



#### 2328-12

#### Preparatory School to the Winter College on Optics and the Winter College on Optics: Advances in Nano-Optics and Plasmonics

30 January - 17 February, 2012

**Optical antennas: Intro and basics** 

J. Aizpurua Center for Materials Physics, CSIC-UPV/EHU and Donostia International Physics Center - DIPC Spain

#### **Optical antennas: Intro and basics**

#### Javier Aizpurua





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http://cfm.ehu.es/nanophotonics

Center for Materials Physics, CSIC-UPV/EHU and Donostia International Physics Center - DIPC Donostia-San Sebastián, the Basque Country, Spain

Winter College on Optics: Advances in Nano-Optics and Plasmonics February 6-17, 2012 The Abdus Salam International Centre for Theoretical Physics, Trieste, Italy

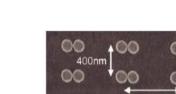
## Outline

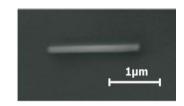
• Optical antennas: Basics

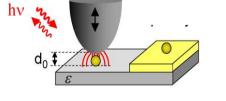
• Playing with modes

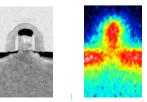
• Optical antennas for Enhanced Spectroscopy: SERS and SEIRA

More applications of Optical antennas

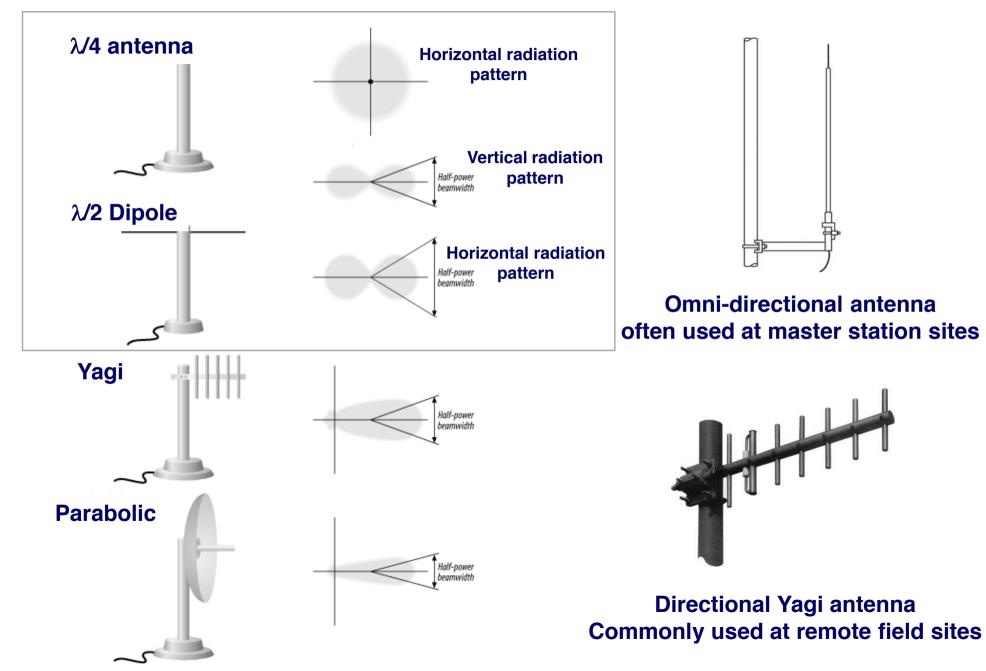








## Different types of radio antennas



#### **Radio Frequency Antennas**



# Half wave dipole antennas



#### **Biconical antenna**



#### Monopole antenna



#### **Combilog** antenna



#### Yagi-Uda antenna



#### Horn antenna

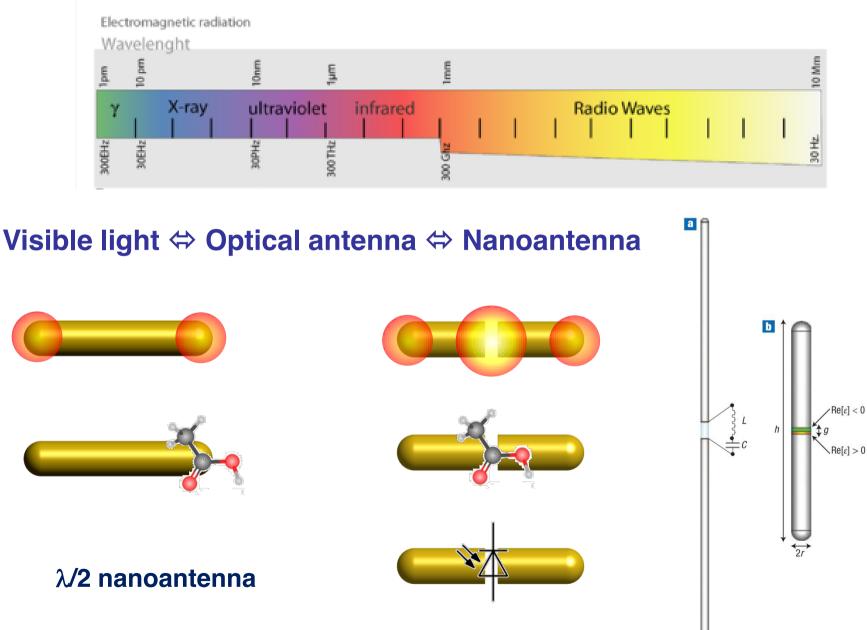


#### Parabolic antenna



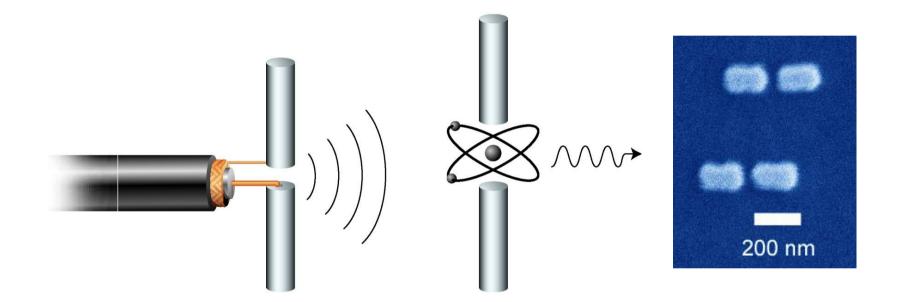
#### Active loop antenna

#### Scaling down in size ← Scaling down in wavelength



### **Optical nanoantennas**

• Design of plasmonic nanoparticles: size, shape, interactions, ...

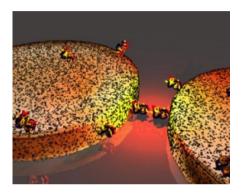


#### Analogy with radio-wave antennas

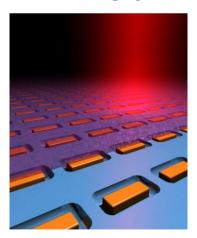
Mühlschlegel et al. Science 308, 1607 (2005)

## **Optical nanoantennas**

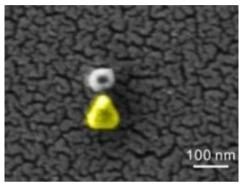
• Applications: biosensing, nonlinear optics / SERS, fluorescence, quantum optics,...



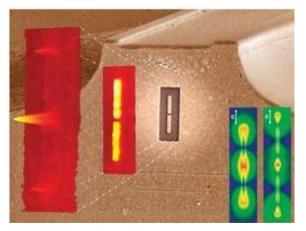
Quidant, ICFO Antenna-gap sensing



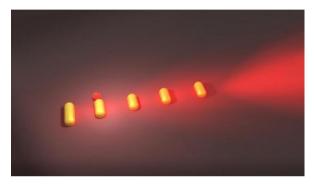
Halas, Nordlander Plasmonic photodetector



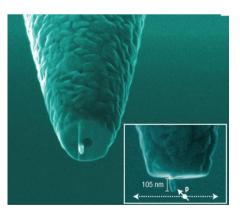
Alivisatos, Giessen Pd-antenna sensor



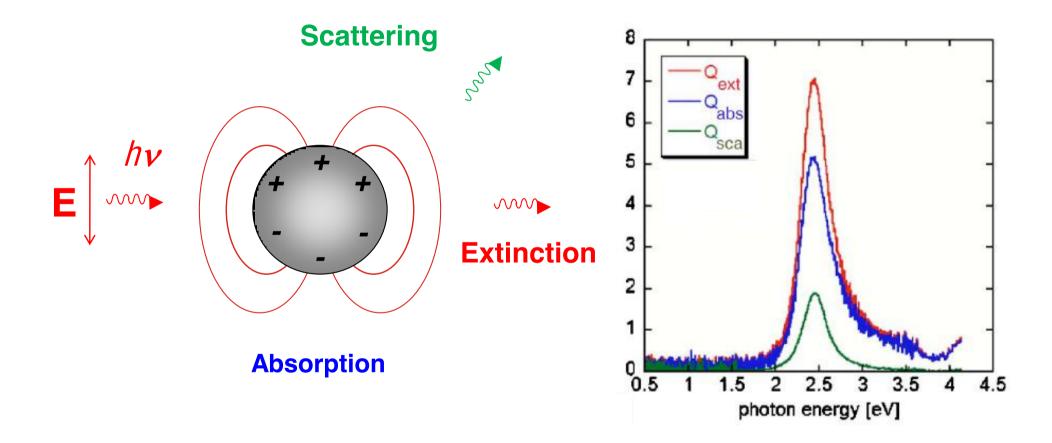
Capasso Antenna QCL



Van Hulst, ICFO - Single molecule, scanning probe



#### The simplest optical antenna: a metallic particle



#### Enhancement of absorption and emission: Bringing effectively the far-field into the near-field

### Metal particle plasmons

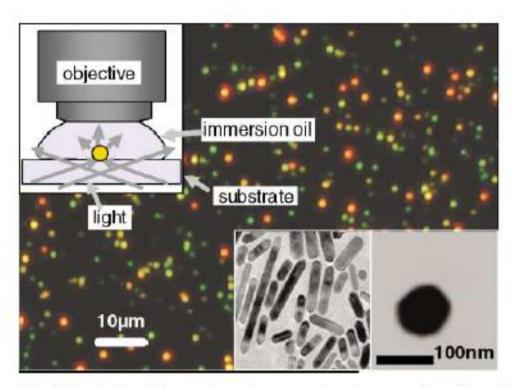
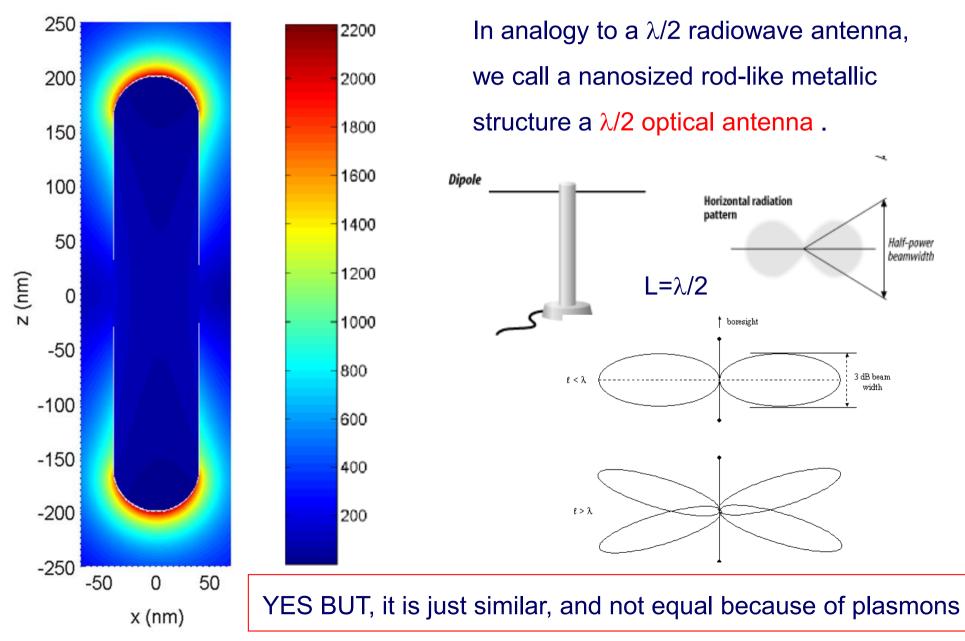


FIG. 2 (color). True color photograph of a sample of gold nanorods (red) and 60 nm nanospheres (green) in dark-field illumination (inset upper left). Bottom right: TEM images of a dense ensemble of nanorods and a single nanosphere.

Standard textbooks:

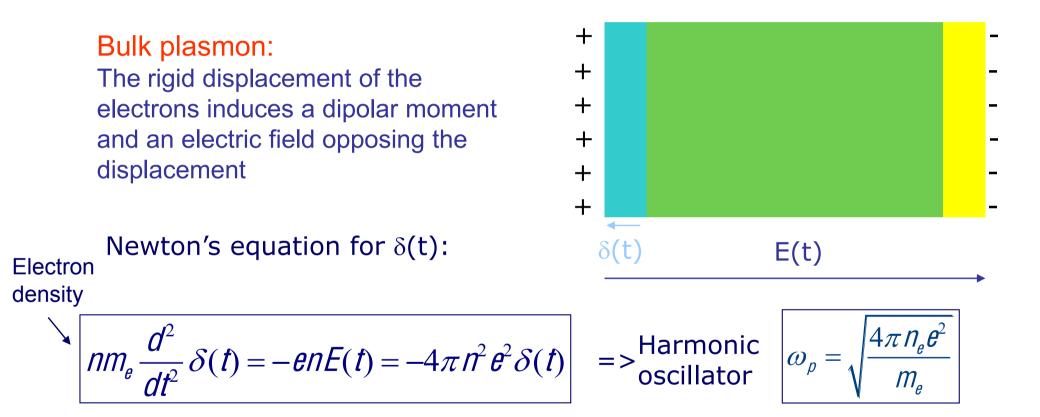
- Kreibig, Vollmer, Optical properties of metal clusters, Springer1995
- Bohren, Huffmann, Absorption and scattering of light by small particles, Wiley 1983

#### Metallic nanorod as a $\lambda/2$ optical antenna



### Bulk plasmons

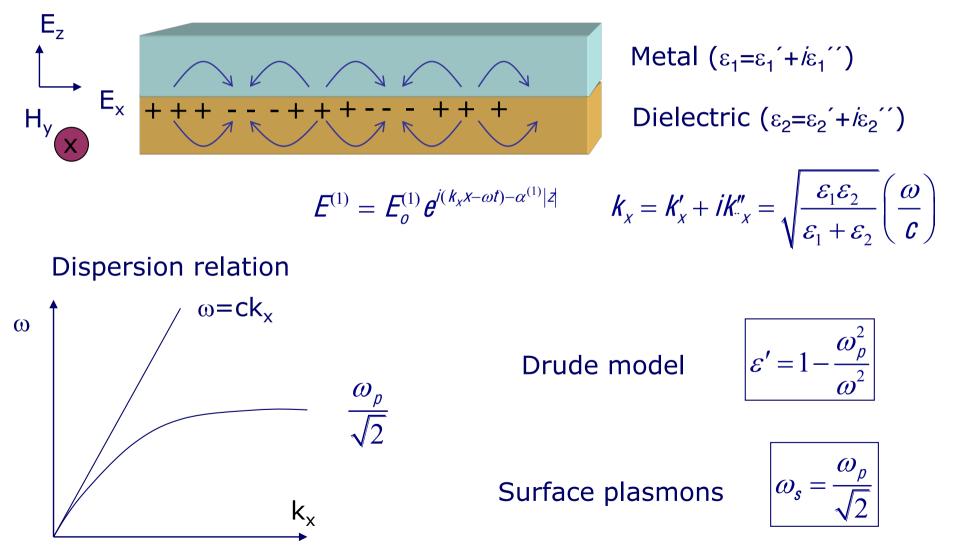
#### A plasmon is a collective oscillation of the conduction electrons



# All the electrons are involved in the oscillation. The energy of those oscillations in typical metals might be triggered out by external probes.

### Surface plasmons

Electromagnetic surface waves which exist at the interface between 2 media whose  $\varepsilon$  have opposite sign.



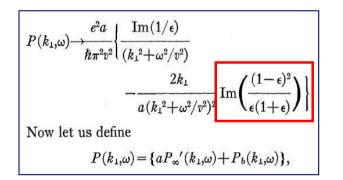
e

#### Plasma Losses by Fast Electrons in Thin Films\*

R. H. RITCHIE

Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee (Received February 7, 1957)

The angle-energy distribution of a fast electron losing energy to the conduction electrons in a thick metallic foil has been derived assuming that the conduction electrons constitute a Fermi-Dirac gas and that the fast electron undergoes only small fractional energy and momentum changes. This distribution exhibits both collective interaction characteristics and individual interaction characteristics, and is more general than the result obtained by other workers. Describing the conduction electrons by the hydro-dynamical equations of Bloch, it has been shown that for very thin idealized foils energy loss may occur at a value which is less than the plasma energy while as the foil thickness decreases below  $\sim v/\omega_p$  the loss at the plasma energy becomes less than that predicted by more conventional theories. The net result is an increase in the energy loss per unit thickness as the foil thickness is decreased. It is suggested that the predicted loss at subplasma energies may correspond to some of the low-lying energy losses which have been observed by experimenters using thin foils.



Now let us define

$$P(k_{\perp},\omega) = \{aP_{\omega}'(k_{\perp},\omega) + P_{b}(k_{\perp},\omega)\},\$$

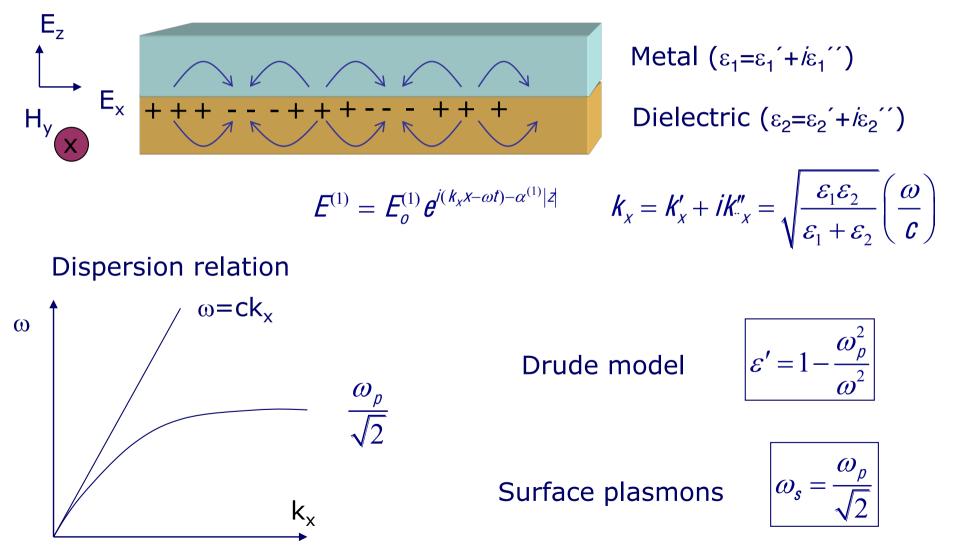
where  $P_{\infty}'$  is the transition probability per unit foil thickness in an infinite foil and  $P_b$  is the term introduced by the boundary effect. Then one may write, inserting the expression for  $\epsilon$ ,

$$P_{b} = \frac{e^{2}}{\pi^{2} \hbar v^{2}} \frac{2k_{1}}{(k_{1}^{2} + \omega^{2}/v^{2})^{2}} \cdot \frac{g\omega_{p}^{4}}{\omega} \cdot \left\{ \frac{\frac{1}{4}\omega_{p}^{2}}{(\omega^{2} - \frac{1}{2}\omega_{p}^{2})^{2} + g^{2}\omega^{2}} - \frac{\omega_{p}^{2}}{(\omega^{2} - \omega_{p}^{2})^{2} + g^{2}\omega^{2}} \right\}.$$
 (25)

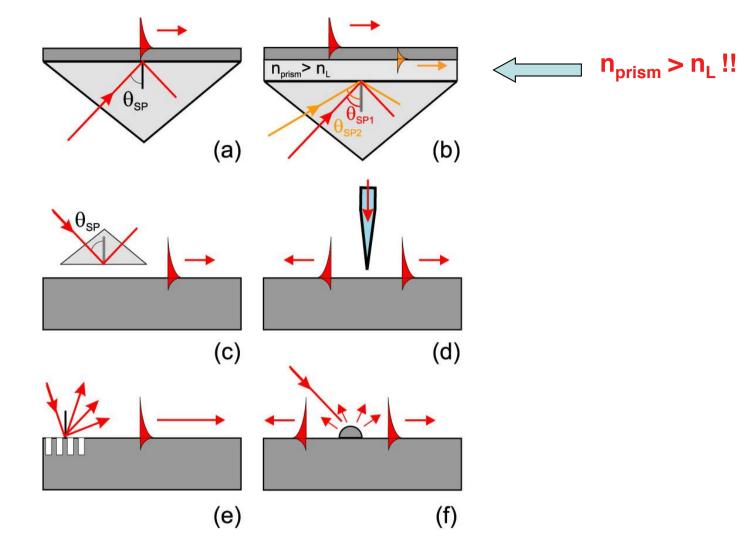
One notes that the effect of the boundary is to cause a decrease in loss at the plasma frequency and an additional loss at  $\omega = \omega_p / \sqrt{2}$ . Call the probabilities for these

### Surface plasmons

Electromagnetic surface waves which exist at the interface between 2 media whose  $\varepsilon$  have opposite sign.



#### Methods of SPP excitation

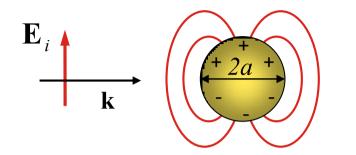


SPP excitation configurations: (a) Kretschmann geometry, (b) two-layer Kretschmann geometry, (c) Otto geometry, (d) excitation with an SNOM probe, (e) diffraction on a grating, and (f) diffraction on surface features.

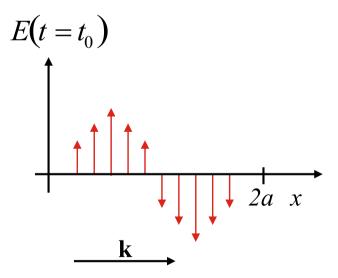
## Nano-optics with localised plasmons

#### Resonances dependence **Characteristics** with size with shape Confined fields: - Nanooptics Enhanced field: - Lighting rod effect • Tunability: - Geometry with material with coupling $\mathbf{m}$ $\mathbf{T}$ -Au • Coupling: - Ag Abs • Wavelength range: - Visible -> Infrared 400 500 600 700 Wave length(nm) $\omega(\mathbf{k})$ $\omega = ck$ **Plasmon polariton** Confined ω<sub>p</sub> + plasmon $\omega_{s}$ -----k

## Light-particle interaction

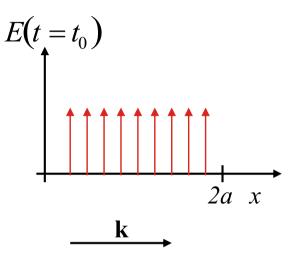


General case:  $\lambda \ll a$ 



Phase shifts in the particles: retardation, multipole excitations

Quasi-static case:  $\lambda >> a$ 



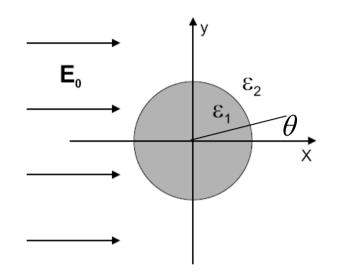
Homogeneous polarization:

All points of an object respond simultaneously to the incoming (exciting) field.

 $\implies$  Helmholtz eq. reduces to Laplace equation:  $\nabla^2 \Phi = 0$ 

 $\nabla^2 \Phi = 0$ 

$$\implies$$
 el. field:  $E = -\nabla \Phi$ 



The electric fields inside (E<sub>1</sub>) and outside (E<sub>2</sub>) the sphere can be obtained from the scalar potentials  $\Phi = \Phi(r, \theta, \varphi)$ 

#### Solve Lapace equation in spherical coordinates:

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

**Boundary conditions:** 

$$\frac{\partial \Phi_1}{\partial \theta} = \frac{\partial \Phi_1}{\partial \theta} \quad (r = a)$$

 $\varepsilon_1 \frac{\partial \Phi_1}{\partial r} = \varepsilon_2 \frac{\partial \Phi_2}{\partial r} \quad (r = a)$ 

continuity of the tangential electric fields

Homogeneous electric field along x-direction:  $\Phi_0 = -E_0 X = -E_0 r \cos \theta$ 

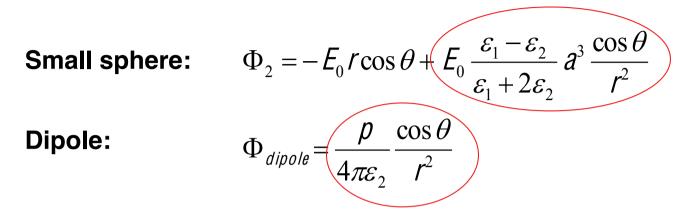
The following potentials satisfy the Laplace equation and boundary conditions:

$$\Phi_1 = -E_0 \frac{3\varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} r \cos \theta$$
  
$$\Phi_2 = -E_0 r \cos \theta + E_0 \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} a^3 \frac{\cos \theta}{r^2}$$

From  $E = -\nabla \Phi$  we obtain

$$\mathbf{E}_{1} = E_{0} \frac{3\varepsilon_{2}}{\varepsilon_{1} + 2\varepsilon_{2}} (\cos\theta \,\mathbf{e}_{r} - \sin\theta \,\mathbf{e}_{\theta}) = E_{0} \frac{3\varepsilon_{2}}{\varepsilon_{1} + 2\varepsilon_{2}} \,\mathbf{e}_{x}$$
$$\mathbf{E}_{2} = E_{0} (\cos\theta \,\mathbf{e}_{r} - \sin\theta \,\mathbf{e}_{\theta}) + \frac{\varepsilon_{1} - \varepsilon_{2}}{\varepsilon_{1} + 2\varepsilon_{2}} \frac{a^{3}}{r^{3}} E_{0} (2\cos\theta \,\mathbf{e}_{r} + \sin\theta \,\mathbf{e}_{\theta})$$

The field is independent of the azimuth angle  $\phi$  which is a result of the symetry implied by the direction of the applied electric field.



The field outside the sphere is the superposition of the applied field and the field of an ideal dipole at the sphere origin

The dipole moment is given by

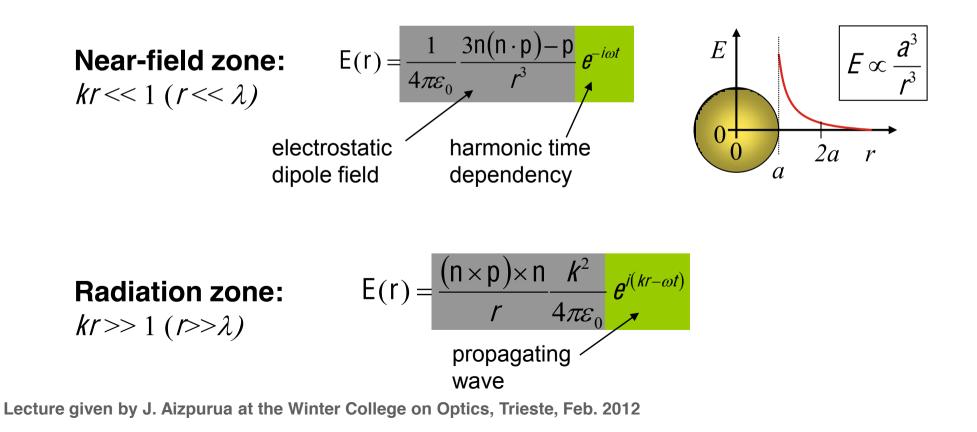
$$\mathbf{p} = 4\pi\varepsilon_0\varepsilon_2 a^3 \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} \mathbf{E}_0$$

Generally the dipole moment is defined by  $\mathbf{p} = \varepsilon_0 \varepsilon_2 \alpha \mathbf{E}_0$ 

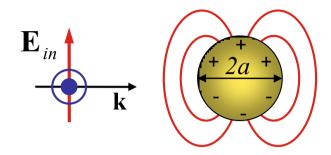
$$\implies \text{Polarizability of the sphere: } \alpha = 4\pi a^3 \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2}$$

We can describe the light scattering of a small sphere by plane wave scattering at an ideal point dipole with dipole moment derived on the previous slides. The dipole field is given by:

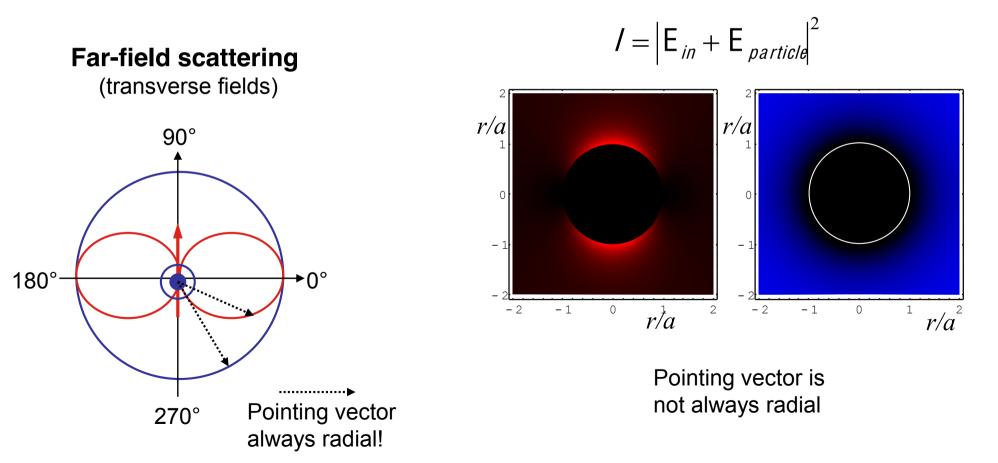
$$\mathsf{E}(\mathsf{r}) = \frac{1}{4\pi\varepsilon_0} \frac{\boldsymbol{\theta}^{ikr}}{r} \left\{ k^2 [(\mathsf{n} \times \mathsf{p}) \times \mathsf{n}] + \frac{1}{r} \left(\frac{1}{r} - ik\right) [3\mathsf{n}(\mathsf{n} \cdot \mathsf{p}) - \mathsf{p}] \right\} \boldsymbol{\theta}^{i\omega t} \quad \text{with} \quad \mathsf{n} = \frac{\mathsf{r}}{r}$$



### Far- and near-field calculations for a sphere $a << \lambda$



**Near-field scattering** 

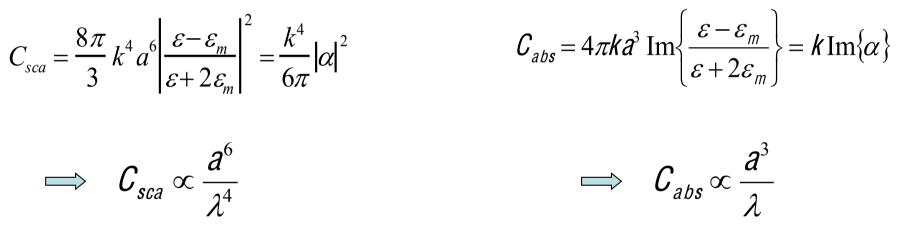


#### Optical cross sections of small spheres

Integrating the Poynting vector  $\mathbf{S}_{sca} (\mathbf{S}_{abs})$  over a close spherical surface we obtain the totally scattered (absorbed) power  $P_{sca} (P_{abs})$  from which we can calculate the scattering (absorption) cross section  $C_{sca} = P_{sca}/I_i$  ( $C_{abs} = P_{abs}/I_i$ ):

#### Scattering cross section:

#### Absorption cross section:



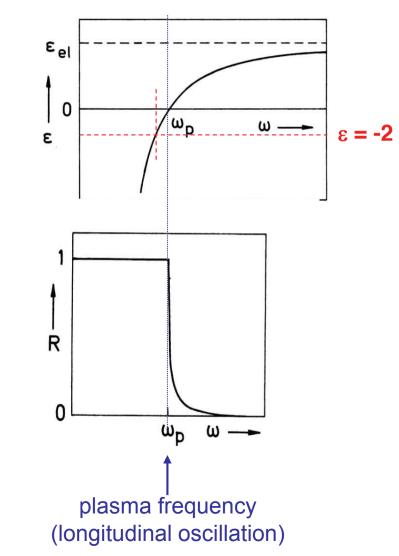
- stronger scattering at shorter wavelength (Rayleigh scattering, blue sky)

- for large particle extinction is dominated by scattering whereas for small particles it is associated with absorption

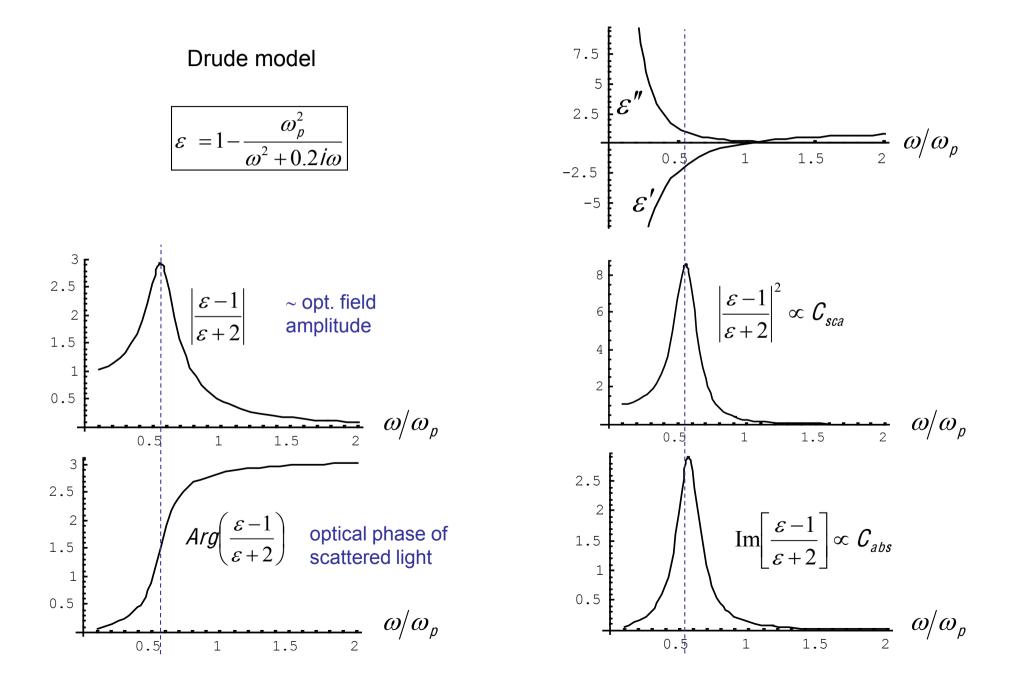
- scattering of single particles <10nm is difficult to measure (low signal/noise and low signal/background)

### **Dielectric function of metals**

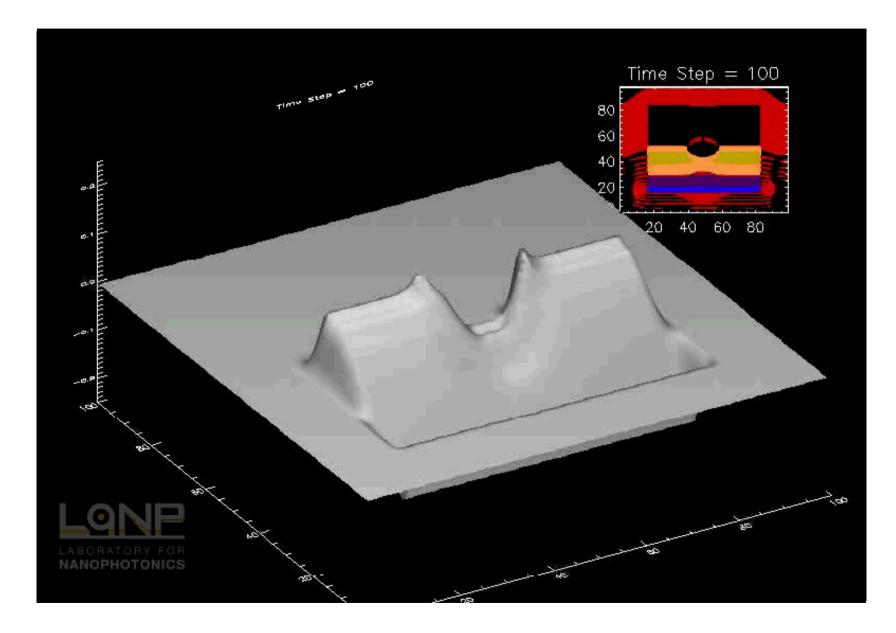
Collective free electron oscillations (plasmons)



#### Optical cross sections of small spheres



## Excitation of a plasmon by a pulse



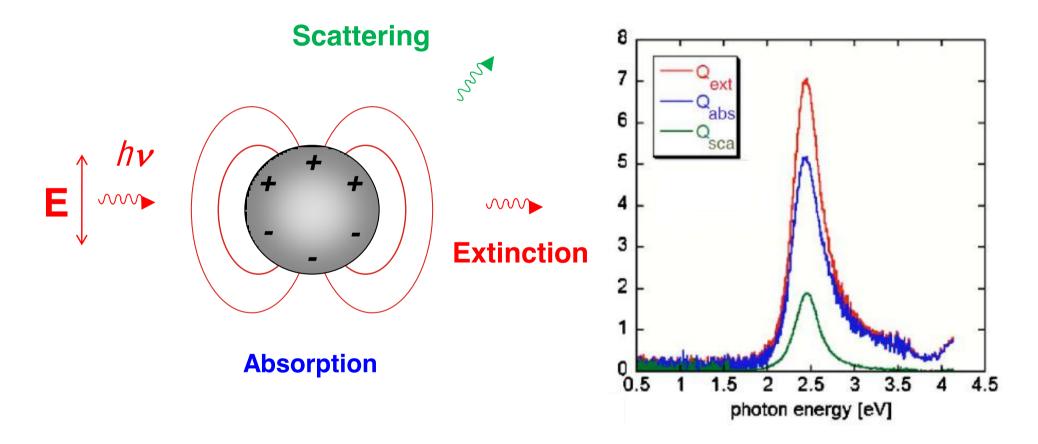
Lecture given by J. Aizpurua at the Winter College on Optics, Trieste, Feb. 2012

**Rice University, Houston** 

## Localised Surface Plasmon: a swimming pool of e-

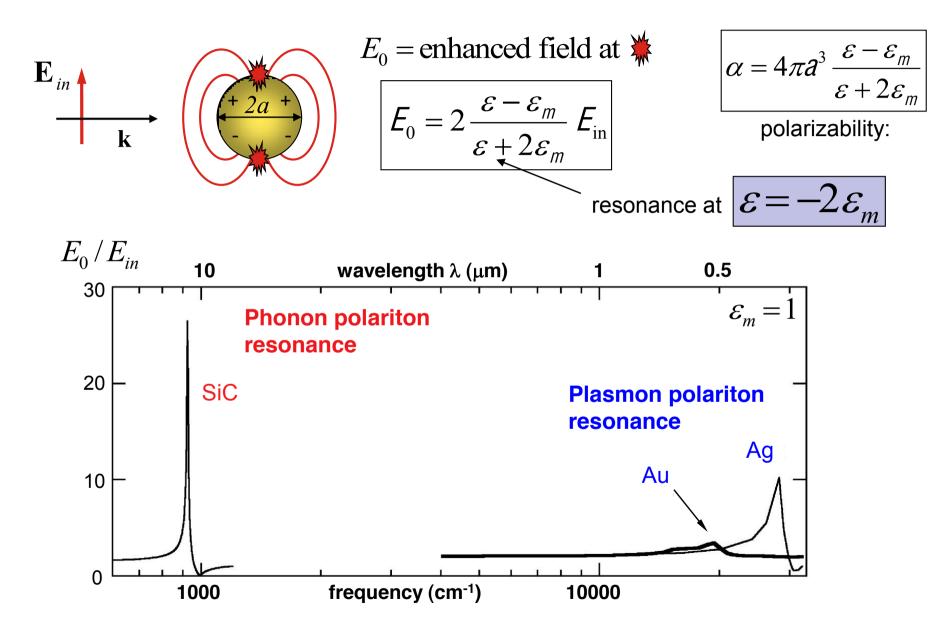


#### The simplest optical antenna: a metallic particle



#### Enhancement of absorption and emission: Bringing effectively the far-field into the near-field

### Small particle resonances



Nanotechnology with plasmonics: before the nanorevolution

#### Lycurgus Cup

#### (British Museum; 4<sup>th</sup> century AD)



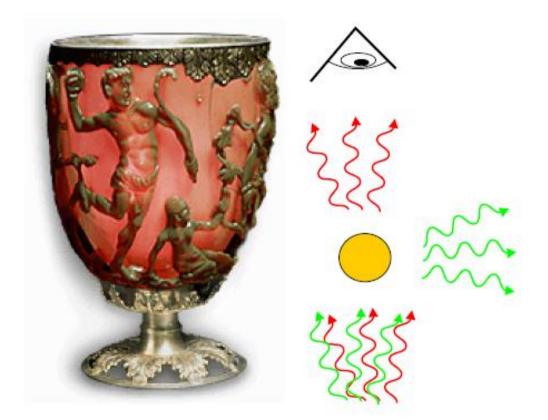


#### **Illumination: from outside**

from inside

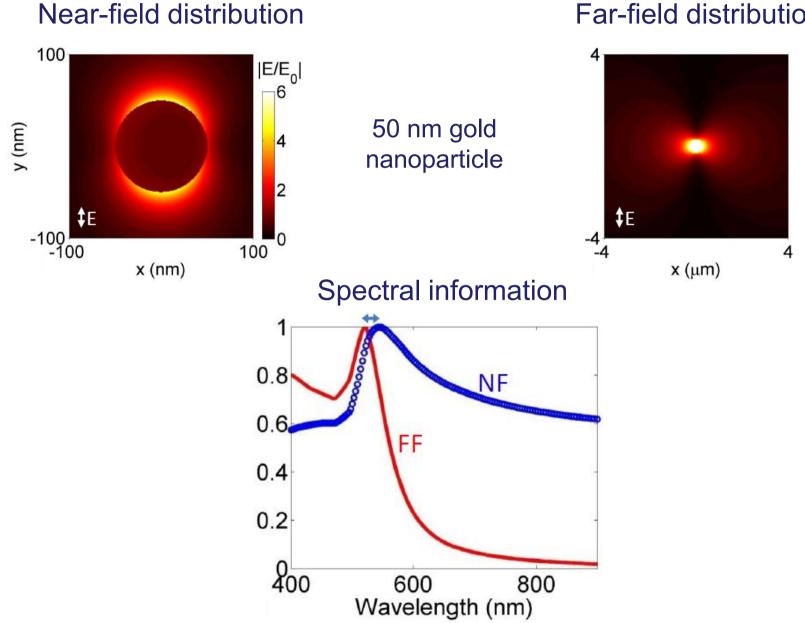
#### (strong absorption at and below 520 nm)

### Extinction vs. scattering



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.

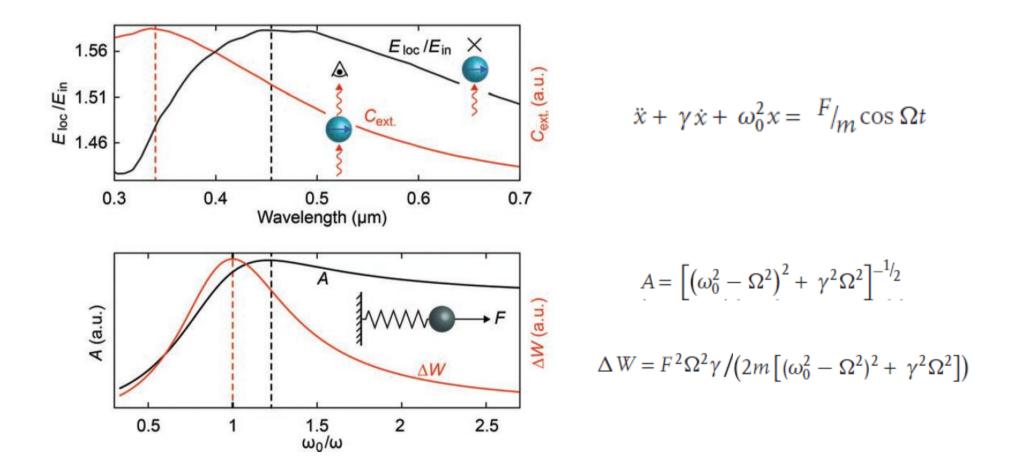
#### Near-field versus far-field



Lecture given by J. Aizpurua at the Winter College on Optics, Trieste, Feb. 2012

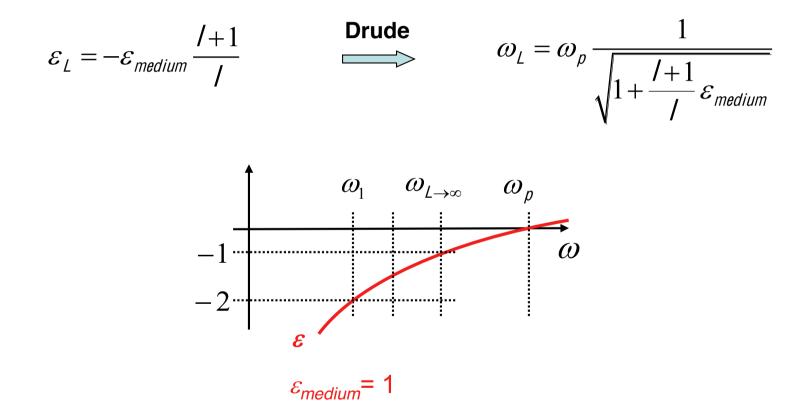
#### Far-field distribution

#### Near-field versus far-field



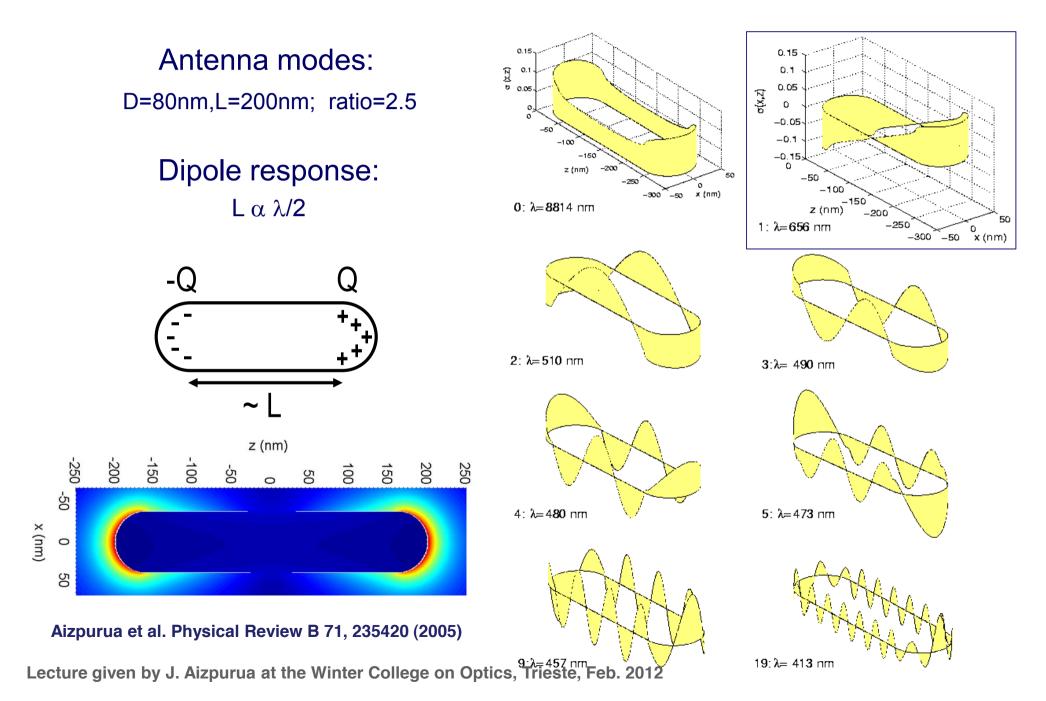
### Higher multipole resonances in quasistatic limit

In the quasistatic limit the Mie theory yields the resonance positions of the higher multipoles at

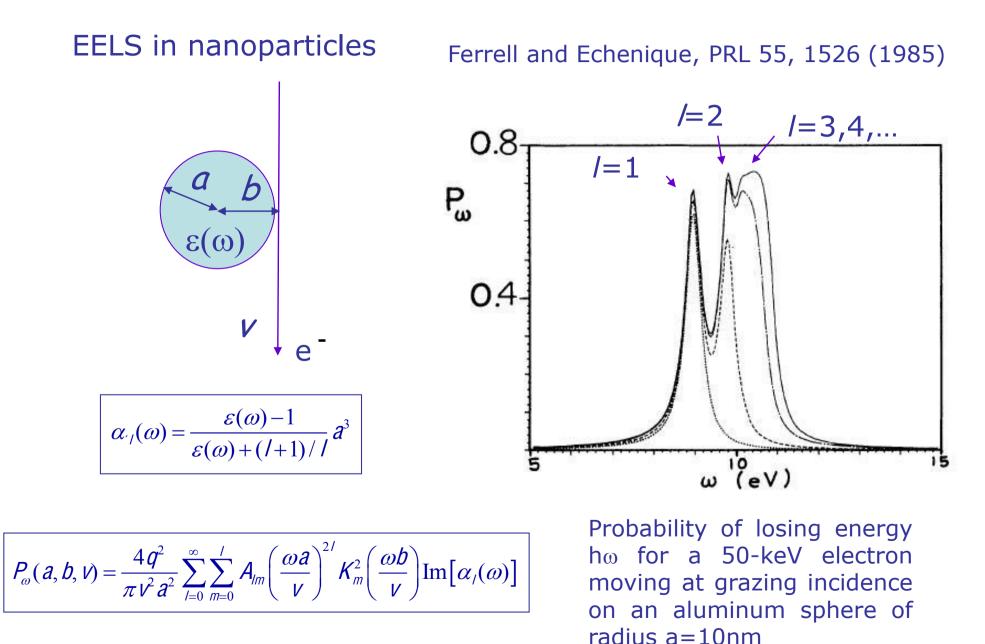


## However, higher multipoles in the quasistatic limit are negligible compared to the dipole contribution (*I*=1)

#### Higher order modes: *I*=1, 2, 3, 4,....



### An alternative to excite high order modes in a sphere



# **Mie-theory**

#### 2.1.3 Exact Electrodynamic Calculation of Spherical Metal Clusters (Mie Theory) Kreibig/Vollmer

The above discussion of the *quasi-static regime* serves as a first rough estimate which only holds for sufficiently small particles and needs to be extended considerably in order to account for larger particle sizes and particle-size distributions.

The general solution of the diffraction problem of a single sphere of arbitrary material within the frame of electrodynamics was first given by Mie in 1908 [2.19]. He applied Maxwell's equations with appropriate boundary conditions in spherical coordinates using multipole expansions of the incoming electric and magnetic fields. Input parameters were the particle size and the optical functions of the particle material and of the surrounding medium. His solution was based upon the determination of scalar electromagnetic potentials from which the various fields werde derived. In particular there are two sets of potentials  $\Pi$ , solving the wave equation

 $\Delta \Pi + |\boldsymbol{k}|^2 \Pi = 0 \tag{2.15a}$ 

in spherical coordinates:

 $\Pi_{e,m}^{inc}$  of the incident plane wave  $\Pi_{e,m}^{in}$  of the wave inside the cluster  $\Pi_{e,m}^{sca}$  of the outgoing scattered wave

The indices e and m indicate the sets of *electrical* and *magnetical* partial waves, respectively. The solutions can be separated in spherical coordinates

 $\Pi = R(r)\Theta(\theta)\Phi(\phi) \tag{2.15b}$ 

and have the form

$$\Pi = \{ \text{cylindrical fct.} \} \cdot \{ \text{Legendre spherical fct.} \}$$

$$\cdot \{ \text{trigonometric fct.} \}$$
(2.15c)

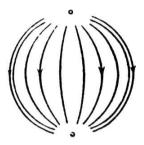
The relevant parameter in all formulas is the size parameter  $x = |\mathbf{k}|R$  which distinguishes the regime of geometrical optics  $(x \gg 1)$  from the one important for clusters  $(x \ll 1)$  (a compressed description is given in [2.9]).

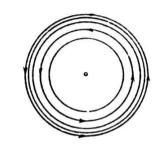
Mie-theory is an electrodynamic theory for optical properties of spherical particles. The solution is divided into two parts: the elctromagnetic one which is treated from first principles (Maxwell equations) and the material problem with is solved by using phenomenological dielectric functions taken from experiments or model calculations

#### See also Bohren/Huffman

# Mie theory - results

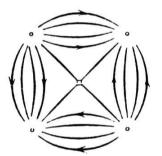
YA'I

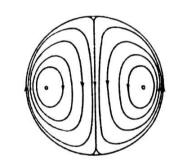




Electric field L = 1

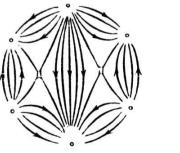
Magnetic field L = 1

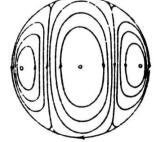




Electric field L = 2

Magnetic field L = 2





Electric field L = 3

Magnetic field L = 3

Fig. 2.6. Electric and magnetic fields far away from the clusters, of the L = 1, 2, and 3 electric partial wave, i.e. the electric dipole, quadrupole, and octupole mode. The same field distributions hold for the magnetic partial waves, if electric and magnetic fields are interchanged (after [2.19]).

Scattering YS1

Absorption YA1

Fig. 2.6 shows farfield distribution at the surface of a large sphere centered at the small cluster

#### Kreibig/Vollmer

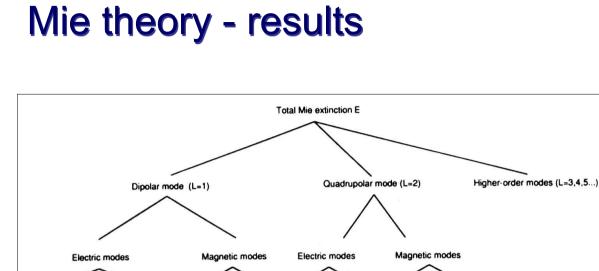
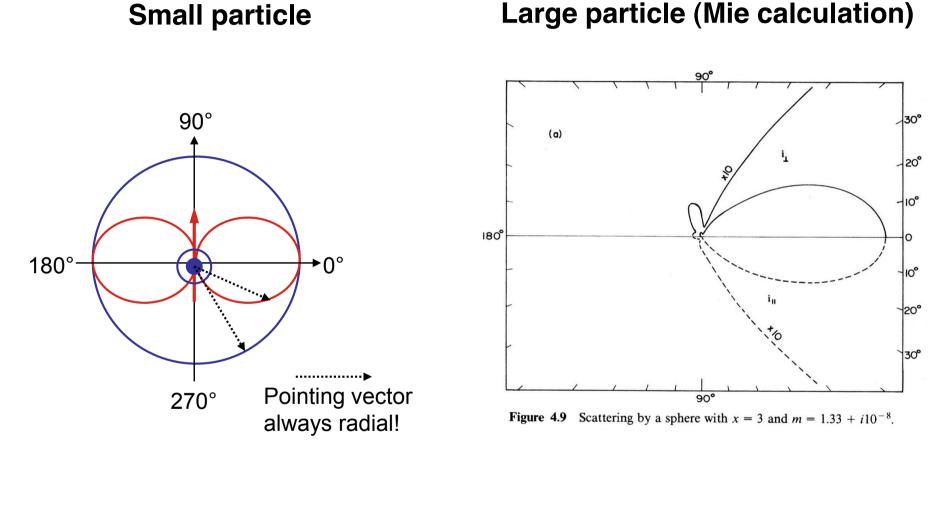


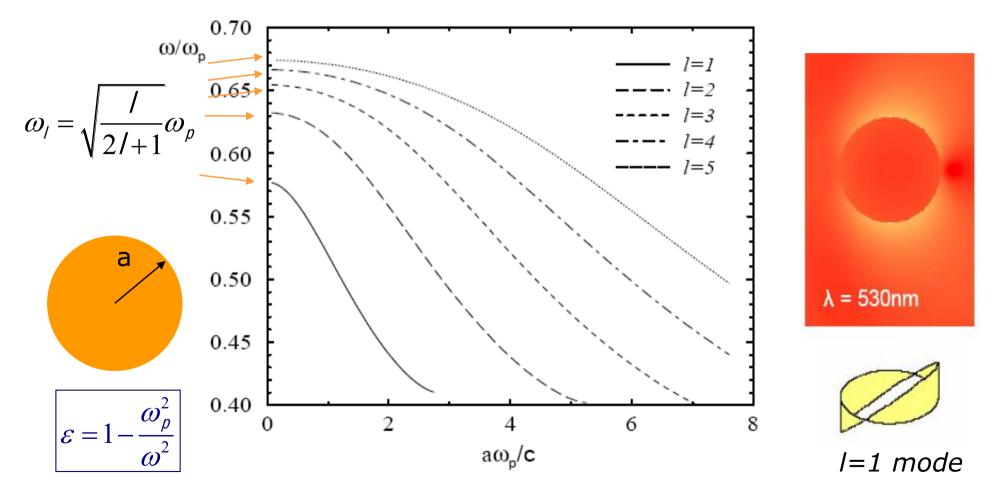
Fig. 2.5. Scheme for decomposing the total Mie extinction spectra in dipolar, quadrupolar and higher modes of electronic excitations. Each multipole contributes by electric and magnetic modes, i.e. plasmons and eddy currents which each consist of absorption and scattering losses.

# Scattering characteristics (far-field)



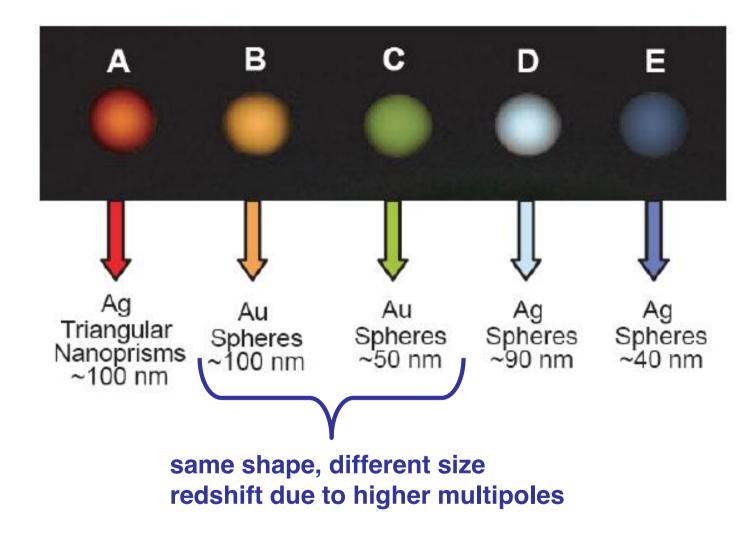
Strong forward scattering

# Spherical plasmons: Mie modes derivaded from Maxwell's equations



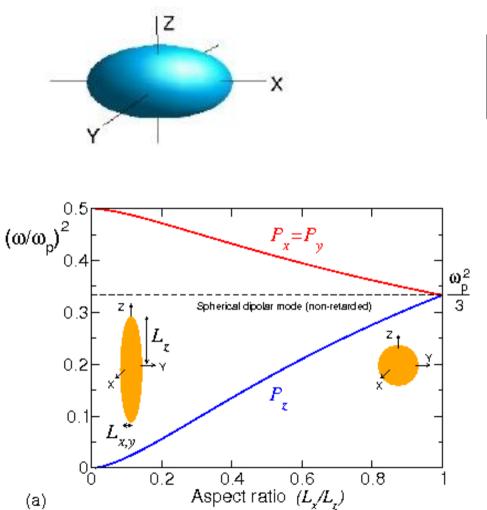
#### **Drude-like metal**

# Effect of finite size on the resonant frequency



Jin et. al., Science 294, 1901 (2001)

# Shape: Polarizability of small ellipsoids



quasistatic approximation:

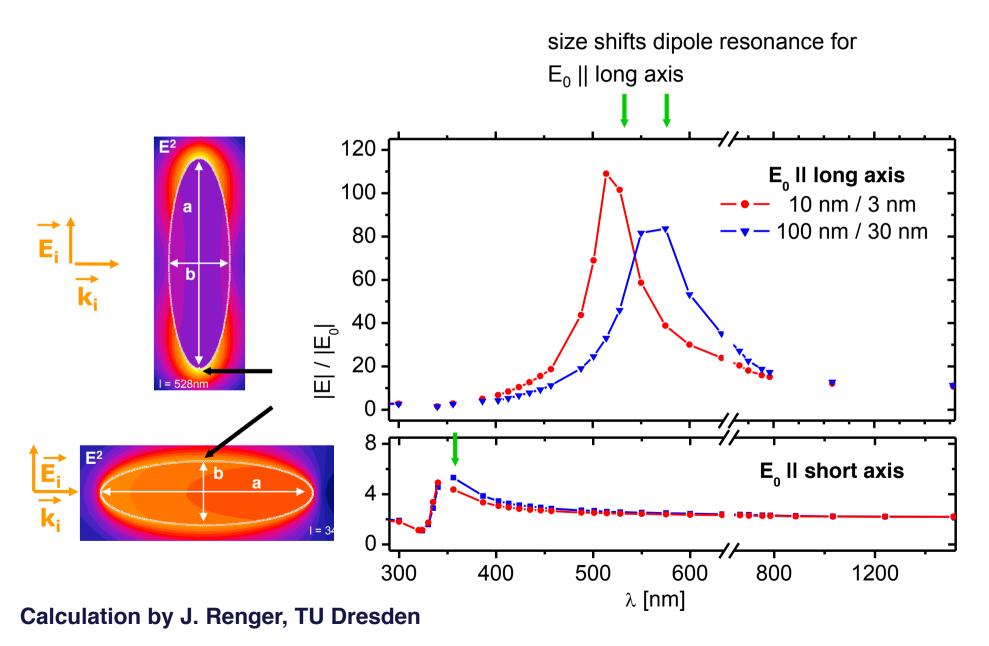
$$\alpha_{xyz} = \frac{4}{3} \pi abc \frac{\varepsilon - \varepsilon_m}{\varepsilon_m + L_{xyz}(\varepsilon - \varepsilon_m)}$$

geometrical factors

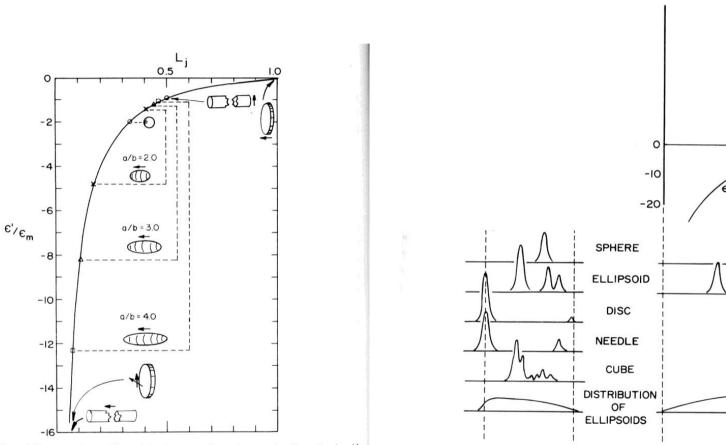
sphere: 
$$L_{x} = L_{y} = L_{z} = \frac{1}{3}$$

generally: 
$$L_x \neq L_y \neq L_z$$
  
 $\rightarrow$  3 resonances at $\varepsilon = \varepsilon_m \left( 1 - \frac{1}{L_{xyz}} \right)$ 

# Silver ellipsoid illuminated by a plane wave



### Plasmon resonances:dependence on the geometry



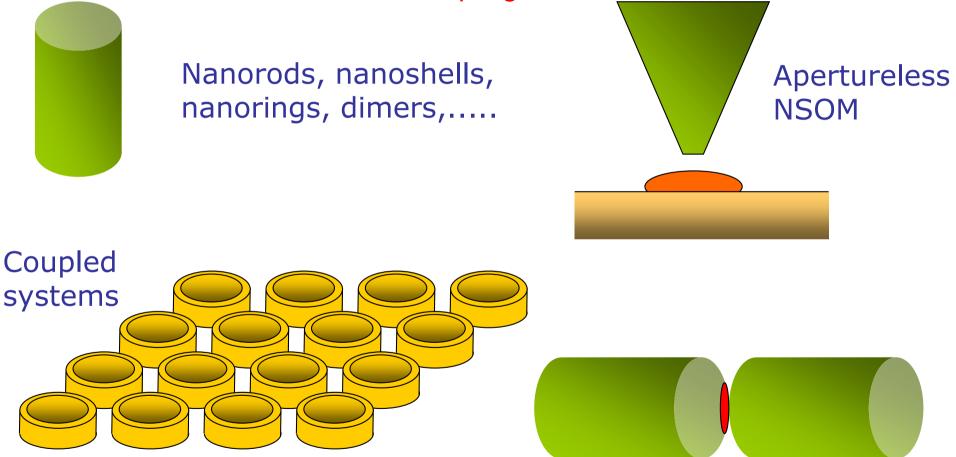
**Figure 12.5** Effect of shape on the position of the lowest-order surface mode of small spheroids. Arrows next to the various shapes show the direction of the electric field.

Figure 12.11 Surface mode frequencies for insulating and metallic particles of various shapes.

 $\omega_{\rm p}$ 

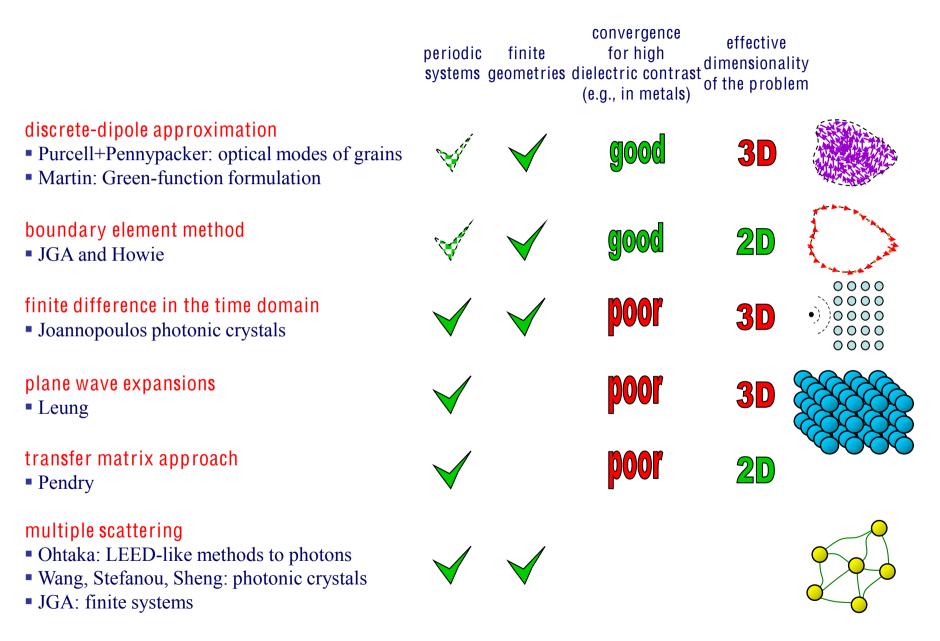
# More complex geometries

Control over the plasmon frequencies by playing with particle shapes and coupling



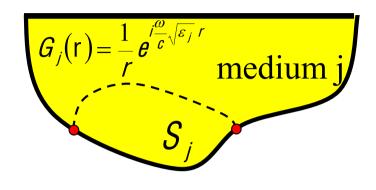
#### Nanometrology, sensing, spectroscopy

### Numerical solutions to the 3D electromagnetic problem



# **Boundary Element Method**

$$E(\mathbf{r}) = i\frac{\omega}{c} A(\mathbf{r}) - \nabla \phi(\mathbf{r})$$
$$A(\mathbf{r}) = A^{\text{ext}}(\mathbf{r}) + \int_{S_j} d\mathbf{s} \ G_j(\mathbf{r} - \mathbf{s}) \ h_j(\mathbf{s})$$
$$\phi(\mathbf{r}) = \phi^{\text{ext}}(\mathbf{r}) + \int_{S_i} d\mathbf{s} \ G_j(\mathbf{r} - \mathbf{s}) \ \sigma_j(\mathbf{s})$$



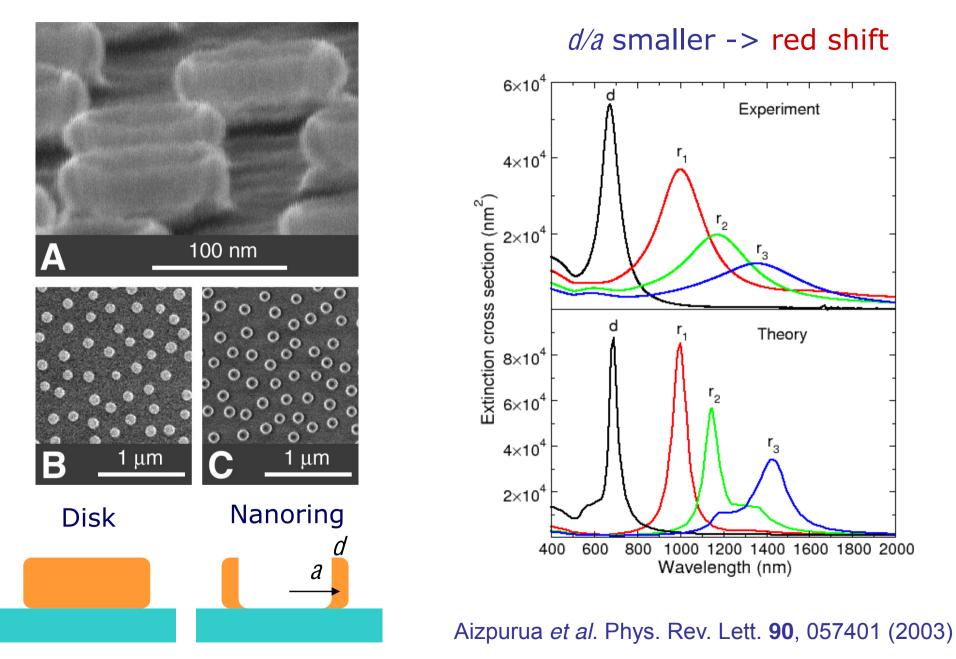
The boundary conditions lead to a set of surface integral equations with the interface currents  $\mathbf{h}_j$  and charges  $\sigma_j$  as variables. For example, the continuity of  $\phi$  leads to

$$\int_{S_j} \mathcal{O}\mathbf{S}' \left[ \mathcal{G}_1(\mathbf{S} - \mathbf{S'}) \ \sigma_1(\mathbf{S'}) - \mathcal{G}_2(\mathbf{S} - \mathbf{S'}) \ \sigma_2(\mathbf{S'}) \right] = \phi_2^{\text{ext}}(\mathbf{S}) - \phi_1^{\text{ext}}(\mathbf{S}),$$

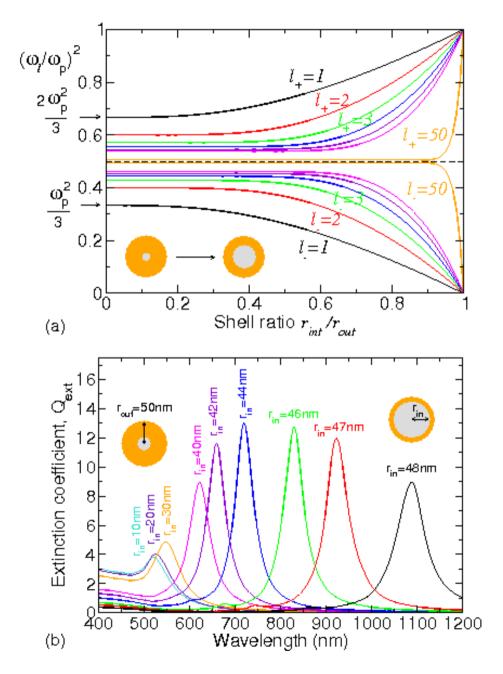
(1 and 2 refer to the interface sides). The surface integrals are now discretized using *N* representative points  $\mathbf{s}_i$ . This leads to a system of 8*N* linear equations with  $\mathbf{h}_1(\mathbf{s}_i)$ ,  $\mathbf{h}_2(\mathbf{s}_i)$ ,  $\sigma_1(\mathbf{s}_i)$ , and  $\sigma_2(\mathbf{s}_i)$  as unknowns.

García de Abajo and Aizpurua, PRB 56, 15873 (1997) García de Abajo and Howie, PRB 65, 115418 (2002)

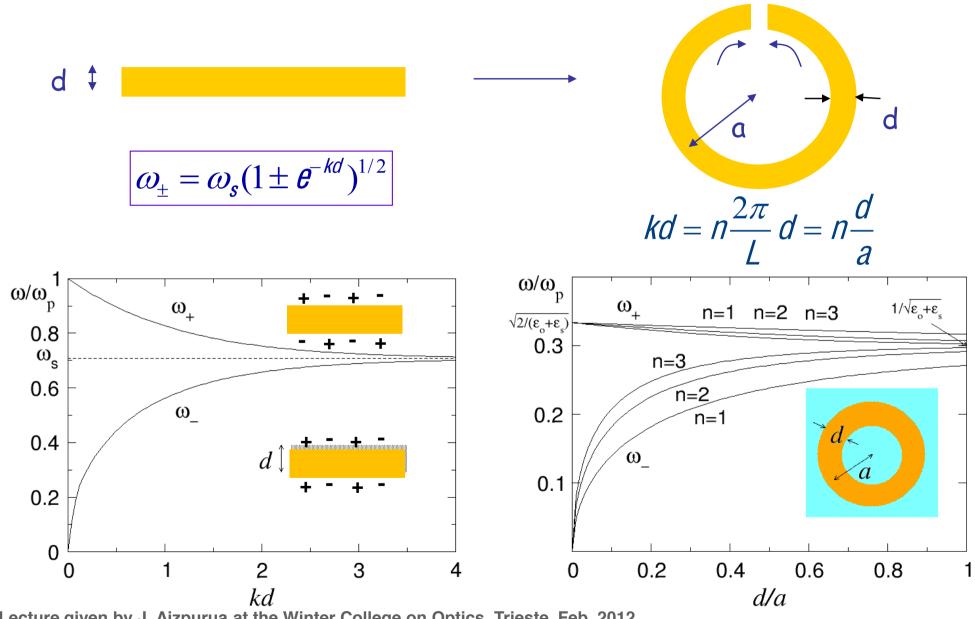
#### An example of Optical Antenna: Optical properties of metallic nanorings



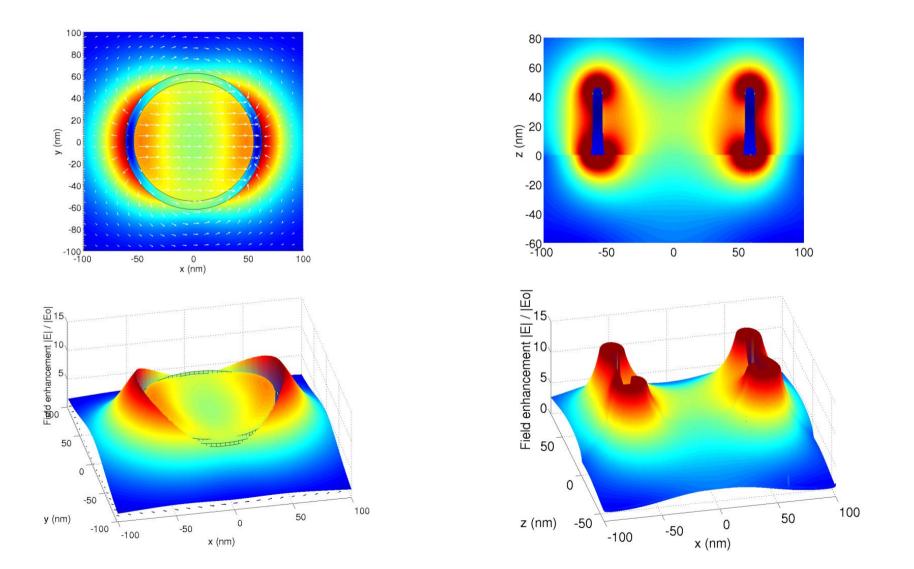
### Optical properties of metallic nanoshells



### Modes in a nanoring. The twisted slab



# Field-enhancement in a nanoring



Lecture given by J. Aizpurua at the Winter College on Optics, Trieste, Feb. 2012

#### **Radio Frequency Antennas**



# Half wave dipole antennas



#### **Biconical antenna**



#### Monopole antenna



#### **Combilog** antenna



### Yagi-Uda antenna



#### Horn antenna

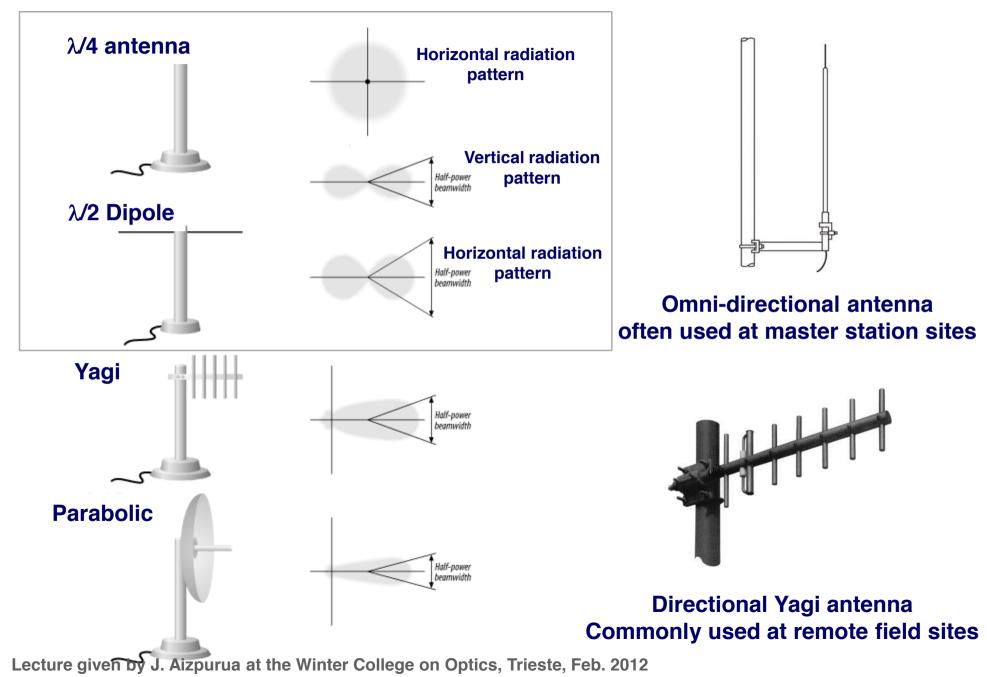


#### Parabolic antenna

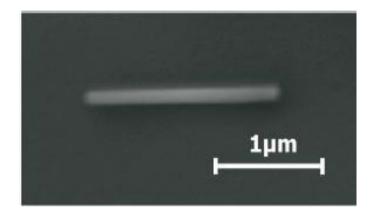


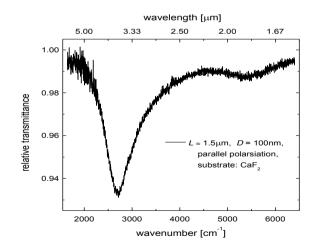
#### Active loop antenna

# Different types of radio antennas

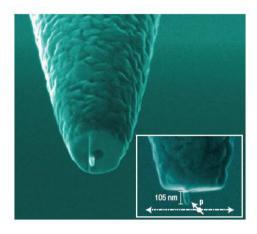


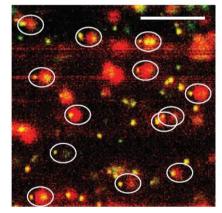
### $\lambda/2$ nanoantenna





### $\lambda/4$ optical antenna

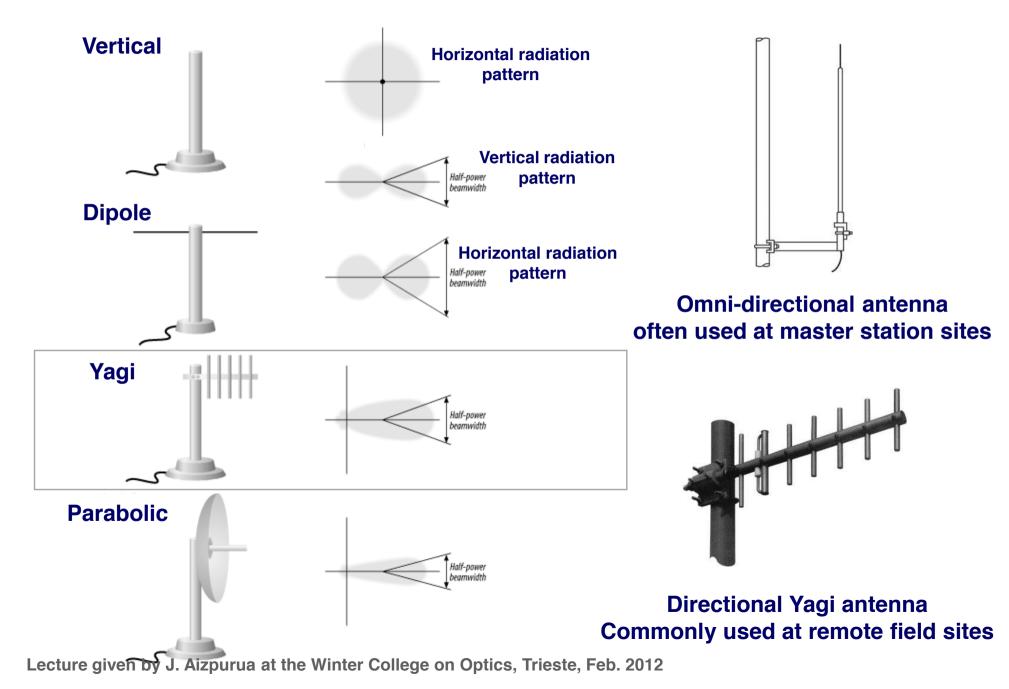




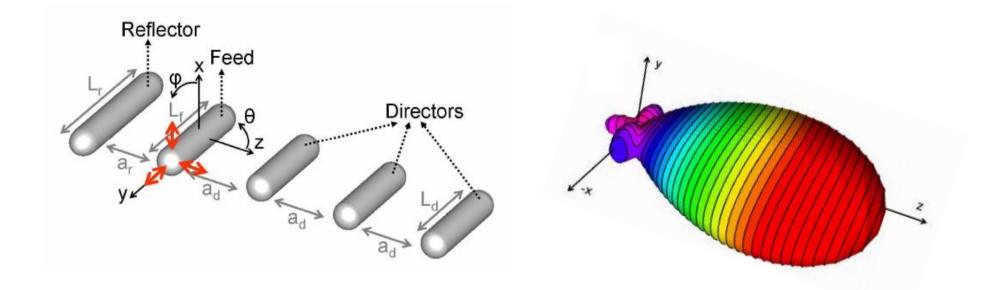
#### Neubrech *et al., App. Phys. Lett. 89, 253104 (2006)*

#### Taminiau *et al., Nature Photonics 2, 234 (2008)*

# Different types of radio antennas

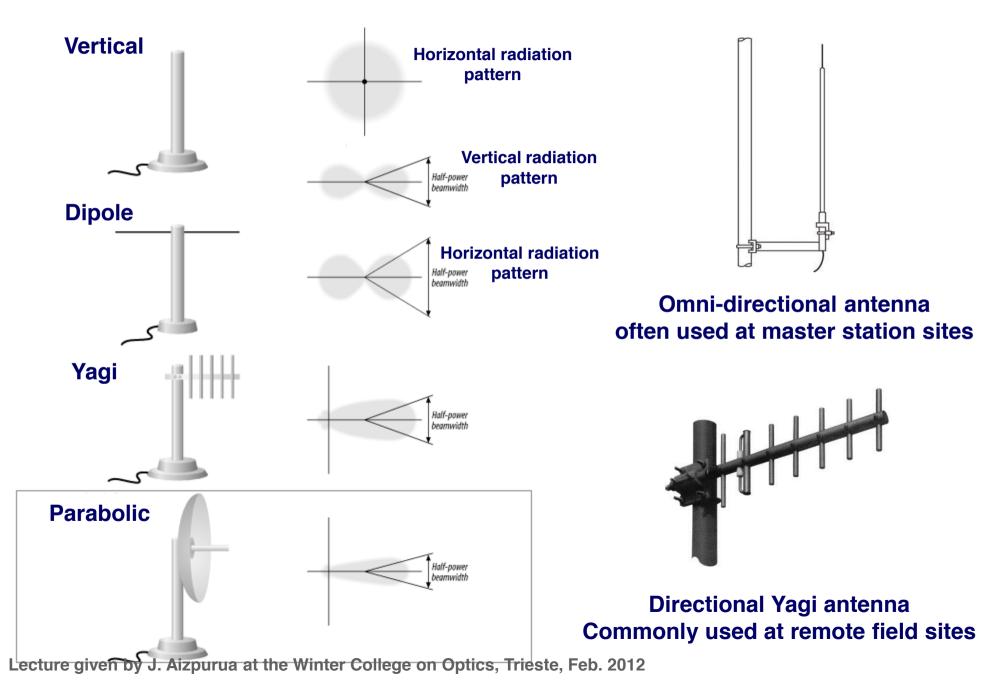


### Yagi-Uda antenna

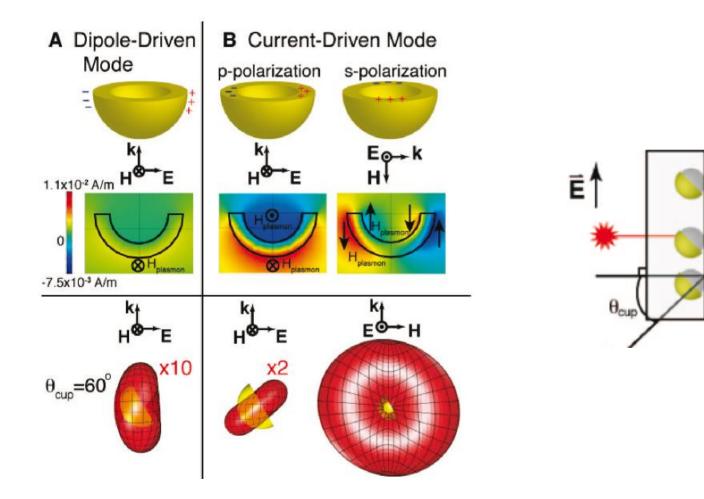


#### Taminiau *et al., Optics Express 14, 10858 (2008)*

# Different types of radio antennas



### Parabolic-like optical nanoantennas

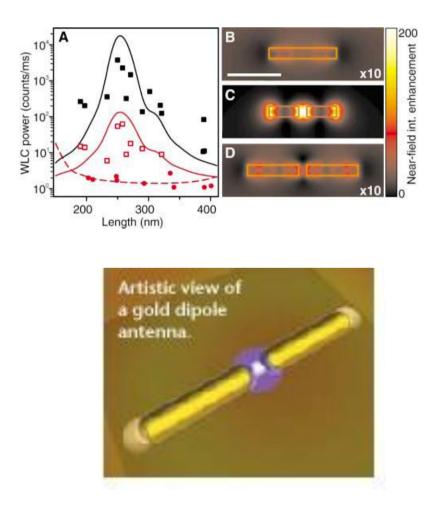


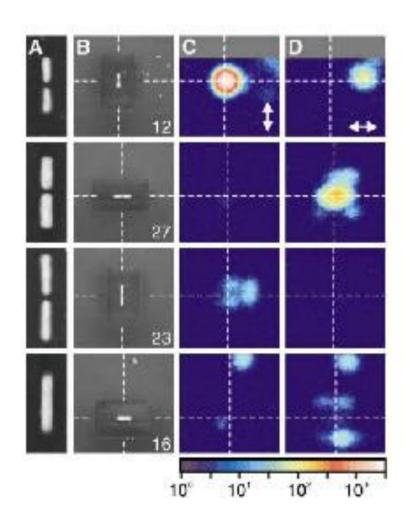
N. Mirin and N. Halas, Nano Letters 9, 1255 (2009)

# **Resonant Optical Antennas**

P. Mühlschlegel,<sup>1</sup> H.-J. Eisler,<sup>1</sup> O. J. F. Martin,<sup>2</sup> B. Hecht,<sup>1\*</sup> D. W. Pohl<sup>1</sup>

SCIENCE VOL 308 10 JUNE 2005

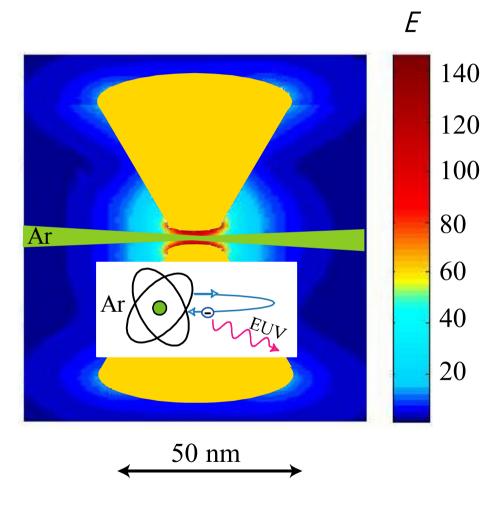




### $\lambda/4$ optical antenna

105 nm

### **Bowtie antennas**

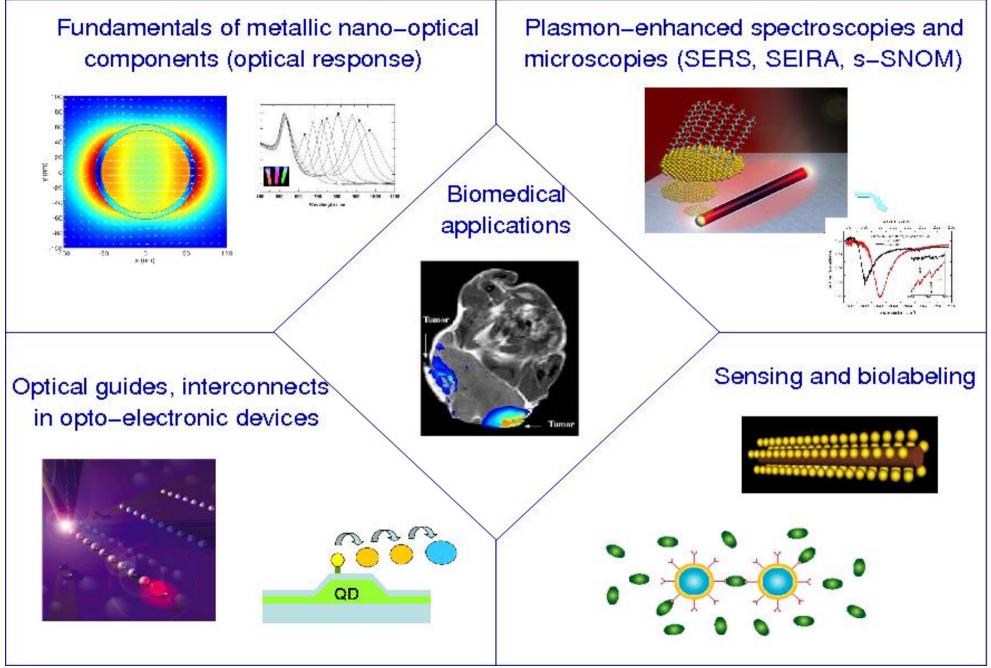


#### Taminiau *et al., Nature Photonics 2, 234 (2008)*

Nature 453, 731 (2008)

Lecture given by J. Aizpurua at the Winter College on Optics, Trieste, Feb. 2012

### **Plasmonic antennas**



### Thank you for your attention!

http://cfm.ehu.es/nanophotonics