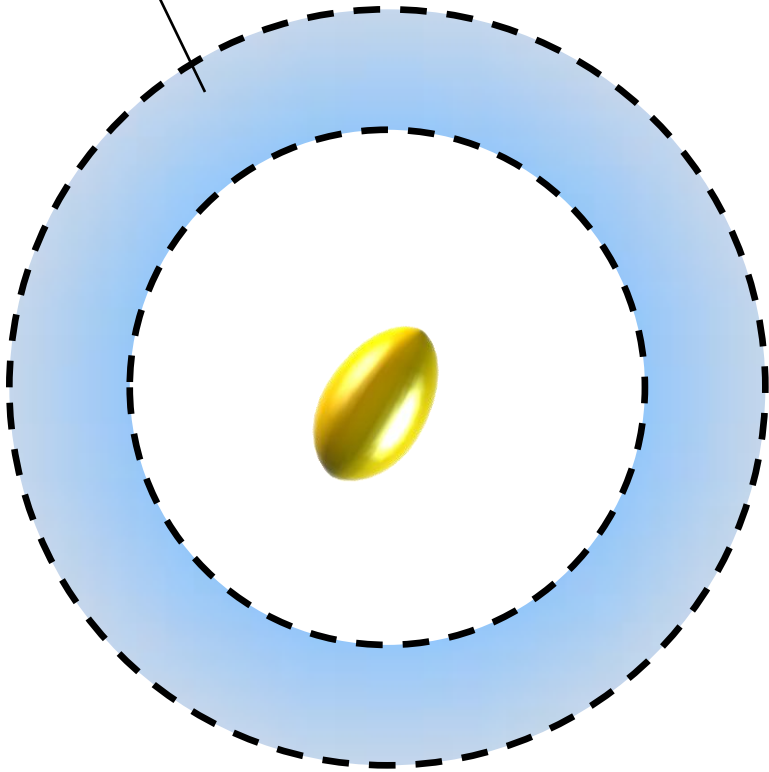
$$\exp(i\tilde{\mathbf{k}}(1+im)\mathbf{R})$$



0

The normalization issue

Complex coordinate transform (PML)
 $X = \alpha(1+im) x$
 $Y = \alpha(1+im) y$
 $Z = \alpha(1+im) z$



$\iiint \mathbf{E} \cdot \boldsymbol{\epsilon}(\omega) \mathbf{E} d^3\mathbf{r}$ is an invariant under space coordinate transforms

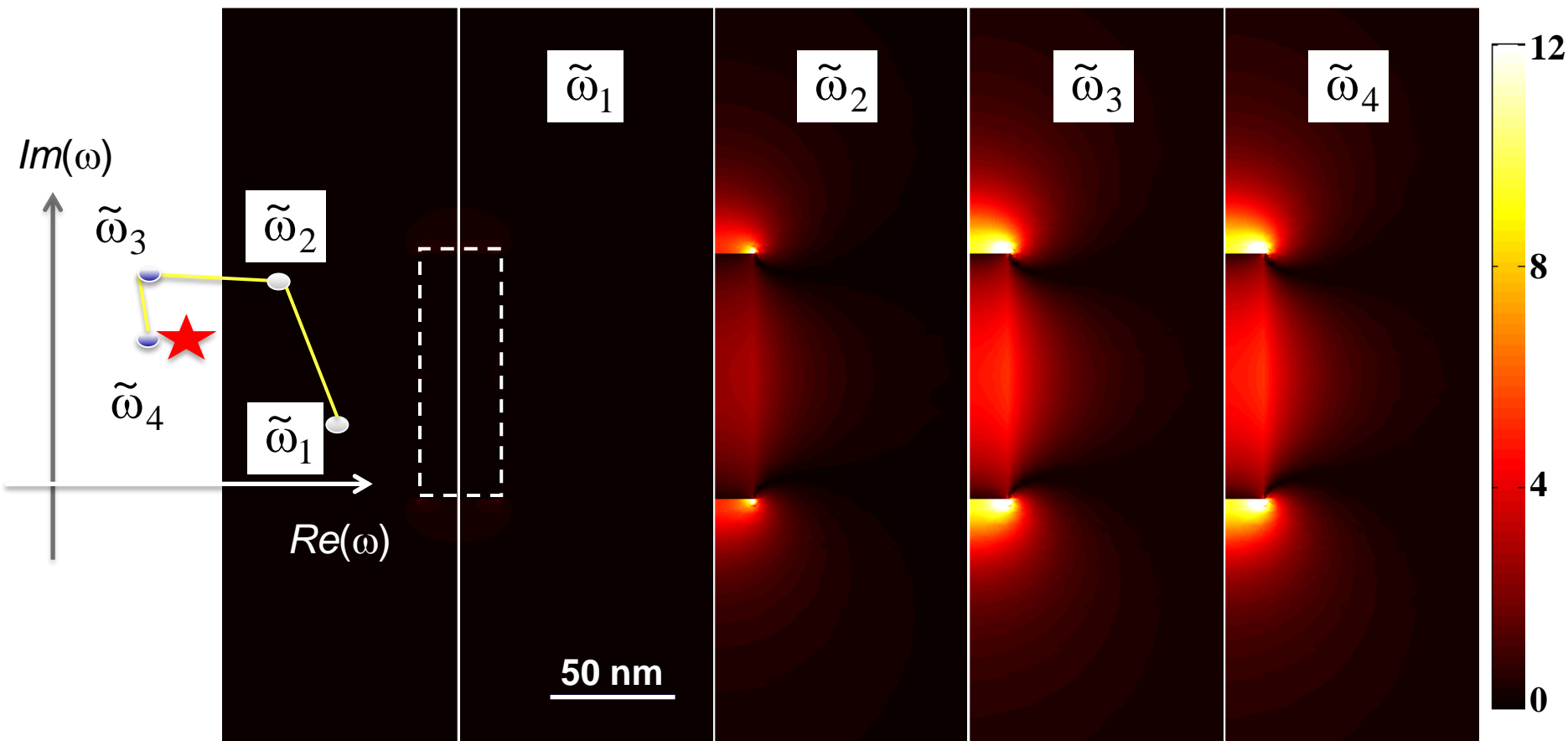


$$V_m = \iiint (\tilde{\mathbf{E}} \cdot \partial(\omega\boldsymbol{\epsilon})/\partial\omega \tilde{\mathbf{E}}) d^3\mathbf{r}$$

is invariant too and can be calculated with any PML, by computing the integral in real space and in the PML.

First (?) time the field in the PML is explicitly considered to evaluate a physical quantity.

Open-source software for resonance calculation

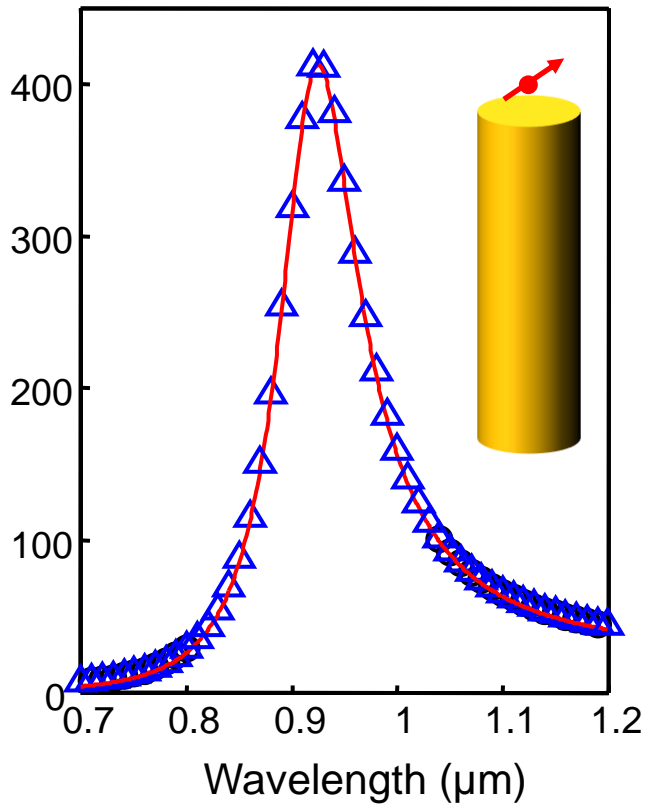


Q. Bai, et al., Opt. Express **21**, 27371 (2013).
Coll. Mathias Perrin/LOMA

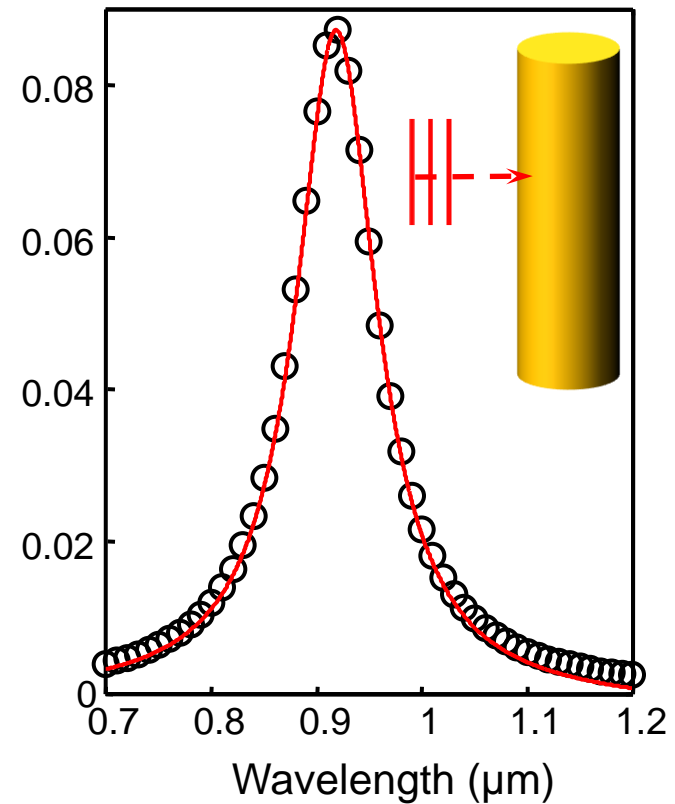
Freeware implemented with COMSOL multiphysics can be downloaded at
www.lp2n.institutoptique.fr

$$\alpha(\omega, \mathbf{r}) = -\frac{\omega}{\omega - \tilde{\omega}_m} \int d^3 \mathbf{r} \Delta\varepsilon(\omega, \mathbf{r}) \mathbf{E}_{\text{inc}}(\omega, \mathbf{r}) \cdot \tilde{\mathbf{E}}_m(\mathbf{r})$$

Purcell factor

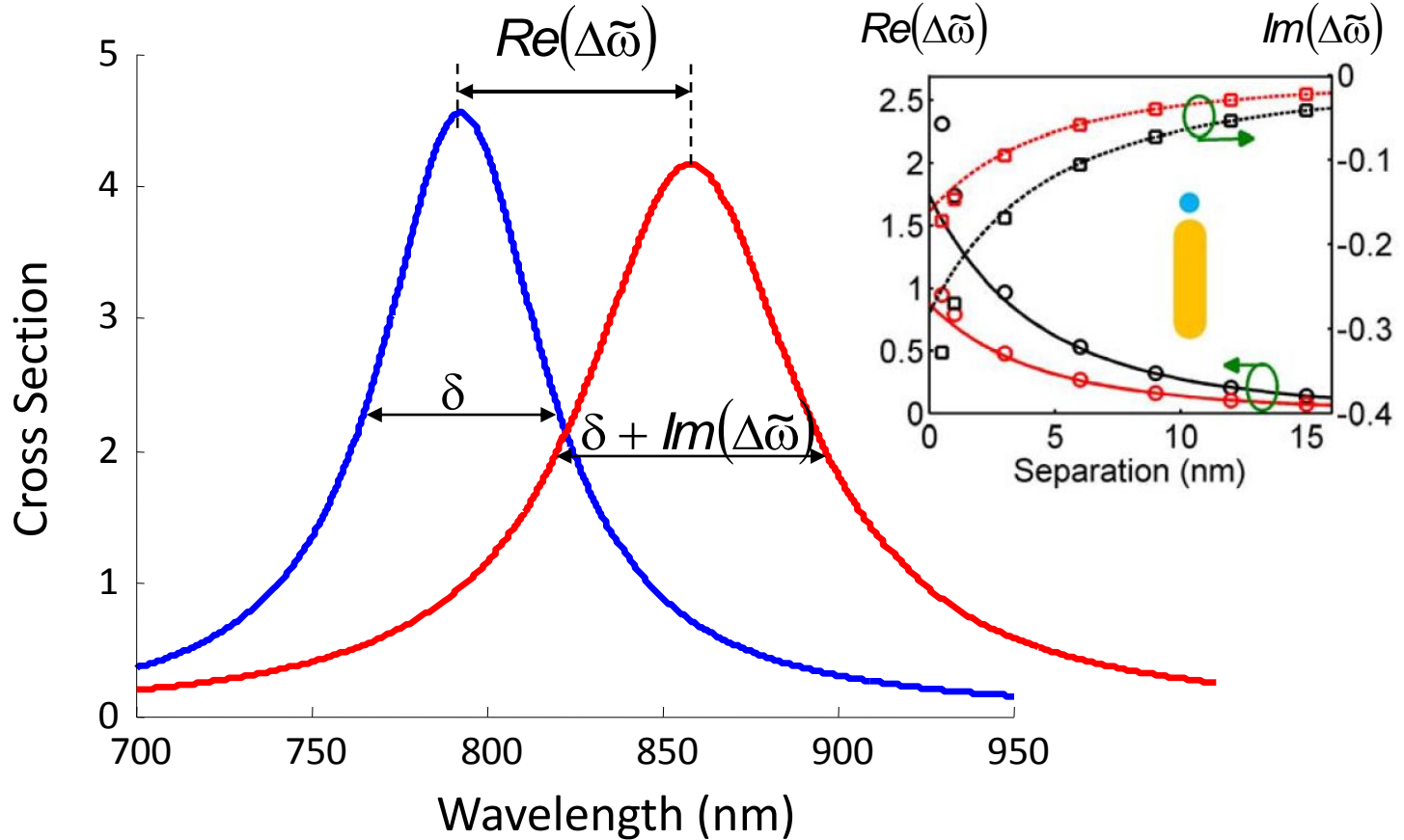
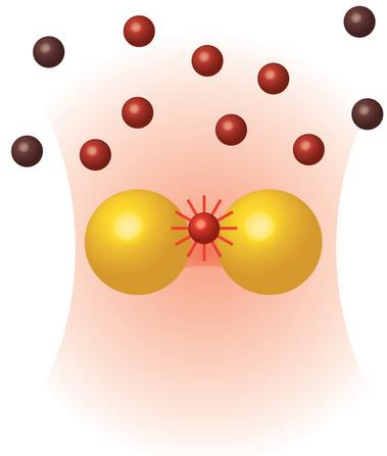


Extinction cross section (μm^2)



Application to sensing

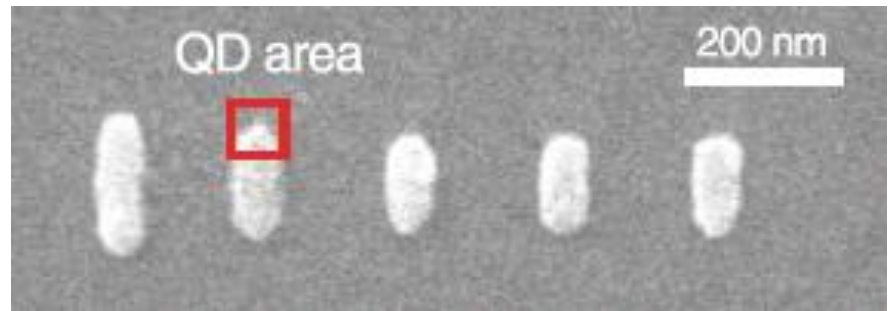
$$\Delta\tilde{\omega} = -\tilde{\omega} \iiint_{\text{perturb.}} \Delta\varepsilon(\mathbf{r}, \tilde{\omega}) \tilde{\mathbf{E}}'(\mathbf{r}) \tilde{\mathbf{E}}(\mathbf{r}) d^3\mathbf{r}$$



Nano-antenna



Yagi-Uda antenna

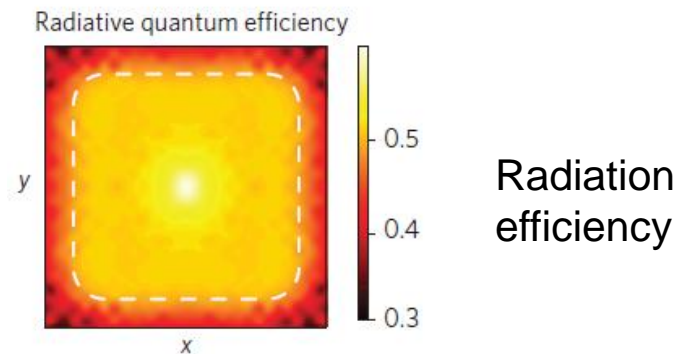
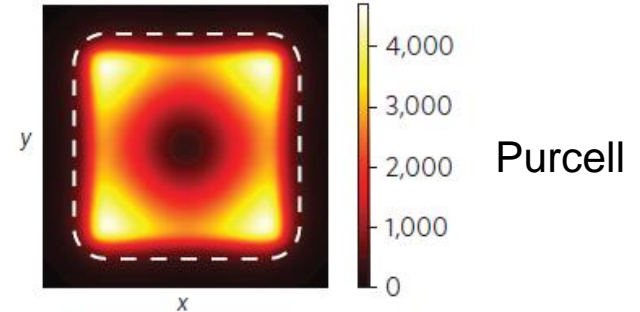
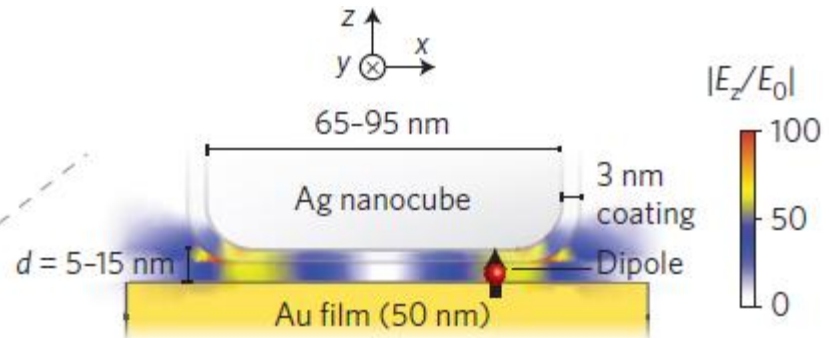
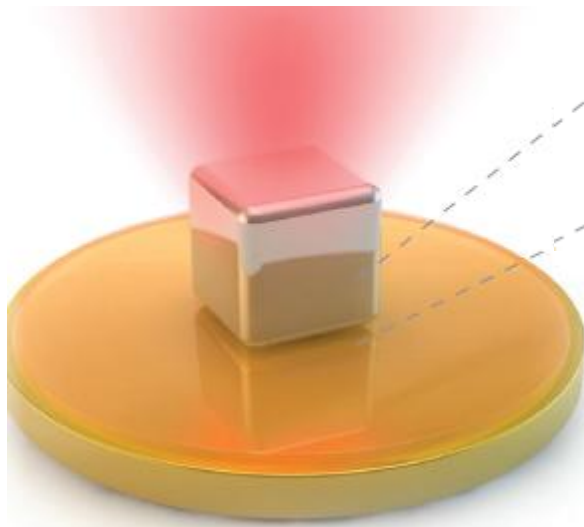
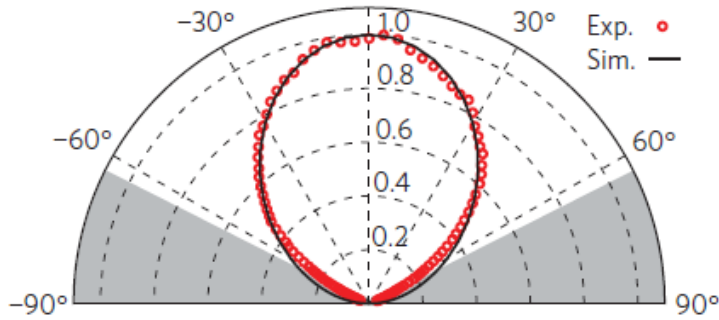


A. Curto et al., Science **329**, 930 (2010).

The quantum-dot luminescence is totally governed by the antenna

- radiation diagram
- Purcell factor

Nano-antenna



G.M. Akselrod et al., Nat. Photon. DOI:
10.1038/NPHOTON.2014.228

“Probing the mechanisms of large Purcell enhancement in plasmonic nanoantennas”

Classical Purcell formula

$$\frac{\Gamma}{\Gamma_0} = F \frac{\omega_0^2}{\omega^2} \frac{\omega_0^2}{\omega_0^2 + 4Q^2(\omega - \omega_0)^2}$$

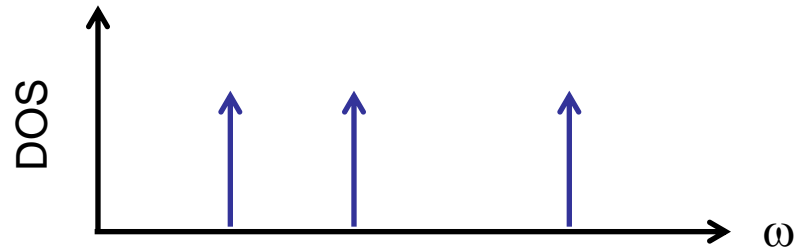
Classical Lorentzian shape

$$F = \frac{3}{4\pi^2} \left(\frac{\lambda}{n}\right)^3 \frac{Q}{V_M} \text{ with } V_M = \frac{\iiint \varepsilon(\mathbf{r}) |\tilde{\mathbf{E}}(\mathbf{r})|^2 d^3\mathbf{r}}{\max(\varepsilon(\mathbf{r}) |\tilde{\mathbf{E}}(\mathbf{r})|^2)}$$

Only valid for large Q
(error scales as 1/Q as
Q → ∞)

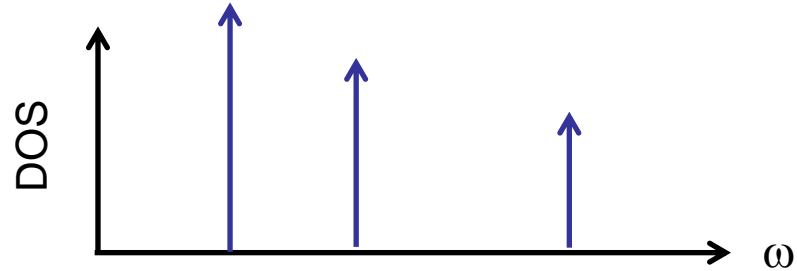
modal-expansion of the LDOS

$$\text{DOS} = \sum_n \delta(\omega - \tilde{\omega}_n)$$



modal-expansion of the LDOS

$$\text{DOS} = \sum_n \delta(\omega - \tilde{\omega}_n)$$

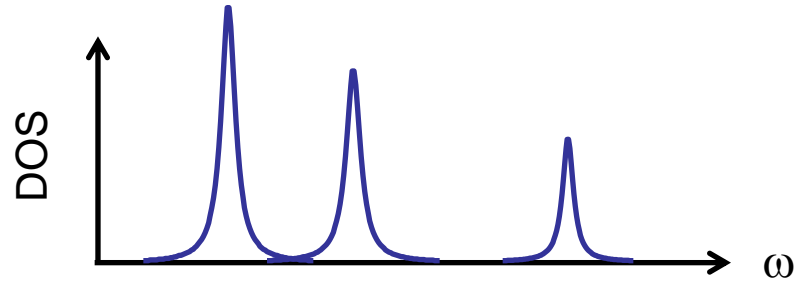


$$\text{LDOS}(\mathbf{r}, \omega) = \sum_n \delta(\omega - \tilde{\omega}_n) \varepsilon(\mathbf{r}) |\tilde{\mathbf{E}}_n(\mathbf{r})|^2$$

$$\langle \tilde{\mathbf{E}}_n, \tilde{\mathbf{E}}_n \rangle = \int_V \varepsilon(\mathbf{r}) |\tilde{\mathbf{E}}_n(\mathbf{r})|^2 d^3\mathbf{r} = 1$$

modal-expansion of the LDOS

$$\text{DOS} = \sum_n \delta(\omega - \tilde{\omega}_n)$$

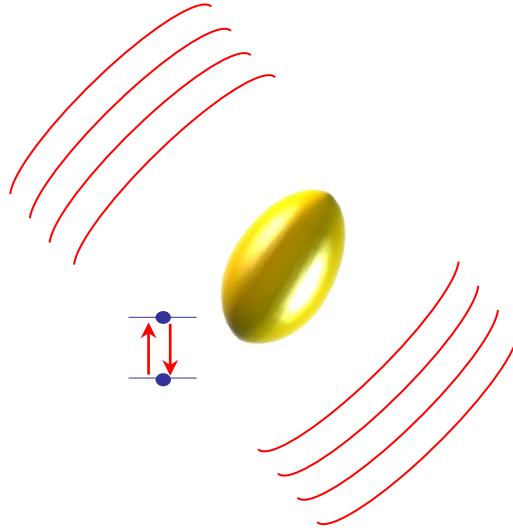


$$\text{LDOS}(\mathbf{r}, \omega) = \sum_n \delta(\omega - \tilde{\omega}_n) \varepsilon(\mathbf{r}) |\tilde{\mathbf{E}}_n(\mathbf{r})|^2$$

$$\langle \tilde{\mathbf{E}}_n, \tilde{\mathbf{E}}_n \rangle = \int_V \varepsilon(\mathbf{r}) |\tilde{\mathbf{E}}_n(\mathbf{r})|^2 d^3\mathbf{r} = 1$$

$$\text{LDOS}(\mathbf{r}, \omega) = \sum_n \frac{\gamma_n}{\pi} \frac{1}{(\omega - \tilde{\omega}_n)^2 + \gamma_n^2} \varepsilon(\mathbf{r}) |\tilde{\mathbf{E}}_n(\mathbf{r})|^2$$

Revisiting the Purcell formula



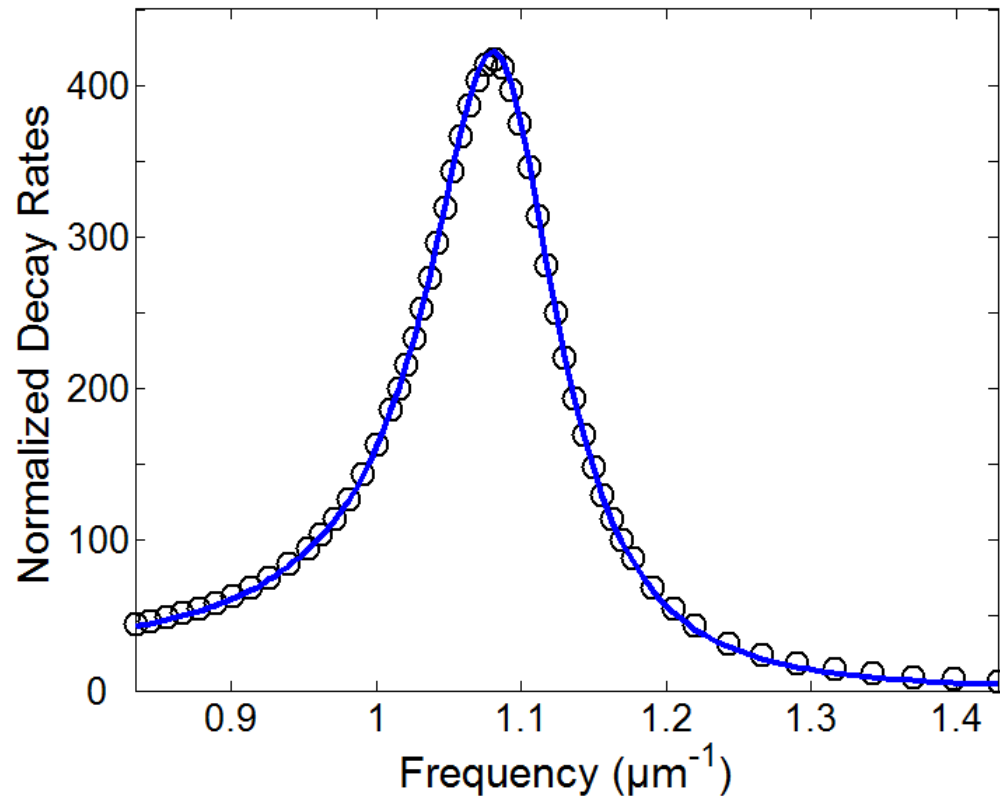
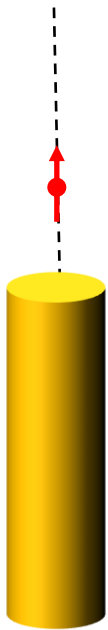
$$\frac{\Gamma}{\Gamma_0} = F \frac{\omega_0^2}{\omega^2} \frac{\omega_0^2}{\omega_0^2 + 4Q^2(\omega - \omega_0)^2} \left[1 + 2Q \frac{\omega - \omega_0}{\omega_0} \frac{\text{Im}(V_M)}{\text{Re}(V_M)} \right]$$

$$F = \frac{3}{4\pi^2} \left(\frac{\lambda}{n} \right)^3 \text{Re} \left(\frac{Q}{V_M} \right) \text{ with } V_M = \frac{\iiint (\tilde{\mathbf{E}} \cdot \partial(\omega\boldsymbol{\epsilon})/\partial\omega \tilde{\mathbf{E}}) d^3\mathbf{r}}{2\varepsilon_0 n^2 (\tilde{\mathbf{E}}(\mathbf{r}_0) \cdot \mathbf{u})^2}$$

Derivation based on *reciprocity arguments*, see C. Sauvan et al., PRL **110**, 237401 (2013) & Q. Bai et al., Opt. Express **21**, 27371 (2013).

Non-Lorentzian response with metallic resonance

Circle: Green-tensor calculation (decay in all modes)
Blue line: revised Purcell formula (with a single mode)

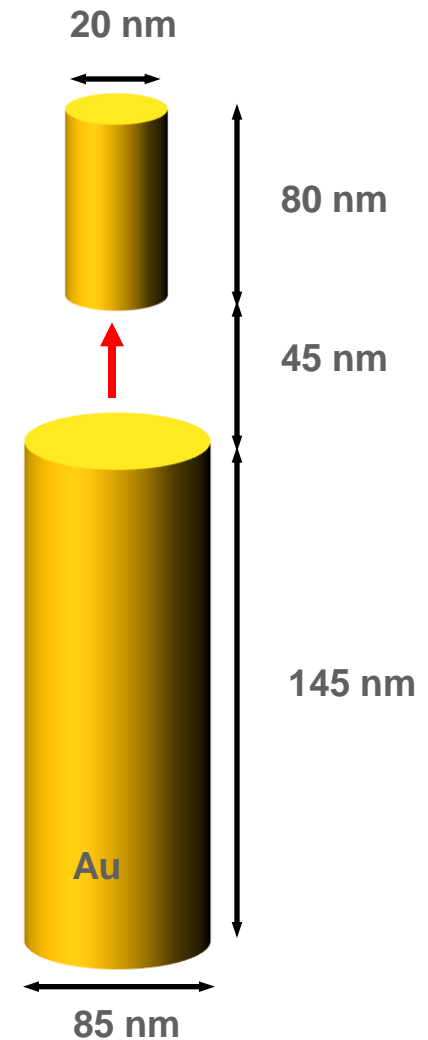
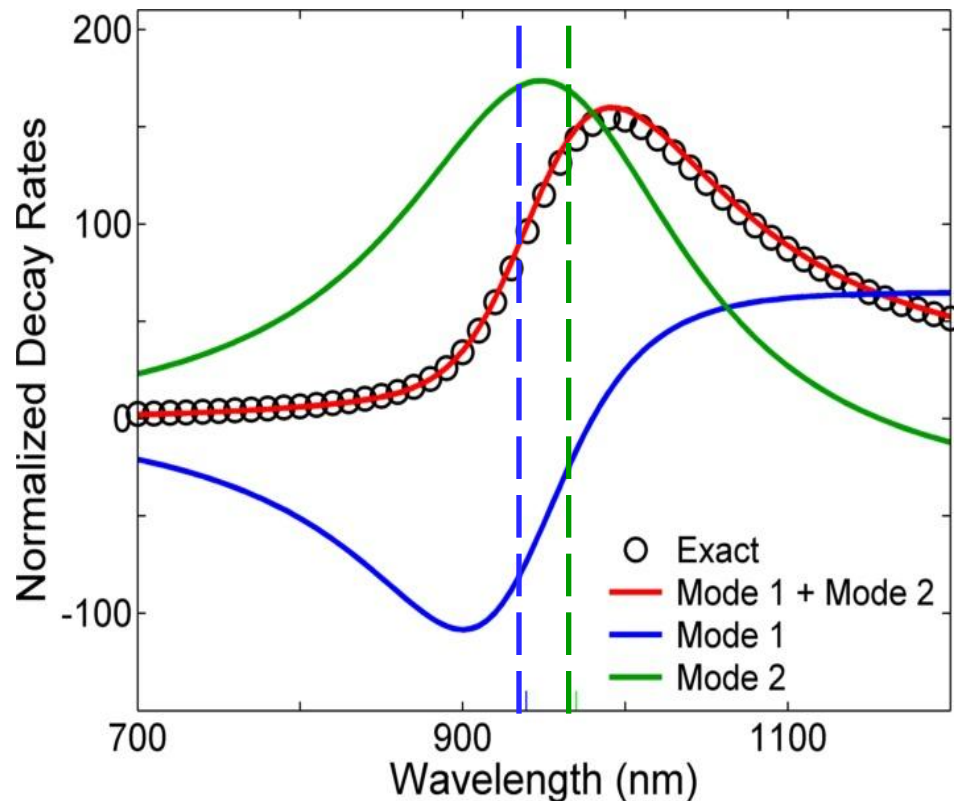


Multi-resonance case

the contribution of a quasi-normal mode to the total power radiated by a source may be detrimental (it may reduce the decay rate), even when the frequencies of the source and the mode are matched.

$$V_1 = (-3 - 7i) \frac{\lambda^3}{10,000}$$

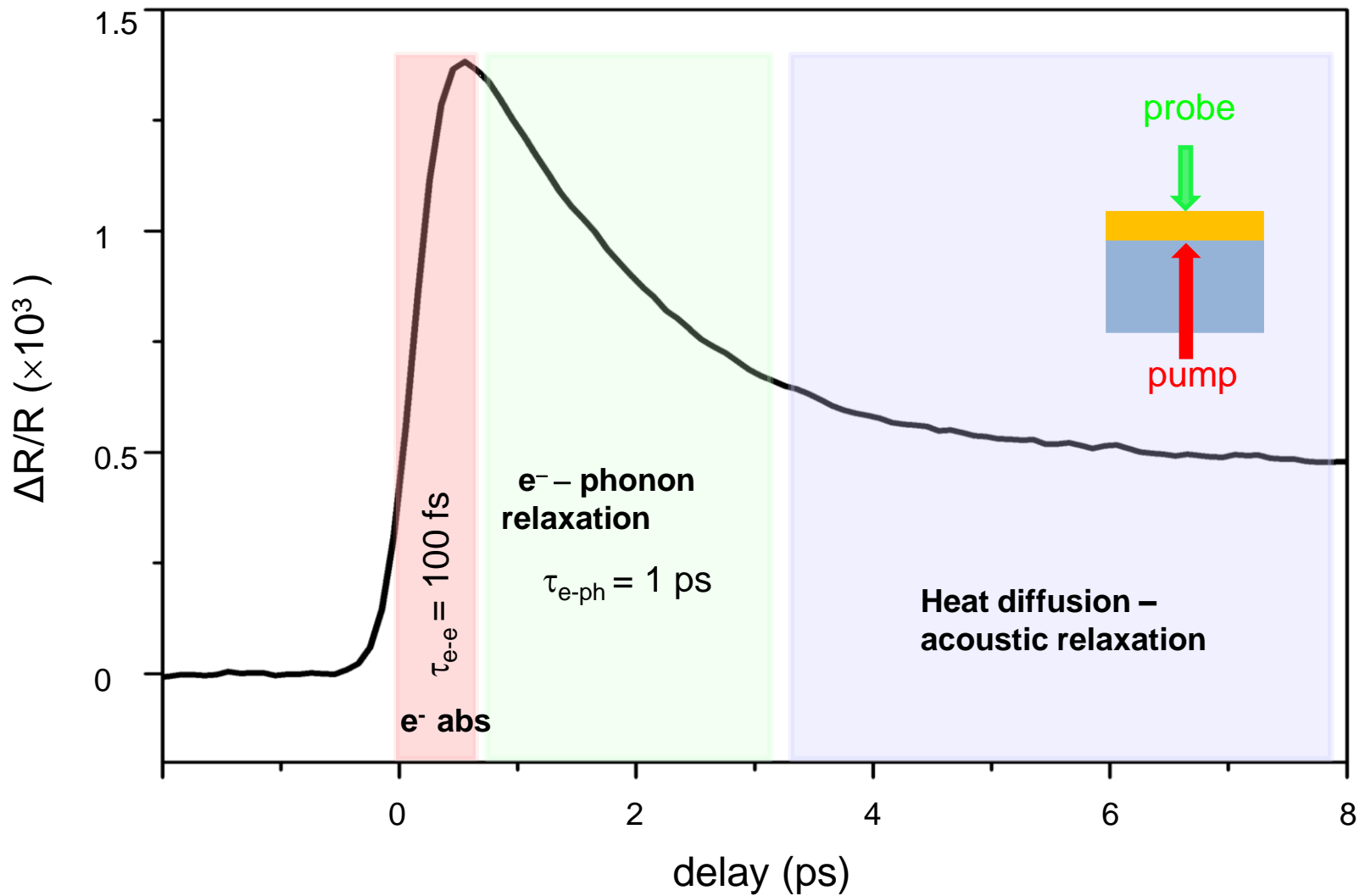
$$V_2 = (4 + 3i) \frac{\lambda^3}{10,000}$$



outline

- Field localization
- Delocalized surface plasmons on metal surfaces
- Wood anomaly
- Localized plasmon (1H20)
- **The « end » of the plasmon**

The plasmon decay and then what?



Hot electrons=hot topic

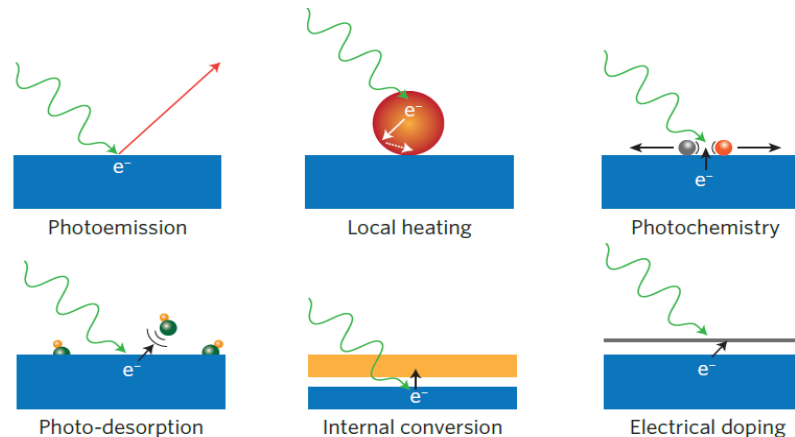
SPP7 2015: first apparition of hot electrons in the main topic list of the SPP conference series

TOPICS
1. Biosensors for Health Care
2. Devices for Telecommunications
3. Electron-Plasmon Interactions
4. Light Concentration for Solar Energy
5. Loss Compensation and Plasmon Lasing
6. Near-Field Instrumentation
7. Nonlocality

First review paper appeared in Jan. 2015



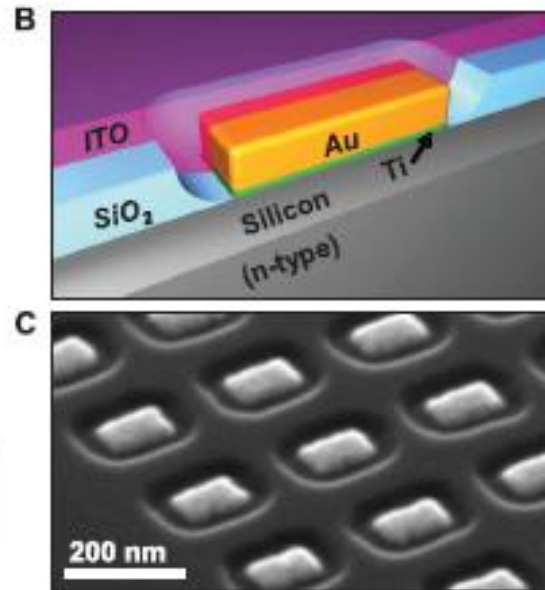
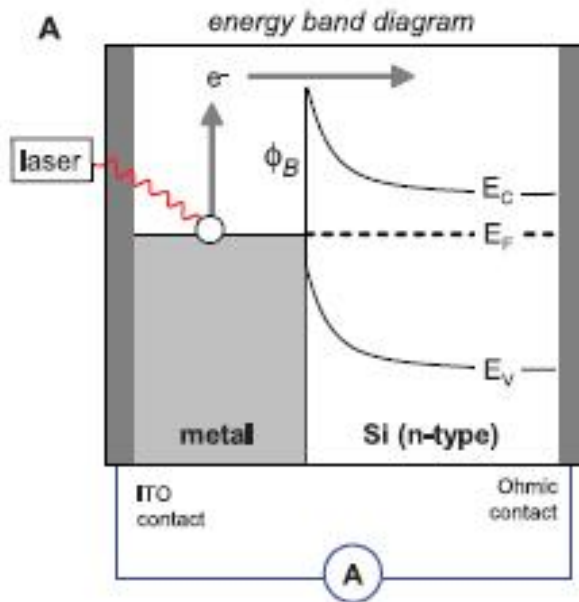
Plasmon-induced hot carrier science and technology



Many applications

Plasmon induced hot carriers

- photodetectors with spectral responses circumventing band gap limitations
- chemical catalysis close to metal surfaces



The end

Même quand il meurt, le plasmon renaît de ses cendres.
Le plasmon est éternel, « offrir donc un plasmon »

