Selected Topics in Advanced Optics

Week 4 – part 1

Olivier J.F. Martin
Nanophotonics and Metrology Laboratory

EPFL

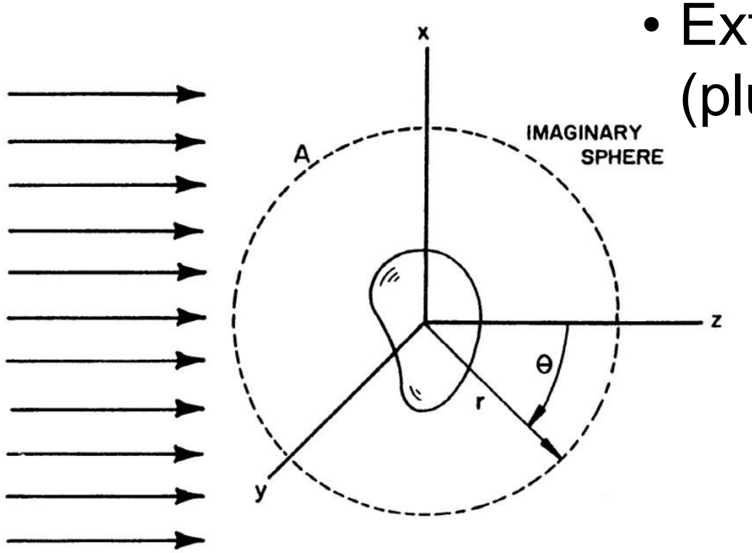
Module 3: Light scattering

- 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).

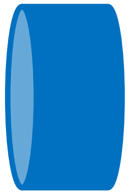
Light scattering determines our perception of the world

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)



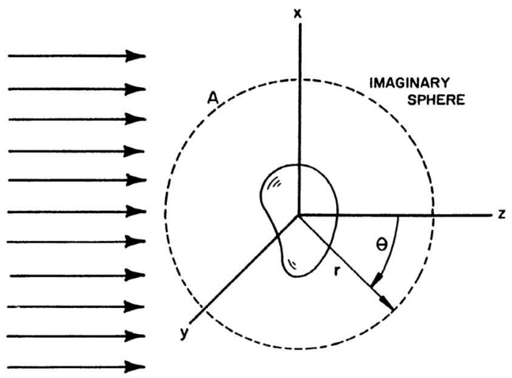
- Power flow across a closed surface around the scatterer:

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

- $W_{abs} > 0$ 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):



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

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

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

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):

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

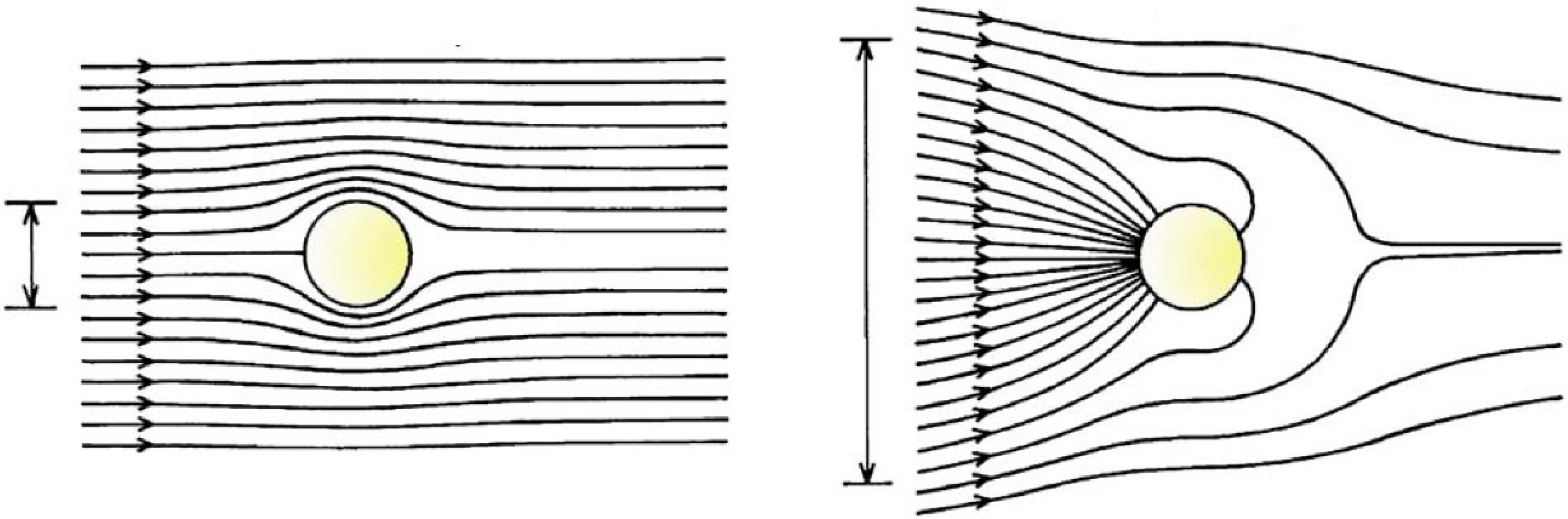
- These cross sections have the dimension of an area !
- By dividing the cross section by the geometrical cross-sectional area G ($G = \pi a^2$ for a sphere) projected onto a plane perpendicular to the incident beam, one obtains the efficiencies:

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

- 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:

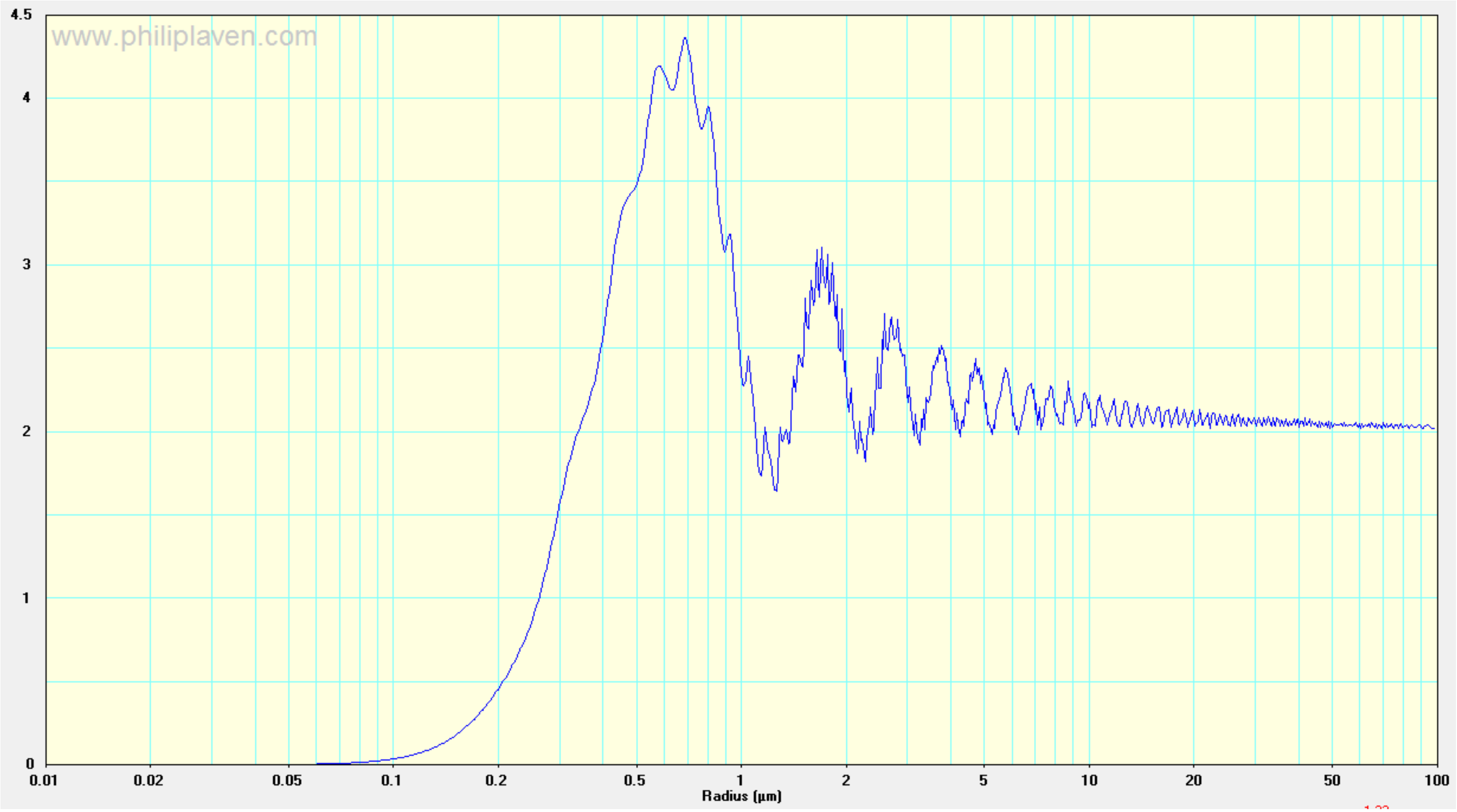


Cross sections

- 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)

Cross sections

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

Sphere, radius=1 μm, n=1.5

Q_{scat}

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

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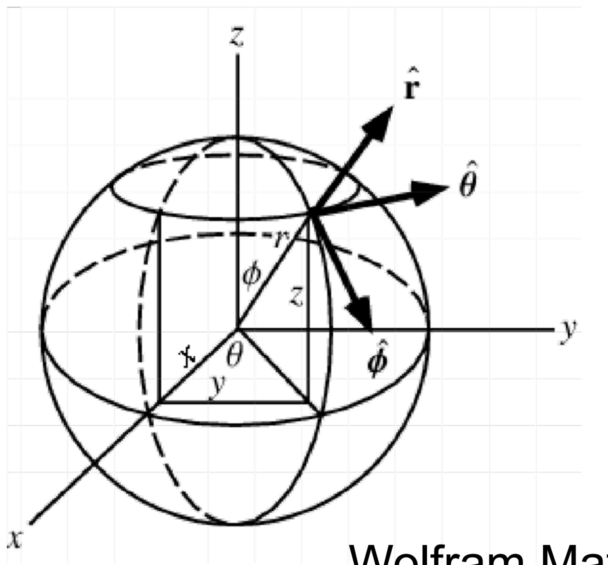
Scattering by a sphere – Mie scattering and VSH

- 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

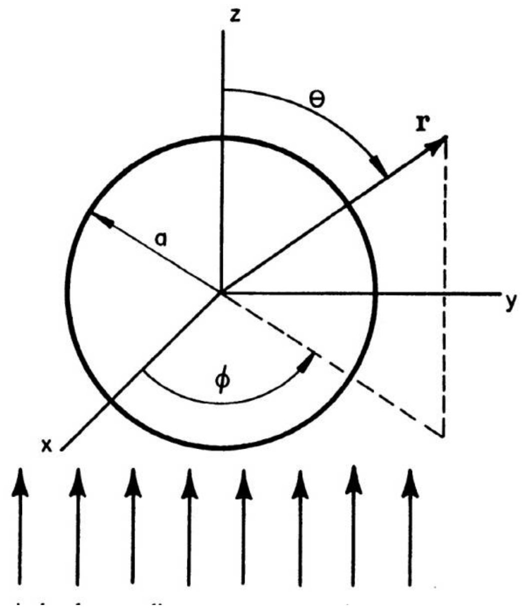
Scattering by a sphere – Mie scattering and VSH

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

$$\boxed{\begin{array}{ll} \nabla^2\mathbf{E}(\mathbf{r})+k^2\mathbf{E}(\mathbf{r})=0 & \nabla\cdot\mathbf{E}(\mathbf{r})=0 \\ \nabla^2\mathbf{H}(\mathbf{r})+k^2\mathbf{H}(\mathbf{r})=0 & \nabla\cdot\mathbf{H}(\mathbf{r})=0 \\ \nabla\times\mathbf{E}(\mathbf{r})=j\omega\varepsilon\mathbf{H}(\mathbf{r}) & \nabla\times\mathbf{H}(\mathbf{r})=-j\omega\varepsilon\mathbf{E}(\mathbf{r}) \end{array}}$$

- 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 $\boldsymbol{r}$, one defines vectorial harmonic functions:

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

- 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

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

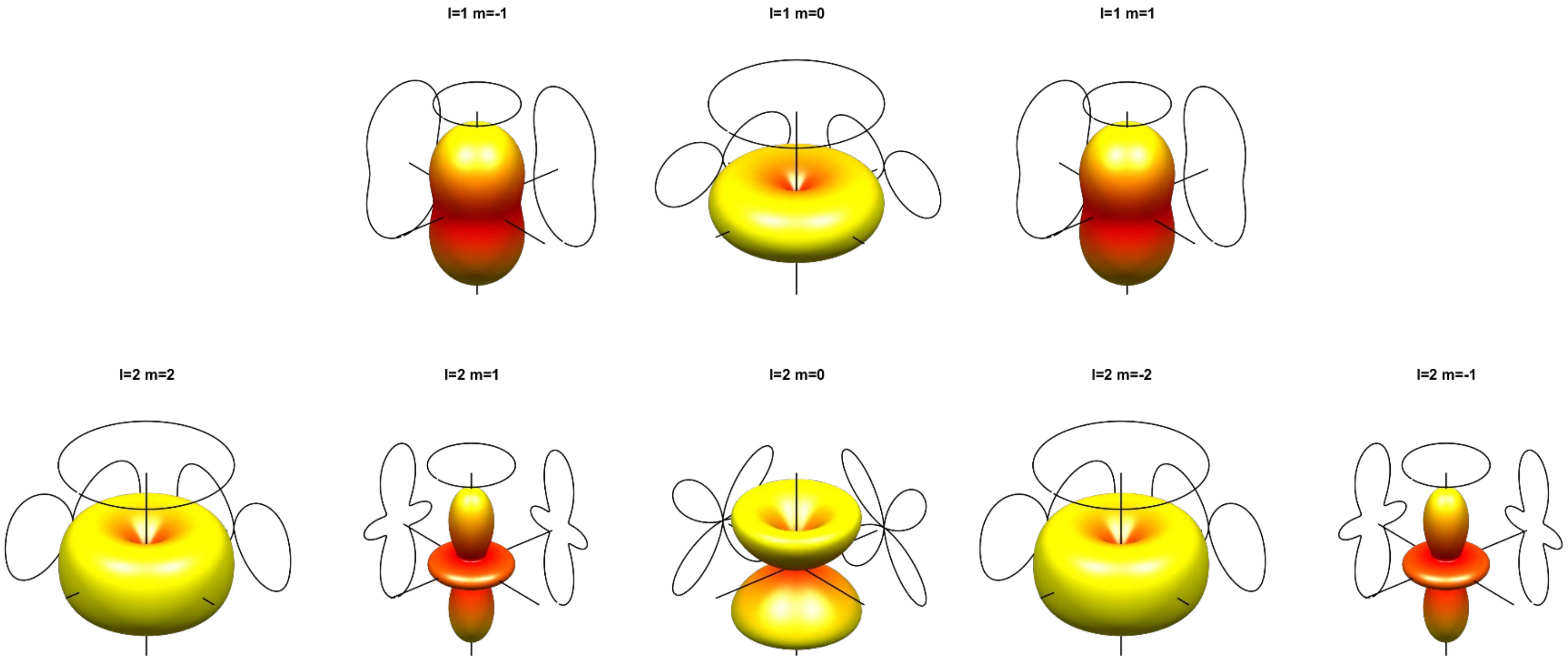
$$\mathbf{N}_{nm} = n(n+1)P_n^m(\cos\theta)\frac{h_n^{(1)}(kr)}{kr}\exp(im\varphi)\mathbf{i}_r + \left[\tau_{nm}(\cos\theta)\mathbf{i}_\theta + i\pi_{nm}(\cos\theta)\mathbf{i}_\varphi\right] \times \frac{1}{kr}\frac{d}{dr}\left[rh_n^{(1)}(kr)\right]\exp(im\varphi).$$

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

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



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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 – 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 <u>for Cartesian dipoles</u>:

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

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

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

Scattering by a sphere – Cartesian multipoles

- Cartesian quadrupoles:

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

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

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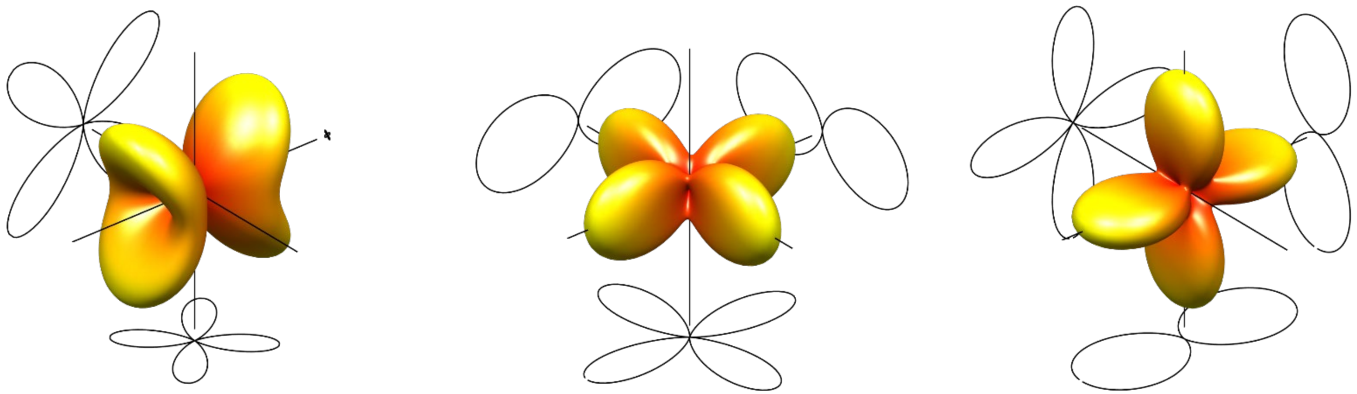
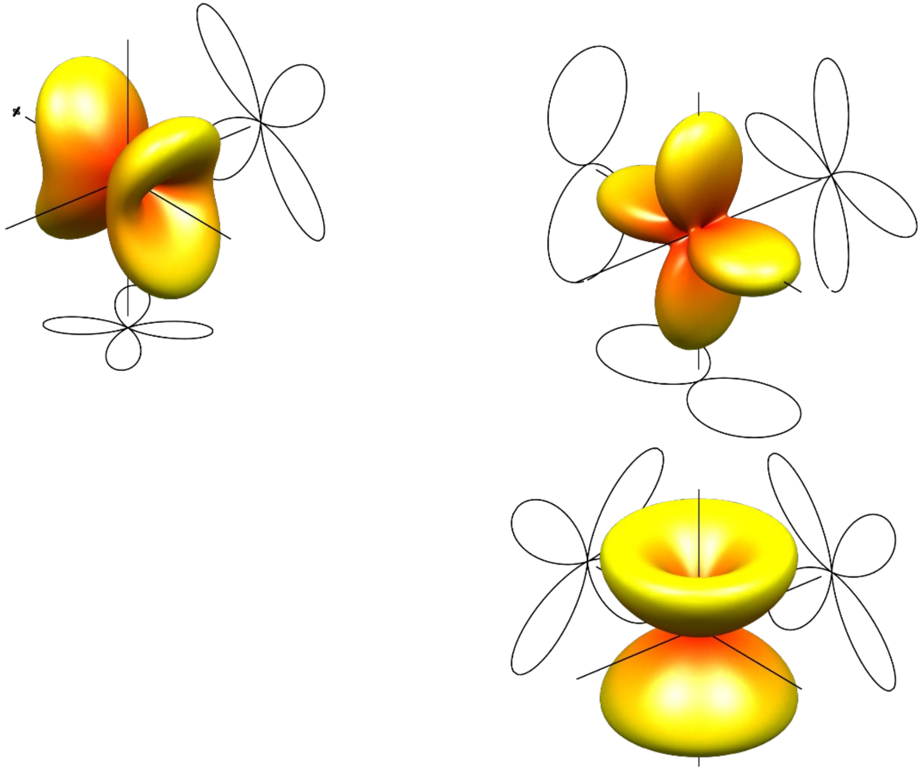
Scattering by a sphere – Cartesian multipoles

- <u>Cartesian quadrupoles</u>:



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



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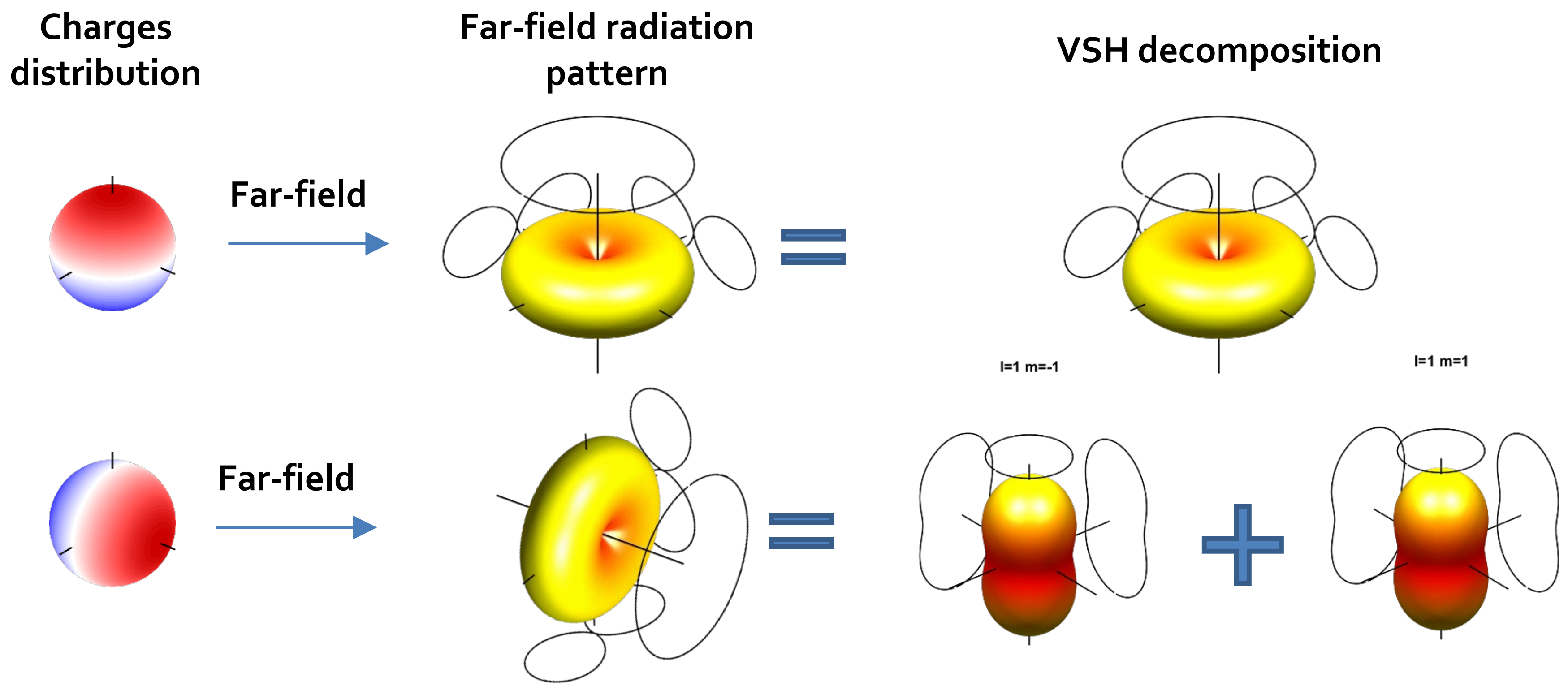
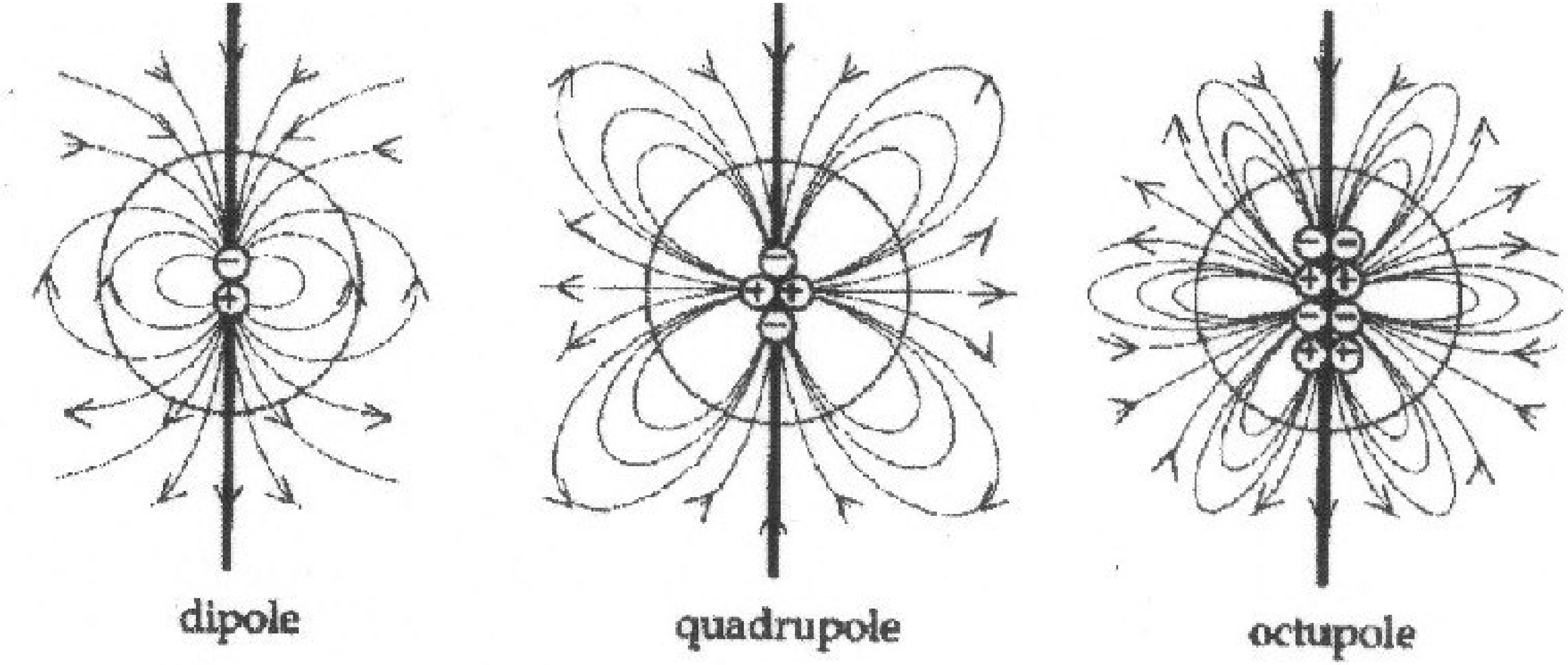
Link between VSH and Cartesian multipoles

- Not all axes are equal!

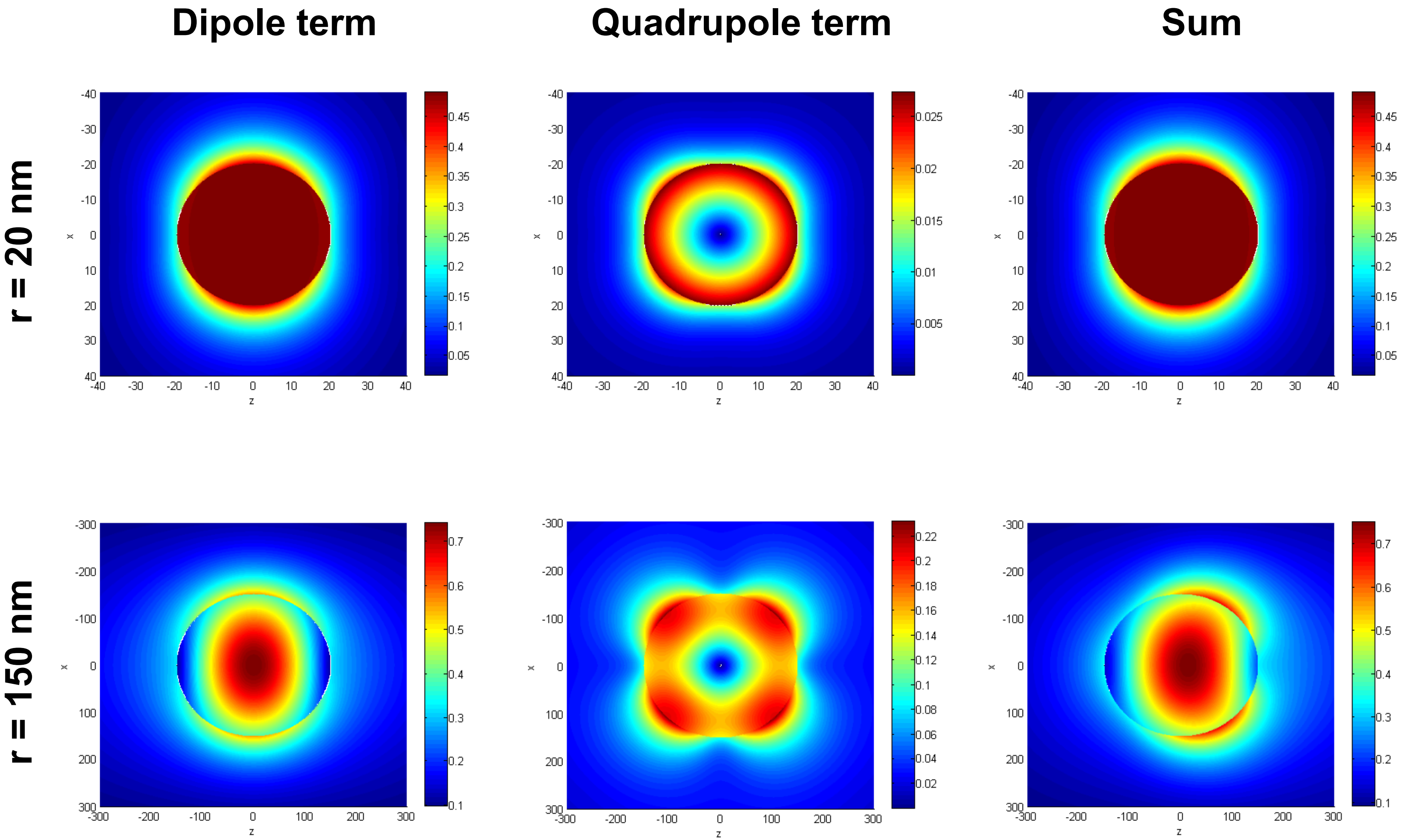


Some Cartesian multipoles

- 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

Selected Topics in Advanced Optics

Week 4 – part 4

Olivier J.F. Martin
Nanophotonics and Metrology Laboratory

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Scattering by particles small compared to the wavelength

- We define two parameters:
  - Size parameter $x = ka = \dfrac{2\pi na}{\lambda_0}$ (n: index of surrounding)
  - Relative refractive index $m = \dfrac{k_1}{k} = \dfrac{n_1}{n}$ (n_1: index of the sphere)

- We look at spheres with a small size parameter:

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

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

Scattering by particles small compared to the wavelength

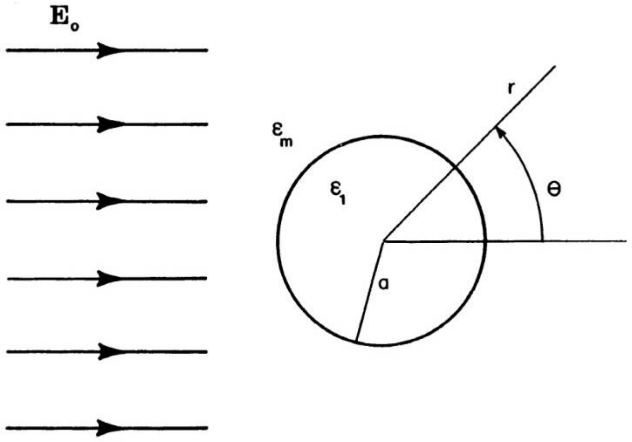
- Finally, one obtains the scattered intensity I_{scat} for a sphere illuminated with incident intensity I_i :

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

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:



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

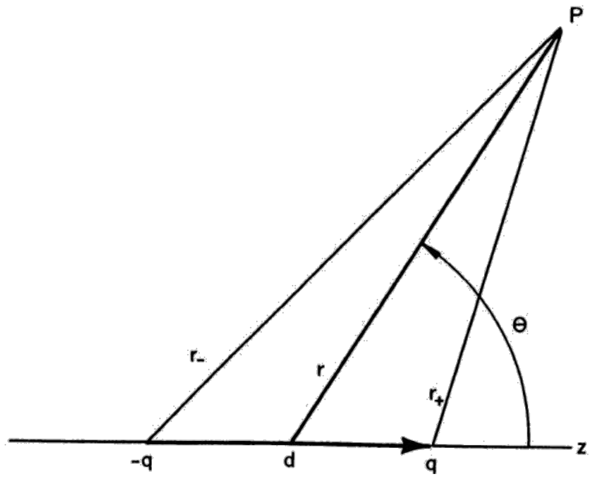
$$Q_{scat} = \frac{8}{3}x^4\left|\frac{\varepsilon_1 - \varepsilon_m}{\varepsilon_1 + 2\varepsilon_m}\right|^2$$

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

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

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:

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