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Review of the null-field method with discrete sources

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Abstract

In this paper the state of the art of the Null-Field Method with Discrete Sources (NFM-DS) will be reviewed. The NFM-DS combines the advantages of the null-field method with the advantages of the method of discrete sources to overcome stability problems of the standard null-field methods encountered in computation of scattering by very elongated particles such as long finite fibres and very flat particles such as flat discs.

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1. Introduction

As there are light and particles almost everywhere light scattering by particles is an important subject not only in natural science but also in modern technologies. Problems of interest range from interstellar dust to red blood cells. Other applications are focused on aerosol particles, soot aggregates, colour pigments and asbestos fibres. To characterize such particles light scattering theories and corresponding computational programs are needed. A number of light scattering theories have been developed, all having their pros and cons. An extensive overview of available theories has recently been published by Kahnert [1].

The T-matrix method or null-field method is one of the most well-known light scattering theories to compute scattering by nonspherical particles. Recent reviews of the literature on this method have been published by Mishchenko et al. [2,3]. The advantage of this method is that the T-matrix is computed, which includes the full solution of the scattering problem. This matrix relates the expansion coefficients of the incident field to the expansion coefficients of the scattered field. By using a precomputed T-matrix, scattering by a rotated or a translated particle or orientation averaged scattering can easily be computed. Additionally multiple scattering or scattering by a particle located near a plane interface can be computed from a stored T-matrix of this particle.

The standard T-matrix method is restricted to particles having an aspect ratio not larger than about 1:4 [4]. To solve the stability problems with particles having a larger aspect ratio expansion of the internal field using discrete sources was introduced into the null-field method.

In retrospect we started to develop the Null-Field Method with Discrete Sources (NFM-DS) 10 years ago. So it is time for a review of the state of the art of this method and to have a close look at the capabilities of the method and at problems still being unresolved. In the first place we did not intend to develop a new concept in

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light scattering theory. Our original interest was to solve some practical problems of the standard T-matrix method especially with particles having a high aspect ratio such as fibres and flat discs. For this reason in the first papers we used diverse names for our new concept of using discrete sources in the null-field method such as Multiple Multipole Extended Boundary Condition Method [5] and Extended Boundary Condition Method with Multipole Sources [6]. Later we decided for the name Null-Field Method with Discrete Sources (NFM-DS) [7]. Every new concept should have a single name.

In this paper the development of the NFM-DS will be reviewed and some exemplary scattering results will be presented to demonstrate the capabilities of the concept. Recent developments will also be mentioned. For a full description of the theory we refer the interested reader to the recently published monograph by Doicu et al. [8] which also includes FORTRAN90 programs on CD-ROM.

2. Null-field method with discrete sources

For internal field expansion the NFM-DS makes use of concepts developed in the framework of the Discrete Sources Method (DSM) [9] and related methods. The main idea of the DSM consists in approximating the solution of the problem by a linear combination of discrete sources. These discrete sources are the fundamental solution of the differential equation of the problem. The introduction of DSM is generally attributed to Kupradze and Aleksidze [10].

There are a variety of methods which use “equivalent sources” for field expansion. The “equivalent sources” may be of any type as long as they are solutions of the wave equation. Spherical waves, dipoles, Mie potentials and Gabor functions have been applied for field expansion.

Therefore there are different names for similar concepts like Method of Fundamental Solutions (MFS), Multiple Multipole method (MMP) [11], DSM, Method of Auxiliary Sources (MAS) [12], Method of Fictitious Sources (MFS) [13] or Yasuura method [14] and the Generalized Multipole Technique (GMT) [15].

A review of these methods has been published in an edited volume by Wriedt [16] and more recently by Fairweather [17]. A comprehensive presentation of the DSM is given by Doicu et al. [18]. For an analytic foundation of the null field method and the DSM in acoustics and electromagnetics the reader is referred to this book.

In the NFM-DS the following systems of functions have been implemented for field expansion: multiple multipoles [5], lowest order spherical vector wave functions [6] and electric dipoles and vector-Mie-potentials [19]. For rotational symmetric particles such as finite fibres the discrete sources are positioned on the axis of symmetry of the particle [20]. With oblate rotational symmetric scatterers it is of advantage to position the discrete sources in a complex plane [6]. In this way scattering by oblate discs [21], flat Cassini shaped particles [22] have been solved. Scattering results by particles having an aspect ratio as large as 100:1 can easily be computed for both extremely oblate and prolate particles.

3. Programs developed

Over the years a number of FORTRAN90 programs have been developed to compute the T-matrix of various scattering problems using the NFM-DS. These scattering problems include

- homogeneous, dielectric (isotropic, chiral) and perfectly conducting, axisymmetric particles;
- homogeneous, dielectric (isotropic, uniaxial anisotropic, chiral) and perfectly conducting, nonaxisymmetric particles;
- axisymmetric, composite particles;
- axisymmetric, layered particles;
- an inhomogeneous, dielectric, axisymmetric particle with an arbitrarily shaped inclusion;
- an inhomogeneous, dielectric sphere with a spherical inclusion;
- an inhomogeneous, dielectric sphere with an arbitrarily shaped inclusion;
- an inhomogeneous, dielectric sphere with multiple spherical inclusions;
- clusters of arbitrarily shaped particles;
- two homogeneous, dielectric spheres;

- clusters of homogeneous, dielectric spheres;
- concentrically layered spheres and
- a homogeneous, dielectric or perfectly conducting, axisymmetric particle on or near a plane surface.

The programs perform convergence tests and write the computed T-matrix to an output file. Using this stored T-matrix the scattering characteristics of a single particle or the scattering characteristics of an ensemble of polydisperse, homogeneous spherical particles can be computed. Additionally the effective wave number of a medium with randomly distributed spheroidal particles can be calculated. This FORTRAN90 programs which we developed over the years have been published recently on CD-ROM together with a monograph on the NFM-DS [8].

If the T-matrix is known, the scattering characteristics, describing the scattered field in the far-field region, can be readily computed. These include the far-field pattern, the differential scattering cross sections (DSCS), the amplitude matrix, the optical cross sections and the phase and extinction matrices. We consider a global coordinate system *OXYZ* with the origin inside the particle and denote by α_p, β_p and γ_p the particle orientation angles. For an incident wave travelling in the direction (β, α) we denote by

$$\mathbf{E}_s(\mathbf{r}) = \frac{e^{jk_s r}}{r}\left\{\mathbf{E}_{s\infty}(\mathbf{e}_r) + O\left(\frac{1}{r}\right)\right\}, \quad r \to \infty,$$

the scattered field in the direction (θ,φ), where $\mathbf{E}_{s\infty}(\mathbf{e}_r)$ is the far-field pattern and $\mathbf{e}_r = \mathbf{r}/r$ is the radial unit vector. Using the decomposition

$$\mathbf{E}_{s\infty} = E_{s\infty,\theta}\mathbf{e}_\theta + E_{s\infty,\varphi}\mathbf{e}_\varphi,$$

where $(\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_\varphi)$ are the spherical unit vectors of the scattering direction (θ, φ); we define the DSCS for parallel and perpendicular polarizations as $\sigma_{dp} = |E_{s\infty,\theta}|^2$ and $\sigma_{ds} = |E_{s\infty,\varphi}|^2$. The DSCS have the dimension of area, and dimensionless quantities are the normalized differential scattering cross sections $\sigma_{dpn} = |E_{s\infty,\theta}|^2/\pi a_c^2$ and $\sigma_{dsn} = |E_{s\infty,\varphi}|^2/\pi a_c^2$, where a_c is a characteristic dimension of the particle. In the following exemplary computational results are presented, therefore the normalized DSCS is plotted in the form of scattering diagrams.

4. Validation

In development of the FORTRAN90 programs which are based on the NFM-DS we heavily made use of program validation by comparing with results obtained from other programs or by comparing with other implementations of the same concept. These programs include the DSM, which is developed for related problems by the group of Eremin at Lomonosov Moscow State University [9]. Examples using DSM for validation have been published for the flat discs [22], concave Cassini oval based shapes [21], long fibres [23] and total internal reflection microscopy (TIRM) [24]. Evanescent wave scattering by a particle on a plane interface using both NFM-DS and DSM has been considered by Doicu et al. [25].

The T-matrix programs developed by Barber and Hill [26] and by Mishchenko [4] suitable for rotationally symmetric scattering particles also proved to be useful for the purpose of program validation. For particles lacking this kind of symmetry or more complex particles we also made use of the MMP by Hafner and Bomholt [11], the Discrete Dipole Approximation (DDA) [27], Finite Difference Time Domain (FDTD) [28], Volume Integral Equation Method (VIEM) [29], and the commercial computational electromagnetics program CST Microwave Studio [30] based on the Finite Integration Technique (FIT) [31]. Results for a dielectric cube comparing NFM-DS with DDA, FDTD and VIEM have been published by Wriedt and Comberg [32]. NFM-DS, DDA and MMP have been compared for a two particle multiple scattering problem by Comberg and Wriedt [33]. NFM-DS, MMP, DSM, DDA and FIT have all been applied to compute scattering by a red blood cell [34].

5. Flat discs

As already mentioned there is a problem with the standard T-matrix method to compute scattering by flat particle having a high aspect ratio. To improve the T-matrix method it helps to expand the internal field of the flat particle into a system of discrete sources positioned in a complex plane [22]. As an exemplary scattering result the DSCS is computed for a flat circular disc of diameter $d = 1000\,\mathrm{nm}$, thickness $t = 40\,\mathrm{nm}$, refractive index $n = 1.46$ and wavelength $\lambda = 532\,\mathrm{nm}$; the plane wave is incident normal to the flat surface and additionally at an angle of 65°. This flat cylindrical particle has an aspect ratio of 25. Using the standard T-matrix method it would not be possible to compute scattering by such a flat particle. The scattering results for both incident p- and s-polarizations are presented in Figs. 1 and 2. It is of advantage to deposition the discrete sources according to the shape of the scattering particle. This has been demonstrated by Hellmers and Wriedt [35] for the oblate, biconcave Cassini disc. If the discrete sources are positioned correctly along the axis in complex plane a lower number of discrete sources is needed and computation of scattering by even larger flat particle is possible.

6. Aggregated fibres

Computations of light scattering by fibrous particles are needed in various scientific branches such as astrophysics, atmospheric science and especially in optical particle characterization. In optical particle characterization there is interest to detect airborne fibrous particles like mineral, glass or asbestos fibres, which are considered to be serious health hazards. Here high aspect ratios are of special interest and so it is required that a light scattering simulation algorithm can handle them. Computational results of scattering by such fibres having a high aspect ratio have been published by Pulbere and Wriedt [20]. In this case discrete sources positioned on the axis of symmetry of the scattering fibre have been applied to cope with the high aspect ratio.

In this section we will introduce a new approach to compute scattering by aggregated fibres and we will present computational results for the first time. Using discrete sources we can compute scattering by a single fibre having a high aspect ratio. But to compute scattering by two long fibres which stick together in some way would be a multiple scattering problem and to solve this problem using a standard multiple scattering concept combining the T-matrices of both fibres would not be possible because the circumscribed spheres of both fibres would intersect. Also other computational approaches such as DDA, FDTD and related methods might be problematic because if the fibres are not aligned parallel to each other this would blow up the computational domain and increase computer time tremendously.

To overcome this problem we developed a multiple scattering method based on a decomposition of the primary fibres. Each fibre is decomposed into basic units having a size parameter of about $ka = 0.5$ and the

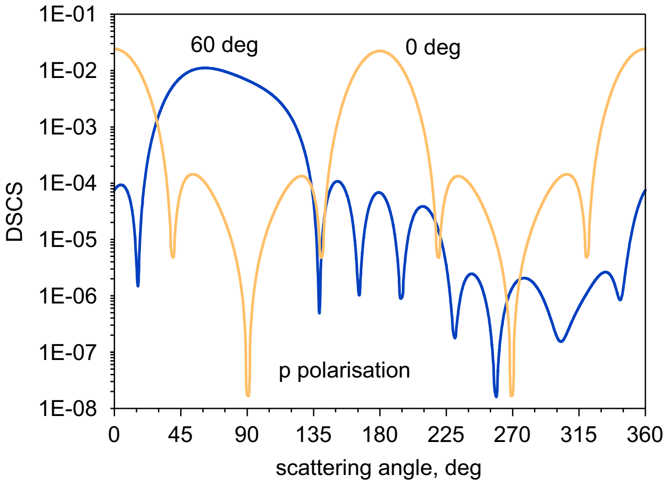

Fig. 1. Scattering diagram of a flat circular disc for incident p-polarization and two incident directions.

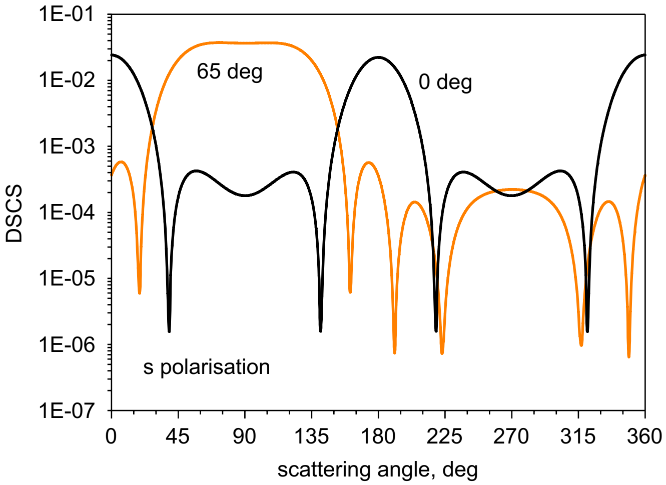

Fig. 2. Scattering diagram of a flat circular disc for incident s-polarization and two incident directions.

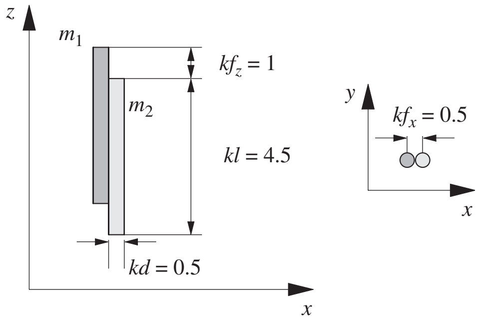

Fig. 3. Geometry of two parallel fibres. Parameters are given as size parameters. Refractive indices: $m_1 = 1.5, 1.3$.

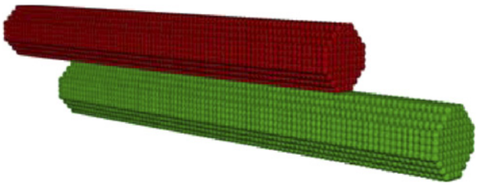

Fig. 4. The 3D view of the fibre geometry given in Fig. 3.

full scattering problem is solved by combining the T-matrices of all those units. In the following examples consisting of two fibres of circular section these basic units would be small sections of fibres. Each section is a short cylinder of circular cross section. Each of the fibres in the example is decomposed into nine short cylindrical sections. As an example, scatterings by two different fibre geometries have been computed. In the first one the fibres are parallel to each other. The geometry of this with all parameters given as size parameters is shown in Fig. 3. The plane wave is incident along the z-axis and the scattering diagram is plotted on the x–y plane both for incident p- and s-polarizations (Fig. 5). For validation DDA has also been used to compute scattering by these parallel fibres. A 3D view of the input fibre geometry used for DDA is shown in Fig. 4. As can be seen from the scattering diagram there is close agreement in the computational results (Fig. 5).

In the second example the fibres are aligned perpendicular to each other, see Fig. 6. A 3D view of the DDA fibre geometry is shown in Fig. 7. From the computed scattering diagram (Fig. 8) we see that there is also close

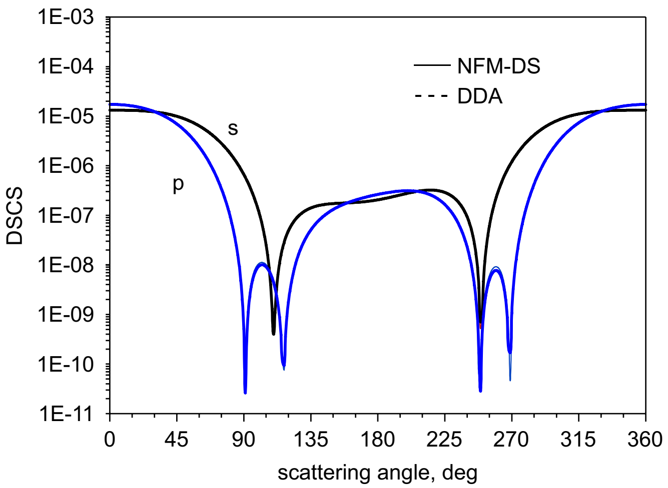

Fig. 5. Scattering diagram of two parallel fibres of geometry given in Fig. 3.

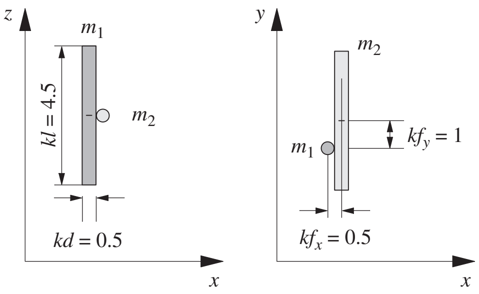

Fig. 6. Geometry of two perpendicular fibres. Parameters are given as size parameters. Refractive indices: $m_1 = 1.5, 1.3$.

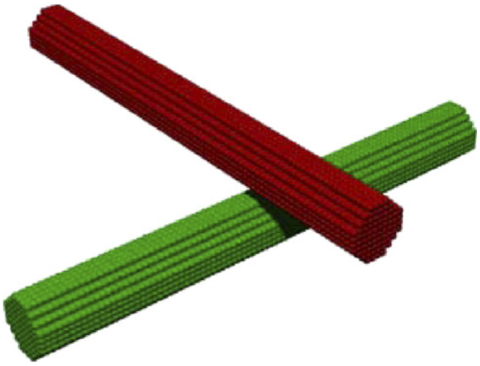

Fig. 7. The 3D view of fibre geometry given in Fig. 6.

agreement between the NFM-DS multiple scattering approach and DDA. Further results on this computational method will be published in near future.

7. Nonspherical, nonaxisymmetric particles

The NFM-DS has also been extended to compute the T-matrix of arbitrarily shaped particles which are nonaxisymmetric. This includes ellipsoids [19] and superellipsoids [36] as examples of such particle shapes. The

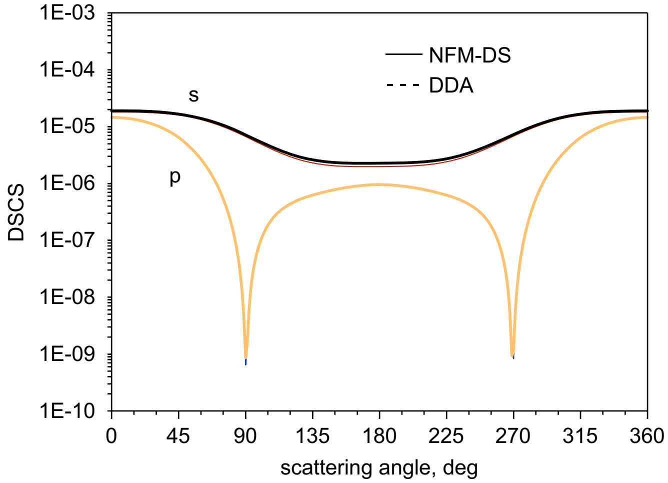

Fig. 8. Scattering diagram of two perpendicular fibres of geometry given in Fig. 6.

normal incident wave is a plane wave. But as in optical particle characterization lasers are commonly used, the Gaussian beam also has been implemented as an incident wave in the NFM-DS [37].

To be able to compute scattering by a particle of almost any shape we decided to use a modified centroid integration scheme to compute the surface integrals to be evaluated for the T-matrix of the scattering particle [38]. This integration scheme is based on a discretization of the particle surface into a number of small triangular surface patches. For a description of any particle surface we are using the Wavefront .obj computational graphics format. In this way computational graphics programs can be used for visualization, generation or modification of particle shapes. The surface integrals to be computed are approximated by

$$\int_S f \, \mathrm{d}S \approx \sum_i f(v_{i,c}) \, \mathrm{area}[v_{i,1}, v_{i,2}, v_{i,3}]. \tag{1}$$

Here, $v_{i,1}, v_{i,2}, v_{i,3}$ are the vertices spanning a triangle and point $v_{i,c}$ denotes the centre of mass of the triangle $[v_{i,1}, v_{i,2}, v_{i,3}]$ given by

$$v_{i,c} = \frac{1}{3} \sum_{j=1}^{3} v_{i,j}. \tag{2}$$

Thus the integral over each small triangle is approximated by multiplying the value of the integrand at the centroid by the triangle area. Of course, the particle shape has do be discretized into a sufficiently large number of small triangular surface patches to achieve convergent computational results.

As an exemplary application a Windows based program has been developed both to generate superellipsoids and to compute scattering by such particles [36]. The superellipsoid represents a family of 3D shapes by a product of two superquadratic curves [39]. This type of shape is well known in computer graphics and it can be used not only to model a wide range of shapes such as spheres, ellipsoids and cylinders but also cubes and cylinders with rounded edges [40]. For triangulation of a superellipsoid surface we make use of an algorithm developed and implemented into a C-program by Widmann [41] for the piecewise linear approximation of implicitly defined surfaces. This algorithm was originally developed for the numerical solution of integral equations on surfaces arising from the boundary integral method. Scattering results for superellipsoids can be found in a paper by Wriedt [36].

Here we would like to demonstrate scattering by an arbitrarily shaped particle using a Gaussian random shape sphere produced using the method developed by Muinonen et al. [42]. The original triangulation produced by this program is not really perfect. There is oversampling at the poles of the body. To improve the triangulation the Rational Reducer software has been used to modify the original Gaussian random sphere resulting in 4148 surface triangles. A 3D view of the particle geometry is shown in Fig. 9. The bounding box of this particle in (x, y, z) is

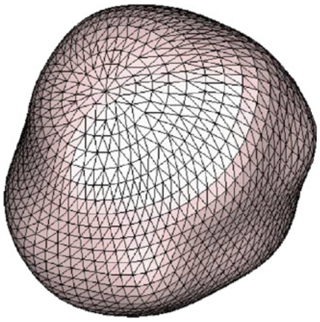

Fig. 9. The 3D view of a Gaussian random particle.

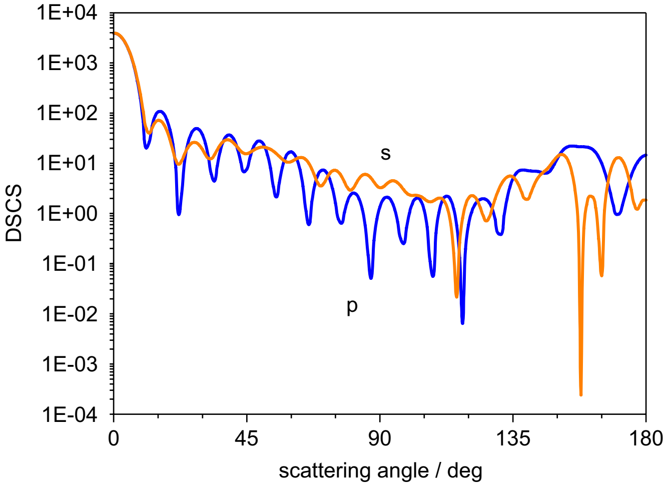

Fig. 10. Scattering diagram of a Gaussian random particle both for incident p- and s-polarizations.

(4.009, 3.612, 3.618 μm), its volume is 26.352 μm^3 and its surface area is 43.371 μm^2. For the interested reader the 3D shape model of this Gaussian random particle will be available on the website http://www.T-matrix.de for the purpose of computing validation results. To compute the scattering diagram an incident wave of $\lambda = 628.31$ propagating along the z-axis has been used. A convergent result has been achieved with $N = 31$, $M = 21$ as parameters of vector spherical wave expansion. The scattering diagram of the Gaussian random shaped particle is shown in Fig. 10 both for incident p- and s-polarizations. The influence of triangulation of a particle shape on the scattering result has been investigated by Hellmers and Wriedt [43]. It has been found that a certain number of surface triangles is needed for correct computation of the T-matrix and that a larger number of triangles will not spoil the computation result but just increase computational time.

8. Particles on a plane interface

Another topic of practical interest is light scattering by particles positioned on a plane surface. Due to fast progress and further miniaturization of nanostructures as parts of electronic devices there is an essential need in characterization of different kinds of defect particles using a particle surface scanner. Different numerical methods have been applied recently to model light scattering by particles on a surface, but some of them are not able to fully account for interactions between particle and a surface. Therefore the NFM-DS has been developed for particle on surface scattering fully taking into account particle surface scattering interaction without any kind of approximation. The theory is fully detailed by Wriedt et al. [44]. Doicu et al. [45] demonstrated that correct computational results can even be computed if the circumscribed sphere of a nonspherical particle on a plane surface does intersect the plane surface a bit.

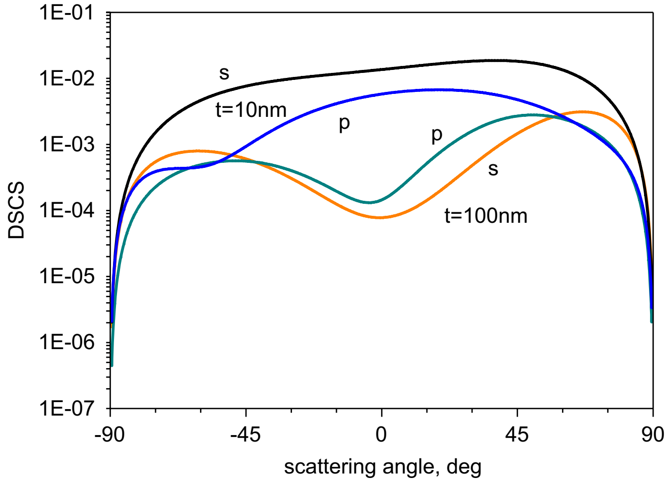

Fig. 11. Scattering diagram of a PSL sphere on a Si substrate with SiO_2 film for both incident p- and s-polarizations.

To calibrate optical particle scanners commonly PSL spheres are used. In the next example a PSL ($n = 1.6$) sphere ($d = 200\,\text{nm}$) is positioned on a SiO_2 film ($n = 1.46$) on a Si substrate ($n = 3.88 - i0.02$). A plane wave with wavelength $\lambda = 532\,\text{nm}$ is incident at 45° from the normal. The unpolarized DSCS is plotted both for incident p- and s-polarizations in Fig. 11. In this case the thickness of the SiO_2 film is varied and one can see that a minimum in the scattering pattern forms if the thickness of the film is increased.

Scattering of a particle positioned on a plane surface is also needed in other applications. Recently interest in nanoparticle of noble metals positioned on a plane interface increases, due to their ability to demonstrate a peak in the extinction spectrum known as localized surface plasmon resonance. There is a strong dependence of plasmon resonance on the size, the shape and the dielectric properties of the particle material. Due to this fact gold and silver nanoparticles are often used in biomedical and sensing devices. Modelling of metal particles is especially difficult due to complex refractive indices depending on wavelength.

8.1. Further applications

The programs developed have been used in a number of practical applications. This ranges from sizing of nonspherical particles using Phase Doppler Anemometry (PDA) [46], development of an asbestos fibre detection instrument [20], characterization of soot aggregates by a light scattering method [47], computing the calibration curve for TIRM [24] and a study for an erythrocyte characterization method [22].

As in optical particle characterization the incident wave is quite often a laser beam with a Gaussian intensity distribution the NFM-DS has also been extended to compute scattering by a Gaussian laser beam [37].

A more recent extension of the NFM-DS is scattering computation for particles on plane surfaces to develop efficient methods for particle surface scanners [44]. Evanescent wave scattering is also considered [25], which is of interest in biosensor development. As many scattering particles are not homogeneous or isotropic the theory has also been adapted to handle scattering by layered particles [48], composite [49], chiral [8] or anisotropic particles [50]. Further computational results especially for particles of complex shape and structure have recently been published by Wriedt [51]. There is interest to characterize surface roughness of solidifying molten metal particles. Hellmers and Wriedt used the NSM-DS to compute scattering of spherical metal particles with a different degree of surface roughness [52].

9. Conclusion

In this paper an overview of the progress in developing the NFM-DS has been given. Some open problems are still left. These include a bianisotropic refractive index of the scattering particle and of the film on which the particle is positioned or of the surrounding medium, nonaxisymmetric compound particles and optimal deposition of discrete sources and faces for surface integration. The full potential of the NFM-DS concept and the computational programs is still waiting to be exploited. There might be further interesting applications in optical particle characterization, surface defect characterization, nano-optics, near-field optics and optical sensing.

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