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# Absorption and scattering of light from ensembles of randomly oriented aggregates

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**Abstract**

Ensembles of aggregates are important in the areas of aerosols and combustion physics. This paper presents one approach to the absorption and scattering of light from aggregates where the individual primary particles are small compared to the wavelength, whereas the aggregate can be large compared to the wavelength. The method is related to the Rayleigh-Debye-Gans (RDG) theory. The difference is that the near field interaction between primary particles is included and that the primary particles can have arbitrary shape, overlap each other and have a space dependent index of refraction. Closed form expressions are presented for the absorption and scattered intensity of an ensemble of aggregates with random position and orientation. These expressions give fast and accurate numerical evaluations of the scattering and absorption from ensembles of aggregates. The numerical results are compared with the ones obtained from the T-matrix method and the discrete dipole approximation method.

# 1 Introduction

The Rayleigh-Debye-Gans theory is an important tool for applications where waves are scattered from objects that are small compared to the wavelength. As long as the material properties are linear it applies to all wavelengths. Even though there are more accurate methods, the RDG method is often still preferred since it greatly simplifies the analysis and is considered to be accurate enough for many applications. The RDG method is also an important tool for understanding the physics of the scattering problem [18]. In the RDG-theory the objects are considered to be non-overlapping spheres. The reason for this is that it utilizes the analytic closed form expression for the polarization of a sphere in a constant electric field. For soot particle aggregates, the RDG theory gives inaccurate results for the scattered intensities and the absorbed powers and this inaccuracy increases as the wavelength decreases. The absorption was analyzed in [11], where the RDG method was compared with the generalized multi-sphere Mie-solution (GMM) [19], which is very accurate for aggregates of non-overlapping spherical particles, and an approximate method referred to as the electrostatic approximation (ESA) [14]. For a sphere radius of 15 nm and a wavelength of 1064 nm the ESA and GMM give quite similar results, whereas RDG gives 1-5% errors. When the wavelength decreased to 532 nm the error for ESA is less than 6 % and RDG gives an error that is twice that of the ESA. The drawback of the GMM and ESA methods are that they can only be applied to aggregates that consist of non-overlapping homogeneous spheres. This is a severe restriction since real aggregates consist of overlapping primary particles, see *e.g.*, transmission electron micrograph (TEM) images of soot aggregates in refs. [18] and [4]. A more useful method for determining absorption and scattering from aggregates is the discrete dipole approximation (DDA) [6]. It is not restricted to spheres and can handle in principle any shape of the primary particles. The drawback with this method is that the averaging over an ensemble of aggregates is very time consuming.

In the paper [9] the RDG method was generalized to particles of arbitrary shape

and inner structure. The polarization is then determined by numerically solving a quasi static problem with the entire object in an external constant electric field. The near field interaction between the primary particles is thus included. In this paper the method in [9] is referred to as the generalized RDG (G-RDG) method. The method was in [9] compared with good agreement with the T-matrix method [3], [17], [1], [12]. In the case of non-overlapping spheres the ESA method and G-RDG method are very similar, even though they use different techniques. The G-RDG method is in this paper applied to absorption and scattering from an ensemble of randomly oriented aggregates. The averaging over an ensemble of aggregates with random orientation is beneficial for the G-RDG method and it can be used for a large class of aggregates and wavelengths. In the numerical section it is compared with the discrete dipole approximation (DDA) [6], and the T-matrix method for the case of non-overlapping spheres and to DDA for aggregates that consist of overlapping spheres. The G-RDG is accurate for aggregates of overlapping and non-overlapping particles with radius 10 nm and wavelengths longer than 532 nm.

The numerical section considers absorption and scattering from ensembles of aggregates that are identical. In a real case one has to take into account the fact that an ensemble consists of aggregates with different number of particles and different fractal dimensions. It was shown in [10] that even a small deviation of the structure of an aggregate leads to large variations in the averaged scattered intensity, whereas the absorption is almost unaffected. A large number of different types of aggregates require a very fast solver, such as the one presented in this paper.

# 2 Prerequisetes

Assume an ensemble of a large number of randomly distributed and oriented aggregates confined in a volume $V$. The volume is illuminated by a monochromatic linearly polarized plane wave with a wavelength $\lambda$. The volume $V$ has three length scales. The first scale is the diameter of the volume, which is much larger than the vacuum wavelength. The second length scale is the typical diameter of an aggregate. This is considered to be in the range $0.1\lambda - 10\lambda$. The third scale is the diameter of a primary particle, or monomer, of an aggregate. In this paper the diameter of the monomers is considered to be less than 0.05 $\lambda$. The mean distance between aggregates is very large compared to the wavelength.

Let $xyz$ be a cartesian coordinate system with its origin at one of the aggregates. With the time convention $e^{-\mathrm{i}\omega t}$ the incident field reads

$$\boldsymbol{E}^i(\boldsymbol{r}) = \hat{\boldsymbol{e}}_i E_0 e^{\mathrm{i}\boldsymbol{k}_i\cdot\boldsymbol{r}} \tag{2.1}$$

where $\hat{\boldsymbol{e}}_i$ is a unit vector, $E_0$ is a real valued amplitude, $\boldsymbol{k}_i = k\hat{\boldsymbol{k}}_i$ is the vacuum wave vector and $k = \omega/c$. The scattered electric far field from the volume is given by

$$\boldsymbol{E}^s(\boldsymbol{r}) = -\frac{k^2 e^{\mathrm{i}kr}}{4\pi\varepsilon_0 r}\hat{\boldsymbol{r}} \times \left( \hat{\boldsymbol{r}} \times \iiint_V \boldsymbol{P}(\boldsymbol{r'}) e^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r'}}\,\mathrm{d}v' \right) \tag{2.2}$$

where $\boldsymbol{r} = r\hat{\boldsymbol{r}}$ and

$$\boldsymbol{q} = k(\hat{\boldsymbol{k}}_i - \hat{\boldsymbol{r}}) \tag{2.3}$$

The induced polarization $\boldsymbol{P}(\boldsymbol{r}')$ is defined as

$$\boldsymbol{P}(\boldsymbol{r}') = \varepsilon_0 \left(\varepsilon_c(\boldsymbol{r}') - 1\right) \boldsymbol{E}(\boldsymbol{r}') e^{-\mathrm{i}\boldsymbol{k}_i \cdot \boldsymbol{r}'}$$

where $\boldsymbol{E}(\boldsymbol{r})$ is the total electric field in the object and where the complex permittivity is related to the complex refractive index $m$ as $\varepsilon_c = \varepsilon_r + \mathrm{i}\dfrac{\sigma}{\omega\varepsilon_0} = m^2$. The factor $e^{-\mathrm{i}\boldsymbol{k}_i \cdot \boldsymbol{r}}$ is added in order to remove the phase of the incident field in the polarization vector. In the rest of the paper the complex permittivity is considered to be constant in the aggregate. It is straightforward to generalize to a space dependent permittivity.

The time average of the scattered power flow density, or intensity, is given by the Poynting vector in the far zone

$$\boldsymbol{S}_s = \frac{1}{2}\mathrm{Re}\left\{\boldsymbol{E}^s(\boldsymbol{r}) \times \boldsymbol{H}^{s*}(\boldsymbol{r})\right\} = \frac{\eta_0}{2}|\boldsymbol{E}^s|^2 \hat{\boldsymbol{r}}$$

where $|\boldsymbol{E}^s|$ is defined by $|\boldsymbol{E}^s| = \sqrt{\boldsymbol{E}^s \cdot \boldsymbol{E}^{s*}}$ and $\eta_0$ is the wave impedance. The normalized scattering intensity is given by

$$I(\boldsymbol{r}) = \frac{|\boldsymbol{E}^s(\boldsymbol{r})|^2}{E_0^2} \tag{2.4}$$

## 2.1 Co and cross polarization

The intensity can be measured in two perpendicular directions. The vectors $\hat{\boldsymbol{k}}_i$ and $\hat{\boldsymbol{e}}_i$ in Eq. (2.1) are perpendicular and hence the three vectors $(\hat{\boldsymbol{k}}_i \times \hat{\boldsymbol{e}}_i, \hat{\boldsymbol{k}}_i, \hat{\boldsymbol{e}}_i) = (\hat{\boldsymbol{f}}, \hat{\boldsymbol{k}}_i, \hat{\boldsymbol{e}}_i)$ define a right-handed cartesian coordinate system. Now introduce spherical coordinates $(r, \alpha, \beta)$ with unit vectors $\hat{\boldsymbol{r}}$, $\hat{\boldsymbol{\alpha}}$ and $\hat{\boldsymbol{\beta}}$, where $\hat{\boldsymbol{r}}$ is directed to the field point and $\alpha$ is the angle between $\hat{\boldsymbol{e}}_i$ and $\hat{\boldsymbol{r}}$, *i.e.*,

$$\begin{aligned}
\hat{\boldsymbol{r}} &= \sin\alpha\cos\beta\hat{\boldsymbol{f}} + \sin\alpha\sin\beta\hat{\boldsymbol{k}}_i + \cos\alpha\hat{\boldsymbol{e}}_i \\
\hat{\boldsymbol{\alpha}} &= \cos\alpha\cos\beta\hat{\boldsymbol{f}} + \cos\alpha\sin\beta\hat{\boldsymbol{k}}_i - \sin\alpha\hat{\boldsymbol{e}}_i \\
\hat{\boldsymbol{\beta}} &= -\sin\beta\hat{\boldsymbol{f}} + \cos\beta\hat{\boldsymbol{k}}_i
\end{aligned} \tag{2.5}$$

The intensity is the sum of the co and cross polarization intensities that are defined as

$$\begin{aligned}
I &= I_{\mathrm{co}} + I_{\mathrm{cross}} \\
I_{\mathrm{co}} &= \frac{1}{E_0^2}|\hat{\boldsymbol{\alpha}} \cdot \boldsymbol{E}^s|^2 \\
I_{\mathrm{cross}} &= \frac{1}{E_0^2}|\hat{\boldsymbol{\beta}} \cdot \boldsymbol{E}^s|^2
\end{aligned} \tag{2.6}$$

In some papers, *e.g.*, [18], the notation $I_{VV}$ is used for $I_{\mathrm{co}}$ and $I_{VH}$ for $I_{\mathrm{cross}}$.

## 2.2 Absorbed power

The time average of the absorbed power in the ensemble is given by

$$W_a = \frac{1}{2}\frac{\sigma}{|\varepsilon_0(\varepsilon_c - 1)|^2}\int_V |\boldsymbol{P}(\boldsymbol{r})|^2 \,\mathrm{d}v = \frac{1}{2}\frac{\omega \mathrm{Im}\{m^2\}}{\varepsilon_0 |m^2 - 1|^2}\int_V |\boldsymbol{P}(\boldsymbol{r})|^2 \,\mathrm{d}v \tag{2.7}$$

where $\sigma$ =conductivity.

## 2.3 The aggregates

The aggregates that are of particular interest in this paper are soot aggregates formed during incomplete combustion, *cf.*, [18] and [4], even though the method can be applied to other structures. The primary particles of an aggregate are sphere-like particles that overlap each other. A commonly used model is that the number of primary particles, $N_A$, of an aggregate increases with the radius of the aggregate according to the formula, *cf.*, [2] and [13],

$$N_A = k_f \left(\frac{R_g}{a}\right)^{D_f} \tag{2.8}$$

Here $D_f$ is the fractal dimension, $k_f$ is fractal pre factor, $a$ is the radius of a primary particle and $R_g$ is the radius of gyration, defined as

$$R_g = \sqrt{\frac{1}{N_A}\sum_{n=1}^{N_A} |\boldsymbol{r}_n - \boldsymbol{r}_{\mathrm{mean}}|^2}$$

$$\boldsymbol{r}_{\mathrm{mean}} = \frac{1}{N_A}\sum_{n=1}^{N_A} \boldsymbol{r}_n$$

where $\boldsymbol{r}_n$ is the vector to the center of primary particle number $n$. These fractal aggregates are considered in the numerical examples.

# 3 Polarization $\boldsymbol{P}(\boldsymbol{r})$ and dipole moment $\boldsymbol{p}$

It is clear from Eqs. (2.2) and (2.7) that the absorption and scattering problems are solved once the polarization $\boldsymbol{P}(\boldsymbol{r})$ is known in the aggregate. The full wave methods, like DDA, determines $\boldsymbol{P}(\boldsymbol{r})$ with high accuracy, whereas the RDG method gives an approximate closed form expression. In this paper the method presented in [9] is utilized. The polarization is obtained by solving a quasi-static problem. It provides a solution that is more accurate than the RDG method and more flexible than the full wave solutions. The polarization vector $\boldsymbol{P}(\boldsymbol{r})$ in the aggregate for a space independent external electric field $E_0\hat{\boldsymbol{e}}_i = E_0(e_{ix}, e_{iy}, e_{iz})$ can approximately be expressed in terms of a polarization matrix $\mathbf{P}(\boldsymbol{r})$ as

$$\boldsymbol{P}(\boldsymbol{r}) = \mathbf{P}(\boldsymbol{r}) \begin{pmatrix} e_{ix} \\ e_{iy} \\ e_{iz} \end{pmatrix} E_0 \tag{3.1}$$

The polarization matrix is symmetric.

## 3.1 Numerical determination of $\mathbf{P}(\boldsymbol{r})$

The method introduced in [9] of determining the polarization matrix utilizes the finite element method. The aggregate is placed with its center close to the origin. A sufficiently large sphere, or cube, is placed around the aggregate. The diameter of the sphere should be large enough such that it does not affect $\mathbf{P}(\boldsymbol{r})$, *i.e.*, on the order of ten times the diameter of the aggregate. First a space independent field $\boldsymbol{E} = (1, 0, 0)$ V/m is applied and $(\mathrm{P}_{11}(\boldsymbol{r}), \mathrm{P}_{21}(\boldsymbol{r}), \mathrm{P}_{31}(\boldsymbol{r}))$ are determined. The simplest way of applying $\boldsymbol{E}$ is to let the potential on the surface of the sphere, or cube, equal $-x$. Next the field $\boldsymbol{E} = (0, 1, 0)$ V/m is applied and $(\mathrm{P}_{12}(\boldsymbol{r}), \mathrm{P}_{22}(\boldsymbol{r}), \mathrm{P}_{32}(\boldsymbol{r}))$ are determined and finally $\boldsymbol{E} = (0, 0, 1)$ V/m is applied to obtain $(\mathrm{P}_{13}(\boldsymbol{r}), \mathrm{P}_{23}(\boldsymbol{r}), \mathrm{P}_{33}(\boldsymbol{r}))$. By solving these three quasi static problems the polarization matrix is known in the entire aggregate. In the numerical section $\mathbf{P}(\boldsymbol{r})$ was obtained by the commercial finite element method program Comsol. It is then very easy and fast to obtain $\mathbf{P}(\boldsymbol{r})$, even for a complicated strucure.

## 3.2 The dipole moments of primary particles

An aggregate consists of $N_A$ primary particles, with volumes $V_{sn}$, $n = 1 \ldots N_A$. There are no restrictions on the shapes of the primary particles and they can overlap. A polarizability matrix can be defined for every primary particle as

$$\mathbf{p}_n = \int_{V_{sn}} \mathbf{P}(\boldsymbol{r}) \, \mathrm{d}v$$

In some applications the notation $\alpha$ is used for the polarizability matrix. The dipole moment for the primary particle for the incident field $\boldsymbol{E}^i(\boldsymbol{r})$ in Eq. (2.1) is given by

$$\boldsymbol{p}_n = \mathbf{p}_n \begin{pmatrix} e_{ix} \\ e_{iy} \\ e_{iz} \end{pmatrix} E_0 \tag{3.2}$$

## 3.3 Averages of $\boldsymbol{P}(\boldsymbol{r})$ and $\boldsymbol{p}_n$

Consider an aggregate with fixed orientation. Let the direction $\hat{\boldsymbol{e}}_i$ of the applied electric field $E_0\hat{\boldsymbol{e}}_i$ alter over all directions and determine the average of the polarization along $\hat{\boldsymbol{e}}_i$. This is equivalent to keeping the direction of $\hat{\boldsymbol{e}}_i$ constant and rotating the aggregate in all directions. It is quite straightforward to see that the average of the polarization parallel to the electric field is

$$\tilde{\boldsymbol{P}}(\boldsymbol{r}) = \hat{\boldsymbol{e}}_i \frac{1}{3} \mathrm{tr}\{\mathbf{P}(\boldsymbol{r})\} E_0 \tag{3.3}$$

where $\mathrm{tr}\{\mathbf{P}(\boldsymbol{r})\} = \mathbf{P}_{11}(\boldsymbol{r}) + \mathbf{P}_{22}(\boldsymbol{r}) + \mathbf{P}_{33}(\boldsymbol{r})$ is the trace of the matrix $\mathbf{P}(\boldsymbol{r})$. The averaged polarization in a direction perpendicular to $\hat{\boldsymbol{e}}_i$ is zero. By introducing the matrix $\tilde{\mathbf{P}}(\boldsymbol{r})$ as

$$\tilde{\mathbf{P}}(\boldsymbol{r}) = \frac{1}{3} \mathrm{tr}\{\mathbf{P}(\boldsymbol{r})\} \mathbf{I},$$

where $\mathbf{I}$ is the unit matrix, the polarization matrix, for a fixed orientation of the aggregate, can be decomposed as

$$\mathbf{P}(\boldsymbol{r}) = \tilde{\mathbf{P}}(\boldsymbol{r}) + \Delta\mathbf{P}(\boldsymbol{r})$$

The polarization vector for a single aggregate with fixed orientation in a fixed external field can be expressed as

$$\boldsymbol{P}(\boldsymbol{r}) = \tilde{\boldsymbol{P}}(\boldsymbol{r}) + \Delta\boldsymbol{P}(\boldsymbol{r}) \tag{3.4}$$

where the average of $\Delta\boldsymbol{P}(\boldsymbol{r})$ over all directions of the external field $E_0\hat{\boldsymbol{e}}_i$ is zero.

The averaged polarizability matrix is defied as

$$\tilde{\mathrm{p}}_n = \frac{1}{3}\mathrm{tr}\{\mathbf{p}_n\}\mathbf{I} \tag{3.5}$$

or, equivalently,

$$\tilde{\mathrm{p}}_n = \int_{V_{sn}} \tilde{\mathbf{P}}(\boldsymbol{r})\,\mathrm{d}v$$

where $V_{sn}$ is the volume of primary particle $n$. The polarizability matrix is decomposed as

$$\mathbf{p}_n = \tilde{\mathbf{p}}_n + \Delta\mathbf{p}_n$$

where $\Delta\mathrm{p}_{nij} = \mathrm{p}_{nij}$ for $i \neq j$ and $\Delta\mathrm{p}_{n11} + \Delta\mathrm{p}_{n22} + \Delta\mathrm{p}_{n33} = \mathbf{0}$.

The average of the dipole moment for the incident field in Eq. (2.1) is

$$\tilde{\boldsymbol{p}}_n = \tilde{\mathbf{p}}_n \begin{pmatrix} e_{ix} \\ e_{iy} \\ e_{iz} \end{pmatrix} E_0 = \hat{\boldsymbol{e}}_i \frac{1}{3}\mathrm{tr}\{\mathbf{p}_n\}E_0 \tag{3.6}$$

The dipole moment of primary particle $n$ for a given orientation of the aggregate is

$$\boldsymbol{p}_n = \tilde{\boldsymbol{p}}_n + \Delta\boldsymbol{p}_n \tag{3.7}$$

where $\Delta\boldsymbol{p}_n$ has zero average value for an ensemble of aggregates and is, in general, not parallel with the incident field.

# 4 Scattering and absorption from a single aggregate

Again consider an aggregate with fix orientation. Without approximations the far-field amplitude and the intensity are obtained from Eqs. (2.2) and (2.4), respectively.

In order to speed up the calculations one can utilize the induced dipole moments $\boldsymbol{p}_n$ for each primary particle and write the scattered far-field as the sum

$$\boldsymbol{E}^s(\boldsymbol{r}) = -\frac{k^2 e^{\mathrm{i}kr}}{4\pi\varepsilon_0 r}\hat{\boldsymbol{r}} \times \left(\hat{\boldsymbol{r}} \times \left(\sum_{n=1}^{N_A} \boldsymbol{p}_n e^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_n}\right)\right) \tag{4.1}$$

where $\boldsymbol{r}_n$ is the position of the center of the primary particle. In terms of the dipole moments the co and cross polarization intensities are

$$\begin{aligned} I_{\text{co}} &= \frac{k^4}{(4\pi E_0 \varepsilon_0 r)^2} \left| \sum_{n=1}^{N_A} \hat{\boldsymbol{\alpha}} \cdot \boldsymbol{p}_n e^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_n} \right|^2 \\ I_{\text{cross}} &= \frac{k^4}{(4\pi E_0 \varepsilon_0 r)^2} \left| \sum_{n=1}^{N_A} \hat{\boldsymbol{\beta}} \cdot \boldsymbol{p}_n e^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_n} \right|^2 \end{aligned} \tag{4.2}$$

The absorbed power is preferably obtained from Eq. (2.7). It is not appropriate to express the absorbed power in terms of the dipole moments, since that introduces errors.

# 5 Scattering and absorption from an ensemble of aggregates

The main results of this paper are the closed form expressions for the scattered intensity and the absorbed power of an ensemble of aggregates. These results are presented here and are derived in the appendix.

Consider the volume $V$ with an ensemble of $M$ identical, but randomly distributed and oriented, aggregates. The mean distance between the aggregates is much longer than the wavelength and hence the phase differences between the scattered fields from the different aggregates are random. The total absorbed power equals $M$ times the average of the absorbed power of one aggregate, where the averaging is over all rotations of the aggregate. Since the distance between aggregates is large and random the total scattered intensity equals $M$ times the average of the intensity of one aggregate.

Each aggregate in the ensemble consists of $N_A$ primary particles. The coordinate system introduced in Eq. (2.5) is used. Then the co and cross polarization intensities are given by

$$\begin{aligned} I_{\text{co}}(\boldsymbol{r}) = \frac{Mk^4}{32\pi^2\varepsilon_0^2 r^2} \sum_{n=1}^{N_A} \Bigg[ \frac{2}{9}\sin^2(\alpha) \sum_{n'=1}^{N_A} \left( \text{tr}\{\tilde{\mathbf{p}}_n\}\text{tr}\{\tilde{\mathbf{p}}_{n'}^*\} \frac{\sin(|\boldsymbol{q}|r_{nn'})}{|\boldsymbol{q}|r_{nn'}} \right) \\ + \frac{3+\sin^2\alpha}{15} \text{tr}\{\Delta\mathbf{p}_n \Delta\mathbf{p}_n^*\} \Bigg] \end{aligned} \tag{5.1}$$

and

$$I_{\text{cross}}(\boldsymbol{r}) = \frac{Mk^4}{160\pi^2\varepsilon_0^2 r^2} \sum_{n=1}^{N_A} \text{tr}\{\Delta\mathbf{p}_n \Delta\mathbf{p}_n^*\} \tag{5.2}$$

where $r_{nn'} = |\boldsymbol{r}_n - \boldsymbol{r}_{n'}| = \sqrt{(x_n - x_{n'})^2 + (y_n - y_{n'})^2 + (z_n - z_{n'})^2}$, $\alpha$ is the angle between $\boldsymbol{r}$ and $\hat{\boldsymbol{e}}_i$, and $\boldsymbol{q}$ is the vector defined in Eq. (2.3). Notice that $\hat{\boldsymbol{r}} \cdot \hat{\boldsymbol{k}}_i =$

$\cos\gamma$, where $\gamma$ is the angle between $\hat{\boldsymbol{r}}$ and $\hat{\boldsymbol{k}}_i$ and thus $|\boldsymbol{q}| = 2k\sin\left(\frac{\gamma}{2}\right)$, which is a commonly used expression. One may use an arbitrary orientation of the aggregate when these intensities are calculated. The results are, as expected, independent of the orientation. If one cannot decompose the volume into sub-particles with corresponding dipole matrices, then one can obtain the intensities by integration as in Eqs. (A.11) and (A.12).

The absorbed power is given by

$$W_a = \frac{M\sigma E_0^2}{6|\varepsilon_0(\varepsilon_c - 1)|^2}\int_{V_a} \mathrm{tr}\{\mathbf{P}(\boldsymbol{r})\mathbf{P}^*(\boldsymbol{r})\}\mathrm{d}v \tag{5.3}$$

Since the matrix $\Delta\mathbf{p}_n$ is symmetric, the explicit expression for $\mathrm{tr}\{\Delta\mathbf{p}_n\Delta\mathbf{p}_n^*\}$ is

$$\mathrm{tr}\{\Delta\mathbf{p}_n\Delta\mathbf{p}_n^*\} = |\Delta\mathrm{p}_{n11}|^2 + |\Delta\mathrm{p}_{n22}|^2 + |\Delta\mathrm{p}_{n33}|^2 + 2\left(|\Delta\mathrm{p}_{n12}|^2 + |\Delta\mathrm{p}_{n13}|^2 + |\Delta\mathrm{p}_{n23}|^2\right)$$

The same expansion is valid for $\mathrm{tr}\{\mathbf{P}(\boldsymbol{r})\mathbf{P}^*(\boldsymbol{r})\}$. This means that the average of the absorbed power is the average of the absorbed power for an electric field in three perpendicular directions.

Equations (5.1), (5.2) and (5.3) are the main results of this paper and are derived in the appendix. By using a finite element method program such as Comsol it is very easy and fast to obtain the intensities and absorbed powers even for complicated structures. It is obvious that one can generalize the expressions to an ensemble that consists of many different aggregates.

# 6 Numerical results

The present method, a generalized Rayleigh Debye Gans method (G-RDG), is an approximate method that has limitations. There are restrictions on the diameter of the primary particles compared to the wavelength and on the fractal dimension of the aggregate. As will be seen a wavelength of 532 nm and a diameter of 20 nm or less gives errors in intensities and absorption that are less than a couple of percent for an aggregate of 20 particles, fractal dimension 1.8 and an index of refraction $m = 1.61 + i0.59$, [5]. The size of the aggregate can be extended to at least one hundred particles. The errors increase with increasing diameter to wavelength ratio and with increasing fractal dimension, and at some stage it is wise to switch to a more accurate method. In that case the discrete dipole approximation is a good alternative since it can handle overlapping spheres. The calculations in this section have been done using the free software DDSCAT [7], for the discrete dipole approximation, MSTM [15] for the T-matrix method and the commercial program Comsol for the finite element method calculations in the G-RDG method.

First the absorption from an ensemble of identical aggregates with random orientation is considered. The examples are similar to an example in [11]. The first example, see Figure 1, shows the averaged absorption for aggregates with fractal dimension $D_f = 1.8$ and $k_f = 2.3$, *cf.*, Eq. (2.8), and 1, 5, 10 and 20 non-overlapping

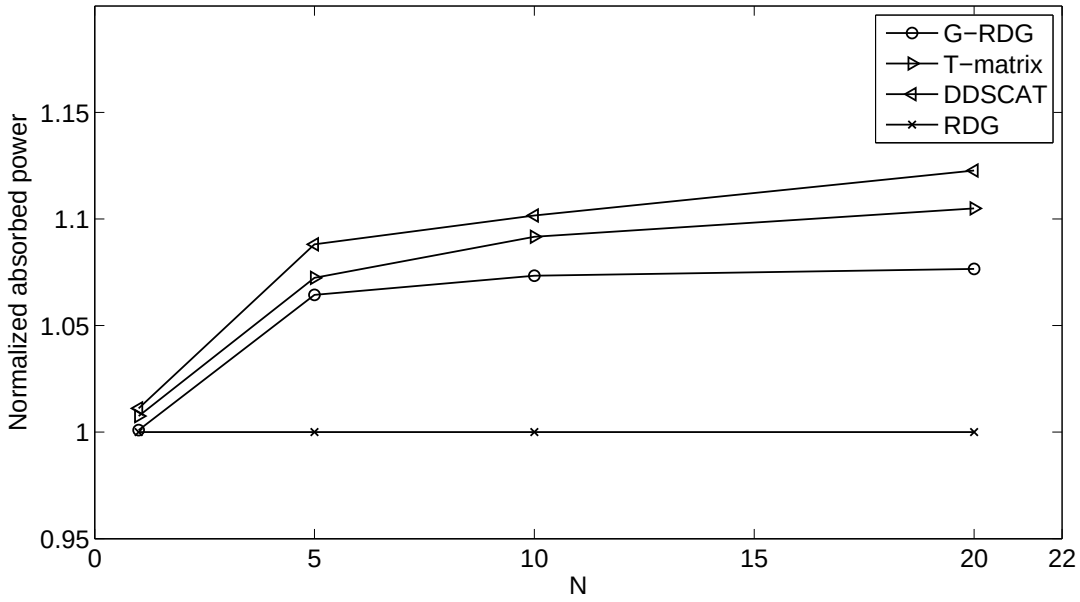


**Figure 1**: A comparison between the averaged absorbed power for the RDG method, the G-RDG method, the DDA method and the T-matrix method for an ensemble of identical aggregates, with random orientation. Each aggregate consists of $N$ non-overlapping spheres with radius 10 nm, $k_f = 2.3$ and $D_f = 1.8$. The wavelength is 532 nm. All of the results are normalized with the absorbed power obtained by the RDG method. The results from the T-matrix method are considered to be very accurate.

spheres. The absorbed power is normalized with the absorbed power using the RDG approximation, *i.e.*,

$$W_{\mathrm{RDG}} = \frac{9E_0^2 V_a \varepsilon_0 \omega \mathrm{Im}\{m^2\}}{2|m^2+2|^2}$$

where $V_a$ is the volume of the aggregate. The T-matrix method, the discrete dipole approximation, the G-RDG method and the RDG method are compared for the wavelength 532 nm. The T-matrix method is known to give very accurate results and its values are assumed to be correct, *cf.*, [16], [12] and [3]. It is seen that the error for the RDG-method is on the order of 10% when $N > 10$ whereas the G-RDG method and the DDA method both have errors on the order of 2%. It is somewhat strange that the DDA method is not more exact since it gives very accurate values for the scattered intensities.

In Figure 2 the G-RDG method is compared with the RDG-method, the DDA method and the T-matrix method. Again the results from the T-matrix method are assumed to be correct. The aggregate consists of $N$ non-overlapping spheres with radius 10 nm and with $k_f = 2.3$ and $D_f = 1.5$. At 1064 nm the G-RDG method is very accurate but as the wavelength decreases the error increases. At the wavelength

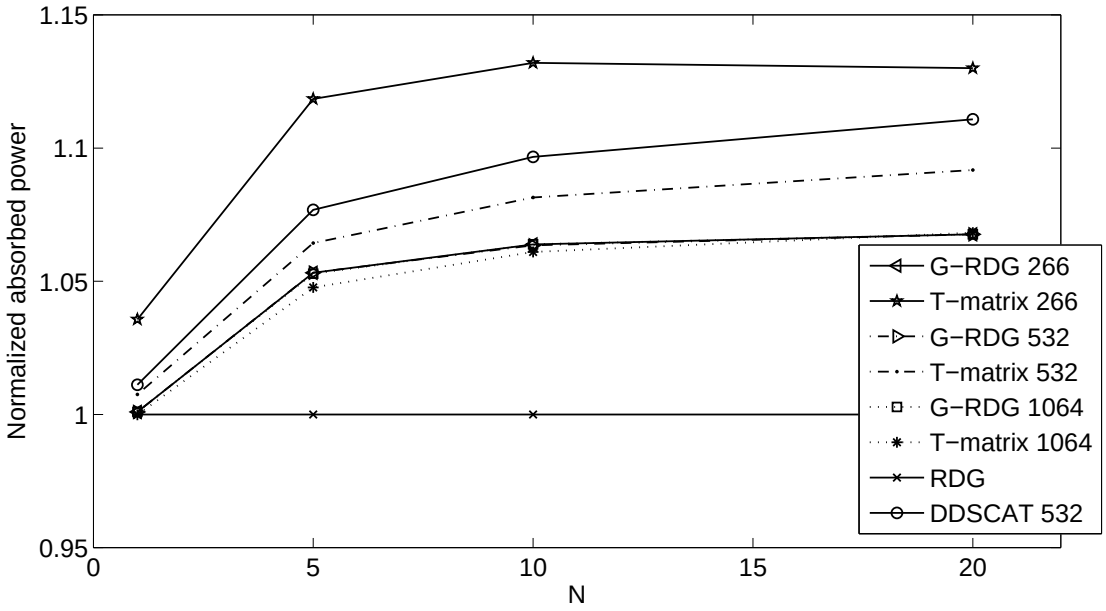


**Figure 2**: A comparison between the averaged absorbed power for the RDG (Rayleigh) method, the G-RDG method, the DDA method and the T-matrix method for an ensemble of identical aggregates, with random orientation. Each aggregate consists of $N$ non-overlapping spheres with radius 10 nm, $k_f = 2.3$ and $D_f = 1.5$. The wavelengths are 1064 nm, 532 nm and 266 nm. All of the results are normalized with the absorbed power obtained by the RDG method. The result for DDA is only shown for 532 nm. The results from the T-matrix method are considered to be very accurate. Notice that the three curves for G-RDG almost coincide.

532 nm the error is on the order of 3%, which in most cases can be accepted. At 266 nm the error is on the order of 5%. The curve of G-RDG levels out and is almost constant for $N > 10$. This is the same behavior that was noticed in [11] for the electrostatic approximation (ESA) method [14]. In fact, ESA and G-RGD are very similar for non-overlapping spherical primary particles. As mentioned before, the drawback with ESA is that it can only be applied to non-overlapping spheres. The RDG approximation gives a quite large error even at 1064 nm, which in accordance with the observations that it underestimates the absorbed power, *cf.*, [11] and [14]. The DDA method is very time consuming and is for this reason only shown for 532 nm. It is then slightly more accurate than the G-RDG method.

In Figure 3 the G-RDG method is compared with the RDG method and the DDA method for the absorbed power for an aggregate with overlapping spheres, see Figure 4. The radius of the spheres are 10 nm and the distance between two adjacent spheres is 13.3 nm. The wavelength is 532 nm. This case can not be analyzed by the T-matrix method since the spheres overlap. As seen above, the accuracy of the DDA method and the G-RDG method are roughly the same at 532 nm. For this case the error in the RDG-method is on the order of 15-20%, which in many cases

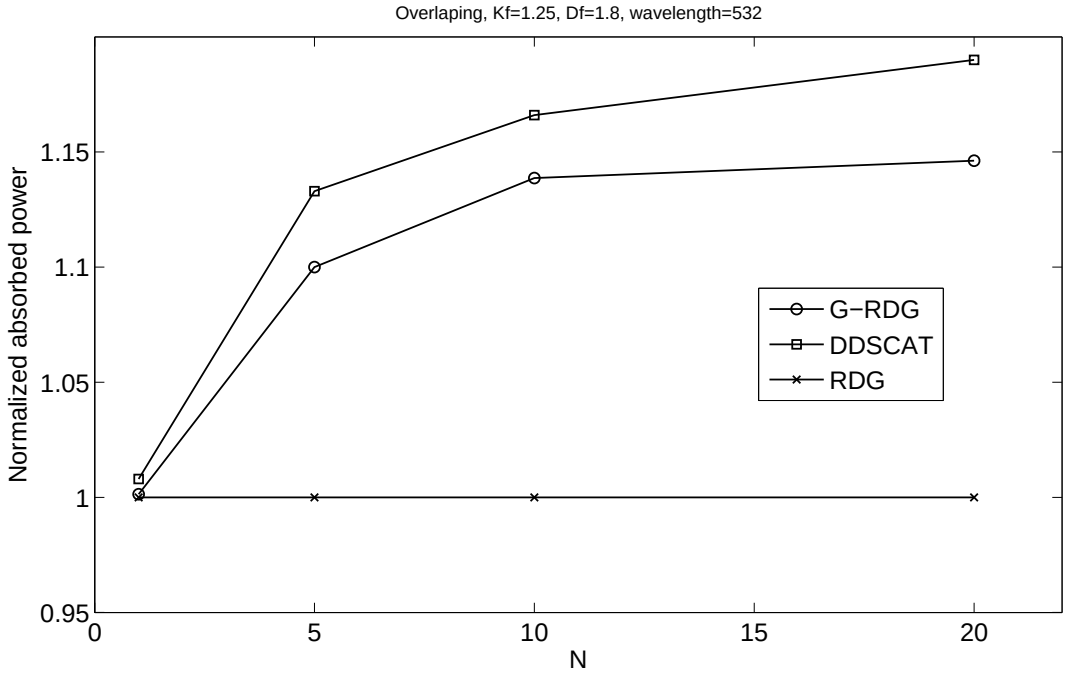


**Figure 3**: A comparison between the averaged absorbed power for the RDG (Rayleigh) method, the G-RDG method and the DDA method for an ensemble of identical aggregates, with random orientation. Each aggregate consists of $N$ overlapping spheres with radius 10 nm, $k_f = 1.25$ and $D_f = 1.8$, *cf.*, Figure 4. The distance between the centers of two adjacent spheres is 13.3 nm. The wavelength is 532 nm. All of the results are normalized with the absorbed power obtained by the RDG method.

cannot be accepted. The large error is due to that the polarization vector in the RDG method is the one obtained from a single sphere in a constant electric field, and that differs from the polarization of overlapping spheres.

Two examples of averaged scattered intensities are presented in Figures 5 and 6. The intensities are the total intensity in the plane perpendicular to the direction of the electric field of the incident wave, *i.e.*, for $\alpha = \pi/2$, see Eq. (2.5). The angle on the horizontal axis figures is the angle measured from the direction of incidence. The intensities are normalized with the intensity at $\theta = 0°$, *i.e.*, the intensity in the forward direction. In the case of non-overlapping spheres, Figure 5, it is seen that G-RDG, DDA and the T-matrix method give the same results for an aggregate of 20 spheres. In the case of overlapping spheres, Figure 6, the T-matrix cannot be used, but it is assumed that the result for DDA is very accurate. As seen from the graph the G-RDG method is also very accurate, even at 532 nm.

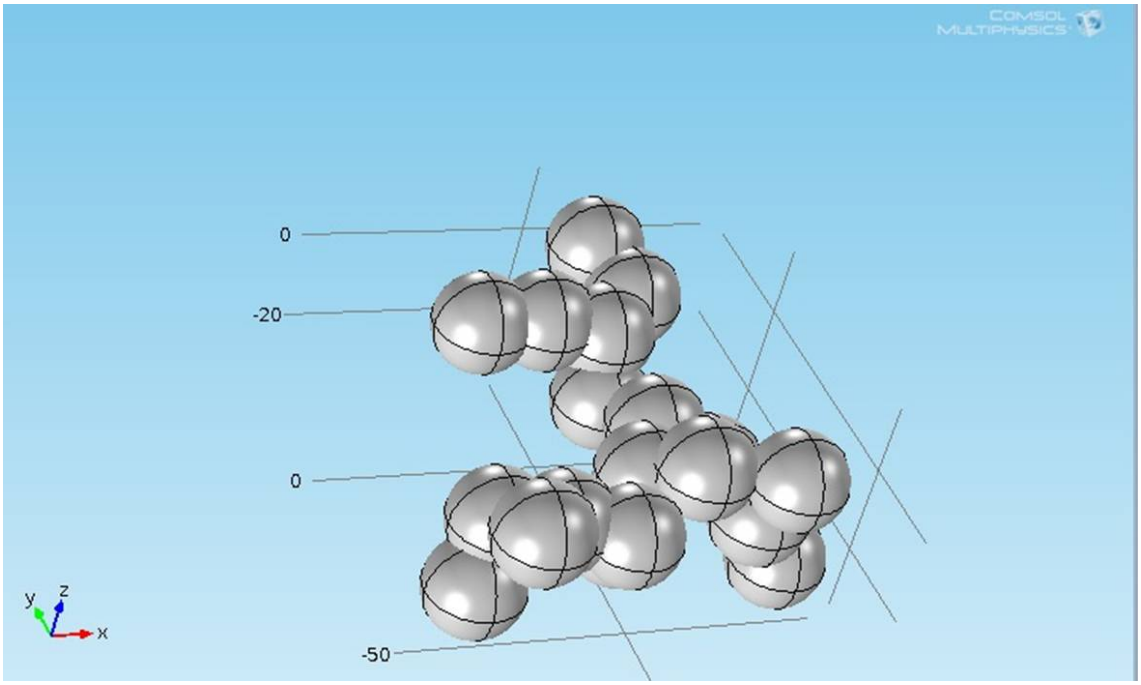


**Figure 4**: The aggregate with 20 overlapping spheres used in Figures 3 and 6. The radius of the spheres is 10 nm, the distance between the centers of two adjacent spheres is 13.3 nm, $k_f = 1.25$ and $D_f = 1.8$.

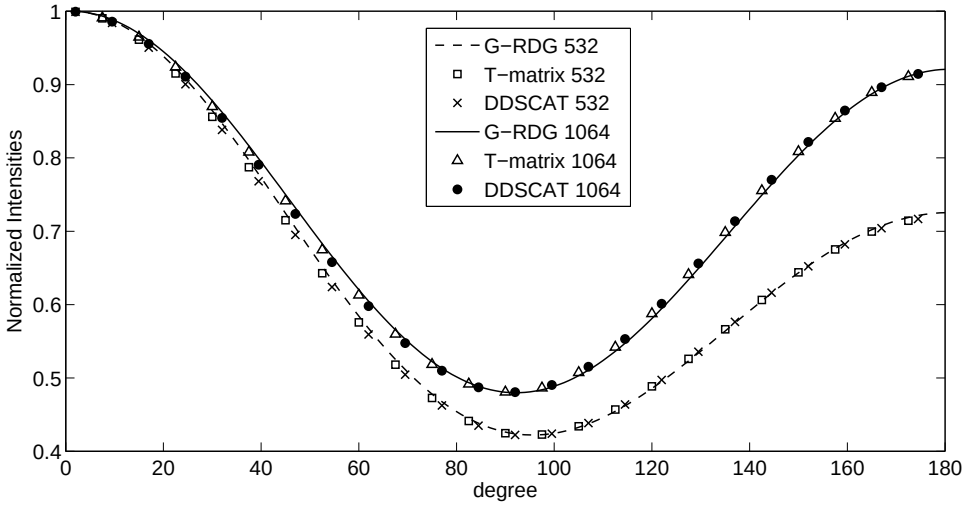


**Figure 5**: The averaged normalized intensity for an an ensemble of identical aggregates, with random orientation. Each aggregate consists 20 non-overlapping spheres with $k_f = 2.3$ and $D_f = 1.5$. The wavelengths are 532 nm and 1064 nm. The scattering plane is perpendicular to $\hat{\boldsymbol{e}}_i$, the angle is the polar angle $\theta$ between $\hat{\boldsymbol{k}}_i$ and $\hat{\boldsymbol{r}}$.

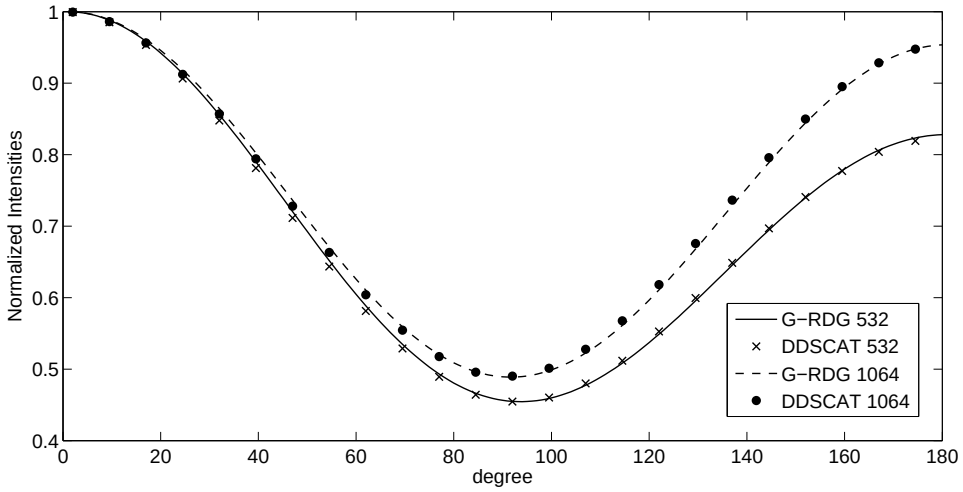


**Figure 6**: The averaged normalized intensity for an an ensemble of identical aggregates, with random orientation. Each aggregate consists 20 overlapping spheres with $k_f = 1.25$ and $D_f = 1.8$. The wavelengths are 532 nm and 1064 nm, the radius of the spheres is 10 nm and the distance between the centers of two adjacent spheres is 13.3 nm. The scattering plane is perpendicular to $\hat{\boldsymbol{e}}_i$, the angle is the polar angle $\theta$ between $\hat{\boldsymbol{k}}_i$ and $\hat{\boldsymbol{r}}$.

### 6.1 Run-time

The run-time for the different methods differs a lot when it comes to averaging over an ensemble of aggregates. The G-RDG is approximatley 40 times faster than DDA, and the T-matrix method is roughly 200 times faster than DDA. The reason that DDA is time consuming is that it needs to redo the entire calculation for each rotation of the aggregate, and at least 125 different angles are needed in the averaging. The T-matrix method calculates the T-matrix for the entire aggregate and this matrix is indepandent of the incident field. The averaging can then be done analyticaly and leads to very fast calculations.

## 7 Conclusions

There are a number of different methods that can be applied to scattering and absorption from aggregates and a large number of papers are devoted to these problems, *cf.*, the reference list in [18], [12] and [8]. In this paper the T-matrix method, the discrete dipole approximation method and the RDG method are compared with the G-RDG method. The T-matrix method is fast and can handle large aggregates, but is restricted to aggregates made of non-overlapping spheres, which is a severe restriction. The discrete dipole approximation can handle any type of aggregate but is very slow when it is applied to ensembles of aggregates. The approximate methods have their advantages of being fast and easy too implement. Thus the RDG

method, is often used since the scattered intensities and absorbed powers are very easy to calculate. The drawback is that the errors already at quite long wavelengths can be unacceptably large. The method presented in this paper is a generalization of the RDG method, and in a sense also a generalization of the ESA method [14]. It provides an easy and very fast way to calculate the absorbed power and scattered intensity from ensembles of aggregates. Thus one can use the finite element method and apply an external constant field in three perpendicular directions to obtain the scattered intensity and the absorbed power for an ensemble of identical, but randomly oriented aggregates. The numerical examples in this paper were obtained using the commercial FEM-program Comsol. One can use other methods like integral equation methods, or even the quasi-static discrete dipole approximation, to obtain the same results. The main advantage is that the method leads to simple closed form expressions for the averages of absorbed power and scattered intensity. The expressions have simple physical interpretations, which is of interest in the inverse problem where the parameters describing the aggregates are to be determined from scattering experiments.

# Appendix A

The averages of the scattered intensity and absorption of an aggregate are derived in this appendix. To do the averaging, the aggregate is kept in a fixed position in an $xyz$-coordinate system and the direction of propagation, $\hat{\boldsymbol{k}}_i$, is altered over all directions and the direction of the electric field, $\hat{\boldsymbol{e}}_i$, is altered over the angular interval $[0, 2\pi]$. This corresponds to a rotation of the aggregate over all Euler angles. Before one can take the average one needs to express the three vectors $(\hat{\boldsymbol{e}}_i, \hat{\boldsymbol{f}}, \hat{\boldsymbol{k}}_i)$, *cf.*, Eq. (2.5) in the cartesian unit vectors $(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{\boldsymbol{z}})$. This is done by introducing the spherical angles $\theta, \phi$ and the angle $\psi$ for the direction of the electric field.

The wave vector of the incident plane wave is given by

$$\boldsymbol{k}_i = k\hat{\boldsymbol{k}}_i = k(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta) \tag{A.1}$$

where $\theta$ is the polar angle from the $z$-axis and $\phi$ is the azimuthal angle from the $x-$axis. The direction $\hat{\boldsymbol{e}}_i$ of the electric field of the incident wave is given by

$$\hat{\boldsymbol{e}}_i = (e_x, e_y, e_z) = \hat{\boldsymbol{\xi}}\cos\psi + \hat{\boldsymbol{\eta}}\sin\psi \tag{A.2}$$

The unit vectors $\hat{\boldsymbol{\xi}}$ and $\hat{\boldsymbol{\eta}}$ are given by

$$\begin{aligned} \hat{\boldsymbol{\eta}} &= \frac{\hat{\boldsymbol{k}}_i \times \hat{\boldsymbol{z}}}{|\hat{\boldsymbol{k}}_i \times \hat{\boldsymbol{z}}|} \\ \hat{\boldsymbol{\xi}} &= \hat{\boldsymbol{\eta}} \times \hat{\boldsymbol{k}}_i \end{aligned} \tag{A.3}$$

## A.1 Derivation of the intensities in Eqs. (5.1) and (5.2)

The normalized scattered intensity is given by

$$I(\boldsymbol{r}) = \frac{|\boldsymbol{E}^s(\boldsymbol{r})|^2}{E_0^2} = \frac{1}{E_0^2}\sum_{m=1}^{M}|\boldsymbol{E}_m^s(\boldsymbol{r})|^2 \tag{A.4}$$

where $\boldsymbol{E}_m^s$ is the scattered electric far-field from aggregate number $m$. All other terms in the right hand side of Eq. (2.4) sum up to zero.

Since all of the aggregates are identical, one can obtain the intensity by altering the directions $\hat{\boldsymbol{k}}_i$ and $\hat{\boldsymbol{e}}_i$ over all angles $\theta$, $\phi$ and $\psi$ for one of the aggregates, *i.e.*,

$$I(\boldsymbol{r}) = M < I_1(\boldsymbol{r}) >= \frac{M}{E_0^2 8\pi^2}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}|\boldsymbol{E}_1^s(\boldsymbol{r})|^2\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi \tag{A.5}$$

The scattered field is expressed in terms of the dipole moments by utilizing Eqs. (4.1) and (3.7)

$$\boldsymbol{E}_1^s(\boldsymbol{r}) = -\frac{k^2 e^{\mathrm{i}kr}}{4\pi\varepsilon_0 r}\hat{\boldsymbol{r}}\times\left(\hat{\boldsymbol{r}}\times\left(\sum_{n=1}^{N_A}(\tilde{\boldsymbol{p}}_n+\Delta\boldsymbol{p}_n)e^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_n}\right)\right) \tag{A.6}$$

The average of $\Delta\boldsymbol{p}_n$ for the ensemble is zero and it is plausible that also the average of $\Delta\boldsymbol{p}_n(\boldsymbol{r})e^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_n}$ is negligible. Also it is assumed that the average of $\Delta\boldsymbol{p}_n\cdot\Delta\boldsymbol{p}_{n'}^* e^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_n-\boldsymbol{r}_{n'})}$ is negligible when $n\neq n'$. It is seen that

$$\hat{\boldsymbol{r}}\times(\hat{\boldsymbol{r}}\times\tilde{\boldsymbol{p}}_n)\cdot\hat{\boldsymbol{r}}\times(\hat{\boldsymbol{r}}\times\tilde{\boldsymbol{p}}_{n'}^*) = \frac{1}{9}\sin^2(\alpha)\mathrm{tr}\{\tilde{\mathbf{p}}_n\}\mathrm{tr}\{\tilde{\mathbf{p}}_{n'}^*\}$$

where $\alpha$ is the constant angle between $\hat{\boldsymbol{r}}$ and $\hat{\boldsymbol{e}}_i$, *cf.*, Eqs. (2.5) and (2.6). The intensity can be approximated by the following expression

$$\begin{aligned} I(\boldsymbol{r}) =& M < I_1(\boldsymbol{r}) >= \frac{2Mk^4}{(4\pi)^4\varepsilon_0^2 r^2}\left[\frac{1}{9}\sin^2(\alpha)\sum_{n=1}^{N_A}\sum_{n'=1}^{N_A}\mathrm{tr}\{\tilde{\mathbf{p}}_n\}\mathrm{tr}\{\tilde{\mathbf{p}}_{n'}^*\}\right. \\ &\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}e^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_n-\boldsymbol{r}_{n'})}\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi \\ &\left.+\sum_{n=1}^{N_A}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}|\hat{\boldsymbol{r}}\times(\hat{\boldsymbol{r}}\times\Delta\boldsymbol{p}_n)|^2\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi\right] \end{aligned}$$

When the intensity is split up in its co and cross polarizations this leads to

$$I_{\mathrm{co}}(\boldsymbol{r}) = \frac{2Mk^4}{(4\pi)^4\varepsilon_0^2 r^2}\left[\frac{\sin^2\alpha}{9}\sum_{n=1}^{N_A}\sum_{n'=1}^{N_A}\mathrm{tr}\{\tilde{\mathbf{p}}_n\}\mathrm{tr}\{\tilde{\mathbf{p}}_{n'}^*\}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi} e^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_n-\boldsymbol{r}_{n'})}\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi\right.$$
$$\left.+\sum_{n=1}^{N_A}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}|\hat{\boldsymbol{\alpha}}\cdot\Delta\boldsymbol{p}_n|^2\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi\right]$$
$$I_{\mathrm{cross}}(\boldsymbol{r}) = \frac{2Mk^4}{(4\pi)^4\varepsilon_0^2 r^2}\sum_{n=1}^{N_A}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}|\hat{\boldsymbol{\beta}}\cdot\Delta\boldsymbol{p}_n|^2\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi$$

One can simplify this further. Since $\boldsymbol{q}$ has a constant length $|\boldsymbol{q}| = k\sqrt{2(1-\hat{\boldsymbol{r}}\cdot\hat{\boldsymbol{k}}_i)}$ and should cover all directions one may write $\boldsymbol{q} = |\boldsymbol{q}|(\sin\bar{\theta}\cos\bar{\phi}, \sin\bar{\theta}\sin\bar{\phi}, \cos\bar{\theta})$ and then

$$\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi} e^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_n-\boldsymbol{r}_{n'})}\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi = 2\pi\int_0^{2\pi}\int_0^{\pi} e^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_n-\boldsymbol{r}_{n'})}\sin\bar{\theta}\,\mathrm{d}\bar{\theta}\,\mathrm{d}\bar{\phi}$$

The identities

$$\int_0^{2\pi} e^{\mathrm{i}A\cos(\bar{\phi}+\phi_0)}\,d\bar{\phi} = 2\pi J_0(A)$$

and

$$\boldsymbol{q}\cdot(\boldsymbol{r}_n-\boldsymbol{r}_{n'}) = |\boldsymbol{q}|\left(\rho_{nn'}\sin\bar{\theta}\cos\left(\bar{\phi}-\arctan\left(\frac{y_n}{x_n}\right)\right)+(z_n-z_{n'})\cos\bar{\theta}\right)$$

where $\rho_{nn'} = \sqrt{(x_n-x_{n'})^2+(y_n-y_{n'})^2}$, can now be utilized. This leads to

$$\begin{aligned}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi} e^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{r}_n-\boldsymbol{r}_{n'})}\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi &= 4\pi^2\int_0^{\pi} J_0(|\boldsymbol{q}|\rho_{nn'}\sin\bar{\theta})e^{\mathrm{i}|\boldsymbol{q}|\cos\bar{\theta}(z_n-z_{n'})}\sin\bar{\theta}\,\mathrm{d}\bar{\theta}\\ &= 8\pi^2\int_0^{1} J_0(|\boldsymbol{q}|\rho_{nn'}\sqrt{1-x^2})\cos(|\boldsymbol{q}|x(z_n-z_{n'}))\,\mathrm{d}x\end{aligned} \tag{A.7}$$

Also the last integral can be solved analytically. To do this one may first realize that the integral must be independent of the orientation of the coordinate system. Thus in a case where $x_n = x_{n'} = y_n = y_{n'} = 0$ one must have that

$$\int_0^1\cos(|\boldsymbol{q}|x(z_n-z_{n'}))\,\mathrm{d}x = \int_0^1 J_0(|\boldsymbol{q}||z_n-z_{n'}|\sqrt{1-x^2})\,\mathrm{d}x = \frac{\sin(|\boldsymbol{q}||z_n-z_{n'}|)}{|\boldsymbol{q}||z_n-z_{n'}|} \tag{A.8}$$

Then consider the general case. One may rotate the coordinate system such that $z_n = z_{n'}$ and then $\rho_{nn'} = r_{nn'}$. The integral has to give the same value which means that the integral in Eq. (A.7) equals

$$\int_0^1 J_0(|\boldsymbol{q}|\rho_{nn'}\sqrt{1-x^2})\cos(|\boldsymbol{q}|x(z_n - z_{n'}))\,\mathrm{d}x = \int_0^1 J_0(|\boldsymbol{q}|r_{nn'}\sqrt{1-x^2})\,\mathrm{d}x$$

From Eq. (A.8) one gets

$$\int_0^1 J_0(|\boldsymbol{q}|\rho_{nn'}\sqrt{1-x^2})\cos(|\boldsymbol{q}|x(z_n - z_{n'}))\,\mathrm{d}x = \frac{\sin(|\boldsymbol{q}|r_{nn'})}{|\boldsymbol{q}|r_{nn'}}$$

By using a symbolic manipulator the following expressions are obtained:

$$\begin{aligned}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi} |\hat{\boldsymbol{\alpha}}\cdot\Delta\boldsymbol{p}_n|^2 \sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi = \\ = \frac{8\pi^2}{15}\left((1+2\sin^2\alpha)\left(|\Delta \mathrm{p}_{n11}|^2 + |\Delta \mathrm{p}_{n22}|^2 + |\Delta \mathrm{p}_{n33}|^2\right)\right. \\ -(1-3\sin^2\alpha)\mathrm{Re}\left\{\Delta \mathrm{p}_{n11}\Delta \mathrm{p}^*_{n22} + \Delta \mathrm{p}_{n11}\Delta \mathrm{p}^*_{n33} + \Delta \mathrm{p}_{n22}\Delta \mathrm{p}^*_{n33}\right\} \\ \left.+(3+\sin^2\alpha)\left(|\Delta \mathrm{p}_{n12}|^2 + |\Delta \mathrm{p}_{n13}|^2 + |\Delta \mathrm{p}^2_{n23}|\right)\right)\end{aligned}$$

and

$$\begin{aligned}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi} |\hat{\boldsymbol{\beta}}\cdot\Delta\boldsymbol{p}_n|^2 \sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi = \frac{8\pi^2}{15}\left(|\Delta \mathrm{p}_{n11}|^2 + |\Delta \mathrm{p}_{n22}|^2 + |\Delta \mathrm{p}_{n33}|^2\right. \\ -\mathrm{Re}\left\{\Delta \mathrm{p}_{n11}\Delta \mathrm{p}^*_{n22} + \Delta \mathrm{p}_{n11}\Delta \mathrm{p}^*_{n33} + \Delta \mathrm{p}_{n22}\Delta \mathrm{p}^*_{n33}\right\} \\ \left.+3\left(\Delta \mathrm{p}^2_{n12} + \Delta \mathrm{p}^2_{n13} + \Delta \mathrm{p}^2_{n23}\right)\right)\end{aligned}$$

However, since $\Delta \mathrm{p}_{11} + \Delta \mathrm{p}_{22} + \Delta \mathrm{p}_{33} = 0$ then
$\mathrm{Re}\left\{\Delta \mathrm{p}_{n11}\Delta \mathrm{p}^*_{n22} + \Delta \mathrm{p}_{n11}\Delta \mathrm{p}^*_{n33} + \Delta \mathrm{p}_{n22}\Delta \mathrm{p}^*_{n33}\right\} = -0.5(|\Delta \mathrm{p}_{11}|^2 + |\Delta \mathrm{p}_{22}|^2 + |\Delta \mathrm{p}_{33}|^2)$.
This leads to Eqs. (5.1) and (5.2), *i.e.*,

$$\begin{aligned}I_{\mathrm{co}}(\boldsymbol{r}) = \frac{Mk^4}{32\pi^2\varepsilon_0^2 r^2}\sum_{n=1}^{N_A}\left[\frac{2}{9}\sin^2(\alpha)\sum_{n'=1}^{N_A}\left(\mathrm{tr}\{\tilde{\mathrm{p}}_n\}\mathrm{tr}\{\tilde{\mathrm{p}}^*_{n'}\}\frac{\sin(|\boldsymbol{q}|r_{nn'})}{|\boldsymbol{q}|r_{nn'}}\right)\right. \\ \left.+\frac{3+\sin^2\alpha}{15}\mathrm{tr}\{\Delta\mathrm{p}_n\Delta\mathrm{p}^*_n\}\right]\end{aligned} \tag{A.9}$$

and

$$I_{\mathrm{cross}}(\boldsymbol{r}) = \frac{Mk^4}{160\pi^2\varepsilon_0^2 r^2}\sum_{n=1}^{N_A}\mathrm{tr}\{\Delta\mathrm{p}_n\Delta\mathrm{p}^*_n\} \tag{A.10}$$

If one cannot decompose the volume into sub-particles with dipole matrices, then one can obtain the intensities by integrations, as

$$I_{\mathrm{co}}(\boldsymbol{r}) = \frac{Mk^4}{32\pi^2\varepsilon_0^2 r^2}\left[\frac{2}{9}\sin^2(\alpha)\int_{V_a}\int_{V_a}\left(\mathrm{tr}\{\tilde{\mathbf{P}}(\boldsymbol{r})\}\mathrm{tr}\{\tilde{\mathbf{P}}(\boldsymbol{r}')^*\}\frac{\sin(|\boldsymbol{q}||\boldsymbol{r}-\boldsymbol{r}'|)}{|\boldsymbol{q}||\boldsymbol{r}-\boldsymbol{r}'|}\right)\mathrm{d}v\,\mathrm{d}v' + \frac{3+\sin^2\alpha}{15}\int_{V_a}\mathrm{tr}\{\Delta\mathbf{P}(\boldsymbol{r})\Delta\mathbf{P}(\boldsymbol{r})^*\}\,\mathrm{d}v\right] \tag{A.11}$$

and

$$I_{\mathrm{cross}}(\boldsymbol{r}) = \frac{Mk^4}{160\pi^2\varepsilon_0^2 r^2}\int_{V_a}\mathrm{tr}\{\Delta\mathbf{P}(\boldsymbol{r})\Delta\mathbf{P}^*(\boldsymbol{r})\}\,\mathrm{d}v \tag{A.12}$$

## A.2 Derivation of the absorption in Eq. (5.3)

The absorbed power is the sum of the absorbed powers of the aggregates in the ensemble. Also here one can get this sum by averaging as, *cf.*, Eq. (2.7)

$$\begin{aligned} W_a &= \frac{M}{(4\pi)^2}\frac{\sigma}{|\varepsilon_0(\varepsilon_c-1)|^2}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}\int_{V_a}|\boldsymbol{P}_1(\boldsymbol{r})|^2\,dV\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi \\ &= \frac{M\sigma}{2|\varepsilon_0(\varepsilon_c-1)|^2}\left(\int_{V_a}|\tilde{\boldsymbol{P}}(\boldsymbol{r})|^2\,dV + \frac{1}{8\pi^2}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}\int_{V_a}|\Delta\boldsymbol{P}(\boldsymbol{r})|^2\,dV\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi\right) \end{aligned} \tag{A.13}$$

The integrals over the angles $\theta$, $\phi$ and $\psi$ can be solved analytically. The results are

$$\begin{aligned} W_a &= \frac{M}{(4\pi)^2}\frac{\sigma}{|\varepsilon_0(\varepsilon_c-1)|^2}\int_0^{2\pi}\int_0^{2\pi}\int_0^{\pi}\int_{V_a}|\boldsymbol{P}_1(\boldsymbol{r})|^2\,dV\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\psi \\ &= \frac{M\sigma E_0^2}{6|\varepsilon_0(\varepsilon_c-1)|^2}\int_{V_a}\mathrm{tr}\{\mathbf{P}(\boldsymbol{r})\mathbf{P}^*(\boldsymbol{r})\}dV \end{aligned}$$

One may notice that $\mathrm{tr}\{\mathbf{P}(\boldsymbol{r})\mathbf{P}^*(\boldsymbol{r})\} = \mathrm{tr}\{\tilde{\mathbf{P}}(\boldsymbol{r})\tilde{\mathbf{P}}^*(\boldsymbol{r})\} + \mathrm{tr}\{\Delta\mathbf{P}(\boldsymbol{r})\Delta\mathbf{P}^*(\boldsymbol{r})\}$, but the latter expression does not simplify the numerical calculations.

## A.3 A comparison with the RDG approximation

In the RDG method all primary particles are considered to be identical spheres with the same dipole moments as a single sphere. By introducing approximations to Eqs (5.1), (5.2) and (5.3) one can get expressions that resemble the expressions for intensities and absorption for the RDG approximation. It is then assumed that all of the primary particles have approximately the same volume, $V_{sn}$, and the same dipole moment. One then introduces $\boldsymbol{p}_{\mathrm{av}} = \frac{1}{N_A}\sum_{n=1}^{N_A}\tilde{\boldsymbol{p}}_n$ and use the approximation

$\boldsymbol{p}_n \approx \boldsymbol{p}_{\mathrm{av}}$. The RDG approximation neglects the cross polarization and thus the intensity is the co polarization intensity given by

$$\begin{aligned} I(\boldsymbol{r}) = I_{\mathrm{co}}(\boldsymbol{r}) &= \frac{N_A^2 M k^4}{(4\pi)^3 E_0^2 \varepsilon_0^2 r^2} (|\tilde{\boldsymbol{p}}_{\mathrm{av}}| \sin\alpha)^2 \int_0^{2\pi} \int_0^{\pi} S(\bar{\theta}, \bar{\phi}) \sin\bar{\theta} \, \mathrm{d}\bar{\theta} \, \mathrm{d}\bar{\phi} \\ &= \frac{M k^4 (|\tilde{\boldsymbol{p}}_{\mathrm{av}}| \sin\alpha)^2}{(4\pi)^2 E_0^2 \varepsilon_0^2 r^2} \sum_{n=1}^{N_A} \sum_{n'=1}^{N_A} \frac{\sin(|\boldsymbol{q}| r_{nn'})}{|\boldsymbol{q}| r_{nn'}} \end{aligned} \tag{A.14}$$

where $S(\bar{\theta}, \bar{\phi})$ is the structure factor

$$S(\bar{\theta}, \bar{\phi}) = \frac{1}{N_A^2} \left| \sum_{n=1}^{N_A} e^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_n} \right|^2 \tag{A.15}$$

One advantage with Eq. (A.14) is that it includes the structure factor and hence the theory described in the review article by Sorensen [18] can be applied. The approximation of the absorption reads

$$W_a = \frac{M N_{\mathrm{A}} \sigma}{2 V_{sn} |\varepsilon_0 (\varepsilon_c - 1)|^2} |\boldsymbol{p}_{\mathrm{av}}|^2 \tag{A.16}$$

where $V_{sn}$ is the volume of a primary particle. The drawback with Eqs. (A.15) and (A.16) is that they are quite inaccurate, even though they give better results than the sphere approximation used in the RDG method.

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