Nano-Optics

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Table of Contents

Basics

Constitutive relations

$$\mathbf{D} = \varepsilon_o \varepsilon \mathbf{E} \qquad (\mathbf{P} = \varepsilon_o \chi_e \mathbf{E}) \,,$$
$$\mathbf{B} = \mu_o \mu \mathbf{H} \qquad (\mathbf{M} = \chi_m \mathbf{H}) \,,$$
$$\mathbf{j}_c = \sigma \mathbf{E} \,.$$

describe the role of matter!

Maxwell:

$$\nabla \times \mathbf{E}(\mathbf{r}) = i\omega \mathbf{B}(\mathbf{r}) \,,$$
$$\nabla \times \mathbf{H}(\mathbf{r}) = -i\omega \mathbf{D}(\mathbf{r}) + \mathbf{j}(\mathbf{r}) \,,$$
$$\nabla \cdot \mathbf{D}(\mathbf{r}) = \rho(\mathbf{r}) \,,$$
$$\nabla \cdot \mathbf{B}(\mathbf{r}) = 0 \,,$$

$\Big| \; \mu^{-1} \; \Big| \; \nabla \times$

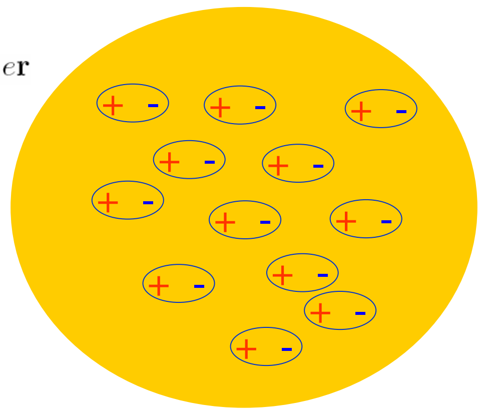
$\mathcal{E}$

$$\nabla \times \mu^{-1} \nabla \times \mathbf{E} - \frac{\omega^2}{c^2} [\varepsilon + i\sigma/(\omega\varepsilon_o)] \, \mathbf{E} = i\omega\mu_o \mathbf{j}_s$$

Dielectric function is a complex quantity

Dielectric function of metals

$$\boldsymbol{\mu} = e\mathbf{r}$$



$$\mathbf{P} = n\boldsymbol{\mu}$$

$$\mathbf{D}(\mathbf{r},t) = \varepsilon_0 \mathbf{E}(\mathbf{r},t) + \mathbf{P}(\mathbf{r},t)$$

$$\mathbf{D} = \varepsilon_0 \varepsilon \mathbf{E}$$

$$\varepsilon = 1 + \frac{|\mathbf{P}|}{\varepsilon_0 |\mathbf{E}|}$$

Drude-Sommerfeld theory

Free electron contribution

$$m_e \frac{\partial^2 \mathbf{r}}{\partial t^2} + m_e \Gamma \frac{\partial \mathbf{r}}{\partial t} = e \mathbf{E}_0 e^{-i\omega t}$$

$$\mathbf{r}(t) = \mathbf{r}_0 e^{-i\omega t} \quad \Rightarrow \quad \boxed{\varepsilon_{\text{Drude}}(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\Gamma\omega}}$$

$$\omega_p = \sqrt{ne^2/(m_e \varepsilon_0)}$$

$$\varepsilon_{\text{Drude}}(\omega) = \boxed{1 - \frac{\omega_p^2}{\omega^2 + \Gamma^2}} + \boxed{i \frac{\Gamma \omega_p^2}{\omega(\omega^2 + \Gamma^2)}}$$

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Drude-Sommerfeld model

Free electron contribution

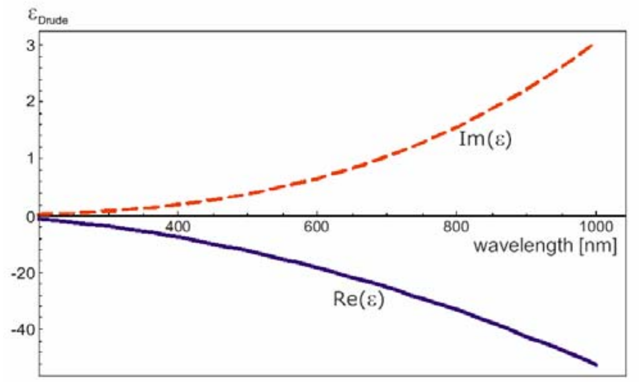

Figure 12.1: Real and imaginary part of the dielectric constant forgold according to the Drude-Sommerfeld free electron model (ω_p=13.8·10^{15} s^{-1},Γ = 1.075 · 10^{14} s^{-1}). The blue solid line is the real part, the red, dashed line is the imaginary part. Note the different scales for real and imaginary part.

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Interband transitions

Bound electron contribution (Interband transitions)

$$m\frac{\partial^2 \mathbf{r}}{\partial t^2} + m\gamma\frac{\partial \mathbf{r}}{\partial t} + \alpha\mathbf{r} = e\mathbf{E}_0 e^{-i\omega t}$$

$$\mathbf{r}(t) = \mathbf{r}_0 e^{-i\omega t} \quad \Rightarrow \quad \varepsilon_{\text{Interband}}(\omega) = 1 + \frac{\tilde{\omega}_{\text{p}}^2}{(\omega_0^2 - \omega^2) - i\gamma\omega}$$

$$\tilde{\omega}_{\text{p}} = \sqrt{\tilde{n}e^2/m\varepsilon_0}$$
$$\omega_0 = \sqrt{\alpha/m}$$

$$\varepsilon_{\text{Interband}}(\omega) = 1 + \frac{\tilde{\omega}_{\text{p}}^2(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2} + i\frac{\gamma\tilde{\omega}_{\text{p}}^2\omega}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2}$$

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Interband transitions

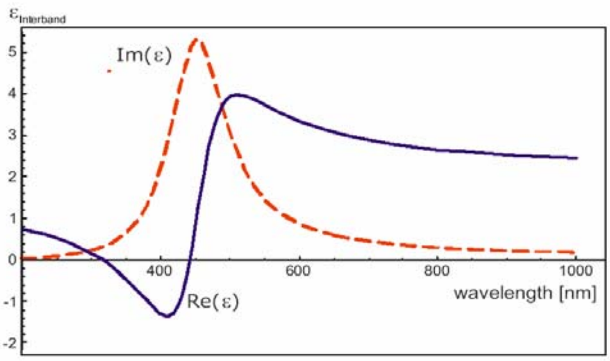

Figure 12.2: Contribution of bound electrons to the dielectric function of gold. The parameters used are $\tilde{\omega}_{\text{p}} = 45 \cdot 10^{14}\ s^{-1}$, $\gamma = 8.35 \cdot 10^{-16}\ s^{-1}$, and $\omega_0 = 2\pi c/\lambda$, with λ=450 nm. The solid blue line is the real part, the dashed red curve is the imaginary part of the dielectric function due to bound electrons.

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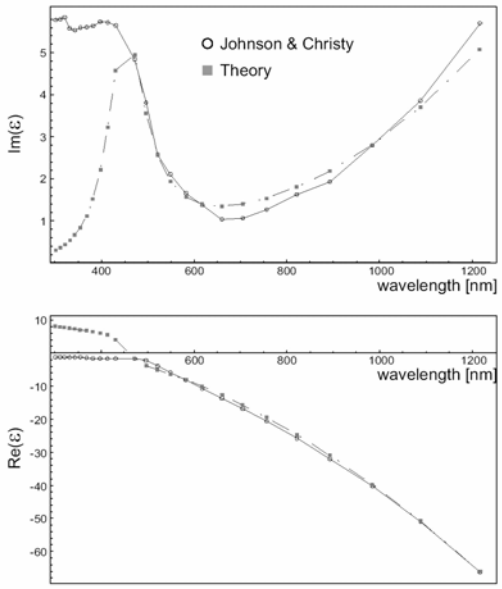
Dielectric function of gold



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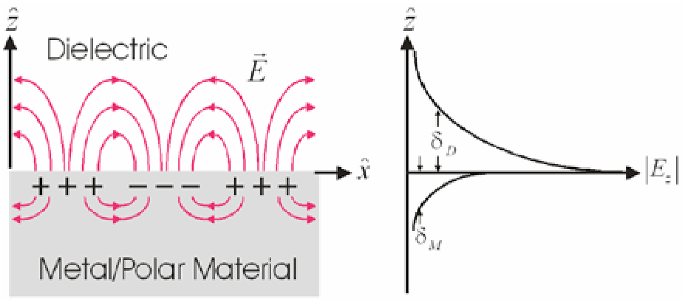
Surface plasmons



surface wave with a certain relation between angular frequency (energy) and wavevector (momentum)

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Solutions of the homogenous Helmholtz equation

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r},\omega) \; - \; \frac{\omega^2}{c^2}\varepsilon(\mathbf{r},\omega)\,\mathbf{E}(\mathbf{r},\omega) \; = \; 0$$

for this system

→ Eigenmode of the system (exists without excitation)

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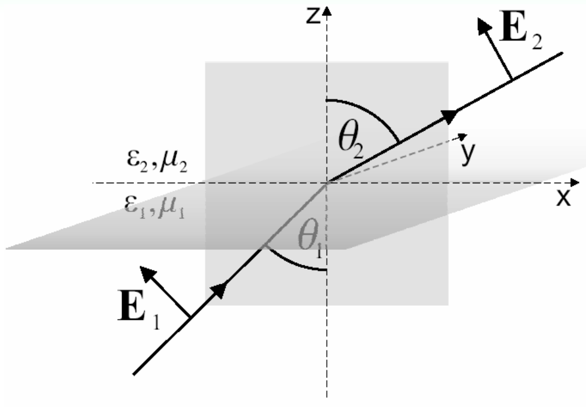
Surface Plasmons



$$\mathbf{E}_{\mathrm{i}} = \begin{pmatrix} E_{\mathrm{j,x}} \\ 0 \\ E_{\mathrm{j,z}} \end{pmatrix} e^{ik_{\mathrm{x}}x - i\omega t} e^{ik_{\mathrm{j},z}z}. \quad j = 1,2$$

p-polarized plane waves

For s-polarized waves → Brewster angle

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Surface Plasmons

displacement fields in both half spaces have to be source free

$\nabla \cdot \mathbf{D} = 0$ → $k_x E_{j,x} + k_{j,z} E_{j,z} = 0, \quad j = 1,2$

$k_{||}$ conserved → $k_x^2 + k_{j,z}^2 = \varepsilon_j k^2, \quad j = 1,2$

Boundary conditions:

continuity of $E_{||}$ → $E_{1,x} - E_{2,x} = 0$

continuity of $D_{\perp}$ → $\varepsilon_1 E_{1,z} - \varepsilon_2 E_{2,z} = 0$

4 equations, 4 unknowns!

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Surface Plasmons

Solutions exist for a vanishing determinant!

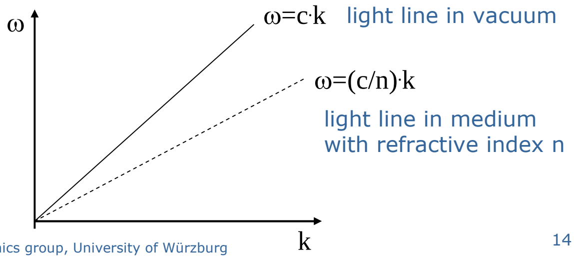
$k_x = 0$ or $\varepsilon_1 k_{2,z} - \varepsilon_2 k_{1,z} = 0$

$$k_x^2 = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} k^2 = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \frac{\omega^2}{c^2}$$

"Dispersion relation"

Compare to the dispersion relation of light in a homogenous medium

$\omega = (c/n) \cdot k$



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$$k_x^2 + k_{j,z}^2 = \varepsilon_j k^2, \quad j = 1,2$$

$$k_{j,z}^2 = \frac{\varepsilon_j^2}{\varepsilon_1 + \varepsilon_2} k^2, \quad j = 1,2$$

$$k_x^2 = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} k^2 = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \frac{\omega^2}{c^2}$$

Define the propagation character of the surface wave

propagating interface modes?

k_x must be real!

$k_{j,z}$ must be purely imaginary!

Assume that imaginary part of ε_1 is small

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We are looking for interface waves that propagate along the interface. This requires a real k_x. Looking at

$$k_x^2 = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} k^2 = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \frac{\omega^2}{c^2}$$

this can be fulfilled if both, the sum and the product of the dielectric functions are either both positive or both negative.

In order to obtain a 'bound' solution, we require that the normal components of the wave vector are purely imaginary in both media giving rise to exponentially decaying solutions. This can only be achieved if the sum in the denominator of

$$k_{j,z}^2 = \frac{\varepsilon_j^2}{\varepsilon_1 + \varepsilon_2} k^2, \quad j = 1,2$$

is negative.

$$\varepsilon_1(\omega) \cdot \varepsilon_2(\omega) < 0$$
$$\varepsilon_1(\omega) + \varepsilon_2(\omega) < 0$$

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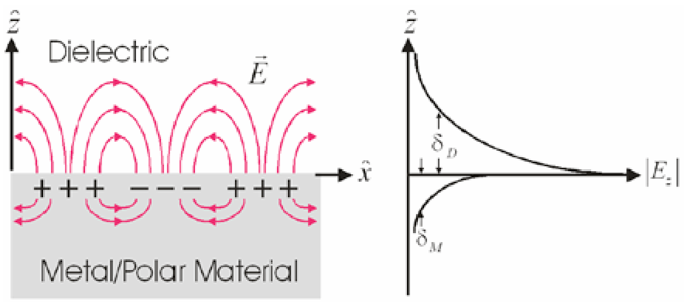
What is it?



Surface – Plasmon – Polariton !

Properties of Surface Plasmons

Now take imaginary part of the metal's dielectric function into account:

$$\varepsilon_1 = \varepsilon_1' + i\varepsilon_1'' \quad \text{with real } \varepsilon_1' \; \varepsilon_1''$$

ε_2 is assumed to be real.

➡ $k_x = k_x' + i k_x''$

Plasmon wavelength (k_x') — damping factor (k_x'')

$|\varepsilon_1''| \ll |\varepsilon_1'|$ ➡

$$k_x' \approx \sqrt{\frac{\varepsilon_1' \varepsilon_2}{\varepsilon_1' + \varepsilon_2}} \frac{\omega}{c}$$

$$k_x'' \approx \sqrt{\frac{\varepsilon_1' \varepsilon_2}{\varepsilon_1' + \varepsilon_2}} \frac{\varepsilon_1'' \varepsilon_2}{2\varepsilon_1'(\varepsilon_1' + \varepsilon_2)} \frac{\omega}{c}$$

Properties of Surface Plasmons

1. Wavelength

$$\lambda_{\rm SPP} = \frac{2\pi}{k'_{\rm x}} \approx \sqrt{\frac{\varepsilon'_1 + \varepsilon_2}{\varepsilon'_1 \varepsilon_2}}\lambda$$

at a wavelength of 632.8 nm
$\varepsilon_1 = -11.6 + 1.2i$ ➡ $\lambda_{SP}=0.96 \cdot \lambda$

2. Propagation length

1/e decay length of the intensity is $1/(2k''_{\rm x})$

for gold at 632.8nm: → **10μm**

3. Decay length

$$k_{1,\rm z} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1'^2}{\varepsilon'_1 + \varepsilon_2}}\left[1 + i\frac{\varepsilon''_1}{2\varepsilon'_1}\right]$$

$$k_{2,\rm z} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_2^2}{\varepsilon'_1 + \varepsilon_2}}\left[1 - i\frac{\varepsilon''_1}{2(\varepsilon'_1 + \varepsilon_2)}\right]$$

$$(1/k_{1,\rm z}, 1/k_{2,\rm z}) = (28 \text{ nm}, 328 \text{ nm})$$

inside the metal (28 nm); in the dielectric (328 nm)

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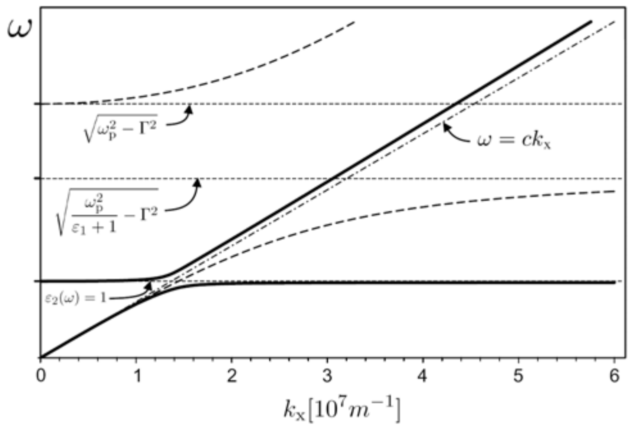
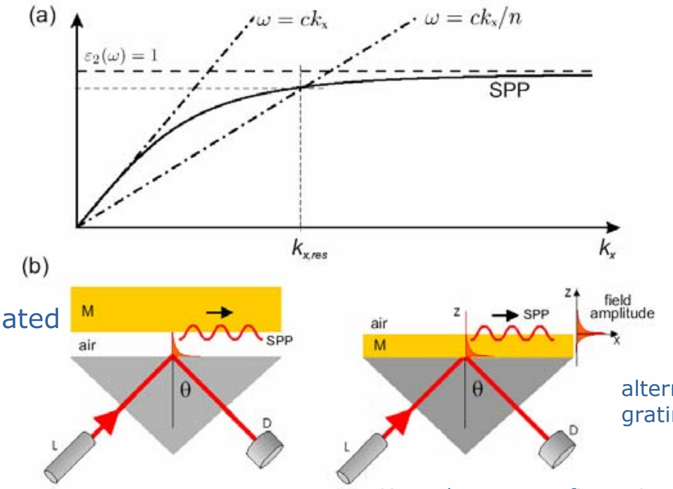


Figure 12.5: Dispersion relation of surface-plasmon polaritons at a gold/air interface. The solid line is the dispersion relation that results from a dielectric function accounting for a single interband transition. The dashed line results from using a Drude type dielectric function. The dash-dotted straight line is the light line $\omega = c \cdot k_x$ in air.

Excitation of SPP

attenuated TIR



alternatively: grating coupler

Otto-configuration

Kretschmann-configuration

Otto configuration

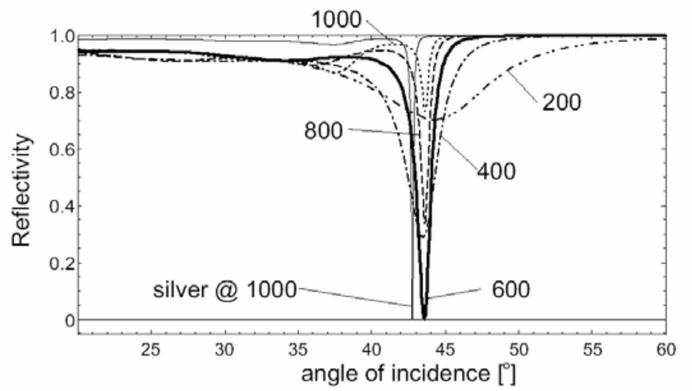


Figure 12.7: Excitation of surface plasmons in the Otto configuration. The reflectivity of the exciting beam is plotted as a function of the incident angle and for different air gaps (in nm). The curves are evaluated for a gold film. For comparison, a single trace is also plotted for silver for which the resonance is much sharper because of lower damping.

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Kretschmann configuration

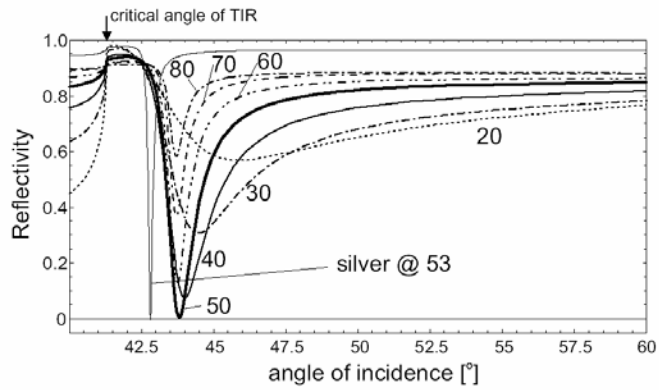

Figure 12.8: Excitation of surface plasmons in the Kretschmann configuration. The reflectivity of the exciting beam is plotted as a function of the incident angle and for different air gaps (in nm). The curves are evaluated for a gold film. For comparison a single trace is also plotted for silver. Note the the much sharper resonance due to the smaller damping of silver as compared to gold. The critical angle of total internal reflection shows up as a discontinuity marked by an arrow.

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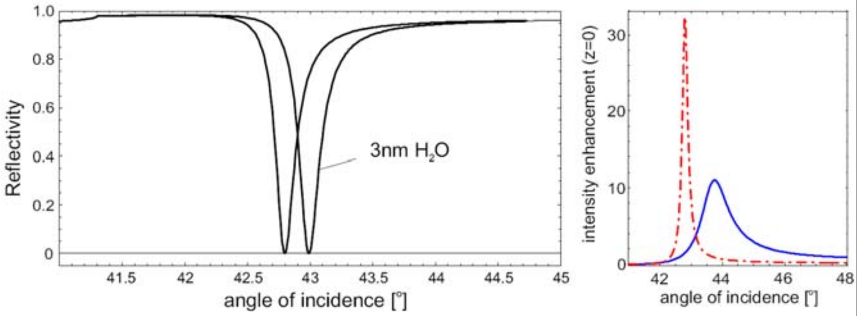
Application

Sensors:



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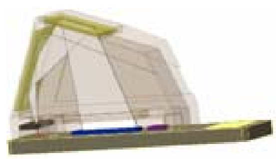
Commercial systems

Biacore 3000 by Biacore International AB (Uppsala, Sweden)

Spreeta by Sensata Technologies (Attleboro, MA)



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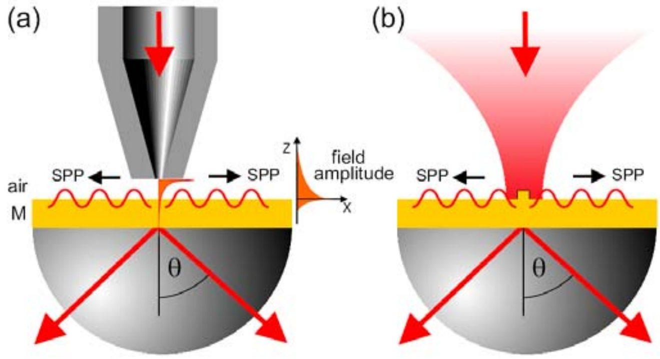
Surface Plasmons in Nano-Optics

Excitation by confined fields:



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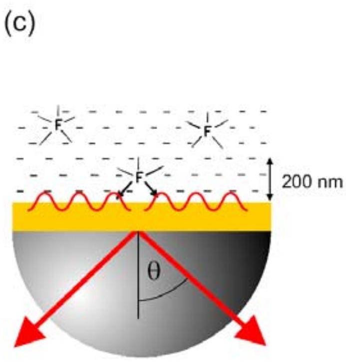

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Excitation & leakage radiation

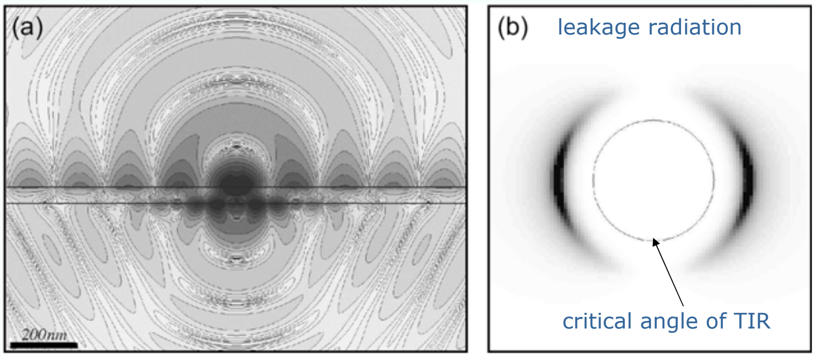

Figure 12.11: Excitation of surface plasmons with a dipole source placed 5nm above a 50nm silver layer supported by a glass substrate. The excitation wavelength is $\lambda = 370nm$ and the dipole moment is parallel to the interface. (a) Lines of constant power density (factor of 2 between successive contour lines) depicted at a certain instant of time. The figure shows the surface plasmon propagation along the top surface of the silver film and also the radiative decay into the lower half space. (b) Radiation pattern evaluated on a horizontal plane in the lower medium. The circle indicates the critical angle of total internal reflection at an air-glass interface. The two lobes result from the radiative decay of surface plasmons excited by the dipole source.

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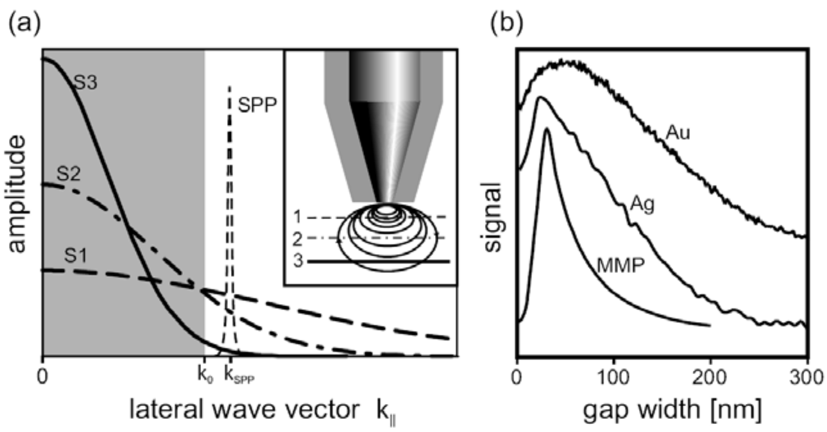
Distance dependence



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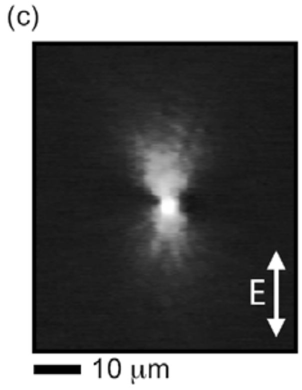
Imaging surface plasmons



leakage radiation.
Microscope focused to the metal film

Imaging surface plasmons

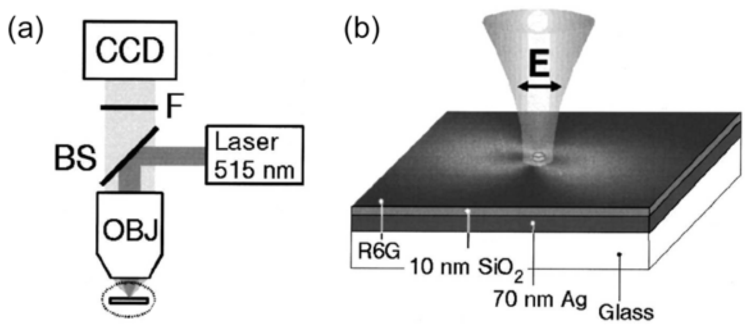
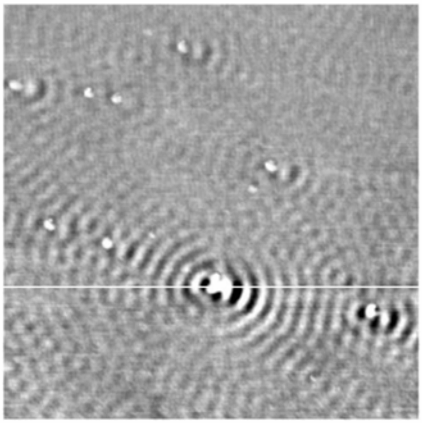
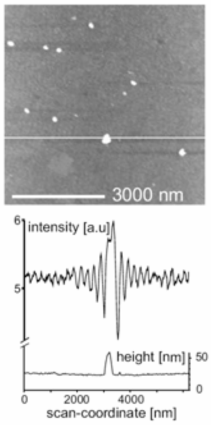


Figure 12.13: Excitation of surface plasmons by a subwavelength-scale protrusion located on the top surface of a metal film. (a) Setup, (b) Close up of the particle-beam interaction area. In this experiment, the surface plasmons are detected by the fluorescence intensity of a thin layer of fluorescent molecules deposited on a dielectric spacer layer. From [20].

Interferences of surface plasmons





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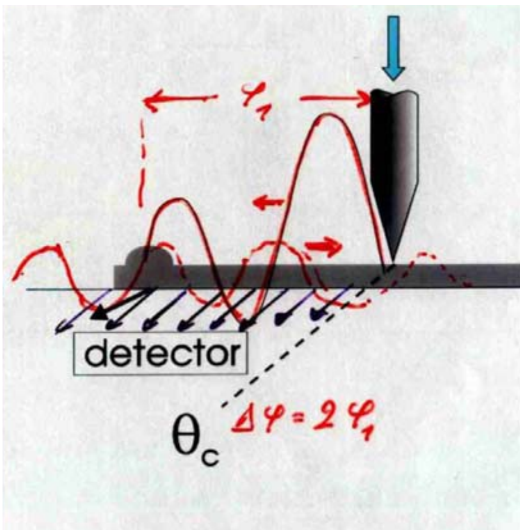

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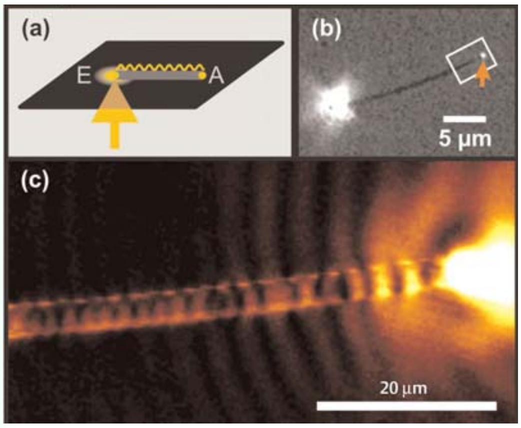

Plasmonischer Lichttransport in einem einkristallinen Nanodraht aus Silber.
a) Fokussiertes Laserlicht (λ = 785 nm) regt am Einkoppel-Ende (E) ein Plasmon an.
b) Mikroskopische Aufnahme eines 18,6 µm langen Drahtes. Der kleine Fleck am Auskoppelende (Pfeil) bestätigt den Lichttransport. c) Rasternahfeldmikroskopische Aufnahme des Auskoppelendes. Der am Drahtende reflektierte Plasmonenanteil interferiert mit dem ankommenden Plasmon.

F.R.Aussenegg, H.Ditlbacher
Plasmonen als Lichttransporter
Physik in unserer Zeit **37**, 220 (2006) pdf (c) 2006 Wiley-VCH

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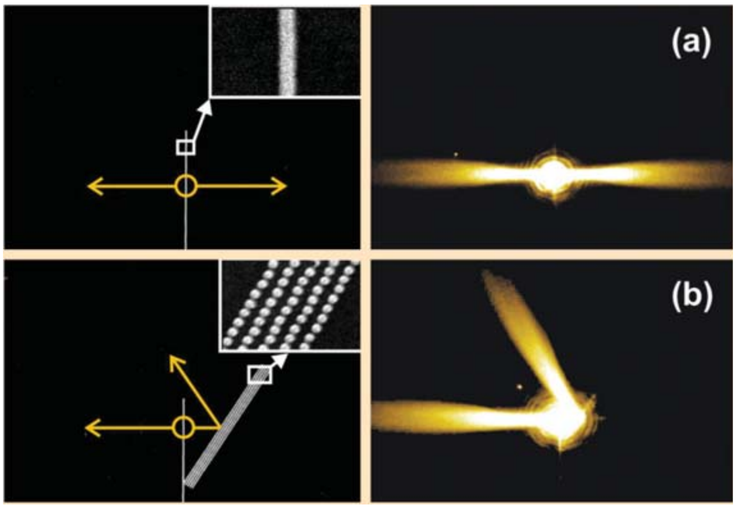

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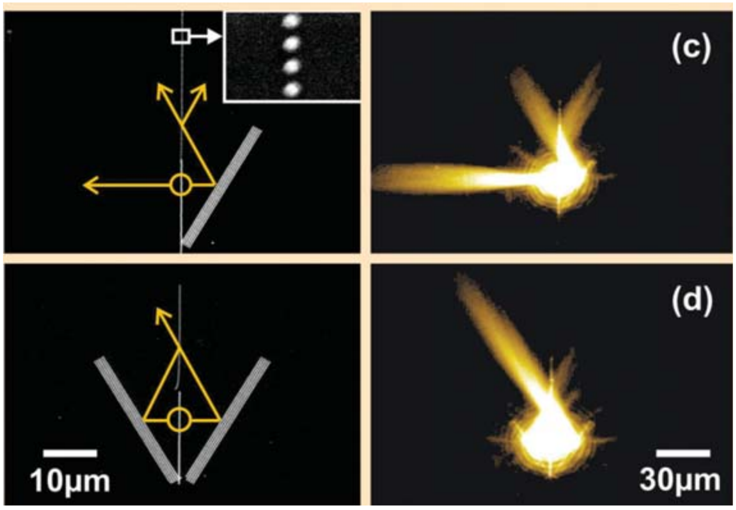

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Quasistatic approximation

Example: Dipole field

$$\mathbf{E}(r\mathbf{n},t) = \frac{1}{4\pi\epsilon_0}\left[k^2(\mathbf{n}\times\mathbf{p})\times\mathbf{n}\frac{e^{ikr}}{r} + [3\mathbf{n}(\mathbf{n}\cdot\mathbf{p})-\mathbf{p}]\left(\frac{1}{r^3}-\frac{ik}{r^2}\right)e^{ikr}\right]e^{i\omega t}$$

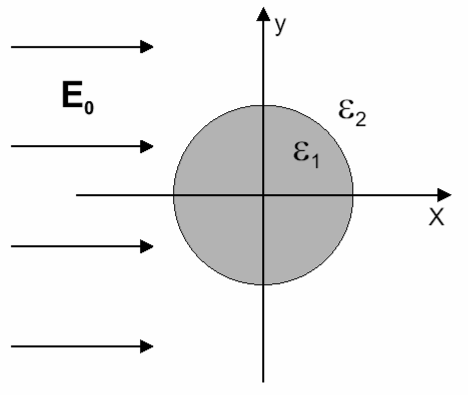
$kr \ll 1$ Near-field zone

$$\mathbf{E}(r\mathbf{n},t) = \frac{1}{4\pi\epsilon_0}[3\mathbf{n}(\mathbf{n}\cdot\mathbf{p})-\mathbf{p}]\frac{e^{i\omega t}}{r^3}$$

= Electrostatic field of a dipole!!!!!
Oscillating with $e^{i\omega t}$!!!!

To find a near-field distribution it is sufficient to determine the solution of an electrostatics problem

Particle plasmons



Electrostatics problem

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Particle plasmons

Laplace eq. in spherical coordinates

$$\frac{1}{r^2 \sin\theta}\left[\sin\theta \frac{\partial}{\partial r}\left(r^2 \frac{\partial}{\partial r}\right) + \frac{\partial}{\partial \theta}\left(\sin\theta \frac{\partial}{\partial \theta}\right) + \frac{1}{\sin\theta}\frac{\partial^2}{\partial \varphi^2}\right]\Phi(r,\theta,\varphi) = 0$$

Ansatz:

$$\Phi(r,\theta,\varphi) = \Sigma_{\mathrm{l,m}}\, b_{\mathrm{l,m}} \cdot \Phi_{\mathrm{l,m}}(r,\theta,\varphi)$$

associated Legendre functions

$$\Phi_{\mathrm{l,m}} = \left\{ \begin{array}{c} r^l \\ r^{-l-1} \end{array} \right\} \left\{ \begin{array}{c} P_l^m(\cos\theta) \\ Q_l^m(\cos\theta) \end{array} \right\} \left\{ \begin{array}{c} e^{im\varphi} \\ e^{-im\varphi} \end{array} \right\}$$

Legendre functions of the second kind

$$\left[\frac{\partial \Phi_1}{\partial \theta}\right]_{r=a} = \left[\frac{\partial \Phi_2}{\partial \theta}\right]_{r=a}$$

$$\varepsilon_1 \left[\frac{\partial \Phi_1}{\partial r}\right]_{r=a} = \varepsilon_2 \left[\frac{\partial \Phi_2}{\partial r}\right]_{r=a}$$

Boundary conditions

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$$\Phi_1 = -E_0 \frac{3\varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} r \cos\theta$$

$$\Phi_2 = -E_0 r \cos\theta + E_0 \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} a^3 \frac{\cos\theta}{r^2}$$

$$\mathbf{E} = -\nabla\Phi \quad \Rightarrow$$

$$\mathbf{E}_1 = E_0 \frac{3\varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} (\cos\theta\, \mathbf{e}_r - \sin\theta\, \mathbf{e}_\theta) = E_0 \frac{3\varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} \mathbf{e}_x$$

$$\mathbf{E}_2 = E_0 (\cos\theta\, \mathbf{e}_r - \sin\theta\, \mathbf{e}_\theta) + \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} \frac{a^3}{r^3} E_0 \,(2\cos\theta\, \mathbf{e}_r + \sin\theta\, \mathbf{e}_\theta)$$

scattered field (econd term) is identical to the electrostatic field of a dipole μ located at the center of the sphere.

$$\boldsymbol{\mu} = \varepsilon_2 \alpha(\omega) \mathbf{E}_0$$

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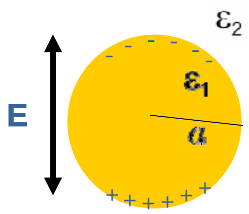
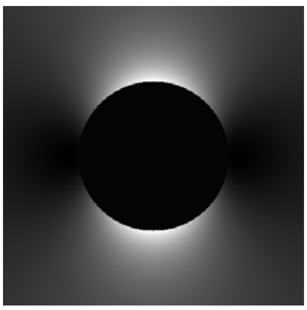
2. Plasmon resonance small particle

Quasi-static limit:

Particles smaller than the skin depth of metal





$$\boldsymbol{\mu} = \varepsilon_2 \alpha(\omega) \mathbf{E}_0$$

Field enhancement is determined only by the material properties, not the size of the particle - but smaller particles give more confinement

$$\alpha(\omega) = 4\pi\varepsilon_o a^3 \frac{\varepsilon_1(\omega) - \varepsilon_2}{\varepsilon_1(\omega) + 2\varepsilon_2}$$

Field enhancement ~ 20 on the surface (for gold)

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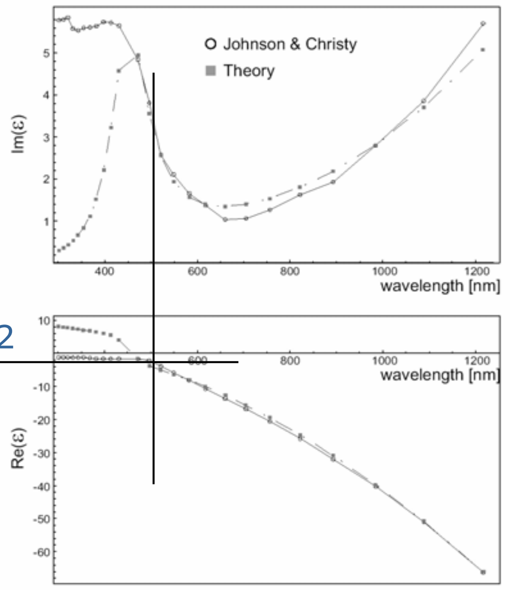
Dielectric function of gold

Re[ε]=-2



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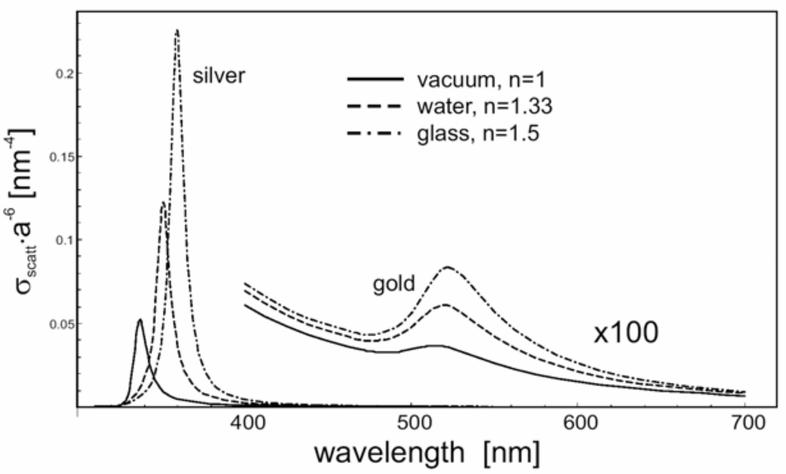
Resonances of gold particles



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Ancient roman Lycurgus cup illuminated by a light source from behind.
Light absorption by the embedded gold particles leads to a red color of the transmitted light whereas scattering at the particles yields a greenish color. From http://www.thebritishmuseum.ac.uk/ science/lycurguscup/sr-lycugus-p1.html.

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