JOURNAL OF MODERN OPTICS, 2001, VOL. 48, NO. 14, 2105–2128

Individual and aggregate scattering matrices and cross-sections: conservation laws and reciprocity

BRIAN STOUT†
Institut Fresnel, UMR du CNRS no. 6133, Faculté des Sciences et Techniques, Centre de Saint Jérome 13397 Marseille Cedex 20, France

JEAN-CLAUDE AUGER‡
Centro de Investigacion en Polimeros, COMEX, Marcos Achar Logaton 2, 55885 Tepexpan, Estado de México

and JACQUES LAFAIT
Laboratoire d'Optique des Solides de l'Université Pierre et Marie Curie, UMR 7601, Case 80, 4 place Jussieu 75252 Paris Cedex 05, France

(*Received 7 March 2001; revision received 26 June 2001*)

Abstract. For systems of multiple spheres, we investigate in detail the 'individual' and aggregate electromagnetic scattering matrices, and their relations with conservation laws, reciprocity and the optical theorem. In order for these relations to adopt their simplest form, care is taken to completely extract both incoming and outgoing phase factors in the definitions. We illustrate that the 'individual' cross-sections in an aggregate are defined only in terms of part of the total field, and consequently do not individually obey conservation laws or reciprocity; these relations should be satisfied for the scattering by the entire aggregate. We demonstrate that for scatterer centred transfer matrices, the conservation laws and reciprocity are automatically satisfied regardless of whether or not sufficient multipolarities were retained in the description of individual scatterers. Derivations and results are worked out in a particularly compact and transparent formalism, including magnetic permeability contrast, and the possibility of complex polarizations.

1. Introduction

Although theoretical treatments of multiple scattering began more than a century ago, recent years have seen considerable improvements in the analytical and numerical aspects of the multiple scattering of aggregates, a subject having numerous applications. These developments have the goal of facilitating the

† This work was during his stay at the Laboratoire d'Optique des Solides, Université Pierre et Marise Curie, Paris.

‡ This work was performed during his doctoral research at the Laboratoire d'Optique des Solides, Université Pierre et Marise Curie, Paris.

Journal of Modern Optics ISSN 0950–0340 print/ISSN 1362–3044 online
http://www.tandf.co.uk/journals
DOI: 10.1080/09500340110076428

computer calculations of complex objects and/or multiple scattering calculations. In order to make contact with experimentally measurable quantities, it is often convenient to define quantities in terms of scattering matrices and cross-sections. Mackowski and others have pointed out the utility of defining the aggregate scattering as a sum of 'individual' contributions [1]. These 'individual' scattering quantities are (in some cases) defined in terms of part of the total field, and as such may not necessarily satisfy all the quantities usually associated with true scattering quantities such as positivity, reciprocity, energy conservation, etc. The satisfaction of such relations is, however, assured by the underlying physical equations of motion, and must be recovered when the 'individual' scattering relations are summed to obtain the quantities corresponding to scattering by the entire aggregate.

In order to define physical quantities in a manner free from numerical parasites, it is important to cleanly extract the contributions coming from the regular translation matrices which contain information on the relative positions of scatterers, but also depend on the location of the (arbitrary) origin of the system. Mackowski has already pointed out the utility of the scatterer centred transfer matrices in this regard when treating orientationally averaged quantities [1, 2]. We find that the scatterer centred transfer matrix is equally useful for the orientation fixed quantities. The regular translation matrices remaining in the formulae, can then be conveniently replaced by the exact incident and outgoing wave phase factors.

We will spend some time developing our notation since for reasons of clarity, we found it useful to continue the recent trend of modifying the historical conventions, notations and derivations. Several authors, including Chew in his recent book [3], have pointed out that in the multiple scattering of scalar waves, considerable simplification of the index notation can be achieved by using a matrix notation in which the continual summations over multipolarity indices can be suppressed. It has also been observed by Tsang *et al.* [4], that working with normalized spherical waves simplifies many scattering formulas. Furthermore, these authors have demonstrated that derivations are facilitated by invoking the vector spherical harmonics (rather than working solely with the vector spherical waves). In this work, we have continued the above trend by normalizing the vector spherical harmonics in addition to normalizing the vector spherical waves. Derivations are also facilitated by remarking that a normalized spherical wave basis renders the regular vector translation matrices unitary.

The system under study is that of an aggregate of non-overlapping scatterers of various sizes, labelled by an index j, and whose electromagnetic properties are described by the (complex) relative dielectric and permeability constitutive functions $\varepsilon_j(\omega)$ and $\mu_j(\omega)$. We adopt a time-harmonic framework with $\exp(-\mathrm{i}\omega t)$ time dependence. The aggregate is immersed in a non-absorbing homogenous medium characterized by ε and μ (real constants). Although much of the formalism is applicable to scatterers of general shape, attention will be focused on aggregates of spheres, the logic being that rather complicated continuous and discontinuous objects may be approximated by an appropriate aggregate/agglomerate of spheres. Furthermore, spheres serve as excellent building blocks due to their comparatively simple electromagnetic response.

In section 2, we introduce our conventions. In section 3, we review some essential aggregate scattering concepts such as field expansions, incident wave phase factors, transfer matrices and the far-field limit. In the discussion of the far field, we introduce the scattered wave phase factors as a consequence of the translation matrices operating on far fields. For completeness, the special case of spherical scatterers is rapidly reviewed in section 4, in order to obtain a useful formula relating internal field coefficients to scattering coefficients. We derive a formula for the amplitude-scattering matrix in section 5. Our formula has a particularly simple form, and is readily applicable to arbitrary polarizations. We propose that it is useful to incorporate both incident and scattered wave phase factors into its definition. We also show the constraints placed on the aggregate transfer matrix due to reciprocity.

In section 6, we derive various aggregate and individual cross-section formulas, paying particular attention to the role of the phase factors. Of particular interest is an optical theorem for individual cross-sections. We will see that in the case of spherical scatterers, the principle of flux conservation permits alternative ways of calculating the total scattering and absorption cross-sections. The concept of individual 'cross-sections' is also clarified in this section via illustrative calculations. Notably, we show that individual extinction and scattering cross-sections are not necessarily positive, unlike aggregate cross-sections. It is further demonstrated that individual 'cross-sections' do not satisfy energy conservation relations, but that these relations are satisfied for the entire aggregate. We further demonstrate in this section that conservation laws are satisfied whether or not a sufficient number of multipolarities have been included in the calculation of the scatterer centred transfer matrix of the system.

2. Conventions and normalizations

The vector spherical harmonics (VSH) form a basis for angular vector functions, and are a valuable calculational aid due to the fact that one can appeal to their general properties, such as orthogonality, in order to simplify many integrals. Unfortunately, there exists no universally accepted convention for the VSHs. We will use *normalized* VSHs as defined by Cohen-Tanoudji [5]. Given a direction $\mathbf{r}$, they consist of a longitudinal spherical harmonics $\mathbf{Y}_{nm}(\mathbf{r})$, parallel to $\mathbf{r}$, and two types of 'transverse' spherical harmonics, $\mathbf{Z}_{nm}(\mathbf{r})$ and $\mathbf{X}_{nm}(\mathbf{r})$ perpendicular to $\mathbf{r}$,

$$\mathbf{Y}_{nm}(\mathbf{r}) \equiv \mathbf{r}\, Y_{nm}(\mathbf{r}), \quad \mathbf{X}_{nm}(\mathbf{r}) \equiv \frac{\mathbf{r} \times \nabla_{\mathbf{r}} Y_{nm}(\mathbf{r})}{[n(n+1)]^{1/2}}, \quad \mathbf{Z}_{nm}(\mathbf{r}) \equiv \frac{\nabla_{\mathbf{r}} Y_{nm}(\mathbf{r})}{[n(n+1)]^{1/2}}, \tag{1}$$

where

$$\nabla_{\mathbf{r}} \equiv \hat{\theta}\frac{1}{r}\frac{\partial}{\partial\theta} + \hat{\phi}\frac{1}{r\sin\theta}\frac{\partial}{\partial\phi}.$$

The Y_{nm} are the normalized scalar spherical harmonics (see equation (A 1) of the appendix).

The source-free electromagnetic wave equation in each homogenous zone (indexed j), has the form

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - k_j^2 \mathbf{E}(\mathbf{r}) = 0, \tag{2}$$

where $k_j = 2\pi/\lambda_j$ is the in-medium wavenumber (in terms of the relative constitutive parameters of the medium, $k_j^2 \equiv \varepsilon_j \mu_j (\omega/c)^2$).

The functions $\mathbf{M}_{nm}(k_j\mathbf{r})$ and $\mathbf{N}_{nm}(k_j\mathbf{r})$ satisfy equation (2) in spherical coordinates [6], and are known as the normalized (outgoing) vector spherical waves

$$\begin{aligned}\mathbf{M}_{nm}(k\mathbf{r}) &\equiv -h_n(kr)\mathbf{X}_{nm}(\mathbf{r}),\\ \mathbf{N}_{nm}(k\mathbf{r}) &\equiv \frac{1}{kr}\{[n(n+1)]^{1/2}h_n(kr)\mathbf{Y}_{nm}(\mathbf{r}) + [krh_n(kr)]'\mathbf{Z}_{nm}(\mathbf{r})\},\end{aligned} \tag{3}$$

where $j_n(x)$ and $h_n(x)$ are respectively the spherical Bessel functions and spherical Hankel functions of the first kind. The prime ($'$) in the definition of $\mathbf{N}_{nm}$, equation (3), denotes differentiation with respect to kr. The notation $\mathcal{R}g\{\}$, stands for 'take the regular part of'. Recalling that $h_n(x) \equiv j_n(x) + \mathrm{i}n_n(x)$, where $n_n(x)$ is the spherical Neumann function (irregular at the origin), 'taking the regular part' means replacing $h_n(x)$ by $j_n(x)$.

We have adopted a normalized definition for the spherical waves, $\mathbf{M}_{nm}$ and $\mathbf{N}_{nm}$ [4], wherein their regular parts obey continuum normalization relations

$$\frac{2}{kk'\pi}\int \mathrm{d}^3r\mathcal{R}g\{\mathbf{A}_{nm}(k\mathbf{r})\} \bullet \mathcal{R}g\{\mathbf{B}_{n'm'}(k'\mathbf{r})\} = \delta_{AB}\delta_{nn'}\delta_{mm'}\delta(k-k'), \tag{4}$$

where $\mathbf{A}$ and $\mathbf{B}$ represent either of the normalized spherical waves $\mathbf{M}$ or $\mathbf{N}$. In equation (6), and throughout the rest of this work, we use a heavy dot ($\bullet$) to denote ordinary 3-vector scalar products. An important property of the spherical waves is that they satisfy the easily memorized curl relations

$$\nabla \times \mathbf{M}_{nm}(k\mathbf{r}) = k\mathbf{N}_{nm}(k\mathbf{r}), \quad \nabla \times \mathbf{N}_{nm}(k\mathbf{r}) = k\mathbf{M}_{nm}(k\mathbf{r}). \tag{5}$$

The oldest and most common spherical wave convention in the multiple scattering literature is the unnormalized vector spherical waves [7, 8], $\mathbf{M}_{nm}^{\text{unnorm}} \equiv \nabla u_{nm} \times \mathbf{r}$ and $\mathbf{N}_{nm}^{\text{unnorm}} \equiv (1/k)\nabla \times \mathbf{M}_{nm}^{\text{unnorm}}$, which are defined in terms of the unnormalized scalar waves $u_{nm}^{\text{unnorm}}(\mathbf{r}) \equiv h_n(kr)P_n^m(\cos\theta)\exp(\mathrm{i}m\phi)$. Use of unnormalized waves gives results compatible with Mie's formalism [9]. The relations between the normalized and unnormalized spherical waves are $\mathbf{A}_{nm} = \gamma_{nm}\mathbf{A}_{nm}^{\text{unnorm}}$, where the normalization coefficients, γ_{nm}, are defined

$$\gamma_{nm} \equiv \left[\frac{(2n+1)(n-m)!}{4\pi n(n+1)(n+m)!}\right]^{1/2}. \tag{6}$$

We will see below that our choice of normalized spherical wave functions confers more convenient mathematical properties on the translation matrices.

3. Spherical wave expansions, transfer matrices and far fields

3.1. *Spherical wave expansions of excitation, internal and incident fields*

In an N-particle aggregate, the excitation field for a particle labelled j is generated by the incident field and the (multiple) scattering of the incident field by the other $N-1$ scatterers. The excitation field does not include the field scattered by the jth particle, but it will include fields scattered by the jth particle and reflected back on it by the other particles in the aggregate.

The 'centre' of the jth particle is denoted $\mathbf{x}_j$. The spherical coordinates relative to this scatterer are then $\mathbf{r}_j \equiv \mathbf{r} - \mathbf{x}_j$. The excitation field may be expressed in terms of regular spherical waves centred on the particle in question

$$\mathbf{E}_{\mathrm{e}}^{(j)}(\mathbf{r}) = E_0 \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \{\mathcal{R}g\{\mathbf{M}_{nm}(k\mathbf{r}_j)\} a_{\mathrm{e},nm}^{(j),\mathrm{M}} + \mathcal{R}g\{\mathbf{N}_{nm}(k\mathbf{r}_j)\} a_{\mathrm{e},nm}^{(j),\mathrm{N}}\}$$
$$= E_0 \mathcal{R}g\{\mathcal{E}^{\mathrm{t}}(k\mathbf{r}_j)\}.a_{\mathrm{e}}^{(j)}, \tag{7}$$

where E_0 is a real parameter which determines the amplitude of the wave. In the second line, the infinite sum, $\sum_{n=1}^{\infty}\sum_{m=-n}^{n}$, has been integrated into an abstract vector space notation. Following Tsang *et al.* [4], we define a generalized index $l \equiv n(n+1) - m$ for which each integral value of l corresponds to a unique physical n, m pair. One can then arrange the spherical wave components in an infinite column vector, $\mathcal{E}(k\mathbf{r})$, which will be truncated in numerical applications at some finite multipolarity $n_{\max}$ ($l_{\max} = n_{\max}^2 + 2n_{\max}$)

$$\mathcal{E}(k\mathbf{r}) \equiv \begin{bmatrix} \mathbf{M}(k\mathbf{r}) \\ \mathbf{N}(k\mathbf{r}) \end{bmatrix} = \begin{bmatrix} \mathbf{M}_1(k\mathbf{r}) \\ \vdots \\ \mathbf{M}_{l_{\max}}(k\mathbf{r}) \\ \mathbf{N}_1(k\mathbf{r}) \\ \vdots \\ \mathbf{N}_{l_{\max}}(k\mathbf{r}) \end{bmatrix}. \tag{8}$$

Analogously, $a_{\mathrm{e}}^{(j)}$ is a column vector of (complex) excitation field coefficients. A superscript (t) means that the column vector is transposed into a row vector, and the dots (.) indicate matrix multiplication on the spherical wave components.

Other fields of interest that can be expanded in terms of regular spherical waves are the fields on the interior of the particles ($j = 1, \ldots, N$), $\mathbf{E}_{\mathrm{I}}^{(j)}$, expressed via the internal field coefficients $a_{\mathrm{I}}^{(j)}$, $\mathbf{E}_{\mathrm{I}}^{(j)}(\mathbf{r}) = E_0 \mathcal{R}g\{\mathcal{E}^{\mathrm{t}}(k_j\mathbf{r}_j)\}.a_{\mathrm{I}}^{(j)}$. The incident field, $\mathbf{E}_{\mathrm{i}}$, is also developed in terms of regular spherical waves. Here however, one develops its coefficients once and for all in the origin of our coordinate system, $\mathbf{E}_{\mathrm{i}}(\mathbf{r}) = E_0 \mathcal{R}g\{\mathcal{E}^{\mathrm{t}}(k\mathbf{r})\}.a_{\mathrm{i}}$. Nevertheless, it will frequently prove necessary to reexpress the incident field in terms of coordinates centred on the individual particles.

The corresponding excitation, internal and incident magnetic fields are derived via the Maxwell equation

$$\mathrm{i}\omega\mu(\mathbf{r})\mu_0\mathbf{H}(\mathbf{r}) = \nabla \times \mathbf{E}(\mathbf{r}), \tag{9}$$

which is readily determined via the action of a curl on the spherical waves basis, equation (5):

$$\mathbf{H}(\mathbf{r}) = E_0 \frac{1}{\mathrm{i}\omega\mu\mu_0} \mathcal{R}g\{\nabla \times \mathcal{E}^{\mathrm{t}}(k\mathbf{r}_j)\}.a$$
$$= E_0 \frac{k}{\mathrm{i}\omega\mu\mu_0} \mathcal{R}g\{[\mathbf{N}(k\mathbf{r}), \ \mathbf{M}(k\mathbf{r})]\}. \begin{bmatrix} a^{\mathrm{M}} \\ a^{\mathrm{N}} \end{bmatrix}. \tag{10}$$

An incident electric field of particular interest is that of the homogenous plane wave, $\mathbf{E}_{\mathrm{i}}^{\mathrm{plane}}$, described by a real wave vector, $\mathbf{k}_{\mathrm{i}}$, and a (possibly complex) polarization vector, $\mathbf{e}_{\mathrm{i}}$:

$$\begin{aligned}\mathbf{E}_{\mathrm{i}}^{\mathrm{plane}}(\mathbf{r}) &= E_0\mathbf{e}_{\mathrm{i}}\exp(\mathrm{i}\mathbf{k}_{\mathrm{i}}\cdot\mathbf{r}) \\ &= -E_0 4\pi\sum_{n,m}\mathcal{R}g\{\mathrm{i}^n\mathbf{M}_{nm}(kr,\theta,\phi)\mathbf{X}_{nm}^*(\hat{\mathbf{k}}_{\mathrm{i}}) + \mathrm{i}^{n+1}\mathbf{N}_{nm}(kr,\theta,\phi)\mathbf{Z}_{nm}^*(\hat{\mathbf{k}}_{\mathrm{i}})\}\bullet\mathbf{e}_{\mathrm{i}} \\ &= E_0\mathcal{R}g\{\mathcal{E}^{\mathrm{t}}(k\mathbf{r})\}.p_{\mathrm{i}},\end{aligned} \tag{11}$$

where the incident plane wave coefficients, p_{i}, are expressed as ordinary (three) vector scalar products between the (possibly complex) incident wave polarization vector, $\mathbf{e}_{\mathrm{i}}$ and the transverse VSHs

$$\begin{aligned} p_{\mathrm{i},nm}^{\mathrm{M}} &= -\mathrm{i}^n 4\pi\mathbf{X}_{nm}^*(\hat{\mathbf{k}}_{\mathrm{i}})\bullet\mathbf{e}_{\mathrm{i}} \equiv 4\pi\mathcal{X}_{nm}(\hat{\mathbf{k}}_{\mathrm{i}})\bullet\mathbf{e}_{\mathrm{i}}, \\ p_{\mathrm{i},nm}^{\mathrm{N}} &= -\mathrm{i}^{n+1}4\pi\mathbf{Z}_{nm}^*(\hat{\mathbf{k}}_{\mathrm{i}})\bullet\mathbf{e}_{\mathrm{i}}. \equiv 4\pi\mathcal{Z}_{nm}(\hat{\mathbf{k}}_{\mathrm{i}})\bullet\mathbf{e}_{\mathrm{i}},\end{aligned} \tag{12}$$

where the modified transverse VSHs, $\mathcal{X}$ and $\mathcal{Z}$ have been defined in order to simplify the notation

$$\mathcal{X}_{nm}^*(\mathbf{r}) \equiv -\mathrm{i}^{-n}\mathbf{X}_{nm}(\mathbf{r}), \quad \mathcal{Z}_{nm}^*(\mathbf{r}) \equiv -\mathrm{i}^{-n-1}\mathbf{Z}_{nm}(\mathbf{r}). \tag{13}$$

3.2. *Scattered fields and transfer matrices*

The scattered fields of an N-particle aggregate can be written as the sum of the outgoing spherical waves, equation (3), centred respectively on the individual particles

$$\mathbf{E}_{\mathrm{s}}^{\mathrm{tot}}(\mathbf{r}) = \sum_j^N \mathbf{E}_{\mathrm{s},N}^{(j)}(\mathbf{r}_j) = E_0\sum_j^N \mathcal{E}^{\mathrm{t}}(k\mathbf{r}_j).f_{\mathrm{s}}^{(j)}. \tag{14}$$

where $f_{\mathrm{s}}^{(j)}$ are the body centred scattering coefficients [3].

An object of great practical interest in aggregate scattering is the 1-body transfer matrix $\mathbf{T}_1^{(j)}$. This matrix yields the scattering coefficient of the jth scatterer in terms of the coefficients of the excitation field impinging on the jth scatterer, $a_{\mathrm{e}}^{(j)}$,

$$f_{\mathrm{s}}^{(j)} \equiv \mathbf{T}_1^{(j)}.a_{\mathrm{e}}^{(j)}. \tag{15}$$

The solution of the multiple scattering problem is achieved by deriving the transfer matrices $\mathbf{T}_1^{(j)}$ for all the particles and then solving for the excitation (or scattering) coefficients which using the translation addition theorem can be expressed as

$$a_{\mathrm{e}}^{(j)} = \boldsymbol{\beta}^{(j,0)}.a_0 + \sum_{l=1,l\neq j}^N \boldsymbol{\alpha}^{(j,l)}.f_{\mathrm{s}}^{(l)}, \tag{16}$$

where we have adopted the useful notation of Chew [3], $\boldsymbol{\alpha}^{(j,l)} \equiv \boldsymbol{\alpha}(k(\mathbf{x}_j - \mathbf{x}_l))$, $\boldsymbol{\beta}^{(j,0)} \equiv \boldsymbol{\beta}(k(\mathbf{x}_j - \mathbf{x}_0))$, and $\mathbf{x}_0$ denoting the chosen origin of the system.

We recall that the translation addition theorem for spherical waves can be written [3]

$$
\begin{aligned}
\mathcal{E}^{t}(k\mathbf{r}) &= \mathcal{E}^{t}(k\mathbf{r}').\boldsymbol{\beta}(k\mathbf{r}_0), \quad r' > r_0, \\
\mathcal{E}^{t}(k\mathbf{r}) &= \mathcal{R}g\{\mathcal{E}^{t}(k\mathbf{r}')\}.\boldsymbol{\alpha}(k\mathbf{r}_0), \quad r' < r_0, \\
\mathcal{R}g\{\mathcal{E}^{t}(k\mathbf{r})\} &= \mathcal{R}g\{\mathcal{E}^{t}(k\mathbf{r}')\}.\boldsymbol{\beta}(k\mathbf{r}_0), \quad \forall|\mathbf{r}'|,
\end{aligned} \tag{17}
$$

where $\boldsymbol{\alpha}(k\mathbf{r}_0)$ and $\boldsymbol{\beta}(k\mathbf{r}_0)$ are respectively the irregular and regular translation matrices. The matrix elements of $\boldsymbol{\alpha}(k\mathbf{r}_0)$ are known functions of spherical Hankel functions and associated Legendre functions [3, 7, 8]. The $\boldsymbol{\beta}(k\mathbf{r}_0)$ matrices functions are regular parts of $\boldsymbol{\alpha}(k\mathbf{r}_0)$, i.e. the spherical Hankel functions are replaced by spherical Bessel functions.

A significant advantage obtained by using normalized spherical waves, is that the Hermitian conjugation (†) (complex conjugation plus transpose) of the regular translation matrices corresponds simply to inverse translation, i.e.

$$
\boldsymbol{\beta}^{\dagger}(k\mathbf{r}_0) = \boldsymbol{\beta}(-k\mathbf{r}_0). \tag{18}
$$

From the third line of the translation-addition theorem, equation (17), it can be shown in both normalized and unnormalized notation that

$$
\boldsymbol{\beta}(-k\mathbf{r}_0)\boldsymbol{\beta}(k\mathbf{r}_0) = \mathbf{I}, \tag{19}
$$

where $\mathbf{I}$ is the identity matrix. Taken together, equations (18) and (19) show that the normalized $\boldsymbol{\beta}$ matrices are unitary.

The homogeneous plane wave has particularly useful translation properties

$$
\begin{aligned}
\mathbf{E}_{\mathrm{i}}^{\mathrm{plane}}(\mathbf{r}) &= E_0\mathbf{e}_{\mathrm{i}} \exp(\mathrm{i}\mathbf{k}_{\mathrm{i}}\cdot\mathbf{r}) \\
&= \exp(\mathrm{i}\mathbf{k}_{\mathrm{i}}\cdot\mathbf{x}_j)E_0 \exp(\mathrm{i}\mathbf{k}_{\mathrm{i}}\cdot\mathbf{r}) \\
&= \exp(\mathrm{i}\mathbf{k}_{\mathrm{i}}\cdot\mathbf{x}_j)E_0\mathcal{R}g\{\mathcal{E}^{t}(k\mathbf{r})\}.p_{\mathrm{i}}.
\end{aligned} \tag{20}
$$

The re-expression of an arbitrary incident field in terms of the jth reference frame can also be accomplished via the third line of the translation-addition theorem, equation (17)

$$
\begin{aligned}
\mathbf{E}_{\mathrm{i}}(\mathbf{r}) &= E_0\mathcal{R}g\{\mathcal{E}^{t}(k\mathbf{r})\}.a_{\mathrm{i}} \\
&= E_0\mathcal{R}g\{\mathcal{E}^{t}(k\mathbf{r}_j)\}.\boldsymbol{\beta}(k\mathbf{x}_j).a_{\mathrm{i}}.
\end{aligned} \tag{21}
$$

Comparison of equations (20) and (21), yields the useful result that the regular translation matrix acting on the coefficients of plane wave coefficients corresponds (in the limit $l_{\max} \to \infty$, equation (8)) to a multiplication by a phase factor

$$
\boldsymbol{\beta}(k\mathbf{x}_j).p_{\mathrm{i}} = \exp(\mathrm{i}\mathbf{k}_{\mathrm{i}}\cdot\mathbf{x}_j)p_{\mathrm{i}}. \tag{22}
$$

Taking into account the expression for the plane wave coefficients, equation (12), this formula for $\boldsymbol{\beta}$ may be written as

$$
\boldsymbol{\beta}(k\mathbf{x}_j).\begin{bmatrix} \mathcal{X}(\hat{\mathbf{k}}) \\ \mathcal{Z}(\hat{\mathbf{k}}) \end{bmatrix} = \exp(\mathrm{i}\mathbf{k}\cdot\mathbf{x}_j)\begin{bmatrix} \mathcal{X}(\hat{\mathbf{k}}) \\ \mathcal{Z}(\hat{\mathbf{k}}) \end{bmatrix}. \tag{23}
$$

Taking the complex conjugate of this expression yields

$$
\begin{aligned}
[\mathcal{X}^{*}(\hat{\mathbf{k}}),\ \mathcal{Z}^{*}(\hat{\mathbf{k}})].\boldsymbol{\beta}^{\dagger}(k\mathbf{x}_j) &= [\,\mathcal{X}^{*}(\hat{\mathbf{k}}),\ \mathcal{Z}^{*}(\hat{\mathbf{k}})\,].\boldsymbol{\beta}(-k\mathbf{x}_j) \\
&= \left[\,\mathcal{X}^{*}(\hat{\mathbf{k}}),\ \mathcal{Z}^{*}(\hat{\mathbf{k}})\,\right] \exp(-\mathrm{i}\mathbf{k}\cdot\mathbf{x}_j).
\end{aligned} \tag{24}
$$

In calculations, these infinite vectors $\mathcal{X}(\hat{\mathbf{k}})$ and $\mathcal{Z}(\hat{\mathbf{k}})$ are truncated to some finite value of the orbital angular momentum number, $n_{\max}$. In order for equation (24) to be satisfied to some given precision, $n_{\max}$ is an increasing function of $|k\mathbf{x}_j|$.

In the expression of equation (14) for the scattered electric fields, the spherical waves, $\mathcal{E}^{\mathrm{t}}(k\mathbf{r}_j)$, are centred respectively on each of the scatterers. It will frequently prove useful to develop these waves about the origin of the aggregate. Using the fact that $\boldsymbol{\beta}(-k\mathbf{x}_j) = \boldsymbol{\beta}^{-1}(k\mathbf{x}_j)$, the second line of the translation-addition theorem, equation (17), can be expressed as

$$\mathcal{E}^{\mathrm{t}}(k\mathbf{r}_j) = E_0 \mathcal{E}^{\mathrm{t}}(k\mathbf{r}).[\boldsymbol{\beta}(-k\mathbf{x}_j)], \quad \mathbf{r} > \mathbf{x}_j. \tag{25}$$

In situations where one is only interested in the behaviour of fields outside of the aggregate, equation (25) allows the total scattered field to be expressed as

$$\mathbf{E}_{\mathrm{s}}^{\mathrm{tot}}(\mathbf{r}) = E_0 \sum_j \mathcal{E}^{\mathrm{t}}(k\mathbf{r}).\boldsymbol{\beta}(-k\mathbf{x}_j).f_{\mathrm{s}}^{(j)}, \quad \mathbf{r} > \mathbf{x}_j \ \forall \ \mathbf{x}_j \tag{26}$$

$$= E_0 \sum_j \mathcal{E}^{\mathrm{t}}(k\mathbf{r}).\boldsymbol{\beta}^{\dagger}(k\mathbf{x}_j).f_{\mathrm{s}}^{(j)} \tag{27}$$

$$= E_0 \sum_j \mathcal{E}^{\mathrm{t}}(k\mathbf{r}).\boldsymbol{\beta}^{(0,j)}.f_{\mathrm{s}}^{(j)}. \tag{28}$$

Defining the aggregate centred scattering coefficients, $a_{\mathrm{s}}^{(j)}$ as

$$\begin{aligned} a_{\mathrm{s}}^{(j)} &\equiv \boldsymbol{\beta}(-k\mathbf{x}_j).f_{\mathrm{s}}^{(j)} \\ &= \boldsymbol{\beta}^{(0,j)}.f_{\mathrm{s}}^{(j)}, \end{aligned} \tag{29}$$

the total scattered field exterior to the aggregate can be written in the aggregate centred coordinates

$$\mathbf{E}_{\mathrm{s}}^{\mathrm{tot}}(\mathbf{r}) = E_0 \sum_j \mathcal{E}^{\mathrm{t}}(k\mathbf{r}).a_{\mathrm{s}}^{(j)}. \tag{30}$$

A particularly useful means of formulating the solution is in terms of the scatterer centred transfer matrices of the entire N-particle system, $\mathbf{T}_N^{(j,k)}$ [1]. This transfer matrix can be seen as grouping all the multiple interactions as interactions between pairs of particles. Since this matrix contains all local and far field multiple scattering information, it directly yields the scattering coefficients in terms of the known incident field

$$\begin{aligned} f_{\mathrm{s}}^{(j)} &= \sum_k \mathbf{T}_N^{(j,k)}.\boldsymbol{\beta}(k\mathbf{x}_k).a_{\mathrm{i}} \\ &= \sum_k \mathbf{T}_N^{(j,k)}.\boldsymbol{\beta}^{(k,0)}.a_{\mathrm{i}}. \end{aligned} \tag{31}$$

In this equation, $\boldsymbol{\beta}(k\mathbf{x}_k)$ represents the regular translation-addition matrix. Thanks to the presence of $\boldsymbol{\beta}(k\mathbf{x}_j)$ in this definition, $\mathbf{T}_N^{(j,k)}$ depends only on the sizes and relative positions of the scatterers, but not on the choice of the origin.

3.3. *Far field*

Quantities of typical interest in scattering calculations involve the fields at points $\mathbf{r}$, far from the aggregate, i.e. $\lim r \to \infty$. In this limit, the outgoing and regular spherical waves adopt simpler forms:

$$\lim_{r\to\infty} \mathsf{M}_{nm}(k\mathbf{r}) \to \mathrm{i}\mathrm{i}^{-n}\frac{\exp(\mathrm{i}kr)}{kr}\mathsf{X}_{nm}(\mathbf{r}) = -\mathrm{i}\frac{\exp(\mathrm{i}kr)}{kr}\mathcal{X}^*_{nm}(\mathbf{r}),$$
$$\lim_{r\to\infty} \mathsf{N}_{nm}(k\mathbf{r}) \to \mathrm{i}\mathrm{i}^{-n-1}\frac{\exp(\mathrm{i}kr)}{kr}\mathsf{Z}_{nm}(\mathbf{r}) = -\mathrm{i}\frac{\exp(\mathrm{i}kr)}{kr}\mathcal{Z}^*_{nm}(\mathbf{r}) \tag{32}$$

and

$$\lim_{r\to\infty} \mathcal{R}g\{\mathsf{M}_{nm}(k\mathbf{r})\} \to \frac{-\mathrm{i}}{2kr}\{\exp(\mathrm{i}kr) - (-1)^n\exp(-\mathrm{i}kr)\}\mathcal{X}^*_{nm}(\mathbf{r}),$$
$$\lim_{r\to\infty} \mathcal{R}g\{\mathsf{N}_{nm}(k\mathbf{r})\} \to \frac{-\mathrm{i}}{2kr}\{\exp(\mathrm{i}kr) + (-1)^n\exp(-\mathrm{i}kr)\}\mathcal{Z}^*_{nm}(\mathbf{r}). \tag{33}$$

The scattered spherical wave vectors in the far-field limit have thus a simple representation in terms of the modified VSHs

$$\lim_{r\to\infty} \mathcal{E}^{\mathrm{t}}(k\mathbf{r}) = -\mathrm{i}\frac{\exp(\mathrm{i}kr)}{kr}[\mathcal{X}^*(\mathbf{r}),\ \ \mathcal{Z}^*(\mathbf{r})]. \tag{34}$$

Inserting this value into the expression for the far field in terms of the scattering coefficients, equation (26), and using the conjugate phase relation, equation (24), the aggregate centred scattered waves are subject to a phase shift, $\exp(-\mathrm{i}k\mathsf{x}_j\cdot\mathbf{r})$,

$$\lim_{r\to\infty} \mathsf{E}^{(j)}_{\mathrm{s}}(\mathbf{r}) = \lim_{r\to\infty} E_0\mathcal{E}^{\mathrm{t}}(k\mathbf{r}).\boldsymbol{\beta}(-k\mathsf{x}_j).f^{(j)}_{\mathrm{s}}$$
$$= -\mathrm{i}E_0\frac{\exp(\mathrm{i}kr)}{kr}\exp(-\mathrm{i}k\mathsf{x}_j\cdot\mathbf{r})[\mathcal{X}^*(\mathbf{r}),\ \ \mathcal{Z}^*(\mathbf{r})].\begin{bmatrix} f^{(j),\mathsf{M}}_{\mathrm{s}} \\ f^{(j),\mathsf{N}}_{\mathrm{s}} \end{bmatrix}. \tag{35}$$

This phase shift is exactly what one would expect in the far field based on different propagation path lengths.

Invoking the relation for the magnetic field, equation (9), and the curl of vector waves, equation (5), the magnetic far-field limit can be expressed

$$\lim_{r\to\infty} \mathsf{H}^{(j)}_{\mathrm{s}}(\mathbf{r}_j) = -\mathrm{i}E_0\frac{k}{\omega\mu\mu_0}\frac{\exp(\mathrm{i}kr)}{kr}\left[-\mathrm{i}\mathcal{Z}^*_{nm}(\mathbf{r}),\ -\mathrm{i}\mathcal{X}^*_{nm}(\mathbf{r})\right].\begin{bmatrix} f^{(j),\mathsf{M}}_{\mathrm{s}} \\ f^{(j),\mathsf{N}}_{\mathrm{s}} \end{bmatrix}$$
$$= \frac{k}{\omega\mu\mu_0}\mathbf{r}\times\mathsf{E}^{(j)}_{\mathrm{s}}(\mathbf{r}), \tag{36}$$

where the properties, equation (A 11) of the modified VSHs were invoked.

4. Spherical scatterers and internal fields

In the interest of completeness, we quickly review in this section, the one-body transfer matrix results for spherical scatterers, and derive the relation between the internal and scattering coefficients. This result is used in section 6 for calculating the absorption cross-section. Invoking the developments of the internal, excitation and scattered fields, and imposing the electric and magnetic field boundary conditions at the surface of a sphere of radius R_j, one readily finds the four equations [6],

$$
\begin{aligned}
\psi_n(\rho_j\chi_j)a_{\mathrm{I},nm}^{(j),\mathsf{M}} &= \rho_j\psi_n(\chi_j)[a_{\mathrm{e}}^{(j)}]_{nm}^{\mathsf{M}} + \rho_j\xi_n(\chi_j)f_{\mathrm{s},nm}^{(j),\mathsf{M}}, \\
\mu\psi_n'(\rho_j\chi_j)a_{\mathrm{I},nm}^{(j),\mathsf{M}} &= \mu_j\psi_n'(\chi_j)[a_{\mathrm{e}}^{(j)}]_{nm}^{\mathsf{M}} + \mu_j\xi_n'(\chi_j)f_{\mathrm{s},nm}^{(j),\mathsf{M}}, \\
\psi_n'(\rho_j\chi_j)a_{\mathrm{I},nm}^{(j),\mathsf{N}} &= \rho_j\psi_n'(\chi_j)[a_{\mathrm{e}}^{(j)}]_{nm}^{\mathsf{N}} + \rho_j\xi_n'(\chi_j)f_{\mathrm{s},nm}^{(j),\mathsf{N}}, \\
\mu\psi_n(\rho_j\chi_j)a_{\mathrm{I},nm}^{(j),\mathsf{N}} &= \mu_j\psi_n(\chi_j)[a_{\mathrm{e}}^{(j)}]_{nm}^{\mathsf{N}} + \mu_j\xi_n(\chi_j)f_{\mathrm{s},nm}^{(j),\mathsf{N}},
\end{aligned}
\tag{37}
$$

where χ_j and ρ_j are respectively the dimensionless size parameter $\chi_j \equiv kR_j = 2\pi R_j/\lambda$ (λ is wavelength in the host medium) and the index contrast parameter, $\rho_j \equiv k_j/k = (\varepsilon_j\mu_j/\varepsilon\mu)^{1/2}$. These equations were rendered rather compact by using Ricatti Bessel functions, $\psi_n(x) \equiv xj_n(x)$, $\xi_n(x) \equiv xh_n(x)$.

Eliminating the internal field from these equations, and invoking the definition of the transfer matrix, equation (15), one finds the classic Mie result that only the diagonal elements of $\mathbf{T}_1^{(j)}$ are non-zero, and given by

$$
\begin{aligned}
\mathbf{T}_{1;nm,\nu\mu}^{(j),\mathbf{MM}} &= \delta_{n\nu}\delta_{m\mu}\frac{\mu_j\psi_n(\rho_j\chi_j)\psi_n'(\chi_j) - \mu\rho_j\psi_n'(\rho_j\chi_j)\psi_n(\chi_j)}{\mu\rho_j\psi_n'(\rho_j\chi_j)\xi_n(\chi_j) - \mu_j\psi_n(\rho_j\chi_j)\xi_n'(\chi_j)}, \\
\mathbf{T}_{1;nm,\nu\mu}^{(j),\mathbf{NN}} &= \delta_{n\nu}\delta_{m\mu}\frac{\mu_j\psi_n'(\rho_j\chi_j)\psi_n(\chi_j) - \mu\rho_j\psi_n(\rho_j\chi_j)\psi_n'(\chi_j)}{\mu\rho_j\psi_n(\rho_j\chi_j)\xi_n'(\chi_j) - \mu_j\psi_n'(\rho_j\chi_j)\xi_n(\chi_j)}
\end{aligned}
\tag{38}
$$

Alternatively, one can obtain the coefficients for the fields in the interior of particle by eliminating the scattered fields in equation (37). Upon invoking the Wronskian relation, one obtains [3, 10]

$$
\begin{aligned}
a_{\mathrm{I},nm}^{(j),\mathsf{M}} &= \mu_j\rho_j\frac{\mathrm{i}}{\mu_j\psi_n(\rho_j\chi_j)\xi_n'(\chi_j) - \mu\rho_j\psi_n'(\rho_j\chi_j)\xi_n(\chi_j)}[a_{\mathrm{e}}^{(j)}]_{nm}^{\mathsf{M}}, \\
a_{\mathrm{I},nm}^{(j),\mathsf{N}} &= \mu_j\rho_j\frac{\mathrm{i}}{\mu\rho_j\psi_n(\rho_j\chi_j)\xi_n'(\chi_j) - \mu_j\psi_n'(\rho_j\chi_j)\xi_n(\chi_j)}[a_{\mathrm{e}}^{(j)}]_{nm}^{\mathsf{N}}.
\end{aligned}
\tag{39}
$$

Inserting after the fraction the one body transfer matrix for a sphere, equation (38), and its inverse, and appealing to the definition of the transfer matrix, equation (15), one obtains an expression for the internal field coefficients of a sphere in terms of the scattered field coefficients

$$
\begin{aligned}
a_{\mathrm{I},nm}^{(j),\mathsf{M}} &= \mu_j\rho_j\frac{\mathrm{i}}{\mu\rho_j\psi_n'(\rho_j\chi_j)\psi_n(\chi_j) - \mu_j\psi_n(\rho_j\chi_j)\psi_n'(\chi_j)}f_{\mathrm{s},nm}^{(j),\mathsf{M}}, \\
a_{\mathrm{I},nm}^{(j),\mathsf{N}} &= \mu_j\rho_j\frac{\mathrm{i}}{\mu_j\psi_n(\chi_j)\psi_n'(\rho_j\chi_j) - \mu\rho_j\psi_n(\rho_j\chi_j)\psi_n'(\chi_j)}f_{\mathrm{s},nm}^{(j),\mathsf{N}}.
\end{aligned}
\tag{40}
$$

5. Amplitude-scattering matrices

The amplitude-scattering matrices describe the scattered waves far from the aggregate resulting from an incident plane wave. Inserting the far-field results of equation (35) for the jth particle, yields

$$\lim_{r\to\infty}\frac{kr\mathbf{E}_s^{(j)}(\mathbf{r})}{\exp(ikr)E_0} = -i\,\exp(-ik\mathbf{x}_j\cdot\mathbf{r})[\mathcal{X}^*(\mathbf{r}),\mathcal{Z}^*(\mathbf{r})].f_s^{(j)}$$
$$= -i\,\exp(-ik\mathbf{x}_j\cdot\mathbf{r})[\mathcal{X}^*(\mathbf{r}),\mathcal{Z}^*(\mathbf{r})].\mathbf{T}_N^{(j,k)}.\boldsymbol{\beta}^{(k,0)}.p_i$$
$$= -i\,\exp(ik(\hat{\mathbf{k}}_i\cdot\mathbf{x}_k - \mathbf{r}\cdot\mathbf{x}_j))[\mathcal{X}^*(\mathbf{r}),\mathcal{Z}^*(\mathbf{r})].\mathbf{T}_N^{(j,k)}.p_i. \qquad (41)$$

Recalling the expression for the plane wave coefficients, equation (12), we obtain

$$\lim_{r\to\infty}\frac{kr\mathbf{E}_s^{(j)}(\mathbf{r})}{\exp(ikr)E_0} = \frac{1}{i}\sum_k 4\pi\,\exp(ik(\hat{\mathbf{k}}_i\cdot\mathbf{x}_k - \mathbf{r}\cdot\mathbf{x}_j))[\mathcal{X}^*(\mathbf{r}),\mathcal{Z}^*(\mathbf{r})].\mathbf{T}_N^{(j,k)}.\begin{bmatrix}\mathcal{X}(\hat{\mathbf{k}}_i)\bullet\mathbf{e}_i\\ \mathcal{Z}(\hat{\mathbf{k}}_i)\bullet\mathbf{e}_i\end{bmatrix}. \qquad (42)$$

This formula prompts the definition of the individual amplitude-scattering dyadic, $\overline{\overline{S}}^{(j)}$ [4] as the dyadic giving the far-field scattered wave originating from the jth particle in terms of the incident wave polarization vector, $\mathbf{e}_i$, and field strength, E_0

$$\lim_{r\to\infty}\mathbf{E}_s^{(j)}(\mathbf{r}) = E_0\frac{\exp(ikr)}{-ikr}\overline{\overline{S}}^{(j)}(\mathbf{r},\hat{\mathbf{k}}_i)\bullet\mathbf{e}_i. \qquad (43)$$

Comparison of equation (43) and equation (42) shows that the amplitude-scattering dyadic is expressed

$$\overline{\overline{S}}^{(j)}\left(\mathbf{r},\hat{\mathbf{k}}_i\right) = -4\pi\sum_l \exp(ik(\hat{\mathbf{k}}_i\cdot\mathbf{x}_l - \mathbf{r}\cdot\mathbf{x}_j))[\mathcal{X}^*(\mathbf{r}),\mathcal{Z}^*(\mathbf{r})].\mathbf{T}_N^{(j,l)}.\begin{bmatrix}\mathcal{X}(\hat{\mathbf{k}}_i)\\ \mathcal{Z}(\hat{\mathbf{k}}_i)\end{bmatrix}$$
$$= -4\pi\sum_l [\mathcal{X}^*(\mathbf{r}),\mathcal{Z}^*(\mathbf{r})].\boldsymbol{\beta}^{(0,j)}.\mathbf{T}_N^{(j,l)}.\boldsymbol{\beta}^{(l,0)}.\begin{bmatrix}\mathcal{X}(\hat{\mathbf{k}}_i)\\ \mathcal{Z}(\hat{\mathbf{k}}_i)\end{bmatrix}, \qquad (44)$$

where both incoming and scattered wave phase factors have been put into the definition of $\overline{\overline{S}}^{(j)}$. As discussed in section 3, the second line of equation (44) is only true in the limit $l_{\max}\to\infty$.

An expression useful for proving an individual scattering optical theorem is obtained from the real part of the scattering matrix in the forward direction with the same polarization. This expression for a possibly complex polarization reads

$$\mathrm{Re}\,[\mathbf{e}_i^*\bullet\overline{\overline{S}}^{(j)}(\hat{\mathbf{k}}_i,\hat{\mathbf{k}}_i)\bullet\mathbf{e}_i]$$
$$= -4\pi\,\mathrm{Re}\left\{\sum_l \exp(ik\hat{\mathbf{k}}_i\cdot(\mathbf{x}_l - \mathbf{x}_j))[\mathbf{e}_i^*\bullet\mathcal{X}^*(\hat{\mathbf{k}}_i),\mathbf{e}_i^*\bullet\mathcal{Z}^*(\hat{\mathbf{k}}_i)].\mathbf{T}_N^{(j,l)}.\begin{bmatrix}\mathcal{X}(\hat{\mathbf{k}}_i)\bullet\mathbf{e}_i\\ \mathcal{Z}(\hat{\mathbf{k}}_i)\bullet\mathbf{e}_i\end{bmatrix}\right\}$$
$$= -\frac{1}{4\pi}\mathrm{Re}\left\{\sum_l \exp(ik\hat{\mathbf{k}}_i\cdot(\mathbf{x}_l - \mathbf{x}_j))p_i^\dagger.\mathbf{T}_N^{(j,l)}.p_i\right\}. \qquad (45)$$

Inspection of equation (45) shows that the amplitude-scattering dyadic has non-zero components only in the transverse directions for the incident and scattered wave. Therefore the amplitude-scattering matrix $S^{(j)}(\theta,\phi;\theta_i,\phi_i)$ can be written as a 2×2 matrix acting on the polarization degrees of freedom of the incident wave, $e_{\theta,i}$ and $e_{\phi,i}$

$$\begin{bmatrix} E^{(j)}_{\theta,\mathrm{s}} \\ E^{(j)}_{\phi,\mathrm{s}} \end{bmatrix} = \frac{\exp(\mathrm{i}kr)}{-\mathrm{i}kr} \begin{bmatrix} S^{(j)}_{\theta\theta} & S^{(j)}_{\theta\phi} \\ S^{(j)}_{\phi\theta} & S^{(j)}_{\phi\phi} \end{bmatrix} \begin{bmatrix} e_{\theta,\mathrm{i}} \\ e_{\phi,\mathrm{i}} \end{bmatrix} E_0, \tag{46}$$

where we identify $S^{(j)}_{\theta\theta} = \hat{\theta} \bullet \overline{\overline{S}}^{(j)} \bullet \hat{\theta}_\mathrm{i}$, $S^{(j)}_{\theta\phi} = \hat{\theta} \bullet \overline{\overline{S}}^{(j)} \bullet \hat{\phi}_\mathrm{i}$, etc. This definition of $S^{(j)}$ differs from one commonly encountered [6] in that it incorporates *both* incident and scattered wave phase factors. A considerable advantage of this definition is that the aggregate amplitude-scattering dyadic (matrix) can be defined simply as the sum of the individual ones,

$$\overline{\overline{S}}^{\mathrm{agg}} \equiv \sum_j \overline{\overline{S}}^{(j)}. \tag{47}$$

An important symmetry relation is imposed on the aggregate amplitude-scattering matrix due to the reciprocity of the Maxwell equations [4]. In the present context, this relation can be found by considering a dipole source, A, located at point $\mathbf{r}_\mathrm{i}$ sufficiently far from the aggregate such that the field at the aggregate may be approximated by a plane wave of wave-vector $\hat{\mathbf{k}}_\mathrm{i} = -\hat{\mathbf{r}}_\mathrm{i}$, and a dipole receiver, B, located at point $\mathbf{r}$ sufficiently far from the aggregate that the far-field limit can be applied. The reciprocity principle tells us that the field response is identical if we interchange the source to be located at B (such that $\hat{\mathbf{k}}_\mathrm{i} \to -\hat{\mathbf{r}}$) and the receiver at $\mathbf{r}_\mathrm{i}$ (such that $\hat{\mathbf{r}} \to -\hat{\mathbf{k}}_\mathrm{i}$). Reciprocity thus yields for the *aggregate* amplitude-scattering dyadic

$$\left[\overline{\overline{S}}^{\mathrm{agg}}(-\hat{\mathbf{k}}_\mathrm{i}, -\hat{\mathbf{r}})\right]^\mathrm{t} = \overline{\overline{S}}^{\mathrm{agg}}(\hat{\mathbf{r}}, \hat{\mathbf{k}}_\mathrm{i}). \tag{48}$$

Using the second line the expression for individual amplitude-scattering dyadics, equation (44), and the properties of the VSHs under inversion, equation (A 8), we find

$$\begin{aligned} \left[\overline{\overline{S}}^{\mathrm{agg}}(-\hat{\mathbf{k}}_\mathrm{i}, -\hat{\mathbf{r}})\right]^\mathrm{t} = -4\pi \sum_{j,l} \sum_{nm,\nu\mu} & \\ \times \{(-1)^{m+\mu} \mathsf{X}_{\nu\mu}(\hat{\mathbf{k}}_\mathrm{i}) [\boldsymbol{\beta}^\dagger(k\mathbf{x}_j) . \mathbf{T}_N^{(j,l)} . \boldsymbol{\beta}(k\mathbf{x}_l)]^{\mathsf{MM}}_{\nu\mu,nm} \mathsf{X}^*_{nm}(\hat{\mathbf{r}}). & \\ + (-1)^{m+\mu} \mathsf{X}_{\nu\mu}(\hat{\mathbf{k}}_\mathrm{i}) [\boldsymbol{\beta}^\dagger(k\mathbf{x}_j) . \mathbf{T}_N^{(j,l)} . \boldsymbol{\beta}(k\mathbf{x}_l)]^{\mathsf{MN}}_{\nu\mu,nm} \mathsf{Z}^*_{nm}(\hat{\mathbf{r}}) & \\ + (-1)^{m+\mu} \mathsf{Z}_{\nu\mu}(\hat{\mathbf{k}}_\mathrm{i}) [\boldsymbol{\beta}^\dagger(k\mathbf{x}_j) . \mathbf{T}_N^{(j,l)} . \boldsymbol{\beta}(k\mathbf{x}_l)]^{\mathsf{NM}}_{\nu\mu,nm} \mathsf{X}^*_{nm}(\hat{\mathbf{r}}) & \\ + (-1)^{m+\mu} \mathsf{Z}_{\nu\mu}(\hat{\mathbf{k}}_\mathrm{i}) [\boldsymbol{\beta}^\dagger(k\mathbf{x}_j) . \mathbf{T}_N^{(j,l)} . \boldsymbol{\beta}(k\mathbf{x}_l)]^{\mathsf{NN}}_{\nu\mu,nm} \mathsf{Z}^*_{nm}(\hat{\mathbf{r}})\}. & \end{aligned} \tag{49}$$

Comparison with the original expression, equation (44), yields relations which must be satisfied by the transfer matrices

$$\sum_{j,l} [\boldsymbol{\beta}^\dagger(k\mathbf{x}_j) . \mathbf{T}_N^{(j,l)} . \boldsymbol{\beta}(k\mathbf{x}_l)]^{\mathsf{AB}}_{\nu\mu,nm} = (-1)^{m+\mu} \sum_{j,l} [\boldsymbol{\beta}^\dagger(k\mathbf{x}_j) . \mathbf{T}_N^{(j,l)} . \boldsymbol{\beta}(k\mathbf{x}_l)]^{\mathsf{AB}}_{nm,\nu\mu}, \tag{50}$$

where A and B can represent either M or N.

One may obtain reciprocity relations free of the translation matrices by employing the first line of equation (44) containing phase factors. Restricting then to the case of $\Omega = \Omega_i$, one then obtains relations for the sum of the transfer matrices

$$\sum_{j,l} [\mathbf{T}_N^{(j,l)}]^{AB}_{\nu\mu,nm} = (-1)^{m+\mu} \sum_{j,l} [\mathbf{T}_N^{(j,l)}]^{AB}_{nm,\nu\mu}. \tag{51}$$

6. Cross-sections

While emphasizing the role of phase factors, we shall derive formulas for multiple scattering cross-sections which have appeared recently or concurrently in the literature (in one form or another). Cross-sections are defined in terms of the power flux, i.e. in terms of the Poynting vector, $\mathbf{S} \equiv \frac{1}{2}\,\mathrm{Re}\,(\mathbf{E} \times \mathbf{H}^*)$. Since the total field consists of the incident field and the fields scattered from each of the particles in the aggregate, the total Poynting vector can be decomposed in three parts $\mathbf{S}^{agg}_{tot} = \mathbf{S}_{inc} + \mathbf{S}^{agg}_{ext} + \mathbf{S}^{agg}_{scat}$, where $\mathbf{S}_{inc}$, $\mathbf{S}^{agg}_{ext}$ and $\mathbf{S}^{agg}_{scat}$ are respectively the incident, extinction and scattered flux of the aggregate. Explicitly, they are defined

$$\begin{aligned}
&\mathbf{S}_{inc} \equiv \tfrac{1}{2}\,\mathrm{Re}\,\{\mathbf{E}_i \times \mathbf{H}_i^*\}, && \text{incident,} \\
&\mathbf{S}^{agg}_{scat} \equiv \tfrac{1}{2}\,\mathrm{Re}\,\{\textstyle\sum_{j,l} \mathbf{E}_s^{(j)} \times \mathbf{H}_s^{(l)*}\}, && \text{scattering,} \\
&\mathbf{S}^{agg}_{ext} \equiv \textstyle\sum_j \mathbf{S}^{(j)}_{ext} \equiv \tfrac{1}{2}\sum_j \mathrm{Re}\,\{\mathbf{E}_i \times \mathbf{H}_s^{(j)*} + \mathbf{E}_s^{(j)} \times \mathbf{H}_i^*\}, && \text{extinction,}
\end{aligned} \tag{52}$$

where the summations are performed on the particle labels.

In order to define cross-sections, one calculates the incident flux, $\mathbf{S}_{inc}$, assuming a homogenous host medium with real constitutive parameters ε and μ (i.e. no absorption). In the computation of cross-sections, one assumes incident plane waves, $\mathbf{E}_i(\mathbf{r}) = E_0 \mathbf{e}_i \exp(\mathrm{i}\mathbf{k}_i \cdot \mathbf{r})$ which yields

$$\begin{aligned}
\mathbf{S}_{inc} &= \frac{1}{2\omega\mu\mu_0}\,\mathrm{Re}\,\{\mathrm{i}\mathbf{E}_i \times \nabla \times \mathbf{E}_i^*\} \\
&= \frac{1}{2\omega\mu\mu_0}\,\mathrm{Re}\{(E_0^2 k \hat{\mathbf{k}}_i - E_0^2 k (\mathbf{e}_i \bullet \hat{\mathbf{k}}_i)\mathbf{e}_i^*)\} \\
&= \frac{E_0^2}{2}\left(\frac{\varepsilon}{\mu}\frac{\epsilon_0}{\mu_0}\right)^{1/2} \hat{\mathbf{k}}_i.
\end{aligned} \tag{53}$$

where in the last line, one invokes the fact that for a homogenous plane wave, the polarization vector, $\mathbf{e}_i$ is perpendicular to the wave vector $\mathbf{k}_i$.

6.1. *Differential scattering cross-section*

The differential scattering cross-section is defined as the angular function of scattered flux per unit solid angle dΩ at large distances from the scatterers divided by the unit power flux of the incident wave in the direction of incidence

$$\begin{aligned}
\frac{d\sigma^{agg}_{scat}(\theta, \phi, \theta_i, \phi_i)}{d\Omega} &\equiv \lim_{r\to\infty} r^2 \frac{\mathbf{r} \bullet \mathbf{S}^{agg}_{scat}(\mathbf{r})}{\hat{\mathbf{k}}_i \bullet \mathbf{S}_{inc}} \\
&= \lim_{r\to\infty} r^2 \frac{|\sum_j \mathbf{E}_s^{(j)}(\mathbf{r})|^2}{E_0^2} = \sum_j \frac{d\sigma^{(j)}_{scat}}{d\Omega},
\end{aligned} \tag{54}$$

where in the second line one uses the relation for the far magnetic field, equation (36). In the last equality, we have taken advantage of the sum over the particle labels to define individual *real valued* differential scattering 'cross-sections' for the jth particle

$$\frac{\mathrm{d}\sigma_{\mathrm{scat}}^{(j)}}{\mathrm{d}\Omega} \equiv \lim_{r\to\infty} r^2 \frac{\sum_l \mathrm{Re}\,\{(\mathbf{E}_{\mathrm{s}}^{(j)}(\mathbf{r}))^* \bullet \mathbf{E}_{\mathrm{s}}^{(l)}(\mathbf{r})\}}{E_0^2}. \tag{55}$$

When carrying out an integration over the scattering angles (as we will do below), it is useful to write out the above product explicitly using the aggregate centred field development, equation (30), and the far-field limit of the scattered electric field, equation (34) to obtain

$$\frac{\mathrm{d}\sigma_{\mathrm{scat}}^{(j)}(\theta,\phi,\theta_{\mathrm{i}},\phi_{\mathrm{i}})}{\mathrm{d}\Omega} = \frac{1}{k^2} \times \sum_l \mathrm{Re}\left\{ [(a_{\mathrm{s}}^{(j),\mathsf{M}})^*, \ (a_{\mathrm{s}}^{(j),\mathsf{N}})^*]. \begin{bmatrix} \mathcal{X}(\mathbf{r}) \\ \mathcal{Z}(\mathbf{r}) \end{bmatrix} \right.$$
$$\left. \bullet [\mathcal{X}^*(\mathbf{r}), \ \mathcal{Z}^*(\mathbf{r})]. \begin{bmatrix} a_{\mathrm{s}}^{(l),\mathsf{M}} \\ a_{\mathrm{s}}^{(l),\mathsf{N}} \end{bmatrix} \right\}. \tag{56}$$

If the subject of interest is the differential cross-section for the entire aggregate, and not some integration over angles, then one may simply insert the results for the aggregate amplitude-scattering matrix, equations (47), (43) and (44), into equation (56) to obtain

$$\begin{aligned}\frac{\mathrm{d}\sigma_{\mathrm{scat}}^{\mathrm{agg}}(\theta,\phi,\theta_{\mathrm{i}},\phi_{\mathrm{i}})}{\mathrm{d}\Omega} &= \frac{1}{k^2} |\overline{\overline{S}}^{\mathrm{agg}}(\mathbf{r},\hat{\mathbf{k}}_{\mathrm{i}}) \bullet \mathbf{e}_{\mathrm{i}}|^2 \\ &= \frac{1}{k^2}\left\{ \left| \sum_{j,l} \exp\left(\mathrm{i}k(\hat{\mathbf{k}}_{\mathrm{i}}\cdot\mathbf{x}_l - \mathbf{r}\cdot\mathbf{x}_j)\right) \right.\right. \\ &\quad \left. \times (\boldsymbol{\theta} \bullet \mathsf{X}(\mathbf{r}).[\mathbf{T}_N^{(j,l)}.p_{\mathrm{i}}]^{\mathsf{M}} + \boldsymbol{\theta} \bullet \mathsf{Z}(\mathbf{r}).[\mathbf{T}_N^{(j,l)}.p_{\mathrm{i}}]^{\mathsf{N}}) \right|^2 \\ &\quad + \left| \sum_{j,l} \exp\left(\mathrm{i}k(\hat{\mathbf{k}}_{\mathrm{i}}\cdot\mathbf{x}_l - \mathbf{r}\cdot\mathbf{x}_j)\right)(\boldsymbol{\phi} \bullet \mathsf{X}(\mathbf{r}).[\mathbf{T}_N^{(j,l)}.p_{\mathrm{i}}]^{\mathsf{M}} \right. \\ &\quad \left.\left. + \boldsymbol{\phi} \bullet \mathsf{Z}(\mathbf{r}).[\mathbf{T}_N^{(j,l)}.p_{\mathrm{i}}]^{\mathsf{N}}) \right|^2 \right\},\end{aligned} \tag{57}$$

where in the last line the vector components and phase factors are written out explicitly. A practical version of this formula in terms of Legendre functions is obtained by inserting the expressions for the VSHs of the appendix, equation (A 2).

It is clear from the above formula that the phase factors play an important role in determining the interference between the scattering sources. As a demonstration of the utility of these formulas, we show in figure 1, the good agreement obtained between calculations of the differential cross-section, using equation (57), and measured values taken by Wang and Gustafson using a microwave analogue technique [11]. The system consists of an aggregate of two identical polystyrene

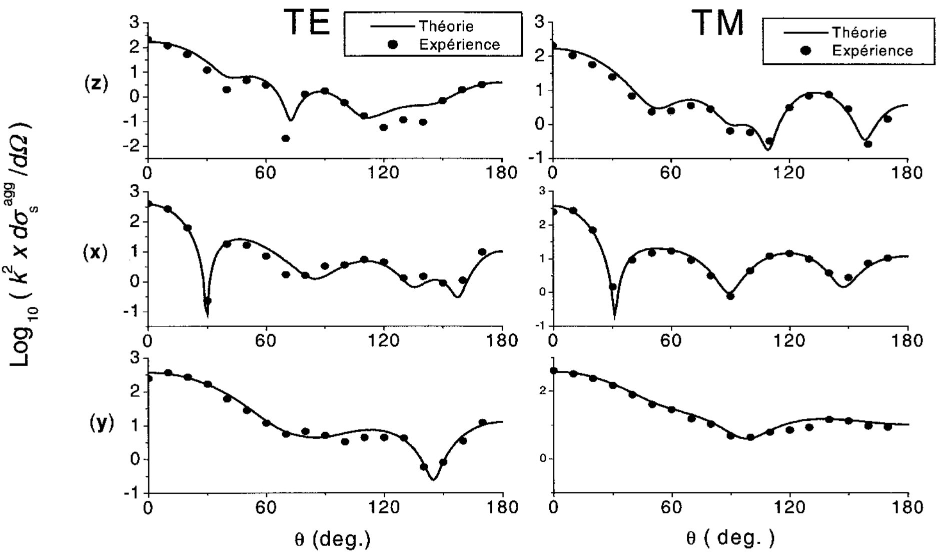

Figure 1. Comparison between the theoretical and experimental differential scattering cross-sections, $d\sigma_s^{agg}/d\Omega$, for identical touching spheres, dielectric contrast parameter $\rho = 1.61 + 0.004i$ and size parameter $\chi = kR = 3.083$. The incident wave-vector ($\theta_i = 0$) lies along the positive $\mathbf{z}$ direction. Measurements taken in the $\mathbf{x}$–$\mathbf{z}$, ($\phi = 0$) plane are plotted as a function of the polar angle, θ, of the outgoing wave. The polarization vector, $\mathbf{e}_i$, of the incident electric field is transverse electric (TE), or transverse magnetic (TM) for $\mathbf{e}_i$ oriented along the $\mathbf{y}$ and $\mathbf{x}$ axis respectively. Comparisons are made when the symmetry axis of the two spheres lies respectively along the $\mathbf{z}$, $\mathbf{x}$ and $\mathbf{y}$ axis (denoted in the parenthesis of the plots).

spheres (of radius R) in contact ($|\mathbf{x}_1 - \mathbf{x}_2| = 2R$) for different orientations and polarizations. We plot the base 10 logarithm of the square of the wavenumber multiplying the differential cross-section, $k^2 \times d\sigma_{scat}^{agg}(\theta, \phi = 0, \theta_i = 0)/d\Omega$, (alternatively written as $|\underline{\underline{S}}^{agg}(\theta, \phi = 0, \theta_i = 0) \bullet \mathbf{e}_i|^2$) as a function of the polar angle, θ, of the outgoing measurement direction.

6.2. *Total scattering cross-section*

Although the differential cross-section is somewhat complicated, results for the total scattering cross-section are simplified by the orthogonality of the VSHs. Carrying out an integral over all angles of the differential cross-sections, equations (54) and (56), the total scattering cross-section for the aggregate, σ_{scat}^{agg}, may be written $\sigma_{scat}^{agg} = \sum_{j=1}^{N} \sigma_{scat}^{(j)}$, where the $\sigma_{scat}^{(j)}$ are the angular integrals of the individual differential 'cross-sections', equation (55),

$$\sigma_{scat}^{(j)}(\theta_i, \phi_i) \equiv \int \frac{d\sigma_{scat}^{(j)}(\theta, \phi, \theta_i, \phi_i)}{d\Omega} d\Omega = \frac{1}{k^2} \sum_{l}^{N} \mathrm{Re}\{f_s^{(j)\dagger} . \boldsymbol{\beta}^{(j,0)} . \boldsymbol{\beta}^{(0,l)} . f_s^{(l)}\}$$

$$= \frac{1}{k^2} \sum_{l}^{N} \mathrm{Re}\{f_s^{(j)\dagger} . \boldsymbol{\beta}^{(j,l)} . f_s^{(l)}\}. \qquad (58)$$

Inserting the expressions for the body centred and aggregate centred scattering coefficients, equations (29) and (31) into the above formula, one obtains

$$\sigma_{\mathrm{scat}}^{(j)}(\theta_{\mathrm{i}},\phi_{\mathrm{i}}) = \frac{1}{k^2}\,\mathrm{Re}\left\{\sum_{l,k,m} a_{\mathrm{i}}^{\dagger}.\boldsymbol{\beta}^{(m,0)\dagger}.\mathbf{T}_N^{(j,m)\dagger}.\boldsymbol{\beta}^{(j,l)}.\mathbf{T}_N^{(l,k)}.\boldsymbol{\beta}^{(k,0)}.a_{\mathrm{i}}\right\}$$
$$= \frac{1}{k^2}\,\mathrm{Re}\left\{\sum_{l,k,m} a_{\mathrm{i}}^{\dagger}.\boldsymbol{\beta}^{(0,m)}.\mathbf{T}_N^{(j,m)\dagger}.\boldsymbol{\beta}^{(j,l)}.\mathbf{T}_N^{(l,k)}.\boldsymbol{\beta}^{(k,0)}.a_{\mathrm{i}}\right\}, \tag{59}$$

where in the second line we invoked the unitarity properties of the regular translation matrix.

Of particular interest is the incident plane wave in which incident phase factors will give rise to interference effects

$$\sigma_{\mathrm{scat}}^{(j)}(\theta_{\mathrm{i}},\phi_{\mathrm{i}}) = \frac{1}{k^2}\mathrm{Re}\left\{\sum_{l,k,m} \exp\left(\mathrm{i}\mathbf{k}_{\mathrm{i}}\cdot(\mathbf{x}_k-\mathbf{x}_m)\right)p_{\mathrm{i}}^{\dagger}.\mathbf{T}_N^{(j,m)\dagger}.\boldsymbol{\beta}^{(j,l)}.\mathbf{T}_N^{(l,k)}.p_{\mathrm{i}}\right\}. \tag{60}$$

One can view the scattered wave factors as having gone into the formation of the $\boldsymbol{\beta}^{(j,l)}$ matrix.

6.3. *Extinction cross-section and optical theorem*

Unlike the incident and scattering flux, there is no shortcut for calculating the extinction flux outside of the optical theorem. One can then either prove the optical theorem by manipulation of the underlying equations [4, 6], or prove the optical theorem by simply calculating the extinction cross-section. We choose the latter, since this approach is perhaps the most straightforward when treating individual extinction 'cross-sections'. In any case, the direct calculation is considerably facilitated by the use of normalized VSHs. Using the definition of the extinction flux, equation (52), and equation (9), the aggregate extinction cross-section can be written as $\sigma_{\mathrm{ext}}^{\mathrm{agg}} = \sum_{j=1}^{N}\sigma_{\mathrm{ext}}^{(j)}$, where the individual extinction 'cross-sections', $\sigma_{\mathrm{ext}}^{(j)}$ are defined as

$$\sigma_{\mathrm{ext}}^{(j)}(\theta_{\mathrm{i}},\phi_{\mathrm{i}}) \equiv \lim_{r\to\infty} -\int \mathrm{d}\Omega\, r^2 \frac{\mathbf{r}\bullet\mathbf{S}_{\mathrm{ext}}^{(j)}(\mathbf{r})}{\hat{\mathbf{k}}_{\mathrm{i}}\bullet\mathbf{S}_{\mathrm{inc}}}$$
$$= \lim_{r\to\infty} -r^2\int \mathrm{d}\Omega \frac{\mathbf{r}\bullet\mathrm{Re}\{\mathrm{i}\mathbf{E}_{\mathrm{s}}^{(j)}\times\nabla\times\mathbf{E}_{\mathrm{i}}^{*} - \mathrm{i}\mathbf{E}_{\mathrm{i}}^{*}\times\nabla\times\mathbf{E}_{\mathrm{s}}^{(j)}\}}{kE_0^2}. \tag{61}$$

The evaluation of $\sigma_{\mathrm{ext}}^{(j)}$ proceeds in a straightforward manner. First, one inserts the far-field limits for the regular and scattered waves, equation (33) and equation (32) respectively, and then eliminates the $\nabla\times$ operator by invoking the spherical wave relation of equation (5). We are left with vector cross products of the transverse VSHs, but these are given by the relations of equation (A 6). The final step is to carry out an integration over all angles, and invoke the orthonormality relations of the VSHs to obtain

$$\sigma_{\mathrm{ext}}^{(j)}(\theta_{\mathrm{i}},\phi_{\mathrm{i}}) = -\frac{1}{k^2}\,\mathrm{Re}\{a_{\mathrm{i}}^{\dagger}.a_{\mathrm{s}}^{(j)}\}. \tag{62}$$

Inserting as usual the regular translation matrices appearing in equations (30) and

(31), we obtain

$$\sigma_{\text{ext}}^{(j)}(\theta_{\text{i}}, \phi_{\text{i}}) = -\frac{1}{k^2}\text{Re}\{a_{\text{i}}^{\dagger}.a_{\text{s}}^{(j)}\}$$
$$= -\frac{1}{k^2}\sum_{l} \text{Re}\{a_{\text{i}}^{\dagger}.\beta^{(0,j)}.\mathbf{T}_N^{(j,l)}.\beta^{(l,0)}.a_{\text{i}}\}. \tag{63}$$

Specializing to incident plane waves, p_{i}, and invoking the phase shift equation, equation (23) and its conjugate, we find

$$\sigma_{\text{ext}}^{(j)}(\theta_{\text{i}}, \phi_{\text{i}}) = -\frac{1}{k^2}\sum_{l} \text{Re}\{\exp(\text{i}k\hat{\mathbf{k}}_{\text{i}} \cdot (\mathbf{x}_l - \mathbf{x}_j))p_{\text{i}}^{\dagger}.\mathbf{T}_N^{(j,l)}.p_{\text{i}}\}. \tag{64}$$

Comparing the above equation to the $\overline{\overline{S}}$ dyadic relation of equation (46) for the forward scattering, we obtain an *individual* particle optical theorem, valid for complex polarizations

$$\sigma_{\text{ext}}^{(j)} = \frac{4\pi}{k^2}\text{Re}\{\mathbf{e}_{\text{i}}^{*} \bullet \overline{\overline{S}}^{(j)}(\hat{\mathbf{k}}_{\text{i}}, \hat{\mathbf{k}}_{\text{i}}) \bullet \mathbf{e}_{\text{i}}\}. \tag{65}$$

Restricted to real polarization vectors, this reproduces a result derived in [4] for isolated scatterers. Recalling the definition of the aggregate amplitude-scattering matrix, equation (48), the sum of the above formula over particle labels leads to an aggregate optical theorem, $\sigma_{\text{ext}}^{\text{agg}} = (4\pi/k^2)\,\text{Re}\{\mathbf{e}_{\text{i}}^{*} \bullet \overline{\overline{S}}^{\text{agg}}(\hat{\mathbf{k}}_{\text{i}}, \hat{\mathbf{k}}_{\text{i}}) \bullet \mathbf{e}_{\text{i}}\}$.

6.4. *Absorption cross-section*

For energy absorbing particles in a lossless medium, the absorption cross-section is generally calculated via energy conservation. That is, since the medium is lossless, all absorption occurs inside the particles, and therefore the total absorbed flux can be obtained by integrating the total flux on any closed surface surrounding the system. This observation leads to the relation $\sigma_{\text{abs}}^{\text{agg}} = \sigma_{\text{ext}}^{\text{agg}} - \sigma_{\text{scat}}^{\text{agg}}$ [4] for the aggregate absorption cross-section.

An alternative method for calculating $\sigma_{\text{abs}}^{\text{agg}}$ is to evaluate the absorption via the flux through the surfaces of the particles, the aggregate cross-section then being expressed as the sum of individual absorption cross-sections $\sigma_{\text{abs}}^{\text{agg}} = \sum_j^N \sigma_{\text{abs}}^{(j)}$. The integral for the flux through the particle surfaces can readily be carried out in the case of spherical scatterers (radii R_j). The normal to the surface in the sense of incoming flux then being $-\hat{\mathbf{r}}$, the individual absorption cross-sections for spherical scatterers are then expressed as

$$\sigma_{\text{abs}}^{(j)} = -\frac{\oint_A \hat{\mathbf{r}} \bullet \mathbf{S}_{\text{I}}^{(j)}\,\text{d}A}{\hat{\mathbf{k}}_{\text{i}} \bullet \mathbf{S}_{\text{inc}}}$$
$$= -\frac{\omega\mu\mu_0 R_j^2}{E_0^2 k}\int \text{d}\Omega\,\hat{\mathbf{r}} \bullet \text{Re}\{\mathbf{E}_{\text{I}}^{(j)} \times \mathbf{H}_{\text{I}}^{(j)*}\}. \tag{66}$$

Inserting the spherical wave developments of the internal electric and magnetic fields, equation (10), then making use of the vector product expressions for the spherical harmonics, equation (A 6), and lastly invoking the orthonormality relations of the VSHs, yields

$$\sigma_{\mathrm{abs}}^{(j)}=\left(\frac{\mu}{\varepsilon}\right)^{1/2}\frac{1}{\left|\rho_j\right|^2k^2}\sum_{n,m}\left[\mathrm{Re}\left\{\mathrm{i}\left(\frac{\varepsilon_j}{\mu_j}\right)^{1/2}\psi_n^*(\rho_j\chi_j)\psi_n'(\rho_j\chi_j)\right\}|a_{\mathrm{I},nm}^{(j),\mathsf{M}}|^2\right.$$

$$\left.+\,\mathrm{Re}\left\{\mathrm{i}\left(\left(\frac{\varepsilon_j}{\mu_j}\right)^{1/2}\right)^*\psi_n^*(\rho_j\chi_j)\psi_n'(\rho_j\chi_j)\right\}|a_{\mathrm{I},nm}^{(j),\mathsf{N}}|^2\right]. \tag{67}$$

It is clear that in the case of a non-absorbing scatterer (i.e. ε_j and μ_j are real constants), that the absorption cross-section is zero.

Using the expression relating the internal field coefficients to the scattering coefficients, equation (40), this result may be written as

$$\sigma_{\mathrm{abs}}^{(j)}=\frac{1}{k^2}\sum_{n,m}\{C_n^{(j)}|f_{\mathrm{s},nm}^{(j),\mathsf{M}}|^2+D_n^{(j)}|f_{\mathrm{s},nm}^{(j),\mathsf{N}}|^2\}, \tag{68}$$

where

$$\begin{aligned}C_n^{(j)}&=\frac{\mathrm{Re}\{\mathrm{i}\rho_j\mu\mu_j^*\psi_n^*(\rho_j\chi_j)\psi_n'(\rho_j\chi_j)\}}{\left|\mu_j\psi_n(\rho_j\chi_j)\psi_n'(\chi_j)-\mu\rho_j\psi_n'(\rho_j\chi_j)\psi_n(\chi_j)\right|^2},\\ D_n^{(j)}&=\frac{\mathrm{Re}\{\mathrm{i}\rho_j^*\mu\mu_j\psi_n^*(\rho_j\chi_j)\psi_n'(\rho_j\chi_j)\}}{\left|\mu\rho_j\psi_n(\rho_j\chi_j)\psi_n'(\chi_j)-\mu_j\psi_n(\chi_j)\psi_n'(\rho_j\chi_j)\right|^2}.\end{aligned} \tag{69}$$

In the case of $\mu_j=\mu=1$, this result reduces to the absorption formula for spherical scatterers derived by Mackowski [12].

A matrix multiplication expression method of writing the above absorption formula is to define a diagonal matrix $\boldsymbol{\Gamma}^{(j)}$ of the form

$$\boldsymbol{\Gamma}^{(j)}=\begin{bmatrix}C^{(j)} & 0\\ 0 & D^{(j)}\end{bmatrix}, \tag{70}$$

where the matrix elements of $C^{(j)}$ and $D^{(j)}$ are respectively $[C^{(j)}]_{nm,\nu\mu}=\delta_{n\nu}\delta_{m\mu}C_n^{(j)}$, $[D^{(j)}]_{nm,\nu\mu}=\delta_{n\nu}\delta_{m\mu}D_n^{(j)}$. In the matrix notation, one can write

$$\begin{aligned}\sigma_{\mathrm{abs}}^{(j)}&=\frac{1}{k^2}f_{\mathrm{s}}^{(j)\dagger}.\boldsymbol{\Gamma}^{(j)}.f_{\mathrm{s}}^{(j)}\\ &=\frac{1}{k^2}\sum_{l,m}a_{\mathrm{i}}^{\dagger}.\boldsymbol{\beta}^{(0,m)}.\mathbf{T}_N^{(j,m)\dagger}.\boldsymbol{\Gamma}^{(j)}.\mathbf{T}_N^{(j,l)}.\boldsymbol{\beta}^{(l,0)}.a_{\mathrm{i}}.\end{aligned} \tag{71}$$

When evaluating for incoming plane waves, the formula takes the form

$$\sigma_{\mathrm{abs}}^{(j)}=\frac{1}{k^2}\sum_{l,m}\exp\left(\mathrm{i}k\hat{\mathbf{k}}_{\mathrm{i}}\cdot(\mathbf{x}_l-\mathbf{x}_m)\right)p_{\mathrm{i}}^{\dagger}.\mathbf{T}_N^{(j,m)\dagger}.\boldsymbol{\Gamma}^{(j)}.\mathbf{T}_N^{(j,l)}.p_{\mathrm{i}}. \tag{72}$$

We remark that as opposed to the individual scattering cross-sections, equation (60), formulas for the individual absorption and extinction cross-sections, $\sigma_{\mathrm{abs}}^{(j)}$ and $\sigma_{\mathrm{ext}}^{(j)}$ are simpler since they contain no translation-addition matrices and consequently, as suggested by Mackowski [1], it generally proves simpler (faster) to calculate the total aggregate scattering cross-section in terms of the $\sigma_{abs}^{(j)}$ and $\sigma_{ext}^{(j)}$, i.e. $\sigma_{scat}^{agg} = \sum_j (\sigma_{ext}^{(j)} - \sigma_{abs}^{(j)})$.

6.5. *Total cross-sections and energy conservation*

Large scale calculations were not the aim of the present work, and we have not discussed the calculation of the N-body transfer matrices. These have been discussed in other works [3, 13], and in a forthcoming work by us. It seemed nevertheless useful to give some results obtained for the total cross-sections using the above formulas in order to clarify the physical content of the individual 'cross-sections'.

The individual absorption cross-sections, $\sigma_{abs}^{(j)}$, have a clear physical interpretation since they represent the absorption in the jth sphere individually and are calculated in terms of the *total* field in the interior of the scatterers. The $\sigma_{abs}^{(j)}$ are thus necessarily non-negative for passive media. Although the individual scattering and extinction 'cross-sections' are practical calculational objects, and contain useful physical information, they should not be viewed as true cross-sections since they involve only *part* of the total field. For instance, although the aggregate scattering must satisfy energy conservation, $\sigma_{ext}^{agg} = \sigma_{scat}^{agg} + \sigma_{abs}^{agg}$, the energy conservation relations are generally *not* satisfied by the individual 'cross-sections' as we have defined them ($\sigma_{ext}^{(j)} \neq \sigma_{scat}^{(j)} + \sigma_{abs}^{(j)}$). In addition, although the individual scattering and extinction 'cross-sections' are real quantities by definition, nothing requires them to be positive (contrary to $\sigma_{abs}^{(j)}$).

For the sake of simplicity, we will give results for a two sphere aggregate consisting of two identical touching spheres with radii $R_1 = R_2 = R$, and plot individual and total cross-sections (divided by R^2) as functions of the size parameter, $\chi = kR = (\omega/c)(\varepsilon\mu)^{1/2}R$. In figure 2, we take zero permeability contrast, $\mu_j = \mu = 1$, and invoke the same dielectric contrast as used in the comparison with experiment (see figure 1), except that we set the imaginary part strictly to zero, $\rho_1 = \rho_2 = 1.61$. The symmetry axis of the sphere is along the $\mathbf{z}$ axis $((\mathbf{x}_1)_z = -(\mathbf{x}_2)_z = -R,\ (\mathbf{x}_1)_{x,y} = (\mathbf{x}_2)_{x,y} = 0)$. The incident wave vector is taken to be oriented along the positive $\mathbf{z}$ direction. In figure 2 (*a*), we illustrate the individual and aggregate extinction cross-sections and remark that for certain size parameters, the extinction 'cross-section' of particle 2 is negative. In figure 2 (*b*), we show the individual and aggregate scattering cross-sections and remark that the individual scattering cross-section of particle 2 is generally larger than that of particle 1 ('lens effect' of particle 1). Energy conservation for the aggregate cross-sections is strictly observed, $\sigma_{ext}^{agg} = \sigma_{scat}^{agg}$.

In figure 3, we keep the same real part of the dielectric contrast as in figure 2, but add some absorption $\rho_1 = \rho_2 = 1.61 + 0.1i$. The individual and aggregate extinction and scattering cross-sections are given in (*a*) and (*b*) respectively. We remark that whereas the tendency for negative extinction individual 'cross-sections' diminishes with increased absorption, the individual scattering 'cross-section' of particle 2 becomes slightly negative at a size parameter of $\chi \approx 3.8$. The individual and aggregate absorption cross-sections are plotted in (*c*). Figure 3 (*d*) displays the aggregate absorption, scattering and extinction cross-sections (all of which are positive, and satisfy energy conservation, $\sigma_{ext}^{agg} = \sigma_{scat}^{agg} + \sigma_{abs}^{agg}$).

It is important to remark that although energy conservation can test the accuracy of an algorithm for the calculation the scatterer centred transfer matrix,

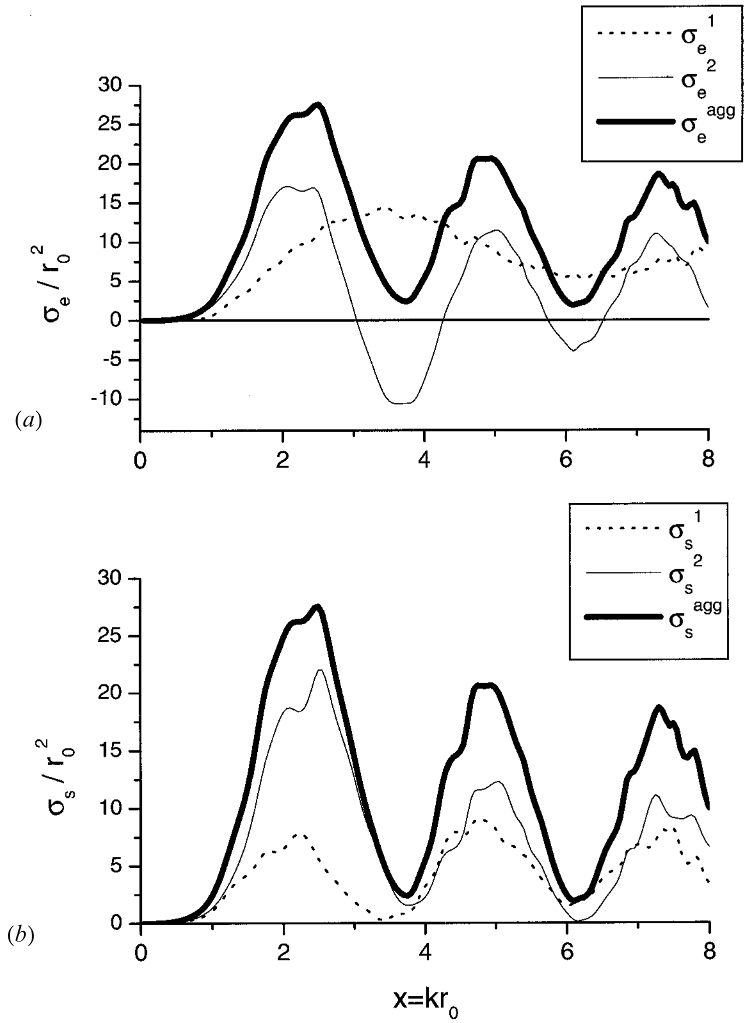

Figure 2. Absorption free aggregate cross-sections for identical touching spheres, $\rho_{\text{sphere}} \equiv k_{\text{sphere}}/k = 1.61$, radius$=R$. The incident wave and symmetry axis both lie on the z axis. Individual and aggregate cross-sections (divided by R^2) are plotted as a function of the size parameter, $\chi = kR$. (*a*) Extinction cross-sections. (*b*) Scattering cross-sections.

$\mathbf{T}_N^{(i,j)}$, it cannot tell us whether or not the individual scatterers were accurately described. For instance, should we correctly calculate $\mathbf{T}_N^{(i,j)}$ in an overly truncated multipolarity space ($n_{\max}$ too small) energy conservation will still hold but the result can be inaccurate due to a poor description of the individual scatterers. This effect is demonstrated in table 1. We present the total extinction scattering and absorption cross-sections for a system of seven identical dielectric touching spheres wherein one sphere is placed at the origin, and a pair of spheres is placed on each of the x, y and z axes, all equidistant from the origin. We consider a plane wave travelling along the positive z direction and a size parameter of $\chi = kR = 3$ for all the spheres. The index of refraction of the spheres is chosen as

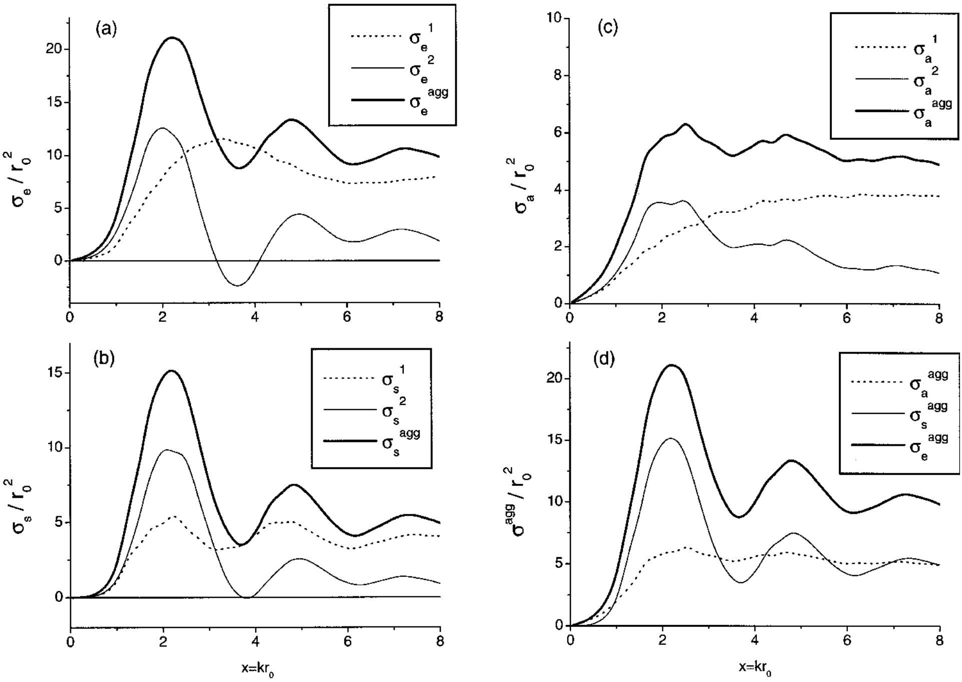

Figure 3. Aggregate cross-sections with absorption. This is the same system as figure 2, except that an imaginary (absorbing) part has been added to the dielectric contrast parameter, $\rho_{\rm sphere} = 1.61 + .1i$. Individual and aggregate extinction, σ_e, and scattering, σ_s, cross-sections (divided by R^2) are plotted in (*a*) and (*b*) respectively as a function of the size parameter, $\chi = kR$. (*c*) Individual and aggregate absorption cross-sections. (*d*) Aggregate absorption and scattering cross-sections ($\sigma_a^{\rm agg}$ and $\sigma_s^{\rm agg}$), and their sum (which is identical to the aggregate extinction cross-section, $\sigma_e^{\rm agg}$).

Table 1. Extinction, scattering and absorption cross-sections for a system of seven dielectric touching spheres located at the origin, and on the x, y and z axes respectively. The refractive index of the spheres is $n_{\rm sph} = 2.5 + 0.01i$, while that of the host medium is $n_m = 1.5$.

$n_{\max}$	$\sigma_{\rm ext}$	$\sigma_{\rm scatt}$	$\sigma_{\rm abs}$	percent
1	0.58176	0.56958	0.01218	(~40%)
2	1.34668	1.30563	0.04105	(~90%)
3	1.49606	1.40997	0.08683	(~99.2%)
4	1.5014	1.4132	0.0882	(~99.4%)
5	1.51003	1.42162	0.08841	(~99.97%)
6	1.5105	1.42201	0.08849	

$n_{\rm sph} = 2.5 + 0.01i$, and that of the surrounding medium as $n_{\rm med} = 1.5$. The calculations were performed using a rigorously calculated scatterer centred transfer matrix, $\mathbf{T}_N^{(i,j)}$ and using the formulae derived in this paper. The total cross-sections are presented for different values of the $n_{\max}$ orbital cut-off. We remark that energy conservation ($\sigma_{\rm ext} = \sigma_{\rm scat} + \sigma_{\rm abs}$) is obeyed for arbitrary cut-off values, but that the results are reliable only for a cut-off of $n_{\max} \geq 3$.

7. Conclusions

We have derived relations imposed on the aggregate centred scattering matrices, $\mathbf{T}_N^{(i,j)}$, by the underlying conservation laws and symmetry relations. We have seen that for appropriate definitions, the optical theorem can be applied to individual scatterers, while the conservation laws only manifest themselves once all the individual field contributions have been summed. Similarly, the reciprocity relations are also satisfied only on the level of the entire aggregate. We have demonstrated some typical applications of these formulae with calculations. In orientation fixed calculations, we have emphasized that in order to avoid truncation errors, it is important to replace regular translation matrices acting on incident and scattered wave coefficients by their exact phase eigenvalues.

An important observation of this work is that energy conservation can serve as a test for the accurate calculation of the transfer matrix, $\mathbf{T}_N^{(i,j)}$, and the cross-section formulae. Once reliable methods have been employed, conservation relations are satisfied regardless of whether or not the calculations are performed for a multipolarity basis sufficiently large to describe the individual scatterers. An important deficiency of the present article is that we have not presented a reliable means of calculating the scatterer centred scattering matrices, $\mathbf{T}_N^{(i,j)}$. This shall be presented in a forthcoming article, which uses this matrix to calculate the local fields present in aggregate scattering.

In this work, we have only discussed systems composed of dielectric spheres, and the reader may well wonder about applications to metallic spheres. Our studies on metallic spheres indicate that there is the possibility for reliable calculations for metallic spheres provided that calculations are performed in the strongly scattering regime ($kR \gtrsim 1$) and/or well separated spheres. Applications to small ($kR \ll 1$) closely packed metallic spheres however can present problems for our formalism due to the existence of large polarization charges which occur in metallic spheres in the quasi-static limit. Such systems apparently require a more refined or modified treatment.

We emphasize that the methods presented in this work can be applied to an arbitrary configuration of spheres. Studies of disordered systems can often be considerably improved by invoking configuration averages of the system [2] (not presented here). Our formulae, for fixed orientations of the incident field with respect to the system may find some of their best applications in the rapidly expanding field of photonic crystals. Our formalism is particularly suited to the studies of finite size effects, since there are no assumptions of 'infinite' crystal lattices, plane wave developments or the Bloch theorem. The theory presented here finds a close analogue in treatments of the scattering by '2-dimensional' systems composed of 'infinite' cylinders. A number of studies of 2D photonic crystal systems [14, 15] and waveguides [16] have recently been reported or are currently in progress.

Appendix: Vector spherical harmonics (VSHs)

The normalized scalar spherical harmonics can be written in terms of the associated Legendre functions $P_n^m(x)$,

$$Y_{nm}(\mathbf{r}) = Y_{nm}(\theta,\phi) = \left[\frac{2n+1}{4\pi}\frac{(n-m)!}{(n+m)!}\right]^{1/2} P_n^m(\cos\theta)\exp(im\phi)$$
$$= \gamma_{nm}[n(n+1)]^{1/2} P_n^m(\cos\theta)\exp(im\phi), \quad \text{(A 1)}$$

where $\mathbf{r}$ is some unit vector and θ and ϕ its angular coordinates. The normalization coefficient, γ_{nm}, is defined in equation (6). Making use of equation (A 1), the normalized VSHs, defined in equation (1), can be explicitly written in terms of the associated Legendre functions

$$\mathbf{Y}_{nm}(\mathbf{r}) = \gamma_{nm}[n(n+1)]^{1/2} P_n^m(\cos\theta)\exp(im\phi)\mathbf{r},$$
$$\mathbf{X}_{nm}(\mathbf{r}) = \gamma_{nm}\left[-\frac{im}{\sin\theta}P_n^m(\cos\theta)\exp(im\phi)\boldsymbol{\theta} + \frac{d}{d\theta}P_n^m(\cos\theta)\exp(im\phi)\boldsymbol{\phi}\right], \quad \text{(A 2)}$$
$$\mathbf{Z}_{nm}(\mathbf{r}) = \gamma_{nm}\left[\frac{d}{d\theta}P_n^m(\cos\theta)\exp(im\phi)\boldsymbol{\theta} + \frac{im}{\sin\theta}P_n^m(\cos\theta)\exp(im\phi)\boldsymbol{\phi}\right].$$

They satisfy the orthonormality condition

$$\int d\Omega_{\mathbf{r}} \mathbf{A}^*_{n'm'}(\mathbf{r}) \bullet \mathbf{B}_{nm}(\mathbf{r}) = \delta_{nn'}\delta_{mm'}\delta_{AB}, \quad \text{(A 3)}$$

where $\mathbf{A}$ and $\mathbf{B}$ can be any one of $\mathbf{Y}$, $\mathbf{X}$ or $\mathbf{Z}$,

With respect to the cross product of $\mathbf{r}$, the VSHs have the properties

$$\mathbf{r}\times\mathbf{Y}_{nm}(\mathbf{r}) = 0,$$
$$\mathbf{r}\times\mathbf{X}_{nm}(\mathbf{r}) = -\mathbf{Z}_{nm}(\mathbf{r}), \quad \text{(A 4)}$$
$$\mathbf{r}\times\mathbf{Z}_{nm}(\mathbf{r}) = \mathbf{X}_{nm}(\mathbf{r}).$$

Using the fact that $\mathbf{Z}_{nm}$ and $\mathbf{X}_{nm}$ are both perpendicular to $\mathbf{r}$, and the relation

$$\mathbf{a}\bullet(\mathbf{b}\times\mathbf{c}) = \mathbf{b}\bullet(\mathbf{c}\times\mathbf{a}) = \mathbf{c}\bullet(\mathbf{a}\times\mathbf{b}) \quad \text{(A 5)}$$

we obtain useful vector product relations

$$\mathbf{X}_{nm}(\mathbf{r})\times\mathbf{Z}^*_{\nu\mu}(\mathbf{r}) = -\mathbf{X}_{nm}(\mathbf{r})\bullet\mathbf{X}^*_{\nu\mu}(\mathbf{r})\mathbf{r},$$
$$\mathbf{Z}_{nm}(\mathbf{r})\times\mathbf{X}^*_{\nu\mu}(\mathbf{r}) = \mathbf{Z}_{nm}(\mathbf{r})\bullet\mathbf{Z}^*_{\nu\mu}(\mathbf{r})\mathbf{r}, \quad \text{(A 6)}$$
$$\mathbf{X}_{nm}(\mathbf{r})\times\mathbf{X}^*_{\nu\mu}(\mathbf{r}) = \mathbf{X}_{nm}(\mathbf{r})\bullet\mathbf{Z}^*_{\nu\mu}(\mathbf{r})\mathbf{r},$$
$$\mathbf{Z}_{nm}(\mathbf{r})\times\mathbf{Z}^*_{\nu\mu}(\mathbf{r}) = -\mathbf{Z}_{nm}(\mathbf{r})\bullet\mathbf{X}^*_{\nu\mu}(\mathbf{r})\mathbf{r}.$$

The complex conjugation properties of the VSHs are analogous to that of the scalar spherical harmonics

$$\mathbf{A}_{n,-m}(\mathbf{r}) = (-1)^m\mathbf{A}^*_{nm}(\mathbf{r}). \quad \text{(A 7)}$$

The parity or inversion properties of the VSH are

$$\mathbf{Y}_{nm}(-\mathbf{r}) = (-1)^n\mathbf{Y}_{nm}(\mathbf{r}),$$
$$\mathbf{X}_{nm}(-\mathbf{r}) = (-1)^n\mathbf{X}_{nm}(\mathbf{r}), \quad \text{(A 8)}$$
$$\mathbf{Z}_{nm}(-\mathbf{r}) = (-1)^{n+1}\mathbf{Z}_{nm}(\mathbf{r}).$$

$$
\begin{aligned}
\mathsf{Y}^*_{nm}(-\mathbf{r}) &= (-1)^{n-m}\mathsf{Y}_{n-m}(\mathbf{r}),\\
\mathsf{X}^*_{nm}(-\mathbf{r}) &= (-1)^{n-m}\mathsf{X}_{n-m}(\mathbf{r}),\\
\mathsf{Z}^*_{nm}(-\mathbf{r}) &= (-1)^{n-m+1}\mathsf{Z}_{n-m}(\mathbf{r}).
\end{aligned} \tag{A 9}
$$

The spherical harmonics with phase factors included

$$
\begin{aligned}
\mathcal{X}_{nm}(\mathbf{r}) &\equiv -\mathrm{i}^{n}\mathsf{X}^*_{nm}(\mathbf{r}),\\
\mathcal{Z}_{nm}(\mathbf{r}) &\equiv -\mathrm{i}^{n+1}\mathsf{Z}^*_{nm}(\mathbf{r}),
\end{aligned} \tag{A 10}
$$

have slightly different, and more convenient properties. For the cross-product with $\mathbf{r}$ one finds, which is useful in far-field situations,

$$
\begin{aligned}
\mathbf{r}\times\mathcal{X}_{nm}(\mathbf{r}) &= -\mathrm{i}\mathcal{Z}_{nm}(\mathbf{r}),\\
\mathbf{r}\times\mathcal{Z}_{nm}(\mathbf{r}) &= -\mathrm{i}\mathcal{X}_{nm}(\mathbf{r}).
\end{aligned} \tag{A 11}
$$

Other properties of interest are their complex conjugate:

$$
\begin{aligned}
\mathcal{X}^*_{nm}(\mathbf{r}) &= (-1)^{n-m}\mathcal{X}_{n-m}(\mathbf{r}),\\
\mathcal{Z}^*_{nm}(\mathbf{r}) &= (-1)^{n-m-1}\mathcal{Z}_{n-m}(\mathbf{r}).
\end{aligned} \tag{A 12}
$$

A useful relation when treating reciprocity is

$$
\begin{aligned}
\mathcal{X}^*_{nm}(-\mathbf{r}) &= (-1)^{-m}\mathcal{X}_{n-m}(\mathbf{r}),\\
\mathcal{Z}^*_{nm}(-\mathbf{r}) &= (-1)^{-m}\mathcal{Z}_{n-m}(\mathbf{r}).
\end{aligned} \tag{A 13}
$$

References

[1] Mackowski, D. W., 1994, *J. opt. Soc. Am.* A, **11,** 2851.
[2] Mackowski, D. W., and Miscshenko, M. I., 1996, *J. opt. Soc. Am.*, **13,** 2266.
[3] Chew, W. C., 1994, *IEEE Press Series on Electromagnetic Waves* (New York: IEEE Press).
[4] Tsang, L., Kong, J. A., and Shin, R. T., 1985, *Theory of Microwave Remote Sensing, Wiley Series in Remote Sensing* (New York: John Wiley & Sons).
[5] CohenTannoudji, C., Dupont-Roc J., and Grynberg, G., 1987, *Photons et Atoms* (Paris: Editions of the CNRS 15, quai Anatole-France), pp. 55–57.
[6] Bohren, C. F., and Huffman, D. R., 1983, *Absorption and Scattering of Light by Small Particles* (New York: Wiley-Interscience).
[7] Stein, S., 1961, *Quart. appl. Math.*, **19,** 15.
[8] Cruzan, O. R., 1962, *Quart. appl. Math.*, **19,** 15.
[9] Mie, G., 1908, *Ann. D. Phys.*, **25,** 377.
[10] Barber, P. W., and Hill, S. C., 1990, *Light Scattering by Particles: Computational Methods, Advanced Series in Applied Physics* (Singapore: World Scientific).
[11] Wang, R. T., and Gustafson, B. A. S., 1984, *Proceedings of the 1983 Scientific Conference on Obscuration and Aerosol Research*, edited by J. Farmer and R. Kohl (Aberdeen, MD: US Army Chemical Research, Development, Engineering Center), pp. 237–247.
[12] Mackowski, D. W., 1991, *Proc. R. Soc.* (London) Ser. A, **433,** 599.
[13] Tzeng, Y. C., and Fung, A. K., 1994, *J. electron. Waves Appl.*, **8,** 61.
[14] Tayeb, G., and Maystre, D., 1997, *J. opt. Soc. Am.* A, **14,** 3323.
[15] Gralak, B., Enoch, S., and Tayeb, G., 2000, *J. opt. Soc. Am.* A, **17,** 1012.
[16] Lo, K. M., McPhedran, R. C., Bassett, I. M., and Milton, G. W., 1994, *J. lightwave Technol.*, **12,** 396.