Selected Topics in Advanced Optics

Week 4 – part 1

Olivier J.F. Martin
Nanophotonics and Metrology Laboratory

EPFL

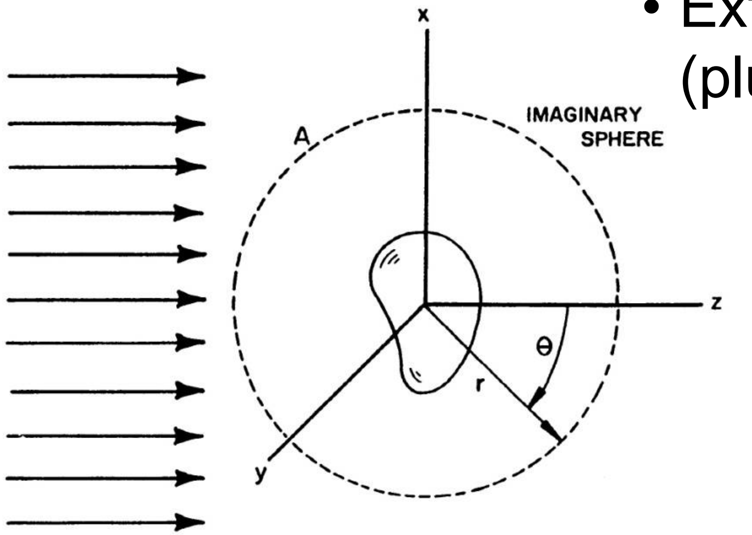
Module 3: Light scattering

- C.F. Bohren & D.R. Huffman, Absorption and scattering of light by small particles (Wiley, New York, 1983).
- H.C. van de Hulst, Light scattering by small particles (Dover, New York, 1981).
- F. Mühlig et al., «Multipole analysis of meta-atoms», *Metamaterials* vol. 5, p. 64 (2011).

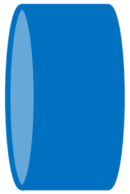
Light scattering determines our perception of the world

Scattering experiment

- When an object is positioned on an incident beam, the amount of measured energy decreases (extinction)



- Extinction = scattering + absorption by the particle (plus possibly absorption in the surrounding medium)



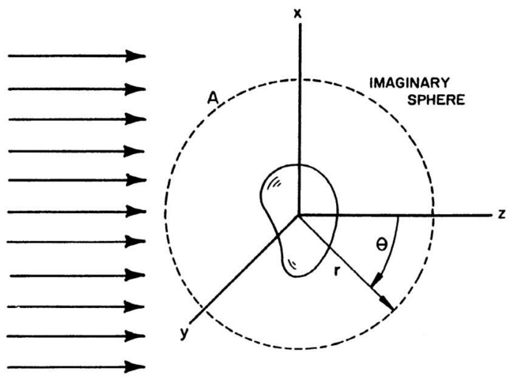
- Power flow across a closed surface around the scatterer:

$$W_{abs} = -\int_A dA\,\mathbf{S}\cdot\mathbf{e}_r$$

- $W_{abs} > 0$ and corresponds to the power absorbed in the object (if energy is generated in the object, then $W_{abs} < 0$)

Scattering experiment

- From the electromagnetic field around the object, we can obtain the Poynting vector
- The time-averaged Poynting vector can be written outside the object as sum of incident and scattered fields, as well as the interaction between the two (extinction):



$$\mathbf{S} = \mathbf{S}_i + \mathbf{S}_{scat} + \mathbf{S}_{ext}$$

$$\mathbf{S}_i = \frac{1}{2}\mathrm{Re}\left\{\mathbf{E}_i \times \mathbf{H}_i^*\right\} \qquad \mathbf{S}_{scat} = \frac{1}{2}\mathrm{Re}\left\{\mathbf{E}_{scat} \times \mathbf{H}_{scat}^*\right\}$$

$$\mathbf{S}_{ext} = \frac{1}{2}\mathrm{Re}\left\{\mathbf{E}_i \times \mathbf{H}_{scat}^* + \mathbf{E}_{scat} \times \mathbf{H}_i^*\right\}$$

Scattering experiment

- Combining these different components of the Poynting vector, we define the different energy flows:

$$W_i = -\int_A dA\, \mathbf{S}_i \cdot \mathbf{e}_r \qquad W_{scat} = \int_A dA\, \mathbf{S}_{scat} \cdot \mathbf{e}_r \qquad W_{ext} = -\int_A dA\, \mathbf{S}_{ext} \cdot \mathbf{e}_r$$

- And rewrite the power absorbed by the object:

$$W_{abs} = W_i - W_{scat} + W_{ext}$$

- For a non-absorbing surronding medium ($W_i = 0$), we finally have:

$$\boxed{W_{ext} = W_{abs} + W_{scat}}$$

- The extinction, absorption and scattering power flows can therefore be calculated from the electromagnetic fields.

Cross sections

- To obtain characteristic parameters that do not depend on the illumination, one normalizes with the incident irradiance I_i (intensity of the incident electromagnetic field):

$$C_{ext} = \frac{W_{ext}}{I_i} \qquad C_{abs} = \frac{W_{abs}}{I_i} \qquad C_{scat} = \frac{W_{scat}}{I_i}$$

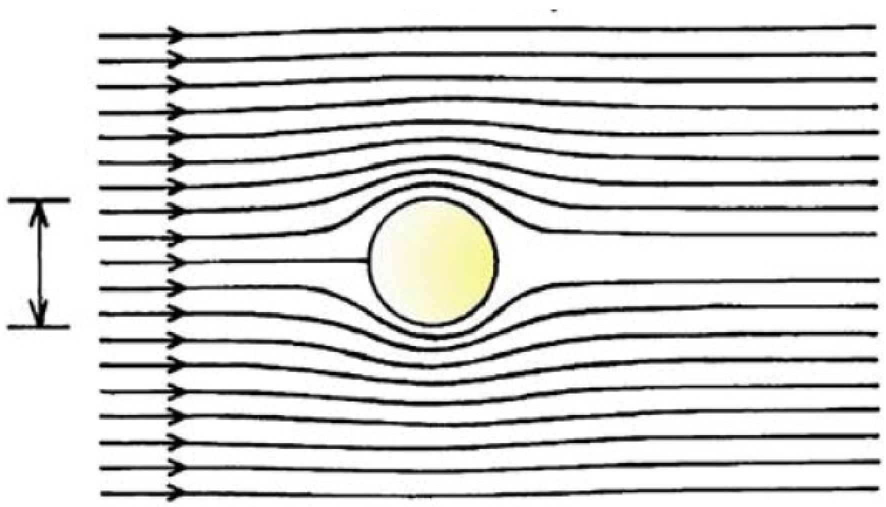
- These cross sections have the dimension of an area !
- By dividing the cross section by the geometrical cross-sectional area G ($G = \pi a^2$ for a sphere) projected onto a plane perpendicular to the incident beam, one obtains the efficiencies:

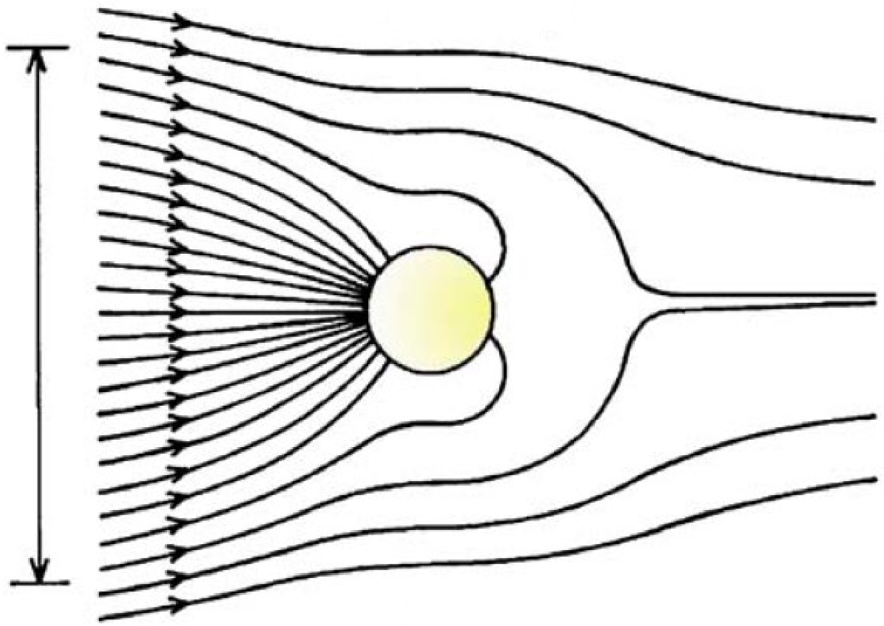
$$Q_{ext} = \frac{C_{ext}}{G} \qquad Q_{abs} = \frac{C_{abs}}{G} \qquad Q_{scat} = \frac{C_{scat}}{G}$$

- The efficiency indicates the effective optical size of the particle, compared to its real, geometrical size.

Cross sections

- The efficiency indicates the effective optical size of the particle, compared to its real, geometrical size
- The effective size can be significantly larger than the physical size
- This can be understood in terms of electromagnetic field distribution:



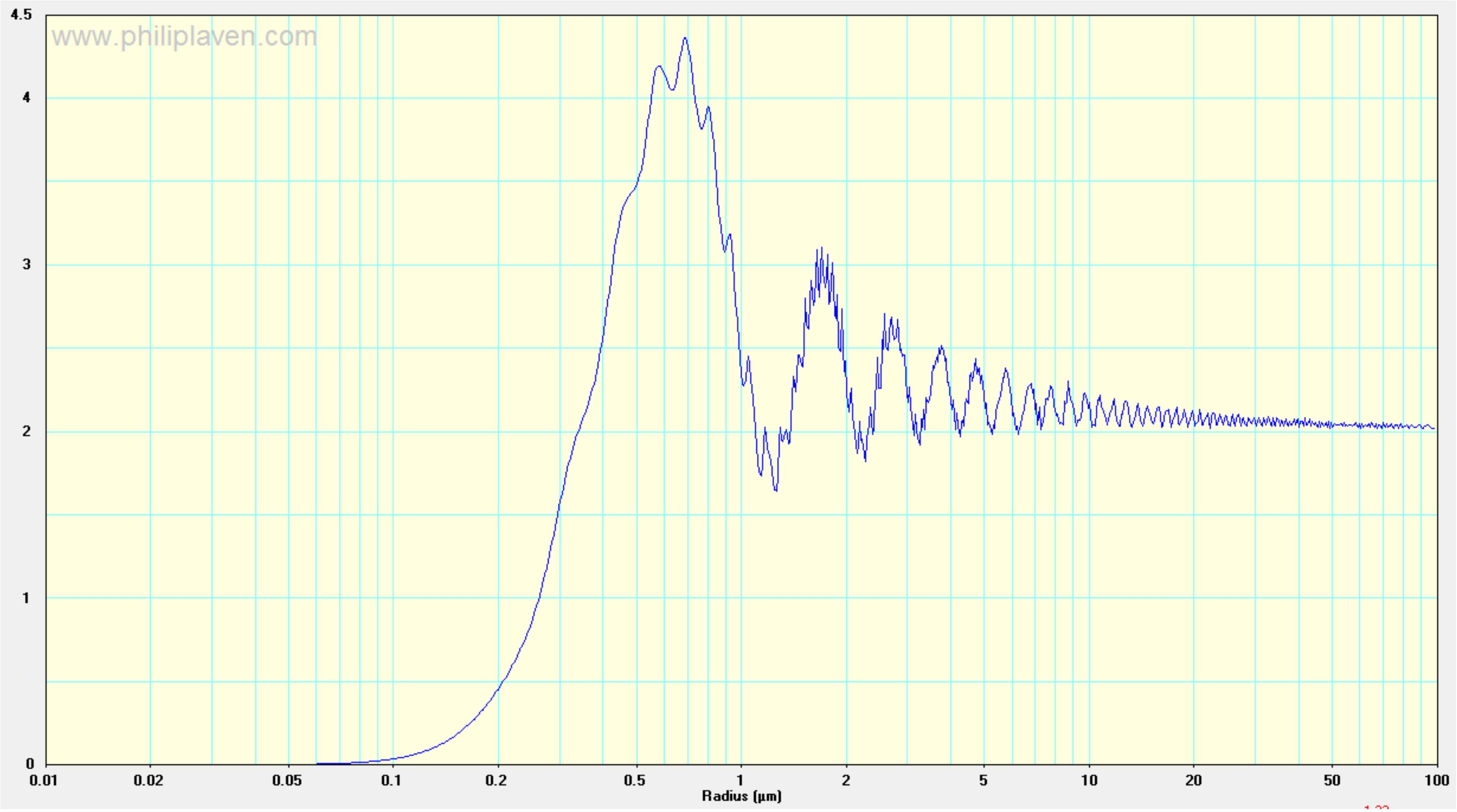


Cross sections

- Often an object appears larger than its physical size!

Sphere, λ_0=1 μm, n=1.5 (λ_{eff}=0.66 μm)

Q_{scat}



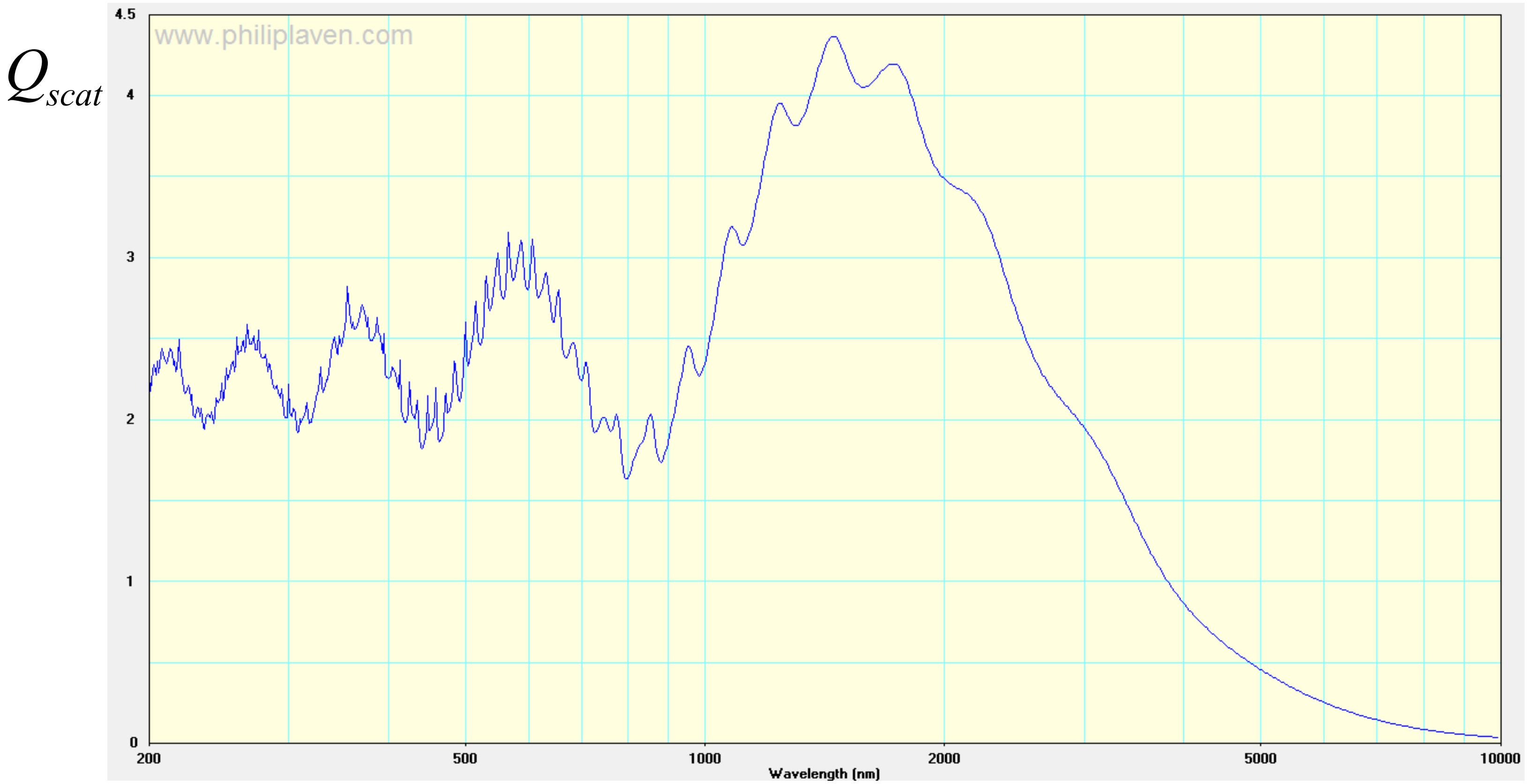
Radius (μm)

Olivier J.F. Martin

Cross sections

- This of course depends on the relative dimension of the object, compared to the wavelength

Sphere, radius=1 μm, n=1.5



Wavelength (nm)

Differential cross section

- For a given object, light is scattered differently in different directions, one defines the differential cross section $\left|\mathbf{X}(\Omega)\right|^2 / k^2$ into a solid angle $d\Omega$, such that

$$C_{scat} = \int_0^{2\pi} d\phi \int_0^{\pi} d\theta \frac{\left|\mathbf{X}(\theta,\phi)\right|^2}{k^2} = \int_{4\pi} d\Omega \frac{\left|\mathbf{X}(\Omega)\right|^2}{k^2}$$

- The differential cross section provides the amount of light scattered into a specific direction and is similar to the cross section known in atomic physics.

Selected Topics in Advanced Optics

Week 4 – part 2

Olivier J.F. Martin
Nanophotonics and Metrology Laboratory

EPFL

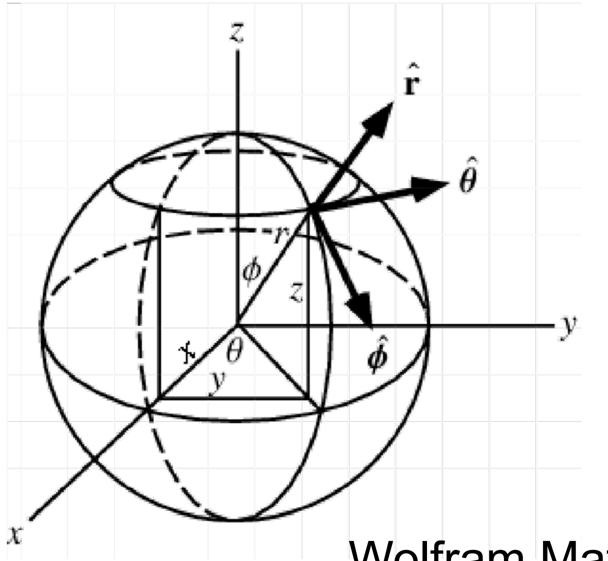
Selected Topics in Advanced Optics

Week 4 – part 3

Olivier J.F. Martin
Nanophotonics and Metrology Laboratory
EPFL

Scattering by a sphere – Mie scattering and VSH

- The Mie theory provides an exact solution for light scattering by a sphere.
- This solution relies on vector spherical harmonics (VSH): solutions of Helmholtz equation in spherical coordinates.
- It mainly consists in solving Maxwell's equations in spherical coordinates inside and outside the sphere and imposing the boundary conditions (continuity of $\boldsymbol{E}$ and $\boldsymbol{H}$ parallel and $\boldsymbol{D}$ and $\boldsymbol{B}$ perpendicular) at the interface between the sphere and the surrounding medium.
- Expressions are simple in spherical coordinates, but all axes are not equal!



Wolfram Mathworks

Scattering by a sphere – Mie scattering and VSH

- Assuming a $\exp(-j\omega t)$ time dependence, and a linear, homogeneous, isotropic medium, Maxwell's equations give us:

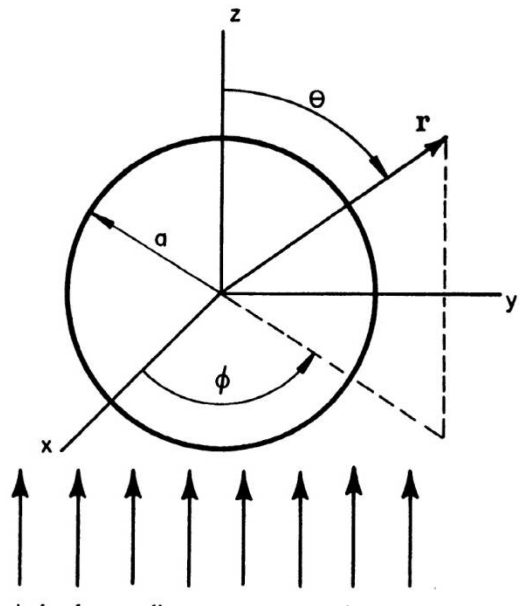
$$\nabla^2\mathbf{E}(\mathbf{r})+k^2\mathbf{E}(\mathbf{r})=0 \qquad \nabla\cdot\mathbf{E}(\mathbf{r})=0$$

$$\nabla^2\mathbf{H}(\mathbf{r})+k^2\mathbf{H}(\mathbf{r})=0 \qquad \nabla\cdot\mathbf{H}(\mathbf{r})=0$$

$$\nabla\times\mathbf{E}(\mathbf{r})=j\omega\varepsilon\mathbf{H}(\mathbf{r}) \qquad \nabla\times\mathbf{H}(\mathbf{r})=-j\omega\varepsilon\mathbf{E}(\mathbf{r})$$

- We obtain solutions for these equations in spherical coordinates using a generating scalar function in the form:

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



Scattering by a sphere – Mie scattering and VSH

- From the scalar generating function and the position vector r, one defines vectorial harmonic functions:

$$\mathbf{M}(\mathbf{r}) = \nabla \times \left(\mathbf{r} \psi(\mathbf{r}) \right) \qquad \mathbf{N}(\mathbf{r}) = \frac{\nabla \times \mathbf{M}(\mathbf{r})}{k}$$

- Which are divergence free and fulfil:

$$\nabla \cdot \mathbf{M}(\mathbf{r}) = 0$$
$$\nabla \cdot \mathbf{N}(\mathbf{r}) = 0$$
$$\nabla \times \mathbf{N}(\mathbf{r}) = k\mathbf{M}(\mathbf{r})$$

- The generating function satisfies the wave equation in spherical coordinates:

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

Scattering by a sphere – Mie scattering and VSH

- Finally, one obtains the scattered field as

$$\mathbf{E}_{sca}(r,\theta,\varphi) = \sum_{n=1}^{\infty}\sum_{m=-n}^{n} k^2 E_{nm}[a_{nm}\mathbf{N}_{nm}(r,\theta,\varphi) + b_{nm}\mathbf{M}_{nm}(r,\theta,\varphi)].$$

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

- With

$$\mathbf{M}_{nm} = \left[i\pi_{nm}(\cos\theta)\mathbf{i}_\theta - \tau_{nm}(\cos\theta)\mathbf{i}_\varphi\right] \times h_n^{(1)}(kr)\exp(im\varphi),$$

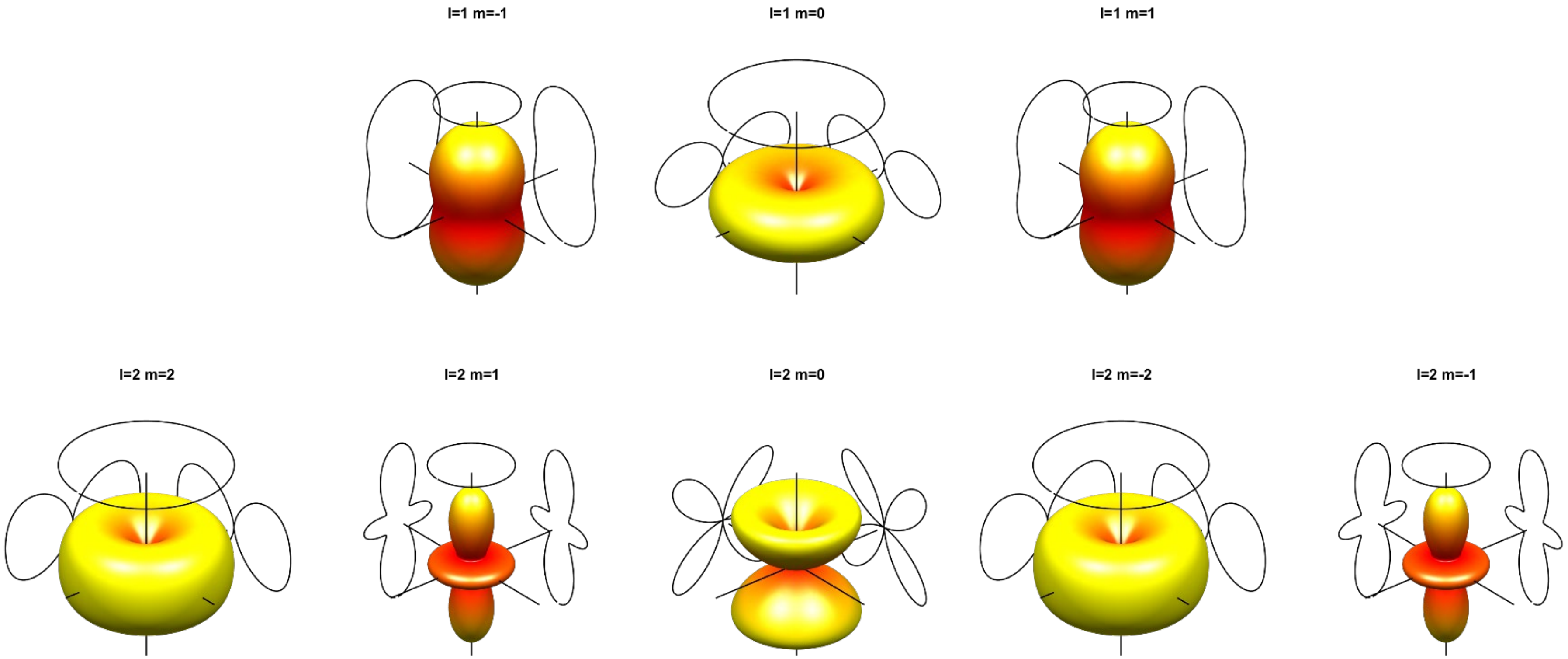
$$\mathbf{N}_{nm} = n(n+1)P_n^m(\cos\theta)\frac{h_n^{(1)}(kr)}{kr}\exp(im\varphi)\mathbf{i}_r + \left[\tau_{nm}(\cos\theta)\mathbf{i}_\theta + i\pi_{nm}(\cos\theta)\mathbf{i}_\varphi\right] \times \frac{1}{kr}\frac{d}{dr}\left[rh_n^{(1)}(kr)\right]\exp(im\varphi).$$

S. Mühlig, Metamaterials vol. 5, p. 64 (2011)

Scattering by a sphere – Mie scattering and VSH

- Radial dependence: Hankel functions of the first kind $h_n^{(1)}$
- Angular dependence: Legendre functions $$\pi_{nm}(\cos\,\theta) = \frac{m}{\sin\,\theta} P_n^m(\cos\,\theta),$$

$$\tau_{nm}(\cos\,\theta) = \frac{d}{d\theta} P_n^m(\cos\,\theta).$$



Olivier J.F. Martin

Scattering by a sphere – Mie scattering and VSH

- Expansion coefficients for a given scatterer are obtained by projecting the field on the VSH:

$$a_{nm} = \frac{\int_0^{2\pi}\int_0^{\pi} \mathbf{E}(r=a)\mathbf{N}^*_{nm}(r=a)\sin\theta\, d\theta\, d\varphi}{\int_0^{2\pi}\int_0^{\pi} |\mathbf{N}_{nm}(r=a)|^2 \sin\theta\, d\theta\, d\varphi},$$

$$b_{nm} = \frac{\int_0^{2\pi}\int_0^{\pi} \mathbf{E}(r=a)\mathbf{M}^*_{nm}(r=a)\sin\theta\, d\theta\, d\varphi}{\int_0^{2\pi}\int_0^{\pi} |\mathbf{M}_{nm}(r=a)|^2 \sin\theta\, d\theta\, d\varphi}.$$

- Scattering cross-section:

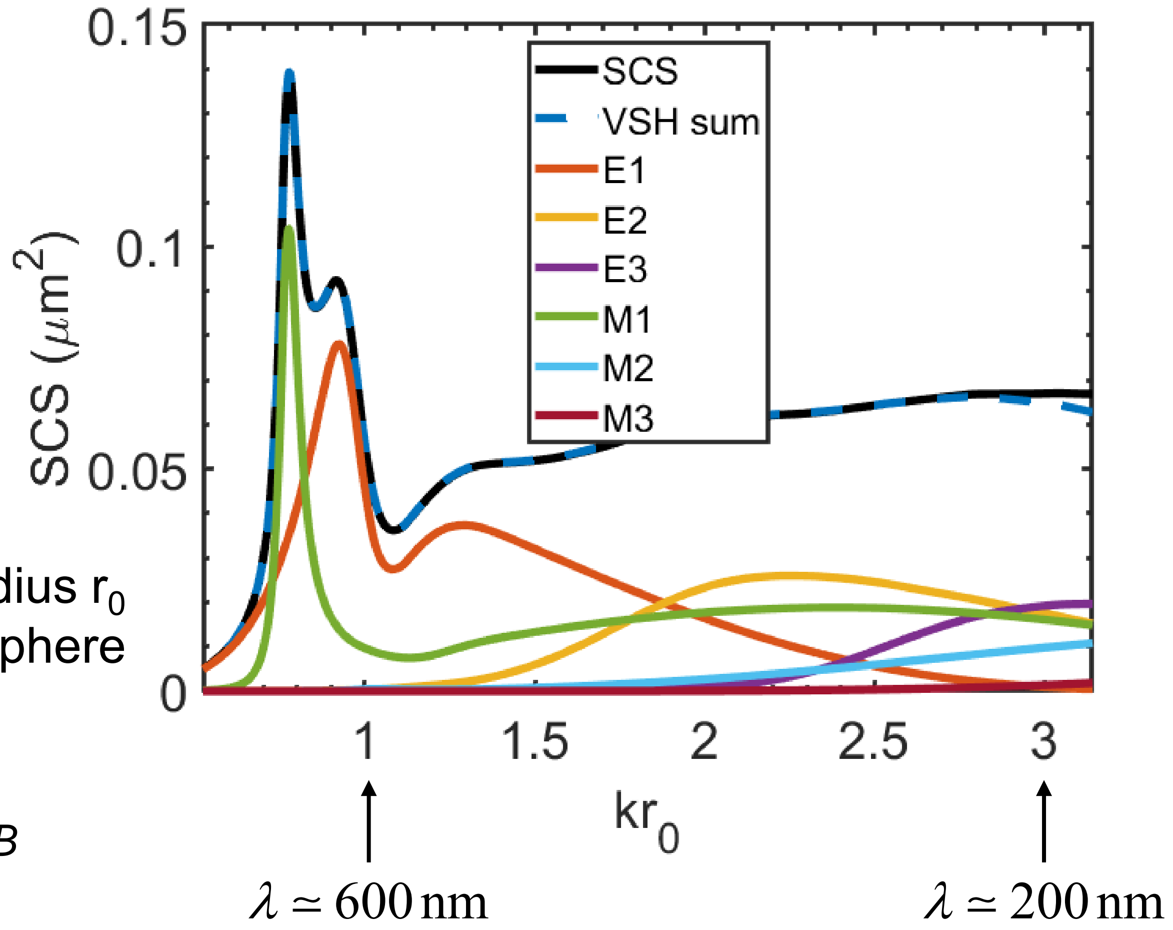
$$C_{sca} = k^2 \sum_{n=1}^{\infty} \sum_{m=-n}^{n} n(n+1)(|a_{nm}|^2 + |b_{nm}|^2).$$

S. Mühlig, Metamaterials vol. 5, p. 64 (2011)

Scattering by a sphere – Mie scattering and VSH

- The first and second VSHs are usually sufficient to represent the scattering cross section
- Important check: the sum of VSH must equal the scattering cross section

100 nm radius r_0
amorphous Si sphere



M. Riccardi, Physical Review B vol. 106, p. 115428 (2022)

Scattering by a sphere – Cartesian multipoles

- One is used to represent the scattering in Cartesian coordinates (not in spherical coordinates)
- VSH → Cartesian multipoles
- Only orders ±1 of VSH are required for Cartesian dipoles:

$$\mathbf{p} = \begin{pmatrix} p_x \\ p_y \\ p_z \end{pmatrix} = C_0 \begin{pmatrix} (a_{11} - a_{1-1}) \\ i(a_{11} + a_{1-1}) \\ -\sqrt{2}a_{10} \end{pmatrix} \qquad \mathbf{m} = \begin{pmatrix} m_x \\ m_y \\ m_z \end{pmatrix} = cC_0 \begin{pmatrix} (b_{11} - b_{1-1}) \\ i(b_{11} + b_{1-1}) \\ -\sqrt{2}b_{10} \end{pmatrix}$$

$$C_0 = \sqrt{6\pi} i / c Z_0 k$$

S. Mühlig, Metamaterials vol. 5, p. 64 (2011)

Scattering by a sphere – Cartesian multipoles

- Cartesian quadrupoles:

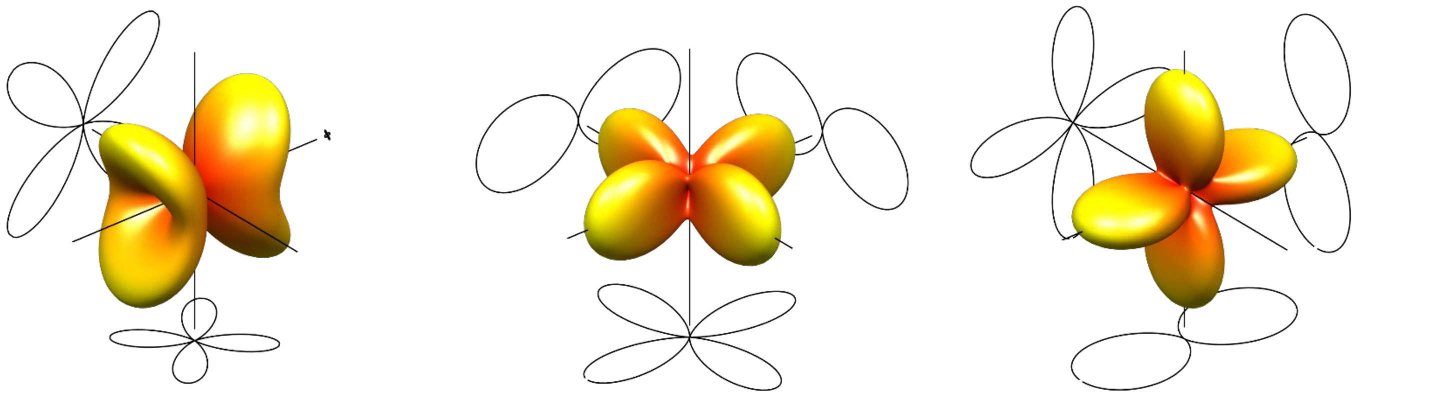
$$\mathbf{Q} = \begin{pmatrix} Q_{xx} & Q_{xy} & Q_{xz} \\ Q_{yx} & Q_{yy} & Q_{yz} \\ Q_{zx} & Q_{zy} & Q_{zz} \end{pmatrix}$$

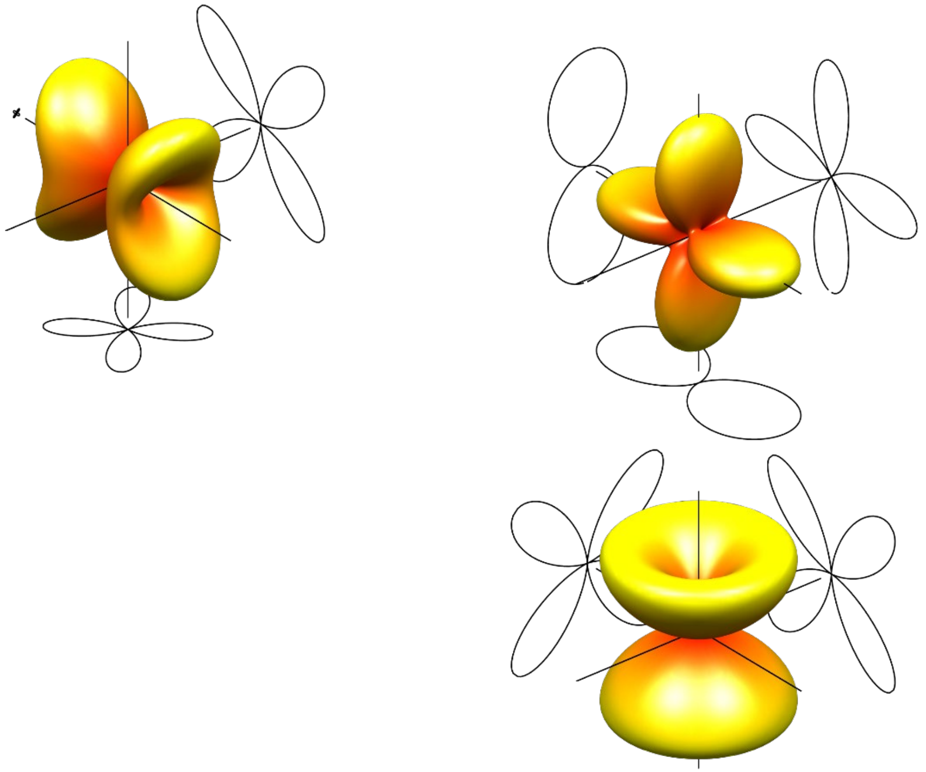
$$\mathbf{Q} = D_0 \begin{pmatrix} i(a_{2,2}+a_{2,-2}) - \frac{i\sqrt{6}}{2}a_{2,0} & (a_{2,-2}-a_{2,2}) & i(a_{2,-1}-a_{2,1}) \\ (a_{2,-2}-a_{2,2}) & -i(a_{2,2}+a_{2,-2}) - \frac{i\sqrt{6}}{2}a_{2,0} & (a_{2,-1}+a_{2,1}) \\ i(a_{2,-1}-a_{2,1}) & (a_{2,-1}+a_{2,1}) & i\sqrt{6}a_{2,0} \end{pmatrix}$$

S. Mühlig, Metamaterials vol. 5, p. 64 (2011)

Scattering by a sphere – Cartesian multipoles

- Cartesian quadrupoles:



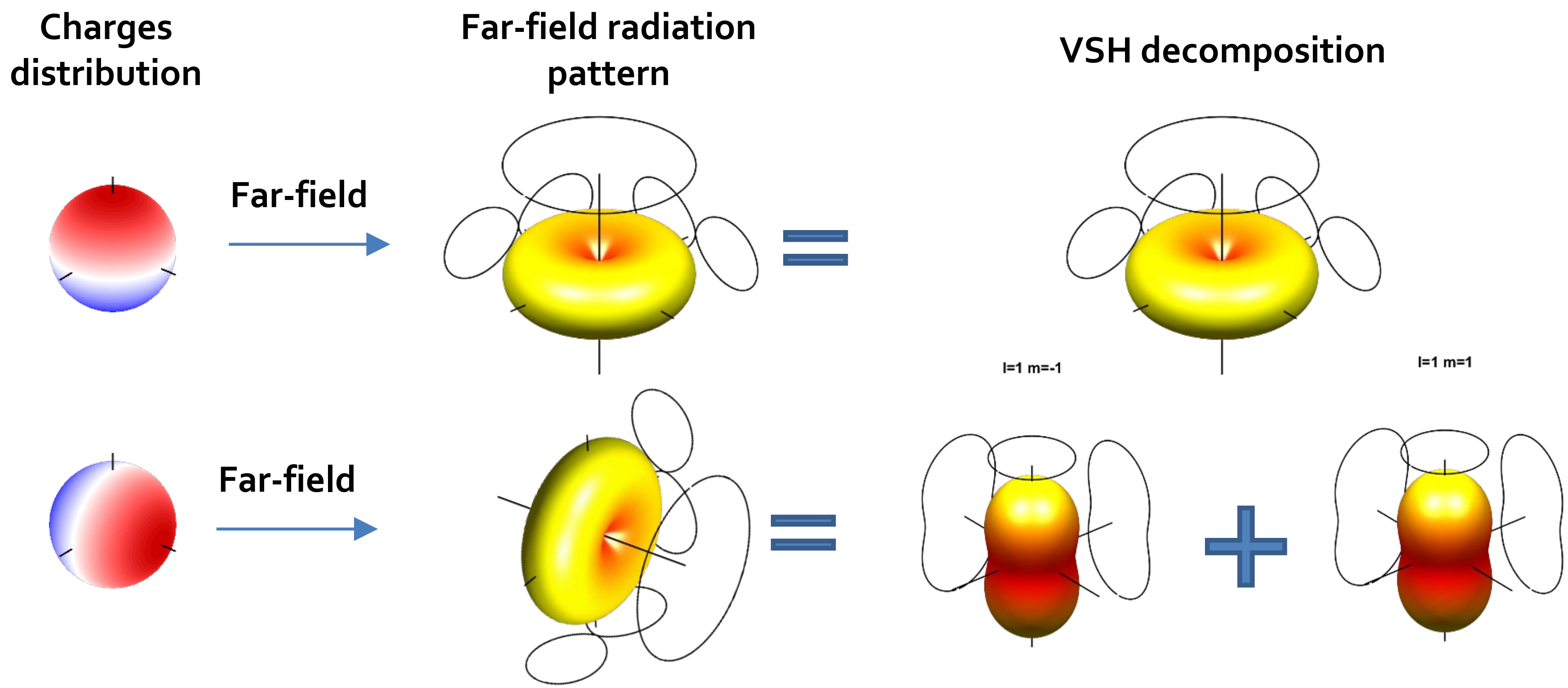


$$\mathbf{Q} = \begin{pmatrix} Q_{xx} & Q_{xy} & Q_{xz} \\ Q_{yx} & Q_{yy} & Q_{yz} \\ Q_{zx} & Q_{zy} & Q_{zz} \end{pmatrix}$$

S. Mühlig, Metamaterials vol. 5, p. 64 (2011)

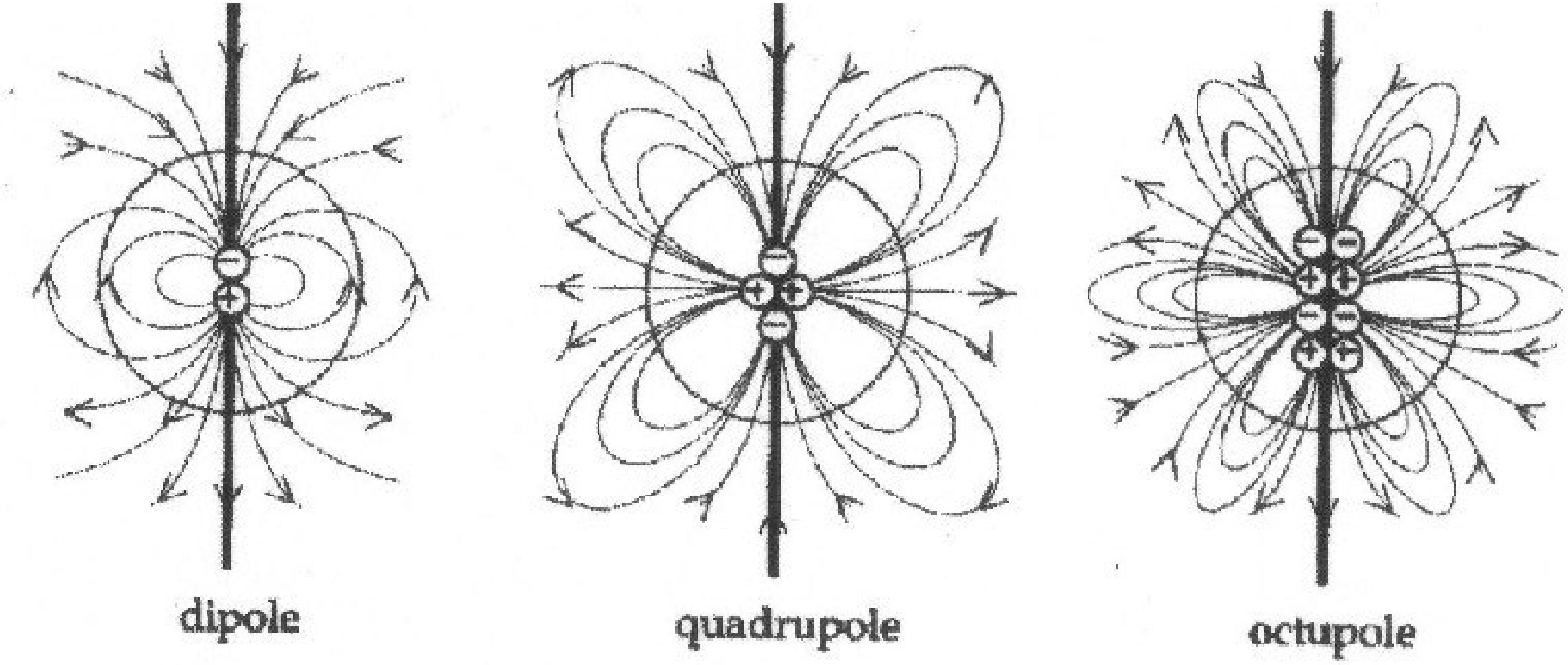
Link between VSH and Cartesian multipoles

- Not all axes are equal!

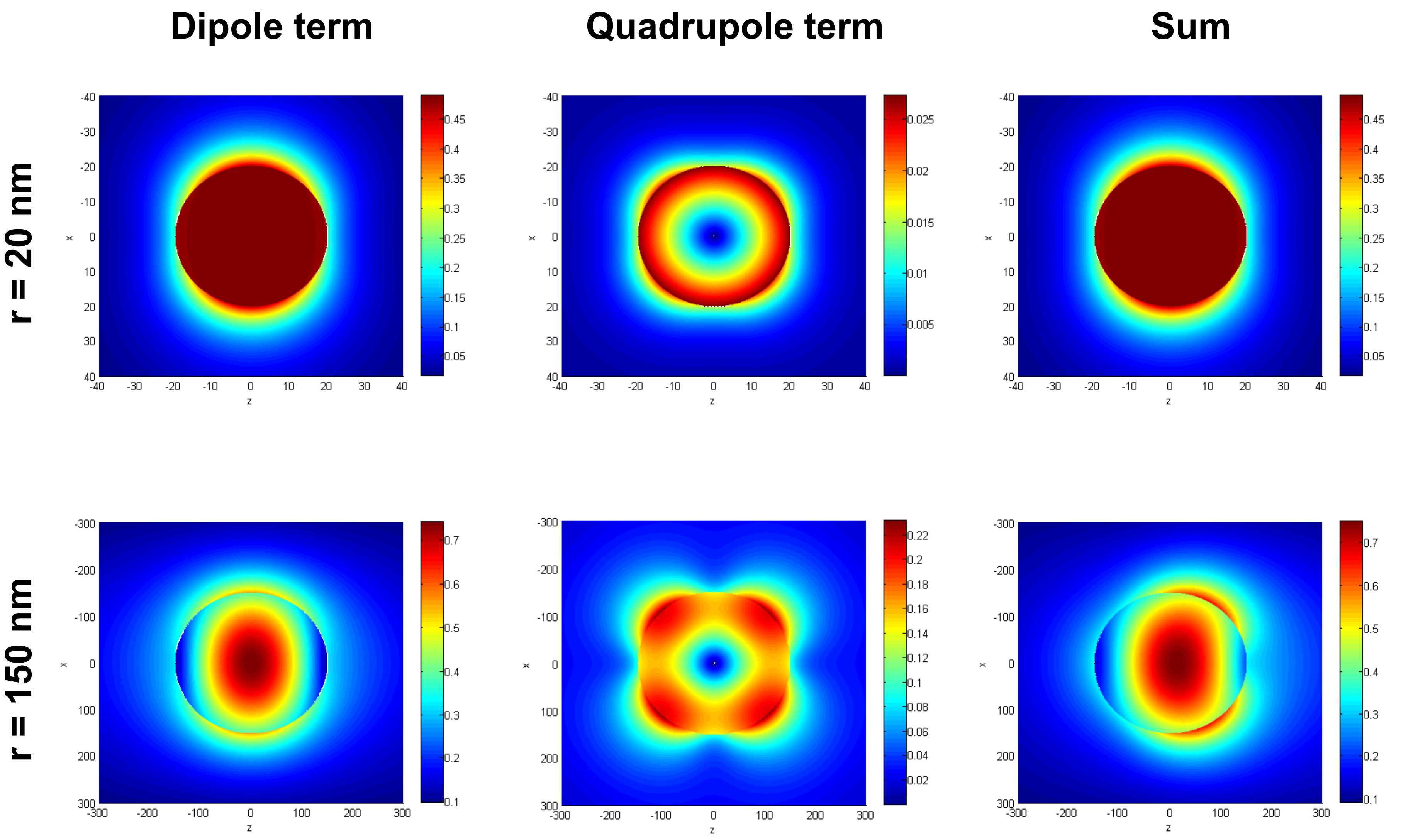


Some Cartesian multipoles

- The radiation pattern can be understood by visualizing the corresponding moving charges



The larger the scatterer, the higher the required multipoles



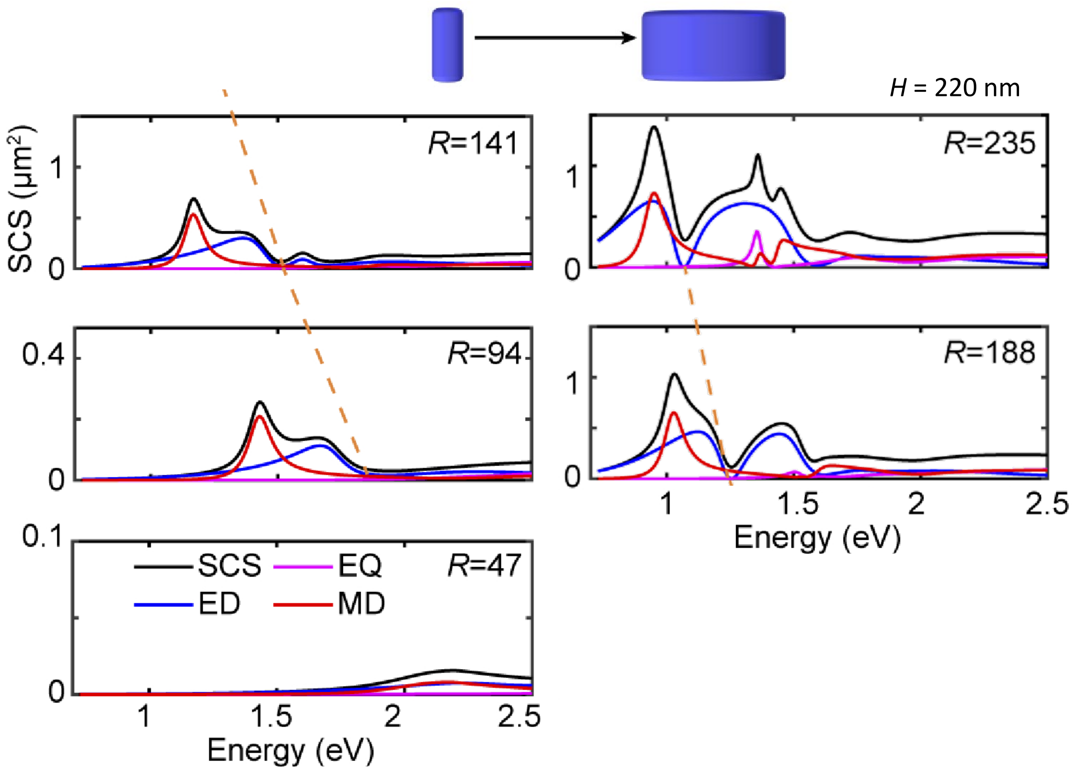
Dielectric sphere n=1.55, λ=600 nm

Olivier J.F. Martin

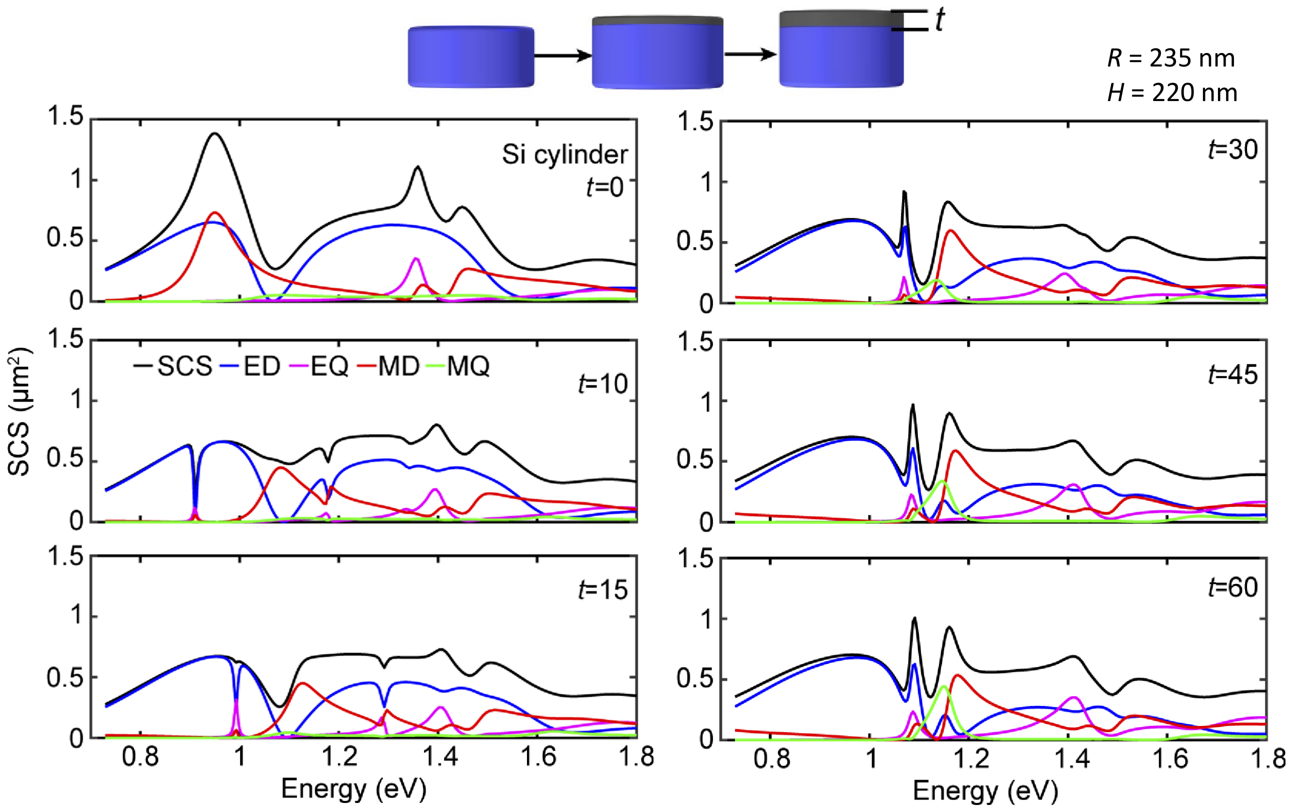
Some practical examples of the utilization of multipoles

- Dielectric structures support a magnetic and an electric dipoles that can couple to the electric dipole of a metallic structure to produce a very rich response

Si cylinder



Si cylinder + Ag disk



D. Ray, Optics Express vol. 29, p. 24056 (2021)

Selected Topics in Advanced Optics

Week 4 – part 4

Olivier J.F. Martin
Nanophotonics and Metrology Laboratory

EPFL

Scattering by particles small compared to the wavelength

- We define two parameters:
  - Size parameter $x = ka = \frac{2\pi n a}{\lambda_0}$ (n: index of surrounding)
  - Relative refractive index $m = \frac{k_1}{k} = \frac{n_1}{n}$ (n_1: index of the sphere)
- We look at spheres with a small size parameter:

$$x = ka = \frac{2\pi n a}{\lambda_0} \ll 1 \qquad and \qquad |m| x \ll 1$$

- Then we can approximate the Hankel functions with their power series (see e.g. M. Abramowitz and A. Stegun, Pocketbook of mathematical functions (Harri Deutsch, Thun, 1984))

Scattering by particles small compared to the wavelength

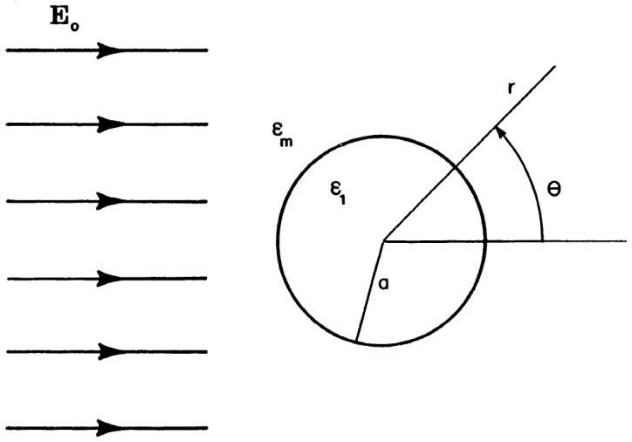
- Finally, one obtains the scattered intensity I_{scat} for a sphere illuminated with incident intensity I_i :

$$I_{scat} = \frac{8\pi^4 n a^6}{\lambda^4 r^2} \left| \frac{m^2 - 1}{m^2 + 2} \right|^2 \left(1 + \cos^2\theta\right) I_i$$

Rayleigh scattering

Scattering by a small particle in the electrostatic limit

- If the particle is small, it makes sense to assume that the incident field is homogeneous
- One can then use an electrostatic approach and the formulae simplify dramatically:



$$Q_{abs} = 4x \operatorname{Im}\left\{\frac{\varepsilon_1 - \varepsilon_m}{\varepsilon_1 + 2\varepsilon_m}\right\}$$

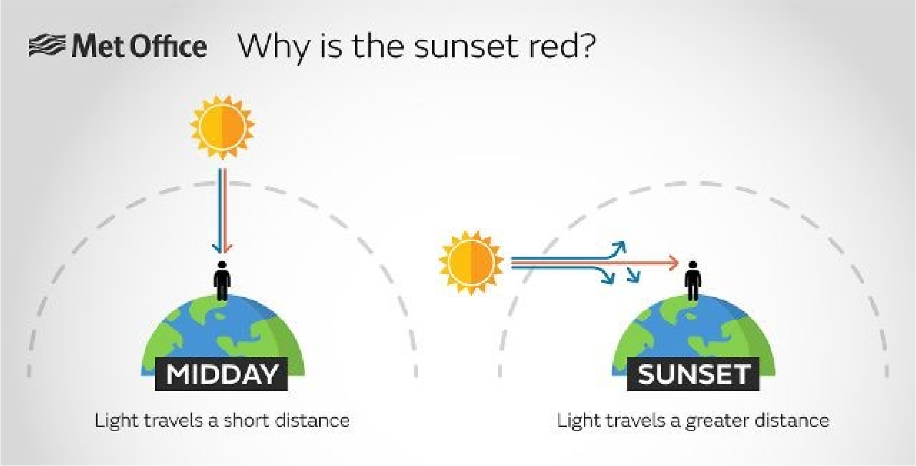
$$Q_{scat} = \frac{8}{3} x^4 \left|\frac{\varepsilon_1 - \varepsilon_m}{\varepsilon_1 + 2\varepsilon_m}\right|^2$$

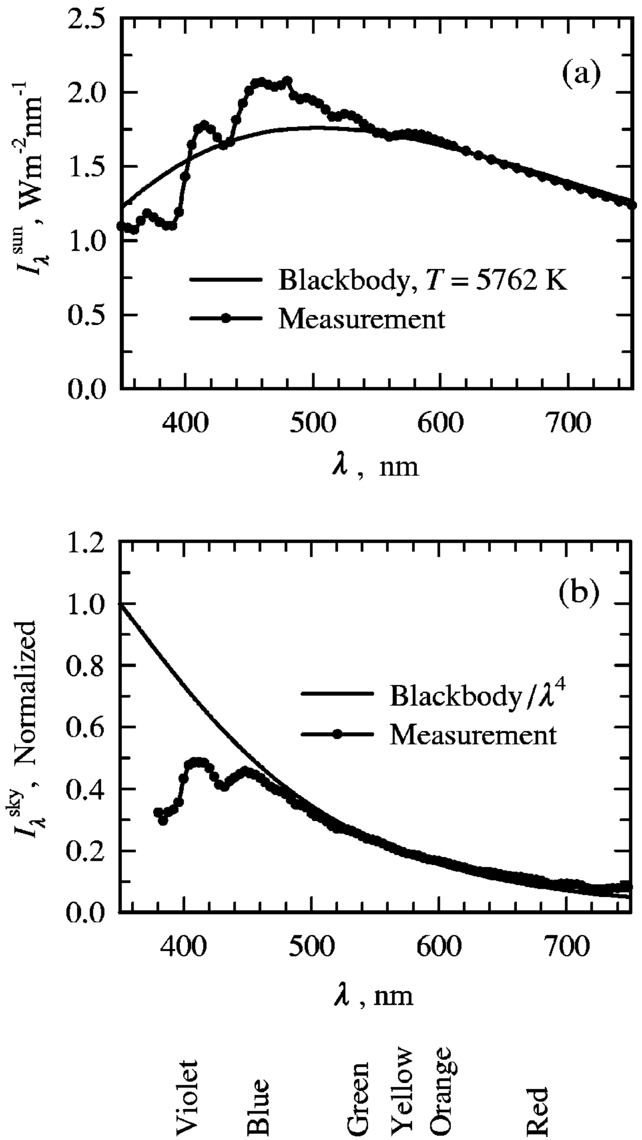
$$Q_{ext} = Q_{abs} + Q_{scat} \qquad x = ka = \frac{2\pi n a}{\lambda_0}$$

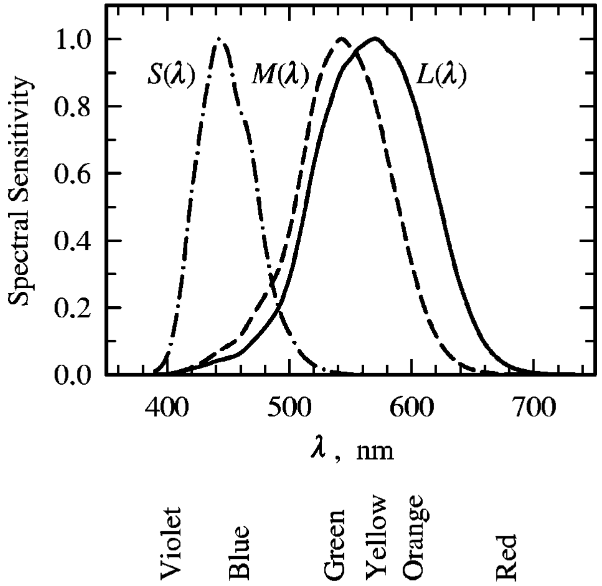
- When extinction is dominated by absorption, its spectrum goes with $1/\lambda$
- When it is dominated by scattering, its spectrum goes with $1/\lambda^4$

The colour of the sky

- Since light scattering goes with $1/\lambda^4$, the sky should have the colour of the shortest wavelength our eye can perceive: violet !
- However, the sky is blue, because of absorption of violet through the atmosphere and physiology of our eye



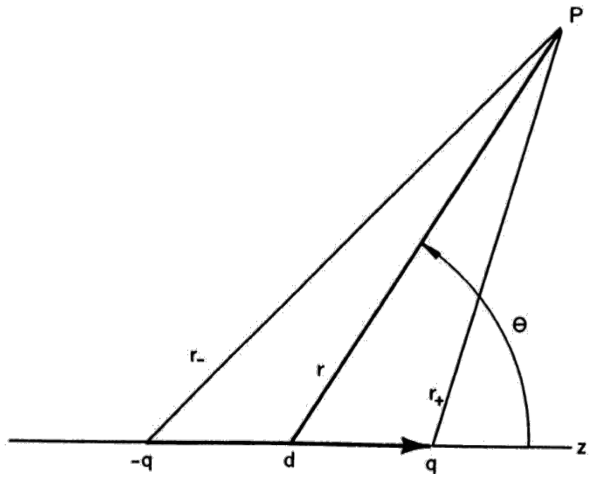




G.S. Smith, American Journal of Physics vol. 73, p. 590 (2005)

Scattering by a small particle in the electrostatic limit

- The previous approach is very useful for light scattering by a dipole (two oscillating charges +q and –q) with dipole moment $\mathbf{p} = p\,\mathbf{e}_z = qd\,\mathbf{e}_z$



- Potential at a point P (assuming $d \to 0$ but qd remains constant):

$$\Phi = \frac{\mathbf{p}\cdot\mathbf{r}}{4\pi\varepsilon_m r^3} = \frac{p\cos\theta}{4\pi\varepsilon_m r^2}$$

- By comparison, we can show that the field scattered by a small sphere is similar to that of a dipole with dipole moment:

$$\boxed{\mathbf{p} = 4\pi\varepsilon_m a^3 \frac{\varepsilon_1 - \varepsilon_m}{\varepsilon_1 + 2\varepsilon_m}\mathbf{E}_0 = \varepsilon_m \alpha \mathbf{E}_0}$$

- Where we introduce the sphere polarizability: $\alpha = 4\pi a^3 \dfrac{\varepsilon_1 - \varepsilon_m}{\varepsilon_1 + 2\varepsilon_m}$

Scattering by a small particle in the electrostatic limit

- The polarizability completely characterizes the sphere and can be used to define the cross sections:

$$C_{ext} = k \operatorname{Im}\{\alpha\}$$

$$C_{scat} = \frac{k^4}{6\pi}|\alpha|^2$$

- These equations are valid if scattering is small compared to absorption; as a consequence using the optical theorem:

$$C_{abs} = C_{ext} - C_{scat} \simeq C_{ext} = k \operatorname{Im}\{\alpha\}$$

Light scattering by a slab

- When measuring a (very large) homogeneous slab, one considers the reflected (R), absorbed (A) and transmitted (T) light intensities
- Optical theorem in that case: $A + R + T = 1$
- In general one cannot measure absorption, but it can be deduced from R and T:

$$\boxed{A = 1 - R - T}$$