Optical scattering resonances of metal nano particles

(PhD Course: Optical at the Nanoscale)

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1. Introduction

In this note we will cover theory for scattering from very small particles, and spherical particles. More complex geometries will be covered with slides.

2. Scattering from very small particles – the quasistatic limit:

In this section a quasi-static approach will be presented for calculating optical scattering by, and absorption in, very small particles. Using a quasi-static approach rather than a fully retardation-based approach will be justified. Optical absorption and scattering cross sections will be expressed in terms of the dipole moment of the particle, which can be calculated from knowledge of the electric field inside the particle. The electric field can be calculated with the methods of electrostatics.

As a first step we will consider the radiation from a time-dependent current distribution. We may expand the electromagnetic fields **E**, **D**, **B**, **H**, the currents **J**, and the relative dielectric constant ε_r in its temporal Fourier-components, i.e.

$$\boldsymbol{E}(\boldsymbol{r},t) = \int_{\omega=-\infty}^{\omega=\infty} \boldsymbol{E}(\boldsymbol{r},\omega)e^{i\omega t}\,d\omega \;= 2Real\left(\int_{\omega=0}^{\omega=\infty} \boldsymbol{E}(\boldsymbol{r},\omega)e^{i\omega t}\,d\omega\right) \quad , \tag{1}$$

where

$$\boldsymbol{E}(\boldsymbol{r},\omega) = \frac{1}{2\pi}\int_{t=-\infty}^{t=\infty} \boldsymbol{E}(\boldsymbol{r},t)e^{-i\omega t}\,dt \;, \tag{2}$$

and similar for the other parameters (**D**, **B**, **H**, **J**, and ε_r).

Assuming linear materials Maxwell's equations can then be formulated ($\mu = \mu_0$, $\boldsymbol{D}(\boldsymbol{r},\omega) = \varepsilon_r(\boldsymbol{r},\omega)\boldsymbol{E}(\boldsymbol{r},\omega)$, $\rho = 0$):

$$\boldsymbol{\nabla}\times\boldsymbol{E}(\boldsymbol{r},\omega) = -i\omega\mu_0\boldsymbol{H}(\boldsymbol{r},\omega), \tag{3}$$

$$\boldsymbol{\nabla}\times\boldsymbol{H}(\boldsymbol{r},\omega) = i\omega\varepsilon_0\varepsilon_r(\boldsymbol{r},\omega)\boldsymbol{E}(\boldsymbol{r},\omega) + \boldsymbol{J}(\boldsymbol{r},\omega), \tag{4}$$

$$\boldsymbol{\nabla}\cdot\varepsilon_r(\boldsymbol{r},\omega)\boldsymbol{E}(\boldsymbol{r},\omega) = 0, \tag{5}$$

$$\boldsymbol{\nabla}\cdot\mathbf{H}(\boldsymbol{r},\omega) = 0, \tag{6}$$

From which we can derive the wave equation ($k_0^2 = \omega^2/c^2$ with c being the vacuum speed of light)

$$-\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E}(\boldsymbol{r},\omega) + k_0^2\varepsilon_r(\boldsymbol{r},\omega)\boldsymbol{E}(\boldsymbol{r},\omega) = i\omega\mu_0\boldsymbol{J}(\boldsymbol{r},\omega). \tag{7}$$

If the currents satisfy Ohm's law, $\boldsymbol{J}(\boldsymbol{r},\omega)=\sigma(\boldsymbol{r},\omega)\boldsymbol{E}(\boldsymbol{r},\omega)$ where σ is the conductivity, we arrive at a wave equation in terms of only the electric field and a complex dielectrictric constant

$$-\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E}(\boldsymbol{r},\omega)+k_0^2\varepsilon(\boldsymbol{r},\omega)\boldsymbol{E}(\boldsymbol{r},\omega)=0, \tag{8}$$

where $\varepsilon(\boldsymbol{r},\omega)=\varepsilon_r(\boldsymbol{r},\omega)-i\sigma(\boldsymbol{r},\omega)/\omega\varepsilon_0$. We may also define the complex refractive index as $n(\boldsymbol{r},\omega)=\sqrt{\varepsilon(\boldsymbol{r},\omega)}$. For metals it is usually the complex refractive index which is tabulated in books and papers. The complex refractive index for gold, silver and copper can e.g. be found in [1].

In the case where the electric field is generated by a current distribution in a homogeneous medium, i.e.

$$-\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E}(\boldsymbol{r},\omega)+k_0^2\varepsilon_{ref}(\omega)\boldsymbol{E}(\boldsymbol{r},\omega)=i\omega\mu_0\boldsymbol{J}(\boldsymbol{r},\omega), \tag{9}$$

a solution satisfying that the radiation should propagate away from the sources is given by

$$\boldsymbol{E}(\boldsymbol{r},\omega)=-i\omega\mu_0\int\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}';\omega)\cdot\boldsymbol{J}(\boldsymbol{r},\omega)d^3r', \tag{10}$$

where the Green's tensor $\overleftrightarrow{\boldsymbol{G}}$ is given by

$$\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}';\omega)=\left(\overleftrightarrow{\boldsymbol{I}}+\frac{1}{k_0^2\varepsilon_{ref}}\nabla\nabla\right)g(\boldsymbol{r},\boldsymbol{r}';\omega),\quad g(\boldsymbol{r},\boldsymbol{r}';\omega)=\frac{e^{-ik_0n_{ref}|\boldsymbol{r}-\boldsymbol{r}'|}}{4\pi|\boldsymbol{r}-\boldsymbol{r}'|}. \tag{11}$$

$$\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}';\omega)=\left(\overleftrightarrow{\boldsymbol{I}}\left[1-\frac{i}{k_0n_{ref}|\boldsymbol{r}-\boldsymbol{r}'|}-\frac{1}{k_0^2\varepsilon_{ref}|\boldsymbol{r}-\boldsymbol{r}'|^2}\right]-\frac{(\boldsymbol{r}-\boldsymbol{r}')(\boldsymbol{r}-\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|^2}\left[1-\frac{3i}{k_0n_{ref}|\boldsymbol{r}-\boldsymbol{r}'|}-\frac{3}{k_0^2\varepsilon_{ref}|\boldsymbol{r}-\boldsymbol{r}'|^2}\right]\right)\frac{e^{-ik_0n_{ref}|\boldsymbol{r}-\boldsymbol{r}'|}}{4\pi|\boldsymbol{r}-\boldsymbol{r}'|}.$$

The expression (10) is a solution to equation (9) because the Green's tensor satisfies

$$-\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}';\omega)+k_0^2\varepsilon_{ref}(\omega)\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}';\omega)=-\overleftrightarrow{\boldsymbol{I}}\delta(\boldsymbol{r}-\boldsymbol{r}'). \tag{12}$$

We can now use Eq. (10) to calculate the radiation field generated by a monochromatic dipole current, a current which exists because the position of the two charges oscillates. The dipole moment can be expressed in the form

$$\boldsymbol{p}(\boldsymbol{r},t)=\left(\boldsymbol{p}_0e^{i\omega t}+C.C.\right)\delta(\boldsymbol{r}-\boldsymbol{r}_0), \tag{13}$$

and the corresponding dipole current is given by ($\boldsymbol{J}(\boldsymbol{r},t)=\frac{\partial}{\partial t}\boldsymbol{p}(\boldsymbol{r},t)$):

$$\boldsymbol{J}(\boldsymbol{r},\omega)=i\omega\boldsymbol{p}_0\delta(\boldsymbol{r}-\boldsymbol{r}_0). \tag{14}$$

Thereby the electric field generated by a dipole current is given by

$$\boldsymbol{E}(\boldsymbol{r},\omega)=\omega^2\mu_0\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}_0;\omega)\cdot\boldsymbol{p}_0\,. \tag{15}$$

Without loss of generality we may choose $\boldsymbol{p}_0=\hat{z}p_0$ and $\boldsymbol{r}_0=\boldsymbol{0}$. Then for large distances to the current source the expression (15) reduces to

$$\boldsymbol{E}(\boldsymbol{r},\omega)=-\omega^2\mu_0p_0\hat{\theta}\sin\theta\,\frac{e^{-ik_0n_{ref}r}}{4\pi r}. \tag{16}$$

The magnetic field can then be calculated using the Maxwell equation (3) resulting in

$$\boldsymbol{H}(\boldsymbol{r},\omega) = -\frac{\omega^2}{c} p_0 n_{ref} \hat{\phi} \sin\theta \frac{e^{-ik_0 n_{ref} r}}{4\pi r} . \quad (17)$$

The radiated power can be calculated from the Poying vector, which has to be based on real fields instead of the complex fields, i.e.

$$\boldsymbol{S}(\boldsymbol{r}) = \langle \boldsymbol{E}(\boldsymbol{r},t) \times \boldsymbol{H}(\boldsymbol{r},t) \rangle = \langle \left(\boldsymbol{E}(\boldsymbol{r},\omega)e^{i\omega t} + \ C.C.\right) \times \left(\boldsymbol{H}(\boldsymbol{r},\omega)e^{i\omega t} + \ C.C.\right) \rangle \quad (18)$$

$$= \boldsymbol{E}(\boldsymbol{r},\omega) \times [\boldsymbol{H}(\boldsymbol{r},\omega)]^* + C.C. = 2Real(\boldsymbol{E}(\boldsymbol{r},\omega) \times [\boldsymbol{H}(\boldsymbol{r},\omega)]^*)$$

$$= 2k_0^4 \frac{c}{\varepsilon_0} \frac{n_{ref}}{4\pi} |p_0|^2 \frac{\sin^2\theta}{4\pi r^2} \hat{r} .$$

The time-averaged power radiated from the dipole can thereby be expressed in terms of the dipole moment

$$P = \oiint \boldsymbol{S}(\boldsymbol{r}) \cdot \hat{n} da = 2k_0^4 \frac{c}{\varepsilon_0} \frac{n_{ref}}{6\pi} |p_0|^2 . \quad (19)$$

We can now apply our knowledge from previously about Green's functions and integral equation methods, and the equation (19), to evaluate scattered power for a small particle.

If we consider a particle with complex dielectric constant $\varepsilon(\mathbf{r},\omega)$ the wave equation (8) can be conveniently written in the form

$$-\boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \boldsymbol{E}(\boldsymbol{r},\omega) + k_0^2 \varepsilon_{ref}(\omega) \boldsymbol{E}(\boldsymbol{r},\omega) = -k_0^2 \left(\varepsilon(\boldsymbol{r},\omega) - \varepsilon_{ref}(\omega)\right) \boldsymbol{E}(\boldsymbol{r},\omega). \quad (20)$$

An incident field $\mathbf{E}_0$ corresponding to the situation without the particle is a solution to the equation

$$-\boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \boldsymbol{E}_0(\boldsymbol{r},\omega) + k_0^2 \varepsilon_{ref}(\omega) \boldsymbol{E}_0(\boldsymbol{r},\omega) = 0. \quad (21)$$

By combining (20) and (21) we arrive at

$$-\boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \left(\boldsymbol{E}(\boldsymbol{r},\omega) - \boldsymbol{E}_0(\boldsymbol{r},\omega)\right) + k_0^2 \varepsilon_{ref}(\omega) \left(\boldsymbol{E}(\boldsymbol{r},\omega) - \boldsymbol{E}_0(\boldsymbol{r},\omega)\right) = i\omega\mu_0 \boldsymbol{J}(\boldsymbol{r},\omega), \quad (22)$$

where

$$i\omega\mu_0 \boldsymbol{J}(\boldsymbol{r},\omega) = -k_0^2 \left(\varepsilon(\boldsymbol{r},\omega) - \varepsilon_{ref}(\omega)\right) \boldsymbol{E}(\boldsymbol{r},\omega). \quad (23)$$

The field outside the particle can now be calculated using

$$\boldsymbol{E}(\boldsymbol{r},\omega) = \boldsymbol{E}_0(\boldsymbol{r},\omega) - i\omega\mu_0 \int \overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}';\omega) \cdot \boldsymbol{J}(\boldsymbol{r},\omega) d^3r'. \quad (24)$$

Notice that if we consider a very small particle we can ignore the dependence of $\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}';\omega)$ on $\mathbf{r}'$ for large distances to the center of the particle, in which case the electric field

$$\boldsymbol{E}(\boldsymbol{r},\omega) = \boldsymbol{E}_0(\boldsymbol{r},\omega) + \omega^2 \mu_0 \overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}_0;\omega) \cdot \boldsymbol{p}_0 , \quad (25)$$

is expressed in terms of the dipole moment

$$\boldsymbol{p}_0 = \varepsilon_0 \int \left(\varepsilon(\boldsymbol{r},\omega) - \varepsilon_{ref}(\omega)\right) \boldsymbol{E}(\boldsymbol{r},\omega) d^3r . \quad (26)$$

It is now possible to calculate the scattered power by inserting (26) in (19).

Consider the incident field of a plane wave, e.g.

$$\boldsymbol{E}_0(\boldsymbol{r},\omega) = \hat{z}E_0 e^{-ik_0 n_{ref}\,x} \ , \tag{27}$$

$$\boldsymbol{H}_0(\boldsymbol{r},\omega) = -\hat{y}\frac{k_0 n_{ref}}{\omega\mu_0}E_0 e^{-ik_0 n_{ref}\,x} \ , \tag{28}$$

with the Poynting vector

$$\boldsymbol{S}(\boldsymbol{r}) = \hat{x}2\frac{k_0 n_{ref}}{\omega\mu_0}|E_0|^2 \ . \tag{29}$$

In this case the power per unit area of the incident field passing through the surface perpendicular to the x-direction is given by (29). If we normalize the scattered power with the power of the incident field per unit area we arrive at the scattering cross section

$$\sigma_{scat} = k_0^4\frac{|p_0|^2/\varepsilon_0^2}{6\pi|E_0|^2} \ . \tag{30}$$

The time-averaged power lost due to absorption (Ohmic losses) is given by

$$P = \int\langle\left(\boldsymbol{E}_0(\boldsymbol{r},\omega)e^{i\omega t} + C.C.\right)\cdot\left(\boldsymbol{J}(\boldsymbol{r},\omega)e^{i\omega t} + C.C.\right)\rangle\, dV \tag{31}$$

$$= \int 2Real(\boldsymbol{E}_0(\boldsymbol{r},\omega)\cdot[\boldsymbol{J}(\boldsymbol{r},\omega)]^*)\, dV$$

$$= 2\omega\ Imag(\boldsymbol{E}_0(\boldsymbol{r},\omega)\cdot[\boldsymbol{p}_0]^*) \ .$$

Thereby the absorption cross section becomes (31) normalized with the magnitude of (29), i.e.

$$\sigma_{abs} = \frac{\omega\ Imag\ (\boldsymbol{E}_0(\boldsymbol{r},\omega)\cdot[\boldsymbol{p}_0]^*)}{\frac{k_0 n_{ref}}{\omega\mu_0}|E_0|^2} = \frac{k_0}{\varepsilon_0 n_{ref}}\frac{Imag\ (\boldsymbol{E}_0(\boldsymbol{r},\omega)\cdot[\boldsymbol{p}_0]^*)}{|E_0|^2} \ . \tag{32}$$

One approach to calculate the field is to solve the integral equation obtained by inserting (23) into (24). In this case we notice that if the distances considered between points inside the particle are very small we can approximate the Green's tensor with

$$\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}';\omega) \cong \left(3\frac{(\boldsymbol{r}-\boldsymbol{r}')(\boldsymbol{r}-\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|^2} - \overleftrightarrow{\boldsymbol{I}}\right)\frac{1}{4\pi k_0^2\varepsilon_{ref}\,|\boldsymbol{r}-\boldsymbol{r}'|^3} \ . \tag{33}$$

If we approximate the incident field according to the same principle for positions inside the particle we arrive at

$$\boldsymbol{E}_0(\boldsymbol{r},\omega) \cong \hat{z}E_0 \ . \tag{34}$$

Within the approximations (33) and (34) that are applicable inside and just outside a very small particle we obtain that the incident field and total field satisfy

$$\boldsymbol{\nabla}\times\boldsymbol{E}(\boldsymbol{r},\omega) \cong 0, \tag{35}$$

which means that to a good approximation we can calculate the field inside the particle with the methods known from electrostatics, i.e. we can express the electric field as minus the gradient of a scalar potential, and then calculate the potential that satisfies the boundary conditions at the surface of the particle, and which leads to a field decreasing as (33) away from the particle.

3. scattering from a very small spherical particle

According to the theory of electrostatics, if a spherical particle with dielectric constant ε_2 surrounded by a material with dielectric constant ε_1, is placed in an electric field $\boldsymbol{E}_0$ being constant across the particle, then the total field inside the particle is given by

$$\boldsymbol{E}(\boldsymbol{r}) = \boldsymbol{E}_0 \frac{3\varepsilon_1}{2\varepsilon_1+\varepsilon_2}, \tag{36}$$

which in our case leads to the dipole moment (26)

$$\boldsymbol{p}_0 = \varepsilon_0(\varepsilon_2 - \varepsilon_1)\frac{4\pi}{3} a^3 \boldsymbol{E}_0 \frac{3\varepsilon_1}{2\varepsilon_1+\varepsilon_2}, \tag{37}$$

where the material constants depend on the angular frequency. The resulting scattering and absorption cross sections are

$$\sigma_{scat} = k_0^4 \varepsilon_1^2 a^6 \frac{8\pi}{3} \left|\frac{\varepsilon_2-\varepsilon_1}{\varepsilon_2+2\varepsilon_1}\right|^2, \tag{38}$$

$$\sigma_{abs} = k_0 n_1 12\pi a^3 \frac{\varepsilon_1 Imag\,(-\varepsilon_2)}{|\varepsilon_2+2\varepsilon_1|^2}. \tag{39}$$

Notice that with the chosen sign convention the imaginary part of refractive index'es and dielectric constants is negative when there are absorption losses. Incidentally, the result in equation (39) is e.g. identical to Eq. (1) in the paper [2].

The above expression (36) can be derived by expressing the incident field in spherical coordinates

$$\boldsymbol{E}_0 = \hat{z}E_0 = -\nabla(-E_0 r\ \cos\theta), \tag{40}$$

and expressing the field inside the sphere (layer i=1) and outside the sphere (layer i=2) as the gradient of a potential, i.e. $\boldsymbol{E}(\boldsymbol{r}) = -\nabla\varphi_i$, where

$$\varphi_i = A_i r\cos\theta + B_i \frac{1}{r^2}\cos\theta, \tag{41}$$

and by requiring that the electromagnetic boundary conditions are satisfied across the surface of the particle, that the field and potential is not infinite in the center of the particle, and that for large distances from the particle the field should be just the incident field. This procedure is easily generalized to layered spheres, such as e.g. a gold coated polystyrene sphere, by expressing the field in each layer in the form (41).

4. Surface integral equation approach to scattering from a very small particle of general shape

For more complex particle shapes (only one material will be considered) a numerical method is required, and we will consider the surface integral equation method for the electric potential. The potential related to the incident field (φ_0) and to the total field (φ) both satisfy the equation $\nabla^2\varphi = 0$. For positions $\boldsymbol{r}$ inside the particle (*V*) the potential can be expressed in terms of the potential and its normal derivative on the particle surface via the surface integral equation, i.e.

$$\varphi(\boldsymbol{r}) = \oiint \left(g(\boldsymbol{r},\boldsymbol{r}')\hat{n}' \cdot \nabla' \varphi(\boldsymbol{r}') - \varphi(\boldsymbol{r}')\hat{n}' \cdot \nabla' g(\boldsymbol{r},\boldsymbol{r}') \right) da' \quad , \ \boldsymbol{r} \text{ inside } V \tag{42}$$

where the scalar electrostatic Green's function is given by

$$g(\boldsymbol{r},\boldsymbol{r}';\omega) = \frac{1}{4\pi|\boldsymbol{r}-\boldsymbol{r}'|}. \tag{43}$$

For positions outside the particle the potential is given by

$$\varphi(\boldsymbol{r}) = \varphi_0(\boldsymbol{r}) + \oiint \left(g(\boldsymbol{r},\boldsymbol{r}')\hat{n}' \cdot \nabla' \varphi(\boldsymbol{r}') - \varphi(\boldsymbol{r}')\hat{n}' \cdot \nabla' g(\boldsymbol{r},\boldsymbol{r}') \right) da' \quad , \ \boldsymbol{r} \text{ outside } V \tag{44}$$

where the latter expression satisfies the "radiating" boundary condition, namely that the scattered field should decrease away from the scatterer similar to the expression (33). By letting $\mathbf{r}$ approach the surface of the scatterer from either side we obtain self-consistent equations for the potential and its normal derivate on e.g. the outside of the particle surface if we apply the electromagnetics boundary conditions across the surface. This integral equation approach has been applied in ref. [3].

5. Scattering from layered spherical particles – Mie scattering theory

In the case of spherical particles it is convenient to expand the electric and magnetic fields in spherical harmonics centered at the center of the particle. For the incident field we will now consider a plane wave with the electric field polarized along the x-axis and propagating along the negative z-axis, i.e [4,5]

$$\boldsymbol{E}_{inc}(\boldsymbol{r}) = \hat{x}E_0 e^{ikz} = E_0 \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} \left(\boldsymbol{m}_{o1n}^{(1)} + i\boldsymbol{n}_{e1n}^{(1)} \right), \tag{45}$$

where $\boldsymbol{m}_{o1n}^{(1)}$ and $\boldsymbol{n}_{e1n}^{(1)}$ are spherical wave functions given by [4,5]

$$\boldsymbol{m}_{\substack{o\\e}1n}^{(1,3)} = \pm \frac{1}{\sin\theta} z_n^{(1,3)}(kR) P_n^1(\cos\theta) \begin{matrix}\cos\phi\\ \sin\phi\end{matrix} \hat{\theta} - z_n^{(1,3)}(kR) \frac{dP_n^1(\cos\theta)}{d\theta} \begin{matrix}\sin\phi\\ \cos\phi\end{matrix} \hat{\phi}, \tag{46}$$

$$\boldsymbol{n}_{\substack{o\\e}1n}^{(1,3)} = \frac{n(n+1)}{kR} z_n^{(1,3)}(kR) P_n^1(\cos\theta) \begin{matrix}\sin\phi\\ \cos\phi\end{matrix} \hat{r} + \frac{1}{kR} \left[kR z_n^{(1,3)}(kR) \right]' \frac{dP_n^1(\cos\theta)}{d\theta} \begin{matrix}\sin\phi\\ \cos\phi\end{matrix} \hat{\theta} \pm \frac{1}{kR\sin\theta} \left[kR z_n^{(1,3)}(kR) \right]' P_n^1(\cos\theta) \begin{matrix}\cos\phi\\ \sin\phi\end{matrix} \hat{\phi} \quad . \tag{47}$$

Here ´ means the derivative with respect to the argument (kR), P_n^1 is a Legendre function, $z_n^{(1)}$ is a spherical Bessel function, and $z_n^{(3)}$ is a spherical Hankel function. The spherical Bessel function is given by

$z_n^{(1)}(x) = \sqrt{\frac{\pi}{2KR}} J_{n+\frac{1}{2}}(x)$, where $J_n(x)$ is the ordinary Bessel function, and the spherical Hankel function is given by $z_n^{(3)}(x) = \sqrt{\frac{\pi}{2KR}} H_{n+\frac{1}{2}}^{(2)}(x)$, where $H_n^{(2)}(x)$ is the Hankel function of order zero and second kind.

The electric and magnetic fields in each layer (j) generated in response to the incident field (45) of a layered spherical particle can be expressed in the form [4,5]

$$\boldsymbol{E}_j(\boldsymbol{r}) = E_0 \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} \left(a_{n.j}^{(1)} \boldsymbol{m}_{o1n}^{(1)} + a_{n.j}^{(3)} \boldsymbol{m}_{o1n}^{(3)} + i b_{n.j}^{(1)} \boldsymbol{n}_{e1n}^{(1)} + i b_{n.j}^{(3)} \boldsymbol{n}_{e1n}^{(3)} \right), \tag{48}$$

$$\boldsymbol{B}_j(\boldsymbol{r}) = -\left(\frac{E_0}{c/\sqrt{\varepsilon_j}}\right) \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} \left(b_{n.j}^{(1)} \boldsymbol{m}_{e1n}^{(1)} + b_{n.j}^{(3)} \boldsymbol{m}_{e1n}^{(3)} - i a b_{n.j}^{(1)} \boldsymbol{n}_{o1n}^{(1)} - i a_{n.j}^{(3)} \boldsymbol{n}_{o1n}^{(3)} \right). \tag{49}$$

Here we have used that the incident field only has terms $\cos\phi$ and $\sin\phi$ and not the general $\cos m\phi$ and $\sin m\phi$ requiring a further summation over m. This simplification is the main reason for choosing an incident wave propagating along the z-axis.

The boundary conditions in this case are that the electromagnetics boundary conditions must be fulfilled across interfaces, i.e. we require continuity of the tangential electric and magnetic field components, the field is not allowed to be infinitely large in the center of the particle, and outside the particle the field must be the sum of the given incident field and a field component propagating away from the particle.

Consider that j=N is the outer medium surrounding the particle.In this case the scattering boundary condition gives $a_{n.j}^{(1)} = 1$, and $b_{n.j}^{(1)} = 1$, and the scattered electric field (**E**-**E**$_0$) and magnetic field (**B**-**B**$_0$) are

$$\boldsymbol{E}_{scat}(\boldsymbol{r}) = E_0 \sum_{n=1}^{\infty} i^n \frac{2n+1}{n(n+1)} \left(a_{n.j}^{(3)} \boldsymbol{m}_{o1n}^{(3)} - i b_{n.j}^{(3)} \boldsymbol{n}_{e1n}^{(3)} \right), \tag{50}$$

$$\boldsymbol{B}_{scat}(\boldsymbol{r}) = -\left(\frac{E_0}{c/\sqrt{\varepsilon_j}}\right) \sum_{n=1}^{\infty} i^n \frac{2n+1}{n(n+1)} \left(b_{n.j}^{(3)} \boldsymbol{m}_{e1n}^{(3)} + i a_{n.j}^{(3)} \boldsymbol{n}_{o1n}^{(3)} \right). \tag{51}$$

The scattered power can now be calculated using the Poynting vector of the scattered field, i.e.

$$P_{scat} = \oint\!\!\oint 2Real(\boldsymbol{E}_{scat}(\boldsymbol{r}) \times [\boldsymbol{H}_{scat}(\boldsymbol{r})]^*) \cdot \hat{n} da$$

$$= -\frac{|E_0|^2}{c/\sqrt{\varepsilon}} \sum_{n=1}^{\infty} \left(\frac{2n+1}{n(n+1)}\right)^2 \oint\!\!\oint 2Imag\left(\left|a_{n.j}^{(3)}\right|^2 \boldsymbol{m}_{o1n}^{(3)} \times \left[\boldsymbol{n}_{o1n}^{(3)}\right]^* + \left|b_{n.j}^{(3)}\right|^2 \boldsymbol{n}_{e1n}^{(3)} \times \left[\boldsymbol{m}_{e1n}^{(3)}\right]^* \right) \cdot \hat{n} da, \tag{52}$$

where we have already applied orthogonality between the even (e) and the odd (o) wave functions (even and odd in ϕ) and the relation

$$\int_0^{\pi} \left(\frac{P_n^1(\cos\theta)}{\sin\theta} \frac{P_{n'}^1(\cos\theta)}{\sin\theta} + \frac{dP_n^1(\cos\theta)}{d\theta} \frac{dP_{n'}^1(\cos\theta)}{d\theta} \right) \sin\theta \, d\theta = \delta_{nn'} \frac{2}{2n+1} (n(n+1))^2. \tag{53}$$

Furthermore, we can show by e.g. inserting the large-argument approximation for the Hankel function that

$$\frac{1}{kR} \left(\left[kR z_n^{(3)}(kR)\right]' \right)^* z_n^{(3)}(kR)' = \frac{1}{(kR)^2} i. \tag{54}$$

The expression (52) now leads to

$$P_{scat} = \frac{2\pi}{k^2}\frac{\sqrt{\varepsilon}}{c\mu_0}|E_0|^2 \sum_{n=1}^{\infty}(2n+1)\left(\left|a_{n.j}^{(3)}\right|^2 + \left|b_{n.j}^{(3)}\right|^2\right), \quad (55)$$

where j and k refer to the surrounding medium.

For the incident field we find

$$\boldsymbol{E}_{inc}(\boldsymbol{r}) \times [\boldsymbol{H}_{inc}(\boldsymbol{r})]^* = -\hat{z}|E_0|^2\sqrt{\frac{\varepsilon_0}{\mu_0}}\sqrt{\varepsilon}. \quad (56)$$

If we now normalize (55) with the magnitude of (56) we find the scattering cross section

$$\sigma_{scat} = \frac{2\pi}{k^2}\sum_{n=1}^{\infty}(2n+1)\left(\left|a_{n.j}^{(3)}\right|^2 + \left|b_{n.j}^{(3)}\right|^2\right). \quad (57)$$

We can calculate the absorption as the total average power into a closed surface surrounding the particle, in which case we should use the total field instead of the scattered field, i.e.

$$P_{abs} = -\oiint 2Real(\boldsymbol{E}(\boldsymbol{r}) \times [\boldsymbol{H}(\boldsymbol{r})]^*) \cdot \hat{n}da =$$

$$-P_{scat} - \oiint 2Real(\boldsymbol{E}_0(\boldsymbol{r}) \times [\boldsymbol{H}_{scat}(\boldsymbol{r})]^* + \boldsymbol{E}_{scat}(\boldsymbol{r}) \times [\boldsymbol{H}_0(\boldsymbol{r})]^*) \cdot \hat{n}da \ , \quad (58)$$

such that the absorption cross section is given by

$$\sigma_{abs} = -\sigma_{scat} + \frac{2\pi}{k^2}\sum_{n=1}^{\infty}(2n+1)\left(Real(a_{n.j}^{(3)} + b_{n.j}^{(3)})\right). \quad (59)$$

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