Accurate light scattering for non spherical particles from Mie-type theory

B. Hourahine, K. Holms and F. Papoff
Department of Physics, University of Strathclyde, 107 Rottenrow, Glasgow G4 0NG, UK
E-mail: benjamin.hourahine@strath.ac.uk
E-mail: f.papoff@strath.ac.uk

Abstract. We report a new approach for accurate calculation of optical cross sections and internal and scattered fields at any point in space for micro- and nanoparticles. Our approach is based on constructing the intrinsic optical modes of general smooth particles and hence optimised surface Green functions that, for any incident field, provide an a priori upper bound on the error and identify the class of incident fields with largest error.

Non-spherical particles with at least one characteristic length on the order of the wavelength of incident light have remarkable optical properties: metallic particles can enhance the surface field by several orders of magnitude [1]; dielectric particles can have exceptionally high quality factors [2] or focus light into sub-wavelength photonic jets [3]. This underpins a large amount of applied and fundamental research, ranging from various types of surface enhanced nonlinear spectroscopy [4] to single photon light-matter interaction [5]. The optical response of these particles depends both on their shape and constituent material, with internal and scattered electromagnetic fields being solutions of the macroscopic Maxwell equations that satisfy the appropriate boundary conditions at the surface of the particle and at infinity. The great variety of particle shapes used in experiments and the lack of an exact theory analogous to Mie’s solution for spheres leads to the use of fully numerical or semi-analytical methods in theoretical investigations [6, 7]. In order to systematically study the dependence of near and far field optical properties on shape and input field, we need a method that fulfils several requirements. To increase the accuracy of the calculated fields the error with which the boundary conditions are fulfilled must be minimised. Furthermore, as this error depends on the incident field, we need an a priori estimate of the error at any point in space for any incident field if predictions are to be considered reliable. Finally, it is desirable that calculation of the resulting fields arising for different input fields must have a low computational cost for their evaluation at any point in space. In this paper we describe an approximate theory in which, like Mie theory, all fields are expanded as sums of vector functions, and each function in the incident field is associated with only one function in both the internal and scattered fields. This approach fulfils all the requirements mentioned above. In the rest of the paper we outline our method, derive an exact formula for the a priori error for any incident field, and compare this method with selected particles where it is possible to obtain essentially exact results. We conclude with an outline of future developments and with a brief comparison of our method with the most common alternatives.
Figure 1. Surface error for a rod shaped metallic particle of diameter one wavelength (700 nm) and aspect ratio two, illuminated by 45° polarised incident plane waves. The GSGF was optimised for an incident field at 45°; the error bound and the actual evaluated error match at this angle, where the error is due to the part of the field inside the optimising set but outside the range of the Green’s function. (term 1 on the right-hand of Eqn. 2). At 0° incidence the error is clearly seen to be due term 2 of Eqn. 2, implying that this field should also be included in the set over which the GSGF is optimised. For incident angles larger than 45°, the error is of the same order as the 45° incident field within \(\{O^0\}\).

In the following we use the Einstein convention of summation over repeated indexes, unless otherwise specified, and the compact notation \(F^\sigma(x) = [E^\sigma(x), H^\sigma(x)]^T\), with \(\sigma = i, s\) for incident, internal and scattered electromagnetic fields respectively. We indicate the field components of \(F^\sigma\) tangent to the particle surface \(S\) at point \(s\) as \(f^\sigma(s)\). We use \(\langle a | b \rangle = \int_S a^\ast \cdot b \, ds\) and \(\|a\| = \langle a | a \rangle^{1/2}\) for scalar products and norms over \(S\). Dielectrics without losses and metals are modelled by real and complex dielectric constants respectively [8]. For these media, the boundary conditions are that the energy of the scattered field flows toward infinity and the components of the total external and internal fields tangent to the surface of the particle are identical. Electromagnetic fields at the surface of the particle and within the internal and external regions are given by the surface integral \(F^\sigma(x) = \int_S G^\sigma(x, s) F^0(s) \, ds\), where \(G\) is the dyadic surface Green function [9, 10] and \(\sigma = i(s)\) labels the internal (scattered) field.

We have previously [11] approximated \(G\) with an operator that we call a Generalised Surface Green Function (GSGF), because they are based on generalised inverses, using these GSGFs to calculate internal and scattered field of elongated particles for incident fields propagating along the symmetry axis. We now substantially generalise that approach by optimising the GSGF over a set of incident fields, \(\{O^0\}\), and provide a rigorous \(a\) \textit{priori} upper bound on the error everywhere in space and for any incident field. We approximate internal and scattered fields by finite combinations of regular and radiating electric and magnetic multipolar fields, respectively [12, 13]. These are exact solutions of the Maxwell equations and are linearly independent and complete over the surface of the particle [8]. Note that the surface residual \(\|f^0 - f^i + f^s\|\) (for Lyapunov [14] surfaces or those continuous up to at least the second derivative)
provides an upper bound to the maximum error on scattered and internal fields that decreases with the distance from $S$ (see Eqn. 3.3 of Ref. [8]), as long as the error in the propagation, i.e., in the evaluation of the multipolar functions used to expand of solutions of the Maxwell equations, is negligible. For this reason the coefficients $c_j$ of the fields’ expansion that minimise the residual provide an optimal description of the fields at any point. These coefficients are the solutions of the self-adjoint linear system $(\mathbf{f}_i|\mathbf{f}_j)c_j = (\mathbf{f}_i|\mathbf{f}_0)$, but solving this system directly leads to large numerical errors [15]. We address this problem by sampling the tangent components of the of electric and magnetic fields at a set of points on the surface, $\{\Pi\}$, and then constructing orthonormal sets of internal and scattering tangential fields, $\{i\}$ and $\{s\}$, which span the same spaces as the functions $\{\mathbf{f}^i\}$ and $\{\mathbf{f}^s\}$ at those points, via singular value decomposition [16] of the sampled functions and subsequent unitary transformation to pairs of principal vectors [17, 18].

The resulting pairs of internal and scattered functions are orthogonal to all but at most one function in the other space, and oriented at an angle $\xi$ to their partner $(\mathbf{i}_j \cdot \mathbf{s}_j = \cos(\xi_j))$. We can then find the internal and scattered fields for a particular incident field by decomposing it into a sum of pairs of principal modes, $\mathbf{f}^0 = \sum_j a_j^i \mathbf{i}_j - a_j^s \mathbf{s}_j$, where $a_j^i/s$ are amplitudes of each of the internal and scattered principal modes for that specific incident field [18]. This approach provides a substantial improvement in numerical stability compared to the original self-adjoint linear system, combined with a determination of the intrinsic principal modes of the scatterer.

A further important difference with respect to previous work [11, 20] is that the surface points are chosen such that Gaussian quadrature [21] can be applied, resulting in errors in scalar products and norms on $S$ due to discretisation that can be safely neglected as they are of an order lower ($o(\epsilon)$) than the leading error, $\epsilon$, of this theory, which itself is due to the use of finite numbers of functions to expand the fields. By changing the number of functions used in the expansions of $\mathbf{f}^i/s$, how close the zeros and singularities of the expanding functions are to $S$ and the number of principal mode pairs ($r$) included in the solution, we minimise the surface error over a set of different incident fields $\{\mathcal{O}^0\}$, so that we have $\|\mathbf{a}_i^0 - \mathbf{f}^i + \mathbf{f}^s\| \leq \epsilon\|\mathbf{a}_i^0\|$ for each input field $\mathcal{O}_i^0$. Here $\epsilon$ is one or two orders of magnitude smaller than the threshold below which the scattering differential cross section converges [20].

The GSGF can also be constructed from the principal modes of the particle; for a set of modes optimised over $\{\mathcal{O}^0\}$ with leading error $\epsilon$, the GSGF is

$$G^\sigma(\mathbf{x}, \mathbf{s}) = \sum_{j \leq r} \frac{1}{\sin^2 \xi_j} \left\{ \frac{+\mathbf{F}_j(\mathbf{x})(i_j - \cos(\xi) s_j)}{-\mathbf{S}_j(\mathbf{x})(s_j - \cos(\xi) i_j)} \right\} ; \sigma = i \ .$$

(1)

Note that Mie theory can be recast in the form of Eqn. 1 with vector spherical harmonics and spherical vector wavefunctions for incident and scattered light respectively [18].

The advantage of using a set of exact solutions of the Maxwell equations to expand the principal modes is twofold: the result fields can be propagated analytically anywhere in space at virtually no computational cost and some field features can be found analytically. Furthermore, from Eqn. 1 we can derive an exact bound for the surface residual for any incident field, including those outside of $\{\mathcal{O}^0\}$ in terms projectors into and outside of the space of $\{\mathcal{O}^0\}$,

$$P_{\{\mathcal{O}^0\}} = \{|\mathbf{a}_i\rangle\langle \mathbf{a}_i|\}^{-1}$$

and $P_{\perp \{\mathcal{O}^0\}} = 1 - P_{\{\mathcal{O}^0\}}$:

$$\|\mathbf{f}^0 - \mathbf{f}^i + \mathbf{f}^s\| \leq \| (1 - G_{\{\mathcal{O}^0\},\iota}) P_{\{\mathcal{O}^0\}} |\mathbf{f}^0\| + \| (1 - G_{\{\mathcal{O}^0\},\iota}) P_{\perp \{\mathcal{O}^0\}} |\mathbf{f}^0\| \ ,$$

(2)

where $G^S = -\hat{n} \times \hat{n} \times [\mathcal{G}^S(s',s)-\mathcal{G}^s(s',s)]$, with $G^S$ providing the response of the particle through projection of incident fields onto the sets $\{i\}$ and $\{s\}$, while $(1 - G_{\{\mathcal{O}^0\},\iota})$ is the orthogonal projector into the kernel of $G_{\{\mathcal{O}^0\},\iota}^S$. The first term on the right of Eqn. 2 gives the error for the part of an incident field within $\{\mathcal{O}^0\}$ which is lost under the action of the Green’s function, while the second term is the contribution from the part of the incident field outside of the
Figure 2. Scattering cross sections for a) Prolate and b) oblate spheroids with aspect ratios of 2 and a refractive index of 1.5. Both planes of light polarisation with respect to the axis of symmetry are used (0° and 90°). Results are shown for the current method (points) and a Mie-like separation of variables method (lines) [19].
optimisation set which is also outside of the space of the principal modes. The surface residual for a field for which the GSGF has not been optimised is then of the same order $\epsilon$ as for a field in $\{O^0\}$ when the second term of Eqn. 2 is $O(\epsilon)$. In other words, we have identified the part of a generic input field that may reduce the precision of the scattering calculation: it is the input field component that is orthogonal to the set $\{O^0\}$ and within the kernel of the GSGF. This is important not only theoretically, but also practically, even without optimisation. When the GSGF is not optimised, the set $\{O^0\}$ corresponds to the finite set of incident fields in which the error is measured directly, but one can estimate the error for an arbitrary incident field once the error has been tested on a finite set of incident fields. In Fig. 1 we show the two right-hand terms of Eqn. 2 and their sum, as compared against the calculated error for an example particle where the Green's function was optimised for a single incident field incident at 45° with respect to the particle axis.

Once the error is low, the solutions are highly reliable. Figure 2 demonstrates the agreement between the results of our theory, and the essentially exact results from the generalisation of Mie theory to one of the few other cases where coordinate separation is possible [19]. Finally in Fig. 3 we demonstrate the range of particles that can be reliably treated with our approach, showing the maximum value of $\epsilon^2$ for a range of rounded dielectric and metallic rods and disks: minimising the residual over a set of plane waves linearly polarised at 45° to the plane of incidence, with angles of incidence to the symmetry axis ranging from 0° to 90° at intervals of 15°. Note that the limiting case of rods or disks with aspect ratio 1 is a sphere with radius $1/2\pi$ in units of the wavelength of the incident field.

In conclusion, our formalism, as in Mie theory, identifies the contribution to internal and scattered field of each of the functions in the expansion of the incident field and allows us to study the dependence of optical properties of resonant particles on their shape and on the input field. For the determination of internal and scattered fields of symmetric particles over

Figure 3. Variation in optimised surface error for families of dielectric (open circles, refractive index $n = 1.5$) and gold (triangles, refractive index $n \sim 0.32 + 3.18i$) rounded disc and rod-like particles as a function of the ratio between the semi-diameters parallel and perpendicular to the symmetry axis. Aspect ratio 1 corresponds to a sphere with radius $r = 1/2\pi$ wavelengths.
large regions of space, the computational complexity of our method is similar to that of the T-matrix and Discrete Sources [20] approaches, but the principal modes have the advantage of allowing a great compression of information and reduction of memory cost. This approach is less demanding than methods such as the Finite Domain Time Difference (FDTD), because the fields are propagated everywhere simply by evaluating the special functions contained in the multipolar fields. The advantage of our approach with respect to the Discrete Dipole Approximation (DDA) is that the number of multipolar fields necessary for the GSGF is smaller than the number of dipoles used in the DDA. Furthermore, GSGF does not suffer from stair-casing errors and field approximations at metal/dielectric interfaces, as the FDTD and DDA, and have the unique features of providing equations and diagnostic for the error and of allowing a partial analytical analysis. The derivation of GSGF for layered particles, particles with inclusions and particles clusters is straightforward, while GSGF for metallic particles with low symmetry and sharp edges are much more challenging; in these cases the FDTD and DDA [6] remain the methods of choice.

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