Effective medium theory to mimic the fields scattered by metallic particles with insulators

Alexis Devilez, Xavier Zambrana-Puyalto, Brian Stout, Nicolas Bonod
Institut Fresnel, CNRS, Université d'Aix-Marseille, Ecole Centrale Marseille,
Marseille, France

Théorie Spectrale des Nouveaux Matériaux

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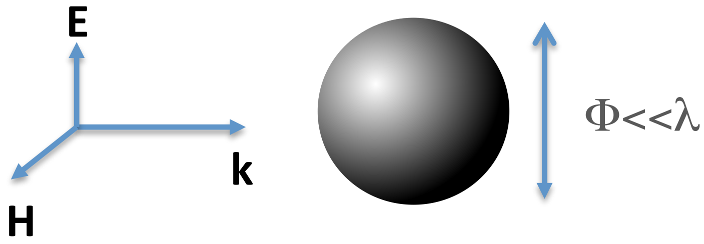
Interaction between light and particles



The EM response is described by the polarisability:

$$\begin{pmatrix} \vec{p} \\ \vec{m} \end{pmatrix} = \begin{pmatrix} \alpha_e & 0 \\ 0 & \alpha_H \end{pmatrix} \begin{pmatrix} \vec{\mathbf{E}} \\ \vec{\mathbf{H}} \end{pmatrix}$$

Far field — Near and intermediary field

$$\vec{\mathbf{E}} = \frac{e^{iwt}}{4\pi\varepsilon_0 r^3}\left[\underbrace{k^2 r^2 \left(\vec{p} \times \hat{r}\right) \times \hat{r}}_{\text{Far field}} + \underbrace{(3 - ikr)(\overrightarrow{(p.\hat{r}).\hat{r}} - \vec{p})}_{\text{Near and intermediary field}}\right]$$

Mie scattering coefficients

$$\alpha_e = -\frac{6\pi}{ik^3} a_1 \qquad a_n \equiv -t_n^{(e)} = \frac{j_n(k_1 R)}{h_n^{(+)}(k_1 R)} \frac{\left[\varepsilon_{21}\varphi_n^{(1)}(k_1 R) - \varphi_n^{(1)}(k_2 R)\right]}{\left[\varepsilon_{21}\varphi_n^{(+)}(k_1 R) - \varphi_n^{(1)}(k_2 R)\right]}$$

$$\alpha_h = -\frac{6\pi}{ik^3} b_1 \qquad b_n \equiv -t_n^{(h)} = \frac{j_n(k_1 R)}{h_n^{(+)}(k_1 R)} \frac{\left[\mu_{21}\varphi_n^{(1)}(k_1 R) - \varphi_n^{(1)}(k_2 R)\right]}{\left[\mu_{21}\varphi_n^{(+)}(k_1 R) - \varphi_n^{(1)}(k_2 R)\right]}$$

$$\varphi_n(z) \equiv \frac{\left[z j_n(z)\right]'}{j_n(z)} \qquad \varphi_n^+(z) \equiv \frac{\left[z h_n^+(z)\right]'}{h_n^+(z)} \qquad z = kr$$

Electromagnetic resonances

$$a_n = \frac{j_n\left(z_b\right)}{h_n^+\left(z_b\right)} \frac{\varepsilon_s \varphi_n\left(z_b\right) - \varepsilon_b \varphi_n\left(z_s\right)}{\varepsilon_s \varphi_n^+\left(z_b\right) - \varepsilon_b \varphi_n\left(z_s\right)}$$

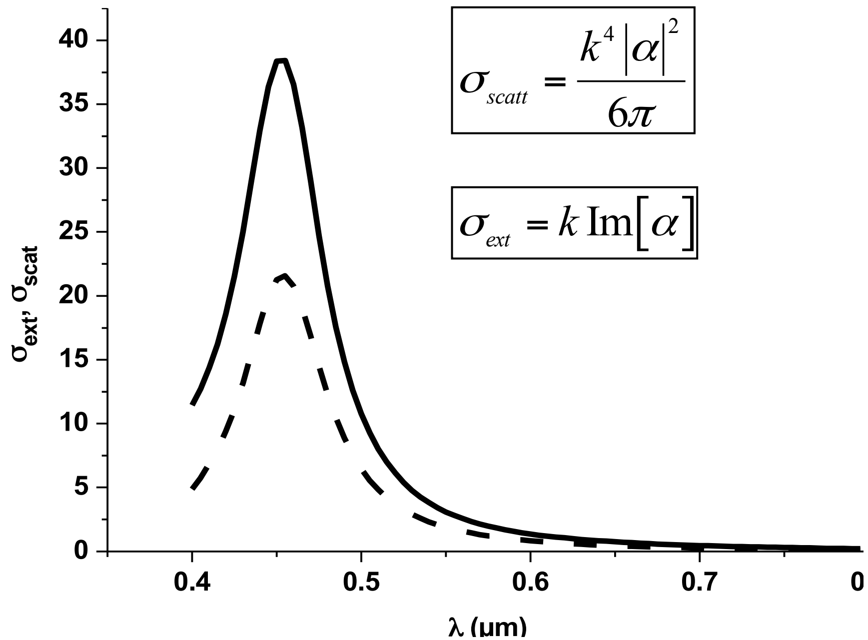
$$\varepsilon_s \varphi_n^+\left(z_b(\omega_p)\right) - \varepsilon_b \varphi_n\left(z_s(\omega_p)\right) = 0$$

Pole ω_p in the complex ω-plane

If the particle is illuminated at $\omega = \mathrm{Re}(\omega_p)$, resonant response

$$\begin{pmatrix} \vec{p} \\ \vec{m} \end{pmatrix} = \begin{pmatrix} \alpha_e & 0 \\ 0 & \alpha_H \end{pmatrix} \begin{pmatrix} \vec{\mathbf{E}} \\ \vec{\mathbf{H}} \end{pmatrix}$$

$$\vec{\mathbf{E}} = \frac{e^{iwt}}{4\pi\varepsilon_0 r^3}\left[k^2 r^2 \left(\vec{p} \times \hat{r}\right) \times \hat{r} + (3 - ikr)((\vec{p}.\hat{r}).\hat{r} - \vec{p})\right]$$



Analytic expression of the polarisability

$$a_n = \frac{j_n(z_b)}{h_n^+(z_b)} \frac{\varepsilon_s \varphi_n(z_b) - \varepsilon_b \varphi_n(z_s)}{\varepsilon_s \varphi_n^+(z_b) - \varepsilon_b \varphi_n(z_s)}$$

$$\begin{cases} \varphi_n(z) \equiv \dfrac{[z j_n(z)]'}{j_n(z)} \\ \varphi_n^+(z) \equiv \dfrac{[z h_n^+(z)]'}{h_n^+(z)} \end{cases}$$

Quasi-static approximation: $\alpha_{e,0} = 4\pi r^3 \dfrac{\varepsilon_s - \varepsilon_b}{\varepsilon_s + 2\varepsilon_b}$

Radiative correction: $\dfrac{1}{\alpha_e} = \dfrac{1}{\alpha_{e,0}} - \dfrac{ik^3}{6\pi}$

Higher order: $\dfrac{1}{\alpha_e} = \dfrac{1}{\alpha_{e,0}} \left(1 - \dfrac{3(kr)^2(\varepsilon - 2)}{5(\varepsilon + 2)} - \dfrac{3(kr)^4}{350(\varepsilon + 2)}(\varepsilon^2 - 24\varepsilon + 16) \right) - \dfrac{ik^3}{6\pi}$

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1. Photonic resonances hosted by spherical particles

2. **Photonic resonances hosted by metallic particles**

3. Silicon particles: a novel platform to enhance light matter interactions?

4. Equivalence between dielectric and metallic particles.

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Plasmonic resonance on metallic particles

Resonance :

Pole of the polarisability in the complex ω-plane

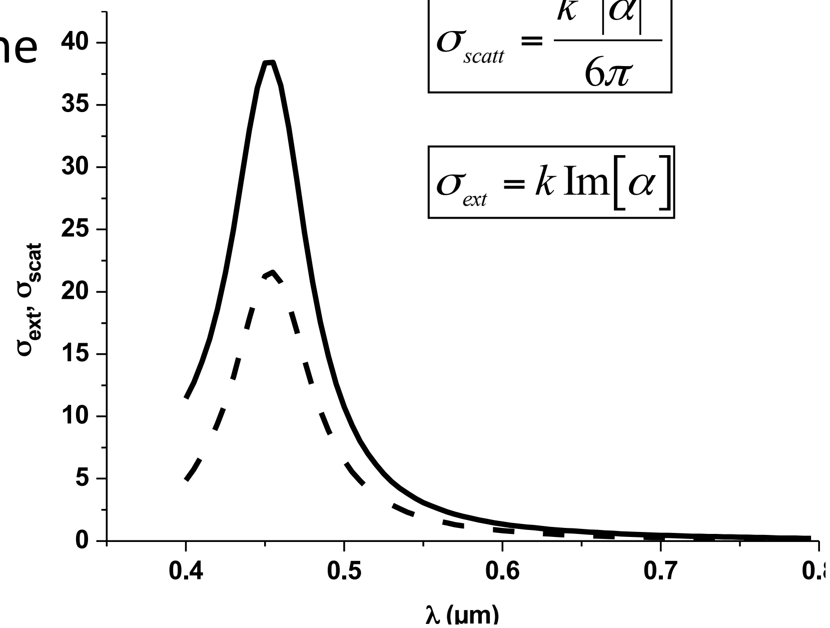
$$\alpha_{e,0} = 4\pi r^3 \frac{\varepsilon_1(\omega) - \varepsilon_2(\omega)}{\varepsilon_1(\omega) + 2\varepsilon_2(\omega)}$$

Drude-Lorentz's model:

$$\varepsilon_1(\omega) = 1 - \frac{\omega^2_{plasma}}{\omega^2 + i\Gamma\omega}$$

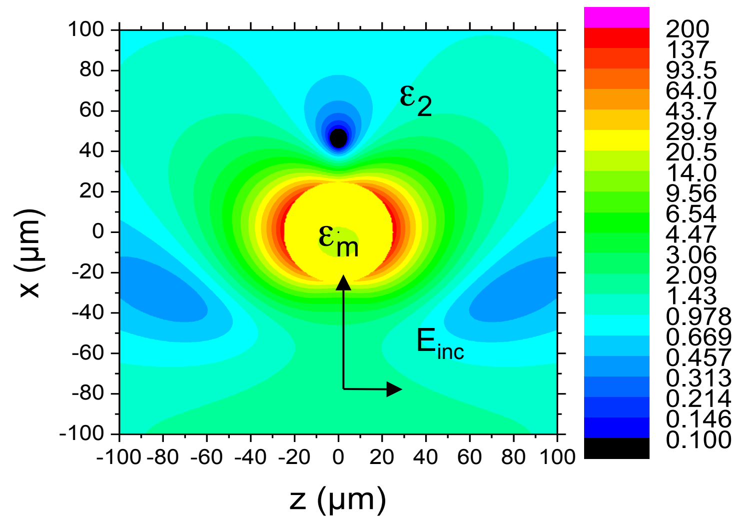
Resonance condition:

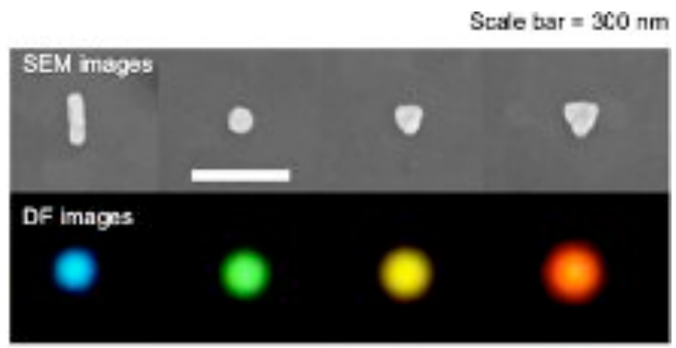
$$\mathrm{Re}(\varepsilon_2) > 0, \mathrm{Re}(\varepsilon_1) < 0$$



$$\sigma_{scatt} = \frac{k^4 |\alpha|^2}{6\pi}$$

$$\sigma_{ext} = k\,\mathrm{Im}[\alpha]$$





Advanced materials 19 (22), 3771-3782 (2007)

The Drude-Sommerfeld's model

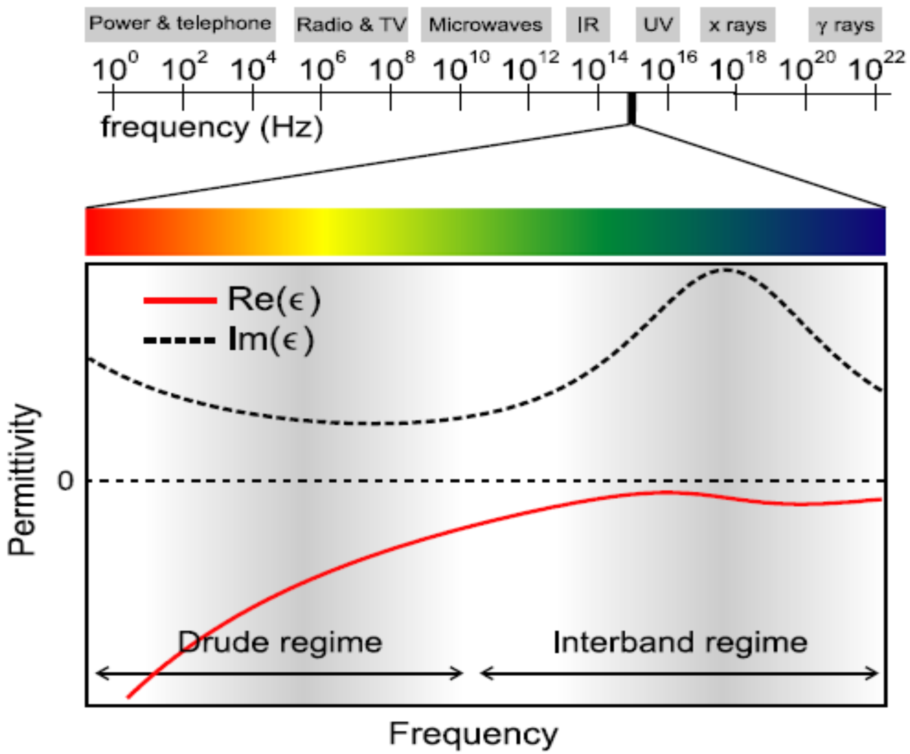
This model accounts for (i) a negative real part and (ii) a significant imaginary part

The optical response in metals is dominated by the collective behavior of the free electron gas.

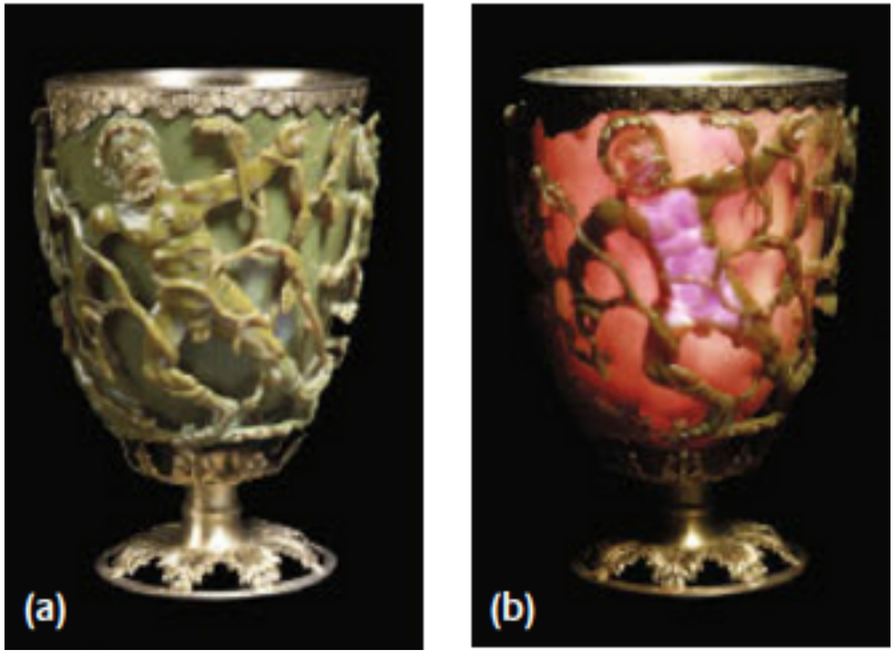
To a first approximation,the conduction electrons in the metal can be treated as an ideal electron gas moving in the background of the positive metal ions.

Using the Drude–Sommerfeld model, the dielectric function of the metal can be expressed as:

$$\varepsilon_1(\omega) = 1 - \frac{\omega^2_{plasma}}{\omega^2 + i\Gamma\omega}$$



Resonant light interaction with particles: Lycurgus cup (4th century BC)



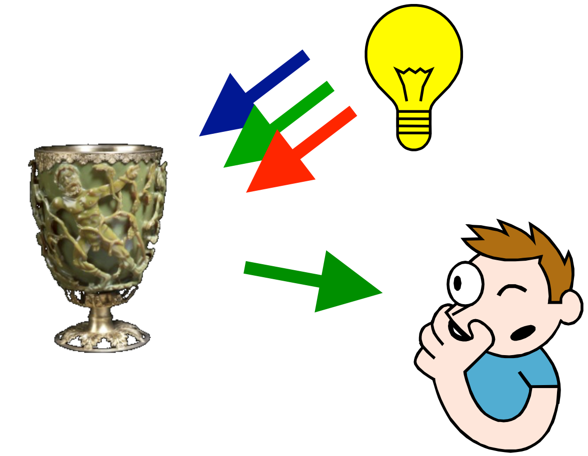
Scene showing the triumph of Dionysus on Lycurgus

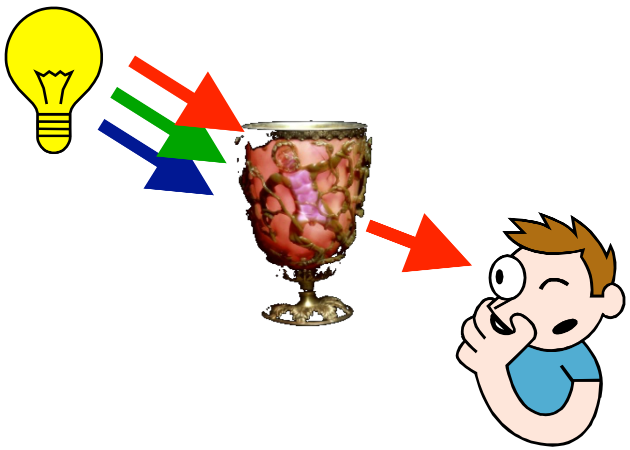
Dichroism: green in reflection, red in transmission.

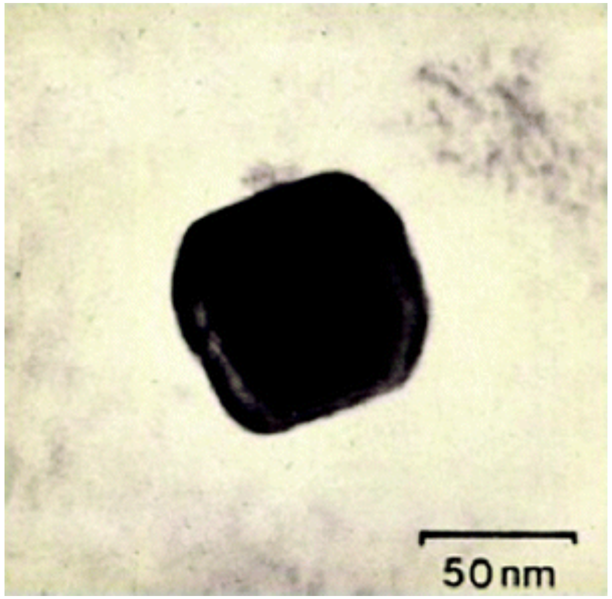
1st report in 1845.

1st studies in 1950.

Lycurgus cup



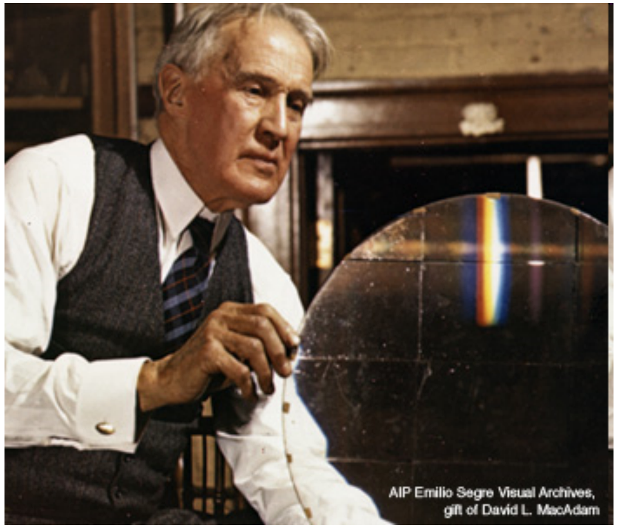




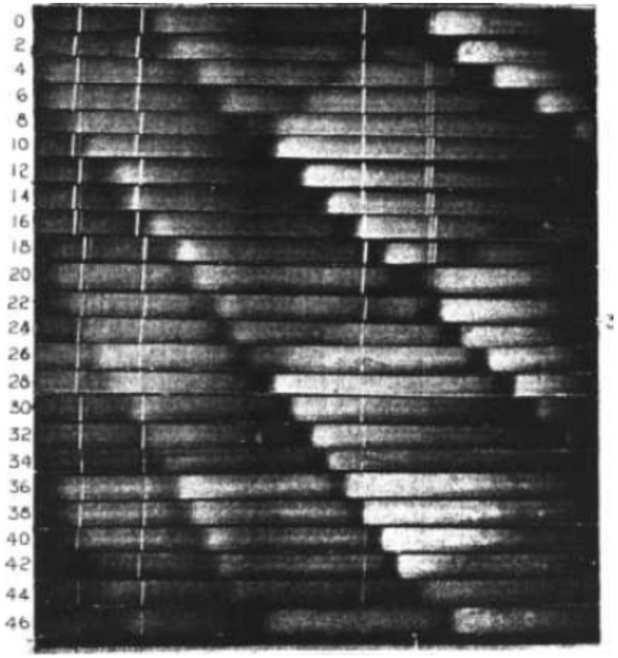
Detection of particles 50-100 nm
in diameter, alloy:
gold (30%) / silver (70%)
Traces of copper

The Wood's anomalies

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



Robert W. Wood: The Scientist who Played with Optics
Wood at Johns Hopkins University, with his mosaic replica diffraction grating. Optics&Photonics News (october 2009)



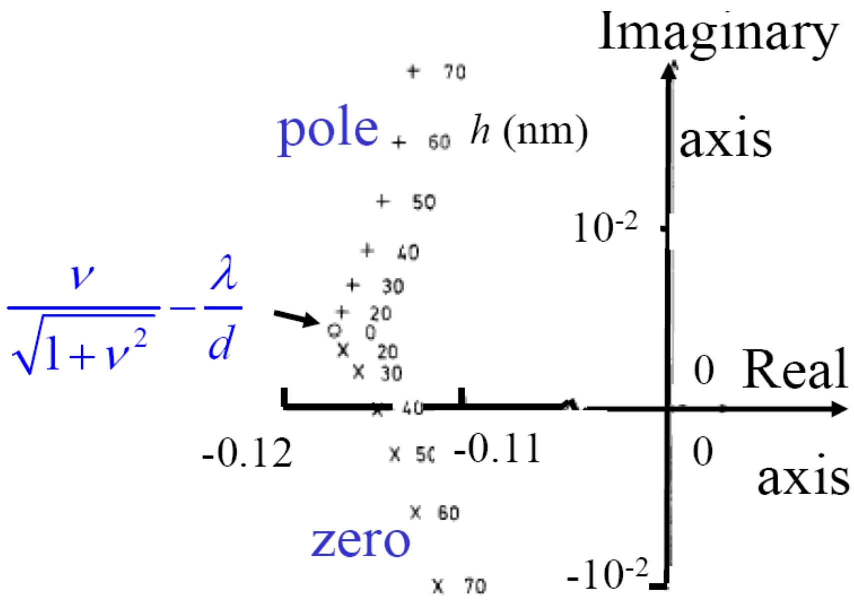
R. W. Wood, Philos. Mag. 4, 396-402 (1902)
R. W. Wood, Philos. Mag. 23, 310 (1912)
L. R. Ingersoll, Astrophys. J. 51, 129 (1920)
R. W. Wood, Phys. Rev. 48, 928-936 (1935)
J. Strong, Phys. Rev. 49, 291-296 (1936)

« On mounting the grating on the table of a spectrometer I was astounded to find that under certain conditions the drop from maximum illumination to minimum, a drop certainly of from 10 to 1. »

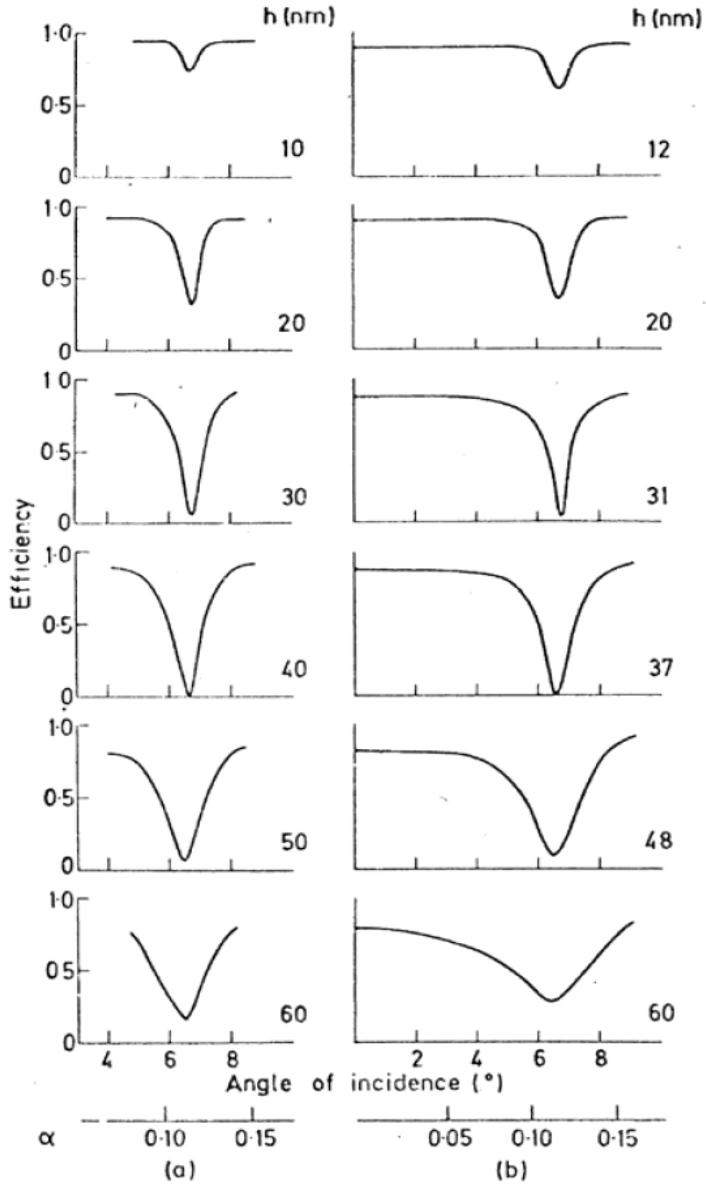
« So far as I know, polarization has never been introduced into the theory of gratings »

« On turning the nicol through a right angle all trace of the bright and dark bands disappeared »

1976: Complete light absorption



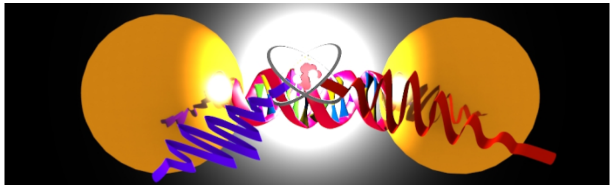
R. Petit et al., Opt. Commun. **19** (1976)

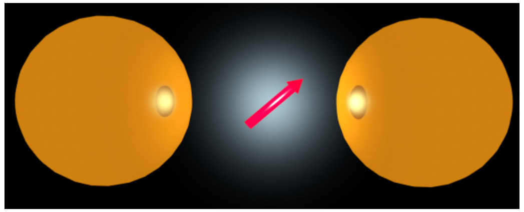


Dratic fall of the reflectance from 90% to 0 below 1%

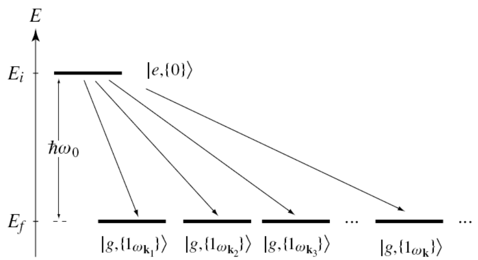
Hutley et al., Opt. Commun. **19** (1976)

Manipulating the spontaneous emission with metallic particles





2 levels system



$$\frac{\gamma}{\gamma_0}=1+\frac{6\pi\varepsilon}{|\mu|^2}\frac{1}{k^3}\mathrm{Im}\left\{\mu^*.\mathbf{E}_s(\mathbf{r}_0)\right\}$$

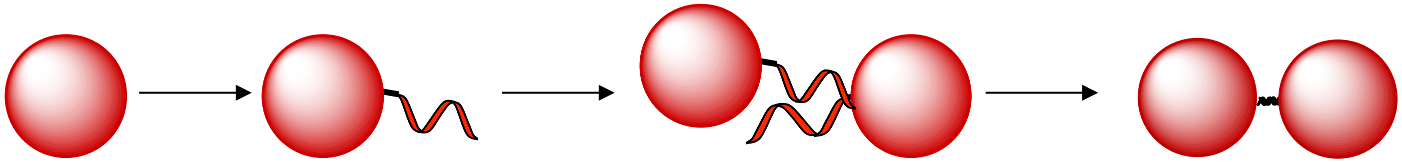
Oscillating dipole

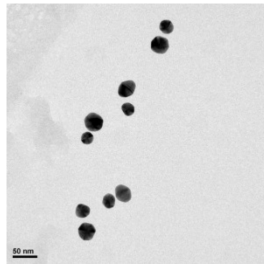
$$\frac{d}{dt^2}\mathbf{p}(t)+\gamma_0\frac{d}{dt}\mathbf{p}(t)+\omega_0^2\mathbf{p}(t)=\frac{1}{m}\mathbf{E}_s(\mathbf{r}_0)$$

$$\mathbf{p}(t)=\mathrm{Re}\left[\mathbf{p}_0 e^{-i\omega t}e^{-\gamma t/2}\right]$$

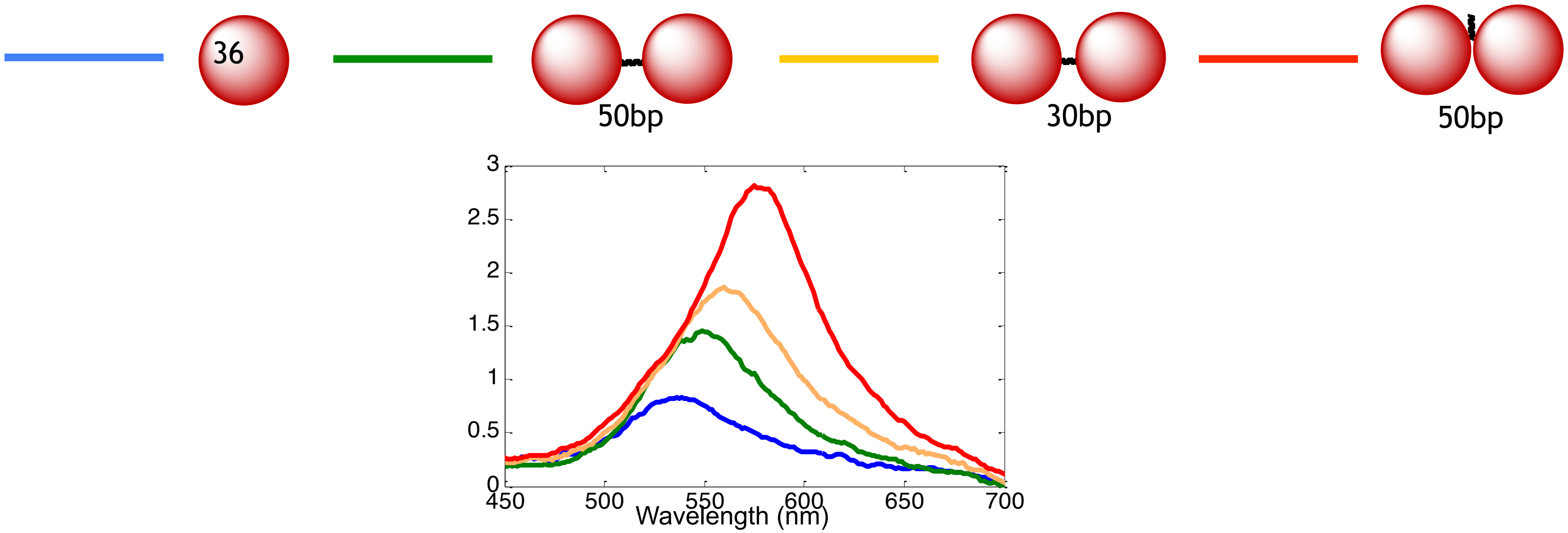
$$\frac{\gamma}{\gamma_0}=\frac{P}{P_0}=1+\frac{6\pi\varepsilon}{|p|^2}\frac{1}{k^3}\mathrm{Im}\left\{\mathbf{p}^*.\mathbf{E}_s(\mathbf{r}_0)\right\}$$

Coupling of 2 gold nanoparticles linked by a double DNA strand



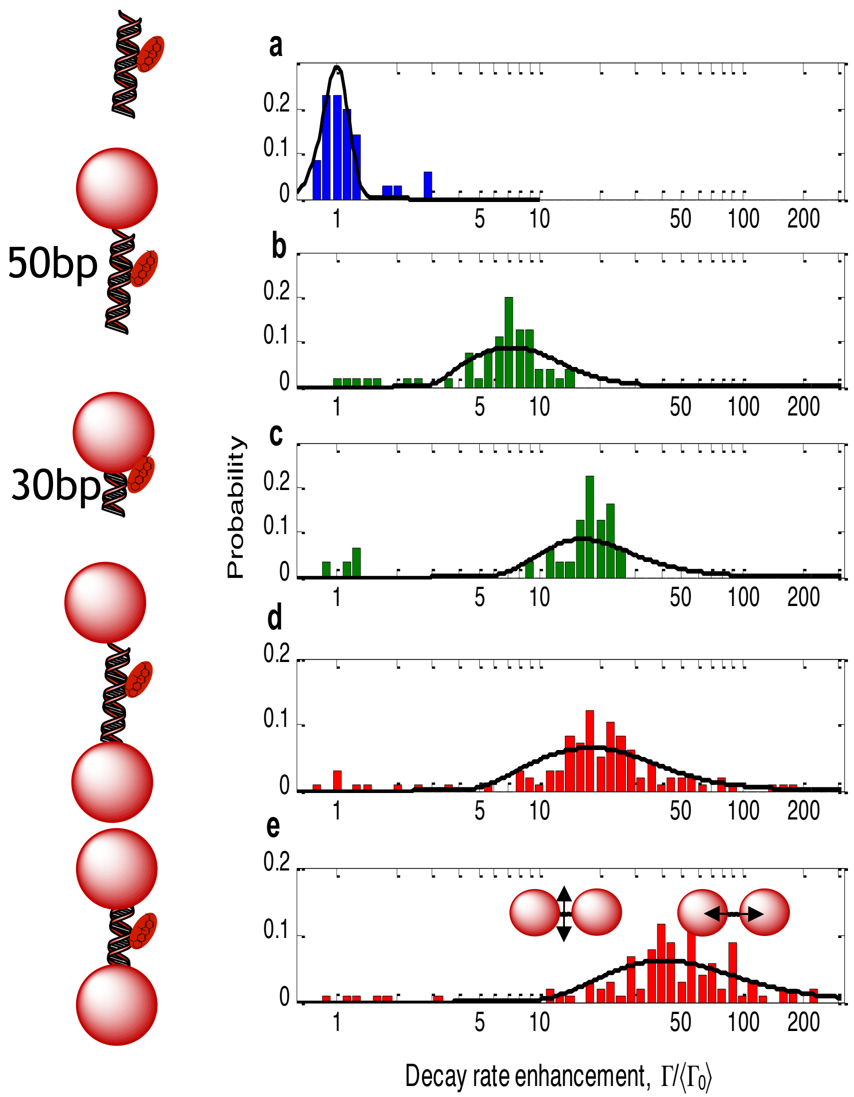


Dimer of 36 nm diameter.



M. P. Busson et al., Nano Lett. **11**, 5060-5065 (2011)

Fast Single photon source

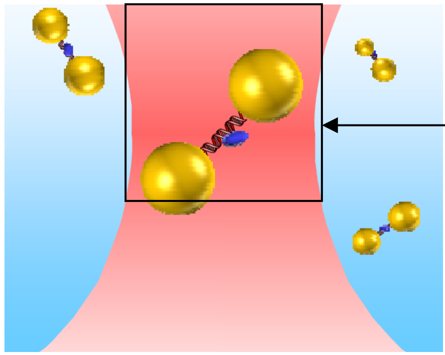


Γ/Γ_0 increased by 2 orders of magnitude: the lifetime fluorescence is decreased from 3 ns to 35 ps.

M. P. Busson, B. Rolly, B. Stout, N. Bonod, S. Bidault, Nature Commun. **3**, 962 (2012)

‘Meta’ molecule: organo-metallic emitter

Colloidal ‘meta’ chromophores that can be characterized as classical emitters in solution with standard fluorescent correlation spectroscopy.



Organo-metallic particle

Sample	Fluorescence enhancement factor [a]	Lifetime τ (ps)	Molar extinction coefficient ε ($M^{-1}cm^{-1}$)	Quantum effiency ϕ (%)
ATTO	1	4300	10^5 [b]	65[c]
mono50	0.51	615	$3.4\ 10^5$	9.3
mono30	0.35	135	$1.2\ 10^6$	1.8
dim50	0.76	185	$1.1\ 10^6$	4.8
dim30	1.35	65	$5.7\ 10^6$	1.6

M. P. Busson et al., Angew. Chem. Int. Ed. **51**, 11083–11087 (2012)

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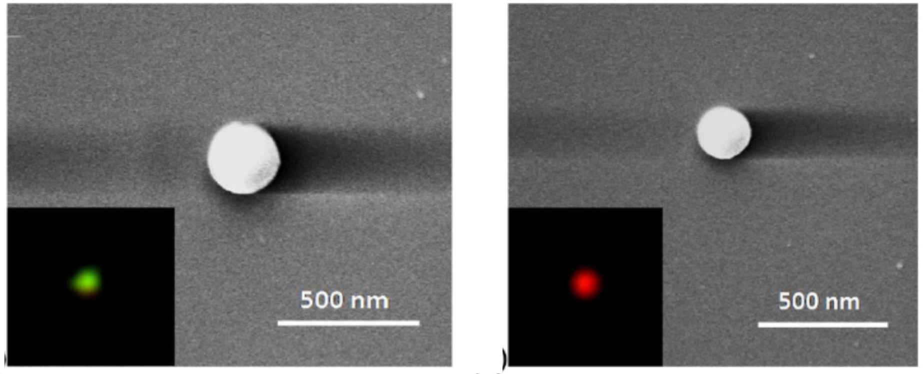
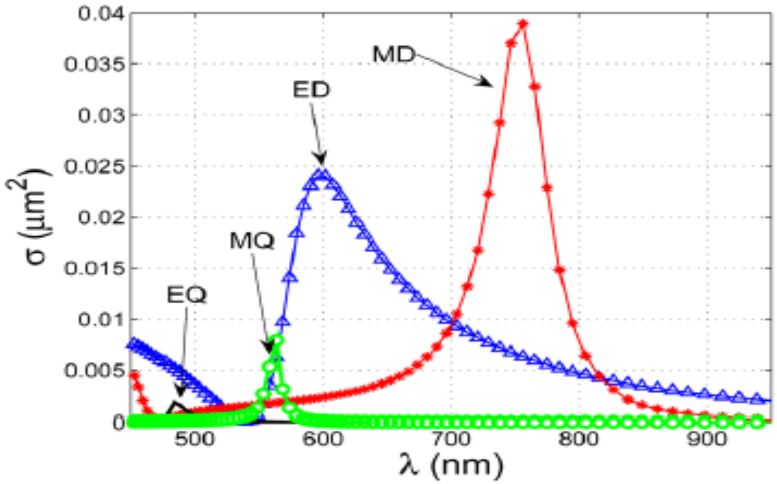
First experimental demonstration





A. B. Evlyukhin et al., Nanolett. 12, 3749–3755 (2012)

$$\begin{pmatrix} \mathbf{P} \\ \mathbf{M} \end{pmatrix} = \begin{pmatrix} \alpha_e & 0 \\ 0 & \alpha_H \end{pmatrix} \begin{pmatrix} \mathbf{E} \\ \mathbf{H} \end{pmatrix}$$

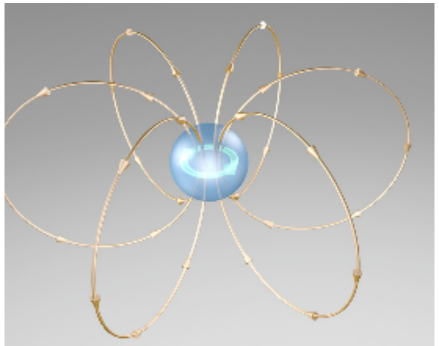
-Resonant interaction between light and individual particles

-Structural color of silicon particles

-Electric and magnetic response with similar amplitudes

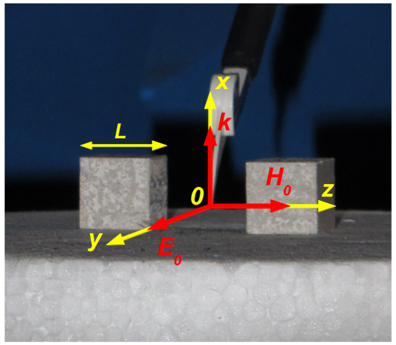
Current research projects

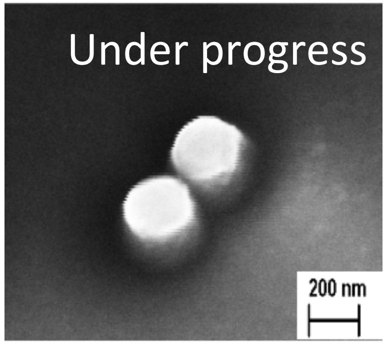
Photonic cavity:
Enhancement of the purcell factor,
Electric and magnetic spontaneous emission



B. Rolly, B. Bebey, S. Bidault, B. Stout, N. Bonod, Phys. Rev. B **85**, 245432 (2012)
X. Zambrana-Puyalto, N. Bonod, Phys. Rev. B **9**1, 195422 (2015)

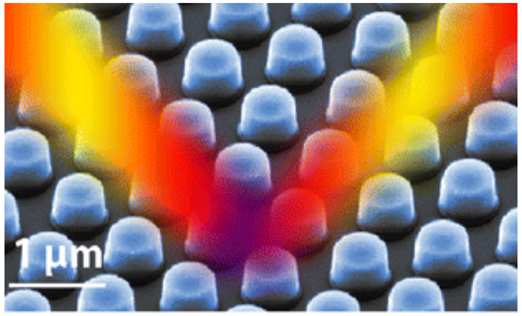
Gap antenna:
Enhancement of the electric and
magnetic fields





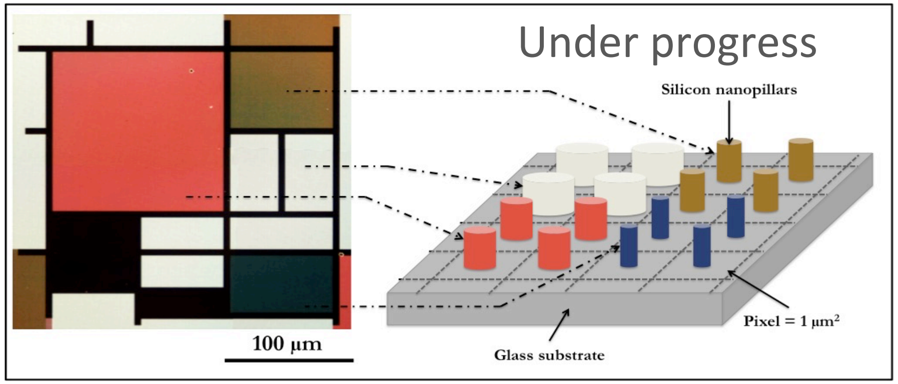
G. Boudarham, R. Abdeddaim, N. Bonod, Appl. Phys. Lett. **104**, 021117 (2014)

Mirrors:



P. Moitra, B. A. Slovick, W. Li, I. I. Kravchencko, D. P. Briggs, S. Krishnamurthy, J. Valentine, ACS Photonics **2**, 692-698 (2015)
See N&Vs: N. Bonod, Nature Mat. **14**, 664-665 (2015)

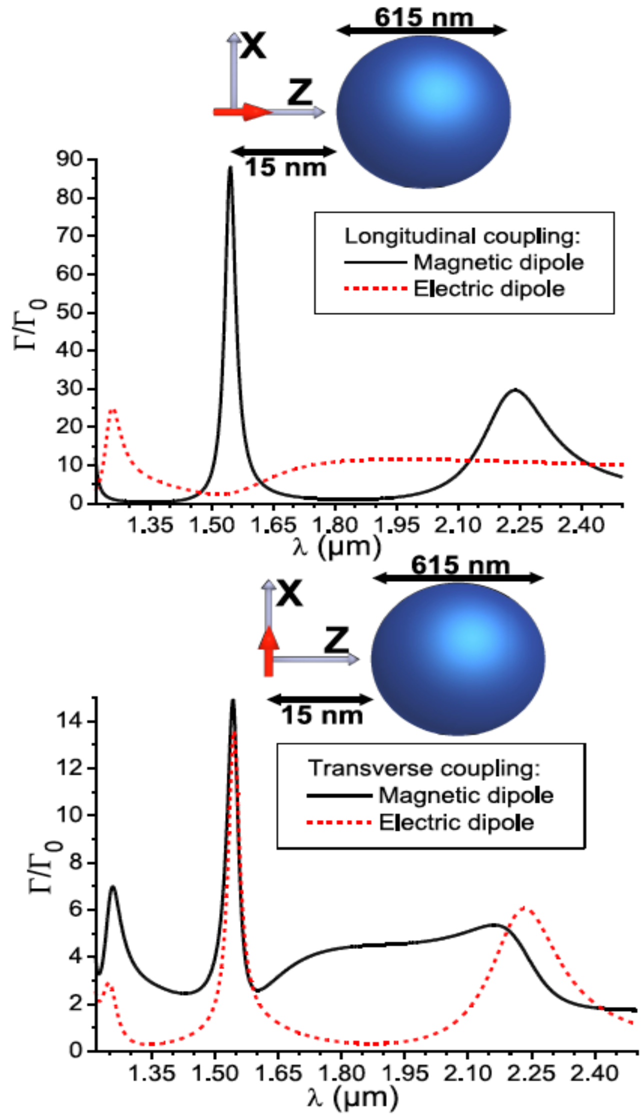
Nanoprinters:



Electric and magnetic emission rates

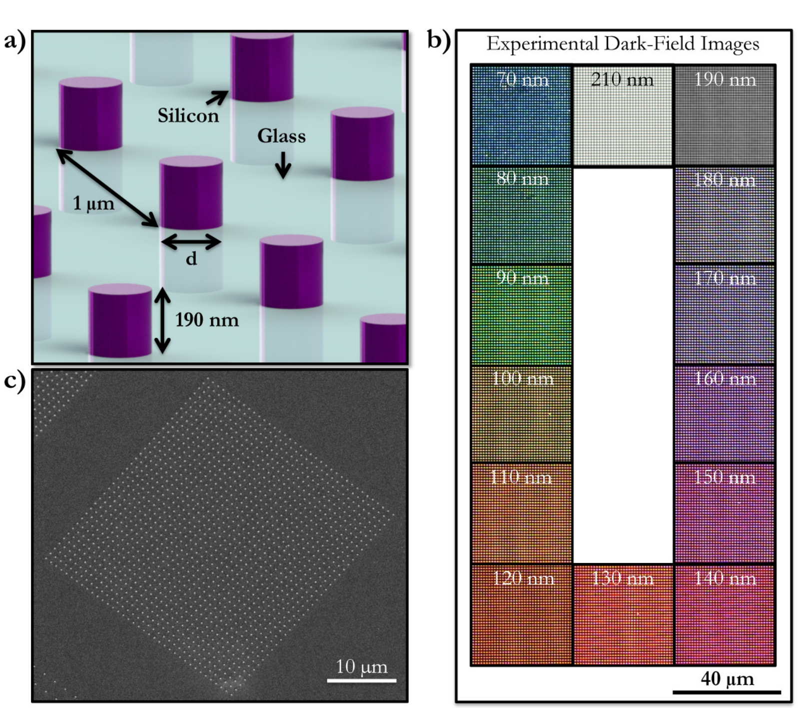
$$\frac{\Gamma_{tot}^{L,u}}{\Gamma_0} = 1 + \mathrm{Re}\left\{ \alpha_1^u \left[3i \frac{e^{ikd}}{(kd)^3}(ikd+1) \right]^2 + \alpha_2^u \left[\frac{3}{\sqrt{5}} \frac{e^{ikd}}{(kd)^4}\left(5i(kd)^2 - 15kd - 15i\right) \right]^2 \right\}$$

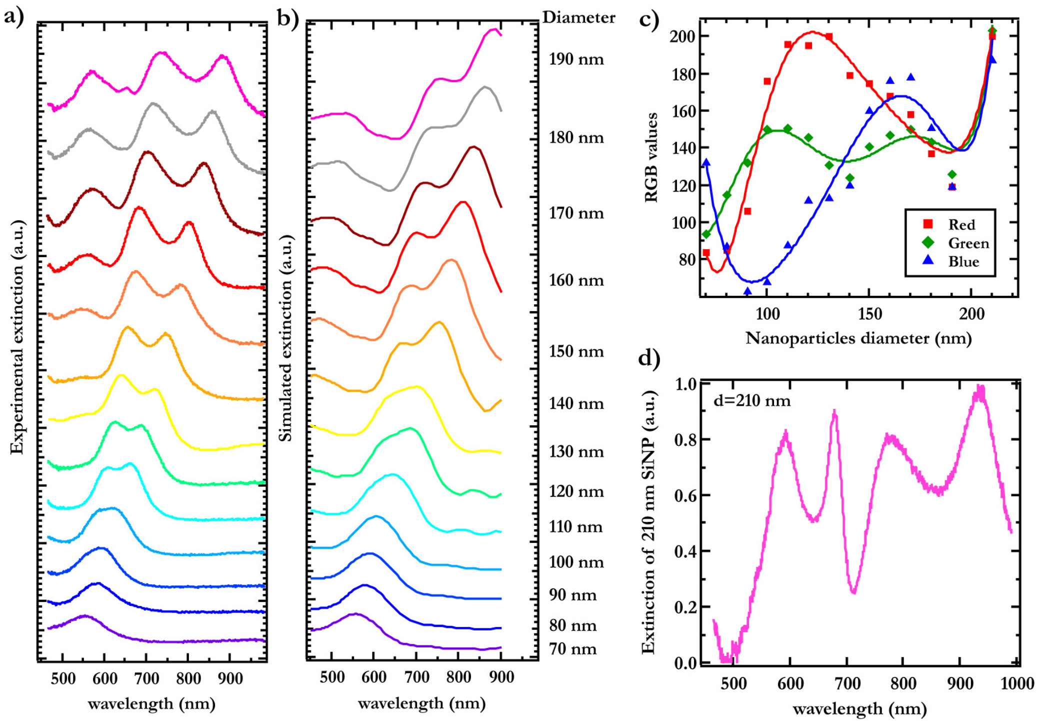
$$\frac{\Gamma_{tot}^{t,u}}{\Gamma_0} = 1 + \mathrm{Re}\left\{ \alpha_1^u \left[\frac{3i}{2} \frac{e^{ikd}}{(kd)^3}\left(-(kd)^2 - ikd + 1\right) \right]^2 + \alpha_2^u \left[\frac{\sqrt{15}}{2} \frac{e^{ikd}}{(kd)^4}\left(-(kd)^3 - 3i(kd)^2 + 6kd + 6i\right) \right]^2 - \alpha_1^v \left[\frac{3i}{2} \frac{e^{ikd}}{(kd)^2}(kd+i) \right]^2 - \alpha_2^v \left[\frac{\sqrt{15}}{2} \frac{e^{ikd}}{(kd)^3}\left(-(kd)^2 - 3ikd + 3\right) \right]^2 \right\}^2$$

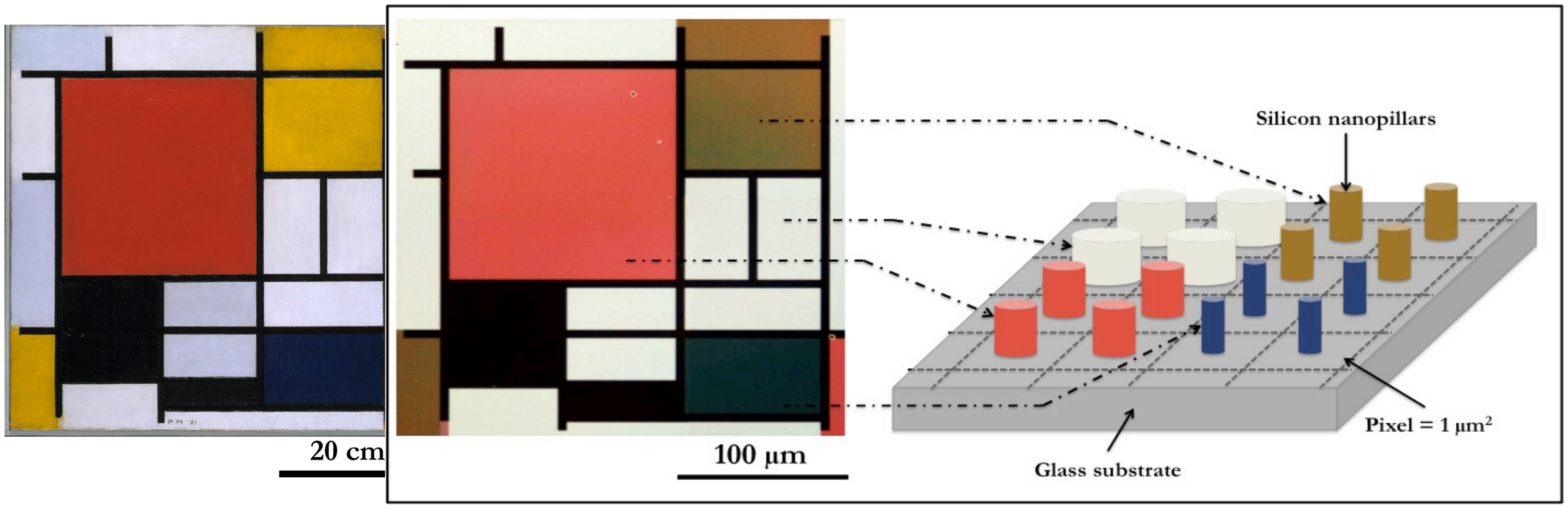


B. Rolly, B. Bebey, S. Bidault, B. Stout, N. Bonod, Phys. Rev. B **85**, 245432 (2012)

Structural colour







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4. Equivalence between dielectric and metallic particles

Alexis Devilez

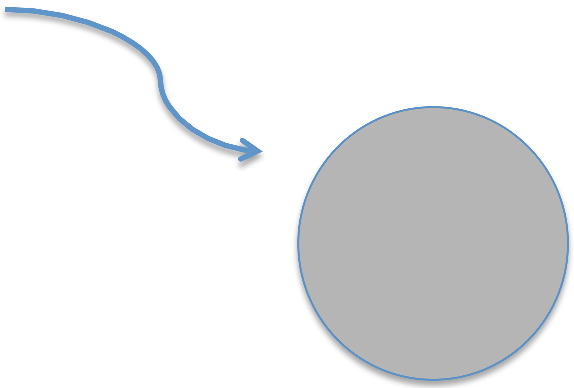
Xavier Zambrana Puyalto

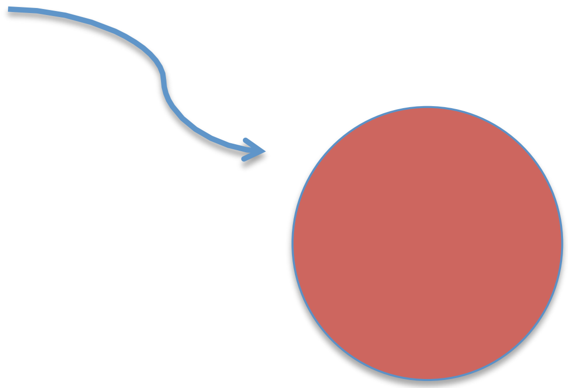
Brian Stout

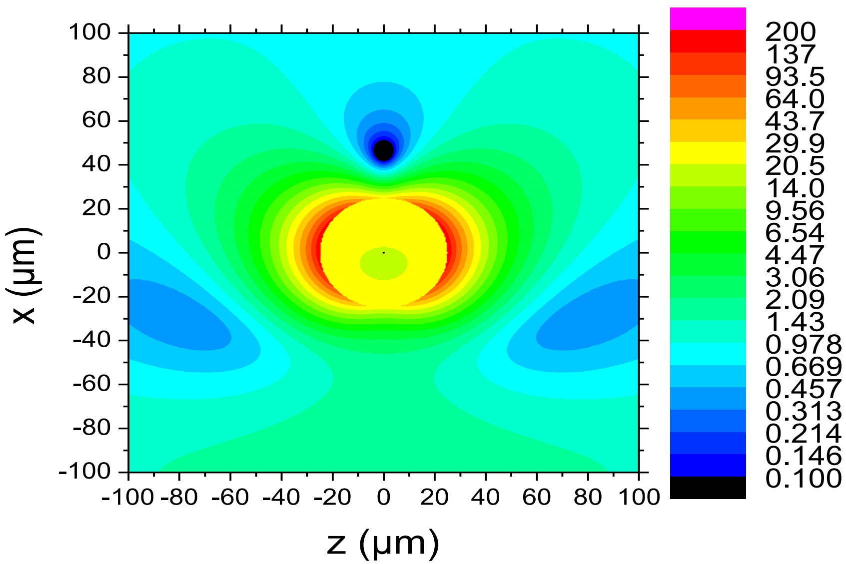
Is it possible to reproduce the electromagnetic response of metallic particles with insulators?

$$z_0 = k_0 r$$

$$r << \lambda$$







????

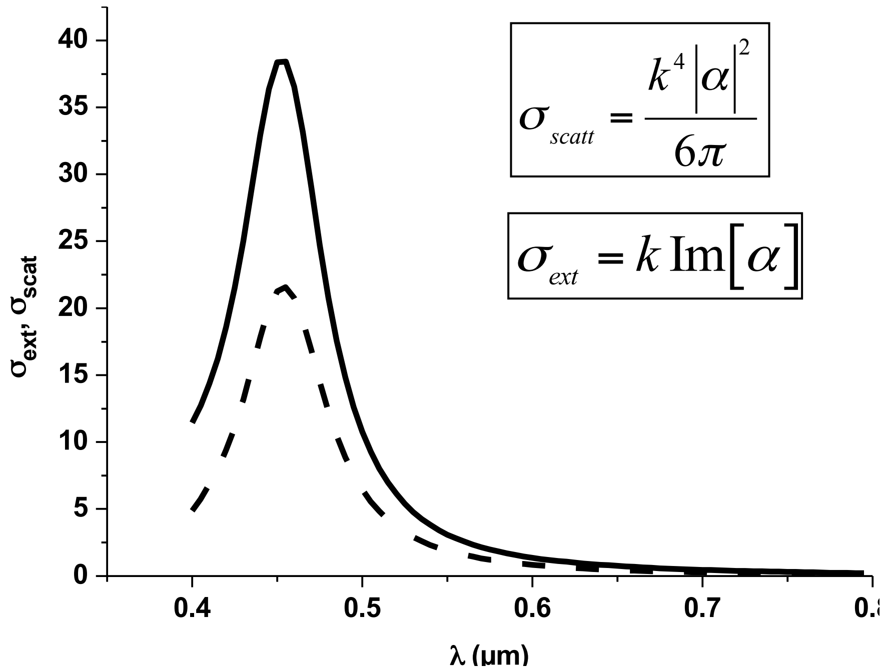
Metal

Dielectrics

Dipolar model of equivalence

$$\alpha_e = -\frac{6\pi}{ik^3} a_1$$

$$a_1 = \frac{j_1(z_b)}{h_1^+(z_b)} \frac{\varepsilon_s \varphi_1(z_b) - \varepsilon_b \varphi_1(z_s)}{\varepsilon_s \varphi_1^+(z_b) - \varepsilon_b \varphi_1(z_s)}$$



$$\sigma_{scatt} = \frac{k^4 |\alpha|^2}{6\pi}$$

$$\sigma_{ext} = k \operatorname{Im}[\alpha]$$

Equivalence:

$$a_1(z_0, \varepsilon_{in}) = a_1(z_0, \varepsilon_{eq})$$

$$\frac{\varepsilon_{in}\varphi_1(z_0) - \varphi_1(z_{in})}{\varepsilon_{in}\varphi_1^+(z_0) - \varphi_1(z_{in})} = \frac{\varepsilon_{eq}\varphi_1(z_0) - \varphi_1(z_{eq})}{\varepsilon_{eq}\varphi_1^+(z_0) - \varphi_1(z_{eq})}$$

$$\varphi_1(z) \equiv \frac{[z j_1(z)]'}{j_1(z)}$$

Equation to solve:

$$\frac{\varphi_1(z_{in})}{\varepsilon_{in}} = \frac{\varphi_1(z_{eq})}{\varepsilon_{eq}}$$

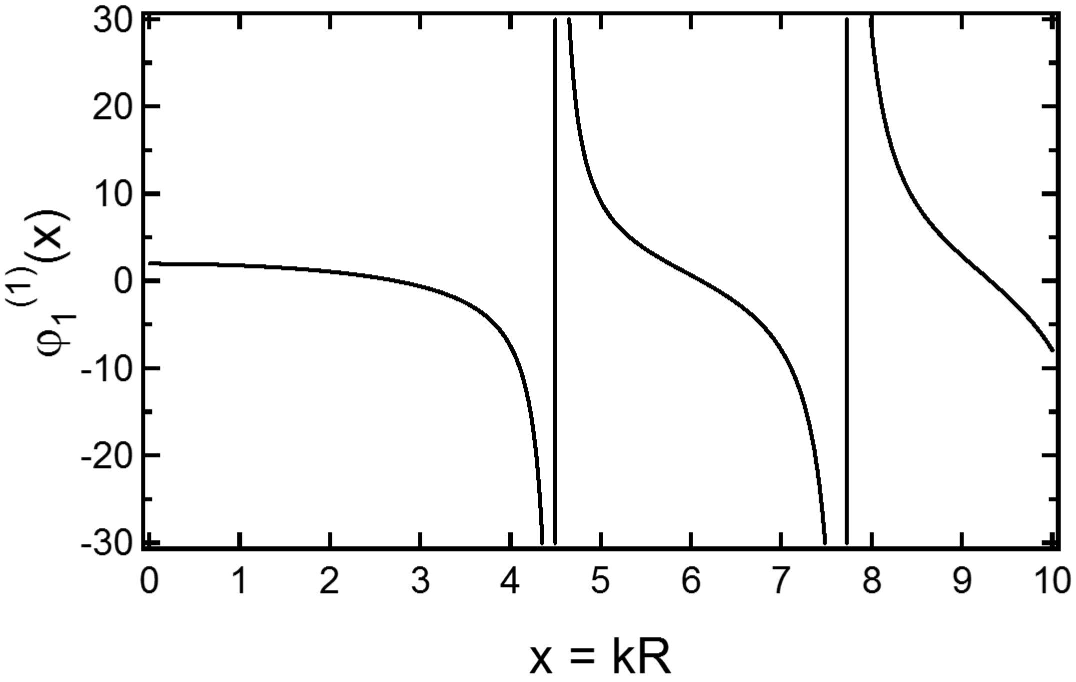
→ Numerical solving

→ **Analytic solving**

The φ_n functions

$$\frac{\varphi_1(z_{in})}{\varepsilon_{in}} = \frac{\varphi_1(z_{eq})}{\varepsilon_{eq}}$$

$$\varphi_1(z) \equiv \frac{[z j_1(z)]'}{j_1(z)}$$



- $\varphi_1^{(1)}$ is a quasi-periodic function and looks like a tan function
- $\varphi_1^{(1)}$ has poles on the real axis which are the zeros of the Bessel functions

Numerical solving: recursivity

$$\varphi_{n-1}^{(1)}(z) = n - \frac{1}{\varphi_n^{(1)}(z) + n/z^2}$$

$$\varphi_n^{(+)}(z) = \frac{1}{n/z^2 - \varphi_{n-1}^{(+)}(z)} - n$$

No clear physical insight

Small & dipolar particles

$$j_1(z) = \frac{\sin(z)}{z^2} - \frac{\cos(z)}{z}$$

$$\varphi_1^{(1)}(z) = 1 + z\frac{j_1'(z)}{j_1(z)}$$

As $kR = z << 1$: The Taylor Expansion seems a natural way

$$j_1(z) = z/3 + \frac{z^2}{30} + o(z^2)$$

$$\varphi_1^{(+)}(z) = 2 - \frac{z^2}{5} + o(z^2)$$

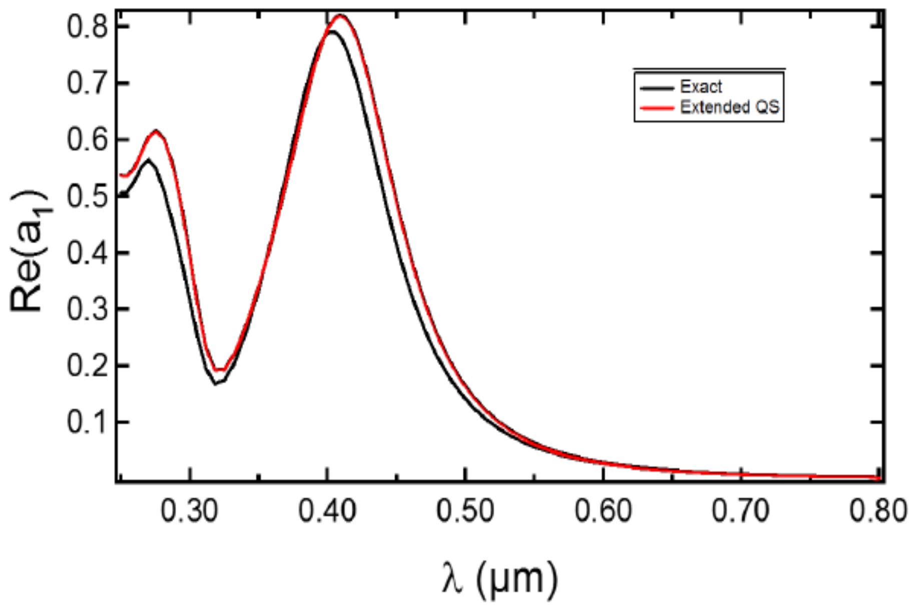
Taylor expansion

$$a_1(z) = -\frac{2iz^3}{3}\frac{\varepsilon_{21}-1}{\varepsilon_{21}+2} - \frac{2iz^5}{5}\frac{(\varepsilon_{21}-2)(\varepsilon_{21}-1)}{\varepsilon_{21}+2} + \frac{4z^6}{9}\left(\frac{\varepsilon_{21}-1}{\varepsilon_{21}+2}\right) + O(x^7)$$

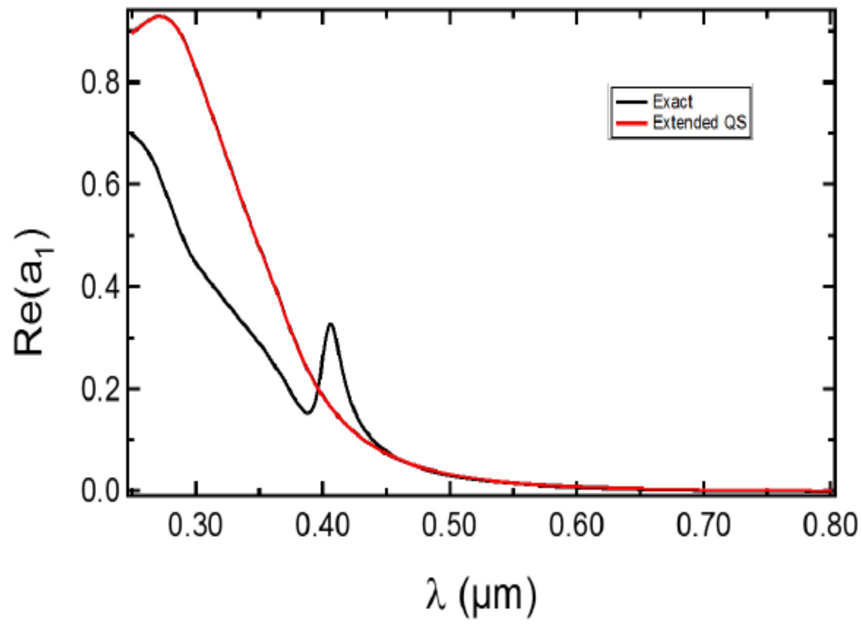
$$b_1(z) = -\frac{iz^5}{45}(\varepsilon_{21}+2) + O(x^7)$$

As in the quasi-static models, no explicit resonance appears for dielectric materials.

Taylor expression & polarisability



Ag particle, D = 100 nm



Si particles, D = 100 nm

$$\left(a_1^{(\mathrm{G.f.})}\right)^{-1} = \left(a_1^{(\mathrm{q.s.})}\right)^{-1} - \frac{3i}{2z} + 1,$$

Weierstrass factorization of the Bessel function

$$j_n(z) = \frac{z^n}{(2n+1)!!} \prod_{k=1}^{\infty} \left(1 - \left(\frac{z}{a_{n,k}}\right)^2\right)$$

G. Watson, A treatise on the theory of Bessel functions, (Cambridge University Press, 1944)

Expression of $\varphi_n^{(1)}$

$$\varphi_n^{(1)}(z) = \frac{[z j_n(z)]'}{j_n(z)} = z\,[ln(z j_n(z))]'$$

$$\varphi_n^{(1)}(z) = n + 1 + \sum_{k=1}^{\infty} \frac{2z^2}{z^2 - a_{n,k}^2}$$

where $a_{n,k}$ are the zeros of the j_n function

Analytic solution of the equivalence problem

Special functions in the multipolar Mie theory:

$$\frac{\varphi_1(z_{in})}{\varepsilon_{in}} = \frac{\varphi_1(z_{eq})}{\varepsilon_{eq}} \qquad \varphi_1(z) \equiv \frac{[z j_1(z)]'}{j_1(z)}$$

Factorization of the Bessel function:

$$j_n(z) = \frac{z^n}{(2n+1)!!} \prod_{k=1}^{\infty} \left(1 - \frac{z^2}{a_{n,\alpha}^2}\right)$$

$a_{n,\alpha}$:zeros of Besssel functions (and poles of functions φ)

G. Watson, A treatise on the theory of Bessel functions, (Cambridge University Press, 1944)

To get:

$$\varphi_n(z) = n + 1 + \sum_{\alpha=1}^{\infty} \frac{2z^2}{z^2 - a_{n,\alpha}^2} \qquad \varphi_1(z) \simeq 2\frac{1-(z/b)^2}{1-(z/a)^2}$$

The zeros of the Bessel functions φ

a and b are 2 constants: $a = a_{1,1} = 4,493$; $b = a_{1,1}/\sqrt{2} = 3.177$

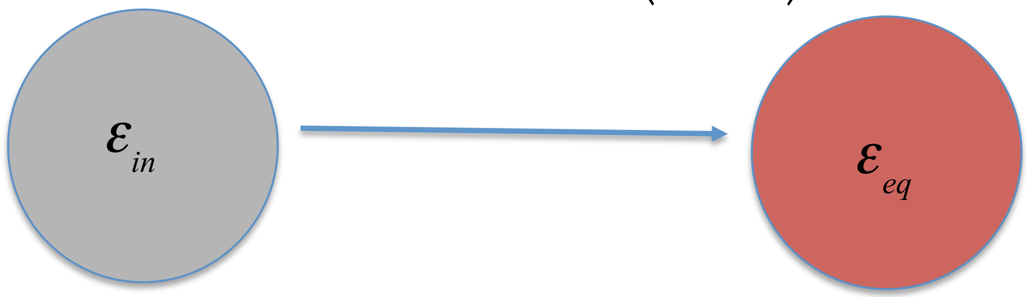
Analytic relation between dielectrics and metals

2nd order polynomial equation:

$$\varepsilon_{in}\frac{1-\varepsilon_{eq}\left(z_0/b\right)^2}{1-\varepsilon_{eq}\left(z_0/a\right)^2}=\varepsilon_{eq}\frac{1-\varepsilon_{in}\left(z_0/b\right)^2}{1-\varepsilon_{in}\left(z_0/a\right)^2}$$

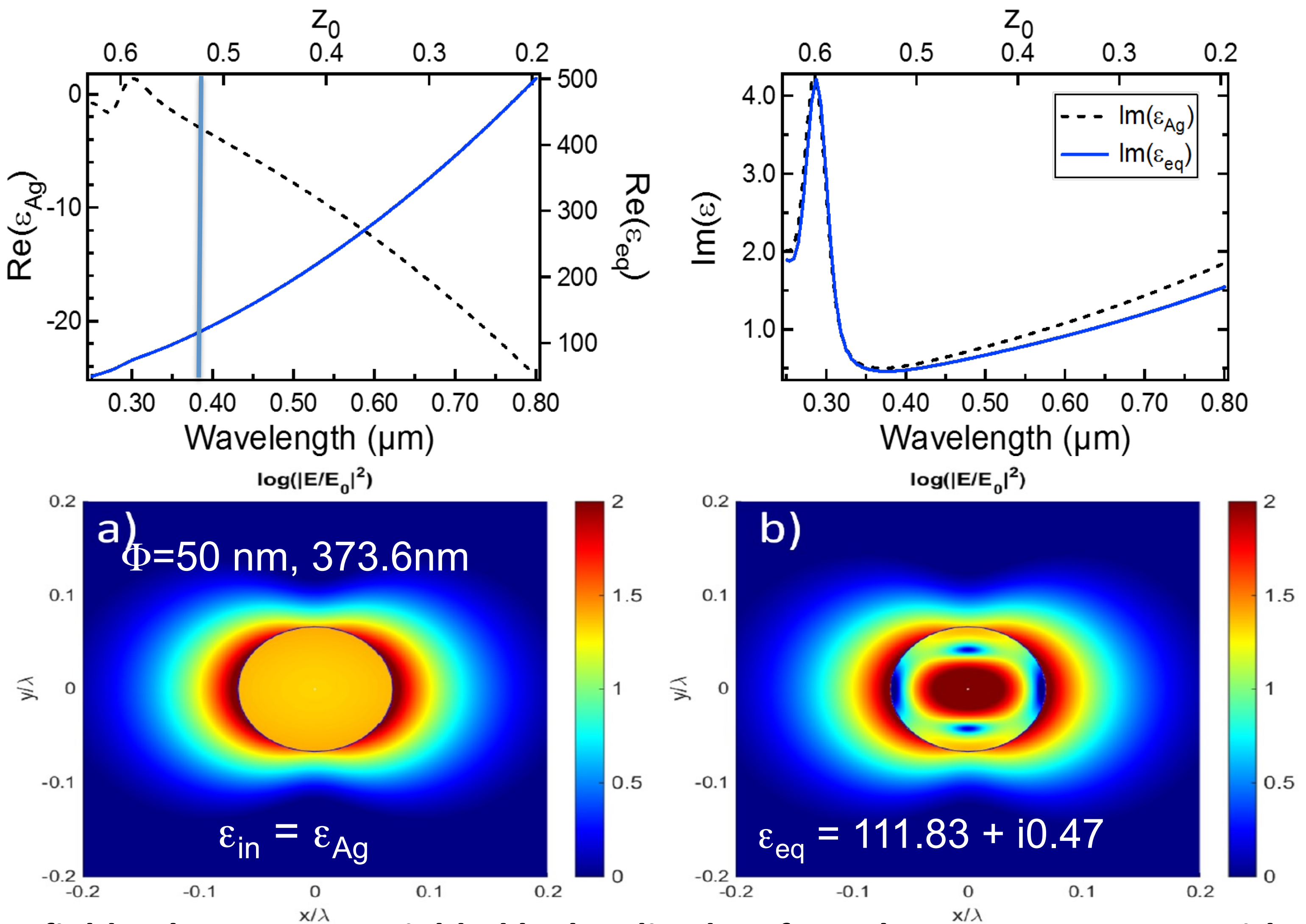
The non trivial solution is:

$$\varepsilon_{eq}=\left(\frac{a}{z_0}\right)^2\frac{1-\varepsilon_{in}\left(z_0/a\right)^2}{1-\varepsilon_{in}\left(z_0/b\right)^2}$$



A. Devilez et al., Phys. Rev. B 92, 241412(R) (2015)

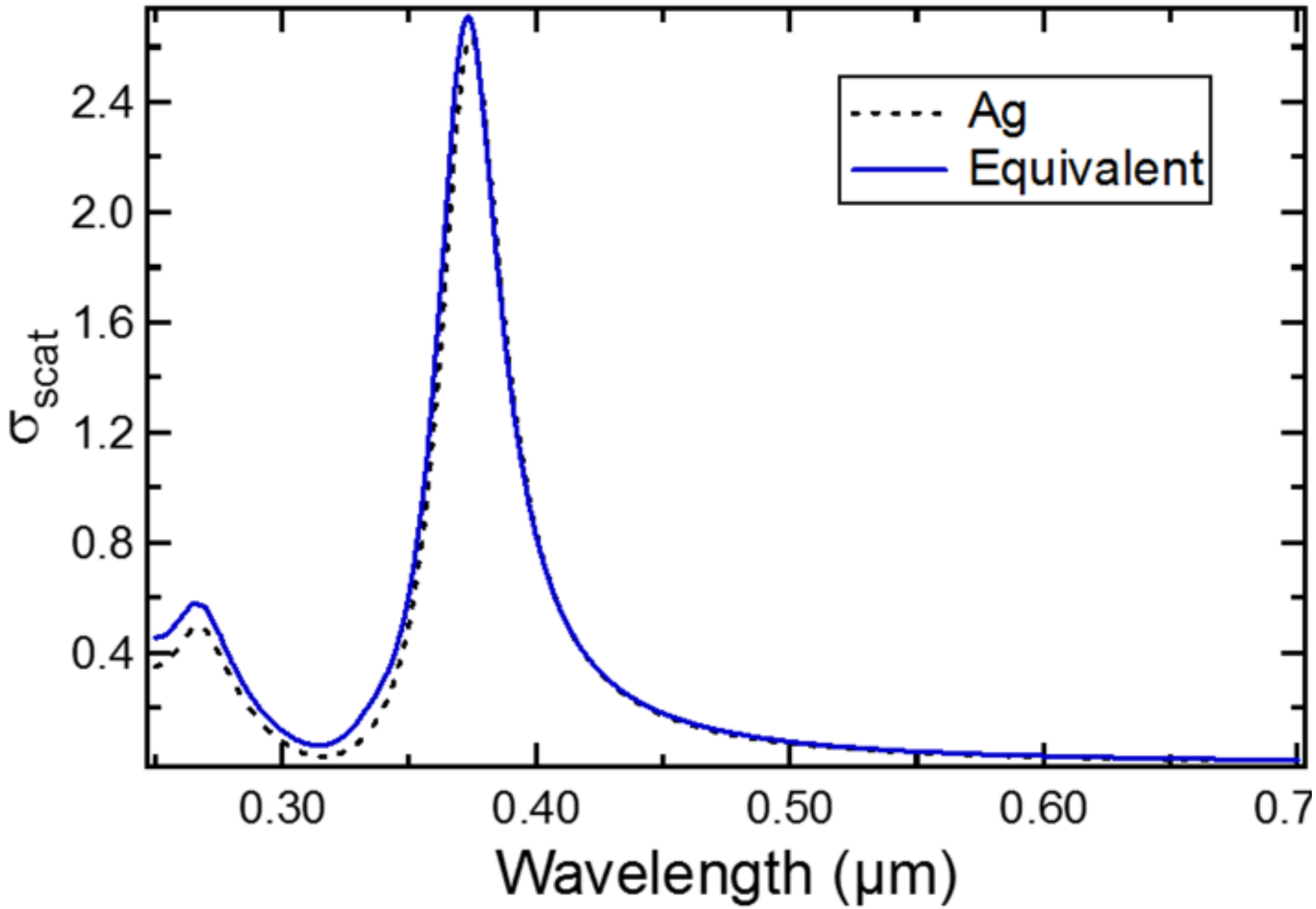
Metal/dielectric equivalence



The strong field enhancements yielded by localized surface plasmon resonances with metallic particles in the visible spectrum can be reproduced by dielectric particles in other frequency domains

A. Devilez et al., Phys. Rev. B 92, 241412(R) (2015)

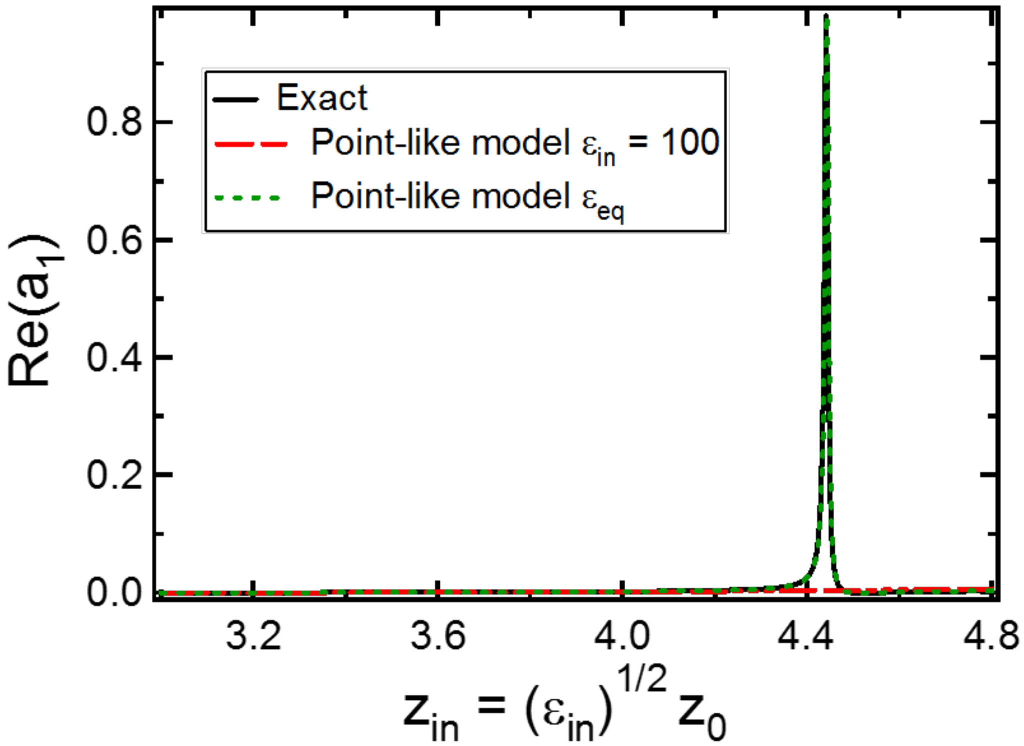
Far field properties



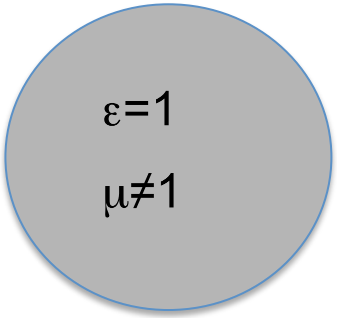
Particle polarizability

$$\left(a_1^{(\mathrm{G.f.})}\right)^{-1} = \left(a_1^{(\mathrm{q.s.})}\right)^{-1} - \frac{3i}{2z} + 1,$$

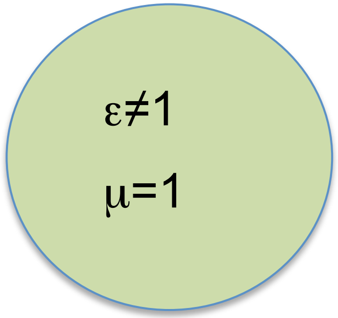
$$\varepsilon_{eq} = \left(\frac{a}{z_0}\right)^2 \frac{1-\varepsilon_{in}\left(z_0/a\right)^2}{1-\varepsilon_{in}\left(z_0/b\right)^2}$$



Localized magnetic « plasmons »



$$z_0 = k_0 r$$
$$r << \lambda$$



$$b_n = \frac{j_n\left(z_b\right)}{h_n^+\left(z_b\right)} \frac{\mu_s \varphi_n\left(z_b\right) - \mu_b \varphi_n\left(z_s\right)}{\mu_s \varphi_n^+\left(z_b\right) - \mu_b \varphi_n\left(z_s\right)}$$

$$b_1(z_0, \varepsilon_{eq}) = b_1(z_0, \mu_{in})$$

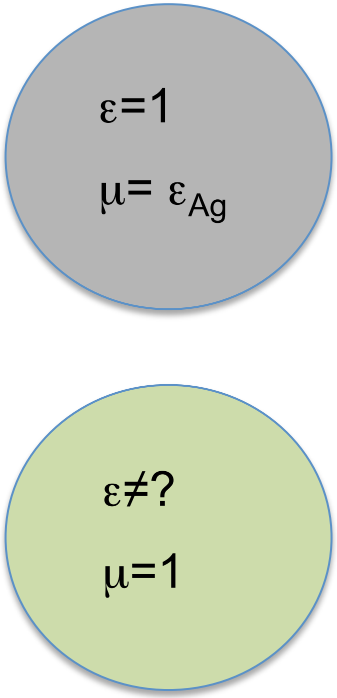
We get a 2nd order polynomial equation whose non trivial solution can be cast:

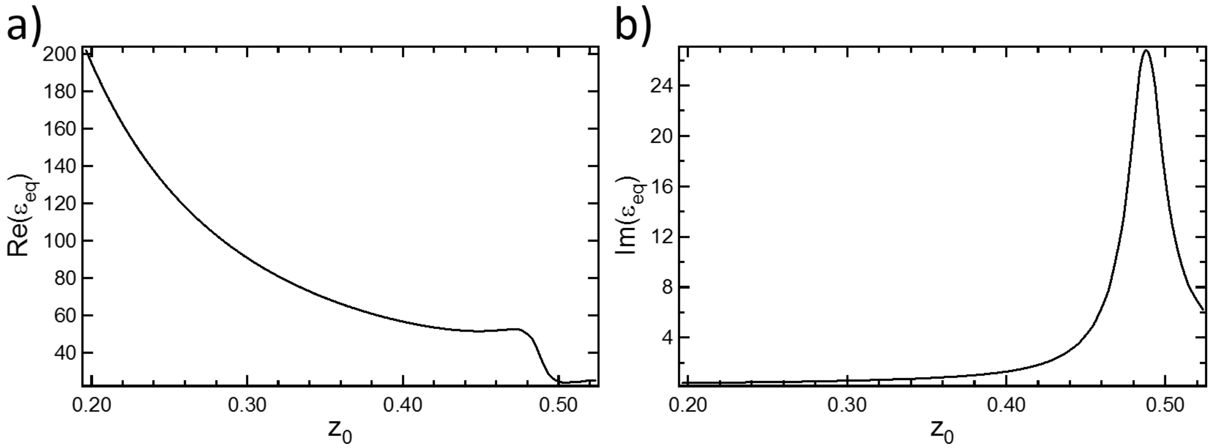
$$\varphi_1\left(z_{eq}\right) = \frac{\varphi_1\left(z_{in}\right)}{\mu_{in}}$$

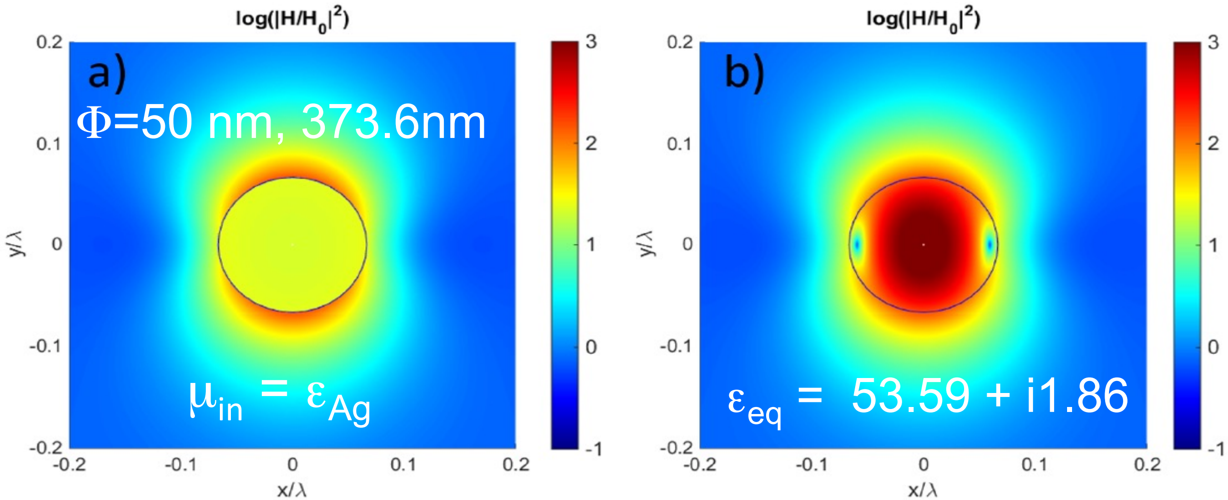
$$\varepsilon_{eq} = -\frac{\left(\mu_{in} - 1 - \mu_{in}^2\left(z_0/a\right)^2 + \mu_{in}^2\left(z_0/b\right)^2\right)}{-\mu_{in}^2\left(z_0/b\right)^2 - \mu_{in}^2\left(z_0/a\right)^2\left(z_0/b\right)^2 + \mu_{in}^2\left(z_0/b\right)^2\left(z_0/a\right)^2 + \left(z_0/a\right)^2}$$

A. Devilez, X. Zambrana-Puyalto, B. Stout, N. Bonod, Arxiv 1506.09180 (2015)

Magnetic ‘plasmon’ resonance







Dielectric material can reproduce the electromagnetic resonances of exotic materials with $\mu<0$

A. Devilez, X. Zambrana-Puyalto, B. Stout, N. Bonod, Arxiv 1506.09180 (2015)

Conclusions

- Two particles made of different materials can scatter the same electromagnetic fields

- Dielectric particles, characterized by positive dielectrics (no free electrons) can yield the same fields than metallic particles (with free electrons).

- Dielectric particles can yield the same fields than those produced by exotic magnetic metals (characterized by negative permeability)

- Dielectric particles can enhance the electric and the magnetic fields at a sub-λ scale.

Thanks for your attention

Purcell factor of silicon Mie resonators

Purcell Factor $\frac{\Gamma(\omega)}{\Gamma_0(\omega)} \equiv \frac{6\pi c^3}{\omega^3}\frac{Q}{V}$

Defined by E.M. Purcell in the radio regime (Phys. Rev. 69, 681 (1946))

-Dipole located in the field's maximum

-Dipole 'oriented' along the field's polarization

-Only one resonant mode in the cavity

In nanophotonics:

-the state of the resonator cannot be described with only 1 electromagnetic mode.

-The definition of effective volume $\int_V |\mathbf{E}_\mu(\mathbf{r},\omega_\mu)|^2 \mathrm{d}V$ does not work very well

A. F. Koenderink, Opt. Lett. 35, 4208 (2010)

If a suitable normalization is used, a **modal expression** can be used:

$$F(\omega) = \frac{6\pi c}{|p|^2\omega}\mathrm{Im}\{\mathbf{p}^* \cdot \overleftrightarrow{\mathbf{G}}^{(e)}(\mathbf{r_0},\mathbf{r_0}) \cdot \mathbf{p}\}$$

$$= \frac{3\pi c^3}{\omega}\sum_\mu \mathrm{Im}\left\{\frac{1}{V_\mu\omega_\mu(\omega_\mu - \omega)}\right\}$$

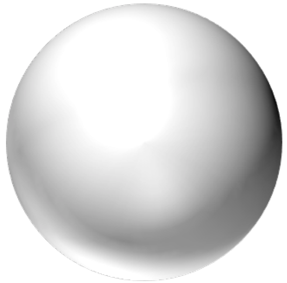
with $V_\mu = \frac{1}{(\mathbf{u_p} \cdot \mathbf{E}_\mu(\mathbf{r_p},\omega_\mu))^2}$

C. Sauvan, J.-P. Hugonin, I. Maksymov, and P. Lalanne, Phys. Rev. Lett. **110**, 237401 (2013).
P. T. Kristensen and S. Hughes, ACS Photon. **1**, 2 (2014).
E. A. Muljarov, M. B. Doost, and W. Langbein, arXiv:1409.6877.

Multipolar fields

System: Homogeneous, isotropic sphere embedded in a homogeneous, isotropic, lossless medium

Eigenmodes of the system are called **multipolar Fields:**



$$J^2[\mathbf{E}^{(x)}_{j,m_z,q}] = j(j+1)\mathbf{E}^{(x)}_{j,m_z,q}$$

$$J_z[\mathbf{E}^{(x)}_{j,m_z,q}] = m_z\mathbf{E}^{(x)}_{j,m_z,q}$$

$$\Pi[\mathbf{E}^{(x)}_{j,m_z,q}] = \begin{cases}(-)^j\mathbf{E}^{(m)}_{j,m_z,q}\\ (-)^{j+1}\mathbf{E}^{(e)}_{j,m_z,q}.\end{cases}$$

Eigenfrequencies can be obtained with a transcendental equation that does not depend on m_z, so:

$$\omega^{(x)}_{j,m_z,q} = \omega^{(x)}_{j,q}$$

The parameter q orders the real part of the frequency:

$$\omega^{(x)}_{j,1} < \omega^{(x)}_{j,2} < \omega^{(x)}_{j,3} < \ldots$$

M. E. Rose, Multipole Fields (Wiley, New York, 1955)

Eigenfrequencies and eigenvectors

Transcendental equations. 1 for x=e, 1 for x=m

↓

$\omega_{j,q}^{(x)}$ Complex eigenfrequencies

↓

$$\mathbf{E}_{j,m_z,q}^{(m)} = A_j^{(m)} R_j\left(k_{j,q}^{(m)}\right)\left[\frac{1}{\sin\theta}\frac{\partial Y_{j,m_z}}{\partial\phi}\hat{\theta} - \frac{\partial Y_{j,m_z}}{\partial\theta}\hat{\phi}\right]$$

$$\mathbf{E}_{j,m_z,q}^{(e)} = \frac{A_j^{(e)}}{\epsilon(r)k_{j,q}^{(e)}r}\left[j(j+1)R_j\left(k_{j,q}^{(e)}\right)Y_{j,m_z}\hat{\mathbf{r}} + \frac{\partial\left(rR_j\left(k_{j,q}^{(e)}\right)\right)}{\partial r}\frac{\partial Y_{j,m_z}}{\partial\theta}\hat{\theta} + \frac{\partial\left(rR_j\left(k_{j,q}^{(e)}\right)\right)}{\partial r}\frac{1}{\sin\theta}\frac{\partial Y_{j,m_z}}{\partial\phi}\hat{\phi}\right]$$

Complex eigenvectors

X. Zambrana-Puyalto et al., Phys. Rev. B 91, 195422 (2015)

Modal expression of the Purcell factor

$$F(\omega) = \frac{6\pi c}{|p|^2\omega}\mathrm{Im}\{\mathbf{p}^* \cdot \overleftrightarrow{\mathbf{G}}^{(e)}(\mathbf{r_0},\mathbf{r_0}) \cdot \mathbf{p}\}$$

$$= \frac{3\pi c^3}{\omega}\sum_{\mu}\mathrm{Im}\left\{\frac{1}{V_\mu\omega_\mu(\omega_\mu - \omega)}\right\}$$

where the effective volume is computed as: $\frac{1}{V^{(x)}_{j,m_z,q}} = \left(p.E^{(x)}_{j,m_z,q}\right)^2$

Multimodal expression :

$$F(\omega) = \frac{3\pi c^3}{\omega}\sum_{j,m_z,q}\left[\underbrace{\mathrm{Im}\left\{\frac{1}{V^{(e)}_{j,m_z,q}\omega^{(e)}_{j,q}(\omega^{(e)}_{j,q} - \omega)}\right\}}_{\text{Electric modes}} + \underbrace{\mathrm{Im}\left\{\frac{1}{V^{(e)}_{j,m_z,q}\omega^{(e)}_{j,q}(\omega^{(e)}_{j,q} - \omega)}\right\}}_{\text{Magnetic modes}}\right]$$

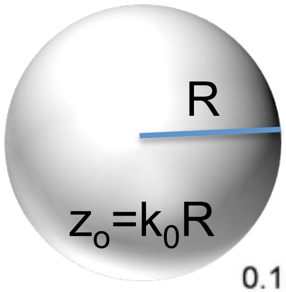
X. Zambrana-Puyalto et al., Phys. Rev. B 91, 195422 (2015)

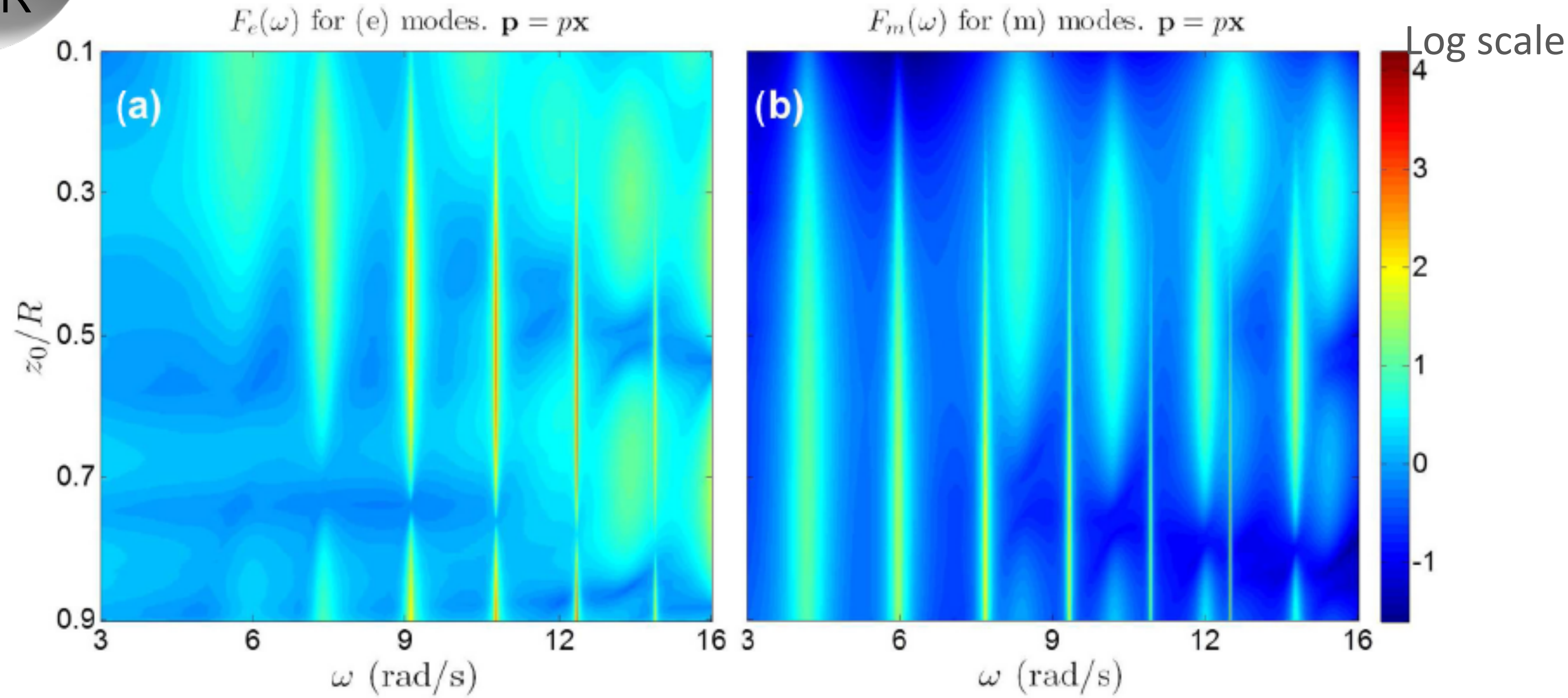
Purcell factor with respect to the dipole position

$$F(\omega) = \frac{3\pi c^3}{\omega} \sum_{j,m_z,q} \left[\mathrm{Im}\left\{ \frac{1}{V^{(e)}_{j,m_z,q}\,\omega^{(e)}_{j,q}(\omega^{(e)}_{j,q} - \omega)} \right\} + \mathrm{Im}\left\{ \frac{1}{V^{(e)}_{j,m_z,q}\,\omega^{(e)}_{j,q}(\omega^{(e)}_{j,q} - \omega)} \right\} \right]$$

Electric modes

Magnetic modes



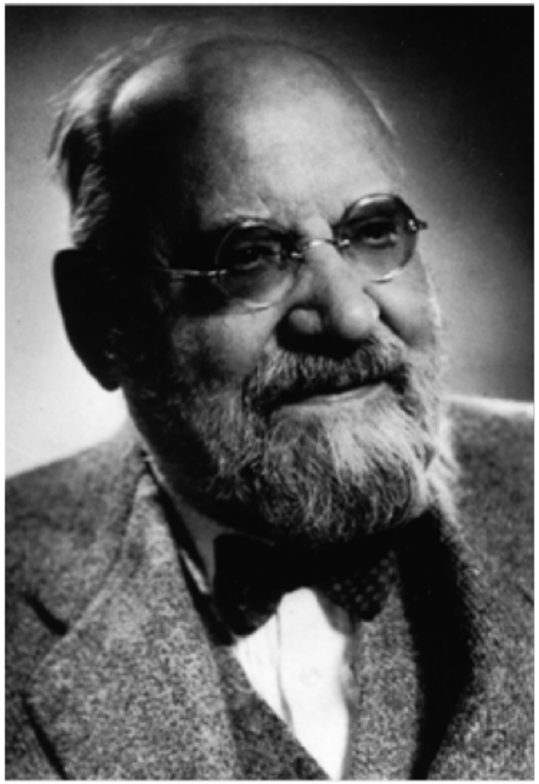


$$\left\langle V^{(x)}_{j,m_z,q} \right\rangle = \int_0^R \int_\Omega \frac{3r^2 \mathrm{d}r \mathrm{d}\Omega}{[\hat{\mathbf{r}} \cdot \mathbf{E}^{(x)}_{j,m_z,q}]^2 + [\hat{\theta} \cdot \mathbf{E}^{(x)}_{j,m_z,q}]^2 + [\hat{\phi} \cdot \mathbf{E}^{(x)}_{j,m_z,q}]^2}$$

X. Zambrana-Puyalto et al., Phys. Rev. B 91, 195422 (2015)

Multipolar Mie theory

Solution of Maxwell equations in spherical coordinates (1908)



Gustav Mie
1868 -1957

Scalar solutions in spherical coordinates

Helmoltz equation in a homogenous, isotropic medium, no source:

$$\Delta \mathcal{A}(\mathbf{r}, \omega) + k_0^2 \mathcal{A}(\mathbf{r}, \omega) = 0$$

The outgoing waves are defined as:

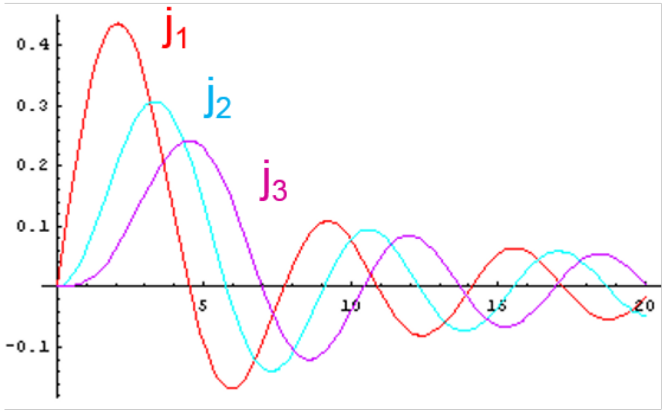
$$\mathcal{A}_{n,m}(k\mathbf{r}) = h_n^{(+)}(kr) Y_{n,m}(\theta, \phi)$$

The regular waves without any singularity at r=0 are defined as:

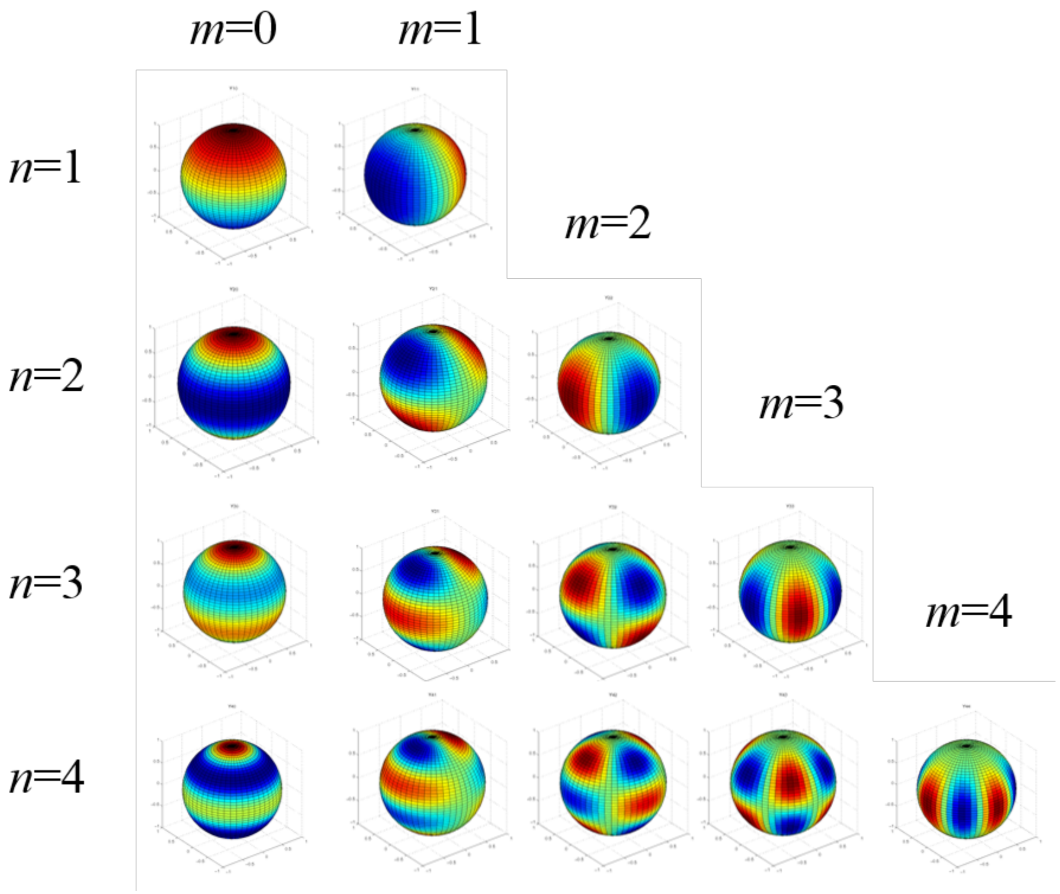
$$Rg\left\{\mathcal{A}_{n,m}(k\mathbf{r})\right\} = j_n(kr) Y_{n,m}(\theta, \phi)$$

Illustration of the spherical wave function basis

Bessel functions $j_n(r)$



Spherical Harmonics : $Y_{n,m}(\theta,\phi)$



Bessel functions describe the radial dependency, while the spherical harmonics describe the angular dependency.

Vector Partial Wave Functions

$$\mathbf{M}_{n,m}(k\mathbf{r}) \equiv \frac{\nabla \times [\mathbf{r}\mathcal{A}_{n,m}(k\mathbf{r})]}{k}$$

$$\mathbf{N}_{n,m}(k\mathbf{r}) \equiv \frac{\nabla \times \mathbf{M}_{n,m}(k\mathbf{r})}{k}$$

$$Rg\left\{\mathbf{M}_{n,m}(kr)\right\} \equiv \frac{\nabla \times [\mathbf{r}Rg\left\{\mathcal{A}_{n,m}(k\mathbf{r})\right\}]}{k}$$

$$Rg\left\{\mathbf{N}_{n,m}(kr)\right\} \equiv \frac{\nabla \times Rg\left\{\mathbf{M}_{n,m}(kr)\right\}}{k}$$

E and H field expansions on Vector Spherical Wave functions

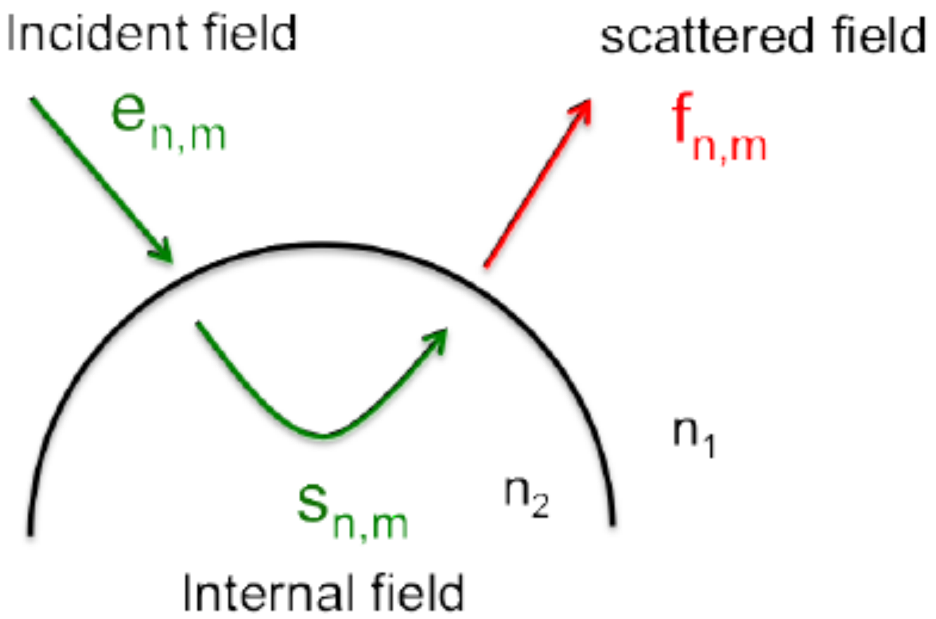
$$\mathbf{E}^{+} = E \sum \mathbf{M}_{n,m}(k\mathbf{r}) f_{n,m}^{(h)} + \mathbf{N}_{n,m}(k\mathbf{r}) f_{n,m}^{(e)}$$

$$\mathbf{E} = E \sum Rg\left\{\mathbf{M}_{n,m}(k\mathbf{r})\right\} e_{n,m}^{(h)} + Rg\left\{\mathbf{N}_{n,m}(k\mathbf{r})\right\} e_{n,m}^{(e)}$$

$$\mathbf{H}^{+} = \frac{kE}{i\omega\mu_0\mu} \sum \mathbf{N}_{n,m}(k\mathbf{r}) f_{n,m}^{(h)} + \mathbf{M}_{n,m}(k\mathbf{r}) f_{n,m}^{(e)}$$

$$\mathbf{H} = \frac{kE}{i\omega\mu_0\mu} \sum Rg\left\{\mathbf{N}_{n,m}(k\mathbf{r})\right\} e_{n,m}^{(h)} + Rg\left\{\mathbf{M}_{n,m}(k\mathbf{r})\right\} e_{n,m}^{(e)}$$

Transfert Matrix



$$\begin{bmatrix} f_{n,m}^{(h)} \\ f_{n,m}^{(e)} \end{bmatrix} = \begin{bmatrix} t_n^{(h)} & 0 \\ 0 & t_n^{(e)} \end{bmatrix} \begin{bmatrix} e_{n,m}^{(h)} \\ e_{n,m}^{(e)} \end{bmatrix}$$

Mie scattering coefficients

$$a_n \equiv -t_n^{(e)} = \frac{j_n(k_1R)}{h_n^{(+)}(k_1R)} \frac{\left[\varepsilon_{21}\varphi_n^{(1)}(k_1R) - \varphi_n^{(1)}(k_2R)\right]}{\left[\varepsilon_{21}\varphi_n^{(+)}(k_1R) - \varphi_n^{(1)}(k_2R)\right]}$$

$$b_n \equiv -t_n^{(h)} = \frac{j_n(k_1R)}{h_n^{(+)}(k_1R)} \frac{\left[\mu_{21}\varphi_n^{(1)}(k_1R) - \varphi_n^{(1)}(k_2R)\right]}{\left[\mu_{21}\varphi_n^{(+)}(k_1R) - \varphi_n^{(1)}(k_2R)\right]}$$