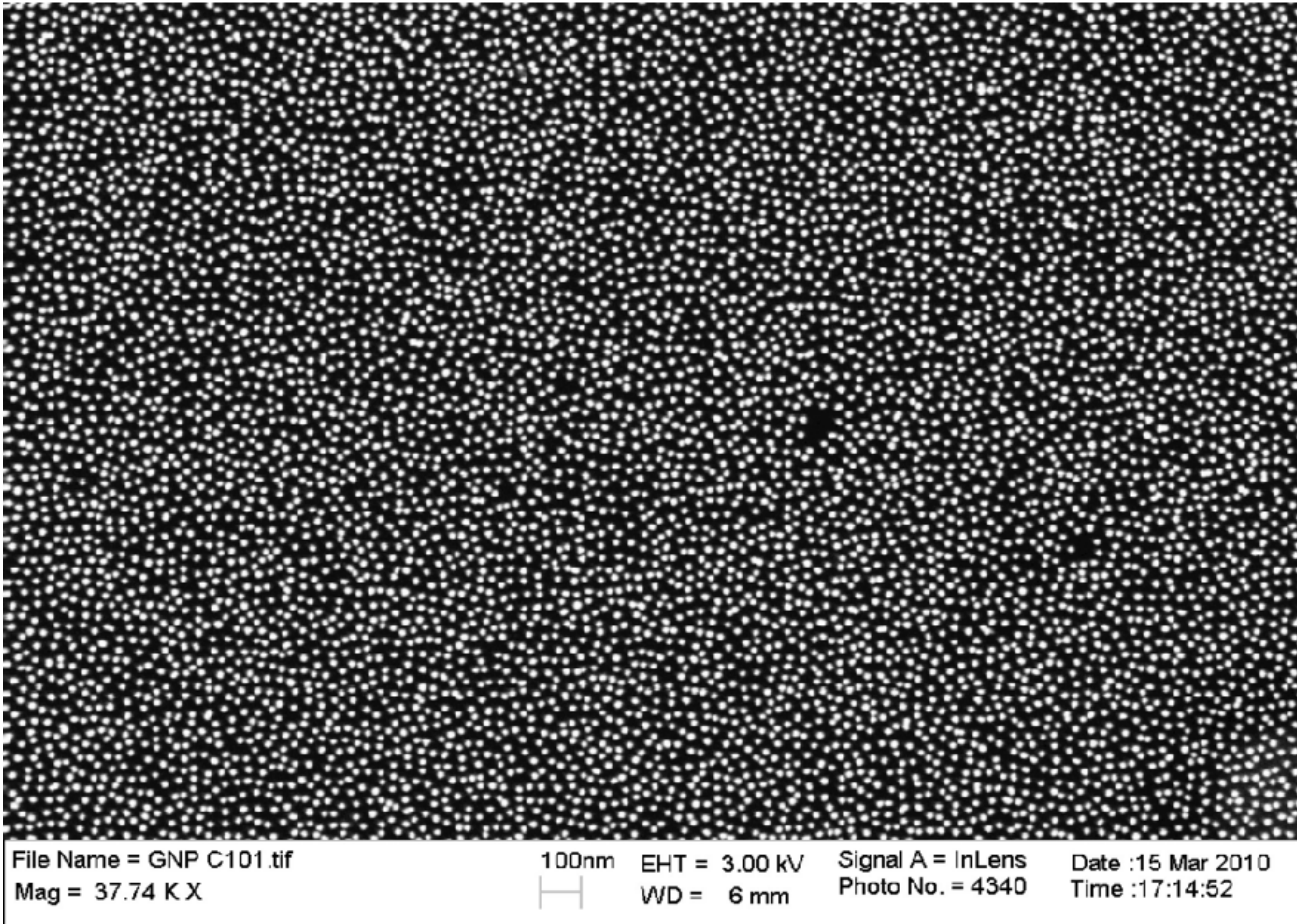
Computational Photonics

Scattering theory: quasi-static sphere

Nanosphere



(courtesy of T. Bürgi, University Geneva)

Quasi-static regime

- in the quasi-static regime phase variations of the field across the surface can be neglected $\Longrightarrow$ electrostatics, no magnetic field
- temporarily the fields still oscillate according to $e^{-i\omega t}$

introducing a scalar potential: $$\mathbf{E}(\mathbf{r}) = -\nabla\Phi(\mathbf{r})$$

has to be a solution to Laplace equation: (absence of charges)

$$\triangle\Phi = 0$$

e.g. spherical coordinates:

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

Quasi-static regime

- general solution of this equation is given by:
  (solved by separation of variables the same way as before)

$$\Phi = \sum_{n=0}^{\infty} \sum_{m=0}^{n} \left(a_{nm} r^n + \frac{b_{nm}}{r^{n+1}} \right) P_n^m(\cos\theta) e^{im\phi}$$

$P_n^m(x)$ associated Legendre polynom

- introducing three potentials:

ϵ_2 Φ_0 illumination

Φ_S scattered

ϵ_1 Φ_1 inside

$\Longrightarrow$

$$\Phi_2 = \Phi_S + \Phi_0$$

Quasi-static regime

- z-polarised incident wave is assumed to be constant in the vicinity of the coordinate center *(quasi-static)*

$$\Phi_0 = -E_0 z = -E_0 r P_1^0 (\cos\theta)$$

- two boundary conditions have to be used to determine a_{nm} and b_{nm}

continuity of tangential electric field

$$\left.\frac{\partial \Phi_1}{\partial \theta}\right|_{r=R} = \left.\frac{\partial \Phi_2}{\partial \theta}\right|_{r=R}$$

continuity of normal dielectric displacement

$$\epsilon_1 \left.\frac{\partial \Phi_1}{\partial r}\right|_{r=R} = \epsilon_2 \left.\frac{\partial \Phi_2}{\partial r}\right|_{r=R}$$

Quasi-static regime

$$\Phi = \sum_{n=0}^{\infty} \sum_{m=0}^{n} \left(a_{nm} r^n + \frac{b_{nm}}{r^{n+1}} \right) P_n^m(\cos\theta) e^{im\phi}$$

$$\Phi_0 = -E_0 z = -E_0 r P_1^0(\cos\theta)$$

- in the inner domain all b_{nm} coefficients are zero as the potential has to remain finite
- in the outer domain all a_{nm} coefficients are zero as the potential has to remain finite
    - illuminating field has only a nonzero component for n=1 and m=0
- other components remain zero, hence 2 equations with 2 unknowns

Quasi-static regime

$$\Phi_1 = -\frac{3\epsilon_2}{\epsilon_1 + 2\epsilon_2} E_0 r \cos\theta$$

$$\Phi_2 = -E_0 r \cos\theta + \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2} R^3 E_0 \frac{\cos\theta}{r^2}$$

$$\mathbf{E}_1 = \frac{3\epsilon_2}{\epsilon_1 + 2\epsilon_2} E_0 \mathbf{e}_z$$

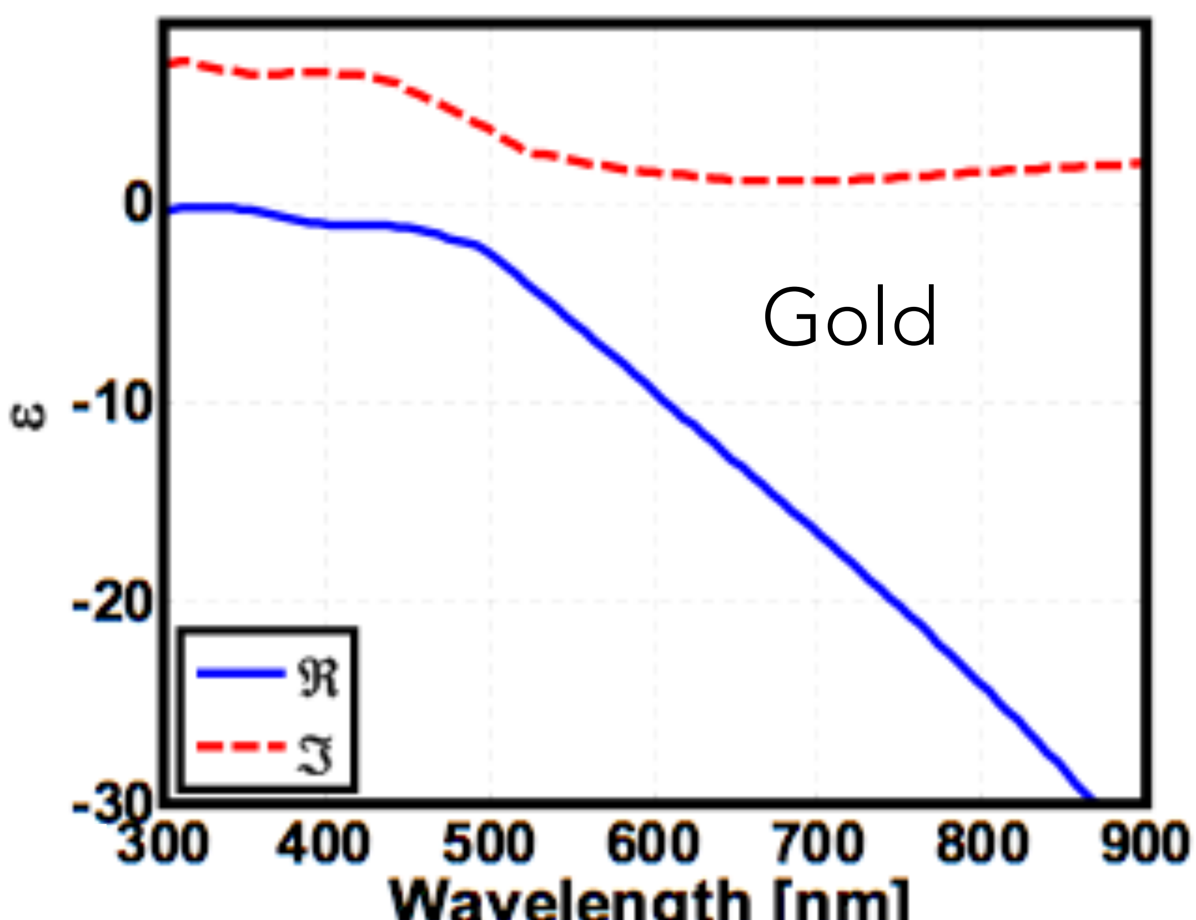
$$\mathbf{E}_2 = E_0 \mathbf{e}_z + \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2} \frac{R^3}{r^3} E_0 (2\cos\theta \mathbf{e}_r + \sin\theta \mathbf{e}_\theta)$$

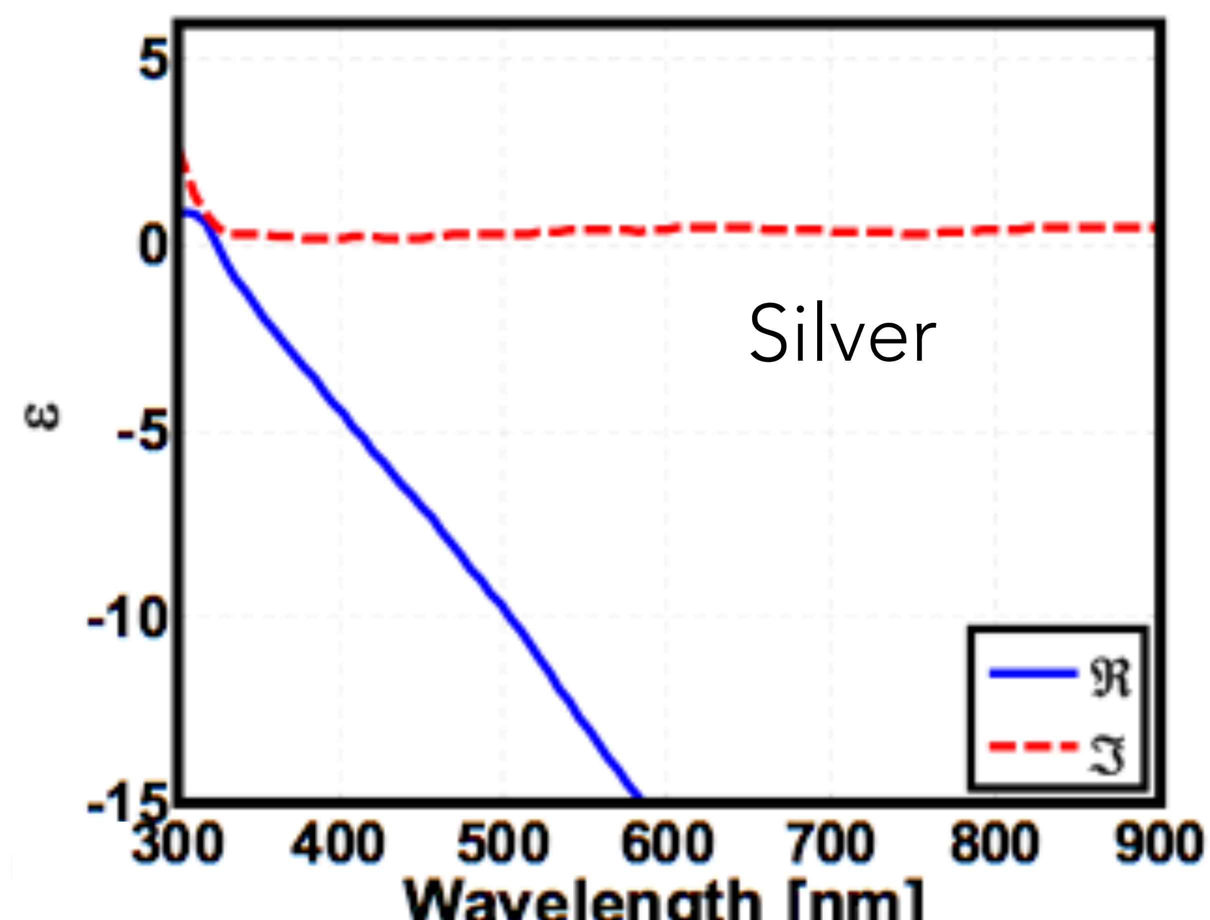
Quasi-static regime

- scattered field corresponds to the field of a dipole with a moment $\mathbf{p}$

$$\mathbf{p} = \epsilon_2 \alpha E_0 \mathbf{e}_z$$

Polarisability: $$\alpha = 4\pi R^3 \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}$$





Quantities of interest

scattering cross section

time average of energy flow is expressed by the real part of the Poynting vector

$$\bar{\mathbf{S}} = \frac{1}{2}\Re\left(\mathbf{E}\times\mathbf{H}^*\right) = \frac{1}{2}\Re\left(E_\theta H_\phi^*\mathbf{e}_r - E_r H_\phi^*\mathbf{e}_\theta\right)$$

total radiation power

$$W = \int_0^{2\pi}\int_0^{\pi} \mid\bar{\mathbf{S}}\mid r^2 \sin\theta \mathrm{d}\theta\mathrm{d}\phi = \frac{\omega k^3 \mid \mathbf{p}\mid^2}{12\pi\epsilon_2} = \frac{\omega k^3\epsilon_2}{12\pi}\mid\alpha\mid^2 E_0^2$$

intensity of the incident field

$$\bar{S}_0 = \frac{\omega\epsilon_2}{2k}E_0^2$$

Quantities of interest

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$$C_{\mathrm{sca}} = \frac{W}{\bar{S}_0} = \frac{k^4}{6\pi^2} \mid \alpha \mid^2$$

absorption cross section

$$C_{\mathrm{abs}} = k\Im(\alpha)$$

extinction cross section

$$C_{\mathrm{ext}} = C_{\mathrm{sca}} + C_{\mathrm{abs}}$$

Quasi-static regime

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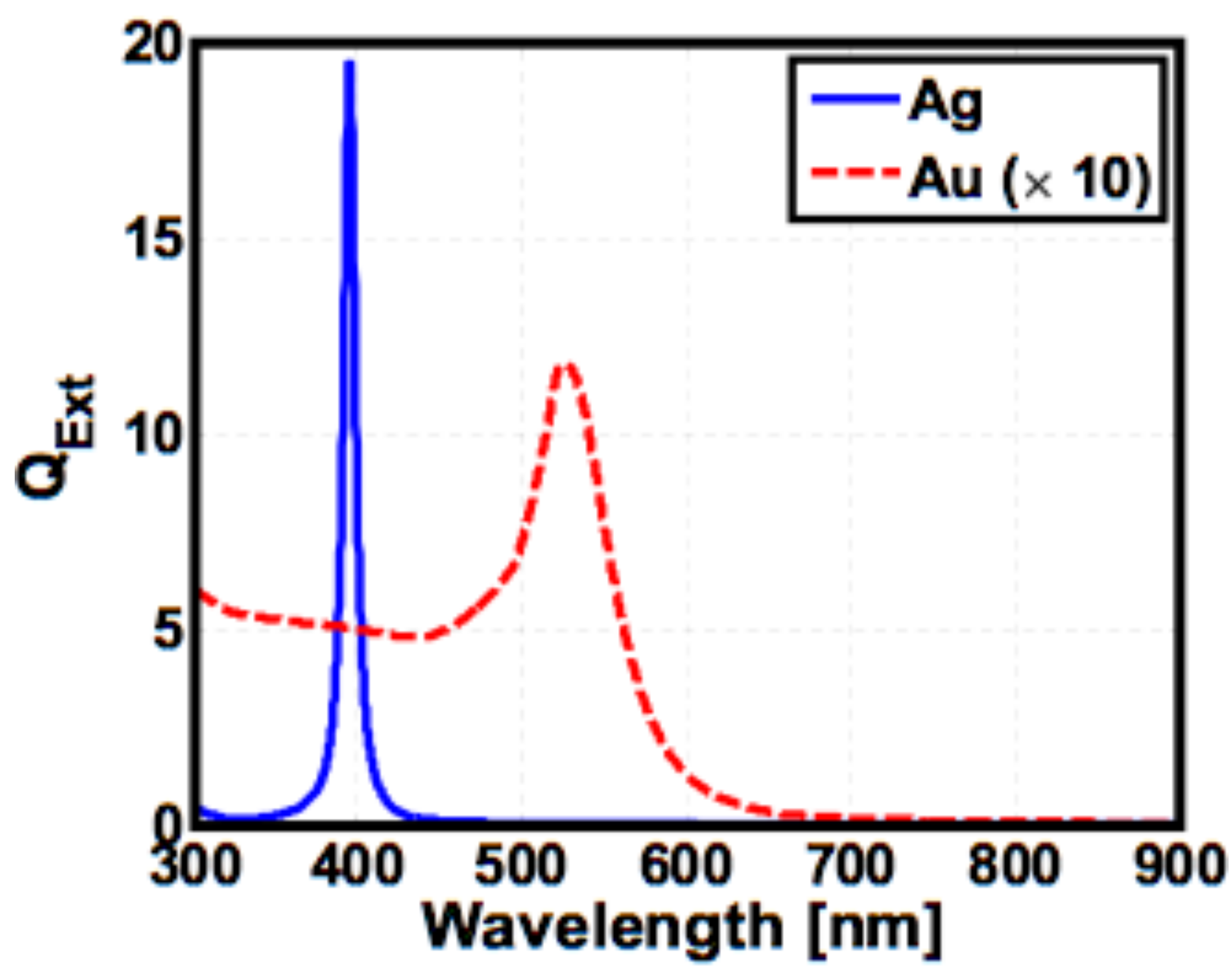

Quasi-static regime

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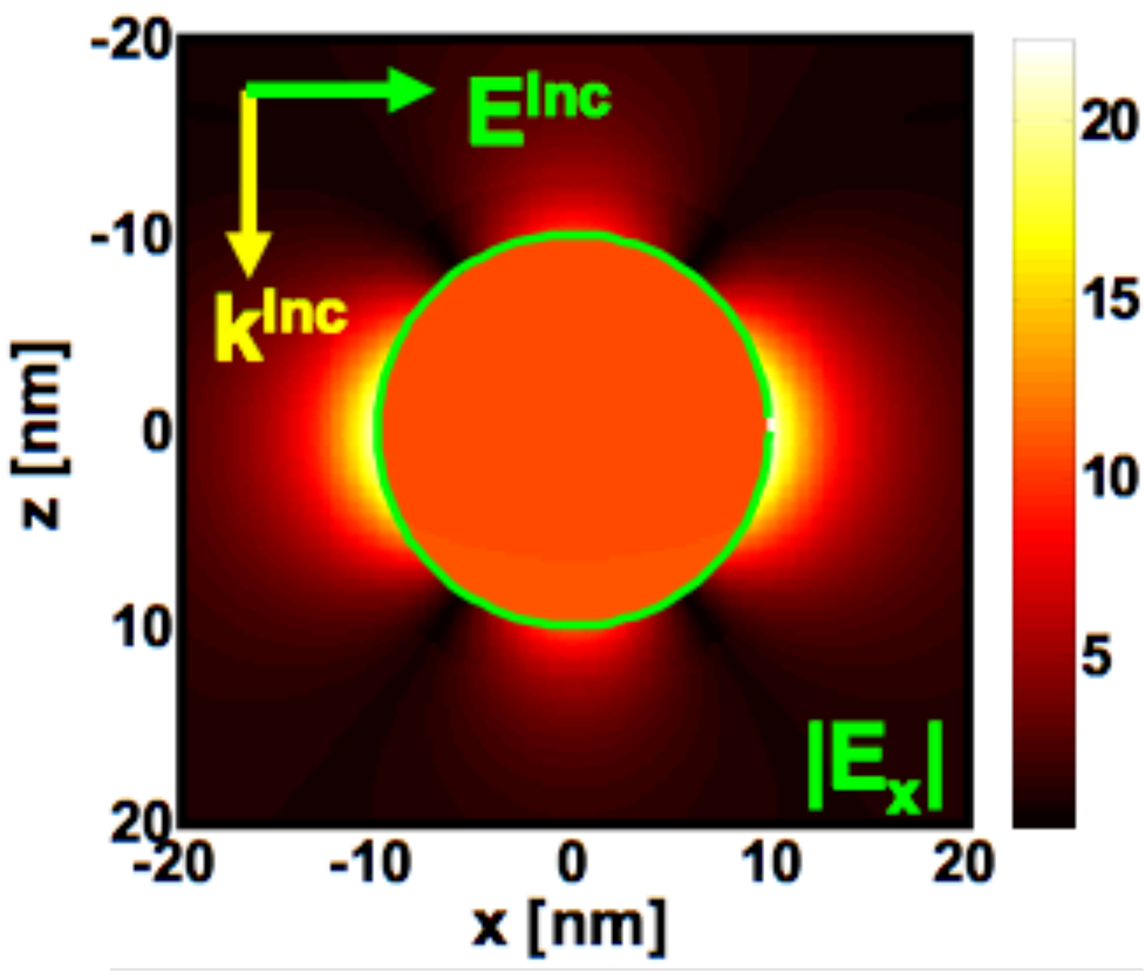
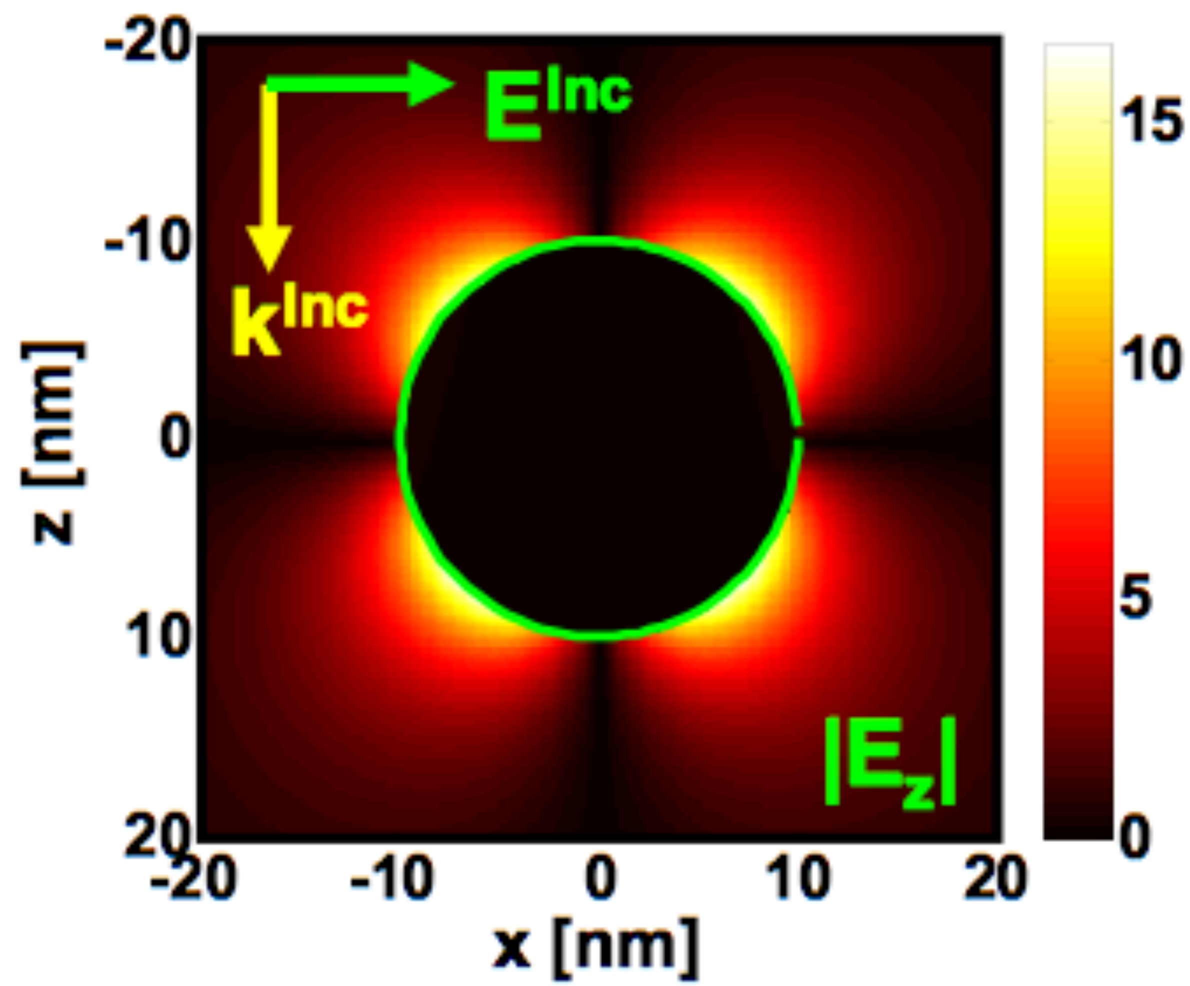




taken from
2020science.org

Strangeness

Lycurgus Cup

Behaves in unexpected ways

Field induced by oscillating dipole

- have ignored time dependence of the external field

$$\mathbf{E} = \frac{1}{4\pi\epsilon_2}\left(-\frac{\mathbf{p}(t_0)}{r^3} + \frac{3\mathbf{r}\left[\mathbf{r}\cdot\mathbf{p}(t_0)\right]}{r^5} + \frac{ik\mathbf{p}(t_0)}{r^2} - \frac{3ik\mathbf{r}\left[\mathbf{r}\cdot\mathbf{p}(t_0)\right]}{r^4} - \frac{k^2}{r^3}\mathbf{r}\times\left[\mathbf{r}\times\mathbf{p}(t_0)\right]\right)$$

$$t_0 = t - kr/\omega$$

- first two terms in are the static field and the scattering field $\left[\propto 1/r^3\right]$
- next two terms, called the induction field, are the field induced by the current due to the dipole oscillation $\left[\propto 1/r^2\right]$
    - last term is called the radiation field $\left[\propto 1/r\right]$

$$\mathbf{H} = -\frac{i\omega}{4\pi}\left(\frac{1}{r^2} - \frac{ik}{r}\right)\mathbf{r}\times\mathbf{p}(t_0)$$

Computational Photonics

Scattering theory: quasi-static sphere

Computational Photonics

Scattering theory: fully dynamic sphere

Mie theory for spheres

$$\nabla^2 \mathbf{E} + k^2 \mathbf{E} = 0 \qquad \nabla \cdot \mathbf{E} = 0$$
$$\nabla^2 \mathbf{H} + k^2 \mathbf{H} = 0 \qquad \nabla \cdot \mathbf{H} = 0$$

$\Rightarrow$ reducing the full vectorial to a scalar problem

from algebra we know that a vector can be construct from a scalar function and an arbitrary constant vector via

$$\mathbf{M} = \nabla \times (\mathbf{c}\psi)$$

divergence of a curl of any vector function vanishes

$$\nabla \cdot \mathbf{M} = 0$$

using vector identities it can be shown that

$$\nabla^2 \mathbf{M} + k^2 \mathbf{M} = \nabla \times \left[\mathbf{c}\left(\nabla^2 \psi + k^2 \psi\right)\right]$$

$\Rightarrow$ ψ needs to satisfy a scalar wave equation

Mie theory for spheres

→ can construct a second vector function $\mathbf{N} = \dfrac{\nabla \times \mathbf{M}}{k}$

→ $\nabla \cdot \mathbf{N} = 0 \qquad \nabla^2 \mathbf{N} + k^2 \mathbf{N} = 0 \qquad \nabla \times \mathbf{N} = k\mathbf{M}$

→ $\mathbf{M}$ and $\mathbf{N}$ have all the required properties of an electromagnetic field

→ free of divergence

→ the curl of $\mathbf{M}$ is proportional to $\mathbf{N}$

→ the curl of $\mathbf{N}$ is proportional to $\mathbf{M}$

→ finding a solution to the scalar wave equation for ψ instead of a field equation

$\mathbf{M} = \nabla \times (\mathbf{r}\psi)$ → wave equation in spherical polar coordinates

Scalar solutions to wave equation

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial \psi}{\partial r}\right)+\frac{1}{r^2\sin\theta}\frac{\partial}{\partial \theta}\left(\sin\theta\frac{\partial \psi}{\partial \theta}\right)+\frac{1}{r^2\sin\theta^2}\frac{\partial^2 \psi}{\partial \phi^2}+k_m^2\psi=0$$

seek a particular solution of the form

$$\psi(r,\theta,\phi)=R(r)\Theta(\theta)\Phi(\phi)$$

and performing a separation of variables procedure

$$\psi_{nm}\propto Z_n^{(J)}(k_m r)P_n^m(\cos\theta)\,e^{im\phi}$$

different Bessel functions radial dependency

$$Z_n^{(1)}(z)=j_n(z)=\sqrt{\frac{\pi}{2z}}J_{n+0.5}(z)$$

$$Z_n^{(2)}(z)=y_n(z)=\sqrt{\frac{\pi}{2z}}Y_{n+0.5}(z)$$

$$Z_n^{(3)}(z)=h_n^1(z)=j_n(z)+iy_n(z)$$

$$Z_n^{(4)}(z)=h_n^2(z)=j_n(z)-iy_n(z)$$

angular dependency by associated Legendre function of the first kind and of degree n and order m

$$P_n^m(\cos\theta)=\frac{\left(1-\cos^2\theta\right)^{\frac{m}{2}}}{2^n n!}\frac{d^{n+m}\left(\cos^2\theta-1\right)^n}{d\left(cos\theta\right)^{n+m}}$$

Vector solutions to wave equation

vector harmonics can be constructed via

$$\mathbf{M}_{nm} = \nabla \times (\mathbf{r}\psi_{nm}) \qquad \mathbf{N}_{nm} = \frac{\nabla \times \mathbf{M}_{nm}}{k}$$

plugging this solution into the vector solution provide

$$\mathbf{M}_{nm}^{(J)}(r,\theta,\varphi) = \left[i\pi_{nm}(\cos\theta)\hat{\mathbf{e}}_\theta - \tau_{nm}(\cos\theta)\hat{\mathbf{e}}_\varphi\right]\Psi_n^{(J)}(kr)e^{im\varphi}$$

$$\mathbf{N}_{nm}^{(J)}(r,\theta,\varphi) = n(n+1)P_n^m(\cos\theta)\frac{\Psi_n^{(J)}(kr)}{kr}e^{im\varphi}\hat{\mathbf{e}}_r + \left[\tau_{nm}(\cos\theta)\hat{\mathbf{e}}_\theta + i\pi_{nm}(\cos\theta)\hat{\mathbf{e}}_\varphi\right]\frac{1}{kr}\frac{d}{dr}\left[r\Psi_n^{(J)}(kr)\right]e^{im\varphi}$$

$$\tau_{nm}(\cos\theta) = \frac{d}{d\theta}P_n^m(\cos\theta) \qquad \pi_{nm}(\cos\theta) = \frac{m}{\sin\theta}P_n^m(\cos\theta)$$

Field expansion for scattered and internal field

$$\mathbf{E}_{\rm inc}(\mathbf{r},\omega) = -\sum_{n=1}^{\infty}\sum_{m=-n}^{n} iE_{nm}\left[p_{nm}(\omega)\mathbf{N}_{nm}^{(1)}(\mathbf{r},\omega) + q_{nm}(\omega)\mathbf{M}_{nm}^{(1)}(\mathbf{r},\omega)\right]$$

$$\mathbf{H}_{\rm inc}(\mathbf{r},\omega) = -\frac{1}{Z}\sum_{n=1}^{\infty}\sum_{m=-n}^{n} E_{nm}\left[q_{nm}(\omega)\mathbf{N}_{nm}^{(1)}(\mathbf{r},\omega) + p_{nm}(\omega)\mathbf{M}_{nm}^{(1)}(\mathbf{r},\omega)\right]$$

$$\mathbf{E}_{\rm sca}(\mathbf{r},\omega) = \sum_{n=1}^{\infty}\sum_{m=-n}^{n} iE_{nm}\left[a_{nm}(\omega)\mathbf{N}_{nm}^{(3)}(\mathbf{r},\omega) + b_{nm}(\omega)\mathbf{M}_{nm}^{(3)}(\mathbf{r},\omega)\right]$$

$$\mathbf{H}_{\rm sca}(\mathbf{r},\omega) = \frac{1}{Z}\sum_{n=1}^{\infty}\sum_{m=-n}^{n} E_{nm}\left[b_{nm}(\omega)\mathbf{N}_{nm}^{(3)}(\mathbf{r},\omega) + a_{nm}(\omega)\mathbf{M}_{nm}^{(3)}(\mathbf{r},\omega)\right]$$

$$\mathbf{E}_{\rm int}(\mathbf{r},\omega) = \sum_{n=1}^{\infty}\sum_{m=-n}^{n} iE_{nm}\left[c_{nm}(\omega)\mathbf{N}_{nm}^{(1)}(\mathbf{r},\omega) + d_{nm}(\omega)\mathbf{M}_{nm}^{(1)}(\mathbf{r},\omega)\right]$$

$$\mathbf{H}_{\rm int}(\mathbf{r},\omega) = \frac{1}{Z_2}\sum_{n=1}^{\infty}\sum_{m=-n}^{n} E_{nm}\left[d_{nm}(\omega)\mathbf{N}_{nm}^{(1)}(\mathbf{r},\omega) + c_{nm}(\omega)\mathbf{M}_{nm}^{(1)}(\mathbf{r},\omega)\right]$$

$$E_{nm} = |\mathbf{E}_0|\, i^n(2n+1)\frac{(n-m)!}{(n+m)!}$$

Boundary conditions

tangential electric and magnetic fields continuous at interface

$$E_{\mathrm{inc},\theta}(R) + E_{\mathrm{sca},\theta}(R) = E_{\mathrm{int},\theta}(R) \quad H_{\mathrm{inc},\theta}(R) + H_{\mathrm{sca},\theta}(R) = H_{\mathrm{int},\theta}(R)$$

$$E_{\mathrm{inc},\phi}(R) + E_{\mathrm{sca},\phi}(R) = E_{\mathrm{int},\phi}(R) \quad H_{\mathrm{inc},\phi}(R) + H_{\mathrm{sca},\phi}(R) = H_{\mathrm{int},\phi}(R)$$

with the expansions and relying on the orthogonality

(the prime indicates a differentiation with respect to the argument in parentheses)

$$a_{nm} = a_n p_{nm} \qquad b_{nm} = b_n q_{nm}$$

Mie coefficients for the scattered field

$$a_n = \frac{\mu\eta^2 j_n(\eta x)[x j_n(x)]' - \mu_{\mathrm{sph}} j_n(x)[\eta x j_n(\eta x)]'}{\mu\eta^2 j_n(\eta x)[x h_n^{(1)}(x)]' - \mu_{\mathrm{sph}} h_n^{(1)}(x)[\eta x j_n(\eta x)]'}$$

$$b_n = \frac{\mu_{\mathrm{sph}} j_n(\eta x)[x j_n(x)]' - \mu j_n(x)[\eta x j_n(\eta x)]'}{\mu_{\mathrm{sph}} j_n(\eta x)[x h_n^{(1)}(x)]' - \mu h_n^{(1)}(x)[\eta x j_n(\eta x)]'}$$

$$x = \frac{\omega}{c}\sqrt{\varepsilon(\omega)\mu(\omega)}a \qquad \eta = \sqrt{\frac{\varepsilon_{\mathrm{sph}}(\omega)\mu_{\mathrm{sph}}(\omega)}{\varepsilon(\omega)\mu(\omega)}}$$

(Some) derived quantities

scattering cross section

$$C_{\text{sca}} = \frac{4\pi}{k^2} \sum_{n=1}^{\infty} \sum_{m=-n}^{n} n(n+1)(2n+1) \frac{(n-m)!}{(n+m)!} \left(|a_{nm}|^2 + |b_{nm}|^2\right)$$

extinction cross section

$$C_{\text{ext}} = \frac{4\pi}{k^2} \sum_{n=1}^{\infty} \sum_{m=-n}^{n} n(n+1)(2n+1) \frac{(n-m)!}{(n+m)!} \Re(p_{nm}^* a_{nm} + q_{nm}^* b_{nm})$$

absorption cross section

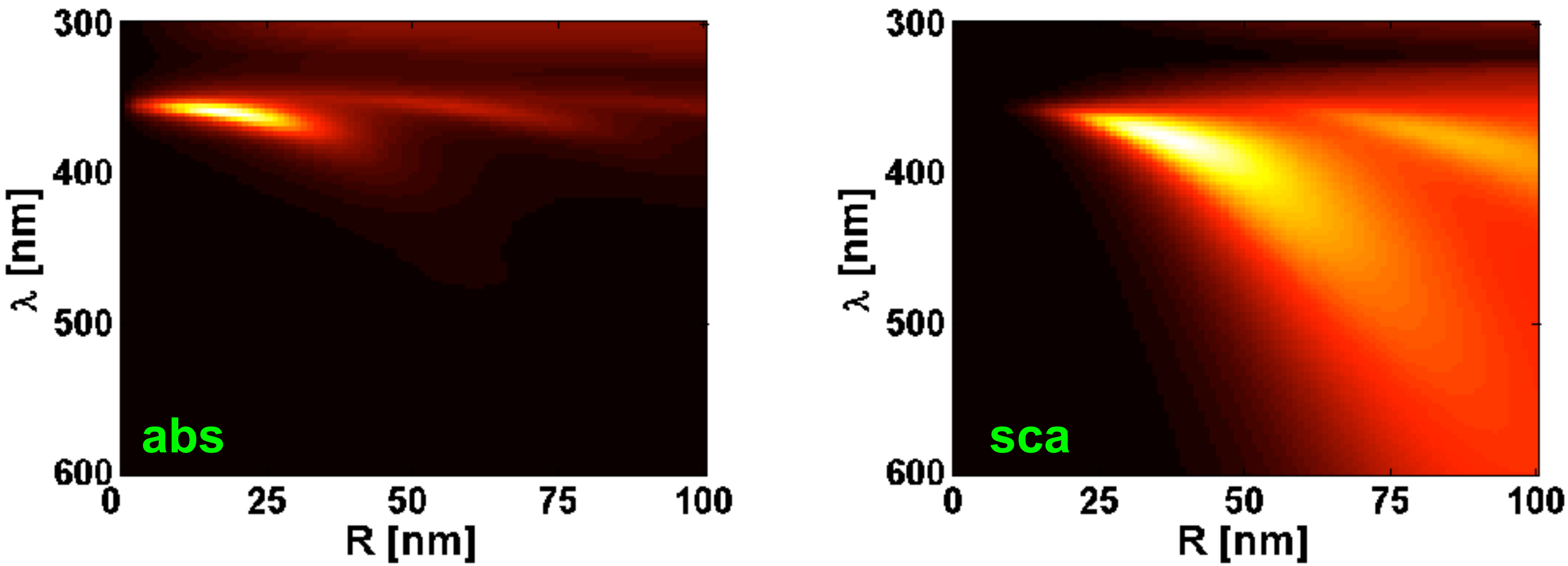
$$C_{\text{abs}} = C_{\text{ext}} - C_{\text{sca}}$$

suggested expansion order

$$N = x + 4x^{1/3} + 2$$

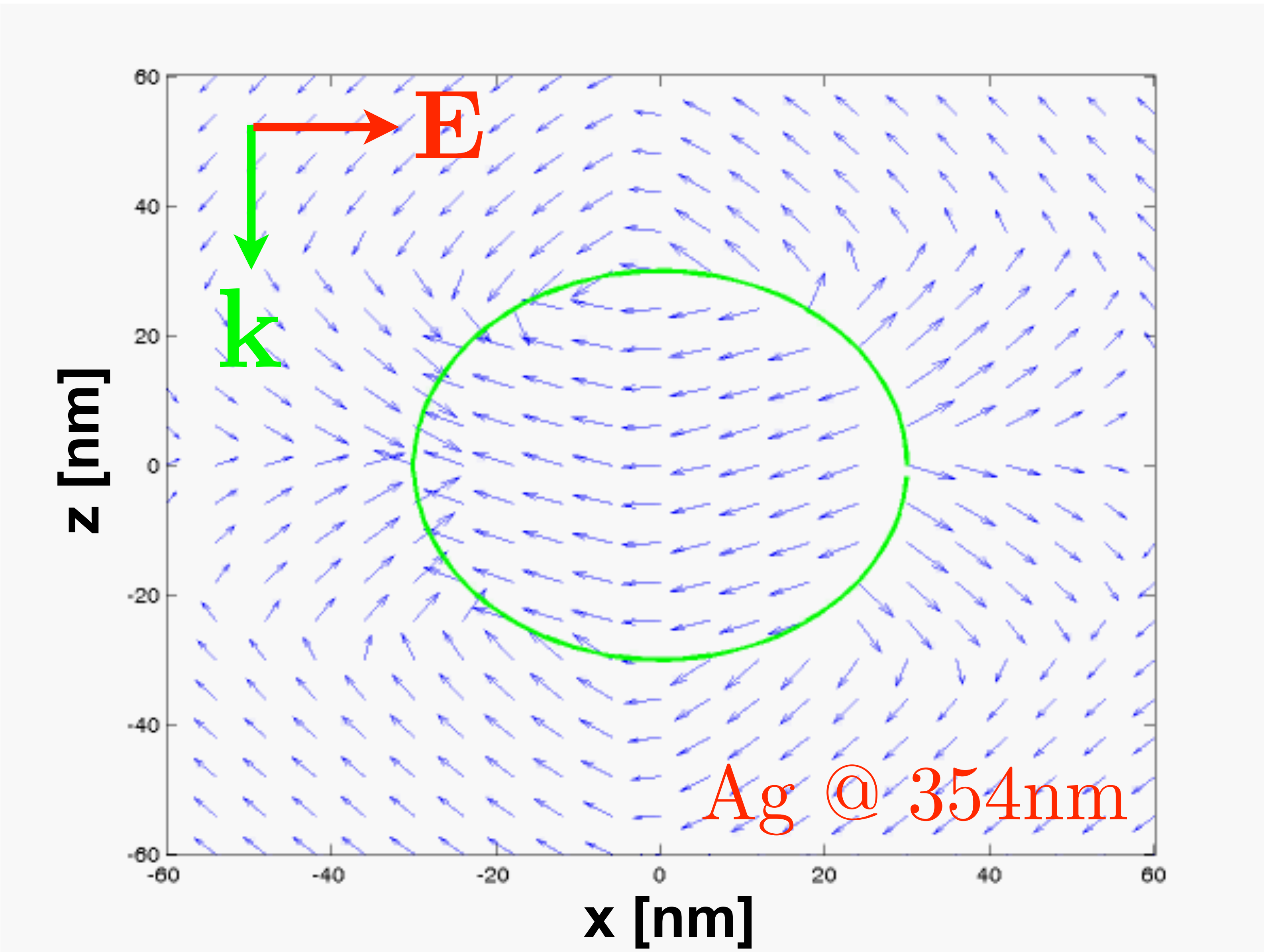
Metallic nanoparticles - size effects

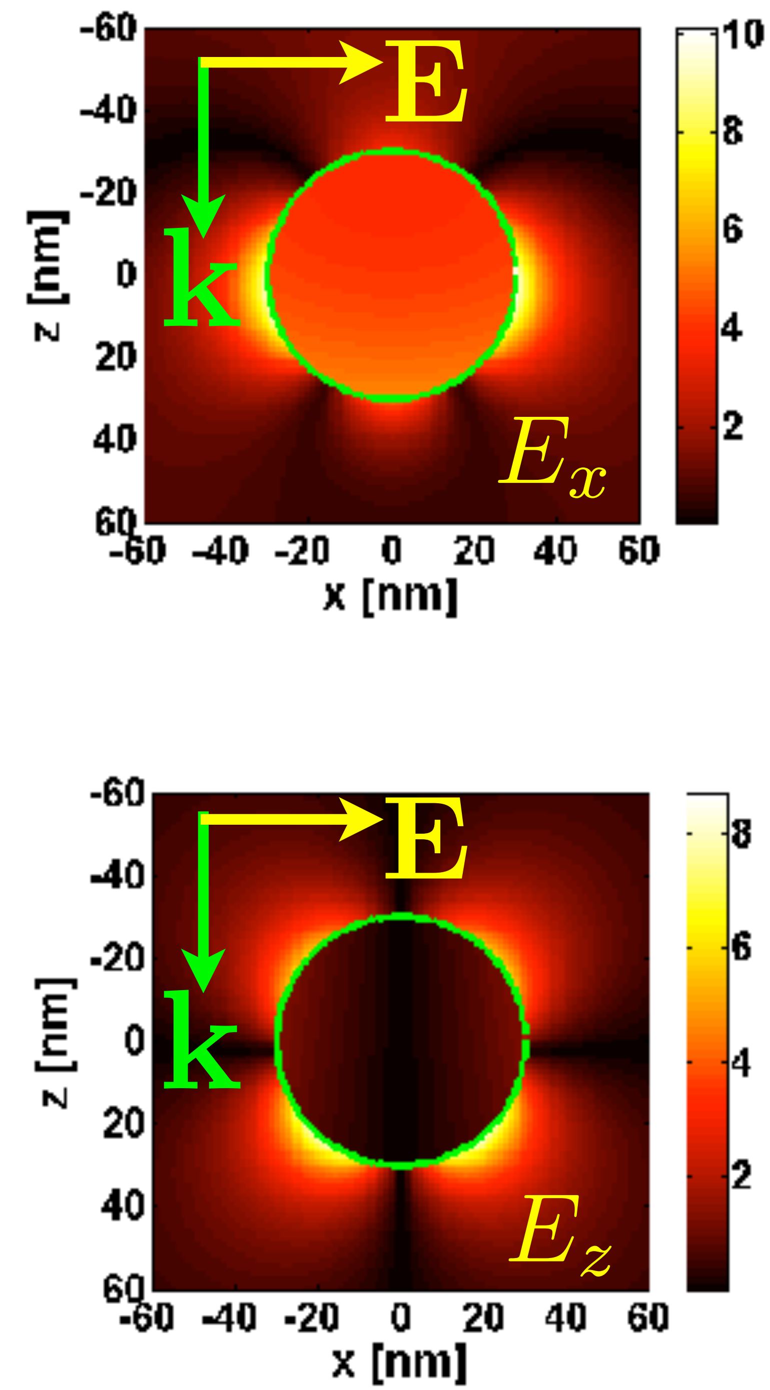
silver sphere surrounded by air



- smaller spheres absorb stronger
- larger spheres → resonance red-shift and higher order resonances

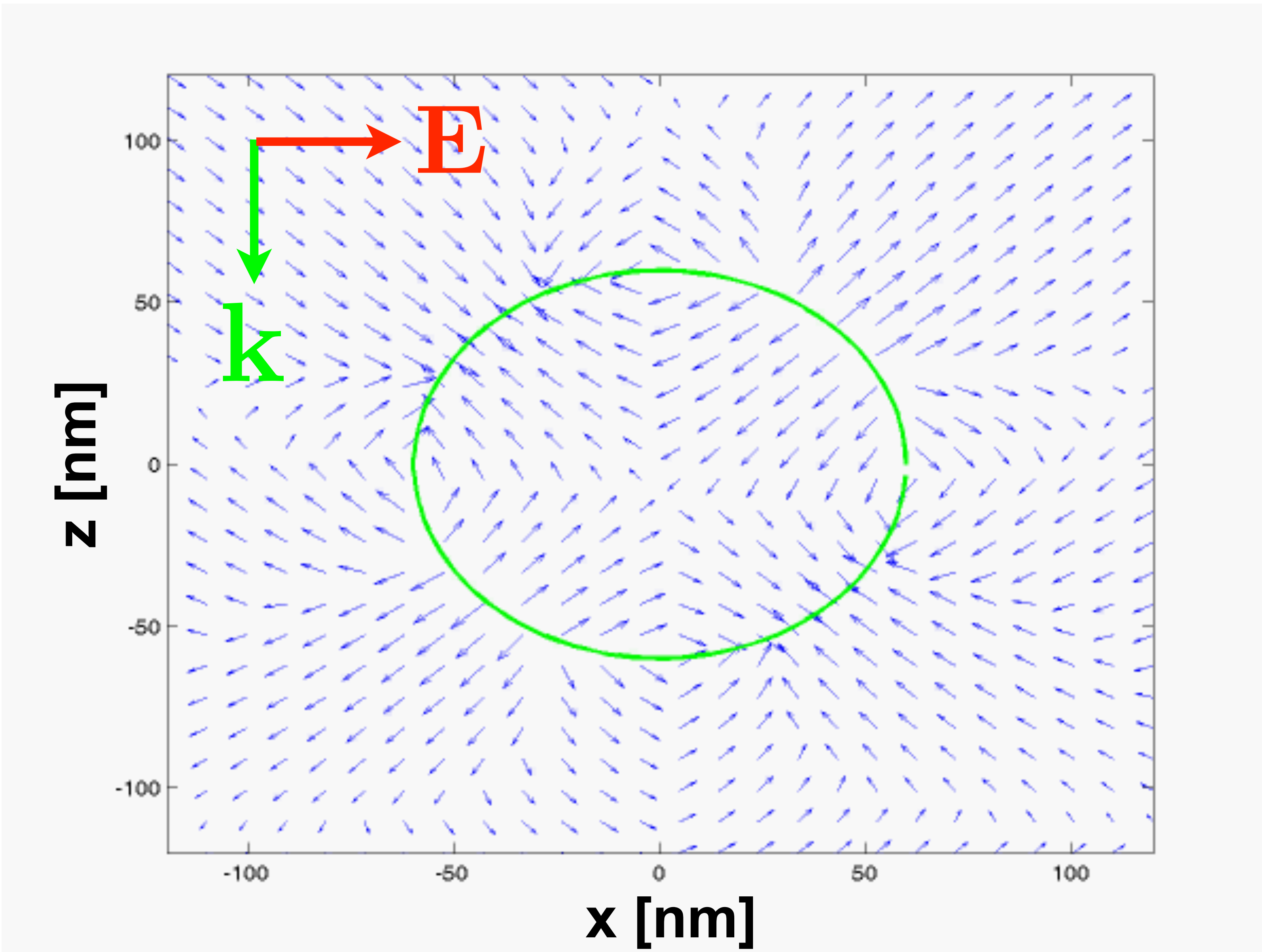
Metallic nanoparticles - the dipolar resonance

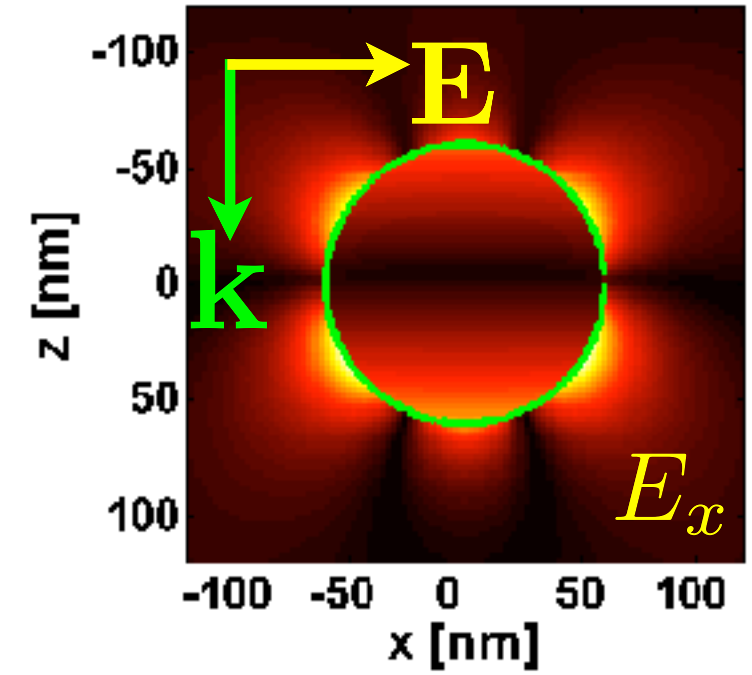


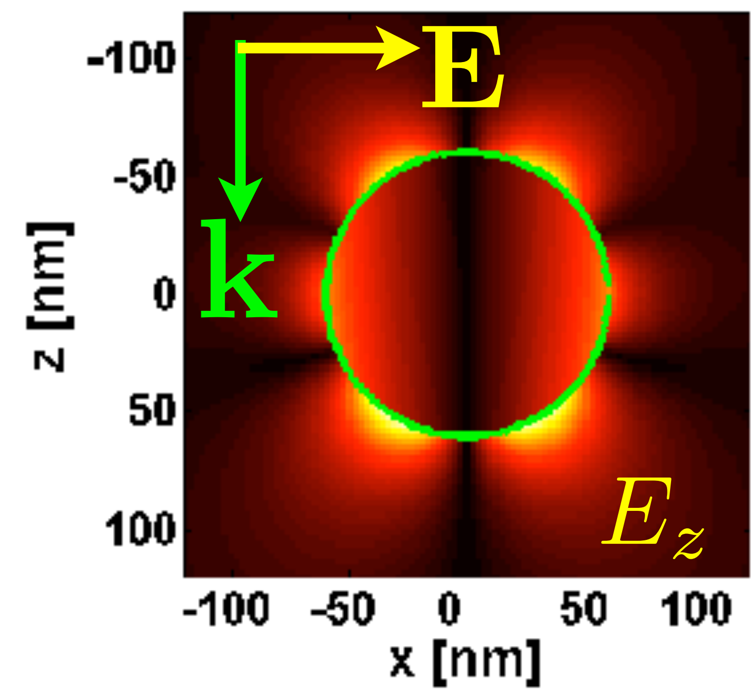


resonant oscillation of the charge density

Metallic nanoparticles - the quadruoplar resonance

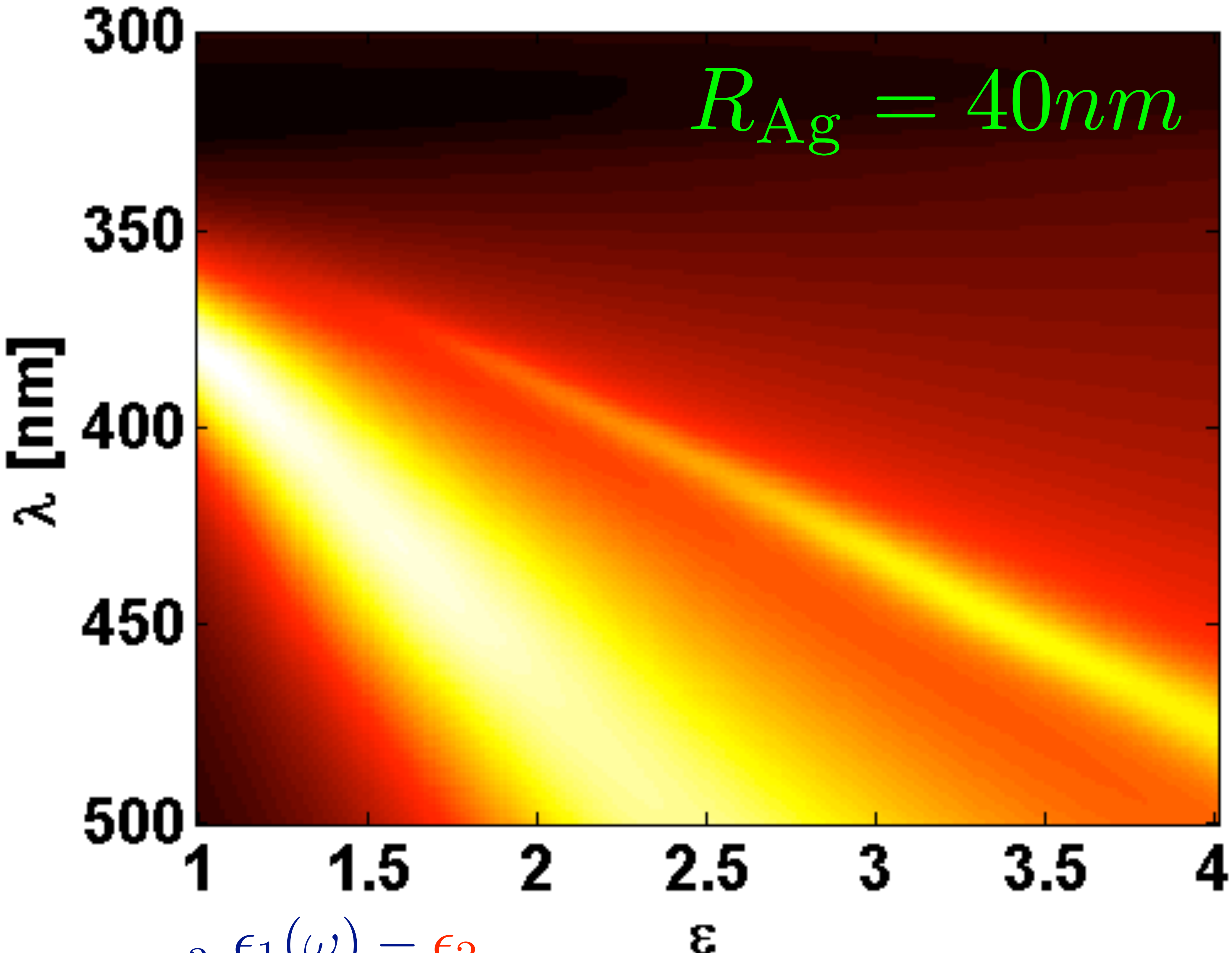






sphere is a cavity → higher order resonances

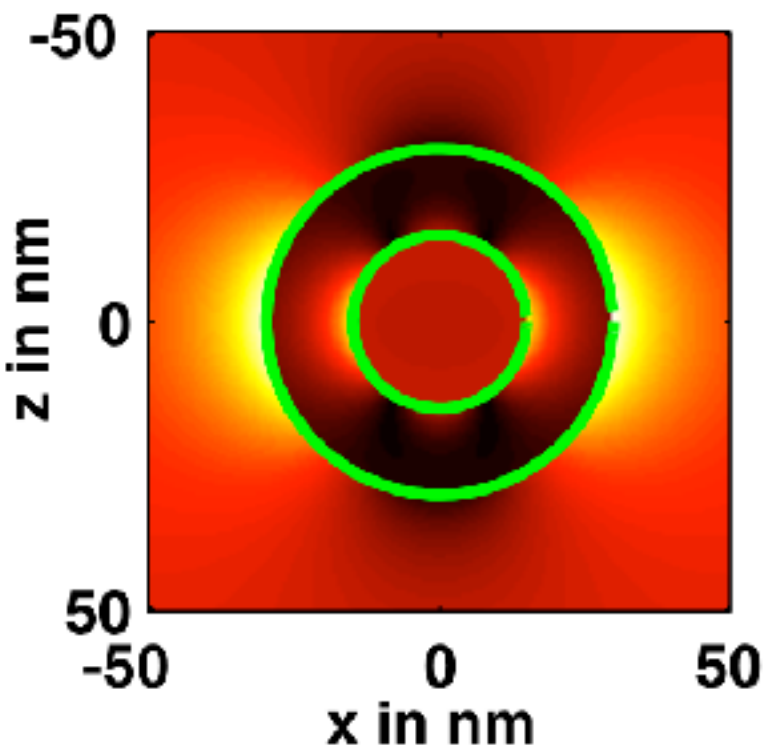
Impact of a dielectric surrounding



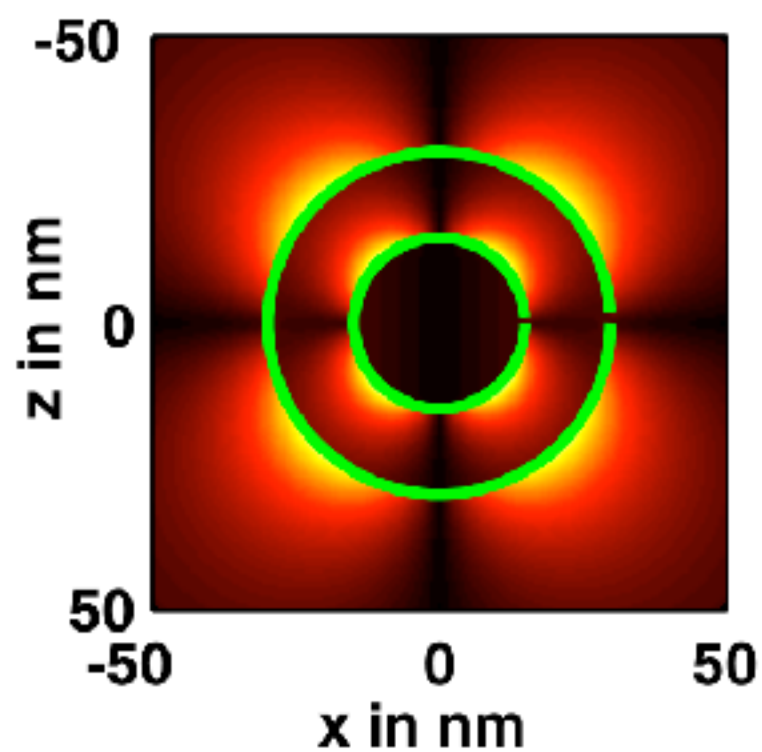
$$\alpha = 4\pi R^3 \frac{\epsilon_1(\omega) - \epsilon_2}{\epsilon_1(\omega) + 2\epsilon_2}$$

Mie theory for spheres

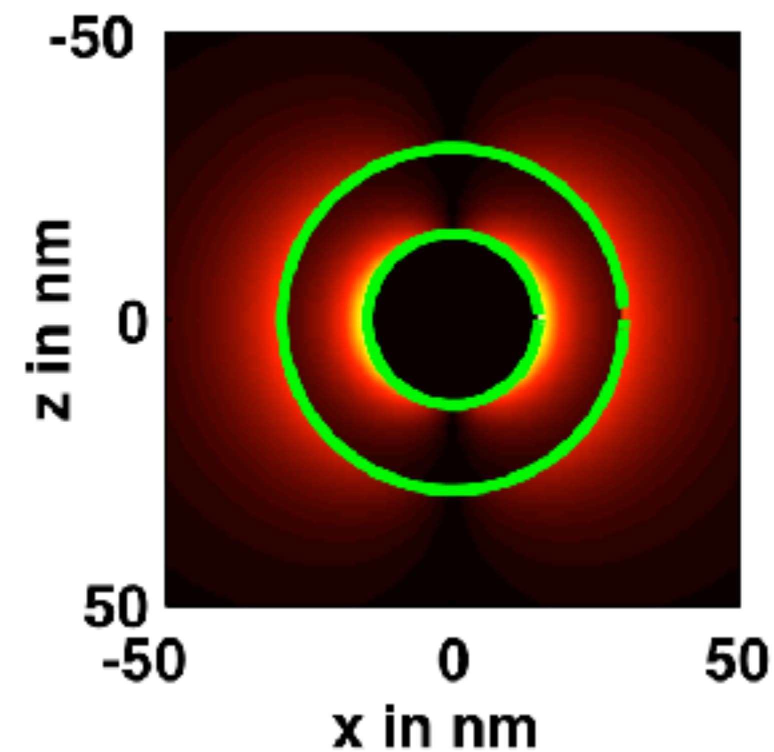
analysing metallic nano particles in 3D with or without covering layers



$|\mathbf{E_x}|$



$|\mathbf{E_z}|$



$|\mathbf{E_y}|$

in the x-z-plane at λ=633nm, r_{Core} = 15 nm, n_{Shell} = 1.5, r_{Shell} = 30 nm

Computational Photonics

Scattering theory: fully dynamic sphere